SUPERCONDUCTIVITY IN ARTIFICIAL Nb/Cu HETEROSTRUCTURES*

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Transport and tunneling measurements of artificially prepared Nb/Cu heterostructures have been made. Samples studied had individual layers in the range 3.6-5000 Å with total thickness ~ 1 μm. The dependence of T_c above 300 Å layer thickness agrees well with proximity effect theory with no adjustable parameters. Below 300 Å the data indicate the T_c of Nb decreases with layer thickness. The coupling strength $2\Delta/kT_c$ exhibits a continuous decrease from the strong coupled value of ~ 3.8 (thick layers) toward the weak coupled value of ~ 3.5 (thin layers). No evidence is found for a change in the Nb LA and TA phonons energies from their bulk values down to ~ 10 Å. These results, in conjunction with Brillouin scattering measurements of zone center phonons, imply large changes in the phonon dispersion relation as a function of layer thickness in these materials.

I. INTRODUCTION

Techniques for reliably preparing artificially layered metals by sputtering have recently been developed (Schuller and Falco 1979). Superconductivity should be a good probe for investigation of possible manifestations of superlattice effects on physical properties of these materials since the relevant length scale, the coherence length, is longer than typical layer thickness. Interesting effects are expected due to the modulation of 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 series of studies using Nb/Cu heterostructures.

II. SAMPLE PREPARATION

The Nb/Cu heterostructures were prepared by a sputtering process described in detail elsewhere (Falco and Schuller 1982). Very briefly summarizing this technique, oriented single crystal sapphire substrates (c axis in the plane of the substrate) were anchored to a variable speed rotating platform which moved them alternately between the two rate controlled beams of sputtered Nb and Cu (rates ~ 40 Å/sec). In this way samples of individual layer thickness in the range 3.6 Å-5000 Å with total film thickness ~ 1 μm were prepared. For all results presented here, the Nb and Cu layer thicknesses were equal. A detailed study of the X-ray structure (Schuller 1980) shows that samples with layer thickness d above 10 Å have long range coherence (~ 10 superlattice layer thicknesses) perpendicular to the layers (z-direction) and ~ 100 Å crystallite size in the x-y plane of the films. The X-ray diffraction lines became progressively broader for samples below this thickness, indicating that the material becomes more and more disordered for the very thin layers. Bragg θ-2θ X-ray diffraction and/or mechanical film thickness measurements (Sloan Dektac) were used to determine individual layer thicknesses in the manner described previously (Schuller 1980).

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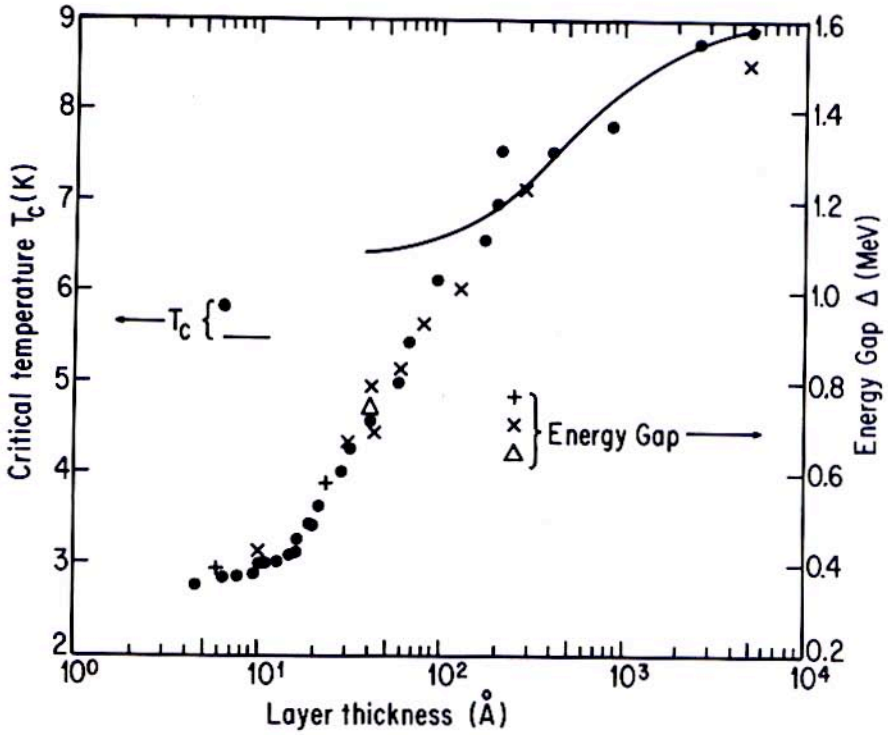

FIGURE 1. Dependence of T_c and energy gap of Nb/Cu heterostructures on layer thickness. Solid line is fit of deGennes-Werthamer theory (described in text).

III. T_c DATA

Both inductive and resistive T_c measurements were made, with agreement of the two techniques to within a few mK (Banerjee, Yang, Falco and Schuller 1982). Each sample showed one sharp transition. Figure 1 shows that for thick layers a T_c of 8.9 K characteristic of pure Nb is found, with T_c decreasing as the layer thickness decreases until it saturates at ~ 2.8 K for thin layers.

The deGennes-Werthamer (dGW) theory of the proximity effect (deGennes and Guyon 1963; Werthamer 1963) has been used to analyze this data. The solid line in Figure 1 is a no adjustable parameter fit of the dGW theory to our data. It can be seen that deviations between theory and experiment begin to occur in the region where d is becoming comparable to the superconducting coherence length ξ_S. However, it has been shown (Hauser, Theuerer and Werthamer 1964) that the dGW model is still applicable for $d < \xi_S$. Thus our results suggest that the properties of very thin layers of Nb are different than the bulk values assumed for calculating the solid curve in Figure 1. Assuming the dGW model to be applicable for $d < \xi_S$, and using the T_c of niobium as an adjustable parameter for d < 300 Å, allows us to extract the T_c of niobium as a function of film thickness. This result, shown in Figure 2, is compared to that of Wolf et al. (Wolf, Kennedy and Nisenoff 1976), where they measured T_c's of single evaporated films of Nb of various thicknesses. The reduced T_c of their thin films, Wolf et al. conjectured, was due to contamination of the surface layer, which becomes

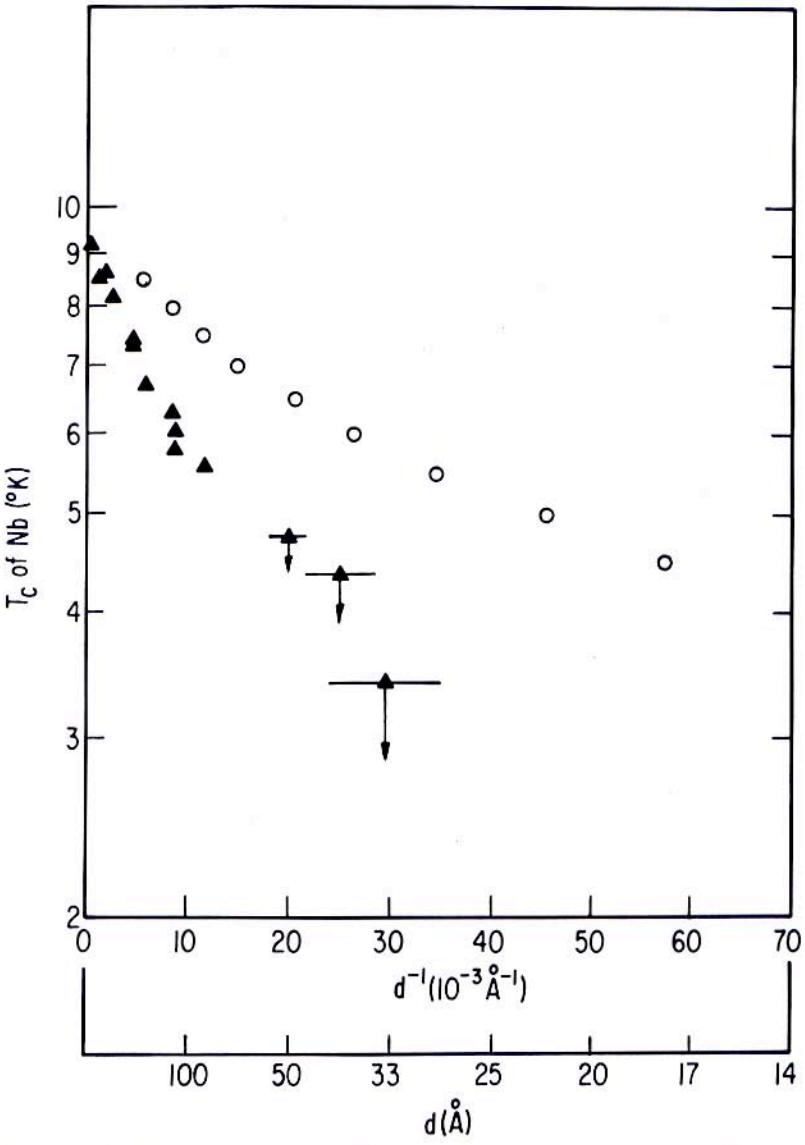

FIGURE 2. ● -- *intrinsic* T_c *of thin Nb layer inferred from fit of deGennes-Werthamer theory to Nb/Cu heterostructure data.* ▲ -- *data of Wolf et al. (Wolf, Kennedy and Nisenoff 1976) on single Nb thin films.*

increasingly important as the films are made thinner. Results of Auger spectroscopy on our layered samples (total thickness always ~ 1 μm) show we have reasonably well-separated layers with clean interfaces and negligible contamination. Our data then imply that the Nb T_c decreases with decreasing layer thickness down to at least 17 Å. Since, as shown by Figure 3, the resistivity

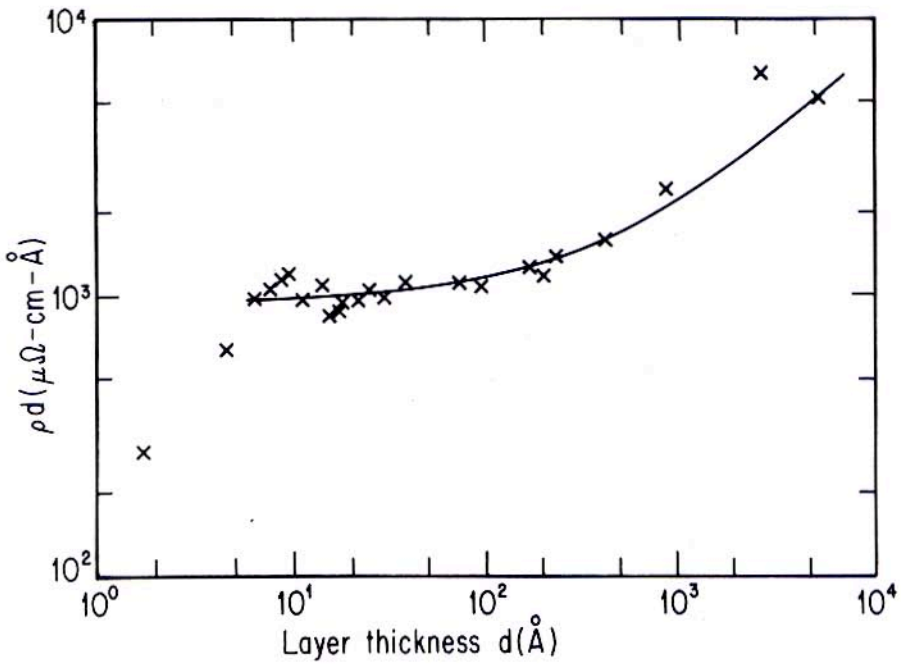

FIGURE 3. Electrical resistivity (measured at 50 K) times layer thickness vs. layer thickness for a series of Nb/Cu heterostructures. Data plotted on log-log scales for convenience in presentation. Solid line provided as a guide to the eye is a straight line on a linear plot.

increases linearly with decreasing layer thickness down to at least 10 Å, this is in qualitative agreement with the idea that the density of states is affected by the decrease in the mean free path.

IV. TUNNELING RESULTS

Tunneling studies were performed on samples prepared in an identical manner to that described above (Yang, Falco and Schuller 1982). The only exception is that in order to use a technique pioneered by Wolf et al. (Wolf, Zasadzinski, Osmun and Arnold 1980), 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 evaporated. The total time elapsed between deposition of the final Nb layer and overcoating with the Al overlayer was less than 2 minutes. The tunneling barrier was then formed by 1-2 days oxidation in the laboratory environment. After this, the sample was placed in another deposition system and a counter electrode (In, Pb or Ag) evaporated. All the data presented here are from junctions which satisfy the generally accepted "Rowell" reliability criteria (Rowell 1969).

The energy gap dependence on layer thickness is essentially the same as the T_c dependence, as can be seen from Figure 1. Also note that these results (as are all our measurements) are independent of the thickness of the Al overlayer. Consequently the coupling strength ($2\Delta/kT_c$) exhibits little variation; decreasing from ~3.8 for thick layers to slightly less than ~3.5 for thin layers. However, the scatter in the data is too large to derive more quantitative conclusions from these data.

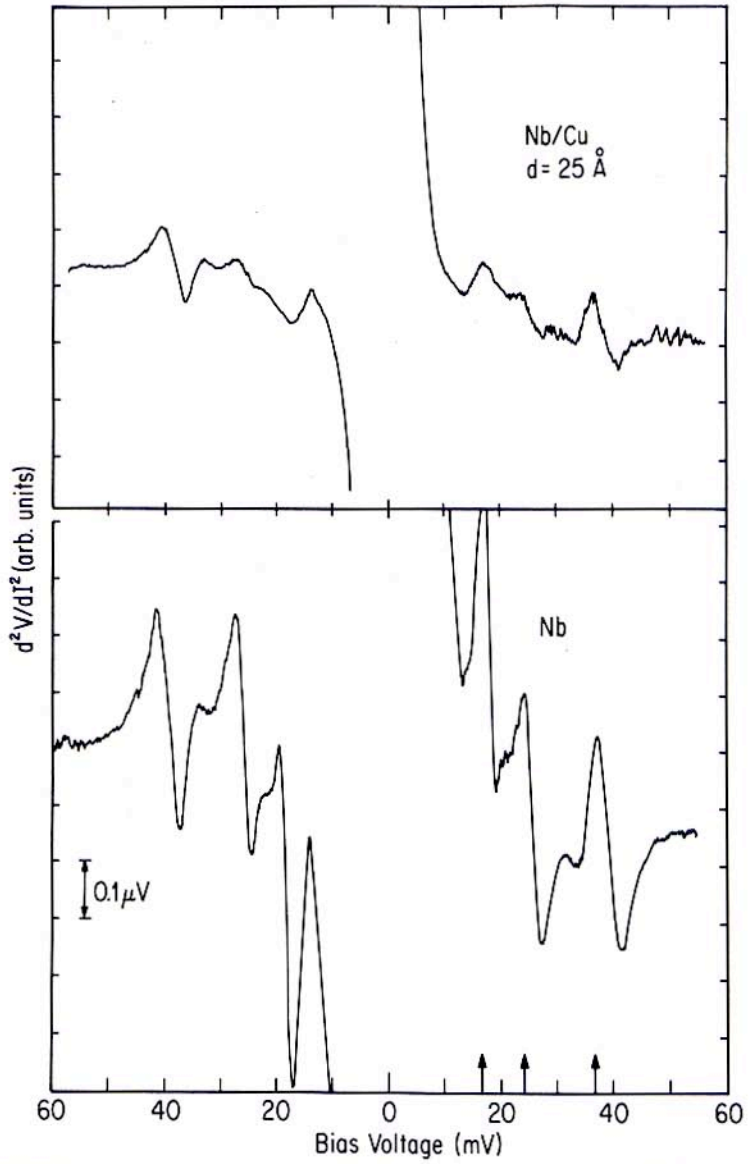

FIGURE 4. Second harmonic "d^2V/dI^2" vs. V curves for a 25 Å layer thickness Nb/Cu heterostructure and a pure Nb film.

Tunneling is also an ideal probe for the observation of phonons in metals (McMillan and Rowell 1969), which will be observable as structure in d^2V/dI^2 vs. V characteristics. Figure 4 shows the measured second harmonic d^2V/dI^2 vs. V curve for a 25 Å Nb/Cu superlattice compared with that of pure Nb. The three prominent features evident in these curves correspond to the longitudinal

acoustic (LA) phonon in aluminum (~ 37 mV), longitudinal acoustic (LA) phonon in Nb (~ 24 mV) and transverse acoustic (TA) phonon in Nb (~ 17 mV). No other phonon structure has been observed in our samples in the 0-50 mV energy region studied. We note that the copper phonon at ~ 29 mV is very weak and is masked by the large Al and Nb phonons.

The aluminum LA peak due to the thin overlayer applied to form the tunnel junction 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 as a function of layer thickness, irrespective of any possible changes in the Nb/Cu phonons. 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 (Chaikin, Arnold and Hansma 1977; Donovan-Vojtovic, Schuller and Chaikin 1979). As shown in Figure 5, this result is also observed experimentally, with data spanning an order of magnitude in Δ^2 (and amplitude).

The Nb phonon peaks only slightly broaden down to layer thicknesses of ~ 32 Å but do not shift in energy (Yang, Falco and Schuller 1982). This broadening causes the LA phonon to be lost in the background for the thinnest layer sample studied of 10.5 Å. However, even for this sample there is no evidence for a shift in energy. The observation of bulk like phonons down to layer thicknesses of 10 Å might seem somewhat surprising at first until it is noted that, since superconductivity mainly samples $2k_F$ phonons, one would not expect to see changes until the thicknesses become comparable to $1/k_F$ (~ one lattice spacing).

We have shown earlier using Brillouin light scattering that the zone center acoustic phonon exhibits a large decrease (20%) in its velocity for layer thicknesses centered around ~ 10 Å (Kueny et al. 1982). This result, in conjunction with the tunneling measurements discussed above showing that the zone boundary phonons are pinned at a fixed energy independent of layer thickness, implies that the dispersion relation shows an anomalous "kink" at thicknesses of ~ 10 Å.

Many high T_c d- and f-band superconductors exhibit phonon anomalies, and it has been shown that T_c is related to these anomalies (see for example various

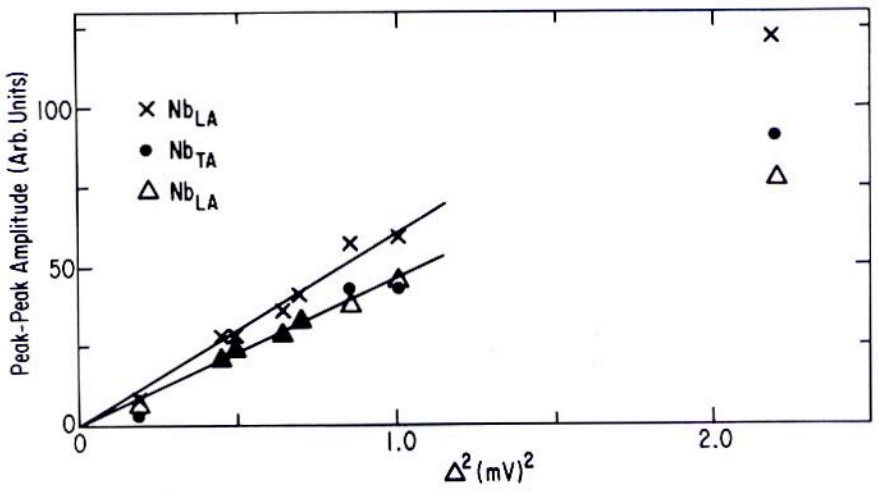

FIGURE 5. Peak-to-peak amplitude of phonon structure in second harmonic curves vs. energy gap for a series of Nb/Cu heterostructures.

articles in Douglass 1976). 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 (for a recent review see Sinha 1980). The strong electron-phonon coupling in these materials can allow these charge fluctuations to give rise to anomalous phonon dispersion and sometimes to cause structural phase changes. In the Nb/Cu heterostructure 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 line widths start showing considerable broadening around layer thicknesses of d ~ 10 Å, indicating that structural changes are taking place below this thickness.

V. SUMMARY

We have investigated the superconducting properties of a new class of artificially prepared layered metals, exhibiting interesting physical properties. The physical properties can be varied by altering the layer thickness, making these materials ideal for a number of studies.

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