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Low-temperature static friction of N₂ monolayers on Pb(111)

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Abstract

Using a quartz crystal microbalance technique, we have measured the interfacial viscosity of nitrogen monolayers deposited on very homogeneous Pb(111) surfaces. At temperatures below 15 K, no dissipation is detected, suggesting that the N₂ films are rigidly coupled to the oscillating electrode. By raising the temperature close to 20 K, we find sliding of the nitrogen film for coverages above about 0.5 nominal layers. The observed temperature dependence is in good qualitative agreement with recent calculations on the static friction of a nitrogen slab in contact with a crystalline Pb substrate.

1. Introduction

Interest in the friction of a nanoscale system sliding on a solid surface [1] has grown considerably in the last ten years thanks to improvements in experimental techniques such as the atomic force microscope (AFM) [2], the surface force apparatus (SFA) [3] and the quartz crystal microbalance (QCM) [4], which have allowed measurements with an unprecedented resolution. Very recently, an AFM study of atomic scale friction has suggested new strategies to overcome the problem of stiction in microdevices [5]. Yet, fundamental questions are still unanswered. For instance, in the case of a solid monolayer adsorbed on a flat surface, one of the simplest possible geometries in which to investigate friction phenomena, does the sliding friction become larger if the substrate is a metal rather than an insulator? On a metallic surface there are two main dissipative processes: the sliding-induced excitation of phonons and of conduction electrons, whose relative contributions are still a matter of debate.

Krim and co-workers sought to explore the nature of electronic friction in a conducting material by measuring with a QCM the total friction associated with slippage of molecularly thin nitrogen films adsorbed on a Pb substrate as they passed through the superconducting

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transition temperature $T_c \sim 7.2$ K [6, 7]. They observed a sharp drop in the dissipation signal as the system was cooled below T_c . Renner *et al* [8] investigated the same system and found no sliding of the N_2 films at all temperatures from 4 to 77 K. We then decided to repeat this experiment in an ultrahigh-vacuum (UHV) system [9] specifically designed for QCM measurements in the temperature range from 4 to 400 K. In the case of Ne monolayers deposited on a very homogeneous Pb(111) electrode below 7 K, we found a pronounced depinning transition separating a low-coverage region, where the film was locked to the metal substrate, from a high-coverage region characterized by slippage [10]. More recent studies of Ne monolayers cooled down across T_c do not show any anomalous change in the dissipation [11]. This negative result may be explained in terms of the very weak coupling between the Ne atoms and the Pb conduction electrons caused by the low polarizability of Ne and lack of a quadrupole moment [12].

Hereafter we present some preliminary results of N_2 on Pb electrodes that show sliding, but only at temperatures above ~ 20 K. In the following, after a brief description of the QCM technique and of the experimental set-up, we summarize the main nitrogen results found so far.

2. Experimental set-up

The QCM is a very good probe with which to study interfacial phenomena. It consists of a small AT-cut quartz disc whose principal faces are optically polished and covered by a metal film. These films act both as conductive electrodes and as active surfaces. By applying an AC voltage across the QCM electrodes, the quartz faces move in a shear oscillating motion characterized by a typical resonant frequency f_0 and a quality factor Q . With an FM technique we can drive the QCM at its fundamental frequency or at one of its odd overtones [13]. The molecules deposited on the electrodes experience a lateral force $F = 4\pi^2 m A f_0^2$, where m is the molecular mass and A is the quartz oscillating amplitude. Any dissipation caused by the sliding friction of the adsorbate causes a reduction in the Q -factor, whose value is proportional to the resonance amplitude. Therefore one can determine the adsorbed film mass from the resonance frequency shift and the interfacial viscosity from a decrease in the resonance amplitude [14].

For this work, we used quartz blanks with the parallel faces optically polished. The rms roughness of the quartz surface was ~ 1 nm. For the realization of the Pb electrodes, the quartz was first annealed under UHV conditions up to 520 K to remove condensed impurities, then introduced in the previously cooled deposition stage and thermalized at the deposition temperature of 155 K. High-purity (99.999%) Pb was deposited by means of a water-cooled Knudsen cell in UHV (base vacuum 10^{-10} Torr) at a constant rate of 0.5 nm s^{-1} up to a thickness of 150 nm on each side of the quartz. The deposition parameters, in particular low substrate temperatures and high fluxes, were chosen in order to avoid three-dimensional growth (Volmer–Weber), obtain a uniform distribution of terraces nearly parallel to the quartz faces, and produce a low roughness over the whole electrode. The resulting films were made up of interconnected grains with a mean size of 0.5 μm and an rms roughness of 4.5 nm over the whole area, which was very low compared to the thickness of the film, as shown in figure 1. Each grain consisted of parallel platelets stacked with different in-plane orientations, mostly arranged parallel to the quartz substrate with a (111) termination, revealed by the high-resolution image shown in the inset of figure 1. A statistical analysis indicates that more than 95% of the exposed surface was tilted by less than 10° and the terrace size distribution appears peaked around 20 nm, with a significant population of terraces as big as 90 nm, both figures being much bigger than the driving oscillation amplitude of the quartz.

The QCM was housed in a specifically designed chamber [9] that combines UHV techniques with low temperatures. In this way, it was possible to significantly reduce the surface

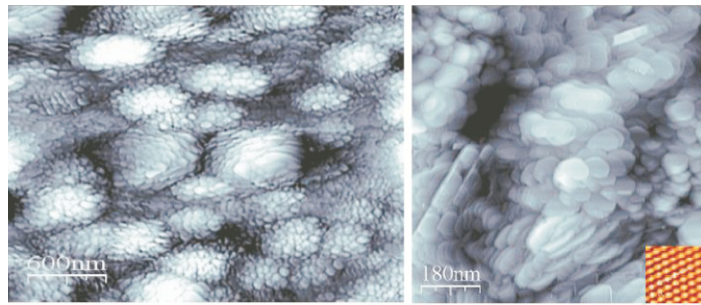


Figure 1. Right: large-scale STM image ($3 \mu\text{m} \times 3 \mu\text{m}$) showing different interconnected grains. The topography has been measured at room temperature in constant current mode. Left: detail of one of the grains ($900 \text{ nm} \times 900 \text{ nm}$). It is possible to see the different orientation of the platelets within each grain. The inset shows a closer view on one of the terraces, revealing its (111) structure, with a distance between two nearest atoms of 0.35 nm .

(This figure is in colour only in the electronic version)

contaminations of the QCM electrodes and to clean them by using a sputtering gun. High-purity N_2 was evaporated directly on one QCM electrode through a stainless steel capillary attached to a sapphire leak valve.

3. Experimental results

Figure 2 shows the resonance frequency shifts and the amplitude variations measured while dosing nitrogen on the Pb(111) electrode at temperatures of 9 and 15 K. The data have been acquired at the third overtone of a 5 MHz quartz plate characterized by a Q -factor of 300 000 at low temperature. The oscillating amplitude of the quartz plates is estimated to be $\sim 5 \text{ nm}$. Accordingly, the lateral force F experienced by a N_2 molecule is $\sim 0.002 \text{ fN}$. The resonance frequency stability of the QCM is $\sim \pm 0.1 \text{ Hz}$, essentially caused by the finite resolution of the digital counter, while that of the oscillation amplitude is $\sim \pm 1 \text{ mV}$. The frequency shift corresponding to the adsorption of a monolayer of N_2 on one QCM electrode is calculated to be equal to 2.8 Hz [6].

It is evident that no change in amplitude is observed while dosing. This is more clearly seen in the bottom graph of figure 2, which displays the normalized slip time τ_s [14] as a function of film coverage. This quantity can be calculated from the measured frequency and amplitude shifts of the QCM, and it represents the time required for the adsorbed film speed to decay to $1/e$ of its initial value after the oscillating substrate is stopped in the absence of a bulk vapour. A slip time equal to zero implies that the film is rigidly locked to the surface. We have repeated the data acquisition at each temperature a few times over a period of a week without finding any particular difference from the data of figure 2.

We have taken similar measurements at a temperature of 19 K, and a representative data set is displayed in figure 3. As before, no change in dissipation is observed until the film coverage reaches a value near 0.8 layers, beyond which dissipation starts, and the film slides. We have then acquired numerous isotherms by slightly changing the acquisition procedure: (i) dosing the nitrogen film continuously or in steps; (ii) changing the dosing rate; (iii) cleaning the Pb surface by heating up the QCM at different temperatures. Figure 4 shows four of these runs, which indicate the data reproducibility in this experiment. Although the data present some scatter, they clearly indicate a depinning transition at a coverage comprised between 0.6 and 0.8 layers. This behaviour is reminiscent of what we recently found [10] in the case of Ne films

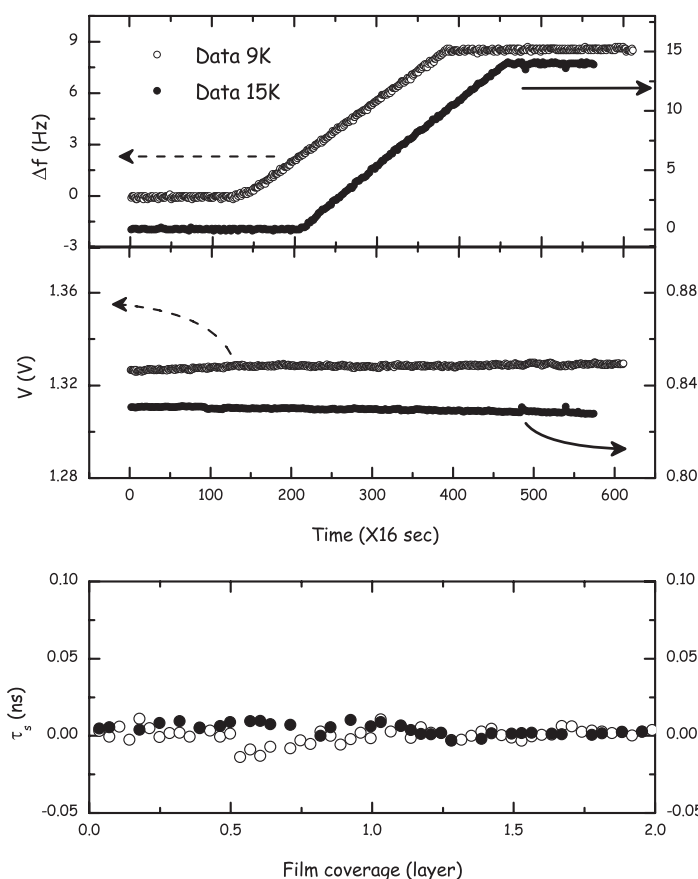


Figure 2. Raw data of the resonance frequency shift (top) and oscillation amplitude (middle) during an isothermal deposition of N_2 on Pb. Bottom: calculated slip time as a function of N_2 film coverage. Full circles refer to data taken at 9 K, empty circles to data at 15 K.

deposited at temperatures below 7 K. At this stage we still do not know whether the difference in the depinning onset observed with the two adsorbates is related to a different structure of the solid films or, more simply, it is caused by scatter in the data. Further structural and QCM studies are necessary to clarify this point.

The observed no-slippage of nitrogen at low temperature may be due to defects and/or contaminants of the Pb surface that act as pinning centres. Although our Pb surface is not an ideal crystalline surface, we do not think that this is the explanation for our results, for the following reasons. The STM characterization of the Pb surface morphology indicates the presence of atomic terraces with a characteristic size of 50 nm, while the QCM oscillating amplitude is of the order of 5 nm and the molecular size is ~ 0.4 nm. Accordingly, in the isothermal scan most of the molecules will experience the surface as being essentially flat. The same behaviour has been observed on three Pb electrodes grown in different times and with slightly different deposition parameters. During the experiment, these surfaces were sputtered at room temperature in order to recover the clean pristine state by removing a few layers of the Pb film. Due to the high mobility of Pb, recovery of surface damage induced by ion irradiation is very efficient already at temperatures around 300 K. Regardless of the variations in the

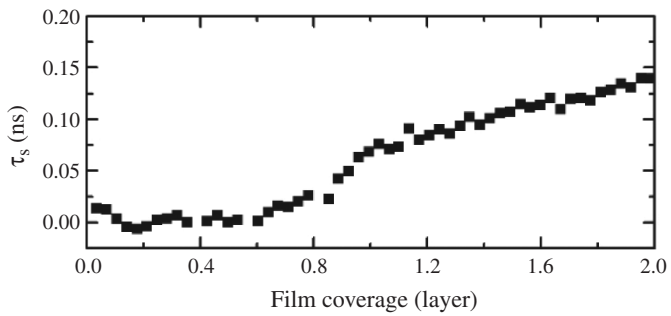
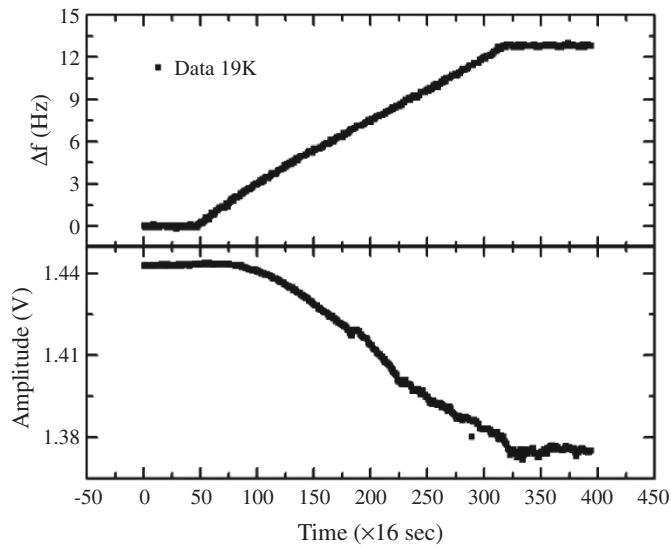


Figure 3. Raw data of the resonance frequency shift (top) and oscillation amplitude (middle) during an isothermal deposition of N₂ on Pb at 19 K. Bottom: calculated slip time as a function of N₂ film coverage.

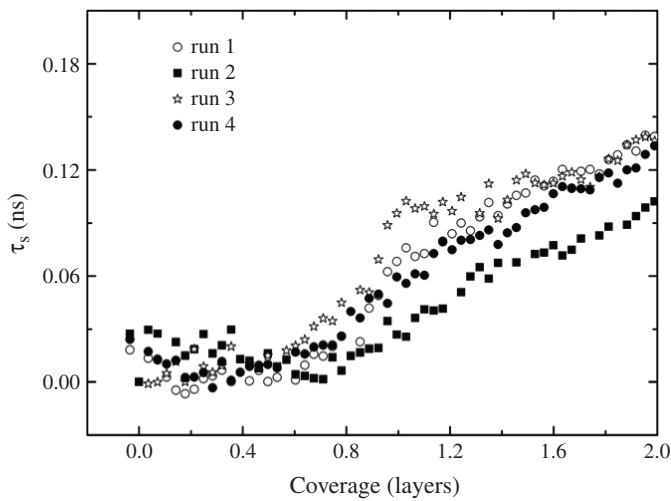


Figure 4. Normalized slip times of N₂ films deposited on Pb at 19 K in different conditions.

resulting morphologies we have never detected any sliding of N₂ below 15 K. The same results have been observed with the other adsorbates so far investigated, namely Ar, Kr and Xe. In contrast, Ne films show a well reproducible sliding below 10 K [10]. Similarly, D₂ monolayers

do slide at low temperatures, although the data are affected by a larger noise because of the low QCM resolution of a deuterium film.

In our opinion, these observations suggest that the static friction of heavy adsorbates at low temperature is not a direct consequence of the Pb surface quality but, more likely, reflects a strong corrugation in the resulting surface potential experienced by the film atoms/molecules. The weak force of inertia exerted by the QCM is not sufficient to move the atoms/molecules from their equilibrium positions and it is necessary to increase the temperature to activate the sliding process.

This interpretation is also consistent with very recent molecular dynamics simulations of the static friction of a N₂ slab on a crystalline Pb(111) substrate [15]. In the temperature range 0 to 20 K, the kinetic energy of the N₂ molecules is much lower than the corrugation of the surface potential, and the static friction force per molecule remains temperature independent and very large. At slightly higher temperatures, a misplacing of planes occurs at the interface, altering the N₂ fcc structure. Above 25 K, this misplacing moves into the bulk of the N₂ slab, and a sharp drop in the static friction is observed.

Such conclusions are in contrast with a recent work by Highland and Krim [16]. Although they agree that N₂ layers are highly susceptible to becoming pinned at low temperature, they find that superconductivity-dependent friction is readily observable whenever the films are not pinned. However, no details are provided on the success rate (e.g. how often the adsorbed films slide after cooling below 10 K) and on the possible reasons for pinning. Certainly, a major difference between our experiment and theirs is that we deposit the adsorbate directly onto the QCM in a controlled way, while Highland and Krim introduce a certain amount of gas to the QCM cell at 85 K and then cool it down to 4.2 K. Further measurements are currently under way in our laboratory to better understand the pinning of N₂ films at low temperatures.

Acknowledgments

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