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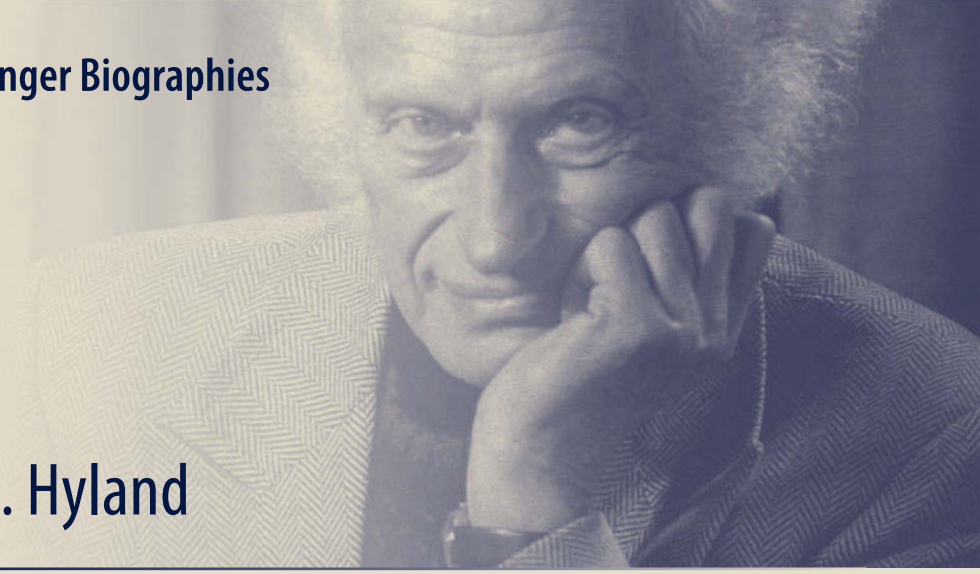

G.J. Hyland

Herbert Fröhlich

A Physicist Ahead of His Time

Springer

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Herbert Fröhlich

A Physicist Ahead of His Time

With a foreword by Fanchon Fröhlich

Springer

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This work is dedicated with esteem
and gratitude to
Fanchon Fröhlich,
philosopher and artist—friend for 50 years

Foreword

It gives me very great pleasure to write a few words of introduction to this biography of my late husband, Herbert Fröhlich.

When my good friend of many years, Gerard Hyland, a former student of Herbert (in fact his last Ph.D. student) shared with me the idea for this book, I was very happy to give him every assistance in piecing together all the available documentation and photographs relating to Herbert's career. It has been a longer journey than expected—books usually are!—but the result is a work that goes far beyond a simple narrative of one man's life. This book touches upon a whole era of physics research in Europe, setting it in the context of the dramatic political and military circumstances of the time. Herbert's experiences, some of which I shared, clearly reveal how science and scientific curiosity are not to be extinguished by mere national enmities; how scientists—then as now—form a mutually self-supporting global community that is often able to overcome the whims of dictators and other political exigencies in their pursuit of scientific progress.

This biography also details the many interactions Herbert and I had with other active scientists and their families. (My strongest memories are of Richard Feynman, S.W. Hayter, Maurice Marois, and Erwin Schrödinger). I am filled with nostalgia as I look at the collected photos, read the anecdotes, and remember not only the many good times described herein, but also the sometimes more difficult ones. For the outsider they will be informative rather than emotive, but still a wonderful record of a decisive period of twentieth-century science and some of the people involved.

I am not a scientist, and my understanding of Herbert's work is not of a technical kind, but rather conceptual, having had the benefit of many discussions with him after our marriage in 1950, particularly when his ideas about a particular problem were still being crystallized—discussions that often included a philosophical dimension. The reception of his work and the respect accorded him by those better qualified than myself to judge its technical merit are ample testimony to its importance. The reader with a basic education in science will learn more in these pages about his many and varied contributions. These reveal that Herbert was—rather like Feynman, some of whose correspondence with my husband is reproduced in this book—a man of broad interests. He frequently began to address one topic, which, having achieved some new insights and recorded these for publication, he would then drop in order to move on to another more challenging problem. The subtitle of this biography "A Physicist Ahead of His Time" resonates with my impression that he frequently left the jigsaw unfinished, but in a form that allowed others to come along and complete it at a later date.

I thank Gerard for undertaking this biography, and for the marvellous book that has resulted, in which I trust the reader will find enjoyment and inspiration.

December 2014 Fanchon Fröhlich

Acknowledgments

It is a pleasure to fulsomely acknowledge the unstinting help I have received over very many years from Fröhlich's widow, Fanchon, in the course of my documentation of her late husband's life and work, for providing me with valuable information and insights into his life and work, and for making available the photographs included in this biography. I am also indebted to former members of Fröhlich's department in Liverpool for sharing their reminiscences with me—in particular, the late Dr. Brian Holland, the late Dr. Ronald Huby, Dr. Harry Newns, Dr. Charles Terreaux—and to other former colleagues elsewhere, especially Dr. Jiri Pokorný (Prague), Prof. Dr. Fritz Popp (Neuss), and Prof. Geoffrey Sewell (London).

The genealogical assistance of Barbara Staudacher (Rexingen), and the archival assistance of Adrian Allen (former Librarian of Liverpool University), Alan Franklin (Manx National Heritage), Joanna McManus (Royal Society Librarian), Dr. Jagdish Mehra (Professor of Science and Humanities at the University of Houston), Angelika Mundorff (Museum Fürstenfeldbruck), Dr. Gerhard Neumeier (Stadtarchiv, Fürstenfeldbruck), and Josephine Zilberkweit (Liverpool) is gratefully acknowledged. In addition, I wish to thank Helen Ireland, Peter Larkin, and Geoffrey Lewis (Warwick University Library), Prof. John Dainton, F.R.S. and Dr. Peter Rowlands (University of Liverpool), Dr. Brian Pollard (University of Bristol), and especially Dr. Laurence Caves (Southam), Dr. Mario Micallef (University of Warwick) and Anthony Teeluck (Hook) for their assistance in various ways.

I also wish to acknowledge the assistance of the Department of Special Collections of the Bodleian Library, Oxford, in making available material from the archives of the Society for the Protection of Science and Learning, and to thank its successor organization, the Council for Assisting Refugee Academics (*CARA*), for permission to reproduce some of the material in this book.

Liberal use has been made of unpublished interviews with Fröhlich, recorded by Profs. Haken and Wagner in Stuttgart, during the 1980s, which Fröhlich made

available to me to supplement those recorded by myself in Liverpool around the same time.

The collaboration during the 1970s with Prof. George Rowlands, a fellow member of the Department of Physics at the University of Warwick, and with Prof. Fred Cummings of the University of California at Riverside in extending some aspects of Fröhlich's work on superfluid He^4 is remembered with pleasure.

It is a particular pleasure to thank my old friend and former colleague Claudio Ronchi for encouraging me to approach *Springer Verlag*, and to gratefully acknowledge the invaluable editorial assistance of Dr. Angela Lahee (Springer Verlag, Heidelberg) and of Charles Rayappan and Gajalakshmi Sundaram (Scientific Publishing Services, Chennai, India).

Finally, I wish to sincerely thank my wife, Mercedes, for her help with proofreading, the compilation of the index, and general word-processing matters.

Contents

Herbert Fröhlich—a drawing by his wife Fanchon, 1973

Chronology

Cited references in square brackets correspond with those in the *Complete Bibliography of H. Fröhlich* at the end of this book. Particularly important works are identified by parenthetical numbers in **bold** type; asterisked references denote his most influential works.

1905: Herbert Fröhlich born in Rexingen (Württemberg) on Saturday, 9 December.

1907: The Fröhlich family moves to Munich.

1911–1915: Herbert attends Primary School in Munich.

1915–1921: Attends the Municipal High School of Commerce in Munich, after which he moves to Reutlingen to spend 6 months at the Technical College for the Textile Industry.

1927: ***age 21***: Takes his *Abitur* at the *Oberrealschule* in Magdeburg; enters the Ludwig-Maximilian University, Munich, to study experimental physics.

1928: Enrols for a doctorate in experimental physics under W. Wien; Wien dies, Fröhlich changes to theoretical physics, and starts attending lectures by Sommerfeld.

1929: Enters the Theoretical Physics Institute under Sommerfeld.

1930: ***age 25***: Awarded D. Phil. (*Magna cum Laude*) for a thesis on the Photoelectric Effect in metals, which is published in *Annalen der Physik* as [F1].

1930–1931: Research Fellow in Munich, where he is the first to apply ‘second quantization’ to electrons in solids [F**2**].

1931: Moves to Freiburg im Breisgau (Württemberg) as Research Fellow, to introduce modern physics in Gustav Mie’s Physical Institute.

1932: *Privatdozent* in Freiburg from December.

1933: *Habilitationsschrift* published as [F7]. *Privatdozent* appointment confirmed by the State of Württemberg early in the year, but is terminated by the Nazis in April.

1933: Visits London in an unsuccessful attempt to obtain, *via* the Academic Assistance Council (*AAC*), financial support for a research position in the *UK*—despite an offer from Schrödinger to be his Assistant at Oxford. Joins his parents in Strasbourg, to where they have fled from from Munich to escape the Nazis.

1934: Accepts an invitation from Frenkel to go to Leningrad (St. Petersburg) to work as a 'Foreign Expert' in A.F. Joffe's Physico-Technical Institute.

1935: ***age 30***: Flees Russia to escape Stalin's purges, and eventually arrives (*via* Vienna) in the *UK* on 30 June to attend a conference in Bristol on the Metallic State; he is awarded a Research Fellowship for 12 months by the *AAC* in July to work with Mott in Bristol.

1936: First book—*Elektronentheorie der Metalle* [F(**i**)]**—is published by Springer. Obtains a grant from Bristol University.

1937: Publishes his first paper on dielectric breakdown [F**16**]*. Moves to Holland for 6 months with a stipend from the Lorentz Foundation to work in Kramers' group in Leiden, from where he publishes [F**17**] dealing with the specific heat of small particles. Returns to Bristol with the promise of financial support from the Electrical Research Association (*ERA*) to continue work on dielectric breakdown.

1938: Collaborates with Heitler and Kemmer [F**20**] in a vector extension of Yukawa's meson theory, which predicts a neutral meson.

1940: ***age 35***: Interned for about 3 months, together with other 'aliens' including Heitler and Heinz London. Publishes [F**33**] (with Nabarro), predicting that monovalent metals should, through an indirect interaction between nuclear spins mediated by conduction electrons, exhibit nuclear ferromagnetism at sufficiently low temperatures—anticipating by 14 years the better-known work of Ruderman and Kittel.

1943: Joins the academic staff of Bristol University as a lecturer in Theoretical Physics.

1946: Attends the International Conference on Fundamental Particles and Low Temperature Physics in Cambridge.

1947: Publishes [F**60**]*, out of which later evolved 'hot' electron physics. Visiting Professor at the Dublin Institute for Advanced Studies, where Schrödinger and Heitler are then based.

1948: Leaves Bristol to take up the first Chair of Theoretical Physics at the University of Liverpool on 1st October.

1949: Oxford University Press (*OUP*) publishes his second book—*Theory of Dielectrics* [F(**ii**)]**.

1950: ***age 45***: Publication of his work (in collaboration with Pelzer and Zienau) on the properties of slow electrons in polar materials (large polarons) [F**72**]*. Visiting Professor at Purdue, where he lectures on *Statistical Mechanics*, and tours principal universities in *USA* and Canada. Identifies the electron-phonon interaction as the basis of superconductivity [F**76**]**, consistent with which is the contemporaneous discovery of the isotope effect [F**77**]. Marries Fanchon Aungst (*b.*1927).

1951: Elected Fellow of the Royal Society on 15th March. Invited to speak [F81] in Washington on the occasion of the 50th anniversary of the *US* National Bureau of Standards (*NBS*).

1952: Publication of his field-theoretical Hamiltonian [F**84**]*** for electrons and phonons in interaction, marking the start of a new era in solid-state physics. Lectures in Lille (under the auspices of the Foreign Universities Interchange Scheme), Zurich, Rennes, and in Paris, at the conference 'Rotational Transformations in Solids' [F82].

1953: Visiting Professor at Purdue during the Lent term, where he lectures on *Electrons in Crystals*; lectures in other *US* Institutions, at the Canadian Research Council, and at the Lorentz-Kamerlingh Onnes Centenary Meeting in Leiden [F85]. Lectures at international theoretical physics conferences in Kyoto and Tokyo [F87, 88] and at a conference on low temperature physics in Houston, *USA*.

1954: Solves his Hamiltonian exactly for a 1-dimensional model of a superconductor, finding, for the first time, an energy spectrum characterised by an essential singularity in the electron-lattice coupling constant and a gap [F**89**]; presents these results at the 10th Solvay Conference in Bruxelles [F93]. Lectures in Copenhagen, Rennes, *ETH* Zurich, and at the International Conference on Semiconductors in Amsterdam [F91].

1955: ***age 50***: Awarded his first honorary degree (D.Sc.) by the University of Rennes, France, in recognition of his contributions to dielectric theory.

1956: Lectures at Purdue on the *Polaron* and on the *Many-body Problem*, and in Bruxelles at l'Institut des Hautes Études. Attends the International Conference on Electron Transport in Metals and Solids held at the Canadian National Research Laboratories, Ottawa, in September.

1957: Second Edition of *Theory of Dielectrics* published by *OUP*. Lectures at a number of American universities, at the University of Paris, and at the Dublin Institute for Advanced Studies.

1958: Visits the University of Strasbourg under the auspices of the Foreign Universities Interchange Scheme.

1960: ***age 55***: Publishes a novel approach to space-time reflections, predicting 4 new mesons with the same isospin as the K-mesons [F**107**]. Visits Australia as a consultant to the Executive of Australian Commonwealth Scientific and Research Organisation (*CSIRO*).

1961: Gives further lectures in *USA* and Canada.

1962: Visits Hungary (under the auspices of the British Council) at the invitation of the Hungarian Government. Lectures on *Polaron Theory* [F114] at the Scottish Universities Summer School on *Polarons and Excitons* at St. Andrews.

1963: Develops further his isobaric spin algebra in [F116]; publishes a new formulation of the electron-phonon interaction in narrow band materials with Machlup and Mitra [F**118**]. Gauss Visiting Professor in Göttingen (May–July).

1964/1965: Visits universities and research laboratories in the *USA* during the Lent term, and is Visiting Professor at the Technical University of Stuttgart during July, where he lectures on *Solid-state Physics*.

1965: ***age 60***

1966: Elected Chairman of the International Commission of Statistical Mechanics and Thermodynamics of the International Union of Pure and Applied Physics (*IUPAP*) until 1972. Visits Leiden as Lorentz Professor, and a number of American universities during the autumn, and speaks [F127] at a symposium on Ferroelectricity in Michigan in honour of P.W. Debye.

1967: Publishes his first paper [F**129**] on the connection between micro and macrophysics. Gives the opening address *Quantum Mechanical Concepts in Biology* at the first meeting in the series ‘From Theoretical Physics to Biology’ organised by l’Institut de la Vie, in Versailles.

1968: Speaks on *The Connection of Micro and Macrophysics* [F**139**]* at the International Conference on Statistical Mechanics in Kyoto, where he is Visiting Professor at the Research Institute for Fundamental Physics. Publishes a theory of superconductivity in materials with incomplete inner shells [F**131**], and presents a model showing how coherent excitations can be established in living systems [F**134**]**. Visits Italy in April (under the auspices of the Italian Research Council and the British Council), and, during the autumn term, the *USA* and the University of Alberta, Canada, where he lectures on *The Connection between Microscopic and Macroscopic Physics*, and where he is awarded LL.D (*Hon. causa*).

1969: Proceedings [F**138**]* of the 1967 Versailles conference (*From Theoretical Physics to Biology*) published; gives the Opening Address at the 2nd Versailles meeting on the same topic. Awarded Sc.D (*Hon. causa*) by Trinity College, Dublin, and is Visiting Professor at Stuttgart, where he lectures on *The Connection between Micro and Macrophysics*.

1970: ***age 65***: Lectures at the Dublin Institute for Advanced Studies during February, and at the University of Stuttgart during July. Broadcasts on German Radio a series of talks [F148] entitled *Theoretische Physik und Biologie* (Theoretical Physics and Biology). Undertakes further visits to the *USA* and Canada during the autumn term.

1971: Visits several European universities during April, July and September, and lectures in Stuttgart on the *Boltzmann Equation*. Attends the 3rd meeting of the series 'From Theoretical Physics to Biology' organised by l'Institut de la Vie; proceedings of the second meeting published [F147].

1972: Awarded the *Max Planck Medal* of the German Physical Society (presented *in absentia*, owing to convalescence from surgery, and accepted on his behalf by his wife, in October in Wiesbaden). Predicts a frequency-specific long-range interaction between coherently oscillating systems [F**150**]*. Speaks at a symposium on Synergetics at Schloss Elmau, Bavaria [F153]. Visits Canada and *USA* (including La Jolla). Undertakes a Royal Society Study Visit to Japan during December and January 1973, visiting Tokyo and Kyoto (Institute for Fundamental Physics); returns home *via* Bombay (Tata Institute).

1973: Retires from the Liverpool Chair, which is marked by the publication of the *Festschrift* 'Cooperative Phenomena'. Becomes *Professor Emeritus* in Liverpool, and is appointed (until 1976) Professor of Solid-state Electronics at the University of Salford, Manchester *UK*. Publishes [F**154**] dealing with non-linearly coupled electric polarization and elastic fields, and a major review article [F**155**]* on the connection of micro and macrophysics.

1974: Lectures at the Erice Summer School on Cooperative Effects [F158].

1975: ***age 70***: Speaks at a colloquium in Berlin to mark the 75th Anniversary of Planck's Quantum Theory [F165]. Gives the opening address at the 5th International Conference on Theoretical Physics and Biology, in Vienna.

1976: Gives a Science Faculty Lecture (*Order and Organisation in Physics and Biology*) in Liverpool to celebrate his 70th birthday, and lectures at a symposium on Interdisciplinary Aspects of Modern Physics, in Parma, Italy.

1977: Speaks at a Workshop on Synergetics at Schloss Elmau, Bavaria [F172].

1978: Speaks at the Pennsylvania Conference on Electrical Insulation and Dielectric Phenomena, organised by the National Academy of Science Conference [F175].

1979: Elected a Foreign Member of the Stuttgart Max Planck Institute for Solid-state Research. Speaks on *Non-local electrodynamics and the Kemmer Equation* at a symposium to mark Kemmer's retirement from the Edinburgh Chair, and at the *IBM* International Workshop on Physical Concepts in Tissue Growth, in Bad Neuenahr (Rhineland-Palatinate), Germany [F178].

1980: ***age 75***: Awarded Honorary Doctorates by the University of Stuttgart (Dr. Rer. Nat.), and by Purdue University (D.Sc.). Lectures in Munich, Alberta, Houston (at a symposium on the Biological Effects of Non-ionising Radiation [F179]) and at the Dublin Institute for Advanced Studies.

1981: Lectures in Cologne, at the Dublin Institute for Advanced Studies, and at the international symposium Biomedical Thermology in Strasbourg [F181]. Visits New Delhi, India (under the auspices of the British Council) and there attends the 1st International Seminar on the Living State.

1982: Lectures in Erice [F185], Palermo, San Miniato (*NATO* Advanced Study Institute on Molecular Models of Photoresponsiveness [F187]), Stuttgart, and at an International Symposium in Bad Neuenahr.

1983: Contributes [F186] to the proceedings of the 1982 Bad Neuenahr Symposium, which he co-edits with F. Kremer, and which is published by Springer under the title *Coherent Excitations in Biological systems* [F(iii)]. Speaks at the International Conference on Non-linear Electrodynamics in Biological Systems, in Loma Linda, California. Visits Moscow.

1984: Lectures in Messina, Sicily.

1985: ***age 80***: International Symposium held at Liverpool to celebrate his 80th birthday. Speaks at the 50th Anniversary of Meson Theory in Kyoto, Japan [F193], and attends the Sixth International Meeting on Ferroelectricity in Kobe.

1986: Chairs the European Physical Society Meeting in Überlingen (Baden-Württemberg), Germany.

1987: Publication of the *Festschrift* 'Energy Transfer Dynamics' in honour of his 80th birthday in December 1985. Presents *Elementary Remarks on High Temperature Superconductors* at the International Workshop on Novel Mechanisms of Superconductivity in Berkeley [F197]. Speaks in Prague at the International Seminar on the Biophysical Aspects of Cancer. Appears on *UK* Granada Television in a Nuclear Science Documentary. Attends an international conference at the University of Bristol to celebrate the 40th anniversary of the discovery of the π- and V-particles.

1988:	*Biological Coherence and Response to External Stimuli* [F(**iv**)] is published by Springer, which he edits and contributes the first chapter. His 1968 paper *Long range coherence and energy storage in biological systems* [F**134**]** is listed as a 'most frequently cited work' by the Institute for Scientific Information (*ISI*). Attends a conference (Meaning and Structure in Biology and Physics: Some Outstanding Questions) in Bermuda, and is Chairman of the last in the series of conferences on Theoretical Physics and Biology organised by l'Institut de la Vie.
1989:	Awarded D.Sc (*Hon. causa*) by North Eastern Hill University, India. Principal speaker at a symposium on Biophysics in Kiev (May), and at a symposium on Biometrology in Stockholm (June).
1990:	***age 85***: Attends the First International Conference on the Study of Consciousness within Science, San Francisco, in February, but is too unwell to speak.
1991:	Dies in the early hours of Wednesday, 23 January, aged 85, in the Royal Liverpool University Hospital, from an infection following an operation there for bowel cancer.

Fig. 1.1 Herbert Fröhlich, 1905–1991

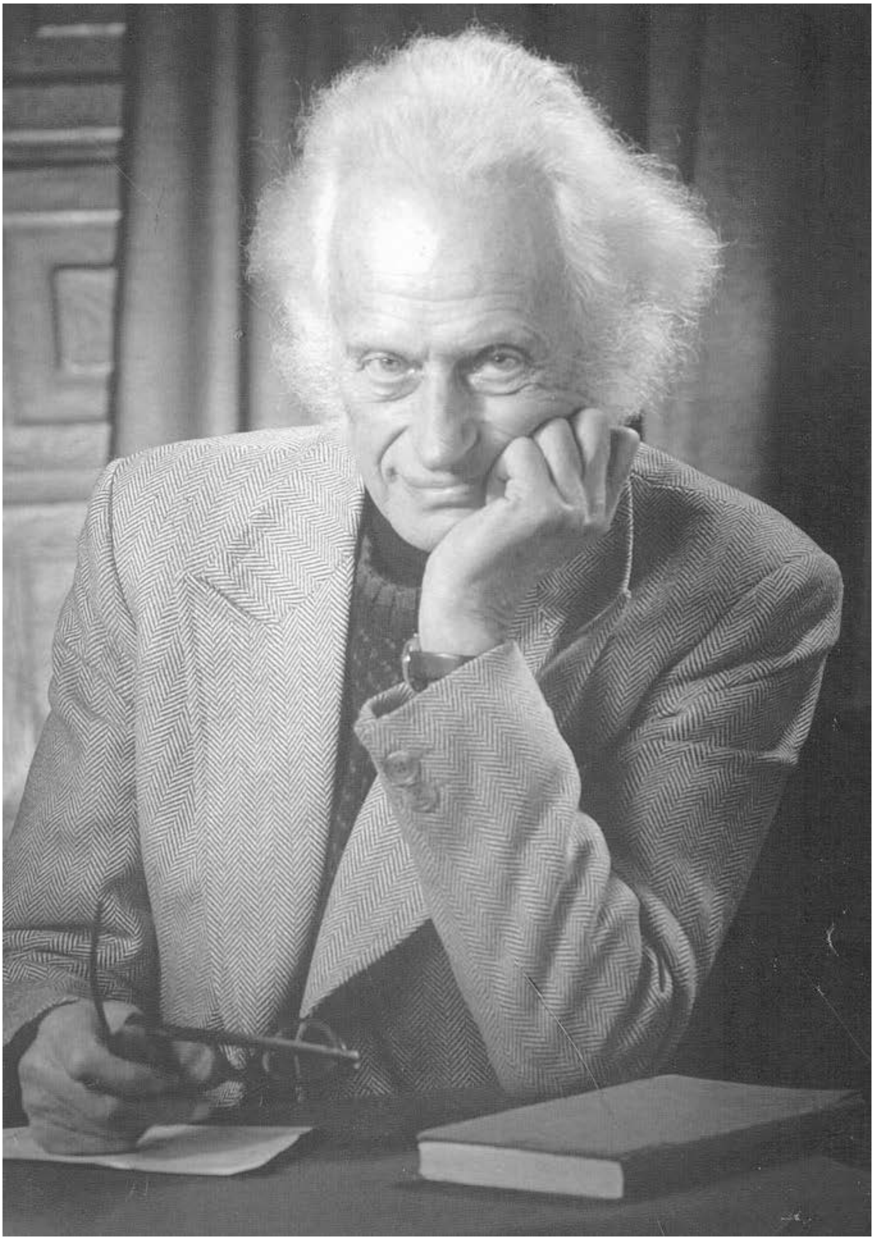

Fig. 1.1 Herbert Fröhlich, 1905–1991

Chapter 1
Prologue

Einstein's *annus mirabilis* of 1905 saw the publication of his investigations into the theory of Brownian motion, his theory of special relativity, and, most revolutionary of all, his paper on light quanta, in which he extended Planck's quantization of material oscillators to electromagnetic radiation, thereby affording an immediate understanding of—amongst other things—the *photoelectric effect*, for which, 16 years later, he was awarded the Nobel Prize. Some 25 years later in 1930, at the University of Munich, the photoelectric effect was the subject of the doctoral dissertation of the theoretical physicist **HERBERT FRÖHLICH**, 1905–91 (Fig. 1.1).

Fröhlich's illustrious career, which started just after the new quantum mechanics had been formulated, and which went on to span some 60 years during which many developments in physics took place, was distinguished by the diversity of the fields in which he was active and to which he contributed so significantly. His contributions often influenced subsequent developments, sometimes in a fundamental way, and often revealed some hitherto unsuspected connection between seemingly quite unrelated areas of physics, and later, even between physics and biology. The hallmark of his particular genius was an ability to recognize when new ideas genuinely had to be introduced and when they did not. It was not only from this ability to balance the radical with the conservative that his most significant contributions arose, but also from his consummate skill in identifying the particular aspect of a given phenomenon on which to focus attention, thereby often unearthing some hitherto unsuspected possibility within a pre-existing theoretical framework, thus removing the need for ad hoc hypotheses. The decisive influence he exerted—often as a 'man ahead of his time'—in fields as diverse as meson theory and biology stands as a strong indictment against fragmentation and over-specialization in theoretical physics, something that was quite alien to his holistic perception of the subject. Indeed, so much ahead of his time was he that he pioneered a number of topics long before some of them were rediscovered later by others who now take much of the credit; these include: the prediction of (i) a neutral meson and a quartet of vector mesons, (ii) a partial anticipation of the Lyddane-Sachs-Teller relation, (iii) an interaction between the nuclear spins in a metal mediated by conduction

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electrons, (iv) 'hot' electron physics, (v) ferroelectric soft modes, (vi) polaritons, (vii) a phonon-mediated electron-electron attraction as the basis of superconductivity, (viii) 'sliding' modes in low-dimensional conducting systems, (ix) the importance of size effects in finite samples, both in solid-state physics and biology.

> **Most influential of all, however, was undoubtedly his introduction, in the early 1950s, of the methods of quantum field theory into solid-state physics, which completely revolutionised the future development of the subject, and of statistical mechanics in general.**

43 years of Fröhlich's working life were spent at the University of Liverpool in the *UK*, where he occupied, with great distinction, the first Chair of Theoretical Physics from 1948 until his retirement in 1973. This occasion was marked by a *Festschrift*, 'Cooperative Phenomena', edited by H. Haken and M. Wagner, and published by Springer Verlag. A post-retirement position at the Dublin Institute for Advanced Studies had been seriously considered, but did not materialise, and he was appointed Professor Emeritus in Liverpool, a position he held until his death on 23 January 1991 at the age of 85. Between 1973 and 1976, he was Professor of Solid-State Electronics at the University of Salford, *UK*, and his 80th birthday saw the publication by Springer of a second *Festschrift*, 'Electron Transfer Dynamics', edited by T.W. Barrett and H.A. Pohl.

Before moving to Liverpool, Fröhlich was Reader in Physics at the University of Bristol to where he had come from Russia in 1935 as a refugee from Stalin's purges, having left his native Germany after being dismissed by the Nazis from his university post in Freiburg in 1933, shortly after his appointment as *Privatdozent* had been ratified. Prior to this, he had been a doctoral student of Sommerfeld in Munich, receiving his D.Phil. in 1930, after only two years of study. Internationally, he was a highly respected figure whose status as a colossus in the world of Theoretical Physics was undisputed. His circle of friends and colleagues included such *illuminati* as Sommerfeld (his doctoral supervisor), Heisenberg, Schrödinger, Bohr, Feynman and Pauli, who, of Fröhlich, once declared, 'Ah yes, there we have someone who can not only calculate, but can also think!' [J. Mehra—personal communication, *c.*1990].

Fröhlich became a Fellow of the Physical Society of Great Britain in 1944, was elected[1] Fellow of the Royal Society in 1951, was awarded the Max Planck Medal of the German Physical Society in 1972,[2] and received numerous Honorary Degrees worldwide (see *Chronology*).

On several occasions between 1954 and 1972, he was invited to submit nominations to the Swedish Royal Academy of Science's Nobel Committee for Physics, and between 1973 and (at least) 1988 he annually proposed a candidate for the Max Planck Medal.

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[1] He had first been proposed in 1946.

[2] Since, at the time, Fröhlich was convalescing after surgery, his wife accepted the Medal on his behalf at a General Conference of the European Physical Society, in Wiesbaden, 3–6 October, 1972, and read the paper [F151] he had written whilst in hospital.

During the late 1960s, at the invitation of Hermann Haken (who had been a young Visiting Research Fellow in Liverpool during the late 1950s), he started to make regular visits to Stuttgart where he gave many stimulating lectures at the Theoretical Physics Institute of which Haken was, by then, Director. The connection with Stuttgart intensified after 1970 with the opening of the Max Planck Institute for Solid-state Physics to where he was invited by its first Director, Ludwig Genzel, and where, from 1979 to 1991, Fröhlich was a Foreign Member, regularly making extended visits. Peter Fulde—a former director of one of the Institute's research groups, who remembers these many visits—is quite sure that Fröhlich's relationship with Germany was not at all embittered by his pre-war experiences, some of which he dismissed as reminiscent of a fairly amusing adventure film.

Fröhlich travelled extensively, visiting many parts of the world, lecturing,[3] and exchanging ideas with other physicists, often as Visiting Professor, such as Gauss Professor in Göttingen, Lorentz Professor in Leiden during the 1960s, and at the Dublin Institute for Advanced Studies on many occasions from 1947 onwards. Between 1966 and 1973, he was Chairman of the Commission on Statistical Mechanics and Thermodynamics of the International Union of Pure and Applied Physics.

In all, he published 200 papers—62 on condensed matter physics (excluding superconductivity, but including papers dealing with the connection between micro and macrophysics), 44 on dielectrics, 25 on superconductivity, 48 on theoretical physics and biology, and 21 on particle physics. In addition, he authored 2 books (one on the electron theory of metals, the other on the theory of dielectrics), and edited two others (both on theoretical physics and biology), the second at the age of 82. From 1948, Fröhlich was an editor of the series *Monographs on the Physics and Chemistry of Materials*, published by Oxford University's Clarendon Press. In addition, he was Editor-in-Chief of the journal *Collective Phenomena* (published by Gordon and Breach), and was on the Editorial Boards of the journals *Physics of Condensed Matter*, *Il Nuovo Cimento*, and the *International Journal of Engineering Science*.

Fröhlich's holistic outlook and constant alertness to the possibility that certain concepts might well have relevance to fields other than those in which they had first arisen, helped resolve some of the most enigmatic mysteries of the physics of his era. His most heroic attribute, however, was undoubtedly the courage to entertain an unusually wide range of novel ideas, and to have the conviction to express them without fear of possible refutation. As his old friend and colleague of pre-war days, Walter Heitler,[4] wrote in his contribution to Fröhlich's 1973 *Festschrift*… 'Herbert's strength consisted in his wealth of *anschaulich* ideas, by which he

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[3] A list of unpublished transcripts of some of his invited lectures at various institutions is given in the *Complete Bibliography of H. Fröhlich* at the end of this book, wherein reference is made also to those lectures that have been published.

[4] Heitler (1973).

grasped physics without much reference to the underlying mathematics. Formalism was not his strength…. Such a gift is today unfortunately rare. Of formalists there are enough.'

This biography grew out of a contribution originally written for a belated centennial celebration of Fröhlich's birth, which was held in Liverpool in 2006, the proceedings of which were published by the University, with myself as co-editor with P. Rowlands (Hyland and Rowlands 2006, 2008). Being his last doctoral student, 1965–68, and having maintained regular contact with him after I moved to the University of Warwick, where I taught theoretical physics between 1968 and 2001, I had numerous opportunities to personally discuss with him not only his many and varied contributions in the past, but also problems on which he was working during the 1970s and 1980s, some of which I collaborated in. These discussions, many of which were recorded, shed much light on the contemporary background to many of his most significant contributions, from which it has been possible to gain a unique perspective and insight into the true significance of his work, which might otherwise not have been possible. During his later years, he used to tell me he often consulted the articles I had written for his two *Festschrifts* whenever he wanted to refresh his memory. Since his death, I have had the benefit of many fascinating meetings with his widow, Fanchon, whose lucid reminiscences have been an invaluable source of inspiration.

I have treated some topics of particular importance in the Fröhlich Canon—such as dielectric breakdown, polaron theory, superconductivity, the connection between micro- and macrophysics, and the prediction of macroscopic quantum states in living systems—in more quantitative detail in order to illustrate his methodology and to give a flavour of the associated mathematics involved.

In addition to the photographs in the body of the text, there is, at the end of each of the following chapters, a Photo Gallery of images of some of the people, places and events referred to in those sections of the chapter identified in the captions.

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Chapter 2
Early Life in Germany

2.1 Childhood and Schooling

Herbert Fröhlich was born at 11.30 am on Saturday 9 December 1905 in the home of his paternal grand-parents, Abraham Jakob and Dorken (*née* Landauer) at Im Brühl, 186 (now Freudenstädter Straβe 31), in Rexingen (Württemberg), a village in the Neckar valley, on the edge of the Black Forest, where an unusually high proportion of the inhabitants were Jewish. Rexingen is near Horb am Neckar, approximately midway between Freudenstadt and Tübingen. Herbert's parents (both b.1878) Jakob (known as Julius) and Frieda, *née* Schwarz (known as Fanny) came from long-established Jewish families[1] in Rexingen, where they were married in 1903. Herbert's birth was registered on 10 December by his father, a dealer in livestock. The family was comfortably off, but had no tradition of university education. Herbert was the first male child in the family—one year younger than his sister Betty,[2] and 10 years older than his brother, Albrecht[3] (known as Ali).

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[1] There are 36 Fröhlich entries in the archives of the old Jewish Cemetery in Rexingen, and 62 relating to his mother's family (Schwarz); the Fröhlich line can be traced back to at least 1744 (In Stein gehauen 1997, 2003, pp. 122–123).

[2] At school Betty (*b*.1904) excelled in mathematics, but having, in those days, no possibility of pursuing it professionally, became, instead, an accomplished pianist. In 1926, as an ardent Zionist, she was the first member of the family to leave Germany for Palestine, at first living on a collective farm with her husband Fritz Lichtenstein. They separated in 1938, and she later married Martin Lustig. She had a daughter and 4 sons, one of whom was killed in the Yom Kippur war of 1973.

[3] Albrecht (1916–2001) later became a distinguished pure mathematician, and was elected Fellow of the Royal Society of Great Britain in 1976. He married in the same year as his brother (1950), and died at the same age of 85 years, leaving a widow (Dr. Ruth Brooks), 2 adopted children and several grandchildren. Herbert and Albrecht were not the only siblings to be elected F.R.S. in the 20th century; others include Christopher and Michael Longuet-Higgins, Maurice and David Hill, Nathaniel and Miriam Rothschild, and Sidnie and Irene Manton.

G.J. Hyland, *Herbert Fröhlich*,
Springer Biographies, DOI 10.1007/978-3-319-14851-9_2

Fig. 2.1 Fröhlich as a youth on the ski slopes, *c.*1923

In October 1907, when Herbert was not yet 2 years old, the family moved to Munich,[4] and decided to become naturalised Bavarians in order to be able to enjoy rights that otherwise would have been denied them. After moving, his father continued his livestock business in partnership with Josef Bikart from premises[5] in Fürstenfeldbruck, a small town some 25 km west of Munich, travelling there each day by train. It was in Munich—a city for which he retained a life-long nostalgia—that Herbert grew up, developing a love of chess and classical music, and attending Primary School (*Volkschule*) from 1911 until 1915, when he entered the Municipal High School of Commerce (*Städtische Höhere Handelsschule*). He was always a very independent child, and in 1921, when his excellent mathematics teacher was replaced by a mediocre one, he left school at the age of 15 without his *Matura*, essentially as an act of defiance, to avoid fulfilling the expectation of his elders that 'such a clever boy' must go to university! He then apprenticed himself to a firm of textile manufacturers (quite possibly belonging to an uncle on his mother's side), spending 6 months at the Technical College for the Textile Industry in Reutlingen, intending to make a career in commerce, which he did until 1926.

During this time, however, he became interested in radio, which in those days one had to build oneself. In a magazine for radio enthusiasts, he read that anyone who seriously intended to build radios needed higher mathematics. Accordingly, he bought what turned out to be a very good book from which he taught himself calculus, and started to do physics as a hobby; but he soon 'caught the bug', and decided he wanted to study it properly. In order to gain admission to university, however, he first had to take his *Abitur*. In those days, each German state had its own regulations, and, after studying these, he decided to go to Magdeburg where external candidates were examined mainly in relation to their proposed course of study later on. Since there were few posts in theoretical physics—other than school teaching, which did not appeal to him—he opted for experimental physics, intending to keep theoretical physics as a hobby. After 6 months as a private student at the *Oberrealschule* in Magdeburg, he took his *Abitur* early in 1927 at the age of 21.

During the 1920s, he was a member of the *Jungjüdische Wandervögel* (a hiking club for Jewish youths) and travelled extensively throughout Germany collecting folk songs; he was, however, never interested in any political activity. He excelled in sporting activities, particularly skiing and mountaineering, a love of which he retained throughout his life (Fig. 2.1).

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[4] From September 1917 until 1933, when they left Germany, they lived at Seidlstraβe, 22.

[5] At first, they had premises at Hauptstraβe 1, moving a few years later to Schöngeisingerstraβe 6, and finally in 1930, to Bullachstraβe 3, near the centre of the town.

2.2 Entrance to University and Early Academic Career in Germany

Fröhlich entered the Ludwig-Maximilian University in Munich as an undergraduate in the summer of 1927, beginning 4 semesters of lectures in physics, mathematics and chemistry. On his first day, he decided that the most important thing to do was to find a subject for a thesis, and accordingly went to a lecture that he had seen advertised entitled *Current Problems in Modern Physics*, seating himself in the front row. This turned out to be rather embarrassing, since, unbeknown to him, the front row was always reserved for full professors: he should have sat at the back—indeed, as a new student, he probably should not have been there at all!

During his second year, he decided he wanted to enrol for a doctorate in experimental physics under the Nobel Laureate Wilhelm Wien, but with Wien's death later that year (1928), Fröhlich changed to his erstwhile 'hobby', theoretical physics, quite ignorant of the fact that Munich was then one of the world centres of this subject. Here, the Theoretical Physics Institute was presided over by one of the great architects of the old quantum theory, Arnold Sommerfeld, some of whose lectures Fröhlich started to attend. In an interview given in 1983 [F188], he gave the following account of those early days:

> I went to one of Sommerfeld's lectures. There one learnt a great deal from the mistakes he made. He never used a manuscript; he would just start making his calculations on the blackboard, make a mistake and start looking for it. Sometimes one of the students would notice what the mistake was. One day I did that. After I had done that several times, he asked me to stay behind, and then said, 'From now on you will direct the problem class.' So I was already a theoretical physicist, and had to make up problems for the others. Then, at the end of my second undergraduate year, Sommerfeld gave me some papers to read during the vacation. I read these, then thought I could do something along those lines, and made a calculation. When I came back, I showed what I had done to Sommerfeld; he asked me to change this and that, and then said, 'Well, there's your thesis' [F188].[6]

Thus, in July 1930, during his third year—and without ever having taken an undergraduate degree (which would have taken 4 years!)—Fröhlich submitted his doctoral thesis on the photoelectric effect in metals, and received his D.Phil. (*Magna cum Laude*) on 22 July (Fig. 2.2).

After one year as a Research Fellow in Munich, during which time he was supported by a scholarship from the German Scientific Society's Emergency Fund, Sommerfeld came to tell him that Gustav Mie, who was then head of Physics Institute of the University of Freiburg (im Breisgau), wanted someone to introduce modern physics there. Since at that time there were very few jobs in theoretical physics, Fröhlich wasted no time in accepting Mie's offer, and in October 1931 went off to Freiburg; the alternative may well have been for him to become a skiing instructor, for he was an enthusiastic and highly competent skier!

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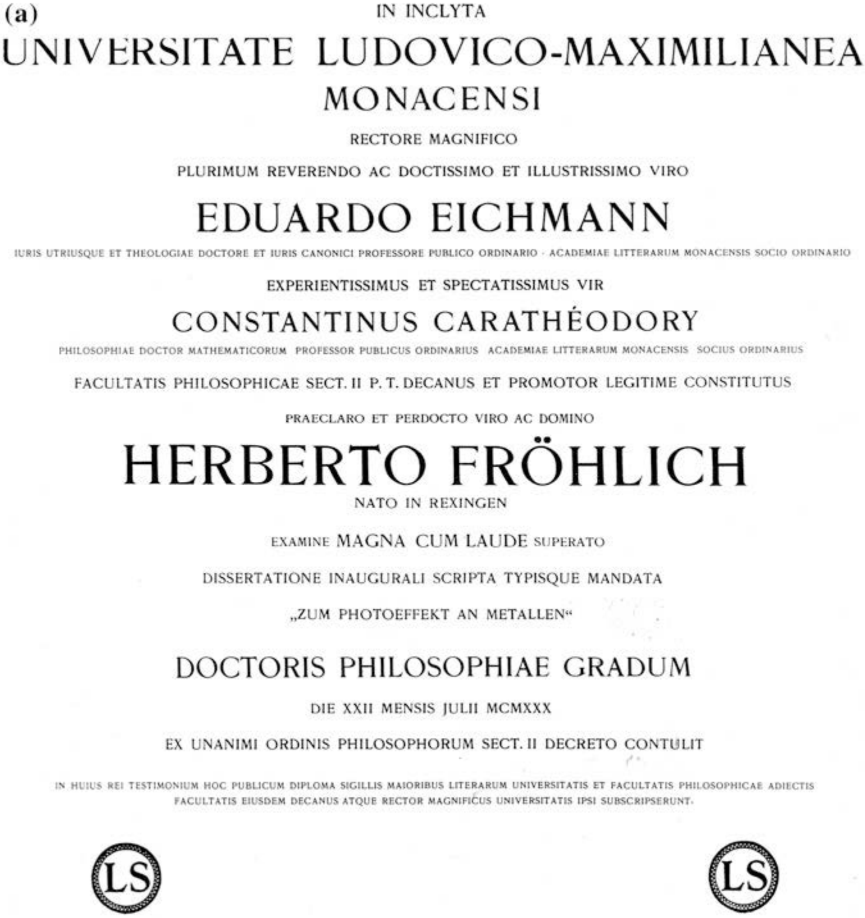
[6] This denotes reference (188) in the *Complete Bibliography of H. Fröhlich*, at the end of this book; similarly [Fx] denotes reference (x).



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MONACENSI
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PLURIMUM REVERENDO AC DOCTISSIMO ET ILLUSTRISSIMO VIRO
EDUARDO EICHMANN
IURIS UTRIUSQUE ET THEOLOGIAE DOCTORE ET IURIS CANONICI PROFESSORE PUBLICO ORDINARIO · ACADEMIAE LITTERARUM MONACENSIS SOCIO ORDINARIO
EXPERIENTISSIMUS ET SPECTATISSIMUS VIR
CONSTANTINUS CARATHÉODORY
PHILOSOPHIAE DOCTOR MATHEMATICORUM PROFESSOR PUBLICUS ORDINARIUS ACADEMIAE LITTERARUM MONACENSIS SOCIUS ORDINARIUS
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PRAECLARO ET PERDOCTO VIRO AC DOMINO
HERBERTO FRÖHLICH
NATO IN REXINGEN
EXAMINE MAGNA CUM LAUDE SUPERATO
DISSERTATIONE INAUGURALI SCRIPTA TYPISQUE MANDATA
„ZUM PHOTOEFFEKT AN METALLEN“
DOCTORIS PHILOSOPHIAE GRADUM
DIE XXII MENSIS JULII MCMXXX
EX UNANIMI ORDINIS PHILOSOPHORUM SECT. II DECRETO CONTULIT
IN HUIUS REI TESTIMONIUM HOC PUBLICUM DIPLOMA SIGILLIS MAIORIBUS LITERARUM UNIVERSITATIS ET FACULTATIS PHILOSOPHICAE ADIECTIS FACULTATIS EIUSDEM DECANUS ATQUE RECTOR MAGNIFICUS UNIVERSITATIS IPSI SUBSCRIPSERUNT.

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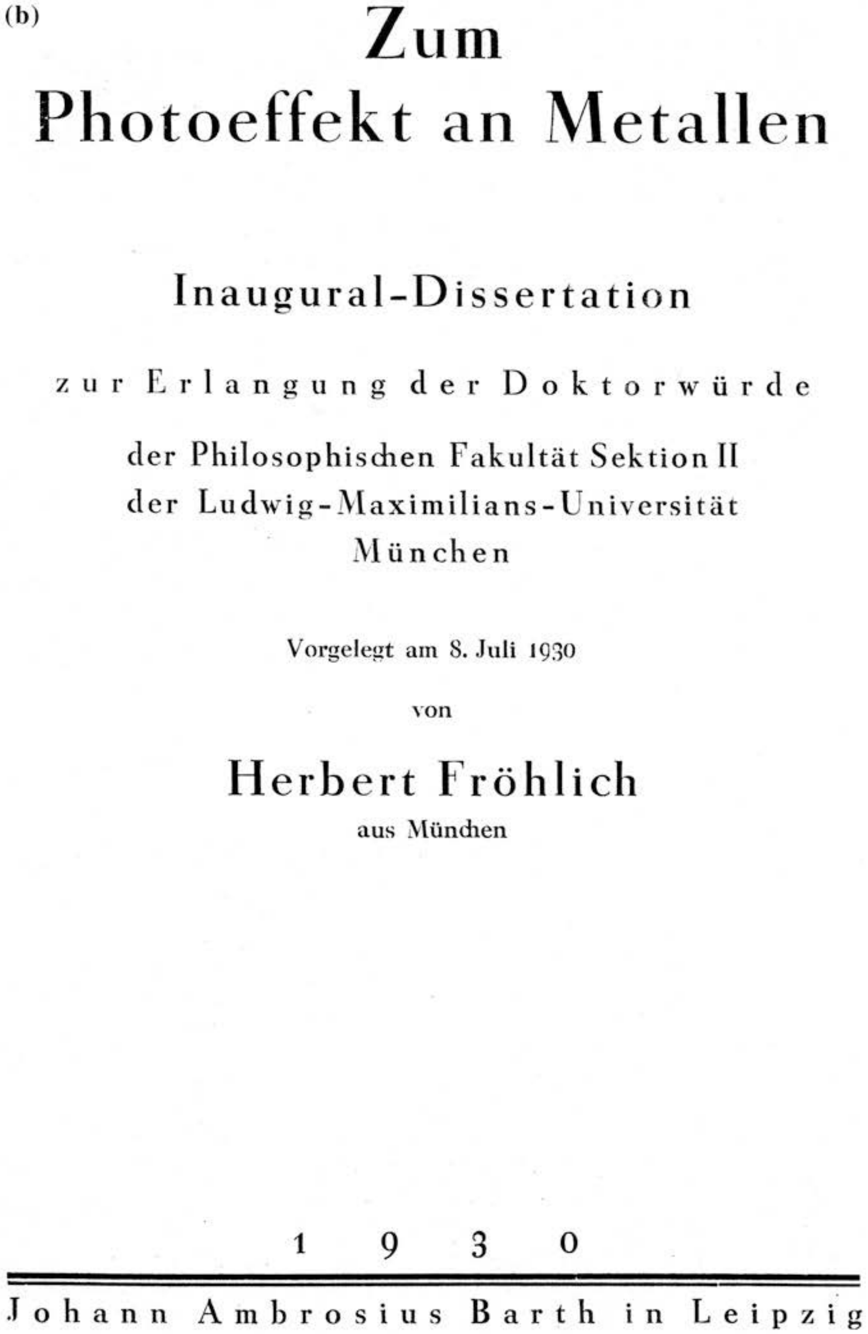
Fig. 2.2 **a** Fröhlich’s Doctoral Diploma from the University of Munich, 1930; **b** the cover of his Doctoral Dissertation [F1]



(b)

Zum
Photoeffekt an Metallen

Inaugural-Dissertation

zur Erlangung der Doktorwürde

der Philosophischen Fakultät Sektion II
der Ludwig-Maximilians-Universität
München

Vorgelegt am 8. Juli 1930

von

Herbert Fröhlich
aus München

1930

Johann Ambrosius Barth in Leipzig

Fig. 2.2 (continued)

Before leaving Munich, Fröhlich published two papers, the first [F1] being his D.Phil. thesis in which he extended W. Wentzel's theoretical treatment of the photoelectric effect, based on Sommerfeld's theory, to the case of thin metallic sheets, drawing attention to the importance of surface conditions, particularly in determining the difference between the normal and selective effects; his calculations agreed in all essential aspects with experiment, The second publication [F2], which proved to be way ahead of its time, arose from the following consideration: just as the form of the thermionic Richardson equation[7] for the current of electrons emitted from a hot metallic surface as derived from quantum mechanics, using Fermi-Dirac statistics, differs from the that given classically on the basis of Maxwellian statistics, so one might anticipate a corresponding difference in the expressions for the fluctuations in the electric current. This was shown to be indeed the case, there being, in addition to a contribution arising from the discreteness of the electronic charge (which turned out to be identical to that given by Schottky's classical derivation—so-called 'shot noise'), a contribution allied to the finiteness of Planck's constant, h, which was proportional to h^3. Unfortunately, however, it was found that the latter lay far below the limits of detection then possible. The real significance of this publication, however, was its application of the technique of so-called 'second quantization'[8] (*Quantelung der Amplituden*)—only 3 years after it had been extended to fermion systems by Jordan and Wigner in 1928 (Jordan and Wigner 1928)—to the Fermi gas constituted by the conduction electrons in a metal, which were treated as the quanta of an electron field, ψ, whose Fourier coefficients, a_k and $a_k^\dagger$, *anti*commute—i.e.

$$\psi = \sum_{\mathrm{k}}^{\infty} \frac{a_{\mathrm{k}}}{\sqrt{L}} e^{ikx+i\omega t}, \quad \text{where } k = n\pi/L,\ n = 1, 2, 3\ldots, \tag{2.2.1}$$

in the case of a one-dimensional system of length L, where, in respect of the Pauli principle,

$$a_k^\dagger a_k \equiv N_{\mathrm{k}} = 0, 1, \text{ and } a_k a_k^\dagger = 1 - N_{\mathrm{k}}, \tag{2.2.2}$$

the mean number of electrons (with momentum $\hbar k$ and energy $\hbar\omega$) being $\bar{N}_{\mathrm{k}}$ (Fig. 2.3).

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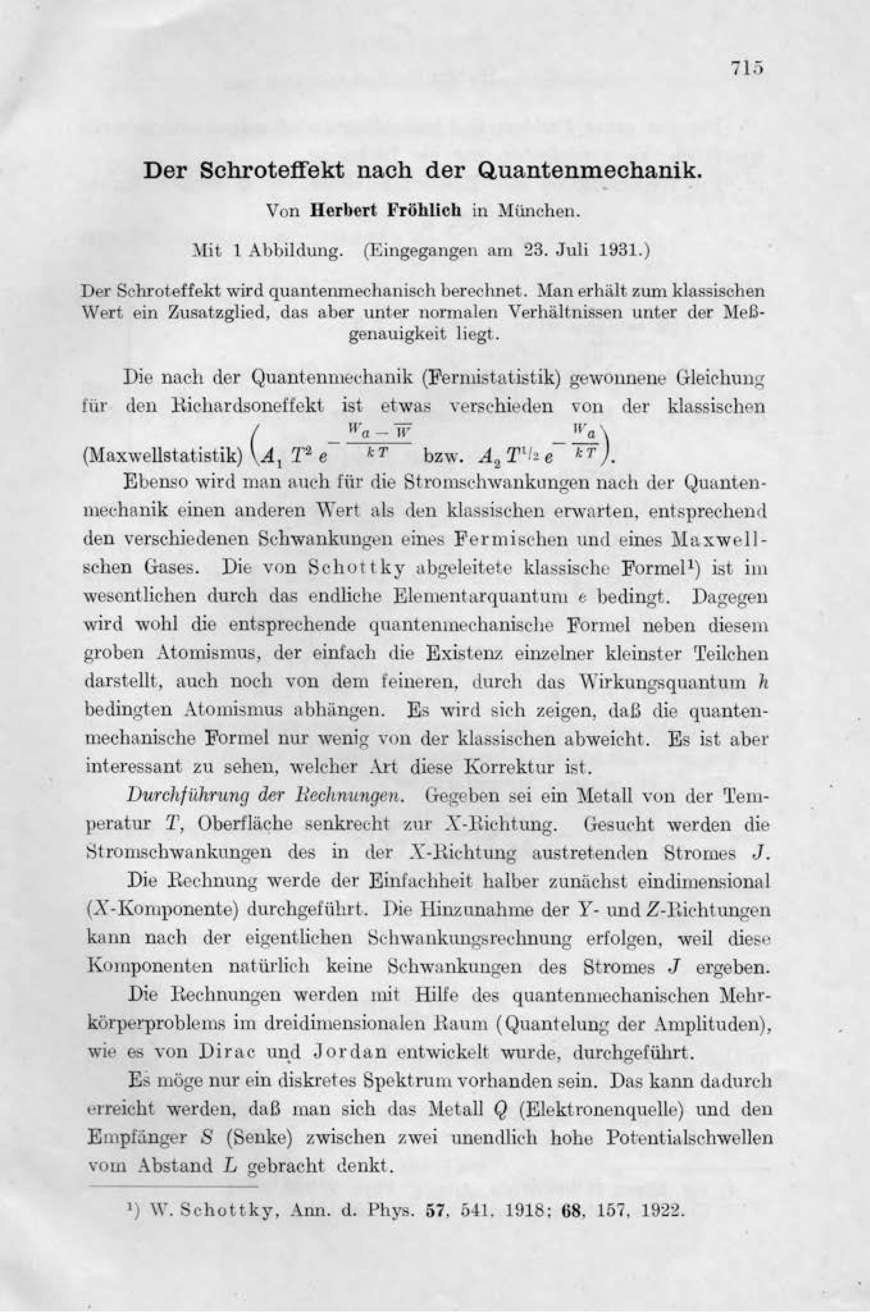
[7] The quantum mechanical form of this equation permits the work function, W, of a metal to be determined from measurement of the thermionic current, which is proportional to $T^2\, exp(-W/kT)$; classically, the pre-exponential factor becomes $T^{1/2}$, whilst the meaning of W is somewhat different.

[8] 'Second quantization' is a technique whereby a many-body system of identical particles is described by a quantum field in three-dimensional space and time, rather than by appropriately symmetrised many-particle Schrödinger wave-functions in multi-dimensional configuration space. The individual particles are the quanta of this field (just as photons are quanta of the electromagnetic radiation field) whose quantization rules dictate the particle statistics, bosons (fermions) being described by commutating (anti-commuting) fields. Many years later, as a spin-off of his work on the connection between micro and macrophysics, Fröhlich demonstrated the equivalence of the two approaches in a way that is very much simpler than that which is usually presented [F155].



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Der Schroteffekt nach der Quantenmechanik.

Von **Herbert Fröhlich** in München.

Mit 1 Abbildung. (Eingegangen am 23. Juli 1931.)

Der Schroteffekt wird quantenmechanisch berechnet. Man erhält zum klassischen Wert ein Zusatzglied, das aber unter normalen Verhältnissen unter der Meßgenauigkeit liegt.

Die nach der Quantenmechanik (Fermistatistik) gewonnene Gleichung für den Richardsoneffekt ist etwas verschieden von der klassischen (Maxwellstatistik) $\left(A_1\, T^2\, e^{-\frac{W_a - \overline{W}}{k\,T}}\right.$ bzw. $\left.A_2\, T^{1/2}\, e^{-\frac{W_a}{k\,T}}\right)$.

Ebenso wird man auch für die Stromschwankungen nach der Quantenmechanik einen anderen Wert als den klassischen erwarten, entsprechend den verschiedenen Schwankungen eines Fermischen und eines Maxwellschen Gases. Die von Schottky abgeleitete klassische Formel[1]) ist im wesentlichen durch das endliche Elementarquantum ε bedingt. Dagegen wird wohl die entsprechende quantenmechanische Formel neben diesem groben Atomismus, der einfach die Existenz einzelner kleinster Teilchen darstellt, auch noch von dem feineren, durch das Wirkungsquantum h bedingten Atomismus abhängen. Es wird sich zeigen, daß die quantenmechanische Formel nur wenig von der klassischen abweicht. Es ist aber interessant zu sehen, welcher Art diese Korrektur ist.

Durchführung der Rechnungen. Gegeben sei ein Metall von der Temperatur T, Oberfläche senkrecht zur X-Richtung. Gesucht werden die Stromschwankungen des in der X-Richtung austretenden Stromes J.

Die Rechnung werde der Einfachheit halber zunächst eindimensional (X-Komponente) durchgeführt. Die Hinzunahme der Y- und Z-Richtungen kann nach der eigentlichen Schwankungsrechnung erfolgen, weil diese Komponenten natürlich keine Schwankungen des Stromes J ergeben.

Die Rechnungen werden mit Hilfe des quantenmechanischen Mehrkörperproblems im dreidimensionalen Raum (Quantelung der Amplituden), wie es von Dirac und Jordan entwickelt wurde, durchgeführt.

Es möge nur ein diskretes Spektrum vorhanden sein. Das kann dadurch erreicht werden, daß man sich das Metall Q (Elektronenquelle) und den Empfänger S (Senke) zwischen zwei unendlich hohe Potentialschwellen vom Abstand L gebracht denkt.

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[1]) W. Schottky, Ann. d. Phys. **57**, 541, 1918; **68**, 157, 1922.

Fig. 2.3 The paper [F2] in which ‘second quantization’ was first applied to electrons in a metal—Reproduced by permission of Springer-Verlag

What was so impressive was not only his evident awareness, already at this early stage in his research career, of such a sophisticated and highly advanced technique of quantum field theory, but also his courage to be the first to apply it to a problem in solid-state physics. It was to be almost another 20 years, however, before Fröhlich fully realised the potential and utility of this technique in the solution of non-relativistic many-body problems (vide Chap. 5).

Once in Freiburg, in addition to lecturing on the new electron theory of metals, which had been born out of applying the recently developed quantum theory of Schrödinger to solid-state physics,[9] Fröhlich continued his researches in metal physics, publishing 5 papers. The first was a long paper [F3] that gave the first theoretical treatment of secondary electron emission from a sheet of metal on which cathode rays are incident; this early work later became rather well-known, and was still being cited almost half a century later (Rösler and Brauer 1981). It was found that in order to produce secondary electrons (whose energies are always below 25 eV), the primary electrons had to have an energy of a least of the order of 10 eV, this lower limit being determined mainly by the Pauli principle; for a primary energy of 100 eV, it was calculated that there is about one secondary electron for each primary. Finally, in the case of thin metallic sheets, it was predicted that the majority of secondary electrons are emitted from the face opposite to that on which the primary electrons are incident.

The second paper [F4] dealt with light absorption and the selective photoelectric effect; the third [F5] was devoted to the determination of the energy levels of metallic electrons from their optical constants; the fourth [F6] concerned the position of the absorption spectra of coloured alkali halides, in which he attempted to explain—on the basis of wave mechanics and assuming weakly bound electrons—the Mollwo relation between the frequency of the absorption maximum and the lattice constant of photochemically coloured alkali halide crystals at room temperature; the final paper [F7], *On the absorption of metals in the visible and ultraviolet*, was the subject of his *Habilitationsschrift*, which was submitted for publication in the journal *Zeitschrift für Physik*, in January 1933 (Fig. 2.4).

Fröhlich became *Privatdozent* at Freiburg in December 1932, and his appointment was confirmed by the State of Baden (to which Freiburg then belonged) the day before the state administration was dissolved early in 1933. It was only in 1932, the year before Hitler became Chancellor, that Fröhlich truly realised what was going on, during an outing with other staff and students during the Christmas vacation:

> It was only at this time that I realised what was going on. Nobody before took it seriously, so I don't think the people themselves took it seriously. On this outing we had a certain young man, an average student, neither good nor bad, not imaginative; and as he walked with me he said: 'When he comes to power, he will never leave again'. And then he said: 'The Jews have decided to destroy Germany'. And I said, 'How do you know this?

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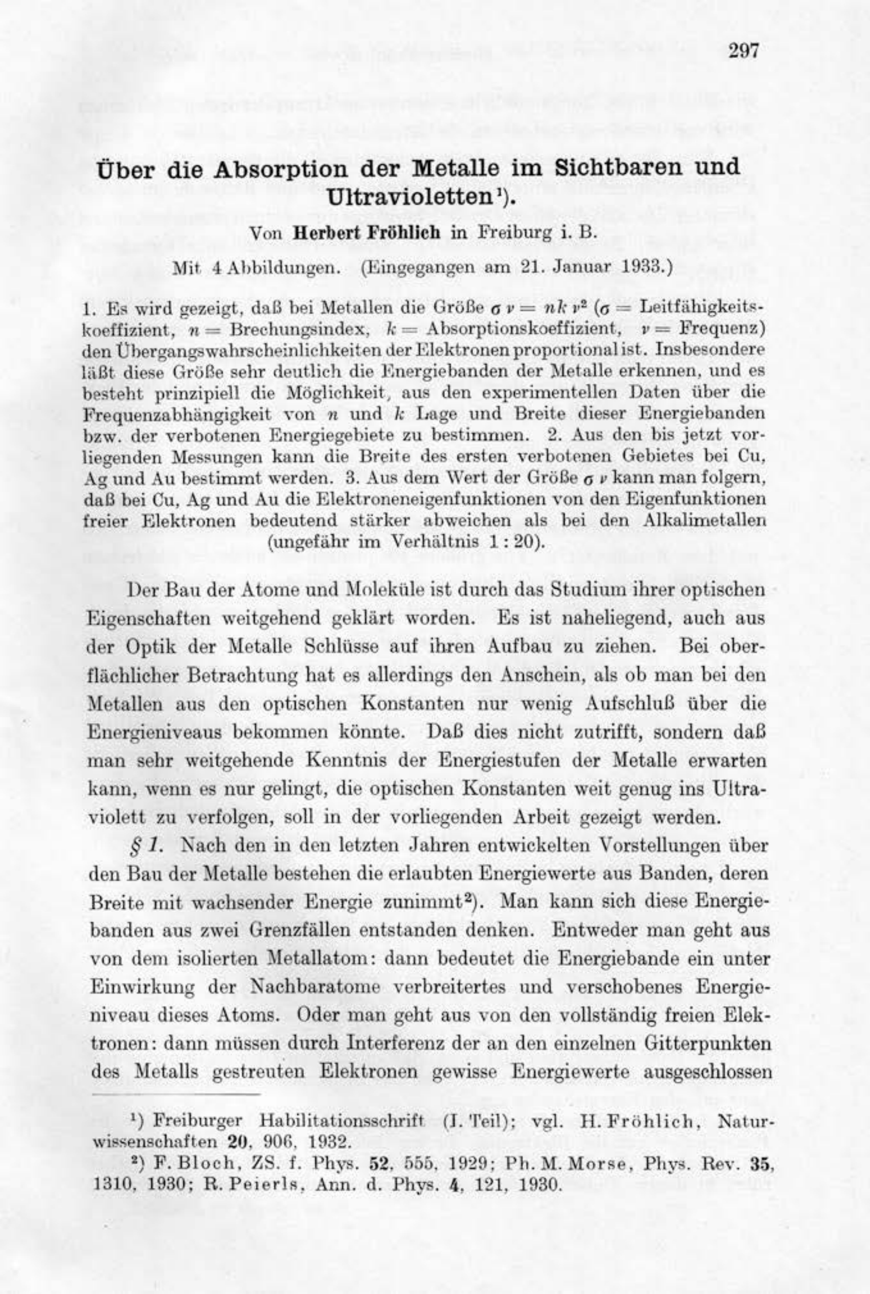
[9] This was an activity in which even Pauli participated (Pauli 1926), although he later came to despise it: 'I don't like this solid-state physics……though I initiated it (with his application of Fermi-Dirac statistics to calculate of the spin paramagnetism [Pauli paramagnetism] of the electron gas in a metal); one shouldn't wallow in dirt' (Hoddeson et al. 1992, p. 159).



Über die Absorption der Metalle im Sichtbaren und Ultravioletten[1]).

Von **Herbert Fröhlich** in Freiburg i. B.

Mit 4 Abbildungen. (Eingegangen am 21. Januar 1933.)

1. Es wird gezeigt, daß bei Metallen die Größe $\sigma \nu = n k \nu^2$ (σ = Leitfähigkeitskoeffizient, n = Brechungsindex, k = Absorptionskoeffizient, ν = Frequenz) den Übergangswahrscheinlichkeiten der Elektronen proportional ist. Insbesondere läßt diese Größe sehr deutlich die Energiebanden der Metalle erkennen, und es besteht prinzipiell die Möglichkeit, aus den experimentellen Daten über die Frequenzabhängigkeit von n und k Lage und Breite dieser Energiebanden bzw. der verbotenen Energiegebiete zu bestimmen. 2. Aus den bis jetzt vorliegenden Messungen kann die Breite des ersten verbotenen Gebietes bei Cu, Ag und Au bestimmt werden. 3. Aus dem Wert der Größe $\sigma \nu$ kann man folgern, daß bei Cu, Ag und Au die Elektroneneigenfunktionen von den Eigenfunktionen freier Elektronen bedeutend stärker abweichen als bei den Alkalimetallen (ungefähr im Verhältnis 1 : 20).

Der Bau der Atome und Moleküle ist durch das Studium ihrer optischen Eigenschaften weitgehend geklärt worden. Es ist naheliegend, auch aus der Optik der Metalle Schlüsse auf ihren Aufbau zu ziehen. Bei oberflächlicher Betrachtung hat es allerdings den Anschein, als ob man bei den Metallen aus den optischen Konstanten nur wenig Aufschluß über die Energieniveaus bekommen könnte. Daß dies nicht zutrifft, sondern daß man sehr weitgehende Kenntnis der Energiestufen der Metalle erwarten kann, wenn es nur gelingt, die optischen Konstanten weit genug ins Ultraviolett zu verfolgen, soll in der vorliegenden Arbeit gezeigt werden.

§ 1. Nach den in den letzten Jahren entwickelten Vorstellungen über den Bau der Metalle bestehen die erlaubten Energiewerte aus Banden, deren Breite mit wachsender Energie zunimmt[2]). Man kann sich diese Energiebanden aus zwei Grenzfällen entstanden denken. Entweder man geht aus von dem isolierten Metallatom: dann bedeutet die Energiebande ein unter Einwirkung der Nachbaratome verbreitertes und verschobenes Energieniveau dieses Atoms. Oder man geht aus von den vollständig freien Elektronen: dann müssen durch Interferenz der an den einzelnen Gitterpunkten des Metalls gestreuten Elektronen gewisse Energiewerte ausgeschlossen

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[1]) Freiburger Habilitationsschrift (I. Teil); vgl. H. Fröhlich, Naturwissenschaften **20**, 906, 1932.

[2]) F. Bloch, ZS. f. Phys. **52**, 555, 1929; Ph. M. Morse, Phys. Rev. **35**, 1310, 1930; R. Peierls, Ann. d. Phys. **4**, 121, 1930.

Fig. 2.4 Fröhlich's 1933 Freiburger Habilitationsschrift [F7]—Reproduced by permission of Springer-Verlag

> He answered: 'I've read the *Protocols of the Elders of Zion*'.[10] 'Do the Jews know this?', I asked: 'Of course', he said. 'Well I'm a Jew, and I don't know it, I replied, to which he answered: 'Well you must be a very great exception'. Although he was completely average in every way, he was also completely mad. And that was an insanity that had spread right through Germany [F188].

With the rise of Nazism, the appointment was short-lived, and he was dismissed in April 1933 under new 'cleansing' laws pertaining to Jews and other undesirables, the Law for the Restoration of the Professional Civil Service (*Gesetz zur Wiederherstellung des Berufsbeamtentums*) having been passed on 7 April 1933. Had this not have happened, Fröhlich would have probably succeeded Königsberger as Professor of Theoretical Physics in Freiburg. On the back of his letter of dismissal Fröhlich made some calculations, but later threw the letter into the waste paper basket. Thinking better of it, he retrieved it, thinking that the calculation might eventually come in useful; he records that this did, in fact, turn out to be the case after the War, but he always refused to reveal to what the calculation referred. Since the Easter holidays were approaching, he decided to go on a skiing holiday.

Just before Fröhlich's dismissal, his brother, Ali, who was then about 17 years old, lent a book on Trotsky to a friend who was distributing anti-Nazi leaflets. This person was later picked up by the *SS*, and unfortunately had the book, which had Ali's name on it, in his possession. Fearing that this would make Ali a target for the *SS*, his parents appealed to a policeman who lived in the same apartment block; he said that all he could do to protect Ali would be to arrest him as an 'Enemy of the State', and have him taken to a police station that was not yet under Nazi control. After spending the night in a cell, he was brought before a judge who gave him a sympathetic hearing and released him. Upon seeing him return home, the policeman's wife's immediate reaction was to ask 'You are not still here are you?' Ali took the hint, and left immediately for Strasbourg,[11] where he had relatives. When the *SA* (*Sturmabteilung*[12]) learned of Ali's release, and being unable to trace him, they took reprisal on his father, arresting him on 29 March at Fürstenfeldbruck railway station, and then beating him up; the next day, he was detained under *Schutzhaft*,[13] but was released a few days later on 3 April.

When Herbert returned from his skiing trip, family was afraid to come down to open the front door. Fortunately, his sister, Betty, who had moved to Palestine in 1926,

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[10] A fraudulent document that first appeared in Russia in 1905, in which was discussed a supposed Jewish plan for world domination; it was widely cited during the 1920s and 30s in defence of anti-semitism.

[11] Strasbourg is the capital of the Alsace region of eastern France, which borders Germany, and is only 25 km west of Rexingen where Herbert had been born.

[12] The original para-military wing of the Nazi Party, which played a key role in Hitler's rise to power during the 1920s and early 1930s. It became disempowered after Hitler's blood-purge in 1934 (Night of the Long Knives), being effectively then superseded by the *SS*.

[13] *Schutzhaft* was a kind of protective custody under the guise of which political opponents of the Nazi regime, and especially Jews, were rounded up so that they could be officially 'protected' from the wrath of the German population.

was staying with them at the time, and it was decided that the remainder of the family, apart from Herbert, would join her there. In the meanwhile, for safety, his parents moved later that Spring to Strasbourg, whilst Betty returned to Palestine to make the necessary arrangements. Herbert, however, remained in Munich, and attempted to raise money from the property and land his father had in Fürstenfeldbruck.

Having made some sales, Herbert arrived at the office of the family's solicitor to sign the relevant documents, only to be told: 'I'm very sorry, but one of my men is in the *SS*; everything is confiscated'. In great anger, Fröhlich immediately went to the Nazi offices and demanded to see the Chief Confiscator, a man by the name of Josef Meisinger.[14] On being led in, Meisinger's first words were: 'I've been waiting for you'. He then continued: 'I could, of course, keep you here, but I shall not do so if you collaborate. You must tell me all your father's bank accounts; we know them already, but we just want to see whether you will collaborate'. Realising what might befall him, he told Meisinger about some accounts, but not others. After that, he saw Meisinger a number of times in attempts to get him to release items that had been confiscated. One day, he took a pile of bills, urging him to release money so that they could be paid. Meisinger laughed and said: 'Every day your father goes and sits in the Café Aubette[15] in Strasbourg, and you mean to tell me he has no money for his son!' Fröhlich immediately decided he must go to Strasbourg to warn his father that his movements were being watched. At the border, he was taken off the train by the *SS*, who told him to remove his shoes:

> They thought I was so stupid as to try to take money out myself. So I took my shoes off, and said 'Do you want my socks too?' This impressed them, and they took me back again [F188].

His dealings with Meisinger continued:

> I felt no fear of this man. If I had shown the least sign of fear, he would have had me taken away and killed, without doubt. At one stage, he made a basic mistake. He said to me: 'I can't see you today, for I must go to Nürnberg to see the Führer'. So I thought to myself that since I was his personal victim, nobody else would touch me, and that this was a good opportunity to get away from Munich. So that same day, I took an antique dealer and a

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[14] Josef Meisinger (1899–1947) was born in Munich, and joined the *SS* is March 1933. In 1934 he moved to Berlin with Heydrich to the office of the Gestapo. In 1940 he was appointed Commander of State Police in Warsaw, where his atrocities appalled even his superiors. He was removed to Tokyo in 1941 where he acted as Gestapo Liason Officer until September 1945 when he surrendered himself to two American war correspondents. He was handed over to the Polish authorities in Warsaw, and executed in 1947 as a war criminal for atrocities in Poland, having become known as the 'Butcher of Warsaw'. Walter Schellenberg described Meisinger as 'one of the most evil creatures among Heydrich's bunch of thugs who carried out the vilest of his orders… He was a frightening individual, a large, coarse-faced man with a bald head and an incredibly ugly face. However, like many men of his type, he had drive and energy and an unscrupulous sort of cleverness' (Schellenberg 1956, pp. 160–161).

[15] An historic building on Place Kléber, built by Jacques-Françoise Blondel, 1765–1772, which still exists. Between 1926 and 1928 it was redecorated by Sophie Taeuber-Arp, Jean Arp and the De Stijl artist Theo van Doesburg.

> furniture dealer round to our house,[16] and they agreed to let me have a cash payment the following morning in return for the key. I then took the train that went to Basel. I knew it stopped on the German side of Basel to let the *SS* men on, so I got off as they got on. I had given my money to an organisation to bring out; you had to trust them, and they got 10 % [F188].

He then made his way to Strasbourg, some 113 km from Basel, to join his parents and brother who were awaiting their visas for Palestine.[17] Before leaving Munich, however, he found time to write a paper [F8] with Hans Bethe, a former student of Sommerfeld, which led to Fröhlich being invited to Russia the following year (vide Chap. 3). In this paper, which dealt with superconductivity, they showed, by exact analysis, that the magnetic interaction between electrons, which Frenkel, in a recent publication (Frenkel 1933), had invoked to account for the stability of the supercurrents, has actually *no* influence in this respect, the only effect being a tiny alteration in the effective mass of the electrons, which otherwise continued to obey the usual laws of non-superconductive conduction. The paper arose from a competition between Fröhlich and Bethe to be the first to spot any error in anything new that came out on superconductivity, and on this occasion it was Fröhlich who won. It must be appreciated that at this time (and indeed for many years to come) superconductivity was *the* central problem in solid-state physics, in that it continued to defy understanding in terms of the newly forged electron theory of metals that had otherwise been so spectacularly successful.

Despite his dealings with Meisinger, Fröhlich recorded that 'We never had any personal animosity towards each other; it was like a game of "cat-and-mouse", in which I was the mouse. But then, in a way, I enjoyed it, because I enjoy danger!' [F188].

One incident from this period that has hitherto never been narrated is the occasion on which Fröhlich was in a restaurant in Munich when Hitler entered. Fröhlich immediately realised that this was his opportunity kill Hitler with the steak knife he was holding. After a few moments reflection, however, he decided against it, notwithstanding the detestation with which he held Hitler, believing mistakenly that he would come to nothing, and was not worth the consequences that such an action would undoubtedly have entailed; accordingly, he continued with his meal. Even without a knife, Fröhlich, who was extremely fit, could easily have killed Hitler with his bare hands, having by then perfected his self-defence skills in order to protect himself against attacks from the Brownshirts (Fröhlich 1985).

---

[16] The family home at Seidlstraβe 22 contained some fine items of *art nouveau* furniture.

[17] With Herbert's financial assistance (made possible by encashing an insurance policy on his father's life and from the proceeds of the sale the family property (in both Munich and Fürstenfeldbruck), his parents and brother moved to Palestine in 1934; his father died there in 1952, and his mother in 1959.

Photo Gallery

The numbers in round brackets at the end of each caption correspond to the section of the text to which the photograph refers.

Unless stated otherwise, all photographs are from private collections.

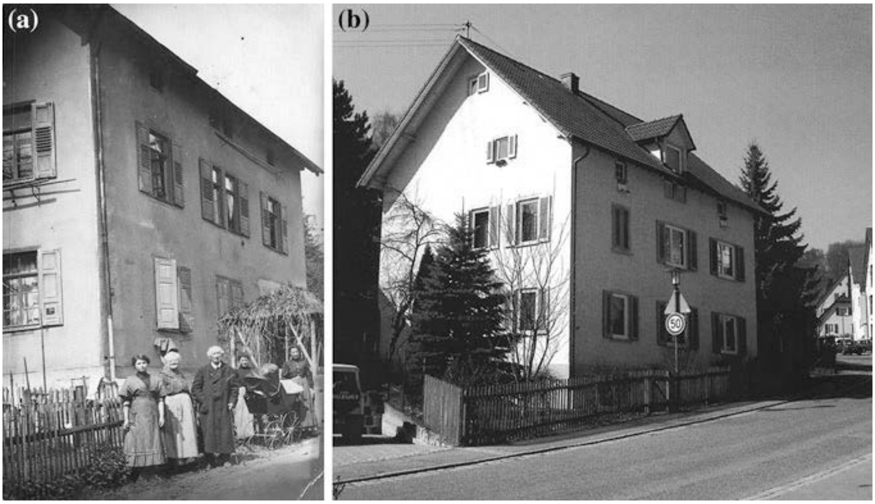
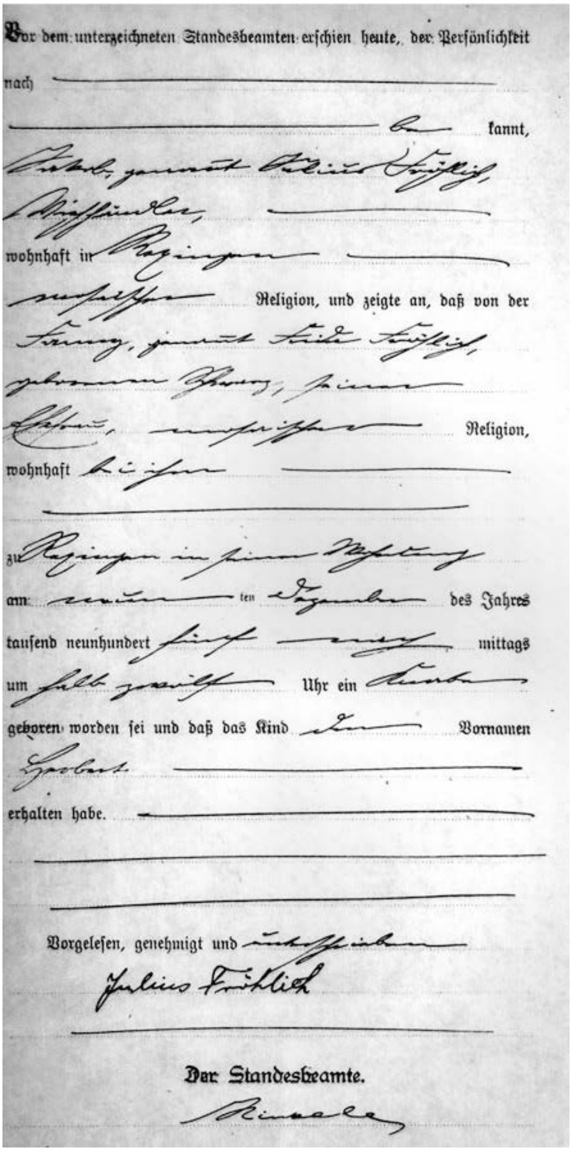


Fig. 2.5 **a** Fröhlich's birthplace in Rexingen, in the home of his paternal grandparents (shown centre, in 1911) at Freudenstädter Straβe 31 (formerly, Im Brühl 186); **b** the same in 2005 (Sect. 2.1)



Vor dem unterzeichneten Standesbeamten erſchien heute, der Perſönlichkeit
nach
be kannt,
Jakob, genannt Julius Fröhlich,
Viehhändler,
wohnhaft in Rexingen
israelitischer Religion, und zeigte an, daß von der
Fanny, genannt Frida Fröhlich,
geborene [illegible], seiner
Ehefrau, israelitischer Religion,
wohnhaft bei ihm
zu Rexingen in seiner Wohnung
am neun ten Dezember des Jahres
tauſend neunhundert fünf vor mittags
um halb zwölf Uhr ein Knabe
geboren worden ſei und daß das Kind den Vornamen
Herbert
erhalten habe.

Vorgeleſen, genehmigt und unterschrieben
Julius Fröhlich

Der Standesbeamte.
Kinkele

Fig. 2.6 Entry of Fröhlich's birth in the Registry Office Records in Rexingen (Sect. 2.1).

In front of the local official (who signed this form) there appeared today the person known as Jakob (Julius) Fröhlich, dealer in livestock and cattle, residing in Rexingen, of Jewish faith, and he announced that Fanny (Frida) Fröhlich, his wife, of Jewish faith, residing with him in Rexingen, gave birth on 9 December 1905 at 11.30 am to a male child, and that the child has been given the name of Herbert. Read aloud, authorised and signed: Julius Fröhlich/the Local Official: Kinkele—Reproduced by courtesy of Barbara Staudacher, Rexingen

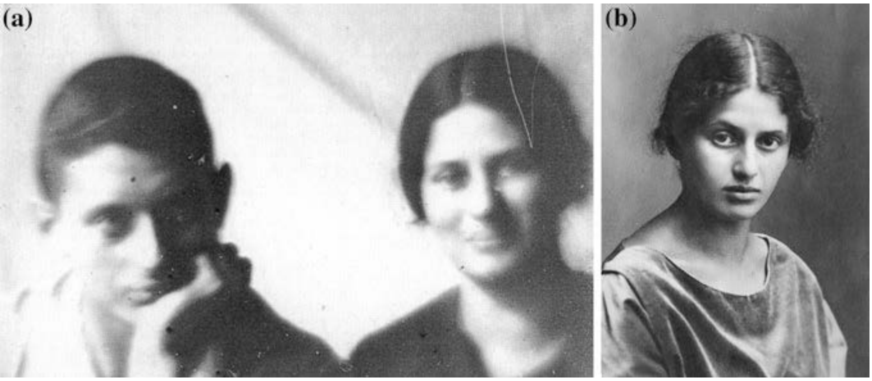


Fig. 2.7 **a** Fröhlich with his sister Betty (*b*.1904); **b** Betty, some years later, *c*.1926 (Sect. 2.1)

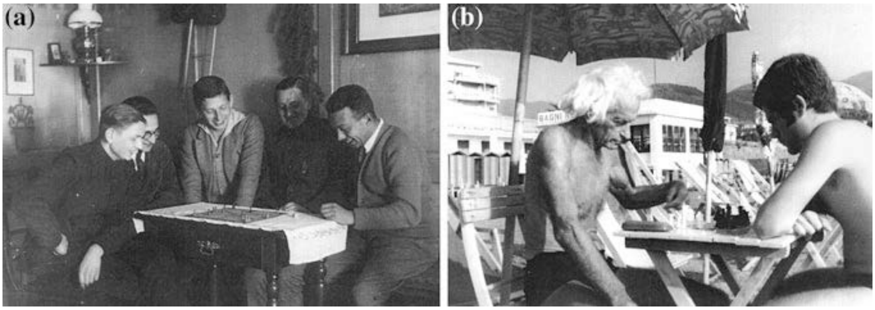


Fig. 2.8 **a** Fröhlich (*centre*) enjoying a game of chess with friends in Munich; **b** another game in Levanto, Italy many years later (Sect. 2.1)

Fig. 2.9 Fröhlich on a skiing trip with friend Eva (Sect. 2.1)

Fig. 2.10 Possibly on a *Jungjüdische Wandervogel* outing (Sect. 2.1)

Fig. 2.11 Fröhlich's parents, Julius and Frieda, in Marienbad, 1914, two years before their second son, Ali, was born (Sect. 2.2)

Fig. 2.12 Fröhlich near his home in Seidlstraβe, Munich, *c*.1930, aged 25 (Sect. 2.2)

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Chapter 3
Moving on: The Years of Exile

3.1 From Germany to Russia, via England

Whilst in Strasbourg, Fröhlich received an invitation from Frenkel to go—as a ‘Foreign Expert’—to the Physico-Technical Institute in what was then Leningrad (now St. Petersburg); the Director of the Institute was A.F. Joffe. This invitation arose from the paper with Bethe [F8], mentioned in Chap. 2, which turned out to be the last one Fröhlich wrote before leaving Germany, in which, it will be recalled, they had shown that a recent attempt by Frenkel (1933) to understand certain aspects of superconductivity was flawed.

Before eventually deciding to accept the Leningrad position, however, Fröhlich explored a number of other possibilities. In June 1933, he wrote to Fowler in Cambridge asking for help in finding a position in England. Fowler immediately forwarded his letter to Redcliffe Salaman of the Academic Committee of the Central Jewish Relief Fund in England. The following month (on 15 July), Fröhlich wrote directly to Walter Adams, the General Secretary of the Academic Assistance

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In this chapter ‘Moving on’ is taken from the title of an interview Fröhlich gave in 1983 [F188], in which he recalled his wartime experiences.

G.J. Hyland, *Herbert Fröhlich*,
Springer Biographies, DOI 10.1007/978-3-319-14851-9_3

Fig. 3.1 Fröhlich as ‘Foreign Expert’ in Leningrad (St. Petersburg), 1934

Council (*AAC*)[1] in the *UK*, requesting financial support. By October, he was in London where he met Blackett to whom he showed a letter he had from Schrödinger (whom he had first met in Munich[2]), expressing the hope that the necessary financial support could be found to enable Fröhlich to work with him in Oxford.[3] Blackett considered this to be a very strong recommendation, and accordingly wrote (as also did Mie on 3 November, Sommerfeld on 6 November and Schrödinger on 7 November) to the *AAC* supporting his application for funding; Sommerfeld had earlier recommended Fröhlich for the position of Assistant to S.N. Bose. Despite this level of support, Fröhlich was informed later in November that the *AAC* was unable to make any grant at that time, but hoped to be able to do so in another 3 months.

Fröhlich was apparently still in London in January 1934, when he visited the General Secretary of the *AAC* who recorded that Fröhlich was not enamoured with the possibility of a position in New Zealand (a possibility that had evidently been discussed), preferring instead to go to Russia to take up Frenkel's offer of a 'proper' position in Leningrad, rather than continuing to seek refugee grants elsewhere—a quest in which he had thus far been singularly unsuccessful. He accordingly contacted Frenkel, accepting his offer, and was told to go to Paris to arrange his visa. This he did, but it took another 6 months before the visa materialised.

In the meantime, he returned to Strasbourg, and went to the Physics Department of the University, where he started writing his first book, *Elektronentheorie der Metalle*, on the new electron theory of metals, a field to which he himself had already actively contributed, having published, by then, some 8 papers [F1–F8]. His motivation for writing the book—apart from financial—was to provide an account of recent developments in a form that was accessible to experimentalists, in the

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[1] The *AAC* was set up on 24 May 1933, at the instigation of William Beveridge, as a British response to help academics from central Europe who had fled from Nazi persecution; its first Chairman was Ernest Rutherford. Initially, its funds, which had been raised by public subscription, were limited to about £13,000. Aware of the imminent influx of refugee scientists from Europe, Lindemann was instrumental in extracting additional financial support from companies such as *ICI*. In 1936, when the threat to free learning had spread beyond Germany, the *AAC* became the Society for the Protection of Science and Learning (*SPSL*), whose Academic Assistance Fund provided research fellowships to distinguished academics. In 1997, the *SPSL c*hanged its name to the Council for Assisting Refugee Academics (*CARA*), and continues to assist refugee academics. Much of the information given in Sect. 3.1 comes from archives of the *SPSL*, which are now administered by *CARA; they can be consulted in the Department of Special Collections of the Bodleian Library, Oxford.*

[2] Fröhlich first met Schrödinger by accident in May 1930 when Schrödinger (during a visit to Munich to give a lecture entitled '*The transformation of the physical concept of the world*') mistakedly entered Fröhlich's room in the Theoretical Physics Institute looking for Sommerfeld, whom he had long known. Having knocked on the door, Schrödinger entered and said: 'I am Schrödinger, and who are you? Is Mr. Sommerfeld here?' Fröhlich then directed him to Sommerfeld's room next door.

[3] Schrödinger had been elected to a Fellowship at Magdalen College prior to his arrival in Oxford on 4 November 1933. A letter from Schrödinger (by then in Oxford) in support of Fröhlich is reproduced in Appendix 2 at the end of this chapter.

hope that this might encourage further experimentation, which, in turn, might hopefully catalyse further theoretical developments.

Fröhlich travelled to Russia by train, spending a week in Warsaw *en route*. At the Russian border, luggage had to be moved to a different train, and he recorded that: 'I didn't give any tip to the porters because I felt that now the country belonged to them, it would be insulting'. Upon arriving in Leningrad (probably) in mid 1934, he was told that since his apartment was not quite ready he would be accommodated in a hotel—the famous *Hotel Astoria* in Saint Isaac's Square, next to the Cathedral, and across from the historic Imperial German Embassy. On entering the hotel, he noticed that Frenkel gave a very big tip to the porter:

> So I then worked out how often I could go in and out of my room on my salary….The question of money was worked out very soon, for whenever someone came to see me from another institute it was called a 'consultation', and I was paid the equivalent of a third of my monthly salary [F188].

After eventually settling in at the Physico-Technical Institute, he continued writing his book, having been further encouraged of the need for such a work from his discussions with experimentalists there. The only treatment available at the time[4] was the *Handbuch* article (of the same title) by Sommerfeld and Bethe, dating from 1933. A contact that proved to be particularly significant was M.P. Bronstein[5] who, following A.H. Wilson in Cambridge (Wilson 1931), had developed a rather detailed theory of *semiconductors* (Bronstein 1932, 1933)—then a quite new field, somewhat disparagingly known as 'dirt physics' ('*Dreckphysik*') because of the crucial role played by impurities in determining the electronic properties of these materials. Accordingly, Fröhlich decided to include a chapter on them in his book, which being, for some time, the *only* textbook to contain a treatment of semiconductors, later proved to be very useful. It is likely that Fröhlich was already acquainted with semiconductors from his time in Freiburg, where the Professor of Theoretical Physics, Königsberger, was one of the pioneers in this field, being the first to propose an activated electrical conductivity ($\sim e^{-E/kT}$) based on the dissociation of metallic atoms into electrons and positive ions. Fröhlich's decision to get involved with semiconductors was, at the time, very courageous, given the low esteem in which work in this field was then held. Typical of this attitude was Pauli's remark:

> One should not work on semiconductors, they are a mess; who knows whether there are semiconductors at all!
>
> (Busch 1989, p. 255).

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[4] Mott and Jones' book, *Theory of the properties of Metals and Alloys*, was not published until 1936.

[5] In addition to his work on semiconductors, Bronstein was a pioneer of quantum gravity, author of works in astrophysics, quantum electrodynamics and cosmology, and wrote a number of popular science books for children. In August 1937, he was arrested during one of Stalin's purges, and executed on 18 February 1938.

Whilst in Leningrad, Fröhlich was offered, and accepted, a part-time position at the Technical University, which required his attendance only one day per week, but for which he received a salary almost equal to that of his full-time job at the Physico-Technical Institute. This additional appointment increased his circle of scientific contacts, some of whom went on to become quite well-known physicists. He enjoyed his time in Leningrad where he was able to maintain a reasonable standard of living and still have money to spare; for his status as a 'foreign expert' permitted him to purchase items that would have otherwise been unobtainable.

This agreeable lifestyle turned out to be short-lived, however. For with the assassination of Sergei Kirov, the Head of the Communist Party in Leningrad, on 1 December 1934, Stalin launched his campaign of terror, otherwise known as the Great Purge. Given his experience in Munich, Fröhlich knew immediately what was happening, and appreciated the seriousness of the situation. As a foreigner, he had to renew his Residence Permit every three months, which involved leaving his passport at an office and collecting it some time later. Being aware that some foreigners had already been expelled or deported to camps, he decided to collect his passport early. Without informing him, an Exit Visa had been placed inside his passport, and it was only by chance that he discovered it: more importantly, he read it to find that the Visa was valid for only 5 days, 4 of which had already gone! He returned immediately to the Institute to inform his colleagues who could not believe what had happened, assuming that there must have been some mistake. Believing the contrary, however, Fröhlich immediately left the house in which he was staying—a course of action that turned out to be fortuitous, since he later learned that the police had come around the next day to see if he was still there. Unable to take out of Russia any of the vast number of roubles he had amassed (since there was little to spend them on), he bought a large quantity of caviar, some semi-precious stones (both of which he later sold), and an ill-fitting suit with a label 'Made in England', which turned out to be a good omen! (Fig. 3.1). Fortunately, he succeeded in finding a ticket counter at the railway station at which it was still possible to use internal roubles to buy a ticket to a destination outside of Russia. Vienna was his preference—Austria not having by then been annexed by the Nazis—but there were no tickets left. He was, however, offered a ticket for a First Class sleeper carriage on a train that went to Rome, via Vienna; being well able to afford it, he bought the ticket.

Just prior to leaving Leningrad, Fröhlich was approached privately by Vladimir Fock who asked him whether, once out of Russia, he might be able to do something to help the plight of Dmitri Ivanenko, given that the authorities were rather sensitive about what was said about them in the West. Ivanenko was a senior Research Fellow and a colleague of Fock and Fröhlich at the Physico-Technical Institute. He had been arrested on 27 February 1935 in the wake of Kirov's assassination—on the grounds that he was a 'socially dangerous element'—and sent to a corrective labour camp in Karaganda in Central-Eastern Kazakhstan. Once out of Russia, Fröhlich did indeed take up Ivanenko's case with the help of Pauli and Dirac, and, although unbeknown to him until 50 years later, was successful. For, by December 1935, Ivanenko's sentence had been commuted to exile in Tomsk, Western Siberia, where he became a professor and Head of the Department of Theoretical Physics between 1936 and 1938. In 1985, when Fröhlich was in Japan to speak at a conference marking the 50th anniversary of the publication of Yukawa's first paper on meson theory, Ivanenko introduced himself, and gave him a reprint of a publication marking his own 80th birthday on which he had written: '*An Herrn Fröhlich, mein bester freund*'. Fröhlich wondered for some time why he was Ivanenko's 'best friend': then he suddenly remembered!

By the time he left Russia in May 1935, Fröhlich had published 2 papers. The first [F9], which was communicated by Fowler to the Proceedings of the Cambridge Philosophical Society on 22 December 1934, dealt with the concept of the number of free electrons, N_F, in a metal from the point of view of quantum mechanics; without having to make any assumptions about the potential field in the metal, he derived the following expression for N_F:

$$N_F = (4/3)D(E_F)E_{\mathrm{kin}}(E_F), \qquad (3.1.1)$$

where $D(E_F)$ the density of states (neglecting spin) at the Fermi energy, E_F, and $E_{\mathrm{kin}}(E_F)$ is the average kinetic energy of those electrons whose total energy is E_F. He noted that his expression included the trivial case of free electrons, and does not explicitly contain the number of valence electrons (Fig. 3.2).

In the second paper [F10], which he submitted to *Physikalische Zeitschrift der Sowjetunion* on 15 March 1935, he calculated the work function in Sommerfeld's model of a metal. A third paper [F11], entitled 'The inner photoelectric effect in semiconductors', dealt with the electron distribution function for a semi-conductor illuminated by monochromatic light, and with the associated Dember effect (the creation of a voltage across a conductor or semiconductor by illuminating one side of it); it was submitted to the same journal on 2 September, two months after his arrival in England, during his stay with Peierls in Manchester, prior to moving to Bristol.

3.2 From Russia to England, via Vienna

Having succeeded in getting out of Russia by train, Fröhlich alighted in Vienna. There he eventually managed to exchange some of his remaining roubles and sell his semi-precious stones, together with the remainder of his rail ticket covering the onward part of the journey to Rome. He then went to the university in an attempt to secure a position in theoretical physics. Although unsuccessful, he did obtain the equivalent of a month's salary for helping someone with a paper to be presented at a forthcoming conference on the 'Metallic State', which was to be held at the University of Bristol in the *UK* at the beginning of July 1935. Having no other prospects, Fröhlich wrote to the organiser of the conference, who happened to be Arthur Tyndall,[6] the Head of the Physics Department, asking if he could attend

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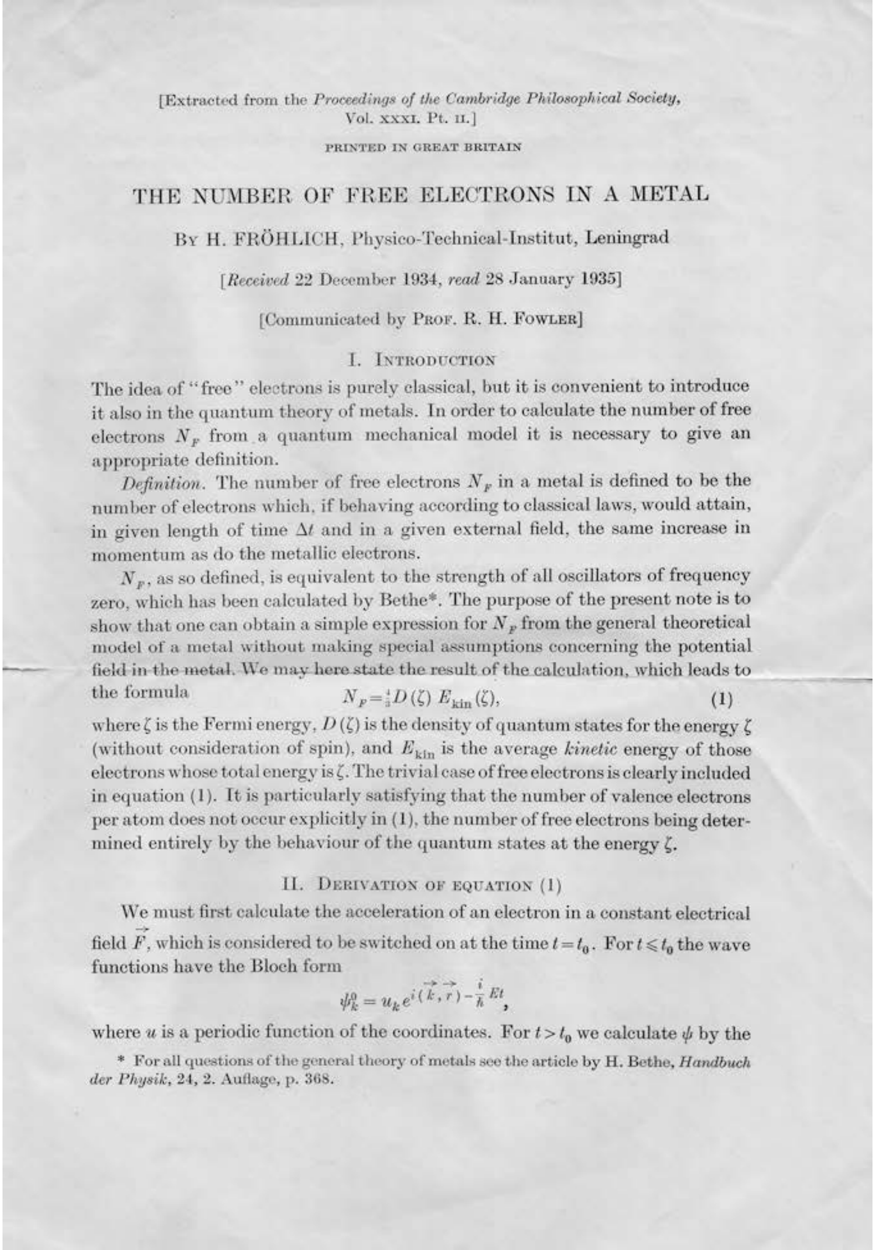
[6] Arthur Mannering Tyndall (1881–1961) was Acting Head of the Bristol Physics Department from 1910, and Henry Overton Wills Professor of Physics from 1919 until his retirement in 1948.



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THE NUMBER OF FREE ELECTRONS IN A METAL

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[Communicated by PROF. R. H. FOWLER]

I. INTRODUCTION

The idea of "free" electrons is purely classical, but it is convenient to introduce it also in the quantum theory of metals. In order to calculate the number of free electrons N_F from a quantum mechanical model it is necessary to give an appropriate definition.

Definition. The number of free electrons N_F in a metal is defined to be the number of electrons which, if behaving according to classical laws, would attain, in given length of time Δt and in a given external field, the same increase in momentum as do the metallic electrons.

N_F, as so defined, is equivalent to the strength of all oscillators of frequency zero, which has been calculated by Bethe*. The purpose of the present note is to show that one can obtain a simple expression for N_F from the general theoretical model of a metal without making special assumptions concerning the potential field in the metal. We may here state the result of the calculation, which leads to the formula

$$N_F = \tfrac{1}{3} D(\zeta)\, E_{\mathrm{kin}}(\zeta), \qquad (1)$$

where ζ is the Fermi energy, $D(\zeta)$ is the density of quantum states for the energy ζ (without consideration of spin), and E_{kin} is the average *kinetic* energy of those electrons whose total energy is ζ. The trivial case of free electrons is clearly included in equation (1). It is particularly satisfying that the number of valence electrons per atom does not occur explicitly in (1), the number of free electrons being determined entirely by the behaviour of the quantum states at the energy ζ.

II. DERIVATION OF EQUATION (1)

We must first calculate the acceleration of an electron in a constant electrical field $\vec{F}$, which is considered to be switched on at the time $t = t_0$. For $t \leqslant t_0$ the wave functions have the Bloch form

$$\psi_k^0 = u_k e^{i(\vec{k}, \vec{r}) - \frac{i}{\hbar} Et},$$

where u is a periodic function of the coordinates. For $t > t_0$ we calculate ψ by the

---

* For all questions of the general theory of metals see the article by H. Bethe, *Handbuch der Physik*, 24, 2. Auflage, p. 368.

Fig. 3.2 A paper [F9] written in Leningrad, 1934. Reproduced with the permission of the Cambridge Philosophical Society himself. Tyndall agreed, and having financed his own travel from the money raised in Vienna, Fröhlich arrived in England on 30 June 1935, when he was granted a Visitor Permit valid for one month.

The person Fröhlich helped in Vienna was Hermann Mark,[7] who had been Professor of Physical Chemistry at the University of Vienna since 1932. In a letter to Max Born in Cambridge, dated 17 June 1935, Mark records that he had had several discussions with Fröhlich, which he (Mark) had found ‘instructive and helpful’, and that together they had come to the view that it could be of interest to set up experiments to study the dependence of the electrical and optical properties of metals on crystal size; Fröhlich later pursued this theoretically, in the context of the electronic specific heat, during his 6 months stay in Leiden in 1937 (vide infra). So impressed was Mark that, in his letter to Born, he asked if there was any way that funds (£180) might be made available to enable Fröhlich to work with him on this topic in Vienna. On 26 June, Born wrote to the General Secretary of the *AAC* supporting Mark’s initiative for Fröhlich, commenting that metallurgical firms should be very much interested in the kind of work proposed on small metal particles, and asking if the *AAC* could help to raise the money.

Before leaving Vienna, Fröhlich co-authored with E. Guth of the Theoretical Physics Institute (and a collegue of Mark) a short Letter to *Nature* [Nature **136**, 179 (1935)] pointing out a flaw (relating to symmetrisation) in a recent article in *Nature* by G. Temple, which claimed that the general principles of quantum mechanics entail the paradox that any two operators representing physical variables must commute. *For some reason, Fröhlich never included this paper in his list of publications, and, accordingly, it has been omitted from the Complete Bibliography at the end of this book.*

After the Bristol conference, which finished on 5 July 1935, and which was reported on in *Nature*, **136**, 115 (1935)—*see* Appendix 1 at the end of this chapter—Fröhlich stayed for a while in Manchester with Peierls who, on 6 July, wrote to the *AAC*, likewise supporting the Vienna initiative, and urging them to find the necessary funding. On 10 July, Peierls received the response that ‘they did not think temporary employment in Vienna would really be to Fröhlich’s advantage, since the conditions in Austria are utterly hopeless.’ The letter went on to say that if funds of £100 or more were available, it would be far better for Fröhlich to use them in England. Although, at that time, they stated that they did not know from what source money could be found—even for his temporary support—by 22 July, the *AAC* had somehow managed to arrange a grant of £182 for Fröhlich, to be spread over 12 months starting on 1 August 1935. In discussion with the *AAC*, Fröhlich agreed

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[7] Mark (1895–1992) knew Schrödinger (having served with him in the army during the First World War), and it is possible, given the good impression Fröhlich had made on Mark, that Mark was (at least partly) instrumental in arranging the afore-mentioned testimonial for Fröhlich from Schrödinger, whose acquaintance Fröhlich had already made in Munich in 1930 (vide Footnote 2 above). Mark is known as the ‘father of polymer science’, and, in collaboration with E. Guth, developed the statistical theory of the rubber molecule, which eventually led to the theory of cross-linked rubber elasticity. He left Austria in 1938, and eventually settled in the *USA* [http://archive.today/E0ce].

that it was not in his interest to use the grant to work in Vienna, and four alternatives were discussed: London (with Blackett), Cambridge (with Fowler), Bristol (with Mott), and Trinity College Dublin (with Ditchburn). Despite the fact that Ditchburn's interests were probably closest to Fröhlich's at that time, Dublin was soon discounted on the grounds that there he would be rather isolated with inadequate library facilities—a circumstance that would have necessitated repeated and expensive visits to England. Finally, by 30 July 1935, it was agreed with Mott that Fröhlich should go to Bristol at the end of September that year.

Fröhlich's arrival in Bristol was, however, slightly delayed because of a visit to Palestine, where his parents, brother and sister were all now living. On 23 September 1935, he was invited to lecture at the Physical Institute of the University of Jerusalem, and during his visit explored the possibility of a position there upon the termination of his *AAC* grant. Evidently, Palestine was attractive to him, for he approached the *AAC* to ask if they could investigate what prospects there might be there. Accordingly, on 16 October, the General Secretary of the *AAC*, Walter Adams, wrote to Chaim Weizmann of the *Zionist Organisation* in London to seek his help in this matter, describing Fröhlich as 'one of the best of the younger German scientists', and offering to make his *AAC* grant available for use in Palestine should a suitable position be found. Weizmann discussed Fröhlich's case with his friends and contacts (including Ornstein who, although based in Holland, was much involved in the establishment of a physics department in Jerusalem) who all agreed with his assessment of Fröhlich, and promised to pursue the matter upon his return to Palestine. Things dragged on until June the following year (1936) when Fröhlich was finally informed that there was no prospect of a position in Palestine, the only vacancy in theoretical physics having already been offered to Felix Bloch.[8] In the meanwhile, Fröhlich seriously considered[9] accepting a second invitation from Frenkel to return to Russia (the General Secretary of *AAC* actually writing to the Russian Ambassador in London in this connection), this time to work at an institute that, unlike the Physico-Technical Institute where he had been originally, had not yet been given over to military research. As it turned out, this did not happen, and he remained in Bristol where he was supported by the *AAC* grant, which expired in August 1936. After this, although unable to offer him a permanent position, the university did make available a small grant enabling him to stay on for a further 6 months.

Already in Bristol at this time were a number of other Jewish refugee physicists from Nazi Germany, the most senior of whom was Walter Heitler, a former student of Sommerfeld and Herzfeld in Munich. Others included the experimentalist Heinz London, the somewhat younger Kurt Hoselitz, and later Robert Sack who arrived in 1938; also there was the (non-Jewish) political refugee (and future spy), Klaus Fuchs, who was then one of Mott's doctoral students.

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[8] Bloch, who was at the time in Stanford, turned down the offer, and remained in Stanford where he was appointed to a Full Professorship. He did, however, accept an Honorary Doctorate from the University of Jerusalem in 1962.

[9] Why he should ever have entertained this course of action is not at all clear, given his earlier experience there, and the fact that Stalin was still conducting his Great Purge.

Appendices

Report in Nature on the conference 'The Metallic State', held in Bristol, 1935

(note the mention of Prof. H. Mark from Vienna)
—*Reproduced with the permission of Nature*

The Metallic State

A CONFERENCE on "The Metallic State" was held in the University of Bristol at the H. H. Wills Physics Laboratory on July 2–5, about forty visitors being present from other parts of England and from the Continent. Most of the visitors were housed in Wills Hall, one of the University halls of residence.

At the opening session of the Conference, Dr. J. A. Prins (Groningen) gave an account of the results obtained at the University of Groningen on the absorption and emission of X-rays by metals. The absorption spectrum in crystalline solids has a fine structure extending several hundred electron volts on the short wave-length side of the edge, and depending in its general features only on the structure, and not on the atomic constitution of the crystal. A theoretical explanation due to Kronig was discussed, which relates the phenomenon to the Bragg reflection of the ejected electrons. Dr. H. W. B. Skinner (Bristol) showed how the study of emission bands in the ultra-soft region can give information about the occupied electronic states in metals. Prof. W. L. Bragg (Manchester) gave a report of some recent results obtained by Sykes on the formation of a superstructure in copper-gold alloys, and Mr. J. D. Bernal (Cambridge) discussed the factors which determine the crystal structure of alloys, pointing out especially that the atomic radii of the constituents have often a decisive influence.

The morning of July 3 was devoted to a discussion of the liquid and amorphous states. Prof. H. Mark (Vienna) gave an account of recent theories of electrical conductivity, and Dr. J. A. Prins gave a report on his own work on amorphous antimony. Dr. G. W. Brindley (Leeds) showed the effect of filing on the intensity of X-ray reflection from metals. In the afternoon there was a discussion on the properties of metallic bismuth, in which Prof. A. Goetz (Pasadena), Dr. N. Thompson (Bristol), Dr. D. Shoenberg (Cambridge) and Dr. H. Jones (Bristol) took part. Bismuth is peculiar in that very small quantities (less than 0·1 per cent) of lead, tin or other metals in solid solution have a very marked effect on its electrical conductivity and diamagnetic susceptibility. Prof. Goetz expressed the opinion that these facts suggest the existence in bismuth of a microcrystalline superstructure of the kind postulated by Zwicky, but an alternative explanation was suggested in terms of the wave mechanical theories due to Peierls and Jones. These theories, however, will require further development before any quantitative account of the facts can be given.

On July 4, Prof. W. Gerlach (Munich) gave an account of the work carried out in his laboratory on the electrical resistance of nickel and of some of its alloys. The resistance of nickel decreases in a magnetic field, the decrease being proportional to the change in σ^2, where σ is the magnetisation. Further, the resistance plotted against temperature shows a kink at the Curie point. A possible explanation of the effect was suggested by Prof. N. F. Mott (Bristol). Dr. E. C. Stoner (Leeds) pointed out that various methods of determining the Weiss intramolecular field constant in nickel give very discordant results, and suggested an explanation.

On July 5, a discussion on supra-conductivity was opened by Dr. J. M. Casimir (Leyden), who gave an account of the fundamental experiments of Meissner, and of de Haas and Mrs. Casimir, and of the important theoretical advances of Gorter, Casimir and London. Mr. K. Mendelssohn (Oxford) gave an account of the behaviour of certain alloys. It appears that though great progress has been made towards the understanding of the thermodynamics and electrodynamics of a supra-conductor, we are still as far as ever from knowing the cause of the phenomenon. Dr. H. Niewodniczański and Mr. C. J. Milner (Mond Laboratory, Cambridge) discussed the electrical conductivity of non-supra-conductors, and here also the results of theory are by no means in accord with experiment, and the opinion was expressed on several sides that all theories are wrong below 4° K. Prof. F. Simon (Oxford) and Dr. E. Teller (London) also discussed the possibility of using nuclear spins to obtain yet lower temperatures than can be obtained with ordinary paramagnetics by the magnetic cooling method.

An interesting feature of the Conference was the frequent mention of experimental facts for which no quantitative theoretical explanation could be given. Considerable advances have recently been made in the quantum theory of the metallic state, and it is now possible for the theorist to give an indication of why certain metals have their particular crystal structures, magnetic properties and electrical conductivities. Nevertheless, the theory is as yet far from being able to give such a complete account of the experimental data as is possible, for example, in the field of atomic spectra.

Nature **136**, 115 (1935)

Schrödinger's Letter of Letter of Support for Fröhlich

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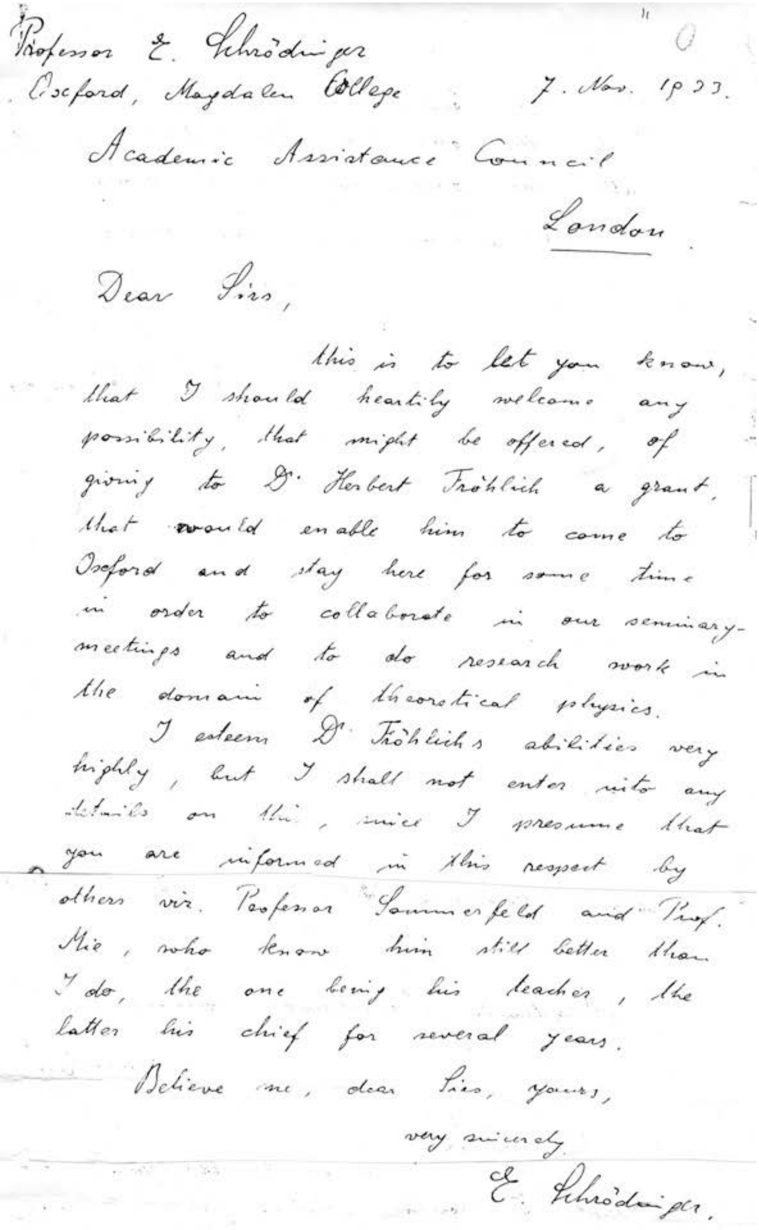

Professor E. Schrödinger
Oxford, Magdalen College
7. Nov. 1933.

Academic Assistance Council
London.

Dear Sirs,

this is to let you know, that I should heartily welcome any possibility, that might be offered, of giving to Dr. Herbert Fröhlich a grant, that would enable him to come to Oxford and stay here for some time in order to collaborate in our seminary-meetings and to do research work in the domain of theoretical physics.

I esteem Dr. Fröhlich's abilities very highly, but I shall not enter into any details on this, since I presume that you are informed in this respect by others viz. Professor Sommerfeld and Prof. Mie, who know him still better than I do, the one being his teacher, the latter his chief for several years.

Believe me, dear Sirs, yours,
very sincerely
E. Schrödinger.

Photo Gallery

The number in round brackets at the end of the caption corresponds to the section of the text to which the photograph refers.

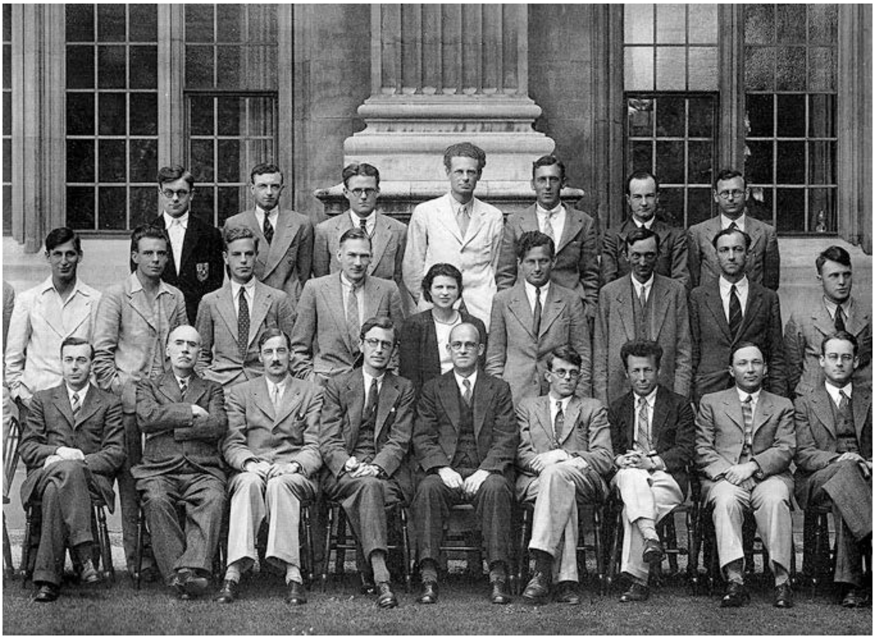

Fig. 3.3 The Bristol Physics Department in the mid 1930s. *Front row* Heitler (3rd from *RH* end), Tyndall (*centre*), Mott (4th from the *LH* end). Fröhlich is 4th from *RH* end of the 2nd row—Reproduced with the permission of the Department of Physics, Bristol (Sect. 3.2)

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Chapter 4
Life in Bristol

4.1 From Metals to Dielectrics: First Theory of Dielectric Breakdown

Once in Bristol, Fröhlich finished his book on the electron theory of metals (*Elekronentheorie der Metalle* [F(*i*)]) in September 1936,[1] having signed the contract already in 1934. It was published the following month by Springer, who had by then moved to Vienna, as Volume XVIII of their Monograph Series 'Structure & Properties of Matter': the Vienna Editor was H. Mark (vide Chap. 3). Although unable to arrange any royalties, Springer did pay him a generous advance amounting to a few thousand marks (Fig. 4.1).

1936 saw the publication of two books on similar subjects, namely A.H. Wilson's *Theory of Metals* (Wilson 1936), and *Theory of metals and alloys* by N.F. Mott & H. Jones, the latter two authors being in the Bristol Physics Department. Fröhlich's book, the title of which was the same as that of a substantial treatise by Sommerfeld and Bethe, which had appeared in *Handbuch der Physik* three years earlier in 1933, was distinguished by its lucidity, the inclusion of many helpful diagrams, and a full discussion of the physics behind any mathematics—a feature that characterised all his writings.

Despite its title, the book—which comprised seven chapters and an Appendix—dealt with a wide range of topics pertaining not only to the metals but also to *semiconductors*, as already mentioned in Chap. 3. The first three chapters included a general discussion of electrons in a periodic potential (following the work of Bloch in 1928), the thermodynamic and optical consequences of applying Fermi statistics to metallic electrons, and a treatment of conductivity, both electrical and thermal, together with a discussion of thermoelectric and galvanomagnetic effects.

The erratum of this chapter can be found under DOI 10.1007/978-3-319-14851-9_8

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[1] In the *Foreword* to his book, Fröhlich acknowledges the help of K. Fuchs and H. London, who were then fellow refugees in Bristol, in correcting the proofs.

G.J. Hyland, *Herbert Fröhlich*,
Springer Biographies, DOI 10.1007/978-3-319-14851-9_4

Fig. 4.1 Fröhlich in Bristol, early 1940s

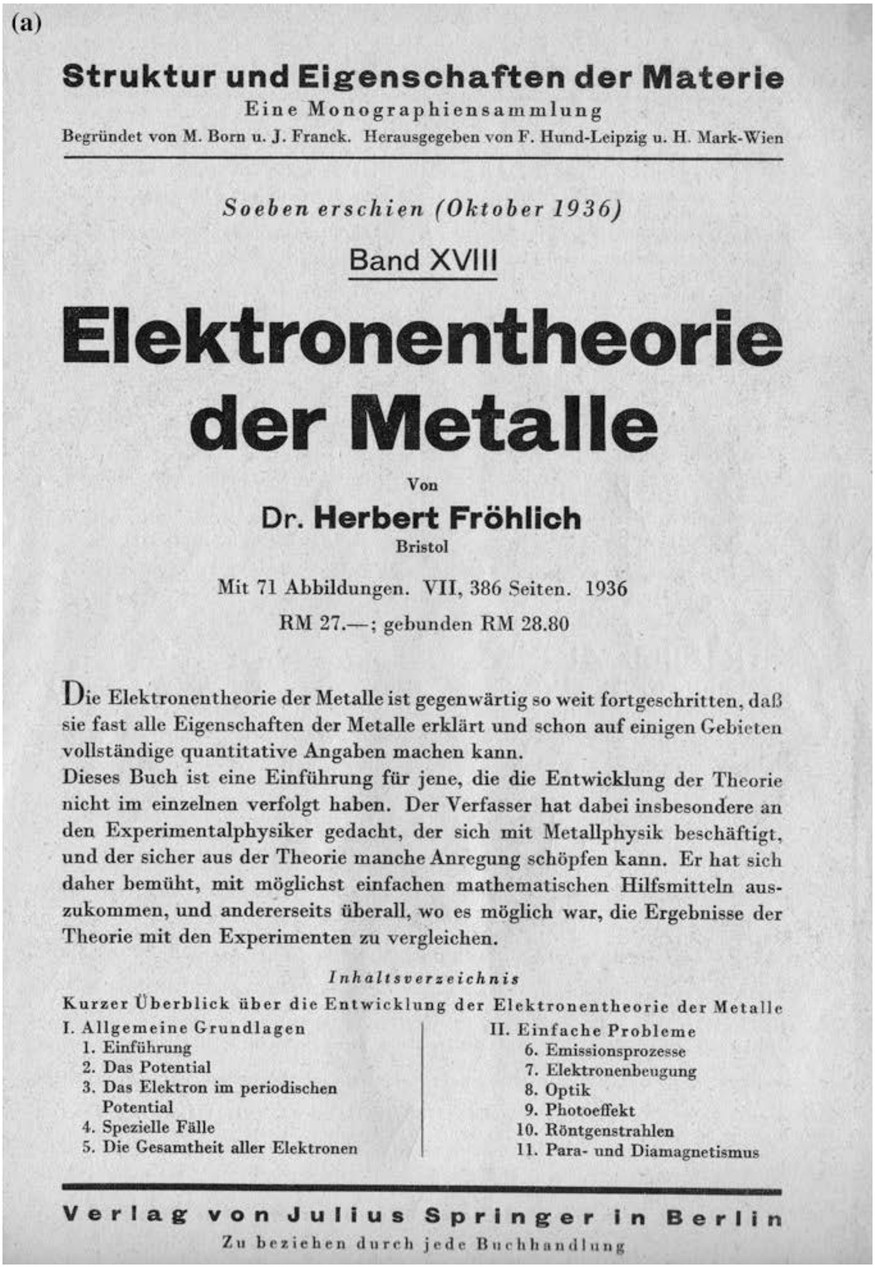
The final three chapters were devoted to metallic binding, ferromagnetism, and to a systematic discussion, in Chap. 7, of the different types of metals—monovalent, divalent and transition—and referred to the then recent work of H. Jones on bismuth. Indeed, it was from a diagram on page 320 of that chapter, showing that a diagonal through the Periodic Table of the Elements separates metals from insulators, that Lark-Horowitz was later led to adopt germanium as the 'best' semiconductor. Semiconductors, per se, was the subject of Chap. 4, and it was principally because of this that, during the Second World War when the technological potential of these materials was starting to be realized, the book was reproduced (untranslated) as a photo-lithoprint in the *USA*[2]; for at that time it was the *only* textbook to contain a comprehensive treatment of semiconductors, covering such topics as electrical conductivity, the thermoelectric and Hall effects, rectification, and optical properties (Fig. 4.2).

So useful was the book considered to be in Germany after the war, that it there became known as 'the Bible'. Indeed, a badly charred copy—a victim of an Allied air raid on Frankfurt (am Main)—was copied out by hand in its entirety by one physicist who, in common with many others, appreciated its great value, particularly in the immediate post-war years when it was unobtainable. Many years later in the 1970s, this physicist showed Fröhlich his tattered manuscript copy, proudly telling him that he had written it all out by hand, only to receive the reply: 'I wrote it all by hand, too!' The physicist was Ludwig Genzel (1922–2003), Professor of Physics at Frankfurt and Freiburg, and later Director (1970–1990) of the *Max Planck Institute for Solid-state Research* in Stuttgart, which he helped to found in 1970 at the request of Heisenberg.

Fröhlich's first publications [F12, F13] from Bristol in 1936 arose from a collaboration with Walter Heitler, continuing the latter's earlier work with Teller on the magnetic cooling method, which exploited the fact that the magnetic moments of a paramagnetic salt can be orientated isothermally by an external magnetic field. In [F13], they showed that the dominant contribution to the thermal conductivity, $\lambda(T)$, of a paramagnetic salt comes, not from lattice vibrations but rather, from the *spins* of the paramagnetic ions. This contribution arises when, in the presence of a temperature difference between two points of the crystal, excited ions travel down the temperature gradient to the cooler region where they deposit their excitation

---

[2] Fröhlich learned about this 'pirate' reprint, and how well it was selling in the *USA*, when, during a visit to Bristol just after the war, Seitz suggested that he should attempt to claim royalties, which Fröhlich duly did. After many unsuccessful attempts at writing to the office of the Aliens' Property Custodian in Washington, he took the opportunity whilst in Washington in 1951 to speak on his new theory of superconductivity at the *NBS* Semicentennial Symposium on Low Temperature Physics [F81], to visit the office in person. The reason for the delay soon became apparent when the official to whom he was directed, after complaining about the complexity of Springer contracts, received a telephone call, which Fröhlich overheard, which revealed that he was still prevaricating over the rights to the wartime song *Lili Marlene*! Clearly, the strategy was to delay things as long as possible to avoid paying out. Eventually this official was replaced, and Fröhlich received an apology for the delay; it remains uncertain, however, whether he ever received any royalties. The book was later republished, untranslated, by Springer in 1969.



(a)

Struktur und Eigenschaften der Materie
Eine Monographiensammlung
Begründet von M. Born u. J. Franck. Herausgegeben von F. Hund-Leipzig u. H. Mark-Wien

Soeben erschien (Oktober 1936)

Band XVIII

Elektronentheorie der Metalle

Von
Dr. Herbert Fröhlich
Bristol

Mit 71 Abbildungen. VII, 386 Seiten. 1936
RM 27.—; gebunden RM 28.80

Die Elektronentheorie der Metalle ist gegenwärtig so weit fortgeschritten, daß sie fast alle Eigenschaften der Metalle erklärt und schon auf einigen Gebieten vollständige quantitative Angaben machen kann.
Dieses Buch ist eine Einführung für jene, die die Entwicklung der Theorie nicht im einzelnen verfolgt haben. Der Verfasser hat dabei insbesondere an den Experimentalphysiker gedacht, der sich mit Metallphysik beschäftigt, und der sicher aus der Theorie manche Anregung schöpfen kann. Er hat sich daher bemüht, mit möglichst einfachen mathematischen Hilfsmitteln auszukommen, und andererseits überall, wo es möglich war, die Ergebnisse der Theorie mit den Experimenten zu vergleichen.

Inhaltsverzeichnis

Verlag von Julius Springer in Berlin
Zu beziehen durch jede Buchhandlung

Fig. 4.2 **a** Springer's advertisement of Fröhlich's first book, *Elektronentheorie der Metalle* [F(i)]: **b** the advert continued—Reproduced with the permission of Springer-Verlag

(b)

Fröhlich, Elektronentheorie der Metalle

III. Leitfähigkeit
  12. Elementare Theorie
  13. Die Gitterschwingungen und ihre Wechselwirkung mit den Elektronen
  14. Elektrische Leitfähigkeit
  15. Wärmeleitfähigkeit
  16. Thermoelektrische Effekte
  17. Galvano-magnetische Effekte
IV. Halbleiter
  18. Allgemeines
  19. Leitfähigkeitsprobleme
  20. Optische Probleme
V. Die metallische Bindung
  21. Einführung
  22. Das Metallmodell der freien Elektronen
  23. Quantitatives
VI. Ferromagnetismus (und Paramagnetismus II)
  24. Austauschkräfte und Wechselwirkung freier Elektronen
  25. Der ferromagnetische Zustand
  26. Die Magnetisierungskurve
  27. Paramagnetismus II
VII. Systematische Diskussion der Metalle
  28. Überblick
  29. Elemente mit abgeschlossenen inneren Schalen
  30. Elemente mit nichtabgeschlossenen inneren Schalen (Übergangselemente)
  31. Wismut
  32. Flüssige Metalle
Anhang
  1. Schrödinger-Gleichung mit Vektorpotential
  2. Beweis des Summensatzes
  3. Integrale zur Fermi-Statistik
  4. Bose-Einstein-Statistik
  5. Virialsatz
  6. Elektronen in nichtkubischen Kristallen
  7. Gitterpotential
  8. Legierungen mit γ-Struktur
Literaturverzeichnis
Sachverzeichnis

Ich bestelle hiermit:

.... Expl. **Fröhlich, Elektronentheorie der Metalle**
(Struktur und Eigenschaften der Materie, Band XVIII.) 1936.
RM 27.—; gebunden RM 28.80

Ferner: ....

....

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(Verlag von Julius Springer / Berlin)

Name (möglichst Stempel): ....

Genaue Adresse: ....

.... *Datum:* ....

19. 36. 270.

Fig. 4.2 (continued)

energy,[3] the mean free path being governed by magnetic interactions. It was found that λ is roughly proportional to T^{-2} (if $kT > W$), or $T^{-3/2}$ (if $kT < W$), where W is the spin-spin interaction energy. At 0.06 K, the calculated value in the case of ammonium alum was $\lambda \approx 3$ erg/cm s deg, but experiments that had been carried out up to that time did not allow a conclusive check on these predictions, since they had been conducted over such a long time that interaction between the spins and the lattice vibrations could not be neglected; accordingly, the experimental value of λ most likely contained a contribution from the lattice vibrations, which would account for the much larger measured value of 240 erg/cm s deg (Kürti and Simon—personal communication to Fröhlich and Heitler 1936). A third paper [F14], published the following year in collaboration with Heitler and Teller, dealt with time effects in the magnetic cooling method, and pointed out that, in the case of a metal, the nuclear spins can transfer their energy to the conduction electrons, the time required being short enough to make possible an application of the method.

Fröhlich next developed a method [F15] that allowed, for the first time, quantitative *analytical* calculations of the binding energy of alkali metals, using the ionisation potential as the only empirical parameter; associated calculations of the lattice constant, heat of sublimation, compressibility and thermal expansion coefficient were all found to be in satisfactory agreement with experimental values.

He then turned his attention from metals to dielectrics, a class of electrical insulators, addressing first the technologically important problem of dielectric breakdown—i.e. to understand the reason these materials cease to be insulating in high external electric fields. Of particular interest was the so-called *intrinsic* breakdown observed in many dielectrics, such as glass, mica, most polymers and, in particular, alkali halides—a class of ionic (polar) materials for which, through the work of von Hippel, the experimental situation was most clearly defined, specifically: (a) the breakdown field is almost the same for different specimens of the same material, and does not depend on their source or method of preparation, (b) breakdown occurs very rapidly on a time scale of the order of 10^{-8} s, which is very much less than that characterising thermal breakdown, which is very much slower, in the order of seconds. Research on alkali halides strongly implicated the involvement of electrons in the breakdown process. Particularly important was the finding that in the case of *mixed* crystals, such as Na–Ag chloride, the intrinsic dielectric strength (i.e. the magnitude of the external electric field above which breakdown occurs) is always higher than that of their components (von Hippel and Lee 1941)—a feature that breakdown apparently had in common with the electrical resistivity of a mixture of metals, where it was known to be due to a decrease of the

---

[3] The similarity with ambipolar conduction in semiconductors (where it is the *electron-hole* creation energy that is transported down a temperature gradient) should be noted; indeed, this situation was later actually considered in a joint paper [F91] with Kittel in 1954, in which they acknowledged the independent, contemporaneous work of Price (1954, 1955). (The earliest reference dealing with ambipolar heat conduction would appear to be that of Davydov and Shmushkevitch 1940). Ambipolar heat conduction later proved to be of importance in Mott-insulating materials, such as the technologically important oxide UO_2 (Hyland 1983).

electron mean-free-path, or, equivalently, their relaxation time, τ. Strictly speaking, the concept of a relaxation time requires that the scattering of the electrons be elastic. τ, which is a function of the electron's kinetic energy (velocity), is not simply the average time between two collisions, but contains also the scattering angle, and is better regarded as the average time between two large angle scatterings. Interpreting the observed increase in dielectric strength of mixed crystals similarly, requires, firstly, the presence of electrons in the conduction band of the material, and secondly that these electrons have a sufficiently high energy, E, that their collisions with the lattice vibrations can be considered to be elastic—i.e. $E \gg \hbar\omega$, where $\hbar\omega$ is a quantum of the relevant lattice vibration (vide infra): this condition is well-satisfied for values of E in the order of eV—so that a relaxation time can again be defined. In the absence of an external electric field, there are, in an ideal (defect free) crystalline insulator at the temperatures of interest, a negligible number of electrons in the conduction band because of the large band gap, I_o ($\sim$ several eV); this no longer holds, however, in the presence of a strong (but sub-breakdown) electric field ($\sim 10^5$ V/cm), as first noted by Zener in an ill-fated[4] early theoretical attempt to understand dielectric breakdown, written during the tenure of a research fellowship in Bristol during 1932–1934 (Zener 1934).

Von Hippel's general idea was that under the influence of an external electric field, breakdown occurs when the field is high enough to accelerate conduction electrons of *any* given energy to the energy I_o when they can ionize the negative ions of the lattice by collision, thereby producing an additional conduction electrons, the process building up in the form of an avalanche. This increase in the number of electrons undermines the insulation of the material, resulting in dielectric breakdown on a rapid time scale. This was the starting point of Fröhlich's development of his own theory of dielectric breakdown, which he later showed was applicable, with appropriate modifications, to a range of materials, not solely the ionic ones considered initially [F16]. At sub-breakdown fields, the existence of a steady current indicates that a conduction electron can, by some means, get rid of the energy it receives from the field. In common with von Hippel, Fröhlich assumed that the operative mechanism was via energy transfer to lattice vibrations. His theory differed from that of von Hippel, however, in that he considered that breakdown occurs when the applied field is just strong enough to be able to accelerate to the energy I_o just those electrons that *already* have an energy that is only slightly below I_o—i.e. fast electrons; his breakdown field is thus *lower* than

---

[4] Zener's idea was that dielectric breakdown was due to a dramatic increase in the number of electrons in the conduction band arising from the fact that in the presence of an electric field, by a process akin to the auto-ionization of free atoms, electrons from the valence band can find their way into the conduction band; this possibility arises in consequence of the fact that in an electric field the usual energy band scheme becomes *tilted* so that states in the (full) valence acquire the *same* energy as those in the (empty) conduction band, thereby permitting tunnelling to occur, the more efficiently, the higher the field. Whilst the number of electrons so produced certainly increases rapidly with increasing field strength, it cannot account for the almost *discontinuous* increase that characterises dielectric breakdown; it can nevertheless account for the presence in the conduction band of electrons that would not be there in the absence of a field.

that predicted on the basis of von Hippel's considerations, which involved acceleration of conduction electrons of *all* energies.

More formally, the breakdown field was defined as that field above which a conduction electron gains more energy from the external electric field than it can transfer to the lattice vibrations, so that a stationary state characterised by a steady current cannot exist.

The energy, A, transferred per second per unit volume from the electric field, F, to an electron is given by:

$$A = JF \tag{4.1.1}$$

where J the current density is given, in terms of the relaxation time τ, and the electric charge, e, by:

$$J = e^2\tau(E)F/m \tag{4.1.2}$$

In order that a steady current may flow, it is necessary that the energy acquisition rate, A, be exactly balanced by the rate of energy transfer, B, to the lattice vibrations. Both A (via τ) and B depend on the electron's energy, E. For the case of an ionic lattice, it can be shown that, provided[5] $E < I_o$, $A \sim E^{3/2}$, and $B \sim E^{-1/2}$. There is thus always an energy E^* such that $A = B$, with $E^* \sim 1/F$. At low energies ($E < E^*$), the electron loses energy, whilst at high energies ($E > E^*$), it gains more energy than it can transfer to the lattice. Provided, however, $E^* > I_0$—i.e. for weak fields—equilibrium is possible, whilst for $E^* < I_0$ (in strong fields) it is not. Thus the critical field above which it is no longer possible to maintain a steady current is determined by $E^* = I_o$, the associated critical (breakdown field) being $F^* \sim 1/I_o$. Thus, the field strength at which breakdown begins has[6] to be calculated from the condition

$$A(E,F) = B(E), \quad \text{with} \quad E = I_o, \tag{4.1.3}$$

or equivalently,

$$F^* = \left[mB(E)/e^2\tau(E)\right]^{1/2}_{E=I_o} \tag{4.1.4}$$

---

[5] Electrons with energy $E > I_0$, interact not only with the lattice vibrations, but also undergo inelastic collisions with the ions of the lattice causing ionization, the associated mean-free path being of the order of the lattice spacing—i.e. about 100 times shorter than that associated with scattering by lattice vibrations. Accordingly, the latter can be neglected, whilst the former do not result in any overall increase in electron density because they are balanced by the inverse process.

[6] It was not possible to give a more precise criterion for calculating the breakdown field.

The breakdown field so calculated is the field strength that is, on average, *first* able to accelerate an electron with an energy *just below* I_o up to the value I_o, when it can produce an abrupt increase in the number of conduction electrons by ionization. The evaluation of the breakdown field F^* is thus reduced to a calculation of $\tau(E)$ and $B(E)$.

Introducing the electronic mean-free-path ℓ, through $\ell = v\tau$, where v is the average velocity of an electron, it follows that

$$F^* \sim \ell^{-1/2} \tag{4.1.5}$$

It follows immediately that F^* can be increased by strategies that reduce the mean-free-path, such as increasing the temperature (which increases the degree of excitation of the lattice, and hence the likelihood of the electron being scattered), introducing foreign ions into the lattice, and by using thin films of dielectric material whose thickness is smaller than the mean-free path of bulk material (of the order of 10^{-5}–10^{-6} cm), in which case the surfaces themselves give rise to scattering. The predicted increase in F^* with decreasing thickness was experimentally confirmed two years later in mica, where reducing the thickness from 10^{-4} to 10^{-5} cm was found to increase F^* from 11×10^6 to 16×10^6 V/cm (Austen and Hackett 1939).

Fröhlich drew attention[7] to these strategies in his first paper on dielectric breakdown in diatomic polar materials, which was published in May 1937 in the *Proceedings of the Royal Society* [F16] (Fig. 4.3).

In the case of alkali halides, the calculation of τ(E) and $B(E)$ was greatly facilitated by the fact that, in the case of such diatomic polar lattices, each elastic vibration is connected with a corresponding electrical polarization *field*, $\boldsymbol{P}$. The conduction electrons interact most strongly with vibrations belonging to the optical branch in which the positive and negative ions vibrate against one another in antiphase giving rise to a large electric dipole moment; the associated frequency is in the infra-red region. The energy of interaction, W, is given by

$$W = e\Sigma_{\mathrm{w}}\phi_{\mathrm{w}} \tag{4.1.6}$$

where ϕ_{w} is the potential of a single polarization wave of wave number w at the position of an electron of charge e, and is determined from

$$\nabla^2\phi_{\mathrm{w}} = 4\pi \mathrm{div}\, \boldsymbol{P}_{\mathrm{w}} \tag{4.1.7}$$

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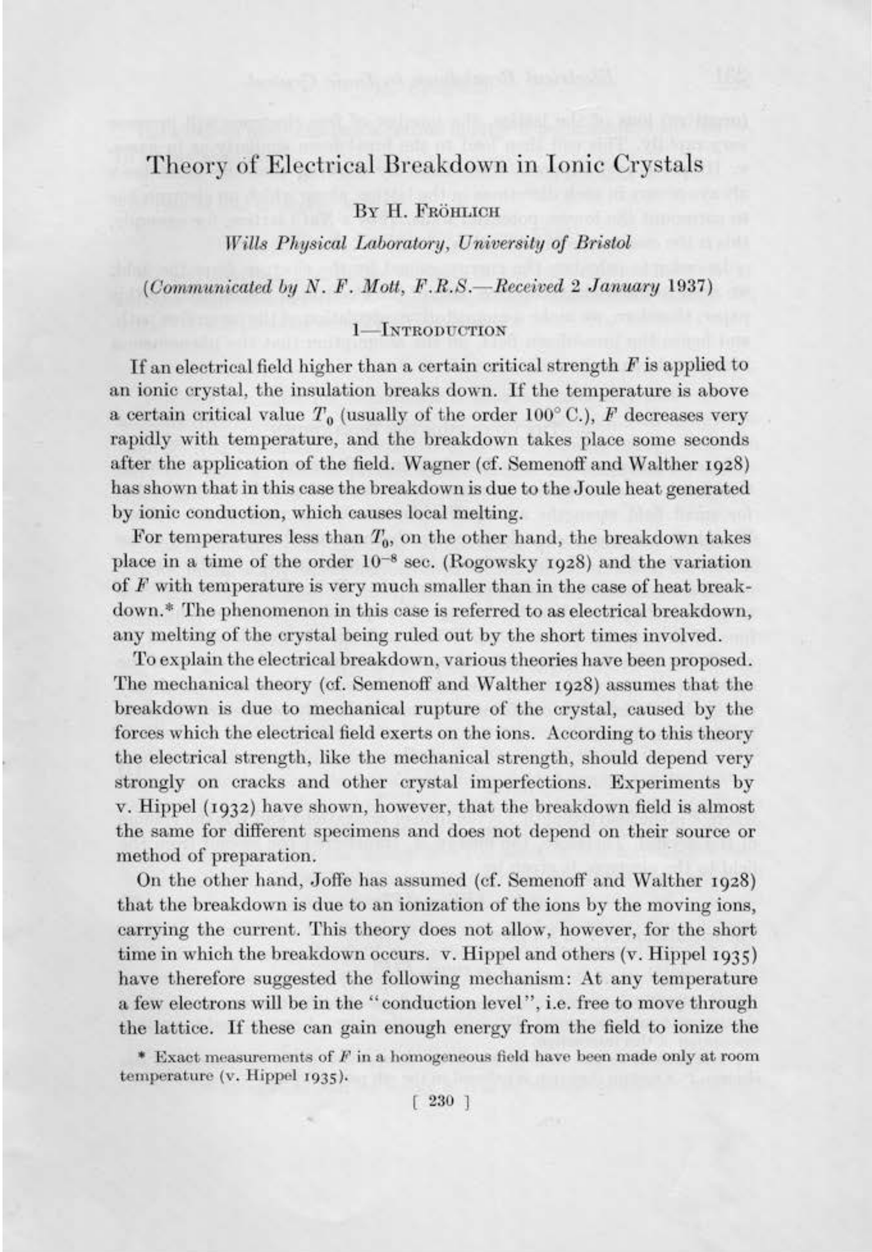
[7] The latter two strategies were elaborated in greater detail three years later in 1940, in [F34], upon his return to Bristol in 1940 after internment.



Theory of Electrical Breakdown in Ionic Crystals

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1—Introduction

If an electrical field higher than a certain critical strength F is applied to an ionic crystal, the insulation breaks down. If the temperature is above a certain critical value T_0 (usually of the order 100° C.), F decreases very rapidly with temperature, and the breakdown takes place some seconds after the application of the field. Wagner (cf. Semenoff and Walther 1928) has shown that in this case the breakdown is due to the Joule heat generated by ionic conduction, which causes local melting.

For temperatures less than T_0, on the other hand, the breakdown takes place in a time of the order 10^{-8} sec. (Rogowsky 1928) and the variation of F with temperature is very much smaller than in the case of heat breakdown.* The phenomenon in this case is referred to as electrical breakdown, any melting of the crystal being ruled out by the short times involved.

To explain the electrical breakdown, various theories have been proposed. The mechanical theory (cf. Semenoff and Walther 1928) assumes that the breakdown is due to mechanical rupture of the crystal, caused by the forces which the electrical field exerts on the ions. According to this theory the electrical strength, like the mechanical strength, should depend very strongly on cracks and other crystal imperfections. Experiments by v. Hippel (1932) have shown, however, that the breakdown field is almost the same for different specimens and does not depend on their source or method of preparation.

On the other hand, Joffe has assumed (cf. Semenoff and Walther 1928) that the breakdown is due to an ionization of the ions by the moving ions, carrying the current. This theory does not allow, however, for the short time in which the breakdown occurs. v. Hippel and others (v. Hippel 1935) have therefore suggested the following mechanism: At any temperature a few electrons will be in the "conduction level", i.e. free to move through the lattice. If these can gain enough energy from the field to ionize the

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* Exact measurements of F in a homogeneous field have been made only at room temperature (v. Hippel 1935).

[230]

Fig. 4.3 Fröhlich's first paper on dielectric breakdown [F16], 1937—Reproduced with the permission of the Royal Society where $\boldsymbol{P}_{\mathrm{w}}$ is the associated polarization per unit volume; it follows that only *longitudinal* waves interact with the electron. Solving this equation yields an expression for the electron-lattice Hamiltonian, H_{int}, which formed the basis not only of Fröhlich's theory of dielectric breakdown, but also a numerous other analyses of electronic processes in ionic crystals, not least the so-called 'polaron problem'—vide Sect. 5.3.

$$H_{\mathrm{int}} = -\frac{4\pi i e^2}{2a^3\sqrt{2N}}\Sigma_{\mathrm{w}} w^{-1}\left[b_{\mathrm{w}}\mathrm{e}^{\mathbf{iw.r}} - b^*_{\mathrm{w}}\mathrm{e}^{-\mathbf{iw.r}}\right], \tag{4.1.8}$$

where

$$P_{\mathrm{w}} = \frac{e}{2a^3\sqrt{2N}}\left[b_{\mathrm{w}}\mathrm{e}^{\mathbf{iw.r}} + b^*_{\mathrm{w}}\mathrm{e}^{-\mathbf{iw.r}}\right], \tag{4.1.9}$$

$2a^3$ is the volume of the unit cell (i.e. a is distance between neighbouring ions), and $2N$ the total number of ions.

In the case of the fast electrons that Fröhlich identified as relevant to breakdown, the dominant scattering is by polarization waves of *short* wavelength, since it is these that have a momentum closest to that of electrons with $E \sim$ eV; the associated frequency, ω, can thus be approximated by that of the *Restrahlen*—i.e. the frequency of the transverse optical vibrations, ω_{T}, with which light interacts.

Detailed calculation using time-dependent perturbation theory to calculate the transition rates connected with the electron-lattice interaction yielded:

$$\tau = \tau_{\mathrm{o}}[1 + 2/(e^{\hbar\omega_{\mathrm{T}}/kT} - 1)]^{-1} = \tau_{\mathrm{o}} f(\mathrm{T}), \tag{4.1.10}$$

where $\tau_{\mathrm{o}} \sim E^{3/2}$ is the time of relaxation at $T = 0$ K, which is is entirely due to the zero-point oscillations of the lattice,[8] and $f(\mathrm{T})$ is a decreasing function of temperature. Thus, from Eqs. 4.1.1, 4.1.2, 4.1.10, $A \sim E^{3/2}$, as noted earlier, whilst the E-dependence of B is found to be:

$$B = const\ E^{-1/2} \tag{4.1.11}$$

Substituting these in Eq. 4.1.4 yields the following expression for the breakdown field, F^*, which, it should be noted, contains *no* arbitrary constants:

$$F^* = [const/I_{\mathrm{o}}] f(T)^{-1/2} \tag{4.1.12}$$

F^*, via $f(T)^{-1/2}$, is a weakly increasing function of temperature, which for $kT \gg \hbar\omega_{\mathrm{T}}$ becomes proportional to $T^{1/2}$; for orientation, it may be noted that for NaCl, for example, $\hbar\omega_{\mathrm{T}}$ is 0.018 eV, or 218 K.

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[8] This contrasts with the case of a metal at $T = 0$ K where, owing to the Pauli principle, an electron cannot loose energy, so that there are no collisions of the conduction electrons with the zero-point oscillations of the lattice.

Thus, the increase in F^* with temperature, which was predicted qualitatively by Eqs. 4.1.4, 4.1.5 is confirmed by microscopic calculation.

At the time, however, there was no experimental data against which the predicted increase in F^* with increasing temperature could be checked, there being data for a variety of alkali halides only at room temperature. The calculated and observed values of F^* were found to agree within an order of magnitude, being of the order of 10^5 V/cm, but were systematically below the experimental ones, consistent with the severity of Fröhlich's criterion for breakdown, which involved *only* those electrons whose energy is *just below* the ionization threshold, I_o, and which thus gives a *lower* bound to the value of F^*.

4.2 Sojourn in Holland

Shortly before the termination of his grant from the University of Bristol, Fröhlich moved, at the beginning of 1937, to Leiden, where, for about 6 months, he was supported by a grant from the Lorentz Foundation. Here, he worked in Kramers'[9] group, publishing 2 papers. The first [F17] addressed the topic he had earlier discussed with Mark during their meeting in Vienna in 1935, namely, the dependence of the electrical properties of metals on crystal size (vide Chap. 3). Using the electronic specific heat as an example, he noted that, in contrast to the case of an infinite system in the thermodynamic limit where the single electron quantum states form a continuum, these states must become quantized in the case of a finitely extended system—the average inter-level separation being of the order of the ratio of the Fermi energy to the number of conduction electrons. At sufficiently low temperatures in a finite system, the inter-level separation exceeds kT, so that the usual form of the Fermi distribution no longer holds, whence thermodynamic properties *different* from those found in the (bulk) thermodynamic limit must be anticipated. Assuming a constant level spacing, Fröhlich investigated the special case of small cubic crystals all of the same size, finding that the constant volume specific heat decreased exponentially at sufficiently low temperatures, in contrast to the familiar linear decrease that characterises bulk systems (Fig. 4.4).

In this, Fröhlich was again ahead of his time, anticipating the subsequent bourgeoning of interest in mesoscopic systems and nanotechnology. It would be another 25 years before the problem was next considered in any detail and in greater generality, taking into account, for example, a distribution of crystal sizes (Kubo 1962).[10]

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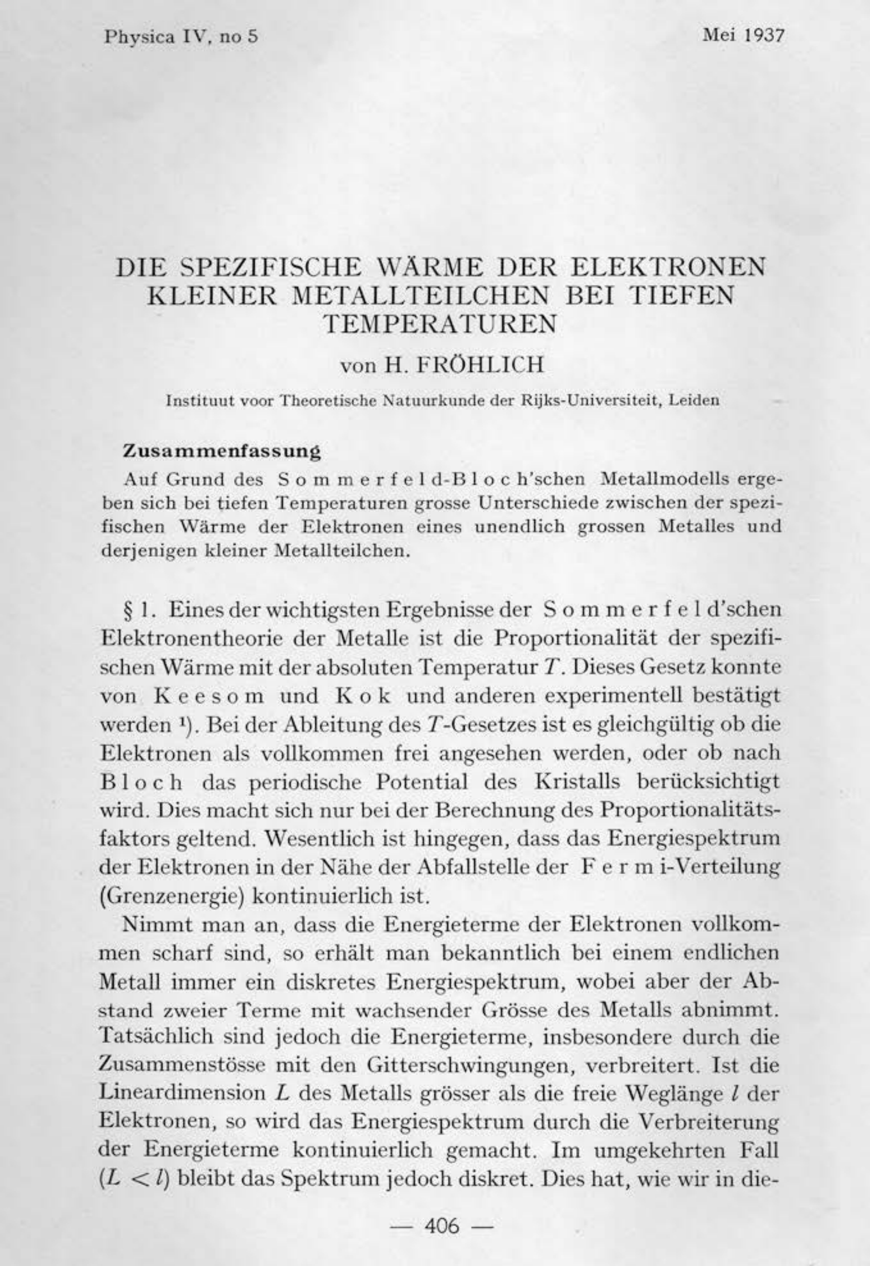
[9] From his earlier work on magnetic cooling [F13], Fröhlich was already familiar with Kramers' work (Kramers 1930) showing that, in an inhomogeneous crystalline electric field, states of a paramagnetic ion of *even* multiplicity are still 2-fold degenerate (Kramers degeneracy).

[10] Mention should also be made of later theoretical work (Denton et al. 1971, 1973), which showed that the effect of admitting a distribution in crystal size is to soften Fröhlich's exponential temperature dependence of the electronic specific heat to a *power* law, the precise form of which depends on the assumed level statistics; under certain conditions, the linear bulk temperature dependence can actually be retrieved.



Physica IV, no 5 Mei 1937

DIE SPEZIFISCHE WÄRME DER ELEKTRONEN KLEINER METALLTEILCHEN BEI TIEFEN TEMPERATUREN

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Zusammenfassung

Auf Grund des Sommerfeld-Bloch'schen Metallmodells ergeben sich bei tiefen Temperaturen grosse Unterschiede zwischen der spezifischen Wärme der Elektronen eines unendlich grossen Metalles und derjenigen kleiner Metallteilchen.

§ 1. Eines der wichtigsten Ergebnisse der Sommerfeld'schen Elektronentheorie der Metalle ist die Proportionalität der spezifischen Wärme mit der absoluten Temperatur T. Dieses Gesetz konnte von Keesom und Kok und anderen experimentell bestätigt werden [1]). Bei der Ableitung des T-Gesetzes ist es gleichgültig ob die Elektronen als vollkommen frei angesehen werden, oder ob nach Bloch das periodische Potential des Kristalls berücksichtigt wird. Dies macht sich nur bei der Berechnung des Proportionalitätsfaktors geltend. Wesentlich ist hingegen, dass das Energiespektrum der Elektronen in der Nähe der Abfallstelle der Fermi-Verteilung (Grenzenergie) kontinuierlich ist.

Nimmt man an, dass die Energieterme der Elektronen vollkommen scharf sind, so erhält man bekanntlich bei einem endlichen Metall immer ein diskretes Energiespektrum, wobei aber der Abstand zweier Terme mit wachsender Grösse des Metalls abnimmt. Tatsächlich sind jedoch die Energieterme, insbesondere durch die Zusammenstösse mit den Gitterschwingungen, verbreitert. Ist die Lineardimension L des Metalls grösser als die freie Weglänge l der Elektronen, so wird das Energiespektrum durch die Verbreiterung der Energieterme kontinuierlich gemacht. Im umgekehrten Fall ($L < l$) bleibt das Spektrum jedoch diskret. Dies hat, wie wir in die-

Fig. 4.4 Fröhlich's paper on the electronic specific heat of small metallic crystals [F17], 1937—Reproduced with the permission of Elsevier

The second paper [F18] was an unsuccessful attempt to understand the λ-transition of liquid helium (He^4) to the superfluid state as an order-disorder transition, based on a diamond lattice structure model of He^4 proposed by F. London the previous year (London 1936), in which only one half of the sites are occupied, thereby simulating a binary alloy. Despite the good agreement obtained with the experimental entropy of transition, the treatment was much later shown to be flawed on energetic grounds (London 1954, p. 35). Notwithstanding this, the model stimulated a certain amount of additional research by others (Keesom 1942). Particularly perceptive—in the light of much later developments in both liquid He^4 and superconductivity—was Fröhlich's remark that the form of the observed specific heat at low temperatures requires an *isolated* lowest level whose separation from higher levels must decrease with increasing temperature up to the λ-point.

In June 1937, towards the end of his stay in Leiden, Fröhlich received a letter from Mott in Bristol informing him that he had been approached[11] by the British Electrical & Allied Industries Research Association[12] (*ERA*) for theoretical help in understanding the technologically important subject of dielectric breakdown; the letter is reproduced in Appendix 1 at the end of this chapter. Mott mentioned that he had told the *ERA* about his [i.e. Fröhlich's] work, in which they had expressed great interest—so much so, that they would probably be prepared to make a grant of £275 for one year, starting on 1 October 1937, to enable him to employ an assistant, not only to extend what he had already done to *non*-polar materials, but also to address the discrepancy between some recent results that they had obtained (Thomas 1938) on the temperature dependence of breakdown and that predicted by Fröhlich's theory. Also of interest to them at this time were the effects of cracks and impurities.

4.3 Dielectrics Revisited, and the First ERA Reports

Fröhlich duly returned to Bristol to take up the *ERA* grant[13] himself, during which time he was also supported by a further grant from the *AAC*, as well as one from Mott's 'Special Fund'. In June 1838, he applied for a permanent visa, which was granted in the August. Upon his arrival in the *UK* in 1935, and again upon his return from Holland in July 1937, he had been initially permitted to stay for only one month; on both occasions, however, the permission was extended each 6 months at

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[11] The approach was probably made by Stanley Whitehead, a long-standing member of the *ERA* staff since 1925, who became its Director in 1946, a post he held until his death in 1956.

[12] The *British Electrical & Allied Industries Research Association* (*ERA*) is the second oldest Research Association in the *UK*; it became incorporated in 1920, when it took over the work of the *Electrical Research Committee* founded in 1914 as the result of an earlier initiative (1912) of the Institution of Electrical Engineers (*IEE*) to encourage research. The first Director of the *ERA*, from 1919 to 1944, was E.B. Wedmore, who was mentioned in the letter reproduced in Fig. 4.6a. The *ERA* was financed equally by government and industry.

[13] The amount Fröhlich actually received from the *ERA* turned out to be only £260, but it was later renewed for a further year, starting October 1938.

the request of the *AAC*.[14] He did not forget the support he had received from the *AAC*, and, in 1939, applied to become a member, and continued to support their work financially for many years, until at least 1967.

His first *ERA* Report [F25], dating from in 1939 (and subsequently published in the Proceedings of the Royal Society as [F26]) extended his earlier theory of 1937 for diatomic polar materials to *any* kind of polar crystal for which there is more than one *Restrahlen* frequency, such as is the case with the technologically important dielectrics quartz and mica, for example. Calculation of the breakdown field for these materials yielded values of the right order of magnitude, but somewhat lower than the experimental values the then available (Thomas 1936); the latter were, however, plagued with uncertainties, which made it difficult to reliably access the success of the extended theory.

Around the same time, Fröhlich, in collaboration with Mott [F23], used the same *field* description of lattice polarization as he had used in his theory of dielectric breakdown to treat what was essentially the *converse* problem—namely, the calculation of the mean-free-path of a *slow* electron with an average thermal energy of $(3/2)kT$ (~ 0.04 eV at room temperature) in the conduction band of an otherwise insulating ionic crystal, for which scattering by the lattice vibrations is much stronger than it is in the case of a fast electron, entailing a shortening of its mean-free-path. Following again his earlier work on dielectric breakdown, it was assumed that the electronic mean-free-path is determined by scattering by longitudinal polarization waves, but now (because the electron is here moving *slowly*) by those of *long* wavelength with frequency ω_L (rather than by those of short wavelength relevant to the case of dielectric breakdown, whose frequency is close to that (ω_T) of transverse waves, and which was approximated by the Restrahlen frequency). Because of the long wavelength involved, it was possible to treat the lattice as a dielectric *continuum,*[15] characterised by high and low frequency dielectric constants ε_∞ and ε_s, associated, respectively, with the electronic polarizability of the ions and their displacement, and by a single longitudinal optic frequency ω_L.

They first pointed out that for such polarization waves whose wavelength is long compared to the interionic distance, but short[16] compared to the size of a diatomic

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[14] Fröhlich did not apply for British naturalisation until November 1943, when he named Sir Edward Appleton, Professor Tyndall, *FRS* and a Dr. E.T. Paris as sponsors; it was not granted, however, until August 1946.

[15] Rather than as a discrete set of point charges, alternatively $+e$ and $-e$, with ionic masses M_+ and M_-, as was necessary in the case of the fast electrons.

[16] In the light of much later developments (vide Sect. 5.2), it should be noted that attention was drawn in passing to the opposite case of polar samples whose size is *smaller* than the wavelength, it being pointed out that the optical properties of a system of such small samples dispersed (with an inter-sample separation greater than the wavelength) in a non-polar medium should differ from those of a corresponding bulk polar sample whose dimensions exceed the wavelength; in particular, it was noted that the scattering of infrared rays should be maximum at a frequency *different* from that of the (bulk) *Restrahlen*. It was a further 10 years, however, before this topic was elaborated further, which Fröhlich did in his second book, '*Theory of Dielectrics*', first published in 1949 (vide Sect. 5.2); much later, it proved to be of great importance in biological systems (vide Sect. 6.3).

polar crystal (which is necessary in order that the distinction between longitudinal and transverse modes is meaningful), ω_{L} is *higher* than ω_{T} because of the accumulation of electric charge near the nodes of the longitudinal mode, finding that

$$\omega_{\mathrm{L}}^2 = (\varepsilon_s - \varepsilon_\infty + 1)\omega_{\mathrm{T}}^2 \tag{4.3.1}$$

This result partly anticipated what is now known as the 'Lyddane-Sachs-Teller' relation published 2 years later (Lyddane et al. 1941), namely:

$$\omega_{\mathrm{L}}^2 = (\varepsilon_s/\varepsilon_\infty)\omega_{\mathrm{T}}^2 \tag{4.3.2}$$

The scattering was treated by first order time-dependent perturbation theory, which revealed that for electron kinetic energies, $E < \hbar\omega_{\mathrm{L}}$, when the electron can only absorb lattice quanta $\hbar\omega_{\mathrm{L}}$, the relaxation time, τ, is given by:

$$\tau = const.[\exp(\hbar\omega_{\mathrm{L}}/kT) - 1], \qquad E < \hbar\omega_{\mathrm{L}}. \tag{4.3.3}$$

This expression for τ, which diverges as $T \longrightarrow 0$ K, should be compared with the *non*-divergent limit, τ_{o}, given by Eq. 4.1.10 (wherein ω_{L} is replaced by the *Reststrahlen* frequency, ω_{T}, appropriate to short waves), which holds for a fast electron (with energy $E > \hbar\omega_{\mathrm{T}}$) that can also *emit* a lattice quantum.

The mean-free-path, ℓ, associated with τ is given by $\ell = v\tau$. If the slow electrons have a Maxwellian distribution, the average mean-free-path, $\overline{\ell}$, is obtained by integrating over energy, which yields (where $k\Theta_{\mathrm{L}} \equiv \hbar\omega_{\mathrm{L}}$),

$$\overline{\ell} = const.\,(T/\Theta)^{1/2}[\exp(\Theta_{\mathrm{L}}/T) - 1], \quad T < \Theta, \tag{4.3.4}$$

the associated mobility, μ, being given by

$$\mu = (e/m)\overline{\ell}/\overline{v} \tag{4.3.5}$$

where $\overline{v} = (3kT/m)^{½}$, by equipartition.

With $\Theta_{\mathrm{L}} = 280$K, the above expression for the mean-free-path was found to describe the general T-dependence obtained from Hall and electrical conductivity data on cuprous oxide then available (dating from 1933), but the absolute values were about eight times too small. Significant improvement was later obtained, however, when polaron effects were taken into account [F72], which results in an increase in $\overline{\ell}$ by a factor[17] $\varepsilon_s\varepsilon_\infty/(\varepsilon_s - \varepsilon_\infty + 1)$. Even better agreement was obtained [F90] with subsequently acquired mobility data on alkali halides, such as NaCl at $T = 82$ and 200 K; for NaCl, $\Theta_{\mathrm{L}} = 340$ K, so that $T < \Theta_{\mathrm{L}}$, as required (Redfield 1953).

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[17] This factor arises from the fact that polaron theory is concerned only with the inertial polarization of the lattice—vide Sect. 5.3.

In view of the subsequent development of (large) polaron theory (vide Sect. 5.3), it is of interest to note that attention was drawn in a footnote of [F23] to the neglect of any possible effect of a screening of the electronic charge, such occurring only in a higher order of approximation.

During the first half of 1940, he continued his work on dielectric breakdown, addressing some of the topics that the *ERA* had identified as being of interest to them in their letter to Mott in 1937. First, he extended his original theory to cover the case of *non*-polar materials, finding [F31] that the effects of temperature, thickness and purity were qualitatively similar to the polar case. Calculation of the absolute value of the mean-free-path was here not possible, however, for a number of reasons: (a) restriction to a single lattice frequency is here not realistic, (b) the simplification afforded in the polar case of treating the interaction of the electron with the lattice in terms of a polarization field is here not available, and must, instead, be determined from the details of the interaction of a neutral atom with an electron. Notwithstanding these difficulties, he was able to calculate the energy dependence of A and B, which were found to be identical to the polar case treated originally. An approximate expression for the breakdown field was obtained, which for $I_o \sim 5$–10 eV, average phonon energies $\sim 10^{-2}$–10^{-1} eV, and a mean-free-path of the order of 10^{-6} cm, yielded $F^* \sim 10^6$ V/cm, in general agreement with experiment.

A second report [F32] of the same date arose in response to concerns expressed by the *ERA* about the validity of his original theory in the light of their experimental finding that in some organic solids F^* *decreased* with increasing temperature (Thomas 1938), which was *opposite* to his predicted increase [F16]. He pointed out, however, that the dielectric composition of the materials in question was very different from that to which his theory was applicable, and suggested to the *ERA* that they repeat their measurements of the temperature dependence of the breakdown field on more appropriate materials. This they did, and the results confirmed his predicted increase in F^* with increasing temperature (Austen and Hackett 1939; Austen and Whitehead 1940), although the observed T-dependence for KBr was found to be somewhat sigmoidal. The increase was confirmed independently (Buehl and von Hippel 1939) but only *below* a certain temperature, T_o ($\sim$320 K), above which F^* decreased (as in the case of the organic materials investigated by the *ERA*); subsequent work on NaCl confirmed this reversal in the T-dependence of F^* near 320 K (von Hippel and Lee 1941), which is far too low to be attributable to thermal breakdown. It would be another 6 years, however, before an attempt was made to understand this reversal, as will be narrated in Sect. 4.7.

Fröhlich concluded [F32] with some qualitative considerations that showed how the observed decrease of F^* with increasing temperature might be understood in terms of the structural composition of the organic materials concerned, which were long chain paraffins with inserted molecular polar groups. Whilst the presence of the polar groups act to increase the dielectric strength of the paraffin by increasing the electron scattering, he pointed out that if these groups were able to rotate this would reduce the potential hills presented by the ions to an electron, resulting in a decrease in F^* in the direction perpendicular to the chains of the host material, whilst parallel to the chains, F^* would be nearly independent of T. Thus provided this effect dominates the increased scattering associated with the presence of the polar groups, the breakdown field in polycrystalline material would decrease with increasing temperature, as observed.

In another report [F34] of the same year (1940), the influence of thickness and of impurities on dielectric strength was investigated in greater detail, following the publication of experimental results by the *ERA* in 1939, which confirmed the qualitative predictions of his first paper of 1937 (Austen and Hackett 1939). Both of these *ERA* reports were actually published during the period when Fröhlich was interned (vide Sect. 4.5), the actual work presumably having been done prior to this.

It should be appreciated that Fröhlich's theory of dielectric breakdown was not universally accepted, the main rivals being those[18] based on von Hippel's mechanism, which involved the acceleration of conduction electrons of *all* energies (as opposed to only those that already had energies just below I_o, as assumed by Fröhlich) up to the ionization threshold, I_o. A particularly illuminating presentation of this rival theory was that given by Callen, which, apart from the differences just referred to, followed closely Fröhlich's own notation and methodology (Callen 1949). Both sides fiercely defended their respective positions, sometimes in jointly authored debates, such as those between Fröhlich, Seeger & Teller (F24, Seeger and Teller 1939), and between Fröhlich and Seitz [F74].

The only other publication on condensed matter physics dating from this period, which was *not* devoted to dielectric topics, was [F33] in collaboration with Nabarro, which was submitted to the Proceedings of the Royal Society in February 1940. This considered the orientation of nuclear spins in a monovalent metal in connection with a proposal for extending the technique of magnetic cooling to lower temperatures by using the magnetism of atomic nuclei. The central idea was that the magnetic interaction between the nuclear spins and the spins of the conduction electrons[19] results in an indirect coupling between the nuclear spins, which can be *significantly larger* than their direct interaction. Using an analogy with the Weiss theory of ferromagnetism, they showed that this indirect interaction results in a second-order phase transition at a Curie temperature T_c below which the nuclear spins exhibit long-range order characteristic of a ferromagnetic; T_c is given by:

$$kT_c \approx \Delta E^2/8E_F \tag{4.3.6}$$

where ΔE is the hyperfine splitting of the free atom, and E_F is the electronic Fermi energy; this yields $T_c \sim 10^{-6}$ K, thus making possible the attainment of such temperatures using the method of magnetic cooling. Seven years later, it was shown, again in collaboration with Nabarro, that the domain structure of this nuclear ferromagnetic state is similar to that of ordinary ferromagnets [F65].

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[18] See, for example, Seeger and Teller (1938).

[19] The possibility of energy transfer from the nuclear spins in a metal to the conduction electrons had been realised some years earlier in collaboration with Heitler & Teller in [F14], where it was noted that this transfer occurs sufficiently rapidly to make possible an application of the magnetic cooling method.

Thus in the absence of an external magnetic field, the specimen is divided into domains that are polarised to saturation, but each with a different direction of polarisation so that the total magnetic moment vanishes. The saturation polarisation is approximately 1000 times smaller than in an ordinary ferromagnetic material because the nuclear magnetic moment is of the order of 1/1000 Bohr magneton.

It should be noted that [F33] anticipated by some 14 years, the better known work by Ruderman and Kittel, based on essentially the same interaction (Ruderman and Kittel, 1954).

4.4 Development and Applications of Vector Meson Theory

The importance of including higher order processes referred to above in connection with polaron theory was well appreciated by Fröhlich from his work [F19] with Heitler, during the previous year, in which they attempted to understand, in terms of the second-order process of virtual[20] emission and re-absorption of mesons[21] (assumed to be a scalar bosons), the discrepancy in the values of the magnetic moments of the proton and neutron from those given by Dirac theory. This they were able to do provided the meson–nucleon interaction was assumed to depend on the relative orientation of the nucleon spin and on the angular momentum of the emitted meson. Shortly after, in the course of a chance meeting at the Royal Society in London between Heitler and Nicholas Kemmer, a former student of Pauli, Kemmer criticized their use of such an interaction on the grounds that it would violate parity. Kemmer proceeded to tell Heitler about his own preference for *vector* mesons, described by the Proca equations, in which this problem is avoided. They decided to collaborate, the result being their joint paper [F20] of 1938 in which they addressed the following topics: (a) the *neutron–proton force*, which, in their theory, was based on the exchange of virtual vector (spin 1) mesons between the nucleons, had a range $\hbar/m_o c$ (= λ), and was always attractive in the ^3S-state (unlike the situation if the meson is described by a scalar field); (b) *the proton–proton force*, which was here obtained in the 4th order of approximation, and found to be repulsive at distances below ½λ. To obtain an attractive force that was equal in strength to that between the neutron and proton it was necessary to admit a *neutral* meson of the same rest mass ($m_o \approx 100$ electron masses) as the charged ones; (c) *the self-energies of the neutron and proton*, which were found to diverge; (d) *the*

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[20] A virtual process is one that violates energy conservation but on such a short time Δt that the energy discrepancy is within the associated energy uncertainty, $\Delta E \sim h/\Delta t$ given by quantum mechanics, where h is Planck's constant. Such processes arise in second order perturbation theory, where they take the form of transitions (in which momentum is conserved) to and from all possible so-called 'intermediate states' involving the emission/absorption of (virtual) quanta of some kind. It would be another 10 years, however, before the implications of virtual processes of emission and absorption for solid-state physics were fully appreciated, as will be narrated in Chap. 5.

[21] At the time, the mesons were referred to as 'heavy electrons'.

magnetic moments of the neutron and proton, which were found to be close to those obtained earlier by Fröhlich and Heitler [F19], notwithstanding the fact that here [in F20] *transversely* polarised mesons are involved, in contrast to [F19] where they were assumed to be spinless (i.e. scalar particles). It should be noted that the *non*-divergent values of the magnetic moment arose from the fact that, in an external magnetic field, the divergent self-energy can be expanded as a power series in the field, in which the coefficient of the term linear in the field strength (the magnetic moment) is *finite* (Fig. 4.5).

Despite divergences and certain other defects—not least the fact that the mesons in question were later (1941, 1950) experimentally found to be *pseudo-scalar*[22]—a possibility first noted by Kemmer (1938), but for some reason not at the time pursued —the importance of this pioneering vector extension of Yukawa's original (scalar) meson theory of 1935 should not be underestimated. For, not only was it instrumental in raising awareness of Yukawa's work (Yukawa 1935) in the West, but, more importantly, it also predicted the existence of a *neutral* meson (then called a 'neutretto') 12 years before neutral pions were discovered experimentally in 1950.

This work was quickly followed by three papers in collaboration with Heitler and Kahn [F22, 29, 30] in which the theory of [F20] was applied to two topics. The first was an attempt to understand a possible discrepancy in the fine-structure of hydrogen, which at the time had not actually been experimentally confirmed, but only inferred from the results of spectroscopic studies on deuterium, which were found to be at variance with the Sommerfeld fine-structure formula (Williams, 1938). In the case of hydrogen, Pasternack had pointed out that the inferred discrepancy could be accounted for if, due to a repulsive deviation from Coulomb's law, the $2S_{1/2}$ level was raised 0.03 cm^{-1}*above* the $2P_{1/2}$ state, in contrast to the prediction of the Dirac equation that these states are *degenerate* (Pasternack 1938). It should be appreciated that this shift—now known as the Lamb shift—was definitively established only in 1947 with the work of Lamb & Retherford employing microwave spectroscopy developed during WWII, which gives the separation to be 0.035287 cm^{-1} (1057.845 ± 0.009 MHz), which is close to Pasternack's estimate (Lamb and Retherford 1947). Rather than attributing it to a defect in Dirac's relativistic wave equation for the electron, Fröhlich, Heitler & Kahn suggested in [F22] that the discrepancy was due to the field of the proton being non-Coulombic because, according to meson theory, '…….the proton spends a certain fraction of time in a dissociated state as a neutron and a positive mesotron distributed around the nucleus in a volume with linear dimensions of order of the electronic radius' [F22]. Using the earlier work of Fröhlich, Heitler and Kemmer [F20] (together with Fröhlich's own technique of solving the Schrödinger equation by a perturbation of the boundary conditions [F21]), they showed that the *S*-states were indeed shifted upwards. The estimated shift of the $2S_{1/2}$ state was about 1/3 cm^{-1}; this is 10 times larger than Pasternack's value, but they stressed that not much weight should be attached to the numerical values. In addition, they noted that the deviation of the proton's field from

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[22] Pseudo-scalar meson theory was much later found to be renormalisable.

On the nuclear forces and the magnetic moments of the neutron and the proton

By H. Fröhlich, *H. H. Wills Physical Laboratory, University of Bristol,*

W. Heitler, *Research Fellow of the Academic Assistance Council, H. H. Wills Physical Laboratory, Bristol,*

and N. Kemmer, *Beit Scientific Research Fellow, Imperial College, London*

(*Communicated by N. F. Mott, F.R.S.—Received* 1 *February* 1938)

Introduction

It was first suggested by Heisenberg that the forces between a proton and a neutron are connected with an exchange of charge between the two heavy particles. This exchange nature of the neutron-proton forces is now generally accepted. It would follow from this assumption that in suitable circumstances a proton (neutron) could emit a positively (negatively) charged particle transforming itself into a neutron (proton). At first sight it seemed that the emission of positive or negative electrons in the β-decay could in this way be made responsible for the nuclear forces. This was, in fact, suggested by Iwanenko (1934) and Tamm (1934). It has also been pointed out by Wick (1935) that the virtual emission of β-electrons might explain the values of the magnetic moments of the proton and the neutron. These theories, however, were not successful. The nuclear forces, for instance, turn out to be too small by a factor of more than 10^{10} and have far too small a range; this is due to the fact that the β-decay constant is extremely small. Since the β-decay is a process which, in nuclear dimensions, takes "geological ages", one might think that the ordinary properties of the heavy particles have no direct connexion with this process and that an approximate theory of the nuclear forces should be possible without the inclusion of the β-decay.

A new hope for such an "exchange theory" of the properties of nuclei is offered by the probable existence of a hitherto unknown type of particle constituting the hard component of cosmic radiation. Since these particles do not lose much energy by radiation, it has been suggested by Neddermeyer and Anderson (1937) that they are (positive and negative) "heavy electrons" with a mass between that of an electron and a proton. From cosmic-ray data the mass of these particles can hardly be determined yet,

Fig. 4.5 Fröhlich, Heitler & Kemmer's meson theory paper [F20], 1938—Reproduced with the permission of the Royal Society

Coulombic occurs at a distance of the order of 10^{-15} m (the classical electron radius, e^2/mc^2), which is at the limit of the validity of the meson theory used, and presciently concluded '.....The information obtained from exact measurements of the fine structure of hydrogen will therefore probably be important for the development of future quantum electrodynamics'. Their approach was criticised by Lamb (1939, 1940), but despite a compelling field-theoretic attempt [F29] to refute his criticism, it was eventually established that the shift is *not* due to a deviation from the Coulomb law, but is rather an *electron* self-energy effect.[23] This early calculation of Fröhlich, Heitler and Kahn [F22] is nevertheless significant, however, in that it was the *first* attempt to understand the nondegeneracy of the $2S_{1/2}$ and $2P_{1/2}$ states—an attempt that predates, by 8 years, Bethe's Shelter Island calculation of 1947, which gave 1040 MHz for the shift (Bethe 1947).

The second topic, which was the subject of the third paper [F30], dealt with the photodisintegration of the deuteron, the aim being to use this simple system (comprising a proton and a neutron) to obtain support for the vector meson theory of [F20] in which there is a strong tensor spin-spin coupling. Their idea was to calculate the cross-section for photodisintegration by γ-rays, taking into account the interaction of a light quantum, not only with the proton's electric charge but also, with the electrically charged mesons[24] that, according to meson theory, surround the proton and neutron. Assuming the deuteron ground-state to be the odd-parity ^3S state, they found that, at high energies, the cross-section is much larger than it is if the interaction with the charged mesons is neglected, as was the case with the earlier calculation of Bethe & Peierls (1935); it was also found that the angular distribution is different. The magnitude of these effects was illustrated for the case of 17 MeV γ-rays, the cross-section being 7 times larger than that given by the earlier calculation, whilst the ratio of the number of protons emitted parallel to the γ-ray to the number perpendicular to it was 0.7, compared with the zero value given by Bethe & Peierls. Although their large cross-section was later shown to be illusory by Rarita et al. in (1941), in consequence of certain inconsistencies in their calculation and the $1/r^3$ singularity in their tensor potential (Rarita et al. 1941), the work of Fröhlich, Heitler & Kahn was again still significant, not only in being again the first to address the problem in this way but also, because it motivated others towards more rigorous calculations.

On the occasion of Fröhlich's 80th birthday in 1985, Kemmer recalled some of their early applications of meson theory in a playlet[25] entitled *Wrong Interactions*, which is reproduced in Appendix 2 at the end of this chapter.

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[23] This is an interaction of the electron with its own electromagnetic field, which, within quantum field theory, arises from the emission and re-absorption of virtual photons by the electron, which effectively smear its point charge over a volume of radius of about 0.1 fm. Near the proton, this entails a slightly weaker attraction than in the case of a point electron. Accordingly, states in which the electron has a high probability of being near the proton (e.g. S states) are less tightly bound than are P-states, for example.

[24] This is a spin-dependent exchange, tensor interaction.

[25] In this connection, it is interesting to record that in 1971 Fröhlich (in collaboration with his wife) actually devised a ballet, which they proposed to Covent Garden, based on fundamental

4.5 Internment

With the fall of France in June 1940, Bristol was declared a ‘protected area’, necessitating the removal of Fröhlich and other ‘*Class C* enemy aliens’, including Heitler and Heinz London, out of the city to Wells (Somerset), were they were accommodated in a house in Vicars Close, near the Cathedral. Local tensions and hysteria were running high, so much so that Heitler’s mother, who had hung Walter’s light coloured trousers on a washing line to dry, was suspected of signalling to German aircraft! Despite the strenuous efforts of Tyndall to secure their return, the three men were removed from their lodgings at 7.30 a.m on 25 June, and interned under a new Home Office Order requiring the internment of all *Class C* Aliens. Having fled from Germany and Russia to avoid internment (and probably far worse), Fröhlich considered it highly ironic that this fate should eventually befall him in England!

They were first taken to a very badly organized camp in Paignton (Devon), which had wonderful views over the sea, but virtually no food. This prompted a concerted series of complaints, as a result of which they were removed to another camp just outside Prees Heath, near Whitchurch, in Shropshire. Here, although there was more food, conditions were still not good, with several people being forced to occupy single tents. The matter was eventually raised in Parliament, after which those who so wished were moved into houses. It was a beautiful summer, and in the interest of maintaining closer contact with each other than would otherwise have been possible, Fröhlich, Heitler and others chose to remain under canvas where they established a ‘university’ (which they described as ‘probably the best in England’), giving lectures to the younger people. Heitler somewhat humourously recalled (Heitler 1973) that the military personnel that ran the camp comprised officers who were not best suited to warfare! On one occasion, following the arrival of a group of Austrians, the Colonel was overheard—much to the amusement of the internees—to ask a subordinate whether he had ‘split up the tribes’! Both Fröhlich and Heitler record how much they enjoyed the lectures of the philosopher-mathematician Friedrich Waismann from whom they learned set theory and mathematical logic, particularly, Gödel’s theorems of 1931, which were then relatively new. In addition, they enjoyed excellent chamber music under the direction of Norbert Brainin who later, in 1947, founded the Amadeus String Quartet. They also participated in amateur theatrical productions, which included performances of Shakespeare plays in which all the parts were played by males, as in Elizabethan times (Fig. 4.6).

Whilst at Prees Heath, the internees discussed what they would do in the event of a Nazi invasion. The favoured plan, having seized guns from the British personnel at the camp, was to go to Liverpool where they would take over a ship and escape before the Nazis arrived there. In the event, this never happened, and around

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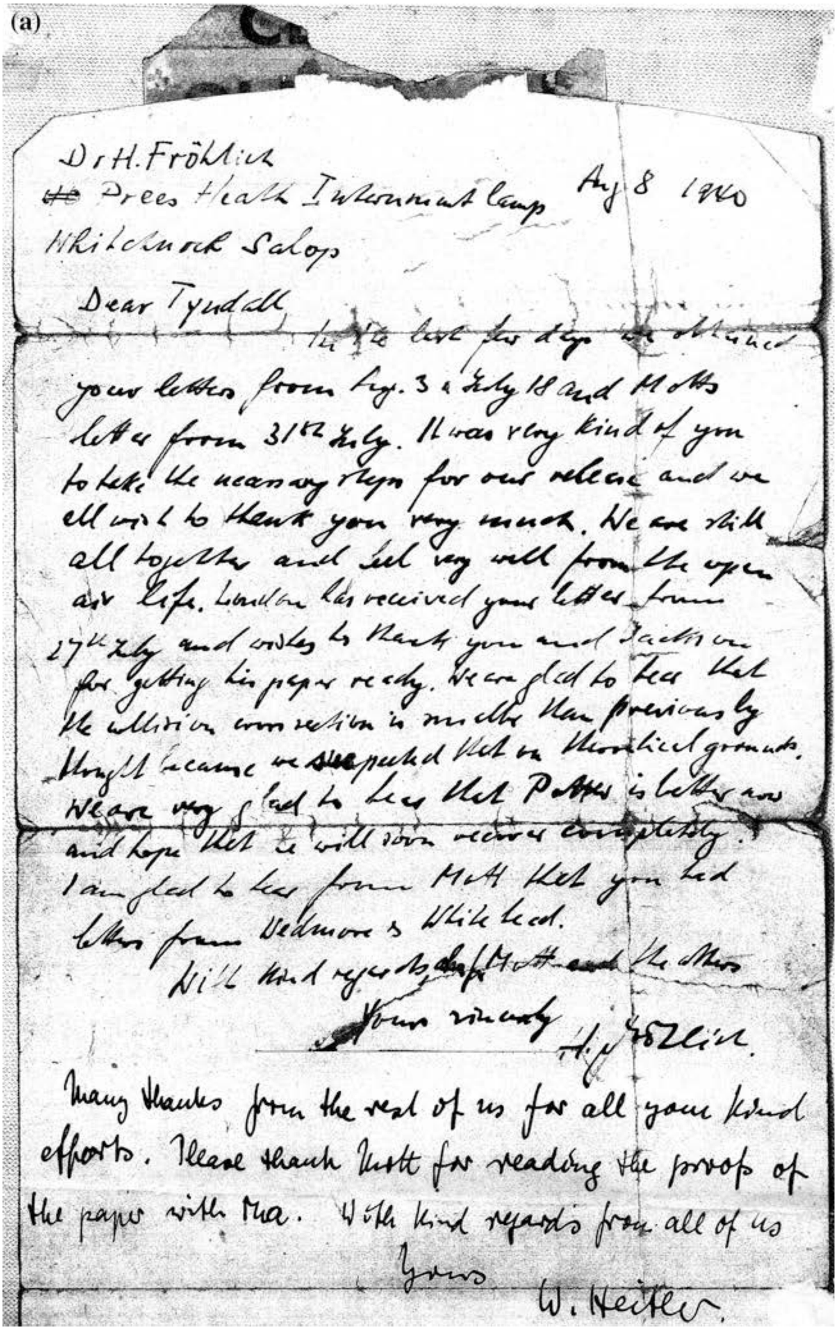
(Footnote 25 continued)
interactions, such as pair-production/annihilation (involving both light quanta and mesons and their antiparticles), β-decay, and the formation of neutral hydrogen atoms from electrons and protons.

(a)



Dr H. Fröhlich
Prees Heath Internment Camp
Whitchurch Salop

Aug 8 1940

Dear Tyndall,

In the last few days we obtained your letters from Aug. 3 & July 18 and Motts letter from 31st July. It was very kind of you to take the necessary steps for our release and we all wish to thank you very much. We are still all together and feel very well from the open air life. London has received your letter from 27th July and wishes to thank you and Jackson for getting his paper ready. We are glad to hear that the collision cross section is smaller than previously thought because we suspected that on theoretical grounds. We are very glad to hear that Potter is better now and hope that he will soon recover completely. I am glad to hear from Mott that you had letters from Wedmore & Whitehead.

With kind regards to Mott and the others

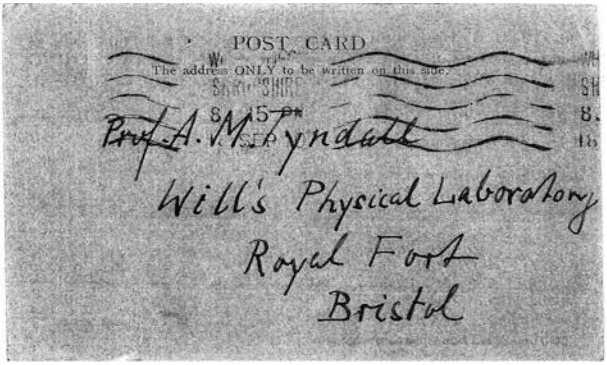
Yours sincerely
H. Fröhlich.

Many thanks from the rest of us for all your kind efforts. Please thank Mott for reading the proof of the paper with Ma. With kind regards from all of us

Yours
W. Heitler.

Fig. 4.6 **a** A letter sent during internment in Prees Heath Camp, near Whitchurch, Shropshire, 1940; **b** the letter's envelope—Reproduced with the kind permission of the Council for Assisting Refugee Academics (*CARA*)

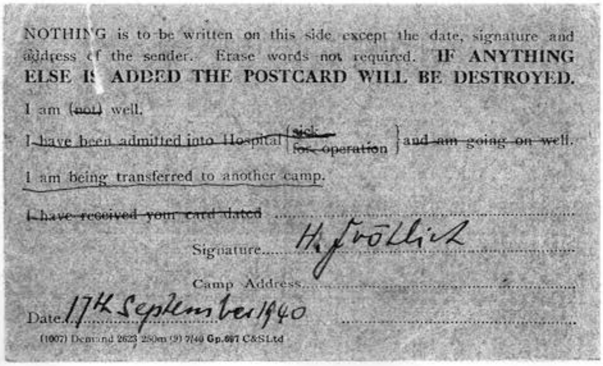
(b)



POST CARD

The address ONLY to be written on this side.

Prof A. M. Tyndall
Will's Physical Laboratory
Royal Fort
Bristol



NOTHING is to be written on this side except the date, signature and address of the sender. Erase words not required. **IF ANYTHING ELSE IS ADDED THE POSTCARD WILL BE DESTROYED.**

I am ~~(not)~~ well.

~~I have been admitted into Hospital~~ { ~~sick~~ / ~~for operation~~ } ~~and am going on well.~~

I am being transferred to another camp.

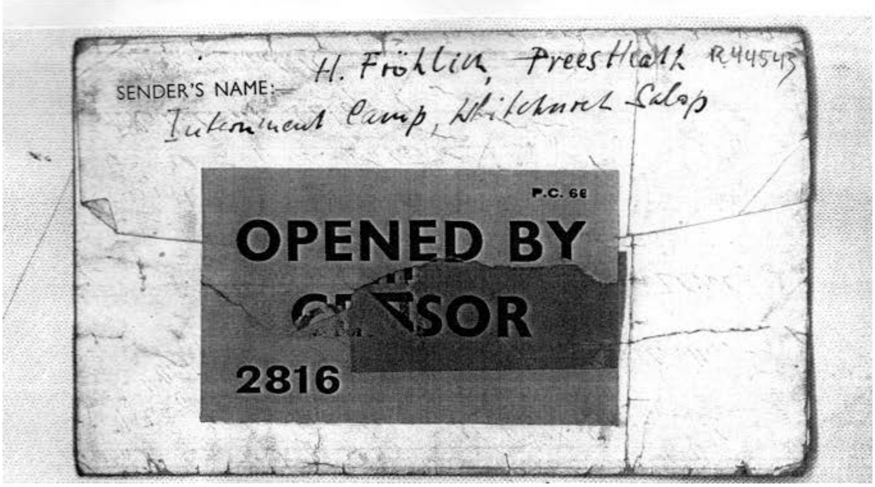
~~I have received your card dated~~ ....

Signature.......... H. Fröhlich

Camp Address..........

Date 17th September 1940 ....

(1007) Demand 2623 250m (9) 7/40 Gp.687 C&SLtd



SENDER'S NAME: H. Fröhlich, Prees Heath R44543
Internment Camp, Whitchurch Salop

P.C. 66

OPENED BY CENSOR

2816

Fig. 4.6 (continued)

18 September, when the tents could no longer withstand the winds, the camp was closed, and the internees shipped off to the Isle of Man. There they were accommodated in houses, which Fröhlich and Heitler found much less agreeable, since it broke up the cameraderie that had been previously built up. On arrival, Fröhlich went through the list of houses with the man in charge, and noticed there was one place that was large enough to accommodate a group. He was told, however, that they couldn't have it, since it had been reserved for orthodox Jews who had special food, to which Fröhlich replied: 'Well, we are the orthodox Jews'. This did the trick, and they were all able to live together as before.[26] A couple of days later, however, he was playing chess with someone in the house who became very angry when he discovered that they were not at all orthodox!

Tyndall persisted with his efforts to have his physicists released, continually assuring the Home Office of their loyalty, and eventually, after almost 3 months (less than 1 week of which had been spent in the Isle of Man), his efforts bore fruit. For on 21 September 1940 he was informed that instructions had been given for Fröhlich's release (Fig. 4.7). Soon after, he and his colleagues were all returned to Bristol, but were forbidden to continue their research into nuclear fission,[27] which they had, however, already given up prior to internment. Owing to an administrative error, they were returned in reverse order of seniority—Fröhlich last but one, followed by a very indignant Heitler!

4.6 Return to Bristol

Back in Bristol, Fröhlich wrote the following letter of gratitude (dated 31 Oct 1940) to a Miss Simpson of the *AAC* (reproduced by kind permission of *CARA*):

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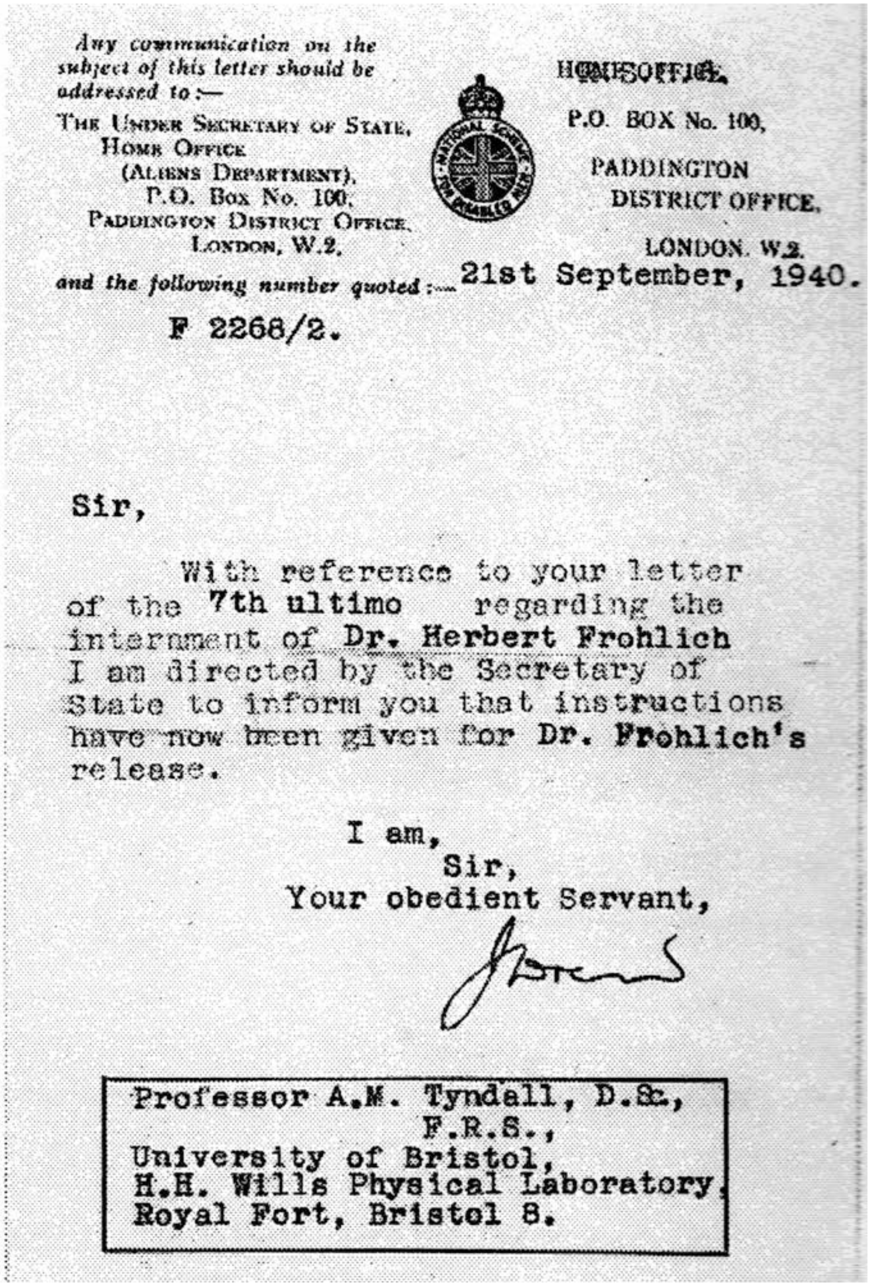
[26] The house could well have been part of the Hutchinson Square Camp in Douglas, where many Jewish intellectuals and artistes were interned (including, for a while, Norbert Brainin), and which became known as the 'University of the Isle of Man'.

[27] There exists an unpublished manuscript co-authored with Heitler (dating from some time between March 1939 and March 1940) entitled *Chain-Reactions in Uranium*, in which they considered the minimum concentration of U^{235} necessary to produce a divergent chain reaction; the absence, however, of the requisite numerical data prevented any definite conclusions from being drawn. Following the Frisch-Peierls Memorandum of March 1940 (*On the construction of a "Super-bomb" based on a Nuclear Chain Reaction in Uranium*), Fröhlich gave up work in this field, never to return to it, despite approaches by Cockroft and others to continue; after the War, he even turned down an invitation to become Head of the Theoretical Physics Division at Harwell, not wanting to be involved with any work that might further nuclear warfare [An extract from the manuscript with Heitler is reproduced in Appendix 3 at the end of this chapter].



Any communication on the subject of this letter should be addressed to:—
The Under Secretary of State,
Home Office
(Aliens Department),
P.O. Box No. 100,
Paddington District Office,
London, W.2.
and the following number quoted:—

HOME OFFICE,
P.O. BOX No. 100,
PADDINGTON
DISTRICT OFFICE,
LONDON. W.2.

21st September, 1940.

F 2268/2.

Sir,

With reference to your letter of the 7th ultimo regarding the internment of Dr. Herbert Frohlich I am directed by the Secretary of State to inform you that instructions have now been given for Dr. Frohlich's release.

I am,
Sir,
Your obedient Servant,

Professor A.M. Tyndall, D.Sc.,
F.R.S.,
University of Bristol,
H.H. Wills Physical Laboratory,
Royal Fort, Bristol 8.

Fig. 4.7 Letter from the Home Office to Tyndall (Head of Physics at Bristol) informing him that Fröhlich is to be released—Reproduced with the kind permission of the Council for Assisting Refugee Academics (*CARA*)

HH Wills Physical Laboratory,
Fort Royal,
Bristol, 8

Dear Miss Simpson,
I enclose cheque for £4 subscription towards your funds for 1940/1. I am glad to be back again. Heitler is now back too, so we are all here again. I would like to thank you and your colleagues for all you have done, and are doing for us.

With kind regards,
Yours sincerely,
H. Fröhlich

During the remainder of the war, Fröhlich was occupied in part-time research for the Ministry of Supply, working initially on an image converter instrument for use on tanks to extend night vision; before it could be perfected, however, radar had come into use.

Despite the irregularity of the wartime, there was no year between 1939 and 1945 in which Fröhlich failed to publish at least one paper; indeed, he published, in total, some 30 papers and *ERA* Reports during this period, most of them dealing with some aspect of dielectric breakdown, including the two topics that the *ERA* had identified in their 1937 letter to Mott—namely, breakdown in *non*-polar materials [F31] and the influence of thickness and impurities on dielectric strength [F34].

Upon his return to Bristol after internment, the *ERA* continued to fund[28] his work —and also that of many of his research students and collaborators (some of whom worked at the *ERA*)—the support continuing after he joined the academic staff in 1943. His work for them was not confined to dielectric breakdown, but included also dielectric loss in both solids and liquids [F36, F39, F41, F44, F45, F51], the theory of the dielectric constant [F43, F44, F45], and other topics of interest to the *ERA*, such as the dielectric properties of dipolar materials [F37, F44, F46, F47, F52] and those containing dipolar long-chain molecules [F50, F52]. In collaboration with R.A. Sack, he also worked on the rheological properties of dispersions [F48, F53], and on the absorption of light by a single layer of alkali metal atoms adsorbed on a dielectric surface, and their emission of photoelectrons [F59].

During this period, Fröhlich published only one paper that was not devoted to dielectric theory, namely [F40], which appeared in *Physical Review* in 1942, in which, following early experimental indications in 1941 that cosmic ray mesons were *pseudo-scalar*, he noted that when the recoil of a nucleon is taken into account, such pseudo-scalar mesons actually make *no* contribution to the nucleon's magnetic moment, in contrast to the prediction of the then fashionable Møller-Rosenfeld theory (Møller and Rosenfeld 1940).

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[28] This support continued for many years, until at least 1961, long after he had moved to Liverpool, despite his persistent refusal to properly complete the appropriate grant application forms, simply writing 'Theoretical work in dielectrics', for which the *ERA* should be given credit for accepting (Powles 1973).

4.7 Dielectric Breakdown Revisited: The Birth of the ‘Hot’ Electron Concept

It will be recalled (vide Sect. 4.3) that since 1939 there had been indications that dielectric strength with a negative temperature coefficient was not confined to the organic materials originally investigated by the *ERA* (Thomas 1938), but was shared by some crystalline alkali halides above a certain temperature in the vicinity of 320 K (Buehl and von Hippel 1939 (KBr)), (von Hippel and Lee 1941 (NaCl)). In their introduction, the latter authors explicitly state that the decrease they had earlier found in KBr [which, incidentally, was not corroborated by the *ERA* (Austen and Whitehead 1940)] could *not* be attributed to the onset of thermal breakdown. A decrease, but at much lower temperatures, had been found also in amorphous materials, such as soda glass (von Hippel and Maurer 1941) and certain varnishes investigated by the *ERA* (Thomas and Griffiths 1942), in the vicinity of 180 K—a temperature so low that the involvement of thermal mechanisms could be ruled out with certainty. A breakdown field that decreased with increasing temperature was, it will be remembered, contrary to Fröhlich’s original theory of 1937, which predicted, instead, a monotonic *increase*. Ironically, it was in the paper confirming[29] the predicted increase in the case of an ionic material to which his original theory was applicable, namely, KBr, that the *ERA* drew attention to their results on varnish. Evidently, Fröhlich’s original theory was not the whole story, and around 1945, in response to the *ERA*’s concerns, he set about developing a theory of breakdown in amorphous dielectrics; the result was the report [F49], which was published in 1947 as [F60].

It will be recalled that in the original theory no account had been taken of inter-electronic collisions, the only collisions considered being between electrons and the lattice vibrations. In his introduction to [F49], Fröhlich pointed out that this neglect of inter-electronic collisions is justifiable only *below* a certain temperature,[30] T_o, above which a conduction electron undergoes more collisions with other conduction electrons than with lattice vibrations. His idea was that this change in collision scenario perhaps underlies the reversal in the sign of temperature coefficient of the breakdown field above above the temperature T_o. It was assumed that the basic idea underlying breakdown was essentially the same as originally—i.e. breakdown occurs when conduction electrons cannot transfer to the lattice all the energy they take up from the applied electric field; the new element that was now introduced concerned way in which this energy transfer occurs. Now, because of the

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[29] The observed increase was, however, somewhat sigmoidal, in contrast to the predicted (non-sigmoidal) dependence.

[30] In the presence of an external electric field, the number of electrons in the conduction band is higher than in the absence of a field, and this number increases with temperature; the electron density is also high just prior to breakdown according to the original theory [F16].

predominance of inter-electron collisions, the energy acquired from the field is first shared amongst the electrons *before* being transferred to the lattice. Accordingly, the electrons may be considered to be in a thermal equilibrium corresponding to a (electron) temperature, T_e, which must be *higher* than that, T, of the lattice, since otherwise no energy would be transferred to the lattice. T_e, is determined by the usual energy balance condition, in which the rate B of energy transfer from the electrons to lattice vibration now depends, not only on T, but also on T_e[31]:

$$A(T_e) = B(T, T_e) \tag{4.7.1}$$

where A is the rate at which energy is transferred to the conduction electrons from the external electric field.

Detailed calculations were made firstly for a amorphous insulator, modelled as a crystalline material with many imperfections. It will be recalled that, in equilibrium at finite temperatures, a finite number of lattice defects are always present even in pure stoichiometric crystals; these defects can be vacant lattice points or ions in interstitial positions, either of which can act as *trapping centres* for electrons that would otherwise be in the conduction band. In addition to a ground-state, such localised electrons have a range, ΔV, of discrete excited states with energies below the lower edge of the conduction band. The number of such traps far exceeds the number of electrons in the conduction band provided the energy of defect formation is less than the energy gap between the conduction and valence bands. In establishing T_e, the conduction electrons share the energy they acquire from the field with these localised electrons, as well as amongst themselves; in turn, provided $\Delta V \gg h\nu$, the localised electrons transfer this energy to the lattice via phonon emission (in quanta $h\nu$) associated with transitions between their excited states, and for $\Delta V \gg kT$, they dictate the form of B, since they are already strongly coupled to be lattice because of their localisation.

It can be shown that, for low fields, the rate balance equation $A = B$ has two solutions for T_e (one stable, one unstable), which converge as the electric field F increases, eventually coalescing at a certain field strength, F^*, above which there is no solution. For $F > F^*$, $A > B$, and the electrons gain more energy from the field than they can transfer to the lattice; F^* is thus the breakdown field—i.e. the field strength at which a stable electron temperature first becomes impossible, and above which T_e starts to increase, and with it the number of conduction electrons; this entails a further increase in the energy transferred from the external electric field which, in turn, further enforces the increase in T_e. Through this positive feedback effect, the process becomes a cooperative one, which continues until eventually the crystal breaks down.

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[31] From a formal point of view, T_e can be considered as the counterpart of the energy, E in the original theory of 1937. E, however, was eventually set equal to the ionization energy, I_o, which is a given quantity; T_e, on the other hand, has to be determined as a solution of the balance condition $A = B$.

In contrast to the situation at $T < T_o$, where independent fast electrons are responsible for breakdown, at $T > T_o$, it is the *collective* action of *slow* electrons (with thermal velocities) that is here responsible, since it is through their collisions both amongst themselves and with the localised electrons (a collective effect) that an electron temperature, T_e, *higher* than that, T, of the lattice, can be defined.

Unlike in his original theory of dielectric breakdown, Fröhlich was here unable to derive quantitative results for the value of F^*, on account of the absence at the time (1945) of any proper theory of the motion of *slow* electrons in amorphous insulators. It was, however, possible to deduce the T-dependence of F^*, which was found to be of the form:

$$\ln F^* = const + \frac{\Delta V}{2kT}, \quad \text{provided } h\nu \ll \Delta V \text{ and } \; kT \ll \Delta V, \tag{4.7.2}$$

i.e. the new theory predicts that F^* decreases with increasing temperature, in agreement with experiments on amorphous materials.

Fröhlich presciently noted that this will 'lead to a theory of the deviations from Ohm's law as well as to a theory of breakdown' [F60, p. 523]. Ohm's law states that the electric current density, J, that flows in response to an applied electric field, F, is proportional to the field—i.e.

$$J = \sigma F \tag{4.7.3}$$

where σ is the electrical conductivity. At low fields, where $T_e = T$, σ is a function of T, and so J is linearly proportional to F. In high fields, however, $T_e > T$, and T_e now depends on F, so that J depends *non*-linearly on F—i.e. deviations from Ohm's law will occur. Out of this there later evolved the technologically important field now known as that of 'hot electron' physics—a term coined by Shockley in 1951 in a publication dealing with non-ohmic conduction, which appeared one year *after* Fröhlich's presentation of his own ideas at Bell Labs (Shockley 1951) (Fig. 4.8).

In an accompanying paper [F61] of 1947, Fröhlich showed[32] that however weak is the field, the inclusion of inter-electronic collisions is actually *essential* for the realisation of a stationary state, and for the correct calculation of the electric current. This important paper changed the course of his subsequent work on dielectric breakdown, and in 1952, after moving to Liverpool, he extended the electron temperature concept, which had earlier been introduced in the context of amorphous materials, to defect-free crystalline materials, the results being summarised in an *ERA* report [F83].

For the case of an ideal ionic crystal, the new expression for the breakdown field yielded a somewhat higher value than that given by his more stringent original theory of 1937; the decrease in breakdown field derived in the amorphous case was

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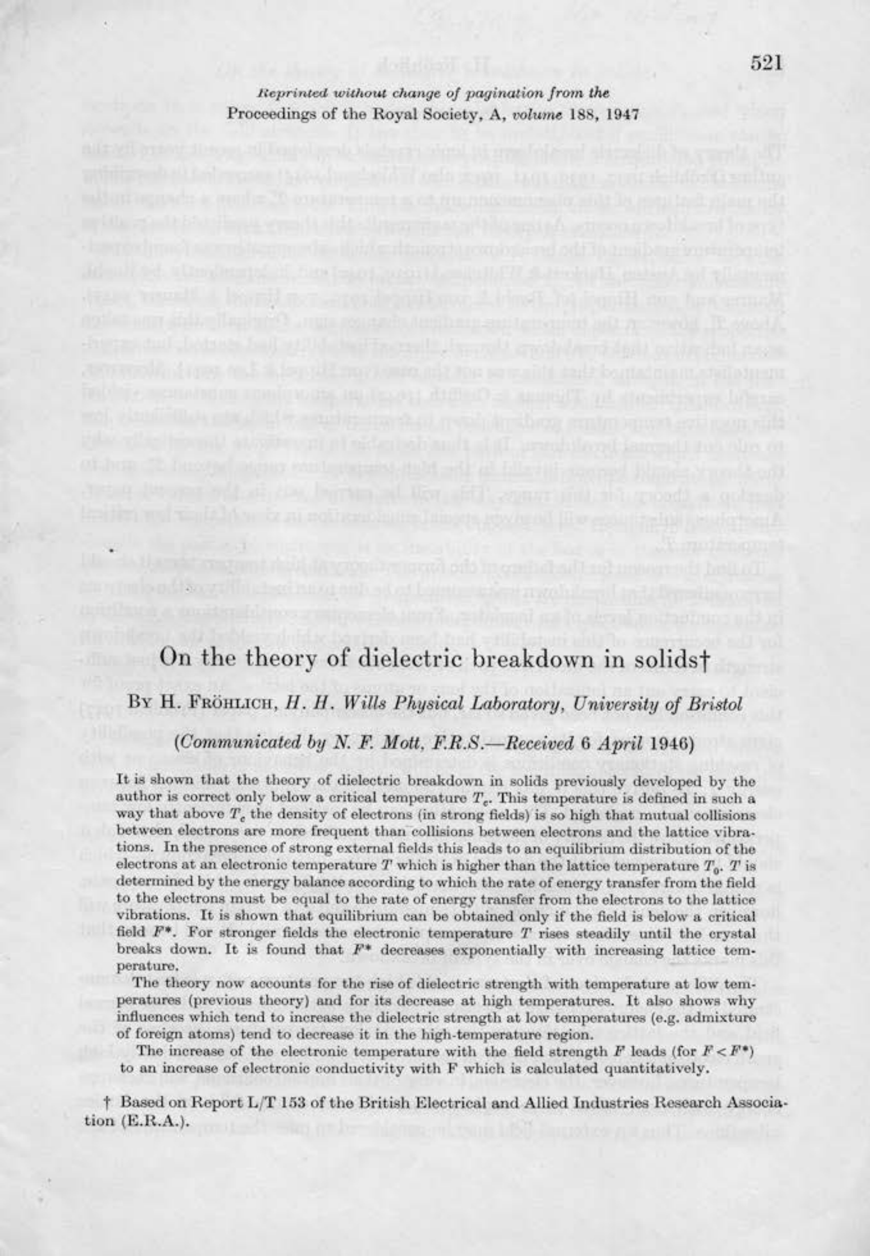
[32] For further discussion, see Paranjape (1973).



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On the theory of dielectric breakdown in solids†

By H. Fröhlich, *H. H. Wills Physical Laboratory, University of Bristol*

(*Communicated by N. F. Mott, F.R.S.—Received* 6 *April* 1946)

It is shown that the theory of dielectric breakdown in solids previously developed by the author is correct only below a critical temperature T_c. This temperature is defined in such a way that above T_c the density of electrons (in strong fields) is so high that mutual collisions between electrons are more frequent than collisions between electrons and the lattice vibrations. In the presence of strong external fields this leads to an equilibrium distribution of the electrons at an electronic temperature T which is higher than the lattice temperature T_0. T is determined by the energy balance according to which the rate of energy transfer from the field to the electrons must be equal to the rate of energy transfer from the electrons to the lattice vibrations. It is shown that equilibrium can be obtained only if the field is below a critical field F^*. For stronger fields the electronic temperature T rises steadily until the crystal breaks down. It is found that F^* decreases exponentially with increasing lattice temperature.

The theory now accounts for the rise of dielectric strength with temperature at low temperatures (previous theory) and for its decrease at high temperatures. It also shows why influences which tend to increase the dielectric strength at low temperatures (e.g. admixture of foreign atoms) tend to decrease it in the high-temperature region.

The increase of the electronic temperature with the field strength F leads (for $F < F^*$) to an increase of electronic conductivity with F which is calculated quantitatively.

† Based on Report L/T 153 of the British Electrical and Allied Industries Research Association (E.R.A.).

Fig. 4.8 Paper [F60] containing the genesis of 'hot electrons', 1947—Reproduced with the permission of the Royal Society not, however, reproduced, as will be discussed further below. This report later (1956) formed the basis of an important paper with B.V. Paranjape [F97] that ‘…offered for the first time a possibility of calculating the breakdown field in terms of measurable quantities (ionic density, electron mobility, Debye temperature and effective mass)’, and in 1961 was the subject of his final *ERA* report on dielectric breakdown, entitled ‘ Hot Electrons’ [F112]. In [F97] it was shown that because of the smallness of the energy exchange involved in collisions between electrons and the lattice vibrations, a high density of conduction electrons is not actually necessary in order that inter-electronic collisions dominate those with the lattice vibrations. Expressions were derived for the breakdown field in both polar and non-polar materials, which in both cases was predicted to weakly *increase* with increasing lattice temperature, as in the case in his original theory of 1937.

In the case of polar materials, where slow electrons interact most strongly with the longitudinal optical vibrations of of *long* wavelength, the crystal could be treated as dielectric continuum characterised by the high and low frequency dielectric constants ε_∞ and ε_s, as done in polaron theory, which by then had been developed (vide Sect. 5.3), and which was accordingly incorporated in the new theory of breakdown; its inclusion was reflected in the appearance of the factor $[1/\varepsilon_\infty - 1/\varepsilon_s]$ in the derived[33] expression for the breakdown field, which took the following form:

$$F^* \cong \frac{me}{\hbar}\omega_L[1/\varepsilon_\infty - 1/\varepsilon_s]\ G(T/\Theta_L). \tag{4.7.4}$$

Here, $k\Theta_L \equiv \hbar\omega_L$, where ω_L is the frequency of long wavelength longitudinal optical mode, and $G(T/\Theta_L)$ is a monotonically, slowly increasing function of temperature, T, with values that range from about 0.5 at $T = 0$ K to about 0.8 as $T \to \infty$. It is to be noted that this expression contains no arbitrary constants, and yields breakdown field values slightly higher than those given by the original theory of 1937, but which are still lower than those given by von Hippel’s criterion.

It is also to be noted that the monotonically *increasing* dependence of the breakdown field with increasing temperature predicted by the new theory is not only opposite to the decreasing dependence predicted for amorphous materials where the concept of an electron temperature first originated [F49, 60], but clearly cannot account for the *reversal* in the T-dependence of F^* observed in KBr and NaCl near 320 K, as had originally been hoped. Presumably, the decrease predicted in the case of amorphous materials is connected with the assumed existence of electrons localised at trapping centres where they are already strongly coupled to be lattice.

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[33] This was based on perturbation theory, which here cannot really be justified, as noted in [F90], because the electrons interact so strongly with the long wavelength optical phonons.

In a later attempt to understand this reversal in terms of hot electrons, O'Dwyer invoked a variant of Fröhlich's 'amorphous' theory in which, with increasing temperature, the number of trapping centres increases as exp $(-W_D/kT)$, where W_D is related to the defect formation energy, until eventually their concentration become such that the electronic structure of the crystal starts to approach that of an amorphous material—i.e. contains many shallow electron traps, associated with which is a breakdown field with a negative temperature coefficient (O'Dwyer 1957, 1964). It should be noted, however, that at the relatively low temperatures at which the reversal is observed (320 K in the case of NaCl), the defect concentration is negligible for typical defect formation energies, $W_D \sim 2$ eV, raising doubts about the validity of such an approach. The whole question of the reversal in the temperature dependence of the breakdown field in crystalline polar dielectrics thus remained very much open, not only theoretically, but also experimentally, given the rather conflicting data then available.

In view of the somewhat convoluted development of Fröhlich's ideas on dielectric breakdown, which spanned almost a quarter of a century, a brief recapitulation the salient points might prove helpful:

(1) There are essentially two quite distinct theories of breakdown: the original theory of 1937, based on consideration of independent fast electrons in ionic crystals [F16], and a later theory, dating from 1945, based on the collective electron concept of 'hot electrons' in amorphous materials [F49, 60], which was later (1952) adapted to defect-free crystalline materials [F83, 97].

(2) In both theories, the criterion for breakdown is derived from consideration of the balance between the rate at which conduction electrons absorb energy from the applied electric field and the rate at which it is dissipated, which in both cases is assumed to be to lattice vibrations.

(3) The two theories differ in the assumptions made about the relative importance of electron-electron collisions in comparison with electronic collisions with lattice vibrations, the former being neglected in the original theory.

(4) It was subsequently shown that it is essential *always* to include electron–electron collisions since without them it is impossible ever to have a state of steady current [F61]; accordingly, the original theory must be considered to be superseded by the hot electron version, adapted to defect-free crystalline materials, as finally formulated in [F97].

(5) In both theories, breakdown is connected with an increase in the number of conduction electrons, associated with which in the case of hot electrons is an uncontrolled increase in the electron temperature, which ultimately destroys the crystal.

(6) The hot electron theory was developed in the hope of accounting for the negative temperature coefficient of the breakdown field found in some materials, but was successful only in the case of amorphous materials. When later adapted to crystalline materials, a positive temperature coefficient was

> predicted, as in the case of the original theory. In both cases, the derived expression for the breakdown field did not contain any adjustable parameters; the values predicted by the hot electron theory were somewhat higher than those of the original theory, but still below the experimental values.

It should be noted that collaboration between theoretical physicists and industry, such as Fröhlich enjoyed with the *ERA*, was not then common, and it was in an attempt to redress this situation that Fröhlich titled his series lectures given at the Royal Institution just after the war, in 1946, *Theoretical Physics and Industry* [F55]. Out of this arose a consultancy with *ICI*, but, despite this, Bristol was not awarded an *ICI* Fellowship; they did, however, offer him funding for further collaboration, which he passed on to the Bristol physics department, having no need of it himself. Another industrial consultancy had arisen earlier when Willis Jackson (later Lord Jackson of Burnley—see also Sect. 6.3), whom Fröhlich already knew via the *ERA*, joined Metropolitan-Vickers in 1936.

Fröhlich then combined his joint interest in meson theory and dielectrics, studying the problem of the decay and capture of slow negative mesons in dielectrics. Here it was shown, on the basis of some earlier[34] work with Pelzer [F67], that the rate at which a meson transfers energy to the dielectric is very much lower than it is in a metal, the associated time of slowing-down being orders of magnitude longer, and possibly in excess of the meson decay time [F63], consistent with the experimental observation of decay electrons. A further contribution in this area was made the following year, in collaboration with Huby, Kolodziejski & Rosenberg [F68], just before Fröhlich left Bristol. Comparing the radiative energy loss with that due to the Auger effect, it was shown that the latter is very much weaker in a dielectric than in a metal, so much so that it is here likely to play only a negligible role.

In addition to these solid-state activities, Fröhlich established a research group in nuclear structure, applying in 1947, in collaboration with Huang, Ramsey and Sneddon [F62, F64], the results of Møller-Rosenfeld meson theory to calculate the approximate binding energies of very light nuclei.

As narrated in Norman Thompson's Memoirs[35] of Bristol at this time, the physics department was large enough to contain an interesting mix of people, but not so large that it fragmented into independent groups. Most people knew everyone else, the atmosphere was friendly, which greatly facilitated exchange of ideas between the different fields represented, which ranged from dielectrics to cosmic rays showers, in which the pion was discovered in 1947 by Powell's group, and for which he was awarded a Nobel prize in 1952. Other research activities included Herbert Skinner's work on soft X-ray emission in metals, and Harry Jones' theoretical work on the electron theory of metals.

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[34] This work was not published, however, until 1948, and so post-dates [F63] in the Fröhlich bibliography at the end of this book.

[35] http://www.phy.bris.ac.uk/history/07.%20Thompson's%20History.pdf; see also N.F. Mott, in *Biographical Memoirs of the Royal Society*, Vol. 38, 147–162 (1992).

In addition, there was a good social life running in parallel with the scientific activities. A popular weekly event was the 'Lab' walk each Sunday morning, when, if one felt like it, one turned up (together with sandwiches) at the Clifton Suspension Bridge at 10 a.m. Fröhlich was apparently almost always present, and it was he who decided where to go; nobody else knew where and nobody very much cared. Everyone just walked and talked, often about physics, until someone decided it was time for lunch, after which the walk continued well into the afternoon until they arrived on a bus route that would take them back into Bristol.

After the War, such was the high regard in which Fröhlich was held that, notwithstanding his original nationality, he was invited to become Head of the Theoretical Physics Division at Harwell, having become naturalised as a British citizen on 31 August 1946. He declined this offer, however, not wanting to be involved with any work that might further nuclear warfare, despite his earlier interest in nuclear fission as evidenced by his pre-war work with Heitler mentioned earlier; Klaus Fuchs[36] was appointed in his place! Accordingly, he remained in Bristol where he had been appointed Lecturer in Theoretical Physics in 1943, staying until 1948. During his time at Bristol, his status rose from that of a refugee, supported by grants from the *AAC*, to the position of Reader.

Sometime in the mid-1940s, Fröhlich arranged for his brother, Ali, who had been living in Palestine since before the war, to be admitted to the university to study mathematics under Hans Heilbronn, himself a refugee from Nazi Germany, during which time the two brothers shared accommodation. Ali went on to become a distinguished mathematician, and a Fellow of the Royal Society; he died in 2001.

In November 1946, Fröhlich was proposed for Fellowship of the Royal Society, by N.F. Mott, but was not elected until 1951 (Fig. 4.9).

During his time in Bristol, Fröhlich's lectures included the following topics: Atomic Theory for Chemists (1939/40/1); Quantum Mechanics (1942); Statistical Mechanics (1943); Theory of Radiation, Statistical Mechanics, Electrodynamics (1944, 1947); Introduction to Theoretical Physics—Mechanics of Mass Points, Potential theory, Relativistic Mechanics, Statistical Mechanics (1944/5); Meson Field Theory (1945/6); Nuclear Theory (1946/7); Special Relativity (1947).

Before Fröhlich left Bristol, he made the first of many visits to the Dublin Institute for Advanced Studies to where his friend and former colleague W. Heitler had moved in 1941, becoming Director of the School of Theoretical Physics in 1946, following Schrödinger's resignation.

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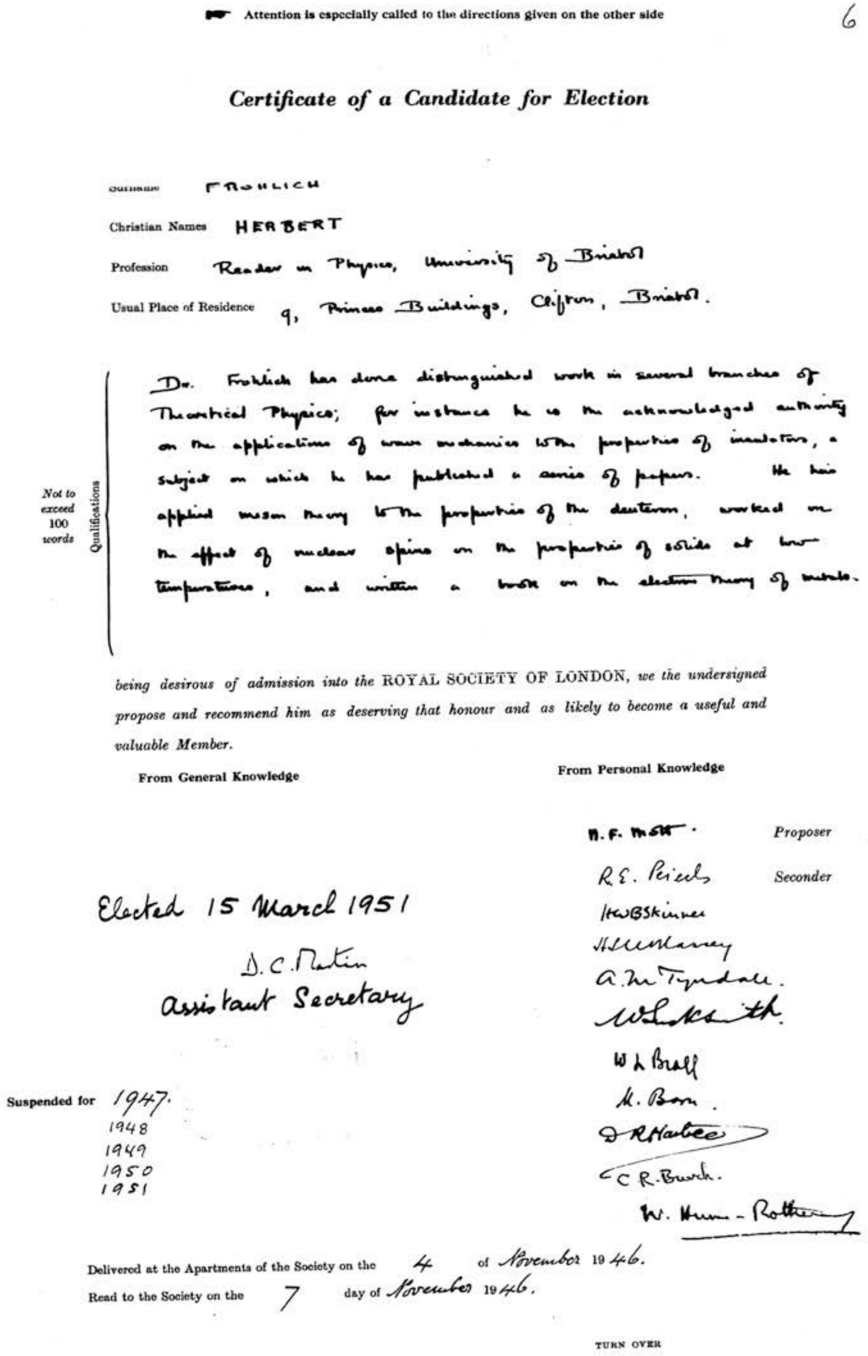
[36] In 1950, Fuchs confessed to passing secrets relating to the Manhattan Project to the Russians; after 9 years imprisonment, he emigrated to E. Germany where he was elected to the Academy of Sciences.



Attention is especially called to the directions given on the other side

6

Certificate of a Candidate for Election

Surname FROHLICH

Christian Names HERBERT

Profession Reader in Physics, University of Bristol

Usual Place of Residence 9, Princes Buildings, Clifton, Bristol.

Not to exceed 100 words

Qualifications

Dr. Frohlich has done distinguished work in several branches of Theoretical Physics; for instance he is the acknowledged authority on the applications of wave mechanics to the properties of insulators, a subject on which he has published a series of papers. He has applied meson theory to the properties of the deuteron, worked on the effect of nuclear spins on the properties of solids at low temperatures, and written a book on the electron theory of metals.

being desirous of admission into the ROYAL SOCIETY OF LONDON, *we the undersigned propose and recommend him as deserving that honour and as likely to become a useful and valuable Member.*

From General Knowledge

Elected 15 March 1951

D. C. Martin

Assistant Secretary

From Personal Knowledge

N. F. Mott. *Proposer*

R.E. Peierls *Seconder*

HWBSkinner

HSWMassey

A.M. Tyndall.

W Sucksmith.

W L Bragg

M. Born.

D R Hartree

C R Burch.

W. Hume-Rothery

Suspended for 1947.
1948
1949
1950
1951

Delivered at the Apartments of the Society on the 4 of November 1946.

Read to the Society on the 7 day of November 1946.

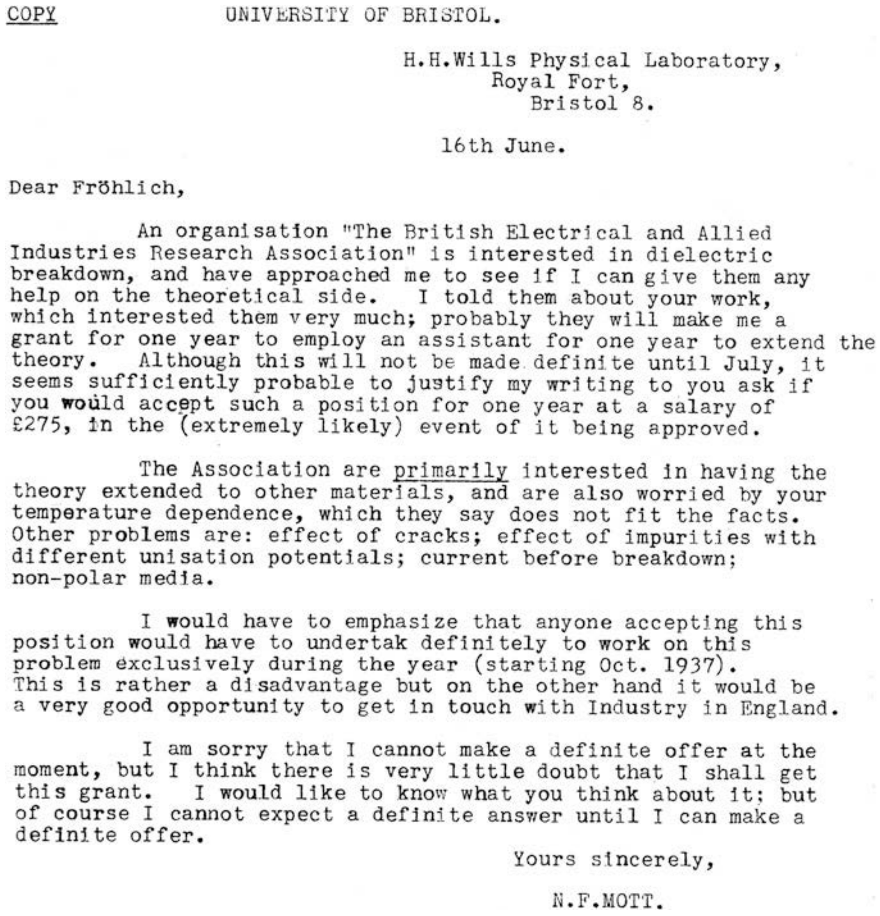
TURN OVER

Fig. 4.9 Royal Society proposal certificate—Reproduced with the permission of the Royal Society

Appendices

Mott's Letter to Fröhlich in Holland concerning work for the ERA

Reproduced with the permission of CARA



COPY UNIVERSITY OF BRISTOL.

H.H.Wills Physical Laboratory,
Royal Fort,
Bristol 8.

16th June.

Dear Fröhlich,

An organisation "The British Electrical and Allied Industries Research Association" is interested in dielectric breakdown, and have approached me to see if I can give them any help on the theoretical side. I told them about your work, which interested them very much; probably they will make me a grant for one year to employ an assistant for one year to extend the theory. Although this will not be made definite until July, it seems sufficiently probable to justify my writing to you ask if you would accept such a position for one year at a salary of £275, in the (extremely likely) event of it being approved.

The Association are primarily interested in having the theory extended to other materials, and are also worried by your temperature dependence, which they say does not fit the facts. Other problems are: effect of cracks; effect of impurities with different unisation potentials; current before breakdown; non-polar media.

I would have to emphasize that anyone accepting this position would have to undertak definitely to work on this problem exclusively during the year (starting Oct. 1937). This is rather a disadvantage but on the other hand it would be a very good opportunity to get in touch with Industry in England.

I am sorry that I cannot make a definite offer at the moment, but I think there is very little doubt that I shall get this grant. I would like to know what you think about it; but of course I cannot expect a definite answer until I can make a definite offer.

Yours sincerely,

N.F.MOTT.

'Wrong Interactions'—a Playlet by N. Kemmer[37]

ACT I, Scene 1

(*The curtain rises, revealing a very dimly lit stage, with no particular shapes or objects clearly discernable, but with the entire space in a state of agitation—not unlike the air above a glowing brazier. Streaks of brightness, like flying sparks, appear and disappear, predominantly in pairs, but occasionally and with increasing frequency, singly.*)

As each actor or group of actors speaks they come into sharper focus.

Carl Anderson	I've found a new particle
Crowd murmuring	Not another!
C.A.	'…..in cosmic rays… with unit charge……. about 200 times more massive than an electron
A group of theorists	Hey! Wasn't there a paper by a Japanese accounting for nuclear forces—two years ago—with a charged boson? Let's see—Ah yes, Yukawa. Would you believe it! He wants a mass of about 200! We'll have to study this
A smaller group among the theorists	…….and Pauli and Weisskopf have that 'anti-Dirac' paper. They show that a charged field of spinless bosons would behave very much like the Dirac field. That must be what Yukawa is doing!
A distant voice (with Japanese accent)	No I'm not!
Bhabha (*From the other side of the stage*)	I'll try to do the same
Proca	I've found an entirely new set of equations for the electron. I'm sure they are consistent
Fröhlich & Heitler	Let's get going. We must modify Yukawa's theory to get spin-dependent nuclear forces
Kemmer	But Proca's equations are just right for Yukawa's boson modified to spin 1. I must apply Pauli-Weisskopf to them!
The Japanese voice	But that's where I started. Like Maxwell's equations!
Voices of many theorists	But they are quite wrong! Electrons have spin ½, not 1

---

[37] 'This Playlet was my contribution to the Symposium held in Liverpool on December 18th 1985, to celebrate Herbert Fröhlich's eightieth birthday, which I had the honour to chair. It does not seem to me that this fragment need be weighed down by explanatory notes and references. Any reader seeking these will find a very good account of this piece of history in: Mukherji, V, Archiv. of Hist. of Exact Sci. **13**, 17, 1974. Short reports of my own that have most of the relevant references are: Kemmer, N., Jour. de Physique **43**, Colloq. Suppl. C-8, p 359, 1982 (in English), and more specifically: Kemmer, N., Physikal. Blätter, **39**, 170, 1983 (in German)'.

Heitler (*meeting Kemmer*)	Hullo, Pauli asked me to get in touch—what are you up to?
Kemmer	I'm busy getting spin into the Yukawa's forces
Heitler	Fröhlich and I are doing the same. We've put a spin interaction into the Pauli-Weisskopf scalar theory
Kemmer	But that's rubbish
Pauli (*Distant voice*)	Hear, hear!
Kemmer (*Pauli's approving voice is heard again overlaid by what seem to be infantile gurgles and chuckles that sound strangely Chinese.*)	You are breaking an absolutely inviolable law; your way doesn't conserve parity!
Kemmer	I think my way is better. Ever heard of the Proca equations?
Heitler	Let's collaborate. We'll have to learn about Proca

ACT I, Scene 2

(*a few months later*)

Yukawa (with Sakata & Taketani)	Here is our paper. We now have the full vector boson theory. It looks like electrodynamics. We've added the 'vector potential' bits now
Fröhlich, Heitler, & Kemmer	Here is our paper, with a vector boson obeying Proca's equations
Bhabha	Here is my paper with a full vector boson theory
All seven (Y, S & T; F, H, & K; B)	We are all saying just the same things! We will start predicting things for Anderson's particle (which he calls 'mesotron'); we've fitted things to give the right neutron/proton force. We all agree that a neutral mesotron is needed for the full nucleon-nucleon force scheme
Kemmer	I've been studying charge independent N–N forces for a long time and wanted to have the whole scheme in our joint……
Fröhlich and Heitler (*interrupting*)	It would have made our paper too long. That's why you are publishing that stuff on your own[38]. That other paper of yours was useful too. You've shown that vector bosons are O.K.; scalar, Pauli-Weisskopf ones aren't. Nor are the pseudo-scalar and pseudo-vector cases that you dragged in

---

[38] Kemmer's paper immediately preceded that with Fröhlich & Heitler [F20] in the same journal.

ACT I, Scene 3

(*at the beginning of this scene the light gradually gets very dim*)

The group of 7, with Kobayashi	The more we compare our predictions with mesotron behaviour, the worse the agreement looks! Perhaps after all our theory is too shaky. We can't get beyond first order perturbation, the coupling is too strong for that and the higher order infinities are horrendous
Fröhlich & Heitler	Even so, we did try to get something sensible on magnetic moments, but nobody believes us
Faint disembodied voice	Wait for renormalisation!
Stueckelberg (*equally faint*) (*Nobody on the stage hears these last two calls.*)	I've nearly got that!

[THERE IS AN INTERVAL FOR REFRESHMENTS, WORLD WAR II, etc.]

ACT II

(*Some years later; the scene is the same as before but it has become a good deal brighter. The 'atmospheric phenomena' continue but we are getting more varied flying sparks appearing and disappearing. The figure of Fröhlich is seen to move slowly to the back of the stage, where throughout the act he seems to be building beautiful structures which begin to look much more solid than anything that had loomed up previously. Other figures join Fröhlich. Occasionally he stops to look back at the action in front.*)

Powell (flanked by Lattes and Occhialini)	All your fine ideas had nothing to do with the mesotron—which we call muon, but here—catch!
(*Powell tosses something to the previous group of theorists who have been joined by quite a few others*)	There's my new pion—that ought to suit you!
A large group of experimenters, including Björklund et al., & Steinberger et al.	And here's a neutral pion for you too! (*They toss it over*)
The theorists	Look—they make a fine isotriplet
Heitler	That's good, it will fit the facts!

Tomonaga, Schwinger, Feynman and Dyson together	We know how to make good sense of field theories—some of them. Quantum electrodynamics makes perfect sense now!
Old theorists	Then tell us how to deal with vector bosons! That's what pions are—isn't it?
Young theorists (Y.T.) and young experimenters (Y.E.)	No!
Y.T	That theory doesn't renormalise, it's horrible!
Y.E	It doesn't fit experiments either!
Old Theorists	What then?
Pauli (*addressing Kemmer*)	That was a stupid, careless thing you did long ago; why didn't you link your isospin ideas with the other stuff? With isospin you get an additional factor (-3) for the deuteron ground state; the vector boson calculations by all of you were wrong!
Old theorists	Oh dear!
Pauli	But the pseudoscalar case…
Kemmer	The one we thought least likely……
Pauli	Yes, that. It seems to fit best!
Chorus of experimenters	Yes it does
Salam	And I can renormalise that one!
Pauli	But that's still not much good because your coupling constant is huge
Experimenters	….and the interaction that fits is not the renormalisable one…
Old theorists	So what is left of our theories? Not the vector boson, it seems
Heitler	But the isospin idea is good
Kemmer	And I'm glad I thought of the pseudoscalar
Young theorists	But with the strong coupling have we any way to improve our theories? It'll be a hard job. We'll look for new methods
(*A complex plane looms; many young theorists climb on to it.*) *As the curtain falls, the light dims somewhat, spark flying increases dramatically*).	

ACT III

(*Many years later. Scene as before, with many new big strong buildings in the background. Fröhlich is still there in a large company. In the foreground the spark flying is not quite as intense but new types of sparks in the three primary colours start to grow prominent. Previous actors look old.*)

Chorus of very young people	How silly you were! It was all a red herring. Your baryons and mesons aren't elementary!
Oldsters	Not elementary—then what……
Young ones	Look at our quarks and gluons!
Oldsters	Does that mean you have field theories that work?
Young ones	You'd be surprised!
Oldsters	What field theories?
Young ones	We use vector bosons!
Oldsters	What! They were quite dead!
Young ones	Ours are alive and kicking. Can you spare some millions to help us find more of them?They're rather different from yours.
Oldsters	Not Proca?
Young ones	No! Yang-Mills
Klein (*faint voice from the past*)	What I nearly had long ago!!
Shaw (*also faintly*)	And what I got pipped at the post with and never published!
Oldsters	Vector bosons! What masses?
Young ones	We don't put in masses. We've gone non-linear. Isospin and symmetries like it are the key
Higgs and Kibble	………. and have to be broken!
Oldsters	And can you renormalise?
t'Hooft	I can, for these gauge theories!
Young ones	It's all very hopeful
Tutti	But oh, so complicated!

Curtain falls with lighting fluctuating more and more strongly and fast.
The shooting sparks become one great swirling mass.

THE END

Fragment of an Unpublished Manuscript with Heitler on Chain Reactions (c.1939/40)

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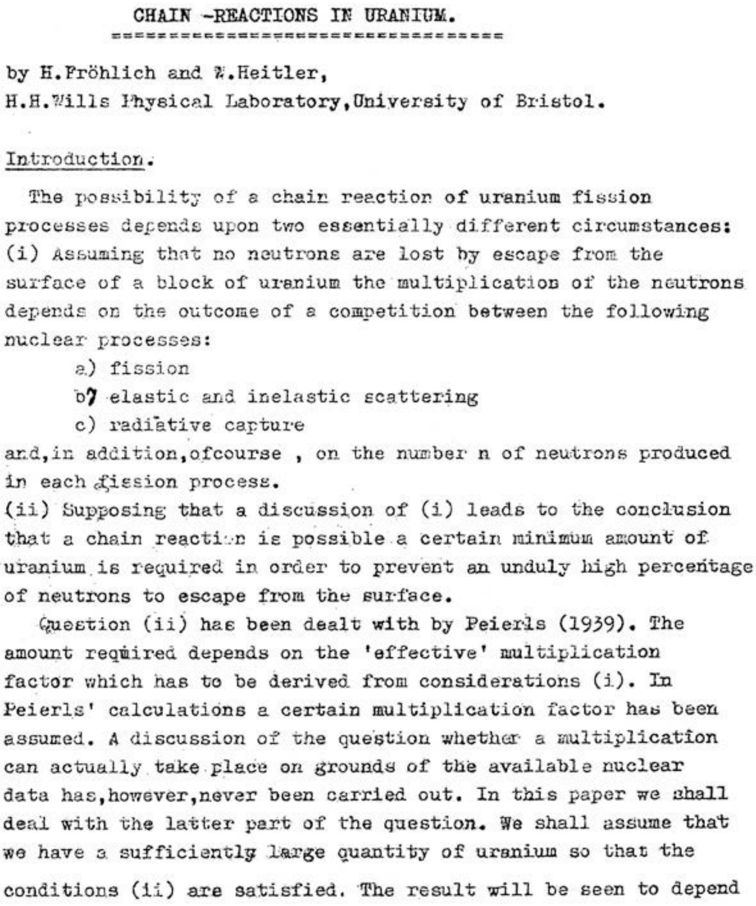

CHAIN -REACTIONS IN URANIUM.

by H.Fröhlich and W.Heitler,
H.H.Wills Physical Laboratory,University of Bristol.

Introduction.

The possibility of a chain reaction of uranium fission processes depends upon two essentially different circumstances: (i) Assuming that no neutrons are lost by escape from the surface of a block of uranium the multiplication of the neutrons depends on the outcome of a competition between the following nuclear processes:

a) fission
b) elastic and inelastic scattering
c) radiative capture

and,in addition,ofcourse , on the number n of neutrons produced in each fission process.
(ii) Supposing that a discussion of (i) leads to the conclusion that a chain reaction is possible a certain minimum amount of uranium is required in order to prevent an unduly high percentage of neutrons to escape from the surface.

Question (ii) has been dealt with by Peierls (1939). The amount required depends on the 'effective' multiplication factor which has to be derived from considerations (i). In Peierls' calculations a certain multiplication factor has been assumed. A discussion of the question whether a multiplication can actually take place on grounds of the available nuclear data has,however,never been carried out. In this paper we shall deal with the latter part of the question. We shall assume that we have a sufficiently large quantity of uranium so that the conditions (ii) are satisfied. The result will be seen to depend

Photo Gallery

The numbers in round brackets at the end of each caption correspond to the section of the text to which the photographs refer.

Unless stated otherwise, all photographs are from private collections.

Fig. 4.10 At a meeting in Liverpool to celebrate Fröhlich's 80th birthday: Left to right - Mott, Fröhlich, Mrs Fröhlich, Kemmer (Sects. 4.3 and 4.4)

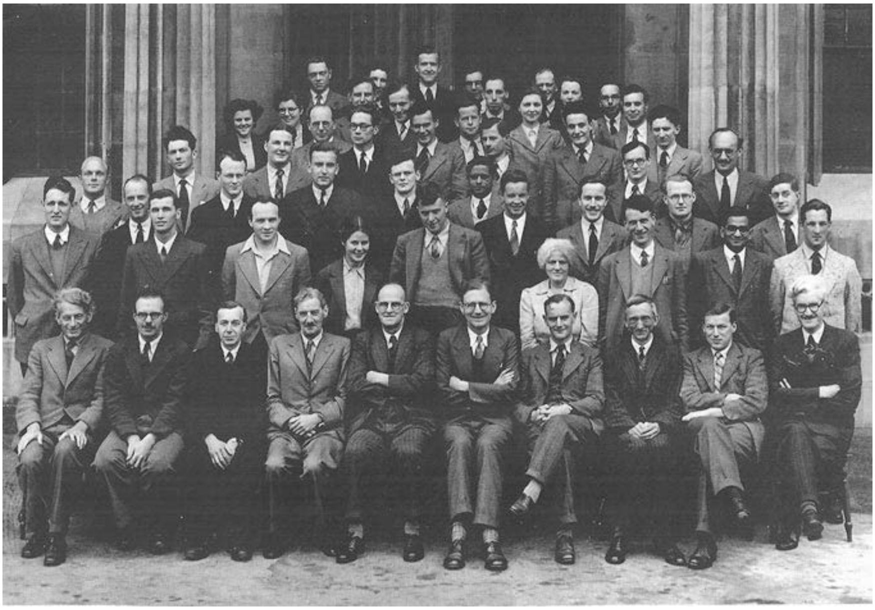

Fig. 4.11 Fröhlich (*LH* end of the front row) as a member of staff of the Physics Department in the late 1940s: Tyndall is 5th, and Mott is 6th from the *LH* end of the front row; Heitler is absent, having moved to the Dublin Institute for Advanced Studies in 1946—Reproduced with the permission of the Department of Physics, Bristol (Sect. 4.6)

Fig. 4.12 Fröhlich & Mott, many years later in 1985

Fig. 4.13 Fröhlich on leave from Bristol at the Dublin Institute for Advanced Study, 1947: *Back row*-Fröhlich (2nd from the *RH* end); others in this row include Born (3rd from the *RH* end), Schrödinger (2nd from the *LH* end). *Front row* Møller (2nd from the *LH* end), Heitler (3rd from the *LH* end), Conway (4th from the *LH* end), Jánossy (*RH* end). In the centre of the front row is the Irish President (Seán T O'Kelly) and his wife—Reproduced by courtesy of the Irish Press plc (Sect. 4.7)

Fig. 4.14 Fröhlich (*RHS*) and Huang (*LHS*) at a garden party at the H.H. Wills Physical Laboratory in Bristol, 1947—Reproduced with the permission of the Department of Physics, Bristol (Sect. 4.7)

Fig. 4.15 Fröhlich and his brother, Ali, in Bristol, sometime between 1945 and 1948 (Sect. 4.7)

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Chapter 5

The Liverpool Years: The First Professor of Theoretical Physics

5.1 From Bristol to Liverpool

In 1948, at Chadwick's instigation, and after long negotiations with the Vice Chancellor to ensure that he would have an effectively independent research institute with the promise of a well-funded theoretical physics library[1] under his own control, and minimal undergraduate teaching responsibilities, Fröhlich moved to Liverpool, to become, at the age of 42, the first Professor of Theoretical Physics (Fig. 5.1), a position for which, incidentally, Schrödinger had proposed himself in 1946.[2]

The Liverpool Chair had first been advertised by the university in May 1947, and applications were received from Benham, Jaeger, Jahn and Temperley. Although only Jahn and Temperley were invited for interview, Jaeger's application did receive serious consideration, partly on account of his work being known to the External Advisors (Mott and Peierls) and to certain members of the Selection Committee. Whilst it was agreed that all three merited careful consideration, it was finally concluded that none of them was likely to fully meet the needs of the subject in Liverpool. After discussion with the External Advisors as to whether an approach should be made to someone who had *not* applied for the Chair, they suggested that consideration be given to Fröhlich, who was already well-known to them; indeed, Mott, in his report to the Selection Committee, revealed that Fröhlich was likely soon[3] to be elected Fellow of the Royal Society. The Committee welcomed this proposal, and Fröhlich was duly invited to Liverpool for preliminary talks with the

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[1] The library soon grew to become the invaluable aid to research that he had foreseen, and upon his retirement from the Chair in 1973 the University granted it the title '*Fröhlich Research Library*' in his honour.

[2] Given Schrödinger's research interests at the time, which included meson theory, Chadwick was at first enthusiastic, but was eventually dissuaded from offering him the position on the advice of Rotblatt [P. Rowlands, personal communication, 2009].

[3] Fröhlich was first proposed in 1946, but was not elected until 1951 (vide Sect. 4.7).

G.J. Hyland, *Herbert Fröhlich*,
Springer Biographies, DOI 10.1007/978-3-319-14851-9_5

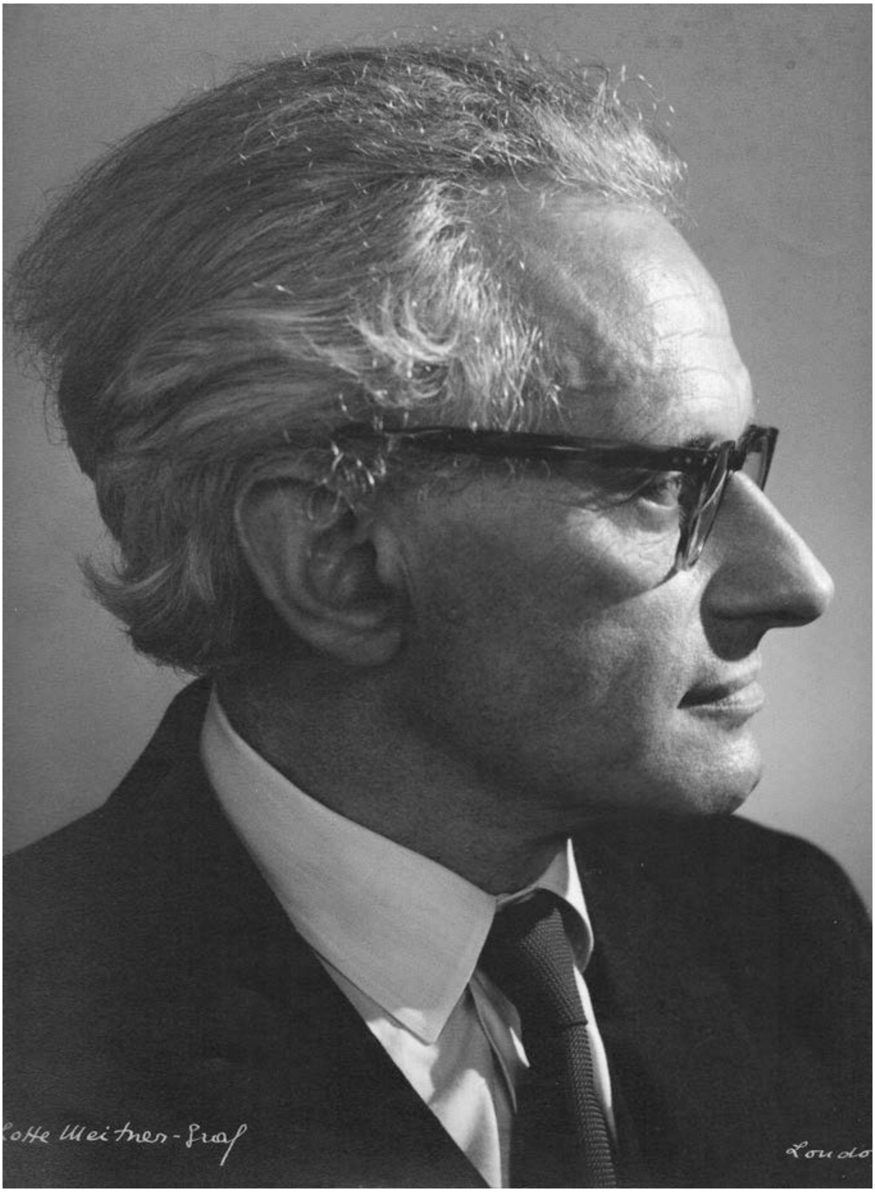
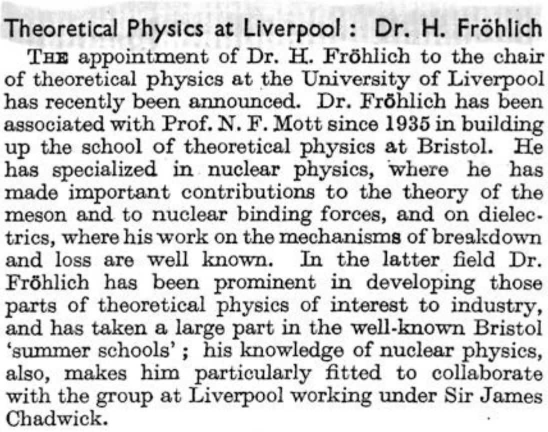


Fig. 5.1 Fröhlich at the time he was elected Fellow of the Royal Society, March 1951



Theoretical Physics at Liverpool : Dr. H. Fröhlich

The appointment of Dr. H. Fröhlich to the chair of theoretical physics at the University of Liverpool has recently been announced. Dr. Fröhlich has been associated with Prof. N. F. Mott since 1935 in building up the school of theoretical physics at Bristol. He has specialized in nuclear physics, where he has made important contributions to the theory of the meson and to nuclear binding forces, and on dielectrics, where his work on the mechanisms of breakdown and loss are well known. In the latter field Dr. Fröhlich has been prominent in developing those parts of theoretical physics of interest to industry, and has taken a large part in the well-known Bristol 'summer schools' ; his knowledge of nuclear physics, also, makes him particularly fitted to collaborate with the group at Liverpool working under Sir James Chadwick.

Fig. 5.2 Announcement in *Nature* (8 May, 1948) of Fröhlich's appointment to the Liverpool Chair of Theoretical Physics—Reproduced with the permission of Nature

Vice Chancellor and Chadwick. On 20 February 1948, in the absence of the External Advisors, he met the Selection Committee, several of whose members already knew him, and they were all convinced that, in view of his high standing, he would be a valuable and much needed contribution to the work of the university. Accordingly, the Committee unanimously recommended that Fröhlich be appointed to the Chair of Theoretical Physics as from 1 October 1948; the appointment was reported in *Nature* on 8 May that year (Fig. 5.2).

From Bristol and the *ERA*, he brought with him a substantial number of co-workers and research students, including H. Pelzer (a member of the Senior Research Staff of the *ERA*, and well-known as the co-author with Wigner of a seminal paper on the rates of chemical reactions), B. Szigeti (a Research Fellow supported by the *ERA*) who was highly experienced in dielectrics and crystal dynamics, A.B. Bhatia (*1851 Exhibition Scholar*), and Kun Huang (an *ICI* Fellow) who went to Edinburgh during the university vacations to work with Max Born on what was to become the well-known treatise '*Dynamical Theory of Crystal Lattices*', which was first published in 1954 (Born and Huang 1954). In addition there was S. Zienau, a research student working on electrons in polar crystals, and later (in 1950—vide Sect. 5.3), co-author, together with Fröhlich and Pelzer, of the famous paper [F72] in which the theory of what became known as the 'large' polaron was first formulated. The only staff member besides Fröhlich was his assistant lecturer, R. Huby. The first research students were:

Miss S.N. Ruddlesden and Mr A.C. Clark, both of whom had recently graduated from Liverpool (Huby 1950, 1988).

A distinguishing characteristic of his department, which, until 1959, occupied an elegant Georgian house at 6 Abercromby Square—and which was rather separate from the rest of the university—was the constant stream of eminent visitors from abroad, such as Bardeen, Bethe, Heisenberg, Heitler, Onsager and Prigogine, which made it a truly international centre of excellence in theoretical physics. To some in Liverpool, this was a source of some disquiet in the immediate post-war years when the visitors included both German and Japanese physicists, such as Hermann Haken and Sadao Nakajima.[4] To Fröhlich, however, racial, political and religious differences were no impediment to sincere scientific discourse, and in any case, as he pointed out, some of the younger people were still school-children during the war; appreciating how difficult it was for them, he was only too happy to be able to help. It was, politically, racially and religiously, a very heterogeneous department, with Arabs, Catholics, Communists, Hindus and Jews, but Fröhlich did his best to ensure that their irreconcilable differences were not a source of disharmony, actually, on occasion, forbidding discussion at coffee or tea of particularly sensitive issues, such as Arab-Israeli conflicts.

Indeed, morning coffee and afternoon tea were undoubtedly the most significant events of the day at which important ideas often emerged from extended and animated discussions, to which the congenial atmosphere of the house was so conducive; apparently, there was a reluctance to use reference books to settle a point, and to go and fetch was was highly unpopular because it broke up discussions! Despite this congeniality, surnames were always used, however—even Fröhlich's wife often referring to him simply as 'Fröhlich'; indeed, few knew what his initial *H* stood for: perhaps they thought it was Hamiltonian, which would not have been inappropriate (Powles 1973)! Departmental outings continued, as in Bristol, but now the preferred destination was usually North Wales.

In 1959, the department was relocated nearby in the new Chadwick Laboratory, on the top 3 floors[5] of its 8-storey tower, the stairs of which Fröhlich daily athletically climbed, two-at-a-time, until he broke a thigh-bone in the 1970s; Theoretical Physics became an independent department of the university three years later in 1962.

As far as his research students were concerned, Fröhlich's typical method of teaching was to toss out a few ideas, leaving the recipients essentially to sort things out for themselves, but giving the occasional pertinent, but often cryptic, suggestion as to how they might proceed—an approach to which the author can personally bear witness. He drew people into discussion by his enthusiasm for the subject, cultivating creative argument with colleagues—something that is rapidly disappearing today—and exercising his razor-sharp mind in constructive criticism, particularly on the occasion of the weekly theoretical physics seminars that were held every

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[4] A touching letter of appreciation and gratitude from Nakajima's father is reproduced in Appendix 2 at the end of this chapter; at this time (1954), Japan did not yet have much contact with the outside world.

[5] On the 8th floor were Fröhlich's office, that of his secretary, and offices for research students and visitors; the 7th floor contained the Seminar/Coffee room, together with offices, whilst the 6th floor contained the Theoretical Physics Library and more offices. Adjacent to the entrance to the tower, he had a reserved parking place, identified by a plaque bearing his name, which he used first for his much-prided Riley 2.5 Saloon, and later for his Rover 2000, which was one of the first in Liverpool.

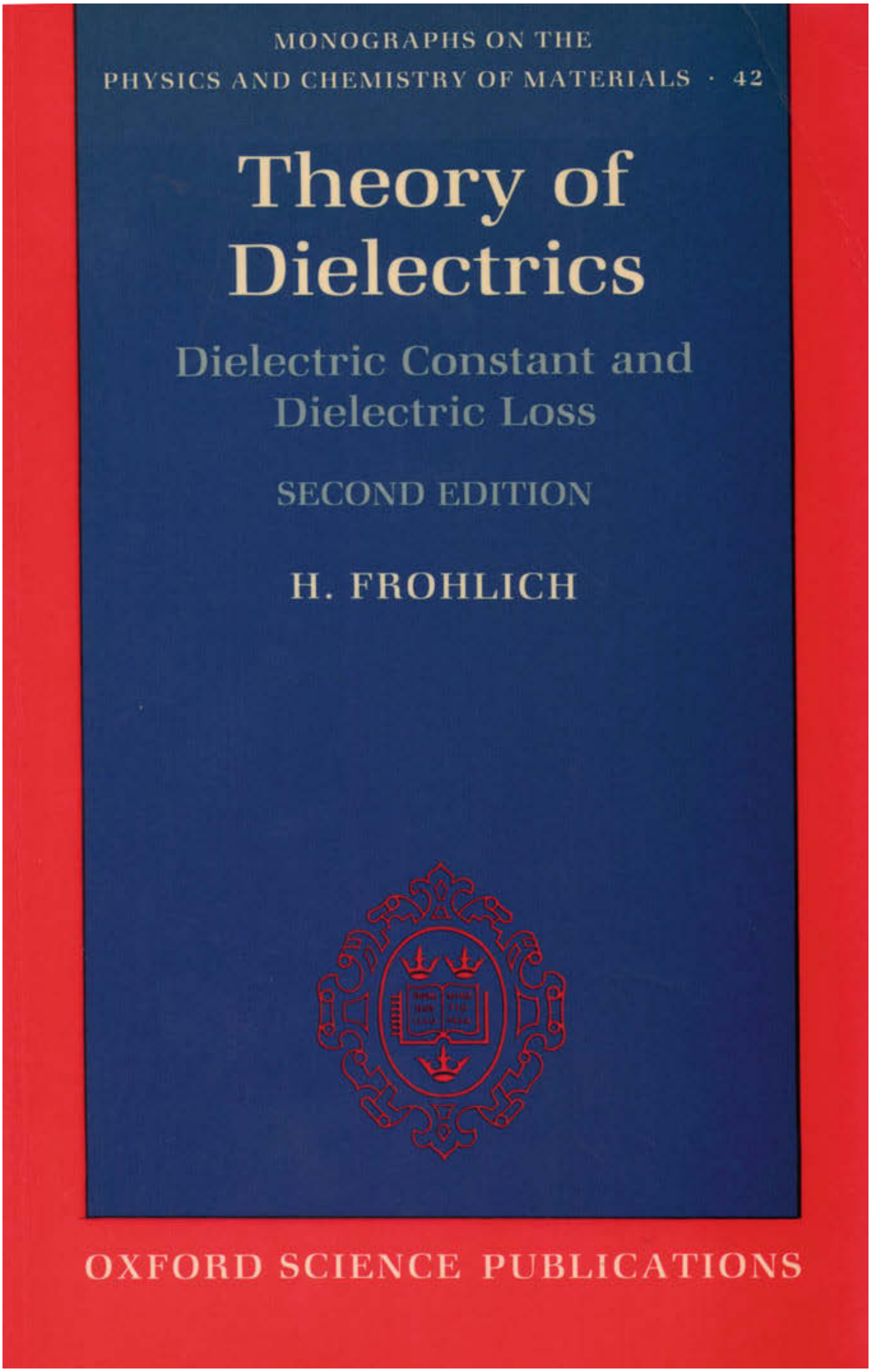
Thursday afternoon at 3.30 p.m. during the university terms. At the same time, he was always very approachable; he was recently described by a former colleague in Germany as ‘friendly, but very cunning!’

5.2 Theory of Dielectrics

Once settled in Liverpool, he completed his second book, *Theory of Dielectrics* [F(ii)], which was published by Oxford University Press in 1949, and which soon became the definitive text on the theory of dielectrics. The book was expressly written for use by applied scientists, including not only physicists but also chemists and biologists, in an attempt to fill a long-felt need for an up-to-date, authoritative, and systematic account of the theory of the dielectric constant and dielectric loss (but excluding dielectric breakdown). In addition, he hoped that it would act as a stimulus for further research, both theoretical and experimental (Fig. 5.3).

After a short introduction to the basic concepts and definitions used in macroscopic dielectric theory for both static and time-dependent fields, and the energy loss associated with the latter, together with some important remarks relating to the temperature dependence of the static dielectric constant, which follow from some quite general thermodynamical considerations (Chap. 1), the subsequent subject matter of the book divides naturally into detailed consideration of static and dynamic properties. The main topic of the former is the theory of the static dielectric constant (Chap. 2), whilst that of the latter concerns dielectric loss (Chap. 3). These are followed by a long chapter devoted to applications to a variety of dielectric materials, including liquids and gases. The book was republished in 1957 with an additional Appendix (B) dealing with the general theory of the dielectric constant, and contained some theoretical work on dielectric loss done subsequent to the 1st Edition. A survey of the theoretical situation at this time was given by Fröhlich in [F103].

Basic to the considerations of Chap. 2 is a recognition of the distinction between two quite different types of interaction in a dielectric, namely those of short range (associated with chemical bonds, van der Waals attraction, and various repulsive forces), and the long-range forces of a dipolar nature, which means that they must be taken into account even at macroscopic distances. The latter feature led Fröhlich to base his work on the Lorentz-Kirkwood method of treating dipolar interaction, namely: ‘*......from a macroscopic specimen select a microscopic spherical region which is sufficiently large to have the same dielectric properties as a macroscopic specimen. The interaction between dipoles inside the spherical region will then be calculated in an exact way, but for the calculation of their interaction with the rest of the specimen the latter is considered as a continuous medium*’ [F(ii), p. 22]. The most important section of Chap. 2 is Sect. 2.7 (based on work published the preceding year [F66]), in which the following expressions are derived for the static dielectric constant, ε_s, which hold *quite generally* for any dielectric material that is not permanently polarized:



MONOGRAPHS ON THE
PHYSICS AND CHEMISTRY OF MATERIALS · 42

Theory of
Dielectrics

Dielectric Constant and
Dielectric Loss

SECOND EDITION

H. FROHLICH

OXFORD SCIENCE PUBLICATIONS

Fig. 5.3 Fröhlich's 2nd book, *Theory of Dielectrics*, first published 1949—Reproduced with the permission of Oxford University Press

$$\varepsilon_s - 1 = \frac{4\pi}{3V} \frac{3\varepsilon_s}{(2\varepsilon_s + 1)} \frac{\langle M^2 \rangle}{kT} = \frac{4\pi}{3V} \frac{(\varepsilon_s + 2)}{3} \frac{\langle M^2_{\mathrm{vac}} \rangle}{kT} \tag{5.2.1}$$

where $\langle M^2 \rangle$ is the macroscopic fluctuation of the *spontaneous*[6] dipole moment of a sufficiently large sphere of dielectric material of volume V embedded in its own medium with dielectric constant ε_s[7]; $\langle M^2_{\mathrm{vac}} \rangle$ are the fluctuations in the case of a sphere in vacuum.

It was shown that when all short-range forces are neglected, and the molecules are assumed to be spherical, Eq. 5.2.1 yields either the Clausius-Mossotti or the Onsager formula, according as whether the molecules are non-polar or polar, respectively. Including short-range forces between nearest neighbours only, Eq. 5.2.1 was then used to obtain a generalisation of Kirkwood's formula for a polar liquid, which contains, for the case of spherical molecules, a corresponding generalisation of the Onsager formula. In turn, these generalisations permitted determination of the domain of validity of the original formulae (in which no account is taken of short-range forces), which have often been used indiscriminantly. It was found, for example, that for dipolar liquids the Onsager formula can be expected to hold only asymptotically at high temperatures.

Another topic treated in Chap. 2 was the temperature dependence of the static dielectric constant of a dipolar solid in the vicinity of an order-disorder transition, in which attention was drawn to the similarity in the dielectric behaviour of disordered solids and liquids.

The other main topic treated in the book (Chap. 3), which involved *dynamic* properties of the materials, was *dielectric loss*—i.e. the energy loss from an *A.C.* electric field due to the heating that occurs in a dielectric material. It was shown, in particular, that the familiar Debye equations for the real and imaginary parts of the complex, frequency-dependent dielectric constant, $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$, respectively, follow under the assumption that, in an external electric field, equilibrium is attained exponentially with time—i.e. $e^{-t/\tau}$, where the relaxation time, τ, in general depends on temperature; the associated dielectric loss is given in terms of ε_1 and ε_2 by $\tan\phi = \varepsilon_2/\varepsilon_1$. Again, Fröhlich stressed that the Debye equations '*have been applied to many substances, not always, unfortunately, with the necessary discrimination with respect to the intended range of validity*', and proceeded to obtain expressions for the relaxation time for dipolar solids and liquids, taking care to specify the conditions necessary for their validity.

In the case of a dilute solution of dipolar molecules in a liquid or an amorphous solid, generalisations of the Debye equations were derived covering a *range* of

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[6] *Spontaneous* here means that $\langle M^2 \rangle$ is a property of the material in the *absence* of a field.

[7] This work was later used [F92, F99] to identify the origin of an important mistake in an expression for the static dielectric constant given by Harris and Alder (1953).

relaxation times, and the associated absorption discussed. Finally, the case of *resonance* absorption was treated, in which the loss is due to displacements of charges bound elastically to an equilibrium position about which they oscillate with a frequency ω_o; near this *resonance* frequency, the power loss (and ε_2) is a maximum. In deriving the associated expressions for ε_1 and ε_2, it was assumed that equilibrium is approached through exponentially damped oscillations—i.e. $e^{-t/\tau}$ was replaced by $e^{-t/\tau}\cos(\omega_o t + \theta)$, where the phase θ was shown to be given by $\tan\theta = -\omega_o\tau$. It was found, in contrast to the case of Debye absorption, that the frequency of maximum absorption is here independent of temperature, whilst the resonance peak becomes narrower and higher with decreasing temperature. As noted in Sect. 4.6, Fröhlich had first considered dielectric loss already in 1941 [F36], and later in 1942 [F39] and 1945 [F51 Addendum], the latter being elaborated in detail in [F56]. In the latter work, he showed, for the case of a rigid dipole oscillating about an equilibrium position, that the usual Lorentz formula for the shape of absorption lines is incorrect except near resonance, and derived a generally valid expression for the loss angle, which must be used at ultra-high frequencies, and which takes the form of the familiar Debye expression at frequencies well above resonance. The new formula agreed with that derived independently by van Vleck and Weisskopf for a system of harmonic oscillators, using a different method. In [F57], Fröhlich applied his own method to van Vleck and Weisskopf's system, reproducing their result.

The final chapter (Chap. 4) was devoted to applications of the results of Chaps. 1 and 2 to a variety of dielectric materials, such as non-polar solids, dipolar liquids and, in particular, ionic crystals about which two extremely important points were made, which subsequently developed into independent subjects. The first (on pages 153–155), the origins of which can be traced to a footnote (vide Sect. 4.3) in a paper with Mott of 1939 [F23] and which was one that proved to be of particular importance much later on in the case of biological systems (vide Sect. 6.3), was the fact that, because the long wavelength polarization waves in an ionic crystal give rise to dipole-dipole forces of long range, these vibrations must depend on the *size* and *shape* of the sample. This dependence has the strongest repercussions in the optical properties of polar specimens whose dimensions are of the order of, or smaller than, the wavelength of the polarization waves, for which the polarization is effectively uniform throughout the sample, and distinction between longitudinal and transverse modes vanishes. Here, neglecting retardation effects, he showed that for a spherical specimen of a diatomic polar lattice the lowest energy mode is a triply degenerate one in which the polarization is uniform throughout[8] its volume (and is thus infrared active). The frequency, ω_s, of this mode was found to be *intermediate* between the longitudinal (ω_L), and transverse (ω_T) frequencies of the corresponding bulk crystal—i.e. a sample of the same material sufficiently large that its dimensions exceed the wavelength, and which absorbs light only at the frequency ω_T.

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[8] Despite this, it is sometimes called a *surface* mode, since its existence is due to the finite size of the sample; hence the *s* subscript on ω_s.

The increase of the absorption frequency to the value ω_s in the case of a 'small' sample is due to the polarization charge at its surface; ω_s is given in terms of the bulk transverse frequency ω_T by:

$$\omega_S^2 = \omega_T^2 \frac{(\varepsilon_s + 2)}{(n^2 + 2)} \tag{5.2.2}$$

In NaCl, for example, where $\varepsilon_s = 5.9$, $n^2 = 2.25$, $\omega_s = 1.36\omega_T$—*c.f.* $\omega_L = 1.62\omega_T$.

This important work by Fröhlich is interesting historically in that it complements the work of Mie, in whose institute in Freiburg Fröhlich was first employed, 1932–33—*see* Sect. 2.2. Mie (1908) showed that in the case of spherical *metallic* samples that are small compared to the wavelength of the light, the light absorption peak occurs at a frequency *between* zero and the plasma frequency, in contrast to the situation in a large metallic crystal where the peak absorption occurs at zero frequency. For both metallic and ionic spheres, ω_s is defined by $\varepsilon(\omega_S) = -2\varepsilon_M$, where $\varepsilon(\omega_S)$ is the appropriate dielectric function of the spherical sample and ε_M is the dielectric constant of the surrounding medium.

Apart from some early clarification and extension by Szigeti in an *ERA* Report (Szigeti 1951), this important work lay dormant theoretically for nearly 20 years before being taken up by others (Engelman and Ruppin 1968) who showed that, although not localised at the surface, Fröhlich's mode is actually the lowest frequency mode of a *series* of surface modes; in addition, they generalised the above to ionic samples of arbitrary shape, and investigated the effect of retardation[9] in diatomic crystals, making contact with *polaritons*—i.e. collective (transverse) excitations having both phonon and photon character, which result from solving the equations of motion for the lattice together with the Maxwell equations (Ruppin and Engelman 1968, 1970). A few years later, Genzel and Martin treated the case of small spherical samples embedded in a non-absorbing medium with dielectric constant ε_M, for which they derived (Genzel and Martin 1972, 1973) the following generalisation of Eq. 5.2.2:

$$\omega_S^2 = \omega_T^2 \frac{[\varepsilon_s(1-f) + \varepsilon_M(2+f)]}{n^2(1-f) + \varepsilon_M(2+f)} \tag{5.2.3}$$

where f is the fraction of the total sample volume occupied by the small spheres.

Experimentally, attention seems first to have been drawn to this frequency shift in 1966 in studies of the infrared dielectric dispersion of UO_2 and ThO_2 (Axe and Pettit 1966), who analysed their data using an expression for ω_s, which anticipated Eq. 5.2.3 for the case $f = 0$. For further discussion of the theoretical-experimental situation, vide Genzel and Martin (1972, 1973).

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[9] Prior to this, dating from the work of Huang in Liverpool (Huang 1951a), polaritons had been introduced only in the case of *infinite* crystals.

The second topic, which proved to be well ahead of its time, and and which was not pursued by others until some years later,[10] appeared on the final page of the book. It referred to 'the possibility of a permanent polarization of ionic crystals', associated with the static dielectric constant, ε_s, diverging to infinity as the transverse optic frequency, ω_T, tends to zero. This behaviour is implied by the following expression for ε_s in terms of n^2 and other ionic parameters quantities, which Fröhlich derived for the case of a diatomic polar lattice:

$$\varepsilon_s - n^2 = 4\pi[(n^2+2)/3]^2 \frac{(e^*)^2 N_o}{M_{red}\ \omega_T^2} \tag{5.2.4}$$

where the reduced ion mass M_{red} is given by $1/M_{red} = 1/M_+ + 1/M_{,-}$, N_o is the number of unit cells per unit volume and e^* is an effective ionic charge. He concluded the book with the following sentence: '*Investigations on these lines should be of importance in view of the properties of crystals like barium titanate. They have not been developed far enough, however, to be included in the present book*'. This proved to be particularly prescient, since the first conclusive experimental evidence of ferroelectric soft modes was not obtained until some 13 years later, using infra-red spectroscopy (Barker and Tinkham 1962), (Spitzer et al. 1962), and neutron spectroscopy (Cowley 1962) on strontium titanate.

With these confirmations of his prediction, Fröhlich returned to this topic in 1967 in his contribution [F127] to Debye's *Festschrift*, in which he also drew attention to another possibility associated with the vanishing of a transverse frequency—this time an *electronic* one—namely, the establishment of a metallic state. In his contribution [F126] to J.C. Slater's *Festschrift*, he had shown how this could occur as an excited state of a system whose ground-state is a Mott insulator—an insulator characterised not by a filled valence band of electrons, but by electrons that are *localised* in order to minimize their mutual Coulomb repulsion. Essentially, the increase in the number of (delocalised) conduction electrons produced by raising the temperature acts to screen the interaction of the remaining localised electrons whose binding energy is thereby reduced, making it progressively easier to create more carriers, so that the process becomes a cooperative one. Eventually the screening becomes so strong that the remaining localised electrons are delocalised—i.e. their transverse frequency vanishes—and the system undergoes a first order phase transition to a metallic state. The novel feature of the model, which became the subject of the author's Ph.D. thesis in 1965 (Hyland 1968), was that Coulomb correlations are responsible for both the Mott-insulating ground-state *and*, via screening effects, for its eventual transformation into a metallic state.

So useful did his *Theory of Dielectrics* prove to be that it was subsequently translated into Russian and Japanese, and was reissued as a paperback by *OUP* in 1986, when *Physics Today* commented: 'The presentation is admirable for its

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[10] Fröhlich's pioneering work was later acknowledged in Cochran's theoretical paper of 1960, dealing with crystal stability and the theory of ferroelectricity (Cochran 1960).

clarity…. All in all, it remains a masterly treatment of an engrossing subject'. Indeed, such was the significance of Fröhlich's many contributions in the field of dielectrics that these alone would have been sufficient to secure his international reputation, for in this field he was the undisputed master—so much so that to the *ERA* he became known as the 'wizard'! His first honorary degree, conferred by the University of Rennes (France) in 1955, was in recognition of his monumental work in dielectric theory.

During a visit to Zurich to speak on dielectrics, at the invitation of Scherrer who at the time was working on ferroelectrics, Fröhlich took the opportunity to look up Pauli whom he had known since the time of his pre-war collaboration with Heitler and Kemmer on nuclear forces, when they were in regular correspondence. To his dismay, Pauli told him that the subject of his lecture was a trivial one, and consequently he would not be attending. To this Fröhlich replied that, in his view, it was far from trivial, claiming that, in his opinion, more mistakes had been made in this field than in any other; he proceeded to remind Pauli that the Lorentz-Lorenz relation relation, for example, is often wrongly used, as he was well aware. After this, Pauli became more enthusiastic, and did in fact attend Fröhlich's talk that afternoon. At the end of the lecture, Scherrer, who was acting as chairman, asked Pauli if he would like to open the discussion, to which he replied:

> There have been more mistakes in this area than in any other theory. I myself already made one this morning, and have no intention of making another! (Fröhlich - personal communication 1983)

Another topic in this field, which was addressed some time later, concerned the energy loss of charged particles moving through a dielectric. In a joint paper [F86] with R.L. Platzman in 1953, attention was drawn to the close relation that exists between this and dielectric loss in an insulator, and a formula was derived for the rate of energy loss due to dielectric relaxation; this formula indicated that, for electrons with kinetic energy below the lowest electronic excitation potential, dielectric relaxation makes a substantial contribution to the total energy loss, which is comparable to that arising from transfer of vibrational quanta, a finding that has implications in radiobiology. The same methodology was later used [F96] to obtain a formula for the yield of secondary electrons from that of photo-electrons, and was developed further in his contribution [F105] to the *Max Planck Festschrift* in 1958, in which he gave a phenomenological theory of the energy loss of fast charged particles moving with an assumed constant velocity through a medium whose properties are described in terms of a complex dielectric function, ε, which, in general, depends on both frequency and wave-vector. The rate of energy loss was found to be proportional to the imaginary part of the *inverse* of ε, in contrast to the case of optical excitation where it is proportional to the imaginary part of ε; the two cases differ in respect of the fact that in the latter the exciting electric field is transverse, whilst in the case of a charged particle it is longitudinal. It was shown that energy loss for certain discrete values of frequency, ω, requires only that ε^{-1} has the form of a δ-function, or equivalently, that $\varepsilon(\omega) = 0$. Previous work

[F94, F95] with Pelzer[11] had shown that this condition is satisfied by longitudinal electric oscillations in solids, which include the case of plasma oscillations. The influence of plasma oscillations of the electron gas in a non-degenerate semiconductor on conduction was briefly considered in a short note [F100] published by Doniach in 1956, in which an estimate was given for the minimum electron density, n_{P}, at which the conduction electrons (moving with thermal velocities) can support plasma oscillations, above which an associated increase in electron mobility, μ, must be anticipated, as well as a departure from the usual $\mu \sim T^{-3/2}$ law for electron-lattice scattering. They noted that if the plasma would *completely* describe the electronic motion, then there would be *no* ordinary scattering at all. Their estimate of n_{P} took the following form (wherein ε is a background dielectric constant):

$$n_{\mathrm{P}} \approx (\varepsilon kT/4\pi e^2)^3 \mathrm{cm}^{-3} \tag{5.2.5}$$

n_{P} can easily satisfy $n_{\mathrm{P}} << n_{\mathrm{D}}$, where n_{D} is the density at which the electron gas becomes degenerate, when only relatively few electrons in a range of order kT about the Fermi surface undergo scattering by lattice vibrations, on which plasma oscillations then have little direct influence.

In passing, it is of interest to record the disappointment felt by some in the Liverpool physics department that, having been brought there by Chadwick—presumably on the strength of the significance of his contribution to the meson theory of nuclear forces—Fröhlich should now be preoccupying himself with something as non-relativistic and seemingly mundane as dielectric theory. It was, however, far from mundane, as he showed on many occasions; indeed, it actually spawned the so-called ‘dielectric’ approach to the many-body problem some 10 years later, towards the end of the 1950s and early 1960s, whilst the fluctuation-dissipation theorem (Callen and Welton 1951) was to a certain extent prefigured in his treatment of the static dielectric constant. Furthermore, in the light of Fröhlich’s later concern with the connection between micro and macrophysics (vide Sect. 6.2), this early interest in dielectric theory is particularly pertinent since the dielectric constant affords an example of a macroscopic quantity that itself straddles, so to speak, the gap between micro and macrophysics.

5.3 Polaron Theory

Around 1949, Fröhlich returned to consider an aspect of the interaction of a *slowly moving* conduction electron with the polarization field it gives rise to in an ionic (polar) crystal, which had been neglected in his 1939 paper with Mott [F23] referred

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[11] This work also showed that energy loss by fast charged particles in discrete steps of $\hbar\omega_{\mathrm{p}}$ (the plasmon quantum) can only be expected if $n\kappa/(n^2 + \kappa^2)^2$ has a sharp maximum at ω_{p}, where n and κ are the optical constants; this condition was shown to hold for the alkali metals and for silver.

to in Sect. 4.3, namely, the effect of the reaction of this polarization *back* on the electron.[12] The need for a theory that takes this into account had first become apparent in the course of his work on dielectric breakdown in amorphous materials [F49, 60], and from the following related consideration, which had been alluded to as early as 1939 in his critique [F24] of Seeger and Teller's formulation of von Hippel's criterion for breakdown (vide Sect. 4.1): in the case of a moving electron, only those parts of the lattice that are sufficiently far from electron (so that the latter executes only a small angle during the course of a lattice vibration) will see the electron as an effectively static charge. The angular distance travelled by an electron at a distance d in a time $1/\omega$ ($\sim$ the period of the lattice vibration) is $v/\omega d$; if this angle is to be small, then $v/\omega d \ll 1$, or equivalently $d \gg v/\omega$—i.e. the slower the electron, the shorter the distance beyond which the lattice will perceive it as stationary. The limiting value of d for which the electron appears static (and the polarization thus proportional to the Coulomb field of the electron) may thus be taken to be of the order of v/ω. Below this distance, the electron passes so quickly that its effect on an ion can be considered as a shock, exciting oscillation after it has passed.

As noted in Sects. 4.3 and 4.7, a slow (thermal) electron interacts most strongly with longitudinal polarization waves of *long* wavelength, $\lambda \gg$ the lattice constant, whilst its de Broglie wavelength similarly exceeds the lattice constant. Accordingly, the lattice can here be modelled as a dielectric *continuum*, described by high and low frequency dielectric constants ε_∞, and ε_s, and, in the simplest case, by a single longitudinal frequency ω_L.

The electric potential, $\Phi(r)$, in the dielectric continuum at a distance r from a *stationary* electron of charge e is given by:

$$\Phi(\boldsymbol{r}) = -(1/\varepsilon_s)e/r \qquad (5.3.1)$$

The static dielectric constant ε_s includes contributions not only from the displacement of ions from their equilibrium positions but also from their electronic polarizability.[13]

In the case of a slowly moving electron, the above considerations indicate that Eq. 5.3.1 holds only at $r > d$. Replacing r by the distance v/ω_L, yields the following net[14] energy of interaction, W, between the electron and the polarization field of the lattice, where the factor $(1/\varepsilon_\infty - 1/\varepsilon_s)$ ensures that only the inertial polarization associated with 'slow' ion displacements is included; the polarization associated with deformation of the electron shells of the ions (the resonance frequency of which is *far* above that which characterises the ion displacements—i.e. ω_L) does *not*

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[12] Recognition of the possible importance of this reaction can actually be traced back to von Hippel (1931, 1932), in connection with his ideas on the mechanism of dielectric breakdown—vide Sect. 4.1.

[13] These two contributions are *not* actually independent, since the former entails a certain amount of electronic displacement [F(ii), pp. 106, 151].

[14] The net interaction energy includes the energy required to polarize the lattice, and is reflected in the factor ½ in Eq. 5.3.2.

influence the energy difference between states of different itinerant electron velocity since, for sufficiently low velocities, it is always excited to its full value:

$$W \sim -\frac{1}{2}(1/\varepsilon_\infty - 1/\varepsilon_\mathrm{s})e^2\omega_\mathrm{L}/v \tag{5.3.2}$$

For small velocities, this potential energy term can exceed the electron's kinetic energy ½ mv^2, as noted by Fröhlich in the series of lectures [F55] entitled *Theoretical Physics in Industry*, which he gave at the Royal Institution in 1946, commenting: 'No detailed study of the influence of polarization on the motion of electrons has yet been made'.

In consequence of the quantum mechanical uncertainty principle, the above classical picture must be modified, however, as follows: associated with the electron's velocity, v, is an uncertainty in position, Δx, given by $\Delta x \Delta p \geq \hbar/2$, where p is the electron's linear momentum (mv); thus $\Delta x \geq \hbar/2mv$. This positional uncertainty must not exceed the distance d introduced above, for otherwise there would be no meaning to the idea of ions beyond this distance 'seeing' the electron effectively as stationary. For the limiting case $\Delta x = d, v = (\hbar\omega/2\mathrm{m})^{1/2}$, whence:

$$d = d_\mathrm{o} \equiv (\hbar/2m\omega_\mathrm{L})^{1/2} \tag{5.3.3}$$

and Eq. 5.3.2 must be replaced by:

$$W \sim -\frac{1}{2}(1/\varepsilon_\infty - 1/\varepsilon_\mathrm{s})e^2/d_\mathrm{o} \tag{5.3.4}$$

Thus d_o is effectively a measure of the 'size' that the (original point) electron assumes in the dielectric in consequence of its interaction with the polarization field of the material. Clearly, the validity of the adopted continuum representation of the lattice requires that $d_\mathrm{o} \gg a$, where a is the lattice constant, a condition that is usually fulfilled; in NaCl, for example, where $a \sim 5.6 \times 10^{-8}$ cm, $\omega_\mathrm{L} = 7 \times 10^{12}$ Hz, $d_\mathrm{o} = 2.8 \times 10^{-7}$ cm. This 'expanded' electron constitutes the 'polaron', and since, for the consistency of the approach, its size d_o must exceed the lattice constant, it is called a 'large' polaron. At $r \gg d_\mathrm{o}$, the polarization field, which is here proportional to the Coulomb field of the electron, can follow the electron's peregrinations through the dielectric continuum; this results in an increase in the mass of the electron to a value m^{**}, which is larger than the Bloch effective mass, m^* it would otherwise have.[15] At $r < d_\mathrm{o}$, however, the polarization is less than the classical value, and the

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[15] The Bloch effective mass, m^*, is a parameter in terms of which the effects of the periodic lattice potential on the dynamics of an electron can be incorporated, allowing the electron to be treated effectively as a free particle. m^* is intimately related to the electronic band structure, being positive (negative) near the bottom (top) of a band, and can be greater or less than the free electron mass, m. Roughly speaking, small values of m^* characterise wide bands and large values narrow bands, for which m^* is inversely proportional to the band-width.

response of the polarization field to the passing electron and, in turn, its self-energy are here governed by the dynamical properties of the polarization field.

In collaboration with Pelzer and Zienau, Fröhlich (Fig. 5.4) realised that this was the place for a field theory devoid of the singularities that had earlier plagued meson theory:

> I decided that electrons in ionic crystals was the place for a singularity-free field theory, and this led to the theory of what is now known as the 'large' polaron [F185].

For, as seen from the above semi-classical considerations, the nature of the interaction itself provides a natural cut-off—unlike the situation in the meson-nucleon case, where it had to be imposed artificially, thereby destroying the relativistic invariance of the theory.

The term, H_{int}, in the Hamiltonian describing the interaction of the slow electron with the polarization field of the lattice is essentially the same as that on which his earlier theory of dielectric breakdown had been based, but appropriately modified in respect the treatment of the crystal as a dielectric *continuum*,[16] characterised by a single longitudinal optic phonon frequency, ω_{L}, whilst to ensure that only the 'inertial polarization associated with ion displacements is included, the effective dielectric constant $(1/\varepsilon_\infty - 1/\varepsilon_{\mathrm{s}})$ of Eq. 5.3.2 is introduced. Equation 4.1.8 is thus modified as follows:

$$H_{\mathrm{int}} = -4\pi i e\left[\frac{\hbar\omega_{\mathrm{L}}}{8\pi V}(1/\varepsilon_\infty - 1/\varepsilon_{\mathrm{s}})\right]^{1/2}\sum_{\mathbf{w}} w^{-1}\left(b_{\mathrm{w}}\mathrm{e}^{\mathbf{iw\cdot r}} - b^*_{\mathrm{w}}\mathrm{e}^{-\mathbf{iw\cdot r}}\right) \qquad (5.3.5)$$

where b_{w} (b_{w}*) are related to the polarization, $\boldsymbol{P}$, by

$$\boldsymbol{P} = \left[\frac{\hbar\omega_{\mathrm{L}}}{8\pi V}(1/\varepsilon_\infty - 1/\varepsilon_{\mathrm{s}})\right]^{1/2}\sum_{\mathbf{w}}\frac{\boldsymbol{w}}{w}\left(b_{\mathrm{w}}\mathrm{e}^{\mathbf{iw\cdot r}} + b^*_{\mathbf{w}}\mathrm{e}^{-\mathbf{iw\cdot r}}\right), \qquad (5.3.6)$$

V is the volume of interest, e is the electron's charge, and $\boldsymbol{r}$ its position vector.

The prefactor of the summation in Eq. 5.3.5 can be rewritten in terms of a dimensionless coupling constant $\alpha\,(\equiv |W|/\hbar\omega_{\mathrm{L}})$ as $-i\,\hbar\omega_{\mathrm{L}}(d_{\mathrm{o}})^{1/4}(4\pi\alpha/V)^{1/2}$, where d_{o} is the 'size' of the polaron, as given by Eq. 5.3.3. With Eq. 5.3.4, α is given by

$$\alpha \equiv \frac{e^2}{2\hbar}(1/\varepsilon_\infty - 1/\varepsilon_{\mathrm{s}})\left(\frac{2m^*}{\hbar\omega_{\mathrm{L}}}\right)^{1/2} \qquad (5.3.7)$$

where m^* is the Bloch effective mass, which may be greater or less than m. α is analogous to the Sommerfeld fine-structure constant in quantum electrodynamics,[17]

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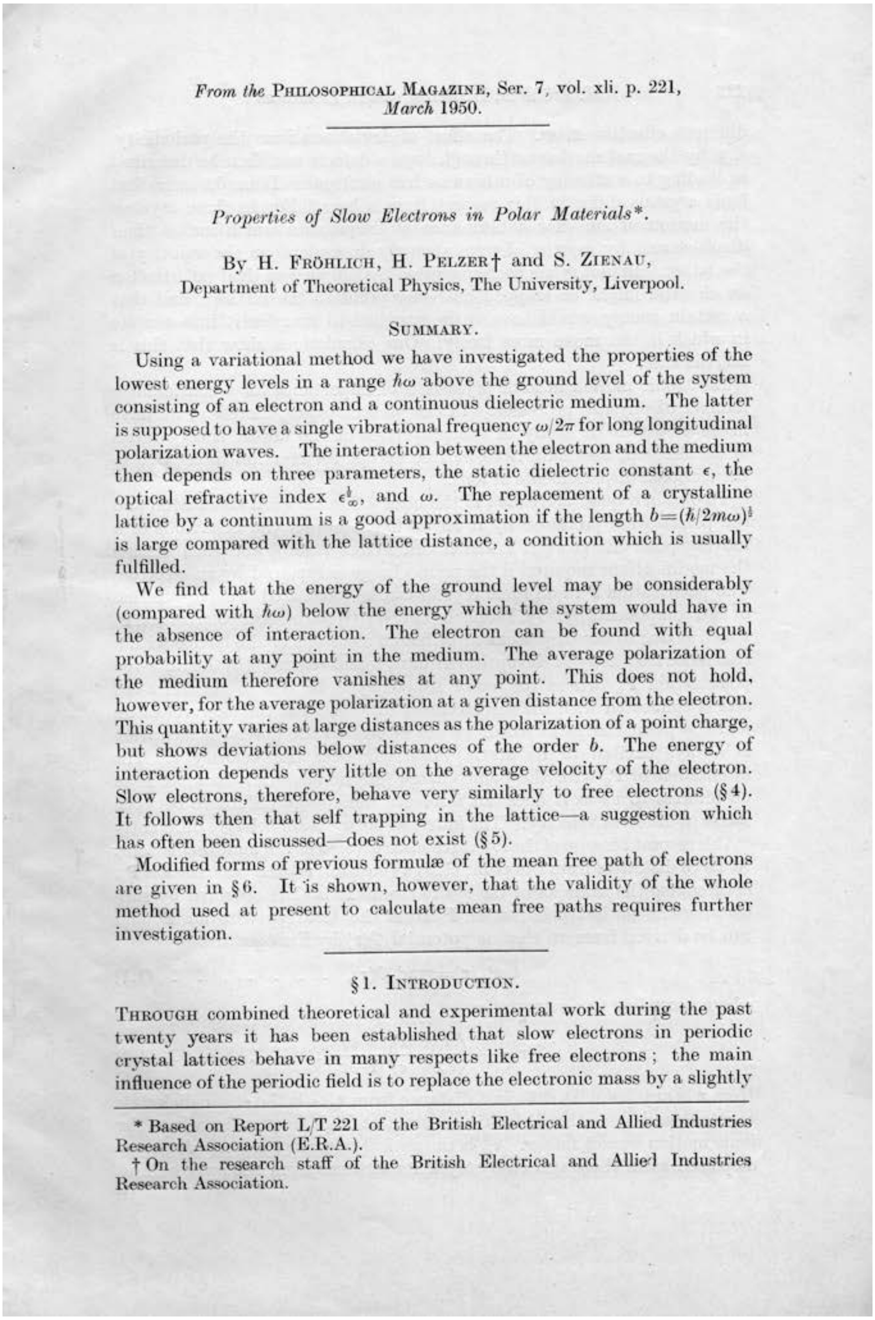
[16] This was done already in [F23]. In the case of the fast electrons involved in Fröhlich's original theory of dielectric breakdown, however, it was necessary to use a point charge model of the polar lattice, since such electrons interact predominantly with polarization waves of *short* wavelength.

[17] Sommerfeld's fine-structure constant determines the strength of the interaction between electrons and photons, and is defined by e^2/hc; its value is ~1/137.



From the Philosophical Magazine, Ser. 7, vol. xli. p. 221,
March 1950.

*Properties of Slow Electrons in Polar Materials**.

By H. Fröhlich, H. Pelzer† and S. Zienau,
Department of Theoretical Physics, The University, Liverpool.

Summary.

Using a variational method we have investigated the properties of the lowest energy levels in a range $\hbar\omega$ above the ground level of the system consisting of an electron and a continuous dielectric medium. The latter is supposed to have a single vibrational frequency $\omega/2\pi$ for long longitudinal polarization waves. The interaction between the electron and the medium then depends on three parameters, the static dielectric constant ϵ, the optical refractive index $\epsilon_{\infty}^{\frac{1}{2}}$, and ω. The replacement of a crystalline lattice by a continuum is a good approximation if the length $b=(\hbar/2m\omega)^{\frac{1}{2}}$ is large compared with the lattice distance, a condition which is usually fulfilled.

We find that the energy of the ground level may be considerably (compared with $\hbar\omega$) below the energy which the system would have in the absence of interaction. The electron can be found with equal probability at any point in the medium. The average polarization of the medium therefore vanishes at any point. This does not hold, however, for the average polarization at a given distance from the electron. This quantity varies at large distances as the polarization of a point charge, but shows deviations below distances of the order b. The energy of interaction depends very little on the average velocity of the electron. Slow electrons, therefore, behave very similarly to free electrons (§ 4). It follows then that self trapping in the lattice—a suggestion which has often been discussed—does not exist (§ 5).

Modified forms of previous formulæ of the mean free path of electrons are given in § 6. It is shown, however, that the validity of the whole method used at present to calculate mean free paths requires further investigation.

§ 1. Introduction.

Through combined theoretical and experimental work during the past twenty years it has been established that slow electrons in periodic crystal lattices behave in many respects like free electrons; the main influence of the periodic field is to replace the electronic mass by a slightly

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* Based on Report L/T 221 of the British Electrical and Allied Industries Research Association (E.R.A.).

† On the research staff of the British Electrical and Allied Industries Research Association.

Fig. 5.4 Fröhlich, Pelzer and Zienau's polaron paper [F72], 1950—Reproduced with the permission of Taylor & Francis except that the magnitude of α is here (via $\varepsilon_\infty, \varepsilon_s$ and ω_L) *material dependent*, entailing the possibility of both $\alpha < 1$ *and* $\alpha > 1$.

The ground-state of the Hamiltonian, H, of the coupled electron-lattice system,

$$H = p^2/2m + H_{\text{latt}} + H_{\text{int}} \tag{5.3.8}$$

was first studied in the weak coupling limit ($\alpha \ll 1$) in [F72] using 2nd order perturbation theory[18] in the context of which the polarization induced in the dielectric continuum by the electron and its reaction back on the electron is described, respectively, in terms of the virtual emission[19] and re-absorption by the electron of longitudinal polarization quanta; the parallel with the meson-nucleon system treated earlier [F20] is clear.

As anticipated in the above semi-classical considerations, these virtual processes endow the electron with a finite self-energy, E_o, and increase its Bloch effective mass, m^*, to the value m^{**}. Assuming the coupling is so weak that there is never more than one virtual quantum excited at any one time, perturbation theory yields the following results:

$$E_o = -\alpha\hbar\omega_L \tag{5.3.9}$$

$$m^{**}/m^* = 1/(1 - \alpha/6) \tag{5.3.10}$$

Unfortunately, however, most polar crystals do not satisfy $\alpha \ll 1$; in NaCl, for example, $\alpha \approx 6$. Accordingly, it was necessary to develop methods that could deal with the case of stronger coupling, where an *arbitrary* number of quanta are excited. The first to be tried, by M. Gurari[20] of Fröhlich's department (Gurari 1953), and independently by others (Lee et al. 1953), was a variational method, the validity of which required that correlations between successively emitted quanta induced by the electron's recoil could be neglected; this neglect is permissible provided the number of quanta ($\sim \frac{1}{2}\alpha$) is not too large, which imposes an upper limit on α—i.e. the method is geared to the case of so-called 'intermediate' coupling. The result for E_o is here found to be identical to that obtained in the weak coupling limit (but is now no longer restricted to $\alpha \ll 1$), whilst the form of m^{**}/m^* is identical to that obtained by expanding the weak coupling result to lowest order—i.e.

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[18] In first order, H_{int} leads to no change it the electron's ground-state energy since at $T = 0$ K there are no real polarization quanta; neither can such be emitted by a *slow* electron, since its kinetic energy is too low.

[19] The reverse sequence is impossible, since there are no real (thermal) polarization quanta at $T = 0$ K. A virtual process is one that violates energy conservation but on such a short time Δt that the energy discrepancy is within the associated energy uncertainty, $\Delta E \sim h/\Delta t$ given by quantum mechanics, where h is Planck's constant. Such processes arise in second order perturbation theory, where they take the form of transitions (in which momentum is conserved) to and from all possible so-called 'intermediate states' involving the emission/absorption of (virtual) quanta of some kind.

[20] His original name was Yisroel Aryeh Leib (http://mentalblog.com/2005/08/rebbes-brother-yisroel-aryeh-leib.html).

$$m^{**} = m^{*}(1 + \alpha/6) \tag{5.3.11}$$

The extreme case of strong coupling ($\alpha \gg 1$) was treated by Fröhlich himself [F90] using a variational trial wave-function of a form suggested by the work of Pekar[21] (1949). The resulting expression for the self-energy was found to be:

$$E_{\mathrm{o}} \sim -0.1\alpha^{2}\hbar\omega_{\mathrm{L}} \tag{5.3.12}$$

It should be noted that this expression is actually *independent* of frequency, ω_{L}, because of the ω_{L}-dependence of α (Eq. 5.3.7). The associated result for the effective mass was:

$$m^{**}/m^{*} = (1 + 0.02\alpha^{4}) \tag{5.3.13}$$

Thus, contrary to Landau's earlier prediction that an electron becomes *self-trapped* in the polarization field it itself produces—i.e. 'digs its own hole' (Landau 1933)—Fröhlich's strong-coupling analysis shows that this does *not* happen: m^{**}/m^{*} is finite, and the electron remains mobile (Fig. 5.5).

For NaCl, in which $\alpha \approx 6$, Gurari's result gives the lower self-energy, and is thus the better solution; indeed, as noted by Fröhlich 'it is not easy to find actual substances for which α is so large as to make [his] the better solution' [F90].

In strong coupling limit ($\alpha \gg 1$), the number of virtual quanta ($\sim\alpha^{2}$) is large, and correlations between successively emitted quanta predominate to such an extent that they result in the formation of a polarization potential well of radius $\sim d_{\mathrm{o}}/\alpha$ around the electron. This entails not only an enhanced effective mass (Eq. 5.3.13), but also an internal structure arising from the possibility of electronic excitation within the well. In this limit, the electron follows the zero-point fluctuations of the polarization field, in contrast with the situation in the weak coupling ($\alpha \ll 1$) where there is never more than one quantum excited at any one time, and where it is the lattice that follows the motion of the electron. Thus in both limits, in consequence of the recoil of the electron, its point charge is effectively spread over a finite volume of radius d_{o} for weak coupling, and of radius d_{o}/α for strong coupling. It is this that removes the necessity of introducing a cut-off, and automatically ensures a non-divergent self-energy. In both limits, consistency with the adopted continuum treatment of the crystalline lattice requires that the distances d_{o} and (d_{o}/α) be much *larger* than the lattice constant, which is why such polarons are often called 'large' polarons, as already mentioned in the semi-classical preamble.

Towards the end of [F90], Fröhlich pointed out that although the intermediate- and strong-coupling expressions for E_{o} yield the *same* self-energy near $\alpha \approx 10$, they did not match *smoothly* here, whilst the values of the polaron effective mass, m^{**},

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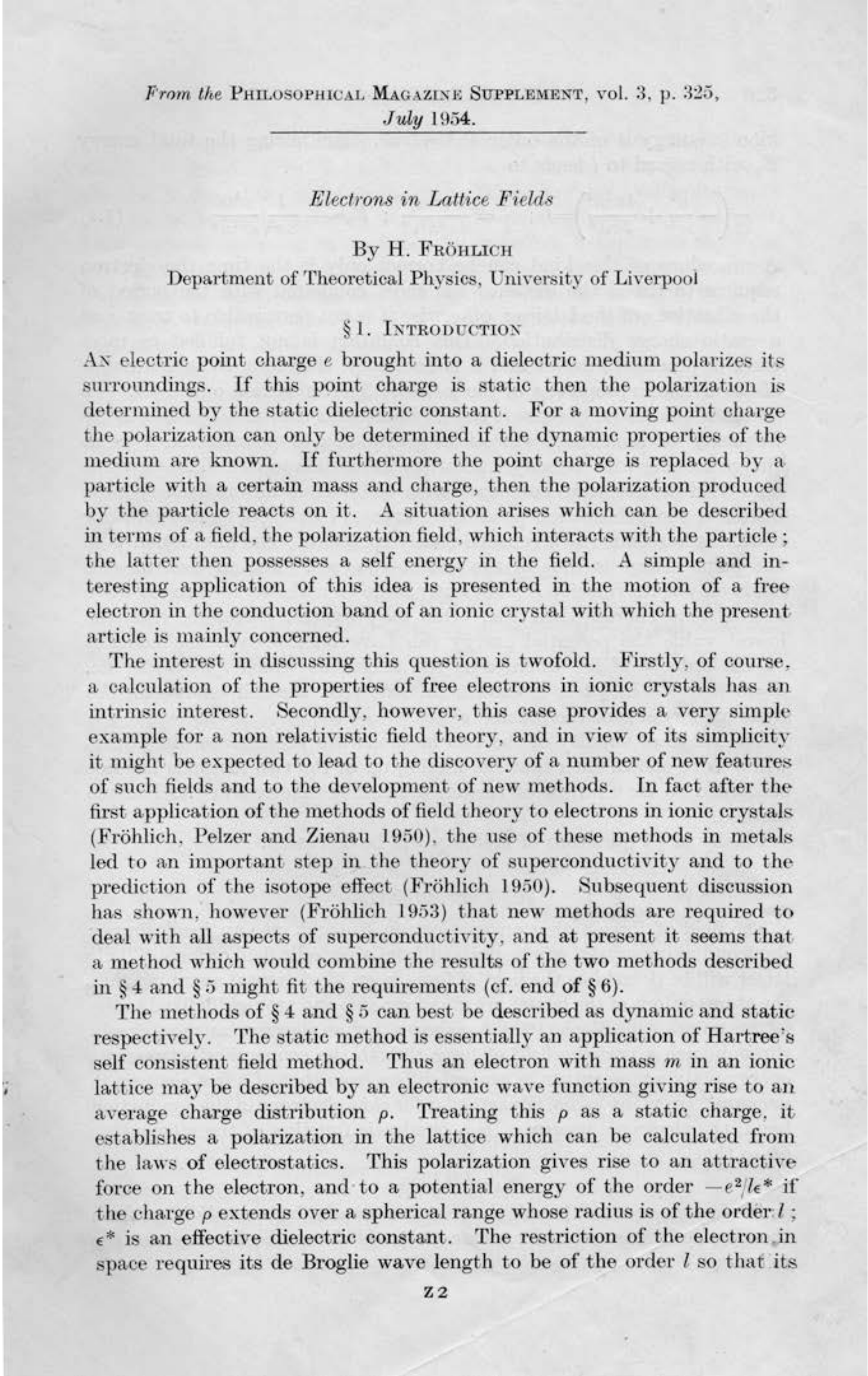
[21] It was Pekar who first coined the term 'polaron' in 1946 (Pekar 1946a, b).



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Electrons in Lattice Fields

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§ 1. INTRODUCTION

AN electric point charge e brought into a dielectric medium polarizes its surroundings. If this point charge is static then the polarization is determined by the static dielectric constant. For a moving point charge the polarization can only be determined if the dynamic properties of the medium are known. If furthermore the point charge is replaced by a particle with a certain mass and charge, then the polarization produced by the particle reacts on it. A situation arises which can be described in terms of a field, the polarization field, which interacts with the particle; the latter then possesses a self energy in the field. A simple and interesting application of this idea is presented in the motion of a free electron in the conduction band of an ionic crystal with which the present article is mainly concerned.

The interest in discussing this question is twofold. Firstly, of course, a calculation of the properties of free electrons in ionic crystals has an intrinsic interest. Secondly, however, this case provides a very simple example for a non relativistic field theory, and in view of its simplicity it might be expected to lead to the discovery of a number of new features of such fields and to the development of new methods. In fact after the first application of the methods of field theory to electrons in ionic crystals (Fröhlich, Pelzer and Zienau 1950), the use of these methods in metals led to an important step in the theory of superconductivity and to the prediction of the isotope effect (Fröhlich 1950). Subsequent discussion has shown, however (Fröhlich 1953) that new methods are required to deal with all aspects of superconductivity, and at present it seems that a method which would combine the results of the two methods described in § 4 and § 5 might fit the requirements (cf. end of § 6).

The methods of § 4 and § 5 can best be described as dynamic and static respectively. The static method is essentially an application of Hartree's self consistent field method. Thus an electron with mass m in an ionic lattice may be described by an electronic wave function giving rise to an average charge distribution ρ. Treating this ρ as a static charge, it establishes a polarization in the lattice which can be calculated from the laws of electrostatics. This polarization gives rise to an attractive force on the electron, and to a potential energy of the order $-e^2/l\epsilon^*$ if the charge ρ extends over a spherical range whose radius is of the order l; ϵ^* is an effective dielectric constant. The restriction of the electron in space requires its de Broglie wave length to be of the order l so that its

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Fig. 5.5 The paper 'Electrons in Lattice Fields' [F90], 1954—Reproduced with the permission of Taylor & Francis differ by a factor of 100; in other words, near $\alpha \approx 10$, *neither* approach affords a realistic description of the situation,[22] and he concluded that '......it seems desirable to develop a method which leads to a continuous transition between the results of the two methods'. For he believed that such a method would be useful not only in dealing with the polaron, but also possibly in connection with the more important problem of superconductivity. In connection with the latter, Fröhlich believed it to be particularly significant that, for $\alpha = 6$, intermediate-coupling theory (Eq. 5.3.11) predicted an increase in m^* by only a factor of 2, and that it would be worthwhile to investigate the extent to which this might continue to be the case when correlations between successively emitted quanta were included in the theory. It is of considerable interest to note, therefore, that Schrieffer told Fröhlich [F185] many years later that the form of the *Ansatz* for his many-electron extension of Cooper's pair wave-function—in which all pairs have the *same* total momentum (vide Sect. 5.4), and which was basic to the eventual solution of the problem by Bardeen et al. (1957)—had actually been motivated by the structure of the variational wave-function of the large polaron in the intermediate-coupling regime, as given by Lee, Low and Pines in 1953. An approach to finding superconducting wave-functions starting from the strong-coupling limit, on the other hand, was considered to be less appropriate since in this limit the dynamics of the lattice, which by this time were known to be involved in superconductivity in an essential way, are effectively suppressed, so that there is *no* isotope effect (vide Sect. 5.4).

It was Fröhlich's cryptic remarks about superconductivity at the end of [F90] that attracted[23] the attention of Feynman who took up Fröhlich's challenge to develop a theory that permits the transition from weak to strong coupling to be achieved in a *continuous* way. This he successfully did by adapting his own Lagrangian path-integral (variational) method that he had earlier used successfully in quantum electrodynamics,[24] which here permitted complete elimination of the phonons (Feynman 1955). The effect of this elimination was to replace the instantaneous form of the electron-phonon interaction given by Fröhlich's interaction Hamiltonian (vide Eq. 5.4.6) by a retarded interaction in which the electron interacts only with itself, in a way that is inversely proportional to the distance travelled from previous times, bearing with it the memory of its history—i.e. the electron behaves as though it were in a potential resulting from its electrostatic interaction with the average charge density at its previous locations. Unfortunately, however, the associated path integral could not be evaluated in closed form, and was thus approximated by one that could be so evaluated. Basic to the

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[22] The intermediate coupling approach is here undermined by its neglect of inter-quanta correlations, whilst α is too small for the strong-coupling approach to be justified.

[23] It must be remembered that, at this time, the solution to the problem of superconductivity by Bardeen, Cooper and Schrieffer was still 2 years in the future.

[24] To do this it was simply necessary to replace photons by phonons, since in *both* cases the field is describable in terms of harmonic oscillators whose interaction with the electron is linear in the field coordinates.

implementation of Feynman's programme was his modelling of the electron's interaction with this potential as an electron bound *harmonically* to a fictitious particle of finite mass that now mimicked the essentials of the electron's interaction with the lattice polarization.

In the extreme weak and strong coupling limits ($\alpha \rightarrow 0$, $\alpha \rightarrow \infty$), the following expressions were obtained for the self-energies:

$$E_{\mathrm{o}} \leq -\hbar\omega_{\mathrm{L}}[\alpha + 0.0123\,\alpha^2 + \mathrm{O}(\alpha^3)], \quad \alpha \rightarrow 0 \tag{5.3.14}$$

$$E_{\mathrm{o}} \leq -\hbar\omega_{\mathrm{L}}[\alpha^2/3\pi + 3ln2 + 3/4 + \mathrm{O}(1/\alpha^2)], \quad \alpha \rightarrow \infty \tag{5.3.15}$$

which, as he commented, are 'at least as accurate as previously known results' (Feynman 1955)—namely those given by the earlier weak and strong coupling theories, Eqs. 5.3.9, 5.3.12. In the range of intermediate coupling regime, however, numerical methods had to be used to evaluate the integrals yielded by Feynman's approach. This was done later by Feynman's collaborator, T.D. Schultz, for $\alpha = 3$, 5, 7, 9, 11 (Schultz 1959), and revealed the superiority of Feynman's self-energy values over the entire range of coupling.

The final section of his paper was devoted to consideration of the effective mass, m^{**}, of the polaron. Here, however, Feynman was unable to find an appropriate extension of his variational principle, which minimised the energy for *finite* polaron momentum, and which at the same time conserved momentum, and had to content himself with a non-rigorous treatment of the bound two-body system in terms of which he modelled the polaron; for low velocities, V, the self-energy was found to be augmented by a kinetic contribution ½ $m^{**}V^2$, via which m^{**} was defined. As in the case of the self-energy, an expression for m^{**}/m^{*} was obtained, which varied *continuously* with the coupling constant, α, although not being based on a variational principle, Feynman admitted it was difficult to adjudge the accuracy of the derived values, especially for large α (i.e. strong coupling).

How Feynman came to get involved with polaron theory is an amusing story that is told in a letter he sent to Fröhlich, which is reproduced, together with the latter's reply, in Appendix 1 at the end of this chapter. Apart from the possibility of adapting his approach to the polaron problem to the nucleon-meson case (which proved to be possible only for the unrealistic case of spinless nucleons and mesons (Mano 1955)), Feynman's real interest was, as just mentioned, in how his treatment of the polaron problem might suggest a strategy whereby the problem of superconductivity might be attacked; note the penultimate paragraph of his letter wherein he asks: 'What do we have to do to understand superconductivity?'

A valuable review of polaron calculational methods by G.R. Allcock, also of Fröhlich's department, appeared in 1956, in which Feynman's solution and other contemporary approaches to Fröhlich's challenge were considered in detail (Allcock 1956).

Powerful as Feynman's approach was in permitting the transition from weak to strong coupling to be treated in a *continuous* way, its utility was physically undermined by the progressive breakdown, with increasing α, in the validity of the underlying treatment of the ionic lattice as a dielectric continuum: a new approach was called for. In the West,[25] this was again initiated by Fröhlich in 1957, this time from considerations of Debye dielectric loss in ionic solids associated with trapped electrons [F101, *see also* F114, F118]. The naive interpretation of the observed loss is to assume that an electron can sit on positive ions in the neighbourhood of a trapping centre (effectively positively charged) thus forming an electric dipole with a number of possible directions of equal energy between which it can make transitions, thus giving rise to the loss. He pointed out that this interpretation is flawed because the various possible sites of the electron will combine to form various quantum states of which the ground-state is, in general, non-degenerate, thus making Debye loss impossible. He noted, however, that the naive interpretation can be retained if the energy levels of these electronic states have, at temperature T, a spread considerably *less* than kT (for then they could be superposed so as again to form *localised* states in a quasi-classical way); this energy spread is governed by the overlap of the electronic wave-functions on neighbouring ions. Fröhlich now crucially realised that when the *displacements* of the ions in the vicinity of the positive ion on which an electron is assumed to be localised are taken into account, the overlap is *reduced* from the value ($\gg kT$) appropriate to a rigid lattice by a factor $\exp(-\varDelta^2/x_o^2)$, where $\varDelta$ is the ion displacement and x_o its zero-point amplitude; this factor is a very sensitive function of $(\varDelta^2/x_o^2)$, and can take very small values, thus reducing the overlap to less than kT.

These ideas were subsequently developed by G.L. Sewell (1958), for the case of a diatomic polar lattice, into what is now known as 'small' polaron theory, in which the interaction of an electron with the ionic displacements is again built in from the start. The electron is treated in the tight-binding approximation (as is appropriate in the case of narrow bands, such as *d*-bands, for example), the eigenstates of the system being constructed as linear combinations of localised states in which the electron is bound to one of the positive ions, into which are now included the displacement of neighbouring ions caused by the electron.[26] The energy levels corresponding to these eigenstates form a band—the so-called 'small' polaron band—whose width is determined by the overlap integral governing the transfer of the electron, *together with its accompanying lattice displacements*, between neighbouring positive ions. These accompanying lattice displacements make the motion of the electron more sluggish, which is reflected in an exponential increase in the (rigid lattice) Bloch effective mass, m^* to the small polaron effective mass, m^{**}—or, equivalently, to an exponential decrease in the Bloch bandwidth, W_o to the small polaron bandwidth, W, given by:

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[25] Around the same time, the problem was considered independently in Japan (Yamashita and Kurosawa 1958).

[26] A somewhat similar strategy can be discerned in a remark made by Fröhlich in the discussion (on p. 1186) of a paper by Bardeen at the International Conference on Electron Transport in Metals and Solids, in Ottawa in 1956, in which he pointed out that a better treatment of a displaced atom or impurity can be obtained by redefining the unperturbed wave-functions (Bardeen 1956).

$$m^{**} = m^{*}\exp(\gamma) \tag{5.3.16}$$

$$W = W_{o}\exp(-\gamma) \tag{5.3.17}$$

The factors $\exp(\pm\gamma)$ arise as a result of the transfer from ion to ion of the lattice displacements that accompany the electron; γ can be written as:

$$\gamma = f(T)\gamma^{o} \tag{5.3.18}$$

where $f(T)$ is an *increasing* function of temperature, reflecting the fact that the random thermal motion of the ions opposes the transfer of their displacements, and γ^{o} is the value of γ at $T = 0$ K; for the simple model used, which is based on localised *s*-states and an optic mode frequency that is independent of wavelength, γ^{o} is typically about 15, when screening effects arising from the electronic polarizability of the ions are taken into account in terms of the high-frequency dielectric constant ε_{∞}.

The width of the small polaron band thus decreases with increasing temperature, and once it becomes less than kT extended states can no longer be defined. The electron then becomes localized in a self-trapped state, as originally envisaged by Landau (1933), but now with the important difference that this occurs *only* at *finite* temperatures. Since this localized state is stabilised by displacement of the surrounding ions, movement of the electron to a neighbouring site by tunnelling is possible only if the *same* lattice displacements can somehow be realised there so that this site is rendered *degenerate* with that at which the electron is initially localised. This can be achieved with the help of thermal vibrations (in particular, those belonging to the acoustic mode), and such thermally assisted motion (a *multi*-phonon process) is known as 'hopping' or 'jumping'. This kind of transport is characterised by an extremely low mobility, μ, that is activated—i.e.

$$\mu = \mu_{o}e^{-E_{a}/kT}, \tag{5.3.19}$$

where the activation energy, E_a is the energy required to recreate at the neighbouring site the lattice deformation that traps the electron initially.

It must be stressed that because of certain simplifying assumptions made (such as the band being based on localised orbitals that are *s*-states), the results are not quantitatively applicable to real narrow band materials in which the orbitals of relevance (such as *d*- or *f*-states) are highly anisotropic and in general degenerate; qualitatively, however, the band narrowing predicted on the simple model used can still be anticipated. The subject of the hopping mobility of small polarons was subsequently investigated in more detail by Lang and Firsov (1963) in a form that can be applied to real materials, such as the technologically important nuclear fuel UO_{2+x}, in which the carriers are (5*f*) holes in the Mott-insulating ground-state of the stoichiometric material (Casado et al. 1994).

The implications of the uncertainty principle on the mobility of carriers in narrow bands, such as those associated with small polarons, and, in particular, its temperature dependence, was the subject of a short note with Sewell in 1959 [F104];

it was estimated that the transition to hopping occurs at mobilities less than about 0.1 cm^2/s V. Sewell revisited the problem in a more extensive article published a few years later (Sewell 1963), as did Fröhlich himself in his contribution to the *Festschrift* for G. Busch [F135] in which he drew attention to some frequently occurring mistakes in the (then) current literature on small polaron transport. It should be noted that Fröhlich's initial work in this area predated Holstein's formulation of small polarons for the case of a molecular lattice where the electron-lattice interaction is of *short* range (Holstein 1959), which became better known than Sewell's treatment for an polar lattice, published the previous year.

Somewhat later, Fröhlich developed a better way of proceeding in the case of low mobility, narrow band materials by constructing wave-functions in which the electron is based on the *oscillating position* of a positive ion, rather than on a (fixed) lattice point, as had been done previously, since, in this way, the electron's movement is facilitated [F117, 118]. By following the ion's oscillation, not only is a considerable amount of the electron-ion interaction already included in lowest order—thereby leaving a relatively small residual electron-phonon interaction to be dealt with—but also the necessity of including *inter*-band transitions is avoided [F125].

Out of this evolved an associated modification of the usual tight-binding method in which an extended Bloch state is based on a superposition of localised wave-functions. This was elaborated in considerable detail by one of Fröhlich's doctoral students, T.K. Mitra, who showed, using localised wave-functions of the above kind in which the electron follows the ion motion adiabatically, that the electron-phonon matrix element governing intra-band transitions is proportional to the bandwidth, and is thus small in the case of narrow bands (Mitra 1969, 1978). The implications of this finding for superconductivity in materials with incomplete narrow energy bands will be considered in the next chapter in Sect. 6.1.

5.4 Theory of Superconductivity, the Introduction of Quantum Field Theory into Solid-State Physics, and Marriage

Important as polaron theory was in its own right, it was for Fröhlich, simply a testing ground for the non-relativistic application of field-theoretical methods, prior to bringing them to bear on the problem of superconductivity in metals, where, of course, *many* electrons have to be considered, and where the relevant lattice vibrations are acoustic, rather than optical. It must be remembered that, in 1950, superconductivity was *the* central problem in solid-state physics—a problem that had continued to defy solution since the advent of quantum theory, despite the efforts of the best minds, including several Nobel Laureates, such as Einstein, von Laue and Heisenberg. At that time, it was generally considered that the phenomenon involved some, yet to be discovered, novel collective feature of the Coulomb interaction between the conduction electrons. Such a consideration was not unreasonable, given that the success of the free-electron model of metals, in which the Coulomb interaction between electrons is neglected, was not then understood. Indeed, it did not become so until the work of Bohm and Pines, which showed that the Coulomb interaction between electrons can be divided into two parts: a long-range part, whose effect is described by longitudinal plasma oscillations, and a residual short-range 'screened' interaction, whose range is about 1 Å (Bohm and Pines 1953). Owing to the large excitation energy of the plasma oscillations, the long-range part can be ignored in many calculations, which explains the success of the free electron model in the case of many[27] metals.

To Fröhlich, however, an approach to the problem of superconductivity in terms of the Coulomb interaction seemed ill-founded, given the extremely small experimental energy difference, δE, between the superconducting and normal states of the order of 10^{-3}–10^{-4} eV per atom, which is minute in comparison with typical Coulomb energies, which are of the order of eV. Accordingly, he formed the opinion that the main problem in developing a theory of the superconducting state was first to find an interaction of the correct magnitude. The fact that such a dramatic effect as superconductivity is characterised by such a *small* energy difference is surely indicative that the difference between the normal and superconductive states is a highly subtle one, and that any understanding of it must be expected to require considerable ingenuity. In this connection, the small energy ms^2 (where m is the mass of an electron and s the speed of sound in a metal), which he knew [F81] played a role in some aspects of the theory of the electrical conductivity of normal metals, suggested itself in consequence of its small size, of the order of 10^{-5} eV. The involvement of the speed of sound focussed attention on the *dynamics of the metal lattice*, since the speed of sound is proportional to $M^{-½}$, where M is the mass of an ion; furthermore, ms^2 is the product of an electron parameter (its mass, m) and a lattice characteristic (the speed of sound, s), suggesting an underlying *electron-lattice interaction*—a suggestion that is consistent with the empirical finding that it is *poor* conductors that become superconductors. For from the work of Bloch, it was known that electrical resistivity is due to the scattering of conduction electrons by thermally excited lattice vibrations, or phonons—i.e. is due to the electron-phonon interaction. Field-theoretically, this scattering expresses itself as absorption/emission of such real phonons by the electron, which give non-zero contributions in 1st order of perturbation theory.

From his experience with field theoretical methods, both relativistic and non-relativistic, Fröhlich realised that the same interaction entails, in addition, *other* possibilities connected with the *virtual*[28] emission and absorption by an electron of phonons. These processes arise in 2nd order perturbation theory, and, in the case of

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[27] These metals are those with conduction bands *sufficiently wide* that an electron's itinerant kinetic energy dominates the potential energy associated with the residual short-range part of the Coulomb repulsion, the dominant contribution to which arises when, in the course of its band motion, the electron arrives at an ion where *another* conduction electron is already present.

[28] A virtual process is one in which violates energy conservation but on such a short time Δt that the energy discrepancy is within the associated uncertainty, $\Delta E \sim h/\Delta t$ given by quantum mechanics, where h is Planck's constant. Such processes arise in second order perturbation theory,

a single electron, had been the basis of polaron theory in the weak coupling limit. Similarly, in the case of a metal at absolute zero where there are no thermal lattice vibrations (real phonons) to scatter an electron, the Coulomb field of an electron can again itself cause a dynamic disturbance in the lattice by attracting positive ions in its vicinity, thereby creating a local increase in the density of positive charge. This, in turn, reacts back on the electron, lowering its energy—i.e. the electron acquires a polaron-like self-energy. In addition, however, and in contrast to the polaron case, there is here *another* effect arising from an influence of the lattice distortion on the motion of *other* conduction electrons in its vicinity—i.e. there is a dynamic *interaction* between electrons, which is transmitted by the dynamics of the lattice. Fröhlich's novel hypothesis was that it is this interaction that perhaps underlies the superconducting state—an hypothesis based on his realisation that Bloch's theory of electrical conductivity (Bloch 1928) was simply *one* aspect of a field theory; another, hitherto unrecognised complementary aspect was that the *same* electron-lattice interaction entails a novel *interaction* between electrons, mediated by the zero point dynamics of the lattice.

Given the success of the free electron model in describing the properties of the non-superconducting state of a metal, Fröhlich assumed that this continues to be the case as far as the direct Coulomb interaction between electrons is concerned, and thus focussed his attention solely on the electron-phonon interaction.

His first attempt to study this interaction used perturbation theory to calculate the associated energy change, using Bloch's matrix elements appropriate to absolute zero where there are only zero-point phonons. To first order, there is no energy change on account of the absence of thermal phonons, the first non-zero contribution, ΔE, arising in *second* order from virtual processes of emission and absorption of zero-point phonons by the electrons[29]:

$$\Delta E = -2 \sum_{\mathrm{k}} \sum_{\mathrm{w}} \frac{|M_{\mathrm{w}}|^2 f_{\mathrm{k}} \left(1 - f_{\mathrm{q}}\right)}{\varepsilon_{\mathrm{q}} - \varepsilon_{\mathrm{k}} + \hbar s w} \tag{5.4.1}$$

where M_{w} is the matrix element for emission of a vibrational quantum of wave-vector w by an electron of wave-vector $\boldsymbol{k}$, which (to conserve momentum) then makes a transition to an intermediate state with wave-vector $\boldsymbol{q}$ $(= \boldsymbol{k} - \boldsymbol{w})$ from which it is re-absorbed into the original state. To satisfy the Pauli principle, the transition must be proportional to the probability f_{k} (≤ 1) that the state $\boldsymbol{k}$ is occupied, and to the probability $(1 - f_{\mathrm{q}})$ that the state $\boldsymbol{q}$ is empty. At $T = 0$ K, where there are no thermal (real) phonons, $|M_{\mathrm{w}}|^2$ is given by Bloch (1928):

(Footnote 28 continued)
where they take the form of transitions (in which momentum is conserved) to and from all possible so-called 'intermediate states' involving the emission/absorption of (virtual) quanta of some kind.

[29] This is the quantum mechanical counterpart of the establishment of the lattice distortion by one electron (emission) and the influence of this distortion on either the same, or a *different* electron (absorption).

$$|M_{\mathrm{w}}|^2 = 4C^2\hbar w/9nVMs^2 \tag{5.4.2}$$

where V is the volume, M is the ion mass, n their number per unit volume, s is the speed of sound, and C is an interaction constant having the dimension of energy (and is of the order of 10 eV, similar in magnitude to the Fermi energy, E_{F}). It was found convenient to introduce an associated dimensionless constant, F, of the order of magnitude of unity, defined by:

$$F = C^2/3E_{\mathrm{F}}Ms^2 \tag{5.4.3}$$

Equation 5.4.1 contains the two effects described in the above semi-classical picture—namely, a polaron-like self-energy, ΔE_1, arising from the term linear in f, whilst the term bilinear in f can formally be interpreted as an *interaction* between two points in momentum space occupied with densities f_{k} and f_{q} (Fig. 5.6).

It should be appreciated, however, that perturbation theory treats the electron-phonon interaction as instantaneous, which it is not. For since the exchanged phonon moves with the speed of sound, s, which is much less than the Fermi velocity, v_{F}, of a conduction electron, the phonons trail behind the electrons: thus a phonon absorbed by an electron at position $\boldsymbol{r}$ at time t will have been emitted by another electron in a *different* location, $\boldsymbol{r}'$ at the *earlier* time t', given by:

$$t' = t - |\boldsymbol{r}' - \boldsymbol{r}|/s \tag{5.4.4}$$

The interaction thus depends not only on the inter-electronic separation $|\boldsymbol{r}' - \boldsymbol{r}|$, but also on $(t - t')$—i.e. it is a retarded interaction.[30]

Whilst the parallel with quantum electrodynamics is clear, they differ in that, in vacuo, the latter yields an effectively instantaneous interaction[31] that is *always* repulsive, *unlike* the present case, where the algebraic form of the energy change due to the phonon-mediated electron-electron interaction given by 2nd order perturbation theory indicates that the interaction is actually *attractive* only for electrons whose energy difference is *less* than the energy of the exchanged lattice quantum, under which condition the lattice follows the electrons. The attractive interaction is thus of a highly *dynamic* nature, depending on the momenta[32] of the two electrons and on the ability of the lattice to dynamically respond to their Coulomb field. For electrons whose energy differs by more than the energy of a lattice quantum, which

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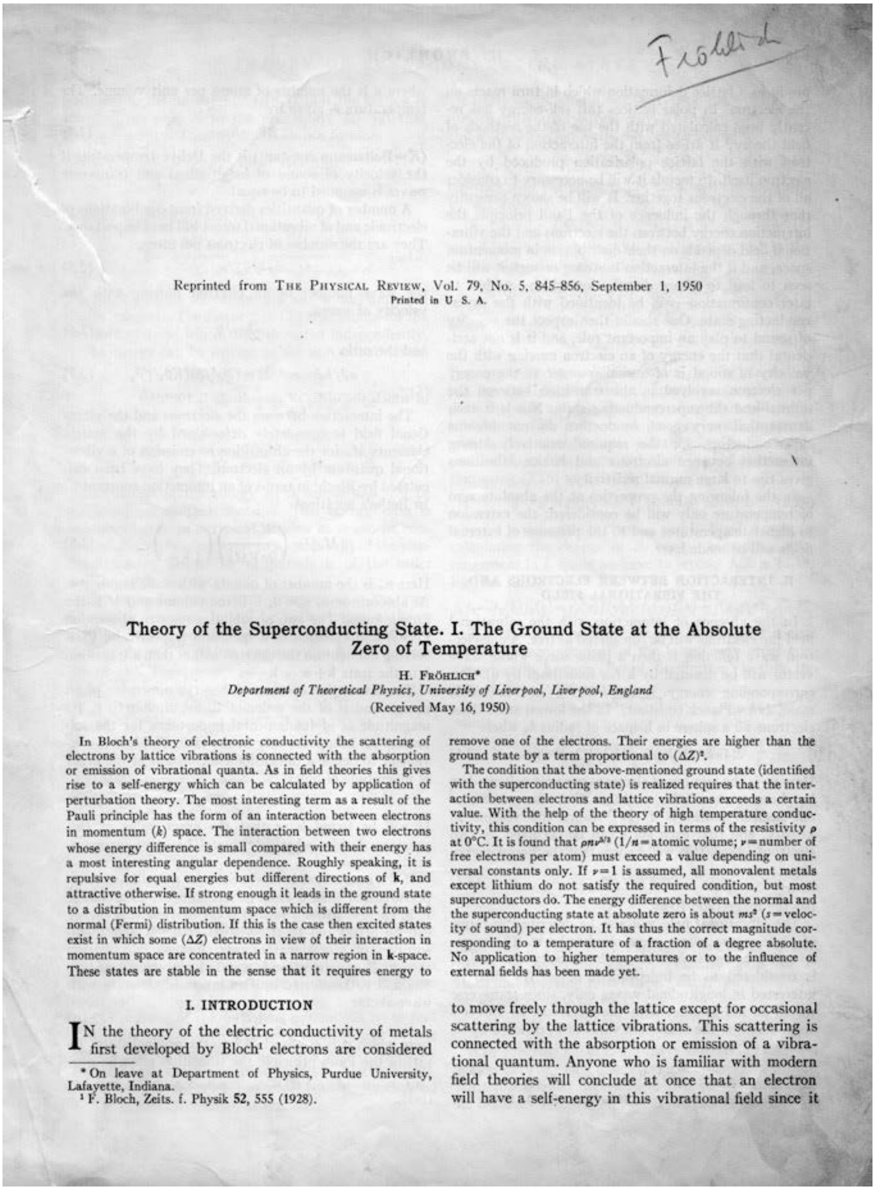
[30] Retardation effects are important in the case of so-called ‘strong-coupling’ superconductors, such as Pb (Eliashberg 1960).

[31] In media, on the other hand, with a refractive index, n, greater than unity, the speed of light is reduced, making it possible for a charged particle to here travel superluminally ($v > c/n$), and emit Čerenkov radiation; this is perhaps the closest parallel to the electron-phonon case, where $v_{\mathrm{F}} > s$.

[32] Since the phonon mediated electron–electron interaction depends of the *momenta* of the interacting electrons, as well as on their spatial separation, the interaction is actually *non*-local. This aspect was considered by Fröhlich many years later [F157] in connection with deriving the basic results of the *BCS* theory *without* solution of the many-body problem (vide Sect. 6.2).



Fröhlich

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Theory of the Superconducting State. I. The Ground State at the Absolute Zero of Temperature

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(Received May 16, 1950)

In Bloch's theory of electronic conductivity the scattering of electrons by lattice vibrations is connected with the absorption or emission of vibrational quanta. As in field theories this gives rise to a self-energy which can be calculated by application of perturbation theory. The most interesting term as a result of the Pauli principle has the form of an interaction between electrons in momentum (k) space. The interaction between two electrons whose energy difference is small compared with their energy has a most interesting angular dependence. Roughly speaking, it is repulsive for equal energies but different directions of $\mathbf{k}$, and attractive otherwise. If strong enough it leads in the ground state to a distribution in momentum space which is different from the normal (Fermi) distribution. If this is the case then excited states exist in which some (ΔZ) electrons in view of their interaction in momentum space are concentrated in a narrow region in $\mathbf{k}$-space. These states are stable in the sense that it requires energy to remove one of the electrons. Their energies are higher than the ground state by a term proportional to $(\Delta Z)^2$.

The condition that the above-mentioned ground state (identified with the superconducting state) is realized requires that the interaction between electrons and lattice vibrations exceeds a certain value. With the help of the theory of high temperature conductivity, this condition can be expressed in terms of the resistivity ρ at 0°C. It is found that $\rho n \nu^{4/3}$ ($1/n$ = atomic volume; ν = number of free electrons per atom) must exceed a value depending on universal constants only. If $\nu = 1$ is assumed, all monovalent metals except lithium do not satisfy the required condition, but most superconductors do. The energy difference between the normal and the superconducting state at absolute zero is about ms^2 (s = velocity of sound) per electron. It has thus the correct magnitude corresponding to a temperature of a fraction of a degree absolute. No application to higher temperatures or to the influence of external fields has been made yet.

I. INTRODUCTION

In the theory of the electric conductivity of metals first developed by Bloch[1] electrons are considered to move freely through the lattice except for occasional scattering by the lattice vibrations. This scattering is connected with the absorption or emission of a vibrational quantum. Anyone who is familiar with modern field theories will conclude at once that an electron will have a self-energy in this vibrational field since it

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[1] F. Bloch, Zeits. f. Physik 52, 555 (1928).

Fig. 5.6 Fröhlich's first paper on superconductivity—Reprinted with permission from H. Fröhlich, Physical Review Vol. 79, 845 (1950) Copyright 1950 by the American Physical Society is the case for the majority of electrons, the interaction is repulsive, and the electrons follow the lattice almost instantaneously, as expected intuitively in consequence their low mass compared with that of an ion, $m \ll M$.

The effect of this interaction, Fröhlich noted, is to shift electrons from the top of the usual spherically symmetric Fermi distribution, f_o, appropriate in the case of no interaction, to higher energies, resulting in a new distribution f_1, which he obtained from f_o by displacing a thin concentric shell of width ms (containing a relatively small number of electrons) from the region of the Fermi surface of f_o, thereby creating a gap between two occupied regions of momentum space: the higher energy shell and the inner Fermi sea. This shift of a fraction of the electrons to higher energies is, however, opposed by the increase in kinetic energy that must accompany such a shift. Taking this into account, it was found that f_1 yields the lower energy *provided* the dimensionless coupling constant, F, exceeded a certain critical value F_o that depended on the number of electrons per ion.

It is possible to express this criterion of the occurrence of superconductivity in terms of the resistivity at 273 K, and it was found to be obeyed by a range of superconductors, and was not obeyed by many metals that do not become superconductors. Thus, for $F > F_o$, the usual Fermi distribution f_o becomes unstable, the shell distribution f_1 yielding a lower energy. This requirement of strong electron-lattice interaction is, of course, consistent with the empirical fact that the metals that exhibit a transition to the superconducting state are those with *poor* electrical conductivity, arising from the electrons being *strongly* scattered by thermal phonons.

The energy difference $\delta E \equiv \Delta E(f_o) - \Delta E(f_1)$ between the two distributions (or equivalently, the energy difference between the normal and superconducting states at absolute zero) was found, using Eq. 5.4.1, to be proportional to Fms^2—i.e. the difference increases linearly (via F) with the strength of the electron-lattice interaction, and involves the small energy ms^2, with which Fröhlich began his deliberations. Experimentally, δE is given by

$$\delta E = H_c^2/8\pi \tag{5.4.5}$$

where H_c is the critical magnetic field above which a superconductor reverts to its normal metallic state. Despite the calculated values being larger than typical experimental values by a factor of between 10 and 100, and the very severe difficulties known to be associated with the use of perturbation theory, Fröhlich was convinced that his phonon-mediated electron-electron attraction was the key to understanding the phenomenon of superconductivity, and tentatively identified the shell distribution, f_1, with the superconducting state. To convincingly establish this, it was necessary to show that f_1 leads to the correct thermal and electromagnetic properties of superconductors. However, apart from showing that a finite energy was necessary to deform the f_1 distribution (which thus exhibits a certain rigidity reminiscent of that which London had shown phenomenologically to lead to the Meissner effect (London 1950)), his own efforts [F76, F78] based on the f_1 distribution were not otherwise successful, and his identification of f_1 with the superconducting state proved somewhat premature. Notwithstanding this, his identification of a phonon-mediated electron-electron interaction proved to be the key that broke the deadlock in understanding superconductivity, as will become apparent.

Fröhlich finalised his initial calculations during a visit to Purdue where he spent the Spring Semester of 1950, and he submitted his paper [F76] to the *Physical Review* on 16 May 1950, before leaving. The referee was Bardeen who immediately recommended its publication. Fröhlich then spent a few days at Princeton, and it was there, at his breakfast table one morning, that he opened the current copy of *Physical Review*, dated 15 May, to find two *Letters* (Maxwell 1950; Reynolds et al. 1950), independently reporting that for mercury the superconducting transition temperature, T_c, depended on the *isotopic* mass, M, in such a way that the product $T_c M^{1/2}$ is approximately constant:

> Upon checking, I found my mass-dependence confirmed, and on 19 May sent a letter [F77] to the Proceedings of the Physical Society to claim confirmation of the basic idea—the electron-phonon interaction [F185] (Fig. 5.7).

This result was effectively already contained in Eq. 6.9 of [F76] (which appeared on 1 September 1950), namely $H_c^2 \sim m/M$, although the isotopic implications were not noted initially, but only later in a *Note added in proof*, in connection with the discovery that $T_c M^{1/2}$ is approximately constant—a result that follows from $H_c \propto M^{-1/2}$ by applying the law of corresponding states. It should be emphasised that not only did the theory predict *an* isotope effect, but that the actual M-dependence was precisely as found experimentally.[33]

It is amusing to note that one of the discoverers of the isotope effect, E. Maxwell told Fröhlich [F185] that, whilst still a young research worker, he was explicitly forbidden to investigate the isotopic dependence of the superconducting transition temperature, on the grounds that any suggestion of ever obtaining a positive result was laughable, in keeping with the prevailing belief that the ions, because of their large mass, could not possibly play any role in the phenomenon of superconductivity. Not being so dissuaded, however, he worked secretly by night, and found the effect.

It must be stressed that, contrary to accounts in many textbooks[34] and elsewhere, Fröhlich's work *predated* the publication of the positive results on the isotope investigations: it was *not* motivated by it, unlike the contemporaneous work of Bardeen (1950). Although the isotope results had been announced, prior to their publication, at a meeting sponsored by the *US* Office of Naval Research in March,

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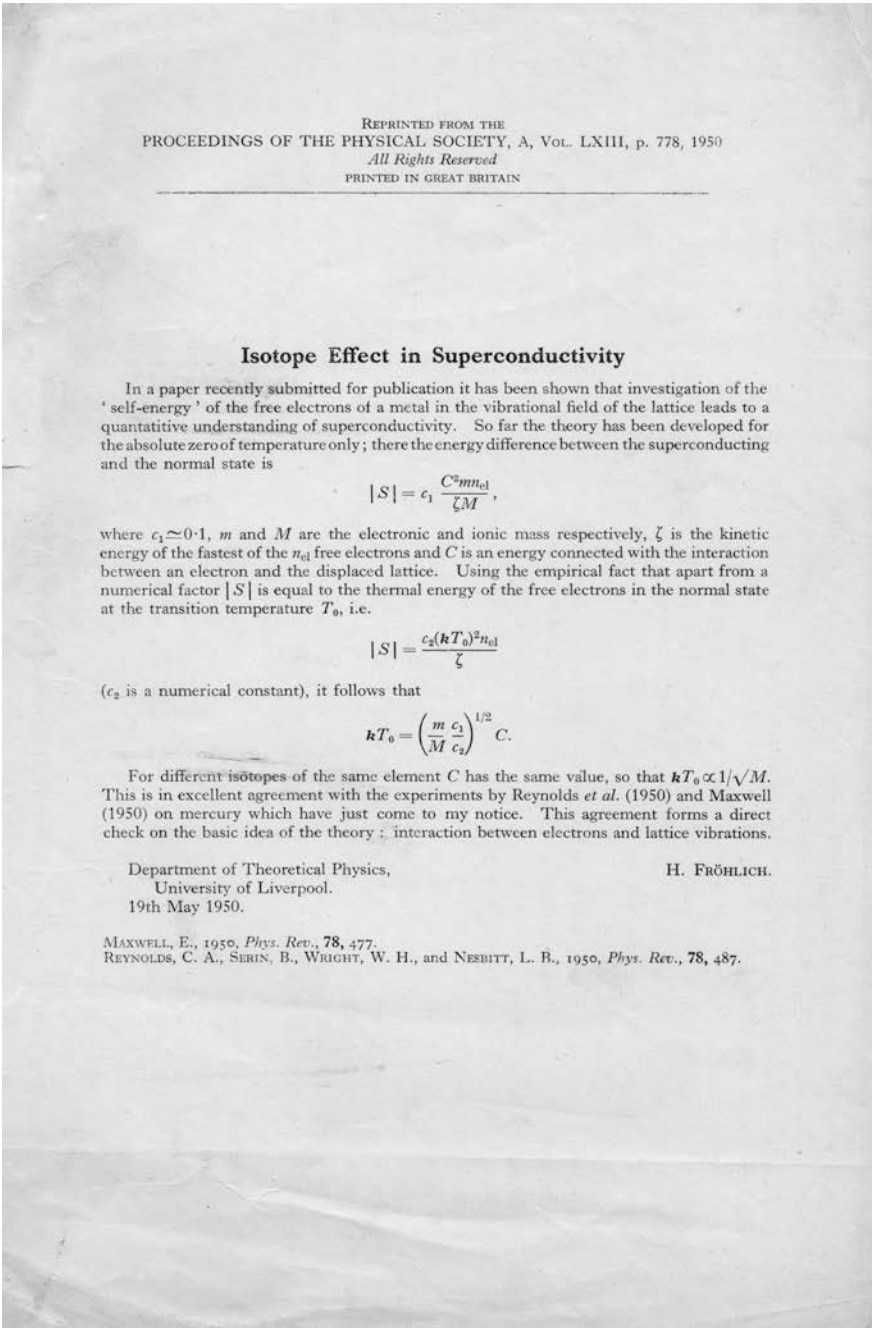
[33] In retrospect, it was fortunate that the metals investigated at the time were ones that displayed this dependence, since it was subsequently discovered that there are superconductors for which there is either no isotope effect, or one for which the dependence of T_c on H_c is much *weaker* than $M^{-1/2}$, as is the case with Transition metals to which Fröhlich returned in the 1960s [F119, 130, 131]—vide Sect. 6.1.

[34] One on the most blatant examples is to be found on page 150 of Kittel's book, *Quantum Theory of Solids,* where one reads: 'The result [of the experimental results on the isotope effect in superconductors] suggested to Fröhlich that the properties of lattice phonons, zero point or thermal, are involved in superconductivity; it is difficult to see how else the atomic mass could enter' (Kittel 1963).



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Isotope Effect in Superconductivity

In a paper recently submitted for publication it has been shown that investigation of the 'self-energy' of the free electrons of a metal in the vibrational field of the lattice leads to a quantatitive understanding of superconductivity. So far the theory has been developed for the absolute zero of temperature only; there the energy difference between the superconducting and the normal state is

$$|S| = c_1 \frac{C^2 m n_{\mathrm{el}}}{\zeta M},$$

where $c_1 \simeq 0{\cdot}1$, m and M are the electronic and ionic mass respectively, ζ is the kinetic energy of the fastest of the n_{el} free electrons and C is an energy connected with the interaction between an electron and the displaced lattice. Using the empirical fact that apart from a numerical factor $|S|$ is equal to the thermal energy of the free electrons in the normal state at the transition temperature T_0, i.e.

$$|S| = \frac{c_2 (\boldsymbol{k} T_0)^2 n_{\mathrm{el}}}{\zeta}$$

(c_2 is a numerical constant), it follows that

$$\boldsymbol{k} T_0 = \left(\frac{m}{M} \frac{c_1}{c_2}\right)^{1/2} C.$$

For different isotopes of the same element C has the same value, so that $\boldsymbol{k} T_0 \propto 1/\sqrt{M}$. This is in excellent agreement with the experiments by Reynolds *et al.* (1950) and Maxwell (1950) on mercury which have just come to my notice. This agreement forms a direct check on the basic idea of the theory: interaction between electrons and lattice vibrations.

Department of Theoretical Physics,
University of Liverpool.
19th May 1950.

H. FRÖHLICH.

MAXWELL, E., 1950, *Phys. Rev.*, **78**, 477.
REYNOLDS, C. A., SERIN, B., WRIGHT, W. H., and NESBITT, L. B., 1950, *Phys. Rev.*, **78**, 487.

Fig. 5.7 Letter concerning the isotope effect [F77], 1950—Reproduced with the permission of the Institute of Physics

Bardeen was ignorant of them until early in May 1950, when he received a telephone call from Serrin (one of the discoverers) telling him about his results. ‘I immediately thought that the electron-phonon interaction must be involved and attempted to construct a theory on this basis’ (Bardeen 1973b, p. 64).

The importance of Fröhlich’s realisation of a phonon-mediated electron-electron interaction, and his conviction that this interaction was basic to the understanding superconductivity cannot be overestimated. More than half a century later, it is now difficult to appreciate just how *avant garde* his claim appeared at the time, when the perceived wisdom ‘knew’, not only that electrons repel one another, but also that the ions, in consequence of their large mass, could play no role in the phenomenon of superconductivity. From his experience with the polaron, however, Fröhlich suspected that this was not necessarily the case, and his thesis proved crazy enough to be correct. Its gradual acceptance was due not so much to the vindicating discovery of the isotope effect, but rather to the fact that a new type of electronic interaction had been found within the *existing* semi-empirical free electron model, *without* having to admit any new ad hoc hypothesis; this was the view of Pauli, for example, who subsequently directed his pupil Schafroth towards superconductivity.

Typical of the desperate situation with respect to superconductivity, which existed prior to Fröhlich’s work, were Pauli’s dictum: ‘theories of superconductivity are wrong’, and Felix Bloch’s claim: ‘theories of superconductivity can be disproved’ [F109]. In this connection, however, the following quotation from a letter from Fritz London to Laszlo Tisza (dated 2 November 1950) is of interest:

> I was in some correspondence with Fröhlich. I am quite sure that his new interaction is the thing needed for superconductivity, but his first attempt to devise the electrodynamics seemed to me not quite to the point. Just today he writes to me that he has got it, and I am convinced that he, if anybody, is the man to do it.

Whilst in Princeton, Fröhlich visited Bell Labs where Bardeen was then working, and they compared their approaches to the problem of superconductivity:

> Although our approaches were different, mine using a variational method and his perturbation theory, both theories were based on the self-energy of the electrons in the phonon field rather than a true interaction between electrons (Bardeen 1973b).

At Bell Labs, Fröhlich met also Schockley, with whom he discussed his 1947 work [F60, F61] on dielectric breakdown, which was based on the introduction of an electronic temperature that is higher than that of the lattice; the following year, Schockley coined the expression ‘hot’ electrons (Shockley 1951) to describe this situation (vide Sect. 4.7). Around this time—and probably not unconnected with his contributions to both these fields of mutual interest—an approach was made by Bell Labs to entice Fröhlich away from Liverpool to become their ‘specially endowed’ professor at Princeton University. This did not materialise, for not only was he unwilling to relinquish his effectively pure research post in Liverpool for one that would undoubtably have entailed undergraduate teaching,[35] but also he had recently married a young American philosophy student, Fanchon Aungst who did not wish to return to America at that time. She had only recently come to England from Chicago, where she had been a pupil of Rudolf Carnap,[36] in order to read Philosophy in Oxford under Peter Strawson at Somerville College. In Oxford, the legendary 'Miss Anscombe' (Elizabeth Anscombe[37]), the analytical philospher and authority on Ludwig Wittgenstein, made a lasting impression on her. Having landed in Liverpool in 1949, on her way to Oxford, Fanchon attended a meeting of the local German Circle, which at the time was frequented by many European intellectuals, such as Baroness Rausch von Traubenberg (Marie-Hilde Rosenfeld,[38] 1889–1964) and Baroness Erisso (Eva von Sacher-Masoch,[39] 1911–91) who was then a Ph.D. student in Liverpool, and it was there, in a house in Gambier Terrace, that she was introduced to Fröhlich by a mutual friend, Erika Wirtz, a lecturer in German at the university. They were married the following year on Monday 26 June 1950, immediately after his return from America, and while she was still studying in Oxford[40]; he was then 44 years old and she, 22. That morning, Fröhlich did not appear at coffee, which was unusual since it was known that he was not away on a trip; neither did he appear at afternoon tea, by which time he was known to be in the department. In response to queries by those present, Szigeti, who also had been absent at morning coffee, explained that Fröhlich had got married that morning, and was now catching up with his work! (Powles 1973). Another contributory factor in his not wanting to leave Liverpool was his love of Chinese food, since, at that time, Liverpool had what he considered to be the best Chinese restaurants in Europe!

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[35] Fröhlich gave undergraduate lectures only during his first few years in Liverpool, when he lectured (invariably without notes) on Quantum Mechanics, Special Relativity and Statistical Mechanics (H. Newns, private communication, 2006).

[36] Before moving to the *USA* in 1935, Carnap (1891–1970) had been a leading member of the Vienna Circle, and an advocate of logical positivism.

[37] Elizabeth Anscombe (1919–2001) graduated from Oxford 1941, held Research Fellowships at Newnham College, Cambridge (1942–45) when she first met Wittgenstein, and at Somerville College (1947–1970; she became Professor of Philosphy at Cambridge (1970–86), the Chair that Wittgenstein had held between 1939 and 1947.

[38] Daughter of Hugo and Helene Rosenfeld, and wife (and assistant) of the physicist Baron (Freiherr) Heinrich Rausch von Traubenberg (1880–1944). Her first husband, Otto Riess was killed in WWI in 1914, whilst her second died of a heart attack in 1944, following her arrest by the Nazis.

[39] An Austrian aristocrat, and a great-niece of Leopold von Sacher-Masoch, author of *Venus in Furs*. She married Major Robert Glynn Faithfull in 1946; their daughter is the former the singer Marianne Faithfull.

[40] After their marriage, Fanchon continued with her studies at Somerville College, Oxford, until 1953, gaining a B. Litt. degree. She then studied art, firstly at the Liverpool Art School, where she held a Liverpool Travelling Scholarship in painting, and later with Peter Lanyon (1918–64) in St Ives, Cornwall. In 1972, she went to Japan to study ink painting in Kyoto. She has exhibited in galleries in Liverpool and in Paris, where, for a number of years, she painted and etched at S.W. Hayter's famous *Atelier 17*. She has also published articles, not only in philosophical journals, such as *Mind*, but also, in art and scientific journals.

Back from the *USA*, Fröhlich presented a report of his theory of superconductivity at the Summer Provincial Meeting of the Physical Society, which that year was held in Liverpool, 7–8 July 1950. His report concluded with a plea for experiments using more isotopes, and in the discussions that followed was informed by Pippard that Shoenberg's Cambridge group was keen to undertake the necessary measurements, if isotopes could be made available, which W.D. Allen of the UK Atomic Energy Authority's Harwell Laboratory immediately offered to arrange. Apparently, however, he had earlier sent isotopes to Mendelssohn's group at Oxford University's Clarendon Laboratory, but the samples had remained on Mendelssohn's desk uninvestigated until he heard about positive results obtained by Shoenberg's group. The latter's results showed that the critical magnetic field in tin decreases with increasing isotopic mass according to $M^{-1/2}$ when the *temperature* remains *constant* (Allen et al. 1950), as originally predicted by Fröhlich's theory [F76, Eq. 6.9]; it was this finding, incidently, that finally persuaded Bohr that Fröhlich was on the right track. Fröhlich attributed Mendelssohn's initial reluctance to investigate isotopes to the then prevailing belief that the ions play no role in the phenomenon of superconductivity, a belief that was possibly fuelled by the fact that Kamerlingh Onnes had failed to find an isotope effect in lead, as early as 1922 (Kamerlingh 1922). For further insights into the isotope affair, the account by Dahl should be consulted (Dahl 1992).

Prior to the publication of the eventual solution of the problem of superconductivity in 1957 (Bardeen et al. 1957), Fröhlich published 3 papers of singular elegance and importance, the last of which, in 1954, was a review of polaron theory entitled *Electrons in Lattice Fields* [F90], which, as mentioned in Sect. 5.3, was what prompted Feynman to apply his path integral approach to the problem; the other two papers dealt with superconductivity.

The first of these [F84], published in 1952, is arguably Fröhlich's most influential contribution to physics, and marked the start of a new era with its introduction of the methods and concepts of quantum field-theory into non-relativistic condensed matter physics. His introduction of creation and annihilation operators for both electrons[41] *and* phonons, permitted the derivation of what is now known as the 'Fröhlich Hamiltonian', H_F: 'A definite Hamiltonian stood where before there was emptiness; a definite mathematical problem was posed' [F125]; '.......from a state of impotence the efforts of physicists were now channelled towards a definite task' [F136].

H_F contains 3 terms: (i) the kinetic energy of the electrons, whose mutual Coulomb interaction is neglected, in keeping with the success of the free electron theory in describing the non-superconducting state: this energy is expressed in terms of anti-commuting, fermion operators, a_k and $a_k^\dagger$; (ii) the energy of the lattice vibrations expressed in terms of commuting, boson operators, b_w and $b_w^\dagger$; (iii) the electron-lattice interaction energy, which involves products of the type $b_w a_k^\dagger a_{k-w}$ whose non-linearity means that each system is coupled to itself—i.e. a change in the

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[41] It should be remembered that Fröhlich had first applied second quantization to electrons already in 1931 [F2], as recounted in Sect. 2.2 of Chap. 2.

electron distribution has an effect on the phonons, which, in turn, affects the electrons; thus:

$$H_\mathrm{F} = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \sum_{\mathbf{w}} \hbar s' w \left(b_{\mathbf{w}}^\dagger b_{\mathbf{w}} + \frac{1}{2} \right) + i \sum_{\mathbf{k},\mathbf{w}} D_{\mathrm{w}} \left(b_{\mathbf{w}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}-\mathbf{w}} - b_{\mathbf{w}}^\dagger\, a_{\mathbf{k}-\mathbf{w}}^\dagger a_{\mathbf{k}} \right) \tag{5.4.6}$$

where ε_k is the one-electron energy, $\hbar^2 k^2/2m$, s′ is the speed of sound in the *absence* of the electron-phonon interaction, and the real quantity D_w is defined by:

$$D_\mathrm{w}^2 = C^2 \hbar s' w/\, 2nVM(s')^2 \tag{5.4.7}$$

where C is a constant with the dimensions of energy, V is the total volume, M is the ion mass, and n their number per unit volume. D_w^2 can be expressed in terms of a dimensionless coupling constant F' and the Fermi energy $\mathrm{E_F}$, according to:

$$D_\mathrm{w}^2 = 4F' E_\mathrm{F}\ \hbar s' w/3nV \tag{5.4.8}$$

where F' is defined by

$$F' = 3C^2/8E_\mathrm{F} M(s')^2 \tag{5.4.9}$$

which is clearly closely related to F defined by Eq. 5.4.3.

He now used a canonical transformation to eliminate, as far as possible, the electron-phonon interaction from the Hamiltonian of Eq. 5.4.6, in the process of which retardation was again neglected. The aim was to focus attention on the (phonon-mediated) electron-electron interaction, rather than on the very much larger polaron-like self-energy, which in perturbation theory was given by ΔE_1. This yielded the following instantaneous electron-electron interaction, H_int, which is *attractive* between electrons near the Fermi surface whose energies differ by less than that of the exchanged lattice quantum of the wave-vector $\boldsymbol{w}$, in agreement with the original approach using perturbation theory:

$$H_\mathrm{int} \sim F \sum_{\mathbf{k},\,\mathbf{q},\,\mathbf{w}} \frac{(\hbar s w)^2}{\left[(\varepsilon_{\mathbf{q}-\mathbf{w}} - \varepsilon_{\mathbf{q}})^2 - (\hbar s w)^2 \right]} a_{\mathbf{q}}^\dagger a_{\mathbf{q}-\mathbf{w}} a_{\mathbf{k}-\mathbf{w}}^\dagger a_{\mathbf{k}} \tag{5.4.10}$$

F and s are renormalised values of F' and s', which are given by:

$$F = F's'/s \qquad s = s'(1 - 2\nu F') \tag{5.4.11}$$

where ν is the number of electrons per ion.

The renormalization of the speed of sound can be considered as a kind of ‘inverse’ of polaron formation, in which instead of an electron carrying with it some lattice deformation, the ions carry with them oscillations in the electron density, which increase their inertia, resulting in a decrease in the speed of sound. The existence of this renormalisation disposed[42] of Wentzel's criticism that the magnitude of the parameter F, necessary for superconductivity to occur in Fröhlich's original theory of 1950, had to be so large that it entailed an instability of the lattice (Wentzel 1951). For Eq. 5.4.11 shows that to ensure that the renormalised speed of sound is positive (condition for lattice stability), it is the *unrenormalised* quantity F' that must be below a certain value; F itself can be arbitrarily large (Fig. 5.8).

It should be appreciated that Fröhlich's Hamiltonian permitted, for the first time, systematic investigation not only of the phenomenon of superconductivity, in particular, but also of the electron-phonon interaction in metals, in general, such as its effect on the density of electronic energy levels; the associated impact on specific heat, C_v, was considered by Buckingham (1951). The possibility that a residual attractive interaction exists in *all* metals, which at low temperatures has an ordering effect on the electron Fermi gas such that its specific heat is reduced, was the subject of a short note in 1963 [F119] in which Fröhlich argued that such a behaviour would entail a modification of the Third Law of thermodynamics such that for systems in equilibrium, $\partial C_v/\partial T \to 0$ as $T \to 0$—*see also* [F125].

In passing, it should be recorded that Fröhlich's introduction of field-theoretic developments into solid state physics was later reciprocated by a gradual flow of concepts from condensed matter physics back into nuclear and particle physics, such collective motion in nuclei, and quark/gluon condensates.

In the early 1950s, Fröhlich spoke on his phonon-mediated electron-electron interaction mechanism at a number of international conferences, including the *NBS* Low Temperature Physics Conference in Washington in 1951 [F81], the Lorentz-Kamerlingh Onnes Centenary Conference on Electron Physics in Leiden (Netherlands) in 1953 [F85], the International Conference of Theoretical Physics in Japan in 1953 [F88] (at which he spoke also on the polaron problem [F87]), and at the 10th Solvay Conference in Bruxelles in 1954, where he first reported [F93] his solution for a one-dimensional model of a superconductor (vide infra). It is clear from the discussions at these meetings, however, that his ideas were by no means unanimously accepted, although the great importance of his phonon-mediated electron-electron interaction was acknowledged by Bohr and Heisenberg at the Leiden Conference [F85]:

> I most thoroughly appreciate the great importance of Fröhlich's contribution to our understanding of the interaction between the electrons through their coupling with the ion lattice (Bohr 1953).

> I completely agree with Prof. Fröhlich that the isotope effect in superconductors suggests very strongly the predominance of an interaction of the kind produced by the zero point lattice vibrations, as he has discussed. Coulomb interaction seems to be less important (Heisenberg 1953).

Further progress towards obtaining superconductive solutions of Fröhlich's Hamiltonian was, however, thwarted by difficulties connected with the use of

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[42] For an earlier refutation of Wentzel's criticism, based on [F76], see Huang (1951b).

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Interaction of electrons with lattice vibrations

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Using methods of modern field theories a canonical transformation of the Hamiltonian of free electrons in the field of the lattice vibrations is performed. This transformation takes account of the bulk of the interaction of the electrons with the vibrational field and leads to a renormalization of the velocity of sound and of the interaction parameter F. An objection of Wentzel's against the use of large F is removed in this way. Even in the case of weak interaction the transformed Hamiltonian contains already in zero order terms which require a modification of the usual procedure in the theory of metals, and which at low temperatures lead to an increase of the effective mass of the electrons. Treatment of strong interaction requires the development of a new method.

1. Introduction

In the theory of metals in its simplest form electrons are treated as free, apart from their interaction with the lattice vibrations. Bloch (1928) has described this interaction in terms of absorption or emission of vibrational quanta. Recently, it has been noticed by the author (Fröhlich 1950, quoted as I) that this implies also the possibility of virtual emission and absorption of quanta and in this way gives rise to an interaction between electrons. In fact, the situation is best described in terms of a field theory in which the electrons are the sources of the vibrational field. Discussion with the help of perturbation theory led to the introduction of an interaction parameter F. It was found that if F is larger than a critical value F_0 then the electron distribution in momentum space differs in the ground state from the normal distribution. This new state was tentatively identified with the superconductive state which led to a prediction of the isotope effect. Starting from a knowledge of this effect, Bardeen (1950) has developed a theory on similar lines.

Further development of the theory has been hindered so far by mathematical difficulties. For the condition $F > F_0$ implies that perturbation theory can no longer be usefully applied to calculate details of the energy spectrum, although it should be expected to lead to the correct magnitude of the energy of the ground state.

The case of normal metals for which $F < F_0$ shows also some points of interest. Thus, as Buckingham (1951) has pointed out, the density of energy levels is altered by the interaction. This may lead to anomalies in the specific heat. Also, as was first suggested by Cooke (1951), certain anomalies in the soft X-ray emission spectrum of Na observed by Skinner (1940) might find their explanation in this way. The only explanation that was available at the time would require an overlapping of energy bands which is very unlikely in Na.

The major part (E_1) of the interaction energy between the electrons and the lattice vibrations is due to an adjustment of the electronic density in the potential field of the lattice vibrations and follows them adiabatically. It is this part which gives rise to the (negative) self energy of a vibrational quantum calculated by

Fig. 5.8 Fröhlich's most influential paper introducing quantum field theory into solid-state physics—Reproduced with the permission of the Royal Society perturbation theory, noted above, and by the absence of any other systematic method: a further *idea* seemed to be lacking, as noted, in particular, again by Bohr and Heisenberg in their discussion of Fröhlich's Leiden paper:

> In fact, as is generally recognised, we have to do in the superconductive phase with a state of the electrons which, although differing very little in energy from the normal one, exhibits a high degree of order… (Bohr 1953)

> It will be reasonable to picture the state with current as a 'solid body' of electrons moving through the ionic lattice[43] (Heisenberg 1953)

In the absence of any such idea, and to refute a criticism from van Vleck and Slater at the *NBS* Meeting in Washington in 1951 that his Hamiltonian could not yield a phase transition, Fröhlich considered the *one*-dimensional model mentioned above, which, provided the interaction of an electron with the lattice is so strong that the recoil of the lattice when an electron is scattered can be neglected, he was able to solve *non*-perturbatively, using a Hartree self-consistent field approximation [F89]. A single lattice mode becomes strongly excited, and acts as a spatially periodic field on the electrons, which produces a *gap* in the single electron energy spectrum, *à la* Peierls (Peierls 1930); the gap was found to be proportional to $\exp(-3/2F)$. This particular form of the dependence of the gap on F *cannot* be expanded as power series about $F = 0$ (the function has an essential singularity at $F = 0$), which indicates the impossibility of ever obtaining such a gap in *any* order of perturbation theory,[44] as was later rigourously shown by Migdal (1958).

Despite this gap, the system is *not* an insulator at absolute zero because the periodic variation in electron density is tied, not to the lattice itself but rather, to the lattice *modes* that are here strongly excited, and which it enforces in a self-consistent way. The energy difference, δE per atom between this state and that in the absence of any electron-lattice coupling was found to be given by $E_F \exp(-3/F)$. The appearance of the Fermi energy, E_F, in place of the energy ms^2, which characterised the perturbation result, means that there is here *no* isotope effect—its absence originating in the 'over-strong' electron-lattice interaction assumed ab initio, via which the dynamical properties of the lattice are effectively suppressed. Furthermore, the presence of the large energy E_F outweighs the reduction arising from the replacement of F by $\exp(-3/F)$, resulting in an unrealistically large energy difference. It may be noted, however, that the rapid variation of the function $\exp(-1/F)$ with F is consistent with the very sensitive empirical dependence of the superconducting transition temperature with pressure, despite F itself varying little under compression.

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[43] This possibility had much earlier been suggested by Frenkel (1933).

[44] In this connection, it should be appreciated that the status of the gap that characterises the f_1 distribution of Fröhlich's paper of 1950 [F76] is quite different, in that it separates two *occupied* regions of the *same* momentum space, and was introduced 'by hand'.

The possibility that a gap characterises[45] the superconductive state has a long history in the phenomenological development of the subject, first appearing in the work of F. and H. London in 1935 (London and London 1935). Three years later it was invoked by Welker in an abortive theoretical attempt (Welker 1938) to understand the Meissner-Ochsenfeld effect—namely, the expulsion of a magnetic field applied above the superconducting transition temperature, T_c, upon cooling through T_c, so that, in the superconductive state, the system behaves as a perfect diamagnet;[46] such perfect diamagnetism had, incidentally, been anticipated by Frenkel the year before it had been discovered, in the paper to which reference was made in Chap. 2 (Frenkel 1933). The idea of a gap resurfaced again in the experimental work of Daunt and Mendelssohn on persistent currents in 1946, where the existence of a gap 'protects' the system against dissipation (Daunt and Mendelssohn 1946). The essential singularity that characterises the gap in Fröhlich's one-dimensional model (which was the first to be derived theoretically, and in an exact way) turned out *not* to be peculiar to the low-dimensionality of the system considered, but was shared by the later work of Cooper in 1956 (vide infra), and by the full three-dimensional solution of Bardeen, Cooper and Schrieffer (*BCS*) the following year (vide infra). Prior to this, Bardeen had shown (Bardeen 1955) that a gap would entail a *non-local* relation between current density and magnetic field of the form that had been suggested by Pippard from his microwave measurements that showed that the depth to which a magnetic field penetrates a superconductor is essentially *independent* of the strength of the field. The non-locality was found to be characterised by a macroscopic distance—the so-called 'coherence length', of the order of 10^{-4} cm (Faber and Pippard 1955)—a quantity that was later to feature prominently in the *BCS* theory. The existence of a gap was first confirmed by measurement of the electronic specific heat[47] in vanadium, which revealed a temperature dependence proportional to exp $(-\Delta/2kT)$, where the gap, Δ, was of the order of kT_c (Corak et al. 1954), and later by measurements of heat conductivity and optical absorption.

In the meanwhile, the thermodynamic properties of this one-dimensional model were elaborated the following year by C.G. Kuper, then a Research Fellow in Fröhlich's department, who showed that the model does indeed exhibit a second-order phase transition (Kuper 1955) in which the gap acts as a temperature-dependent 'order parameter'. With increasing temperature, excitation of electrons across the gap reduces the periodic variation in electron density, which, in turn, reduces the amplitude of the resonant lattice modes, thereby narrowing the gap, which eventually vanishes above a certain temperature (the transition temperature),

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[45] Whilst the existence of a gap proved to be necessary for superconductivity, it was not *sufficient*, since insulators are themselves characterised by a gap.

[46] There is, however, an essential difference between normal diamagnetism and the Meissner effect, in that the former can support a uniform, static, non-zero magnetic induction, whilst the latter cannot (vide Sect. 6.2).

[47] At the Solvay Meeting of 1954, Fröhlich noted that the transition to the superconductive state should be reflected in a 'qualitative change' in the electronic specific heat, and suggested that this be investigated experimentally [F93]; Corak's experimental work was published the same year.

T_c, given by $kT_c \approx E_F \exp(-3/2F)$. The electronic specific heat was found to be exponential near $T = 0\ K$ and to exhibit a discontinuity at T_c.

Despite some unrealistic features—such as the transition temperature being large compared to the Debye temperature, a condition that is not fulfilled in real superconductors[48] (and which here arises from the presence of the large Fermi energy, E_F in the prefactor of the exponential term in the expression for T_c)—it is interesting that, some 20 years later, this one-dimensional model was to find application (Bardeen 1973a, b) to the so-called paraconductivity attributed to sliding modes in quasi one-dimensional organic and other low dimensional systems—*see also* [F173].

Crucial to the eventual *BCS* solution of the problem of superconductivity in 1957 was Leon Cooper's demonstration in 1956 (Cooper 1956) that, in the case of just *two* electrons above the Fermi sea, Fröhlich's phonon-mediated attractive interaction results in a *single* bound-state of zero[49] centre-of-mass momentum, *no matter how weak* the attraction—essentially because the Pauli principle blocks any possible decay channels; the energy was again found to exhibit an essential singularity in the coupling constant.

The extension of Cooper's work to many electron proved, however, to be quite complicated because of the Pauli principle, which prevents the pairing off all the electrons in Cooper pairs in any straightforward way—*except* when all the pairs have the *same* net momentum, which in the ground-state must be zero.

This *pairing*, which proved to be the concept that had earlier been missing,[50] had, incidentally, been independently anticipated qualitatively somewhat earlier by M. Schafroth (1954), following his work in Liverpool, to where, in 1952, he had been sent by Pauli to work with Fröhlich. The very first mention, of pairing, however, seems to have been made 8 years earlier by Ogg (1946) in connection with superconductivity in metal-ammonia solutions, which he attributed to Bose-Einstein condensation of *pairs* of electrons, a suggestion reiterated[51] by Onsager in 1951.

The work of *BCS* was based on an *approximate* form of Fröhlich's electron-electron interaction (Eq. 5.4.10) in which its momentum dependence (as contained in the factor $G(\varepsilon_{\mathbf{q}-\mathbf{w}} - \varepsilon_{\mathbf{q}}) \equiv (\hbar s w)^2[(\varepsilon_{\mathbf{q}-\mathbf{w}} - \varepsilon_{\mathbf{q}})^2 - (\hbar s w)^2]^{-1})$ is neglected, and is replaced by a negative *constant* within a shell of width $\hbar\omega_D$ centred on the Fermi surface, where ω_D is the Debye frequency, and by zero elsewhere—i.e.

$$G \to \begin{cases} -1, & \text{for } |\varepsilon_{\mathbf{q}-\mathbf{w}} - \varepsilon_{\mathbf{q}}| < \hbar\omega_D \\ 0, & \text{for } |\varepsilon_{\mathbf{q}-\mathbf{w}} - \varepsilon_{\mathbf{q}}| > \hbar\omega_D \end{cases} \tag{5.4.12}$$

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[48] It was much later pointed out by Kuper at the Fröhlich Centenary Meeting in Liverpool in 2006, that what Fröhlich had constructed was not actually a model for superconductivity in one-dimension, but rather a charge-density wave (Kuper 2008).

[49] For zero total momentum, the two electrons are most tightly bound.

[50] In this connection, it should be appreciated that there is actually *no* pairing in Fröhlich's one-dimensional model, but rather a macroscopic occupation of a moving lattice wave that carries the electrons along with it, each symbiotically sustaining the other.

[51] Cited as 'Private Communication' in Kuper (1967).

In addition, they included the short-range repulsion, $V_c(\boldsymbol{w})$, that remains after the long-range part of the inter-electronic Coulomb interaction has been taken care of in terms of plasma oscillations. Their electron-electron interaction Hamiltonian is thus based on terms of the form:

$$h_{\text{int}} \sim [-F + V_c(\boldsymbol{w})] a_{\mathbf{q}}^{\dagger} a_{\mathbf{q}-\mathbf{w}} a_{\mathbf{k}-\mathbf{w}}^{\dagger} a_{\mathbf{k}} \tag{5.4.13}$$

It can be seen from Eq. 5.4.10 that the attractive interaction is strongest when the two electrons have the *same* energy, which will be the case if they have equal and opposite momenta ($\boldsymbol{k}$, $-\boldsymbol{k}$), so that the pair has zero centre-of-mass momentum—i.e. their centre of gravity is at rest. *BCS* chose their variational ground-state wave-function to ensure that the maximum number of such pairs of electrons (with of zero[52] centre-of-mass momentum) take advantage of the attraction, and *only* such pairs (in which the electrons also have *anti*-parallel spin) are considered. The resulting many-pair state is a *single* quantum state, which is cooperatively produced and exhibits coherence[53] with an associated coherence length of the same order of magnitude as that predicted by Pippard, namely, 10^{-4} cm, mentioned above. It should be noted, however, that in contrast to the earlier work of Schafroth, the many-electron wave-function does *not* describe bosonic pairs of electrons, but rather correlations between pairs of electrons separated by a distance of the order of 10^{-4} cm; since this far exceeds the average inter-electronic separation, electrons belonging to millions of *other* pairs will be found within this distance. Accordingly, the pairs cannot be considered to be independent entities; instead, they are spatially interlocked in a highly intricate way that guarantees their collective coherence, and in consequence, the pairs do *not* satisfiy Bose commutation relations. As Bardeen stressed in his contribution to Fröhlich's 1973 *Festschrift* '.....the key thing is pairing, not pairs. Although often used, the concept of (boson) pairs is misleading; they are not stable above the transition temperature, they overlap strongly and would not exist but for their interaction.' (Bardeen 1973b)—i.e. pairing is inherently a cooperative effect, which is best treated in *momentum* space, rather than in position space where correlations between *more* than just *two* electrons would have to be considered.

The energy difference, δE, between the ground-state of this system and that in the absence of any interaction was now found to be given essentially by $ms^2\exp(-2/F)$; this value is much smaller than that given by Fröhlich's original perturbative calculation, where the energy difference was proportional to ms^2F, because F now enters via the factor $\exp(-2/F)$, which is much smaller than F. It should be noted that the *BCS* result is a synthesis of the energy ms^2 that characterised Fröhlich's original perturbative calculation, and the essential singularity factor of his one-dimensional model; the desirability of developing a method that 'forms a link between the two methods

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[52] It will be recalled that in the ground-state of an ideal Bose gas, *all* the particles have zero momentum (Einstein condensation).

[53] Coherence means that the different parts of the system behave dynamically in a perfectly correlated way—i.e. are *in phase* with each other.

discussed so far' had already been stressed by Fröhlich 3 years earlier [F93]. As with the case of Cooper's single-pair calculation, the many-pair ground-state is found to be stable for all positive values of the coupling constant F, *however small*. It follows from Eq. 5.4.13, however, that the effect of including the short-range part of the direct inter-electronic Coulomb repulsion[54] is to reduce F to a value F^*, so that the criterion for superconductivity becomes $F^* > 0$, i.e. the phonon-mediated electron-electron attraction must be strong enough to *dominate* the short-range part of the direct Coulomb repulsion.

In the case when each pair has the same *non*-zero total (centre-of-mass) momentum, the pairs move cooperatively together (coherence), again as a *single* quantum state,[55] effectively realising the 'solid body' envisaged by Heisenberg, thereby ensuring that the flow is stable against dissipation; for the break-up of any particular pair necessarily involves *all other* pairs, which would thus require an enormous expenditure of energy.

Although the assumption of an associated 'rigidity' in the superconducting many-electron wave-function had been phenomenologically shown to lead to the Meissner effect (London 1950), as already noted, this effect *cannot* be satisfactorily derived within the *BCS* theory because of problems of gauge invariance arising from the approximate form of the electron-electron interaction used (Schafroth 1958). As will be noted in the next chapter (Sect. 6.2), however, an exact, a model-independent derivation was eventually given by Sewell in 1990, in terms of the macroscopic wave-functions earlier introduced by Fröhlich in the late 1960s, following the work of Yang (vide Sect. 6.2).

At finite temperatures, some Cooper pairs will be broken up, so that a superconductor contains individual (unpaired) electrons in addition to Cooper pairs, the former increasing in number with increasing temperature, according to $\exp(-\Delta/2kT)$, where Δ is the energy gap in a superconductor—i.e. the energy required to unbind a Cooper pair thereby creating two separate, *un*paired, electrons. It is these unpaired electrons that are responsible for the electronic specific heat of a superconductor, which must, accordingly, be expected to be proportional to $\exp(-\Delta/2kT)$; as already noted, this was precisely as found by experiment(Corak et al. 1954). Unlike in the early approach of Welker, but in common with Fröhlich's one-dimensional model, the gap the *BCS* theory is a *decreasing* function of temperature, and vanishes at the transition temperature, in accordance with the 2nd order nature of the superconducting-to-normal phase transition.

---

[54] Subsequent work by others showed that the effect of taking into account the *retarded* nature of the phonon-mediated electron-electron attraction is to reduce this direct short-range Coulomb repulsion, in consequence of the latter being retarded by a time $1/\omega_P$ (where ω_P is the electronic plasma frequency), which is much shorter than that, $1/\bar{\omega}$, which characterises the former (where $\bar{\omega}$ is an average lattice frequency); this reduction in the short-range repulsion favours the occurrence of superconductivity—vide, for example Eliashberg (1960).

[55] Consistent with this is the absence of thermoelectric effects in a superconductor; in particular, the absence of a Peltier effect indicates that the supercurrent-carrying state has zero entropy.

It is of interest to record (as already mentioned in Sect. 5.3) that, many years later, Schrieffer told Fröhlich [185] that the form of his many-electron extension of Cooper's pair wave-function, in which all pairs have the *same* total (zero) momentum, was actually motivated by the structure of the variational wave-function of the large polaron in the so-called 'intermediate-coupling' regime, first considered by Fröhlich in collaboration with Gurari in 1953 (Gurari 1953), and, independently, by others the same year (Lee et al. 1953).

It was later realised by Yang (vide Sect. 6.2), however, that the pairing correlations between electrons near the Fermi surface with equal and opposite spin and momenta, which characterise the many-pair *BCS* wave-function, *can* be collectively expressed *spatially* in terms of a two-point *macroscopic* wave-function, $\Phi_2(\boldsymbol{x}, \boldsymbol{y})$, having the form of a bound state—i.e. $\Phi_2(\boldsymbol{x}, \boldsymbol{y})$ is large only when $|\boldsymbol{x} - \boldsymbol{y}|$ is below a length (the coherence length, $\sim 10^{-4}$ cm) characteristic of a particular material. It is in terms of $\Phi_2(\boldsymbol{x}, \boldsymbol{y})$ that the zero-momentum pairs with anti-parallel spin reflects itself macroscopically as a pair condensate. $|\Phi_2|^2$ is thus in the nature of an 'order parameter', the search for which Fröhlich had advocated already in 1953, the year *before* his one-dimensional model calculation was published, commenting:

> Apart from the order of magnitude of the energy we have not yet, however, been able to derive any further properties of superconductors. ……I think, however, that the solution will not come from mathematical considerations only, but will require a new physical concept which will help us find an appropriate approximation[56]. This should involve an 'order' parameter similar to the case of second order [phase] transitions. In superconductivity we have so far not been able to find such a parameter, and I think our efforts should be directed along such lines [F88].

Notwithstanding significant differences between the eventual *BCS* theory and that of Fröhlich's earlier work—in particular, the former's treatment of pairing correlations, which turned out to be intimately connected with the required new physical concept sought by Fröhlich, namely *coherence*—it remains a scandalous mystery to those properly acquainted with the history of the subject why Fröhlich was not included in the Nobel Citation shared by Bardeen, Cooper and Schrieffer in 1972 (although the number of people who can share the same Nobel prize is, admittedly, limited to three). For several essential features of their work, not least, the Hamiltonian used- which, apart from the inclusion of a direct, short-ranged screened Coulomb repulsion between electrons, was a simplication of the actual phonon-mediated electron–electron interaction derived by Fröhlich 5 years earlier using a canonical transformation. In addition, the form of their many-pair wave-function was actually motivated by the work of Fröhlich's collaborators in Liverpool, whilst the *BCS* expression for the energy difference between the superconducting and normal states is a synthesis of the results of his earlier work [F76, F89].

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[56] This is in direct contrast to the opinion of A.H. Wilson who, in the Preface to the 2nd edition of his book *The Theory of Metals*, published the same year (1953), attributes the difficulties in understanding superconductivity as possibly 'due to the inadequacies of the mathematical techniques employed rather than to the omission of some vital physical principle.'

Fröhlich's pivotal contribution to the theory of superconductivity was, however, fulsomely acknowledged by Bardeen in a letter to Fröhlich, dated 22 July 1960, wherein he wrote:

> The introduction of this interaction by Fröhlich in 1950 and the simultaneous verification of its importance by the discovery of the isotope effect gave the break-through that pointed the way towards the development of a successful theory of superconductivity [F136].

Fascinating reviews by Fröhlich of the state of superconductivity at various epochs (some of which contain valuable historical insights) can be found in [F109, F125, F136, F146, F185]. Of particular interest is [F109], which contains an insightful section entitled *The psychology of Superconductivity.*

5.5 Return to Particle Physics

Lest the impression be gained from the earlier parts of this chapter that Fröhlich's interest in particle physics ceased with his disillusionment over the divergences that plagued further development of his pre-war work on the meson theory of nuclear forces, recounted in Chap. 4, it must be emphatically stated that this was not the case; it was a subject in which he maintained a profound interest throughout his life, and one to which he persistently returned, right up to the end, publishing some 11 papers between 1958 and 1985.

His post-war interest in particle physics was reawakened in the late 1950s with the discoveries of parity violation and *CP* invariance, the former 'striking a chord' with his early pioneering attempt with Heitler [F19] to understand the so-called anomalous magnetic moments of the neutron and proton in terms of scalar meson theory, which had been criticized by Kemmer precisely because their admitted spin-spin interaction *violated* parity! (vide Sect. 4.4 and Kemmer 1965). Neither did the associated discovery at the time of *CP* invariance come as too much of a surprise to Fröhlich. For this invariance indicates some deep connection between electric charge, generally considered to be an internal property of a particle, and the structure of the external space-time 'occupied' by the particle. Indeed, he had long suspected from the *universality* of electric charge that electric charge was more a property of electrodynamics than of the particular particles that 'carry' it, in which case, he believed, electrodynamics in its present form would need to be extended. To him, these experimental discoveries of the late 1950s were catalytic, and he accordingly embarked on what he later described [F121] as an ambitious programme '......whose aim is the derivation of the properties of particles and fields from geometrical considerations.'

Arguing, in 1960, that the conventional (passive) treatment of reflections in terms of point transformations is unphysical, he developed a novel approach involving the introduction of a new angular space in terms of which space reflections could be considered *actively* as special cases of *continuous* transformations [F106, F108]. He introduced his approach as follows:

The new treatment is based on a simple but important difference between reflexions and continuous rotation. I shall illustrate this first for an ordinary two-dimensional Euclidean space. Assume a coordinate system in which the x-direction runs from left to right, say, and the y-direction is obtained from it by a 90° anti-clockwise rotation. Consider an irregular triangle in this two-dimensional space described by the coordinates of three points (x_1, y_1), (x_2, y_2), (x_3, y_3). A rotation replaces these three by three different coordinate pairs (x_k', y_k'). As is well known in geometry there are two ways of interpreting the new set of co-ordinates: (i) the triangle has not been moved, but the co-ordinate frame has been rotated, (ii) the co-ordinate frame remains the same, but the triangle has been moved appropriately. The first interpretation has no physical (geometrical) meaning in terms of the figure. A co-ordinate system is quite an arbitrary device; completely different types of co-ordinates might be introduced without changing the geometrical properties. The second interpretation has, however, a very definite geometrical meaning: it tells us, for instance, that the angles of the triangle are unchanged by the 'rotation'. To have a closer analogy with field equations we replace the triangles by the three straight lines forming it. They are described by three equations between y and x, $a_k x + b_k y + c_k = 0$. Rotation of the coordinate frame by an angle θ (first interpretation) corresponds to a replacement of (x, y) by (x', y'), say, $x = x' \cos\theta - y' \sin\theta; y = x' \sin\theta + y' \cos\theta$. The appropriate motion of the triangle on the other hand corresponds to a replacement of a_k, b_k by $a_k' = a_k \cos\theta + b_k \sin\theta$; $b_k = -a_k \sin\theta + b_k \cos\theta$ (second interpretation). Carried out together the two transformations leave the form of the equations invariant.

Consider now a reflexion in which the value of each x-coordinate is replaced by its negative; $x_1 = -x_1$, etc. The first interpretation says simply that the frame has been replaced by another one in which the x-axis runs from right to left, instead of from left to right. The second interpretion, however, can no longer be offered in terms of continuous displacements and rotations. It would require the triangle to be turned inside out (or rather its two-dimensional analogue). Thus if we decide to avoid this latter action, then no physical interpretation can be given to reflexions.

The possibility of a physical interpretation of these reflexions in a two-dimensional system can be regained, however, if use is made of a third dimension which permits rotation around the y-axis. This third dimension then represents an angle ϕ which for $\phi = 0$, say, yields the first frame and for $\phi = \pi$ the reflected one.

Interpretation of the angle ϕ in terms of the x - y plane, suggests a formal connection Pauli spin, which has definite values (± ½) only for two directions, say $\phi = 0$ and $\phi = \pi$. The interpretation of intermediate angles would then have to be given in terms of a mixture of the original triangle, and of the reflected one. Thus right from the beginning, the present description considers the possibility of both these triangles. The co-ordinate ϕ decides which one is realized. Invariance of the above three equations under reflexion thus involves (*i*) replacement of the coordinate frame (x, y) *by* $(- x, y)$ and (*ii*) rotation of the triangle around the y axis by 180°, leading to the replacement of a_k by - a_k.

The case of Lorentz transformations is, of course, more difficult than the above case, but it offers a similar distinction between continuous transformations and reflexions. The former always offer a physical interpretation, either as (three-dimensional) rotations, or as relative motion. The latter would require actions like turning a body inside out, which would require internal degrees of freedom. Our programme must then be the development of a description which right from the beginning permits the treatment of certain properties, say momentum (p_k, p_0) together with all the reflected ones, $(- p_k, p_o)$; $(p_k, - p_o)$; $(- p_k, - p_o)$; $k = 1, 2, 3$ denotes the spatial, 0 the time part). This should be expected to involve the use of new angular co-ordinates.

> Following the above discussion I feel that point transformations other than mere coordinate replacements should he considered as unphysical and should be replaced by continuous transformations through introduction of new angular coordinates [F106, F108, adapted].

This new angular space permitted the definition of dynamical variables that could be interpreted in terms of isobaric spin, electric charge and mass, and led to a wave-equation[57] of the following form, whose solutions described bosons:[58]

$$(B_{\mu}\partial_{\mu} + M)\,\Psi = 0 \tag{5.5.1}$$

where M is a mass operator,[59] and the B_{μ} are the counterparts of Kemmer's β-matrices in the new angular space.

Crucial to the programme was the representation of the β_{μ}-matrices in terms of direct products of four sets of Pauli matrices ($\boldsymbol{\rho}$, $\boldsymbol{\rho}'$), ($\boldsymbol{\sigma}$, $\boldsymbol{\sigma}'$) by:

$$\beta_{\mathrm{k}} = \frac{1}{2}\,(\rho_1\sigma_{\mathrm{k}} + \rho_1'\sigma_{\mathrm{k}}'), \qquad \beta_4 = \frac{1}{2}\,(\rho_2 + \rho_2'), \tag{5.5.2}$$

which permitted a classification of wave-functions, Ψ, in terms of spin-pair functions referring to ($\boldsymbol{\rho}$, $\boldsymbol{\rho}'$), ($\boldsymbol{\sigma}$, $\boldsymbol{\sigma}'$); this yielded 4 different wave-functions, Ψ_{M}, Ψ_{π}, Ψ_{K} and Ψ_{v}. The angular space defined by ($\boldsymbol{\sigma}$, $\boldsymbol{\sigma}'$) is the angular part of ordinary x_{k}-space, since according to Eq. 5.5.1, rotation of the x_{k}-space is identical to a unitary transformation of the ($\boldsymbol{\sigma}$, $\boldsymbol{\sigma}'$). On the other hand, the ($\boldsymbol{\rho}$, $\boldsymbol{\rho}'$) require the definition of a *new* angular space, which was considered to be an internal space of the bosons; the existence of this space permitted treatment of any unitary transformation of the ($\boldsymbol{\rho}$, $\boldsymbol{\rho}'$) as a particular case of a continuous transformation, in conformity with the new advocated approach to reflections.

The mass operator, M, in Eq. 5.5.1 must commute with the B_{μ}, and with T^2, T_3 and Q_3, where, $\boldsymbol{T}$ and Q_3 are, respectively, the operators for isospin and electric charge, but these requirements do not completely determine its general form. Originally [F106], the following form was used:

$$M = C(1 + I^2)^2\,(T^2 + I^2), \tag{5.5.3}$$

---

[57] The starting point of the derivation of this wave-equation was Kemmer's meson equation (Kemmer 1939).

[58] In order to separate the 4 irreducible representations of the new underlying isospin algebra it was, at this time, necessary to introduce operators that could not be expressed in terms of the algebra; this defect was subsequently rectified [F115] in collaboration with Terreaux in 1963, when it was realised that the elements of the *centrum* of the underlying algebraic ring themselves permit the required separation.

[59] The introduction of a mass operator marked a dramatic departure from his earlier treatment of rest mass as an electromagnetic self-energy effect, which had led him in 1958 to propose [F102], in keeping with the fashion of the time, a simple empirical relation for the masses of baryons and mesons in terms of the fine-structure constant.

where C is a constant, and I is a kind of momentum operator that connects the constituent dashed and undashed spaces of the new angular space. In conjunction with the wave-equation Eq. 5.5.1, it was found that:

i) Ψ_π represents an isobaric spin triplet, the 3 particles having electric charges (1, 0, -1) and zero mechanical spin, which were tentatively identified with the π-mesons; their mass is $2C$.
ii) Ψ_K represents 2 isobaric spin doublets, the 4 particles having electric charges (1, 0, -1, 0), 3rd component of isobaric spin (½, -½, -½, ½), and zero mechanical spin, which were tentatively identified with K-mesons: their mass is $7C$.

A check on these identifications was provided by the predicted K:π mass ratio of 7:2, which is very close to the experimental value of 966:273; this permitted determination of the constant C whose value turned out to be 137 electron masses.

With light quanta in mind (as the only other boson known at the time), he noted that while the above expression for M allowed for a field with *zero* isobaric spin and mass, it was not sufficient to determine the external space time parts of the associated wave function. To remedy this, the following term was subsequently added, which had *no* effect on the above predictions:

$$c[\frac{1}{2}\,(\rho_3+\rho_3')]^2\sum_\mathrm{k}[\frac{1}{2}\,(\sigma_\mathrm{k}+\sigma_\mathrm{k}')]^2 \tag{5.5.4}$$

where c is a constant [F107].

It was then found that:

iii) Ψ_M describes an isospin singlet, and has 10 components in external space-time, which satisfy the Maxwell equations, i.e. it describes light quanta of spin 1. It should be noted that the ‘mass’ constant c here simply gives a measure for the electromagnetic vector potential.
iv) Ψ_v describes 2 isobaric doublets, with the same electric charge and isospin properties as does Ψ_K (i.e. the K-mesons), but now with mechanical spin 1; their mass is given by:

$$m_\nu = [7C(7C+2c)]^{1/2} \tag{5.5.5}$$

where C is the constant that parametrises the expression for M corresponding to π- and K-mesons (Eq. 5.5.3); Fröhlich called the 4 new bosons ν-mesons. The appearance of the constant c in the expression for their mass, M_v, is perhaps unexpected, given that it parametrised the operator (Eq. 5.5.4) that yielded the Maxwell equations. Provided C and c have equal signs, m_ν exceeds the mass the K-mesons. Light quanta and the ν-mesons require the use of the 10-dimensional representation of the β_μ, while the π- and K-mesons require the 5-dimensional one.

Thus not only did the wave- equation Eq. 5.5.1 describe all bosons known at the time (1960), but it predicted the existence of a further 4 vector mesons (ν-mesons).

This prediction followed simply from the symmetries of the wave-equation (Eq. 5.5.1), *without* any consideration of interactions. Fröhlich thus considered it to be particularly significant when he learned that, precisely from considerations based on the empirical properties of the weak interaction, Lee and Yang had been led, the previous year, to the conclusion that 4 new bosons should exist with properties identical to those he had predicted (Lee and Yang 1960). Lee and Yang had called the new particles 'schizons', but, in a short note [F111] Fröhlich proposed that they be named *φανχων* (from *φανοσ* and *χων*) after his wife, Fanchon (Fig. 5.9). The same year (1961), K* vector mesons resonances[60] having precisely these properties were experimentally detected (Alston et al. 1961), but for some reason this was never alluded to by Fröhlich, perhaps because he was well aware of the unsatisfactory nature of his *ad hoc* determination of the form of the mass operator, which he admitted '....... should be replaced by a compelling deivation', but which '.....will require deeper insight than has been achieved so far into properties of the new angular spaces.' [F107].

After 1961, many other mesons were discovered, which are not predicted by the above theory as it stands. In 1964, however, the whole direction of theoretical research in this area was dramatically altered with the advent of the quark model.

Prior to this, however, in his contribution [F108] to the Pauli memorial issue of the journal *Helvetica Physica Acta*, after presenting a more systematic treatment of his earlier work [F106, 107], he went on to consider, in more detail than previously, the implications of his treatment of reflections in terms of continuous transformations, noting that allowing for all possible combinations of space and time reflections *quadruples* the number of wave-equations. The other new element, however, was a 'Note added in proof', in which he reported that replacing the Kemmer β_μ by the corresponding Dirac matrices leads, under certain conditions, to a wave equation for the electron-neutrino field. This was the starting point of the subsequent work of Fröhlich's assistant in Liverpool, Ch Terreaux, who showed that the decomposition of the restricted Lorentz group into a direct product of two-dimensional unimodular transformations permits Fröhlich's quartet of fermion wave equations to be obtained in a **systematic** way (Terreaux 1962); Terreaux went on to show that the existence of spin-½ particles entails the existence of just *two* kinds of electric charges in Minkowski space. Furthermore, his equations of motion for leptons exhaust only *half* of the total number of fermion equations, inviting speculation as to what the remaining half might describe.

In a long paper [F116] published the following year (in 1963) in *Nuclear Physics*—which was prefaced by a quotation adapted from the alchemical axiom of Maria Prophetissa[61]: *One becomes two, two becomes three, and out of the three comes the*

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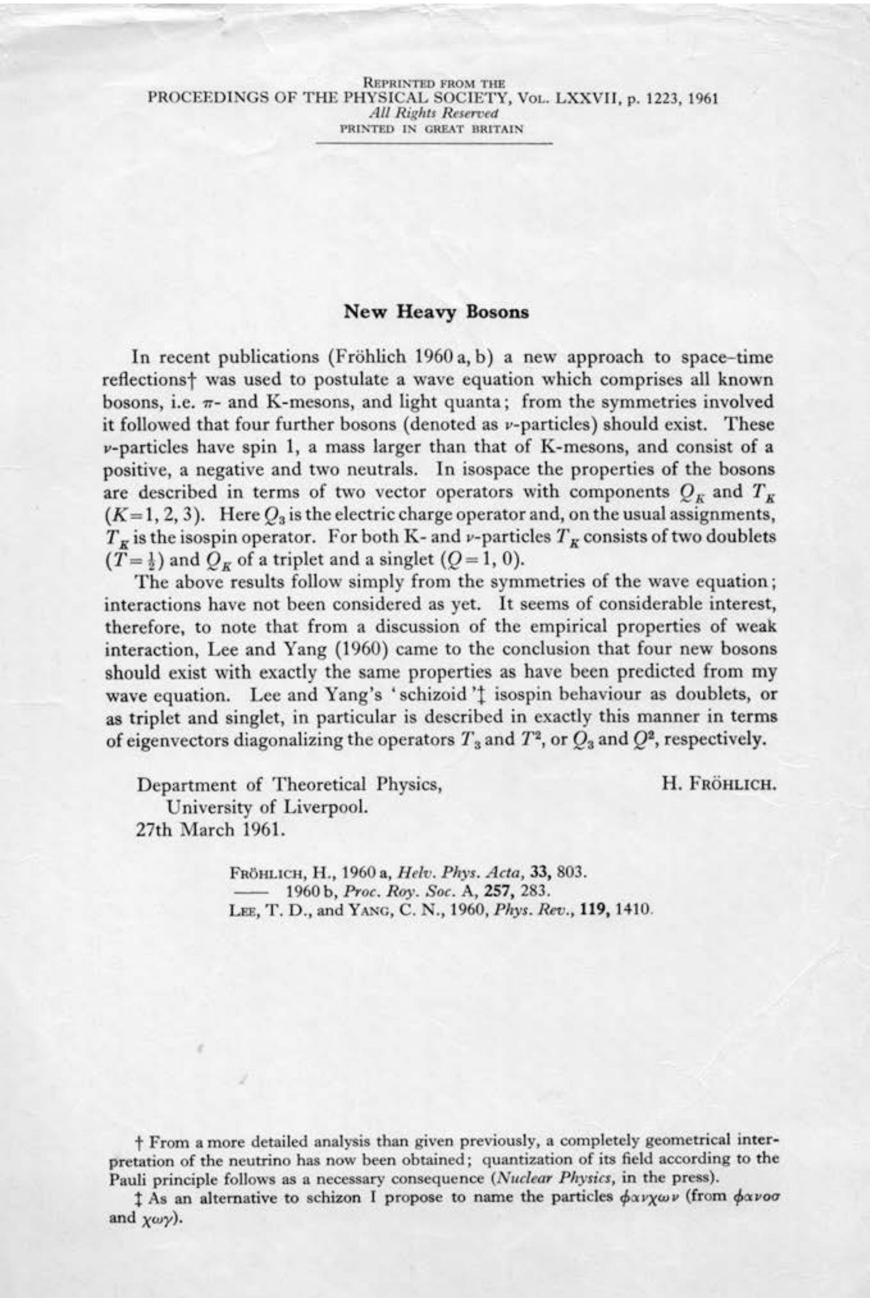
[60] The K* resonances have the same quark combinations as the K-mesons.

[61] The axiom had earlier been used by Jung as a metaphor for individuation. Like Pauli, Fröhlich was much fascinated by Jung's concept of the collective unconscious.



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New Heavy Bosons

In recent publications (Fröhlich 1960 a, b) a new approach to space–time reflections† was used to postulate a wave equation which comprises all known bosons, i.e. π- and K-mesons, and light quanta; from the symmetries involved it followed that four further bosons (denoted as ν-particles) should exist. These ν-particles have spin 1, a mass larger than that of K-mesons, and consist of a positive, a negative and two neutrals. In isospace the properties of the bosons are described in terms of two vector operators with components Q_K and T_K ($K=1, 2, 3$). Here Q_3 is the electric charge operator and, on the usual assignments, T_K is the isospin operator. For both K- and ν-particles T_K consists of two doublets ($T=\frac{1}{2}$) and Q_K of a triplet and a singlet ($Q=1, 0$).

The above results follow simply from the symmetries of the wave equation; interactions have not been considered as yet. It seems of considerable interest, therefore, to note that from a discussion of the empirical properties of weak interaction, Lee and Yang (1960) came to the conclusion that four new bosons should exist with exactly the same properties as have been predicted from my wave equation. Lee and Yang's 'schizoid'‡ isospin behaviour as doublets, or as triplet and singlet, in particular is described in exactly this manner in terms of eigenvectors diagonalizing the operators T_3 and T^2, or Q_3 and Q^2, respectively.

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27th March 1961.

H. FRÖHLICH.

FRÖHLICH, H., 1960 a, *Helv. Phys. Acta*, **33,** 803.
—— 1960 b, *Proc. Roy. Soc.* A, **257,** 283.
LEE, T. D., and YANG, C. N., 1960, *Phys. Rev.*, **119,** 1410.

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† From a more detailed analysis than given previously, a completely geometrical interpretation of the neutrino has now been obtained; quantization of its field according to the Pauli principle follows as a necessary consequence (*Nuclear Physics*, in the press).

‡ As an alternative to schizon I propose to name the particles φανχων (from φανοσ and χωγ).

Fig. 5.9 Prediction of new mesons, 1961 [F111], which he proposed to name after his wife—Reproduced with permission of the Institute of Physics *one as the fourth*—he went beyond his earlier phenomenological treatment, showing that continuous *space* reflections could be represented by non-linear transformations of a triad of 3-dimensional vectors, and that these transformations were equivalent to rotations of an associated *tetrad* in a 4-dimensional space.[62] It was found that the Lorentz invariance of the definitions of an unreflected and a fully reflected triad could only be maintained if, from a relativistic point of view, the triad has *axial* symmetry (Fig. 5.10).

Further study in this paper of the angular structure of the new space revealed that its properties could be interpreted solely in terms of homogeneous Lorentz transformations. Correspondingly, it was found that the structure was such as to permit the introduction of wave-equations not only for the light quanta treated previously, but also (quite remarkably) for *neutrinos* as well. The equation for the latter turned out to be *identical* to that which he had first mentioned in the '*Note added in proof*' to [F108], and then obtained in 1961 from consideration of the implications of his continuous treatment of reflections on the structure of *momentum space* [F110]. This earlier work had yielded a completely geometrical interpretation of the neutrino, in which neutrino charge found interpretation as a coordinate in terms of which the *distinction* between left and right-handedness could be described, quantization of its field according to the Pauli principle following as a necessary consequence (Fig. 5.11).[63]

The possibility of a similar geometrical interpretation of the wave-equations for massive particles was considered to require, however, an essential extension to the purely angular structure admitted hitherto, involving the introduction of a *length*, consistent with the close connection between wave-equations and translations—i.e. inhomogeneous Lorentz transformations.

Four years later, hoping that the necessary geometrical concepts might already be available in electrodynamics, he showed that all 10 operators required for the definition of local generators of inhomogeneous Lorentz transformations exist in quantum (but *not* in classical) electrodynamics, since the required operators, which are based in a non-local way on the vector potential, do *not* commute with the fields they rotate and translate; consequently, field quantization no longer needed to be considered as a purely empirical feature but was seen, for the first time, to be actually imposed by *geometrical* requirements [F121]. In turn, the Maxwell equations acquired a corresponding geometrical significance, expressing in the local limit the invariance of the current density J^{μ} under translations; J^{μ} itself was expressed as the local limit of the derivative (with respect to an appropriate non-local coordinate) of a non-local scalar field that vanishes locally. Accordingly, the

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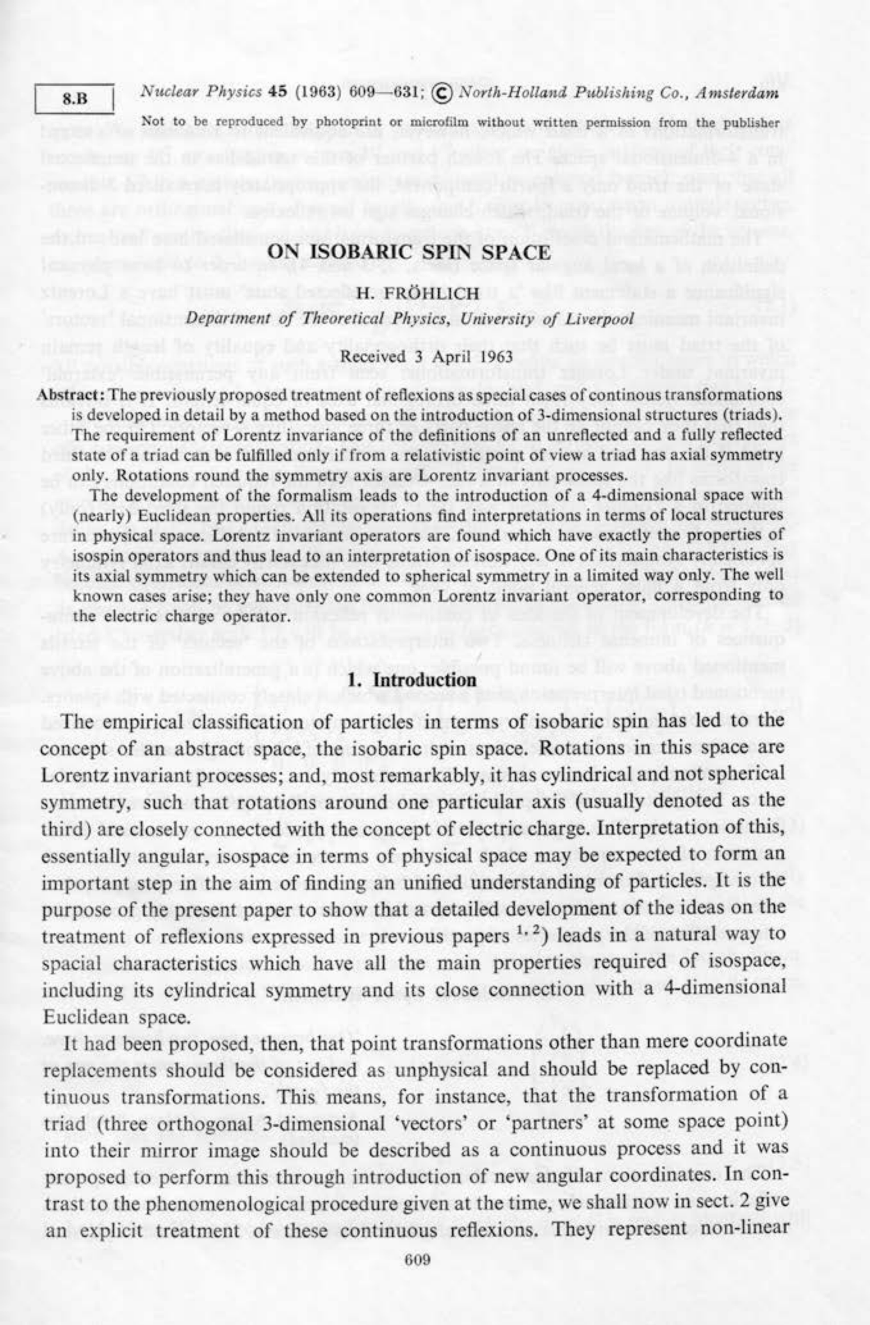
[62] At the time, the tetrad was introduced in a purely ad hoc way. Only much later, during the 1980s, did Fröhlich realise that such a tetrad, with the *same,* remarkable relativistically invariant properties, already exists in the case of Dirac particles, where it is constituted by the 16 bilinear covariants (vide Sect. 6.4).

[63] In view of later developments [F193], it is of interest to note that it was in [F110] that Fröhlich first introduced *bilocal* extensions of the bilinear Dirac covariants, motivated by the bilocal structure of the commutators of the associated particle fields.



8.B *Nuclear Physics* **45** (1963) 609—631; © *North-Holland Publishing Co., Amsterdam*

ON ISOBARIC SPIN SPACE

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Abstract: The previously proposed treatment of reflexions as special cases of continous transformations is developed in detail by a method based on the introduction of 3-dimensional structures (triads). The requirement of Lorentz invariance of the definitions of an unreflected and a fully reflected state of a triad can be fulfilled only if from a relativistic point of view a triad has axial symmetry only. Rotations round the symmetry axis are Lorentz invariant processes.

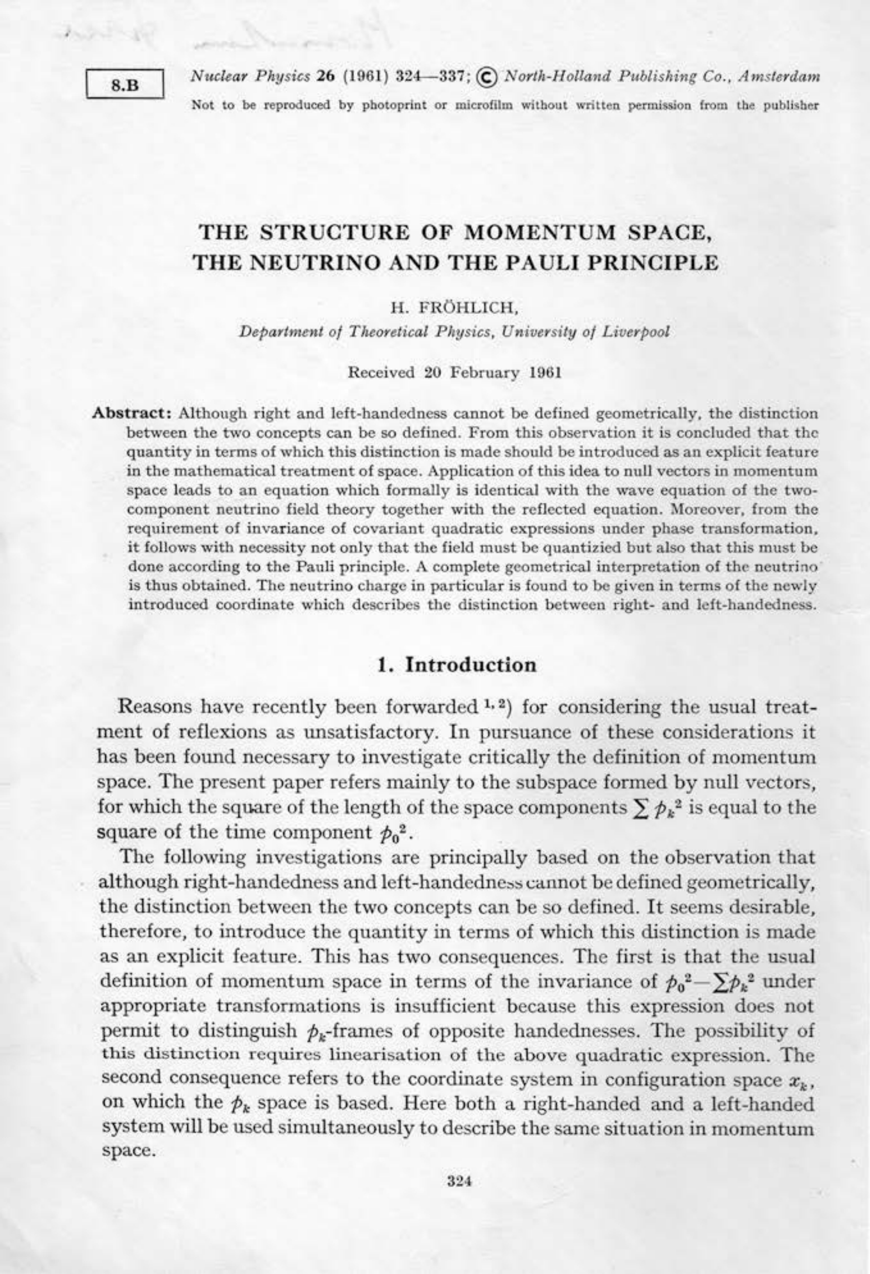
The development of the formalism leads to the introduction of a 4-dimensional space with (nearly) Euclidean properties. All its operations find interpretations in terms of local structures in physical space. Lorentz invariant operators are found which have exactly the properties of isospin operators and thus lead to an interpretation of isospace. One of its main characteristics is its axial symmetry which can be extended to spherical symmetry in a limited way only. The well known cases arise; they have only one common Lorentz invariant operator, corresponding to the electric charge operator.

1. Introduction

The empirical classification of particles in terms of isobaric spin has led to the concept of an abstract space, the isobaric spin space. Rotations in this space are Lorentz invariant processes; and, most remarkably, it has cylindrical and not spherical symmetry, such that rotations around one particular axis (usually denoted as the third) are closely connected with the concept of electric charge. Interpretation of this, essentially angular, isospace in terms of physical space may be expected to form an important step in the aim of finding an unified understanding of particles. It is the purpose of the present paper to show that a detailed development of the ideas on the treatment of reflexions expressed in previous papers [1,2]) leads in a natural way to spacial characteristics which have all the main properties required of isospace, including its cylindrical symmetry and its close connection with a 4-dimensional Euclidean space.

It had been proposed, then, that point transformations other than mere coordinate replacements should be considered as unphysical and should be replaced by continuous transformations. This means, for instance, that the transformation of a triad (three orthogonal 3-dimensional 'vectors' or 'partners' at some space point) into their mirror image should be described as a continuous process and it was proposed to perform this through introduction of new angular coordinates. In contrast to the phenomenological procedure given at the time, we shall now in sect. 2 give an explicit treatment of these continuous reflexions. They represent non-linear

Fig. 5.10 Paper on isobaric spin space [F116]—Reproduced with the permission of Elsevier



8.B

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THE STRUCTURE OF MOMENTUM SPACE, THE NEUTRINO AND THE PAULI PRINCIPLE

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Received 20 February 1961

Abstract: Although right and left-handedness cannot be defined geometrically, the distinction between the two concepts can be so defined. From this observation it is concluded that the quantity in terms of which this distinction is made should be introduced as an explicit feature in the mathematical treatment of space. Application of this idea to null vectors in momentum space leads to an equation which formally is identical with the wave equation of the two-component neutrino field theory together with the reflected equation. Moreover, from the requirement of invariance of covariant quadratic expressions under phase transformation, it follows with necessity not only that the field must be quantizied but also that this must be done according to the Pauli principle. A complete geometrical interpretation of the neutrino is thus obtained. The neutrino charge in particular is found to be given in terms of the newly introduced coordinate which describes the distinction between right- and left-handedness.

1. Introduction

Reasons have recently been forwarded [1,2]) for considering the usual treatment of reflexions as unsatisfactory. In pursuance of these considerations it has been found necessary to investigate critically the definition of momentum space. The present paper refers mainly to the subspace formed by null vectors, for which the square of the length of the space components $\sum p_k^2$ is equal to the square of the time component p_0^2.

The following investigations are principally based on the observation that although right-handedness and left-handedness cannot be defined geometrically, the distinction between the two concepts can be so defined. It seems desirable, therefore, to introduce the quantity in terms of which this distinction is made as an explicit feature. This has two consequences. The first is that the usual definition of momentum space in terms of the invariance of $p_0^2 - \sum p_k^2$ under appropriate transformations is insufficient because this expression does not permit to distinguish p_k-frames of opposite handednesses. The possibility of this distinction requires linearisation of the above quadratic expression. The second consequence refers to the coordinate system in configuration space x_k, on which the p_k space is based. Here both a right-handed and a left-handed system will be used simultaneously to describe the same situation in momentum space.

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Fig. 5.11 Paper on the structure of momentum space, the neutrino and the Pauli principle [F110] —Reproduced with the permission of Elsevier

Maxwell equations themselves had now to be considered as the local limit of a *bilocal* theory.[64] He later noted at the end of [F124] that same conclusion must indeed be drawn from the usual presentation of electric current densities in particle physics. For these densities are quadratic in the particle field operators, $\psi(x)$, whence, in consequence of the singularities involved their commutators, expressions like $\psi^{\dagger}(x)Q\,\psi(x)$, (where Q is an operator) have no meaning *except* in terms of the limit of $\psi^{\dagger}(x')Q\,\psi(x)$ as $x \to x'$.

Attributing particular significance to uniform dilations, which change the metric but leave Maxwell's equations invariant, he concluded [F121] with the following profound statement, of particular relevance to his more general programme of geometrisation, and which reveals his concern about the origin of frames of reference:

> Such invariance must also be demanded of any basic theory of particles; for establishment of a metric would require the existence of measuring instruments of length. They would consist of particles, whose existence cannot be postulated in a theory whose aim would be to derive this existence as one of its main consequences [F121].

Further consideration of these bilocal aspects of electrodynamics subsequently led to the introduction [F124] of a non-local generator of dual transformations under which the Maxwell equations *with* sources, J^{μ}, remain invariant—in contrast to the usual local treatment where $J^{\mu} = 0$ is necessary for such invariance. The new non-local generator involved a 4-dimensional integral over an infinitesimal region of a quantity closely connected with the scalar product ***E.B*** of the electric and magnetic fields; it had integer eigenvalues and represented a new quantized property of the electromagnetic field, the precise nature of which remains to be established.

In conclusion, it must be acknowledged that, unlike the situation with Fröhlich'contributions in many other areas of theoretical physics, references to these works in the literature are conspicuous by their absence, from which it can only be concluded that, despite their predictions and undoubted ingenuity, they had little or no impact or influence on the future developments in this field; whether they were too radical, too little understood, or simply too far removed from contemporary fashions to merit serious attention must remain the subject of speculation. Many years later, however, considerable interest was expressed by Russian physicists, some of whom (including L.B. Okun) Fröhlich met during a visit to Moscow in 1983.

---

[64] During the 1970s, Fröhlich showed that the Maxwell equations *with* sources can be indeed obtained as the local limit of a set of homogeneous (source-*free*), bilocal equations satisfied by the Maxwell spinor. In bilocal extension, this complex spinor field acquires an additional component, which acts as a potential for the sources. This work was never published on account of the difficulties connected with eliminating the *magnetic* sources that necessarily accompany the electric 4-current density in the local limit; it was, however, summarised in the author's extension of the idea to the gravitational field (Hyland 1979).

Appendices

Feynman's Letter to Fröhlich concerning the Polaron, and Fröhlich's Reply

13 SEP 1954

CALIFORNIA INSTITUTE OF TECHNOLOGY
PASADENA

NORMAN BRIDGE LABORATORY OF PHYSICS

September 8, 1954

Dr. H. Fröhlich
Department of Theoretical Physics
University of Liverpool
Liverpool, England

Dear Frohlich:

We have very pretty librarians. Last week I went in to enjoy the scenery and picked up Advances of Physics as a cover. I read your article. I became more interested in it than in the librarians and resolved to accept your challenge to find a method uniformly valid over the entire range of α. Enclosed is my solution.

It uses the lagrangian form of quantum mechanics. The references are R. P. Feynman, Rev. Mod. Phys. 20, 367 (1948) and Phys. Rev. 80, 440 (1950), but particularly Phys. Rev. 84, 108 (1951) Appendix C only. It could be translated no doubt to conventional methods, but I have not done so.

Can you tell me if the problem has already been solved, or whether I should publish the results. What I sent you is a rough first draft of a paper. I would like your comments before I publish, so I am waiting for your answer (which I hope comes soon).

The absolute accuracy of the result can be tested in two ways. At small α the exact coefficient of α^2 can be obtained from second order perturbation theory. I set it up but haven't done the numerical integrals necessary. For large α^2 one can use more elaborate trial functions in Landau's method to get a better coefficient to the α^2 term. These could be compared to my result. Have either of these two things been done?

Are there any further references I should make to other work? I don't know what Buckingham did. Is it published? I rather suspect from your hints what it was. If I guess right he probably uses a coulomb potential somewhere and does hard work. If he used an harmonic potential it would be much easier and more accurate, and if I guess right about what he did, leads to (34).

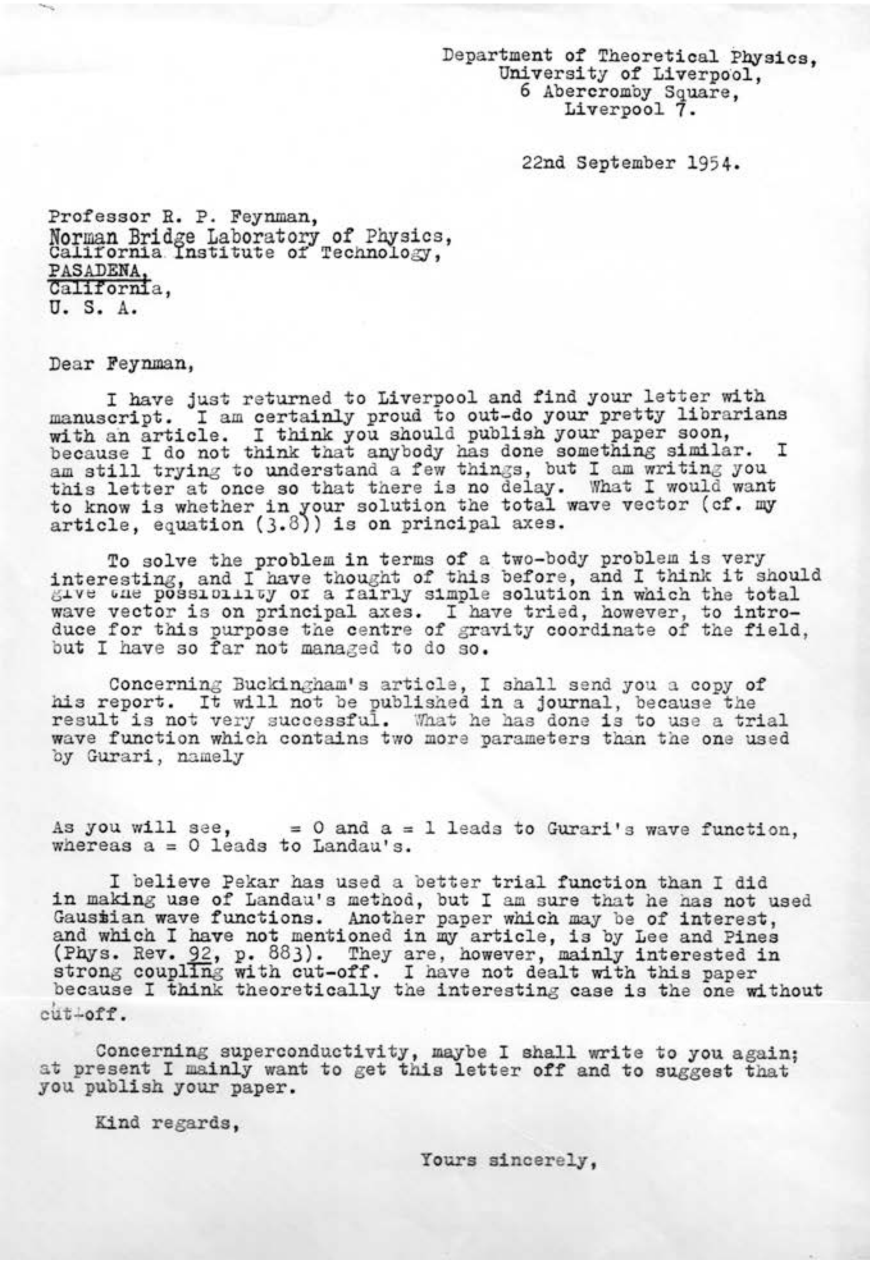
What do we have to do to understand superconductivity?

I hope for an early reply so that I may get the paper out if it is worthwhile.

Sincerely, Dick Feynman.
R. P. Feynman

RPF:n
Enc.

(No harder is case for which lattice vibrations have more than one frequency).



Department of Theoretical Physics,
University of Liverpool,
6 Abercromby Square,
Liverpool 7.

22nd September 1954.

Professor R. P. Feynman,
Norman Bridge Laboratory of Physics,
California Institute of Technology,
PASADENA,
California,
U. S. A.

Dear Feynman,

I have just returned to Liverpool and find your letter with manuscript. I am certainly proud to out-do your pretty librarians with an article. I think you should publish your paper soon, because I do not think that anybody has done something similar. I am still trying to understand a few things, but I am writing you this letter at once so that there is no delay. What I would want to know is whether in your solution the total wave vector (cf. my article, equation (3.8)) is on principal axes.

To solve the problem in terms of a two-body problem is very interesting, and I have thought of this before, and I think it should give the possibility of a fairly simple solution in which the total wave vector is on principal axes. I have tried, however, to introduce for this purpose the centre of gravity coordinate of the field, but I have so far not managed to do so.

Concerning Buckingham's article, I shall send you a copy of his report. It will not be published in a journal, because the result is not very successful. What he has done is to use a trial wave function which contains two more parameters than the one used by Gurari, namely

As you will see, = 0 and a = 1 leads to Gurari's wave function, whereas a = 0 leads to Landau's.

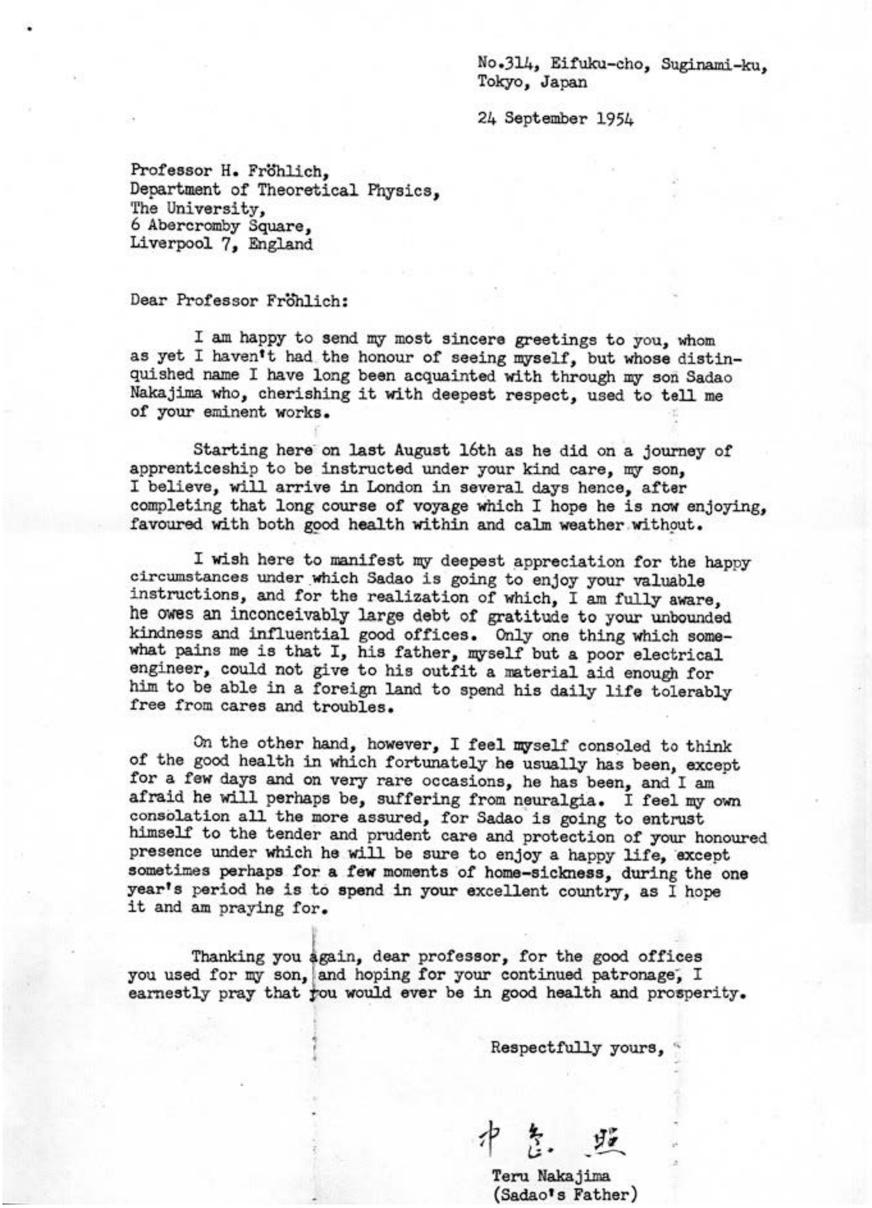
I believe Pekar has used a better trial function than I did in making use of Landau's method, but I am sure that he has not used Gaussian wave functions. Another paper which may be of interest, and which I have not mentioned in my article, is by Lee and Pines (Phys. Rev. 92, p. 883). They are, however, mainly interested in strong coupling with cut-off. I have not dealt with this paper because I think theoretically the interesting case is the one without cut-off.

Concerning superconductivity, maybe I shall write to you again; at present I mainly want to get this letter off and to suggest that you publish your paper.

Kind regards,

Yours sincerely,

An Appreciative Letter from Japan



No.314, Eifuku-cho, Suginami-ku,
Tokyo, Japan

24 September 1954

Professor H. Fröhlich,
Department of Theoretical Physics,
The University,
6 Abercromby Square,
Liverpool 7, England

Dear Professor Fröhlich:

I am happy to send my most sincere greetings to you, whom as yet I haven't had the honour of seeing myself, but whose distinquished name I have long been acquainted with through my son Sadao Nakajima who, cherishing it with deepest respect, used to tell me of your eminent works.

Starting here on last August 16th as he did on a journey of apprenticeship to be instructed under your kind care, my son, I believe, will arrive in London in several days hence, after completing that long course of voyage which I hope he is now enjoying, favoured with both good health within and calm weather without.

I wish here to manifest my deepest appreciation for the happy circumstances under which Sadao is going to enjoy your valuable instructions, and for the realization of which, I am fully aware, he owes an inconceivably large debt of gratitude to your unbounded kindness and influential good offices. Only one thing which somewhat pains me is that I, his father, myself but a poor electrical engineer, could not give to his outfit a material aid enough for him to be able in a foreign land to spend his daily life tolerably free from cares and troubles.

On the other hand, however, I feel myself consoled to think of the good health in which fortunately he usually has been, except for a few days and on very rare occasions, he has been, and I am afraid he will perhaps be, suffering from neuralgia. I feel my own consolation all the more assured, for Sadao is going to entrust himself to the tender and prudent care and protection of your honoured presence under which he will be sure to enjoy a happy life, except sometimes perhaps for a few moments of home-sickness, during the one year's period he is to spend in your excellent country, as I hope it and am praying for.

Thanking you again, dear professor, for the good offices you used for my son, and hoping for your continued patronage, I earnestly pray that you would ever be in good health and prosperity.

Respectfully yours,

中島 照

Teru Nakajima
(Sadao's Father)

Photo Gallery

The numbers in round brackets at the end of each caption correspond to the section of the text to which the photographs refer.

Unless stated otherwise, all photographs are from private collections.

Fig. 5.12 The first home of Theoretical Physics, at 6 Abercromby Square, Liverpool (the white building), 1948–59 (Sect. 5.1)

Fig. 5.13 The first members of the Department of Theoretical Physics, *c*.1951. *Back row* from the *left* Pelzer, Newns, Zienau, Bauer, O'Dwyer, Le Couter, Clark, Huby; *middle row* from the *left* Fröhlich, Bhatia, Szigeti, Miss Ruddleson; *front row* Mrs Zienau, Mrs Szigeti, Avril Rees (Fröhlich's secretary, who later married Huang)—Reproduced through the courtesy of Dr. H. Newns (Sect. 5.1)

Fig. 5.14 The Three Professors: *Left to Right*: Huang, Fröhlich, Bhatia, *c*.1951—Reproduced by courtesy of Dr. H. Newns (Sect. 5.1)

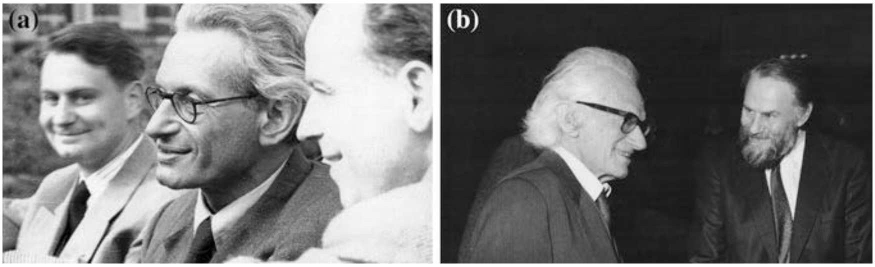


Fig. 5.15 **a** Fröhlich and Haken (*LHS*) in Liverpool, sometime in the mid-1950s; **b** Fröhlich and Haken, many years later in 1985, at the Stuttgart Max Planck Institute, on the occasion of Fröhlich’s 80th birthday (Sect. 5.1)

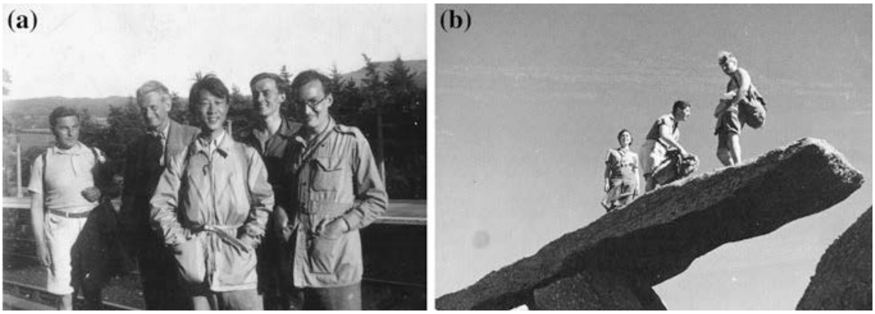


Fig. 5.16 **a** Departmental outing to North Wales: Huang and Huby are in the front row, mid-1950s. **b** Climbing, later that day—Reproduced by courtesy of Dr. H. Newns (Sect. 5.1)

Fig. 5.17 The Chadwick Tower—the top 3 floors of which were the home of Theoretical Physics, 1959–2003: Fröhlich's office was the right-hand half of the *top floor* on the side shown in the photograph, overlooking the River Mersey (Sect. 5.1)

Fig. 5.18 *Left to Right*: Fröhlich, Pelzer and Zienau in Liverpool, 1950 (Sect. 5.3)

Fig. 5.19 **a** Fanchon Fröhlich in Paris, around the time of her marriage in 1950. **b** At another location in Paris (Sect. 5.4)

Fig. 5.20 **a** Arriving in Japan for a conference, 1953—Fröhlich and Feynman are clearly identifiable. **b** Conference delegates with Fröhlich and Feynman (Sect. 5.4)

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Chapter 6
The Liverpool Years: From Professor to Professor Emeritus

6.1 Superconductivity Revisited

In 1963, Fröhlich returned to superconductivity, this time in connection with the then recent finding that in the case of transition metals characterised by incompletely filled *d*-bands, the isotope effect is much weaker than the $M^{-\frac{1}{2}}$ dependence that characterises non-transition metals, and, in some cases, is absent altogether (Matthias et al. 1963). He pointed out that this is actually to be expected if the energy bands associated with the incompletely filled inner shells are sufficiently narrow. For the $M^{-\frac{1}{2}}$ dependence of T_c arises from the assumption that only a small fraction $(\sim \hbar\omega_D/E_F)$ of conduction electrons avail themselves of the attractive interaction, where ω_D is the Debye frequency. When the band-width is itself of the order of $\hbar\omega_D$, however, nearly *all* the electrons partake in the interaction, and the isotope effect is lost [F119].

In the same paper, [F119], he went on to make some comments concerning the magnitude of the dimensionless parameter F that characterised the strength of the electron-phonon interaction in his original work [F76] of 1950. F is defined (Eq. 5.4.3) by $C^2/3E_FMs^2$, where C is a parameter with the dimensions of energy that characterises Bloch's electron-phonon matrix elements, E_F is the Fermi energy, s the speed of sound, and M the ion mass. In the case of inner shells with fairly localised wave-functions, the usual perturbative treatment of the electron-lattice interaction in terms of ionic displacements cannot be expected to be valid, since those parts of the electronic wave-function near the ions should follow the ion displacements almost adiabatically. Recognising this, it became clear that a significant amount of the electron-phonon interaction is incorporated *already in lowest order*, leaving rather little for scattering; accordingly, the resultant interaction parameter, C, must itself be expected to be small in the case of a narrow band. Fröhlich pointed out that if C and E_F vary in the *same* way with band-width, W, then $F \propto W$ – a result that is in sharp contrast to

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The erratum of this chapter can be found under DOI 10.1007/978-3-319-14851-9_8

G.J. Hyland, *Herbert Fröhlich*,
Springer Biographies, DOI 10.1007/978-3-319-14851-9_6

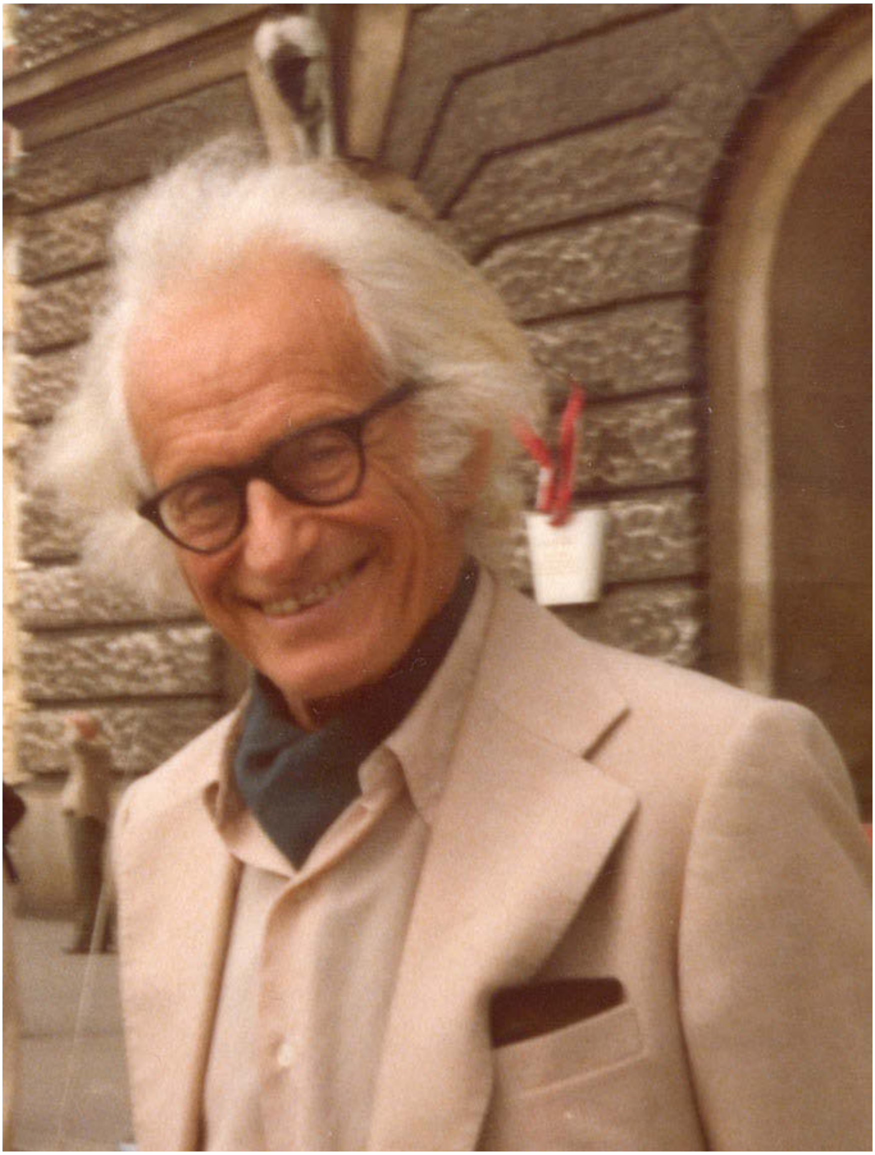

Fig. 6.1 Fröhlich in Vienna, at a *l'Institut de la Vie* conference in 1975

the dependence sometimes inferred within the context of the *BCS* theory, namely $F \propto 1/W$.

Using the modified tight-binding method that had been developed specifically to incorporate as much as possible of the electron-lattice interaction already in lowest order (vide Sect. 5.3), he showed 5 years later, in collaboration with Mitra [F132], that $C \propto W$, whence $F \propto W$, as conjectured earlier. Thus, for transition metals characterised by incompletely filled narrow *d*-bands with small *W*, the phonon-induced electron-electron interaction is indeed much *smaller* than generally believed, which, of course, does not favour superconductivity (Fig. 6.1).

A few years later, he identified a mechanism via which superconductivity in materials with incomplete inner shells can actually occur *without* the involvement of the lattice, and so would automatically account for the absence of an isotope effect, as found in some cases [F130, 131]. This mechanism is based on a screening of the *d*-band plasma by the more mobile *s*-electrons,[1] such that for long waves the frequency, ω_d of the *d*-plasma becomes proportional to wave number. This new 'acoustic-like' mode[2] then entails an attractive interaction between the *s*-electrons in the same way as does the ordinary acoustic lattice mode, and hence the possibility of superconductivity[3] (Fig. 6.2).

The possibility of transforming the *d*-plasma mode into an 'acoustic-like' mode follows, under certain conditions, from consideration of the frequency and wave-vector dependent dielectric function $\varepsilon\,(\boldsymbol{q}, \omega)$ of the *s*-*d* system, in terms of which the modification of the interaction between any two electrons by the remainder can be approximated. Neglecting any lattice contribution, $\varepsilon\,(\boldsymbol{q}, \omega)$ can be written as follows:

$$\varepsilon(\boldsymbol{q}, \omega) = \varepsilon_o(\boldsymbol{q}, \omega) + \alpha_s + \alpha_d \tag{6.1.1}$$

where $\alpha_s(\alpha_d)$ is the polarisability of the *s*(*d*) electrons, and $\varepsilon_o(\boldsymbol{q}, \omega)$ is the contribution from band-to-band transitions.

In order that the *s*-electrons screen the *d*-plasma, it is necessary that α_s be positive, whilst α_d must be negative, so that the *d*-electrons anti-screen; this requires that the following inequalities hold:

$$q^2 v_d^2 < \omega^2 < q^2 v_s^2 \tag{6.1.2}$$

where $v_s(v_d)$ are the mean electron velocities at the Fermi surface.

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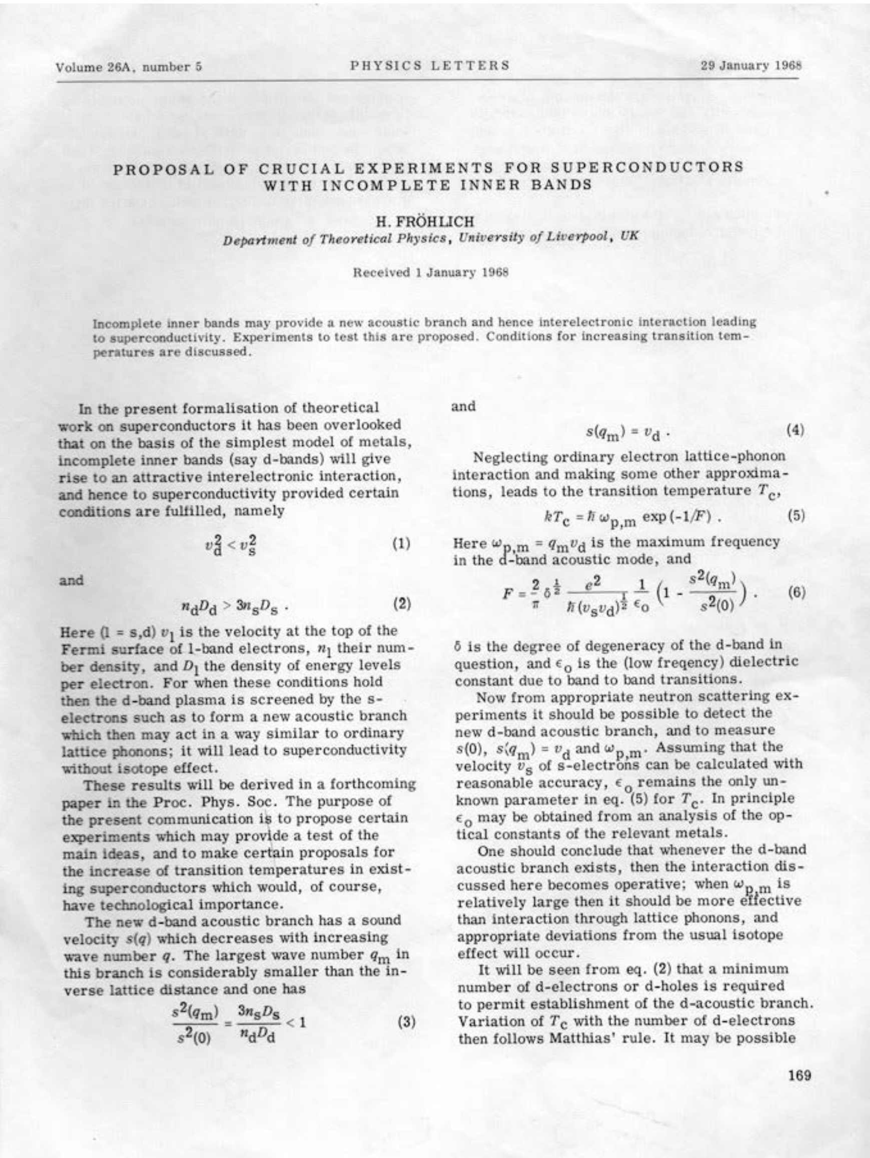
[1] This treatment in terms of two plasmas is certainly consistent with the empirical finding that in transition elements that exhibit superconductivity, the *d*-orbitals overlap to a much greater extent than they do in those transition elements that exhibit magnetic order instead.

[2] He suggested that this 'acoustic' mode—the possibility of which appears to have been first alluded to by Pines (1956)—should be detectable using neutron scattering [F130]. Some years later, he estimated the effect of friction, finding the mode to be strongly damped, but nevertheless observable at sufficiently low temperatures at wavelengths less than the electronic mean free path [F144].

[3] At this time, Fröhlich was unaware of a similar suggestion made independently 3 years earlier by Radhakrishnan (1965).



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PROPOSAL OF CRUCIAL EXPERIMENTS FOR SUPERCONDUCTORS WITH INCOMPLETE INNER BANDS

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Incomplete inner bands may provide a new acoustic branch and hence interelectronic interaction leading to superconductivity. Experiments to test this are proposed. Conditions for increasing transition temperatures are discussed.

In the present formalisation of theoretical work on superconductors it has been overlooked that on the basis of the simplest model of metals, incomplete inner bands (say d-bands) will give rise to an attractive interelectronic interaction, and hence to superconductivity provided certain conditions are fulfilled, namely

$$v_{\mathrm{d}}^2 < v_{\mathrm{s}}^2 \tag{1}$$

and

$$n_{\mathrm{d}} D_{\mathrm{d}} > 3 n_{\mathrm{s}} D_{\mathrm{s}} \,. \tag{2}$$

Here (l = s,d) v_{l} is the velocity at the top of the Fermi surface of l-band electrons, n_{l} their number density, and D_{l} the density of energy levels per electron. For when these conditions hold then the d-band plasma is screened by the s-electrons such as to form a new acoustic branch which then may act in a way similar to ordinary lattice phonons; it will lead to superconductivity without isotope effect.

These results will be derived in a forthcoming paper in the Proc. Phys. Soc. The purpose of the present communication is to propose certain experiments which may provide a test of the main ideas, and to make certain proposals for the increase of transition temperatures in existing superconductors which would, of course, have technological importance.

The new d-band acoustic branch has a sound velocity $s(q)$ which decreases with increasing wave number q. The largest wave number q_{m} in this branch is considerably smaller than the inverse lattice distance and one has

$$\frac{s^2(q_{\mathrm{m}})}{s^2(0)} = \frac{3 n_{\mathrm{s}} D_{\mathrm{s}}}{n_{\mathrm{d}} D_{\mathrm{d}}} < 1 \tag{3}$$

and

$$s(q_{\mathrm{m}}) = v_{\mathrm{d}} \,. \tag{4}$$

Neglecting ordinary electron lattice-phonon interaction and making some other approximations, leads to the transition temperature T_{c},

$$kT_{\mathrm{c}} = \hbar\, \omega_{\mathrm{p,m}} \exp(-1/F) \,. \tag{5}$$

Here $\omega_{\mathrm{p,m}} = q_{\mathrm{m}} v_{\mathrm{d}}$ is the maximum frequency in the d-band acoustic mode, and

$$F = \frac{2}{\pi} \delta^{\frac{1}{2}} \frac{e^2}{\hbar (v_{\mathrm{s}} v_{\mathrm{d}})^{\frac{1}{2}}} \frac{1}{\epsilon_{\mathrm{o}}} \left(1 - \frac{s^2(q_{\mathrm{m}})}{s^2(0)}\right) . \tag{6}$$

δ is the degree of degeneracy of the d-band in question, and ϵ_{o} is the (low freqency) dielectric constant due to band to band transitions.

Now from appropriate neutron scattering experiments it should be possible to detect the new d-band acoustic branch, and to measure $s(0)$, $s(q_{\mathrm{m}}) = v_{\mathrm{d}}$ and $\omega_{\mathrm{p,m}}$. Assuming that the velocity v_{s} of s-electrons can be calculated with reasonable accuracy, ϵ_{o} remains the only unknown parameter in eq. (5) for T_{c}. In principle ϵ_{o} may be obtained from an analysis of the optical constants of the relevant metals.

One should conclude that whenever the d-band acoustic branch exists, then the interaction discussed here becomes operative; when $\omega_{\mathrm{p,m}}$ is relatively large then it should be more effective than interaction through lattice phonons, and appropriate deviations from the usual isotope effect will occur.

It will be seen from eq. (2) that a minimum number of d-electrons or d-holes is required to permit establishment of the d-acoustic branch. Variation of T_{c} with the number of d-electrons then follows Matthias' rule. It may be possible

Fig. 6.2 Fröhlich's plasma model for superconductivity in materials with incomplete inner shells [F130]—Reproduced with the permission of Elsevier

The frequency, ω_p, of the longitudinal electric collective mode of the system is given by the solution of $\varepsilon(\boldsymbol{q}, \omega) = 0$, which, under the condition Eq. 6.1.2, takes the form:

$$\omega_p^2 = q^2 s^2(q) \tag{6.1.3}$$

where the velocity s is given by:

$$s^2(q) = \frac{\omega_d^2 f_d}{q^2 \varepsilon_o(\boldsymbol{q}) + 3\,\omega_s^2 f_s / v_s^2} \tag{6.1.4}$$

$\omega_s(\omega_d)$ are the unscreened plasma frequencies, which are given by:

$$\omega_s^2 = 4\pi n_s e^2 / m_s, \text{ and similarly for } \omega_d^2 \tag{6.1.5}$$

f are the oscillator strengths for each band; in terms of f and the density of energy levels per electron, D, at the relevant Fermi surface, the mean Fermi velocities, v, are given by:

$$mv^2 = 3f/D \tag{6.1.6}$$

Equations 6.1.3 and 6.1.4 show how the d-plasma is transformed into an acoustic-like mode through the screening effect of the more mobile s-electrons; ω_p becomes linear in q as $q \longrightarrow 0$ (long waves), whilst Eq. 6.1.4 shows that the 'sound' velocity, $s(q)$, decreases with increasing q, becoming equal to v_d at the maximum wave-vector, q_m, of the 'acoustic' mode—i.e. $s(q_m) = v_d$.

It can be shown that $\omega_p{}^{\max}(\equiv q_m v_d)$ is given by:

$$\omega_p^{2\,\max} = \omega_d^2 f_d \gamma^2, \tag{6.1.7}$$

where γ contains the effect of the screening, and can be written in the following form:

$$\gamma^2 \equiv \varepsilon_o^{-1}(1 - 3n_s D_s / n_d D_d) \tag{6.1.8}$$

from which it follows that:

$$n_d D_d > 3 n_s D_s, \tag{6.1.9}$$

which imposes a lower limit on n_d.

Under condition Eq. 6.1.2, and in terms of $\omega_p, \varepsilon(\boldsymbol{q}, \omega)$ takes the form:

$$\varepsilon(\boldsymbol{q}, \omega) = \varepsilon_1(q)(1 - \omega_p^2/\omega^2) \tag{6.1.10}$$

where $\varepsilon_1(q)$ represents $\varepsilon_o(q)$ modified by the s-screening effect. For $\omega^2 < \omega_p^2$, $\varepsilon(\boldsymbol{q}, \omega) < 0$—i.e. there is an *attractive* interaction between any two s-electrons, and hence Cooper pairing resulting in a superconductive state as in the case of the conventional *BCS* theory based on the phonon-mediated electron-electron attraction, as described in Sect. 5.4 of Chap. 5.

An appropriate adaptation of the *BCS* expression for the superconducting transition temperature, T_c, yields:

$$kT_c = \hbar\omega_\mathrm{p}{}^{\max}\exp(-1/F) \tag{6.1.11}$$

where F can be shown to be given by:

$$F = \frac{2\delta^{1/2}e^2}{\pi\hbar(v_\mathrm{s}v_\mathrm{d})^{1/2}}\gamma^2 \tag{6.1.12}$$

where δ is the degeneracy of the *d*-band.

Fröhlich noted that the prefactor of the exponential in the above expression for T_c, $\omega_\mathrm{p}{}^{\max}$, is (via Eqs. 6.1.5 and 6.1.7) proportional to the *effective* number of free *d*-electrons, $n_\mathrm{d}f_\mathrm{d}$, which increases proportionally with n_d for nearly empty bands, reaches a maximum, and becomes zero for filled bands; the values of n_d itself, are however, restricted by Eq. 6.1.9. In addition, the magnitude of $\omega_\mathrm{p}{}^{\max}$ is (via the screening factor γ^2) dependent on the value of ε_o, which is quite possibly much larger than unity, so that $\gamma^2 \ll 1$, whence $\omega_\mathrm{p}{}^{\max} \ll \omega_\mathrm{d}$. Assuming now, in the usual way, that the 5-fold degenerate *d*-band splits into a triplet (t_{2g}) and a doublet (e_g), then '……$\omega_\mathrm{p}{}^{\max}$ represents a curve very similar to Matthias' well-known curve[4] of the dependence of T_c on the filling of the *d*-shell.' [F130]. He went on the note that in Eq. 6.1.11, the screening factor γ^2 occurs also in the interaction constant F, '….. thus accentuating the feature that metals with nearly empty or nearly full *d*-bands do not become superconductive on the basis of the interaction discussed at present, although they may become so at a very low temperature through the [usual] lattice-phonon-induced interaction' [F130].

Clearly, the model just described exhibits no isotope effect, since $\omega_\mathrm{p}{}^{\max}$ does not contain the ion mass.

He further noted that the above expression for T_c predicts (via the n_s dependence of γ^2) that T_c increases as the number of *s*-electrons is decreased (but only up to a certain limit), and suggested how this might be realised by the introduction of a certain amount of ionic bond into a superconductive transition metal or alloy by using admixtures of ionic compounds such as oxides wherein the number of *s*-electrons vanishes [F130, 131]. This suggestion can only be considered to have been highly prescient, given the subsequent discovery in the 1980s of the new high temperature superconductors based on oxides containing incomplete inner shells, such as La_2CuO_4, doped with divalent ions such as Ba, Ca, or Sr.

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[4] See Matthias (1955)

To Fröhlich, the appeal of this simple model was that both the indirect electron-electron attraction necessary for superconductivity *and* the direct, screened Coulomb repulsion between electrons, which tends to counteract it, were treated in a *unified* way. To see this, rewrite Eq. 6.1.10 as follows:

$$\varepsilon(\boldsymbol{q},\omega)^{-1} = \varepsilon_1(\boldsymbol{q},\omega)^{-1}\omega_{\mathrm{p}}^2/\left(\omega^2-\omega_{\mathrm{p}}^2\right)+\varepsilon_1(\boldsymbol{q},\omega)^{-1} \tag{6.1.13}$$

When introduced into the expression for the interaction between two electrons, the first term yields the attraction between electrons (mediated by the 'acoustic' *d*-mode), whilst the second represents Coulomb repulsion appropriately screened by the *s*-electrons. The magnitudes of both contributions arise from the *same* type of approximation such that the total is always attractive for $\omega^2 < \omega_{\mathrm{p}}^2$.

Quite generally, some relation between the two must be expected even in the phonon-mediated case, since the underlying electron-phonon interaction is basically Coulombic, and as such must contain considerable electronic contributions, as had been noted already some years earlier [F119]. To be able to show this quantitatively, however, would require approximations that hold equally for both electron-electron *and* electron-ion interactions—a programme that was deemed to be over-ambitious. The *s-d* model, by contrast, presented a case in which both the attraction and repulsion interactions could be treated within the *same* approximation.

Long before the new high T_{c} materials were discovered, Fröhlich's great friend, the experimentalist Bernd Matthias, in his search for high temperature superconductors, had used anomalously low melting points as a guide (Matthias 1967); Fröhlich pointed out that this implied, assuming the Lindemann formula had some validity, relatively low lattice frequencies. It will be recalled from Chap. 5 that already in 1952, in his most influential paper in which he introduced the methods of quantum field theory into solid-state physics, he had shown that the electron-phonon interaction has repercussions not only on the electrons, where it leads to the electron-electron interaction upon which superconductivity depends, but also on the lattice, wherein it entails a renormalisation of the velocity of sound, such that strong electron-phonon interaction implied a *low* sound velocity, which is consistent with a low melting point.

Noting that many of the superconductors with the highest T_{c} known at the time had incomplete inner shells, he decided to study this problem by using a generalisation of the above model based on *s* and *d*-electrons in which the ions were now introduced as an additional, third plasma [F149]. The mutual Coulomb interaction of the three plasmas was now found to produce *two* overlapping ion acoustic branches, one of which is a long wavelength mode to which the *d*-plasma contributes, thereby raising the ion frequency; in the other branch, on the other hand, which, provided the velocity of the *d*-electrons is sufficiently low, is restricted to *short* wavelengths, the *d*-plasma acts to screen the ion oscillation whose frequency is thereby lowered. It is thus found that the short waves, which are most important near melting, are softened by the *same* interaction as is responsible for superconductivity. Although in the short wave region, treatment of the ions as a plasma cannot strictly be justified, the fact that the extraordinary lattice dispersion relations found in niobium using inelastic neutron scattering can be described in terms of hard long waves and short soft waves (Nakagawa and Woods 1963) strongly supports the general idea.

After a detailed examination of available experimental data up to 1970, Rothwarf concluded that there is indeed evidence of acoustic plasmons, and that ‘overall the model gives a satisfactory explanation for a number of diverse superconducting *and* normal properties of transition metals’. Particularly impressive was considered to be the model’s ability to account for the pressure-dependence of the superconducting transition temperature in these materials, where *both* signs of dT_c/dp are found, and for the pressure-induced superconductivity found in the case of some other elements with incomplete inner shells, such as Ce, U and Y (Rothwarf 1970).

Fröhlich’s later approach [F197] to the new high T_c metal-oxide superconductors was similar, in that it focused not on an interaction between electrons and lattice vibrations, but rather on a pairing between *d*-electrons via their interaction with the much higher frequency oscillations of the electrons of the O^{2-} ions, which together with the incompletely filled inner *d*-shell ions and other dopants, characterize the new materials. What led him to this was the fact that some of these materials have the perovskite structure, a structure that is shared by many oxide ferroelectrics wherein the electrons of the O^{2-} ions get displaced by mode softening. Whilst this does not occur in the case of the high T_c perovskites, he suggested that a polar oscillation of these O^{2-} electrons should nevertheless exist, characterised by a frequency ω_o, well above that of the lattice. Adapting the *BCS* formula $kT_c \approx \hbar\omega \exp(-1/F)$ to this possibility, requires that the lattice frequency, ω, be replaced by the much higher frequency ω_o, whilst since F is inversely proportional to the Fermi energy, E_F (vide Eq. 5.4.3), it follows that $F \sim n^{-2/3}$, where n is the number density of *d*-electrons. The possibility of a rather high T_c then arises, provided n is sufficiently low, which, he noted, can be achieved by the introduction of dopant ions; a low value of n does, however, limit the current that can be carried. For an application of this model to a specific metal-oxide, *see* Hyland (1987).

6.2 Statistical Mechanics and the Connection Between Micro and Macrophysics

Welcome as the eventual *BCS* solution to the problem of superconductivity was when it was first published in 1957, Fröhlich did not relinquish the conviction that such a striking phenomenon as superconductivity must surely be derivable from microphysics *without* the necessity of resorting to detailed solution of the many-body problem. His 1961 review [F109] of the theory of superconductivity concluded with this sentence:

> The similarity of substances as different as liquid helium and the electron fluid of superconductors certainly points to some very general features which should be closely connected with quantization in macroscopic physics.

Having read this, Yang immediately wrote to Fröhlich informing him that the sought-after ‘general feature’ was what he had termed ‘off-diagonal-long-range-order’ (*ODLRO*) (Yang 1962); Yang’s work was a development and generalisation of earlier theoretical considerations of O. Penrose for the case of the boson superfluid He^4 (Penrose 1951). *ODLRO* is an extreme non-local property of entities known as reduced density matrices, in which it is reflected by the existence of long-range correlations based on the phases of certain so-called *macroscopic* wave-functions (vide infra), and an associated *coherence* involving dynamic order over macroscopic distances from which follow the remarkable properties of flow.

The reduced density matrices are complex, non-local, time-dependent macroscopic scalar fields, the first two of which are defined as follows:

$$\Omega_1(\boldsymbol{x}',\boldsymbol{x}'';t) \equiv Tr[\psi^\dagger(\boldsymbol{x}'')\psi(\boldsymbol{x}')\Omega(t)], \tag{6.2.1}$$

$$\Omega_2(\boldsymbol{x}',\mathbf{y}';\boldsymbol{x}'',\mathbf{y}'';t) \equiv Tr[\psi^\dagger(\boldsymbol{x}'')\psi^\dagger(\boldsymbol{y}'')\psi(\boldsymbol{y}')\psi(\boldsymbol{x}')\Omega(t)], \tag{6.2.2}$$

where Tr denotes trace, the $\psi(\psi^\dagger)$ are wave operators satisfying non-relativistic Bose or Fermi commutations relations, and $\Omega(t)$ is the density matrix describing the whole unique system, which satisfies (for both bosons and fermions):

$$i\hbar\ \partial_t\Omega(t) = [H, \Omega(t)] \tag{6.2.3}$$

where $[H, \Omega]$ is the commutator of Ω with the Hamiltonian, H, defined by:

$$H = \hbar^2/2m \int \partial\psi(\boldsymbol{x})\partial\psi^\dagger(\boldsymbol{x})d^3\boldsymbol{x} + {}^1\!/_2\iint V(|\boldsymbol{x}-\boldsymbol{y}|)\psi^\dagger(\boldsymbol{x})\psi^\dagger(\boldsymbol{y})\psi(\boldsymbol{y})\psi(\boldsymbol{x})d^3\boldsymbol{x}d^3\boldsymbol{y} \tag{6.2.4}$$

$V(|\boldsymbol{x} - \boldsymbol{y}|)$ is assumed to be a short range potential; in the case of fermions, $\boldsymbol{x}$ and $\boldsymbol{y}$ include both space *and* spin coordinates.

Ω_1 and Ω_2 are related through the following integral condition (where N is the total mean number of particles under consideration) by:

$$\int \Omega_2(\boldsymbol{x}',\mathbf{y};\boldsymbol{x}'',\mathbf{y})d^3\boldsymbol{y} = (N-1)\Omega_1(\boldsymbol{x}',\boldsymbol{x}'') \tag{6.2.5}$$

Ω_1 satisfies $\Omega_1(\boldsymbol{x}',\boldsymbol{x}'') = \Omega_1^*(\boldsymbol{x}'',\boldsymbol{x}')$, and can thus be expressed as follows in terms of two real, non-local fields $\sigma\,(\boldsymbol{x}', \boldsymbol{x}'')$ and $\chi\,(\boldsymbol{x}', \boldsymbol{x}'')$, which are, respectively, even and odd functions of $\boldsymbol{x}'$, $\boldsymbol{x}''$ (so that $\lim_{x'=x''}\chi\,(\boldsymbol{x}',\boldsymbol{x}'') = 0$):

$$\Omega_1(\boldsymbol{x}',\boldsymbol{x}'') = \sigma\,(\boldsymbol{x}',\boldsymbol{x}'')\ \exp\ i\chi\,(\boldsymbol{x}',\boldsymbol{x}'') \tag{6.2.6}$$

The particle density, $\sigma(\boldsymbol{x}, t)$ is defined by

$$\sigma(\boldsymbol{x},t) = \lim \Omega_1(\boldsymbol{x}',\boldsymbol{x}'';t), \tag{6.2.7}$$

and the current density, $\boldsymbol{J}(\boldsymbol{x}, t)$, by:

$$\boldsymbol{J}(\boldsymbol{x},t) = \lim \frac{\hbar}{2mi}(\partial_{x'} - \partial_{x''})\Omega_1(\boldsymbol{x}',\boldsymbol{x}'';t), \tag{6.2.8}$$

where lim denotes the local limit, $\boldsymbol{x}' = \boldsymbol{x}''$.

$\Omega_2(\boldsymbol{x}', \boldsymbol{y}'; \boldsymbol{x}'', \boldsymbol{y}'')$ satisfies:

$$\begin{aligned}\Omega_2(\boldsymbol{x}',\mathbf{y}';\boldsymbol{x}'',\mathbf{y}'') &= \Omega_2^*(\boldsymbol{x}'',\mathbf{y}'';\boldsymbol{x}',\mathbf{y}'),\\ \Omega_2(\boldsymbol{x}',\mathbf{y}';\boldsymbol{x}'',\mathbf{y}'') &= \pm\Omega_2(\boldsymbol{y}',\mathbf{x}';\boldsymbol{x}'',\mathbf{y}''),\\ \Omega_2(\boldsymbol{x}',\mathbf{y}';\boldsymbol{x}'',\mathbf{y}'') &= \pm\Omega_2(\boldsymbol{x}',\mathbf{y}';\boldsymbol{y}'',\mathbf{x}''),\end{aligned} \tag{6.2.9}$$

where $+$ holds for bosons, $-$ for fermions.

Being *spatially* non-local fields, the reduced density matrices were the perfect vehicle with which Fröhlich could pursue his earlier proposal to introduce non-local features into hydrodynamics in order to: (i) be able to describe transverse displacements in a macroscopic continuum [F125], and (ii) overcome difficulties that surrounded the quantization of transverse motion in a continuous fluid, the resolution of which otherwise necessitated the introduction of atomistic structure (Allcock and Kuper 1955). The latter meant, as he noted towards the end of [F109], that '......in quantum mechanics the usual (local) hydrodynamics does not represent a consistent[5] theory', and concluded that '......from this modification conditions might possibly arise under which quantization would lead to a finite energy for the excitation of transverse motion'. Such a gap characterising excitations in superfluid He^4 with non-zero curl, known as rotons,[6] was originally proposed by Landau (1941, 1947), and is a feature that this (boson) system has in common with (fermion) superconductors. Some years later, and just prior to his work with the reduced density matrices, Fröhlich raised the related matter of defining transverse collective coordinates, which, he noted, must necessarily contain a microscopic element [F125].

Reduced density matrices were attractive from another point of view, namely that they 'straddle', so to speak, the micro-macro divide, in the sense that whilst macroscopic quantities, such as density and current fields, can be defined in terms of them (Eqs. 6.2.7 and 6.2.8), their equations of motion are governed by quantum mechanics, i.e. by microphysics. Fröhlich's strategy was that solution of the exact equation of motion (Eq. 6.2.3) for the full density matrix, Ω, should never be attempted, but instead used to generate exact equations of motion of the Ω_n:

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[5] For further consideration of this see [F128].

[6] In German literature 'curl' is denoted as 'rot'—hence rotons!

> Complete solution is not at all desirable. For it would contain such an immense amount of information that extracting from it the 'interesting' macroscopic properties would be a task about equivalent with finding the complete solutions in the first place [F129].

It turns out, however, that these equations of motion are infinitely coupled, in the sense that the equation of motion for Ω_n involves Ω_{n+1}. Thus, for example, the equation of motion for Ω_1 takes the following form, which involves Ω_2:

$$\begin{aligned} i\hbar\, \partial_t \Omega_1(\boldsymbol{x}',\boldsymbol{x}'';t) &+ \hbar^2/2m\left(\partial^2_{\boldsymbol{x}'} - \partial^2_{\boldsymbol{x}''}\right)\Omega_1(\boldsymbol{x}',\boldsymbol{x}'') \\ &= \int [V(|\boldsymbol{x}'-\boldsymbol{y}|) - V(|\boldsymbol{x}''-\boldsymbol{y}|)\Omega_2(\boldsymbol{x}',\mathbf{y};\boldsymbol{x}'',\mathbf{y})d^3\boldsymbol{y} \end{aligned} \tag{6.2.10}$$

Accordingly, some approximations[7] have to be introduced in order to truncate the hierarchy of equations if a *closed* equation of motion of a particular Ω_n is to be obtained.

To gain some experience with this methodology, Fröhlich's first considered the derivation of the macroscopic Navier-Stokes' equations of classical hydrodynamics [F129], assuming a fluid of identical atoms that interact through a short-range, 2-body potential, $V(|\boldsymbol{x}-\boldsymbol{y}|)$. Since the fluid density, σ, and velocity field, $\boldsymbol{v}$, that enter these equations are defined (via Eqs. 6.2.7 and 6.2.8, together with 6.1.12 below) in terms of Ω_1, it sufficed to restrict attention to the equation of motion for Ω_1. The terms in the latter involving Ω_2 do *not* contribute to the local limit of $\partial_t\Omega_1$, which immediately yields, as an *exact* equation, the equation of continuity:

$$\partial_t\sigma(\boldsymbol{x},t) + \operatorname{div}\boldsymbol{J}(\boldsymbol{x},t) = 0, \tag{6.2.11}$$

where the fluid density $\sigma(\boldsymbol{x}$, t) and the current density $\boldsymbol{J}(\boldsymbol{x}, t)$ are as defined by Eqs. 6.2.7 and 6.2.8.

Since $\boldsymbol{J}(\boldsymbol{x}, t) \equiv \sigma(\boldsymbol{x}, t)\boldsymbol{v}(\boldsymbol{x}, t)$, he noted that an operator for the velocity field cannot exist, because it would involve division by the highly singular density operator (a sum of δ-functions); the *c*-number velocity field, $\boldsymbol{v}(\boldsymbol{x}, t)$, however is defined by:

$$\boldsymbol{v}(\boldsymbol{x},t) = \frac{\hbar}{2m}\lim\left(\partial_{\boldsymbol{x}'} - \partial_{\boldsymbol{x}''}\right)\chi(\boldsymbol{x}',\boldsymbol{x}'';t), \tag{6.2.12}$$

where χ is the phase of Ω_1—*see* Eq. 6.2.6. Furthermore, he pointed out that even though the velocity field is a macroscopic quantity, it nevertheless involves Planck's constant, $\hbar$. If the phase $\chi(\boldsymbol{x}', \boldsymbol{x}'')$, factorises into a function of $\boldsymbol{x}'$ only, and a function of $\boldsymbol{x}''$ only, then $\boldsymbol{v}$, is longitudinal (*curl*-free)—i.e. the flow is irrotational; to allow for the possibility of a transverse flow characterised by *curl*$\boldsymbol{v} \neq 0$, it is thus necessary that $\chi(\boldsymbol{x}', \boldsymbol{x}'')$ contains a *non*-factorisable contribution.

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[7] The simplest approximation would be to factorize Ω_2, for example, into appropriately symmetrised products of Ω_1, but this would be valid only in the case of non-interacting particles, or asymptotically, in the case of interacting particles—i.e. at separations greater than the range of their interaction.

The other Navier-Stokes' equation, describing the time-dependence of the fluid velocity field, $\boldsymbol{v}(\boldsymbol{x}, t)$, was obtained from lim $(\partial_{x'} - \partial_{x''})\partial_t \Omega_1$, which yields the following exact equation of motion:

$$m\sigma(\boldsymbol{x})[\partial_t \boldsymbol{v} + \boldsymbol{v}(\boldsymbol{x})\text{grad}\ \boldsymbol{v}(\boldsymbol{x})] = -2\text{div}\,\underline{\underline{T}}^{\text{o}} - \int \{\partial_{\text{x}} V(|\boldsymbol{x} - \boldsymbol{y}|)\} P(\boldsymbol{x},\boldsymbol{y}) d^3 y, \tag{6.2.13}$$

where $\underline{\underline{T}}^{\text{o}}$ is the internal kinetic energy density tensor, with components $T^{\text{o}}_{rs}(\boldsymbol{x})$ defined by:

$$T^{\text{o}}_{rs}(\boldsymbol{x}) \equiv -(\hbar^2/2m)\ \lim(\partial_{x'} - \partial_{x''})_r (\partial_{x'} - \partial_{x''})_s \sigma(\boldsymbol{x}', \boldsymbol{x}''). \tag{6.2.14}$$

$P(\boldsymbol{x}, \boldsymbol{y}) \equiv \Omega_2(\boldsymbol{x}, \mathbf{y}; \boldsymbol{x}, \mathbf{y})$, and satisfies:

$$P(\boldsymbol{x},\boldsymbol{y}) = P(\boldsymbol{y},\boldsymbol{x}). \tag{6.2.15}$$

Whilst the form of the *LHS* of Eq. 6.2.13 corresponds with that of the Navier-Stokes' equation for $\partial_t \boldsymbol{v}$, the *RHS* does not, because of the term involving the integral of the two-body potential, V, with the two-point correlation function $P(\boldsymbol{x}, \boldsymbol{y})$; this term arises from the term involving Ω_2 in the equation of motion for Ω_1. However, by expressing $P(\boldsymbol{x}, \boldsymbol{y})$ in terms of *deviations* from a translationally and rotationally invariant hydrodynamic equilibrium characterised by zero fluid velocity, $\boldsymbol{v} = 0$, a constant fluid density, σ_0, and by a two-point correlation function, P_0, that is a function only of $|\boldsymbol{x} - \boldsymbol{y}|$ ($=r$), Fröhlich showed that retaining only terms that are linear[8] in the deviation from equilibrium, and assuming they vary slowly in comparison with the 2-body potential V, then the *RHS* of Eq. 6.2.13 assumes the form of the Navier-Stokes' (partial differential) equation—i.e.

$$-\text{grad}\ p(\mathbf{x}) - \eta_1 \text{curlcurl}\ \boldsymbol{v}(\boldsymbol{x}) + \eta_2 \text{grad div}\ \boldsymbol{v}(\boldsymbol{x}), \tag{6.2.16}$$

where $p(\boldsymbol{x}) \equiv 2/3\ T^{\text{o}} + p_1$. It is to be emphasised that the non-kinetic part, p_1, of the pressure, p, and the viscous coefficients, η_1 and η_2, are given only *formally* by integrals involving the 2-body potential and the parameters that characterise the deviation from equilibrium. Calculation of magnitudes and temperature-dependences of these *material-specific* quantities requires more explicit knowledge of $P(\boldsymbol{x}, \boldsymbol{y})$ and of the 2-body potential; this is a task for microscopic theory, and was beyond the scope of his programme, as he stressed.

The general structure of the Navier Stokes' equations, on the other hand, is *not* material-specific, but is satisfied by *all* fluid systems near equilibrium, being essentially a consequence of the symmetry of the two-point correlation function, $P(\boldsymbol{x}, \boldsymbol{y})$ and of the translational and rotational invariance that characterises an

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[8] The apparent inconsistency of retaining the *non*-linear convective derivative term ($\boldsymbol{v}$grad)$\boldsymbol{v}$ on the *LHS*, was subsequently addressed and resolved (Hyland and Rowlands 1970).

isotropic fluid; furthermore, the equations are independent of the statistics of the particles constituting the fluid—i.e. they hold equally for bosons and fermions. This clear separation between general and material-specific aspects mirrors that which obtains in classical hydrodynamics, where solution of the Navier-Stokes' equations requires knowledge of the pressure, which is outside the realm of hydrodynamics; the required information is contained in the fluid-specific equation of state, which must additionally be given.[9]

Fröhlich thus completely vindicated his belief not only that '.....surely it must be possible to achieve this [i.e. the derivation of these macroscopic equations] from the quantum-mechanical many-body problem without going into the complications exhibited in so many papers on this subject', but also that classical hydrodynamics indeed arises as 'a limiting case of a non-local theory' [F129].

Having been responsible for the introduction of quantum field theoretical techniques in condensed matter physics, Fröhlich was now becoming increasingly convinced of their rampant misuse, and lamented the neglect of general considerations evidenced in many of the *tour de force*, mathematically motivated, approaches to the many-body problem that started to appear in the late 1950s and early 1960s, which often contained features that evaded interpretation, making it difficult, if not impossible to assess their validity. Indeed, he repeatedly stressed that detailed solution of the *N*-body problem—even if it were possible—is, in principle, undesirable and pointless, since many macroscopic quantities (such as pressure, for example) have no meaning at a microscopic level; he went on to note, however:

> This does not, of course, make detailed treatments (using various approximation methods) superfluous, but puts them into their proper place, which is the calculation of magnitudes and other properties (*e.g.* temperature dependence) of various (material) parameters entering the formulation of macroscopic equations [F139].

Accordingly, he advocated the following two-stage strategy: (i) derive the *structure* of the relevant macroscopic equations from microphysics with the aid of quantities such as the reduced density matrices in terms of which *macroscopic* fields of interest can be defined, but whose exact equations of motion are governed by quantum mechanics, i.e. by microphysics; (ii) use more detailed many-body techniques to calculate the magnitudes and temperature dependences of quantities that arise as (formally defined) parameters in the derived macroscopic equations, such as the viscous coefficients in the case of the Navier-Stokes' equations. This philosophy, concerning the way in which the connection between micro and macrophysics should be approached, can be traced to his review of von Laue's

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[9] A similar distinction between generally valid differential equations that describe spatio-temporal evolution and equations pertaining to properties of particular materials is to be found in the domain of the macroscopic electrodynamics of ponderable media, where the Maxwell equations are the counterpart of the Navier-Stokes' equations, whilst the so-called constitutive relations have a status equivalent to that of the equation of state in hydrodynamics.

book, *Theorie der Supraleitung*, which appeared some 20 years earlier in *Nature* on 10th January 1948, wherein he wrote:

> In discussions on this subject it is often suggested that a more successful treatment of the many-body problem requires an improvement in mathematical technique rather than new physical ideas. Yet even an attempt to derive the properties of ideal gases would find the atomic physicist (if assumed to have no knowledge of macrophysics) at a loss without the introduction of new physical concepts. He would probably start with a discussion of the motion of two and then three weakly interacting particles, and afterwards be led to the conclusion that consideration of more particles is very complicated and unlikely to lead to any simple results. It is only after the introduction of new physical concepts which do not exist in atomic physics – such as pressure and entropy – that other simple laws of physics (the gas laws) can be found. It may well be that further concepts of this kind will be discovered and that they will be essential in solving problems like superconductivity.
> [*The full review is reproduced in Appendix 1 at the end of this chapter*]

One such 'further concept' turned out to be that of off-diagonal-long-range order (*ODLRO*); Yang noted that in the case of fermion systems, because of the Pauli principle, the lowest order reduced density matrix in which *ODLRO* can occur is $\Omega_2(\boldsymbol{x}', \mathbf{y}'; \boldsymbol{x}'', \mathbf{y}'')$, wherein it is reflected by the following factorization into 2-point macroscopic wave-functions $\Phi_2(\boldsymbol{x}, \boldsymbol{y})$ when the two pairs $(\boldsymbol{x}', \boldsymbol{y}')$ and $(\boldsymbol{x}''; \boldsymbol{y}'')$ [*the off-diagonal aspect*] are well-separated [*the long-range aspect*], which exhibits long-range phase correlations:

$$\Omega_2(\boldsymbol{x}', \mathbf{y}'; \boldsymbol{x}'', \mathbf{y}'') \rightarrow \Phi_2^*(\boldsymbol{x}'', \boldsymbol{y}'')\Phi_2(\boldsymbol{x}', \mathbf{y}'), \tag{6.2.17}$$

Expressing the complex field Φ_2 in terms of a real amplitude function (R) and real phase function (S) through $\Phi_2 = R\mathrm{e}^{\mathrm{i}S}$, the *RHS* of Eq. 6.2.17 takes the following form in which the long-range phase correlation (coherence) is evident:

$$\Phi_2^*(\boldsymbol{x}'', \boldsymbol{y}'')\Phi_2(\boldsymbol{x}', \mathbf{y}') = R(\boldsymbol{x}', \mathbf{y}')R(\boldsymbol{x}'', \boldsymbol{y}'')\mathrm{e}^{\mathrm{i}[S(\boldsymbol{x}', \boldsymbol{y}') - S(\boldsymbol{x}'', \boldsymbol{y}'')]} \tag{6.2.18}$$

In the case of a single particle, it is this relative phase that is responsible for typical quantum mechanical interference effects. In the case of a system comprising a *large* number of particles (a macroscopic system), on the other hand, due the thermal disorder, the associated phases usually average out, and thus have no influence on the macroscopic behaviour of the system, which thus behaves classically. In the presence of *ODLRO*, this does *not* happen, and the influence of the phase persists up to a macroscopic level, so that the system behaves as a *macroscopic* quantum system.

Non-asymptotically, Eq. 6.2.17, must be replaced by:

$$\Omega_2(\boldsymbol{x}', \mathbf{y}'; \boldsymbol{x}'', \mathbf{y}'') = \Phi_2^*(\boldsymbol{x}'', \boldsymbol{y}'')\Phi_2(\boldsymbol{x}', \mathbf{y}') + \Lambda_2(\boldsymbol{x}', \mathbf{y}'; \boldsymbol{x}'', \mathbf{y}'') \tag{6.2.19}$$

where $\Lambda_2(\boldsymbol{x}', \mathbf{y}'; \boldsymbol{x}'', \mathbf{y}'')$ vanishes unless all four coordinates are close together—i.e. does *not* exhibit *ODLRO*.

In general, a macroscopic wave-function, Φ, describes a system in which there is a macroscopic number, N_c, of identical particles (or groups of particles) all of which are in the *same* state, forming a *condensate*, so that each behaves in exactly the same way; the entire system is then effectively an N-fold replica of any one of them. Because of the macroscopic number of particles involved, the fluctuations in the number of particles in a given elemental sub-volume is so small that instead of being interpreted as a probability density (as in the case of microscopic systems) $|\Phi(\boldsymbol{x})|^2$ can here be re-interpreted as the actual condensate density, so that the following[10] normalisation holds:

$$\int |\Phi(\boldsymbol{x})|^2 d^3\boldsymbol{x} = N_\mathrm{c} \sim \mathrm{O}(N) \tag{6.2.20}$$

i.e. the wave-function Φ has both macroscopic normalisation and extent, and is thus called a 'macroscopic wave-function'. Being a field over the three-dimensional $\boldsymbol{x}$-space, occupied by the system, it is quite different from the N-particle ($3N$-dimensional configurational space) Schrödinger wave-function, $\Psi(\boldsymbol{q}_1, \boldsymbol{q}_2, \ldots, \boldsymbol{q}_\mathrm{i}, \ldots, \boldsymbol{q}_\mathrm{N})$, which is a function of all the N particles coordinates, $\boldsymbol{q}_\mathrm{i}$. and which, *unlike* $\Phi(\boldsymbol{x})$, is normalised to unity, in keeping with its probabilistic interpretation.

Returning to the case of superconductors, the macroscopic wave-function necessarily depends on *two* space points[11] $(\boldsymbol{x}, \boldsymbol{y})$, and is large only when $|\boldsymbol{x} - \boldsymbol{y}|$ is below a length characteristic of a particular material, and is thus normalised to N (Yang 1962):

$$\int |\Phi_2(\boldsymbol{x}, \boldsymbol{y})|^2 d^3\boldsymbol{x}\, d^3\boldsymbol{y} = \mathrm{O}(N) \tag{6.2.21}$$

It cannot be stressed too strongly that the above prediction of two-point macroscopic wave-functions $\Phi_2(\boldsymbol{x}, \boldsymbol{y}; t)$ in the case of a fermion system follows solely from the fact that Ω_2 is the lowest order reduced density matrix that can exhibit *ODLRO*, and is quite independent of any microscopic theory, although the *BCS* ground-state *happens* to be consistent with it. In terms of the latter, the condensate consists of pairs of electrons with equal and opposite momenta and spin, whilst the bounded spatial structure of $\Phi_2(\boldsymbol{x}, \boldsymbol{y})$ finds interpretation in terms of the macroscopic coherence length ($\sim 10^{-4}$ cm) that characterises these (Cooper) pairs (vide Sect. 5.4).

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[10] This is simply an N-fold 'magnification' of the normalisation $\int |\phi(x)|^2\, d^3\boldsymbol{x} = 1$, which holds in the case of a *single* particle described by a 'microscopic' wave-function, $\phi(\boldsymbol{x})$, for which $|\phi(\boldsymbol{x})|^2$ is a *probability* density.

[11] It should be noted that this was *not* the case in Fröhlich's 1966 paper [F123] in which he showed that the hydrodynamic equations of compressible fluids, together with the London equations, lead to the Ginzburg-Landau equation for the macroscopic wave-function in a superconductor; for here, the macroscopic wave-function depended on t and a *single* space coordinate only. Later in, 1970, in a short review published in *Nature* [F146], he commented that use of macroscopic wave-functions of this form—instead of the two-point form, $\Phi_2(\boldsymbol{x}, \boldsymbol{y}; t)$—might well be responsible for the difficulty in connecting the Ginzburg-Landau equation with microscopic theory, *other than* near the transition temperature.

Fröhlich [F139] first presented the integro-differential wave-equation satisfied by $\Phi_2(\boldsymbol{x}, \boldsymbol{y})$ at an international conference on statistical mechanics held in Kyoto in 1968 during his chairmanship of the Commission on Statistical Mechanics and Thermodynamics of the International Union of Pure and Applied Physics (*IUPAP*). Apart from replacing the non-local phonon-induced electron-electron attraction by a local, 2-body potential, $V(|\boldsymbol{x} - \boldsymbol{y}|)$, his derivation of the macroscopic wave-equation was exact; the equation takes the form:

$$\begin{aligned} i\hbar\ \partial_t\Phi_2(\boldsymbol{x},\mathbf{y}) = {} & \left[-\hbar^2/2m\left(\partial_x^2+\partial_y^2\right) + V(|\boldsymbol{x}-\boldsymbol{y}|)\right]\Phi_2(\boldsymbol{x},\mathbf{y}) \\ & + \int [V(|\boldsymbol{x}-\boldsymbol{z}|)+V(|\boldsymbol{y}-\boldsymbol{z}|)]\{\Omega_1(\mathbf{z},\boldsymbol{z})\Phi_2(\boldsymbol{x},\mathbf{y}) \\ & - {}^1\!/_2\,[\Omega_1(\boldsymbol{x},\boldsymbol{z})\Phi_2(\boldsymbol{z},\boldsymbol{y}) + \Omega_1(\boldsymbol{y},\boldsymbol{z})\Phi_2(\boldsymbol{x},\boldsymbol{z})]\}d^3\boldsymbol{z} \end{aligned} \tag{6.2.22}$$

Restricting consideration to the case of equilibrium, characterised by translational invariance and an absence of currents, A.W.B Taylor, a colleague in Fröhlich's department, and a former research student of his, showed that this macroscopic wave-equation Fourier transforms into one of the two basic expressions of the *BCS* theory in the formulation of De Gennes (Taylor 1970), namely:

$$2(\mu - \eta_{\mathrm{k}})\varphi_{\mathbf{k}} = \sum_{\mathbf{K}} V_{\mathbf{K}}\varphi_{\mathbf{k}-\mathbf{K}}\left(1 - f_{\mathbf{k}}^{+}\right) \tag{6.2.23}$$

where $\Phi_2(\boldsymbol{x}, \boldsymbol{y}) = \varphi(|\boldsymbol{x} - \boldsymbol{y}|) \exp(-2i\mu t/\hbar)$, and $\varphi(|\boldsymbol{x} - \boldsymbol{y}|) = \Sigma_{\mathbf{k}}\varphi_{\mathbf{k}} \exp i\boldsymbol{k} \cdot (\boldsymbol{x} - \boldsymbol{y})$; similarly, $V_{\mathbf{K}}$ and $f_{\mathbf{k}}$ are the Fourier coefficients of the potential $V(|\boldsymbol{x} - \boldsymbol{y}|)$ and $\Omega_1(|\boldsymbol{x} - \boldsymbol{y}|)$, respectively. In addition,

$$f_{\mathbf{k}}^{+} \equiv f_{\mathbf{k},\sigma=1} + f_{\mathbf{k},\sigma=-1} \tag{6.2.24}$$

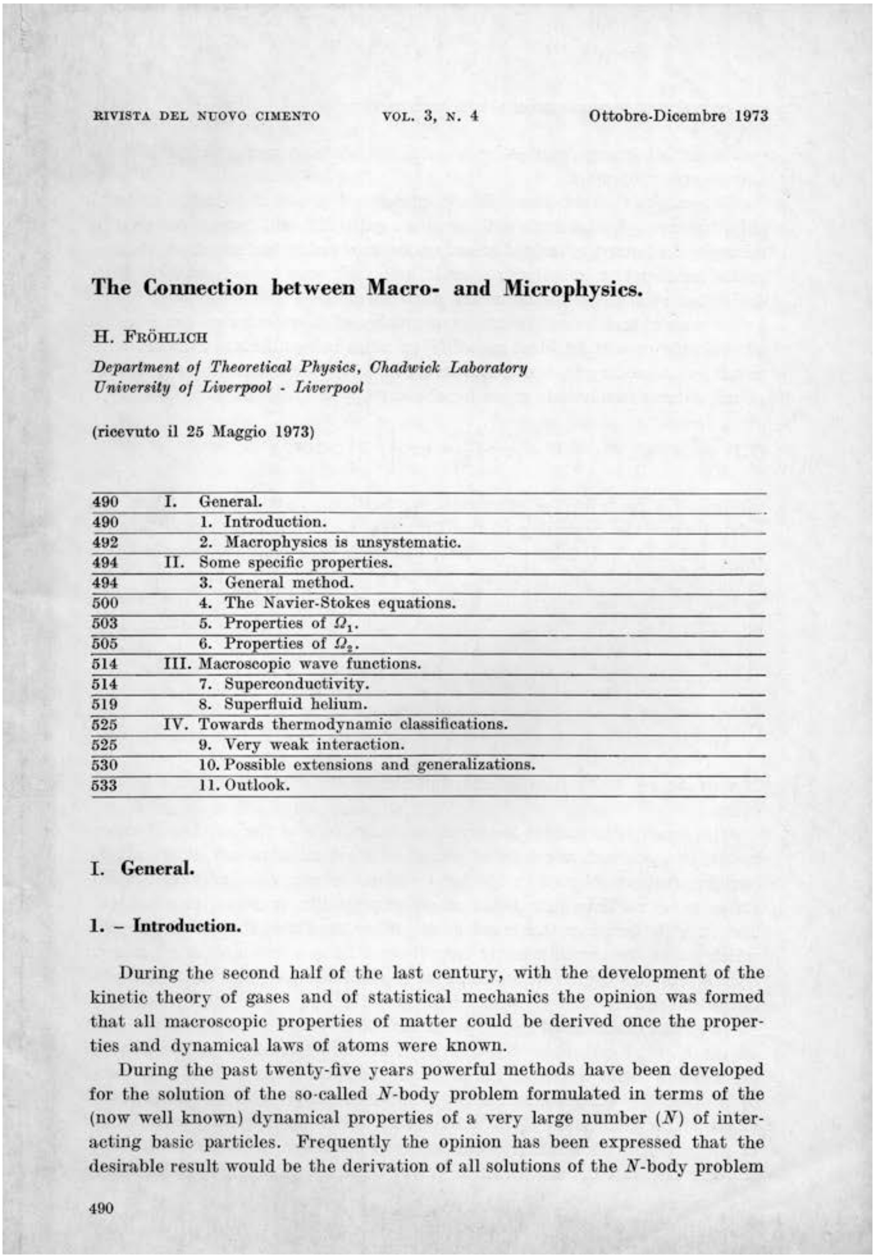
where σ is the spin coordinate, and η_{k} is a renormalised single electron energy, defined by:

$$\eta_{\mathrm{k}} \equiv \hbar^2 k^2/2m - {}^1\!/_2 \sum_{\mathbf{K}} V_{\mathbf{K}} f_{\mathbf{k}-\mathbf{K}}^{+} \tag{6.2.25}$$

The second basic expression in the *BCS* theory, which provides a connection between the Fourier components of Φ_2 and Ω_1, was later obtained by Fröhlich himself from consideration of the short-range properties of Ω_2, and for the ground-state takes the following form:

$$\frac{1}{2}\left(f_{\mathbf{k},\sigma} + \mathrm{f}_{-\mathbf{k},-\sigma}\right) - f_{\mathbf{k},\sigma}\mathrm{f}_{-\mathbf{k},-\sigma} = \varphi_{\mathbf{k}}\varphi_{\mathbf{k}}^{*} \tag{6.2.26}$$

This work was included in a long review of his ideas on the connection of micro and macrophysics, which was published in *Rivista del Nuovo Cimento* in 1973 [F155], just before his retirement from the Liverpool Chair (Fig. 6.3).



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The Connection between Macro- and Microphysics.

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I. General.

1. – Introduction.

During the second half of the last century, with the development of the kinetic theory of gases and of statistical mechanics the opinion was formed that all macroscopic properties of matter could be derived once the properties and dynamical laws of atoms were known.

During the past twenty-five years powerful methods have been developed for the solution of the so-called N-body problem formulated in terms of the (now well known) dynamical properties of a very large number (N) of interacting basic particles. Frequently the opinion has been expressed that the desirable result would be the derivation of all solutions of the N-body problem

Fig. 6.3 Fröhlich's 1973 review paper on the connection between macro- and microphysics [F155]—Reproduced with the kind permission of Società Italiana di Fisica

Other topics dealt with in this review article, apart from a detailed discussion of the boson superfluid He^4 (vide infra), included: (i) a particularly simple way of demonstrating the equivalence of the N-particle configuration space and second-quantized formalisms by using the hierarchy of equations of motion of the reduced density matrices, Ω_n; (ii) the use of reduced density matrices to obtain a new justification for statistical mechanics, which circumvents reliance on the ergodic theorem.[12] In connection with the latter, he noted that the hierarchy of equations of motion of the reduced density matrices, Ω_n, are *exact*, and do not contain any *statistical* assumptions, since they are defined *not* in terms von Neumann's density matrix, but rather the density matrix that describes the temporal evolution the whole *unique* system; statistical features arise only when several subsystems are considered. Nevertheless, if treated appropriately, he believed that this hierarchy should permit the introduction of thermodynamic quantities, and yield all relations that hold between them [F155]. He went on to show that this expectation is indeed fulfilled in the case of weak interaction and translational invariance, when the Fourier transform, f_p, of Ω_1*wrt* the non-local coordinate $(\boldsymbol{x}' - \boldsymbol{x}'')$ satisfies a Boltzmann-like equation, which although reversible, describes an irreversible approach to equilibrium. Defining equilibrium by the time-independence off_p, required that the collision term on the *RHS* of the equation vanish, which, in turn, required that f_p be a Bose or Fermi distribution.

More generally, f_p is a function of the associated centre-of-mass coordinate $\boldsymbol{x} \equiv ½(\boldsymbol{x}' + \boldsymbol{x}'')$, which in a gas would correspond to the so-called Wigner distribution. Unlike $f_\mathrm{p}(0), f_\mathrm{p}(\boldsymbol{x})$ can take *negative* values, concerning which Fröhlich commented:

> There is a large worried literature about this possibility, which many authors think contains contradictions, Actually, however, the simple-minded interpretation of $f_\mathrm{p}(\boldsymbol{x})$ as a 'number' of particles does not apply in such cases. The only quantity that must never be zero, the fluid density, $\sigma(\boldsymbol{x})$, never is negative ($\sigma(\boldsymbol{x}) = \Sigma_\mathbf{p} f_\mathbf{p}(\boldsymbol{x})/V^{½}$, where V is the volume of the system). Negative $f_\mathrm{p}(\boldsymbol{x})$ may and should arise when quantum mechanical interference becomes relevant [F155].

The exact equation of motion of $f_\mathrm{p}(\boldsymbol{x})$ follows directly from that (Eq. 6.2.10) of Ω_1:

$$\partial_\mathrm{t} f_\mathrm{p}(\boldsymbol{x}) + m^{-1}\boldsymbol{p}\ \partial_\mathrm{x} f_\mathrm{p}(\boldsymbol{x}) = (i/2\pi\hbar)\int W(\boldsymbol{x}',\boldsymbol{x}'')\mathrm{e}^{-\mathrm{i}\boldsymbol{p}\cdot\xi/\hbar}\mathrm{d}^3\boldsymbol{\xi} \tag{6.2.27}$$

where $W(\boldsymbol{x}',\boldsymbol{x}'') \equiv \int [V(|\boldsymbol{x}'-\boldsymbol{y}|) - V(|\boldsymbol{x}''-\boldsymbol{y}|)]\Omega_2(\boldsymbol{x}',\mathbf{y};\ \boldsymbol{x}'',\boldsymbol{y})d^3\boldsymbol{y}$, and $\boldsymbol{\xi} \equiv \boldsymbol{x}' - \boldsymbol{x}''$.

It will be observed that whilst the *LHS* of Eq. 6.2.27 has the form of the *LHS* of a Boltzmann equation, the *RHS* does not, and the conditions under which it takes the form of a Boltzmann-like *collision integral* remain to be established.[13]

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[12] He showed that already for $N = 20$, it would take a time longer than the age of the Universe for the system to pass through all of its microstates; accordingly, for $N \gg 1$, there is simply insufficient time for all microstates to be sampled equally often, as required by the ergodic theorem, which thus cannot be relevant to a justification of statistical mechanics.

[13] Fröhlich returned to this problem in 1971 in a series of 3 lectures given in Stuttgart. Derivation of the conditions under which a Boltzmann equation for the electron-phonon system can be derived *without* the use of the objectionable random phase approximation, was the subject of an earlier collaboration [F120] with A.W.B. Taylor, his former pupil.

The following year, he generalized his derivation of the basic expressions of the *BCS* theory (Eqs. 6.2.23 and 6.2.26) to the case of the actual non-local electron–electron interaction, *without* making any approximations [F157] that might violate gauge invariance.[14] The importance of this exact 'macroscopic' derivation of these equations, '....without reference to detailed models and obscure many-body techniques' [F142], cannot be over-emphasised, and vindicates his earlier conviction that this should not only be possible, but also desirable if the general validity of the *BCS* results is to be established.

> A need for this exists, in particular, in connection with the application of the Josephson formulae in the determination of the ratio *e/h* of fundamental constants. Clearly a formula used in this context must be known to be exact, and hence model independent [F155].

Subsequent work, along the lines initiated by Fröhlich, focussed on the electrodynamics of superconductors, in particular how the Meissner effect can be derived in an exact, gauge-invariant way. This was achieved by showing, from considerations of local gauge covariance,[15] that *ODLRO* cannot coexist with a uniform magnetic induction (provided the field is below a certain critical value that is proportional to the square-root of the free energy difference between the superconducting and normal states) (Sewell 1990). Sewell later showed by simple, general arguments that flux quantization[16] (in units of *ch/2e*) and persistent currents are consequences of the Meissner effect, thermodynamic stability, and the single-valuedness of the macroscopic wavefunction. It was found that the Meissner effect itself arises from a rigidity of the macroscopic wave-function in the presence of a magnetic field—a rigidity that is here the macroscopic counterpart of that originally proposed by London (1950, pp. 146–155) in the case of the many-electron Schrödinger wave-function in a superconductor (Sewell 1997, 2002).

In the case of the boson superfluid He^4, *ODLRO* appears already in the first reduced density matrix, as first noticed by Penrose (1951)—i.e. $\Omega_1(\boldsymbol{x}', \boldsymbol{x}'')$ factorises according to:

$$\Omega_1(\boldsymbol{x}',\boldsymbol{x}'') = \Phi_1^*(\boldsymbol{x}'')\Phi_1(\boldsymbol{x}') + \Lambda_1(\boldsymbol{x}',\boldsymbol{x}''), \tag{6.2.28}$$

where $\Phi_1(\boldsymbol{x})$ is the macroscopic wave-function (a complex field over space and time), and $\Lambda_1(\boldsymbol{x}', \boldsymbol{x}'') \rightarrow 0$ for large $|\boldsymbol{x}' - \boldsymbol{x}''|$. The real quantity $|\Phi_1(\boldsymbol{x})|^2$ is the condensate density, the value of which can be obtained experimentally from the

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[14] The *approximate* form of the electron-electron interaction term used in the *BCS* Hamiltonian *destroys* gauge invariance, making it impossible to define a unique current operator (Schafroth 1958).

[15] It is should be noted that the gauge principle here plays an *essential* role, in contrast to the situation with previous attempts to derive the Meissner effect, which are flawed precisely because they *violate* gauge invariance (Schafroth 1958)!

[16] Much later, Fröhlich made the important point that magnetic flux quantization is *not* restricted to superconductors, but is a completely general property of all materials—including biological ones, where, as he noted, the small size of cells should facilitate its observation [F(iv), Chap. 1]—vide Sect. 6.3.7

reversal in the trend of the X-ray or neutron diffraction peaks on cooling through T_λ, as first pointed out by (Cummings et al. 1970, 1981); for the observed peaks mirror the pair correlation function, $P(\boldsymbol{x}, \boldsymbol{y})$, which, together with Φ_1, is contained in the 2nd reduced density matrix, $\Omega_2(\boldsymbol{x}', \boldsymbol{y}'; \boldsymbol{x}'', \boldsymbol{y}'')$. The values of the condensate density deduced in this way are consistent with previous zero temperature, theoretical estimates of about 10 % of the total fluid density (Penrose and Onsager 1956; McMillan 1965). This reduction from the 100 % value that characterises an ideal Bose gas is due to the hard-core repulsion between the He^4 atoms, which depletes the ground-state by 90 %, consistent with the large zero-point energy of the system. This depletion is represented by the non-local field $\Lambda_1(\boldsymbol{x}', \boldsymbol{x}'')$ whose phase does not, in general, factorise, so that the associated velocity field can be transverse; by contrast, the condensate velocity field, $\boldsymbol{v}_c$, is necessarily longitudinal (i.e. $curl\boldsymbol{v}_c = 0$), being the gradient of the phase of Φ_1.

The following exact integro-differential macroscopic wave-equation satisfied by $\Phi_1(\boldsymbol{x}; t)$, in which Φ_1 is coupled to Λ_1 through terms involving the 2-body potential,[17] was first derived by Fröhlich in 1968 [F139], and used the following year to derive a proto-type two-fluid model based on the density and velocity fields associated with the condensate and depletion [F141][18]:

$$\begin{aligned} i\hbar\, \partial_t \Phi_1(\boldsymbol{x}) = -(\hbar^2/2m)\partial_{\boldsymbol{x}}^2 \Phi_1(\boldsymbol{x}) + \Phi_1(\boldsymbol{x}) \int V(|\boldsymbol{x}-\boldsymbol{y}|)\Omega_1(\boldsymbol{y},\boldsymbol{y}) d^3\boldsymbol{y} \\ + \int V(|\boldsymbol{x}-\boldsymbol{y}|)\Phi_1(\boldsymbol{y})\Lambda_1(\boldsymbol{x},\boldsymbol{y}) d^3\boldsymbol{y} \end{aligned} \tag{6.2.29}$$

It was subsequently noted, however, on the basis of a Monte Carlo calculation of Ω_1 (McMillan 1965), that Λ_1 does not vary slowly enough in comparison with V to validate certain expansions used by Fröhlich in the derivation of his two-fluid equations. This defect can be corrected by expanding instead about a *local* equilibrium in terms of the spatial gradients of the relevant *macroscopic* fields; in this way, a closed set of associated hydrodynamic equations was obtained for the condensate and depletion (Hyland and Rowlands 1971). The connection between this set of equations and those of the phenomenological two fluid model, formulated in terms of the superfluid and normal fluid, was later established by identifying the velocity field of the normal (non-superfluid) component with the velocity of entropy transport. This latter work yielded an expression for the superfluid density in terms of the condensate density, which had the correct limiting behaviour as $T \to 0$ and as

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[17] In the case of a hard-core potential, such as characterises that between He^4 atoms, $V(|\boldsymbol{x}-\boldsymbol{y}|)$ should be replaced by a screened potential $V(|\boldsymbol{x}-\boldsymbol{y}|)\,[1 - R(|\boldsymbol{x}-\boldsymbol{y}|)]$, where R is significant only when $|\boldsymbol{x}-\boldsymbol{y}|$ is within the dimensions of the core [F141]. The screening function originates from $\Omega_2(\boldsymbol{x}, \mathbf{y}; \boldsymbol{x}, \mathbf{y})$, and is necessary to ensure that $\Omega_2(\boldsymbol{x}, \mathbf{y}; \boldsymbol{x}, \mathbf{y}) \to 0$ for $|\boldsymbol{x}-\boldsymbol{y}| < 2a_o$, where a_o is the hard-core radius.

[18] Most important in this paper is a 'Note added in Proof', which points out that for longitudinal flow the *whole* density flows with the velocity of the condensate, whereas for transverse flow the energy density of flux is *higher* as long as the condensate density is non-zero, consistent with the existence of a gap for transverse motion mentioned above.

$T \rightarrow T_\lambda$ (Hyland and Rowlands 1972); it is to be noted that, unlike the condensate, the superfluid density[19] approaches ideal Bose gas value of 100 % as $T \rightarrow 0$. Around the same time, another attempt to connect the 2 two-fluid models was made by Haug and Weiss, using a somewhat different methodology (Haug and Weiss 1972).

The problem was attacked in a more fundamental way by Fröhlich the following year (1973). Starting with a detailed analysis of the properties of Ω_2, whose *short-range* behaviour was found to dominate the behaviour of Ω_1 (even though it makes only a minute contribution the normalisation condition Eq. 6.2.5), he showed that the domination of the majority of the system (based on Λ_1) by the minority Φ_1-condensate arises from a *partial* factorisation of $\Omega_2(\boldsymbol{x}', \mathbf{y}'; \boldsymbol{x}'', \mathbf{y}'')$ into terms of the form $\Phi_2^*(\boldsymbol{x}'', \mathbf{y}'')\Phi_2(\boldsymbol{x}', \mathbf{y}')$, according to:

$$\Omega_2(\boldsymbol{x}',\boldsymbol{y}';\boldsymbol{x}'',\boldsymbol{y}'') = \Phi_2^*(\boldsymbol{x}'',\boldsymbol{y}'')\Phi_2(\boldsymbol{x}',\boldsymbol{y}') + \Lambda_2(\boldsymbol{x}',\boldsymbol{y}';\boldsymbol{x}'',\boldsymbol{y}''), \tag{6.2.30}$$

but where, in *contrast* to the case of fermion superconductors, Φ_2 does *not* have the form of a bound-state, so that the spatial integral of $|\Phi_2|^2$ is here of order N^2, as opposed to N [F155]. When $\boldsymbol{x}'$ is far from $\boldsymbol{y}'$, $\Phi_2(\boldsymbol{x}', \boldsymbol{y}')$ must be proportional to $\Phi_1(\boldsymbol{x}')\ \Phi_1(\boldsymbol{y}')$, to ensure that $\Omega_1(\boldsymbol{x}', \boldsymbol{x}'')$ exhibits *ODLRO*—i.e. has the form given by Eq. 6.2.28. Thus in general, $\Phi_2(\boldsymbol{x}, \boldsymbol{y})$ must have the following form:

$$\Phi_2(\boldsymbol{x},\boldsymbol{y}) \sim \Phi_1(\boldsymbol{x})\Phi_1(\boldsymbol{y}) + g(\boldsymbol{x},\boldsymbol{y}) \tag{6.2.31}$$

where $g(\boldsymbol{x}, \mathbf{y})$ is a short-range function whose normalization integral, like that of Φ_2 in the case of superconductors, is of the order of N, *not* N^2—i.e.

$$\int g^*(\boldsymbol{x},\mathbf{y})g(\boldsymbol{x},\mathbf{y})d^3\boldsymbol{x}d^3\boldsymbol{y} = \mathrm{O}(N) \tag{6.2.32}$$

The domination[20] of the majority of the system, as represented by the incoherent component Λ_1 (Eq. 6.2.28), by the coherent minority represented by Φ_1 is thus seen to arise in an *indirect* way via the short-range field $g(\boldsymbol{x}, \mathbf{y})$, and can be expressed in a form similar to Eq. 6.2.26 that holds in the case of superconductors, except that in the case of a boson superfluid (He^4), $g_\mathbf{k}g^*_\mathbf{k}$ is found to constitute a lower limit on the fluctuation $<n_\mathrm{k}^2> - f_\mathrm{k}^2$, where $<n_\mathbf{k}> = f_\mathbf{k}$, the Fourier component of Ω_1 in equilibrium—i.e. $\Sigma_\mathbf{k} <n_\mathbf{k}> = N$, the total mean number of particles.

Later work showed that $g(\boldsymbol{x}, \mathbf{y}) \neq 0$ entails an an additional term, $\int V(|\boldsymbol{x}-\boldsymbol{y}|)\Phi_1^*(\boldsymbol{y})g(\boldsymbol{x},\boldsymbol{y})d^3\boldsymbol{y}$, on the *RHS* of Eq. 6.2.29, via which Φ_1 is coupled to $\Phi_1{}^*$; it was found that this term is essential if the normal state ($\Phi_1 \equiv 0$) is to be able to

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[19] The first attempt to understand how the minority (10 %) condensate 'pilots' the much larger depletion in a superfluid manner was made by Cummings (1971).

[20] This domination is even more marked in the case of superconductors where, in terms of the density of states at the Fermi energy, $D(\mathrm{E_F})$ and the energy gap Δ, $|\Phi_2|^2 \approx D(E_F)\Delta$, so that $|\Phi_2|^2/\Omega_1(x,x) \sim 10^{-5} - 10^{-4}$, even at $T = 0$; comparison with the case of He^4 is not straightforward, however, because of the operation of the Pauli principle in one case but not the other.

develop an instability leading to the establishment of a state with a non-zero Φ_1, in keeping with the λ-transition exhibited by liquid He^4 (Hyland and Rowlands 1974).

The problem of superfluidity thus turned out (perhaps unexpectedly) to be more subtle than that of superconductivity, essentially because of the strong short-range, hard-core interaction between He^4 atoms.

6.3 From Theoretical Physics to Biology

6.3.1 Historical Preliminaries

Around 1965, Fröhlich and his wife were in Alpbach (Austrian Tyrol), where she was attending a conference on science and life, while he indulged his love of mountain–climbing.[21] There, quite by chance, she met Maurice Marois (1922–2004), a professor of Medicine at the Sorbonne and founder in 1960, together with François de Clermont-Tonnerre,[22] of *l'Institut de la Vie* in Paris. They duly embarked on a discussion of the relation of physics to life, she casually telling Marois that her husband was a famous theoretical physicist to whom he was later introduced. Keen to pursue the contact, Marois suggested that they should meet in Paris, where, during a lunch, Fröhlich's wife happened to mention that, according to Wigner, 'life' was impossible from the point of view of quantum mechanics (Wigner 1961).[23] At this, Marois became excited, and asked Fröhlich what could be done to bridge the gap between physics and biology. At the time, he was rather reluctant to get involved, since not only had he never really been interested in this question, but also because he was then immersed in pure theoretical physics from which he did not wish to be deflected. Marois, however, persisted and eventually Fröhlich agreed to help organise what was to be the first of many successful international conferences entitled 'Theoretical Physics and Biology', which were to be held, under the auspices of *l'Institut de la Vie*, at Versailles, and elsewhere. These conferences, which continued, biennially, until 1988, were attended by highly eminent physicists and biologists, including such people as Onsager, Prigogine, Crick, Edelman, Cooper and Wigner himself.

That Fröhlich had agreed to get involved was due to the fact that the encounter with Marois had reminded him of something he had realised about living systems already before the Second World War, following a conversation he had had, in Bristol around 1938, with an endocrinologist friend of his, Max Reiss (a refugee

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[21] This he did *without* the use of ropes or crampons.

[22] François de Clermont-Tonnerre (1906–79) was Deputy Commander of the Allied Landings in N. Africa, and then Chief-of-Staff of General Giraud. He took part in the Tunisian Campaign, undertook aerial missions over Berlin, and after the war was made a Chevalier of the Legion of Honour.

[23] This was later the subject of a chapter in 'Symmetries and Reflections', published in 1967 (Wigner 1967). For a more recent assessment, see Swain (2002).

from Prague, and a founder of psychiatric endocrinology), who had told him that a small electrical potential difference of about 100 mV is maintained across the membrane of a *living* cell. Upon ascertaining the thickness of the membrane layer (of the order of 10^{-6} cm), Fröhlich immediately realised that this implied an enormous electric field of the order of 10^5 V/cm. At that time, it will be remembered, Fröhlich was working on dielectric breakdown (vide Sect. 4.1), and he immediately appreciated that in such a high field dielectric breakdown would undoubtedly occur in non-biological materials, unless special precautions were taken. He accordingly concluded that such a remarkable dielectric property must surely be exploited by biological systems in the execution of some vital function. He did not stop at this, however, but went on to consider associated dynamical properties. Assuming an elastic constant corresponding to a sound speed of 10^5 cm/s, he estimated that the frequency at which the inner and outer surfaces of the cell membrane vibrate against one another is about 50 GHz. At this time, such frequencies (in the millimetre microwave region) were technologically unavailable. 'How typical', Fröhlich exclaimed, 'for Nature to take advantage of our experimental incapabilities!' Encouraged however, by his contact at the *ERA*, Willis Jackson (later Lord Jackson of Burnley), who told him that such frequencies would probably be available before too long (Jackson was no doubt aware of the contemporaneous development of *RADAR*, then employing centimetre waves), Fröhlich approached the biologist Victor Rothschild (3rd Baron Rothschild 1910–1990) for suggestions of appropriate biological systems to investigate once experimental facilities became available. Rothschild suggested blood cells—a highly perceptive suggestion, given later developments (vide infra); but it was now 1939, and no further progress was made.

From his experience with superconductivity during the intervening years, Fröhlich had learned that one idea does not make a theory,[24] and thus suspected that in addition to the remarkable dielectric properties of living cells with which he was already aware, another idea had to be introduced: this he took to be *coherence*. Consistent with his general alertness to the possibility that a given concept might well have a relevance to fields other than that in which it had first arisen, and refusing to believe that 'one cannot learn anything from superconductivity other than a theory of superconductivity' [F165], he started to consider the possibility that coherence might *not* be confined to systems near thermal equilibrium—as is the case with superfluids and superconductors, wherein it underpins, amongst other things, the dynamic order that is characteristic of flow in these systems—but can perhaps be realised also in living biological systems at room temperature. In contrast to superfluids and superconductors, however, living systems are open, dissipative systems that are held *far* from thermal equilibrium by their metabolic activity, so that coherence might here be reasonably expected to be reflected as a new kind of *dynamic order* allied to biological activity; perhaps the closest

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[24] Here, his initial identification of the phonon-induced electron-electron interaction had to be supplemented by a second idea, namely, *pairing*, which, following the work of Yang, collectively expresses itself macroscopically as long-range phase correlations, or *coherence*.

comparable *non*-biological system in which coherence is realised at room temperature, as a consequence of non-linear interactions, is the pumped laser.

In the case of the very much more structurally complex systems encountered in biology, the question naturally arises as to how coherence might be established. Clearly, an atomic or electronic approach, such as was possible in the case of superfluids and superconductors, is here not feasible, in consequence of the much greater structural complexity of biological systems. But, as Fröhlich noted, this complexity is essentially the same, whether or not the system is alive; what distinguishes a living system is its orderly functioning. This functioning, he maintained, must be reflected in the strong excitation of relatively few degrees of freedom that dominate the remainder when the system is alive, and which constitute a physical discriminant of ‘aliveness’. Since such orderly functioning is a holistic property, it is in turn reasonable to suppose that these few dominating degrees of freedom are in the nature of *collective* modes that are lifted far from the thermal equilibrium to which the majority of modes continue to subscribe.

It was in connection with attempting to identify relevant collective modes that the remarkable dielectric properties of living biological systems suggested themselves to Fröhlich. By the late 1960s, he was aware that, in addition to the vibrations of the inner and outer surface of the cell membrane against one another, at around 50 GHz, which he had considered originally in the late 1930s, there were ones associated with membrane proteins that are strongly polarised by the high trans-membrane electric field of a metabolically active cell; another possibility was the stretching vibration of a hydrogen bond (Careri 1969). Subsequent work indicated other kinds of dipolar vibrations characterised by both lower and higher frequencies, such as those calculated for a number of giant breathing and rocking modes in double helix structures, with frequencies around 10^9 Hz (Prohofsky and Eyster 1974; Prohofsky 1987), those in the vicinity of 1×10^{12} Hz estimated for certain vibrations of blue proteins (Brill 1978), and those in the near 5×10^{13} Hz, reported for *H*-bonded amide structures (Careri 1973).

The presence of electric dipoles of various kinds thus started to emerge as ubiquitous feature of biological systems, and in consequence of the long range of the Coulomb field of an electric dipole, Fröhlich realised that there must indeed exist bands of collective longitudinal electric modes associated with the totality of dipoles of a given kind, each characterised by the same frequency.

He first presented his ingenious ideas in the opening paper *Quantum Mechanical Concepts in Biology* [F138] at an international conference, held at the Trianon Palace in Versailles in 1967 under the auspices of *l’Institut de la Vie*. This paper is an example of cross-fertilization *par excellence,* and of his approach in general—namely, one in which a calculation is always preceded by an idea. For, apart from its Appendices, the paper contains absolutely no mathematics; instead, it is concerned with enunciating, in a physically clear way, his novel ideas, and with consideration of their ramifications, such as, for example, the possible role of his conjectured biological coherence in the control of cell division. In this connection, he noted another physical characterising feature of biological systems, namely their elastic deformability, whereby the system can stabilize itself (lower its energy) by exploiting the fact that the polarization energy depends on the shape of the confining system (vide Sect. 5.2). The coherent excitation of a polarization wave will be accompanied by an associated coherent deformation that exerts a stress of the surface of a cell, which increases with its size. He conjectured that this stress might act as a stimulus for cell division, provided the density of cells is sufficiently low that there is no contact inhibition so that the elastic deformation necessary for stabilization can occur. Fröhlich concluded his paper with the following statement:

> I wish to remark that the above suggestions are meant to be highly speculative. They should demonstrate, however, that application of quantum mechanical concepts can lead to new points of view which might be used as guiding points in the search for undiscovered regularities [F138].

6.3.2 A Simple Non-linear Model for a Dynamic Coherent Excitation

For the next 25 years, Fröhlich tirelessly led this search. Already by 1968—even before the proceedings of the Versailles conference had been published (which was not until 1969)—he had succeeded in showing [F133, F134] how such dynamical order could be established through a kind of non-equilibrium phase transition. He did this using a simple model of an active biosystem comprising: (i) a large number, Z, of identical units, each unit being capable of electric dipole oscillations at the same frequency ω_o; the long range Coulomb interaction between the individual oscillators then gives rise to a band of longitudinal electric polarization modes of the material as a whole, covering a frequency range $\omega_1 \leq \omega \leq \omega_2$, which may be shifted considerably from ω_o, but which is relatively narrow: (ii) each unit is assumed to be fed locally with metabolic energy at a constant rate, so that each of the Z modes receives energy at the same rate, s: (iii) it is assumed that each unit can exchange energy locally with the surrounding medium, which is treated as a heat bath[25] that is in thermal equilibrium at a fixed temperature, the energy exchange being in quanta $\hbar\omega$, and at a rate that is nearly independent of ω.

The net rate of energy loss, L_{1r}, of the mode with frequency ω_r containing n_r quanta is given by:

$$L_{1r} = \phi[n_r \exp(\hbar\omega_r/kT) - (1 + n_r)] \tag{6.3.1}$$

where the function ϕ may depend on temperature.

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[25] The heat bath comprises, amongst other things, cell water (both bound and free), mobile ions, certain electronic degrees of freedom and elastic displacements.

The existence of a *range* of collective frequencies, $\omega_1 \leq \omega \leq \omega_2$, permits *non-linear* exchanges with the heat bath involving the absorption of a quantum $\hbar\omega_\mathrm{r}$ in conjunction with the emission of a quantum $\hbar\omega_\mathrm{q}$ (or vice versa) in the range

$$0 < \hbar|\omega_\mathrm{r} - \omega_\mathrm{q}| \leq \hbar(\omega_2 - \omega_1) \tag{6.3.2}$$

The net rate of energy loss, $L_{2\mathrm{r}}$, of the mode ω_r due to such processes is taken to be of the form:

$$L_{2\mathrm{r}} = \chi \sum_\mathrm{q} \left[n_\mathrm{r}(1 + n_\mathrm{q}) \exp \hbar(\omega_\mathrm{r} - \omega_\mathrm{q})/kT - n_\mathrm{q}(1 + n_\mathrm{r}) \right] \tag{6.3.3}$$

where the function χ may depend on temperature.

The forms of Eqs. 6.3.1 and 6.3.3 are dictated by the requirement that in thermal equilibrium—i.e. in the absence of energy supply, $s = 0 - n_\mathrm{r}$ is given by the usual Planck distribution

$$n_\mathrm{r}^\mathrm{o} = \frac{1}{\exp(\hbar\omega_\mathrm{r}/kT) - 1} \tag{6.3.4}$$

The condition for stationarity

$$s = L_{1\mathrm{r}} + L_{2\mathrm{r}}, \tag{6.3.5}$$

entails the following expression for n_r

$$n_\mathrm{r} = [1 + s/(\phi + \chi N)] \frac{1}{\exp(\hbar\omega_\mathrm{r} - \mu)/kT - 1} \tag{6.3.6}$$

where μ is defined by:

$$\exp(-\mu)/kT \equiv \frac{\phi + \chi \sum_\mathrm{q} \left[(1 + n_\mathrm{q}) \exp(-\hbar\omega_\mathrm{q}/kT) \right]}{(\phi + \chi N)} \tag{6.3.7}$$

where N is the total number of polarization quanta—i.e. $N = \Sigma_\mathrm{q} n_\mathrm{q}$.

μ can formally be interpreted as a *chemical potential*, and here arises as a consequence of the competition between the dissipation inherent in the system and the metabolic pumping, which fixes the *total* number of polarization quanta; it should be noted that $\mu = 0$ in the absence of the non-linear, two-quanta process, $L_{2\mathrm{r}}$—i.e. for $\chi = 0$. A non-zero chemical potential entails, of course, the possibility of a redistribution of polarization quanta amongst the modes, analogous to the Einstein condensation that occurs in a Bose gas of a fixed number of material particles in thermal equilibrium as the temperature is lowered. This can be seen from Eq. 6.3.6, which shows that to ensure $n_\mathrm{r} > 0$, requires

$$\hbar\omega_1 > \mu > 0 \tag{6.3.8}$$

where ω_1 is the lowest frequency in the band of collective polarization modes. Thus a kind of Bose condensation into the *lowest* frequency mode occurs when μ approaches $\hbar\omega_1$ very closely, which it does when s exceeds a critical value, s_o; thus in contrast to the equilibrium condensation in an ideal Bose gas, which is achieved by reducing the energy of the system by lowering the temperature, the condensation in the case of a metabolically active biosystem actually requires a supercritical rate of energy *supply* (Fig. 6.4).

Thus, below the critical rate, s_o, the heat-bath succeeds in imposing its equilibrium Planck distribution on the polar modes. At $s > s_o$, however, this is no longer possible, and the energy in excess of that which can be accommodated in thermal equilibrium is channelled (via the two-quanta processes in Eq. 6.3.3) into the polarization mode of lowest frequency, ω_1, wherein it is stored. After a sufficient time, this single mode, which extends throughout the whole system, becomes strongly excited mechanically (i.e. has zero entropy[26]) to an amplitude far in excess[27] of that which would obtain in thermal equilibrium; it thereby achieves a macroscopic significance as a 'coherent excitation' in which the constituent dipolar units oscillate in phase with one another.[28] This coherent excitation dominates the dynamical behaviour of the system whose other modes remain only incoherently excited thermally, as they are in the absence of metabolic activity. It is interesting, historically, to recall that the purely mechanical nature of this energy storage as a discriminant of a living system had been recognised by Schrödinger already in 1943 (and somewhat later also by F. London) in his influential book 'What is Life?', based on a series of Lectures given at the Dublin Institute for Advanced Studies, wherein can be found the following statement:

> The living organism seems to be a macroscopic system which in part of its behaviour approaches that purely mechanical (as opposed to thermodynamical) conduct [behaviour] to which all systems tend as the temperature approaches the absolute zero and all molecular disorder is removed (Schrödinger 1944).[29]

Of particular importance is the polarization mode of zero wave-vector, corresponding to a 'giant' dipole oscillation of the whole system, in which the positive

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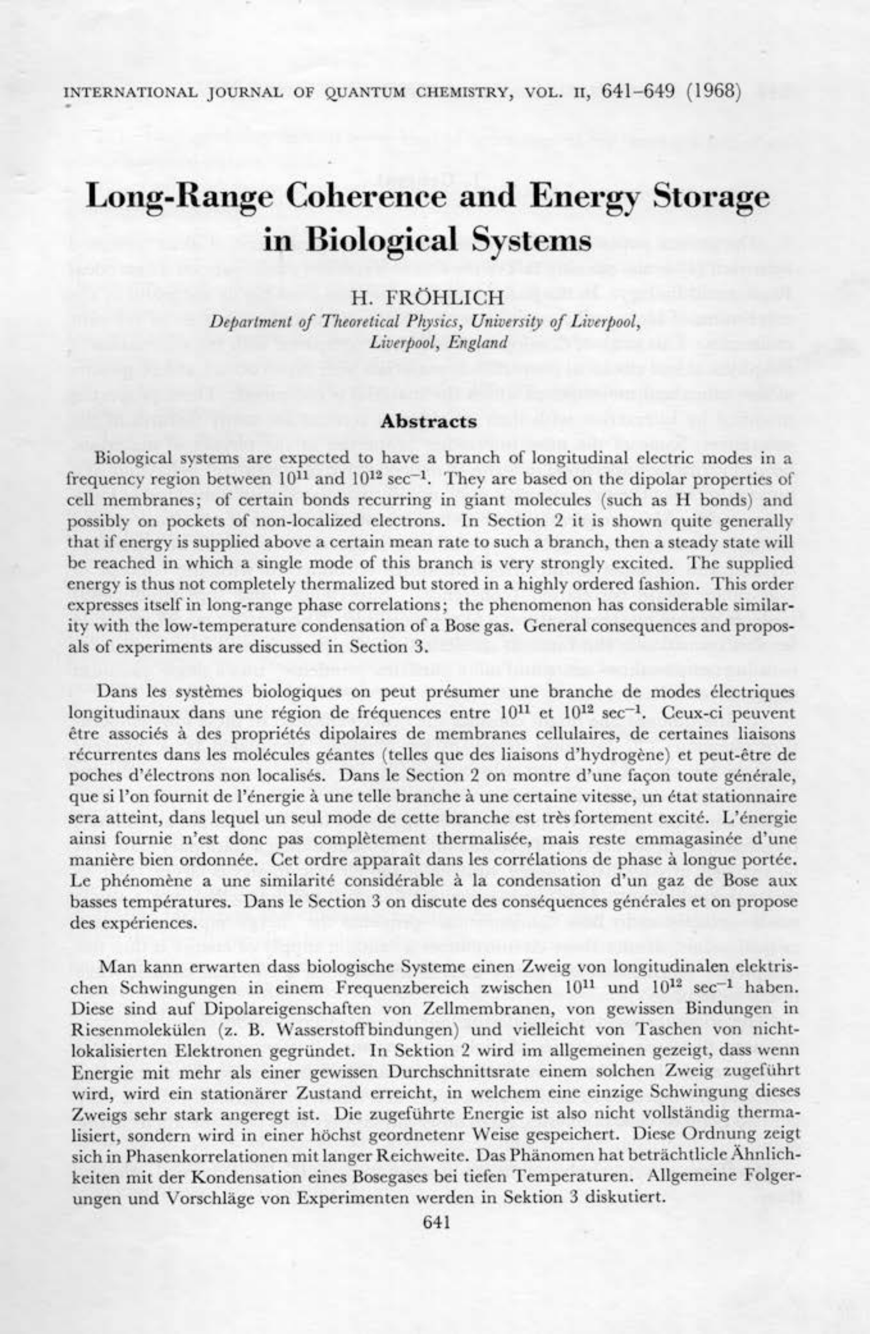
[26] It is important to realise that this does *not* entail any violation of the Second Law of Thermodynamics since there is an associated *increase* in the entropy of the remainder of the system (the heat bath).

[27] Field–theoretically, this is equivalent to the occupation number of this mode (the condensate) becoming macroscopically large.

[28] This is almost the *inverse* of the situation that obtains in the case of dielectric breakdown, where absorption of energy from the applied electric field by conduction electrons *raises* their collective temperature *above* that of the lattice—vide Sect. 4.7.

[29] It is essential to appreciate, however, the fundamental difference between the mechanical behaviour here referred to, which is an equilibrium feature realised at absolute zero, and that predicted by Fröhlich for living systems at ambient temperatures, which is an extreme ***non***-equilibrium effect.



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Long-Range Coherence and Energy Storage in Biological Systems

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Abstracts

Biological systems are expected to have a branch of longitudinal electric modes in a frequency region between 10^{11} and 10^{12} sec^{-1}. They are based on the dipolar properties of cell membranes; of certain bonds recurring in giant molecules (such as H bonds) and possibly on pockets of non-localized electrons. In Section 2 it is shown quite generally that if energy is supplied above a certain mean rate to such a branch, then a steady state will be reached in which a single mode of this branch is very strongly excited. The supplied energy is thus not completely thermalized but stored in a highly ordered fashion. This order expresses itself in long-range phase correlations; the phenomenon has considerable similarity with the low-temperature condensation of a Bose gas. General consequences and proposals of experiments are discussed in Section 3.

Dans les systèmes biologiques on peut présumer une branche de modes électriques longitudinaux dans une région de fréquences entre 10^{11} et 10^{12} sec^{-1}. Ceux-ci peuvent être associés à des propriétés dipolaires de membranes cellulaires, de certaines liaisons récurrentes dans les molécules géantes (telles que des liaisons d'hydrogène) et peut-être de poches d'électrons non localisés. Dans le Section 2 on montre d'une façon toute générale, que si l'on fournit de l'énergie à une telle branche à une certaine vitesse, un état stationnaire sera atteint, dans lequel un seul mode de cette branche est très fortement excité. L'énergie ainsi fournie n'est donc pas complètement thermalisée, mais reste emmagasinée d'une manière bien ordonnée. Cet ordre apparaît dans les corrélations de phase à longue portée. Le phénomène a une similarité considérable à la condensation d'un gaz de Bose aux basses températures. Dans le Section 3 on discute des conséquences générales et on propose des expériences.

Man kann erwarten dass biologische Systeme einen Zweig von longitudinalen elektrischen Schwingungen in einem Frequenzbereich zwischen 10^{11} und 10^{12} sec^{-1} haben. Diese sind auf Dipolareigenschaften von Zellmembranen, von gewissen Bindungen in Riesenmolekülen (z. B. Wasserstoffbindungen) und vielleicht von Taschen von nichtlokalisierten Elektronen gegründet. In Sektion 2 wird im allgemeinen gezeigt, dass wenn Energie mit mehr als einer gewissen Durchschnittsrate einem solchen Zweig zugeführt wird, wird ein stationärer Zustand erreicht, in welchem eine einzige Schwingung dieses Zweigs sehr stark angeregt ist. Die zugeführte Energie ist also nicht vollständig thermalisiert, sondern wird in einer höchst geordnetenr Weise gespeichert. Diese Ordnung zeigt sich in Phasenkorrelationen mit langer Reichweite. Das Phänomen hat beträchtlicle Ähnlichkeiten mit der Kondensation eines Bosegases bei tiefen Temperaturen. Allgemeine Folgerungen und Vorschläge von Experimenten werden in Sektion 3 diskutiert.

Fig. 6.4 Fröhlich's 1968 paper [F134] in the International Journal of Quantum Chemistry, which in 1988 became one of the most frequently cited works in its field—Reproduced with the permission of John Wiley & Sons charge of each constituent dipolar unit moves in synchrony in the same direction by the same amount, and the negative charges similarly, but in the opposite direction, so that the entire dipolar system macroscopically replicates the motion of a single dipole; for an external electromagnetic radiation can interact with this mode[30] provided it has the same frequency, and provided its vacuum wavelength is much greater than the size of the oscillating system.[31] This condition is fulfilled in the case of the basic units involved in *photosynthesis*, and later the same year (1968), Fröhlich applied these ideas to this problem [F137]. Here absorption of *light* replaces the metabolic energy supply considered originally, causing excitation of the *transverse* branch of excitonic modes. Again invoking non-linear interaction of these modes with their surroundings, transfer of the incoming light energy into the lowest frequency mode occurs, wherein it is stored, resulting in its strong excitation; in addition, attention was drawn to the fact that the light absorption should show two peaks, one associated with bulk properties, the other, with the surface. It must be emphasised, however, that the coherent excitation does not necessarily take the form of a giant dipole oscillation; rather, such simply represents one particularly striking possibility.

Provided frictional losses[32] are sufficiently small, the coherent mode can be stabilised through its non-linear coupling with the elastic field of the material; in this way is realised a state of *dynamical order* that in thermal equilibrium would be extremely improbable and highly transient, but which is here an *emergent* property associated with the self-organising ability of an open, dissipative system when *far* from thermal equilibrium. He noted that in consequence of this stabilization, the frequency of the lowest lying longitudinal electric mode *when coherently excited* must, in general, be anticipated to be *lower* than it is in the case of incoherent excitation, i.e. the mode is 'softened' to some extent, whilst the value of the critical pumping rate will itself be lower than it would otherwise be. Since the elastic deformability depends, in general, on the *state* of biological evolution—older (larger) cells being more easily deformed—the quantitative characteristics of a particular dynamic coherent excitation must be expected to be similarly dependent.

He stressed that maintenance of the associated dynamical order is contingent on the *continued* inflow of energy at the appropriate rate; indeed, it is quite possible that a particular coherent excitation is realised only as and when it is required for

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[30] As explained in Sect. 5.2, there are actually three such modes with frequencies intermediate between those of the usual longitudinal and transverse bulk modes, and which (because of the long range of the Coulomb interaction) depend on the macroscopic size and shape of the system (F(ii), Engelman and Ruppin 1968).

[31] For further details, see Hyland (2002).

[32] The effect of frictional losses (such as viscous damping), which contribute to the total dissipation in the system, is to *increase* the threshold power, s_o, above the value it would otherwise have; provided the metabolically available power *exceeds* this higher value, the coherent excitation will still be realised. An interesting and important case of a coherent oscillation on which viscous damping is minimal is the metabolically maintained longitudinal oscillation of microtubules in the brain; for here, the interplay between material properties *peculiar to these structures* results in 'slip' boundary conditions (Pokorný et al. 1997).

the execution of some essential biological function—i.e. that it exists only for a certain period of time at some specific stage in the evolution of the bio-system when, for example, its elastic deformability is such as to permit stabilisation of the coherent mode appropriate to its degree of excitation.

These matters were the subject of a fascinating series of radio broadcasts (in German) in 1970, which were subsequently published [F143].

6.3.3 A New Long-Range Frequency-Selective Attraction

In 1972, before he had discovered the existence of experimental support for these ideas, Fröhlich showed [F150] that two large, electrically neutral systems (such as cells or giant molecules) can attract one another, provided they are properly 'tuned'—i.e. oscillate coherently with the same frequency. Assuming the vibrations to be predominantly polar, he showed that the new attractive force varies as R^{-3}, where R is the separation of the two systems; it is thus of *longer* range than the usual van der Waals' attraction (which is proportional to R^{-6}). The quantitative *range* of the force was found to be governed by the dielectric properties of the intervening medium at the frequency of coherent mode (Fig. 6.5). In this way, the possibility exists of a frequency-specific attraction on both an inter- *and* intra-cellular level, thereby providing a novel basis of inter-cellar communication and for understanding of the specificity of the attraction between enzymes and their substrates, as well as the pairing of homologous chromosomes in meiosis, in terms of 'frequency recognition', as first pointed out by B.W. Holland,[33] a former *ICI* Research Fellow in Fröhlich's department in Liverpool during the 1960s (Holland 1972).

Being reliant on the existence of coherent excitations, the attractive interaction can be switched on and off according as to whether or not the rate of metabolic energy supply is sufficient to excite the necessary coherent vibration. In the case of an inadequate level of metabolic activity ($s < s_o$), the attraction could be 'switched-on' by exposure to external microwave radiation[34] [F176], provided the radiation can supply the deficit power ($s_o - s$) for a sufficient length of time, which is minimal at resonance—i.e. when the frequency of the external field is the same as that of the coherent excitation (Bhaumik et al. 1976). In this way, a dispersive repulsive

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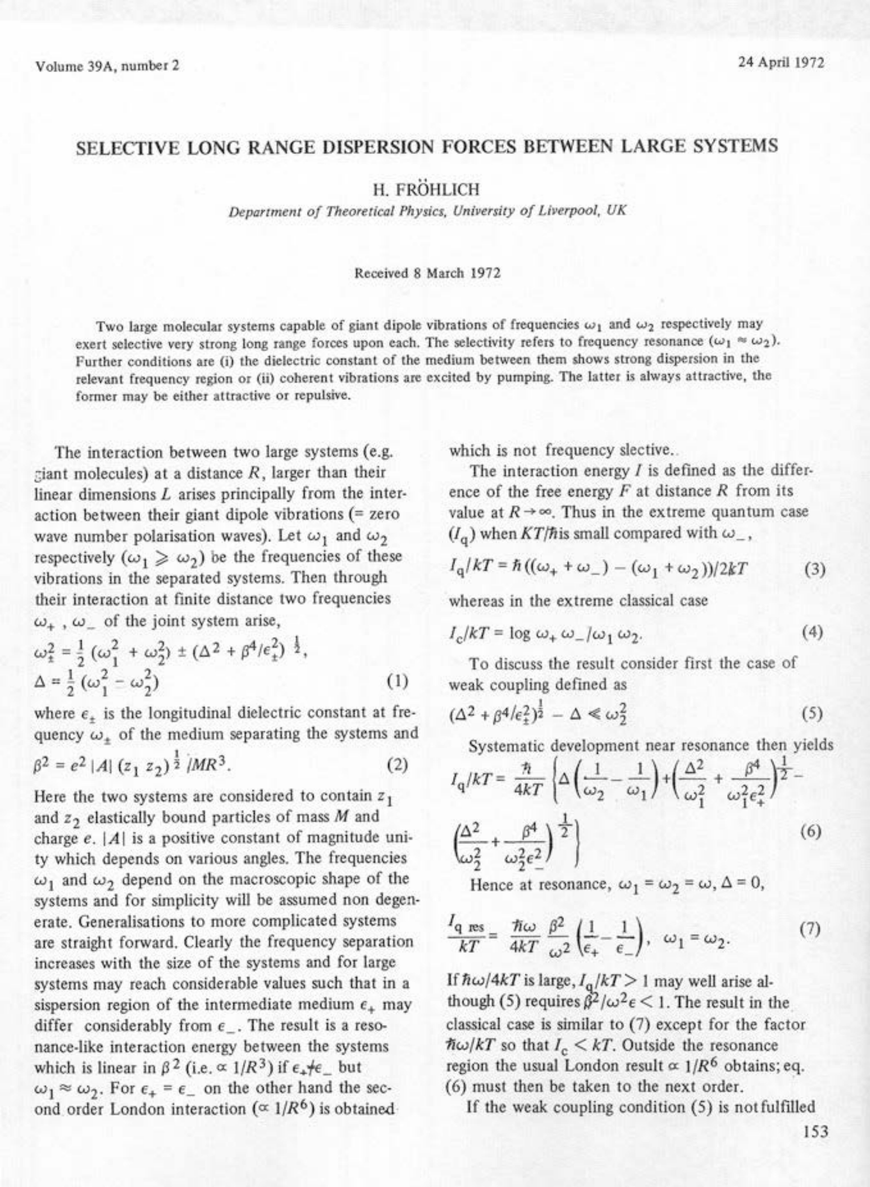
[33] Holland pointed out that in the case of two separated oscillating dipoles, which for simplicity he assumed to be parallel, each having no static dipole moment, there is an attractive force between them when they oscillate in anti-phase, and a repulsive force when they have the same phase. Thus, the average force over a sufficiently long time, τ, will tend to zero *unless* the frequencies are *nearly identical*, with a frequency difference *less* than $1/\tau$.

[34] It is possible that the deficit could be (non-resonantly) supplied by a heat current associated with small temperature *differences* ($\sim 0.1°$) produced by localised microwave heating—homogeneous changes in temperature by several degrees being found to yield negligible effects (Kremer et al. 1985)—in which case, we have the interesting situation of a non-thermal effect (the establishment of coherence) being produced thermally by localised heating.



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SELECTIVE LONG RANGE DISPERSION FORCES BETWEEN LARGE SYSTEMS

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Two large molecular systems capable of giant dipole vibrations of frequencies ω_1 and ω_2 respectively may exert selective very strong long range forces upon each. The selectivity refers to frequency resonance ($\omega_1 \approx \omega_2$). Further conditions are (i) the dielectric constant of the medium between them shows strong dispersion in the relevant frequency region or (ii) coherent vibrations are excited by pumping. The latter is always attractive, the former may be either attractive or repulsive.

The interaction between two large systems (e.g. giant molecules) at a distance R, larger than their linear dimensions L arises principally from the interaction between their giant dipole vibrations (= zero wave number polarisation waves). Let ω_1 and ω_2 respectively ($\omega_1 \geqslant \omega_2$) be the frequencies of these vibrations in the separated systems. Then through their interaction at finite distance two frequencies ω_+, ω_- of the joint system arise,

$$\omega_\pm^2 = \tfrac{1}{2}(\omega_1^2 + \omega_2^2) \pm (\Delta^2 + \beta^4/\epsilon_\pm^2)^{\frac{1}{2}},$$
$$\Delta = \tfrac{1}{2}(\omega_1^2 - \omega_2^2) \tag{1}$$

where $\epsilon_\pm$ is the longitudinal dielectric constant at frequency $\omega_\pm$ of the medium separating the systems and

$$\beta^2 = e^2\,|A|\,(z_1 z_2)^{\frac{1}{2}}/MR^3. \tag{2}$$

Here the two systems are considered to contain z_1 and z_2 elastically bound particles of mass M and charge e. $|A|$ is a positive constant of magnitude unity which depends on various angles. The frequencies ω_1 and ω_2 depend on the macroscopic shape of the systems and for simplicity will be assumed non degenerate. Generalisations to more complicated systems are straight forward. Clearly the frequency separation increases with the size of the systems and for large systems may reach considerable values such that in a sispersion region of the intermediate medium ϵ_+ may differ considerably from ϵ_-. The result is a resonance-like interaction energy between the systems which is linear in β^2 (i.e. $\propto 1/R^3$) if $\epsilon_+ \neq \epsilon_-$ but $\omega_1 \approx \omega_2$. For $\epsilon_+ = \epsilon_-$ on the other hand the second order London interaction ($\propto 1/R^6$) is obtained which is not frequency slective.

The interaction energy I is defined as the difference of the free energy F at distance R from its value at $R \to \infty$. Thus in the extreme quantum case (I_q) when $KT/\hbar$ is small compared with ω_-,

$$I_q/kT = \hbar((\omega_+ + \omega_-) - (\omega_1 + \omega_2))/2kT \tag{3}$$

whereas in the extreme classical case

$$I_c/kT = \log \omega_+ \omega_- / \omega_1 \omega_2. \tag{4}$$

To discuss the result consider first the case of weak coupling defined as

$$(\Delta^2 + \beta^4/\epsilon_\pm^2)^{\frac{1}{2}} - \Delta \ll \omega_2^2 \tag{5}$$

Systematic development near resonance then yields

$$I_q/kT = \frac{\hbar}{4kT}\left\{\Delta\left(\frac{1}{\omega_2} - \frac{1}{\omega_1}\right) + \left(\frac{\Delta^2}{\omega_1^2} + \frac{\beta^4}{\omega_1^2\epsilon_+^2}\right)^{\frac{1}{2}} - \left(\frac{\Delta^2}{\omega_2^2} + \frac{\beta^4}{\omega_2^2\epsilon_-^2}\right)^{\frac{1}{2}}\right\} \tag{6}$$

Hence at resonance, $\omega_1 = \omega_2 = \omega$, $\Delta = 0$,

$$\frac{I_{q\,\mathrm{res}}}{kT} = \frac{\hbar\omega}{4kT}\frac{\beta^2}{\omega^2}\left(\frac{1}{\epsilon_+} - \frac{1}{\epsilon_-}\right), \quad \omega_1 = \omega_2. \tag{7}$$

If $\hbar\omega/4kT$ is large, $I_q/kT > 1$ may well arise although (5) requires $\beta^2/\omega^2\epsilon < 1$. The result in the classical case is similar to (7) except for the factor $\hbar\omega/kT$ so that $I_c < kT$. Outside the resonance region the usual London result $\propto 1/R^6$ obtains; eq. (6) must then be taken to the next order.

If the weak coupling condition (5) is not fulfilled

Fig. 6.5 Paper dealing with the selective long-range interaction between biological systems [F150]—Reproduced with the permission of Elsevier interaction that is realised in thermal equilibrium [F150] can, with the onset of coherence, be transformed into an attractive one (Hyland 1998). It was later realized that this frequency-specific attraction could form the basis of a field-assisted ‘intelligent’ biochemistry (Del Giudice 1993), whereby a precise sequence of biochemical reactions can be internally orchestrated and externally influenced by exposure to electromagnetic fields of appropriate frequencies and intensities for sufficient lengths of time.

Experimental support for this attractive interaction was later provided by the phenomenon of *rouleaux* formation in a population of red blood cells, in which, below a certain separation of the order of 1 μm (which is orders of magnitude greater than the range of chemical forces), the cells rush together to form coin-like stacks (*rouleaux*) at a much faster rate than is expected on the basis of Brownian motion (Rowlands 1994, wherein references to earlier work can be found). It is particularly significant, in the light of the foregoing considerations, that when the membrane potential is removed, or when metabolism is inhibited, the rate reduces to the Brownian value. Furthermore, in the case of a mixture of different mammalian blood cells, it is found that the rouleaux formation takes place only between cells of the *same* species (presumably characterised by the same coherent frequency), again in contrast to the behaviour expected on the basis of Brownian motion. Rothschild’s original suggestion (*loc. cit.*) to use blood as a system with which to investigate Fröhlich’s early ideas was thus highly prescient.

6.3.4 A Static Coherent Excitation

The following year (1973), elaborating some qualitative considerations he had presented in 1967 at the first meeting organised by *l’Institut de la Vie* [F138], Fröhlich showed [F154] how a quite different kind of coherent excitation—a *static* one—can arise in which the electric dipole moment induced in a bio-molecule by the enormous transmembrane electric field gets (meta) stabilised—via elastic displacements, in consequence again of the *shape* dependence of the dielectric polarization energy—as a quasi-ferroelectric state characterised by a *non*-zero average polarization—i.e. $<P> \neq 0$. In contrast to the dynamic excitation (for which the average electric polarization vanishes, $<P> = 0$, but $<P^2> \neq 0$), this static one is characterised by a minimum energy, rather than a critical energy *rate*. It should be noted that, as early as 1970, he had pointed out [F145] how this elastically stabilised highly polar state might help in overcoming certain difficulties in understanding the action of enzymes.

Experimental support for the static coherent excitation later[35] came from the dramatic change in the field dependence of the dielectric response of Langmuir-Blodgett

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[35] For earlier supporting evidence, the work of Mascarenhas (1975) and of Kolias and Melander (1976) should be consulted.

layers of haemoglobin, which is observed when an applied electric field reaches 10^5 V/cm; the effect was found to last for a considerable time, during which the system behaved as a ferroelectric (Hasted et al. 1981).

6.3.5 The Experimental Situation, and the Problem of Replication

The first experimental evidence in support of the dynamic coherent excitation came in 1974 from spectroscopic millimetre microwave studies made in the former *USSR* on a variety of living systems during the 1960s and early 1970s, the existence of which Fröhlich discovered quite by accident. One Saturday morning in the winter of 1974/5, during which there were severe fuel restrictions in the *UK* owing to shortage of coal due to industrial strike action, he went into the departmental library in an attempt to keep warm. There, quite by accident, he opened the current issue of Soviet journal *Uspekhi* to find a report (Devyatkov 1974) of a meeting of the *USSR* Academy of Sciences, which had been held the previous January; the report contained nine contributions demonstrating striking influences of low intensity coherent *mm* microwave radiation on a variety of living systems, ranging from yeast to the bone marrow of mice.

The observed spectra were found to have the following properties in common: (i) the effects of irradiation were strongly dependent on the frequency of the microwaves, (ii) there was an associated power (intensity) threshold below which no effect was observed, and above which the effects of exposure depended only weakly on further increases of power over several orders of magnitude, (iii) the occurrence of some effects depended on the duration of irradiation, exposure for a certain minimum time being necessary for an effect to manifest itself. The first two of these properties are entirely consistent with those expected [F159] if a dynamic coherent excitation is involved, whilst the third property was later found [F176, Hyland (2002)] also to be understandable.

In view of the paucity of detail in the Russian reports, Fröhlich set about persuading various experimental groups to attempt to reproduce some of the results. From this followed numerous research programmes, some of which he actively participated in, particularly in Germany, at the Max Planck Institute in Stuttgart (under the direction of F. Keilmann) and the Institute for Biophysical Radiation Research near Munich (under the direction of W. Grundler), where the influence of ultra-weak microwave radiation on the growth rate of single cells of the yeast *Sacccharomyces cerevisiae* was intensively investigated between 1977 [F170] and 1992 (Grundler and Kaiser 1992). In the latter publication, irradiation in the G_1-phase was studied at intensities as low as 5 pW/cm^2 (amplitude modulated at 8 kHz), revealing an *extremely sharp* dependence on frequency[36] ($Q \sim 10^4$), similar to the response of a self-sustained (limit cycling) oscillator—namely, a central maximum (enhanced growth rate), flanked on each side by minima (depressed growth rate), the width of the resonance (near 41 GHz) being found to *decrease* with decreasing irradiation power.

Other highly resonant interactions with ultra-weak microwave radiation characterize the ‘switch-on’ of certain epigenetic processes, such as the induction of λ-phage (near 70 GHz) first found in 1979 (Webb 1979), which was later independently corroborated (Lukashevsky and Belyaev 1990).

Experimental evidence that the cell membrane is a site of coherent oscillations—as originally envisaged by Fröhlich—was provided in 1985 by the *similarity* in the microwave absorption spectra of red blood cells and their ‘ghosts’—i.e. cells that are devoid of cytoplasm and organelles—the resonant frequency here being near 36.7 GHz (Blinowska et al. 1985).

Another set of experimental results consistent with the existence of coherent excitations in active biosystems came from laser Raman studies on metabolically active bacterial cells (such as *E. coli*) which revealed: (i) high intensity peaked spectra near 5×10^{12} Hz, which appear[37] only at certain stages in the evolution of the systems investigated (Webb et al. 1977; Webb 1980; Bannikov et al. 1980), (ii) an anti-Stokes/Stokes intensity ratio close to *unity* (as opposed to the thermal value of 0.5 at physiological temperatures), consistent [F171] with the existence a strongly supra-thermally excited mode in the *E. coli* system investigated.

Other more recent experimental work consistent with Fröhlich’s coherent excitations includes: (i) the detection of coherent nuclear motion in membrane proteins at frequencies in the *sub*-millimetre band, typically THz (Vos et al. 1991), (ii) the detection of coherent nanomechanical motion in the cell wall of the yeast *S. cerevisiae* at around 1 kHz, which is allied to metabolic activity[38] (Pelling et al. 2004). In this connection, it is relevant to note that oscillating electric (near) fields in the kHz and MHz range have been detected by a variety of techniques in the immediate vicinity of these and certain other cells near cell division (Smith et al. 1987; Pohl and Pollock 1986, 1988).

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[36] It should be noted that the failure of other attempts (e.g. Furia et al. 1986; Gos et al. 1997) to reproduce the effect found by Grundler et al. do not *necessarily* invalidate the latter’s positive results, on account of crucial **differences** in experimental protocol, which **undermine the fidelity of the purported replications**—such as the stage in the cell cycle at which irradiation takes place, the way in which growth rate is measured, the density of cells, the method used for synchronization, etc.

[37] Again, as in the case of experiments to investigate whether exposure to weak microwave radiation at specific frequencies has any effect on cell division, not all attempts to replicate these *positive* findings have been successful—e.g. Layne and Bigio (1986)—but, in many cases, the same criticism as mentioned in the preceding Footnote applies, namely that the purported replication experiments are, for various reasons, actually *different* experiments.

[38] It will be recalled that this is of the same order as the *modulation* frequency used by Grundler and Kaiser (1992).

On the other hand, attempts to detect a *radiative* electromagnetic field associated with a biological coherent excitation continue,[39] although the emission of coherent biophotons from living systems *is* now established—but only across the visible (Popp and Chang 1998). The possibility of large up-conversions and mode coupling between Fröhlich's microwave coherent excitations and visible photons in biological systems has been considered by Swain (2006) *see also* (Popp 2006, 2008).

6.3.6 A Synthesis of the Two Kinds of Coherent Excitation: A Model for Electrical Brain-wave Activity

A particularly ingenious synthesis of Fröhlich's static and dynamic coherent excitations, together with the long-range frequency-specific attraction associated with the latter—which represents what might be the considered to be the pinnacle of his "*oeuvre biologique*"—produced a model in terms of which it was possible to understand certain aspects of the electroencephalogram (*EEG*), or brain-waves, as known at the time. Specifically, (i) how, despite their small amplitude, such excitations can persist in the presence of thermal noise, without requiring the involvement of enormous volumes of brain tissue, and (ii) the hyper-sensitivity of brain function to ultra-weak external electromagnetic fields. The extremely low frequencies of the *EEG* ($\sim 0.5 - 60$ Hz) in comparison to those associated with dynamic coherent excitations ($\sim$GHz), strongly suggested to Fröhlich that they must arise in some rather subtle, *secondary* way. He first presented his ideas a Work Session of the Neurosciences Research Programme held at *MIT* in November 1974, organised by W. Ross Adey and Suzanne Bawin, the proceedings of which were published in their Bulletin in 1977 [F167], and later summarised in other publications [F176, F194, F195, Fröhlich and Hyland (1995)]. His model was based on periodic, autocatalytic (self-sustaining) reactions within an enzyme system assumed localised in the Greater Membrane of the brain, involving the cyclical excitation and de-excitation of the enzymes through chemical reaction with a substrate system. These reactions are assumed to be maintained by the predicted long-range attractive interaction proportional to R^{-3}. If in their excited state (but not in their ground-state) the enzymes possess a large electric dipole moment that is (meta)stabilised as a static coherent excitation by elastic deformation, an oscillatory electric polarization will accompany these periodic chemical reactions.

If S is the number of substrate molecules and N (Z) the number of excited (non-excited) enzymes in a certain region, then the rate of increase in the number of activated enzymes, when spontaneous transitions back to the non-polar ground-state (characterised by the rate constant β) are included, is given by:

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[39] In this connection it should be appreciated that because of the longitudinality of Fröhlich's coherent excitation (in bulk), radiation can only arise as a side-effect of the existence of internal bounding surfaces that endow the excitation with a degree of tranversality (Hyland 2002).

$$dN/dt = \alpha NZS - \beta N \tag{6.3.9}$$

For every enzyme excited, one substrate molecule is chemically destroyed. If, however, the energy released by the spontaneous transitions to the ground-state is assumed to excite a high-frequency coherent mode in the remaining substrate molecules, then through the associated long-range, frequency-selective R^{-3} force, fresh substrate molecules will be attracted into the region, which will replenish their population; if the rate of attraction is γ, then:

$$dS/dt = -\alpha NZS + \gamma S \tag{6.3.10}$$

Finally, Z satisfies the following equation, where, like γS, the term $\lambda(A - Z)$ arises from the long-range attractive interaction:

$$dZ/dt = -dN/dt + \lambda(A - Z) \tag{6.3.11}$$

It was assumed that equilibrium of non-excited enzymes is reached very quickly (equivalent to assuming $\lambda >> \alpha, \beta$), so that Z can be replaced by A, and thus neglected in analysing the dynamics of N and S. The two remaining (Lotka-Volterra) equations, Eqs. 6.3.9 and 6.3.10, have oscillating solutions (relative to their time-independent values, N_o and S_o), representing period chemical reactions, the frequency of which, to first order in small quantities, is $(\gamma\beta)^{1/2}$—i.e. the frequency is determined, not by any coherent mode, but by parameters that characterise the much slower chemical reactions. As a consequence, electric vibrations with the *same* frequency are generated at each enzyme site, the spatial arrangement of which determines the extent to which these signals are observable at a distance, since external electrical detection will normally integrate the contribution of all the enzymes in the system.

For an appropriate spatial arrangement, the polarized enzymes may exhibit, via short-range interactions, a tendency towards forming a ferroelectric state, which will increase the rate of activation; this increase will be offset, however, by the resistive damping of the electric current associated with the periodically changing polarization. It was found, provided the damping is not too large, that the oscillations of the polarization field exhibits *limit cycle*[40] behaviour, making them highly sensitive to (external) electrical and (internal) chemical influences. It is with these *electrical limit cycles* [the frequency of which *differs* from $(\gamma\beta)^{1/2}$ (Kaiser 1977)] that brain-waves find interpretation in the model, where, in consequence of the tendency

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[40] A limit cycle is an oscillation in a nonlinear system that, whatever way it starts, approaches at later time a unique, self-sustaining state of stable periodic motion. It will be recalled that the response of yeast to ultra-weak microwaves was found to be similar to the response of a self-sustained (limit cycling) oscillator—namely, a central maximum (enhanced growth rate), flanked on each side by minima (depressed growth rate) (Grundler and Kaiser 1992).

towards the ferroelectric state, large areas of the enzyme sub-system must *oscillate coherently,* as is indeed found experimentally (Elul 1974).

In turn, it was noted that in consequence of the possibility of inducing a collapse of this electric limit cycle by exposure to a weak *external* oscillatory electric field, sufficient energy can be made available to initiate certain physiological processes (such as nerve conduction, for example), the threshold of which *exceeds* the energy available in the incoming signal alone. In effect, the limit cycle stores the energy of the incoming signal until its (finite) storage capacity is reached, after which it collapses—i.e. the role of the external field is simply to “trigger” the collapse of the limit cycle, the stored energy thereby released effectively amplifying the (small) incoming signal energy, possibly to a physiologically significant level.[41] Theoretical study of the interaction of external electric fields with the limit cycle was undertaken almost immediately by one of Fröhlich’s collaborators in Germany, and was the subject of ongoing research for many years (Kaiser 1992, 1996), one of the most interesting findings being that in the presence of an external oscillatory electric field the model can, under certain conditions, exhibit deterministic chaos (Kaiser 1985).

Quite independently of these developments, attention turned, during the burgeoning of interest in chaos theory during the 1980s, to the seemingly stochastic appearance of empirical *EEG* signals, in attempts to ascertain whether the observed behaviour could be attributed to the existence of low-dimensional strange (chaotic) attractors, rather than to random noise, as had hitherto been done. From time-series analyses of experimental *EEG* data, obtained on humans under a wide variety of different conditions—ranging from high levels of conscious activity to epileptic seizure—the existence of relatively low-dimensional attractors was ubiquitously established (Gallez and Babloyantz 1991). Perhaps somewhat surprisingly, the attractor dimension was found to *increase* with increasing the degree of conscious awareness, as also did the number of positive Lyapunov-exponents; evidently, the higher the level of chaos, the greater is the “flexibility” of the system—a flexibility that guarantees a *much greater robustness* than would otherwise obtain.

The inferred presence of chaotic attractors of various dimensions in the dynamical system responsible for the observed brainwave activity clearly invalidates the restriction to two variables made in earlier analyses of Fröhlich’s model, and argues in favour of reinstating the dynamics of the Z population, since at least 3 degrees of freedom are necessary for the possibility of deterministic chaos; a start in this direction was made some time ago (Uçar and Hyland 2000). Even if the generalised model *is* found to yield a strange attractor under appropriate conditions, it still cannot necessarily be considered as a realistic model for human brain-wave activity, since the *lowest* embedding dimension deduced from time-series analyses of *EEG* data is found (Babloyantz and Destexhe 1986) to be equal to 5 (corresponding to the case of *petit mal* seizure)—i.e. two more than characterise the model; it must be

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[41] This could be relevant also to the understanding to the ultra-sensitive response of some living biosystems to ultra-weak microwave radiation mentioned earlier.

remembered, however, that quantitative estimation of the correlation and embedding dimensions is plagued with a certain degree of uncertainty.

It must thus be concluded that the real value of Fröhlich's approach to this problem is not quantitative, but rather illustrative, namely, to show how application of the concept of coherence and its consequences to a simple model of an active biosystem can afford a novel insight into a long-established phenomenon—an insight that is, furthermore, framed in the same non-linear dynamical language that is now routinely used to analyse time-series *EEG* data from which information into the nature of the phenomenon has in recent years been obtained, and which can now be used to refine the original model accordingly. In this way, it may then possible to correlate attractor topology, for example—and hence the associated degree of conscious awareness—with specific model parameters whose physical interpretation is clear. In turn, it might then be possible to identify ways (both physical and chemical) of changing the values of these parameters and, in turn, the attractor topology: i.e. the model offers the possibility of systematic study of the dependence of the degree of conscious awareness (as here represented by attractor topology) on the values of specific physical parameters characterising the underlying deterministic, non-linear model of brainwave activity, such as the electrical resistivity of brain tissue, for example, which governs the dissipation in the system, which is essential its limit cycle behaviour.

Fröhlich's brain-wave model is a good example of the 'broad-brush' approach that, on occasion, characterised his work, namely, tossing out a few novel or suggestive ideas, but leaving the elaboration of the details and implications to others.

6.3.7 Some General Considerations

The real importance of Fröhlich's pioneering work on coherent excitations in living systems is that it directed attention from biological structure to biological *function*. His work continues to generate considerable interest because of: (i) the variety of possibilities it offers of understanding the non-thermal, ultra-sensitivity of living systems to very weak electromagnetic radiation at specific frequencies, (ii) the rather unexpected role that *macroscopic quantum effects* apparently play in living systems (Smith 1998)—a role that has been invoked in consciousness studies (Hyland 1998).

In assessing the status of Fröhlich's daring speculation concerning the possibility of coherent excitations in living systems at room temperature as an emergent feature of their far-from-thermal equilibrium, dissipative character, it is essential to appreciate that this possibility transcends any particular *mechanism* whereby coherence might be established, such as the three models discussed above. For these particular theoretical models show only the *possibility* of such excitations; they do not enjoy a monopoly—indeed, quite different models might well lead to similar results:

> In fact if, as we think, biological activity makes great use of these excitations, the manner of establishing them need not be unique [F195, p. 259].

Fröhlich considered that the role of theoretical physics is here to tentatively introduce physical concepts via which our understanding of living systems might be deepened. To decide whether they are actually appropriate, however, requires close collaboration with experiment, since theoretical physics cannot here make predictions in the way that it can for materials near thermal equilibrium, where they are in the realm of linear response; the far-from-equilibrium nature of living systems precludes any such systematic approach. Furthermore, many biological properties may be the result of long evolutionary processes, and whilst theory can suggest a number of possibilities, it cannot predict the way in which evolution has *actually* progressed:

> It must be asked therefore whether the relevant properties are possible, rather than whether they are probable [F(iv), p. 2].

On the other hand, as he noted [F186 (p. 1), F195 (p. 243)], it is here permissible to ask questions that in physics are strictly forbidden (except when dealing with machines, and similar constructions), such as: 'What is the *purpose* of a particular excitation?'

Turning to the problem of the experimental reproducibility of non-thermal biological effects allied to coherent excitations, it must first be emphasised that, in many cases, positive results were obtained only after many failures, even by the same experimental group. Furthermore, as already noted, it must be appreciated that there are often subtle (or not so subtle) differences in experimental protocol that can undermine the fidelity of purported replication experiments by independent groups, such as differences in the stage in the cell cycle at which investigations were carried out, and differences in exposure conditions (near vs. far field, differing durations of exposure, the use of different irradiation intensities, etc.). There are thus many parameter 'windows' that have to be respected before replication can possibly be realised.

Four further points should be noted: (i) non-thermal influences are often 'opposite' to thermally produced ones, as illustrated, for example by the growth rate of *E. coli*, which is increased by heat, but decreased (in a highly frequency–dependent manner) by exposure to low intensity microwave radiation. Accordingly, it is quite possible for such contra-thermal non-thermal effects to be thermally obliterated, unless sufficiently low intensities are used, consistent with the finding that non-thermal effects are often more pronounced the lower the irradiating intensity; (ii) great care must be exercised to ensure that a particular experimental intervention does not interfere with endogenous biological activity; a good example of this is the development of a flow-through system to minimize the time for which an active biological system is exposed to potentially deleterious effects of the laser beam used in Raman effect experiments (Drissler and Santo 1983); (iii) the physiological state of a living system generally depends on its history, which cannot be assumed to have been the same even for identical species; (iv) because of the highly non-linear character of living systems, the possibility exits that attempts to replicate a given experiment are undermined by the presence of deterministic chaos (Kaiser 1987), [F(iv), p. 4], whereby the slightest difference between nominally identical systems can entail vastly differing evolutions.

Fröhlich's biological coherent excitations—the dynamic one, in particular—have stimulated a vast amount of theoretical work, both at the level of the rate equation (and its generalization) and with attempts to develop a Hamiltonian formulation of his model (Hyland 2002) and to connect the model with symmetry breaking (Del Giudice[42] et al. 1988a, b—*see also* Dürr 2006). In addition, this work led to the establishment of series of international conferences, such as those organised by *l'Institut de la Vie*, which continued for many years until around 1988, and those that have been held biennially at Charles University in Prague, the first of which (*Biophysical Aspects of Cancer*) Fröhlich attended in 1987. The centenary in 2005 of his birth was celebrated in Prague in July 2005 by an international meeting *Coherence and Electromagnetic Fields in Biological Systems* (Liboff 2005), and by similar meetings at the International Institute for Biophysics in Neuss in August 2005, and in Liverpool, the following April (Hyland and Rowlands 2006, 2008).

The theoretical and experimental situation as of 1980 was reviewed in a long paper [F176], dealing with non-thermal effects of microwaves on biological systems (Fig. 6.6).

Subsequent work was presented in contributions to two books: *Coherent Excitations in Biological Systems* [F(iii)], and *Biological Coherence and Response to External Stimuli* [F(iv)], published by Springer, in 1983 and 1988, respectively, both of which Fröhlich edited (Fig. 6.7)[43]; for the second (published when he was 82) he also contributed the opening chapter. Therein, in addition to reviewing and updating earlier work, and following Bloch (1968), he drew attention to the important point that magnetic flux quantization is not restricted to superconductors, but is a completely general property of all materials—including biological ones, where, as he noted, the small size of cells should facilitate its observation. In 1986, he also published 2 further extensive and valuable review articles on his coherent excitations [F194, F195]. In all, he published (either singly or in conjunction with others) 48 papers devoted exclusively to matters relating to either theoretical or experimental issues connected with his predicted biological coherent excitations, which is 3 more than he published in the field of dielectrics.

Interest did not cease with Fröhlich's death, as evidenced, for example, by the publication 7 years later of a monograph by Pokorný and Wu (1998) entitled *Biophysical Aspects of Coherence and Biological Order*, and by the on-going conferences in Prague.

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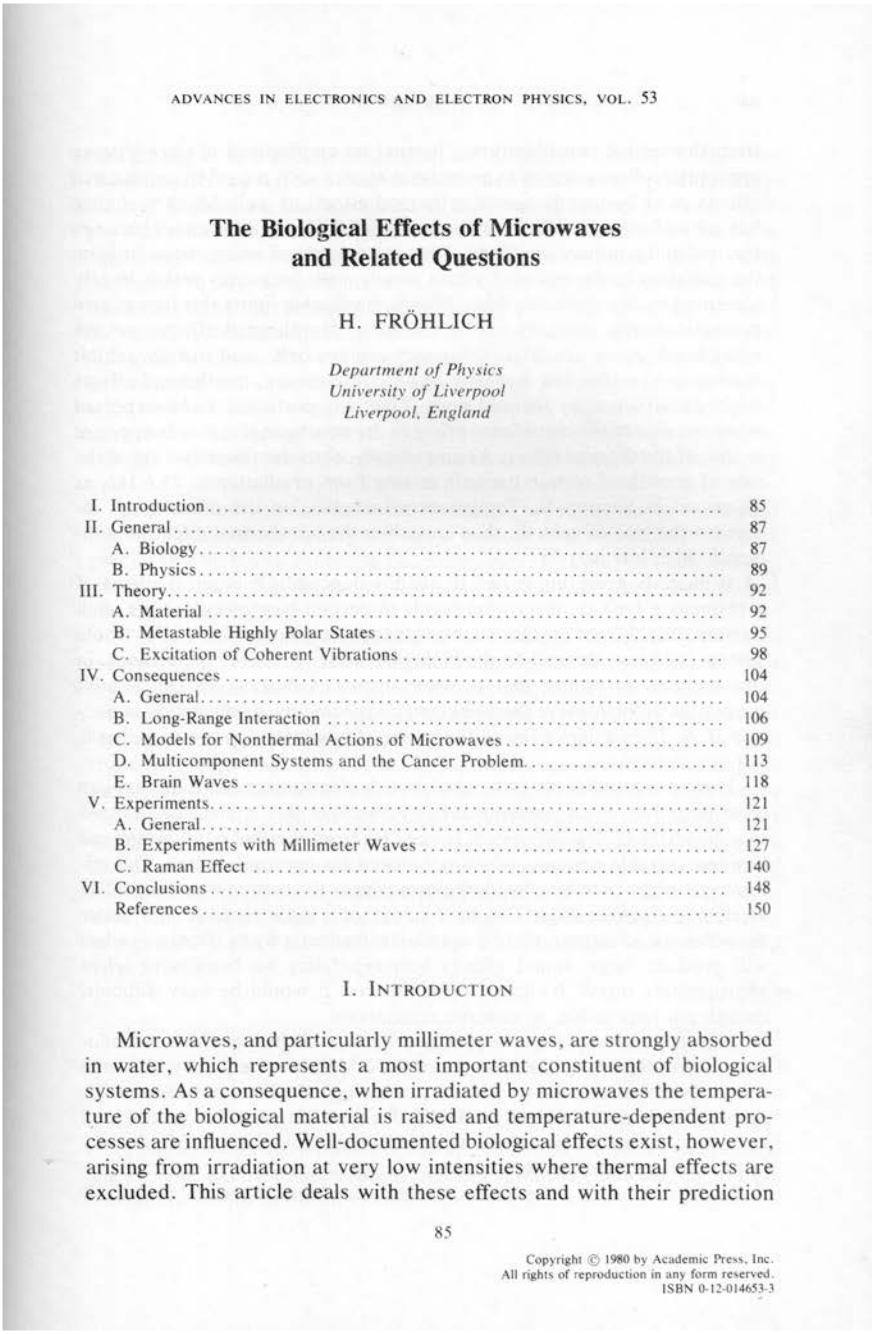
[42] Later development of work by Preparata (1995) predicted the existence in water of coherent domains containing strong evanescent 60 THz electric fields (Arani et al. 1995); in contrast to Fröhlich's case, the coherence is here realised *without* pumping, in consequence an instability (a kind of macroscopic Lamb shift) in the usual ground-state when account is taken of a coupling between certain electronic excitations of the water molecules and the zero-point fluctuations of the electromagnetic field—*see also* (Del Giudice 2008).

[43] In the case of [F(iii)], F. Kremer was joint editor.



ADVANCES IN ELECTRONICS AND ELECTRON PHYSICS, VOL. 53

The Biological Effects of Microwaves and Related Questions

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I. INTRODUCTION

Microwaves, and particularly millimeter waves, are strongly absorbed in water, which represents a most important constituent of biological systems. As a consequence, when irradiated by microwaves the temperature of the biological material is raised and temperature-dependent processes are influenced. Well-documented biological effects exist, however, arising from irradiation at very low intensities where thermal effects are excluded. This article deals with these effects and with their prediction

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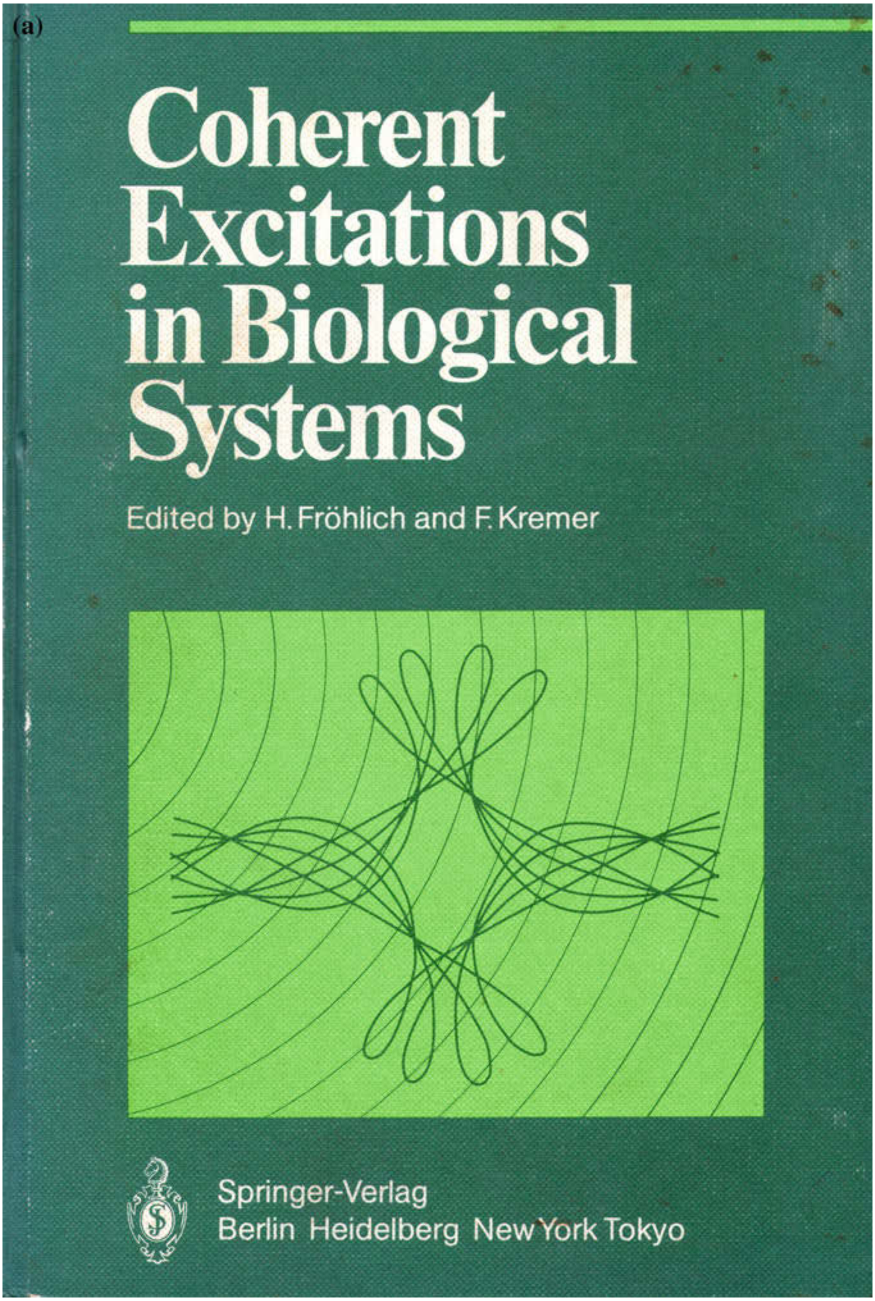
Fig. 6.6 Review article on the biological effects of microwaves [F176]—Reproduced with the permission of Elsevier

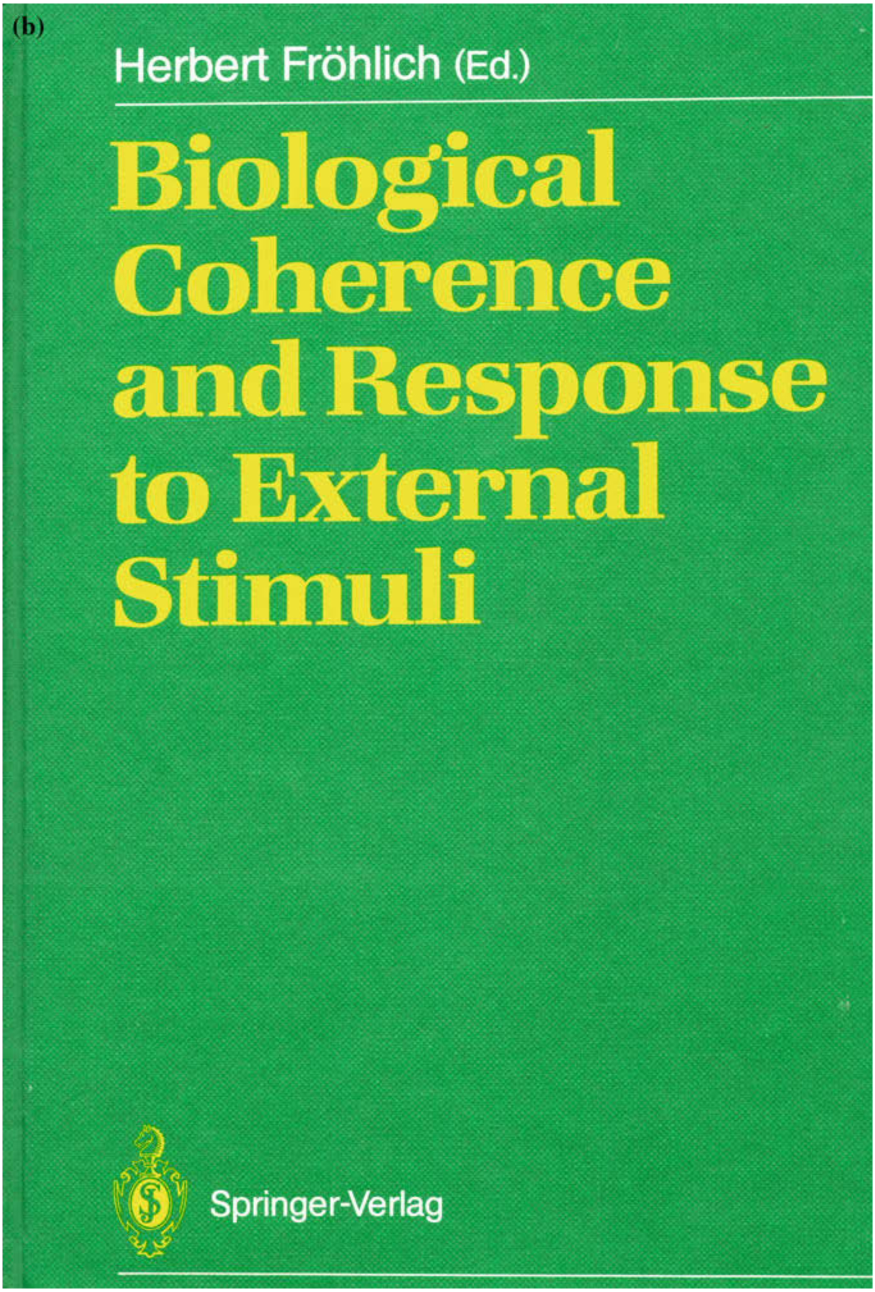


Coherent Excitations in Biological Systems

Edited by H. Fröhlich and F. Kremer

Springer-Verlag
Berlin Heidelberg New York Tokyo

Fig. 6.7 Two Springer publications edited by Fröhlich, dealing with coherent excitations in biological systems: **a** 1983; **b** 1988



(b)

Herbert Fröhlich (Ed.)

Biological Coherence and Response to External Stimuli

Springer-Verlag

Fig. 6.7 (continued)

6.3.8 Cancer

As already mentioned, the real importance of Fröhlich's pioneering work on coherent excitations in living systems is that it directed attention from (quasi-static) biological structure to dynamic biological function. Of particular interest, in this respect, were his ideas [F(iv), F174, F176] concerning cancer, as understood at the time. It will be recalled that in his first biological publication [F138], Fröhlich noted that in a cell in which a polarization wave is coherently excited, the associated elastic deformation of its surface could well act as a stimulus for cell division. Ten years later in 1977, he went on to show [F168] how a coherent excitation of an entire organ can exert a control over the division of its cells, and considered the implications for cancer.

He first noted that in normal tissues and organs, the rate of cell division is governed by the size and shape that an organ has reached at each stage of its growth, division ceasing once the organ is fully grown—except when it is required to replace damaged/dead cells. If the origin of this control could be identified, he believed that this could afford not only valuable insight into the problem of cancer where this control is absent or defective, but might also suggest novel therapeutic strategies. Clearly, any such control requires a long-range interaction between all (or most) of the cells in a particular organ or tissue. His idea, dating from the mid 1970s, when understanding of intra-cellular structure, cancer genetics and microbiology was far less advanced than now, was that the required control is perhaps based on a *global* coherent excitation that extends throughout the whole organ in question (its frequency being organ-specific), to which the coherent vibrations of the constituent cells each contribute with the appropriate phase; it was assumed that the site of each cellular excitation was the *DNA* protein complex in the cell nucleus [F168]. The global coherent vibration can be excited only once the organ has reached a size such that the number of contributing cells is large enough to permit the necessary *collective* polarization mode to be defined; just how large a system must be for this to be possible is governed by the magnitude of the fluctuations in the relevant macroscopic quantities [F143]. Below this size, cells can divide freely, the electro-strictive stress at the cell surface acting as a stimulus, as mentioned above. Once the cells are locked into the global coherent excitation, however, this stimulus ceases, since the deformations of individual cells become sublimated into an overall deformation of the whole organ; this affords a novel insight into the so-called 'contact inhibition' exhibited by cells in normal tissue.

Because of the long range of the interactions involved, such a global excitation itself depends on the size and shape of the organ,[44] and its spatial structure must,

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[44] Even neglecting the non-linear electro-strictive coupling referred to above, the frequency of the global coherent excitation must be expected to be somewhat lower than that of an individual cell in isolation. For owing to the Coulomb interaction between the individual cellular dipoles, their (common) frequency broadens into a band of the collective (i.e. global) modes, and it is the *lowest* such frequency that characterises the global coherent excitation.

in general, be expected to be much more complicated that of a giant dipole, and, accordingly, only very weakly optically active[45]; in addition, the long-range attraction between cells oscillating coherently with the same frequency [F150] contributes to the stability of the tissue.

Owing to their participation in the global coherent excitation of the whole organ, individual cells loose their autonomy, resisting, in particular, any attempt to have their own coherent frequency altered. Should a change in a cell's coherent frequency be a necessary precursor of cell division (as seems likely in view of the necessity of *DNA* to first uncoil), division will be inhibited unless energy is available to liberate the cell from the global coherent mode in which it is frequency/phase-locked; the required energy was assumed to come from nutrients, the amount of which gets progressively depleted with time. With successive cell division, the strength of the global vibration increases with the participation of daughter cells as they are frequency-entrained back into resonance,[46] and it is thereby able to exert a stronger influence over the autonomy of individual cells; accordingly, more energy (*less* of which is now available) is then required to free a cell before it can divide. These two negative feedback effects re-enforce one another, so that cell division becomes a *self-limiting* process, eventually ceasing all together (except when necessary to replace damaged/dead cells), whence the organ stops growing.

Within this scenario, Fröhlich's approach to cancer was the following: if, for some reason—such as an accumulation of intra–cellular mutations, or because of invasion by foreign molecules (carcinogens)—some cells loose the 'correct' frequency, they will thereby cease to be in resonance with the global excitation. Provided not too many cells are so affected, the global coherent vibration will be only slightly weakened, and still strong enough to exert control over the division of the remaining untransformed cells, and to entrain newly divided cells back into resonance (provided their frequency is not too different from that of the global coherent excitation). If, however, the number of aberrant cells exceeds a certain critical value, the global coherent excitation will be weakened to such an extent that it is rendered incapable of exerting its controlling and corrective influences. There then occurs a kind of phase transition to a disordered state of uncontrolled cell division characterised by a loss of coherence; this state, Fröhlich identified as *cancer.*

In this connection, it is of interest to note that a report in *Nature* of a meeting (*Oncogenes and growth control*) held ten years later at the European Molecular Biology Laboratory in Heidelberg contained the following statement:

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[45] Since (unlike the case of a cell) the size of an organ can be much *larger* than typical electromagnetic wavelengths, it is possible that the global coherent excitation is here predominantly *longitudinal.*

[46] After division, once the *DNA* complex has recoiled, the frequency will revert to the value corresponding to a cell in isolation, which, in general, will differ from that of the global coherent mode.

> Perhaps the most interesting new theme of the meeting was that normal cells, or their products, can exert control over those of their number that have been transformed into cancer cells (Newmark 1987).

Fröhlich concluded his 1977 review paper with the following statement:

> An important conclusion is then that, in the disordered (cancerous) state, order might be restored by external irradiation with the correct frequency. This, of course, would be the most startling influence of electromagnetic waves on biological systems[47] [F168].

There is a clear parallel here with the modality known as 'microwave resonance therapy', which was subsequently developed in the Ukraine during the 1990s (Sit'ko et al. 1996).

Fröhlich's identification of the cancerous state as one in which the **coherence** that characterises the non-cancerous state has broken down, later received experimental support from biophoton studies (Popp and Chang 1998) in the visible region, which revealed that photon emission from cancerous tissue is more abundant and *much less coherent* than is that from normal tissue; furthermore, the emission displays an opposite dependence on cell density, in parallel with the absence of contact inhibition amongst cancer cells.

Within the last 5 years, it has been shown how the mono-clonality of tumours might, in principle, be accommodated within the framework of Fröhlich's original approach; for this, and other related matters, see Hyland (2009). Recently, a number of very interesting papers have appeared (Pokorný 2009; Pokorný et al. 2014) in which a specific site of Fröhlich's coherent excitations, as relevant to cancer, is identified—namely, the microtubules of the cytoskeleton. In the establishment of the excitation, mitochondria are assumed to play two crucial roles: (i) their strong static electric field polarises the microtubules, (ii) the energy left over after their production of *ATP* and *GTP*—which has hitherto been considered as 'waste'—is used to pump the microtubular dipoles into coherent oscillation. In the case of mitochondrial dysfunction (a signature of human cancers, Warburg 1956), this energy supply is disrupted, entailing an associated disruption in the coherent excitation, and with it, a breakdown in the biological control it would otherwise exert—a novel insight that suggests that cancer treatment should target mitochondrial dysfunction.

6.4 Unfinished Business

During the late 1970s and 1980s, as a relaxation from his work in biology, Fröhlich briefly returned to particle physics, and was surprised, but gratified, to find that a tetrad with the *same* relativistically invariant properties as that which he had introduced in an ad hoc way 20 years earlier in his work (vide Sect. 5.5) on isobaric spin space [F116] was actually already contained in Dirac electron theory. Here, the tetrad is constituted by the 16 Dirac bilinear covariants, $\bar{\psi}Q\psi$, where Q involves

---

[47] These ideas were, to some extent, adumbrated in the early work of Lakhovsky (1951).

products of the Dirac γ-matrices. He found that the tetrad is most simply constructed in terms of $\rho^{\mu} = (\rho^{\mathrm{m}}, iI), \sigma^{\nu} = (\sigma^{\mathrm{n}}, iI)$, where ρ^{m} and σ^{n} are the six 4 × 4 matrices in terms of which the *non*-manifestly covariant form of the Dirac equation was first formulated, i denotes the Minkowski 'i', and I the identity. The tetrad, $\underline{\underline{U}}$, is thus a 4 × 4 array, with elements $\psi^{\dagger}\rho^{\mu}\sigma^{\nu}\psi$, each of which is a component of one of the 5 relativistic fields defined by the 16 bilinear covariants.

$$\underline{\underline{U}} \equiv \begin{pmatrix} J_1 & J_2 & J_3 & iS_0 \\ \pi_1 & \pi_2 & \pi_3 & iP \\ \mu_1 & \mu_2 & \mu_3 & is \\ iS_1 & iS_2 & iS_3 & -J_0 \end{pmatrix} \tag{6.4.1}$$

where J^{μ} is the electric 4-current density $(\sim i\bar{\psi}\gamma^{\mu}\psi)$, S^{μ} is the pseudo-vector spin current density $(\sim i\bar{\psi}\gamma^5\gamma^{\mu}\psi)$, π^{k} and μ^{k} are, respectively, the electric and magnetic moment components of the antisymmetric tensor $\left(\bar{\psi}[\gamma^{\mu}\gamma^{\nu} - \gamma^{\nu}\gamma^{\mu}]\psi/2i\right)$, s is the scalar $(\bar{\psi}\psi)$ and P is the pseudo scalar $(i\bar{\psi}\gamma^5\psi)$.

The tetrad was found, by inspection,[48] to be orthogonal, satisfying 38 quadratic relations that express the equality in length of all the rows (4 relations, such as $J^2 - S_o^2 = J_{\mathrm{o}}^2 - S^2$) and their mutual perpendicularity (6 relations, such as $\underline{J} \cdot \underline{S} - S_{\mathrm{o}}J_{\mathrm{o}} = 0$[49]), and similarly for the columns; in addition, there are 18 'cross' relations that mix columns and rows, such as $J_{\mathrm{o}}J - S_{\mathrm{o}}S = \underline{\mu} \times \underline{\pi}$. Perhaps surprisingly, all these properties were found to be relativistically invariant, as was the case with the tetrad introduced in [F116], a crucial role in ensuring this being played by the cross relations. He noted that the derivation of these quadratic relations is here very much simpler than the usual procedure (Kofink 1940; Takahashi 1983) using the Fierz identities (Fierz 1937). All 38 quadratic relations can actually be derived from 9, so that out of the 16 bilinear covariants only 7 (=16 − 9) are independent, consistent with the bilinear covariants being independent of the overall phase of the bispinors out of which they are constructed; there is, however, no unique choice for these 7 quantities, but $\underline{J}$, J_{o} and $\underline{S}$ recommend themselves through their evident physical interpretation.

In line with the bilocal considerations at the end of [F124] noted in Sect. 5.5, Fröhlich investigated whether the bilinear covariants, correspondingly generalised to $\bar{\psi}(x'')Q\psi(x')$, similarly form a tetrad with the *same* geometrical properties as locally. In his Yukawa Memorial Lecture [F193] in Kyoto in 1985, he reported that this was indeed the case, and furthermore, that the geometrical properties of the tetrad can be expressed with the help of the generators of SU_4, one for the rows, one

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[48] Subsequent unpublished work by the author showed that the orthogonality of the tetrad directly follows from the well-known relations satisfied by the Pauli matrices, in terms of which matrices the Dirac ρ and σ matrices can be expressed. The 'cross' relations (vide infra), on the other hand, which involve a mixing of rows and columns, are sufficient conditions to ensure the positivity of the determinant of the tetrad.

[49] This orthogonality was first realised in 1931, soon after the publication of Dirac's work (Uhlenbeck and Laporte 1931).

for the columns, whilst the cross-relations that mix rows *and* columns require $SU_4 \times SU_4$. Feynman was at the Kyoto meeting, and told Fröhlich that, in the local case, he too was familiar with certain algebraic relations between some of the 5 fields constituted by the bilinear covariants, but was unaware of their geometrical significance in terms of a tetrad, and had not pursued the matter any further because in the local case the dynamics of these fields require for their description the introduction of terms that are *not* expressible in terms of the fields alone. Fröhlich informed him that in bilocal extension, this is no longer the case, it being then possible to derive generalised (Dirac-like) wave-equations that collectively describe, *in closed form*, the dynamics of the tetrad fields.

Representing the tetrad as a 16-component column functional, these equations take the form:

$$\left[\Gamma'_{\mu}\partial'_{\mu} + m\right]U(x',x'') = 0, \quad \left[\Gamma''_{\mu}\partial''_{\mu} + m\right]U(x',x'') = 0, \tag{6.4.2}$$

where Γ'_{μ} and Γ''_{μ} are the left and right direct products of the Dirac γ_{μ} with the identity, and U satisfies $U(x',x'') = U^*(x'',x')$. Apart from their dependence on (x', x'') the structure of these two sets of equations is formally similar in structure to that of the (local) Bargmann-Wigner equations.

Introducing 'external' and 'internal' coordinates, X and ξ, through $X \equiv \frac{1}{2}(x' + x'')$, $\xi \equiv \frac{1}{2}(x' - x'')$, the 32 equations take the form:

$$\left[\frac{1}{2}(\Gamma'_{\mu} + \Gamma''_{\mu})D_{\mu} + \frac{1}{2}(\Gamma'_{\mu} - \Gamma''_{\mu})\Delta_{\mu}\right]U(X,\xi) = -2m\, U(X,\xi), \tag{6.4.3}$$

$$\left[\frac{1}{2}(\Gamma'_{\mu} - \Gamma''_{\mu})D_{\mu} + \frac{1}{2}(\Gamma'_{\mu} + \Gamma''_{\mu})\Delta_{\mu}\right]U(X,\xi) = 0 \tag{6.4.4}$$

where, in the presence of an external 4-potential, A_{μ},

$$D_{\mu} \equiv \partial^{X}_{\mu} - ie/\hbar c\left[A_{\mu}(x') - A_{\mu}(x'')\right], \tag{6.4.5}$$

$$\Delta_{\mu} \equiv \partial^{\xi}_{\mu} - ie/\hbar c\left[A_{\mu}(x') + A_{\mu}(x'')\right] \tag{6.4.6}$$

The matrix combinations $(\Gamma'_{\mu} \pm \Gamma''_{\mu})$ each satisfy the Duffin-Kemmer β_{μ}-relations (Kemmer 1939), but are not independent. Indeed, it has long been known that β_{μ} can be expressed in terms of the two commuting sets of matrices, Γ'_{μ} and Γ''_{μ}, according[50] to $\beta_{\mu} = \frac{1}{2}(\Gamma'_{\mu} + \Gamma''_{\mu})$, whence the *local* Duffin-Kemmer (*DK*) equation,[51] takes the form:

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[50] It will be recalled that a similar representation was used much earlier in [F107]—vide Eq. 5.5.2.

[51] This equation describes bosons (mesons) of different spin, in contrast to the tetrad equations, which collectively describe spatio-temporal evolution of the 5 different tensor fields associated with the *same* Dirac particle.

$$\left[\frac{1}{2}\left(\Gamma'_{\mu}+\Gamma''_{\mu}\right)\partial_{\mu}+m\right]\psi=0 \tag{6.4.7}$$

For $m \neq 0$, this yields (Takahashi 1969), upon operating with $\frac{1}{2}(\Gamma'_{\mu}-\Gamma''_{\mu})$:

$$\frac{1}{2}\left(\Gamma'_{\mu}-\Gamma''_{\mu}\right)\partial_{\mu}\psi=0. \tag{6.4.8}$$

It will be noticed that the structure of Eqs. 6.4.7 and 6.4.8 is *identical* to that of the terms involving the *X*-space derivative, D_{μ}, in the above bilocal equations—i.e. the terms in the *DK* equation involving the local derivative ∂_{μ} become the *X*-space derivative terms in bilocal extension; there is, of course, no counterpart in the local *DK* equations of the terms involving the internal ξ-space derivative.

Kemmer also mentioned the combination $\frac{1}{2}(\Gamma'_{\mu}-\Gamma''_{\mu})$, which, he noted, had earlier been introduced by de Broglie, but which is *not* expressible in terms of the β_{μ} matrices alone. Here, however, the combination arises naturally, and enjoys the same status as $\frac{1}{2}(\Gamma'_{\mu}+\Gamma''_{\mu})$.

Fröhlich's bilocal extension based on the Dirac bilinear covariants can thus be considered to extend to space-time coordinates the 'distinction' between Γ'_{μ} and Γ''_{μ} (and indeed also between dashed and undashed operators that played such an important part in [F106–108]); this is realised by introducing distinct x' and x'', such that Γ'_{μ} becomes allied to the evolution of $U(x', x'')$ *wrt* to x', and Γ''_{μ} to the evolution *wrt* x''. The same holds at the level of the definition of the tetrad as a 4 × 4 array in terms of the commuting ρ and σ matrices given above, in the sense that ρ is allied to $\psi^{\dagger}$, whose coordinates are distinguished, as x'', from those, x', of ψ on which σ acts.

In an unpublished manuscript dating from 1986 (which is reproduced in Appendix 2 at the end of this chapter), 8 of the above 32 equations were presented. He considered it to be of particular significance that whilst the pseudo-vector spin current density, $S^{\mu}(\sim i\bar{\psi}\gamma^{5}\gamma^{\mu}\psi)$ does not satisfy an equation of continuity in local theory, it *does* so in the internal ξ-space of the admitted bilocal extension:

$$\Delta_{\mu}S^{\mu}=0, \quad \text{for all } \xi \tag{6.4.9}$$

This result is potentially relevant to establishing the physical (as opposed to formal) significance of ξ-space. To investigate this further, Fröhlich noted that S^{μ} transforms in the same way as does a magnetic current, in that it couples to the *dual* of an external electromagnetic field, but in such a way that the coupling vanishes in the local limit. Despite this, he was able to devise a novel experimental arrangement in which the vanishing energy current density of a Dirac particle in a direction orthogonal to its spin, which was taken to be itself orthogonal to an external oscillating electric field, was found to arise from an *exact* cancellation between local and non-local contributions—an insight that is, of course, contingent on the ab initio adoption of a non-local approach. More important, however, was considered to be the development of experimental methods to detect the *non*-local current, possibly utilising superconductors in which, as will be recalled from Sects. 5.4, 6.2, the supercurrent is itself based on a 2-point macroscopic wave-function, $\Phi_2\,(\boldsymbol{x}, \boldsymbol{y}; t)$.

Thus although based on standard Dirac theory, and as such contains nothing fundamentally new, the bilocal extension that lifts the $x' = x''$ degeneracy of conventional theory affords the possibility of new, hitherto unappreciated perspectives (such as that just noted for S^{μ}, for example), provided the associated ξ-space is taken seriously.[52] The closest connection with conventional local theory is, of course, via the 'external' X-coordinate, although it should be noted that, in bilocal generalisation, the continuity equation which holds locally for the electric current density, J^{μ}, namely $\partial_{\mu} J^{\mu} = 0$, becomes $D_{\mu}\, J^{\mu} = 0$, which reduces to $\partial^{X}_{\mu} J^{\mu} = 0$, *only* for $A^{\mu} = 0$.

Fröhlich spoke on these matters, and their extension to Duffin-Kemmer fields, at a symposium held at the University of Edinburgh in October 1979, to mark Kemmer's retirement, and 5 years later summarised some subsequent work in a letter to him dated 30 March 1984.

It was by considering the invariances of the 32 generalised Dirac-like equations (Eqs. 6.4.3, 6.4.4) under the extended group of transformations associated with the new internal ξ-space, together with the transformations involving interchanges the two sets of matrices, Γ'_{μ} and Γ''_{μ}, that Fröhlich hoped to obtain, through some kind of symmetery breaking, a deeper understanding of both the separation of Dirac particles into leptons and quarks, and also their mass spectra, *without* having to admit features that are alien to Minkowski space-time. One such transformation is $x' \Leftrightarrow x''$, under which X-space remains invariant, but ξ-space is fully reflected: $\xi^{\mu} \rightarrow -\xi^{\mu} : \boldsymbol{P}(\xi)\, \boldsymbol{T}(\xi)$.

But by now he was over 80 years old, and the task of editing [F(iv)] over the next 3 years absorbed most of his time and energy, so that very little of the above was ever published. There exist, however, copious manuscript notes containing attempts at further elaboration, which look highly promising, particularly concerning the separation of Dirac particles into leptons and quarks and their mass spectra, where 3 generations of lepton and quark masses could be accounted for.

Positive results in this direction would, of course, vindicate his earlier conviction that an understanding of mass necessitates an extension involving the introduction of a length, which would now be connected with an inherently *non*-local feature of space-time structure. The necessity of such an extension was already apparent from his earlier work on continuous reflections, wherein the form of the mass operator was *not* derivable in a compelling way.

The manuscripts referred to above are lodged in the Special Collections and Archives of the University of Liverpool (Reference code: GB 0141 D.56), *where they may be consulted.*

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[52] In the sense that the introduction of ξ-space is *not* to be regarded simply as a formal device to facilitate calculation, at the end of which the local limit is always taken, as is done in the case of the reduced density matrices, for example (vide Sect. 6.2).

Appendices

Fröhlich's 1948 Review of Von Laue's Theorie der Superleitung

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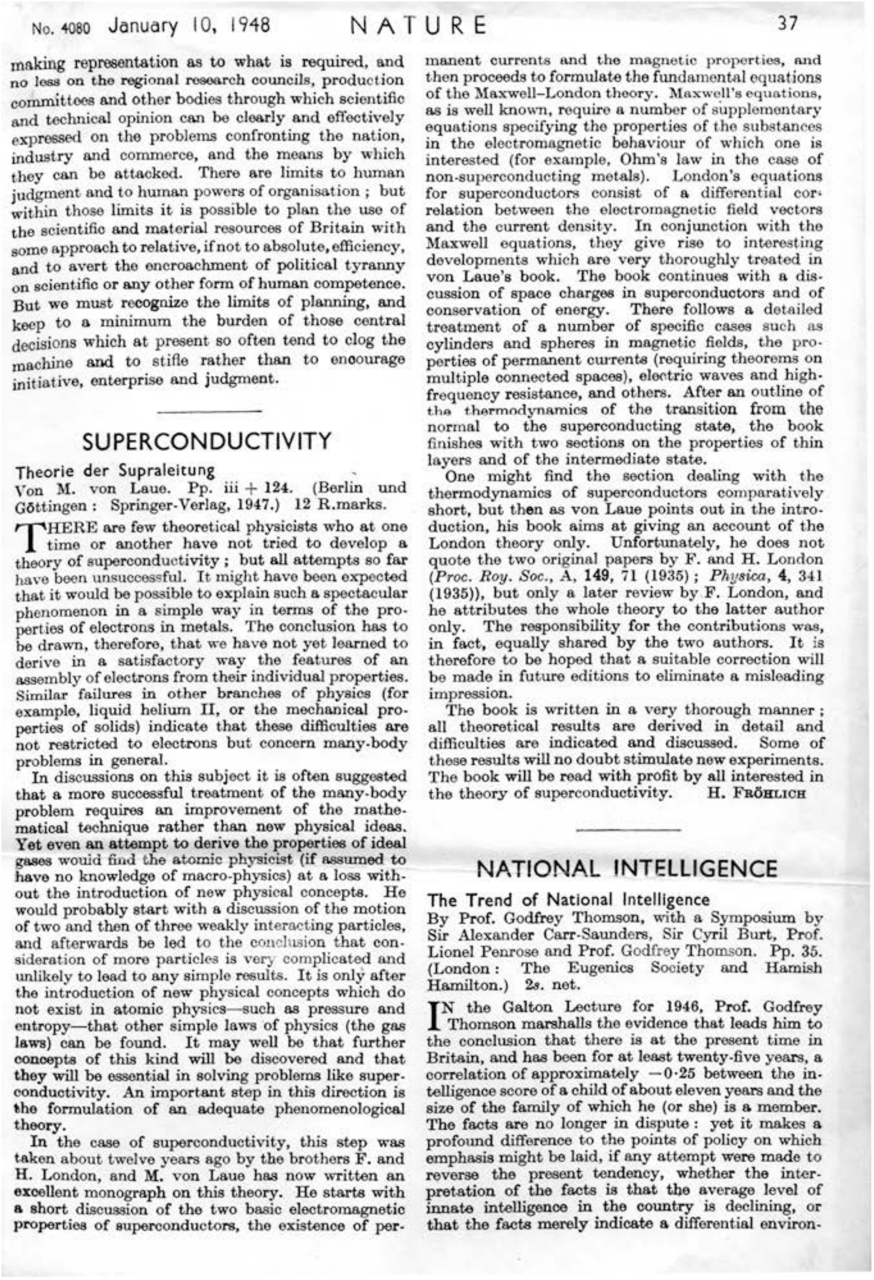

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making representation as to what is required, and no less on the regional research councils, production committees and other bodies through which scientific and technical opinion can be clearly and effectively expressed on the problems confronting the nation, industry and commerce, and the means by which they can be attacked. There are limits to human judgment and to human powers of organisation ; but within those limits it is possible to plan the use of the scientific and material resources of Britain with some approach to relative, if not to absolute, efficiency, and to avert the encroachment of political tyranny on scientific or any other form of human competence. But we must recognize the limits of planning, and keep to a minimum the burden of those central decisions which at present so often tend to clog the machine and to stifle rather than to encourage initiative, enterprise and judgment.

SUPERCONDUCTIVITY

Theorie der Supraleitung
Von M. von Laue. Pp. iii + 124. (Berlin und Göttingen : Springer-Verlag, 1947.) 12 R.marks.

THERE are few theoretical physicists who at one time or another have not tried to develop a theory of superconductivity ; but all attempts so far have been unsuccessful. It might have been expected that it would be possible to explain such a spectacular phenomenon in a simple way in terms of the properties of electrons in metals. The conclusion has to be drawn, therefore, that we have not yet learned to derive in a satisfactory way the features of an assembly of electrons from their individual properties. Similar failures in other branches of physics (for example, liquid helium II, or the mechanical properties of solids) indicate that these difficulties are not restricted to electrons but concern many-body problems in general.

In discussions on this subject it is often suggested that a more successful treatment of the many-body problem requires an improvement of the mathematical technique rather than new physical ideas. Yet even an attempt to derive the properties of ideal gases would find the atomic physicist (if assumed to have no knowledge of macro-physics) at a loss without the introduction of new physical concepts. He would probably start with a discussion of the motion of two and then of three weakly interacting particles, and afterwards be led to the conclusion that consideration of more particles is very complicated and unlikely to lead to any simple results. It is only after the introduction of new physical concepts which do not exist in atomic physics—such as pressure and entropy—that other simple laws of physics (the gas laws) can be found. It may well be that further concepts of this kind will be discovered and that they will be essential in solving problems like superconductivity. An important step in this direction is the formulation of an adequate phenomenological theory.

In the case of superconductivity, this step was taken about twelve years ago by the brothers F. and H. London, and M. von Laue has now written an excellent monograph on this theory. He starts with a short discussion of the two basic electromagnetic properties of superconductors, the existence of permanent currents and the magnetic properties, and then proceeds to formulate the fundamental equations of the Maxwell–London theory. Maxwell's equations, as is well known, require a number of supplementary equations specifying the properties of the substances in the electromagnetic behaviour of which one is interested (for example, Ohm's law in the case of non-superconducting metals). London's equations for superconductors consist of a differential correlation between the electromagnetic field vectors and the current density. In conjunction with the Maxwell equations, they give rise to interesting developments which are very thoroughly treated in von Laue's book. The book continues with a discussion of space charges in superconductors and of conservation of energy. There follows a detailed treatment of a number of specific cases such as cylinders and spheres in magnetic fields, the properties of permanent currents (requiring theorems on multiple connected spaces), electric waves and high-frequency resistance, and others. After an outline of the thermodynamics of the transition from the normal to the superconducting state, the book finishes with two sections on the properties of thin layers and of the intermediate state.

One might find the section dealing with the thermodynamics of superconductors comparatively short, but then as von Laue points out in the introduction, his book aims at giving an account of the London theory only. Unfortunately, he does not quote the two original papers by F. and H. London (*Proc. Roy. Soc.*, A, **149**, 71 (1935) ; *Physica*, **4**, 341 (1935)), but only a later review by F. London, and he attributes the whole theory to the latter author only. The responsibility for the contributions was, in fact, equally shared by the two authors. It is therefore to be hoped that a suitable correction will be made in future editions to eliminate a misleading impression.

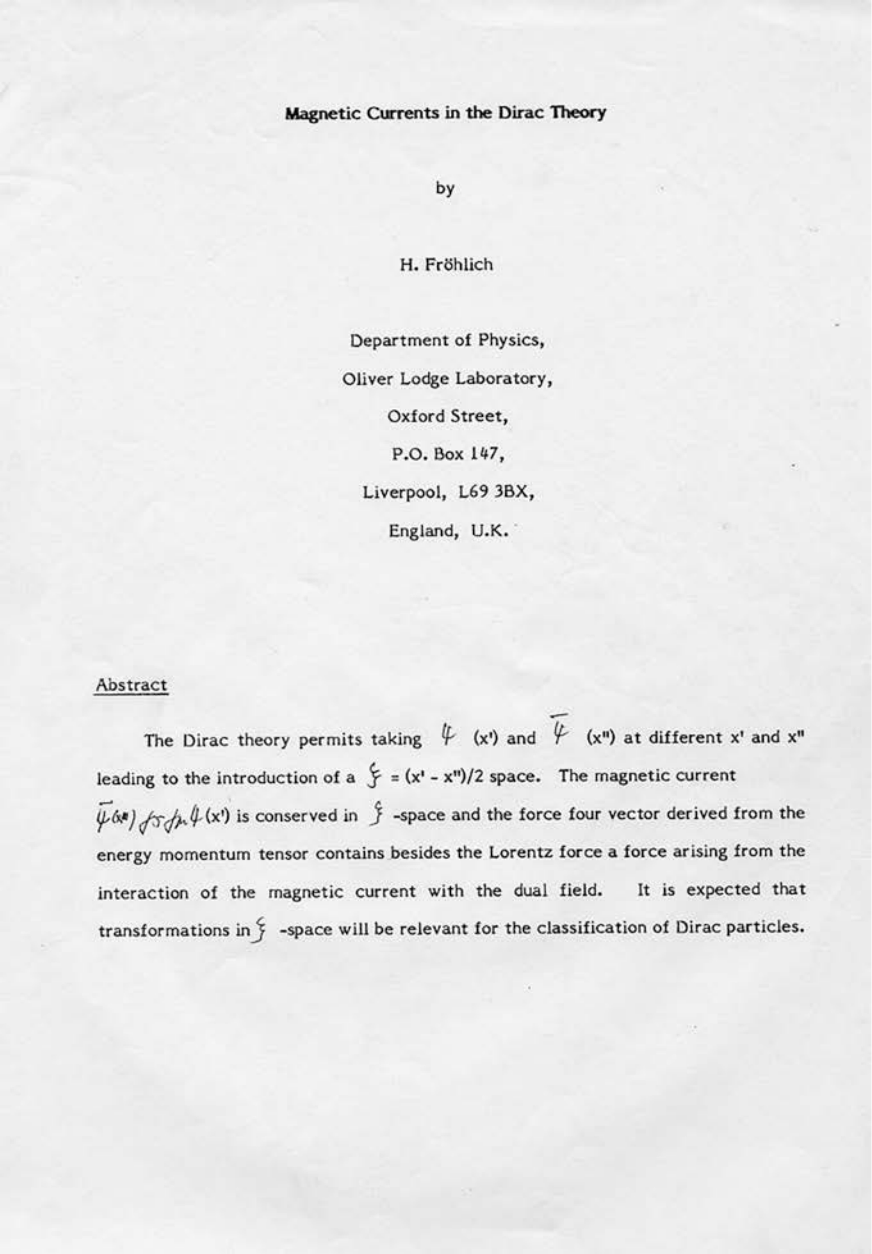
The book is written in a very thorough manner ; all theoretical results are derived in detail and difficulties are indicated and discussed. Some of these results will no doubt stimulate new experiments. The book will be read with profit by all interested in the theory of superconductivity. H. FRÖHLICH

NATIONAL INTELLIGENCE

The Trend of National Intelligence
By Prof. Godfrey Thomson, with a Symposium by Sir Alexander Carr-Saunders, Sir Cyril Burt, Prof. Lionel Penrose and Prof. Godfrey Thomson. Pp. 35. (London : The Eugenics Society and Hamish Hamilton.) 2*s*. net.

IN the Galton Lecture for 1946, Prof. Godfrey Thomson marshalls the evidence that leads him to the conclusion that there is at the present time in Britain, and has been for at least twenty-five years, a correlation of approximately −0·25 between the intelligence score of a child of about eleven years and the size of the family of which he (or she) is a member. The facts are no longer in dispute : yet it makes a profound difference to the points of policy on which emphasis might be laid, if any attempt were made to reverse the present tendency, whether the interpretation of the facts is that the average level of innate intelligence in the country is declining, or that the facts merely indicate a differential environ-

An Unpublished Manuscript of 1986 on a Bilocal Extension of the Dirac Bilinear Covariants



Magnetic Currents in the Dirac Theory

by

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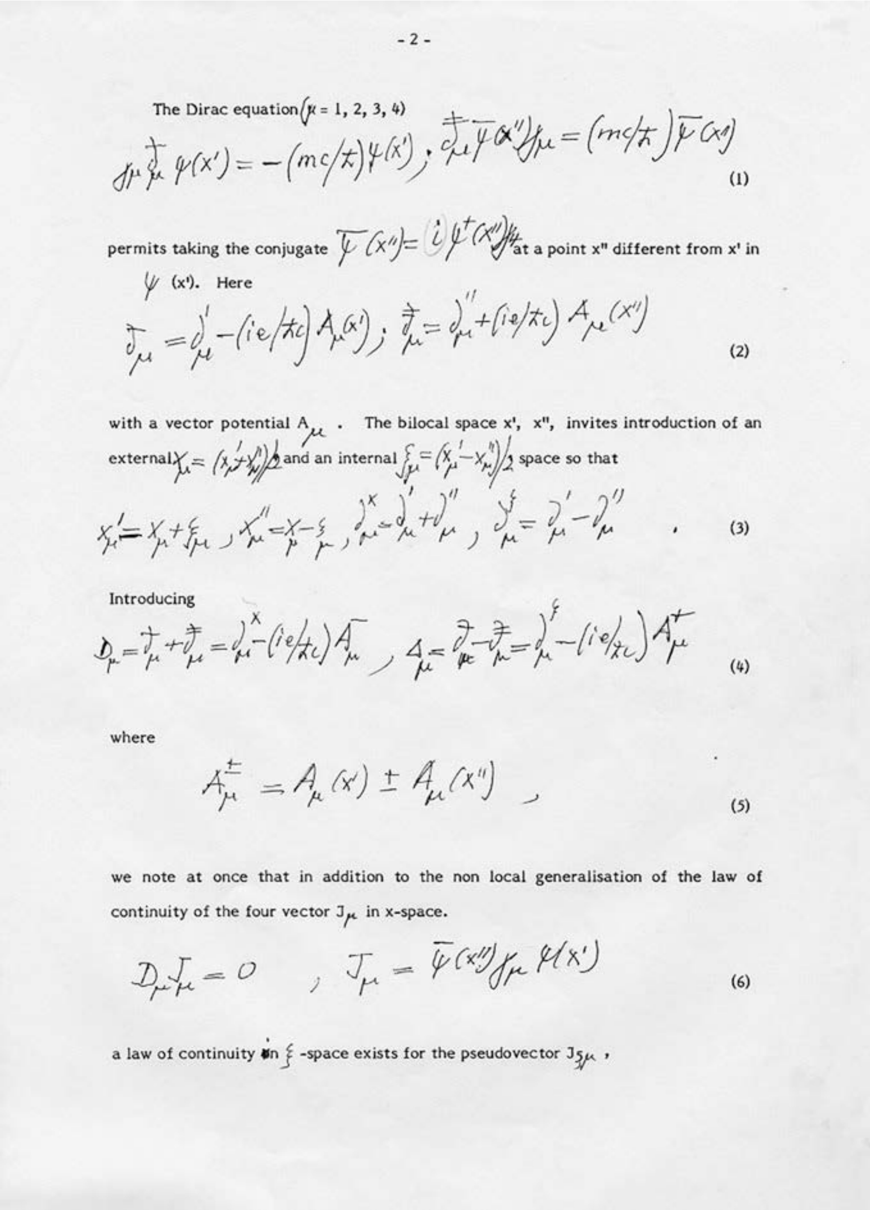
Abstract

The Dirac theory permits taking ψ (x') and $\bar{\psi}$ (x") at different x' and x" leading to the introduction of a $\xi = (x' - x'')/2$ space. The magnetic current $\bar{\psi}(x'')\gamma_5\gamma_\mu\psi(x')$ is conserved in ξ -space and the force four vector derived from the energy momentum tensor contains besides the Lorentz force a force arising from the interaction of the magnetic current with the dual field. It is expected that transformations in ξ -space will be relevant for the classification of Dirac particles.



The Dirac equation $(\mu = 1, 2, 3, 4)$

$$\gamma_\mu \vec{\partial}_\mu \psi(x') = -(mc/\hbar)\psi(x')\,;\quad \overleftarrow{\partial}_\mu \bar{\psi}(x'')\gamma_\mu = (mc/\hbar)\bar{\psi}(x'') \tag{1}$$

permits taking the conjugate $\bar{\psi}(x'') = i\psi^{+}(x'')\gamma_4$ at a point x" different from x' in ψ (x'). Here

$$\vec{\partial}_\mu = \partial'_\mu - (ie/\hbar c)A_\mu(x')\,;\quad \overleftarrow{\partial}_\mu = \partial''_\mu + (ie/\hbar c)A_\mu(x'') \tag{2}$$

with a vector potential A_μ . The bilocal space x', x", invites introduction of an external $X_\mu = (x'_\mu + x''_\mu)/2$ and an internal $\xi_\mu = (x'_\mu - x''_\mu)/2$ space so that

$$x'_\mu = X_\mu + \xi_\mu\,,\quad x''_\mu = X_\mu - \xi_\mu\,,\quad \partial^X_\mu = \partial'_\mu + \partial''_\mu\,,\quad \partial^\xi_\mu = \partial'_\mu - \partial''_\mu \quad . \tag{3}$$

Introducing

$$D_\mu = \vec{\partial}_\mu + \overleftarrow{\partial}_\mu = \partial^X_\mu - (ie/\hbar c)A^-_\mu\,,\quad \Delta_\mu = \vec{\partial}_\mu - \overleftarrow{\partial}_\mu = \partial^\xi_\mu - (ie/\hbar c)A^+_\mu \tag{4}$$

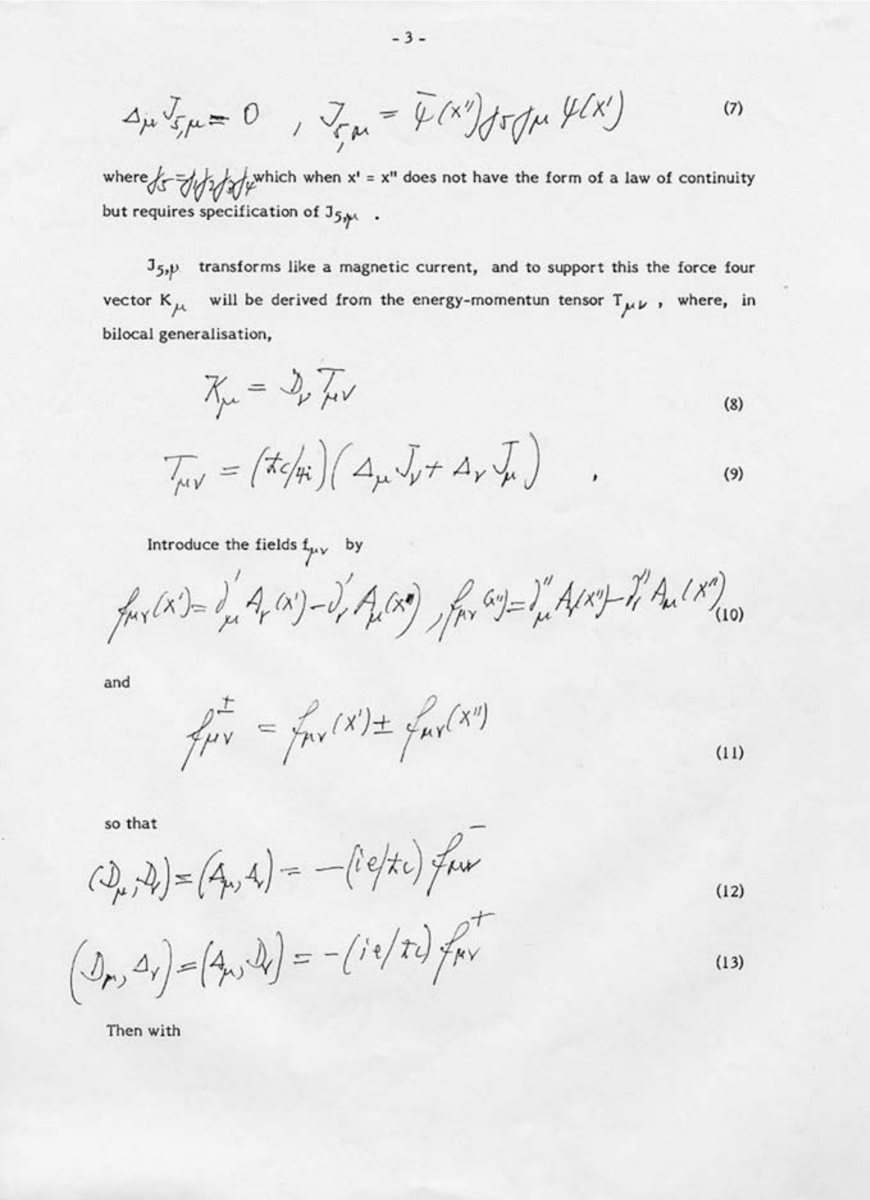
where

$$A^\pm_\mu = A_\mu(x') \pm A_\mu(x'')\,, \tag{5}$$

we note at once that in addition to the non local generalisation of the law of continuity of the four vector J_μ in x-space.

$$D_\mu J_\mu = 0\,,\quad J_\mu = \bar{\psi}(x'')\gamma_\mu \psi(x') \tag{6}$$

a law of continuity in ξ -space exists for the pseudovector $J_{5\mu}$,



- 3 -

$$\Delta_\mu J_{5,\mu} = 0 \quad , \quad J_{5,\mu} = \bar{\psi}(x'')\gamma_5\gamma_\mu\psi(x') \tag{7}$$

where $\gamma_5 = \gamma_1\gamma_2\gamma_3\gamma_4$ which when x' = x" does not have the form of a law of continuity but requires specification of $J_{5,\mu}$.

$J_{5,\mu}$ transforms like a magnetic current, and to support this the force four vector K_μ will be derived from the energy-momentun tensor $T_{\mu\nu}$, where, in bilocal generalisation,

$$K_\mu = D_\nu T_{\mu\nu} \tag{8}$$

$$T_{\mu\nu} = (\hbar c/4i)(\Delta_\mu J_\nu + \Delta_\nu J_\mu) \quad , \tag{9}$$

Introduce the fields $f_{\mu\nu}$ by

$$f_{\mu\nu}(x') = \partial'_\mu A_\nu(x') - \partial'_\nu A_\mu(x') \quad , \quad f_{\mu\nu}(x'') = \partial''_\mu A_\nu(x'') - \partial''_\nu A_\mu(x'') \tag{10}$$

and

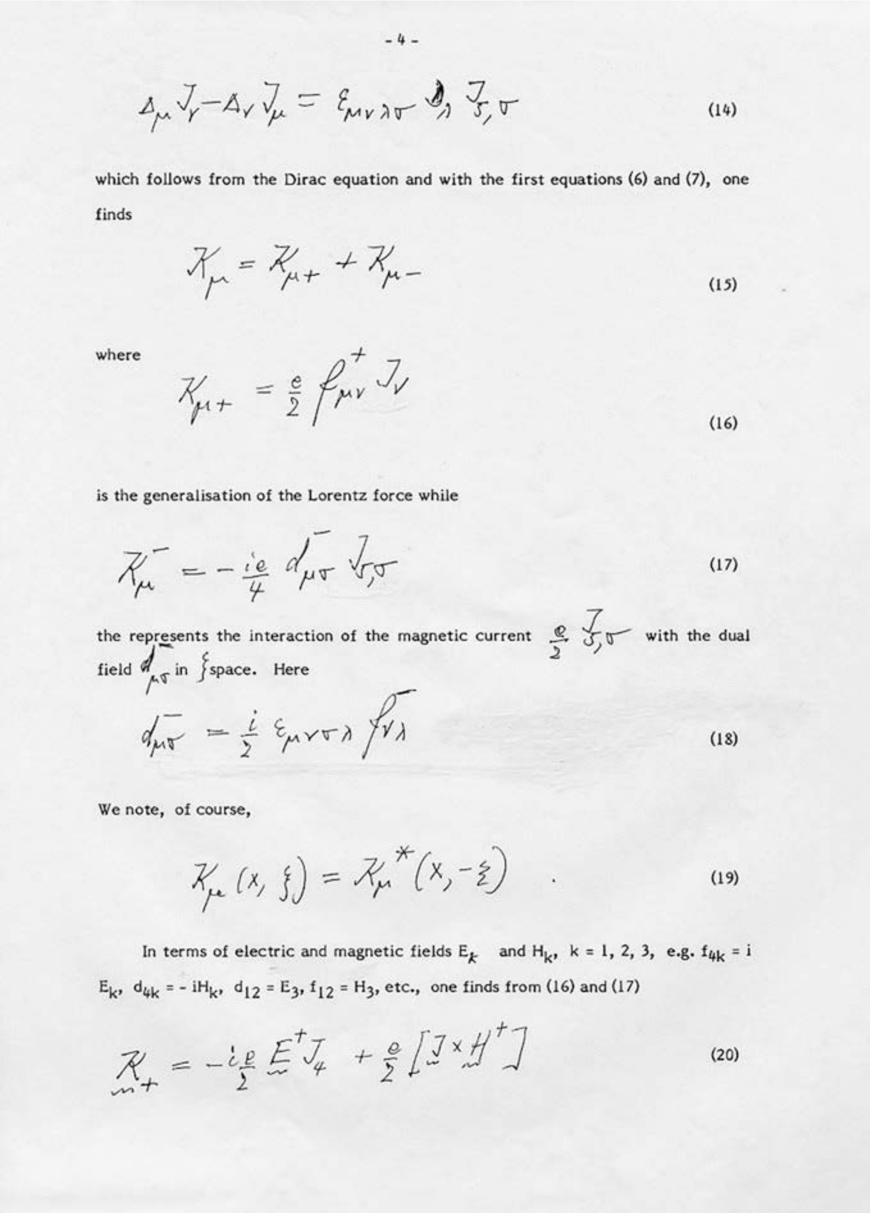
$$f^{\pm}_{\mu\nu} = f_{\mu\nu}(x') \pm f_{\mu\nu}(x'') \tag{11}$$

so that

$$(D_\mu, D_\nu) = (\Delta_\mu, \Delta_\nu) = -(ie/\hbar c) f^{-}_{\mu\nu} \tag{12}$$

$$(D_\mu, \Delta_\nu) = (\Delta_\mu, D_\nu) = -(ie/\hbar c) f^{+}_{\mu\nu} \tag{13}$$

Then with



$$\Delta_\mu J_\nu - \Delta_\nu J_\mu = \varepsilon_{\mu\nu\lambda\sigma} \partial_\lambda J_{5,\sigma} \tag{14}$$

which follows from the Dirac equation and with the first equations (6) and (7), one finds

$$K_\mu = K_{\mu+} + K_{\mu-} \tag{15}$$

where

$$K_{\mu+} = \frac{e}{2} f^+_{\mu\nu} J_\nu \tag{16}$$

is the generalisation of the Lorentz force while

$$K^-_\mu = -\frac{ie}{4} d^-_{\mu\sigma} J_{5,\sigma} \tag{17}$$

the represents the interaction of the magnetic current $\frac{e}{2} J_{5,\sigma}$ with the dual field $d^-_{\mu\sigma}$ in ξspace. Here

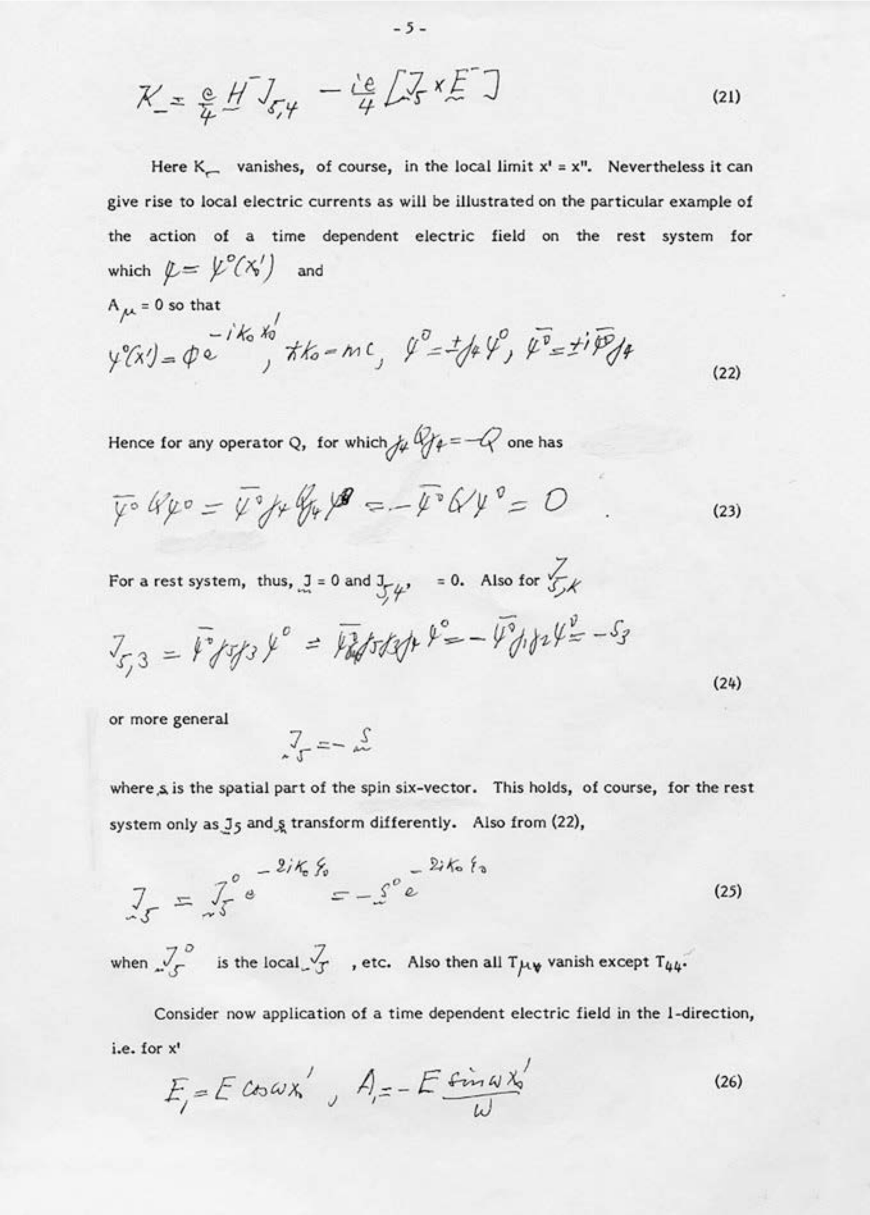
$$d^-_{\mu\sigma} = \frac{i}{2} \varepsilon_{\mu\nu\sigma\lambda} f^-_{\nu\lambda} \tag{18}$$

We note, of course,

$$K_\mu(x, \xi) = K_\mu^*(x, -\xi) \quad . \tag{19}$$

In terms of electric and magnetic fields E_k and H_k, $k = 1, 2, 3$, e.g. $f_{4k} = i E_k$, $d_{4k} = - iH_k$, $d_{12} = E_3$, $f_{12} = H_3$, etc., one finds from (16) and (17)

$$\underset{\sim}{K}_+ = -\frac{ie}{2} \underset{\sim}{E}^+ J_4 + \frac{e}{2}\left[\underset{\sim}{J} \times \underset{\sim}{H}^+\right] \tag{20}$$



$$K_{-} = \frac{e}{4}\underset{\sim}{H}^{-} J_{5,4} - \frac{ie}{4}[\underset{\sim}{J}_5 \times \underset{\sim}{E}^{-}] \tag{21}$$

Here K_{-} vanishes, of course, in the local limit x' = x". Nevertheless it can give rise to local electric currents as will be illustrated on the particular example of the action of a time dependent electric field on the rest system for which $\psi = \psi^0(x_0')$ and

$A_\mu = 0$ so that

$$\psi^0(x') = \phi e^{-ik_0x_0'}, \quad \hbar k_0 = mc, \quad \psi^0 = \pm\gamma_4\psi^0, \quad \bar{\psi}^0 = \pm i\bar{\psi}^0\gamma_4 \tag{22}$$

Hence for any operator Q, for which $\gamma_4 Q\gamma_4 = -Q$ one has

$$\bar{\psi}^0 Q\psi^0 = \bar{\psi}^0\gamma_4 Q\gamma_4\psi^0 = -\bar{\psi}^0 Q\psi^0 = 0 \quad . \tag{23}$$

For a rest system, thus, $\underset{\sim}{J} = 0$ and $J_{5,4}$, = 0. Also for $J_{5,k}$

$$J_{5,3} = \bar{\psi}^0\gamma_5\gamma_3\psi^0 = \bar{\psi}^0\gamma_4\gamma_5\gamma_3\gamma_4\psi^0 = -\bar{\psi}^0\gamma_1\gamma_2\psi^0 = -S_3 \tag{24}$$

or more general

$$\underset{\sim}{J}_5 = -\underset{\sim}{S}$$

where $\underset{\sim}{S}$ is the spatial part of the spin six-vector. This holds, of course, for the rest system only as $\underset{\sim}{J}_5$ and $\underset{\sim}{S}$ transform differently. Also from (22),

$$\underset{\sim}{J}_5 = \underset{\sim}{J}_5^0 e^{-2ik_0\xi_0} = -\underset{\sim}{S}^0 e^{-2ik_0\xi_0} \tag{25}$$

when $\underset{\sim}{J}_5^0$ is the local $\underset{\sim}{J}_5$, etc. Also then all $T_{\mu\nu}$ vanish except T_{44}.

Consider now application of a time dependent electric field in the 1-direction, i.e. for x'

$$E_1 = E\cos\omega x_0', \quad A_1 = -E\frac{\sin\omega x_0'}{\omega} \tag{26}$$

and similar for x'' so that

$$E_1^- = -2E \sin \omega x_0 \sin \omega \xi_0 \tag{27}$$

This will be treated as a first order in E perturbation.

Hence

$$\mathcal{K}_{-2} = -i\frac{e}{4}\left[\underset{\sim}{J}_5 \times \underset{\sim}{E}^-\right]_2 = -\frac{ie}{4} J_{5,3} E_1^- = \frac{ie}{2} E J_{5,3}^0 e^{-2ik_0\xi_0} \sin \omega x_0 \sin \omega \xi_0 \, . \tag{28}$$

In the required order, as there is no x_x', x_x'' dependence, on has

$$D_1 = -\frac{ie}{\hbar c} A_1^- = \frac{ieE}{\hbar c} \frac{2\cos \omega x_0}{\omega} \sin \omega \xi_0 \, , \tag{29}$$

$$\Delta_1 = -\frac{ie}{\hbar c} A_1^+ = \frac{ieE}{\hbar c} \frac{2\sin \omega x_0}{\omega} \cos \omega \xi_0 \, , \tag{30}$$

$$D_4 = \frac{1}{i}\partial^x \, , \quad \Delta_4 = \frac{1}{i}\partial^\xi \tag{31}$$

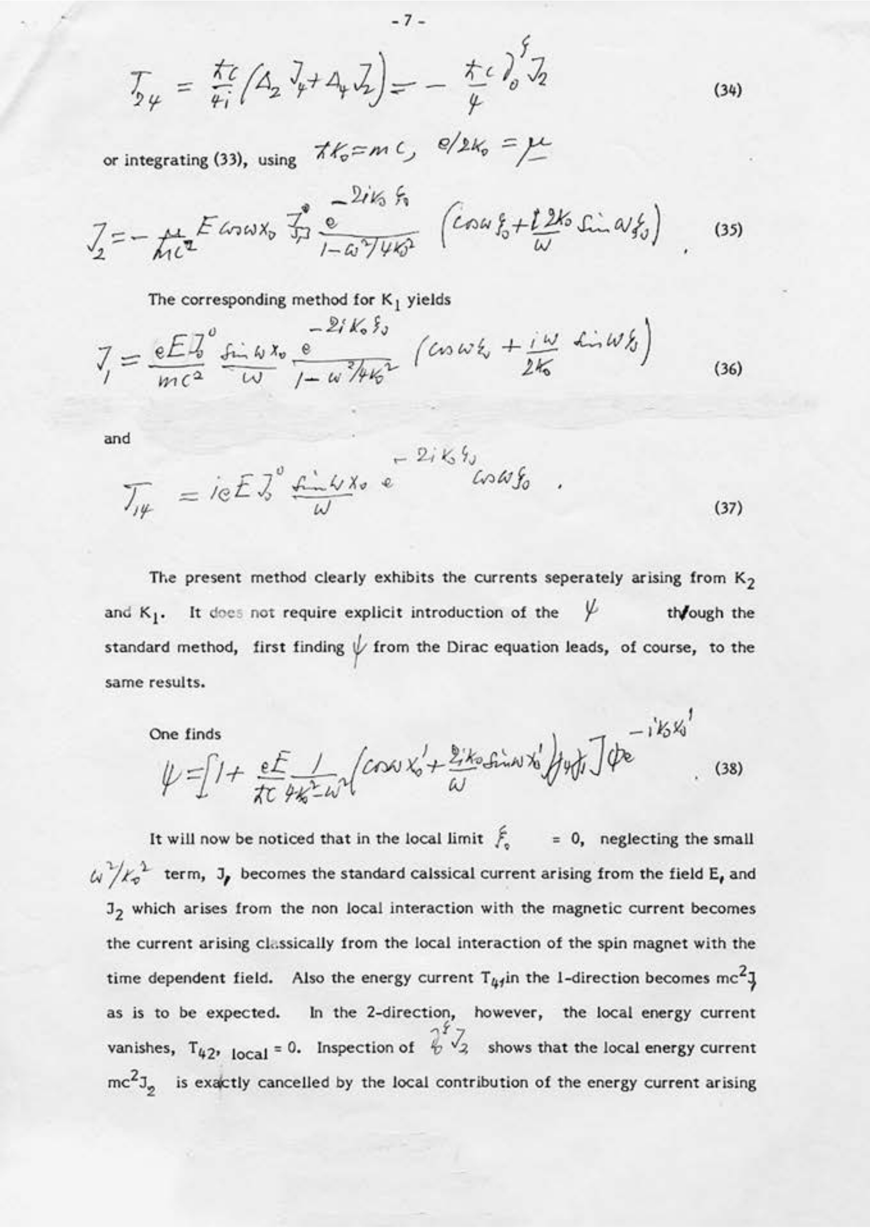
and all other Δ and D operators vanish. Hence as $D_1 T_{2,}$, is negligible (8) yields

$$\mathcal{K}_{,2} = \frac{1}{i}\partial_0 T_{24} \tag{32}$$

or integrating (28),

$$T_{24} = \frac{e}{2} E J_{5,3}^0 \frac{\cos \omega x_0}{\omega} e^{-2ik_0\xi_0} \sin \omega \xi_0 \, , \tag{33}$$

Further more from (9) using (31)



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$$T_{24} = \frac{\hbar c}{4i}\left(\Delta_2 J_4 + \Delta_4 J_2\right) = -\frac{\hbar c}{4}\partial_0^{\xi} J_2 \tag{34}$$

or integrating (33), using $\hbar k_0 = mc$, $e/2k_0 = \mu$

$$J_2 = -\frac{\mu}{\hbar c^2} E \cos\omega x_0 \, J_0^0 \, \frac{e^{-2ik_0\xi_0}}{1-\omega^2/4k_0^2}\left(\cos\omega\xi_0 + \frac{i2k_0}{\omega}\sin\omega\xi_0\right) . \tag{35}$$

The corresponding method for K_1 yields

$$J_1 = \frac{eEJ_0^0}{mc^2}\frac{\sin\omega x_0}{\omega}\frac{e^{-2ik_0\xi_0}}{1-\omega^2/4k_0^2}\left(\cos\omega\xi_0 + \frac{i\omega}{2k_0}\sin\omega\xi_0\right) \tag{36}$$

and

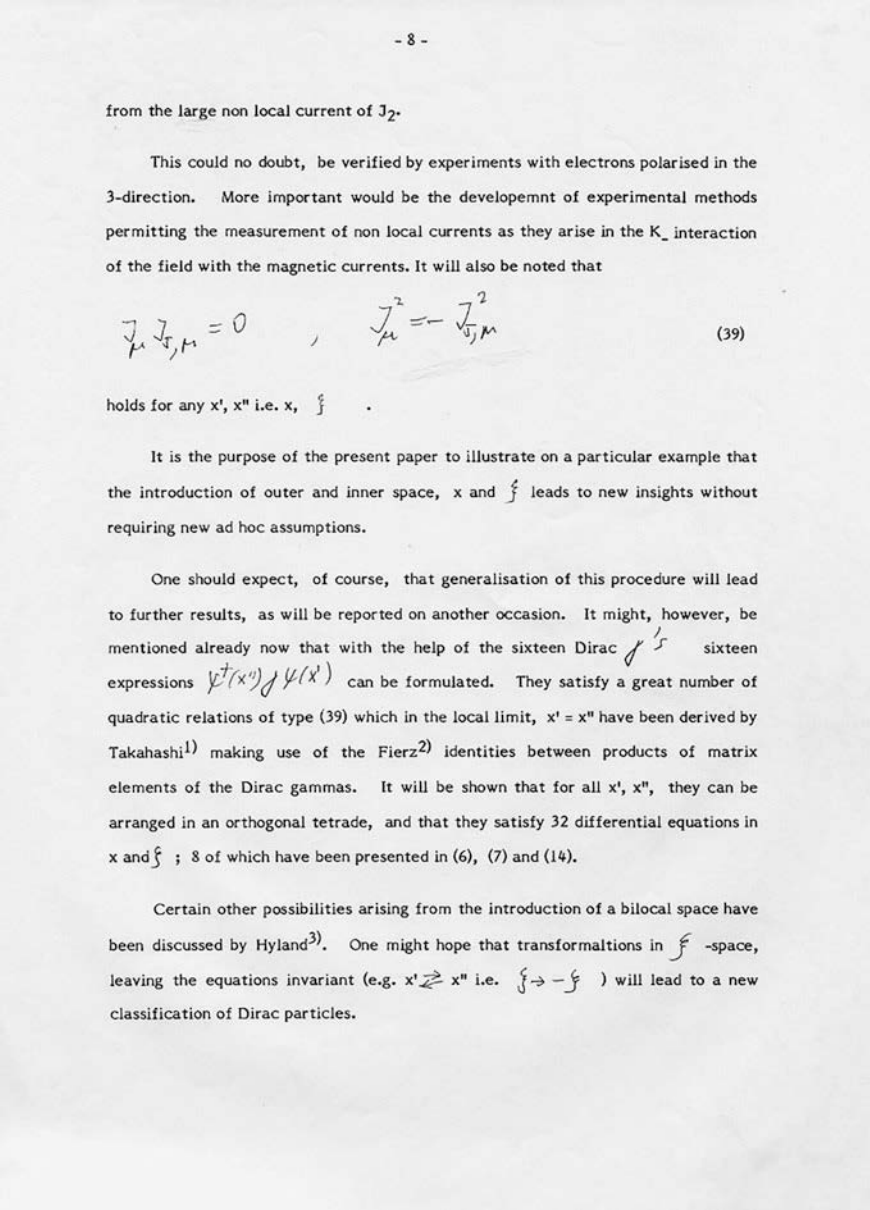
$$T_{14} = ieEJ_0^0\frac{\sin\omega x_0}{\omega} e^{-2ik_0\xi_0}\cos\omega\xi_0 . \tag{37}$$

The present method clearly exhibits the currents seperately arising from K_2 and K_1. It does not require explicit introduction of the ψ through the standard method, first finding ψ from the Dirac equation leads, of course, to the same results.

One finds

$$\psi = \left[1 + \frac{eE}{\hbar c}\frac{1}{4k_0^2-\omega^2}\left(\cos\omega x_0' + \frac{2ik_0}{\omega}\sin\omega x_0'\right)\gamma_4\gamma_1\right]\phi e^{-ik_0x_0'} . \tag{38}$$

It will now be noticed that in the local limit $\xi_0 = 0$, neglecting the small ω^2/k_0^2 term, J_1 becomes the standard calssical current arising from the field E, and J_2 which arises from the non local interaction with the magnetic current becomes the current arising classically from the local interaction of the spin magnet with the time dependent field. Also the energy current T_{41} in the 1-direction becomes mc^2J_1 as is to be expected. In the 2-direction, however, the local energy current vanishes, $T_{42,\,\text{local}} = 0$. Inspection of $\partial_0^{\xi} J_2$ shows that the local energy current mc^2J_2 is exactly cancelled by the local contribution of the energy current arising



from the large non local current of J_2.

This could no doubt, be verified by experiments with electrons polarised in the 3-direction. More important would be the developemnt of experimental methods permitting the measurement of non local currents as they arise in the K_- interaction of the field with the magnetic currents. It will also be noted that

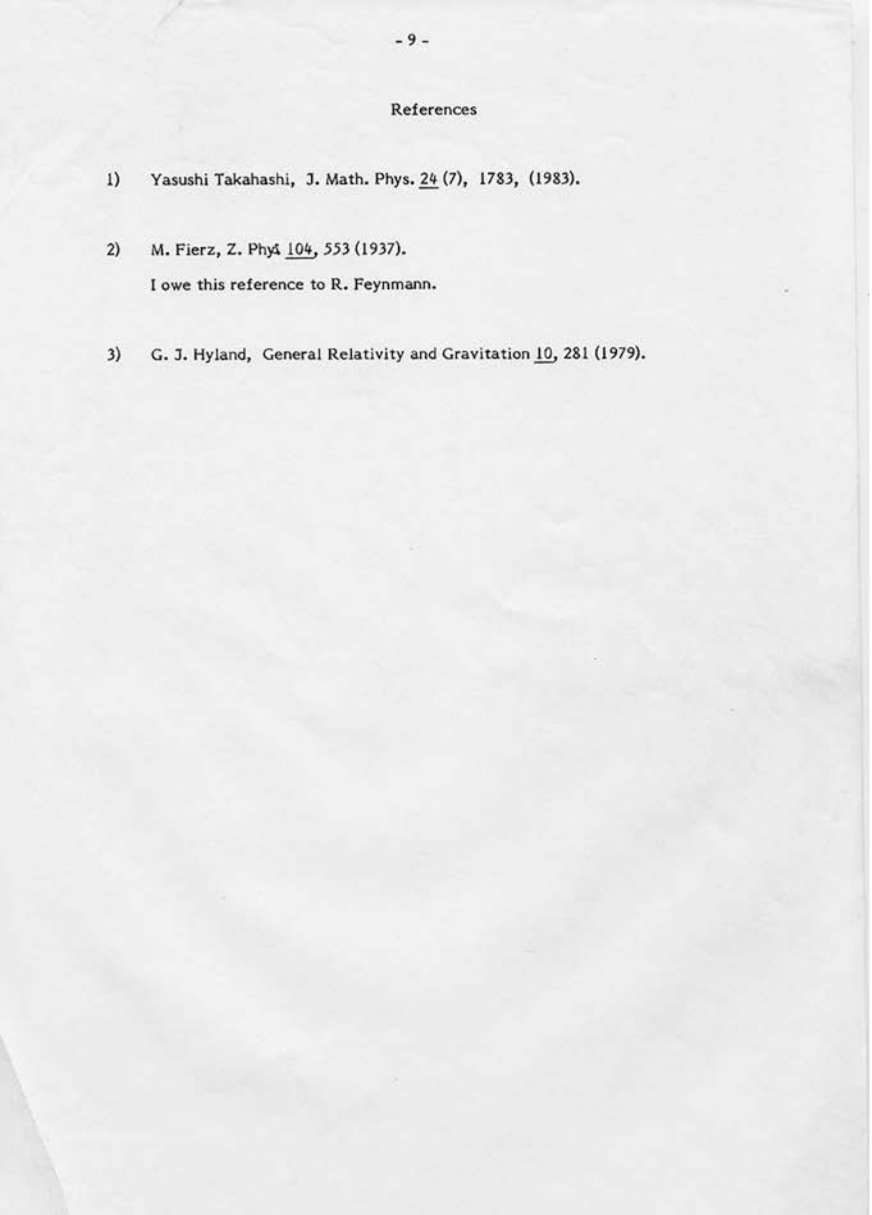
$$J_\mu J_{5,\mu} = 0 \quad , \quad J_\mu^2 = - J_{5,\mu}^2 \tag{39}$$

holds for any x', x" i.e. x, ξ .

It is the purpose of the present paper to illustrate on a particular example that the introduction of outer and inner space, x and ξ leads to new insights without requiring new ad hoc assumptions.

One should expect, of course, that generalisation of this procedure will lead to further results, as will be reported on another occasion. It might, however, be mentioned already now that with the help of the sixteen Dirac γ's sixteen expressions $\psi^\dagger(x'')\gamma\psi(x')$ can be formulated. They satisfy a great number of quadratic relations of type (39) which in the local limit, x' = x" have been derived by Takahashi[1] making use of the Fierz[2] identities between products of matrix elements of the Dirac gammas. It will be shown that for all x', x", they can be arranged in an orthogonal tetrade, and that they satisfy 32 differential equations in x and ξ ; 8 of which have been presented in (6), (7) and (14).

Certain other possibilities arising from the introduction of a bilocal space have been discussed by Hyland[3]. One might hope that transformaltions in ξ -space, leaving the equations invariant (e.g. $x' \rightleftarrows x''$ i.e. $\xi \rightarrow -\xi$) will lead to a new classification of Dirac particles.



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1) Yasushi Takahashi, J. Math. Phys. 24 (7), 1783, (1983).

2) M. Fierz, Z. Phys. 104, 553 (1937).
I owe this reference to R. Feynmann.

3) G. J. Hyland, General Relativity and Gravitation 10, 281 (1979).

Photo Gallery

The numbers in round brackets at the end of each caption correspond to the section of the text to which the photographs refer.

All photographs are from private collections.

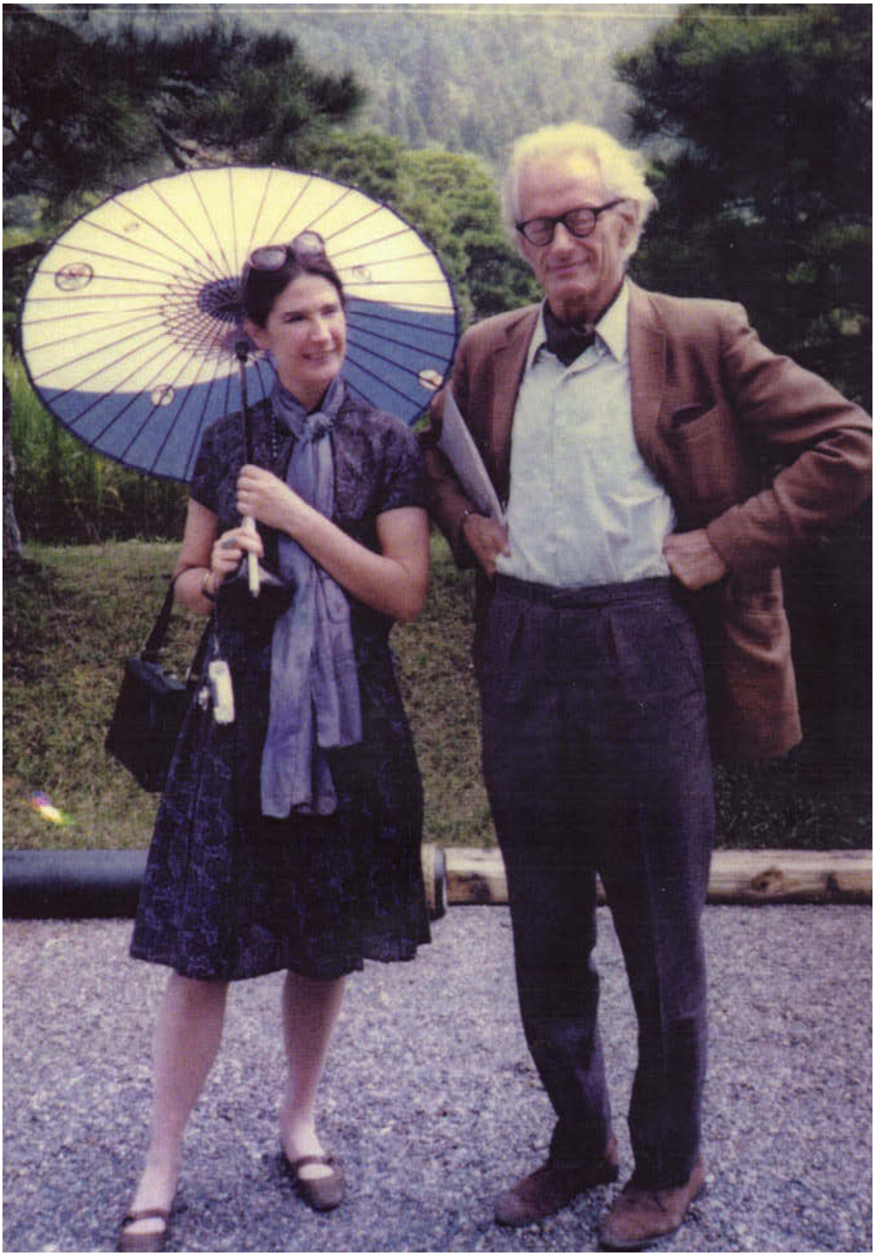

Fig. 6.8 Fröhlich and his wife in Kyoto, Japan, 1968, for the International Conference on Statistical Mechanics at which he presented [F139] (Sect. 6.2)

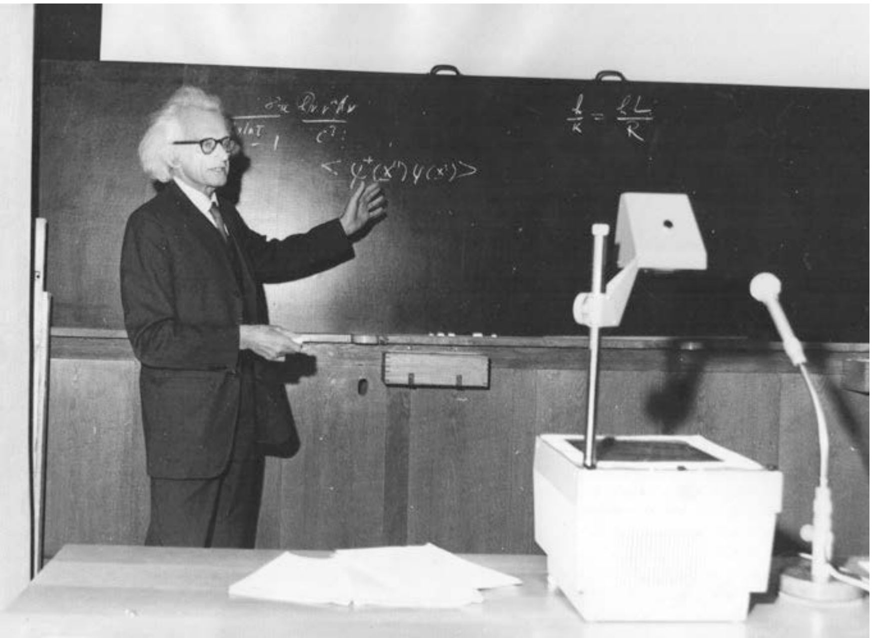

Fig. 6.9 Fröhlich indicating the 1st reduced density matrix during a lecture on the connection between micro and macrophysics at the Dublin Institute for Advanced Studies, 1969, (Sect. 6.2)

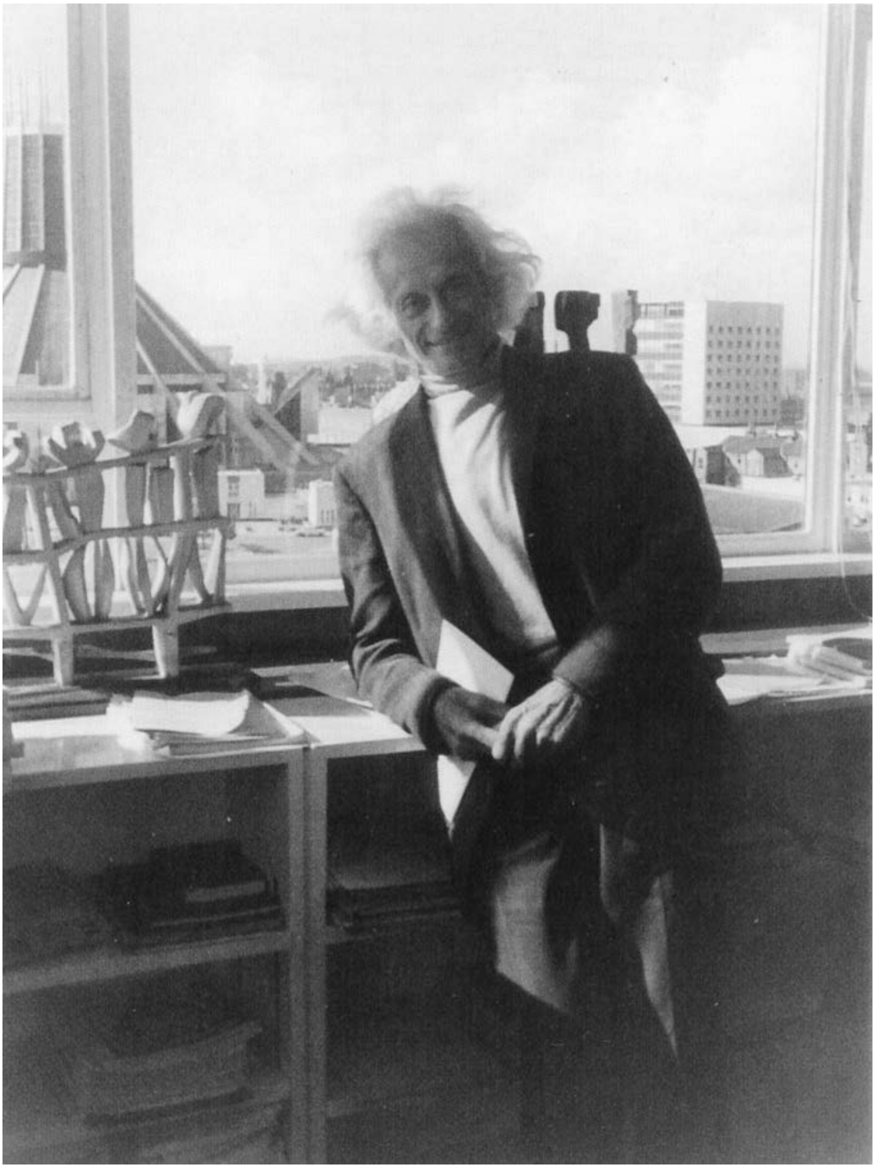

Fig. 6.10 Fröhlich in his office just before retiring from the Liverpool Chair in 1973 (Sect. 6.2)

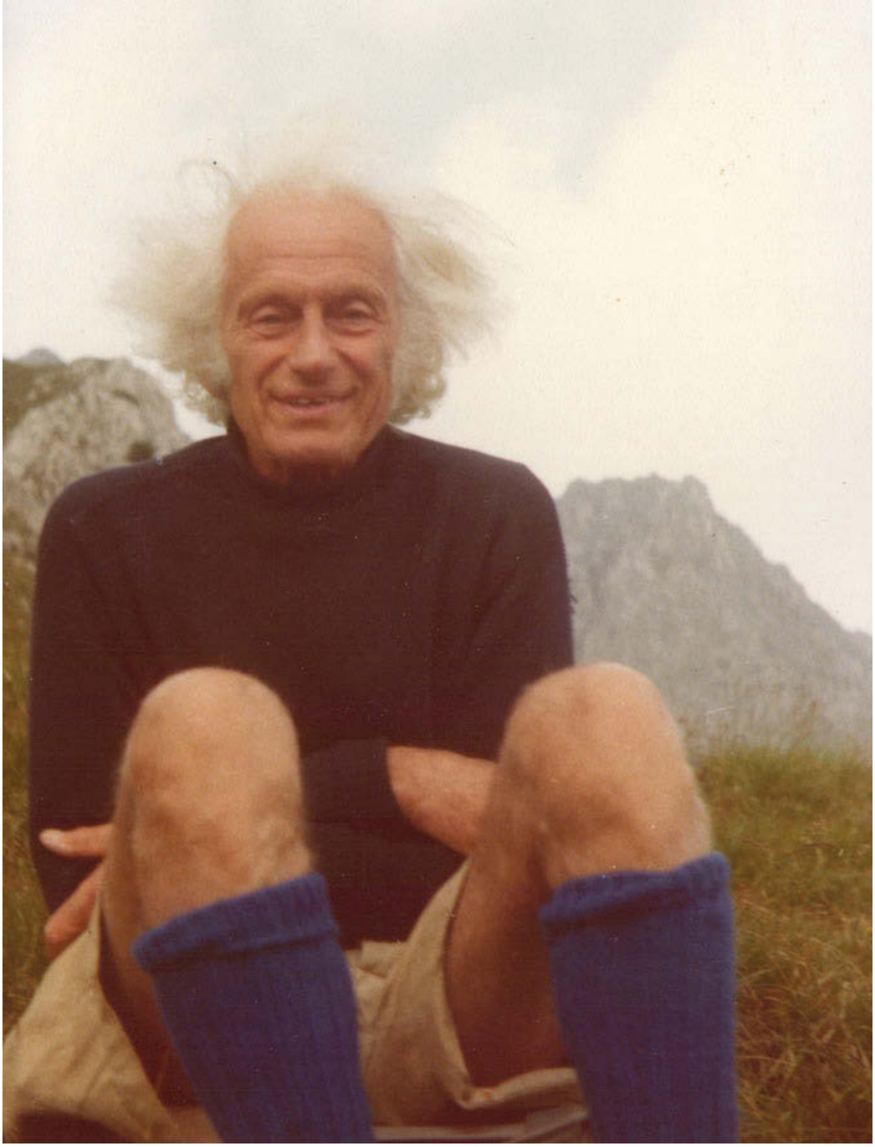

Fig. 6.11 Resting after a climb above Innsbruck (Sect. 6.3)

Fig. 6.12 At the 5th *l'Institut de la Vie* International Conference on Theoretical Physics and Biology, Vienna, 1975; also shown is Rudolph Peierls (Sect. 6.3)

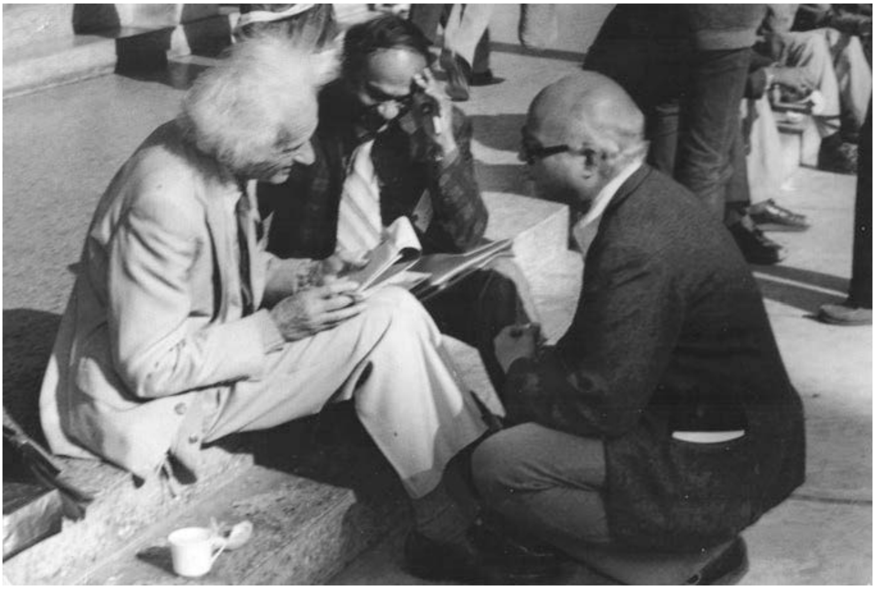

Fig. 6.13 In New Delhi, India, at the 1st International Seminar on the Living State, 1981 (Sect. 6.3)

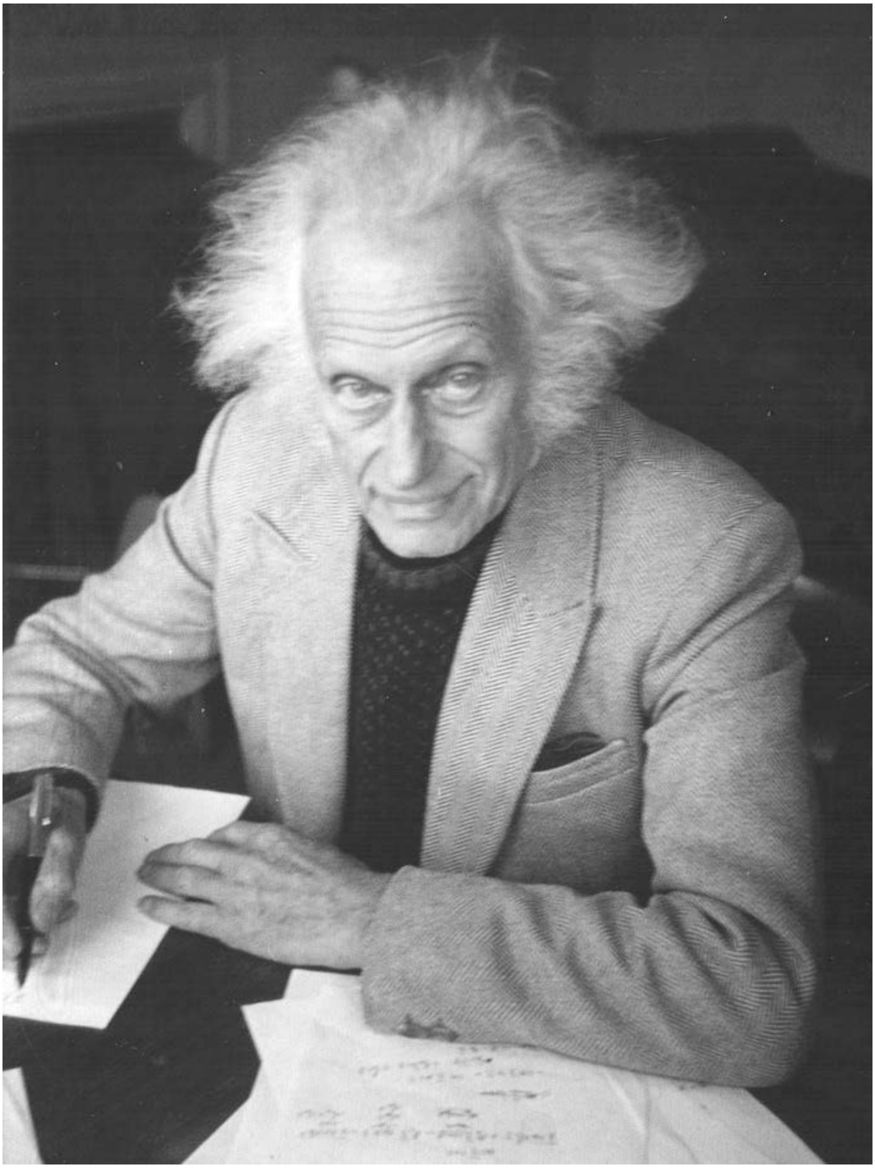

Fig. 6.14 Working on the tetrad at his desk at home, *c*.1985 (Sect. 6.4)

Fig. 6.15 Creative hands, *c*.1985 (Sect. 6.4)

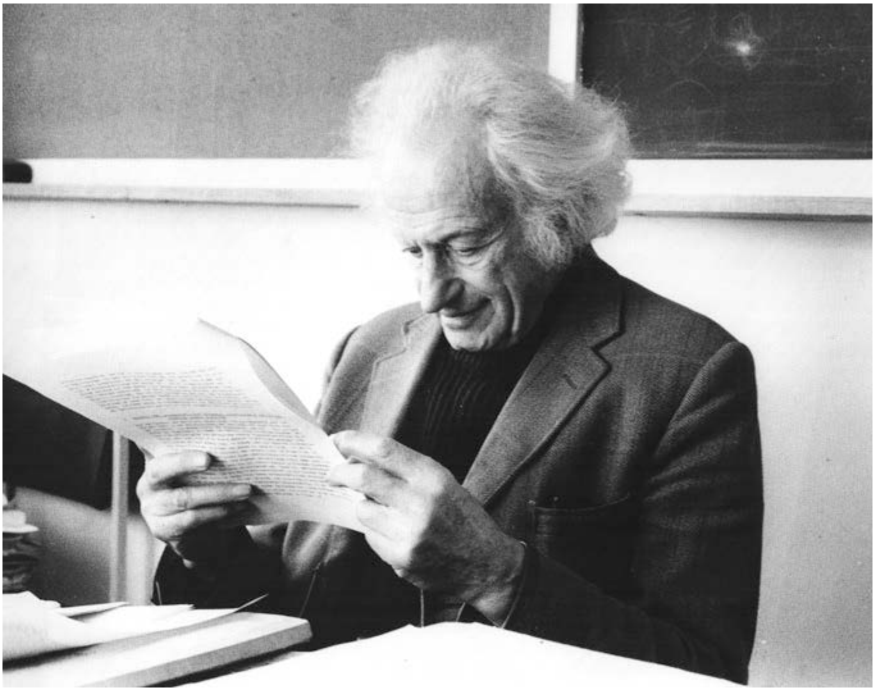

Fig. 6.16 Reading in his office, 1987 (Sect. 6.4)

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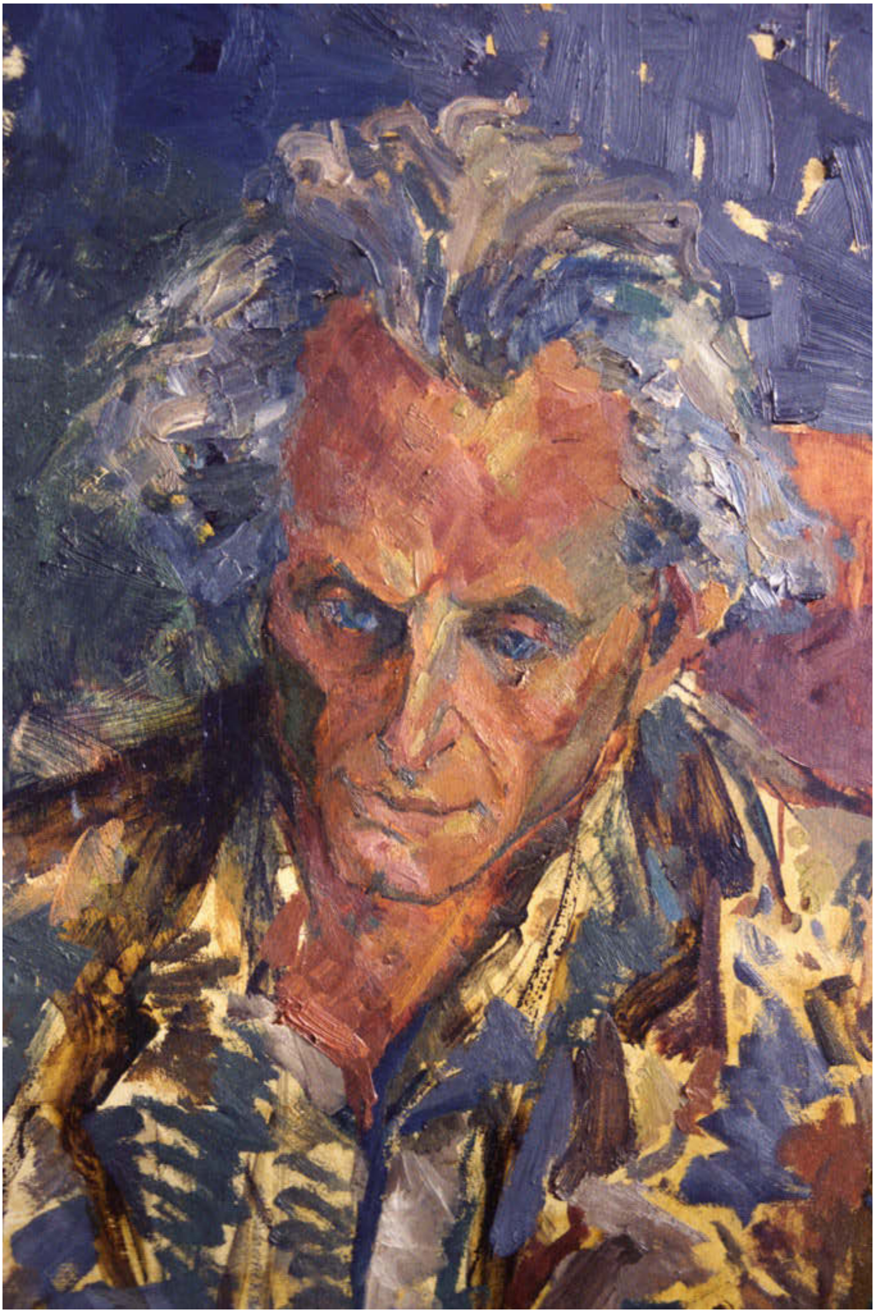

Fig. 7.1 Fröhlich—an oil painting by his wife, Fanchon

Chapter 7
Epilogue

The characteristic hallmark of Fröhlich's particular genius was his ability to recognize when new ideas genuinely had to be introduced and when they did not. In this he was assisted by an ability to understand physics intuitively, rather than through an unhealthy dependence on mathematics—a trend that had become increasingly prevalent during the late 1950s, and one that he lamented and caustically criticised in the introduction to his 1961 review of superconductivity theory (Fig. 7.1):

> This article will be of interest, I hope, to the non-specialist. It addresses itself, however, also to specialists, in particular to those who excel in the use of a single technique such that their understanding has the form of a δ-function. There is the hope that some of them might realise that a calculation should be preceded by an idea [F109].

Fröhlich's strength did not lie so much in technical fluency, but rather in the wealth of ideas he constantly entertained and juggled with—recall Pauli's comment that in Fröhlich……'we have someone who can not only calculate but can also think', and Heitler's reminiscence (Heitler 1973) of working with him in Bristol ……'Herbert's strength consisted in his wealth of *anschaulich* ideas, by which he grasped physics without much reference to the underlying mathematics' (Heitler 1973). It is for this reason that many of his most important works contain valuable insights into physical reality, which are not thoroughly worked out.[1] A good example is the problem of superconductivity, whose ultimate solution by others contained much of the essence of Fröhlich's earlier work.

It was not only from his unique ability to balance the radical with the conservative that his most significant contributions arose, but also from his consummate skill in identifying, for a particular phenomenon, the 'Achilles' heel' on which to

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The erratum of this chapter can be found under DOI 10.1007/978-3-319-14851-9_8

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[1] One physicist known to the author affectionately once described Fröhlich as something of a 'Picasso-like' figure in the world of 20th century theoretical physics!

G.J. Hyland, *Herbert Fröhlich*,
Springer Biographies, DOI 10.1007/978-3-319-14851-9_7

focus attention. In this way, he was often able to access the root of a problem in a way that not only expedited its eventual solution, but also occasionally revealed some hitherto unsuspected possibility within a pre-existing theoretical framework, thereby removing the need for ad hoc hypotheses.

What could have been more conservative (yet paradoxically radical) than his identification of the electron-phonon interaction as the basis of superconductivity? What more radical than his continuous treatment of reflections? And what more seemingly paradoxical than his demonstration that, under appropriate conditions, supply of metabolic energy in a living system can actually result in the establishment of a dynamic order, rather than the heating expected intuitively?

His outlook on physics was truly holistic, and he was constantly alert to the possibility that certain concepts might well have relevance to fields other than those in which they had first arisen. His most heroic attribute, however, was undoubtedly the courage to entertain an unusually wide range of novel ideas, and to have the conviction to express them without fear of possible refutation.

The decisive influence he exerted, often as a 'man ahead of his time', in fields as different as meson theory and biology stands as a strong indictment against fragmentation and over-specialization in theoretical physics—something which was quite alien to his holistic perception of the subject. Perhaps somewhat less apparent, however, is the essential unity and coherence that, nevertheless, underlies much of his work—properties that are, of course, not unconnected with the fact that his most significant contributions actually arose from cross-fertilization between fields of physics so different that they now constitute virtually independent disciplines between which there is little communication.

Fröhlich, however, belonged to the generation of physicists that predated the onset of such divisions—the generation that had been the first to apply, with such outstanding success, the new quantum theory to so many diverse problems in physics, and who witnessed some of the most significant discoveries and events in 20th century physics, such as the relativistic quantum mechanics of Dirac, the development of quantum field theory and of Yukawa's meson theory, the discoveries of numerous particles (starting with the neutron) and of the violation of parity in weak interactions. In addition, there was, of course, the foundation of modern solid-state physics (which had begun with Pauli's paper on the paramagnetism of a non-interacting Fermi gas), as well as developments in statistical mechanics and many-body theory, which grew out of Fröhlich's introduction of the concepts and techniques of quantum field theory into solid-state physics in the early 1950s, arguably the most influential of all his contributions.

Understandably, the subsequent, ever-increasing trend towards fragmentation and over-specialization was, by Fröhlich's generation, considered anathema. It was truly a 'golden age', dominated by such intellectual giants as Bohr, Feynman, Heisenberg, Pauli, Schrödinger and Sommerfeld, all of whom Fröhlich knew personally, and by whom he was deeply respected. It must, of course, be remembered that in Fröhlich's early days the community of theoretical physics was a small and rather closely-knit one, which greatly benefited from much closer contact and exchange than is now possible. Amongst his contemporaries, the theoretician Felix Bloch and the experimentalist Bernd Matthias, both of whom he was especially close to, should not go unrecorded.

For all his eminence, Fröhlich remained always accessible to the two generations of researchers who studied under him and benefited so much from his wise counsel always so generously given; on them his magnetic personality made an indelible impression. His enthusiasm for physics was infectious, and his incisive, critical insight legendary. But he was so much more than the sum of his many publications: he was an incredible human being.

In 1972, he was awarded the Max Planck Medal of the German Physical Society, but was unfortunately unable to receive it in person, since he was convalescing after surgery; accordingly, his wife accepted the Medal on his behalf at a General Conference of the European Physical Society in Wiesbaden, 3–6 October, 1972, and read a short paper [F151] he had written whilst in hospital. In 1979, he was elected Foreign Member of the Stuttgart Max Planck Institute, and was awarded numerous Honorary Doctorates from universities across the world, such as Alberta (Canada), Trinity College Dublin, North Eastern Hill University (Shillong, India), Purdue, and Stuttgart; his first honorary degree was conferred by the University of Rennes (France) in 1955, in recognition of his monumental work in dielectric theory.

His 60th, 70th and 80th birthdays were marked by celebrations in Liverpool (and elsewhere), the latter with a one-day symposium, the proceedings of which, together with those from other celebratory meetings in Stuttgart and Palm Coast, Florida, were later published by Springer as a *Festschrift* (Barrett and Pohl 1987); this was the second *Festschrift* to be dedicated to him, the first (Haken and Wagner 1973) having been published on the occasion of his retirement from the Liverpool Chair in 1973.

Away from physics, with which he maintained a life-long love affair, his interests included skiing, mountaineering, and walking; indeed, whilst walking on a hill-side in Italy, he put out a forest fire when he was in his late 60s! He also much enjoyed music, particularly *Lieder*, which he loved to sing, and opera, which he regularly attended, whenever it came to Liverpool. In addition, he shared with his wife, herself a talented artist (Fig. 7.1), an on-going interest in abstract art and experimental theatre. He was not only a strong supporter of contemporary theatre in Liverpool, such as the Great George's Community Arts Project during the early 1970s, but also strove to promote the production of an experimental play by his wife, written under the pseudonym of Leslie Faust in the late 1960s. The play, entitled *I have a beetle in my black box*, was a mixture of Liverpool beat and Wittgenstein logic, the title being based on that of the famous thought experiment that Wittgenstein introduced in his *Philosophical Investigations*, in connection with his investigations into pain.

He remained extremely fit[2] up to his last years, doing thirty push-ups on his finger tips each morning, and regularly standing on his head for ten minutes in order to increase blood-flow to the brain; indeed he advised colleagues to do the same (Kemmer—personal communication to the author, *c*.1985). He never lost his athletic figure, despite breaking a thigh-bone when he was in his 70s during a visit to a park in Germany, which continued to give him trouble for the rest of his life, even though he had it pinned and re-pinned.

He worked right up to the end, and despite an increasing involvement with biology from 1967 onwards, remained interested and active in many other areas of physics, such as the problem of superconductivity in metals with incomplete inner shells, and later, in 1987, at the age of 81, by a short, but characteristically novel contribution to one of the first international conferences on the new high temperature superconductors; in addition, during the last decade of his life, he began to explore the implications of a bilocal extension of the conventional Dirac theory in the quest for a possible understanding of the separation of Dirac particles into leptons and quarks. When he no longer chose to drive, he daily took a taxi to his office in the Oliver Lodge Physics Laboratory. Indeed, it was there at his desk that he became ill, necessitating his immediate admission to the Royal Liverpool Hospital, where he underwent an operation that confirmed bowel cancer; he died from an infection, aged 85, a few days later, during the early hours of Wednesday, 23 January 1991.

In the December prior to his death, his 85th birthday had been celebrated in Liverpool, on which occasion he was presented with a vaguely Epsteinish bronze head of himself by the Liverpool artist Eva Goldsmith.

With his death, the world of Theoretical Physics lost an internationally renowned and much respected figure: extinguished was a beacon that, for some 60 years, had shone so illustriously and reliably, its penetrating rays illuminating not only some of the most enigmatic areas of the subject but also, and perhaps more significantly, often revealing some hitherto unsuspected connection between seemingly quite unrelated areas, and, during his later years, even between physics and biology.

His legacy to Science, and to Physics in particular, is immense, and his great humanity an example to be emulated.

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[2] Overall, he enjoyed excellent health, apart from suffering from pernicious anaemia, which was diagnosed shortly after his arrival in Bristol in 1935, when he became quite ill, and nearly died; indeed, his doctor told him that he was surprised that he had not died years earlier. It must be remembered that, at a time, the only treatment was concentrate of liver juice. After injections for vitamin B12 became available in the 1950s, he injected himself daily, which kept the anaemia under control.

Photo Gallery

Unless stated otherwise, all photographs are from private collections.

Fig. 7.2 Fröhlich amongst his peers, at the Lorentz-Kamerlingh Onnes Centenary Conference on Electron Physics, Leiden, 1953: *Front row* standing, from the *LH* end: Heisenberg (3rd), Bohr (4th), Peierls (5th), **Fröhlich** (6th), F London (7th); *front row* standing from the *RH* end: Rosenfeld, Pauli. Dirac is 4th from the *LH* end of the 2nd standing row; also in this row are Pippard (between Heisenberg and Bohr), and Bloch (between Fröhlich and London). Fierz is squatting at the extreme *RH* end of the *front row*—Reproduced with the permission of the Huygens and Kamerlingh Onnes Laboratories, University of Leiden

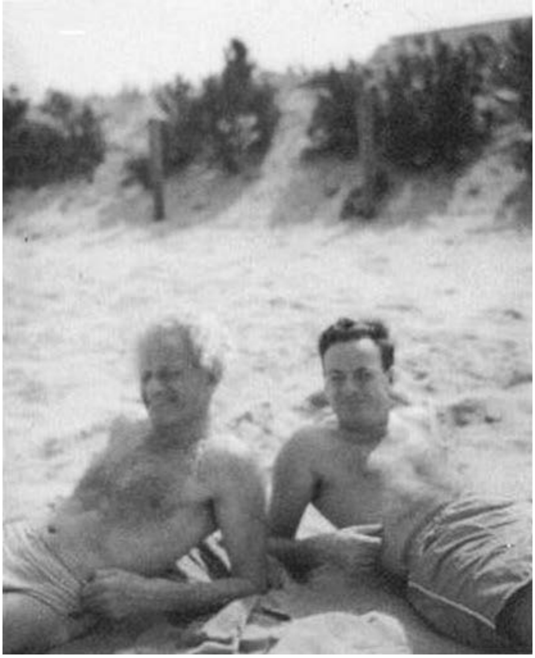

Fig. 7.3 Relaxing with Feynman on a beach somewhere

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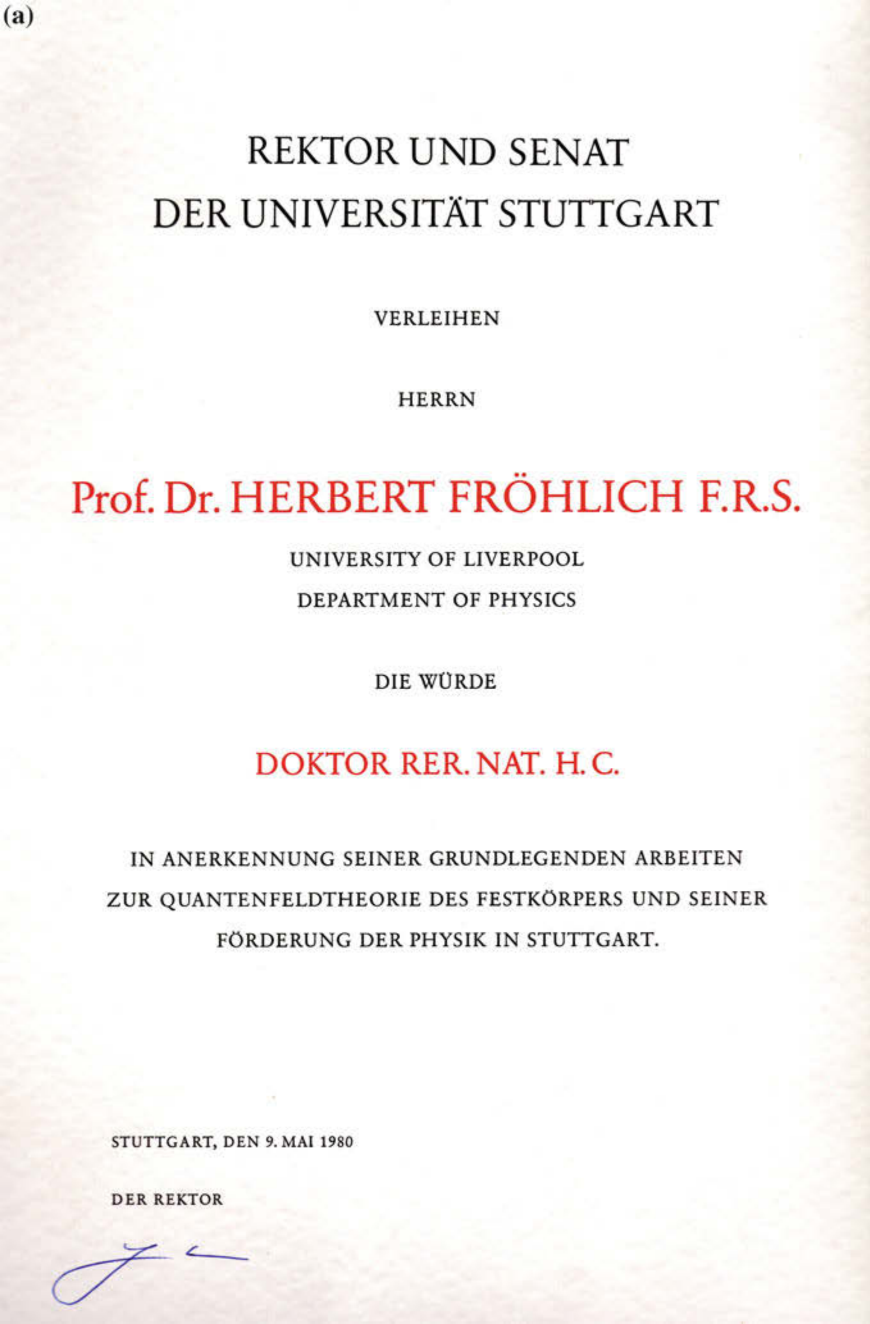

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UNIVERSITY OF LIVERPOOL
DEPARTMENT OF PHYSICS

DIE WÜRDE

DOKTOR RER. NAT. H. C.

IN ANERKENNUNG SEINER GRUNDLEGENDEN ARBEITEN ZUR QUANTENFELDTHEORIE DES FESTKÖRPERS UND SEINER FÖRDERUNG DER PHYSIK IN STUTTGART.

STUTTGART, DEN 9. MAI 1980

DER REKTOR

Fig. 7.4 **a** Honorary Degree Diploma from the University of Stuttgart; **b** Fröhlich in his doctoral robes at Trinity College, Dublin on the occasion of his honorary degree in 1969

Fig. 7.4 (continued)

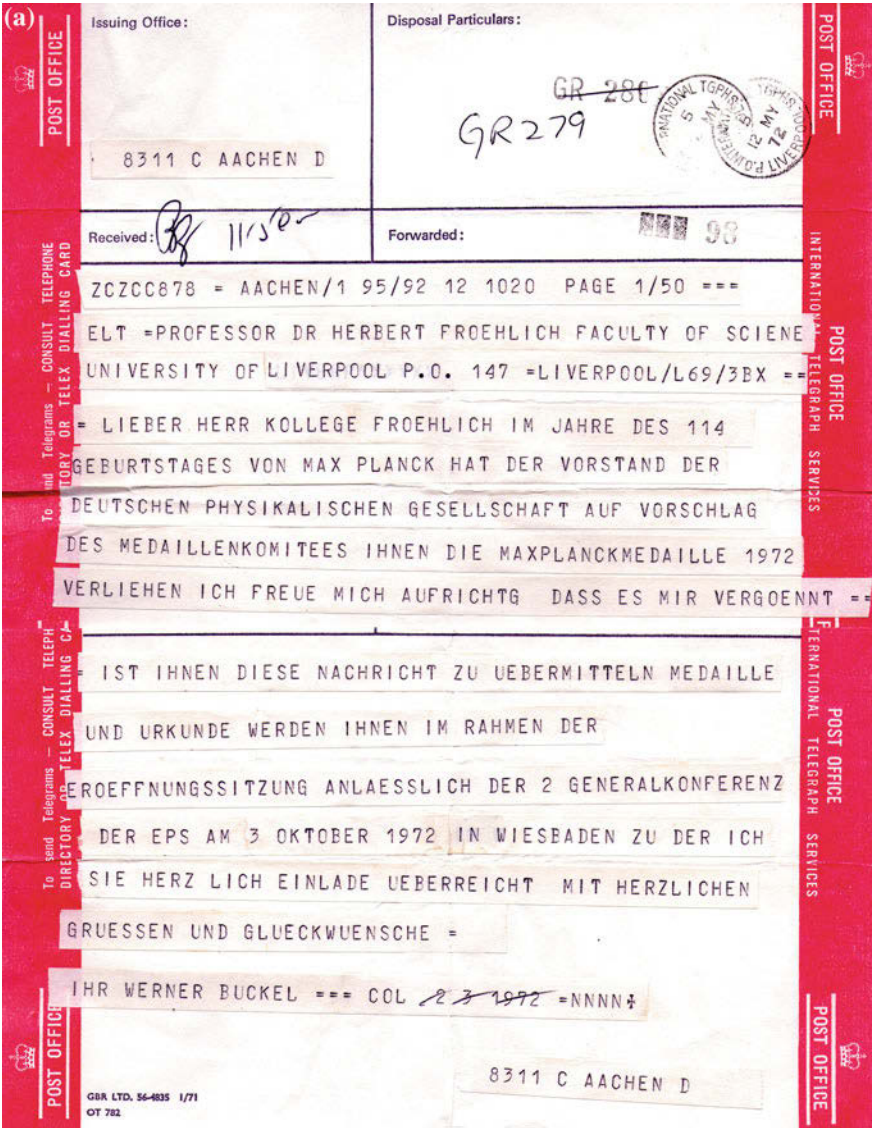

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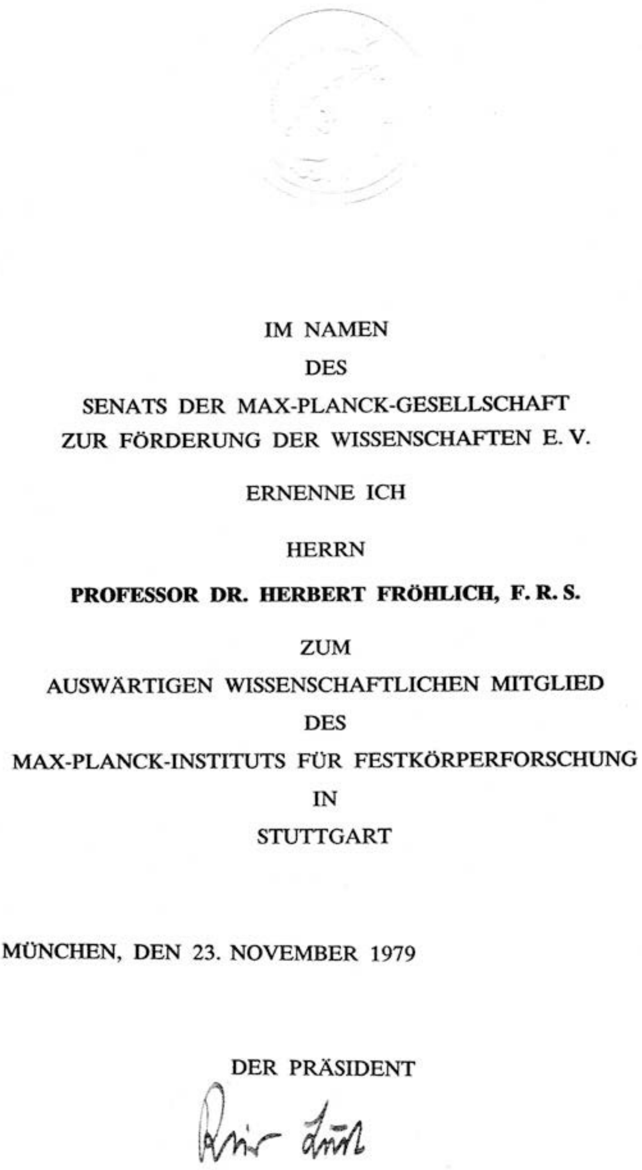
Fig. 7.5 **a** Telegram announcing the award of the *Max Planck Medal* to Fröhlich in 1972; **b** Mrs. Fröhlich accepting the *Max Planck Medal*, on behalf of her husband, from the President of the German Physical Society, Prof. Dr. Werner Buckel, in Wiesbaden, October 1972—Reproduced by courtesy of Physikalische Blätter

Fig. 7.5 (continued)



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MÜNCHEN, DEN 23. NOVEMBER 1979

DER PRÄSIDENT

Fig. 7.6 Membership certificate of the Stuttgart Max Planck Institute

Fig. 7.7 Fröhlich's 80th birthday celebration in Liverpool, with Sewell (*centre*) and the author; *behind* Fröhlich is his brother, Ali

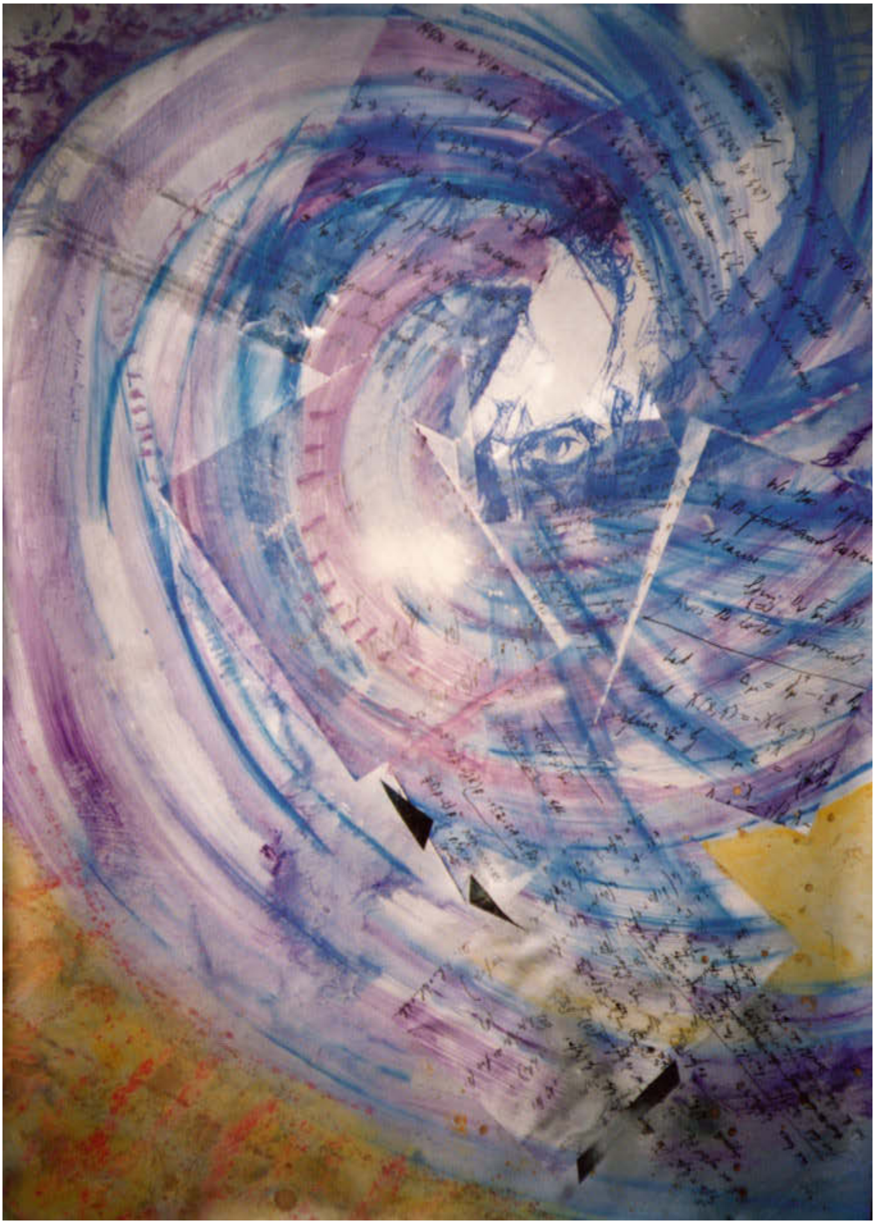

Fig. 7.8 An abstract composition of 1991 by Fanchon Fröhlich, incorporating fragments of her husband's manuscripts and the 1973 portrait of him by herself shown at the beginning of this book

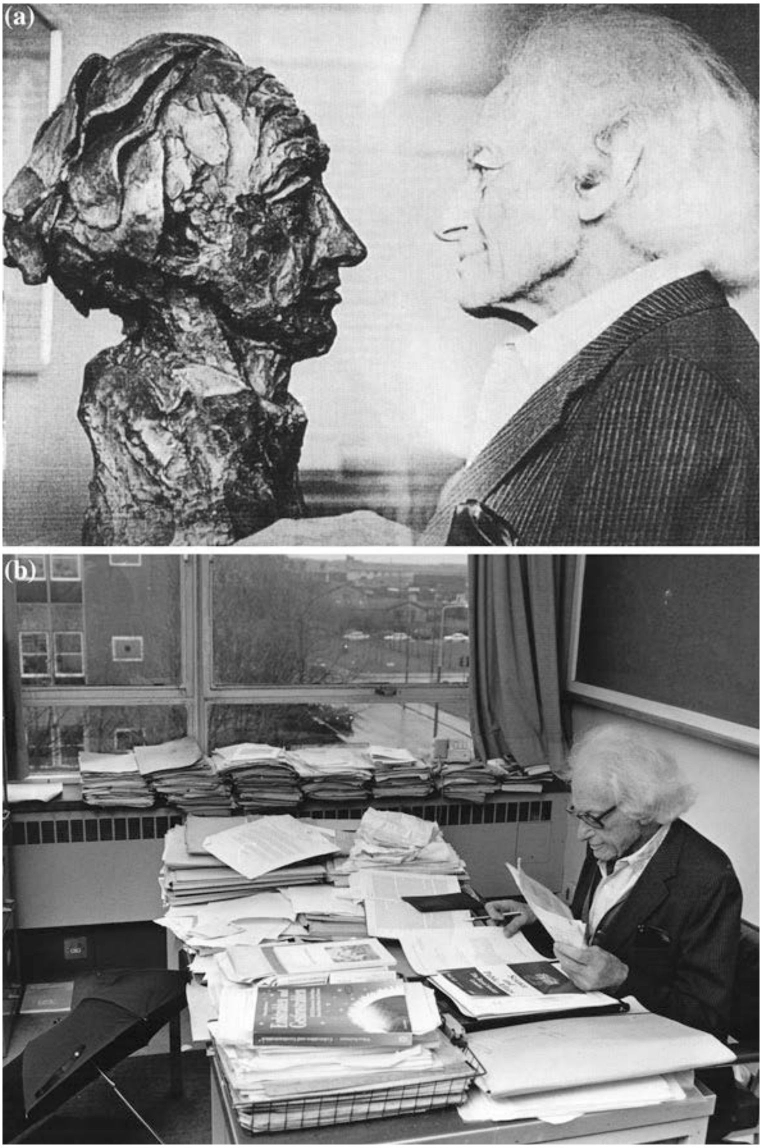

Fig. 7.9 **a** Fröhlich at 85, facing the bronze head by Eva Goldsmith; **b** working in his office later that day—Reproduced by courtesy of the Liverpool Echo

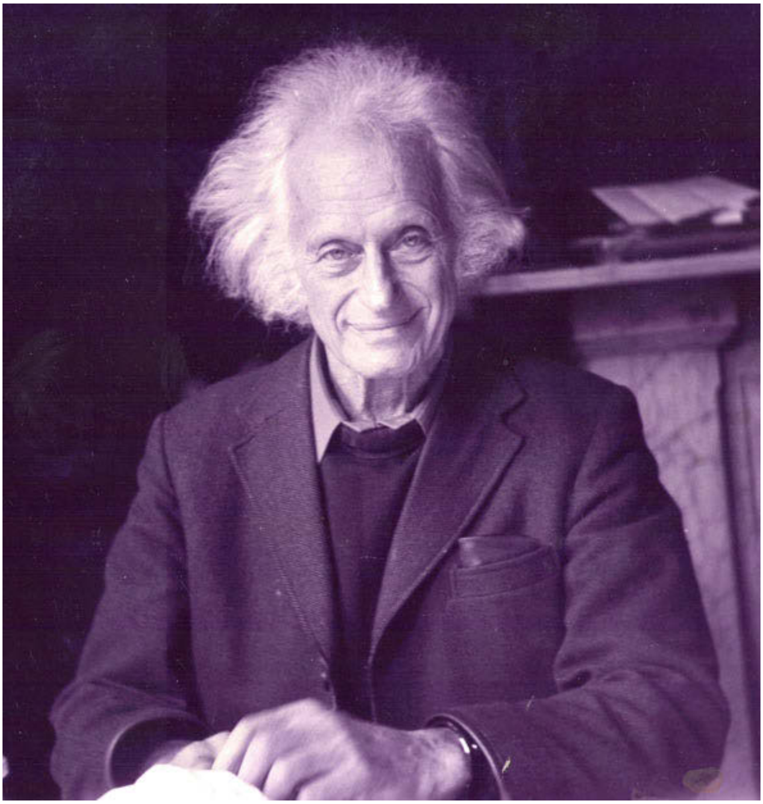

Fig 7.10 The 'grand old man': the final picture, on the occasion of his 85th birthday, 9 December 1990

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Errata to: Chapters 4, 5, 6, 7 and Book Backmatter in Herbert Fröhlich

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The original version of chapters 4, 5, 6, 7 and Book Backmatter were inadvertently published with black and white figures. The correct version of the figures have been replaced.

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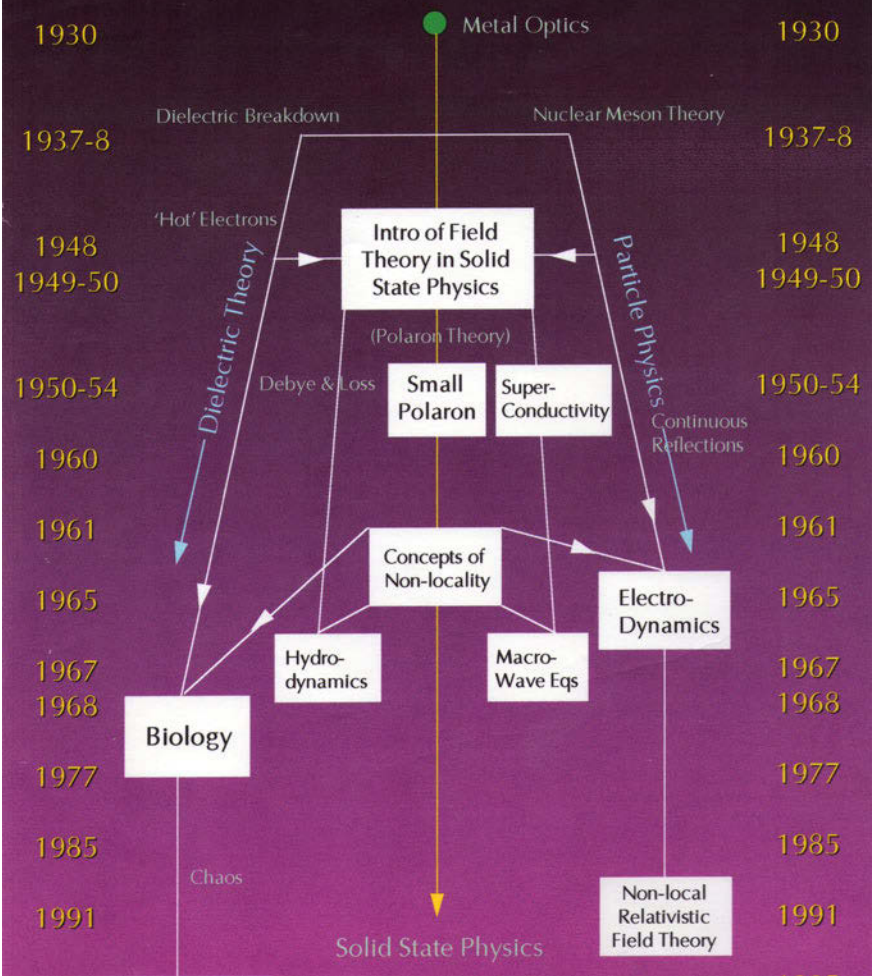

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1965 Electro-Dynamics 1965

Hydro-dynamics

Macro-Wave Eqs

1967 1967

1968 Biology 1968

1977 1977

1985 1985

Chaos

1991 Non-local Relativistic Field Theory 1991

Solid State Physics

Complete Bibliography of H. Fröhlich

The asterisked references denote his most influential works.

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(i)*	1936	'*Elektronentheorie der Metalle*'. (Electron Theory of Metals): Springer Verlag, Berlin. Reprinted, Ann Arbor, *USA*, 1943. Republished by Springer 1969.
(ii)*	1949	'Theory of Dielectrics'. Oxford: Clarendon Press (2nd Edition. 1958; subsequently translated into Russian and Japanese).
(iii)	1983	'Coherent Excitations in Biological Systems'. (eds Fröhlich, H. and Kremer, F.). Springer Verlag, Berlin, Heidelberg, New York, London, Paris, Tokyo.
(iv)	1988	'Biological Coherence and Response to External Stimuli'. (ed Fröhlich, H): Springer Verlag, Berlin, Heidelberg, New York, London, Paris, Tokyo.

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(2)	1931	*Der Schroteffekt nach der Quantenmechanik* (Shot Effect in Quantum Mechanics), *Z. Physik* **71**, 715–719.
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