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# Non-Ergodic Dynamics of an Electron Glass

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**Abstract.** — We present the first experimental evidence for the existence of an electron glass. The glassy relaxation is monitored by time dependent conductance measurements following an excitation of electrons to conditions far from equilibrium. At low temperatures the samples exhibit memory-effects observable for hours, as is characteristic of glasses, demonstrating the existence of a glassy *electronic* phase. A simple model, based on the dynamics of non-interacting Anderson-localized electrons, is analyzed to show that dynamic constraints imposed by particle conservation alone can indeed exhibit the observed non-ergodic behavior.

## 1. Introduction

Glasses have for long been known as systems with internal dynamics so slow that they cannot reach thermodynamic equilibrium within a time compatible with our ability to observe. While homogeneity of space makes the equilibrium state of a solid to be a crystal, the non-equilibrium glass does not require (nor does it acquire) such an ordered atomic structure with translational symmetry. Schrödinger [1] was the first to recognize the very interesting capability of such structures lacking translational symmetry for long term memory. The underlying property is the huge number of distinguishable quasi-equilibrium configurations in which they can exist for very long times. Most impressive of Schrödinger's application of this property is his "aperiodic solid" model of genetic memory, borne out by the discovery of the genetic function of DNA. Strangely, this part of Schrödinger's work did not stimulate wide interest in aperiodic solids, or disordered systems as they are now known. The impetus for that came rather later, with the discovery of Anderson localization [2], and Mott's extensive work on the subject [3].

One implication of Anderson localization is that the electrons in disordered systems can themselves become very sluggish in their motion and take extremely long to equilibrate internally, thus making possible a glassy electronic state [4]. This goes rather against intuition: Electrons being very much lighter than atoms, they have been conceived as moving very rapidly, as they indeed do in good crystals. Motion in Anderson-localized systems occurs by transitions between localized states. Localized states cannot be degenerate, so an activation energy is required for transfer of electrons between such states. Moreover, such inelastic transitions

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entail rates proportional to  $\exp[-2r/\xi]$  ( $r$  is the distance between the sites, and  $\xi$  is the radius of the localized function), rather than the rate for elastic transitions which is proportional to  $\exp[-r/\xi]$ . Anderson localization thus reduces, exponentially, the speed of electronic rearrangement in such a medium. Notice that for such a slowed-down process the medium itself must be in a non-equilibrium state, to provide a potential without translational symmetry for the electrons. Another feature peculiar to electrons is that a sluggish relaxation process can happen even when inter-particle interaction is unimportant [4]. Interactions between particles is the accepted cause for inhibiting motion in the glassy state. In localized electronic systems the slowness of transitions between localized states (which conserve particles) may *per se* provide sufficient constraint on the motion. This article presents a simple such model which is compatible with the observed glassy relaxation. The presence of interactions (*e.g.* Coulomb repulsion) puts additional constraints (keeping electrons apart from each other) on their motion, thus enhancing their glassy behavior.

Sluggish response of disordered electronic systems due to Anderson localization has been predicted by several authors [4,5], but clear observation of glassy behavior was rarely claimed [6]. Recently, Ben-Chorin *et al.* [7] reported on non-ergodic transport in Anderson localized films of indium-oxide and ascribed the phenomena to the salient features of hopping transport in non-equilibrium states. It was argued that the microscopic processes leading to the observed slow dynamics are due to electronic transitions and do not involve motion of atoms. In other words, the phenomena observed were ascribed to the system being an electronic glass. This article brings new results of field-effect measurements and introduces a compelling analogy between this electron-glass and conventional glasses.

## 2. Experimental

The experiments employed a MOSFET-like structure, the indium oxide film serving as the semiconductor, and a 100  $\mu\text{m}$  thick glass plate as the dielectric separating it from a gate electrode. The device is shown schematically in the insert of Figure 1, and described in detail in reference [7]. Films were prepared by evaporating 99.999% pure  $\text{In}_2\text{O}_3$  onto a glass substrate at a rate of 0.2–0.7  $\text{\AA}/\text{s}$  in an atmosphere of pure  $\text{O}_2$  at a pressure of  $2 \times 10^{-5}$  mbar. Conductivity was measured using a two terminal ac technique. Source-drain voltages were of the order of 100 mV rms, well in the ohmic regime. The high resistance of the samples (up to 300 G $\Omega$ ), and the parasitic capacitance of the measuring wires dictated the use of low frequencies (1-10 Hz) for the phase sensitive measurements thus limiting the temporal response of the measurements to 3-30 seconds. The longest experimental time, dictated by instrumental stability, was  $\sim 4$  hours.

In some of the relaxation measurements the system was excited optically using a 6300  $\text{\AA}$  LED, installed on the probe near the sample. To facilitate uninterrupted long-time scans the samples were immersed in a 100-liter, liquid-He storage-dewar at  $T = 4.11$  K.

## 3. Results and Preliminary Discussion

Figure 1 shows typical results of the change of conductance due to sweeps of the gate voltage  $V_g$ .  $V_g = -80$  V was maintained during cooling to 4 K, and for 12 hours after to allow the sample to equilibrate and to establish a baseline (determined by instrumental stability). Thereafter, consecutive conductance *versus* gate-voltage traces were recorded, and labeled by the time elapsed since the experiment began. Each trace took about 2 minutes to complete.  $V_g$  was held at 0 between successive sweeps. The traces exhibit an intriguing effect: The conductance *versus*  $V_g$  shows a local minimum at the 'cool-down- $V_g$ ' of  $-80$  V. Contrary to intuition, around this  $V_g$  the conductance increases, no matter whether electrons are added or

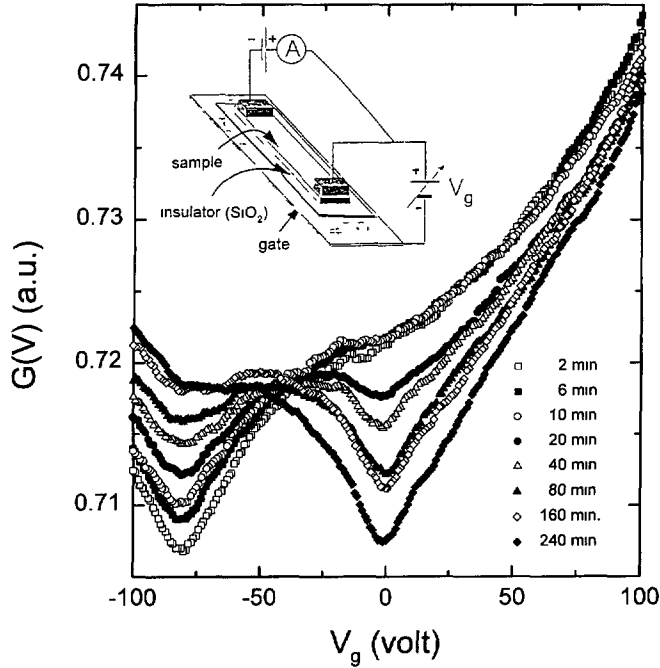


Fig. 1. — The field effect for a 68 Å film with  $R_{\square} = 75 \text{ M}\Omega$  where, initially,  $V_g \approx -80 \text{ V}$  was kept for 12 hours. At  $t \equiv 0$ ,  $V_g = 0$  was applied and maintained during the intervals between subsequent sweeps. The insert illustrates the sample geometry.  $T = 4.11 \text{ K}$ .

removed from the device. As was explained elsewhere [7], this surprising effect results from two features that, arguably, are generic to charge transport in the hopping regime:

- exciting an Anderson insulator from thermal equilibrium *enhances* its conductivity - the conductivity is at a *minimum* when the system is in thermal equilibrium,
- the relaxation of the excited electronic system to equilibrium is a sluggish, non-ergodic process.

A sudden change in *either direction* of  $V_g$  causes a departure from equilibrium, thereby enhancing its conductance. The sluggish relaxation of the system makes this process observable even when  $V_g$  is swept relatively slowly. Figure 1 reveals a unique feature which marks 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  $V_g = 0$  requires many hours to form while the original minimum at  $V_g = -80$  takes as long to “heal” (note that the dip at  $V_g = -80 \text{ V}$  is visible even after four hours). This behavior resembles the slow mechanical deformation in 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 Figure 1. We comment that the above vague terminology regarding time duration reflects the observed logarithmic time dependence (*viz.* Fig. 2) which shows *no* characteristic time for the duration of the experiment. The only measure of the speed of relaxation is how much the conductance decays in, say, a decade of time. The temporal dependence of the conductance following an excitation is illustrated

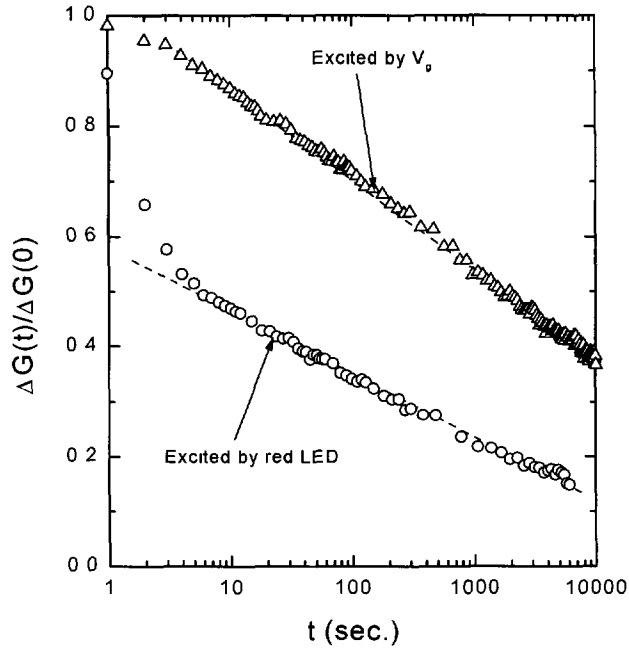


Fig. 2. — Relaxation of the normalized excess-conductance of the same film as in Figure 1, following different excitations. Upper curve: excitation is achieved by a voltage step of 100 V at the gate. Lower curve: excitation is by a 10 seconds exposure to a red LED. The initial  $\Delta G(0)/G$  for both excitations is  $\sim 1\%$   $T = 4.11$  K.

in Figure 2. It shows the relaxation of the excess conductance  $\Delta G(t)$ , created by a voltage step on the gate or, alternatively, by illumination. The voltage step from  $V_g = -80$  V  $\equiv V_0$  to  $V_\infty = 0$  is applied at  $t = t^0$  and maintained thereafter. For excitation by light the source is turned on, its intensity adjusted until  $\Delta G$  equals  $\Delta G(t^0)$  of the gate experiment, and it is then *turned off*. The gate experiment shows that  $\Delta G(t)/\Delta G(0)$  decays as  $\log(t)$ , where  $\Delta G(t) = G(t) - G_\infty$ ,  $G_\infty$  is the asymptotic value  $G(t = \infty)$  and  $\Delta G(0)$  is the change of conductance measured at  $t = t^0$  (by “0” in these log plots is meant our smallest resolvable time, typically 3-30 s, *cf.* Sect. 2 above). This peculiar time dependence has been observed in all our  $V_g$  excitation experiments and may thus be taken as characteristic of the studied electronic system. The lack of time-scale inherent to a  $\log(t)$  dependence complicates comparison of time-domain processes.

The excess conductance  $\Delta G(t)/\Delta G(0)$  induced by light also follows a  $\log(t)$  for  $t > 10$  s, but is qualitatively different dependence at early times (*cf.* Fig. 2). An accurate determination of the decay-law at  $t < 10$  s is impossible due to the already mentioned limited time resolution for such intervals. Nevertheless, extrapolation of  $\Delta G(t > 10 \text{ s})/\Delta G(0)$  to early times shows clearly that initial relaxation is *faster*. We suspect that the difference in the initial relaxation is due to different initial non-equilibrium states generated by the two types of excitations. Specifically, the range of electron excitation energies  $\Delta E$  is larger for optical excitation. In the field experiment  $\Delta E \approx 10\text{--}10^2$  K while in the optical experiment  $\Delta E$  is much larger. Since electronic states at higher energies tend to be less localized, the transition rates are larger, so relaxation is expected to be faster, as is observed. After the highly excited electrons decay to levels comparable with the initial conditions in the gate experiment, the relaxation in the two experiments should be similar, again as observed.

Finally we note that applying a strong magnetic field decreases  $G$  and, at the same time slows down the relaxation.

#### 4. Quantitative Considerations

This section attempts to show that the simplest model of a system of non-interacting Anderson localized electrons can account for the glassy behavior observed in our experiments. The basic assumption of the model involves s-like electronic states with a uniform localization radius  $\xi$  and a constant (one-particle) density of states  $N(E) = N(E_F)$ . Given the simplicity of the model, we cannot expect a precise theoretical prediction for the observed  $\Delta G(t)$ , but show that the observed duration of relaxation and the magnitude of the excess conductance as well as their dependence on disorder are compatible with the observations.

**4.1. CALCULATION OF  $\Delta G(0)$ , THE INITIAL EXCESS CONDUCTANCE.** — We first calculate the initial excess current density  $\delta j$ . It is calculable as current through a unit area of a plane, defined by a unit vector  $\sigma$  parallel to the field  $\mathbf{F}$ . For a random distribution of sites the choice of the plane position is arbitrary because the relevant probabilities do not depend on the position of the plane.

Suppose there are  $N$  electrons/cm<sup>3</sup> and charge transport is by hopping. The contribution to  $j$  from hops of length  $r$  traversing the plane is  $eNP(r)dr(W_R - W_L)r \cdot \sigma$  where  $W_R$ , ( $W_L$ ) are hopping rates to the “right” (“left”) and  $P(r)dr$  is the probability that an electron can find a suitable site at a distance  $r \pm dr$ . The difference in the rates  $W_R$  and  $W_L$  is due to  $\mathbf{F}$ .

At  $t = 0$ ,  $N(0)$  electrons per unit volume are assumed excited into sites above the Fermi energy (by more than  $k_B T$ ), and uniformly distributed in space. They are most likely to hop into other sites above the Fermi energy, where the probability that they are empty is nearly 1. Only those electrons whose final site energy differ by no more than  $k_B T$  from the initial site contribute to the current (see Eq. (1)). The difference  $W_R - W_L$ , for electrons hopping into a site a distance  $r$  from the initial site is [8]:

$$W_R - W_L = \frac{\nu_{ph}(eFr/k_B T)\exp(-2r/\xi)}{\sinh^2(E/2k_B T)} \quad (1)$$

where  $E$  is the energy gain or loss of the electron and  $\nu_{ph}$  is a phonon frequency. The distribution of sites with a distance  $r$  from the initial site and with  $E < k_B T$  is  $4\pi N(E)k_B T r^2$ , so the excess current density at  $t = 0$  is:

$$j = eN(0) \int dr dE \nu_{ph} N(E) \left( \frac{eF}{k_B T} \right) r^4 \frac{\exp(-2r/\xi)}{\sinh^2(E/2k_B T)} \approx \left( \frac{6}{3 \times 32} \right) e^2 \nu_{ph} N(0) N(E) F \xi^2. \quad (2)$$

The incremental conductivity  $\Delta g$  due to the excited electrons is  $\delta j/F$ . Using  $l$  for length (between ‘source’ and ‘drain’),  $w$  for width,  $t$  for thickness, and  $N_{tot} = N(0)lwt$  for the total number of excited electrons, the incremental conductance is:

$$\Delta G = \left( \frac{\delta j}{F} \right) \frac{wt}{l} \approx \frac{1}{16} e^2 \nu_{ph} N_{tot} N(E) \frac{\xi^5}{l^2}. \quad (3)$$

With quantities typical of our experiments,  $\xi = 10 \text{ \AA}$ ,  $N(E) = 10^{20} \text{ cm}^{-3}(\text{eV})^{-1}$ ,  $l = 0.5 \text{ cm}$ ,  $N_{tot} = 10^{12}$ ,  $\nu_{ph} = 10^{12} \text{ s}^{-1}$ , one gets  $\Delta G \approx 10^{-11} \Omega^{-1}$ . The value  $N = 10^{12}$  corresponds to  $\Delta V_g = 100 \text{ V}$  applied to the plate-capacitance composed of the sample, the glass plate and the gate electrode. The calculated effective  $\Delta E$  for this case is a few mV.

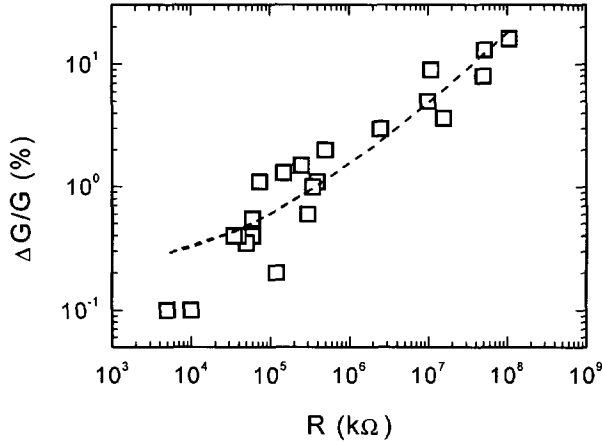


Fig. 3. — The fractional change of conductance,  $\Delta G/G$ , as a function of the films sheet resistance,  $R_{\square}$ , at  $T = 4.11$  K for a  $\Delta V_g = 100$  V excitation. The dashed line depicts the theoretical dependence expected by equation (5). The range of resistances shown is spanned by localization-lengths varying between  $\xi = 18$  Å to  $\xi = 6.5$  Å.

The magnitude of excess conductance,  $\Delta G$ , does not depend on temperature according to equation (3). It should be borne in mind, however, that the existence of these effects hinges on the validity of the condition  $\Delta E > k_B T$ .

The small thickness renders the films to be in the 2-dimensional regime of the Mott law [8]

$$G = G_0 \exp \left[ -\frac{T_0}{T} \right]^{1/3} \quad \text{with} \quad T_0 = \frac{3}{N(E)k_B t \xi^2}. \quad (4)$$

The exponential dependence  $G(T)$  mandates very careful stabilization of the temperature; otherwise the true  $\Delta G(t)$  is masked by thermally induced fluctuations. Combining equations (3, 4) one gets:

$$\frac{\Delta G}{G} \propto \frac{R}{(\text{Log}(R))^{15/2}} \quad (5)$$

where  $R \equiv 1/G$  is the film resistance. Crude as the estimate is, it compares favorably with our results, as shown in Figure 3.

**4.2. CONDITIONS FOR OBSERVATION OF GLASSY BEHAVIOR.** — For sufficiently strong disorder (films with  $\xi < 15$  Å), relaxation is observed throughout the duration of the experiments, of the order of  $10^3$  s at  $T = 4$  K. A central point of the paper is to show that our simple model is compatible with such relaxation times. The time needed for equilibration, as observed by measurement of  $\Delta G(t)$ , can be estimated by calculating the rate of hopping through the critical link in the percolation network. It is emphasized that this is a *minimal* time for relaxation, based on an assumption that the last stages of relaxation occur from near equilibrium. It is possible for carriers at high-energy sites not connected to the percolation cluster with any  $\zeta \leq \zeta_c$  or, in cases of initial spatial inhomogeneity, to require many successive critical steps to reach an equilibrium position. If such conditions apply to a significant number of carriers, the relaxation process can be observable for times longer than the estimate based on a critical link. The calculation below is therefore a lower limit for the observable relaxation time.

The rate of hopping through a link in the percolation network is given by  $\tau' = RC$ , where  $R$  is the Miller-Abrahams [9] resistance for that link,  $R = (k_B T / e^2 \nu_{ph}) \exp(2r/\xi) \exp(\varepsilon/k_B T)$ , and  $C$  the random network capacitance  $C = e^2 / kT \exp(-\varepsilon/k_B T)$  ( $\varepsilon$  is the higher of the two site energies of the link). Thus,  $\tau' = \nu_{ph}^{-1} \exp(2r/\xi)$  and  $\tau = \nu_{ph}^{-1} \exp(\zeta_c)$  where  $\zeta_c = C_{2D} [N(E) \xi^2 t k_B T]^{-1/3}$  using the two-dimensional version of the percolation model. With the values used before for  $N(E)$ ,  $T = 4$  K. and [10]  $C_{2D} = 3.5$ , one obtains:  $\tau = 10^6$  s for  $\xi = 10$  Å,  $\tau = 700$  s for  $\xi = 15$  Å, and  $\tau = 10^{-1}$  s when  $\xi = 20$  Å. Thus, in this range of sample parameters and temperatures, the glass transition occurs somewhere between  $\xi = 10$  Å to  $\xi = 15$  Å.

## 5. Summary

The above considerations and their agreement with experimental observations support the contention that an electronic glass is an observable entity in particular in view of the observed response to a magnetic field. Some problems require further elucidation. First and foremost, it is the detailed form of the temporal dependence of the excess conductance,  $\Delta G(t)$ . The present set of experiments established that  $\Delta G(t)$  follows consistently a  $\log(t)$  law when  $\Delta E$  is not too large. Most models that purport to account for non-exponential relaxation phenomena yield a 'stretched-exponent' law [11] or a power law which do not give a satisfactory agreement with our data.

Another problem to pursue is the possible role of electron-electron interactions. Although the above analysis shows that interactions are not required, it cannot rule out their possible contribution. But additional experiments indicate that *long-range* Coulomb interactions are not involved. In these experiments we compared  $\Delta G(t)$  of several pairs of samples. The members of a pair were made identically, except that a metal film was put in proximity (separated by a 60–100 Å  $\text{SiO}_2$  layer) to the sample. The pair was excited to the same initial  $\Delta G$ . The ensuing  $\Delta G(t)$  of the 'screened' *versus* that of the 'unscreened' differed, but only in the initial, fast relaxation part of  $\Delta G(t)$  (*cf.* Sect. 3). The asymptotic  $\log(t)$  relaxation extended over the *same time-period for both members of the pair*, showing that slow relaxation does not hinge on long-range Coulomb interactions.

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