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Perspectives of voltage control for magnetic exchange bias in multiferroic heterostructures

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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° deterministic 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 mechanization 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.

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 [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 [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.

1.1. *Introduction for EB effect: origin and physical mechanism*

EB effect refers to a shift (H_{EB}) of magnetic hysteresis loop along the magnetic field axis caused by the exchange coupling of antiferromagnetic (AFM) and FM layers [2,4–6]. 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 [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 [5,8]. Since the EB effect is important in both fundamental research and the information storage [9–13], many different systems containing FM—AFM interfaces have been studied for three types [7]: (1) core/shell nanoparticles (e.g. Co/CoO [14–18], Ni/NiO [19–22], Fe/FeO$_x$ [23–26]); (2) non-uniform magnetic materials (e.g. spin glass [27,28], complex magnetic oxides [29–34], Heusler alloys [35,36]); (3) bilayer and multilayer thin films [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 [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 [28]. However, the controllability of atomic layers can be realized in thin films [41–45].

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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 origin 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 $H_{\mathrm{EB}} = (nJS_{\mathrm{FM}} \cdot S_{\mathrm{AFM}})/(M_{\mathrm{FM}}t_{\mathrm{FM}})$ [8,12,13]. Where n is the number of interaction spins of FM (S_{FM}) and AFM (S_{AFM}), J is the interaction intensity, M_{FM} and t_{FM} 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} $K_{\mathrm{AFM}} \geq J$, the exchange coupling can happen [7]. Where K_{AFM} is the anisotropic 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_{B}), H_{EB} comes to zero [63,64]. We may naturally think that T_{B} is associated with Neel 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 overlayer'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 microelectronic 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 conformed that purely electric control is possible by measuring the magnetoresistance of an exchange coupled spin valve. In 2009, Bibes 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-

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