

Recent advances in exchange bias of layered magnetic FM/AFM systems

LIU ZhongYuan*

State Key Lab of Metastable Materials Science & Technology, Yanshan University, Qinhuangdao 066004, China

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The exchange bias (EB) has been investigated in magnetic materials with the ferromagnetic (FM)/antiferromagnetic (AFM) contacting interfaces for more than half a century. To date, the significant progress has been made in the layered magnetic FM/AFM thin film systems. EB mechanisms have shown substantive research advances. Here some of the new advances are introduced and discussed with the emphasis on the influence of AFM layer, the interlayer EB coupling across nonmagnetic spacer, and the interlayer coupling across AFM layer, as well as EB related to multiferroic materials and electrical control.

exchange bias, coercivity, layered magnetic thin films, interlayer coupling, electric control of exchange bias

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For the magnetic materials with ferromagnetic (FM)-antiferromagnetic (AFM) interfaces, after cooling through the AFM Néel temperature of T_N (lower than the FM Curie temperature of T_C in general) in an applied magnetic field, the measured hysteresis loop exhibits different coercivity fields in absolute value in field-descending and ascending branches. Specifically, the loop center is shifted away from the zero-field position (Figure 1). Generally, a negative loop shift occurs relative to the cooling field direction. This loop shift is usually called as exchange bias (EB) or exchange biasing field (H_{EB}). This phenomenon was firstly observed by Meiklejohn and Bean [1] in a core/shell particle system of Co/CoO, and it was attributed to the exchange interaction at the interface between FM Co core and AFM CoO shell [2,3].

In the past more than half century since the discovery of EB, the EB effect has been extensively investigated in many different magnetic systems containing the FM-AFM interfaces because of its importance in fundamental research and

technological applications in information storage [4–8]. In brief, the magnetic materials exhibiting the EB effect can be separated into three main different categories [4]: 1) the core/shell particle systems such as Co/CoO [9–13], Ni/NiO [14–17], Fe/Fe oxides [18–21], and ZnO-Fe/Fe_xO_x [22]; 2) the inhomogeneous magnetic materials, for example, spin glasses [23,24], magnetic complex oxides [25–32], and Heusler alloys, [33,34]; 3) layered magnetic thin film systems [35–128].

The research in the core/shell systems has been strongly restricted because of the limited technological routes of fabrication. Usually, the AFM shells are so thin that the Néel temperature T_N is much lower than the bulk value. The interfacial exchange biasing is sufficiently strong to pin the magnetic moments in the FM core. Moreover, it is difficult to identify the FM-AFM interfacial nature and the crystallinity of the ultrathin AFM shells. Hence, the core/shell nanostructures are not ideal systems for investigation of fundamental features of EB. Nevertheless, the EB effect in nanostructured magnetic materials has been reported to strongly suppress the superparamagnetism [35,36], showing

*Corresponding author (email: liuzy0319@yahoo.com)

great potential applications in high density information storage. A renewed interest has been activated in the studies of EB in the core/shell particle systems. The progress in this part has been summarized in a review by Nogués et al. [8].

In the inhomogeneous magnetic materials, there are no well-defined FM-AFM interfaces. The FM or AFM exchange interactions are actually generated in different area or domains due to the arrangement of magnetic ions. The FM-AFM interfaces are thus randomly formed. It is difficult to extract the useful information for understanding the EB. The inhomogeneous magnetic materials are thus not the ideal systems for investigation on fundamental aspects of EB, though some of them have been reported to exhibit pretty interesting EB-related phenomena [24].

The EB is well known to be strongly dependent on many factors related to the FM and AFM layers such as the microstructure of both the AFM and FM layers (e.g., grain size, crystalline orientation, domain configuration and roughness) and the spin structure at the FM-AFM interface. To date, progress in the studies of EB has been made in the layered magnetic thin films. This is mainly because of the following reasons. The manipulation of the EB relies on the fine control of the FM-AFM interface at the atomic level, and the required experimental and analytical tools are well-developed for the layered thin films. The EB and its related phenomena have been utilized in many different technological applications since their discovery, and most of them are found in the thin-film forms, particularly the spin valve and

tunneling devices in recent years [4–8]. Thereby, the explosive studies have been triggered on the layered FM-AFM thin film systems. In response to the influences of those factors, many remarkable features are usually observed to appear along with the loop shift or exchange biasing field of H_{EB} (Figure 1), for example, enhanced coecivity [37–40], asymmetric magnetization reversals [41–56], training effect [56–73], and memory effect [74]. Though the EB-related phenomena have been extensively investigated for more than half a century, good understanding of the underlying EB mechanisms has actually been obtained in just the past decade. In this review, some of the recent advances in the studies of EB are introduced and discussed.

1 AFM layer dependence of EB

The EB originates from the exchange interaction at the FM-AFM interface. From consideration of an ideal interface model originally proposed by Meiklejohn and Bean [1–3], the exchange interaction between neighboring spins at the FM-AFM interface can give rise to an expression of H_{EB} as $H_{EB} = (nJS_{FM} \cdot S_{AFM}) / (M_{FM}t_{FM})$, where n is the number of interactions per unit area between FM spins of S_{FM} and AFM spins of S_{AFM} at the interface. J is the interaction strength between S_{FM} and S_{AFM} spins. M_{FM} and t_{FM} are the magnetization and thickness of the FM layer, respectively.

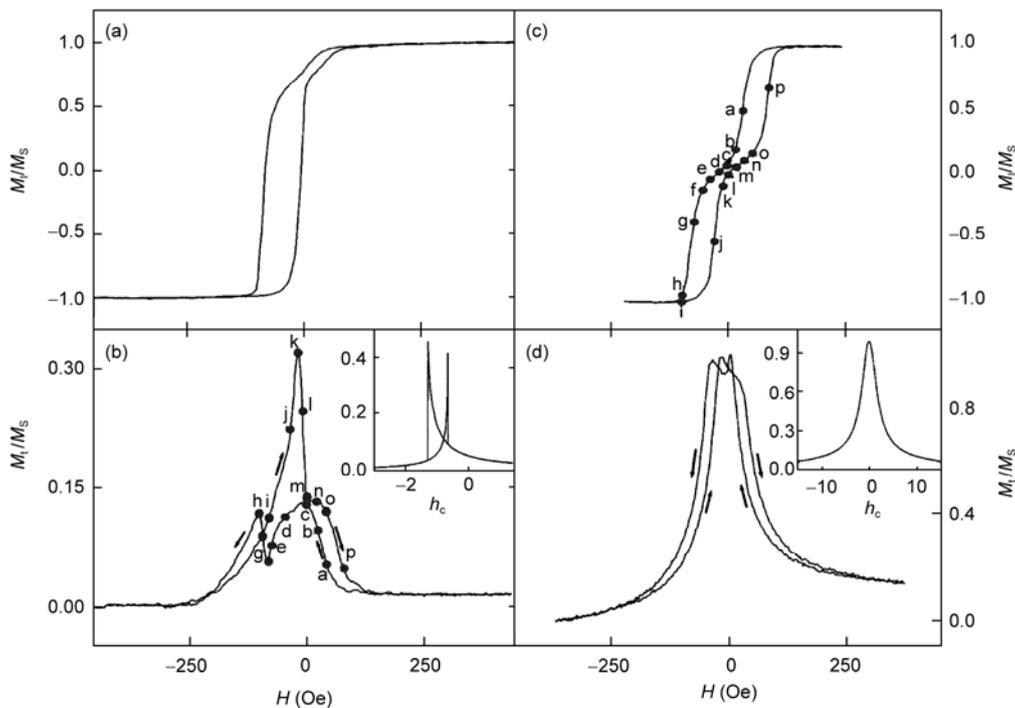


Figure 1 M_L - H and M_T - H loops along the as-deposited unidirectional (a) and (b) and hard (c) and (d) axes for the NiO/Ni₈₁Fe₁₉ bilayer (M_L and M_T represent the longitudinal and transverse components of magnetization, respectively). The insets in (b) and (d) are the calculated M_T - H loops at $\alpha=87^\circ$ and 0° , respectively, from the rotation model. In the calculation, the scaled coercivity h_c is taken to be 0.5 [49].

Thereby, H_{EB} is expected to show an inversely linear dependence on the FM layer thickness t_{FM} , that is, $H_{EB} \propto 1/t_{FM}$. This inversely linear relation is experimentally supported if the FM layer thickness of t_{FM} is larger than a critical value [75,76], being suggestive of the interfacial nature of EB. The critical FM thickness depends on the FM materials and the growth condition.

Compared to the FM layer, the AFM layer exhibits quite complex influences on H_{EB} . It has been revealed that the EB is dependent on many factors related to the AFM layer. In the abovementioned expression of H_{EB} , the exchange coupling is considered to involve just the interfacial spins in the FM and AFM layers. The AFM layer thickness does not appear, suggesting that H_{EB} should be independent on the AFM layer thickness. However, the experimental investigations indicate that H_{EB} has strong dependence on the AFM layer thickness (Figure 2) [77]. When the AFM layer is sufficiently thin, H_{EB} exhibits an abrupt drop with the decrease in the AFM thickness in a narrow range. When the AFM thickness is less than a critical value, H_{EB} is rendered to zero. This critical AFM thickness depends on the specific AFM material and the measurement temperature. This AFM thickness dependence of H_{EB} is a common phenomenon. When the AFM layer is relatively thick, the dependence of H_{EB} on the AFM thickness becomes quite complex. Relying on the specific layered system, its layered configuration and microstructure, and the measurement temperature, the variation of H_{EB} with the increase in the AFM thickness can be generally separated into two main categories: 1) H_{EB} increases to a constant value when the AFM layer is thick enough [78–84,87]; 2) H_{EB} first increases to a maximum and then decreases with increasing the AFM thickness [77,78,80,84–88].

In the relatively thick range, the decrease of H_{EB} with decreasing the AFM thickness was observed to follow the inversely linear relation of $H_{EB} \propto 1/t_{AFM}$ in some layered thin film systems, for example, the NiFe/CoO system with $t_{CoO} > 10$ nm [89]. This inverse relation suggests that the EB should be affected by the spin structure and the domain structure within the AF layer, being consistent with the micromagnetics calculations by considering the influence of

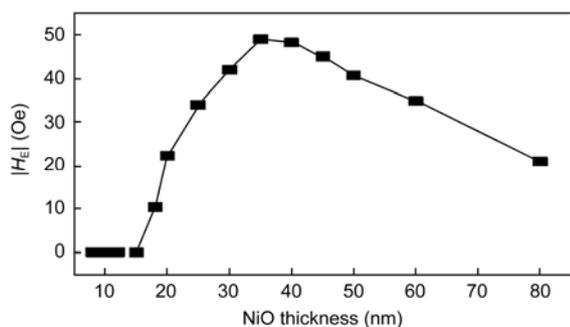


Figure 2 EB field as a function of the NiO thickness for the NiFe/NiO bilayer [77].

the spin structure and the domain walls in the AFM layer [90–92]. Although the AFM domain structure is difficult to be directly observed, it is able to be indirectly reflected via the domain configuration in the FM layer because of the exchange coupling at the FM-AFM interface. In a bilayered system of NiFe/NiO, the domain imaging studies of the FM layer indicate the clear variation of FM domain configuration with the NiO thickness [77]. Yang and Chien [93] carried out the studies on the EB in a trilayer system of Permalloy (200 Å)/FeMn (t_{AFM})/Co (100 Å) via a special cooling procedure. Their experimental results are suggestive of the formation of spiral domain walls in the AFM layer for $t_{AFM} < 15$ nm as proposed by the Mauri's model [91], demonstrating the key role of AFM domain walls in the EB. Recently, Morales et al. [94] investigated the role of AFM bulk spin structure in the EB of a trilayered Ni (50 nm)/FeF₂ (t_{AFM})/Permalloy (50 nm) system by performing the field cooling with the either parallel or antiparallel magnetizations in the two FM layers. In their studies, the AFM layer was chosen to be so thick that the interlayer coupling across the AFM layer could be ruled out, implying that the formation of spiral domain walls through the AFM layer is not possible. Their results reveal the significant difference in H_{EB} between those two cooling configurations. This interesting difference confirms that the EB involves not only the interfacial spins but the bulk AFM spin structure. According to the domain state model [95], the formation of domains throughout the AFM layer depends on the cooling process with the parallel or antiparallel magnetizations in the two FM layers, providing a reasonable explanation of the observed difference in H_{EB} .

For the existence of EB, it is well known that a sufficiently strong anisotropy of $t_{AFM}K_{AFM}$ is required in the AFM layers. Theoretically, $t_{AFM}K_{AFM} \geq J$ (J is the exchange coupling strength) should be satisfied [4]. The K_{AFM} relies on the AFM thickness. For the thick enough AFM layer, K_{AFM} tends to be the bulk value. However, K_{AFM} becomes weaker with the decrease in t_{AFM} . K_{AFM} can be overly weak to maintain the EB if the AFM layer is thin enough, although the exchange coupling can still exist. It is well known that the EB has a strong dependence on temperature. With the increase of temperature, H_{EB} decreases and is rendered to zero at a critical temperature, that is, the reputed blocking temperature of T_B . It is natural to think that T_B should be related to the Néel temperature T_N of the AFM layer. However, T_B is lower than T_N , and the difference between T_B and T_N depends on the layered FM/AFM systems and the AFM materials. For the AFM layers such as CoO, the studies by Ambrose and Chien [89,96] reveal the finite size effect, that is, the Néel temperature T_N decreases with decreasing the AFM thickness, following the finite-size scaling relation with a shift exponent of $\lambda = 1.55 \pm 0.05$. The investigations on the EB in a bilayered system of NiFe/CoO show that T_B and T_N have the similar dependence on the AFM layer thickness, though T_B is always slightly lower

than T_N [89]. This observation suggests that the EB is greatly affected by the finite-size scaling of the AFM Néel temperature T_N . For the sufficiently thin AFM layers, the observed sharp drop of H_{EB} with decreasing the AFM thickness has been thus believed for a period to originate from the finite size effect of T_N . However, recent studies on ultrathin CoO layers in a $\text{Fe}_3\text{O}_4/\text{CoO}$ multilayer [97] reveal that with decreasing the CoO layer thickness, the exchange-coupled AFM layer exhibits the enhanced T_N because of the magnetic proximity effect, while T_B displays the decrease. This AFM thickness dependence of T_B may not be explained by the finite size effect, remaining a topic for further research.

2 Interlayer EB coupling in trilayered FM/NM/AFM systems

Among the experimental studies on the EB, most of them have been conducted in the magnetic systems with the direct FM-AFM contact. The EB has long been considered to originate from a nearest-neighbor exchange coupling at the FM-AFM interface, being in the interfacial nature. For instance, many investigations suggest that the EB is induced by a small number of pinned uncompensated AFM spins at the interface [96], and thus the main part of the uncompensated AFM spins can be coupled to the FM layer and be rotatable with it [97,98]. This will lead to the enhanced coercivity. By inserting of a nonmagnetic spacer layer between the FM and AFM layers, however, the studies on the trilayered FM/NM/AFM systems demonstrate that an interlayer EB coupling exists across a nonmagnetic spacer, being a general phenomenon [99–109]. This finding strongly supports that the origin of EB is beyond the interfacial effect.

In the Permalloy/Au/CoO trilayers, the studies by Göke-meijer et al. [99,100] reveal that a long-range interlayer EB coupling exists between the FM and AFM layers across a nonmagnetic spacer. The coupling strength was observed to decay exponentially with the increase of the NM thickness and persist to as much as 50 Å or more, depending on the NM materials. This discovery is contrary to the prevailing assumption that EB is a short-range interaction at the FM/AF interface. It was proposed that the conduction electrons may be involved in the mediation of the coupling. The other investigations of EB in the trilayered FM/NM/AFM systems with different NM spacers confirm the existence of the interlayer EB coupling across a NM spacer and the exponential decay of the coupling strength, but with the decay length much shorter than 50 Å [101–103]. The observed rapid suppression of EB by a NM spacer was attributed to pinholes by some researchers [104]. In the trilayered systems of FeNi/Cu/FeMn [105] and NiO/Cu/NiFe [106], the oscillatory interlayer EB coupling was claimed to be observable as a function of the NM spacer thickness. It was

directly connected with the oscillatory interlayer coupling across the spacer in the FM/NM/FM trilayers [105] or was considered to be thermally assisted on the basis of the temperature-dependent competition between the RKKY-like coupling and the AFM coupling within the AFM layer as well as the interlayer dipolar interaction [106]. By inserting an ultrathin Pt spacer between the $[\text{Pt}/\text{Co}]_n$ multilayer and the AFM layers of FeMn [107] and IrMn [108], considerable enhanced EB was observed. The bias field H_{EB} reached a maximum for a spacer layer thickness of a few angstroms, and it then decreased quickly with the further increase of the spacer. H_{EB} enhancement is attributed to the role of the Pt spacer in increasing the perpendicular component of the ML magnetization.

Though the experimental studies give the inconsistent results about the short-range or long-range nature of the exchange coupling between the FM and AFM layers, all of them confirm that the EB is not necessarily produced by a nearest-neighbor (direct exchange) or next nearest-neighbor (superexchange) coupling mechanism. Recently, Valev et al. [109] have probed the EB in a trilayer of Fe/Cu/CoO via magnetization-induced optical second harmonic generation (MSHG). It was found that the appearance of EB at the blocking temperature T_B is accompanied by the formation of pinned uncompensated spins at the AFM/spacer interface. The alignment of those pinned uncompensated spins under the influence of the FM interface is considered to be directly responsible for EB. It was observed that the MSHG signal still shows a strong magnetic contribution from the CoO interface even when the loop shift drops to zero at the spacer thickness of 3.5 nm. This result indicates that even when no loop shift is observable at a spacer thickness, the magnetic interaction between FM and AFM layers can be still sufficiently strong to induce order in the AFM layer. Therefore, the effective distance of the FM influence on the magnetic order at the AFM interface should be longer than that determined from the loop shift. Most probably, the exchange coupling at the FM-AFM interface has the long-range nature.

3 Interlayer coupling in trilayered FM/AFM/FM systems

In the trilayered FM/AFM/FM systems, if the AFM separating spacer is sufficiently thin, the two FM layers are expected to be coupled together via the antiferromagnetically ordered spin structure in the AFM separating spacer. This interlayer coupling is actually induced by the exchange coupling at the top and bottom FM-AFM interfaces. The coupling nature can be FM or AFM, depending on the AFM spacer thickness. Recently, the interlayer coupling across the NiO spacers has been observed in the $[\text{Pt}/\text{Co}]_n/\text{NiO}/[\text{Co}/\text{Pt}]_n$ multilayers with perpendicular anisotropy (Figure 3) [110, 111]. With the increase of the NiO spacer thickness

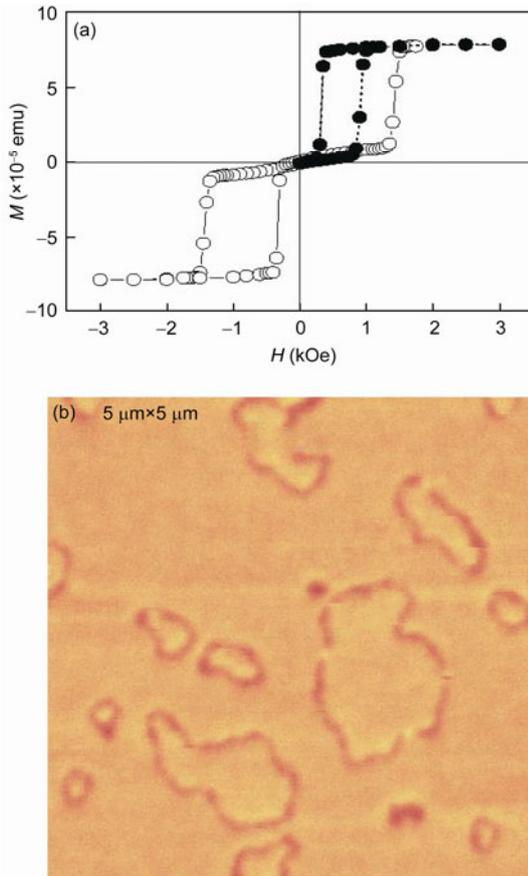


Figure 3 (a) Major (\circ) and minor (\bullet) M - H loops along the out-of-plane easy axis for glass/Pt (100 Å)/[Pt (5 Å)/Co (4 Å)] $_3$ /NiO (11 Å)/[Co (4 Å)/Pt (5 Å)] $_3$ /Pt (50 Å). The minor loop is measured after a positive saturation of the whole system, in a field range where the magnetically hard lower multilayer is magnetically fixed. In this sample, the large shift of 619 Oe indicates a strong AF coupling across the 11 Å NiO layer. (b) The magnetic force microscopy image at the remanent state ($H = 0$ Oe). Only closed domain walls are visible due to the AFM interlayer coupling across the thin NiO spacer [110].

in the thin range, the interlayer coupling oscillates between the FM and AFM nature (Figure 4). The oscillating period of ~ 5 Å is about two monolayer of the (111)-textured NiO spacer. Moreover, in the [Pt/Co] n /NiO/[Co/Pt] n multilayer with the strongest AFM interlayer coupling across a ultrathin NiO spacer of just 11 Å thickness, the measured temperature dependence of the biasing field H_{EB} shows that the blocking temperature T_B is as high as 250 K [110]. In the bilayer Ni/NiO (28 Å) system with the in-plane anisotropy, the measured T_B is just 38 K [112]. The [Pt/Co] $_3$ /NiO (11 Å) multilayer with perpendicular anisotropy is determined to have a T_B of 220 K [113]. This significantly enhanced T_B is highly dependent on the in-plane and out-of-plane anisotropy constants of the intervening antiferromagnet as proposed by Baruth and Adenwall [114]. Moreover, the AFM interlayer coupling should also lead to the further enhancement of T_B to 250 K. In the trilayer NiFe/NiO/Co with the in-plane anisotropy, Camarero et al. [115] have

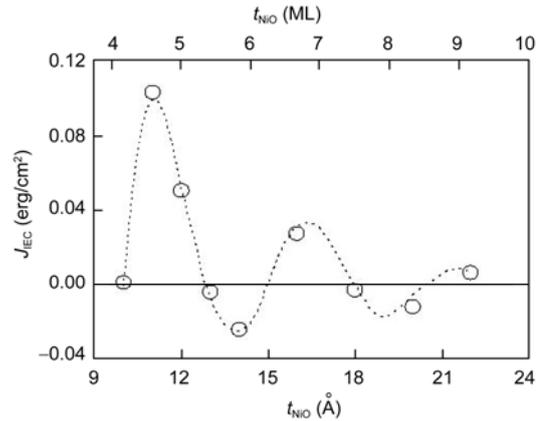


Figure 4 The interlayer coupling strength (J_{IEC}) as a function of NiO thickness (in units of both Å and ML) at room temperature. J_{IEC} is determined by where M_S and t_{FM} are the saturation magnetization and total thickness of all upper Co layers, respectively; H_{MLS} is the minor loop shift with $H_{MLS} > 0$ (< 0) corresponding to the AFM (FM) coupling. The dotted line is a guide to the eyes [110].

observed a 90° coupling across the NiO spacer between NiFe and Co layers, and the coupling exists for a NiO thickness up to 25 nm. By systematically varying the thicknesses of the bottom FM and the AFM layers, Kuch et al. [116] have successfully realized atomic-level control of the FM-AFM interface morphology. Using magneto-optical Kerr-effect measurements and layer-resolved spectro-microscopic magnetic domain imaging of single-crystalline FM/AFM/FM trilayers, they have observed the mediated magnetic coupling across the interface by step edges of single-atom height.

4 Electric-field control of EB via magnetoelectric (ME) effect

In the multiferroic materials with the coexistence of ferromagnetism and ferroelectricity (FE), ME coupling effect allows the electric-field control of magnetization and the magnetic-field control of the electric polarization. The potential applications of ME in a new generation of devices in spintronics have stimulated interest in the multiferroic materials in recent years. Research effort has been made to look for materials with the FE and FM coexistence. In nature, however, the single-phase compounds with the simultaneous FE and FM presence are scarce. In contrast, the single-phase compounds exhibiting the coupled FE and AFM behavior are relatively common, but they have received less attention. In combination with the FM layers, their AFM character can be used to induce EB. By the electric control of EB via the ME effect, the electrically tuning of the magnetization of the FM layer becomes possible.

Recently, Borisov et al. [117–119] have reported the successful electric-field control of EB in the Cr_2O_3 /[Co/Pt] n multilayers via the ME effect of the AFM Cr_2O_3 pinning

layer. This electrical control of EB has suggested new possibilities for tailoring magnetoresistive components with low power consumption.

The hexagonal oxides of YMnO_3 and LuMnO_3 are the multiferroic materials exhibiting the coexistence of the coupled FE and AFM characters. The recent investigations on their EB effect [120,121] have shown that an electric-field can be used to tune the EB in the FM/AFM heterostructures, thus manipulating the magnetic switching of the FM layer. Those results exhibit the promising perspectives of oxides with the coupled AFM and FE characters in spintronics, though the practical application of YMnO_3 and LuMnO_3 in spintronics is strongly restricted because their Néel temperatures are much lower than room temperature.

BiFeO_3 is the only multiferroic material that exhibits the higher Néel temperature than room temperature. The EB effect of BiFeO_3 has been studied recently in detail [122–127]. It has been pointed out that the EB of BiFeO_3 could be related to the FE and AFM domain structures [122–124], which can be manipulated by the electric-field [123–128]. Allibe et al. [128] have investigated the giant magnetoresistance (GMR) effect of a spin valve deposited on top of a BiFeO_3 layer. They have demonstrated the electrical manipulation of the EB and related resistance value in the spin valve.

5 Conclusion

In a little more than decade, significant progress has been made in the deeper understanding of the underlying mechanisms for the EB. The intensive investigations on the AFM thickness dependence of the EB have demonstrated that the exchange coupling at the FM-AFM interface involves not only the interfacial AFM spins but the spin structure in the AFM layer. The studies on the interlayer EB coupling across a nonmagnetic spacer between the FM and AFM layers have indicated that the EB is beyond the interfacial nature, most probably having the long-range characteristics. Via controlling the AFM spacer and its spin structure, it has been demonstrated that the interlayer coupling can be induced between the FM layers separated by an AFM spacer. By depositing the FM layer on top of the multiferroic layer with the simultaneous presence of AFM and FE behavior, recent studies have shown the electrical control of the EB and eventual electrical manipulation of the magnetization in the FM layer, demonstrating the promising applications of multiferroic materials in a new generation of electric-field controlled devices in the field of spintronics.

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