Oscillation-induced static deflection in scanning force microscopy

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Employing an atomic force microscope (AFM) in conjunction with a quartz crystal microbalance, we have investigated how a high-frequency lateral oscillation of the substrate influences the imaging process of the AFM. It was found that the time-averaged deflection of the cantilever (both vertical and lateral) changed when the oscillation of the quartz resonator was turned on. The vertical-tip–substrate distance increased, while the lateral force of sliding decreased at the same time. A mode of imaging based on this effect was demonstrated. The oscillation was periodically interrupted at a rate of 73 Hz and the corresponding periodic modulation of the deflection was filtered and amplified using lock-in amplifiers. Slowly scanning the sample and displaying the outputs of the lock-in amplifiers versus \( x \) and \( y \) produced an image of the oscillation-induced static (OIS) deflection. Various mechanisms by which a lateral oscillation can generate a time-averaged vertical force are discussed. The fact that the vertical OIS deflection scaled linearly with amplitude and, also, that the OIS deflection was stronger on the sloped portions of the sample than on the flat regions, suggests a geometric interpretation. We term the phenomenon “cobble stone effect.” Experiments in liquids showed that the generation of an OIS deflection required immediate contact between the sample and the tip: a search for an OIS deflection in the presence of a liquid-filled gap between the sample and the tip yielded a negative result. Hydrodynamic forces are thereby ruled out as dominating factors. © 2007 American Institute of Physics. [DOI: 10.1063/1.2424445]

I. INTRODUCTION

The forces between a sharp tip and a planar surface are not at all a simple matter. These forces are the basis of atomic force microscopy and have been investigated in much detail in this context.\(^1\) There are numerous complications, including nonlinear interaction potentials,\(^2\) various types of hysteresis and memory,\(^3–5\) hydrodynamic forces,\(^6\) capillarity,\(^7\) the finite compliance of the sample,\(^8\) and not to mention damage and wear.\(^9\) While this wide variety of forces often precludes an unambiguous, quantitative interpretation of atomic force microscopy (AFM) images, it opens new avenues for the study of complex materials as well. Numerous modes of dynamic scanning force microscopy have been devised, which have considerably broadened our knowledge of complex samples and of forces on the colloidal scale.\(^10,11\)

In the work reported here, we have explored the consequences of a high-frequency lateral oscillation of the substrate. The sample is deposited on the front surface of a quartz crystal microbalance, where the latter is oscillated with an amplitude in the range of a few nanometers.\(^12\) Importantly, there is a large difference between the resonance frequency of the quartz crystal (which is 5 MHz) and the frequency of amplitude modulation (which was 73 Hz in our case). The cantilever only responds to the (slow) frequency of modulation. When the instrument scans at a rate of—let us say—20 pixels/s, the cantilever deflection is averaged over more than \(10^5\) cycles of oscillation for every single pixel. Even if the detection electronics were able to pick up a megahertz movement of the lever (which it is not), the lever would not be able to follow the megahertz motion of the crystal due to its inertia.

The fact that a periodic lateral movement of the sample creates a static vertical force is not necessarily a surprise. Such rectification phenomena are characteristic of nonlinear interactions (and the tip-sample interaction is known to be nonlinear). For a weakly nonlinear interaction, the efficiency of rectification should scale with the square of the amplitude of oscillation. A well known example of such an effect is the normal stress in polymer rheology.\(^13\) When a high-molecular-weight polymeric fluid is sheared rapidly, forces perpendicular to the direction of shear develop, which have to do with the alignment of chains. These forces must scale (at least) quadratically with the shear amplitude for reasons of symmetry. A second example of an oscillation-induced static force is the ponderomotive force in electrodynamics.\(^14\) This force requires a spatial gradient in the strength of the driving electric field. The ponderomotive force is employed in optical tweezers, where it pulls the colloidal sphere towards the laser focus. A hydrodynamic analog is conceivable. Finally, there is a rationalization of the oscillation-induced static (OIS) deflection which borrows from everyday experience. A cartoon is shown in the inset in Fig. 3. Consider a wheel
rolling on cobble stones. If the speed is high, the time-averaged position of the wheel is close to the peak of the asperities. The wheel does not have time to dive into the crevices in between the stones. For a somewhat slower motion, the wheel does follow the shape of the surface a little closer, leading to smaller time-averaged height of the axis above the surface of the street. We term this interpretation “cobble stone effect.” Putting it into more physical terms, it is essential that the interaction between the wheel and the street is strongly nonlinear. The vertical force onto the wheel abruptly ceases once the wheel and the street are out of contact. What causes the time-averaged vertical displacement is the concerted action of the nonlinear force (pushing the wheel up) and inertia (preventing the wheel from adhering to the street).

Lateral oscillations have been frequently exploited in scanning force microscopy. For example, Dinelli et al. have studied the compliance of the sample by laterally oscillating it, while it was in contact with the AFM tip. They term the technique “lateral force modulation.” These authors used small amplitudes in order to avoid nonlinear interactions. The goal was linear mechanical spectroscopy on the nanoscale. Krotil et al. have used higher amplitudes and thereby reached the onset of sliding. A few groups have specifically employed a high-frequency (megahertz) excitation to probe the tip-sample interaction. At high frequencies, the trajectory of the tip is mostly governed by the balance between the tip-sample force and inertia (“acceleration force microscopy”). The spring constant of the cantilever then is of minor importance. The use of high-frequency excitation eliminates certain relaxations, mostly occurring on soft samples such as rubber surfaces or biological specimens.

Oscillation-induced vertical forces have recently been discussed in the context of acoustic sensing. Dultsev et al. have observed that small particles can shake off the surface of quartz crystal microbalance by ramping up the amplitude to exceptionally high values. Edvardsson et al. have performed similar investigations. These authors monitored how the instrumental setup employed here is simple: The sample is deposited on a quartz crystal resonator. Quartz resonators have resonance frequencies in the megahertz range. The amplitude of motion can be up to a few tens of nanometers. Again, the detector is too slow to resolve a megahertz deflection of the cantilever, should it exist. We only observe the time-averaged deflection, where the averaging period is much longer than the period of oscillation (but shorter than the period of modulation). When the AFM tip scans the oscillating surface, the rapid oscillation prevents intimate contact. This has two consequences: the rapid movement of the substrate drives the tip away from the surface and it reduces time-averaged force of friction. To the best of our knowledge, the literature does not mention such an effect in the context of scanning force microscopy. Michael Rodahl (Q-sense AB, Sweden) has verbally reported on this phenomenon.

II. EXPERIMENT

Figure 1 shows a sketch of the experimental setup. The substrate was a quartz crystal resonator (149211-1, Maxtek Inc., Santa Fe Springs, CA) with a diameter of 1 in. and a fundamental frequency of 5 MHz. The crystal was coated with gold electrodes. The electrodes were keyhole shaped in order to provide energy trapping. The crystal was mounted in the holder CHC100 (Maxtek) with the front electrode connected to ground. It was driven by a network analyzer (E5100, Agilent) with a power of 10 dBm unless stated otherwise. A power of 10 dBm corresponds to an electrical amplitude of \( U_{el,0} = 1 \text{ V} \). Most of the measurements were carried out on the fundamental at 5 MHz. The \( Q \) factor of the resonance was determined with the network analyzer as \( Q = 15 \ 000 \). The dissipation is partly caused by viscous losses in the crystal and partly by Ohmic resistance of the switch (Fig. 1). The frequency of the resonator was fixed and chosen to be on the peak of the resonance.

From the driving voltage and the \( Q \) factor, one calculates the amplitude of oscillation \( u_0 \) as

\[
    u_0 = \frac{4}{(\pi n)^2 d_{33} Q U_{el,0}},
\]

where \( n \) is the overtone order \((n=1 \text{ for the fundamental})\) and \( d_{33} = 3.1 \times 10^{-12} \text{ m/V} \) is the piezoelectric strain coefficient. Inserting the values, we find an amplitude of 20 nm. Multiplying by the radial frequency \( \omega \), one finds a maximum speed of \( v_0 = \omega u_0 = 0.6 \text{ m/s} \).

During imaging, the oscillation was periodically interrupted by means of an electronic switch (ZAD 3H, Minicircuits). The switch was controlled by a square wave generator (Kontron 20 MHz programmable function generator, Series 8200). The square wave was used in order to obtain sharp edges in images such as shown in Fig. 7. When using the lock-in amplifier (LIA) for detection, modulating with a sine wave rather than a square wave would not have made much of a difference because the LIA outputs a weighted integral of the signal, in any case. We used a modulation frequency of 73 Hz unless stated otherwise. The choice of the modulation...
frequency was guided by the following two considerations: On one hand, one tries to modulate as fast as possible in order to stay away from 1/f noise and, also, to allow for fast imaging. On the other hand, a measurement of the OIS deflection as a function of modulation frequency showed a decrease of the OIS deflection even below the bandwidth of the resonance (see Sec. III). The resonator itself responds to a change in drive level within a time of $\tau = (2 \pi \Gamma)^{-1} \sim (960 \text{ Hz})^{-1}$, where $\Gamma$ is the half-band–half-width of the resonance (153 Hz for the crystal used here). The response time of the resonator would have allowed for modulation frequencies in the range of a few hundred hertz. However, there appeared to be a second, slower time constant for the cantilever deflection in response to a sudden change of oscillation amplitude. The choice of the modulation frequency of 73 Hz reflects the need to stay below this second, slower response rate.

The atomic force microscope was the unit Nano Wizard (JPK Instruments, Berlin, Germany). This stand-alone setup was placed on a custom-made table fixing the quartz holder in a defined position. The vertical and lateral deflections can be externally monitored via analog outputs supplied by the “access box.” Both signals were fed into lock-in amplifiers (EG&G dual phase lock-in amplifier, model 5210), which were referenced to the square wave generator used for modulation. The sensitivities of the amplifiers were 10 mV (full scale). The time constants were 30 ms. The amplifiers were set to display magnitude ($R$) and phase ($\theta$) on the analog outputs. The output of the magnitudes $R$ were fed back into the access box and displayed as images employing the control software of the AFM. We also monitored the phase $\theta$ but did not find interesting features. In the following, we only discuss the magnitude of the cantilever response $R$ which is synonymous to “OIS deflection.”

Calibration was carried out on the basis of the comparison of the voltage output of the lock-in amplifier to the variance of gray values of an artificial image taken under conditions of slow modulation (such as shown in Fig. 2). The conversion factor was 1.2 V/nm.

III. RESULTS AND DISCUSSION

In order to demonstrate the existence of an oscillation-induced static force on a qualitative basis, we show an artificial “image” taken on a single spot (Fig. 2). The image size was chosen as 1 nm (the smallest size allowed by the software), leaving the cantilever almost in place. The image therefore displays a time sequence. In this case, the data acquisition rate (the inverse data acquisition time per pixel) was chosen to be lower than the modulation frequency. As a consequence, stripes appear in the image. Evidently, stripes only occur if there is a synchronization in the timing of the line scans and the timing of the modulation. Such a synchronization can be achieved by slight adjustments of the modulation frequency on a trial-and-error basis. As the picture shows, the response of the tip to a change in amplitude is not simple. The stripes are not just light and dark. Rather, the tip jumps quickly when the oscillation is turned on, leading to a sharp edge. However, it does not return to the ground state in the same rapid way after the excitation is turned off. The fact that the tip retracts from the sample during the “on” periods (rather than the “off” periods, where “on” and “off” indicate whether or not the crystal was driven) became evident when modulating very slowly (periods of a few seconds). The bright stripes could be assigned to the “on” periods based on visual observation. No stripes occurred in the images of the lateral deflection when the image was taken on a fixed spot. Since the tip did not move, the friction force was zero and stayed zero (on average) even with the oscillation turned on. Stripes did occur in lateral deflection images when the tip moved (cf. Fig. 7). In the latter case, the stripes visualize the oscillation-induced reduction of the friction force.

Figure 3 shows the amplitude dependence of the OIS deflection. While taking an artificial image of a 1 x 1 nm² spot, the electrical drive level of the crystal was increased in steps of 2.5 dBm. Panel (a) shows line averages as a function of the line index. From the slight slope on the plateaus, one
concludes that there is some drift of the OIS force with time. Given that the OIS force was found to vary strongly with $x$ and $y$ (cf. Fig. 5), we attribute these drifts to a lateral drift of the tip on the sample. Panel (b) shows the dependence of the OIS deflection on amplitude. Clearly, the OIS deflection scales linearly with oscillation amplitude. The linear dependence on amplitude came as somewhat of a surprise, because the normal stresses in rheology depend on the square of the shear stress. A linear dependence would be forbidden by symmetry. For an isotropic medium, the normal stresses must be independent of the direction of shear. Such an invariance with regard to a sign reversal is incompatible with linear scaling. The phenomenon encountered here clearly is not compatible with a quadratic dependence on amplitude and must therefore have another explanation. We invoke the cobble stone effect as outlined in the Introduction in order to explain the linear amplitude-deflection relation. The interpretation in terms of the cobble stone effect is also corroborated by the finding that the OIS deflection is large on parts of the sample which are strongly sloped (Fig. 5, see the discussion below).

Figure 4 shows the dependence of the OIS force on the modulation frequency. During acquisition of an artificial image, the modulation frequency was chosen to have values of 50, 73, 200, 500, and 1000 Hz. The OIS deflection dropped substantially with frequency between 50 and 200 Hz. This range of frequencies is much below the expected response rate of the crystal itself, which is around $2\pi \Gamma \sim 960$ Hz. If the dynamics of the OIS effect was entirely due to the $Q$ factor of the crystal, the response time of the OIS deflection should be the same as the time needed to turn the crystal on and off. This time is equal to $(2\pi \Gamma)^{-1}$, where $\Gamma$ is the half-band–half-width of the resonance. Since the response times of the OIS deflection and the response time of the crystal are different, the decrease of the OIS amplitude with frequency must be related to the interaction between the crystal and the cantilever and not to the dynamics of the crystal itself.

In Fig. 5, we demonstrate that the OIS deflection can be a basis of an imaging mode. This particular image was taken on a bare gold surface. Panels (a) and (b) show the mean vertical and the mean lateral deflections (averaged over times longer than the modulation period), whereas panels (c) and (d) show the vertical and the lateral OIS deflections, respectively. The acquisition time was around 4 h. Most notably, the vertical OIS deflection [panel (c)] is large at those spots which have a large slope in the topographic image [panel (a)]. An area which shows this effect is circled in panels (a) and (c). We rationalize this finding as depicted in the inset of Fig. 3. With regard to the lateral OIS deflection [panel (d)], the situation is less clear. The lateral OIS deflection appears to correlate to gradients in the lateral force. Some of the lines going from the upper left to lower right in panel (b) appear as double lines in panel (d).

The linear dependence of the OIS force on amplitude and the fact that the OIS force is strong on the edges of the topographical features support the interpretation in terms of the cobble stone effect [inset in Fig. 3(a)]. The vertical tip deflection is governed by the highest asperity in reach of the tip, where “in reach” designates the area covered by the oscillation. On a sloped portion of the sample, the maximum height is proportional to the amplitude of oscillation and to the local slope.

In order to demonstrate material contrast, we deposited a drop of the soft polymer polyisoprene (70 000 g/mol, deposited from a 3 mg/mol solution in toluene with a Nano-Plotter, Gesellschaft für Silizium-Mikrosysteme mbH-GESIM, Dresden, Germany) onto the crystal and imaged the edge of the drop (Fig. 6). The drop is situated on the lower right. The dashed line in the upper panel denotes the edge of the drop as inferred from the topographic image (not shown). While the OIS deflection disappeared on the drop far away from the contact line (lower right), there appeared to be an increased OIS deflection close to the three phase line. The absence of OIS deflection in the center of the drop is easily explained by the finite penetration depth of the ultrasonic shear waves in the soft polymer. Above a thickness of a few microns (depending on the modulus of the material), the upper surface stays at rest. Interestingly, the transition into this quiet region is not given by a smooth decrease but rather by an increase and an abrupt drop. This increase is seen in both
the trace and the retrace data and therefore is not a kinetic artifact. We tentatively attribute this maximum in the OIS deflection to the film resonance.\textsuperscript{28,29} When the thickness of the drop equals a quarter of the wavelength of sound, the film itself forms an acoustic cavity, which is resonantly driven by the movement of the substrate. The situation is analogous to a vibrating reed. In this thickness range, the top of the polymer layer oscillates with an amplitude even larger than the amplitude at the bottom of the film. Possibly, the increased vertical OIS deflection at intermediate thickness is related to this phenomenon.

Imaging a gold surface in a liquid environment, we found the vertical OIS force to have completely disappeared. However, the lateral OIS deflection was still sizable, as demonstrated in Fig. 7. The data acquisition rate in this case was less than the modulation frequency, leading to a striped pattern. The bright and dark portions of the image are the "off" and "on" periods, respectively. We interpret this image in the sense that the oscillation reduces the time-averaged friction force.

Finally, we investigated whether the generation of an OIS deflection requires immediate contact with the surface or whether it can be mediated across a liquid gap. Standard lubrication theory predicts that a fluid streaming in the wedge between a sphere and a plate should not produce a net vertical force.\textsuperscript{30} Lubrication theory assumes a small Reynolds number. In principle, the inclusion of the nonlinear term of the Navier-Stokes equation might lead to a vertical force in analogy to the Bernoulli force. Weak Bernoulli forces above an oscillating surface have been recently discussed in the context of acoustic second harmonic generation.\textsuperscript{31} In order to search for such an effect, we monitored the vertical OIS deflection while approaching a small sphere glued to a cantilever towards the oscillating crystal surface. The diameter of the sphere was 12 $\mu$m. Figure 8 shows the deflection-distance curve (upper part) and the corresponding OIS deflection (lower part). Within the resolution of the instrument, the OIS deflection vanished when the tip had detached. If there were to be an OIS force mediated by the liquid, one would expect a slightly nonzero OIS deflection within a range of the escape depth of the shear wave (250 nm, dashed ellipse in Fig. 8). We have searched an OIS deflection with increased sensitivity and an increased time constant of the lock-in amplifiers, thereby decreasing the detection limit to a about 0.01 nm. The result was negative. This finding rules out hydrodynamic explanations of the OIS force.

![FIG. 6. (Color online) OIS deflection image taken on the edge of a drop of polyisoprene deposited on the gold electrode. The drop is on the lower right of the image. The OIS deflection is almost entirely absent on the soft, sticky polymer. The size of the image is 13 $\times$ 13 $\mu$m$^2$. (a) Vertical OIS deflection, scanning from left to right. The dashed line denotes the edge of the droplet as inferred from the topographic image (not shown). (b) Vertical OIS deflection, scanning from right to left. (c) Lateral OIS deflection, scanning from left to right.](image1)

![FIG. 7. (Color online) Image of the lateral OIS deflection taken on a bare gold surface in water. The modulation frequency was below the data acquisition rate. The stripes correspond to the on and off periods (cf. Fig. 2). In this case the tip did scan, resulting in a finite lateral deflection. The lateral deflection (proportional to the coefficient of sliding friction) was much decreased by the oscillation. The size of the image is 10 $\times$ 10 $\mu$m$^2$.](image2)

![FIG. 8. OIS deflection vs piezotravel acquired in water with a 12 $\mu$m sphere glued to the cantilever. The upper trace shows the time-averaged deflection. Once the sample is out of contact, the OIS deflection (lower trace) vanishes. A static oscillation-induced force requires direct contact between the sample and the tip; it is not produced across a layer of liquid.](image3)
IV. SUMMARY

Oscillating the substrate in atomic force microscopy at a frequency of a few megahertz and an amplitude of a few nanometers leads to a time-averaged vertical deflection and a decrease in the time-averaged friction force. For the case investigated here, the vertical deflection could be traced back to a geometrical effect, termed “cobble stone effect.” The strength of the OIS deflection is small on soft samples. The vertical OIS deflection requires immediate contact between the tip and the sample; it is not mediated across a liquid gap.

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