Superconductivity in *d*- and *f*-Band Metals

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THEORY OF RESISTIVITY "SATURATION"

*Philip B. Allen**

Department of Physics
State University of New York
Stony Brook, New York, USA

High T_c superconductors at room temperature and above have resistivities rising less rapidly with T than Bloch-Grüneisen theory predicts. Critical discussion of seven possible mechanisms is given. Three mechanisms (anharmonicity, Fermi smearing, T-dependent energy bands) are consistent with Boltzmann theory, and four (localization, Debye-Waller effects, Phonon drag and ineffectiveness, and non-classical conduction channels) are not. It is argued that the last mechanism is the most plausible.

I. THE SATURATION PHENOMENON VERSUS NORMAL METALLIC RESISTIVITY

Figure 1 shows the electrical resistivity of Nb_3Sn as a function of temperature (Woodard and Cody, 1965.). The shape of $\rho(T)$ departs strongly from Bloch-Grüneisen theory (Ziman, 1960). It was pointed out by Fisk and Lawson (1973) that the same shape of $\rho(T)$ is seen in nearly all high T_c d-band metals (and other highly resistive metals as well), whereas low T_c materials tend to have a more normal behavior. Sometimes the effect is called a "bulge" - implying that the anomalous feature is excess resistivity for temperatures near θ_D. This seems to me incorrect; if there were only a bulge, then $\rho(T)$ above θ_D should be normal, i.e., (a) nearly

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linear in T and (b) extrapolating back through ρ_o, the residual resistance. This second constraint - that the intercept is fixed - is often forgotten. It follows from the basic variational solution of the Bloch-Boltzmann equation (Allen, 1978),

$$\rho_{ideal}(T) = \rho_o + [(n/m)_{eff}\, e^2 \tau_{ph}(T)]^{-1} \tag{1}$$

$$1/\tau_{ph}(T) = (2\pi/\hbar)\, k_B T\, \lambda_{tr}(T) \tag{2}$$

$$(n/m)_{eff} = N(o) \langle v_x^2 \rangle \tag{3}$$

$$\lambda_{tr}(T) = 2 \int_o^\infty \frac{d\Omega}{\Omega}\, \alpha^2_{tr} F(\Omega) \left[\frac{\hbar\Omega/2k_B T}{\sinh(\hbar\Omega/2k_B T)} \right]^2 \tag{4}$$

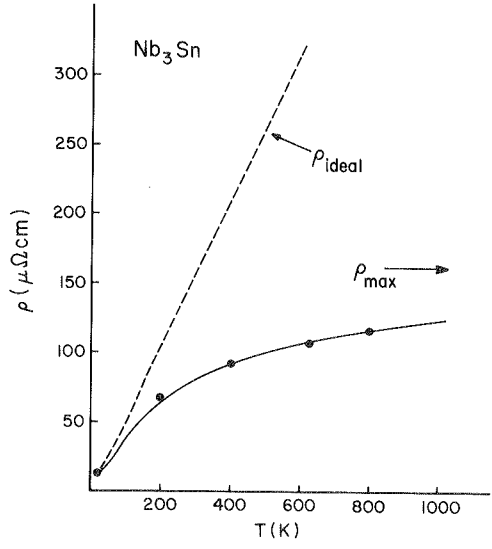

FIGURE 1. Resistivity of Nb_3Sn versus temperature. Dots are data of D.W. Woodard and G.D. Cody (Phys. Rev. 136, A166 (1964).) The solid line is a fit using eq. (6) with ρ_o = 13.5μΩcm and ρ_M = 163μΩcm. Assuming $\lambda_{tr} = \lambda = 1.7$, the fitted value of $(n/m)_{eff}$ gives a Drude plasma frequency 3.7 eV which compares well with 3.4 eV calculated by L.F. Mattheiss et al. (Phys. Rev. B17, (1978).) The dashed curve is ρ_{ideal}, the Boltzmann resistivity calculated from eqs. (1-4) using the phonon density of states of P. Schweiss et al. (in "Superconductivity in d- and f-Band Metals," D.H. Douglass, ed., Plenum, New York, 1976; p. 189) for the shape of $\alpha^2_{tr}F(\Omega)$. The corresponding mean free path at 300K is 7Å, using the Fermi velocity of Mattheiss et al.

where N(o) is the density of states at the Fermi surface, $\langle v_x^2 \rangle$ is the mean square x-component of electron velocity at the Fermi surface, and $\alpha^2_{tr}F(\Omega)$ is a function very similar to $\alpha^2F(\Omega)$ in superconductivity, with a slightly different weighting used in the Fermi surface average. At high temperatures ($T \gtrsim \theta_D$) the factor in brackets in eq. (4) becomes unity and $\lambda_{tr}(T)$ achieves its maximum value, a constant, λ_{tr}, very similar to λ in superconductivity. A simplified version of equation (1) valid at T=0 and at $T \gtrsim \theta_D$ is

$$\rho_{ideal}(T) \approx \rho_o + \rho_1 T \qquad (5)$$

where ρ_1 involves two coupling constants, λ_{tr} and $(n/m)^{-1}_{eff}$.

From eq. (5) it follows that $\rho(T)$ of Nb_3Sn (fig. 1) is badly amiss at high T. One cannot tell (without a detailed theory) whether there is excess resistivity in the "bulge" region, but no detailed theory at all is required to see that at high T, $\rho(T)$ falls below the required linear behavior. A better name for the phenomenon is "deviation from linearity" (d.f.ℓ.), which carries no prejudice as to the cause. Fisk and Webb (1976) have introduced a more descriptive term, "saturation." This word conveys the prejudice (which I think is correct) that the downward turn in $\rho(T)$ represents the approach to Mott's (1971) maximum metallic resistivity (or minimum metallic conductivity), $\rho_{max} = 1/\sigma_{min}$, and is associated with very short electron mean free paths (ℓ). Unfortunately we have no clear microscopic picture for the conduction mechanism in Mott's regime. Mooij (1973) has assembled much evidence that in d-band alloys, ρ saturates at a value $\rho_{max} \approx 150\mu\Omega$ cm.

II. PARALLEL RESISTOR FORMULA

An empirical formula used by Wiesmann *et al.* (1977) fits a great deal of data.

$$1/\rho(T) = 1/\rho_{ideal}(T) + 1/\rho_{max} \qquad (6)$$

If eq. (5) is used for ρ_{ideal}, then there are three adjustable parameters. For A15 metals, ρ_{max} is found to be $\sim 150\mu\Omega$ cm. Only ρ_o varies significantly when eq. (6) is fit to data for the same material with varying degrees of radiation damage (Gurvitch, 1978). The electron-phonon parameter ρ_1 derived from fitting (6) to experiment, agrees with independent theoretical estimates of ρ_1 by Allen *et al.*

(1978a) for $Nb_3A\ell$ and Nb_3Ge. These estimates consist of calculating $(n/m)_{eff}$ from band theory and estimating $\lambda_{tr} \sim \lambda$, taking λ from T_c. This procedure was used successfully by Chakraborty *et al.* (1976) for elements like Nb where "saturation" is not seen at 300K and below. Infrared experiments (Mattheiss *et al.*, 1978) confirm the value of $(n/m)_{eff}$. Detailed theoretical calculations (Pinski *et al.* 1978) of λ and λ_{tr} for Nb and Pd show that these two numbers differ by only 10%. From the theory used to estimate ρ_1 it is found that if Boltzmann theory worked, ℓ at 300K would be 3-4Å in Nb_3Ge and $Nb_3A\ell$.

The success of eq. (6) suggests that it may be more than an accident. Perhaps an extra conduction channel parallel to the Boltzmann channel is physically real. If so, it should be a general occurrence, available in all metals. This does not contradict the data. In elements not showing saturation, ρ_{ideal} is sufficiently small compared to 150μΩ cm that it is difficult or impossible to detect the "parallel resistor."

III. WHAT IS BLOCH-BOLTZMANN THEORY?

The first step is to understand thoroughly the ordinary theory of metallic conduction formulated by Felix Bloch (1928). The Bloch-Boltzmann equation for electrons in steady-state in a dc external electric field E is

$$- e \underset{\sim}{E} \cdot \underset{\sim}{v}_k \, (-\partial f/\partial \varepsilon_k) = \sum_{k'} Q_{kk'} \phi_{k'} \tag{7}$$

where the distribution function F_k for electrons in state $|k\rangle$ with velocity $\hbar \underset{\sim}{v}_k = \partial \varepsilon_k / \partial \underset{\sim}{k}$ is $f(\varepsilon_k) + \phi_k \, (-\partial f/\partial \varepsilon_k)$ and f is the Fermi function. The scattering operator $Q_{kk'}$ is the sum of terms for impurity scattering, phonon scattering, and electron-electron (Coulomb) scattering. This equation has been linearized, which means that it only gives the Ohmic part of the current. This equation rests on four assumptions:

(A) The $\underset{\sim}{E}$ field and the currents are described by the semiclassical theory. That is, the E field "accelerates" electrons according to $\hbar \dot{\underset{\sim}{k}} = -e\underset{\sim}{E}$, and the current is caused by the resulting excess of electrons with a band velocity pointing along $\underset{\sim}{E}$,

$$\underset{\sim}{j} = -e \sum_k \underset{\sim}{v}_k \, \phi_k \, (-\partial f/\partial \varepsilon_k) \tag{8}$$

(B) Scattering events are statistically independent of each other; they are separated by enough wavelengths that the electron "recovers" from previous collisions before experiencing the next one. For free electrons the criterion is $k_F \ell >> 1$.

(C) The phonon part of the collision operator is calculated using only the first term in the Taylor series

$$\mathcal{H}_{ep} = \sum_{\ell} [\underset{\sim}{u}_\ell \cdot \underset{\sim}{\nabla}_\ell V + \frac{1}{2} \underset{\sim}{u}_\ell \underset{\sim}{u}_\ell : \underset{\sim}{\nabla}_\ell \underset{\sim}{\nabla}_\ell V + \ldots] \qquad (9)$$

where u_ℓ is the displacement of the ℓ^{th} atom and V is the crystal potential. Specifically, the phonon part of Q is second order in $(u \cdot \nabla V)$. Higher powers of $(u \cdot \nabla V)$ would give effects higher order in $(k_F \ell)^{-1}$ and so are omitted according to (B). Higher order terms in the series (8), treated to second order in perturbation theory, will give corrections of order $(u/a)^2$ and higher; these are omitted.

(D) The phonons are assumed to be in thermal equilibrium.

In spite of the four assumptions, this theory is remarkably sophisticated and includes many processes to infinite order of perturbation. This sometimes surprises theorists unfamiliar with the details. Feynman diagrams provide a classification scheme which is helpful (to some) in understanding the content and limitations of eq. (7). Figure 2 shows representative graphs which are completely included in eq. (7). It is surprising that no renormalization effects occur to alter eq. (7) at the level of the graphs depicted in fig. 2. The most careful proof is by Holstein (1964). He found that for ac fields, when $T < \theta_D$ and $\omega \lesssim \omega_D$, renormalization effects do occur, but they cancel as $\omega \to 0$.

Representative graphs which are not summed by eq. (7) are shown in fig. 3. Each graph has been classified according to which of the four assumptions A-D is violated. Graphs A will be discussed in Sec. XI, B in VII, C in VIII, and D in IX.

IV. ANHARMONICITY

There are two effects within Boltzmann theory which can cause the "ideal" resistivity to depart from linearity at high temperature. Unfortunately, they don't seem sufficient to explain saturation. They are described in this section and the next. The simplest is the fact that phonon frequencies change with temperature partly because of thermal expansion. For convenience we call this "anharmonicity,"

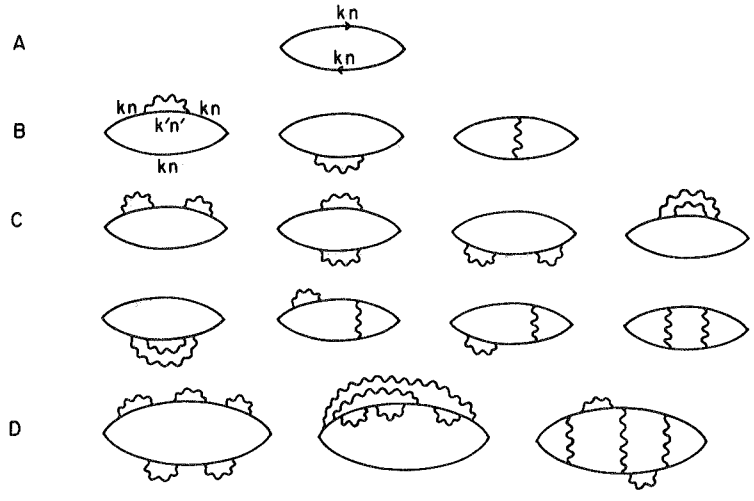

FIGURE 2. Representative Feynman graphs summed in Bloch-Boltzmann theory. Subsets A,B,C are zeroth, first, and second-order graphs respectively; D shows a few of the fifth-order graphs which are summed.

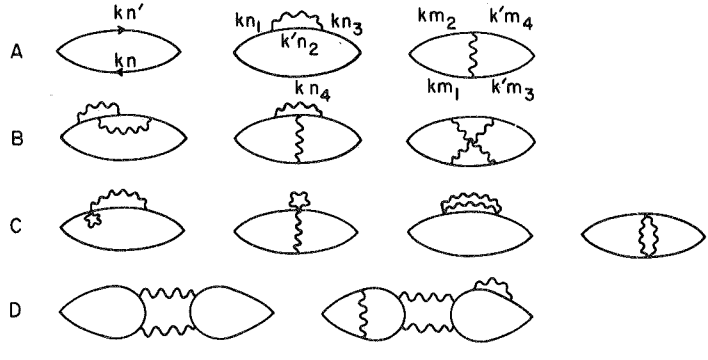

FIGURE 3. Feynman graphs omitted in Bloch-Boltzmann theory. Subsets A,B,C,D are the lowest-order graphs omitted because of assumptions A,B,C,D. In the special cases (n'=n) or ($n_1=n_3=n_4$) or ($m_1=m_2$ and $m_3=m_4$), graphs A reduce to graphs of fig. 2.

even though the mechanism is possibly not anharmonic coupling. To estimate the importance of anharmonicity, Allen *et al.* (1976) followed McMillan (1968) and wrote

$$\lambda_{tr} = \eta/M\langle\omega^2\rangle \tag{10}$$

Thus the effect of anharmonicity on $\rho(T)$ at high T is through a factor $\langle\omega^2\rangle^{-1}$. Normal behavior is for $\langle\omega^2\rangle$ to diminish with increasing T, causing $\rho(T)$ to rise above linearity. However, many A15 metals have anomalous anharmonic behavior, and $\langle\omega^2\rangle$ is known to increase. It has been estimated that at most 1/3 of the d.f.ℓ. can be explained this way (Bader and Fradin, 1976).

V. FERMI SMEARING

Eq. (8) can be written

$$\sigma(T) = \int_{\infty}^{\infty} d\varepsilon\ \sigma(\varepsilon,T)\ (-\partial f/\partial\varepsilon) \tag{11}$$

$$\sigma(\varepsilon,T) = \sum_k e^2 v_{kx}^2\ \tau_k\ \delta(\varepsilon_k-\varepsilon) \tag{12}$$

where τ_k is *defined* by the relation $\phi_k = -e\ \underset{\sim}{v}_k \cdot \underset{\sim}{E}\ \tau_k$ and $\underset{\sim}{E}$ is in the $\hat{x}$ direction. Mott (1936) pointed out that in d-band systems, $\sigma(\varepsilon,T)$ may vary with ε on a scale k_BT, so that $(-\partial f/\partial\varepsilon)$ in (11) may *not* be replaced by $\delta(\varepsilon)$. The resulting corrections are called "Mott Fermi smearing." The effect on the T-dependence of ρ can be significant when energy bands are narrow and the Fermi velocity changes rapidly with ε. Many authors have made model calculations purporting to show how saturation in A15 metals can be explained this way (Cohen, Cody, and Halloran, 1967; Bader and Fradin, 1976; Nakayama and Tsuneto, 1978; Ting, Synder, and Williamson, 1979). Unfortunately it is extremely difficult to calculate this effect, and most of the existing literature on the subject is wrong. A reasonable procedure (Allen, 1976) starts by defining a density of state $N(\varepsilon)$, mean square velocity $v_x^2(\varepsilon)$, and scattering rate $\tau(\varepsilon)$ through the relations

$$N(\varepsilon) \equiv \sum_k \delta(\varepsilon_k-\varepsilon) \tag{13}$$

$$N(\varepsilon)v_x^2(\varepsilon) \equiv \sum_k v_{kx}^2\ \delta(\varepsilon_k-\varepsilon) \tag{14}$$

$$N(\varepsilon)v_x^2(\varepsilon)\tau(\varepsilon) \equiv \sigma(\varepsilon,T)/e^2 \qquad (15)$$

Calculating $\tau(\varepsilon)$ requires solving Boltzmann's equation. It is not permissible to use the standard variational solution which assumes $\tau(\varepsilon)$ = const. Roughly speaking, $1/\tau(\varepsilon)$ behaves as $N(\varepsilon)$ with some additional factors which (hopefully!) vary less rapidly with ε. Note that $N(\varepsilon)$ *cancels* out of $\sigma(\varepsilon,T)$ leaving $v_x^2(\varepsilon)$ as the parameter to be smeared. Attempted calculations along these lines (Laubitz *et al.*, 1977; Allen and Chakraborty, 1978; Pinski *et al.*, 1978) have been moderately successful for the elements. There is no doubt that the effect on ρ_{ideal} can be significant. However, it is hard to believe that Fermi smearing - a complex and band-structure-dependent effect, can account for the simple and regular behavior of so many materials. Why should $\rho(T)$ not sometimes bend upwards, or else bend too far in the downwards direction?

VI. ALTERATIONS OF BAND STRUCTURE WITH T

Two suggestions are sometimes made based on intuitive plausibility: (i) putting a T-dependent band structure into eq. (7), or (ii) introducting a phenomenological lifetime broadening (Testardi and Mattheiss, 1978; Wiesmann *et al.*, 1978; Greig and Morgan, 1973) in addition to the thermal broadening of eqs. (11-15). The latter receives some support from a CPA theory by Chen *et al.* (1972); also Brouers (1978), but this theory omits electron-hole interactions and deals only with a single band. A rigorous theory (making neither of these simplifications, but valid only for weak disorder) has been given by Chakraborty and Allen (1978, 1979). Their theory can account for T dependent shifts in $\sigma(\omega)$ at optical frequencies in semiconductors, and gives a generalization of Boltzmann theory in the dc limit (discussed further in sec. XI.). Unfortunately their theory is in no way equivalent to a modified version of eq. (7). No justification is found for either intuitive procedure, except that for the trivial effects of thermal expansion, a T-dependent band structure can be used in (7). This latter is a small effect, less significant than the thermal shifts of phonon frequencies already discussed in sec. IV.

VII. CONNECTION WITH ANDERSON LOCALIZATION

The remaining sections deal with attempts to explain saturation outside the framework of Bloch-Boltzmann theory. Of the omitted graphs in fig. 3, the hardest to deal with are those of 3B. They contribute to ρ terms which are higher order in $(k_F\ell)^{-1}$. It is known (Langer and Neal, 1966) that a straightforward expansion of σ in powers of $(k_F\ell)^{-1}$ has logarithmically divergent coefficients in third order and higher. There is as yet no accepted method for solving this problem. It seems likely that the difficulties with perturbation theory relate to an actual physical singularity, the onset of Anderson localization (Anderson, 1958; Thouless, 1974). This subject remains difficult and controversial. Only simple models involving a single band have been studied, whereas A15 metals have many overlapping bands. At T=0 it is generally agreed that σ will vanish above a critical value of the disorder, d_c. There is some controversy about whether σ vanishes continuously (as $(d_c-d)^P$ for some positive P) or discontinuously (as $\theta(d_c-d)$). Older evidence suggested the latter, with the value of σ just below d_c being σ_{min}. This strongly suggests a connection between saturation and localization. More recent evidence suggests the former, which seems incompatible with saturation, i.e. there is no hint of an *excess* conductivity before localization occurs.

The situation is even less clear at finite temperature. For $d>d_c$, either thermally assisted hopping or activation above a mobility edge will allow σ to be finite. For $d<d_c$, little work has been done. The data of fig. 1 suggest the question whether vibrational disorder can *cause* localization, and if so how will $\sigma(T)$ behave above and below the threshold. In summary, there is yet not much evidence for a close connection between saturation and localization.

VIII. DEBYE-WALLER FACTORS

It has been proposed at various times that when an electron scatters from the atomic displacement, a Debye-Waller factor should enter, weakening the strength of the scattering potential, just as for x-ray scattering. This is undoubtedly true, and is described diagramatically by dressing the electron-phonon vertex by additional closed phonon loops which represent virtual emission and reabsorption of the same phonon. (fig. 3C, first two). Clearly this will give a diminished high-T resistivity, as observed

experimentally (Rossiter, 1977; Visscher, 1978). There are two difficulties. First, the Debye-Waller expansion parameter is (u/a), a number which behaves roughly the same in all metals, approaching the Lindemann limit at melting. There is nothing to discriminate strong-scattering metals where saturation is seen from weak-scattering metals like Cu or Aℓ where it is not. Second, there is a theoretical objection (Sham and Ziman, 1963) that multi-phonon scattering enters in the same order of perturbation theory as Debye-Waller effects, (fig. 3C, second two) and clearly enhance the resistivity. The degree to which the two effects cancel is unknown. Speculations of exact cancellation seem unfounded. It is difficult to estimate the effects well and the matter awaits further theoretical development.

IX. PHONON DRAG

It is known that in a metal carrying a current, the phonons are not exactly in thermal equilibrium. In an idealized case one can imagine that the phonons are dragged with the current, so that in the rest-frame of the drifting electrons, the phonons and electrons are both close to thermal equilibrium. Fewer current-degrading collisions occur and the conductivity is enhanced (Ziman, 1960). This picture applies only if phonons decouple from everything except electrons. In actuality, phonons couple to each other (by anharmonic coupling) and to defects, allowing relaxation toward equilibrium. Thus phonon drag is expected to be significant only in pure metals at low temperatures where anharmonic and defect effects are small. A theory is achieved by generalizing eq. (7) to two coupled Boltzmann equations, one for electrons and one for phonons. Holstein (1964) has shown that these coupled Boltzmann equations can be derived by summing up the additional graphs of fig. 3D.

X. PHONON INEFFECTIVENESS

Morton *et al.* (1978) and Cote and Meisel (1978) have independently proposed the hypothesis that phonons with $Q\ell < 1$ become ineffective scatterers. Little justification has been offered except that it gives an economical explanation for the resistivity data and for the degradation of T_c by radiation damage. Two things are bothersome. First, in

the spirit of Mott's ρ_{max}, saturation should be independent of which mechanism causes the principal scattering (defects, phonons, or Coulomb scattering, whereas "phonon ineffectiveness" treats only the phonon mechanism. Second, amorphous s-p metals have T_c values comparable to their crystalline counterparts and tunneling shows excess strength in $\alpha^2F(\Omega)$ at low Ω. If phonon ineffectiveness applies, one would expect the small Ω phonons with small Q to have a deficiency of weight in α^2F.

Both groups proposing phonon ineffectiveness have invoked Pippard's (1955) work on ultrasonic attenuation. Pippard showed that if the impressed phonon has $Q\ell < 1$, then electrons are ineffective in degrading the ultrasonic energy. This is because electrons are dragged by the impressed phonon wave. The connection between Pippard's ideas and saturation seems obscure. The thermal phonons present during electrical conduction are incoherent and cannot all drag electrons. It seems to me that if there is truth in the idea of "phonon ineffectiveness," it is contained in the phonon drag processes of sec. IX.

XI. BEYOND SEMICLASSICAL THEORY

Finally we turn to the graphs of fig. 3A. These graphs were summed in the generalized Boltzmann equation of Chakraborty and Allen (1978, 1979), as a solution to the problem of how to include temperature-dependent electron bands into conductivity theory. Both the semiclassical acceleration equation and the semiclassical current (eq. 8) must be supplemented by non-classical terms which give the interband dipole transitions and the interband currents. These graphs by themselves would give significant contribution to $\sigma(\omega)$ only for values of ω in the interband energy range. However, when summed to high order along with the graphs of fig. 2, a new channel for d.c. conduction is found. The resulting theory, when solved to first order in the new effects, has exactly the form of the parallel resistor model, eq. (6). The "shunt resistor" ρ_{max} represents the new dc current channel available when interband currents, interband excitation by the $\tilde{E}$ field, and interband scattering are allowed to mix with the usual semiclassical processes. Disorder both allows and inhibits these processes. For example, a virtual interband excitation by the $\tilde{E}$ field could not affect the dc current unless a virtual interband scattering event were available to restore energy conservation. On the other hand, the amount of current carried in this fashion is limited by collisions just

as in the semiclassical case. The expression obtained for $\sigma_{min} = 1/\rho_{max}$ is quite complicated. Collisions appear in both numerator and denominator, leaving a result which is formally independent of the strength of the disorder. The order of magnitude of the term is $\sigma_{min} \sim nhe^2/mE_B$ where E_B is a band separation. This has the right size to account for saturation. Comparing with Boltzmann conduction the ratio is $\hbar/\tau E_B$, similar to $1/k_F\ell$. However, this is only a first order solution. It is not clear why the phenomenological eq. (6) should be successful in the range where the second term dominates. Why do higher order terms not appear?

XII. FINAL COMMENTS

On studying "saturation" we have the double misfortune of a complicated physical parameter (conductivity) and a complicated system (A15 metals). However, we have the good fortune of a clean experiment where disorder can be tuned in a simple reversible way (temperature) and where the results, although unexplained, are simple (eq. 7). It is my prejudice that none of the proposed explanations, except the non-classical channel of sec. XI, have the requisite combination of universality and microscopic validity. If the theory of sec. XI is the correct explanation , we are still lacking a complete or adequately simple picture of the phenomenon.

There is a simple verbal description (V. Heine, private communication) of conduction in the Mott regime, even though there is still no simple mathematical description. Electrons in d-band compounds spend quite a lot of time circulating around the transition metal atom they happen to be associated with, before moving on to the next one. When disorder is weak, the process of motion from one unit cell to the next has phase coherence, and Bloch states are formed which carry currents (until interrupted by scattering.) However, if the disorder is high, phase memory may be lost within a unit cell (Ohkawa 1978) and the resulting eigenstate carry no current. Collisions now play a dual role of helping create the current through transitions between states, and preventing the current from continuing too far in the new state. Thus it is possible for ρ_{max} to be independent of the strength of the disorder.

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