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Note: This paper was presented at the 64th Annual Conference on Magnetism and Magnetic Materials.

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ABSTRACT
Two types of electric control of exchange bias (EB) by resistive switching (RS), i.e. conductive-filament-RS (type I) and interface-barrier-RS (type II) were observed in the Si/SiO_2/Ti/Pt/FeO_x/Co/ITO multilayer devices, which were fabricated by magnetron sputtering. It is difficult for the type I device to control EB, which may be due to that the quantity of conductive filaments is not enough to modify the antiferromagnetic structure of FeO_x near the Co/FeO_x interface. However, the electric control of EB can be accomplished in the type II device. Compared with low-resistance-state (LRS), the exchange bias field (H_E) increases a little but the coercivity (H_C) increases significantly at high-resistance-state (HRS). We consider that the migration of the oxygen vacancies under different voltages is able to mediate the interfacial barrier height, leading to the bipolar RS effect and the change of EB as well. This provides a way for designing new types of spintronic devices based on electric control.

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INTRODUCTION
Electric control of exchange bias (EB) in ferromagnet/antiferromagnet (FM/AFM) bilayer provides a pathway for full electric control of magnetism, which is of great importance in developing energy-efficient spintronic devices. Although in the past decade people have made many attempts to explore the effect in various materials and devices, each method has its own limitation, such as irreversibility, volatility, operation at quite low temperatures and large voltage, demand for special substrates and so on. Very recently, people have proposed a new way to realize non-volatile, repeatable electric control of EB at room temperature (RT) by virtue of resistive switching (RS) with an antiferromagnetic resistive layer. By applying certain voltages, the device exhibits different EB effects at high-resistance-state (HRS) and low-resistance-state (LRS). Conductive filaments forming in the AFM layer and rupturing near the FM-AFM interface are considered to result in the combined RS and EB phenomena. However, this kind of electric control of EB works only if high-density magnetic filaments can be formed and ruptured at the FM-AFM interface during the RS process. Moreover, the choice of materials is strongly limited because specific oxides (e.g. NiO) are required for the AFM layer.

Apart from the conductive filament mechanism, it is well known that the RS behavior can also be triggered by changing the interfacial barrier height after applying appropriate voltages. However, whether or not the EB can be controlled by interface-barrier-type RS effect has remained unknown. To our knowledge, α-Fe_2O_3 is a typical RS material, which often demonstrates bipolar RS behavior with the interface-barrier-mediation mechanism. On the other hand, α-Fe_2O_3 is also a typical antiferromagnetic material with very high Néel temperature (∼955 K) and much
significant EB effect can be obtained in FM/α-Fe₂O₃ bilayers.\textsuperscript{16,17} Therefore, in this work, iron-oxide (FeO\textsubscript{x})/Co (x ≤ 1.5) bilayers were chosen for exploring the electric control of EB based on the RS effect. By changing the value of x, both the conductive-filament-RS (type I) and interface-barrier-RS (type II) can be accomplished and the corresponding electric controls of EB have been investigated.

**EXPERIMENTAL DETAILS**

The device’s stacking structure is schematically displayed in Fig. 1(a). The Ti/Pt/FeO\textsubscript{x}/Co/ITO multilayer films were deposited by magnetron sputtering at room temperature on Si (100) substrate with native oxide on surface. The base pressure was lower than 5.0×10⁻⁶ Pa and the Ar pressure was kept at 0.3 Pa during the film deposition. First, a 2 nm-thick Ti film and a 50 nm-thick Pt film were deposited successively on the substrate as the bottom electrode. Afterwards, the FeO\textsubscript{x} layer was deposited by using a α-Fe₂O₃ commercial target with mixed gases of Argon and Oxygen. Two different FeO\textsubscript{x} films, i.e. FeO\textsubscript{x}(1) and FeO\textsubscript{x}(2), were made at lower (P\textsubscript{O₂}: P\textsubscript{Ar} = 1 : 2) and higher (P\textsubscript{O₂}: P\textsubscript{Ar} = 2 : 3) oxygen partial pressures, respectively. Finally, for making the top electrode, a 2.6 nm-thick Co layer and a transparent 70 nm-thick ITO film were deposited successively on the FeO\textsubscript{x} layer by using shadow mask in circular shape with the diameter of about 160 μm. All the thicknesses mentioned above are nominal. To establish EB in the FeO\textsubscript{x}/Co bilayer, a constant magnetic field of about 200 Oe was applied parallel to the film plane during the film deposition.

The RS behavior was characterized by a Keithley-2400 meter with the wires’ connection schematically shown in Fig. 1(a). The microstructure was characterized by a JEOL 2200FS TEM equipped with an Omega filter for energy-filtered imaging, working at 200 kV accelerating voltage. The in-plane magnetic hysteresis (M-H) loops were measured by a SQUID-VSM (Quantum Design) and a commercial focused magneto-optical Kerr effect magnetometer (FMOKE, NanoMOKE3) with the spot size of about 100 μm. All the measurements and characterizations were performed at room temperature.

**RESULTS AND DISCUSSION**

The bright-field low-resolution cross-sectional TEM image of a Si/SiO\textsubscript{2}/Ti/Pt/FeO\textsubscript{x}(2)/Co/ITO multilayer device set at LRS is shown in Fig. 1(b), which exhibits good stacking structure and sharp interfaces. From Fig. 1(b), the thicknesses of Pt, FeO\textsubscript{x} and ITO layers are measured to be about 50.3, 65.3 and 67 nm, respectively, in good agreement with the nominal ones. Fig. 1(c) and 1(d) show the high-resolution TEM (HRTEM) images at two different positions near the Co-FeO\textsubscript{x}(2) interface for the same device as that described in Fig. 1(b). As shown in Fig. 1(c), the thickness of Co layer is about 2.6 nm, which is identical to the nominal value. Moreover, as displayed in Fig. 1(c) and 1(d), different orientations of lattice fringes in the FeO\textsubscript{x}(2) layers indicates its polycrystalline structure. From the dotted yellow area in Fig. 1(c) and 1(d), the interplanar spacing are both measured to be about 0.2520 nm, corresponding to the (110) orientation of α-Fe₂O₃. This indicates that the FeO\textsubscript{x}(2) layer, grown under higher oxygen partial pressure,
has been almost fully oxidized to stoichiometric $\alpha$-$\text{Fe}_2\text{O}_3$. On the other hand, the FeO$_x$(2) layer has a prominent (110) orientation, which plays a crucial role in producing exchange bias in the FM/$\alpha$-$\text{Fe}_2\text{O}_3$ bilayer. Although TEM characterization has not been performed in the device with FeO$_x$(1) layer, it is expected that similar results will be obtained except that more oxygen vacancies may exist in the FeO$_x$(1) layer.

Because the magnetic property may also reflect the oxidation level or the concentration of oxygen vacancy for the FeO$_x$ layer, FeO$_x$(1) and FeO$_x$(2) single layer film samples were fabricated on the same substrates and at the same conditions as those for the multilayer device, and their field dependences of magnetization were measured by SQUID-VSM. As shown in Fig. 2(a), the magnetic moments for those two sample are very small. After deduction of the linear diamagnetic background signals, two weak $M$-$H$ loops (see Fig. 2(b)) can be observed with the saturation magnetization of about several emu/cm$^3$, which is much smaller than Fe$_3$O$_4$ ($\sim 415$ emu/cm$^3$) or $\gamma$-$\text{Fe}_2\text{O}_3$ ($\sim 262.7$ emu/cm$^3$) at room temperature, suggesting that FeO$_x$(1) and FeO$_x$(2) films are both nearly antiferromagnetic. Even so, it can be seen that the magnetization of FeO$_x$(1) is obviously larger than that of FeO$_x$(2), implying that the oxidation level of FeO$_x$(1) is less than that of FeO$_x$(2). Fig. 2(c) and 2(d) display the $M$-$H$ loops of the Ti/Pt/FeO$_x$(1)/Co/ITO multilayer film samples with FeO$_x$(2) and FeO$_x$(1) layers at various temperatures, respectively. Obviously, the FeO$_x$(2)/Co bilayer exhibits larger exchange bias field ($H_E$) and coercivity ($H_C$) than those for the FeO$_x$(1)/Co bilayer. For example, at room temperature, $H_E/H_C$ equals to 138 Oe/386 Oe for the former and 47 Oe/172 Oe for the latter. With increasing the temperature, $H_E/H_C$ decreases slowly for the former while much faster for the latter, as shown in Fig. 2(e) and 2(f). At 400 K, $H_E$ holds at 74 Oe for the former while approaches to zero for the latter, implying that FeO$_x$(2) layer has larger blocking temperature in comparison with FeO$_x$(1) layer. All these above results indicate that FeO$_x$(2) is close to stoichiometric ($x \approx 1.5$) while FeO$_x$(1) is less stoichiometric ($x < 1.5$) and may contain more oxygen vacancies.

![Figure 2](https://example.com/fig2.png)

**FIG. 2.** The field dependences of magnetization for the Si/SiO$_2$/Ti/Pt/FeO$_x$(1) film (in red) and Si/SiO$_2$/Ti/Pt/FeO$_x$(2) film (in black) before (a) and after (b) deduction of the linear background. The $M$-$H$ loops for the Si/SiO$_2$/Ti/Pt/FeO$_x$/Co/ITO film samples with FeO$_x$(2) (c) and FeO$_x$(1) (d) layers at $T = 300$ K, 350 K and 400 K, respectively. The temperature dependences of $H_C$ and $H_E$ for the Si/SiO$_2$/Ti/Pt/FeO$_x$/Co/ITO film samples with FeO$_x$(2) (e) and FeO$_x$(1) (f) layers.

Figure 3(a) shows a typical unipolar RS behavior in the Si/SiO$_2$/Ti/Pt/FeO$_x$(2)/Co/ITO device, indicating the cell switched from HRS to LRS or vice versa without changing the voltage.
polarity. The current-voltage (I-V) curves are displayed in both linear (Fig. 3(a)) and semi-logarithmic scales (Fig. 3(b)). The current increase is limited by a current compliance (CC) with its value denoted as $I_{CC}$. The device will be permanently damaged if $I_{CC}$ is too large, while the RS process may not be triggered with too small $I_{CC}$ due to not enough voltage applied. Therefore, in order to well characterize the RS behavior, appropriate $I_{CC}$ needs to be used during the I-V curve measurement. After a lot of tests, $I_{CC}$ was finally set to be 10 mA and 1 mA for the two devices discussed in Fig. 3 and Fig. 4, respectively. From Fig. 3(b), one can see three obvious
processes of ‘forming’, ‘reset’ and ‘set’ with the corresponding voltages of about 7.4 V, 0.9 V and 3.3 V, respectively. The resistance difference between HRS and LRS is significant with the on/off ratio of about 1000 at the read voltage of 0.1 V. This kind of RS behavior is often explained by conductive filament model,\textsuperscript{23,24} which is also applied in NiO\textsuperscript{23,25} and some other oxides.\textsuperscript{23,24} In this work, this kind of unipolar RS effect is called ‘type I’. Fig. 3(c) shows the M–H loops measured by FMOKE when the device is at HRS and LRS, and no obvious change of $H_E$ and $H_C$ (see Fig. 3(d)) can be observed in this type I device. This is most possibly due to that the quantity of conductive filaments formed at LRS is not enough to modify the AFM structure near the Co–FeO$_x$ interface and thus the change of EB is negligible. It is noted that no obvious conductive filaments can be easily observed in the HRTEM images in Fig. 1(c) and Fig. 1(d), also supporting this argument.

Figure 4(a) shows a typical bipolar RS behavior in the Si/SiO$_2$/Ti/Pt/FeO$_x$(1)/Co/ITO device, indicating that switching from HRS to LRS or vice versa needs to be operated upon changing the voltage polarity. The $I$–$V$ curves are displayed in both linear (Fig. 4(a)) and semi-logarithmic scales (Fig. 4(b)) with $I_{CC}$ set to be 1.0 mA. From Fig. 4(b), it can be seen that the corresponding voltages for ’set’ and ’reset’ process are about $-4.2$ and $3.2$ V, respectively. The on/off ratio is larger than 1000 at the read voltage of 0.1 V. Such bipolar RS effect is called ‘type II’, whose mechanism is attributed to mediation of the interface-barrier-height under certain voltages.\textsuperscript{11,12} Fig. 4(c) shows $M$–$H$ loops measured by FMOKE when the device is at LRS and HRS, which shows that the $M$–$H$ loop is obviously broadened after the device switches from LRS to HRS. As displayed in Fig. 4(d), $H_E/H_C$ equals to 116 Oe/66 Oe and 123 Oe/87 Oe at LRS and HRS, respectively. The corresponding voltages for ‘set’ and ‘reset’ processes are about $-E_8$ and $-E_7$, respectively. The on/off ratio is larger than 1000 at the read voltage of 0.1 V. Such bipolar RS effect is called ‘type II’. The inherent mechanism responsible for the latter case can not be fully understood at present and needs further studies. This work provides a new way for electric control of EB, which is potentially used in new generation spintronic devices.

**ACKNOWLEDGMENTS**

This work was supported by National Key Research and Development Program of China (2016YFA0300803), National Natural Science Foundations of China (Nos. 51971109, 51771053, 51471085), the Fundamental Research Funds for the Central Universities, and the open research fund of Key Laboratory of MEMS of Ministry of Education, Southeast University.

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