## Magnetic-Field-Tuned Quantum Phase Transition in the Insulating Regime of Ultrathin Amorphous Bi Films

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A surprisingly strong variation of resistance with a perpendicular magnetic field, and a peak in the resistance versus field, R(B) has been found in insulating films of a sequence of homogeneous, quenchcondensed films of amorphous Bi undergoing a thickness-tuned superconductor-insulator transition. Isotherms of magnetoresistance, rather than resistance, versus field were found to cross at a well-defined magnetic field higher than the field corresponding to the peak in R(B). For all values of B, R(T) was found to obey an Arrhenius form. At the crossover magnetic field the prefactor became equal to the quantum resistance of electron pairs  $h/4e^2$ , and the activation energy returned to its zero-field value. These observations suggest that the crossover is the signature of a quantum phase transition between two distinct insulating ground states, tuned by magnetic field.

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Superconductor-insulator (SI) transitions of disordered two-dimensional (2D) conductors have been studied extensively for about two decades because they offer the opportunity to investigate a wide variety of quantum phenomena [1]. Of particular interest are transitions of strongly disordered films, tuned by a perpendicular magnetic field. The dirty-boson picture was proposed to describe the magnetic-field-tuned transition from superconductivity. In this picture the insulator consists of Bose-condensed, field-induced vortices and localized Cooper pairs [2]. An early experiment by Paalanen, Hebard, and Ruel reported a peak in the magnetoresistance of  $InO_{x}$  films on the insulating side of the SI transition [3]. The behavior of the Hall resistance at fields close to the peak field led these authors to suggest that there was a crossover from the state proposed by Fisher in which there are localized Cooper pairs, to one in which transport is dominated by single-particle excitations. They referred to this as crossover between Bose and Fermi insulators [3]. This peak in R(B) in the insulating regime of the fieldtuned SI transition has been the subject of numerous investigations in recent years. With improvements in sample fabrication procedures and the introduction of new materials, changes of resistance of several orders of magnitude have been reported [4–8]. Also of interest in the present context are observations of Arrhenius activated behavior, i.e., a hard gap in  $InO_x$  and  $TiN_x$  films both in zero field in insulating films, and in magnetic fields on the insulating side of the SI transition [7,9,10]. In this Letter we report an apparent perpendicular magnetic-field-tuned quantum phase transition between two separate insulating ground states in thin films on the insulating side of the disorder or thickness-tuned superconductor-insulator transition. A central piece of evidence for this assertion is that isotherms of magnetoresistance (MR) defined as [R(B,T) - R(0,T)]/R(0,T) cross at a well-defined

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magnetic field higher than that corresponding to the peak in R(B).

The data employed in the present work were obtained from studies of homogeneous amorphous Bi (a-Bi) films that were grown by quench-condensation in situ at liquid helium temperatures on (100) SrTiO<sub>3</sub> (STO) single-crystal substrates precoated in situ with a 15 Å underlayer of amorphous Sb (a-Sb). Films grown by deposition onto substrates held at liquid helium temperatures and precoated in situ with thin underlayers of either a-Ge or a-Sb are known to be homogeneous [11]. The underlayers have zero conductance within instrumental resolution. The experiments involve repeated cycles of deposition and measurement carried out in a dilution refrigerator system designed to study the evolution of electronic properties with film thickness [12]. All the measurements were carried out using a four-terminal configuration employing a dc current source with currents in the linear regime of the currentvoltage (I-V) characteristic.

Representative examples of the evolution of R(T) with thickness of several insulating a-Bi films are shown in Fig. 1(a). Representative data of R(B) and the field dependence of the MR at 600 mK in films ranging in thickness from 19.74 to 21.12 Å are presented in Fig. 1(b). Peaks in R(B) are observed in films thicker than 20.53 Å. The values of the magnitudes of the peaks in R(B) and the fields at the peaks both increase with film thickness. It is important to note that large peaks in R(B), at fields above the critical field of the SI transition have not been previously reported for superconducting films grown on substrates with a-Ge or a-Sb underlayers. On the other hand, in the case of nominally granular quench-condensed films, grown on substrates that are not precoated, R(B) increases dramatically with increasing field, rising to values several orders of magnitude higher than the normal resistance [13]. Such films also exhibit nonmonotonic variations of R(T)



FIG. 1 (color online). (a) Zero-field resistance vs temperature of a sequence of nominally homogeneous *a*-Bi films with thicknesses from 19.74 Å (top) to 21.12 Å (bottom) in average nominal increments of 0.2 Å. Notice that the resistances of these films monotonically increase with decreasing temperature and do not exhibit the local minima found in nominal granular films. (b) MR as a function of field at 600 mK in films of different thicknesses. The labels are thicknesses in units of angstroms. The inset is the original sheet resistance vs magnetic field at 600 mK, again for films of different thicknesses.

which are not found in precoated films. Giant magnetoresistance peaks have been found in studies of films quench condensed onto substrates perforated with nanometer scale arrays of holes [14].

We now turn to the temperature dependence of R(B) and the MR for films of specific thicknesses. Representative data of R(B, T) for films, 20.91 and 21.12 Å thick, are presented in Figs. 2(a) and 2(b). The peak height becomes higher with decreasing temperature, which is consistent with results reported for  $InO_x$  and  $TiN_x$  films. The peak field  $B_{peak}$  is a function of temperature and can be fit with the form  $B_{peak} = B_0 + \alpha T^{\beta}$  over the range of temperatures studied, as shown in Fig. 2(c). From the measurements,



FIG. 2 (color online). Sheet resistance vs perpendicular magnetic field at different temperatures for the (a) 20.91 Å and (b) 21.12 Å thick films. The magnetic field at the MR peak vs temperature is plotted in (c). The two thicknesses are labeled with numbers whose units are angstroms. The arrows in (a) and (b) indicate the resistance peaks.

 $B_0 = 1.92 \pm 0.04$ ,  $\alpha = 2.63 \pm 0.06$ , and  $\beta = 2.78 \pm 0.10$  for the 20.91 Å thick film, and  $B_0 = 2.37 \pm 0.07$ ,  $\alpha = 3.791 \pm 0.09$ , and  $\beta = 2.41 \pm 0.13$  for the 21.12 Å thick film. It is unclear as to whether any of the theoretical models for the peak, which will be considered later, are consistent with these observations.

The temperature dependencies of the resistances of the 20.91 and 21.12 Å thick films at temperatures below 1 K can be fit by an Arrhenius form,  $R = R_0 \exp(T_0/T)$ , in fields ranging from 0 to 10 T. This is shown in Figs. 3(a) and 3(b). The field dependencies of the activation energy  $T_0(B)$ , and the prefactor  $R_0(B)$ , are plotted in the lower halves of Figs. 3(c) and 3(d). The activation energy exhibits a peak at a magnetic field close to  $B_0$  described in the previous paragraph.

The measurement of resistance at temperatures below 300 mK is difficult for several reasons. The *I*-*V* characteristics become nonlinear at currents larger than 1 pA. The resistance itself becomes so large that combined with the capacitances in the measuring circuit, with its heavy filtering, results in an extraordinarily long time constant. Also, R(T) can exceed the input impedance of the voltage amplifier, which can lead to erroneous results. Therefore, data below 300 mK were questionable and were excluded.

The most striking result is the occurrence of a crossover in the plot of the MR vs *B*, as shown in Figs. 3(c) and 3(d). The magnetic field at the crossing point  $B_c$  corresponds to two features of the Arrhenius fit. First, the activation energy at this crossing field returns to the value it exhibited at zero field. Therefore,  $T_0(B) - T_0(0)$  is always positive when  $B < B_c$  and negative when  $B > B_c$ . Second, the prefactors  $R_0$  in these two films are equal in value to  $h/4e^2$ , which is the quantum resistance for electron pairs. Parenthetically the first appearance of positive magnetoresistance at 600 mK for the film thicker than 20.53 Å also coincides with the zero-field prefactor falling below  $h/4e^2$ . These three features lead us to suggest the existence of a quantum critical point at  $B = B_c$  with the MR rather than R as the observable. Indeed, if Arrhenius conduction were to extend to zero temperature, in the zero-temperature limit, we would expect

$$MR(B, T)|_{T \to 0} = \left[\frac{R_0(B)}{R_0(0)} \exp\left(\frac{T_0(B) - T_0(0)}{T}\right) - 1\right]_{T \to 0}$$
$$= \begin{cases} \infty, & B < B_c \\ \frac{h/4e^2}{R_0(0)} - 1, & B = B_c \\ -1, & B > B_c \end{cases}$$
(1)

even though all resistances would diverge.



FIG. 3 (color online). Arrhenius plots of the (a) 20.91 Å and (b) 21.12 Å thick films in six representative magnetic fields. The resistances increase by more than three decades in these two films within the temperature range from 1 to 0.28 K. The MR, the activation energy  $T_0$ , and the prefactor  $R_0$  vs magnetic field of the 20.91 and 21.12 Å thick films are plotted in (c) and (d). The temperatures in (c) are 300 mK, 400 mK, 450 mK, 500 mK, 700 mK, 800 mK, 900 mK, and 1 K. The temperatures in (d) are 300 to 500 mK with 25 mK as the common increment and 500 to 900 mK with 100 mK as the increment.

Further support for the idea of a quantum phase transition comes from the success of finite size scaling. Here we use the scaling form first introduced by Fisher [2]:

$$R = R_c \mathcal{F}\left(\frac{|B - B_c|}{T^{1/\nu_z}}\right).$$
 (2)

However, we use MR as the observable in place of the resistance. Both films' data, within a certain range of fields and at sufficiently low temperatures can be scaled with critical exponent product  $\nu z = 0.65 \pm 0.08$ . This is shown in Fig. 4. With the assumption z = 1, this product would correspond to the universality class of a 2 + 1 dimensional *XY* model. Similar values have been found for magnetic field and electrostatically tuned SI transitions [15,16]. The data points close to the peak in *R*(*B*) and at high temperatures fail to scale, which may be due to the limits on the quantum critical regime.

It is interesting that the MR rather than the *R* isotherms as a function of *B* cross as a function of magnetic field. The low temperature zero-field resistance must result from a combination of effects including the motion of strongly localized electrons as well as participation of presumably localized Cooper pairs. The application of a magnetic field to the film adds vortices and the behavior of these added vortices results in a highly resistive phase that appears to disappear at a field-tuned quantum phase transition. That this high resistance phase and the observed crossover are associated with Cooper pairing is supported by the robust observation that at the crossover magnetic field the prefactor of the Arrhenius fit to the data is the quantum resistance for electron pairs.

Additional evidence for the presence of vortices in the film near the magnetoresistance peak is the anisotropy of magnetoresistance. At 400 mK for the 21.12 Å thick film, our preliminary results of the MR in a 2.5 T parallel field is 20.95% [17], while it is 198.7% in a 2.5 T perpendicular field. This result is consistent with previous observations by Markovic *et al.* [18], which were also interpreted as the



FIG. 4. Scaling of the MR of (a) the 20.91 Å and (b) the 21.12 Å thick films. Both plots employ data from below and including 500 mK. The magnetic field range of 20.91 Å film is from 2.5 to 10 Tesla, while it is 5 to 10 Tesla for the 21.12 Å film. In the case of the 20.91 Å thick film, there is a shorter upper branch due to the closeness of  $B_c = 3.9T$  and the peak in R(B), while  $B_c = 7.3T$  in the case of the 21.12 Å thick film.

evidence of vortices in the insulating Bi films at low magnetic fields. With the ability to apply higher fields, we found the anisotropy diminishes when the field is larger than the peak field and vanishes near  $B_c$ . For instance, at 400 mk the MRs in parallel and perpendicular fields of 7.3 T are 56.2% and 60.6%, respectively. This result is consistent with the idea that local superconductivity and vortices disappear close to the field-tuned quantum phase transition.

To the best of our knowledge none of the models of the SI transition predict a quantum phase transition such as the one reported here, although it is quite possible that they may be extended to include one [2,19–25]. The condition of  $R_0$  equal to  $h/4e^2$  delineates a phase boundary in these thickness and field-tuned insulating films as evidenced by two observations: the magnetoresistance peak is found only in the thicker films when the zero-field prefactor falls below  $h/4e^2$  and the prefactor at the crossover field  $B_c$  is  $h/4e^2$ . This suggests that quantum fluctuations of vortices play a role in the present observations, that  $B_c$  is the critical field for the vanishing of local superconductivity, and that the transition is from a Bose insulator with localized Cooper pairs to a Fermi insulator.

One might ask why these effects have not been observed previously, given the significant number of studies of the field-tuned superconductor-insulator transition. Most studies have focused on films that are superconducting in the absence of a magnetic field. Thus there is no zero-field reference resistance as would be needed to evaluate the magnetoresistance. Second, the crossover field is at 3.9 and 7.3 T for the two films reported here, with the 7.3 T crossover a property of the less disordered film. With further reduction of disorder with an increase of thickness, the crossover could move to unattainably high values of magnetic field and be unobservable.

In summary, isotherms of the MR have been observed to cross at a well-defined magnetic field higher than that of the peak in R(B) of quench-condensed insulating films of *a*-Bi. Curves of R(T) at all magnetic fields follow an Arrhenius form for temperatures below 1 K. The prefactor of this form becomes equal to the quantum resistance for pairs and the activation energy returns to its zero-field value at the crossover field. Data near the crossover are consistent with finite size scaling and the universality class of the (2 + 1)D XY Model. We suggest that these observations are evidence of a quantum phase transition between two distinct insulating phases, which might be a Bose insulator to a Fermi insulator.

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