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Intrinsic electron-glassiness in strongly-localized Be films

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Abstract

We present results of out-of-equilibrium transport measurements made on strongly-localized Beryllium films and demonstrate that these films exhibit all the earmarks of intrinsic electron-glasses. These include slow (logarithmic) relaxation, memory effects, and more importantly, the observation of a memory dip that has a characteristic width compatible with the carrier-concentration of beryllium. The latter is an empirical signature of the electron-glass. Comparing various non-equilibrium attributes of the beryllium films with other systems that exhibit intrinsic electron-glasses behavior reveals that high carrier-concentration is their only common feature rather than the specifics of the disorder that rendered them insulating. It is suggested that this should be taken as an important hint for any theory that attempts to account for the surprisingly slow relaxation times observed in these systems.

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INTRODUCTION

The interplay between static disorder and Coulomb interactions may precipitate a glassy state in an Anderson insulator. This ‘electron-glass’ scenario was discussed in several papers [1–4]. In theory, this property is generic to *all* degenerate Fermi systems with localized states interacting via a Coulomb potential. Experimental evidence for these glassy effects, however, has been somewhat scarce, presumably due to specific material requirements. It turns out that only systems with relatively high carrier-concentration n exhibit relaxation times that can be conveniently monitored by transport measurements. Conductance relaxations that persist for many seconds, and memory effects characteristic of *intrinsic* [5] electron-glass, seem to be peculiar to systems with $n > 10^{19}\text{cm}^{-3}$ [5]. A prominent group of materials that exhibit electron-glass behavior with long relaxation times are granular metals; Al [6], Bi [7], Pb [7], Ni [8], and Au [9], all having high carrier-concentration $n \gtrsim 10^{22}\text{cm}^{-3}$.

Hitherto, the only non-granular systems that exhibited intrinsic electron-glass behavior were crystalline and amorphous indium-oxide films ($\text{In}_2\text{O}_{3-x}$ and In_xO respectively) [5], which are ionic compounds.

In this work we report on the low temperature transport properties of strongly-localized Be films, and demonstrate that they exhibit intrinsic glassy effects. This is the first non-granular mono-atomic system to show these effects. These include logarithmic relaxation of the out-of-equilibrium conductance and, more importantly, a memory-dip that has all the earmarks of intrinsic electron-glass. Although beryllium, like a typical metal, has a Fermi energy E_F of few electron-volts, it has an unusually low density of states at E_F [10]. A signature of the Be peculiar density of states, namely, at the Fermi energy, the density-of-states decreases with energy,

is actually observed in our field effect measurements as will be demonstrated below. The low density of states of beryllium is presumably the main reason why strong localization is achievable in this material just by making the sample thin enough (yet still physically continuous).

EXPERIMENTAL

Samples used in the experiments reported here were $18 \pm 5 \text{Å}$ films deposited as described elsewhere [11] on $140 \mu\text{m}$ glass slides. These were silver-painted on their backside so as to form a gate for the field effect measurements. The samples were typically $400 \mu\text{m}$ wide and $600 \mu\text{m}$ long strips and had sheet resistance R_{\square} in the range $23\text{k}\Omega$ to $120\text{k}\Omega$ at 295K and $100\text{k}\Omega$ - $160\text{M}\Omega$ at $\approx 4\text{K}$. The strongly-localized nature of the films at this temperature range was tested by measuring their conductance versus temperature dependence $G(T)$ in the range 4 - 50K . Below $\approx 10\text{K}$, this dependence is consistent with $G(T) \propto \exp[-(\frac{T_0}{T})^{1/2}]$ with T_0 in the range 100 - 900K . Figure 1 illustrates this behavior for two of the samples that were used in this study.

The different values of R_{\square} in the studied series of samples were obtained by a judicious oxidation of the Be films in an oxygen-enriched chamber. The change of the samples resistance was constantly monitored during the oxidation process. At the range of sample thickness $d \approx 18 \text{Å}$, the room-temperature sheet-resistance of the samples is $\approx 20\text{k}\Omega$. Such samples are usually deep into the hopping regime at liquid helium temperatures, and a small change in their thickness d translates into a large change of R_{\square} at 4K . The sheet-resistance of samples much thicker than 18Å may not reach the quantum resistance at 4K (recall that $R_{\square} > \hbar/e^2$ is a pre-requisite for electron-glass behavior [5]). For example, in a previous study [12] a beryllium film with a nominal thickness

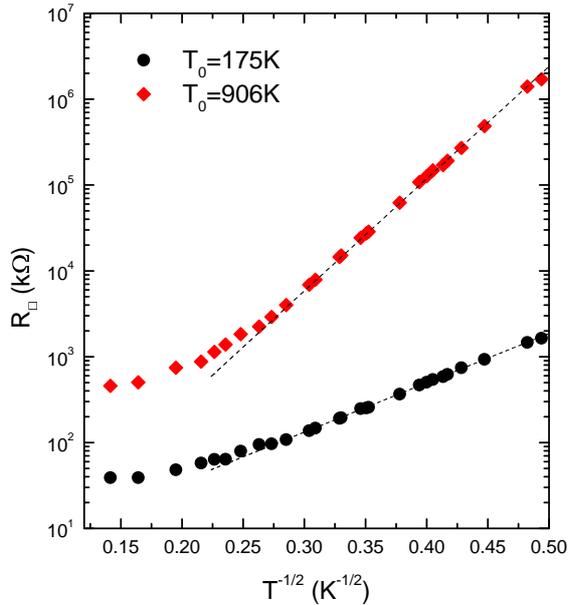


FIG. 1: Resistance versus temperature plots for two of the beryllium films used in this study.

d of ≈ 20 Å, only slightly thicker than the films studied here, exhibited $G(T) \propto \exp[-(\frac{T_0}{T})^{1/2}]$ with $T_0 = 1.6$ K. This should be compared with $T_0 \gtrsim 100$ K in the samples used in this work. The much smaller value for T_0 , and the associated larger localization length in this case means that strongly-localized behavior is attained only at temperatures that are well below the range covered here. Such samples are not included in our present study where one of our goals is to compare the results with previously studied electron-glasses, which were measured at or near 4K.

The conductivity of the samples was measured using a two-terminal ac technique employing a 1211-ITHACO current pre-amplifier and a PAR-124A lock-in amplifier. Measurements reported below were performed with the samples immersed in liquid helium at $T = 4.1$ K maintained by a 100 liters storage-dewar, which allowed long term measurements of samples as well as a convenient way to maintain a stable temperature bath. Unless otherwise indicated, the ac voltage bias was small enough to ensure linear response conditions (judged by Ohm's law being obeyed within the experimental error). Fuller details of measurements techniques are given elsewhere [13].

A variety of techniques were employed to characterize the films microstructure. Fig.2 shows a Transmission Electron Microscope (TEM) micrograph of a Be film prepared in the same way and with similar thickness as the samples used for the transport studies. The physical continuity of the film is evident in the figure. On care-

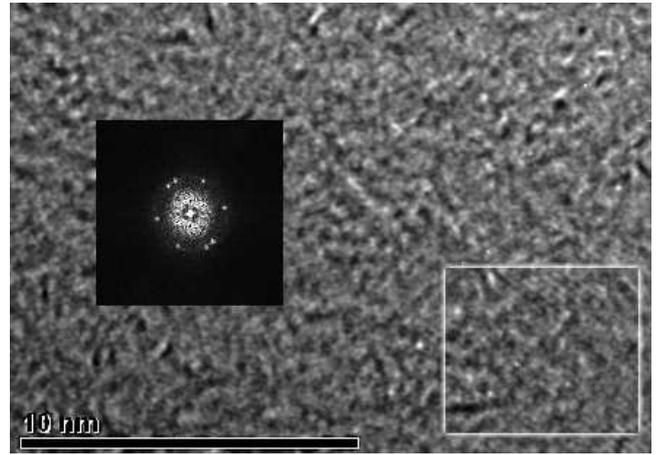


FIG. 2: Transmission Electron Microscope micrograph of ≈ 20 Å Be film deposited on carbon-coated Cu grid. The black square is a Fourier transform of the framed area on the right. The latter shows 2.33 Å fringes associated with the d-spacing of the $\langle 100 \rangle$ planes of the hexagonal BeO (reflected as satellites in the Fourier transform plate).

ful examination, the micrograph shows occasional fringes that indicate the presence of small crystals. These were identified as BeO by direct imaging and further, by their diffraction pattern (interestingly, the diffraction pattern of Be, being light on electrons, was presumably too weak to register a clear pattern over the background set by the amorphous carbon support-film). The BeO crystals were clearly observable in TEM dark-field imaging off their $\langle 100 \rangle$ diffraction line. This enabled an estimate of their crystallographic size and partial volume in the samples. Randomly distributed BeO crystallites of sizes up to 50 - 70 Å were observed in these dark-field images. We estimate that less than $\approx 10\%$ of the film area is occupied by fully oxidized Be crystallites, and therefore transport presumably occur through non-oxidized Be phase. Yet, the insulating BeO crystallites, somewhat restrict the volume available for conductivity (much like punching holes in the film would). A result of this geometrically-constrained structure is that the transport properties of the films show some mesoscopic effects that one usually encounters in smaller systems measured at similar temperatures and comparable degree of disorder [14, 19].

The physical continuity of the Be phase in the film was ascertained by performing local electron-energy-loss-spectroscopy (EELS) on the parts of the film that were not occupied by BeO crystals. The EELS spectra taken from these areas was consistent with that of metallic Be. A slight shift of energy, $\leq +3\%$ of the peaks position in the spectra was detected, possibly due to strain. The presence of free Be in the samples was also confirmed by X-ray Photoemission Spectroscopy, which was carried out on the actual samples that were used for the transport measurements.

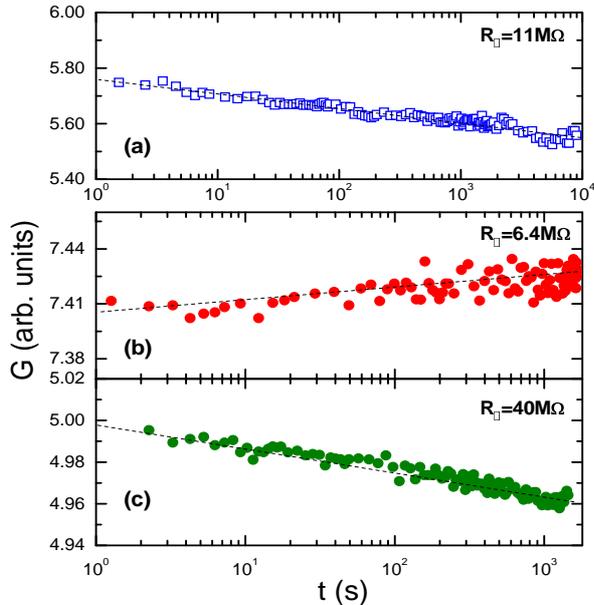


FIG. 3: Non-equilibrium transport behavior of typical Be films under different protocols; (a) After a quench-cool from $T \approx 100\text{K}$ to $T = 4.11\text{K}$. (b) Starting from near-equilibrium, $G(t)$ was recorded from the instance the source-drain voltage was increased to $10V_{ac}$ from the (Ohmic) 20mV_{ac} bias. (c) Same sample as (b) after the 20mV_{ac} has been restored. Each graph is labeled with the average R_{\square} of a sample under the measurements conditions.

RESULTS AND DISCUSSION

We turn now to the non-equilibrium transport properties of the films. The first signature of glassy behavior in these films is encountered upon quench-cooling the sample to 4K ; after an initial fast drop (reflecting the change in temperature), the conductance G keeps on decreasing slowly (logarithmically) long after the sample has reached the bath temperature. A typical quench-cooling protocol is shown in Fig.3a. The figure also illustrates the slow conductance excitation process upon ‘stressing’ [15] the film with a non-Ohmic source-drain voltage Fig.3b. The ensuing relaxation of G after the source-drain voltage was set back to its Ohmic value is shown in Fig.3c. These excitation-relaxation curves are clearly similar to those previously observed in glassy $\text{In}_2\text{O}_{3-x}$ samples [15]

A controlled way to take the system out of equilibrium is a change of the potential difference between the sample and a near-by gate. This technique has been widely used in the study of several electron-glasses [5–7, 13]. Among other things, it may be used to estimate a typical relaxation time τ under a given set of conditions [16]. An example of such protocol is illustrated in Fig.4. In this protocol, one uses the conductance relaxation law $\Delta G(t/t_0) = \Delta G(1\text{sec}) - a \cdot \log(t/t_0)$ where t_0

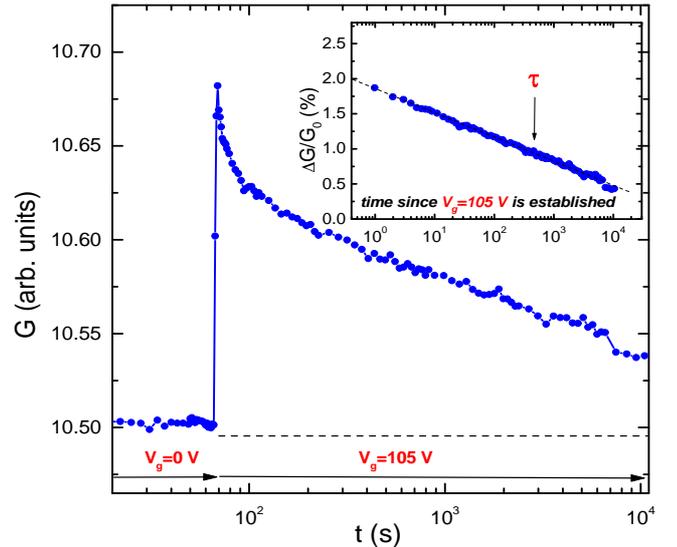


FIG. 4: A gate-excitation protocol for a specific Be sample ($R_{\square} = 40\text{M}\Omega$). The insert depicts the characteristic $\log(t)$ dependence of the EG from which the typical relaxation time τ is estimated using the baseline conductance $G_0(105\text{V})$ (marked here by the dashed line).measured independently.

is the experimental resolution time, and the equilibrium value of G at $V_g = 105\text{V}$ to extract the value of τ defined by $G(\tau) \equiv \frac{1}{2}G(1\text{sec})$.

A characteristic feature that is believed to be common to all intrinsic electron-glasses is a memory-dip; this is a cusp-like minimum in $G(V_g)$ centered at the gate voltage V_g where the system was allowed to equilibrate [5–7, 17]. A conspicuous memory-dip was consistently observed in all our Be films. Fig.5 shows this feature for two Be samples in the studied series. For both samples, $G(V_g)$ scans were taken after a $\simeq 24$ hour equilibration under $V_g = 0$ volt. Note first that the memory-dips have the same shape and width independent of R_{\square} and independent of whether the $G(V_g)$ scans were taken by sweeping V_g through the equilibrium- V_g or *symmetrically* around it [13] (c.f., the lower graph of Fig.5).

In previously studied electron-glasses the width of the memory-dip was found to systematically depend on the carrier-concentration n of the system [17]. On the basis of the $G(V_g)$ data, we have estimated the typical width in our Be films in the same manner as was done in [17]. This involves several stages; First the change of charge ΔQ associated with the cusp-width is estimated from ΔV_g by taking heed of the sample-gate capacitance. The relevant energy is then calculated using the beryllium $(\partial n / \partial \mu)_{E_F}$ and the screening length. This procedure gave the energy-width Γ^* (as defined in [17]) as $\approx 8\text{meV}$. Using the empirical relation between the width and n (Fig.4 of [17]) such Γ^* corresponds to n of order $10^{21}\text{--}10^{22}\text{cm}^{-3}$. This is consistent with our Hall ef-

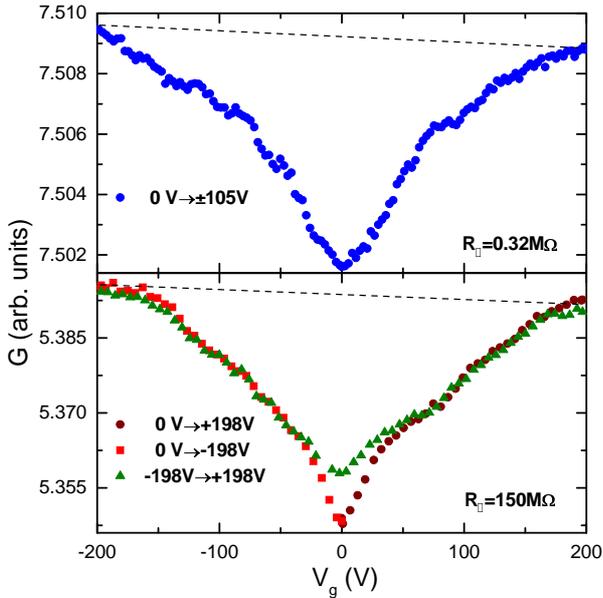


FIG. 5: The conductance versus gate voltage for two Be films (labeled by their R_{\square}) illustrating the memory-dip structure. After the sample was allowed to equilibrate under $V_g=0$ for ≈ 24 hours, scans were taken either from $V_g=0$ in each voltage direction (symmetrical $G(V_g)$ scans) or, from -198 V to $+198$ V. Note that the structure is skewed due to the contribution of the anti-symmetric (equilibrium) field-effect (see text).

fect measurements that gave $n \cong 7\text{-}8 \cdot 10^{21} \text{cm}^{-3}$ as well as with the concentration predicted by band-structure calculations ($n = 0.016$ state/atom [18] tantamount to $n \simeq 1.7 \cdot 10^{21} \text{cm}^{-3}$). This correlation between the width of the memory-dip and the carrier concentration of the material is an important empirical test for the intrinsic nature of the electron-glass [5].

The $G(V_g)$ traces (Fig.5) reveal some mesoscopic fluctuations (reproducible with V_g scans) superimposed on the memory-dip (note e.g., the modulation of $G(V_g)$ around $V_g \approx -120$ V, and $+90$ for the 0.32 M Ω and the 150 M Ω samples respectively). As alluded to above, these presumably result from the small effective volume for conductance due to the presence of the insulating BeO regions.

The relative magnitude of the memory-dip $\Delta G/G$ grows monotonously with R_{\square} . Interestingly, $\Delta G/G(R_{\square})$ for Be is almost identical to that measured under the same conditions in other electron-glasses. In Fig.6 we compare the results of the current study with some old data [19] taken on $\text{In}_2\text{O}_{3-x}$ films exhibiting quite similar behavior. A similar agreement is observed between that data of Fig.6 and the results obtained on granular aluminum films (c.f., Fig.17 of [20]).

On the other hand, the anti-symmetric part (c.f., Fig.6) of the $G(V_g)$ for the beryllium samples has the

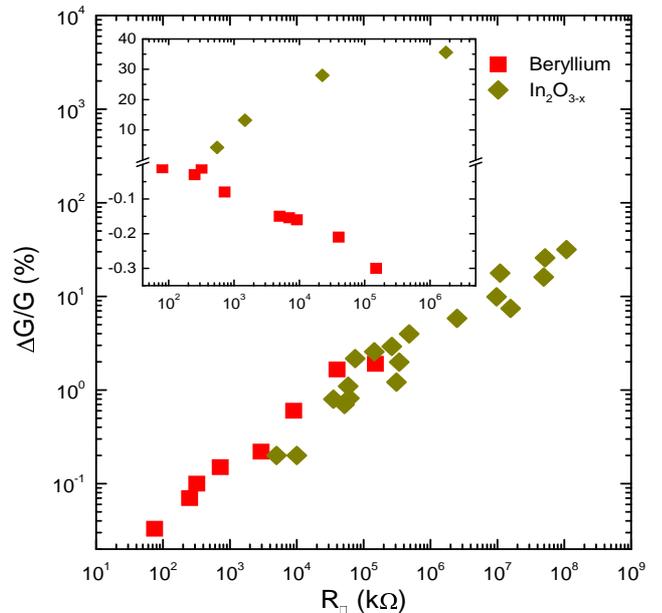


FIG. 6: The relative magnitude of the memory-dip versus R_{\square} for the studied Be samples. The data of [15] are shown for comparison. Insert: The slope (relative change of G per 400 V of V_g) of the anti-symmetric part of $G(V_g)$ compared with typical results for $\text{In}_2\text{O}_{3-x}$ samples.

opposite slope to that observed in $\text{In}_2\text{O}_{3-x}$ and In_xO [5, 13]. The sign of this slope is controlled by the energy derivative of the thermodynamic density of states $\partial n/\partial \mu$ at the Fermi level; $\partial G(V_g)/\partial V_g \propto \frac{\partial}{\partial E}(\partial n/\partial \mu)_{E_F}$. The anti-symmetric part of $\partial G(V_g)/\partial V_g$ we observe in all our Be films (represented by the dashed curves in Fig.5) is consistent with the negative slope of $\partial n/\partial \mu|_{E_F}$ found in theoretical calculations for the Be band-structure [21]. These calculations assumed an ideal Be crystal which might not be relevant for the disordered structure. It is not uncommon however that band-structure features calculated for the perfect crystal persist in the disordered material (as actually observed for some optical properties of Be [22]). The magnitude of the slope depends also on the film resistance as is shown for both $\text{In}_2\text{O}_{3-x}$ and Be in the insert to Fig.6. Being a low density system, $\frac{\partial}{\partial E}(\partial n/\partial \mu)_{E_F}$ in $\text{In}_2\text{O}_{3-x}$ is much larger than in metals, which in turn makes $\partial G(V_g)/\partial V_g$ larger.

It is interesting to note that the currently known electronic systems that exhibit intrinsic glassiness (with associated long relaxation times) are quite diverse in most other aspects. For example, in terms of microstructure, there are in this group representatives of all types of disordered structures; poly-crystalline ($\text{In}_2\text{O}_{3-x}$, and Be), granular or discontinuous (Al, Pb, Au, Ni), and amorphous (In_xO , Bi). Most of these systems contain oxide, whether as an intrinsic part of the material

($\text{In}_2\text{O}_{3-x}\text{In}_x\text{O}$), or to stabilize a granular structure (e.g., Al). However, the discontinuous Ni films, being prepared and measured under high-vacuum conditions [8], is oxygen-free and show the same effects as the other intrinsic electron-glasses [8]. In some of these systems there may be local order due to superconductivity (Pb, Bi, the high n version of In_xO , Be, Al) or magnetism (Ni) at the temperatures of the experiments but not in others. Finally, all these systems obey some form of activated conductivity, $G(T) \propto \exp[-(\frac{T_0}{T})^\alpha]$ with $0.3 < \alpha < 1$, however, no single value of α is singled out in the group. In other words, the conductivity versus temperature law $G(T)$ exhibited by these systems is not due to a specific hopping mechanism. It does not set this group apart from other hopping systems that do not exhibit long relaxation times. Indeed, a $G(T)$ law that resembles is observed in many disordered semiconductors while their relaxation times are very short [23] (presumably due to their low n [5]).

In fact, the *only* common feature of the materials that show long relaxation times appears to be their relatively high carrier-concentration (in addition of course to being strongly-localized thus exhibiting hopping conductivity). The common, out-of-equilibrium features that all these electron-glasses exhibit are suggestive of a generic mechanism. The current work supports the conjecture that sluggish relaxation and the associated memory effects of the electron-glass is intimately connected with high carrier-concentration [5]. This should be the pivotal point of any theoretical model that purports to account for these phenomena.

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