TITLE:

Atomic-Level Stick-Slip

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SYNONYMS:

Atomic-scale stick-slip
Atomic lattice stick-slip
Atomic stick-slip

DEFINITION:

Atomic-level stick-slip refers to the behavior of a sliding interface, usually an atomic force microscope tip sliding along a crystalline surface, whereby the tip sticks and then slips laterally with respect to the surface in a periodic fashion. The periodicity coincides with the surface lattice.

SCIENTIFIC FUNDAMENTALS:

History of atomic-level stick-slip

Atomic-level stick-slip friction behavior is a widely observed phenomenon in atomic force microscopy (AFM) and has been reviewed in detail in the literature.[1-3] Atomic-level stick-slip was first discovered by Mate *et al.* who were measuring friction with the AFM between a tungsten tip and a graphite (0001) surface.[4] The lateral force exhibited stick-slip behavior with the periodicity of the graphite lattice. Since then, atomic-level stick-slip behavior has been observed on a wide range of materials, from soft materials like stearic

acid crystals with silicon nitride tips to a diamond tip on a diamond surface. Typical atomic-level stick-slip behavior is illustrated in Fig. 1. This example shows the lateral force experienced by a silicon nitride tip sliding from left to right on the (0001) surface of muscovite mica. In the image, one sees a periodic lattice. The line trace shows that the lateral force starts from zero and builds up to a maximum value. During this phase of the measurement, the tip is sticking to the surface with no relative slip (although there may be some lateral deformation of the tip and sample). The arrow indicates the occurrence of the first slip event. The slip involves the tip moving the equivalent of one lattice spacing along the surface. The tip then sticks again until the maximum lateral force is reached once more, and the next slip occurs, and so on. The periodicity of the slip events is 0.529 nm, which is equal to the lattice constant of the mica (0001) surface. The well-defined force at which the tip slips, F_f, is called the static friction force.

The AFM signals measured correspond to the slope of the end of the AFM cantilever beam, which bends or twists due to forces normal (F_z) or parallel (F_x, F_y) to the surface, as illustrated in more detail in Fig. 2. Morita *et al.* have carried out a systematic study of atomic-level stick-slip on a range of materials, demonstrating precise determination of the slip motions that take place.[1] As seen in Figs. 2 and 3, torsional or buckling rotations at the end of cantilever occur due to frictional forces acting either transverse (F_x) or parallel (F_y), respectively, to the long axis of the cantilever's projection onto the sample. The data in Fig. 3, and from many other experiments, demonstrate that, on an ordered sample, the tip, whose surface atoms are not necessarily ordered, prefers to reside in positions *in registry with the sample lattice*. More on the importance of interfacial commensurability will be discussed below. This periodic interaction is responsible for *all* atomic-level contrast images obtained with contact-mode AFM. One must not imagine the AFM tip smoothly tracing out atomic corrugations as with a scanning tunneling microscope (STM), but instead realize that the relative tip-sample motion is *discontinuous*.

The first few observations of this phenomenon were acquired with highly anisotropic samples, such as graphite and mica, which exhibit strong covalent bonding within each layer but weaker van der Waals or electrostatic forces between the layers. These materials cleave easily to expose their basal planes. It was suggested that the periodic forces occurred because a flake of the layered material had become attached to the tip.[1] Thus, the tip and sample structures were commensurate, and a periodic interaction would be expected. However, further measurements reported stick-slip on materials that did not possess such bonding anisotropy, such as NaCl, gold and diamond. Atomic-level stick-slip can thus occur between the sample and the tip itself.

Stick-slip and contact size

Initially, several researchers misunderstood data like those in Fig. 1, thinking that true atomic resolution was achieved. One aspect that contributed to this misunderstanding is that there is no way to distinguish between the buckling and bending deformation modes of the cantilever (see Figs. 2c and 2d). As a result, atomic-level stick-slip behavior was misinterpreted as being a topographic signal from the corrugation, as seen in STM images. But this was not the case. The lack of true atomic resolution in contact AFM can be understood in light of the contact mechanics. When the tip is in contact with a given sample, for typical tip radii, loads and elastic constants, the contact is larger than a single atom. For example, a 20 nm radius silicon nitride tip exerting a 1 nN load on a mica sample

produces a contact area involving nearly 15 mica unit cells, as estimated using the Hertz theory. Furthermore, Hertz theory neglects tip-sample adhesion, which, if included, makes the estimated contact area even larger and can ensure a substantial contact area even at the lowest possible applied loads. Atomistic models confirm this argument.[2] As a result, contact-mode AFM cannot have single-atom resolution as an STM does. In fact, Mate's original paper presented similar calculations which showed the contact area to be far greater than a single atom contact.[4] This observation has several consequences; for example, the lateral resolution of features is limited by the contact area, and, as a result, point defects are not imaged. It therefore remains to be explained why, despite having a multiple atom contact and (most likely) a non-commensurate tip structure, the interaction between the tip and sample possesses the periodicity of the sample's atomic lattice.

Comparison with conventional stick-slip motion

The term *stick-slip* must be used carefully: Historically, *stick-slip* refers to macroscopic behavior involving multiple contacting asperities.[3] A creaking door hinge, a bowed violin string, screeching tires and earthquakes are all examples of macroscopic stick-slip. Furthermore, stick-slip in micrometer-scale single-asperity contacts has been observed frequently using the surface forces apparatus. A rich variety of phenomena are involved in these examples, but the unifying principle is that the instability results from the dependence of friction upon the interfacial sliding speed or the static contact time in combination with the presence of some elastic compliance in the system. Specifically, if friction during sliding is lower than in the static case, if sliding friction reduces with increasing sliding speed, or if friction grows with time in static contact, then stick-slip instabilities can result.[3]

Consider a force applied through an elastic spring of a given stiffness to a slab of material in static contact with another material. The remote end of the spring is moved at a fixed pulling speed. Initially, because the surfaces are stuck together, the spring stretches and thus the lateral force the spring exerts on the slab increases. Once this force exceeds the static friction force, sliding begins. If friction is lower at higher interfacial sliding speeds, then this leads to increasingly faster relaxation of the spring force until it is no longer large enough to maintain sliding (*i.e.*, it falls below the kinetic friction force for that relative sliding speed). The system then sticks again and the cycle repeats. This behavior is influenced by factors such as the surface roughness and sliding speed-dependent effects particularly evident in viscous or viscoelastic materials. In contrast, in *atomic-level stick-slip*, the interface is atomically smooth, wear does not occur, and the contact may involve only solid, largely elastic materials, although the behavior is also seen in viscoelastic materials. No strengthening of static friction with contact time, or decrease in sliding friction with sliding speed is required.

Consequently, the pulling speed dependence of atomic-level stick-slip friction deviates from that typically seen for macroscopic stick-slip behavior. Macroscopic interfaces typically exhibit decreased static friction with increasing pulling speed resulting from the dependence of friction on interfacial contact time mentioned above. In contrast, a near-logarithmic *increase* of the static friction force with increasing pulling speed is attributed to reaction rate-limited slipping from one lattice site (or potential energy minimum) to the next.[5] The atomic-scale stick-slip process can be viewed as a series of chemical reactions with potential energy barriers that limit motion, where slipping is the "reaction." Tip atoms

in the "stuck" position try to surmount these barriers at a natural attempt frequency. The slower the pulling speed, the more likely it is that they will be able to slip out of their local potential minima at a lower applied lateral force. If the tip is pulled at a high speed, the likelihood that these natural escape attempts will contribute to the slip process is reduced, and the force required to counteract friction will be greater than if the tip atoms were moved along more slowly. Observations of atomic-scale static friction decreasing with pulling speed have also been reported. As mentioned in the theory section below, the precise explanation for the speed and temperature dependence is a matter of ongoing debate.

Commensurability at the interface

Interfacial commensurability–*i.e.*, periodic coincidence of the lattices–is not a necessary condition for the occurrence of stick-slip. For example, atomic-level stick-slip has been observed with AFM tips made out of amorphous silicon nitride and oxide.[1] Even if the tip atoms are ordered, they will not necessarily be in an arrangement that is commensurate with the sample's lattice. Without a commensurate interface, a sufficiently large tip would have no preferred relative positions in which to reside, and therefore smooth sliding could be expected. In light of this argument, the regular appearance of atomic-level stick-slip for a wide range of tip materials and sizes is surprising. Not only does this phenomenon occur for many tip and sample materials, but it has also been observed in humid and dry air, liquid and vacuum, and from cryogenic to elevated temperatures.

While atomic-level stick-slip is *often* observed with crystalline samples, it is not *always* observed. Conditions can vary so that a given tip can alternate between producing stick-slip motion and not. This behavior is the topic of much discussion among experimentalists, but no systematic study of the specific conditions that govern the occurrence of stick-slip friction has been carried out.

Theoretical approaches

Several theoretical efforts to explain and model atomic-level stick-slip behavior, specifically in the context of force microscopy, have appeared in the literature. These studies can be divided into analytical approaches and molecular dynamics (MD) simulations. The analytical approaches primarily address the *mechanics* of stick-slip behavior, *i.e.,* a potential energy distribution (frequently referred to simply as a potential) is assumed and the resulting behavior studied. Most of the analytical approaches build on the Prandtl-Tomlinson model.[6,7] Some of these models represent the tip as a single atom or a single entity without internal degrees of freedom, although multi-atom (tens of atoms) tips have also been considered. Scanning is simulated by increasing the lateral displacement between the fixed end of the cantilever and the sample. The tip initially resides in a potential minimum that is determined by the tip-sample interaction. Because finite static friction due to tip-sample interactions inhibits sliding of the tip, elastic energy is built up in the cantilever and in elastic deformations of the tip and sample themselves.

The total energy of the system is comprised of the interaction energy and the elastic energy stored in the cantilever and the deformed contact (Fig. 4). If the (lateral, or torsional) spring is compliant enough compared with the corrugation of the potential energy, a critical point is eventually reached, where the elastic strain energy becomes sufficient to move the system out of the potential minimum. As a result, slip between tip

and sample takes place. In this slip stage, the cantilever and the contact quickly relax, the previously stored energy is released, and the motion is brought to a stop as the tip finds a new potential minimum, the closest one being one unit cell over. This stick-slip motion generates vibrations both in the sample and the cantilever. The phonons excited in this process carry energy away from the interaction region. Since phonon group velocities are much higher than typical AFM tip scanning speeds (even when slipping), this relaxation occurs before the next stick event. The collective results of the analytical models can be summarized as follows:

1. The stick-slip instability can be interpreted as the system (tip and sample) residing in or searching for potential energy minima, where the energy is the sum of the tip-sample interaction and elastic energy stored in the torsion of the cantilever and the lateral deformation of the contact.

2. Sufficiently small stiffness values of the cantilever springs and the contact itself and a sufficiently strong tip-sample interaction are required to produce the stick-slip instability. If this is not the case, then the stick-slip instability can be prevented, and near-frictionless sliding can occur.[8] This phenomenon is discussed in more detail in the next section.

3. The energy stored and then dissipated will be distributed amongst the cantilever, the tip and the contact, depending on their relative (lateral) stiffness values and damping constants.

4. Friction decreases with increasing temperature due to thermally-activated hopping across potential barriers. Increased scanning (pulling) speeds will lead to increased friction because of the reduced amount of time given to allow thermally-assisted sliding to occur. However, the attempt frequency, and precise temperature and speed dependencies are matters of debate.

5. The entire system, which involves the tip-sample interaction, the lateral contact stiffness and the cantilever's torsional stiffness, is non-linear in nature. The resulting dynamics can be chaotic depending on the pulling speed and the tip-sample interaction.

While these insights are clearly important, they do not provide any information on the details of vibrations (energy dissipation) in the contact zone, the physical origin of the interaction forces, or the possibility of relaxation and displacement of tip atoms in the contact. Some further insight into the origins of stick-slip behavior has been provided by molecular dynamics (MD) simulations. MD simulations have revealed that stick-slip can vary with applied load, scan speed, and scan direction with respect to crystallographic directions. Slip has been shown to occur for some systems via a dislocation mechanism, whereby tip atoms that initially reside in surface *fcc* positions relieve lateral strain by shifting to *hcp* sites.[9] The slipped and unslipped atoms are separated by a dislocation that propagates through the contact.

Interpretations of MD data must be carried out with caution, as MD approaches suffer from several significant limitations. These are primarily the following:

1. In most cases, because of computational limits, the modeled tip is approximately ten times smaller than those used in AFM experiments.

2. Also, because of computational limits, scanning speeds are several orders of magnitude faster than what is achieved in AFM experiments. Typical MD simulation speeds are 10^{-1} m/s to 10^2 m/s *vs.* typical AFM experimental speeds of 10^{-7} m/s to 10^{-5} m/s.

3. Simplifying assumptions are often made regarding the interaction potentials, including, in some cases, the use of a very generic Leonard-Jones potential.

New approaches, such as multi-million atom simulations, or accelerated MD, are beginning to address these issues. As well, despite these limitations, MD provides many insights, including atomic-level descriptions of vibrational motion and energy dissipation mechanisms active during stick-slip motion, revealing that excitations are indeed highly localized in the contact zone.

Stick-slip transitions: Superlubricity, and multiple slip

Recently, Socoliuc *et al.* used AFM to observe the existence of smooth sliding with no stick-slip when the load was sufficiently low, corresponding to extremely low energy dissipation.[10] As the load increased, a transition to stick-slip behavior occurred (Fig. 5). This can be understood in the context of the Prandtl-Tomlinson model. As shown in Fig. 4a, stick-slip instabilities no longer occur when the surface corrugation is sufficiently weak or the cantilever spring is sufficiently stiff. Specifically, the parameter $\gamma_T = \left(2\pi^2 V_0\right) / \left(k_{exp} a_l^2\right)$ describes the relation between the lateral corrugation of the tip–substrate interaction V_0, the substrate lattice parameter a_l, and the experimental lateral stiffness of the system k_{exp}. The experimental lateral stiffness can be evaluated from the expression: $\frac{1}{k_{exp}} = \frac{1}{k_{lever}} + \frac{1}{k_{tip}} + \frac{1}{k_{cont}}$, which includes the effect of the lateral stiffness of the cantilever k_{lever}, tip structure k_{tip}, and tip-sample contact k_{cont}.

Atomic stick-slip behavior is observed only if $\gamma_T > 1$, *i.e.,* when the system is sufficiently compliant or the interfacial corrugation is sufficiently strong. When $\gamma_T < 1$, sliding occurs without stick-slip instabilities. This phenomenon has been termed "superlubricity." The term is somewhat misleading, as there can still be remaining dynamic dissipation. So far however, the friction force observed in these cases has been lower than the detectable limit of the AFMs used, and correspondingly the friction loops have no observable hysteresis within the experimental uncertainty.

Superlubricity has been accomplished in a variety of ways. For example, Dienwiebel *et al.* observed superlubricity for a graphite flake attached to the tip sliding on a graphite surface.[11] Superlubricity was only observed when the flake and the surface were oriented to be incommensurate, in agreement with the idea that incommensurability renders the corrugation of the interfacial potential sufficiently weak to allow the suppression of stick-slip instabilities. Superlubricity has also been observed for sharp AFM tips sliding over atomically flat surfaces at sufficiently low loads.[10,12] In these

experiments, the superlubricity was enabled by the sufficiently low interfacial corrugation resulting from the low applied loads and, as shown by Socoliuc *et al.*,[10] it was correlated with low values of the interfacial lateral stiffness. The demonstrated agreement with the predictions of the Tomlinson model is impressive. Recently, methods have been described that can be readily applied to practical situations: By oscillating the tip or sample at frequencies corresponding to normal resonances of the system, one can intermittently achieve low loads and thus low interfacial corrugations with very little actuation required. Such an oscillating tip is able to slide stably at the instants where the corrugation (total load) is low, resulting in a lower net friction. This same technique has been applied to macroscopic systems.

Another type of transition was predicted by Johnson and Woodhouse who showed that under certain conditions slip may occur over an integer number of lattice spacings.[13] This phenomenon is called a *multiple slip*. In fact, a multiple slip was observed in the original Letter reporting atomic-level stick-slip by Mate *et al.*,[4] but since then it has been rarely discussed until recently.[12,14] Johnson and Woodhouse identified the relationships between the lateral (*i.e.,* torsional) cantilever stiffness, the lateral stiffness of the elastically deformed contact itself, and the corrugation of the lateral force interaction, as key parameters controlling the transition to multiple slips. An adjustable damping factor was introduced which represents the dynamic energy dissipation in the tip or sample materials or in the cantilever itself. The transition from single to double slips occurs when high-frequency fluctuations in the lateral force, triggered by the slip instability, overshoot the corrugated lateral tip-sample interaction force. The possibility of an overshoot reduces with increased damping.

Analytical approaches have been employed to describe transitions from single to multiple slip in atomic-scale friction (Fig. 4). For example, Conley *et al.* used a combination of numerical methods to deal with issues of the complex dynamics in atomic-scale friction.[15] The authors considered a quasistatic limit and transitions between multiple slip modes by solving the equation of motion numerically. An analytical solution of the transition between different slip regimes for the simple case of the one-dimensional Tomlinson model in the quasistatic limit was recently reported by Medyanik *et al.*[12] The authors analyzed the energy landscape and showed how the number of local energy minima increases with increasing interfacial corrugation (Fig. 6a). Single and multiple slips correspond to sliding of the tip to the nearest, the next nearest, or the next-next nearest, *etc.*, local minimum. Slipping to further minima can occur only with sufficiently low energy dissipation during slip. Transition from single to multiple slip occurs with increasing load which indicates that corrugation increases with load. Specifically, the existence of multiple slip regimes is governed by the Tomlinson parameter, γ_T, reaching characteristic values. In other words, $\gamma_T = 1$ represents the transition from smooth sliding to slipping by one lattice site. The possibility of slips of higher multiplicity occurs for larger critical values of γ_T. In the same paper,[12] the authors reported experimental observation of the dependence of stick-slip behavior on load. The experiments were performed on a highly-oriented pyrolitic graphite (HOPG) sample and, as shown in Fig. 6b, the system exhibited superlubricity at the lowest applied load. At higher loads, stick-slip instabilities occurred with the periodicity of the HOPG lattice, while increasing the load even further leads to slips over integer multiples of the lattice spacing, as predicted by the model.

Remaining Questions

There has not yet been any clear conclusion indicating under exactly what conditions stick-slip behavior occurs. Often, images like the one shown in Fig. 1 are *not* obtained. It is possible that under the same loads, with the same sample and with the same cantilever, some unknown change in the tip occurs and stick-slip is suddenly observed. The reasons for this are not established. Furthermore, no one has studied whether friction varies with load in the same manner in the presence and absence of stick-slip.

Another unresolved question pertains to stick-slip periodicity. Most accounts so far report *one* stick-slip event per surface unit cell, even when the unit cell contains more than one atomic species, such as the surface of an alkali halide crystal. One exception is the large unit cell of Si(111)-7×7, as measured in ultrahigh vacuum (UHV) with tips coated with polytetrafluoroethylene, where multiple stick-slip events per unit cell were resolved.[16]

As discussed above, energy released during sliding is carried away by phonons excited in the sliding process. Phonon frequencies are eleven orders of magnitude higher than AFM scanning frequencies, and the relevant dissipation processes occur quickly. It has been found recently in experiments involving Si tips on KBr samples that slip times in atomic stick-slip can be as long as 10 ms. Such time scales are currently inaccessible to conventional atomistic simulations. However, new accelerated simulation techniques, or the use of more powerful computers and efficient algorithms, may provide routes to addressing this challenge.

KEY APPLICATIONS:

While macroscale stick-slip processes are capable of producing significant entities, from music to mountains, atomic-scale stick-slip has yet to be incorporated in any direct application. Stick-slip is used by microelectromechanical systems for precision applications, demonstrating positioning down to 10 nm.[17] It is conceivable that stick-slip methods could be extended to atomic-scale positioning by integrating nanoscale contacts into a device, such that atomic-level stick-slip at the contact point can be used as a precise and accurate indicator of position. Nonetheless, such applications have not yet been realized in a device. At the moment, the most commonly used application for atomic-level stick-slip is the nanoscale lateral spatial calibration of AFMs. This procedure is one of a few and perhaps the fastest of the ways to calibrate an AFM at this scale.
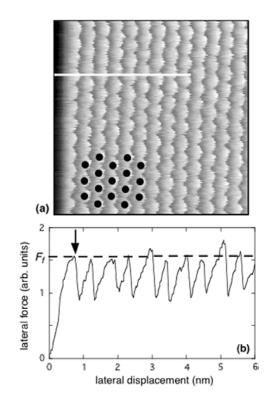
FIGURES:

Figure 1. (a) Lateral force AFM image of the muscovite mica (0001) surface. Image size: 7.5 x 7.5 nm². The fast scan direction is from left to right. The filled circles represent the lattice of the mica unit, whose symmetry and periodicity (0.529 nm) coincide with the lateral forces. (b) Line trace of the section indicated in (a). The lateral force exhibits "stick-slip" behavior, where the lateral force builds up to some well-defined maximum value, and then quickly relaxes (arrow). During the relaxation, the tip slips by one unit cell. This behavior repeats itself with the lattice periodicity. From Ref. [2].
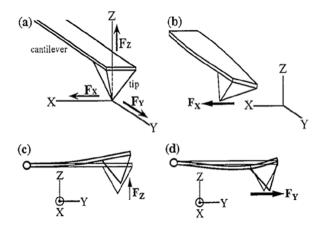
Figure 2. (a) Force components which act on the tip apex of an AFM cantilever. F_x, F_y, and F_z are the forces across, along and normal to the cantilever respectively. These forces cause torsion (b), bending (c), and buckling (d) respectively. From Ref. [1].
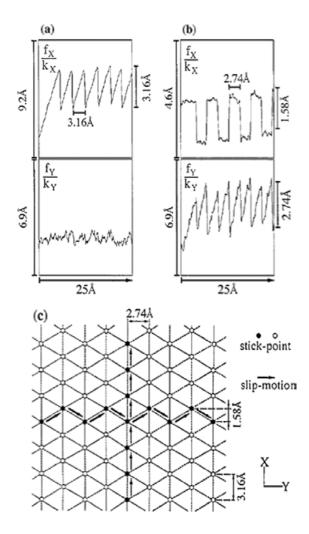
Figure 3. Friction data for lateral cantilever twisting (f_x/k_x) and buckling (f_y/k_y) due to frictional forces parallel to a MoS_2 surface acting on a Si_3N_4 tip, where f_x, f_y and k_x, k_y are the friction force along x and y and the torsional and flexural stiffness of the cantilever, respectively. In (a), the cantilever was scanned along the *x* direction indicated in (c) (*i.e.,* perpendicular to its long axis). Stick-slip behavior resulted in periodic lateral twisting of the cantilever and no appreciable back-and-forth longitudinal buckling of the cantilever. In (b), the cantilever was scanned along the *y* direction indicated in (c) (*i.e.,* parallel to its long axis). This time, the cantilever buckled back and forth as it stuck (top) and twisted back and forth (bottom) while scanning. Individual stick points and the path of the tip were mapped out, as indicated in (c), which corresponded to the MoS_2 unit cell. (From Ref. [1])
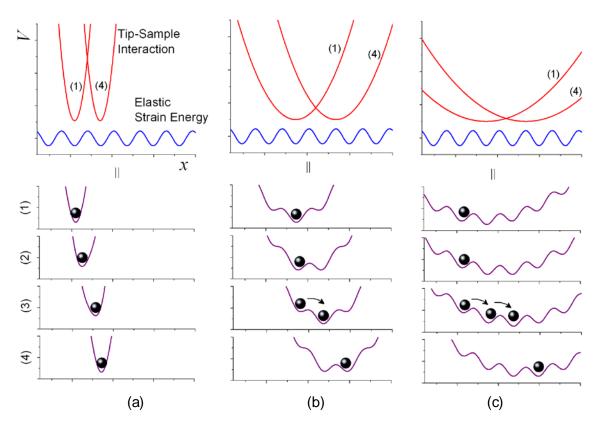
Figure 4: Energy *V vs.* displacement *x* described for the 1-D Prandtl-Tomlinson model for (a) continuous sliding, (b) stick-single slip, and (c) stick-double slip. Upper plots illustrate the separate energetic contributions from the periodic tip-sample interaction potential (blue) and the strain energy in the elastic components of the system (red). The lower plots show the total energy changing as the scan progresses, with a sphere indicating the location of a monatomic tip.
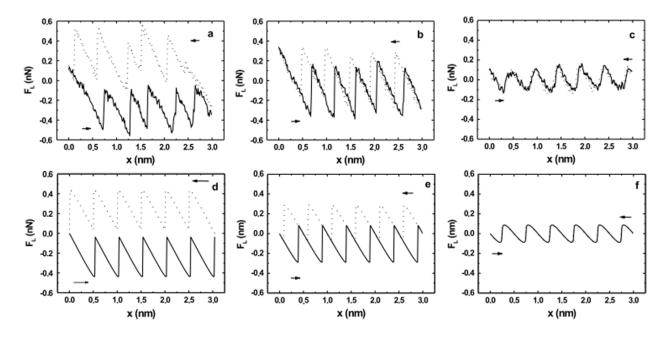
Figure 5. (a) –(c) Experimental friction loops showing the lateral force acting on the tip sliding from right to left (dotted line) and then left to right (solid line) in the (100) direction on the NaCl(001) surface in UHV. The externally applied load was (a) 4.7 nN, (b) 3.3 nN, and (c) 0.47 nN. (d) –(f) Corresponding numerical results from the Tomlinson model for (d) $\gamma_T = 5$, (e) $\gamma_T = 3$, and (f) $\gamma_T = 1$. The lateral stiffness for the calculation is chosen to be $k_{exp} = 1$ N/m and the lattice constant $a_l = 0.5$ nm. When $\gamma_T \leq 1$, smooth sliding is seen and hysteresis between the forward and the backward lateral forces disappears (*i.e.*, there is no dissipation). From Ref. [10].
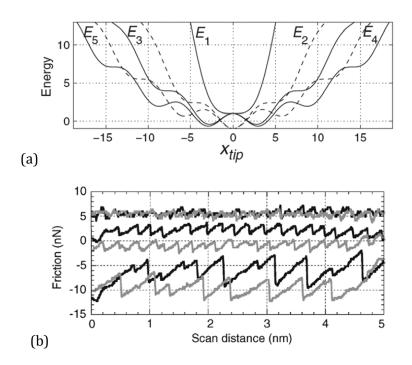
(a)

(b)

Figure 6. (a) Energy landscapes (energy *vs.* position of the tip) corresponding to five different values of lateral contact stiffness. For each energy curve E_i (except $i = 1$) there are i local minima that correspond to stable equilibrium states of the system. Therefore as the system becomes unstable due to the motion of the cantilever, there are i possible destinations in which to slip. (b) Friction behavior on the (0001) surface of HOPG exhibits transition to multiple slips: Smooth sliding is observed at the lowest load (top), single slip is observed at intermediate loads (middle), and mostly double slips occur at the largest load (bottom). From Ref. [12].

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