PHONON "IMPURITY BAND" IN DILUTE SOLUTIONS OF In IN Pb OBSERVED BY SUPERCONDUCTING TUNNELING

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A single, light, substitutional impurity atom in a cubic crystal can cause a triply degenerate, discrete, bound vibrational state to appear above the high-frequency cutoff of the phonon spectrum.[1] A dilute substitutional alloy is expected to exhibit an "impurity band" of such states broadened by their mutual interaction.[2] In this Letter we describe experiments in which the sensitive technique of tunneling between superconductors demonstrates the presence of such an impurity band of phonons in dilute solutions of In in Pb, and we determine its shape using programs which were described previously[3] for the inversion process of calculating the phonon spectrum $\alpha^2(\omega)g(\omega)$ from the tunneling data. $\alpha(\omega)$ is the mean electron-phonon coupling constant for phonons of frequency ω, g the density of phonon states.

In Fig. 1 we show typical data on the second derivative of tunnel current versus voltage for an Al-I-$Pb_{0.97}In_{0.03}$ junction at 0.85°K. For comparison, a curve is shown for pure lead in the same voltage region just above the longitudinal peak. The structure near a phonon energy 9.5 $\times 10^{-3}$ eV is the impurity band. The composition was determined by atomic absorption spectroscopy.[4] Other compositions from ~1 to ~6% gave similar curves with appropriately different intensity in the impurity band, but little apparent change in its over-all width.

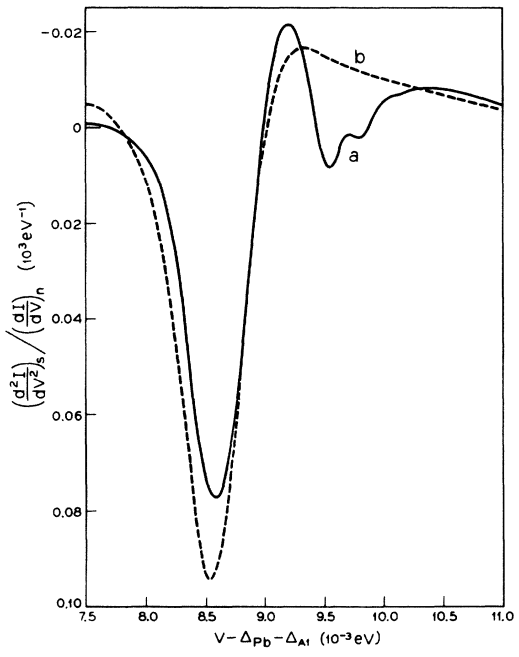

FIG. 1. Curve (a): Second derivative d^2I/dV^2 of tunnel current versus voltage for a Al-I-$Pb_{0.97}In_{0.03}$ tunnel junction at 0.83°K in the region of phonon energies ~9 $\times 10^{-3}$ eV. Curve (b): Similar data for a Al-I-Pb junction.

Figure 2 shows the phonon spectrum [in the sense $\alpha^2(\omega)g(\omega)$] calculated from data like those of Fig. 1. As the aluminum film is superconducting at 0.85°K, the density of states in the $Pb_{0.97}In_{0.03}$ is first unfolded from the tunneling characteristic assuming a BCS density-of-states variation in the Al and allowing for excited quasiparticles. Also shown for comparison is the pure-lead phonon spectrum as calculated in the previous Letter.[3] The total amplitude of the impurity band is ~12% of that in the longitudinal peak, consistent with ~3 × the concentration of 3% In. The electron-local-mode coupling should not be very different from that for the Pb phonons; it appears to be slightly greater. Also note that approximately twice as much intensity is borrowed from the transverse modes as from the longitudinal ones, as expected.

Assuming that the interatomic force constants are unchanged by substitution, the local mode

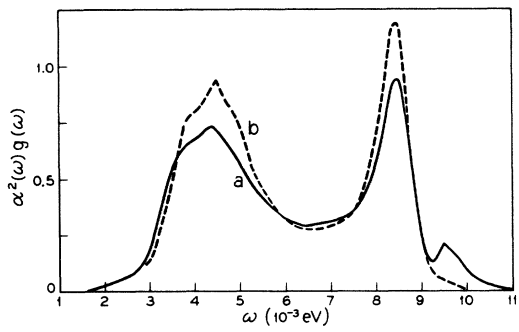

FIG. 2. (a) Calculated phonon spectrum $\alpha^2 g(\omega)$ for $Pb_{0.97}In_{0.03}$ as a function of phonon energy $\hbar\omega$. (b) Calculated $\alpha^2 g(\omega)$ for pure Pb.

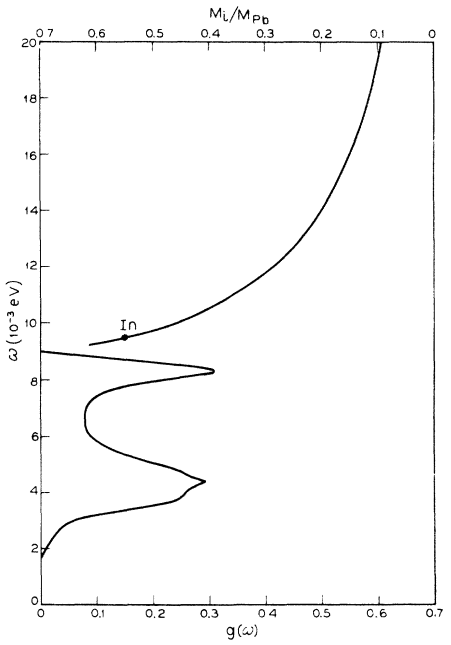

FIG. 3. Calculated position of local mode as a function of impurity mass ratio to Pb, M_I/M_{Pb}. The lower curve is the estimated Pb $g(\omega)$ used in this calculation. Point for In mass is marked.

for a mass ratio M_I/M_{Pb} occurs at a frequency ω_0 given by the equation[1]

$$\int d\omega\, g(\omega) = \frac{M_{Pb}-M_I}{M_{Pb}} \int \frac{\omega^2 d\omega\, g(\omega)}{\omega_0^2-\omega^2}. \qquad (1)$$

$g(\omega)$ was estimated from $\alpha^2 g$ for lead by approximating α^2 by a linear function equal to 1.11 at 4.4 meV, 1.34 at 8.4 meV.[3] The small tail of the spectrum from 9 to 10 meV was assumed spurious because it is not indicated by neutron data, and cut off by extrapolating g linearly to zero at 9.0 meV. With this g the frequency ω_0^2 was calculated as a function of M_I/M_{Pb}, and the result is shown in Fig. 3. The remarkable agreement of the result for M_{In}/M_{Pb} with the peak of the impurity band at 9.5 meV is fortuitous in view of the sensitivity to the force-constant assumption. We do not believe this result to be sensitive to the assumptions about g, although the M_I/M_{Pb} value of 0.74 at which a localized mode first appears is rather sensitive to the form of cutoff at 9 meV.

In addition to a breadth of $\sim\frac{1}{2}$ to 1 meV, the band shows at least one shoulder at ~9.8 meV. Since approximately half the In atoms have an In nearest neighbor, the existence and approximate magnitude of such structure are not unexpected. The fact that the breadth also does not vary with composition may indicate some instrumental smearing or a surface effect.[5]

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[1]I. M. Lifshitz, J. Phys. U.S.S.R. 7, 211, 249 (1943); 8, 89 (1944); Dokl. Akad. Nauk SSSR 48, 83 (1945); M. Lax, Phys. Rev. 94, 1391 (1954).

[2]J. S. Langer, J. Math. Phys. 2, 584 (1961).

[3]W. L. McMillan and J. M. Rowell, Phys. Rev. Letters 14, 108 (1965).

[4]We are indebted to T. Y. Kometani for the analysis of our specimens. Films evaporated simultaneously with the tunnel junctions were dissolved in acid and flamed in a conventional atomic-absorption apparatus.

[5]More recent results for an alloy with <1% In do indicate narrowing of the band and a decrease in relative strength of the shoulder. Some of the observed width is certainly experimental smearing.