

Glassy Dynamics in Disordered Electronic Systems Reveal Striking Thermal Memory Effects

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Memory is one of the unique qualities of a glassy system. The relaxation of a glass to equilibrium contains information on the sample's excitation history, an effect often referred to as "aging." We demonstrate that under the right conditions a glass can also possess a different type of memory. We study the conductance relaxation of electron glasses that are fabricated at low temperatures. Remarkably, the dynamics are found to depend not only on the ambient measurement temperature but also on the maximum temperature to which the system was exposed. Hence the system "remembers" its highest temperature. This effect may be qualitatively understood in terms of energy barriers and local minima in configuration space and therefore may be a general property of the glass state.

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A glassy system is often characterized by a typical glass temperature T_G , below which its dynamics slow down dramatically. The sluggishness is ascribed to multiple local minima separated by energy barriers in configuration space. At high temperatures the system can explore the entire configuration space and thus it is ergodic. As the temperature is lowered below T_G the system is trapped in a subsystem of metastable states and it is no longer ergodic. The slow relaxation of glasses to equilibrium occurs via many-body transitions through the local minima at which the glass is trapped for a relatively long time before it manages to cross a barrier and move to another state. For glasses driven by thermal fluctuations (as opposed to quantum tunneling), overcoming a barrier is controlled by thermal activation, leading to a characteristic relaxation rate, λ , which decreases sharply as the temperature is lowered. For a class of glasses known as "strong," this is described by Arrhenius behavior, $\lambda \propto \exp\{-U/kT\}$, U being a typical barrier height of the order of T_G [1]. Fragile glasses manifest an even sharper temperature dependence.

This Letter deals with the situation where a glass never had the chance to visit the entire configuration space. What is the temperature dependence of the dynamics if the system is prepared in a glassy state and is "born" nonergodic? We investigate the dynamics of electron-glass systems that are fabricated using a special technique that enables the preparation of glassy systems at cryogenically low temperatures. Our main result is that under these conditions the dynamics depend not only on the working temperature but also on the maximal temperature that was experienced by the glass, T_{\max} . Hence, the dynamics of the system holds information on the glass temperature history.

The "electron glass" (EG) is a relative newcomer to the family of glasses. Electronic properties of strongly

disordered interacting systems (Anderson insulators) exhibit glassy behavior such as slow relaxation of the conductance G to equilibrium, memory effects, and aging (for reviews see Refs. [2,3]). These phenomena were predicted theoretically several decades ago [4–8] and were termed the electron glass [7] because the glassy properties are attributed to the conduction electrons. Experimentally, a growing number of systems have been reported to show such glassy behavior including discontinuous Au [9,10], amorphous and polycrystalline indium oxide films [11–15], ultrathin Pb film [16], granular aluminum [17,18], thin beryllium films [19], NbSi [20], Tl_2O_{3-x} [21], GeSbTe [22], and discontinuous films of Ag, Al, and Ni [10,23]. The conductance in these systems was shown to decay logarithmically with time after an abrupt cooldown or an electrical excitation out of equilibrium,

$$G(t) \propto G_0 - S \log(t), \quad (1)$$

where G_0 is the conductance at $t = 1$ sec just after the excitation. Such logarithmic slow relaxations over several decades in time have been reported in many glassy systems such as flux relaxation in superconductors [24], volume of crumpling paper [25], spin glasses [26], and porous silicon [27].

The samples studied in this Letter are discontinuous metal films that have been shown to exhibit glassy dynamics [10]. A major advantage of these samples is that they can be fabricated using a technique called "quench condensation" by which a thin metal film is evaporated on a substrate that is held at cryogenic temperatures and ultrahigh vacuum as illustrated in Fig. 1. This technique allows one to fabricate samples at temperatures in the range 10–300 K [10,28]. Since even at $T = 300$ K the conductance exhibits slow

relaxation, one can assume that throughout the entire fabrication temperature range the system is in the glass state. Conductance was measured by the two-probe methods using either ac techniques (frequency lower than 11 Hz) or dc techniques without apparent difference between the two. A voltage was applied between the drain and the source electrodes while making sure that the sample kept within the regime of linear Ohmic response and the current was measured using a current amplifier. A more detailed description of the experimental system and measurement may be found elsewhere [10].

An effective way to excite an EG out of equilibrium is by applying a gate voltage V_g between a metallic plate and the sample, which are arranged in a MOSFET configuration (see Fig. 1). A change of V_g instantly alters the underlying potential, introduces or depletes electrons and thus pushes the system out of equilibrium. This causes the conductance to abruptly increase by an amount denoted ΔG_0 , followed by a slow logarithmic relaxation towards equilibrium. ΔG_0 is a measure of how much the conductance is affected by the energy excitation as will be discussed below. Hence, in the remainder of this Letter the conductance is scaled by ΔG_0 ; consequently, the slope will be also presented in the normalized units

$$s \equiv \mathbb{S}/\Delta G_0. \quad (2)$$

Studying the temperature dependence of the relaxation of the conductance as a consequence of such an excitation reveals a striking effect. The dynamics are found to be strongly affected by the maximal temperature at which the sample was allowed to equilibrate. Figure 2(a) shows the conductance as a function of time following an abrupt V_g change for a discontinuous Au film that was prepared at $T = 140$ K. A clear logarithmic slow relaxation of the conductance is observed, consistent with Eq. (1). Heating the sample to 170 K for ~ 1600 sec and cooling back down has a relatively small ($\sim 20\%$) effect on the resistance, indicating that there is no big change in the microstructure; however, the dynamics is found to slow down dramatically. The slope of the logarithmic curve is found to decrease by a factor of 3 as a result of the heating-cooling cycle. Repeating this process has no further effect on the dynamics demonstrating that it is the first heating to $T = 170$ K that has the remarkable influence on the relaxation.

A related finding is shown in Fig. 2(c) which depicts the conductance relaxation of a series of samples which were heated to various maximal temperatures T_{\max} for relatively long periods, and cooled back to $T = 130$ K such that the resistance $R_{130\text{ K}} \sim 100\text{ M}\Omega$ for all samples. It is seen that

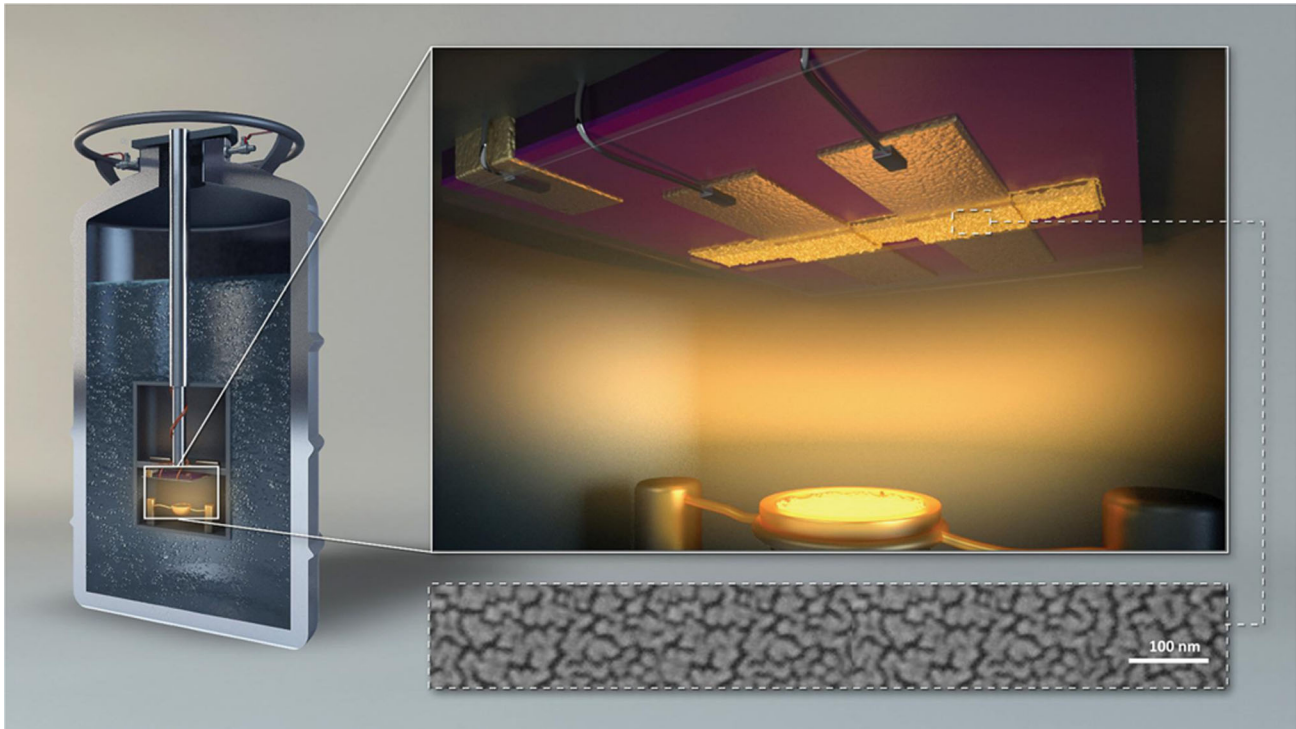


FIG. 1. A Si/SiO substrate with preprepared Au contacts is placed on a sample holder (in gray) situated in a high vacuum measuring probe which is immersed into a liquid He bath. Thin films are evaporated through a shadow mask (transparent layer in the sketch) and condense on the SiO substrate (pink) at cryogenic temperatures. A voltage source is connected between the Si layer which acts as a metallic gate (purple) and the sample for application of a gate voltage (far left electrode). Source drain voltage across the sample is applied for conductivity measurement. Bottom: A scanning electron microscope image section of a typical quench-condensed Au sample taken at $T = 300$ K, illustrating the discontinuous nature of the film. The quenched disorder in the geometry, combined with the long-ranged Coulomb interactions, leads to frustration which is at the heart of the glassiness of this system.

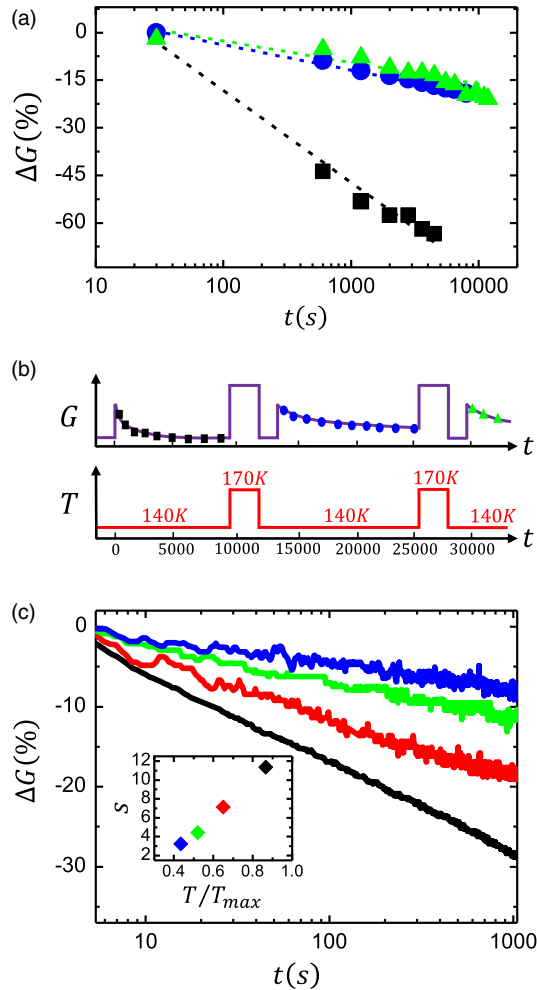


FIG. 2. (a) $\Delta G = [G(t) - G_0]/\Delta G_0$ as a function of time for a discontinuous Au film which was excited out of equilibrium at 140 K. Black squares are the initial curve in which $T = T_{\max} = 140$ K ($R = 2M\Omega$). Blue circles depict the conductance versus time measured after the sample was heated to 170 K and cooled back to 140 K, thus defining $T_{\max} = 170$ K ($R = 2.4M\Omega$). Green triangles are the results after a second heating-cooling cycle. The experimental protocol is illustrated in panel (b). (c) Conductance as a function of time measured at $T = 130$ K for four discontinuous Au films having T_{\max} of 150 (bottom black line), 200 (red line), 250 (green line), and 300 K (top blue line). For all samples the resistance at $T = 130$ K was $\sim 100M\Omega$. Inset: The slope of the $G(t)$ curves ($s = S/\Delta G_0$) as a function of T/T_{\max} for the four samples demonstrating the dependence of Eq. (3).

the slope of these curves s increases monotonically with T_{\max} and can be approximately described by

$$s \propto \frac{T}{T_{\max}}, \quad (3)$$

as illustrated in the inset.

Apparently the system remembers that it was exposed to the temperature T_{\max} . The higher this temperature, the slower the dynamics are at $T < T_{\max}$. Evidently, the highest

temperature experienced by the sample is encrypted in the dynamics of the system.

This behavior is seen for samples spanning a large range of fabrication temperatures. The dependence of s on the measurement temperature, T , for three discontinuous Au films prepared at different temperatures and accordingly having different T_{\max} is seen in Fig. 3. Once again, the dependence of the dynamics on both T and T_{\max} is evident. For all three samples $s \approx 7$ at $T = T_{\max}$ and it decreases considerably as T is lowered. The slope is found to be a function of T/T_{\max} and roughly follows Eq. (3). Similar $s(T)$ curves to those shown in Fig. 3 were obtained for over 20 Au samples with resistances ranging between a few kohms to a few hundred Mohms and sizes ranging from $20 \times 50 \mu\text{m}$ to 7×7 mm. About half of them were fabricated at room temperature and were exposed to air prior to the measurement (3 such samples are presented in the Supplemental Material [29]) and the rest had various lower T_{\max} . We observed similar results on 4 quench condensed Ni films and 2 Al films. Similar to other EGs [2,3], we observe no dependence of s on cooling rate within the experimental abilities, in contrast to other types of glasses. It is important to note that a film grown on a cryocooled substrate are amorphous while samples that were fabricated or heated to room temperature may indeed be crystalline [30]. However, we did not observe any difference in the results obtained on different samples that were prepared at different temperatures. This is consistent with the conjecture that the conductance is governed by hopping between islands and not by the atomic order in the islands.

The fact that the dynamics depend strongly on T is naturally understood in terms of energy scales of the

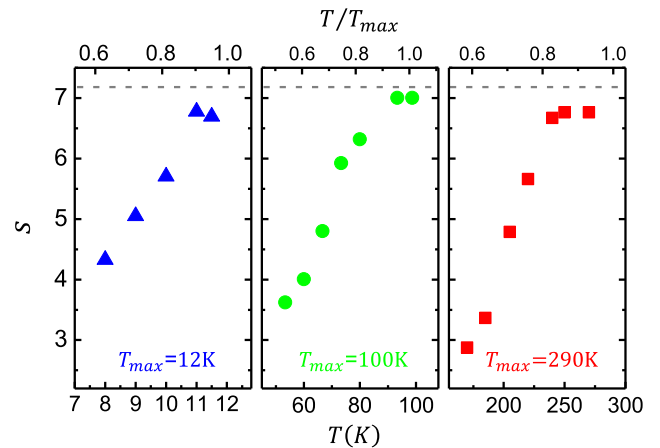


FIG. 3. The conductance relaxation slope s as a function of the temperature T and of T/T_{\max} for three discontinuous Au films having T_{\max} of 12, 100, and 290 K from left to right, respectively. All samples were allowed to equilibrate at T_{\max} for a few hours and then were cooled down to T , just before the measurement. The dashed line marks the experimental limit at large s which is set by the measurement protocol [2,31].

potential landscape versus thermal energy. But how can one understand the dependence on T_{\max} ? In the following we suggest an explanation that may be generic to glassy systems prepared at temperatures at which the system is already glassy.

A general property of glasses is that the presence of interactions may cause the system to be trapped in metastable states in configuration space. The energy barriers between these states have a wide distribution, which leads to a slow relaxation process toward equilibrium over many orders of magnitude. Heating the glass to higher temperature exposes the system to an even wider distribution of barriers, and consequently causes the relaxation process to become slower after the sample is cooled back to the lower temperature.

Assuming thermal activation as the relevant process driving the relaxations and an approximately uniform density of barriers \mathcal{D} in the interval (U_{\min}, U_{\max}) , the distribution of relaxation rates is given by [32,2]

$$P(\lambda) = \mathcal{D} \frac{dU}{d\lambda} = \frac{\mathcal{D} \cdot T}{U_{\max} - U_{\min}} 1/\lambda \equiv C/\lambda. \quad (4)$$

The dynamics are governed by many simultaneous processes, which leads to a logarithmic form of relaxations since

$$\sum_{\lambda} e^{-\lambda t} = \int_{\lambda_{\min}}^{\lambda_{\max}} e^{-\lambda t} P(\lambda) d\lambda \approx C[-\gamma_E - \log(\lambda_{\min} t)], \quad (5)$$

with γ_E the Euler-Mascheroni constant, and λ_{\min} and λ_{\max} the lower and upper cutoffs of the relaxation rate distribution [33]. As the system relaxes to equilibrium, each relaxation lowers the conductance by an amount δ_G , i.e., $G(t) \propto -\delta_G C \log(t)$, where δ_G may also depend on the temperature. In order to eliminate this nonuniversal dependence it is sensible to normalize the slope by ΔG_0 since

$$\Delta G_0 = \int_{\lambda_{\min}}^{\lambda_{\max}} \delta_G P(\lambda) d\lambda = C \delta_G \log\left(\frac{\lambda_{\max}}{\lambda_{\min}}\right) \approx C \delta_G U/T. \quad (6)$$

Thus, we find that the normalized conductance relaxation $G(t)/\Delta G_0$ scales logarithmically, with a slope proportional to T/U .

The experimental results indicate that for our systems, which were prepared at low temperature and therefore never had the chance to explore large parts of the configuration space, the relevant energy scale of the barriers is $U \propto T_{\max}$; hence, the distribution of relaxation depends on T_{\max} . Since thermal activation dominates the relaxation processes also at the lower temperature T , we obtain a logarithmic relaxation with a slope: $s_{\text{new}} = W(T/T_{\max})$, as indeed depicted in Figs. 2 and 3 and expressed in Eq. (3).

The factor W depends on the waiting time at $T = T_{\max}$, which was kept constant through the measurements.

An intuitive way to understand this result is to consider a simple potential landscape in the phase space illustrated in Fig. 4 undergoing the following sequence of events: (a) The system is prepared at temperature T_1 . Here the electrons are limited to exploring potential wells in configuration space that are separated by barriers U where $U \propto k_B T_1$. (b) The system is excited by a gate voltage change at $t = 0$, thus modifying the underlying electronic potential landscape. The relaxation is dominated by barriers whose magnitude is set by $k_B T_1$. (c) The system is heated to $T_{\max} = T_2 > T_1$. This enables electrons to enter deeper wells separated by higher barriers proportional to $k_B T_{\max}$. Upon a sequential cool down, the system is frozen in regions of phase space with larger potential wells. (d) After excitation of the system the relaxation is now dominated by barriers whose magnitude is set by $k_B T_{\max}$ and the dynamics slow down considerably.

Conductance dynamics in electron glasses was argued to be driven by ‘‘intrinsic’’ effects, i.e., processes that are governed by the conduction electrons [19,21,34]. Yet, one may wonder whether the above temperature dependence is related to ‘‘extrinsic’’ processes since heating the sample may effect the microstructure of the films. The experimental findings, however, show that morphology change does not affect the temperature dependence of the dynamics. The Supplemental Material [29] shows 3 discontinuous Au films having the same T_{\max} (300 K) but very different geometries (due to different preparation conditions) as clear

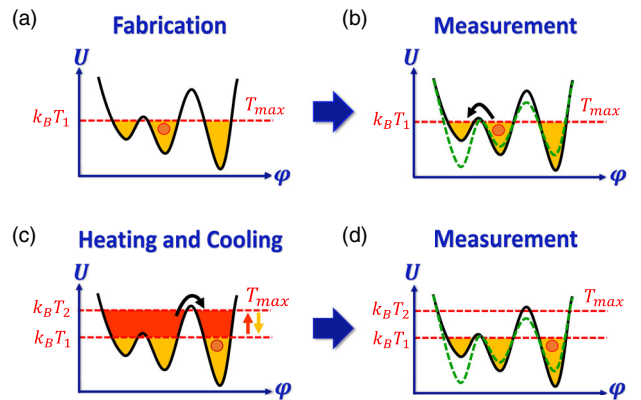


FIG. 4. A sketch of a section of the potential landscape in phase space. The black solid lines describe the landscape in the initial state. Applying a gate voltage changes this potential line (green dashed lines) and ignites a relaxation process. For all steps the system tends to minimize its free energy, i.e., to thermally equilibrate to the configuration with the lowest possible potential. At the first stage [(a) and (b)] the accessible energy is bounded by barriers of the order of the fabrication temperature T_1 . Heating to a new maximal temperature T_2 results in the system exploring deeper potential wells and, hence, characterized by slower relaxation processes after recooling to T_1 , as discussed in the main text [(c) and (d)].

from the scanning electron microscope scans of the three samples. Despite the significantly different geometries, all samples exhibit similar temperature dependence of the glass dynamic, demonstrating that the conductance sluggishness is not influenced by changes in the microstructure but only by T_{\max} .

Naturally, temperature is a crucial quantity in determining the dynamics of a glass. In recent years a number of nontrivial effects of temperature have been reported such as rejuvenation memory [35,36], temperature chaos [37,38], cooling rate memory effects [39,40], and possible cooling induced sample maturing. The dependence of the glassy properties on the highest temperature T_{\max} presented in this Letter is a novel, intriguing temperature effect which demonstrates that the dynamics of a glass can reveal the glass's highest temperature. This is made possible by the use of the quench condensation technique that enables the preparation of samples at temperatures at which the system is already glassy. This unique memory may be a general feature of glasses prepared at $T < T_G$ for which the sample could only explore a limited fraction of the configuration space. In a sense, T_{\max} takes the role of an effective T_G in controlling the relaxation processes of the glass. It would be interesting to explore additional glasses to find the regime of applicability of this novel phenomenon.

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