A quench-cooling procedure compared with the gate-protocol for aging experiments in the Electron-Glass

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Abstract

Anderson-insulating indium-oxide films excited far from equilibrium exhibit a variety of memory effects including aging. Full-aging has been recently demonstrated in this system using two different experimental protocols. The first, (gate-protocol) employed a MOSFET structure and involved switching between two gate voltages. In a different procedure, the system was subjected to a non-ohmic longitudinal field \( F \) for a waiting-time \( t_w \), and the relaxation of \( G \) was monitored after the field was switched back to its linear response value. In this paper we describe yet another protocol that involves measuring the response of the system that has been ‘aged’ at some low temperature \( T \) of the quench-cooling protocol are pointed out. The results of aging experiments based on the better-controlled, gate-protocol performed with different systems are compared and discussed.

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I. INTRODUCTION

The combination of sufficiently strong disorder and interactions in a degenerate Fermi system may precipitate a glassy state in an electronic system. Such a scenario, referring to the state as ‘electron glass’, was offered in several theoretical papers two decades ago [1,2,3]. Further elaboration of the early models, including quantum effects, suggested that the glass phase occurs on the insulating side of the metal-insulator transition [4]. Experimental evidence for glassy effects in disordered electronic systems were reported in few cases [5,6,7,8]. The experiments showed several transport features that are typical of other glasses, e.g., slow relaxation and memory effects including aging. Aging is an intriguing memory effect common to many glassy systems [8]. The system is said to exhibit aging if the response (e.g., relaxation from some excitation) depends on the system history in addition to the time \( t \). That is in contrast to ergodic systems where the response depends only on \( t \). In electron glasses, this phenomenon may be observed in the relaxation of the excess conductance \( \Delta G(t) \) created by changing the gate voltage from a state at which it equilibrated for \( t_w \) to a state where it was at full equilibrium (employing a MOSFET structure) [9,10].

This will be referred to here as a ‘gate-protocol’. It turns out that the aging function \( \Delta G(t,w) \) in this case is just a function \( f(t/w) \) [9,10]. This so called ‘full’ aging behavior has been rarely observed in such a clean form in any other glassy system. Recently, another aging protocol was employed in Anderson-localized indium oxide films by “stressing” the sample with a large electric field during \( t_w \) (‘stress-aging-protocol’) [11]. The latter procedure is fundamentally different than the gate-protocol in that during the waiting time the system is excited as opposed to relaxed in the gate-protocol. Full aging was realized in this protocol as well. Although the relaxation laws of the two protocols were not identical, both show a \( \log(t) \) dependence for \( t \ll t_w \) and a slower relaxation for \( t \geq t_w \). This means that the signature of \( t_w \) is printed on the form of each relaxation curve (in addition of course to the its effect on the amplitude of \( \Delta G \), which is a necessary ingredient for the aging phenomenon to hold).

In this paper we describe yet another protocol to observe aging in an electron glass. Motivated by recent theoretical work that shows aging in the electron glass [12], the protocol involves quench-cooling the sample from a high temperature \( T_H \) to low temperature \( T_L \ll T_H \), and letting it partially relax for \( t_w \). The aging is tested by re-exciting the sample with a sudden change of the gate voltage, and the relaxation of the excess conductance \( \Delta G(t) \) is measured from this time onward (‘T-protocol’). Several weaknesses, inherent in the T-protocol are pointed out. Qualitatively however, the results are similar to these obtained with the gate-protocol and exhibit full-aging behavior; the ensuing relaxation \( G(t,w) \) could be cast as \( f(t/w) \) over a wide range of \( t_w \). Finally, we compare the aging function obtained with the more reliable gate-protocol with the trap model suggested by Bouchaud and Dean. The implications for models of aging in quantum glasses [13], where dynamics in phase space is controlled by tunneling rather than by thermal activation, are discussed.

II. EXPERIMENTAL

A. Sample preparation and measurement techniques

Several different batches of samples were used in the present study. All were thin films (50±2Å thick) of crystalline indium-oxide that were e-gun evaporated on 110μm cover glass. Sample size was typically 1x1 mm and their sheet resistance R\( \square \) were in the range 1.5-55 MΩ at 4.11K. Gold film (≈1000Å thick) was evaporated on the backside of the glass and served as the gate electrode. Conductivity of the samples was measured using a two terminal ac technique employing a 1211-ITHACO current source.

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B. Description of the aging protocols

The T-protocol used in the experiments described below involved the following steps. First, the sample, attached to a measuring probe along with a calibrated Ge thermometer, is raised above the 4He liquid level. The conductance of the sample and the reading of the Ge thermometer are continuously monitored in the process of warming the sample to a temperature $T_H$. During the time the sample dwells at $T_H$ the gate voltage $V_g$ is set to an initial value, usually $V_g^o = 0$. Care was taken to ensure that the sample is always in helium atmosphere to minimize irreversible changes of the sample during temperature cycling. With this setup, warming up the sample and settling its temperature at $T_H$ usually took 3-10 minutes. The reverse operation, cooling the sample from $T_H$ to 4.11K could be accomplished in 3-4 seconds. As will become clear below, this stage of the protocol is critically important for an aging experiment of the T-protocol type. The sample conductance $G$ was then plotted as function of the time that elapsed from the moment the Ge thermometer reached 4.11K. Such a plot is shown in figure 1 for a typical run. Note that, after the quench, $G$ decreases logarithmically. This is the natural (‘history-free’) relaxation law of the electron glass; as will be shown below it may persist for almost six decades in time. In this protocol, however, the sample is allowed to relax only for $t_w$, at which time the gate voltage is quickly changed (typically over 5 seconds) from $V_g^o$ to $V_g^n$. This causes $G$ to increase, and then, while $V_g$ is held constant, it relaxes again towards a new equilibrium value set by $V_g^n$ and the bath temperature (c.f., figure 1). Finally, the time dependence of $G$ is recorded for a time span that extends at least three times $t_w$. The same procedure is then repeated with exactly the same set of parameters ($T_H$, $V_g^o$, $V_g^n$, and the changeover time of $V_g^o$ to $V_g^n$) except by using each time a different value for $t_w$. The set of plots $G(t,t_w)$, where $t$ is measured with respect to the time at which $V_g$ settled at $V_g^n$, is used as the data for the analysis of the aging behavior as detailed in the next section.

The gate protocol (more fully described e.g., in reference 9) starts after the sample equilibrated at $T_L$ under $V_g^o$. Then, $V_g$ is switched to $V_g^n$ and allowed to relax for $t_w$. Finally, $V_g^n$ is re-instated, and the excess conductance that ensues (relative to the equilibrium value of $G$) is plotted versus time $t$. A constant bath temperature is maintained for the entire process.

III. RESULTS AND DISCUSSION

The results reported here were reproduced on two different batches of samples. In the following however, we describe in detail the results on a single sample where extensive measurements were done, and where the conductance at a given temperature was least affected by the repeated temperature cycling. For the sample shown here the variance of $G$ at $T = 4.11K$ was within 1% for the entire series involving the cycling between $T_H$ and 4.11K. This is an impressive figure considering that, for the sample used here, $G$ changes by two orders of magnitude in this temperature range. The set of data that resulted from using the T-protocol with six different waiting times and with $T_H \approx 100K \pm 5K$ is plotted in figure 2. The quantity $\Delta G(t)$ plotted in the figure is the excess conductance affected by switching of $V_g^o$ to $V_g^n$. Namely, $\Delta G(t) = G(t) - G_b$ where $G(t)$ is the conductance versus time starting from the moment $V_g$ attained the target value $V_g^n$, and $G_b$ is the ‘background conductance’, a time dependent quantity reflecting the relaxation of $G$ following the quench. This subtraction also compensates for the ±1% variation in the value of $G$ immediately after a quench in different runs. $G_b$ is a function of both time and $V_g^o$ and is obtained by extrapolating towards the end-point of each run along the $\log(t)$ curve defined.
by the $t_w$ relaxation (the curve recorded during the time interval marked by $G(t_0)$ and $G(t_w)$ in figure 1, for example).

Before discussing the results, the reasons for choosing the various parameters, in particular the $100K$ for $T_H$, should be mentioned. A low temperature for $T_H$ is beneficial in reducing the quench time and the risk of irreversible sample change due to the thermal cycling. There is however a concern that favors a relatively high $T_H$ in this experiments, and it is of a rather fundamental nature. In addition to slow relaxation, a common feature of all glasses is a slow approach to a steady state; For example, applying a large voltage (field) across an electron-glass results in a time dependent increase of the conductance, a process that may last for days [11, 14]. The same behavior is observed when the bath temperature is changed from $T_1$ to $T_2 > T_1$, and the effect is actually quite prominent as illustrated in figure 3 for a specific case. Clearly, this is a complicating feature that introduces additional parameters to control in an already involved procedure [15]. Fortunately, these (and other) non-ergodic effects become exponentially smaller with temperature [16] and they are practically absent in our samples at $T \geq 90−100K$. Using these temperatures for $T_H$ instantly removed all traces of memory from the system, which made the ensuing results independent of the dwell time at $T_H$.

It is essential that the quench-cooling process will be fairly fast and smooth. If during the cooling process the sample is allowed to spend time at an intermediate temperature $T_i$ (in the range where non-ergodic effects are noticeable), a signature of this $T_i$ will appear at the ensuing relaxation. This is one of the weaknesses of all protocols based on quench cooling. Another problem that may actually become more severe when the cooling process is fast is the appearance of extra noise in the measurement after the quench. This takes the form of sudden bursts of fluctuations, with intermittent periods of activity that subsides after few hours. The problem presumably results from release of mechanical energy due to strains created in the process of quench-cooling the substrate/sample structure. Repeated thermal cycling from temperatures somewhat higher than the intended $T_H$ (‘training’) usually helped to minimize this disturbance.

The other important parameter in the experiment is $\delta V_g = |V_{g}^{n} - V_{g}^{o}|$ for which we chose $100V$ as a value large enough to have a sizeable re-excitation and small enough not to destroy the memory of the system [13, 17].

Figure 4 shows the respective outcome of the gate-protocol for the same sample for comparison. The results of the two aging protocols exhibit several common features. In both cases, the initial amplitude increases with
The process of the logarithmic conductance goes down along the dashed arrow in figure 5. This is the asymptotic value of $\Delta G$ changing associated with the equilibrium field-effect, which is due to $2b)$. This however is merely the excess conductance as- 

sociated with the equilibrium field-effect to $G$ when $V_g$ is switched to $V_g^n = 100V$. Namely, $V_g^o = 0V$, and $V_g^n = 100V$. Same sample as in figures 1 and 2.

The quality of data collapse is almost as good in the T-protocol as in the gate-protocol except for the region $t \gg t_w$ where the scatter is more noticeable. The main source of error in T-protocol is presumably the uncertainty in the extrapolated function of $G_b$, which naturally is more pronounced for large $t/t_w$. The best one can do is to correct for the effect of variations in the bath temperature during the relaxation process. In the current case, temperature variations was apparently not the only reason for the imperfect collapse for the $t \gg t_w$ regime. A probable cause is the presence of some burst

FIG. 5: The memory cusp as recorded in a field effect plot $G(V_g)$ (for the sample in figures 1 and 2) short time (approximately 40 sec.) after the quench from 100K (squares), and after 12 days (circles). The former trace was taken by first sweeping $V_g$ to +100V then recording $G(V_g)$ by continuously sweeping to −100V with 8V/sec. sweep rate. The ‘$t \rightarrow \infty$’ trace was taken by sweeping from $V_g$ to either extreme with 4V/sec. sweep rate. The dotted straight line connecting the end points of $G(V_g)$ is the field effect that is presumed to be recorded if measured instantly after the quench.
FIG. 6: The conductance $G$ versus time following a quench from $100K$ to $4.11K$ (circles) and the initial amplitude for the relaxation $\Delta G(t_0)$ following the switch from $V_g^0 = 0V$ to $V_g^n = 100V$ for the different waiting times in figure 2.

noise discussed above.

While the T-protocol has a number of weaknesses as an experimental procedure, it also has an advantage over the aging protocols previously used. In both, gate-protocol and stress-protocol the sample is initially at equilibrium, and $t_w$ is the time it spends in an out of equilibrium state. This requires that for each $t_w$ one has to wait for the system to come to equilibrium, which, as may be inferred from figure 6 is a long wait. In the T-protocol, on the other hand, the starting point is an ergodic state, so initializing the system for the next run is a fast process. Also, this protocol seems the more natural way to probe ‘aging’ in the sense used in everyday’s life; the system starts out ”young”, is then ”aged” for $t_w$, and the response $\Delta G$ due to the $V_g^0 \rightarrow V_g^n$ change is a measure of how deeply the system has ”aged”. This is judged by the initial amplitude and by the temporal dependence of $\Delta G$.

As an experimental procedure however, the T-protocol is manifestly not as easy as the two previously used protocols. The gate protocol, in particular, is less ‘invasive’, much easier to control and implement, and therefore more reliable.

Aging in the most general form means that the response of the system (in our case the excess conductance $\Delta G$) is a function of both $t$ and $t_w$. It would appear that, to exhibit such a behavior, it is sufficient that the system has a wide spectrum of relaxation times. Indeed, the response due to the application of an exciting agent after $t_w$ will be composed of only those components that have already relaxed. Therefore it is natural to expect that the ensuing response will be both larger and include slower components as $t_w$ increases. Clearly, these are the two ingredients that characterize the aging phenomenon.

It is easy to show in the T-protocol that $\Delta G(0)$, the initial amplitude of $\Delta G$ depends on $t_w$ in a unique way. This comes naturally from the basic properties of the electron glass, namely the logarithmic relaxation, and the time evolution of the memory cusp. The log($t$) relaxation of $G$ following a quench from a high temperature is shown in figure 6 for the sample under study. Therefore, at time $t_w$ after the quench one finds:

$$G(t_w) = G(t_0) - a_0 \cdot \log(t_w) \quad (1)$$

where $t_0$ is the time resolution of the experiment (typically, $t_0 = 1$ sec.). When $V_g$ reaches $V_g^n$, the conductance increases to $G(V_g)$ (see figures 1 and 5 for the definition of the various parameters). The initial amplitude for the relaxation after the switch of $V_g$ is $\Delta G(0) \equiv G(V_g) - G(t_w)$ and with (1):

$$\Delta G(0) = G(V_g) - G(t_0) + a_0 \cdot \log(t_w) \quad (2)$$

The first two terms of the right hand side of (2) include $\delta G_{eq}$ the equilibrium field effect, which as discussed above is associated with the change with energy of the thermodynamic density of states. They also depend on the sweep rate of $V_g$, the value of $T_H$, and the cooling rate involved in the quench. The third term reflects the expected dependence on $t_w$; obviously the longer the system ages the more susceptible it becomes to re-excitation (‘rejuvenation’) i.e., a larger response will occur, simply because the excitation affects only those components that have already relaxed. The dependence of $\Delta G(0)$ on the waiting time for the six aging runs used in figure 2 is depicted in figure 6.

While these considerations account for aging in its general form, the data collapse upon scaling by $t/t_w$ illustrated in figures 2 and 4 cannot be explained just by using the above assumptions, and a more fundamental treatment appears to be needed. The heuristic model offered to account for full-aging in the gate protocol is much harder to justify for the T-protocol. The model involved two elements. The first is the logarithmic relaxation law. This was shown to follow from the basic features of hopping transport and it is supported by recent Monte-Carlo simulations. The second was an assumption of some symmetry inherent to the gate-protocol when $\delta V_g \equiv |V_g^n - V_g^0|$ is small enough. It was later found that full aging is still observed even when this symmetry is absent. The lack of symmetry is even more obvious in the stress-aging-protocol which nonetheless show very good data collapse. Moreover, the heuristic model could explain the data collapse only in the regime $t \ll t_w$ where the logarithmic law is obeyed while the master plots in all three protocols deviate from the logarithmic law for $t \gg 0.2t_w$. For the gate protocol the signal/noise is good enough to confirm full aging behavior at least up to $t \approx 2t_w$. For later times $\Delta G$ reaches quickly the noise level as illustrated in figure 7 and we cannot be sure that there is still a perfect collapse. Nevertheless, there seems to be a wide enough time interval over which full aging is observed in the $t \gg t_w$ regime, and it is not describable by a logarithmic time depen-
dence. The right model for full aging in the electron glass must be able to account for this regime as well as for the $t \ll t_w$ regime.

A model that gives full aging in both $t \ll t_w$ and $t \gg t_w$ regimes has been suggested by Bouchaud and Dean [22]. In their 'tree' model, the two-time response is given as: $1 - \sin(\pi x) \left( \frac{t}{t + t_w} \right)^y$ and $\frac{\sin(\pi x)}{x} \left( \frac{t}{t + t_w} \right)^x$ for $t < t_w$ and $t \gg t_w$, respectively and where $y \equiv 1 - x$. The data in figure 7 are plotted to allow comparison with the $t \gg t_w$ regime. Note that to be consistent with the model, the value of $x$ has to be about 0.87 (c.f., figure 7). This is the average value of the best-fit exponent found in eight aging experiments based on the gate-protocol. The values for $x$ in these runs ranged between 0.85 to 0.91. This constrains the value of $y$ to be 0.09 – 0.15 for the relaxation in the $t \gg t_w$ regime. To reconcile the experimental results (figure 6) with the model for the $t \ll t_w$ regime requires $y \leq 0.0085$, and given the scatter in the data of figure 7 we cannot rule out agreement with the model.

This agreement however may be fortuitous. The Bouchaud and Dean model assumes dynamics of a classical glass, where the barriers in phase space are crossed by thermal activation. This is not appropriate for the case of a quantum glass like the electron glass [4]. In particular, the value of the exponent $x$ in the model is given by $T/T_g$ where $T_g$ is a glass temperature [11]. This gives the aging function a specific temperature dependence, in contrast with the experiment [4]. On the other hand the overall resemblance of the model predictions to actual data suggest that the underlying approach may have merit for real systems. This might encourage researchers to look for a modification of the model that are more appropriate for a glass with tunneling controlled dynamics rather than by thermal activation.

Using the gate-protocol it was shown before [9] that the aging function fits well a stretched exponent expression: $\Delta G(t, t_w) \propto \exp \left[ -\beta \left( \frac{t}{t_w} \right)^\alpha \right]$ with $\beta \approx 2.75$ and $\alpha \approx 0.21$. This expression, with strikingly similar parameters $\alpha$ and $\beta$, were found to fit many aging experiments on both crystalline and amorphous indium-oxide films. The gate-protocol, recently applied to granular Al films, exhibited a near-perfect full-aging, and seem to fit quite well to the stretched-exponent expression above with $\beta \approx 2.8$ and $\alpha \approx 0.185$ [10]. Full-aging behavior was observed in experiments based on the stress-protocol on In$_2$O$_3$-x films, and the results could be fitted by the stretched-exponent with $\beta \approx 2.32$ and $\alpha \approx 0.215$ [14]. Finally, the aging function of the T-protocol could be fitted by a stretched exponent with $\beta \approx 2.65$ and $\alpha \approx 0.22$ (after subtracting 0.58-0.6 that represents $\delta G_{eq}$ from the data in figure 2b). It is important to note that these fits to the stretched exponent function should be merely regarded as a convenient classification scheme; To see that the stretched exponent cannot be the correct description of these experiments suffice is to notice that for $t \ll t_w$, the function reduces to a power-law dependence with exponent $\alpha$. A value for $\alpha$ of the order of 0.2 may reasonably mimic a $\log(t)$ dependence over a limited range, which is why this, in-principle discrepancy (c.f., figure 6), does not stand out in aging experiments where $t_w$ was restricted to less than 4 decades. What the similarity of parameters does tell us is that the aging function of several systems must have a similar shape. This is not a trivial observation. The studied systems; crystalline indium-oxide, several versions of amorphous indium-oxides [24], and granular Al are totally different in terms of microstructure, and they usually exhibit quite different conductance versus temperature laws $G(T)$ [24]. The feature that is common to all these systems is that they are strongly localized, which is the pre-requisite to be in the electron glass phase [4], and their $G(T)$ is consistent with hopping transport mechanism [24]. We suspect that the latter is an important ingredient in bringing about the 'unified' aging behavior; Namely, it is the logarithmic relaxation inherent to the hopping mode of transport [15] that is common to all of the full-aging examples listed above. Actually, the only difference between the results of different systems (and different protocols), is the value at which the aging function $f(t/t_w)$ deviate from the generic $\log(t)$ behavior. The challenge is to find the physical scheme that controls this aspect of the full-aging behavior.
The phenomenon.

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