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# Microscope of glassy relaxation in femtogram samples: Charge offset drift in the single electron transistor

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By measuring the long-term charge offset drift in single electron tunneling transistors, we have observed a transient relaxation after fabrication which is correlated with the presence of amorphous insulator. The temperature and time dependence of the transient relaxation are both in agreement with an extension of the standard model for two-level systems in glasses. This technique, which is sensitive to atomic scale motion in femtogram-sized samples, offers the possibility of a technique for investigation of glassy relaxation.

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The study of glassy relaxation in amorphous materials at low temperature is both venerable and yet still active at present.[1] In general, most of the experiments performed to date have been on macroscopic samples. In particular, thermal conductivity, specific heat, and nonequilibrium heat release measurements have all been performed primarily on bulk samples; recently, such measurements have been extended into the thin-film regime involving samples with microgram or nanogram masses.[2,3] These studies have shown the need for, and the interesting physics that can result from, measurements in very small samples. For example, in a recent measurement using these techniques,[4] the specific heat of a thin-film amorphous Si sample showed a lack of tunneling states in an amorphous material.

In this paper, we show that it is possible to go beyond this regime and look at even smaller sample sizes using electrical measurements. For example, the gap between calorimetric measurements (macroscopic) and molecular dynamics simulations[5,6] (microscopic, typically about 1 million atoms at most) precludes direct comparison; by measuring glassy relaxation corresponding to just a few mobile clusters, it may be possible to bridge that gap. Molecular dynamics simulations for amorphous alumina[7] reveal that the structure is $SiO_4$ tetrahedra and (predominantly) $SiO_6$ octahedra, with a broad range of O–Al–O bond angles centered at 93°; low-temperature glassy relaxation occurs via orientational (bond angle) changes in the rigid tetrahedra and octahedra, with a length scale set by the fourfold and fivefold rings. In addition, as mentioned above, thin-film samples may show behavior different from bulk,[4] and the ability to probe the intermediate size regime may uncover other such differences.

Recently, studies on nanoelectronic devices, in general, have shown electrical characteristics which can depend on atomic positions. These characteristics are generally considered to be "noise" which arise from "defects" through either the defect position (atomic motion) or charge state (charge traps).[8–10] In addition to being unwanted noise, such electrical measurements can also provide information about exquisitely small ensembles of atomic motion, including the well-known case of "two-level fluctuators"[8,11] (TLFs) which allow real-time measurements of the state of a single microscopic defect. However, results in the intermediate size regime, where there is collective motion of a number of such defects, has not been shown in these electrical measurements.

Single electron tunneling (SET) transistors have excellent sensitivity when used as charge electrometers, with the ability to measure capacitive charge changes below $10^{-4}e$.[12] If there is disordered material in the insulating regions surrounding the thin-film SET transistor, any structural relaxation—if it is accompanied by a change in the charge distribution of the defect—can give rise to a change in the capacitively coupled "charge offset" $Q_0$, which represents a (possibly time-dependent) phase shift in the transistor control curve. In general, the resulting fluctuations in transistor current fall into two regimes: standard $1/f$ noise[13] (typical charge fluctuation about $0.001e$) at short times, and large drift ($1e$ or greater) at times of hours or longer.[14] In particular, we have recently shown that in SET transistors based on the $Al/AlO_x/Al$ tunnel barrier, there can be a nonequilibrium relaxation of the time dependence of the charge offset drift in freshly fabricated devices.

In this paper, we investigate in detail this "transient relaxation" (TR) in $Q_0(t)$. We show that the TR in $Q_0(t)$ shows the same long-time tail as the heat release in macroscopic silica samples when quenched to temperatures below 1 K,[15] although with a sensitivity to femtogram samples rather than milligram-size samples. We show the transient has a time dependence which can be predicted from the standard two-level system (TLS) model,[16] which also predicts the heat release in macroscopic samples. As an example of the interest in measuring the same glassy relaxation in smaller samples, we discuss one difference between the TR and the heat release: the conditions under which the relaxation recurs.

We fabricated standard three-terminal SET transistors with $Al/AlO_x/Al$ tunnel junctions (Fig. 1 insets).[17] On some of these devices, we put an extra layer about 80 nm thick of amorphous $AlO_x$ [Fig. 1(a) inset] deposited by electron-gun evaporation of $Al_2O_3$; typical base pressure was $1\times10^{-5}$ Pa, during deposition 3 to $10\times10^{-5}$ Pa. For the devices represented here, the devices were cooled below 4 K within 1 1/2 days after deposition.

Figure 1 shows the charge offset drift as a function of time $Q_0(t)$ in SET transistors, for devices with (1a) and without (1b) extra amorphous insulator deposited onto the transistor island. One can clearly see in Fig. 1(a) that the charge offset is changing in a random way much more rapidly at early times than at late times; initially, the magnitude of the

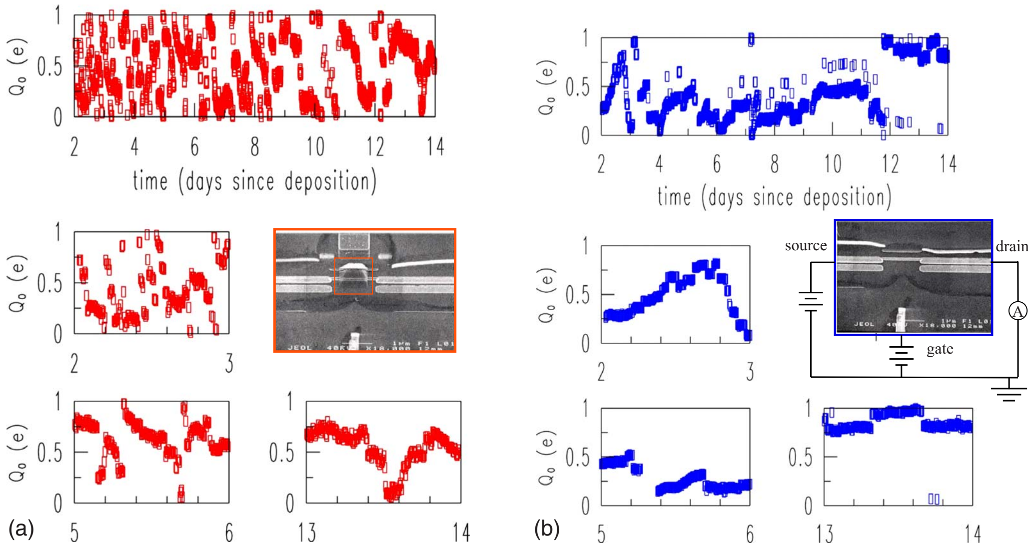

FIG. 1. (Color online) Charge offset $Q_0(t)$ as a function of running time since device fabrication ($t=0$). Upper panel: full time dependence; Lower panels: magnification of selected slices of time, showing systematic dependence on time since deposition. $I(Q_0)$ is a periodic function with period $1e$, and thus $Q_0(t)$ is only defined modulo $1e$; as a result, we plot $Q_0(t)$ in a reduced zone scheme by suppressing the possible integer offset. The inset shows a micrograph of the SET transistor [Al lines (bright) on an oxidized Si substrate (dark)]. (a) Red data from device with extra $AlO_x$ deposited (shown in inner red square in micrograph) shows transient relaxation in time [rate of random change in $Q_0(t)$ decreases with running time]; (b) blue data from device without extra $AlO_x$ shows no transient relaxation in time. Inset shows schematic of measurement circuit: (voltage bias on source, current flow measured at drain, capacitively coupled gate electrode). Uncertainty in $Q_0(t)$ is about $\pm 0.01e$; for more information on the measurement technique, please consult (Ref. 14). $H=1$ T. The temperature started at 25 mK, and was raised above 1 K at $t=9$ days; note that $Q_0(t)$ is temperature independent.

time rate of change in the charge offset [i.e., $|dQ_0(t)/dt|$] is so large that subsequent measurements of $Q_0$ (spaced five minutes apart) appear random. In contrast, such a TR may also exist in the device without the extra insulator [Fig. 1(b)], but to a much smaller extent. We have generally observed:[14] (i) TR in devices when measured immediately after fabrication; devices measured later will show an equilibrium (stationary) level of $Q_0(t)$ but no TR; (ii) TR often in devices with extra insulator, and rarely in devices with just the native and tunnel junction insulators. For the devices discussed in this paper, full details of fabrication and measurement parameters can be found in Ref. 14.

We can quantify this observation of TR by measuring the time rate of change in the charge offset $Q_0(t)$ as measured in different bins of time, as follows: starting with the data in Figs. 1(a) and 1(b), we obtain "duration times," i.e., the successive lengths of time before $Q_0(t)$ changes by more than a certain amount ($0.1e$). We then form Markov plots (semi log plots of number of duration times versus duration time); the values of $\gamma$ come from the slopes of Markov plots; error bars come from statistical uncertainty in the Markov plots.

The results of this analysis are shown in Fig. 2. We can clearly see that in two different devices with extra insulator sitting on top of the island (black and red data), there is a strong decrease in the time rate of change $\gamma$ as a function of running time since deposition of the Al film; in contrast, for a device without such extra insulator, there is no such systematic decrease apparent. As is evident from the line, the TR has a general time dependence of $\gamma \propto 1/\text{time}$, identical to the behavior of the nonequilibrium heat evolution in glasses.[15] This analysis of relaxation rate versus running time, and the resulting qualitative dependence $\gamma \propto 1/t$, are the central experimental results of this paper.

Having demonstrated the basic behavior of the long-term (time scale of hours or longer) drift, we can also examine the

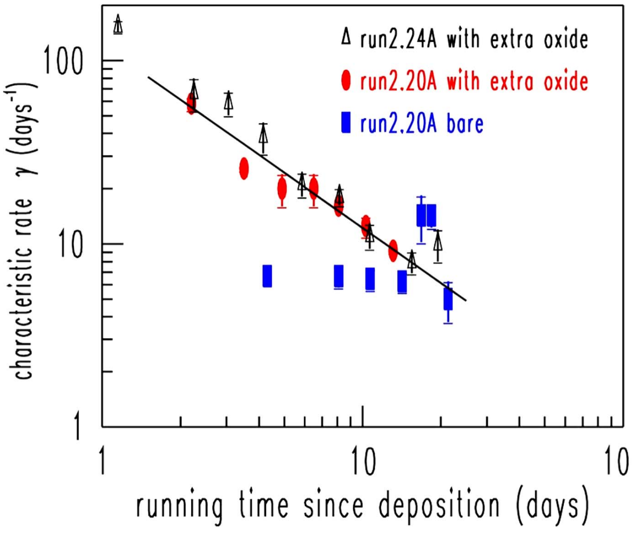

FIG. 2. (Color online) Log-log plot of rate of change in $Q_0(t)$ with running time for three different devices. The red and blue sets of data correspond to the data in Fig. 1; the black data (2.24A) is an additional set from a different device which also had extra $AlO_x$ (320 nm) deposited on top. Two of the three sets of data clearly show the transient relaxation, where the rate of change in $Q_0(t)$ decreases systematically with running time over about a decade; the line is a guide to the eyes with a dependence of $\gamma \propto 1/\text{time}$.

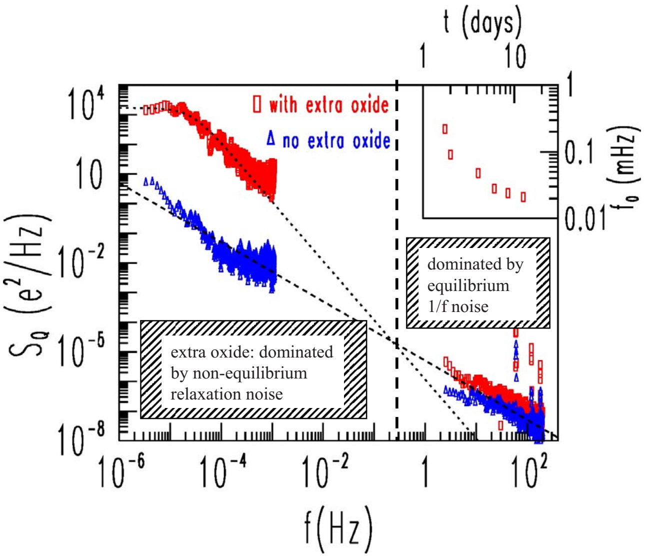

FIG. 3. (Color online) Main: power spectral density of charge fluctuations versus frequency; (i) with extra $AlO_x$, showing the Lorentzian shape of the long-term drift (below 1 mHz, measured at $t=7$ days); (ii) without extra $AlO_x$ (vertically shifted for clarity), showing the lack of a knee at low frequencies. Also shown is the $1/f$ spectrum of the short-term noise (above 1 Hz) for both devices. Inset: knee frequency versus running time.

short-term $1/f$ noise (time scale of seconds or shorter). We can explicitly show the difference between short-term and long-term by examining the power spectral density $S_Q(f)$ of both types of fluctuations, as shown[18] in Fig. 3. The short-term noise at higher frequencies has the typical $1/f$ spectrum. It is clear that, in order for the long-term drift to generate the $1e$ changes evident, its power spectral density must lie significantly higher than the $1/f$ extrapolation from the short-term noise;[19] this is apparent from the figure which shows that the long-term drift corresponds to a Lorentzian spectrum $S_Q(f) \propto \frac{1}{1+(f/f_0)^2}$, with a characteristic knee frequency $f_0$. Consistent with the TR of the long-term drift, both the knee frequency and the amplitude of the Lorentzian change with time, with the knee frequency moving to lower frequencies (as shown in the inset) as the running time increases. In contrast, the devices without extra insulator [e.g., Fig. 1(b)] have the featureless $1/f$ spectrum without a knee frequency (the small upturn noticeable at the low-frequency end is typical of "rolling" $1/f$ noise[13]).

We can note two additional distinctions between these two types of fluctuations: first, in contrast to the long-term drift $Q_0(t)$, the amplitude $S_Q(f)$ of the short-term $1/f$ noise ($f>1$ Hz) does not decrease over the same time period (not shown). Second, the temperature dependence of these two fluctuations also exhibit qualitatively different behavior: $Q_0(t)$ is independent of temperature, as exemplified in Fig. 1 which ranges from 0.03 K to 1.2 K. In contrast, $S_Q(f)$ at 10 Hz increases by about a factor of three, similar to other reports.[20]

To summarize the experimental results: (i) the TR is correlated with the presence of amorphous insulator;[14] (ii) the rate of change in $Q_0(t)$ is proportional to 1/time; (iii) the presence of the TR is correlated with the presence of a knee frequency in the power spectral density; and (iv) the TR is time dependent but not temperature dependent; the short-term $1/f$ noise is not time dependent, but is temperature dependent.

We can compare the experimental results to a model we previously developed.[14] This model is driven by the first two empirical observations listed above, which are very reminiscent of the nonequilibrium heat evolution in amorphous materials. This heat evolution can be explained naturally in the TLS model[16] as due to TLS which are frozen into nonequilibrium energy states by the quench, and are slowly relaxing to the ground state while releasing energy. We have extended the TLS model to predict the absolute magnitude, and temperature and time dependences of $\gamma$, by calculating the rate of release,[14] which for energies $E$ between $E_{\min}$ and $E_{\max}$ ($V$ is sample volume) is

$$\dot{N} = V\bar{P}(E_{\max} - E_{\min})/(2t), \tag{1}$$

here, $\bar{P}$ is the average density of TLS. We note that this rate does not depend on temperature, and is indeed inversely proportional to time (both in agreement with the data, as in Figs. 1 and 2).

As one example of the utility of this "microscope" for the examination of glassy relaxation, we can estimate the density of TLS from the rate of relaxations. We have (i) material volume $V$ of area 0.5 $\mu m^2$ and thickness of 80 nm; (ii) $E_{\min}=0$, $E_{\max}=10$ K,[6] and we assume (iii) every relaxation results in an electrostatic atomic reconfiguration which changes $Q_0$ (and thus $\dot{N}=\gamma$). From Fig. 2, $\dot{N}=\gamma \approx 5/\text{day}$, after $t$=three weeks. We obtain $\bar{P} \approx 1.5 \times 10^{38}$ $J^{-1}$ $cm^{-3}$, which is only about a factor of five higher than that reported in a wide range[1] of amorphous materials as deduced from heat-evolution measurements.[15] We believe that this similarity in quantitative values strongly suggests that this technique is probing the same glassy relaxation measured by the calorimetry techniques. We must note that this estimate of the density of TLS depends on several assumptions as defined above, and thus can be considered only semiquantitative; we believe that further studies may allow tests of these assumptions.

As a second example of the utility, we consider the implications for TLS dynamics implicit in the noise data. Consider, for instance, the data in the lower left panel of Fig. 1(a), showing data between days five and six. In some cases, the charge offset $Q_0(t)$ shows abrupt changes which might be due to a single TLS relaxing to a lower energy state, if the capacitive coupling between the TLS and the SET transistor is fairly large. In other cases, such as the data between 5.3 and 5.6, $Q_0(t)$ show a smooth gradual change in its value, while at other times (between 5.0 and 5.2), $Q_0(t)$ appear to be basically constant. These latter observations strongly suggest that oftentimes the TLS are interacting, so that the smooth gradual evolution of $Q_0(t)$ can naturally be explained by subsequent relaxation of TLS which are made energetically favorable by the previous relaxation of other TLS.

In addition, we can consider the effect of such interactions on the power spectral density data, and its implications for spin glasses: Fig. 3 shows a Lorentzian with regions that are flat and proportional to $1/f^2$, but with no intermediate $1/f$ region. In contrast, the standard model for $1/f$ noise,[13] coming from independent TLS with a distribution of energies, predicts a wide $1/f$ region. The lack of this region in our data (as indicated both by the spectral shape and by the shift in knee frequency with time) suggests that the TLS have strong interactions driving their dynamics, consistent with the transient relaxation observed in Fig. 1(a). This result also has implications for, for instance, the long-standing issue of whether or not spin glasses demonstrate $1/f$ noise versus a steeper frequency dependence;[21] we suggest that the resolution of this issue is in whether measurements are made on fresh versus fully relaxed samples.

To summarize, we have performed measurements of the long-term drift of the charge offset in SET transistors. We have observed that there is a TR in the drift after fabrication. The qualitative temperature and time dependences are different from those of the short-term ($1/f$-like) noise. We have demonstrated that an extension of the TLS model for glasses and amorphous materials can explain the quantitative and qualitative behaviors of the drift as driven by nonequilibrium relaxation of the disordered atoms.

In conclusion, we believe that we have developed a technique for examining glassy relaxation in very small (femtogram) samples. We can note among other intriguing possibilities.

(1) The time dependence of the glassy relaxation on the microscale can be examined in the correlation times indicated by the knee frequency of the Lorentzian spectrum.

(2) The detailed time dependence can also be elucidated by examination of data such as that shown in Fig. 1(a) (days 5–6), which shows both abrupt changes, but also slow evolutions over courses of minutes or hours. The slow evolutions suggest that sometimes microscopic atomic motions may occur on a *very* slow time scale. Such time dependence on the intermediate scale, where a few clusters (two-level systems) are being observed cannot be examined in the averaged measurements of macroscopic samples, nor in TLF measurements of individual defects. Such time dependence on this intermediate scale might be correlated with molecular-dynamics simulations.[5,6]

(3) The microscopic nature of the defects, and thus the glassy relaxation, may be different for thin-film samples.[4] For instance, one difference[14] between our observations of transient relaxation and the nonequilibrium heat evolution in macroscopic glasses: the heat evolution recurs upon thermal cycling of the sample, whereas the transient relaxation in $Q_0(t)$ is apparently frozen in at device fabrication. This possibility may bear on the "universality" question[1] in amorphous materials. In this vein, it may be interesting to apply our technique to amorphous Si, which has recently been reported to have a lack of TLS.[4]

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