Superconducting Transitions in Ultrathin, Amorphous, Metallic Multilayers

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(Received 24 February 1989)

Multilayers of ultrathin superconducting a-$Mo_{79}Ge_{21}$ and normal Ge-rich a-$Mo_{1-x}Ge_x$ were prepared to study the effects of disorder on superconductivity in reduced dimensions. The superconducting transition temperatures of the multilayers as a function of composition and thickness of the normal metal show a systematic enhancement over the single-film T_c. This effect can be attributed to a crossover from 2D single-film behavior to 3D bulk behavior as electron diffusion between individual superconducting layers increases. The nature of this crossover sheds light on the microscopic mechanisms reducing T_c.

PACS numbers: 74.70.Mq

It is well known that disorder has a profound influence on conduction processes in metals.[1] These effects are even more pronounced in reduced dimensions. At the same time, disorder can influence the cooperative behavior of materials. In particular, the superconducting transition temperature of disordered thin films has been observed to decrease systematically from the bulk value as the thin film thickness is reduced.[2] Experiments on homogeneously disordered (i.e., amorphous) films have attributed this reduction in T_c to the enhanced Coulomb interactions expected in disordered systems.[3,4] This conclusion is based on the success of recent theories to account semi-quantitatively for the observed behavior.[5,6] No direct confirmation of the role of Coulomb effects has been demonstrated however. Hence alternative theories[7] or interpretations cannot be definitively ruled out.

In order to test more directly for the effects of enhanced Coulomb interactions on the reduction in T_c in disordered superconducting thin films, we have attempted to alter the screening in these films and to examine the effect on the transition temperature. Related experiments have been reported recently by Bergmann and Wei, which probe the effects of screening on the temperature dependence of the normal-state resistance in sandwiches of thin disordered metal films separated by an insulating layer.[8] The physical idea behind these types of experiments is as follows. In 2D disordered systems, the Coulomb interaction (at finite frequencies) has a $1/q^2$ divergence at long wavelengths; hence long-range contributions are thought to be particularly important. According to current theories enhancements in the Coulomb interaction, and in particular the long-wavelength divergence, inherently compete with superconductivity and cause a significant reduction in the transition temperature as the films are made thinner. Therefore, if one can alter this long-range Coulomb interaction by means of a highly conducting ground plane, a corresponding effect on T_c should be observed.

We have attempted to further clarify the origins of the reduction of T_c in disordered superconductors, and to explore specifically the effect of screening at various length scales, through a study of the superconducting transitions in a series of superconductor/nonsuperconductor (S/N) multilayers. In these multilayers the distance between the superconducting layers (and hence their mutual screening) has been systematically varied from 200 down to 20 Å. At the same time, we have systematically varied the conductivity of the N layers from insulating to metallic. This increased conductivity of the N layers makes the diffusion of the electrons more three dimensional, which further increases the screening. It also introduces a reduction in T_c due to the proximity effect and permits Josephson coupling between the layers. The complication introduced by the proximity effect was compensated by comparing the T_c of a given multilayer with that of a corresponding $N/S/N$ single-layer sandwich structure designed to have an identical proximity-effect reduction of T_c. This strategy also compensates for any effect of the S/N interface itself on T_c. Therefore, an unambiguous comparison can be made between the 2D behavior of a single layer and any 3D effects which may occur in the multilayer.

The multilayers were fabricated by means of sequential cosputtering of alternate layers of superconducting and nonsuperconducting amorphous Mo-Ge alloys using procedures described previously in detail.[9-11] The Mo-Ge system forms a homogeneous, amorphous alloy over the composition range from pure Ge to 80 at.% Mo for thin films grown at room temperature.[12] Therefore, the electronic properties can be varied as a function of Mo concentration from those of an insulator, through the metal-insulator transition, and into the realm where superconductivity exists.[10] The conductivity and critical temperature increase with Mo concentration and T_c ultimately reaches 7.4 K near 80% Mo. The bulk structural normal-state and superconducting properties of these al-

loys have been studied extensively in thick films.[10,12,13] The tunneling properties of pure a-Ge are also known through previous work.[14] Thus considerable information about the amorphous Mo-Ge material system exists that can be used to help in the analysis of our data and to check the quality of the layering.

For the S layers in our samples, we chose 25-Å ultrathin films of $Mo_{79}Ge_{21}$. The T_c of bulk $Mo_{79}Ge_{21}$ is 7.4 K and is reduced to 4.0 K in a 25-Å film. For the N layers the thickness was varied from 20 to 200 Å and the Mo concentration was varied between 0 at.% (pure a-Ge) and 42 at.% Mo. To compensate for the proximity effect, in a given matched set of samples the thickness of the N layers was chosen as shown in Fig. 2(a). In each single-layer sandwich and in the corresponding multilayer, the top and bottom N layers were made half the thickness of the basic N layers of the multilayer. According to the conventional theory of the proximity effect,[15,16] by symmetry this procedure should lead to identical proximity-effect reductions of T_c.[17]

The quality of the layering is extremely important, since we are concerned with relatively small changes in the transition temperatures of very thin films. To determine the quality of the layering, we have studied x-ray diffraction on a multilayer from each set. In each case sharp peaks are observed at low angles, as is expected for well defined layers with different scattering factors. The spacing between peaks corresponds to the periodicity expected from the deposition rates to within a few percent. Further confirmation of the quality of the layering and the presence or absence of an interlayer short is provided by cross-sectional TEM's prepared via ultramichrotomy.[18] Smooth, continuous layers were observed in each case. High-resolution images suggest the interface mixing to be on the order of a few angstroms. Thus, from a structural standpoint, high-quality layering has been achieved.

Additional evidence of the layering quality can be obtained through examination of the transport properties in the normal state. The low-temperature resistivity of each sample was measured using the van der Pauw technique at 15 K where $\rho(T)$ is essentially flat, above the region where fluctuation conductivity effects may contribute significantly. Since Mo-Ge is an amorphous material, the resistivity should not depend on film thickness, and in a multilayer one would expect each layer to behave independently. In this case, each sample may be described by a set of N parallel conductors, where N is the effective number of full N layers (counting two half layers as one) with thickness d_N. In these terms the sheet conductance is given by $G_\square = N_S \sigma_S d_S + N_N \sigma_N d_N$, where $\sigma_{S,N}$ is the conductivity of the individual layer, and $d_{S,N}$ is its thickness. In both the sandwich structures and the multilayers the effective number of S and N layers is the same, so to test the above equation, we plot the sheet conductance per layer as a function of d_N

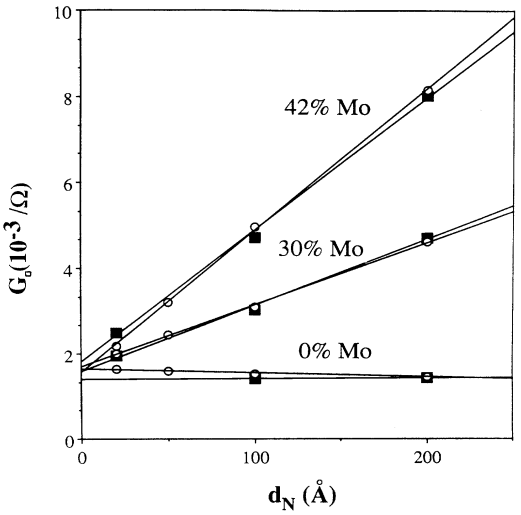
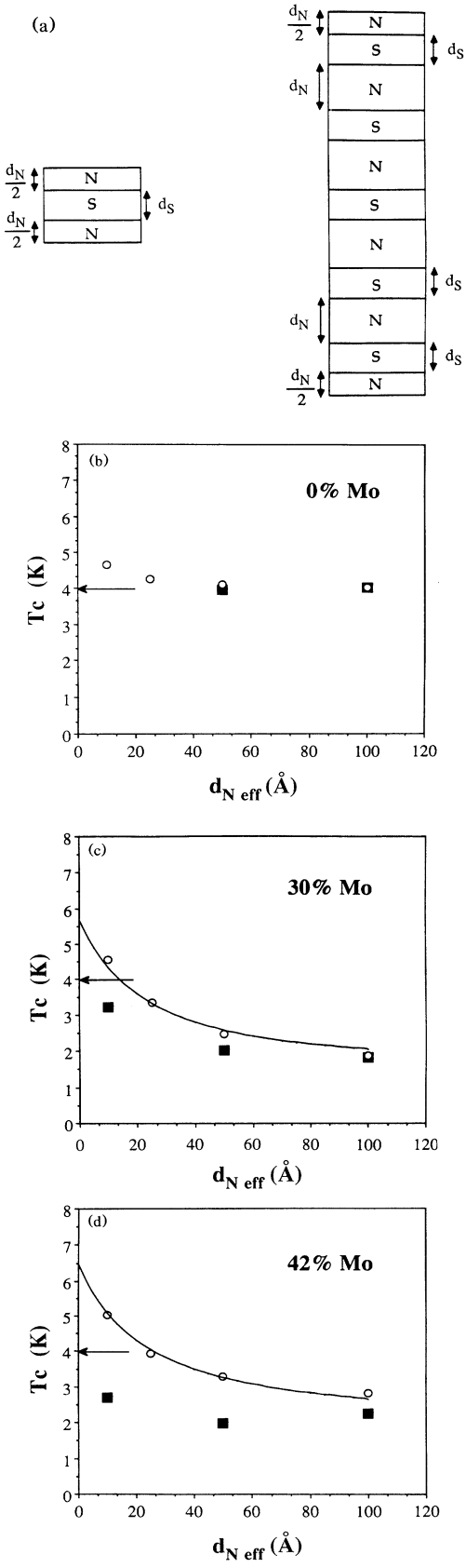


FIG. 1. Sheet conductance per layer as a function of d_N for three matched pairs of samples; ■ denote single-film structures, O denote multilayers.

(since $d_S = 25$ Å is constant in all cases). The conductance per layer is shown in Fig. 1 where it is confirmed that both the multilayers and sandwich structures have the same slope (and hence σ_N) for a given Mo concentration. Also in each case the intercept corresponds to the sheet conductance of an individual S layer. Finally, we note that the material parameters obtained from the slopes and intercepts of these curves correspond to those measured in bulk films of similar composition within 10%.[10] This agreement demonstrates directly that the conductivity remains essentially constant, even in such thin layers, and confirms that no gross changes have occurred in the normal-state properties. Hence, the electrical properties of the sandwich structures and multilayers can be compared on an equal basis with considerable confidence.

Consider now the transition temperatures of these samples. The critical temperature of each sample was measured using the van der Pauw technique, with phase-sensitive detection at a frequency of 27 Hz and current densities of 1–10 A/cm^2. The transition temperatures as a function of d_N^{sandwich} are shown in Fig. 2 for three different sets of matched single-layer sandwich and multilayer structures. Here T_c is defined as the midpoint of the resistive transition for samples with transition widths (10%–90%) less than 0.03 K. For those with broader transitions, T_c is taken as the inflection point. Where direct comparisons have been made, the T_c's defined these ways are very close to the extrapolation of the inverse Aslamasov-Larkin fluctuation conductivity,[19] which approaches T_c linearly in 2D. We note that the transition widths decrease systematically as the N layer becomes more metallic. In addition, for a given Mo concentration in N, the width also decreases with d_N,



presumably reflecting the more three-dimensional behavior.

The samples with 0% Mo illustrate the behavior of insulating N layers, while those with 30% and 42% Mo illustrate that for metallic N layers. Clearly, the behavior is different in the two limits. For the insulating N layer, the transition temperatures of the multilayer and sandwich structures correspond, and are independent of d_N except for the smallest d_N (20 Å), where the T_c of the multilayer is slightly higher. At this d_N the S layers begin to be coupled via tunneling, since the tunneling length in a-Ge is known to be about 10 Å.[14] Since the T_c's in the multilayers with larger d_N are near the single-film T_c value, these samples evidently retain their 2D character. However, as N becomes metallic, the changes in T_c are more dramatic. The critical temperatures of the multilayers lie significantly above the single-layer sandwich structures and are strongly dependent on d_N.

Note that for both of the multilayers with metallic N layers, the T_c of the multilayer rises above that of a single layer with insulating over and underlayers as d_N becomes small. By contrast, for a single-layer sandwich with the metallic over and underlayers the T_c is always lower. Hence, any increased screening due to the metallic nature of the N layers is not enough to raise T_c, at least not above its value in the presence of the proximity effect with the metallic N layer. On the other hand, the increased diffusion between the S layers present in the multilayers is apparently sufficient to raise T_c toward its bulk value despite the proximity effect. In fact, modeling of the observed dependence of T_c with d_N using proximity-effect theory (solid lines in Fig. 2) can only be carried out with reasonable parameters provided that the intrinsic T_c of the S layers is taken to systematically increase toward the bulk value as the N layers become more metallic.[20] The value of this T_c can be inferred from the limiting value of T_c of the multilayer as d_N goes to zero.

These data demonstrate that the original reduction in T_c is an intrinsic 2D effect and not due solely to some type of proximity or interface effect, or due to some other change in the intrinsic properties of the individual S layers as they are made thin. They put stringent constraints on the specifics of any theory proposed to account for the reduction of T_c in disordered superconductors. For example, they appear to put an upper limit of 20 Å on the cutoff of the $1/q^2$ divergence in the long-range Coulomb interactions in the 2D limit.

FIG. 2. (a) Schematic of single-film structure and multilayer; $S=25$ Å $Mo_{79}Ge_{21}$, $N=Mo_{1-x}Ge_x$ where $1-x$ is varied for each matched pair. (b)–(d) Critical temperatures of single-film structures (■) and multilayers (○) as a function of d_N for (b) $N=0\%$ Mo, (c) $N=30\%$ Mo, (d) $N=42\%$ Mo. Arrows indicate T_c for the original $N/S/N$ structure where N is pure a-Ge.

Although increased diffusion between S layers in the multilayer structure appears to raise T_c towards the bulk value, we note that the relevant thermal diffusion length $\xi_T = (\hbar d/2\pi k_B T)^{1/2}$ appears to have changed very little. For example, if we use the values of D obtained from bulk critical-field slopes,[9] we find that ξ_T at 4 K only varies from 30 to 34 Å between the samples with $N = 30\%$ Mo and $N = 42\%$ Mo, yet the $n = 42\%$ Mo multilayers have much higher T_c's. This apparent sensitivity of T_c to small changes in ξ_T suggests that the samples may be in a critical regime where for small ξ_T the electrons are confined to a single layer, but as ξ_T increases slightly, the probability for electron transfer between S layers (or the effect of such transfer) is significantly enhanced. As pointed out by Bergmann and Wei, if electron transfer between layers in a multilayer structure is sufficiently strong, any effects due to Coulomb interactions should be the same as those in a single layer whose total thickness is the sum of each individual layer thickness.[8] This argument relies on a short-ranged Coulomb interaction, which is also supported by our data as discussed in detail above. Any additional effects on T_c in the multilayers due to 3D Josephson coupling or magnetic coupling between S layers are unknown. We have not attempted to quantitatively account for the differences observed between single-film and multilayer structures in the spirit of theories of T_c reduction in ultrathin films. However, we feel that a comprehensive treatment which includes these effects and the proximity effect on an equal footing will be required.

In summary, we have observed a continuous crossover of the transition temperatures in matched sandwich and multilayer structures from 2D single-film behavior to 3D bulk behavior as electron diffusion between thin superconducting layers increases. This study explicitly demonstrates that simple interface effects or interactions with the substrate are not responsible for the reduction of T_c in these ultrathin, disordered Mo-Ge films. The 2D values of the critical temperatures observed in multilayers with insulating N layers suggest that if enhanced Coulomb interactions are responsible for the reduced T_c in single thin films, then the relevant wavelengths are less than 20 Å. Instead we find that actual 3D diffusion to adjacent superconducting layers is required before the reduced T_c of the single film can regain its bulk value.

We gratefully thank S. Park and P. Broussard for valuable discussions during the early stages of this work. We also thank P. Broussard for guidance on proximity-effect calculations. In addition, we thank V. Matijasevic for assistance in sample preparation; A. Marshall and A. Schwartzman for TEM pictures; and H. Fukuyama, G. Bergmann, and A. Kapitulnik for discussions. This research was supported by the NSF Division of Materials Research through the Center for Materials Research at Stanford University.

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