Ultrasound induced lubricity in microscopic contact

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A physical effect of ultrasound induced lubricity is reported. We studied the dynamic friction dependence on out-of-plane ultrasonic vibration of a sample using friction force microscopy and a scanning probe technique, the ultrasonic force microscope, which can probe the dynamics of the tip–sample elastic contact at a submicrosecond scale. The results show that friction vanishes when the tip–surface contact breaks for part of the out-of-plane vibration cycle. Moreover, the friction force reduces well before such a break, and this reduction does not depend on the normal load. This suggests the presence on the surface of a layer with viscoelastic behavior. © *1997 American Institute of Physics.* [S0003-6951(97)04335-0]

The friction force microscope (FFM),[1] has opened a way for the study of nanoscale friction phenomena. In the past years, a variety of tribological phenomena have been investigated.[1–5] Recently, experiments have been carried out in ambient, controlled humidity and ultrahigh vacuum environments to understand the conditions in which a single or a multiasperity contact prevails under these conditions.[6,7] Nevertheless, a full understanding of friction and tribology on the nanoscale is still a long way away. It is difficult to investigate the nature of the tip–surface interaction and differentiate the contribution of various forces since FFM only measures the overall lateral force acting on the tip.

A few years ago, a new scanning probe technique, the ultrasonic force microscope (UFM),[8] was introduced, enabling the modulation of the tip–surface distance on the *ultrasonic* submicrosecond time scale. Using UFM, a direct study of the dynamics of the tip–surface contact can be achieved. As a result of the first observations, it was reported that friction is modified by out-of-plane ultrasonic vibration of the sample.[9] A reduction in friction was observed for a range of materials,[10] but only briefly examined. We propose to investigate this phenomenon by combining FFM and UFM techniques.

The microscope used for the experiments is a commercial multimode atomic force microscope (AFM),[11] modified in order to implement the UFM mode.[8,9] The experiments were carried out by laterally moving the sample back and forth relative to the tip, at a set load, as shown schematically in Fig. 1(a). The lateral length was sufficiently long (20–200 nm) to realize dynamic friction for most of the cycle. The friction force was measured by a lock-in amplifier.[12,13]

In the UFM configuration, the sample glued to a piezoplate is vibrated out of plane at ultrasonic frequency (2–3 MHz in our experiments). The free resonance frequency of the cantilever is more than two orders of magnitude lower than the excitation frequency, and so, although there is some cantilever vibration at that frequency, its amplitude is small compared with the excitation amplitude.[14] Any subharmonic response would occur only at higher excitation amplitudes.[15] The tip–sample distance is, thus, modulated [see Fig. 1(b)]. As shown in Fig. 2, if the ultrasonic amplitude is low (a_0) so that the tip–surface distance is modulated over the linear part of the force F versus distance z curve, $F(z)$, the average normal force over one cycle remains unchanged. If the amplitude is high enough (a_{c1} for a set load F_1) to reach the pull off, the average normal force changes causing an additional cantilever deflection. This change in force strongly depends on the set load and the slope of the $F(z)$ curve. For amplitudes higher than a_{c1}, the tip–surface contact is broken for part of the vibration cycle.[8,16]

In our experiments, we recorded the effect of the ultrasonic amplitude on (i) the friction force and (ii) cantilever deflection. The time interval at which the ultrasonic amplitude increases was chosen longer than the sliding period in (i) and shorter than the AFM z-feedback response in (ii), respectively. Comparing the two measurements, one can relate friction to the modulated tip–surface contact area.

We chose three different flat (on the nanoscale) samples to avoid topographical artefacts: polished (100) silicon wafer, cleaved mica, and optically polished glass. Two different types of cantilevers with the tip have been used: a silicon ultralever (nominal tip radius $R=10$ nm, cantilever elastic constant $k_c=0.24$ N/m) and a silicon–nitride microlever ($R=100$ nm, $k_c=0.05$ N/m).[11] All the experiments have been carried out in an ambient environment ($\cong 23\,^{\circ}$C, 30% of relative humidity).

Measurements without ultrasound show that the friction force versus load curve is almost linear for all the cantilever–sample combinations used. This suggests that a multiasperity contact is taking place.[6,17]

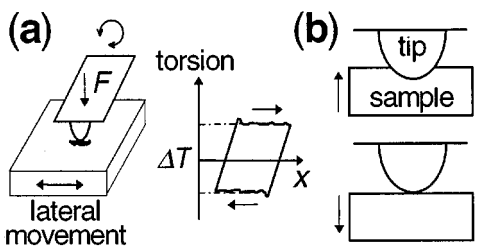

FIG. 1. (a) The tip and the surface slide relative to each other at normal load F. The friction force is proportional to ΔT. (b) The cantilever cannot follow the ultrasonic vibration due to inertia, and the tip–sample distance is modulated.

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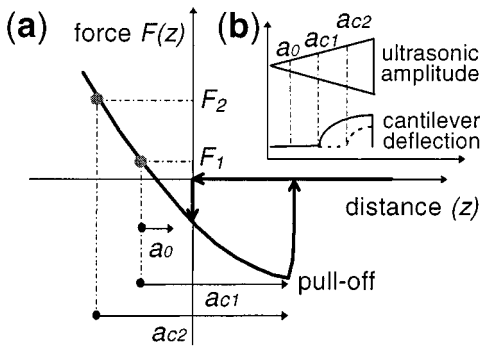

FIG. 2. (a) Scheme of force vs distance dependence $F(z)$ for a tip–sample system. For a set load F_1 and a low value of ultrasonic amplitude (a_0), the average force acting on the tip is unchanged and no variation in cantilever deflection occurs (b). For a higher value of the ultrasonic amplitude (a_{c1}) an additional force rises due to the strong nonlinearity of $F(z)$, and the cantilever deflects [(b) solid line]. For a higher set load F_2, such deflection starts at a higher ultrasonic amplitude a_{c2} [(b) dotted line] (see Ref. 8).

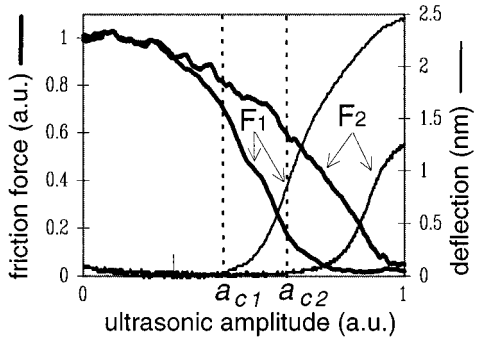

FIG. 3. Experimental measurements of dynamic friction (thick line) and cantilever deflection (thin line) dependencies on the ultrasonic amplitude (Si sample, Si_3N_4 microlever). The loads applied are 0 nN (F_1) and 2 nN (F_2). The sliding speed is 50 nm/s. The maximum friction values are normalized. The ultrasonic amplitude has not been calibrated, but in this case, is estimated to be at the maximum between 10 and 20 Å (see Ref. 8). Friction diminishes even at a low ultrasonic amplitude of about 1–2 Å when the contact is not yet broken and the friction reduction onset does not vary appreciably with the applied load. Friction vanishes when the contact is intermittently broken for part of the vibration cycle. The pull-off force varies by less than 5% over the full range of ultrasonic amplitudes.

In Fig. 3, the representative dependencies of dynamic friction and ultrasonic force on the cantilever deflection are presented (Si sample, Si_3N_4 microlever). The graph shows measurements at two different values of the normal load. As the ultrasonic amplitude is increased from zero, the friction force decreases at first slowly. This decrease always commences at very low ultrasonic amplitudes irrespective of the normal load. When the amplitude reaches a critical value a_{ci} ($i=1$ or 2), a cantilever deflection shift onsets due to the strong nonlinearity of the force curve. At amplitudes above a_{ci}, the friction force rapidly goes to zero. It is to be noted that a_{ci} strongly depends on the normal load, as expected (see Fig. 2).[8] The results obtained for other cantilever–sample combinations exhibit the same behavior in the range of lateral movement speed from 50 to 500 nm/s and of normal loads from -10 to 20 nN. Negative values of the total load can be obtained since an adhesive force acts between the tip and the surface. In our experiments, the pull-off force was estimated between 5 and 20 nN, depending on the tip.

At a macroscopic level, a reduction of friction due to the vibration of one of the two sliding bodies was already reported in the literature, but the vibration direction (lateral rather than out of plane) and the vibration frequency (kHz rather than MHz) were very different from the present case.[18]

In another experiment (Si sample, Si_3N_4 microlever), we studied the dependence of static friction on sample out-of-plane ultrasonic vibration. A static friction force was generated by applying a small lateral vibration amplitude to the sample so that the tip did not slide, and stuck to the surface while continuing to experience an oscillating tangential force. For three values of the lateral vibration length (in the ratio 1:0.6:0.4), the maximum applied shear force under this sticking regime was found to be linearly proportional to the lateral vibration length. The static friction force was measured as the dynamic friction.[13] In Fig. 4, it is shown that the static friction force begins to decrease at very low ultrasonic amplitudes irrespective of the maximum applied shear force. At high ultrasonic amplitudes above the onset of intermittent contact, the static friction vanishes.

Let us consider a tip in contact with a sample under a static load. The sample is now vibrated out of plane at small amplitudes such that the tip is always in contact with the sample; in one cycle, the tip is pushed or driven in by an amount equal to the vibration amplitude. As we showed in Fig. 2, for small amplitudes, the average normal load is equal to the initial set value. Amontons law states that the friction force raises linearly with the normal force.[19] This is clearly contrary to the present observation. Modulating the distance also modulates the contact area. It may be argued that a slip could take place at the minimum contact area, and then the friction force reduces with increasing ultrasonic amplitude. Our static friction measurements, however, show that at ultrasonic amplitudes less than a critical value, it is not possible to have slip (see Fig. 4), and that this critical value is independent of the maximum shear force. A mechanism based on slip at the minimum contact area in each cycle may,

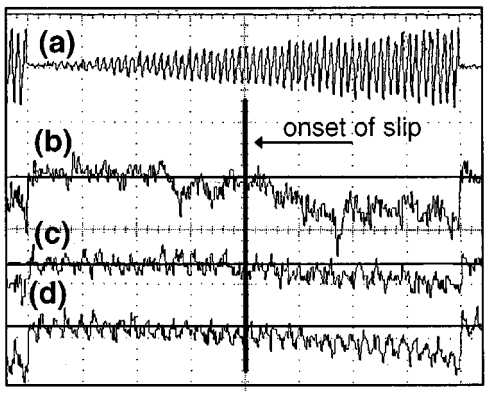

FIG. 4. A small lateral vibration is applied to the sample so that the tip does not slip and a static friction force is generated. The lateral movement length is varied in the ratio 1:0.6:0.4 [from (b) to (d), respectively]. An out-of-plane ultrasonic vibration ramped in amplitude (a) is introduced in all three cases. The ultrasonic amplitudes applied in this experiment correspond to the *low amplitude* regime that is well before the break of the contact. The three torsion signals start decreasing roughly at the same ultrasonic amplitude.

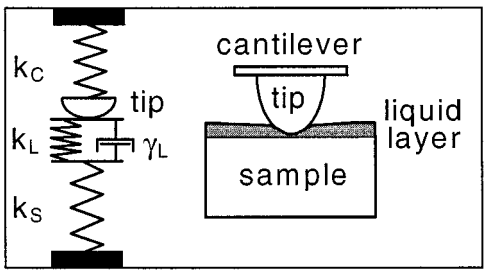

FIG. 5. Proposed model for the dynamics of a solid-liquid-solid contact.

therefore, be discounted as an explanation for the phenomenon shown in Fig. 3.

Another mechanism, which could explain this friction reduction at low amplitudes, is the modification of the pull-off force and, therefore, of the net normal load with increasing amplitude. This is not the case, as our measurements showed a pull-off force reduction of less than 5% at amplitudes at which the friction force had already vanished.

As an explanation of the effect of dynamic friction reduction at very low ultrasonic amplitudes, we could suggest another mechanism. It is known that for a tip–surface contact in an ambient environment with load values as those used in our experiments, the contact is not solid–solid but solid–liquid–solid,[20–22] that a liquid layer formed by water and, possibly, organic contaminants organizes in a solidlike configuration between the tip and the sample sustaining the load.[4,23–25] Such a layer might present a viscoelastic behavior,[26] represented in the rheological model shown in Fig. 5 by a dashpot γ_L and a spring k_L. This viscoelastic effect could be expected to be pronounced at ultrasonic frequencies (from a MHz to GHz frequency range). Without ultrasonic vibration, it is this compliant layer, which is most deformed ($\cong 10$ Å as could be deduced from UFM measurements), whereas the deformation of the sample is an order of magnitude less ($\cong 1$ Å for the loads and tips that we used).[27] Viscosity does not allow the soft layer to relax while the stress of the sample is relieved for part of the vibration cycle. Therefore, a negative pressure can be reached when the sample quickly moves away from the tip, losing the solidlike structure of a compressed liquid layer and causing a noticeable reduction in dynamic friction even at small ultrasonic amplitudes. It is to be noticed that there should be no dependence on the load in this model, as the initial compression of the soft layer does not affect the onset of friction reduction. The same is valid for the static friction case. Therefore, such viscoelastic behavior of a liquid layer consistent with all our data could well be the origin of the observed friction reduction at low ultrasonic amplitudes.

At high amplitudes of the ultrasonic vibration the contact is broken for part of the cycle. Here, the average friction force may be expected to fall sharply with increasing amplitude, as an increasing part of the cycle registers noncontact. The onset amplitude of this regime is marked by a_{ci} in Fig. 3.

We have demonstrated that the friction force dramatically reduces when out-of-plane ultrasound is applied to the sample and completely disappears when the tip–surface contact is broken for part of the ultrasonic cycle. This reduction begins at low amplitudes well before the break of the contact, and such onset does not depend on the applied load.

To explain this effect, we suggest that a liquid layer exhibiting viscoelastic behavior is present in between the tip and the sample. To further investigate this phenomenon, we propose to work in a controlled humidity environment and to vary the tip and sample surface hydrophilic and chemical properties.

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