EFFECT OF ALLOYING ON THE PHONON SPECTRUM OF LEAD-INDIUM ALLOYS*

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We report the observation of two new and interesting features in the phonon spectrum of lead-indium alloys, inferred from electron tunneling measurements. Even for alloys containing as much as 26% indium, the main features of the pure-lead phonon spectrum are practically unchanged, and the impurity band[1] remains a distinct part of the spectrum.

All the tunnel junctions were prepared at room temperature by evaporating a thin aluminum layer onto a glass slide and allowing its surface to oxidize. The alloy film was subsequently deposited so that an Al/insulator/alloy junction was formed. Since the vapor pressure of lead is much higher than that of indium, the alloy films will in general have lower indium concentrations than the initial alloy prior to evaporation. For this reason the composition of the alloy films was estimated from four-terminal measurements of the residual resistance ratio of the films. The thickness of the alloy films ranged between 1000 and 2500 Å, thus the residual resistance ratio for these alloy concentrations is not dependent on boundary scattering. Resistive measurements showed that the width of the superconducting transition was less than 5×10^{-2} °K for all the alloy films which compares favorably with the widths of 2×10^{-2} °K obtained for pure-lead films. Furthermore several specimens of similar composition (similar residual resistance ratios) exhibited the same energy gap, transition temperature, and tunneling characteristics. We therefore believe our films to be homogeneous.

Measurements of d^2V/dI^2 were carried out using a harmonic-detection system in which the fundamental was suppressed by means of a bridge. This apparatus will be described in detail elsewhere.[2] Measurements of the dynamic resistance dV/dI of the tunnel junctions with both metals normal (n) and again with both metals superconducting (s) enable one to obtain the relative dynamic conductance $\sigma=(dV/dI)_n/(dV/dI)_s$. Thus $d\sigma/dV$, which reflects the phonon structure in the tunneling density of states, was obtained from $(d\sigma/dV)\propto\sigma^3\times(d^2V/dI^2)$. The data presented here were obtained with modulation levels of about 60 μV rms, and experiments using lower modulation levels showed that the essential features of the data remain unchanged.

All the tunneling measurements were carried out in the temperature range from 0.95 to 1.1°K, where both the alloy and the aluminum film were superconducting; the temperature was kept constant within a few millidegrees during each run. At these temperatures the energy gaps of the pure lead and the alloys have reached their limiting value ($T/T_c\sim1/7$). For aluminum films this corresponds to a low enough reduced temperature so that the different tempreatures at which the various runs were made produced only negligibly small changes in the σ and $d\sigma/dV$ curves presented here. The effect of the phonon spectrum of aluminum on the features of σ in the region well removed from the energy gap is 100 times smaller than structure arising in σ due to the phonon spectrum of lead and the alloys.[3]

It has been shown that the broad features of the tunneling curves σ and $d\sigma/dV$ of pure lead may be interpreted in terms of a "simple" phonon spectrum consisting of two Lorentzians, T and L, associated with the density of transverse and longitudinal phonons, respectively.[4] The fine structure in $d\sigma/dV$ is related to the critical points of the phonon spectrum.[5] These results provide a basis for the interpretation of $d\sigma/dV$ in terms of the effect of alloying on the broad features of the phonon spectrum. To unravel the detailed structure of the phonon spectrum of the alloys it would be necessary to carry out the elaborate computational procedure of McMillan and Rowell.[6]

Figure 1 shows σ as a function of energy for pure lead and three alloys containing 3, 12, and 26 at.% indium. These compositions lie well within the range of solid solubility of indium in lead, the alloy lattice remaining face-centered cubic. Figure 2(a) shows details of the high-energy region of the $d\sigma/dV$ curves, while Fig 2(b) covers the range from 2 to 12 meV.

We shall first consider the possible influence of alloying on the electron-phonon coupling. The measured values of $2\Delta_0/kT_c$ range between 4.28 and 4.36 for all the samples, suggesting that the coupling is little affected by alloying.

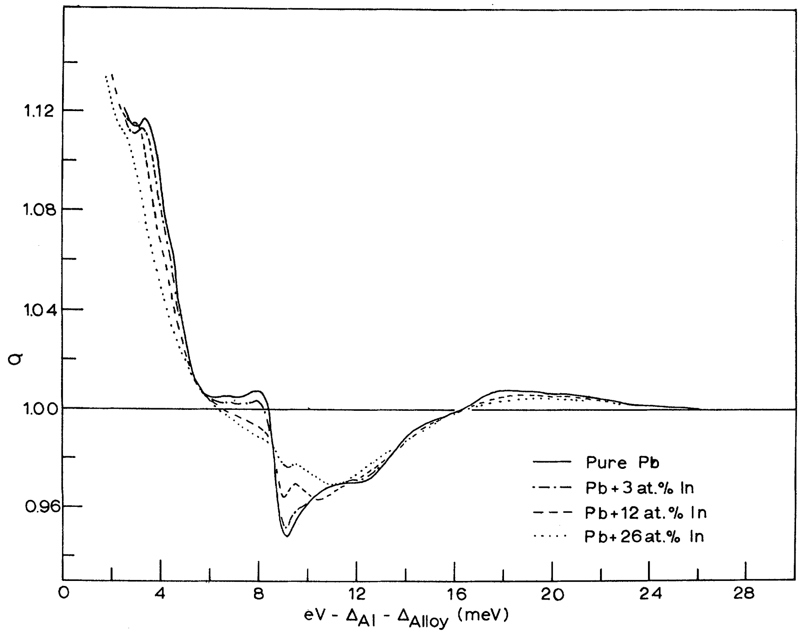
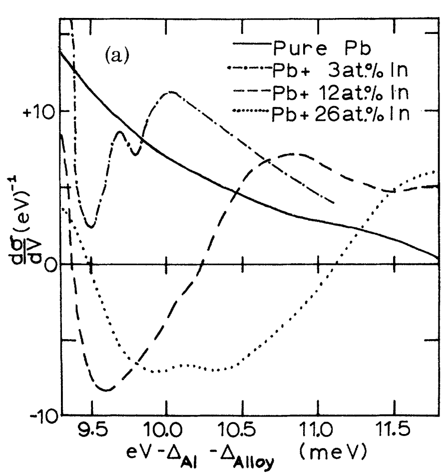


FIG. 1. The relative dynamic conductance σ for an Al/insulator/Pb tunnel junction (solid line), and three Al/insulator/alloy junctions (dashed and dotted lines).

The same conclusion is reached by examining the deviation of σ from its BCS value at the point of maximum slope (near 8.5 mV, Fig. 1). It has been shown[7,8] that this deviation is also an indicator of the strength of the coupling.

We now turn to Fig. 2, and call attention to the two important features of our results: (i) the influence of varying indium concentration on the phonon impurity band (~9.5 to ~11 meV) first observed by Rowell, McMillan, and Anderson[1]; and (ii) the effect of alloying on the features of the phonon spectrum below ~9.5 meV.

The impurity band [Fig. 2(a)].—The Pb + 3 at.% In alloy shows the splitting attributed to



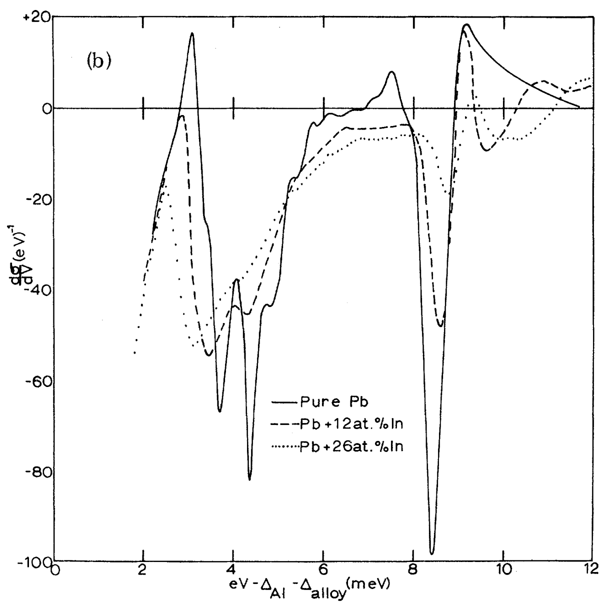


FIG. 2. (a) $d\sigma/dV$ versus energy in the region of the phonon impurity band. (b) $d\sigma/dV$ in the energy range from 2 to 12 meV.

the differing influence of In-In and In-Pb nearest neighbors.[1] As the indium concentration is increased, this splitting becomes washed out as might be expected, but it is remarkable that the structure in the $d\sigma/dV$ curves which is identifiable with the impurity band continues to be clearly distinguishable. One might have expected that in concentrated alloys impurity effects would be absorbed into the main body of the lattice vibrational spectrum; such apparently is not the case.

The phonon spectrum below 9.5 meV [Fig. 2(b)].—It is evident that the amplitudes of the structure associated with the T (3.5 to 5 meV) and L (8 to 9 meV) modes[5,9] are decreased with increasing alloying. This is to be expected since the growth of the impurity band would reduce the phonon density of states in these alloys near the transverse and longitudinal peaks. In spite of the difference in mass and valence between the Pb and In atoms it is surprising that there is no large shift in energy of the L and T peaks. Within the present theoretical knowledge of the phonon spectra of substitutional alloys, a detailed quantitative analysis of these data is difficult. We are extending these measurements to cover the full range of solid solubility of indium in lead; these as well as other tunneling experiments on this alloy system will be presented in detail elsewhere.

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EVIDENCE FOR IMPURITY STATES ASSOCIATED WITH HIGH-ENERGY CONDUCTION-BAND EXTREMA IN n-CdTe

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The resistivities of several samples of n-CdTe have been found to increase by factors greater than 10^4 between 1 and 28 000 kg/cm^2 at room temperature. The interpretation of these results in terms of deionization into an impurity level[1] yields a pressure coefficient for the separation of this level from the lowest (000) conduction band large enough to suggest that the level is associated with the higher lying (100) conduction band, a model originally suggested by Paul[2] to explain Sladek's[3] results on n-GaAs.

12 samples of n-CdTe, similar to those described by Segall, Lorenz, and Halsted,[4] were measured. All of the samples show the same qualitative characteristic of a relative insensitivity to pressure at low pressures, followed by a sharp rise at high pressures. We show in Fig. 1 the ρ vs P relation for five undoped samples in which a decrease in resistivity at zero pressure is believed to be correlated with an increasing excess of Cd. The samples