

Title	Competition of perpendicular magnetic anisotropy and exchange magnetic anisotropy in a Pt/Co/ $\alpha$ -Cr <sub>2</sub> O <sub>3</sub> (0001) thin film
Author(s)	Shiratsuchi, Yu; Noutomi, Hayato; Oikawa, Hiroto et al.
Citation	Journal of Applied Physics. 2011, 109(7), p. 07C101
Version Type	VoR
URL	<a href="https://hdl.handle.net/11094/89981">https://hdl.handle.net/11094/89981</a>
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Cite as: J. Appl. Phys. **109**, 07C101 (2011); <https://doi.org/10.1063/1.3535555>

Submitted: 21 September 2010 • Accepted: 21 October 2010 • Published Online: 17 March 2011

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## Competition of perpendicular magnetic anisotropy and exchange magnetic anisotropy in a Pt/Co/ $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>(0001) thin film

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(Presented 15 November 2010; received 21 September 2010; accepted 21 October 2010; published online 17 March 2011)

We investigated perpendicular magnetic anisotropy and exchange magnetic anisotropy in a Pt/Co/ $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>(0001) thin film grown on an  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>(0001) substrate. The film exhibits perpendicular magnetic anisotropy below a Co thickness of 1.2 nm at room temperature. Independent of the magnetic easy direction of the Co layer, the perpendicular exchange bias (PEB) appears in a direction perpendicular to the film below 80 K. The maximum unidirectional magnetic anisotropy energy estimated from the exchange bias field is 0.33 erg/cm<sup>2</sup>, which is higher than the reported PEB strength. The perpendicular exchange bias is accompanied by the in-plane remanent magnetization and an increase in the in-plane coercivity. We speculate that the increases in the in-plane remanent magnetization and the in-plane coercivity are caused by the spin canting of Cr<sup>3+</sup> in the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>(0001) layer. © 2011 American Institute of Physics. [doi:10.1063/1.3535555]

### I. INTRODUCTION

Exchange magnetic anisotropy is a type of interface magnetic anisotropy generated at an interface between a ferromagnetic (F) and an antiferromagnetic (AF) layer.<sup>1,2</sup> One promising feature of exchange magnetic anisotropy is the shift in the magnetization curve that occurs along the field axis, which is known as the exchange bias. The exchange bias is used to fix the magnetization of the F layer in spintronic devices such as spin valves. Currently, several spintronic devices make use of the in-plane oriented magnetization of the F layer, and thus the exchange bias lies in the film plane. However, spintronic devices that make use of perpendicular magnetization offer several advantages over the in-plane magnetization devices, including high integrability and low power consumption. In perpendicular magnetization devices, the exchange bias must be perpendicular to the film, that is, perpendicular exchange bias (PEB) must be achieved. PEB has recently been observed in systems such as [Pd/Co]<sub>n</sub>/IrMn,<sup>3</sup> [Pt/Co]<sub>n</sub>/CoO,<sup>4</sup> and [Pt/Co]<sub>n</sub>/ $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> substrates,<sup>5</sup> as well as Co/ $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layers.<sup>6–8</sup> Furthermore, a high PEB strength of 0.1 erg/cm<sup>2</sup> has been observed in systems using IrMn as an AF layer.<sup>3</sup> Generating high PEB strengths using an AF layer that does not contain rare species, such as Ir, is required if spintronic devices are to become widely available.

In previous studies, we reported that a PEB could be induced through the use of  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>(0001) as the AF layer.<sup>7,8</sup> Specifically, we reported that a PEB of approximately 0.29 erg/cm<sup>2</sup> was generated in a Pt/Co/ $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>(0001) thin film.<sup>8</sup> In the report, the magnetization behavior was investigated only for the magnetic field applied in the direction perpendicular to the film. Therefore, the influence of the magnetic easy direction of the Co layer on the high PEB was not clear. In the present study, we investigated the influence of the

magnetic easy direction of the Co layer on the PEB by varying the Co thickness from 0.5 to 2.4 nm.

Samples were fabricated using a direct current magnetron sputtering system. The base pressure of the magnetron sputtering system was less than 5×10<sup>-7</sup> Pa. The structure of the samples was as follows: Pt[5]/Co[*t*<sub>Co</sub>]/ $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>[30]/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub>(0001) substrate (the numbers in brackets represent the thickness of the film in nanometers). The  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer was deposited at 773 K on an  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>(0001) substrate that was smoothed by annealing in air.<sup>9</sup> A reactive sputtering method using a mixture of Ar + O<sub>2</sub> gases was adopted for the preparation of the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer. A Co layer and a Pt capping layer were deposited on the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer at room temperature. The Co thickness, *t*<sub>Co</sub>, was varied from 0.5 to 2.4 nm. Structural characterizations of the films were performed through reflection high-energy electron diffraction (RHEED), X-ray diffraction (XRD) and atomic force microscopy. The RHEED observations were performed *in situ*. The RHEED patterns showed that the prepared films exhibited the following orientation relationship in the growth direction:

$$\text{Pt}(111) \parallel \text{Co}(111) \parallel \alpha\text{-Cr}_2\text{O}_3(0001) \parallel \\ \alpha\text{-Al}_2\text{O}_3(0001) \text{ Substrate}$$

It is worth noting that the lattice constants of the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer determined through XRD were almost identical to those of bulk  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>. The surface roughness of the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer was 0.18 nm, calculated in a 1 × 1  $\mu$ m<sup>2</sup> region (root mean square value). Detailed structural information about the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer will be reported elsewhere. The magnetic properties of the film stack were investigated using a superconducting quantum interference device (SQUID) magnetometer and through vibrating sample magnetometry (VSM). The direction of the applied magnetic field was either parallel or perpendicular to the film plane. Measurements with the SQUID magnetometer were performed in the range of 5–140 K, and VSM

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measurements were performed at room temperature. When measurements were performed below room temperature, the sample was heated once to 330 K and then cooled to the measurement temperature in a magnetic field of +30.0 kOe, applied along the direction of measurement. The Néel temperature of  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> is reported to be approximately 307 K.<sup>10</sup>

First, we discuss the change in the magnetic easy direction with a change in the Co thickness. It is known that the effective magnetic anisotropy energy per unit volume,  $K_{\text{eff}}$ , can be expressed as follows:

$$K_{\text{eff}} = K_V - 2\pi M_S^2 + (K_S + K_{\text{Int}})/t_{\text{Co}}, \quad (1)$$

where  $K_V$  is the bulk term of the magnetic anisotropy,  $M_S$  is the saturation magnetization, and  $K_S$  and  $K_{\text{Int}}$  are the surface/interface terms of the magnetic anisotropy. In our investigations,  $K_{\text{eff}}$  was estimated from the magnetization curves for a field direction that was either parallel or perpendicular to the film.  $K_{\text{eff}}$  was estimated using the following relationship:

$$K_{\text{eff}} = \left( \int H dM \right)_{H \perp \text{Film}} - \left( \int H dM \right)_{H // \text{Film}}. \quad (2)$$

In this definition,  $K_{\text{eff}}$  represents the effective perpendicular magnetic anisotropy energy. The measurements were obtained at room temperature. The result is shown in Fig. 1. The sign of  $K_{\text{eff}}$  changed from negative to positive with decreasing Co thickness, and the transition occurred at approximately 1.2 nm. This trend indicates that samples with a Co thickness less than 1.2 nm exhibit perpendicular magnetic anisotropy. From the linear fit of the data for Co thicknesses between 1.2 and 2.4 nm, the values of  $K_V - 2\pi M_S^2$  and  $K_S + K_{\text{Int}}$  were determined as  $-1.1 \times 10^7$  erg/cm<sup>3</sup> and 0.66 erg/cm<sup>2</sup>, respectively. The obtained value of  $K_S + K_{\text{Int}}$  was in the range of the reported values, that is, 0.40–0.74 erg/cm<sup>2</sup> in the Pt/Co multilayer.<sup>11</sup> This result indicates that the perpendicular magnetic anisotropy at the Co/ $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> interface is of the same order of magnitude as the anisotropy at the Pt/Co interface. In the following paragraphs, we discuss the influence of the magnetic easy direction on the PEB of a film stack with a Co thickness of 1.0 or 1.6 nm and a magnetic easy direction that is either perpendicular or parallel to the film plane.

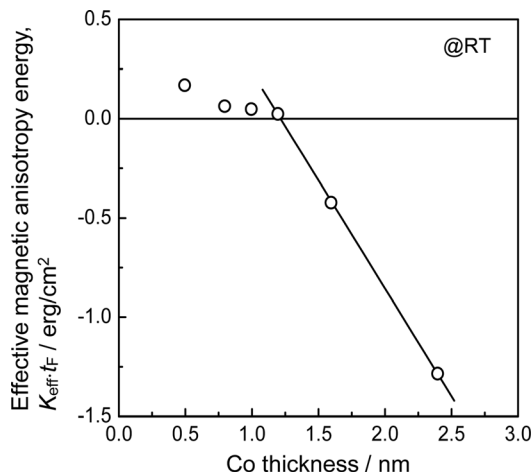


FIG. 1. The Co thickness dependence of the effective perpendicular magnetic anisotropy energy. The solid line represents the linear fit of data for Co thicknesses of 1.2–2.4 nm. The positive (or negative) value indicates that the magnetic easy direction is perpendicular (or parallel) to the film.

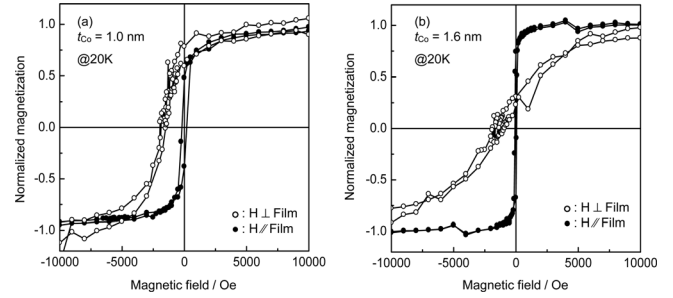


FIG. 2. The magnetization curves of the film with Co thicknesses of (a) 1.0 nm and (b) 1.6 nm. The measured temperature was 20 K. The open circles represent the data in a direction perpendicular to the film, and the closed circles represent the data in a direction parallel to the film.

Figure 2 shows the magnetization curves measured at 20 K. The Co thicknesses were (a) 1.0 nm and (b) 1.6 nm. For both films, the magnetization curve in the direction perpendicular to the film shifted in the negative direction along the field axis. This indicates that the appearance of the PEB is independent of the magnetic easy direction of the F layer. In addition to the PEB, a low in-plane exchange bias of several 10 Oe was observed, which was much lower than the PEB. This low in-plane exchange bias was probably caused by the spin canting of Cr<sup>3+</sup>, as discussed below. The PEB can be explained by the Cr<sup>3+</sup> spin orientation in the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer. As mentioned above, the  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer exhibited a (0001) orientation perpendicular to the film. According to the Cr<sup>3+</sup> spin orientation in bulk  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub>, the Cr<sup>3+</sup> spins are aligned along (0001).<sup>12</sup> Therefore, the Cr<sup>3+</sup> spins in the fabricated  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> layer must be aligned in a direction perpendicular to the film, which induces the PEB. This mechanism is different from the PEB mechanism for the system with the Mn-based AF layer, in which the PEB requires the perpendicular magnetic anisotropy of the F layer.<sup>13</sup>

Another characteristic of our films is the high PEB field. The shift in the magnetization curve, that is, the exchange bias field  $H_{\text{Ex}}$ , was approximately 1720 Oe for the film with  $t_{\text{Co}} = 1.0$  nm and approximately 1150 Oe for the film with  $t_{\text{Co}} = 1.6$  nm. The unidirectional magnetic anisotropy energy,  $J_K$ , was estimated as 0.26 and 0.32 erg/cm<sup>2</sup> using the relationship  $J_K \equiv H_{\text{Ex}} \cdot M_S \cdot t_F$ . For all of the investigated samples,  $J_K$  was in the range of 0.17–0.33 erg/cm<sup>2</sup>. The obtained value was consistent with our recent report<sup>8</sup> and higher than the reported value for the system that used  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> as an AF layer.<sup>5,6</sup>

Finally, we discuss the temperature-related changes in the in-plane remanence ratio ( $M_r/M_S$ ) and the in-plane coercivity ( $H_C$ ) for the film with  $t_{\text{Co}} < 1.2$  nm. Recall that the magnetic easy direction at room temperature was perpendicular to the film in this thickness range (see Fig. 1). As seen in Fig. 2(a), the magnetization reversal was gradual in the direction perpendicular to the film, whereas the in-plane magnetization reversed sharply. Furthermore, the remanence ratio and the coercivity were higher in the in-plane direction than in the perpendicular direction. These results imply that the in-plane magnetic anisotropy is induced in this temperature range. To demonstrate the correlation between the appearance of the PEB and the induced in-plane magnetic anisotropy, the temperature dependence of the PEB and  $H_C$  for the direction perpendicular to the film is shown in Fig. 3(a) and the temperature dependence of  $M_r/M_S$  and  $H_C$  for the in-plane direction

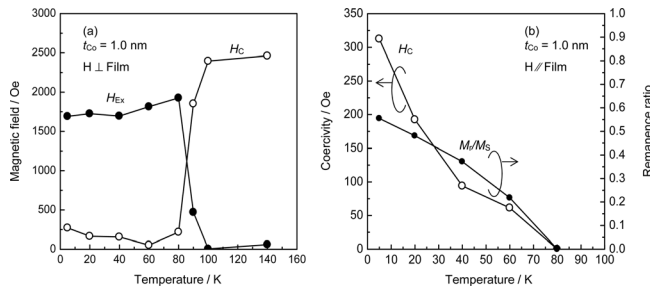


FIG. 3. (a) The temperature dependence of (a) the exchange bias field and the coercivity in the direction perpendicular to the film and (b) the remanence ratio and the coercivity in the direction parallel to the film. The Co thickness was 1.0 nm.

is shown in Fig. 3(b). The  $t_{Co}$  was 1.0 nm. As shown in Fig. 3(a), the PEB field gradually increased as a function of temperature, up to 80 K, and then sharply decreased until it reached zero at approximately 100 K. The gradual increase in the PEB below 80 K is probably caused by the spin canting of  $Cr^{3+}$ . The sharp drop of PEB can be explained by Mielkejohn and Bean's model (M-B) model.<sup>1,8,14</sup> In the M-B model, the appearance of the exchange bias is explained by the competition of the interface exchange coupling energy,  $J_{int}$ , and the magnetic anisotropy of the AF layer,  $K_{AF} \cdot t_{AF}$ , where  $K_{AF}$  is the magneto-crystalline anisotropy and  $t_{AF}$  is the thickness of the AF layer. The exchange bias appears when the condition  $J_{int} < K_{AF} \cdot t_{AF}$  is satisfied. Conversely, if  $J_{int} > K_{AF} \cdot t_{AF}$ , an enhancement in coercivity is observed because the AF spins reverse along with the magnetization of the F layer. As shown in Fig. 3(a), a sharp drop in the coercivity was observed along with the appearance of the exchange bias. That is, the sharp change of PEB and  $H_C$  occurs when  $J_{int} \approx K_{AF} \cdot t_{AF}$  is satisfied. The detailed mechanism of this characteristic temperature dependence is reported elsewhere.<sup>8</sup>

Along with the appearance of the PEB, both in-plane  $M_r/M_s$  and in-plane  $H_C$  increased below 80 K, as shown in Fig. 3(b). From these results, it is likely that the PEB induced the in-plane magnetic anisotropy. Although the detailed mechanism is currently unclear, we speculate that the spin canting of  $Cr^{3+}$  leads to the in-plane component of exchange coupling at the interface, which subsequently induces the in-plane magnetic

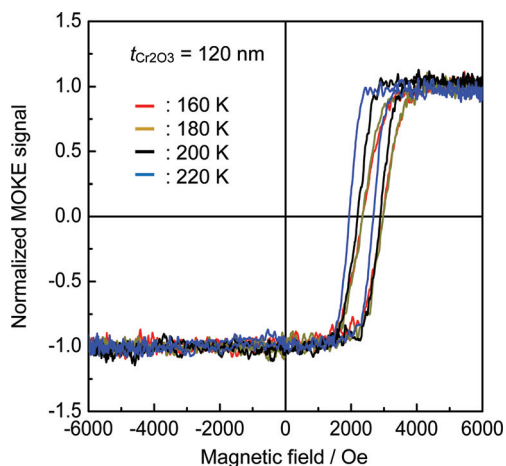


FIG. 4. (Color online) The MOKE loops in the temperature range of 160–220 K for the film with a  $Cr_2O_3$  thickness of 120 nm. The applied magnetic field was perpendicular to the film.

anisotropy. If spin canting is occurring, an in-plane exchange bias should be observed. The low in-plane exchange bias mentioned in Fig. 2 supports this argument. Furthermore, we suggest that the spin canting of  $Cr^{3+}$  in the  $\alpha$ - $Cr_2O_3(0001)$  thin film occurs below 180 K.<sup>15</sup> These hypotheses were indirectly verified through the gradual reversal of the magnetization in the direction perpendicular to the film. The MOKE loops for a film with a  $\alpha$ - $Cr_2O_3(0001)$  layer thickness of 120 nm are shown in Fig. 4. The magnetic field was applied in a direction perpendicular to the film. The magnetic field during the cooling of the sample was  $-4.0$  kOe, and thus the sign of the exchange bias was positive. The measurement temperatures were 120, 260, 180, and 220 K. Although the details will be discussed elsewhere,<sup>8</sup> the onset temperature of the PEB increased to approximately 230 K because of the high  $\alpha$ - $Cr_2O_3$  thickness. A sharp magnetization reversal was observed between 220 and 180 K in a direction perpendicular to the film, which became gradual below 180 K. Although the gradual reversal of the magnetization in a direction perpendicular to the film is indirect evidence of spin canting of  $Cr^{3+}$ , it also supports the above discussions on the mechanism of the in-plane magnetic anisotropy.

In summary, we investigated the PEB in a Pt/Co/ $\alpha$ - $Cr_2O_3(0001)$  thin film grown on an  $\alpha$ - $Al_2O_3(0001)$  substrate. We demonstrated the appearance of perpendicular unidirectional magnetic anisotropy measuring approximately  $0.17$ – $0.33$  erg/cm<sup>2</sup> and showed that its direction was not affected by the magnetic easy direction of the film. The appearance of perpendicular exchange bias was accompanied by the induction of the in-plane magnetic anisotropy. We speculate that the induced in-plane magnetic anisotropy is caused by the spin canting of  $Cr^{3+}$ .

## ACKNOWLEDGMENTS

This work was partly supported by the Encouragement of Young Scientists (B) and Priority Assistance for the Formation of Worldwide Renowned Centers of Research—The Global COE Program (Project: Center of Excellence for Advanced Structural and Functional Materials Design) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan.

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