Atomic-Scale Friction of a Tungsten Tip on a Graphite Surface

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Using an atomic force microscope, we have observed atomic-scale features on the frictional force acting on a tungsten wire tip sliding on the basal plane of a graphite surface at low loads, $< 10^{-4}$ N. The atomic features have the periodicity of the graphite surface and are discussed in terms of a phenomenological model for the tip motion described by the sum of a periodic tip-surface force and the spring force exerted by the wire.

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While the macroscopic friction between two solids rubbing together can be readily measured, little is known about the atomic mechanism by which the frictional forces are generated. Numerous studies,[1] however, have shown that friction can depend dramatically on the chemical and atomic nature of surfaces and can be extremely sensitive even to submonolayers of adsorbed atoms or molecules. Nevertheless, an atomic picture of the microscopic dynamics of friction has yet to be developed.

In this Letter, we report the first observation where the atomic structure of a surface manifests itself directly in the dynamical frictional properties of an interface. In particular, we have observed that the frictional force on a tungsten tip sliding on the basal plane of graphite for small loads ($< 10^{-4}$ N) displays features with the atomic periodicity of the surface. When the magnitude of the frictional forces exceeds a critical value depending on the spring constant of the tip, sudden slips of the tip across the surface are observed which also have the periodicity of the graphite surface. We discuss these results in terms of a phenomenological model for the tip motion described by the sum of a periodic tip-surface force and the force exerted by the level spring of the tip assembly.

Our experiments were conducted with an atomic force microscope[2] operating in ambient laboratory air. Our instrument[3] uses optical interference for measuring the deflection of a level with a known spring constant and a tip at its end. Our typical level and tip combination was fabricated from tungsten wire, 12 mm long and 0.25 or 0.50 mm in diameter, one end of which is bent at a right angle and electrochemically etched in NaOH solution to a sharp point (1500 to 3000 Å in radius) to serve as a tip. The wire spring constants were 150 and 2500 N/m for the 0.25- and 0.50-mm diameter wires, respectively. For these experiments, our sample-tip geometry was arranged for the interferometer to measure the forces on the tip parallel to the surface, as shown in Fig. 1. Experiments were conducted on the basal plane of a single grain of highly oriented polycrystalline graphite. The samples were cleaved before experiments, but contaminants such as water from the air ambient may exist at the tip-surface interface.

For three different loads on the tip, Fig. 2 shows the wire deflection parallel to the surface and the corresponding frictional force experienced by the tip as a function of sample position, as the sample is moved back and forth parallel to the surface plane at a velocity of 400 Å/s. Over the range 40 Å/s to 4000 Å/s, the frictional forces show little dependence on the velocity. For the lowest load, 7.5×10^{-6} N in Fig. 2(a), only a slight frictional force opposes the tip sliding across the surface. At a larger load, 2.4×10^{-5} N in Fig. 2(b), the tip initially moves with the sample until, at point A, the wire exerts enough force on the tip to overcome the static frictional force, and the tip starts to slide across the surface. However, the sliding process is not uniform, but instead the frictional force displays a corrugation with an approximate periodicity of 2.5 Å, the same periodicity as the honeycomb structures of the graphite surface. When the sample direction is reversed at point B, the tip again moves with the sample until it starts to slide at point C. At even larger loads, 5.6×10^{-5} N in Fig. 2(c), the corrugation in the frictional force has a distinctively different shape, as the tip undergoes a series of sudden slips as it slides across the surface. Within the 200-μs time constant of our electronics, the slips are instantaneous, while between slips the tip moves with the surface.

The slips in Fig. 2(c) are irregular since the direction of the scan is not along a symmetry direction of the

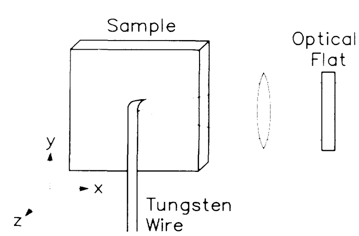

FIG. 1. Schematic diagram of apparatus. The base of the tungsten wire is held fixed, while the sample is moved in x, y, and z directions. Wire deflections parallel to the surface are measured from the intensity change in the interference pattern between light reflected off the wire and light reflected off the optical flat.

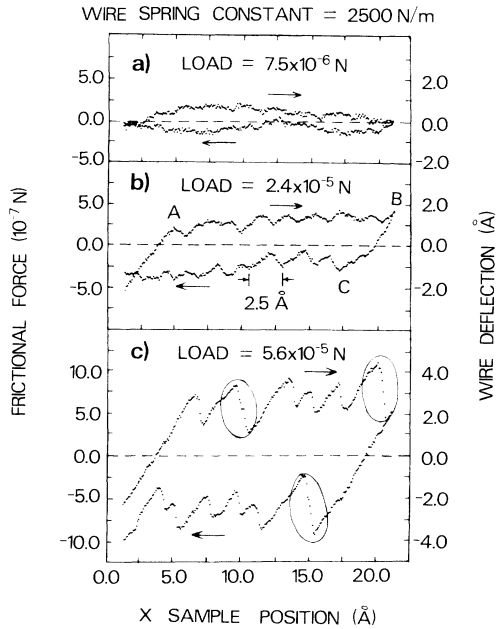

FIG. 2. The wire deflection parallel to the surface and the corresponding frictional force on the tip as a function of sample position for three different loads. The circled sections in (c) indicate where double slips occur.

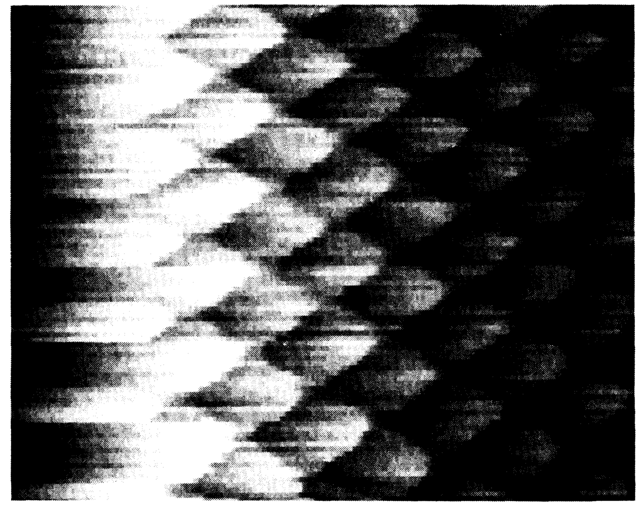

FIG. 3. The frictional force in the x direction as a function of x and y. The intensity of the image is scaled to the frictional force with the bright areas corresponding to a high force. The full-scale change from dark to bright corresponds to 1.8×10^{-6} N. Only scans in left-to-right direction are shown. The size of the image is 20 by 20 Å,[2] with no correction for distortion from the piezoelectric scanners. The load on the tip is 5.6×10^{-5} N, and the wire spring constant is 2500 N/m.

graphite surface. That the slips actually have the periodicity of the surface can be seen in Fig. 3. Here, we show how the frictional force in the x direction behaves during the stick-slip process as the sample is rastered in the xy plane with a 5.6×10^{-5}-N load on the tip and a wire spring constant of 2500 N/m. The sample is moved back and forth with a velocity of 400 Å/s in the x direction and 2 Å/s in the y direction. The region at the left of the image is where the tip moves initially with the sample. The critical frictional force before the first slip occurs varies slightly depending on the position of the tip with respect to the graphite lattice and has an average value of 1.5×10^{-6} N in the image shown. The slips occur where the image changes suddenly from bright to dark. When two contours of slip cross, a "double slip" occurs, which are also observed in Fig. 2(c) and indicated by circles. Figure 3 clearly illustrates how the atomic structure of the surface influences the frictional properties of this interface.

In Fig. 4, we show how the average frictional force the tip experiences during the slip process varies with load, using a wire spring constant of 155 N/m. The onset of atomic-scale stick slip occurs at $\simeq2\times10^{-6}$ N load for the 155-N/m wire, substantially lower than the $\simeq5\times10^{-5}$-N onset observed for the 2500-N/m wire. The data fall near the straight line in the figure, indicating how the frictional force would behave if it were linear with load with a coefficient of friction of 0.012. The apparent coefficients of friction varied somewhat

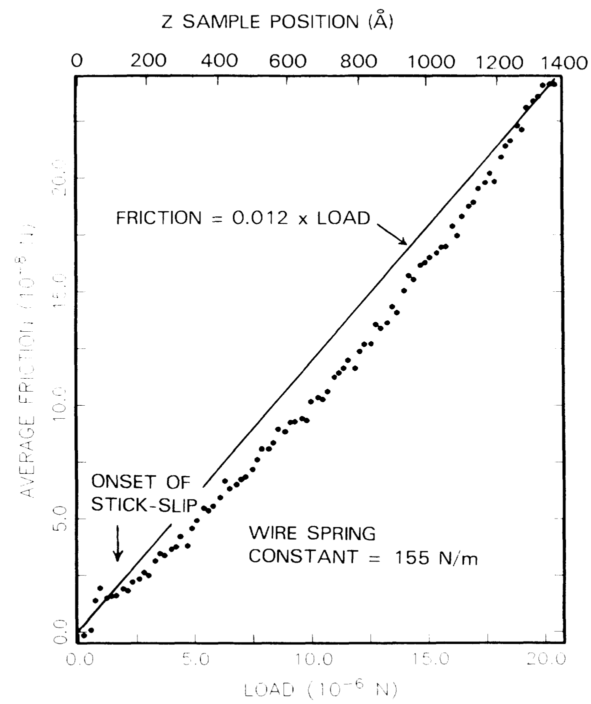

FIG. 4. Average frictional force as a function of load on the tip as it slides across the surface. The load is computed by multiplication of the wire spring constant by the z sample position, shown by the scale at the top, after the sample has made contact with the tip.

from tip to tip and ranged from 0.005 to 0.015 in our experiments. Similar values have been reported by Skinner, Gane, and Tabor[4] for tungsten tips sliding on graphite in vacuum for a slightly higher range of loads (10^{-5} to 4×10^{-4} N).

The frictional forces observed in Figs. 2 and 3 during sliding can be interpreted qualitatively in terms of the tip experiencing a frictional force with two components, one conservative and periodic, and the other nonconservative, constant, and opposing the direction of motion. If the relative motion of the surface and tip were simply that of two rigid collections of atoms, the effective force would be a conservative force oscillating about zero. Slow reversible elastic deformation of the sample or tip would also contribute to the conservative force. The origin of the nonconservative, direction-dependent force component could be phonon generation, viscous dissipation, or plastic deformation or wear. Because the atomic-scale features of the frictional force remain unchanged after many repetitions of the same line scan and are reversible with load, it seems unlikely that significant plastic deformation or wear is occurring. For the case shown in Fig. 2(b), the periodic frictional force would have a peak-to-peak modulation of $\simeq 2\times10^{-7}$ N and the nonperiodic frictional force a value of $\simeq 3\times10^{-7}$ N. Our results also indicate that both the periodic and nonperiodic forces increase with load. However, if the modulation in the frictional force increases to the point where the spatial derivative of the force exceeds the wire spring constant, the tip position becomes unstable, and the tip will "slip" across the surface until the next stable position is found. Figure 5 illustrates how this occurs. So, when the wire spring constant is lowered from 2500 to 155 N/m, the load at which the onset of stick-slip motion occurs is also reduced, as mentioned earlier. Since the slips should occur where the derivative of the force equals the wire spring constant, the slips in Fig. 3 should mark the contour of a constant force derivative along x of 2500 N/m.

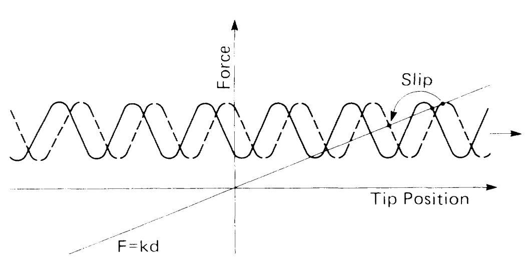

FIG. 5. The frictional force for two sample positions (solid and dashed lines) as the sample moves left to right, and the negative of the force from the wire deflection ($-F=kd$, where k=spring constant and d=wire deflection). The tip position on the surface is stable when the forces on the tip balance, as indicated by the intersection of the plots. As the surface moves underneath the tip, a slip occurs when, at the intersection point, the derivative of the frictional forces equals the spring constant.

For the loads used in this study, the apparent area of contact is large on the atomic scale. Using the model of Mamin *et al.*,[5] which takes into account the deformation of the softer graphite surface, and assuming a tip radius of 3000 Å, we estimate an apparent area of contact of 1.6×10^{6} Å^2 for the image in Fig. 3. However, the surface of the tip is not smooth on an atomic scale, and only the apexes of asperities on the tip will make contact with the smoother graphite surface, resulting in the actual area of contact being much smaller than the apparent area.

In standard models of friction,[6] the frictional force is assumed to be proportional to the actual area of contact, i.e., $F=sA$, where the proportionality constant is the shear strength of the junction. For frictional contacts where only the apexes of asperities touch, substantial theoretical work[7] indicates that the actual area of contact is proportional to load, independent of whether the asperities deform elastically or plastically, if a statistical distribution of asperity heights is assumed. In Fig. 4, since the frictional force acting on the tip is nearly linear with load, a model where the apexes of the asperities of the rougher tip surface interact with the graphite surface is consistent with our results. The observed nonlinearity of the friction with load could arise from the shear strength of the junction increasing with load or from deformation of the softer graphite surface, resulting in the normal lever displacement (and therefore the actual load) being slightly smaller than the sample displacement, which was used for computing the load.

It is surprising that the interaction of the large contact area of the tip, which is presumably inhomogeneous and quite disordered, results in the fractionally large periodic component observed in the frictional force. Suppose that at any localized region of the tip-surface contact, the frictional force is the sum of a periodic, conservative part and a nonconservative part opposing the direction of motion. Integrated over the entire contact area of the tip, the net periodic force will be small. In contrast, the nonconservative part will increase proportionally with the integration area. We conclude that *locally* the periodic force must be much greater than the nonconservative force. One possibility, as pointed out by Pethica for interpreting scanning tunneling microscope images of graphite,[8] is that the asperities drag one or more small flakes of graphite along the sample. If the graphite is aligned at the interface, the interaction would be coherent, generating a large periodic force. Another possibility[5] is that most of the load is supported by a nearly frictionless film of water or other lubricant, which is penetrated by a single microtip responsible for the frictional force. We hope that future work will clarify these issues.

To conclude, we have clearly shown that the atomic structure of a graphite surface influences the frictional dynamics of a tip sliding on this surface. Preliminary results in our laboratory also indicate that atomic-scale stick-slip can be observed on mica surfaces. These studies should lead eventually to an understanding of the atomic dynamics of friction.

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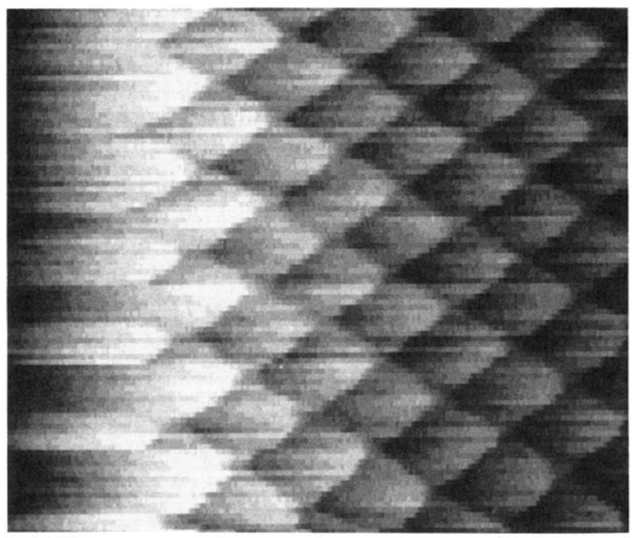

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