Effects of cantilever buckling on vector piezoresponse force microscopy imaging of ferroelectric domains in $BiFeO_3$ nanostructures

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Systematic studies are presented on the effects of cantilever buckling in vector piezoresponse force microscopy (V-PFM) imaging of polarization domains in thin-film based (001)-oriented $BiFeO_3$ nanostructures, as observed through the coupling of out-of-plane and in-plane PFM images. This effect is a strong function of the laser spot position on the cantilever, being strongest at the free end, and insignificant at 60% of the cantilever length from the pivot point. This finding provides a unique approach to V-PFM imaging of ferroelectric polarization domains, yielding three dimensional PFM images without sample rotation in the plane. © *2010 American Institute of Physics*.
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Piezoresponse force microscopy (PFM)[1,2] is one of the most widely used scanning probe microscopy techniques to study piezoelectric/ferroelectric properties mainly due to its nanoscale spatial resolution, high piezoactuation sensitivity, and ease of implementation.[3–8] PFM detects the local electromechanical response of a ferroelectric sample by assuming that the induced AFM cantilever oscillation purely results from the sample surface oscillations due to the converse piezoelectric effect. In reality, a PFM image is actually a superposition of several artifacts that may arise during the measurements.[9,10] One of which, of interest in this work, stems from the cantilever buckling oscillations that occur when domains with in-plane (IP) polarization are aligned parallel to the long axis of the AFM cantilever.[11–13] Although the buckling motion of the cantilever is a result of IP piezoresponse, the output signal is strongly coupled in the out-of-plane (OP) PFM images. Surprisingly, despite the extensive use of this technique to probe samples with IP polarization, there has been no detailed experimental work to study the effects of cantilever buckling in vector PFM (V-PFM) images.

Depending on the relative orientations of the applied field and the polarization vector, the sample deformation can be in the form of elongation, contraction, or shear, which translates into the motion of the tip in contact with the surface. Domains with polarization in the OP direction induce vertical deflections of the cantilever (OP signal) as depicted in Fig. 1(a). For domains with an IP polarization, the resulting motion of the cantilever is dependent on the orientation of the polarization vector with the cantilever axis. For the case of domains with IP polarization direction perpendicular to the cantilever axis, the shear strain induces lateral oscillations, which are detected as torsion of the cantilever in the IP signal [Fig. 1(b)]. For the case of IP domain polarization parallel to the cantilever axis, the shear strain from the sample induces buckling oscillations, which are detected as OP signal [Fig. 1(c)]. This results in the coupling of the vertical and the buckling deflection of the cantilever to the OP signal, which can give rise to erroneous information in the PFM data. For ferroelectric thin films with IP polarization vectors, the buckling effect can be the dominant contribution to the total OP signal.

In this letter, we present a detailed study of the buckling effects of a cantilever during V-PFM domain imaging of a patterned $BiFeO_3$ (BFO) nanostructure where imaging was performed as a function of laser position along the cantilever, to investigate the buckling effect along the entire cantilever length. The sample consisted of an epitaxial (37 nm) BFO/(70 nm) $SrRuO_3$ (SRO) thin film heterostructure deposited by rf magnetron sputtering on a (001) $SrTiO_3$ substrate. The isolated structure was patterned directly on the blanket BFO film using the focused-ion beam technique; the process details are described elsewhere.[14] The PFM measurements were carried out in a commercial AFM (MFP-3D™, Asylum Research) using conductive diamond coated AFM tips, an ac bias of 17 kHz and 1 V_{rms} was applied to the tip, while grounding the SRO electrode.

Figure 2 shows the AFM topography and the corresponding PFM images of the BFO isolated structure. Typi-

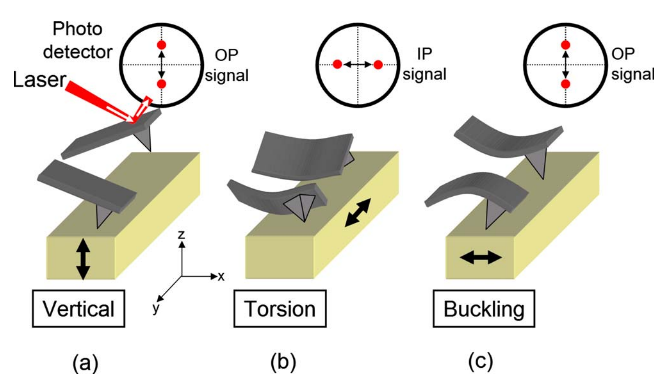

FIG. 1. (Color online) The three possible cantilever motions with respect to the orientation of the cantilever and polarization directions.

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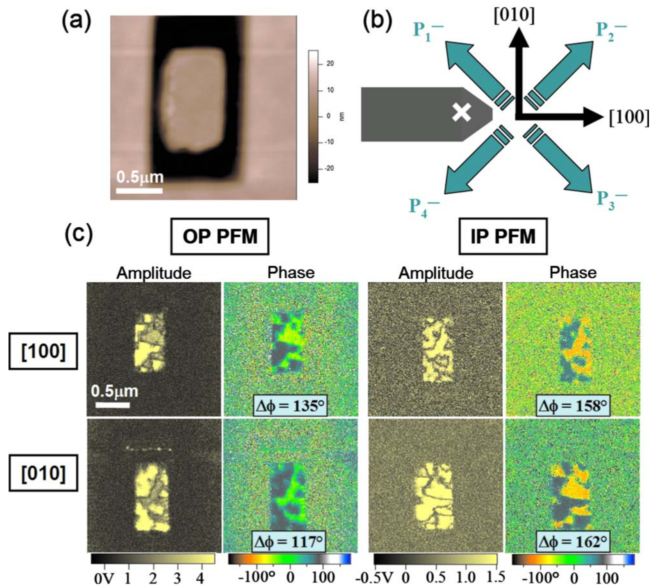

FIG. 2. (Color online) (a) Topography image of the $BiFeO_3$ nanostructure; (b) Schematic showing the cantilever alignment with the substrate orientation and the polarization directions of the BFO thin film, during PFM domain imaging. (c) IP and OP PFM images, obtained along the [100] and [010] directions.

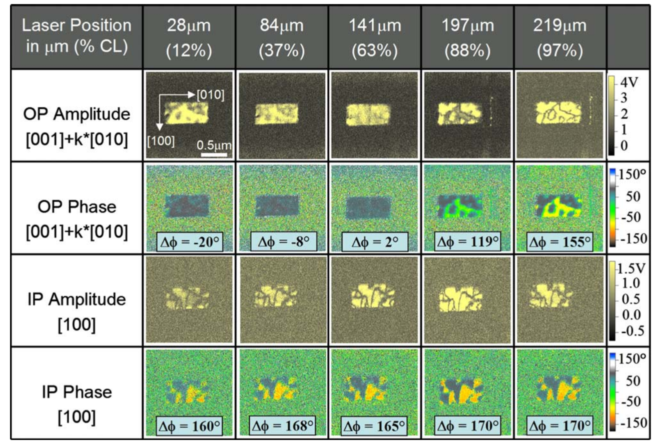

FIG. 3. (Color online) Montage of IP and OP PFM domain images taken along the BFO [010] direction with different laser spot positions along the length of AFM cantilever.

cally, to construct three-dimensional polarization maps of the BFO structure, a set of OP and IP PFM images are obtained with the cantilever aligned along two orthogonal directions. In this case, images were acquired along the substrate [100] and [010] directions [Fig. 2(b)].[9,11] The topography image [Fig. 2(a)] shows a structure with 1×0.5 μm^2 lateral dimensions surrounded by 40 nm deep trenches, therefore completely isolated from the surrounding BFO film. Comparing the [100] OP and IP PFM images, it is observed that there is no correlation between the two sets of images. This is also true for the [010] PFM image, which implies that there is little or no coupling between the OP and IP PFM images in the same direction.

On the other hand, comparing only the OP PFM images in both the [100] and [010] directions, there are two interesting observations that can be made. First, the [100] and [010] OP phase images show that there is a phase contrast of 135° and 117°, respectively, between the two opposite domains; this indicates that there are two vertical domain orientations within the BFO structure. Second, both PFM amplitude images show a nonuniform image contrast. Interestingly, when compared with the corresponding phase images, the amplitude image contrast is dependent on phase orientation, where domains that appear darker in the phase image show a brighter contrast in the amplitude image and vice versa. These two observations are found to be contradictory to earlier reports of PFM data on mosaic-like BFO films. It was found that most of the BFO films have polarization in one of the vertical directions. For the case of BFO films deposited on an SRO bottom electrode and with no top electrode layer, polarization should point down toward the SRO bottom electrode, due to asymmetric charge compensation at the bottom and top interfaces.[15] Therefore, only a single variant domain structure is expected in the OP PFM images. Furthermore, when comparing the [100] and [010] OP PFM images, it is found that the two sets of images are not the same, which was not the case in our previous report where the piezoresponse was independent of the scan direction.[14] However, when comparing the [100] OP with the [010] IP PFM images, a strong correlation is observed. Likewise, the [010] OP image resembles the [100] IP PFM image. The similarity between the images suggests a strong coupling between IP and OP PFM images, indicating that IP polarization dominates the OP PFM signal [due to the buckling effect shown in the schematic Fig. 1(c)]. The same measurements were carried out in the blanket BFO film region where a similar effect is observed, indicating that this effect is not unique to the isolated BFO structure.

In addition, it can be postulated that the buckling effect varies along the cantilever length. To study this effect, PFM images were obtained as a function of laser position along the cantilever axis. The cantilever has a total length of 225 μm, while the laser spot size is 55 μm in diameter. The PFM images in Fig. 3 were acquired as a function of the laser position with respect to the cantilever length, where the pivot point and free end of the cantilever are the 0 and 225 μm marks, respectively. Five sets of OP/IP domain images were obtained along the [010] direction from the 28 to 219 μm mark and for all laser positions the same deflection set point of 1.0 V was used during imaging.[16]

From comparison of the OP images, it is clear that the average phase image contrast between the two opposite domains in the nanostructures increases from 119° at 197 μm to 155° at 219 μm. As the amplitude image contrast becomes more similar between the opposite domains, domain walls appear as dark lines in the amplitude image. More interestingly, a comparison of the [010] OP PFM images at the 219 μm mark with [100] IP PFM images reveals that they are almost identical. Therefore, the buckling effect is most pronounced and would likely be further enhanced if the laser spot is moved closer to the cantilever free end. In contrast, it is found that the buckling effect decreased significantly when the laser spot is located at the 141 μm mark, where the PFM contrast between the two domains is only 2°. Here, there is no apparent domain structure in the amplitude image and the previously observed domain walls have also disappeared. The above observations raise two important points; (1) the contrast observed in images between 141 and 225 μm mark is entirely due to IP contrast, with negligible contribution from OP contrast; (2) for images close to the

141 μm mark, the IP contribution diminishes and only the OP contrast remains.

As the laser spot position is moved further inwards toward the pivot point, an interesting contrast reversal in both the amplitude and phase images is observed at the 84 μm mark, and becomes more obvious at the 28 μm mark. This shows that the phase orientation dependent contrast still exists between the 28 and 141 μm marks, except the effect is reversed. The presence of the contrast reversal suggests a model whereby the buckling effect is not detected when the laser spot is at the 141 μm mark. This is related to the fact that OP signal is sensitive to the cantilever angle,[10] not the cantilever displacement, and for the case at the 141 μm mark, the slope along the buckling lever is 0, independent of the IP amplitude. Closer to, or further from, the pivot point, the slope changes from positive to negative, or negative to positive, during each periodic voltage cycle, yielding strong phase shifts for images to either side of this point.

A comparison of the IP images acquired between the 197 and 28 μm marks shows that the IP PFM signal decreases as the laser position is moved toward the pivot point. This is expected, since the torsional motion of the cantilever is weakest near the pivot point. More importantly, although the IP signal is weaker, the contrast in the PFM images still remains constant. Note that the IP signal at the 219 μm mark is weaker than the 197 μm mark due to the fact that at the 219 μm mark 50% of the laser beam is not reflected back to the photodiode.

In conclusion, the disparity found in the OP PFM images is attributed to the difference in cantilever response distributed over its entire length due to the buckling effect. It is observed in OP images that the phase undergoes a 180° reversal when moving from the 197 to 28 μm mark and the fulcrum point of this vibration can be extrapolated to 140 μm mark. Therefore, the buckling effect is greatly reduced at this position, which is approximately 60% of the cantilever length from the pivot point. Also, it was found that the buckling effect is most dominant when the laser is aligned close to the free end of the cantilever. Finally, we have demonstrated that one can obtain the full set of three-dimensional PFM domain images without rotating the sample by acquiring images at different laser positions along the cantilever, which presents a unique approach to V-PFM.

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