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Integration of contact size dependence and thermal activation in atomic friction

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ABSTRACT

Nano-asperity friction force has been found to depend on both the sliding velocity and the contact size. The former is a consequence of stress-assisted, thermally activated process, which is often explained by the one-degree-of-freedom Tomlinson model that assumes a virtual point contact sliding over a periodic tip–surface interaction potential. However, this model cannot provide an accurate description of the rate-determining process when coupled with the contact size dependence. In this synergistic experimental/modeling study, atomic friction measurements were conducted on cleaved mica and HOPG surfaces with varying normal force and sliding velocity, and the spatially-nonuniform saddle-point configurations were calculated from a Peierls framework. The agreement between experiments and theory suggests that the detailed characteristics of the interface slip field govern the atomic friction.

One critical question in the study of mechanically interacting rough surfaces is how single asperities deform in response to the normal and shear forces [1–5]. In the past several decades, the development of friction force microscopy (FFM) has enabled the quantitative measurement of friction behavior for nanoscale asperities with the contact sizes lying below ∼100 nm. The first noteworthy observation is the serrated force–displacement curves with stick–slip periodicity matching the substrate lattice constants. The Prandtl–Tomlinson model is often used to explain this observation, in which a point contact is sliding over a periodic potential that represents the tip–substrate interaction [4,5]. The point contact is connected to the loading apparatus (i.e., the cantilever in the friction force microscopy), and the low spring constant and high magnitude of the corrugated potential lead to the stick–slip behavior. While the model has been successfully applied in atomic friction, one obvious drawback is that all the interface atoms in contact are "forced" to move simultaneously, i.e., only one degree of freedom is allowed. We now address the consequences of this drawback in several recent experiments. Socoliuc et al. [6] found that the stick–slip behavior may disappear when the normal contact is reduced. Their interpretation based on Tomlinson model is the reduction of the corrugated potential magnitude (as the decrease of the applied normal force). The required reduction was found to be one or two orders of magnitude. In contrast, we view that the friction process involves an inhomogeneous slip of the interface atoms, and the boundary between atoms with slip displacement differing by one lattice constant is a dislocation. When the contact size is larger than the dislocation core size (i.e., the distance that slip field varies rapidly by one Burgers vector in a Peierls dislocation viewpoint), the friction behavior is achieved

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by dislocation nucleation and motion which result into a stress drop on the force–displacement curve. The disappearance of the stick–slip behavior corresponds to a small contact size that is comparable to or smaller than the dislocation core size. More importantly, this Peierls perspective does not invoke the change of the corrugated potential magnitude.

Another drawback with the Tomlinson model is related to the thermally activated friction. The atomic friction force often exhibits a logarithmic dependence on the sliding velocity [7,8]. Gnecco et al. [7] tested the frictional behavior of NaCl (100) surface with normal force on the order of nano-Newtons and with the sliding velocity, V, ranging from 5 nm/s to 1 μm/s. The friction force is found to be proportional to $\ln V$. A subsequent work by Riedo et al. [8] for cleaved mica surface demonstrated further that there is an asymptotic value of the friction force at very large V, and the variation of normal force (from 4 to 12 nN) does not change the $\ln V$ dependence. Also if the surrounding temperature decreases, the friction force will increase [9–11]. For instance, Jansen et al. [11] measured the friction force on a highly ordered pyrolytic graphite (HOPG) surface and showed the temperature dependence of friction force in the range of 100–300 K. All these observations suggest that the friction is a stress-assisted, thermally activated process. When tested at low velocity and high temperature, even if the applied force is lower than the athermal friction force, the atoms in contact may have a longer time (in the case of low sliding velocity) or a higher kinetic energy (in the case of high temperature), and thus a higher probability, to overcome the activation energy barrier. In spite of the wide acceptance of this view, we call into questions the determination of the activation energy by Tomlinson model. That is, in both Gnecco et al. [7] and Riedo et al. [8] that use the one-degree-of-freedom Tomlinson model, the corresponding saddle-point configuration for the activation energy calculation is apparently a homogeneous slip, which cannot treat contact size dependence as explained in the preceding paragraph.

In view of the restricted homogeneous motion of all interface atoms in the Tomlinson model, we argue that the fortuitous success of this model is the nanometer-sized contact in these experiments. The Peierls dislocation model [12] will degenerate into the one-degree-of-freedom Tomlinson model at small contacts, but will deviate from it when the contact size is larger than the dislocation core size. The hypothesis to be tested is that if the activated energy can be accurately calculated from an inhomogeneous slip of the interface atoms without the need of changing the magnitude of the corrugated potential, how will the results be compared to atomic friction measurements with a wide change of contact size and sliding velocity? Next we will present both theoretical calculations and experimental measurements.

The FFM experiments were conducted in an Atomic Force Microscopy (Agilent PicoPlus) in an atmosphere control chamber, where a small amount of nitrogen gas was continuously flown into this chamber. The cantilever had a monolithic silicon probe with a nominal radius of 10 nm. The normal force constant was in situ calibrated using a standard pre-calibrated cantilever with known bending

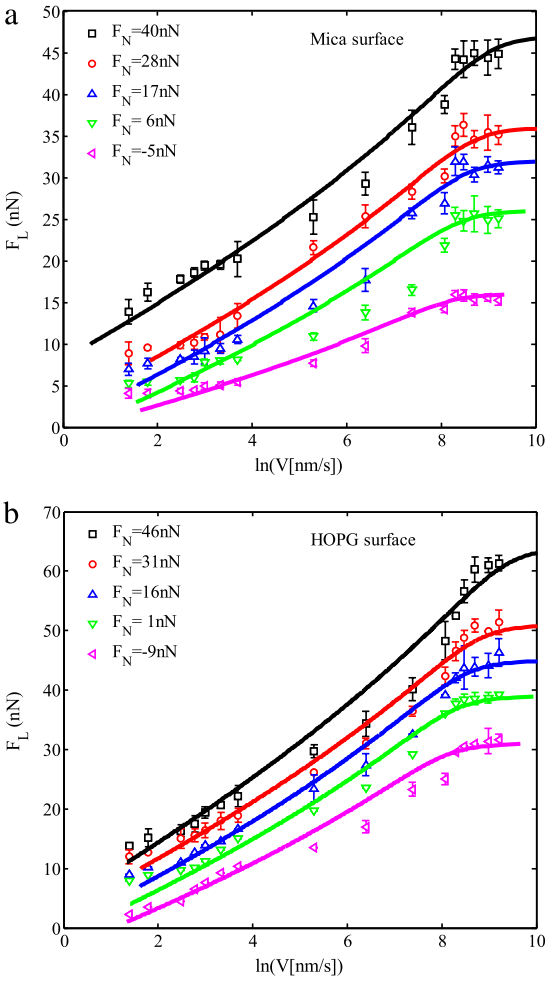

Fig. 1. Friction force (extracted as the half value of the bounds of the stick–slip curves during forward and background sliding tests) increases as a function of sliding velocity. Solid lines are fitting results from theoretical model in Eq. (3). FFM experiments were conducted on (a) mica and (b) HOPG surfaces.

stiffness. A simple and reliable way for friction force calibration with high accuracy and effectiveness was performed in situ using the recently developed levitation method [13–15]. The resulting normal and lateral stiffnesses were $k_{normal}^{cantilever} = 0.096$ N/m and $k_{lateral}^{cantilever} = 1.14$ N/m, respectively. In our first set of experiments, freshly cleaved and atomically smooth mica surfaces were used at room temperature and low humidity (<4%). The second set was conducted on freshly cleaved HOPG at room temperature and low humidity (2.7 $\pm$ 0.2%). Different scan dimensions were employed in order to obtain various scanning velocities in Fig. 1. Each data point in Fig. 1 corresponds to one friction measurement, from which the friction force is calculated as the half of the stick–slip peak force during forward scanning subtracted by the peak force during backward scanning.

Our measurements in Fig. 1 have the same characteristics as the previous measurements [7,8]. In a wide range of sliding velocity, the friction force F_L is proportional to $\ln V$. Eventually at sufficiently high V, the friction force reaches