Perspectives of voltage control for magnetic exchange bias in multiferroic heterostructures

Q. Yang\textsuperscript{a}, Z. Zhou\textsuperscript{a}, N.X. Sun\textsuperscript{a,b}, M. Liu\textsuperscript{a}

\textsuperscript{a} Electronic Materials Research Laboratory, Key Laboratory of the Ministry of Education & International Center for Dielectric Research, Xi'an Jiaotong University, Xi'an 710049, China
\textsuperscript{b} Electrical and Computer Engineering Department, Northeastern University, Boston, MA 02115, USA

\begin{abstract}
Exchange bias, as an internal magnetic bias induced by a ferromagnetic–antiferromagnetic exchange coupling, is extremely important in many magnetic applications such as memories, sensors and other devices. Voltage control of exchange bias in multiferroics provides an energy-efficient way to achieve a rapidly 180\degree determinisitic switching of magnetization, which has been considered as a key challenge in realizing next generation of fast, compact and ultra-low power magnetoelectric memories and sensors. Additionally, exchange bias can enhance dynamic magnetoelectric coupling strength in an external-field-free manner. In this paper, we provide a perspective on voltage control of exchange bias in different multiferroic heterostructures. Brief mechanism and related experiments are discussed as well as future trend and challenges that can be overcome by electrically tuning of exchange bias in state-of-the-art magnetoelectric devices.
\end{abstract}

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\section{1. Introduction}

Being able to maintain the stabilization of magnetization in spin-valve structures, the exchange bias (EB) effect has become one of the most decisive factors in the excellent performance of spintronic devices \cite{1}. For example, the EB effect plays an important role in the area of high-resolution magnetic sensors because of its minimal heat dissipation at highest performance \cite{2,3}. This review intends to provide an overview of the technological perspectives for the EB effect, one that manages to see the wide picture of its origin, intrinsic properties, application prospects, research status and future trend. Comprehending the characteristics of EB and the significance of voltage controllability in memory technologies is included in section 1. Two general manifestations of interface exchange interactions have been demonstrated in section 2. Sections 3, 4, 5 present existing studies on perspectives for the development of voltage controlled EB in different structural systems, which include multiferroic oxide heterostructures, shape memory alloy/piezoelectric laminate and FM/AFM/FE heterostructures. Section 6 gives the scenarios for future trend of voltage controlled EB and an overall summary is displayed in Section 7.

\section{1.1. Introduction for EB effect: origin and physical mechanism}

EB effect refers to a shift (\(\Delta H_{EB}\)) of magnetic hysteresis loop along the magnetic field axis caused by the exchange coupling of antiferromagnetic (AFM) and FM layers \cite{2,4,5}. When the temperature drops to the Neel temperature, \(T_N\), a unidirectional anisotropy will be induced in the FM layer and produces an EB effect at the interfaces of FM–AFM materials \cite{7}. The EB effect was first discovered in 1956 by Meiklejohn et al. based on Co–CoO system and was believed to be a result of the interface exchange coupling \cite{5,8}. Since the EB effect is important in both fundamental research and the information storage \cite{9–13}, many different systems containing FM–AFM interfaces have been studied for three types \cite{7}: (1) core/shell nanoparticles (e.g. Co/CoO \cite{14–18}, Ni/NiO \cite{19–22}, Fe/FeO \cite{23–26}); (2) non-uniform magnetic materials (e.g. spin glass \cite{27,28}, complex magnetic oxides \cite{29–34}, Heusler alloys \cite{35,36}); (3) bilayer and multilayer thin films \cite{37,39}. It is known that the EB effect strongly depends on the microstructures of the FM and AFM layers. As for the core/shell nanoparticles with potential for the high-density memories, they are limited to the preparation technology which can hardly control the interface properties precisely as well as the crystallinity of ultrathin AFM shell layer \cite{37,40}. And the non-uniform magnetic materials are restricted for the random distribution of magnetic ions which will generate the FM–AFM interfaces randomly \cite{28}. However, the controllability of atomic layers can be realized in thin films \cite{41–45}.
The sophisticated preparation technologies and analysis methods of films enable intensive studies for the EB effect [46–51]. Although EB effect has been studied for almost a half century, the physical mechanism started to be understood for only two decades. Generally, researchers believe that the EB effect is origins from the spin exchange coupling of the FM–AFM interface [8,12,13]. The pioneering EB papers of Meiklejohn et al. assumed an ideal interface and found the $H_{EB} = (n)S_M S_AFM/(M_{FM} M_{AFM})$ [8,12,13], where $n$ is the number of interaction spins of FM ($S_M$) and AFM ($S_{AFM}$), $J$ is the interaction intensity, $M_{FM}$ and $M_{AFM}$ are the magnetization and thickness of FM layer, respectively. As indicated in this expression, the EB fields related to interfacial character are inversely proportional to the thickness of coupled FM layer ($t_{FM}$). And the $t_{FM}$ is associated with the types of FM materials and the growth conditions, which demonstrates the EB effect obtains interfacial properties. This model supposed that the interface is perfect and did not reflect the influence of AFM layer. However, the experimental results have indicated that $H_{EB}$ is strongly affected by the thickness of AFM layer ($t_{AFM}$) [52]. The $H_{EB}$ dependence of $t_{AFM}$ in the multilayer thin films is highly related to the material system, the assignment of configuration, the microstructure and the experiment temperature [52–59]. Despite the complexity, the behavior can be primarily divided into two types: (1) $H_{EB}$ increased parallel with $t_{AFM}$ and reaches its saturation point when the $t_{AFM}$ is large enough [52–59]; (2) $H_{EB}$ declined after reaching the peak with an increasing $t_{AFM}$ [52,53,55,58–60]. Since the domain structure of AFM layer is hard to observe, the exchange coupling between FM and AFM layers can help to reflect it indirectly by the detection of domain structure variation in FM layer [52]. Yang et al. studied the changes of $H_{EB}$ and AFM-layer domain structure based on permalloy (200 Å)/FeMn ($t_{AFM}$)/Co/100 Å structure in 2000 [61]. And the observed spiraling domain structure indicated that the domain structure of AFM layer is vital to the exchange coupling [61]. In 2009, Morales et al. delved into Ni (50 nm)/FeF$_2$ ($t_{AFM}$/Permalloy (50 nm), finding that besides FM–AFM interfacial properties the antiferromagnetic bulk spin structure also contributes to the EB effect [62].

Actually, the appearance of EB effect requires the AFM layer having strong anisotropy field magnetic anisotropy [7]. Only if the $t_{AFM}$ $K_{AFM} \geq J$, the exchange coupling can happen [7]. Where $K_{AFM}$ is the anisotropy constant relating to $t_{AFM}$ and temperature, and $J$ is the interaction intensity, $K_{AFM}$ decreases with reduced $t_{AFM}$, and the EB effect will disappear if the $K_{AFM}$ is too weak despite of the existing interfacial spin exchange coupling [7]. Moreover, the EB effect is closely related to temperature, which can affect the pinning ability of AFM layer to FM layer. As temperature goes lower, the $K_{AFM}$ value becomes higher, and the EB effect increases [7]. Researchers have found that if the temperature is higher than a critical temperature ($T_0$), $H_{EB}$ comes to zero [63,64]. We may naturally think that $T_0$ is associated with Néel temperature, $T_N$, nevertheless, the exactly relationship remains to be discovered.

1.2. Voltage controlled EB effect in multiferroics: evolution and challenge

In recent years, magnetoelectric (ME) couplings have been introduced to FM–AFM structures and voltage controllable EB systems provide a new perspective as a novel control method [65–67]. Simultaneously occupying ferromagnetic (FM) and ferroelectric (FE) orders, multiferroic materials manage to manipulate magnetism by an electric field (E-field) or vice versa [68–85]. This means that $H_{EB}$ (magnetic property) can be regulated with E-field, and paves way for non-volatile, lightweight, and energy-efficient magnetic memories [86–90]. When the E-field is applied across the thickness direction of FE substrate, a strain on the piezoelectric phase of this substrate is generated, leading to a stress on the overlay’s magnetic phase through ME coupling [91]. Therefore, multiferroic materials, simultaneously occupying ferromagnetic (FM) and ferroelectric (FE) orders, have gained tremendous flurry of research interests [68–70,72–83,92–100]. Significant progress has been made in multiferroics, such as sensors, magnetoelectric random access memories (MERAMs), etc. [101–105]. They are potentially the most promising way to overcome power and temperature issues in optimizing the performance of micro-electronic devices [106]. There are existing studies focusing on the development of novel ME couplings, the exploration of different E-field controlling mechanisms (e.g. the strain-mediated converse ME coupling [107–113], the manipulation of domain structure [98,114–116], the interfacial charge mediated ME coupling [82,117,118]), and the E-field control of exchange coupling [119]. Though the strain-mediated ME couplings in multiferroic heterostructures can achieve a large ME coupling, they are limited for only realizing 90° magnetic rotations [120,121]. Recently, a breakthrough is achieved by Wang et al. [122]. They realized 180° magnetic moment reversal through electrostriction-induced magnetic anisotropy [122]. However, 180° deterministic switching still needs to be further explored for it is essentially required to satisfy the memory switching requirement [123]. The mentioned voltage control of FM/AFM EB proposed in multiferroics offers another prospective way for 180° magnetism switching [124].

In 2005 Binek and Doudin [125] suggested a focus on exchange bias with E-field for the achievement of energy-efficient MERAMs. This led to the invention of MERAM by Chen et al. [66]. Since then many activities in the search for EB using magnetoelectric materials have been triggered. In 2006, Kleemann’s group [66,126] proposed a concept of MERAMs which confirmed that purely electric control is possible by measuring the magnetoresistance of an exchange coupled spin valve. In 2009, Bibe et al. [105] proposed a typical MERAM as shown in Fig. 1, in which, the bottom FM layer of magnetic tunneling junction (MTJ) is coupled to AFM–FE layer. By switching the polarization 180°, the AFM vector switch 180° and then drive the FM layer reversal 180°. Two resistive states (parallel and antiparallel) are created by voltage-assisted 180° magnetization reversal through exchange bias. In 2010, a stress-mediated voltage controlled EB in multiferroic heterostructure has first been demonstrated by Polisetty et al. [4] and gave a novel method for the EB regulation. Finally, in 2014, Street et al. [127] reported an important technical breakthrough by doping the magnetoelectric control layer, Cr$_2$O$_3$, with 3% of B$_2$O$_3$. This crucially enhances the Néel temperature from 307 K to ~400 K and thus paves the way for technically acceptable EB-based spintronic devices in the near future.

Nevertheless, there are still many challenges in the voltage control of EB. The complex switching mechanisms, the challenges in understanding the physics behind this interface phenomena and the significant disadvantages caused by the presence of an external magnetic bias field (e.g., decrease of spatial resolution and signal-to-noise ratio) have made the utilization of EB effect a technologically difficult area both theoretically and experimentally [2,65,123]. Thus, over the past decade, numerous efforts have been put into the investigation of controlling EB [2,120,128,129]. To the best of our knowledge, although there are existing reviews related to the EB systems [7,10,11,130], few of them are target at giving a detailed introduction for the voltage control of EB effect based on multiferroics. It would, therefore, be interesting to provide an overview of the technological perspectives for voltage controlled EB effects. In this paper, we will present the remarkable progress in E-field control of magnetism in multiferroics achieved in recent few years, and discuss the challenges remain to be solved.

Several material systems and tuning strategies such as voltage control of EB in FM/multiferroic oxides heterostructures or in ferromag-
netic shape memory alloys (FSMAs) will be introduced, as well as the realization of 180° deterministic magnetization switching. Microscopic mechanisms behind different systems and approaches used to control exchange bias with electric fields remains a subject on which the proposed models are still very tentative. Other than delved into microscopic mechanisms, we focused more on recent research progress. In short, EB systems that based on multiferroic materials keep attracting numerous efforts for the robust, easy-fabricated and well-performed properties, which indicate that they are ideal for MERAMs and other E-field controllable memory technologies.

2. Two manifestations for interface exchange interactions

In conventional antiferromagnets like IrMn, NiO and CoO, it is impossible to control magnetism with E-field; however, multiferroics may offer exactly such an opportunity [131]. Studies done on multiferroic materials including YMnO3 and BFO show that strong exchange interactions can be obtained [131]. In fact, two general manifestations of interface exchange interactions between FM and AFM layers have been demonstrated so far [131]. The first one is related to exchange bias of the magnetic hysteresis loop [131]. Some spins of interface are pinned by AFM layer, therefore, the underlying FM layer must overcome these pinned spins before magnetization can be switched, resulting in exchange bias [132]. The second one is an enhancement of the FM coercive field [131]. The interface spins that not pinned will rotate with the FM layer when the H-field is swept; this is the so-called spin drag effect, which increases the coercivity of FM material [132]. Based on the action of uncompensated pinned spins mentioned here, in 2011, Borisov et al. separated a specific exchange bias-related contribution to the ferromagnetic coercive field for the first time [5]. We’ll discuss the material systems and tuning strategies in next three sections.

3. Perspectives for voltage control of EB in FM/multiferroic oxide heterostructures

In this section, the EB controlled by E-field in typical FM/multiferroic oxides heterostructures have been studied, and these three heterostructures presented latter have improved advantages compared with their former ones.

3.1. Control of EB in FM/Cr2O3

Many researchers believe that Cr2O3 is a suitable material for various spintronic applications for its significant coupling phenomenon at room temperature (below Neel temperature at the same time) [65,133–136]. Although, Cr2O3 does not have ferroic orderings, FM/Cr2O3 has been well-accepted by multiferroic research area and become a classic structure which is well investigated in many multiferroic papers [3,137–140], and we’ll treat it the same as the ideal multiferroic heterostructures.

In 2005, Borisov et al. [65,131] managed to control the EB field with various cooling treatments with combinations of E-field and magnetic field (H-field) based on the magnetoelastic heterostructure of Cr2O3 (111)/[Co (0.3 nm)/Pt (1.5 nm)]3/ Pt (1.5 nm). They reversed the orientation of AFM interface magnetization (S_{AFM}) and eventually obtained a large EB field (H_{EB}). The H-field offset can be quantified by the exchange bias fields (\mu_0H_{EB}), as illustrated in Fig. 2a. And they also explored the relationship between E_{RF} (E-field for freezing field) and \mu_0H_{EB} to investigate the domain formation and inverse mechanism. The functions between E_{RF} and \mu_0H_{EB} are found out to be hysteresis loops, as demonstrated in Fig. 2b. It can be found that, after ME field cooling (MEFC), the value of \mu_0H_{EB} is always negative (~20 mT) when E_{RF} ≤ 0, resulting from the positive remnant of the FM state. As E_{RF} gets a slightly higher (≤60 kV/m), the \mu_0H_{EB} has a distinct increment from negative to positive and eventually comes to maximum (~18 mT) while E_{RF} ≈ 100 kV/m. This indicates a formation of new domain structure which increases the positive polarization.
Fig. 2. (a) Normalized hysteresis curves of Cr$_2$O$_3$(111)/Pt 0.5 nm[Co 0.3 nm/Pt 1.5 nm]/Pt 1.5 nm measured after magnetic field cooling in $\mu_0H_T = 0.6$ T and $E_0 = 0$ from $T = 350$ to 298 K (1) and after magnetoelectric field cooling to 250 K in $\mu_0H_T = 0.6$ T and $E_0 = -500$ kV/m (2) and $E_0 = 500$ kV/m (3), respectively. The lines are to guide the eyes. The exchange bias fields $\mu_0H_{EB}$ referring to the loops 2 and 3 are indicated by arrows. (b) Exchange bias field $\mu_0H_{EB}$ vs electric freezing field $E_0$ as obtained from hysteresis loops after MEFC from $T = 350$ to 298 K in $E_0$ and magnetic fields $\mu_0H_T = 0.1$ (curve 1); 0.3 (curve 2), and 0.6 T (curve 3) (main panel and upper inset at enhanced scale). The lines are guides to the eye. The lower inset shows the experimental data $E_0$ vs $\mu_0H_T^{2/3}$ of the electric threshold field $E_0 = E_B (\mu_0H_B = 0)$ [65].

![Image](https://via.placeholder.com/150)

Fig. 3. Isothermal electric switching of the exchange-bias field. (a) Exchange-biased hysteresis loops of Cr$_2$O$_3$(0001)/Pd 0.5 nm/[Co 0.6 nm Pd 1.0 nm]/ at $T = 303$ K after initial magnetoelectric annealing in $E = 0.1$ kV mm$^{-1}$ and $\mu_0H_T = 77.8$ mT. Hysteresis loops are measured by polar Kerr magnetometry in $E = 0$, respectively. The red squares show the virgin curve with a positive exchange-bias field of $\mu_0H_{EB} = +16$ mT. Isothermal-field exposure in $E = -2.6$ kV mm$^{-1}$ and $\mu_0H = +154$ mT gives rise to a loop with a negative exchange-bias field of $\mu_0H_{EB} = -13$ mT (green triangles). (b) The red squares show the virgin reference loop. The blue circles show the hysteresis loop after isothermal-field exposure in $E = +2.6$ kV mm$^{-1}$ and $\mu_0H = -154$ mT, giving rise to the same negative exchange bias of $\mu_0H_{EB} = -13$ mT. (c) $\mu_0H_{EB}$ versus number of repeated isothermal switching through exposure to $E = +2.6$ kV mm$^{-1}$ (blue circles) and $E = -2$ kV mm$^{-1}$ (red squares) at constant $\mu_0H = -154$ mT, respectively [134].

of $S_{AFM}$. To analyse the inverse relationship between the threshold field $E_0$ and the magnetic freezing field, $\mu_0H_{EB}$, a model based on AFM interfacial spin energy is constructed, and the simulation result is shown in the lower inset of Fig. 2b. This work offers exactly such an opportunity to gain electrical control of exchange interactions. Nevertheless, the reversibility, uniformity and operability at room temperature are still the great challenges for the isothermal electric tuning. Therefore, further efforts should be made to promote the E-field control of repeatable magnetization switching to the realization of practical devices.

In fact, reversible switching is possible if the direction of biasing magnetic field is changed [141]. And, in 2010, He et al. [134] achieved repeatable magnetization switching in FM/Cr$_2$O$_3$ heterostructure after applying inverse interface magnetization. As illustrated in Fig. 3a and b, by reversing the E-field isothermally ($T = 303$ K) while maintaining two same field product $EH$, one can change the initial EB fields, $\mu_0H_{EB} = +6$ mT, to the same value of $\mu_0H_{EB} = -13$ mT. Since the electric current monitored stayed below 0.01 $\mu$A, the field inducing becomes the possible reason for the AFM single-domain state switching in the Cr$_2$O$_3$ heterostructure. Fig. 3c represents the $\mu_0H_{EB}$ with repeated isothermal switches ($T = 303$ K), the E-fields for switching are $E = +2.6$ kV mm$^{-1}$ and $-2.0$ kV mm$^{-1}$ (blue circles and red squares, respectively). On account of the difference existing in the interface magnetization, the
\( \mu_0 H_{EB} \) values are asymmetric while under positive and negative exchange bias. These results based on FM/\( \text{Cr}_2\text{O}_3 \) heterostructure realize repeatable magnetization switching with the help of E-field, making a significant improvement compared to the work Borisov's group did in 2005. Meanwhile, isothermal EB switching in MERAM has become feasible by using very thin films of \( \text{Cr}_2\text{O}_3 \), whose (0001) faces irrespective of their accidental roughness always have ME switchable ferromagnetic surface magnetization [106,126,134].

3.2. Exchange bias in FM/\( \text{YMnO}_3 \)

The availability of ferroelectric antiferromagnets, such as the multiferroic perovskites, is another fascinating way to modulate magnetic structure with E-field [142]. Coupling between AFM component and an adjacent ferromagnet provides permanent, non-volatile control of ferromagnetism switching [142]. This promises to be an exciting area of research and suggests the possibility of ultra-low power spintronic applications [142].

\( \text{YMnO}_3 \) (YMO) is a multiferroic perovskite of this kind. Being able to maintain multiferroic properties over a large temperature range (i.e., the ferroelectricity exists up to 900 K, the AFM property emerges when the temperature is below 90 K), YMO has been well received [143–147]. It manages to couple FE and AFM orders together, enabling E-field to control exchange bias of the AFM-pinned FM layer [143]. Some reports have found that it’s viable for epitaxial YMO (0001) films with remnant electric polarization to obtain exchange bias in FM materials like NiFe [143,145,146]. The E-field induced exchange bias in FM/YMO heterostructures constitutes an important step toward the new generation of spintronic devices, and have aroused a hot wave in research [143,144].

In 2006, Laukhin et al. [143] discussed the voltage control of exchange bias shift by fabricating NiFe (Permalloy-Py, 15 nm) / \( \text{YMnO}_3(0001)/\text{Pt} \) (8 nm) heterostructure, in which they tuned the exchange-bias coupling, and eventually switched the magnetization of the FM layer with E-field. Their exploration of magnetotransport properties was based on the ME coupling between the multiferroic YMO (AFM and FE) layer and FM layer, and the experimental results can be seen in Fig. 4. The narrow, symmetrical hysteresis loop of \( V_E = 1.2 \) V shown in Fig. 4a reflects the inhibitions of coercivity and the \( H_{EB} \). This sample was cooled from 300 K to 2 K, and \( H_{EB} \) around 60 Oe can be directly observed without thermal cycling. And the arrow in Fig. 4a illustrates schematically that it’s possible to realize the E-field-induced magnetization switching of the FM material (Py). Fig. 4b shows the biasing-voltage \( (V_E) \) dependence of the magnetization, and the zoom on the left clearly indicates that the magnetization \( (M) \) cannot return back to the initial state \( (M > 0) \) despite the reduction of \( V_E \). Nevertheless, the E-field can modify the magnitude of \( M \). In fact, the dynamic switching of \( H_{EB} \) with an applied E-field remained elusive until Laukhin et al. reported their work of \( \text{Y MnO}_3 \) focusing on these very low temperatures [131]. However, the switch of magnetization in FM layer has a strong dependence on external H-field and temperature. Further researches are still needed.

3.3. Control of EB in FM/\( \text{BiFeO}_3 \)

\( \text{BiFeO}_3 \) (BFO) has indirect coupling between ferroelectric and antiferromagnetic order parameters, and the coupling mechanisms based on the electrically controllable polarization effect have been explored successfully. Researchers revealed that the different domain wall patterns offer varied properties; canted moment from \( 71^\circ \) domain walls in BFO film coupled to FM layer that allows 180\(^\circ\) magnetic moment switching [148–150]; while 109\(^\circ\) domain wall BFO film introduces EB into FM/BFO system, by which the FM moment can be manipulated by changing EB in BFO films [151–153]. Béa et al. represented the first direct observation of exchange bias with a multiferroic material at room temperature with no training effect [101] and confirmed the relationship between \( H_{EB} \) and the size of multiferroic domains later on [86]. Despite of the absence of voltage, their work paves the way towards the room-temperature electrical control of magnetization with BFO. In fact, the FM/BFO heterostructure combines the properties of BFO and the interface exchange coupling with the FM layer [154]. Therefore, controlling a large ferromagnetic magnetization with solely an applied E-field is possible [154]. This is our focus since it’s attractive to the spintronics community in the reduction of energy consumption [154].

Wu et al. have made some investigations on how voltage controlled EB in \( \text{La}_0.7\text{Sr}_0.3\text{MnO}_3/\text{BiFeO}_3 \) systems [132,155]. Fig. 5 shows their work done in 2010 [155]. They can repeatedly reverse the ferroelectric polarization with voltage and thus, switch two exchange-bias states (low and high) by the way of strain-mediated coupling. Without the aid of bias magnetic field or field cooling, the maximum modulation of exchange-bias was about 0.15 \( \text{H}_C \), which reflected the existence of an exchange-bias field shift (\( \sim 125 \) Oe). As for the mechanisms related to this E-field effect on \( H_{EB} \), the authors attributed the phenomena to the following two aspects. Firstly, based on the effects of strain, the reverse of voltage pulses brings out the switch of BFO ferroelectric polarization, generating the strain-mediated interface magnetism due to the magnetostriuctive effect of LSMO layer. Although the strain influence accounts and cannot be ruled out in this case, it is a key factor during the voltage controlling exchange bias. Secondly, authors imagined an atomic-scale mechanism based on Fe–Mn coupling: Negative (positive) \( V_E \) pulls (pushes) the carriers to (away from) the interface, therefore, the carriers can modulate the magnetotransport properties of LSMO, which eventually reflects in the aspects of exchange bias and magnetoresistance. The role that domain structure of BFO may play was eliminated because that different domain structures exhibit nothing different in exchange-bias magnitude. This result was opposite to what found in \( \text{Co}_0.9\text{Fe}_0.1 \) (CoFe)/BFO, demonstrating E-field induced interface magnetism can be a crucial reason. In 2013 they further studied the direct, bipolar electrical control
of exchange bias based on $\La_{0.7}\Sr_{0.3}\MnO_3/BiFeO_3$ system [132]. The bipolar modulation effect under the help of a finite voltage within the initial BFO polarization was demonstrated once again without field cooling, temperature cycling or external H-field [132]. This would provide a low-power alternative for the conventional magnetization control mechanisms despite of the need of low temperature.

4. Control of EB in shape memory alloy/piezoelectric laminate

Some ferromagnetic shape memory alloys (FSMAs) of Ni–Mn alloys also perform well in EB controlling because the increase of AFM exchange coupling can give rise to a large exchange bias field ($H_{EB}$) for the voltage control [35,156–161]. The H-field generates martensitic transformation as well as the arrangement of martensite variants, then FM and AFM exchange coupling emerges [35,156,161]. Large strain in this process can produce interactions between structures and the magnetism [35,156,161], which eventually change the magnetic properties [35,156].

Based on FSMAs, Yang et al. [156] obtained a strong ME effect in 2015. And magnetization switching without external bias magnetic field was finally realized due to the strain mediated converse ME coupling. They developed an FSMA of $\Mn_{50.12}\Ni_{30.31}\Sn_{10.57}$

0.7Pb ($\Mg_{1/3}\Nb_{2/3}\O_3$ – 0.3PbTiO$_3$ (MNS/PMN-PT) for the study of $H_{EB}$ with an applied E-field. They found that the induced strain will affect magnetic properties or the orientation of AFM and FM interface magnetization ($\Phi$). Fig. 6b shows the results measured after field cooled (FC) process. From the results, we can calculate $H_{EB}$ by the left and right coercive fields ($H_1, H_2$): $H_{EB} = -(H_1 + H_2)/2$. For the initial state ($E_{dc} = 0$ kV/cm), a considerably large negative shift ($H_{EB} = 1086$ Oe) of the loop’s center is observed. When the electric is up to 4 kV/cm, a clearly decrease of 27 Oe ($H_{EB} = 1059$ Oe) emerges. And if the $E_{dc}$ returns back to 0 kV/cm, the $H_{EB}$ of the MNS laminate reverted to the previous state, indicating that the E-field was the cause for the shift of hysteresis loops. Fig. 6c displays the change of saturation magnetization $M_S$ ($\Delta M_S$) and exchange bias field ($\Delta H_{EB}$) with temperature control while the sample was under an $E_{dc}$ of 4 kV/cm. The results suggested that, the E-field induced exchange bias field, $H_{EB}$, was more sensitive to monitor the strain-mediated ME effects than $M_S$ when the MNS/PMN-PT heterostructure was under different temperatures. The values of $\Delta H_{EB}$ are much more conspicuous since both magnetic properties and $\Phi$ can be modified by strain, making $\Delta H_{EB}$ a better index than $\Delta M_S$ in the voltage controlled progress when temperature varies. This work manages to switch the magnetization without an external bias magnetic field and the obtained $H_{EB}$ is considerably large. However, the low temperature will still hinder its practical application.

5. 180° magnetization switching by controlling EB in FM/AFM/FE heterostructures

Although the non-volatility of voltage controlled magnetism modulation has been realized in magnetoelectric random access memories (MERAMs), the 180° deterministic, dynamic magnetization switching for information storage still eagers for further exploration [120,162]. In 2011, Liu et al. proposed another possible concept for electrically controlled information storage [120].

Fig. 5. Electric-field control of exchange bias. (a) The gate-voltage-pulse sequence used for the measurements. (b,c) Measurements of normalized exchange bias and peak resistance for the gate-pulse sequence shown in (a). Each point is determined from an MR sweep at 5.5 K after pulsing the gate with voltage $V_g$. The exchange bias modulates with ferroelectric polarization; the data shown for (b) were obtained with a negative remnant magnetization in the LSMO channel whereas the data shown for (c) were obtained in positive remnant magnetization. Error analysis was done on (b) by taking multiple MR sweeps. Standard deviations of the individual peak locations were obtained and the error was calculated for each point using standard error propagation techniques. Owing to the large number of data and the long time it takes to run such an experiment, the corresponding measurement was not made on (c). Both measurements were made without field cooling the system at any time. The maximum normalized values of exchange bias correspond to shifts of $\sim 100$ Oe in (b) and $\sim 200$ Oe in (c, d). Examples of individual MR curves from the upper and lower resistive states where the exchange-bias values were determined [155].

Fig. 6. (a) Schematic diagram of the MNS/PMN-PT laminate. (b) M–H loops for the MNS/PMN-PT laminate at 4 K with and without $E_{dc}$. (c) Temperature dependence of the changes of $\Delta M_S$ and $\Delta H_{EB}$ with the application of $E_{dc} = 4$ kV/cm. Here the value of $M_S$ is calculated as $M_S = (M_{S1} + M_{S2})/2$, where $M_{S1}$ and $M_{S2}$ are the positive and negative saturation magnetization, respectively [156].
They explored the E-field modulation of exchange bias based on FeMn/Ta (5 nm)/FeMn (15 nm)/Ni80Fe20 (2 nm)/FeGaB (14 nm)/Ta (20 nm)/PZN-PT (011) (lead zinc niobate–lead titanate) multiferroic heterostructures. The investigation for voltage control of the magnetization switching was carried out with an external bias magnetic field \( H_E \) applied at the angle of 55° to the magnetic easy axis. As presented in Fig. 7a, when the E-field was reduced from 6 kV cm\(^{-1}\) to 4 kV cm\(^{-1}\) (\( t = 100 \) s), near 180° deterministic magnetization switching can be realized with the help of \( H_E = 28 \) Oe. However, this procedure was nonreciprocal, which meant that the magnetization cannot return back to its initial state even if the E-field changed from 4 kV cm\(^{-1}\) to 6 kV cm\(^{-1}\). This irreversibility can hardly meet the demand of repeated magnetization switches in dynamic memories. In order to solve this problem, a magnetic impulse generated by a giant electromagnet was utilized to inverse the magnetization back. As demonstrated in Fig. 7b, 180° dynamic magnetization switching in FeGaB film can be observed when the magnetic impulse was applied with an square-wave voltage and a bias magnetic field, \( H_E = 28 \) Oe. Additionally, dynamic magnetization switching was also realized when \( H_E = 0 \) Oe, as shown in Fig. 7c. The energy required for the producing of magnetic impulse and square-wave voltage was relatively low. Thus, the near 180° dynamic magnetization switching realized in AFM/FM/FE multiferroic heterostructures at ambient temperature enhanced the low power dynamic memory technologies.

Furthermore, in 2015, Chen et al. \cite{163} extended the reversible E-field control of magnetization switching based on Ta (4 nm)/Pt (2 nm)/IrMn (8 nm)/CoFeB (55 nm)/Ta (5 nm)/PMN-PT (011) multiferroic heterostructure. They studied the angular dependence of exchange bias by tuning the ratios and relative orientations of different anisotropy dynamically with voltage and a help of magnetic fields ranging from \(-15 \) to \(-35 \) Oe, stressing the importance of E-field controlled exchange bias for the design of ultralow power magnetoelectric devices. What’s more, they finally realized reversible magnetization reversal at zero magnetic fields by an optimization of anisotropy configuration. Despite of the requirement for a precise sample design, both of the two works hold promising applications in the novel spintronic devices.

6. Future trend of voltage controlled EB

Recently, spin Hall effect (SHE), which can eventually generate spin–orbit torque (SOT) and induce magnetization switching, have aroused much research interest in AFM/FM systems \cite{164, 166}. This SOT-induced switching promises an external-field-free switching of the perpendicular magnetization, which is attractive in ultrahigh-density information storage \cite{167}. The inverse SHE, in which a pure spin current is converted to a charge current, was also observed in AFM/FM systems \cite{164, 168, 169}. In these systems, AFM can exert an internal effective field on the adjacent FM through the exchange bias. Therefore, multidimensional manipulation of magnetization may be possible. If AFM/FM is combined with FE substrates, the voltage regulation can be involved. Moreover, the recently discovered magnetic skyrmion with spin texture provides a new storage structure which can potentially result in significantly improved densities and much lower power dissipation compared to conventional domain wall devices \cite{170, 171}. If the AFM/FM systems, FE substrates skyrmion structures combine together, novel phenomena may be possible when the voltage is applied. We think this kind of regulation can offer a new way for the electrical manipulation of magnetization and its application in spintronics.

7. Conclusions

As a conclusion, we give perspectives on what have been observed and what can be expected for the future with regard to voltage control of exchange bias. Based on varied multiferroic heterostructures, one manages to see the wide picture of regulation process: firstly, the exchange bias of FM/multiferroic (\( \text{Cr}_2\text{O}_3 \), \( \text{YMn}_2 \), \( \text{BiFeO}_3 \)) bilayer can be controlled by polarization switching due to co-existence and coupling of AFM and FE properties in multiferroic layer. Secondly, FSMA/FE heterostructure creates a tunable exchange bias due to martensitic phase transformation in FSMAs. Lastly, the controllable exchange bias can realize the 180° magnetization switching and increase the ME coupling coefficient in FM/AFM/FE multiferroic heterostructures. We believe that the development of voltage controllable exchange bias provides a possibility for smaller, faster, more efficient spintronics/electronics devices.

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