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Calculation of an Atomically Modulated Friction Force in Atomic-Force Microscopy.

D. TOMÁNEK(*), W. ZHONG(*) and H. THOMAS(**)

(*) *Department of Physics and Astronomy and Center for Fundamental Materials Research, Michigan State University - East Lansing, MI 48824-1116*

(**) *Institut für Physik, Universität Basel Klingelbergstrasse 82, CH-4056 Basel, Switzerland*

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Abstract. – We investigate the microscopic mechanism of energy dissipation in the friction force microscope (FFM), which is a modification of the atomic-force microscope for application to friction. Based on *ab initio* results for the interaction between Pd and graphite, we determine the atomic-scale modulation of the friction force and the corresponding stick-slip motion at the interface during the relative motion between these solids. We propose two idealized versions of the FFM and show that the friction force depends not only on the Pd-graphite interaction potential, but even more critically on the construction parameters of such a microscope.

Friction between two solids is one of the most important and complex processes which affect everyday's life. So far, the science of friction—tribology—has described this phenomenon mainly in a macroscopic and phenomenological way [1]. More recently, successful attempts have been undertaken to observe friction forces on the atomic scale [2] and to understand the underlying microscopic mechanisms in case of sliding friction without wear [3]. Independently, the availability of supercomputers has made predictive calculations [4, 5] of atomic-scale friction forces possible.

The recent experimental progress in nanotribology has been facilitated by the development of a modified version of the atomic-force microscope [6] (AFM) for application to friction, which is sometimes called the friction force microscope (FFM). Like the AFM, the FFM consists of an «atomically sharp» tip of a material A, suspended on a soft cantilever, which is brought into nondestructive contact with a well-defined substrate B. Measuring the vertical deflection of the cantilever is used to keep the applied load F_{ext} constant during the surface scan. An independent measurement of the cantilever deflection in the direction of

the trajectory provides information about the microscopic friction force F_f which inhibits sliding between materials A and B.

It has been recognized already a long time ago that in the ideal case of a single crystal B with a defect-free surface, and in the absence of dislocations and plastic deformations, the only source of friction and its modulation should be atomic-scale corrugations of the A-B interaction potential [7]. These modulations can be calculated [4, 5] and have been successfully observed on highly oriented pyrolytic graphite using the FFM with a tungsten tip [2]. In this system, the A-B interaction is much weaker than the cohesive forces in A or B. If this condition is dropped, wear is likely to occur at the A-B interface, observable as substrate plowing or wetting of the tip by substrate atoms [4]. For dissipative friction without wear, the motion of the tip across the surface must give rise to a sequence of instabilities. The degrees of freedom predominantly involved in these instabilities may be either those of the substrate or those of the tip, and one can distinguish «substrate-induced» from «tip-induced» friction.

As for the theory, two different approaches have been used to determine atomic-scale friction. A molecular dynamics calculation with parametrized pair potentials has been used to simulate the stick-slip motion and to determine the friction force between a Si FFM tip and a Si substrate [4]. An independent approach, based on an *ab initio* density functional calculation, has been used to determine the upper limit of the average friction force $\langle F_f \rangle$ from the potential energy barriers $\Delta V(F_{ext})$ during the «surface diffusion» of Pd on graphite [5]. In this letter, we address the mechanism which leads to the excitation of microscopic degrees of freedom and hence to energy dissipation. We make use of the first-principles Pd-graphite interaction potential [5] and based on these results, we determine atomic-scale modulations of the friction force F_f along the horizontal trajectory of the Pd tip on graphite in the quasi-static limit of relative velocity $v \to 0$. We show that F_f depends sensitively not only on the corrugation and shape of the Pd-graphite interaction potential, but also on the specific construction parameters of the FFM. This latter point is of utmost importance if friction forces obtained with different microscopes are to be compared to one another.

Two models of a friction force microscope are shown in fig. 1. In both models, the

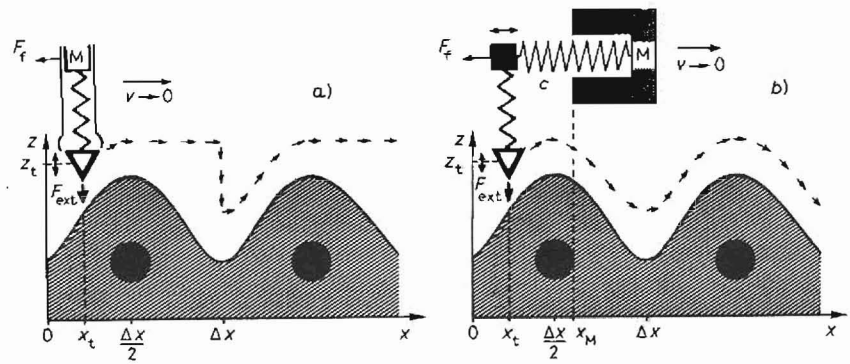

Fig. 1. – Two models for the friction force microscope (FFM). In both models, the external suspension M is guided along the horizontal surface x-direction at a constant velocity $v = \mathrm{d}x_M/\mathrm{d}t \to 0$. The load F_{ext} on the «sharp» tip (indicated by V) is kept constant along the trajectory $z_t(x_t)$ (shown by arrows). *a*) A «maximum-friction microscope», where the tip is free to move up, but gets stuck at the maximum z_t between $\Delta x/2$ and Δx. *b*) A «realistic-friction microscope», where the position of the tip x_t and the suspension x_M may differ. In this case, the friction force F_f is related to the elongation $x_t - x_M$ of the horizontal spring from its equilibrium value.

suspension of the tip moves quasi-statistically along the surface x-direction with its position x_M as the externally controlled parameter. The tip is assumed to be stiff with respect to excursions in the surface y-direction. We restrict our discussion to the case of tip-induced friction and assume a rigid substrate which applies for friction measurements on graphite [2, 5]. In the «maximum-friction microscope» [5], the full amount of energy needed to cross the potential energy barrier ΔV along Δx is dissipated into heat. This process and the corresponding friction force can be observed in an imperfect atomic-force microscope which is shown in fig. 1*a*). A vertical spring connects the tip and the external microscope suspension M. The horizontal positions of the tip and the suspension are *rigidly* coupled, $x_t = x_M = x$. For $0 < x < \Delta x/2$, a constant load F_{ext} on the tip is adjusted by moving the suspension up or down. For $\Delta x/2 < x < \Delta x$, however, the tip gets stuck at the maximum value of z_t. At Δx, the energy ΔV stored in the spring is abruptly and completely released into internal degrees of freedom which appear as heat.

The calculated potential energy $V(x)$ during this process is shown in fig. 2*a*). We considered a monatomic Pd tip sliding along a trajectory connecting adjacent hollow and bridge sites on graphite, for a load $F_{ext} = 10^{-8}$ N. In order to predict F_f, we have used the results of our previous *ab initio* calculation for Pd on graphite [5] which have been conveniently parametrized [8]. The force on the tip in the negative x-direction, as defined in fig. 1*a*), is given by

$$F_f(x_M) = \begin{cases} \partial V(x_M)/\partial x_M\,, & \text{if } 0 < x_M < \Delta x/2\,, \\ 0\,, & \text{if } \Delta x/2 < x_M < \Delta x \end{cases} \tag{1}$$

and shown in fig. 2*b*). The nonzero value of the average friction force $\langle F_f \rangle$, indicated by the dash-dotted line in fig. 2*b*), is a clear consequence of the mechanism which allows the tip to get stuck. $F_f(x_M)$ is a nonconservative force since it depends strongly on the scan direction. In the absence of the «sticking» mechanism, F_f is given by the gradient of the potential *everywhere*, as indicated by the dashed line in fig. 2*b*). It is independent of the scan direction and hence conservative. In this case, F_f inhibits sliding for $0 < x_M < \Delta x/2$ and promotes sliding for $\Delta x/2 < x_M < \Delta x$, so that the friction force averages to zero.

A more realistic construction of the friction force microscope is shown in fig. 1*b*). In this microscope, the AFM-like tip-spring assembly is *elastically* coupled to the suspension in the

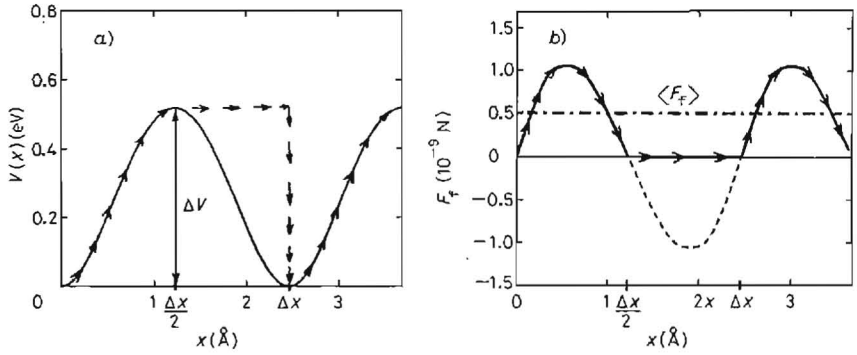

Fig. 2. – Potential energy of the tip $V(x)$ (*a*)), the friction force $F_f(x)$ and the average friction force $\langle F_f \rangle$ (*b*)) in the «maximum friction» microscope with a monatomic Pd tip on graphite. The arrows indicate the tip trajectory corresponding to a relaxed vertical position z_t for a constant load on the tip $F_{ext} = 10^{-8}$ N.

horizontal direction, so that x_t may differ from x_M. For a given x_t, the tip experiences a potential $V(x_t, z_t) = V_{int}(x_t, z_t) + F_{ext} z_t$ consisting of the tip-surface interaction V_{int} and the work against F_{ext}. The tip trajectory $z_{t,min}(x_t)$ during the surface scan is given by the minimum of $V(x_t, z_t)$ with respect to z_t. For this trajectory, $V(x_t) = V(x_t, z_{t,min})$ represents an effective tip-substrate potential.

This potential $V(x_t)$ depends strongly on F_{ext} and is corrugated with the periodicity of the substrate due to variations of the chemical bond strength and of $z_{t,min}$. It is reproduced in fig. 3*a*) in the case of a monatomic Pd tip on graphite and $F_{ext} = 10^{-8}$ N. The corrugation of the potential $V(x_t)$ will elongate or compress the horizontal spring from its equilibrium length which corresponds to $x_t = x_M$. The friction force is given by

$$F_f(x_M) = -c(x_t - x_M), \tag{2}$$

where c is the horizontal spring constant. The total potential energy V_{tot} of the system consists of $V(x_t)$ and the energy stored in the horizontal spring,

$$V_{tot}(x_t, x_M) = V(x_t) + \frac{1}{2} c(x_t - x_M)^2. \tag{3}$$

For a given horizontal position x_M of the FFM suspension, the equilibrium position of the tip x_t is obtained by minimizing V_{tot} with respect to x_t. We obtain

$$\frac{\partial V_{tot}}{\partial x_t} = \frac{\partial V(x_t)}{\partial x_t} + c(x_t - x_M) = 0 \tag{4}$$

or, with eq. (2),

$$F_f = -cx_t + cx_M = \frac{\partial V(x_t)}{\partial x_t}. \tag{5}$$

A graphical solution of eq. (5) is shown in fig. 3*b*) and the resulting relation $x_t(x_M)$ is shown in fig. 3*c*). If the force constant c exceeds the critical value $c_{crit} = -[\partial^2 V(x_t)/\partial x_t^2]_{min}$, which for Pd on graphite and $F_{ext} = 10^{-8}$ N is $c_{crit} = 23.2$ N/m, we obtain a single solution x_t for all x_M. This situation is indicated by the dotted line in fig. 3*b*) and *c*) for a hard spring with $c = 40.0$ N/m. The friction force F_f is given by eq. (2) and shown by the dotted line in fig. 3*d*). F_f is independent of the scan direction and hence conservative, resulting in $\langle F_f \rangle = 0$. Hence no friction should occur in the AFM, which is the limiting case of an FFM for $c \to \infty$.

A more interesting case arises if the horizontal spring is soft, $c < c_{crit}$. This situation is shown by the solid line in fig. 3*b*) and *c*) for $c = 10.0$ N/m. In this case, the solution $x_t(x_M)$ of eq. (5) displays a sequence of instabilities. These instabilities lead to a stick-slip motion of the tip with increasing x_M, similar to «plucking a string». The hysteresis in the $x_t(x_M)$ relation, shown in fig. 3*c*), results in a dependence of the force F_f on the scan direction. The friction force $F_f(x_M)$ in this case is shown by the solid line in fig. 3*d*). It is a nonconservative/dissipative force and averages to a nonzero value $\langle F_f \rangle = 3.03 \cdot 10^{-10}$ N, given by the dash-dotted line. The energy released from the elongated spring into heat is represented by the shaded area in fig. 3*d*).

The present theory predicts occurrence of friction only for very soft springs or a strongly corrugated potential $V(x_t)$. The latter fact can be verified experimentally since the corrugations $\Delta V(x_t)$ increase strongly with increasing applied load [5]. Consequently, for a given c, the friction force is zero unless a minimum load F_{ext} is exceeded. On the other hand, for a given F_{ext}, no friction can occur if c exceeds a critical value $c_{crit}(F_{ext})$.

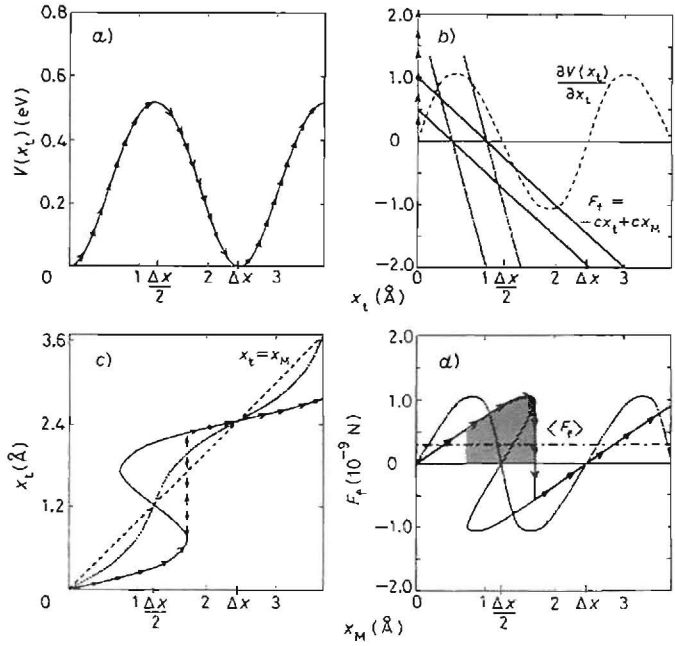

Fig. 3. – Microscopic friction mechanism in the «realistic-friction microscope». The calculations are for a monatomic Pd tip on graphite and $F_{ext} = 10^{-8}$ N. Results for a soft spring, giving nonzero friction, are compared to a zero-friction microscope with a hard spring. *a*) Potential energy of the tip $V(x_t)$. *b*) A graphical solution of eq. (5) yielding the equilibrium tip position at the intersection of the derivative of the potential $\partial V(x_t)/\partial x_t$ (dashed line) and the force due to the horizontal spring F_f. Solid lines, for different values of x_M, correspond to a soft spring with $c = 10.0$ N/m, and dotted lines correspond to a hard spring with $c = 40.0$ N/m. *c*) The calculated equilibrium tip position $x_t(x_M)$. *d*) The friction force F_f as a function of the FFM position x_M and the average friction force $\langle F_f \rangle$ for the soft spring (dash-dotted line).

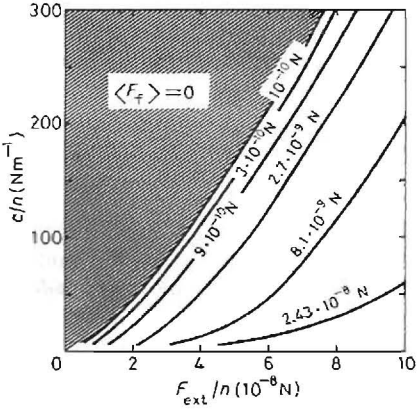

Fig. 4. – Contour plot of the average friction force $\langle F_f \rangle$ between a Pd tip and graphite, as a function of the load F_{ext} and the force constant c. All forces and the force constant are normalized by the number of tip atoms n in contact with the substrate.

A similar situation occurs during sliding between large commensurate flat surfaces of A on B. In that case, c is given by the elastic constants of A at the interface [7], hence cannot be changed independently. Since c is rather large in many materials, zero friction should be observed for moderate applied loads in the absence of wear and plastic deformations. For a multiatom «tip» which is *commensurate* with the substrate, the tip-substrate potential is proportional to the number of tip atoms at the interface, n, and so is the critical value c_{crit} for nonzero friction. In this case, the effective FFM spring depends both on the external spring and the elastic response of the tip material. The inverse value of c_{crit} is given by the sum of the inverse values of the corresponding spring constants. For a large tip which is *incommensurate* with the substrate, no friction should occur [3].

The average friction force $\langle F_{\text{f}} \rangle$ as a function of the load F_{ext} and the force constant c is shown as a contour plot in fig. 4 for a monatomic or a larger commensurate Pd tip on graphite. Clearly, the applicable load range is limited by the underlying assumption of contact without wear. This figure illustrates that not only the friction force F_{f}, but also the friction coefficient $\mu = \langle F_{\text{f}} \rangle / F_{\text{ext}}$, depend strongly both on the interaction potential between the two materials in contact and on the intrinsic force constant c of the friction force microscope. This clearly makes the friction force dependent on the construction parameters of the FFM. There is also one advantage in this fact—c can be chosen in such a way that nonzero friction occurs even at small loads F_{ext}.

In summary, we calculated the atomic-scale modulation of the friction force and the corresponding stick-slip motion at the interface during the relative motion between Pd and graphite. We proposed two idealized versions of a friction force microscope. We showed that the friction force depends not only on the Pd-graphite interaction potential, but even more critically on the construction parameters of such a microscope.

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