Slow Conductance Relaxation in Insulating Granular Al: Evidence for Screening Effects

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(Received 9 July 2010; published 6 May 2011)

It is shown that the conductance relaxations observed in electrical field effect measurements on granular Al films are the sum of two contributions. One is sensitive to gate voltage changes and gives the already reported anomalous electrical field effect. The other one is independent of the gate voltage history and starts when the films are cooled down to low temperature. Their relative amplitude is strongly thickness-dependent which demonstrates the existence of a finite screening length in our insulating films and allows its quantitative estimate (about 10 nm at 4 K). This metalliclike screening should be taken into account in the electron glass models of disordered insulators.

DOI: 10.1103/PhysRevLett.106.186602 PACS numbers: 72.80.Ng, 61.20.Lc, 73.23.Hk

During the past 20 years, slow and glassy conductance relaxations were found in several disordered insulating systems [1–3]. These relaxations have been revealed and often studied by means of field effect measurements. In such experiments, disordered insulating films are used as (weakly) conducting channels of metal-oxide-semiconductor field-effect transistor (MOSFET) devices which allow us to measure their conductance response to gate voltage (V_g) changes. After a quench at, e.g., 4 K, a slow and endless decrease of the conductance is found as long as V_g is kept constant, and any V_g change triggers a new conductance relaxation. Moreover, the system keeps some memory of its V_g history: Any stay under a fixed V_g value remains printed for some time in $G(V_g)$ sweeps as a conductance dip centered on this value. We have also shown recently in granular Al thin films that these V_g-induced relaxations display aging; i.e., the dynamics of the system depends on the time spent at low temperature, a characteristic property of glassy systems [4].

Several experimental findings suggest that these slow conductance relaxations could reflect the properties of the ‘‘electron glass’’ [5,6], a glassy state theoretically predicted in the 1980s for disordered insulators [7,8]. According to theoretical and numerical studies, a system of electrons with ill-screened interactions and disorder will need an infinite time to reach its equilibrium state at low temperature, the relaxation towards equilibrium being characterized by a conductance decrease of the system [9]. Up to now, the electron glass problem has received considerable theoretical developments but only few experimental illustrations, which explain the interest in the electrical field effect results. Moreover, most of the experiments have focused on the electrical conductance and its V_g-induced relaxations, but less is known concerning the dielectric properties and their possible time evolution at low T.

Since even after a long stay under a fixed V_g the conductance decrease shows no sign of saturation, it is not possible to define the conductance relaxation relative to the equilibrium value, which is unknown. Instead, short excursions to V_g values never explored before are often used to define a V_g history-free reference conductance G_{ref} [2–5]. This history-free conductance was sometimes called the ‘‘off-equilibrium’’ conductance since the system has never been allowed to equilibrate at these V_g’s. In the present Letter, we show that G_{ref} is not constant in time after a cooldown to 4.2 K in granular Al films thicker than 10 nm. We also show that this feature demonstrates the existence of a screening length in our insulating granular films and allows its quantitative estimate.

Our granular Al films were prepared by e-gun evaporation of Al under a partial pressure of O_2, as described elsewhere [3]. By changing the O_2 pressure, we can tune the resistance of the films from metallic to insulating. For insulating samples, x rays and TEM studies have revealed an assembly of crystalline Al grains with a typical size of a few nanometers. They are believed to be separated by thin insulating Al_2O_{3-x} layers. MOSFET devices are made by deposition of granular Al films on top of heavily doped Si wafers (the gate) covered by a 100 nm thick thermally grown SiO_2 layer (the gate insulator).

The film conductance G was measured by using a two terminal ac technique, employing a FEMTO current amplifier DLPCA 200 and a lock-in amplifier SR 7265. Source-drain voltage was such that G stays in the Ohmic regime. All the electronic equipments sensitive to room temperature drifts were placed in a thermalized chamber with a T stability better than 0.1 K. This last point was crucial in order to follow G variations with a precision of $10^{-3}\%$ during weeks of measurements.

For 4.2 K measurements, the MOSFET devices are mounted in a box filled with He exchange gas and plunged into a 100 l liquid He dewar. The sample temperature follows the mK variations of the liquid He bath around 4.2 K, and a carbon glass thermometer close by was used to correct these temperature variations. The time required to cool down the sample from room temperature to 4.2 K is about 10 min.

For Fig. 1(a), a MOSFET device with a 20 nm thick granular Al channel was cooled down to 4.2 K and maintained at this temperature under $V_{geq} = 0$ V. Fast V_g sweeps from -15 to $+15$ V (250 s long) were taken every 6000 s after the cooldown. All the $G(V_g)$ curves display a conductance dip centered on V_{geq} which reflects the memory of the relaxation associated with the stay under V_{geq}. Far enough from V_{geq} (here for $|V_g| > 5$ V), G is roughly constant, and this baseline reflects the off-equilibrium conductance G_{ref} previously mentioned. Looking at the time evolution of $G(V_g)$ curves, two features are salient. First, the amplitude of the dip increases as a function of time. This result is well known from previous studies [2–5]: The longer the stay under V_{geq}, the more pronounced the dip is. Second, and this is the new feature we aim to discuss here, the baseline conductance also decreases as a function of time. As highlighted in Fig. 1(b), the G decrease is well described at any V_g's by a ln$t$ dependence, more pronounced in the dip region because of the superposed baseline relaxation and dip growth.

To be more quantitative, we can define a parameter whose physical meaning will become clear later, the slope ratio (SR) of the G relaxation slope in Fig. 1(b) at V_{geq} to the G_{ref} relaxation slope (at -10 V, for example). For the sample of Fig. 1, SR $\simeq 2.9$. We have measured 4 different 20 nm thick films with $R_\square$ values from 10 MΩ to 10 GΩ. A baseline relaxation was always observed, with SR at 4 K between 2 and 3. For one film, the gate and the gate insulator were, respectively, Al and alumina, and consistent results were obtained which excludes any role of a specific gate insulator material. Detailed investigation of the vertical electrical homogeneity of the films has also shown that the baseline relaxation is not related to a specific layer close to the film-substrate interface [10].

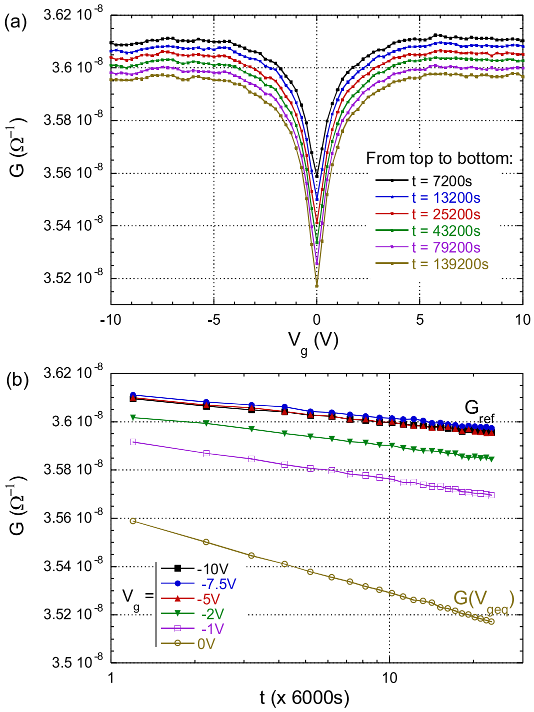

FIG. 1 (color online). (a) $G(V_g)$ curves measured at different times t after a cooldown to 4.2 K. $V_g = V_{geq} = 0$ V between V_g sweeps. (b) Corresponding $G(t)$ curves for negative V_g's in and out of the conductance dip region. The sample was 20 nm thick with $R_\square = 550$ MΩ at 4.2 K. See the text for the details.

We checked that this baseline relaxation was independent of the V_g history by using the "two-dip" protocol [5]. After some time under V_{geq1}, V_{geq} was changed to V_{geq2} for the rest of the experiment. We observed the formation of a new dip at V_{geq2} and the erasure of the old one at V_{geq1} (as already known [3,5]), but the baseline relaxation in any V_g range far enough from V_{geq1} and V_{geq2} continues as if no V_{geq} change was imposed.

One may think that the baseline relaxation is induced by the V_g sweeps themselves. Indeed, when measuring $G(V_g)$ curves, a small dip starts to form at each measured point. The baseline thus reflects a short time relaxed G value (10 s being typical for our sweep parameters). We know from the two-dip protocol results that a dip formed during 10 s will be erased roughly in about 100 s [2,3]. In our case, we wait for 6000 s between two V_g sweeps; thus, no memory of the previous sweep is expected out of the dip region. In one experiment, we checked that the baseline relaxation was unaffected by the suspension of V_g sweeps during 30 h.

An experimental artifact that may explain the baseline relaxation is the existence of a T drift after the cooldown. Because of the rapid divergence of the film resistance at low T, the observed baseline variations correspond typically to drifts of a few mK ($\simeq 2.5$ mK for the whole relaxation observed at 4.2 K in Fig. 1). We have performed different tests (thermometer stability and comparison with and without He exchange gas) which indicate that the T drifts are much smaller than the baseline relaxation amplitude. The effect of the film thickness discussed below will definitely ruled out any interpretation in terms of a T drift.

If the baseline relaxation is a property of the granular film itself, how can we explain it? A simple hypothesis is to state that the granular Al film, although electrically insulating, has a metalliclike screening length L_{sc} that is smaller than its thickness T_h ($T_h = 20$ nm in Fig. 1). Then, only the layer of the film located at a distance smaller than L_{sc} from the gate insulator is sensitive to V_g changes. The conductance dip ΔG_{dip} reflects the relaxation of this layer, whereas the conductance baseline G_{ref} reflects the relaxation of the rest of the film. The conductance relaxation measured at V_{geq} is then the sum of the two contributions: $\Delta G(V_{geq}, t) = \Delta G_{\mathrm{dip}}(t) + \Delta G_{\mathrm{ref}}(t)$. Since the relaxations at any V_g are well described by a ln$t$ dependence, we can write $\Delta G_{\mathrm{ref}}(t) = -A_{\mathrm{ref}} \ln t$ and $\Delta G_{\mathrm{dip}}(t) = -A_{\mathrm{dip}} \ln t$. The

SR is by definition equal to $(A_{\rm dip} + A_{\rm ref})/A_{\rm ref}$. If the film is homogeneous, it is natural to suggest that $A_{\rm dip}$ and $A_{\rm ref}$ are, respectively, proportional to L_{sc} (the V_g sensitive layer thickness) and $T_h - L_{sc}$ (the V_g insensitive layer thickness). Then, SR $= T_h/(T_h - L_{sc})$ as long as $T_h > L_{sc}$. The results for the 20 nm films (SR between 2 and 3) give estimates for the screening length L_{sc} between 10 and 13 nm.

If our simple model is correct, then the baseline relaxation relative to that measured at V_{geq} must change with the thickness of the films. This is illustrated in Fig. 2, where the experiment of Fig. 1 was reproduced for two granular Al films 10 and 100 nm thick. A clear thickness dependence is visible. Qualitatively, the baseline relaxation is almost absent for the 10 nm thick film [Fig. 1(a), SR $= 20$], while it is almost equal to that measured at V_{geq} for the 100 nm thick film [Fig. 1(b), SR $= 1.1$]. The agreement with the above simple picture is also quantitative. These SR values give L_{sc} estimates of about 10 nm, close to the results obtained from 20 nm thick films (10–13 nm). We stress that the SR variations observed for different thicknesses are much more important than those observed for a given thickness and different $R_\square$'s.

Our simple model also predicts a thickness dependence for the amplitude of the conductance dip $\Delta G/G = [G_{\rm ref} - G(V_{geq})]/G_{\rm ref}$. $\Delta G/G$ data measured for 10, 20, and 100 nm thick films are shown in Fig. 3 as a function of their percolation critical resistance R_C [11]. In 2D films (T_h less than the percolation radius L_0), $R_C \simeq R_\square$, but for thicker ones, $R_C \simeq (T_h/L_0)R_\square$. Taking $L_0 \simeq 20$ nm [12], we estimate R_C as $R_\square$ for 10 and 20 nm films and $5R_\square$ for 100 nm films. All the samples were measured after being kept under $V_{eq} = 0$ V for $t = 20$ h after a cooldown at 4.2 K. As can be seen and was already reported [1,3], $\Delta G/G$ increases significantly with R_C. Besides, a clear reduction of $\Delta G/G$ is visible for a given R_C as T_h increases, in qualitative agreement with the fact that the conductance dip originates from a layer of a fixed thickness. The agreement is also quantitative. As long as $T_h < L_{sc}$, $\Delta G/G(t)$ does not depend on T_h. Let us note its value $C(t)$ [of course, $C(t)$ depends on R_C]. When $T_h > L_{sc}$, one has $\Delta G/G(t) = A_{\rm dip}\ln t/(G_0 - A_{\rm ref}\ln t) \simeq A_{\rm dip}\ln t/G_0 = C(t)L_{sc}/T_h$ $[\Delta G/G(t) \ll 1]$. Then, plotting $(T_h/L_{sc}) \times (\Delta G/G)$ for all the samples should give one single curve $C(t, R_C)$. This is confirmed in the inset in Fig. 3 for which we have assumed a screening length of 10 nm for all the films. An increase of the $\Delta G/G$ data scatter with the thickness is also observed in Fig. 3. It is related to some departure from a perfect vertical homogeneity of the films, as discussed in Ref. [10].

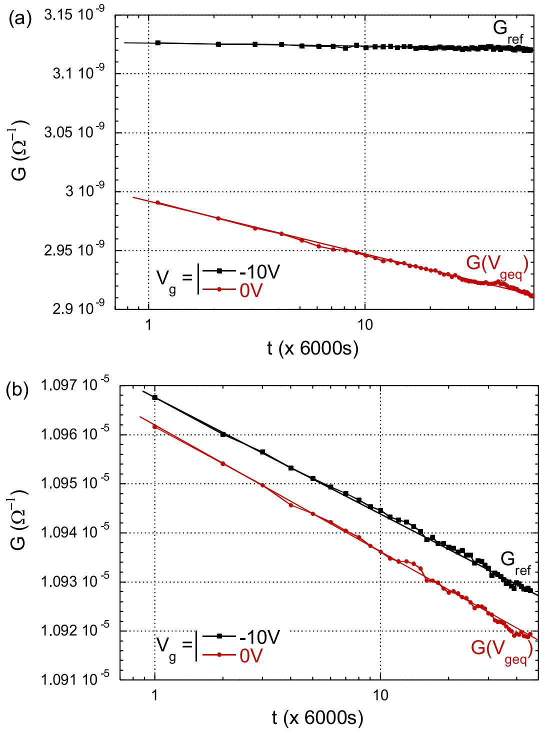

FIG. 2 (color online). $G(t)$ measurements after a cooldown at 4.2 K for (a) a 10 nm thick film, $R_\square = 6$ GΩ, and (b) a 100 nm thick film, $R_\square = 2$ MΩ. The $G(t)$ relaxations are plotted at 0 V (V_{geq}) and at -10 V (baseline relaxation $G_{\rm ref}$).

We now come to a discussion of our results. First, note that the thickness dependence of SR clearly demonstrates that the conductance relaxation exists in the "bulk" of the film and is a property of the granular Al itself (and not of its interface with the gate or the surface oxide layer). The bulk relaxation after a cooldown at 4.2 K is well described by a $\ln t$ dependence without any sign of saturation over weeks of measurement. Such a $\ln t$ dependence was also observed for the time increase of the conductance dip amplitude after a cooldown [2,3] (more complex laws are observed when V_{geq} is fixed a time t_e after the cooldown; see [4]). It

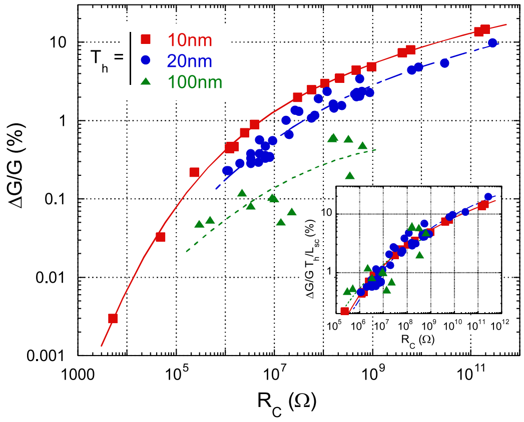

FIG. 3 (color online). $\Delta G/G$ values measured 20 h after a cooldown at 4.2 K as a function of R_C (see the text for the details). The films are 10 (squares), 20 (circles), and 100 nm (triangles) thick. The lines are guides for the eye. In the inset, $(\Delta G/G) \times (T_h/L_{sc})$ versus R_C. Typical error bars on each point are about 10%.

is a natural relaxation law for a system having a $1/\tau$ distribution of relaxation times, as expected for an electron glass [13].

In our granular Al thin films, the electronic transport at finite T is believed to result from electron tunneling between metallic Al grains. A lower bound for L_{sc} is then the typical grain size, which was found in recent TEM measurements to be 2–4 nm. A second important microscopic length scale is the percolation radius L_0 of the critical resistance network. Since in our films $R_C \lesssim 100\ \text{G}\Omega$, the diffusion of charge carriers through the critical resistance network is expected to be faster than the time scale of the measurements (the intergrain capacitance and the capacitance of the granular Al channel to the gate are, respectively, $\simeq 10^{-19}$ and $\simeq 10^{-11}$ F). Thus, L_0 should be an upper bound for L_{sc} as indeed observed: L_0 estimates from conductance fluctuations measurements are between 20 and 40 nm [12], a few times our L_{sc} value.

We note that no baseline relaxation was reported on 20 nm thick indium oxide films which may indicate that $L_{sc}(4\ \text{K}) > 20$ nm in this system. Interestingly enough, L_0 was found to be $\simeq 300$ nm in "crystalline" films [14], i.e., $\simeq 10$ times our estimate for granular Al films, reflecting a less dense percolating network in indium oxide consistent with a larger screening length.

Our observations address the important question of screening in a disordered insulator, a subject which has been little explored both theoretically and experimentally. Electron glass models are generally developed in the limit of strongly localized electrons [7–9] (Refs. [15,16] are exceptions). But in real systems, there is a mobility of the charge carriers at finite T which will give rise to a metalliclike screening. Numerical studies on disordered insulators have indeed found a transition between a metallic screening at high T, where most of the electrons are diffusive, and a slow dielectric response at low T, where most of the electrons remain located in finite size clusters [17–19]. Such a transition was observed around 1 K in capacitive measurements on a doped semiconductor [20]. According to the usual formula, $L_{sc}^2 = \epsilon\epsilon_0/e^2 dN/d\mu(E_F)$, where $dN/d\mu$ is the thermodynamic density of states. In a glassy phase and for a finite time t, the system cannot explore all the configurations, and the thermodynamic density of states has to be replaced by a pseudoequilibrium density of states $dN/d\mu(t)$ for which only relaxations faster than t are allowed. The first theoretical attempts to do so [7,21] have found that the short time ($\simeq$ Maxwell time) screening length diverges as T goes to 0 and decreases as a function of time [21].

In summary, we have described in insulating granular Al films the existence of a V_g insensitive conductance relaxation. Its thickness dependence demonstrates the existence of a metallic screening length of about 10 nm at 4 K. Our results provide a new way to study the screening length and its relaxation in disordered insulators where an anomalous electrical field effect has been found. They also point to the need for more theoretical studies: How are the classical electron glass models predictions affected by the existence of a metallic screening length ?

This research has been partly supported by the French-National Research Agency ANR (Contract No. 05-JC05-44044).

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