Scaling of the Insulator-to-Superconductor Transition in Ultrathin Amorphous Bi Films

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Study of the electrical conductances of a series of ultrathin Bi films as a function of increasing thickness has revealed behavior which can be correlated with the onset of superconductivity, even in the insulating state. The conductances of these films scale empirically with a single parameter which falls to zero when the films become superconductive. These observations suggest a direct transition between insulating and superconducting behavior. This transition occurs at a normal-state sheet resistance close to the quantum resistance for pairs, $R_q = h/4e^2$.

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The study of the interplay between superconductivity and localization in the two-dimensional limit is perceived as a fascinating problem for two reasons [1]. First, the two phenomena are evidently antithetical, as superconductivity follows from circumstances in which the charge carriers exhibit coherent behavior over macroscopic distances, whereas localization, especially in the strong limit, refers to the restriction of such behavior to a microscopic spatial scale. Second, the case of two dimensions is special in that it is the lower critical dimensionality both for superconductivity and for the insulator-to-metal transition.

One approach to the problem of the interplay between superconductivity and localization has been to investigate the thickness dependence of superconductivity in ultrathin films. In the case of alloys [2] and intermetallic compounds [3,4], it has been observed that the transition temperature drops with decreasing film thickness and superconductivity disappears at apparently arbitrary values of the sheet resistance in the normal state. Theoretical models which have been fitted with this behavior consider the reduction of the screening of the Coulomb interaction with increasing disorder and sheet resistance which accompany decreasing thickness [5,6]. However, these models are actually valid only in the weak-scattering limit and thus may not provide realistic predictions for the disappearance of superconductivity.

The notion of a superconductor-insulator phase transition has emerged from efforts to interpret very detailed measurements of the thickness dependence of the resistance of sets of amorphous Bi films deposited onto amorphous Ge substrates held at liquid-helium temperatures. In these studies an abrupt loss of superconductivity was observed. Curves of resistance versus temperature for different films exhibited either insulating or superconducting behavior in the limit of $T \rightarrow 0$. When plotted together on the same graph the apparent separatrix between the curves of the superconducting and insulating films was very close to the quantum resistance for pairs, $h/4e^2$ or 6455 Ω [7,8]. Similar results have been found for ultrathin films of $DyBa_2Cu_3O_{7-x}$ [9].

Theoretical models of disordered thin films treating Cooper pairs as bosons describe a superconductor-insulator transition at $T=0$ driven by quantum fluctuations with disorder as the control parameter for the transition [10–12]. This framework predicts a magnetic-field-driven transition [13] which appears to have been observed experimentally [14]. It should be noted, however, that tunneling studies on quench evaporated films, which indicate that the order parameter drops continuously to zero as films become insulating, would appear not to support this approach [15].

In this Letter we present evidence supporting the idea that in zero magnetic field, in the limit of zero temperature, there is a direct insulator-to-superconductor transition. The electrical transport in a sequence of 31 sequentially deposited amorphous Bi (*a*-Bi) films of increasing thickness grown on amorphous Ge (*a*-Ge) substrates exhibits behavior which anticipates the onset of superconductivity. In particular, a scaling function has been found for the conductivity, characterized by a parameter which vanishes with the onset of superconductivity. One might expect such scaling as a natural consequence of a direct phase transition between insulator and superconductor. One-parameter scaling was also found for nonsuperconducting Pd films which were fabricated for comparison with the superconducting films [16]. In that instance, however, the characteristic parameter fell to zero in the limit of zero resistance at high temperature, in a manner very different from the behavior reported here for Bi films.

Samples were prepared by incremental deposition of vaporized metals supplied by effusion cells in a high-vacuum environment onto substrates held at temperatures lower than 18 K [7]. Small amounts of metal were deposited in sequential steps resulting in a set of films with thickness increments so small that successive films usually overlapped in the range of conductances within the accessible temperature range (0.4–14 K). Great care was taken during the study of a particular metal to keep the substrate and source temperatures as well as the vacuum conditions constant during sequential evaporations. The

substrates were prepared by depositing *in situ* an approximately 6-Å-thick layer of *a*-Ge (about a monolayer) onto glazed alumina substrates. The films of Bi, subsequently deposited, wet the *a*-Ge and coat it uniformly, although the procedure may not guarantee that they are single phase [17]. The films of Pd were deposited directly onto glazed alumina substrates, as it is known that Pd will coat such surfaces uniformly [16]. Thicknesses were nominal and were derived from readings of a calibrated crystal-oscillator monitor which is directly sensitive to deposited mass per unit area. Resistance was measured as a function of temperature using a four-point probe technique employing preevaporated electrical leads. The *I-V* characteristics were linear. The details of these experimental techniques have been described elsewhere [18].

Figure 1 shows plots of $\log G$ vs $\ln T$ for a number of different *a*-Bi films of various thicknesses. The parameter G is the conductance in units of $4e^2/h$ and T is the temperature. The range of behaviors includes films which exhibit strongly activated conduction as well as the first few superconducting films in the sequence. The primary observation is that for the insulating films, the curve of G vs $\ln T$ of each individual film can be shifted along the $\ln T$ axis to overlap that of the film adjacent to it in the sequence of films. This operation can be applied successively to *all* insulating films in the sequence over the *entire* temperature range. Mathematically, this implies that the electrical conductance can be written in a scaled form, $G=G(T,d)=G(T/T_0^I(d))$, where d is the film thickness and T_0^I is a characteristic parameter, with the superscript I referring to insulating behavior. The ratio of the values of T_0^I of two adjacent films was then determined by the shift on the $\ln T$ axis required to superpose their plots of G vs $\ln T$. The actual values of T_0^I were found by noticing that for the thinnest films the conductance follows the form $G=G_0\exp(-T_0^I/T)$, with G_0 approximately equal to 0.174, independent of thickness. Although the physical origin of this form is unclear, it provides a better fit than any of the usual choices associated with hopping conduction. Then, starting with the value of T_0^I for the thinnest film, determined directly from the fit, values of T_0^I for all the subsequent films were determined by successive multiplication by the ratios of the scaling parameters for adjacent films. Once T_0^I was determined for each film, all of the data collapsed onto a single curve [19]. This procedure worked over a wide range of conductances from the regime in which they were exponentially activated functions of temperature well into that in which they were nearly logarithmic functions.

The superconducting films with thicknesses near the superconductor-insulator crossover were analyzed in an analogous manner employing the parameter T_0^S. It was found that by shifting data of G vs $\ln T$, the low-temperature tails of the various conductance curves could be brought together to follow an envelope trajectory. For those films having sufficiently long conductance tails at low temperatures, the data collapsed onto a single curve. Values of T_0^S were found by noticing that the low-temperature tails of films which were superconducting, but were close to the threshold for the transition to nonsuperconducting behavior, fell to zero as $\exp(-T_0^S/T)$. Using T_0^S of one of these films as a reference, values of T_0^S for the other films were determined without any additional adjustable parameters. In this instance, however, the high-temperature conductivity data did not collapse onto a single curve.

The final result of the above procedure is illustrated in Fig. 2. There, $\log G$ is plotted against $\ln(T/T_0^S)$ and $\ln(T/T_0^I)$ for 31 insulating and 10 superconducting films, respectively [20]. Comparing Figs. 1 and 2, one can see how dramatically the data can be collapsed onto two separate curves.

In Fig. 3, values of both $\log T_0^I$ and $\log T_0^S$ for the separate *a*-Bi films are plotted as a function of the quan-

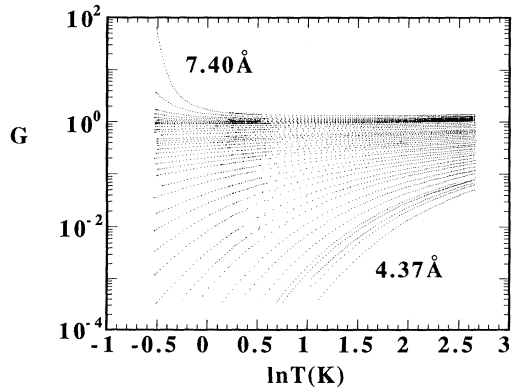

FIG. 1. Logarithm of the conductance G, in units of $4e^2/h$, vs $\ln(T)$ for a number of different Bi films. The thickness of the first (thinnest) and last (thickest) films are indicated.

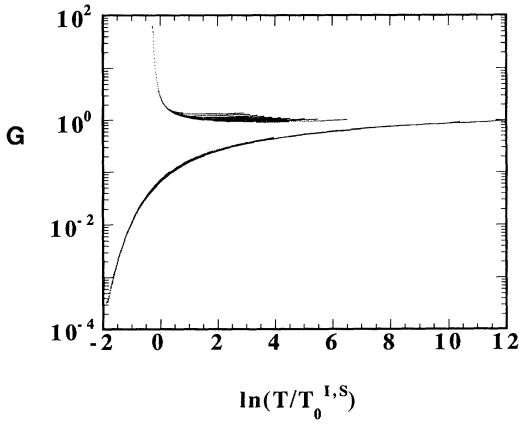

FIG. 2. Logarithm of the conductance G vs $\ln(T/T_0^I)$ and $\ln(T/T_0^S)$ for insulating and superconducting Bi films. The upper and lower sets of curves correspond to T_0^S and T_0^I, respectively.

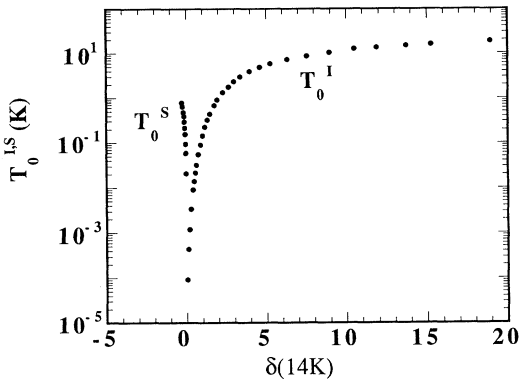

FIG. 3. Plots of $\log T_0^I$ and $\log T_0^S$ vs $\delta(14\text{ K})=R/R_0-1$, where R is the sheet resistance at $T=14$ K and R_0 is the algebraic average of the sheet resistances at 14 K of the last nonsuperconducting film and first superconducting film. The value of R_0 is 6.42 kΩ which is very close to the quantum resistance for pairs, $R_q=h/4e^2$.

tity $\delta(14\text{ K})=R/R_0-1$, where R is the sheet resistance of a given film at 14 K and $R_0=6.42$ kΩ is the algebraic average of the resistances of the last nonsuperconducting and first superconducting films of the sequence, evaluated at a temperature of 14 K. This value of R_0 is very close to the quantum resistance for pairs $h/4e^2$. The striking result of Fig. 3 is that both T_0^I and T_0^S fall abruptly to zero near $\delta(14\text{ K})=0$. In contrast, the parameter T_0 determined for Pd films (not shown) fell continuously to zero as $R\rightarrow 0$ [16].

One can fit the limiting variation of T_0^I, with $\delta(14\text{ K})$ near zero, using the form $T_0^I\sim[\delta(14\text{ K})]^i$. The exponent i is found to be 2.8 ± 0.4. The error bars are estimated from the values of the exponents obtained using different fitting ranges. The corresponding analysis for $T_0^S\sim[-\delta(14\text{ K})]^s$, for which the data are less extensive, yields $s=1.5\pm0.2$. Although the resistance, even at 14 K, is temperature dependent ($dR/dT<0$), this dependence is so weak, especially for films close to the onset of superconductivity, that $\delta(14\text{ K})$, as defined, is a reasonable choice for the control parameter. To illustrate this, if one repeats the above analysis using resistances measured at 10 K, the values of i and s are found to be 2.8 ± 0.3 and 1.4 ± 0.2, respectively, which, within the experimental error, are the same values obtained using resistances measured at 14 K.

Within the experimental error, the exponents i and s differ by a factor of 2. This lack of symmetry between the nonsuperconducting and superconducting forms may be intrinsic. It might also follow from the possibility that the data for superconducting films are not in the critical regime of the hypothesized phase transition. To be in this regime requires that a film be both close to the threshold and at sufficiently low temperatures. Comparison of plots of T_0^I and T_0^S vs $\delta(14\text{ K})$, as shown in Fig. 3, suggests that it is possible that the asymptotic limit is achieved for insulating films but not superconducting ones. An additional possibility is that the physics probed by the resistance measurements is different for insulators and superconductors. A final possibility relates to the control parameter for the transition. It has been assumed that the changing of thickness in experiments of this type only affects the degree of disorder of the film. If it were also to change the carrier concentration, as has been indicated by related experiments [9], then the failure to obtain symmetric results might be attributable to not having identified the correct scaling variable. However, results essentially identical to those shown above are obtained if the thickness is used in place of the sheet resistance as the control parameter.

The absence of physical and chemical inhomogeneity at mesoscopic length scales was clearly essential for the above scaling analysis to work. It was not possible to scale accurately data for systems of Al and Pb films also grown on a-Ge substrates. There is direct evidence from the presence of small, discontinuous features in their resistive transitions that these sets of films contained admixtures of more than one superconducting phase.

The essence of a phase transition, apart from its direct thermodynamic manifestations, such as heat-capacity anomalies, is the scaling of appropriate physical variables in its vicinity. We have observed scaling of the conductances of insulating and superconducting films, and the vanishing of the parameters characterizing this scaling at the insulator-to-superconductor crossover. These observations are evidence that there is a direct transition between insulating and superconducting behavior at the critical resistance $h/4e^2$. The full understanding of this transition will require study of films with parameters even closer to the transition than those reported here, as well as investigations which extend to much lower temperatures.

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[20] It should be noted that the analysis leading to Fig. 2 was carried out using a linear representation of G, although the plot in Fig. 2 displays $\log G$.