

Multiferroic magnetoelectric composite nanostructures

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Multiferroics are attracting increasing interest and provoking much research activity driven by the profound physics of these materials, the coexistence and coupling of ferroelectric and magnetic orders, and the potential applications in novel multifunctional devices such as sensors, transducers, memories and spintronics. Multiferroic magnetoelectric (ME) composite systems, such as ferromagnetic–ferroelectric heterostructures, which offer a novel route for integrating ferroelectric and ferromagnetism, have been widely studied in recent years. In these ME composite systems, ME coupling is strain-mediated, that is, the strain induced in one component, either by magnetostriction in the ferromagnetic or by the piezoelectric effect in the ferroelectric, is transferred to the other component, altering the polarization or magnetization. This article reviews the magnetic-field control of electric polarization and its converse effect, electric-field control of magnetization, in multiferroic ME composite nanostructures. The review focuses on three kinds of ME nanostructures: vertical heterostructures, horizontal heterostructures and particulate nanocomposite films. Theoretical approaches, such as Green's function methods, the phase field model and first-principles methods, have been used to simulate and predict the ME coupling effect in such nanostructures. Herein we briefly describe the potential applications of the ME nanostructured composites using representative examples, and outline the challenges and promising future for this field.

Research on the magnetoelectric (ME) effect, discovered more than a century ago [1], progressed through pioneering work in the 1950s and 1960s, and has now seen a resurgence driven by long-term technological aspirations. With the trends toward device miniaturization, there is ever-increasing interest in combining electronic and magnetic properties into multifunctional materials to produce a single device component that can perform more than one task. Multiferroic ME materials are particularly appealing not only because they have the properties of their parent compounds, but also because interactions between the magnetic and electric orders lead to additional functionalities.

From the viewpoint of material constituents, multiferroic ME materials can essentially be divided into two types: single-phase [2,3] and composite [4,5]. According to the original definition, a single-phase multiferroic material is one that possesses at least two of the 'ferro' properties, such as ferroelectricity, ferromagnetism or ferroelasticity. While ME composites are multiphase materials composed of different phases, neither phase supports the ME effect.

Magnetoelectric coupling describes the influence of a magnetic (electric) field on the polarization (magnetization) of a material. It may arise directly between the two order parameters as in single-phase multiferroics, or indirectly via strain/stress as in ME composites. Several recent articles have summarized and reviewed research progress in single-phase multiferroic ME materials [2,3], and accordingly, this review will instead focus on composite multiferroic materials.

The ME effect in composite materials is known as a product tensor property, first proposed by van Suchtelen in 1972 [5], that results from the cross interaction between the two phases in the composite. As illustrated schematically in Figure 1, the composite ME effect is a result of the product of the magnetostrictive effect (magnetic/mechanical effect) in the magnetic phase and the piezoelectric effect (mechanical/electrical effect) in the piezoelectric phase.

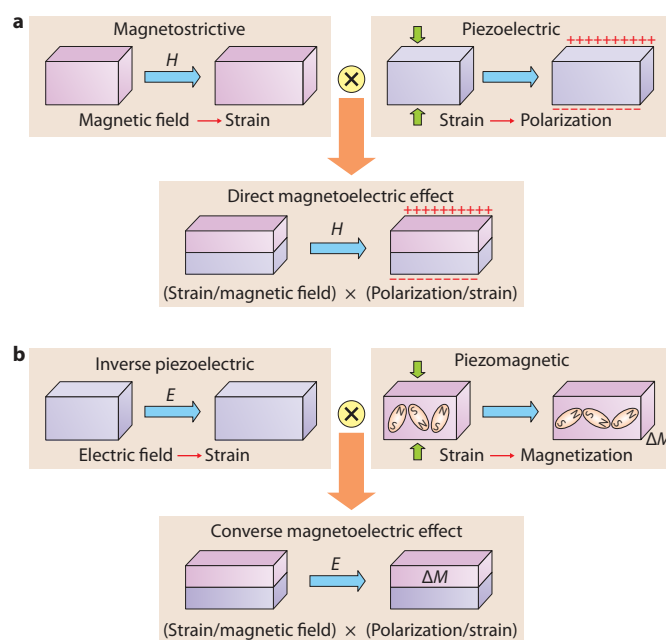


Figure 1. Schematic illustration of strain-mediated ME effect in a composite system consisting of a magnetic layer (purple) and ferroelectric layer (pink). (a) Direct ME effect: an in-plane magnetic field (H) induces strain in the magnetic component due to the magnetostrictive effect, which is mechanically transferred to the ferroelectric component inducing a dielectric polarization through the piezoelectric effect. (b) Converse ME effect: an out-of-plane electric field (E) induces strain in the ferroelectric component due to the inverse piezoelectric effect, which is mechanically transferred to the magnetic component, inducing a magnetization change (ΔM) or domain reorientation through the piezomagnetic effect.

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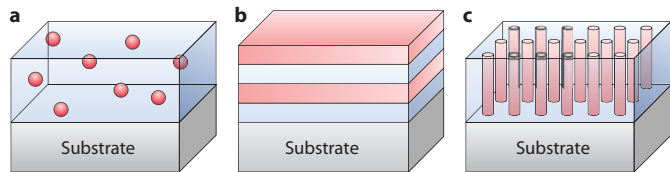


Figure 2. Schematic illustration of three kinds of ME composite nanostructures with common connectivity schemes: (a) 0-3 particulate nanocomposite films with magnetic particles (0) embedded in a ferroelectric film matrix (3); (b) 2-2 horizontal heterostructure with alternating ferroelectric (2) and magnetic (2) layers, or simply a ferroelectric (or magnetic) thin film grown on a magnetic (or ferroelectric) substrate; and (c) 1-3 vertical heterostructure with one-phase nanopillars (1) embedded in a matrix of another phase (3).

The composite ME effect can therefore be described as follows [6].

$$\text{Direct ME effect} = \frac{\text{Magnetic}}{\text{Mechanical}} \times \frac{\text{Mechanical}}{\text{Electric}} \quad (1)$$

$$\text{Converse ME effect} = \frac{\text{Electric}}{\text{Mechanical}} \times \frac{\text{Mechanical}}{\text{Magnetic}} \quad (2)$$

Many bulk ME composites have been found to exhibit such a strain-mediated ME effect above room temperature. Multiferroic ME films, in comparison with bulk ME composites, have some unique advantages. For example, ferroelectric/piezoelectric and magnetostrictive phases could be tuned and controlled at the nanoscale, representing a new scale for exploring ME coupling mechanisms. Furthermore, the two constituent phases in bulk ME composites are usually combined by co-sintering or adhesive bonding, which inevitably results in loss at the interface. In composite films, however, the different phases can be combined at the atomic level, and thus interface losses could be reduced significantly. By combining different phases with similar crystal lattices, epitaxial or superlattice composite films can also be designed, which facilitates the understanding of ME coupling at the atomic scale. Multiferroic ME films are therefore promising candidates for use in integrated magnetic/electric devices, such as sensors, micro-electromechanical systems, high-density memories and spintronics.

The renaissance of multiferroic ME films has recently been accelerated by advances in thin-film growth techniques, such as the pioneering work of Zheng *et al.* [7], supported by improved theoretical calculations [8–12]. The new growth techniques have provided routes to novel structures and phases, and allow the properties of traditional functional materials to be modified by strain engineering. Improved theory has aided in the design of new multiferroics, and helped our understanding of the coupling between magnetic and ferroelectric orders. The number of studies devoted to research on multiferroic ME films has increased remarkably in the last five years. Recently, Nan *et al.* [4] presented a comprehensive review of the historical perspectives, status and future directions of multiferroic ME composites, focusing on bulk ME composites. In the present review, the focus is on the experimental and theoretical progress in multiferroic ME films and nanostructures from the perspective of the ME effect, that is, magnetic field control of electric polarization and electric control of magnetization. Finally, we discuss the applications of the nanostructured multiferroic ME composites and summarize the challenges and promising future for research and technology in this field.

Magnetic-field control of electric polarization

The notation 0-3, 2-2, 1-3, etc. is used to describe the structure of a two-phase composite [13], where each number denotes the connectivity of each phase. For example, a 0-3 particulate composite is composed of

single-phase particles (denoted by 0) embedded in a matrix of another phase (denoted by 3), as shown in Figure 2. The common connectivity schemes examined so far include 0-3 particulate films, 2-2 horizontal heterostructures, and 1-3 vertical heterostructures of multiferroic ME composites consisting of ferroelectric and magnetic phases. Ferroelectric materials, including BaTiO₃ (BTO), PbTiO₃ (PTO), Pb(Zr,Ti)O₃ (PZT) and BiFeO₃ (BFO), and magnetic materials, including CoFe₂O₄ (CFO), NiFe₂O₄ (NFO), Fe₃O₄, La_{1-x}Sr_xMnO₃ (LSMO) and metals, with different nanostructures have been prepared by physical deposition techniques (*e.g.* pulsed laser deposition, PLD) and chemical solution processing.

Magnetic-field control of electric polarization can be described by the dynamic ME coefficient: $\alpha = \partial P / \partial H$ or $\alpha_E = \partial E / \partial H$, where P , E and H are the electric polarization, electric field and magnetic field, respectively, and α_E is most often used.

Particulate nanocomposite films

In 0-3 composites (Figure 2(a)), a magnetic phase is generally distributed in a ferroelectric matrix. For example, by employing a sol-gel method, Wan *et al.* [14] obtained 0-3 polycrystalline composite films with CFO nanoparticles dispersed in PZT. Zhong *et al.* [15] used Bi_{3.15}Nd_{0.85}Ti₃O₁₂ (BNT0) as the ferroelectric phase to constitute x BNT0/(1- x)CFO 0-3 polycrystalline films by a similar method. Both films exhibited ferroelectric and ferromagnetic behaviors, and ME coefficients were measured in both studies. Liu *et al.* [16] developed a modified sol-gel process to fabricate PZT/CFO composite films, and ME coupling between CFO and PZT in the film was demonstrated by external magnetic field-induced electric polarization change. However, as judged from the polarization–electric field hysteresis loops of the composite films, which do not exhibit a well-saturated shape, together with the influence of large leakage current, further confirmation is needed to determine whether the magnetic field-induced electric polarization change in fact reflects ME coupling in the composite films.

Murugavel *et al.* [17] prepared (100)-oriented PZT/NFO composites on (001) SrTiO₃ (STO) substrates by PLD. In their composite, NFO nanoparticles were randomly dispersed in the PZT matrix, and the measured magnetic field-dependent ME coefficient α_E showed similar saturation field-dependent behaviors to bulk composite ceramics. However, the maximum values of α_E for these composites, of the order of 10 mV cm⁻¹ Oe⁻¹, are lower than that reported for bulk PZT/NFO particulate composites [18]. This might be due to lattice clamping effects by the substrate. The sign of the ME coefficient is reversed between the two oppositely poled states, which demonstrates that the ME signals originate from ferroelectric domains with an inversion symmetry-breaking relationship.

Horizontal heterostructures

Horizontal nanostructures (Figure 2(b)) consisting of alternating layers of a ferroelectric perovskite and magnetic spinel usually exhibit only weak ME effects due to a large in-plane constraint from the substrate [9]. The ME effect could, however, be comparable to that in the bulk counterparts. Horizontal nanostructures are easy to fabricate and the leakage problem found in 0-3 and 1-3 composites may not apply due to the blocking of current flow by resistive ferroelectric layers. A direct ME effect could be observed in these materials. Due to their ease of processing and integration in devices, 2-2 heterostructures are the most widely investigated of the ME composite nanostructures [19–31].

Zhang *et al.* [20] reported a 2-2 heterostructure of CFO/BTO grown on a (001) STO substrate prepared by PLD. Their composite exhibited enhanced ME response, with an ME voltage coefficient of about 66 mV cm⁻¹ Oe⁻¹, comparable to that for bulk ceramic composites. To release the large clamping effect, He *et al.* [21] modified the interface between the ME composite and the substrate by using solution processing to add a LaNiO₃ (LNO) layer as a buffer and bottom electrode in a PZT/CFO layered structure. The results showed that LNO can induce a different preferential orientation in the PZT layer, thus leading to different ferroelectric behavior. The modification

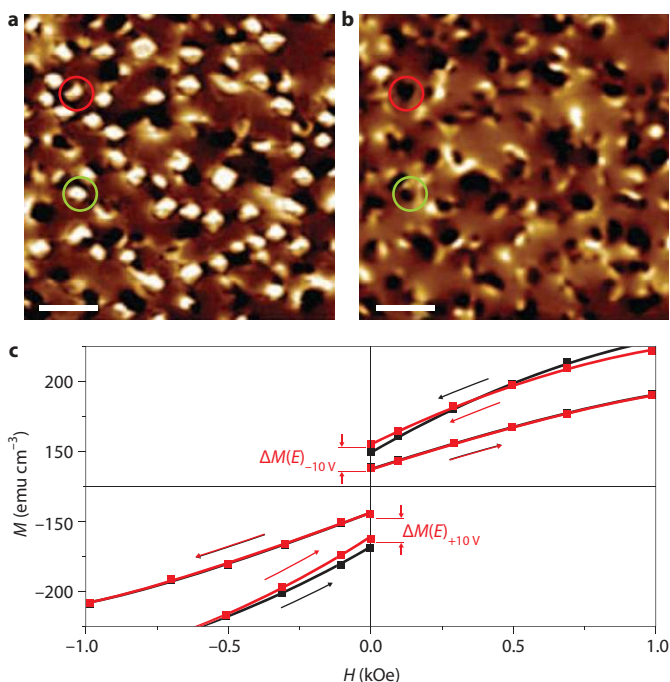


Figure 3. Changes in magnetic configuration of a $(\text{BiFeO}_3)_{0.65}/(\text{CoFe}_2\text{O}_4)_{0.35}$ film upon electrical poling. Magnetic force microscopy image taken (a) after magnetization in an upward-oriented 20 kOe perpendicular field, and (b) after electrical poling at +12V along the out-of-plane direction (scale bars, 1 μm). (c) Magnetization curves taken before (black) and after (red) electrical poling of $\sim 10\%$ of the total film area. Adapted from Ref. 39, reproduced with permission (© 2005 ACS).

effectively relaxed the constraint effect from the substrate and thereby enhanced the ME response of the thin PZT/CFO bi-layered film.

To explore the ME mechanism in the layered composite films thoroughly and quantitatively, more work on the influence of constraint stress on the ME effect is needed. Li *et al.* [22] developed a novel magnetic field-dependent Raman scattering method to study the strain-mediated ME coupling in horizontal heterostructures. The magnetostriction of the CFO layer generates stress that is transferred mechanically to the PZT layer, resulting in Raman mode changes. The observed magnetic-field-induced softening of the soft mode in the PZT layer bears a resemblance to direct ME output in the bilayers, providing evidence for a strain-mediated ME coupling mechanism in the multiferroic bilayers.

Similar research on layered ME nanostructures has been conducted by many groups. For example, PZT/ $\text{Ni}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ multilayer films reported by Ryu *et al.* [23], and $\text{Fe}_3\text{O}_4/\text{BaTiO}_3$ and PZT/ $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ bilayer films obtained by Ziese *et al.* [24] and Ma *et al.* [25], respectively, are all of high quality and exhibit coexisting ferroelectric and ferromagnetic behaviors. The ME coefficients in this composite films range from 3 to 30 $\text{mV cm}^{-1} \text{Oe}^{-1}$. Murugavel *et al.* [26] and Singh *et al.* [27] reported superlattice-based multiferroic ME films. Both the $\text{Pr}_{0.85}\text{Ca}_{0.15}\text{MnO}_3/\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ and $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3/\text{BaTiO}_3$ superlattices grown on single-crystal SrTiO_3 in these studies showed good dielectric and ferromagnetic behaviors, as well as magnetoresistance and magnetocapacitance effects.

A special, simple horizontal heterostructure has recently been constructed by growing an epitaxial ferroelectric PZT film on a manganite single crystal of LSMO [28], and conversely, by growing an epitaxial magnetic $\text{LaCa}(\text{Sr})\text{MnO}_3$ film on a ferroelectric single crystal (*e.g.* BTO and $\text{Pb}(\text{Mg},\text{Nb})\text{O}_3\text{-PbTiO}_3$) [29,30]. The efficient mechanical coupling across the interface results in a remarkable ME effect in such simple horizontal heterostructures. For example, Wu *et al.* [28] constructed a multiferroic heterostructure by growing a 110 nm-thick $\text{Pb}(\text{Zr}_{0.3}\text{Ti}_{0.7})\text{O}_3$ epitaxial film on single-crystal $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$. A strong

temperature-dependent ME effect was observed in this composite as a result of the magnetostrictive properties of the manganite crystal. The ME voltage in this composite was approximately 87% of the theoretical value predicted using a phenomenological thermodynamic model, and was estimated to be as large as $600 \text{ mV cm}^{-1} \text{Oe}^{-1}$ at 120 K. Wang *et al.* [31] later deposited BTO directly on CFO ceramics, which are cheaper and easier to process than costly single-crystal substrates. The ME coefficient of the CFO-based composite was about $38 \text{ mV cm}^{-1} \text{Oe}^{-1}$, which is one order of magnitude lower than that for epitaxial films grown on carefully selected single-crystal substrates. Thus, the improvement in strain transfer efficiency across the interface between the two phases is a key issue.

Theoretical modeling

In theoretical analyses, certain characteristics of the films, such as residual stress/strain resulting from the lattice misfit between the film and substrate and spontaneous polarization in epitaxial films, have been considered in order to understand the ME response in nanostructured films. As a result, the constitutive equations for the coupling magnetic–mechanical electric interactions in the nanostructured films can be expressed using a direct notation for tensors, as follows [9].

$$\sigma = c\varepsilon - e^T E - c\varepsilon^{\text{ms}} - \sigma_s \quad (3)$$

$$D = \varepsilon\varepsilon + \kappa E + \alpha H + P_s \quad (4)$$

$$B = \mu(\varepsilon, E, H)H + M_s \quad (5)$$

where σ , S , D and B are the stress, strain, electric displacement and magnetic induction; c , ε and μ are the stiffness, dielectric constant and permeability; and e is the piezoelectric coefficient. The parameters e^T and ε^{ms} are the transpose of e and the magnetostrictively induced strain related to the magnetic field-dependent magnetostriction constants of the ferromagnetic phase, respectively. The residual stress σ_s (or residual strain ε_s), spontaneous polarization P_s and magnetization M_s are included for multiferroic composite films and are not found in the constitutive equations for bulk composites [6]. The ME coupling interaction and magnetically induced polarization for nanostructured composite films calculated using Green's function and these modified constitutive equations [9] reveal that the 1-3 vertical heterostructures (Figure 2(c)) could exhibit an ME response larger than that in the bulk counterparts if there is no leakage problem. In contrast, the 2-2 horizontal heterostructures (Figure 2(b)) show much weaker ME coupling assuming a complete in-plane constraint effect. Liu *et al.* [32] has obtained similar results by Landau–Ginsburg–Devonshire phenomenological thermodynamic theory, and Petrov *et al.* [33] continues to model the ME interactions in ferrite NFO/PZT nanostructures in the form of nanobilayers, nanopillars and nanowires to further understand the size- and shape-dependent scaling behavior of the ME effect in nanostructured composites.

The phase-field model [34], which accounts for elastic energy in the constrained thin film, including the effect of the free film surface and the constraint of the substrate, has recently been developed for study of the ME coupling effect in 1-3 nanocomposite thin films [35]. Phase-field calculations of the magnetic field-induced electric polarization in a 1-3 BTO/CFO nanocomposite film gave similar results to the Green's function method, and illustrates that the magnetic field-induced electric polarization is highly dependent on the film thickness, the morphology of the nanocomposite, and the substrate constraint, providing a number of degrees of freedom in controlling coupling in nanocomposite films.

Electric field control of magnetization

In addition to magnetic-field control of electric polarization as discussed above, the converse ME effect—electric-field control of magnetization—is also of importance. Controlling magnetic anisotropy or the magnetization direction in a ferromagnetic material directly by applying

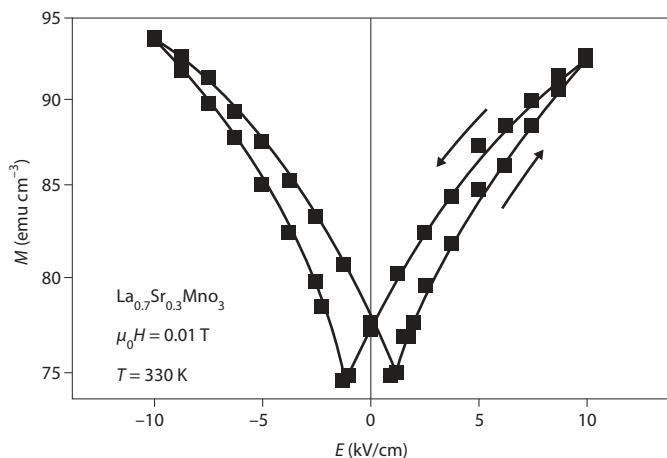


Figure 4. In-plane magnetization (M ; at 0.01 T and 330 K) of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ film grown on single-crystal PMN-PT (001) measured using a vibrating sample magnetometer with external electric field (E) applied across the PMN-PT substrate. Adapted from Ref. 30, reproduced with permission, reproduced with permission (© 2007 APS).

an electric field rather than current has become a main issue in the fields of spintronics and multiferroics. The electric field actuation of magnetic properties has been reported in the dilute magnetic semiconducting (Ga,Mn)As system [36] and ultra-thin ferromagnetic metal films [37,38]. However, this effect in these systems is limited to low temperatures (far below room temperature) and/or high electric fields (1 MV cm^{-1} or above). Alternatively, electric-field control of magnetic behavior can be realized by the coupling ME effect in multiferroic ME composite systems, either through a strain-induced ME effect across the interface [29,39–45], bias exchange [46–50], or a charge-driven ME effect [37,38,51].

Strain-induced ME effect

In multiferroic ME composite nanostructures, as shown in Figure 1 (b), an external electric field results in a change in the shape of the ferroelectric phase through the converse piezoelectric effect during switching. The strain in this case passes on to the magnetic phase, which alters the magnetic anisotropy of the magnetic phase via magnetostriction. Thus, electric-field control of the magnetic behavior of magnetic nanostructures can be achieved by strain-mediated ME coupling, as observed recently by many researchers [39–50].

Vertical heterostructures. In the BFO/CFO 1-3 vertical heterostructure film (Figure 2(b)), in which ferrimagnetic CFO nanopillars are embedded in a BFO film, magnetic force microscopy images (Figure 3) have shown that an electric field can change the magnetic configuration of the CFO pillars [39,40]. The strength of the ME coupling between the ferroelectric matrix and the ferrimagnetic nanopillars, given by the static perpendicular ME susceptibility $\alpha = \Delta M / \Delta E$, was estimated from the electric field-induced change of magnetization (ΔM) measured from the plots in Figure 3(c) to be approximately $1.0 \times 10^{-2} \text{ G cm V}^{-1}$ ($0.126 \text{ Oe cm V}^{-1}$). Here, the mechanism responsible for the electric field-induced magnetization switching involves the intimate lattice coupling between the two ferroic constituents of the nanocomposite, brought about by three-dimensional heteroepitaxial growth. In this case, when an electric field is applied to the piezoelectric matrix, its shape changes through the converse piezoelectric effect during switching, dynamically altering the magnetic anisotropy of the ferrimagnetic pillars via magnetostriction.

Horizontal heterostructures. For 2-2 horizontal heterostructures, Eerenstein *et al.* [29] demonstrated a large electrically induced magnetic change in an epitaxial heterostructure consisting of a ferromagnetic LSMO $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ film grown on a 0.5 mm-thick ferroelectric

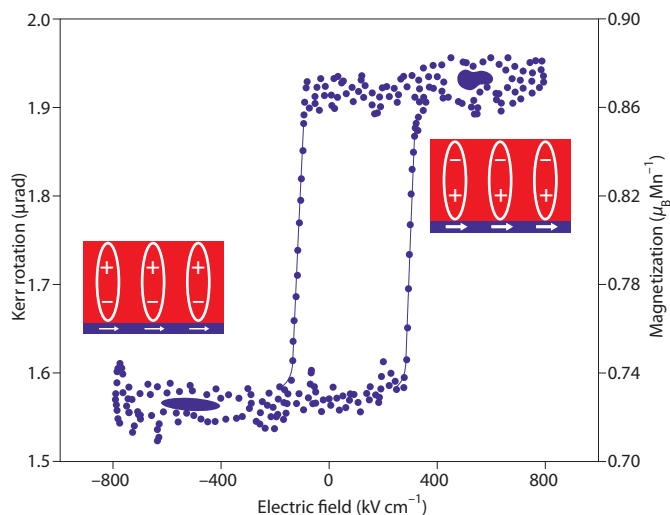


Figure 5. Magnetolectric hysteresis curve at 100K showing the magnetic response of a PZT (250 nm) on LSMO (4 nm) heterostructure as a function of electric field applied across the PZT film, as measured using a MOKE magnetometer. (Inset) Magnetic and electric states in the LSMO layer (blue) and PZT layer (red). The size of the arrows indicates the magnetization amplitude. Adapted from Ref. 51, reproduced with permission (© 2009 Wiley-VCH Verlag GmbH & Co. KGaA).

BTO substrate. Owing to the strain coupling via ferroelastic non- 180° BTO domains, the switching of BTO domains alters the local strain, magnetic anisotropy and thus the magnetization in the LSMO. In a composite of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ film on a single-crystal $\text{Pb}(\text{Mg},\text{Nb})\text{O}_3\text{-PbTiO}_3$ (PMN-PT) substrate, Thiele *et al.* [30] observed an obvious electric-field modification of magnetization (Figure 4). By applying an electric field to reversibly compress the isotropic in-plane lattice parameter of the PMN-PT substrate, a large strain-mediate ME coupling coefficient of $\alpha = \mu_0 dM/dE \leq 6 \times 10^{-8} \text{ s m}^{-1}$ ($0.06 \text{ Oe cm V}^{-1}$) at ambient temperature was derived from the M - E hysteresis loop in this magnetic-film/ferroelectric-substrate system.

Very recently, Liu *et al.* [41] prepared multiferroic composite structures of $\text{Fe}_3\text{O}_4/\text{PZT}$, $\text{Fe}_3\text{O}_4/\text{PMN-PT}$ and Fe_3O_4 with $\text{Pb}(\text{Zn},\text{Nb})\text{O}_3\text{-PbTiO}_3$ (PZN-PT) by spin-spray deposition of Fe_3O_4 ferrite film on ferroelectric PZT, PMN-PT and PZN-PT substrates. In the $\text{Fe}_3\text{O}_4/\text{PMN-PT}$ structure, a large microwave ME coefficient of 67 Oe cm kV^{-1} was observed. In the $\text{Fe}_3\text{O}_4/\text{PZN-PT}$ structure, a record high electrostatically tunable ferromagnetic resonance field range of 860 Oe with a linewidth of 330–380 Oe was demonstrated, corresponding to an ME coefficient of $108 \text{ Oe cm kV}^{-1}$. The static ME coupling of the multiferroic heterostructures was studied by electric field-induced changes in M - H hysteresis loops. Magnetization changes were caused by the electric field-induced strain due to the converse magnetoelastic effect. Therefore, electric-field tuning of magnetization is still realized in this system through strain-mediated ME coupling. Analogously, Yang *et al.* [42] observed sharp and reversible changes in magnetization under a electric field in a sample consisting of a CFO film on single-crystal (001) PMN-PT. The relative changes in M - E hysteresis loops were obtained for both the in-plane and out-of-plane magnetizations, suggesting that the electric field-induced change in magnetic anisotropy via strain plays an important role in the ME coupling in the CFO/PMN-PT heterostructure.

Instead of oxide-based ferromagnets, Sahoo *et al.* [43] deposited a 10 nm-thick film of ferromagnetic iron on a single-crystal BTO substrate. Large magnetization changes appeared in this system in response to ferroelectric switching and structural transitions of BTO under electric field and temperature control, respectively. Their studies provide additional evidence of alteration of the induced magnetic anisotropy in the horizontal heterostructures by interface strain coupling.

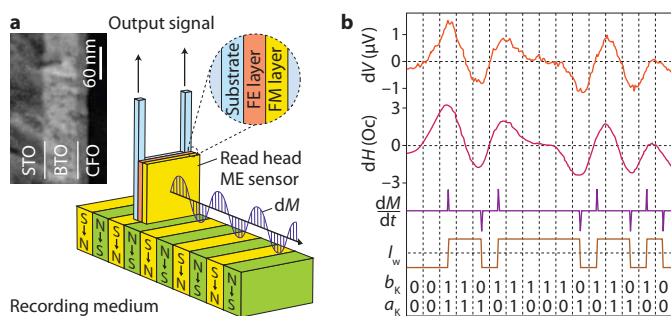


Figure 6. (a) Schematic illustration of an ME read-head sensor based on a bilayer CFO/BTO heterostructure. (Inset) A transmission electron microscopy image of the CFO/BTO heterostructure grown on a single-crystal STO substrate. (b) Voltage output waveforms (upper) for thin bilayer CFO/BTO films induced by different functions of δH at 1 kHz simulating the expected output voltage signals from a section of a magnetic recording track when using the ME read head. The bits b_k written with I_w apply the required δH on the head, inducing a response voltage δV following the pattern of the bits. Adapted from Ref. 72, reproduced with permission (© 2008 AIP).

Brandlmaier *et al.* [44] put forward a pathway for realizing strain manipulation of magnetic anisotropy in heterostructures consisting of a thin magnetite film and a piezoelectric actuator. Their results suggest that by optimizing the strain transfer efficiency and strain orientation, larger magnetization orientation shifts could be obtained. A magnetization orientation shift of 17° could be realized in the case of perfect strain transfer across the actuator–ferromagnetic film interface.

In addition to strain-mediated coupling, exchange bias has also been employed for electric-field control of the magnetic properties in magnetic films. The concept of using the exchange bias in multiferroic ME composite nanostructures derives from studies on the single-phase multiferroic BFO film, which acts as a ferroelectric antiferromagnet at room temperature [52], as well as work on the electric control of exchange bias in multiferroic materials. The exchange bias of multilayer $\text{Cr}_2\text{O}_3/(\text{Co},\text{Pt})_3$ can be electrically reversed but requires thermal cycling [46], whereas the exchange bias in YMnO_3 –permalloy heterostructures can be electrically tuned directly [47]. However, all of these results were observed at very low temperature.

The combination of multiferroic BFO with a ferromagnetic metal layer offers an exciting opportunity for modulating or controlling the magnetic structure by applying an electric field at room temperature. An electrically driven change in ferroelectric polarization and thus antiferromagnetic order through the ferroelectric–antiferromagnetic coupling in multiferroic BFO could switch the magnetization of an overlying thin ferromagnetic film (e.g. $\text{Co}_{0.9}\text{Fe}_{0.1}$ [48] and CoFeB [49,50]) by quantum-mechanical exchange. On the other hand, an electrically driven change in the electrical polarization of a ferroelectric that is also ferroelastic produces mechanical strain, which is transferred to an overlying ferromagnetic thin film. This deformation can also modify the preferred orientations of the magnetic dipoles and therefore the macroscopic magnetization [45]. Thus, in heterostructures consisting of BFO and thin ferromagnetic layers, strain-mediated coupling still exists. Studies in this area suggest the possibility of new device technologies, particularly in spintronic applications [53,54].

Charge-mediated ME effect

In 2-2 horizontal heterostructures containing ultrathin ferromagnetic films, an electric field could result in the accumulation of charge (e.g. spin-polarized electrons or holes) at the interface, which produces a change in the interface magnetization as a consequence of spin-dependent screening of the electric field. Recently, Weisheit *et al.* [37] observed a coercivity change of up to 4.5% in 2-nm-thick FePt films with the application of voltage. They attributed the electric field-induced modification of magnetocrystalline anisotropy to a change

in the number of unpaired d electrons under the applied electric field. However, a liquid electrolyte is required in order to apply a very high electric field at the surface in their system. Recently, Maruyama *et al.* [38] improvement on this by building all-solid-state ultrathin $\text{Fe}(001)/\text{MgO}(001)$ junctions in which a large change ($\sim 40\%$) in magnetic anisotropy could be induced by applying a high electric field (about 100 mV nm^{-1}). They tentatively attributed the effect to a change in the relative occupation of the $3d$ orbital of iron atoms adjacent to the MgO barrier.

Such a charge-driven ME effect can also be realized in multiferroic horizontal heterostructures containing ultrathin ferromagnetic films. A remarkable observation of the electric-field control of magnetism via the charge-mediated ME effect has been reported recently by Molegraaf *et al.* [51] in a bilayer $\text{PZT}(250 \text{ nm})/\text{LSMO}(4 \text{ nm})$ composite nanostructure. The charge-mediated ME coupling in this multiferroic nanostructure takes advantage of the sensitivity of a strongly correlated ultrathin magnetic film to competing electronic ground states. Resulting from direct control of magnetism via charge carrier density, the magnetic behavior of the nanostructure shows a hysteretic response as a function of the applied electric field (see Figure 5) with modulation of the LSMO magnetization as the PZT switches polarization. This M – E loop is demonstrative of cross-coupling between ferroic ground states, and affords a coupling coefficient of $\Delta M/\Delta E = 0.8 \times 10^{-3} \text{ Oe cm V}^{-1}$ at 100 K.

Theoretical modeling

Although the electric-field control of magnetization in ME composite nanostructures has been observed experimentally, our fundamental understanding of this phenomenon remains limited. By first-principles density functional calculations of a model Fe/BaTiO_3 horizontal superlattice, Duan *et al.* [21] demonstrated a ME effect that arises from an electronic hybridization between titanium and iron atoms that is not mediated by strain. The displacement of atoms at the interface caused by ferroelectric instability alters the overlap of atomic orbitals at the interface, which in turn affects the interface magnetization, resulting in an ME effect. A similar effect was found by Yamauchi *et al.* [55] for the $\text{Co}_2\text{MnSi}/\text{BaTiO}_3$ interface, and by Niranjana *et al.* [56] for the $\text{Fe}_3\text{O}_4/\text{BaTiO}_3$ interface. Rondinelli *et al.* [57] predicted that the ME effect at the $\text{SrRuO}_3/\text{SrTiO}_3$ interface may be mediated by free carriers, by which application of an electric field results in the accumulation of spin-polarized electrons or holes at the interface, producing a change in the interface magnetization. Very recently, Duan and colleagues [58,59] explored the effect of surface ME in response to the direct influence of an external electric field on the magnetic properties of ultrathin ferromagnetic metal films. Their calculations showed that spin-dependent screening leads to spin imbalance of the excess surface charge, resulting in notable changes in surface magnetization and surface magnetocrystalline anisotropy.

A nonlinear thermodynamic approach was recently employed by Pertsev [60] to model an ME effect via strain-induced spin reorientation transitions in ferromagnetic films. Based on the previous experimental observation that magnetization reorientation can be induced by tuning the film strains externally in many ferromagnetic thin films [61,62], Pertsev found that when such a strain-induced transition is caused by applying an electric field to a piezoelectric substrate, ME switching takes place. Such a converse ME effect is expected to be experimentally accessible in heterostructures such as $\text{CFO}/\text{PZN-PT}$ or $\text{Ni}/\text{PZN-PT}$. However, all of these results await experimental verification.

Measurements of the ME coupling effect

To measure the direct ME effect ($\alpha_E = \partial E/\partial H$) in ME nanostructures, it is common to record the dynamic change in voltage or charge induced by the application of a magnetic field parallel to the film plane. This is the same method as applied for bulk ME composites. Generally, there are two characteristic modes, active and passive, for measurement of

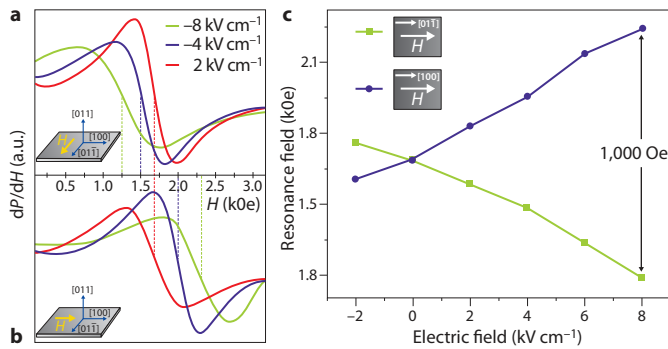


Figure 7. FMR absorption spectra for Fe_3O_4 film grown on single-crystal PMN-PT under various electric fields with an external magnetic field applied along the (a) tensional and (b) compressive directions of the PMN-PT substrate. (c) Generated resonance field change under an electric field with various magnetic field orientations (see inset). Adapted from Ref. 41, reproduced with permission (© 2009 Wiley-VCH Verlag GmbH & Co. KGaA).

the direct ME effect [31]. The active mode is similar to measurement of the magnetodielectric effect, and requires a test current to be passed through the sample to obtain the capacitive signal change induced by a magnetic field. Alternatively, a direct-current pulse test voltage can be applied to the samples to obtain the polarization signal change induced by a magnetic field, giving the polarization–electric field (P – E) hysteresis loop [16]. Thus when making ME measurements for composite samples in the active mode, by direct measurement of the P – E loop or dielectric constant change under an applied magnetic field, the contribution from the magnetoresistive and interfacial capacitive effects (or Maxwell–Wagner interfacial polarization effect [63]) must be considered.

In passive-mode measurement, no test current or voltage is applied to the sample. Instead, the ME voltage induced by a magnetic field applied parallel to the film plane is read directly (Figure 6). Thus, the magnetoresistive and interfacial capacitive effects appearing in the active mode would not contribute to the ME output signals measured in passive mode. When measuring the dynamically induced voltage in films by applying a small alternating-current (AC) magnetic field superimposed on a magnetic bias field, however, much care has to be taken to distinguish the real ME signal from the Faraday effect [31]. Thus, in ME composite nanostructures, the small magnitude of the ME response signals mean that measurement of the direct ME coefficient can be complicated by other parasitic effects such as leakage, interfacial polarization and faradaic effects, which could be a cause of the discrepancies among the ME coefficients reported in the literature.

By comparison, a variety of approaches have been developed to probe the converse ME coupling effect, that is, electric-field control of magnetization. A direct method is to use a vibrating sample magnetometer to measure the magnetization–magnetic field (M – H) behavior (see Figures 3(c) and 4). By applying an electric field to samples (typically out-of-plane), the change in M – H can be measured [29,39]. At high frequency, the magnetic tunability of ME composite nanostructures, for applications in tunable microwave devices, can be measured through electric field-induced changes in the ferromagnetic resonance (FMR) field (see Figure 7) [42]. Magnetic force microscopy and magneto-optical Kerr effect (MOKE) magnetometry have recently been used to probe the change in magnetic domains on application of an electric field (Figures 3(a,b) and 5) [38,39,51]. The MOKE observation (Figure 5) in particular is an easy approach for the study of electric field-controlled magnetism in ME nanostructures.

Applications

Bulk ME composites that exhibit a large ME effect at above room temperature and at low bias are promising for use in a range of technological

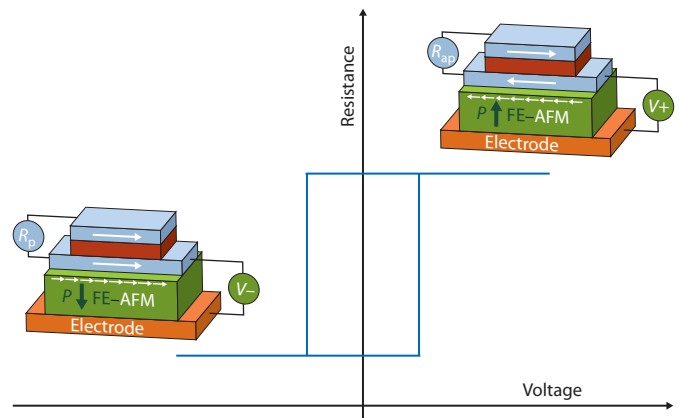


Figure 8. Conceptual sketch of a possible MERAM element. The binary information is stored by the magnetization direction (thick white arrows) of the bottom ferromagnetic layer (blue), is read by the resistance of the magnetic trilayer (R_p when the magnetizations of the two ferromagnetic layers are parallel), and is written by applying a voltage (V) across the multiferroic ferroelectric–antiferromagnetic layer (FE-AFM; green). If the magnetization of the bottom ferromagnetic layer is coupled to the spins in the multiferroic (small white arrows), and if the ME coupling is sufficiently strong, reversing the ferroelectric polarization (P) in the multiferroic changes the magnetic configuration in the trilayer from parallel to antiparallel, and the resistance from R_p to antiparallel (R_{sp}). This process achieves a hysteretic dependence of device resistance on voltage. Adapted from Ref. 76, reproduced with permission (© 2008 M. Bibes).

applications, including magnetic field sensors, transducers, filters, oscillators, phase shifters and memory devices [4,64–70]. Magnetolectric composite nanostructures similarly have potential applications as micro-devices in these fields. For example, magnetolectric composite nanostructures could be used as micro-sensors in place of traditional magnetoresistive read heads, which require a constant DC test current in order to detect the change in resistance/amplitude during operation. A novel magnetic recording read head based on the ME effect has been theoretically proposed by Vopsaroiu *et al.* [71]. The newly designed ME read head does not need a DC test current because data are read back directly as an induced ME output voltage. This highlights one of the clear benefits regarding the issues of power consumption and Joule thermal heating in high-resistance tunneling magnetoresistive sensors. Recently, Zhang *et al.* [72] demonstrated a prototype of such an ME read head based on bilayer CFO/BTO (Figure 6(a)). The output voltage waveforms of the bilayer CFO/BTO thin film follow the wave functions of the AC magnetic excitation signal closely (Figure 6(b)), illustrating the feasibility of multiferroic ME nanostructures for use in ME read heads.

The tuning of magnetization for magnetic devices has typically been achieved by applying a magnetic bias field using an electromagnet. Magnetolectric coupling in ME materials shows great promise for achieving electrostatically controllable magnetization, and novel, electrostatically tunable microwave magnetic devices such as filters, resonators and phase shifters based on ME composites have been proposed. For example, Liu *et al.* [41] considered the possibility of tuning magnetization in $\text{Fe}_3\text{O}_4/\text{PMN-PT}$ and $\text{Fe}_3\text{O}_4/\text{PZN-PT}$ composite films using an electric field (Figure 7(a)). They achieved a shift in the FMR absorption spectra of the composite films by applying an electric field. Changes in the spectral shifts according to the orientation of the external magnetic field have also been reported [42]: a total resonance field shift of $\delta H_r = 1,000$ Oe at 8 kV cm^{-1} was found when the external magnetic field was applied parallel to the [100] and [011] directions (Figure 7(b)), which could lead to the design of magnetostatically tunable magnetic devices with large effective magnetic field tunability.

Due to the hysteretic nature of the ME effect, ME nanostructures may also find applications in memory devices. The linear ME effect has a positive or negative sign depending on the annealing conditions (under parallel or antiparallel magnetic and electric fields). Thus, the coupling could, in principle, permit data to be written electrically and read magnetically in memory technologies. In the BFO/CFO vertical heterostructure, this could be achieved by combining a low, uniform magnetic field with the switching electric field to selectively switch CFO pillars with one magnetic configuration. Based on experimental results, Zavaliche *et al.* [73] proposed a concept and experimental setup for electrically assisted magnetic recording using ME nanostructures with high perpendicular magnetic anisotropy and an electric field to assist magnetic recording.

With multiferroics, the coexistence of several order parameters and ME coupling can be exploited simultaneously in novel types of memory elements [74–76]. As ferroelectric polarization and magnetization are used to store binary information in ferroelectric random access memory (FeRAM) and magnetic random access memory (MRAM), respectively, the coexistence of magnetization and polarization in a multiferroic ME material could allow the realization of four-state logic in a single device [75]. Beyond the combination of ferroic properties in a single device, the electrical control of magnetization via ME coupling offers the opportunity to combine the respective advantages of FeRAM and MRAM in the form of non-volatile magnetic storage bits that are switched by an electrical field. A sketch of a possible magnetoelectric RAM (MERAM) element was proposed by Bibes *et al.* (Figure 8) [76]. The basic operation of such an MERAM combines ME coupling with interfacial exchange coupling between a multiferroic material and a ferromagnet in a way that allows the magnetization of the ferromagnetic layer to be switched using a voltage [40]. The binary information is stored by the magnetization direction of the bottom ferromagnetic layer, is read by the resistance of the magnetic trilayer (when the magnetizations of the two ferromagnetic layers are parallel), and is written by applying a voltage across the multiferroic ferroelectric–antiferromagnetic layer.

Perspectives

Multiferroic ME nanostructures have become an important topic of ever-increasing interest in the past few years due to their compatibility with micro-fabrication techniques, which is a prerequisite for their incorporation into microelectronic devices. However, ME composite nanostructures are only a few years old, and thus there remain many open questions regarding ME coupling in the nanostructures. The first question that needs to be answered is physical: how do we control the identity, placement and function of every important atom in an ME nanostructure in ways that are practically applicable in real-world ME materials and devices? Controlling the growth of desired ME nanostructures with precise composition, atomic arrangements and interfaces is the most interesting issue but also a tough problem. Indeed, there have been ongoing quests in the field of nanomaterials and nanodevices, with recent progress also driven by directed assembly processes. High-quality horizontal heterostructures such as ME superlattices hold particular promise for the rational design of new multiferroics and their devices. The second question is conceptual: how do we understand systems that are too large to be handled by brute-force calculation, but also too small to be tackled by statistical methods? First-principles density functional calculations have proved to be a powerful tool in understanding and predicting the behavior of single-phase multiferroics, and such calculations could be carried forward to ME composite nanostructures.

Like bulk ME composites, sensors, transducers and heterogeneous read/write devices are among the suggested technical implementations of the composite ME effect in ME nanostructures. Recent preliminary experiments have shown the promising potential of novel devices based on the concept of ME coupling in ME nanostructures, and further detailed experiments are on the way.

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