



Multiferroic and magnetoelectric properties of $\text{Ba}_{0.85}\text{Ca}_{0.15}\text{Zr}_{0.1}\text{Ti}_{0.9}\text{O}_3\text{-CoFe}_2\text{O}_4$ core-shell nanocomposite

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ABSTRACT

Lead-free magnetoelectric (ME) composites with remarkable ME coupling are required for the realization of eco-friendly multifunctional devices. This work demonstrates the ME properties of $\text{Ba}_{0.85}\text{Ca}_{0.15}\text{Zr}_{0.1}\text{Ti}_{0.9}\text{O}_3\text{-CoFe}_2\text{O}_4$ (BCZT-CFO) core-shell composites synthesized via co-sol-gel technique. Room temperature ferroelectric and ferromagnetic characterization have shown that the samples are magnetic and ferroelectric along with an adequate magnetoelectric coupling of 12.15 mV/(cm Oe). The strong dependence of electric parameters on applied magnetic DC bias fields demonstrated in ferroelectric and magnetoelectric measurements provide a framework for the development of potential magnetoelectric devices. Also, the high sensitivity of magnetoelectric coupling towards the applied AC magnetic field can be used for its application in magnetoelectric sensors.

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1. Introduction

The possibility of miniaturization of devices by incorporating multitasking materials is the impetus behind the research of multiferroic materials recently [1]; even though it was discovered long ago [2]. These materials possess two or more ferroic orders such as ferromagnetic, ferroelectric or ferroelastic and there is often coupling between these, which is known as magnetoelectric (ME) effect. This unique coupling mechanism gives an additional control to input and store data such as electric field control of magnetization and vice versa. The observed weak ME coupling in single phase natural compounds brings more interest in composite multiferroics especially piezoelectric-magnetostrictive composites [3]. Lead-based $\text{PbTiO}_3\text{-PbZrO}_3$ (PZT)-ferrite systems and lead-free systems such as (K,Na)NbO₃-ferrite, (Bi,Na)TiO₃-ferrite and BaTiO₃-ferrite systems have been extensively studied over the past decade [4–9]. Lead-based ceramics have been in the forefront for years due to their outstanding piezoelectric behavior ($d_{33} = 500\text{--}600$ pC/N) [10, 11]. The major reason for these enhanced piezoelectric properties is due to the existence of a morphotropic phase boundary (MPB), where three

phases coexist. Such enhanced properties are being attributed to the reduction in energy barrier near MPB for polarization rotation. But the global restrictions on lead-based compounds due to Pb-toxicity, urges the research world to find a better alternative to PZT. However the existing lead-free systems such as (K,Na)NbO₃ ($d_{33} \sim 250$ pC/N), (Bi,Na)TiO₃ ($d_{33} \sim 200$ pC/N) and BaTiO₃ ($d_{33} \sim 150$ pC/N) lacks an MPB similar to PZT and hence possesses much lower piezoelectricity [12–15]. Among these lead-free systems; BaTiO₃ is a perovskite oxide whose electrical properties can be altered by substituting in either “Ba” site or “Ti” site or both with Calcium, Zirconium etc. Calcium substitution in “Ba” site has shown a phase transition from rhombohedral to cubic while Zirconium substitution in “Ti” site leads to a tetragonal to cubic phase transition [16]. Liu and Ren have shown that an MPB can be achieved by combining $\text{Ba}(\text{Z}_{0.2}\text{Ti}_{0.8}\text{O}_3)$ and $(\text{Ba}_{0.7}\text{Ca}_{0.3})\text{TiO}_3$ systems ($50\text{Ba}(\text{Z}_{0.2}\text{Ti}_{0.8}\text{O}_3) - 50(\text{Ba}_{0.7}\text{Ca}_{0.3})\text{TiO}_3$ or $\text{Ba}_{0.85}\text{Ca}_{0.15}\text{Zr}_{0.1}\text{Ti}_{0.9}\text{O}_3$ (BCZT)) [17]. The high performance lead-free piezoelectric system, BCZT, was delivered a very high piezoelectric $d_{33} \sim 620$ pC/N, and a high permittivity ~ 3060 at 2 MHz, comparable to the ever market dominated lead-based PZT. Bao et al. presented another variant of BCZT, with a slightly higher critical temperature of 114 °C at the expense of a lower d_{33} (450 pC/N) [16]. Thus, BCZT system is a better alternative to the conventional lead based PZT systems. This offers wide possibilities in piezoelectric as well as

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