Mechanical nonlinear generation with coupled torsional harmonic cantilevers for sensitive and quantitative atomic force microscopy of material characteristics

O. Sahin, C. F. Quate and O. Solgaard
E. L. Ginzton Laboratory, Stanford University, Stanford, CA 94305

ABSTRACT

Tapping-mode has been the most widely used mode of operation in atomic force microscopy. Recent studies have shown that higher harmonics of the cantilever vibrations carries information about material properties such as stiffness, viscoelasticity or capillary forces. A major problem with higher harmonic imaging is the low signal to noise ratio. Here we present a micromachined cantilever that enhances the signal levels at a particular higher harmonic by 40 dB. We demonstrate that the higher harmonic signal is sensitive to the thickness of an oxide film thermally grown on silicon, and enable mapping of chemical composition variations across a polymer surface.

Keywords: atomic force microscopy, cantilever, higher harmonics, tapping mode, material properties

1 INTRODUCTION

Atomic force microscopy (AFM) is used to image materials with nanoscale lateral resolution. Although imaging with atomic resolution has been demonstrated, new methods that simultaneously map various material properties and topography are needed. Tapping-mode atomic force microscopy has been the most widely used mode of operation [1]. In this technique the AFM cantilever is vibrated near its fundamental resonance frequency in the vicinity of the sample so that the tip of the cantilever periodically contacts the sample. Due to non-linear tip-sample forces, the dynamics of the cantilever motion carries extensive information about the sample. The two measured quantities, amplitude and phase of the cantilever vibration, only relate to the average values of interaction forces however [2,3]. Recent studies have shown that higher harmonics of the cantilever vibrations has the potential to image material properties such as stiffness, viscoelasticity or capillary forces [3-8]. The signal to noise ratio at the higher harmonics is however not sufficient for practical measurements [9]. Previously we have demonstrated 20 dB enhancement of the higher harmonic response in a specially micromachined cantilever that uses a higher order vibration mode to resonantly enhance a particular harmonic [10].

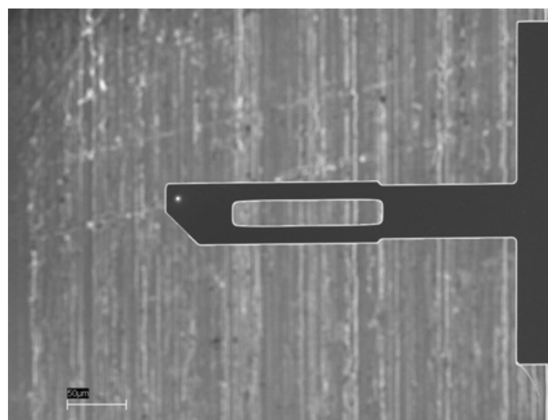

Fig. 1 SEM image of a torsionally coupled harmonic cantilever. The cantilever is 300 um long and 3 um thick. The width near the base is 40 um. The width of the arms is 15 um and the dimensions of the rectangular opening are 20 um by 130 um. The tip is located 12 um away from the longitudinal axis. The rectangular opening reduces the torsional bending stiffness and the torsional resonance frequency.

2 ENHANCING HIGHER HARMONIC GENERATION

Here we present recent advancements we made in higher harmonic imaging. We have designed, fabricated and experimentally demonstrated the use of a new class of micromachined cantilevers called coupled torsional harmonic cantilevers. These cantilevers have an asymmetric shape with an offset tip (see Fig. 1). When used in tapping-mode, the tip-sample forces excite torsional modes through higher harmonic generation.

In addition to the resonant enhancement, the torsional modes have a geometrical advantage over the flexural

modes traditionally used to detect cantilever motion. Because the width of the cantilever is much smaller than its length, a small tip displacement due to torsional bending will generate a larger angular displacement compared to flexural modes. The torsional vibrations can therefore be effectively measured with regular four quadrant position sensitive photo-detectors that are commonly used in atomic force microscopes.

When a higher harmonic matches a torsional resonance frequency, the vibrations at that harmonic are resonantly enhanced because of the large quality factors of the torsional modes (~1000). This operation principle is similar to the resonant enhancement demonstrated in ref. 10. Because of the geometrical advantage explained above, measuring torsional-mode deflection is a much more efficient method for detecting higher-harmonics.

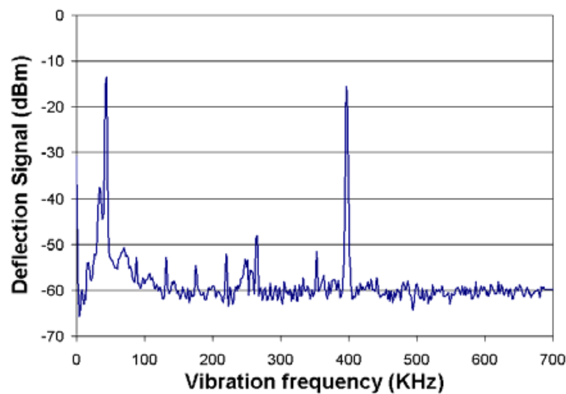

Fig. 2 Torsional vibration spectrum of a torsionally coupled harmonic cantilever. The torsional resonance frequency of this cantilever is at the exact 9th integer multiple of the fundamental flexural resonance frequency. The signal at the 9th harmonic is resonantly enhanced and has 45 dB signal to noise ratio in 1 KHz bandwidth.

In Fig. 2 we show the torsional vibration spectrum of a coupled torsional cantilever used in tapping-mode. The cantilever is driven at its first flexural resonance frequency of 44.14 KHz. It is the tip-sample forces that generate the torsional vibration spectrum shown in Fig. 2. The first peak in this spectrum is also at 44.14 KHz. Although torsional vibrations have a component at this frequency, the signal measured at this frequency is mainly due to the cross talk from the large flexural vibration. A small misalignment of the cantilever and photo detector can result in significant cross talk. There are other peaks in the vibration spectrum that appear at the integer multiples of the driving frequency. These are the higher harmonics. The torsional resonance frequency of this cantilever is at 397.26 KHz, which is also the 9th harmonic of the drive frequency. Therefore, the signal at this harmonic is much stronger. It has a signal-to-noise ratio of 45 dB in 1 KHz bandwidth.

There are a number of strategies available to tune the ratio of the torsional resonance frequency to the fundamental flexural resonance frequency to a desired integer. The cantilever in Fig. 1 has a rectangular opening that reduces the torsional stiffness of the cantilever. Reduced torsional stiffness results in a reduction in the torsional resonance frequency. On the other hand, the flexural modes of the cantilever are symmetric to the longitudinal axis and the presence of the hole does not affect the flexural stiffness as long as the effective width is the same. To match the integer ratio precisely, we have fabricated cantilevers with slight differences in the widths of the rectangular openings and parallel arms. Fabrication of the cantilevers was based on a silicon on insulator process described in detail previously [10].

2.1 Nanomechanical Measurements

Theoretical modeling of higher-harmonic generation has shown that the amplitude of a higher harmonic strongly depends on the stiffness of the sample under test [6,11]. In order to observe this experimentally, we have prepared a thin film of thermal oxide with a thickness gradient grown on a silicon wafer. The thickness gradient is obtained by polishing the wafer at a shallow angle after a uniform oxidation step. The polished wafer has a bare silicon surface on one side of the wafer, which develops an extremely thin native oxide layer after the polishing step. The thickness of the thermal oxide layer increases linearly beyond the native oxide region. A schematic of the cross section of the silicon wafer is given in Fig. 3. The Young's modulus of silicon dioxide is less than the Young's modulus of silicon. Therefore, the effective stiffness of the surface is that of the silicon where the film is negligibly thin, and then it is monotonously reduced to the stiffness of silicon dioxide as the thickness of the thermal oxide increases.

We have scanned the surface of the wafer along the thickness gradient in tapping mode with a coupled torsional harmonic cantilever. The amplitude of the enhanced higher harmonic is given in Fig. 3. The schematic of the wafer cross-section is drawn to relate the measured amplitudes and the position on the wafer. On the native oxide, the signal level has a constant value, while the harmonic amplitude drops rapidly to a minimum, as the film gets thicker. Beyond a certain thickness, the amplitude signal is slightly decreased as the thickness of the oxide is further increased. This oscillatory response of the harmonic amplitude with respect a monotonous

change in effective Young's modulus of the surface has been predicted previously [11]. The response of the harmonic amplitude as well as the location of the first minimum depends on the spring constant of the cantilever, the free-oscillating and set point amplitude of the vibrations, and the geometry of the tip. A careful choice of the cantilever design and tapping conditions will optimize the sensitivity over a specific stiffness range. The measurements in Fig. 3 are done with a cantilever spring constant of 1 N/m. Silicon and silicon dioxide are relatively stiff materials, so the sensitivity to variations in the thickness of the thermal oxide can be improved by using a cantilever with a larger spring constant.

The measurements show that for the first 300 nm of the oxide film, the sensitivity to the film thickness is approximately 0.1 nm/√Hz. This calibration is done by slowing the scan speed and reducing the scan size around the linear region of the harmonic amplitude response, and then measuring the minimum thickness variation that results in a variation in the harmonic amplitude above the noise level.

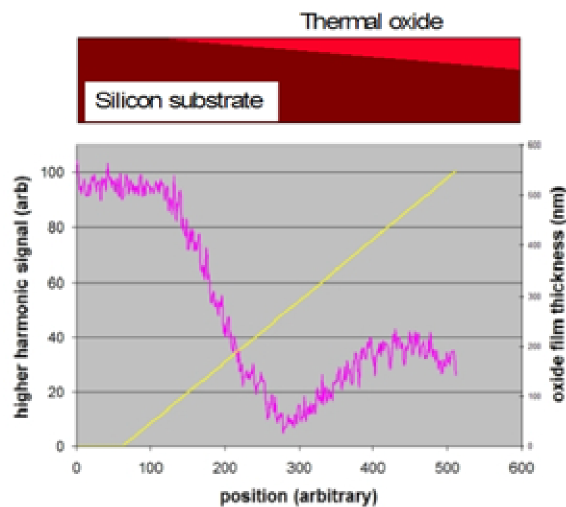

Fig. 3 Schematic of the thermally oxidized wafer polished at an angle and the measured higher harmonic amplitude across the wafer. The schematic cross section is shown to illustrate the location of the signals measured.

2.2 Mapping Chemical Compositions

Mechanical properties of materials highly depend on their chemical components. This relation allows us to map chemical composition variations across surfaces with the harmonic imaging technique. The imaging is done in tapping mode while the amplitude of the enhanced higher harmonic is monitored. Therefore the topography and higher harmonic image is generated simultaneously and they are perfectly registered. In Fig. 4, the topography and higher-harmonic image of a polymer surface are shown. The surface is composed of two types of polyethylene. One is amorphous and the other is ordered to some degree, resulting in a difference in hardness. The topography image on the right barely shows the features on the surface, whereas the harmonic image has three distinct colors, dark brown, light brown, and white. These levels correspond to the two types of polyethylene and the silicon substrate. Silicon is the stiffest of all and appears as white (the highest signal). Amorphous regions are relatively softer and they appear as dark brown.

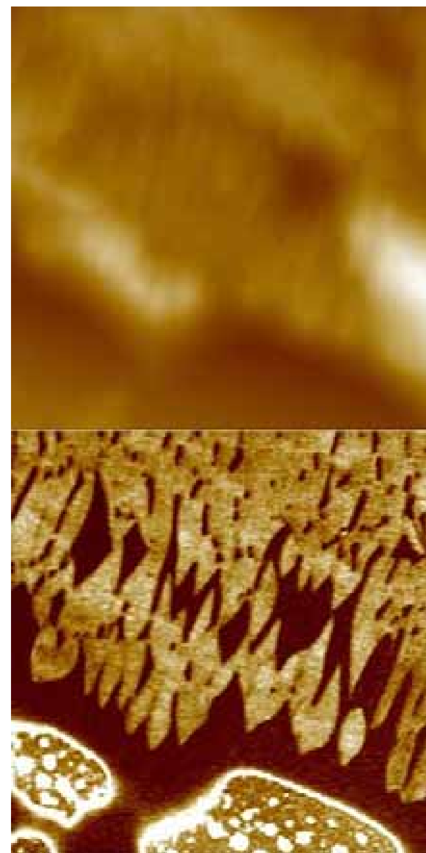

Fig. 4 Topography (top) and higher harmonic amplitude (bottom) image of a polymer surface on a silicon substrate. The harmonic image shows three color levels, white, light brown, and dark brown, corresponding to the three materials on the surface; silicon, amorphous polyethylene, and ordered polyethylene. Scan area is 5.6 microns

3 CONCLUSIONS

The enhanced higher harmonic generation achieved with the coupled torsional harmonic cantilever presented in this paper enables sensitive measurement of mechanical properties with nanoscale lateral resolution. The applications of harmonic imaging demonstrated here show the potential of nanomechanical measurements for the study and design of materials engineered at the nanoscale.

References

[1] Q. Zhong, D. Inniss, K. Kjoller, and V. B. Elings, "Fractured polymer/silica fiber surface studied by tapping mode atomic force microscopy," *Surf. Sci.* **280**, L688 (1993).

[2] J. P. Cleveland, B. Anczykowski, A. E. Schmid, and V. B. Elings, "Energy dissipation in tapping-mode atomic force microscopy" *Appl. Phys. Lett.* **72**, 2613 (1998).

[3] A. S. Paulo and R. Garcia, "Unifying theory of tapping-mode atomic force microscope" *Phys. Rev B.* **66,** 041406(R) (2002).

[4] R. Hillenbrand, M. Stark, and R. Guckenberger, "Higher-harmonics generation in tapping-mode atomic force microscopy: Insights into tip-sample interaction" *Appl. Phys. Lett.* **76**, 3478 (2000).

[5] W. Stark and W. M. Heckl, "Fourier transformed atomic force microscopy: tapping mode atomic force microscopy beyond the Hookian approximation" *Surf. Sci.* **457**, 219 (2000).

[6] O. Sahin and A. Atalar, "Simulation of higher harmonics generation in tapping-mode atomic force microscopy" *Appl. Phys. Lett.* **79**, 4455 (2001)

[7] M. Stark, R. W. Stark, W. M. Heckl, and R. Guckenberger, "Inverting dynamic force microscopy: from signals to time resolved forces" *PNAS* **99**, 8473 (2002).

[8] R. W. Stark and W. M. Heckl*, Rev. Sci. Instrum*. **74**, 5111 (2003).

[9] T. R. Rodriguez and R. Garcia, "Tip motion in amplitude modulation (tapping mode) atomic force microscopy: Comparison between continuous and point-mass models" *Appl. Phys. Lett.* **80**, 1646 (2002)

[10] O. Sahin, G. Yaralioglu, R. Grow, S. F. Zappe, A. Atalar, C. F. Quate, and O. Solgaard, "High resolution imaging of elastic properties using harmonic cantilevers" *Sensors and Actuators A,* **114,** 183 (2004)

[11] O. Sahin, A. Atalar, C. F. Quate, and O. Solgaard, "Resonant harmonic response in tapping-mode atomic force microscopy," *Phys. Rev. B.* **69** 165416 (2004)