Disorder and Magnetic Field Dependence of Slow Electronic Relaxation

Zvi Ovadyahu
The Racah Institute of Physics, The Hebrew University, Jerusalem 91904, Israel

Michael Pollak
Department of Physics, The University of California Riverside, California 92521
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We show that the relaxation of the excess conductivity in a hopping system driven far from equilibrium is systematically affected by static disorder and magnetic field. In following the glasslike relaxation for times much longer than has been done before we observe a new time dependence of the relaxation. A positive magnetoresistance and a strong effect of the magnetic field on the relaxation indicate that spins may play an important role in the nonequilibrium properties of such systems. [S0031-9007(97)03583-7]

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Many disordered systems are characterized by a very slow, nonexponential time dependence of relaxation to equilibrium. There is no all-inclusive explanation or form for the temporal relaxation law, but many experiments suggest that it is a feature common to the combination of disorder and interactions [1]. It was also suggested that constraints not due to interactions, e.g., conservation of particles, may be sufficient in some cases to cause nonergodic relaxation [2]. This suggestion was made in the context of electronic Fermi glasses (commonly call Anderson insulators). Nonergodic relaxation is particularly interesting for these because the dynamics of electrons has been traditionally considered as very rapid. Such aspects of Anderson insulators were studied to a far lesser degree than their equilibrium properties.

Recently Ben-Chorin *et al.* [3] reported on nonergodic transport in Anderson localized films of indium oxide and ascribed the phenomena to the hopping transport in nonequilibrium states. Here we describe further experiments on this system using a technique that allows monitoring the relaxation for much longer times than hitherto observed. This yields new information on the temporal dependence of the relaxation in such systems. We also show, for the first time, that the dynamics of the relaxation process is distinctly affected by magnetic field H, and discuss the implications of these results.

Our experiments employed a MOSFET-like (metal-oxide-semiconductor field-effect transistor) structure, where the semiconductor was crystalline indium oxide, and a 100 μm thick glass plate was the dielectric separating it from a gate electrode. Full details of sample fabrication are given elsewhere [4]. The film conductance G was measured as a function of gate voltage V_g using a two terminal ac technique. Source-drain voltages were in the Ohmic regime. The high resistance of the samples, and the parasitic capacitance of the measuring wires dictated the use of low frequencies (0.5–10 Hz depending on the resistance) for the phase sensitive measurements. The samples were immersed in a cryostat placed inside a 9 T magnet maintained at $T = 4.11$ K.

Figure 1 shows the change of a sample conductance when V_g is swept from $+100$ to -100 V. Before the sweeps, $V_g = 0$ V (at which the sample was cooled down) was maintained during 12 h to allow equilibration at this V_g at $T = 4.11$ K. Thereafter the experiment was begun (at time $t = 0$) by shifting V_g to 50 V and recording consecutive $G(V_g)$ traces at the indicated times. $G(V_g)$ shows a local minimum at $V_g = 0$ V. Thus, intriguingly, G increases from $G(V_g = 0)$ whether electrons are added or removed from the device. As was explained elsewhere [3], this surprising effect results from two features that are arguably generic to charge transport in the hopping regime: (1) Exciting an Anderson insulator far from thermal equilibrium *enhances* its conductivity which is at a local *minimum* when the system is in thermal equilibrium. (2) The relaxation of the excited electronic system to equilibrium can be a sluggish, nonergodic [2] process. A sudden change *in either direction* of V_g causes a departure from equilibrium, thereby enhancing G. The sluggish relaxation of the system makes this process observable even when V_g is swept quite slowly. Figure 1 reveals a unique feature which earmarks the glassy state of this electronic system: $G(V_g)$ develops a local minimum at any V_g held fixed for an extended time. The new conductance dip at $V_g = 50$ V takes many hours to build up while the original minimum at $V_g = 0$ V takes as long to "heal" (note that the dip at $V_g = 0$ V is discernible even after many hours). An easy to visualize mechanical analog is the deformation of a glass under a durable pressure of a stylus. When shifting the stylus the dip gradually disappears (due to surface tension) while another dip forms under the new stylus position. A scanning probe stylus would produce traces similar to Fig. 1. The dips in the $G(V_g)$ traces reflect residues of the "original" equilibrium and formation of a "new" equilibrium state. The inset in Fig. 1 shows

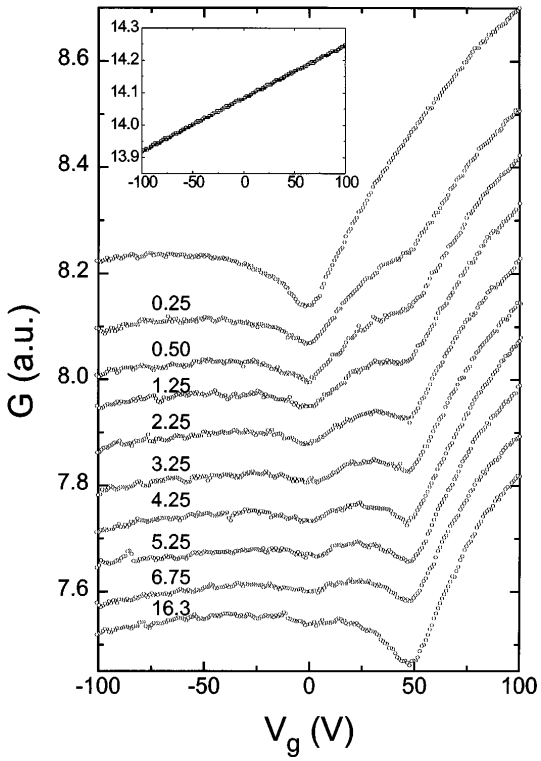

FIG. 1. Conductance versus gate voltage for a typical two-dips experiment. The sample resistance is 26 MΩ. The top trace in the figure shows the initial state with a well developed dip at $V_g = 0$. The other traces (shifted for clarity) were taken at later times, measured from the time a "final" $V_g = 50$ V was imposed between subsequent sweeps. The traces are labeled by the respective time (in hours) that elapsed since the final V_g was applied, and each took 4 min to complete. The inset shows a trace taken identically, approximately 18 h into the run, but after the sample was warmed up to 17 K (all the other data were taken at $T = 4.11$ K). Note that in this case *neither* dip appears.

the field effect of the same sample taken at $T = 17$ K. There $G(V_g)$ exhibits a "normal" field effect indicating that relaxation to equilibrium is faster than the rate of change of V_g. Thus, for this state of disorder, the glass temperature must be somewhere between 4 and 17 K.

We note in passing that similar double-dip behavior was also reported by Adkins *et al.* [5] in a cermet, and by Salvino *et al.* [6] on silicon oxide, but these were attributed to glassy behavior of ions. We believe that such a behavior is characteristic of any electrically active glass.

We have used the "two-dips experiment" to monitor the dynamics of many samples as a fucntion of disorder and magnetic field. We focus here on measurements on three samples [7] with different states of disorder, characterized by the sheet resistance of the sample at $T = 4.11$ K. Figure 2 shows $G(H)$ for the three. The film thickness was 55 Å, and the magnetic field H was oriented parallel to the film plane. Under these conditions, the magnetoresistance of all the samples was positive, presumably due to spin effects [8].

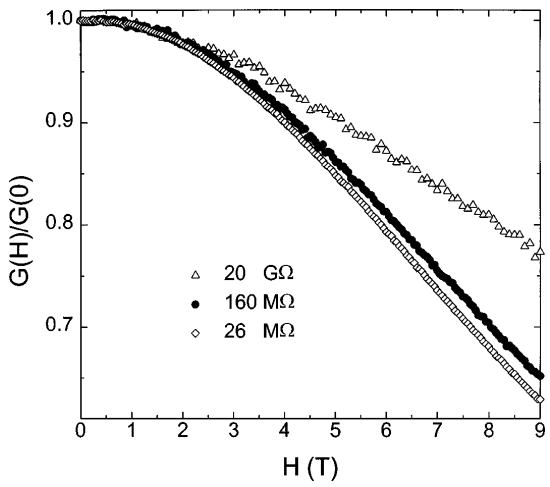

FIG. 2. The normalized conductance versus magnetic field for the three samples studied ($T = 4.11$ K).

For a more systematic study we define a dimensionless parameter $A(t)$ as the ratio of the amplitude of the decaying dip to its value at $t = 0$ (as measured from the "asymptotic" baselines). The behavior of $A(t)$ was found to be fairly insensitive to the conditions of measurements (e.g., a range of sweep rates which was typically 0.1–2 V/sec, the value of the "cool down" V_g). Figure 3 shows $A(t)$ at $H = 0$ for the three measured samples, and Fig. 4 compares $A(t, H = 0)$ with that of $A(t, H = 9\ \mathrm{T})$ for two of these samples. Notice these salient features:

(1) $A(t)$ for the first several hours is always nonexponential and can be described as a power law, $A(t) \propto t^{-\theta}$ with the value of θ varying slowly with t. For $t < 5$ h, and at $H = 0$, θ varied in the range 0.27–0.29 while at $H = 9$ T it was 0.20–0.22 in all the studied samples. For $t < 1$ h θ was considerably smaller, and $A(t)$ plotted well [9] as $\ln(t)$ (i.e., $\theta \to 0$). In some cases we observed that for $t \geq \tau_{\max}$, $A(t)$ crosses over to a faster (possibly

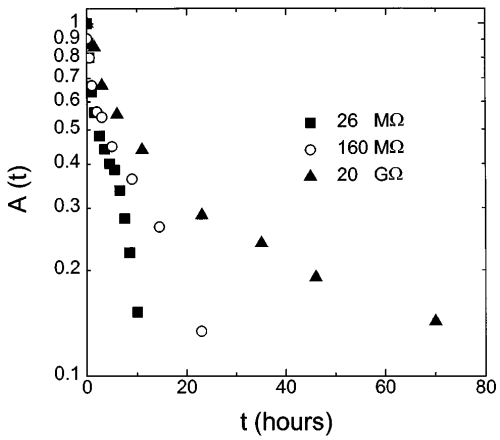

FIG. 3. The behavior of $A(t)$ for the three studied samples. A is defined as $A \equiv \Delta G(t)/\Delta G(0)$ where $\Delta G(t)$ is the dip magnitude at time t measured relative to the natural baseline. The temperature was held constant at $T = 4.11$ K and $H = 0$.

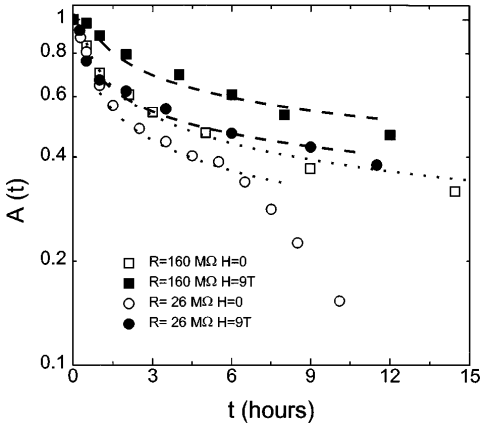

FIG. 4. As in Fig. 3, and comparing between the behavior of A at $H = 0$ and at $H = 9$ T for two samples. (Note that R in the legend is the value of the sample resistance at zero field and is used here just to characterize the disorder.) The dashed and dotted lines are fits of $A(t) \propto t^{-0.21}$ and $A(t) \propto t^{-0.27}$, respectively, to the data.

exponential) time dependence. This $\tau_{\max}$ increases systematically with the sample disorder.

(2) The relaxation rate is affected by the disorder. The more resistive the sample the slower is its relaxation (Fig. 3).

(3) In the presence of $H = 9$ T, the relaxation rate is substantially slower (Fig. 4).

The experimental correlation between the change of conductivity and the change in the relaxation rate due to both disorder and field (as well as the low glass temperature), strongly suggests an electronic mechanism. The approach to equilibrium of the carriers that we imparted an excess energy occurs via quantum-mechanical transitions between localized states—the same types of processes giving rise to conductivity near equilibrium. The constraint of particle conservation dictates that at least two sites must change their state of occupation, a cause for slow relaxation and high resistance. Increasing the disorder reduces the conductivity in two ways—by increasing energy level spacing and by reducing the localization radius. As was originally shown by Kurobe and Kamimura, a strong magnetic field introduces another constraint by impeding certain transitions, thus affecting both the conductivity and the relaxation rate. In their model electrons are localized within a single orbital which cannot accommodate two electrons with parallel spins. When spins are aligned by H, transitions from singly occupied to singly occupied states are blocked, thus reducing the conductivity and the rate of relaxation. In less localized situations the constraint is weaker, but can still exist due to the exchange term [10].

It was argued [2] that in contrast to most glassy systems, in a strongly disordered system even noninteracting electrons can become glassy as a result of particle conservation. In Ref. [3] we showed that the slow relaxation and thus the observed dip of G with V_g can be accounted for by a simple model of localized noninteracting electrons. Here we show that such a model can qualitatively account for the shape of the dip. Current due to hopping transitions between sites i, j separated by an energy E_{ij} is proportional to $[f_i(1 - f_j) + f_j(1 - f_i)]/\sinh^2(E_{ij}/2kT)$. The denominator restricts contributions to current to $|E_{ij}| \leq kT$, and $[f_i(1 - f_j) + f_j(1 - f_i)]$ is virtually zero when i, j are farther than kT from E_F. Holes and electrons excited into that region (as they are with excitation far from equilibrium) enhance $[f_i(1 - f_j) + f_j(1 - f_i)]$ and thus enhance G. On the other hand, for insertion of particles within kT from E_F the change in $[f_i(1 - f_j) + f_j(1 - f_i)]$ is quite small. It follows that when V_g is so small that particles are inserted only within kT of E_F (say, when $V_g < V_0$) [11], the change in G is very small. But, as V_g increases above V_0, G increases rapidly. Saturation is expected when the rate of insertion of nonequilibrium particles equals the rate of particles decaying to within kT from E_F. The regime of V_g is thus usefully divided into two. When $V_g < V_0$, ΔG is expected to remain small. The width of this regime is expected to increase proportionally to T. For $V_g > V_0$, ΔG is expected to rise more rapidly.

Figures 2 and 4 show the effect of H on G and $\Delta G(t)$. H clearly reduces G and slows the relaxation, and thus *qualitatively*, has the same effect as increasing the disorder. But *quantitatively* H and disorder do not affect G and relaxation in the same way. This can be easily seen by comparing the data in Figs. 2, 3, and 4.

One may ask whether there is a simple relationship (as explained below) between the resistance $1/G$ and the relaxation, monitored by $\Delta G(t)$. G can be altered by several experimental parameters say $X, Y, \ldots$ (e.g., H, T, the disorder). Suppose one changes G with X, and, separately, with Y, such that the final G is the same in both cases. Should one expect $\Delta G(t)$ to be also the same? The answer is generally no, because G and $\Delta G(t)$ are determined by transitions among different sets of states. G is determined by transition rates among states confined to lie within about kT of E_F while relaxation involves states farther from E_F. Thus one cannot expect $\Delta G(t)$ to be determined uniquely by G. Such could be reasonably expected only if the change in $f_i(1 - f_j)w_{ij}$ (w_{ij} is the transition rate from i to j) in each pair (i, j) were affected in the same way by X as it is by Y. It is thus understandable that H may have a stronger effect on relaxation than the magnetoresistance suggest, though the degree to which this happens here appears rather large. We shall return to this point later.

So far we did not consider interactions, but these can, of course, be important. Charge interactions can bring about a dip (or a gap) in the one-particle density of states near E_F. This dip has its own characteristic relaxation, which will affect $\Delta G(t)$—the energy gradually decreases as, in response to the interaction potential, the inserted

particle is gradually "dressed" by other particles (thus also reducing its mobility). The dip in the DOS will also affect the dip in $G(V_g)$ since the possibility to insert particles as V_g is varied tracks the DOS.

Spin-spin interactions can also be important, as they generate an effective local magnetic field, and we already mentioned the effect of H on G and on $\Delta G(t)$. Spin interactions thus can affect the relaxation in accordance with the characteristic decay to an equilibrium configuration of the spins. Such interactions can perhaps explain the large effect of H on $\Delta G(t)$.

During the extended relaxation time the ionic system is not likely to remain static. The detailed behavior of the process is then convoluted by the dynamics of the ions. But whether the ions are static or not it should be realized that to account for the results reported above the *electronic* system *must be glassy*. The measured change of G with V_g cannot be attributed to a polarization of ions—note that the field which can excite ions out of equilibrium is *transverse* to the measured current—the symmetry of the problem thus does not allow an interpretation in terms of a depolarization current due to relaxation of ions (and no longitudinal current could be measured due to the application of V_g). Also, if the electrons were not glassy the "memory" effects discussed above could not have existed due to the screening of the internal fields. Our measurements thus lend further experimental evidence in support of the prediction that glassy effects could be due to the internally slow dynamic of electronic systems [12]. The observation of relaxation for many hours is not commonly associated with electron dynamics. Slow electron relaxation, on time scales of seconds, has been observed in capacitance measurements [13].

In summary, we described a set of experiments pertinent to slow relaxation in a disordered electronic system. The results, and, in particular, the dependence of the relaxation rate on disorder and magnetic field, suggest that the origin of the phenomenon is electronic in nature, and apparently involves spin effects.

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[11] The value of V_0 can be estimated from $V_0 \approx e\nu(0)kT/C$ where $\nu(0)$ is the two-dimensional density of states [$\nu(0) = 5 \times 10^{35}$ J^{-1} m^{-2} in the present case] and C the effective capacitance per unit area. Using the measured C this typically gives $V_0 = 2{-}5$ V for $T = 4$ K in good agreement with our experiments.
[12] Motion of ions *may* also occur during these very long relaxation times which is why it is not claimed that the relaxation is *purely* electronic. But, as discussed above, the electronic system *must be glassy* in the first place for these effects to be seen [see also M. Pollak and Z. Ovadyahu, J. Phys. (to be published)].
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