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Electron-Tunneling Studies of Dilute Pb-Based In Alloys*

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The technique of electron tunneling has been used to obtain the effective phonon spectra, $\alpha^2(w)F(w)$, and the parameters defining the superconductivity for dilute Pb-In alloys by solving the Eliashberg equations. These quantities were extracted from the measured energy gap of the alloy films and the measured normalized conductance of the thin-film tunnel junctions of the form Al–Al_2O_3–Pb-In. All the measurements were carried out around 1 K, at which temperature both the aluminum and the alloy films were in the superconducting state. The normal-state data were taken by raising the temperature above the transition temperature of both the films. A band of frequencies, the so-called impurity band, appeared beyond the high-frequency cutoff of the phonon spectrum of pure Pb. For all the alloys studied the impurity band was found to consist of at least two peaks which are attributed to the vibrations of isolated indium atoms and the vibrations of the pairs of indium atoms, both surrounded by the host lead atoms. The position of the first peak has been found to be constant (9.57 ± 0.03 meV), and its width has been found to vary linearly with concentration of indium. The position of the second peak remains constant up to 2-at.% indium but increases with the further addition of indium. The width of the second peak also varies linearly with the indium concentration. The fraction of modes in the impurity band has been found to be a factor 1.5–2.0 less than the concentration of indium. A determination of the ratio of the energy gap to the superconducting transition temperature for the alloy films shows that the electron-phonon interaction remains nearly constant for all the alloys studied.

I. INTRODUCTION

The problem of the disordered lattice has been of considerable interest for some time. This is due to the large effects of imperfections on the properties of the solids and to the fact that imperfections are always present. Although a number of theoretical methods have been developed for the study of the disordered lattice, the problem still remains, at least in regard to the phonon spectrum, not solved rigorously. The most recent extensive work on such phonon spectra has been done by Dean[1] on one- and two-dimensional disordered lattices and by Payton and Visscher[2] on one-, two-, and three-dimensional lattices. All these calculations are for limited-size lattice only. The central result of these calculations is that when light impurities are added to the heavy host lattice, a number of peaks appear beyond the high-frequency cutoff of the phonon spectrum of the host lattice. Dean has attributed these peaks to the vibrational modes of impurity clusters of different

sizes. At low concentrations of the light impurities, there is a main peak believed to be caused by the vibrational modes of the isolated impurity atoms and other smaller peaks placed symmetrically around the main peak. The smaller peaks have been attributed to the vibrations of higher-order impurity clusters. Elliott and Taylor[3] have studied the problem of the vibrational properties of a crystal with a randomly arranged small concentration of impurities by a thermal Green's-function technique. The Green's functions lead directly to average and weighted displacement and momentum correlations which can be related to observable properties such as the phonon density of states and thermal energies. Their method treats correctly multiple interactions of a vibrational quantum with a single defect, but treats interference between defects only approximately. Their method is thus correct to first order of impurity concentration but only approximately correct to higher orders. We shall refer to their results later. Taylor[4] has extended the analysis and has attempted to describe the vibrations of lattices with high defect concentrations. As far as the localized vibrational modes are concerned, the results of calculations by Elliott and Taylor[3] are only partly in agreement with the experimental results.

The situation on the experimental side is not very good either. Until about seven years ago, the only satisfactory method for the study of the phonon spectrum was the method of inelastic neutron scattering. Because of the complexity of the method, most of the work on phonon spectra using inelastic neutron scattering has been done in pure solids only. In recent years the method of electron tunneling into a superconductor has become convenient to study the phonon spectrum of strong-coupling and intermediate-coupling superconductors. Rowell and McMillan[5] were the first to use this method for a detailed study of the phonon spectrum of pure lead, which was also used as a test case for the method of electron tunneling as a tool for phonon-spectrum studies. From the measured ratio of the tunneling conductances in the superconducting and normal state, which is proportional to the ratio of the electron densities of states, Rowell and McMillan calculated the gap parameter and the phonon spectrum for lead. To be more specific, the method of electron tunneling gives the product $\alpha^2(w)F(w)$, where $\alpha^2(w)$ is the electron-phonon coupling function and $F(w)$ is the phonon density of states. The results obtained by Rowell and McMillan for the effective phonon spectrum, $\alpha^2(w)F(w)$, for Pb are in very good agreement with the results on the phonon spectrum by inelastic neutron scattering. In the last few years the method of electron tunneling has been used for the phonon-spectrum studies of many strong-coupling and intermediate-coupling superconductors and also for some superconducting alloys and intermetallic compounds.

The main aim of the present work is to make use of the method of electron tunneling into superconductors to investigate the phonon spectrum of a disordered system and to quantitatively study the effects of increasing the disorder. The alloys studied in the present investigations are the dilute Pb-based In alloys. This alloy system was chosen because of (a) strong electron-phonon coupling, (b) easy miscibility of In in Pb, (c) relatively large mass ratio of the components, and finally (d) because of the fact that the existence of the impurity band has been shown for one dilute alloy of this system.[6] The technique of electron tunneling provides information about the gap parameter, the renormalization constant, and, of course, the effective phonon spectra.

In Sec. II we give a brief outline of the relevant theoretical results which have been used to obtain the phonon spectrum from the measured normalized conductance. The experimental techniques are discussed in Sec. III. The experimental results and their discussion are given in Sec. IV. In Sec. V we give the conclusions drawn from the present work.

II. THEORY

The integral equations for normal and pairing self-energies (Eliashberg equations) of a dirty superconductor are[7]

$$\xi(w)=[1-Z(w)]w=\int_{\Delta_0}^{w_C} dw'\,\mathrm{Re}\,\frac{w'}{[w'^2-\Delta'^2(w')]^{1/2}} \times\int dw_q\,\alpha^2(w_q)F(w_q)\,[D_q(w'+w)-D_q(w'-w)]\,, \quad (1)$$

$$\phi(w)=\Delta(w)Z(w)=\int_{\Delta_0}^{w_c} dw'\,\mathrm{Re}\,\frac{\Delta'(w')}{[w'^2-\Delta'^2(w')]^{1/2}} \times\{\int dw_q\,\alpha^2(w_q)F(w_q) \times[D_q(w'+w)+D_q(w'-w)]-U_c\}, \quad (2)$$

where $D_q(w)=(w+w_q-i0^+)^{-1}$, $\Delta(w)$ is the energy-dependent energy gap, $\Delta_0=\Delta(\Delta_0)$, $F(w_q)$ is the phonon density of states, and $\alpha^2(w_q)$ is an effective electron-phonon-coupling function for phonons of energy w_q. In Eq. (2) U_c is the Coulomb pseudopotential given approximately by

$$U_c=\frac{N(0)V_c}{1+N_0V_c\ln(E_F/w_c)}\,, \quad (3)$$

where V_c is the static screened-Coulomb interaction averaged over the Fermi surface, $N(0)$ is the electronic density of states at the Fermi surface unrenormalized by the electron-phonon interac-

tion, and $w_c \simeq 5w_0$, where w_0 is the maximum phonon frequency.

The tunneling density of states in a superconductor is given by[7]

$$\frac{N_s(w)}{N(0)} = \mathrm{Re}\ \frac{|w|}{[w^2 - \Delta^2(w)]^{1/2}} \qquad (4)$$

and is directly measured by the ratio of the superconducting and normal conductances of a superconductor–normal-metal junction at $T = 0$ K. It is also possible to compute the tunneling density of states from the characteristics of a superconductor-superconductor junction at finite temperature, as was done in our case. One can then by an iterative procedure, described in detail by Rowell and McMillan,[5] obtain $\alpha^2(w)F(w)$ and U_c, which give the best fit between the experimentally measured density of states and the one computed from Eqs. (1), (2), and (4).

III. EXPERIMENTS

The production of thin-film tunnel junctions of the form Al–Al_2O_3–metal is a very standard technique and has been discussed extensively.[5] We will not give the details of junction preparation but discuss only some of the important points. The tunnel junctions used were of the form Al–Al_2O_3–Pb-In and were evaporated onto glass substrates at room temperature at pressure less than 10^{-5} Torr. The alloy films were prepared by the flash evaporation technique in which small pellets of the previously prepared alloy were individually dropped into a resistively heated Ta boat kept at a temperature that was higher than the boiling point of the alloy. Each pellet was evaporated completely before the next one was dropped. This method of evaporation ensured the homogeneity of the alloy film. The thickness of the alloy film was about 1000 Å which, as it should be, was greater than the phonon-emission mean free path of the tunneling electrons. All the measurements were carried out around 1 K, at which temperature both the aluminum and the alloy films were in the superconducting state. The tunneling characteristics both in the superconducting and normal states were measured by a standard modulation technique which combined harmonic detection with the bridge technique.[8] The system used was capable of determing the normalized conductance of the junction to within a few parts in 10^5. The superconducting energy gaps in aluminum and the alloys were also measured from the I-V characteristics. We observed a single and sharp energy gap for all the alloys studied which gave further support to the assumed homogeneity of the alloy films. Any junctions showing spurious gap behavior, whether because of edge effects or alloy inhomogeneities, were discarded. A further criterion was used to determine the quality of the junction. In the Eliashberg gap equations (1) and (2) there are two opposing terms that determine the superconducting properties of a material: the electron-phonon term involving $\alpha^2(w)F(w)$, which is the term enhancing superconductivity, and the electron-electron pseudopotential term U_c, which opposes the pair formation. Over a wide range of materials this Coulomb term does not vary considerably, remaining somewhere about 0.11. If any of the junctions after inversion gave a value of U_c very much different from a previously determined value for lead[5] (0.12 ± 0.02), the junction was discarded. In all the alloys studied, the value of U_c obtained after the inversion of the gap equations was within (0.12 ± 0.01). Using this criterion we believe that reliable values of $\alpha^2(w)F(w)$ have been obtained.

IV. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

Tunneling measurements were carried out on materials Pb, $Pb_{99.5}In_{0.5}$, $Pb_{99}In_1$, $Pb_{98}In_2$, $Pb_{96}In_4$, $Pb_{94}In_6$, and $Pb_{92}In_8$. In addition, resistance-ratio measurements and T_c measurements were done on the alloy films. Some of the results of these measurements are given in Table I. The value of $2\Delta_0/kT_c$, which is a measure of the strength of the electron-phonon interaction, increases slightly with the addition of In. The addition of intermediate-coupling In to strong-coupling Pb does not decrease the strength of the electron-

TABLE I. Resistance ratio, energy gap, T_c, $2\Delta_0/kT_c$, U_c, and the percentage modes in the impurity band for different alloys.

Material	$\frac{R_{293} - R_0}{R_0}$	Δ_0 (meV)	T_c(K)	$2\Delta_0/kT_c$	U_c	Percentage modes in impurity band
Pb	58.5	1.38	7.190	4.45	0.129	...
$Pb_{99.5}In_{0.5}$	16.2	1.39	7.181	4.48	0.117	...
$Pb_{99}In_1$	15.3	1.40	7.173	4.54	0.114	0.66
$Pb_{98}In_2$	11.3	1.40	7.150	4.53	0.114	1.16
$Pb_{96}In_4$	5.9	1.39	7.075	4.54	0.121	2.69
$Pb_{94}In_6$	4.9	1.39	7.061	4.56	0.130	3.08
$Pb_{92}In_8$	3.2	1.38	7.025	4.56	0.114	5.53

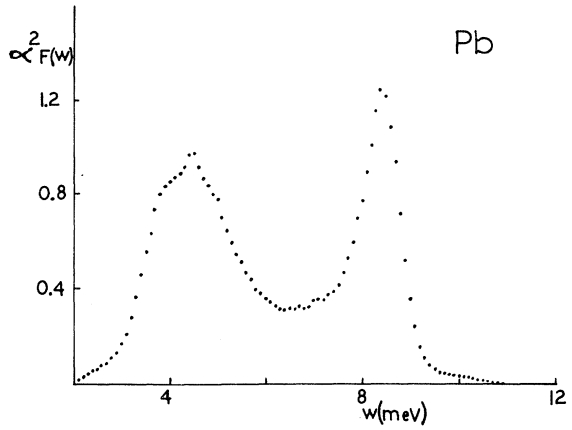

FIG. 1. $\alpha^2(w)F(w)$ for pure Pb.

phonon coupling.

The values of $\alpha^2(w)F(w)$ for pure Pb and different alloys, obtained by inverting Eqs. (1) and (2), are plotted in Figs. 1–4. $\alpha^2(w)F(w)$ for pure Pb agrees very well with the results of Rowell and McMillan.[5] Some of the van Hove critical points are seen clearly for the case of pure Pb. The main features of the phonon spectra of pure Pb are practically unchanged with the addition of small amounts of In except for the appearance of the impurity band beyond the high-frequency cutoff of the pure-Pb phonon spectrum. As the impurity concentration is increased, the impurity band broadens. At the same time, the strengths of the

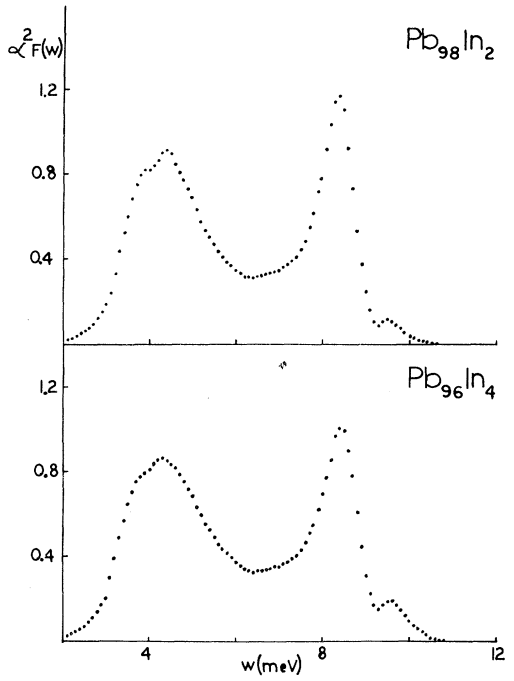

FIG. 3. $\alpha^2(w)F(w)$ for $Pb_{98}In_2$ and $Pb_{96}In_4$.

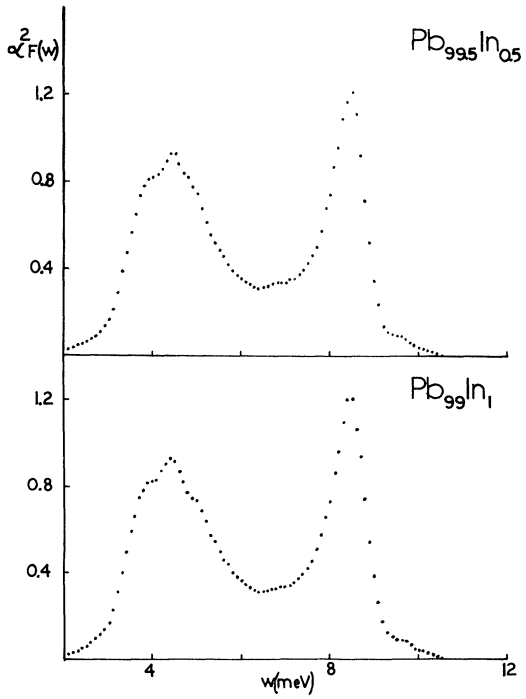

FIG. 2. $\alpha^2(w)F(w)$ for $Pb_{99.5}In_{0.5}$ and $Pb_{99}In_1$.

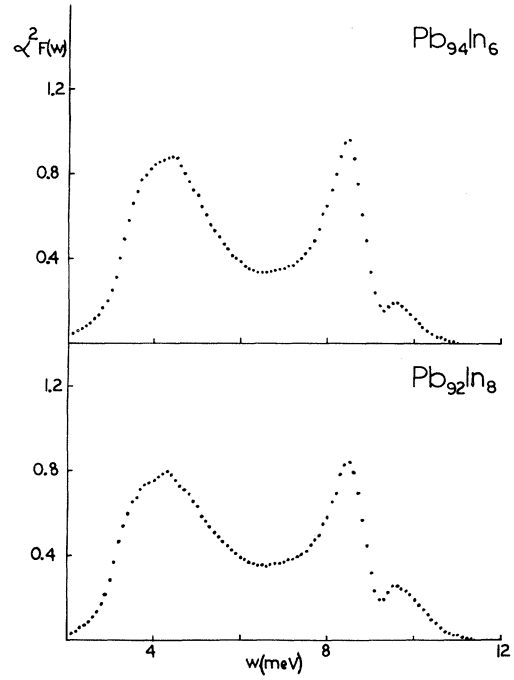

FIG. 4. $\alpha^2(w)F(w)$ for $Pb_{94}In_6$ and $Pb_{92}In_8$.

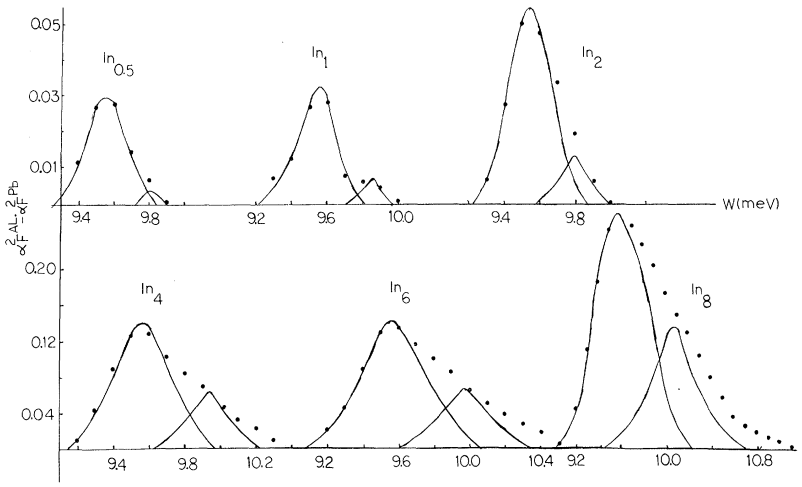

FIG. 5. Difference between the phonon spectra of different alloys and of pure Pb in the impurity-band region only.

longitudinal and the transverse peaks decrease. That is, the impurity band grows at the expense of the longitudinal and the transverse peaks. A close examination of the impurity band reveals that the impurity band has a structure. To see this structure it is essential to take out the background of the pure-Pb phonon spectrum. That is what has been done in Fig. 5. In this figure the difference between the effective phonon spectra of different alloys and of pure Pb is plotted in the impurity band region only (points). It is quite evident that for all the alloys studied the impurity band can be resolved into at least two well-defined peaks. The origin of these two peaks is believed to be the vibrational modes of the single impurity atom surrounded by the heavy host atoms and the vibrational modes of the pairs of the impurity atoms. These two peaks for all the alloys are also shown in Fig. 5 (solid lines). The method used to resolve these two peaks is as follows. The left half of the first peak is drawn first. The right half of the first peak is the reflection of the left half. The left half of the second peak is the difference between the observed points and the right half of the first peak. Finally the right half of the second peak is the reflection of the left half. Note that for higher-concentration alloys the impurity band contains something more than the two peaks described above. This residue could partly be due to the vibrations of clusters of impurities containing more than two impurity atoms. At very low concentrations there are not many of such clusters and thus there is not any residue. We have observed that the position of the first peak is always constant (9.57 ± 0.03 meV). On the other hand, the position of the second peak stays constant at lower concentrations but moves out with increasing amounts of In. This is shown in Fig. 6. We believe that the change in position of the second peak is only because of the errors introduced by the limitations of the method used to resolve the two peaks. At higher concentrations, vibrational modes of higher-order clusters also introduce some errors in resolving the two peaks. No theoretical results are available for the comparison of the position of the second peak.

Let us go back to the results of the calculations made by Elliott and Taylor[3] on the effects of light impurities on the phonon spectrum. They considered only the effects of the mass changes produced by the impurities, and their results are good up to first order in impurity concentration. Thus

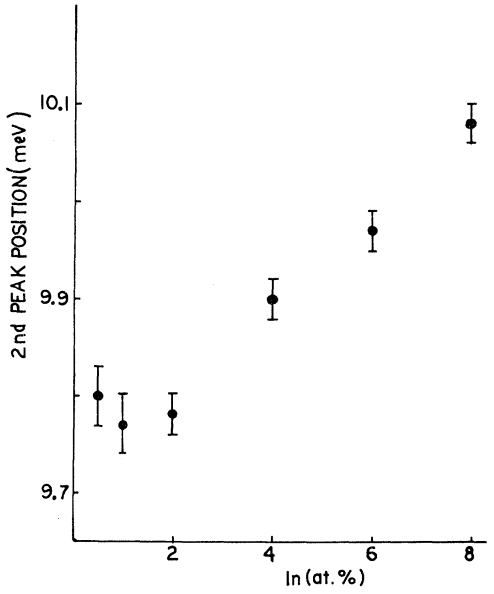

FIG. 6. Position of the second peak vs indium concentration.

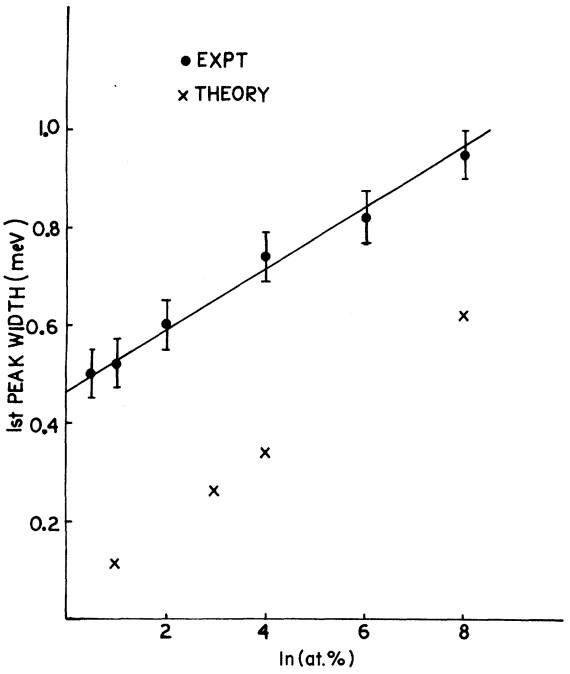

FIG. 7. Width of the first peak vs indium concentration.

their impurity band would correspond to our first peak only. They predict the width of the impurity band to vary linearly with impurity concentration. In our case the width of the first peak has been found to vary linearly with impurity concentration, as shown in Fig. 7. This is in agreement with the theory. The observed linear plot does not pass through the origin, which is in contradiction with the theory. Also shown in Fig. 7 are the theoretical points calculated for the Pb-In system by Taylor.[9] The observed and the calculated curves have the same slope. This indicates that there is always some intrinsic width. Definitely some of the observed width is because of the instrumental smearing. Also, there can be some width because of the finite lifetime of the phonons.

The width of the second peak has also been found to vary linearly with In concentration, as shown in Fig. 8. This plot also has a positive intercept. The ratio of the areas under peak two and peak one is shown as a function of In concentration in Fig. 9. The area under peak one should vary linearly with concentration since the number of Pb-In-type bonds is proportional to concentration. Similarly, the area under peak two should vary as (concentration)^2 since it is proportional to the number of In-In-type of bonds. Thus the ratio of the two areas should vary linearly with concentration. This has been observed as expected. This gives support to the assumed model that peaks one and two are due to the Pb-In and In-In types of

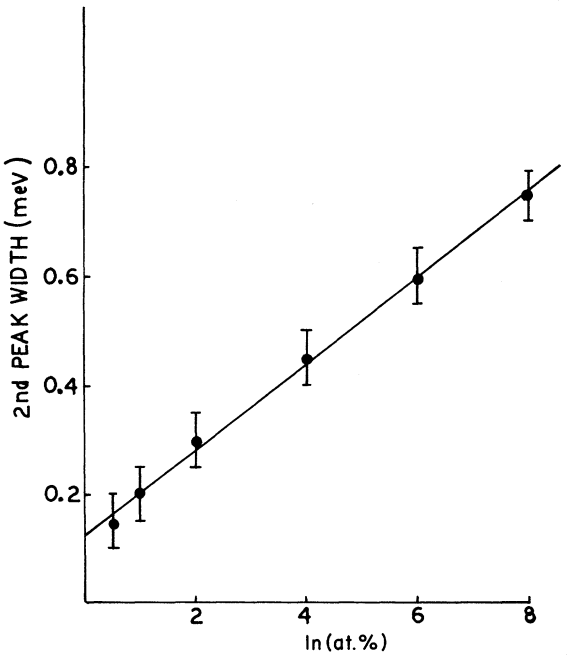

FIG. 8. Width of the second peak vs indium concentration.

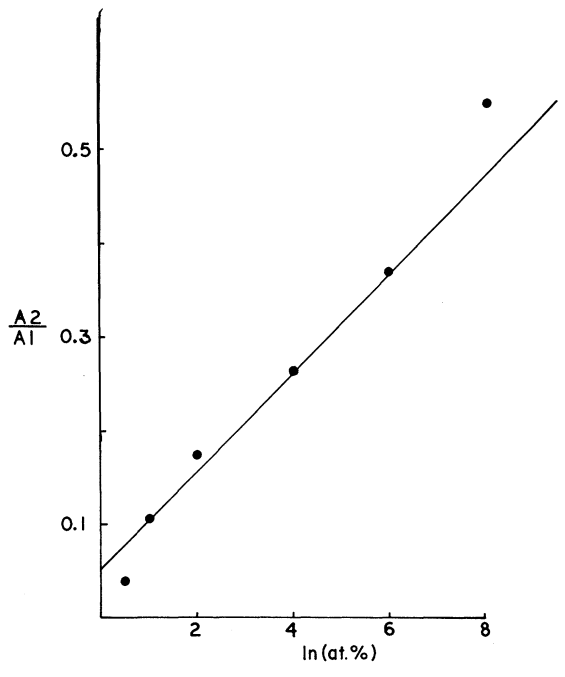

FIG. 9. Ratio of areas under peak two and peak one vs indium concentration.

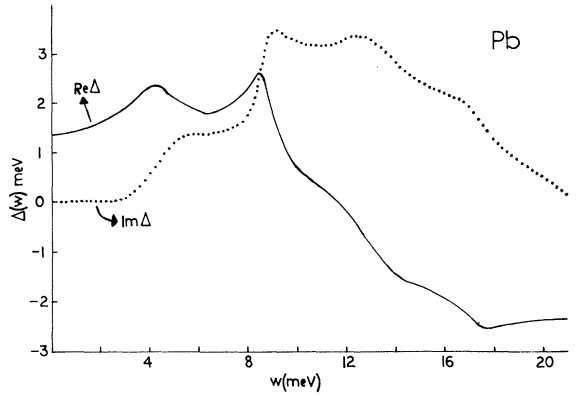

FIG. 10. Real and imaginary parts of the gap parameter for pure Pb vs energy.

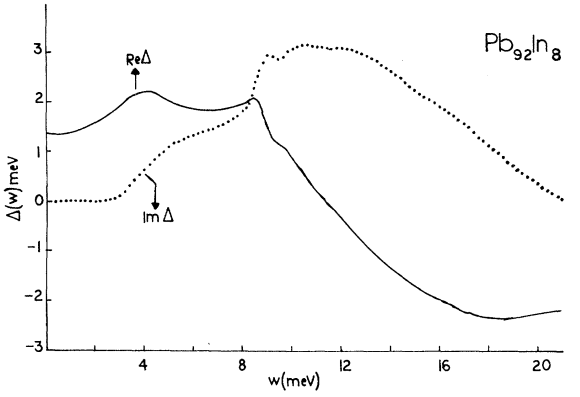

FIG. 11. Real and imaginary parts of the gap parameter for $Pb_{92}In_8$ vs energy.

nearest-neighbor interactions, respectively.

The fraction of the modes in the impurity band should be equal to the impurity concentration.[3] In the present case the impurity band has been found to contain a fraction of modes which is always less than the impurity concentration. Table I lists the fraction of modes in the impurity band for different alloys. The fraction of the modes is always a factor 1.5–2.0 less than the impurity concentration. This can be because of a different value for $\alpha^2(w)$ in the impurity band where In vibrations are dominant. Our results on the fraction of the modes in the impurity band are in contradiction with the results of Rowell *et al.*[10] They find that for a 3-at.%-In alloy in Pb, 5% of the total number of states appear in the impurity band. This could be partly due to the difference in the method used to count the number of states in the impurity band. Our method of mode counting is to subtract the background because of the pure-Pb spectrum from the impurity band and then count the number of modes in the impurity band. Figure 1 for the phonon spectrum of Pb shows a significant background in the impurity-band region.

Figures 10 and 11 show the energy dependence of the real and imaginary parts of the gap parameter for Pb and $Pb_{92}In_8$, respectively. Scalapino, Schrieffer, and Wilkins[11] have shown that for a simple phonon spectrum with a single peak at energy ω_0 a peak at energy $\Delta_0+\omega_0$ is expected in the real and imaginary parts of the gap parameter. Figure 10 for the real and imaginary parts of the gap parameter for Pb shows peaks at the transverse- and longitudinal-phonon energies. In the case of $Pb_{92}In_8$ alloy, in addition to the two peaks at the transverse- and longitudinal-phonon energies, there is an extra peak at the energy where the impurity band occurs. At this energy the impurity-band phonons can be exchanged. Also, there is a weak structure at the energies where multiphonon processes can take place.

V. CONCLUSIONS

The technique of electron tunneling has been very successful in providing the information about the phonon spectra of dilute Pb-based In alloys. The general features of the phonon spectra of these alloys have been found to show a fairly good agreement with the machine calculations on the one-, two-, and three-dimensional disordered systems. The width of the main peak in the impurity band has been found to vary linearly with the impurity concentration, as predicted by Elliott and Taylor. $2\Delta_0/kT_c$ for all the alloys studied does not show any significant change, indicating that the strength of the electron-phonon coupling in the dilute Pb-In alloys does not change very much. The fraction of modes in the impurity band is always a factor 1.5–2.0 smaller than the impurity concentration. This result has been explained as being due to the changes in $\alpha^2(w)$ in the impurity band where In vibrations are dominant.

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Phonon Fluorescence in Superconductors and the Propagation Characteristics of High-Frequency Phonons in Ge:Sb and $Al_2O_3:V^{3+}$

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We present a detailed account of phonon generation by superconducting tunnel junctions (Sn-*I*-Sn) and by Sn and $Pb_{0.5}Tl_{0.5}$ films pumped by a heat pulse. The spectrum and propagation characteristics of the generated phonons is studied through the resonance absorption by Sb-donor levels in uniaxially compressed Ge. At all values of generator power (up to a few watts) the emitted phonons show a large density at a value of energy equal to the superconducting energy gap (2Δ). In contrast, the spectrum emitted by a constantan heater is shown to be in quantitative agreement with the blackbody-radiation model. The experiments show that the mean free path of longitudinal and transverse phonons in the superconductor changes discontinuously when $\hbar\omega = 2\Delta$. Theoretical calculations show that significant reabsorption of phonons of energy $> 2\Delta$ results in a nonlinear buildup of the intensity of the 2Δ phonons. The propagation characteristics of these phonons in Ge:Sb show a frequency and polarization dependence in excellent agreement with the Griffin-Carruthers theory of resonance-fluorescence phonon scattering by donor levels. The magnetic field tunability of the generated 2Δ phonons in Sn is utilized to study the ground state of V^{3+} in Al_2O_3. Transverse phonons of energy 1.02 meV propagating along the *c* axis are resonantly absorbed as expected according to theory.

I. INTRODUCTION

The primary motivation for high-frequency phonon-propagation research is its application to a variety of problems at thermal frequencies. At frequencies above about 5×10^{10} Hz several interesting effects become observable. For example, the energy gap (2Δ) of most superconductors lies in the region of 0.4 to 4 meV ($\sim10^{11}$ to 10^{12} Hz). Thus, high-frequency phonon-propagation studies might reveal important parameters about the electron-phonon interaction in superconductors and their relationship to other measurements. Also, the energy levels of a variety of defects in insulators and semiconductors lie in the range of thermal-phonon frequencies. In many cases the levels of interest are strongly coupled to the strain and hence phonon excitation may be a more fruitful (and possibly simpler) method to study such levels than conventional far-infrared photon absorption. In addition, the problem of phonon-defect interaction is of interest in itself. Finally, lattice anharmonicity and dispersion (in the acoustic modes) become important in many solids (and liquid helium) at frequencies of 10^{11} to 10^{12} Hz. Thus, it is clear that monochromatic-phonon-propagation experiments at high frequencies are potentially of interest to the current theories of heat transport (thermal-phonon lifetimes), electron-phonon interactions, and nonlinear parameters in a variety of different materials.

In this paper we present a detailed account of our work[1,2] on the spectrum of phonons generated by (a) thin-film superconducting tunnel junctions[3] (Sn-*I*-Sn) and (b) by superconducting (Sn and $Pb_{0.5}Tl_{0.5}$) films pumped by a heat pulse. The spectra and propagation characteristics of the generated phonons are studied through the observation of resonance absorption by Sb-donor levels in uniaxially compressed Ge and by time-of-flight techniques. We show that the spectrum consists of a narrow band of phonons centered at the superconducting energy gap 2Δ. Furthermore, the emitted-phonon frequency is tuned by application of a magnetic field parallel to the plane of the film. This tunability is used to study the ground-