

## Viscosity of the Electron Gas in Metals\*

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With a view toward explaining measurements of ultrasonic attenuation in metals, the coefficient of shear viscosity of a free electron gas interacting with thermal phonons and local crystal inhomogeneities is computed. The methods employed are essentially those developed for the problem of electrical conductivity. By means of a variational principle, a general formal solution is obtained. The effective mean free path is found to be somewhat smaller and of a somewhat more complicated (though not qualitatively different) temperature dependence than that associated with electrical conduction. The effect of a transverse magnetic field is determined for the case when a time of relaxation exists.

### 1. INTRODUCTION

MEASUREMENTS on lead and tin<sup>1,2</sup> indicate several seemingly general features of absorption of ultrasound in metals at the lowest temperatures: (a) Attenuation is exponential and is qualitatively alike for shear and compressional waves. (b) The frequency dependence of the attenuation coefficient appears to be quadratic at low frequencies. (c) From a flat maximum at absolute zero, the attenuation coefficient falls rapidly with increasing temperature. (d) When a sample is allowed to become superconducting, the attenuation falls off sharply (though apparently not discontinuously) as the temperature is lowered below the transition temperature. Mason has shown, by estimating the electronic mean free path from electrical conductivity data, that the static shear viscosity of the electron gas could account for the observed attenuation at very low temperatures.<sup>3</sup>

This paper will investigate the static shear viscosity of the electron gas in normal metals. The theory closely parallels that for the electrical conductivity (as elaborated, for example, in the account by Wilson<sup>4</sup>), the difference lying in the second singular spatial direction introduced when transport of momentum is considered. All the usual assumptions of the theory for the conduction problem will be retained; validity of Boltzmann's transport equation, perturbation treatment of the electron-phonon interaction, independent particle description of the electron gas, electronic energy an isotropic quadratic function of wave number, and Debye spectrum for the lattice vibrations. As no simultaneously realistic and tractable model of the polyvalent metals has as yet been proposed, the treatment will further be restricted to a single Brillouin zone.

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<sup>1</sup> H. E. Bömmel, Phys. Rev. **96**, 200 (1954).

<sup>2</sup> L. MacKinnon, Phys. Rev. **98**, 1181 (1955).

<sup>3</sup> W. P. Mason, Phys. Rev. **97**, 557 (1955).

<sup>4</sup> A. H. Wilson, *The Theory of Metals* (Cambridge University Press, New York, 1954).

### 2. FORMAL THEORY

If a conductor having  $n$  conduction electrons (effective mass  $m$ , charge  $e$ ) per unit volume is in shear motion with constant local velocity  $u$  in the  $x$  direction and uniform velocity gradient  $\beta = \partial u / \partial z$  in the  $z$  direction, the electrons will follow the lattice in the same state of steady flow and will experience no applied forces. The Boltzmann equation for the electron distribution function will be

$$-\frac{\hbar}{m} k_z \frac{\partial f}{\partial z} = \left( \frac{\partial f}{\partial t} \right)_{\text{coll}} \quad (2.1)$$

The equilibrium distribution is given by the Fermi function

$$f_0 = \left[ \exp \left( \frac{E - \zeta}{\kappa T} \right) + 1 \right]^{-1}, \quad (2.2)$$

where  $E = \hbar^2 k^2 / 2m$  is the energy of an electron with wave vector  $\mathbf{k}$  and velocity  $\mathbf{v} = \hbar \mathbf{k} / m$ , and the Fermi energy  $\zeta$  and velocity  $v_0$  are given by

$$\zeta = \frac{1}{2} m v_0^2 = \frac{\hbar^2}{2m} \left( \frac{3n}{8\pi} \right)^{\frac{2}{3}} \quad (2.3)$$

In a frame of reference moving with the local flow velocity,  $f_0(k)$  becomes  $f_0(|\mathbf{k} - m\mathbf{u}/\hbar|)$ . If the flow velocity is small ( $u \ll v_0$ ) the departure from thermal equilibrium will be small, i.e.,  $(f - f_0) \ll f_0$ . The Boltzmann equation may then be written

$$-\frac{\hbar^2 \beta}{m} k_x k_z \frac{\partial f_0}{\partial E} = \left( \frac{\partial f}{\partial t} \right)_{\text{coll}}, \quad (2.4)$$

and a solution found of the form

$$f = f_0 + k_x k_z S(E) \frac{\partial f_0}{\partial E} \quad (2.5)$$

The collision term may be put into the form

$$\left( \frac{\partial f}{\partial t} \right)_{\text{coll}} = -k_x k_z \mathcal{L}(S), \quad (2.6)$$

where  $\mathcal{L}$  is a linear operator, and the transport deter-

mined by solving

$$\mathcal{L}(S) = \frac{\hbar^2 \beta}{m} \frac{\partial f_0}{\partial E}, \quad (2.7)$$

for the nonequilibrium distribution function  $S$ .

The kinetic shear stress due to the velocity gradient of the electron flow is, in the observer's frame of reference,

$$T_{xz} = -\frac{2m^4}{\hbar^3} \int (v_x + u) v_z f(\mathbf{v}) d^3v. \quad (2.8)$$

Dividing this by the transverse flow velocity gradient gives the coefficient of shear viscosity of the electron gas:

$$\begin{aligned} \eta &= \frac{T_{xz}}{\beta} \approx -\frac{\hbar^2}{4\pi^2 m \beta} \int k_x k_z f(\mathbf{k}) d^3k \\ &= \frac{4(2m^5)^{\frac{1}{2}} \zeta^{\frac{3}{2}}}{15\pi^2 \hbar^5 \beta} S(\zeta). \end{aligned} \quad (2.9)$$

(Terms of order  $kT/\zeta$  are neglected.)

In the event the collision term is of the simple form

$$\left(\frac{\partial f}{\partial t}\right)_{\text{coll}} = -\left(\frac{f-f_0}{\tau}\right), \quad (2.10)$$

$\tau(E)$  is identified as the time of relaxation, and one has

$$S = \hbar^2 \beta \tau / m, \quad (2.11)$$

without further computation. Defining the electronic mean free path  $l \equiv v\tau$  and using (2.3), the coefficient is, in this case, given by the familiar formula

$$\eta = \frac{1}{5} n m v_0 l, \quad (2.12)$$

where  $l(\zeta)$  has been written simply as  $l$ . In general, however,  $\mathcal{L}$  is an integral operator, the collision term is not of the form (2.10), and one must solve the integral equation (2.7).<sup>5</sup>

The assumptions of steady flow and of uniform velocity gradient are not realized in practice when motion of the conductor is maintained by an acoustic wave. However, if the effective time of relaxation is much less than the period of the wave and if the effective mean free path is much less than the wavelength, then the static shear viscosity will be an accurate measure of the viscous attenuation. When the mean free path approaches the wavelength—a condition which can be met in very pure metals at low temperatures in the upper range of practicable frequencies—the electrons will see an alternating velocity gradient which disrupts the transport of momentum down the velocity gradient and brings about a relative decline in the magnitude of  $\eta$  with increasing frequency. The high-frequency complications will not be considered in the present paper.

<sup>5</sup> The viscosity of the thermal phonon gas may be treated in the same way. If the mean free path is the same as for heat conduction, one finds that  $\eta$  for the phonon gas is proportional to  $T$  times the lattice thermal conductivity.

### 3. COLLISION OPERATOR FOR THE LATTICE VIBRATIONS

The first-order transition probabilities (single phonon emission and absorption) for an electron interacting with the acoustic lattice vibrations may, given the deformation of the potential seen by the electron, be calculated by means of perturbation theory. If the interaction produces only a small departure from thermal equilibrium, the collision term is approximately linear in the nonequilibrium part of the distribution function. The result appropriate to the conduction problem is given by Wilson (reference 4, p. 260). For the viscosity problem, using the form (2.5) for the distribution function, the collision term is

$$\begin{aligned} \left(\frac{\partial f}{\partial t}\right)_{\text{coll}} &= \frac{C^2 \Delta}{8\pi^3 \hbar M \kappa T} \int \frac{q^2 N(\mathbf{q})}{\nu(\mathbf{q})} d^3q \\ &\times [k_x k_z S(E) - k_x' k_z' S(E')] \\ &\times \{f_0(k)[1-f_0(k')] \Omega(E-E'+\hbar\nu) \\ &+ f_0(k')[1-f_0(k)] \Omega(E-E'-\hbar\nu)\}. \end{aligned} \quad (3.1)$$

Here,  $C$  is the coupling constant measuring the strength of the interaction,  $E'$  is the energy, and

$$\mathbf{k}' = \mathbf{k} + \mathbf{q} \quad (3.2)$$

is the wave vector of an electron scattered by absorption of a phonon of wave vector  $\mathbf{q}$  and frequency  $\nu(\mathbf{q})$ .  $N(\mathbf{q})$  is the phonon distribution function, and

$$\Omega(x) \equiv (\hbar/x) \sin(xt/\hbar) \quad (3.3)$$

has the character of a  $\delta$  function for large  $t$ . The required constants are defined as follows:  $M$  = mass of lattice atom,  $n_a$  = number of atoms per unit volume,  $\Delta$  = volume per atom ( $\Delta = a^3$ ),  $v_s$  = velocity of sound,  $q_0$  = maximum thermal phonon wave number, and  $\Theta$  = Debye's characteristic temperature.

For isotropic metals, two integrations may be performed by introducing the angles  $\theta$  and  $\phi$  in the  $\mathbf{q}$  space, with  $\mathbf{k}$  the polar axis and  $\theta$  the angle between  $\mathbf{k}$  and  $\mathbf{q}$ . The required integrations over  $\phi$  are

$$\begin{aligned} \int_0^{2\pi} k_x q_x d\phi &= \int_0^{2\pi} k_z q_x d\phi = 2\pi k_x k_z (q/k) \cos\theta, \\ \int_0^{2\pi} q_x q_x d\phi &= \pi k_x k_z (q^2/k^2) (3 \cos^2\theta - 1). \end{aligned} \quad (3.4)$$

In the integration over  $\theta$ , the expansion  $E' - E = q \cos\theta \times (dE/dk) + \frac{1}{2} q^2 (d^2E/dk^2)$  and the singular property of the  $\Omega$  functions determine the slowly varying function of  $\theta$ :

$$\begin{aligned} 1 + \frac{2q}{k} \cos\theta + \frac{q^2}{2k^2} (3 \cos^2\theta - 1) \\ = 1 - \frac{3q^2}{2k^2} \pm \hbar\nu + \frac{3q^4}{8k^4} \mp \frac{3q^2}{4k^2} \left(\frac{\hbar\nu}{E}\right) + \frac{3}{8} \left(\frac{\hbar\nu}{E}\right)^2. \end{aligned} \quad (3.5)$$

Integration over  $\theta$  then produces a factor

$$\int_0^{2\pi} \Omega(E-E' \pm h\nu) \sin\theta d\theta = \frac{\pi(m/2)^{\frac{1}{2}}}{qE^{\frac{1}{2}}}. \quad (3.6)$$

The integration over  $q$  extends from  $q=0$  to  $q=q_0$  defined by

$$hqv_s = \kappa\Theta. \quad (3.7)$$

To cast this in a convenient form, put

$$w \equiv \frac{E-\zeta}{\kappa T}, \quad z \equiv \frac{h\nu}{\kappa T}, \quad (3.8)$$

and use the relations

$$f_0(E)[1-f_0(E+h\nu)] = -\kappa T \frac{\partial f_0}{\partial E} \left( \frac{e^w+1}{e^{w+z}+1} \right) e^z, \quad (3.9)$$

$$f_0(E-h\nu)[1-f_0(E)] = -\kappa T \frac{\partial f_0}{\partial E} \left( \frac{e^w+1}{e^{w-z}+1} \right).$$

If one assumes for the thermal phonons the elastic continuum spectrum

$$2\pi\nu(\mathbf{q}) = qv_s, \quad (3.10)$$

and the equilibrium (Planck) distribution<sup>6</sup>

$$N(\mathbf{q}) = (e^z - 1)^{-1}, \quad (3.11)$$

the collision term may now be written

$$\left( \frac{\partial f}{\partial t} \right)_{\text{coll}} = k_x k_z \frac{(m/2)^{\frac{1}{2}}}{\hbar^2 \Lambda E^{\frac{1}{2}}} \left( \frac{T}{\Theta} \right)^3 \frac{\partial f_0}{\partial E} \int_{-\Theta/T}^{\Theta/T} \left( \frac{e^w+1}{e^{w+z}+1} \right) \frac{z^2 dz}{|1-e^{-z}|}$$

$$\times \left\{ ES(w) - S(w+z) \left[ E + \kappa T z - 3 \left\{ D \left( \frac{T}{\Theta} \right)^2 - \frac{(\kappa T)^2}{8E} \right\} z^2 \right. \right.$$

$$\left. \left. - \frac{3D\kappa T}{2E} \left( \frac{T}{\Theta} \right)^2 z^3 + \frac{3D^2}{2E} \left( \frac{T}{\Theta} \right)^4 z^4 \right] \right\}, \quad (3.12)$$

with

$$\Lambda \equiv \left( \frac{4\pi}{3} \right)^{\frac{1}{2}} \frac{4M\alpha\kappa\Theta}{3\hbar^2 C^2}, \quad D \equiv \frac{(6\pi^2)^{\frac{3}{2}} \hbar^2}{4ma^2} = \frac{\zeta}{2^{\frac{1}{2}}(n/n_a)^{\frac{1}{2}}}.$$

This should be compared with the analogous result for the conduction problem (reference 4, p. 263).

The various terms in (3.12) may be ordered as follows:

- (1) For the significant energies,  $E \sim \zeta$ ,
- (2)  $D(T/\Theta)^2 z^2 \sim (D^2/E)(T/\Theta)^4 z^4 \sim \zeta$ ,
- (3)  $\kappa T z \sim (D\kappa T/E)(T/\Theta)^2 z^3 \sim \kappa\Theta \sim 10^{-3}\zeta$ ,
- (4)  $(\kappa T)^2 z/E \sim (\kappa\Theta)^2/\zeta \sim 10^{-6}\zeta$ .

<sup>6</sup> It appears that the long-standing objection to this procedure, originally raised by Peierls, has been clarified. For a self-consistent treatment of electron-phonon disequilibria, see I. I. Hanna and E. H. Sondheimer, Proc. Roy. Soc. (London) 239, 247 (1957).

With error of order  $10^{-3}$ , then, the collision operator for interaction with the acoustic lattice vibrations, as defined by Eq. (2.6), is given by

$$\mathcal{L}_i(S) = \frac{(m/2)^{\frac{1}{2}}}{\hbar^2 \Lambda E^{\frac{1}{2}}} \left( \frac{T}{\Theta} \right)^3 \frac{\partial f_0}{\partial E} \int_{-\Theta/T}^{\Theta/T} \left( \frac{e^w+1}{e^{w+z}+1} \right) \frac{z^2 dz}{|1-e^{-z}|}$$

$$\times \left\{ ES(w) - S(w+z) \left[ E - 3D \left( \frac{T}{\Theta} \right)^2 z^2 + \frac{3D^2}{2E} \left( \frac{T}{\Theta} \right)^4 z^4 \right] \right\}. \quad (3.13)$$

#### 4. "IDEAL" VISCOSITY

At sufficiently high temperatures ( $T \gg \Theta$ ) the integrand of the collision operator (3.13) may be expanded in powers of  $z$ . The electron-phonon interaction yields, in this limit,

$$\left( \frac{\partial f}{\partial t} \right)_{\text{coll}} = -k_x k_z S(E) \frac{\partial f_0}{\partial E} \frac{(m/2)^{\frac{1}{2}}}{\hbar^2 \Lambda E^{\frac{1}{2}}} \left( \frac{T}{\Theta} \right)^3$$

$$\times \int_0^{\Theta/T} 2 \left[ 3D \left( \frac{T}{\Theta} \right)^2 z^3 - \frac{3D^2}{2E} \left( \frac{T}{\Theta} \right)^4 z^5 \right] dz. \quad (4.1)$$

$\mathcal{L}_i$  is now a simple multiplicative operator and the collision term is of the form (2.10), with time of relaxation

$$\tau_i = \frac{2\hbar^2 \Lambda E^{\frac{1}{2}}}{D(m/2)^{\frac{1}{2}}} \left( \frac{\Theta}{T} \right) \left[ 3 - \frac{(\zeta/E)}{2^{\frac{1}{2}}(n/n_a)^{\frac{1}{2}}} \right]^{-1}, \quad (4.2)$$

mean free path  $l_i = v\tau_i$ , and "ideal" viscosity (due to scattering by the lattice vibrations alone) given by

$$\eta_i = \frac{1}{5} n m v_0 l_i. \quad (4.3)$$

( $l_i$  is evaluated at  $E = \zeta$ .)

The quantity  $\tau_i$  in (4.2) differs from that for electrical conduction by the factor in square brackets. For the significant energies ( $E \cong \zeta$ ) in true metals ( $n/n_a > \frac{1}{4}$ ), this factor has a value between one and three.  $\tau_i(\zeta)$  is thus always less for the viscosity than for the conductivity at high temperatures, by a factor of up to  $\frac{1}{3}$ . For semimetals, the limit of integration  $\Theta/T$  must be replaced by  $(2E/D)^{\frac{1}{2}}(\Theta/T)$ , whereupon  $\tau_i$  becomes identical with that for the conductivity (see reference 4, p. 264).

At intermediate and low temperatures,  $\mathcal{L}_i$  remains and integral operator and one must solve the integral equation (2.7). If only the electron-phonon interaction is considered, this is, when one uses (3.13),

$$E \frac{\partial f_0}{\partial E} \int_{-\Theta/T}^{\Theta/T} [S(w+z) - S(w)] \left( \frac{e^w+1}{e^{w+z}+1} \right) \frac{z^2 dz}{|1-e^{-z}|}$$

$$= - \frac{\hbar^4 \Lambda \beta}{(m^3/2)^{\frac{1}{2}}} \left( \frac{\Theta}{T} \right)^3 E^{\frac{1}{2}} \frac{\partial f_0}{\partial E}$$

$$+ \frac{\partial f_0}{\partial E} \int_{-\Theta/T}^{\Theta/T} S(w+z) \left[ 3D \left( \frac{T}{\Theta} \right)^2 z^2 - \frac{3D^2}{2E} \left( \frac{T}{\Theta} \right)^4 z^4 \right]$$

$$\times \left( \frac{e^w+1}{e^{w+z}+1} \right) \frac{z^2 dz}{|1-e^{-z}|}. \quad (4.4)$$

The homogeneous equation obtained by setting the right hand side of (4.4) equal to zero is solved by  $S(w) = \text{const}$ ; a solution of the inhomogeneous equation then exists only if the inhomogeneous part is orthogonal to a constant, which establishes the condition

$$\begin{aligned} \frac{\hbar^4 \Lambda \beta}{(m^3/2)^{1/2}} \left(\frac{\Theta}{T}\right)^3 \int_{-\infty}^{\infty} E^3 \frac{\partial f_0}{\partial w} dw &= \int_{-\infty}^{\infty} \frac{\partial f_0}{\partial w} dw \int_{-\Theta/T}^{\Theta/T} S(w+z) \\ &\times \left[ 3D \left(\frac{T}{\Theta}\right)^2 z^2 - \frac{3D^2}{2E} \left(\frac{T}{\Theta}\right)^4 z^4 \right] \\ &\times \left( \frac{e^w + 1}{e^{w+z} + 1} \right) \frac{z^2 dz}{|1 - e^{-z}|}. \end{aligned} \quad (4.5)$$

At sufficiently low temperatures ( $T \ll \Theta$ ) the integral term on the right-hand side of (4.4) may be treated as a perturbation. A solution of the form

$$S(w) = S_0 + S_1(w), \quad |S_0| = \text{const} \gg |S_1|, \quad (4.6)$$

may then be obtained in which  $S_0$  is determined by (4.5) and  $S_1$  is (in principle) obtainable from (4.4). The result is

$$\begin{aligned} S(w) \cong S_0 &= \frac{\hbar^4 \Lambda \zeta^{3/2} \beta}{3(2m^3)^{1/2} D} \left(\frac{\Theta}{T}\right)^5 \\ &\times \left[ J_5 \left(\frac{\Theta}{T}\right) - \frac{(T/\Theta)^2}{2^{4/3} (n/n_a)^{1/3}} J_7 \left(\frac{\Theta}{T}\right) \right]^{-1}, \end{aligned} \quad (4.7)$$

where

$$J_p(x) \equiv \int_0^x \frac{z^p dz}{(e^z - 1)(1 - e^{-z})}. \quad (4.8)$$

This procedure is due to Bloch. Its justification is discussed in the older literature on the conduction problem.<sup>7</sup> As the method is limited compared to the variable principle now available, it will not be considered at greater length here.

According to Eq. (2.9), the "ideal" viscosity is now given by (4.3) if an effective time of relaxation is formally defined by

$$\begin{aligned} \tau_i(\zeta) &= \frac{mS(\zeta)}{\hbar^2 \beta} = \frac{\hbar^2 \Lambda \zeta^{3/2}}{3(2m)^{1/2} D} \left(\frac{\Theta}{T}\right)^5 \\ &\times \left[ J_5 \left(\frac{\Theta}{T}\right) - \frac{(T/\Theta)^2}{2^{4/3} (n/n_a)^{1/3}} J_7 \left(\frac{\Theta}{T}\right) \right]^{-1}. \end{aligned} \quad (4.9)$$

Although the derivation does not permit such extension, (4.9) includes the high-temperature result (4.2), for  $J_p(x) \cong x^{p-1}/(p-1)$  when  $x \ll 1$ . In the neighborhood

<sup>7</sup> For example, in A. Sommerfeld and H. Bethe, *Handbuch der Physik* (Verlag Julius Springer, Berlin, 1933), Vol. 24, Part 2, p. 499.

of absolute zero,  $J_p(\Theta/T) \cong J_p(\infty) = \text{const}$ , and

$$\tau_i \cong \frac{\hbar^2 \Lambda \zeta^{3/2}}{3(2m)^{1/2} D J_5(\infty)} \left(\frac{\Theta}{T}\right)^5. \quad (4.10)$$

Here, as for  $T \gg \Theta$ , the temperature dependence is the same as for the conductivity, but the magnitude of  $\tau_i$  is exactly  $\frac{1}{3}$  that for the conductivity for all  $n/n_a > \frac{1}{4}$ . At finite temperatures, the term in  $J_7(\Theta/T)$  spoils somewhat the  $T^{-5}$  dependence (to the extent of about 5% at  $T/\Theta = 0.05$ ), the ideal viscosity falling more slowly than the ideal conductivity with increasing temperature.

### 5. "RESIDUAL" FLUIDITY

If the conduction electrons are scattered only by randomly located inhomogeneities of atomic dimensions, and if the scattering is elastic, the collision term of the Boltzmann equation may be written (compare reference 4, p. 268)

$$\begin{aligned} \left(\frac{\partial f}{\partial t}\right)_{\text{coll}} &= \frac{\Delta}{\pi \hbar^2} \int |B_{kk'}(\theta)|^2 k'^2 dk' \sin \theta d\theta d\phi \\ &\times (k_x' k_z' - k_x k_z) S(E) \Omega(E - E') \partial f_0 / \partial E. \end{aligned} \quad (5.1)$$

$B_{kk'}(\theta)$  is the matrix element for the scattering. The required integration over  $\phi$  is

$$\int_0^{2\pi} k_x' k_z' d\phi = 2\pi k_x k_z (1 - \frac{1}{2} \sin^2 \theta), \quad (5.2)$$

and that over  $k'$  is

$$\int_0^{\infty} \Omega(E - E') k'^2 dk' = \pi (2m^3)^{1/2} E^{3/2} \hbar^{-2}. \quad (5.3)$$

In this case, a time of relaxation  $\tau_r$  exists for all temperatures, given by

$$\frac{1}{\tau_r} = \frac{3(2m^3)^{1/2} E^{3/2} \Delta}{4\pi \hbar^4} \int_0^{\pi} |B_{kk'}(\theta)|^2 \sin^3 \theta d\theta. \quad (5.4)$$

A factor  $\frac{3}{2} \sin^2 \theta$  in the integrand replaces the corresponding factor  $(1 - \cos \theta)$  for the conduction problem. Since the scattering is probably mostly in the forward direction, and since  $\frac{3}{2} \sin^2 \theta > (1 - \cos \theta)$  for  $\theta$  less than about  $100^\circ$ , the time of relaxation and mean free path  $\tau = v \tau_r$  are most likely smaller for the viscosity problem. In the neighborhood of absolute zero, the ideal fluidity  $1/\eta_i$  vanishes as  $T^5$ , leaving a "residual" fluidity  $1/\eta_r$  given by

$$\eta_r = \frac{1}{3} n m v_0 \tau_r. \quad (5.5)$$

The collision operator for this residual scattering is

given by

$$\mathcal{L}_r(S) = \frac{E^{\frac{1}{2}}}{(m/2)^{\frac{1}{2}} v_r} S(E) \frac{\partial f_0}{\partial E} \quad (5.6)$$

It is independent of the temperature.

6. THE VARIATIONAL PRINCIPLE

The variational principle developed by Kohler for the conduction problem<sup>8</sup> is easily adapted to the viscosity problem. It permits rigorous solution (to any desired degree of accuracy) of the linearized transport equation for all collision mechanisms treated simultaneously, without any assumption as to the additivity of the associated partial fluidities. Its validity depends only upon quite general properties of the total collision operator.

In terms of the operator  $L$  defined by

$$\left(\frac{\partial f}{\partial t}\right)_{\text{coll}} = -\frac{\hbar^2 \beta}{m} L(F), \quad (6.1)$$

where  $F(\mathbf{k})$  is the nonequilibrium distribution function defined by

$$f = f_0 + F \partial f_0 / \partial E, \quad (6.2)$$

and where  $|F \partial f_0 / \partial E| \ll f_0$ , the Boltzmann equation (2.4) is

$$L(F) = k_x k_x \partial f_0 / \partial E. \quad (6.3)$$

For any two functions  $G(\mathbf{k})$  and  $H(\mathbf{k})$  define

$$(G, H) \equiv \int GL(H) d^3k. \quad (6.4)$$

The variational principle is now stated as follows: If  $F$  is the solution of (6.3), then of all functions  $G$  satisfying the condition

$$(G, G) = \int G k_x k_x \frac{\partial f_0}{\partial E} d^3k, \quad (6.5)$$

$F$  is the one which makes  $(G, G)$ , as given by (6.4), a maximum.

The proof follows the same steps as for the conduction problem (reference 4, p. 301). The relations  $(F, G) = (G, F)$  and  $(G, G) \geq 0$  result, respectively, from the symmetry of the transition probability between incident and scattered electronic wave vectors and from the essentially positive character of the transition probability. Equation (6.3) and the condition (6.5) also imply that  $(G, F) = (G, G)$ . From these three relations it follows that  $(G, G) \leq (F, F)$ , thus proving the maximum principle.

For the case of isotropic metals, the Boltzmann equation is

$$\mathcal{L}(\sigma) = \frac{\partial f_0}{\partial E} = \frac{1}{\kappa T} \frac{\partial f_0}{\partial w}, \quad (6.6)$$

where

$$\sigma(E) \equiv (m/\hbar^2 \beta) S(E). \quad (6.7)$$

Now,

$$F(\mathbf{k}) = (\hbar^2 \beta / m) k_x k_x \sigma(E), \quad (6.8)$$

and

$$L(F) = k_x k_x \mathcal{L}(\sigma). \quad (6.9)$$

The integration over angles may be performed directly, so that  $(F, F)$  is proportional to  $\int_0^\infty E^{\frac{1}{2}} \sigma \mathcal{L}(\sigma) dE$  or, for a degenerate electron gas, to  $\kappa T \int_{-\infty}^\infty E^{\frac{1}{2}} \sigma \mathcal{L}(\sigma) dw$ . The solution to (6.6) is therefore determined by maximizing

$$(\sigma, \sigma) \equiv \kappa T \int_{-\infty}^\infty E^{\frac{1}{2}} \sigma \mathcal{L}(\sigma) dw, \quad (6.10)$$

subject to the condition

$$(\sigma, \sigma) = \int_{-\infty}^\infty E^{\frac{1}{2}} \sigma \frac{\partial f_0}{\partial w} dw. \quad (6.11)$$

7. GENERAL SOLUTION

Let

$$\sigma(w) = \sum_{r=0}^\infty \sigma_r w^r, \quad (7.1)$$

$$d_{rs} \equiv -\kappa T \int_{-\infty}^\infty E^{\frac{1}{2}} w^r \mathcal{L}(w^s) dw, \quad (7.2)$$

$$\alpha_r \equiv - \int_{-\infty}^\infty E^{\frac{1}{2}} w^r \frac{\partial f_0}{\partial w} dw. \quad (7.3)$$

Then the variational integral (6.10) is

$$(\sigma, \sigma) = \sum_r \sum_s d_{rs} \sigma_r \sigma_s, \quad (7.4)$$

and the condition that this be a maximum subject to condition (6.11) requires that the expansion coefficients  $\sigma_r$  satisfy

$$\sum_s d_{rs} \sigma_s - \alpha_r = 0. \quad (7.5)$$

The formal solution is now, from (2.9),

$$\eta = -\frac{4(2m^3)^{\frac{1}{2}}}{15\pi^2 \hbar^3} \int_{-\infty}^\infty E^{\frac{1}{2}} \sigma(w) \frac{\partial f_0}{\partial w} dw = \frac{4(2m^3)^{\frac{1}{2}}}{15\pi^2 \hbar^3} \sum_r \alpha_r \sigma_r = \frac{4(2m^3)^{\frac{1}{2}}}{15\pi^2 \hbar^3} \frac{\mathfrak{D}_{\alpha\alpha}}{\mathfrak{D}}. \quad (7.6)$$

Here,

$$\mathfrak{D} \equiv \begin{vmatrix} d_{00} & d_{01} & \cdots \\ d_{10} & d_{11} & \\ \vdots & & \end{vmatrix} \quad \text{and} \quad \mathfrak{D}_{\alpha\alpha} \equiv \begin{vmatrix} 0 & \alpha_0 & \alpha_1 & \cdots \\ \alpha_0 & d_{00} & d_{01} & \\ \alpha_1 & d_{10} & d_{11} & \\ \vdots & & & \end{vmatrix} \quad (7.7)$$

are infinite determinants which are in general not convergent except in ratio.

<sup>8</sup> M. Kohler, Z. Physik 124, 772 (1948); 125, 697 (1949).

Now, define the finite determinants

$$\mathfrak{D}^{(n)} \equiv \begin{vmatrix} d_{00} & d_{01} & \cdots & d_{0, n-1} \\ d_{10} & d_{11} & & \vdots \\ \vdots & & & \\ d_{n-1, 0} & \cdots & & d_{n-1, n-1} \end{vmatrix},$$

$$\mathfrak{D}_{\alpha\alpha}^{(n)} \equiv \begin{vmatrix} d_{00} & d_{01} & \cdots & d_{0, n-1} & \alpha_0 \\ d_{10} & d_{11} & & \vdots & \vdots \\ \vdots & & & & \\ d_{n-1, 0} & \cdots & & d_{n-1, n-1} & \alpha_{n-1} \\ \alpha_0 & \cdots & & \alpha_{n-1} & 0 \end{vmatrix}, \quad (7.8)$$

$$\mathfrak{D}_{\alpha}^{(n-1)} \equiv \begin{vmatrix} d_{00} & d_{01} & \cdots & d_{0, n-2} & \alpha_0 \\ d_{10} & d_{11} & & \vdots & \vdots \\ \vdots & & & & \\ d_{n-1, 0} & \cdots & & d_{n-1, n-2} & \alpha_{n-1} \end{vmatrix},$$

$$\mathfrak{D}_{\alpha\alpha}^{(n)'} \equiv |\text{Minors of } \mathfrak{D}_{\alpha\alpha}^{(n)}|.$$

According to a theorem of Sylvester, if  $M$  and  $M'$  are corresponding  $m$ -rowed minors of any determinant  $D$  and its adjoint  $D'$ , respectively, then  $M'$  is equal to  $D^{m-1}$  times the algebraic complement of  $M$ . Let  $M'$  be the 2-rowed minor formed by the 4 elements at the bottom right hand corner of  $\mathfrak{D}_{\alpha\alpha}^{(n)'}$ . Then  $D^{m-1} = D = \mathfrak{D}^{(n)}$ , the algebraic complement of  $M$  is  $\mathfrak{D}^{(n-1)}$ , and the theorem yields the relation

$$\begin{vmatrix} \mathfrak{D}_{\alpha\alpha}^{(n-1)} & \mathfrak{D}_{\alpha}^{(n-1)} \\ \mathfrak{D}_{\alpha}^{(n-1)} & \mathfrak{D}^{(n)} \end{vmatrix} = \mathfrak{D}_{\alpha\alpha}^{(n)} \mathfrak{D}^{(n-1)}. \quad (7.9)$$

Expanding (7.9), summing over  $n$ , and using one obtains

$$\mathfrak{D}^{(1)} = d_{00}, \quad \mathfrak{D}_{\alpha\alpha}^{(1)} = -\alpha_0^2, \quad (7.10)$$

$$\frac{\mathfrak{D}_{\alpha\alpha}}{\mathfrak{D}} = \lim_{n \rightarrow \infty} \frac{\mathfrak{D}_{\alpha\alpha}^{(n)}}{\mathfrak{D}^{(n)}} = \frac{\alpha_0^2}{d_{00}} + \sum_{n=2}^{\infty} \frac{[\mathfrak{D}_{\alpha}^{(n-1)}]^2}{\mathfrak{D}^{(n-1)} \mathfrak{D}^{(n)}}. \quad (7.11)$$

The coefficient of viscosity is, therefore

$$\eta = \eta_0 + \frac{4(2m^3)^{\frac{1}{2}}}{15\pi^2 \hbar^3} \sum_{n=2}^{\infty} \frac{[\mathfrak{D}_{\alpha}^{(n-1)}]^2}{\mathfrak{D}^{(n-1)} \mathfrak{D}^{(n)}}, \quad (7.12)$$

with the first approximation

$$\eta_0 = \frac{4(2m^3)^{\frac{1}{2}}}{15\pi^2 \hbar^2} \left( \frac{\alpha_0^2}{d_{00}} \right). \quad (7.13)$$

When the collision operator  $\mathcal{L}$  incorporates terms due to collisions with both phonons [Eq. (3.13)] and inhomogeneities [Eq. (5.6)], the required matrix elements for a degenerate electron gas are

$$\alpha_0 = \xi^{\frac{1}{2}},$$

$$d_{00} = -\kappa T \int_{-\infty}^{\infty} E^{\frac{1}{2}} [\mathcal{L}_i(1) + \mathcal{L}_r(1)] dw$$

$$= \frac{3(2m)^{\frac{1}{2}} D \xi^{\frac{1}{2}}}{\hbar^2 \Lambda} \left( \frac{T}{\Theta} \right)^5 \left[ J_5 \left( \frac{\Theta}{T} \right) - \frac{(T/\Theta)^2}{2^{4/3} (n/n_a)^{\frac{1}{3}}} J_7 \left( \frac{\Theta}{T} \right) \right]$$

$$+ \frac{2\xi^{\frac{1}{2}}}{(m/2)^{\frac{1}{2}} \nu_r}. \quad (7.14)$$

Inserting these in (7.13) and making use of (2.3) yields, for the first approximation,

$$\frac{1}{\eta_0} = \frac{1}{\eta_i} + \frac{1}{\eta_r},$$

that is, additivity of the partial fluidities [Eqs. (4.3), (4.9), and (5.5)]. This is the analog of Matheson's rule for the electrical conductivity; it does not hold in higher approximation.

Calculation of successive approximations is straightforward. On the basis of Sondheimer's results for the electrical conductivity,<sup>9</sup> corrections to  $\eta_0$  may be expected to be quite small. Extension to semimetals and to nonpolar semiconductors involves only minor alterations in  $\mathcal{L}$ . For polar semiconductors, where scattering of the conduction electrons by the optical lattice vibrations must be considered, extension of the theory could be accomplished along the lines established by Howarth and Sondheimer for the conduction problem.<sup>10</sup> The validity of the useful results depends, of course, upon the convergence of the sum in (7.11) and upon the convergence of  $\lim_{n \rightarrow \infty} (\mathfrak{D}_{\alpha\alpha}^{(n)}/\mathfrak{D}^{(n)})$  to  $\mathfrak{D}_{\alpha\alpha}/\mathfrak{D}$ .

8. TRANSVERSE MAGNETO-VISCOSITY

When a magnetic field (0,  $H$ , 0) exists in a metal, the Boltzmann equation for the electron gas is

$$-\frac{\hbar}{m} k_z \frac{\partial f}{\partial z} + \omega_c \left( k_x \frac{\partial f}{\partial k_x} - k_z \frac{\partial f}{\partial k_z} \right) = \left( \frac{\partial f}{\partial t} \right)_{\text{coll}}, \quad (8.1)$$

where

$$\omega_c = eH/mc \quad (8.2)$$

is the cyclotron frequency of the electrons. Since the operator  $(k_x \partial/\partial k_x - k_z \partial/\partial k_z)$  transforms  $k_x k_z$  into  $(k_z^2 - k_x^2)$  and vice versa, the appropriate form for the distribution function in an infinite metal is

$$f = f_0 + [k_x k_z S_1(E) + (k_z^2 - k_x^2) S_2(E)] \frac{\partial f_0}{\partial E}. \quad (8.3)$$

Assume the existence of a time of relaxation, and small departure from thermal equilibrium. When one neglects small terms, but retains the terms in  $H$ , the Boltzmann equation reduces to

$$(\hbar^2 \beta/m) k_x k_z - \omega_c [4k_x k_z S_2 - (k_z^2 - k_x^2) S_1] = (1/\tau) [k_x k_z S_1 + (k_z^2 - k_x^2) S_2]. \quad (8.4)$$

Equating coefficients of  $k_x k_z$  and  $(k_z^2 - k_x^2)$  yields

$$(S_1/\tau) + 4\omega_c S_2 = \hbar^2 \beta/m, \quad -\omega_c S_1 + (S_2/\tau) = 0, \quad (8.5)$$

with the solution

$$S_1 = (\hbar^2 \tau \beta/m) [1 + 4\omega_c^2 \tau^2]^{-1},$$

$$S_2 = (\hbar^2 \omega_c \tau^2 \beta/m) [1 + 4\omega_c^2 \tau^2]^{-1}. \quad (8.6)$$

<sup>9</sup> E. H. Sondheimer, Proc. Roy. Soc. (London) **203**, 75 (1950).  
<sup>10</sup> D. J. Howarth and E. H. Sondheimer, Proc. Roy. Soc. (London) **219**, 53 (1953).

The coefficient of viscosity is therefore the zero-field value  $\eta(0)$ , with  $S_1(\zeta)$  replacing the former  $S(\zeta)$ :

$$\eta(H) = \eta(0)[1 + 4\omega_c^2\tau^2]^{-1}. \quad (8.7)$$

The magnetic field suppresses the viscosity by shortening the mean free path in the direction of transport. Apart from differences in the magnitude of  $\tau$ , the term  $4\omega_c^2\tau^2$  in (8.7) replaces  $\omega_c^2\tau^2$  in the analogous result for the conduction problem. This is owing to charge transport being reversed by turning through  $180^\circ$  while transverse momentum transport is reversed by turning through  $90^\circ$ , or in one-half the time. The assumption of

a time of relaxation limits the validity of (8.7) to  $T \gg \Theta$ . However, as shown by Sondheimer and Wilson for the electrical conductivity,<sup>11</sup> such a formula is probably more widely applicable than its derivation would suggest.

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<sup>11</sup> E. H. Sondheimer and A. H. Wilson, Proc. Roy. Soc. (London) **190**, 435 (1947).

## Absence of Diffusion in Certain Random Lattices

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This paper presents a simple model for such processes as spin diffusion or conduction in the "impurity band." These processes involve transport in a lattice which is in some sense random, and in them diffusion is expected to take place via quantum jumps between localized sites. In this simple model the essential randomness is introduced by requiring the energy to vary randomly from site to site. It is shown that at low enough densities no diffusion at all can take place, and the criteria for transport to occur are given.

### I. INTRODUCTION

A NUMBER of physical phenomena seem to involve quantum-mechanical motion, without any particular thermal activation, among sites at which the mobile entities (spins or electrons, for example) may be localized. The clearest case is that of spin diffusion<sup>1,2</sup>; another might be the so-called impurity band conduction at low concentrations of impurities. In such situations we suspect that transport occurs not by motion of free carriers (or spin waves), scattered as they move through a medium, but in some sense by quantum-mechanical jumps of the mobile entities from site to site. A second common feature of these phenomena is randomness: random spacings of impurities, random interactions with the "atmosphere" of other impurities, random arrangements of electronic or nuclear spins, etc.

Our eventual purpose in this work will be to lay the foundation for a quantum-mechanical theory of transport problems of this type. Therefore, we must start with simple theoretical models rather than with the complicated experimental situations on spin diffusion or impurity conduction. In this paper, in fact, we attempt only to construct, for such a system, the simplest model we can think of which still has some expectation of representing a real physical situation

reasonably well, and to prove a theorem about the model. The theorem is that at sufficiently low densities, transport does not take place; the exact wave functions are localized in a small region of space. We also obtain a fairly good estimate of the critical density at which the theorem fails. An additional criterion is that the forces be of sufficiently short range—actually, falling off as  $r \rightarrow \infty$  faster than  $1/r^3$ —and we derive a rough estimate of the rate of transport in the  $V \propto 1/r^3$  case.

Such a theorem is of interest for a number of reasons: first, because it may apply directly to spin diffusion among donor electrons in Si, a situation in which Feher<sup>3</sup> has shown experimentally that spin diffusion is negligible; second, and probably more important, as an example of a real physical system with an infinite number of degrees of freedom, having no obvious oversimplification, in which the approach to equilibrium is simply impossible; and third, as the irreducible minimum from which a theory of this kind of transport, if it exists, must start. In particular, it re-emphasizes the caution with which we must treat ideas such as "the thermodynamic system of spin interactions" when there is no obvious contact with a real external heat bath.

The simplified theoretical model we use is meant to represent reasonably well one kind of experimental situation: namely, spin diffusion under conditions of

<sup>1</sup> N. Bloembergen, *Physica* **15**, 386 (1949).

<sup>2</sup> A. M. Portis, *Phys. Rev.* **104**, 584 (1956).

<sup>3</sup> G. Feher (private communication).