QUANTITATIVE IMAGING OF ELASTIC, PIEZOELECTRIC AND FRICTIONAL PROPERTIES OF MATERIAL SURFACES USING ATOMIC FORCE ACOUSTIC MICROSCOPY

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Abstract

In Atomic Force Microscopy the deflection and torsion of micro-fabricated elastic beams with a sensor tip are used to generate high-resolution images of sample surfaces. In dynamic modes, the cantilever is vibrated while the surface is scanned. Between the tip and the sample the force interaction occurs at least during a fraction of a vibration cycle. In Atomic Force Acoustic Microscopy flexural and torsional cantilever vibrations are excited by out-of-plane and in-plane ultrasonic sample surface vibrations, respectively. The contrast of AFAM images contains local elastic, adhesive and frictional sample properties. In the piezo-mode, an electric ac-voltage applied at the sensor tip contacting excites local surface vibrations and hence probes the local piezoelectric activity. Quantitative evaluations of AFAM images in the linear elastic interaction region yield local indention and shear moduli of sample surfaces.

Introduction

In Atomic Force Microscopy (AFM) [1] high-resolution images of sample surfaces are obtained by exploiting the deflection and torsion of micro-fabricated elastic beams with a sensor tip, which is in force contact with the sample. In dynamic AFM methods, the cantilever is vibrated while the sample surface is scanned. At least during a fraction of a vibration cycle the tip is in force contact with the sample. As imaging quantities the amplitude and the phase of the cantilever vibration, the change in the mean cantilever deflection, and the contact resonance frequencies can be used. Techniques allowing topography measurements are e.g. the contact and the tapping mode [2]. With other operation modes additional sample surface properties can be imaged, like e.g. friction (Friction Force Microscopy) [3], elasticity (Force Modulation, Pulsed Force Mode, Ultrasonic Force Microscopy) [4-6], and magnetic (Magnetic Force Microscopy) [7] and ferroelectric domains (piezo-mode techniques) [8].

In Atomic Force Acoustic Microscopy (AFAM) [9-14] ultrasonic fields in the sample are detected and imaged. Flexural and torsional cantilever vibrations are excited by out-of-plane and in-plane sample surface vibrations, respectively. The ultrasound is transferred from the sample to the cantilever via the interaction forces between the sensor tip and the sample surface. The contrast of images obtained with bending modes of the cantilever depend on local elastic and adhesive sample surface properties, while torsional mode images contain the shear elasticity and the friction forces in the contact.

In the piezo-mode [8], local sample surface vibrations are excited via the inverse piezoelectric effect by an electric ac-voltage applied at the sensor tip. Thus, the local piezoelectric activity of the sample determines the resulting cantilever vibration. In the ultrasonic piezo-mode, frequencies close to a contact resonance of the AFM cantilever are used to enhance the vibration amplitude and thus the image contrast [15-16].

The quantitative evaluation of the contrast of dynamic AFM images in order to determine local material properties is still a challenge. The contact resonance spectroscopy allows one to deduce elastic constants such as the indentation and the shear modulus from flexural and torsional contact resonance frequencies, respectively, provided that low enough excitation amplitudes are used permitting to linearize the interaction force [15-18]. Absolute measurement accuracy is = 30%, especially when stiff materials are investigated. The relative accuracy within an image is better than 5 %. A quantitative analysis of the nonlinearity of the interaction forces in AFAM to determine adhesion and friction parameters and of the piezo-mode to obtain local piezoelectric constants is the subject of ongoing research projects.

Physical background

To render possible a quantitative evaluation of AFAM and ultrasonic piezo-mode images it is convenient to use rectangular shaped cantilever beams, because their flexural and torsional vibration modes can easily be calculated. The cantilever is described as an elastic beam clamped at one end and with the sensor tip at the other end. Its free flexural and torsional resonance frequencies depend on its geometrical data and material parameters. A contact of the sensor tip with a sample surface stiffens the system and the resonance frequencies increase [9-18].

In general, vertical and lateral forces act in the tip-sample contact, which may contain elastic, adhesive (including electric and magnetic parts), damping, and friction forces. If a static load F_c is applied to the cantilever which presses the sensor tip into the sample and shifts it into the repulsive region of the force curve, the interaction is mainly determined by elastic forces. For an out-of-plane excitation vertical and for an in-plane excitation lateral elastic forces prevail and cause flexural and torsional cantilever vibrations, respectively. From the measured flexural and torsional contact resonance frequencies the vertical and the lateral stiffness of the contact k* and k^*_{lat} can be calculated [15-18]. The analysis of lateral vibrations has to take into account the tip length [10] and its lateral stiffness [19].

The Hertzian model of elastic contacts [20] relates the contact stiffness to the contact parameters:

$$k^* = \sqrt[3]{6E^{*2}RF_c}\ , \tag{1}$$

$$k^*_{lat} = 8a_cG^*, \tag{2}$$

$$a_c = \sqrt[3]{\frac{3RF_c}{4E^*}}\ . \tag{3}$$

Here, R is the tip radius, a_c is the radius of the contact area, and E* and G* are the reduced Young's and shear modulus of the contact, respectively. They depend on the Young's and shear moduli and the Poisson ratios E_t, E_s, G_t, G_s, $?_t$, and $?_t$ of the tip and the sample:

$$\frac{1}{E^*} = \frac{1-\nu_t^2}{E_t} + \frac{1-\nu_s^2}{E_s}\ , \tag{4}$$

$$\frac{1}{G^*} = \frac{2-\nu_t}{G_t} + \frac{2-\nu_s}{G_s}\ . \tag{5}$$

Eqs. (1) to (5) allow to determine the local Young's and shear modulus of the sample surface if the geometrical data and material parameters of the cantilever and sensor tip are known. The quantitative contact resonance spectroscopy is analogue to nanoindentation measurements, generalized to the determination of indentation moduli of anisotropic materials [15].

The cantilever data are sources of inaccuracies and errors in the experiments. Normally, the shape of the cantilever deviates from a rectangular beam of constant cross section changing its vibration spectrum. A problem is mode-coupling which occurs due to inhomogeneities in the cantilever properties and its mounting on the chip. Also the shape of the tip, which essentially influences the contact area, is not known accurately enough, and may be not spherical as assumed in the equations above. It may not be even of rotational symmetry because it is a single crystal. Additionally, it may change its shape during experiments because of tip wear. The lateral stiffness of the tip is not known, so that till now, it is difficult to obtain the same lateral contact stiffness k^*_{lat} from different torsional modes of the cantilever.

High-excitation amplitudes cover nonlinear regions of the interaction forces and the flexural cantilever vibrations are then influenced by adhesion. High amplitude torsional vibrations will cause slip in the tip-sample contact and thus contain friction forces [14].

In the piezo-mode, the sample surface vibrations, which generate the cantilever oscillations, are excited locally via the inverse piezoelectric effect. Thus, the local piezoelectric activity of the sample mainly contributes to the contrast in piezo-mode images. This part is superposed by elastic and adhesive tip-sample interactions. Contact resonances of the cantilever are exploited for image contrast enhancement in the piezo-mode [15-16]. Ferroelectric domains in the sample polarized perpendicularly to the surface, when excited in the piezo-mode, cause out-of-plane surface vibrations, which generate flexural cantilever vibrations [15-16]. It has been shown that in-plane polarized ferroelectric domains, when excited in the piezo-mode, add a lateral vibration component [21] enabling one to generate torsional cantilever resonances as well [18]. The local electric and strain fields generated around the sensor tip are extremely inhomogeneous accounting for a complicated quantitative evaluation. The main parameters playing a role in the imaging quantities are discussed in [15].

Examples of imaging and quantitative evaluation

The dynamic AFM methods have been applied to many different materials, e.g. crystals [13], multi-domain ferroelectric ceramics and thin films [15, 16], polymeric materials [9, 18], nanocrystalline magnetic thin films [22], diamond-like carbon layers [23], and clay in rocks [24], have been examined. Friction and stick-slip phenomena have been studied by measuring the torsional resonances of the cantilever, for example in bare and lubricated silicon samples [25].

Fig. 1 shows the topography and an AFAM contact-resonance image of a nanocrystalline Ni sample with a grain size of 167 nm. The cantilever spring constant was 48 N/m, the first two free flexural resonance frequencies were 166 kHz and 1031 kHz, respectively. The contact-resonance image was taken with the first flexural mode. The image size is 1.5 x 1.5 µm^2.

The flexural contact resonances yield the vertical contact stiffness (Fig. 2, image size 1.5 x 1.4 µm^2) and finally the local indentation moduli M. The results at the three points indicated in Fig. 2 are 234 (1), 184 (2), and 176 GPa (3), respectively. The data agree with indentation moduli calculated from literature values of Ni single crystal constants for different crystal orientations: $M_{(111)}$ = 223 GPa, $M_{(110)}$ = 218 GPa, $M_{(100)}$ = 202 GPa. A point by point quantitative comparison is not possible yet because the crystal orientations at the indicated positions are not known.

Fig. 3 shows the topography and vertical (f = 332 kHz, 1st bending mode) and lateral (f = 3.1 MHz, 1st torsional mode) piezo-mode images (size: 6 x 6 µm²) of a piezoelectric ceramic (Lead Zirconate Titanate, PIC 151). A Pt-Ir coated cantilever, spring constant k_c = 2.3 N/m, first two free bending resonances f_1 = 69.2 kHz and f_2 = 435.5 kHz, was used. In contrary to the topography, in the piezo-mode images ferroelectric domains of different polarization are visible.

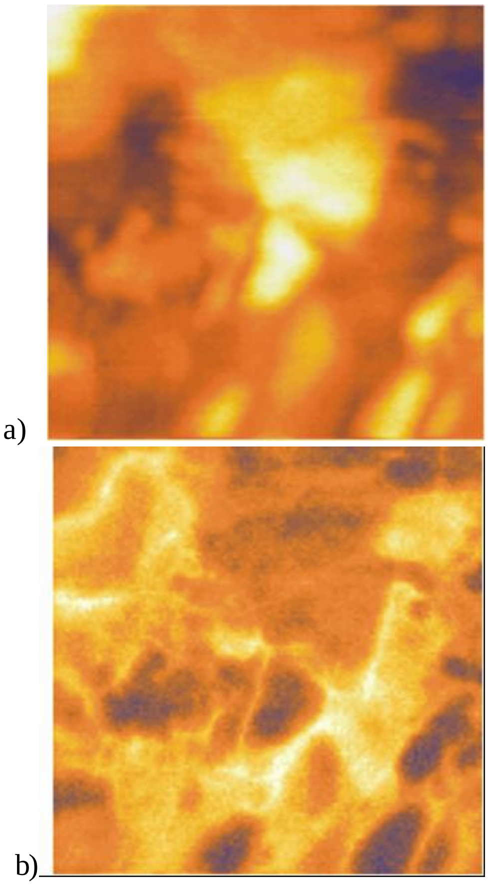

Figure 1 : a) Topography, height scale 10 nm, b) contact-resonance AFAM image, frequency scale 730 – 750 kHz, of nanocrystalline Ni

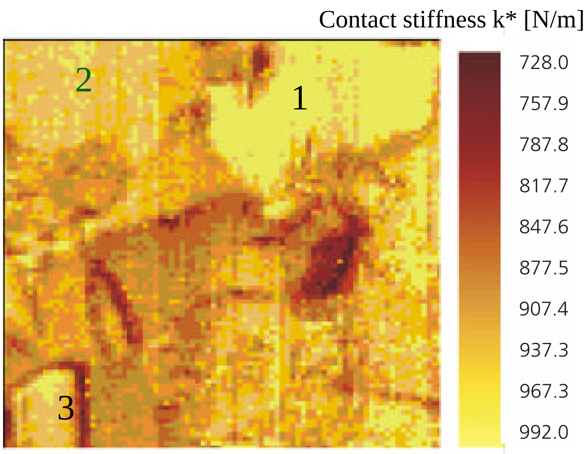

Figure 2 : Contact–stiffness AFAM image of Ni

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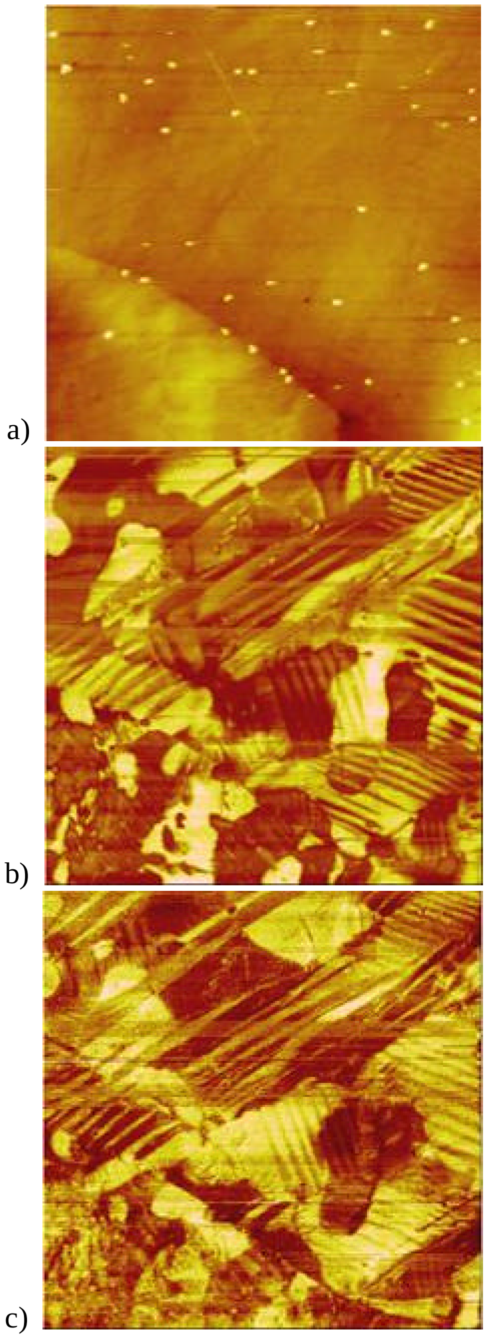

Figure 3 : a) Topography, height scale 20 nm, b) vertical and c) lateral piezo-mode images of PIC 151

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