determined by the analysis of tunneling currents. This technique allows the forbidden range of energies in the complex band structure of a solid to be probed directly.

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‡Howard Hughes Doctoral Fellow.

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FAR-INFRARED ABSORPTIVITY OF NORMAL LEAD*

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The Holstein-Boltzmann equation has been used to calculate the far-infrared absorptivity of normal Pb in and above the phonon-frequency region. The absorptivity exhibits a threshold at $\bar{\nu} \approx 35$ cm^{-1} and weak structure at ≈ 70 cm^{-1} related to the transverse and longitudinal peak in the phonon density of states, respectively. It increases rapidly through the phonon region, eventually saturating at the Holstein value. These results agree with the recent experiments of Joyce and Richards.

What happens when a high-frequency electromagnetic wave is incident on a pure metal at very low temperature? To the extent that the metal can be approximated by a free-electron gas there exists no mechanism for the loss of electromagnetic energy; the absorptivity is zero.[1] Some years ago, Holstein[2,3] showed that, even at $T = 0$ and for an infinite residual resistance ratio, there exist, in actuality, two dissipative mechanisms. One is due to the presence of a surface.[2] The electrons acquire an oscillatory energy from the electric field in passing through the skin depth δ_f; this energy is then converted to heat upon diffuse collision with the surface and subsequently dissipated in the interior of the metal. The other mechanism is purely a bulk process.[3] The electron simultaneously absorbs an incident photon and emits a phonon. From a quantum mechanical point of view the "free" conduction electrons are able to absorb the photon because collision with the surface or a phonon allows for the conservation of both energy and momentum. Holstein considered the high-frequency limit, i.e., $\omega \gg \omega_D$, a typical phonon frequency (e.g., for Pb, $\hbar\omega_D \approx 8.3$ meV), where the volume absorption process is independent of

frequency. In this range the excited electron-hole pair can interact with all the zero-point phonon modes, resulting in a constant effective collision time τ_{eff}. The aim of the present Letter is to extend the Holstein volume absorption (A_v) to lower frequencies through the phonon frequency region. In this regime it is not energetically possible for the excited electron-hole pair to interact with all the phonons; hence, $\tau_{\text{eff}}(\omega)$ and A_v become frequency dependent and naturally reflect the phonon density of states of the metal.

To calculate A_v, a perturbation expansion in the electron-phonon (EP) coupling can be used at high frequencies.[3] The result to lowest order in the EP coupling is $A_v = 2/\omega_p \tau_{\text{eff}}$, where $\omega_p = (4\pi ne^2/m_{\text{op}})^{1/2}$, the plasma frequency, m_{op} is the optical mass, n is the electron density, and τ_{eff} is simply related to the usual high-temperature resistivity relaxation time τ; for a Debye model, $\tau_{\text{eff}} = 5kT\tau/2\hbar\omega_D$. A measure of the EP coupling is $\hbar/\tau_{\text{eff}}$ itself[4] and in each order of the perturbation expansion one has $\hbar/\tau_{\text{eff}}$ divided by a typical energy denominator, $\hbar\omega$, the energy separation of the electron-hole pair. Thus, the perturbation parameter is $1/\omega\tau_{\text{eff}}$. For metals with strong EP coupling (e.g., Pb, Hg, Nb), $1/\tau_{\text{eff}} \approx \omega_D$ and as $\omega \to \omega_D$, $\omega\tau_{\text{eff}} \to 1$, i.e., the perturbation expansion is not valid; at lower frequencies one must include multiphonon processes. Holstein,[4] in his general analysis of transport phenomena in an electron-phonon system, has shown that one need only consider two basic types of multiphonon contributions to the conductivity (for zeroth order in c_s/v_F, where c_s is the velocity of sound and v_F is the Fermi velocity): (1) a sequence of single phonon exchanges between the electron-hole pair, the so-called ladder diagrams (cf. Fig. 6, Ref. 4), (2) a series of single phonon emissions and reabsorptions by the electron (hole), i.e., the self-energy parts. The summation of all the latter processes necessitates the use of the exact propagator[4] for the contribution of each electron/hole (solid) line to the diagrams in Fig. 6, Ref. 4. It is the rapid variation with energy of the self-energy part contained in the propagator, in the range $0 \leq |\epsilon - \epsilon_F| \leq \hbar\omega_D$, that leads to the above-discussed frequency dependence of $\tau_{\text{eff}}(\omega)$. Strictly speaking, it is the imaginary or dissipative part of the self-energy which contributes to $\tau_{\text{eff}}(\omega)$. The real part of the self-energy gives rise to frequency-dependent dispersive effects. These effects are included (in a natural way) in the present calculation. However, the characteristic features of the bulk absorption are dominated by the dissipative real collisions of electrons with phonons. The dispersive effects would be most clearly manifested in an experiment which is sensitive to the shift in the electron energy levels as a function of energy, such as cyclotron resonance.[5]

Summation of all the ladder diagrams is equivalent to the solution of the following transport equation[4] ($T = 0$):

$$i(\vec{q}\cdot\vec{v}_k - \omega)\Phi_k = v_{ky} + (\pi/\hbar)\sum_{k',\pm} |V_{kk'}|^2(\Phi_{k'} - \Phi_k)\{\delta(\epsilon - \epsilon' \pm \hbar\omega_{kk'})[f^{(\mp)}(\epsilon' + \hbar\omega) + f^{(\mp)}(\epsilon')] + (i/\pi)\mathrm{P}(\epsilon - \epsilon' \pm \hbar\omega_{kk'})^{-1}[f^{(\mp)}(\epsilon' + \hbar\omega) - f^{(\mp)}(\epsilon')]\}, \quad (1)$$

and a subsequent evaluation of the conductivity,

$$\sigma_{xy} = (2e^2/\Omega)\textstyle\sum_k v_{kx}\Phi_k[f^{(-)}(\epsilon) - f^{(-)}(\epsilon + \hbar\omega)]/\hbar\omega. \quad (2)$$

The Holstein-Boltzmann equation, (1), reduces to the usual linearized Boltzmann equation for the dc and extreme anomalous skin-effect limits.[4] The left-hand side of (1) contains the Fourier transform of the space and time derivatives of the "distribution function" per unit field, Φ_k; on the right-hand side the electron velocity v_{ky} is the driving term (the field is taken to be in the y direction) and the second term can be described as a quantal modification of the difference between the "scattering-in" and "scattering-out" contributions familiar to transport theory. $|V_{kk'}|^2$ is the electron-phonon matrix element and the delta-function (principal-value) term represents real (virtual) effects associated with this interaction. The term proportional to Φ_k can be rewritten[4] as

$$-\Phi_k\{\Gamma(\epsilon) + \Gamma(\epsilon + \hbar\omega) + i[M(\epsilon + \hbar\omega) - M(\epsilon)]\}/\hbar, \quad (3)$$

where $M(\epsilon)$ [$\Gamma(\epsilon)$] is the real [imaginary] part of the electron self-energy; expressed in this form (3) is similar to the usual attenuation term of the Boltzmann equation with $\Gamma(\epsilon)/\hbar$ playing the role of an energy-dependent collision frequency. The self-energy is of central importance in the calculation and fortunately has been determined with great accuracy for Pb[6] (corrections are of order c_s/v_F).

The self-energy is related to $\alpha^2(\bar{\nu})F(\bar{\nu})$,[6,7] which is obtained numerically from tunneling data, where $\alpha^2(\bar{\nu})$ is the square of the EP coupling and $F(\bar{\nu})$ is the phonon density of states as a function of energy ν (cm^{-1}).[8]

As a first consideration in the solution of (1) we shall set the wave vector[1] $q=0$. After solving (1) in this limit, we shall use the solution in a Boltzmann equation and include the nonlocal aspects ($q\neq 0$). We make the usual Ansatz $\Phi_k = v_{ky}\mathcal{F}(\epsilon)$. Now, (1) and (2) are simplified to

$$-i\omega\mathcal{F}(\epsilon) = 1 + 2\pi/\hbar\sum_{k',\pm}|V_{kk'}|^2[(\hat{v}_k\cdot\hat{v}_{k'}/V_k^2)\mathcal{F}(\epsilon')-\mathcal{F}(\epsilon)]\{\cdots\}, \tag{4}$$

$$4\pi\sigma(\omega)/\omega = (\omega_p/\omega)^2\int_{\epsilon_F-\hbar\omega}^{\epsilon_F} d\epsilon\mathcal{F}(\epsilon)/\hbar, \tag{5}$$

where (5) holds for a cubic material and ω_p is suitably averaged over the frequency spectrum.

For $\omega\approx\omega_D$ the integral equation (4) contains no obvious smallness parameter. However, a typical electron-phonon collision involves a large-angle scattering and thus the coefficient of $\mathcal{F}(\epsilon)$, $\hat{v}_{k'}\cdot\hat{v}_k/v_k^2$, is an effective small parameter. (4) was solved numerically by an iteration in this parameter and the iteration scheme was analyzed in detail for an Einstein model. The expansion parameter, for $\omega\approx\omega_D$, is $\frac{1}{2}\gamma'/\gamma$, where $\gamma'\equiv\langle v_Q^2(1-Q^2/2k_F^2)\rangle$, $\gamma\equiv\langle v_Q^2\rangle$, and

$$\langle v_Q^2\rangle \equiv \int_0^{2k_F} Q^3(v_Q^2)dQ \Big/ \int_0^{2k_F} Q^3 v_0^2 dQ, \tag{6}$$

a normalized average of the dressed electron-ion pseudopotential. The effective expansion parameter decreases with increasing frequency and the iteration tends to the Holstein solution at high frequency. The values of γ', γ for Pb have been determined as follows: γ is proportional to the first moment of $\alpha^2(\bar{\nu})F(\bar{\nu})$,[6,9] and the value so obtained is $\gamma=0.037$. This agrees quite well with the value calculated[6] from the Heine-Abarenkov (HA)[10] model pseudopotential, $\gamma_{HA}=0.038$. Using the HA pseudopotential, McMillan[11] has numerically determined $\gamma-\gamma'=0.0213$. Thus, $[\gamma'/\gamma]_{Pb}=0.44$.

An "interpolation solution" has been shown to agree within 2% of the second iteration solution of the Einstein model over the entire frequency range of interest. This interpolation solution essentially accounts for the effect of the "scattering-in" term by scaling the term in (3) with the factor $(\gamma-\gamma')/\gamma$. Using the above values of γ, γ' and the self-energy[6] for Pb, the normalized bulk conductivity, $\sigma_1+i\sigma_2\equiv(\omega/\omega_p)^2 4\pi\sigma(\omega)/\omega$, was computed and is shown in Fig. 1. Although the local limit is strictly not valid until $\bar{\nu}>240\ \mathrm{cm}^{-1}$ (as will be indicated), it is instructive to discuss $\sigma_{1,2}$ to elucidate the phonon effects; also $\sigma_{1,2}$ will be utilized in the nonlocal calculation below.

The transverse and longitudinal peaks in $\alpha^2(\bar{\nu})F(\bar{\nu})$ at $\bar{\nu}=35$ and $67\ \mathrm{cm}^{-1}$ are reflected in the two dips (at 45 and $77\ \mathrm{cm}^{-1}$) in σ_2. The same peaks are exhibited in σ_1 by a threshold at $\bar{\nu}\approx 35\ \mathrm{cm}^{-1}$ and a weak knee at $\bar{\nu}\approx 70\ \mathrm{cm}^{-1}$. These dominant features of $\alpha^2(\bar{\nu})F(\bar{\nu})$ are somewhat washed out in $\sigma_{1,2}$ by two energy integrations: the first for the self-energy and the second for the conductivity.

The function $\mathcal{F}(\epsilon)$ varies slowly with energy at both high ($\omega\gg\omega_D$) and low ($\omega\ll\omega_D$) frequency. In these limits some insight can be gained by using the Drude form for the conductivity,

$$\sigma_1+i\sigma_2 = [\omega\tau_{eff}+i(\omega\tau_{eff})^2]/[1+(\omega\tau_{eff})^2]. \tag{7}$$

At high frequency ($\omega\tau_{eff}\gg 1$), $\sigma_2\sim 1-(\omega\tau_{eff})^{-2}$, $\sigma_1\sim(\omega\tau_{eff})^{-1}$ as indicated in Fig. 1; from these limiting values one obtains the constant (Holstein) value for $\tau_{eff}(\mathrm{Pb})=4.7\times10^{-14}$ sec. This is numerically accurate to the extent that the ratio[12] $\langle Q^2 v_Q^2/\bar{\omega}_Q\rangle/\langle v_Q^2/\bar{\omega}_Q\rangle$ is equal to the scale factor $\langle Q^2 v_Q^2\rangle/\langle v_Q^2\rangle$. Since $Q^2 v_Q^2$ is strongly peaked[10] at $Q/2k_F=0.4$, in a region where the angular average of the phonon spectrum $\bar{\omega}_Q$ is relatively flat,[13] the ratios are expected to be equal within $\approx 10\%$ (see below). At low frequency $[\omega\tau_{eff}(\omega)]^{-1}$ rapidly goes to zero, so that σ_1 also is $\sim[\omega\bar{\tau}_{eff}(\omega)]^{-1}\propto\omega^2$; in a more exact calculation it is $\propto\omega^4$. For σ_2, however, the correspond-

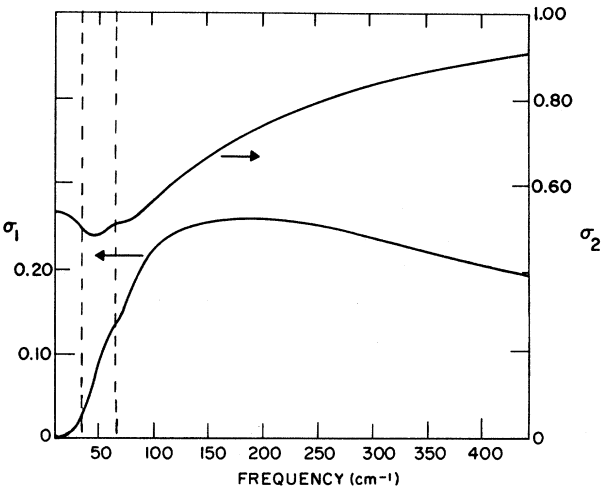

FIG. 1. The ($q=0$) normalized real (σ_1) and imaginary (σ_2) parts of the bulk conductivity of Pb. Dashed lines at 35, $67\ \mathrm{cm}^{-1}$.

ing Drude form (7) must be modified by the low-frequency limit of the real part of the self-energy. The effect is to change the mass in the normalizing factor ω_p and thus $\sigma_2 \rightarrow m_{op}/m^{**}$ as $\omega \ll \omega_D$. From Fig. 1, $m^{**}/m_{op} = 1.87$; m^{**} (cf. Sect. V.C. of Ref. 4) differs from the specific heat and low-frequency cyclotron-resonance mass m^* (for Pb, the theoretical[6] $m^*/m_{op} = 2.54$) by the "scattering-in" contribution. The value obtained for m^{**} is accurate to the extent that the $\langle Q^2 v_Q{}^2/\bar{\omega}_Q{}^2\rangle/\langle v_Q{}^2/\bar{\omega}_Q{}^2\rangle$ is equal to $\langle Q^2 v_Q{}^2\rangle/\langle v_Q{}^2\rangle$. The argument for this approximate equality is the same as above.[14] The effective mass m^{**} increases[15] with frequency (as shown in Fig. 1) until the threshold of σ_1.

In calculating the absorptivity A we must now consider the nonlocal aspects ($q \neq 0$). We adopt a procedure similar to the one used in Sect. IV of Ref. 5 and we assume that the technique used above to handle the "scattering-in" term is valid to a good approximation with a finite $\bar{q}$. The result is to reproduce the Reuter-Sondheimer (RS)[16] solution (specular case) for the surface impedance Z [$A = (c/\pi)\mathrm{Re}Z$],

$$Z = -(8i\omega/c^2)\int_0^\infty dq/[q^2 - \tfrac{3}{4}\delta_f{}^{-2}\kappa(q)] \qquad (8)$$

with, however,

$$\kappa(q) = \int_{\epsilon_F - \hbar\omega}^{\epsilon_F} K(qv_F/\Omega(\epsilon))\,d\epsilon/\hbar\Omega(\epsilon), \qquad (9)$$

$\hbar\Omega(\epsilon) \equiv -\hbar\omega - (1-\gamma'/\gamma)\{M(\epsilon) - M(\epsilon+\hbar\omega) + i[\Gamma(\epsilon) + \Gamma(\epsilon+\hbar\omega)]\}$, $\delta_f = c/\omega_p$, and $K(s)$ defined in (A10), Ref. 16. An analogous result holds for the diffuse case. The main consideration, now, is whether the anomalous (nonlocal) or local limit, for (8), is applicable in the phonon frequency region. This criterion can be most clearly determined in the series expansion of the surface impedance (6.1) to (6.6) in Dingle.[2,17] For this purpose the series parameter can be written as $\xi = \frac{3}{4}(v_F/\delta_f\omega)^2[1 + i/\omega\tau_{eff}(\omega)]^{-3}$. The anomalous limit obtains for $|\xi| > 1$ and the local limit for $|\xi| < 1$. Since $[\omega\tau_{eff}(\omega)]_{min} \approx 1$, the appropriate limit is determined simply by $v_F/\delta_f\omega$. The band parameters for Pb are obtained from the Chambers[18] value of σ/l, which is equal to 8.46×10^{22} (cm sec)$^{-1}$ in Gaussian units. From this value, assuming a spherical Fermi surface for Pb, we obtain $v_F = 1.24\times10^8$ cm/sec and the effective $\hbar\omega_p = 7.43$ eV ($\omega_p{}^2/4\pi v_F = \sigma/l$); thus, for $\hbar\omega > 30$ meV (or 240 cm^{-1}) the local limit is applicable. For the phonon regime we use the large-q asymptotic expansion of (9) and retaining the two leading terms we have

$$A = \frac{8\sqrt{3}}{9}\left(\frac{4\omega^2 v_F}{3\pi\omega_p{}^2 C}\right)^{1/3} + \frac{8\sqrt{3}}{9}\left(\frac{16c}{3\pi v_F}\right) \times \left(\frac{4\omega^2 v_F}{3\pi\omega_p{}^2 C}\right)^{2/3} \mathrm{Re}\left\{\frac{i\bar{\Omega}e^{-i\pi/6}}{\omega}\right\}, \qquad (10)$$

where $\bar{\Omega} \equiv \int_{\epsilon_F - \hbar\omega}^{\epsilon_F} d\epsilon\,\Omega(\epsilon)/\omega$. As a convenience and to an excellent approximation we replace $i\bar{\Omega}/\omega$ in (10) by $(\sigma_1 + i\sigma_2)^{-1}$. The results, using (10), are shown in Fig. 2. The main features of the theoretical (solid line) absorptivity are (1) a point of inflection and threshold at $\nu \approx 30$ cm^{-1}, (2) a rapid rise in A due to Γ [or $\hbar/\tau_{eff}(\omega)$] above 30 cm^{-1}, (3) a very weak knee at $\nu \approx 70$ cm^{-1}. Below $\nu \approx 30$ cm^{-1} the absorptivity is due to the anomalous skin effect (ASE) with, however, phonon renormalization effects. In this frequency region,[19] the curly brackets in (10) are replaced by $-m^{**}(\omega)/2m_{op}$, which is equal to 1.0 at $\nu = 30$ cm^{-1} and is 1.87/2 at 5 cm^{-1}. The dashed line in Fig. 2 represents the results of (10) with $\Gamma = 0$. In the sense of the Dingle series in (10) A_v and the ASE absorption are additive (over the frequency range considered in Fig. 2) but with the important renormalization effects included in the latter. Also shown in Fig. 2 are the experimental results of Joyce and Richards[20] shifted slightly to fit the theoretical curve in the threshold region. The agreement is satisfactory; the differences can be attributed to (1) experi-

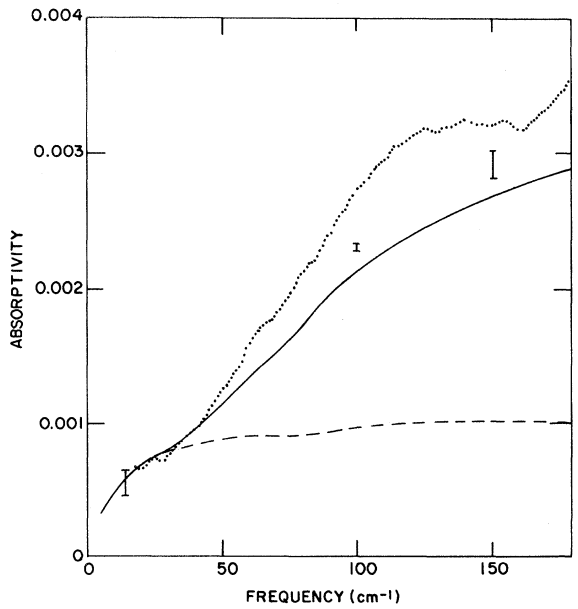

FIG. 2. The theoretical (solid line) and experimental (dotted line) absorptivity of Pb.

mental error (quoted in Ref. 20 and included in Fig. 2), (2) the next-higher term of the Dingle series—a careful estimate yields positive corrections of 6% at 100 cm^{-1} and 13% at 180 cm^{-1}, (3) a small ($\lesssim 10\%$) q-dependent correction to the $1-\gamma'/\gamma$ factor, (4) variations in the choice of the band parameters, especially v_F.

In the near-infrared and optical region the volume and surface absorptivity are additive.[2] At the higher frequencies $A_v \to 2/\omega_p \tau$ (in the local limit) and for $\hbar\omega_p = 14$ eV, $A_v = 0.002$; thus, assuming diffuse scattering, the absorptivity saturates at 0.005.

As it turns out, the HB integral equation (1) need only be solved (as such) for a strong-coupling EP metal with $v_F/\delta_f \omega_D \approx 1$. For Pb in the phonon regime (1) can be solved by iteration[21] in $(\vec{q}\cdot\vec{v}_k)^{-1}$ (assuming the specular case): $\Phi = \Phi^{(0)} + \Phi^{(1)}$,[21] where $\Phi^{(0)} = v_{ky}/i\vec{q}\cdot\vec{v}_k$, etc. Equivalently, in terms of diagrams,[4] to obtain A in the form of (10) one need only consider the two lowest-order ladder diagrams (zero- and one-rung ladders, but with exact electron/hole propagators). The contributions of the two diagrams to $\sigma(\omega)$ are then expanded in $(\vec{q}\cdot\vec{v}_k)^{-1}$, retaining the first two terms (each rung brings an additional $\Gamma/\hbar\vec{q}\cdot\vec{v}_k$). The statement (made above and in footnote 89 in Ref. 4) that (1) reduces to the Boltzmann equation in the ASE limit holds in the "nonrelaxation" region ($\omega\tau \ll 1$, at finite temperature), where ω can essentially be set euqal to zero. We are considering $\omega\tau_{\text{eff}} \gtrsim 1$, so there are ω-dependent phonon effects in the ASE limit, but note they first appear in the higher-order (q^{-2}) term.

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*The work reported here was initiated while the author was a member of the technical staff at Bell Telephone Laboratories, Murray Hill, N. J. A preliminary version of this work is reported in H. Scher, Bull. Amer. Phys. Soc. 12, 672 (1967).

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