Contact resonances in voltage-modulated force microscopy

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A study of the frequency dependence of the signal in piezoresponse scanning force microscopy of ferroelectric materials has been performed. It is found that, for soft cantilevers, the signal is governed by the cantilever elastic properties. Both ferroelectric-electromechanical and electrostatic interaction contributions to the overall signal were found to depend on the frequency of the testing voltage. Indications for optimal measurement regimes are given. © 2003 American Institute of Physics. [DOI: 10.1063/1.1592307]

Piezoresponse scanning force microscopy (PFM) is now a standard method for the study of ferroelectric phenomena. A small oscillating testing voltage is applied between the conductive tip of a scanning probe microscope and the bottom electrode of a ferroelectric sample. This ac voltage induces mechanical oscillations of the cantilever, which are then retrieved from the global deflection of the cantilever using a lock-in technique. The scenario initially proposed was that the surface of the ferroelectric sample is oscillating due to the converse piezoelectric effect (CPE), and that these oscillations are transmitted to the cantilever. Meanwhile it was shown that the electrostatic (ES) interaction between the tip/cantilever and the bottom electrode of the sample may play a significant role in the formation of the PFM contrast. Hong et al. even neglected the influence of the CPE on the cantilever vibration, and used the term “dynamic-contact electrostatic force microscopy” (DC–EFM) for the same technique (same experimental setup). Recently, the contrast in PFM was thoroughly analyzed and the limits for different contrast mechanisms were defined. Given the earlier findings, we use in the following the more general term contact-voltage modulated force microscopy (c-VMFM) instead of PFM or DC–EFM.

Labardi et al. provided a first report concerning the frequency dependence of the c-VMFM signal from TGS single crystals. However, they did not discriminate between the CPE and ES contributions. In this letter we will show that the properties of the cantilever strongly influence the VMFM measurements. The spring constant of the cantilever does not only control the (static) force applied to the sample, but also its own dynamical properties. In turn, the latter influence the magnitude and the phase of the VMFM signal. In order to study this influence we performed an analysis of the frequency dependence of the c-VMFM signal. We show that both CPE and ES interaction have frequency-dependent contributions to the c-VMFM signal, and that they can be separated by local hysteresis measurements.

The experimental setup is similar to that used in Refs. 1–11. A commercial scanning probe microscope (Autoprobe CP Research, Thermomicroscopes) working in contact mode and a lock-in amplifier (EG&G Instruments, model 7260) were employed to measure the vibrations of the cantilever. The mechanical oscillations were induced by applying a small ac voltage between the tip (Micromash, CSC12- and CSC11-series coated with W2C) and the bottom electrode of the sample. The ac amplitudes applied ranged from 50 mV up to 1 V, in order to keep the cantilever vibration within 3% of the static deflection. The ac voltage applied did not result in a change of the ferroelectric domain structure in our experiments. The spring constants of the cantilevers ranged from $k = 0.02$ N/m to $k = 0.5$ N/m corresponding to first free resonance frequencies ($f_{r1}$) of 9 up to 32 kHz. Hysteresis loop measurements were performed using a computer-controlled Keithley 2400 source meter in series with the ac source. The frequency of the triangular wave was 5 mHz. The samples used for investigations were chemical solution deposited lead zirconate titanate films, 100 nm thick, incompletely crystallized, showing rosette-type ferroelectric islands (1–2 μm in lateral size) surrounded by a pyrochlore phase. These samples were chosen because they permitted a comparison, on the same sample, of the induced cantilever vibrations for a contact of the tip with ferroelectric and nonferroelectric materials, respectively, preserving exactly the same ES interaction between the cantilever and the platinum bottom electrode.

The cantilever is the most important element of an atomic force microscope (AFM). Its role is twofold: (a) carrying the tip, it controls the interaction between tip and surface and (b) it provides information to the system about the tip displacement. Most commercial AFMs use an optical beam to detect the cantilever bending which is directly related to the force exerted on the tip and the tip displacement. In this work we refer only to this type of detection. This method works quite well in the quasistatic regime (vibrations below 1 kHz) such as in the usual topography imaging contact mode. Being an elastic beam, the cantilever has its own resonance frequencies corresponding to different vibration modes. The cantilever vibrations are governed by the boundary conditions (forces acting on the cantilever) and they have to be taken into account in the detection process. This is especially important when the cantilever is driven into oscillation at or near one of its resonance frequencies, when the vibration is most sensitive to changes in boundary condi-
tions, and this fact is already exploited in noncontact-VMFM (EFM and Kelvin force microscopy).

Figure 1 compares the frequency dependence of the noncontact-VMFM signal with the c-VMFM signal. The spectra were obtained while keeping the cantilever fixed above (O) and in contact with (—) the pyrochlore region of the sample surface, and are typical for all cantilevers measured in this work. The vibration modes (and therefore the resonance frequencies) of a cantilever are different in contact and in the noncontact case,12 as sketched in Fig. 1(c) for the first flexural modes. It has been calculated that the n° contact resonance (f°C) lies between f°C and f°C of the free cantilever (cantilever fixed only at one end).12 The reason why in our experiments f° is higher than f°C is the fact that in contrast to Ref. 12, the forces driving the cantilever into oscillation in VMFM of nonferroelectric surfaces are electrostatic, acting on both tip and cantilever. The force acting on the cantilever is distributed along its length and this significantly changes the induced deflection. Assuming a planar geometry the first harmonic of the ES force has the form $F_0 = -\alpha (V_{dc}-CPD)V_{ac}\sin(\omega t)$, where $\alpha = (1/2)\partial C/\partial z < 0$ is the derivative of the system capacitance with respect to the coordinate z normal to the surface, Vdc and Vac are the dc and the amplitude of the ac component of the applied voltage, and the contact potential difference (CPD) was typically $\approx 0.5$ V in our experiments. The response of the cantilever to this excitation force is supposed to be $h_0 = F_0/\kappa$, where $\kappa$ is the equivalent spring constant of the cantilever.6 As this formula has a static origin, it can only be valid at low frequencies. The frequency dependence of the cantilever response $A_0$ to an ac excitation may be empirically described near a resonance by the formula $A_0 = A_0(\omega^2 - \omega_0^2)^{1/2}$ and $\tan \varphi = \gamma_0(\omega^2 - \omega_0^2)^{1/2}$. Here, $A_0$ is the amplitude of oscillation at the resonance $f_0 = \omega_0/2\pi$, $\gamma$ is a damping coefficient, and $\omega$ is the driving angular frequency. This means that the phase of the cantilever oscillation for $\omega < \omega_0$ is shifted $180^\circ$ compared to the phase at $\omega = \omega_0$. This $180^\circ$ phase shift can be clearly seen in Fig. 1(b) for both contact and noncontact cases. This leads to the conclusion that the amplitude $h_0 = \beta_0 F_0/\kappa$ and the phase $\varphi_0$ of the electrostatically induced cantilever deflection have a complex dependence on frequency. Near resonances, $\beta_0$ and $\varphi_0$ have the form of $A_0$ and $\phi$. The phase $\varphi_0$ of the oscillation is especially important in VMFM, because it provides information on the sign of the CPD, surface charge, or polarization. Below any resonance, it can be noted that the cantilever oscillations in contact and in noncontact mode have a phase difference of $180^\circ$. This is caused by the different boundary conditions and by the fact that the local angular deflection and not the tip position of the cantilever is detected.13 The $180^\circ$ phase change of the cantilever oscillation at the resonance frequency means that the slope of the bias dependence $\eta_0 = \partial h_0/\partial V_{dc}$ changes its sign when passing through resonance.

If a Hertzian contact is assumed, the contact stiffness is $k^* = \frac{1}{6}E^*\pi^2R_0^4$,9,12–14 where $E^*$ is the reduced Young modulus of the tip-to-sample contact, R is the radius of the tip apex, and $F_0$ is the static (indentation) force. This implies that increasing $F_0$ leads to an increase of $k^*$, i.e., a shift of $f'_0$. In the point mass model, an increase of $F_0$ by a factor of 2 should result in an increase of $f'_0$ by roughly 12% (neglecting the increase of R). However, we did not detect any noticeable shift of $f'_0$ with increasing $F_0$ up to a factor 3. The reason for this behavior can be found in Fig. 2(a), showing a force spectroscopy measurement. From this calibration curve it is clearly seen that the contact force (the “setpoint” value) is ten times smaller than the adhesion force between tip and sample surface (2 nN compared to 23 nN). This means the static force between cantilever and sample is essentially the adhesion force when soft cantilevers are used under ambient atmosphere. Unfortunately, cantilevers capable of higher loading forces have $f'_0$ above the upper frequency limit of our experimental setup.

Recording the sample topography and the c-VMFM signal for different ac testing frequencies revealed an enhancement of the VMFM contrast near $f'_0$. Figure 2(b) shows the sample topography with a flat (nonferroelectric) background (“P”) and clusters (“rosettes”) of ferroelectric grains about 200 nm in lateral size (“F”). Figures 2(c) and 2(d), respectively.
In conclusion, we have shown that c-VMFM measurements performed with soft cantilevers are strongly influenced by the experimental conditions via the dynamical properties of the whole (electro)mechanical system composed of cantilever, tip, sample and adhesion layer. For the imaging of the ferroelectric domain structure we propose the use of a frequency near a contact resonance of the system, but not exactly on the peak, because its position is controlled by the nonuniform adhesion. An important advantage of the contact resonance is that the ac amplitude needed for testing can be very much reduced (in our case down to 50 mV), and this makes the method suitable for very thin films, below 50 nm in thickness. Quantitative measurements of the material properties should be performed with stiff cantilevers (in the strong indentation regime) but only for testing frequencies at least one order of magnitude below $f_{1\omega}$. The drawback is that the measurement will be affected by the large contact force applied, typically in the 1 μN range.

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