

Sliding Friction of Solid Xenon Monolayers and Bilayers on Ag(111)

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We have measured the force required to slide one- and two-atom-thick solid films of xenon along a Ag(111) surface, and observe that the friction associated with the bilayer is approximately 25% greater than that associated with the monolayer. Our result sheds new light on the nature of atomic-scale frictional energy dissipation mechanisms, and demonstrates how localized to an interface such mechanisms can be.

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The fundamental origins of friction, an important physical phenomenon in light of both its everyday familiarity and its enormous economic impact, have been discussed and debated for at least 300 years, with very little resolved [1]. With the advent of new experimental techniques capable of studying the force of friction which results when a finite number of atoms is made to slide on a crystalline substrate [2], a new field involving investigations at atomic length and/or time scales, nanotribology, [3,4] has evolved.

At the atomic scale, friction is believed to originate primarily via sliding induced excitation of atomic lattice vibrations (phonons) [1], whose lifetimes are on the order of 10^{-9} – 10^{-12} s. Electronic contributions to the energy dissipation (associated with electronic motion excited by the sliding process) have also been suggested as potential contributors to frictional energy dissipation if one or both of the materials in contact is an electrical conductor [5]. Direct experimental evidence that atomic lattice vibrations and/or electronic effects do in fact give rise to friction has, however, been lacking.

We report here a quartz crystal microbalance study of the force required to slide solid monolayers and bilayers of xenon along a Ag(111) surface (Fig. 1). We observe that the friction associated with the bilayer is 27% greater than that associated with the monolayer, a result which is completely inexplicable from a macroscopic viewpoint. The result does, however, fall within the range of values currently expected for friction arising from a combination of electronic and phononic effects. The possibility of non-negligible electronic contributions to friction is of great interest, since to date the vast majority of fundamental theoretical treatments of wearless friction, i.e., that not associated with the wearing away of material [6], have considered only the phonon contributions [1]. A variety of practical applications become possible, moreover, in cases where electronic contributions dominate. For example, adsorbed films or particles on metal substrates could potentially be transported along the surface via electronic friction forces associated with an electrical current.

Monolayers and bilayers of xenon adsorbed on Ag(111) surfaces have been studied for many years, resulting in a

high degree of experimental precision and theoretical accuracy concerning the overall system parameters and the associated interaction potentials [7]. At 77.4 K, xenon condenses onto Ag(111) as an “uncompressed” close-packed triangular solid monolayer with an atom-atom spacing of 0.452 nm, or 5.624 atoms/nm². The xenon monolayer is highly incommensurate with the Ag(111) substrate, whose nearest-neighbor spacing is 0.236 nm. It accommodates further atoms by compressing until it reaches a spacing of 0.439 nm (5.97 atoms/nm²), the same spacing as that of (111) bulk solid xenon. Further accommodation of atoms into the first layer then becomes increasingly difficult. Subsequent atoms which are unable to force their way into the first layer form a two-dimensional gas phase in the second layer [8], which ultimately solidifies into a layer whose spacing is equal to that of the compressed monolayer, 0.439 nm. The second layer being perfectly commensurate with the first layer, one must assume that the two move in unison while sliding [9].

We measured the friction forces by condensing xenon gas at 77.4 K onto the (111) surfaces of silver films which had been electron-beam evaporated *in situ* at room temperature and in ultrahigh vacuum conditions onto quartz crystal microbalances with resonant frequencies $f \approx 8$ MHz and quality factors $Q \approx 10^5$. Shifts in f and Q resulting from xenon gas condensation indicate the number of adsorbed particles per unit area, and the degree to which they slip about on the silver surface due to the horizontal oscillatory motion of the microbalance. We experimentally observe that the xenon films slide one

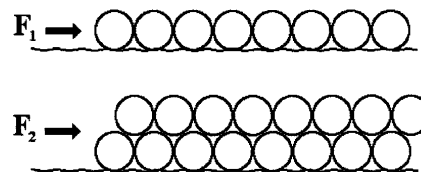


FIG. 1. Schematic of the experiment reported here. The force required to slide a two-atom-thick film of xenon along Ag(111) is compared to that required to slide a one-atom-thick film by monitoring the degree to which the films slide in response to horizontal shaking of the Ag(111) surface.

tenth of the peak-to-peak amplitude of this oscillation, with our best estimate of the actual sliding distance being about 2 nm. Since the sliding distance is well below the typical Ag(111) grain size (which is at least 30 nm [10]), the friction which we measure appears not to be dominated by the influence of substrate heterogeneity and defects [11]. These effects, if present at significant levels, would degrade the stepwise nature of an adsorption isotherm recorded on the surface [12].

Figure 2 depicts typical frequency and quality factor shift data versus the xenon gas pressure to which the sample is exposed (as measured by a spinning rotor gauge), where we have identified various features of the stepwise curve with the various stages of xenon film growth. At the lowest pressures the gas adsorption occurred under kinetic, rather than equilibrium conditions.

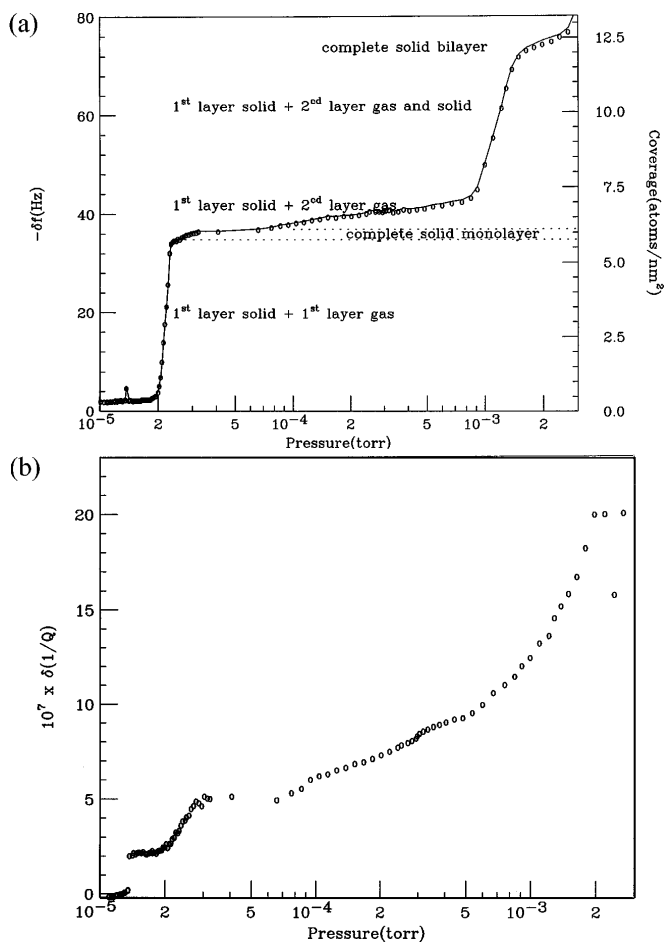


FIG. 2. Microbalance frequency shift and inverse quality factor shift versus residual gas pressure for xenon adsorbed on Ag(111) at 77.4 K. The various stages of xenon film growth are labeled, and identified with the particle coverage in atoms/nm², as determined by independent x-ray and electron scattering experiments. The solid line represents our estimate for the true film coverage, including corrections for slippage effects. Comparison of monolayer and bilayer friction levels were carried out for coverages of 5.97 and 11.94 atoms/nm², respectively.

Repetitive runs at varying condensation rates revealed, however, no significant differences in the friction versus coverage traces which we measured for coverages beyond the half monolayer level. The data points in Fig. 2 depict the actual measured frequency [13] and quality factor shifts. The frequency shift data slightly underrepresent the actual mass of the adsorbed film on account of film slippage effects (an extreme example of this occurs in the case of superfluid helium, which completely decouples from the oscillation of the underlying substrate [14]). The solid line in Fig. 2 represents our estimate for the true film coverage, correcting for slippage. The abrupt change in slope or “knee” in the frequency versus pressure curve near 35 Hz is indicative of a sudden decrease in the compressibility of the adsorbed film, unambiguously marking the completion point for the uncompressed monolayer [15]. The frequency shift at this point is within experimental error of that expected for adsorption on a flat surface, so the surface area probed by the first layer is essentially the same as that probed by the second layer [16]. The discontinuity in the $\delta(1/Q)$ curve at low pressure is indicative of an abrupt transition from pinning to sliding as coverage is increased at the initial stages of adsorption. This feature, although not an artifact, is not reproduced from one sample to another.

Characteristic slip times τ and friction coefficients η (the friction force per unit area per unit velocity) were determined via the relations [17]

$$\delta(Q^{-1}) = 4\pi\tau\delta f, \quad \eta = \rho_2/\tau, \quad (1)$$

where ρ_2 is the mass per unit area of the xenon film. Shorter slip times and higher friction coefficients thus correspond to greater friction forces coupling the film to the substrate.

Average particle slip times τ versus coverage for the Fig. 2 data set are presented in Fig. 3, where one monolayer has been defined as 5.970 atoms/nm², the spacing of the compressed monolayer. The trace exhibits

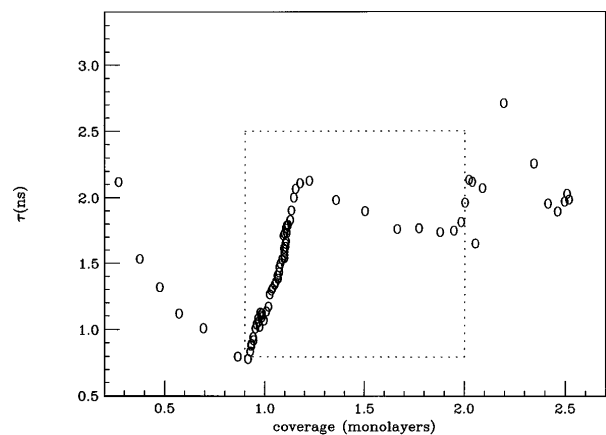


FIG. 3. Slip time τ versus coverage (1 monolayer = 5.970 atoms/nm²). The dotted box encloses the monolayer to bilayer regime which is redisplayed in Fig. 4.

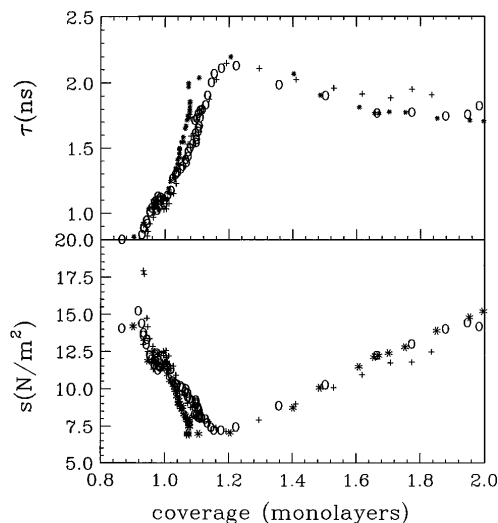


FIG. 4. Slip time τ and shear stress $s = \eta v$ (for $v = 1$ cm/s) versus coverage for three different Ag(111) surfaces. The shear stress for the two-atom-thick bilayer 15.1 ± 0.5 N/m² is approximately 25% greater than that associated with the one-atom-thick monolayer, 11.9 ± 0.4 N/m².

a high degree of sensitivity to the phase of the adsorbed film, including a small plateau in the vicinity of one monolayer, and distinct maxima in coverage regimes where relatively large numbers of adsorbed particles in a condensed gas phase are present, just beyond coverages of 0, 1, and 2 monolayers. The most straightforward explanation of the shape of the curve is that particles in the gas phase slide more readily than particles in the solid phase. This being the case, the slip times deduced for coverages corresponding to monolayer plus second layer gas phases would include contributions from the second layer gas-phase particles sliding on the first layer. We were therefore careful to make comparisons of friction forces for solid monolayers and bilayers with equal lattice spacings, and only in regions where the two-dimensional gas was at a minimum. These points occur at the coverages of 5.97 and 11.94 atoms/nm² in Fig. 2, which have been redefined in Figs. 3 and 4 as one and two monolayers.

The data within the dotted box in Fig. 3 are redisplayed in Fig. 4 along with data recorded on two other Ag(111) film substrates. The lower portion of the figure depicts the shear stress $s = \eta v$, or the force per unit area which would be required to slide the films at a constant speed of 1 cm/s (the order at which the films are slipping). The friction force per unit area associated with the two-layer-thick films, 15.1 ± 0.5 N/m², is 27% greater than that associated with sliding of the one-atom-thick film, 11.9 ± 0.4 N/m². Although we have not examined thicker film formation in detail for this study, it is clear that the friction force does not continue to increase substantially with increased coverage. This is consistent with results

from a previous study involving Kr films, where it was qualitatively observed that the friction did not increase significantly upon condensation of bulk crystallites [3].

Electronic friction arises from interfacial electronic interactions and is expected to be at most 5% greater for a two-atom-thick bilayer than a one-atom-thick monolayer [18]. The enhancement of phonon friction due to a decreased substrate-first layer spacing arising from the additional adhesive attraction of the second layer to the substrate has also been estimated to be no more than a few percent [19]. Preliminary computational results indicate, however, that phonon friction can be significantly (perhaps by as much as 100%) greater for a bilayer than monolayer, by nature of the fact that there are more vibrational modes present into which mechanical energy can be dissipated [20]. The experimental value of 27%, which is inexplicable from the point of view of any macroscopic treatment, falls well within the range of values expected from an atomic-scale viewpoint. As theoretical and simulational efforts become increasingly sophisticated in this area, an estimate of the proportion of energy losses associated with the motion of electrons to that associated with the excitation of atomic lattice vibrations for this system will become attainable. Our observation that the magnitude of the friction force is determined primarily by the first layer meanwhile demonstrates just how localized to an interface frictional energy dissipation mechanisms can be.

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