Apparent Destruction of Superconductivity in the Disordered One-Dimensional Limit

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We present the results of a model-system study of the competition between superconductivity and disorder in narrow superconducting wires. As one moves from the two-dimensional regime toward the one-dimensional limit, large and systematic reductions in the superconducting transition temperature are obtained. The observed behavior extrapolates to the total destruction of superconductivity in the disordered one-dimensional limit. Our findings are in clear disagreement with a recent theoretical treatment. In addition, the superconducting fluctuations appear to be modified by disorder for the narrowest samples.

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It is now well understood that increasing disorder leads to Anderson localization and the related enhancement of the Coulomb interactions.[1,2] Such disorder-induced enhancement of the Coulomb interaction inherently competes with conventional superconductivity. In fact, it has been experimentally observed in homogeneous two- and three-dimensional systems that superconductivity is far more sensitive to the presence of disorder than are the normal-state properties.[3-8] Furthermore, since the effects of localization and increased Coulomb interactions upon the normal-state properties increase with decreasing dimensionality, the relevance of disorder upon superconductivity should become very large for few-dimensional systems. However, there exist serious discrepancies in our present understanding that require elucidation. The importance of a more complete understanding of the consequences of disorder cannot be underestimated. For example, the newly discovered high-temperature superconducting oxide systems are few-dimensional in nature and possess relatively large resistivities. Thus disorder may play an important role in their superconductivity. Finally, this work argues strongly that the effects of disorder should not be ignored in the important class of quasi-1D superconductors, including the organic systems.

In the weakly localized region, where the precursor effects of localization may be treated with perturbation theory, it is predicted for homogeneous systems that the reduction in the superconducting transition temperature, ΔT_c, normalized to the T_{c0} of the otherwise identical material, behaves as[9]

$$\Delta T_c/T_{c0} \sim (h/4\pi^2 E_F \tau)^{\mathcal{D}-1} .$$

Here $\mathcal{D}$ is the dimensionality, E_F is the Fermi energy, h is Planck's constant, and τ is the elastic-scattering time. Note that perturbation theory breaks down in 1D. Experimentally, the competition between superconductivity and disorder has been most closely studied in 2D,[4-8,10,11] where the observed effects are large and in general agreement with theoretical predictions.[12,13] In the more technologically important 3D case, the understanding has been hampered by several problems. For 3D systems in the region of weak disorder, it is experimentally difficult to distinguish nonlocalization disorder-induced effects upon T_c, e.g., the broadening of structure in the density of states and alterations of the phonon structure. Also, among the theoretical calculations there exist large disagreements in the predicted T_c suppression, which may originate in the handling of short-wavelength diffusion cutoffs.[14,15] It now seems reasonable that the agreement between theory and experiment is most likely for 2D, where there is only logarithmic sensitivity to such cutoffs. Thus, results in other dimensions are needed to advance our understanding of the consequences of disorder upon superconductivity. The relevant characteristic length for superconductivity in the presence of disorder is the thermal diffusion length $L_T \equiv (hD/2\pi k_B T)^{1/2}$, which is of order 100 Å for disordered metal at 4 K. D is the electron diffusion constant. As length scales below 1000 Å are now possible with state-of-the-art electron-beam lithography, the approach to 1D is now experimentally accessible. Finally, let us point out that we will restrict ourselves to the case of homogeneous systems, as opposed to inhomogeneous or granular systems for which the behavior is observed to be quite different.[16]

Our aim in this work is to study the impact of disorder upon superconductivity as we move continuously from the 2D regime toward the 1D limit by varying both the sample width w and the film thickness d. Our measurements were made upon a series of ultrathin amorphous superconducting $Mo_{0.8}Ge_{0.2}$ films, which were synthesized with advanced vapor-deposition techniques. The specifics of their synthesis and characterization appear elsewhere.[6-8] We have previously established that these films are homogeneous and exhibit constant equivalent bulk properties down to film thicknesses of 10 Å. Furthermore, the normal-state properties are consistent with predictions for the 2D weakly localized regime. The bulk zero-temperature coherence length [$\xi(0) \cong 50$ Å] and diffusion constant ($D \cong 0.5$ cm^2/s) are obtained from the upper-critical-field slope. From D we calculate that $L_T \cong 100$ Å at 4 K for our films.

These films were patterned with a trilevel electron-beam lithographic process[17] utilizing a 120-keV Phillips scanning transmission electron microscope with a 20-Å spot diameter. The pattern transfer and sample etching were done by means of a reactive plasma etching process, with the use of a mixture of CF_3Br and Ar to etch the Mo-Ge films. Extensive efforts were taken to rule out extraneous sources of the the observed behavior. To avoid sample-to-sample variations, up to ten different wires with different widths were written in an area of $(150\ \mu m)^2$ upon a single film. Each sample included a wide bridge ($w = 1$–$4\ \mu m$), which was used as a reference for the 2D superconducting transition. Note that all samples were exposed to the same electron-beam flux, and no evidence of damage was observed or expected for our amorphous films. Finally, the sample processing steps were varied extensively with no resultant change in the measured behavior. Subsequent scanning-transmission-electron-microscope examination was used to determine the actual wire width, w, and revealed no sign of sample voids or defects. Samples were fabricated in four-point configurations, and were typically 10–20 μm long.

The mean-field transition temperature of each sample, $T_c(d,w)$, was obtained from the measured resistive transitions. The sample current densities were kept low to avoid heating or current-induced vortex motion, and were in the range of 1–10 A/cm^2. No change in the resistive transitions was observed for current densities 10–50 times larger. Note that the characteristic critical currents for similarly fabricated thick films are of order 10^4–10^5 A/cm^2.

Figure 1 displays the results of a set of four wires written on a *single* film of thickness 50 Å. The data clearly display a systematic and monotonic decrease in the transition temperature as a function of decreasing sample width. The results were observed to be reproducible from sample to sample. Note that although the transition widths indicate increased fluctuations for the narrowest samples, there is a clear depression of T_c notwithstanding. Overall, reductions in T_c from the 2D-film value $T_c(2D)$ were discernible for wires as wide as 0.5 μm, while the largest observed reduction was 30% for the narrowest wire that we were able to fabricate ($w \cong 325$ Å). Near T_c the Aslamasov-Larkin (AL) contribution to the fluctuation conductivity, $\sigma'_{AL}(T)$, was observed to dominate the Maki-Thompson term for our films.[8,18] As $\sigma'_{AL}(T) \sim (T-T_c)^{-1}$ in 2D, the T_c for wires with

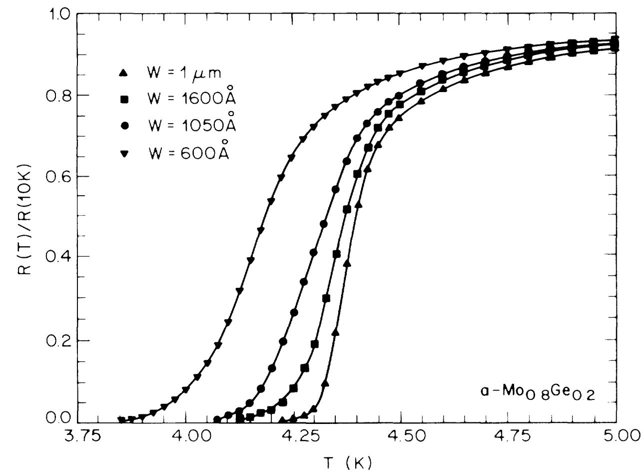

FIG. 1. Resistive transitions for a set of four wires with different widths w upon a single film of thickness 50 Å. Note the systematic decrease in T_c with decreasing sample width.

$w > 750$ Å was obtained from a linear extrapolation of the reciprocal excess fluctuation conductivity to zero. It was observed that the T_c obtained in this fashion was essentially coincident with the midpoint of the resistive transition. For decreasing sample width below 750 Å, the 2D AL behavior progressively breaks down, and the transition width becomes increasingly large. Here the T_c was estimated from the resistive midpoint. The increased fluctuations for the narrowest samples will be discussed in more detail later.

If we vary the sample width w for fixed film thickness, the reduction in the transition temperature, $\Delta T_c \equiv T_c(2D) - T_c(w)$, is observed to vary as w^{-2}. As shown in previous work, the transition temperature decreases with film thickness from the coupling to disorder.[6–8,10–13] Therefore, a useful manner to study the width dependence for different film thicknesses is to examine the normalized reduction in the transition temperature $\Delta T_c/T_c(2D)$. Figure 2 displays the results for $\Delta T_c/T_c(2D)$ versus sample width for several film thicknesses. As shown, we find $\Delta T_c/T_c(2D)$ to be *independent* of film thickness. Also shown is the apparent $(w/w_c)^{-2}$ behavior.

There are several points to be made regarding this behavior. First, the mean-field transition-temperature data extrapolate to zero [i.e., $\Delta T_c/T_c(2D) = 1$] at a critical sample width $w_c \approx 150$ Å, which is suggestively close to the thermal diffusion length L_T for the temperature range examined. As L_T is the characteristic length scale for the determination of the dimensionality, this extrapolation implies that superconductivity is *destroyed* by the presence of disorder in 1D. Second, the observed w^{-2} dependence is seemingly inconsistent with 1D behavior. For the normal-state properties, the relevant measure of disorder in 1D is the resistance per characteristic

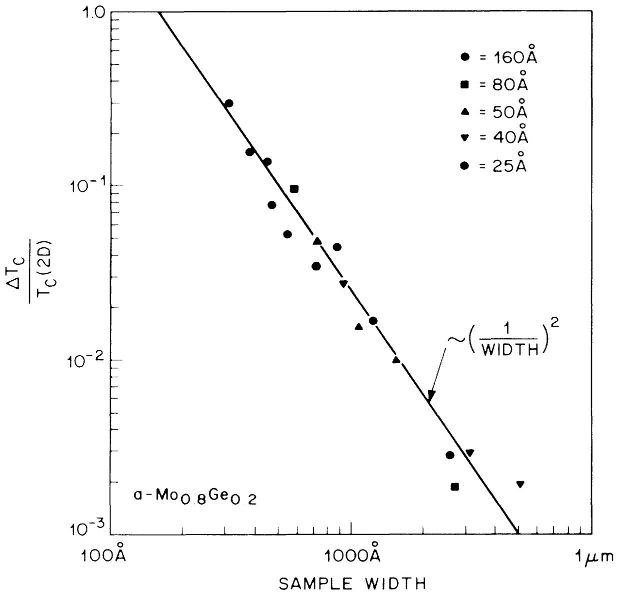

FIG. 2. Log-log plot of the normalized reduction in transition temperature $\Delta T_c/T_c(2D)$ vs sample width w, shown for several different film thicknesses 160 Å $\leq d \leq$ 25 Å. Note that the apparent $1/w^2$ behavior extrapolates to $T_c(w_c)=0$ for $w_c \cong 150$ Å. The $T_c(2D)$ for the films were $d=160$ Å, $T_c(2D)\cong 6.2$ K; $d=80$ Å, $T_c(2D)\cong 4.9$ K; $d=50$ Å, $T_c(2D)\cong 4.35$ K; $d=40$ Å, $T_c(2D)\cong 3.9$ K; $d=25$ Å, $T_c(2D)\cong 3.1$ K.

length.[1] Hence, this only involves the cross-sectional area of the sample (wd). More simply put, halving the sample width while doubling its thickness should have no effect upon the effects of disorder of 1D. The observed behavior clearly disagrees with this expectation.

Taken together, these two points indicate that we are not in the 1D limit. This is consistent with our evaluation of L_T being less than our sample widths. Thus, the observed w^{-2} behavior of $\Delta T_c/T_c(2D)$ is apparently characteristic of the *crossover* from 2D to 1D, extrapolating to the *destruction* of superconductivity in the disordered 1D limit. To be fair, we must point out that this w^{-2} extrapolation is empirical, and may well change closer to the 1D limit. However, the trend is immediate and clear.

The resistive transitions become increasingly broad for sample widths well below 1000 Å, indicating substantial fluctuations for the narrowest samples. For example, for a film with $d=160$ Å and $T_c(2D)=6.19$ K, the resistive transition width for the $w=4$ μm sample was approximately 40 mK, whereas by $w=550$ Å it had become almost 300 mK (with $T_c \cong 5.715$ K). To some degree such broadening is expected. Sufficiently close to the transition ($T>T_c$) the fluctuation correlation length $\xi^+(T) = \xi(0)(T/T_c-1)^{-1/2}$ becomes equal to about half the sample width, and the superconducting fluctuations cross over to 1D behavior.[18] For example, if $w=500$ Å and $\xi(0)=50$ Å, this crossover occurs at $T/T_c=1.04$. Such numbers are in good agreement with the observed broadening *above* T_c.

However, we also observe a slowly decreasing tail in the resistive transition extending increasingly *below* T_c for the narrowest samples. At present, we cannot explain this behavior. The unbinding of topological defects in 2D is known to produce resistance below the mean-field T_c.[19] Such unbinding would be expected to increase as the sample size approaches the temperature-dependent average vortex-antivortex separation. To apply to our case such finite-size effects would have to be very large, since for our low values of normal-state sheet resistance (100–700 Ω) the effects in 2D are very slight. Additionally, thermally activated phase-slip centers are known to produce finite resistance below T_c in 1D, although they have only been systematically studies for clean systems.[20] However, in this case one expects the resistance to fall off exponentially with temperature below T_c, which does not agree with the results of our narrowest samples. We point out that the role of quantum fluctuations is at this time not known.[21] Therefore, we may only conclude that the superconducting fluctuations are enhanced as the disordered 1D limit is approached.

Finally, let us examine the consequence of our findings upon quasi-1D systems, examples of which include transition-metal chalcogenide and organic systems. There are admittedly important differences. Our amorphous wires incorporate substantially greater disorder than high-quality quasi-1D crystals, so much so that true single-atom chains of our material would clearly be strongly localized. Quasi-1D crystals also possess neighboring chains in close proximity which result in important modifications of the screened Coulomb interaction compared with that of a single chain. However, it is important to note that we observe substantial effects upon superconductivity for cross-sectional areas 4–5 *orders of magnitude larger* than those of single chains in quasi-1D systems. We believe that the magnitude of the reductions observed in this work strongly suggests that the consequences of disorder cannot be ignored for superconducting quasi-1D systems.

Recently, Ebisawa, Fukuyama, and Maekawa calculated the leading-order correction to the transition temperature for narrow superconducting wires in the weakly localized regime.[22] This calculation contains the same fundamental physics as their 2D theory, which was in basic agreement with our previous results for $T_c(2D)$ vs $1/d$ for these same films. Their prediction for the T_c reduction in 1D behaves as $\Delta T_c/T_c(2D) \sim (wd)^{-1/2}$, which clearly disagrees with our results. Furthermore, their predicted reduction is many times stronger than our observations. With the use of parameters appropriate to our samples, their calculation yields substantial reductions ($\simeq 50\%$) for sample cross sections of order (3

μm)^2. Note that our experiment is not in the 1D limit, and as we approach it we progressively leave the weakly localized regime. Still, such a large qualitative and quantitative disagreement with experiment is clearly disconcerting. More importantly, it raises the question whether the theory requires "only" a careful reappraisal of the all-important cutoffs, or whether a more fundamental revision of the consequences of disorder upon superconductivity is in order.

In conclusion, we have made a systematic study of the competition of superconductivity and disorder in the approach to 1D. We observe a strong and systematic reduction in the transition temperature with decreasing sample size. As all indications are that we are not in the 1D limit, the observed behavior is apparently characteristic of the crossover from 2D to 1D. Furthermore, this behavior extrapolates to the complete destruction of superconductivity in the disordered 1D limit. Finally, our results strongly suggest that the consequences of disorder cannot be ignored for the quasi-1D superconducting systems.

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