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Tunneling studies of a metallic superlattice

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Tunneling studies of artificially prepared Nb-Cu superlattices have been performed. Individual layers were in the range 8–5000 Å with total sample thicknesses ~1 μm. The coupling strength $2\Delta/kT_c$ shows a continuous decrease from values of ~3.8 (thick layers) appropriate for a strong-coupled superconductor toward the weak-coupled value of ~3.5 (thin layers). Contrary to earlier theoretical predictions and reports of indirect experimental evidence, we find no evidence for electronic states within the superconducting energy gap. The energies of the Nb longitudinal and transverse acoustic (LA and TA) phonons are found to be unaltered from their bulk values down to layer thicknesses of 32 Å. At ~10 Å the TA peak remains unaltered in energy but significantly broadens while the LA peak becomes too broad to measure. These results, in conjunction with earlier Brillouin scattering measurements, imply large changes in the phonon dispersion as a function of layer thickness.

Thin-film deposition techniques are now at a stage where metallic superlattices of good structural quality can be reliably prepared.[1] Possible manifestations of superlattice effects on physical properties of metals can be expected to be largest in systems where one or both of the constituents are superconducting since the relevant length scale for superconductivity is the coherence length ξ ($\xi = 16\,000$ Å for Al, 380 Å for Nb, etc.). This is much longer than layer thickness which can now be reliably prepared.[2] Interesting effects are expected due to the modulation of the properties imposed by the layering process. These include the possible development of new interfacial phonons, the existence of quasiparticle states inside the forbidden superconducting gap, etc. In order to investigate some of these problems we have performed an extensive tunneling study using Nb-Cu superlattices.

We find even for the thinnest layers studied (~10-Å layer thickness) no evidence that the energies of the zone-boundary phonons are substantially modified from those in the bulk. In the energy range accessible to our tunneling states (10–50 meV) we find no evidence for the existence of extra phonon peaks which are not present in the bulk. Contrary to earlier theoretical predictions[3] no additional electronic states are observed in the forbidden superconducting gap. However, a decrease in the coupling strength (i.e., ratio of superconducting gap energy Δ to transition temperature T_c) is found for the thinnest layers.

The Nb-Cu superlattices were prepared by a sputtering technique described earlier.[1,2] The layer thicknesses are extracted from standard θ-2θ x-ray diffraction using a simple theory which relates the position of superlattice reflections directly to layer thickness.[4] The tunneling measurements were performed with the use of a technique pioneered by Wolf, Zasadzinski, Osmun, and Arnold.[5] Immediately after the final Nb layer of each multilayer was prepared, the Ar sputtering gas was pumped from the chamber and a thin (20–80-Å) aluminum overlayer was evaporated or sputtered. The total time for this was less than 2 min. The tunneling barrier was then formed by long (24–48-h) oxidation in the laboratory environment. After oxidation the sample was placed in another deposition system and a counter electrode (In, Pb, or Ag) was evaporated. In this fashion, junctions could be reproducibly formed with almost ideal resistances for tunneling measurements (~50 Ω). The measurements were performed in a ^{4}He cryostat using standard tunneling techniques.[6] All the data presented here are from junctions which satisfy the generally accepted "Rowell" reliability criteria.[6] We have also performed some experiments with thick ($\gtrsim$200-Å) Cu as the top layer backed by Nb (~3000 Å) and find similar phonon structure results to the ones reported in Pb/Cu proximity sandwiches.

The energy gaps were determined from the position of the sharp rise in the I-V characteristics at 1.5 K (or from the peak in the dV/dI characteristic as shown in Fig. 1).[6] Independent determinations of the gaps were also obtained by fitting a thermally smeared density of states to the experimentally measured dV/dI characteristics. Both determinations give superconducting gaps which agree to within ~5%. Figure 2 shows the energy gap versus the layer thickness for a series of samples prepared and measured in random order over a period of two months. It is important to note that gaps measured from junctions with different Al barrier thicknesses closely agree and fall on a universal curve without any corrections.

To determine the coupling strength ($2\Delta/kT_c$) we have made use of inductive and resistive T_c measurements performed earlier on these same samples.[7]

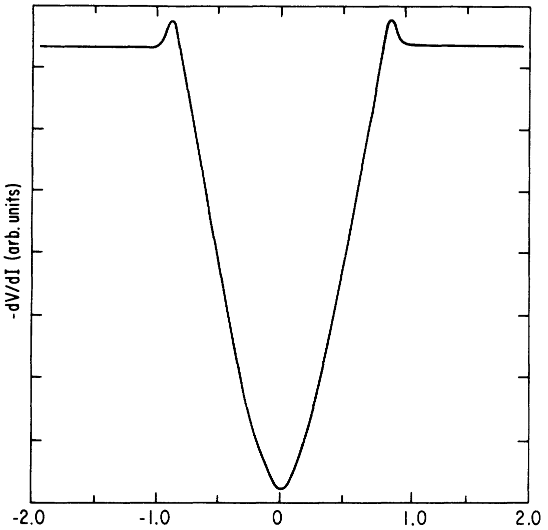

FIG. 1. First harmonic (dV/dI) vs bias voltage characteristic for a superconducting Nb-Cu superlattice.

Figure 3 shows that within the scatter in the data $2\Delta/kT_c \simeq 3.8$ for thick films, as is found for bulk niobium. As the layer thickness is decreased a trend toward the weak-coupling value of 3.54 is observed. Unfortunately the scatter is too large to make more quantitative statements about this behavior.

van Gelder[3] has calculated the superconducting density of states for a system that has a square modulation of the superconducting pair potential. He finds new states inside the forbidden energy gap due to this periodic modulation. Using the Mattis-Bardeen formulation,[8] Liu and Leibowitz related these changes in the density of states to the temperature dependence of the ultrasonic attenuation within a few mk of T_c.[9] They have performed an experiment in which the periodic gap modulation was imposed by subjecting a single crystal of In to a tilted magnetic field at ~ 3.4 K. A comparison of the temperature dependence of the ultrasonic attenuation with the theoretical calculation has been used as evidence for the existence of such states in the gap.[9]

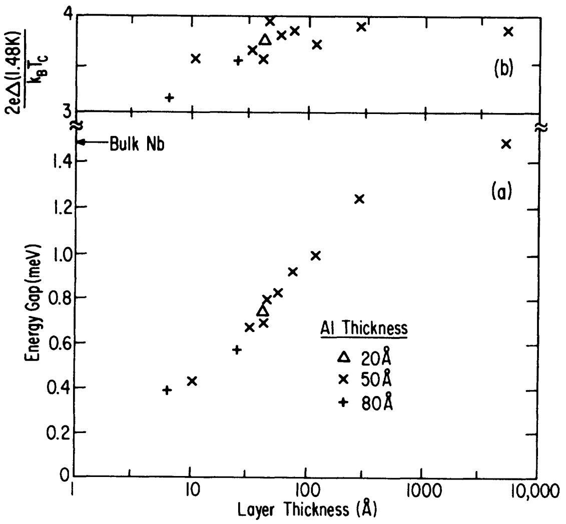

FIG. 2. Energy gap vs layer thickness for various thicknesses of Al tunneling overlayer.

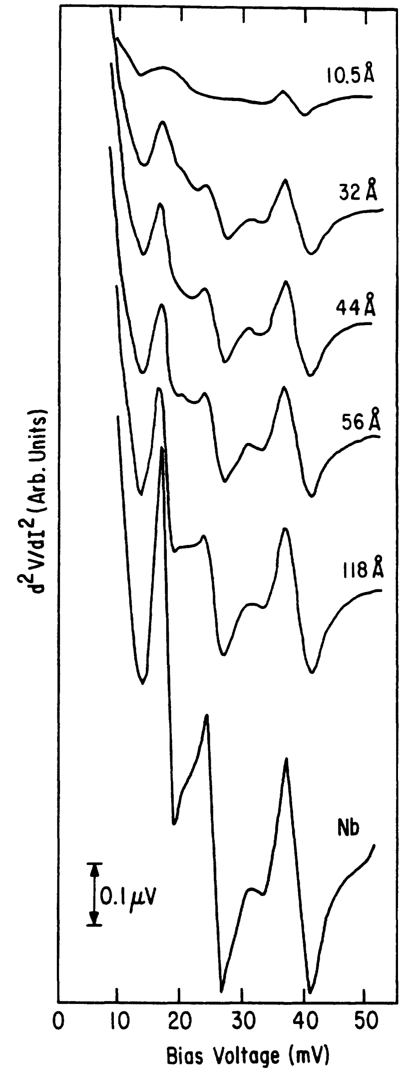

FIG. 3. Second harmonic signal (d^2V/dI^2) vs bias voltage for a series of Nb-Cu superlattices of various layer thicknesses. Peaks occur at the Nb TA phonon (~ 17 mV), Nb LA phonon (~ 25 mV), and Al LA phonon (~ 37 mV).

Of course, tunneling is a very direct measure of the superconducting density of states. Our tunneling data on Nb-Cu superlattices down to 1.48 K shows no evidence for the existence of such superlattice induced states. Since our experiments have been performed for layer thicknesses d ranging from 8–5000 Å the whole range $d \ll \xi_{Nb}$ to $d \gg \xi_{Nb}$ has been spanned. On the other hand, tunneling is a measure of the thermally smeared superconducting density of states. It is possible that the thermal smearing is large enough to make the states in the gap unobservable at ^{4}He temperatures. Of course, if correct, the ultrasonic measurements[9] indicate that thermal smearing is not an important factor for the observation of this effect. We are planning low-temperature measurements down to ~ 50 mK to investigate this point further.

The existence of strong electronic scattering at the Nb-Cu interfaces due to contamination could also affect these results. To check this point ion mill Auger measurements were performed on samples which had been left in the laboratory for over six months. These studies show no contamination by oxygen or carbon below the first ~ 60 Å from the surface.[10] Consequently, we conclude that the disagreement with the zero-temperature van Gelder theory might be real and should be the subject for further theoretical and experimental work.

Superconducting tunneling is also an ideal probe for the observation of phonons in metals.[11] If large van Hove singularities are present, these will be directly observable as structure in d^2V/dI^2 vs V characteristics. Figure 3 shows the measured second-harmonic signal d^2V/dI^2 vs V curves for a series of Nb-Cu superlattices in the voltage range outside of the superconducting gap. All measurements were performed at 1.48 K and in a field of 300 G to quench the superconductivity in the In counter electrode. Three large structures are immediately evident in this series of curves, corresponding to the longitudinal acoustic (LA) phonon in aluminum (~ 37 mV), the LA phonon in Nb (~ 25 mV), and the transverse acoustic (TA) phonon in Nb (~ 17 mV). These energies correspond well to phonon energies in pure bulk Al (Ref. 12) and Nb (Ref. 13) shifted out to higher energies by the superconducting gap. In the energy region studied (0–50 mV) no other phonon structure has been observed in our samples. We should note that the copper phonon [at ~ 29 mV (Ref. 14)] is very weak and is masked by the large Al and Nb phonons so that we do not expect it to show up in this kind of a measurement.

The presence of the aluminum peak serves a valuable role as an internal calibration for the positions and amplitudes of the structures due to other phonons. The energy of the aluminum structure should not shift, irrespective of any possible changes in the Nb-Cu phonons as a function of layer thickness. That this is true can be seen from Fig. 3. In addition, the peak-to-peak amplitude of the phonon structure is expected to scale to first order with Δ^2, as has been shown earlier for a variety of systems in a proximity configuration.[14,15] This result is also observed experimentally, with data spanning an order of magnitude in Δ^2 (and amplitude).

It is interesting to note that no evidence is found for additional surface phonons. The Nb phonon peaks do not shift in energy and only slightly broaden down to layer thicknesses of 32 Å with the broadening causing the LA phonon to be lost in the background for the thinnest layer sample of ~ 10 Å. However, even for this sample there is no evidence for a shift in energy. The observation of bulklike phonons down to layer thicknesses of 10 Å might seem somewhat surprising at first sight. On the other hand, since superconductivity mainly samples $2k_F$ phonons one would not expect to see changes until the thicknesses become comparable to $1/k_F$ ($\sim$one lattice spacing). We have shown earlier using a Brillouin light scattering technique, that the zone-center acoustic phonon exhibits a large decrease ($\sim 20\%$) in its velocity for layer thicknesses centered around ~ 10 Å.[16] This, in conjunction with the fact shown above that the zone-boundary phonons are pinned at a fixed energy, implies that the dispersion relation shows an anomalous "kink" at thicknesses of ~ 10 Å.

We should note that an alternative possibility to explain our data is that only the top Nb layer is being probed in each sample and that the phonons in this Nb layer are unaltered down to ~ 10 Å. We do not believe this to be the case for the following reasons: The coherence length and phonon emission length[17] are both longer than the thickest layers studied here. Proximity effect calculations[7] and tunneling into thick ($\gtrsim 200$-Å) Cu backed by Nb (~ 3000 Å) shows clear superconducting structure demonstrating that the layers are coupled.

Phonon anomalies have been found earlier in many high T_c d- and f-band superconductors and it has been shown that the superconducting transition temperature is related to these anomalies.[18] Theoretical work has shown that there is a tendency towards the formation of a charge-density wave in systems where there is a high density of states at the Fermi surface.[19] Owing to the strong electron-phonon coupling these charge fluctuations give rise to anomalous phonon dispersion and sometimes cause structural phase changes. In addition, in the Nb-Cu superlattice system a charge-density wave is artificially imposed on the lattice due to the periodic composition modulation. It is interesting to note that the x-ray linewidths start showing considerable broadening around layer thicknesses of $\simeq 10$ Å, indicating that structural changes are taking place below this thickness. Clearly, further theoretical and experimental work is needed to clarify the relationship between anomalous phonon dispersion, superconductivity, and localization effects in layered metals.

In summary, we have performed tunneling studies on the metallic superlattice Nb-Cu. A systematic variation found for the energy gap in conjunction with earlier T_c measurements shows a continuous change from strong coupling in thick layers to weak coupling for thinner layers. The theoretical prediction of states inside the forbidden superconducting energy gap due to the periodic modulation of the order parameter is not observed. The energies of the Nb phonons are found to be independent of layer thickness. This, in conjunction with earlier Brillouin scattering measurements, implies that the dispersion relation for acoustic phonons is anomalous in the region of 10-Å layer thickness.

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