

The development of the quantum-mechanical electron theory of metals: 1928—1933

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We trace the fundamental developments and events, in their intellectual as well as institutional settings, of the emergence of the quantum-mechanical electron theory of metals from 1928 to 1933. This paper continues an earlier study of the first phase of the development—from 1926 to 1928—devoted to finding the general quantum-mechanical framework. Solid state, by providing a large and ready number of concrete problems, functioned during the period treated here as a target of application for the recently developed quantum mechanics; a rush of interrelated successes by numerous theoretical physicists, including Bethe, Bloch, Heisenberg, Peierls, Landau, Slater, and Wilson, established in these years the network of concepts that structure the modern quantum theory of solids. We focus on three examples: band theory, magnetism, and superconductivity, the former two immediate successes of the quantum theory, the latter a persistent failure in this period. The history revolves in large part around the theoretical physics institutes of the Universities of Munich, under Sommerfeld, Leipzig under Heisenberg, and the Eidgenössische Technische Hochschule (ETH) in Zurich under Pauli. The year 1933 marked both a climax and a transition; as the laying of foundations reached a temporary conclusion, attention began to shift from general formulations to computation of the properties of particular solids.

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INTRODUCTION

The electron theory of metals underwent dramatic development between the first proposal by Paul Drude (1900) and Hendrik Antoon Lorentz (1904—1905, 1909) at the turn of the twentieth century, of a free-electron theory of metals and the writing by Arnold Sommerfeld and Hans Bethe (1933, hereafter cited as SB) of their monumental review in the 1933 *Handbuch der Physik*. From relatively crude classical conceptions, the field reached the point where serious calculations of the properties of particular metals could be undertaken.

The initial breakthroughs were the application by Wolfgang Pauli in 1926 and Sommerfeld in 1927 of Fermi-Dirac statistics in a semiclassical framework to a free-electron gas (Pauli, 1927; Sommerfeld, 1927), and the fundamental shaping by Felix Bloch in 1928 of the quantum

mechanics of electrons in a crystal lattice (Bloch, 1928); these were followed by the further development in 1928—1933 of the quantum-mechanical basis of the modern theory of solids by many theoretical physicists, including Bethe, Bloch, Werner Heisenberg, Rudolf Peierls, Lev Landau, John Slater, Léon Brillouin, and Alan Wilson. From a small number of problems worked on at relatively few institutions, the quantum theory of solids expanded into a substantial field of research at numerous centers in different countries.

The development divides into two distinct phases. The first, charted in an earlier paper by two of us (Hoddeson and Baym, 1980) was devoted to finding the general quantum-mechanical-statistical framework; this phase began with the semiclassical work of Pauli and Sommerfeld and culminated in Bloch's epochal thesis (1928). The second phase, from 1928 to about 1933, which we examine here, saw the realization that the new quantum theory could explain, at least qualitatively and occasionally even quantitatively, the varied properties of solids; a rush of interrelated successes, each following in the wake of previous ones, established the network of concepts that structured the modern quantum theory of solids.

The story revolves in large part around the theoretical physics institutes of the University of Munich, the University of Leipzig, and the Eidgenössische Technische Hochschule (ETH) in Zurich. Their directors of research—respectively, "Geheimrat" Sommerfeld and his students Heisenberg and Pauli—having recently been at the center of the development of quantum mechanics, were eager to explore and test their new capability on problems beyond atoms and molecules. Solid state, by providing a large and ready number of concrete problems treatable by quantum theory, functioned initially as an ex-

tensive proving ground, and somewhat later as a target of opportunity, for the new mechanics. For unlike other subfields of physics (e.g., nuclear physics and quantum electrodynamics), whose theory depended strongly on new experimental findings, solid state was in these years a field whose theoretical effort aimed primarily at explaining phenomena that had been observed for decades.

Heisenberg and Pauli, in retrospect, played a remarkably pivotal role in the development of the quantum theory of solids. Beyond their own concern with exploring quantum mechanics, they turned to a variety of problems of solids with students, *Assistenten* (essentially the modern postdoctoral fellows), and visiting fellows. This generation, grounded in the new mechanics, shaped the fundamental building blocks of the modern theory of solids, explaining in microscopic terms remarkably many experimentally observed phenomena (for example, paramagnetism, diamagnetism, magnetoresistance, the Hall effect, and the behavior of semiconductors), and in turn providing deeper insight into the workings of quantum mechanics itself.

The year 1933 marked both an intellectual and an institutional break in the development, heralded with the appearance of many reviews, most notably by Bethe and Sommerfeld, but also by Brillouin (1930a, 1931), Peierls (1932a), Bloch (1933), Slater (1934), Lothar Nordheim (1934), and others (e.g., Borelius, 1935), of the quantum electron theory of metals. These served as texts in new graduate programs that trained the first generation of specialists in quantum solid-state physics. At the same time, for political as well as intellectual reasons, many of the earlier workers in quantum solid-state theory left the field, while new workers joined, and the center of research shifted from Germany to the United States and England. At this juncture, these reviews expressed optimistically and proudly that all the observed phenomena (even superconductivity) appeared to be, if not solved, then at least soluble in terms of the existing quantum theory. Attention of the quantum theorists now shifted from qualitative and conceptually oriented problems towards more quantitative comparison of theory with experiment. With Eugene Wigner and Frederick Seitz's pivotal 1933 papers on the band structure of sodium as a prototype (Wigner and Seitz, 1933, 1934), the development of approximate methods for dealing with real rather than ideal solids became the principal focus.

Our aim in this paper, rather than to present a comprehensive history of the second phase of development of the quantum theory of metals, is to delineate this period by tracing in detail certain fundamental developments and events, in their intellectual as well as institutional settings, that gave this era of solid-state history its character; in so doing we draw upon original papers, correspondence, and interviews with participants. We focus here on theoretical, rather than experimental developments, particularly on the examples of band theory, magnetism, and superconductivity, the former two immediate successes of the quantum theory, the latter a persistent failure. Our focus is on only one sector of the

study of solids; we do not touch upon the independent tradition of the study of the mechanical properties of solids, whose roots lie in metallurgy. Only quite a bit after this period did this line of research make contact with the quantum theory of solids. Following a review in the first section of the developments that comprise the first stage in the quantum-mechanical electron theory of metals, we turn, in Secs. II, III, and IV, to the test areas of transport phenomena (resulting in band theory), magnetism, and superconductivity; and the concluding section examines the intellectual and institutional transition of the field in 1933.

I. THE QUANTUM-MECHANICAL FRAMEWORK, 1926–1928

A. Problems circa 1925

Mysteries in the theory of metals at the time of the discovery of quantum mechanics involved both fundamental issues, such as why the properties of a metal differ from those of an insulator, and the explanation of particular phenomena such as the specific heat, electrical and thermal conductivities, magnetism, and the relationships between electrical and magnetic effects (see Seeliger, 1921; Solvay, 1927; Hume-Rothery, 1931; Mehra, 1975).

The explanation of the specific heat of metals presented a dilemma: the substantial contribution, $3Nk/2$, of the electrons in metals predicted by the classical free-electron-gas model of Drude and Lorentz, in which the electrons obey the equipartition law, was not observed. On the other hand, the theory of Einstein (1907, 1911), Debye (1912), and Born and von Kármán (1912, 1913), neglecting the electrons, led to a value in good agreement with experiment. (For further discussion see Hoddeson and Baym, 1980.) One could not simply assume that the number of electrons is much smaller than the number of atoms without contradicting optical results; through an undeciphered mechanism, either arising from interactions or perhaps analogous to Planck's quantization of radiation energy, the electrons appeared not to be obeying classical equipartition (see, for example, Jeans, 1921, pp. 302–306 and 400).

In the area of transport, the electron-gas model of Lorentz notably, as well as of Drude and others, gave good agreement with the experimental ratio of the electrical and thermal conductivity—the Wiedemann-Franz law—but the theory could not compute these quantities separately. According to the classical theory the electrical conductivity varied as $T^{-1/2}$, where T is the temperature, if the number of electrons and their mean free path were assumed to be independent, as well as independent of T . But experiment (for example, as described by Grüneisen, 1928) showed the electrical conductivity of pure metals to vary as $1/T$. For the theory to agree with these findings, one would have had to make the apparently unreasonable assumption that the mean free path is much longer at

room temperature than the interatomic distance (and even longer at low temperatures). Similarly, the heat conductivity at high temperatures was computed to be proportional to $T^{1/2}$, but observed to go in direct proportion to T .

Baffling as well was the curious phenomenon of superconductivity, first observed in 1911 by Gilles Holst, working under Heike Kamerlingh Onnes: why should all traces of electrical resistance suddenly disappear in certain metals (including lead, tin, and mercury) and in many alloys, when the temperature falls below a critical value close to absolute zero (Kamerlingh Onnes, 1913a; Flim, ca. 1965)? Furthermore, the considerable empirical information gathered about semiconducting substances, such as, metal oxides and selenium, could not be reconciled with any general theoretical model, e.g., the free-electron gas.¹

How to compute the paramagnetism, diamagnetism, or ferromagnetism of solids was not understood. Questions included why is the paramagnetism of ordinary metals, such as the alkalis, weak and finite as the temperature goes to zero? Why, contrary to the classical theorems of Niels Bohr (1911) and Hendrika Johanna van Leeuwen (1919, 1921; Van Vleck, 1932, pp. 100–102), is there a nonzero diamagnetism? And, given Pierre Weiss's experimentally successful phenomenological mean-field theory of ferromagnetism (1907, 1911), what determined the values of its parameters? Among the many problems concerning the relationship of electricity and magnetism were why does the Hall effect coefficient sometimes have a positive sign and how does one compute the magnitude of the observed change of resistance of metals in strong magnetic fields, the magnetoresistance?

Since no coherent basis was available for establishing the correct microscopic theory, pre-quantum-mechanical attempts to solve the problems of metals represent a groping for reasonable conceptions. Out of these emerged a large number of valid notions, for example, the fundamental idea that microscopic charged particles in a metal behave in many ways like free particles and are responsible for electrical transport (Drude, 1900), and that gas degeneracy was somehow the solution to the specific-heat dilemma (e.g., Schrödinger, 1924)²; these would eventually become part of the modern theory of solids. But since the framework was wrong, fitting the theory to the data, particularly as more numerous experiments were made, required adding an increasing number of *ad hoc* assumptions, most of which proved untenable. Ultimately the pastiche of valid, as well as incorrect, models, and assumptions in the electron theory of metals represented a state of confusion and complexity reminiscent of the state

of physics two decades earlier, prior to the introduction of the quantum.

B. The semiclassical theory of Pauli and Sommerfeld, 1926–1928

The break came in late 1926. Pauli in Hamburg became interested in the fundamental question: what are the areas of applicability of the newly developed Fermi-Dirac and Bose-Einstein quantum statistics? (See Fermi, 1926a, 1926b; Dirac, 1926; Einstein, 1924, 1925; Bose, 1924). As he wrote on 22 November to Erwin Schrödinger, "Recently, I have also been occupied with gas degeneracy [Schrödinger, 1924]. With a heavy heart I have become converted to the idea that Fermi . . . not Einstein-Bose, is the correct statistics. I want to write a short note about an application of it to paramagnetism"³ (Pauli, 1926a). This note (Pauli, 1927), an attempt by Pauli to deepen his understanding of quantum statistics by using paramagnetism as a test, began the quantum theory of metals. Pauli showed that in order to calculate the paramagnetism of (free) electrons in metals, he must assume that they obey Fermi-Dirac statistics—an assumption, he writes, "made by Fermi in analogy to one by the author," his exclusion principle. As a consequence of applying this principle to electrons in a metal, only a small number of electron spins could be aligned by the magnetic field, causing the spin susceptibility to be 2 orders of magnitude smaller at room temperature than the Curie susceptibility, agreeing with observations.

Although in subsequent years Pauli would often express his disdain for solid-state problems—e.g., in 1931, referring to the residual resistance, which his *Assistant*, Peierls, had just calculated, as "Größenordnungsphysik . . . ein Dreckeffekt und im Dreck soll man nicht wühlen" (order of magnitude physics . . . a dirt effect and one should not wallow in dirt) (Pauli, 1931⁴)—in treating the problem of the spin paramagnetism, Pauli opened the way to the development of the modern quantum theory of solids. Later, Pauli, like Heisenberg, would play a pivotal role in motivating, supporting, and criticizing work in the field, especially by Bloch and Peierls.

The next crucial steps were taken by Sommerfeld in Munich, for the task of applying Fermi-Dirac statistics to the specific-heat dilemma and reworking the old Drude-Lorentz theory was not to Pauli's taste. Although Sommerfeld seems to have had little prior intellectual attachment to the electron theory of metals, and by 1927 had not yet written on solid-state physics or quantum statis-

¹For recent historical accounts of semiconductor prehistory see Hempstead (1977) and Kaiser (1978).

²For recent discussions, see Hanle (1977) and Belloni (1978); also Mehra (1975).

³The Pauli correspondence is cited in the references as WP1 and WP2, where WP1 refers to the first volume, edited by Hermann *et al.* (1979), and WP2 to the second volume, edited by von Meyenn *et al.* (1985) of the collected Pauli correspondence.

⁴We thank Karl von Meyenn for sending us a copy of this and other letters prior to their publication.

tics,⁵ the problem posed by extending Pauli's work appealed to his expertise in applying sophisticated mathematical methods to a wide variety of physical problems. Unlike Heisenberg and Pauli, he appears as a rule to have been more interested in the mathematical solution of a problem than in its underlying physics.

In 1906, Sommerfeld had been called from the chair he held at the Technische Hochschule Aachen to the chair of theoretical physics at the University of Munich, and given a large institute. He was already at this time a well-known figure in the scientific community, and had become, as editor of the physics volume of the *Enzyklopädie der Mathematischen Wissenschaften* since 1898, a correspondent of many eminent physicists. Sommerfeld's institute would have an impressive history of work on solids, which helped to nurture the early development of the quantum theory of solids. Even before World War I, a generation of physicists who would carry out pioneering solid-state studies emerged from here, among them Peter Debye, Peter Paul Ewald, Max von Laue, and Brillouin. In 1912, von Laue, W. Friedrich, and P. Knipping performed in the institute the historic x-ray diffraction experiments that provided the first experimental look inside the crystal lattice.⁶

By the 1920s, Sommerfeld's institute was one of the major international centers for theoretical physics, attracting students and traveling fellows—including Heisenberg, Pauli, Bethe, Peierls, Gregor Wentzel, Walter Heitler, and Fritz London—members of the new generation that developed quantum mechanics and its applications. These students and fellows would in turn build up new centers with an active exchange of ideas as well as scientists, helping to spread the Sommerfeld teaching and research tradition. Most of the creators of the quantum

theory of solids in the years 1926–1933 were connected at some point with Sommerfeld's institute (Manegold, 1970; Benz, 1973; Eckert, 1986).

The physical arrangement of the institute allowed for maximum communication among those working there; in three large connecting rooms of comparable size were Sommerfeld's personal office and library, the office and library of Sommerfeld's *Assistent*, and a work room for approximately 20 graduate students and visitors. A fourth tiny room contained a spiral staircase leading to the basement where the early x-ray diffraction experiments were carried out (Bethe 1981a).⁷

Communicating his interests enthusiastically in his courses, articles, and numerous lectures, both at Munich and elsewhere,⁸ Sommerfeld involved large numbers of students and colleagues in research on the theory of metals. He taught three courses: a six-semester undergraduate course to approximately 100 students (three times a week over a three-year period), a weekly seminar on current research topics to a group of about 20, and a special topics course to about 20 advanced graduate students (twice a week in alternate semesters) (Peierls, 1981a; Bethe, 1981a).⁹ Sommerfeld's *Assistent* also taught a course twice a week to approximately 20 students; in alternate years, the subject was quantum mechanics, which was not included in the main Sommerfeld sequence (Bethe, 1981a). Sommerfeld's undergraduate course, out of which eventually grew the famous Sommerfeld textbooks, included classical mechanics, the mechanics of continuous bodies, thermodynamics and statistical mechanics, electrodynamics, optics, and mathematical physics.¹⁰

The seminar pursued outstanding recent developments

⁵Drude, although corresponding with Sommerfeld at the time of his electron theory of metals (Drude, 1900), does not refer to the theory in the four letters that he wrote to Sommerfeld between 1899 and 1901 in the Sommerfeld Nachlass (the Sommerfeld collection at the Deutsches Museum, referred to here as DM). While not actively working in the field, Sommerfeld did follow its developments, lecturing in 1908, 1910, and 1912 (*Archive for History of Quantum Physics*, a collection of microfilms, transcripts and tapes, hereafter referred to as AHQP; see footnote 90) on the electron theory of metals, and related questions of kinetic theory of gases. His first *Assistent*, Debye, wrote his *Habilitation* thesis on electrons in metals (Debye, 1910). Furthermore, Karl Herzfeld, as Sommerfeld's *Assistent* in Munich, lectured in 1920–1922 on metals, the theory of gases, and magnetism.

⁶For the role of Munich in these experiments see Ewald (1962), Forman (1970), Chap. 1 of Hoddeson, Braun, Teichmann, and Weart (1987), and interviews with Debye, Einstein, Ewald, and Friedrich (AHQP).

⁷Bethe (1981a) reminisced about the creative work carried out in the institute's basement by Sommerfeld's machinist Karl Selmayr, who out of little balls and wire would construct excellent models of crystals, which were sold throughout the world. F. Hund, for example, traveled to Munich several times in order to obtain crystal models for the physics institutes in Göttingen, Rostock, and Leipzig (Hund, 1982).

⁸A good example is Sommerfeld's lecture course at the 1931 Michigan Summer Symposium (Goudsmit, 1961; Dennison, 1967; see also Meyer *et al.*, 1944). Another example is the lectures Sommerfeld delivered in Japan at the Law School of Tokyo Imperial University, in December 1928 on "Selected Problems on Wave Mechanics and Theory of Electrons." We wish to thank Atsushi Katsuki and Shuntiki Hirokawa for a copy of notes from these lectures.

⁹Peierls's recent memoirs (1985) contain a wealth of reminiscences expanding on the material in the Peierls (1981) interview of Hoddeson.

¹⁰According to Bethe (1981a), Sommerfeld—at heart a mathematical physicist—gave a particularly beautiful series of lectures in the mathematical physics section.

in all areas of theoretical physics;¹¹ in 1926–1928 it was devoted mainly to topics in quantum mechanics. In this seminar Sommerfeld transmitted his personal style and taste in research by working closely with its members, often drawing them out by his “principal technique . . . to appear dumber than any of us” (Houston, 1964). The advanced topics course, also in this period concerned with problems in quantum mechanics, focused in 1927–1928 on the quantum electron theory of metals. Bethe, a research student, and Peierls, a third-year student, were among those whose interest in solid-state physics was kindled in this course (Peierls, 1981a), in which, Bethe recalls, Sommerfeld “told us what he had discovered in the last week. It was very fascinating” (Bethe, 1981a, 1981b). Max Born later compared Sommerfeld’s method of personal instruction to

the tutoring at the old British Universities, but less methodical and formal. . . . Often before or after the Colloquium he was seen at the Hofgarten-Cafe, discussing problems with some collaborators and covering the marble tables with formulae. It is reported that one day an integral resisted all attempts at reduction and was left unfinished on the table; the next day Sommerfeld, returning to the same table, found the solution written under the problem, obviously meanwhile worked out by another mathematician taking his cup of coffee with greater leisure. . . . A great part was played by invitations to join a ski-ing party on the ‘Sudelfeld’ two hours by rail from Munich. There he and his mechanic Selmayr . . . were joint owners of a ski-hut. In the evenings, when the simple meal was cooked, the dishes washed, the weather and snow properly discussed, the talk invariably turned to mathematical physics, and this was the occasion for the receptive students to learn the master’s inner thoughts (Born, 1952–1953).

In early 1927, Sommerfeld, on seeing (Pauli, 1956) proofs of Pauli’s paper on paramagnetism, realized that the approach of using Fermi-Dirac statistics with the free-electron-gas model might work as well for other problems of metals, particularly those Drude discussed at the turn of the century using classical statistics. Reworking the program of the electron theory of metals by applying a particular mathematical formulation, the Fermi-Dirac statistics, suited Sommerfeld’s personal style in physics. He quickly produced a series of successful results (discussed in detail in Hoddeson and Baym, 1980), showing, for example, that the new statistics decreased the specific heat of the electrons in a metal at room tem-

perature to an unmeasurably small value.¹² Optimistically he wrote in *Die Naturwissenschaften* of his “overall impression” that “without any doubt . . . the new statistics removed the contradictions in the older theory” (Sommerfeld, 1927, p. 831). While this impression turned out to be overstated, Sommerfeld’s work in Munich in 1927 would lay the foundation for the quantum theory of solids.

The work begun in Sommerfeld’s circle on the quantum theory of metals soon spread to many other centers. Those who helped to bring the theory to the United States included Carl Eckart, William Houston, Edward Teller, Bethe, Bloch, William Allison, Nathaniel Frank, I. I. Rabi, Edward Condon, and Philip Morse (Slater 1975, pp. 164 and 165; Bethe, 1981a). Peierls and Bethe were among those who would bring the theory to England. Heisenberg and Pauli, still intellectually close to their former mentor, directed important work on the theory at Leipzig and Zurich, respectively. Sommerfeld’s influence was also felt abroad through his articles, e.g., by Nordheim, then working with R. H. Fowler in Cambridge (Nordheim, 1962), as well as through his lectures. For example, Walter Brattain, after attending Sommerfeld’s course at the 1931 Michigan summer symposium on the electron theory of metals (Goudsmit, 1961) delivered a special series of lectures on the theory at Bell Laboratories (Becker, 1931a, 1931b; Brattain, 1954, 1974, 1975).¹³

Sommerfeld left Munich from August 1928 to May 1929 for travels around the world. His lectures during this trip, e.g., in Tokyo in December 1928,¹⁴ helped further to disperse the early quantum theory of metals. During this year, Peierls went to Leipzig to work with Heisenberg (Peierls, 1981a), and Bethe went to Frankfurt and then to Stuttgart to work as Ewald’s *Assistent*.

C. The quantum-mechanical theory of Bloch, 1928

Sommerfeld’s theory served as a precursor of the fully quantum-mechanical theory, pinpointing agreements and discrepancies with experiment and indicating where quantum-mechanical building blocks were needed. Even within his own circle in Munich, serious failings of Sommerfeld’s theory were apparent; predictions often disagreed with experimental findings, e.g., on the size and functional dependence of the resistivity, the magnetoresistance, and various galvanomagnetic and thermoelectric effects such as the Hall effect. While Sommerfeld was aware of these problems, as Peierls reflected recently, he was optimistic that in one way or another they would be resolved. But to do so would require a fully quantum-

¹¹For example, Peierls reported on the recent work of Dirac and Jordan on transformation theory. Bethe reported on perturbation theory as described in the galley proofs of Schrödinger’s original papers on quantum mechanics, which Sommerfeld had obtained for use in his seminar. The group was mixed, including undergraduates, graduates, professors, and visitors (Bethe, 1981a; Peierls, 1981a).

¹²According to Peierls (1981a), Sommerfeld was in good touch with recent experiments, in particular, those of Grüneisen in Berlin on the temperature dependence of the electrical resistivity.

¹³The historical link between this course and the discovery of the first transistor is traced in Hoddeson (1980, 1981).

¹⁴See Sommerfeld’s lectures in Japan (footnote 8).

mechanical theory of metals.

The crucial first step in developing this theory was taken by Bloch, Heisenberg's first student in Leipzig. As Bloch recalls, Heisenberg considered problems of solids "as a field to which quantum mechanics could fruitfully be applied" (Bloch, 1981). Through his work on the helium spectrum (Heisenberg, 1926a), Heisenberg realized as early as 1926 that the quantum-mechanical exchange interaction was the likely source of the local field in Weiss's theory of ferromagnetism (see Sec. III below). From his study of the work of Heitler and F. London on homopolar bonding in the hydrogen molecule (Peierls, 1981a; Heisenberg, 1926b, 1926c; Pauli, 1926b; Heitler and London, 1927; Van Vleck, 1932, p. 337), Heisenberg had grown convinced that he could derive the local field by their approach, and he had made sufficient progress on the theory of ferromagnetism that when Bloch arrived in Leipzig in the fall of 1927, Bloch felt that, "Well Heisenberg has it already in a nutshell . . . I don't want to just simply work it out." Furthermore, "I'm not going to compete with Heisenberg" (Bloch, 1964, 1976, 1981). Instead, Bloch chose to study the quantum mechanics of electrons in metals. (However, approximately a year later, he would embark on a major effort to develop further Heisenberg's treatment of the ferromagnetism problem.)

The Leipzig Physical Institute was in 1926 still a stronghold of old-fashioned classical physics, influenced by the views and habits of the "Geheimräte" Otto Wiener and Theodor Des Coudres, who in the quarter century of their regime scarcely allowed the infiltration of the new quantum ideas into their academic life.¹⁵ Within a year the Institute changed radically, occasioned by the deaths of Des Coudres and Wiener and the departure of Georg Jaffé. Sommerfeld played an essential role in filling the newly vacant posts, first with Wentzel in 1926 and then, in 1927, with Debye, who was at the time in Zurich, and Heisenberg.

The pedagogical style that the Sommerfeld team ushered in was modeled largely after that of Munich, with the stimulating research environment revolving around courses and seminars (Leipzig, 1927): the theory lecture course, 4 hours per week, dealt with classical mechanics, thermodynamics, electrodynamics, and optics in a four-semester cycle. The special 3-hour weekly lecture treated subjects such as "modern problems of atomic physics" and "quantum mechanics." Advanced students were offered a seminar on the "structure of materials," organized jointly by Heisenberg and Wentzel [and from 1929 Friedrich Hund, after Wentzel became a Professor (Ordinarius) in Zurich]. The seminars were informal, on a "high level," usually small (consisting of approximately six students, assistants, and professors), and focused on research by the participants or important articles in the

current journals. "Then we sat together and we started to play ping-pong—it was all very informal" (Bloch, 1981). While experimentalists did not usually attend the seminars, and in general the relationship between theorists and experimentalists was tenuous (Bloch, 1981), contact was established through a weekly colloquium arranged by Debye.

Bloch, who had studied in Zurich with Schrödinger, arrived in the winter semester of 1927–1928, primarily on the advice of Debye, to continue his studies (Bloch, 1964). Peierls came from Munich to Leipzig as a student in the fourth semester in the spring of 1928, at the time Sommerfeld was preparing to set out on his world tour (Peierls, 1980, see p. 28; 1963); Teller and Rabi followed in the fall, as did Houston, after a sojourn of several months with Sommerfeld, to spend the remaining time of his Guggenheim fellowship (Houston, 1964). Peierls recently reflected on the happy environment for theoretical solid-state research there, arising out of Heisenberg's realization that "there was an open problem . . . electrons in metals were one proving ground for quantum mechanics." Peierls also recalled the differences between working with Heisenberg and Sommerfeld. While both were approachable, Sommerfeld was such a busy man that "you didn't call on him quite as easily as on Heisenberg." On the other hand, Heisenberg was about half Sommerfeld's age, very modest, and "his ambition to excel in table tennis was more obvious than his ambition to be a great physicist" (Peierls, 1981a).

Bloch's thesis work began with a question Heisenberg posed: How are the ions in the lattice to be dealt with (Bloch, 1981)? Bloch took a major step forward by approximating the lattice by a three-dimensional periodic potential, and ignoring the mutual interaction of the electrons so as to reduce the problem to a one-body calculation. Then drawing upon the idea Heitler and London had used in their treatment of the hydrogen molecule, of constructing electronic wave functions starting from a basis of unperturbed single-atom ground-state orbitals (Heitler and London, 1927)—Bloch's familiarity with the Heitler-London method dated from 1926, when Heitler and London were in Zurich and all three would enjoy walks together (Bloch, 1981)—he solved in perturbation theory the single-electron problem.

Bloch assumed a potential in which the electrons were bound to the lattice with an energy much larger than the kinetic energy of their motion through it—the "tight-binding" method—so that, most of the time, any given electron revolves about the nucleus of a certain atom and rarely ever moves to a different atom. By solving the Schrödinger equation in Fourier-analyzed form, Bloch discovered the important theorem that the wave function of an electron energy eigenstate in a perfect periodic lattice has the form (now known as a "Bloch state") of a product of a free wave and a periodic function u with the period of the lattice: $e^{ik \cdot r} u(r)$. By implying that electrons would move freely through a perfect lattice, this theorem explained why Sommerfeld's semiclassical model worked so well: despite the ions, the electrical conductivi-

¹⁵Des Coudres (1862–1926) and Wiener (1862–1927) were typical classical physicists of lesser rank. For a picture of the period see McCormmach (1982).

ty of a perfect lattice of identical atoms would be infinite, with finite conductivity a result of lattice imperfections or ionic motion. In Bloch's published article on his thesis (Bloch, 1928, 1930a) he rederived this pivotal "Bloch theorem" using group theory, then fashionable (Bloch, 1981), and also laid the foundation of the quantum theory of electrons in lattices by developing many basic principles, as well as techniques still in use today.¹⁶

II. BAND THEORY, 1928–1933

The key to understanding the electronic transport and optical properties of solids was band theory; with this major conceptual building block in place one could finally account for both fundamentals, such as the difference between metals and insulators or the nature of semiconductors, and particular phenomena, such as the Hall effect and magnetoresistance.

The band picture, in retrospect so evident once the form of the solutions of the Schrödinger equation in a periodic potential was understood, came into full focus only over the three years following Bloch's thesis. By carrying out the first calculation of electron wave functions in a metal that took the ions into account, Bloch's paper laid the foundations of band theory. As Bloch showed, the electron energy-momentum relation was no longer simply quadratic; hidden within was the structure from which the concept of the "hole" would later emerge. While Bloch derived in this paper only the ground-state band wave functions and energies, he recalls that the concept of many bands was "completely obvious" from the start: "since an atom has excited states, to each excited state there would belong a band," with gaps between (Bloch, 1981). However, the role of bands and band gaps in determining the properties of solids was not yet explicitly recognized.

Contemporaneously with Bloch in Leipzig, Bethe in

Munich was writing his thesis (Bethe, 1928, 1981b; Bernstein, 1979) under Sommerfeld in 1927–1928 on the solid-state problem of electron scattering in crystals. Although the discovery by C. J. Davisson and Lester Germer of electron diffraction in 1926 was generally perceived as a confirmation of quantum mechanics, a number of technical problems remained. The experimental diffraction maxima did not occur at the predicted energies. "And so," Bethe recalls, "Sommerfeld asked me, 'well, please clear that up and tell us why that is.'" Bethe, following closely the methods developed at Sommerfeld's institute in 1917 by Ewald in his "dynamic" theory of x-ray diffraction (Ewald, 1917, 1927), explained how the electrons having negative potential energy in the metal have greater kinetic energy inside than outside, with a consequent shortening of their wavelength, thus explaining the discrepancy. Bethe dealt with, among other topics, the phenomenon of "selective reflection," in which electrons impinging on a metal in certain energy intervals are observed to be totally reflected. To explain this effect he carried out, in close correspondence with Ewald, a "weak-binding" approximation for the wave function of an electron in a periodic crystal, starting from his realization—independent of Bloch—that the electron wave functions must be of the form, $e^{ik \cdot r} u(\mathbf{r})$.

Setting up the mathematical machinery for developing band theory, the same as that later employed by Peierls and others, Bethe showed, as Ewald found earlier for x rays (Ewald, 1917, pp. 592ff), that for certain incident directions and energy intervals one cannot construct propagating solutions for electrons in the crystal. And the connection of these intervals with the forbidden gaps between bands would not, however, be made until 1930 by Morse (1930). And the concept of band gaps, although lurking about in Bethe's calculations—even so far as his writing out the usual weak-binding secular problem that exhibits gaps—was not made sufficiently explicit for his thesis to play a significant role in the further development of band theory.

At Eastertime 1928, as Bloch was finishing his thesis, Peierls arrived in Leipzig. Heisenberg, having explored the Heitler-London method in ferromagnetism (Heisenberg, 1928a; Bloch, 1976), suggested that Peierls study the usefulness of this approach to the conductivity problem by constructing many-electron wave functions that took into account from the start the electron-electron interactions. (Bloch's calculation, by contrast, was based on single-electron wave functions with no account of electron-electron interactions.) But, Peierls recalls, "I struggled very hard but couldn't get away from the conclusion that . . . this model . . . would have no conductivity" (Peierls, 1981a).

Heisenberg then suggested that Peierls look at the Hall effect, the buildup of a transverse voltage as an electrical current passes through a metal in a magnetic field. Sommerfeld's semiclassical theory, based on free electrons, could not essentially improve upon the classical result for the Hall voltage, which although agreeing well with observation for the alkalis and certain other metals

¹⁶Bloch's work on electrical conduction is discussed in Hoddeson and Baym (1980). Also significant was the work on conduction by Houston, and later by Nordheim. While in Munich in the spring of 1928, Houston had examined the problem of the temperature dependence of electrical resistivity (Houston, 1928, 1929). Realizing that the zero-point vibrations of the lattice scatter x rays, Houston attempted an analogous description of the scattering of electron "waves," in terms of the mean thermal displacements of individual atoms. The correct calculation, which Bloch carried out shortly afterwards, required employing the actual phonon modes in the full Boltzmann equation. Houston (1964) recalls Sommerfeld describing his work as "the first decent treatment of the resistance law." Despite their common interests, there seems to have been no significant interaction between Bloch and Houston in this period. Nordheim (1931) refined Bloch's work, including, for example, the more accurate "rigid-ion" description of the interaction of electrons with ions, and extended it to describe further phenomena such as conduction in alloys and thermoelectric phenomena.

(copper, gold, silver, lead, palladium, and manganese), could not account for the variations of the Hall voltage with temperature or magnetic field, or explain why for certain metals it gave the wrong magnitude or sometimes even the wrong sign (SB, pp. 366 and 562).

The clue to understanding this “anomalous” or “positive” Hall effect lay in going beyond free-electron theory and fully exploiting the nonquadratic relation Bloch found between electron energy (E) and crystal momentum (k). This relation implies in particular that electrons in the upper part of the band (Peierls, like Bloch, considers only the ground-state band, also in the tight-binding approximation) have a group velocity decreasing with crystal momentum, opposite to the behavior of free electrons; in other words, the electrons have a negative effective mass due to the negative curvature of $E(k)$. Peierls (1980, p. 30) recalls that in unraveling the positive Hall effect “I . . . first had to convince myself that the effect of the magnetic field on the wave vector of the electron was the same as for a free electron of the same velocity, but that the mean velocity of the electron was given by dE/dk , and therefore different from that for a free electron of the same k , if the energy function $E(k)$ was different. It was obvious, in particular, that in Bloch’s tight-binding model the energy would flatten off near the band edge, so that the current would there go to zero.”

Peierls submitted the full account of the positive Hall effect to the *Zeitschrift für Physik* (Peierls, 1929a) at the end of 1928, and described the theory at a meeting of the Deutschen Physiologischen Gesellschaft in Leipzig on 19–20 January 1929 (Peierls, 1929b). In this paper he calculates the system’s response to electric and magnetic fields by first showing that (in a one-band model) the time rate of change of the components of an electron wave packet in electric and magnetic fields is given by the matrix element of the Lorentz force, generalizing Bloch’s early argument (Bloch, 1928) on the behavior of Gaussian wave packets in an electric field. Then, with considerable insight, he uses this result to justify writing, in “analogy with the corresponding formula of classical mechanics,” the effect of the fields on the time rate of change of the electron distribution function in terms of the Lorentz force. Including electron-lattice collisions by generalizing Bloch’s integral equation (derived from the Boltzmann equation) for the electron distribution function, he derives a result for the Hall constant that reduces in the limit of a slightly filled band to the classical result; however, in the limit of a nearly filled band it reduces instead to the classical result for carriers of *positive* charge, whose number equals the number of *unfilled* states in the band. Peierls almost makes explicit the idea of the “hole,” that vacancies near the top of an otherwise filled band behave as positively charged particles of positive effective mass. In fact, he points out (Peierls, 1929a, p. 264) how his result is connected with Pauli’s 1925 reciprocity principle (Pauli, 1925a) that draws a correspondence between an atomic state having a certain number of electrons outside a closed shell and the state with the same number of holes (Lücken) in the closed shell. Furthermore, in pointing out

that the electrical conductivity must vanish in the case of a completely occupied band, Peierls, although not mentioning it, found the basic characterization of electrical insulators.

The picture of the hole as a positively charged entity would not be fully delineated until the middle of 1931, when Heisenberg used Peierls’s work on the Hall effect as one illustration of the “far-reaching analogy between the terms of an atomic system with n electrons and a system which is n electrons short of having a closed shell”¹⁷ (Heisenberg, 1931a; Spence, 1958, p. 58). Showing that a hole is described by a complex-conjugate wave function (a result perhaps more familiar now from the point of view of second quantization), he concludes that for states near the top of the band “the holes (Löcher) behave exactly like electrons with *positive* charge under the influence of a disturbing external field . . . the electrical conduction in metals having a small number of holes can in every connection be written as the conduction in metals having a small number of positive conduction electrons.”

P. A. M. Dirac had already, by December 1929, formulated the concept of the hole in quantum electrodynamics,¹⁸ a vacancy in the sea of negative-energy electrons. [Although he initially suggested that this hole could be the proton, he properly identified it as an “anti-electron” or positron in mid-1931 (Dirac, 1931, p. 61) (a month prior to Heisenberg’s paper).] The analogy, so obvious today, between the solid-state hole and the Dirac hole, and the fact that both holes were invented in the same period, has led to a common belief of an historical connection between the two concepts. As Bloch had recently mused on such a connection, “There was [in this period] so much interplay between all the physicists . . . that as soon as somebody had an idea, another one took it up and put it in a different form and used it somewhere else . . .” (Bloch, 1981). However, there is no internal evidence of a relationship in the development of the two concepts. Neither the Peierls-Heisenberg nor the Dirac papers, in presenting their pictures of holes, refer to the other. Rather, both concepts appear to have a common root in Pauli’s work on almost-filled shells of many-electron atoms, a correspondence both Heisenberg (1931) and Dirac (1930, 1931) draw upon. While the analogy between the two holes may have been apparent to some [“certainly when Dirac’s paper on the hole theory came

¹⁷Peierls (1985, p. 38) recalls Heisenberg telling him, “This situation looks very similar to one I have encountered in atomic spectra, where the spectrum of an atom with one or two electrons in the last shell is very similar to that of an atom that has one or two electrons missing from the complete shell.” Peierls is uncertain as to when this “significant conversation” took place, but reflects that were it at the time Heisenberg introduced him to the problem, “then he knew the answer from the beginning . . . and left me just to work out the mathematical details.”

¹⁸The physical picture of the Dirac hole appears first in Dirac (1930), and is discussed further in Dirac (1931).

out, it was obvious . . . that there was an analogy with the electrons in metals and vacant places," Peierls recalls (Peierls, 1981a; see also Mott, 1979)], one does not see mention of it in the literature of the period. Bethe recently ascribed this omission in part to the fact that the Dirac holes were not generally taken seriously until after the positron was discovered in 1932 (Bethe, 1981a); although the infinite continuum of states in the Dirac theory made it very hard to construct a finite theory of quantum electrodynamics, in a solid "it was clear there was a finite number of states, so it was very obvious that an unoccupied state was a hole and how it would behave" (Bethe, 1981a).

In the spring of 1929, Peierls moved to Zurich from Leipzig to work with Pauli at the ETH. Like those in Leipzig, the available physics posts in Zurich in the 1927 round of appointments were occupied by Sommerfeld's students. After Heisenberg declined the post as Debye's successor, Pauli was offered, and accepted, the position. Pauli would form around himself at the ETH a group of physicists who would make major contributions to the early development of the quantum theory of solids. In particular, Heisenberg's half-year trip to the United States in the spring of 1929¹⁹ led to important additions to Pauli's group. Bloch, after completing his dissertation, came in the winter semester of 1928–1929 to succeed Ralph Kronig as Pauli's *Assistent*.²⁰ Peierls, sufficiently advanced now to begin a doctoral thesis, came on Heisenberg's recommendation (Peierls, 1963, 1980).

A number of visiting fellows, who might otherwise have gone to Heisenberg, also went to Zurich; Landau, J. Robert Oppenheimer, Rabi, and Léon Rosenfeld worked in Pauli's group for portions of 1929 (Pauli, 1929a; also note b, Pauli, 1929b, WP1, p. 497). "I have now a rather bustling operation here in Zurich," Pauli (1929b) wrote in May 1929 to Sommerfeld. For Pauli, like Heisenberg, solid state was in these years a prime testing ground for quantum mechanics, and his description in his letter to Sommerfeld of the problems of those working with him at this time illustrates the extent to which Pauli regarded solid state as an area in which to employ students and research fellows: "Mr. Bloch is at present occupied with working out a theory of superconductivity . . . Mr. Peierls is working on a theory of thermal conductivity in solid bodies." Pauli (1923, 1925b) had himself struggled several times with applying quantum mechanics to the problem of thermal conductivity.

The physicists in Leipzig and Zurich formed a family. Debye in Leipzig and Paul Scherrer in Zurich could look back on a collaboration of more than 10 years (at Göttingen as well as Zurich). Heisenberg, Pauli, and

Wentzel had all studied with Sommerfeld at the beginning of the 1920s, as had Debye some 20 years in the past. Sommerfeld remained a father figure to his students: "Hopefully you will continue to run a Kindergarten for physics babies like Pauli and me!" Heisenberg (1929a) wrote to Sommerfeld in 1929.

While the interaction between Leipzig and Munich and between Zurich and Munich was formed by the relationships of the pupils, Heisenberg and Pauli, to Sommerfeld, the interaction between Leipzig and Zurich, by contrast, was shaped by the less formal and more intense relationship of the two peers. Particularly during the 1920s, these two interacted deeply in science, yet their characters could scarcely have differed more: Heisenberg openly friendly, an early riser as ambitious in sports as in science, and a nature lover; Pauli aggressive, often woundingly critical, moody, and preferring night clubs over morning outings.²¹ But they shared their enthusiasm for physics, and continued their active exchange as heads of their own institutes.

Some weeks before his return to Leipzig, Heisenberg wrote Pauli from America: "So you want to have Peierls as your Assistent next semester? To me that naturally seems completely correct in principle, but I think you should also send good physicists to me in L[eipzig] as compensation; I would especially like Bloch to come to L again for a while. Can this be done? I would find it very nice if we could arrange such an exchange of physicists between Zurich and L, but it must be mutual, for otherwise I would be left all alone" (Heisenberg, 1929b). As Heisenberg requested, Bloch went to Leipzig as Heisenberg's *Assistent*, while Peierls became Bloch's successor in Zurich as Pauli's *Assistent*. Their *Habilitation* studies²² in the area of quantum mechanics of solids strongly influenced for both the course of their careers.

At Pauli's suggestion, Peierls took up further problems in solid-state physics. Maintaining an interest in lattice vibrations in an anharmonic crystal, Pauli suggested that Peierls study heat conduction in nonmetallic solids (Peierls, 1977). Peierls's work on heat conduction in insulators, completed in late October 1929, became his doctoral thesis, submitted to Leipzig (Peierls, 1929c). In it Peierls carried out a critical analysis of how lattice vibrations come into thermal equilibrium at low temperatures; introducing the important [and soon to become controversial (see Brillouin, 1962)] concept of Umklapp processes, he found that in a pure material, conservation of "crystal

¹⁹His visit to the University of Chicago on this trip resulted in the well-known lecture volume (Heisenberg, 1930a).

²⁰Kronig was Pauli's *Assistent* in the summer of 1928. See Kronig, 1982, as well as WP1 (Hermann *et al.*, 1979), note on p. 442, and Kronig, 1960.

²¹WP1 (Hermann *et al.*, 1979) is a helpful source of the relationship between these two. David Cassidy furnishes a striking description of their opposite characters in his forthcoming biography of Heisenberg. We thank Cassidy for showing us parts of his manuscript prior to publication. See also Daniel Serwer (1977) and Bleuler (1984).

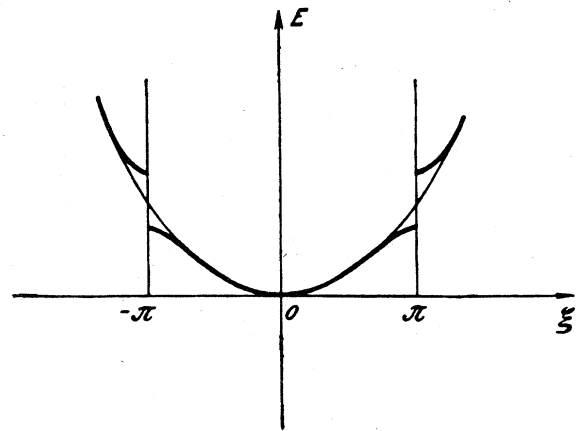
²²The *Habilitationsschrift* put one on the road to becoming a Professor, and gave one the privilege of announcing formal lectures and inviting students to them.

momentum" of lattice vibrations implies that as the Umklapp processes become frozen out with decreasing temperature, the thermal conductivity rises exponentially. Peierls immediately reacted to his discovery by asking "Could that be superconductivity?"; an inquiry Pauli encouraged with the remark, "Well, if this can explain superconductivity, then you can certainly have your Habilitation;" however, "it didn't, of course" (Peierls, 1981a).

Peierls then turned to applying his arguments to metals. In a paper submitted to the *Annalen der Physik* six weeks after his thesis paper (Peierls, 1930a), he points out the role of Umklapp processes in keeping the lattice vibrations in equilibrium and limiting the electrical and thermal conductivity at low temperatures. Unlike Bloch, who assumed the lattice vibrations to remain in thermal equilibrium, Peierls writes down coupled Boltzmann equations for both the electron and lattice vibration distributions. He also uses, for the first time, the "rigid-ion" approximation to describe the interaction of electrons with oscillating ions (thanking Pauli for pointing out that this is not an exact procedure).

The first section of this rather longish paper, on the two limiting cases—tight and weak binding—for electrons in solids, was to play a seminal role in the further development of band theory. Peierls's earlier explanation of the anomalous Hall effect depended on the negative curvature of the electron energy, as a function of wave number, near the top of the band. But so far only the tight-binding approximation had been examined, and this was clearly not a good approximation for real metals (Peierls, 1977), for, as Bloch had recently argued, the magnetic susceptibility of ordinary metals indicated a density of states at the Fermi surface (in modern language) more nearly that of free electrons (Bloch, 1929a). The basic problem bothering Peierls was how to connect the tight-binding limit, with its novel and apparently important structure, to the free-electron limit, closer to experiment, which does not have such negative curvature. Examining the case of weakly bound electrons in one dimension, he found, as every solid-state student now learns, that whenever two free-electron states, separated by a reciprocal lattice vector, have an energy difference comparable with the potential matrix element, gaps and hence negative curvature appear in the spectrum, disappearing only when the electrons are exactly free. "I still remember the excitement . . . how thrilled I was to see that" (Peierls, 1963). His starting equations were, as we noted, just those written down by Bethe in his thesis, although Peierls by this point, having the experience of his Hall effect work, knew what question to ask of the model calculation.

His result, which he illustrated with the classic figure, reproduced here (Fig. 1), thus established the concept of the band gap as characteristic of electrons in solids (Peierls, 1930a, Fig. 1, p. 126). Peierls, in looking back on this work, wrote recently, "Few pieces of work have given me as much pleasure as this discovery, which required only a few lines of calculation, both because it satisfied me that the nature of the Bloch bands was now qualita-



Energiewerte erster Näherung

FIG. 1. Peierls's (1930a) illustration of the formation of band gaps in a weak periodic potential.

tively the same all the way from tight binding to almost free electrons, and because of the neat method of approximation I had invented" (Peierls, 1980, pp. 31 and 32).

Actually, the transition from tight to weak binding, and the existence of gaps had been worked out by M. J. O. Strutt, in mid-1928 (Strutt, 1928a),²³ for a sinusoidal potential in one dimension, the Mathieu problem, although this work did not receive the immediate attention of solid-state theorists. The first application of Strutt's approach was made by Morse, soon after receiving his Ph.D. at Princeton in June 1929 under the supervision of Karl Compton (Morse, 1930). Working during the summer for Davisson at the Bell Telephone Laboratories on the interpretation of the experiments of Davisson, Germer, G. P. Thomas, and others on electron diffraction from metal surfaces (Morse, 1977, see pp. 92–100), Morse began a general analysis of the solutions of the Schrödinger equation for an electron in a periodic potential. The starting point, as he acknowledges in his paper on this work, was equations similar to those in Bethe's thesis; and he derived the important conclusion that "the periodic variation of the potential inside the crystal creates bands of forbidden energies inside the crystal, even for electron energies greater than the maximum potential energy, a somewhat surprising result." Morse goes on in his paper to work out in detail the example of the separable potential in three dimensions that is a sum of cosines, whose solution reduces to the one-dimensional Mathieu problem studied by Strutt; in applying this exactly soluble model to the Davisson-Germer experiments he thus made the first explicit connection between the band structure of electrons in solids and the diffraction of electrons imping-

²³The paper of M. J. O. Strutt (1928a) follows two earlier ones, Strutt (1927) and (1928b), applying the Mathieu equation to various problems in modern physics.

ing on solids. Not having completed this work at Bell, he continued it at Princeton in the fall, and “after a great number of further computations, done by myself on the department’s desk calculating machine,” submitted his paper to the *Physical Review* in April 1930 (Morse, 1977, pp. 97 and 98). Although by this time Peierls’s paper containing the weak-binding calculation had appeared, and Morse included a reference to it, Peierls’s work does not appear to have had a significant influence on Morse’s.

Meanwhile, Brillouin, who had worked under Sommerfeld in 1912–1913 and was now a lecturer at the *École Supérieure d’Electricité* on radio and Professor of Science at the University of Paris, became interested in problems of electrons in solids, in preparing his book, *Les Statistiques Quantiques et Leurs Applications*, published in early 1930 (Brillouin, 1930). Brillouin had extensive experience in both statistical mechanics and mathematical problems of wave propagation, including development of the WKB method in quantum theory (Brillouin, 1962),²⁴ but at the time of this book had not yet made independent contributions to the quantum theory of metals. The chapter on the mean free path of electrons in solids contains a description of energy bands, but only a rather mathematical one based essentially on Strutt’s one-dimensional calculation. Brillouin also appeared to have learned of Morse’s work prior to the publication of his own, most likely during the previous summer on his trip to the United States to lecture at the Michigan Summer School in Ann Arbor.²⁵ But Peierls’s work on the Hall effect and the weak-binding calculation are not noted, although the chapter ends with a passing last-minute reference to Peierls’s critique, in the same paper (Peierls, 1930a) as his weak-binding calculation, of Bloch’s theory of electrical conductivity (Brillouin, 1930b, p. 294).

The full application of Peierls’s ideas on formation of energy gaps to realistic solids was begun by Brillouin, working alone in Paris in the summer of 1930 (Néel, 1981). In two papers, presented on his behalf by J. Perrin to the Académie des Sciences at its 28 July meeting (Brillouin, 1930c, 1930d), he first generalizes Peierls’s (1930a) result to show that in three dimensions the surfaces of discontinuities in energy versus wave number for nearly free electrons form polyhedra in momentum space—the Brillouin zones—and then, counting states, argues that each zone corresponds to a single atomic state and shows how to transform from the extended zone scheme to the fundamental zone. He makes the connection, as did

Morse, between the conditions for a discontinuity in the energy and for Bragg scattering, one he remarks that Peierls did not make in his paper.

These new ideas were synthesized a week later in a paper, submitted to the *Journal de Physique et le Radium* (Brillouin, 1930e), which is the fundamental source of the established technology of the geometry of Brillouin zones. Going beyond his recent book, which relied on exact and one-dimensional solutions, he realizes here the generality of energy bands. He also describes the curvature of the energy-wave number relation in terms of an effective mass m^* , remarking that m^* can be negative, although he does not make contact with Peierls’s Hall-effect work. The paper concludes with an attempt to establish a phenomenological connection between “propagation anomalies” (i.e., the Fermi surface reaching a zone boundary) in polyvalent metals and lower electrical conductivity in these materials, but in the absence of any dynamical theory his arguments emerge as inconclusive.²⁶

Brillouin later described his recollections of this fertile summer: “At first I did not realize that I was doing something that might become really important. I did it for the fun of it, following my own line of investigation by sheer curiosity and taking a great deal of pleasure in making carefully all the drawings needed to explain the properties of these *Brillouin Zones*.” He was also pleased by the response to his work and was “especially proud of a very affectionate letter from my old teacher, Sommerfeld, who praised warmly my contribution and said that he was so happy to be now able to understand clearly the interconnection between isolated atomic electronic levels and free electrons in metals.” Unfortunately during the war this letter, along with most of Brillouin’s prewar papers, “mysteriously disappeared” (Brillouin, 1962, pp. 17 and 18).

Brillouin took the opportunity of the translation of his book into German a year later (Brillouin, 1931) to make extensive revisions of the chapter on electrons in solids, including (nearly verbatim) his *Journal de Physique* paper and bringing the discussion of the electrical conductivity up to date by including Bloch’s recent correction (the T^{-5}) of the low-temperature dependence found in his thesis; he also used the occasion to express deep concern about the reality of Peierls’s Umklapp processes, arguing

²⁴After leaving Germany in 1933, Bloch became acquainted with Brillouin during a short stay in Paris, during which Bloch lived with the Langevin family. Bloch recalls that Brillouin was a most interactive and lively person, with “a very sound grasp on reality” through his engineering background, and a great expert on waves, especially radio waves (Bloch, 1981).

²⁵Brillouin’s complete citation on p. 264, “Morse, *Phys. Rev. t.* (1929),” indicates that he had not at the time of writing of his book seen the final version of Morse’s paper.

²⁶The richness of the geometries of Fermi surfaces would first be studied by Bethe for the 1933 review (SB), where electron energies computed using Bloch’s tight-binding model were used to draw the ideal Fermi surfaces for a number of simple lattices—cubic, face-centered cubic, and body-centered cubic (SB, p. 401). For the drawings Sommerfeld and Bethe commissioned R. Rühle, who had done the figures for Jahnke and Emde’s tables of functions. Bethe recalls that “it was clear to me . . . that it made a great difference whether [the Fermi surfaces] were nearly a sphere or were some interesting surface,” and that for the problem of magnetoresistance, “it was very important how anisotropic the Fermi surface is” (Bethe, 1981a).

that since “these processes can be represented as a simple superposition of a Bragg reflection and a normal scattering I have the impression that one must leave the Umklapp processes out of the theory . . .” (Brillouin, 1931, pp. iv and v).²⁷

By the end of 1930 all the crucial pieces of band theory were waiting to be assembled. The existence of band gaps was well understood. In addition to the Bloch tight-binding and Peierls’s weak-binding calculations, one had Morse’s general arguments, as well as Strutt’s exact example. At year’s end Kronig and William Penney submitted for publication their simple analytically soluble one-dimensional model of a periodic square-well potential (Kronig and Penney, 1931),²⁸ which verified from another point of view the general features of the quantum states and energies of electrons in solids. Furthermore, the concept of holes, although not yet clearly described, was implicit in Peierls’s work on the Hall effect. These ideas would be fused in 1931 by Wilson in two classical papers on semiconductors (Wilson, 1931a, 1931b).

Wilson’s interest in solid-state physics grew out of his attempts, starting in 1929 while a research fellow at Emmanuel College in Cambridge, to explain Peter Kapitza’s recent experimental discovery there that the resistance of metals in strong magnetic fields increases linearly with the field. The problem of the influence of magnetic fields on electrical conduction, or “magnetoresistance,” being outside the scope of the simple Sommerfeld theory (which provided no theory of the electron mean free path), had become in this period an important test problem in the theory of metals and attracted the attention not only of Wilson, but of Bloch in 1928 (Bloch, 1929a), Peierls (1930b) and Landau in 1930 (Landau, 1930), and Bethe in 1931 (Bethe, 1931a). Finding Cambridge, under Rutherford’s influence, “highly concerned with nuclear physics” (Wilson, no date), Wilson obtained a Rockefeller Foundation Fellowship to go to Leipzig, where he could join Heisenberg and Bloch, then *Assistent*, who were very interested in magnetism. Others in Leipzig at that time were Hund, Teller, and Debye.

Immediately after Wilson’s arrival in Leipzig the first week of 1931, Heisenberg, sensing the significance of Peierls’s work (1930b, 1931) on effects of magnetic fields in metals, asked Wilson to deliver an explanatory colloquium. In Wilson’s words, “There were two problems, . . . one was that I had Peierls’ papers and didn’t really understand them, and secondly . . . to give a seminar in German at which I would be cross-questioned back

and forth would be a bit of an ordeal.” Wilson recalls being particularly impressed with Heisenberg’s, as well as Debye’s, ability to see through problems by very simple physical arguments. Contrasting the atmosphere at Leipzig with the more mathematical approach he found at Cambridge, “particularly with Dirac and [R.H.] Fowler,” he remarked, “I’d been to something like four seminars before I had to give mine, and on nearly every occasion Heisenberg would stop whoever was talking and say, ‘This is alright, this is mathematics, but not physics’ and he would say ‘Wie kann man das physikalisch anschaulich machen?’ [How can we make that physically intuitive?]” (Wilson, no date).

Wilson sat down to study the Peierls and Bloch papers in detail. To him the problem was that Bloch, in showing that tightly bound electrons could in fact move through the lattice, had “proved too much” (Wilson, no date), that all solids would be metals. Were insulators simply very poor conductors? However, implicit in Peierls’s papers on the Hall effect lay the clue, not carried further by Peierls, that a filled band would carry no current. “Suddenly one morning” Wilson realized that “I’ve been looking at it all wrong, of course it’s perfectly simple . . .” (Wilson, no date): he could make the basic Bloch-Peierls theory of electrical conductivity “intuitively more plausible if one assumed that the quasi-free electrons, like valence electrons in single atoms, could form either open or closed shells” (Wilson, 1980, p. 45). Here, finally, was the answer to the old question of the difference between metals and insulators: insulators have completely filled bands, while metals have partially filled bands, a situation nicely summed up in Wilson’s remark in his first paper on semiconductors, “we have the rather curious result that not only is it possible to obtain conduction with bound electrons, but it is also possible to obtain non-conduction with free electrons.”

Wilson told his idea to Heisenberg who said, “I really must get Bloch in” (Wilson, no date). Wilson recalls Bloch’s reply after hearing his arguments, “No, it’s quite wrong, quite wrong, quite wrong, not possible at all,” for Bloch had, since his thesis, assumed that the difference between metals and insulators was only quantitative, determined by the size of the electron overlap integral, which measures the ease with which an electron can hop from atom to atom. Attempting to refute Wilson, Bloch pointed out that although solids formed of monovalent elements would have only half of the uppermost band filled, and would be metals, the divalent alkaline earths would have just enough electrons to fill the top band exactly and hence should be insulators.

But the following day Wilson was able to point out to Bloch that, unlike in an idealized one-dimensional lattice, the bands in a three-dimensional solid can in fact overlap, so that rather than the bands being filled in the order of the corresponding atomic states, several bands in a polyvalent material could be partially filled. “It therefore followed that an elemental solid . . . with an odd valency had to be a metal, whereas elements with an even valency might produce either a metal or an insulator” (Wilson,

²⁷However, by 1933 Brillouin would accept the validity of the Umklapp process. See Brillouin (1933a, 1933b, 1933c).

²⁸Kronig and Penney (1931) note that van der Pol and Strutt (1928) had previously considered the special case of the periodic square-well problem, but in a classical physics context. On Kronig’s suggestion, Penney, whom he had met during a visit to Cambridge and London in 1929–1930, came in 1930 to Groningen, where Kronig had obtained a permanent lectureship (Kronig, 1982).

1980, pp. 45 and 46). After a week Bloch was convinced, and he would soon summarize Wilson's concepts in a short paper given at the seventh Deutschen Physikertages in Bad Elster, in September 1931 (Bloch, 1931); among his illustrations were those reproduced here as Figs. 2(a) and 2(b), showing the difference between a metal and an insulator, respectively, and Fig. 2(c) showing an impurity level in a semiconductor.

Wilson proposed to Heisenberg that he broaden his colloquium to deal more generally with bands. They decided that Wilson should give two colloquia, spaced approximately 3 months apart, and should also consider semiconductors, which the new concept of electrons in filled or unfilled shells might also illuminate. Heisenberg had become interested in semiconductors through the experimentalist B. Gudden, at Erlangen (Wilson, 1981); but at the time, knowledge about semiconductors was so scant that it was uncertain whether they even existed. While Grüneisen in his 1928 review of metallic conductivity in the *Handbuch der Physik* (Grüneisen, 1928) had distinguished semiconductors as a class of solids with a pronounced minimum in their resistance as a function of temperature, it was not clear (as Wilson notes in the introduction to his second paper on semiconductors) that this behavior might not simply be caused by oxide surface layers on otherwise metallic substances.²⁹

In his first colloquium, in February, Wilson described his theory of the difference between metals and insulators and put forward a simple picture of a semiconductor as an insulator with a gap between what we now call the valence and conduction bands, small enough that electrons could easily be excited across the gap at finite temperature. The details were written up in the *Proceedings*

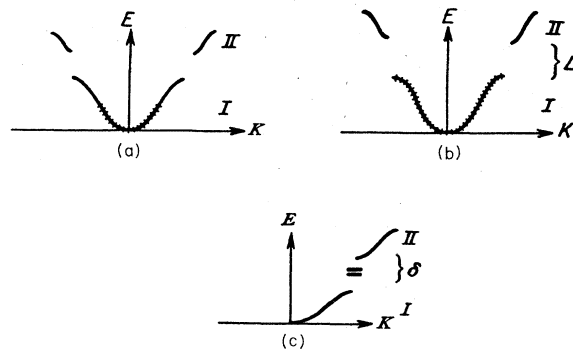


FIG. 2. Wilson's picture of the energy bands in (a) a metal, (b) an insulator, and (c) an impurity level in a semiconductor, as portrayed by Bloch (1931).

of the Royal Society (Wilson, 1931a; communicated by Dirac, since Fowler was in the United States at the time). Referring in his paper to the previous work of Bloch, Peierls, Morse, Brillouin, and Kronig and Penney as indicating the general existence of energy bands, he gives a very clear review of the weak- and tight-binding approximations, and, in particular, calculates (for the first time) the wave functions and energies of *p*-state bands in tight binding. (Bloch's original calculation was an *s*-state band.) The following discussion, detailing his earlier arguments to Bloch, shows how overlap of *s* and *p* bands in alkaline earths can lead to their being conductors; this section is remarkable as the first use of band theory to distinguish qualitatively the properties of realistic solids. Wilson then goes on to describe his simple model of a semiconductor, showing that the chemical potential lies halfway in the band gap, and calculating the specific heat, spin paramagnetism, and electrical conductivity as limited by emission or absorption of a "sound quantum" by electrons. He concludes by remarking that while the interpretation of the experimental results on semiconductors is still difficult, and even their very existence "remains an open question . . . the theory is on the right lines."

This spring in Leipzig was also the time that Heisenberg wrote his paper on the Pauli exclusion principle (Heisenberg, 1931a) in which, as an outgrowth of his interest in the theory of magnetic effects in metals, particularly Peierls's (1929a) Hall-effect paper, the concept of the solid-state "hole" first appears. In fact, Heisenberg's paper was received by the *Annalen der Physik* the day after Wilson's was received by the Royal Society. It is noteworthy that while he had many discussions with Heisenberg on electrons in metals, Wilson in his paper never treats the unoccupied states in the valence bands as hole degrees of freedom, but rather works in terms of the electron states; nor does Heisenberg's paper refer to Wilson.

Between his first and second colloquia, Wilson learned through Heisenberg of Gudden's view that semiconductor behavior was always caused by impurities (Wilson, 1981). The second colloquium, which was attended by a group of

²⁹Indeed, accepted experiments of H. J. Seemann (1927) and Schulze (1931) indicated that pure silicon, in the absence of oxide films, was a good metal. That such metallic silicon, when covered with an oxide layer, could exhibit the observed increase of conductivity with temperature might be explainable, as Wilson further remarks (Wilson, 1931b), by Frenkel's theory (1930) of transmission of thermally activated electrons across oxide films, a theory that Frenkel also applied to explain the temperature dependence of the conductivity of granular thin films with the charming analogy, "This relation can be illustrated by the fact that the gaps between adjacent rails in a railway line decrease in the summer and increase in the winter time, and not vice versa."

The "canard" that silicon was a good metal would, as Wilson noted (1980), linger through the prewar period [for example, in Wilson (1939), p. 44], and even into recent times. [The 51st edition of the *Handbook of Chemistry and Physics* (1970) lists (p. F190) the electrical resistivity of Si at 0°C as only six times that of Cu at 20°C. The properties of silicon first began to be clarified by wartime research, particularly by Seitz and co-workers at the University of Pennsylvania, and J. Scaff, R. Ohl, and others at Bell Telephone Laboratories (Hoddeson, 1980; Seitz, 1981; Seitz, as quoted in Mott, 1980b, pp. 63 and 64).

experimentalists from Erlangen headed by Gudden and which lasted several days, addressed the role of impurities, and the detailed model is described in Wilson's second paper (1931b). The paper begins with a brief argument making an interesting contact between conductivity and optical properties, namely, that while the experimental conductivity of cuprous oxide indicates, according to his earlier paper, an excitation energy of order 0.6 V, optical absorption implies an intrinsic band gap of order 2 V, and hence "the observed conductivity . . . must be due to the presence of impurities." The model he considers is an impure insulator in which an electron associated with an impurity has an energy in the band gap close to the conduction band, so that it can be excited thermally into the conduction band. [This is the donor model; the concept of acceptor levels appears first to have been introduced by Peierls (1932a) and clarified by W. Schottky in 1933 in explaining experiments of F. Waibel on copper oxide (Schottky and Waibel, 1933, especially pp. 862 and 863).] Such an impurity level is shown in Fig. 2(c) (from Bloch's talk). After determining the chemical potential, Wilson calculates the electrical conductivity and points out that the Hall coefficient is given by the classical formula, inversely proportional to the conduction-electron density. Finally, fitting to the Hall coefficient of cuprous oxide observed by Vogt in 1930, Wilson deduces an impurity concentration of order 10^{17} per cubic centimeter, "conclusive proof that the conductivity is due to impurities and is not intrinsic."

Wilson went on later in the year to apply band concepts in developing a pioneering theory of rectification at a metal-to-semiconductor junction (Wilson, 1932), in which electrons penetrate, by quantum-mechanical tunneling, a symmetric potential barrier in the transition layer between the metal (here copper) and the oxide. The positive direction of the electron current was predicted to be from the metal to semiconductor, unfortunately opposite to later experiment, as Wilson acknowledged some years after in his well-known book on semiconductors (Wilson, 1939).³⁰ Nevertheless this paper would provide the basis of attempts to explain experimental work on rectification during the 1930s (Hoddeson, 1981), although at the time, as Wilson reflects (1981), not much notice was taken of it by physicists.

Wilson, by bringing together the elements of band theory in a simple conceptual picture, closes this chapter in the development of the fundamental quantum theory of solids. He emerges as an important figure in the transition of solid-state theory from its early conceptual to its later practical orientation, for not only did his model make it possible to begin to approach realistic solids, but, because his papers were so clear, they would be widely

read by subsequent generations of experimental and theoretical researchers.

III. MAGNETISM, 1928—1933

A second area of solid-state phenomena for which the quantum theory would provide the necessary theoretical building blocks was magnetism—a development in many ways paralleling that of electrical transport. The seminal papers on paramagnetism by Pauli (1927), diamagnetism by Landau (1930), and ferromagnetisms by Heisenberg (1928b) were, like Sommerfeld's pathbreaking work on conduction (1927), based on drastically simplified models, shorn of all irrelevant (and realistic) detail to bring out the essential physical ideas. Only with application of the understanding of electrons in solids reached through band theory, developed out of concern with electrical transport phenomena, could detailed comparisons be made between theory and magnetic properties of solids.

A. Paramagnetism and diamagnetism

Pauli's work on paramagnetism, carried out in late 1926 and published in 1927 (Pauli, 1927), was pivotal not only to the development of the understanding of electrical transport, as discussed above, but to the theory of magnetism in solids. Our earlier article (Hoddeson and Baym, 1980) gave background for Pauli's study of this problem (see also WP1, pp. 19, 53, 55, and 56, and Pauli, 1920), and we do not repeat the history here except to recall that while Pauli had a longstanding interest in magnetic problems, his motivation was not to explain paramagnetism as such, but to use the phenomenon to answer the more fundamental question of which statistics applied to matter.

Pauli concludes his paramagnetism paper with a brief numerical comparison of his formula for the paramagnetic susceptibility with then available observations of the susceptibility of the alkali metals Na, K, Rb, and Cs. The difference in all cases appeared to imply the presence of a weak diamagnetism, and in Rb and Cs a diamagnetism of comparable size to the paramagnetism, but he makes no attempt to understand the residual diamagnetism in terms of contributions from the bound core electrons and the conduction electrons.

The diamagnetism of ionic cores was by this time reasonably well understood. The fundamental formula for the susceptibility, given by Pauli himself (1920), after Langevin, as a sum over the mean-square radii of the electron orbits, gave results comparable to measured atomic and ionic susceptibilities (see the review of Van Vleck, 1932, especially pp. 100–102). On the other hand, according to the classical argument given by Bohr (1911), Lorentz (1914, p. 188), and "Miss" van Leeuwen (1919, pp. 49–51; 1921), the diamagnetic susceptibility of a free-electron gas must vanish, and thus there should be no conduction-electron diamagnetism. Mathematically this result emerges from the fact that the energy of an electron in a magnetic field is proportional to its velocity squared,

³⁰Wilson's book (1939) was written as a text for experimental physicists; see Wilson (1981). The effort of theorists and experimentalists to communicate about physics was a relatively new trend in Great Britain.

independent of the field. Bohr and Lorentz also explained this phenomenon physically by noting that while electrons in magnetic fields move in circles, those that bounce off the boundary slowly circulate around the edge in a direction opposite to the orbital motion of the interior freely circulating electrons, producing a magnetic moment that exactly cancels that of the interior electrons.

The question we may ask at this point is whether, after Pauli's paper, there was any evidence for a nonzero conduction-electron diamagnetism. Had Pauli taken available experimental numbers for the ionic core diamagnetic susceptibilities³¹—and even by the 1930 Solvay Conference he had not done so, in print at least—he would have concluded that the susceptibility of Rb and Cs could essentially be explained as a sum of his paramagnetic contributions plus the measured core diamagnetism. However, for Na and K, he would have found the surprising result that what was needed was further paramagnetism [an effect arising from the enhancement of the paramagnetic susceptibility from the Pauli value by electron-electron interactions, which, through the Pauli principle, favor electron spin alignment (Sampson and Seitz, 1940)]. For none of the alkalis was there any need to invoke conduction-electron diamagnetism. The only hint that such diamagnetism played a role was the enormous diamagnetic susceptibility of bismuth (as well as antimony (de Haas and van Alphen, 1930)).³²

By sorting out the magnetic contribution of the spins in the alkalis, Pauli indirectly set out the problem that a theory of diamagnetism would have to explain. The invention of quantum mechanics provided new tools for studying this phenomenon, which Bloch recalls attracted theoreticians of the period because "it was a very clean quantum mechanics problem" (Bloch, 1981). According to Peierls (1981b), "it was part of one's general interest in metals." The first published attempt to deal with conduction-electron diamagnetism from a quantum-mechanical point of view was by Francis Bitter (1930) at Caltech in late 1929, in which he essentially computes the expectation value of the Pauli-Langevin atomic diamagnetism formula using free electronic wave functions spread over a unit cell. Such an estimate must, on dimensional grounds, be reasonable, and indeed, Bitter's results for the diamagnetic susceptibility are ~ 1.25 – 2 times the correct answer. It is worth noting that Bloch's 1928 theory of electrons in solids appears not to have influenced Bitter, although Houston had returned to Caltech

from Leipzig in the fall of 1928.

The full quantum-mechanical problem of electrons orbiting in a magnetic field was solved by Landau in his paper (1930) on diamagnetism. In 1929 Landau, at age 21, went on a 2-year trip, supported at first by the Soviet People's Commissariat of Education (Narkompros) and then as a Rockefeller Fellow, to European research centers, including Zurich, Copenhagen, and Cambridge, as well as stops in Germany (among them Berlin and Leipzig), Holland, and Belgium (Berestetskii, 1958; Livanova, 1980). How Landau became involved in this problem is unclear. Although he had not published before on solid-state physics, he was a student in Leningrad, which felt Paul Ehrenfest's influence from his 5-year prewar stay there, where A. F. Ioffe was a leading solid-state experimentalist. Motivation possibly came from Ehrenfest, whom Landau met in Berlin in late 1929 (Livanova, 1980), at the time Ehrenfest, in response to a published note by Raman (1929a, 1929b) and sundry letters, was republishing in German his 1925 paper (Ehrenfest, 1925, originally in Dutch, and Russian) on the diamagnetism of bismuth (Ehrenfest, 1929). According to Peierls, who became acquainted with Landau on his first visit to Zurich, Landau had the problem well under control on his arrival there in late 1929; at this time he was going about solving everything for himself, and most likely had worked out the quantum-mechanical problem of an electron in a magnetic field for its own interest (Peierls, 1980). However, Landau delayed submitting his paper until May 1930 when he was at the Cavendish Laboratory, during which time he had discussions about experiment with Kapitza, whom he met for the first time.

In his paper Landau, beginning with the remark that, "It has until now been more or less quietly assumed that the magnetic properties of electrons, other than spin, originate exclusively from the binding of electrons in atoms," proceeds to show, by a clever algebraic technique, that a quantum-mechanical electron in a uniform magnetic field \mathbf{H} is described by a harmonic oscillator of frequency $e\hbar H/mc$, the Larmor frequency. He then turns directly to the statistical mechanics of a degenerate free-electron gas in a field, and derives the famous result that the diamagnetic susceptibility of the gas in a weak field is exactly $-\frac{1}{3}$ of the Pauli spin susceptibility, rather than zero as in the classical theory. He also notes that the diamagnetic moment should have a strong periodicity in the field, an effect whose experimental discovery W. J. de Haas and P. M. van Alphen would report in their communication to the Royal Dutch Academy of Sciences at the end of 1930 (de Haas and van Alphen, 1930). Although de Haas and van Alphen give a reference to Landau's paper in their communication, they do not make any contact with his theoretical prediction of the periodicities. In fact, Landau himself despaired of observing the effect, suggesting in his paper that inhomogeneities would wash it out. The paper concludes with a brief qualitative attempt to understand Kapitza's recent experiments on magnetoresistance, and an acknowledgment of his discussions with Kapitza.

³¹For example, Sucksmith (1926), who notes the lack of "a satisfactory theory explaining the fact that a large number of elements exhibit a paramagnetic susceptibility independent of temperature," and the apparent smallness of the measured susceptibilities below 500°C, compared with their values at the boiling points.

³²Pre-band-theory attempts to explain the diamagnetism of bismuth were based upon bound electrons having orbits that embraced several atoms (Ehrenfest, 1929a, 1929b).

David Shoenberg recalls³³ that Landau explained to him “many years later” how he came to make his remark about the practical limitations on seeing the oscillations: “He replied that since he knew nothing about experimental matters he had consulted Kapitza whom he was visiting at the time, and Kapitza had told him that the required homogeneity was impracticable.” de Haas and van Alphen in fact carried out their experiments on single crystals of bismuth;³⁴ what Landau could not realize was that the lattice effects that would be shown to enhance the diamagnetic susceptibility of bismuth so dramatically also greatly increase the oscillation period and therefore the observability of the effect in bismuth.

Landau’s results were immediately accepted. As Peierls recalls (1980), “neither Pauli nor I had any doubt in feeling confident that Landau had got the right argument.” Pauli described Landau’s work in his Solvay talk of October that year (Pauli, 1932),³⁵ referring to it as being verbally communicated, and gave in addition the result for the case of nondegenerate statistics and arbitrary-strength field, where again the weak-field susceptibility is $-\frac{1}{3}$ the spin contribution. Kapitza, in the discussion of Pauli’s talk, remarked, “Landau’s new theory, in which the free electrons contribute to the magnetism of the substance, gives us great hope to see all these phenomena [such as effects of impurities and imperfections] explained by a common picture” (Pauli, 1932, p. 243).

Just how Landau’s picture modified the Bohr-Lorentz

argument for the vanishing of the diamagnetic susceptibility was not immediately clear. A peculiar feature of the Landau diamagnetic susceptibility of a degenerate electron gas, one that does not appear to have been discussed in the literature of the time, is that even though it is nonzero as a result of quantization of the orbits, it is independent of Planck’s constant \hbar . How then did one recover the classical limit? The form derived by Pauli for nondegenerate statistics (proportional to \hbar^2 , and thus going to the classical vanishing value as \hbar goes to zero) showed more clearly how the nonvanishing result was a quantum phenomenon. The independence of \hbar in the Landau result arises from the fact that degeneracy reduces the nondegenerate result, as in paramagnetism, by a “density-of-states” factor proportional to T/T_f , where T is the temperature and T_f the Fermi temperature; since T_f is proportional to \hbar^2 , the dependence on \hbar curiously cancels out, a structure obscured in Landau’s direct solution of the degenerate-gas problem.

The relation between Landau’s theory and the classical Bohr-Lorentz argument was soon addressed by C. G. Darwin in Edinburgh, and by Teller, then Heisenberg’s *Assistent* in Leipzig. Landau, by calculating the partition function rather than the magnetic moments of the electron states, did not have to deal explicitly with the question of the diamagnetic contribution of the electrons near the boundary compared with those in the interior. To satisfy himself that Landau’s result can be derived in terms of the electron magnetic moments, Darwin (1931; talk in Solvay, 1932; see also Seemann, 1929) studied the electron orbits in the exactly soluble model in which the container is replaced by a harmonic-oscillator well, and he was able to recover “the features of Bohr’s argument about the creeping of the electron round the boundary wall,” and to see how the cancellation between the two contributions no longer occurred in the quantum theory. Teller (1931),³⁶ on the other hand, working with a more realistic confining potential, evaluated the statistically averaged magnetic moment contributions of the boundary and interior electrons to show how Landau’s result emerged.

The nature of the paramagnetism and diamagnetism of conduction electrons was now understood in principle. However, the free-electron result that the diamagnetism equalled minus one-third of the paramagnetism offered no insight into the vexing anomaly of bismuth (Peierls, 1932b), which could not be explained in terms of core diamagnetism alone. As in the problem of electron transport, meaningful comparison of theory with experiment required including effects of the lattice. Landau, in his paper, understood that even in a lattice the motion of the

³³We thank Dr. Shoenberg for kindly showing Paul Hoch this manuscript, and thus making it available for use by the International Project on the History of Solid State Physics. See also Shoenberg (1965, 1978). Shoenberg tells (1965, p. 667) how around 1937 he visited Kapitza’s laboratory in Moscow and observed the oscillatory variation in bismuth with Landau right on the spot for detailed interpretation. In this way, they made what Shoenberg believes was the first determination of the Fermi surface.

³⁴The single crystals of bismuth used in these experiments were grown by L. V. Shubnikov, who, like Landau, on graduating from Leningrad Polytechnical University was sent by the Narkompros on an extended scientific visit to western Europe. Between 1926 and 1930 he worked in Leiden with de Haas. By improving upon a method of Kapitza’s, Shubnikov succeeded in producing single crystals of bismuth, which made possible the discovery of the Shubnikov–de Haas effect, the periodic change of electrical resistivity in bismuth as a function of magnetic field at low temperatures; this effect helped to motivate the de Haas–van Alphen experiments. Whether Landau made contact with Shubnikov during his 1929–1930 visit to Europe is unclear (Balabekyan, 1966).

³⁵Pauli (1932). Landau is referred to on pp. 186 and 238. The Sixth Solvay meeting—attended by Kapitza, Debye, Heisenberg, Brillouin, Dorfman, Weiss, Darwin, Stern, Brillouin, Langevin, Dirac, Einstein, Bohr, Fermi, Van Vleck, and others—was devoted to magnetism. The active discussion of Pauli’s paper gives a picture of the state of the application of quantum mechanics to magnetism at the turn of the 1930s.

³⁶Teller acknowledges discussions of the Landau work with Pauli, Van Vleck, and Peierls; a variant of Teller’s argument, together with a pedagogical explanation of the connection with Landau’s derivation, was given the following spring by Van Vleck (1932, paragraph 81).

electrons “can in a certain sense still be considered as free . . . [and] that the principal characteristic effect in the magnetic field remains unchanged In particular, the relation between para- and diamagnetism, is altered, and it is possible that in certain cases the latter can exceed the former, so that we get a diamagnetic substance like bismuth.”

The necessary extension of Landau’s theory was undertaken by Peierls in Zurich, and described in a paper he submitted in November 1932, the month after his arrival at Enrico Fermi’s Institute in Rome (Peierls, 1932b).³⁷ Having had a continuing interest in the behavior of electrons in solids in the presence of magnetic fields, as well as in band theory, Peierls soon recognized that the theory of diamagnetism of conduction electrons in real solids entailed two difficulties—first, the complexity of computing by Landau’s method the exact eigenstates of electrons in a periodic potential with magnetic field, and second, the conceptual difficulty that the broadening of levels caused by collisions with impurities and phonons in metals at most temperatures and magnetic fields exceeds the spacing between the levels, thus threatening to wipe out or modify substantially the Landau effect (Peierls, 1981b).

Peierls easily overcame the first difficulty by examining the case of very weak magnetic fields, which could be treated as a small perturbation. The second difficulty would, he argued, be resolved when the widths of the states induced by collisions were small compared with the temperature, although they might be large compared with the spacing between unperturbed levels, for then collisional broadening of the levels would have small effect on the equilibrium thermodynamics. The important practical result that emerged from Peierls’s paper was an expression for the diamagnetic susceptibility involving the electron energy-momentum relation $E(k)$ at the Fermi surface.³⁸

Although encountering only “polite interest” (Peierls, 1963) from Fermi, Peierls continued on diamagnetism, and in January 1933 submitted a second paper (Peierls,

1933b) in which he considered the limit of strong magnetic fields, where the level spacing is now large compared to the temperature. He recalls that in examining the case of free electrons, “it suddenly dawned on me that you would in that case get a (more or less) periodic variation in the susceptibility as a function of the . . . reciprocal of the magnetic field intensity. And this of course immediately reminded me of the funny result of de Haas and van Alphen,” which he had learned about during a visit to de Haas in Leiden in late 1930 or 1931. As Peierls recounts, de Haas

talked about this strange effect . . . which mystified him. And I remember he told me since he didn’t understand what was going on, he was trying to look for the dependence of this effect on everything, including time. So he kept one particular specimen of bismuth in his cupboard, and every few months remeasured the effect to see if it was going to change. . . . I found this phenomenon quite mystifying, but I don’t think I attempted to find an explanation at that time. (Peierls, 1981b)

It was clear from the agreement of the theory that Peierls derived in his paper with measurements by de Haas and van Alphen on bismuth in strong fields that he now had the correct explanation.

The work by Peierls on strong-field diamagnetism is another of those curious situations where a scientist realizes only after making a discovery that he had earlier encountered but not been receptive to, the crucial ideas and facts underlying the discovery. For not only had Peierls not recognized the connection to de Haas and van Alphen’s work until after he had his theory in hand, but he had not remembered that Landau had already presaged such an effect 3 years earlier. “Presumably I never read Landau’s paper carefully, having had its main contents explained by him before publication, or if I saw the remark, I accepted Landau’s assurance that it was unobservable, and promptly forgot it” (Peierls, 1980, p. 36). Peierls does not recall talking to Landau about (Peierls’s) working out the theory of the effect, but paints an insightful picture in conjecturing, “if I did, I imagine that he might have said that it was already known to him in his paper. Although as long as he thought that my result was correct, which obviously he must have done because he had obtained it himself, he might well not have bothered to point this out. This was quite within Landau’s nature” (Peierls, 1981b).

The third side of this triangle, the lack of influence of Landau’s work on the de Haas—van Alphen experiment, has been recently commented on by Shoenberg (no date):

The remarkable coincidence is that theoretical prediction and experimental observation of the oscillatory effect should have occurred almost simultaneously with neither side being aware of the other side’s contribution. In fact the motive behind the Leiden experiments had nothing to do with Landau’s remark, but was based on a long standing hunch of de Haas that there should be a close correlation between diamagnetic susceptibility and the change of electrical resistance in a magnetic field.

³⁷Peierls derived the result here for the case of tight binding; however, later work showed that the result is in fact more general.

³⁸In particular, for parabolic bands the diamagnetic susceptibility becomes essentially proportional to the inverse of the electron effective mass m^* at the Fermi surface, so that large diamagnetic susceptibilities arise when $E(k)$ has large curvature, as turned out to be the case in bismuth (Jones, 1934a, 1934b). A similar form for the paramagnetic susceptibility, given by Bethe (SB, p. 476), in terms of the electron density of states at the Fermi surface, would allow inclusion of lattice effects in the Pauli result; here the susceptibility turns out to be proportional to m^* , opposite to the behavior of the diamagnetic susceptibility. Jones’s two papers on alloys and bismuth are especially noteworthy for making the first connection between structure and the electron theory of metals, explaining how the electron energies induce a distortion of the lattice in bismuth, analogous to the modern “Peierls transition” in one-dimensional structures.

Even though they proceeded along independent paths, the experimental and theoretical discoveries of the de Haas—van Alphen effect provide one of the first major examples of successful agreement between theory and contemporary experiment in quantum solid-state physics. As in electrical transport, the theory had now reached the level of sophistication where a working relation between theory and experiment could develop.

B. Ferromagnetism

While Pauli in Hamburg in late 1926 wrote his thoughts on paramagnetism to Heisenberg in Copenhagen, Heisenberg in his letters told Pauli of his own mulling on the problem of ferromagnetism (collected in WP1). The correspondence between these two friends brings out, on the one hand, the common origins of their current interest in magnetism—the search for the symmetries of the wave function and statistics of a many-electron system—and on the other, their different concerns—Pauli's with the statistics of gases, which would lead to the Sommerfeld-Bloch free-electron development, and Heisenberg's with few-electron systems, which would lead to the study of solids as extended molecular systems and to the modern theory of ferromagnetism.

After his arrival in Copenhagen in May to take up the position of Lecturer (Robertson, 1979), the twenty-four-year-old Heisenberg worked on establishing the connection between the Pauli exclusion principle and the antisymmetry of the wave function of a several-electron system (Heisenberg, 1926d), turning to “a practical problem . . . the helium atom with two electrons” (Heisenberg, 1963) as a simple test case. He soon wrote two papers, submitted in June and July; the first, on the properties of two and more like-particle systems (Heisenberg, 1926e), introduces the notion of the (dynamically conserved) symmetry of the wave function, the fully antisymmetric (determinantal) wave function for a many-electron system, and the concept of the exchange interaction, or “resonance” as Heisenberg termed it. The second paper, on the calculation of the spectrum of the helium and other two-electron atoms (Heisenberg, 1926a), explicitly introduces the Coulomb exchange integral.³⁹ The connection of these arguments to the question of statistics, a problem in this period only beginning to come into focus,⁴⁰ was to be a theme in the correspondence with Pauli.

Responding on 28 October (Heisenberg, 1926b) to the Pauli letter of 19 October (Pauli, 1926b) on gas degeneracy, collisions, and fluctuations, that expressed his (Pauli's) now “considerably milder” view of Fermi-Dirac statistics,

³⁹These papers are discussed in Miller (1984).

⁴⁰“I was interested in two electrons and not in many electrons. Therefore I could forget about Bose and Fermi statistics” (Heisenberg, 1963).

Heisenberg attributed his long delay in answering to the fact that Pauli's letter “constantly made the rounds here, and Bohr, Dirac and Hund are scuffling with us about it.” Heisenberg continues, “With regard to the Dirac statistics we are in agreement . . . if the atoms obey your exclusion principle [Verbot], then so must the gas.” A week later on 4 November (Heisenberg, 1926c); for further detail see Hoddeson and Baym, 1980, Heisenberg reveals to Pauli, “I myself have thought a little bit about the theory of ferromagnetism, conductivity and similar filth [S . . . erein]. The idea is this: in order to use the Langevin theory of ferromagn[etism], one must assume a large coupling force between the spinning electrons (for *only these* turn). This force shall be obtained, as for helium, indirectly from the resonance.” Heisenberg thus took here the two crucial steps towards the theory of ferromagnetism: the first was to identify, for the first time, the elementary magnetic moments responsible for ferromagnetism as those of the recently discovered spinning electrons,⁴¹ and the second was to realize that the exchange interaction could give the strong coupling required to align the moments.

Heisenberg had already accepted, by Christmas 1925, Samuel Goudsmit and George Uhlenbeck's hypothesis that the electron spin had associated with it an intrinsic magnetic moment (Heisenberg, 1925),⁴² and was aware that the gyromagnetic ratio g of the electron spin required by spectroscopic measurements agreed with that of ferromagnets, determined by the Einstein—de Haas—Barnett effect. The latter measurements, although from the first beset by uncertainties (Galison, 1982),⁴³ had by 1925 indicated with good accuracy a g factor of 2 (Barnett and Barnett, 1925). Heisenberg's comment in his letter to Pauli, “for only these turn,” in fact refers to these experiments, as is clear from his later paper on ferromagnetism (Heisenberg, 1928b), in which he writes “it follows from the known factor $g=2$ in the Einstein-de Haas effect (a value measured only in ferromagnetic substances), that in a ferromagnetic crystal only the intrinsic moments of the electrons are oriented, and not the atoms at all”⁴⁴ (Heisenberg, 1928c, 1928d).

⁴¹For the discovery of spin see, for example, Jammer (1966, pp. 146—153) and references therein.

⁴²Earlier, Heisenberg was not ready to accept Kronig's prior suggestion of the spin; see, for example, Bohr (1926), where Bohr relates to Kronig, “I have since had quite a difficult time in trying to persuade Pauli and Heisenberg, who were so deep in the spell of the magic duality that they were most unwilling to greet any outway of the sort.”

⁴³The topic was a major subject in the 1930 Solvay Conference, where P. Weiss reviewed “Les phénomènes gyromagnétiques,” concluding that experiments, mainly by S. J. Barnett and L. J. H. Barnett (1925), provided “une présomption très forte en faveur de l'attribution du ferromagnétisme à l'électron pivotant.” See Solvay (1932), p. 354.

⁴⁴Heisenberg (1928d, p. 115) writes that “the orbital moments in the crystal are *not* freely orientable, on the whole they compensate and do not contribute to the crystal's magnetism.”

The existence of strong internal forces between the fundamental magnetic moments in a magnetic material underlay Weiss's phenomenological "mean molecular field" theory of 1907 (Weiss, 1907, 1911, 1930), a theory drawing on Langevin's 1905 statistical-mechanical theory of the paramagnetism of magnetic dipoles (Langevin, 1905), as well as earlier work by Ewing from 1886 to 1900 on internal forces in magnetic materials (Ewing, 1890, 1893; Weiss and Foex, 1926). However, in order to find agreement with experiment, Weiss had to assume, *ad hoc*, forces orienting the atoms in a ferromagnet linear in the magnetization, yet of order 10^3 – 10^4 times stronger than could be explained from simple magnetic interactions. Heisenberg's key idea was to show how strong, nonmagnetic forces between electrons that favor spin alignment arise from the quantum-mechanical exchange interaction, in the same way as he had just shown that they produce level splittings between electrons in singlet (para) and triplet (ortho) states of two-electron atoms.

As he continues on 4 November, "I believe that one can in general prove: parallel orientation of the spin vectors always gives the *smallest* energy. The energy differences in question are of *electrical* order of magnitude, but fall off with increasing distance *very quickly*. I have the feeling (without knowing the material even remotely) that this in principle could be extended to give a meaning for ferromagnetism. To resolve the question of why, whereas most materials are not ferromagnetic, certain ones are, one must simply calculate quantitatively, and perhaps one can make plausible why circumstances are most favorable for Fe, Kr [*sic*] and Ni. Similarly, in conductivity the resonant wandering of the electrons à la Hund comes into play"⁴⁵ (Heisenberg, 1926a). Pauli, interestingly, made a notation on this letter: "ferromagnetism doesn't work [geht nicht!] (gas degeneracy)" (Heisenberg, 1926c, see note a), a comment likely reflecting his own fresh discovery that degeneracy of the conduction electrons leads to a striking suppression of their (para)magnetizability. To Pauli's communication of his pessimism (letter not available), Heisenberg responded on 15 November, "I nevertheless consider the idea that it [ferromagnetism] has to do with the resonance very attractive" (Heisenberg, 1926f).

Heisenberg's letter to Pauli confirms his admitted lack of familiarity with the problem; in particular, as his reference to Langevin indicates, he had not yet gone back to the literature, for Langevin had dealt only with paramagnetism and diamagnetism. Later, in his classic article

⁴⁵Heisenberg refers here to the concept of tunneling, or "resonant wandering," which Hund (1927) had just introduced to understand molecular spectra. Hund and Heisenberg were both in Copenhagen in 1926–1927, and had in fact, "dreamt up a theory of conductivity" on the day of the letter to Pauli (Hund, 1926, 1984). This discussion foreshadowed the theory of conductivity that Bloch would develop more than a year later under Heisenberg's direction.

on ferromagnetism, Heisenberg would correctly refer instead to Weiss. The origin of Heisenberg's interest in ferromagnetism is uncertain. One source is likely his friendship with physicists at the University of Hamburg, for whom magnetism was a favorite topic. Sommerfeld's student Wilhelm Lenz, and Lenz's own student Ernst Ising, both then at Hamburg, had recently developed (1920, 1924) the "Ising model" of ferromagnetism (in which spins, which can be either "up" or "down," interact with their nearest neighbors) (Lenz, 1920; Ising, 1925; Brush, 1967).⁴⁶ In 1926, Pauli and Wentzel, also Sommerfeld students, held positions in Hamburg, while Otto Stern, who with Walter Gerlach had in 1920 discovered spatial quantization in molecular beams, directed Hamburg's program in experimental physics. Discussions about rotating electrons, with visitors such as Goudsmit in February 1926 and Yakov Frenkel in April 1926, were common at Hamburg during the period (Pauli, 1926c, 1926d). Heisenberg visited at least once, in January 1926 (Heisenberg, 1926g). In addition, Debye organized a magnetism week from 21–26 June 1926 in Zurich, attended by Schrödinger, Pauli, Sommerfeld, Langevin, Stern, and Weiss (Pauli, 1926e; Schrödinger, 1926; Mrs. Schrödinger, 1951).⁴⁷ Although Heisenberg apparently did not attend this meeting, he visited his teacher Sommerfeld in Munich the following month (Heisenberg, 1969, p. 104), when Schrödinger was also present, and they may well have talked about magnetism—the topic of Sommerfeld's advanced topic lecture course at this time (Munich, 1926).

Heisenberg did not submit his theory of ferromagnetism for publication until 20 May 1928 (to the *Zeitschrift für Physik*), a year and a half after spelling out his intuitions to Pauli. Two factors contributed to the delay. First, he did not have available in 1926 the group-theoretical machinery he would use to compute the energies of a many-spin system in terms of the exchange interaction; this would be developed by Wigner, Heitler, London, and Hermann Weyl in the interim.⁴⁸ Second,

⁴⁶Ising correctly showed that the one-dimensional model would not exhibit ferromagnetism; however, his erroneous arguments that neither would the three-dimensional "spatial model" led to the Lenz-Ising model's not being seriously worked on until the mid-1930s.

⁴⁷Published reports from the Zurich conference have not been found.

⁴⁸In this period, Wigner, Hund, Heitler, London, Weyl, and others were extending the two-electron picture Heisenberg had used to study helium to many-electron systems, using the representations of the permutation group, which had been used successfully to analyze atomic spectra. This approach looked so promising at the time that all who were working on the quantum theory of metals in 1928 studied it, going through the classic papers by Wigner and von Neumann and using as a text Speiser (1923) (see Bethe, 1981a; Peierls, 1981a). The major papers (by Wigner, Hund, Heitler, and London) are listed in Heisenberg (1928b).

during this time he became fully occupied with the conceptual foundations of quantum mechanics, developing the uncertainty principle the following spring. Heisenberg's thoughts appear not to have returned to ferromagnetism until after his move to Leipzig in the fall of 1927, when he suggested it as a thesis problem to Bloch. As Bloch chose instead to examine the electrons in metals, Heisenberg continued on ferromagnetism himself.

Once in Leipzig, Heisenberg contacted the Berlin group-theory experts, London, Wigner, and John von Neumann, to learn how to deal with calculating the exchange energy of a many-electron system. He later recalled the "close cooperation between Berlin and Leipzig I went to Berlin to discuss the matter because it was kind of an application of London's ideas on quantum chemistry"⁴⁹ (Heisenberg, 1963; Bloch, 1964, p. 22). London, who had studied under Sommerfeld, was at the time Schrödinger's *Assistent* at the University of Berlin. Wigner was *Assistent* to Richard Becker, another Sommerfeld student and at that time Professor of Theoretical Physics at the Technische Hochschule in Berlin.⁵⁰ von Neumann, then also in Berlin, occasionally collaborated with Wigner.

Two months prior to Heisenberg, in March 1928, Frenkel at the Physical-Technical Institute in Leningrad⁵¹ submitted a paper, published in the same volume of the *Zeitschrift für Physik* as Heisenberg's, on the magnetic and electrical properties of metals at absolute zero (Frenkel, 1928a). Frenkel announced his paper to Sommerfeld on 8 March 1928, in a letter continuing an intensive discussion that they had begun at the Volta conference in Como the year before (on the Sommerfeld free-electron-gas model for metals) (Frenkel, 1928b, 1928c). In his paper Frenkel briefly speculates that a spontaneous magnetic moment can appear, as a consequence of a coupling arising from Heisenberg's resonance phenomenon, between the individual spin moments of the free electrons, and between the spin vectors of the free and bound electrons,⁵² "yielding, in certain circumstances, an unusually large negative value for the 'magnetic' energy . . ." However, unlike Heisenberg, Frenkel did not develop this suggestion into a quantitative theory. No mention of Frenkel's paper appears either in Heisenberg's publica-

tions (1928b, 1928c) or in his letters to Pauli, although hearing from Sommerfeld of solid-state studies in Leningrad might possibly have motivated Heisenberg to begin in earnest to work out his earlier ideas on ferromagnetism.⁵³

Heisenberg used his scientific correspondence with Pauli, who was by now in Zurich, as a way of formulating his ideas; a series of seven letters and postcards, written between 3 and 21 May 1928, allows us to take a close look at the final stage in the development of his theory of ferromagnetism (Heisenberg, 1928a, 1928c, 1928e, 1928f, 1928g, 1928h, 1928i). In the first letter, on 3 May 1928, Heisenberg (1928a) reveals that he considered ferromagnetism a diversion from "more important problems . . . annoying myself with Dirac's quantum field theory," problems which "today I have nothing new to say about . . ." Instead, "I've dealt with ferromagnetism."

Of the three approximations available to compute the strong interactions between electron spins, the Pauli-Sommerfeld free-electron model, the Bloch tight-binding method, and the London-Heitler method of treating the "exchange of the valence-electrons of any two atoms in the lattice," Heisenberg argues that when the last "is the best approximation, one can, in certain circumstances obtain ferromagnetism," i.e., when the exchange term, $J_{(12)}$, is positive. Then using group-theoretic methods to calculate the energy levels, and neglecting their fluctuations in evaluating the partition function, he arrives at "the Weiss [mean-field] formula" for the magnetization [although here for the first time the hyperbolic tangent, for spin $\frac{1}{2}$, replaces the Langevin function in Weiss's theory (1907, 1911)].

Four days later Heisenberg (1928e) writes, "Today I would like to continue my epistle on ferromagnetism, and first write about the main objections, which you have naturally recognized for quite some time. . . . The major swindle lies in that I have inserted for all terms of the same total angular momentum j the mean energy rather than the real term value." He then proceeds to include fluctuations in the energy levels, using a Gaussian distribution, and now arrives at a modified Weiss formula. Only for a lattice with at least 8 neighbors (z) does this result give a spontaneous magnetization that goes to zero at a critical temperature. For z at least 6 he finds another solution, which "has no physical meaning." [This branch of spontaneous magnetization, together with another (unmentioned by Heisenberg) present for *all* z , appears to arise from an extraneous power of the exchange interaction parameter that Heisenberg slipped into the last term of the fluctuation correction that he reported to Pauli; it is corrected in the published version.] He concludes, "All in all, however, I find that one already understands the

⁴⁹Bloch (1964) recalls that the whole Leipzig group, including Heisenberg, Wentzel, and students, was "quite frequently at colloquia in Berlin."

⁵⁰Sommerfeld (1926), in recommending Becker for this chair in 1926, wrote, "Such a happy combination for theoretical physics and technology you could hardly find in another candidate." Later on, Becker and his school would play an important role in the technological application of the theory of ferromagnetism.

⁵¹Also at this institute at this time were Landau and J. Dorfman. See V. Frenkel (1974).

⁵²Frenkel credits Dorfman with the idea of coupling between the spins of the free electrons and those bound in nonclosed shells (Frenkel, 1928a, p. 35).

⁵³Sommerfeld warned another member of his circle, Georg Joos, who wanted to embark on a theory of ferromagnetism, "You had better wait. Heisenberg will presumably hit the center of the target again" (Sommerfeld, 1928).

origin of F[erro]M[agnetism] substantially better.” Later in the day, Heisenberg (1928f) sent Pauli a short postcard in which he adds that z refers to the number of “nearest neighbors,” and that Fe, Co, and Ni satisfy his $z \geq 8$ condition for “ferromagnetism in the Weiss sense.”

Unfortunately, Pauli’s letters to Heisenberg from this period are lost. As Heisenberg’s letters reflect, Pauli expressed serious objections concerning both the physical assumptions and the mathematics, which served the important function of forcing Heisenberg to refine his theory. From letters to Bohr the following month, we learn that Pauli in fact considered Heisenberg’s work on ferromagnetism “very beautiful” (Pauli, 1928a) and one reason for learning group theory from Weyl, although Weyl’s “philosophy and life style,” Pauli adds (1928b), “are not to my taste.”

Heisenberg deals, on 10 May (1928c), with more of Pauli’s objections: “I believe that I can now answer most of your questions, to a degree.” First, he argues that the magnetic moments of the atomic cores can be neglected. He next turns to discuss the sign of the exchange integral J , admitting, “I have not succeeded in achieving a half-way useful evaluation of J I have the dark suspicion that J first becomes positive for p and d -states, thus not for s -states. . . . It seems not unlikely to me that in Fe-Co-Ni the d -states bear the guilt for ferromagnetism.” He then comments on the desirability of understanding the relationship between his theory and Pauli’s theory of paramagnetism, and ends with further explanation of his calculation of the partition function. (One can imagine Pauli trying to reproduce Heisenberg’s somewhat erroneous result from the sketchy calculation Heisenberg sent earlier.) In a postcard, written three days later (1928g), Heisenberg adds that he has just succeeded “in gaining clarity about the sign of $J_{(12)}$ J is negative for small principle quantum numbers [n], as for London and Heitl[er]; in contrast for large n it becomes positive. The boundary lies at $n = 3$, but it can just as well be 2 or 4.”

The next day, he responds (1928h) to a comment of Pauli’s concerning his own work on paramagnetism: “Actually, I also believe that your explanation of the susceptibility of the alkalis is correct; one can perhaps say that your theory is useful for metals of very large conductivity, while the third [Heitler-London] method is useful for those of smaller c[onductivity]. In reality both are but very crude approximations.” Finally, on 21 May, Heisenberg (1928i) writes Pauli, “well, I’ve sent Scheel [the editor of the *Zeitschrift*] a manuscript about ferromagnetism. . . . The assumption of the Gaussian distribution for the energy values, which for sufficiently low temperatures leads to false results, still seems unsatisfactory to me. . . . Perhaps I’ll set one of my people here on to the calculation. . . .”

Heisenberg’s article (1928b) closely follows the lines of his correspondence with Pauli in both 1926 and 1928. After noting that neglect of electron interactions leads, according to Pauli, to paramagnetism or diamagnetism, he describes the basis of his theory: “The empirical phenomenon that ferromagnetism presents is very similar

to the situation we met earlier in the case of the helium atom.” The clue is the splitting of the two-electron helium atom into singlet and triplet terms by the exchange interaction. He continues, “We will try to show that the Coulomb interaction together with the Pauli principle suffice to give the same result as the molecular field postulated by Weiss. Only very recently have the mathematical methods for treating such a complicated problem been developed by Wigner, Hund, Heitler and London.” Recalling the Heitler-London expression for the exchange integral, and explaining how the exchange energy can tend to align spins, he then launches into a very formal calculation of the energy levels in terms of the characters of the permutation group, finally specializing to nearest-neighbor interactions with a common exchange integral, and introducing the Gaussian approximation.⁵⁴ His resulting version of the Weiss formula implies that a spin must have at least 8 nearest neighbors for the system to become ferromagnetic,⁵⁵ a result he continues to regard as significant; it also implies that the system must become paramagnetic again at low temperature, but he does not “believe that this result has physical meaning. It arises mathematically through the assumed Gaussian distribution of the energy values.”

Heisenberg concerns himself in the final section with the applicability of his theory to real ferromagnetic materials, Fe, Co, and Ni. He finds that to fit the transition temperatures of these ferromagnets requires an exchange integral J_0 of order one-hundredth of the hydrogen atom ground-state energy, while because of the exponential fall-off of the exchange effect, ferromagnetism should not occur in Fe or Ni solutions. However, “Very much more difficult is to answer the question of the sign of J_0 .” Spelling out in more detail his arguments to Pauli that principal quantum number $n = 3$ is the first likely place that J_0 becomes positive, he concludes that the two conditions $n \geq 3$ and $z \geq 8$ together are “far from sufficient to distinguish Fe, Co, Ni from all other substances.” Heisenberg was aware of the limitations of his theory and ends with the remark, “It was of course only to be expected that the temporary theory sketched here offers but a qualitative scheme into which ferromagnetic phenomena will perhaps later be incorporated. . . . I hope later to go into these questions as well as a thorough comparison of theory with experimental results.”

⁵⁴The familiar “Heisenberg model” of ferromagnetism, with Hamiltonian $\sim \sum J_{ij} \sigma_i \cdot \sigma_j$, does not in fact appear in Heisenberg’s paper, but would be given later in this “expressive form” by Dirac (1929, p. 731) without reference to Heisenberg.

⁵⁵Interestingly, Heisenberg’s account of fluctuations, while inadequate, does give a noticeably better predicted critical temperature T_c , than a “mean-field” calculation for $z \geq 8$. For example, for $z = 12$ mean-field theory gives $T_c/J_0 = 6$, the modern “exact” result is 4.02, while Heisenberg’s calculation yields 4.73; for $z = 8$, the results are 4, 2.53, and, for Heisenberg, 2; for $z = 6$, the results are 6 and 1.68, while Heisenberg finds no ferromagnetism.

Heisenberg's correspondence with Pauli on ferromagnetism includes two letters in the summer of 1928 (Heisenberg, 1928j, 1928k), in the second mentioning that he has written a further paper on ferromagnetism for the "Sommerfeldfestschrift" (Heisenberg, 1928d) dealing with the interaction of several valence electrons. Still mulling over his "unpleasant swindle," the Gaussian distribution, he continues, "I'd like very much if Weyl could try this problem. I've completely given it up. The whole question seems important to me on account of the similarity between my model and Ising's. My present view is that Ising should have obtained ferromagnetism if he had assumed sufficiently many neighbors (perhaps $z \geq 8$) That Ising uses this ['wild spatial'] model as an argument against ferromagnetism seems to me an indication that he did not understand in perspective his own work."

Heisenberg's immediate involvement with the foundations of ferromagnetism ends at this point; although the mathematical description of the cooperative effects in his model proved too difficult for the time, Heisenberg intuitively identified the correct physical basis of ferromagnetism as a quantum phenomenon, and thus opened the field of the quantum theory of ferromagnetism. The many problems left unsolved would in subsequent years be addressed by specialists in this field. Heisenberg's remarkable work between 1926 and 1928 further stands out as the first exploration of the physical consequences of electron-electron interactions in solids within the framework of quantum mechanics.

The main development of the quantum theory of ferromagnetism continued in Zurich, where Bloch, who was Pauli's *Assistant* in the 1928–1929 academic year, followed a study of magnetoresistance in the fall (Bloch, 1929a) with the first of a series of papers on ferromagnetism (Bloch, 1929b). Recognizing Heisenberg's work as a correct insight, Bloch felt challenged to improve the "not very reliable" mathematics in which it was expressed (Bloch, 1981), and began with the role played by the conduction electrons. The question had been raised by J. Dorfman and co-workers (Dorfman and Jaanus, 1929; Dorfman and Kikoin, 1929),⁵⁶ in Ioffe's institute in Leningrad, who argued from the observed anomaly in the thermoelectric effect at the Curie point in Ni that the specific-heat discontinuity there arises from the conduction electrons, and therefore they must be the crucial actors in ferromagnetism, rather than the bound electrons considered by Heisenberg in his Heitler-London approach. By studying the free-electron model, Bloch could avoid Heisenberg's Gaussian assumption. Pauli was interested in this aspect of the problem, possibly, as Bloch later offered, because it extended Pauli's own treatment of paramagnetism, also based on a study of the conduction electrons (Bloch, 1981).

To determine whether conduction electrons can be the

source of ferromagnetism, Bloch in his paper (1929b) carries out the original calculation of the now familiar exchange energy of a free-electron gas,⁵⁷ but discovers that only at low electron densities (or equivalently in narrow bands), too low for the alkalis, does the attractive exchange interaction dominate the zero-point energy of the electrons to produce a ferromagnetic state.⁵⁸ The zero-point motion of the electrons, he concludes, must be taken into account in deciding whether a metal can be ferromagnetic. While his calculation neglects the influence of the atoms on the electrons, these, he points out, can be taken into account by using periodic (Bloch) wave functions in the exchange integral; the important contribution, he finds (in a calculation not described here), "comes when the two electrons are close together, in the neighborhood of the same atom." He is in fact led back to the region of applicability of the Heitler-London approach used by Heisenberg. The answer is not obvious, for as he remarks, "the exchange integral can become negative, decreasing substantially the possibility for ferromagnetism. On the other hand, we have shown earlier that a periodic potential can lower the zero-point energy of the electrons so that in some circumstances a condition [for ferromagnetism] can be fulfilled."

In carrying out his free-electron calculation, Bloch applied the determinantal method recently developed by the American John Slater (1929) in his theory of complex atomic spectra; he thanks Slater in his paper for showing him the manuscript and for a number of friendly discussions. Slater, who visited Heisenberg's institute in Leipzig as a Guggenheim fellow during the summer and fall of 1929, recalls showing Bloch a preprint of this work in Zurich, which Bloch was "greatly taken with."⁵⁹ In this period Bloch and Slater would have an important influence on each other's work—Bloch's earlier work on metallic structure, and Slater's on complex atomic spectra converging in 1929 on the problem of ferromagnetism.

Slater had developed his theory of complex spectra in the spring of 1929 at Harvard, in an attempt to understand why Douglas Hartree's self-consistent field method

⁵⁷Wigner and Seitz later cited this result in their calculation of the energy of the interacting electron gas (Wigner and Seitz, 1934, p. 512, note 5).

⁵⁸The problem of ferromagnetism of a uniform electron gas would be shortly revisited by Edmund C. Stoner (1930a, 1930b). As Stoner observed, a model of ferromagnetism due to free electrons would yield Curie temperatures much higher than those observed.

⁵⁹Slater says in his biography that he came to Zurich, en route to Leipzig, to participate in a conference—the 1–4 July ETH "lecture week," that year on x rays and quantum theory (Zurich, 1929; Pauli and Scherrer, 1929)—and that he first met Bloch there. The datings of the Slater paper (received 8 June, and sent off, Slater says, before he went to Europe) and the Bloch paper (submitted 10 June), and the fact that both acknowledge the other, suggest that they in fact first interacted in early June (Slater, 1975, pp. 62 and 123).

⁵⁶Their result, however, is inconclusive; see, for example, Van Vleck (1932, p. 345, footnote 43) and Stoner (1930a, 1930b).

(Hartree, 1928; Darwin, 1958)⁶⁰ was so successful in analyzing atomic spectra (Slater, 1963, 1967, p. 52). Using a method based on Dirac's antisymmetric determinantal wave function for a many-electron system (Dirac, 1929), he constructed the many-electron function as a determinant of spin orbitals, and discovered that it gave a rather accurate self-consistent field.⁶¹ This approach, which included "at the very beginning" the correct antisymmetry properties, was simpler than the group-theory method then in vogue, a point Slater emphasized at the start of his abstract: "Atomic multiplets are treated by wave mechanics, without using group theory." He recalls that when he arrived in Europe "everyone knew of the work"; it was rumored that "Slater had slain the Gruppenpest" (Slater, 1975, p. 62), and so physicists could, as Bethe put it recently (1981a), "happily . . . forget all the group theory that we had learned." Bloch recalls (1981), "we were all relieved that one had a much more familiar way of expressing the content than all those general high-brow group-theoretical arguments."

During his stay with Heisenberg in Leipzig, Slater entered the discussion of the merits and relationship of Heisenberg's exchange and Bloch's tight-binding methods, and would contribute substantially to unraveling the origin of ferromagnetism. By comparison of the two approaches in the context of the cohesion of metals, he saw first that they formed different unperturbed bases for attacking metals by perturbation theory—analogue, as he later described, to the relation between the "Heitler-London and the Hund-Mulliken molecular-orbital approaches, respectively, to the molecular problem"⁶² (Slater, 1975, p. 126), pointing out in his paper that "they are essentially equivalent in their results when properly handled" (Slater, 1930b). With this understanding, Slater pushed Bloch's recent demonstration that the nonmagnetic state of conduction electrons at metallic density has lower energy than the magnetic to "the quite general conclusion that the outer electrons, which are largely if not entirely responsible for both cohesion and conduction, cannot produce ferromagnetic effects." He continues, "It is a very attractive hypothesis to suppose that in the iron group the existence of the $3d$ and $4s$ electrons provides . . . the two electron groups apparently necessary for ferromagnetism; for it is only in the transition groups that we have two such sets of electrons, and this criterion

⁶⁰Hartree was also present at the July meeting at the ETH (Slater, 1975).

⁶¹The variational—now called Hartree-Fock—method of improving Hartree's approach was suggested in November 1929 by V. Fock (1930a, 1930b) and Slater (1930a) independently.

⁶²Hund had always favored the one-electron approximation, considering ferromagnetism a matter of multiplet level splitting and a competition between two energies of the same order of magnitude, that for excitation to states of the necessary symmetry, and the gain by level splitting. He wrote in his diary on 16 January 1929, "Heisenberg's result $z \geq 8$ in ferromagnetism debatable [angreifbar]." He later remarked, "Heisenberg's understanding of ferromagnetism by means of the exchange integral was not to my taste" (Hund, 1984).

would go far toward limiting ferromagnetism to the metals actually showing it." Such inner electrons, if in well-separated orbits, could, Slater conjectures, have lower energy in a spin-aligned state. Finally, in his analysis of the cohesion of metals—a problem similar to that of the Heisenberg model of ferromagnetism, only with generally negative exchange interactions—he presents equations for the wave functions of the model, basically a Schrödinger equation in difference form, that would be used in subsequent studies by Bloch and Bethe. [Dirac's algebraic formulation (1929) of the Heisenberg model in terms of Pauli spin matrices had not yet taken hold.]

The following spring back at Harvard, Slater computed the sizes of incomplete atomic shells of various atoms in the periodic table, and showed that the $3d$ orbitals in the iron group satisfied the condition for spontaneous magnetization that their size be small enough for the energy decrease caused by the exchange effect to outweigh, à la Hund (footnote 62) the energy increase from excitation of electrons at the top of the band (Slater, 1930c). He notes that if ferromagnetism "depends on the existence of incomplete shells within the atoms . . . the metals most likely to show it would be Fe, Co, Ni, and alloys of Mn and Cu (Heusler alloys)."

Up to this point, the work of Bloch and Slater was concerned with understanding the physical basis of the Heisenberg model, rather than with its mathematical solution. But once Bloch came to Utrecht, during the fall of 1929, visiting H. Kramers on a Lorentz Foundation fellowship (the second part of this year was spent as an assistant in Haarlem), he suddenly felt "quite free" after the busy time as Pauli's *Assistant*, and in this frame of mind began to "pick up old things" (Bloch, 1981). He turned to the question of the physical predictions of the Heisenberg model itself, particularly to improving Heisenberg's treatment of ferromagnetism at low temperatures, a region in which both Weiss's mean-field theory and Heisenberg's calculations are invalid. Replacing Heisenberg's group-theoretical approach by that of Slater determinants, Bloch discovered "spin waves," the states corresponding to single or few spin flips in the fully aligned ground state. "I said, why if electrons can hop, spins can also hop" (Bloch, 1981). Deriving a closed expression for their energy eigenvalues, Bloch went on to relate the low-lying spectrum to the thermodynamics, making the remarkable connection that the fluctuations arising from the spin waves at low temperatures in one- and two-dimensional lattices destroy the possibility of ferromagnetism, while in three dimensions they give a $T^{3/2}$ falloff in the magnetization, compatible with the then existing data (Bloch, 1930b).⁶³ In an allusion back to

⁶³The discovery of spin waves was in fact made simultaneously and independently by Slater, who in the concluding section of his paper on the cohesion of metals illustrated the effect of spin fluctuations in the normal state with a calculation of the excited states of a fully aligned chain of spins, computing the single flipped spin state exactly and the multiply-flipped states approximately. Slater did not, however, as Bloch notes, draw the implications of spin waves for ferromagnetism.

Heisenberg's calculation he remarks that "not only the number of nearest neighbors, but also their arrangement plays a role."

Bloch gave a physical picture of the spin waves at the third Leipziger Vorträge (Bloch, 1930c), held in the summer of 1930 while he was still based in Haarlem. This meeting, one of the earliest to specialize in solid-state physics, was organized by Debye with the help of his *Assistant* Henri Sach, and attended by von Laue, Bethe, Ioffe, Grüneisen, Bloch, Peierls, and Nevill Mott. The talks included Bloch's on magnetism, Grüneisen's on the temperature dependence of the electrical and thermal resistance of metals, and Peierls's on the behavior of metallic conductors in strong magnetic fields (Peierls, 1930b). The degree to which solid state was functioning as a target of opportunity for quantum mechanics is underscored by Debye's hope in the preface to the Proceedings that the talks would not only illuminate pure electron diffraction and interference experiments—i.e., the tools that enabled physicists to see inside the lattice—"but also to verify what the wave-mechanical conception can achieve in explaining the properties of metals."

In his review of magnetism delivered at the October 1930 Solvay meeting, Pauli summarizes Heisenberg's, Bloch's, and Slater's progress in understanding ferromagnetism (Pauli, 1932). The open questions he feels are threefold. First, why are so few substances ferromagnetic? Slater's relation between ferromagnetism and orbit size does not, to Pauli, "seem solidly based." A second question is the magnitude of the saturation magnetization, which "is not compatible with the hypothesis of an electron freely circulating about an atom, and it seems rather that several atoms must participate. . . ." Third is the problem of including the influence of the crystal lattice on the direction of magnetization. He continues, "Under [actual] conditions Bloch's approximation is rather bad; one should, however, consider as established his general results that ferromagnetism is possible under conditions very different from those in which the Heitler-London method is applicable, and that in general it is not sufficient only to consider the signs of the exchange integrals." On the difficult problem of solving the Heisenberg model, Pauli lays down what would be a challenge to many subsequent generations, "that an extension of the theory of Ising to a three-dimensional lattice might give ferromagnetism. . . ."

The following spring, Bethe turned to the solution of the Heisenberg model. At Fermi's institute in Rome as a Rockefeller Foundation fellow at the time, he decided, as he wrote to Sommerfeld in May 1931 (Bethe, 1931c), to "treat the problem of ferromagnetism decently [and] . . . really calculate the eigenfunctions." In this letter he commented in detail on the limitations of Bloch's spin-wave theory, which Bethe felt did not discuss the solutions "precisely enough." Bethe, in his paper (1931b), analyzes the one-dimensional chain of spins with the exchange interaction J either positive, corresponding to the Heisenberg ferromagnet, or negative, corresponding to the "normal" (or now the antiferromagnetic) case, relevant

for the cohesion problem. Starting with the fully aligned state, he determines the wave functions of states with an arbitrary number of reversed spins, starting with his famous *Ansatz* for the case of two interacting spin waves, and then generalizing it. The calculation, although incomplete in many respects, e.g., "in the [$J < 0$] case, the solution of lowest energy is not yet established," is notable as the first exact solution of an interacting quantum many-body problem. Bethe expected to extend his one-dimensional analysis soon to the physically interesting three-dimensional case in a following work, but did not succeed. He would argue, in the later *Handbuch* article, that when J is positive, all three-dimensional lattices are ferromagnetic, whereas, as Bloch had found, two-dimensional lattices and linear chains never are (SB, pp. 607–618).

Bethe's results did not impress Bloch. In a letter to Peierls from Copenhagen on 6 November 1931, Bloch (1931b; this letter was kindly given to us by Bloch) complained that the work did not deal adequately with the low-temperature regime, which Bloch had studied in his spin-wave paper: "It appears to me that Bethe's tedious algebraic manipulation [Ixerei] is somewhat academic in character, in particular because it does not sufficiently discuss the neighborhood of the lowest eigenvalues. I believe that in this regime, however, my calculations are reasonable, since they neglect only the exclusion of spins on the same site and this cannot play a role in a very dilute spin gas."

Bloch had himself progressed on ferromagnetism during the last several months, and in summer 1931, at the end of his year as Heisenberg's *Assistant*, sat down to write his *Habilitationsschrift* (Bloch, 1932a). Much of this "long and learned" paper (Bloch, 1964), published in 1932, was worked out during a long hospital stay as Bloch recovered from a broken leg following a climbing accident (Bloch, 1981; Peierls, 1985). Nominally devoted to exchange-interaction problems and residual magnetization in ferromagnets, the paper presents an exceptional wealth of formalism which has become part of the fabric of the modern theory of condensed matter physics and collective phenomena. Beyond its contribution to the theory of domain walls, this work serves as a bridge between the quantum theory of ferromagnetism in the early 1930s and present theories of many-particle systems.

Bloch begins by introducing in the Heisenberg problem, as formulated by Slater, two sets of "second-quantized" operators per site that create or annihilate spins pointing to the right or left (as spins in those days usually pointed, rather than up or down as they now do). Then by showing how the Hamiltonian remains invariant under rotation of the coordinate system he derives the representation of quantum-mechanical angular momentum in terms of two harmonic oscillators, here the two sets of spin creation and annihilation operators [thus originating the technique exploited later by Julian Schwinger (1952; see also Wigner, 1959)]. Bloch then turns to statistical-mechanical questions, and in a clear reference to Bethe's recent paper, points out that "in fact to answer many [sta-

tistical] questions, knowledge of the stationary states is in principle [prinzipiell] not needed.” To illustrate his point, he introduces the now familiar connection between temperature and imaginary time, showing how finite-temperature statistical problems can be described in terms of a Schrödinger-like equation in imaginary time. Considering next the dynamics of the Heisenberg ferromagnet, he derives the Heisenberg representation equations of motion of the creation and annihilation operators. Then neglecting the fluctuations of the operators, i.e., treating them as a pair of complex order parameters whose squares give the spin densities, and making a long-wavelength expansion—“full analogous to replacing the lattice by a continuum in the Debye theory of specific heat”—he derives differential equations of motion for the order parameters. In the limit of a nearly-aligned system, the equations reproduce the long-wavelength spin-wave spectrum Bloch had earlier derived.

In the final section, Bloch focuses on the role of magnetic dipole interactions in leading to domain structure and residual magnetization, making the first connection between the Heisenberg model and the magnetization structure of real ferromagnets. Motivation for this work came in part from Bloch’s discussions with Becker. “We met in a little village in between [Leipzig and Berlin] once, just to talk about ferromagnetism” (Bloch, 1981). In his paper, Bloch writes, “We want to show here that . . . the ordinary dipole forces between the spins influence the grouping of the spins in a crystal in a decisive way The weak magnetic energies can do this since we are dealing with a very large system in which, despite the strong exchange interaction, the energies of various stationary states lie extremely close together so that even very small secular disturbances can still have a very great influence.” Including the dipole energy approximately in his long-wavelength equation for the order parameters, an approximation which Bloch points out in proof was criticized by Landau, Bloch shows that the order parameters in the limit in which only the spin density of one spin orientation or the other is large, become determined by precisely a nonlinear time-dependent Schrödinger equation. (The nonlinear terms reflect the magnetic dipole interaction.) The domain wall, he shows in a calculation familiar in all detail to students of the Ginzburg-Landau equation (Ginzburg and Landau, 1950), is then described by the now well-known “kink” solution, $\sim \text{sech}x$, of the nonlinear equation, in which the spin density changes in space from entirely “right” to entirely “left.” The more exact equations, he points out, also contain a kink solution, derived by Heisenberg by a variational method. Bloch recalls discussing this work on order parameters with Landau in 1931 while visiting Kharkov (Bloch, 1981), and indeed similar ideas would underlie Landau’s later work, such as his theory of phase transitions in 1937 and the Ginzburg-Landau equation of superconductivity.

With Bloch’s *Habilitationsschrift*, the initial development of the quantum theory of ferromagnetism reached the stage where one could hope, with application of realistic electronic wave functions in metals, to understand

properties of physical ferromagnets. Parallel to the development of the single-electron quantum theory of metals out of Pauli’s study of paramagnetism as a test case in the statistics of gases, the quantum theory of ferromagnetism evolved out of Heisenberg’s concern with few interacting-electron systems, in particular the helium atom, his favorite test case. Indeed one may regard Heisenberg’s works on helium as a second independent root of the quantum theory of solids. The one-electron theory of metals was insufficient to explain ferromagnetism, and the major works examined here, by Heisenberg, Bloch, Slater, and Bethe, indicated clearly that a satisfactory explanation required taking into account the collective interactions between electrons in the crystal lattice. These works, in their attempts to treat many-electron interactions, contain the beginnings of the modern theory of collective phenomena, a theory which would reach fruition more than two decades later.⁶⁴

IV. SUPERCONDUCTIVITY, 1929–1933

Although first observed in 1911, superconductivity remained in the decade following the invention of quantum mechanics a conspicuously stubborn and insoluble problem. Between 1929 and 1933, more than a dozen theoretical physicists, including Bohr, Pauli, Heisenberg, Bloch, Landau, Brillouin, W. Elsasser, Frenkel, and Kronig, armed with the successes of the quantum theory of metals and new observations, were optimistic that the new tool would also help them to explain superconductivity. In their approaches to superconductivity, these theorists portray their confidence in the power of the new mechanics, even in the face of continual frustration at the failure of their theories to agree either with experiments or with theories proposed by colleagues. As Bethe lamented in 1933 on the failure of the quantum theory to explain superconductivity, compared with its success with normal conduction, in superconductivity “only a number of hypotheses exist, which until now have in no way been worked out and whose validity cannot therefore be verified” (SB, p. 555).

To illustrate we examine two of the most prominent conceptions then under discussion: the spontaneous current theories of Bloch, Landau, and Frenkel, centering on the notion of a current-bearing equilibrium state (1929–1933), and the electron-lattice or electron-chain theory of Bohr and Kronig (1932–1933). To suggest the larger picture of research on superconductivity, we also sketch three of the other theories in the air at this time: Elsasser’s (1932), in terms of relativistic electrons, R. Schachenmeier’s (1932), based on exchange between conduction and bound electrons (1932), and Brillouin’s (1933), in which superconductivity is associated with electrons “trapped” in metastable states.

⁶⁴For a history of the development of the theory of collective phenomena, see Hoddeson *et al.* (1987b).

Available experimental information on superconductors in the 1920s was quite incomplete.⁶⁵ The outstanding feature of superconductors—whose understanding was the major theoretical focus—was the loss of resistivity at very low temperatures, a loss which, as Kamerlingh Onnes and co-workers in Leiden demonstrated following their initial discovery, was indeed total.⁶⁶ By 1913, the disappearance of superconductivity in strong magnetic fields was discovered (Kamerlingh Onnes, 1913b; Tuyn and Kamerlingh Onnes, 1925), and realized soon after to be an effect not of local heating in “bad places,” but rather of the field itself.

In 1923 J. C. McLennan established in Toronto the second cryogenic laboratory (after Leiden) to engage in superconductivity research,⁶⁷ and in 1925, when experimentalists headed by Walther Meissner at the Physikalisch Technische Reichsanstalt (PTR) in Berlin liquefied helium (Meissner, 1925), the PTR became the third such laboratory. While Leiden and Toronto experiments centered on the phenomenology of superconductivity, including effects of magnetic fields and the changes of properties through the superconducting transition, the initial PTR program concerned the problem of “whether all metals become superconducting” (Meissner, 1925, p. 691). Within 3 years, Meissner’s group had analyzed 40 metals, adding to the list of the superconducting materials tantalum, titanium, thorium, and niobium; in 1929, they also measured chemical compounds and alloys and found that even materials composed of insulating and nonsuperconducting metals, e.g., copper sulfate, could become superconducting, indicating that superconductivity was not simply an atomic property (Meissner, 1928, 1929). Evidence that superconductivity was not a solid-state “dirt effect,” like normal conductivity, was provided by Leiden measurements that showed that superconductivity did not depend essentially on the purity or crystalline order of the material (Sizoo, 1926); Meissner, on the other hand, found that even the purest single crystals of a normal conductor like gold do not necessarily become superconducting when

cooled down to temperatures as low as 1.3 K, the PTR’s lowest operating temperature (Meissner, 1926). Adding to the mystery was the Leiden observation that heat conduction, which is primarily by electrons, remains continuous through the superconducting transition, although the electrical conductivity becomes infinite (de Haas and Bremmer, 1931).

Two essential features of superconductors were learned too late to influence microscopic work between 1929 and 1933. The first, as revealed in 1933 in the classic experiment of Meissner and Robert Ochsenfeld (1933; see also Meissner, 1934) in Berlin—published in October 1933 in *Die Naturwissenschaften*—is that they expel magnetic flux;⁶⁸ this effect would suggest that a more fundamental characterization of superconductors was perfect diamagnetism, rather than vanishing resistivity. The second, which would come into focus between 1932 and 1934 through the work in Leiden of Willem Keesom, J. A. Kok, and others (e.g., Keesom and Kok, 1932; Keesom, 1934), as well as Ehrenfest (1933), A. J. Rutgers (1934), Cornelius Gorter, and Hendrik G. B. Casimir (Gorter and Casimir, 1934a, 1934b, 1934c), was that the transition to the superconducting state was reversible, and the superconducting state—unique only if, as demonstrated by Meissner and Ochsenfeld, flux was not frozen into superconductors—could be described by thermodynamics.

The handful of theoretical attempts to understand superconductivity from microscopic principles prior to the development of quantum mechanics was limited by the failure to understand the behavior of electrons in normal metals.⁶⁹ For example, F. A. Lindemann (1915) and J. J. Thomson (1922) constructed theories of ordered electron structures which avoided the electron specific-heat dilemma, at the same time providing a model for superconductivity. Lindemann’s theory of normal and superconductors rested on the hypothesis “that far from forming a sort of perfect gas the electrons in a metal may be looked upon as a perfect solid;” thus, à la Born—von Kármán, the electron specific heat became greatly reduced from its classical value. Superconductivity followed; if the repulsive force between the electrons and ions were sufficiently short ranged and the ionic motion small, as might occur at low enough temperatures, “the electron space-lattice can move unimpeded through the atom space-lattice.”

⁶⁵The experimental situation in superconductivity circa 1928 is discussed by Grüneisen (1928); for the early 1930s see Meissner (1932), and Grayson Smith and Wilhelm (1935). A more complete discussion of the early period of superconductivity, as well as further references, is given in Dahl (1984, 1986) and in Hoddeson, Baym, Heims, and Schubert (1987). We are grateful to H. Schubert for allowing us to draw extensively upon his efforts in this section.

⁶⁶See, for example, Kamerlingh Onnes (1914) and Tuyn (1929). W. J. de Haas and J. Voogd (1931a) refined the characterization of superconductors in 1931: “We therefore regard the vanishing of the resistance within a few hundredths of a degree as the most characteristic phenomenon of supraconductivity in pure metals.”

⁶⁷For a description of McLennan’s laboratory, see McLennan (1923). Work of the laboratory is reviewed in Burton (1934).

⁶⁸Mrs. G. L. de Haas-Lorentz (1925) had in fact already discussed the penetration depth of superconductors, starting from the question of whether a magnetic field, held completely outside a superconductor by screening currents, can exert influence on the superconductor. Having been published in Dutch, the work remained largely unnoticed prior to the discovery of the Meissner-Ochsenfeld effect.

⁶⁹Assorted early theories are reviewed by Kretschmann (1927). This article, written as Sommerfeld was developing the semiclassical free-electron theory of metals, does not benefit from the perspective of that theory.

Thomson's attempt to explain metallic conduction was expressed in terms of "chains of electrons lying along a line of a lattice . . . traveling along that line carrying energy and electricity from one part of the solid to another." As in Lindemann's theory, the ordered electrons would have a greatly reduced specific heat. Furthermore, "the amount of energy communicated" between the chain and lattice, Thomson writes, "will fall off very rapidly as the ratio of the duration of the collision to the time of [atomic] vibration increases. . . . thus when the temperature gets so low that the time taken by an electron to pass [an interatomic distance] is comparable with the time of vibration of the atom, any diminution in the temperature will produce an abnormally large increase in the conductivity, and thus the metal would show the superconductivity discovered by Kammerlingh [*sic*] Onnes." Unlike Lindemann, who attempted to explain perfect conductivity, Thomson predicted greatly enhanced normal conductivity. The electron space-lattice and chain concepts, a foreshadowing of the need for collective ordering of the electrons in superconductors, would reappear in Bohr's and Kronig's later theories of superconductivity.

Albert Einstein, describing problems of electron conduction in metals at the meeting in Leiden on 11 November 1922 to celebrate the 40th anniversary of Kamerlingh Onnes's professorship (Einstein, 1922; Yavelov, 1980), shared the need to avoid free electrons: "it looks as though, according to today's state of our knowledge, free electrons do not exist in metals at all." In superconductors, "it seems unavoidable that supercurrents are carried by [electron transfer along] closed molecular conduction chains." However, he remarked with great foresight, "with our far-reaching ignorance of the quantum mechanics of composite systems we are very far from being able to compose a theory out of these vague ideas.

By the end of the 1920s quantum theory had developed to the stage where solving superconductivity seemed a more realistic goal. The turning point was again Bloch's thesis in mid-1928, which put the theory of the conductivity of normal metals on a firm foundation and strongly suggested, through his calculation of the low-temperature resistivity, that perfect conductivity could not be obtained simply from using the single-electron approach at extremely low temperatures. [Bloch did not in his thesis attempt to deal with superconductivity, except to remark at the very end, after illustrating "the possibility of a transition between two completely different laws of conductivity" (that between degenerate and nondegenerate electrons), that "the phenomenon of superconductivity shows that such a transition actually occurs, which . . . remains up to now not clarified."] Bloch's thesis had an immediate impact on Bohr, who, with his standing interest in the electron theory of metals since his own thesis in 1911, was apparently thinking about solving superconductivity from a single-electron picture. As Bohr wrote to Heisenberg at Christmas 1928: "How have you been doing with superconductivity? Bloch's beautiful work, which you so kindly sent me, and from which I had much pleasure, taught me of course that the way out which I indicated was not

possible"⁷⁰ (Bohr, 1928). He would make another attempt three years later, which would again fall victim to Bloch.⁷¹

Bloch began thinking seriously about superconductivity when he moved from Leipzig to Zurich in autumn 1928, soon after finishing his thesis, to become Pauli's *Assistent* (Bloch, 1981). His theory, which he never published,⁷² shared with one that Landau formulated contemporaneously and finally published in 1933⁷³ (Landau, 1933) the key idea that in equilibrium the thermodynamically favored superconducting ground state, corresponding to a minimum of the free energy, bears a finite spontaneous current below the critical temperature, while at higher temperatures current-free equilibrium states have statistically greater probability.

Pauli's attitude towards this work, Bloch recalled, was "get on with it so as to be finally done with all these 'dirt-effects'" (Bloch, 1980). "Pauli, after all, was a physicist, and as such he could not entirely ignore the interesting problems in solid-state physics, but he didn't really have his heart in it" (Bloch, 1981). However, Pauli himself was clearly involved in superconductivity, telling Bohr in January 1929, "On the question of superconductivity I could not come to any definite result" (Pauli, 1929c). He proudly described Bloch's pursuit of superconductivity in numerous letters in the spring of 1929: In March to Oskar Klein, who had succeeded Heisenberg as

⁷⁰We thank David Cassidy for alerting us to this communication. According to Bloch (1981), "Heisenberg tried [superconductivity] at one point . . . [with] some idea of condensation in angular momentum space." Heisenberg did not publish on superconductivity in this period, but did return to the problem after the war; see Heisenberg (1947, 1948, 1949).

⁷¹In Bohr (1932a), the article on superconductivity that he sent to *Die Naturwissenschaften* and then withdrew in the proof stage, Bohr remarks, immediately after discussing Bloch's conductivity work, "On the basis of the independent-electron picture, no explanation can be given for Kamerlingh Onnes's discovery." Bohr's notes and correspondence on superconductivity in this period are collected in the Bohr Scientific Manuscripts (cited as BSM), AHQP (see footnote 90).

⁷²Since Bloch's work was never published, our knowledge of it comes from references by others—e.g., a short description by Bethe (SB, Sec. 44, "Ansätze für Theorie der Supraleitung," pp. 555–558) of the idea "Bloch and Landau suggested," Landau (1933) and Brillouin (1935)—as well as Bloch's interviews and retrospective articles, e.g., Bloch (1980, 1966).

⁷³Referring in his 1933 paper to his 1929 work, Landau wrote "similar thoughts were also simultaneously expressed by Bloch." Landau, we recall, worked briefly at Pauli's institute in late 1929, and although Bloch was based in Leipzig at the time, he did turn up in Zurich for visits, possibly giving the two opportunity to discuss their similar explanations of superconductivity. Bloch recalls that he conceived of his idea before he came to work with Pauli, but that he and Landau did not communicate about their related notions for some time (Bloch, 1981).

Bohr's assistant in 1927 in Copenhagen, Pauli reported that "Bloch here has made progress with a theory of superconductivity. I will not assert that he has already succeeded in finding an explanation... but his results bid fair hopes. In any case, I now believe that the way conjectured by Bohr last fall was completely false" (Pauli, 1929d). In a postcard to Bohr the next month: "Bloch's theory of superconductivity seems to be getting very beautiful!" (Pauli, 1929e). To Munich he wrote in May, "Now I have a rather big enterprise here in Zurich. Mr. Bloch is busy working out a theory of superconductivity. The job isn't finished, but it seems to be working" (Pauli, 1929f). In the same letter, he also refers to the Peierls study of heat conduction in insulators, which both Peierls and Pauli hoped might provide a model for superconductivity. And finally, to Kronig in June, "Bloch is pursuing superconductivity and is altering his theory on a daily basis (Thank God before publication!)" (Pauli, 1929g).

Both Bloch and Landau felt that ferromagnetism and superconductivity were closely joined phenomena. The currents of ferromagnetism and the currents of superconductivity "both persisted... there must be a common cause" (Bloch, 1981). Indeed, Bloch worked on the two problems at the same time, with the thought in the back of his mind that the answer to both lay in the electron-electron interactions. As he later wrote, an "appealing interpretation [of superconductivity] was suggested through analogy with ferromagnetism, where remanent magnetization had been explained by recognizing that parallel orientation of the magnetic moments of the atoms leads to a lower energy than random orientation. Similarly, it seemed plausible to interpret current flow in a superconductor as the result of a correlation between the velocities of the conduction electrons that is energetically favored and, therefore, manifests itself at sufficiently low temperatures" (Bloch, 1966, p. 27).

However, Bloch made far less progress in superconductivity than in ferromagnetism. In each of many calculations, he found that the minimum energy state bore no current, a result which came to be known as Bloch's first theorem on superconductivity. Brillouin later stressed the importance of this theorem at the May 1935 meeting of the Royal Society of London, since it "practically forbids any interpretation of superconductivity within the frame of classical theory" (Brillouin, 1935). The point, as Brillouin had demonstrated with a simple classical argument (Brillouin, 1933b), is that were any current to flow, one could always decrease the total energy by applying a potential difference of one sign or the other across the conductor for an instant; thus, the energy could not have been at a minimum. (The only way out, Brillouin realized, was for the current not to be "stable but only metastable," which seemed ruled out since "Meissner's experiment proved decisively that supra-currents were stable." Missing at this stage was the understanding of supercurrent-carrying states as effectively-stable local minima.) So stymied was Bloch that in exasperation he formulated his now-famous tongue-in-cheek second theorem, that every theory of superconductivity can be

disproved. The frequency with which this unpublished theorem was quoted indicates its appeal to other physicists studying superconductivity in this period (London, 1935, see p. 25; Bethe, 1981a).

Landau had little more success. Pushing the analogy between superconducting and ferromagnetic states to the fullest, he assumes in his published article (Landau, 1933) that superconductors contain local "saturation currents" which flow in different directions, producing no net current unless organized by an applied field. Landau provides no microscopic justification for such a picture, and indeed his paper is not based on quantum mechanics; rather he assumes a phenomenological description of the energy as a sum of the spatially-varying local magnetic field energy plus a term "designated, in correspondence with the density gradient term in the theory of surface tension, as the capillary term," proportional to the curl of the local current [where the Londons (London and London, 1935) would later use the current itself] in the superconductor. While the analogy with ferromagnetism (and the theory of surface tension) turns out to be false, the paper remains interesting for the seeds it contains of Landau's later work on phase transitions and the Ginzburg-Landau theory of superconductors (Ginzburg and Landau, 1950), particularly the expansion of the free energy near the transition temperature T_c in terms of an order parameter in both those theories. Here Landau expands the free energy near T_c in the magnitude i of the saturation current, $F = F_0 + ai^2/2 + bi^4/4$, with the familiar assumption that the coefficient a changes from positive to negative as the temperature is lowered through T_c . Although the order parameter of superconductivity is misidentified, the theory gives the correct qualitative behavior of the entropy and specific heat near T_c . Landau also ventures the possibility that the orientable internal magnetization in his model can lead to magnetic flux expulsion up to a particular external field strength. Whether he knew of the work of Meissner and Ochsenfeld, published over half a year after his paper was submitted, we do not know. The paper provides no internal evidence; Landau characteristically gives few references in his papers. He concludes with a hint that his theory is unsatisfactory, as it suggests a $(T_c - T)^{1/2}$ behavior of the critical magnetic field, compared with the linear dependence on temperature near T_c recently observed (de Haas and Voogd, 1931b).

Frenkel in Leningrad put forth at the end of 1932 a related version of the spontaneous-current theory (Frenkel, 1933), arguing that the magnetic forces between electrons—forces "totally neglected hitherto in the electron theory of metals"—would encourage them to move in stable parallel streams rather than randomly, yielding a local current. Deducing that the magnetic interactions produce a large electron effective mass, he concludes that "so long... as the electrons in a metal move collectively as an organized crowd of sufficiently large size, their motion can remain unaffected by the heat motion of the crystal lattice, the quanta... of the heat waves being insufficient to knock out even a single electron." In analo-

gy with ferromagnetism, he also presents the picture that there exist, "in a superconducting body, regions with whirl currents whose orientations . . . vary in an irregular manner from one region to another." Frenkel too ventures that a metal in the superconducting state in an external field "must behave like a *diamagnetic* body with a large negative susceptibility . . . the interior of such a body will be screened from external magnetic fields by the system of surface currents induced by the lattice." The Meissner effect, reported in October 1933, would verify this intuition. Two months after Frenkel's theory was published, Bethe and Herbert Fröhlich (1933) severely criticized it in a paper sent to the *Zeitschrift für Physik*, showing by a more precise argument that the magnetic interactions lead to only a tiny correction of the effective mass, and that "all the formulae of the usual conductivity theory remain fully in force."

The idea of spontaneous currents as states of lowest electron energies was, according to Bethe, in the 1933 *Handbuch* article, "extremely tempting; however, until now there has been no success in constructing a model with the required properties" (SB, p. 556). An alternative approach, adopted independently by Bohr and Kronig, was to treat supercurrents as a coherent motion of the entire ground-state electron distribution, a quantum resurrection of the earlier Lindemann-Thomson models. The lively correspondence between Bohr, Bloch, and Kronig between June 1932 and January 1933 on superconductivity provides a rare insight into this development, showing in particular how, through Bloch's persistent criticism, Bohr lost confidence in his theory, as eventually Kronig did in his.

Bohr had returned to superconductivity in the spring of 1932. In mid-June he wrote to Bloch to try out his thoughts, "I would awfully much like to talk with you on several questions concerning metallic properties, . . . namely a thought touching on superconductivity which I got and cannot get away from, even if I am far from understanding the connection between superconducting properties and ordinary electric conduction." His idea is that "superconductivity concerns a coordinated motion of the whole electron lattice," the many-electron wave function of a supercurrent-carrying state being only a slight (effectively long-wavelength) modulation of the normal ground-state wave function. Because the electron motion involves long wavelengths, "the current will not be significantly disturbed by the thermal oscillation of the metallic lattice. . . . The transition to a state where the electrons move uncoordinated between one another should be somewhat analogous to a melting of a solid body, but it has not been possible so far for me to make my understanding of the process of this transition clear." Expressing his faith that the solution to superconductivity lies in the new mechanics, he writes, "Just as quantum mechanics has first made it possible to bring the picture of 'free' electrons in metals in closer correspondence with experiment, it seems also that first through quantum mechanics . . . can one understand how the two lattices can move through each other without resistance and sig-

nificant deformation. I should be very happy to hear a few words from you on how you look upon all this" (Bohr, 1932b).

Bloch's reaction was rather doubting. On his way to Brussels in late June to arrange the next Solvay conference, Bohr met with Bloch in Berlin to discuss his theory. As he related to Delbrück, "I had a very lively discussion with Bloch . . . and I think that I succeeded in some way to overthrow his scepticism." He also writes that "I have the day before yesterday in Liege with Rosenfeld's help written a little article which I have sent to *Die Naturwissenschaften*" (Bohr, 1932c). The article, entitled "Zur Frage der Supraleitung" and dated June 1932, spells out in more detail the basic conception he wrote to Bloch, and gives estimates of the magnitudes of the supercurrent associated with his modified wave function, from the limits of nearly-free to tightly-bound electrons. Bohr acknowledges "illuminating discussions" with Bloch and Rosenfeld. Although the paper was accepted for the 11 July issue (Bohr, 1932a), Bohr, as a result of Bloch's continuing objections, withdrew the article in the proof stage.

An undated handwritten note by Bloch (1932b) at this time reveals the depth of this criticism of Bohr's theory; it suggests that Bohr "let it lie, on account of doubts: (1) experimental facts, a) magnetic fields (could possibly be understood), b) McLennan's experiments [on the superconducting transition in the presence of alternating currents] (cannot be explained). (2) Proof of the general existence of current-carrying solutions doubtful. Difference between conductors and insulators are not sufficiently considered. . . ." He continues, "(3) The concept could become meaningful since it deals with bringing the secured features of atomic mechanics into agreement with the empirical facts of superconductivity. (4) The concept is basically different from attempts to treat the interactions of the electrons as a rigid lattice in an intuitive [classical] picture. The latter is ruled out because the melting energy would be too high. The picture of a moving lattice is neither in agreement with the binding of the electrons to the ions nor with its motion. (5) The change of state at the critical point should not be determined by typical quantum-mechanical features in an obvious way."

Bloch's letters to Bohr are more deferential. In mid-July, he wrote from Leipzig telling how "very happy I also was about the discussions in Berlin and welcome very much that you are letting a note about superconductivity appear. . . . My comments then were not meant as 'negating criticism,' rather I consider it entirely possible that your ideas really contain the solution of the riddle. I merely wanted to point out that one needs to be somewhat more careful in the evaluation of the order of magnitude of the currents and believe, so far, that the discussion of the model of tightly bound electrons has *something* to do with the problem. In any case, it would interest me a great deal to see the proofs of your note!" (Bloch, 1932c). Then, 2 weeks later, Bloch sent Bohr detailed remarks on his manuscript, adding, "I'm quite in agreement with the whole 'tone' of the note and perceive only the lack of more precise conceptions still somewhat unsatisfactory."

He also asks Bohr if he would hold up publication until October when he can come to Copenhagen (Bloch, 1932d). Bohr took Bloch's comments seriously, and in early September wrote back that "I have held back my article on superconductivity because I had second thoughts about it all on grounds of the paradoxes that show up when one treats the lattice field as fixed in handling the electron system" (Bohr, 1932d).

As the discussion between Bohr and Bloch ensued, Kronig in Groningen developed a similar electron lattice theory, which he subsequently published in two papers (1932a, 1932b) in the 1932 *Zeitschrift für Physik*. The first (received 31 August) proposes that the interactions between electrons lead to their forming a rigid lattice intermeshed with the ionic lattice. The electron system can be superconducting since "in analogy with Bloch's theory for a single electron, translation [by an electron lattice constant] of the whole electron lattice can experience no resistance."

On learning of Kronig's theory from McLennan in mid-October, Bohr wrote (1932e) to Kronig, enclosing a copy of the proofs of his paper and inviting him to Copenhagen to talk with Bloch, Rosenfeld, and himself. He remarks that he delayed returning the proofs in part because of his difficulty in bringing his theory into agreement with McLennan's experiments on the dependence of the superconducting transition temperature on the presence of high-frequency alternating currents, but after discussing McLennan's most recent results with Bloch and Rosenfeld, he felt it would be right now not to wait longer to publish his note, possibly in somewhat altered form.⁷⁴ Kronig [whose early invention of the spin of the electron met resistance from Pauli and Heisenberg, to Bohr's later "consternation and deep regret" (Bohr, 1926)] wrote assertively to Bohr the next day that the new physical content of Bohr's paper is in fact "covered" by his own, and comments parenthetically, "My result is somewhat more specialized but offers... more prospects for quantitative

evaluation." He adds, "Unfortunately, it was scarcely possible to mention your work, even in an added note, since I had already completed the corrections. . . . I enclose a copy of the proof, since you can best see the content of my conceptions in that" (Kronig, 1932c).

The next week Kronig visited Copenhagen,⁷⁵ and the discussion between Bohr, Bloch, and Kronig went into full swing. Bohr and Bloch raised the objection that for the electron lattice to carry a current it would have to be able to tunnel through the N (the number of electrons) potential hills between lattice sites, which becomes impossible as N grows large. Kronig responded (1932d) after returning to Groningen, involving the zero-point motion of the electrons, and at the same time presenting Bohr with a mathematical argument (based, it would appear, on a faulty expansion of the wave function in the magnitude of the current carried) that Bohr's proposed current-carrying wave function, a modulation of the ground state, could not be a steady-state solution of the Schrödinger equation. Kronig also mentions that he is submitting a second paper, and has sent a copy to Bloch asking him to send it on to Bohr.

Bloch sent Kronig's manuscript to Bohr, as requested, a few days later with frank comments on Kronig's idea of a large electron zero-point motion allowing it to overcome the potential hills: "I would like straightaway to . . . make you aware of the point in Kronig's work which seems to me is wrong. Kronig discusses the case of the linear electron lattice and finds that the mean square ϵ^2 of the zero point amplitude grows as $\log N$. . . [and] that the matrix elements of the interaction with the ion lattice . . . under circumstances become independent of N or even disappear with growing N . To that one can remark: 1) If the zero point amplitudes really become so large, then one can naturally no longer speak at all of a lattice. 2) The Kronig result is very specially tied to the one dimensional case." He goes on to explain, using arguments familiar from his spin-wave paper, that "The logarithmic growth of ϵ^2 with $\log N$ comes . . . mainly from the long elastic waves which, on account of their small frequency, have a very large zero point amplitude. In the two and three dimensional case, the long wavelength waves, on account of their relatively small number, play practically no role, and one can also easily show that then for the largest values of N that ϵ^2 becomes independent of N . (It would also be sad if it were other, for then an NaCl lattice even at absolute zero would fall apart)" (Bloch, 1932e).

Kronig wrote again to Bohr a week later (1932e), acquiescently: "Mr. Bloch pointed out to me, and with full justification, that the given conception cannot be carried over to the three dimensional case. . . . There are then the following possibilities: 1. The idea of the electron lattice is entirely unusable. 2. Electron exchange saves the

⁷⁴McLennan's experiments (McLennan *et al.*, 1932a, 1932b) showed a decrease of the superconducting transition temperature in the presence of an alternating current. The issue here was whether these experiments indicated that "superconductivity is not, as assumed, a property in a metal at low temperatures independent of the presence of a current, but that it must be closely knitted to the current mechanism itself" (Bohr, 1932f), and thus does not arise simply from a structure such as an electron lattice, described in terms of a modulation of the ground-state wave function. The second McLennan paper (1932b, received 12 September 1932) discusses the high-frequency currents as "confined to the outside of the . . . wire" by the skin effect, unlike in the first paper, where a connection with the skin effect is dismissed; the fact that McLennan's effect was not a bulk phenomenon seemed to Bohr to have made the experiments a less crucial test of his conception. It was not realized, of course, before the Meissner effect, that supercurrents are actually carried in the surface, rather than by bulk motion of the electrons.

⁷⁵Shortly after Kronig's visit, Bohr (1932f) drafted several versions of an addendum to his article to explain the differences between his and Kronig's theory.

three dimensional case. . . . 3. Superconductivity must not be ascribed to a translational movement of all electrons but rather to the building of a one-dimensional electron lattice." He goes on to mention a note from Meissner, which gives the first indications of his experiments on penetration of fields in superconductors: "Mr. Meissner writes further: It follows, that in the vast majority of cases the superconducting current in all probability is a surface-current flowing on the surface of conductors, so that only the outermost electrons can be displaced by the electron lattice. . . . This would be in accord with the chain picture. I have asked Mr. Meissner to tell me in detail how he comes to his conception, but have up to now received no answer."

The concept of a "translation of individual linear chains of electrons in the lattice" would be the basis of the revised version of Kronig's second paper on superconductivity, published at the end of the year (Kronig, 1932b). As he wrote to Bohr (Kronig, 1932f) in mid-December, "On the basis of my mathematical considerations, this change of conception in fact appears to me altogether unavoidable." He remarks that, "As before I would like to believe that the transition point corresponds to a phase change in which the conduction electrons go over from an unordered distribution, similar to a liquid to an ordered state." He also inquires whether Bohr is considering publishing something of his original note, or whether he would prefer to "shroud the existence of this unpublished work in the cloak of silence."

Bohr replied to Kronig just after Christmas, reiterating, "As far as I can see, the case stands just as we discussed it in Copenhagen. . . . It remains my conviction that the difference between the two phases corresponding to superconductivity and usual metallic conductivity is a purely quantum problem, which quite escapes visualization by means of basically mechanical pictures, and on grounds of the large differences in our conceptions is it difficult for me to agree with details in your second treatment"⁷⁶ (Bohr, 1932g). A few days later, Bloch in writing Bohr provided a more technical coup de grâce: "As far as I have understood your letter to Kronig, I am completely in agreement; the more I have thought about it, the more I consider the Kronig work to be in error. The last hope which Kronig places in . . . the one-dimensional lattice is unjustified, because the condition, which according to Kronig would cause the overtaking of the potential hill,

simply means that the zero-point oscillations are so big that this lattice cannot exist. I do not know whether Kronig at the time I wrote to him about it understood me completely. In any case his answer allowed the possibility of giving up the idea of the electron lattice" (Bloch, 1932f).

Neither Kronig nor Bohr progressed further with electron-lattice theories of superconductivity. Even while making no substantial progress himself, Bloch through his role of critic, emerged from the debate as the main authority of the time in the microscopic theory of superconductivity. In his summary of superconductivity in the 1933 *Handbuch* article (SB, pp. 556 and 557), Bethe did not discuss Bohr's work, but suggested that despite flaws in Kronig's theory (brought out earlier by Bloch), it might, "with substantial deepening of its foundations," be the basis of a useful theory.⁷⁷

Solutions to superconductivity in this period were sought in many other directions. Brillouin in 1933 hypothesized that it would occur in a metal in which the curve of single-electron energy as a function of crystal momentum contained secondary minima (Brillouin, 1933a, 1933c). His argument was the following: an electric field applied to a metal induces a current; at ordinary temperatures, when the field is removed, the electron distribution in momentum space becomes symmetric through coupling of the electrons to the lattice vibrations,⁷⁸ and the current induced by the field disappears. However, at very low temperatures this symmetry restoration mechanism, for electrons in the secondary minima, is forbidden for kinematical reasons; such electrons are metastably trapped, producing a persistent current, "which appears to me to represent the essential character of superconductivity." As the temperature is raised, the exponential onset of high-frequency lattice vibrations destroys the metastability and the persistent current disappears, but such a mechanism could not explain why the transition to superconductivity is sharp, nor in fact why the residual resistance due to impurities also disappears at the critical temperature. When Bloch, leaving Nazi Germany, passed through Paris in June 1933, he invoked his "first theorem" in pointing out to Brillouin that the hypothesized metastable states would be unstable against a small common displacement of all the electrons in momentum space. Brillouin added a short appendix to his paper describing Bloch's objection, but could not resolve the issue; later in the year he published an incon-

⁷⁶In Bohr's letter, he counters Kronig's argument against his wave function by explicitly constructing a small current state for a single electron in a one-dimensional periodic potential, a result Kronig (1933) pointed out to Bohr did not agree with exact solutions for his model with Penney. This last argument of Kronig clearly lingered with Bohr, for as J. R. Schrieffer (1986) recalls, Bohr remarked to him in Copenhagen in 1958, during a discussion of the recently developed BCS theory of superconductivity, in essence, "We must go back to fundamentals, and first understand the Kronig-Penney model."

⁷⁷Frenkel (1934) was one of the few to attempt to extend Kronig's electron-chain theory, describing supercurrents as a collective electron motion—"like a chain gliding over a toothed track." The concept of collective electron-lattice motion, although not applicable to superconductivity, has had a rebirth in modern theories of charge-density waves in solids.

⁷⁸In this paper Brillouin finally recognizes Umklapp processes, "transitions anormales de Peierls," as a possible relaxation mechanism.

clusive analysis of whether perturbations can lead to the degradation of a supercurrent, as proposed by Bloch (Brillouin, 1935).

Elsasser (1933), in Frankfurt, suggested in early 1932 an explanation in terms of Dirac's new relativistic electron theory, in which superconductivity arises from a term he extracts in the relation of an individual electron's velocity and momentum proportional to the curl of the electron spin density (not quite the analogous term in the Gordon decomposition of the Dirac current); this term he claims represents an "unmechanical . . . momentumless" current transport. However, Bethe pointed out that on symmetry grounds the expectation value of this term is zero in equilibrium. He comments more generally: "It seems certain that one cannot succeed by considering only single-electron energies," a point he illustrates with the argument that any two single-electron states with wave vectors k and $-k$ have equal energies. Each surface of a given energy in wave-number space thus "contains as many single electron currents oriented to the right as to the left. The total current of all electronic states belonging to a certain energy, therefore, is always zero" (SB, p. 556).

Schachenmeier (1932), in Berlin, tried around the same time to understand superconductivity in terms of electron-electron Coulomb interactions giving rise to "resonant" exchange between conduction electrons and those bound to ions. For resonance frequencies above the maximum thermally excited lattice frequency, he argued, electrons would move through the vibrating lattice without being scattered by irregularities, thus forming a supercurrent. The critical temperature, calculated in terms of the lattice Debye frequency, gave order of magnitude agreement with experiment. Schachenmeier too would be taken to task by Bethe (somewhat to Bethe's later despair, and eventual amusement), who pointed out that he, as well as Kretschmann (1932), "neglect fundamental facts of wave mechanics."⁷⁹ Both, e.g., make a distinction between the valence electrons which are bound to individual atoms and "free" electrons" (SB, p. 558), through failure to antisymmetrize the total electron wave function. Despite its execution, Schachenmeier's paper, an early attempt to employ quantum-mechanical electron-electron interactions to explain superconductivity, hints at modern ideas of mixed valences.

Bethe's evaluation had a portentous postscript. Schachenmeier would not accept Bethe's criticism, and as Bethe wrote to Sommerfeld in late February 1934 from Manchester, "Schachenmeier is busy writing letters to me, which, much more briefly than he, I nevertheless respond to" (Bethe, 1934a). Two days later, Walter Henneberg, a former Sommerfeld student who was now

working at the AEG in Berlin, reported to his old teacher: "Recently, I had a turbulent debate with Professor Schachenmeier; he is very insulted by Bethe's criticism. . . . I found it very alienating that Schachenmeier reproached Dr. Bethe for being abroad." In fact, Bethe had by this time fled Nazi Germany. Henneberg continues, "Sch's opinion is that Dr. Bethe wrote the article with the certain prospect in mind that he would go abroad and thus allow himself to be insulted. I don't believe that I succeeded in convincing Schachenmeier that Dr. Bethe had completed the article at a time when he did not yet think of going abroad. . . . Schachenmeier as an Aryan is advantaged from the beginning and my impression is that he will make use of that. Since Schachenmeier is working in our research institute (nobody knows however what he is doing), I would like you to treat my communication as confidential" (Henneberg, 1934).

Schachenmeier's refusal to recognize Bethe's criticism went so far as his mentioning Bethe only in a note added in proof to a further paper (1934a) on superconductivity, which he submitted at the end of March 1934, claiming in the note that the *Handbuch* article appeared after his writing the paper. Schachenmeier was also apparently quite disturbed that Bethe had only referred to him in small type in the *Handbuch* article, giving Bethe the irresistible opportunity to answer Schachenmeier a few months later in the *Zeitschrift für Physik* in an article written almost entirely in small type (Bethe, 1934b; see also Schachenmeier's reply, 1934b).

Like the electron theory of metals just before Sommerfeld's work, or atomic theory preceding the invention of quantum mechanics, the microscopic understanding of superconductivity circa early 1933 was but a hodgepodge of partial explanations containing *ad hoc* assumptions of questionable validity. None of the theories of superconductivity by this time was sufficiently developed to permit quantitative comparison with experiment. That the explanation depended on many-body interactions was recognized, but not properly dealt with. While the application of quantum mechanics to solids had culminated by 1933 in a remarkably successful theory of transport phenomena and a promising theory of ferromagnetism, superconductivity remained a phenomenon untreatable within the existing quantum-theoretical framework.

Did the obvious failure to explain superconductivity in quantum-mechanical terms suggest that in certain circumstances the quantum theory might be inapplicable, as happens in classical mechanics when velocities approach the speed of light or when quantum-size actions are involved? There is no evidence that any leading theorist in the period drew such a conclusion. While in late 1926 outstanding solid-state problems, such as spin paramagnetism, electrical conductivity, and specific heat, were used as a proving ground for the newly-invented quantum mechanics, 6 years later the one remaining fundamental problem in the quantum theory of metal was not seen as such a test. Rather, as Bohr, for example, correctly emphasized in his correspondence with Kronig, current

⁷⁹As Peierls (1977) described it, Kretschmann seemed fair game for criticizing; he was "generally known to be somebody who quibbled about the current theory without really understanding it. I don't know whether that's a fair assessment, but that was our impression at the time."

models were assumed to be still too crude to deal with the full quantum problem. Bethe summed up the prevailing attitude in the 1933 *Handbuch*: “despite being unsuccessful up until now, we may assert that superconductivity will be solved on the basis of our present-day quantum-mechanical knowledge” (SB, p. 558).

CONCLUSION

The year 1933 marked both a climax and a transition in the development of the quantum theory of solids. As the laying of theoretical foundations reached a temporary conclusion, attention began to shift from general formulations to computation of the properties of particular solids. Institutionally, solid-state physics began to show signs of independence, while the community of physicists working in this area underwent rapid change.

A surge of review articles in the years 1931 to 1934 (e.g., Brillouin, 1931; Peierls, 1932a; Bloch, 1933; SB; Slater, 1934; Nordheim, 1934) made the theory accessible to the students who would form the first generation of solid-state specialists. The Sommerfeld-Bethe review would in particular serve as their “Bible” for more than 2 decades.⁸⁰ The first textbooks on the quantum theory of solids were also conceived in this period: among them that by Brillouin (1931), based on his articles in 1927–1929 on the application of quantum statistics;⁸¹ by Wilson (1936), based on his thesis; and by Mott and Jones (1936),⁸² which remains an everyday reference. The reviews projected the feeling, then widely held (Peierls, 1981a; Bethe, 1981a), that the fundamental theoretical issues in the quantum theory of solids were for the most part resolved. “One gains the impression that its problem,” Peierls wrote (1932a) of the electron theory of metals, “to explain the typical conditions of metals from molecular properties, and to derive the quantitative laws that exist is, with exceptions . . . solved;” similarly Bloch (1933) remarked, “the electron theory of metals in so far that it is based on the conception of independent conduction electrons allows one to understand qualitatively or

quantitatively the most important properties of metals.”

This confidence in the theory was, however, tied to the complementary belief that the subject was becoming less manageable. For example, Bethe wrote to Sommerfeld in 1931 that he doubted much more could come of certain detailed calculations until the eigenfunctions of the metallic crystal are found, an “almost hopeless” prospect—“There is no sense to calculate further, since the Bloch theory represents too coarse an approximation” (Bethe, 1931d). Peierls reflected recently, “There you had a situation where there was a breakthrough, and you exploited that, and when it was finished, you had explained all the outstanding paradoxes and seen that things work in general, then it became less exciting. To do new things which were not just routine applications became harder, as it does in every subject” (Peierls, 1977).

That something basic was still being overlooked was constantly underlined by the unsolved problem of superconductivity. Bloch suggested that “the greatest failing still attached to . . . [the theory] is the absence of a point of view that allows one to grasp the interactions between the conduction electrons in a rational way.” These “delicate pulls,” he correctly surmised, “do not affect most properties of metals, but must play a prominent role in certain phenomena, notably, ferromagnetism and superconductivity” (Bloch, 1933, pp. 238–239). Brillouin noted (1931, p. v) that the theories all rested on simplifying assumptions, and that the interactions between electrons have been almost entirely neglected. “It is not out of the question that a more complete examination (for example, taking into account exchange phenomena) will bring fundamental changes in our conception.” These intuitions would be confirmed 25 years later, when new experimental techniques, new methods of preparing purer and more perfect samples, and a quantum field theory for treating the many-body problem would make possible another theoretical reshaping. (See, for example, Pines, 1981.)

After 1933, the central issue for the quantum theory of solids would be to explain the behavior of real solids, applying approximate mathematical models to the 1933 framework. The simple approximate study of the band structure of sodium by Wigner and Seitz (1933, 1934) marked an important step in this transition, and would within a few years give rise to an industry of band-structure computations for realistic solids.^{83,84}

The growing identity of solid-state physics was revealed from 1930 on in graduate courses and conferences in

⁸⁰Karl Scheel, editor of the *Handbuch*, had asked Sommerfeld to author the article. Sommerfeld agreed, provided Bethe did 90% of the work (and received 90% of the honorarium). Indeed, Sommerfeld wrote only the first 36 of its 290 pages, focusing on his own semiclassical treatment of 1927–1928, and Bethe the rest—the “modern wave-mechanical theory”—a job done in approximately 6 months of full-time work, spread out over about 1 year. Sommerfeld apparently never tried to understand the material Bethe covered, nor did he, as Bethe recalled, discuss the writing of these later sections much with him, although the two did go over the initial outline. The review was essentially unedited, and printed “pretty much in the way I had written it” (Bethe, 1981a, 1981b; Sommerfeld, 1933).

⁸¹This was first published in French (1930) and then substantially expanded in German (1931) and Russian (Brillouin, 1962).

⁸²Mott and Jones (1936, p. 320) contains a more complete listing of current texts and review articles.

⁸³Their method was to compute the electron eigenfunctions assuming each atom to be centered in a spherical “Wigner-Seitz” cell, and each electron to be acted on by a Hartree-Fock field. Bethe read the Wigner-Seitz work while in the last stages of preparing the *Handbuch* article and quickly added a section on it (Bethe, 1981a).

⁸⁴The history of the rise of band-structure calculations is treated by P. Hoch, with a contribution from K. Szymborski, “The Development of the Band Theory of Solids, 1933–1960,” to be published in Hoddeson *et al.* (1987a).

which topics began to be grouped in categories such as "solid bodies," "physics of solids," and "solid state." For example, at the ETH, Scherrer scheduled a 2 hour per week lecture course in the summer of 1930 on the "Physics of Solid Aggregate States." In Leipzig, Heisenberg presented the "Quantum Theory of Solid Bodies" in his special topics course during the winter semester of 1930–1931 (Leipzig, 1930; Heisenberg, 1931b), forewarning his students that "much is still unclear," but "that by the end of the lecture course . . . some things will clear up and perhaps it will stimulate one or more of you to carry out studies." Two years later, Hund offered a lecture course in Leipzig on the "Theory of the Solid State" (Leipzig, 1932), and that same year Wigner initiated a course at Princeton also titled "Theory of the Solid State," attended by approximately 15 graduate students, including Seitz, who prepared the course notes.⁸⁵ While in the late 1920s individual talks on solid state were increasingly included in conference programs,⁸⁶ by the early 1930s meetings devoted entirely to solid-state topics were not uncommon, for example, the third and fifth Leipziger Vorträge, in 1930 and 1933.

At just the time that the problems of the quantum theory of solids began to appear less fundamental, new discoveries were opening up the fields of nuclear physics, cosmic-ray physics, and quantum electrodynamics (Brown and Hoddeson, 1983). Many of those who had contributed to the theory of solids between 1926 and 1933—including Pauli, Heisenberg, Bloch, Landau, Peierls, and Bethe—took up the challenges of these new areas. In both Leipzig and Zurich, interest in solid-state physics declined sharply after the discovery of the neutron. Nordheim recently reflected: "In the early '30s . . . people gravitated to newer things, cosmic radiation and nuclear physics at that time, . . . also the discovery of the neutron, and then of artificial radioactivity, and . . . the development of the first accelerator . . ."⁸⁷ (Nordheim, 1962). However, not everyone abandoned the quantum theory of solids; Wilson, Slater, and Landau were among those who remained active in the area throughout this transition.

⁸⁵We would like to thank Professor Wigner for sending us a copy of these notes, which are in his personal collection at Princeton.

⁸⁶For example, the 1927 Solvay Conference on "Electrons and Photons" included a talk by W. L. Bragg on x-ray diffraction and reflection in crystals (Solvay, 1927).

⁸⁷Certain physicists saw the same situation in reverse. Shoenberg in Cambridge decided to enter solid state in this period, instead of nuclear physics, which, he recalled recently, "was getting too technical, a changeover from the sealing-wax-and-string to the big machine age, a lot of electronics." Shoenberg continues, "I didn't want to go into that . . . I also felt that in nuclear physics all the big things had been done, and then students were left with the detailed jobs. I got the impression that there was far more to be done in Kapitza's line than in nuclear physics." Shoenberg then took up the problem, suggested by Kapitza, of magnetostriction of bismuth (Shoenberg, 1981).

For physicists of Jewish background in Germany, this change of fields was reinforced by the sudden deterioration of their situation from April 1933 on, including their wholesale dismissal from positions. Bethe, who emigrated to England in 1933, and subsequently to the United States at the end of 1934, feels that had he not left Germany at this time, he would have done more in solid-state physics. "I imagine that probably after a few more years, I would also have been captivated with nuclear physics. But it came earlier because I got into contact with people who were doing nuclear physics. England was full of nuclear physicists when I came there in '33" (Bethe, 1936, 1981a). Peierls (1981a) says that his move in 1933 to Cambridge, a strong center of nuclear physics, encouraged him to enter that field. Similarly, Bloch, who moved to the U.S. in 1934, after stays in Copenhagen, Paris, and Rome, felt that these political events had a direct influence on his change of interests then (Bloch, 1981).

The growth of solid-state facilities between 1930 and 1933 geared towards research in real physical systems would keep the field active and attract new workers. The three prime academic examples of such facilities are the Bristol school, which grew up around John E. Lennard-Jones, Mott, and Harry Jones, the physics department at MIT under Slater's direction, and the group within the physics department at Princeton surrounding Wigner. Bristol drew heavily on strong traditions in crystallography, low-temperature physics, and metallography, and attracted young solid-state theorists partly through a growing recognition of the potential industrial relevance of solid-state theory (Keith and Hoch, 1986). At MIT, Slater, who assumed the physics department chairmanship in 1930, built the department up with a strong solid-state theory emphasis (Slater, no date).⁸⁸ Wigner came to solid state via chemistry and crystal structure with the strong feeling that the Sommerfeld-Bloch-Peierls theory did not begin to answer fundamental questions of the structure of real crystals, such as "why the crystal exists; what is its binding energy" (Wigner, 1981). Furthermore, major industrial laboratories in America, including Bell Telephone Laboratories and General Electric, recognizing that active research was essential to application of the new theoretical advances, decided to appoint many well-trained solid-state physicists (Hoddeson, 1980, 1981; Wise, 1985).⁸⁹

The transformation of the field, together with the shift of the center of solid-state activity from Germany to England and the United States, caused by the emigration from Germany of refugee physicists and the growth of new institutions, brought to a close the heroic era, the "Heldentage," in the development of the quantum theory of solids.

⁸⁸Direct ties between Slater's department and Sommerfeld's institute were built by earlier visits to Munich, e.g., by Frank, Morse, and William Allis, in Slater's new department (Slater, 1975, p. 164).

⁸⁹The onset of the Great Depression prevented these laboratories from fulfilling their hiring plans for several years.

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⁹⁰The following abbreviations are used in the references. DM represents documents in the archives of the Deutsches Museum in Munich. AHQP represents the Archive for the History of Quantum Physics, a collection of microfilms of documents, interview transcripts, and interview tapes, available at a number of institutions including the archives of the Niels Bohr Institute in Copenhagen, the Niels Bohr Library of the American Institute of Physics Center for History of Physics in New York City, and the American Philosophical Society Library in Philadelphia. BSM represents the collection of Bohr Scientific Manuscripts in the AHQP. WP1 represents the first volume of the collected Pauli correspondence, edited by Hermann *et al.* (1979). WP2 represents the second volume of the collected Pauli correspondence, edited by von Meyenn *et al.* (1985).

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