# Microstructure, Dimensionality, and Depression of the Transition Temperature in Disordered Superconducting Films

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Composite In-$InO_x$ films are reproducibly fabricated with two classes of microstructure, islanded at low oxide content and amorphous-composite at high oxide content. The islanded films, near the percolation threshold, exhibit a two-dimensional depression of the mean-field transition temperature proportional to normal-state sheet resistance. The corresponding behavior in the amorphous composite films with similar resistivities is three-dimensional with a quadratic dependence on normal-state resistivity. Consistency with respective theory for each case is discussed.

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A characterization of how disorder leads to a depression of the mean-field transition temperature $T_{c0}$ of thin-film superconductors has been the objective of numerous recent studies on a variety of materials.[1,2] An attempt to unify the many disparate interpretations can be found in the recent work of Deutscher, Goldman, and Micklitz,[2] who suggest a crossover between percolation-dominated behavior of large grain structures and localization-dominated behavior of more finely textured structures. However, because of unknown exponents and scale lengths there is no straightforward operational procedure to facilitate such a classification even when detailed microstructural information is available. This problem is further compounded by the question of whether a film's dimensionality should be 2D or 3D.

In this Letter we directly address these issues with a presentation of data on composite In-$InO_x$ thin films with thicknesses ranging from 50 to 600 Å which have been fabricated to have either an islanded and/or granular or amorphous-composite morphology, as verified by transmission electron microscopy (TEM). The disorder-induced $T_{c0}$ depression $\Delta T_{c0}$ of the islanded films is found to be proportional to the normal-sheet resistance $R_N$ and is in good qualitative agreement with a percolation description[3] of a weakly coupled random 2D network of Josephson junctions, each junction defined at the shared boundary of neighboring islands. In contrast, $\Delta T_{c0}$ of the amorphous-composite films varies as the square of the room-temperature normal-state resistivity $\rho_N$, a first-time confirmation of the dependence predicted by Fukuyama, Ebisawa, and Maekawa[4] by use of perturbation theory in the weakly localized regime of a homogeneous 3D system. (Similar behavior in $Nb_3Sn$ and $V_3Si$ films[5] with significantly lower resistivities is most likely due to smearing in the density of states rather than intrinsic disorder which is considered in the microscopic theory.[4]) The underlying and more fundamental dependence of $\Delta T_{c0}$ on the inverse square of the product of the Fermi wave vector $k_F$ and the electron mean free path $l$ [i.e., $\Delta T_{c0} \propto -(k_F l)^{-2}$] is found to be overestimated in the theory by more than a factor of 6.

The films for this study were prepared by reactive ion-beam sputter deposition in which an indium target is argon sputtered in a partial pressure $p_{oxy}$ of oxygen. The film microstructure is directly related to the oxide content which increases with $p_{oxy}$, as illustrated in the logarithmic plot (Fig. 1) of $\rho_N$ vs $p_{oxy}$ normalized to the deposition rate, $r$. At low $p_{oxy}$, where the In-metal-adatom mobility is high, the films have a rough surface texture. The abrupt dependence of $\rho_N$ on $p_{oxy}$ in this region (left-hand portion, region I of Fig. 1) indicates that, for a *chosen thickness,* films can be fabricated near the percolation threshold where the resistance is extremely

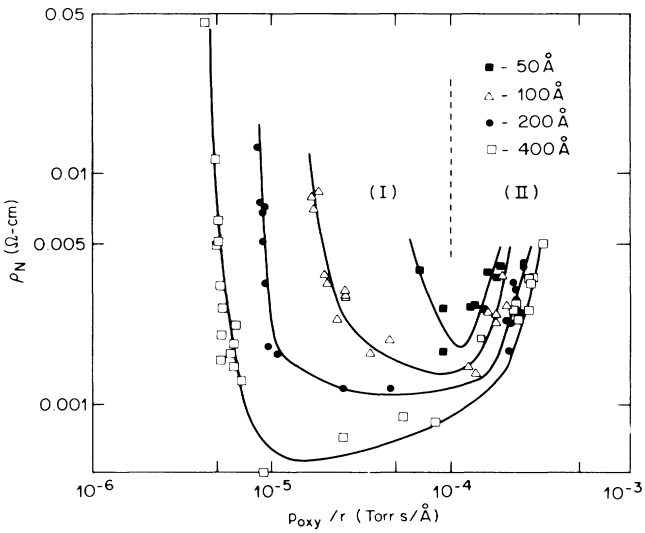


FIG. 1. Plot on logarithmic axes of the normal-state room-temperature resistivity $\rho_N$ of as-made In-$InO_x$ films vs the partial pressure of oxygen normalized to the deposition rate. The solid lines are guides to the eye for the four thicknesses indicated by the symbols. The dashed line separates two regions of behavior as discussed in the text.

sensitive to small changes in the microstructure. TEM micrographs of selected samples from each of these critical regions reveals a microstructure in which predominantly single-crystal disk-shaped islands of indium, with diameters comparable to the film thickness, are almost touching each other.[6] At higher $p_{oxy}$, the reduced adatom mobility associated with the presence of greater amounts of immobile oxide results in an agglomerated to granular to amorphous-composite transformation[7] concomitant with an increase in film smoothness, homogeneity, and resistivity. The vertical dashed line in Fig. 1 delineates the approximate boundary where this transformation takes place. To the right of this boundary, region II, where the oxide content is high, amorphous microstructure and depletion of the free-electron density cause the increase in $\rho_N$. The average oxygen concentration varies between 0 and 40 at.% in region I and 40 and 60 at.% in region II.

A 40% normal-state criterion[8] was used to determine the $T_{c0}$'s which were above the 1.2-K limit of the cryostat. The data corresponding to the rough-textured films located in region I of Fig. 1 are plotted in Fig. 2 as a function of $R_N = \rho_N/d$, where $d$ is the film thickness. Note that most of the points for the 100-, 200-, and 400-Å thick films are near critical percolation for each thickness. (Only one of the 50-Å films was superconducting with $T_{c0} > 1.2$ K.) Thus the solid-line regression fit to all of the data is a good indicator of the average $T_{c0}$ depression in this percolating system near the critical concentration. Using the theoretical result[4] for a percolating network of Josephson-coupled grains together with the temperature-dependent BCS energy gap, we find from a numerical calculation that the expression

$$T_{c0} = T_{cx} - 0.3e^2\rho_N T_{cx}/\pi\hbar L \tag{1}$$

is accurate to 5% in the interval $\frac{1}{2}T_{cx} \lesssim T_{c0} \leq T_{cx}$. The unknown characteristic length $L$ is related to microstructure and $T_{cx}$ is the transition temperature unrenormalized by disorder. Agreement with the solid line of Fig. 2 is obtained for $L = 0.3d$ and $T_{cx} = 3.68$ K, a value close to the 3.4-K bulk transition temperature of indium. Thus, good qualitative agreement with the theory[3] is obtained with the presumption that $L$, $d$, and the island size have comparable magnitudes, a presumption well supported by the TEM evidence.[6,7]

A strikingly different result, that $\Delta T_{c0}$ depends on $\rho_N$, rather than $R_N$, is exhibited by the data for the smooth-textured films, all points in region II of Fig. 1. The theoretical prediction for the $T_{c0}$ depression in 3D homogeneous films is given by[4]

$$\ln(T_{c0}/T_{cx}) = -[3\sqrt{3}/2\pi(k_F l)^2]\{(1/g^*)^2 + 2\pi[g^{-1} - (\mu^*/g)^2 \ln\omega_D\tau]\}, \tag{2}$$

where $g$, the BCS coupling constant, $\mu^*$, the effective Coulomb interaction, and $g^*$, a parameter defined in Ref. 4, are constants on the order of unity. This expression is valid only for electrons with low diffusivity $D$ which have an elastic-scattering rate $\tau^{-1}$ greater than the Debye frequency $\omega_D$ where the Debye energy $\hbar\omega_D$ is less than the Fermi energy $\varepsilon_F$. Magnetoconductance measurements on similar smooth-textured films,[9] in which the electron–inelastic-diffusion length at 5 K is found to be 100 Å and the interaction length $(\hbar D/kT)^{1/2}$ at 2 K is calculated to be less than 100 Å,

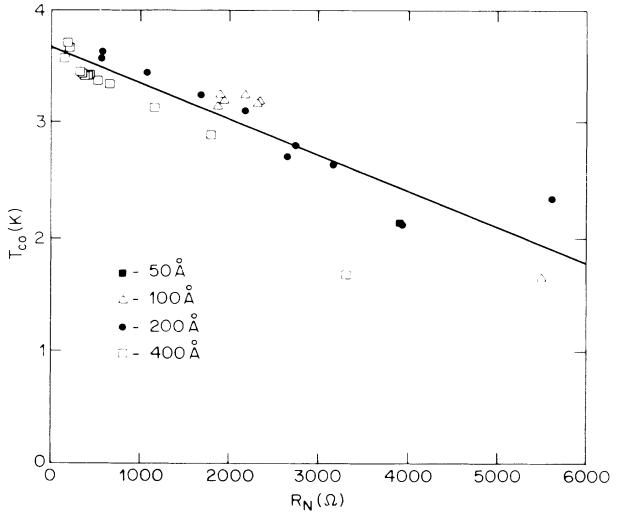


FIG. 2. Plot of $T_{c0}$ as a function of $R_N$ for the rough-textured films with thicknesses indicated by the symbols. The solid line is a regression fit to the data.

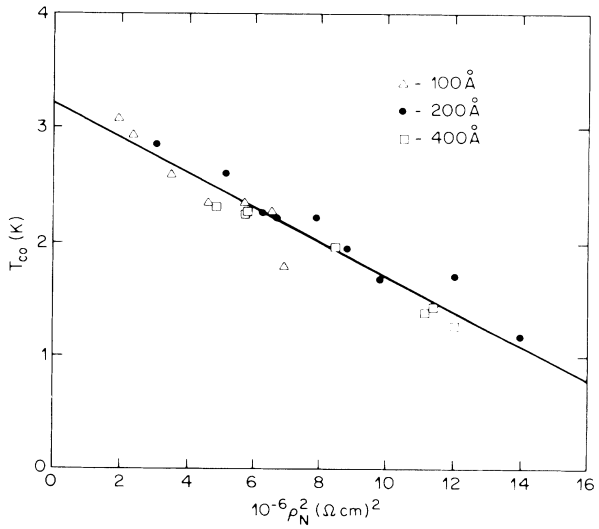


FIG. 3. Plot of $T_{c0}$ as a function of $\rho_N^2$ for the smooth-textured films with thicknesses indicated by the symbols. The solid line is a regression fit to the data.

also suggest the appropriateness of our using a 3D treatment to describe the disorder-induced depression of $T_{c0}$. We present, in the following, two schemes for testing Eq. (2).

First, on the assumption that for $T_{c0} \lesssim T_{cx}$ the density of states at $\varepsilon_F$ is only weakly dependent on disorder,[10] then $(k_F l)^{-2} \propto \rho_N^2$ and Eq. (2) predicts $\Delta T_{c0} \propto -\rho_N^2$. The agreement with such a 3D dependence on $\rho_N$ is demonstrated in Fig. 3. The solid-line regression fit of this figure extrapolates to $T_{cx} = 3.22$ K at $\rho_N = 0$, a value 0.18 K below the bulk transition temperature $T_{cB}$ of indium. Had a linear dependence on $\rho_N$ been used, $T_{cx}$ would be significantly higher, more than 0.8 K above $T_{cB}$. The evidence for a quadratic dependence is thus based on the notion that $T_{cx}$ should be close to $T_{cB}$. This is confirmed in annealing behavior, discussed below, in which $T_{c0}$ increases towards an *upper bound* no higher than $T_{cB}$.

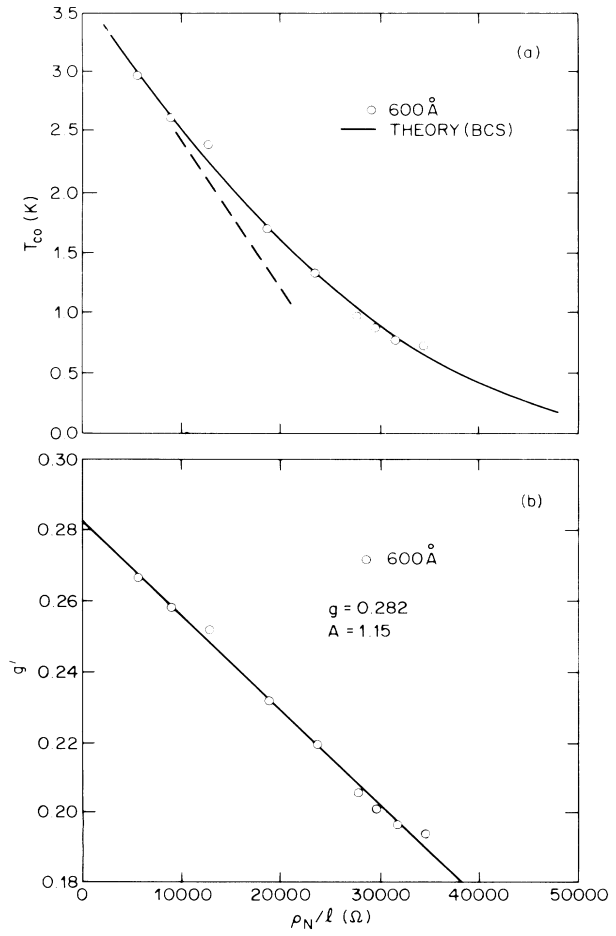


FIG. 4. Dependence of (a) $T_{c0}$ and (b) $g'$ on $\rho_N/l$ for a single annealed smooth-textured film. The solid lines are theory, Eqs. (3) and (4), that use the BCS equation and the dashed line is Eq. (5).

In our second method, $\rho_N$ is varied by annealing at temperatures $\lesssim 160\,^\circ$C and the corresponding value of $k_F l$ is determined by use of electric-field-effect measurements of mobility. We expand our interpretation of earlier data[11] in Fig. 4(a) which shows the dependence of $T_{c0}$ on $\rho_N/l$ for a single 600-Å-thick film. Note that since $(k_F l)^{-2} = e^2 \rho_N / 3\pi^2 \hbar l$ the initial slope of this plot, indicated by the dashed line, confirms the $(k_F l)^{-2}$ dependence predicted in the limit of weak disorder $(T_{c0} \lesssim T_{cx})$ by Eq. (2). Using $g = 0.28$ for bulk indium, together with the values $\mu^* = 0.18$, $g^* = 0.40$, and $\omega_D \tau = 0.18$, inferred from the data for this film,[12] we find that the theory overestimates the dashed-line initial slope in Fig. 4(a) by more than a factor of 6. As the discrepancy cannot be appreciably mitigated with any physically reasonable adjustment of these parameters, it appears that the 3D microscopic theory has overestimated the effects of Coulomb repulsion of paired electrons in the presence of disorder. The expansion parameter of the 3D perturbative theory,[4] defined as $1/\pi k_F l$, ranges from 0.068 for the film with the lowest value of $\rho_N/l$ up to 0.17 for the film with the highest $\rho_N/l$. Since the leading-order correction to $T_{c0}$ is proportional to the square of this parameter, which ranges from 0.0046 to 0.029, one expects the theory to be applicable. Similar theory[13] calculated for 2D, however, is in better agreement with results on thin films of W-Re,[14] Zn,[15] and Mo-Ge.[16] For the case of the Mo-Ge films, however, extrapolation to bulk thicknesses $(d \to \infty)$ and then use of 3D theory gives a $T_{cx}$ which is appreciably higher[17] than $T_{cB}$, a result consistent with our conclusion that the 3D theory[4] overestimates the $T_{c0}$ depression in bulk films.

An important and simplifying insight into the $\rho_N/l$ dependence shown by the data of Fig. 4(a) and predicted by the theory of Eq. (2) is shown by calculation of an effective BCS coupling constant $g'$ directly from the BCS equation,

$$T_{c0} = 1.13 \Theta_D \exp(-1/g'). \tag{3}$$

With the Debye temperature $\Theta_D = 112$ K for bulk In, the linear dependence shown in Fig. 4(b) resulting from this procedure validates an expansion of $g'$ of the form

$$g' = g[1 - A(k_F l)^{-2} + \cdots] = g(1 - Ae^2 \rho_N / 3\pi^2 \hbar l + \cdots), \tag{4}$$

where $A$ is a constant and $g$ is the BCS coupling constant unrenormalized by disorder. The values $A = 1.15$ and $g = 0.282$ determined from the linear fit shown in Fig. 4(b) are used with Eqs. (3) and (4) to calculate the solid curve shown in Fig. 4(a). The value $T_{cx} = 3.67$ K is calculated in the limit $k_F l \to \infty$. A direct comparison with the theory of Eq. (2) for $T_{c0} \lesssim T_{cx}$ can be made by substitution of Eq. (4) into Eq. (3) to find the leading

correction to $T_{c0}$, that is,

$$T_{c0}=T_{cx}-\frac{AT_{cx}}{g(k_Fl)^2}=T_{cx}-\frac{e^2AT_{cx}}{3\pi^2\hbar g}\frac{\rho_N}{l}. \quad (5)$$

As mentioned above, the experimental value for $A$ of 1.15 is a factor of 6 below the value 7.67 calculated directly from the theory. It is remarkable that only the leading term in the expansion of $g'$ accounts so well for the more than a factor of 4 experimentally observed disorder-induced depression of $T_{c0}$. The procedure outlined above parallels keeping the leading correction term in Eq. (2), where Fukuyama, Ebisawa, and Maekawa computed the correction to $(g')^{-1}$, except that here we have expanded $g'$. It is plausible to expect that the $(k_Fl)^{-2}$ correction in $g'$—which is proportional to the density of states—would be analogous in form to the localization and interaction corrections to the normal conductivity.

In closing, we emphasize the importance of dimensionality implied by the present results. Earlier work[1,18] on 100-Å smooth-textured films verified the Kosterlitz-Thouless interpretation of thermally excited vortices undergoing a 2D phase transition and unbinding at a temperature $T_c$ well below $T_{c0}$. These observations raise the most interesting question of how the effects (either 2D or 3D) of microscopic disorder on $T_{c0}$ are manifested in 2D vortex processes occurring over long length scales. We note that the power-law current-voltage behavior, which is definitive signature of vortex unbinding in our smooth-textured films,[18] is *not* observed in the low-resistance tail regions of our *inhomogeneous* rough-textured films, even though the $T_{c0}$ depression of these films is 2D in character. In conclusion, the present work has shown how inhomogeneous (rough texture, region I) disorder gives rise to a 2D $T_{c0}$ depression which is described well by percolation theory[3] and how homogeneous (smooth texture, region II) disorder gives rise to a 3D $T_{c0}$ depression which is quantitatively overestimated by heretofore-untested microscopic weak-localization theory.[4]

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