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Switching of perpendicular exchange bias in Pt/Co/Pt/α-Cr_2O_3/Pt layered structure using magneto-electric effect

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Switching of the perpendicular exchange bias polarity using a magneto-electric (ME) effect of α-Cr_2O_3 was investigated. From the change in the exchange bias field with the electric field during the ME field cooling, i.e., the simultaneous application of both magnetic and electric fields during the cooling, we determined the threshold electric field to switch the perpendicular exchange bias polarity. It was found that the threshold electric field was inversely proportional to the magnetic field indicating that the *EH* product was constant. The high *EH* product was required to switch the exchange bias for the film possessing the high exchange anisotropy energy density, which suggests that the energy gain by the ME effect has to overcome the interfacial exchange coupling energy to reverse the interfacial antiferromagnetic spin. *© 2015 AIP Publishing LLC.*
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Controlling magnetization direction is a key technique of magnetic and spintronics devices. A straight way is to use a magnetic field, which is utilized in high density storage devices like the hard disk drive. Although the magnetic field is still useful, its high energy consumption is often a problem, in particular, in the micro/nano magnetic devices. As alternate ways, the current-induced[1–3] and the voltage-induced[4] magnetization switching have been investigated toward the application in the spintronics devices such as magnetic random access memory. These techniques are promising to ensure the low power consumption and to improve the CMOS compatibility of the spintronics devices. In this work, as one of the voltage-induced magnetization techniques, we investigated the magneto-electric (ME) switching of the exchange bias using the ME effect of an antiferromagnetic (AFM) α-Cr_2O_3.[5,6]

The exchange bias is generated by an interfacial magnetic anisotropy at the ferromagnetic (FM)/AFM interface[7] and it manifests as the shift of a magnetization curve along the magnetic-field axis. The ME-controlled exchange bias was first reported in 2005 by Borisov *et al*. as the switching of the exchange bias polarity after the ME-field cooling (MEFC).[8] After that, the isothermal switching[9] and the MEFC effect in the thin film system[10,11] have been reported up to date. The efforts using an all-thin-film system have been started very recently[10,11] and we have to accumulate the experimental and theoretical data to mature this technique and to clarify the physics. In this work, we focus on the threshold condition to switch the exchange bias polarity by the MEFC using the Pt/Co/Pt/α-Cr_2O_3/Pt layered structure. In particular, using the films possessing the different exchange anisotropy energy density J_K, we investigated the change in the threshold energy gain by the ME effect to switch the exchange bias polarity with J_K. We also discuss the switching mechanism by comparing our results with the previous reports.

The samples were fabricated by a DC magnetron sputtering system operating at a base pressure of 10^{-7} Pa. The stacking structure of the samples was Pt(5.0 nm)/Co(0.8 nm)/Pt(0.5 nm)/α-Cr_2O_3(150 nm)/Pt(20 nm)/α-Al_2O_3-substrate. In order to alter J_K of the film, we adopted two types of the samples; one is the film with the textured α-Cr_2O_3 layer and the other is the film with the twinned α-Cr_2O_3 layer. The crystalline quality of the α-Cr_2O_3 layer is controlled by the growth condition of the Pt buffer layer. For the former type of sample, the Pt buffer layer was deposited at room temperature and then annealed for 1.8 ks at 673 K, giving rise to the textured Pt(111) layer. For the latter one, the Pt buffer layer was deposited above 773 K giving rise to the twinned Pt(111) layer. On these Pt buffer layer, the α-Cr_2O_3 layer was deposited at 773 K by means of the reactive sputtering method using the Ar and O_2 gas mixture. On the α-Cr_2O_3 layer, the Pt spacer layer, the Co layer, and the Pt capping layer were deposited at room temperature. The details of the preparation method are described in the previous papers.[12,13] Independent of the growth condition of the Pt buffer layer, the crystallographic orientation normal to the film surface was Pt(111), Co(111), and α-Cr_2O_3(0001), as confirmed by *in situ* reflection high-energy electron diffraction. Magnetic characterizations were carried out by means of vibrating sample magnetometer (VSM) and anomalous Hall effect (AHE). The magnetization curves for in-plane and out-of-plane directions measured using VSM confirmed that all studied films exhibited perpendicular magnetic anisotropy.[14] Thus, the exchange bias investigated in this paper is a perpendicular exchange bias.[12] The ME effect was investigated using the AHE measurements and it was characterized by

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the perpendicular exchange bias after the MEFC in which both magnetic field H and electric field E are applied during cooling.[8] The AHE measurements were carried out at 260 K. To access the measurement temperature, the sample was first heated to 317 K, above the Néel temperature of bulk α-Cr_2O_3, and then cooled under the ME field conditions. In this work, the magnetic and electric fields were applied in the direction perpendicular to the film. The AHE measurements were carried out at $E = 0$ kV/cm. For the AHE measurements, the samples were patterned into the microdots of 20 μm diameter by means of photolithography and Ar ion milling. Using the microdots, the leakage current and the resistivity were also characterized by two probe method.

Prior to show the results on the ME investigation, we show the leakage current and the resistivity of the fabricated microdot, which are essential information to ensure the ME effect shown below. Figure 1(a) shows the leakage current as a function of the electric field measured at 317 K. The sample was avoided from the electrical break down at the high electric field of 1200 kV/cm. The leakage current was $\sim 1 \times 10^{-4}$ A/cm^2 at 80 kV/cm and 4.5×10^{-2} A/cm^2 at 1200 kV/cm. These values are comparable to the reported values measured at room temperature for the 1-μm-thick α-Cr_2O_3 layer prepared by a pulsed laser deposition[15] but higher than that of the 250-nm-thick α-Cr_2O_3 layer prepared by an RF magnetron sputtering.[10] Besides, as shown in Fig. 1(b), the electrical resistivity increases with decreasing temperature. At 260 K, the resistivity reaches above 73 MΩ·cm at 1200 kV/cm. These results ensure the sufficient insulating feature of the microdot to investigate the ME effect. It should be noted that as shown in the inset of Fig. 1(a), there is insulator surrounding the microdot to avoid the direct contact between the top and bottom electrodes and the leakage current can be generated in this insulator. The leakage current shown in Fig. 1(a) was calculated assuming that the leakage current was concentrated in the microdot, and thus the leakage current and the resistivity shown are maximum and minimum values, respectively.

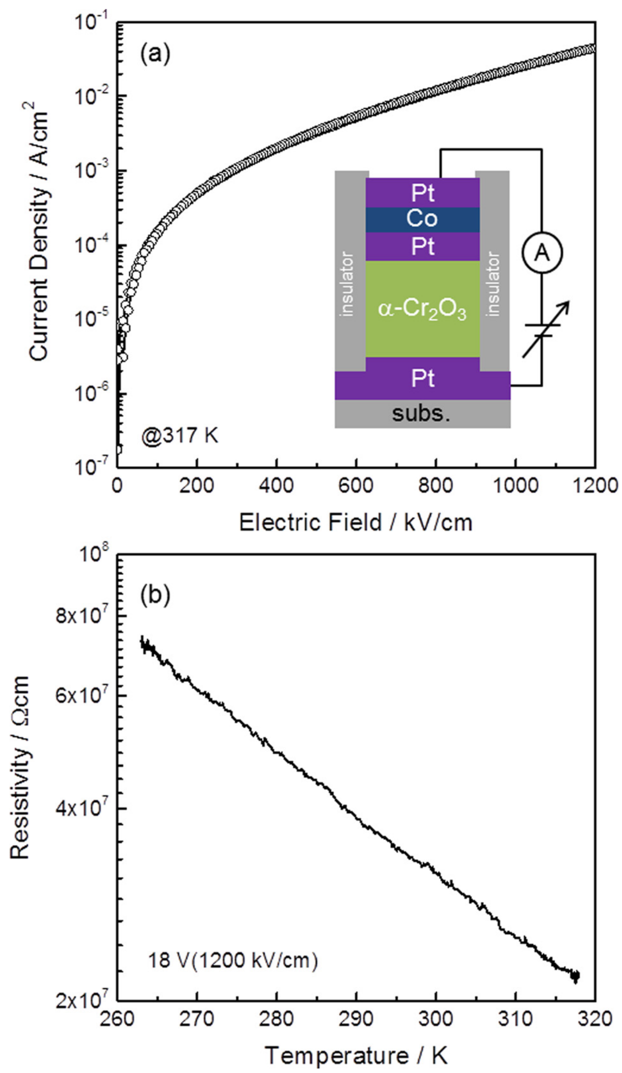

FIG. 1. (a) I-V curve measured at 317 K and (b) temperature dependence of resistivity of the film with the twinned α-Cr_2O_3 layer. Inset of (a) represents a schematic cross-sectional view of the microdot.

Figure 2 shows the normalized AHE loops measured after the MEFC. When the positive magnetic field (+9 kOe) and the negative electric field (−1000 kV/cm) were applied during the cooling, the exchange bias of 0.75 kOe was observed in the negative direction (blue curve). This curve is almost same as that measured after the magnetic field (+9 kOe) cooling without the electric field. When the electric field during the MEFC was changed to +1200 kV/cm in fixing the magnetic field to +9 kOe, the exchange bias 0.66 kOe was observed in the positive direction (red curve). Since the magnetic field applied during the MEFC is identical for two curves, the observed switching of the exchange bias was induced by the ME effect of α-Cr_2O_3. It has been argued that from the symmetry arguments, the switching of the exchange bias polarity is coupled with the surface magnetization of the α-Cr_2O_3.[16,17] By the ME effect, the surface magnetization is induced depending on the sign of the product of EH. As shown in Fig. 2, when the sign of the EH product is positive, the positive exchange bias was observed. Considering that the exchange coupling with interfacial uncompensated Cr spin and Co spin is antiferromagnetic,[18] the positive surface magnetization generates the positive exchange bias in agreement with the above discussion. One

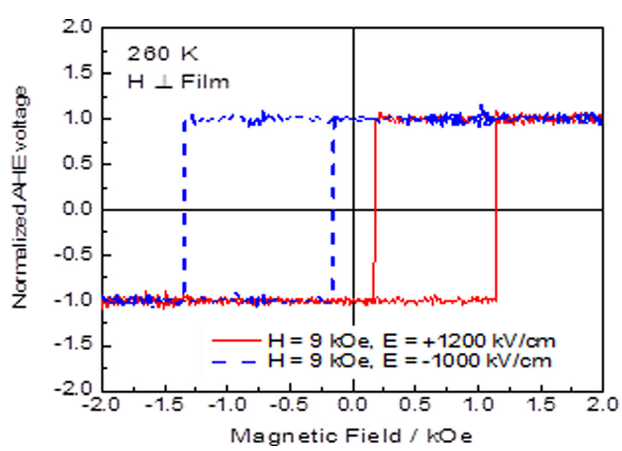

FIG. 2. Normalized AHE loops after the MEFC for the film with the twinned α-Cr_2O_3 layer. Broken blue and solid red lines represent the data after the MEFC of $H = +9$ kOe, $E = -1000$ kV/cm and $H = +9$ kOe, $E = +1200$ kV/cm, respectively.

may still wonder that the switching of the exchange bias was triggered by the leakage current in the α-Cr_2O_3 layer. However, as shown in Fig. 1, the leakage current is at most 4×10^{-2} A/cm^2 which is insufficient to excite the spin flop phase of α-Cr_2O_3 by the current-induced Oersted field, e.g., about 0.28 Oe at 1 nm from the center of microdot assuming that the leakage current is concentrated at the center of the microdot. Besides, the maximum leakage current density is 11–13 orders smaller than the typical threshold current densities to move the magnetic domain wall of FM nanowire[19] and that to switch the magnetization direction by the spin transfer torque.[20] Furthermore, the unipolar switching of the exchange bias with respect to the electric field strength supports the ME mechanism. The exchange bias after switching (+0.66 kOe) is slightly lower than that before the switching (−0.75 kOe). This reduction of the exchange bias was observed in the bulk α-Cr_2O_3 and indicates that the AFM domain is not still fully reversed.

The previous report suggests that the higher electric field to switch the exchange bias polarity is required in the α-Cr_2O_3 thin film system compared to the bulk α-Cr_2O_3 system.[10] However, its critical condition, i.e., the threshold electric field to switch the exchange bias polarity was not investigated in detail. We determined the threshold electric field to switch the exchange bias polarity from the change in the exchange bias field with the electric field strength during the MEFC. In Fig. 3(a), the result for the film with the twinned α-Cr_2O_3 layer is shown. The exchange bias field was switched from about −0.75 kOe to about +0.66 kOe sharply at the threshold electric field. It is also seen that the threshold electric field decreases with increasing the magnetic field strength during the MEFC. The threshold electric field strength was defined as the electric field at which the exchange bias field becomes zero and it was shown in Fig. 3(b) as a function of the magnetic field strength. The threshold electric field is inversely proportional to the magnetic field strength in agreement of the ME mechanism. The *EH* products at the threshold condition $(EH)_{\text{threshold}}$ are 6714 kV/cm·kOe for the film with the twinned α-Cr_2O_3 layer.

According to our previous reports,[18,21] the switching of the exchange bias should be accompanied with the reversal of the interfacial uncompensated Cr spin and thus the energy gain to reverse the interfacial AFM spin would be related to the interfacial exchange coupling strength. We compare the *EH* products for the film possessing the different J_K [ergs/cm^2] $= H_{EX} \cdot M_S \cdot t_{FM}$, where H_{EX} is the maximum exchange bias field [Oe], M_S is the saturation magnetization [emu/cc], and t_{FM} is the ferromagnetic layer thickness [cm]. Using this definition of J_K, the J_K for the film with the twinned α-Cr_2O_3 layer is 0.072 ergs/cm^2 as shown in Fig. 3(b). In Fig. 3(b), the threshold electric field for the film with the textured α-Cr_2O_3 layer possessing $J_K = 0.039$ ergs/cm^2 is also plotted. For the latter film, the $(EH)_{\text{threshold}}$ is 4212 kV/cm·kOe, lower than the film with the twinned α-Cr_2O_3 layer possessing high J_K. Note that the change in J_K with the crystalline quality of the α-Cr_2O_3 may be related to the structural difference such as the grain size but it is unclear at present.

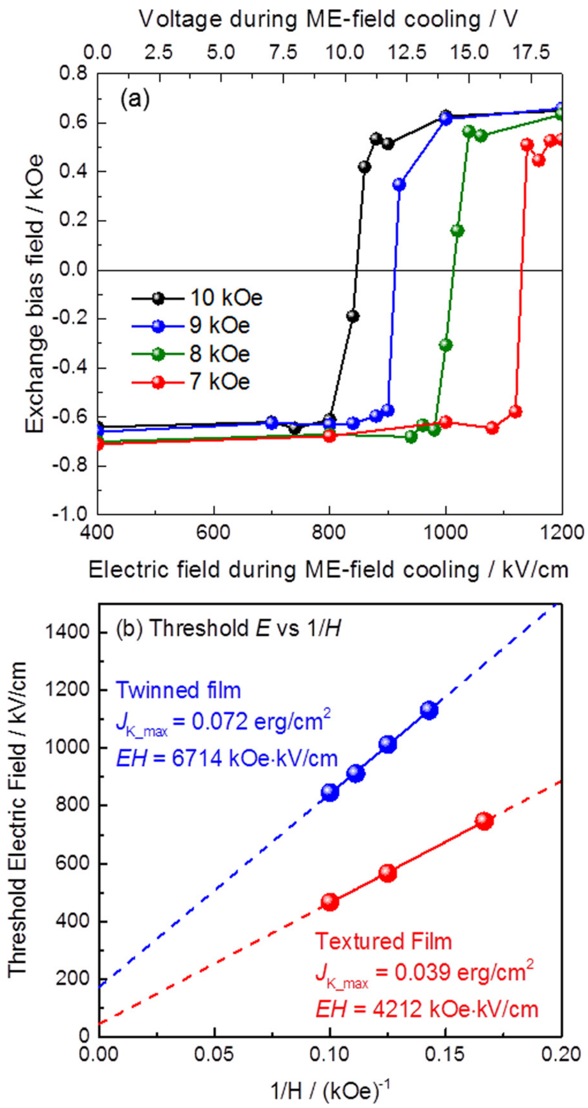

FIG. 3. (a) Changes in perpendicular exchange bias field with the electric field during the MEFC for the film with the twinned α-Cr_2O_3 layer. The magnetic field applied during the MEFC was changed as +10 kOe (black), +9 kOe (blue), +8 kOe (green), and +7 kOe (red). (b) Change in the threshold electric field to switch the exchange bias polarity with the magnetic field strength during MEFC. Red and blue dots represent the value for the films with the textured and twinned α-Cr_2O_3 layer, respectively. The horizontal axis is represented by the inverse of magnetic field 1/*H*. The solid and dotted lines are guide to eye. J_K value shown in (b) was calculated using the exchange bias field at 260 K after the magnetic-field cooling condition with $H = +9$ kOe and $E = 0$ kV/cm.

Because this topic is out of the scope of this paper, it will be reported elsewhere.

We discuss the change in the $(EH)_{\text{threshold}}$ with J_K from the viewpoints of the interfacial AFM spin reversal[16,17,22] by the ME α-Cr_2O_3. In our experiment, since the magnetic field during the MEFC was fixed at +10 kOe, enough high to saturate the FM magnetization, the FM spin orientation is fixed during the switching of the exchange bias polarity. In this situation, in order for the interfacial AFM spins to reverse, they have to overcome the interfacial exchange coupling J_{int} with the FM spin. In addition, the AFM spins also overcome the potential barrier by the magnetic anisotropy of α-Cr_2O_3 layer K_{AFM}, in particular, at the reversed AFM domain nucleation.

Thus, the energy to reverse the interfacial AFM spins gained by the ME effect, i.e., the *EH* product, should be related to J_{int} and K_{AFM}. On the one hand, it was reported that the perpendicular exchange bias in Pt/Co/α-Cr_2O_3/Pt layered system is caused by the magnetic domain formation in the AFM layer,[18] which suggests that the magnetic domain model for the exchange bias[23] may be applicable. The magnetic domain wall model predicts that J_K is proportional to $2\sqrt{A_{AFM}K_{AFM}}$ and J_{int} for the strong and weak interfacial exchange coupling cases, respectively.[23] Hence, the *EH* product to switch the exchange bias polarity can change with J_K through J_{int} and K_{AFM}.

Here, we compare the $(EH)_{threshold}$ values with the previous reports. The $(EH)_{threshold}$ values obtained in this work are about 130–180 times and 10–14 times larger than those for the $[Pt/Co]_3$/Pt/α-Cr_2O_3-substrate system[8,9] and the Pt/Co/Pt/α-Cr_2O_3 thin film system,[10] respectively, but it is only 3–5 times larger than the $(EH)_{threshold}$ to reverse the AFM domain of bulk α-Cr_2O_3.[24] It is likely that the systems with the α-Cr_2O_3 thin film require the high *EH* product. In addition to the above mentioned influence of J_K on $(EH)_{threshold}$, the low ME coefficient α_{ij} and/or the high magnetic anisotropy in the α-Cr_2O_3 thin film can be considered. Because both α_{ij} and magnetic anisotropy constant of α-Cr_2O_3 can be altered by the lattice distortion and the displacement of Cr^{3+} and/or O^{2-} ions in a crystal lattice,[25] the crystallographic analysis of the α-Cr_2O_3 thin film is necessary to clarify the details. This is now under the investigation using the synchrotron X-ray diffraction technique and will be reported in near future.

Finally, we briefly discuss the influence of the Pt spacer layer on the switching of the exchange bias polarity. As a preliminary result, we confirmed that the switching of the exchange bias polarity itself occurs for the film without the Pt spacer layer, while this type of film requires the higher *EH* product than the film with the Pt spacer layer.[26] This is possibly because the Pt spacer layer decreases J_K[27] and this tendency agrees with the above results and discussions. The details of the effect of the Pt spacer layer are now under the investigation using the X-ray circular dichroism and will be reported in the near future.

In summary, we investigated the switching of the perpendicular exchange bias polarity by the MEFC in the Pt/Co/Pt/α-Cr_2O_3/Pt layered structure. The exchange bias field switched at the threshold electric field is inversely proportional to the magnetic field. The required energy gain to switch the exchange bias polarity is high for the film with the high exchange anisotropy energy density. This suggests that the interfacial AFM spin reversal by the ME effect has to overcome the interfacial exchange coupling with FM spins.

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[1] L. Berger, Phys. Rev. B **54**, 9353 (1996).
[2] J. C. Slonczewski, J. Magn. Magn. Mater. **159**, L1 (1996).
[3] E. B. Myers, D. C. Ralph, J. A. Katine, R. N. Louie, and R. A. Buhrrman, Science **285**, 867 (1999).
[4] T. Maruyama, Y. Shiota, T. Nozaki, K. Ohta, N. Toda, M. Mizuguchi, A. A. Tulapurkar, T. Shinjo, M. Shiraishi, S. Mizukami, Y. Ando, and Y. Suzuki, Nat. Nanotechnol. **4**, 158 (2009).
[5] D. N. Astrov, Sov. Phys. JETP **10**, 628 (1960).
[6] V. J. Folen, G. T. Rado, and E. W. Stalder, Phys. Rev. Lett. **6**, 607 (1961).
[7] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956).
[8] P. Borisov, A. Hochstrat, X. Chen, W. Kleeman, and Ch. Binek, Phys. Rev. Lett. **94**, 117203 (2005).
[9] X. He, N. Wu, A. N. Caruso, E. Vescovo, K. D. Belashchenko, P. A. Dowben, and Ch. Binek, Nat. Mater. **9**, 579 (2010).
[10] T. Ashida, M. Oida, N. Shimoura, T. Nozaki, T. Shibata, and M. Sahashi, Appl. Phys. Lett. **104**, 152409 (2014).
[11] K. Toyoki, Y. Shiratsuchi, T. Nakamura, C. Mitsumata, S. Harimoto, Y. Takechi, T. Nishimura, H. Nomura, and R. Nakatani, Appl. Phys. Express **7**, 114201 (2014).
[12] Y. Shiratsuchi, T. Fujita, H. Oikawa, H. Noutomi, and R. Nakatani, Appl. Phys. Express **3**, 113001 (2010).
[13] Y. Shiratsuchi, Y. Takechi, K. Toyoki, Y. Nakano, S. Onoue, C. Mitsumata, and R. Nakatani, Appl. Phys. Express **6**, 123004 (2013).
[14] Y. Shiratsuchi, H. Oikawa, S. Kawahara, Y. Takechi, T. Fujita, and R. Nakatani, Appl. Phys. Express **5**, 043004 (2012).
[15] S.-H. Lim, M. Murakami, S. E. Lofland, A. J. Zambano, L. G. Salamanca-Riba, and I. Takeuchi, J. Magn. Magn. Mater. **321**, 1955 (2009).
[16] A. F. Andreev, JETP Lett. **63**, 758 (1996).
[17] K. D. Belashchenko, Phys. Rev. Lett. **105**, 147204 (2010).
[18] Y. Shiratsuchi, H. Noutomi, H. Oikawa, T. Nakamura, M. Suzuki, T. Fujita, K. Arakawa, Y. Takechi, H. Mori, T. Kinoshita, M. Yamamoto, and R. Nakatani, Phys. Rev. Lett. **109**, 077202 (2012).
[19] T. Koyama, K. Ueda, K.-J. Kim, Y. Yoshimura, D. Chiba, K. Yamada, J.-P. Jamet, A. Mougin, A. Thiaville, S. Mizukami, S. Fukami, N. Ishiwata, Y. Nakatani, H. Kohno, K. Kobayashi, and T. Ono, Nat. Nanotechnol. **7**, 635 (2012).
[20] S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H. D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, and H. Ohno, Nat. Mater. **9**, 721 (2010).
[21] Y. Shiratsuchi, T. Nakamura, K. Wakatsu, S. Maenou, H. Oikawa, Y. Narumi, K. Tazoe, C. Mitsumata, T. Kinoshita, H. Nojiri, and R. Nakatani, Appl. Phys. Lett. **100**, 262413 (2012).
[22] J. Martin and J. C. Anderson, IEEE Trans. Magn. **2**, 446 (1966).
[23] D. Mauri, H. C. Siegmann, P. S. Bagus, and E. Kay, J. Appl. Phys. **62**, 3047 (1987).
[24] A. Iyama and T. Kimura, Phys. Rev. B **87**, 180408(R) (2013).
[25] J. O. Artman, J. C. Murphy, and S. Foner, Phys. Rev. **138**, A912 (1965).
[26] Y. Shiratsuchi *et al.*, "Dynamical switching of perpendicular exchange bias in Pt/Co/α-Cr_2O_3/Pt layered system" (unpublished).
[27] Y. Shiratsuchi, T. Fujita, H. Noutomi, H. Oikawa, and R. Nakatani, IEEE Trans. Magn. **47**, 3909 (2011).