

Instability induced tunneling and repeatable charge injection to SiO₂ surfaces by electrostatic force microscopy

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(Received 5 April 2004; accepted 7 June 2004)

The dynamic response and stability of a voltage biased oscillating cantilever in the proximity of an insulating sample surface is investigated. As the tip approaches the sample surface, the cantilever can jump between two different oscillation modes. The jump is detected as an abrupt increase in the amplitude. This abrupt increase in amplitude can bring the tip into tunneling range of the sample surface and electrons can tunnel between the metallic tip and insulator surface. The electron tunneling decreases the electrostatic force and force gradient sensed by the cantilever. These changes reduce the oscillation amplitude, causing the cantilever to return to the original oscillation mode. The tunneling of tens of electrons from a metallic tip to a SiO₂ surface is detected by measuring the electrostatic force between the tip and the sample. Manipulation of a small number of electrons can be achieved by changing the polarity of the voltage applied, allowing the injection and extraction of electrons to and from an insulator surface. © 2004 American Institute of Physics. [DOI: 10.1063/1.1777397]

I. INTRODUCTION

Electrostatic force microscopy (EFM) is a powerful method to characterize the electrical properties of thin dielectric films.¹ In EFM a voltage biased metallic cantilever is oscillated in the proximity of a sample surface while recording the change in the amplitude and phase of the cantilever due to variations of the electrostatic force [Fig. 1(a)]. The highest sensitivity of the EFM is achieved if the cantilever is brought as close to the sample surface as oscillation stability allows and the oscillation is close to the resonance frequency.²

When the amplitude of the sinusoidal oscillation of the cantilever is small compared with the average tip sample gap, the force acting between the tip and sample is approximately sinusoidal. Under this condition, the resonance curve is Lorentzian in shape and there is only a single-amplitude value which corresponds to a given gap and drive frequency.² Under typical experimental conditions (drive frequency below resonance), as the tip approaches the sample, the oscillation amplitude increases. When it becomes a finite fraction of the average gap, a nonsinusoidal force on the tip is produced. During a cycle of the oscillations, the magnitude of the force sensed by the cantilever becomes much larger when the tip is closest to the sample as compared to when it is furthest from the surface. Due to this nonlinear force sensed by the tip, the resonance curve becomes distorted and multiple values of the amplitude are possible for a given gap and drive frequency. This causes an instability as the tip approaches the surface, which is described and discussed in detail in Ref. 3.

Figure 1(b) shows an example of this instability as the tip approaches the surface. The sudden change of the oscil-

lation amplitude (instability) is related to the transition between two different oscillation modes of the cantilever.³ Switching between two amplitude values was observed previously even for cases when no voltage was applied between tip and sample.⁴ Various theoretical models have been developed that explain the multiple solutions of the cantilever oscillations in proximity to the sample.³⁻⁵

In recent experiments of single-electron tunneling^{6,7} between a metallic tip and an insulating surface, it has been observed that the gap where the tunneling occurs can be very similar to the gap where the instability is encountered [Fig. 2(a)]. To detect single-electron tunneling events, the cantilever should be mechanically oscillated at a very small drive amplitude, a couple of thousand hertz below its natural resonance frequency. If the drive amplitude of the cantilever is increased, the instability occurs further away from the sample surface, well before single-electron tunneling events are observed. When this large amplitude instability occurs, the minimum tip-sample gap (distance between the oscillating tip end and surface) is abruptly reduced and the tip can come very close to the sample surface without making contact with it. Electrons can tunnel between the tip and the surface at this small gap. The sign of the dc voltage applied between tip and sample controls the direction of the tunneling. In this paper the instability induced tunneling of tens of electrons to and from an insulating surface is investigated.

II. EXPERIMENTAL DETAILS

The EFM measurements are performed in ultrahigh vacuum (base pressure 10⁻⁹ Torr) and the sample used is a 20 nm high quality thermally grown SiO₂ film on Si substrate. The cantilever is mechanically oscillated at a frequency below its natural resonance frequency, by applying an ac voltage to the piezoelectric bimorph attached to the back of the cantilever [Fig. 1(a)]. The drive frequency is chosen so that when the tip comes close to the sample, the

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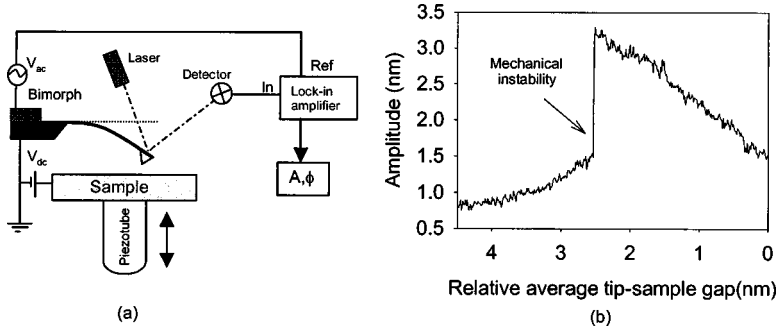


FIG. 1. (a) Schematic diagram of the experimental setup for cantilever amplitude and phase measurement of a voltage biased sample. The tip-sample gap is modulated periodically by moving the piezoelectric tube up and down. (b) A typical abrupt increase in amplitude (mechanical instability) is detected as the sample is moved toward the tip.

cantilever resonance frequency approaches the drive frequency. A dc voltage is applied to the sample with respect to the Pt coated cantilever (± 5.6 V for these experiments). The tip-sample gap is modulated periodically by applying a triangular voltage to the piezotube attached to the sample. The voltage applied to the piezotube moves the tip periodically toward and away from the sample surface. Dynamic amplitude and phase curves are acquired as the tip moves relative to the sample.

Figure 2(b) shows the amplitude variation for a voltage biased cantilever as it approaches a SiO_2 surface. As the tip-sample gap is reduced, a gradual increase in amplitude is detected due to the increased force gradient that shifts the resonance frequency toward the drive frequency. At an average gap of 9 nm, a rapid increase in the amplitude is observed, followed by an immediate abrupt decrease in amplitude. This abrupt increase and decrease in the amplitude [Fig. 2(b)] is much larger in magnitude than that of the single-electron tunneling event shown in Fig. 2(a).

The rapid increase is caused by the previously described mechanical instability encountered by the cantilever.³ The cantilever oscillations make an abrupt transition from one oscillation mode to another. When this abrupt transition occurs, the tip-sample minimum gap suddenly decreases. If this minimum gap falls below that needed for electron tunneling, electrons can tunnel between the tip and surface. This charge can fill up (or empty) trap states in the surface near the tip, modifying the local surface potential. The charge transfer causes an abrupt decrease in the electrostatic force gradient acting between tip and sample. The decreased force gradient causes the oscillation amplitude to jump back to the initial oscillation mode, corresponding to a smaller amplitude. After the charge transfer, the oscillating tip continues to approach the sample surface. A second point of instability is encountered and the amplitude jumps again to a larger value [see

Fig. 2(b)]. Since at this point, the charge transfer has already occurred, no electrons tunnel, and the oscillation remains in the second oscillation mode. By further moving the tip toward the surface, a gradual decrease of the amplitude is observed as the cantilever follows the large amplitude oscillation mode.³

To clarify the charge transfer mechanism observed in Fig. 2(b), the average optical deflection signal is recorded simultaneously with the oscillation amplitude as the tip is moved toward the surface (Fig. 3). The optical deflection signal is extracted from the split photodiode after filtering out the high frequency (150 kHz) oscillation signal. The optical deflection signal is low-pass filtered (cutoff frequency 100 Hz). Monitoring the optical deflection is a very sensitive way to detect the average force on the tip.⁶ In Fig. 3(a) the cantilever oscillation amplitude variation is shown as the tip approaches the sample with the first instability and tunneling (abrupt rise and fall) followed by the second instability. The corresponding optical deflection signal shows an attractive (negative) force between tip and sample before and during the instability and charge transfer (no contact made). After the charge transfer, further reduction of the average tip-sample gap produces a less attractive deflection (until the second instability is reached). This less attractive deflection is due to a reduced force on the tip caused by the charge transfer and the reduced amplitude (greater minimum gap).

In Fig. 3(c) the amplitude variation versus gap is shown over a larger range of tip-sample gap. In this data set, the tip is moved toward the sample until contact is made. As the tip approaches the sample, the oscillation amplitude at a certain gap becomes zero. This is the snap-in point where the spring constant of the cantilever equals the attractive force gradient sensed by the tip. This is also the point where the tip starts to make contact to the surface [shown in Figs. 3(c) and 3(d)].

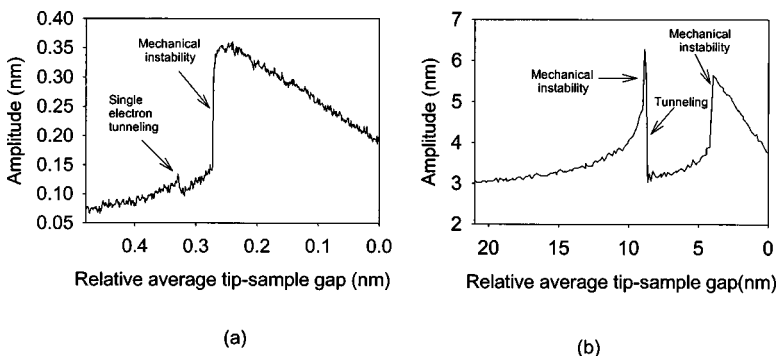


FIG. 2. (a) A single-electron tunneling event is followed by an instability in oscillation amplitude as the sample approaches the tip. (b) For a cantilever driven at large amplitude as the tip approaches the sample, an abrupt jump to large amplitude (mechanical instability) is instantly followed by a decrease in amplitude (tunneling) before the tip moves to a position at which it jumps to large amplitude again (second mechanical instability).

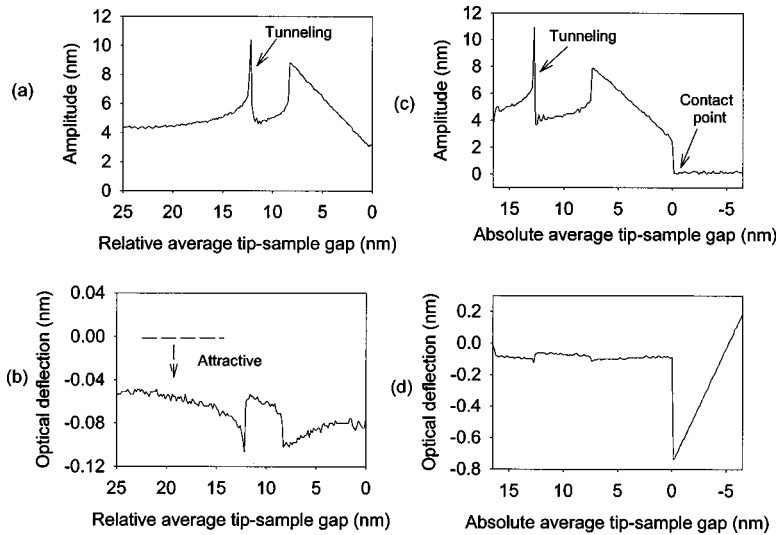


FIG. 3. Oscillation amplitude (a), (c) and optical deflection signal (b), (d) as the tip is moved toward the surface without contacting (a) and until contact is made (c) with the surface. When the tunneling occurs (abrupt decrease in amplitude) no contact is made between tip and sample.

After the cantilever snap-in to the surface, the negative optical deflection signal becomes more positive as the tip is further pushed toward the sample surface. In this regime, a combination of attractive and repulsive forces is sensed by the cantilever. Figure 3(d) shows that in both situations, when the charge is initially transferred after the first instability and the second instability is encountered, the cantilever senses a purely attractive force and no contact is made between the tip and sample. Once the cantilever starts to sense repulsive forces, other charge transfer mechanisms (such as contact charging) may occur.

A major difference between single-electron tunneling [Fig. 2(a)] and the charge transfer observed in Figs. 2(b) and 3 is the large “spike” in the cantilever amplitude before this charge transfer occurs. For the single-electron tunneling case (small amplitude drive), the tip-sample gap is continuously reduced and no mechanical instability is observed before tunneling occurs. This fine control of the gap allows reaching a condition where single-electron tunneling detection is possible. In the case of large drive amplitude, the instability causes an abrupt amplitude increase (tip-sample gap is rapidly reduced) and an uncontrollable number of electron transfer events often occur. The minimum tip-sample gap distance in Fig. 3(c) is ~ 1.7 nm, which is within the tunneling range for electrons.³ Since no contact is made between tip and sample, the most probable charge transfer mechanism in these experiments is tunneling. Whether tunneling occurs is partially determined by the mechanical instability in the cantilever oscillation. Simulation of the amplitude as the average tip-sample gap is reduced allows a prediction of whether the instability will bring the tip within tunneling range.

III. SIMULATIONS

For a mechanically oscillated cantilever, the amplitude and phase of the oscillations are modified by the force gradient sensed by the tip. For simulating the dynamic response of the oscillating cantilever, only the long-range electrostatic force acting between tip and sample is included in the model. The details of this simulation were presented previously.³

The model assumes that the average tip-sample gap is modulated by the movement of the base of the cantilever. The cantilever motion causes a modulation of the electrostatic force (dependent on the tip-sample gap). During one cycle of oscillation the cantilever will sense a much larger force when it is near the sample surface as compared when it is farther away. The periodic electrostatic force is Fourier decomposed in a series of terms at harmonics of the modulation frequency. Assuming a sinusoidal motion of the tip (consistent with high quality factor), the differential equation that describes the cantilever motion can be solved analytically.³

Two characteristics of the cantilever response, as the tip-sample gap is reduced, are the shift of resonance to a lower frequency and the change of the resonance curve from a Lorentzian shape to a distorted resonance curve. Two branches of the resonance curve can be distinguished, the left branch that describes the cantilever response for frequencies below resonance and the right branch that describes the cantilever response for frequencies above and a finite range below the cantilever resonance. As the tip-sample gap is reduced the resonance curve becomes more distorted and shifted toward lower frequency values.³ In Fig. 4 the resonance curves at different tip-sample gaps are shown. Far from the surface the cantilever is on the lower left branch [Figs. 4(a) and 4(b)] of the resonance curve (lower amplitude). As the tip-sample gap is reduced the resonance curve is shifted toward lower frequencies and gets more distorted. In addition the amplitude is increasing in magnitude. If the drive frequency is fixed as in the present experiment [the drive frequency is marked by a straight line in Figs. 4(a)–4(f)], the left branch of the resonance curve can shift so much toward the left that there is a gap below which there is no possible solution on the left branch. Once this gap is reached the cantilever oscillation jumps to the right branch [Fig. 4(c)]. The transition from the left branch (lower amplitude) to the right one (higher amplitude) will cause an abrupt increase in the amplitude. The increased amplitude will bring the tip very close to the surface where electrons can tunnel. The modification of the surface potential, once the electrons tunnel, causes a reduction in the force gradient sensed by the cantilever. The smaller force gradient causes the resonance to

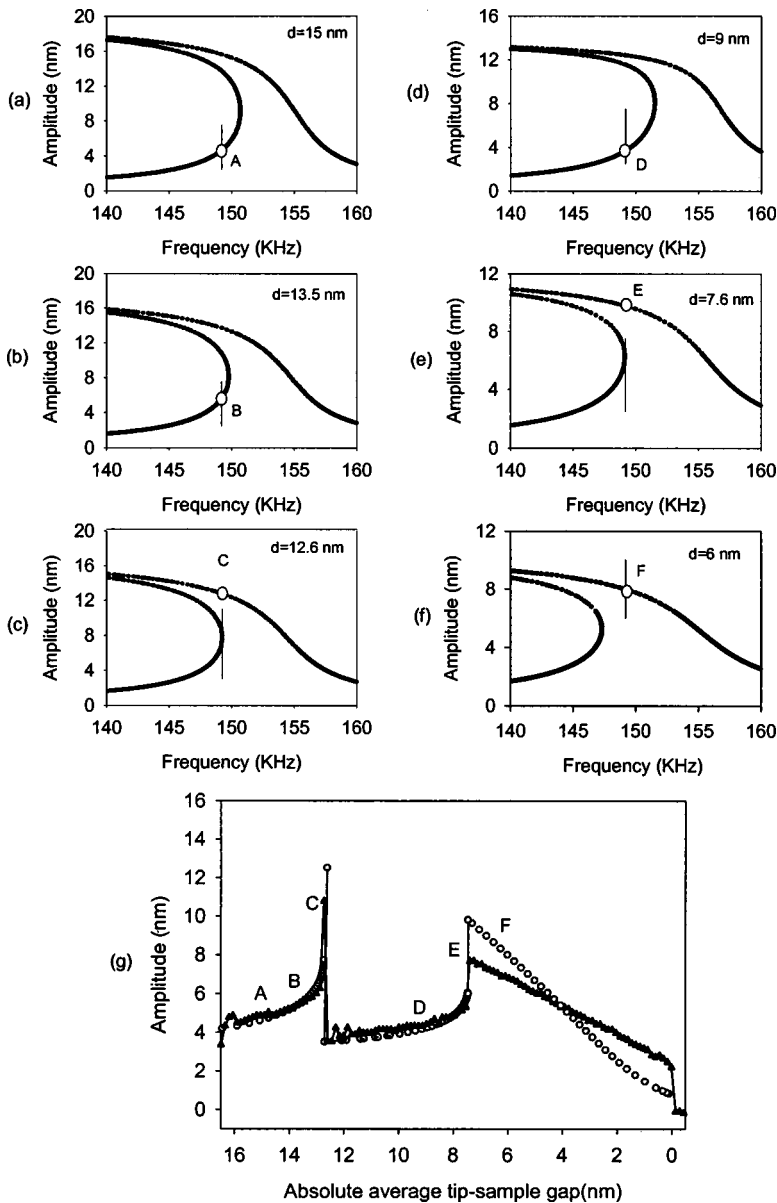


FIG. 4. Simulated resonance curves of the cantilever oscillation (a)–(f) for decreasing tip-sample gap before (a)–(c) and after (d),(e),(f) electron tunneling. When the tip reaches the gap where no solution is possible on the left branch of the resonance curves (c),(e) the oscillation jumps to the right branch (large amplitude). (g) A comparison between experimental and simulated cantilever response versus average gap with tip-sample gaps labeled with letters corresponding to the resonance curves shown in (a)–(f). The experimental result is shown with triangles and the simulation result with circles.

shift to higher frequency, and the cantilever jumps back to the lower left branch of a new resonance curve.

Once on the left branch (lower amplitude) of the new resonance curve, as the tip-sample gap is further reduced, the cantilever oscillation will follow again the left branch [Fig. 4(d)] until it encounters the gap where no solution exists on the left branch and jumps to the right branch [Fig. 4(e)]. Since the surface is already charged, no further electrons can tunnel to the surface and the cantilever oscillation remains on the new branch mode. This second mechanical instability is characterized by a jump to larger amplitude and once on the right branch, the cantilever oscillation follows this branch (amplitude decreases) as the gap is further reduced [Fig. 4(f)].

Figure 4(g) shows the amplitude variation versus tip-sample gap, with the marked gap values where the resonance curves [Figs. 4(a)–4(f)] are plotted. The simulations predict well the gaps where the two instabilities occur and also the jump in amplitude when the instability is encountered. The experimental conditions are: resonance frequency of the can-

tilever 155.68 kHz, drive frequency 6.486 kHz below resonance, quality factor 25 946, oxide thickness 20 nm, and the dc voltage applied to sample is 5.6 V. For the simulations, the radius of the tip ($r=37$ nm) and the spring constant of the cantilever (1.8 N/m) are adjusted to fit the experimental curve and the values used fall in the ranges specified by the manufacturer.⁸

After fitting the experimental data, the abrupt decrease in amplitude associated with tunneling corresponds to ≈ 40 electrons. These electrons may tunnel to trap states near the SiO₂ surface or states inside the oxide film. For thermally annealed SiO₂ films on silicon the measured defect density^{1,9} is often in the range of 10^{12} cm⁻². This density would correspond to tens of available states underneath the tip. These states are found on the surface, inside the dielectric film, and at the interface between the SiO₂/Si. At certain locations on the SiO₂ surface, a single-tunneling charge transfer event (spike) is observed, as shown in Figs. 2(b) and 3. At other locations, a second-tunneling charge transfer event may be observed. In Fig. 5, a trace is shown where two instabilities

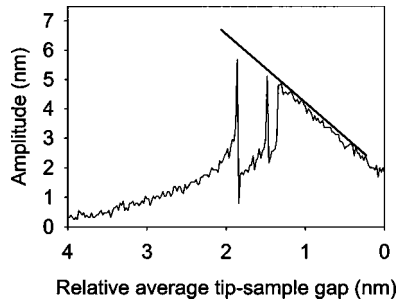


FIG. 5. Two consecutive tunneling events mediated by mechanical instability, before a final mechanical instability. The dotted line shows the alignment of the peak of the amplitude spikes with the larger amplitude mode.

and tunneling charge transfer events are detected before the cantilever jumps permanently to the second (larger) oscillation mode. The spikes in amplitude for the two tunneling cases line up with the amplitude change expected for the larger oscillation mode (a line is drawn to show the aligning of the amplitude values). Consecutive tunneling may occur if in a previous tunneling process not all the available states are filled/emptied. While over certain locations on the SiO₂ surfaces several consecutive tunneling events can be observed, the most common case is a single-mechanical instability and tunneling charge transfer event.

For a clean metallic surface, no charge transfer is detected, because any electrons that tunnel are compensated instantaneously by the applied voltage source. The presence of long lifetime states (greater than the measurement time) makes possible the detection of the tunneling events between the metallic tip and the insulating surface. In contrast to our previous single-electron tunneling experiments,⁶ the tunneling process detected in this experiment is determined by the instability of the cantilever oscillation, which abruptly reduces the tip-sample gap such that electrons can tunnel between the tip and the insulating surface. The number of electrons that tunnel depends on the density of surface states on the thin dielectric film, the parameters of the tip, and the applied voltage.

IV. CHARGE TRANSFER BETWEEN TIP AND SAMPLE

The direction of the electron tunneling process depends on the sign of voltage applied between tip and sample. If the sample is biased at a negative voltage with respect to the tip, electrons will tunnel from the surface to the tip. For a positive voltage on the sample, electrons will tunnel from the tip to the sample surface. To explore the possibility of two way electron transfer between tip and sample, the dc voltage polarity applied to the sample was changed, allowing for the transfer of electrons to or from the sample surface. Two consecutive traces are acquired as the tip approaches and is retracted from the surface, for a chosen polarity of a dc voltage.

Figure 6(a) shows a schematic of the cantilever motion and the change in amplitude as the tip twice approaches and is retracted from the sample. Initially the cantilever is far from the surface (A) and as it comes closer to the surface the amplitude abruptly increases as it encounters the first instability and tunneling (B). It then continues toward the surface

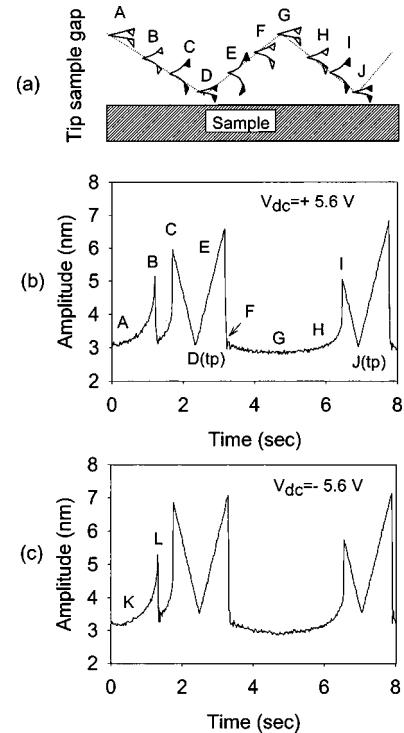


FIG. 6. (a) Schematic of the cantilever motion. Amplitude response of the oscillating cantilever for two consecutive approaches and retractions from the surface for a positive (b) and a negative (c) voltage applied to the sample. Tunneling occurs only during the first approach to the sample after the polarity of the voltage applied to the sample is reversed. Points *D* and *J* are the turning points (tp) as the direction of the cantilever motion is reversed and the tip is retracted from the surface.

where it encounters the second instability point and jumps to the large amplitude mode (C). The tip is further pushed toward the surface while in the larger amplitude mode until the direction of motion for cantilever is reversed (turning point, D). The tip is then pulled away from the surface following the larger amplitude mode (E) until the oscillations fall to the lower amplitude mode (F). In Fig. 6(a), the cantilever is darkened at those positions where it is found in the larger amplitude mode. The tip is retracted from the surface (G) until it reaches the initial height above the surface, as seen at (A). Then a different approach starts. The amplitude increases again as the tip approach the surface (H) until the tip encounters a new instability point and jumps to the larger amplitude mode (I) and then follows this mode as long as the tip is pushed toward the surface (J). At this point (J) the tip is retracted from the surface.

In Fig. 6(b), the amplitude trace is shown for a positive voltage (+5.6 V) applied to the sample. On first approach, the cantilever reaches the first mechanical instability (B) and electron tunneling occurs, causing the fall of the oscillation amplitude to a lower amplitude. The second instability is then encountered (C) and the cantilever jumps to the higher amplitude mode. Since all the surface states are filled, no more tunneling is possible and the oscillation remains at the larger amplitude mode. The tip is further moved toward the surface until the direction is reversed (D) and the tip starts to retract. The tip follows the large amplitude mode (E) and then falls back to the lower amplitude mode (F). It eventu-

ally reaches the starting point (G). On the second approach at the same bias voltage (+5.6 V), the amplitude increases (H) until the first instability point is encountered (I). However, since all the surface states were already filled from the previous tunneling charge transfer event (B), no more tunneling occurs and the tip stays in the larger amplitude mode (I to J). At (J), the direction of the cantilever motion is reversed and the tip is retracted from the surface. This trace shows that since during the first approach, electrons tunnel (B) and fill up the trap states, no other tunneling is possible when the sample is approached a second time.

If the sign of the voltage applied to the sample is reversed (-5.6 V) after the two approach/retract cycles shown in Fig. 6(b), then as the tip approaches the sample again (K), new tunneling events are detected again after the first mechanical instability (L). In this case, electrons are moved from the surface to the tip. If the tip is withdrawn and approaches a second time while the same negative voltage is applied, no tunneling charge transfer event is detected. This is due to the fact that the electrons that filled the surface states (L) had already tunneled to the tip during the first approach. These measurements show that the electron lifetime of the surface states filled by tunneling is larger than 7 s for the sample investigated, as no tunneling is detected when the cantilever approaches the surface a second time [Figs. 6(b) and 6(c).] Previous studies of contact charging SiO₂ surfaces showed that the time for charge dissipation can be larger than tens of minutes.¹⁰

This transfer of the charge back and forth between tip and sample has been observed many times and tunneling events are always detected during the first approach to the

sample after the voltage is reversed. If the voltage is unchanged, subsequent approaches of the tip of the sample do not show any charge transfer. The fact that no electrons tunnel if the surface states are filled/emptied, and that charge can be transferred only if the voltage sign is reversed, further supports the claim that the abrupt decrease in amplitude is due to tunneling. Since the direction of electron tunneling is controlled by the polarity of the dc voltage applied to the sample, the injection or the extraction of a small number of electrons from an insulating surface can be achieved.

V. SUMMARY

The stability of an oscillating cantilever in the proximity of a sample surface is simulated and investigated experimentally. The instability that is encountered is due to nonlinear cantilever dynamics when it is near the surface. The abrupt increase in the amplitude caused by this instability can take the tip into tunneling range of the surface and electrons can be transferred between tip and sample. The repeatable injection and extraction of tens of electrons between tip and insulating sample is demonstrated.

¹R. Ludeke and E. Cartier, Appl. Phys. Lett. **78**, 3998 (2001).

²Dror Sarid, *Scanning Force Microscopy with Application to Electric, Magnetic and Atomic Forces* (Oxford University Press, New York, 1994).

³L. J. Klein and C. C. Williams, J. Appl. Phys. **95**, 2547 (2004).

⁴A. Kühle, A. H. Sorensen, and J. Bohr, J. Appl. Phys. **81**, 6562 (1997).

⁵L. Nony, R. Boisgard, and J. P. Aimé, J. Chem. Phys. **111**, 1615 (1999).

⁶L. J. Klein and C. C. Williams, Appl. Phys. Lett. **81**, 4589 (2002).

⁷L. J. Klein and C. C. Williams, Appl. Phys. Lett. **79**, 1828 (2001).

⁸NSC14/Ti-Pt from www.spmtips.com

⁹R. Ludeke and H. J. Wen, Appl. Phys. Lett. **71**, 3123 (1997).

¹⁰T. Uchihashi, T. Okusako, Y. Sugawara, Y. Yamanishi, T. Oasa, and S. Morita, J. Appl. Phys. **79**, 4174 (1996).

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