Infrared-Induced Sluggish Dynamics in the GeSbTe Electron Glass

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Infrared-induced sluggish Dynamics in the GeSbTe Electron-Glass.

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Abstract

The electron-glass dynamics of Anderson-localized GeSbTe films is dramatically slowed-down following a brief infrared illumination that increases the system carrier-concentration (and thus its conductance). These results demonstrate that the dynamics exhibited by electron-glasses is more sensitive to carrier-concentration than to disorder. In turn, this seems to imply that many-body effects such as the Orthogonality Catastrophe must play a role in the sluggish dynamics observed in the intrinsic electron-glasses.

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The interplay between disorder and Coulomb interactions makes Anderson insulators a natural candidate to exhibit out-of-equilibrium transport properties characteristic of glasses [1–12]. Glassy features such as slow relaxation, ageing, and other memory effects related to such an electron-glass (EG) scenario have been observed in a number of systems [13]. A feature common to all these systems is relatively high carrier-concentration n. Intrinsic electron-glass effects with relaxation times longer than seconds seem to be peculiar to Anderson insulators with n spanning the range of $\approx 4\text{x}10^{19}\text{cm}^{-3}$ (crystalline indium-oxide) to $\simeq 10^{22}\text{cm}^{-3}$ (beryllium) [13]. The relaxation time τ in amorphous indium-oxide with $n<8\text{x}10^{19}\text{cm}^{-3}$ dropped by more than two orders of magnitude when n was reduced by a mere factor of ≈three [14]. This is also consistent with the absence of intrinsic EG effects in lightly-doped semiconductors; experiments on phosphorous-doped silicon exhibited relaxation times much shorter than a second [15]. Condensed-matter phenomena that favor high carrier-concentration is usually a many-body effect (screening, superconductivity). It is therefore of interest to study this aspect in a controlled manner. The fast dependence of τ on n observed in [14] employed amorphous indium-oxide samples with different *compositions* to obtain different carrier-concentrations. The recent observation of coexistence of electron-glass phase with persistent-photoconductivity in $GeSb_xTe_y$ [16] gives us a unique opportunity to check this issue in a *single* sample. Persistent-photoconductivity (PPC) is used in this work to change the carrier-concentration of the system by a considerable amount without changing the main structural features of the sample [17]. It is shown that, in the PPC-state, the electron-glass dynamics is slowed down by a more than an order of magnitude relative to the dark-state. This dramatic effect may help in elucidating the mechanisms responsible for the slow dynamics exhibited by intrinsic electron-glasses as it hints on the importance of many-body effects.

Samples used for this study were prepared by e-gun depositing a $GeSb_2Te_5$ alloy unto room temperature substrates. The substrates were 0.5μm SiO_2 layer thermally grown on <100> silicon wafers having bulk resistivity $\rho \simeq 2\text{x}10^{-3}\Omega$cm, deep into the degenerate regime. The wafers were employed as the gate electrode in the field-effect measurements. Polycrystalline samples of $GeSb_xTe_y$ were obtained by crystallizing the as-deposited (amorphous) films a temperature of 460±5K. 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≈4.1K held by a 100 liters storage-dewar. This allowed up to two months measurements on a given sample while keeping it cold (and in the dark). Optical excitations employed AlGaAs diode operating at ≈0.88±0.05μm, mounted on the sample-stage at a distance of ≈12mm from the sample. The diode was energized by a computer-controlled Keithley 220 current-source. Fuller details of sample preparation and characterization are described elsewhere [16].

The protocol used for assessing the electron-glass dynamics is the two-dip-experiment (TDE). This is illustrated in Fig.1 showing a series of conductance vs. gate-voltage $G(V_g)$ traces taken at different times for a $GeSb_xTe_y$ sample in the dark-state. The first $G(V_g)$ trace was taken 24 hours after the sample was cooled down to T=4.1K and allowed to equilibrate while V_g=5.8V was held between the sample and the gate. The resulting field-effect trace is composed of two components; an anti-symmetric component reflecting the underlying (thermodynamic) density-of-states (DOS), and a superimposed dip, centered around the gate-voltage where the systems was allowed to relax (V_g=5.8V in this particular case). The latter, so called memory-dip (MD), is the distinguishing feature of the intrinsic electron-glass [17], reflecting an underlying Coulomb-gap [5, 9, 12, 13]. The width of the memory-dip is comparable with that of the amorphous version of indium-oxide [14] with the same carrier-concentration $(6\pm3)\text{x}10^{20}\text{cm}^{-3}$. A second trace shown in Fig.1a was taken after the gate-voltage was moved to V_g=-5.8V and left there for 10 minutes before a new $G(V_g)$ trace was taken. This trace shows a dip centered at the newly imposed gate-voltage while

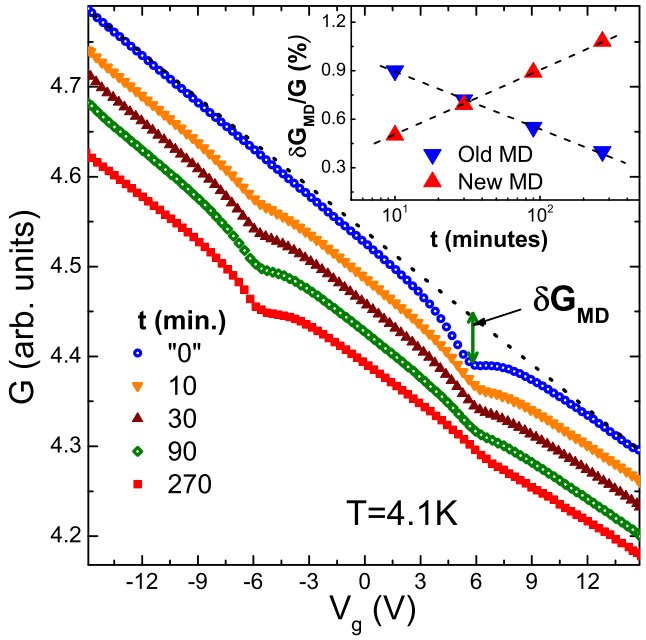

FIG. 1: (color online) The two-dip-experiment performed on a $GeSb_xTe_y$ film with $R_\square$=4.4MΩ in the dark-state. Curves are shifted for clarity. The dotted line depicts the underlying thermodynamic density-of-states. Inset shows the time dependence of the old and new memory dips (see text for details). Sample lateral dimensions are W=1 mm, L=$\frac{1}{2}$ mm.

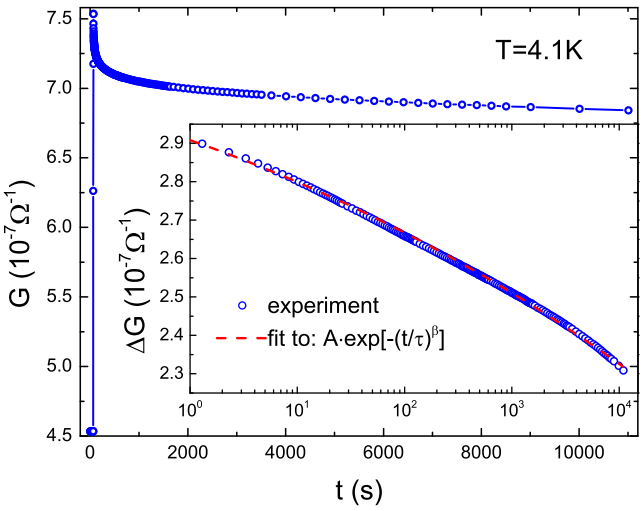

FIG. 2: (color online) The time dependence of the conductance before, during, and after exposing the sample (as in Fi1.1) to infrared (3 seconds at 1mA). The inset depicts fitting the relaxation of G in the induced PPC-state. Fitting parameters are: A=3.375, τ=2x10^8 sec, β=0.1.

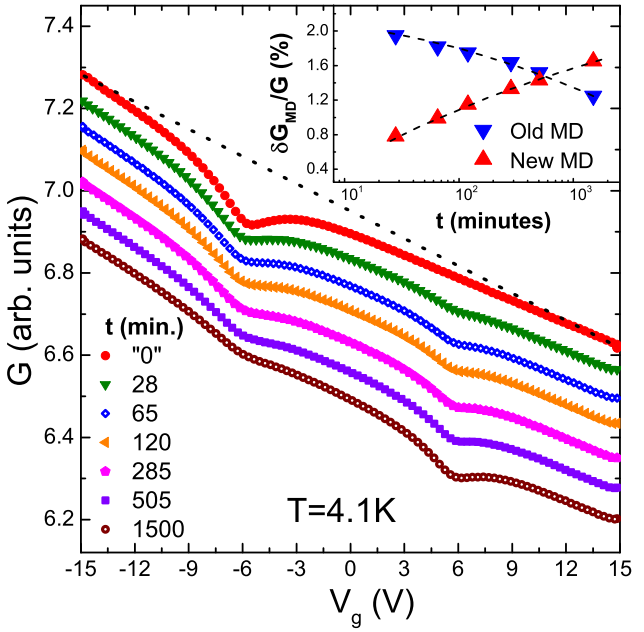

FIG. 3: (color online) The two-dip-experiment as in Fig.1 but after the sample is put in its PPC-state. Curves are shifted for clarity. The dashed lines are guides for the eye.

the old dip is diminished in magnitude (marked in the figure as δG_{MD}). This protocol is repeated to produce the other curves shown in Fig.1a while V_g is parked at -5.8V between gate-sweeps taken at later times.

The inset to Fig.1 shows the time dependence of the magnitude of the old and new MD's which, in agreement with previously studied electron-glasses, exhibit logarithmic dependence [10, 18]. A typical time characterizing the dynamics of the TDE protocol is the time where the magnitude of the old MD equals that of the new one. This turns out to be ≈33 minutes, which again is similar to the result obtained for In_xO film with comparable carrier-concentration measured under identical protocol [14] (including history).

This dynamics is observed when the sample is kept in the dark. A dramatic change is observed when the system is cast into its persistent-photoconductivity (PPC) state [16]. This is achieved by exposing the sample to the infrared source causing the conductance G to increase by ≈50%. The effect of the infrared excitation and the ensuing conductance relaxation are shown in Fig.2

The inset to Fig.2 depicts the relaxation of the excess conductance ΔG created by the IR excitation. This ΔG(t) fits well a stretched exponent, $\Delta G(t) \propto \exp[-(t/\tau)^\beta]$ with β=0.1±0.01. The same relaxation law with the *same* β (for samples measured at T=4.1K) has been seen in more than dozen other $GeSb_xTe_y$ films with sheet resistances $R_\square$ ranging from 2.5kΩ to 25MΩ [16]. The infrared excitation has also increased the magnitude of the MD as can be seen in the top curve for $G(V_g)$ in Fig.3. This trace was taken ≈22 hours after the infrared source was turned off. It is the starting stage for a TDE protocol of the system in its PPC-state. The same protocol was used in the $G(V_g)$ series shown in Fig.3 as in the previous two-dip experiment of Fig.1 but it had now to be extended over much longer time to accommodate data

points where $\delta G_{MD}(t)/G$ of the new MD exceeds the amplitude of the old MD. As may be seen from the inset to Fig.3 the typical time of the TDE in this case is ≈580 minutes, about 17-times longer than for the system in its dark-state. Note (inset to Fig.3) that $\delta G_{MD}(t)/G$ curves for both the new and old MD's deviate from the logarithmic law that characterize their time dependence in the dark-state TDE series (Fig.2). This presumably is a result of the system drifting towards its dark-state with its associated smaller MD amplitude [16]. It is not however the reason for the perceived slowdown of the dynamics in the PPC-state; the same typical time for the TDE would be gathered by using the extrapolated crossing point for logarithmic curves based on the first 2-3 points in the series.

The more than an order of magnitude increase in the TDE typical time (Fig.3 vs. Fig.1) was also observed on two other $GeSb_xTe_y$ films with $R_\square$=2.1MΩ and $R_\square$=8.5MΩ. We also confirmed that both, the enhanced magnitude of the MD and the slower dynamics of the PPC-state are reversible; by keeping the sample for a minute or two above 30-40K then re-cooling to T=4.1K the dark-state on all its previous features are restored, including the faster dynamics.

It is difficult to get a direct experimental reading of the change in the previously established MD in the dark-state immediately after turning off the infrared source; the fast change of G(t) in the initial stage of the PPC relaxation would overwhelm a $G(V_g)$ measurement. There is however an indirect way to infer something about this issue from $G(V_g)$ measurements performed in later times, starting from ≈12 minutes after the infrared illumination. Results of these measurements are shown in Fig.4. Note that 12 minutes after the infrared exposure the MD amplitude $\delta G_{MD}(t)/G$ is ≈85% of its maximum value (obtained at intermediate times; at long times $\delta G_{MD}(t)/G$ actually *goes down* as the PPC-state dissipates and the dark-state value is recovered [16]). At the same time however, the *shape* of the MD differs from its asymptotic form. This can be seen in the inset to Fig.4; normalizing the amplitude of the 12 minutes MD to that of the 9-hours MD fails to show the simple scaling that characterizes how the memory-dip evolves with time under other protocols. In particular, the MD shape is time independent following quench-cooling the sample from high temperatures [19]. The wider MD shape of the 12-minutes trace is consistent with a "hotter" or more energetic electronic state than in either the dark-state or the asymptotic PPC-state [19].

The "hotter" nature of the 12-minutes MD suggests that the infrared exposure disturbed the relaxed configuration of the dark-state. Judging by the ΔG produced by the illumination, the added carriers density in the PPC-state Δn is a sizeable fraction of the carrier-concentration of the dark-state [16], in this particular case of the order of $\approx 10^{19}cm^{-3}$. Changing the charge-density of the interacting system naturally takes it out of equilibrium. It is less clear that it could also slow-down its dynamics. This needs elaboration.

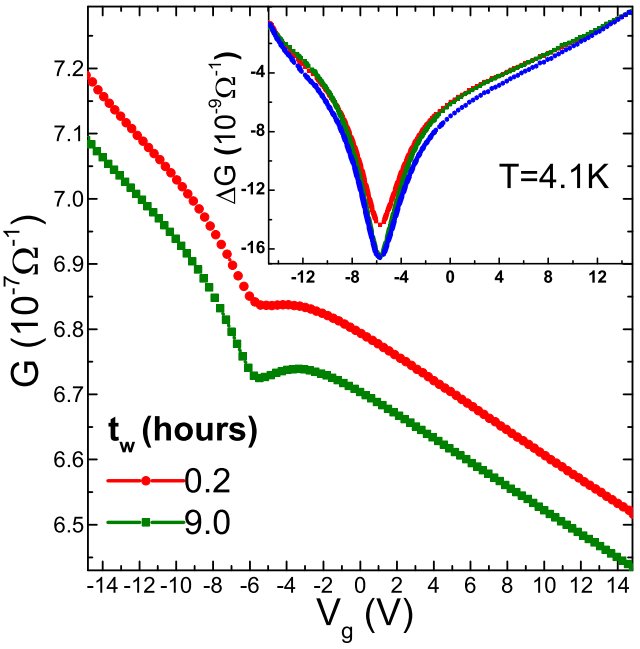

FIG. 4: (color online) Conductance vs. gate-voltage sweeps for the sample taken at the indicated times after the sample was exposed to infrared radiation (using the protocol described in Fig.2). The inset shows the corresponding MD obtained by subtracting a straight line from the respective $G(V_g)$ trace (with corresponding labels as in the main figure). The blue circles are data points for the 12-minutes MD multiplied by a constant (1.157) to match the ΔG value of the 9 hours trace at equilibrium V_g. This illustrates that the 12-minutes MD is wider than the long-time value {these MD's appear to coincide at the end-points of the V_g measured-range "by-construction", due to the way they were obtained from the composite $G(V_g)$}.

Relaxation from an excited state of the electron-glass proceeds by transitions between localized states. So does the system conductance. One should be careful however in drawing conclusions about relaxation from observations pertinent to conductance. In the first place, there are many more sites involve in relaxation than those participating in dc transport [13, 21]. Conductance is controlled by the relatively fast transitions in the current-carrying network [20] while relaxation must also involve transitions in the 'dead-wood' regions of the system. Local dynamics in these regions, which occupy most of the system volume, is also orders of magnitude slower than that of the sites that are part of the current-carrying-network. The global relaxation rate is a hierarchical process composed of avalanche-like events propagating through the *entire* system [21]. Secondly, while conductance is essentially controlled by single-particle transitions, relaxation requires energy-diminishing events and

these ultimately necessitate simultaneous *multi*-particle transitions [21]. These transitions are required to approach the lowest energy configuration and it is plausible that their relative importance grows with the carrier-density [22, 23].

Conductance and relaxation may also differ *qualitatively:* In several electron-glasses where temperature dependence was measured over a range of few degrees around 4K it was found that while conductance increases (exponentially) with temperature, relaxation remained constant, or even becomes *slower* [24] with it. This work demonstrates the distinction between conductance and relaxation in a different way; adding carriers to the system by photo-excitation boosts one while impeding the other. It is also important to note that the enhanced conductance makes it unlikely that the dynamics slowdown in the PPC state is due to increased disorder. While it is the interplay between interaction and disorder that induces glassy behavior, it appears that the inter-particle interaction (naturally being affected by a change in the carrier-concentration) is the more dominant factor in slowing down relaxation rates.

The mechanism responsible for the detrimental effect of carrier-concentration on relaxation may be either specific to the way Δn is generated [or that $\Delta n=\Delta n(t)$], or it is of a generic nature. These are discussed in turn.

Relaxation slowdown results from introduction of constraints on spatial re-arrangement of charges. A mechanism specific to the PPC-state might result from the local displacement of the ions from which charge was photo-generated [25]. It is not clear however that these defects could be more effective in hindering charge organization than other defects so abundant in these materials such as grain-boundaries, vacancies, etc. which are unaffected in the dark-state$\rightarrow$PPC-state transition [16]. Indeed, there is no hint that these added defects play a role even in the conductance noise; the amplitude of the 1/f power-spectrum is actually somewhat smaller in the PPC state.

Adding charge to a system naturally increases its conductance. In the hopping regime this occurs by effectively reducing inter-site energy-differences. However, it may also introduce another element that offsets this effect. Doping a semiconductors for example also introduces scattering centers which compromise the mobility. In the hopping regime added charge may decrease tunneling probability by reducing wavefunctions overlap. Such a mechanism is the Anderson orthogonality-catastrophe (AOC) [24, 26]. Localized-states in Anderson insulators that exhibit long-lived MD are typically *multiply*-populated [27]. These are essentially small disordered metals which actually accentuates the AOC effect over the 'clean limit' [28]. Tunneling probability between these mesoscopic elements, and the presence of neighboring ones acting as bath, are reduced due to the AOC. The reduction is more important the smaller is the "bare" tunneling probability, slower transitions are made *much* slower, fast ones are barely affected [24, 28]. This *non*-specific mechanism for the slowdown does not depend on the method used to increase n, provided that the extra charge is spread evenly throughout the sample volume (as in the process of doping the sample or alloying it with another component that changes the composition of the system). This requirement guarantees that the increase in carrier-concentration includes the 'dead-wood' regions. As noted above, the slowest transition-rates involved in energy relaxation are due to the dynamics in these regions.

The AOC scenario is supported by the observation that each and every Anderson insulator with large carrier-concentration tested to date has exhibited intrinsic EG effects (and indirectly, by the short relaxation times in lightly-doped semiconductors [15]). More experiments will be required to elucidate these issues. In particular, using the PPC technique with materials that are borderline in terms of their carrier-concentration and extending the measurements to lower temperatures may produce useful information.

In summary, persistent-conductivity has been used to change the carrier-concentration in the electron-glass phase of $GeSb_xTe_y$ films [29]. This resulted in an enhanced conductance and a substantial slowing-down of the relaxation dynamics relative to the dark-state. This highlights the fundamental difference between conductance and energy relaxation of electron-glasses and further illustrates the many-body aspects of these phenomena. Neglecting many-body effects in treating the dc conductivity of Anderson insulators may not lead to obvious discrepancies with experiments. But using single-particle models to account for the long relaxation times of the intrinsic electron-glasses is unlikely to yield much progress.

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