

Transition to exponential relaxation in weakly disordered electron glasses

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The out-of-equilibrium excess conductance of electron-glasses $\Delta G(t)$ typically relaxes with a logarithmic time dependence. Here it is shown that the $\log(t)$ relaxation of a weakly disordered In_xO film crosses over asymptotically to an exponential dependence $\Delta G(t) \propto \exp\{-[t/\tau(\infty)]\}$. This allows for assigning a well-defined relaxation-time $\tau(\infty)$ for a given system disorder (characterized by the Ioffe-Regel parameter $k_F\ell$). Near the metal-insulator transition, $\tau(\infty)$ obeys the scaling relation $\tau(\infty) \propto [(k_F\ell)_C - k_F\ell]$ with the same critical disorder $(k_F\ell)_C$ where the zero-temperature conductivity of this system vanishes. The latter defines the position of the disorder-driven metal-to-insulator transition which is a quantum-phase transition. In this regard the electron glass differs from classical glasses, such as the structural glass and spin glass. The ability to experimentally assign an unambiguous relaxation time allows us to demonstrate the steep dependence of the electron-glass dynamics on carrier concentration.

DOI: [10.1103/PhysRevB.97.214201](https://doi.org/10.1103/PhysRevB.97.214201)**I. INTRODUCTION**

Slow relaxation is a widespread phenomenon manifested in a variety of physical systems. A temporal law exhibited in many such instances is the stretched exponential (SE) where a measurable M relaxes with time t as $M \propto \exp[-(t/\tau)^\beta]$ with $0 < \beta < 1$ and τ as a typical relaxation time. Several approaches were offered to account for the physical origin of this time dependence [1–6]. Phenomenologically, however, the SE may be ascribed to a distribution of relaxation-times $P(\tau)$ that, in turn, determines the value of β and τ .

Another form of relaxation that is ubiquitous in disordered systems is a logarithmic law $M \propto -\log(t)$ [7–12,17]. Technically, a logarithmic time dependence may be regarded as a special case of the stretched-exponential law (when $\beta \rightarrow 0$). A $\log(t)$ relaxation conforms to an extended range of relaxation-times $P(\tau)$ whereas $\beta = 1$ is the case for a narrow distribution [4].

Demonstrating experimentally a $\log(t)$ dependence is more demanding than a stretched exponential in that one has no freedom in fitting; no parameter is involved in verifying a logarithmic form whereas three parameters are typically used to fit a SE to measured data. A $\log(t)$ relaxation is also special in being limited to intermediate times; it ought to cross over to a different form for both short and long times [7].

A $\log(t)$ relaxation has been claimed in a number of diversified phenomena; structural recovery from a heat shock [7], autocorrelations in spin glass [8], magnetization relaxation [9], relaxation of levitation force [10], exchange kinetics in polymers [11], and dynamics of glass-forming systems [12]. In some experiments the logarithmic relaxation was monitored for more than five decades [13], but the expected transition to the asymptotic behavior has so far escaped detection. The main reasons for the difficulty in reaching this limit is that the magnitude of the relaxed part of the observable is usually small to begin with, and most of it dissipates in the early stage of the relaxation. The finite signal-to-noise ratio and drift of instruments make it hard to identify the expected asymptotic

behavior when the $P(\tau)$ distribution has components that extend over many hours.

The experiments presented in this paper demonstrate that the transition from logarithmic to exponential relaxation may consistently be observed in electron glasses with short relaxation times. These observations were made on Anderson-insulating amorphous indium oxide In_xO films with the lowest carrier-concentration N yet reported to exhibit intrinsic electron-glass features. Data for the exponential-relaxation regime allow us to uniquely determine the relaxation-time $\tau(\infty)$ that presumably represents the high end of $P(\tau)$. Using samples with different degrees of disorder it is shown that $\tau(\infty)$ appears to vanish at the metal-insulator critical point while obeying a scaling relation characteristic of a quantum-phase transition.

II. EXPERIMENT**A. Sample preparation and characterization**

Samples used in this paper were thin 200-Å-thick films of In_xO . These were made by e -gun evaporation of 99.999%-pure In_2O_3 onto room-temperature Si wafers in a partial pressure of 3×10^{-4} mbars of O_2 and a rate of 0.3 ± 0.1 Å/s. The Si wafers (boron doped with bulk resistivity $\rho \leq 2 \times 10^{-3}$ Ω cm) were employed as the gate electrode in the field-effect and gate-excitation experiments. The samples were deposited on a SiO_2 layer (2-μm thick) that was thermally grown on these wafers and acted as the spacer between the sample and the conducting Si:B substrate.

The as-deposited films had sheet-resistances of $R_\square > 500$ MΩ at room temperature. They were then thermally treated. This was performed by stages; the samples were held at a constant temperature T_a starting from $T_a \approx 320$ for 20–30 h then T_a was raised by 5–10 K at the next stage. This was repeated until the desired R_\square was attained (see Ref. [14] for full details of the thermal-annealing process). This process yielded samples with $R_\square = 20$ –40 kΩ that at $T \approx 4$ K spanned the

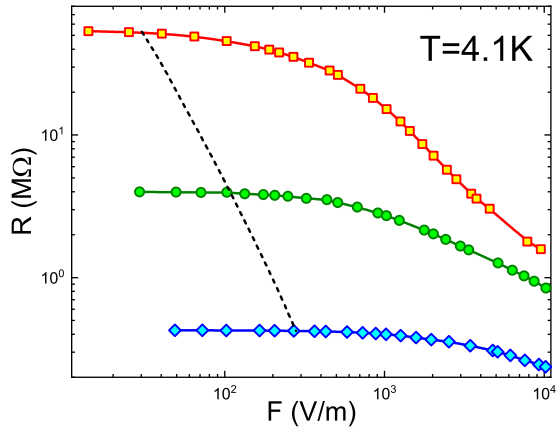


FIG. 1. The dependence of sample resistance on the applied field for three typical In_xO films, all with identical dimensions of 1×1 mm. The dashed line connects points on each $R(F)$ curve that their deviation from ohmic behavior exceeds the experimental error.

range of 100 k Ω to 90 M Ω . A maximum $T_a \approx 355$ K and 14 days of treatment were required to get the lowest resistance used in the paper. The carrier-concentration N of these samples, measured by the Hall effect at room temperatures, was in the range of $N \approx (8.5 \pm 0.3) \times 10^{18} \text{ cm}^{-3}$.

The motivation for choosing a low- N version of In_xO for these experiments was the observation that the electron-glass dynamics becomes faster as their carrier concentration falls below $N \lesssim 4 \times 10^{19} \text{ cm}^{-3}$ [15,16] whereas, all other things being equal, their excess conductance ΔG is more conspicuous [14]. These expectations were borne out in our experiments which made it possible to quantify the system dynamics as it approaches the quantum-phase transition.

B. Measurement techniques

Conductivity of the samples was measured using a two-terminal ac technique employing a 1211-ITHACO current preamplifier and a PAR-124A lock-in amplifier. Measurements were performed with the samples immersed in liquid helium at $T \approx 4.1$ K held by a 100-l storage dewar. This allowed up to 2-month measurements on a given sample while keeping it cold. These conditions are essential for measurements where extended times of relaxation processes are required at a constant temperature especially when running multiple excitation-relaxation experiments on a given sample.

The gate-sample voltage (referred to as V_g in this paper) in the field-effect measurements was controlled by the potential difference across a 10- μF capacitor charged with a constant current. The rate of change in V_g is determined by the value of this current. The range of V_g used in this paper reached in some cases ± 60 V which is equivalent to the ± 15 V used in previous studies [17] where the gate-sample separation was 0.5 μm as compared with the 2- μm SiO_2 spacer used here.

The ac voltage bias in conductivity measurements was small enough to ensure near-ohmic conditions. The voltage used in the relaxation experiments was checked to be in the linear-response regime by plotting the current-voltage characteristics of each sample. Figure 1 illustrates the dependence of the conductance on the applied field for typical samples.

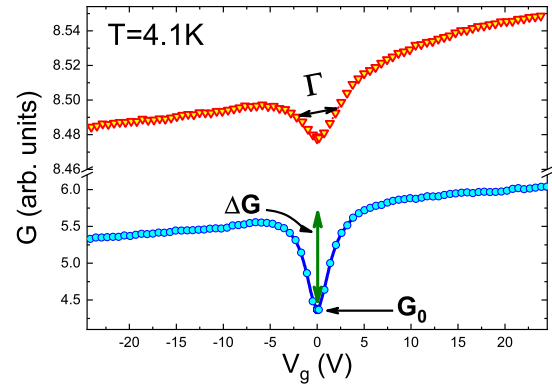


FIG. 2. Conductance versus gate voltage for two of the In_xO samples used in the paper. The data for $G(V_g)$ were taken with a sweep rate of 0.5 s for both samples. The typical width of the memory-dip Γ and the definition of ΔG and G_0 are marked with arrows. The memory-dip relative magnitudes $\Delta G/G_0$ are $\approx 0.3\%$ and $\approx 30\%$ for the samples with $R_{\square} = 105 \text{ k}\Omega$ and 45 M Ω , respectively.

C. Results and discussion

Figure 2 shows the dependence of the conductance G on gate-voltage V_g for two of the studied In_xO samples. Each $G(V_g)$ plot shows two main features; an asymmetric component characterized by $\partial G(V_g)/\partial V_g > 0$ in the entire range of -50 to $+50$ V, that reflects the increased thermodynamic density of states with energy (the thermodynamic field effect), and a cusplike dip centered at $V_g = 0$ where the system was allowed to relax before sweeping the gate voltage (the memory dip). Note that $\Delta G/G_0$ for the 45-M Ω sample is $\approx 30\%$ —the highest value ever reported for any electron glass with comparable R_{\square} at $T \approx 4$ K and measured with a nominal sweep rate in the range of (0.3–1) V/s. Similarly, the 105-k Ω sample has $\Delta G/G_0 \approx 0.3\%$, still large enough for the memory dip to stand out despite the sloping thermodynamic field effect. Note that the width Γ of the memory dip of these samples is quite narrow consistent with their low N [15]. Samples with different degrees of disorder, using this batch of films, were used to monitor how, following identical excitation and measurement conditions, the system approaches its equilibrium state.

An effective and reproducible way to excite the system is the gate protocol in which the excitation is affected by switching the gate-voltage V_g from its equilibrium value of V_{eq} to a new one of V_n . This takes the system out of equilibrium, and this is reflected in the appearance of a time-dependent excess-conductance $\Delta G(t)$. The minimum time the sample was equilibrated under V_{eq} in this paper was 12 h. In all the experiments described here the total gate-voltage swing $|V_n - V_{\text{eq}}|$ was much larger than the memory-dip width Γ defined in Fig. 2. The relaxation to the equilibrium under the newly established V_n was monitored through the measured $\Delta G(t)$. In previously studied electron glasses, this invariably led to $\Delta G(t) \propto -\log(t)$ persisting over three or more decades. The same is true for the relaxation law observed in other experiments where logarithmic relaxation was observed [7,9,10,12]. In the electron glasses used here, however, the $\log(t)$ dependence was only observed for the first two decades of the relaxation. A clear deviation from a logarithmic law

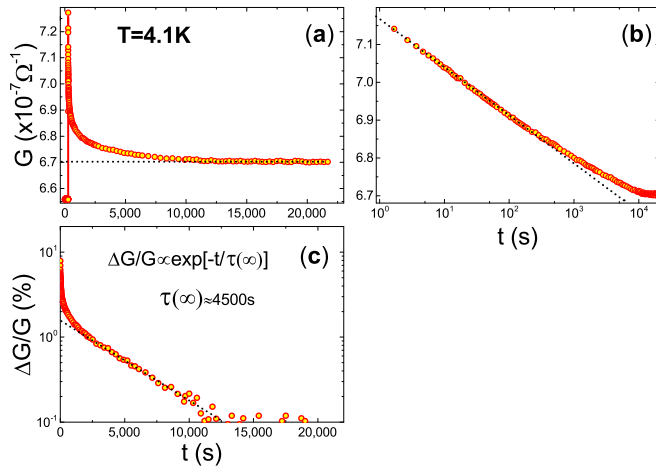


FIG. 3. Results of using the gate protocol (see the text) on a sample with $R_{\square} = 1.5 \text{ M}\Omega$. (a) Conductance as a function of time; after $\approx 50 \text{ s}$ of monitoring G under $V_g = 0 \text{ V}$ the gate voltage was swept to $V_g = 20 \text{ V}$ at a rate of 10 V/s . The dashed line is $G(\infty)$, the asymptotic value of the conductance. $G(\infty)$ differs from $G(V_{\text{eq}})$ due to the component of the thermodynamic field effect. (b) Conductance relaxation starting from the time $V_g = 20 \text{ V}$ was established showing the extent of the $\log(t)$ dependence (delineated by the dashed line). (c) The plot of $G(t) - G(\infty)$ demonstrating an exponential relaxation behavior (dashed line is the best fit).

to another time dependence was consistently observed in all samples of this and three other prepared batches of In_xO . Figure 3 shows the results of the gate protocol for an insulating sample with $R_{\square} = 1.5 \text{ M}\Omega$. As the figure illustrates, the excess conductance initially follows $\Delta G(t) \propto -\log(t)$ for a time period lasting up to $\approx 250 \text{ s}$, [Fig. 3(b)], then ΔG deviates from the logarithmic law, showing a tendency for the conductance to saturate. It turns out that for $t \gtrsim 1500 \text{ s}$ the excess conductance $\Delta G(t)$ for this sample may be well fitted to a stretched-exponential dependence with $\beta \approx 0.95 \pm 0.1$ [Fig. 3(c)]. Within the experimental error this β is close enough to unity. Accordingly, we fit the data in this regime assuming $\beta \equiv 1$, that is, $\Delta G(t) \propto \exp[-t/\tau]$. It seems that, for these samples, the low end of the rate distribution is narrow enough to make it impossible to distinguish β from unity.

Qualitatively similar results were obtained for the gate protocol applied to a $105\text{-k}\Omega$ sample, the sample with the lowest degree of disorder that was used for this purpose. These results are shown in Fig. 4: The figure shows again the limited range over which the excess conductance follows a $\log(t)$ dependence and the transition to an exponential-relaxation regime at later times. Apart from the lower signal-to-noise ratio (due to the smaller $\Delta G/G_0$ associated with the lower R_{\square} sample), the main difference between the data in Figs. 4 and 3 is in the relaxation-time $\tau(\infty)$. This time turns out to be independent of the particular values of V_{eq} and V_n as long as $|V_n - V_{\text{eq}}|$ is larger than the width of the memory-dip Γ . This aspect was tested in the sample of Fig. 4 using eight different runs of the gate protocol with $20 \leq |V_n - V_{\text{eq}}| \leq 90 \text{ V}$ and in seven runs for a sample with $R_{\square} = 120 \text{ k}\Omega$ using voltage swings in the range of $40\text{--}80 \text{ V}$. In either case, the same behavior of $G(t)$ and the same value of $\tau(\infty)$ were obtained.

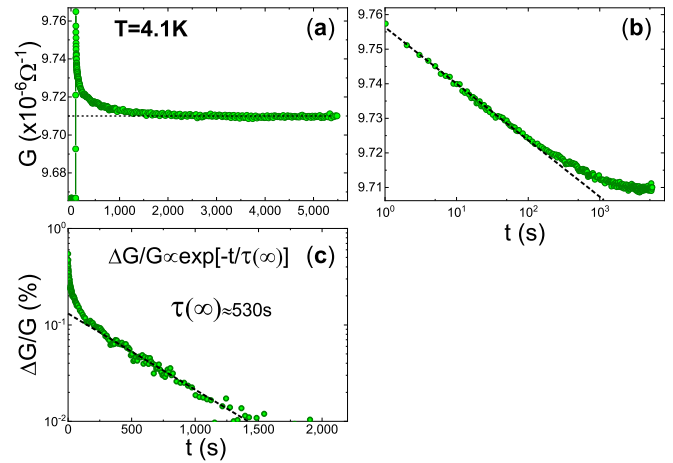


FIG. 4. The same as in Fig. 3 but for a sample with $R_{\square} = 105 \text{ k}\Omega$. Note the difference in the time scale as compared with Fig. 2 and the constant $G(t)$ for $t \gtrsim 2000 \text{ s}$.

Note that it is the relatively short equilibration time of these samples that makes these repeated measurements on the given sample practical (in addition to the benefit of short relaxation times in minimizing the problem of the instruments' drift).

In the range of the equilibration times used in this paper, the relaxation-time $\tau(\infty)$ is also independent of the time the sample was held under V_{eq} before switching to V_n . Figure 5 compares the results of the gate protocol for two different times the system was allowed to equilibrate under V_{eq} .

On the other hand, the relaxation-time $\tau(\infty)$ does depend on R_{\square} as it may already have been noticed by comparing the time scales in Figs. 3 and 4. In fact, the relaxation time of these electron glasses decreases systematically with their resistance and, as will be shown below, eventually vanishes when the sample resistance is sufficiently small.

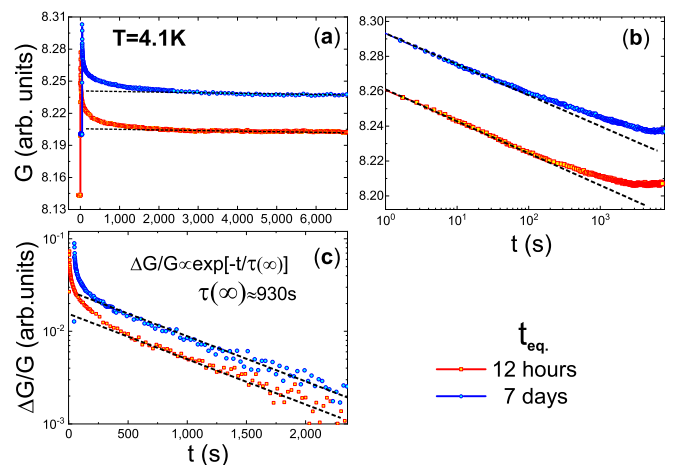


FIG. 5. Comparing the results of the gate protocol (with $V_{\text{eq}} = 0$ and $V_n = 46 \text{ V}$) for a sample with $R_{\square} = 120 \text{ k}\Omega$ using two different equilibration times t_{eq} . (a) $G(t)$ data for the entire duration of the protocol with $V_{\text{eq}} = 0$, $V_n = 46 \text{ V}$, and sweep rate of 10 V/s . The curves are displaced for clarity. (b) $G(t)$ for the relaxation. The origin of time is taken as the instance when V_g reached V_n . Note the similar appearance of the curves (dashed lines are the respective best fits).

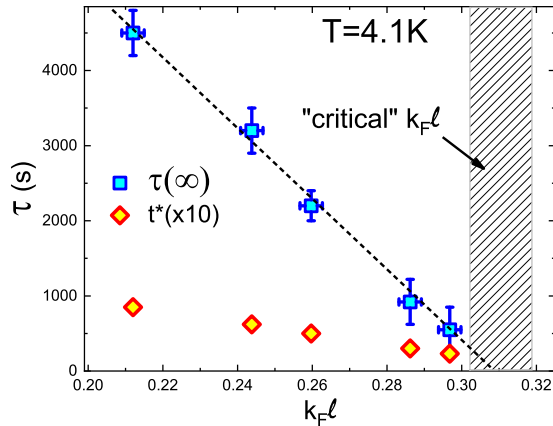


FIG. 6. The dependence of the relaxation-times $\tau(\infty)$ (defined in Figs. 3–5) and t^* (see the definition in the text) on the disorder parameter $k_F \ell$ near the critical regime of the metal-to-insulator transition (marked by the hatched area).

This trend has an implication on the origin of the slow relaxation in these systems. The reduction of $\tau(\infty)$ with R_{\square} is achieved in In_xO by thermal annealing. The changes in the structural properties of the material during the annealing process were extensively studied in Ref. [18] by electron diffraction, energy-dispersive spectroscopy, x-ray interferometry, and optical techniques. These studies revealed that the change in the resistance from the as-deposited deeply insulating state all the way to the metallic regime is mainly due to an increase in the material *density*. In particular, the samples retained their amorphous structure and composition throughout the entire process [18]. Moreover, the dynamics associated with structural changes monitored during annealing and recovery of the samples was qualitatively different than that of the electron glass and did not change its character throughout the entire range of disorder [16]. The diminishment of $\tau(\infty)$ with R_{\square} cannot then be identified with the elimination of some peculiar structural defects that were hypothesized as being responsible for the slow relaxation and other associated nonequilibrium transport phenomena in electron glasses [19].

On the other hand, as all the samples studied here had essentially the same N , the difference in their relaxation times with their different resistances may hint on the role of *quenched* disorder. This motivated us to measure $\tau(\infty)$ in films of the same preparation batch that underwent different degrees of thermal annealing to fine-tune their disorder.

A dimensionless measure of disorder that is often used in metal-insulator and superconducting-insulator studies is the Ioffe-Regel parameter $k_F \ell$. This may be estimated by $k_F \ell = (3\pi^2)^{2/3} \hbar \sigma_{\text{RT}} e^{-2} N^{-1/3}$ where σ_{RT} is the sample conductivity at room temperature. We use this measure to plot in Fig. 6 the dependence of $\tau(\infty)$ on disorder. The figure shows that $\tau(\infty)$ appears to vanish at (or near) a specific $(k_F \ell)_C$. Interestingly, this point in disorder coincides with the metal-insulator transition (MIT) for In_xO determined [20] in the same version of In_xO used in the current paper (in terms of having a similar carrier-concentration N).

The diminishment of the relaxation time as the metallic phase is approached from the insulating phase is in line with the

notion that glassy behavior is an inherent feature of Anderson insulators [21–31].

The dynamics at the metal-insulator critical point (defined by disorder where the dc conductivity vanishes as $T \rightarrow 0$) is likely slower than deeper into the metallic phase; sections of the sample may still be insulating, and their slow dynamics could influence the transport in the just-percolating diffusive channel [32,33]. Glassy effects may then persist some distance into the diffusive phase, but their magnitudes will be small, and the observed dynamics will speed up sharply as $k_F \ell$ increases past the critical disorder.

The trend for faster dynamics as $k_F \ell \rightarrow (k_F \ell)_C$ is also reflected during the $\log(t)$ relaxation regime. This may be quantified by choosing (arbitrarily) a time-scale t^* defined through $2G(t^*) = G(1) - G(\infty)$. This yielded t^* of 23–85 s for the samples with $k_F \ell$ in the range of 0.29–0.22, respectively. The dependence of t^* on disorder is shown in Fig. 6.

The scaling relation $\tau(\infty) \propto |k_F \ell - (k_F \ell)_C|^\nu$ with $\nu \approx 1$ obeyed by the relaxation time (Fig. 6) is essentially the same as the scaling behavior of the zero-temperature conductance $\sigma(0) \propto |k_F \ell - (k_F \ell)_C|^\nu$ reported in Ref. [20] for the MIT of this material. The transition to the electron-glass phase thus emerges as a *quantum-phase transition* rather than following the scenario conceived by Davies *et al.* [21]. These authors anticipated a glass temperature by analogy with the spin-glass problem [21]. Instead, the experiments suggest that the glassy effects are just finite-temperature manifestations of a zero-temperature disorder-driven transition in the same vein that an exponential temperature dependence of the conductance attests to the Anderson-insulating phase.

Our results emphasize the dominant role of the disorder in controlling the slow relaxation of electron glasses. Note that the present system has the lowest carrier concentration among the electron glasses studied to date that, in turn, it is also the *least disordered* electron glass; the disorder required to make the system insulating is smaller the smaller the Fermi energy (or equivalently N). It is due to these conditions that it was possible to observe the transition to the ergodic regime of relaxation.

As the transition is approached from the insulating side, the relaxation time concomitantly decreases with the reduced quenched disorder thus curtailing the time scale over which electron-glass attributes may be observed by conventional transport techniques.

It is illuminating to compare the glass dynamics reported here using In_xO with carrier-concentration $N \approx (8.7 \pm 0.2) \times 10^{18} \text{ cm}^{-3}$ with the dynamics observed in measurements made on another version of the same material with $N \approx (7.5 \pm 0.5) \times 10^{19} \text{ cm}^{-3}$ [34]. In the present paper the $\log(t)$ relaxation was shown to persist for two decades in time, whereas in the In_xO version with $N \approx (7.5 \pm 0.5) \times 10^{19} \text{ cm}^{-3}$ the logarithmic law was still observable for *two more* decades in time [34]. Figure 7 shows the magnitude of the memory dip versus the resistivity of these versions of In_xO that structurally are essentially identical except that due to a small change in composition they differ in carrier concentration. The figure demonstrates that, all other things being equal, the relative magnitude of the out-of-equilibrium excess conductance is considerably larger in the currently studied version of In_xO which, in turn, means that its much faster dynamics is not due to the limiting small signal, and conversely, the higher- N version

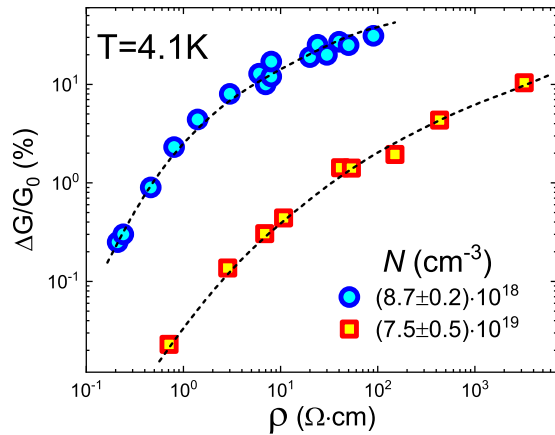


FIG. 7. The relative magnitude of the memory dip (see Fig. 2 for the definition) as a function of resistivity for two versions of In_xO films measured under the same conditions. The dashed lines are guides for the eye.

slower dynamics may actually be more than two decades. In other words, going down one decade in carrier concentration the electron-glass dynamics becomes faster by *at least* two decades.

This result may help to put in perspective the failure of some Anderson insulators, most notably Si and GaAs to exhibit intrinsic electron-glass features. The absence of lightly doped semiconductors from the list of systems that show glassy effects made it hard for some researchers to accept that the electron glass is just another property of the interacting Anderson-insulating phase. It has been suggested [18] that the failure to observe a memory dip in Anderson insulators with low carrier concentration is their fast relaxation times; to resolve the memory dip by a field-effect measurement requires relaxation times longer than the sweep time used in the $G(V_g)$ measurements. It was then conjectured that the relaxation time of Anderson insulators sharply depends on N , so only systems with large carrier concentrations exhibit a memory dip [18].

The finite relaxation time of the low- N version of In_xO studied here gives an unambiguous experimental point of reference in support of this conjecture. We expect that if a version of In_xO with $N \approx 10^{17} \text{ cm}^{-3}$ (typical for lightly doped semiconductors in their hopping regime) could be made and if its resistance at low temperatures could be made to be compatible with field-effect measurements, then its relaxation time will be too short to allow a memory dip to be observed just as is the case for Si and GaAs.

Carrier concentration is evidently an important parameter in determining the relaxation time of electron glasses, but it should be emphasized that other factors may play a significant role. In particular, the electron-phonon coupling is obviously an essential ingredient in energy relaxation processes of the electronic system. Further reduction of transition rates relative to the “bare” rates controlled by disorder may occur for nonlocal interactions. These may bring into play additional constraints [3] as well as effects related to coupling of the tunneling charge to other degrees of freedom (polaronic effects [33] and the orthogonality catastrophe [35–37]).

It is yet unclear how important is the long-range Coulomb interaction to the observed slow dynamics. Relaxation times that extend over thousands of seconds are observable at temperatures where the hopping length, which is the effective screening length in the insulating regime, is on the order of $\approx 10 \text{ nm}$. It seems therefore that, in addition to strong enough quenched disorder, medium-range interactions may be sufficient to account for the relaxation times observed in the experiments. This issue is now under investigation by using a metallic ground plane in proximity to the sample to modify the long-range Coulomb interaction in a controlled way.

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