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Relaxor-normal ferroelectric phase transition and significantly enhanced electromechanical strain behavior in $Bi(Ni_{1/2}Ti_{1/2})O_3$–$PbTiO_3$–$Pb(Mg_{1/3}Nb_{2/3})O_3$ ternary system close to the morphotropic phase boundary

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ABSTRACT

A pseudo-cubic to tetragonal phase transformation was found to be accompanied by a relaxor-normal ferroelectric phase transition in new perovskite-type $(0.65-x)Bi(Ni_{1/2}Ti_{1/2})O_3$–$0.35PbTiO_3$–$xPb(Mg_{1/3}Nb_{2/3})O_3$ ceramics. A morphotropic phase boundary was identified in the range of $0.4 < x < 0.6$ where the maximum quasi-static $d_{33} = 550$ pC/N was achieved. However, a large-strain platform of ~0.35% was generated under 6 kV/mm (dynamic $d_{33}^* = 583$ pm/V) within $0.15 \le x \le 0.4$. Particularly, the d_{33}^* and the strain hysteresis were found to be closely related with the $Pb(Mg_{1/3}Nb_{2/3})O_3$ content, field magnitude and signal frequency. The temperature-dependent measurement demonstrated that enhanced electrostrains appeared not only near a freezing temperature T_f, but also near an ergodic to ferroelectric phase transition temperature T_{fr}, both of which are ascribed to a reversible field induced ergodic to ferroelectric phase transition. An interesting finding would be that more similar free energies between ergodic and ferroelectric phases near T_{fr} have brought about smaller driving fields and strain hysteresis but lower maximum strains.

1. Introduction

Lots of attention has been in recent years given to the electric field induced strain behavior of a material for potential actuator applications. Apart from conventional $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) based ferroelectric materials, Bi-containing perovskite-type ferroelectric ceramics have also attracted much interest owing to their relatively high Curie temperatures (T_c) and good piezoelectric properties [1–3]. Particularly, giant electrostrains of up to ~0.45% were reported in a few Bi-based ferroelectric materials such as $(Bi_{1/2}Na_{1/2})TiO_3$ [4–7], $Bi(Mg_{1/2}Ti_{1/2})O_3$ (BMT) [8], $BiFeO_3$ [9,10] based solid solutions and so on.

Piezoelectric properties were known to be significantly enhanced in the proximity of a traditional morphotropic phase boundary (MPB) between two kinds of ferroelectric compositions of different symmetries such as rhombohedral (R) or pseudo-cubic (PC) phases and tetragonal (T) phases. In some cases, the composition induced morphotropic phase transformation from R or PC to T was accompanied by a relaxor-normal ferroelectric phase transition owing to the change in the degree of compositional inhomogeneity in perovskite lattices [11,12] or in the domain size or dynamics [13]. Flat free energy profiles of two ferroelectric phases close to the MPB would facilitate the polarization vectors to more sufficiently align along the direction of applied electric fields [14–18], thus leading to higher piezoelectric activities. However, the piezoelectric strain was usually limited within ~0.1% in most of PZT based solid solution ceramics probably owing to the clamping effect of grain or domain boundaries, although larger strain values are definitely necessary for actual applications. In additional to the electric field induced antiferrelectric to ferroelectric phase transition based on a volume effect between two different unit cells, giant electrostrains have been also reported in a couple of relaxor ferroelectric ceramics such as $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN) [19], La doped PZT [20], and some above-mentioned Bi-based perovskites [4–9] near a critical freezing temperature T_f where ergodic and nonergodic relaxor phases coexist [21]. A series of recent studies have demonstrated that the mechanism for such giant electrostrains could be attributed to a reversible phase transformation from a disordered relaxor state to a long-range ferroelectric ordered state driven by an external electric field [22–25]. Unfortunately, this kind

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