In the face of these uncertainties, we can at best come to negative conclusions. First, it is clear that the closed sheet of the dHvA data does not correspond to the dominant wing contour C_4 of the RFSE. It is likely that the closed sheet is C_5, which then would be the c axis view of the same piece of the Fermi surface for which A_4 is the a axis view. If the dominant wing C_4 is assigned to band-7 of Wood's calculation then the A_4-C_5 sheet should presumably be identified with band 5. The difficulty here is that the A_4 area, whether determined from the dHvA or the RFSE, is twice the APW area of both bands 7 and 5.[6] Secondly, the numerical agreement between the area of C_4 and the "broken sheet" of the dHvA data makes less plausible the suggestion that partial magnetic breakdown occurs at the c axis zone boundary. The disappearance of oscillations for **H** slightly off the c axis, which was one of the factors in Goldstein and Foner's analysis, could be explained if the wings were compressed in the k_c direction, so that they look more like elliptical cylinders than the extended structures shown in their drawings. Finally, the assignment of the 0.081 Å^{-2} oscillation of the dHvA data to Wood's band 8 is inconsistent with C_6. Despite the good agreement of C_6 with the APW results, both with respect to shape and area, the absence of corresponding contours in the A and B crystals, particularly for **H** parallel to c, indicates that the true band-8 surface may well be much more complicated than envisaged in Wood's model.[31]

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[31] Alternatively, it is conceivable that C_6 is not related to band 8 at all. For example, if the wings of C_4 are altered slightly so that extremal diameters can exist (e.g., prolate ellipsoids truncated by the junction at the body of the butterfly), then one can show that there will be a set of orbits lying entirely on the wings which generate the contours C_6 and C_5 (case IV) in substantial detail. In particular, the gap in C_5 is predicted, and the quasidegeneracies along the k_a and k_b axes are topological coincidences. This model predicts a wing C_4 which is too large by a factor of 1.3, however.

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Ideal Resistivity of Bismuth-Antimony Alloys and the Electron-Electron Interaction

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The magnitude of a T^2 term occurring in the low-temperature electrical resistivity of bismuth and bismuth-antimony alloys has been measured. Antimony concentrations were less than 3 at.%. The rate of increase of this term with increasing antimony concentration (decreasing free-carrier concentration) suggests that it may be associated with electron-electron scattering due to an interaction which is appreciably phonon-mediated (Fröhlich-BCS type).

INTRODUCTION

A NUMBER of measurements of the electrical resistivity of bismuth at low temperatures have shown that electron and hole mobilities corresponding to an "ideal" component vary as T^{-2} below 30°K.[1–5] Although the magnitude of the T^2 resistivity term increases slightly in a range covering a few degrees near 10°K, above and below this range mobilities are proportional to T^{-x}, where $1.9 < x < 2.1$. For single crystals, the magnitude of the T^2 component does not change with orientation.[4]

The T^2 dependence has been difficult to understand on the basis of the electron-phonon scattering usually associated with the ideal resistivity at low temperatures. The cutoff wave vector for acoustic phonons active in intravalley electron scattering at low temperatures must be the largest dimension of the Fermi surface, since in bismuth this dimension is much less than the Debye cutoff vector. The phonon energy corresponding to the largest dimension of the Fermi surface

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[1] G. K. White and S. B. Woods, Phil. Mag. **3**, 342 (1958); Can. J. Phys. **33**, 58 (1955).
[2] A. L. Jain, Phys. Rev. **114**, 1518 (1959).
[3] R. N. Zitter, Phys. Rev. **127**, 1471 (1962).
[4] A. N. Friedman, Phys. Rev. **159**, 553 (1967).
[5] S. M. Bhagat and D. D. Manchon, Phys. Rev. **164**, 966 (1967).

is equal to the thermal energy at $\sim 10°K$.[5,6] Since the phonon statistics for bismuth at liquid-helium temperatures are well described by the Debye model (shown by the thermal conductivity of the lattice[5]), the temperature dependence of the electron-phonon contribution to the electrical resistivity should proceed from a T^3 or stronger dependence at temperatures much less than 10°K to a linear dependence at temperatures somewhat larger than 10°K and higher. Although an approximately T^2 dependence of this contribution can be expected over a range of 1 or 2°K, clearly the electron-phonon scattering will not yield a T^2 dependence throughout the range observed.[7] Furthermore, no adjustment of the phonon cutoff wave vector will lead to this behavior. Perhaps the electron-phonon scattering is responsible for the anomalous increase in resistivity which saturates above 10°K, although at the present time this cannot be shown.

An alternative explanation of the T^2 dependence could be that the low-temperature limit of the ideal resistivity is dominated by electron-electron scattering. Recently, measurements of the Lorenz numbers of transition metals have been discussed in this regard.[8–10] (It has also been suggested that several nontransition metals exhibit an electron-electron term in the resistivity.[11]) Such measurements are difficult for bismuth, since the thermal conductivity is dominated by a lattice component. If such measurements could be made, they might not be diagnostic, since electron-phonon scattering in bismuth is wide angle above only a few degrees Kelvin. The Lorenz number would be constant with temperature at higher temperatures for either electron-electron or electron-phonon scattering.

The alloy $Bi_{1-x}Sb_x$, with x small, is somewhat unusual in that electron mean free paths remain long even when the antimony concentration becomes several atomic percent. It is possible to measure accurately the ideal resistivity of such alloys with x in the range $0<x<3$ at.%. Since the band overlap responsible for the free carriers decreases with increasing x,[12] the strength of the T^2 term in the resistivity can be measured as a function of the free-carrier density. We will discuss such measurements and the information which they might reveal regarding the electron-electron interaction in bismuth.

MEASUREMENTS

The electrical resistance of bismuth and several bismuth-antimony alloys was measured at liquid-helium temperatures using a galvanometer amplifier, to an accuracy of better than 0.5%, and with temperatures measured to a comparable accuracy or better with a helium-gas thermometer and vapor-pressure thermometry. The cryostat and temperature control system have been described by White and Woods.[1] The geometric factors necessary to determine the resistivity were accurate to better than 1%. On pure bismuth it was found that compensating the earth's magnetic field with Helmholtz coils did not affect the measurements. (The residual resistance ratio was 21.) As a precaution, however, sample currents were maintained below 0.1 A to prevent effects from self-magnetic fields due to the current flow. Potential and current leads were attached to the specimens with bismuth-cadmium solder.

All the specimens were in the shape of rods with a circular cross section approximately 8 mm in diameter. The distance between potential leads was typically 40 mm. The large specimen dimensions were chosen to avoid any possible complications due to size effects. (For the residual resistance of our specimens, no size effects are expected with diameters greater than 1 or 2 mm.[4]) The starting materials for the specimens were bismuth and antimony of 6–9 grade, purchased from Cominco Ltd., Montreal, Can. For the alloys, bismuth and antimony were zone-refined before alloying.

To complement measurements on pure bismuth single crystals discussed in the literature,[5] measurements were made on a nominally pure polycrystalline specimen. The measurements are shown in Fig. 1. The resistivity is well described by

$$\rho = A + BT^2 + CT^n. \quad (1)$$

The coefficient B is the same for trigonal and bisectrix orientations of the single crystals and for the polycrystalline specimen (Table I). (Measurements by Okada[13] and by Friedman[4] show an anisotropy of $\sim 20\%$ in the ideal resistivity at room temperature which vanishes at low temperatures.) Changing the residual resistance by a factor of more than 15 does not affect the magnitude of the T^2 term, and Matthiesen's rule is well obeyed. The exponent n in the third term is difficult to evaluate, since this term is small, but appears to fall in the range

[6] A discussion by Friedman (Ref. 4) suggests an "effective" Debye temperature of 50°K due to the limiting dimension of the Fermi surface. This discussion is incorrect, since the largest dimension is $\lesssim 10\times10^{-21}$ g cm sec^{-1}. See V. S. Edelman and M. S. Khaikin, Zh. Eksperim. i Teor. Fiz. **49**, 107 (1965) [English transl.: Soviet Phys.—JETP **22**, 77 (1966)]; A. P. Korolyuk, Zh. Eksperim. i Teor. Fiz. **49**, 1009 (1965) [English transl.: Soviet Phys.—JETP **22**, 701 (1966)]; M. S. Khaikin and V. S. Edelman, Zh. Eksperim. i Teor. Fiz. **47**, 874 (1964) [English transl.: Soviet Phys.—JETP **20**, 587 (1965)]. The momentum corresponding to a phonon of energy $k_B\times1°K$ is 1.06×10^{-21} g cm sec^{-1}.

[7] It is suggested in Ref. 5 that a T^2 temperature dependence could be obtained for an electron-phonon resistivity term in the range 1.25–4.2°K, using a cutoff or effective Debye temperature of 9°K. This discussion depends on $T^3J_3(\Theta/T)\alpha T^2$ in this range, where J_3 is a Debye integral. We have evaluated the J_3 integral numerically, and a T^2 dependence does not occur even approximately in the range 1.25–4.2°K, much less throughout a much wider range.

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[11] J. C. Garland and R. Bowers, Phys. Rev. Letters **21**, 1007 (1968).

[12] Yi-Han Kao, R. D. Brown, III, and R. L. Hartman, Phys. Rev. **136A**, 858 (1964).

[13] T. Okada, J. Phys. Soc. Japan **12**, 1327 (1957).

3–5. (This term is negligible below 5°K.) The exponent n cannot be determined more accurately by extending measurements to higher temperatures, since this term saturates in the range 10–15°K.[4] Perhaps the CT^n term rather than the BT^2 term should be associated with the electron-phonon resistivity.

Polycrystalline $Bi_{1-x}Sb_x$ alloys were prepared by heating appropriate proportions under vacuum in a silica tube to an initial temperature of ~700°C, mechanically agitating, mixing for several hours at ~350°C, and then quenching the melt. Specimens were subsequently annealed for several days under vacuum at ~260°C. Chemical analysis of several pieces of each alloy specimen showed that the antimony concentration was constant on the millimeter scale over the specimen, within the accuracy of the analysis (~3%). Homogeneity on the micron scale was examined by electron microprobe. This method does not yield high accuracy for our low-concentration alloys; the statistical uncertainty due to limited counting rates was about 10%. The results show that the antimony concentration does not vary by more than about 10% over distances of 100 μ. Resistivity measurements on the 2% alloy before and after annealing gave identical values for the coefficient B, showing either that microscopic homogeneity is not an important factor or that the specimens are very homogeneous. The resistivity of all the alloys varied in the same manner with temperature as for pure bismuth, although different values were obtained for A, B, and C in Eq. (1). The electrical resistivities of these alloys are shown in Fig. 1. Composition, residual resistance ratio, and magnitude of the coefficient B for bismuth and the alloys are summarized in Table I.

TABLE I. Specimen parameters.

Specimen	$\rho_{300}/\rho_{4.2}$	Lateral dimensions (mm)	B ($\mu\Omega$ cm/°K^2)
Bismuth, trigonal axis[a]	370	3.30×3.60	0.008
Bismuth, bisectrix axis[a]	320	3.30×3.95	0.008
Bismuth, polycryst.	21	~8.0 diam	0.0078±0.0004
$Bi_{99.1}Sb_{0.9}$, polycryst.	9.5	~9.0 diam	0.0106±0.0005
$Bi_{98.3}Sb_{1.7}$, polycryst.	9.5	~8.0 diam	0.0129±0.0007
$Bi_{97.8}Sb_{2.2}$, polycryst.	8.6	~8.0 diam	0.0155±0.0007

[a] See Ref. 5. To calculate B, we have assumed a room-temperature resistivity of 115 $\mu\Omega$ cm in both cases.

The variation of B with antimony concentration is shown in Fig. 2. As discussed by Ziman,[14] with two terms in the resistivity, departures from Matthiesen's rule are expected to be greatest when the two terms are of comparable magnitude. Since Matthiesen's rule is well obeyed for "pure" bismuth through a wide range of residual resistivity, it should be well obeyed in the alloys, where the ideal resistivity is somewhat smaller in comparison to an increased residual resistivity. (Further, it is worth remarking that the ratio A/B does not change by more than a factor of 2 for the alloys.)

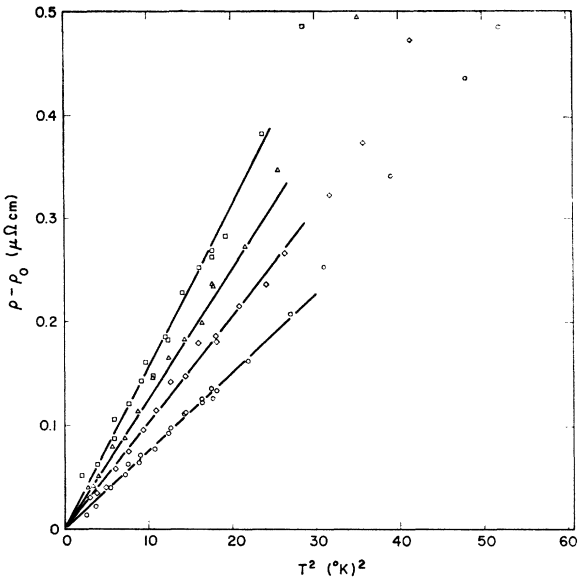

FIG. 1. Ideal resistivity $\rho-\rho_0$ versus T^2 for pure bismuth and bismuth-antimony alloys. ○, pure bismuth; ◇, $Bi_{99.1}Sb_{0.9}$; △, $Bi_{98.3}Sb_{1.7}$; □, $Bi_{97.8}Sb_{2.2}$.

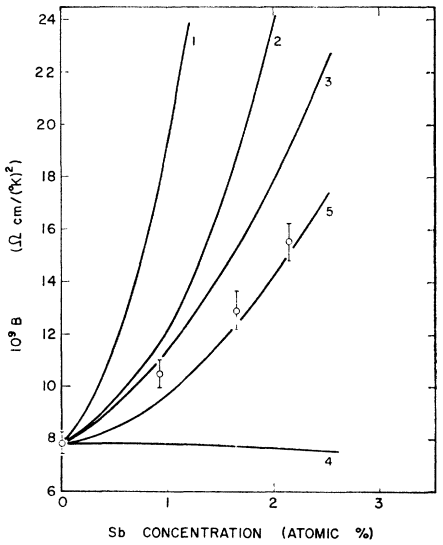

FIG. 2. Coefficient B [Eq. (1)] as a function of antimony concentration. Circles are experimental points. Curves 1–5 give the variation for the various scattering processes discussed in the text: 1, hole-hole scattering via a Coulomb interaction potential; 2, electron-electron scattering via a Coulomb interaction potential; 3, hole-hole scattering via a phonon-mediated interaction potential; 4, electron-electron scattering via a phonon-mediated interaction potential; 5, electron-phonon scattering (here the k_F^{-2} variation characteristic of either electron-phonon or hole-phonon scattering is shown, assuming that statistical factors, scattering angles, etc., yield a hypothetical T^2 dependence for electron-phonon scattering).

[14] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1960), p. 285.

DISCUSSION

Before we consider the possibility that electron-electron scattering dominates the low-temperature limit of the ideal resistivity of bismuth, a few brief comments will be made on the band structure. de Haas–van Alphen measurements on bismuth-antimony alloys by Kao *et al.*[12] and by Brandt *et al.*[15] show that the shape of the three ellipsoids in the electron band does not change appreciably as the degeneracy energy decreases with increasing antimony concentration. In addition, effective-mass ratios for different orientations on the Fermi surface are preserved, although at all points the effective mass decreases with alloying because of the well-known nonparabolic dispersion relation.[12,15,16] The hole band is parabolic, and therefore the single-hole ellipsoid will remain constant in shape as well.

With electron scattering in low-temperature transport measurements dependent on wave vectors and effective masses at the Fermi surface, $k_F(\theta,\phi)$ and $m_F^*(\theta,\phi)$, the dependence of ideal resistivity on the free-carrier density can be described in terms of particular values $k_F(0,0)$ and $m_F^*(0,0)$ and of angular functions which do not change with alloying. (This does not hold for electron-hole scattering, since the shape of the total Fermi surface does not remain constant, a point which we will return to later.) $k_F(0,0)$ and $m_F^*(0,0)$ will be denoted by k_F and m_F^*.

By either the Boltzmann transport equation[17] or the Landau theory of transport in Fermi liquids,[18–21] the quasiparticle-quasiparticle interaction leads to a term in the electrical resistivity of the general form

$$\rho' = \frac{K(k_BT)^2 m_F^{*4}}{e^2\hbar^7 k_F^3}\langle\{[4\pi e^2/K(2k_F)^2] \times[\epsilon_{\rm Coul}(q/2k_F)]^{-1} f_{\rm Coul}(\theta,\phi) + V_{\rm ph}(0,0) \times[\epsilon_{\rm ph}(q/2k_F)]^{-1} f_{\rm ph}(\theta,\phi)\}^2\rangle_{\rm av}, \quad (2)$$

where K is the dielectric constant. The angular functions involve angular coordinates of two initial and two final quasiparticle states on the Fermi surface, although we have not expressed this explicitly. The scattering function in curly brackets is appropriately averaged over the Fermi surface, and is nonzero for umklapp processes or for scattering events in which velocity is not conserved due to anisotropy of the electron and hole effective masses. $\epsilon_{\rm Coul}$ and $\epsilon_{\rm ph}$ express screening of the Coulomb and phonon-mediated ($V_{\rm ph}$) contributions to the particle-particle interaction. Angular dependence of the screening is included in $f_{\rm Coul}$ and $f_{\rm ph}$. q is a characteristic screening parameter of the screening function of whatever form.

The electron-electron interaction in semiconductors or semimetals with many-valley conduction or valence bands has been discussed in detail by Cohen.[22] In at least some cases, when screening of the intravalley electron-electron interaction is highly effective, the dominant contribution to the electron-electron interaction is an attractive intervalley term of the phonon-mediated Fröhlich-BCS type. This contribution is apparently sufficiently strong for superconductivity to occur in degenerate semiconductors such as GeTe and $SnTiO_3$.[23]

A crude estimate for the effectiveness of screening for the intravalley Coulomb scattering in bismuth can be obtained by considering the average interparticle spacing r_s. Taking a value of 100 for the dielectric constant,[24] an average effective mass $\sim 0.1 m_e$,[25] and a free-carrier density $\sim 5\times 10^{17}$ cm^{-3},[25] r_s is of order 10^{-1} times the Bohr radius a_B^* appropriate to K and m^*. (A more precise value is not justified by the assumptions involved.) Thus, the intravalley screening is approximately the same or more effective than in good metals, which do not achieve the $r_s \ll a_B^*$ high-density limit.[26] Screening of intravalley Coulomb interactions should be sufficiently effective so that intervalley phonon-mediated interactions become relatively important. (Screening of phonon-mediated intervalley transitions will be much less effective, since for bismuth the momentum exchange must be much greater than the maximum Fermi momentum.) The phonon-mediated interaction potential $V_{\rm ph}(0,0)$ does not depend on k_F or m_F^* for either acoustic or optical modes, in contrast to the Coulomb potential. (Angular variation of the phonon-mediated interaction is included in $f_{\rm ph}$.)

Although the relative magnitudes of phonon-mediated and Coulomb contributions to the particle-particle interaction may depend critically on $\epsilon_{\rm Coul}$ and $\epsilon_{\rm ph}$, the most rapid variation of ρ' with k_F and m_F^* arises from the explicit factors appearing in Eq. (2). If the interaction were exclusively Coulomb, ρ' would be proportional to m_F^{*4}/k_F^7, disregarding the dependence through screening. The effect of screening, when $r_s \ll a_B^*$, is roughly to decrease the exponents of m_F^* and k_F by a power less than unity. Since m_F^* and k_F change in the same direction with alloying, for electron-electron scattering these changes will at least partially cancel. The effects of screening can be included by changing the exponent of k_F to a value which almost certainly falls

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[26] D. Pines and P. Nozières, *Theory of Quantum Liquids* (W. A. Benjamin, Inc., New York, 1966).

in the range 6.5–7.0. If the phonon-mediated contribution were dominant in Eq. (2), ρ' would be approximately proportional to m_F^{*4}/k_F^3, with a weak additional dependence introduced by screening. The critical element in this discussion is an additional factor k_F^{-4} appearing when the Coulomb interaction is dominant. For a semimetal, we expect the electron-electron contribution to the resistivity to depend on k_F and m_F^* in some manner intermediate to the two extremes.

The relative contributions of electron-electron, electron-hole, and hole-hole scattering to the resistivity would be very difficult to assess, requiring a detailed evaluation of ϵ_{Coul}, ϵ_{ph}, f_{Coul}, and f_{ph}. The analysis by Cohen for even spherically symmetric Fermi-surface segments is complicated,[22] and we will not attempt the more difficult analysis required for bismuth. We will consider four special cases for ρ': (a) hole-hole scattering via a Coulomb interaction potential, (b) electron-electron scattering via a Coulomb interaction potential, (c) hole-hole scattering via a phonon-mediated interaction potential, and (d) electron-electron scattering via a phonon-mediated interaction potential. For either interaction, electron-hole scattering alone would yield a dependence of ρ' on k_F and m_F^* intermediate to electron-electron and hole-hole cases.

Values for m_{Fe}^*, k_{Fe}, and k_{Fh} (m_{Fh}^* is constant) as a function of alloying are determined from the data of Kao *et al.*,[12] of Brandt *et al.*,[15] and of Ellett *et al.*[16] Here, it is important to remember that for a nonparabolic band, the inertial mass appropriate to Eq. (2) is not the same as the cyclotron mass measured in De Haas–van Alphen experiments, for example. The constant shape of the electron surfaces as the number of electrons is decreased shows that the Lax ellipsoidal, nonparabolic model is appropriate. (The more complicated model required when this model is not descriptive yields electron Fermi-surface segments which change shape as the electron density changes.[27]) According to the Lax model, the inertial mass at energy E in the electron band is

$$m_e^*=m_e^*(E=0)(1+E/Eg)\,, \tag{3}$$

and the cyclotron mass is

$$m_c^*=m_c^*(E=0)(1+2E/Eg)\,. \tag{4}$$

In calculating the inertial mass we have taken the values 0.015 eV for the energy gap E_g in pure bismuth and 0.027 eV for the electron degeneracy energy in pure bismuth.[25] No further assumptions are required, e.g., constant energy gap with alloying.

The behavior of ρ' with increasing antimony concentration for the cases (a)–(d) discussed above is shown in Fig. 2. In addition, the k_F^{-2} variation characteristic of electron-phonon scattering (assuming a momentum-independent mean free path) is displayed. Within the accuracy of the experimental results for B, m_{Fe}^*, k_{Fe}, m_{Fh}^*, and k_{Fh}, the BT^2 term in the resistivity is compatible with electron-phonon scattering (to be distinguished from electron-electron scattering, which is phonon-mediated). However, as we have discussed, this scattering mechanism does not lead to the required T^2 temperature dependence. Conceivably, a calculation of the electron-phonon scattering including details of both the electron and phonon spectra could yield the proper T^2 temperature dependence, but we regard this as an unlikely event. Another possible source for a T^2 term in the electrical resistivity would be the second term in the temperature expansion of the transport integral.[28] For a scattering mechanism yielding a T^2 second term, the first term must be constant in temperature, i.e., the residual resistivity. This explanation would require the BT^2 term to change with changes in the residual resistivity. The experimental results for bismuth show that the BT^2 term is independent of the residual resistivity, which rules out this mechanism.

If, in fact, an "electron-electron" resistivity term is a more reasonable explanation, Fig. 2 indicates that the phonon-mediated contribution to the interaction potential is at least comparable in magnitude to the Coulomb contribution when averaged over the Fermi surface. Relative magnitudes of the two interactions would depend on the contributions of electron-electron, electron-hole, and hole-hole scattering to ρ', but in any case the phonon-mediated interaction must be large.

With regard to the requirements for superconductivity discussed by Cohen,[22] it must be noted that the Fermi surface of bismuth is severely anisotropic, and in this case even a strong phonon-mediated contribution to the particle-particle interaction in a transport average may not be sufficient for superconductivity. We emphasize, if it is necessary, that results of our experiments do not constitute a proof that particle-particle interactions in the electron assembly are responsible for the T^2 term in the resistivity of bismuth. (There is, in fact, no incontestable proof that any of the T^2 resistivity terms discussed in the literature are electron-electron in origin.) In principle, an experiment to exploit isotope effects might be useful in resolving this question. Although either a T^2 electron-phonon or a phonon-mediated electron-electron resistivity term would be proportional to the reciprocal of the isotope mass, electron-electron scattering including a Coulomb contribution to the interaction would yield a term displaying this dependence only partially.

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[27] M. H. Cohen, Phys. Rev. **121**, 387 (1961).

[28] A. H. Wilson, *The Theory of Metals* (Cambridge University Press, New York, 1953), p. 13.