Observation of Normal-Metal Phonons with Proximity-Effect Tunneling

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We have measured the tunneling characteristics (dV/dI and d^2V/dI^2 versus V) of thin film junctions of the form Al-I-M-Pb, where M is Ag, Cu, or Al. These tunneling characteristics have *dips* at voltages corresponding to peaks in the phonon density of states of M and *peaks* at voltages corresponding to the peaks in the phonon density of states of Pb. From the amplitude of the dips due to the phonons of M we can crudely estimate the electron-phonon coupling constant λ_M for Al and Cu.

A great deal of theoretical and experimental work has shown that a normal metal may be induced superconducting by the proximity effect.[1] We have studied the tunneling characteristics of sandwiches of Ag, Cu, and Al backed by Pb. Using tunneling into the normal-metal side we have observed structures at voltages associated with the phonons of the normal metal for the first time. We have therefore shown that induced superconductivity involves Cooper pairing between electrons in the normal metal which respond to *phonon interactions* in the normal metal. Our results should allow a more detailed test of proximity-effect theories as well as the use of proximity-effect tunneling to probe the properties of normal metals in a more sensitive way than is afforded by T_c or gap measurements.

Analysis of the conductance of tunnel junctions fabricated of strong-coupling superconductors has produced the most detailed information on the electron-phonon interaction presently available[2] in these materials. For a normal metal the effect of the electron-phonon interaction on the tunneling conductance is much smaller[3-5] (two orders of magnitude smaller for normal Pb versus superconducting Pb). By utilizing the proximity effect we regain the sensitivity of the superconducting state to the electron-phonon interaction so that phonon structure may be observed for normal metals.

Our tunnel junctions were fabricated by (1) evaporating 2000 ± 500 Å of Al, (2) oxidizing it in air at atmospheric pressure from 10 min to 10 h or heating the substrate in air at 100°C for 10 min to 1 h, (3) edge masking with 2000 ± 200 Å of SiO_2, (4) evaporating the normal-metal layer with an electron-beam gun, and finally (5) evaporating 3000 ± 300 Å of Pb from a resistively heated boat. The time delay between evaporations 4 and 5 was approximately 1–3 sec at pressures of order 2×10^{-6} to 2×10^{-5} Torr.

The criteria used for obtaining usable junctions was (1) normal-state resistances of 10–10 000 Ω for a 0.2-mm×1-mm junction at the University of California at Santa Barbara (UCSB) and 50–5000 Ω for a 0.4-mm×2-mm junction at the University of California at Los Angeles (UCLA), and (2) superconducting conductances at zero bias which were less than 0.1% of the normal-state conductance at 1.1 K (where the Al counter electrode is superconducting) at UCSB or approximately equal to the value quoted by Bermon[6] for the measured gap at 1.4 K at UCLA. All data reported in this paper have been reproduced at least twice in each laboratory.

We measured the tunneling characteristics by applying an ac modulation current at ~1000 Hz (1120 Hz at UCSB and 1019 Hz at UCLA) and measuring the voltage at the primary frequency for resistance and twice the primary frequency for d^2V/dI^2, with a lock-in amplifier. The magnitude of the modulation voltage (set at 10 mV bias) was 1 mV rms for the data presented in Figs. 1–3.

Energy gaps at 1.1 and 1.4 K were determined from I versus V curves using the construction of McMillan and Rowell for the sum of the half-gaps[2] and the construction of Douglass and Meservey[7] for the Al gap. Our data analysis was not, however, sensitive to the details of the energy-gap construction used since (1) only ratios of energy gaps for different materials enter in our equation for λ, and (2) the position of the phonon structure is uncertain to ± 0.5 mV.

Tunneling into a strong-coupling superconductor gives *dips* in d^2V/dI^2 near voltages associated

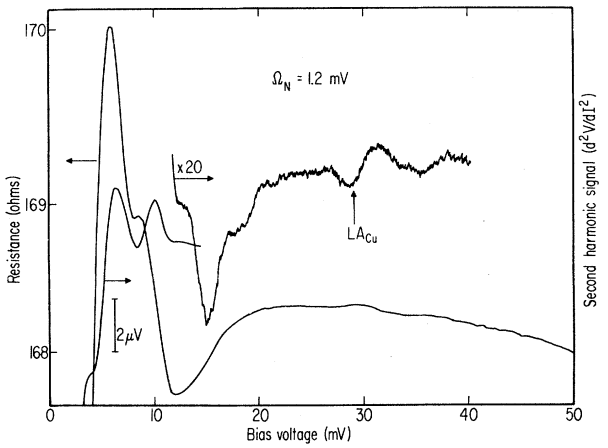

FIG. 1. Resistance and d^2V/dI^2 versus V for an Al-I-Cu-Pb tunnel junction. Above $\approx$ 10 mV the d^2V/dI^2 sensitivity has been increased by a factor of 20. Note the *peaks* near 6 and 10 mV. The arrow at 29 mV is the predicted position (based on neutron scattering) for *dips* due to the LA phonons of Cu.

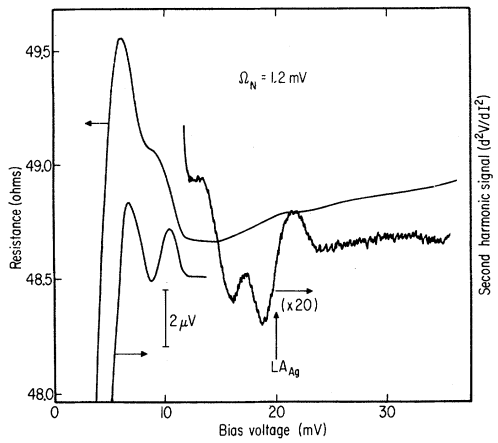

FIG. 2. Resistance and d^2V/dI^2 versus V for an Al-I-Ag-Pb tunnel junction. Note the Pb phonon *peaks* near 6 and 10 mV. The arrow at 20 mV is the predicted position for a ***dip*** due to LA phonons of Ag.

with peaks in the phonon density of states of the superconductor[2]—specifically dips at $V=(E_p+\Omega)/e$, where E_p is the energy of a peak in the phonon density of states, and Ω is the superconducting energy gap. Tunneling into a thin normal metal backed by a superconductor gives *peaks* in d^2V/dI^2 at those same voltages.[8] Thus we would expect *peaks* due to the Pb phonons and hope to see *dips* due to the Ag, Cu, and Al phonons.

Figure 1 shows data taken at 1.40 ± 0.05 K at UCLA for an Al-I-Cu-Pb junction with a (200 ± 20)-Å Cu layer. Note the *peaks* in d^2V/dI^2 near voltages corresponding to the peaks in the Pb phonon density of states (4.5 and 8.5 mV) shifted out by the measured energy gap of the junction (1.2 ± 0.1 mV). Up to 20 mV the structure may be identified as an inverted Pb characteristic.[9] Above 25 mV we would hope to see structure due to the Cu layer. Specifically, a force-constant analysis of neutron scattering data gives a phonon density of states with the largest peak near 28 mV for Cu.[10] Thus, as discussed above, we would expect a *dip* centered near 29 mV; this position is indicated by an arrow on the figure. Inspection of the curve reveals that there is indeed a dip near this position.

Figure 2 shows data taken at 1.40 ± 0.05 K for an Al-I-Ag-Pb junction with a (200 ± 20)-Å Ag layer. A force-constant analysis of neutron scattering data for Ag gives a phonon density of states with its largest peak near 19 mV.[10] Thus we would expect a dip near 20 mV; this position is indicated by an arrow. There is structure close to this bias voltage. Unfortunately, there is a small amount of structure at this voltage due to the Pb electrode. Nevertheless, careful comparison of Fig. 2 with Fig. 1 reveals that the dip followed by an overshoot with a Ag layer is much larger than found experimentally for a Cu layer.

In fact, comparison of the Ag and Cu layer data reveals that the only significant differences are that (1) only the Cu layer data have a dip near 29 mV, and (2) the Ag layer data have a much stronger dip near 20 mV.

Figure 3 shows data taken at (1.4 ± 0.05)°K for

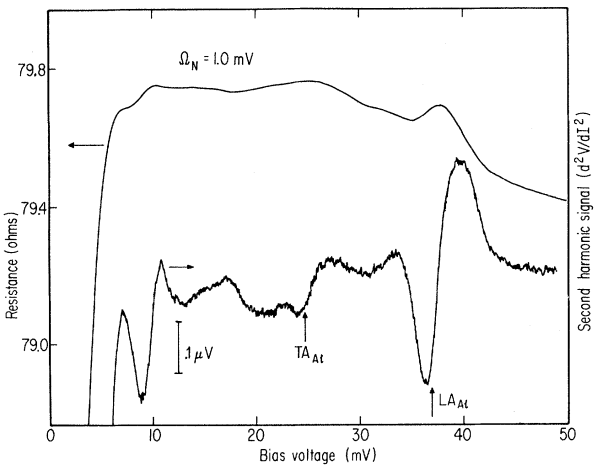

FIG. 3. Resistance and d^2V/dI^2 versus V for an Al-I-Al-Pb tunnel junction. Note again the Pb phonon *peaks* near 6 and 10 mV. The arrows at 25 and 37 mV are the predicted positions for ***dips*** due to transverse and longitudinal Al phonons, respectively.

an Al-*I*-Al-Pb junction with a (200 ± 20)-Å Al layer backed by Pb. At this temperature the Al would not have been superconducting without the Pb backing. Note that the inverted Pb phonon structure is reduced when compared with Ag and Cu data. A partial oxidation of the Al film before the Pb evaporation would produce a reduced Al-Pb coupling and hence a reduced Pb phonon structure. Below 1.1 K, when Al is superconducting, we have observed dips near 37 mV without the proximity effect; however, the structure is much weaker (smaller than the Cu phonon dips in Fig. 2). Furthermore we observe very similar structure in this junction at 4.2 K. Thus, we believe that the enhancement of the amplitude is due to the enhancement of the Al energy gap by the Pb backing. The enhancement is so large that we see, for the first time to our knowledge, dips at energies associated with the transverse phonons in Al. Specifically, the force-constant analysis of neutron scattering data gives a largest peak in the phonon density of states near 36 mV.[11] This peak is due to longitudinal phonons. There is a smaller peak near 24 mV due to transverse phonons. Since the measured energy gap for this junction was 1.0 mV, we have drawn arrows at 25 and 37 mV for comparison with our data. Note that there are indeed dips near these voltages.

Finally, we have used the amplitudes of the Cu and Al phonon structure to obtain crude estimates of λ_{Cu} and λ_{Al}, the electron-phonon coupling constants for these metals. The Eliashberg gap equations may be used to calculate the tunneling density of states. For strong coupling superconductors these equations predict deviations from simple BCS behavior with structure corresponding to the phonon density of states.[2] In order to compare the amplitude of these deviations for different materials we note that if the phonon density of states has the same shape[12] for these materials the amplitudes will scale approximately as $A_1/A_2 = (\lambda_1\Omega_1/\lambda_2\Omega_2)^2$ where A refers to the deviation from BCS, λ is the electron-phonon coupling constant, and Ω is the energy gap. Note that this analysis depends on the induced gap and not the M-Pb coupling, as distinct from other proximity-effect determinations of λ.[1]

For the case of proximity-effect tunneling the structure observed at the normal-metal phonon frequencies may be obtained by using the self-consistency equation for the BCS potential [Eq. (11) of McMillan[13]] in its strong-coupling form. This equation is completely analogous to the Eliashberg gap equations with the replacement of the self-consistent BCS potential by the induced self-energy. The net effect is that the amplitude scaling now depends on the induced gap. We therefore take for the Ω's the measured energy gaps on the normal-metal side of the proximity-effect sandwiches.

Fortunately Al, Cu, and Pb all have the same crystal structure (fcc) and thus similarly shaped phonon densities of states. (Compare, for example, Fig. 3 of Ref. 10 with Figs. 1 and 2 of Ref. 11.) Thus we can obtain estimates for the ratio of Ω's for Al, and Cu relative to those for Pb by using our tunneling data for A and Ω: Our values for Ω are 1.2 ± 0.1 and 1.0 ± 0.15 for Cu and Al backed by Pb and 1.36 ± 0.05 mV for pure Pb. Relative values for A as obtained by numerically integrating the second-derivative structures at the LA phonon frequencies are[14] 0.005 ± 0.001, 0.064 ± 0.013, and 1, respectively.

Thus, using ratios relative to $\lambda_{Pb} = 1.55$,[2,15] we obtain $\lambda_{Cu} = 0.13 \pm 0.03$ and $\lambda_{Al} = 0.5 \pm 0.1$. We are given some confidence in our scaling approximation by comparison of our value $\lambda_{Al} = 0.5 \pm 0.1$ with two estimates based on the McMillan equation for T_c[16]: McMillan's original estimate[16] of 0.38 and a more recent estimate[17] of 0.46.

For comparison with our normal-metal results, theoretical calculations by Das[18] and by Nowak[19] give $\lambda_{Cu} = 0.15 \pm 0.02$, and 0.12 ± 0.02 respectively. Using two different results for form factors, Allen[20] calculates $\lambda_{Cu} = 0.083$ and 0.45. Recent experimental measurements by Hoyt and Mota[21] based on extrapolations from fcc superconducting alloys of Cu with Ga give $\lambda_{Cu} = 0.155$.

In summary, we have observed the phonons of Cu and Ag with electron tunneling by inducing superconductivity in these normal metals with the proximity effect. Further, we have shown that the phonon structure of Al can be greatly enhanced in the same way. In fact, it can be enhanced enough that the transverse phonons can be observed in addition to the previously observed longitudinal phonons. Finally, we have made crude estimates of the size of the electron-phonon coupling constants, λ, for Cu and Al based on the ratio of the amplitudes of their phonon structure to that of Pb.

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Oscillatory Magnetoresistance in 4*Hb*-TaS_2 *†

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Oscillations with frequencies in the range 0.04 to 3 MG have been observed in the magnetoresistance of 4*Hb*-TaS_2. The low frequencies observed are consistent with sections of Fermi surface resulting from band folding due to the charge-density-wave superlattice, and the angular dependence of the frequencies suggests cylindrically shaped pieces of the Fermi surface. For particular lead geometries the oscillatory portion of the experimental voltage displays an unusual current dependence in that it scales linearly with $|\bar{I}|$ but does not depend on the direction of $\bar{I}$.

Shubnikov–de Haas oscillations have been observed in the magnetoresistance and Hall resistance of the layer compound 4*Hb*-TaS_2 in the temperature range 0.62–4.2 K and in magnetic fields up to 220 kG. Five frequencies in the range 0.04 to 3 MG have been identified and correspond to sections of Fermi surface containing from 10^{-4} to 10^{-2} electrons per atom. These correspond to very small cross sections and are consistent with a Fermi surface broken up by the charge-density-wave (CDW) superlattice. The angular dependence for field orientations between parallel and perpendicular is consistent with that expected for cylindrically shaped sections of Fermi surface. Measurements for particular lead and crystal geometries show an unusual and novel behavior of the oscillations in that they appear as terms which depend only on the magnitude of the current $|\bar{I}|$ and not on the direction of $\bar{I}$.

The crystals were grown by iodine vapor transport in a gradient of 720–700°C for four weeks following the method of Wattamaniuk, Tidman, and Frindt.[1] Polyhedral crystals several millimeters in diameter have been mounted with the current flow parallel to the layers in the $[10\bar{1}0]$ direction. Because of the geometrical variation of the crystal cross section and the large anisotropy of conductivity the current may be quite non-