Quantitative measurement of tip-sample interactions in amplitude modulation atomic force microscopy

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The author introduces an algorithm for the reconstruction of the tip-sample interactions in amplitude modulation atomic force microscopy ("tapping mode"). The method is based on the recording of amplitude and phase versus distance curves and allows the reconstruction of tip-sample force and energy dissipation as a function of the actual tip-sample distance. The proposed algorithm is verified by a numerical simulation and applied to a silicon sample in ambient conditions. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2355437]

The amplitude modulation atomic force microscopy (AFM)—also known as "tapping mode"—is the workhorse under the scanning probe microscopy techniques applied in ambient conditions and liquids.[1–6] This technique is based on a cantilever with integrated tip oscillating near the sample surface (see Fig. 1). During the approach toward the sample surface the cantilever oscillation is influenced by the tip-sample interaction which can be detected by a change of the oscillation amplitude A and the phase ϕ. Unfortunately, the analysis of these signals is greatly complicated by the nonlinearity of the tip-sample force, resulting in instabilities and hysteresis (see, e.g., Refs. 7–13). Nonetheless, it has been shown how the energy dissipation between the tip and the sample is linked to the phase[14–18] and the tip-sample force versus time can be deduced by inverting the cantilever trajectory.[19,20]

In this letter we present an approach enabling the quantitative reconstruction of tip-sample force and dissipation in amplitude modulation AFM. The proposed algorithm is based on the systematic recording of amplitude and phase versus distance curves. Analytical formulas can be applied to these data sets for the calculation of tip-sample force and energy dissipation versus the tip-sample distance. The reliability of the method is demonstrated by a numerical simulation. An experimental application on a silicon sample demonstrates its applicability.

Our analysis is based on the equation of motion for the cantilever which is given by

$$m\ddot{z}(t) + (2\pi f_0 m/Q_0)\dot{z}(t) + c_z(z(t) - D - A)$$
$$= a_d c_z \cos(2\pi f_d t) + F_{ts}[z(t),\dot{z}(t)]. \tag{1}$$

Here, $z(t)$ is the position of the tip apex at the time t; c_z, m, Q_0, and $f_0=\sqrt{(c_z/m)}/(2\pi)$ are the spring constant, the effective mass, the quality factor, and the eigenfrequency of the cantilever, respectively. The first term on the right hand side of the equation represents the external driving force of the cantilever. It is modulated with the constant excitation amplitude a_d at a fixed frequency f_d. The nonlinear tip-sample interaction force F_{ts} is introduced by the second term.

In the following, we solve the equation of motion by an analytical approach and consider only the steady-state solution given by the ansatz

$$z(t \geqslant 0) = D + A + A\cos(2\pi f_d t + \phi), \tag{2}$$

where ϕ is the phase difference between the excitation and the oscillation of the cantilever. For the mathematical treatment of the nonlinear tip-sample force it is mathematically advantageous to expand F_{ts} into a Fourier series. With the assumption that the tip touches the surface only slightly during an individual oscillation cycle, it is sufficient to consider only the first harmonics of the system.[15,21] Introducing only these terms into the equation of motion we obtain two coupled equations,[22]

$$\frac{f_0^2 - f_d^2}{f_0^2} = I_{even} + \frac{a_d}{A}\cos\phi, \tag{3a}$$

$$-\frac{1}{Q_0}\frac{f_d}{f_0} = I_{odd} + \frac{a_d}{A}\sin\phi, \tag{3b}$$

where the following integrals have been defined:

$$I_{even} = \frac{2f_d}{c_z A}\int_0^{1/f_d} F_{ts}[z(t),\dot{z}(t)]\cos(2\pi f_d t + \phi)dt, \tag{4a}$$

$$I_{odd} = \frac{2f_d}{c_z A}\int_0^{1/f_d} F_{ts}[z(t),\dot{z}(t)]\sin(2\pi f_d t + \phi)dt. \tag{4b}$$

For the further analysis of Eqs. (3) we examine these two integrals in more detail.

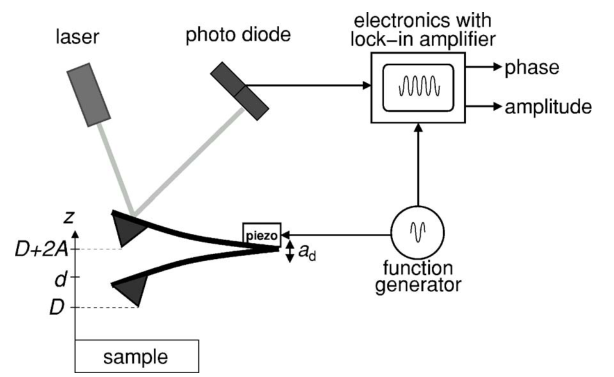

FIG. 1. Schematic setup of an amplitude modulation atomic force microscope. A frequency generator is used for the oscillation of the cantilever. Its oscillations are commonly detected with the laser beam deflection method, but other detection methods might be used as well. The oscillation amplitude A and the phase difference ϕ between the cantilever driving and oscillation are detected with a lock-in amplifier. The tip is assumed to oscillate between $D=d-A$ and $D+2A=d+A$.

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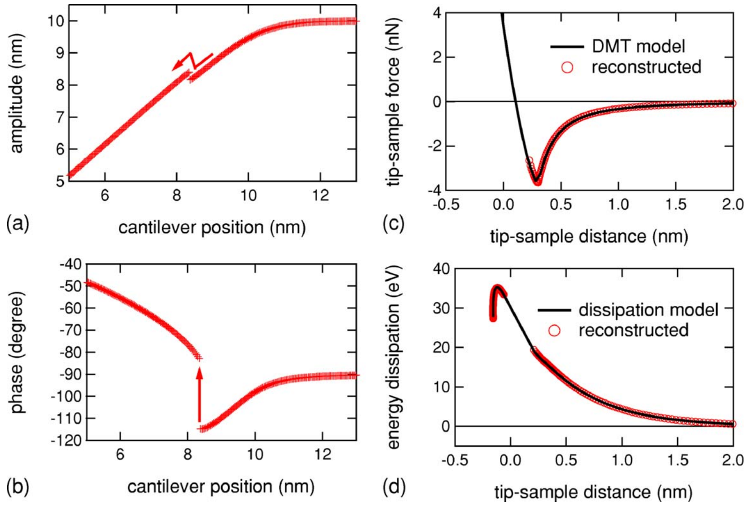

FIG. 2. (Color online) Numerical verification of the proposed algorithm. Based on the equation of motion [Eq. (1)] we numerically calculated the (a) amplitude and (b) phase vs distance curves during the approach toward the sample surface. Both curves show the typical instability and (c) only the data points before the jump are used for the reconstruction of the tip-sample force, (d) while the complete data set can be used for the energy dissipation curve. The assumed tip-sample interactions are plotted by the solid lines.

Introducing Eq. (2) into the first integral, I_{even} can be transformed to

$$I_{\mathrm{even}} = \frac{2}{\pi c_z A^2}\int_D^{D+2A} F_{\mathrm{ts}}\frac{z-D-A}{\sqrt{A^2-(z-D-A)^2}}dz. \tag{5}$$

The amplitudes commonly used in tapping mode are considerably larger than the interaction range of the tip-sample force. Consequently, the tip-sample forces are nearly zero in the integration range between $D+A$ and $D+2A$. For this large amplitude approximation[23,24] the last term can be expanded at $z \to D$ to $(z-D-A)/\sqrt{A^2-(z-D-A)^2} \approx -\sqrt{A/2(z-D)}$ and we get

$$I_{\mathrm{even}} \approx -\frac{\sqrt{2}}{\pi c_z A^{3/2}}\int_D^{D+2A}\frac{F_{\mathrm{ts}}}{\sqrt{z-D}}dz. \tag{6}$$

As a result of this computation we obtain an integral equation from Eq. (3a),

$$\underbrace{\frac{c_z A^{3/2}}{\sqrt{2}}\left[\frac{a_d\cos(\phi)}{A}-\frac{f_0^2-f_d^2}{f_0^2}\right]}_{\kappa} = \frac{1}{\pi}\int_D^{D+2A}\frac{F_{\mathrm{ts}}}{\sqrt{z-D}}dz. \tag{7}$$

The left hand side of this equation contains only experimentally accessible data and we define this term as κ. The benefit of our transformations is that the integral equation can be inverted[24] and as a final result we get

$$F_{\mathrm{ts}}(D) = -\frac{\partial}{\partial D}\int_D^{D+2A}\frac{\kappa(z)}{\sqrt{z-D}}dz. \tag{8}$$

With this integral equation it is now straightforward to calculate the tip-sample force from a spectroscopy experiment. First, one has to calculate the κ values as a function of the actual tip-sample distance $D=d-A$. In a second step the tip-sample force is numerically calculated from Eq. (8).

Further information about the tip-sample interaction can be obtained from Eq. (3b) since the integral I_{odd} is directly connected to the energy dissipation.[25,26] Introducing Eq. (2) into the integral it can be shown that $I_{\mathrm{odd}}=-\Delta E/(\pi c_z A^2)$, where ΔE is the energy dissipation per oscillation cycle. As a consequence we get the following formula from Eq. (3b):

$$\Delta E = [(1/Q_0)(f_d/f_0)+(a_d/A)\sin\phi]\pi c_z A^2. \tag{9}$$

The result follows also from the conservation of energy principle and is equivalent to the result of Cleveland *et al.*[15] However, in difference to this former work we suggest to plot the energy dissipation as a function of the nearest tip-sample distance $D=d-A$ in order to have the same scaling as for the tip-sample force.

A verification of the proposed algorithm is shown in Fig. 2. We performed computer simulations of the method by calculating numerical solutions of the equation of motion with a fourth-order Runge-Kutta method.[27] In order to introduce a reasonable tip-sample interaction we considered a tip-sample force with a conservative and dissipative part, i.e., $F_{\mathrm{ts}}=F_{\mathrm{con}}+F_{\mathrm{diss}}$. The tip is modeled as a sphere with radius R that interacts with a flat surface and experiences long-range attractive forces. If the tip comes very close to the sample surface, the repulsive forces between the tip and the sample are modeled by the well-known Hertz model. The adhesion forces are considered by a simple offset. This approach has been used in previous tapping-mode studies[12,13] and results in a force law given by

$$F_{\mathrm{con}}(z) = \begin{cases} -A_H R/6z^2 & \text{for } z \geqslant h_0 \\ \frac{4}{3}E^*\sqrt{R}(h_0-z)^{3/2}-A_H R/6h_0^2 & \text{for } z < h_0. \end{cases} \tag{10}$$

The effective modulus $E^*=1/((1-\mu_t^2)/E_t+(1-\mu_s^2)/E_s)$ depends on the elastic moduli $E_{t,s}$ and the Poisson ratios $\mu_{t,s}$ of the tip and the sample, respectively.

In order to consider also a dissipative tip-sample interaction a viscous damping term with a distance dependent damping coefficient is added, $F_{\mathrm{diss}}=F_0\exp(-z/z_0)\dot{z}$. The energy dissipation caused by this type of dissipation is given by[28]

$$\Delta E = 4\pi^2 f_d A F_0 z_0 \exp\left(-\frac{D+A}{z_0}\right) I_1\left(\frac{A}{z_0}\right), \tag{11}$$

where I_1 is the modified Bessel function of first kind.

Figures 2(a) and 2(b) display the resulting amplitude and phase versus distance curves during approach, respectively. The following parameters have been used: $A_H=0.2$ aJ, $R=10$ nm, $h_0=0.3$ nm, $\mu_t=\mu_s=0.3$, $E_t=130$ GPa, $E_s=1$ GPa, $F_0=10^{-6}$ N s/m, and $z_0=0.5$ nm. The eigenfrequency, the driving frequency, the natural Q factor, and the spring constant of the cantilever were chosen to be $f_0=f_d=300$ kHz, $Q_0=300$, and $c_z=40$ N/m, respectively. The amplitude as well as the phase curve show the often observed discontinuity caused by an instability. The resulting jumps are marked by arrows (see, e.g., Refs. 7–13 for a discussion of this phenomenon).

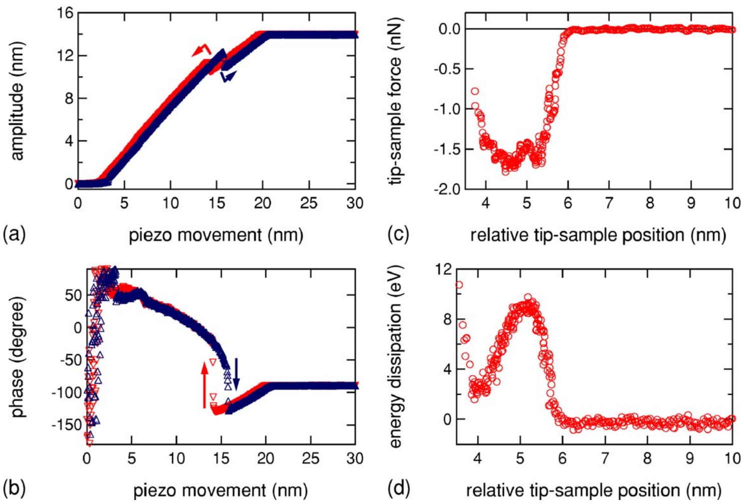

FIG. 3. (Color online) Typical dynamic force spectroscopy experiment on a silicon wafer in air (parameters: $f_d = f_0 = 328.61$ kHz, $c_z = 33.45$ N/m, $Q_0 = 537$). (a) A measurement of the oscillation amplitude as a function of the oscillation amplitude shows jumps at different positions during approach and retraction. (b) The jumps are also observed in the phase vs distance curves. (c) Using the proposed algorithm the tip-sample force can be reconstructed. This curve is calculated from the approach data. Only the data points before the jump are used for the reconstruction of the tip-sample force. (d) The energy dissipation per oscillation cycle can be easily obtained from the conservation of energy principle.

The subsequent reconstruction of the tip-sample interactions based on the data points of the amplitude and phase versus distance curves is shown in Figs. 2(c) and 2(d). The assumed tip-sample force and energy dissipation are plotted by the solid lines while the reconstructed data are shown by the symbols. The agreement demonstrates the reliability of the method. However, it is important to mention that the often observed instability in the amplitude and phase versus distance curves limits the reconstruction of the tip-sample force. Due to the resulting discontinuity in the nearest tip-sample distance D the κ values are not smooth enough for a reliable integration by Eq. (8) and the data points after the jump should not be used for a reliable application of the proposed algorithm. Nonetheless, the reconstruction of the energy dissipation is not affected by the instability and gives reliable values also after the jump.

An application of the method to experimental data obtained on a silicon wafer is shown in Fig. 3 where we used only the data points before the jump to reconstruct tip-sample force and energy dissipation. As a consequence the experimental force curve shows only the attractive part of the force between the tip and the sample showing a minimum of -1.8 nN. This result is in agreement with previous studies stating that the tip senses only attractive forces before the jump.[8,10,12]

In summary, we presented a method for the reconstruction of the tip-sample interactions in amplitude modulated atomic force microscopy (tapping mode). Based on the analysis of the equation of motion we gave explicit formulas for the reconstruction of the tip-sample force and energy dissipation as a function of the actual tip-sample distance. The reliability of the algorithm was demonstrated by a numerical simulation showing the agreement between the assumed tip-sample interaction models and the reconstructed interaction curves. However, due the often observed jumps in the amplitude and phase versus distance curves the reconstruction of the tip-sample force was limited to data points before this instability.

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