

Recollections

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## Recollections

BY C. HERRING

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*Herring, (William) Conyers. Born New York 1914. Educated Princeton University. Research physicist Bell Telephone Laboratories. Research on many branches of physics, particularly magnetism.*

### 1. ONE-ELECTRON ENERGY BANDS

As a graduate student in the 1930s I got my principal education on the electronic quantum mechanics of solids from the Sommerfeld–Bethe article in the *Handbuch der Physik*. It seemed quite clear from this that the one-electron wavefunctions and energies of a self-consistent field solution for a crystal would often have characteristics intermediate between those of the tight-binding approximation and those of the almost-free-electron theory, and that therefore one would need to find new and less restrictive ways of calculating them. The Wigner–Seitz method, which had just been developed, was one such, although its initial validity was limited to states neighbouring the  $k = 0$  state of the conduction band of a non-transition metal. Beyond this, one very appealing handle was to study the rigorous properties of Bloch states that could be inferred from crystal symmetry. Louis Bouckaert and Roman Smoluchowski, who were working as postdoctorals with Wigner in the academic year 1935–6, undertook to study these symmetry properties for electrons in the common cubic lattices; their work culminated in the now famous B.S.W. paper of 1936. I held many discussions with them, and became intrigued by the study of the topological behaviour of the functions  $E_n(k)$  describing the energies of the bands  $n$ , and particularly by the topology of the evolution of these functions as the potential energy function, i.e. the Hamiltonian of the problem, was changed continuously. The development of this interest came while Wigner, under whom I was working as a graduate student at Princeton, was away for the second term at Wisconsin. In our long-distance communications, he encouraged my interest and made the further very fruitful suggestion that it would be of interest to supplement the study of spatial symmetry properties with a study of time-reversal symmetry, a subject to whose general formulation (as distinguished from its application to crystals) he had already made fundamental contributions. Before the end of the academic year I had picked up enough ideas to keep me busy, working at home on Staten Island over the summer. The result was my thesis ‘On energy coincidences in the theory of Brillouin zones’. This had two parts. The first was an analysis of consequences of time-reversal symmetry, and the development of criteria for predicting when it would require the ‘sticking together’ of bands that would be independent if only spatial symmetry were considered. The second and longer part

of the thesis dealt with energy coincidences between different bands, i.e.

$$E_n(k) = E_{n'}(k)$$

for  $n' \neq n$ , that were not required by symmetry, and noted the interesting fact that some such coincidences are indestructable, in the sense that they cannot be made to disappear (though they can be moved about in  $k$ -space) by any infinitesimal change in the lattice potential. I found this work intellectually fascinating, but had grave misgivings about its applicability to real electronic energy bands, because I felt that correlation effects (see below) must make the picturing of metal electrons in one-electron terms a very poor approximation. Fortunately, I realized that the results would have a much more clearly valid applicability to the frequency spectrum of phonons, so I did not feel that I was being awarded a degree under entirely false pretenses.

In the following academic year (1937), John Slater visited Princeton (Institute for Advanced Study) for a sabbatical semester. His methods of reasoning and working were very different from Wigner's, or from my own, and did not at first appeal to me, but I was very impressed by his productivity: in that one semester, by simply barging ahead with calculations in areas that seemed to him to contain 'pay dirt', he succeeded in completing four papers, two of which have emerged in historical perspective as classics. (These were the invention of the APW method and the formulation of the theory of ferromagnetic exchange coupling for an insulator in band-theoretical terms, respectively.) In the hope that some of this facility might rub off on me, I applied to the National Research Council for a fellowship to do postdoctoral research with him at M.I.T. The fellowship was awarded, so I spent the years 1937-9 at M.I.T. However, my strongest intellectual contacts were not with Slater, but with John Bardeen, who had gone from Princeton to Harvard a year earlier to be a Junior Fellow there, and with some of the younger people, mostly experimental, at M.I.T. I was particularly impressed, in view of my developing interest in band calculations (see below), by Bardeen's improvements to the Wigner-Seitz approach, especially in the direction of accurate calculation of the effective mass at the bottom of a band.

My original intention in these postdoctoral years was to develop more realistic models of lattice vibrations, particularly in metals. Since I was convinced that interatomic force constants could not be adequately described in terms of mere nearest neighbour forces or in terms of simple pair potentials, I decided to try to determine the force constants from first principles by suitably generalizing the Wigner-Seitz method of calculating lattice energies. Though such calculations were eventually made in the late 1950s by Toya, and have since become quite commonplace thanks to the concept of pseudopotentials, I found the task too difficult for me at the time, and decided that one would need first to develop a new and more tractable formalism for the calculation of band structures in general. For the simple metals it seemed that one ought to be able to capitalize on the great similarity of their wavefunctions to plane waves; yet it was already known that the strong

departures from plane-wave form near the atomic nuclei had very important consequences for the energies of the states. Realizing that these departures were necessitated by the requirement that the valence electron wave functions be orthogonal to those of the cores, I thought of working with a non-orthogonal basis formed by orthogonalizing plane waves to the core states. To my surprise, even a single plane wave of this sort often gave a remarkably good quantitative approximation to the correct crystal wavefunction. So I started developing the approach systematically, and published an exposition of what is now known as the orthogonalized plane wave method. A fortunate opportunity presented itself immediately to test this in a full-scale band calculation: A. G. Hill, with whom I was at that time sharing an apartment, had started some calculations with Seitz at Rochester on the band structure of beryllium, but had not finished them. Although primarily an experimentalist, he was very interested in doing more work on this project, and he asked if I would be interested in collaborating with him. Since beryllium is divalent, and crystallizes in the hcp structure with two atoms per primitive cell, its bands approximately fill two Brillouin zones instead of half a zone as in the alkali metals. Thus calculation of its band structure and cohesive energy requires a method that is capable of handling states near the zone boundaries: an approximation using merely a parabolic band of constant effective mass would clearly be inadequate, and Shockley's 'empty-lattice' test had just shown that sizable errors could occur in Slater's earlier scheme of fitting boundary conditions only at the centres of cell faces. So I proposed that we should combine OPW calculations for states at the zone boundary with Wigner-Seitz-Bardeen calculations for states near the bottom of the band. The project proved enormously laborious, but eventually we managed to get it finished, and it was on the whole quite successful.

In these same years at M.I.T., Marvin Chodorow, a graduate student under Slater, was working on the application of Slater's new APW method to copper. As his office adjoined mine, we had many conversations on this work. A noteworthy feature of it was his construction of an empirical ion-core potential for copper, significantly better than any that had been used before. Though the thesis was never published in full, use of the Chodorow potential has been revived in recent years.

While this was going on, and afterward, I started to work on several applications of the OPW method. One was an improved calculation of compressibility, etc., for lithium; another, moving towards my original goal of calculating interatomic force constants, was an attempt, with H. B. Huntington, to evaluate the corrections to Fuchs's simple electrostatic theory of shear constants of simple metals. But such work went slowly, and after spending the next two years at Princeton and Missouri, respectively, I became involved, as did everyone else, in war work for the period 1941-5. After the war, I became involved with other things, and it was with surprise that I learned several years later, in an encounter with Frank Herman at an A.P.S. meeting in New York, that he was trying to do a thesis at Columbia University on the electronic bands of diamond, using the OPW method. I followed his work

with interest, as I began to think about possible band structures for the semiconductors silicon and germanium which were being intensively investigated at Bell Laboratories, where I now was. After finishing this diamond work and moving to R.C.A., Herman collaborated with a graduate student at Princeton, J. Callaway, in applying similar methods to the band structure of germanium.

As just noted, there was, in the late 1940s and early 1950s, a great deal of interest in energy bands, especially in valence and conduction band edges, in germanium and silicon. In the early part of this period, many theoretical discussions simply assumed 'for simplicity' that the band edges would be at the centre of the Brillouin zone, and would be non-degenerate, with isotropic effective masses. My experience with calculations for beryllium, and in due course my awareness of the results Herman was getting for diamond, convinced me that such a picture for the electron and hole states in silicon and germanium was extremely unlikely. However, there remained many possibilities, and it took some years to sort them out. Transport properties, particularly the anisotropy of magnetoresistance in single crystals, seemed to provide some of the most promising handles. Early successful uses of this approach were my collaboration with G. L. Pearson to infer from his experiments that the conduction band edge in silicon was on the  $\Delta$  lines with a mass anisotropy of five to one, and the inference by Meiboom & Abeles that the conduction band edge in germanium was at the L points, with a much larger mass anisotropy. Both these conclusions were soon confirmed by cyclotron resonance experiments. The magnetoresistance approach was less successful for the valence bands. I became convinced that the valence band edges in germanium must be along the  $\Delta$  lines, because of the vanishing magnetoresistance for magnetic fields in the [100] directions. Such vanishing would have to occur from symmetry for [100] valleys, but could occur only by accident for many other band structures, including the *a priori* plausible one of a degenerate band edge at  $k = 0$ . It turned out that just this 'accident' does in fact occur. By the middle 1950s, elucidation of the band structures of semiconductors was proceeding fairly rapidly, thanks primarily to cyclotron resonance, but also to such things as piezoresistance, optical properties, etc.; these provided a wealth of opportunities for comparing theory and experiment.

By contrast, there had not occurred any great expansion in opportunities for comparing theoretical band structures with experimental data for the case of metals. There were still the old standbys: electronic specific heat, paramagnetic susceptibility, some features of optical absorption, etc., but the great modern science of Fermiology had not yet been born. One new thing, however, did come along at about this time: the Knight shift. C. H. Townes, who had been a colleague of mine at Bell Laboratories in the late 1940s, had moved to Columbia University, and got involved in the newly blossoming field of nuclear magnetic resonance. Working at Brookhaven, a student of his, W. D. Knight, discovered that the n.m.r. frequencies of nuclei in metals are usually appreciably greater than the frequencies of the corresponding nuclei in non-metallic compounds. Townes proposed the interpretation that the field on the nucleus was enhanced by the contact hyperfine term

associated with the polarization of the conduction electrons, i.e. with their Pauli paramagnetism. The magnitude of the effect should be proportional to the product of the spin susceptibility and the average  $s$  character of the Fermi-surface electrons. Since crude ways of estimating the latter could be significantly in error, he asked me to collaborate with them in working out some reasonably good estimates for metals whose band structures had been calculated. Some of the comparisons of theory and experiment were gratifying, others could more aptly be described as intriguing; such comparisons have subsequently become the object of a sizable literature.

Despite the advances associated with the APW and OPW methods of calculating band energies and wavefunctions, such calculations were still quite laborious, and there was a great need for something simpler that could be used to make calculations of such things as phonon frequencies, etc. In the 1930s, the Russian physical chemist, Hellmann, had noted (as I too later noted in developing the OPW method) that the rapid oscillations of a valence-electron wavefunction near the nucleus of an atom are necessitated by the requirement of orthogonality to the cores. He proposed replacing the orthogonality requirement by a positive effective potential in the core region and gave a rough derivation of such a positive potential on the basis of the Fermi-Thomas model of the atom. He then proposed that one could choose this repulsive potential empirically, rather than relying on first-principles calculation of it: namely, one could choose a few parameters describing the repulsive potential in such a way that a nodeless eigenfunction in the empirical potential would have the same energy as a true valence-electron eigenfunction in the atom. He and Kassatochkin applied this concept, in a very crude but surprisingly effective way, to the calculation of cohesive energies and other properties of a large number of non-transition metals. I had occasion to study this work in the course of preparing a review article on thermionic emission in collaboration with M. H. Nichols, and I was very impressed by the possibilities of the method. After some years I called it to the attention of J. C. Phillips, who had come to Bell Laboratories in the late 1950s as a postdoctoral worker. When Phillips concluded his stint at Bell Laboratories, he moved to Berkeley, where in collaboration with a student, Leonard Kleinman, he succeeded in putting the concept of what are now called 'pseudopotentials' on a more logical basis. As is well known, use of pseudopotentials spread like wildfire in the 1960s and their logical basis was still further explored.

## 2. CORRELATION ENERGY

Although people have known from the early days of quantum mechanics that the true ground-state wavefunction of a many-electron system must be of more complicated form than a determinant of one-electron wavefunctions, and although the importance of the 'correlation-energy' correction to a band-theory calculation of the cohesive energy of a metal had been clearly recognized by Wigner and Seitz in 1933 and 1934, the magnitude and role of correlation energy remained inade-

quately understood in a considerable part of the solid-state community for many years. Perhaps the complication of Wigner's important paper of 1934, which contained a surprisingly good estimate of the correlation energy of a free-electron gas, prompted wishful thinking in the direction of judging such effects to be smaller than he had estimated. At least one serious error of logic appeared in the literature: a paper in 1933 by Brillouin<sup>(1)</sup> in which he undertook to sum the Brillouin–Wigner perturbation series to all orders in the electron–electron interaction in a free-electron gas, and obtained the preposterous result that, because of the behaviour of matrix elements in a system of infinite volume, all terms would vanish, so that there would be no correlation energy! I suspect that even Slater, although he had taken brilliant account of correlation effects in such things as his paper on ferromagnetism and spin waves (mentioned above), did not adequately appreciate the importance of correlation energy in general. As late as 1951, Löwdin,<sup>(2)</sup> who had been interacting with Slater's group at M.I.T., published a calculation of the binding energy of sodium that claimed to get agreement with experiment without including any correlation term. Even those who fully appreciated the importance of correlation energy did not in the early years have a very clear conception of all its implications. Thus, for example, Bardeen, who had found correlation effects to be decisively important in the theory of the work function, and who had always included an important correlation term in his calculations of cohesive energy, at one time gave serious consideration to the prediction of Hartree–Fock theory that because of the exchange effect, the density of states in energy should go to zero at the Fermi energy of a free-electron gas at zero temperature. Most of the rest of us were also briefly confused on this point, although Wigner had indicated that correlation effects would destroy this singular behaviour.

These last sentences touch on the topic of quasi-particles. As noted above, I had had grave doubts when I wrote my thesis that fine details of the dependence of energy on wavevector in one-electron energy bands could have any real physical significance, because correlation effects must surely be large. In the late 1940s, as many points of contact developed between experiments on semiconductors and the theory of their energy band structures, it became clear to many of us that there must exist exact quantum states of the many-electron system with just the same quantum numbers and topology of energy spectrum as the one-electron states of band theory, provided the energy of excitation above the ground state is less than the band gap, i.e., the energy required to produce an additional electron–hole pair at large separation. I do not know who first pointed this out in the literature, or even if it was explicitly pointed out in these early days, but the fact was certainly known to cognoscenti. Later, of course, Kohn and others developed the concept very precisely. Some of us naturally reflected, too, on the possibility that a similar renormalization of single-particle excitation states might occur in metals. While it was clear that for metals one could not define infinite-life quasi-particle states of finite excitation energy, because there was no energy gap, the lifetime width of an excitation should become small compared with the energy of the excitation as

this energy approaches zero. In other words, quasi-particle states can be defined that have an asymptotically precise meaning as one approaches the Fermi surface. But the idea was so simple and so fuzzy that I am not sure whether anything was published on it before Landau's theory of Fermi liquids in 1956. Landau was typically willing to trust his intuition far beyond where logic would take him and, in this case, it really paid off. I do recall, however, publishing some rather fuzzy ideas with the same general philosophy, on the subject of itinerant-electron magnons, in a 1951 paper with Kittel.

### 3. SURFACE PHYSICS

I have already mentioned Bardeen's thesis, in which he showed that an important term in the work function comes from the dipole layer formed at the surface by the departure of the charge distribution in and near the surface cells from the symmetrical form that it has in the interior cells, and in which he showed the importance of exchange and correlation effects for the calculation of this dipole moment. Since the dipole moment must depend on conditions at the surface, and in particular, must be different for different crystallographic faces of the same metal, it became clear that previous theories (of which there were many) that had tried to express the work function of a metal as a property of its bulk lattice could never be very accurate. In his Harvard period, Bardeen made a start at estimating the differences between the dipole moments of different crystal faces. A much more systematic attack on this question was carried out by Smoluchowski just before World War II. I was very interested in this work, as my contacts with experimentalists of Nottingham's group at M.I.T. had got me interested in thermionic emission, and this interest continued and strengthened in my 1940-1 year at Princeton, where M. H. Nichols of Wayne B. Nottingham's group was now doing postdoctoral work. Smoluchowski's work was done, I believe, just as I was moving from my subsequent year at Missouri to New York to do war work. I now remember corresponding with him about it. But several years of hectic involvement with antisubmarine warfare had put all this so completely out of my head that when the after the war I again started thinking about thermionic emission problems at Bell Laboratories, I read Smoluchowski's paper through to the end with great interest, before discovering, in the acknowledgments, that I had read it before.

My interest in thermionic emission got me involved, in the immediate pre-war years, in the theory of thermoelectric effects and the thermodynamics of irreversible phenomena. I used some rather fuzzy theories from the Dutch school to derive some correct results about contact potentials between conductors at different temperatures and about the cooling effect accompanying thermionic emission. Some years later it became clear that such results could be derived much more clearly and rigorously on the basis of linear-response theory and the Onsager relations. But though Onsager's work had been done some years earlier, its application to many practical problems, such as these, lagged, and even the logic of its derivation



was not nearly as clear as it became after Kubo's work of the mid-1950s. Actually, although I did not know it at the time, the results I obtained at this time had already been obtained (though by equally unsatisfactory reasoning) by Carl Wagner some years earlier.

Because of the importance of surface details, and in particular, of surface patchiness, for thermionic phenomena, I became interested in the late 1940s in the phenomenon of thermal etching of surfaces. While thermal etching can occur in the course of evaporation or crystal growth, it may also sometimes occur as a normal way of approaching thermodynamic equilibrium; in such a case it must be that the surface can lower its surface free energy by developing a hill-and-valley structure. So I became interested in the surface tension of solids, and its variation with crystallographic orientation. At one of Nottingham's annual physical electronics conferences at M.I.T., in the late 1940s, I ran into a young metallurgist named George Kuczynski, then (or perhaps soon after) at Sylvania, who was doing fundamental experiments on the sintering of metal particles. I realized that the interpretation of sintering phenomena would have to involve many of the same concepts as were involved with thermal etching, and so was born my interest in sintering. I maintained contact with Kuczynski and his colleagues for several years, and in the course of a number of experimental and theoretical papers it became clear that precise equations could be formulated to describe many sintering-type phenomena, and that from approximate solutions of these equations one could identify which of several mechanisms of transport were operative in any given case. In this period, wire-pulling experiments, as developed by Shaler, Wulff, and others on the basis of similar, cruder, experiments of the 1930s, provided improved quantitative measurements of surface tensions of solids, and verified the phenomenon of diffusional creep.

#### 4. STATISTICAL MECHANICS

While I was a graduate student at Princeton – I think it was probably in the Spring of 1937 – R. H. Fowler spent a term there and gave some lectures on new and exciting developments in the area of statistical mechanics of solids. I remember particularly his discussion of the theory of order–disorder transformations in alloys, with the Bethe cluster approach being presented as a great improvement over the theory of molecular field type of Bragg and Williams. The libration–rotation transformations were likewise treated.

In the summer of 1940 I took part in the annual summer symposium on theoretical physics at the University of Michigan. I shared a room with Gregory Wannier, whom I had first encountered at Princeton several years earlier. He was hard at work trying to finish up a paper on the statistical mechanics of the two-dimensional ferromagnet, work which he had begun in Holland with Kramers, but which he was having to complete without any contact with his collaborator, because the German occupation had cut all connections. He was very excited over the discovery of a

similarity transformation that made an exact connection between the behaviour at high temperatures and that at low. Thanks to this relation, Kramers & Wannier were able to deduce a rigorous expression for the Curie temperature in terms of the exchange integral, for whose validity it was necessary only to assume that there was a single singularity at the Curie temperature. Several years later, Wannier visited me at my home in New York, where I was engaged in war work. Onsager's exact solution for the two-dimensional ferromagnet had just been published, and Wannier was very depressed. He was carrying a copy of Van der Waerden's *Moderne Algebra* and announced that he would now have to learn a whole new field of mathematics before he could understand Onsager's paper and be back in the swim of things. By the time he joined the staff at Bell Laboratories in the late 1940s, he had indeed mastered the new approach, and he and Phil Anderson kept the chalk flying in the tea room with ideas for further extensions.

## 5. MISCELLANEOUS AREAS

When Wannier arrived in Princeton – I think it was in the autumn of 1936 – as a new postdoctoral from Switzerland, he was just in the course of changing his field from quantum field theory to solid state. His first major research project was a formulation of exciton theory for general energy bands. His desire to make the treatment analogous to the local picture of excitons in terms of localized atomic states led him to the concept of what are now called Wannier functions. The concept received practically simultaneous application in Slater's paper of the same year on magnetic exchange and ferromagnons, referred to above.

Despite early work by Bloch, Stoner and others, and despite the subsequent development of the itinerant model of magnetism into a whole school of thought by Stoner, most magneticians for many years considered such models to be much less useful and less realistic than models based on localized atomic moments. In this period some of the most important efforts to describe ferromagnetic phenomena in terms of realistic energy bands, rather than the idealized models used by Stoner and his school, were made by Slater. Just before his 1937 paper just mentioned, he had written two papers on the ferromagnetism of nickel, which attempted a semiquantitative description in terms of an almost filled d band. However, these papers made very inadequate allowance for correlation effects. A little later, some of Van Vleck's students took up itinerant-electron ferromagnetism: Brooks showed how the theory of ferromagnetic anisotropy could be formulated for a band model; Hurwitz showed that even a very large correlation energy could be taken into account in what he called a 'minimum-polarity model'. After the distractions of World War II had cleared away, Van Vleck returned to the question of correlation energy, and showed that if the quantum mechanical fluctuations in the ground state of metallic nickel allowed two d holes to be on the same nickel atom even a small proportion of the time, the Hund's-rule coupling of these holes could suffice to account for the ferromagnetism of nickel. I had some long conversations with

Van Vleck while he was preparing this work for publication in 1953 and suggested a consideration, which I subsequently elaborated in my book: the energy cost of such 'polar' fluctuations might be significantly reduced by screening by the more mobile electrons of the s-p band. But it was only with the development of Fermiology in the 1960s that itinerant magnetism became fully respectable. For example, I remember that when I submitted two papers to the *Physical Review* in 1951 on the subject of the description of Bloch wall states and long wavelength magnons in itinerant models, the referee, although finding nothing wrong with the papers, objected that it was wasteful to devote so much space to such an unrealistic subject as the itinerant model of magnetism.

The benefits of the close interaction between experimentalists and theorists at Bell Laboratories were nowhere better illustrated than in some of the developments having to do with dislocations and plasticity. In the late 1940s, Shockley was very interested in dislocations, and he succeeded in getting an imaginative young applied mathematician, Thornton Read, to join him in thinking about them. They realized early that the rows of etch pits that revealed subgrain boundaries and lineage structure in the routine testing of semiconductor crystals for perfection could be used to test an idea that I believe was originally due to Burgers, namely that small-angle grain boundaries should consist of reasonably well spaced dislocations. They got experimentalists to work at simultaneously measuring grain boundary energies (by the angles of grain boundary grooves), and the spacings of the corresponding dislocation arrays in etched samples cut normally to the grain boundary. They deduced and confirmed a  $\theta \log \theta$  dependence of the grain boundary energy on the angle  $\theta$  between the grain boundary and a simple rational orientation. In another example, some of us working in basic physics happened to hear of the accidental discovery of thin tin whiskers growing out of the plating inside some sealed channel filters that had been used in telephone applications, and that mysteriously developed short circuits, traced ultimately to the whiskers. It occurred to us that because of their small diameter (one or two micrometres) such whiskers might well be perfect crystals free of dislocations, in which case they should be capable of withstanding very large strains without suffering plastic deformation. John Galt volunteered to do some experiments on the whiskers, and these confirmed the expectations. The subject of plasticity in whiskers eventually developed quite a sizeable literature. A third and especially important development in dislocation physics came in 1950, when, in a remarkable time coincidence, Thornton Read and F. C. Frank simultaneously and independently conceived the concept of dislocation multiplication by regeneration of loops around a pair of pinning points (the Frank-Read source).

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