Anomalous parallel-field negative magnetoresistance in ultrathin films near the superconductor-insulator transition

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A parallel-field negative magnetoresistance has been found in quench-condensed ultrathin films of amorphous bismuth in the immediate vicinity of the thickness-tuned superconductor-insulator transition. The effect appears to be a signature of quantum fluctuations of the order parameter associated with the quantum critical point.

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I. INTRODUCTION

Quantum phase transitions (QPTs) are brought about by the variation of an external parameter of the Hamiltonian of a system, which changes the ground state.[1] The superconductor-insulator (SI) transition in two dimensions (2D), tuned by disorder or magnetic field, is believed to be a quantum phase transition. The understanding of the SI transition as a QPT has been inferred from the analysis of transport data using finite-size scaling. However, recent data on the field- and disorder-tuned SI transitions suggest the existence of a finite intermediate regime of metallic behavior not anticipated by the theory.[2] Subsequent explanations of this regime have included a metallic Bose glass or a Bose metal,[3] metallicity produced by the influence of dissipation,[2] and effects resulting from the influence of fermionic excitations not included in boson models.[4] In some instances the metallic regime may be attributable to the electrons not cooling. Because of these complications, it would be useful if there were an explicit signature of quantum fluctuations that could serve as an indicator of the SI transition. A recent calculation[5] appears to offer this possibility. Employing a perturbative approach, a negative correction to the parallel-field magnetoresistance (MR) attributable to quantum fluctuations has been found near the parallel-field SI transition of films (and wires). The total negative MR results from the "Aslamazov-Larkin" correction being overwhelmed by negative contributions from the "density of states" and "Maki-Thompson" terms. In this paper, we report an anomalous, *parallel*-field negative MR whose occurrence is correlated with the thickness-tuned SI transition of ultrathin homogeneous films. This effect may derive from corrections to the conductivity associated with quantum fluctuations even though the effect is found near the condition of critical disorder rather than critical parallel magnetic field.

II. EXPERIMENTAL

Resistance measurements were made using a bottom loading Kelvinox 400 dilution refrigerator, employing four-probe techniques. Electrical leads were filtered at room temperature using π-section filters with a cutoff frequency of about 10 Hz. Power dissipation in the measurement process was kept below 1 pW. The substrate was mounted on a sample holder that could be transferred between the mixing chamber of the refrigerator and an attached ultrahigh vacuum growth chamber using a liquid-helium-cooled transfer stick.[6] In these experiments the plane of the substrate, mounted on a rotatable sample holder, was restricted to be close to the nominally parallel orientation to accommodate additional heat sinking needed to facilitate cooling below 0.1 K.

Films were grown on substrates held at liquid-helium temperatures while mounted on the sample stick with the growth chamber at a pressure of 10^{-10} Torr. The substrates were epipolished single-crystal $SrTiO_3(100)$ wafers, precoated (*in situ*) with a 6-Å-thick film of amorphous Ge (*a*-Ge). To prevent annealing, substrate temperatures were held below 12 K during growth, and below 18 K during other processing and handling. Film thicknesses were increased in increments as small as 0.04 Å, as measured using a calibrated quartz-crystal monitor. The latter was calibrated *ex situ* using a profilometer. Films processed in this fashion are believed to be homogeneously disordered and not granular.[7] Critical features of the present experiments were the possibilities of changing the thickness of a film in tiny increments and of growing films homogeneous in thickness to one part in 10^4.[6] The phenomena reported here occurred over a nominal thickness range of order 0.8 Å out of approximately 9.0 Å, and would not have been seen without such stringent control.

III. RESULTS AND ANALYSIS

The evolution of $R(T)$ of 11 films with thicknesses ranging from 8.5 to 9.3 Å is shown in Fig. 1. Thinner and thicker films, grown in other runs (not shown), were insulating and glass-like in their responses, or fully superconducting, respectively. It should be noted that there are metallic regimes at low temperatures in this data, for both insulator- and superconductor-like films. From this set of experiments alone, one cannot demonstrate that these regimes are not a consequence of failure to cool the electrons. However, the existence of an intermediate metallic regime[2,3,8–10] separating superconducting and insulating films is not the issue in the present work. Apart from this possible metallic behavior at the lowest temperatures, the films sort into two categories, those behaving like insulators ($dR/dT<0$) and those behaving like superconductors ($dR/dT>0$).

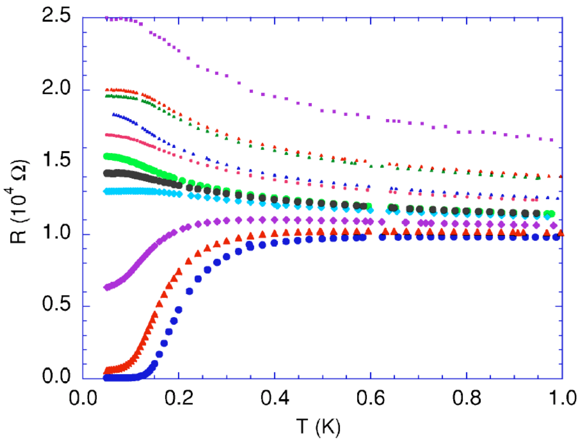

FIG. 1. (Color online) Evolution of $R(T)$ for a series of 11 different thicknesses of Bi. Film thicknesses are: 8.5 (top curve), 8.7, 8.8, 8.85, 8.91, 8.99, 9.05, 9.09, 9.19, 9.25, and 9.3 Å (bottom curve).

Films with thicknesses of less than 8.99 Å are in the insulating state and have positive $R(B)$ at all temperatures. The negative MR first appears in the 8.99-Å-thick film and was studied at temperatures between 0.05 and 0.3 K. An example of the temperature dependence of the MR for the 9.05-Å film, which is representative, is exhibited in Fig. 2. In the lowest fields, $dR/dB>0$. With increasing field, a maximum is reached. At all but the highest temperatures, a regime in which $dR/dB<0$ is then entered. With further increase in field, there is a minimum in $R(B)$, followed by a regime in which the resistance is a linear function of field. This linear behavior at high fields is found at all temperatures from 0.05 to 1 K and in fields from 2 to 12 T.

The magnetic fields in these measurements were only nominally parallel to the substrate plane. The misalignment was estimated to be at most the order of 1° to 2°. This would lead to a perpendicular field component of about 1/30th that of the applied field. At low applied fields, the resultant perpendicular component is insignificant. However, as the magnetic field is increased, this will eventually no longer be true. At high fields we find a linear dependence of $R(B)$, an expected effect if there were flux flow resistance due to a perpendicular field component.[11] With the above estimate of the misalignment this linear regime would appear to start at perpendicular field components of the order of 600 Oe.

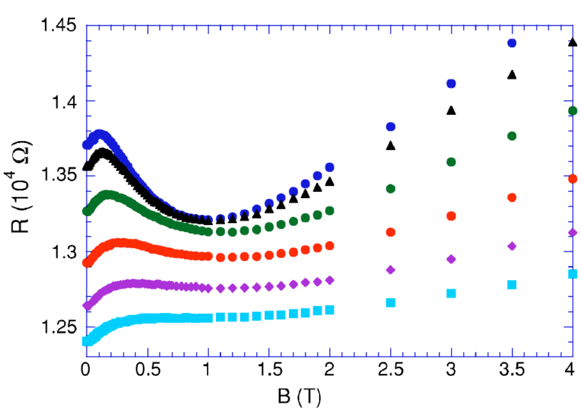

FIG. 2. (Color online) Resistance as a function of parallel magnetic field at 50 (top curve), 100, 150, 200, 250, and 300 mK (bottom curve) for the 9.05-Å-thick film. Data is shown at low field to emphasize the negative magnetoresistance that appears at low temperature. In fields higher than those shown, the $R(B)$ behavior is quite linear.

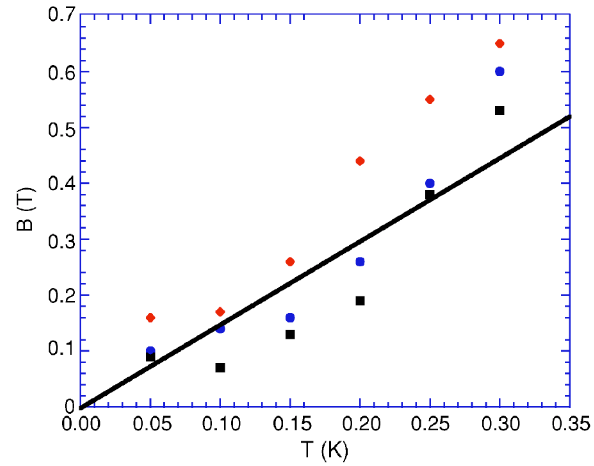

FIG. 3. (Color online) Magnetic field at the peak of $R(B)$ curves vs temperature for 8.99-, 9.05-, and 9.09-Å-thick films from top to bottom. The line corresponds to $\mu B/k_BT=1$.

IV. DISCUSSION

There are systematic aspects of the low-field data exhibiting positive MR, which lead us to attribute it to a spin polarization of the carriers transported by hopping in the *a*-Ge substrate. The effect is most pronounced in the thinnest films, where contributions to the conductivity from the substrate would be proportionally more important than in thicker ones. The peak disappears entirely for films whose thickness exceeds 9.09 Å. This would be expected when transport through the film became dominant. A theory of positive MR in the hopping regime has been given by Matveev and collaborators.[12] It is based on the idea that in zero magnetic field a significant number of hopping sites can be doubly occupied with the electrons forming a spin singlet. In a magnetic field strong enough to polarize the carriers, transitions involving such sites are forbidden as electron pairs cannot form singlets. This leads to a positive MR that saturates when the spins were fully polarized. This picture has been verified experimentally in semiconducting films.[13] In the films of the present work, the conductance channel exhibiting low-field positive MR competes with that exhibiting negative MR, which is a parallel channel. When the positive MR saturates, the negative MR dominates, resulting in a relatively sharp peak at the lowest temperatures for the thinnest films. The peak field should occur when the condition $\mu_BB\sim k_BT$ is satisfied. In Fig. 3 we plot the field at the MR peak versus T for films of three different thicknesses. The line on the figure corresponds to $\mu_BB=k_BT$.

It is necessary to understand the systematics of the negative MR effect in order to justify relating it to quantum fluc-

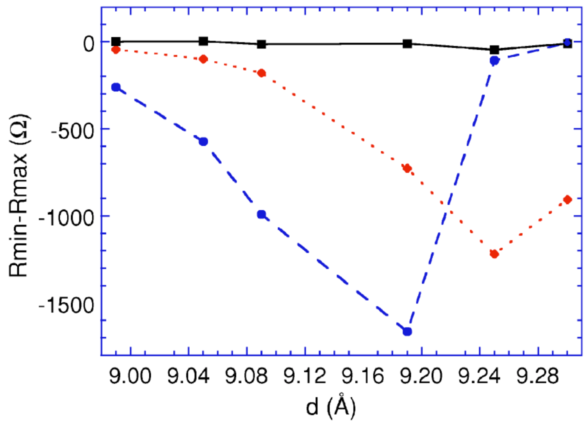

FIG. 4. (Color online) Difference between resistances of the trough and peak of $R(B)$ curves $(R_{\mathrm{min}}-R_{\mathrm{max}})$ as a function of thickness d at 0.05, 0.200, and 0.3 K from bottom to top.

tuations associated with a quantum critical point. In Fig. 4 we plot the fractional change in resistance from the peak to the trough of the negative magnetoresistance as a function of film thickness at 0.050, 0.200, and 0.3 K. Measurements at other temperatures have been suppressed for clarity. The negative MR is not found in any of the films at 0.3 K, and is strongest at the lowest temperature, 0.050 K, for the 9.19-Å-thick film. It is first found in the 8.99-Å-thick film. As the temperature increases the maximum effect moves towards films of greater thickness before eventually disappearing. If one correlates thickness with the sheet resistance of the films at the lowest temperature, the maximum effect occurs near or above a resistance of 6300 Ω which is very close to the quantum resistance for pairs. This is very close to what one would judge to be the SI transition from examination of Fig. 1. The actual SI transition may correspond to a film in the gap between the 9.09- and 9.19-Å-thick films.

We propose that the negative MR effect is associated with fluctuations in the quantum critical region. Its magnitude would be expected to be a measure of the strength of these fluctuations. The increase of the range of the thicknesses over which the effect is seen, together with its weakening as temperature is increased, is consistent with the boundaries of the quantum critical region being determined by the condition $k_BT>\hbar\omega_c\sim|d-d_c|^{\nu z}$, where $\hbar\omega_c$ is the energy scale of the quantum fluctuations, ν is the correlation length exponent, and z is the dynamical critical exponent.[14] Quantum critical behavior would be expected to be cut off at high temperatures when k_BT exceeds some microscopic energy scale in the problem. The negative MR effect disappears at a temperatures above 0.3 K. The thickness at which the maximum effect is found shifts to thicker films as T is increased. This shift would imply that the crossover boundaries defining the quantum critical regime are not symmetric.

Reports of negative magnetoresistance in disordered thin films and wires are not new. Xiong, Herzog, and Dynes (XHD)[15] studied the behavior of quench-condensed, homogeneous, amorphous thin-film Pb wires. They reported a low-field negative MR below the mean-field transition temperature with the field transverse to the wire axis and perpendicular to the plane of the film. Similar behavior was also reported by Marković and collaborators,[16] who studied MoGe wires grown on carbon nanotube substrates. We focus the discussion on the work of XHD as details are available. XHD suggested that the negative MR was enhanced by Coulomb correlations specific to one-dimensional geometries. They further speculated that it might be the result of negative superfluid density fluctuations close to the SI transition. This was proposed by Kivelson and Spivak.[17] Apart from geometry being ultrathin films rather than narrow ultrathin wires, there are a number of other differences between the present work and that of XHD.[15] First, the magnetic field is *parallel* to the plane of the film, whereas it is *perpendicular* to the plane in XHD and in the theory of Kivelson and Spivak.[17] Second, the negative MR of XHD is found above 1 K, whereas in the present work, it exists only below 300 mK. In XHD a number of possible mechanisms for negative MR other than negative superfluid density are considered and ruled out. Nonequilibrium charge imbalance processes associated with phase-slip centers can be excluded in the present work as these processes are found in wires and not in films. Also, the effects we observe are found in the zero-current limit where the current-voltage characteristic is linear. Phase-slip centers would have well-defined signatures in the current-voltage characteristics, which are not seen. Another possibility raised by XHD relates to the quenching by the magnetic field of spin fluctuations associated with electrons singly occupying states in the a-Ge layer. (In their work it was actually an a-Sb layer, but as shown by Hauser,[18] a-Sb and a-Ge are very similar in their properties. These localized electrons were characterized in a-Ge using high-field calorimetry by van den Berg and Löhneysen long ago.[19]) With spin fluctuations quenched by the magnetic field, superconducting fluctuations would be enhanced, leading to a negative MR. In the present work, the range of fields over which negative MR is growing extends to much higher values than those at which spin fluctuations are suppressed using our previous argument. Thus, the negative MR observed is not likely due to the suppression of spin fluctuations of localized electrons.

Another set of potentially relevant experiments are the studies of the SI transition in perpendicular magnetic fields, where a peak, followed by negative MR, is found at fields larger than the critical field of the SI transition. This was observed in In_2O_3 films some years ago by a Bell Laboratories group,[20] and has been reported more recently by groups in Russia, Korea, Israel, and the U.S., respectively.[21–25] One might argue that the data shown here is actually the same physics, but that the magnetic-field scale is dramatically reduced because the films are close to criticality with regard to disorder. This is not likely to be the case. A feature of some of the more detailed reports of a high-field resistance peak[24,25] is that the resistance in the region of the peak at fixed magnetic field is described by $\exp(T_0/T)$. This is not found in our data. Also in our work the peak and the regime of negative MR disappear above some film thickness and there is no trace of them in fields up to 12.5 T in films that are nominally superconducting.

There are a number of other models yielding negative MR in films, such as the work of Beloborodov and collaborators[26] and that of Galitski and Larkin.[27] Since these involve perpendicular rather than parallel magnetic fields they are not as relevant as the work of Lopatin, Shah, and Vinokur,[5] although they may involve similar physics. Yip[28] proposed another mechanism for negative MR. He considered superconductivity confined at a two-dimensional interface with strong surface spin-orbit interaction and showed that an in-plane Zeeman field can induce supercurrent flow. In other words, spin polarization induces supercurrent flow. Although this calculation refers specifically to the superconducting state, the idea might have relevance to the fluctuation regime.

Although the calculations of Lopatin, Shah, and Vinokur[5] are not specific to the present experimental geometry in that their quantum critical point is approached by driving the transition temperature to zero with a parallel magnetic field rather than by controlling disorder, the common features of being close to the quantum critical point and the effect occurring in parallel field, suggest that the underlying mechanism in that calculation and the physics involved in the present work are likely to be the same. A negative MR would then be a signature of critical fluctuations associated with the quantum critical point and an important diagnostic for the SI transition. A detailed calculation relevant to the present configuration would settle the issue.

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