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

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

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 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:

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

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

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

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

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

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

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

[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.

[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 mass[4] m, take the matrix element to depend only on k:

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

and use the simplest trial functions.[5]

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

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

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 formula[5] 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 Mannari[1] 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, $\beta\omega n(\omega)\to 1$, 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 metals[6] 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

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

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 $\sigma_t=C_Vv_f^2\tau_t/3$, where

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

For $T\gg\theta_D$, the $(\beta\omega)^2$ term is negligible; $\tau_t\to\tau_e$, 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

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

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

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

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

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

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.