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SUPERCONDUCTIVITY OF Nb/Cu SUPERLATTICES[†]

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The superconducting transition temperature T_c of Nb/Cu superlattices has been investigated as a function of layer thickness. 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 in conjunction with current proximity theory shows that T_c of Nb decreases with layer thickness. This is interpreted as changes in the electronic density of states due to a decrease in the mean-free path.

Recently a great deal of interest has been generated in the field of artificially layered metallic films, mainly due to the interesting mechanical[1] and magnetic[2] properties reported. In this paper, we report the results of a study of the superconducting transition temperature T_c as a function of layer thickness of Nb/Cu-Layered Ultrathin Coherent Structures (LUCS).[3] The data has been analyzed using the de Gennes-Werthamer (dGW) theory of the proximity effect,[4-6] and the results suggest that electronic properties of niobium change due to a decrease of the mean-free path induced by the layering process.

Multilayered samples of equal layer thickness d were prepared by sequential deposition of Nb and Cu on 90° oriented single crystal sapphire substrates.[3,7] High sputtering rates (~ 40 Å/sec) were used to minimize contamination of the films. The layer thicknesses were varied by changing the angular speed of a table rotating the substrates above the sputtering guns; the total thickness of each sample being kept constant at ~ 1 μm. For 10 Å < d < 75 Å, Bragg θ-2θ X-ray diffraction was used to determine the layer thicknesses.[7] For all other layer thicknesses, where X-rays could not be used, the layer thicknesses were determined by dividing the total thickness of the film by the total number of revolutions of the table. In the common region where both techniques could be used, this method agrees with the X-ray results to within < 5% as well as with calculations based on the sputtering rates and the geometric sizes of the sputtered beams.

T_c's of the samples were measured inductively using the bridge described in Ref-8. T_c onsets were very sharp and the results agreed within a few mK with the T_c's measured resistively using the standard four probe technique. Each of the samples showed one sharp transition. Figure 1 shows that below a layer thickness of ~ 15 Å, the T_c is approximately constant at 2.8°K. Above this thickness, the T_c increases as the layer thickness is increased until it saturates around 8.9°K; the T_c of pure niobium prepared under identical conditions.

The Werthamer refinement[5,6] of the de Gennes model[4] of proximity effect is used to analyze this data. The equations which relate the sample T_c to physical parameters of the constituents are:

$$\ln\left(\frac{T_{cs}}{T_c}\right) = \chi(\xi_s^2\ k_s^2) \qquad (1)$$

$$\ln\left(\frac{T_c}{T_{cn}}\right) = -\ \chi(-\ \xi_n^2 k_n^2) \qquad (2)$$

$$[N\ \xi^2 k\ \tan kd]_s = [N\ \xi^2 k\ \tanh kd]_n \qquad (3)$$

$$\xi_{s,n}^2 \equiv \frac{\pi \hbar\ k_B}{6\ T_c\ e^2\ (\gamma\ \rho)_{s,n}} \qquad (4)$$

$$\chi(Z) = \psi\left(\frac{1}{2} + \frac{1}{2}\ Z\right) - \psi\left(\frac{1}{2}\right) \qquad (5)$$

ψ is the digamma function, T_{cs}, T_{cn} and T_c are the critical temperatures of the superconductor, normal metal and the sample; d_s and d_n the superconductor and normal metal thicknesses (equal in our case); ρ is the low temperature resistivity; γ is the coefficient of the normal state electronic specific heat and ξ is the effective coherence length defined in Equation (4). N the density of states at the Fermi level is assumed to be directly proportional to γ, thus γ is used in Equation (3). Note that the "normal metal" may either be a superconducting metal (in which case $T_{cn} \neq 0$) or a metal such as Cu with $T_{cn} = 0$. In this limit $\ln(T_c/T_{cn}) \to \infty$ and $k_n \to 1/\xi_n$. A knowledge of γ, ρ, T_{cs} and T_{cn}

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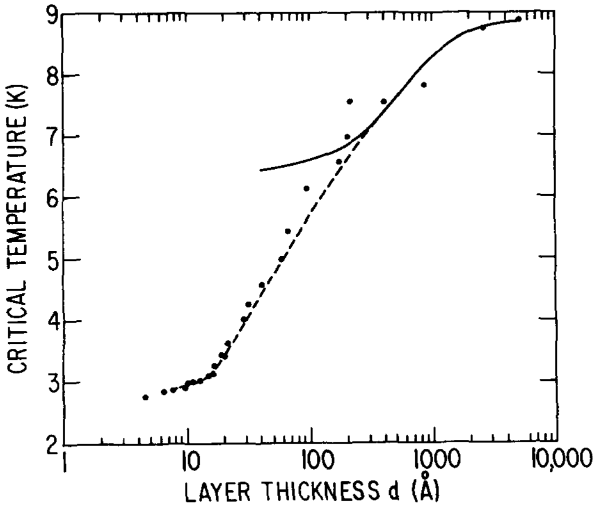

Fig. 1 T_c of Nb/Cu samples versus layer thickness. The solid curve is the dGW fit with no adjustable parameters. The dotted line is the dGW fit using niobium T_c as an adjustable parameter.

in conjunction with Equations (1), (2) and (3) allows T_c to be calculated.

The values of γ used (in J/m^3K^2) are:

$\gamma_{Nb} = 7.5\text{x}10^2$;[9] $\gamma_{Cu} = 1.24\text{x}10^2$.[6]

T_{cn} copper transition temperature is assumed to be 0°K, and T_{cs} is taken to be 8.91°K; the T_c of a niobium film prepared under identical conditions as the samples. Values of $\rho\ell$, where ρ is the resistivity and ℓ the mean free path, varying by a factor of 5 were used to determine the resistivities with only minor effect on the final results. For the data presented here the following experimental parameters for the resistivities of Cu and Nb are used:

$$(\rho\ell)_{Cu} = 6.0 \text{ x } 10^{-15} \text{ ohm-m}^2$$

(determined from the Nb/Cu samples) and

$$(\rho\ell)_{Nb} = 1.5 \text{ x} 10^{-15} \text{ ohm-m}^2 .$$[11]

From the measured resistivities of pure Nb and Cu films we have determined $(\ell_{max})_{Cu} = 2000$ Å and $(\ell_{max})_{Nb} = 160$ Å. For $d < \ell_{max}$ we have used $\ell = d$; in other words the mean-free paths are layer thickness limited up to ℓ_{max}. Beyond this point, they are limited by the above intrinsic mean-free paths.

In the dGW model the following boundary conditions are used:

$$\frac{d\Delta(r)}{dx} = 0 \text{ at metal-insulator or metal-vacuum interface}$$

$$\frac{1}{\Delta}\frac{d\Delta}{dx} = \text{continuous at the normal metal-superconductor interface.}$$[5]

It should be pointed out that the conventional discussion of the dGW model applies to sandwiches made of only two layers. To apply the model in the case of our multilayered samples the first of the boundary conditions has to be applied at the layer midpoints. Thus the 'd' used in Equation (3) to calculate T_c should be put equal to half the layer thickness as opposed to the actual layer thickness.[12]

The solid line in Fig. 1 is a calculation using the dGW theory with no adjustable parameters. It is seen that for d ⩾ 300 Å, the dGW theory is in good agreement with the data. However, below a layer thickness of 300 Å, the measured T_c's of our samples fall off faster and to a much lower value than predicted by the dGW model. Furthermore if one performs the de Gennes version[13] of a Cooper limit[14] calculation using an average (N_oV) of Nb/Cu LUCS system given by

$$(N_oV)_{Nb/Cu} = \frac{N_sV_s(N_sd_s) + N_nV_n(N_nd_n)}{N_sd_s + N_nd_n} \qquad (6)$$

and using

$$T_c = \frac{\theta_D}{1.45}\exp(-1/N_oV) \qquad (7)$$

one obtains $(T_c)_{\text{Cooper limit}} \sim 5.4$°K;[3] whereas we obtain an experimental saturation value of ~2.8°K. This limit simply corresponds to very thin layers where the electrons experience the average pairing interactions of the N and S materials.

Deviations from the dGW model begin to occur in the region where d is becoming comparable to ξ_s. However, Hauser and Theuerer[15] have shown the dGW model to be applicable for $d < \xi_s$. Thus the disagreements mentioned above suggest that properties of niobium begin to change as we deposit it in very thin layers. Assuming the dGW model to be applicable for $d < \xi_s$, and now using the T_c of niobium as an adjustable parameter for d <300 Å, the theory can be fitted to our data. This fitting procedure then allows us to extract the T_c of niobium as a function of film thickness. This result, shown in Fig-2, can be compared to those of Wolf et al.[16] where they measured T_c of single evaporated films of Nb of various thicknesses. The low T_c of the thinner films, Wolf et al. conjectured, was due to contamination of the surface layer, which becomes increasingly important as the films are made thinner. Results of Auger spectroscopy in our layered samples (total thickness always ~1μm), show that oxygen penetration is not more than 60 Å; carbon even less (Fig-3). Since the contaminated volume is <1% of the total volume of the sample, this amount of oxygen penetration should not change the measured properties of Nb significantly. Furthermore, from the phase diagram of Nb-Cu, there is a maximum of about 4% interdiffusion of the two materials. We thus have reasonably well

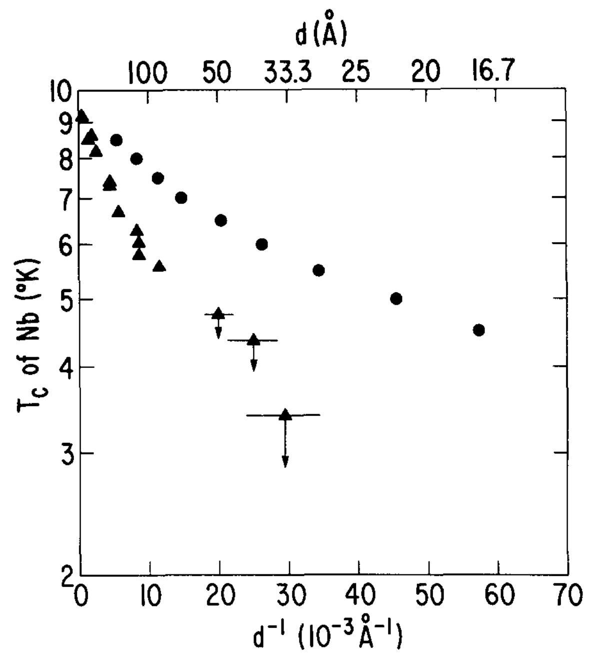

Fig. 2 Plot of T_c of niobium versus inverse layer thickness. ▲- data of Wolf et al. ●- Inferred from our Nb/Cu data using T_c of Nb as an adjustable parameter in dGW theory.

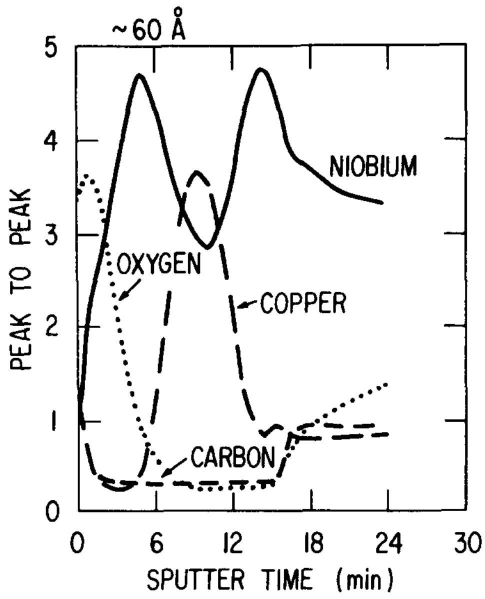

Fig. 3 Auger spectrum of a Nb/Cu sample as a function of ion milling (sputtering) time. Notice the rapid decay of oxygen and carbon. The buildup of oxygen after 16 min. occured after cessation of sputtering with the sample still under vacuum.

phase separated layers with clean interfaces and negligible contamination.

Possibly the T_c of niobium is changing due to the shortened mean-free paths, which are limited by the layer thickness as shown by independent resistivity measurements. Radiation damage studies[17] on niobium have found the T_c to decrease with increasing damage, showing that the niobium T_c is strongly dependent on the mean-free path. Measurements of the T_c of Nb by Crow et al.[18] and Asada and Nose[11] also show dependence on mean-free path. Such changes in T_c have been attributed to changes in the electronic density of states as discussed in Ref. 18-19. We have some evidence to the fact of lowering of the density of states. Magnetic susceptibility measurements on three samples show the susceptibility to decrease with the layer thickness; in all three cases the susceptibility is lower than the average value of pure niobium and copper. We may mention in passing that Testardi et al.[20] have reported no significant change in T_c of niobium by radiation damage. This does not seem to agree with our conclusions nor with the other radiation damage studies cited in the references.

In conclusion, above 300 Å the thickness dependence of T_c of Nb/Cu multilayered films can be explained using standard proximity effect theories with no adjustable parameters. Below this thickness, the data implies that the niobium T_c is decreasing with decreasing thickness. This inference is in qualitative agreement with experiments on single layer thin Nb films and with the idea that the density of states of Nb is affected by the decrease in the mean-free path.

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