

Electrical conduction in metals

The modern theory was created fifty years ago by Felix Bloch; it still provides the basis for understanding conduction, but many challenges remain.

Philip B. Allen and William H. Butler

Fifty years ago in the *Zeitschrift für Physik*, two papers were published which laid the foundations for the modern quantum theory of solids. Arnold Sommerfeld¹ quantized the theory of the electron gas, and Felix Bloch² showed how this could be generalized to a realistic theory for electrons in solids. In a delightful article in *PHYSICS TODAY* (December 1976 page 23) Bloch has recounted the circumstances in which he came to work on the electron theory of metals in Leipzig as Heisenberg's first graduate student.

Bloch's thesis is one of several papers that appeared after 1926 and opened up a whole new field by applying the quantum theory to an old problem. In Bloch's words "almost any problem that had been tossed around years before could now be reopened and made amenable to a consistent treatment." As we read Bloch's paper today, it is apparent that he is very modest. In this article we will attempt to describe how even fifty years later, the process of understanding and implementing Bloch's ideas on electron transport is far from over.

Early theories

Modern theory of electrical conduction in metals began with Paul Drude's paper³ of 1900 which treats the process in terms of a recently discovered particle, the electron. The theory rested on an assumption that under scrutiny seems at first absurdly oversimplified, namely that one should treat conduction electrons as a gas of weakly interacting particles. However, it is precisely this curious fea-

ture of Drude's model that remains at the heart of the modern theory as well; the great successes of both theories suggest that it does have an element of truth. The shininess and high conductivity of metals follow clearly from the hypothesis, unifying their ac and dc properties. The theory could have predicted the transparency of metals in the ultraviolet, a phenomenon that was not discovered experimentally until 1933.

By applying methods from the kinetic theory of gases, Drude was able to give a quantitative derivation of the Wiedemann-Franz law, that the ratio of thermal to electrical conductivities is a universal number, the Lorenz number, times the absolute temperature. If both electrical and thermal currents are carried by electrons, the same mean free path and mean speed enters both formulas. Specifically, the thermal conductivity is

$$\kappa = \frac{1}{2} k_B n l v$$

and the electrical conductivity,

$$\sigma = \frac{e^2}{4k_B T} \cdot n l v$$

(n is the density of particles, v their speed, and l their mean free path). The Lorenz number is thus $3(k_B/e)^2$ which is in reasonable agreement with experiment. The chief source of error was, in fact, in the measured values k_B and e . Accurate values of these constants were not available until Max Planck's paper of October 1900, in which he deduced k_B and e from the data on blackbody radiation.

But all was not well. The theory was "refined" by H. A. Lorentz⁴ who introduced Boltzmann's equation from rarefied gas theory. Among other minor corrections, he found the theoretical Lorenz number to be reduced by a factor 2/3 from Drude's value, weakening the

agreement with experiment. The full degree of disagreement between classical theory and experiment was most clearly described by Niels Bohr in his doctoral dissertation, which has been called⁵ "perhaps the most deflationary publication of all time in physics." The difficulties of Drude's theory are well known:

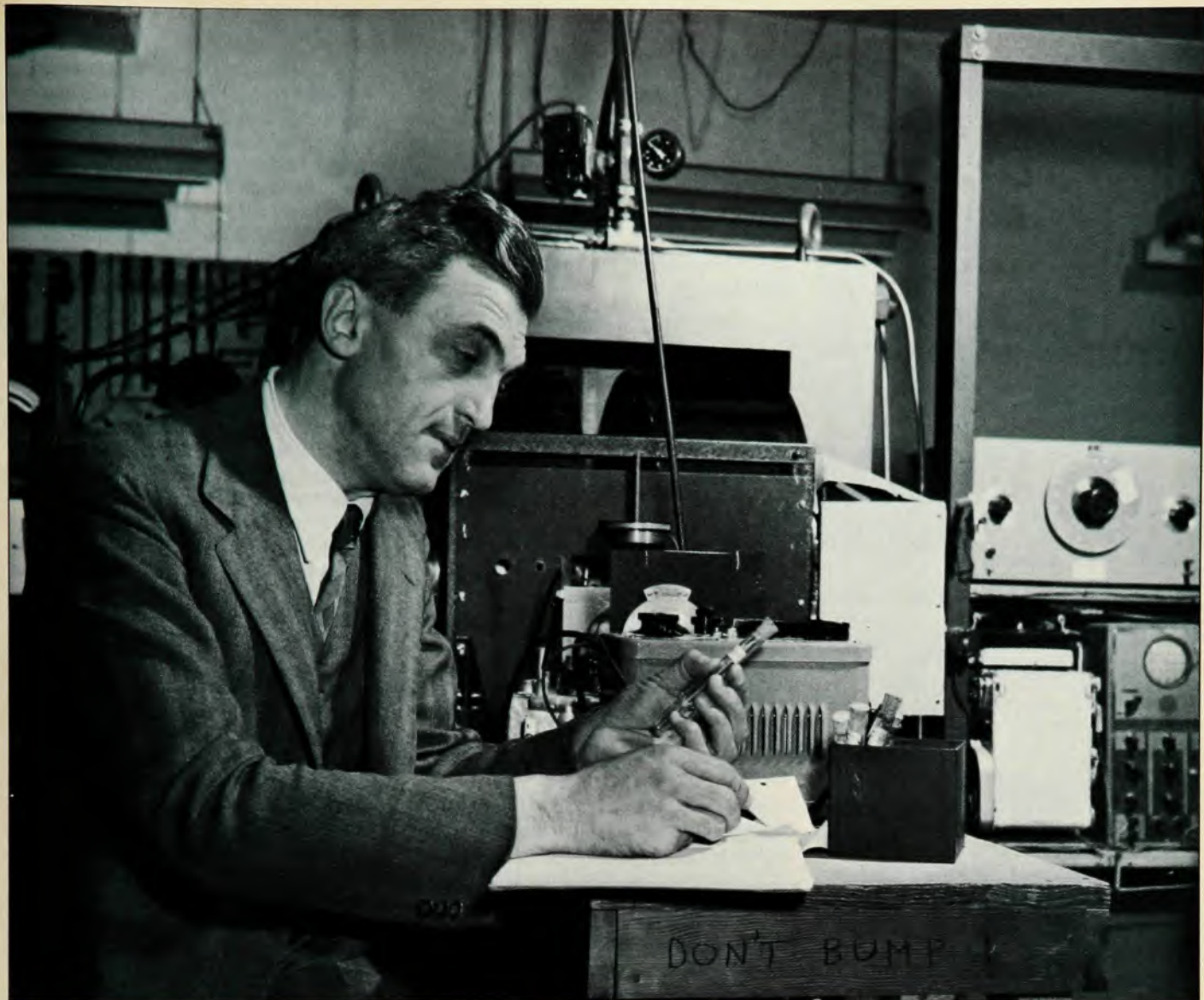
- ▶ prediction of an electronic heat capacity $C = 3Nk_B/2$, which is not observed,
- ▶ failure to explain magnetic properties, and
- ▶ failure to explain the temperature dependence of the conductivity.

Quantum theories

The first problem was cured by Sommerfeld¹ who found that because of the Pauli principle, only a small fraction of the electrons (specifically, $k_B T/E_F$ where the Fermi energy, E_F , is about 10 eV) can be thermally excited and contribute to the heat capacity. Wolfgang Pauli and Werner Heisenberg showed that quantum theory is the key⁵ to the second problem, although the puzzles are by no means completely solved even today. Many of the concepts necessary for the solution of the third problem were given in Sommerfeld's 1928 paper. Treating the electrons as a quantum instead of a classical gas brings in a new velocity, the Fermi velocity, which is temperature-independent. The observed T -dependence of resistivity can then be assigned to the electron scattering rate.

The value of the Lorenz number from Sommerfeld's theory is larger than Drude's value by a factor of $(\pi/3)^2$, and is in excellent agreement with experiment. However, Sommerfeld's theory provided no more justification than Drude's for treating electrons as a gas. It remained for Bloch to provide a satisfactory fundamental theory.

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FELIX BLOCH 1952

Bloch's 1928 paper is best known for "Bloch's theorem" which states that in a periodic medium such as a crystal, the electron eigenstates can be chosen as plane waves modulated by a function with the periodicity of the medium. However, this theorem is but a small part of the work. It had, in fact, been found a few months earlier by E. E. Wittmer and Leon Rosenfeld,⁶ as Bloch acknowledges in his thesis, and can also be thought of as a direct result of Floquet's theorem (published in 1883)—Bloch, however, used the recently introduced group-theoretic methods to deduce the theorem.

From the theorem, Bloch proved that the eigenstates he had found carry a non-vanishing current, and thus have a complete analogy to the corresponding plane-wave eigenstates of the quantum free electron gas. This eliminated one important objection to Drude's "absurd" rarefied gas hypothesis by showing how the electrons could travel through the lattice without scattering off the ions, but it did not explain how they could avoid interacting strongly with each other.

The greatest accomplishment of

Bloch's paper seems to us to be his analysis of how metallic electrons respond to an externally imposed electric field, and how they scatter from the lattice defects and vibrations. The difficulty of this problem has been summarized by William Visscher.⁷ "The obstacle with which any attempt to calculate σ is soon confronted is that in the absence of random scatterers it is infinite. If perturbation theory is to be used, it must be used with care, because every term in the expansion in powers of the scattering strength is either zero or infinity." Bloch's solution to this problem required an intuitive leap, which was verified by perturbation theorists thirty years later. First he noticed that a wave-packet built of "Bloch states" centered at wave-vector $\mathbf{k}$ drifted in $\mathbf{k}$ -space under the influence of the electric field. The drift rate is now called the "semiclassical acceleration;" its value is $d\mathbf{k}/dt = -e\mathbf{E}/\hbar$, and it is a quantum analogue of the classical force law used in the earlier classical gas theories of conduction. He then argued that the wave-packet drift implied that in a metal the occupation probability $F(\mathbf{k})$ of the Bloch states (the

quantum analogue of the classical distribution function) would evolve in time according to $dF/dt = -e\mathbf{E}/\hbar \cdot dF/d\mathbf{k}$. Next he discussed the interaction of electrons with lattice vibrations and derived the form of the relevant matrix elements. Time-dependent perturbation theory showed that the interaction gives rise to energy and wave-vector conserving inelastic transitions between different electron states by emission or absorption of lattice quanta, or phonons. Finally, mentioning great computational difficulties ("*grosse rechnerische Schwierigkeiten*") he abandoned rigor and presented a reasonable model for the matrix elements and made a statistical discussion of the change in occupation probability due to electron scattering events. This discussion is closely parallel to the classical method of Boltzmann and Lorentz. However, Bloch inserted new factors of the type $1 - F(\mathbf{k}')$ to ensure that no scattering could occur into the state $\mathbf{k}'$ (according to the Pauli principle) if the state were already occupied. The legitimacy of using the statistical probability $F(\mathbf{k}')$ at this point is by no means obvious from



ARNOLD SOMMERFELD c. 1930



PAUL DRUDE 1906



EDUARD GRUNEISEN 1937

Schrödinger's equation. However, Bloch was able to derive a remarkable consequence which must have increased his confidence considerably. He proved that if the lattice vibration obeyed Einstein's statistics in thermal equilibrium, then the only electron occupation function which would remain stationary under the mutual scattering between two states $\mathbf{k}$ and $\mathbf{k}'$ of energies $\epsilon_{\mathbf{k}}, \epsilon_{\mathbf{k}'}$ was the newly discovered Fermi-Dirac equilibrium distribution function

$$F_0(\mathbf{k}) = [\exp(\epsilon_{\mathbf{k}} - \mu)/k_B T - 1]^{-1}$$

Bloch then had an equation showing how a steady state current obeying Ohm's law would be reached owing to competition between the electric field pulling the occupancy function in $\mathbf{k}$ -space and the collisions with phonons driving it back towards the Fermi-Dirac equilibrium value. This Bloch-Boltzmann equation (or "semiclassical" Boltzmann equation) remains the central result in the theory of transport in metals and semiconductors. Solving this equation did not prove trivial. Deriving the result that ρ increases linearly with T at high temperature was relatively easy, but the correct low temperature solution, $\rho \propto T^5$, eluded Bloch until⁸ 1930 (see the box below).

Modern results

Where do we stand today in our ability to understand and apply Bloch's theory? Let us postpone a discussion of the deeper microscopic justification of Bloch's theory, and examine first how well the theory works for phenomena in real and well-behaved metals. Figure 1 shows the measured resistivity of nearly pure palladium, an element that has attracted much interest for many years. The Bloch theory as extended by Eduard Grüneisen⁹ in 1933 fits nicely the overall shape of the data. However, a more detailed examination of the low temperature region by Grüneisen showed larger discrepancies

than had been seen in most other elements up to that time. The source of these discrepancies, whether they arose from details of the structure of palladium or from fundamental defects of the theory, was not clear.

Much more is now known about Pd than was known fifty years ago. The shape of the Fermi surface has been measured by such experiments as de Haas-van Alphen oscillations. Neutron scattering has determined the phonon frequency spectrum. Computational band-theorists have learned how to compute electron energy levels and find good agreement with Fermi-surface measurements. Thus the ingredients are available for turning Bloch's equations into a quantitative and predictive theory. However, this program is complicated by two factors. One is purely technical: no simplifying features such as spherical Fermi surface or Debye-like phonon spectrum are consistent with our current knowledge. The algebra of solving Bloch's Boltzmann theory is thus a tedious, but soluble, problem. The second difficulty is that a realistic self-consistent treatment of electron-phonon matrix elements is still not available. Currently theorists are mostly avoiding this problem by making judicious guesses whose consequences can then be compared with experiment.

In collaboration with Frank Pinski, we have recently¹¹ calculated $\rho(T)$ for palladium. The results are shown in figure 1 (colored curve). The calculation can be put into close analogy with Bloch's original work by defining as an intermediate step an effective, weighted density of phonon states for transport processes. In the Bloch-Grüneisen theory, that function is simply proportional to Ω^4 (Ω is the phonon frequency). The two functions are compared in figure 2. The theory is shown in somewhat more detail in the box on the opposite page (below, right).

Bloch-Grüneisen theory

Bloch's semiclassical Boltzmann equations can be solved exactly when the Fermi surface is spherical and the scattering is elastic, but the principal scattering in pure metals is by creation and absorption of lattice vibrations. Bloch solved this only in the limits of high² and low⁸ temperatures and proved that the behavior in between was more difficult to solve. Grüneisen⁹ was undismayed and proposed that since Bloch's low-temperature formula, blindly extrapolated, matched his high-temperature formula, it should be used for all values of T . The resulting "Bloch-Grüneisen formula" is

$$\rho(T) = 4\rho'(T^5/\theta_D^4) \int_0^{\theta_D} dx x^3 C_E(x) \quad (1)$$

where ρ' is $d\rho/dT$ at high T , θ_D is the Debye temperature, and $C_E(x) = [x/2\sinh(x/2)]^2$, is the Einstein function for the specific heat (in units of k_B) of a one dimensional oscillator of frequency ω . The dimensionless parameter x is $\hbar\omega/k_B T$. At high T , equation 1 becomes $\rho = \rho' T$, and at low T , $\rho = 497.7 \rho' T(T/\theta_D)^4$. (Grüneisen had earlier noted empirically the close similarity in temperature dependence between $\rho(T)$ and $TC(T)$ where C is the total specific heat, and on this basis had proposed, incorrectly, that the low-temperature behavior of ρ should be as T^4 .) Equation 1 is now known to be nearly as accurate a solution as Grüneisen could have hoped. However, the underlying model of spherical Fermi surface, Debye phonons (longitudinal branch only), and bears little resemblance to reality for most metals.

As is clear from figure 1, the current microscopic theory fits the data somewhat less well than the older approximate model of Bloch and Grüneisen. However, it is impressive that the microscopic calculation, with *no adjustable parameters*, gives an excellent account of the absolute magnitude and overall T -dependence of $\rho(T)$. This puts us in a much better position than before to assess the significance of the remaining discrepancies. Discrepancies between theory and experiment are easiest to detect at low or high temperatures, because in these ranges theory predicts simpler behavior.

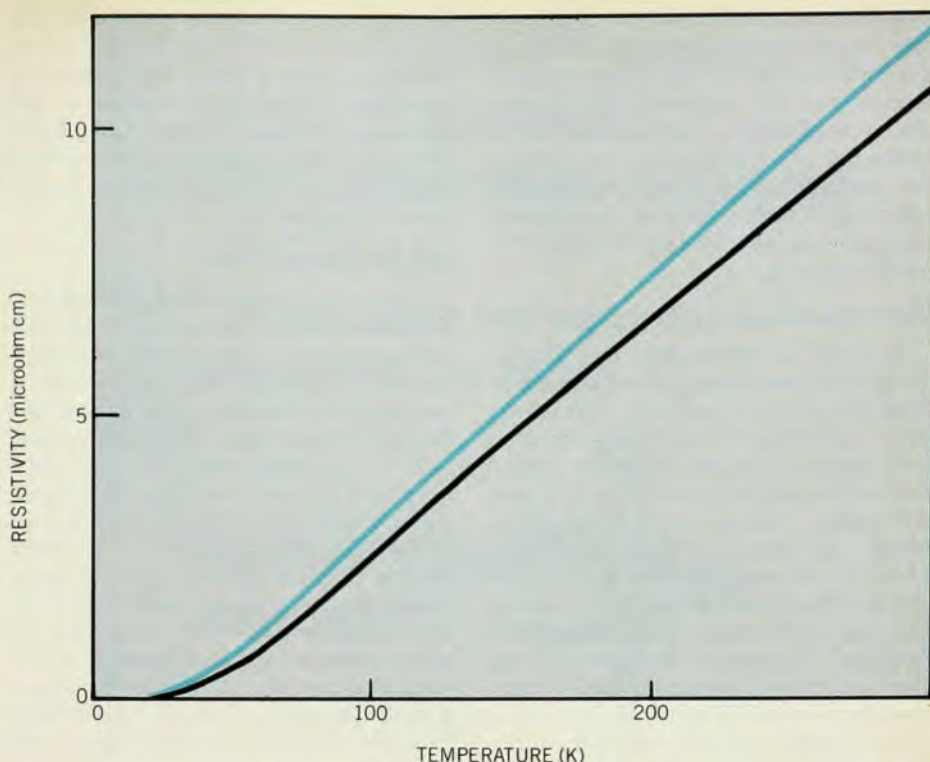
Low temperatures

At low temperatures, Bloch's theory predicts a simple T^5 behavior for the electrical resistivity in pure metals due to inelastic scattering by phonons. There are, of course, other processes that contribute to the resistivity, such as the scattering of electrons due to their Coulomb interaction, and scattering by impurities. An empirical rule, formulated by Augustus Matthiessen in the 1860's, states that the contributions to the resistivity of an impure metal are simply additive, consisting of the pure metal resistivity plus a temperature independent "residual resistivity" due to impurities. If this rule were obeyed, the experimental determination of the low temperature exponent n ($\rho(T) = \rho_0 + aT^n$) would be considerably easier, although still difficult since aT^n is usually much smaller than ρ_0 in the temperature range of power law behavior. However, experiments tend to show strong deviations from this behavior; the low-temperature limit is not such a simple function of ρ_0 and T and contains cross terms. Many different exponents n have been found with little clear regularity, and it is fair to say that confusion prevails.

Recent experiments¹² thus leave us with at least three questions:

- ▶ Is Bloch's limiting T^5 behavior ever observed?
- ▶ Does electron-electron Coulomb scattering appear as a T^2 contribution?
- ▶ What is the source of the systematic deviations from Matthiessen's rule that are seen at low temperatures?

Most researchers believe that these questions can eventually be sorted out in the context of Bloch's theory. The complexity of the theory is such that it is not yet clear what behavior is permitted when there is more than one scattering mechanism. Matthiessen's rule follows from Bloch's theory in a first approximation, but improved approximations allow deviations, which are difficult to calculate exactly. Even in pure materials with only phonon scattering permitted, the "rigorous" T^5 limit is obscured theoretically by the observation that at low enough T , the phonons can no longer be regarded as in equilibrium, and more complicated



Experimental and calculated resistivities of palladium. The Bloch-Grüneisen theory (black curve) fits the data so well that the difference would be hardly perceptible in this figure. There are substantial *relative* differences below 50 K, however. The colored curve shows the results of the modern theory with no adjustable parameters.

Figure 1

equations must be solved.

The direct scattering of electrons from each other by Coulomb interactions plays surprisingly little role in the resistivity. This would be understandable in an isotropic medium with a spherical electron-energy dispersion, because momentum

conserving collisions would conserve current. However, most metals are sufficiently far from spherical symmetry that current conservation is completely broken down. It was observed by Lev Landau and Isaak Pomeranchuk¹³ and W. G. Baber¹⁴ that this should give rise to a T^2

Modern theory

Current knowledge of phonon spectra and Fermi surface dimensions is limited mainly by crystal growing arts and experimental diligence. A correspondingly improved theory of resistivity is available, and we proposed that it should be called the "modernized Bloch-Grüneisen formula."

$$\rho(T) = \left[\frac{1}{3} e^2 N(0) \langle v^2 \rangle \tau \right]^{-1} \quad (2)$$

$$\frac{1}{\tau} = \frac{2\pi}{\hbar} k_B T \lambda_{tr}(T) \quad (3)$$

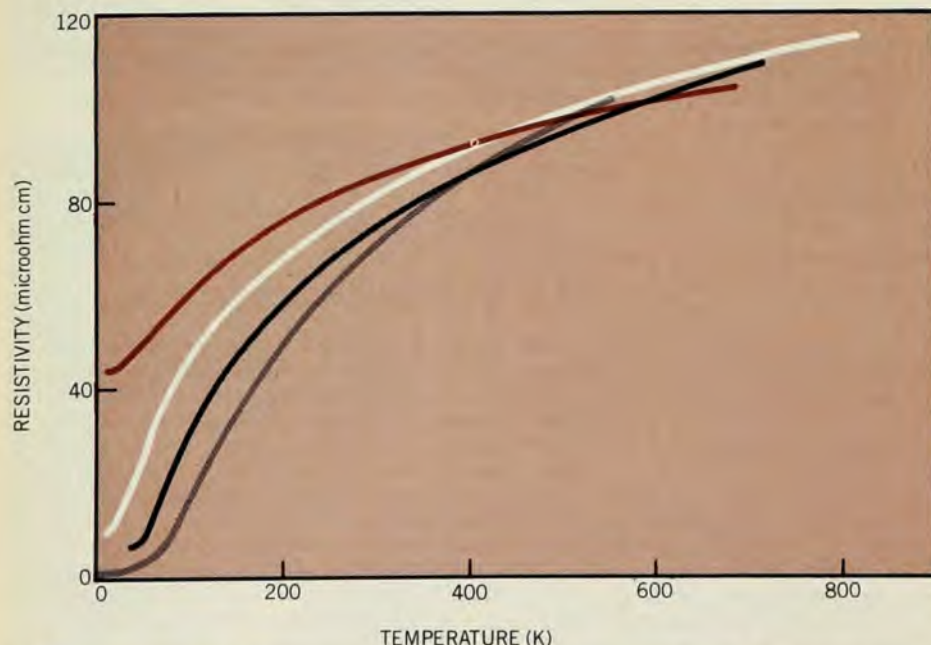
$$\lambda_{tr}(T) = 2 \int_0^{\Omega_{max}} \frac{d\Omega}{\Omega} \alpha^2_{tr} F(\Omega) C_E(\hbar\Omega/k_B T) \quad (4)$$

where $N(0)$ is the density of states (both spins) at the Fermi energy and $\langle v^2 \rangle$ is the mean square Fermi velocity. These parameters can be calculated if the band structure is known and often differ greatly from the free-electron values. The behavior of the scattering rate $1/\tau$ is controlled by the dimensionless coupling constant $\lambda_{tr}(T)$, whose temperature dependence is determined by the Einstein function $C_E(x)$. At high temperatures $C_E = 1$ and $\lambda_{tr}(T)$ becomes a constant, λ_{tr} , whose value ranges between 0.1 and 2.0. The name λ_{tr} is meant to denote the transport (tr) analog of the dimensionless coupling constant λ that determines the superconducting transition temperature and whose value can be deduced from T_c . Similarly, $\alpha^2_{tr} F(\Omega)$ is the analog of the function $\alpha^2 F(\Omega)$ from superconductivity theory, which is the phonon density of states, $F(\Omega)$, weighted with the electron-phonon interaction. The difference between the transport quantities λ_{tr} and $\alpha^2_{tr} F$, and the analogous quantities in superconductivity is a factor of the type $(1 - \cos\theta)$, which causes the behavior of $\alpha^2_{tr} F(\Omega)$ to be as Ω^4 at low temperatures, rather than Ω^2 as seen in superconductivity. Just as equation 1, the above do not represent an exact solution of the Bloch-Boltzmann equation but an accurate variational estimate. Unlike equation 1, however, this version takes full account of the complexities of the Fermi surface and phonon spectrum.

contribution to $\rho(T)$ which would dominate over phonon scattering at both very low and very high T . This effect has not yet received joint experimental and theoretical verification. In transition metals where the effect is expected to be large, low temperature $n = 2$ exponents are sometimes found. However, the expected reappearance of a T^2 term at high T is usually missing.

High temperatures

At high temperatures, the Bloch-Grüneisen theory predicts an increase in resistivity proportional to T . Deviations from that simple behavior are expected for a variety of reasons. Lattice expansion and normal anharmonicity will usually make ρ increase more rapidly than linear, as does Coulomb scattering. Mott-Fermi smearing (the $k_B T$ broadening of the Fermi surface) can cause either positive or negative deviations from linearity. Recently it has been realized that in many transition-metal compounds, ρ versus T curves flatten out at high T (negative deviation from linearity). This seems to be almost a universal effect among high T_c superconducting materials. Some examples are shown in figure 3. Many explanations have been proposed, some invoking processes that may be accounted for in the context of Bloch's theory. However, some theorists believe that it represents a more fundamental breakdown of the framework of the theory itself. It can be argued, for example, that most of the sources of deviation that can be accounted for within that framework allow both positive and negative deviations; the "saturation" behavior shown in figure 3 is, however, too consistently observed to be such an accidental result.



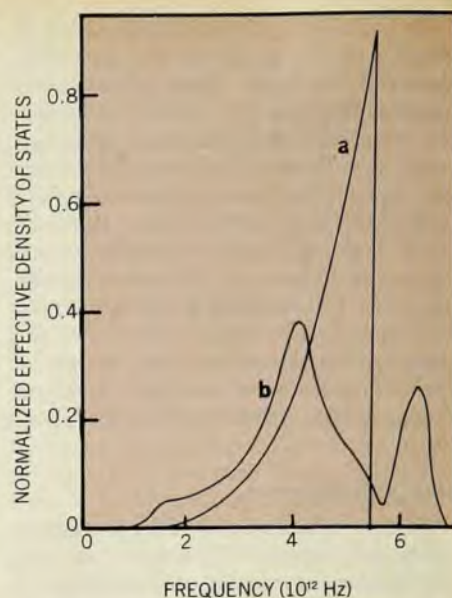
Experimental resistivities for representative A 15 metals: Nb₃Sn white; Nb₃Ge color; Nb₃Sb gray; and V₃Si, black. All four of these except Nb₃Sb have high superconducting transition temperatures. The highest transition temperature is found for Nb₃Ge ($T_c \lesssim 23$ K), which so far has only been grown in thin films with high residual resistivity.

The electron-gas model on which the Bloch-Boltzmann theory is founded is justified by the choice of Bloch states to describe the equilibrium states of the electrons. Perhaps at high temperatures the disorder is so great that new equilibrium states are needed and the gas model must be abandoned.

Microscopic theories

In the last 25 years, many microscopic derivations have appeared of Bloch's transport theory. One of the earliest and easiest is by Walter Kohn and Joaquin Luttinger.¹⁶ These derivations show that Bloch's theory is valid only to lowest order in $(k_F l)^{-1}$ or $(\hbar/\tau E_F)$ where k_F , l , τ , E_F are Fermi wavevector, mean free path, scattering lifetime, and band width, respectively. Experiments seem to indicate that the theory is valid as long as the mean free path is larger than roughly 10 or 20 Å. This result is more reassuring than an earlier critique by Rudolf Peierls¹⁷ which suggested that $\hbar/\tau k_B T$ was the expansion parameter. A particularly careful derivation by Theodore Holstein¹⁸ shows that when the electron-phonon coupling is strong, the mass-renormalization effects which appear in specific heat vanish in dc transport, leaving Bloch's theory intact. Efforts to push beyond lowest order in $(k_F l)^{-1}$ have not so far been well rewarded. Kohn and Luttinger discussed corrections to order $(k_F l)^{-2}$, but it now appears that this term may have an infinite coefficient.

A fundamental problem which confronts perturbation theory is the modern idea of localization especially as worked out by Philip Anderson.¹⁹ Any kind of disorder (induced by impurities or thermal fluctuations, for example) seems to be



Comparison of effective densities of state. Curve a shows the dimensionless function $2\lambda_{tr}(\Omega/\Omega_D)^4$ that can be used in equation 4 to reproduce the Bloch-Grüneisen formula. The parameters λ_{tr} and Ω_D have been adjusted to fit the data of figure 1. Curve b shows the dimensionless, weighted phonon density of states $\alpha^2_{tr} F(\Omega)$ that gives the theoretical curve for $\rho(T)$ shown in figure 1.

able at a critical strength to cause Bloch's extended electron states to localize, a process not described by normal perturbation theory. Conduction in localized states proceeds by "hopping" and lies outside Bloch's theory. There are now several well-documented cases of activated hopping in semiconductors and other non-metallic systems.²⁰ Between the regimes of Bloch's metallic conduction and hopping conduction sits the concept due to Nevill Mott²¹ of the minimum metallic conductance (or the maximum metallic resistance.) It was Zachary Fisk and George Webb²² who introduced the word "saturation" to describe the behavior of figure 3. The idea (which as yet has little detailed justification) is that this is how the metal approaches the maximum resistance. Mott has pointed out that simple dimensional arguments give a resistivity $\rho = \hbar a/e^2$ for a crystal with lattice constant a . This value is of the same order of magnitude as the observed limits seen, for example, in figure 3. The simple phenomenological formula²³

$$1/\rho = 1/\rho_0(T) + 1/\rho_{\max}$$

where $\rho_0(T)$ is the resistivity given by the Bloch theory, fits quite a lot of data. For metals having the A 15 structure, the value of $\rho_{\max}$ is about 150 micro-ohm cm, and appears to be independent of temperature and defect concentration. The widespread occurrence of behavior of this type seems to support Mott's concept and to demand greater theoretical efforts to go beyond Bloch's theory.

After 50 years of effort, many successes, and much progress, we still don't fully

understand either how to use Bloch's theory to explain the low T anomalies, or how to go beyond it to explain the high T effects. For ordinary phenomena, however, the theory remains unchallenged in its usefulness, and the 1928 paper stands as a milestone in physics.

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