Transition to exponential relaxation in weakly-disordered electron-glasses

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Abstract

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$_x$O films crosses-over asymptotically to an exponential dependence $\Delta G(t)\propto-\log(t)$ 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 $\tau(\infty)/(k_F\ell)C\propto-\log(t)$ 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 (MIT) 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.

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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: $M\propto\exp[-(t/\tau)^\beta]$. This allows assigning a well-defined relaxation-time $\tau(\infty)$ for a system with a distribution of relaxation-times $P(\tau)$ in turn, determines the value of $\beta$ and $\tau$.

Another form of relaxation that is ubiquitous is logarithmic $\propto-\log(t)$ law which may consistently be observed in electron-glasses with short relaxation-times. These observations were made on Anderson-insulating amorphous indium-oxide films In$_x$O 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.

EXPERIMENTAL

Sample preparation and characterization

Samples used in this study were thin 200 Å thick films of In$_x$O. These were made by e-gun evaporation of 99.9999% pure In$_2$O$_3$ onto room-temperature Si wafers in a partial pressure of 3x10$^{-4}$mBar of O$_2$ and a rate of 0.3±0.1Å/s. The Si wafers (boron-doped with bulk resistivity $\rho\leq$2x10$^{-4}$Ωcm) were employed as the gate-electrode in the field-effect and gate-excitation experiments. The samples were deposited on a SiO$_2$ layer (2μm 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-resistance $R_{\square}>$500MΩ at room-temperature. They were then
thermally treated. This was done by stages; the samples were held at a constant temperature $T_a$ starting from $T_a \approx 320$ for 20-30 hours then $T_a$ was raised by 5-10K at the next stage. This was repeated until the desired $R_\square$ was attained (see [17] for fuller details of the thermal-annealing process). This process yielded samples with $R_\square=20-40k\Omega$ that at $T\approx4K$ spanned the range of 100k$\Omega$ to 90M$\Omega$. A maximum $T_a \approx 355K$ and 14 days of treatment were required to get the lowest resistance used in the study. The carrier-concentration $N$ of these samples, measured by the Hall-Effect at room-temperatures, was in the range $N\approx(8.5\pm0.3)x10^{18}cm^{-3}$.

The motivation for choosing a low-$N$ version of In$_x$O for these experiments was the observation that the electron-glass dynamics becomes faster as their carrier-concentration falls below $N\lesssim 4x10^{18}cm^{-3}$ [15, 17] while, all other things being equal, their excess conductance $\Delta G$ is more conspicuous [17]. These expectations were borne out in our experiments which made it possible to quantify the system dynamics as it approaches the quantum phase transition.

**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 $T\approx4.1K$ held by a 100 liters storage-dewar. This allowed up to two months 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 experiment on a given sample.

The gate-sample voltage (referred to as $V_g$ in this work) in the field-effect measurements was controlled by the potential difference across a $10\mu F$ capacitor charged with a constant current. The rate of change of $V_g$ is determined by the value of this current. The range of $V_g$ used in this study reached in some cases $\pm 60V$ which is equivalent to the $\pm 15V$ used in previous studies [13] where the gate-sample separation was 0.5$\mu m$ as compared with the 2$\mu m$ 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. 1: The dependence of sample resistance on the applied field for three typical In$_x$O films, all with identical dimensions of 1x1mm. The dashed line connects points on each R(F) curve that their deviation from Ohmic behavior exceeds the experimental error.](image)

**Results and discussion**

Figure 2 shows the dependence of the conductance $G$ on gate-voltage $V_g$ for two of the studied In$_x$O samples. Each $G(V_g)$ plot show two main features; an asymmetric component characterized by $\partial G(V_g)/\partial V_g>0$, in the entire range of -$50V$ to $+50V$, that reflects the increased thermodynamic density-of-states with energy (the thermodynamic field-effect), and a cusp-like 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 45M$\Omega$ sample is $\approx 30\%$ - the highest value ever reported for any electron-glass with comparable $R_\square$, at $T\approx 4K$ and measured with a nominal sweep-rates in the range of (0.3-1)V/s. Similarly, the 105k$\Omega$ 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 $\Gamma$ 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 $V_{eq}$ to a new one $V_n$. 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 study was 12 hours. In all the experiments described here the total gate-voltage swing $|V_n-V_{eq}|$ was much larger than the memory-dip width $\Gamma$ defined in Fig.2. The relaxation to the equilibrium under the newly estab-
lished $V_n$ 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 to another time dependence was consistently observed in all samples of this and three other prepared batches of In$_x$O. 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.3b), then $\Delta G$ deviates from the logarithmic law, showing a tendency for the conductance to saturate. It turns out that for $t\geq 1500\,\text{s}$ 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.3c). Within the experimental error this $\beta$ is close enough to unity. Accordingly, we fit the data in this regime assuming $\beta = 1$, 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 $\beta$ from unity. Qualitatively similar results were obtained for the gate-protocol applied to a 105k$\Omega$ 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-noise ratio (due to the smaller $\Delta G/G_0$ associated with the lower $R_\square$ sample), the main difference between the data in Fig.4 and Fig.3 is in the relaxation-time $\tau(\infty)$. This time turns out to be independent of the particular value of $V_{eq}$ and $V_n$, as long as $|V_n-V_{eq}|$ is larger than the width of the memory dip $\Gamma$. This aspect was tested in the sample of Fig.4 using eight different runs of the gate-protocol with $20\leq |V_n-V_{eq}| \leq 90$ volts, and in seven runs for a sample with $R_\square=120\,\text{k}\Omega$ using voltage-swings in the range of 40-80 volts. In either case, the same behavior of $G(t)$ and the same value of $\tau(\infty)$ was observed. 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 mini-
the sample was held under $V_n=46$ V for a sample with $R_\square=120$ k$\Omega$ using two different equilibration times, $t_{eq}$. (a) $G(t)$ data for the entire duration of the protocol with $V_{eq}=0$ V, $V_n=46$ 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 $V_n$ reached $V_n$. Note the similar appearance of the curves (dashed lines are the respective best-fits).

In the range of equilibration times used in this work, 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 been noticed by comparing the time-scales in Fig.3 and Fig.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.

This trend has 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 [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 increase of 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 resistance 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 $k_F\ell=(3\pi^2)^{2/3}\cdot h\cdot \delta_{RT}\cdot e^{-2}\cdot N^{-1/3}$ where $\delta_{RT}$ 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 study (in terms of having a similar carrier-concentration $N$).

The diminishment of the relaxation-time as the metallicity 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 $T\to0$), 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 magnitude 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 23-85s for the samples with $k_F\ell$ in the range 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 [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 is 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$_x$O with carrier-concentration $N \approx (8.7 \pm 0.2) \times 10^{18}$ cm$^{-3}$ with the dynamics observed in measurements made on another version of the same material with $N \approx (7.5 \pm 0.5) \times 10^{18}$ cm$^{-3}$ [24]. In the present work the log($t$) relaxation was shown to persist for two decades in time whereas in the In$_x$O version with $N \approx (7.5 \pm 0.5) \times 10^{18}$ cm$^{-3}$ the logarithmic law was still observable for two more decades in time [24]. Figure 7 shows the magnitude of the memory-dip versus the resistivity of these versions of In$_x$O that structurally are essentially identical except that due to a small change of 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$_x$O which, in turn, means that its much faster dynamics is not due to the limiting small signal and conversely, the higher-$N$ version 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-time larger than the sweep-time used in the G(V) 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$_x$O studied here gives an unambiguous experimental point of reference in support of this conjecture. We expect that if a version of In$_x$O with $N \approx 10^{17}$ 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 non-local interactions. These may bring into play additional constraints.
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 of 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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