

Quench-cooling procedure compared with the gate protocol for aging experiments in electron glasses

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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_L for a duration t_w after it was quench-cooled from high temperature T_H . As in the previous protocols, this procedure results in full-aging behavior. The advantages and shortcomings 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.^{1–3} Further elaboration of the early models, including quantum effects, suggested that the glassy phase occurs on the insulating side of the metal-insulator transition.⁴ Experimental evidence for glassy effects in disordered electronic systems were reported in a few cases.^{5–7} 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 back 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 $\Delta G(t, t_w)$ in this case is just a function f of t/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 imprinted on the form of each relaxation curve (in addition of course to the its effect on the *amplitude* of ΔG ,

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 $G(t, t_w)$ could be cast as $f(t/t_w)$ 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,⁴ 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 ± 2 Å thick) of crystalline indium-oxide that were e -gun evaporated on $110 \mu\text{m}$ cover glass. Sample size was typically 1×1 mm and their sheet resistance R_{\square} were in the range 1.5 – 55 M Ω at 4.11 K. Gold film (≈ 1000 Å 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 preamplifier and a PAR-124A lock-in amplifier. Except when otherwise noted, the measurements reported here were performed with the samples immersed in liquid helium at $T=4.11$ K held by a 100 liter storage-dewar, which allowed long term measurements of samples as well as a convenient way to maintain a

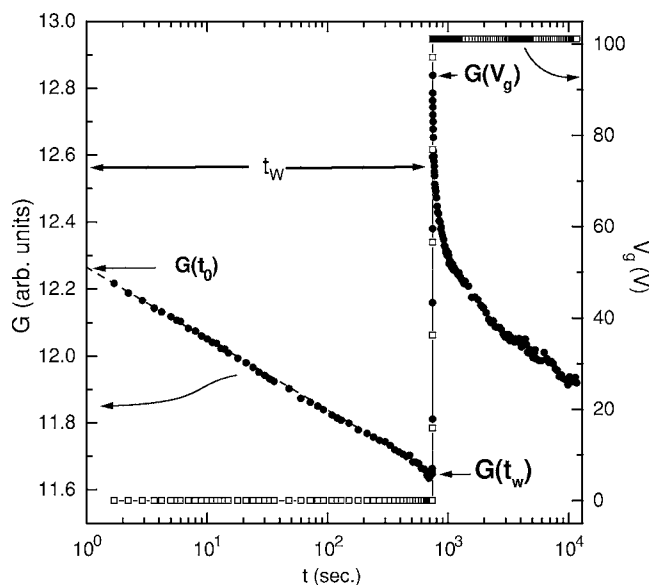


FIG. 1. A typical run of the T protocol. The data marked by full circles is the sample conductance (left y axis), and the squares stand for the corresponding values of the gate voltage (right y axis). Note the initial logarithmic relaxation, lasting $t_w=745$ s in this case. Sample resistance at $T=4.11$ K is 10.4 M Ω .

stable temperature bath. The ac voltage bias was small enough to ensure linear response conditions (judged by deviation from Ohm's law being within the experimental error). Fuller details of sample preparation, characterization, and measurements techniques are given elsewhere.¹³

B. Description of the aging protocols

The T protocol used in the experiments described below involved the following steps. First, the sample, attached at the end of a measuring probe along with a Ge thermometer, is raised above the ^4He liquid level. The conductance of the sample and the reading of the Ge resistor 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 min. The reverse operation, cooling the sample from T_H to 4.11 K could be accomplished in 3–4 s. 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 4.11 K. Such a plot is shown in Fig. 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 s) 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., Fig. 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 Ref. 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^o is reinstated, 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 $T=4.11$ K was within 1% for the entire series involving the cycling between T_H and 4.11 K. 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 $T_H \approx 100$ K \pm 5 K is plotted in Fig. 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, $\Delta G(t)$ is $G(t) - G_b$ 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 variations in the values of G of the different runs immediately after a quench. 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 $\ln(t)$ curve defined by the t_w relaxation [the curve recorded during the time interval marked by $G(t_0)$ and $G(t_w)$ in Fig. 1, for example].

Before discussing the results, the reasons for choosing the various parameters, in particular the 100 K 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 Fig. 3 for a specific case. Clearly, this is a complicating feature that introduces

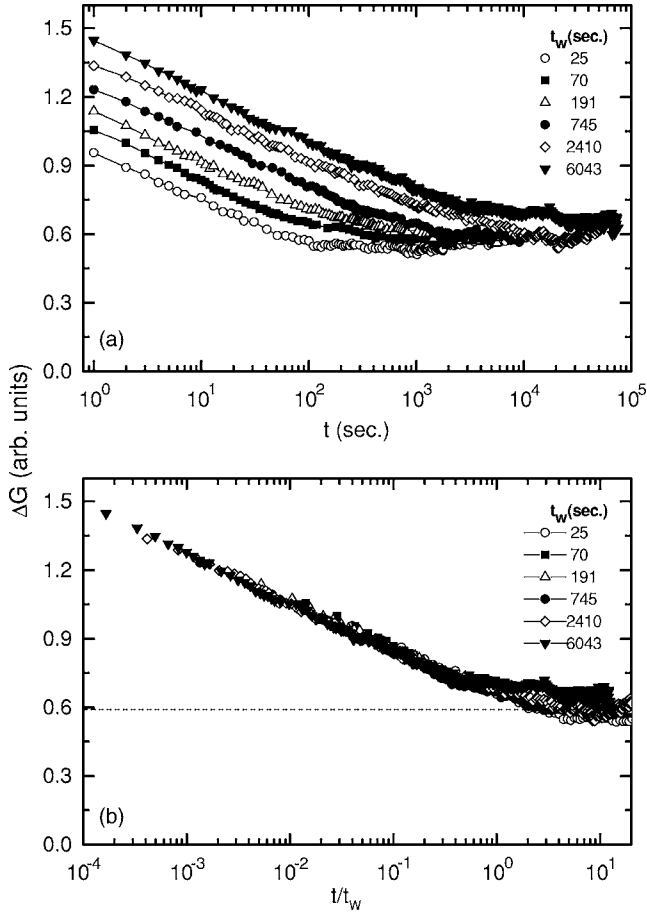


FIG. 2. The excess conductance ΔG (see text) as function of time (a), and time in units of t/t_w (b) for various waiting times. Same sample as in Fig. 1.

additional parameters to control in an already involved procedure.¹⁵ Fortunately, these (and other) nonergodic effects become exponentially smaller with temperature¹⁶ and they are practically absent in our samples at $T \geq 90$ –100 K. 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 nonergodic 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 a few hours. The problem presumably results from the release mechanical energy of trapped 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 100 V as a value large enough

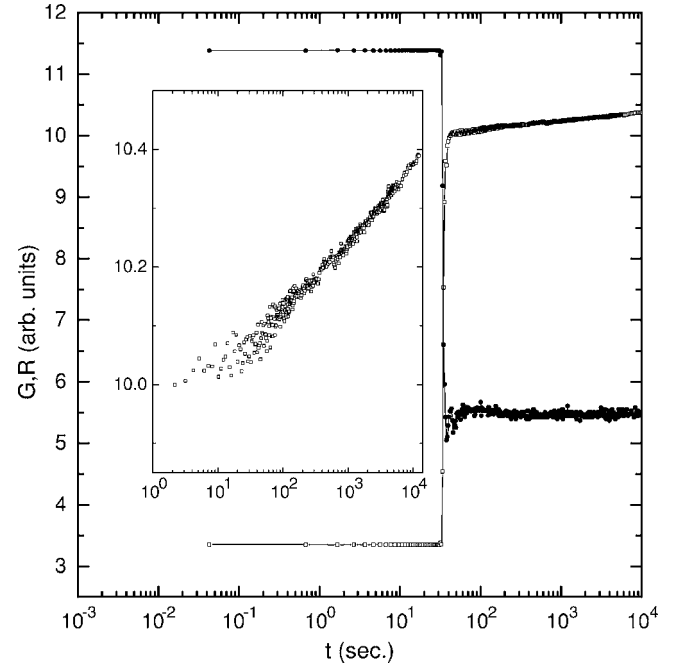


FIG. 3. The sample conductance G (squares) and the Ge thermometer resistance R (circles) during heating (started at $t \approx 35$ s) to $T=6.22$ K from an equilibrated state at $T=4.11$ K and at later times, when the system temperature has reached a constant value (for $t \geq 50$ s). Note (inset) that G continues to increase logarithmically long after the temperature has stabilized as judged by the Ge thermometer. The sample resistance is 1.42 M Ω at $T=4.11$ K.

to have a sizeable reexcitation 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 t_w , and the ensuing relaxation is more sluggish at any given t [compare Fig. 2(a) and 4(a)]. Also, upon normalizing ΔG by t_w measured in each case, the curves collapse onto a common master plot [Figs. 2(b) and 4(b)], and both master plots exhibit $-\ln(t)$ for $t \ll t_w$ and a slower dependence for $t \geq t_w$. A notable difference is the nonzero asymptotic value of $\Delta G(t/t_w)$ in the T protocol [Fig. 2(b)]. This, however, is merely the excess conductance associated with the equilibrium field effect, which is due to changing V_g^o to V_g^n (a positive change in this particular case). This can be appreciated with the help of Fig. 5 that shows the conductance versus gate voltage for two stages of the aging process. The figure shows the “memory cusp,” which is a specific modulation in the conductance versus gate-voltage plot $G(V_g)$. It is seen by sweeping the gate voltage over a range including V_g^o where the system is equilibrating. As noted elsewhere,¹³ the modulation that constitutes the cusp evolves with time during the equilibration process. Immediately after a quench from high temperature $G(V_g)$ shows just an antisymmetric field effect, which has essentially the same shape as the equilibrium result. With time, $G(V_g=0)$ goes down along the dashed arrow in Fig. 5, (which is the process of logarithmic relaxation observed in Fig. 6), revealing the characteristic shape of a cusp with a

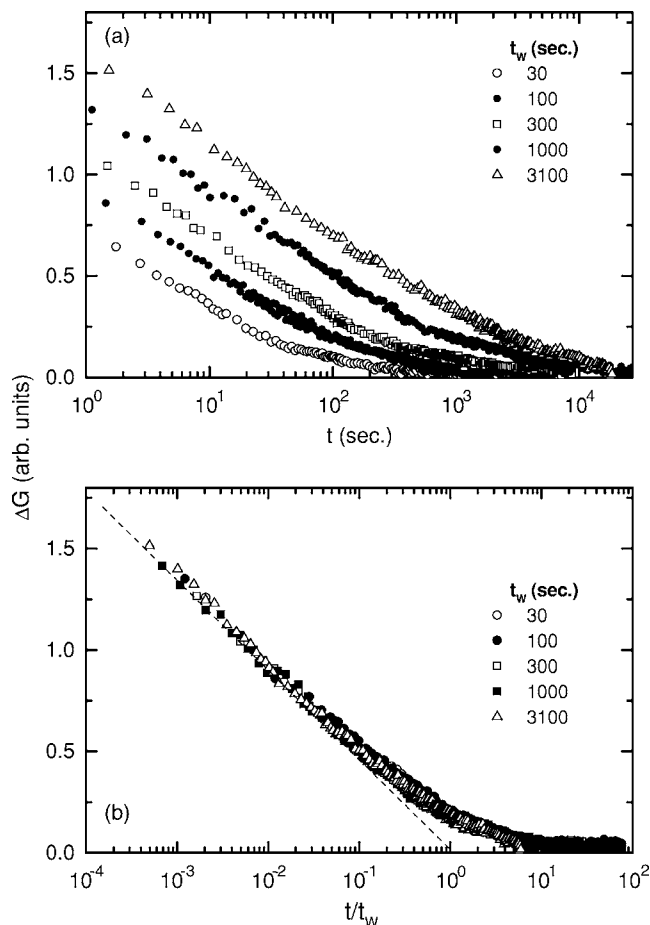


FIG. 4. Aging experiment using the gate-protocol with five identical values of waiting times t_w . The same values of V_g^o and V_g^n used in the T -protocol of Fig. 2 were employed. Namely, $V_g^o=0$ V and $V_g^n=100$ V. Same sample as in Figs. 1 and 2.

local minimum centered at $V_g^o=0$. A model accounting for such dynamics has been proposed by Yu.¹⁸

Note that the anti-symmetric part of $G(V_g)$ does not depend on the waiting time nor on the sweep rate of V_g .¹³ The contribution of the equilibrium field effect to G when V_g is switched to $V_g^n=100$ V from $V_g^o=0$ V is δG_{eq} and can be estimated from Fig. 5 as $\delta G_{eq} = \frac{G(V_g^n) - G(-V_g^n)}{2}$. [Alternatively, one may use $\delta G_{eq} = G(V_g^n) - G(t_0)$ from the $G(V_g=0, t)$ plot by extrapolating to the value of the conductance at t_0 along the well defined $\ln(t)$ dependence as in Fig. 6. The δG_{eq} obtained by these two ways coincide to within the experimental error]. This value of δG_{eq} is drawn in Fig. 2(b) as a dashed line, and is in fair agreement with the asymptotic value of $\Delta G(t/t_w)$.

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, which did not improve the collapse in the current case. A probable cause is the presence of some burst noise discussed above.

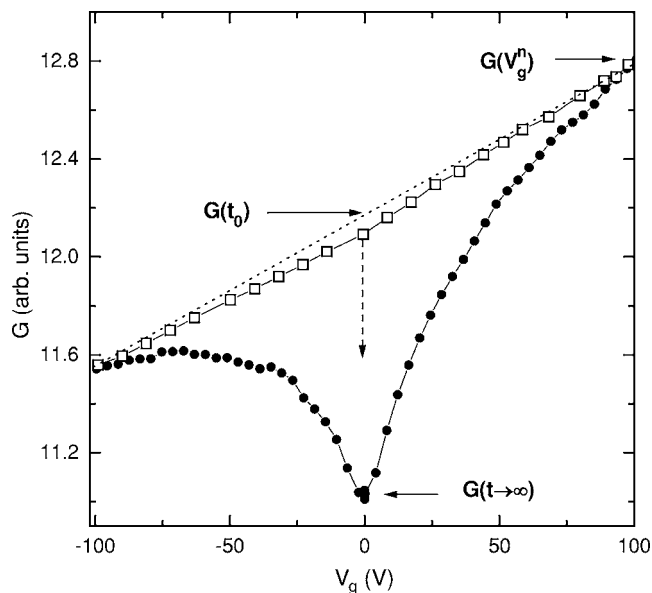


FIG. 5. The memory cusp as recorded in a field effect plot $G(V_g)$ (for the sample in Figs. 1 and 2) short time (approximately 40 s) after the quench from 100 K (squares) and after 12 days. The former trace was taken by first sweeping V_g to +100 V then recording $G(V_g)$ by continuously sweeping to -100 V with 8 V/s sweep rate. The “ $t \rightarrow \infty$ ” trace was taken by sweeping from V_g to either extreme with 4 V/s 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.

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 the stress protocol the sample is initially at equilibrium, and t_w is the time it spends in an out of equilibrium state. This

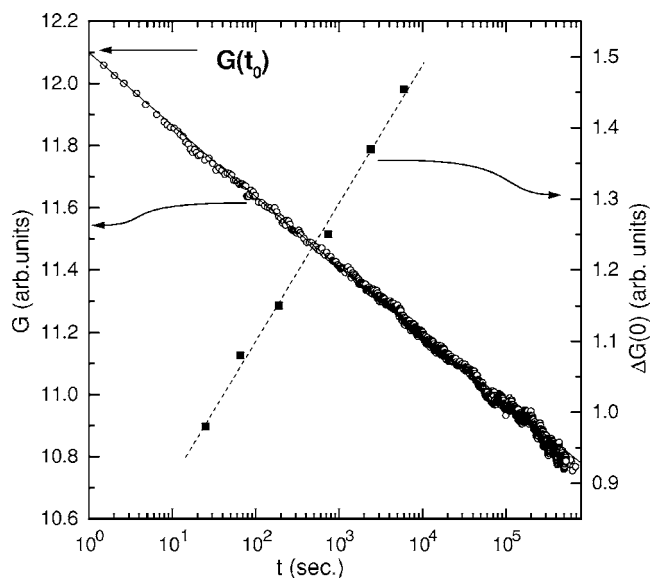


FIG. 6. The conductance G versus time following a quench from 100 to 4.11 K (circles) and the initial amplitude for the relaxation $\Delta G(t_0)$ following the switch from $V_g^o=0$ V to $V_g^n=100$ V for the different waiting times in Fig. 2.

requires that for each t_w one has to wait for the system to come to equilibrium, which, as may be inferred from Fig. 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 ΔG due to the $V_g^o \rightarrow V_g^n$ change is a measure of how deeply the system has “aged.” This is judged by the initial amplitude and by the temporal dependence of ΔG .

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 ΔG) 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 ΔG 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 $\ln(t)$ relaxation of G following a quench from a high temperature is shown in Fig. 6 for the sample under study. Therefore, at time t_w after the quench one finds

$$G(t_w) = G(t_0) - a_o \ln(t_w), \quad (1)$$

where t_0 is the time resolution of the experiment (typically, $t_0=1$ s). When V_g reaches V_g^n , the conductance increases to $G(V_g)$ (see Figs. 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 Eq. (1)

$$\Delta G(0) = G(V_g) - G(t_0) + a_o \ln(t_w). \quad (2)$$

The first two terms of the right-hand side of Eq. (2) include δG_{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 reexcitation (“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 Fig. 2 is depicted in Fig. 6.

While these considerations account for aging in its general form, the data collapse upon scaling by t/t_w illustrated in Figs. 2 and 4 cannot be explained just by using the above

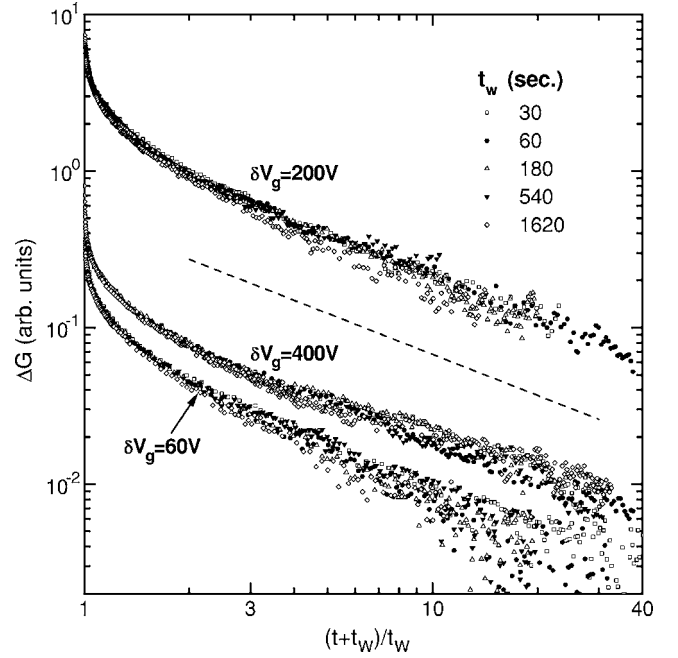


FIG. 7. The excess conductance ΔG in a gate-protocol experiment as function of the normalized time. The figure shows the results of three aging series each with five values of waiting times. These were taken with two different samples; The two bottom curves are data for a single 40.5 M Ω sample, where the two sets shown used different values of δV_g as indicated. The upper set was taken with a 51 M Ω sample with $\delta V_g = 200$ V. The latter data were shifted for clarity by multiplying ΔG by 10. Note that for $t \gg t_w$ the data is consistent with the power-law behavior predicted by Bouchaud and Dean. The dashed line illustrates a power law with exponent -0.87 for comparison.

assumptions, and a more fundamental treatment appears to be needed. The heuristic model offered to account for full-aging in the gate protocol¹⁹ 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 transport¹⁹ and it is supported by recent Monte Carlo simulations.²⁰ The second was an assumption of some symmetry inherent to the gate protocol when $\delta V_g \equiv |V_g^n - V_g^o|$ is small enough. It was later found that full aging is still observed even when this symmetry is absent.²¹ The lack of symmetry is even more obvious in the stress-aging-protocol which nonetheless show very good data collapse.¹¹ 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 $t \geq 0.2t_w$. 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 ΔG reaches quickly the noise level as illustrated in Fig. 7.

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 $1 - \frac{\sin(\pi x)}{\pi(1-x)} \left(\frac{t}{t+t_w}\right)^y$ and $\frac{\sin(\pi x)}{\pi x} \left(\frac{t_w}{t+t_w}\right)^x$ for $t \ll t_w$ and $t \gg t_w$, respectively, and where $y \equiv 1-x$. The data in Fig. 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 (cf., Fig. 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 (Fig. 6) with the model for the $t \ll t_w$ regime requires $y \leq 0.0085$, and given the scatter in the data of Fig. 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 such as 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.⁹ 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 before⁹ that the aging function fits well a stretched exponent expression $\Delta G(t, t_w) \propto \exp[-\beta(\frac{t}{t_w})^\alpha]$ with $\beta \approx 2.75$ and $\alpha \approx 0.21$. This expression, with strikingly similar parameters α and β , 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 $\text{In}_2\text{O}_{3-x}$ 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 sub-

tracting 0.58–0.6 that represents δG_{eq} from the data in Fig. 2(b)].

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 it to notice that for $t \ll t_w$ the function reduces to a power-law dependence with exponent α . A value for α of the order of 0.2 may reasonably mimic a $\ln(t)$ dependence over a limited range, which is why this, in-principle discrepancy (cf., Fig. 6), does not stand out in aging experiments where t_w was restricted to less than four 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,²³ 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 prerequisite 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 transport¹³ 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* $\ln(t)$ behavior. The challenge is to find the physical scheme that controls this aspect of the full-aging phenomenon.

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