Project co-financed by the European Union from the European Regional Development Fund

Sławomir PAWŁOWSKI, Grzegorz DOBIŃSKI, Marek SMOLNY
University of Łódź, Faculty of Physics and Applied Informatics, Łódź
slpawlowski@gmail.com, gdobinski@mvii.uni.lodz.pl,
msmolny@mvii.uni.lodz.pl
Andrzej MAJCHER, Mirosław MROZEK
Institute for Sustainable Technologies – National Research Institute, Radom
andrzej.majcher@itee.radom.pl, miroslaw.mrozek@itee.radom.pl

IMAGING SYSTEM USING HIGHER-HARMONIC IN THE TAPPING MODE OF THE ”TERRA AFM” MICROSCOPE

Key words

Atomic force microscope, tapping mode, synchronous detection, phase imaging.

Abstract

Terra AFM is the atomic force microscope designed and built by the authors as a device for research applications in advanced technologies in industry and in teaching. In tapping-mode, in atomic force microscopy, the interaction between the tip and the sample is, in fact, non-linear and consequently higher harmonics of the fundamental resonance frequency of the oscillating cantilever are generated. In this paper, we present the Terra AFM system using the method of synchronous detection that allows simultaneously recording the amplitudes and phases of the fundamental resonance frequency and of the higher harmonics. The used detection system, composed of 16 bit 100 mega-samples per second (MSPS) analogue-to-digital converter (ADC) and field-programmable gate array (FPGA) device, allows measuring the amplitude and phase of the cantilever within one oscillation cycle and with good signal-to--noise ratio. As a result, good-quality images at higher harmonics could be obtained with the use of conventional cantilevers. The obtained results prove that higher-harmonics imaging can be used to distinguish between different materials. High spatial resolution (about 1 nm) of the presented system is also demonstrated.

Introduction

The main criterion for the development of an atomic force microscope AFM Terra was the ease of use while maintaining high measurement parameters [1]. Modular mechanical design, control, and software were used, which allows making changes and modifications to extend the range of applications. Such changes are due to the experience gained with the use of the built microscopes and are focused on their better adaptation to the objects of research.

The use of the AFM microscope for characterization of objects of heterogeneous rheological structure is often done with the use of an imaging phase. Thus, improvement of the microscope "Terra AFM" is an imaging module utilizing advanced measuring amplitude and the phase of harmonics of the signal from the probe operating in tapping mode (intermittent contact mode). The module improves the quality of the measurement properties of visco-elastic structures, such as composite materials and biological objects.

The most widely used mode of operation of AFM is the tapping mode. The main reason is that, in this mode, the lateral interaction forces between the tip and the sample are minimized. In the tapping mode, the cantilever is excited to vibrate (with free amplitude) at or close to the resonant frequency (this resonant frequency is one of its flexural resonances) of the fundamental mode and is brought close to the studied sample surface so that the tip makes intermittent contacts (tapping) with the surface once in every oscillation period. The contact with the surface alters the amplitude and phase of the cantilever vibration. The vibrations are detected with an optical system where a laser beam reflects from the back of the cantilever and then falls onto a position-sensitive photodiode. As the cantilever is scanned across the surface, the vibration amplitude is maintained at a set-point value (below the free amplitude) through a feedback loop that adjusts the height of the cantilever base. Therefore, the feedback signal reflects the topography of the sample surface. The tip-sample interaction is in fact non-linear. As a consequence, this results in the appearance of higher harmonics of the fundamental oscillation (resonance frequency) of the cantilever [2, 3].

The phase shift between excitation and response of the vibrating cantilever depends on the energy dissipation during the tip-sample contact [3, 4, 5]. Balantekin and Atalar [6] showed that the amount of power dissipated in a sample is related to the mechanical properties of the sample, such as viscosity and elasticity. According to their theoretical considerations, for a given sample, elastic properties one can determine approximately the sample-damping constant by measuring the average power dissipation.

A theoretical analysis carried out by Stark and Heckl [7] showed that information on the elastic properties of the sample surface is contained in the higher harmonics of the fundamental oscillation signal in tapping-mode AFM. In addition, Sahin et al. [8] showed that higher harmonics offer the potential for imaging and sensing material properties at the nanoscale. They pointed out that resonantly enhanced higher harmonics are sensitive to the stiffness of the material under investigation.

The measurement techniques used commonly in commercial AFM allow one to control only the amplitude and phase of the fundamental mode (the first order Fourier component in frequency domain) of the cantilever, and information on the higher-order Fourier components, related to the non-linear interaction between the tip and the sample, is lost. In this paper, we present an AFM system using the method of synchronous detection that allows simultaneously recording the amplitude and phase of the higher harmonics of the fundamental oscillation of the cantilever. As a result, information on the non-linear tip-sample interaction, and, in particular, an insight into the mechanical properties of the sample, can be obtained. Because of the fact that higher harmonics signals obtained with the use of conventional cantilevers and conventional detectors/lock in amplifiers systems have low signal-to-noise ratios, the authors of previous works concerning the analysis of higher harmonics used specially designed harmonic cantilevers that enhanced one of higher harmonics [2], used specially designed torsional harmonic cantilevers [3], or enhanced higher harmonics by driving a conventional cantilever close to a submultiple of its resonant frequency [6]. Only Stark et al. [9] demonstrated that higher-harmonics imaging with the use of a conventional cantilever is possible, although they used the cantilever with a low resonant frequency to obtain higher harmonics within the limited bandwidth of the photodiode preamplifier and recorded higher-harmonics images consecutively.

1. The design of the microscope

The atomic force microscopy, AFM Terra (Fig. 1a), consists of three main parts: the measuring head, electronic control system and software, and an external PC.

The measuring head is a part of the apparatus, which is placed in a test sample, scanner, measuring amplifiers, current converters in four segments photodiode and pre-sample approximation to the probe (Fig. 1b). The measuring unit head comprises the following modules: a photodiode, semiconductor laser, surveillance cameras and photodiode positioning system, and a laser guidance system.

The measuring head assembly has two degrees of freedom in the horizontal plane in perpendicular directions. The AFM holder remains stationary relative to the sample, and its movement actuators perform the measuring assembly. In order to keep the compact dimensions of the laser, the beam track-measuring unit was devastated using a plane mirror, focusing the laser beam on the edge of the probe (cantilever). Two degrees of freedom allow the measuring unit to guide the laser beam on the cantilever.

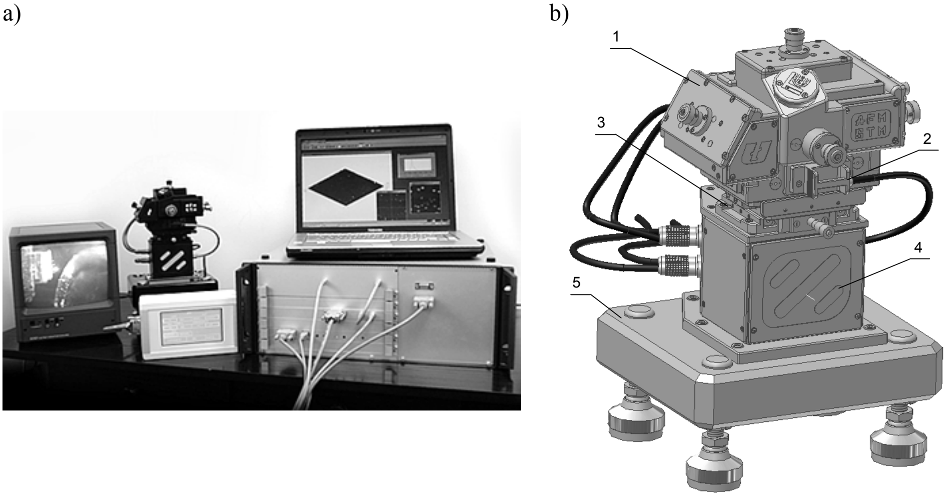

Fig. 1. Atomic force microscope Terra AFM (a) and view of a solid model of the head AFM (b); 1 – measuring unit, 2 – removable module AFM / STM, 3 – XY table, 4 – scanner, 5 – base

The photodiode also has two, independent of the measuring unit, degrees of freedom, enabling guidance of the photosensitive element reflected from the cantilever beam of laser light. The mixing head and a photodiode were implemented by using miniature micrometre screws.

The closure of the positioning mechanisms was implemented by using tension springs. The precise handling “wheelchair head” (the fitting cantilever) is provided by the use of miniature, backlash-free linear roller bearings. To avoid the effect of air movement, the measurement space was built inside the head, without the possibility of direct viewing of the sample and tip scanning. In order to observe the process of scanning and execute precise movements, preview actuators using two miniature CCD cameras were applied.

The measuring unit is placed on scanner unit (Fig. 2b). The structure of the scanner consists of exchangeable piezoelectric tube system, moto reducer, and the traverse. The arrangement of the piezoelectric tube is used for precise positioning of the test sample material. The tube is built on a base inside the sleeve serving as mechanical guards. The piezotube is finished with a magnetic table to fix the samples. The bracket system allows the use of interchangeable piezoelectric tubes of different lengths.

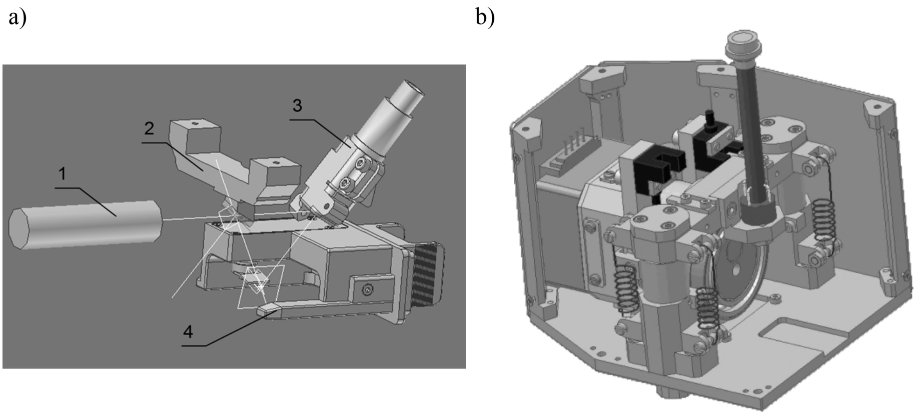

Fig. 2. View of the elements inside the measuring unit of the microscope (a) and scanner unit with the module of piezo tube (b); 1 – laser, 2 – handle with mirror, 3 – arm photodiode, 4 – truck head of the cantilever

The scanner system is used for the approximation of the initial pusher eccentric mechanism, in which the conversion of the rotational movement to the progressive movement of the eccentric tappet occurs. The components of the scanner were attached to a torsion box enclosure that provides adequate rigidity and mechanical protection of the precision scanner. As a part of the drive, a stepper motor cooperating with a miniature transmission wave and eccentric was used.

Modules placed in a cassette with the VME bus communication and operator panel create the electronic control system microscope. The operator panel is designed to facilitate the process of adjustment of the microscope optics for the user. Imaging parameters of this process is done with the touch screen LCD, working independently from the PC.

The modules of the control unit perform the following functions and devises:

- Proportional-integral controller ensuring the maintenance of a distance probe – sample (atomic scale) during the scanning process;
- The master controller and a communication system comprising a programmable FPGA, and a 32-bit microprocessor with peripherals; and,
- Providing a high voltage amplifier control voltages piezo-ceramic tube scanner ±225 V (for each, the four segments of the inner tube and its surface responsible for the shift in the direction of the axis Z).

In the loop proportional-integral regulator, due to the cooperation of electronic circuits to the mechanics, such as the scanning tube, measuring bar, and "optical lever", the feedback system was used with the parameters controlled in a wide range of gain and time constants. The loop uses a 12-bit resistive ladder (AD7547), which is able to increase the dynamics of the gain control at the level of 72 dB and adjust the time constant in the range from 10 s to 400 ms.

The controller also includes systems of differential amplifier input signals, the system pre-setting the initial scan based on the D/A converter (AD669), and a detection system that exceeded the threshold current for process control and automatic approximation of switching circuits and memorizing (AD7512 and AD585).

Terra AFM microscope software, implemented in an external PC, is primarily used for the analysis and presentation of the measurement data obtained. In addition, the software includes a virtual model of the microscope with a set of variables describing the current status of the device and the scanning process control algorithms.

2. Imaging system using harmonic vibration probe

For Terra AFM microscope working in tapping mode, a method of enabling simultaneous, synchronous measurement of both amplitude and phase of harmonics of the oscillating signal from the probe was developed (Fig. 3).

An algorithm for a Discrete Fourier Transform (DFT) is implemented in FPGA circuits. It calculates the amplitude value of the first (or the any) harmonic, which is fed to the feedback loop circuit in order to stabilize the operating point of the microscope. The computational complexity of the conventional DFT algorithm is $O(N^2)$. To reduce the computational time, most frequently the fast Fourier transform (FFT) algorithm is used for which the computational complexity is $O[N \log_2(N)]$. This algorithm is also used by us. To further reduce the computational time, one can use the recursive DFT algorithm, also called "the sliding DFT (SDFT) technique," which performs an N-point DFT on time samples within a sliding window. The SDFT initially computes the DFT of the N time samples. Then, the time window is advanced one sample, and a new N-point DFT is calculated. The large advantage of this process is that each new DFT is efficiently computed directly from the results of the previous DFT. The SDFT process can be described as follows:

$$S_m(n) = \left[S_m(n-1) + x(n) - x(n-N)\right]\left[\cos\left(\frac{2\pi m}{N}\right) + j\sin\left(\frac{2\pi m}{N}\right)\right]$$

where $S_m(n)$ is the new spectral component and $S_m(n-1)$ is the previous spectral component. The subscript m denotes that the spectra are related to the m-th DFT bin. The computational complexity of each successive *N*-point output for the SDFT algorithm is O(*N*) [10].

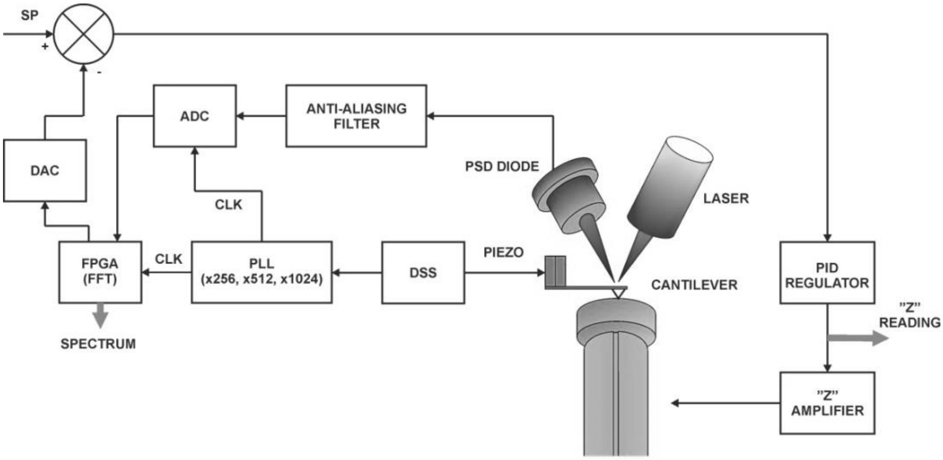

Fig. 3. Block diagram of the excitation and detection of digital vibration amplitude of the probe using the discrete Fourier transformation

Excitation of the probe vibration signal is generated by the digital direct synthesis circuit DDS, and it is also a programmable reference signal to a Phase Locked Loop (PLL), which is used to reproduce the clock signal frequency by a factor of *2n* (e.g., 64, 128, 256). The clock pulses produced in this way are used, in turn, for the timing of operating at speeds up to 100 MHz analogue-to-digital converters, as well as parts of FPGA logic resources that are responsible for reading the data and calculate the amplitude and phase of vibration probe. Since the sampling frequency is an integer multiple of the fundamental frequency of vibration of the measuring probe, the calculated frequency spectrum components using the DFT correspond to the actual harmonic amplitudes of the signal from the probe. This sampling method eliminates spectral leakage phenomenon known in digital signal processing and, in this case, there is no need to use windowing and, consequently, the introduction of correction factors to the calculated amplitudes of the signal components.

3. Imaging system verification

The proper operation of the system has been tested using a reference sample (HP-LDPE, Veeco). This sample consists of a mixture of polystyrene and polyolefin forming a thin layer applied to a silicon substrate. We used a Bruker RTESPA cantilever with a resonance frequency of 303.9 kHz and a spring constant 40 N/m. For convenience, the PLL chip was set to multiply the excitation frequency 256 times. As a result, the ADC and FPGA chip were working with a 77.8 MHz clock. The analogue PID controller was fed with an error signal proportional to the magnitude of the first harmonic of the fundamental oscillation signal determined by 256-point DFT. No additional averaging/post-processing algorithms were used. Sample images are shown in Fig. 4.

It is very well known that the image quality depends on the signal-to-noise performance and that the minimum acceptable signal-to-noise ratio is necessary to detect a given contrast level. The obtained images of the surface topography were of good quality. They possessed relatively high contrast and a low level of noise. This means that good signal-to-noise ratio was achieved. In comparison with other detection methods, good signal-to-noise ratios in connection with a high detection speed are specific and large advantages of the applied method of synchronous detection. As a consequence, this method is found to be very useful and well suited to applications in which oscillation modes are used. It is also worth noting that the application of advanced digital data filtration algorithms to the data obtained from the detection system will lead to further improvement of the signal-to-noise ratio.

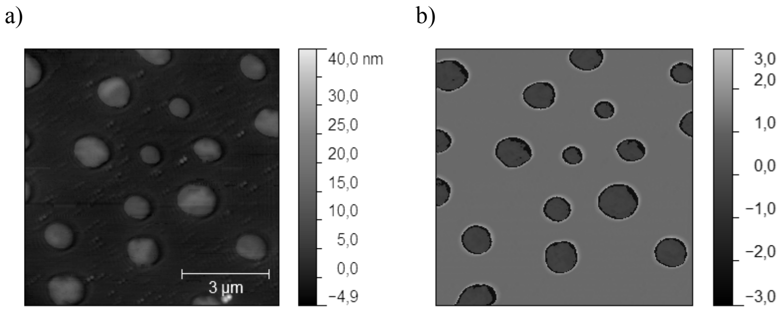

Fig. 4. The amplitude feedback – topographic image (a) and the phase feedback – phase contrast image (b) of reference sample

To test usability of the applied method of synchronous detection to simultaneously detect higher harmonics of the fundamental oscillation signal, we used the same reference sample. We used a Bruker V shaped silicon cantilever (type SNL 10) with a spring constant 0.12 N/m. The cantilever was driven at a fixed frequency close to the fundamental flexural resonance frequency 66.9 kHz.

The set-point amplitude was adjusted to 12.7% of the free amplitude. The second and third flexural resonance frequencies of the cantilever were measured to be 337.6 kHz and 920.7 kHz, respectively; that is, they are equal to 5.05 and 13.76 times the fundamental flexural resonance frequency of the cantilever, respectively. It is also to be noted that there was no measurable higher-harmonic content in the cantilever deflection signal when the cantilever was away from the sample.

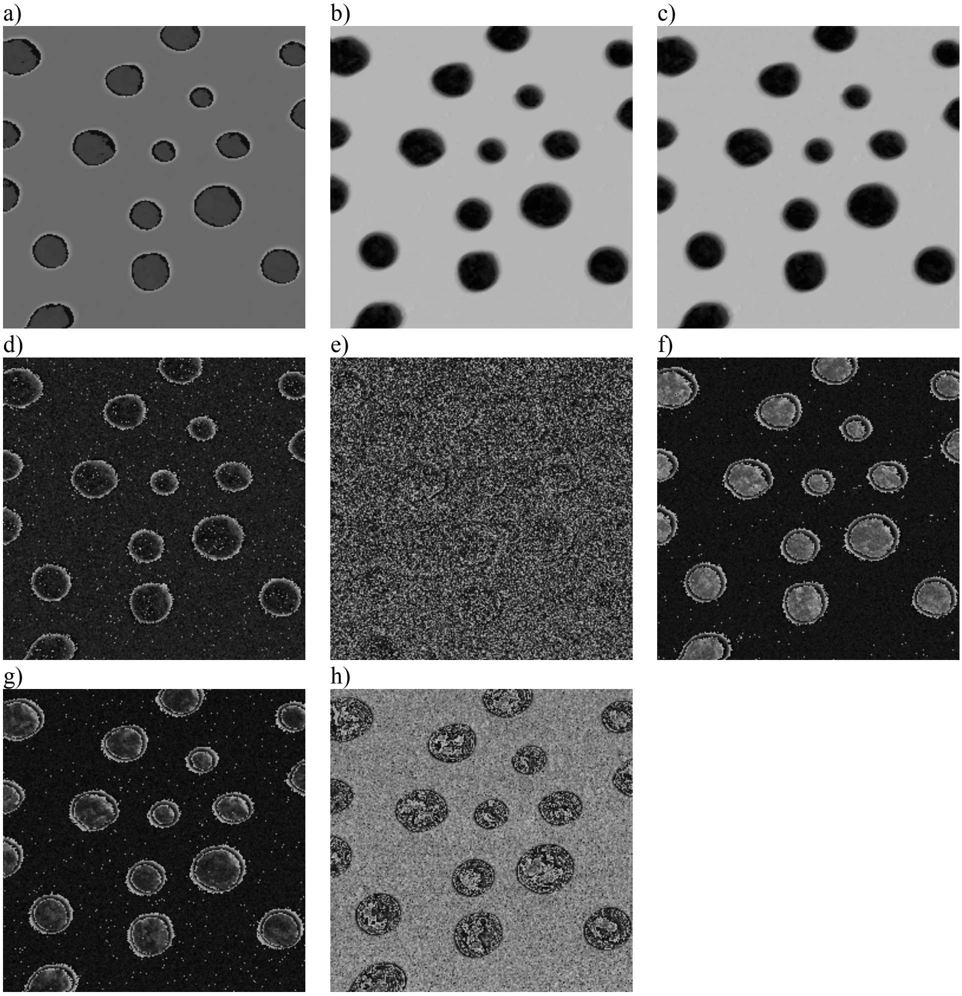

Fig. 5. The phase shift images of the same sample obtained with the reference system with detection module amplitude and phase of harmonics respectively: the first (a), a second (b), a third (c), fourth (d), fifth (e), sixth (f), the seventh (g), the eighth (h) harmonic

Fig. 5 present images of the same surface area of the PS–LDPE sample, recorded using the phases of the first 8 harmonics of the fundamental oscillation signal. In all these images, the PS and LDPE regions can be distinguished. Of all images in Fig. 5, those at the 1st (Fig. 5a), 4th (Fig. 5d), and 6th (Fig. 5f) harmonics are characterized by best signal-to-noise ratio and contrast. The images prove that the phase imaging at higher harmonics can be used to distinguish between different materials, even when a conventional cantilever is used. Note also, that for eight of them (Figs. 5d, f, g), the phase differences between the PS and LDPE regions are larger than that for the traditionally used phase image of the first harmonic (Fig. 5a), which shows the usefulness of the higher harmonics imaging for revealing mechanically heterogeneous regions of the sample. In the context of phase imaging at higher harmonic, it is worth noting that recently, in investigations of biological objects, Dulebo et al. [11] presented second-harmonic phase images revealing additional features at the nanometre scale that were not present in the first-harmonic phase images.

Imaging using the higher harmonics in the feedback control sample position *Z* allows one to carry out further work to clarify the correlation between those obtained images of the phase and mechanical properties of the tested structures.

Conclusions

The design of the Terra AFM microscope follows the changes and developments in the wider area of nanotechnology. Many measurement sessions performed by both authors and other researchers in scanning microscopy techniques allowed sampling to predict the most useful solution in the existing applications of the microscope.

The presented method using harmonic imaging allows simultaneous, synchronous measurement of both the amplitude and phase of harmonic signal from the oscillating probe. The used detection system, composed of 16-bit MSPS ADC and FPGA device, allows one to measure the amplitude and phase of the cantilever within one oscillation cycle and with good signal-to-noise ratio. As a result, good-quality images at higher harmonics could be obtained with the use of conventional cantilevers. In comparison with other detection methods, good signal-to-noise ratios in connection with high detection speeds are specific and large advantages of the applied method of synchronous detection.

Interpreting the nature of the contrast in phase images is difficult because of the multiple contributions to the energy dissipation [3]. Phase images at the 4th and 6th harmonics were characterized by the best signal-to-noise ratio and contrast.

The microscope with the analysis of harmonic imaging allows sensitive, real-world structures, without interfering in their construction and minimizing the impact of the gauge on the test object, and allows for deeper analysis of the mechanical properties of the test material.

Scientific work executed within the Strategic Programme "Innovative Systems of Technical Support for Sustainable Development of Economy" within Innovative Economy Operational Programme.

References

1. Majcher A., Mrozek M., Zbrowski A., Olejniczak W., Pawłowski S., Piskorski M.: STM/AFM microscope for application in industrial advanced technologies and the high schools education. Maintenance Problems 3/2011, pp. 177–188.
2. Sahin O., Yaralioglu G., Grow R., Zappe S.F., Atalar A., Quate C., Solgaard O.: High-resolution imaging of elastic properties using harmonic cantilevers. Sens. Actuators A 114 (2004), pp. 183–190.
3. Sahin O., Erina N.: High resolution and large dynamic range nanomechanical mapping in tapping-mode atomic force microscopy" Nanotechnology, 19, 445717 (2008), p. 9.
4. Cleveland J.P., Anczykowski B., Schmid A.E., Elings V.B.: Energy dissipation in tapping-mode atomic force microscopy. Appl. Phys. Lett. 72 (1998), pp. 2613– 2615.
5. Martínez N.F., García R.: Measuring phase shifts and energy dissipation with amplitude modulation atomic force microscopy. Nanotechnology 17 (2006), pp. 167–172.
6. Balantekin M., Atalar A.: Power dissipation analysis in tapping-mode atomic force microscopy. Phys. Rev. B 67, (2003), pp. 193–404.
7. Stark R.W., Heckl W.M.: Fourier transformed atomic force microscopy: tapping mode atomic force microscopy beyond the Hookian approximation. Surface Science 457 (2000), pp. 219–228.
8. Sahin O., Quate C.F., Solgaard O., Atalar A.: Resonant harmonic response in tapping-mode atomic force microscopy. Phys. Rev. B 69 (2004), pp. 165–416.
9. Stark R.W., Heckl W.M.: Higher harmonics imaging in tapping-mode atomic-force microscopy. Rev. Sci. Instrum. 74 (2003), 5111.
10. Lyons R.G. (Ed.): Streamlining Digital Signal Processing, IEEE Press, Wiley, New Jersey, 2007.
11. Dulebo A., Preiner J., Kienberger F., Kada G., Rankl C., Chtcheglova L., Lamprecht C., Kaftan D., Hinterdorfer P.: Second harmonic atomic force microscopy imaging of live and fixed mammalian cells. Ultramicroscopy 109 (2009), pp. 1056–1060.

System obrazowania wykorzystujący wyższe harmoniczne w trybie kontaktu przerywanego mikroskopu „Terra AFM”

Słowa kluczowe

Mikroskop sił atomowych, tryb kontaktu przerywanego, detekcja synchroniczna, obrazowanie fazowe.

Streszczenie

Mirroskop Terra AFM jest mikroskopem sił atomowych opracowanym i zbudowanym przez autorów jako urządzenie do zastosowań badawczych, przemysłowych i edukacyjnych w obszarze zaawansowanych technologii. W każdym mikroskopie sił atomowych pracującym w trybie kontaktu przerywanego oddziaływanie pomiędzy sondą i próbką ma charakter nieliniowy, co powoduje powstawanie wyższych harmonicznych częstotliwości podstawowej drgań sondy. W artykule przedstawiono system mikroskopu Terra AFM wykorzystujący metodę detekcji synchronicznej umożliwiającą jednoczesne wyznaczanie amplitudy i fazy wyższych harmonicznych przebiegu podstawowego. Głównymi elementami opracowanego systemu detekcji są przetwornik analogowo-cyfrowy o rozdzielczości 16 bitów i szybkości próbkowania 100 MSPS oraz układ programowalny FPGA pozwalający na pomiar amplitudy i fazy w okresu przebiegu podstawowego drgań sondy z dobrą wartością stosunku sygnału do szumu. Prowadzi to do otrzymywania dobrej jakości obrazów przy wyższych harmonicznych z użyciem typowej sondy mikroskopu AFM. Przedstawiono przykłady uzyskiwanych obrazów, które wskazują na przydatność systemu do rozróżniania obszarów próbek zbudowanych z różnych materiałów. Potwierdzają one również wysoką rozdzielczość przestrzenną (około 1 nm) opracowanego systemu.