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# Recent development and status of magnetoelectric materials and devices

## Yuxin Cheng, Bin Peng\*, Zhongqiang Hu\*, Ziyao Zhou, Ming Liu

Electronic Materials Research Laboratory, Key Laboratory of the Ministry of Education & International Center for Dielectric Research, Xi'an Jiaotong University, Xi'an 710049 China

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### ABSTRACT

The magnetoelectric (ME) materials and related devices have been attracting increasing research attention over the last few years. They exhibit strong ME coupling effect at room temperature, and electric field control of magnetization or magnetic field control of ferroelectric polarization can be achieved. The ME coupling effect brings novel functionalities to develop ultra-fast, low-power, and miniaturized electronics. Recent progress shows the performance of ME materials is further improved and the materials are used to develop many new types of electronics such as high-speed memory, radio frequency resonator, compact ME antenna, and weak magnetic field sensor. In this review, we present the overview in those fields with emphasis on both the opportunities and challenges for the application of ME materials and devices in the cutting-edge technologies.

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### 1. ME materials and operation mechanism

The research of the magnetoelectric (ME) saw a renaissance in 2005 [1] of multiferroic materials [2]. The ME possesses ferroelectric (FE) and ferromagnetic (FM) properties in the same phase and linear coupling effect [3] (see Fig 1). The multifunctional materials are fascinating for the coupling among many ferroic order parameters, and likely to provide another parameter of controlling more than one logic states for the whole new applications [4].

Among many generic ME material types, the single-phase multiferroic crystals exhibit the coexistence of magnetism and ferroelectricity and require long-range ordering of magnetic moments and electric diploes. BiFeO<sub>3</sub> is of great interest since it meets the requirement for many practical applications. Non-multiferroics material is another type to show ME properties and anti-ferroelectric (AFE) and anti-ferromagnetic (AFM) materials belong to this group such as Cr<sub>2</sub>O<sub>3</sub>. One outstanding feature of chromia is its operating temperature reaches above 40 degrees. The number of nonmultiferroics is rather small and only a few devices are produced within this type [6,7].

ME composites, on the other hand, consist of separated magnetostrictive and piezoelectric layers are more useful in a variety of applications. These composites typically show giant ME coupling

Corresponding authors

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Fig. 1. The Venn diagram for magnetoelectric and multiferroics [5]. The upper left circle shows ferromagnetic materials and ferroelectric materials are in the upper right circle. Hence, the overlap area of these two circles contains ME materials. The outer large circle is ferro- or ferri-materials.

response compared to those found in single phase materials [8]. Much efforts have been made to achieve strong ME coupling with ME composites, especially the modulation through strain mediated coupling [9], while another two ways of modulation are charge carrier and spin exchange. Of these three mechanisms, the research

E-mail addresses: pengbin@xjtu.edu.cn (B. Peng), zhongqianghu@xjtu.edu.cn (Z. Hu).

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Fig. 2. Magnetization variation as a function of applied electric field under 100 K. The insect shows the abrupt switches of magnetization and polarization [10].

on strain-mediated effects have been widely made and the studies of the other two are in the early phases [10].

### 1.1. Stain-mediated composite multiferroics

The strain-mediated composites formed by materials that show large magnetic and piezoelectric responses. The ME coupling occurs when strain induces converse piezoelectric effect in the magnetic phase or magnetostriction in ferroelectric phase. The first step is to translate the applied field to strain and then indirectly generate polarization in the composites by converting the strain. Since the coupling is the production of piezoelectric and magnetostriction effects, the giant ME effects require large piezoelectric coefficients for ferroelectric phase and large magnetostriction materials. Among the ferroelectric materials, BaTiO<sub>3</sub> and all PZT-based ceramics have been widely studied and the magnetic materials include Tb1-xDyxFe2 (Terfenol-D), Metglas and other ferrites are of great interests for practical applications.

To apply the material in real life, it is also important to con-38 39 sider the factors such as processing difficulty, strain transferring 40 connectivity, electrical resistance and chemical stability. Parameters such as permittivity, piezoelectric strain constant, mechanical qual-41 ity factor, magnetostriction, Curie and Neel temperatures should 42 be carefully reviewed. In general, the most important materials 43 44 properties include piezoelectric loss and piezoelectric voltage constant by piezoelectric materials and magnetostriction coefficient 45 for magnetostrictive material to achieve considerable ME response 46 47 in the composites.

### 1.2. Charge-mediated composite multiferroics

One challenge to realize efficient switching between different 52 magnetic states is to apply the electric field to change the charge 53 carrier density in the composites [6,7]. Charge carrier density can 54 be varied largely at the low voltages which proves to be a promis-55 ing method for future energy-efficient devices. The applied field 56 can control many different material properties such as supercon-57 ductivity [8,9] and ferromagnetic metals [10]. The Fig. 2 shows the 58 magnetoelectric hysteresis curve responding to the applied E-field in the PZT/LSMO system at 100 K. The insects illustrate the mag-60 netic and electric direction for the two layers respectively.

61 The modulation of multiferroic magnetism properties with ex-62 ternal electric field is well explored including coercivity and critical 63 temperature, the direct modulation of magnetization is still in the 64 relatively early research step. In Fig. 3, Zhou demonstrated that 65 the FMR response of in CoFe/BSTO heterostructure shifts at differ-66 ent applied voltage [11]. Several groups also carried out the first



Fig. 3. Ferromagnetic resonance field vs. applied voltage in CoFe/BSTO heterostructure, from [11].



Fig. 4. The application of electric field changes polarization of FE layer, through the FE/AFM and AFM/FM coupling, finally changes the FM layer magnetization.

principle calculation of ME coupling in Fe [12,13], Fe<sub>3</sub>O<sub>4</sub> [14], and SrRuO<sub>3</sub>/BaTiO<sub>3</sub> [15].

### 1.3. Exchange-bias mediated composite multiferroics

Exchange interaction is a well-known concept and used in many magnetic applications such as next generation storage [11, 12]. As is shown in the Fig. 4, the exchange bias device consists of a FE/AFM layer contacting the ferromagnet. The exchange bias effect occurs at the interface of these two layers when there is a strong magnetic exchange and the spins of FM layer are pinned by the ferromagnet [4]. For these systems, the electric field changes the polarization of FE layer, and afterwards through the coupling of FE/AFM changes the AFM layer magnetization. Subsequently with the exchange bias between AFM and FM layer changes the FM layer state. Therefore the coupling might be potentially used to control FM magnetism with electric field through the exchange anisotropy effect [13]. The exchange anisotropy is induced by cooling the heterostructures under the Néel temperature and keeping the FM layer saturated. FeCo/BiFeO<sub>3</sub> system shows that ferroelectric domain wall of BiFeO<sub>3</sub> has effect on the exchange bias [14]. The same effect was also observed in NiFe/YMnO<sub>3</sub> where the large change in exchange bias occurs under different values of electric field [15].

### 2. Magnetoelectric devices and applications

Bulk structures and fine thin-film ME composites show strong 124 ME coupling according to [16]. Based on the ME coupling mech-125 126 anism, ME applications can be classified into several groups as 127 shown in Table 1. Direct ME coupling or magnetic field controlling polarization are used in devices such as sensitive sensors [17], 128 energy harvesters [18]. For the converse ME coupling, E-field con-129 trol of permeability and E-field control of spintronics enable the 130 131 voltage tunable inductors [19]; bandstop filters [20], tunable res-132 onators [21], respectively.

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