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Analysis of the interaction mechanisms in dynamic mode SFM by means of experimental data and computer simulation

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Abstract. The performance of a scanning force microscope (SFM) operated in the dynamic mode at high oscillation amplitudes is determined by the response of the system to a given set of interaction forces between the probing tip and the sample surface. To clarify the details of the cantilever/tip dynamics two different aspects were investigated in experiment and computer simulation. First, the interaction forces dominating the oscillatory motion of the probe were varied by applying an additional electrostatic force field. It is shown that such variations in the attractive part of the interaction potential can cause a switching between two different oscillation states and thereby significantly contribute to the contrast obtained from phase imaging. Secondly, the interaction forces were kept constant but the system response itself was varied by modifying the effective quality factor of the oscillating cantilever with an active feedback circuit. This provides a means to influence the transition from the attractive to the partly repulsive interaction regime, i.e. the onset of the intermittent contact or tapping mode.

Operating an SFM in the dynamic mode at high amplitudes (> 10 nm) offers the possibility of minimizing the contact time of the probing tip with the sample surface and thereby reduce lateral or friction forces involved in the scanning process. It also allows the collection of additional data related to different sample properties by recording the phase shift between the force driving the cantilever and its oscillation. In the last few years these features of operating the SFM in the dynamic mode [1, 2] were shown to be very useful to characterize several different kinds of sample surfaces, e.g. thin organic films, polymers, biological samples or even liquid droplets [3]. Although this has led to a steady increase in the number of possible applications, there are still several details of the interaction process between the tip and the sample that need further clarification. The overall goal must be to relate the experimentally accessible data, such as the amplitude and phase signal, more or less directly to specific sample properties, such as topography, elasticity and viscoelasticity.

Because highly nonlinear interaction forces are involved when the oscillating tip is in close proximity to a solid surface, the analysis of the dynamic system becomes quite complex. Therefore supplementary computer simulations based on proper mathematical models are useful to investigate the details of the interaction process. Basically, the equation of motion describing the dynamic properties of the probing tip has to be solved in such a way that the influence of different parameters characterizing the probe as well as the sample surface can be examined. There have been several reports recently on different approaches to this problem, providing analytical [4] as well as numerical [5–11] solutions. Most of them are based on the point-mass model, but there are also approaches which describe the complete flexural motion of the cantilever beam supporting the probing tip, as this becomes more relevant when the system is driven well above its resonance frequency [12]. Thus by simulating the dynamic system one can gain useful information on the complex interaction process of the oscillating tip and the sample surface.

1 Experimental and simulation methods

The simulation results presented here are all based on the point-mass model with the interaction forces being derived from MYD/BHW calculations [11, 13, 14] and applying the Verlet algorithm [15] to solve the equation of motion numerically. All experiments were performed with a NanoScope III MultiMode stage (Digital Instruments) equipped with an additional lock-in amplifier (EG&G Instruments, Princeton Applied Research, Model 5302) to measure the phase lag between the driving force and the cantilever response quantitatively. Rectangular cantilevers made of doped silicon (Nanosensors) with a nominal length of $L = 125\,\mu\text{m}$ were used.

By analogy with measurements of quasistatic force-distance curves in contact mode, the amplitude and phase shift as a function of the z-position were quantitatively investigated in the dynamic mode by means of simulation and

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experiment. As reported previously [6, 8] this process, called dynamic force spectroscopy (DFS), allows us to differentiate clearly between operating the SFM in the purely attractive and the partly repulsive regimes of the interaction forces. Comparison of the experimental data with the simulation revealed that the onset of the intermittent repulsive contact or tapping mode is indicated by a small step-like discontinuity in the amplitude signal and by a much more pronounced jump in the phase signal as the oscillating tip approaches the sample surface (Fig. 1a). The calculation of the forces acting on the probing tip at the point of closest approach during an oscillation cycle showed that these discontinuities mark the transition from the pure attractive to the intermittent repulsive contact regime of interaction (Fig. 5a). These findings were recently confirmed by Kühle et al., who came to similar conclusions [10]. By carefully adjusting the setpoint (used by the feedback loop to keep the amplitude signal constant) well above the discontinuity one can deliberately put the system into the attractive mode of operation. However, in most cases this state is unstable and due to the hysteresis shown in the DFS curve the system has the tendency to stay in the tapping mode once the tip has propagated into the repulsive part of the interaction forces. As a consequence of the hysteresis effect the SFM can even be operated in the attractive as well as in the partly repulsive mode at exactly the same setpoint settings. The fact that the phase signal exhibits a significant change at the transition point between the two system states can strongly influence the contrast obtained in phase imaging, but might also be used as a diagnostic to monitor the type of forces dominating the cantilever/tip dynamics.

2 Results

2.1 Modification of the interaction forces

In order to examine the influence of attractive forces on the dynamic system in more detail a bias voltage was applied between the SFM tip and the sample, which consisted of a chromium layer on a glass substrate. By adjusting the bias voltage we could control the additional attractive forces and thereby analyze their effect on the cantilever dynamics. Figure 1b shows a typical DFS curve obtained under the influence of an applied bias voltage. Comparison with the undistorted case (Fig. 1a) reveals that the additional attractive force field leads to a delay in the onset of the tapping mode, as marked by the discontinuities in the amplitude and phase signals. As depicted in Fig. 1c this effect is dependent on the applied voltage, i.e. the strength of the force field. This shows that the phase signal can exhibit significant changes while scanning at a given setpoint due to different force gradients between the probing tip and a specific surface site.

The effect becomes apparent when scanning over a test pattern structure like the one shown in Fig. 2a,b. A bias voltage is applied between the SFM tip and every second chromium line on the glass substrate. While scanning at a relatively high setpoint the phase signal allows us to identify the lines with an effective voltage difference to the probing tip (Fig. 2c). Whenever the tip is oscillating above one of those lines the additional force gradient prevents the onset of the repulsive tapping and the corresponding phase jump that would mark this transition does not occur at the given

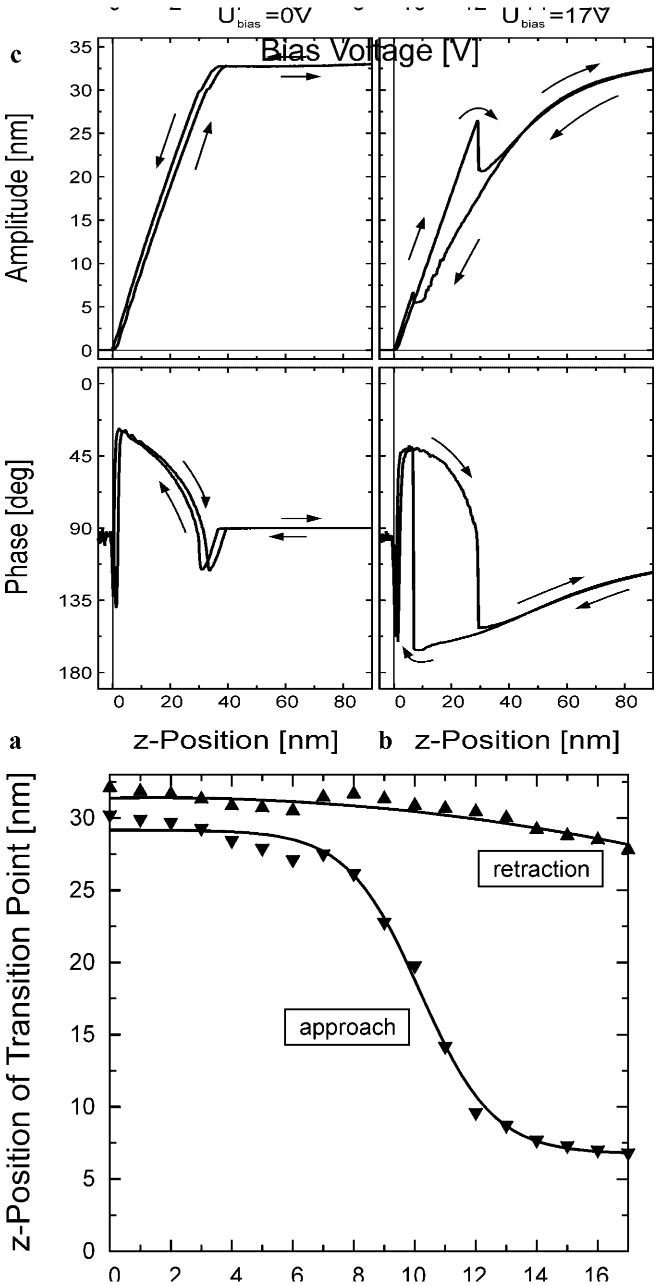

Fig. 1. a,b Experimental DFS curves as obtained by measuring the oscillation amplitude and phase shift as a function of the average probe-sample distance. While the dynamic system was driven at the resonance frequency of the free oscillating cantilever ($f = 325.09$ kHz), a bias voltage was applied between the doped silicon tip and the sample, consisting of a chromium layer on a glass substrate. **c** Position of the transition point between the purely attractive and partly repulsive interaction regime, as a function of the applied bias voltage

setpoint. On the other hand, there is almost no contrast between the lines connected to the tip potential and the glass substrate. However, when scanning at a lower setpoint the contrast of the phase image is changed significantly (Fig. 2d): while the topography image shows no difference, the phase contrast disappears almost completely, apart from edge effects. Due to the decreased setpoint the transition from the

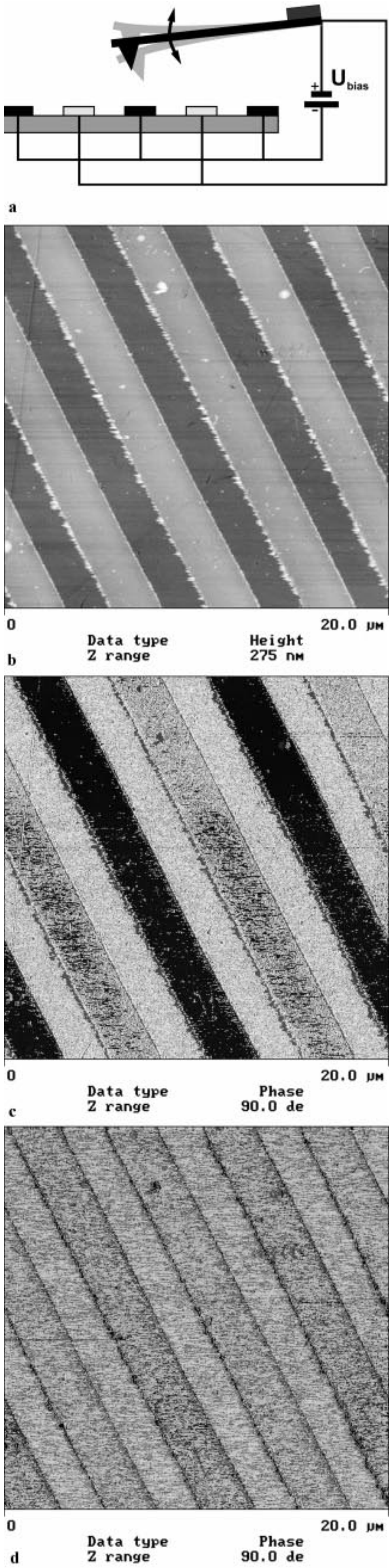

Fig. 2. a Schematic representation of the setup used for scanning on a test pattern structure consisting of chromium lines on a glass substrate, with a bias voltage applied between the tip and every second line of the sample. **b** Topographic SFM image of the structure and corresponding phase images at a high **c** and low **d** setpoint value

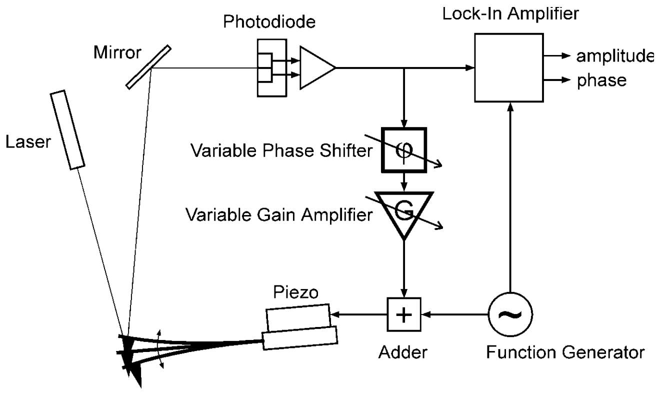

Fig. 3. Schematic diagram of the SFM with additional feedback circuit used for active control of the system response function. The setup allows the effective quality factor of the dynamic system to be changed by more than an order of magnitude

attractive to the intermittent repulsive interaction regime occurs on every spot of the test pattern, regardless of any voltage difference that might be present between the tip and the sample. The dynamic system stays in the intermittent contact mode all the time, so that the phase contrast would only be dominated by effects related to local differences in energy dissipation, e.g. caused by viscoelastic damping [16]. This example illustrates that changes in the local attractive forces can significantly contribute to the contrast observed in phase imaging by switching between two different dynamic modes, and that this effect strongly depends on the chosen setpoint.

2.2 *Modification of the system response*

Another important factor determining the tip-sample dynamics is the response of the system to a given set of interaction forces. The main parameter reflecting the response function is the shape of the resonance curve of the cantilever beam, as characterized by the quality factor $Q = \Delta f / f_{\mathrm{res}}$, with f_{res} and Δf denoting the resonance frequency and the width of the resonance curve, respectively. In order to examine their influence on the cantilever dynamics the interaction forces were kept constant, and instead the system response to these forces was altered.

In the experiments the *effective* quality factor of a given cantilever beam can be changed by means of an extra feedback circuit, which is added to the standard SFM setup and offers an active control of the damping of the dynamic system. By applying an appropriate feedback force to the oscillating cantilever its motion can be regulated in such a way that the modified response function of the system exhibits an increased (or decreased) quality factor. Different coupling mechanisms were proposed to apply feedback forces and thereby to regulate the cantilever response, like photothermal forces [17] or capacitive forces [18]. The setup used here offers the advantage that it does not require any additional mechanical setup, but applies the additional force via the standard piezo which is used to drive the cantilever base (Fig. 3) [19]. The main components of the feedback circuit are a variable phase shifter ϕ and an amplifier with adjustable gain G. The equation of motion describing the modified dynamic system reads:

$$m\ddot{z}(t) - \alpha\dot{z}(t) - kz(t) = F_0 \cos(\omega t) + G\mathrm{e}^{\mathrm{i}\phi} z(t) \,, \qquad (1)$$

where m is the effective cantilever/tip mass, α the damping constant and k the spring constant of the cantilever beam. F_0 and ω denote the magnitude and frequency, respectively, of the original harmonic driving force.

Separate investigations have shown that the cantilever motion stays almost perfectly sinusoidal ($z(t) \approx (A\sin(\omega t))$ even while tapping at very low setpoint settings [16]. Furthermore, by assuming that the appropriate phase shift of $\phi = \pm\pi/2$ is chosen and that all frequency shifts are small compared with the resonance frequency, the following approximation becomes valid:

$$e^{\pm i\pi/2} z(t) \sim \dot{z}(t) . \tag{2}$$

This leads us to a simplified equation of motion:

$$m\ddot{z}(t) - \alpha_{\text{eff}}\dot{z}(t) - kz(t) = F_0 \cos(\omega t) \quad \text{with} \;\; \alpha_{\text{eff}} = \alpha + G . \tag{3}$$

Thus by adjusting the gain of the feedback circuit it is possible to change the effective damping, which is directly related to the quality factor of the dynamic system $Q_{\text{eff}} = m\omega/\alpha_{\text{eff}}$.

DFS curves obtained in experiments under active feedback control with an increased quality factor (Fig. 4) exhibit a significant delay of the discontinuities in the amplitude and phase signal while the tip is approaching the surface. In particular, the phase signal can again be interpreted in terms of monitoring the current system state: it indicates that the increased sensitivity postpones or even prevents the onset of the tapping mode. Therefore the active damping circuit provides a means to control and enhance the size of the attractive interaction regime and thereby to make this operation mode more stable.

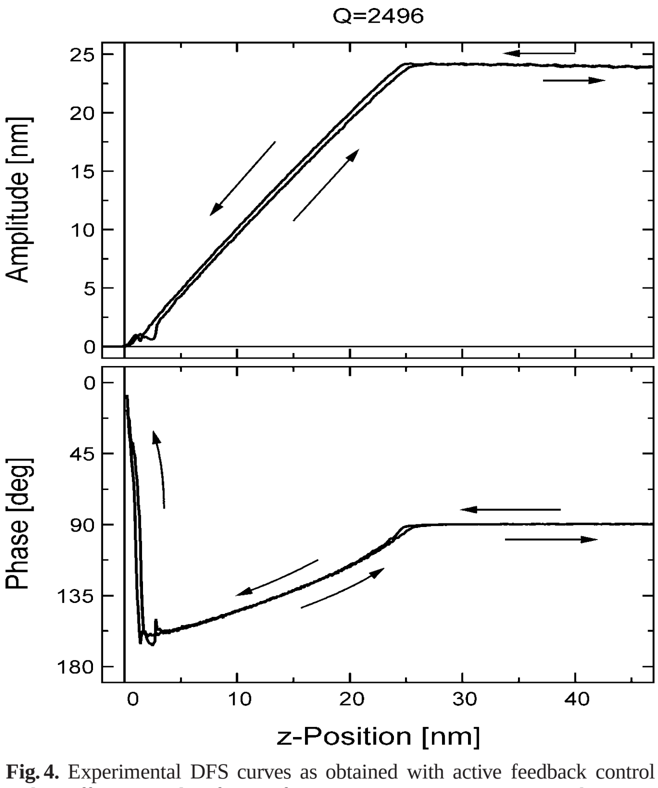

Fig. 4. Experimental DFS curves as obtained with active feedback control and an effective quality factor of $Q \approx 2496$ in air. Comparison with Fig. 1a shows that the increased sensitivity leads to a significant delay of the onset of the repulsive tapping, as marked by the discontinuities in the amplitude and phase signal. The original, unmodified cantilever exhibited a value of $Q \approx 448$ in air and the dynamic system was driven at the resonance frequency of the free oscillating cantilever ($f = 297.72$ kHz)

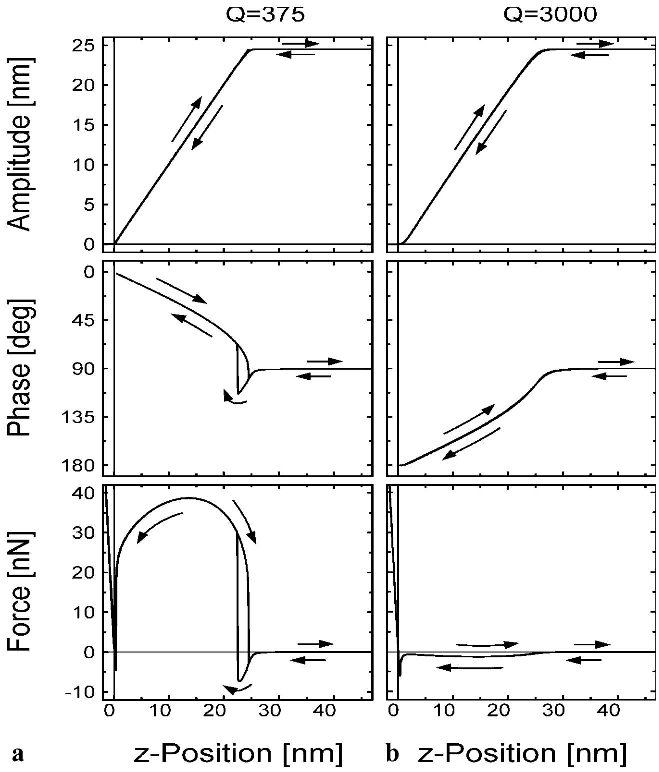

Fig. 5a,b. Calculated DFS curves as obtained by numerical computer simulations with a quality factor of **a** $Q = 375$ and **b** $Q = 3000$. Note that the discontinuities observed in the amplitude and phase signals are directly related to the transition from attractive to repulsive forces acting on the probing tip at the point of closest approach of each oscillation cycle. The increased quality factor leads to complete suppression of the repulsive intermittent contact

In order to validate these experimental findings numerical DFS calculations were performed. The computer simulations offer the possibility of changing the quality factor directly and thereby studying its influence on the oscillating system. Figure 5 shows the resulting DFS curve for an increased value of Q. The calculated force curve verifies that the increased quality factor leads to a delay or, in this case, even to a complete suppression of the onset of the intermittent contact as the tip is approaching the surface. This result reflects the fact that the system is becoming more sensitive to the attractive interaction forces, owing to the increased slope of the resonance curve, i.e. the increased value of Q.

3 Discussion and conclusion

To summarize the results, the two effects observed under variation of the interaction forces and the system response can both be interpreted by the following model: first, while the oscillating tip is approaching the surface, attractive interaction forces lead to a decrease of the effective spring constant of the cantilever and thus to a shift of the resonance curve to lower frequencies. If the cantilever is driven constantly at the resonance frequency of the free oscillating system, this shift must result in a steady decrease of the amplitude signal. On the other hand, the whole oscillating system is continuously approaching the sample and thereby bringing the point of closest approach of the probing tip closer to the surface. These two competing processes determine the onset of the intermittent contact with the sample surface: once the decrease of the oscillation amplitude is compensated by the reduced distance to the sample, the tip starts to tap, and is from there on in intermittent repulsive contact with the surface. The position of this transition point is directly determined by two factors: the strength of the attractive forces and the slope of the resonance curve, i.e. the sensitivity of the system response to these forces. Therefore an increase of the attractive forces or an increase of quality factor both favor the process of decreasing the oscillation amplitude. This results in a delay or even in complete suppression of the onset of the intermittent contact, as shown in the experiments and computer simulations.

Another factor determining the transition from the attractive to the repulsive part of the interaction regime is the oscillation amplitude. Separate investigations based on experimental and simulated DFS measurements showed that a reduction of the oscillation amplitude, i.e. the system's energy, also favors the operation of the SFM in the attractive regime.

In conclusion, it was shown that a lateral distribution of different attractive interaction forces on a sample surface can actually lead to a switching between the two states of the dynamic system while scanning. Therefore this effect has to be carefully considered when data obtained by phase imaging are analyzed. Furthermore, the feedback circuit presented can be utilized to stabilize and control the operation of a dynamic SFM in the attractive interaction regime and thereby to improve the image quality when scanning on sensitive sample surfaces [20].

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