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Lattice Conductivity, Lorenz Numbers, and Nernst-Ettingshausen Effect in Tungsten at Liquid-Helium Temperatures*

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A further analysis of previously published measurements of the dc electrical and thermal transport coefficients of a tungsten crystal is presented. The coefficients were measured as functions of a strong magnetic field at several temperatures in the range of liquid He^4. Problems in separating the lattice thermal conductivity from the electronic thermal conductivity by a simultaneous study of the magnetoresistivities are discussed. The limiting conductivity obtained from such a study of the tungsten data is found to be in reasonable agreement with calculations based on the assumption of strongly coupled phonons scattered by nearly free electrons. From the same data, the Lorenz number is deduced as a function of temperature and found to be in excellent agreement with an electron-electron scattering formula given by Ziman. The transverse (Hall and Righi-Leduc) conductivities are used to deduce a transverse Lorenz number which displays an unexpected temperature dependence that is not explained. An apparent phonon drag effect, very similar to that found by Long *et al.* in the transverse (Nernst-Ettingshausen) thermoelectric coefficient of antimony, is found in the tungsten data for the same coefficient, and is quantitatively explained by a simple model. An electronic specific-heat coefficient is also deduced from the Nernst-Ettingshausen data, and is found to have a value intermediate to the results of augmented-plane-wave (APW) and relativistic augmented-plane-wave (RAPW) Fermi surface calculations.

I. INTRODUCTION

A recent paper[1] presented the results of an experimental determination of six dc electrical and thermal transport coefficients in a tungsten crystal at liquid-helium temperatures in a strong magnetic field. In that paper, the behavior of each kinetic coefficient was discussed independently of the other kinetic coefficients. In this paper, relationships between several of the coefficients are examined. It is only by studying these relationships that the data take on very much relevance. In Secs. II and III, the simultaneous behavior of the electrical and thermal magnetoconductivity tensors is studied, the object in Sec. II being to obtain the lattice conductivity, and in Sec. III, the Lorenz numbers of the lon-

gitudinal and transverse coefficients. Throughout the paper, the kinetic thermal magnetoconductivity $\vec{\lambda}''$ and the adiabatic conductivity $\vec{\lambda}$ are used synonymously, their difference having been found negligible.[1] In Sec. IV, the anomalous temperature dependence of the Nernst-Ettingshausen thermoelectric coefficient is discussed in terms of a phonon drag model proposed by Grenier,[2,3] and the zero-temperature extrapolation of the effect is used to determine an electronic density of states.

II. LATTICE CONDUCTION

The thermal conductivity λ of a metal is the sum $\lambda_g + \lambda_e$ of lattice and electronic terms. The lattice thermal conductivity λ_g of most conductors is quantitatively negligible compared to the electronic thermal conductivity λ_e.[4-6] If a reliable and reasonably precise measurement of λ_g can be made, it may, however, be fundamentally important. In metals at low temperatures, the phonon current will usually be limited by an intrinsic mechanism; the scattering of phonons by conduction electrons. In that case, λ_g measures approximately the same features of the electron-phonon interaction as those measured by that part (ρ_i in the Matthiessen approximation) of the electrical resistivity due to electron-phonon scattering.[6,7] In those metals for which the Bloch contribution to ρ_i is negligible at low temperatures, λ_g is the only direct transport measure of the normal-process coupling. This will be seen to be the case for tungsten.

A measurement of λ_g depends upon finding some method which will drastically reduce λ_e without substantial change of λ_g. The standard method used to determine λ_g of a metal in its nonsuperconducting state is that of alloying, usually by the addition of solute atoms of different valence, but approximately equal mass. This method obviously introduces uncertainties in one's knowledge of λ_g of the *pure crystal*, though the error *may* be small. The only method specifically applicable to a pure-crystalline normal-state conductor is the magnetic field method.[8]

A. Magnetic Field Method

The magnetic field method used in this work is based upon the relation[1]

$$\lambda_{11}(H, T) = TL_1(T)\sigma_{11}(H, T) + \lambda_g(T) , \qquad (1)$$

where λ_g and the Lorenz number L_1 must not depend upon the applied magnetic field H, and the thermal and electrical magnetoconductivities λ_{11} and σ_{11} must each decrease monotonically with the same dependence upon H. Equation (1) is illustrated by the tungsten data[1] (Fig. 1). If a material is a good normal conductor, very strong magnetoresistance effects are required in order to reduce the term $TL_1\sigma_{11}$ to a value comparable to that of λ_g. For this reason, one should not hope to determine λ_g at liquid-helium temperatures by the magnetic field method unless the conductor is an electronically compensated single crystal of very small residual resistivity, with the direction of the applied magnetic field chosen to eliminate saturation effects due to open orbits.

The magnetic field method was applied to a crystal of tungsten at the temperatures of liquid hydrogen (15–20 K) by de Haas and de Nobel,[9] and in subsequent work by de Nobel.[10,11] More recently, the lattice conductivity of tungsten at $T > 90$ K has also been studied by Williams and Fulkerson using alloying methods,[12] and at $T > 80$ K by van Witzenburg and Laubitz[13] using a magnetic field method.

Reviews of the work of de Nobel and other early workers have been given by Klemens[5] and Wilson.[14] Their prognosis for the magnetic field method is not optimistic. The de Nobel experiment and the Sondheimer-Wilson theory show that L_1 is not, in general, independent of H.[15]

The de Nobel[16] L_1 was clearly field dependent at the highest fields, while the straight lines (Fig. 1) found in this work indicate field independence, despite the fact that, owing to higher crystal purity, the *effective* fields used in this work were much higher than those used by de Nobel. At the highest laboratory field (36 kOe) applied by de Nobel, the ratio $\rho_{11}(H)/\rho(0)$ of the magneotresistivity to the zero-field resistivity reached a maximum value of $\simeq 1600$ at the lowest temperature, $\simeq 14$ K. At the highest laboratory field (22 kOe) applied in the pre-

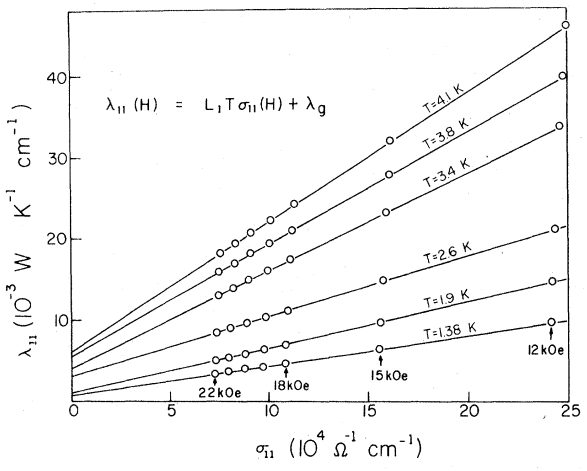

FIG. 1. Simultaneous (in field) high field behavior of the electrical magnetoconductivity σ_{11} and the thermal magnetoconductivity λ_{11}. The λ_{11} intercept at $\sigma_{11} = 0$ is interpreted as the lattice conductivity. Each straight line corresponds to a single temperature and a range of magnetic field strengths. Only selected points in a continuous field sweep are shown. Lorenz numbers are determined from the slopes of the lines. Currents and fields were along orthogonal ⟨100⟩ directions.

sent work, the ratio $\rho_{11}(H)/\rho(0)$ reached a value in excess of 76000 at a temperature of 1.3 K. It seems, therefore, that L_1 is field independent for the tungsten crystal at liquid-helium temperatures.

If the above conclusion is correct, the difference in this result and that of de Nobel is presumably due to the dominance of different and more simple scattering mechanisms at $T < 4.1$ K than in the range $14 < T < 20$ K available to de Nobel. Klemens[5] has given the condition $\gamma_0 > \gamma_i$ for field independence of L_1, where γ_0 is the thermal resistivity at zero field and temperature and γ_i is the part of the thermal resistivity due to intrinsic processes in an ideal crystal. (In the Matthiessen approximation $\lambda_g^{-1} = \gamma_0 + \gamma_i$.) Later in this paper it will be seen that the condition $\gamma_0 > \gamma_i$ is satisfied by the data of the present work. In de Nobel's work, at liquid-hydrogen temperatures, such was not the case. Furthermore, the criterion $\gamma_0 > \gamma_i$ was written in the context of a γ_i due to electron-lattice scattering, whereas it will be seen that γ_i in the present case was due to electron-electron scattering. At hydrogen temperatures, a significant remnant of the electron-phonon scattering should be present.[17]

An experiment is presently being planned which will test the field independence of L_1 in the same crystal at fields $\simeq 60$ kOe. The tentative conclusion supported by the analysis to follow, however, is that L_1 of tungsten at liquid-helium temperatures does satisfy the field-independence condition, and this experiment has, therefore measured the lattice thermal conductivity.

B. Possible Contributions to the Resistivity λ_g^{-1}

The values of λ_g determined through Eq. (1) and Fig. 1 are shown plotted logarithmically against temperature in Fig. 2. The solid line of Fig. 2 is that of the function $\lambda_g = 3.7 \times 10^3 T^2$ in units of erg $\text{sec}^{-1}\,\text{cm}^{-1}\,\text{K}^{-1}$, which is the best fit to the data when a T^2 relation is assumed. A quadratic dependence upon T is expected for two probable scattering mechanisms: the scattering of lattice waves by electrons and by dislocations.[4] High precision cannot be claimed. The coefficient 3.7 should only be considered reliable to $\pm 30\%$, assuming a quadratic T dependence is correct. The uncertainty in λ_g increases with decreasing temperature.

One could conceivably fit the λ_g values to a cubic T dependence, the dependence expected when the phonon current is limited by boundary scattering. It is, therefore, prudent to estimate the expected magnitude of a λ_g limited by boundary scattering. This is done by means of the kinetic relation $\lambda_g = \frac{1}{3} C_g U_s \Lambda$, where C_g is taken to be the Debye approximation to the lattice specific heat, U_s the velocity of sound, and Λ the mean width of the crystal.[5] The result is a λ_g^{-1} at 3 K of $\simeq 10^{-8}$ $\text{erg}^{-1}\,\text{cm sec K}$ as compared to an experimental value $> 3 \times 10^{-5}$ in the same units. It is safe to eliminate boundary scattering from further consideration.

The possibility that isotope scattering could appreciably limit the lattice conductivity should also be considered as a contribution to the gross temperature dependence, though the expected temperature dependence of T^{-1} is clearly not dominant. The most rapid estimate is again obtained from the kinetic relation, with the phonon mean free path taken as $0.054(2\pi)^{-4}\{a(\overline{A})^2/\overline{(\delta A)^2}\}(\Theta/T)^4$, where a is the lattice constant, Θ the Debye temperature, and $\overline{(\delta A)^2}$ is the variance of the atomic masses about the mean value $\overline{A}$.[18] Such a calculation yields approximately the same conductivity as that calculated for boundary scattering. It is thus concluded that the contribution of isotope scattering to λ_g^{-1} was negligible.

Finally, the scattering of lattice waves by dislocation lines must be considered. If the lattice conductivity were limited by dislocations, it should be approximated by the relation[19] $\lambda_g = (23k^3/h^2 U_s \mathfrak{N} b^2) T^2$, where k and h are the Boltzmann and Planck constants, $\mathfrak{N}$ is the average number of dislocation lines per unit area, and b is the magnitude of the Burgers vector. Taking b as one lattice constant and equating the preceding relation to the measured λ_g, it is found that a dislocation density of $\mathfrak{N} \simeq 10^{12}$ lines cm^{-2} would be required to limit λ_g to the observed value. This is at the extreme upper limit for heavily deformed metal crystals.[20] A smaller, more

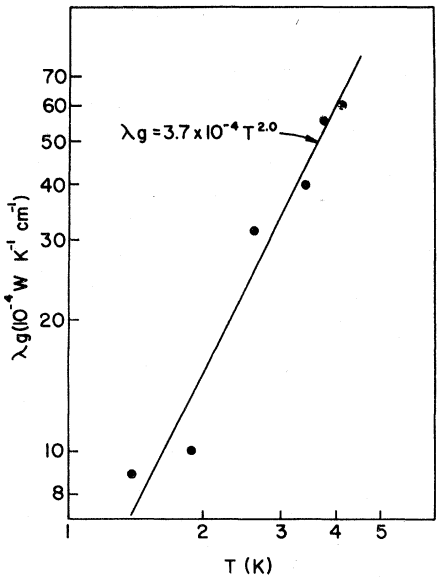

FIG. 2. Lattice conductivity λ_g logarithmically plotted against temperature T. The quadratic temperature dependence is characteristic of electron-phonon scattering at low temperatures.

realistic choice of b would increase $\mathfrak{N}$. It seems unreasonable, therefore, to expect that dislocations contribute in any appreciable way to λ_g^{-1}. The origin of λ_g^{-1} would thus seem to be entirely due to scattering by electrons.

C. Scattering by Electrons

The multiple, but singly connected, sheets of the Fermi surface of tungsten are highly distorted from the ideal spherical case.[21,22] The scattering of phonons by electrons should therefore be described by a model in which both longitudinal and transverse lattice waves interact directly with the electrons.[23] An expression for the conductivity of phonons in a Debye spectrum with all polarizations equally coupled to a parabolic band of electrons was derived by Makinson.[7,24,25] The Makinson formula may be written

$$\lambda_g = \frac{3\hbar k^3 MN}{\pi m^{*2} C^2} \mathcal{J}_3\left(\frac{\Theta}{T}\right) T^2, \tag{2}$$

where N is the density of atoms of mass M, m^* is the cyclotron effective mass of the electronic carriers, $\mathcal{J}_3(\Theta/T)$ is a Debye function, and C is the electron-phonon coupling constant of the order of the Fermi energy. It is customary to evaluate C in terms of some other transport property determined by the same interaction. Makinson[24] and Ziman[7] have expressed C in terms of $\lambda_{e\infty}$, the electronic component of the high-temperature thermal conductivity. Although it does not make a large error, this is not a good practice for tungsten, because the high-temperature thermal conductivity of tungsten is augmented by contributions from the lattice and the mechanism of ambipolar diffusion,[12,26] which produce an uncertain compensation to the U-process resistivity and other inadequacies of a high-temperature measure of C, as discussed by Klemens.[5] Klemens[5,27,28] has made an improved calculation of the electron-phonon scattering contribution to the partial resistivity λ_e^{-1}, and argues that the low-temperature limit of his result is a more appropriate measure of C for Eq. (2) at low temperatures. Although the Klemens argument is probably correct, it is only applicable when electron-phonon scattering makes a measurable contribution to λ_e^{-1}, and such, it will be seen, is not the case for tungsten at liquid-helium temperatures.

Volkenshteyn *et al.*[17] have found that the temperature dependence of the electrical resistivity of tungsten is described by a Debye-Grüneisen-Bloch function with $\Theta = 365$ K for temperatures from $T > 20$ K to room temperature, the maximum temperature of their measurements. An estimate of C in terms of the room-temperature electrical resistivity would seem, therefore, to be the least objectionable experimental approach to finding C. The Bloch formula[29] may be written

$$\rho_i = \frac{3\pi m^{*2} NC^2}{e^2 \hbar k M n^2 \Theta} \mathcal{J}_5\left(\frac{\Theta}{T}\right)\left(\frac{T}{\Theta}\right)^5, \tag{3}$$

the symbols of which have been previously defined, except n, the density of electronic carriers.

In applying Eqs. (2) and (3) to the data, the value $n = 14.9\times10^{21}$ cm^{-3} was used, as determined from the total Fermi-surface volume.[21,22] The resistivity of tungsten at some specific room temperature, say 300 K, is not available with great precision, most workers having avoided that problem by studying a resistivity ratio. Handbook values range from 5.65[30] to 5.4[31] $\mu\Omega$cm and a value $\rho(300) = 5.5$ $\mu\Omega$cm was found during the present study. The value of m^* which seems appropriate to Eqs. (2) and (3) is a weighted average $\overline{m^*}$ based upon an assignment of each measured cyclotron mass[22] for a specific magnetic field orientation to a certain fraction of n, this fraction having been determined from a six-band model[32] which gave reasonable agreement with the measured galvanomagnetic effects.[1,32] On that model $\overline{m^*} = \sum_i n_i m_i^*/n = 1.67 m_0$. Using $\Theta = 365$ K, and the remaining quantities as readily determined from the handbooks,[30,31] several combinations (Table I) of ρ_i, λ_g, and C were determined, assuming $T \ll \Theta$ in Eq. (2) and $T = 300$ K in Eq. (3).

The earlier contention that the lattice conductivity of the tungsten crystal was measured by the magnetic field method and that the conductivity is limited by the scattering of lattice waves by electrons is strongly supported by the results of Table I. Considering the stated precision in the measurement of λ_g, and the theoretical deficiencies of Eqs. (2) and (3) for application to a complex Fermi surface and nontrivial phonon spectrum, the factor-of-2 agreement shown in Table I can be regarded as very good. The 3% uncertainty in $\rho(300)$ is of little consequence.

An extrapolation of the experimental result $\lambda_g = 3.7\times10^{-4} T^2 (W\ \text{cm}^{-1}\ K^{-1})$ to higher temperatures on the basis of Eq. (2) yields $\lambda_g = 5\times10^{-5}\mathcal{J}_3(\Theta/T)\,T^2$. At $T = 100$ K this result predicts $\lambda_g = 2$ W cm^{-1} K^{-1}. Re-

TABLE I. Combinations of the ρ_i, λ_g, and C. Columns 1–3 show three reported values of the electrical resistivity ρ of tungsten at $T = 300$ K, the coupling constant C calculated from Eq. (3) for that ρ_i, and the value of the lattice conductivity λ_g calculated from Eq. (2) using the value of C derived from ρ_i. Column 4 shows the measured value of λ_g, the C calculated from Eq. (2) using the measured λ_g, and the ρ_i calculated from Eq. (3) using the value of C derived from λ_g.

	1	2	3	4
$\rho(\mu\Omega\text{cm})$	5.4	5.5	5.65	12.3
C(eV)	2.76	2.79	2.82	4.16
$\lambda_g(10^3$ erg cm^{-1} sec^{-1}K$^{-1})$	$8.4T^2$	$8.2T^2$	$8.0T^2$	$3.7T^2$

cent experiments[12,13] at $T = 100$ K found values of λ_g in the range $0.4 < \lambda_g < 0.9$ in the same units. Electron-phonon scattering thus appears to contribute some 20–45% to the resistivity λ_g^{-1} at $T = 100$ K.

The most interesting feature of these results is that the λ_g predicted by $\rho(300)$ is too large. A supposed advantage in having chosen $\rho(300)$ instead of $\lambda_{g\infty}$ for an evaluation of C is that the direction of the error should be certain. Unless the U-process contribution to $\rho(300)$ is removed, Eq. (3) should overestimate C and, hence, underestimate λ_g.

The discrepancy can be removed by the unlikely[23] assumption that only the longitudinal lattice waves are directly coupled to the electrons, the transverse waves being indirectly coupled through the 3-phonon N process.[33] The lattice conductivity would then be reduced by a factor of $\frac{1}{3}$ relative to the electrical resistivity,[5] and $\rho(300) = 5.5$ $\mu\Omega$cm would predict a temperature coefficient of 2.7 for λ_g compared to the experimental value 3.7 ± 1.1.

A more appealing explanation is a possible variation of C as the temperature decreases due to the greater importance of small q phonons at low temperatures. The coupling constant should vary inversely as q.[23]

The choices of m^* and Θ bear some further elaboration in relation to the previous discussion. Only C is affected by the ratio m^{*2}/M. The comparison of ρ_i and λ_g is independent of m^*. For Θ, however, it should be noted that, although the aggregate Fermi surface of tungsten contains a number of electrons per atom $n/N \simeq \frac{1}{4}$, the individual sheets have $n_i/N < \frac{1}{4}$. With exclusively intrasheet scattering, as is implicit in Eq. (3), the Θ_R effective in the transport effects should be some average $\overline{\Theta^*}$ of effective Θ_i^* values, where $\Theta_i^* = (4n_i/N)^{1/3}\Theta_R$ when $n_i/N < \frac{1}{4}$, as shown by Sondheimer.[34] When the same naive average as was used to compute $\overline{m^*}$ is used, it is found that $\overline{\Theta^*} \simeq \frac{2}{3}\Theta_R$, where Θ_R should be $\geq \Theta_D$ of the specific heat. Above $T = 50$ K, Θ_D is essentially constant with a value $\simeq 315$ K.[35] The value of Θ_R should be $> \Theta_D$ if longitudinal phonons are more strongly coupled to electrons than are transverse phonons, with a maximum value $\Theta_R \simeq [3/(1 + 2U_T^2/U_L^2)]^{1/2}\Theta_D \simeq 1.4\Theta_D$, where U_T and U_L are the sound velocities for transverse and longitudinal waves as calculated from the elastic constants.[36] The maximum Θ to substitute in Eq. (3) would, therefore, be $\overline{\Theta^*} \simeq \frac{2}{3}(1.4)(315) = 294$ K. This would be in marked disagreement with other experiments[17,37] and would increase the discrepancies in the present case.

The problems in a correct evaluation and interpretation of λ_g of tungsten are apparently greater than shown by the comparative agreement in Table I, but the low temperature λ_g would seem to be superior to ρ_i as a test of an *ab initio* calculation of C by virtue of the fact that a choice of Θ is not required.

III. LORENZ NUMBERS

The generalizations of the Wiedemann-Frzna-Lorenz law to the electrical and thermal magnetoconductivity tensors $\hat{\sigma}$ and $\hat{\lambda}$, for a magnetic field normal to the current direction in an isotropic[1] conductor are given by

$$\lambda_{11} = L_1 T\sigma_{11} + \lambda_g , \tag{4}$$

$$\lambda_{12} = L_2 T\sigma_{12} , \tag{5}$$

where λ_{12} and σ_{12} are the Righi-Leduc and Hall conductivities.[1] In general, the longitudinal Lorenz number L_1 and the transverse number L_2 are functions of H and T. The determination of λ_g by the magnetic field method was based upon Eq. (1), a special form of Eq. (4) for the case where H is large, and L_1 is a function of T only. In the phenomenological interpretation of the Sondheimer-Wilson theory,[38] it is easily shown[2] that L_1 and L_2 should be independent of H in the limits of very small or very large H. Let τ_σ^{-1} be the phenomenological frequency of the processes that limit the electrical conductivity, and let τ_λ^{-1} be the phenomenological frequency of the processes that limit the electronic contribution to the thermal conductivity. In the weak field limit, we have

$$\lim_{H\to 0} L_1 \equiv L_{10} = (\tau_\lambda/\tau_\sigma)L_0 , \tag{6}$$

$$\lim_{H\to 0} L_2 \equiv L_{20} = (\tau_\lambda/\tau_\sigma)^2 L_0 , \tag{7}$$

and in the strong-field[1] for, as calrified by Herring,[39] zero scattering) limit,

$$\lim_{T\to\infty} L_1 \equiv L_{1\infty} = (\tau_\sigma/\tau_\lambda)L_0 , \tag{8}$$

$$\lim_{T\to\infty} L_2 \equiv L_{2\infty} = L_0 , \tag{9}$$

where L_0 is the Sommerfeld number $\pi^2k^2/3e^2 = 2.45 \times 10^{-8}$ V^2 K^{-2}. In general, the τ's are T dependent and unequal. In the zero-T limit of a real crystal they are independent of T and equal. When $T > \Theta$ they are T dependent, but may be equal.

A. Longitudinal Number L_1

The straight lines of Fig. 1 and the successful interpretation of the $\sigma_{11} = 0$ intercept as λ_g support the validity of Eq. (4) in the form of Eq. (1) and a description of L_1 in terms of Eq. (8). The T dependence of L_1 determined from the slopes L_1T of Fig. 1 is shown in Fig. 3. As predicted by Eq. (8), the values of $L_1 = L_{1\infty} \to L_0$ asymptotically as $T \to 0$, within experimental error.

A good fit to the results is obtained by the function $(L_{1\infty}/L_0 - 1) = 3.4 \times 10^{-2}T^2$ (Fig. 4). A useful analysis is provided by assuming the additivity of reciprocal relaxation times:

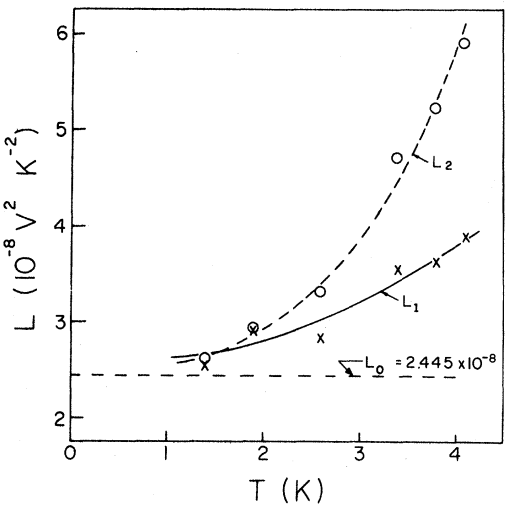

FIG. 3. Lorenz numbers $L_1=(\lambda_{11}-\lambda_g)/T\sigma_{11}$ and $L_2=\lambda_{12}/T\sigma_{12}$. Electron-electron scattering is responsible for the T dependence of L_1, but L_2 was expected to be independent of temperature with the free-electron value L_0 approached asymptotically by both L_1 and L_2. The T dependence of L_2 may be a drag effect.

$$\tau_\sigma^{-1}=\tau_{\sigma 0}^{-1}+\tau_{\sigma t}^{-1}\,, \tag{10}$$

$$\tau_\lambda^{-1}=\tau_{\lambda 0}^{-1}+\tau_{\lambda t}^{-1}\,. \tag{11}$$

The frequencies of temperature-dependent processes are denoted $\tau_{\sigma t}^{-1}$ and $\tau_{\lambda t}^{-1}$, while the frequencies of those processes which persist to zero temperature are denoted $\tau_{\sigma 0}^{-1}$ and $\tau_{\lambda 0}^{-1}$. The additivity of scattering rates is not necessarily equivalent to the Matthiessen approximation.[40] If, as in the present case, the values of L_1 and L_2 are obtained from total (all parts of the electron distribution) conductivities, then the scattering rates related to L_1 and L_2 by Eqs. (6)–(9) are rates averaged over the total Fermi surface. Use of the average rates obtained from Eqs. (6)–(9) in Eqs. (10) and (11) thus reduces the latter equations to statements of Matthiessen's rule.

The tendency $L_{1\infty}\to L_0$ as $T\to 0$ (Fig. 3) implies $\tau_{\sigma 0}=\tau_{\lambda 0}\equiv\tau_0$, and the approximate temperature independence[1] of the resistivity ρ implies $\tau_0\ll\tau_{\lambda t}$. These conditions with Eqs. (8), (10), and (11) yield the result

$$(L_{1\infty}/L_0-1)\simeq\tau_0/\tau_{\lambda t}\simeq 3.4\times10^{-2}\,T^2. \tag{12}$$

The result suggests electron-electron scattering. Consider the Matthiessen approximations $\rho=\rho_0+\rho_i$ and $\gamma=\gamma_0+\gamma_i$ at zero magnetic field. By Eqs. (4) and (6) with ρ_i and λ_g negligible, $\rho_0\simeq L_{10}T(\gamma_0+\gamma_i)$. But, $\rho_0=L_0T\gamma_0$, and by Eqs. (6) and (8) $L_0/L_{10}=L_{1\infty}/L_0$. Therefore, we have

$$(L_{1\infty}/L_0-1)\simeq\gamma_i/\gamma_0=L_0T\gamma_i/\rho_0. \tag{13}$$

If γ_i is a consequence of electron-electron scattering, a simple expression given by Ziman[41] might be expected to approximate γ_i in Eq. (13). In terms of the notation of Eqs. (2) and (3), the Ziman expression and Eq. (13) combine to give

$$\frac{L_{1\infty}}{L_0}-1\simeq\frac{ek^2m^{*3}}{30\hbar^6n^2}\left(\frac{\pi m^{*3}}{3n}\right)^{1/6}\frac{T^2}{\rho_0}\,. \tag{14}$$

If, as in the development of Table I, m^* is chosen to be the average cyclotron mass $\overline{m^*}=1.67\,m_0$, and using the measured[1,21] values $n=14.9\times10^{21}$ cm^{-3} and $\rho_0=1.8\times10^{-10}$ Ωcm, Eq. (14) predicts $(L_{1\infty}/L_0-1)\simeq14\times10^{-2}T^2$. Exact agreement with Eq. (12) is obtained when $m^*=1.1m_0$.

In view of the many assumptions leading to Eq. (14) and its evaluation, the quantitative agreement with experiment is remarkable, and perhaps fortuitous. The result would seem to establish electron-electron scattering as the source of the ideal thermal resistance in tungsten at helium temperatures, and the high-field Lorenz number as a powerful method for the study of the ideal resistance.

In the earlier paper[1] that presented the experimental transport coefficients which are the basis

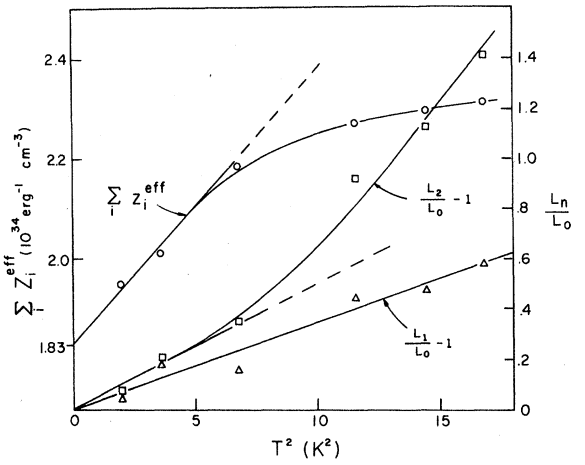

FIG. 4. Function (L_1/L_0-1) of the longitudinal Lorenz number L_1 at high field is found to be quadratic in temperature T. The result is a consequence of electron-electron scattering in the thermal resistivity. The density-of-states parameter $\sum_i Z_i$ effective in the kinetic Nernst-Ettingshausen coefficient is believed to be temperature dependent as a consequence of phonon drag, with the quadratic behavior of $\sum_i Z_i$ below 2.6 K indicative of normal-process scattering. The $T=0$ extrapolation of $\sum_i Z_i$ is the quasifree electron density of states. The function (L_2/L_0-1) of the transverse Lorenz number L_2 has a low-temperature behavior similar to that of $\sum_i Z_i$. This may be a consequence of the same drag mechanism, as both effects are independent of T in the standard theory.

for this paper, there was a discussion of the small temperature dependence of the electrical magnetoresistivity, which expressed some doubt as to the dominance of electron-electron scattering in the ideal electrical resistivity of tungsten determined by others.[17,37] There now seems to be no significant basis for that doubt.

B. Transverse Number L_2

The transverse Lorenz number L_2 is defined by Eq. (5) and is determined from the slopes L_2T of the plots shown in Fig. 5. In a sufficiently strong field, L_2 is expected to be independent of both H and T [Eq. (9)]. The straight lines (Fig. 5) show that $L_{2\infty}$ is independent of field over the range of the data, but that $L_{2\infty}$ is not independent of temperature. In Fig. 3, $L_{2\infty}$ is shown to have a T dependence similar to, but stronger than that displayed by $L_{1\infty}$. Very few studies of $L_{2\infty}$ have been made, and there do not appear to be any other reported observations of a T-dependent $L_{2\infty}$ in the literature. However suspect one may view the result, it should establish the danger in attempting to make a direct substitution of the Righi-Leduc effect for the Hall effect.[42]

The strong-field transverse transport coefficients have been shown[39,43] to be independent of scattering, and the result $L_{2\infty}=L_0$, Eq. (9), is more general than the Sondheimer-Wilson theory. The proofs

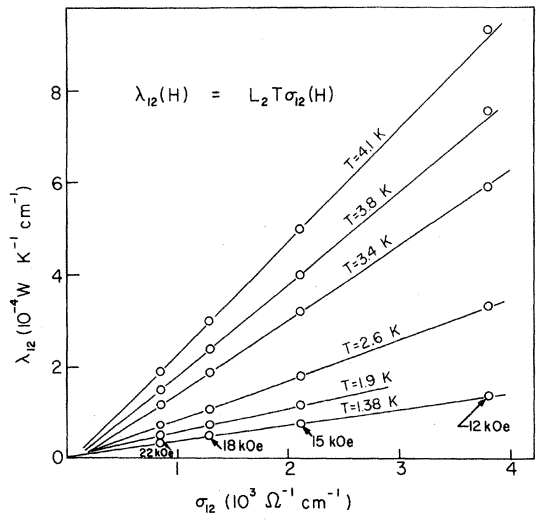

FIG. 5. Simultaneous (in field) high-field behavior of the Hall conductivity σ_{12} and the Righi-Leduc conductivity λ_{12}. Each straight line corresponds to a single temperature and a range of magnetic field strengths, as indicated. Only selected points in a continuous field sweep are shown. All lines should intersect at zero conductivity, as a consequence of the fact that the magnetic field cannot produce a significant direct effect upon the lattice. The scatter of the uncorrected intercepts is a good indicator of the precision in a plot of this type.

that $L_{2\infty}=L_0$ do not appear to account for drag effects between the electron and phonon distributions. When the $L_{2\infty}$ data are plotted in the form $(L_2/L_0 - 1)$ (Fig. 4), it bears a close resemblance at the lowest temperatures to the effective density of states computed from the kinetic Nernst-Ettingshausen effect (Fig. 4). As will be discussed in Sec. IV, the temperature dependence of the Nernst-Ettingshausen density of states is successfully explained by a phonon drag model. This prompted an attempt to describe the temperature dependence of $(L_2/L_0 - 1)$ as a phonon drag effect[44]; however, lengthy efforts to calculate the effect have failed to produce a nonzero result based upon reasonable premises. In order to get the desired result, it seems necessary that hot carriers be preferentially dragged by the lattice current. The Nernst-Ettingshausen enhancement is consistent with preferential drag of cold carriers, which seems more plausible.

Proofs that $L_{2\infty}=L_0$ seem to be mainly in the context of point defect and lattice scattering. Having established the importance of electron-electron scattering in Sec. II, it seems possible that the enhancement of $L_{2\infty}$ could be an electron-electron interaction effect.[45]

The possibility remains that the experimental result is false. The experiment must be repeated in the near future. It seems very unlikely, however, that any improvement in precision could be great enough to remove the temperature dependence altogether. All of the plots (Fig. 5) are quite linear and tend to intersect with little scatter at $\sigma_{12}=\lambda_{12}=0$, as predicted by Eq. (5). The error would therefore have to be a smoothly systematic positive error in the thermometer calibrations, increasing from nearly zero at the lowest temperature to a factor greater than 2 at the highest temperature. One should expect the precision of the thermometers to be less at higher temperatures, but the errors should be random and of the order of a few percent. Magnetoresistance-effect errors from sample and thermometers were removed by standard field-reversal averaging. The effect appears genuine.

IV. KINETIC NERNST-ETTINGSHAUSEN COEFFICIENT

In the strong-field limit of the Sondheimer-Wilson theory,[1,14] the kinetic Nernst-Ettingshausen coefficient ϵ''_{12} is given by

$$\epsilon''_{12}=-\frac{\pi^2 k^2 c T}{3H}\sum_i Z_i \ , \qquad (15)$$

where Z_i is the density of states in energy of the ith independent band of carriers. The $\sum_i Z_i$ should be related to the electronic specific-heat coefficient γ (not to be confused with the same symbol

used previously to denote thermal resistivity) by the expression $\sum_i Z_i = 3\gamma/\pi^2 k^2$. Values of γ calculated in this fashion were reported previously[1] and were observed to be anomalously temperature dependent, but within the rather wide range[35] of previously reported numbers.

If one treats the Sondheimer-Wilson Z_i as a phenomenological parameter, the temperature dependence of ϵ''_{12} is partially explained by a simple model of normal-process phonon drag[46] which was successfully adapted to the calculation of a very similar effect in antimony.[2,3] In this normal drag model, the electronic specific heat C_e is augmented by a dragged fraction β of the lattice specific heat C_g which causes the transport effect to measure $\gamma_{\text{trans}} = \gamma_{\text{equi}} + \gamma_{\text{drag}} = C_e T^{-1} + \beta C_g T^{-1}$. In the Debye approximation, we have

$$\gamma_{\text{drag}} = \frac{\beta 12\pi^4 N k}{5\Theta}\left(\frac{T}{\Theta}\right)^2 , \tag{16}$$

and γ_{equi} is equal, or nearly equal,[3] to the result of a measurement of an equilibrium property. The relative contributions of many-body effects to transport and specific-heat measurements of γ_{equi} is unknown. With $\Theta = 365$ K and $N = 6.35\times10^{22}$ cm^{-3}, Eq. (16) predicts $\gamma_{\text{drag}} \simeq 0.095\times10^{-4}\beta T^2$ (cal mole^{-1} K^{-2}), or a drag contribution to $\sum_i Z_i$ of $0.067\times10^{34}\beta T^2$ (erg^{-1} cm^{-3}). When the $\sum_i Z_i$ values computed from the measured ϵ''_{12} and Eq. (15) are plotted against T^2, the expected linear behavior is observed at temperatures below 2.6 K (Fig. 4). The linear portion is described by the function $0.055\times10^{34}T^2$ (erg^{-1} cm^{-3}). The drag model and data are thus in agreement for $\beta \simeq 0.8$. The most simple model predicts $\beta = \frac{1}{3}r$, where $r(0 < r < 1)$ is an efficiency parameter which measures the fraction of the free energy of the phonon system transferred to the electron system. If the gradient of the free energy (phonon gas pressure) is isotropic[3] (which it surely is not in tungsten), then $\beta = r/n$ for a lattice specific heat which varies as T^n. Therefore, $r = 2.4$, when $n = 3$ and $\beta = 0.8$. The most simple model, which has $0 < r < 1$, does not predict a sufficiently large β.

The zero-temperature intercept of the $\sum_i Z_i$ function is 1.83×10^{34} erg^{-1} cm^{-3}, which corresponds to a specific-heat coefficient $\gamma_{\text{equi}} = 2.6\times10^{-4}$ cal mole^{-1} K^{-2}. Reported[35,47] values of γ_{equi} as determined from equilibrium properties range from 1.8 to 10 of the units 10^{-4} cal mole^{-1} K^{-2}; the correct specific-heat value of γ_{equi}, however, is not likely[48] to be much larger than 2 units. Nonrelativistic augmented-plane-wave (APW) calculations[47,49] of the Fermi surface of tungsten predict $\gamma_{\text{equi}} \simeq 3$ units, while reduced exchange APW [49] and relativistic APW [50] calculations predict $\gamma_{\text{equi}} \simeq 1.7$ units. The APW results, however, do not account for many-body enhancements which are apparently present to some degree in any type of γ_{equi} measurement. The value $\gamma_{\text{equi}} \simeq 2.6$ units reported here thus looks rather good, but it would be unwise to place much weight on the exact value of the result, for it is a consequence of only three data points. The measurements must be extended to lower temperatures, because the γ_{drag} result should be regarded as equally suspect.

Above 2.6 K the $\sum_i Z_i$ curve, Fig. 4 departs from its linear dependence upon T^2 and appears to approach a constant value. Traces of the same behavior were observed for antimony.[2] This departure from Eq. (16), which is based upon a model[46] of normal-process drag, may be due to the onset of umklapp-process drag. If the change in T dependence with increasing T is a consequence of an increasing probability for umklapp processes, and if the phenomenological interpretation of Eq. (15) does not fail as inelastic processes become more dominant, one might expect to see $\sum_i Z_i$ or γ_{trans} pass through a maximum at slightly higher temperatures and eventually become smaller than γ_{equi}. A negative contribution to γ_{drag} in the Nernst-Ettingshausen effect would correspond to the positive drag contribution to the normally negative thermoelectric power of the alkali metals,[51] provided such positive contributions are drag effects.[52]

V. SUMMARY AND CONCLUSION

Many questions remain as to the validity of the magnetic field method as a measure of the lattice conductivity of a metal, but the method appears to be correct in tungsten at liquid-helium temperatures; although, it is apparently incorrect for tungsten at liquid-hydrogen temperatures[10] where a different scattering mechanism prevails. The lattice conductivity was found to be limited by electron-phonon normal-process scattering, but an electron-phonon coupling constant 50% larger than that expected from the room-temperature electrical resistivity was required to obtain a quantitative fit to the most plausible theory.[24] The difference was probably due to the nonspherical aspects of the tungsten Fermi surface.

Although the dominant scatterers of phonons were electrons, the dominant intrinsic scatterers of electrons were electrons. The ideal electrical resistivity is, therefore, not available as a measure of the electron-phonon coupling in tungsten at very low temperatures, thus enhancing the value of a lattice conductivity measurement.

The value of the Sondheimer-Wilson theory[14] as a phenomenological tool was illustrated by its application to the zero-field and high-field magnetoconductivity Lorenz numbers L_1. This resulted in a very successful explanation of the temperature dependence of the *high-field* number in terms of an

expression developed by Ziman[41] for the purpose of describing an electron-electron scattering contribution to the *zero-field* ideal thermal resistivity. No success was found, however, in attempting to explain the large, and completely unexpected, temperature dependence of the transverse effects Lorenz number L_2.

The apparently successful resolution of the Nernst-Ettingshausen effect into density of states and phonon drag contributions underlines[2,3] the value of the Nernst-Ettingshausen effect as a simultaneous, and no-less-precise, measure of two quantities which are traditionally measured by separate studies of the specific heat and the thermoelectric power.

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