of the wave-function amplitude over the FS when they become available. Since all of the calculations of the amplitude of $\bar{\sigma}$ involve approximations which either eliminate the details of the wave-function amplitude completely or are not applicable to this case, there has been no attempt to make detailed comparisons to them. It is hoped that these measurements and others will stimulate more detailed theoretical studies of the effect.

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Inelastic Electron Scattering and the Lorenz Ratio of Liquid Metals

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Recent experimental work has led to the surprising conclusion that the Lorenz ratios of some of the liquid metals exhibit substantial negative deviations from the ideal Sommerfeld value, hence indicating that inelastic electron scattering effects may be an important factor in some liquid metals. Motivated by these observations, we present in this paper a calculation of the electronic transport properties of a liquid metal, which takes into account the effects of inelastic electron scattering from the ionic density fluctuations to leading order in the small dimensionless parameter $E_0/k_B T$. Here E_0 denotes a typical inelastic energy transfer and $k_B T$ the Boltzmann factor. This calculation, based on the nearly-free-electron model, is carried through by use of the exact sum rules of Placzek and of de Gennes on the dynamic structure factor $S(q, \omega)$ of the classical ionic liquid component of the metal. The effects of inelastic electron scattering on the electrical resistivity are found to be negligible. Non-negligible corrections, however, associated with small-angle inelastic processes, are found to enter the electronic thermal resistivity. The corresponding depression in the Lorenz number is expressed in terms of the electron-ion pseudopotential, the static liquid structure factor, and the collective-mode frequencies associated with the density fluctuations of the liquid component. Inspection of the theoretical expression for the deviation in the Lorenz ratio reveals, however, that it is too small to account for the experimentally observed deviations. It is concluded that, within the framework of the nearly-free-electron theory, the effects of inelastic electron scattering are not the dominant cause of the rather large anomalies in the observed Lorenz ratios.

I. SYNOPSIS

In the currently existing nearly-free-electron theory[1-4] of the electronic transport properties of liquid metals, the electrons are considered to be scattered *elastically* from the ionic density fluctuations of the liquid component of the metal. A simple, but important, consequence of this approximation is that the electronic thermal conductivity κ_e is related to the electrical resistivity ρ by means of the Wiedemann-Franz relation[5]

$$\rho \kappa_e / T = L_0 = \tfrac{1}{3}\pi^2 (k_B/e)^2.$$

Recent experimental data for several liquid metals show, however, significant negative deviations of the Lorenz ratio $L = \rho \kappa_e / T$ from the ideal Sommerfeld value L_0. For example, Yurchak and Smirnov[6] and Duggin[7] have observed a deviation in the Lorenz ratio of liquid Ga of the order of -20%, while Filippov[8] has reported deviations in liquid Sn and Pb of the order of -30 and -12%, respectively. Deviations varying between -20 and -40% were reported for Cu some time ago.[9] Moreover, these deviations may be a fairly strong function of temperature.[6,8,10] Such *negative*

deviations cannot be accounted for by the existence of a finite thermal conduction by the liquid component of the metal.[11] Also, use of a simple textbook formula[13] shows that the effects of electron-electron scattering, which would act to decrease the electronic thermal conductivity while having no effect on the electrical resistivity, are completely negligible. Thus, *assuming the soundness of the experimental work*, it would appear that the anomaly in the Lorenz ratio is associated with the electron scattering from the ionic density fluctuations of the liquid, and, specifically, with a distinct difference in the corresponding transport relaxation times for electrical and thermal conduction. This suggests that *inelastic* electron scattering from the ionic density fluctuations may be a significant factor in some liquid metals. As is well known in the theory of electronic transport phenomena in metals,[12,13] when inelastic electron scattering is operative, the electronic thermal resistivity receives a contribution from small-angle inelastic scattering processes, a corresponding contribution not appearing in the electrical resistivity. This effect causes a depression of the Lorenz ratio with respect to the classical value L_0.[14] That inelastic scattering effects may be important in liquid metals was suggested by Duggin[7] in presenting his experimental measurements on liquid Ga.

Motivated by these observations, we present in this paper a calculation of the electronic transport properties of a liquid metal which takes into account the effects of inelastic electron scattering to leading order in the small dimensionless parameter $y = E_0/k_B T$. Here

$$E_0 = \hbar^2 k_F^2/2M$$

denotes a typical inelastic energy transfer, T the absolute temperature, k_B Boltzmann's constant, M the ionic mass, and k_F the Fermi wave vector of the electron system. The results of this calculation will enable us to determine whether or not inelastic scattering effects are in fact important enough to be responsible for the observed deviations in the Lorenz number. Following the work of Mannari[15] and of Baym,[16] we first express the electrical resistivity ρ and the electronic thermal conductivity κ_e in terms of the dynamic structure factor $S(q, \omega)$ of the ionic liquid component of the metal. The leading-order inelastic corrections to the usual nearly-free-electron-theory results for ρ and κ_e are then obtained by explicit use of the exact sum rules of Placzek[17] and of de Gennes[18] on the structure factor. It turns out that the inelastic corrections to the electrical resistivity are completely negligible. Nonnegligible effects, however, associated with the small-angle inelastic scattering processes, are found to enter the electronic thermal conductivity. The deviation in the Lorenz ratio from the Sommerfeld value L_0 brought about by the new term found in κ_e is expressed in terms of the static structure factor $S(q)$, the Fourier transform $v(q)$ of the effective electron-ion interaction, and the collective-mode frequencies[18] ω_q associated with the

ionic density fluctuations. In principle, this result would provide a new opportunity to test theoretical calculations of the basic liquid-metal properties $S(q)$, $v(q)$, and ω_q, namely, against the experimentally observed deviations in the Lorenz ratio. Inspection of the theoretical expression for the deviation in the Lorenz ratio reveals, however, that it is much too small to account for the experimentally observed deviations. We conclude, therefore, that within the framework of the nearly-free-electron theory the effects of inelastic electron scattering from the ionic density fluctuations are not the cause of the rather large anomalies in the experimentally determined Lorenz ratios of liquid Ga, Sn, Pb, and Cu. In view of the difficulty of thinking of another simple mechanism which could give rise to such strong anomalies, we stress here the importance of further experimental confirmation of the presently reported deviations in the Lorenz ratios of the liquid metals.

II. CALCULATION OF TRANSPORT COEFFICIENTS

The effects of the inelastic scattering of electrons from the ionic density fluctuations in a liquid metal may be readily incorporated into the usual nearly-free-electron theory[1-4] by following the work of Mannari[15] and of Baym.[16] Here the rates of transitions among the free-electron states induced by the scattering from the ionic density fluctuations are calculated in the Born approximation in terms of the dynamic structure factor $S(q, \omega)$ of the ionic liquid:

$$\Gamma(\mathbf{k+q} \rightarrow \mathbf{k}) = \frac{2\pi}{\hbar} \frac{|v(q)|^2}{N} f_{k+q}(1-f_k)$$

$$\times \int_{-\infty}^{\infty} d\omega\, S(q, \omega)\delta(\epsilon_{k+q} - \epsilon_k - \hbar\omega). \quad (2.1)$$

In this equation the left-hand side denotes the electronic transition rate for transitions from the free-electron momentum state $\hbar(\mathbf{k+q})$ to the state $\hbar\mathbf{k}$, f_k denotes the electronic state distribution function, $\epsilon_k = \hbar^2 k^2/2m$ is the free-electron energy, m is the electronic mass, and $v(q)$ is the electron pseudopotential, i.e., the Fourier transform of the effective electron-ion interaction.[1-4] The total number of electrons in the system, of volume Ω, is ZN, where N denotes the total number of ions and Z the valence of the metal. The dynamic structure factor is defined as the Fourier transform of the liquid density fluctuation time correlation function[18]:

$$S(q, \omega) = (2\pi N)^{-1} \int_{-\infty}^{\infty} e^{-i\omega t}\, dt\, \langle \rho_q(0)\rho_{-q}(t) \rangle_L, \quad (2.2)$$

where ρ_q denotes the ionic liquid density fluctuation operator

$$\rho_q = \sum_j \exp(-i\mathbf{R}_j \cdot \mathbf{q}),$$

and the angular brackets denote an ensemble average

over the liquid component of the metal. $\mathbf{R}_j$ denotes the coordinate of the jth ion of the liquid component. The transition rate (2.1), together with the inverse rate $\Gamma(\mathbf{k}\rightarrow\mathbf{k}+\mathbf{q})$, may be fed into the standard first-order variational procedure of transport theory[19] in order to obtain the following expressions for the electrical resistivity ρ and electronic thermal conductivity κ_e[20]:

$$\rho = \frac{6\pi\Omega m}{Ne^2\hbar^2\epsilon_F} \int_0^1 dx \, | \, v(x) \, |^2 \, x^3$$

$$\times \int_{-\infty}^{\infty} S(q, \omega)n(\omega)\bar{\omega} \, d\omega, \quad (2.3)$$

$$\kappa_e^{-1} = \frac{6\pi\Omega m}{Ne^2\hbar^2\epsilon_F L_0 T} \int_0^1 dx \, | \, v(x) \, |^2 \, x^3 \int_{-\infty}^{\infty} S(q, \omega)n(\omega)\bar{\omega}$$

$$\times [1 + (3\bar{\omega}^2/4\pi^2)(1/x^2 - \tfrac{2}{3})]. \quad (2.4)$$

In (2.3) and (2.4) we have introduced the notation $\bar{\omega} = \hbar\omega/k_B T$ and $x = q/2k_F$, and ϵ_F denotes the Fermi energy. Also, $n(\omega)$ denotes the Planck function

$$n(\omega) = 1/[\exp(\hbar\omega/k_B T) - 1].$$

There are two pertinent observations that can be made from the results (2.3) and (2.4). First, in the limit of high temperatures $\bar{\omega}\rightarrow 0$, in which the finite energy transfer $\hbar\omega$ between the electron and ionic systems may be neglected, we may put $n(\omega)\bar{\omega} = 1$ and neglect the second term in (2.4) proportional to $\bar{\omega}^2$. In this case the integrals appearing in (2.3) and (2.4) are the same, and it follows that the Wiedemann-Franz law holds: $\rho\kappa_e/T = L_0$. The electrical resistivity is given by

$$\rho = \frac{6\pi\Omega m}{Ne^2\hbar^2\epsilon_F} \int_0^1 dx \, | \, v(x) \, |^2 \, x^3 S(x), \quad (2.5)$$

which is the usual Bhatia-Krishnan-Gerstenkorn-Ziman (BKGZ) result,[1–4] where $S(x)$ denotes the static liquid structure factor

$$S(q) = \int_{-\infty}^{\infty} S(q, \omega) \, d\omega.$$

The second point is that for finite $\bar{\omega}$ we see from the second term of (2.4) that there is a significant contribution to the thermal resistivity $1/\kappa_e$ coming from small values of the momentum transfer $x = q/2k_F$. This is the contribution to the thermal resistivity arising from small-angle inelastic scattering processes that we mentioned in Sec. I. It will be this term that provides the main effect in the present calculation.

We wish to calculate the corrections to the transport coefficients obtained in the $\bar{\omega}\rightarrow 0$ limit that are of first order in the parameter

$$y = E_0/k_B T = \hbar^2 k_F^2/2M k_B T.$$

This parameter, whose magnitude measures the relative importance of inelastic scattering, is typically of the order of 10^{-3}–10^{-2} at the melting point T_M. In order to carry out this calculation it will first be convenient to express the dynamic structure factor $S(q, \omega)$ in terms of a closely related function, namely, the spectral distribution of density fluctuations $A(q, \omega)$,[21]

$$A(q, \omega) \equiv (2\pi N)^{-1} \int_{-\infty}^{\infty} dt \, e^{-i\omega t} \langle \{\rho_\mathbf{q}(0), \rho_{-\mathbf{q}}(t)\} \rangle_L.$$

In terms of $A(q, \omega)$, $S(q, \omega)$ is just[21]

$$S(q, \omega) = A(q, \omega)[n(\omega) + 1].$$

The integrals over frequency which have to be performed in Eqs. (2.3) and (2.4) then have the form

$$\int_{-\infty}^{\infty} d\omega \, A(q, \omega)n(\omega)[n(\omega) + 1]\bar{\omega}^m \quad (m = 1, 3).$$

The next step of the calculation is to replace the product of Planck functions appearing in these integrands by the high-temperature expansion

$$n(\omega)[n(\omega) + 1]$$

$$= (k_B T/\hbar\omega)^2 [1 - \tfrac{1}{12}\bar{\omega}^2 + (7\bar{\omega}^4/720) - \cdots].$$

This expansion, of course, is appropriate for liquid metals (lithium excepted), since the dominant weight of the spectral density function is confined to values of $\hbar\omega$ less than $k_B T$. The resulting integrations over ω in (2.3) and (2.4) may then be reduced to a sum of terms, each of which involves a moment of the spectral density function

$$a_{(n)}(q) = \int_{-\infty}^{\infty} d\omega \, A(q, \omega)\bar{\omega}^n, \quad (2.6)$$

where n is an odd integer and $n \geq -1$. The final step of the calculation is to evaluate the moments (2.6) in the high-temperature limit $\bar{\omega}\rightarrow 0$. These moments have been evaluated by Placzek[17] and by de Gennes.[18] First, we note that in the high-temperature limit $a_{(-1)}(q)$ is just the static structure factor $S(q)$. The first- and third-order moments are[17,18]

$$a_{(1)}(q) = \hbar^2 q^2/M k_B T, \quad (2.7)$$

$$a_{(3)}(q) = 3a_{(1)}(q)[a_{(1)}(q) + \tfrac{1}{3}(\hbar\omega_q/k_B T)^2], \quad (2.8)$$

where ω_q denotes the liquid collective-mode frequency, given in terms of the conventional pair-density function $g(r)$ of the ionic liquid and the ion-ion potential $V(r)$ by the relation[18]

$$M\omega_q^2 = \int d\mathbf{r} \, g(r)[1 - \cos(qx)]\frac{\partial^2 V(r)}{\partial x^2}, \quad (2.9)$$

where x denotes the x component of the radial distance $\mathbf{r}$. We will require only the first three moments for a consistent evaluation of the transport properties to leading order in y.

The resulting expressions for the transport coefficients

are

$$\rho = \frac{6\pi\Omega m}{e^2 N\hbar\epsilon_F} \int_0^1 dx \mid v(x) \mid^2 x^3[S(x) - \tfrac{2}{3}y\gamma x^2], \quad (2.10)$$

$$\kappa_e^{-1} = \frac{\rho}{L_0 T} + \frac{36y\Omega m}{\pi Ne^2\hbar\epsilon_F L_0 T} \int_0^1 dx \mid v(x) \mid^2 x^3(1 - \tfrac{2}{3}x^2)$$
$$\times[1 - \tfrac{1}{12}(\hbar\omega_x/k_B T)^2], \quad (2.11)$$

where we have defined

$$\gamma = 1 - (7/60)(\hbar\omega_x/k_B T)^2$$

and ρ appearing in (2.11) is given by (2.10). We now note that the correction of order y appearing in (2.10) for ρ may be neglected; i.e., the effects of inelastic scattering on the electrical resistivity are negligible. This is because for all x we have the inequality

$$S(x) \gg \tfrac{2}{3}x^2\gamma y \quad (2.12)$$

very well satisfied for liquid metals. To see this we first note that for small x, $S(x)$ may be estimated by the relation[22,23]

$$S(0) = \tfrac{3}{2}(k_B T/Z\epsilon_F) \sim 10^{-3} - 10^{-2} \text{ (typically)},$$

while for $x \sim 1$ we know experimentally that $S(1) \sim 1$.[24] Also, it follows from the definition (2.9) that $\omega_x^2 \propto x^2$ for $x \ll 1$, while for large x, $\hbar\omega_x$ approaches a constant $\hbar\omega_E$ which is of order of the Debye energy $k_B\Theta_D$ of the metal.[25] The maximum value of $\hbar\omega_x/k_B T$ is therefore of order Θ_D/T_M, typically less than unity, so that for all x, $\gamma \simeq 1$. Thus, since $y \sim S(0)$, we have (2.12) very well satisfied for all x. It follows that the expression (2.10) for ρ may be replaced by the BKGZ formula (2.5). On the other hand, the correction to $1/\kappa_e$ given by the second term of (2.11) is in general non-negligible, since it gives a contribution relative to the elastic contribution of order $y/S(x)$ for a given value of x, and is therefore important for small x, for which this ratio is of the order of unity. The results for the thermal conductivity may be written in the form

$$\rho\kappa_e/T = L_0/(1 + \alpha_1), \quad (2.13)$$

where

$$\alpha_1 = \frac{6}{\pi^2} \frac{E_0}{k_B T} \frac{\langle(1 - \tfrac{2}{3}x^2)[1 - \tfrac{1}{12}(\hbar\omega_x/k_B T)^2]\rangle}{\langle S(x)\rangle}. \quad (2.14)$$

The angular brackets denote the wave-vector averages

$$\langle f(x)\rangle \equiv \int_0^1 dx \mid v(x) \mid^2 x^3 f(x). \quad (2.15)$$

This completes our formal calculation of the effects of inelastic electron scattering.

In concluding this section we recall that Eqs. (2.3) and (2.4) for the transport coefficients were derived on the basis of the usual first-order variational procedure of treating the Boltzmann equation.[19,20] If the electron scattering were completely elastic, this procedure would be exact.[19] Since we are working close to the elastic limit, we expect the use of the variational procedure

to introduce only a very small error in the above calculations of ρ and κ_e.

III. DISCUSSION OF RESULTS OBTAINED

We have presented a calculation of the electronic transport properties of a liquid metal in which the effects of the inelastic scattering of electrons from the ionic density fluctuations have been taken into account to first order in the small parameter $y = E_0/k_B T$. To this order, the effects of inelastic electron scattering on the electrical resistivity were shown to be negligible, while non-negligible corrections, arising from small-angle inelastic processes, were found for the electronic thermal conductivity κ_e. The resulting electronic Lorenz ratio may be written down from (2.13) as

$$L = L_0/(1 + \alpha_1),$$

where α_1 is the ratio of wave-vector averages defined in (2.14). It will be convenient for our subsequent discussion to use the BKGZ result (2.5) for ρ in order to reexpress α_1 in the form

$$\alpha_1 = \frac{8Z}{\pi} \frac{E_0}{k_B T} k_F l_0 \left\langle \frac{(1 - \tfrac{2}{3}x^2)[1 - \tfrac{1}{12}(\hbar\omega_x/k_B T)^2]}{(\tfrac{2}{3}\epsilon_F)^2} \right\rangle, \quad (3.1)$$

where l_0 is the electronic mean free path for conduction,

$$1/\rho = ZNe^2 l_0/\Omega\hbar k_F. \quad (3.2)$$

In the wave-vector average in (3.1) we have divided out by the $q = 0$ value of the electron pseudopotential[4] $-\tfrac{2}{3}\epsilon_F$ in order to render these averages dimensionless. We recall that the wave-vector average is defined by Eq. (2.15).

We may now estimate the order of magnitude of the inelastic term α_1 and hence determine whether or not inelastic scattering effects are large enough to account for the anomalies in the experimentally determined Lorenz ratios. The magnitude of α_1 is determined by the product of the dimensionless quantity

$$a = (8Z/\pi)(E_0/k_B T)k_F l_0$$
$$= (8Z/\pi)(m/M)(T_F/T)k_F l_0, \quad (3.3)$$

with the dimensionless wave-vector average defined in (3.1). In (3.3), T_F denotes the Fermi temperature $k_B T_F = \epsilon_F$. At the melting point, use of (3.3) and (3.2) shows that $a \sim 1$ typically. For example, at T_M we have $a = 0.9$ for Na, $a = 1.25$ for Li, and $a = 0.72$ for Ga. Thus, the order of magnitude of α_1 is determined by the order of magnitude of the wave-vector average appearing in α_1. This average is a little difficult to estimate without explicit recourse to detailed numerical calculations.[4,26,27] Its precise value will depend quite sensitively on the shape of the pseudopotential and, to a lesser extent, on the relative importance of the collective-mode term.[32] An *extreme* upper limit to the magnitude of the average may be obtained by neglecting the

collective-mode term in the integrand of the average and by neglecting the shape of the electron pseudopotential by taking $|v(x)| = \frac{2}{3}\epsilon_F$ for all x. In this case the average appearing in α_1 is 0.14, indicating a deviation in the Lorenz ratio of order -14%. In the more realistic situation,[28–31] however, $v(x)$ is substantially less than $\frac{2}{3}\epsilon_F$ in the important range of wave vectors $x \lesssim 1$. Thus, typically we would expect much smaller values for the wave-vector averages, and hence for the effects of inelastic electron scattering. For example, if for liquid Ga, $v(q)$ is chosen to be a Coulomb potential screened by the Lindhard dielectric function, and the collective-mode term is neglected,[32] we find by numerical integration that $\alpha_1 = 2.1\%$.

We have to conclude, therefore, that within the present framework of the nearly-free-electron theory, the effects of inelastic electron scattering from the ionic density fluctuations are not the cause of the rather large anomalies in the experimentally determined Lorenz ratios of liquid Ga, Sn, Pb, and Cu. It is difficult to think of another simple mechanism that would be important enough to give rise to such anomalies. We have already pointed out that the effects of electron-electron scattering are negligible. Corrections to the transport coefficients arising from the use of finite temperature in the Fermi-Dirac statistics are probably also too small.[33] Another mechanism which

could be considered is the possibility of a large effective electron-electron interaction induced by the ionic density fluctuations. However, such an effect could also be expected to be present in the metal just below the melting point, and to the author's knowledge there does not appear to be any experimental evidence for this. Finally, we mention that the present calculation could predict large corrections to the ideal Lorenz ratio if a suitable shape of electron pseudopotential were to be employed. Such a shape, which would have to be strongly peaked at small wave vectors, would be radically different from the conventionally accepted shape for metals, and is highly unlikely. We would stress, then, the importance of further experimental confirmation of the presently reported deviations in the Lorenz ratios of the liquid metals.

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