Slow Dynamics of the Electron-Glasses; the Role of Disorder

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Abstract

We examine in this work the role of disorder in contributing to the sluggish relaxation observed in intrinsic electron-glasses. Our approach is guided by several empirical observations: First and foremost, Anderson localization is a prerequisite for observing these nonequilibrium phenomena. Secondly, sluggish relaxation appears to favor Anderson-insulators with relatively large Fermi-energies (hence proportionally large disorder). These observations motivated us to consider a way to measure the underlying disorder in a realistic Anderson insulator. Optical study using a series of amorphous indium-oxide (In\textsubscript{2}O\textsubscript{3}) establish a simple connection between carrier-concentration and the disorder necessary to approach the metal-insulator transition from the insulating side. This is used to estimate the typical magnitude of the quenched potential-fluctuation in the electron-glass phase of this system. The implications of our findings on the slow dynamics of Anderson-insulators are discussed. In particular, the reason for the absence of a memory-dip and the accompanying electron-glass effects in lightly-doped semiconductors emerges as a natural consequence of their weak disorder.

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INTRODUCTION

Theoretical considerations anticipating nonequilibrium effects in Anderson-localized systems were described in a number of papers [1–12]. These were based on the interplay between disorder and Coulomb interactions leading to an electron-glass phase.

Over the last few decades there were several experimental studies that seem to give support to these expectations [13–15]. The dynamics that characterizes the approach to equilibrium of these systems is sluggish; relaxation of the excess conductance produced by driving the system far from the equilibrium, was observed to persist for many hours in some cases [16]. The long relaxation of the electronic system makes it possible to observe a modulation of the single-particle density-of-states (DOS) in field-effect experiments [17]. This feature, called a ‘memory-dip’ (MD), appears as a cusp-like minimum in the conductance versus gate-voltage. An example is illustrated in the inset to Fig.1. The memory-dip, presumably [8, 9], a reflection of an underlying Coulomb-gap [20, 21], is the identifying feature of intrinsic electron-glass [3, 6, 9, 11, 22].

To date, a memory-dip has been observed in seven different Anderson insulators (listed in Fig.1). The figure shows an empirical correlation between the typical width \(\Gamma\) of the MD and the carrier-concentration \(N\) of the material. The list of Anderson insulators that exhibit electron-glass properties include all types of degenerate Fermi systems; n-type semiconductors (Tl\textsubscript{2}O\textsubscript{3-x}, In\textsubscript{2}O\textsubscript{3}, In\textsubscript{2}O\textsubscript{3-x}), p-type semiconductors (GeSb\textsubscript{x}Te\textsubscript{y}, GeTe), and a metal (Be).

The systematic dependence of the MD characteristic width on carrier-concentration, an electronic property, is consistent with the expectation that the phenomenon is intrinsic. Due to lack of screening in the Anderson-insulating phase, Coulomb interaction can be comparable in magnitude to the quenched disorder, and therefore these competing ingredients responsible for glassy behavior are always present in Anderson insulators.

There seem to be more requirements on the material to allow observation of electron-glass effects. Note that all the system in Fig.1 are made from materials with carrier-concentration \(N\) limited to a range \(10^{22}\text{cm}^{-3} \geq N \geq 5\times 10^{19}\text{cm}^{-3}\). The absence of systems with \(N > 10^{22}\text{cm}^{-3}\) from this list is not surprising; a prerequisite for observing electron-glass effects is Anderson-
localization, which is hard to achieve in a system with large $N$ unless by making it granular (on which we remark later). Less clear is the limit of low carrier-concentration. No memory-dip has been reported in a system with carrier-concentration smaller than $\approx 10^{19}\text{cm}^{-3}$ in any Anderson-localized system like Si or GaAs. Two-dimensional samples of these materials may be tuned to show insulating behavior and were extensively studied in the hopping regime. Their near-equilibrium transport properties (like conductivity versus temperature) are not qualitatively different than those of the systems in Fig.1. The absence of a MD in these systems has been a vexing question for quite some time.

It was conjectured \cite{22} that relaxation processes in lightly-doped semiconductors (with $N \leq 10^{17}\text{cm}^{-3}$) are too fast to allow their signature to be captured by field-effect measurements. This was inspired by the observation of relaxation dynamics in a series of amorphous indium-oxide (In$_2$O) films \cite{22,23}. These can be fabricated with different $N$ values, covering $N \approx 5 \times 10^{18}\text{cm}^{-3}$ to $N \approx 5 \times 10^{21}\text{cm}^{-3}$ by controlling the In-O ratio \cite{24}. The typical relaxation times of these films sharply diminished \cite{23} once the carrier-concentration was reduced below $N \approx 10^{20}\text{cm}^{-3}$. Note that, given the sample-gate capacitance (as well as the other parasitic circuit capacitances), the field-effect temporal resolution is severely limited for high resistance samples. Consistent with this conjecture, ultrafast relaxation in a lightly-doped semiconductor was reported in phosphorous-doped Si \cite{25}.

What needs to be clarified is the role of carrier-concentration in affecting the dynamics of electron-glasses.

A phenomenon that is highly sensitive to the value of $N$ is often indicative of a many-body mechanism playing a role. Mechanisms that were considered in this regard include correlated many-particle transitions \cite{26}, and the Anderson orthogonality catastrophe \cite{27}. A model by Leggett al, based on coupling to an electronic-bath \cite{28}, is consistent with many of the observed features \cite{27}. These mechanisms, in conjunction with quenched disorder and hierarchical constraints, are likely contributing factors in the sluggish relaxation to some degree. However, to account for relaxation times of the order of hours with these scenarios still requires that the bare tunneling probabilities are much smaller than those involved in the dc conductivity.

The purpose of this work is to investigate the role played by disorder in slowing down charge rearrangement processes involved in the energy relaxation of electron-glasses. This is motivated by an alternative interpretation of the decisive role that seems to be played by the carrier-concentration of the system. Note that, at the transition, the magnitude of disorder in an Anderson insulator with large carrier-concentration must be larger than that of a system with low carrier-concentration, which may be summarized by a simple relation between the Fermi energy of the system and the critical disorder. It will be shown in this paper that this remains true deep into the insulating regime. Therefore, the empirical observation that the relaxation becomes slower once $N$ becomes larger, may turn out to be related to the weaker disorder rather than to various many-body effects (although the latter may be quite effective in further slowing down the relaxation).

The first step in examining this conjecture calls for assessing the magnitude of the disorder in the Anderson-insulating phase and its relation to the carrier-concentration of the system. We describe a set of experiments on several batches of amorphous indium-oxide with different carrier-concentrations, and with different degree of disorder in each batch. These data are analyzed to demonstrate a simple relation between disorder and the Fermi-energy of a batch with a given $N$ deep into the insulating regime. This, augmented by further arguments, is used to explain why Anderson-localized systems with low carrier-concentrations are unlikely to exhibit intrinsic electron-glass effects with relaxation times longer than few seconds.

To understand the motivation for our approach to the problem, we first review the main experimental findings pertinent to the electron-glass dynamics.

**Basic features of the dynamics in glassy Anderson-insulators**

Due to the lack of a concrete time-scale in their temporal relaxation (generally, a power-law), dynamics of glasses cannot be uniquely quantified. Tests to study, on a relative basis, how various agents affect the dynamics were performed on Anderson insulating crystalline indium-oxide, In$_2$O$_{3-x}$ and on In$_x$O with various compositions \cite{23,27,29}. These studies were also limited to effectively two-dimensional (2D) samples with sheet resistances $R_{\square}$ in the range of $\approx 1$M$\Omega$ to $\approx 500$M$\Omega$ where the signal to noise of the glassy features is favorable. With these system however, it was possible to test dynamics over a large range of lateral dimensions, from 2$\mu$m to 10mm \cite{21}.

The first observation, already alluded to above, is that dynamics seem to become faster when the carrier-concentration is smaller \cite{22}. Secondly, all features associated with slow dynamics disappear as the system crosses over to the diffusive regime \cite{22}.

Over the temperature range $\approx 2-8K$ there was no indication of dynamics freezeout \cite{27,29}. This is in stark contrast with the behavior of classical glasses where dynamics is quickly frozen below a certain temperature \cite{31}. The sample conductance over this temperature range typically changes by 1-2 orders of magnitude.

Transition-rates associated with conductivity of Anderson-insulators can be expressed as: $\omega \exp[-r/\xi]$
were $r$ is the hopping length and $\xi$ the localization-length. These rates typically range between $\approx 10^{10} \text{sec}^{-1}$ to $\approx 10^5 \text{sec}^{-1}$ based on the transition probability through a bottle-neck resistor with $r/\xi \approx 6-15$ (a typical value for the hopping regime) and assuming attempt frequency $\omega$ of the order of $10^{12} \text{sec}^{-1}$ which is commonly used as the prefactor in hopping conductivity. Here $r$ is the hopping-length and $\xi$ the localization-length. Relaxation rates of the studied electron-glasses are obviously slower by many orders of magnitude.

Another demonstration that conductance and relaxation processes appear to be different was recently observed in photoconductivity experiments on GeSb$_x$Te$_y$ films in their glassy regime; Adding charge to the system by optical excitation enhanced the conductance but it slowed-down the relaxation dynamics [32].

These observations suggest that relaxation and conductivity involve different processes. It is natural then to consider extrinsic effects as a viable mechanism for the slow relaxation exhibited by these systems. There are several candidates to choose from: ion-motion, surface-traps, grain-boundaries [33], and negative-U centers [34] are all potential sources for slow dynamics. Their coupling to charge-carriers might be the reason for the observed conductance relaxation observed in the experiments. In the first place, this would immediately explain why relaxation from an out-of-equilibrium state is controlled by different processes than those involved in the $dc$ conductance. It is also appealing as a source of a very slow phenomenon, intimately connected with the ubiquitous 1/f-noise phenomenon [35].

This however, is one of the problems with extrinsic mechanisms; flicker noise is indeed ubiquitous, intrinsic electron-glassiness is not. 1/f-noise may be observed in the metallic as well as in the degenerate transport regime while the electron-glass is an exclusive property of the localized phase. Furthermore, as mentioned above, a memory-dip, which is the identifying feature of the intrinsic electron-glass, has not been seen in any lightly-doped semiconductors while 1/f noise is quite evident in all these systems. GaAs samples in particular, exhibit prominent 1/f noise yet no slow conductance relaxation that might have compromised their operation as bolometers has been observed [36].

Other shortcomings of extrinsic scenarios include difficulties to account for the dependence of the memory-dip on temperature and disorder [22], they cannot explain the systematic dependence of the memory-dip width on carrier-concentration (Fig.1), and they cannot account for the peculiar evolution of the MD shape with temperature [22,31]. Finally, the failure of lightly-doped semiconductors to show a memory-dip remains enigmatic in this approach.

A more promising route to pursue is the ‘purely’ electronic scenario which accounts for most of the observed features [3,9,11], and as argued below, the reason for the absence of intrinsic glass effects in low carrier-concentration systems emerges naturally from this picture. In this approach, both conductivity and relaxation in the electron-glass proceed via charge-carriers transitions between localized states. The qualitative difference between transitions involved in relaxation and those that control conductivity is that conductance must include activated processes while relaxation, in our scenario, is dominated by tunneling between states differing in energy by $\delta \ll k_B T$ [27].

The driving force for the relaxation is minimizing the electrostatic energy $E_{el} = \sum_{i,j} \frac{e^2}{r_{ij}(r_{ij})}$ under the constraints set by the disorder (assumed to be quenched). A simple example of an energy-reducing event due to charge re-arrangement is illustrated in Fig.2.

A transition of the type shown in Fig.2 for example, would result in an energy release of the order of $\delta \approx \frac{e^2}{r_{12}}$, that amounts to a substantial excess energy injected into the system (except for a rare $r_2 \approx r_1$ event). This may enhance the excess conductivity via two possible mechanisms:

First, by generating an excess of nonequilibrium phonons. Although some of these generated phonons would escape to the bath, it does not take much of overheating to appreciably affect the conductivity in this regime. Consider for example, a typical electron-glass with $R_\square \approx 10 M\Omega$ at $T=4K$. This, when excited, will have an excess conductance $\Delta G/G$ of the order of $\approx 1\%$ (see, e.g., Fig.6 in [37]). Overheating by $\approx 5mK$’s (at 4K) is enough to generate this excess conductance given the $R(T)$ of such a sample (stretched exponential with a power of 1/3 or 1/2 with activation energies of $\approx 5000K$ and 8000K for the indium-oxide and Be sample [37] respectively). Even much less overheating will be needed deeper in the electron glass regime; $\Delta G/G$ increases algebraically with disorder but the sensitivity of $G$ to temperature increases exponentially with it.

Secondly, a transition event at any point $R_i$ in the system may induce transitions in another site $R_j$ either di-
rectly via Coulomb interaction (when \(|r_i-r_j|<r_h|\) or indirectly via a succession of ‘avalanches’ (for \(|r_i-r_j|>r_h|\))\[^{38,39}\] where \(r_h\) is the hopping-length. As alluded to above, there is no metallic screening in the Anderson-localized phase but at finite temperatures the Coulomb interaction is effectively cutoff at distances larger than the hopping-length \[^{38,39}\]. These avalanches, much like the Domino-Effect, ultimately spread to modulate the space-energy configuration of the CCN thus affecting the measured conductance. Relaxation of the excess-conductance would last as long as such energy-reducing transitions occur. It should be noted that the probability of these transitions to occur depends also on the electron-phonon coupling strength (direct electron-electron inelastic transitions are essentially suppressed in the strongly-localized system relative to its value in the diffusive regime \[^{38}\]).

There is also a quantitative difference between transitions involved in the dc conductance and those contributing to relaxation processes. Conductance is determined by relatively fast transitions between pairs of sites composing the current-carrying-network (CCN). These are the pairs of sites connected by the relatively high transition probabilities in the Miller-Abrahams conductance distribution. In the hopping regime the CCN typically occupies a tiny fraction of the system volume; most of the material is ‘dead-wood’; these are the high resistance regions of the system \[^{40-42}\], and this is naturally where the slowest transitions occur. Energy-relaxation in the Anderson insulator involves energy-reducing transitions anywhere in the system including in particular the dead-wood regions.

The relative weight of the dead-wood gains in prominence when the disorder gets larger or when the temperature gets smaller. In our conjecture, this would account for the experimentally observed increase of the relative magnitude of the MD resistance whether affected by disorder, field or temperature \[^{43}\].

The dead-wood, occupying the bulk of the system volume, holds most of the excess-energy associated with the out-of-equilibrium state. Therefore, conductance relaxation in this picture is essentially controlled by the dead-wood slow-dynamics.

**Dynamics in the dead-wood**

The question we need to address concerns the transition rates of tunneling events of the general type illustrated in Fig.2. Tunneling probability depends exponentially on the distance \(\mathcal{L}\) and on an effective-barrier \(V^*\). Tunneling distance \(\mathcal{L}\) for electrons could easily be much larger than that typically found for ions and may extend over \(10^2\)-\(10^3\) atoms in solids. This balances out their transition rates as compared with ions with their \(\approx10^4\) times heavier-mass. Tunneling probability may be vanishingly small if \(\mathcal{L}\) is long enough. However, the most likely transition-scale at the asymptotic regime of the relaxation is perhaps of the order of the Bohr-radius \(a_B\). Transitions over longer distances likely proceed by series of short events. Such ‘serial’ events, as well as other ‘hierarchical processes’, would further slow down the dynamics and should be treated separately.

The quantum transmission through a slab of an Anderson insulator of a finite cross section and length has been worked out by Nikolić and Dragomirova \[^{44}\]. They studied the statistics of the associated eigenvalues as function of disorder and for several lengths. The distribution of transmission eigenvalues was found to be very wide. In the presence of moderate disorder it included a finite portion of near resonance channels which may play a role in local hierarchical processes but they do not directly contribute to relaxation. On the other hand, the distribution of transmission probability was heavily skewed in favor of low transmission channels \[^{43}\] even for weak disorder while the resonances naturally disappeared when critical disorder was imposed. It is plausible that including electron-electron interaction in such calculations will only enhance the peak of the low transition probabilities in the spectrum. Slow relaxation events should therefore be abundant in the system.

Tunneling probability \(\gamma\) through a simple (square) barrier may be estimated by a WKB expression:

\[
\gamma \propto \exp \left( -2 \left( \frac{2m^*V^*}{\hbar^2} \right)^{1/2} \cdot \mathcal{L} \right) ; V^* = V - E
\]

(1)

where \(m^*\) is the effective-mass of the charge carrier, \(V\) the barrier-height and \(E\) the particle energy (which will be taken as \(E_F\)). The spatial form of \(W(r)\) in a realistic Anderson insulator obviously requires a more elaborate treatment than addressed by the WKB approximation (probably a numerical work along the lines of \[^{42}\]). However, the exponential dependence on \(m^*\) \(\mathcal{L}\) and \(V^*\) should still be reflected in a more detailed treatment. This was recently demonstrated in a numerical study extending the calculations of \[^{44}\]; in the localized regime, the probability to find transmission trajectory with value smaller than a given \(\gamma\) is exponential with \(W^{1/2}\) and \(\mathcal{L}\) \[^{15}\].

To estimate how slow a typical tunneling event may be, one needs to know the magnitude of the \(V^*\). We shall assume it to be of order of the potential-fluctuation \(W\). The value of \(W\) is the distinguishing factor in determining whether electron-glass effects can be observed by field-effect measurements in a given system. \(W\) may vary by orders of magnitudes while \(a_B\) is typically in the range of 20-50Å. Larger values for \(a_B\) are usually due to very light effective-mass which counteracts the effect of longer tunneling-distance on the tunneling probability.

To anticipate the discussion below, recall that a pre-requisite for electron-glass behavior is Anderson-localization, which means that the disorder energy \(W\)
The ‘critical’ disorder in Anderson-insulators

There are several ways to characterize the magnitude of disorder in the Anderson-localized phase. For example, the value of the localization length $\xi$ is, in a way, a measure of disorder. However, determining $\xi$ involves transport measurements at low temperatures where one probes just the CCN thus ignoring the most disordered part of the sample. Another choice is the Ioffe-Regel parameter $k_F \ell$ that may be estimated from Hall effect and resistivity measurements at relatively high temperatures [17]. $k_F \ell$ may be taken as a measure of disorder whenever $\rho$ is dominated by the elastic mean-free-path $\ell$. This condition is well-obeyed in the vicinity of $k_F \ell \gtrsim 1$ where the conductivity is scale-independent [17] and the estimate of $k_F \ell$ is then least sensitive to the specific temperature at which $\rho$ is measured. In the regime $k_F \ell < 1$ neither $k_F \ell$ have their usual meaning but $k_F \ell$ may still be a useful parameter to characterize disorder; it is just a dimensionless parameter that decreases monotonically with disorder. On the other hand, $k_F \ell$ is not simply related to static disorder once $k_F \ell \ll 1$ (or $k_F \ell > 1$, a regime irrelevant for this discussion anyhow). In these limiting cases the room-temperature conductivity may be dominated by inelastic processes rather than by static disorder.

Given a system with a certain $k_F \ell$ one still requires a way to assign a value for its $\mathcal{W}$. Theoretical estimates may be used for the ‘critical disorder’ $\mathcal{W}_c$ the disorder necessary to just localize the entire band. Estimates based on a non-interacting picture yield the ratio $\mathcal{W}_c/1 \approx 16.5$ [14]. Here $I$ is of the order of the bandwidth, typically several electron-volts. In these models the Fermi-energy is taken at mid-band where the density-of-states is highest. The models however are more vague when the Fermi-energy is near the band-edge where $\partial n/\partial \mu$ may be rather small. Unfortunately, this is invariably the situation in real Anderson-insulators [14]. It is certainly the case for each of the seven electron-glasses listed in Fig.1.

A more helpful approach then is to rely on the physics of the diffusive regime and extrapolate to the transition point defined by the value of $k_F \ell$ at the transition to the localized phase obtainable from experiments. A measure of disorder for a diffusive system is $\mathcal{W} = \hbar/\tau$ where $\tau$ is the transport mean-free-time.

On the metallic side the ratio $\hbar/\tau = \frac{2}{k_F \ell}$, so at the metal-insulator transition:

$$\mathcal{W}_c = \beta E_F$$

where $\beta$ depends on the specific value of $(k_F \ell)_c$ - the Ioffe-Regel parameter evaluated at the transition to the localized phase:

$$\beta = \frac{2}{(k_F \ell)_c}$$

The experiments described next, suggest that the proportionality between $E_F$ and $\mathcal{W}$ is still valid deep into the insulating regime $(k_F \ell \ll 1)$, covering the entire range of resistances probed in the electron-glass studies.

Gauging disorder by monitoring optical properties; tuning disorder in the amorphous indium-oxides

A system that allows a continuous tuning of disorder for both sides of the metal-insulator is amorphous indium-oxide In$_x$O. It may also be prepared with a vastly different carrier-concentration (between $\approx 5 \times 10^{18}$ cm$^{-3}$ to $\approx 6 \times 10^{23}$ cm$^{-3}$) by controlling the In/O ratio. This makes it possible to study the dependence of the electron-glass properties on carrier-concentration and disorder [24], as well as the metal-insulator [24] and the superconductor-insulator transition [49].

The feasibility of tuning the system resistivity by heat-treatment is a rather unique trait of In$_x$O. It is possible to vary the room-temperature resistance of the as-prepared In$_x$O sample by up to 5 orders of magnitude while maintaining its amorphous structure and composition (and hence carrier-concentration). A before-and-after diffraction pattern illustrating the preservation of amorphicity in the heat-treatment is shown in Fig.3. This figure demonstrates that the amorphous structure remains intact during thermal annealing. Actually, one is hard pressed to see a difference in the before-and-after diffraction patterns (Fig.3). It takes a careful measurement of the diffraction-ring intensity-profile to discern the difference; a narrowing of the diffraction rings, as shown in Fig.4. The change in the resistance is essentially due to modified mobility; Hall effect studies showed only a small change as a result of the annealing process [46, 50]. It was also found that the material volume decreases during the process which could be detected by measuring the thickness of the film for example, by x-ray interferometry as demonstrated in Fig.5.
FIG. 3: Electron diffraction patterns (using 200kV beam) of a ≈50 nm In\textsubscript{x}O film for the as-prepared specimen with sheet resistance $R_{\square}>10^8\Omega$ (a) and after thermal annealing for 34 days yielding $R_{\square}≈7\times10^4\Omega$ (b). The general appearance of the pattern seem unchanged in the annealing but a careful comparison of the strong diffraction ring between the (a) and (b) micrographs reveals slightly less fuzziness in the annealed sample (illustrated in Fig. 4 below).

FIG. 4: (color online) Intensity profile of the strongest rings in the diffraction pattern of both (a) and (b) micrographs shown in Fig.3 as function of distance $r$ from the diffraction center. Data were taken by averaging line-scans using imagej.

FIG. 5: (color online) Reflection-interferometry using a Cu Kα x-ray source (wavelength=1.54Å) for the before-after annealing stages of a similar film as in Fig.3. The inset shows the Fourier transform of the main plot exposing both the reduced film thickness in the annealed sample and the enhanced visibility of the interference pattern.

FIG. 6: (color online) Absorption versus photon energy for a before-and-after In\textsubscript{x}O sample (characterized by carrier-concentration $N≃8.5\times10^{19}\text{cm}^{-3}$). Dashed lines delineate the respective values of the energy-gap.

This result was part of an extensive work designed to measure the change in the optical properties that accompany the thermal-annealing of In\textsubscript{x}O samples with different carrier-concentrations. We bring here fuller results and interpretation of these data. The volume-change caused by annealing the sample was reflected in the absorption versus energy plot as reduction of the optical-gap. This is illustrated in Fig.6 for one of the studied batches.

The dependence of absorption coefficient $\alpha(\omega)$ on energy $\omega$ of all our In\textsubscript{x}O samples obeys the relation:

$$\alpha(\omega)h\omega = B(h\omega-E_g)$$

A volume-reduction that occurs concomitantly with a smaller optical-gap is often observed in pressure studies...
of amorphous materials \[51\text{–}53\]. The increase in wavefunctions overlap due to the reduced volume leads to a wider band-width and to a modified $\partial n/\partial \mu$ (E). A schematic of such a change in the conduction-band shape is depicted in Fig. 7. Note that this depiction is consistent with the observed before-and-after absorption-curves and with the sum-rule of the number of states. However, it does not into account a modification in the mobility-edge shift offered in \[46\].

The similarity in the effects produced by applying hydrostatic pressure \[51\text{–}53\] and thermally-annealing the sample is not surprising; in both cases energy is supplied to a metastable system allowing it to cross barriers associated with its being a structural glass.

Our study include a detailed comparison of the optical properties with simultaneously measured changes of the disorder parameter $k_F\ell$. The values for $k_F\ell$ and the Fermi-energy $E_F$ used here are based on free-electron formulae; $E_F=\frac{k^2(3\pi^2N)^{1/3}}{2m}$ and $k_F\ell=(3\pi^2)^2/2 \sigma_{RT}/e^2 N^{1/3}$ where $\sigma_{RT}$ is the conductivity (measured at room-temperature) and $m^*$ is the effective mass.

Both, optical and electrical measurements were performed in-situ on thick (900-1100Å), effectively 3D films of In$_x$O. Data for the optical gap $E_g$ were collected at intermediate stages of the annealing process starting from the as-prepared sample with typically $k_F\ell \leq 10^{-3}$ and ending in $k_F\ell > 1$. The process was repeated with films of various carrier-concentrations. Fig. 8 shows the dependence of $E_g$ on $k_F\ell$ for each of the studied samples.

The systematic trend that emerges here is that the optical gap $E_g$ is a monotonous function of $k_F\ell$ or rather, both $k_F\ell$ and $E_g$ depend monotonically on the sample volume. Secondly, for a given change in $k_F\ell$, the concomitant change in $E_g$ depends on the carrier-concentration $N$ of the batch; $\Delta E_g$ is bigger for the batch with the larger $N$. The change of the refractive index n (judged by the change in the prefactor B in Eq.3 \[54\]) during the annealing process showed a similar trend. Fig. 9 shows a systematic correlation between $E_F$ and the change in disorder of $n$ and $E_g$ for each of batches studied (for a unity-change in $k_F\ell$).

**ANALYSIS AND DISCUSSION**

Before proceeding it may be a good idea to check whether our assumptions so far are not at odds with reality. We have focused on In$_x$O because of available data pertaining to this system with a wide range of carrier-concentration and disorder. In particular, the metal-insulator transition has been studied in this material for In$_x$O versions with $N \approx 10^{19}$cm$^{-3}$ and $N \approx 10^{21}$cm$^{-3}$. Both versions were found to cross the transition at a similar $k_F\ell \approx 0.32\pm 0.02$ \[24\]. To localize In$_x$O with $E_F \approx 0.3$eV, which is the highest Fermi-energy in the series studied here, $W_C$ has to be $\geq 1.9$eV (to be consistent with Eq.3). Is such disorder a viable occurrence in this system?
FIG. 9: (color online) $\Delta E_g$ and $\Delta n$ are the respective changes in the optical-gap $E_g$ and the refractive-index $n$ when the disorder is changed (by thermal annealing) from $kF\ell\approx0.02$ to $kF\ell=1$. The data points stand for each of the batches studied (labeled by symbols corresponding to each of the batches in Fig.8).

A main source of disorder in $In_xO$ is chemical [24, 46]. This is associated with deviation from stoichiometry; relative to the ionic compound $In_2O_3-x$, there are 5-30% oxygen vacancies in $In_xO$ [24]. To preserve chemical neutrality some of the indium atoms must assume a valence of +1 instead of the +3 they have in the stoichiometric compound. When randomly distributed these valence-fluctuations form a background potential of $e^2/\tau\approx5eV$ (assuming an average interatomic-separation of $r\approx3\;\AA$ [53]). This type of disorder is quite prevalent in non-stoichiometric compounds, metallic-oxides, high-Tc materials etc. The associated disorder $W$ in these materials may reach 4-5eV.

In addition to chemical-disorder, there is an off-diagonal disorder in the material that is partially alleviated in the thermal annealing process. This disorder is related to the distributed nature of the inter-atomic separation (the random values of inter-atomic separation). The distribution of inter-particle distances (and thus wavefunction overlap) obviously gets narrower as the volume decreases and the system approaches the ‘ideal’ closed-packed amorphous structure. The diminishment of this, off-diagonal disorder, is clearly reflected in the enhanced visibility of the x-ray interference pattern of the annealed sample in Fig.4 and also in the electron diffraction Fig.3 and Fig.4 (albeit less conspicuously).

It has been a controversial issue whether off-diagonal disorder can lead to localization [50, 58]. It should be noted that, in the $In_xO$ system, the metal-insulator transition is crossed by the combined effect of changing the atomic overlap (which in turn affects the density of states as schematically described in Fig.6) while simultaneously modifying the off-diagonal-disorder. Localization (at the less-annealed regime) is also aided by the underlying chemical disorder, and possibly by the Coulomb interaction [59].

Two features of the data in Fig.7 and Fig.8 are worth noticing: First, for a given batch (fixed $N$) there is a systematic and monotonous dependence of $E_g$ on $kF\ell$ although the transport measures essentially just the elements of the CCN while optics is sensitive to the whole sample. The correlation between the two measurements is indicative of an underlying common conductance-distribution; being disordered the system is inherently inhomogeneous but it is so in a generic way (as embodied in the percolation treatment of hopping conductivity). Inhomogeneity associated with large thickness or composition variations across the sample will in general not show the correlation between transport and a bulk measurement. Secondly, the change in both the optical-gap and the refractive index between $kF\ell=1$ and $kF\ell\approx0$ exhibits a near-linear correlation with the Fermi-energy of each batch (Fig.8 and Fig.9).

As noted above, during annealing the sample volume decreases (Fig.5), which among other things, causes the refractive index to increase. $\Delta n$ may then be used to estimate the associated change in the system energy per particle $\delta E$. Let us take the largest swing of $\Delta n$ in the series being $\approx20\%$ for the sample with $E_F\approx300$ meV (Fig.8). The average change of the interparticle-separation in this case is $\approx7\%$ (an estimate consistent with the thickness change upon annealing measured independently for this sample) giving $\delta E\approx0.07x5eV=350$ meV. Note that this energy is close to the respective change of the optical gap for this batch (Fig.8). The near-linearity of $\Delta n$ and $\Delta E_g$ with $E_F$ guarantees that proportional results are obtained for the entire series of samples studied here. This means that the disorder-energy in the range $kF\ell=1$ to $kF\ell=0$ changes by no more than $E_F$ of the batch under study.

For the electron-glass phase of $In_xO$ however the relevant range of $kF\ell$ is only $0<kF\ell<0.32$. $In_xO$ sample with $kF\ell\approx0.02$ has a resistance of $\approx1G\Omega$ at 4K [24], therefore the change in $\delta E$ is even smaller than $E_F$. The entire change of disorder relevant for the electron-glass phase of $In_xO$ can be concisely summarized as $\delta E<\beta E_F$ where $\beta\approx0.3$. This can be combined with Eq.2 to give a measure of the disorder $W_{EG}$ in the electron-glass regime (namely, deep in the Anderson-localized phase):

$$W_{EG} \approx (\beta + \beta')E_F = \beta'E_F$$  \hspace{1cm} (5)

Most of the disorder in $W_{EG}$ is actually due to $W_C=\beta E_F$. An order of magnitude estimate for $W$ in the diffusive regime may be taken as $W=h/\tau = hV_F/\ell$ ($\tau$ is the transport mean-free-time). With $E_F=h^2k_F^2/2m^*$ this yields for the ratio $W/E_F=2/kF\ell$. As the transition of $In_xO$ is approached from the diffusive regime $kF\ell\rightarrow0.32$ and $W\rightarrow W_C\approx6.2E_F$. Increasing the disorder above this point, further localizes the system but, as the resistance is exponential with disorder in this regime, a relatively
small added amount of disorder $\approx 0.3E_F$ is necessary to get deep ( $kp\ell \ll 1$) into the insulating state.

The near-linear relation between disorder and Fermi-energy established here for In$_x$O samples is significant; these samples cover more than a decade range in carrier-concentration and over this range the dynamics is changing by almost three orders of magnitude [23]. This is a result of the exponential dependence of relaxation dynamics on $W_{\text{EG}}$ as is argued next.

Our conjecture is that a relation between $W$ and $E_F$ of the form expressed by Eq. 5 and with $\beta^*$ of similar magnitude as in In$_x$O is generally obeyed by Anderson insulators.

If lightly-doped semiconductors follow the same trend, their dynamics should be much faster than in electron-glasses with $N \geq 3 \times 10^{20} \text{cm}^{-3}$. To illustrate, let us use eq. 1 on relative basis to compare the typical tunneling probability for the In$_x$O sample with $N \approx 3.3 \times 10^{20} \text{cm}^{-3}$ with that of a lightly-doped semiconductor like Si. Equation 1 with $m^* = 0.3m_0$, $V^* \approx 1.7$-1.9eV, and $\mathcal{L} \approx 20\AA$ gives $\gamma \approx 10^7$. The parameters for Si-MOSFET in the Anderson insulating regime [49] are similar except for $V^*$; given a carrier-concentration of $N \approx 10^{17} \text{cm}^{-3}$, typical of the insulating phase in this system [60], $E_F$ is $\approx 100 \times$ times smaller. Since $\beta^*$ is likely only smaller when the Fermi-energy is reduced [18], the respective $W$ for Si should be at least two orders of magnitude smaller than that of the In$_x$O sample, which means five to six orders of magnitude faster dynamics (assuming comparable electron-phonon coupling strength and a similar spectral nature of the disorder). If conductance relaxation in the In$_x$O may be observed for days, it may not last more than seconds in lightly-doped semiconductor like Si. With such a relaxation time it would be practically impossible to observe the memory-dip in field-effect experiments. It is hard to see how any parameter peculiar to lightly-doped semiconductor can compensate for their weak disorder. Note that the estimate made above is just for the bare tunneling, it does not take into account coupling to the environment, which would further increase the discrepancy between these rates [27].

A test of these considerations is to instill a disorder of few electron-volts in a lightly-doped semiconductor: by our conjecture, slow relaxation and intrinsic electron-glass effects should be observable in this case. Unfortunately, the measurement of such a sample may turn out to be a tall order. With such strong disorder ($W/E_F \gg 1$) it is doubtful whether the conductance is measurable even at temperatures that exceed the Fermi-energy while, like in any other electron-glass one should maintain $T \ll E_F/k_B$ to make such a measurement, which for these systems means $T \ll 1K$. At these temperatures the resistance for a sample with such $W$ will balloon out of reach.

The difficulty can be demonstrated by reference to an experiment using as a model, a thin film of In$_2$O$_{3-x}$, a system where the resistance can be manipulated over a considerable range. In the experiment, shown in Fig. 10, the room-temperature resistance $R_{\text{RT}}$ of the In$_2$O$_{3-x}$ film is changed by UV-treatment [51]. Samples with $R_{\text{RT}}$ spanning the range of $\approx 16$-180k$\Omega$ were generated from a single batch in this method. At $T \approx 4.1K$ these samples had resistances spanning $\approx 6$ orders of magnitude. The value of $R_{\text{RT}}$ may be taken as a (linear) measure of disorder only when the sample is in the diffusive regime, $R_{\text{RT}} \lesssim h/e^2$. Strong-localization behavior apparently sets-in for $R_{\text{RT}} \gtrsim 40k\Omega$ where the linear relation between disorder and $R_{\text{RT}}$ does not hold. Over the range of $R_{\text{RT}} = 16$-32k$\Omega$, a mere factor of $\approx 2$ in disorder, the resistance at 4.1K changes by $\approx 4$ orders of magnitude. By extrapolation from the regime where $R_{\text{RT}}$ is a linear measure of disorder, changing it by an order of magnitude would increase the resistance (at $\approx 4K$) by $\approx 20$ orders of magnitude, yielding a resistance that defies current measurements techniques. We suspect that similar catastrophe would occur upon cranking-up the disorder in lightly-doped semiconductors.

A more feasible test, is a low-temperature study of a weakly-localized system (kp$\ell \gg 1$). This can be realized for instance by using a two-dimensional (2D) film of In$_2$O$_{3-x}$ which has been extensively studied in the weakly-localized regime. Being 2D, the system should crossover to the strongly-localized regime [41] at sufficiently low temperature despite having sub-critical disorder (in the 3D sense). At sufficiently low temperatures its resistance could be as high as a sample with kp$\ell \ll 1$ of this material that exhibits glassy effects at say $\approx 4K$ [15, 22, 27]. The logic presented above anticipates that the weakly-disordered sample would exhibit at best very weak memory-dip in field-effect experiments.

![Figure 10](image-url) FIG. 10: (color online) The sheet resistance of a In$_2$O$_{3-x}$ film at $T=4.1K$ versus its value at room temperature. The dashed line depicts the expected value of resistance extrapolated along the line where the room-temperature resistance may be a linear measure of disorder. The dotted curve is a guide for the eye.
(it may not show a null effect due to occasional regions with potential-fluctuation of large amplitude).

It should be emphasized that by itself, large resistance does not guarantee electron-glassiness; resistance may result from many factors not related to the type of disorder discussed here. For example, regions of hard-gaps such as a series inclusion of band-insulator or isolated islands of superconductivity in the current-path \[24\] may exhibit huge resistances without necessarily showing glassy effects on extended time scales.

Another corollary of the proposed picture is that relaxation-times cannot be arbitrarily long. This is a result of the maximum value of \(W\) available in reality. Naturally occurring defects in condensed matter systems have an energy of the order of band-widths, which limits their ability to Anderson-localize a system. Indeed, to localize a typical metal with Fermi-energy of \(\approx 3-4\) eV, one has to mix it with another material, usually a band-insulator, rendering the system granular.

Some of the electron-glass transport features are exhibited by granular systems in their activated regime. In particular, they show a memory-dip and slow relaxation \[62\]. The element of the relaxation in the granular systems, common with the electron-glass, is re-distribution of charges in space to lower the ‘electrostatic’ energy of the system. Their dynamics however is expected to be different than that of Anderson-insulators; charge re-distribution between different grains is probably controlled by Coulomb blockade constraints. Charging energies of typical grains could be quite large, which may lead to extremely long relaxation times. This was demonstrated by the Bar-Ilan group; in their experiments on several granular systems it was shown that below few degrees Kelvin dynamics became so slow that it was impossible to follow the evolution of a memory-dip \[63\]. The constraints posed by charging-energies may be partially alleviated by random fields in the insulating matrix in which the metallic granules are embedded but then the dynamics is controlled by extrinsic elements. These issues deserve further study.

In summary, based on the empirical attributes of the electron-glass dynamics, lead us to conclude that a significant part of the slow relaxation observed in systems with relatively large carrier-concentration has to do with their much stronger disorder. The elements of the relaxation process are assumed to be tunneling events that proceed to ultimately minimize the system energy under the constraints of disorder and Coulomb interaction. The slowest transitions naturally occur in the dead-wood regions of the system that in the hopping regime occupy most of the system volume. Slow ‘fluctuators’ are abundant in these regions even for a rather short tunneling distance once the disorder is appreciable. An estimate of the disorder-energy \(W\) for a realistic sample that shows electron-glass properties was made using \(\text{In}_x\text{O}\) as a model system. This was based on measurements of the metal-insulator transition made on these compounds, free-electron concepts, and on optical data relevant for their strongly-disordered regime. It was shown that \(W\) is proportional to the Fermi-energy of the Anderson-insulating system for the entire range of disorder relevant for the electron-glass measurements. All other things being equal, and given the exponential dependence of the tunneling rate on \(W\), one expects energy-relaxation in lightly-doped semiconductors where \(E_F\) is two orders of magnitude smaller, to be many orders of magnitude faster as indeed established experimentally \[21\].

There is still the challenge of accounting for the protracted relaxation-times observed in the experiments. This is a problem common to all glasses but the flexibility of experimenting with electron-glasses seem to offer more scope for progress. From the theory point of view the problem has proved to be difficult; Even when a complete knowledge of the disorder is at hand there are other pieces of the puzzle that need careful elaboration. In particular, coupling to the environment may play a role in further slowing-down transition-rates. Environmental degrees of freedom that follow adiabatically the tunneling object would modify the transition rate through mass enhancement (polaronic effects). More generally, coupling to the environment will suppress tunneling due to the Anderson orthogonality catastrophe (AOC) \[27\]. Recent work focused on new aspects of the AOC (originally conceived for clean systems \[64\]) in the strongly-localized and interacting regime \[65, 66\]. However, a model incorporating coupling to the environment for a medium lacking screening (like an Anderson-insulator), is more pertinent for our experiments. To our knowledge, this aspect has not been adequately addressed by any work so far. Other effects that probably contribute to slow relaxation are hierarchical constraints (the ‘domino’ effect being a special case), and correlated many-electron transitions. Fundamental questions related to these scenarios are yet to be resolved. However, to understand why a memory-dip is not likely to be observed in lightly-doped semiconductors by transport measurements, it may suffice to consider just the role of disorder.

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