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Enhanced rhombohedral domain switching and low field driven high electromechanical strain response in $BiFeO_3$-based relaxor ferroelectric ceramics

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ABSTRACT

A low-field driven high electromechanical strain response (large-signal d_{33}^* of ~1100 pm/V at 3.5 kV/mm) was found in a new (0.75-*x*)$BiFeO_3$-0.25$PbTiO_3$-*x*$Pb(Mg_{1/3}Nb_{2/3})O_3$ ternary relaxor ferroelectric ceramic. *In-situ* synchrotron X-ray diffraction measurements suggested that such a large strain be ascribed to a collective effect of electric field induced PNRs' growth, domain switching and rhombohedral-tetragonal (R–T) phase transition with increasing fields, among which ergodic PNRs' growth into R ferroelectric microdomains dominated the fastest increase of strains and the formation of the maximum strain hysteresis. Most interestingly, obviously enhanced R domain orientation along the electric field direction was believed to make a unique contribution as a result of its slightly reduced lattice distortion, compared with other Bi-containing relaxor ferroelectrics. An illustration of the domain morphology evolution in an ergodic relaxor was depicted to disclose the formation of strain hysteresis based on the delay in dynamics during electric cycling.

1. Introduction

Bismuth-based perovskite ferroelectric materials have attracted lots of attention in recent years either in the field of high-temperature piezoelectric materials [1–3] or large-strain ceramic actuators [4–9]. Particularly, giant strains of ~0.4% were reported in a few Bi-based perovskite relaxor ferroelectric systems such as A-site complexly occupied $(Na_{0.5}Bi_{0.5})TiO_3$ (BNT) based compositions [4–8], and B-site complexly occupied $Bi(Mg_{0.5}Ti_{0.5})O_3$ (BMT) based compositions [9,10]. The generation of large strains was usually found to be associated with the evolution of the dielectric relaxor behavior and mainly ascribed to the reversible phase transition from an ergodic relaxor state to a long-range ferroelectric state [5–11]. This mechanism has been recently updated by contributing the transition from polar nanoregions (PNRs) to short-range polar order, to the *c*-axis oriented growth of PNRs [12]. Unfortunately, the field strength required for triggering giant strains in these Bi-based perovskite ferroelectrics is very large (~6–8 kV/mm), such that the value of large-signal d_{33}^* (normalized strain S_{max}/E_{max}) is usually less than 600 pm/V.

As an important member of the Bi-based perovskite family, $BiFeO_3$ (BF) owns a high Curie temperature (T_c = 1100 K) and a rhombohedral (*R*) structure with an R_{3c} space group at room temperature (RT) [13,14]. A huge shift of Bi^{3+} and Fe^{3+} ions, a counter rotation of oxygen octahedra along the (1 1 1) direction and a similar electronic structure of Bi^{3+} to Pb^{2+} have enabled it to have intrinsically high spontaneous polarization [13,14]. A large polarization value of ~100 μC/cm^2 was reported for BF thin films due to the strain effect introduced by the lattice mismatch between the film and the substrate [15]. This strain effect was believed to transform the structure of BF films into a mixed phase state with stripe-shaped *R* phase embedded in tetragonal (*T*) matrix [16,17], resulting in a large electric field induced strain of over 5%. On the other hand, theoretical predictions based on first-principles calculations or *ab* initio calculations have shown that the large polarization value of BF is inherently high and relatively insensitive to strain [18,19]. This conclusion was later on proved by the measurements on good single crystals as well as bulk ceramics [20,21]. However, only a small strain (~0.07%) was obtained in BF ceramics measured at 60 kV/cm, *i.e.*, well below the coercive field of 100 kV/cm [22]. Recently, a large peak-to-peak bipolar strain of up

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