Stress-aging in the electron-glass

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Abstract

A new protocol for an aging experiment is studied in the electron-glass phase of indium-oxide films. In this protocol, the sample is exposed to a non-ohmic electric field \( F \) for a waiting time \( t_w \) during which the system attempts to reach a steady state (rather than relax towards equilibrium). The relaxation of the excess conductance \( \Delta G \) after ohmic conditions are restored exhibit simple aging as long as \( F \) is not too large.

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Aging is a common phenomenon in non-equilibrium systems. The term ‘aging’ refers to a continuous change in the properties of the system when it is maintained in some fixed external conditions (such as temperature, pressure, etc.) for a waiting-time $t_w$. This change may be reflected in the dynamic response of the system due to an application of a post-aging disturbance. For example, the viscoelastic response of a polymer to a mechanical stress will depend on the time $t_w$ it was ‘aged’ at a temperature $T$ prior to applying the stress.

Systematic studies of various glassy systems revealed that aging might manifest itself in different measurements but all share a common feature: After the external conditions that affect a certain property $P$ are changed, $P$ relaxes towards its new equilibrium value in a way that reflects both the time $t$ and the ‘aging’ time $t_w$, namely, $P(t) = P(t, t_w)$.

A more specific form of aging called ‘simple-aging’ has been recently reported to occur in several glasses where $P(t, t_w)$ could be described as a simple master function $P(t/t_w)$.

The experimental protocol usually employed in aging studies involves relaxation towards an equilibrium state during $t_w$. In this note, we report on a different protocol where the system is under a constant stress $F$ and attempts to reach a steady state during $t_w$. It turns out that the relaxation that ensues after the stress is relieved exhibits simple aging as long as the stress is not too large. The master function $P(t/t_w)$ is affected by the particular magnitude of the stress, and above a certain field the relaxation curves fail to collapse. This is shown to correlate with the loss of memory in the system.

Our experiments were performed using thin films of crystalline $\text{In}_2\text{O}_{3-x}$ in the hopping regime. The response $P$ is taken as the conductance $G$, and the stress $F$ is the electric field applied along the film. Measurements were carried out at $T=4.11\text{K}$ with the samples immersed in liquid $^4\text{He}$ inside a storage dewar. This enabled high temperature stability over long times. A germanium thermometer mounted on the sample stage was used to correct for residual temperature fluctuations and drift. The conductance of the samples was measured using a two terminal configuration. For measurements with $F > 10 \text{ V/cm}$ $G$ was measured by a dc technique, biasing the sample with a voltage source (Keithley’s K617) while measuring the resulting current (the voltage across a $10^5 \Omega$ series resistor). This procedure was used during the stress application. For smaller values of $F$, we used ac techniques employing a current pre-amplifier (ITHACO 1211) and a lock-in amplifier (PAR 124). This was also used to measure the conductance $G$ before each run as well as for the relaxation after $F$ was reset to the ohmic regime ($F$ typically smaller than $1\text{V/cm}$).
The steps performed in this series of experiments, and results for a typical case are illustrated in figure 1. The sample conductance and the accompanying stress-field $F$ were monitored continuously versus time. Initially, $G(t)$ is recorded while keeping $F = F_0$ chosen to be in the ohmic regime (i.e., $\partial G / \partial F|_{F_0} \approx 0$) to establish a baseline ‘equilibrium-$G$’ = $G(F_0,0)$. Then, $F$ was switched to $F_n \gg F_0$ which caused an appreciable increase in conductance (figure 1). Finally, having recorded $G(F_n,t)$ for a time $t = t_w$, $F$ is switched back to its original value $F_0$. This results in an initial sharp decrease of $G$ followed by a slow relaxation process where the conductance decreases and asymptotically approaches $G(F_0,0)$ (the dashed line in figure 1). The relaxation of the excess conductance $\Delta G(t) = G(t \geq t_w) - G(F_0,0)$ is plotted in figure 2a where the origin of the time scale $t = 0$ is the time when $F_0$ was re-established. The same $\Delta G(t)$ curves are plotted in figure 2b as function of $t/t_w$ illustrating a near-perfect data collapse to a master function $\Delta G(t/t_w)$. It is emphasized that no free parameters are involved in this collapse; the only step taken to get the master function $\Delta G(t/t_w)$ is dividing each $\Delta G(t)$ curve by its measured $t_w$.

The master function that results from the present protocol (to be referred to as ‘$F$-protocol’) is quite similar to that of the aging protocol used by Vaknin et al [3] (‘$V_g$-protocol’). In both cases, $\Delta G(t/t_w) \propto -\log(t)$ for $t < t_w$ and both show equally good simple aging (compare figure 2b with figure 2 in reference 3). Note that these two protocols are fundamentally different. The $V_g$-protocol conforms to the commonly used procedure where during $t_w$ the system is relaxing as manifested by the fact that $\Delta G(t)$ is logarithmically decreasing function of $t$. By contrast, the system is excited during $t_w$ in the F-protocol, and the associated $\Delta G$ increases logarithmically with time (inset to figure 1). Note that the $V_g$-protocol is carried out under ohmic conditions throughout the entire process while strong non-ohmic conditions are used during $t_w$ in the F-protocol. During this time, the electronic system absorbs energy from $F$ and, as will be shown below, some memory of the system is impaired in result. It is therefore somewhat surprising that the F-protocol yields as good simple aging as the $V_g$ protocol. In fact, the only feature in the master function that reflects the difference between the two protocols is the extrapolated value for $t/t_w$ to $\Delta G(t/t_w) = 0$ (c.f., figures 2 and 3). In the $V_g$-protocol this happens at $t'/t_w \equiv t^*$ which is usually =1. This is due to a certain symmetry inherent to this protocol. When this symmetry is impaired e.g., by using large swings of gate voltages, this $t^*$ becomes larger than unity [7, 8]. In the F-protocol $t^*$ is consistently larger than unity and increases
systematically with $F_n$ reaching a value of $\approx 10$ (inset of figure 3) before the curves fail to collapse (figure 4). This incidentally means that over the range of fields where simple aging is observed, the master function carries a memory of both $t_w$ and the specific value of $F_n$ (namely, the value of $t^*$ for a given sample). The inset to figure 3 may be interpreted as implying that when $F_n \to 0$, $t^* \to 1$, which in other words is just saying that the sample is under “symmetrical” (i.e., Ohmic) conditions both during $t_w$ and throughout the subsequent relaxation process. Obviously, this situation cannot be realized in practice with the $F$-protocol.

When $F_n$ exceeds a certain value the $\Delta G(t)$ curves fail to collapse upon normalization by $t_w$ (figure 4). For still higher fields $\Delta G(t)$ becomes independent of $t_w$ and assumes the ‘history-free’ law $\Delta G(t) \propto -\log(t)$. This presumably results from the fact that a large $F_n$ has a similar (though not exactly equivalent) effect as that of raising the system temperature. Above some $F_n$, this effective temperature will bring the system to an ergodic state (above the ‘glass temperature’), and the ensuing relaxation upon the switch to $F_0$ should be similar to a quench-cool process. Namely, $\Delta G(t)$ should contain no memory of the past and aging behavior is lost as indeed observed.

The influence of the stress-field, and in particular, its detrimental effect on the memory of the electron glass, can be monitored in a field-effect experiment. This was performed using a sample configured as a FET structure by depositing a gate electrode (Au film) on the backside of the 100$\mu$m glass substrate. The sample was cooled to 4.11K holding its gate voltage $V_g$ at 0V, and was allowed it to relax at this temperature for $\approx 12$ hours. Then, while monitoring $G$ (using ac techniques), $V_g$ was swept to +100V, kept there for 15 seconds after which $V_g$ was swept to -100V. The resulting $G(V_g)$ curve (figure 5) revealed a memory of the cool-down-$V_g$ in the form of a minimum centered at $V_g=0$. After allowing the system to relax again under $V_g=0$, the procedure was repeated except that during 10 of the 15 seconds dwell-time at $V_g=0$, a non-ohmic dc field $F_n$ was applied between the source and drain. The $G(V_g)|F_n$ traces resulting from this procedure exhibit a “memory-cusp” that has a progressively smaller magnitude when $F_n$ is increased (c.f., figure 5). This illustrates the memory loss caused by the stress-field as alluded to above. Moreover, above a threshold $F_n$ the anomalous cusp at $V_g=0$ completely disappears, and $G(V_g)$ reflects just the normal (anti-symmetric) form of the field-effect. It is in this range of fields that the aging behavior is washed out.
In summary, we have demonstrated that the conductance of an electron glass carries a memory of a non-ohmic electric field $F$ applied in the past as well as its duration $t_w$. This information is reflected in the relaxation of the excess conductance $\Delta G(t)$ monitored following a switch of $F$ (at $t=0$) to its ohmic regime. It was also shown that the non-ohmic fields degrade the memory in the system and that simple aging is obeyed only as long as this memory loss is small. Our experiments thus illustrate that 'simple-aging' and 'memory' are inter-related properties of the electron glass.

Finally, it is remarkable that simple-aging is observed in many different systems (electron-glass, spin-glass, polymers, viscous-fluids). That such a simple scaling scheme should so generally hold is a challenge for theory. This seems to imply the existence of a common feature, non-specific to the type of glass being studied [10]. To our knowledge, this common ingredient is yet to be identified.

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Figure captions

1. The sample conductance $G$ versus time during a stress-aging experiment. $F_0=0.5V/cm$ is used except during $t_w$ where $F_n=95V/cm$ is maintained. $R\Box=230M\Omega$ at $T=4.11K$. The inset illustrates the logarithmic law by which $G$ increases under a constant $F_n$ (for $t_w \approx 5$ days and under $F_n=315V/cm$ in this example).

2. Relaxation curves of the excess conductance after an excitation by $F_n=100V/cm$ for different values of $t_w$ (a). Sample with $R\Box=57M\Omega$. The same data as in (a) is plotted in (b) versus $t/t_w$. The dashed line shows the extrapolated value of the logarithmic part of the master function to $\Delta G(t/t_w)=0$ to define $t^*$.

3. $\Delta G(t/t_w)$ for three different values of the stress-field $F_n$, measured on the same sample ($R\Box=57M\Omega$). Each master-function is labeled by its $F_n$ (in units of V/cm). At least three different $t_w$ were used in any such plot with $t_w$ ranging between 10 to 1620 seconds. The inset shows $t^*$ as a function of the stress-field for this sample (circles ) and for two other samples ($R\Box=11M\Omega$-squares, $R\Box=40M\Omega$-triangles).

4. $\Delta G(t/t_w)$ for the same sample as in figure 3 ($R\Box=57M\Omega$) while using a sufficiently high stress-field such that simple aging is no longer obeyed.

5. Field effect $\Delta G(V_g)$ traces measured for the same sample as in figures 3 ($R\Box=57M\Omega$) illustrating the ‘loss of memory’ due to various stress fields. See text for the experi-
mental procedure. The trace taken with $10^{-1}\text{V/cm}$ is the “baseline-memory” for the series. Note that appreciable reduction in the anomalous cusp (dip around $V_g=0$, c.f., reference 7) for $F_n \geq 400\text{V/cm}$ that coincides with the demise of simple aging in this sample (c.f., figures 3 and 4).
Figure 1
Figure 2
$F_n = 400 \text{ V/cm}$

$\Delta G/G (\%)$

$t/t_W$

$t_w (\text{sec.})$

$10^1$

$10^2$

$10^3$

Figure 4
Figure 5