

Superconductivity-Related Insulating Behavior

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We present the results of an experimental study of superconducting, disordered, thin films of amorphous indium oxide. These films can be driven from the superconducting phase to a reentrant insulating state by the application of a perpendicular magnetic field (B). We find that the high- B insulator exhibits activated transport with a characteristic temperature, T_I . T_I has a maximum value (T_I^p) that is close to the superconducting transition temperature (T_c) at $B = 0$, suggesting a possible relation between the conduction mechanisms in the superconducting and insulating phases. T_I^p and T_c display opposite dependences on the disorder strength.

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In the realm of electrical conductors, insulators and superconductors belong to the opposite extremes. While both are a result of the quantum mechanical nature of the conduction process, the details of this process are markedly different. In a superconductor, a zero-resistance, quantum many-body, state emerges at low temperatures (T) inhibiting all scattering of electrons that can produce resistance. On the other hand, in an insulator, the motion of electrons is greatly hindered rendering them unable to carry current in the $T = 0$ limit. At low T , and at zero magnetic field (B), superconducting thin films have an immeasurably low resistivity (ρ). An application of B perpendicular to the film's surface weakens superconductivity until, at a well-defined critical B (B_c), superconductivity disappears and an insulating behavior sets in. This B -induced superconductor-insulator transition (SIT), believed to be a continuous quantum phase transition [1,2], has been the subject of a large number of studies, both theoretical [3–5] and experimental [6–10].

In this Letter, we focus on the reentrant insulator terminating the superconducting phase. Two major findings will be emphasized. First, activated transport is observed over a wide range of B , above B_c , firmly establishing the presence of a well-defined insulating phase. Second, the maximum value of the characteristic temperature for activated conduction at the insulating peak, T_I^p , is close to the superconducting transition temperature, T_c , of the film at $B = 0$, reflecting on a possible relation, and perhaps on the common origin, of the superconducting and insulating phases [11,12].

Our data were obtained from studies of disordered thin films of amorphous indium oxide (a :InO). We prepared the films by e -gun evaporating high purity (99.999%) In_2O_3 onto clean glass or sapphire substrates, in a high vacuum system. The thicknesses of the films used in the present study were between 200–300 Å as measured *in situ* by a quartz crystal thickness monitor. Transmission electron microscopy studies revealed that all the samples were amorphous and crystalline inclu-

sions were never observed. Most samples were lithographically defined to Hall-bar patterns. The voltage probes separation (L) was twice the width (W) of the Hall bar. One sample (Na1c) was patterned in a shape of a meandering line such that $W = 2200 \times L$ (with $L = 25 \mu\text{m}$), which enabled us to measure high ρ values in the insulating state, though low ρ values could not be measured with this two-probe geometry. On our Hall-bar samples, four-probe ρ measurements were carried out by dc and/or low frequency (2–13 Hz) ac lock-in techniques, with excitation currents of 10 pA–10 nA. The samples were cooled either in a dilution refrigerator with base T of 70 mK or in a He-3 refrigerator with base T of 250 mK. One of the samples (Ma1) was subjected to heat treatment in vacuum at $\sim 60^\circ\text{C}$ for a few hours. This annealing process causes a slow decrease of ρ , resulting in a reduction of the room temperature ρ of the sample Ma1 from 2.35 to 1.79 k Ω . As long as the annealing is carried out in vacuum, Hall measurements show that the carrier concentration remains unchanged [13]. Therefore, the decrease of ρ in a :InO films has been attributed to increased mobility due to a reduction of the static disorder [14].

We begin the presentation of the data with a description of the nature of our films at $B = 0$. In Fig. 1, we plot two ρ versus T curves taken from a single a :InO film. The top curve, taken from the as-deposited film, is typical of an insulator: As T is lowered from 6 K, ρ increases monotonically and seems to diverge as $T \rightarrow 0$. To reveal more information on the insulating behavior, we replot, in the inset of Fig. 1, the same ρ data but this time versus $1/T$. The dashed line is a fit of the data to an Arrhenius behavior,

$$\rho = \rho_0 \exp(T_I/T), \quad (1)$$

indicating the existence of a mobility gap at the Fermi energy, with $k_B T_I$ the magnitude of the gap (k_B is the Boltzmann's constant and T_I is the activation temperature). For this curve, $T_I = 4.3$ K. The bottom curve in Fig. 1 was obtained after annealing the film as described

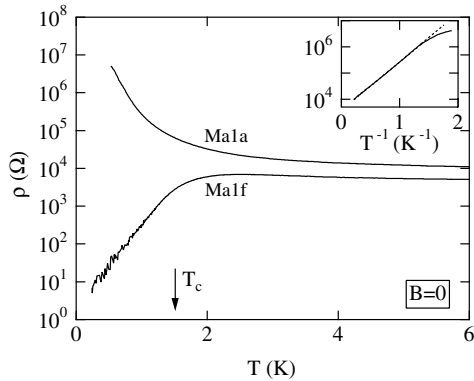


FIG. 1. Zero magnetic field ρ versus T curves of a single film at two different stages of annealing, Mala and Malf. Mala is insulating with $T_I = 4.3$ K. Inset: ρ versus T^{-1} curve for Mala (solid line), and the fit to Eq. (1) (dashed line). At low T , deviation from the activated behavior is observed. Vertical arrow in the main figure marks T_c ($= 1.5$ K) of the superconducting film Malf.

above. The annealing process is similar to that used in [15] to tune the film's properties. The effective disorder in the film has been significantly reduced by the annealing making it fully superconducting with a broad (0.9 K) transition into a superconducting state at $T_c = 1.5$ K. We note that the film's resistivity continues to drop until the signal disappears in the noise with no sign of a kink, or reentrant behavior, which is usually associated with gross inhomogeneities and imperfections in the films [16].

We now turn to the main topic of this work, which is the B dependence of our samples. In Fig. 2(a), we show ρ vs B isotherms, taken at several T s between 0.25–1.13 K, for a typical superconducting film. In this figure, we focus on the lower B range of our data, which includes the critical point of the B -driven SIT. The transition point is identified with the crossing point of the different isotherms at $B_c = 0.69$ T. For $B < B_c$, the superconducting phase prevails with ρ increasing with T , while for $B > B_c$ an insulating behavior takes over and ρ is now a strongly decreasing function of T . For this sample, the value of ρ at the transition is 5.85 k Ω . Our data in this B range are in accordance with previous observations of the B -driven SIT [6,7,9,10,17,18].

The picture changes dramatically when we consider a broader range of B . In Fig. 2(b), we present magnetoresistance curves of sample Na1c, taken over our entire available B range, at several T s between 0.07–1 K. As expected, when B is increased beyond B_c ($= 0.45$ T for this film), the insulating behavior initially becomes more pronounced. The surprising feature in our data is that, even though ρ increases by more than 5 orders of magnitude from its value at B_c [the ordinate in Fig. 2(b) is plotted using a logarithmic scale] [19], beyond approximately 9 T this trend reverses, the insulator becomes increasingly weaker, and, eventually, ρ approaches a value

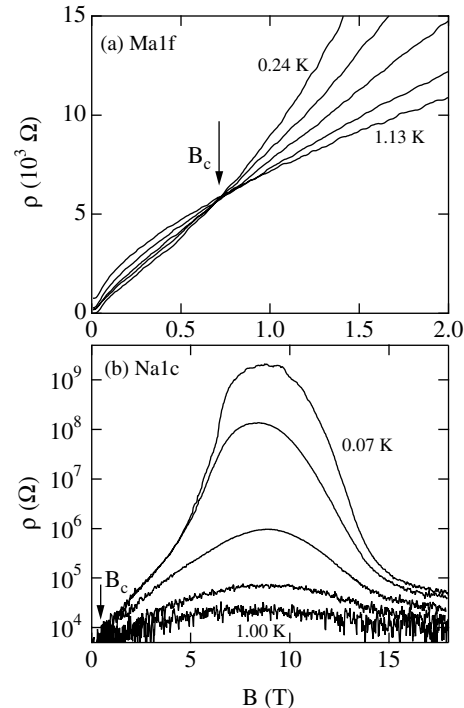


FIG. 2. ρ versus B isotherms (a) at a low B range for film Malf at $T = 0.24, 0.60, 0.78, 0.98,$ and 1.13 K (b) at a large B range for sample Na1c at $T = 0.07, 0.16, 0.35, 0.62,$ and 1.00 K. The critical point of the B -driven SIT, B_c , is indicated by the vertical arrow.

of 70 k Ω (at 0.07 K). This represents a drop of more than 4 orders of magnitude from the peak value of 2 G Ω . The demise of the insulating behavior seems to take place in two steps. An initial, rapid, drop beyond 10 T followed by a transition, at 14 T, to a slower decrease that continues all the way to, and beyond, our highest B of 18 T. It is unclear at which B value this trend will revert to the positive magnetoresistance expected at very high B .

Inspecting the details of the T dependence of ρ in this B -induced insulator reveals a similar picture. Generally, the B -induced insulator follows an Arrhenius T dependence similar to the high-disorder insulator of Fig. 1. In the inset of Fig. 3, we plot the ρ data in Fig. 2(b) vs $1/T$ at three different B values in the vicinity of the peak. The solid lines are fits to an Arrhenius form as in Eq. (1). These fits yield $T_I(B)$, the activation temperature, which now depends on B . In Fig. 3, we plot T_I versus B for the sample of Fig. 2(b). At B_c , the onset of the insulating behavior, T_I is zero. It then increases with B reaching a maximum at 9 T and, similar to ρ itself, decreases at higher B . A rough extrapolation of our data indicates that T_I will vanish at 20 T, although at $B \geq 14$ T our estimates of T_I suffer from increasingly large errors, making this extrapolation inaccurate.

The nonmonotonic behavior of the B -induced insulator has been observed before in a :InO films [17,20] and in other materials [21,22]. In these experiments, the value of

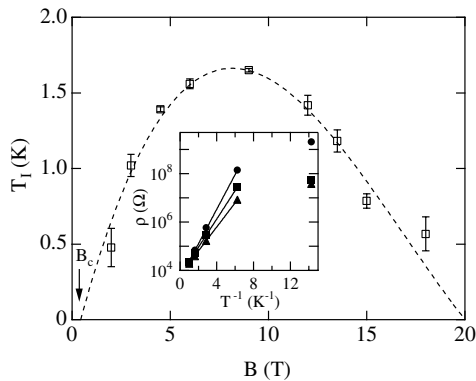


FIG. 3. Inset: ρ versus T^{-1} at $B = 6$ (squares), 9 (circles), and 12 T (triangles) for sample Na1c. The solid lines are fits to Eq. (1). The lowest T data points do not fit to the Arrhenius behavior. The main figure shows T_I , calculated from the fits to Eq. (1), as a function of B . T_I has a peak at 9 T. T_I estimates for 4 T $> B > 14$ T suffer from large errors since the low- T ρ value is not high enough to ensure activated behavior. The vertical arrow marks B_c ($= 0.45$ T), where $T_I = 0$. The dashed line is a guide to the eye.

ρ at the peak was significantly lower than in our data, probably because of the lower value of disorder in these films relative to ours. The appearance in our work, of a much higher ρ peak and more than 4 orders of magnitude in ρ fitting to an Arrhenius form, enabled us to firmly establish the presence of the insulating state and, more importantly, to obtain a conclusive measurement of the activation behavior in the vicinity of the peak. This leads to the central new result of our work: the value of T_I at the insulating peak, $T_I^p = 1.65$ K, is conspicuously close to the $B = 0$ value of T_c for this film, 1.27 K. To test the generality of this observation, we plot, in Fig. 4, T_c at $B = 0$ for several of our superconducting samples against their T_I^p s. Overall, T_I^p is close to T_c . In our samples, T_I^p had values between 0.7 and 1.9 K and T_c had values

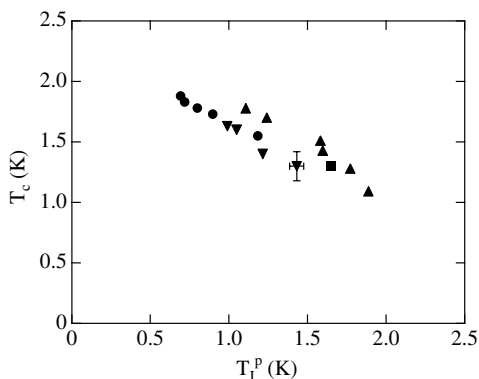


FIG. 4. T_c s at $B = 0$ for several superconducting samples are plotted against T_I^p s at the high- B insulating peaks. Each physical sample is marked with a different symbol and samples have been annealed to vary T_c . Error bars indicated are typical of most data points.

between 0.9 and 1.9 K. We recall here that, T_I measured in samples that are insulating at $B = 0$ (similar to Mala in Fig. 1) had values as high as 8 K. In the cases where we are able to follow one physical sample through several anneal stages, we see that, while T_c increases upon lowering of the disorder, T_I^p decreases, indicating that a more complete theory is required to account for the details of the T_c - T_I^p dependence.

We next suggest that it may be possible to account for the presence of an insulating peak with T_I^p close to T_c by adopting the point of view in which the properties of our films are not uniform over the entire sample [11,23,24]. We recall the recent theoretical work of Ghosal *et al.* [25], who studied numerically the behavior of 2D superconductors in the presence of a random potential as a function of disorder strength. They considered films that are structurally homogeneous, but did not make any assumptions about the spatial uniformity of the superconducting gap (Δ_S). They showed that, in the high-disorder limit, Δ_S can become spatially nonuniform despite the homogeneous nature of the films. These spatial variations of Δ_S lead to the formation of regions of large Δ_S , interceded with regions of vanishingly small Δ_S , creating an intermixture of superconducting and insulating phases [26]. The resulting situation is similar to that existing in granular films that are composed of macroscopic grains of superconductors embedded in an insulator [27]. In such granular films, the application of an external B drives the SIT by breaking long-range phase coherence while individual grains remain superconducting as verified by tunneling measurements [28]. In the insulating regime obtained under these conditions, the conduction process is limited by tunneling of normal electrons between the grains. Since these electrons are created by thermally breaking Cooper pairs in the superconducting grains, this process results in an activated transport with a characteristic temperature of order T_c [12]. Our observation that T_I^p is close to T_c of the film at $B = 0$ may point to such a scenario. A further increase of B beyond the insulating peak leads to the destruction of superconductivity in the grains, eliminating the superconductivity-induced barrier for transport, and resulting in the rapid drop of the resistivity, observed in Fig. 2(b) beyond 10 T.

We summarize our findings in the form of an experimental phase diagram [3], shown in Fig. 5, plotted in the B -disorder plane. It is constructed based on our data from 11 different a :InO films at 33 different anneal stages. The normal-state conductivity of the films at $T = 4.2$ K ($\sigma_{4.2K}$) is used as a measure of disorder. Conductivity at higher T could have equally been used with no essential change to the phase diagram. Two kinds of data points are plotted. The crosses mark the B_c of each film and thus define the boundary between the superconducting and insulating phases. The dashed line is a best fit to the B_c data. Extrapolation of this fit to $B = 0$ gives a $\sigma_{4.2K}$ value of $3.8 \pm 0.1 e^2/h$ (e is the electronic charge and h is

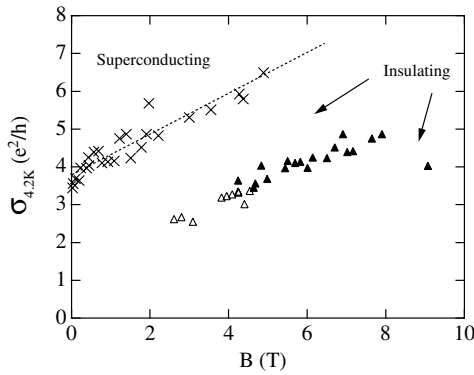


FIG. 5. Experimental phase diagram for a :InO superconducting thin films. The crosses mark the B_c of each sample at the SIT. The dashed line best fits the B_c data. Empty and filled triangles mark the positions of the B -induced insulating peak for samples that, at $B = 0$, are insulating and superconducting, respectively.

Planck's constant). Triangles are used to identify the B position of the insulating peak for samples that, at $B = 0$, were insulating (empty triangles) or superconducting (filled triangles). The triangles distinguish the region where the superconducting grains dominate the transport at low B from the region where superconductivity in the grains is progressively destroyed with increasing B . It is not clear that the triangles mark any phase boundary. Our experimental phase diagram is similar to the schematic phase diagram for a 2D superconductor suggested in [17].

In conclusion, we have studied superconducting thin films of a :InO that were driven through a SIT by the application of perpendicular B . Activated transport is observed over a wide range of B , above B_c , firmly establishing the presence of a well-defined insulating phase. The maximum value of the characteristic temperature for activated conduction at the insulating peak is close to the superconducting transition temperature at $B = 0$. These observations are discussed from a point of view in which the properties of our films are not uniform over the entire sample. We suggest that our results point to a possible relation between the conduction mechanisms in the superconducting and insulating phases in these disordered films.

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