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Nonlinear Detection of Ultrasonic Vibrations in an Atomic Force Microscope

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A new method is proposed to detect ultrasonic vibration of the samples in the Atomic Force Microscope (AFM) using nonlinearity in the tip-sample interaction force curve $F(z)$. Small amplitude ultrasonic vibration less than 0.2 nm is detected as an average displacement of a cantilever. This Ultrasonic Force Mode (UFM) of operation is advantageous in detecting ultrasonic vibration with frequencies up to the GHz range, using an AFM cantilever with a resonant frequency below 100 kHz. It was found that a strong repulsive force is acting after an ultrasonic amplitude threshold of the is crossed, with the amplitude of this threshold depending upon the average force applied to the tip.

KEYWORDS: atomic force microscope, ultrasonics, nonlinear vibrations, viscoelastic properties evaluation, nonlinear atomic force microscope, ultrasonic force mode, UFM

1. Introduction

It may seem difficult to detect ultrasonic frequencies (MHz to GHz frequency range) in an atomic force microscope (AFM)[1,2] having a soft cantilever with a resonant frequency below 100 kHz. Since the dependence of the atomic force F on the distance z between the tip and surface is essentially nonlinear,[3,4] then, small displacements (down to 0.1 Å) and small forces (down to 10^{-11} N) excited by ultrasound can be measured by AFM. If one vibrates the surface of the object by an ultrasonic wave, this nonlinearity will result in a shifting of the average position of the AFM cantilever and in a generation of the higher harmonics of the ultrasonic vibration.

In order to detect the ultrasonic vibration and its harmonics special modification of the cantilever and the AFM setup must be affected. The measurement of the average position of a cantilever can be realized in all existing devices.

2. Theoretical Model of the Nonlinear Detection. Ultrasonic Force Mode (UFM)

2.1 *Dynamic model of the cantilever-object system*

To evaluate the response of the AFM to the vibration of an object it is useful to model the tip-cantilever system as an harmonic oscillator, possessing an effective mass m, spring constant k and effective dissipation γ.[2,4]

The atomic force F is applied to the tip and depends nonlinearly, as schematically illustrated in Fig. 1, upon the distance between the coordinate of the cantilever, z_c, and the instantaneous position of the surface z. The coordinate z is the summation of the AFM stage position, z_s, and the instantaneous displacement of the ultrasonic wave, $A\cos(\omega t)$, where A and ω are amplitude and frequency, respectively. If origin of the coordinates is placed at the non-deflected position of the cantilever, the system obeys the following second order nonlinear differential equation:

$$m\ddot{z}_c+\gamma\dot{z}_c+kz_c=F(z_c-z_s-A\cos\omega t)\equiv F(z). \qquad (1)$$

This equation can be integrated using linearisation of the $F(z)$ dependence[2,4] or, for the general case, solved numerically by computer simulation. However, for the two extreme cases, very low and very high vibration frequency ω, the solution can be found directly from eq. (1).

2.2 *Low vibration frequency (quasi-static) approximation*

If the frequency ω is much lower than both the resonant frequency of the cantilever, ω_0, and the relaxation frequency, ω_r:

$$\omega\ll\omega_0,\ \omega_r, \quad \text{where} \quad \omega_0=\sqrt{k/m}, \quad \omega_r=(k/\gamma), \qquad (2)$$

then the time derivatives of z_c in eq. (1) vanish and eq. (1) is reduced to

$$kz_c=F(z_c-z_s-A\cos\omega t), \qquad (3)$$

corresponding to the static situation described in the literature.[2,4–6] If the cantilever is soft compared with the force derivative (i.e. $k\ll F'(z)$) the cantilever is trapped near $F(z)\cong 0$ and the topography of the object

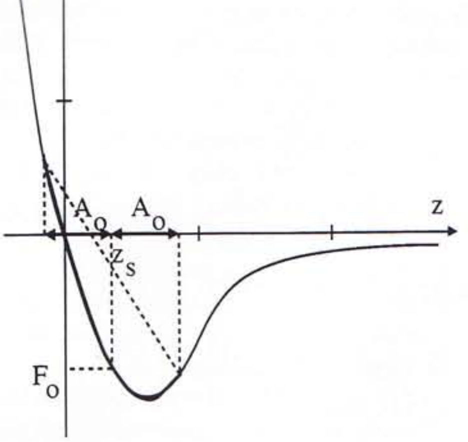

Fig. 1. Schematic illustration of the dependence of the atomic interaction force F on the tip—surface distance z (force curve).

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rather than its elasticity is measured.

2.3 *High vibration frequency (ultrasonic) approximation*

In the high frequency limit, when ω is much higher than the resonant frequency of the cantilever ω_0

$$\omega \gg \omega_0 = \sqrt{k/m}, \tag{4}$$

the inertial force defined by the finite mass of the tip and the cantilever predominates rather than the elastic restoring force of the cantilever spring. Considering that the cantilever of the AFM usually has a resonant frequency ω_0 below 100 kHz, ω has to be in the ultrasonic frequency range. We call this limit the "Ultrasonic Approximation" and this mode of the AFM operation as the "Ultrasonic Force Mode (UFM)".

The oscillation of the z coordinate of an object under ultrasonic frequency (ω in the argument of $F(z)$ in the right-hand side (r.h.s) of eq. (1)) acts as a driving force for the tip-cantilever system described by the (l.h.s) of this equation. When the difference between ω and ω_0 is large, satisfying the condition of the eq. (4), the nonlinearity of $F(z)$ results in the oscillation of cantilever coordinate z_c at the base frequency ω and its harmonics. Also there is a constant deflection of the cantilever, z_0. Therefore, we expand z_c as

$$z_c = z_0 + \sum_{n=1}^{\infty} z_n \cos n\omega t, \tag{5}$$

where z_0 is constant deflection of the cantilever and $z_1, \cdots, z_n$ are the amplitudes of the oscillations with the fundamental frequency ω and its harmonics, $n\omega$. Substituting eq. (5) into eq. (1) we have

$$\begin{aligned} -m\omega^2 \sum_{n=1}^{\infty} n^2 z_n \cos n\omega t - \gamma\omega \sum_{n=1}^{\infty} n z_n \sin n\omega t \\ + kz_0 + \sum_{n=1}^{\infty} z_n \cos n\omega t \\ = F\left(z_0 + \sum_{n=1}^{\infty} z_n \cos n\omega t - z_s - A\cos\omega t\right). \end{aligned} \tag{6}$$

The maximum of the r.h.s. of the eq. (6) should have an upper limit because the force acting between the tip and the surface cannot be infinitely large due to the finite rigidity of the object, then we can assume $|F(z)| \le F_0$, where F_0 is the maximum force. Dividing both sides of the eq. (6) by ω^2, we obtain the following relation

$$\begin{aligned} \Bigg| -m \sum_{n=1}^{\infty} n^2 z_n \cos n\omega t - \gamma/\omega \sum_{n=1}^{\infty} n z_n \sin n\omega t + kz_0/\omega^2 \\ + k/\omega^2 \sum_{n=1}^{\infty} z_n \cos n\omega t \Bigg| \le F_0/\omega^2 \end{aligned}$$

and, taking the limit of ω to infinity, we have

$$\lim_{\omega\to\infty} \left| \sum_{n=1}^{\infty} n^2 z_n \cos n\omega t \right| = 0.$$

From which it follows that

$$\lim_{\omega\to\infty} \sum_{n=1}^{\infty} n^2 z_n \cos n\omega t = 0. \tag{7}$$

Due to the orthogonal properties of the oscillating Fourier components in the l.h.s of eq. (7), the coefficient of each component must also be zero at very high vibration frequencies, so that

$$\lim_{\omega\to\infty} z_n = 0 \quad \text{for} \quad n = 1, 2, \cdots. \tag{8}$$

To evaluate the response of the cantilever to ultrasonic vibration, let us integrate eq. (6) over one period of oscillation $T = 2\pi/\omega$. According to eqs. (5) and (8), z_c in eq. (6) can be replaced by z_0 at very high frequencies. Then

$$\begin{aligned} \int_0^T \Bigg(-m\omega^2 \sum_{n=1}^{\infty} n^2 z_n \cos n\omega t - \gamma\omega \sum_{n=1}^{\infty} n z_n \sin n\omega t + kz_0 \\ + k \sum_{n=1}^{\infty} z_n \cos n\omega t \Bigg) \mathrm{d}t \\ = \int_0^T F(z_0 - z_s - A\cos\omega t)\, \mathrm{d}t, \quad \text{where} \quad T = \frac{2\pi}{\omega}. \end{aligned} \tag{9}$$

The integral of the oscillating terms in the l.h.s. of eq. (9) containing $\cos n\omega t$, $\sin n\omega t$, is performed over a integer number of periods. Therefore, after integration these terms vanish. Taking the integral, we have

$$kz_0 T = \int_0^T F(z_0 - z_s - A\cos\omega t)\, \mathrm{d}t. \tag{10}$$

Let us assume that the $F(z)$ force curve does not depends upon the time derivatives of the coordinate z. This case corresponds to the absence of viscoelastic relaxation in the sample-tip interaction. Then, the time variable t in the eq. (10) can be replaced by the dimensionless variable $x = 2\pi t/T$ and this integral can be written as

$$kz_0 = \frac{1}{2\pi} \int_0^{2\pi} F(z_0 - z_s - A\cos x)\, \mathrm{d}x. \tag{11}$$

The r.h.s. of eq. (11) defines a new function, dependent upon the distance between the tip and the object and upon the amplitude of ultrasonic vibration, while possessing no frequency dependence (provided $F(z)$ has no viscoelastic behavior). This function can be calculated from the original force curve $F(z)$ once the average position z_0 is assumed. It allows us to evaluate the UFM response to the ultrasonic vibration by solving eq. (11). This equation is similar to the low frequency case of eq. (3), if a modified force curve $F_m(z)$ is defined as

$$kz_0 = F_m(z_0 - z_s, A),$$

where

$$F_m(\zeta, A) = \frac{1}{2\pi} \int_0^{2\pi} F(\zeta - A\cos x)\, \mathrm{d}x. \tag{12}$$

It is easy to see that if the original force curve is linear (i.e. $F(z) = -k_f z$), the averaged shift of the cantilever z_0 vanishes. In this case $F_m(z) = F(z)$ and the function is not affected by ultrasound. Therefore, the response of the UFM to ultrasound directly reflects a *nonlinearity* in the $F(z)$ curve.

3. Experimental Setup

The experimental setup was based on the modified commercial AFM (Seiko Instruments Inc. SFA 330) and its schematic diagram is shown in Fig. 2. A microfabricated Si_3N_4 cantilever with a resonant frequency of 33 kHz and spring constant k of 0.024 N/m was used. A polished (100) oriented Si wafer of 400 μm thickness was bonded to a PZT ceramic piezoelectric transducer with a resonant frequency of about 3 MHz using thin layer of crystalline salol (phenyl salicilate). The piezoelectric transducer in turn was bonded to the AFM stage and its electrodes were connected to the output BNC connector using soft wires.

A function generator with 50 Ω, +10 dB·m maximum output was used to drive the piezoelectric transducer at frequencies ranging from 0 to 9 MHz using cw, tone-burst, saw-tooth, and triangle modulated RF signals. This system excited ultrasonic vibrations in the sample at the resonant frequency of the transducer, 3 MHz, and its third harmonic at 9 MHz. The vertical vibrations were effectively excited also at lower frequencies ranging from 0 to about 500 kHz below the mechanical resonances. In this frequency range the sensitivity of the thin plate piezoelectric transducer generally has an approximately uniform frequency dependence. Then it is possible to evaluate the displacement sensitivity of the piezoelectric transducer by applying a DC voltage and measuring the resulting displacement of the cantilever. Therefore, the DC displacement sensitivity of the transducer estimated in this way was about 0.1 nm/V. Useful to mention here, that this estimation is valid only at lower frequencies, whereas at resonant and adjacent frequencies another approach (i.e. optical interferometry) has to be used.

4. Experimental Results and Discussion

Figure 3 shows: (1)—driving tone bursts of the piezoelectric transducer with a carrier frequency of 3 MHz, peak to peak amplitude of 2 V, and envelope frequency of 700 Hz and (2)—a resultant deflection of the cantilever measured by the differential PD. In (2), a shift of the averaged cantilever position z_0 following the envelope of the tone burst is observed. This is due to the nonlinear response to the ultrasonic vibration of the sample. Using a vibration with a carrier frequency in the range of 300 to 500 kHz, much lower than the resonance frequency of the piezoelectric transducer, an estimation of the vibration amplitude by the zero-frequency measurements (see §3) was possible. Then the amplitude required for nonlinear response was estimated to be 0.2–0.5 nm. An analogous (but not calibrated regarding to the vibration amplitude) shift of the averaged cantilever position due to the nonlinear response was observed at vibration frequencies up to 9 MHz.

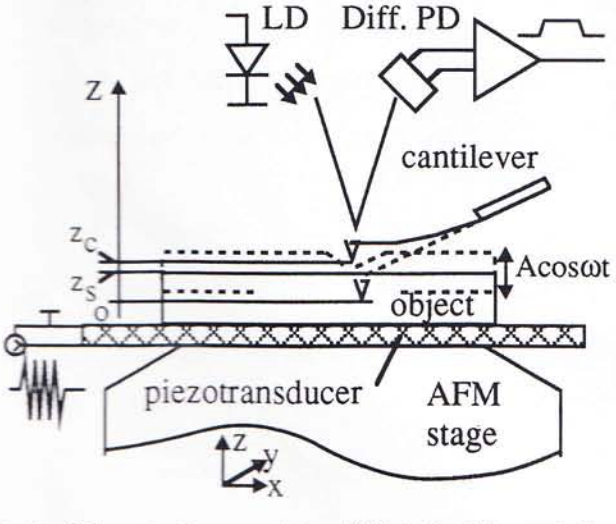

Fig. 2. Schematic diagram of the AFM in the Ultrasonic Force Mode (UFM) of operation; LD—laser diode, Diff. PD—position sensitive differential photo-diode.

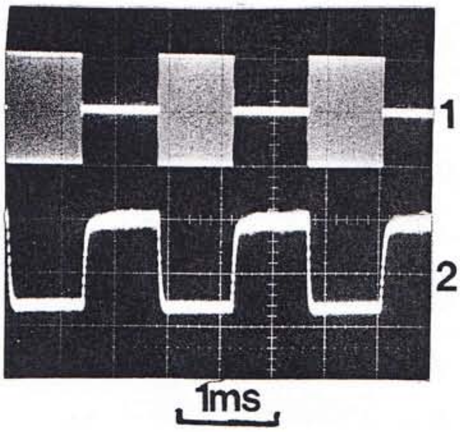

Fig. 3. Experimental verification of the nonlinear detection of ultrasound in the UFM: (1)—the driving signal for the ultrasonic tone bursts (RF frequency—3 MHz, RF amplitude—2 V p-t-p, and envelope frequency 700 Hz); (2) the resultant deflection of the cantilever z_0 measured by the differential PD (sensitivity 0.5 nm/div, negative change of PD signal corresponds to the positive (repelling) displacement of the cantilever), the deflection of the cantilever follows the envelope of the tone burst; a microfabricated Si_3N_4 cantilever with a spring constant $k \sim 0.024$ N/m and a resonance frequency $\sim$33 kHz was used.

Next, we investigated a dependence of the cantilever deflection upon the ultrasonic vibration amplitude of samples under the average force F_0 applied to the tip. The deflection of the cantilever was measured as the piezoelectric transducer was driven by a saw-tooth modulated signal with the carrier frequency of 3 MHz, peak to peak amplitude of 2 V, and envelope frequency of 700 Hz. To diminish the effect of the Z feedback loop on the cantilever position, integral regulation with the lowest possible gain was chosen. The results of these measurements for different values of the average force F_0 are shown in Fig. 4.

It was found that at small amplitudes of ultrasonic vibration, a weak attractive or an almost non-existent force was observed. This force became a repulsive force after the amplitude of ultrasonic vibration reached a certain threshold level, A_t. When we shift the average force F_0 to negative values (from $F_0 = 1.2$ nN to $F_0 = -1.2$ nN) so that an attractive force is applied to the cantilever by the sample, the transition took place

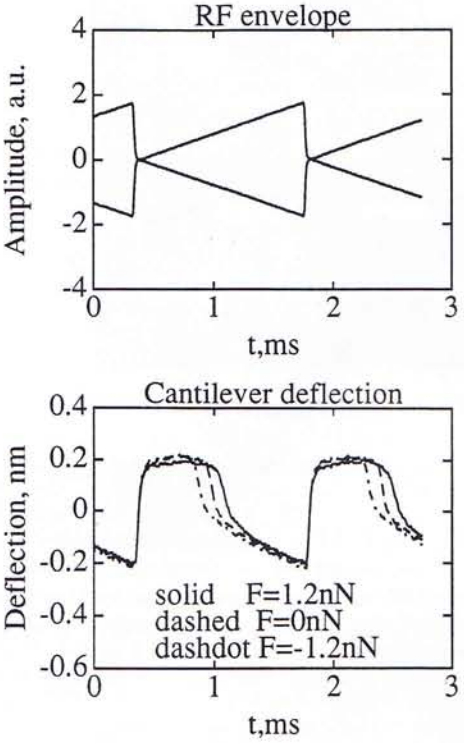

Fig. 4. Experimental study of the cantilever deflection vs amplitude of the ultrasonic wave for different average force F_0 applied to the tip—$z(A)$ curves; a) an envelope of the saw-tooth-modulated RF signal on the piezotransducer (RF frequency—3 MHz, RF amplitude—2 V p-t-p, and envelope frequency—700 Hz); b)—cantilever deflection for the average force F_0 equal to 1.2 nN, 0 nN, and −1.2 nN.

at a smaller amplitude of ultrasonic vibration, and a noticeable decrease in the transition threshold A_t (up to 40%) was observed.

This behavior may be related to the following fact. The higher average force level F_0 corresponds to the average position of the tip z_s close to the object on the repulsive branch of force curve $F(z)$ (Fig. 1). In this region the nonlinearity of the $F(z)$ force curve is small and an ultrasonic vibration of large amplitude is needed to reach the nonlinear region near the minimum on the force curve. The moment when this position is reached can be detected by a shift in the average cantilever position. Changing the average force to a negative value increases the distance between the tip and the object and allows us to reach the nonlinear region with a smaller amplitude A_t, resulting in a decrease of the threshold. This result suggests the possibility of estimating the shape of the $F(z)$ curve and paves the way for evaluating the local elasticity of the object by means of the UFM with nanoscale resolution.

5. Conclusion

We proposed a new method to detect an ultrasonic vibration in the samples of the AFM using the nonlinearity in the force curve $F(z)$ between the tip and the sample. A small amplitude ultrasonic vibration (less than 0.2 nm) is detected as an average displacement of the cantilever. It was found that a strong repulsive force is acting after the threshold of ultrasonic amplitude is crossed, with the amplitude of the threshold depending upon the average force applied to the tip. This transition may allow new sources of the contrast in AFM images. After solving the mechanisms of these newly found phenomena, it could provide new information on local viscoelastic properties and near field acoustic imaging of the subsurface structure.

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