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SOUND ATTENUATION IN METALS: REFORMULATION OF PIPPARD THEORY TO INCLUDE MICROSCOPIC BAND STRUCTURE AND SCATTERING

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Abstract - Pippard's theory is written in Boltzmann equation form which allows both band effects and microscopic scattering to be included. A new formulation of deformation potential theory is an essential ingredient. Numerical calculations are given for Cu, using realistic electron and phonon eigenstates and the rigid-muffin-tin model for electron-phonon coupling. The calculations cover the crossover from hydrodynamic to quantum regimes, and show satisfactory agreement with experiment.

Pippard's[1] theory of ultrasonic attenuation in metals clarifies the crossover from a hydrodynamic regime, where electrons are in local thermal equilibrium at each point of the sound wave of wave vector Q ($Q\ell<<1$), to a quantum regime, where electrons have mean free paths large compared to the sound wavelength ($Q\ell>>1$). The purpose of this paper is to generalize Pippard's theory to the case of Bloch electrons[2] in real metals, with a microscopic description of the scattering of electrons via interactions with impurities and thermal phonons. Such a generalization has eluded solution, partly because of the difficulty of understanding Pippard's procedure[2] and partly because of some subtleties in the meaning of the deformation potential[3]. We present a procedure simpler than Pippard's, and two new theorems[3] needed to clarify deformation potentials. Parts of this program were accomplished by Blount[4], but our method is simpler and more complete. Finally we present the first calculations for the entire regime of $Q\ell$ for a non-free-electron metal, Cu.

The impressed ultrasonic wave gives a spatially varying lattice displacement $u\exp[i(\underset{\sim}{Q}\cdot\underset{\sim}{r}-\omega t)]$ which drives the electron distribution $F_k(r,t)$ away from the equilibrium (Fermi-Dirac) distribution $f(\epsilon_k)$ by an amount $(-\partial f/\partial\epsilon_k)\phi_k$, where ϕ_k is like an electronic energy boost. In the quantum regime ϕ_k consists of electron-hole pair excitations created directly by decay of ultrasonic phonons. In the hydrodynamic regime, electron scattering events permit only a small deviation from local equilibrium. The task is to find a Boltzmann rate equation which can be solved for ϕ_k, from which the attenuation can subsequently be derived. The general form of a Boltzmann equation is

$$\partial F_k/\partial t+\dot{\underset{\sim}{r}}\cdot\partial F_k/\partial\underset{\sim}{r}+\dot{\underset{\sim}{k}}\cdot\partial F_k/\partial\underset{\sim}{k}=[\partial F_k/\partial t]_{collision}. \quad (1)$$

The acceleration $\hbar\dot{\underset{\sim}{k}}$ equals the force experienced by an electron, which has two components. The local strain $\partial u_\alpha/\partial x_\beta=iQ_\alpha u_\beta(\underset{\sim}{x}t)=S_{\alpha\beta}$ causes an electron scattering matrix element $\langle k+Q|\underset{\sim}{u}\cdot\nabla V|k\rangle=D^{ep}_{\alpha\beta}(k)S_{\alpha\beta}$ which acts like a potential and whose gradient gives the longitudinal force on the electron k. The superscript "ep" on D distinguishes this "electron-phonon" deformation potential from a "band structure" deformation potential D^b needed later. There is in general also a transverse force $-e\underset{\sim}{E}$ where $\underset{\sim}{E}$ is $-(1/c)\partial\underset{\sim}{A}/\partial t$ and the vector potential $\underset{\sim}{A}$ has as its source the local ionic current flowing. Thus we have

$$\hbar\dot{k}=-iQ_\alpha[iQ_\beta D^{ep}_{\beta\gamma}(k)u_\gamma]-eE_\alpha. \quad (2)$$

The first two terms on the left hand side of the Boltzmann equation have their usual forms, so (1) becomes in linear approximation

$$(-\partial f/\partial\epsilon_k)[i(\underset{\sim}{Q}\cdot\underset{\sim}{v}_k-\omega)\phi_k-(\underset{\sim}{Q}\cdot\underset{\sim}{v}_k)(\underset{\sim}{Q}\cdot\underset{\sim}{D}^{ep}(k)\cdot\underset{\sim}{u})+e\underset{\sim}{v}_k\cdot\underset{\sim}{E}]=[dF_k/dt]_{scatt}. \quad (3)$$

The scattering term on the right hand side of (3) is greatly modified by the presence of the ultrasonic wave. Collision events will attempt to restore the distribution F_k to a local equilibrium which is at rest locally in the frame of the moving ions, and which has a Fermi surface altered in shape because of the local strain. The linearized form is

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$$[dF_k/dt]_{scatt}=-\sum_{k'}R_{kk'}\psi_{k'} \tag{4}$$

where $R_{kk'}$ is the usual scattering operator[5,6] containing elastic scattering by impurities and inelastic scattering by thermal phonons. The factor ψ_k measures the deviation from the local equilibrium,

$$F_k\equiv f(\varepsilon_{k-m\underset{\sim}{v}/\hbar}+D^{ep}_{\alpha\beta}(k)S_{\alpha\beta})+(-\partial f/\partial\varepsilon_k)\psi_k \tag{5}$$

where $\underset{\sim}{v}(\underset{\sim}{r}t)=-i\omega\underset{\sim}{u}$ is the local ion velocity and $S(\underset{\sim}{r},t)$ is the local strain. Taylor expansion of (5) allows ψ_k to be compared with the original deviation function ϕ_k

$$\psi_k=\phi_k+im\omega\underset{\sim}{u}\cdot\underset{\sim}{v}_k+iQ\cdot\underset{\sim}{\underset{\sim}{D}}^{ep}(k)\cdot\underset{\sim}{u}. \tag{6}$$

Although intuitively appealing, the results (4), (6) are not trivially obvious. In particular, one might expect intuitively that $\underset{\sim}{\underset{\sim}{D}}^{ep}$ in (5) and (6) should be the band deformation potential $\underset{\sim}{\underset{\sim}{D}}^{b}(k)$ which gives the energy shift of an electron state $d(\varepsilon_k-\mu)/d\underset{\sim}{\underset{\sim}{S}}$ in response to a static external strain. These two deformation potentials[3] are not the same, but are simply related by[3]

$$D^{ep}_{\alpha\beta}(k)=D^{b}_{\alpha\beta}(k)+mv_{k\alpha}v_{k\beta}. \tag{7}$$

We have been able to justify (4) and (6) microscopically by generalizations of a procedure used by Holstein[7] to justify the second term of (6) for the case of electron-phonon scattering. Derivations will be given in a subsequent paper[8].

The Boltzmann theory given in eqs.(3), (4) and (6) must be supplemented by an equation to determine $\underset{\sim}{E}$ and a prescription for calculating the attenuation. A self-consistent procedure determining $\underset{\sim}{E}$ is contained in the following equations.

$$\underset{\sim}{j}_{ra}+\underset{\sim}{j}_{er}+\underset{\sim}{j}_{ion}=0 \tag{8}$$

$$(\underset{\sim}{j}_{ra})=-(e/\Omega)\sum_k v_{k\alpha}\phi_k(-\partial f/\partial\varepsilon_k) \tag{9}$$

$$(j_{ion})_\alpha=+(ZeN/\Omega)(-i\omega u_\alpha) \tag{10}$$

$$(\underset{\sim}{j}_{er}+\underset{\sim}{j}_{ion})_\alpha=(m/m^*)_{\alpha\beta}(j_{ion})_\beta \tag{11}$$

$$=(e/\Omega)(-i\omega u_\beta)m\sum_k v_{k\alpha}v_{k\beta}(-\partial f/\partial\varepsilon_k) \tag{12}$$

Equation (8) is Pippard's condition which states that the $\underset{\sim}{E}$-field adjusts itself so that there is zero net local electrical current. The intraband electronic current and the ionic current are given by straightforward expressions (9), (10), where ZN/Ω is the number of electrons per unit volume, n. Holstein[7] noticed that the ultrasonic perturbation also drives electrical current via virtual interband transitions, and proved that this current partially cancels the ion current, leaving only a fraction (m/m^*) where $(m/m^*)_{\alpha\beta}$ is the effective mass tensor $\sum v_{k\alpha}v_{k\beta}(-\partial f/\partial\varepsilon_k)$. Combining (8), (9), and (12), the constraint which fixes the value of $\underset{\sim}{E}$ is

$$0=\sum_k v_{k\alpha}(\phi_k+im\omega\underset{\sim}{u}\cdot\underset{\sim}{v}_k)(-\partial f/\partial\varepsilon_k). \tag{13}$$

Finally we must calculate the attenuation, α. Following procedures of Ziman[5] we have shown that entropy is created irreversibly in collisions at a rate $(1/2)\sum_{kk'}R_{kk'}\psi^*_k\psi_{k'}$. Then α is

$$\alpha=\sum_{kk'}R_{kk'}\psi^*_k\psi_{k'}/v_S\rho_I\omega^2u^2 \tag{14}$$

where $v_S=\omega/Q$ is the sound velocity and ρ_I the ion mass density.

Equations (3), (4), (6), (7), (13) and (14) constitute a complete theory of ultrasonic attenuation including band structure and scattering effects. In general these equations must be solved on a computer. However, in the "constant τ approximation"

$$\sum_{k'}R_{kk'}\psi_{k'}\to(\psi_k-\langle\psi_k\rangle)/\tau\,(-\partial f/\partial\varepsilon) \tag{15}$$

analytic solutions can be found. These solutions exactly reproduce Pippard's results[2]. In order to express the answers in Pippard's form it is necessary to use the theorem[3]

$$\langle D^{ep}_{\alpha\beta}(k)\rangle\equiv\sum_k D^{ep}_{\alpha\beta}(k)\delta(\varepsilon_k)/N(0)=\langle mv_{k\alpha}v_{k\beta}\rangle \tag{16}$$

where N(0) is the density of states $\sum_k\delta(\varepsilon_k)$ and eq. (16) serves to define the average used in eq.(15) also. This theorem is derived in ref.3 from Galilean invariance considerations, but can be understood also from eq.(7) since $d\langle\varepsilon_k-\mu\rangle/d\underset{\sim}{\underset{\sim}{S}}$ is zero by definition. The deformation potential $D^p_{\alpha\beta}(k)$ used by Pippard is related to ours by $\hbar|\underset{\sim}{v}_k|\underset{\sim}{\underset{\sim}{D}}^p(k)=\underset{\sim}{\underset{\sim}{D}}^b(k)$. In the free electron limit, Pippard's answers reduce to his well known formulas[1], which have been rederived by graphical perturbation theory by various authors. Following procedures similar to Grünewald[9] (who considered longitudinal phonons in the free electron limit), we have also succeeded[8] in reproducing Pippard's results[2] for band electrons and general phonons, using Feynman-Dyson-Matsubara perturbation theory.

When the full microscopic scattering term, eq.(4), is used then the Boltzmann equation can only be solved numerically. The results of our computer calculation for the metal Cu are shown in Figs.1 and 2 and compared with the experimental results of MacFarlane and Rayne[10]. It should be noted that there are no free adjustable parameters in the calculations. The methods of this calculation are similar to the methods reported elsewhere[11,12]. The following ingredients were used: (i) a dense mesh of points on the Fermi surface (specifically 492 points in 1/48th of the Brillouin zone) and Korringa-Kohn-Rostoker wave functions calculated with $\ell\leq3$ spherical harmonics; (ii) phonon polarization vectors and frequencies obtained from Born-von Karman force constants fitted to inelastic neutron scattering data; (iii) electron-phonon matrix elements (in $R_{kk'}$) using the 'rigid muffin tin model' and; (iv) a value of $(1/\tau)_{imp}=2.2\times10^{10}s^{-1}$ quoted in ref.10, obtained from residual resistivity. To simplify the calculation we have made the approximation that the deformation potential

$D^{ep}_{\alpha\beta}(k)$ is independent of $\underset{\sim}{k}$ and have replaced it by its Fermi surface averaged value $m\langle v_{k\alpha} v_{k\beta}\rangle$. The systematic underestimation of the attenuation can be qualitatively explained as the effect of the neglected anisotropy of $D^{ep}_{\underset{\sim}{k}}(k)$. The general trends as a function of temperature and angle of propagation are nicely reproduced.

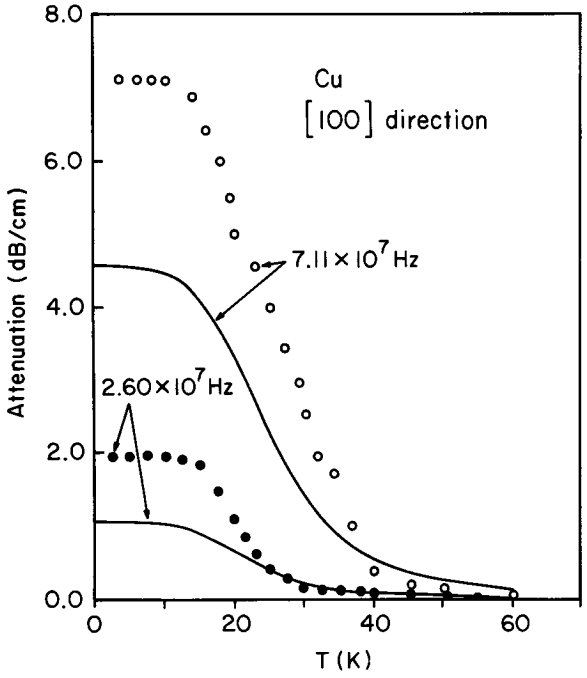

Fig. 1: Attenuation versus temperature for LA phonons in the [100] direction at two different frequencies. Impurity scattering was included in the theory (solid lines using $1/\tau = 2.2\times10^{10} s^{-1}$ in the "constant τ" approximation. This gives $Q\ell = 2.0$ and 5.4 for f=26MHz and 71.1MHz respectively, at T=0. At higher temperatures, $Q\ell$ becomes very small due to electron-phonon scattering. Data are from ref. 10.

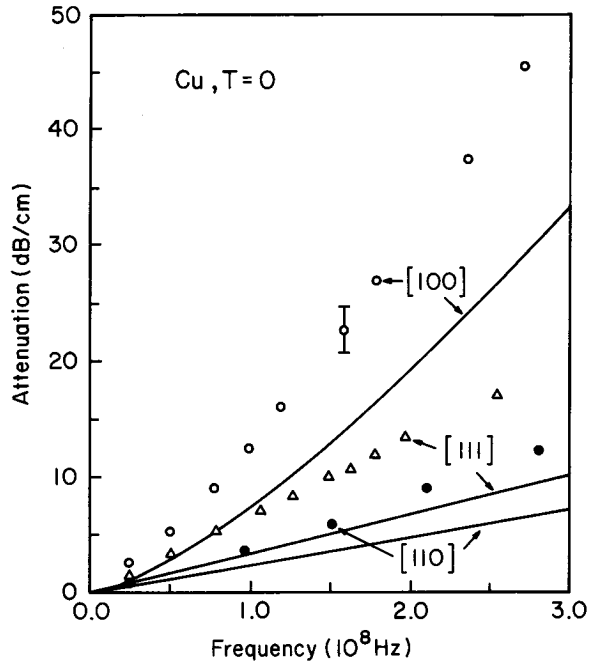

Fig. 2: Low temperature attenuation versus frequency for LA phonons in 3 crystallographic directions in Cu. Data are from ref. 10.

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REFERENCES

1. A.B. Pippard, Phil. Mag. 41, 1104 (1955).
2. A.B. Pippard, Proc. Roy. Soc. A257, 165 (1960); A.B. Pippard, The Dynamics of Conduction Electrons (Gordon and Breach, New York, 1965), pp.110-120.
3. F.S. Khan and P.B. Allen, Phys. Rev. B29, 3341 (1984).
4. E.I. Blount, Phys. Rev. 114, 418 (1959).
5. J.M. Ziman, Electrons and Phonons (Oxford Univ. Press, Oxford, 1960).
6. P.B. Allen, Phys. Rev. B17, 3725 (1978); see eqs. 30,32,33.
7. T. Holstein, Phys. Rev. 113, 479 (1959).
8. F.S. Khan and P.B. Allen, to be published. See also F.S. Khan, Ph.D. Thesis (Dept. of Electrical Engineering, S.U.N.Y., Stony Brook, 1983).
9. G. Grünewald, Z. Phys. 268, 197 (1974).
10. R.E. MacFarlane & J.A. Rayne, Phys. Rev. 162, 532 (1967).
11. W.H. Butler, J.J. Olson, J.S. Faulkner, and B.L. Gyorffy, Phys. Rev. B14, 3823 (1976).
12. W.H. Butler, F.J. Pinski, and P.B. Allen, Phys. Rev. B19, 3708 (1979).