Evidence of Vortices on the Insulating Side of the Superconductor-Insulator Transition

N. Marković, A. M. Mack, G. Martinez-Arizala, C. Christiansen, and A. M. Goldman
School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455
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The magnetoresistance of ultrathin insulating films of Bi has been studied with magnetic fields applied parallel and perpendicular to the plane of the sample. Deep in the strongly localized regime, the magnetoresistance is negative and independent of field orientation. As film thicknesses increase, the magnetoresistance becomes positive, and a difference between values measured in perpendicular and parallel fields appears, which is a linear function of the magnetic field and is positive. This is not consistent with the quantum interference picture. We suggest that it is due to vortices present on the insulating side of the superconductor-insulator transition. [S0031-9007(98)06683-6]

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In the limit of zero temperature, ultrathin films of metals become either insulating or superconducting depending on the strength of the disorder [1] or the applied magnetic field [2]. The superconductor-insulator (SI) transition has been described by a boson Hubbard [3] model which also predicts a metallic phase for films on the margin between insulating and superconducting behavior whose resistance is "universal." Within this model, at $T = 0$, the superconducting state is a Cooper pair condensate with localized vortices, and the insulating state is a vortex condensate with localized Cooper pairs. There has been no full generalization to nonzero temperature. The boson Hubbard model has been challenged by the results of tunneling experiments [4] carried out on thin films of metals which have been interpreted as showing that the amplitude of the order parameter is extremely small or zero at the SI transition and in the insulating state. On the other hand, Hall and longitudinal resistance studies on indium oxide films [5], driven out of the superconducting state by magnetic fields, were interpreted as evidence of two different insulating phases, one presumably a Bose insulator of localized Cooper pairs and the other the usual Fermi insulator of localized electrons. If this interpretation were correct, films insulating by virtue of disorder might behave similarly. Furthermore, if there were vortices and Cooper pairs on the insulating side of the transition, then one might expect to see some evidence of vortex motion in the magnetoresistance of such films.

The magnetoresistance (MR) of disordered two dimensional electronic systems [6] in both strongly localized (SL) and weakly localized (WL) regimes is found to be dominated by two basic mechanisms: an orbital effect [7,8], which is due to quantum interference between electron paths, and a spin effect [9,10], which arises from Zeeman splitting of the electron spin states. The former is flux driven, and therefore highly sensitive to whether the magnetic field is applied in the direction perpendicular to the plane of the sample or parallel to it, while the latter is isotropic and depends only on the magnetic field strength. In systems with strong spin-orbit (SO) coupling, both mechanisms predict a negative MR in the SL regime and a positive MR in the WL regime. The characteristic anisotropy due to the orbital effect, as well as the signature of the spin effect, has been observed in a number of experiments [11,12]. Here we report the results of MR measurements carried out on ultrathin insulating films of Bi close to the SI transition in magnetic fields both parallel and perpendicular to the plane of the sample. The observed anisotropic component of the MR is a linear function of the magnetic field, always positive, and it increases monotonically through the SL-WL crossover. This is not consistent with it being an orbital effect. We suggest that this contribution arises from the flow of vortices, which may offer new evidence in favor of the boson Hubbard model.

The ultrathin Bi films used in this study were evaporated on top of a 10 Å thick layer of amorphous Ge, which was predeposited onto a 0.75 mm thick single crystal of $SrTiO_3$ (100). The substrate temperature was kept below 20 K during all depositions and all the films were grown *in situ* under UHV conditions ($\sim 10^{-10}$ Torr). Under such circumstances, successive depositions could be carried out without contamination to increase the film thickness in increments of 0.25 Å up to 15 Å. Film thicknesses were determined using a previously calibrated quartz crystal monitor. Films prepared in this manner are believed to be homogeneous, since it has been found that they become connected at an average thickness on the order of one monolayer [13]. Resistance measurements were carried out using a standard dc four-probe technique with current bias up to 50 nA. Current-voltage characteristics were linear up to at least 1 μA. Magnetic fields up to 20 kG parallel to the plane of the sample, and up to 12 kG perpendicular to the plane of the sample were applied using a superconducting split-coil magnet.

The temperature dependence of the conductance is logarithmic at higher temperatures, as expected for a weakly localized 2D system, while at the lowest temperatures it becomes activated and could be fit by $G = G_0 \exp(-[T_0/T]^x)$ where $x = 1/2$. At intermediate temperatures, there is a crossover region between the logarithmic and exponential behavior, which moves towards lower temperatures in thicker films.

Figure 1 shows the MR as a function of magnetic field at 0.7 K for the four most insulating films in both parallel and perpendicular fields. The thinnest film, sample 1, shows a negative MR with a fractional change of up to 12% in a 20 kG field, which seems to be field-orientation independent. Although not shown on the graph, additional data taken at different temperatures confirm this isotropic behavior. Sample 2, which is 0.25 Å thicker, also shows a negative MR, but the fractional change is now less than 4% in a 20 kG field. For this sample, the parallel MR is greater than the perpendicular. This anisotropy becomes more obvious for sample 3, which shows a small positive MR for both directions of the field. The MR of sample 4 is even more positive, and the difference between the parallel and the perpendicular fields increases further.

The MR changes sign from negative to positive in both parallel and perpendicular magnetic fields at the temperatures and the thicknesses for which the system is at the crossover between the SL and the WL regime (crossover being loosely defined as the region where the behavior of the conductance changes from activated to diffusive). A similar sign change has been observed in thin films of Au, Ag, and Cu by Hsu *et al.* [14], where the MR was considered to be dominated by the orbital effect.

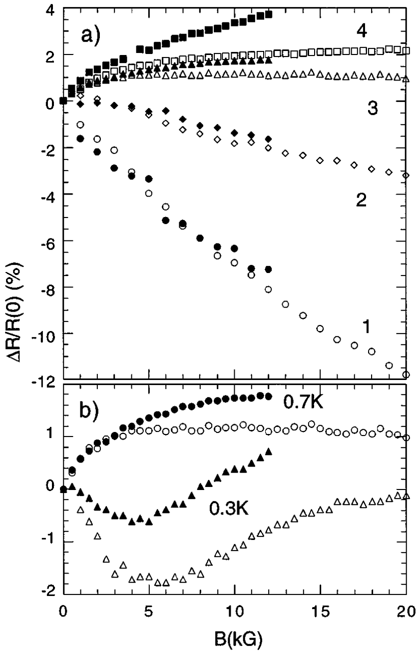

FIG. 1. Magnetoresistance as a function of magnetic field for (a) four films of different nominal thicknesses: 10.50 Å (circles), 10.75 Å (diamonds), 11.00 Å (triangles), and 11.25 Å (squares); (b) sample 3 ($d = 11.00$ Å) at two different temperatures: 0.7 K (circles), and 0.3 K (triangles). Filled symbols represent the perpendicular field, and open symbols represent the parallel field.

In our case, the MR is dominated by a spin effect. One can conclude that from the fact that plotting the longitudinal MR against $\mu_B B/k_B T$ makes all the data collapse on a single curve. This also implies that the orbital contribution is negligible in parallel field. In the variable range hopping regime, a mechanism proposed by Eto [9] predicts an isotropic, negative MR due to the Zeeman effect even in the presence of SO interactions. A model by Maekawa and Fukuyama [10], also based on the Zeeman effect, predicts a positive MR in the WL regime. Our results are in qualitative agreement with these pictures.

We define the anisotropic component of the MR as the difference between the transverse and the longitudinal magnetoresistances $R_a = (\Delta R_\perp - \Delta R_\parallel)/R(0)$ [15]. Figure 2 shows that R_a is a linear function of the magnetic field for ten different thicknesses. We will argue that this anisotropy cannot be due to any standard orbital effect. In the SL regime, in which conduction occurs through variable range hopping, the orbital MR can be described by a model originally proposed by Nguyen, Spivak, and Shklovskii (NSS) [7] in which an applied magnetic field increases the conductance. The critical percolation path approach [8] yields a fractional change in the resistance $\Delta R/R(0) \propto B^2$ at low fields, which increases with decreasing temperature. A similar behavior is predicted in the presence of SO scattering, although the magnitude of the MR is expected to decrease as the SO scattering increases [16,17].

In a 2D system, the sample thickness is smaller than the hopping length, which forces all hops to be in the plane of

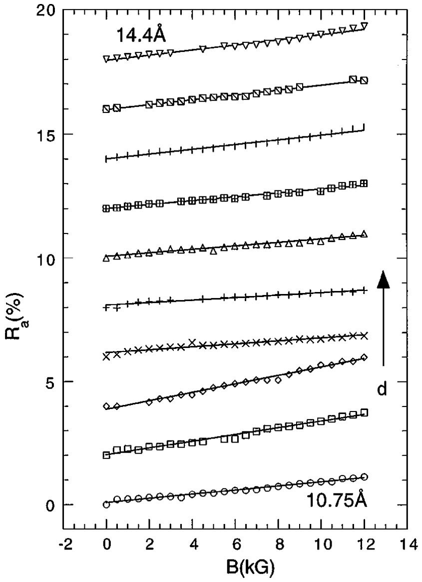

FIG. 2. The difference between the perpendicular and parallel magnetoresistance R_a as a function of magnetic field for ten films of different thicknesses at 0.7 K. Full lines are linear fits. The zeros of the vertical scale are offset for clarity.

the sample. This restriction on hop orientations leads to a MR anisotropy and a MR in a perpendicular field which should be much larger than in a parallel field. Even though the MR of sample 2 is slightly anisotropic, this anisotropy cannot be due to the orbital mechanism described above as the magnitude of the MR is smaller in perpendicular than in parallel fields. In addition, a predicted quadratic behavior with magnetic field was not observed.

In the WL regime, the orbital mechanism is different. In systems with strong SO coupling there is a positive MR, which becomes larger with decreasing temperature as the electron phase coherence time increases. Quantitatively, the theory predicts $\Delta R/R(0) \propto B^2$ in the low field limit and $\Delta R/R(0) \propto \ln B$ in the high field limit [6,18]. In a 2D system, a field parallel to the plane will make no contribution to the flux through the time reversed loops, and no parallel MR is expected. Samples 3 and 4 considered in Fig. 1 are in the WL regime where the conductance shows a logarithmic dependence on temperature. The MR is positive, and its magnitude is larger in perpendicular field, which might be expected from the model described above, but further analysis reveals some inconsistencies. Namely, it was not possible to obtain a satisfactory fit to the WL orbital effect theory [18], which predicts $R_a \propto B^2$ in the low field. Instead, R_a is clearly a linear function of the magnetic field for a wide range of thicknesses and temperatures. Furthermore, the anisotropy due to the orbital effect is expected to change sign [14] at the SL-WL crossover. The anisotropic response first occurs at the temperatures and thicknesses at which the films are in the SL regime, and it persists into the WL regime, but its magnitude is always positive and it changes monotonically through the crossover, as shown on Fig. 3. All this suggests that standard orbital effects cannot be the mechanism responsible for the anisotropy.

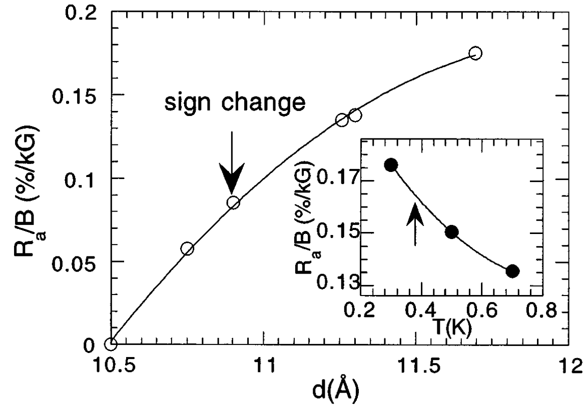

FIG. 3. The slope of R_a vs B, which can be thought of as a measure of the anisotropy, as a function of film thickness for six different films at $T = 0.7$ K. Inset: The slope of R_a vs B as a function of temperature for sample 3 ($d = 11.00$ Å). The lines are guides to the eye. The arrows show where the system is at the SL-WL crossover.

Orbital MR has been observed in a number of experiments on several other systems [11,12] and is often used to obtain various scattering times. One might therefore ask why we do not see any orbital effect? It is known that orbital MR is suppressed in the presence of strong SO coupling [16,17], which might explain why the contribution to the MR from the orbital effect is so small compared to indium-oxide films. It is possible that this contribution would become overwhelmed by the effects of superconducting ordering before becoming significant (this would not be the case for Au, Ag, and Cu films, which can get deep into the WL regime where the orbital effect is substantial, without becoming superconducting). Indeed, closer to the SI transition, R_a starts to deviate slightly from the linear field dependence (the very top of the Fig. 2), acquiring a small quadratic term. This might be due to the orbital effect, or possibly Maki-Thompson (MT) effect, which has the same field dependence as the WL orbital effect [19].

If the boson-Hubbard model actually described the insulating state near the SI transition, then the insulator might be able to sustain pointlike vortices because of the nonvanishing Cooper pair density. If these vortices were to move freely [20] in response to currents, they might produce a flux flow contribution to the MR of the system. This would result in $R_\perp - R_\parallel$ always being positive and proportional to the magnetic field [21], which is the observed behavior. Increasing the thickness (decreasing the disorder) and lowering the temperature drives the system deeper into this putative Bose insulator state, (the most insulating films do not exhibit this effect and are Fermi insulators) where Cooper pairs and vortices are more likely to form. The anisotropy due to flux flow in this regime would therefore become more pronounced with increasing thickness and decreasing temperature, as observed in the measurements. In other words, the linear response, except for its relatively small magnitude, very much resembles the MR due to flux flow in superconducting films, which our samples become when made just slightly thicker. If the process were a dynamical effect, i.e., the vortexlike MR response was occurring in the presence of some kind of order parameter amplitude and phase fluctuations, this picture might not be incompatible with the interpretation of tunneling spectra on the insulating side of the transition that the superconducting gap and the average order parameter amplitude are both zero [4]. Alternatively, and consistent with the original boson Hubbard picture, a nonvanishing superconducting order parameter amplitude could persist into the insulating regime, even though superconductivity is destroyed by phase fluctuations. Phase fluctuations can reduce the gap [22], as they are pair breaking [23]. This can lead to gapless superconductivity. It is also possible that the local gap is destroyed by disorder, but the fermionic degrees of freedom are highly suppressed and maybe even dynamically irrelevant [20]. In support of this view are indications of superconducting effects in films of In_2O_3 driven into the insulating state by magnetic fields, in the work of Paalanen *et al.* [5] and in insulating films of In_2O_3 in the work of Gantmakher and co-workers [24].

Further experimental work and a theoretical model suitable for the intermediate disorder at finite temperatures are needed to resolve this issue. In particular, we are attempting to detect the presence of vortices in the insulating regime by searching for vortex shot noise in a manner similar to the study of Knoedler and Voss [25] carried out on granular aluminum films in the superconducting state.

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