A shear-force mechanism between a chemically etched scanning near-field optical microscope tip and different chemically treated atomic force microscope cantilevers has been experimentally and theoretically investigated as a function of the tip-to-sample distance for different amplitudes of the tip oscillation. The experimental results show, in agreement with the theoretical predictions, that as the tip approaches the cantilever, the electrostatic force is the most influential in the shear-force mechanism, independently of the nature of the tip or the sample. As the tip-to-sample distance decreases, other forces come into play, and the type of interaction depends on the chemical nature of tip and sample surfaces. Thus, for hydrophobic cantilevers, the decrease in the vibration amplitude is mostly due to the solid friction forces resulting from electrostatic interactions. However, if the sample surface is hydrophilic, there is a decrease in the electrostatic force, a water meniscus is formed, and the decrease in the tip amplitude is mostly due to dynamic friction related to capillarity.

Keywords—Capillary forces, coulombic forces, friction, hydrophilicity, hydrophobicity, scanning near-field optical microscopy, shear force.

I. INTRODUCTION

The large majority of scanning near-field optical microscope (SNOM) configurations are based on shear-force control of the tip–sample distance. Shear force was first discussed by Betzig et al. [1] and Toledo-Crow et al. [2]. They found that the vibration of a tapered optical fiber laterally oscillating at its resonance frequency and approaching to a sample surface is damped when the tip-to-sample distance is decreased below a few tens of nanometers. Although the name “shear force” is widely accepted, it is somehow misleading, since the damping is not related to shear stress. The origin of the damping mechanism is still unclear. At this point, there is no satisfying theory to explain this interaction between tip and sample surface. Some authors suggest that one of the most important reasons for a decrease in the vibration amplitude is due to the presence of a contamination layer at the sample surface [3], [4], resulting in capillary forces. When the formation of a water contamination layer is prevented, by working, for instance, in vacuum [5] or with hydrophobically treated surfaces [6], damping is related to dry friction.

The objective of this paper consists of determining, theoretically and experimentally, which are the forces acting on the tip as it approaches the sample surface. A theoretical model to explain the interaction forces is briefly presented, and subsequent experiments are done to measure the forces. The experimental approach is based on a measurement of approach curves with a near-field probe onto an atomic force microscope (AFM) cantilever used as test sample. The SNOM probes used in this work are obtained by chemical etching in hydrofluoric acid of single-mode optical fibers and subsequent evaporation of an aluminum layer [7]. Different fiber tips are used to study the influence of the aluminum layer in the shear force. Furthermore, chemical treatments of the cantilever allow studying the differences of the tip–cantilever interaction when the sample is hydrophobic or hydrophilic, expressing the influence of a water contamination layer. Finally, the influence of the tip diameter is studied as well. Two parameters are monitored simultaneously: the SNOM shear-force signal, which is the oscillation amplitude of the near-field optical fiber probe and gives access to lateral forces, and the AFM signal, which relates to the AFM cantilever bending force and measures vertical components of the damping force.

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II. THEORETICAL CONSIDERATIONS

At the nanometer scale that is of interest here, the electromagnetic interaction is the most important of the fundamental forces observed in nature. It can be seen as the origin of many other related forces such as friction, adhesion, van der Waals, capillarity, etc. In this section, we study the various expressions of this electromagnetic interaction and describe the related forces acting between the tip and sample in the case of SNOM. A theoretical estimation of the order of magnitude is presented for the different forces, based on a simplified model valid when the tip-to-sample separation is much shorter than the tip radius. The sample is simply considered as a flat semi-infinite medium, and the tip is approximated by a sphere of radius \( R \). The forces are thus determined based on the interaction energy per unit area as a function of the tip-to-sample separation.

Electrostatic forces are due to free charges in the tip that induce image charges on the nearby sample surface. As indicated in [8], an attractive coulombian force is acting on these charges that can be expressed as

\[
F_{\text{electr}} = \frac{-q_1q_2}{4\pi\varepsilon_0\varepsilon_r\varepsilon_s\varepsilon_m^2} \left( \frac{\varepsilon_s - \varepsilon_m}{\varepsilon_s + \varepsilon_m} \right)
\]

where \( q_1 \) is the image charge of \( q_2 \), the two charges being separated by distance \( r \). \( \varepsilon_0, \varepsilon_s, \) and \( \varepsilon_m \) are the dielectric constants of free space, sample, and medium, respectively. From this formula, it is clear that when there is a medium different than air between tip and sample, a decrease in the coulombian force is obtained. Thus, in the case of water (\( \varepsilon_{\text{m,water}} = 78 \)), the coulombian interaction decreases almost two orders of magnitude with respect to air. The direction of the coulombian force is colinear with the vector connecting the two charges. In the general situation of a vibrating tip, it has a horizontal as well as a vertical component.

It has to be noted that in the presence of water or electrolyte, the electrostatic interaction between tip and sample is modified by the presence of mobile charges in the surrounding liquid. Counter ions from the polar liquid screen the charges present at the surface of the solid (tip and sample). When the tip is approaching the sample, these two charged layers interact then repulsively through a double layer interaction force [9].

Van der Waals force is related mostly to the nonzero instantaneous dipole moments of all atoms and molecules that lead to material induced polarization at the atomic scale. Using Derjaguin’s equation, it can be seen that the van der Waals force between a sphere and a flat surface is inversely proportional to the square of the tip-to-sample distance [10]. However, the proportionality factor (Hamaker constant) is on the order of \( 10^{-20} \) J for air or vacuum and is further reduced in water. This implies that van der Waals forces will be nonnegligible only for very small tip-to-sample distances, typically in the angström range. In the approximation considered here, van der Waals force has only a vertical component.

In the presence of water vapor, condensation onto sample and tip may lead, at small tip-to-sample distance, to formation of a water meniscus that fills the gap. This formation critically depends on relative humidity and geometry of the tip and leads to a capillary force. For a spherical tip of radius \( R \) and a flat sample, this force can be expressed as [10]

\[
F_{\text{cap}} = \frac{4\pi R \gamma_L \cos \theta}{(1 + D/d)}.
\]

Here, \( \gamma_L \) is the specific surface energy for water (0.072 J/m²), \( D \) is the tip-to-sample separation, and \( \theta \) is the contact angle between the tangent to the sphere and the meniscus. The geometrical parameter \( d \) corresponds to the meniscus height minus the tip-to-sample distance (see [10]). Again, in the simplified model considered here, the capillary force is vertical.

Viscous force is acting on the tip when vibrating in any medium with nonzero viscosity. Based on the Navier–Stokes equation of fluid dynamics [11], one can see that for a simplified situation where the tip apex is a flat disk of radius \( r \), this force is horizontal, proportional (through the viscosity coefficient of the surrounding medium) to the velocity of the tip and inversely proportional to the tip-to-sample distance. Viscous force is evaluated here for a typical value of the tip velocity of \( 5 \times 10^{-4} \) m/s.

When contact occurs, the presence of nonzero resultant force acting at the tip apex leads to friction that is proportional to the resultant force through a friction coefficient. Two friction modes can be distinguished: a dynamic friction (“slip”) and a static mode (“stick”). In the dynamic friction, the tip apex vibrates with a certain velocity and slides onto the sample surface when contact occurs with the AFM cantilever. In the static mode, the tip extremity sticks onto the cantilever surface, with zero velocity of the tip apex. Static friction is expressed by horizontal friction force acting at the contact point, whose magnitude is given [12] by

\[
F_f = \mu N
\]

where \( \mu \) is the static coefficient of friction, which in silica takes the value of 0.8 [13], and \( N \) is the magnitude of the sum of all vertical forces at a tip-to-sample separation equal to the sample roughness. Similar expression characterizes the dynamic friction force, the dynamic friction coefficient being typically 2 to 10 \( \times \) smaller than the static one for materials involved in this study.

Table 1 summarizes for a typical tip radius of 100 nm the different forces acting on the tip vibrating above a Si cantilever. The values are based on the simplified models described above and are reported for several tip-to-sample distances. To evaluate the importance of the surrounding medium, values are given for the same tip vibrating in air as well as in water. Very different orders of magnitude are obtained for the different forces. One sees that coulombian forces are dominant down to very small distances. When working in water, capillary forces are important as well at large distances. However, viscous forces that were often considered in the past as responsible for shear-force damping seem to play only a minor role. As already mentioned above, van der Waals forces come into play only at very short
Table 1
Magnitude of the Forces Acting on the Vibrating SNOM-Tip

<table>
<thead>
<tr>
<th>Force</th>
<th>Tip-to-sample distance [nm]</th>
<th>Amplitude (air) [N]</th>
<th>Amplitude (water) [N]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coulombian</td>
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<td>-5 \times 10^{-2}</td>
<td>-7 \times 10^{-5}</td>
</tr>
<tr>
<td></td>
<td>1</td>
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<td>5</td>
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<td>-3 \times 10^{-4}</td>
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<td></td>
<td>10</td>
<td>5 \times 10^{-5}</td>
<td>-7 \times 10^{-5}</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>1 \times 10^{-3}</td>
<td>-2 \times 10^{-3}</td>
</tr>
<tr>
<td>Van der Waals</td>
<td>0.1</td>
<td>-8 \times 10^{-4}</td>
<td>-8 \times 10^{-6}</td>
</tr>
<tr>
<td></td>
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<td>-8 \times 10^{-2}</td>
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<td>3 \times 10^{-5}</td>
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<tr>
<td></td>
<td>20</td>
<td>2 \times 10^{-4}</td>
<td>-2 \times 10^{-4}</td>
</tr>
<tr>
<td>Capillary</td>
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<tr>
<td></td>
<td>20</td>
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<td>4 \times 10^{-2}</td>
</tr>
<tr>
<td>Viscosity</td>
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<td>4 \times 10^{-2}</td>
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<tr>
<td></td>
<td>20</td>
<td>2 \times 10^{-4}</td>
<td>-2 \times 10^{-4}</td>
</tr>
</tbody>
</table>

Distances, and are quite often masked by electrostatic forces resulting from charge effects, as is well known from AFM force studies. Note that the values given here are deduced from very simple models that allow making only a qualitative analysis of the relative importance of the different forces involved.

III. EXPERIMENTAL CONSIDERATIONS

Approach curves are recorded when approaching the SNOM tip, oscillating at its resonance frequency, to the flat part of an AFM cantilever, measuring simultaneously the damping of the SNOM tip vibration amplitude and the bending of the cantilever. Two approach curves are thus simultaneously recorded: one for the SNOM probe (SNOM signal) and one for the AFM cantilever (AFM signal). Analysis of these two parameters as a function of the vertical damping force exerted on the cantilever due to the tip interaction allows identifying the mechanism responsible for shear-force damping.

The complete experimental setup is schematically presented in Fig. 1. The SNOM tip is mounted on a holder, and a lateral vibration at the tip eigenfrequency is piezoelectrically induced. Tip eigenfrequency is around 11.6 kHz. The tip and its holder can be moved vertically with a linear motor over a range of 25 mm. The tip vibration amplitude is measured with a laser feedback interferometer that offers subnanometer accuracy. The laser used in the feedback interferometer emits 4 mW of red light at 674 nm (Toshiba), and the interference signal is detected through a lock-in amplifier. A complete description of the interferometer setup has been given elsewhere [14]. The AFM signal is recorded as the deflection of the AFM cantilever measured optically with a two-quadrant detector. Only the bending is relevant in the present experiment, the tip oscillating in a plane parallel to the cantilever axis. The AFM laser emits in the range 630–680 nm a total light power smaller than 5 mW. Both SNOM and AFM signals are amplified and fed to a computer. The fine approach between the SNOM tip and the AFM cantilever is done via a piezo tube, which supports the cantilever, and offers a total vertical range of 3 mm for a voltage ramp of 250 V. Commercial silicon cantilevers are used (Lot Oriel). SNOM probes are fabricated in house from chemically etched optical fibers [7]. The Al-coated tips are obtained by deposition of a 100-nm-thick layer of aluminum on a bare SiO$_2$ fiber tip.

The changes in the tip-to-sample interaction are studied based on different chemical properties of the probes. For the SNOM probe, bare SiO$_2$ tips as well as Al-coated tips have been used. For the AFM probe, the Si-cantilevers have been chemically functionalized in several ways. Specific water affinity is obtained by deposition of organic monolayers (thiols). Coating with octadecanethiol [CH$_3$(CH$_2$)$_{17}$SH] leads to hydrophobic cantilevers, while treating the silicon with 16-mercaptoundecanoic acid [COOH(CH$_2$)$_{15}$SH] leads to hydrophilic cantilevers. Functionalization protocol starts with the deposition of a chromium layer (~1.3 nm) on the silicon cantilever, followed by evaporation of a 15-nm-thick layer of gold. After the evaporation, the cantilever is rinsed in pure ethanol. The estimated thickness of the thiol layer is 21.14 and 19.05 Å for the hydrophobic and hydrophilic layers, respectively [15].

Coarse approach of the SNOM probe toward the cantilever surface is performed with the linear motor. When an interaction is detected, fine approach is performed with the AFM piezotube, recording the AFM and SNOM signals. Both directions are recorded to obtain the complete approach/withdraw cycle. Furthermore, resonance curves are taken for the SNOM tip at several points during the cycle to study the behavior of the fiber tip mechanical resonance during the interaction of the SNOM tip with the cantilever surface. All approaches are performed onto the flexible part of the cantilever with an initial tip oscillation amplitude of 4 nm, except when specified differently.

IV. RESULTS

The tip-to-sample interaction depends on several parameters, such as the tip excitation amplitude, the chemical nature of tip and cantilever, or the tip geometry. The forces have been measured as a function of these parameters; the results are presented and discussed below.

Fig. 2(a) and (a$'$) shows a typical example of the SNOM and AFM curves. Concerning the SNOM curve [Fig. 2(a)], there are three different regions in the approach: a first region of free oscillation, in which the tip vibrates at its initial excitation amplitude; a very sharp transition zone, where
damping takes place; and, finally, a contact zone. Similarly, a typical AFM curve [Fig. 2(a)] presents a steady region, in which the cantilever is at rest; an attraction region, in which the cantilever is suddenly attracted by the tip; and a contact region, in which tip and cantilever are stuck together before being separated at large withdraw distances. Usually, the approach/withdraw cycle presents hysteresis: only in perfectly elastic processes adhesion force is equal in magnitude to the initial attraction. In the situation presented here, very sharp damping of the lateral tip oscillation is due to contact with the cantilever: an attraction force of about 1 nN is observed between tip and cantilever, and complete damping is instantaneously observed in the lateral tip oscillation due to static friction force. The same effect is observed in the withdraw part of the curves. The tip, released from the cantilever, instantaneously vibrates with an amplitude corresponding to free oscillation.

A. Influence of the SNOM Tip Geometry

The effect of the tip geometry is studied by recording approach curves on a bare Si-cantilever, with two different SNOM tips. Fig. 2 displays the results: (a) and (b) are SNOM signals for a standard and broken tip, respectively, and (a’) and (b’) display the corresponding AFM signals. The “standard” tip is a conical tip, with complete Al coating showing an optical aperture of about 100 nm. Fig. 3 shows a scanning electron microscope image of this tip (profile and cross section). The “broken” tip has been mechanically damaged; the cone is broken, leading to a larger surface of the tip apex for an optical aperture diameter of about 600 nm. In the case of a broken tip [Fig. 2(b) and (b’)], a larger attraction force of 7 nN is observed, which is attributed to the larger area (thus, the larger amount of surface charges) characterizing the broken tip. The presence of a small kink is attributed to a double-layer repulsive force that is superimposed onto the attractive coulombian force in the presence of a water meniscus. This meniscus is formed when the tip comes in contact with the cantilever, and is due to water/hydrocarbon contamination of tip and cantilever surfaces in ambient. It has a thickness of approximately 6 nm [4]. As for the standard tip discussed above, the contact between tip and cantilever surface results through horizontal friction force in complete damping of the lateral vibration of the tip. Looking at the withdraw part of the cycle, one sees that for the broken tip, the tip starts to vibrate when still in contact with the cantilever. The vibration amplitude regularly increases, corresponding to dynamic friction of the tip slipping over the cantilever surface in the meniscus. At the inflexion point marked with an arrow in Fig. 2(b), the tip is released from the cantilever, as can be deduced from the corresponding point in Fig. 2(b’). The remaining part of the withdraw curve shows that the oscillation amplitude increases with a smaller slope, while the vertical attraction force is reduced from about 11 nN back to zero over a tip-to-sample distance of 130 nm. Note for comparison that in the case of a standard tip, the attractive force at the release point is 17 nN, and that the return to free tip oscillation occurs instantaneously. This behavior, independent of the initial tip oscillation amplitude, is related to the combination of the repulsive double-layer contribution to the electrostatic force and the attractive capillary force, in the presence of a larger water meniscus in case of the broken tip. This combination leads altogether to a smaller resultant attractive force observed in the release process, as well as to the more gradual return to free tip vibration and cantilever equilibrium position when the tip is released from the cantilever.

B. Influence of the Tip Nature

As seen above, electrostatic charges play a crucial role in the interaction forces between the SNOM tip and the
cantilever surface. Their influence is further illustrated by the comparative analysis of several measurements, performed with bare SiO$_2$ as well as Al-coated tips, onto bare Si cantilevers. SNOM and AFM approach curves for a bare glass tip are shown in Fig. 4(a) and (a'). These curves must be compared with results displayed in Fig. 2(a) and (a'), corresponding to identical parameters but recorded with an Al-coated tip. In both cases, the sample is an Si cantilever surface.

The SNOM curves show that while the damping of the Al-coated tip vibration is very sharp [Fig. 2(a)], the situation in which the bare tip is used [Fig. 4(a)] presents a large transition region (corresponding to a total vertical bending force on the cantilever of approximately 27 nN) between vibration and complete damping. In the case of a bare glass tip, the lateral vibration is partially damped when the cantilever comes in contact with the tip, but the tip apex still slips over the cantilever surface. This dynamic friction is again observed in the withdraw part of the curve. Free oscillation is occurring when the tip is released from the cantilever, with tip vibration occurring when the attraction force is 5 nN. Note that this force is much smaller than the 17 nN observed in Fig. 2(a') for a standard Al-coated tip. Analysis of the mechanical resonance curve of the bare tip, displayed in Fig. 4(b), shows, however, a small regular shift toward higher resonance frequency when pressing the tip onto the cantilever surface. In pure dynamic friction, no shift should be observed. A possible explanation could be that the tip is locally stuck during the approach due to surface roughness. The plateau observed in the withdraw part of Fig. 4(a) is attributed to a topographical damage of the tip, resulting from excessive pressure applied in the approach. The slightly different free tip oscillation...
tation amplitude recorded at the very end of the cycle indicates that the damage is permanent. Another possible explanation for this plateau has been recently given by Brunner et al. [16]. They attribute this behavior to the meniscus formation between tip and sample. Taking into account that aluminum and silicon have similar attraction to water, one concludes that the observed differences between Al-coated and bare tips are not related to cantilever contamination, but are rather due to the larger density of electrostatic charges characterizing the Al-coated tip, leading to more important coulombian forces.

In the situations considered above, the flexibility of the cantilever results in contact with the tip when an attractive force is applied. To study the shear-force mechanism in absence of this attraction, the same experiment is repeated approaching the tip onto the fixed part of the cantilever. The results for tip vibration amplitude are displayed in Fig. 5 for an Al-coated tip [Fig. 5(a)] as well as for a bare fiber tip [Fig. 5(b)]. Again, one sees that the transition zones are wider for a bare fiber, as expected from the less efficient damping of reduced surface charges. In the approach part of the curve, a transition zone of 20 nm is observed for Al-coated tip, as opposed to 80 nm for a bare fiber. A similar conclusion is drawn from the analysis of the transition zone in the withdraw part of the curves, where the respective widths are 80 nm for the Al-coated tip and 170 nm for the bare fiber. Comparing Fig. 5(a) with Fig. 2(a) (for an Al-coated tip), and comparing Fig. 5(b) with Fig. 4(a) (for a bare glass tip), one concludes that the cantilever flexibility does not influence the nature of the shear-force mechanism. It simply reduces the width of the transition zones by allowing damping through friction forces at an earlier stage in the approach, since contact occurs earlier.

C. Influence of the Tip Oscillation Amplitude

Fig. 6 shows the SNOM and AFM approach curves for an Al-coated SNOM tip onto an Si-cantilever when the tip is excited at its resonant frequency for three initial oscillation amplitudes, corresponding to 2, 20, and 200 nm, respectively. From the SNOM signals [Fig. 6(a)–(c)], it is clear that as the tip excitation amplitude increases, the width of the transition zone from free oscillation to complete damping increases as well. For small oscillation amplitudes, complete damping of the vibration occurs very abruptly within a few nanometers and is due to static friction as discussed before. For larger excitation amplitudes, the damping of the oscillation takes place in several steps, over a large range of distances. In Fig. 6(c), for instance, damping from free oscillation to complete stop occurs over almost 400 nm, and the tip is already in contact with the sample. Simultaneous analysis of the AFM signals [Fig. 6(a)–(c)] shows that at medium excitation amplitude [Fig. 6(b) and (b)], the damping in the approach part of the curve is due to static friction, while in the withdraw part of the curve, dynamic friction is present. At large excitation amplitude, dynamic friction is observed in both transition zones, and irreversible damage is again induced on the tip. Note that in the approach part of all three curves, the vertical force responsible for the tip attraction is \(4\,\text{nN}\), independent of the tip excitation, as expected from its electrostatic nature.

These observations confirm that at large excitation amplitudes, the tip apex sweeps the cantilever surface over a much longer span. The energy stored in the tip is larger, thus, a larger vertical force must be applied to completely stop the lateral vibration of the tip [20 nN in Fig. 6(c) as opposed to 5 nN in Fig. 6(a)]. For smaller amplitudes, the role of the cantilever roughness is more important, smaller bumps being sufficient to completely stop the lateral vibration of the tip and make it stick in the cantilever surface. Another important element in the analysis of the tip movement is the water meniscus that is formed between the tip and the cantilever in ambient environment. A small excitation amplitude allows the formation of a stable meniscus, and the tip oscillates in water/hydrocarbons. Therefore, the friction force \(F_f\) slows down the tip, explaining the almost vertical transition observed for the SNOM signal in the case of Fig. 6(a) between free and totally damped oscillation. The relatively large bending force observed in the withdraw part of the AFM curve (24 nN for a small excitation amplitude of 2 nm, compared to 15 nN for 200-nm excitation amplitude), confirms this behavior. For a small tip lateral motion, the tip is trapped in the water meniscus by the vertical component of the capillary force. However, when the excitation amplitude is large, the meniscus is disturbed and can even be destroyed due to the large traveling distance of the tip over the cantilever. As a consequence, capillary forces in the
meniscus can no longer retain the tip, which is released from the cantilever. Both the SNOM and the AFM approach curves thus evidence the crucial role of the electrostatic force, the presence of a contamination layer (water), and the existence of a capillary force that retains, to some degree (which depends on the tip initial excitation amplitude), the tip on the cantilever.

D. Influence on the Cantilever Chemical Treatment

The chemical affinity of the cantilever surface has been modified by thiol treatment of the Si surface to study the interaction with different types of contamination layers. The SNOM probe is an Al-coated tip. The approach curves as well as the resonance frequency curves are shown in Fig. 7, in which (a) and (a’) correspond to the SNOM and the AFM approach curves with the hydrophobic cantilever. The corresponding curves for a hydrophilic cantilever are labeled (b) and (b’). To control the mechanical behavior, resonance frequency curves have been simultaneously recorded at several decreasing oscillation amplitudes. Fig. 7(c) displays mechanical resonance curves recorded with the hydrophobic cantilever, while Fig. 7(c’) displays curves recorded with the hydrophilic cantilever.

Both SNOM curves present the same initial decrease of the oscillation amplitude due to the coulombian and friction forces, as predicted by the theory. However, the transition region to a complete damping of the oscillation is very broad and covers several tens of nN. A possible explanation for the lack of sharp transition could come from the presence, in both hydrophobic and hydrophilic cases, of a thiol layer in which the tip can slip much more easily than in the water contamination layer characterizing all the experimental situations pre-

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Fig. 4. SNOM (a) and AFM (a’) approach curves for a bare SiO₂ tip. (b) Tip resonance frequency curves recorded at reduced oscillation amplitudes. The Greek labels identify the corresponding recording points.
Fig. 5. SNOM approach curves on the fixed part of the cantilever.
(a) Al-coated tip. (b) Bare tip.

The tip vibration amplitude for the case of hydrophobic cantilever decreases continuously to a complete stop, while in the hydrophilic situation it first reaches a plateau before being completely damped. The presence of this plateau is attributed to the double-layer repulsive force present in the case of a hydrophilic cantilever, characterized by the formation of a large meniscus. The combined effect of the reduced attractive coulombian force and the large meniscus allows observing the double-layer force. The analysis of the resonance curves shows that in the hydrophilic situation, a shift toward higher frequency is observed, confirming that static friction is responsible for damping of the tip vibration. In the case of a hydrophilic cantilever, no frequency shift is observed, dynamic friction related to electrostatic and capillary forces in the meniscus being the major source of damping.

Analysis of the withdraw part of the curves confirms this conclusion: a smaller vertical force is measured at the release point for the hydrophilic case. Furthermore, the effect of the capillary forces present in the hydrophilic situation is expressed by the difference observed in the withdraw transition zone of the SNOM curves: in the hydrophobic situation, the amplitude of the tip vibration increases as soon as pressure is released, while in the presence of water, attractive capillary forces result in some static friction.

Note that when the same experiment is performed onto a bare Si-cantilever on one hand, and onto an Au-coated Si-cantilever on the other hand, the same decrease in the electrostatic force is observed for the more hydrophilic surface (Si). Furthermore, the analysis of the shift in the resonance frequency of the tip confirms the static friction characterizing the hydrophobic surface, and the presence of an intermediate situation where dynamic friction is observed for the more hydrophilic surface.

V. CONCLUSION

This work allows a better understanding of the forces acting on the SNOM tip in interaction with a flat sample. It shows that coulombian forces are the most important ones to initiate the damping of the lateral oscillation of the SNOM probe. When the tip approaches the sample surface, it normally comes in contact with a water contamination layer, except in situations where a hydrophobic treatment has been performed. When water is present, a meniscus is formed and capillary forces come into play. Furthermore, coulombian forces are reduced, and a double-layer repulsive contribution competes with the coulombian part of the electrostatic force. As a consequence, dynamic friction, which is related to these electrostatic and capillary forces, is responsible for the damping in the tip oscillation. For the case of a hydrophobic sample, static friction, related to the coulombian force, is the principal cause of the decrease in the tip amplitude. These results are confirmed by studying the resonance curves of consecutive decreasing oscillation amplitudes in both situations. No frequency shift is found in the hydrophilic situation while a shift toward the increasing frequencies can be observed in the hydrophilic situation.

Additional studies concerning the tip geometry show that the tip diameter is a critical parameter in the tip-to-sample interaction. The main cause of damping in tips with large diameters is due to dynamic friction, related to the combination of double-layer repulsive forces and attractive capillary forces. On the other hand, for standard tips, the meniscus formed is smaller and the damping is mainly due to static friction.

Tip nature also plays a role in the forces responsible for vibration damping. Thus, the vibration of bare glass tips approaching the sample is damped by dynamic friction and goes through a long transition region from free oscillation to complete stop. This is not the case for Al-coated tips, which have a sharp transition, and damping is solely due to static friction.

It would be interesting to repeat the same experiments in controlled conditions (clean room). As we have shown for hydrophobic surfaces, we expect that when working in this environment, the predominant force in the amplitude damping will be friction force.
Fig. 6. SNOM and AFM approach curves for different excitation amplitudes of the fiber tip. (a) and (a') 2 nm, (b) and (b') 20 nm, (c) and (c') 200 nm.
Fig. 7. SNOM (a) and AFM (a') approach curves for the case of a hydrophobic cantilever. SNOM (b) and AFM (b') approach curves when using a hydrophilic cantilever. The initial tip oscillation amplitude is 20 nm. The corresponding resonance frequency curves recorded at reduced oscillation amplitude are displayed for the hydrophobic (c), respectively hydrophilic (c') cantilevers.

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