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Giant discrete resistance fluctuations observed in normal-metal tunnel junctions

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Giant discrete resistance fluctuations are reported in large (50×50 μm^2) tunnel junctions of Au-Al_2O_3-Au over the temperature range 4–140 K, with applied dc bias voltages of 0–2 V. The fluctuations are believed to result from single electrons trapped on defect sites within the insulating barrier of the tunnel junction. The temperature and bias-voltage dependence of the characteristic switching lifetimes of these fluctuations have been measured, and departures from previously reported behavior are observed.

I. INTRODUCTION

In recent years it has been observed that very small normal-metal tunnel junctions[1] with insulating barriers can display discrete resistance fluctuations. These fluctuations, often referred to as random telegraph signals (RTS's), are usually a small fraction of the total junction resistance. Similar effects have been observed in superconducting tunnel junctions,[2–5] metal constrictions,[6] and in various semiconductor structures, including metal-oxide-semiconductor field-effect transistors,[7–10] and are usually attributed to the trapping of single electrons on ionic defects within the insulating barrier of the junction.

These RTS resistance fluctuations are characterized by two lifetimes, τ_H and τ_L, which designate the mean (statistically averaged) lifetimes of the high- and low-conductance states, respectively. The variation of the RTS lifetimes τ_H and τ_L with temperature and applied dc bias voltage V_b have been studied by many workers,[2–4,7–10] who generally find the following behaviors: First, the RTS's are usually seen only in small junctions, below about 1 μm^2, and disappear in larger junctions, where they become indistinguishable from ordinary $1/f$ background noise. Typically, RTS amplitudes are about 1% of the total junction resistance.

Second, the lifetimes show two distinct temperature dependencies. Above a characteristic temperature T_0, both τ_H and τ_L decrease exponentially with increasing temperature, indicative of thermally activated behavior, while below T_0 both lifetimes become nearly independent of temperature.[2,4] This latter behavior is taken as the signature of quantum tunneling.

Finally, both τ_H and τ_L are strongly dependent on the applied bias voltage, becoming smaller (usually with exponential dependence) as V_b is increased. The explanation for this voltage dependence is still unclear, although various models have been proposed,[2,7,11] including a two-well model[2,8] in which defects within the barrier are presumed to undergo hopping transitions between adjacent sites.

In this paper we report the observation of discrete (RTS) resistance fluctuations in a novel *normal-metal–insulator–normal-metal* tunnel junction. Our junctions are extremely large, typically 10^3 μm^2, and yet display giant RTS's, with resistance changes between high- and low-conductance states of a factor of 10 or more occasionally observed. (This surprising result, we believe, is because the *effective* junction area that dominates the tunneling behavior is actually quite small.) In some respects, these junctions resemble switches whose on and off states are governed by the random statistics of trapped single electrons. Further, the large resistance changes have made it possible to map out the dependence of the switching lifetimes with unusual precision and suggest that the dependence of τ_H and τ_L with both temperature and bias voltage is significantly more complicated than previously realized.[2] These giant RTS's have been observed to date in more than 100 specimens.

II. EXPERIMENTAL DETAILS

As shown in Fig. 1(a), our tunnel junctions employ Au electrodes, typically 1000 Å thick, with an insulating barrier of Al_2O_3. The junction area is 50×50 μm^2 with a barrier thickness of nominally 30 Å. The room-

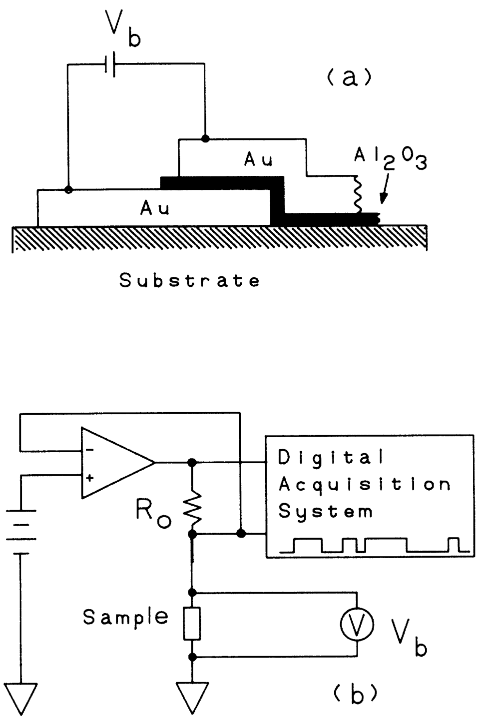

FIG. 1. (a) Schematic drawing of the 50×50 μm^2 Al_2O_3 tunnel junctions used in our experiment. The metal electrodes are Au films, about 1000 Å in thickness, and the substrate is optically polished sapphire. (b) Simplified diagram of the instrumentation used in the experiment.

temperature tunneling resistance ranges from 10 kΩ to 10 MΩ for our samples and increases by about a factor of 3 as the temperature is lowered.

Samples were fabricated using standard photolithographic techniques. First, ≈ 100 Å of Cr was evaporated onto a polished sapphire substrate to enhance the adhesion of Au electrodes to the substrate. Next, a 1000-Å-thick film of gold was evaporated onto the Cr. Then, after masking off part of the Au film, a 50×50 μm^2 area of the sample was coated with a ≈ 70 Å layer of Al, using an ion-mill sputtering system with a deposition rate of ≈ 3 Å/sec and an ambient Ar pressure of 8×10^{-5} Torr. The sample was then exposed to atmospheric (ambient) air for 2 h to oxidize the surface of the aluminum film. Finally, as shown in the figure, the junction was completed by evaporating 1000 Å of gold onto the Al_2O_3 to form the counterelectrode.

The sample resistance was measured by using a standard four-terminal scheme. The contact to the junction Au electrodes was made with an indium pad, resulting in an overall contact resistance that was 6 orders of magnitude smaller than the amplitude of resistance fluctuations.

Data were taken over a temperature range of 4–140 K, using a Janis Supertran variable-temperature cryostat and a Lake Shore model DRC-82C temperature controller. For all measurements, the junctions were biased with a dc voltage (typically 0–2 V) and the time dependence of the sample current was recorded, digitized, and analyzed by computer. A schematic diagram of the instrumentation is shown in Fig. 1(b). A low-noise instrumentation amplifier (Burr Brown INA110) was used in a feedback configuration to keep the dc bias voltage constant across the sample despite large fluctuations in sample current.

To obtain values of the lifetime in either the high- or low-conductance states, it was necessary to record more than 140 000 digitized values of the sample current, amounting to roughly 2000 resistance transitions. The lifetimes of the high- and low-conductance states were obtained by sampling and averaging the duration of the two conductance states. To minimize digitizing errors, the sampling frequency was chosen to be at least 20 times greater than the RTS switching rate, with a maximum of 100 kHz. Overall estimates of the error of lifetime measurements include contributions from sampling errors and finite counting limitations. The percentage error in a measurement of lifetime τ for N conductance transitions with a sampling frequency of f_0 is $[(1/f_0\tau)^2+1/N]^{1/2}$. Ordinarily, this error is about 3% or less.

III. EXPERIMENTAL RESULTS

All samples (about 100) displayed discrete RTS resistance fluctuations, with relative variations ranging from 5% to 2000%, with 100% being typical. Typical data are shown in Fig. 2(a), which shows the time dependence of

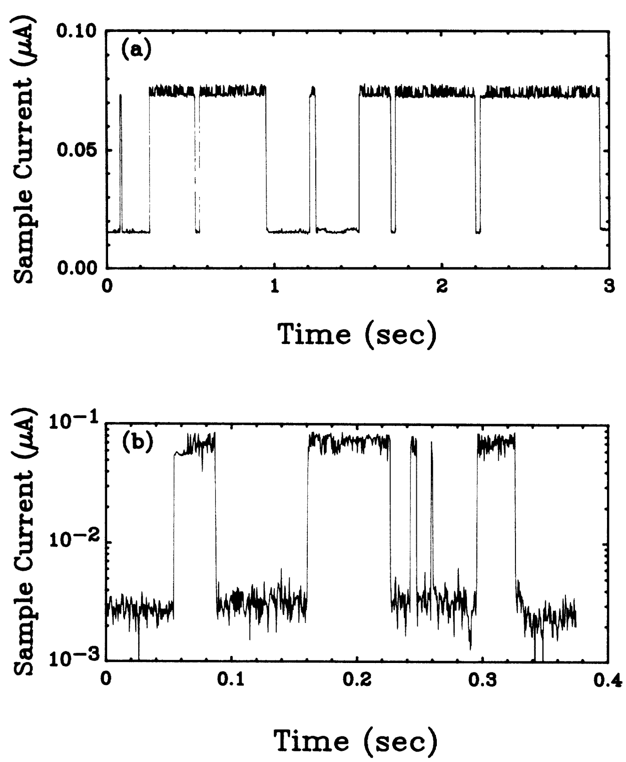

FIG. 2. (a) Time dependence of the current through a Au-Al_2O_3-Au tunnel junction illustrating the discrete conductance fluctuations. The data were recorded at $T=58$ K with a bias voltage of $V_b=233$ mV. The sample resistances in the two states were about 300 kΩ and 1.6 MΩ. (b) Data are shown for a different sample, plotted logarithmically. These data were recorded at $T=77$ K, with an applied bias voltage of 210 mV. The resistances of the high- and low-conductance states were 3.6 and 85 MΩ, respectively.

the current through a sample at fixed bias voltage of 233 mV and a temperature of 58 K. This sample actually shows three conductance states, the apparent "noise" on the high-current state reflecting rapid transitions between two closely spaced conductances. Figure 2(b) illustrates extremely large-amplitude RTS's (×24) for another sample. These data were taken at 44 K with a bias voltage of 210 mV. Note that these fluctuations are typically 10^3 larger than those previously reported.[3,5,7–9]

About half of the samples required an initial "trigger" bias voltage of 0.5–6.0 V before exhibiting large-amplitude RTS's. When this trigger voltage was applied, the sample resistance changed erratically and irreversibly (unlike RTS's, which are reversible fluctuations). However, once the trigger voltage had been removed and replaced by a smaller voltage (typically, <0.5 V), then the background resistance stabilized except for large (reversible) RTS's. The trigger-induced permanent change of sample resistance suggests that the tunnel barrier is modified by the large bias voltage, with creation of the defects that display large-amplitude RTS's. The precise mechanism for the creation of these defects (e.g., whether by dielectric breakdown or an electromigration process) is not known.

Figure 3 shows a typical histogram of the distribution of the duration of the high- and low-conductance states of a sample displaying two-state fluctuations. The mean lifetime of each state can be obtained either by a regression analysis of the slopes of these distributions,[4] or by a numerical average of the individual state durations. It is easily shown that both methods lead to the same result in the large-N limit.

Our measurements of the temperature dependence of the RTS lifetimes, illustrated in Figs. 4 and 5, are generally consistent with previous observations[2,4] in similar systems of two distinct types of temperature variation. The first, which occurs below a characteristic tempera-

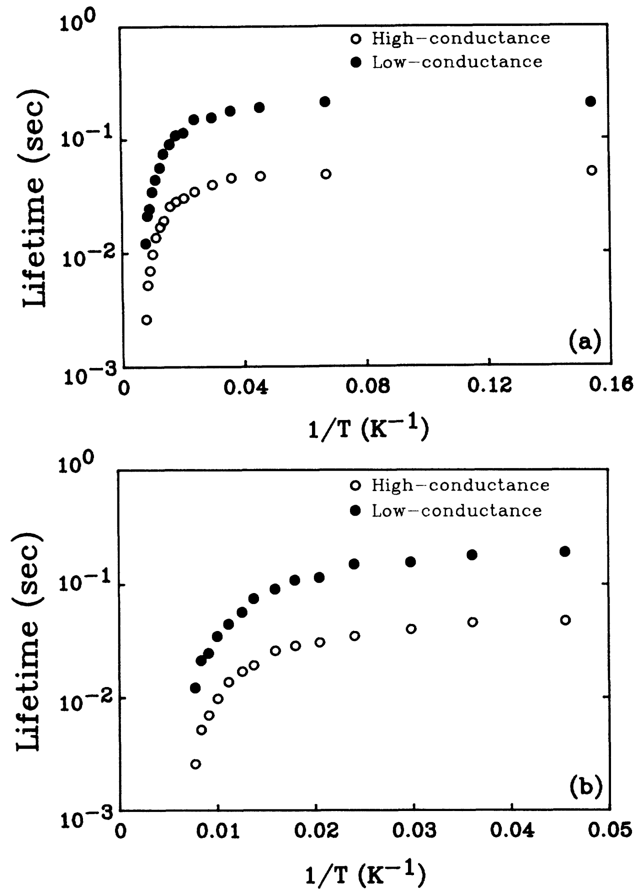

FIG. 4. High- and low-conductance lifetimes are plotted as a function of $1/T$, for a fixed bias voltage $V_b = 1.174$ V. (a) shows the full temperature range (4–130 K), while (b) shows the restricted temperature range (20–130 K). The figures demonstrate that the dependencies of the lifetimes on $1/T$ are not simple exponential, suggesting that the crossover temperature T_0 is greater than 130 K.

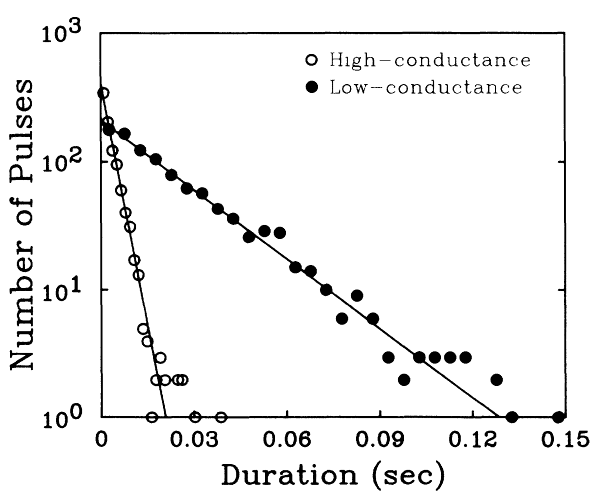

FIG. 3. Histogram showing the number of pulses as a function of pulse duration. The mean lifetimes obtained from averaging the pulse duration are $\tau_H = 24.6$ msec and $\tau_L = 3.44$ msec. Lifetimes obtained from the slope of the data points are $\tau_H = 25.5$ msec and $\tau_L = 3.46$ msec.

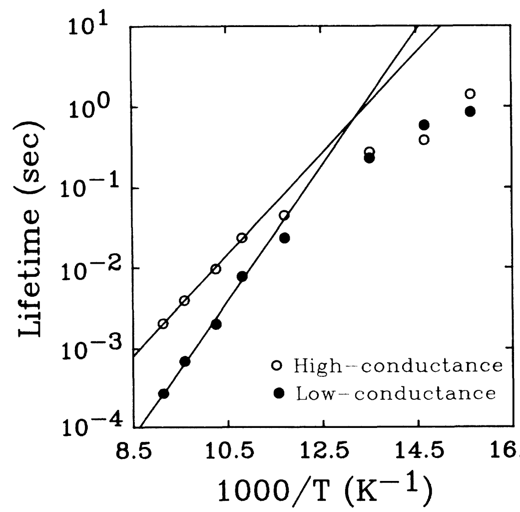

FIG. 5. High- and low-conductance lifetimes are plotted as a function of $1/T$ for a fixed bias voltage $V_b = 165$ mV. For this sample, the crossover temperature T_0 is about 90 K, above which the temperature dependence becomes exponential. The thermal activation energies of the high- and low-conductance states are $E_H = 124$ meV and $E_L = 167$ meV.

ture T_0, is a nearly-temperature-independent behavior of the switching lifetime and is often ascribed to tunneling processes. The second, which occurs above T_0, is an exponential dependence of the switching lifetime on temperature of the form $\exp(U/k_BT)$ (U is a characteristic thermal activation energy) and is interpreted as a thermally activated process. The crossover temperature that defines the boundary between these two regimes is observed to be a nonuniversal property of defects.

We have observed that the switching behavior of our samples becomes relatively unstable as the temperature and bias voltage is increased. Usually, the switching RTS associated with a given defect state will "live" about 20 h at low temperatures (10–15 K) and at bias voltages below about 500 mV. However, for $T > 70$ K or bias voltages $V_b > 1$ V, the switching RTS tends to be unstable in the sense that the switching process itself becomes short lived. Obviously, if a particular defect has $T_0 > 70$ K, then this instability makes it difficult to measure the thermally activated behavior.

For each switching RTS, we have observed that a minimum threshold bias voltage V_{th} is required to set the process in motion. (This threshold bias V_{th} should not be confused with the trigger voltage previously discussed. The trigger voltage, which was not always observed, results in irreversible damage to the dielectric that is associated with the creation of the defect state. In contrast, V_{th} is associated with a reversible process that is linked to the onset of the RTS for an already existing defect.) We stress that the RTS's stop suddenly at V_{th}; extrapolated measurements of the lifetimes to lower voltages show clearly that the RTS should be clearly measurable at voltges below V_{th} and that the threshold effect is not a simple "slowing down" of the fluctuations.

We find that there is a qualitative association between the value of V_{th} and the crossover temperature T_0, with a rough proportionality between the two quantities. Although the value of T_0 has previously been observed to be the same for all defects within the same kind of junctions,[2,12] this is not the situation for our samples. As a consequence, we have not been able to observe thermally activated behavior in those defects which have a large V_{th} (>0.5 V) because such defects invariably have $T_0 > 70$ K and are extremely erratic and difficult to measure. This point is illustrated in Fig. 4, which plots the high- and low-conductance lifetimes of the RTS associated with a single defect, with the sample biased at 1.174 V ($V_{th} \approx 1$ V). The data are shown over a temperature range of 4–130 K, and it is seen that over most of this range both lifetimes are nearly temperature independent, thus signifying tunneling behavior. The departure from T-independent behavior occurs at roughly 50 K, while T_0, which signifies the onset temperature for simple exponential behavior, was evidently above the temperature range investigated for this sample.

Figure 5 shows similar data for a defect in which $V_{th} \approx 0.1$ V. For this low value of V_{th}, we find thermally activated behavior. Below T_0, however, there is a broad crossover region which is presumably a combination of tunneling and thermally activated processes. Note that

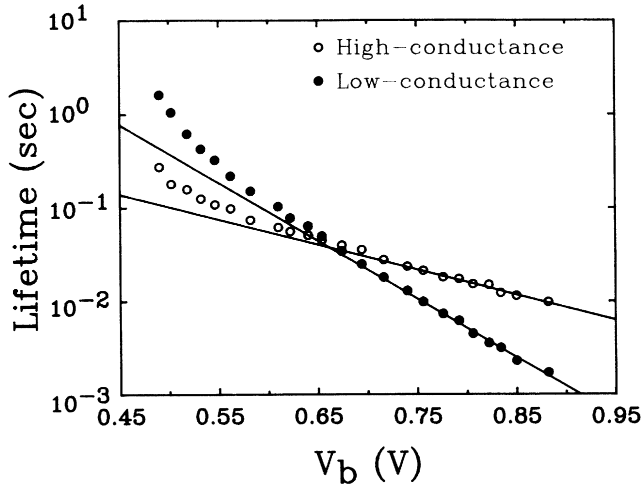

FIG. 6. Lifetimes are plotted as a function of bias voltage at $t = 4.6$ K. In this semilogarithmic plot the data clearly show nonlinear dependence. The slope $d(\log\tau)/dV_b$ of the exponential component changed about $\times 2.7$ and $\times 2.5$ in the high- and low-conductance states, respectively.

the activation energies for this switching defect are 124 and 167 meV for the high- and low-conductance states, respectively; these values are typical of single defect states in our junctions.

We have also measured the bias-voltage dependence of the switching RTS for ten defects and have observed two classes of behavior: The first is a simple exponential dependence ($\ln\tau \propto -V_b$) of the switching lifetimes on bias voltage, as observed also by other workers.[2,4] The second is a more complicated exponential dependence, which we have observed for several defects. This complicated behavior is illustrated in Figs. 6 and 7, where the data have

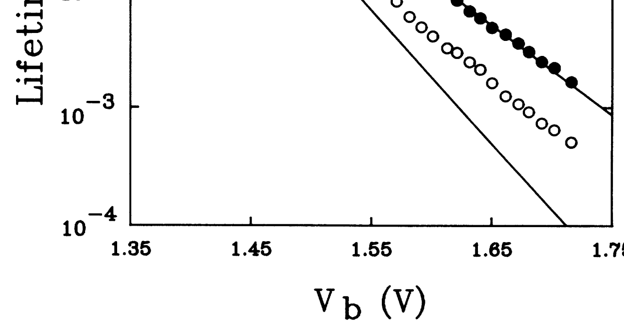

FIG. 7. Plot of high- and low-conductance lifetimes as a function of bias voltage at $T = 6.67$ K. Note that the low-conductance-state lifetime is (nearly) exponential, while the lifetime of the low-conductance state clearly deviates from simple exponential behavior.

been plotted logarithmically with the curvature of the plotted data points on a semilogarithmic scale representing a departure from simple exponential behavior; all data showing this behavior have lifetimes that become less sensitive with increasing bias voltage. Note that for the data in Fig. 6 the slope $d(\ln\tau)/dV$ changes by a factor of 2 for both the high- and low-conductance states. In Fig. 7, we show data for a switching RTS in which the low-conductance state shows nearly perfect exponential behavior, but the high-conductance state clearly shows nonexponential behavior.

IV. DISCUSSION

It is well known that the tunneling current through an insulating barrier can be influenced substantially by electric charges within the barrier. Calculations by Schmidlin[13] suggest that the fractional change of tunnel current, $\delta I/I_{\rm low}$, resulting from electric charge placed into a neutral barrier, may readily be an order of magnitude. For example, if the density of charges uniformly placed within the middle of an Al_2O_3 barrier of thickness d is e/d^3, then $\delta I/I_{\rm low}\approx 400\%$, where the barrier height is assumed to be 1.8 eV. As a rule of thumb, we can presume that a single electron can effectively block conduction through a volume d^3 of the barrier.

This conclusion suggests that discrete resistance fluctuations arising from single trapped electrons can be observed only in uniform tunnel junctions for which L (the transverse dimension of the junction) is not too much larger than d, the barrier thickness. For example, the resistance fluctuations of the discrete RTS across a 1 μm^2 Al_2O_3 barrier tunnel junction having a thickness of 30Å are expected to be less than 0.001%. It is this reason that observations of discrete RTS's have previously been restricted to junctions of typically 1 μm^2 or smaller.

However, paradoxically, it is also possible to observe discrete RTS's arising from single charged defects in extremely large junctions, provided that the junction has a slightly nonuniform barrier thickness. Because the tunneling current varies exponentially with barrier thickness, it is easily shown that nearly all the tunneling current in such cases occurs in a very small section of the junction—that section in which d has its minimum value.

To illustrate this point, consider an $L\times L$ tunnel junction which has $L=50$ μm, barrier thickness $d=30$ Å, and barrier height $E_b=4$ eV. The electrical resistance R of this junction obeys the well-known tunneling relationship

$$R\sim(1/L^2)\exp(dE_b^{1/2}) ,$$

where the equation yields correct numerical values for d(Å) and E_b(eV). Because of the exponential dependence of R on E_b, such a junction will have the same resistance as an extremely tiny junction having $L=30$ Å and $d=20$ Å. Variations of barrier thickness of this scale are unlikely in native oxide barriers, but are easily achieved in synthetic (e.g., Al_2O_3) barriers. In such junctions, the preponderance of tunneling current occurs in the region where d is smallest. Should a switching defect be located in this portion of the junction barrier, extremely-large-amplitude RTS resistance fluctuations may be observed.

We believe this effect is responsible for the giant RTS fluctuations shown in Figs. 2(a) and 2(b). For our samples, the uniformity of the barrier thickness could be controlled by varying the thickness of the Al layer prior to the oxidation step. For Al film thicknesses below about 50 Å, the Al formed discontinuous beads on the surface of the Au, leading to shorted junctions. For Al film thicknesses of about 100 Å, a uniformly oxidized barrier was created which resulted in no detectable RTS. However, in the intermediate regime of 50–100 Å film thickness, large RTS's could be observed, with approximately 70 Å leading to the largest effects. In this thickness range, we believe a continuous insulating layer of Al_2O_3 was created, but with layer thickness fluctuations of about 50% across the sample.

Here we propose a simple model to explain how a single electron trapped on a defect site can lead to the observation of a bias-voltage threshold in our data. As shown schematically in Fig. 8, we consider a defect state within the insulating barrier that has two characteristic energies, $E_{\rm empty}$ and $E_{\rm occ}$, which are associated with the absence and presence, respectively, of an electron trapped on the site, and we further assume that $E_{\rm empty}>E_{\rm occ}$, as shown. We do not at this point address the reason for the difference in the two energy levels. (As mentioned previously, Rogers and Buhrman suggest that the ionic defect itself undergoes a positional transition to an adjacent position in the barrier.) The drawing shows an arbitrary bias voltage V_b that shifts the relative Fermi levels of the two junction electrodes. It is clear that a minimum forward bias voltage V_e is required before charge can tunnel elastically from the Fermi level of the left electrode into state $E_{\rm empty}$. Once this charge is trapped, however, the defect will switch to state $E_{\rm occ}$ and will remain there unless the voltage bias exceeds V_0, the threshold voltage that enables the charge to tunnel into an empty state above E_F on the right electrode. Depending on the relative positions of $E_{\rm empty}$ and $E_{\rm occ}$ with respect to the Fermi level, V_0 can be greater or less than V_e. According to this picture, either $E_{\rm occ}-E_F$ or $E_F-E_{\rm empty}$ (whichever is larger) determines the bias voltage threshold $V_{\rm th}$ observed in our experiments. Note that in the drawing the

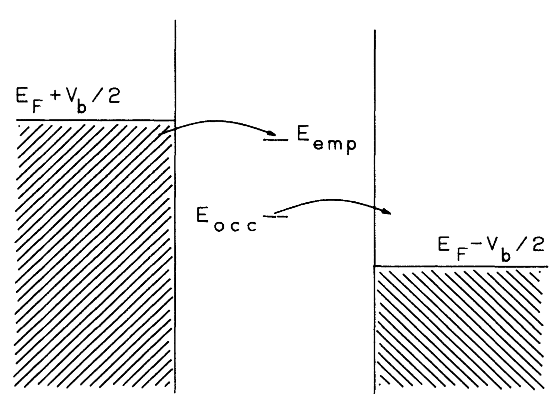

FIG. 8. Energy-level representation of the model discussed in the text. The two electrodes are shown with an applied bias voltage V_b.

threshold voltages will be the same for both the forward and reverse-bias directions. If the defect site is located asymmetrically in the barrier (i.e., closer to one electrode than to the other), however, then the forward and reverse threshold voltages $V_{+\mathrm{th}}$ and $V_{-\mathrm{th}}$ will be different.

If the applied bias voltage V_b is less than V_{th}, then the junction remains in its initial state, which can be either occupied (low conductance) or empty (high conductance). If the initial state is occupied (empty), then $E_F - E_{\mathrm{empty}}$ is larger (smaller) than $E_{\mathrm{occ}} = E_F$. If the bias thresholds of both polarities $V_{+\mathrm{th}}$ and $V_{-\mathrm{th}}$ are known (allowing for the possibility that the defect is not in the center of the barrier), then this energy difference is given by $V_{+\mathrm{th}} V_{-\mathrm{th}} / (V_{+\mathrm{th}} + V_{-\mathrm{th}})$.

According to this picture, it is now possible to provide a qualitative interpretation of the temperature dependence of the lifetimes. As discussed previously, those defects with values of V_{th} (greater than 0.5 V) show tunneling (nearly-temperature-independent) behavior below 100 K. The absence of thermally activated behavior in these samples is presumably a result of the large thermal activation energies associated with these larger values of V_{th}.

We believe the simple exponential behavior of the RTS lifetimes on bias voltage is adequately explained by the model of Rogers and Buhrman.[2,8] However, the curious nonexponential behavior illustrated in Figs. 6 and 7 does not fit the framework of their model.[14] Although we do not understand the reason for this nonexponential behavior, we have investigated several obvious possibilities and have ruled out both the modulation of the effective barrier thickness and the height of the barrier energy with bias voltage.

In summary, we have observed extremely large discrete resistance fluctuations in $50 \times 50\ \mu m^2$ Al_2O_3 barrier tunnel junctions, which we believe result from switching defects located in the narrowest portion of the junction. We have measured the lifetimes of the switching RTS as a function of voltage and temperature and have observed that the lifetimes of some defect states deviate from simple exponential dependence.

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[14]A nonexponential lifetime dependence upon bias voltage has been reported by Ralls *et al.* (Ref. 6) and was interpreted as defect heating. However, our results do not appear to result from defect heating: Since the lifetime *decreases* with increasing temperature, one would expect voltage-induced heatng to decrease the lifetime faster than the simple exponential function, whereas our data show the reverse effect.