Electric-field effect in ultrathin films near the superconductor-insulator transition

N. Marković,* C. Christiansen, G. Martinez-Arizala, and A. M. Goldman

School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455

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The effect of an electric field on the conductance of ultrathin films of metals deposited on substrates coated with a thin layer of amorphous Ge was investigated. A contribution to the conductance modulation symmetric with respect to the polarity of the applied electric field was found in regimes in which there was no sign of glassy behavior. For films with thicknesses that put them on the insulating side of the superconductor-insulator transition, the conductance increased with electric field, whereas for films that were becoming superconducting it decreased. Application of magnetic fields to the latter, which reduce the transition temperature and ultimately quench superconductivity, changed the sign of the reponse of the conductance to electric field back to that found for insulators. We propose that this symmetric response to capacitive charging is a consequence of changes in the conductance of the a-Ge layer, and is not a fundamental property of the physics of the superconductor-insulator transition as previously suggested.

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Investigations of the effect of a perpendicular applied electric field on insulating granular Au films¹ and amorphous In_2O_3 films² revealed that the conductance increased for both polarities of field. This symmetric response was ascribed to nonequilibrium transport phenomena specific to very disordered systems. Long relaxation times and memory, aging and hysteresis associated with the symmetric response support the picture that these systems are Coulomb glasses with long equilibrium times.^{3,4} More recently, the inhomogeneous nature of charge transport and the slow relaxation in such systems has been studied theoretically^{5,6} and experimentally as a function of disorder and magnetic field.^{7,8}

A similar symmetric response to an applied electric field was found in ultrathin amorphous films of Bi and Pb Refs. 9,10 grown on amorphous Ge (a-Ge). We speculated that in such very resistive films the variation of the conductance with electric field was proportional to the dependence of the electronic density of states on energy, since the excited state caused by the applied field cannot relax in very glassy systems.¹⁰ This conjecture has been substantiated by recent simulations of the time development of the Coulomb gap in a Coulomb glass.⁵ There are several problematic aspects to this interpretation: in ultrathin films of Bi and Pb, the symmetric response persisted into regimes where the glassy behavior vanished, such as at high-temperatures and in moreconductive (thicker) films. Furthermore, when the films were thick enough to become superconducting, the response changed sign and the conductance above the transition temperature *decreased* as a function of the applied electric field.⁹ The largest-fractional-conductance modulation was observed deep in the insulating regime, and it decreased as film thickness increased and the insulator-to-superconductor transition was approached. At the transition, the symmetry disappeared, and for a small range of thicknesses around the transition, the field effect was approximately antisymmetric, before changing sign in films that were thick enough to be superconducting at low temperatures. The carrier density in these studies could not be increased enough to drive a nonsuperconducting film into the superconducting state, almost certainly a consequence of the high density of trapping sites.

A qualitative interpretation of these results that was presented, included consideration of the possibility of Cooper pairing in insulating films and symmetry between the insulating and the superconducting states.

Here we report on investigations of the electric-field effect in ultrathin films of Bi at temperatures that traverse a range of temperatures an order of magnitude lower in temperature than those of our previous work. In addition, the superconductor-insulator transition was tuned not only by changing film thickness, but by the application of a perpendicular magnetic field. Films thick enough to become superconducting and exhibiting a conductance above their transition that decreased with applied electric field, were made to revert to the behavior of insulating films whose conductance decreased with electric field. This ensued on application of a perpendicular magnetic field. Although these results are a consistent extension of earlier work, they lead to a different interpretation. We now propose that the symmetric response to an electric field for both superconducting and insulating films is due to the conductivity of the amorphous Ge layer being increased by capacitive charging, rather than any underlying symmetry between the insulating and superconducting states.

These investigations were carried out on a series of ultrathin Bi films, evaporated on top of a 10-Å thick layer of *a*-Ge, which was predeposited onto a SrTiO₃ (100) substrate. The substrate temperature was kept below 20 K during all depositions, and all of the films were grown *in situ* under ultrahigh vacuum conditions ($\sim 10^{-10}$ Torr). The film thickness was gradually increased through successive depositions in increments of 0.1–0.2 Å. Films grown in this manner are believed to be homogeneous, since they become connected at an average thickness of about one monolayer.¹¹

For capacitive charging studies, the electric field was applied perpendicular to the plane by biasing the film relative to a 100-nm thick Au-metal gate on the back of the SrTiO₃ substrate, which was 0.75 mm thick. Even though these substrates are of macroscopic thickness, a substantial charge can be induced at relatively low-gate voltages because of their high dielectric constant below 10 K ($\kappa \varepsilon_0 \sim 8000-20000$).¹²



FIG. 1. Sheet resistance as a function of temperature for a 14-Å thick Bi film in different magnetic fields: 0, 1, 2, 3, 4, 5, 8, 11, and 12 kG, from bottom to top.

Resistance measurements were carried out between depositions using a standard four-probe dc technique. Low-bias currents (<50 nA) were used to avoid Joule heating of the sample and to make sure that the voltage across the sample was a linear function of the applied current. As the film thickness increased from 7 Å to 15 Å, the temperature dependence of the resistance of the system changed from insulatorlike (dR/dT < 0) to superconductor-like (dR/dT > 0) at low temperatures.¹³ There was no sign of the quasireentrant behavior observed in granular films.¹⁴ In order to study the electric-field response of a film across the magnetic-field driven superconductor-insulator transition, we focussed on films that were superconducting in zero field, but could be driven insulating by applying a magnetic field perpendicular to the plane of the sample.¹⁵

The sheet resistance of a representative film as a function of temperature at different magnetic fields is shown in Fig. 1. In zero field, the resistance decreases as the temperature decreases, indicating the onset of superconductivity. At the lowest temperatures, as the magnetic field is gradually increased, the temperature coefficient of the resistance, dR/dT, changes sign, indicating insulating behavior.

The change in the conductance with gate voltage in zero magnetic field of the same film at four temperatures is shown in Fig. 2. It is evident that the conductance decreases with applied gate voltage at temperatures for which dR/dT>0 (compare to the zero-field curve in Fig. 1.), and the effect becomes smaller at higher temperatures. This is similar to the result Martinez-Arizala *et al.*,⁹ obtained for superconducting Pb films. No relaxation effects were observed when gate voltage was applied in this regime.

The change in conductance as a function of gate voltage at a temperature of 0.15 K in different magnetic fields is shown in Fig. 3. The conductance is found to decrease for both polarities of the gate voltage in low fields, even though the effect is not perfectly symmetric. At some value of the magnetic field, where dR/dT is still positive, but small, the effect becomes approximately antisymmetric. The data for positive polarity of the gate voltage look remarkably similar to the data shown in Fig. 2, even though the sample was



FIG. 2. Fractional change in conductance of a 14-Å thick Bi film as a function of gate voltage at 0.15 K (triangles), 0.2 K (diamonds), 0.3 K (squares), and 0.4 K (circles).

driven into the insulating state by increasing the magnetic field in one case, and by increasing the temperature in the other.

In a magnetic field that is high enough to bring the sample into the dR/dT < 0 regime, the conductance increases as the gate voltage is applied, as shown in Fig. 4. Even though the effect is relatively small, it is reproducible. The results are remarkably similar to the case when the superconductorinsulator transition is tuned by changing the film thickness.⁹ Furthermore, similar results are obtained if the sample is driven out of the superconducting state by increasing the temperature. The sign and value of dR/dT seem to indicate whether the field effect will be positive or negative, and whether it will be symmetric or antisymmetric.

We now address the physical nature of the capacitive charging experiment. In particular we will consider the issue of why the conductance-gate-voltage characteristic resembles the density of states of a disordered system. The Efros-Shklovskii density of states (DOS) for two-



FIG. 3. Absolute change in conductance of a 14-Å thick Bi film as a function of gate voltage at 0.15 K in zero magnetic field (circles), and in a perpendicular magnetic field of 1 kG (squares), 2 kG (diamonds), 3 kG (crosses), and 4 kG (triangles).



FIG. 4. Absolute change in conductance of a 14-Å thick Bi film as a function of gate voltage at 0.15 K in a perpendicular magnetic field ranging from 1 kG (bottom) up to 12 kG (top).

dimensional disordered systems is a V-shaped entity with its apex centered on the Fermi energy (E_F) .^{16–18} Conductance studies do not usually probe the DOS of electronic systems, since disturbances to the electron-energy-distribution relax, and carriers added during a measurement are quickly screened.¹⁹ This does not occur in the case of tunneling, as the physical process, characterized by the tunneling time, is short in comparison with the time-of-charge relaxation. As a consequence, tunneling is the standard approach to the measurement of the density of states.^{20–25}

The usual situation in a capacitive-charging experiment is one in which the electron-distribution relaxes after the gate voltage and carrier-concentration are changed. The new carrier-concentration determines the value of the chemical potential, and the minimum of the DOS tracks that new value.¹⁶ We pointed out earlier that in glassy systems with long relaxation times, there may not be a path from the excited state resulting from injected carriers back to the state of minimum free energy, because of barriers in the free-energy landscape. Thus changing the gate voltage would change E_F , but the minimum of the DOS would not track it. In this circumstance, if the conductance were proportional to the density of states, as is the case in the hopping regime,²⁶ a simple field effect conductance-modulation study could map out the dependence of the DOS on energy. Indeed, recent simulations have shown that the time development of the Coulomb gap in the DOS can involve very long time scales due to electron hopping and rearrangement.⁵

In the case of the ultrathin films of metals deposited on top of a thin *a*-Ge layer grown on a $SrTiO_3$ substrate, the geometry is very critical to understanding the evolution of the response to capacitive charging. For the very thinnest films, the order of a 10-Å thick layer of metal on top of a 6-Å to 10-Å thick layer of *a*-Ge, the system behaves as an electron glass, and the conductance modulation, which can be a significant fraction of the total conductance, is a direct measure of the DOS by the argument given above. The relevant density of states is in effect with that of the *a*-Ge layer, with the metal layer acting as a source of electrons. In fact, tunneling measurements of the electronic density of states of amorphous $\text{Ge}_{1-x}\text{Au}_x$ show a very similar shape as a function of the gate voltage as that found in our experiments.²¹ The response is symmetric in voltage because of the symmetry of the DOS for a disordered system.

As film thickness is increased, the glasslike response of the conductance disappears, but there is a small conductance change that still exhibits a V-shape, characteristic of the DOS of a disordered system. This behavior can be understood if one appreciates the continued presence of the a-Ge underlayer, in which charge can be confined. When the gate voltage is increased, charges are drawn out of the electrodes connected to the film, and the metal-film's carrier concentration is changed by a small amount. The gate voltage and the effective capacitance to the gate determine the amount of charge in the *a*-Ge layer, which determines the chemical potential. The metal layer permits a rapid change of the chemical potential, but internal to the *a*-Ge the charges have a long relaxation time, because the *a*-Ge is extremely disordered and separated from the metal film by a Schottky barrier.²⁷ By the arguments given above, this results in changes in the conductance of that layer proportional to its DOS. The total fraction of the modulation of the conductance attributable to the a-Ge layer is only a small fraction of the total conductance, so the overall effect is small.

When the metal film becomes superconducting other physics comes into play. Increasing or decreasing the gate voltage from zero increases the conductance of the a-Ge layer. The combination of the superconducting layer and the *a*-Ge layer can be considered to be a proximity sandwich. Increasing the conductance of the a-Ge layer will make it more metallic, causing the electrons in the sandwich to have a higher probability of being in the *a*-Ge layer. Thus the average-attractive electron-electron interaction that they experience will be reduced, lowering the effective transition temperature of the sandwich. The proximity of a metallic layer has been shown to reduce the superconducting transition temperature²⁸ in some cases. At fixed temperature, in the regime where the resistance is decreasing with decreasing temperature, a reduced transition-temperature derived from capacitive charging, will result in an increase of the resistance with increasing gate voltage (of either sign). Thus the effect of initiating the superconducting transition in the overlay film is to flip over the V-shaped response to the gate voltage. Again, because the *a*-Ge layer at this point contributes only a small part of the total conductance of the composite film, the effect is small. Correspondingly, quenching superconductivity with a magnetic field, flips the conductance-voltage characteristic back to its behavior in the insulating regime.

It should be noted that the original goal of these studies, which was not realized, was to induce superconductivity through changing the carrier concentration by capacitive charging. Superconductivity has been induced in a similar experimental configuration by Schön *et al.* in organic crystals,²⁹ but at much-higher gate voltages than were used in our studies of ultrathin metal films. It remains the subject of future investigations as to whether superconductivity can be induced in ultrathin metal-films deposited on *a*-Ge by

capacitive charging, if the gate voltage were large enough to transfer sufficient charge to both fill all of the traps in the a-Ge and change the carrier concentration in the metal layer.

In summary, we have performed a field-effect conductance-modulation experiment on ultrathin films of Bi deposited onto amorphous Ge near the thickness- and magnetic field-tuned superconductor-insulator transitions. We have swept through the superconductor-insulator transition using a magnetic field, and we see the same change of sign in the field-effect response observed when the superconductorinsulator transition is traversed by changing thickness. The

*Present address: Department of Physics, Harvard University, Cambridge, MA.

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results of these studies together with our previous work on both Pb and Bi,^{9,10} lead us to propose that the small symmetric response in this regime, where no glassy behavior was observed, is due to increasing the conductivity of the insulating amorphous germanium underlayer, rather than some symmetry between the insulating and the superconducting states.

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- 27 By depositing very small amounts of metal on top of the *a*-Ge layer, we may in fact be doping the Ge, and the observed conductance is partially due to the doped Ge film. When the Bi film on top actually becomes metallic, it shunts the Ge film, and a Schottky barrier forms at the interface.
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