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Localization and interaction effects in ultrathin amorphous superconducting films

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Ultrathin superconducting films of amorphous Mo-Ge were made that remain homogeneous down to ~ 20 Å. The expected localization behavior was observed in their resistivity-versus-temperature and sheet resistance ($R_\square$) data. A systematic reduction of the transition temperature with increasing $R_\square$ was observed for several compositions, consistent with recent theories of localization and Coulomb interaction effects on superconductivity. Critical-field data provide further tests of these theories.

With the growing understanding of the approach to Anderson localization, there is now much interest in how localization and the related changes in the Coulomb interaction affect the physical properties of disordered metals. Of particular interest is the effect on superconductivity, with which localization and increased Coulomb interactions inherently compete.[1,2] Recent theories predict that in two dimensions such competition will sharply reduce the transition temperature T_c with increasing sheet resistance $R_\square$.[1] Amorphous superconducting films hold special interest for examining such interplay. Amorphous metals combine microscopic disorder with homogeneity on the length scales for superconductivity (i.e., the superconducting coherence length ξ). This leads to considerable simplification of both the theory and the interpretation of experiment. Additionally, ultrathin amorphous films can be made two dimensional (2D) with respect to both superconductivity and diffusion. In this paper, we present the results of an experimental study of superconductivity in ultrathin amorphous Mo-Ge films. The films exhibit a strong reduction of T_c with increasing $R_\square$ as expected from theory. Important discrepancies, however, are observed between the present theory and experiment.

The a-Mo-Ge films we studied were co-magnetron-sputtered at ambient temperatures from single-element sources. Rapid substrate rotation (300 rpm) during deposition, with evaporation rates less than one monolayer per rotation, dramatically improved homogeneity and coverage for ultrathin films. Samples were prepared at various compositions and thicknesses, on different substrates, and with or without thin protective layers of a-Ge or a-Si. For films thicker than about 80 Å ($R_\square \leq 200\ \Omega$), both the resistivity ρ and T_c were independent of substrate or the existence of protective layers. This was not true for the thinner films. Specifically, ultrathin samples deposited in the same run on a-Si$_3$N$_4$ had lower resistivities than those on polished single-crystal sapphire, indicating more uniform coverage of a likely smoother surface. The presence of protective layers also led to lower resistivities and slightly higher T_c's for the thinnest films, presumably by the prevention of oxidation or other chemical interaction. Using the reasonable criterion, for a given composition, that the best films are those with the lowest ρ, a series of "optimized" samples were made on a-Si$_3$N$_4$ using a 10-Å a-Ge underlayer, successively sputtered Mo-Ge, and a 20–25-Å a-Si overlayer, which was then oxidized. All films were stable against thermal cycling and photolithographic processing. Analysis of selected samples with x rays and transmission electron microscopy showed no sign of crystalline inclusions. Tunneling into the thinnest films (≤ 100 Å) using the oxidized a-Si as the tunnel barrier showed sharp, single gap structure, no evidence for a proximity layer, and gap values as expected from weak-coupled BCS theory.

The data consist of many runs made over an extended period of time. The optimized samples comprise the most complete data and extend to the highest $R_\square$. Resistivity was measured at 300 K and at the resistive maximum (due to the presence of superconducting fluctuations) at low temperatures by the van der Pauw technique. Also, for the two thinnest films, $R(T)$ was measured up to 77 K, and exhibited the $\ln(T)$ dependence expected for 2D localization and/or interaction effects.[3] Superconducting T_c's were measured resistively on patterned samples, and were smooth and well defined. Figure 1 shows the low-temperature ρ vs $R_\square$ for the optimized samples. Note that ρ has risen only about 5% by $d=40$ Å, indicating highly uniform films. Indeed, the behavior of the rise in ρ with increasing $R_\square$, even down to 20 Å, is consistent with that expected from localization and interaction effects, with perhaps a small contribution from boundary scattering.[4] The comparison with localization and/or interaction theory is displayed in the inset, which shows the excess conductance $\Delta G_\square = -\Delta R_\square / R_\square^2$ vs $R_\square$. As can be seen $\Delta G_\square$ is constant, independent of $R_\square$ as expected from theory. The magnitude of $\Delta G_\square$ is also reasonable.[5] Hence there is no evidence that the inherent bulk resistivity of our films is increasing. The importance of this fact is that the bulk T_{c0} may depend strongly on ρ.[2,6] Clearly, in studying the dependence of T_c on $R_\square$ in thin films it is important to ensure that the bulk T_{c0} is itself unchanging. The maintenance of a constant bulk ρ is a reassuring (if not sufficient) condition that this is so. The possibility of interface effects affecting T_c is addressed below.

As seen in Fig. 2, for each composition there is a marked and systematic decrease in T_c from the bulk value, T_{c0}. For $R_\square$ less than about 400 Ω, the decrease in T_c is linear; for larger $R_\square$, T_c decreases more slowly. Recall that in this initial linear regime, for a given composition, the data (and

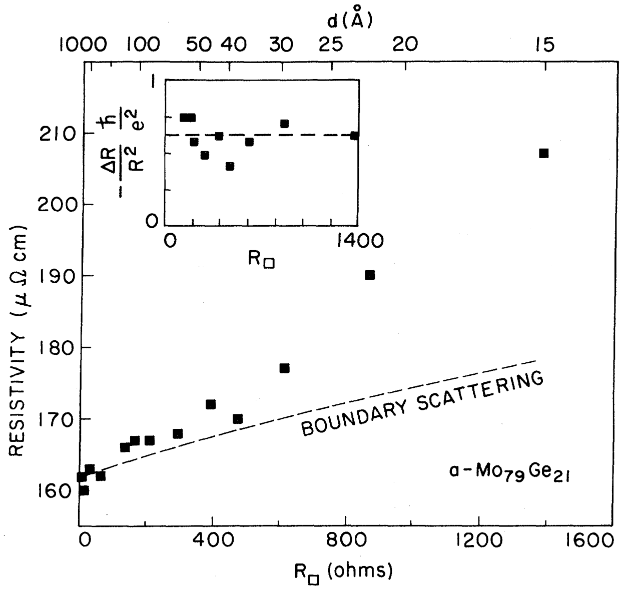

FIG. 1. Low-T ρ vs $R_\square$ (and d) for optimized samples, with estimated boundary scattering effects. Inset shows $\Delta G_\square$ vs $R_\square$ with

$$(\Delta R_\square/R_\square)=[\rho(\text{bulk})-\rho(R_\square)]/\rho(R_\square) \ .$$

thus the conclusions) are independent of sample preparation. A similar depression of T_c with increasing $R_\square$ in disordered non-transition-metal films was reported (but not definitively interpreted) long ago by Strongin, Thompson, Kammerer, and Crow.[7] Raffy, Laibowitz, Chaudhari, and Maekawa have reported a reduction in T_c with increasing $R_\square$ in a-W-Re alloys.[8] Hence the behavior is apparently universal.

Recently, Maekawa and Fukuyama (MF) theoretically studied the effects of localization on 2D superconductors.[1,9] Their perturbation-theory result is

$$\ln\left(\frac{T_c}{T_{c0}}\right) = -\frac{1}{2}\frac{g_1N(0)e^2R_\square}{2\pi^2\hbar}\left[\ln\left(5.5\frac{\xi_0}{l}\frac{T_{c0}}{T_c}\right)\right]^2 - \frac{1}{3}\frac{g_1N(0)e^2R_\square}{2\pi^2\hbar}\left[\ln\left(5.5\frac{\xi_0}{l}\frac{T_{c0}}{T_c}\right)\right]^3 \ .$$

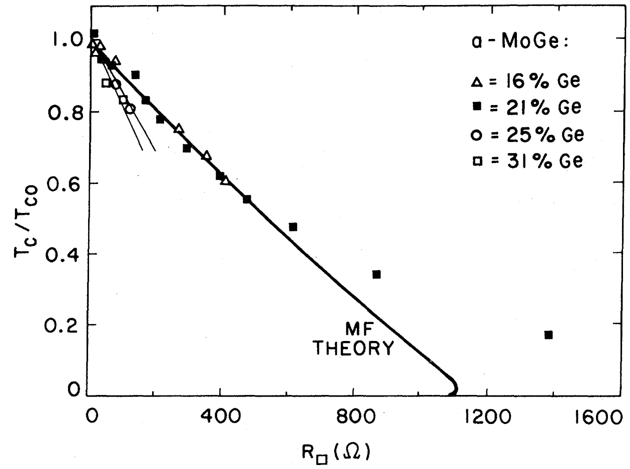

FIG. 2. (T_c/T_{c0}) vs $R_\square$ for various compositions, with fit to theory. Values of T_{c0} are 7.6, 7.2, 6.8, and 5.8 K for increasing Ge content.

The first term is due to the reduction of the density of states (correlation gap precursor) and the second is a (vertex) correction to the electron-electron interaction. Note that for our values of (ξ_0/l), the second term is the predominant contribution to the reduction in T_c. This vertex correction reflects an exchange correction to the electron screening, which is affected by the anomalous diffusion present in disordered metals. This reduction in screening enhances the renormalized Coulomb interaction parameter μ^*, thereby reducing the BCS coupling constant. To compare our results with theory we determine the product $\xi_0 l$ from the critical-field slope of our bulk (thick) samples $[\xi(0)\simeq 45\ \text{Å}]$. Making the reasonable assumption for the mean free path $l \sim 4$ Å (roughly an interatomic distance), the only remaining parameter is the effective coupling constant $g_1N(0)$, which is expected to be of order unity. Figure 2 shows the fit to the data. The fit gives a reasonable value for $g_1N(0)$ of about 0.6. This interpretation implies that the change in μ^* dominates the reduction of T_c. Note that the rate of the T_c depression increases with increasing Ge content. Finally, note that as $R_\square$ increases, the data deviate from theory, and there is no evidence for the predicted reentrant behavior. Thus, at least within this framework, the data suggest that higher-order (positive) corrections are required in the theory for large $R_\square$. Note that we cannot absolutely rule out possible influences of interface (i.e., proximitylike) effects on T_c due to the protective layers used with the thinnest samples. However, for such effects to account for the observed discrepancy with theory, it is necessary that such effects increase T_c—an unlikely but remarkable result if true. If such interface effects reduce T_c, then in our fit to theory we have overestimated $g_1N(0)$. It is worth pointing out that the measured T_c reduction is roughly an order of magnitude greater than that expected from the Kosterlitz-Thouless transition.[10]

Because of its intrinsic interest and as a further test of the theory, we also studied the perpendicular upper critical-field $H_{c2}(T)$ behavior of selected samples. Again, if one is to separate out effects on $H_{c2}(T)$ due to localization and interaction effects from those resulting from gross structure in the film, as in granular or percolation systems, sample uniformity is relevant. The data are shown in Fig. 3 along with fits (dashed lines) to the standard GLAG (Ginzburg, Landau, Abrikosov, and Gor'kov) theory.[11] The prediction of the theory of Maekawa, Ebisawa, and Fukuyama[12] (MEF) is shown in the inset for the same four samples. Within the MEF theory, once $g_1N(0)$ is determined by fitting the T_c data, $H_{c2}(T)$ is completely determined, thus providing a consistency check on the theory. Note that both the negative curvature near T_c and critical-field slope at T_c grow with decreasing T_c (increasing $R_\square$). Furthermore, the slopes at high field decrease slightly. Clearly, the data progressively deviate from the GLAG result. The deviations at high field are qualitatively different from the MEF theory as well. Some caution is required in this conclusion. As shown by the uncertainty bars on the data, the resistive transitions of

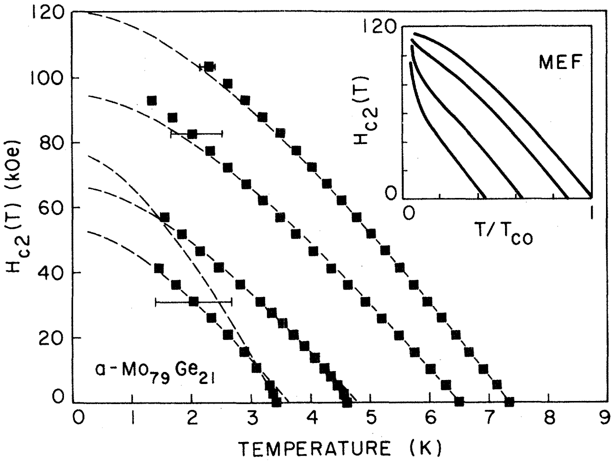

FIG. 3. Upper critical-field data, with GLAG fits (dashed lines). Inset shows MEF theory for same samples.

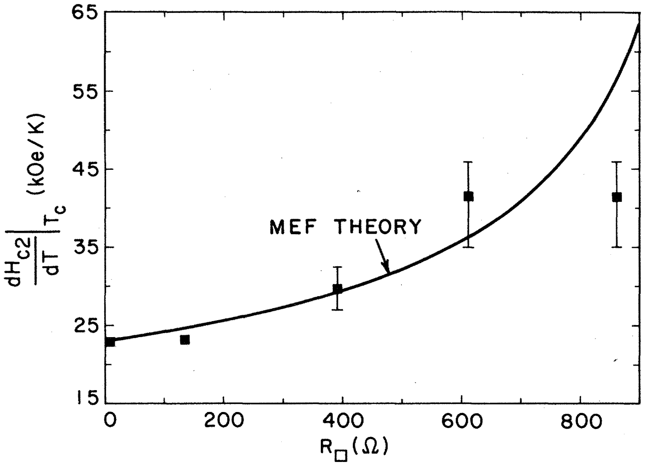

FIG. 4. Low-field critical-field slope, with theory (no free parameters). Error bars due to rapid rollover of $H_{c2}(T)$ with increasing H, not from an uncertainty in measurement.

the thinnest films broaden substantially at high field, making a precise determination of $H_{c2}(T)$ difficult. Such broadening is to a large extent expected due to intrinsic fluctuation effects (field-enhanced fluctuation conductivity above the transition, and the vortex lattice melting below).[13] Nevertheless, the discrepancy seems to be qualitatively real. An improved theory incorporating fluctuation and localization on an equal basis will be required before a quantitative test of the theory will be possible.

Therefore we looked at the critical-field slope at T_c, where we can better avoid the problem of large fluctuations. The data are shown in Fig. 4, along with the prediction of MEF, using the identical parameters found to fit T_c vs $R_\square$ curves. Note that the data agree well with the theory for the same range of $R_\square$ as the T_c fit. Hence the T_c and the low-field H_{c2} data can be interpreted self-consistently.

In conclusion, using carefully prepared ultrathin amorphous superconducting films, we have found that the superconducting transition temperature of the films decreases rapidly with increasing sheet resistance. The results are in general agreement with recent theories of localization and/or interaction effects in homogeneous 2D superconductors and demonstrate that the effects of disorder on superconductivity are dramatic, even in the weakly localized regime. Discrepancies with theory are found, however, which suggest that important corrections to the present theory are required at high magnetic fields and as one enters the more strongly localized regime.

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