Evidence of spatially inhomogeneous pairing on the insulating side of a disorder-tuned superconductor-insulator transition

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Measurements of transport properties of amorphous insulating In_xO_y thin films grown on $SrTiO_3$ substrates have been interpreted as evidence of the presence of superconducting islands on the insulating side of a disorder-tuned superconductor-insulator transition. Although the films appear not to be granular, their behavior is similar to that observed in granular films. The results support theoretical models in which the destruction of superconductivity by disorder produces spatially inhomogeneous pairing with a spectral gap.

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I. INTRODUCTION

Disordered films have been used to study the interplay between localization and superconductivity, a problem originally treated by Anderson[1] and Abrikosov and Gor'kov,[2] who considered a low-disorder regime. For strong disorder, approaches include fermionic mean-field theories,[3–5] and theories that focus on universal critical properties near the superconductor-insulator transition in which case the transition belongs to the dirty boson universality class.[6] The latter theory, and its many extensions, have been motivated by experiments in which the superconductor-insulator transition was traversed in films using a number of tuning parameters, including film thickness, perpendicular and parallel magnetic fields, and charge density.[7]

One of the surprises emerging from the study of the magnetic field tuned superconductor-insulator transition has been the observation of large peaks in the magnetoresistance at fields above the transition. This effect was first reported by Hebard *et al.*[8] who suggested that the state induced when superconductivity was quenched was a Bose insulator, characterized by localized Cooper pairs. They proposed that the peak was a signature of a crossover to a Fermi insulating state of localized electrons. This resistance peak has been the subject of more recent studies involving In_xO_y films,[9–11] microcrystalline TiN films,[12] and high-temperature superconductors.[13]

The focus of recent attempts to explain this feature of the experiments has been on the role of disorder in producing inhomogeneity of superconducting order. The picture is that the insulator consists of a dropletlike electronic texture of superconducting islands immersed in a normal metal or insulating matrix. A similar texture has also been considered in the context of the metal-insulator transition of two-dimensional electron gases.[14] The notion that disorder could imply inhomogeneity of superconducting order on some length scale was first discussed by Kowal and Ovadyahu[15] many years ago.

When quantum fluctuations are included in fermionic theories of superconducting films with sufficiently high enough levels of disorder, a spatially inhomogeneous pairing amplitude develops, which retains a nonvanishing spectral gap.[16] Inhomogeneous pairing can be induced in disordered superconductors by magnetic fields as has recently been established using a Hubbard model.[17] For sufficient disorder, inhomogeneous pairing can also be brought about by thermal fluctuations.[18] The most recent refinement of these ideas is the suggestion that the insulator can undergo a transition to a superinsulating state with infinite resistance, which is the electromagnetic dual to the superconducting state.[19] Using a model of a film which is a two-dimensional Josephson-junction array, the transition is pictured as a charge binding-unbinding transition separating an insulator with a large but finite thermally activated resistance from a low-temperature phase with essentially infinite resistance, which has been found in films of In_xO_y (Refs. 10 and 11) and TiN.[12]

Studies of disorder and magnetic field tuned superconductor-insulator transitions have usually been carried out on films that are either amorphous or granular. For the former, the disorder is on an atomic scale, and for the latter, on a mesoscopic scale in which case the films consist of metallic grains or clusters coupled by tunneling. Amorphous films can be produced when metal atoms such as Pb or Bi are deposited onto substrates held at liquid-helium temperatures (quench evaporation) that are precoated with a wetting layer of amorphous Ge or Sb,[20] by quench evaporation of Be,[21] or by careful growth of Mo_xGe_y, In_xO_y, or TiN using a variety of techniques.

Granular films, develop superconductivity in stages. If the grains are small and weakly connected, the film is an insulator. For grains larger than some characteristic size, and sufficiently coupled, "local superconductivity" develops below some temperature. The opening of a spectral gap in the density of states of the grains[22] results in a relatively sharp upturn in the resistance below this temperature, which is usually close to the transition temperature of the bulk material. For well enough coupled grains, there may be a small drop in resistance at that temperature, followed by an upturn. Global superconductivity or zero resistance occurs when there is a Josephson-coupled percolating path across the film.

We have measured the temperature and magnetic-field dependence of the resistance, and nonlinear conductance-voltage characteristics of amorphous In_xO_y films prepared by electron-beam evaporation on single-crystal $SrTiO_3$ substrates and subsequently annealed. The films when cooled to low temperatures start out on the insulating side of the disorder-driven insulator to superconducting transition. Although from structural characterization studies, they appear to not be granular, they nevertheless exhibit local superconductivity at the lowest temperatures. The application of a perpendicular magnetic field brings about a substantial increase in resistance with a maximum in $R(B)$ that is found at decreasingly small fields with decreasing temperature. This observation suggests the presence in the insulator of superconducting droplets or islands, characterized by a nonvanishing superconducting pair amplitude and coupled by tunneling. Many of the droplets are Josephson coupled, but the areal density of such droplets is not high enough to produce a percolating, zero-resistance path across the film leading to global superconductivity. The action of a magnetic field is to reduce the effective fraction of the film that is coherently coupled. The increase of the resistance in the regime of local superconductivity with decreasing temperature in zero magnetic field can be interpreted as evidence of the superconducting gaps of the droplets increasing with decreasing temperature with the dominant conduction channel being single-particle tunneling between droplets.

The conductance-voltage characteristics of the film are nonlinear, exhibiting thresholdlike features suggestive of single-particle tunneling between superconductors. On the other hand, these characteristics may be better explained as evidence of depinning by current of a Cooper pair charge structure. Earlier work on granular gallium ultrathin films in the vicinity of the thickness-tuned superconductor-insulator transition revealed a voltage threshold for enhanced conduction that was independent of temperature and magnetic field. This was interpreted as evidence for the depinning of a Cooper pair charge structure.[23] Similar voltage thresholds have been found in superconducting In_xO_y and TiN films in the high resistance regime induced by magnetic field.[10–12,19] These have been suggested as evidence for a new collective charge state induced by magnetic field.

Although the data suggest a similar field-induced collective charge state, there are several differences from the results of other investigations. First, the films we have studied start out as insulators exhibiting local superconductivity rather than as superconductors. Second, in the field-induced high resistance regime, a nonzero constant conductance is found below the voltage threshold that exhibits a temperature dependence consistent with Mott variable range hopping. The other experiments find Arrhenius-like temperature dependencies. The conductance-voltage characteristics in zero field at low temperatures are qualitatively similar to those in field, with only the subthreshold conductance changing with applied field. An element common to the present work and some other investigations is the emergence of metallic behavior in sufficiently high fields. The characteristic Mott hopping temperature is found to decrease with increasing field, and if extrapolation is a valid procedure would be predicted fall to zero in the limit of very high magnetic fields. Another difference is that in the present work, the values of sheet resistance in the field-induced insulating state appear to be much lower than those in the other investigations at similar temperatures. We speculate that some of these differences result from the substrates being $SrTiO_3$, a material with a high dielectric constant that can act to screen or partially screen Coulomb effects.

In the next section, we describe the preparation and characterization of the films. This is followed by Sec. III in which the data are presented. In Sec. IV, the implications of the data are discussed.

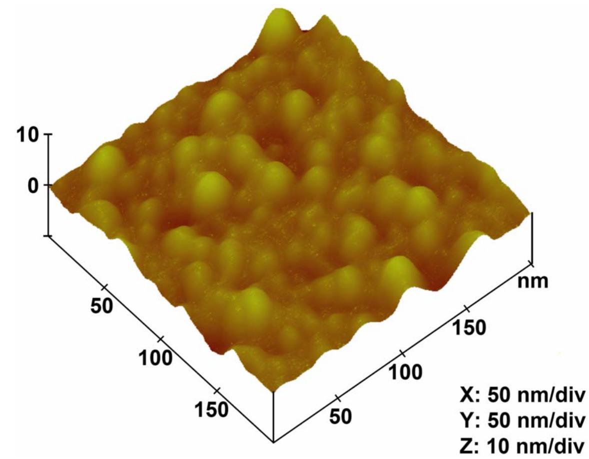

FIG. 1. (Color online) Atomic force microscope image of film 2 with a maximum vertical range of 8.5 nm and an average feature diameter of 18.0 nm. The thickness of this film was 22 nm.

II. FILMS

The 22 nm thick films used in this study were deposited at a rate of 0.4 nm/s by electron-beam evaporation onto (001) $SrTiO_3$ epipolished single-crystal substrates precoated with Pt electrodes, 10 nm in thickness. The starting material was 99.999% pure In_2O_3. A shadow mask defined a Hall bar geometry in which the effective area for four-terminal resistance measurements was $500 \times 500\ \mu m^2$. As-grown films exhibited sheet resistances of about 2600 Ω at room temperature and about 23 k Ω at 10 K. By annealing at relatively low temperatures (55–70 °C) in a high-vacuum environment (10^{-7} Torr), film resistances were lowered, and depending upon the annealing time were either insulating or superconducting at low temperatures.[24] Low-temperature rather than high-temperature annealing avoids changes in morphology. As reported by Gantmakher *et al.*[9] at room temperature the resistances of annealed films were found to be unstable. However, at low temperatures (40–1400 mK) and in vacuum, they were stable. Scanning electron microscope (SEM) studies did not reveal any In inclusions, and could be correlated with atomic force microscope (AFM) studies which revealed for a 22 nm thick film, roughness in the form of surface features with a height of 8.5 nm, and with bases about 18 nm in diameter. The implication of these characterization efforts is that the films are homogeneous and amorphous, do not contain isolated grains or In inclusions, but are rough. A representative AFM image of one of these films is shown in Fig. 1. A nearly featureless SEM photograph is shown in Fig. 2. The shadings of the image resemble the features shown in Fig. 1.

One cannot categorically assert that from scans such as those of Figs. 1 and 2 completely rule out the possibility that the films are not granular. There is always the possibility of structure on scales smaller than those resolved, and chemical inhomogeneities that lead to effective granularity that would not be detected using these methods.

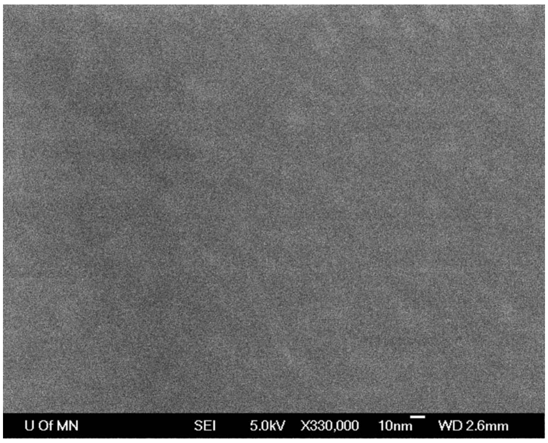

FIG. 2. Scanning electron microscope photograph of film 2. The size bar is 10 nm in length.

III. EXPERIMENT

Measurements were carried out in an Oxford Kelvinox-25 dilution refrigerator housed in a screen room, with electrical leads equipped with π section and RC filters. For measurements of resistance, the current was set in the range of 10–100 pA, to avoid the possibility of heating. Figure 3 shows a plot of $R(T)$ for two films which were studied in detail. For each, dR/dT is negative at the lowest temperatures. In the case of film 1, there is a local minimum in $R(T)$ at about 350 mK. Both films exhibit a sharp upturn in $R(T)$ between 200 and 300 mK, with the effects to be discussed below, occurring for film 1 at higher temperatures than for film 2. These behaviors are suggestive of local superconductivity.[22] At higher temperatures, $R(T)$ of both films could be fit rather well by a Mott variable range hopping form, $R = R_0 \exp[(T_0/T)^{1/4}]$. This is shown in the inset of Fig. 3 over a limited range of temperature. No single functional form could be used to fit the data in the regimes of upturn or local minimum of $R(T)$.

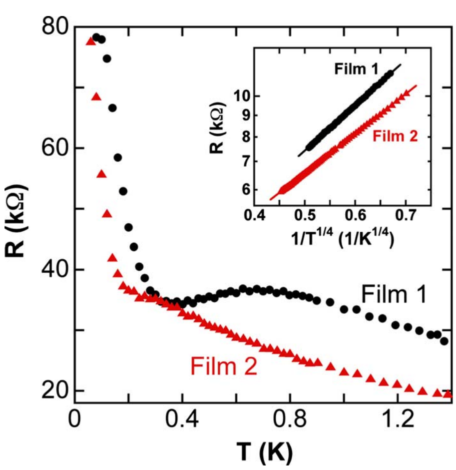

FIG. 3. (Color online) Resistance vs temperature for films 1 and 2. Inset: fits of Mott variable range hopping to higher temperature resistance data in zero field, at high temperatures for both films. The fitting parameters are $T_0 = 39.2$ K and $R_0 = 2119$ Ω ($T = 5$ to 15 K) and and $T_0 = 21.4$ K and $R_0 = 2252$ Ω ($T = 4$ to 24 K), respectively.

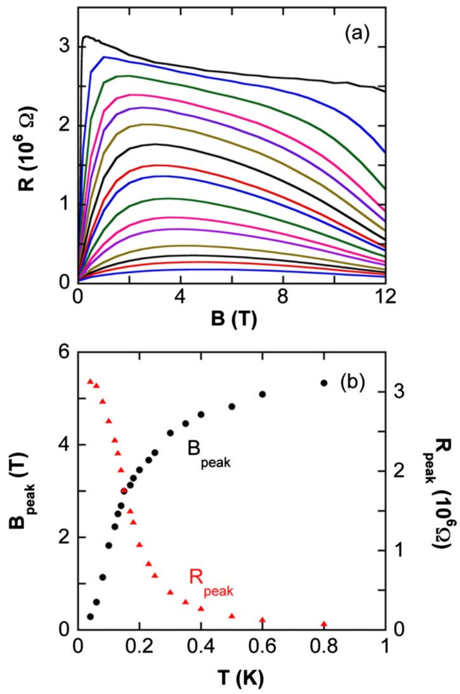

FIG. 4. (Color online) (a) Resistance vs magnetic field, $R(B)$, for film 1. The temperatures are 40 (top), 80, 100, 120, 130, 140, 150, 170, 180, 200, 230, 250, 300, 350, 400, and 500 mK (bottom). (b) The fields (left axis) and the resistances (right axis) of the peaks in $R(B)$ plotted as a function of temperature.

The sheet resistances of films 1 and 2 were both approximately 78 k Ω at 40 mK. In small perpendicular magnetic fields, their resistances increased by up to a factor of 40. The maximum in $R(B)$ as shown in Fig. 4(a) for film 1 is followed, at the lowest temperatures, by a relatively slow decrease in resistance with increasing field. The resistance maximum moved to lower fields, with decreasing temperature. The behavior of film 2 resembled the higher temperature data for film 1, presumably because film 2 exhibited weaker traces of superconductivity as evidenced by the absence of a local minimum in $R(T)$ in the zero field. This variation in properties from film to film is expected, as small changes in chemistry and/or morphology can have a large effect on disordered film properties. The temperature dependencies of the fields, B_{peak} and resistances R_{peak} for film 1 are presented in Fig. 4(b). A qualitatively similar, but weaker enhancement of resistance was previously reported for insulating In_xO_y films by Gantmakher *et al.*[9] A larger enhancement was reported for ultrathin insulating Be thin films[21] and for insulating TiN films.[12] However, none of these works report the systematic effects shown in Fig. 2(b). Another difference, as mentioned above is that most other works find an Arrhenius dependence of $R(T)$ in the presence of magnetic field. We find in the low-voltage linear regime behavior consistent with three dimensional Mott variable range hopping (VRH) in fields from 2 T up to 12 T at temperatures ranging from 0.17 K up to 1.23 K. Representative data are shown in Fig. 5.

Although these curves are best described by 3D Mott VRH, they exhibit much larger values of T_0 than found at

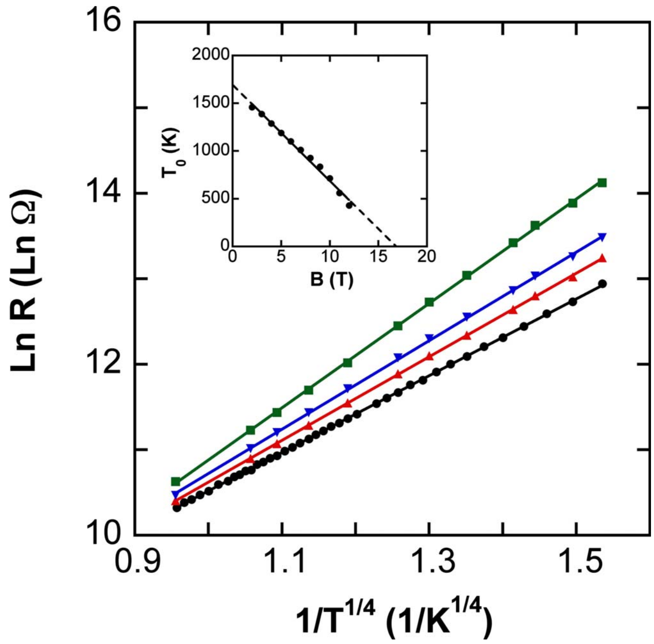

FIG. 5. (Color online) Fits with the three-dimensional Mott variable range hopping form in various magnetic fields for film 1. The fields are $B=2$ (top), 10, 11, and 12 T (bottom). Inset: T_0 vs B. The line is added as a guide to the eye, which intersects the B axis at 16.8 K and the T_0 axis at 1693 K.

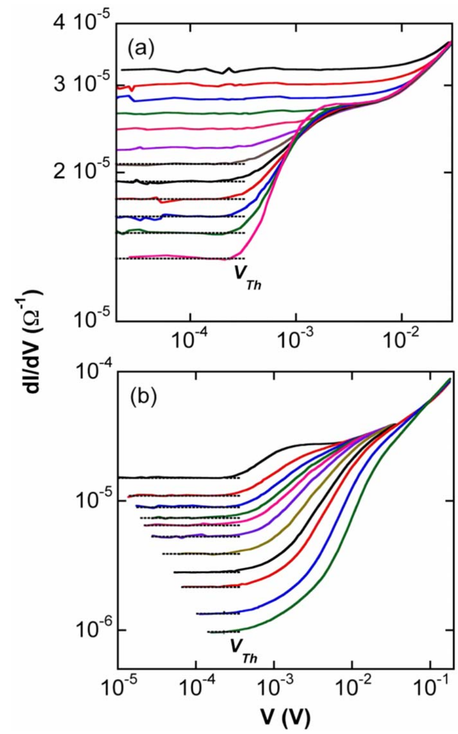

FIG. 6. (Color online) (a) Differential conductance dI/dV vs voltage V, for film 2 in zero field at 90 (bottom), 100, 110, 120, 130, 140, 150, 180, 220, 300, 400, and 500 mK (top). (b) dI/dV vs V at 100 mK in various perpendicular magnetic fields; 0 (top), 0.01, 0.02, 0.03, 0.04, 0.06, 0.1, 0.175, 0.25, 0.5, and 1 T (bottom).

higher temperatures in zero magnetic field (see the inset to Fig. 3). A linear extrapolation to higher fields than those accessed indicates that T_0 would fall to zero at a field of approximately 17 T suggesting that high fields could produce a metallic state such as that proposed for amorphous Be,[25] and recently reported for TiN films.[12] The Mott VRH fit fails at the lowest temperatures. In this regime it was not possible to identify any simple form that would describe the data. It should be emphasized that in no instance could the resistance be described by Arrhenius-type conduction $R(T) \propto \exp[T/T_0]$ as reported by other groups.

Differential conductance-voltage characteristics were also studied in the high resistance regime.[12,21,23,26–29] These are shown in Fig. 6 for film 2, which was studied in detail. Film 1 exhibited qualitatively similar features.

The curves in Fig. 6 exhibit threshold voltages for enhanced conduction, which are approximately independent of temperature and magnetic field. Below these threshold voltages, the differential conductance is constant. However, its value varied with temperature and magnetic field. It exhibited temperature dependencies as indicated above that could be characterized by Mott VRH at temperatures above 170 mK and perpendicular magnetic fields above 2 T. The nonlinear effects that may be due to electron heating are found at voltages well above the observed conductance thresholds.[29] The fact that the low-voltage nonlinearities vanish at temperatures above approximately 200 mK, suggests that these features of the conductance are associated with the presence of a nonvanishing pairing amplitude. Although the low-voltage behavior of the conductance-voltage characteristics are reminiscent of the single-particle tunneling characteristics of superconductor-insulator-superconductor (SIS) junctions, an alternative explanation as evidence of the depinning of a charge structure is more compelling. The nonlinearities of the I-V characteristics just above the low-voltage threshold can be described by the following equation:

$$I = G_{\mathrm{Linear}} V + A(V - V_{\mathrm{Threshold}})^{\zeta}. \quad (1)$$

Here, G_{Linear} is the subthreshold conductance, $V_{\mathrm{Threshold}}$ is the threshold voltage, and A is a proportionality constant. This nonlinearity produced a nonuniversal exponent ζ that was a monotonically decreasing function of increasing temperature in zero field. Typical values for film 2, for which these effects were studied in detail, were $\zeta \approx 2.9$ at 80 mK and $\zeta \approx 2.1$ at 140 mK. As a function of magnetic field, ζ exhibited a minimum, which coincided with the resistance peak. Typical values were $\zeta \approx 2.0$ at 1.5 T and $\zeta \approx 2.2$ at 12 T when $\zeta \approx 2.4$ in zero magnetic field. These voltage thresholds are qualitatively similar to those that are found when charge-density waves or disordered charge structures are depinned.[30] Thresholds were only found at low temperatures and vanished at approximately 200 mK, which as mentioned above, implies that they are associated with the presence of Cooper pairs.

IV. DISCUSSION

The fact that the magnetic field needed to induce the high resistance regime decreases with decreasing temperature is a counter-intuitive result. One possible inference is that there exists an increasing magnetic length-scale, possibly of the form $[\Phi_0/H]$ where Φ_0 is the flux quantum. If this length were to diverge at a nonzero temperature, it would imply the existence of a continuous phase transition to an insulator. If the divergence were to occur in the zero-temperature limit, the putative transition would be a quantum phase transition.

Although there are significant differences, the present results are reminiscent of the findings of large magnetoresistances at fields above the magnetic field-induced superconductor-insulator transition of superconducting In_xO_y and TiN films. The differences are the following: first, the effect emerges from a locally superconducting, rather than a superconducting state, and second, the shape of the curves is not that of a well-defined peak, symmetric about a peak field with a maximum location in field that is independent of temperature. The shapes of the curves in the present work imply that the high resistance state is more robust with respect to applied fields that the high resistance regimes found in other investigations. Nevertheless at high enough fields, the resistance falls. A third difference is that the values of resistance in the high resistance regime are not nearly as high, over roughly the same temperature range, as those found in these other investigations. A fourth difference is that the linear regime of the conductance-voltage characteristics exhibits a temperature dependence of the resistance can be described by Mott variable range hopping, rather than an Arrhenius form, over a fairly extended range of temperatures and magnetic fields. Finally, there are no qualitative differences between the conductance-voltage characteristics in zero field at low temperatures and those in field for which the subthreshold resistance has become very large.

The physical pictures proposed to explain the magnetoresistance peaks in In_xO_y and TiN films all rely on the development of inhomogeneity in the pairing amplitude, produced by the application of magnetic field. The theoretical picture recently proposed involving the insulating state having its origin in superconducting pairing is very likely relevant.[19] The explanation of the present results may involve most of the features of this picture, but would appear to differ in the magnitude of the conductivity (it is higher) and the nature of the excitations. An important feature may be that the present films are grown on $SrTiO_3$ substrates, which have enormous dielectric constants below 10 K ($\kappa > 20000$ for $T < 10$ K) because of their quantum paraelectric character. The juxtaposition adjacent to the film of a half space with a high dielectric constant would serve to screen the Coulomb interaction, which is responsible for the extraordinarily low conductances observed in other experiments, leading to higher conductivities and excitations at nonzero temperatures, which obey Mott variable range hopping.

Although there is no detail physical model available that explains the data, the observation of local superconductivity in zero magnetic field, by analogy with data from granular superconducting films, suggests a starting point that includes the presence of superconducting droplets in an insulating matrix. As mentioned previously, clusters of the type envisioned can emerge from models of superconductivity in the presence of disorder on a microscopic scale. The AFM and SEM scans of the films imply, but do not prove, that the films are not granular at some length scale smaller than that resolved. They also do not exclude the possibility of some chemical inhomogeneity affecting the superconducting properties that would not be resolved at all. If the cluster are real and the films were granular this would not *a priori* preclude the applicability of the model of Ref. 19, contingent upon the detailed geometry.

One might ask how the present films differ from those studied by Kowal and Ovadyahu.[15] Their films are presumably more disordered than ours, and thus further into the insulating regime. Their magnetoresistances are always negative, as the main effect of magnetic field would then be to weaken the inhomogeneous pairing amplitude, leading to negative magnetoresistance.

A final point relates to the nature of the transport in the limit of very high magnetic fields. The activation energy in the Mott variable range hopping that is observed, decreases with increasing magnetic field, and extrapolates to zero at about 17 T, well above the range of fields used in the present studies. This would imply that the films are metallic in the high-field limit, a conclusion previously reached for Be and TiN films and mentioned above.[12,25]

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