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# Uniaxial magnetocrystalline anisotropy for $c$ -plane oriented $\text{Co}_{100-x}\text{M}_x$ ( $M$ : Cr, Mo, W) film with stacking faults

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Stacking faults (SFs) in Co-based alloy grains in a  $\text{Co}_{100-x}\text{M}_x$  ( $M$ : Cr, Mo, and W) film are evaluated by means of in-plane x-ray diffraction. Moreover, the correlation between SFs and uniaxial magnetocrystalline anisotropy  $K_u$  is discussed in connection with the spin-orbit interaction. The ratio of the integrated intensities of the (10.0) to (11.0) diffractions corrected by Lorentz and atomic scattering factors has been proposed as an index for SFs in hcp films with a  $c$ -plane sheet texture. This ratio is equal to 0.25 for perfect hcp stacking, while it is 0 for perfect fcc specific stacking. It has a one-to-one correspondence with the probability of  $-A-B-C-$  atomic-layer stacking  $P_{\text{fcc}}$ . Using this index, pure sputtered Co films are found to have a  $P_{\text{fcc}}$  of 10%. The addition of only 5 at. % of Mo or W into the Co grains reduces  $P_{\text{fcc}}$  to 2%.  $K_u$  was found to increase with the addition of material (e.g.,  $K_u$  was  $4.0 \times 10^6$  ergs/cm<sup>3</sup> for 5 at. % Mo), although the atomic magnetic moment of Co decreases monotonously. A  $P_{\text{fcc}}$  of 10% is found to lower  $K_u$  in a pure Co film by more than a factor of 2 when the spin-orbit interaction is taken into account. © 2009 American Institute of Physics. [DOI: 10.1063/1.3079768]

## I. INTRODUCTION

Co-based alloys are used as magnetic recording media because of their high uniaxial magnetocrystalline anisotropy  $K_u$ . However, they are required to have even higher  $K_u$  for realizing high-density media in the future. From a structural standpoint,  $K_u$  for Co-based alloys is thought to be determined by alloy composition,<sup>1</sup> the crystal-axial ratio  $c/a$ ,<sup>2</sup> and the stacking faults (SFs) introduced into hcp grains.<sup>3-7</sup> There have been few reports of methods for quantitatively evaluating SFs for Co-based alloy films. One method is directly counting the fcc stackings in grains observed in cross-sectional transmission electron microscope images.<sup>5</sup> Another method involves analyzing the broadening of x-ray diffraction (XRD) peaks generated by irradiating samples with synchrotron radiation<sup>6,7</sup> by Warren's formula.<sup>8</sup> However, both these methods do not lend themselves to statistical studies and immediate material development since they are time consuming and expensive. In this context, we proposed an intensity ratio measured by using laboratory-based XRD as a convenient index for evaluating SFs induced in hcp grains<sup>9-11</sup> (hereafter, hcp grains with SFs are simply described as "pseudo-hcp grains,"<sup>12</sup> as shown in Fig. 1). The diffractions used in this proposed method are observed in an in-plane XRD profile and originate from pseudo-hcp grains that have a  $c$ -plane sheet texture and that are randomly oriented in the in-plane direction. In this paper, SFs are quantitatively evaluated, and the correlation between SFs and  $K_u$  for Co-based alloy films is discussed in connection with the spin-orbit interaction.

## II. EXPERIMENTAL PROCEDURE

All samples prepared in the present study were fabricated by direct-current (dc) magnetron sputtering under an Ar gas pressure of 0.6 Pa onto glass substrates at room temperature. The layered structure of the samples was glass/NiFeCr (20 nm)/Ru (20 nm)/ $\text{Co}_{100-x}\text{M}_x$  (20 nm)/C (7 nm). Cr, Mo, and W were selected as the additional element  $M$  since they have large solid solubilities in host Co and are almost unpolarized themselves in Co  $M$  alloys, simplifying the analysis of the spin-orbit interaction.  $\text{Co}_{100-x}\text{M}_x$  films were cosputtered with Co and the additional metal targets by varying the discharge power for each target. The error in the thickness was confirmed to be at most 2%. The film compositions were verified by x-ray fluorescence analysis, and the crystalline structures were examined by XRD using Cu  $K\alpha$  radiation (RIGAKU Corp., ATX-G). The incident angle of x rays in in-plane XRD was fixed at  $0.4^\circ$ , so that the profiles

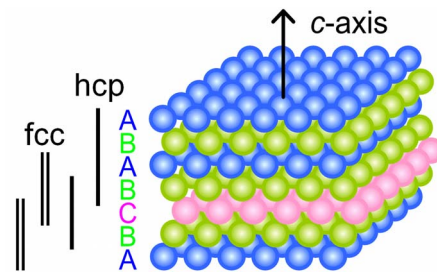


FIG. 1. (Color online) A typical example of the pseudo-hcp structure. The pseudo-hcp structure is defined as layered stacking form retaining uniaxial symmetry and consists of three kinds of closed-packed atomic planes, A, B, and C. As shown by solid and double lines, pure-fcc and pure-hcp parts cannot be discriminated anymore (Refs. 9-13), which is a different treatment from Ref. 4.

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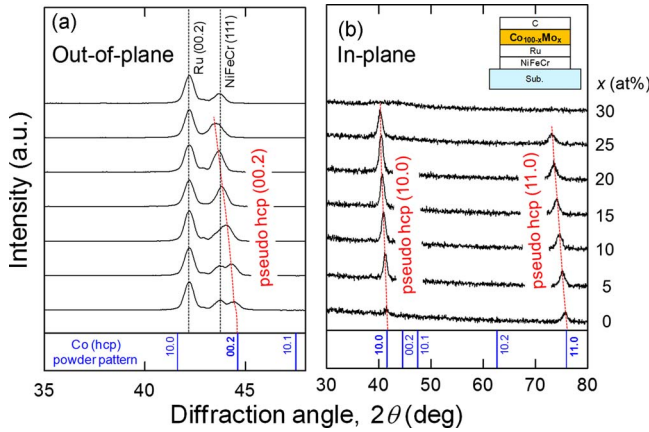


FIG. 2. (Color online) (a) Out-of-plane and (b) in-plane XRD profiles for  $\text{Co}_{100-x}\text{Mo}_x$  films with a layer structure of glass/NiFeCr (20 nm)/Ru (20 nm)/ $\text{Co}_{100-x}\text{Mo}_x$  (20 nm)/C (7 nm) for various Mo contents  $x$ . Ru and NiFeCr exhibit  $c$ -plane sheet texture of the hcp and pseudo-hcp structures. A powder pattern for hcp Co is also shown for reference.

reflect structural information at a depth of approximately 20 nm from the film surface. The irradiated area on the sample was fixed during the in-plane XRD measurement. The magnetic properties were measured using a vibrating sample magnetometer and a torque magnetometer. By accounting for the self-energy,  $K_u$  can be expressed as

$$K_u = L_{2\theta}^{\text{sat}} + 2\pi M_s^2. \quad (1)$$

Here,  $L_{2\theta}^{\text{sat}}$  is the saturated torque coefficient of twofold component and  $M_s$  is the saturation magnetization.

### III. RESULTS AND DISCUSSION

#### A. Structure

As a typical example, Fig. 2 shows (a) out-of-plane and (b) in-plane XRD profiles for  $\text{Co}_{100-x}\text{Mo}_x$  films with various Mo contents. As  $x$  increases to 25%, the diffraction lines around  $44^\circ$  observed in (a) and those from  $41^\circ$  and  $75^\circ$  observed in (b) gradually shift toward lower angles. These diffractions were identified as those from the hcp (00.2), (10.0), and (11.0) planes, respectively. Therefore, it is conjectured that a pseudo-hcp solid solution with a  $c$ -plane sheet texture is formed in  $\text{Co}_{100-x}\text{Mo}_x$  films ( $x \leq 25$  at. %). The integrated diffraction intensity from the (11.0) plane  $I_H$  is larger than that from the (10.0) plane  $I_L$  for the Co film [see Fig. 2(b)], whereas  $I_L$  becomes larger than  $I_H$  as  $x$  increases. This tendency can be physically summarized by  $I_L/I_H$  corrected by Lorentz factors  $L_L$ ,  $L_H$  and atomic scattering factors  $f_L$ ,  $f_H$ . Here, corrected  $I_L/I_H$  is defined as

$$\text{corrected } I_L/I_H = (I_L/I_H) \cdot (|f_H^2/L_H)/(|f_L^2/L_L). \quad (2)$$

Subscripts  $L$  and  $H$  correspond to (10.0) and (11.0) diffractions, respectively. Through the introduction of Eq. (2), effects of the Debye–Waller factor and thermal diffuse scattering was neglected for simplification. The Lorentz factor  $L_{L,H}$  used in this study is a product of geometric factor and polarization factor given by

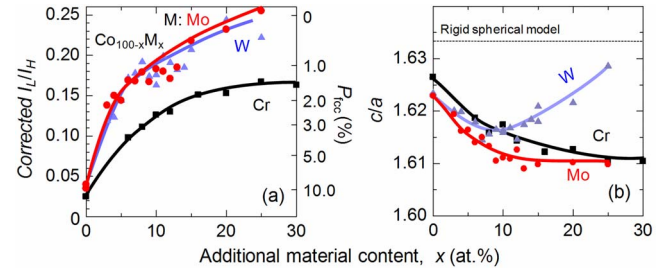


FIG. 3. (Color online) (a) Integrated intensity ratio  $I_L/I_H$  corrected by Lorentz and atomic scattering factors for  $\text{Co}_{100-x}\text{M}_x$  films and (b)  $c/a$  of  $\text{Co}_{100-x}\text{M}_x$  grains as a function of  $M$  content  $x$ . In (a), the right vertical axis indicates the probability of  $-A-B-C-$  atomic-layer stacking,  $P_{\text{fcc}}$ . The broken line in (b) indicates  $c/a$  value for the rigid sphere model.

$$L_{L,H} = (1 + \cos^2 2\theta_{L,H})/2 \sin^2 \theta_{L,H} \cos \theta_{L,H}. \quad (3)$$

For the present disordered solid solution, the compositional-averaged atomic scattering factor can be used as  $f_L$  and  $f_H$ . According to crystallographic approach, structure factor reveals that the  $I_L$  and  $I_H$  do not depend on the stacking sequence of atomic layers; in detail,  $I_H$  includes no information of the SFs, whereas  $I_L$  shows an imbalance of the atomic-layer number among  $A$ ,  $B$ , and  $C$  layers, which indirectly reflects information regarding the SFs. For example, corrected  $I_L/I_H$  is 0.25 for perfect hcp stacking and it is 0 for perfect fcc stacking.<sup>9–11</sup>

Figure 3(a) shows corrected  $I_L/I_H$  for various  $\text{Co}_{100-x}\text{M}_x$  films as a function of additional material content  $x$ . The right vertical axis represents the probability of  $-A-B-C-$  atomic-layer stacking,  $P_{\text{fcc}}$ , which was demonstrated to have a one-to-one correspondence with the corrected  $I_L/I_H$ .<sup>11</sup> The pure Co film has a corrected  $I_L/I_H$  of only 0.04; when Cr, Mo, and W are added to the Co film,  $I_L/I_H$  increases to over 0.14. Corrected  $I_L/I_H$  of 0.04 and 0.14 correspond to  $P_{\text{fcc}}$  of 10% and 2%, respectively. This demonstrates that only a 5 at. % addition of Mo or W into Co grains reduces  $P_{\text{fcc}}$  from 10% to 2%. Figure 3(b) shows  $c/a$  for  $\text{Co}_{100-x}\text{M}_x$  films with a pseudo-hcp structure plotted against  $M$  content  $x$ . The lattice constants  $c$  and  $a$  are respectively derived from the lattice spacings of (00.2) and (11.0) planes,  $d_{00.2}$  and  $d_{11.0}$ , as follows:

$$c = 2d_{00.2}, \quad a = 2d_{11.0}. \quad (4)$$

The measurement error of  $c/a$  is very small, being approximately 0.003, judging from the three data points for the Co film [see  $x=0$  in Fig. 3(b)]. In the case of  $M=\text{Cr}$  and Mo,  $c/a$  decreases monotonously with increasing  $x$ . On the other hand, for  $M=\text{W}$ ,  $c/a$  has a minimum at around  $x=10$  at. %. Thus, common characteristics of Co  $M$  films ( $M$ : Cr, Mo, W) were not observed in the dependence of  $c/a$  on  $M$  content. Averaged in-plane grain sizes derived from Scherrer's equation for Co  $M$  films were 6.7–10.1 nm.

#### B. Intrinsic magnetic properties

Figures 4(a) and 4(b) show the magnetization per one Co atom,  $m^{\text{Co}}$ , and the uniaxial magnetocrystalline anisotropy  $K_u$  as functions of  $M$  content  $x$ . Here,  $m^{\text{Co}}$  was derived by assuming that  $M$  in pseudo-hcp Co  $M$  grains was completely

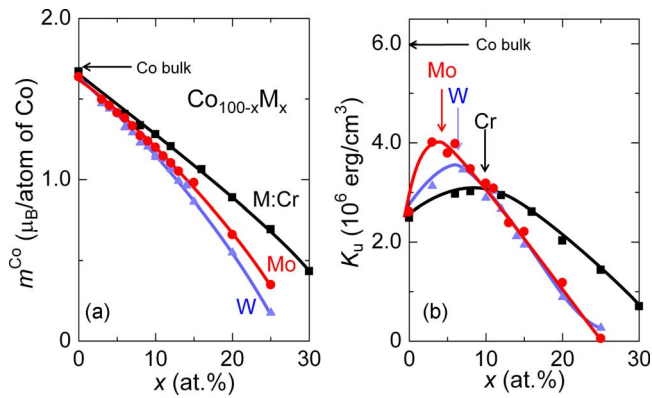


FIG. 4. (Color online) Dependence of (a) moment per Co atom  $m^{\text{Co}}$  and (b)  $K_u$  for  $\text{Co}_{100-x}\text{M}_x$  films on  $M$  content  $x$ . Arrows in the figure indicate the magnitude of  $m^{\text{Co}}=1.7\mu_B$  (Ref. 14) and  $K_u=5.97\times 10^6$  ergs/cm<sup>3</sup> (Ref. 15) for Co bulk samples.

nonpolarized. As Fig. 4(a) shows,  $m^{\text{Co}}$  decreases monotonously from  $1.64\mu_B$  with the addition of the nonmagnetic elements Cr, Mo, and W. In contrast, Fig. 4(b) shows that  $K_u$  has maximum values of  $4.0\times 10^6$  ergs/cm<sup>3</sup> at 5 at. % Mo,  $3.5\times 10^6$  ergs/cm<sup>3</sup> for 7 at. % W, and  $3.0\times 10^6$  ergs/cm<sup>3</sup> for 10 at. % Cr.

### C. Effect of stacking faults on $K_u$

In general, the spin-orbit interaction  $\lambda LS$  is thought to be a dominant factor in determining  $K_u$  of Co  $M$  film, where  $\lambda$  is the coefficient of the spin-orbit interaction,  $L$  is the orbital moment, and  $S$  is the spin moment. The atomic magnetic moment of Co in Co  $M$  films,  $m^{\text{Co}}$ , is given by

$$m^{\text{Co}} = m_L^{\text{Co}} + m_S^{\text{Co}} \sim m_S^{\text{Co}} \propto S, \quad (5)$$

where  $m_L^{\text{Co}}$  is the orbital magnetic moment and  $m_S^{\text{Co}}$  is the spin magnetic moment per Co atom. As Eq. (5) shows,  $m^{\text{Co}}$  is linear with respect to  $S$ ; this is because Co is a 3d transition metal, so that  $m_L^{\text{Co}}$  is negligible small.<sup>16</sup> Therefore,  $K_u$  determined from a Co atom versus  $m^{\text{Co}}$  plot is expected to reveal physical information on  $\lambda L$  ( $K_u \propto \lambda LS \propto \lambda L m^{\text{Co}}$ ).

Figure 5 shows the relationship between  $K_u$  and the atomic magnetic moment per Co atom for Co  $M$  films. In this figure, the dotted lines indicate equi-value lines for  $P_{\text{fcc}}=2.5\%$ , 3.0%, and 10%. For all additional elements,  $K_u$  has a linear dependence on  $m^{\text{Co}}$  when  $m^{\text{Co}}$  is less than  $1.2\mu_B$ . Moreover, it was found that the samples in this region have  $P_{\text{fcc}}<2\%$  (see shaded region in Fig. 5). This implies that  $\lambda L$  is almost constant since the stacking structure of the atomic planes is close to perfect hcp order (i.e.,  $-A-B-A-B-$ ) for Co  $M$  films. On the other hand, for  $m^{\text{Co}}$  over  $1.2\mu_B$ ,  $K_u$  drops below the extrapolated lines from the linear region (indicated by the broken lines in Fig. 5). By considering the atomic stacking order,  $P_{\text{fcc}}$  is found to increase with distance from the broken line and it is 10% for the Co film ( $1.64\mu_B$ ). This indicates that  $\lambda L$  is reduced remarkably by the collapse of the hcp stacking order of the atomic plane. These results indicate that if perfect hcp stacking of the atomic plane is realized, the Co film with  $1.7\mu_B$  will have a  $K_u$  of 5.8

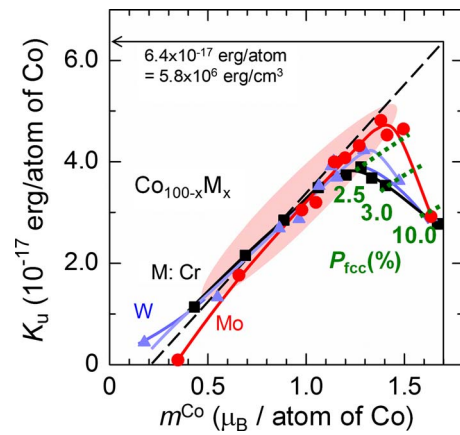


FIG. 5. (Color online) Relationship between  $K_u$  and  $m^{\text{Co}}$  per Co atom for Co  $M$  films. Dotted lines are equi-value lines of  $P_{\text{fcc}}=2.5\%$ , 3.0%, and 10%. Shaded region indicates  $P_{\text{fcc}}<2\%$  (i.e., atomic plane stacking is close to perfect hcp order). The solid arrow indicates that a Co film with  $1.7\mu_B$  will have  $K_u=5.8\times 10^6$  ergs/cm<sup>3</sup> if perfect hcp stacking is achieved.

$\times 10^6$  ergs/cm<sup>3</sup> (corresponding to  $6.4\times 10^{-17}$  ergs/atom, see solid arrows in Fig. 5), which is almost the same value as that for bulk-hcp Co.<sup>15</sup> In other words,  $K_u$  for the pure Co film with pseudo-hcp with a  $c$ -plane sheet texture sputtered used in this study is reduced by more than a factor of 2 due to  $P_{\text{fcc}}$  of about 10%. We are currently verifying the validity of this conclusion by evaluating the magnetic characteristics at low temperature.

### ACKNOWLEDGMENTS

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- <sup>1</sup>For example, A. Ishikawa and R. Sinclair, *IEEE Trans. Magn.* **32**, 3605 (1996).
- <sup>2</sup>For example, S. Chikazumi, T. Wakiyama, and K. Yoshida, *Proceedings of the International Conference on Magnetism*, 1964 (unpublished), p. 756.
- <sup>3</sup>G. B. Mitra and N. C. Halder, *Acta Crystallogr.* **17**, 817 (1964).
- <sup>4</sup>G. Ju, B. Lu, R. J. M. van de Veerdonk, X. Wu, T. J. Klemmer, H. Zhou, R. Chantrell, A. Sunder, P. Asselin, Y. Kubota, and D. Weller, *IEEE Trans. Magn.* **43**, 627 (2007).
- <sup>5</sup>Y. Takahashi, K. Tanahashi, and Y. Hosoe, *J. Appl. Phys.* **91**, 8022 (2002).
- <sup>6</sup>M. F. Toney, E. E. Marinero, M. F. Doerner, and P. M. Rice, *J. Appl. Phys.* **94**, 4018 (2003).
- <sup>7</sup>T. Kubo, Y. Kuboki, R. Tanuma, A. Saito, S. Watanabe, and T. Shimatsu, *J. Appl. Phys.* **99**, 08G911 (2006).
- <sup>8</sup>B. E. Warren, *X-ray Diffraction* (Addison-Wesley Pub. Co., Reading, MA, 1968), p. 284.
- <sup>9</sup>S. Saito, A. Hashimoto, and M. Takahashi, *Digests of the 32nd Annual Conference of The Magnetic Society of Japan*, 2007 (unpublished), Paper No. 13pE-1.
- <sup>10</sup>M. Takahashi and S. Saito, *J. Magn. Magn. Mater.* **320**, 2868 (2008).
- <sup>11</sup>S. Saito, R. Yanagisawa, D. Hasegawa, and M. Takahashi, *Digests of the 32nd Annual Conference of The Magnetic Society of Japan*, 2008 (unpublished), Paper No. 12pA-13.
- <sup>12</sup>A. Hashimoto, S. Saito, N. Itagaki, and M. Takahashi, *J. Phys. D* **41**, 012002 (2008).
- <sup>13</sup>A. Hashimoto, S. Saito, N. Itagaki, and M. Takahashi, *Appl. Phys. Lett.* **89**, 262508 (2006).
- <sup>14</sup>A. J. P. Meyer and P. Taglang, *C. R. Hebd. Seances Acad. Sci.* **231**, 612 (1950).
- <sup>15</sup>R. Pauthenet, Y. Barnier, and G. Rimet, *J. Phys. Soc. Jpn.* **17**, 309 (1962).
- <sup>16</sup>For example, R. A. Reck and D. L. Fry, *Phys. Rev.* **184**, 492 (1969).