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Research Article

Intermittent-Contact Heterodyne Force Microscopy

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Heterodyne Force Microscopy opens up a way to monitor nanoscale events with high temporal sensitivity from the quasistatic cantilever mechanical-diode response taking advantage of the beat effect. Here, a novel heterodyne ultrasonic force method is proposed, in which the cantilever is driven in amplitude-modulation mode, at its fundamental flexural eigenmode. Ultrasonic vibration in the megahertz range is additionally input at the tip-sample contact from the cantilever base and from the back of the sample. The ultrasonic frequencies are chosen in such a way that their difference is coincident with the second cantilever eigenmode. In the presence of ultrasound, cantilever vibration at the difference frequency is detected. Similarly as in heterodyne force microscopy, it is expected that the phase response yields information with increased sensitivity due to the beat effect.

1. Introduction

The mechanical-diode (MD) approach is based on the detection of the quasistatic response of an Atomic Force Microscopy (AFM) cantilever when the forces actuating upon the tip vary nonlinearly in the ultrasonic time scale [1, 2]. Up to now, the mechanical-diode response has been mostly exploited with the AFM working in contact or near-contact mode [3, 4]. Heterodyne Force Microscopy (HFM) [5] introduced a novel method, in which the cantilever tip is in contact with the sample surface, and ultrasound is excited both at the tip (from a transducer at the cantilever base) and at the sample surface (from a transducer at the back of the sample) at adjacent frequencies, and mixed at the tip-sample gap. By this procedure, the tip-sample distance is modulated in beats, and extremely small phase shifts of the sample ultrasonic vibration can be easily monitored via the much larger phase shift of the cantilever vibration induced at the difference (beat) frequency (beat effect). Here, I propose a novel heterodyne ultrasonic force method, named hereafter Intermittent-Contact Heterodyne Force Microscopy (IC-HFM) in which the cantilever is driven in tapping mode, at its fundamental resonance. Ultrasonic vibration in the megahertz range is additionally input at the tip-sample contact from the cantilever base and from the back of the sample. The ultrasonic frequencies are so chosen that their difference is coincident with the second-order cantilever resonance. Here, I demonstrate that in the presence of ultrasound, cantilever vibration at the difference frequency is detected. As explained below, the results can be attributed to the activation of a mechanical-diode signal during the time that the tip and the sample are in contact. The cantilever response can be controlled by varying the difference frequency, which is kept nearest to the second-order cantilever resonance. Phase data are expected to provide an increased sensitivity via the beat effect.

2. Principle of Measurement: Intermittent-Contact Heterodyne Force Microscopy (IC-HFM)

The principle of measurement in IC-HFM is illustrated in Figure 1. The fundamental resonance of the cantilever ω_0 is excited using a piezoelement at the cantilever base. The AFM is operated in tapping mode, with the feedback keeping constant the amplitude of the fundamental flexural cantilever resonance. Additionally, ultrasonic vibration at frequencies ω_1 and ω_2 is input at the tip-sample contact from ultrasonic piezos located at the cantilever base and at the back of the sample, respectively. Frequencies ω_1 and ω_2 are chosen in such a way that $|\omega_2 - \omega_1|$ equals the second-order cantilever resonance. As the cantilever is driven

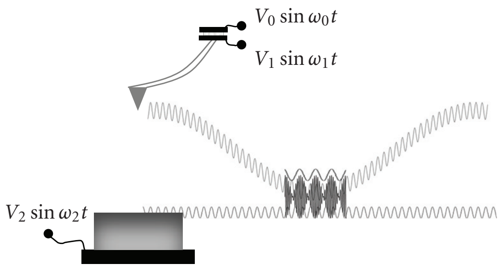

Figure 1: A schematic diagram illustrating IC-HFM (see text).

in its fundamental mode, it comes in contact with the sample surface for a certain time every period. During this contact time, the tip-sample distance is modulated in beats at the difference frequency $|\omega_2 - \omega_1|$ due to the tip and sample ultrasonic vibration and, provided that the tip and sample ultrasonic vibration amplitudes are sufficiently large, a mechanical-diode force will act upon the cantilever and resonantly excite its second-order vibration mode. As in HFM [5], the phase cantilever response at the difference frequency $|\omega_2 - \omega_1|$ is expected to provide information about tip-sample interactions with increased time sensitivity due to the beat effect.

3. Experiment

The measurements have been implemented by appropriately modifying a commercial AFM instrument (NANOTEC). The NANOTEC electronics was used for AFM operation in tapping mode, with the feedback keeping constant the amplitude of fundamental flexural cantilever resonance ω_0. Ultrasonic piezoelements were additionally attached to the sample and the tip holders (see Figure 1). Function generators were used to simultaneously excite the sinusoidal vibration of the sample surface and the cantilever tip at two frequencies ω_1 and ω_2 in the megahertz range ($\approx$4 MHz). The difference frequency $|\omega_2 - \omega_1|$ was purposely chosen to be coincident with the second-order cantilever resonance. Electronic mixing of the synchronous signals from both generators provided the reference signal for a lock-in amplifier to track the cantilever response at the difference frequency $|\omega_2 - \omega_1|$ in amplitude and phase. Rectangular Si cantilevers were used: VISTA probes provided by Nanoscience Instruments, with nominal spring constant $3\,\mathrm{N\,m^{-1}}$, and resonance frequencies $\omega_0 \approx 55$ KHz and $\omega_{\mathrm{second\_mode}} \approx 350$ KHz. The samples consisted in titanium nitride (TiN) coatings prepared by dc magnetron sputtering onto polished AISI 304 stainless steel discs in a vacuum chamber at room temperature using a water-cooled Ti target [6].

4. Results and Discussion

Figures 2(a) and 2(b) show the fundamental and the second-free flexural cantilever resonances, respectively. Figure 2(c) shows the spectral cantilever response measured with the FFT facility of our oscilloscope (Agilent DSO6104A) with

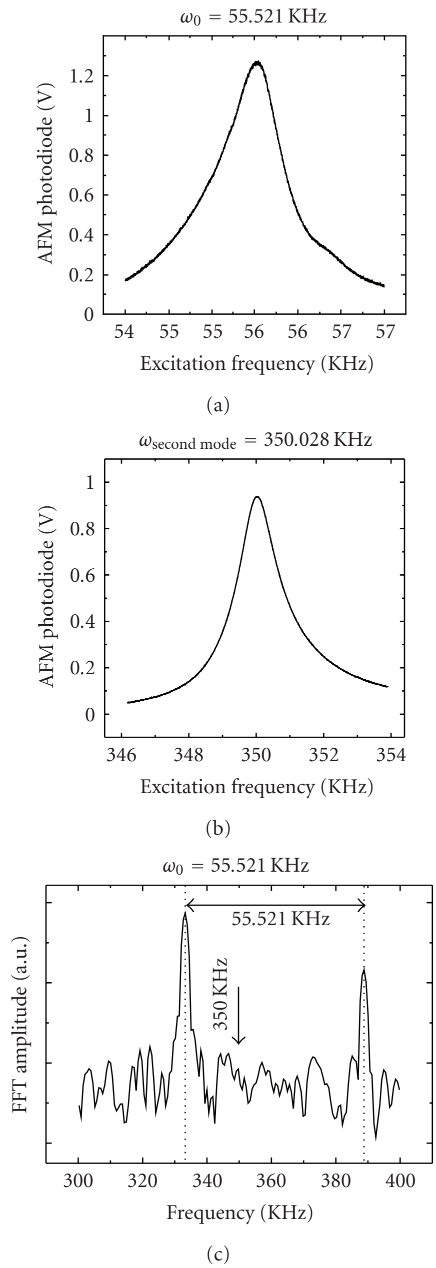

Figure 2: (a) Fundamental and (b) second-order flexural cantilever resonances measured with the cantilever tip out-of-contact with the sample surface (free cantilever modes), in the absence of ultrasonic vibration. (c) FFT of the AFM photodiode signal in tapping mode operation: excitation frequency $\omega_0 = 55.521$ KHz, set-point amplitude $V = 15\% V_0$.

the AFM operated in tapping mode. Peaks at 333.65 KHz and 388.55 KHz are apparent, which correspond to the 6th and 7th harmonics of the fundamental cantilever resonance. As it is well known [7–12], in tapping mode operation, a certain amount of power is shifted to higher harmonics due to the distortion of the harmonic motion of the cantilever at the bottom of each tapping oscillation cycle. Typically, high-amplitude harmonics always appear close to the resonance

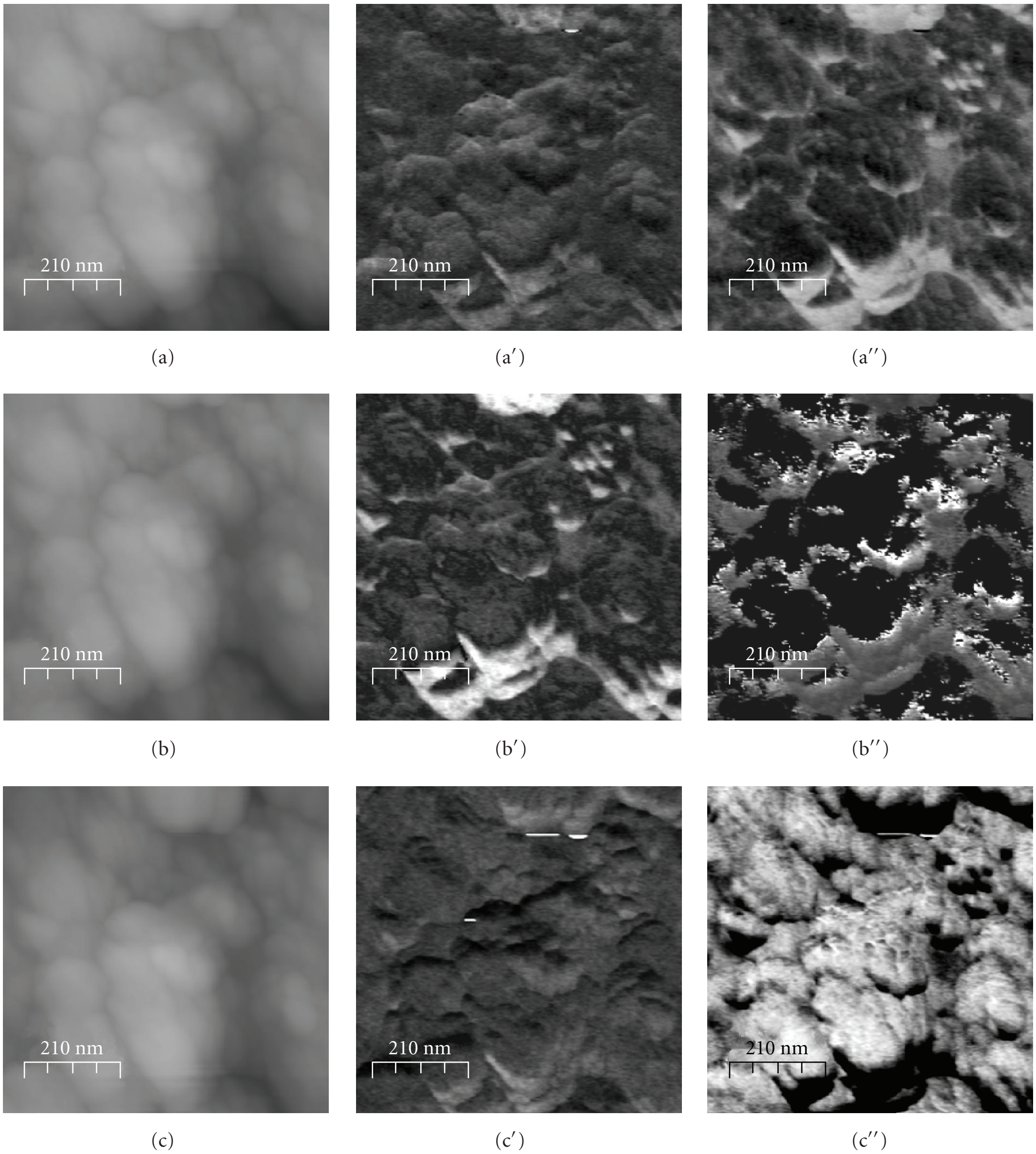

Figure 3: Topography (a)–(c) and IC-HFM amplitude (a′)–(c′) and Phase (a′′)–(c′′) images recorded over a same (700 × 700) nm^2 surface region of a TiN coating. Images with the same letter (i.e., (a), (a′), and (a′′)) were simultaneously recorded. Tapping parameters: ω_0 = 55.52 KHz, V = 31%V_0. Ultrasonic parameters: (a)–(a′′) ω_1 = 4.800 MHz, ω_2 = 4.449 MHz; (b)–(b′′) ω_1 = 4.800 MHz, ω_2 = 4.450 MHz; (c)–(c′′) ω_1 = 4.800 MHz, ω_2 = 4.451 MHz. V_1 and V_2 were kept constant. Greyscale range: (a)–(c) 35 nm; (a′) 3.6 V, (b′) 6.5 V, (c′) 3.2 V; (a′′) 4.2 V, (b′′) 10 V (out-of-scale), (c′′) 5.0 V.

frequencies of higher cantilever resonant modes, indicating a dependence of harmonics generation on cantilever resonances. In our case, the frequency corresponding to the second-order cantilever resonant mode lies in between the 6th and 7th harmonics of the fundamental resonance, as indicated by the arrow in Figure 2(c). In the absence of ultrasound, cantilever vibration in its second-order resonant mode could not be observed from the FFT spectra (see Figure 2(c)). Nevertheless, in the presence of ultrasound, when ultrasonic signals of $\omega_1 = 4.800$ MHz and $\omega_2 \approx 4.450$ MHz were simultaneously input to the cantilever tip and sample surfaces, respectively, while keeping the AFM in tapping mode operation (see Figure 1), a signal at $|\omega_2 - \omega_1| \approx 350$ KHz was clearly detected by means of the lock-in amplifier (see Figure 3). The lock-in used for the detection of the difference frequency was independent from the AFM electronics employed for tapping mode operation. The reference signal for the lock-in amplifier to detect cantilever vibration at the difference frequency of tip and sample ultrasonic vibrations was provided by electronically mixing the synchronous signals from the ultrasonic signal generators, using a simple mixer. A slight modification of the difference frequency led to significant variations in the signal detected by the lock-in amplifier. Figure 3 shows amplitude and phase outputs of the lock-in amplifier in different cases, together with the simultaneously recorded topographic image provided by the tapping-mode measurements. A maximum amplitude value of the signal detected by the lock-in amplifier was obtained when the difference frequency of tip and sample ultrasonic vibrations was coincident with the second-order cantilever resonance mode (Figures 3(b)–3(b′′), difference frequency of 350 KHz). Nevertheless, for this frequency, the phase response could not be properly measured. For difference frequency values slightly above or below the second cantilever resonance (Figures 3(a)–3(a′′), difference frequency of 349 KHz, and Figures 3(c)–3(c′′), difference frequency of 351 KHz, respectively), the phase contrast is reversed.

The excitation of cantilever vibration at the difference frequency of tip and surface ultrasonic vibrations is attributed to the activation of the mechanical-diode effect during the tip-sample contact time, as illustrated in Figure 1, in the same manner that it occurs in HFM [4, 5]. The procedures of Scanning Near Field Ultrasonic Holography (SNFM) [13] and Resonant Difference Frequency Atomic Force Ultrasonic [14] are similarly performed to HFM, with the difference (beat) frequency chosen in the range of hundreds of KHz, above the first cantilever resonance frequency in [13], and coincident with a high-order cantilever contact resonance in [14]. Recently, it has been demonstrated that GHz vibrations from an acoustic resonator can be detected by operating the AFM in tapping mode using the amplitude of the fundamental cantilever mode to control the feedback and collect the topography, and the second-order mode to detect acoustic information [15]. In IC-HFM, the operation mode is actually the same as in [15], save that here the resonant excitation of the second-order cantilever mode is activated by mixing of surface and tip ultrasonic vibrations via the nonlinearity of the tip-surface interaction.

The simultaneous excitation of the first and second cantilever modes in amplitude modulation (AM) AFM operation is currently attracting a great deal of interest [16–19]. It has been demonstrated that the sensitivity of the AM-AFM to map compositional changes can be enhanced by the simultaneous excitation of the first and second flexural modes [18]. A theory has been developed that considers coupling of the different eigenmodes of the cantilever by the virial of the tip-surface forces, and explains the origin of the high force sensitivity observed in multifrequency force microscopy experiments [19]. Here, it is demonstrated that it is possible to detect and control cantilever vibration at frequencies near the second-order cantilever resonance by simultaneously exciting ultrasonic vibration at the tip and surface, while the AFM is driven in tapping mode. Tip-sample interactions events that result in ultrasonic phase delays of surface versus tip vibration should be easily detected with high sensitivity taking into account the beat effect. The scope of the present work is limited to demonstrate the feasibility of the procedure, rather than to provide a detail understanding of the observed contrast. As it will be discussed elsewhere [20] TiN coatings may exhibit important differences in elastic contrast due to strain, structural defects, or coexistence of different phases. Contrast in IC-HFM images in Figure 3 may originate because of elasticity inhomogeneities.

5. Summary

The results presented here demonstrate the detection of cantilever vibration at frequencies coincident or next to its second-order resonant mode, excited by ultrasonic vibration from the cantilever tip and from the sample surface mixed via the nonlinearity of the tip-surface contact, while the AFM is operated in tapping mode. The amplitude in the response is at a maximum when the difference frequency of surface and tip ultrasonic vibration is exactly coincident with the second-order cantilever resonance. Contrast in the phase images of the cantilever response is reversed depending on whether the resulting difference frequency is above or below the frequency of the second-order cantilever resonance.

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