



# Laminate composite magnetoelectric multiferroics optimized by global derivative-free optimization method

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## ABSTRACT

The magnetoelectric multiferroics where magnetism and ferroelectricity coexist in one material have recently attracted renewed interest due to its potential applications in novel functional devices. Natural multiferroic single-phase compounds are rare and an alternative approach to obtain a magnetoelectric (ME) effect is through multilayered composites of a ferroelectric and a ferromagnetic material. An applied electric field creates a piezoelectric strain in the ferroelectric, which produces a corresponding strain in the ferromagnetic material and a subsequent change in magnetization. Various efforts to improve the value of ME coupling coefficient  $\alpha$  have been made by modifying preparation techniques of the samples, by the proper choice of materials or different structures and by choosing different thickness of the samples. In this study, we have applied numerical optimization for arriving at the solution for maximum ME coupling coefficient  $\alpha$  of a laminar ME composite by making use of the anisotropy of the ferroelectric phase. We have used a global derivative-free optimization method based in directional direct search coupled with specific multistart strategies for setting up the optimization problem. The effective ME coupling coefficients  $\tilde{\alpha}_{ij}$  are computed using the asymptotic homogenization method. Optimal composite microstructure with a range of the constituent ferroelectric single-crystal configurations that enhances the overall  $\alpha$  is identified. Optimal composite would have the [001]-axis of the ferroelectric phase oriented out-of-plane of the lamina. Yet the elasticity of the composite is found to be anisotropic at the optimal orientations of the ferroelectric phase. Stress-mediated enhancement of the ME coupling is demonstrated using the analysis of the inplane elastic stiffness of the composite.

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## 1. Introduction

Magnetoelectric multiferroics are increasingly being researched because they provide an additional feature of electric/magnetic control of magnetization/polarization. Magnetoelectric (ME) multiferroics are a new class of materials possessing simultaneously ferroelectric and ferromagnetic properties in the same phase exhibiting linear coupling and have recently drawn increasing interest due to their potential applications in multifunctional devices such as nonvolatile memory elements, magnetic field sensors and actuators, nano-electronics, energy harvesters, ferroelectric photovoltaics etc. [1–5]. Natural multiferroic single-phase compounds are rare and their ME coupling responses are either weak or occur at temperatures too low (since most multiferroics exhibit low Curie temperatures) for practical applications [6]. Alternately, ME effect can be obtained through fabricating compos-

ites of a ferroelectric and a ferromagnetic material in the form of thin film multilayered heterostructures, laminated, particulate or fiber–matrix composites etc [7–10]. The aim of this technique is to create materials that display the “product property” (that is the composite exhibits properties that are not present in the constituent phases) of ME coupling yet retaining properties of parent compounds. Here the coupling between ferroelectric and ferromagnetic order parameter is mostly mediated by an elastic stress/strain between the two phases. Ferromagnetic order manifests in the alignment even in the absence of an applied magnetic field, of local magnetic moments in a material that results in uniform permanent magnetization. Analogously in ferroelectrics, atomic displacements produce a uniform electric polarization even in the absence of an applied electric field [11,12].

Much research has been devoted in the area of composite ME systems since they exhibit more coupling by an order of magnitude than that of the single phase multiferroics [2,9,7]. Many tractable and tunable parameters such as the microstructure and stoichiometry are available in order to optimize the ME response in ME

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composites [13]. These systems operate by coupling the magnetic and electric properties between two materials, generally a ferroelectric material and a ferrimagnetic material, via the lattice (i.e. piezomagnetism couples to piezoelectricity). The induced magnetization  $\mathbf{M}_i$  is proportional to the applied electric field  $\mathbf{E}_j$  through the magnetoelectric coefficients  $\alpha_{ij}$ . It is defined [1] through its measurement via electric field( $\mathbf{E}$ )-induced magnetization

$$M_i = \alpha_{ki} E_k$$

and via the magnetic field( $\mathbf{H}$ )-induced polarization

$$P_k = \alpha_{ki} H_i$$

Here the tensor  $\alpha_{ij}$  corresponds to induction of polarization by a magnetic field or of magnetization by an electric field and is designated as the linear ME effect [14,13]. An applied electric field creates a piezoelectric strain in the ferroelectric, which produces a corresponding strain in the ferrimagnetic material and a subsequent piezomagnetic change in magnetization or the magnetic anisotropy [8].

Selecting materials possessing highly contrasting lattice parameters (thereby ensuring spontaneous immiscibility of phases) such as the perovskite-spinel system of electrostrictive and magnetostrictive phases in a laminar composite architecture could yield better ME coupling than single-phase materials or particulate composites [4]. Various efforts to improve the value of ME coupling coefficient have been made by modifying preparation techniques of the samples, by the proper choice of materials or different structures and by choosing different thickness of the samples [15,16]. Thanks to the composite structure and various connectivity options it throws up, the ME response can be tailored through the judicious choice of phase characteristics, volume fraction, shape and microstructure of the constituents. Several analytical and computational models have been developed to quantify the magnetoelectric coupling in ME multiferroic composites [17,18,2,19–22] consequent to the strain-mediated two-phase model proposed by Harshe et al. [7,23]. Models pertaining to superlattice films of ferroelectrics [24] or superlattices consisting of a combination of FE-paraelectric material [25] suggest that electrostatic coupling could be a design parameter to enhance dielectric properties. The electrostatic interaction, originates from bound charges inside the ferroelectric material, would be prominent in multilayer thin film systems where exist a polarization mismatch between the layers in the event of layer thickness being much smaller of the order of nanometers. Nonetheless, for composite laminate made by juxtaposing ferroelectric and ferromagnetic layers of large thickness, the electrostatic/magnetostatic interactions would be screened. Recent advancements in horizontal multilayered heterostructures present better opportunities for fabrication of highly ordered composite magnetoelectric systems which can deliver excellent ME coupling responses [26,27,8,28]. Yet, a proper computational experiment scouting all the available options for material design by the judicious exploitation of the underlying anisotropy of the component phases in a ME composite setting is still lacking. In this paper, we are optimizing the microstructure of a laminar ME composite of two ferroic materials (ferromagnetic cobalt ferrite,  $\text{CoFe}_2\text{O}_4$  and ferroelectric barium titanate,  $\text{BaTiO}_3$ ) that possessed contrasting lattice parameters. As another strategy to maximize the ME coupling, we search -by employing the method of computational optimization- for the optimal microstructural configuration that could enhance this effect especially utilizing the inherent anisotropy of the ferroelectric phase in composite multiferroics. Enhancement of the piezoelectricity by preferentially orienting ferroelectrics is well established [29–31] and this property could be crucial to the overall response of a composite where it is a component.

Mechanical stress linking the electrical and magnetic characteristics of the composites should be transmitted seamlessly between constituent phases. Sintered bulk samples of  $\text{CoFe}_2\text{O}_4$  with  $\text{BaTiO}_3$  magnetoelectric composites, in general, show ME coupling much smaller than the predicted values [18,7]. (“Bulk material” here refers to the macroscopic scale granular material distinct from thin films or nanoscale materials and hence it implies polycrystals). The reasons are attributed to the low resistivity of ferrites that eventually resists the building up of the poling field and the generation of leakage current through the sample that impedes the piezoelectrically generated charges. Apart from this, there exists deleterious problems inherent to bulk and particulate samples such as percolation due to interdiffusion and conduction and chemical reaction between the constituent phases, mechanical defects and diminished poling behaviour [13]. These issues can be overcome by juxtaposing and connecting two independent phases in horizontal layered (or laminated) geometries [32,18,7]. In this work, we have used a laminate composite configuration of multilayer  $\text{CoFe}_2\text{O}_4$ – $\text{BaTiO}_3$  (or BTO – CFO) system tapping the anisotropy exhibited by the classical ferroelectric  $\text{BaTiO}_3$  especially in its single crystalline phase [29,33]. Here we use a *universal anisotropy index* [34]  $A^U = 5(G^V/G^R) + K^V/K^R - 6$  to quantify the (elastic) anisotropy of  $\text{BaTiO}_3$ . Here  $K$  and  $G$  are the bulk and rigidity moduli and the superscripts  $V$  and  $R$  represent the Voigt and Reuss estimates.  $A^U$  is identically zero for a locally isotropic crystal. The departure from zero of  $A^U$  would provide a measure of the extent of anisotropy of a crystal. Voigt and Reuss estimates of  $K$  and  $G$  are calculated using Hill’s method [35] from the knowledge of measured elastic constants  $C_{\mu}$  of polycrystal [36] and single crystal [37]  $\text{BaTiO}_3$  for computing  $A^U$ . The anisotropy of single crystalline  $\text{BaTiO}_3$  is evidently manifested in the value of  $A^U = 1.33$  compared to near isotropic  $A^U = 0.01$  for polycrystalline  $\text{BaTiO}_3$ . The elastic anisotropy would lead to anisotropic distribution of strain field  $\mathbf{x}$  which inextricably permeates to the piezoelectricity through the relation  $\mathbf{x} = \mathbf{d}^t \mathbf{E}$ , where  $\mathbf{d}^t$  is the transpose of piezoelectric coefficient tensor and  $\mathbf{E}$  is the electric field. Since the lattice mismatch [38] between  $\text{CoFe}_2\text{O}_4$  and  $\text{BaTiO}_3$  is large ( $\sim 0.52$ ), it can give rise to an heteroepitaxial strain in ME thinfilms [26,39] or enhanced interface stress in other horizontal layered media [18,7]. For thin films, with increasing thickness the strain energy increases and the epitaxial layer would relieve it through introducing defects, irreversible deformation and surface roughening. After certain critical thickness ( $h_c$ , which is of the order of nanometers), the film goes to relaxed growth where the misfit strain energy is diminished [40]. However, for polycrystalline materials the misfit appears in areas where the film acquires texture and in that case the grain size matters. Nonetheless, in this work the microstructure of magnetoelectric composite is composed of laminae of micrometer size which is far greater than the critical film thickness (in a thinfilm setting). Thus the misfit strain energy would have marginal, if not little impact on the laminate model that is considered in this work.

Four major aspects govern the ME response of the laminated composites viz., the physical properties of the constituents, the respective thickness and number of the constituent layers, the orientation of the constituents and the type of bonding between the constituents [13]. In this study, we have addressed the former three of the four major aspects defining  $\alpha_{ij}$  except the bonding between the constituents. Here, we have applied a derivative-free optimization for arriving at the solution for maximum ME coupling coefficient  $\alpha$  of a laminar ME composite with the thickness and orientation of ferroelectric phase (i.e., BTO) as design variables. As the ME coupling  $\alpha$  could not be expressed explicitly as function of the afore-mentioned design variables though it inextricably

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