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Onset of high-temperature superconductivity in the two-dimensional limit

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The transition between insulating and superconducting behavior in the low-temperature limit has been explored with use of ultrathin Dy-Ba-Cu-O *c*-axis-oriented films prepared by molecular-beam epitaxy. The transition appears as a separatrix between sets of $R(T)$ curves that exhibit either insulating or superconducting behavior in the $T \rightarrow 0$ limit. Superconductivity is found only when the normal-state sheet resistance is below a value close to $h/4e^2$, or 6450 Ω. Hall-effect data relate the depression of T_c with increasing sheet resistance to a reduction in hole concentration.

The interplay between superconductivity and localization is a problem of fundamental interest. The phenomena are competing, with superconductivity being a manifestation of long-range phase coherence of electron-pair states, and localization, which is usually a consequence of disorder, being an effect in which coherent electronic behavior is spatially limited. As a result, one anticipates that as disorder increases, so that states become localized, superconductivity will disappear. Although there have been many experimental and theoretical studies of the interplay between localization and superconductivity, a complete detailed physical understanding has not yet emerged.[1] The case of thin films is special because the lower critical dimensionality is 2 for both superconductivity and localization. Thus, in two dimensions a superconducting phase transition is barely possible, and electronic states are localized even for arbitrarily weak disorder. The situation is different in three dimensions for metallic systems as a critical degree of disorder is needed to localize states. High-temperature superconductors are particularly interesting for a number of reasons. Their layered structure implies an inherent two dimensionality under some circumstances, and there is a connection between insulating and superconducting behavior, since doping the insulating parent compounds with charge carriers results in metallic superconducting behavior.[2]

In this paper we report a study of the onset of superconductivity in ultrathin films of Dy-Ba-Cu-O. These films were prepared *in situ* using molecular-beam epitaxy (MBE) employing techniques described elsewhere.[3] In contrast with previous work on normal metals where the onset of superconductivity was determined from the thickness dependence of the sheet resistance as a function of temperature $R(T)$ on incrementally grown films,[4,5] here $R(T)$ was altered both by fabricating separate films of different thicknesses, and by exposing the thinnest films to vacuum at room temperature for extended periods of time.

The issue of the existence of a sheet-resistance threshold for superconductivity was first raised in the context of films grown on glass substrates held at low temperatures where results on a number of different soft metals showed that superconductivity was found when the normal-state sheet resistance fell below a threshold value close to $h/4e^2 = 6450$ Ω.[4] In films which were not superconducting, a local minimum in $R(T)$ was observed near the bulk T_c and was the precursor to superconductivity as the sheet resistance was decreased from values larger than $h/4e^2$. The films, which are believed to consist of metallic grains coupled by Josephson tunneling, exhibited no substantial depression of their superconducting transition temperatures from bulk values, once the threshold was attained.

Smooth, nongranular films are believed to form, however, when Bi or Pb is deposited onto amorphous Ge substrates. In this case no local minimum in $R(T)$ was observed and T_c was suppressed substantially from its bulk value by a magnitude which was a function of film thickness. Furthermore, the threshold for superconductivity appeared as a separatrix between sets of $R(T)$ curves which exhibited insulating or superconducting behavior in the $T \rightarrow 0$ limit, with a resistance close to $h/4e^2$ at $T=0$ resembling an unstable fixed point of the various sets of curves $R(T)$.[5]

The issue of the onset of superconductivity has arisen recently in the context of high-T_c films. Hebard and co-workers[6] reported a limiting "electrical" thickness for superconductivity of the order of one lattice constant in ion-beam-thinned $YBa_2Cu_3O_7$ films. They observed that a crossover to nonsuperconducting behavior occurred when the value of the sheet resistance at 100 K, taken to be the normal-state resistance, exceeded $h/4e^2$. Earlier, Valles and co-workers[7] studied substantially thicker films (1700 Å) and found a critical resistivity induced by ion bombardment. In the present work, Dy-Ba-Cu-O films with as-prepared thicknesses less than 100 Å have been studied, and found to yield results which are in detail remarkably like the limiting behavior found for Bi films grown on amorphous Ge substrates at low temperatures. Not only is $h/4e^2$ an upper bound on the normal-state sheet resistance for samples which are probably superconducting as $T \rightarrow 0$, but it is also the resistance corresponding to the separatrix dividing $R(T)$ curves of superconducting and insulating films.

The thicknesses given here for ultrathin films are all nominal and are calculated assuming a growth rate identical to the growth rates of thicker films prepared under identical conditions. Although the films were prepared with deposition rates which would have given a ratio of [Dy]:[Ba]:[Cu] of 1:2:3, their actual compositions were somewhat different.[3] The detailed morphology of the thinnest films is not known. However, x-ray diffraction analysis on films as thin as 100 Å suggests that they consist mostly of *c*-axis-oriented crystallites. Transmission-electron-microscope (TEM) pictures of similar 100-Å films deposited onto prethinned substrates exhibit moire patterns extending over thousands of angstroms. These are disrupted by islandlike structures. Scanning-electron-microscope studies exhibited the same-size islands against a featureless background. Cross-sectional TEM studies of thicker samples revealed a sharp boundary between the $SrTiO_3$ substrate and the Dy-Ba-Cu-O film with well-defined lattice planes. These observations suggest that in the earliest stages of growth the films are ordered and there is relatively wide area, uniform coverage of the substrate followed by the development of islands as the thickness increases. A more detailed account of this work will be presented elsewhere.[8,9]

Electrical contacts were made by evaporating four 500-Å-thick silver pads, taking care to provide radiation shielding between most of the film surface area and the silver evaporation source, and pressing the bond wires to the silver pads. The sheet resistances were measured using the van der Pauw technique[10] with an estimated accuracy of about 2%. Films were cooled in a continuous-flow cryostat over the temperature range from 2.5 to 300 K. Normal-state resistances were determined in the linear region of the *I-V* characteristics with the current typically below 1 μA. Films were 0.6×0.6 cm in area. After a given film was loaded into the cryostat, the sample chamber was filled with N_2 gas and pumped down to 1 mTorr. Successive measurements of $R(T)$ were then taken without reopening the cryostat. For the thinnest films (<60 Å thickness), R in the normal state was observed to increase by about 500 Ω/day when allowed to remain in the sample chamber. Thus, the sheet resistance could be varied either by the production of films of different thicknesses or by allowing films to age. Films with thicknesses less than 30 Å were never superconducting and exhibited values of $dR/dT<0$ down to the lowest temperatures.

Sheet resistances as a function of temperature are shown for films of various thicknesses and at different stages of aging in Fig. 1. As R increases, T_c decreases and dR/dT decreases. As $R\rightarrow h/4e^2$ from below, dR/dT at high temperatures is close to zero. At low temperatures there is either a discernible superconducting transition, or for films close to the threshold $dR/dT>0$ at the lowest temperatures, suggesting an ultimate transition to the superconducting state at a lower temperature. However, if $R>h/4e^2$ (6540 Ω), $dR/dT<0$ at the lowest temperatures, suggesting that the films are insulating in the $T\rightarrow 0$ limit. The above-described behavior appears to be independent of the substrate, since qualitatively similar films have been produced on $SrTiO_3(100)$ and $LaAlO_3(100)$ substrates.

A second feature of the data is the dependence of the transition temperature on "normal-state" sheet resistance, which is shown in Fig. 2. The transition temperature has been arbitrarily taken to be the temperature at which the resistance falls below the noise level of the measuring system, and the normal-state sheet resistance was arbitrarily chosen to be that at 240 K. As dR/dT is

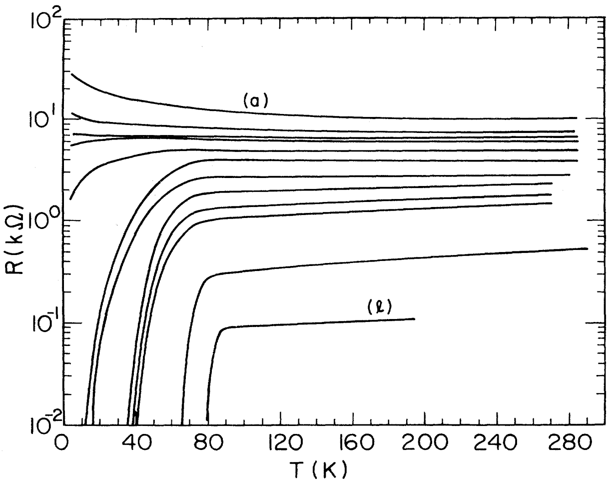

FIG. 1. Sheet resistance as a function of temperature for Dy-Ba-Cu-O films of various nominal thicknesses, some of which have been aged in vacuum at room temperature. Curve (e) is for a 35-Å-thick film deposited onto $SrTiO_3(100)$. Curves (b)–(d) and (a) result from successive aging steps. Curve (g) is for a 35-Å-thick film deposited onto $LaAlO_3(100)$, and curve (f) is the result of aging. Curve (j) is for a 40-Å-thick film deposited onto $LaAlO_3(100)$, and curves (i) and (h) result from aging. Curves (k) and (l) are films with thicknesses of 60 and 120 Å, respectively, deposited onto $SrTiO_3(100)$.

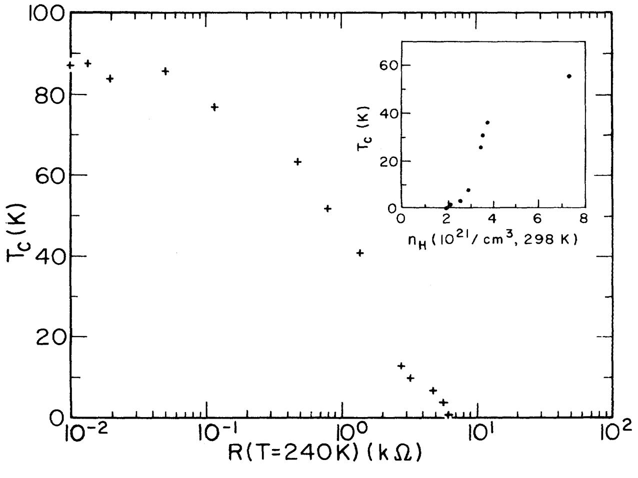

FIG. 2. Transition temperatures as a function of sheet resistance at 240 K for the superconducting films of Fig. 1. The inset shows the variation of T_c with hole concentration for similar films where the hole concentration has been determined using measurements of the Hall effect.

close to zero for values of the resistance approaching $h/4e^2$, the precise temperature at which the normal-state resistance is determined appears to be unimportant. These data show that $h/4e^2$ is an upper bound on the normal-state sheet resistances compatible with superconductivity at an attainable temperature, or a possible extrapolation to superconducting behavior at low temperatures. Given the fact that measurements down to 2.5 K represent a reduction of the temperature to roughly 0.025 of the bulk transition temperature of Dy-Ba_2-Cu_3-O_7, and that measurements on Bi films taken down to 0.45 K were only 0.075 of the transition temperature of thick amorphous Bi films, the present measurements may be considered to be equivalent to a threefold fractional reduction of the lowest temperature explored previously in studies of low-T_c films. Even this increase of fractional reduction in temperature is not sufficient to ensure against the possibility of unexpected effects occurring below the temperature range spanned by the measurements.

The above considerations suggest that the resistance threshold for superconductivity in Dy-Ba_2-Cu_3-O_7 thin films resembles that of ultrathin films considered to be nominally smooth, rather than that of granular films. In the latter, the superconducting transition appeared to be a two-step process, first involving the establishment of an equilibrium order parameter on the grains, followed at low temperatures by phase coupling of the grains.[4] The phase coupling leading to superconductivity appears to be controlled by the normal-state resistance, with the same threshold value as is found here. In granular films there was also a ubiquitous local minimum in $R(T)$ in high-resistance films which is either associated with percolative processes or with phase fluctuations.[4]

One type of anomalous behavior not shown in Fig. 1 was observed in the form of a film with a sheet resistance of 4200 Ω at room temperature where the resistance monotonically increased with decreasing temperature, reaching a value of 8700 Ω at about 10 K before beginning to fall. The curve of $R(T)$ for this film would cross several of the curves in Fig. 1, and it is possible that film might have become superconducting below the lowest available temperature of 2.5 K, despite R being greater than $h/4e^2$ at the maximum in $R(T)$. This behavior is suggestive of the film having many crystallites with their c axes oriented in the substrate plane, which unfortunately was not confirmed by structural studies before the film was destroyed. It is well known that in single crystals of $YBa_2Cu_3O_7$, the temperature coefficient of resistance is negative below about 200 K if the current is along the crystal c axis.[11] Furthermore, the crystalline orientation of a film is affected by small changes in substrate temperature during growth.[12] In fact, our substrate-heater configuration was changed slightly between the time that the anomalous film described above was grown, and the growth of all the films with $R(T)$ displayed in Fig. 1. For transport along the c axis, which would presumably not be metallic near T_c, the entire concept of a resistance threshold of the type discussed here might not be meaningful. This issue requires further experimental study of samples with confirmed in-plane orientation of the c axis.

It should be noted that although the thin films shown in Fig. 1 have their c axes oriented perpendicular to the plane, they do not exhibit large positive values of dR/dT characteristic of thicker films. Furthermore, values of dR/dT, although positive, are not large and fall to zero as the sheet resistances approach $h/4e^2$. Thus the films become less "metallic" as they approach the threshold. These observations, on as-prepared and aged films, are different from those on the ion-beam-thinned films studied by Hebard and co-workers,[6] where the metallic behavior appears to persist until the threshold is achieved. The differences between the two types of films will require detailed intercomparison of their microstructures which may be different because of different approaches to processing.

An additional technical point concerns what actually happens when the films are aged at low pressures. the most obvious process that would produce an increase in the sheet resistance and a reduction of the superconducting transition temperature would be the loss of oxygen by diffusion.[13] This is likely, given that Hall-effect measurements reveal that the hole concentration falls as the films are made thinner or altered by aging. Shown in the inset of Fig. 2 is the reduction of T_c with decreasing hole concentration. These Hall-effect studies show that the hole concentration has a limiting value of 2×10^{21} cm^3 in the limit of zero transition temperature. If there were no similar reduction in carrier concentration in the low-T_c case, which is actually not known experimentally, these results would suggest that the mechanisms for T_c suppression are not the same in high- and low-T_c superconductors although their superconductor-insulator transitions are similar. In the low-T_c case suppression is associated with a reduced density of states.[14]

In summary, the behavior of the electrical transport properties of ultrathin c-axis-oriented Dy-Ba-Cu-O films as a function of thickness is reminiscent of that of quench-evaporated, smooth, low-T_c superconducting films. With the caveat that definitive results may require study at lower temperatures, the sets of curves of $R(T)$ for various films resemble renormalization flows to an unstable fixed point at $T=0$ and at a resistance close to $h/4e^2$ as was found for Bi, the only amorphous film studied previously, and the only metal studied previously which does not form a superconducting alloy with Ge.[15] The value $h/4e^2$ is not necessarily universal, but may be a limiting value of the threshold resistance, as the corresponding resistances for Pb and Al were observed to be slightly higher.[5] The matter of transport along the c axis is not addressed in this work and remains an open question. The present results nevertheless suggest that the explanation of the superconductor-insulator transition in high-T_c films will require arguments other than those based on Josephson-coupled arrays of low-capacitance junctions which are discussed in Ref. 4. The latter theories are, however, very useful models of the behavior of arrays of low-capacitance tunneling junctions fabricated using lithographic techniques.[16] Promising general arguments may be those based on localization and quantum transport in two dimensions,[17] or a phase transition at $T=0$ associated with charge-flux duality in two dimensions.[18,19] In the case of the latter, a proof of exact duality in zero magnetic field, required for the threshold to be precisely $h/4e^2$, has not yet been found.

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