Electric field modulation of exchange bias at the Co/CoO_x interface

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We demonstrated the electric field (EF) effect on exchange bias (EB) in a perpendicularly magnetized Co/CoO_x layered structure. The antiferromagnetic CoO_x layer was formed by naturally oxidizing the Co surface. The modulation of the EB field and the coercivity by gate voltage application through a dielectric layer was clearly observed below the blocking temperature T_B. The modulation ratio of the EB field exhibited strong temperature dependence, and it increased as the temperature approached T_B. One possible cause of the EB modulation detailed herein is the modulation of the electronic state at the Co/CoO_x interface, which is fundamentally different from the case in which multiferroic antiferromagnets are used.

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I. INTRODUCTION

In layered structures with a ferromagnet (FM)/ antiferromagnet (AFM) interface, exchange bias (EB) [1] is induced by field cooling (FC) below the Néel temperature T_N of the AFM. The EB effect originates from the exchange interaction between atoms across the FM/AFM interface and adds a unidirectional magnetic anisotropy (MA) in the layered FM [2,3]. In giant or tunnel magnetoresistive devices, the EB is often utilized to fix the magnetization direction of the magnetization pinned layer [4–6]. Moreover, EB assists magnetization switching by the current-induced spin-orbit torque [7].

According to the Meiklejohn-Bean (MB) model [1,2], which is used generally to describe the EB effect, the magnitude of EB ($|H_{EB}|$) is expressed as $|H_{EB}| = K_J/tM_s = |Js_{FM}s_{AFM}|/tM_s$, where J is the exchange constant between FM and AFM spins, s_{FM} (s_{AFM}) is the spin vector of FM (AFM), t is the (effective) FM thickness, and M_s is the saturation magnetization. Thus the EB effect may be manipulated by controlling these magnetic properties. Application of an electric field (EF) on a FM/AFM system is one of the ways to manipulate EB. This has been demonstrated in systems with antiferromagnetic materials, which show the magnetoelectric (ME) effect [8–12]. In such materials, the EF application directly modulates s_{AFM} through the ME coupling, which is the internal coupling between the antiferromagnetic and ferroelectric orders, resulting in a change in the H_{EB}. EB control based on the above mechanism is impossible in systems such as the CoO/FM and NiO/FM (CoO and NiO are AFM) because there is no ME effect [10].

However, the CoO is an insulator; thus, the magnetic properties at the CoO/FM interface can be controlled via EF gating [13–16]. In this method, modulating the electronic structure at the FM/insulator interface leads to changes in their magnetic properties [17–21]. Therefore, by EF gating, EB is expected to be modulated in these systems without using the ME effect. In this manuscript, we show that EB in a Co thin film with a naturally oxidized antiferromagnetic CoO_x layer can be modulated by EF application [see Fig. 1(a)]. The EB modulation was not caused by redox reactions [22,23], suggesting that EF-induced modulation of the electronic state, including charge doping at the FM/AFM interface, causes changes in EB. This scenario is fundamentally different from that of the ME effect-driven case.

II. EXPERIMENTAL DETAILS

The layered structure used in this experiment is shown in Fig. 1(b). Ta(3.3 nm)/Pt(3.0)/Co(1.0) layers were deposited on a thermally oxidized Si substrate using RF sputtering. We have observed that the Pt underlayer has an fcc (111) texture [24,25]. Therefore the Co layer on fcc (111)-Pt is predicted to have an fcc (111) texture. The sample was exposed to air for 10 min to naturally oxidize the Co surface [26]. The oxidation state has been checked by using x-ray absorption spectroscopy in Ref. [27]. To prevent further oxidization, the sample was covered with a 5-nm-thick HfO_2 layer, formed at 150 °C in an atomic layer deposition (ALD) chamber. Because of the interfacial MA caused by orbital hybridizations of Pt-Co [28] and Co-O [29], the sample has perpendicular MA. The areal saturation magnetic moment tM_s of the oxidized sample is 0.39 mA at 300 K, which was determined using a superconducting quantum interference device. Because the tM_s for pure metallic Co film with a thickness of 1 nm is 1.65 mA at 300 K [25,26], the thicknesses of the nonoxidized (metallic) Co and CoO_x layers are estimated to be 0.24 and 0.76 nm, respectively [26]. The Curie temperature T_C for the sample after the oxidation process is ∼370 K, which was determined by the temperature T dependence of the M_s (not shown). The sample was patterned into a Hall bar structure with a 30-μm-wide channel by photolithography and Argon (Ar) ion milling. The sample was again covered with a 40-nm

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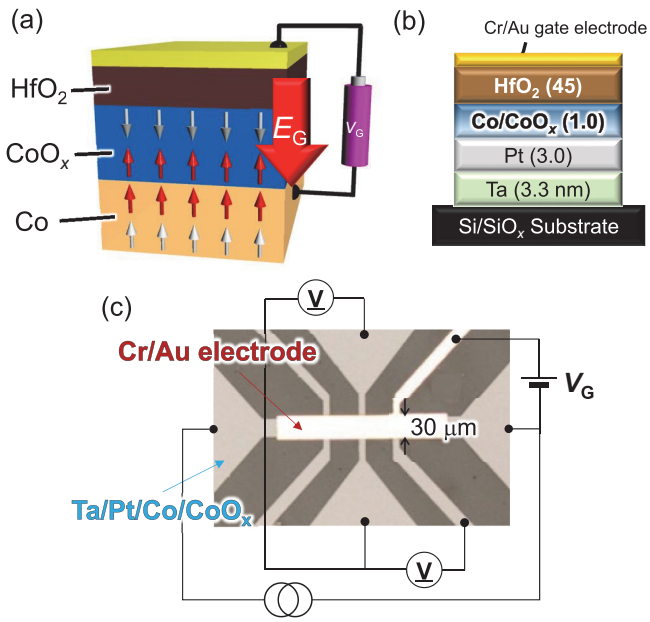

FIG. 1. (a) Schematic of the device. The gate electric field modifies the exchange coupling at the Co/CoO_x interface. (b) Layered structure. (c) Optical image of the fabricated micro-wire. The wire has contact pads for Hall and sheet resistance measurements.

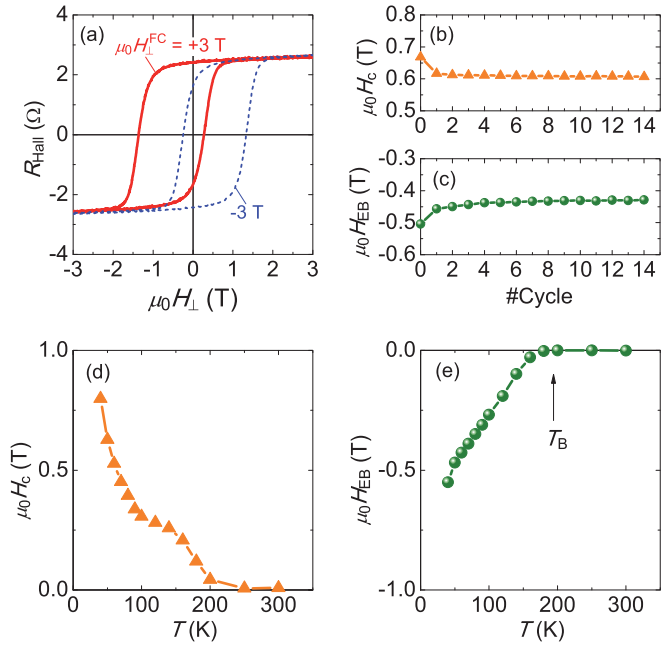

FIG. 2. (a) R_{Hall} hysteresis loops at 40 K for $\mu_0 H_\perp^{FC} = \pm 3$ T and $V_G = 0$ V. (b) H_c and (c) H_{EB} at 60 K plotted as a function of the number of Hall loop measurements. [(d) and (e)] T dependences of H_c and H_{EB} for $V_G = 0$ V, respectively.

HfO_2 gate insulator using the ALD technique at 150 °C. Finally, a Cr(2)/Au(10) counter electrode was formed on top of the channel through the gate insulator layer. In this study, the application of a positive (negative) gate voltage V_G is defined as the direction of increase (decrease) in the electron density at the Co surface. The V_G application is expected to cause charging/discharging at the topmost Co surface because CoO_x is an insulating material. The EF effect on the EB was primarily investigated using the anomalous Hall resistance R_{Hall} measurement, where R_{Hall} is proportional to the perpendicular component of the magnetization. A dc current of 100 μA was used to measure R_{Hall}.

III. RESULTS AND DISCUSSION

EB was introduced by the FC from 300 K, which is higher than the T_N of bulk CoO_x (~290 K) [30,31]. An external magnetic field during cooling ($H_\perp^{FC}$), in a direction parallel to the magnetic easy axis (perpendicular direction) was used for the FC process. The R_{Hall} loops for $\mu_0 H_\perp^{FC} = \pm 3$ T at $T = 40$ K are shown in Fig. 2(a). The loops were measured by sweeping the perpendicular magnetic field $H_\perp$ at $V_G = 0$ V. The center of the hysteresis loops clearly shifts toward the negative (positive) $H_\perp$ direction for the case of (positive) $H_\perp^{FC}$, indicating that the perpendicular EB effect at the Co/CoO_x interface certainly exists. To determine accurate values of the coercivity H_c and the exchange bias field H_{EB}, the hysteresis loops were repeatedly measured 15 times, and the final values were employed to rule out the training effect [see Figs. 2(b) and 2(c)]. We define H_c and H_{EB} as $(H_1 - H_2)/2$ and $(H_1 + H_2)/2$, respectively, where $H_{1(2)}$ is the magnetic field at which the up-(down-) swept R_{Hall} curve crosses the horizontal axis. Figures 2(d) and 2(e) display H_c and H_{EB} defined as a function of T, respectively. $|H_{EB}|$ as well as H_c decreases with increasing T. When $T > 200$ K, H_{EB} becomes zero, indicating that the blocking temperature T_B at which EB vanishes was approximately 200 K.

Next, the EF effect on EB was examined. Figure 3(a) shows the hysteresis curves at 50 K for $V_G = \pm 10$ V, obtained after the FC process ($\mu_0 H_\perp^{FC} = +3$ T). H_c at $V_G = +10$ V is larger than that at -10 V [26]. Although the V_G dependence of H_c is somewhat nonlinear [32,33], it shows monotonous change. More importantly, H_{EB} for $V_G = +10$ and -10 V, which are indicated by the dashed vertical lines, are different, indicating that the application of V_G modulates H_{EB} as well. H_c and H_{EB} as a function of V_G at 50 K are shown in Figs. 3(b) and 3(c). Both monotonically depend on V_G. In order to check the volatility of the present EF effect, we performed the repetitive Hall measurement by changing the V_{G-} value at 40 K. Figures 3(d)–3(g) shows V_G, the difference in $H_c[\mu_0 \delta H_c = \mu_0 H_c(N) \mu_0 H_c(N = 14)]$, $H_{EB}[\mu_0 \delta H_{EB} = \mu_0 H_{EB}(N) \mu_0 H_{EB}(N = 14)]$, and the sheet resistance R_{sheet} as a function of the cycle number N for each Hall measurement. H_c, H_{EB}, and R_{sheet} change rapidly according to V_G change and no hysteresis is observed—i.e., the changes in H_c and H_{EB} are completely reversible. Thus the EF effect is volatile.

The differences in H_c and H_{EB} between $V_G = +10$ and -10 V (ΔH_c and ΔH_{EB}) are +88.3 and −10.6 mT at 50 K, respectively. Figure 3(h) shows ΔH_{EB} as a function of T. In the employed T range, positive (negative) V_G always results in a H_{EB} increase (decrease), i.e., the sign of the H_{EB} change is independent of T. Although the magnitude of ΔH_{EB} decreases abruptly to zero as T approaches T_B (~200 K), it is almost independent of T below 150 K. In contrast, H_{EB} itself shows

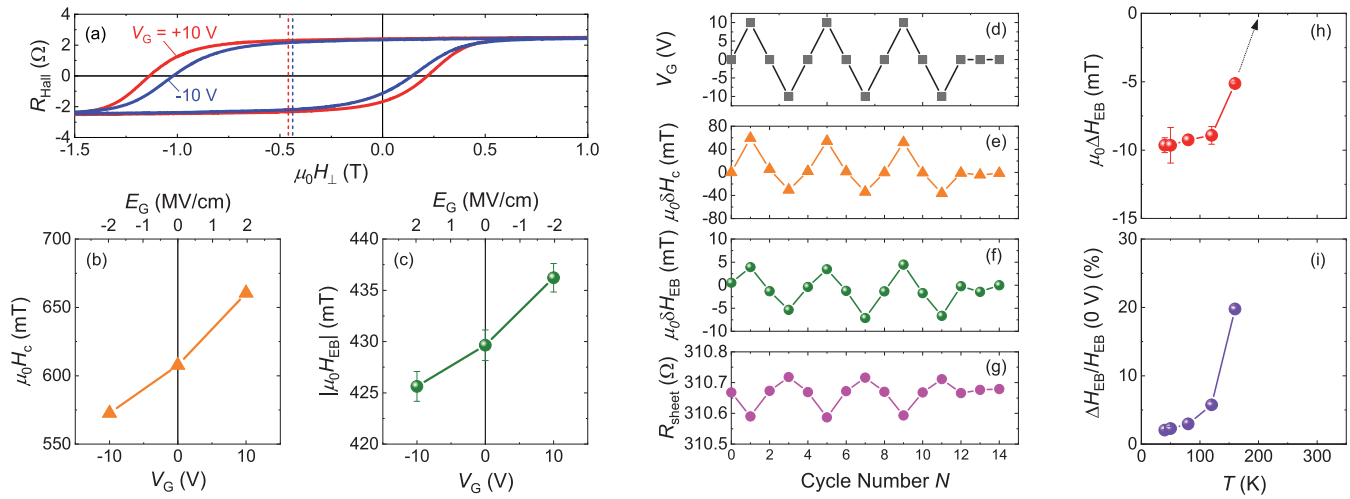

FIG. 3. (a) Results of gating experiments at 50 K for $\mu_0 H_\perp{}^{FC} = +3$ T. (b) H_c and (c) H_{EB} as a function of V_G and gate electric field E_G; E_G is expressed as $V_G/(t_{HfO2} + t_{CoOx})$, where the t_{HfO2} and t_{CoOx} represent the thickness of each insulating layer. [(d)–(g)] V_G, $\mu_0 \delta H_c$, $\mu_0 \delta H_{EB}$, and R_{sheet} as a function of the cycle number N for each Hall measurement. [(h) and (i)] T dependence of the change in H_{EB} (ΔH_{EB}) and modulation ratio of $H_{EB}[\Delta H_{EB}/H_{EB}(0V)]$, respectively.

strong T dependence [see Fig. 2(e)]. As a result, the modulation ratio of $H_{EB}[\Delta H_{EB}/H_{EB}(0\,V)]$ increases with T, as shown in Fig. 3(i).

Next, we discuss the origin of the EF-induced H_{EB} modulation observed here. The important point is that the modulation of H_{EB} is confirmed even at low values of T (the lowest T in the present experiment was 40 K). If the modulation is induced by a thermally activated phenomena, e.g., a redox reaction [22,23], the modulation is expected to exponentially increase with T. ΔH_{EB}, however, is nearly constant at low T [see Fig. 3(d)]. In addition, the V_G modulation of R_{sheet} by $V_G = \pm 10$ V for the present sample is at most 0.04% [26,34]. Moreover, it has no exponential relationship with T in the range from 40 to 300 K, as shown in Fig. 4(a). Note that the R_{sheet} modulation induced by the redox reaction is on the order of 10% at 300 K [26], which is much larger than the present case. Our previous study has shown that the voltage induced redox does not occur in the same system when the HfO_2 insulator layer is formed at 150 °C [26]. The results discussed above strongly suggest that thermally activated phenomena are not the case with the present result. The modulation of t by the EF might be the origin of the present EB change. However, the t change is calculated to be only 1.5 pm, even when the R_{sheet} change is assumed to be totally caused by the t change. It is quite small; thus, to explain the present results only by the t change is difficult. In addition, the modulation of the ME effect in the AFM layer [8–12] can be ruled out because CoO_x is not a multiferroic material [10]. Therefore the modulation of the electronic state at the Co/CoO_x interface, including the charge doping, becomes the most reasonable explanation for the EF-induced H_{EB} change observed here.

Within the MB model, the modulation of K_J and/or tM_s results in the H_{EB} modulation. As shown in Fig. 4(b), the modulation ratio of tM_s is approximately 0.8% per 10 V at 250 K, the value of which is consistent with a previous report [36]. In addition, the modulation of tM_s is almost T-independent, which is confirmed from the T dependence of

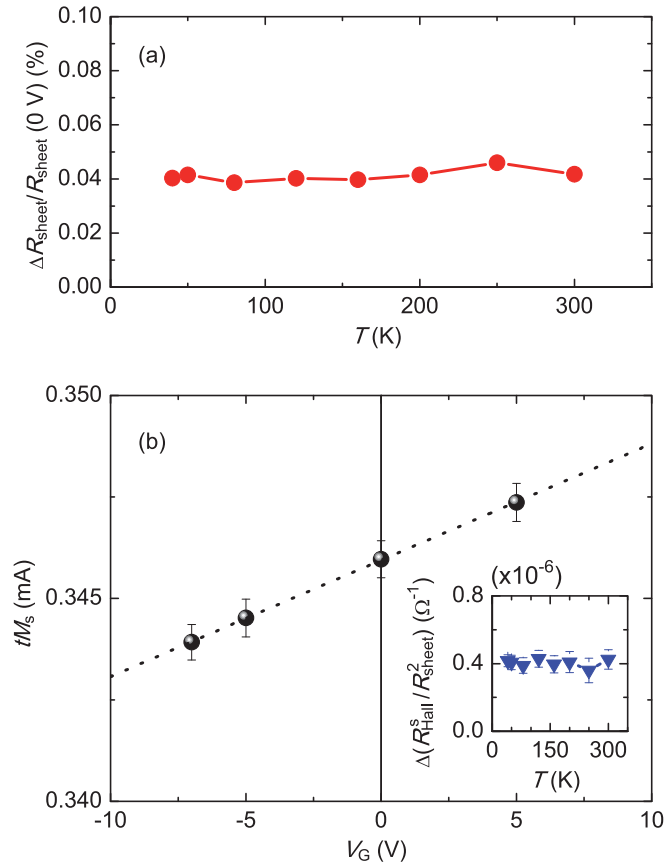

FIG. 4. (a) T dependence of R_{sheet} modulation $\Delta R_{sheet}/R_{sheet}$ (0 V), where ΔR_{sheet} is the difference in R_{sheet} between $V_G = +10$ and -10 V. (b) V_G dependence of tM_s at 250 K. The inset shows the difference in $R^s{}_{Hall}/R^2{}_{sheet}$ between $V_G = +10$ and -10 V [$\Delta R^s{}_{Hall}/R^2{}_{sheet}(0\,V)$, where $R^s{}_{Hall}$ is R_{Hall} at which the magnetization is saturated] as a function of T. $\Delta R^s{}_{Hall}/R^2{}_{sheet}$ is almost constant with T, suggesting that tM_s is independent of T under the assumption that the magnetization is proportional to $R^s{}_{Hall}/R^2{}_{sheet}$ [35].

the R_{Hall} modulation [see the inset of Fig. 4(b)]. tM_s, however, increases (decreases) with positive (negative) V_G. Thus the sign of the H_{EB} change (positive V_G increases H_{EB}) cannot be explained by the tM_s change, i.e., not the modulation of tM_s, but rather of K_J, plays a crucial role in the EF effect on H_{EB}. If the variation of tM_s by ± 10 V is $\pm 1\%$, the EF-induced K_J modulations ΔK_J by ± 10 V are calculated to be 8.8 and 4.3 μJ/m^2 at 50 and 160 K, respectively. Note that K_J modulation is expected to produce changes even in T_N and T_B [37], which may be relevant to the rapid increase in the modulation ratio of H_{EB} toward T_B [see Fig. 3(e)], as in the case of EF-induced MA and the remnant magnetic moment near T_C [36,38].

One interpretation of the EF effect on K_J demonstrated here is as follows. K_J originates from the exchange interaction between the spins of atoms at the FM/AFM interface [1,2]. According to the theoretical study for the generalization of MB model by Mauri *et al.* and Malozemoff [39,40], K_J is determined by the following two equations:

$$K_J = A_{\text{A-F}}/\xi(\lambda < 1), \tag{1}$$

$$K_J = 2(K_A A_A)^{1/2}(\lambda > 1), \tag{2}$$

where K_A, A_A, $A_{\text{A-F}}$, and ξ are the crystalline anisotropy of the AFM, the exchange stiffness of AFM layer and AFM/FM interface, and the distance between interfacial AFM and FM atoms, respectively. Furthermore, λ is defined as $A_{\text{A-F}}/2\xi(K_A A_A)^{1/2}$. In this model, the interfacial anisotropy of FM does not involve K_J. Using $K_A = 2.92 \times 10^7$ J/m^3 of CoO [41], the strength of exchange interaction of CoO-CoO (-2 meV) and CoO-Co (-2 meV) from Ref. [42], and the lattice constant of CoO $a_{\text{CoO}} = 0.4267$ nm $\sim \xi$, λ is calculated to be 0.2; therefore, Eq. (1) is the appropriate expression in our system. Thus the EF modification of $A_{\text{A-F}}$ is expected to cause the change in K_J. This is one of the possible scenarios for the EB modulation. However, the EF induced re-arrangement of the electron structure may produce a change in other parameters, i.e., K_A, A_A, and ξ, which can also be attributed to the EB modulation. Further investigation of magnetic and structural parameters, including stoichiometry of CoO, is required to clarify which parameter change dominates the EB modulation.

Although the EF effect on $A_{\text{A-F}}$ has not been reported, it is possible that the EF modulation of the orbital hybridization [19], which may include the orbital occupancy [17,18], at the Co/CoO$_x$ interface is its microscopic mechanism. This is analogous to the case of the EF effect on the exchange interaction of ferromagnetism [36,43–45]. Another possibility is that the EF-induced modulation of the interfacial Dzyaloshinskii–Moriya interaction (iDMI), which has been intensively studied in layered structures with a structural inversion asymmetry [46–50]. Theoretical study has suggested that the iDMI between magnetic atoms in FM and AFM layers acts on the FM atom as an effective magnetic field, which is considered to contribute to EB [51,52]. In our case, the iDMI at the Co/CoO$_x$ interface might be modulated by the EF, resulting in the EB modulation.

Using the EF control of EB, magnetization switching is expected to be possible. However, further enhancement of EB modulation magnitude is required. Recently, it has been suggested that the efficiency of MA modulation by the EF strongly depends on the internal strain in the FM layer [53–57]. This likely originates from the strain-dependent electron structure of the FM at the Fermi surface. Thus, by tuning the amount of strain in the FM layer, enhancing the EB modulation might be possible. In addition, with this method, high speed operation is expected to be possible, which is advantageous over other methods, such as redox effect.

IV. SUMMARY

In summary, we investigated the EF-induced modulation of EB in a Co/CoO$_x$ structure in which the antiferromagnetic CoO$_x$ layer is formed by natural oxidization. Clear EF-induced modulation of the EB field was observed below T_B. The efficiency of changes in the EB field increased near T_B. The most reasonable mechanism of the EB change demonstrated here is the modulation of the interfacial electronic structure, including electron doping at the Co/CoO$_x$, in contrast to the thermally activated chemical reaction of FM or AFM layer. This mechanism is fundamentally different from the conventionally studied multiferroic AFM case. Although further expansion of the window of the EF-induced change in the EB is required, this method might open a new route in order to develop fast and efficient magnetization switching.

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