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Magnetoelectric switching of perpendicular exchange bias in Pt/Co/α-Cr_2O_3/Pt stacked films

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We report the realization of magnetoelectric switching of the perpendicular exchange bias in Pt/Co/α-Cr_2O_3/Pt stacked films. The perpendicular exchange bias was switched isothermally by the simultaneous application of magnetic and electric fields. The threshold electric field required to switch the perpendicular exchange bias was found to be inversely proportional to the magnetic field, which confirmed the magnetoelectric mechanism of the process. The observed temperature dependence of the threshold electric field suggested that the energy barrier of the antiferromagnetic spin reversal was significantly lower than that assuming the coherent rotation. Pulse voltage measurements indicated that the antiferromagnetic domain propagation dominates the switching process. These results suggest an analogy of the electric-field-induced magnetization with a simple ferromagnet. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4918940]

The manipulation of magnetization is a fundamental concept on which devices for spintronics applications are based. A straightforward technique to achieve this is the implementation of the current-induced Oersted field, which is used in high-density storage devices such as hard disk drives. However, the high energy consumption of the method can be problematic, particularly in micro/nano devices. The spin-polarized charge current has attracted attention as an alternative way to manipulate magnetization because it can directly interact with ferromagnetic (FM) spins via a spin-transfer torque.[1,2] This technique requires high current densities, above 10^6 A/cm^2,[3,4] which might generate undesirable heat dissipation in the devices. The electric-field control of magnetization is another candidate, which does not involve this problem. In this method, an electric field applied across a magnetoelectric (ME) insulator induces magnetization switching; several materials, such as $TbMnO_2$ (Ref. 5) and $BaFeO_3$,[6] have been proposed as ME insulators. α-Cr_2O_3 is one of the proposed ME insulators exhibiting antiferromagnetic (AFM) features;[7,8] because of these, an exchange bias is induced by coupling with the FM layer. In our previous papers,[9,10] we reported that a perpendicularly directed exchange bias above 0.4 erg/cm^2 could be induced in Pt/Co/α-Cr_2O_3/Pt stacked films. This high perpendicular exchange bias is related to the ME-controllable boundary magnetization.[11–14] For bulk α-Cr_2O_3 (Refs. 13 and 15) and α-Cr_2O_3 thin films,[14,16] the electrical switching of the exchange bias has been achieved by the simultaneous application of magnetic and electric fields. Two switching modes have been proposed for this process: ME-field cooling[14–16] and isothermal switching.[13] While the former mode was reported for both bulk α-Cr_2O_3 (Ref. 15) and α-Cr_2O_3 thin films,[14,16] the latter mode using the α-Cr_2O_3 thin film is still challenging. In this study, the isothermal switching of the exchange bias using α-Cr_2O_3 thin films is demonstrated. We also address and discuss the character of the switching mechanism using the results of pulse voltage measurements.

Pt(5)/Co(0.8)/α-Cr_2O_3(200)/Pt(20) stacked films were used, where the numbers in the parentheses denote the thickness of each layer in nanometers. The films were prepared on an α-Al_2O_3(0001) substrate by DC magnetron sputtering using a base pressure of the sputtering chamber below 10^{-6} Pa. The Pt buffer layer was deposited on the substrate at room temperature and was then annealed for 1.8 ks at 673 K to smoothen the surface. Subsequently, the α-Cr_2O_3 layer was deposited at 773 K using an Ar and O_2 gas mixture, i.e., reactive sputtering, whereas the Co and the Pt capping layers were deposited at room temperature. The sputtering gas pressure was kept at 0.5 Pa for the Pt and α-Cr_2O_3 layers and at 0.6 Pa for the Co layer. These procedures generated a crystalline orientation normal to the film of Pt(111), α-Cr_2O_3(0001), and Co(111), which was confirmed by reflection high-energy electron diffraction.[10,17] No other crystallographic orientations were detected in the out-of-plane X-ray diffraction profile. The X-ray reflectivity measurements revealed that the Co/α-Cr_2O_3 interface roughness was approximately 0.3 nm, indicating that a sharp FM/AFM interface was realized.

It is sometimes argued that when a Co layer is directly deposited on α-Cr_2O_3, as in our films, Co could be oxidized and a spacer layer, such as Pt, is required to avoid oxidization.[13,15] The chemical and magnetic states of Co were characterized using soft X-ray absorption (XAS) and X-ray magnetic circular dichroism (XMCD), respectively. The XAS and XMCD measurements were performed at the BL25SU beamline of the SPring-8 synchrotron radiation facility, which is equipped with twin helical undulators.[18] The XMCD signals were collected by the difference of the XAS for the positive (μ_+) and the negative

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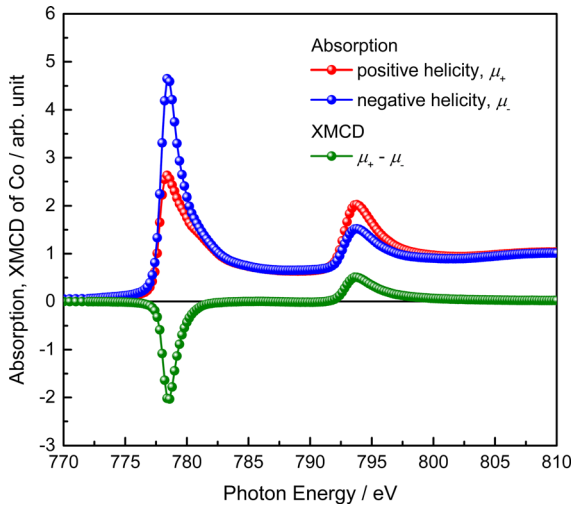

FIG. 1. XAS and XMCD spectra at the Co $L_{2,3}$ edges measured at room temperature. The red and blue symbols represent XAS for the positive and the negative helicities, respectively, and the green circles represent the XMCD spectrum. A magnetic field of 19 kOe was applied perpendicular to the film plane.

(μ_-) helicities of the incident X-rays. Figure 1 shows the XAS and the XMCD spectra at the Co $L_{2,3}$ edges measured at room temperature. The absorption spectra were collected at a magnetic field of 19 kOe. The XAS spectrum displays the metallic features of the films, and no multiplet-structure was observed. The sum rule analysis[19,20] gave the magnetic moments of Co as the effective spin moment $m_s^{\text{eff}} = m_s + 7\langle T_z\rangle = 1.67 \pm 0.13\,\mu_B$ and the orbital magnetic moment $m_l = 0.11 \pm 0.02\,\mu_B$, assuming the expectation value of the magnetic dipole operator $\langle T_z\rangle \ll 1$ and a hole number of 2.45.[21] The total magnetic moment was similar to that of bulk Co and, consequently, considering that the Co layer thickness was a few monolayers, Co oxidization was negligible in our film. It must be noted that the thicknesses of the Co and the Pt capping layers were specifically designed for the implementation of the surface-sensitive total electron yield method in the XAS and the XMCD measurements, details of which are presented in our previous paper.[22]

Prior to the investigation of the ME effect, we examined the magnetic easy direction using magnetization curves measured at room temperature. The magnetization curves for the in-plane and out-of-plane directions confirmed that the studied films exhibited perpendicular magnetic anisotropy.[23] The saturation magnetization M_S and the uniaxial magnetic anisotropy energy density K_U were 1447 emu/cc and 1.48×10^6 erg/cc, respectively. For the calculation of M_S and K_U, the spin polarization of the Pt layer was neglected; this assumption does not affect the estimation of the exchange anisotropy energy density J_K ($J_K = H_{EX} \cdot M_S \cdot t_{FM}$, where H_{EX}: the exchange bias field and t_{FM}: the FM layer thickness) when the spin-polarized Pt is coupled with Co. The switching of the perpendicular exchange bias was detected by the anomalous Hall effect (AHE), which was measured using a Hall-bar device fabricated photolithography and Ar ion milling. The device was designed so that the voltage could be applied across the α-Cr_2O_3 layer, whereas the Pt buffer layer acted as the bottom electrode (Fig. 2(a)). To obtain the target measurement temperature, which varied from 253 to 312 K, the sample was first heated to 312 K, above the Néel temperature of α-Cr_2O_3, and then cooled under a magnetic field of +6 kOe. Note that the Néel temperature of the fabricated α-Cr_2O_3 layer was about 285 K, which was estimated by the temperature dependence of H_{EX}. Before measuring the AHE curve, a magnetic field of −10 kOe and an electric field between 0 and −2500 kV/cm were simultaneously applied. The AHE loops were acquired as a function of the perpendicular magnetic field after switching the electric field off. The directions of the magnetic and electric fields were perpendicular to the film and the positive direction for both fields was defined from the top to the bottom of the device (Fig. 2(a)). Using this setup, the leakage current and the resistivity

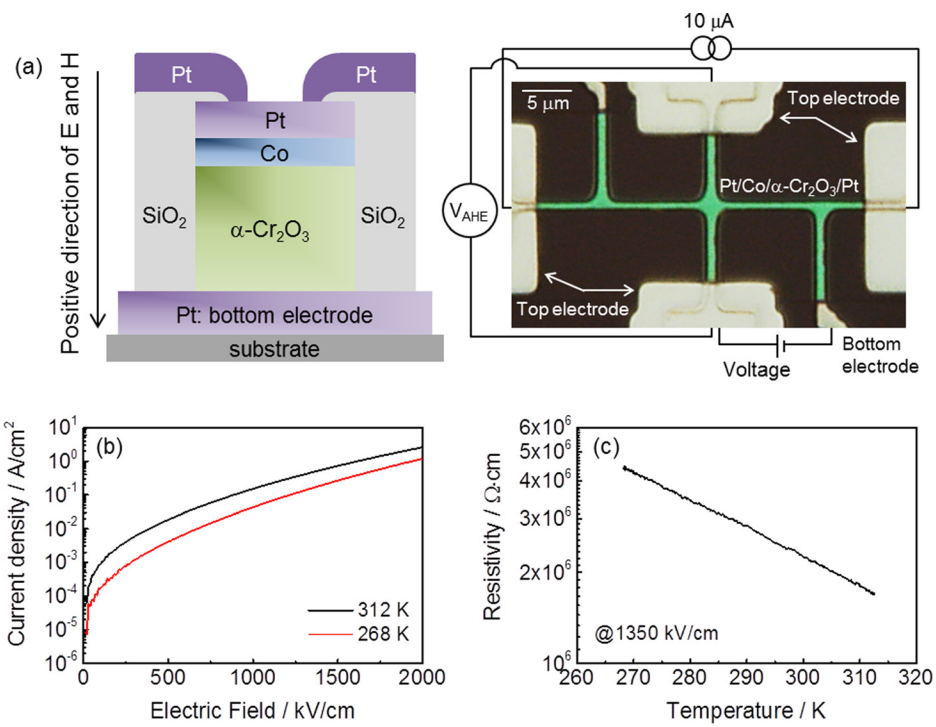

FIG. 2. (a) Schematic drawing of the cross-sectional structure and experimental setup using the Hall device. The photograph was taken by an optical microscope for the same structure used for the investigation. (b) I-V curves measured at 312 K (black) and 268 K (red). (c) Temperature dependence of resistivity measured at $E = 1350$ kV/cm.

across the α-Cr_2O_3 layer were also measured by the two-terminal method. As shown in Fig. 2(b), at 312 K, the leakage current was 0.15 A/cm^2 (2.6 A/cm^2) at 1000 kV/cm (2000 kV/cm) and these values decreased with decreasing temperature. The resistivity was sufficiently high to ensure the insulating feature, above 3.6×10^6 Ω·cm, in the temperature regime applied for the ME investigation, i.e., <278 K (Fig. 2(c)).

Figure 3(a) shows the AHE loops measured at 253 K. After the magnetic field cooling (red curve), an exchange bias of −1.2 kOe was observed in the negative magnetic field direction. As shown by the blue curve, by applying a magnetic field of −10 kOe and an electric field of −2000 kV/cm, simultaneously, the exchange bias was reversed and it was observed in the positive magnetic field direction. Since the temperature was constant during the field application, this switching occurred isothermally. Figure 3(b) displays the variation of the exchange bias field with the applied electric field under a constant magnetic field of −10 kOe for all studied temperatures. As the electric field strength increased, the sign of the exchange bias field switched from negative to positive at the threshold electric field, E_{th}. At 278 K, the change of the exchange bias field was gradual, probably because the negative and the positive exchange biased states were thermally degenerated similarly to the Boltzmann distribution.

Before discussing the switching condition and the ME energy required to switching the exchange bias, we should address the issue of the switching direction discussed below. In principle, the isothermal switching of the exchange bias is reversible: the exchange bias can be reversed from positive to negative and vice versa only by the electric field, while maintaining the magnetic field constant. E_{th} is asymmetric for negative-to-positive and positive-to-negative switching.[13] This originates from the unidirectional nature of the exchange magnetic anisotropy and the difference of E_{th} values would be expected to increase with increasing J_K. In our experiment, switching was observed only for the negative-to-positive case, where J_K promotes AFM spin reversal. In contrast, in the positive-to-negative switching case, J_K hinders AFM spin reversal and the high electric field requires. In our film, the E_{th} value required for the positive-to-negative switching exceeded the breakdown voltage because of the higher J_K than that of the system using bulk α-Cr_2O_3.[13,15]

As an opening gambit, we assume the coherent rotation model to derive the total energy per unit area. Keeping in mind that the FM spin was fixed by the high magnetic field (−10 kOe) during the exchange bias switching, the Zeeman energy and the magnetic anisotropy energy of the FM layer became the constants. Taking into account that the Co and uncompensated Cr spins are antiferromagnetically coupled at the interface,[22] the magnetic energy per unit area can be expressed as

$$F = K_{AFM} t_{AFM} \sin^2\theta - \alpha_{33} E H t_{AFM} \cos\theta - J_K \cos\theta$$
$$= K_{AFM} t_{AFM} \sin^2\theta - \alpha_{33} E t_{AFM} \left(H + \frac{J_K}{\alpha_{33} E t_{AFM}} \right) \cos\theta, \quad (1)$$

where K_{AFM} and t_{AFM} are the magnetic anisotropy energy and the thickness of the AFM layer, respectively, α_{33} is the ME coefficient along the c axis, and θ is the angle between the interfacial uncompensated AFM spin and the magnetic field direction. The $\alpha_{33} E$ product in Eq. (1) is regarded as the E-induced magnetization as naturally derived from the definition of the ME effect; the $\alpha_{33} EH$ represents the energy gain by the ME effect.[7,8,24] Then, we can define the switching condition of the E-induced magnetization as

$$\alpha_{33} EH + \frac{J_K}{t_{AFM}} = 2K_{AFM}. \quad (2)$$

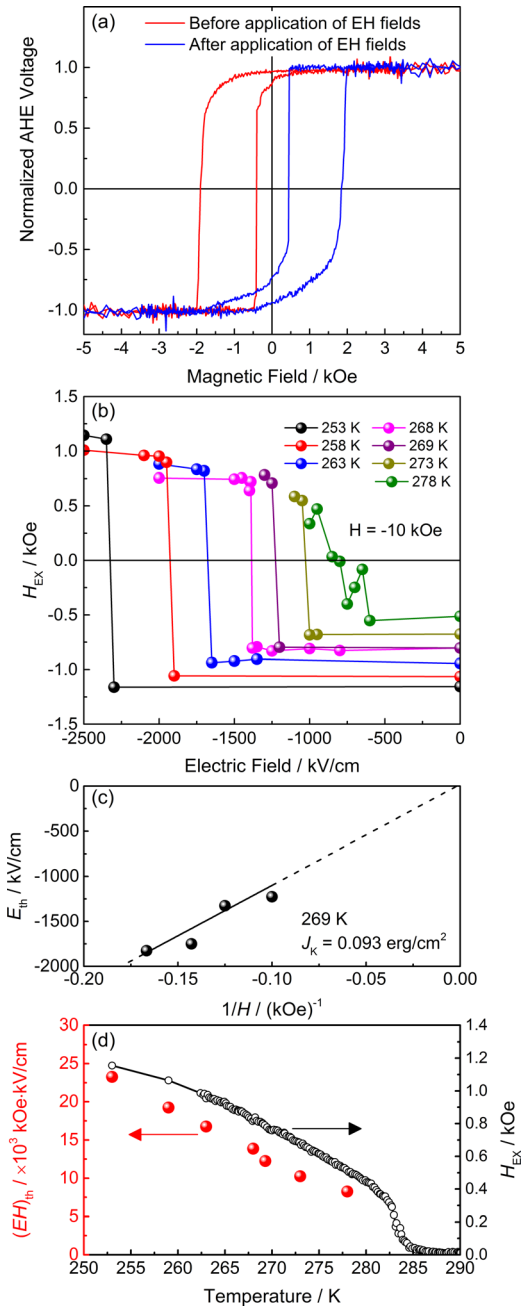

FIG. 3. (a) AHE loops before (red) and after (blue) the application of the magnetic (−10 kOe) and the electric (−2500 kV/cm) fields, measured at 253 K. (b) Change in H_{EX} with the electric field under the magnetic field of −10 kOe, measured at various temperatures. (c) Change in E_{th} with the magnetic field strength, measured at 269 K. The lines in the figure are guides for the eye. (d) Temperature dependence of the EH product required to switch the perpendicular exchange bias, estimated for a magnetic field of −10 kOe. The right vertical axis shows the temperature dependence of H_{EX}.

According to Eq. (2), the threshold EH product required to switch the exchange bias $(EH)_{th}$ should be constant as long as J_K and K_{AFM} are fixed by maintaining the temperature constant. In fact, as shown in Fig. 3(c), E_{th} is roughly proportional to $1/H$ agreeing with the above discussion.

Considering that K_{AFM}, α_{33}, and H_{EX} depend on the temperature, Eq. (2) indicates that $(EH)_{th}$ changes with temperature. Figure 4(d) shows the temperature dependence of $(EH)_{th}$. In the figure, H_{EX} is also shown. Since M_S is almost constant in the measurement temperature regime, the temperature dependence of H_{EX} provides that of J_K. According to the above discussion, H_{EX} promotes the AFM spin reversal, and the increase of H_{EX} could reduce $(EH)_{th}$. However, $(EH)_{th}$ increases with decreasing temperature. In addition, the $(EH)_{th}$ value estimated assuming the K_{AFM}[25] and α_{33} (Ref. 26) values for the bulk α-Cr_2O_3 is of order of 10^7 higher than the experimental value. This indicates that the energy barrier of the AFM spin reversal is significantly lower than K_{AFM}, suggesting the reversed AFM domain nucleation triggers the switching.[24] More specifically, the right side of Eq. (2) should be replaced with the nucleation field of the reversed AFM domain, which is related to the coercivity. In a general ferromagnet, the coercivity increases with decreasing thickness and this tendency should be analogous for the E-induced magnetization. Thus, $(EH)_{th}$ for α-Cr_2O_3 thin film could be higher than that for bulk α-Cr_2O_3. In fact, the $(EH)_{th}$ value was, for example, about 10 000 kOe·kV/cm at 268 K; this value is over 300 times larger than the reported value for bulk α-Cr_2O_3.[13,15]

Finally, we investigated the switching process by using a pulse voltage with a pulse duration ranging from 10 ns to 1 ms. For this study, a pulse voltage of −35 V (−1750 kV/cm) was applied with a static magnetic field of −10 kOe and a measurement temperature of 268 K. Before applying each pulse, the magnetic state was initialized by field cooling from 312 K to 268 K under a magnetic field of +6 kOe. Because of the limited maximum pulse duration of the pulse generator (100 ns), long pulse durations above 100 ns were substituted by the combined application of pulse voltages with 50 ns or 100 ns duration. Figure 4(a) shows the change in the exchange bias field as a function of pulse duration. Below the pulse duration of 100 ns, the exchange bias field gradually increased and at approximately 100 ns, the exchange bias polarity switched from negative to positive. For long pulse durations, above about 1 μs, the exchange bias field became almost constant. In the intermediate state, the two loops with the negative and the positive exchange bias overlapped, indicating that the reversed and the unreversed regions coexisted. In this type of loop, the remanent ratio reflects the ratio of the two regions, i.e., the ratio of the reversed AFM domain. In Fig. 4(b), the change of the remanent ratio in the increasing branch of the magnetization curve with the pulse duration is shown. With increasing pulse duration, the remanent ratio gradually decreases and saturates above the duration time of about 1 μs, in agreement with the variation of H_{EX}. Martin and Anderson reported that the AFM domain switching of bulk α-Cr_2O_3 partially occurs above a pulse duration of the order of microseconds,[24] which roughly agrees with our findings. Consequently, the above results suggest that the AFM domain-wall propagation dominates the exchange-bias switching process. In principle, the AFM domain-wall velocity can be estimated by this type of experiment; the direct observation of the AFM domain is currently under investigation. Furthermore, the higher electric field may generate faster exchange-bias switching and investigations regarding this issue are also in progress.

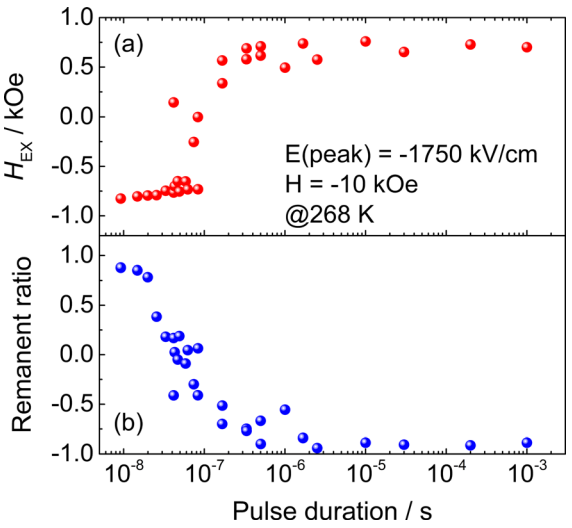

FIG. 4. Changes in (a) H_{EX} and (b) the remanent ratio in the increasing branch of the AHE loop with voltage pulse duration, measured at 268 K. The peak voltage (electric field) was set at −35 V (−1750 kV/cm) and the magnetic field was fixed at −10 kOe during the application of the pulse voltage.

In summary, we demonstrated the isothermal switching of the perpendicular exchange bias in Pt/Co/α-Cr_2O_3/Pt stacked films by the simultaneous application of magnetic and electric fields. The $(EH)_{th}$ value and the switching behavior triggered by the pulse voltage imply the analogy of the E-induced magnetization with a simple ferromagnetic magnetization; (1) the energy barrier for the AFM domain reversal is considerably lower than the energy barrier estimated assuming the coherent rotation and (2) the switching process of the exchange bias is dominated by AFM domain-wall propagation.

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