# Single-electron tunneling to insulator surfaces detected by electrostatic force

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The detection of single-electron tunneling events between a metallic scanning probe tip and an insulating surface is demonstrated by an electrostatic force method. When a voltage-biased oscillating atomic force microscopy tip is placed within tunneling range of the surface of an insulator, single-electron tunneling events are observed between the tip and electronic states at the surface. The events cause an abrupt reduction in cantilever oscillation amplitude, due to the instantaneous reduction of the force gradient at the tip. In most cases, only a single electron tunnels to or from the surface. Experimental data show that no physical contact is made during the tunneling events. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1525886]

The scanning tunneling microscope (STM) has played a powerful role in understanding and characterizing the atomic scale structure and electronic properties of conducting and semiconducting materials.[1] While the STM provides atomic scale spatial resolution, its application is limited to samples that provide adequate conductivity. A tunneling current typically greater than 1 pA or $\sim 10^7$ electrons/s is required. Any electron state with a lifetime greater than $10^{-7}$ s cannot be directly "seen" by the STM. Therefore direct tunneling to electrically isolated electron states in insulating materials by STM is not possible.

Many attempts have been made over the years to image insulating surfaces with the STM. If an insulating film is thin enough, the STM can be used to tunnel "through" the film, rather than to individual states in the film. Indirect evidence for individual trap states in a thin $SiO_2$ film was observed with STM as excess "telegraph noise" in the tunneling current[2] in early experiments. Attempts have also been made to image the surface of insulators using an alternating current STM.[3] Atomic scale results from this approach, however, have not been convincing. Fowler–Nordheim tunneling has been useful for characterizing some of the properties of thin insulating films,[4] and more recently the surface of diamond.[5] In all of this previous scanning probe work, no direct single electron tunneling to/from individual electron states near an insulator surface has been demonstrated.

In this letter, direct measurement of single electron tunneling events between a metallic tip and an insulating surface is reported. This research builds upon previous work in which electrostatic force detection of single electron tunneling events between a specially fabricated probe and a conducting surface was demonstrated.[6,7] In that work, the probe consists of an oxidized silicon atomic force microscope (AFM) cantilever/tip with a $\sim$100 nm metallic dot at its apex. An electrostatic force microscopy (EFM) method is used to detect the charge on the dot. An electron tunneling to or from the dot causes an abrupt change in the electrostatic force signal detected.

The same principle can be applied to a standard metallic tip and a localized electronic state on the insulator surface. In this case, the state on the surface acts as the "metallic dot" in the previous work.[6,7] When a single electron tunnels to a localized, electrically isolated state at the surface of the insulator, the electrostatic force and force gradient on the tip is abruptly reduced, producing a detectible cantilever response.

Several EFM modes can be used to observe single electron tunneling events. In the method described here, the cantilever is mechanically oscillated below its resonance frequency with a piezoelectric bimorph, and a dc voltage is applied between the tip and back contact of the sample. The applied voltage creates an electrostatic force gradient at the tip. When the tip is brought close to an insulating surface, and an electron tunneling event occurs, the electrostatic force gradient is reduced, shifting the cantilever resonance frequency to a higher value (further from the drive frequency). This abruptly reduces the amplitude of the oscillation amplitude of the cantilever.

It is important to note that if a *metallic* tip is placed near a *metallic* sample, tunneling may also occur. However, it will not be detected (change the cantilever amplitude), as it does not change the electrostatic potential (and force gradient) between the tip and sample. In this situation, the potential difference is fixed by the voltage source. It is the presence of the localized electronic state (with long lifetime), which allows the event to be detected.

Figure 1 contains a schematic diagram of the experimental setup for an oxide sample, along with an equivalent circuit of the electrostatic measurement. The capacitance between a metallic tip (above an oxide surface) and the back contact of the sample includes contributions from two series capacitances, the tip–surface capacitance $C_{ts}(z)$ and the oxide capacitance $C_{ox}$. The two capacitors act as a voltage divider. If the system is modeled using a parallel plate approximation, the electrostatic force gradient acting on the tip is

$$F' = \frac{C_{ox}^3 C_{ts}(z)}{[C_{ox}+C_{ts}(z)]^3}\frac{V_{dc}^2}{z^2} = \frac{C_{ox}C_{ts}(z)}{[C_{ox}+C_{ts}(z)]}\frac{V_{ts}^2}{z^2},$$

where $z$ is the gap (vacuum), $V_{ts}$ is the potential difference

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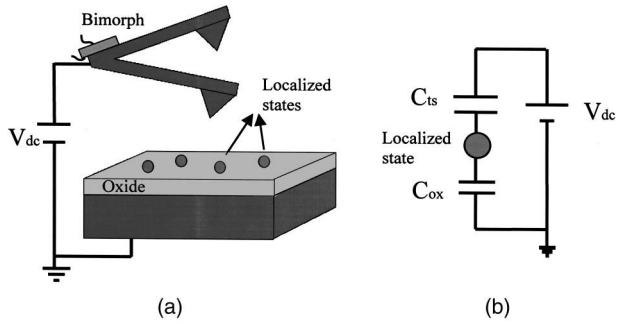


FIG. 1. (a) Experimental setup. (b) Equivalent electric circuit for electron tunneling measurements. $C_{ox}$ is the oxide capacitance and $C_{ts}$ is the tip–sample capacitance.

between the tip and sample (oxide surface), and $V_{dc}$ is the applied voltage between tip and back contact.

A localized, isolated electronic state on or near the oxide surface can be modeled as a floating metallic island. If electrons at the Fermi-level of the tip see an unoccupied state at the same potential near the sample surface, tunneling will occur between the tip and the state with a probability that depends upon the tunneling barrier height and width. The sign of the applied dc voltage determines whether an electron will tunnel to or from the electronic state. When an electron (of charge $e$) tunnels, the magnitude of the potential difference between the tip and the surface will always be reduced according to the following formula:

$$|V_{ts}| = |V_{dc}| \frac{C_{in}}{C_{in}+C_{ts}} - \frac{|e|}{C_{in}+C_{ts}}.$$

Since the force gradient is proportional to the square of the voltage between tip and surface $V_{ts}$, a tunneling event will always cause the force gradient to decrease, causing a reduction in the oscillation amplitude (for drive frequencies below resonance).

Single-electron tunneling measurements are performed by positioning a platinum coated silicon AFM tip above a high quality 20-nm-thick $SiO_2$ film on a silicon substrate at a distance of $\sim$2 nm. The measurements are made in high vacuum at $10^{-9}$ Torr after heating the sample to 650 °C for 45 min. The cantilever resonance frequency is 282 kHz, cantilever spring constant $k = 50$ N/m, and the drive frequency is 3200 Hz below resonance. The tip is scanned to/from the surface, while recording the amplitude and phase of the electrostatic force signal at the drive frequency $\omega$ (with a lock-in amplifier). No feedback loop is used. For applied dc voltages $V_{dc}$ greater than 3.5 V or less than $-3.5$ V (relative to flatband), single electron tunneling events are often observed as the tip approaches the sample. Calculations show that the 3.5 V bias corresponds to $\sim$1 V drop between the tip and the oxide surface with a 2 nm gap and a 20 nm oxide thickness.

In Fig. 2(a), it can be seen that as the tip is scanned toward the sample, the EFM signal rises slowly. At one point, an abrupt drop in amplitude occurs, signaling a single tunneling event. In this case, when the electron tunnels to a state at the surface, the force gradient instantaneously decreases, causing the cantilever oscillating amplitude to drop by 0.03 nm. This reduction in amplitude takes the tip out of tunneling range. As the tip continues its movement toward the surface, the amplitude begins to increase again (due to closer proximity). In this data set, no further tunneling occurs before mechanical instability causes the amplitude to abruptly jump to a higher level. This instability has been observed and studied previously.[8] Its importance to this work is only to point out that if mechanical instabilities occur before tunneling takes place, single electron tunneling events will not be observed. In these measurements, the mechanical instability always abruptly increases the oscillation amplitude, while the tunneling events always cause an abrupt decrease in amplitude.

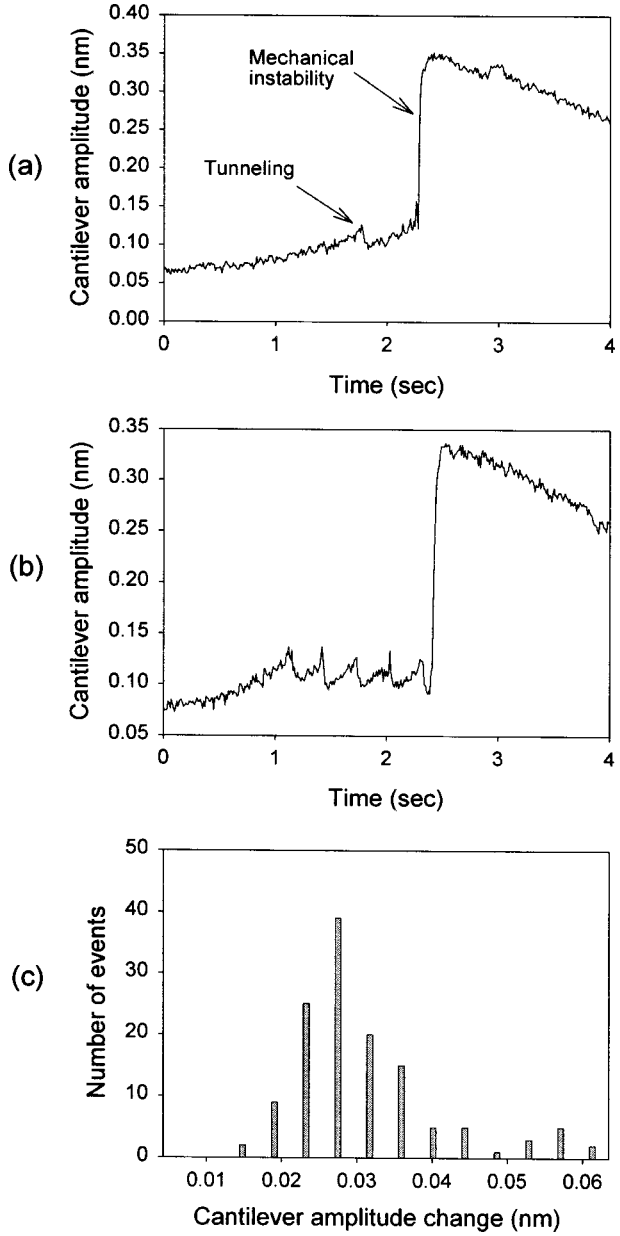


FIG. 2. (a) A single electron tunneling event is observed as the oxide sample is scanned toward the tip. (b) Five consecutive tunneling events detected at a second location on the oxide surface as it approaches the tip. (c) A histogram of the amplitude change occurring for 131 different tunneling events at different locations on the oxide surface. The first peak is centered on an amplitude change of 0.028 nm (single electron tunneling). A second peak is found at 0.056 nm amplitude change (two electrons tunneling).

In Fig. 2(b), five consecutive tunneling events are observed at another location before the instability point. This may be explained by the presence of more than one available state in this region of the oxide surface. Another possible

explanation is that a single state exists below the tip, with a lifetime that is large enough to be detected (lock-in time constant 10 ms), but smaller than the time between events (~.3 s). Note the uniformity of the amplitude change for the five events. Hundreds of measurements like those shown in Figs. 2(a) and 2(b) show that the number of electrons tunneling to the $SiO_2$ surface depends upon the location on the sample. In most regions, only single tunneling events are observed, while in a few others, multiple events are seen before mechanical instability occurs. Tunneling events have also been observed to the surfaces of mica and GaAs.

To establish that each event shown in Fig. 2 is due to a single electron, the magnitude of the abrupt amplitude change was measured for 131 different tunneling events. The data was taken at many different locations on the surface, at both positive and negative bias voltage. A histogram of the results is shown in Fig. 2(c). A bin size of 0.005 nm in this histogram is used.

Since the cantilever amplitude change occurring with each event is proportional to the number of electrons transferred, the amplitude change is expected to be quantized. The measurements are performed by scanning the tip *continuously* toward the surface until tunneling range is reached. The most likely event under this condition is for a single electron to tunnel. The first peak in the histogram (0.028 nm) should correspond to single tunneling events. If that peak corresponded to a charge transfer of multiple electrons, it would be expected that other peaks would exist at smaller values. However, this is not the case. Since the histogram shows that there are no peaks below the peak centered on 0.028 nm, this peak corresponds to single electron events. Note that there is a small second peak around 0.057 nm, which is almost twice the magnitude of the first. This peak corresponds to two-electron events.

The width of the main peak in the histogram may be understood in two ways. First, the finite signal-to-noise ratio causes a variation in the magnitude of the amplitude change. Second, the variations in the surface potential from location to location and depth of the electronic states in the oxide are also expected to broaden the histogram peaks.

To prove that charge transfer does not occur by physical contact, the average optical deflection signal from the cantilever is recorded simultaneously with the cantilever oscillation amplitude. The deflection signal is low pass filtered at 30 Hz to eliminate the ac component of the cantilever motion at the drive frequency. For the dc voltages typically applied (>3.5 V), the cantilever is slightly bent toward the surface (attractive force) as it approaches. In Fig. 3, a single tunneling event is shown before the mechanical instability occurs, with a simultaneous measurement of the average optical deflection signal. It is observed that during the tunneling event, the cantilever is in the attractive regime (deflection signal negative) and does not make contact with the surface. After the event, as the sample continues toward the tip, eventual contact is made (point at which the slope of the deflection curve changes sign). With further movement, the force becomes net repulsive (deflection positive). The optical deflection data supports the conclusion that the tunneling events occur before contact with the surface is made. Based upon the piezotube scanner calibration, in this data set a single electron tunneling event occurs at a gap of approximately 1.2 nm.

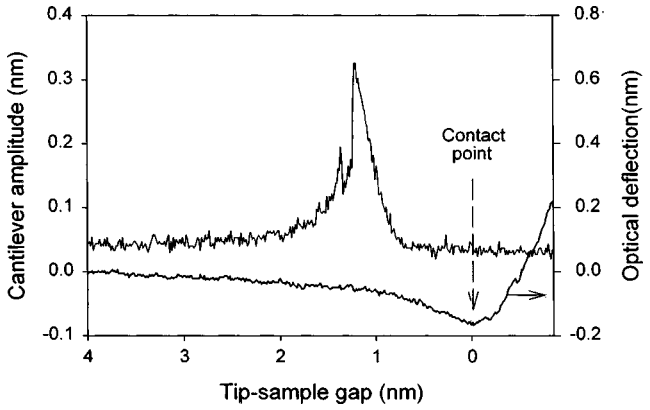


FIG. 3. A tunneling event detected in the oscillation amplitude before mechanical instability occurs, with the corresponding optical deflection signal as the sample approaches the tip.

In summary, single electron tunneling to insulator surfaces has been demonstrated by a scanning probe method. The distribution of the magnitude of the events shows that the charge transferred is quantized, with almost all events corresponding to a single electron. It is shown that no contact is made with the sample surface during the events. This single electron tunneling approach, in principle, will provide a method to characterize both the location and energy of localized, isolated electronic states in or on insulating materials with *atomic scale spatial resolution*. This information is not available from existing scanning probe methods.

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