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Evolution of relaxor behavior and high-field strain responses in $Bi(Mg_{1/2}Ti_{1/2})O_3$-$PbTiO_3$-$Pb(Ni_{1/3}Nb_{2/3})O_3$ ferroelectric ceramics

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ABSTRACT

A composition-induced phase transition from a diffuse-type ferroelectric to a relaxor ferroelectric and then to a weak relaxor ferroelectric was observed in a new (0.68-x)$Bi(Mg_{1/2}Ti_{1/2})O_3$-0.32$PbTiO_3$-x$Pb(Ni_{1/3}Nb_{2/3})O_3$ (BMT-PT-xPNN) ternary system. Meanwhile, a gradual evolution process of nonergodic-ergodic-nonergodic realxor states was identified at room temperature from the BMT-rich side to the PNN-rich side. Two strain maxima of ~0.42% and ~0.29% under 7 kV/mm were obtained in the x = 0.2 and x = 0.65 samples, respectively. It is worthy to note that the high-field strain of the x = 0.2 composition was thermally stable but seriously hysteretic. By comparison, the x = 0.65 composition exhibited a weakly hysteretic strain and a relatively small threshold electric field. This phenomenon was attributed to larger sizes and slower dynamics of polar nanoregions (PNRs) in BMT-rich compositions than those in PNN-rich compositions. Moreover, it was believed that the strain hysteresis of BMT-PT-xPNN relaxor ferroelectrics would be dominated by the growth process of PNRs into ferroelectric microdomains during loading and their subsequent dissociation process during unloading.

1. Introduction

Ferroelectric materials that generate large electrostrains for actuator applications have attracted a great deal of attention in recent years. At present, the piezoelectric actuator materials have been dominated by conventional $Pb(Zr,Ti)O_3$ piezoelectric ceramics [1]. However, the strain value available in this class of materials is usually limited to ~0.1%, which would not be compatible for some applications where a large strain is valued. By comparison, relaxor ferroelectric materials have exhibited large potentials because of their exceptionally large strain values [2]. The achievement of large strains with low hysteresis in lead-based relaxor ferroelectrics such as $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) was believed to result from either domain engineering or electric-field induced phase transformation [2,3]. Apart from classical lead-based relaxor ferroelectrics, Bi-containing perovskite-type relaxor ferroelectrics such as $(Bi_{1/2}Na_{1/2})TiO_3$ (BNT), $Bi(Mg_{1/2}Ti_{1/2})TiO_3$ (BMT), and $BiFeO_3$ (BF)-based ceramics were also reported to have large electrostrains of up to 0.3–0.4% [4–7]. A systematic study on strain characteristics of Bi-based perovskite materials has been carried out in recent years [8–13]. BNT-based relaxor ferroelectrics usually exhibited a serious strain hysteresis and a large temperature sensitivity, and required high threshold electric fields for generating large strains [8,9]. The BF-based ceramics exhibited low driving fields and large strains of over 0.5%, but the temperature stability of the strain behavior still needed to be enhanced [10,11]. By comparison, the BMT-based systems had medium strains and strain hysteresis [12]. However, it would be possible to obtain temperature-insensitive or frequency-insensitive strains because of their specific domain structures [13]. In addition, a few attempts in achieving low threshold electric fields or good temperature stability have been also made recently in some groups [14–16].

Relaxor ferroelectrics are characterized by diffuse and frequency-dependent permittivity near the dielectric maxima. Polar nanoregions (PNRs) are considered to be an important microscopic feature and believed to play an important role in producing various macroscopic properties as a result of the existence of local random fields [17,18]. Upon cooling, the PNRs in an ergodic relaxor state can freeze into a nonergodic relaxor state near a critical freezing temperature T_f [18,19]. Large electrostrains have been reported in a couple of relaxor ferroelectric ceramics such as PMN [20], La doped $Pb(Zr_xTi_{1-x})O_3$ [21], and some Bi-based perovskites [4,5] near their T_f values because the coexistence of ergodic and nonergodic relaxor phases could offer similar free

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