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An alternative scheme to measure single-point hysteresis loops using piezoresponse force microscopy

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ABSTRACT

We present a simple and low cost procedure to obtain the electromechanical response, in a single-point, of non-conductive materials. The technique makes use of amplitude modulated voltage pulses in an atomic force microscope with standard configuration. Material response obtained, as signals of amplitude and phase from a lock-in amplifier as well as the input signal introduced to the conductive tip are stored in the AFM images, which act like a data acquisition system. The acquired data are processed with a free–program that eliminates the necessity of voltage pulses with constant time-widths, enabling the system to study the domain stability in piezo- and ferro-electric materials. We provide the electronic circuit diagram, flowcharts, and a free software for the implementation and execution of this procedure. Our goal is to provide an alternative scheme to measure the strain-hysteretic behavior of nonconductive materials without the need to invest in expensive software or AFM-moduli.

1. Introduction

Atomic force microscopy (AFM) is a technique that allows obtaining topographic information with high resolution in the micro to nanometer range [1]. Under electronic control and adequate ambient conditions, it also has the capacity to acquire quantitative information of diverse surface properties at nanometric level under different types of excitations such as voltage pulses [2–5], currents [6] or mechanical vibrations [7–9]. In particular, excitations with amplitude modulated voltage pulses (v-PAM) allow the detection of different physical or chemical phenomena such as ferro or piezo-electricity [3,4], charge injection [10], ionic conduction [11], motion of oxygen vacancies [12], surface cationic concentrations [13] and electrochemical reactions [14,15] through analysis of local strain- hysteretic behavior.

The v-PAM excitations have been used to compare the ferroelectric hysteresis loops obtained during the polarization switching in continuous and pulse modes [16]. In that work, they used the pulse mode to investigate the domain stability by changing the pulse widths. Later, Jesse et al. extended the use of the v-PAM excitations to acquire real-space imaging of imprint, coercive bias, remanent and saturation responses, and domain nucleation voltage on the nanoscale, in order to associate this data to the material microstructure [17].

Experimentally, in single-point experiments (*i.e.*, the tip does not scan the material's surface), the measurements of the strain-hysteretic signal of the material are made introducing v-PAM excitations through a conductive AFM-tip, as in switching piezoresponse force microscopy (S-PFM) [17,18]. However, those measurements require additional modules to the standard AFM configuration, to control the signal sent to the AFM-tip and typically a data acquisition (DAQ) system for the storing of the material's response; also, the data analysis sometimes demands expensive software, resulting in an additional investment cost.

This paper presents an alternative and simple procedure to obtain the local strain-hysteretic behavior such as phase switching and amplitude butterfly loops in single-point measurements of materials subject to amplitude modulated voltage pulses. The v-PAM is sent through the probe as in S-PFM [17,19]. However, in our proposal it is possible to apply v-PAM excitations with different pulse widths and, advantageously, store the strain-hysteretic information on the AFM standard images as in a typical topographic measurement, *i.e.*, this procedure does not need a DAQ system. For

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the analysis of strain-hysteretic AFM images, we developed free software in *JAVA* that gives the local hysteresis loops. Thus, this procedure allows studying domain stability in the case of piezo-ferroelectric materials [20,21] and possibly relaxation time in electrochemical phenomena [14,22]. The *JAVA* program is not limited to analyze strain-hysteretic AFM images; it also works with information from a DAQ system or data obtained with manually controlled pulses.

2. Principle of PFM and S-PFM

Ferroelectric materials present an intrinsic lattice polarization that can be reversed by means of the application of an external field, greater than the coercive field of the ferroelectric material. Ferroelectrics are also piezoelectric [23], *i.e.,* they present a mechanical deformation with the application of an electric field, phenomenon known as inverse piezoelectric effect. Ferroelectricity occurs because the crystal structure lacks a center of symmetry. Among ferroelectrics, oxides are the most studied and popular due to their mechanical robustness, which confers them a wide variety of practical applications [23].

Piezoresponse Force Microscopy measures the local electromechanical surface displacement of a ferroelectric material as consequence of an external sinusoidal *ac* signal sent though a conductive probe. The probe operates at a frequency close to its contact resonance to enhance the sensitivity and it acts as a movable upper electrode. To close the circuit, the ferroelectric material is generally deposited or grown on a conductive substrate or film, which functions as the bottom electrode. The electromechanical surface displacement is transmitted through the tip transforming into deflection or torsion of the cantilever. These probe movements are detected by the optical lever method, where a laser beam is reflected by the cantilever on a position sensitive four-quadrant photodetector. The deflection or torsion of the probe, *i.e.,* the out-of-plane or in-plane displacements, respectively, are acquired from the photodiode signal. As the tip slides across the ferroelectric sample, the photodiode detector follows the vibration of the probe and sends the signal to a lock-in amplifier where the signal is amplified and filtered using the frequency of the *ac* signal as a reference. The lock-in amplifier allows obtaining the amplitude and phase signals that are returned to the AFM system to form the typical PFM images.

To induce local polarization reversal, the small radius of curvature (R = 10–50 nm) of the probe is used as an advantage since a low *dc* voltage applied between the probe and bottom electrode generates a high electric field of the order of hundreds of kV/cm [24]. This electric field is usually higher than the coercive field of most ferroelectrics. By applying positive or negative *dc* voltage, it is possible to force ferroelectric domains to have opposite polarity. The reading process can be performed as typical PFM measurements.

In single point measurements using S-PFM, the amplitude and phase acquisition is similar to PFM. However, the electrical signal sent through the probe is composed of the typical *ac* signal, working at the contact-resonance frequency, but this is mounted on variable amplitude voltage pulses (*dc* signal) to produce local switching. A pulse consists of one-half with null value and the other with positive or negative *dc* value. The envelope of the signal resembles a triangular signal. The amplitude and phase signals have a dependence on the electrical signal introduced through the probe, resulting in the formation of ferroelectric hysteresis loops. The analysis of these hysteresis loops allows obtaining information on coercive bias, work of switching, d_{33} coefficient, domain stability, domain nucleation voltage, and so on [17].

3. Method description

3.1. Pulses applied through conductive probe and the obtaining of the strain-hysteretic signal

The voltages applied to the material *via* the AFM tip are produced with a function generator to form a v-PAM signal as the illustrated in Fig. 1a. Pulses can be of the same time width (τ_1) and amplitude increments (Δh) (see Fig. 1b), but advantageously in our analysis, it is not a requirement, and they can be different, as in Fig. 1c. For the final signal (the input signal applied to the tip), an *ac* signal is added to the generated v-PAM, forming the waveform schematized in Fig. 1d. In such case, a pulse dubbed the "ON-state" is followed by a no voltage application referred to as "OFF-state". The ON-state is used, for example, to study the domain dynamics of ferroelectric materials while the OFF-state minimizes the electrostrictive effect [24], and gives information about the domain stability [22]. The same pulse configuration is used in electrochemical strain microscopy measurements to study ionic motion and electrochemical reactions of non-ferroelectric materials [14,25].

The strain-hysteretic signal is obtained from the vertical deflection channel of the position-sensitive detector (PSD) and it is sent to a lock-in amplifier that uses the *ac* signal as a reference to give amplitude (R) and phase (θ). Then, R and θ are returned to the AFM system to form the corresponding images, rendering the use of a DAQ system unnecessary (at least in local measurements where only some cycles are taken at one point).

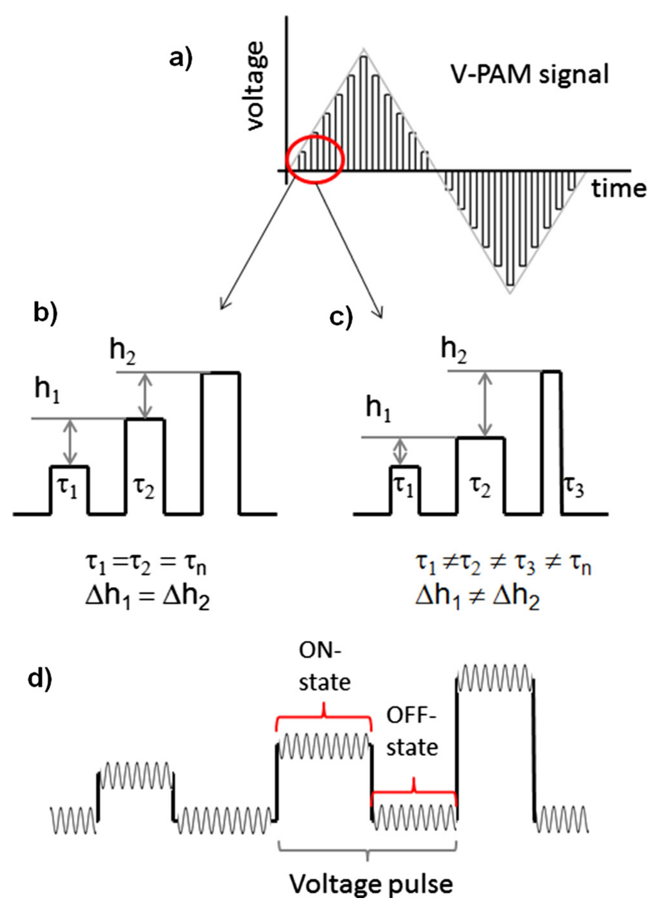

Fig. 1. (a) Waveform (cycle) applied to the AFM tip (v-PAM), Sequence of voltage pulses with (b) equal duration time and (c) different duration time. (d) Sinusoidal signal (*ac*) superimposed to the ON- and OFF-bias pulses.