Deformation, Contact Time, and Phase Contrast in Tapping Mode Scanning Force Microscopy

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The general features of tapping mode operation of a scanning force microscope are presented. Relevant factors of tapping mode such as forces, deformation, and contact times can be calculated as functions of tapping frequency, amplitude damping, and sample elastic and viscoelastic properties. Typical contact times per oscillation are about 10^{-7} s for hard samples and 6×10^{-7} s for soft materials, i.e., between one and two orders of magnitude smaller than their equivalents in contact mode force microscopy. The model proposed allows the determination of the phase lag between excitation signal and cantilever response. Major factors to phase contrast are viscoelastic properties and adhesion forces with little participation from elastic properties. Experiments performed on droplets of glycerin deposited on graphite illustrate the ability to image them by recording phase changes.

1. Introduction

The tapping mode operation of a scanning force microscope represents one of the latest developments in scanning probe microscopies.[1−3] In this mode, the cantilever−tip ensemble is oscillated at a frequency near its resonance. The equilibrium separation between tip and sample is smaller than the oscillation amplitude; as a consequence the tip strikes the sample once each cycle. Large amplitudes, up to 100 nm, provide the cantilever with enough energy to overcome adhesion forces. Damage to the sample is reduced with respect to contact mode scanning force microscopy (SFM) because lateral and shear forces are smaller.

In the last two years this mode has found a variety of applications, in particular, for imaging samples such as Langmuir−Blodgett films,[4] polymers,[5] and biomolecules.[6−9] Recently, few theoretical models and calculations have been proposed to describe the operation of tapping mode SFM.[10−12] Some of these works consider the cantilever as a nonlinear driven oscillator. This assumption allows calculation of the relevant physical parameters that control its operation and, in some cases, direct experimental comparison.[13]

The calculations have mainly been applied to determine the dependence of the force on tip−sample distance and driven frequency (it will be called tapping frequency hereafter). The results show some unexpected complexity of tapping operation. For instance, it is deduced that the force on the sample has a maximum with respect to tip−sample distance.[10] Spatz et al.[11] have demonstrated the existence of a relationship between the tapping frequency and the applied force and the asymmetry of this with respect to the resonant frequency of the free cantilever. Operating the instrument at frequencies lower than the noncontact cantilever resonant frequency minimizes the applied force.

These works have established some of the foundations for a dynamic description of tapping operation; however, they have been restricted to the calculation of a few experimental parameters such as the force. There are many other parameters such as deformation, contact time, and their dependencies on tapping frequency, amplitude damping, and elastic or viscoelastic properties of the sample that have not been addressed. Moreover, the measurement of the phase lag between the cantilever movement and the external force acting on it has been suggested[14] as a way to obtain chemical contrast. However, a detailed explanation of the contrast mechanism has not yet been provided.

Here, we describe the general features of tapping operation. We propose a model to calculate the dependence of sample deformation on cantilever−sample distance, tapping frequency, and sample mechanical properties. We also study the influence of those parameters in the time the tip remains in contact with the sample.

Finally, we examine possible sources of phase contrast such as viscoelasticity and adhesion forces. Experiments performed on glycerin droplets deposited on graphite illustrate the ability to image them by recording the phase variations when the tip goes from the substrate to the liquid.

2. Model

2.1. Equation of Motion. Tapping mode SFM operation can be simulated as a nonlinear driven oscillator with damping. Then, the movement of the cantilever is governed by the following equation:

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$$m\frac{d^2z}{dt^2} = -k_c z - \frac{m w_0}{Q}\frac{dz}{dt} + F_0 \cos(wt) + F(z_c,z) \qquad (1)$$

k_c, w_0, and Q are the elastic constant, resonant frequency ($\omega_0 = 2\pi f_0$), and quality factor of the free cantilever, respectively. The sinusoidal term is the excitation signal applied to the cantilever. $F(z_c,z)$ is the tip−sample interaction, where z_c is the tip−sample distance when the cantilever is at its equilibrium position (Figure 1). The resistive force (damping) is considered to have only terms proportional to the velocity.[15]

In tapping mode rigid cantilevers ($k_c = 20-50$ N/m) and large amplitudes (50−100 nm) are common. Those values give to the cantilever enough energy to avoid being trapped by attractive forces. Large amplitudes imply that the tip experiences different force laws during an oscillation. As a consequence, the above equation has to be solved numerically.

The movement of the cantilever in the tapping operation goes through noncontact and contact tip−sample conditions. These situations are separated by the interatomic distance a_0. For distances larger than a_0 the tip−sample interaction is calculated through the van der Waals force between a sphere and a flat surface.[16] For distances smaller than a_0, the repulsive force between the tip and the sample is simulated by the indentation force derived from Hertz's model.[16]

$$F(z_c,z) = -\frac{AR}{6(z_c+z)^2} \qquad z_c + z > a_0 \quad (2a)$$

$$F(z_c,z) = -\frac{AR}{6a_0^2} + \frac{4E\sqrt{R}}{3-3\nu^2}(a_0 - z - z_c)^{3/2} \qquad z_c + z \le a_0 \quad (2b)$$

R is tip radius, A is Hamaker's constant, and E and ν are the Young's modulus and Poisson's coefficient of the sample, respectively.

In this model, if a spherical and nondeformable tip is assumed, then the total tip−sample force is relaxed in the deformation of the sample. This can be considered as a good approximation for a wide variety of samples. Tips are made of hard materials such as silicon and silicon nitride, whose Young's moduli are 130 and 144 GPa, respectively.

2.2. Viscoelasticity. Tapping mode SFM is being used to image samples such as Langmuir−Blodgett films, cell membranes, biomolecules, and some polymers that could be called soft materials ($E \le 10$ GPa). Here, viscosity is supposed to play an important role in their deformation. As a first approximation, we model the dynamic response of those samples by a linear time-dependent model.[17]

$$\sigma = E\epsilon + \eta\frac{d\epsilon}{dt} \qquad (3)$$

σ, ϵ, and η are the stress, strain, and viscosity, respectively. The ratio between Young's modulus and the viscosity coefficient defines a relaxation frequency, ω_v, that would be useful to explain the response of these samples to external forces.

It is assumed that the viscosity force is proportional to the deformation velocity and contact area. Then, the interaction force in the contact region is

$$F(z_c,z) = -\frac{AR}{6a_0^2} + \frac{4E\sqrt{R}}{3-3\nu^2}(a_0 - z - z_c)^{3/2} - \frac{\pi\eta R}{h}(a_0 - z - z_c)\frac{dz}{dt} \qquad z_c + z \le a_0 \quad (4)$$

where h is the thickness of the sample.

2.3. Parameters. In the text we assume the following values unless otherwise stated: cantilever constant $k_c = 20$ N/m; cantilever free resonant frequency $f_0 = 200$ KHz; quality factor $Q = 500$; free oscillation amplitude 100 nm; tip's curvature radius $R = 20$ nm; Poisson's coefficient $\nu = 0.3$; Hamaker's constant $A = 10^{-19}$ J. Those parameters are representative of many experimental situations. The values of the viscosity used below $\eta = 30$ and 400 Pa s describe several polymers and biomolecules.[18] For those samples a value $h = 10$ nm has been used. Young moduli of $E = 0.1$, 0.3, and 70 GPa have been used. As a reference, the Young's moduli of some cell membranes have been estimated within the 0.1−6 GPa range.[19] For most of the calculations we have chosen as tapping frequency the resonant frequency of the cantilever.

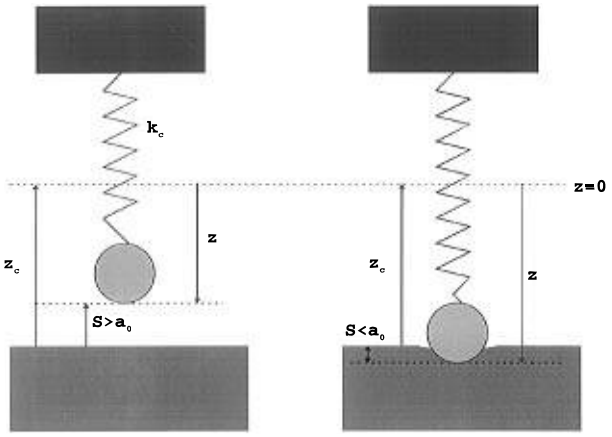

Figure 1. Schematic model of cantilever motion in tapping mode. The cantilever is represented by a spring of constant k_c, z is the tip displacement with respect to its equilibrium position, and z_c is the tip−sample separation at equilibrium.

3. Damped Amplitude

The damped amplitude is the sum of z_c and the sample deformation. The dependence of the damped amplitude on separation is shown in Figure 2a. The shape of the curves illustrates a nonlinear behavior. This nonlinearity is more noticeable for soft samples, and it is responsible for the dependence of the force on separation. As expected, less damping is obtained with soft samples. For hard samples, the curve has a slope close to 1. Here sample deformation is much smaller than z_c; as a consequence, the damped amplitude can be approximated as the tip−sample separation at equilibrium. Experiments performed on silicon wafers (hard sample) and copolymer films (soft sample) are in agreement with above results.[20]

Figure 2b shows the dependence of the damped amplitude on tapping frequency. The presence of repulsive forces during contact and their associated negative force gradients shifts the resonance to higher frequencies and breaks the symmetry of the curves. The shift increases with the stiffness of the sample because the slope of the repulsive interaction is higher. Similarly, the resonant frequency displacement depends on tip−sample distance, increasing as z_c diminishes. These results are in agreement with those of Spatz et al.[11]

4. Deformation and Force

Sample deformation is one of the parameters that better reflects the influence of mechanical properties on tapping operation as well as the most significant parameter for evaluation of sample damage. The curve's shape reflects the existence of a maximum with respect to z_c (Figure 3). Qualitatively it can be explained as follows: the deforma-

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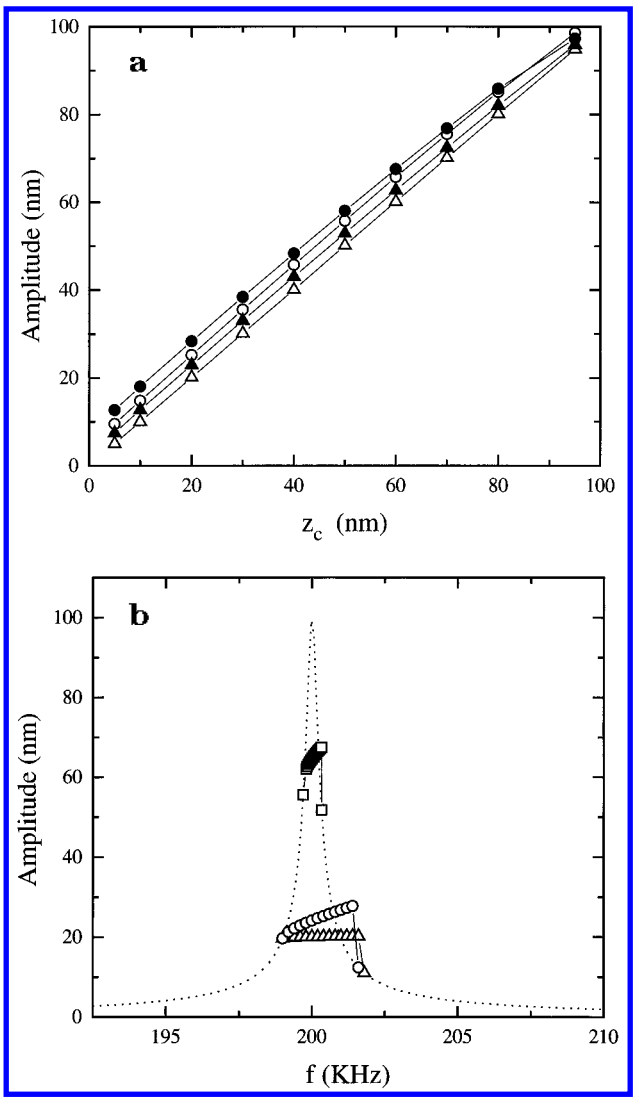

Figure 2. Damped amplitude dependence with respect to equilibrium separation z_c (a) and tapping frequency (b). E = 70 GPa (open triangles) and E = 0.3 GPa (open circles). Viscoelastic samples are characterized by E = 0.1 GPa and η = 30 Pa·s (solid circles) and by E = 0.1 GPa and a viscosity coefficient of 400 Pa·s (solid triangles). (b) The damped amplitude is represented for a soft material (E = 0.3 GPa) and tip−sample separations z_c = 20 nm and 60 nm (open circles and open squares, respectively) and for a hard material (E = 70 GPa) and z_c = 20 nm (open triangles). The dotted line represents the cantilever free amplitude.

tion can be estimated by subtracting the equilibrium distance z_c from the damped amplitude. The bow showed by the amplitude (Figure 2a) produces a maximum in the deformation with respect to tip−sample separation. In particular the shapes obtained for viscoelastic samples are similar to the experimental results found with copolymer films.[20]

Viscoelastic materials attenuate the deformation induced by the strokes of the tip. For instance, the deformation for a viscous sample (E = 0.1 GPa and η = 400 Pa s) is a factor of 2 smaller than that for an elastic one (E = 0.3 Gpa). As a consequence, there is an effective increase of stiffness with frequency for those materials, whenever tapping frequencies are higher than the relaxation frequency of the material. In fact there is experimental evidence of this effect when imaging cell membranes.[6] These results emphasize the suitability of tapping operation with respect to contact mode to image soft samples such as cell membranes or some polymers.

Other calculations (not shown here) indicate a substantial decrease in deformation when the cantilever force constant and free amplitude are decreased. However, the parameter's choice has a lower limit. For small amplitudes and/or soft cantilevers the van der Waals force may dominate the behavior of the cantilever. Then the equation of motion can have solutions other than the above one. The cantilever may get trapped by the sample, have a chaotic motion, or oscillate without touching the sample.

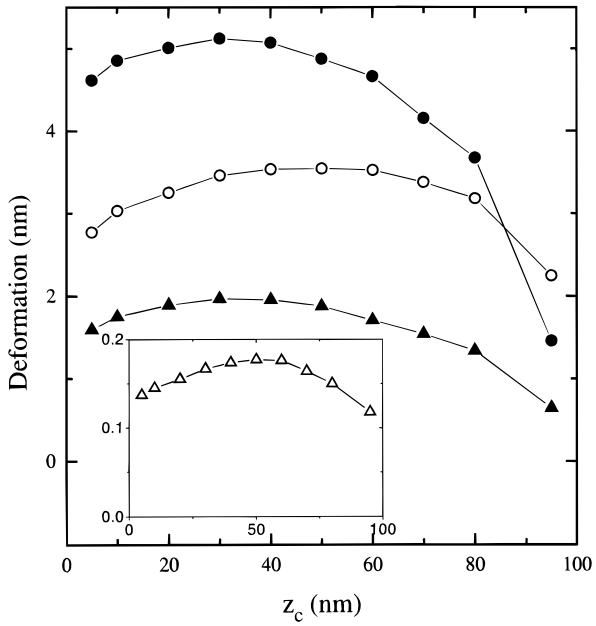

Figure 3. Average sample deformation dependence with tip−sample separation for different samples. Inset curve for E = 70 GPa. Symbols as in Figure 2.

The values obtained outline the relevance of sample deformation to influence lateral resolution. Previous models to calculate lateral resolution in scanning force microscopy have considered nondeformable samples.[21,22] But this approach may not be suitable to describe the behavior of soft materials.[23−25]

Typical values for the forces exerted on the sample are within the 10−40 nN range (Figure 4). The force is averaged over the contact time, and it has been calculated considering only elastic deformations. Peak force values may be higher. The average force presents a maximum with respect to the tip−sample equilibrium position. This maximum has its origin in the bowlike shape of the amplitude curve (Figure 2a). Additionally, the figure reveals that the force exerted on the sample cannot be calculated as the cantilever constant multiplied by the amplitude reduction. For instance, for a free amplitude of 100 nm and a damped amplitude of 82 nm the force estimated by Hook's law would be 360 nN (k_c = 20 N/m), while the value obtained from the calculations is 10 nN (E = 0.3 GPa). These results emphasize the dynamic aspects inherent to tapping operation.

5. Contact Time

The time that the tip is interacting repulsively with the sample is called the contact time, t_c. This time is a useful quantity to understand the behavior of tapping operation

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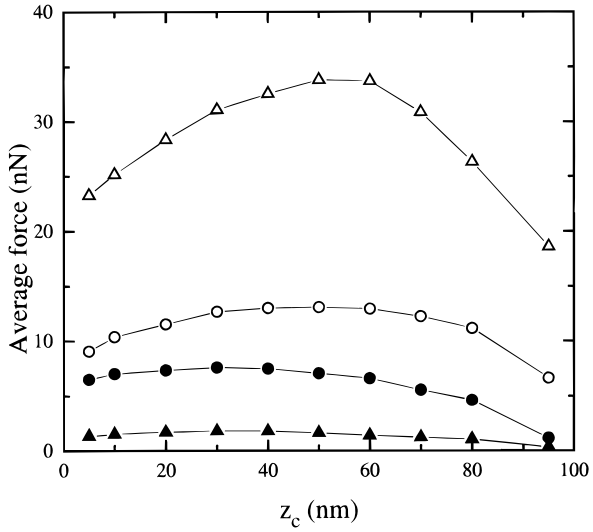

Figure 4. Average elastic force on the sample as a function of tip−sample equilibrium separation z_c. Symbols as in Figure 2.

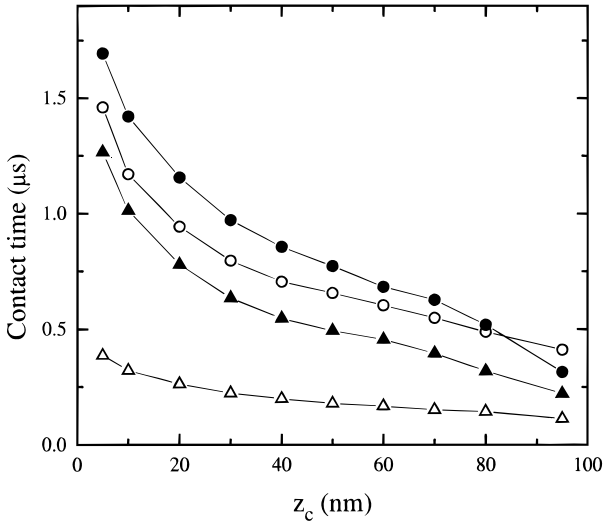

Figure 5. Contact time dependence with tip−sample equilibrium separation. Symbols as in Figure 2.

as well as to estimate the amount of damage done to the sample.

Figure 5 shows the dependence of contact time on tip−sample separation for different samples. As could be predicted, t_c increases with tip−sample proximity. For small separations relative to the free amplitude, there is a steep increase of t_c. Here, there is contact during almost all the oscillation cycle.

There is also a noticeable dependence of t_c on Young modulus. For purely elastic materials a difference in the Young's modulus of about 20 produces contact times delayed by a factor of four. Softer samples have higher t_c values. As a rule, we have found that for elastic materials the product of the force times t_c is almost constant and independent of the material. Then forces and contact times are inversely proportional.

The increase of t_c with tapping frequency is more puzzling (Figure 6). This is almost linear above the resonant frequency of the free cantilever. This asymmetry with respect to resonance could be related to the asymmetry shown by the damped amplitude (Figure 2b). In addition to smaller sample deformation,[13] operating the instrument at tapping frequencies below f_0 produces shorter contact times.

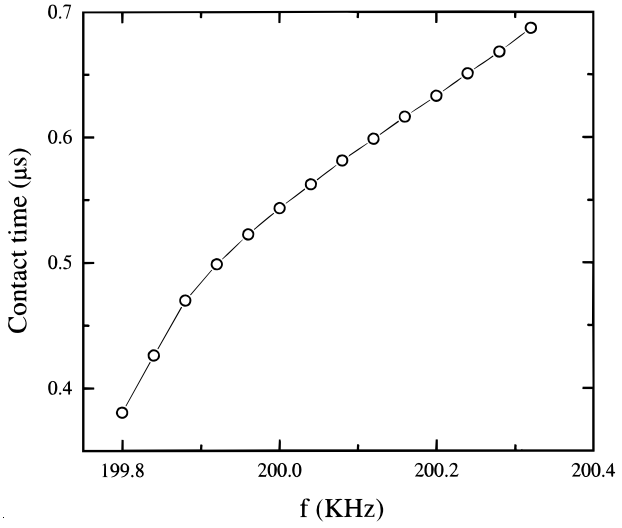

Figure 6. Contact time dependence with tapping frequency for a soft elastic sample (E = 0.3 GPa); z_c = 60 nm.

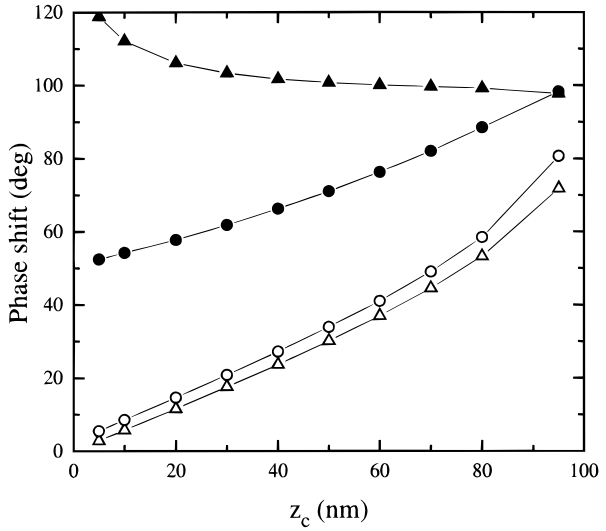

Figure 7. Phase lag between external excitation and cantilever oscillation as a function of z_c for different samples (symbols as in Figure 2).

Ratios between contact times and oscillation periods are about 0.02 and 0.12 for hard and soft elastic materials, respectively (z_c = 80 nm). The t_c determined above supports the observation that tapping operation is gentler on the sample than contact SFM. For instance, for an hypothetical feature on a hard sample (E = 70 GPa) of 4 nm scanned at 1 μm/s, t_c in contact SFM is 4×10^{-3} s while in tapping mode the accumulated t_c on the feature will be about 8×10^{-5} s, i.e., almost two orders of magnitude smaller. As a consequence the friction energy can be estimated to be about two orders of magnitude smaller in tapping operation.

6. Phase Contrast

The phase lag between the excitation signal and cantilever response is a parameter that contains relevant information about the type of interactions that the cantilever−tip system experiences. Figure 7 presents the oscillation phase shift with respect to tip−sample distance z_c for several materials. For elastic materials the phase decreases approximately, from 90° (tapping frequency here is close to resonance) to 0°, as the cantilever approaches the sample. This is a consequence of the resonant frequency displacement of the cantilever to higher fre-

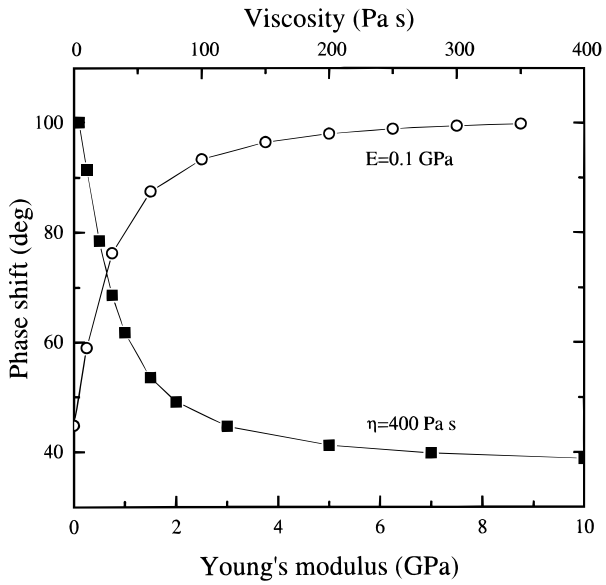

Figure 8. Effect of viscosity on the phase shift of cantilever oscillation. $z_c = 60$ nm. The relationship between tapping and relaxation frequencies ($\omega_v = \eta/E$) explains the asymptotic limits. For $\omega \ll \omega_v$ the phase shift is dominated by viscoelastic properties while for $\omega \gg \omega_v$ it is dominated by the resonant frequency changes.

quencies due to tip−sample contact. The shift is smaller for softer samples because the frequency change is smaller.

Viscoelastic materials have a more complex phase dependence. There are two contributions to the phase. One comes from the displacement of the resonant frequency to higher values (Figure 2b), while the other is due to the viscous response of the sample. For large tip−sample separations (relative to the free amplitude), phase shifts above 90° are possible. The proximity of the sample may reduce the shift for low viscosities, but it never reaches 0°. For a given z_c, the higher the viscosity, the higher the phase shift. For the sample with $\eta = 400$ Pa s the phase shift is dominated by the viscous response of the sample.

Figure 8 summarizes the dependence of the phase on viscoelasticity. The phase shift increases as the sample viscosity increases and the elastic modulus decreases. For high viscosities, the phase shift contribution from viscoelasticity increases the total phase shift above 90°. However, this phase shift is absent for samples where the elastic response dominates over the viscous one. Therefore, viscoelastic properties can be a major or a dominant source to obtain phase contrast. In the slightly different context of force modulation imaging, the influence of viscosity on phase shifts was also pointed out.[26]

We have also studied the influence of adhesion forces on phase shifts. The forces have been modified by changing the value of the Hamaker constant. Under reasonable experimental conditions, a change of 5 nN in the adhesion force can produce a phase shift of about 2°. A value well within the range of sensitivity of most instruments. This result suggests that variations of the capillary forces in samples with regions with different hydrophilic/hidrophobic properties could also be a source of phase contrast.

Both results stress phase measurement as a means to achieve contrast in heterogeneous samples. On the other hand, calculations performed with purely elastic samples show a phase shift of 5° between soft and hard materials (ratio of 200 between elastic properties). However, this shift is reduced to 0.1° when the tapping operation is run

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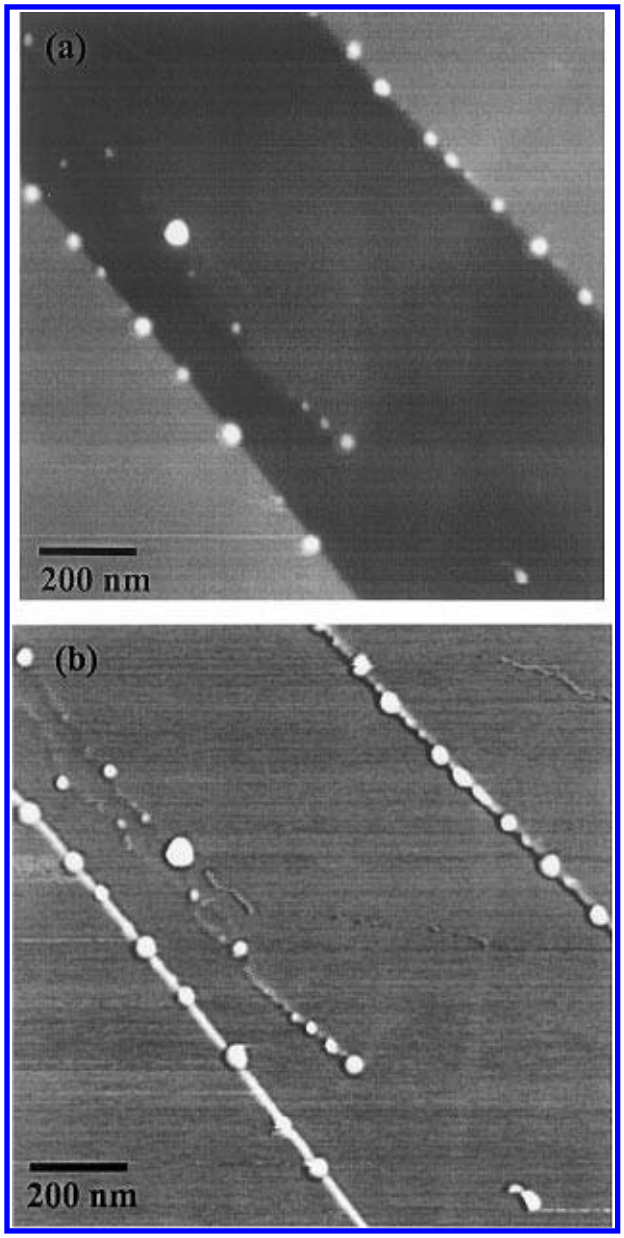

Figure 9. Tapping mode image of glycerin droplets on graphite: (a) topography; (b) phase contrast image; independent on their size all the droplets have the same shift with respect to the substrate (40°). Image size 1.3 μm × 1.3 μm. Tapping mode data: tapping frequency, 350 KHz; free amplitude, 27 nm; amplitude reduction, 5 nm.

at constant amplitude damping. For practical purposes, the elastic component to phase contrast may be considered negligible.

To illustrate the potential of phase contrast imaging and to confirm qualitatively some of the above results, we have performed experiments with glycerin ($C_3H_5(OH)_3$) deposited on highly oriented pyrolitic graphite. A 20 μL drop of glycerin was deposited at room temperature on the substrate for 30 s. Then it was removed from the surface with filter paper. This process leaves nanometer-size droplets on the surface. The observed diameters range from 20 to 60 nm. The apparent height goes from 1.5 to 10 nm. The drops are preferentially located along ridges and steps of the substrate (Figure 9). They form patterns similar to those observed with droplets of KOH water solutions.[27] We have never succeeded in imaging the droplets by contact mode SFM. Most likely the lateral

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forces drag them during imaging. However, the substantial reduction of lateral forces in tapping mode and the viscoelastic properties of nanometer size droplets[28,29] allow their imaging by tapping mode.

The phase contrast image (Figure 9b) shows that topographic changes are not involved in phase contrast. In the image, graphite terraces are separated by 2.4 nm steps, but the phase values on them are identical. When the tip encounters the step from below, the twisting of the cantilever gives rise to a phase shift, as can be observed along the first step on left, but this effect disappears when the scanning direction is changed. Although quantitative confirmation of the theoretical results is hard to obtain at present, partly because of the difficulty in knowing the mechanical properties of nanometer-size glycerin droplets adsorbed on solid supports, the phase shift of the droplets is higher by 40° than that of the bare and more rigid substrate, as the model has predicted. Furthermore, the phase shift is independent of the droplet's size (between 20 and 60 nm diameter), which again emphasizes that contrast is not related to topography.

7. Conclusions

Tapping operation of a scanning force microscope can be considered as a combination of noncontact and contact SFM. As such, it has significant differences with respect to the operation of SFM in the contact mode. The force applied on the sample is easily controlled and calculated in contact SFM by measuring the cantilever's deflection, but in tapping operation there is not an analytical expression to calculate the force. Furthermore, the normal force exerted on the sample can no longer be estimated as the product of the cantilever constant and the amplitude reduction. Several parameters determine the force applied to the sample: free amplitude, amplitude damping, tip−sample separation, tapping frequency, and cantilever and sample mechanical properties. This multifactor dependence outlines the dynamic aspects inherent to tapping operation.

Contact times depend on the sample's mechanical properties. They are between one and two orders of magnitude smaller than those in contact SFM; this in turn minimizes the dragging of the sample by the tip. This supports the experimental observation that tapping produces a gentler treatment on the sample in spite of the fact that normal forces may be comparable. Tapping operation provides another means of minimizing sample damage with viscoelastic materials. This happens whenever the tapping frequency is above the relaxation viscoelastic frequency of the sample.

Changes in adhesion forces and viscoelasticity may be a source of chemical contrast when phase measurements are recorded. This could be the major source of imaging contrast with soft materials (polymers, cell membranes, liquids, or molecular films). Experiments on glycerin droplets illustrate phase contrast imaging of soft and weakly adsorbed layers on solid supports.

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