Phys. Status Solidi RRL **5**, No. 9, 350–352 (2011) / **DOI** 10.1002/pssr.201105326

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Scaling effect of flexoelectric $(Ba,Sr)TiO_3$ microcantilevers

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Received 5 July 2011, revised 27 July 2011, accepted 28 July 2011

Keywords flexoelectricity, microcantilevers, $(Ba,Sr)TiO_3$, sensors

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The flexoelectric microcantilever offers an alternative approach for the development of micro/nano-sensors. The transverse flexoelectric coefficients μ_{12} of barium strontium titanate microcantilevers were measured at room temperature, and found to keep the same value of 8.5 μC/m for microcantilevers with thickness ranging from 30 μm to 1.4 mm. The calculated effective piezoelectric coefficient and electrical energy density of flexoelectric cantilevers are superior to those of their piezoelectric counterparts, suggesting that the flexoelectricity-induced polarization can be significantly increased as structures are scaled down due to the scaling effect of strain gradient, holding promise for flexoelectric micro/nano cantilever sensing applications.

Microcantilever structures were firstly designed for imaging in the atomic force microscopy (AFM) [1], and now have been used in numerous physical, chemical [2], and biological [3] sensing applications. Analogous to contact and tapping modes of AFM, microcantilever sensors can be operated by detecting the changes in resonance response or deflection, caused by mass loading, surface stress variation, or changes in damping conditions. Several microcantilever readout approaches with high precision were reported, including optical [4, 5], capacitive [6], piezoresistive [7] and piezoelectric [8] methods. Among them, the piezoelectric readout technique requires the deposition of piezoelectric materials, such as PZT and ZnO, on the cantilever [9], allowing for a more compact system and showing promise for microcantilever array applications. Piezoelectric cantilevers are also known for low-power consumption due to the high impedance and low driving voltage. However, piezoelectric thin films and nanostructures usually present low piezoelectric properties in comparison with their bulk counterparts, which is a challenge for piezoelectric microcantilever sensing applications. In order to further improve the microcantilever sensitivity, while retaining the advantages of piezoelectric readout, a flexoelectric microcantilever sensing structure was investigated and the initial results are reported in this Letter. The flexoelectric effect is the coupling between mechanical strain gradient and electric polarization described by

$$P_l = \mu_{ijkl} \frac{\partial \varepsilon_{ij}}{\partial x_k} , \qquad (1)$$

where P_l is the flexoelectric polarization, μ_{ijkl} the flexoelectric coefficient (a fourth-rank polar tensor with nonzero components μ_{11}, μ_{12}, μ_{44} in a cubic crystal), ε_{ij} the elastic strain, and x_l the position coordinate [10–12]. It was observed that all insulate solids possess flexoelectricity, but more significant flexoelectricity exists in materials with high nondispersive dielectric permittivity, like lead magnesium niobate (PMN), barium strontium titanate (BST), barium titanate (BT) and lead zirconate titanate (PZT), whose flexoelectric coefficients were measured by Ma and Cross [13]. Among these ferroelectric materials, $\mu_{12} \sim 100$ μC/m of BST at its Curie temperature is about one order of magnitude higher than those in PMN and PZT [13]. Resta [14] theoretically proved that the flexoelectric tensor is a bulk response of the solid, without surface contribution in the thermodynamic limit. Recently, Zubko et al. [15] reported experimental characterization of the complete flexoelectric tensor for $SrTiO_3$ (ST). Sharma's group made atomistic predictions of flexoelectric properties for both BT and ST

[16]. These recent works suggest a critial role of flexoelectricity in a variety of size-dependent physical phenomena related to ferroelectrics including enhanced size-dependent piezoelectricity and elasticity in BT nano-cantilever [17, 18], a strong indentation size-effect in BT [19], and a giant flexoelectric effect in ferroelectric HoMnO$_3$ epitaxial thin films [20]. However, at present there are limited experimental studies on micro/nanoscale flexoelectric structures such as microcantilevers, largely due to the nanofabrication challenges.

In this Letter we report on the fabrication and flexoelectric measurement of BST ceramic beams with the thickness down to micrometers. Our objective is to study the flexoelectric effect of microcantilevers, and to investigate the scaling effect of the effective piezoelectric properties induced by flexoelectricity and its potential for microcantilever sensing applications.

The BST ceramic beams (25 mm × 10.5 mm × 1.4 mm) with the composition of Ba : Sr = 65% : 35% were prepared by conventional solid state processing. The measured room temperature dielectric constant and dielectric loss were found to be 4100 and 0.3%, respectively.

The BST ceramic beams were then diced and lapped into different thicknesses. The samples were sputtered by 500 Å thick Cr, followed by 3000 Å thick gold, as electrodes. The bottom surface of the sample is fully covered with Cr/Au film, while electrodes of an area ranging from 1 mm^2 to 10 mm^2 were prepared on the top surface. The electrode design and typical electroded samples are shown in Fig. 1(a). The flexoelectricity measurements were carried out at room temperature with the experimental setup shown schematically in Fig. 1(b). The ceramic beam sample is rigidly clamped at one end. Transverse vibration of the beam is driven by a piezoelectric actuator. The driven voltage was supplied by a power amplifier (KH7602M) and a 2 Hz sinusoidal signal from a signal generator (AFG3000). The Polytec OFV-5000 laser vibrometer was used to measure the displacement at specific locations along the cantilever, from which the mode shape $w(x_1)$ can be obtained using the free beam mode theory. The transverse strain gradient along the thickness direction of the cantilever can then be calculated by:

$$\frac{\partial \varepsilon_{11}}{\partial x_3} = \frac{\partial^2 w(x_1)}{\partial x_1^2}\,, \tag{2}$$

where x_1 is the axial distance from the clamped end to the measurement point. The generated polarization P_3 can be calculated from the current, I, monitored by a lock-in amplifier (Stanford Research system, Model SR830):

$$P_3 = \frac{I}{2\pi f A}\,. \tag{3}$$

Here f is the driving frequency, and A is the electrode area. Based on the measured displacement and current, the polarization and the strain gradient and μ_{12} can be obtained using Eqs. (1)–(3). Figure 2 shows the measured μ_{12} of BST cantilever beams with different thicknesses. It can be observed that the flexoelectric polarization is proportional to the strain gradient, as shown in the inset of Fig. 2, where the slope remains almost unchanged when the dimensional size is scaled down. The μ_{12} value calculated from the slope of the inset in Fig. 2, was found to be about 8.5 µC/m, which is much lower than previously reported results (100 µC/m) obtained from cantilevers with thickness of sub-mm to millimeters [13], due to the low permittivity (4100) of the BST material at room temperature. Nevertheless, the confirmed scale-independent flexoelectric coefficient holds great potential for flexoelectric N/MEMS.

The effective piezoelectric coefficient, d_{33}^{eff}, of a simple flexoelectric BST cantilever beam can be expressed as [21]

$$d_{33}^{\text{eff}} = \frac{6\mu_{12} l^2}{E h^3}\,. \tag{4}$$

Here h is the thickness of the beam, E is the Young's modulus of BST ($E = 153$ GPa). One can see that d_{33}^{eff} is inversely proportional to the cubic of the thickness of the

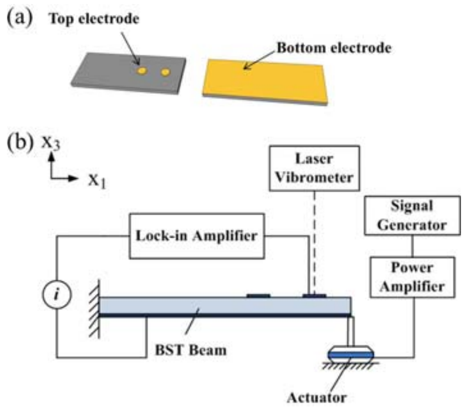

Figure 1 (online colour at: www.pss-rapid.com) (a) Typical electrode configuration. (b) Experimental setup for the measurement of flexoelectric coefficients.

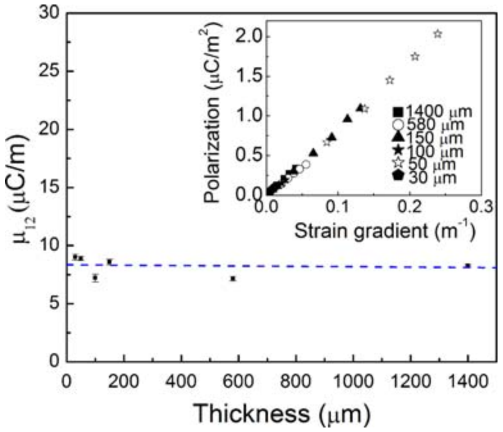

Figure 2 Transverse flexoelectric coefficient μ_{12} and the measured relationship between polarization and strain gradient (inset) in the BST microcantilever with different thicknesses.

beam, hence, it can be significantly enhanced when the beam thickness decreases to micro/nano-meter scale.

On the other hand, the d_{33}^{eff} of a bimorph piezoelectric bender subjected to an external tip force perpendicular to the beam can be expressed by the following equation [22]:

$$d_{33}^{\mathrm{eff}} = \frac{3d_{31}l'^2}{h'^2}\,. \tag{5}$$

where d_{31} is the transverse piezoelectric coefficient, l' is the length of the bimorph and h' is the thickness of the bimorph. The calculated d_{33}^{eff} of BST microcantilevers compared to those of ZnO, PZT (sol–gel, sputtering) and PMNT single crystal cantilever bimorphs, as a function of thickness h, is shown in Fig. 3 (the length-to-thickness ratio (l/h) was set to be a constant value of 50). Note that d_{33}^{eff} is different from the normalized effective piezoelectric constant calculated for BT by Majdoub [18], though similar size dependent effective piezoelectric properties can be observed. Piezoelectric coefficients of conventional piezoelectric materials are given in Table 1. It can be observed that the effective piezoelectric coefficient of flexoelectric cantilevers became higher than those of well-known piezoelectric bimorphs when the thickness of cantilevers is scaled down to sub-micrometers, which can be further increased using optimized BST ceramics (e.g. the highest reported $\mu_{12} = 100$ µC/m of BST [13]). Clearly, the significantly enhanced effective piezoelectric coefficient can be obtained with flexoelectric (FE) micro/nano-cantilevers due to the scaling effect of flexoelectricity.

In addition to the effective piezoelectric coefficient d_{33}^{eff}, another important effective piezoelectric property can be described as $1/2(d \times g)\,\sigma^2$ (refers to electrical energy density) under applied stress σ, where d is the (effective) piezoelectric coefficient (largest value for this material), g is the corresponding voltage coefficient. High electrical energy density is desirable for highly sensitive sensors. The electrical energy density of BST FE microcantilever ($h = 100$ nm, $l/h = 50$, $\mu_{12} = 8.5$ µC/m) can be 2×10^{-3} J/m^3, which is higher than that of piezoelectric cantilevers using the best known conventional piezoelectric material [25].

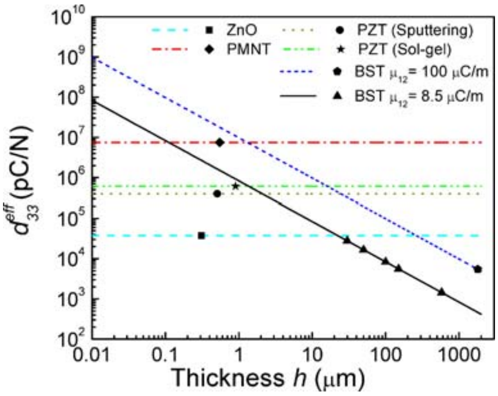

Figure 3 (online colour at: www.pss-rapid.com) Effective piezoelectric coefficients (d_{33}^{eff}) of piezoelectric bimorphs and flexoelectric microcantilevers as a function of thickness h under a constant length-to-thickness ratio ($l/h = 50$), see Refs. [13, 23, 24].

Table 1 Piezoelectric coefficient of different piezoelectric materials, see Refs. [23, 24].

piezoelectric materials	d_{31} (pC/N)
ZnO	–5
PZT (sol–gel)	–82
PZT (sputtering)	–53
PMN-PT	–1000

In summary, the scaling effect of flexoelectricity was studied using BST microcantilever beams with the thickness down to 30 µm. The measured transverse flexoelectric coefficient μ_{12} of ~8.5 µC/m remains constant for microcantilevers with various thicknesses. The calculated effective piezoelectric coefficient d_{33}^{eff} and electrical energy density of FE cantilever beams using the measured μ_{12} increase greatly with the decreasing beam thickness, promising for flexoelectric microcantilever sensing applications.

Acknowledgements This research is supported by Army Research Office (ARO) under grant W911 NF-10-1-0357. The authors appreciate the helpful discussions with Dr. William Clark.

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