Electrical Transport of Spin-Polarized Carriers in Disordered Ultrathin Films

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Slow, nonexponential relaxation of electrical transport accompanied by memory effects has been induced in quench-condensed ultrathin amorphous Bi films by the application of a parallel magnetic field. This behavior, which is very similar to space-charge limited current flow, is found in extremely thin films well on the insulating side of the thickness-tuned superconductor-insulator transition. It may be the signature of a collective state that forms when the carriers are spin polarized at low temperatures and in high magnetic fields.

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The competition between interactions and disorder leads to behavior which is glasslike. Disordered electronic systems such as doped semiconductors and disordered metal films, which exhibit electrical versions of glass behavior, are described as Coulomb or charge glasses. The experimental investigation of the response of the conductances of these systems to capacitive charging (the field effect) has proved to be a useful way to study manifestations of glassy behavior such as hysteresis, slow nonexponential relaxation, and memory [1–4]. Aspects of these investigations have been captured by the simulations of Yu [5] based on the model of Baranovskii, Shklovskii, and Efros [6]. Recently the study of glasslike behavior in disordered two-dimensional (2D) systems has been focused on magnetic field-induced phenomena. Bielejec and Wu [7] have reported hysteresis and slow nonexponential relaxation of the conductance of ultrathin granular Al films, induced by magnetic fields at low temperatures and the temperature dependence of the resistance noise spectral density.

In this Letter we report slow, nonexponential relaxation and memory effects in nominally homogeneous, quench-condensed ultrathin films of amorphous Bi (a-Bi). These effects were induced by the application of a parallel magnetic field in a manner similar to that reported in Ref. [7]. However a systematic study of the variation of the voltage drop along the film with time suggests that the transport resembles space-charge-limited current flow [8]. The sheet resistances of the films exhibiting these effects placed them well on the insulating side of the thickness-tuned superconductor-insulator transition [9] and were never less than $10^5\ \Omega$, at 300 mK, a full decade larger than those of the granular films studied in Ref. [7]. We have also identified field and temperature parameters associated with the onset of the effect. These lead us to suggest that the observed behavior is a collective rather than a single-particle phenomenon. We speculate that these observations are a signature of a Wigner glass such as that discussed by Chakravarty and collaborators [10].

Amorphous Bi films were grown using a system that combines an Oxford Instruments dilution refrigerator (Kelvinox 400) with a "bottom loading" sample transfer-film growth system [11]. The geometry of the evaporation sources and ultrahigh vacuum environment were nearly identical to those used in earlier work [12]. The vapor sources were commercial Knudsen cells, and the flux density at the substrate was uniform to better than one part in 10^4, permitting effects associated with very tiny changes in thickness to be studied. Although the absolute values of thicknesses are nominal, the relative thicknesses and increments of thickness are quite accurate. They are determined by timing the depositions at a fixed rate of 0.01 Å/s. The rate monitoring was measured by depositing a film at a measured rate for a known time and measuring its thickness using a profilometer. All electrical lines to the sample were filtered with a cutoff frequency of about 500 Hz. The current source was filtered with a cutoff frequency of about 10 Hz. Measuring instruments were optically isolated from the controlling computer.

The epipolished $SrTiO_3$ wafers that were employed as substrates were first cooled to helium temperatures, and then precoated with a-Ge, before Bi film growth was initiated. The resultant a-Bi films are believed to be disordered on microscopic length scales [13]. Curves of $R(T)$ for a series of 19 films with thicknesses starting at 11.15 Å, and up to 13.35 Å, are shown in Fig. 1. As the investigation of the superconductor-insulator transition was the initial goal of the measurements, low temperature magnetotransport data, which may have revealed glasslike behavior, were not collected for the first few films of the sequence. Glasslike behavior was observed in films shown in bold in Fig. 1. It was not found in films thicker than 11.85 Å. Superconductivity or superconducting fluctuations did not appear until the nominal film thickness reached 13.35 Å.

For the thinnest films, at temperatures above about 100 mK and in zero magnetic field, $R(T)$ could be fit by the functional form $\exp[(T_0/T)^{0.7}]$. The exponent is different from 0.5, the expectation for variable range hopping including Coulomb effects [14], but close to the value 0.75 found in many other studies [15]. The sample temperature could not be stabilized above 1 K in the large dilution refrigerator. This limited the precision of the

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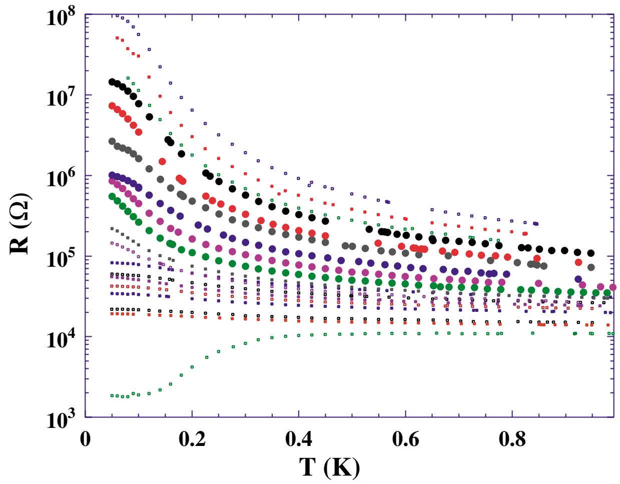

FIG. 1 (color online). Sheet resistance vs temperature for a series of films with thicknesses, from top to bottom, of 11.15, 11.25, 11.37, 11.38, 11.43, 11.48, 11.55, 11.65, 11.75, 11.85, 11.95, 12.03, 12.17, 12.27, 12.4, 12.55, 12.65, 12.85, and 13.35 Å. Those films for which glasslike behavior was studied, as described in the text, are in bold. Although the use of sheet resistance is the preferred way to discuss two-dimensional systems, one should note that resistivity $\rho = Rd$, where d is thickness.

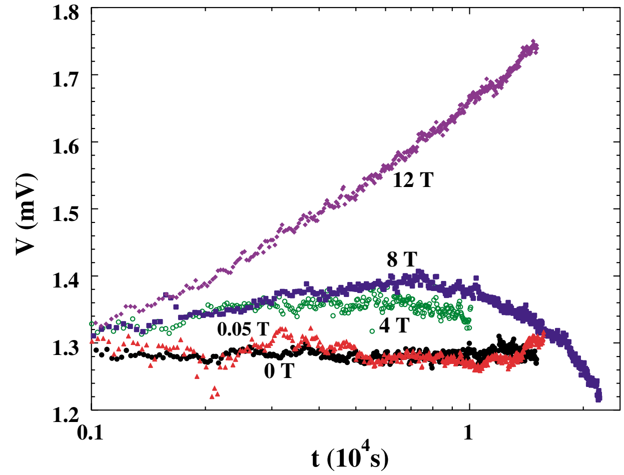

FIG. 2 (color online). Voltage vs time at $T = 50$ mK for an 11.38 Å thick film in zero magnetic field and in several magnetic fields.

exponent, possibly accounting for it being different from the value obtained in previous studies.

The measurement technique, which revealed slow relaxation, involved first setting a current in the film using a constant current source. In all of the measurements, this current was 7×10^{-11} A. (This was the smallest convenient current in a range of currents for which the I-V characteristics of the films in steady state were linear in zero magnetic field.) The films were 2.5 mm long and 0.5 mm in width. Electrical leads were 100 Å thick preevaporated Pt films. In zero magnetic field, voltage, as measured across two leads, 0.5 mm apart and centered between the current leads, developed in response to the current in a time on the order of the RC time constant of the measuring circuit. For films exhibiting slow nonexponential relaxation, after this initial transient, the voltage continued to change. As a technical matter, our observations of ultraslow relaxation were not a consequence of downward drift in the magnetic field produced by improper operation of the superconducting magnet in the persistent current mode. To rule this out, measurements were repeated with the magnet in the driven mode, and the results were found to be identical to those obtained in the persistent mode. The voltages measured in field were linear in current, but extensive studies of the current dependence of the response were not made for fear of heating the film.

Slow relaxation effects were first noticed in an 11.38 Å thick film at 50 mK. The change of voltage with time was slower the higher the magnetic field as shown in Fig. 2. In fields less than 0.1 T, after the initial transient response, there was no slow relaxation. In 12 T at 50 mK, at long times, the voltage grew logarithmically with time. After about 10^4 s the fraction of the measured voltage drop, changing with time, was about 1/3 of the total measured voltage. The effect vanished above 200 mK even in a 12 T field.

From Fig. 2 one can see that the drift curves obtained in intermediate fields were not logarithmic in time and sometimes not even monotonic. Simple logarithmic dependencies were found only in the highest fields at long times. This nonmonotonic temporal evolution of the voltage was originally thought to be a signature of the voltage compliance limit of the current source being reached. It was subsequently realized that such behavior is similar to space-charge-limited current flow in the presence of trapping [16]. In this circumstance charge enters the film from the metallic current leads at its ends. A charge front forms and migrates away from the leads. The charge eventually spreads uniformly through the film. A simple picture is to consider the current electrodes as plates of a planar capacitor. The space-charge regions that extend away from the electrodes into the film are effectively extensions of these plates. For electrons the charge front would progress from the negative electrode and for holes from the positive electrode. As the front moves into the film, the electric field increases as the effective spacing of the plates decreases. When the space charge is spread out over the film the electric field and the voltage fall. In models of this behavior, the maximum in the voltage vs time is related to the transit time of this charge front along the film. The rise and fall of the electric field is quite apparent in the 8 and 4 T curves of Fig. 2. This downturn in the voltage was not seen in films measured in a field of 12 T, where the voltage changed monotonically with time. In this case, the movement of the charge front is so slow that it never enters the region between the voltage electrodes during the measuring time.

The thickness dependencies of the glasslike behavior at a fixed magnetic field of 12 T and at a fixed temperature of 50 mK are shown in Fig. 3. The fraction of the current responding in a glasslike manner decreases with increasing film thickness and the effect vanishes completely when the film thickness reaches 11.95 Å. The very long times in these measurements precluded a truly systematic examination of the dependence of the voltage on current, temperature, and magnetic field for each film. As a consequence, it is not known whether there is a well-defined transition to this slow relaxing state or whether the onset is a crossover.

An important observation relating to memory is shown in Fig. 4. With the temperature at 50 mK, the magnetic field set at 12 T, the current source connected, and the voltage across the sample increasing, the current source was switched off for a short time, and then switched back on. The voltage was observed to drop to a low (actually negative) value in a time given by the circuit time constant when the source was switched off. When it was restored, the voltage rose within the circuit time constant to the value it exhibited before the current source was disconnected at which point it resumed its upward drift. The negative voltage found with the current source off suggests that the film was losing stored space charge during that time. Another memory effect, which is not shown, involved temperature. Starting at a temperature of 50 mK and a field of 12 T, the current source was turned off and the temperature raised to 100 mK. The current was then restored and the voltage was seen to first grow to its previous value at 50 mK, and then decay to a value associated with 100 mK, after which it continued to drift up, but at a different rate.

One can rule out all explanations of glasslike behavior associated with superconductivity [17,18] as the films in which slow relaxation was found did not exhibit either superconductivity or superconducting fluctuation effects in zero magnetic field. In the work of Ref. [7], the 25 Å thick Al films were superconducting in zero field and were measured in fields above the paramagnetic limit, raising the possibility that their observations may somehow be associated with residual superconducting phenomena [18].

Another mechanism that can inhibit conduction, possibly leading to slow relaxation effects, is the decrease in the wave function overlap produced by a magnetic field. This yields a positive magnetoresistance, but is not likely to be applicable, because the threshold field for nonexponential relaxation, 0.1 T, is so low. This will lead to a magnetic length much larger than the hopping distance and will thus not affect the hopping rate [5].

The argument that spins are relevant to our observations depends on the effect being found in a magnetic field parallel to the plane of the film. A theory of positive magnetoresistance resulting from the polarization of the spins of electrons involved in conduction by hopping has been given in Ref. [19]. 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 field strong enough to polarize the carriers, transitions involving such sites would be forbidden as electron pairs cannot form singlets. The field would thus

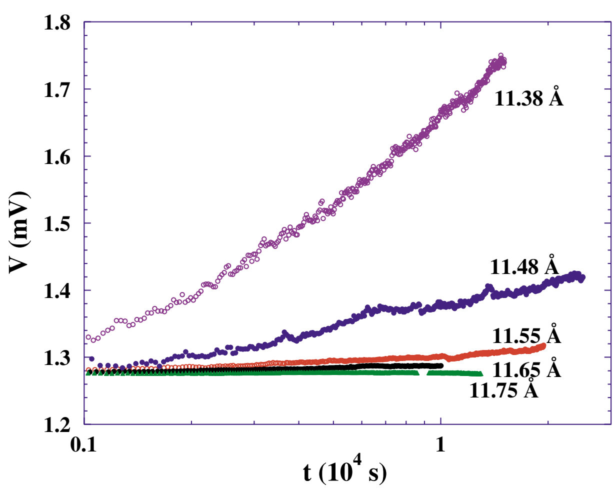

FIG. 3 (color online). Voltage vs time for a series of films of different thicknesses at a temperature of 50 mK and in a magnetic field of 12 T for films of several thicknesses. The vertical axis has been adjusted to take out contributions from small voltage offsets that are due to thermal emfs that cannot be removed in the standard way as the bias must remain fixed. Some data are not shown for clarity.

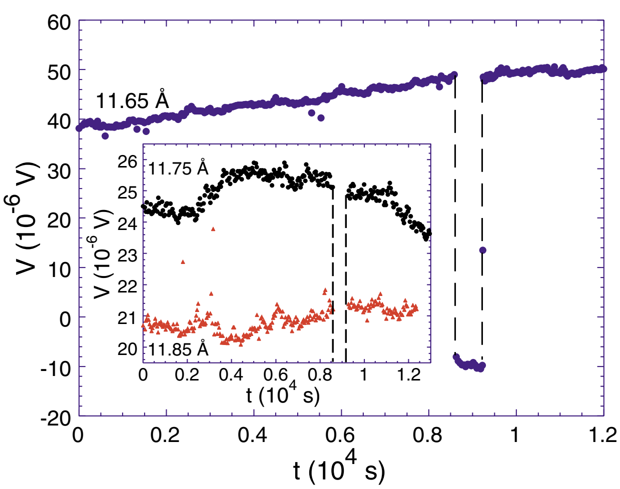

FIG. 4 (color online). The main curve is the voltage vs time for an 11.65 Å thick film. The important feature is the response when the current source is turned off for a short while and then turned back on. The inset panel shows similar data for 11.75 and 11.85 Å thick films. Since the effect is smaller for those thicknesses the voltage axis has been offset. The dashed lines are guides for the eye.

shut off certain hopping transitions. This picture has been considered as an explanation of the magnetoresistive behavior of Si-based two-dimensional electron gases exhibiting variable range hopping [20]. It is conceivable that transport could be sufficiently inhibited by this mechanism to result in space-charge limited conduction.

However other features of the data can be used to rule out single-particle effects. Focusing on the 11.38 Å thick film, one can compute the ratio of the Zeeman energy $\mu_B H$ associated with the threshold field of 0.1 T with the thermal energy $k_B T$ at 50 mK. The former is 9.25×10^{-25} J, whereas the latter is 6.9×10^{-25} J, corresponding $\mu_B H/k_B T = 1.34$, which is qualitatively consistent with a picture involving spin polarization of carriers involved in hopping conduction. Slow relaxation in this film in a field of 12 T disappears above 200 mK, which corresponds to a ratio $\mu_B H/k_B T = 40$. This is not consistent with the effect being due to single-carrier spin polarization. If such were the case the phenomenon would disappear when the Brillouin function fell below its saturation value, or when $\mu_B H/k_B T < 1$ and not when $\mu_B H/k_B T \gg 1$. An additional feature inconsistent with single-particle spin polarization is the absence of saturation in the change of the slow relaxation with increasing magnetic field (see Fig. 2.). In the work of Ref. [20], the magnetoresistance saturated as expected, since spin polarization as given by a Brillouin function saturates when $\mu_B H/k_B T \sim 1$. Saturation was not observed in our data for the 11.38 Å thick film in fields up to 12 T at 50 mK. This corresponds to $\mu_B H/k_B T = 160$. In fact, the change in the relaxation is most pronounced between 8 and 12 T. These observations suggest that there must be another energy scale in the problem other than $k_B T$, which determines the onset of the glasslike state.

We can speculate that slow relaxation effects are due to a collective state occurring in a disordered, spin-polarized, two-dimensional electronic system. For collective charge effects to occur electron-electron interactions must be important. The usual measure of their role is the electron gas parameter r_s, which is the average interparticle separation in units of the effective Bohr radius, $a_B = \hbar^2 \epsilon / me^2$. Here ϵ is the dielectric constant. Unfortunately we could not estimate r_s from the carrier concentration, as the Hall resistance of these samples was too small to be measured, suggesting a very low carrier mobility or equal numbers of electrons and holes.

One possibility for the collective state is the disordered 2D Wigner glass in a magnetic field that was discussed in Ref. [10]. The application of an electric field to such as system would result in the glass sliding [21]. The dynamics of this sliding might resemble our observations. The determination of the actual picture will require measurements that test specific theoretical predictions.

In summary, we have observed a severe inhibition of the motion of charge induced by the application of a parallel magnetic field. This leads to space-charge-limited conduction which exhibits many features of a glass, perhaps a Wigner glass. The detailed theoretical description of this state is an open question.

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