

Direct Calculation of Electronic Properties of Metals from Neutron Scattering Data

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The cross section for the scattering of conduction electrons in a metal by the lattice oscillations is written in terms of the observed slow-neutron inelastic-scattering cross section. This enables one to include all multiphonon processes, Debye-Waller factors, and umklapp processes in the electron-lattice interaction without having to use a phonon description of the lattice oscillations. Expressions which involve only the effective electron-lattice matrix elements and the observed neutron-scattering data are given for the electron self energy, phonon-induced effective mass, electrical and thermal conductivities, and viscosity.

SOLID-STATE physicists have expended an enormous amount of labor over the years calculating the consequences of electron-phonon interactions in real metals. They have labored similarly extracting information on the one-phonon spectrum from slow neutron scattering data. It is the purpose of this paper to show how one can pass directly from essentially unanalyzed neutron scattering data to a calculation of those metallic properties that are influenced by the interaction of the electrons with lattice oscillations. The method proposed permits one to take into account all multiphonon processes, Debye-Waller factors, umklapp processes, couplings to transverse phonons and anharmonic phonon effects, without ever having to go through an intermediate description of the lattice oscillations in terms of phonons.¹

The inelastic scattering of a slow neutron from a metal is a process really very similar to the inelastic scattering of a conduction electron from the lattice oscillations. Both couple, not to individual phonons, but to the ion density; and for both a Born approximation is valid.

In the pseudopotential approximation, the scattering cross section for a process in which the neutron loses momentum \mathbf{k} and energy ω is, aside from trivial factors, the number $S(\mathbf{k}, \omega)$ of available vibrational states of the lattice with momentum \mathbf{k} and energy ω . In terms of $\rho(\mathbf{k}, t)$, the Fourier transform of the operator $\rho(\mathbf{r}, t)$ for the density of nuclei, S is given by

$$S(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \rho(\mathbf{k}, t) \rho^\dagger(\mathbf{k}, 0) \rangle \quad (1)$$

where the expectation value is in the equilibrium ensemble for the metal.

Conduction electrons in Bloch states, in a metal with rigid ion cores, are scattered via a screened potential by the fluctuations of ion density. As was pointed out by

¹ L. J. Sham and J. M. Ziman in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1963), Vol. 15, review the previous work that went beyond the simple one-phonon picture of the interaction of electrons with the lattice. They also review the attempts that have been made to view the electron-lattice interaction as a diffraction problem. Attention is also called to the work of I. Mannari [Progr. Theoret. Phys. (Kyoto) 26, 51 (1961)] on electrical conductivity, in which he points out that the scattering cross section for conduction electrons is connected with the time-dependent pair correlation function of the ions.

Migdal,^{2,3} this scattering in the normal metal is correctly described, to lowest order in $(m/M)^{1/2}$, by the Born approximation. The scattering probability is thus proportional to the number $S'(\mathbf{k}, \omega)$ of available (fully interacting) states for density fluctuations. This number is just S with the elastic Bragg peaks subtracted out:

$$S'(\mathbf{k}, \omega) = S(\mathbf{k}, \omega) - 2\pi\delta(\omega) |\langle \rho(\mathbf{k}) \rangle|^2. \quad (2)$$

Consider first the electron-lattice collision term in the Boltzmann equation for the electronic distribution function. In a collision an electron in Bloch state \mathbf{p} with energy ϵ scatters to a state \mathbf{p}' with energy ϵ' by creating a density fluctuation with momentum \mathbf{k} and energy ω . (For $\omega < 0$ this is effectively an absorption process.) Let the matrix element of the screened potential be $\langle \mathbf{p}' | v(\mathbf{k}, \omega) | \mathbf{p} \rangle$ for this process. Then the rate at which the scattering occurs is given by the golden rule as

$$2\pi\delta(\epsilon - \epsilon' - \omega) f(\mathbf{p})(1 - f(\mathbf{p}')) N S'(\mathbf{k}, \omega) |\langle \mathbf{p}' | v(\mathbf{k}, \omega) | \mathbf{p} \rangle|^2, \quad (3)$$

where $f(\mathbf{p})$ is the density of electrons in the Bloch state \mathbf{p} , $1 - f(\mathbf{p})$ is the density of available final states \mathbf{p}' , and N is the number of ions per unit volume. The entire collision term is

$$\left(\frac{\partial f(\mathbf{p})}{\partial t} \right)_{\text{coll}} = \sum_{\mathbf{p}' \mathbf{k}} \int_{-\infty}^{\infty} d\omega \delta(\epsilon - \epsilon' - \omega) N |\langle \mathbf{p}' | v(\mathbf{k}, \omega) | \mathbf{p} \rangle|^2 \\ \times [f(\mathbf{p}')(1 - f(\mathbf{p})) S'(-\mathbf{k}, -\omega) \\ - f(\mathbf{p})(1 - f(\mathbf{p}')) S'(\mathbf{k}, \omega)]. \quad (4)$$

The identification of S' with the experimentally observed equilibrium density of states for lattice fluctuations assumes that phonon equilibration rates are much more rapid than electron-phonon scattering rates. At temperature $T (= 1/K\beta)$ the equilibrium S' obeys the detailed balancing condition

$$S'(-\mathbf{k}, -\omega) = e^{-\beta\omega} S'(\mathbf{k}, \omega). \quad (5)$$

² A. B. Migdal, Zh. Eksperim. i Teor. Fiz. 34, 1438 (1958) [English transl.: Soviet Phys.—JETP 34, 996 (1958)]; A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963), Chap. IV.

³ R. E. Prange and L. P. Kadanoff, Phys. Rev. 134, A566 (1964).

In the "one-phonon approximation"

$$S'(\mathbf{k}, \omega) = \frac{2\pi}{M} \sum_{\lambda} (\mathbf{k} \cdot \mathbf{e}_{\mathbf{k}\lambda})^2 \delta(\omega^2 - \omega_{\mathbf{k}\lambda}^2) \frac{\omega/\omega_{\mathbf{k}\lambda}}{1 - e^{-\beta\omega}}, \quad (6)$$

where $\mathbf{e}_{\mathbf{k}\lambda}$ is the polarization vector of a phonon with polarization index λ ; on substituting (6) into (4) one recovers the ordinary Boltzmann equation collision term.

To give the reader a feeling for how the transport coefficients depend on S' , we give the results of the most elementary variational calculations with the Boltzmann equation in which we linearize the collision term, assume free electrons with an effective mass⁴ m , take the matrix element to depend only on k :

$$|\langle \mathbf{p}' | v(\mathbf{k}, \omega) | \mathbf{p} \rangle|^2 \rightarrow |v(k)|^2 \delta_{\mathbf{p}', \mathbf{p}-\mathbf{k}} \quad (7)$$

and use the simplest trial functions.⁵

The electrical conductivity is given by $\sigma_e = ne^2\tau_e/m$ where

$$\tau_e^{-1} = \frac{m}{12\pi^3 Z} \int_0^{2p_f} k^3 dk |v(k)|^2 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S'(k, \omega) \beta \omega n(\omega). \quad (8)$$

Z is the number of conduction electrons per ion and $n(\omega) = (e^{\beta\omega} - 1)^{-1}$. Eq. (8) is a generalization of the Grüneisen formula⁵ for the conductivity in that included in S' are multiphonon processes, Debye-Waller factors, umklapp processes, etc. Substituting (6) into (8) gives the usual Grüneisen formula. Eq. (8) has been derived by Mannari¹ for the special case of a liquid metal.

Only for temperatures much greater than θ_D , the Debye temperature, is τ_e strictly interpretable as a relaxation time. In this limit, $\beta\omega n(\omega) \rightarrow 1$, so that the ω integral becomes $S'(\mathbf{k})$, the static structure factor (minus the Bragg peaks at reciprocal lattice vectors). Thus, we see that the recent use of the resultant formula by Ziman and co-workers in the study of liquid metals⁶ has, in fact, included all multiphonon processes. The temperature dependence of the high-temperature conductivity is determined only by the temperature dependence of $S'(\mathbf{k})$.

We find for the electron viscosity $\eta = 2n\epsilon_f\tau_v/5$, where

$$\tau_v^{-1} = \frac{m}{12\pi^3 Z} \int_0^{2p_f} dk (3k^3 - 3k^5/4p_f^2) |v(k)|^2 \times \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S'(k, \omega) \beta \omega n(\omega). \quad (9)$$

In general, τ_v differs from τ_e , the integration giving more weight to $k/p_f < (8/3)^{1/2}$ than τ_e and less to the region

$(8/3)^{1/2} < k/p_f < 2$. This can well lead to differences in the temperature dependence of τ_e and τ_v .

The thermal conductivity becomes $\sigma_t = C_V v_f^2 \tau_t/3$, where

$$\tau_t^{-1} = \frac{m}{12\pi^3 Z} \int_0^{2p_f} dk |v(k)|^2 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S'(k, \omega) \beta \omega n(\omega) \times \left[k^3 + \left(\frac{\beta\omega}{\pi} \right)^2 k(3p_f^2 - k^2/2) \right]. \quad (10)$$

For $T \gg \theta_D$, the $(\beta\omega)^2$ term is negligible; $\tau_t \rightarrow \tau_e$, so that the Wiedemann-Franz law obtains.

The method of describing the electron-lattice interaction in terms of S' is not limited to the Boltzmann equation. The entire perturbation expansion for the electron-phonon interaction, commonly written in terms of $D(\mathbf{k}, z)$, the phonon Green's function, is more correctly written with $\mathbf{k} \cdot D(\mathbf{k}, z) \cdot \mathbf{k}$ replaced by

$$\int \frac{d\omega}{2\pi} \frac{S'(\mathbf{k}, \omega)}{z - \omega} (1 - e^{-\beta\omega}).$$

For example, the phonon contribution to the effective electron-electron interaction is

$$V_{e\text{eff}} = \frac{1}{2} \sum_{\{\mathbf{p}\}, \mathbf{k}} \langle \mathbf{p}'' | v(\mathbf{k}, z) | \mathbf{p} \rangle \langle \mathbf{p}' | v(-\mathbf{k}, -z) | \mathbf{p}' \rangle \times \int \frac{d\omega}{2\pi} \frac{S'(\mathbf{k}, \omega)}{z - \omega} (1 - e^{-\beta\omega}) c_{\mathbf{p}'', \dagger} c_{\mathbf{p}', \dagger} c_{\mathbf{p}', \dagger} c_{\mathbf{p}, \dagger}, \quad (11)$$

where z is the energy transfer in the interaction.

Following the procedure due to Migdal,² we can write the electron self energy in terms of S' as

$$\Sigma(\mathbf{p}, z) = \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{d\omega}{2\pi} \frac{1 + n(\omega) - f_0(\epsilon)}{z - \epsilon - \omega} \frac{S'(\mathbf{k}, \omega)}{1 + n(\omega)} \times \sum_{\mathbf{p}'} |\langle \mathbf{p}' | v(\mathbf{k}) | \mathbf{p} \rangle|^2 \delta(\epsilon' - \epsilon_f). \quad (12)$$

Then, using (7), we find as the phonon contribution to the electron effective mass at $T \ll \theta_D$:

$$\frac{m^*}{m} = 1 + \frac{mN}{2\pi^2 p_f} \int_0^{2p_f} k dk |v(k)|^2 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{S'(k\omega)}{\omega}. \quad (13)$$

At high temperatures m^* approaches m .

To sum up, we see that the only quantity that remains to be calculated theoretically is the effective matrix element $\langle \mathbf{p}' | v(\mathbf{k}) | \mathbf{p} \rangle$, since $S(\mathbf{k}, \omega)$ is directly obtained from neutron scattering experiments. There do not exist, at present, sufficiently detailed maps of $S(\mathbf{k}, \omega)$, for a simple metal like lead or aluminum at a representative number of points in \mathbf{k} space, to carry out detailed evaluations of the formulas given here.

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⁴ This effective mass m should not include effects of electron-phonon interactions. See Ref. 3.

⁵ J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1963), Chap. IX, details the elementary variational calculations of electronic transport coefficients from the Boltzmann equation.

⁶ Reviewed in Sham and Ziman, Ref. 1.