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## $\frac{11}{2}$  Deconotives of voltage control for magnetic exchange his in  $\frac{11}{12}$  Perspectives of voltage control for magnetic exchange bias in  $\frac{77}{78}$ <sup>13</sup> multiferroic heterostructures and the state of t  $14$

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#### 21 века в област в о  $22$   $\ldots$   $\ldots$   $22$   $\ldots$   $\ldots$   $28$ A R T I C L E I N F O A B S T R A C T

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23 Tarticle history: The Community of the Exchange bias, as an internal magnetic bias induced by a ferromagnetic–antiferromagnetic exchange and containing the containing the containing containing the containing containing 24 Received 1 December 2016 coupling, is extremely important in many magnetic applications such as memories, sensors and other <sup>25</sup> Received in revised form 17 January 2017 **devices. Voltage control of exchange bias in multiferroics provides an energy-efficient way to achieve a** 26 Accepted 30 January 2017<br>https://www.akaba.org/interministic switching of magnetization, which has been considered as a key challenge in 27 The magnetoelectric state with the Sampact and ultra-low power magnetoelectric memories and sensors. 93 28 94 Additionally, exchange bias can enhance dynamic magnetoelectric coupling strength in an external-field-29 95 free manner. In this paper, we provide a perspective on voltage control of exchange bias in different 30 96 multiferroic heterostructures. Brief mechanization and related experiments are discussed as well as future <sub>31</sub> Exchange bias trend and challenges that can be overcome by electrically tuning of exchange bias in state-of-the-art <sub>97</sub>  $\frac{32}{2}$ magnetoelectric devices.

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## **1. Introduction**

tus 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.](#page--1-0) Sections [3,](#page--1-0) [4,](#page--1-0) [5](#page--1-0) 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](#page--1-0) gives the scenarios for future trend of voltage controlled EB and an overall summary is displayed in Section [7.](#page--1-0)

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65 131 0375-9601/© 2017 Elsevier B.V. All rights reserved.66 and the contract of the con

### 37 103 *1.1. Introduction for EB effect: origin and physical mechanism*  $38$

39 105 40 Being able to maintain the stabilization of magnetization in the stabilization of the stabil 41 spin-valve structures, the exchange bias (EB) effect has become one  $\frac{1}{2}$  is the first first of the structure of the structures of the structures, the exchange bias (EB) effect has become one 42 of the most decisive factors in the excellent performance of spin-<br> $\frac{6}{3}$  of anticomplement (the Neel tank) and the most decisive in the step i 43 tronic devices [\[1\].](#page--1-0) For example, the EB effect plays an important  $\frac{1}{2}$  instruction in the EM layer and analyzes on FB  $^{44}$  role in the area of high-resolution magnetic sensors because of its  $^{45}$   $^{45}$   $^{46}$   $^{46}$   $^{46}$   $^{47}$   $^{48}$   $^{47}$   $^{48}$   $^{47}$   $^{48}$   $^{47}$   $^{48}$   $^{47}$   $^{48}$   $^{47}$   $^{48}$   $^{47}$   $^{48}$   $^{49}$   $^{49}$ 45 minimal heat dissipation at highest performance  $[2,3]$ . This review  $\frac{1}{2}$  and first discovered in 1056 by Meillaish at also for  $\frac{1}{2}$ . The case of  $\frac{1}{2}$ <sup>46</sup> intends to provide an overview of the technological perspectives when and we believed to be a series when the interface overboxe. <sup>47</sup> for the EB effect, one that manages to see the wide picture of  $\frac{113}{113}$  coupling  $\frac{1580}{113}$  Since the EB effect is important in both functionally <sup>48</sup> its origin, intrinsic properties, application prospects, research sta-<br>assay the information storme 10, 131 many different systems is a second to the information storme 10, 131 many different system  $\frac{49}{115}$  is origin, means to compute the information storage  $\frac{9-13}{15}$ , many different sys-<br> $\frac{49}{115}$  in any different sys- $\frac{100}{20}$  the initial contains the containing thems containing FM—AFM interfaces have been studied for three  $\frac{116}{116}$  $\frac{51}{12}$  in significant of voltage controllations of intensive estimates types [\[7\]:](#page--1-0) (1) core/shell nanoparticles (e.g. Co/CoO [\[14–18\],](#page--1-0) Ni/NiO  $\frac{117}{117}$  $52$  118 included in section 1, two general mannestations of methods  $[19-22]$ , Fe/FeO<sub>x</sub>  $[23-26]$ ; (2) non-uniform magnetic materials  $118$  $\frac{53}{118}$   $\frac{1}{18}$   $\frac{1}{18}$  $54$  100 13 5, 4, 5 present existing studies on perspectives for the de-<br> $\frac{1}{25,36}$ ; (3) bilayer and multilayer thin films [\[37–39\].](#page--1-0) It  $^{55}$  velopinent of voltage controlled Eb in unferent structural systems, is known that the EB effect strongly depends on the microstruc- $^{56}$  will not helight indicate the indicate the FM and AFM layers. As for the core/shell nanoparticles  $^{122}$  $_{57}$  alloy/piezoelectric laminate and FM/AFM/FE heterostructures. Sec-<br>with potential for the high-density memories, they are limited to  $_{123}$  $_{58}$  tion 6 gives the scenarios for future trend of voltage controlled EB the preparation technology which can hardly control the interface  $_{124}$  $_{59}$  and an overall summary is displayed in Section 7. The section properties precisely as well as the crystallinity of ultrathin AFM  $_{125}$  $_{60}$  shell layer  $[37,40]$ . And the non-uniform magnetic materials are  $_{126}$ 61 127 restricted for the random distribution of magnetic ions which will 62 128 *E-mail addresses:* [ziyaozhou@xjtu.edu.cn](mailto:ziyaozhou@xjtu.edu.cn) (Z. Zhou), [mingliu@mail.xjtu.edu.cn](mailto:mingliu@mail.xjtu.edu.cn) 63 (M. Liu). Music controllability of atomic layers can be realized in thin films [\[41–45\].](#page--1-0) 129 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 [\[2,4–6\].](#page--1-0) 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 [\[7\].](#page--1-0) 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 [\[5,8\].](#page--1-0) Since the EB effect is important in both fundamental generate the FM—AFM interfaces randomly [\[28\].](#page--1-0) However, the con-

<sup>(</sup>M. Liu).

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of films enable intensive studies for the EB effect [\[46–51\].](#page--1-0)

<sup>3</sup> Although EB effect has been studied for almost a half century, magnetic (FM) and ferroelectric (FE) orders, have gained tremen- <sup>69</sup> <sup>4</sup> the physical mechanism started to be understood for only two dous flurry of research interests [68–70,72–83,92–100]. Significant <sup>70</sup> <sup>5</sup> decades. Generally, researchers believe that the EB effect is ori- progress has been made in multiferroics, such as sensors, mag- <sup>71</sup>  $^6$  gin from the spin exchange coupling of the FM—AFM interface  $[8,$  netoelectric random access memories (MERAMs), etc.  $[101-105]$ .  $\frac{72}{2}$  $7$   $12,13$ ]. The pioneering EB papers of Meiklejohn et al. assumed an  $\;$  They are potentially the most promising way to overcome power  $\;$   $\;$   $^{73}$ 8 ideal interface and found the  $H_{EB} = (nJ S_{FM} \cdot S_{AFM})/(M_{FM} t_{FM})$  [\[8,12,](#page--1-0) and temperature issues in optimizing the performance of micro-<sup>9</sup> 13]. Where *n* is the number of interaction spins of FM (S<sub>FM</sub>) and electronic devices [\[106\].](#page--1-0) There are existing studies focusing on <sup>75</sup> <sup>10</sup> AFM (S<sub>AFM</sub>), *J* is the interaction intensity, M<sub>FM</sub> and t<sub>FM</sub> are the the development of novel ME couplings, the exploration of dif-<sup>11</sup> magnetization and thickness of FM layer, respectively. As indicated ferent E-field controlling mechanisms (e.g. the strain-mediated <sup>77</sup> <sup>12</sup> in this expression, the EB fields related to interfacial character are converse ME coupling [107–113], the manipulation of domain <sup>78</sup> <sup>13</sup> inversely proportional to the thickness of coupled FM layer  $(t_{FM})$ . structure [98,114–116], the interfacial charge mediated ME cou-<sup>14</sup> And the  $t_{FM}$  is associated with the types of FM materials and the pling [82,117,118]), and the E-field control of exchange coupling  $\frac{80}{2}$ <sup>15</sup> growth conditions, which demonstrates the EB effect obtains inter-  $[119]$ . Though the strain-mediated ME couplings in multiferroic  $81$ <sup>16</sup> facial properties. This model supposed that the interface is perfect heterostructures can achieve a large ME coupling, they are lim- <sup>82</sup> <sup>17</sup> and did not reflect the influence of AFM layer. However, the exper- ited for only realizing 90° magnetic rotations [120,121]. Recently, <sup>83</sup> <sup>18</sup> imental results have indicated that H<sub>EB</sub> is strongly affected by the a breakthrough is achieved by Wang et al. [\[122\].](#page--1-0) They realized <sup>84</sup> <sup>19</sup> thickness of AFM layer (t<sub>AFM</sub>) [52]. The H<sub>EB</sub> dependence of t<sub>AFM</sub> in 180° magnetic moment reversal through electrostriction-induced <sup>85</sup> <sup>20</sup> the multilayer thin films is highly related to the material system, magnetic anisotropy [122]. However, 180° deterministic switch- <sup>86</sup> <sup>21</sup> the assignment of configuration, the microstructure and the exper- ing still needs to be further explored for it is essentially re- <sup>87</sup> <sup>22</sup> iment temperature [\[52–59\].](#page--1-0) Despite the complexity, the behavior quired to satisfy the memory switching requirement [123]. The  $^{88}$  $^{23}$  can be primarily divided into two types: (1)  $H_{\rm EB}$  increased parallel mentioned voltage control of FM/AFM EB proposed in multifer-  $^{89}$ <sup>24</sup> with *t<sub>AFM</sub>* and reaches its saturation point when the *t<sub>AFM</sub>* is large roics offers another prospective way for 180° magnetism switching <sup>90</sup> <sup>25</sup> enough  $[52-59]$ ; (2)  $H_{EB}$  declined after reaching the peak with  $[124]$ . <sup>26</sup> an increasing t<sub>AFM</sub> [\[52,53,55,58–60\].](#page--1-0) Since the domain structure of In 2005 Binek and Doudin [125] suggested a focus on exchange <sup>92</sup> <sup>27</sup> AFM layer is hard to observe, the exchange coupling between FM bias with E-field for the achievement of energy-efficient MER- <sup>93</sup>  $^{28}$  and AFM layers can help to reflect it indirectly by the detection of AMs. This led to the invention of MERAM by Chen et al. [66].  $^{94}$ <sup>29</sup> domain structure variation in FM layer [\[52\].](#page--1-0) Yang et al. studied the Since then many activities in the search for EB using magneto- <sup>95</sup> <sup>30</sup> changes of H<sub>EB</sub> and AFM-layer domain structure based on permal- electric materials have been triggered. In 2006, Kleemann's group <sup>96</sup> <sup>31</sup> loy (200 Å)/FeMn (t<sub>AFM</sub>)/Co(100 Å) structure in 2000 [\[61\].](#page--1-0) And [66,126] proposed a concept of MERAMs which conformed that <sup>97</sup> <sup>32</sup> the observed spiraling domain structure indicated that the domain purely electric control is possible by measuring the magnetoresis-  $98$ <sup>33</sup> structure of AFM layer is vital to the exchange coupling [\[61\].](#page--1-0) In tance of an exchange coupled spin valve. In 2009, Bibes et al. [105] <sup>99</sup> <sup>34</sup> 2009, Morales et al. delved into Ni (50 nm)/FeF<sub>2</sub> (*t*<sub>AFM</sub>)/Permalloy proposed a typical MERAM as shown in Fig. 1, in which, the bot-<sup>100</sup> <sup>35</sup> (50 nm), finding that besides FM—AFM interfacial properties the tom FM layer of magnetic tunneling junction (MTJ) is coupled to <sup>101</sup> <sup>36</sup> antiferromagnetic bulk spin structure also contributes to the EB AFM–FE layer. By switching the polarization 180°, the AFM vec- <sup>102</sup> 37 effect [62]. Two switch 180° and then drive the FM layer reversal 180°. Two 103 Although EB effect has been studied for almost a half century, [12,13\].](#page--1-0) The pioneering EB papers of Meiklejohn et al. assumed an [13\].](#page--1-0) Where *n* is the number of interaction spins of FM  $(S_{FM})$  and growth conditions, which demonstrates the EB effect obtains interimental results have indicated that *H*<sub>EB</sub> is strongly affected by the thickness of AFM layer  $(t_{AFM})$  [\[52\].](#page--1-0) The  $H_{EB}$  dependence of  $t_{AFM}$  in changes of *H*<sub>EB</sub> and AFM-layer domain structure based on permaleffect [\[62\].](#page--1-0)

<sup>38</sup> Actually, the appearance of EB effect requires the AFM layer resistive states (parallel and antiparallel) are created by voltage- <sup>104</sup> <sup>39</sup> having strong anisotropy field magnetic anisotropy [\[7\].](#page--1-0) Only if the assisted 180° magnetization reversal through exchange bias. In <sup>105</sup> <sup>40</sup>  $t$ <sub>AFM</sub>  $K$ <sub>AFM</sub> ≥ *J*, the exchange coupling can happen [\[7\].](#page--1-0) Where  $K$ <sub>AFM</sub> 2010, a stress-mediated voltage controlled EB in multiferroic het- <sup>106</sup> <sup>41</sup> is the anisotropic constant relating to  $t_{\text{AFM}}$  and temperature, and erostructure has first been demonstrated by Polisetty et al. [4] and <sup>107</sup> <sup>42</sup> J is the interaction intensity.  $K_{\rm AFM}$  decreases with reduced  $t_{\rm AFM}$ , gave a novel method for the EB regulation. Finally, in 2014, Street <sup>108</sup> <sup>43</sup> and the EB effect will disappear if the K<sub>AFM</sub> is too weak despite et al. [127] reported an important technical breakthrough by dop- <sup>109</sup> <sup>44</sup> of the existing interfacial spin exchange coupling [\[7\].](#page--1-0) Moreover, ing the magnetoelectric control layer, Cr<sub>2</sub>O<sub>3</sub>, with 3% of B<sub>2</sub>O<sub>3</sub>. This <sup>110</sup>  $^{45}$  the EB effect is closely related to temperature, which can affect crucially enhances the Néel temperature from 307 K to  ${\sim}400$  K  $^{-111}$ <sup>46</sup> the pinning ability of AFM layer to FM layer. As temperature goes and thus paves the way for technically acceptable EB-based spin- <sup>112</sup> <sup>47</sup> lower, the *K*<sub>AFM</sub> value becomes higher, and the EB effect increases tronic devices in the near future.  $48$  [\[7\].](#page--1-0) Researchers have found that if the temperature is higher than Nevertheless, there are still many challenges in the voltage con-<br> <sup>49</sup> a critical temperature ( $T_{\rm B}$ ),  $H_{\rm EB}$  comes to zero [\[63,64\].](#page--1-0) We may trol of EB. The complex switching mechanisms, the challenges in <sup>115</sup>  $^{50}$  naturally think that  $T_B$  is associated with Neel temperature,  $T_N$ , understanding the physics behind this interface phenomena and <sup>116</sup> Actually, the appearance of EB effect requires the AFM layer 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

<sup>1</sup> The sophisticated preparation technologies and analysis methods stress on the overlayer's magnetic phase through ME coupling [91]. <sup>67</sup> 2 of films enable intensive studies for the EB effect [46–51]. Therefore, multiferroic materials, simultaneously occupying ferro-  $68$ stress on the overlayer's magnetic phase through ME coupling [\[91\].](#page--1-0) dous flurry of research interests [\[68–70,72–83,92–100\].](#page--1-0) Significant progress has been made in multiferroics, such as sensors, magnetoelectric random access memories (MERAMs), etc. [\[101–105\].](#page--1-0) and temperature issues in optimizing the performance of microthe development of novel ME couplings, the exploration of different E-field controlling mechanisms (e.g. the strain-mediated converse ME coupling [\[107–113\],](#page--1-0) the manipulation of domain structure [\[98,114–116\],](#page--1-0) the interfacial charge mediated ME coupling [\[82,117,118\]\)](#page--1-0), and the E-field control of exchange coupling heterostructures can achieve a large ME coupling, they are limited for only realizing 90◦ magnetic rotations [\[120,121\].](#page--1-0) Recently, magnetic anisotropy [\[122\].](#page--1-0) However, 180◦ deterministic switching still needs to be further explored for it is essentially required to satisfy the memory switching requirement [\[123\].](#page--1-0) The mentioned voltage control of FM/AFM EB proposed in multiferroics offers another prospective way for 180◦ magnetism switching [\[124\].](#page--1-0)

In 2005 Binek and Doudin [\[125\]](#page--1-0) suggested a focus on exchange bias with E-field for the achievement of energy-efficient MER-AMs. This led to the invention of MERAM by Chen et al. [\[66\].](#page--1-0) Since then many activities in the search for EB using magneto-[\[66,126\]](#page--1-0) 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. [\[105\]](#page--1-0) proposed a typical MERAM as shown in [Fig. 1,](#page--1-0) in which, the bottom FM layer of magnetic tunneling junction (MTJ) is coupled to AFM–FE layer. By switching the polarization 180◦, the AFM vecassisted 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. [\[4\]](#page--1-0) and gave a novel method for the EB regulation. Finally, in 2014, Street et al. [\[127\]](#page--1-0) reported an important technical breakthrough by doping the magnetoelectric control layer,  $Cr<sub>2</sub>O<sub>3</sub>$ , with 3% of  $B<sub>2</sub>O<sub>3</sub>$ . 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.

<sup>51</sup> nevertheless, the exactly relationship remains to be discovered. The significant disadvantages caused by the presence of an ex- <sup>117</sup> 52 118 ternal magnetic bias field (e.g., decrease of spatial resolution and 118 <sup>53</sup> 1.2. Voltage controlled EB effect in multiferroics: evolution and signal-to-noise ratio) have made the utilization of EB effect a tech-<sup>119</sup> 54 challenge **120** 120 and the mologically difficult area both theoretically and experimentally  $[2, 120]$  $[2, 120]$ 55 121 [65,123\].](#page--1-0) Thus, over the past decade, numerous efforts have been <sup>56</sup> In recent years, magnetoelectric (ME) couplings have been put into the investigation of controlling EB [\[2,120,128,129\].](#page--1-0) To the <sup>122</sup> <sup>57</sup> introduced to FM–AFM structures and voltage controllable EB best of our knowledge, although there are existing reviews related <sup>123</sup> 58 systems provide a new perspective as a novel control method to the EB systems  $[7,10,11,130]$ , few of them are target at giving  $124$ 59 [\[65–67\].](#page--1-0) Simultaneously occupying ferromagnetic (FM) and ferro- a detailed introduction for the voltage control of EB effect based 125  $60$  electric (FE) orders, multiferroic materials manage to manipulate on multiferroics. It would, therefore, be interesting to provide an  $126$  $61$  magnetism by an electric field (E-field) or vice versa [\[68–85\].](#page--1-0) overview of the technological perspectives for voltage controlled  $127$ <sup>62</sup> This means that H<sub>EB</sub> (magnetic property) can be regulated with LEB effects. In this paper, we will present the remarkable progress <sup>128</sup>  $^{63}$  E-field, and paves way for non-volatile, lightweight, and energy- in E-field control of magnetism in multiferroics achieved in recent  $^{129}$  $64$  efficient magnetic memories [\[86–90\].](#page--1-0) When the E-field is applied few years, and discuss the challenges remain to be solved. Sev-  $130$ <sup>65</sup> across the thickness direction of FE substrate, a strain on the eral material systems and tuning strategies such as voltage control <sup>131</sup> <sup>66</sup> piezoelectric phase of this substrate is generated, leading to a cof EB in FM/multiferroic oxides heterostructures or in ferromag- <sup>132</sup> 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 best of our knowledge, although there are existing reviews related to the EB systems [\[7,10,11,130\],](#page--1-0) 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 ferromagDownload English Version:

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