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Phonon Contribution to the Far Infrared Absorptivity of Superconducting and Normal Lead

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ABSTRACT

Measurements of the far infrared absorptivity of superconducting and normal Pb show structure due to the Holstein phonon generation process. This structure is compared with a phenomenological theory including the generation of a real acoustic phonon as the photon scatters a normal electron or breaks a Cooper pair, and to a non-local calculation of the surface resistance from strong coupling superconductivity theory.

We have used the techniques of far infrared spectroscopy to measure the absorptivity of superconducting and normal lead throughout the region of the phonon frequencies. Our initial experiments[1] in the range from 15 cm^{-1} to 200 cm^{-1} provided the first example of structure on the absorptivity of a metal due to the direct excitation of phonons. We used a "Golden Rule" treatment of the Holstein[2] phonon generation process to account for most of the features of our experimental data.

This report describes further investigations of the Holstein effect in lead. We have extended our measurements to a frequency of 350 cm^{-1}, which is sufficiently greater than the energy gap ($2\Delta \approx 22$ cm^{-1}) and the highest phonon frequencies (80 cm^{-1}) that we expect the optical properties of the superconductor and normal metal to be identical. We have used

Allen's theoretical treatment[3] of the Holstein process in the superconducting state to obtain an improved fit of the Golden Rule calculation to our data. Finally, we have compared our data to Shaw and Swihart's nonlocal calculations[4] of the surface resistance of lead for frequencies up to 100 cm^{-1}. The quantitative agreement found shows that the complex conductivities calculated from strong-coupling superconductivity theory can explain both real and virtual phonon structure on the surface resistance.

We measured the absorptivity of our samples by a calorimetric method. A chemically polished single crystal sample of pure lead ($R_{300}/R_{1.2} \simeq 85000$) was mounted in a highly absorbing cavity and illuminated with radiation from a far-infrared Michelson interferometer. A doped germanium thermometer cemented to the back of the sample measured the temperature rise due to radiation absorption. The experiments done at 1.2K on samples in the superconducting state were compared to experiments on normal state lead done in a magnetic field of ~1200 Oe applied parallel to the sample surface. The data were analyzed by dividing the superconducting absorption spectrum by that of the normal metal, since structure due to the background and resonant modes of the sample cavity should cancel out of this ratio.

Our experimental ratio of superconducting to normal absorption is shown by the points in Fig. 1. The ratio rises rapidly from the energy gap at $2\Delta=22$ cm^{-1} to a maximum at 35 cm^{-1}. The decrease from 35 cm^{-1} to 55 cm^{-1} results from the increase in absorptivity of the normal metal due to phonon creation. The ratio then rises in two steps, centered at ~65 cm^{-1} and ~95 cm^{-1}, which are due to the onset of absorption in the superconductor. In addition, the ratio gradually decreases from 120 cm^{-1} to 240 cm^{-1}, remaining constant to the upper frequency limit (350 cm^{-1}) of our ex-

periment. The scale factor of the data in Fig. 1 was established by setting the value of A_S/A_N at high frequencies to its expected value of unity.

The Holstein process involves the scattering of a conduction electron which absorbs a photon and emits a phonon. We were able to use a simple phenomenological theory to calculate the transition probability for this process and explain the major features of our data. The initial state consists of a photon of energy ω and a filled Fermi sea of conduction electrons at T=0. The final state is an electron of energy ε_1, a hole of energy ε_2, and a phonon of energy Ω. The density of final states is assumed to be the convolution of the independent densities of states of the electron (N_e), hole (N_h), and phonon (F). The Holstein volume absorptivity is then proportional to

$$P_v \propto \omega^{-1} \int N_e(\varepsilon_1) N_h(\varepsilon_2) \alpha^2(\Omega) F(\Omega) \delta(\varepsilon_1+\varepsilon_2+\Omega-\omega) d\varepsilon_1 d\varepsilon_2 d\Omega. \qquad (1)$$

The function $\alpha^2(\Omega)$ is the square of the electron-phonon matrix element.

The Holstein absorptivity in the normal metal is obtained by substitution of a constant N_o for the electron and hole densities of states, while the expression for the superconductor is obtained by using a BCS density of states for N_e and N_h and including the effects of superconducting coherence on the electron states. The integrals were evaluated using Rowell and McMillan's values[5] for $\alpha^2F(\Omega)$, modified by Allen[3] to include the anisotropic coupling of the electrons to the longitudinal phonons. Composite spectra for both the superconducting and normal states were then calculated by adding the Holstein absorptivity P_v to the anomalous skin effect absorptivity P_s. P_s in the normal metal was obtained from Dingle's tables[6] of surface resistance, using the electronic mean free path obtained from our resistivity measurements and Chambers' value[7] $\sigma/\ell=9.4\times10^{10}\ \Omega^{-1}\ cm^{-2}$. The

superconducting skin effect absorptivity was calculated in the local limit using values of $\sigma_1(\omega)$ and $\sigma_2(\omega)$ derived from strong-coupling superconductivity theory for Pb.[8] The ratio of P_v to P_s was estimated from Scher's calculations[9] to be 0.8, quite close to the value $P_v/P_s=1$ which gave the best fit to our data.

The theoretical ratio A_S/A_N calculated in this manner is shown by the solid curve in Fig. 1. The shape of this curve clearly accounts for the major features of the data.

A better fit to our data is obtained from a computation of the non-local conductivities $\sigma_1(\underset{\sim}{k},\omega)$ and $\sigma_2(\underset{\sim}{k},\omega)$ from strong-coupling superconductivity theory, and an integration over the allowed values of $\underset{\sim}{k}$ for each ω. Shaw and Swihart[4] have calculated R_S/R_N in this manner for several frequencies up to 95 cm^{-1}. Fig. 2 shows our data (curve A) compared to their calculations for diffuse (curve B) and specular (curve C) boundary conditions for the electrons. No adjustable parameters are used in the fit of the theory to our data. Both theoretical curves accurately reproduce the steepness of the absorption edge above the energy gap and show the Holstein effect structure at the correct frequencies. The accuracy of the fit raises the possibility of using the infrared absorptivity to obtain information on the phonon density of states in materials which cannot be easily investigated by the standard method of superconducting tunneling.

Calculations of A_S/A_N from either the local or the extreme anomalous limit of the strong coupling theory show no significant phonon structure. Preliminary results from a series of alloys of PbBi and PbIn confirm the disappearance of the phonon structure when the electronic mean-free-path is less than the skin depth. The absence of phonon structure on the thin

film data of Palmer and Tinkham[10] can be understood in this way.

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FIGURE CAPTIONS

Fig. 1. Ratio of experimental absorptivity in superconducting and normal states compared with phenomenological theory.

Fig. 2. Experimental absorptivity ratio (curve A) compared with non-local strong coupling theory assuming diffuse (curve B) and specular (curve C) boundary conditions for electrons.

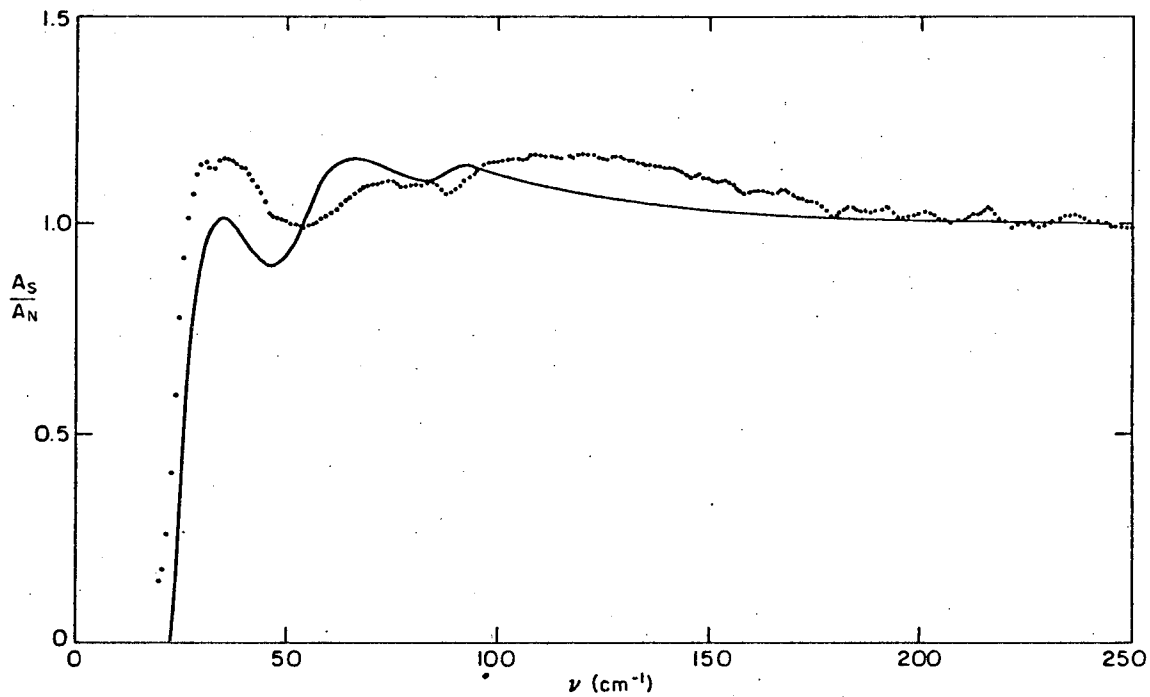

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Fig. 1

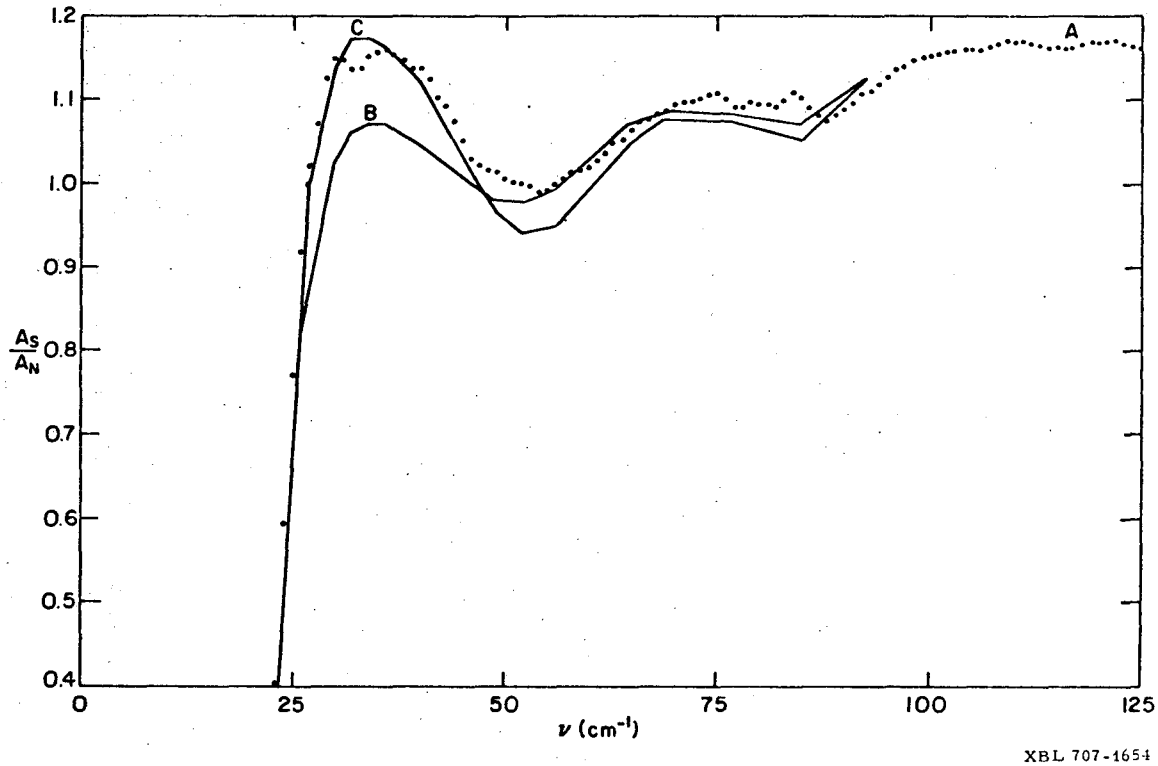
1.2
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.4
$\frac{A_S}{A_N}$
0
25
50
75
100
125
ν (cm^{-1})
A
B
C
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Fig. 2

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