Low Temperature Metallic State of Ultrathin Films of Bismuth

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Measurements of resistance vs. temperature have been carried out on a sequence of quench-condensed ultrathin films of amorphous bismuth (a-Bi). The resistance below about 0.1K was found to be temperature independent in a range of films with thicknesses spanning the superconductor-to-insulator transition that would be inferred by analyzing data obtained above 0.14K. Film magnetoresistance was temperature dependent in the same temperature range over which the resistance was temperature independent. This implies that the low temperature metallic regime is intrinsic, and not a consequence of failure to cool the electrons.

Research on ultrathin superconducting films has been focused on superconductor-to-insulator (SI) transitions [1], which are believed to be quantum phase transitions (QPTs) [2]. In a QPT, tuning an external parameter of the Hamiltonian alters the ground state. The external parameters in previous work have been disorder, varied by changing the thickness of quench-condensed films, and perpendicular and parallel magnetic fields [3,4,5,6,7,8].

A complication in studying QPTs is that they occur at zero temperature, whereas data is acquired down to the lowest accessible temperatures, which are naturally nonzero. Finite-size scaling is used to infer the existence of the QPT, and to extract critical exponents [3]. A successful finite-size scaling analysis is evidence of a QPT, but is not a proof. If other physics intervenes at temperatures lower than those accessed in the measurements, one may draw incomplete, if not erroneous, conclusions [3].

For this reason it is essential to strive to extend measurements to lower temperatures in search of the “true” ground state.

Metallic behavior at low temperatures was recently reported in studies of the field-driven transition of homogeneous MoGe [11] films that would have been considered to be superconducting in the $T \rightarrow 0$ limit, based on a finite-size scaling analysis. Earlier, quench-condensed granular films exhibited metallic behavior in zero magnetic field, beginning at temperatures($\sim 2K$), just below those at which the resistance initially began to drop towards zero [12]. In previous work on ultrathin films of a-Bi grown on an a-Ge layer, there were no indications of a metallic regime down to temperatures of order 0.14K [13].

In this letter we report measurements of $R(T, H)$ of a-Bi films over a range of thicknesses spanning the putative SI transition. We find metallic behavior in zero magnetic field, as evidenced by a nonzero temperature-independent resistance at the lowest measured temperatures, which were of order 0.050K [13]. Considering only data taken at temperatures above 0.14K, this same set of films would appear to be undergoing a thickness-tuned SI transition. A metallic regime was also present when the films were subjected to magnetic fields applied in the plane. Such fields mainly affect the amplitude of the order parameter, as they do not introduce vortices as perpendicular fields do.

Amorphous Bi films were deposited in situ at liquid helium temperatures onto single-crystal SrTiO$_3$(100) substrates, pre-coated (in situ) with a 6Å thick film of a-Ge [13]. This was done in a chamber held at a pressure of $10^{-10}$ Torr. To prevent annealing, substrate temperatures were held below 12K during growth, and below 18K during other processing and handling. Film thicknesses were increased in increments as small as 0.05 Å, as measured using a calibrated quartz crystal monitor. Films processed in this fashion are believed to be homogeneous and disordered [5]. However, a previous experiment showed that a-Ge plays an active role in electrical transport [4]. Critical features of the present experiments are the ability to change the nominal thickness of a film in tiny increments and to grow films that are homogeneous in thickness to one part in $10^4$. This was accomplished by using a large source-to-substrate distance (60cm) and employing Knudsen cells as vapor sources, and maintaining ultrahigh vacuum conditions during growth. All of the effects reported here occurred over a nominal thickness range of order 0.2Å out of approximately 9.0Å, and would not have been seen without such stringent control.

Resistance measurements were carried out in a Kelvinox 400 dilution refrigerator, using either DC or AC four-probe techniques. Electrical leads into the cryostat were filtered at room temperature using π-section filters with a cutoff frequency of about 500 Hz. Extra filtering was employed when a DC current source was used, bringing the cutoff frequency down to about 10 Hz. Power dissipation in the measurement process was kept below 1 pW. Only magnetic fields parallel to the film plane could be applied, with values extending up to 12.5T. In the process of insuring adequate cooling of the films, rotation of the sample platform was restricted to a few degrees. Parallel alignment was adjusted very carefully at room temperature. In high fields, even a slight misalignment will result in a substantial perpendicular field component, which could dominate the high-field physics. For
example, an error of 1° at a field of 12T would result in a perpendicular component of about 0.2T. This is greater than the observed critical component of about 0.2T. It is required that the observed critical component of about 0.2T be smaller than the 0.2T. This lower bound of the critical component of about 0.2T is given the fact that one full turn of the rotator control corresponded to an error of 1°.

The evolution of $R(T)$ of eleven films with thicknesses ranging from 8.5 Å to 9.3 Å is shown in Fig. 1. Thinner films and thicker films, grown in other runs (not shown) were insulating and glass-like in their responses, or fully superconducting, respectively. Focusing on temperatures above 0.14K, insulating and superconducting films appear to be clearly separated into two groups. Although we do not show it here, resistance data of this set of films could be collapsed in the usual manner employing a finite size scaling form, $R(\delta, T) = R_F(\delta/T^{\nu z})$, with a critical exponent product $\nu z$ of 1.1 ± 0.1 in agreement with previous work [1]. The coherence length and dynamical critical exponents are $\nu$ and $z$ respectively, and $\delta$ is the control parameter given by $d - d_c$, where $d$ is film thickness and $d_c$ is a critical thickness. This scaling breaks down when temperature-independent, metallic, resistance data below 0.14K are included in the analysis. It should be noted that this metallic regime was found for both insulator- and superconductor-like films, as might be inferred from the data above 0.14K.

In interpreting a temperature independent $R(T)$, one must consider the possibility that as the measured temperature decreases, the electrons do not cool. This is an important issue for disordered ultrathin films, which, as well as being antennae for electromagnetic radiation, have tiny heat capacities. Apart from heating by the measuring current, which is unlikely because I-V characteristics were always linear, electromagnetic noise is likely the major source of heating. Noise can be external or internal to the cryostat. The latter would be Johnson noise in the electrical leads that are in equilibrium with black-body radiation. These leads are at temperatures higher than that of the film over much of their length. To be certain that the electrons have cooled would require measurement of some well-understood property of the film and its use as a thermometer, or the provision of a separate thermometer that is in certain thermal contact with the film that has a response to possible heating effects from the environment identical to that of the film. Since neither of these possibilities was available, we rely on indirect arguments. These will emerge in the course of the discussion of $R(T, H)$, which follows.

In Fig. 2, $R(H)$ at various temperatures is plotted for the 9.3 Å thick film. At the highest temperatures, in the lowest fields, $dR/dH$ is slightly positive. With increasing field, a regime in which $dR/dH$ becomes negative is entered. This is followed by a minimum in $R(H)$, and finally an upturn at higher fields. In sufficiently high fields superconducting fluctuations are completely quenched. The regime in which $dR/dH < 0$ is possibly associated with the suggestion of Kivelson and Spivak [17], that a local order parameter density can fluctuate from point to point in sign as well as in magnitude in disordered systems near the SI transition. This can bring about a negative magnetoresistance in a perpendicular magnetic field. As stated previously the small error in alignment can result in a small component of magnetic field perpendicular to the plane.

The systematics of the magnetoresistance provide evidence that the metallic regime is intrinsic. The negative magnetoresistance grows as $T$ decreases as shown in Fig. 2, but becomes abruptly positive when $R(T)$ becomes temperature independent at low temperatures. It continues to increase with further decrease of $T$. This can be seen in the inset of Fig. 2 which emphasizes the low field regime. The magnetoresistance in the metallic regime is a function of temperature, both in its initial change with field and for all values of field. If the saturation of the resistance were a consequence of electrons not cooling then these effects would not be expected.

The upturn of $R(H)$ in high parallel fields is hard to explain quantitatively as it is a consequence of the combined action of the large parallel component of the field and the very much smaller, but unknown perpendicular component. This linear dependence of $R(H)$ on field would be expected if there were flux flow resistance due to a perpendicular field component [13]. It should be noted that at high fields there is a significant contribution to the resistance which is quadratic with field. This could be associated with the weakening of the order parameter amplitude fluctuations by the parallel component of field.

Further interesting effects are seen if the temperature dependence of the resistance is plotted for fields above the magnetoresistance minimum. The quenching of superconducting fluctuations in this regime is shown in Fig. 3 for the 9.19 Å thick film. Its low temperature behavior is always metallic. As the field is increased, the domain in temperature over which $dR/dT = 0$ broadens significantly and reaches a maximum width of about 100mK. This maximum width can be seen much more dramatically in Fig. 4 which shows data from the 9.09 Å thick film, in which the temperature of the onset of the metallic regime is plotted as a function of its resistance. For this film, the resistance of the widest metallic regime was 12,900Ω, which is remarkably close to twice the value of the quantum resistance for pairs. This resistance is actually equal to the zero-field resistance of the film in the low temperature limit. This particular film is the first of those in the sequence of films shown in Fig. 1 that exhibits superconducting fluctuations as evidenced by a downturn in $R(H)$ at higher temperatures. All of the films exhibiting superconducting fluctuations display a similar response to an applied parallel magnetic field. It is important to note that the systematics of this variation of resistance with field are different from what was reported by Mason and Kapitulnik [13]. This may be a consequence of the fact that the field in this work was parallel rather than perpendicular to the film plane.
One may compare the present results with those obtained in studies of quench-condensed $\alpha$-Ga films which behaved in a manner very similar to the films of the present work, but became metallic at temperatures as high as $2K$ \[2\]. In that work, the temperatures at which $dR/dT$ first fell to zero were dependent on the measuring current. In the subsequent work of Ref. \[4\], on similar films, it was proposed that the nonlinearities and metallic behaviors in films that appeared to become insulating at high temperatures were a consequence of charge motion, whereas for those that appeared to become superconducting, they were due to vortex motion. This was arrived at because the systematics of the data suggested that insulating and superconducting states at $T = 0$ would only be found in the limit of zero measuring current. Although the I-V characteristics were nonlinear, the systematics of the data could also be used to eliminate the possibility of heating. It is conceivable that similar nonlinear features of the I-V characteristics for $\alpha$-Bi films exist, but their observation would require much lower current levels than were employed in the measurements.

Metallic regimes at low temperatures in ultrathin films that exhibit local superconductivity have been the subject of several theoretical works, which emphasize the bosonic nature of these systems as well as, in some instances, the role of dissipation \[21\][22][23][24]. The data we have presented cannot validate any of these theories, some of which make very specific predictions, such as glass-like behavior \[23\]. On the other hand, the data can be used to make a case for the metallic regime being intrinsic and not a consequence of failure to cool the electrons.

The values of either thickness or magnetic field that result in maximally wide metallic regimes could be evidence of the metallic quantum critical point of the SI transitions proposed in the context of phase-only Bose Hubbard models \[25\][26]. We conjecture that when the tuning parameter of the quantum phase transition is close to its critical value, evidence of quantum critical fluctuations, in this case, a metallic regime, extends to high temperatures, resulting in behavior such as that exhibited in Fig. 4. In this scenario metallic behavior at other fields (or thicknesses) is seen at nonzero temperatures because the free motion of vortices and charges that respond to the measuring current masks the superconducting and insulating ground states, as discussed above. The actual value of the resistance of the separatrix for the thinnest film exhibiting superconducting fluctuations is very close to $h/2e^2$ which is double the quantum resistance for electron pairs. It turns out that this is the value of the critical resistance obtained from finite-size scaling of Monte Carlo data in simulations of the phase-only Bose Hubbard model \[26\]. This scenario implies that there is a quantum critical point separating insulating and superconducting phases, with the metallic regime that obscures it resulting from a nonzero measuring current. Alternatively, there might not be a nonzero critical point at all. In this instance, the striking magnetic field dependence of the temperature onset of the metallic regime such as shown in Fig. 4 would be just a feature of the phase diagram of the putative Bose metal regime.

In summary, the extension of measurements of $R(T)$ of ultrathin, quench-condensed films of $\alpha$-Bi down to temperatures of order $0.05K$ have revealed metallic behavior over a range of film thicknesses near the SI transition. This suggests that the apparent superconducting and insulating ground states inferred from the analysis of data obtained at temperatures above $0.14K$ and discussed in many theories, may not be realized. Evidence was presented supporting the results being intrinsic, and not a consequence of the electrons in the film not being cooled. Lower temperatures and lower measuring current densities may be needed to find the “true” ground state, at least for films in the transition regime between insulating and superconducting behavior.

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\bibitem{13} The actual low temperature limit on the sample holder was close to $0.020K$ when the refrigerator was cooled to its base temperature. This was determined from other work using a calibrated thermometer mounted on it. The calibration for the thermometer used for the present studies had a lower limit of $0.050K$, hence the use of the term “measured temperature.” All of the data shown refers to the latter, but
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the metallic regime discussed here actually persisted to the
temperature reached when the refrigerator was allowed to
cool to its base temperature.
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FIG. 1. Evolution of $R(T)$ for a series of eleven different thicknesses of Bi. Film thicknesses are: 8.5, 8.7, 8.8, 8.85, 8.91, 8.99, 9.05, 9.09, 9.19, 9.25, and 9.3 Å. The 50mK resistance of the nominally critical curve is very close to 12,900 Ω, twice the quantum resistance for pairs.

FIG. 2. Evolution of $R(H)$ at various temperatures for the 9.3Å thick film, with H in-plane. Temperatures, for curves from top to bottom are: 500, 250, 200, 150, 100, and 50mK. Inset: the same data plotted as percentage change in the resistance vs. field. The range is limited to 1 T, so as to highlight the low-field behavior. The two upper curves were obtained at 100 mK and 50 mK, which are in the metallic regime.

FIG. 3. Evolution of $R(T)$ of the 9.19Å film as a function of in-plane magnetic field. Field values from top to bottom are: 12.5, 12, 11.6, 11.5, 11, 10, 9, 8, 7, 6, 5, 4, 3, 2, and 0 T.

FIG. 4. Plot of temperature $T_{flat}$ at which $R(T)$ becomes temperature independent vs. the value of that resistance for the 9.09Å thick film. A sharp peak at a sheet resistance of 12,900 Ω is evident.
