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LETTER TO THE EDITOR

Influence of 5d transition elements on the magnetocrystalline anisotropy of hcp-Co

Nobuaki Kikuchi[†], Osamu Kitakami[†], Satoshi Okamoto[†], Yutaka Shimada[†], Akimasa Sakuma[‡], Yoshichika Otani[§] and Kazuaki Fukamichi[§]

 † Research Institute for Scientific Measurements, Tohoku University, 2-1-1 Katahira, Sendai 980-8577, Japan
‡ Magnetic and Electric Materials Research Laboratory, Hitachi Metals Ltd,

¹ Magnetic and Electric Materials Research Laboratory, Infacti Metals Elu

Kumagaya 360-0743, Japan

§ Department of Materials Science, Graduate School of Engineering, Tohoku University, Sendai 980-8579, Japan

E-mail: kitakami@rism.tohoku.ac.jp

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Abstract. We have studied the effects of 5d transition elements (X = W, Re, Ir, and Pt) on the magnetocrystalline anisotropy energy (MAE) of epitaxially grown hexagonal-close-packed (hcp)-Co (1010) single crystal films. The MAE of Co–X significantly changes, depending on X. With increasing numbers of valence electrons in X, the sign of the first anisotropy constant K_1 changes from positive to negative, indicating that the easy axis of magnetization rotates from the *c*-axis to the *c*-plane. The second anisotropy constant K_2 monotonically increases with the number of valence electrons in X. All these variations were found to be systematic and are discussed in conjunction with the structural analyses.

From the practical point of view, hexagonal-close-packed (hcp)-Co-based alloy is one of the most important magnetic materials, especially for high-density magnetic recording media. It is well known that 5d transition elements, such as Ta and/or Pt, improve the recording characteristics of Co thin-film media [1–5]. In spite of intensive studies, however, very little reliable data are available about the effects of these 5d transition elements on the magnetocrystalline anisotropy energy (MAE) of hcp-Co. Since 5d transition elements possess very large spin–orbit coupling, it is expected that these elements significantly influence the MAE of ferromagnetic 3d metals. For example, for ordered 3d–5d alloys with a AuCu structure, such as FePt and CoPt, the 5d element greatly enhances the MAE through its large spin–orbit coupling and strong 3d–5d hybridization [6–8]. Although some theoretical work has clarified the electronic state of 3d, 4d [9, 10], and 5d elements [11, 12] in 3d ferromagnets in order to explain existing experimental results [13, 14], no systematic studies have been carried out for their effects on the MAE because of difficulty in theoretical calculations.

Recently, we have investigated the effects of Ta and Pt on the MAE of Co and Co–Cr alloy thin-film recording media [15]. In this letter, the previous work is extended to a series of 5d elements X (X = Hf, Ta, W, Re, Ir, and Pt), and we report a unique feature of the MAE of Co–X alloys with a hcp structure.

The Cr(211) buffer layer and the Co_{1-x}X_x (1010) layer were epitaxially grown on MgO(110) at a substrate temperature of 300 °C by a sputtering method. The sputtering chamber had a base pressure of $\leq 5 \times 10^{-7}$ Torr, and the depositions were carried out at an argon gas pressure of 5 mTorr. The deposition rates for Cr and Co-X were 0.70



Figure 1. The pole figures of Co–Ir $\{10\overline{1}0\}$ and MgO $\{111\}$ from Co–Ir(800 Å)/Cr(400 Å)/MgO(110). We note the excellent epitaxial growth of Co–Ir $(10\overline{1}0)$ on MgO(110) with the orientation relationship of Cr–Ir $[0001] \parallel \text{MgO}[100]$.



Figure 2. The lattice parameters *a* and *c* and the axial ratio c/a of hcp-Co_{1-*x*}X_{*x*} (X = W, Re, Ir and Pt) as a function of 5d element concentration *x*.

and 0.25 Å s⁻¹, and their thicknesses were controlled to 400 and 800 Å, respectively. The film compositions were determined by x-ray photoelectron spectroscopy (XPS) using Mg K α radiation. The lattice constants and the crystallographic orientation were carefully checked by x-ray diffractometry, and the resultant orientation relationship was identified as MgO(110)[100] || Cr(211)[110] || Co–X(1010)[0001]. The pole figures of MgO{111} and Co–Ir{1010} in figure 1 clearly show an excellent single crystal growth of Co–Ir(1010). Similar epitaxial growth was also confirmed for all Co–X samples prepared in the present study, except Hf and Ta, for which it is difficult to form a uniform solid solution with Co [16, 17]. The room-temperature magnetic properties were measured with a vibrating sample magnetometer and a torque magnetometer. Both the Sucksmith–Thompson method [18] and torque magnetometry were used for determination of the magnetic anisotropy constants K_1 and K_2 and both methods gave the same anisotropy constants.

Figure 2 shows the variation of the lattice constants a and c and the axial ratio c/a as a



Figure 3. Typical magnetization curves of (a) pure Co and (b) $Co_{0.935}W_{0.065}$ single crystal films. The solid lines and dotted lines, respectively, show the curves measured in MgO[100] and [110]. It is noticed that the curve shape in the hard axis (in this case MgO[110]) changes from convex to almost linear by addition of W, indicating that the higher order term K_2 is appreciably suppressed.

function of the 5d element concentration x for various kinds of hcp $Co_{1-x}X_x$ (X = W, Re, Ir, and Pt). It should be noted that all 5d elements except Pt show the same trend; a linearly increases and c remains almost constant with increasing x, resulting in a remarkable decrease of the axial ratio c/a from 1.623 (x = 0) to 1.600 ($x \sim 0.2$); the value should be 1.633 for ideal stacking sequence. In contrast, Pt yields obvious deviation from this trend; both a and c increase linearly with x in accordance with the well known Vegard's law, yielding a slight decrease in the axial ratio c/a. Since the MAE is very sensitive to the crystal lattice, it is expected that Pt would have different effects on the MAE of hcp-Co from other 5d elements. In fact, the MAE of Co–Pt is different from the trend of other Co–X alloys, as will be mentioned later.

The typical M-H curves of pure Co and $Co_{0.935}W_{0.065}$ are shown in figures 3(a) and 3(b), respectively. Both samples exhibit ideally uniaxial properties where the *c*-axis of the hcp structure coincides with MgO[100]. It is noticed that the M-H shape along the hard axis (in this case MgO[110]) changes from convex to almost linear by addition of W, indicating that the higher-order term K_2 is suppressed appreciably. Figure 4 shows a plot of the saturation magnetization M_s , and the anisotropy constants K_1 and K_2 of Co–W, Re, Ir and Pt as functions of the 5d element concentration x. For W with the number of valence electrons n = 6, K_1 increases and K_2 decreases with increasing x. The maximum anisotropy energy per Co atom is 1.7 times larger than that of pure Co. Note that the significant reduction of K_2 gives rise to linear hard magnetization curve, as shown in figure 3(b). Re (n = 7) reduces both K_1 and K_2 monotonically. A surprising behaviour of the MAE was found in the Co-Ir (n = 9) system. As the Ir content increases, the sign of K_1 changes from positive to negative around x = 0.06, indicating that the easy axis of magnetization rotates from the *c*-axis (in this case MgO[100]) to the orthogonal direction (MgO[110]), as shown clearly in figure 5. From the results mentioned above, the 5d element with a larger n tends to change the magnetization easy axis of hcp-Co from the *c*-axis to the *c*-plane. This tendency can be seen more clearly in figure 6 by replotting figure 5. By rewriting the expression for the magnetic anisotropy energy $E(\theta) = K_1 \sin^2 \theta + K_2 \sin^4 \theta$, the energy can be divided into the two-fold and four-fold



Figure 4. Variation of the saturation magnetization M_s , and the magnetic anisotropy constants K_1 and K_2 of hpc-Co_{1-x}X_x (X = W, Re, Ir and Pt) as a function of 5d element concentration x.



Figure 5. Angular dependence of the MAE of $Co_{1-x}Ir_x$. Note that the easy axis of magnetization changes from *c*-axis (in this case MgO[100]) to *c*-plane (MgO[110]).



Figure 6. (a) Two-fold A_2 and (b) four-fold A_4 symmetry components of the MAE of hcp-Co_{1-x} X_x (X = W, Re, Ir and Pt) as a function of the number of valence electrons of 5d transition element X. The arrows correspond to the values for pure hcp-Co.

symmetry components as

 $E(\theta) = -1/2(K_1 + K_2)\cos 2\theta + K_2/8\cos 4\theta = -A_2\cos 2\theta + A_4\cos 4\theta \quad (1)$

with $A_2 = (K_1 + K_2)/2$ and $A_4 = K_2/8$. Figures 6(a) and 6(b), respectively, show the two components A_2 and A_4 against the 5d element for fixed x. It is worth noting that the two-fold symmetry component changes its sign from positive to negative with increasing number of valence electrons n in the 5d elements. This tendency is significantly enhanced with increasing the concentration x. For the four-fold symmetry component, A_4 is decreased by the addition of the 5d element and this effect is less pronounced for the element with a larger n. Thus the two-fold and four-fold symmetry components of the MAE of Co–W, Re, and Ir vary systematically as a function of n. In contrast with these 5d elements whose lattice parameters are the same at a fixed x (figure 2), the MAE of Co–Pt is obviously different from this trend. As can be seen in figure 6, A_2 remains almost constant but A_4 increases appreciably with increasing x, reflecting the significant increase in K_2 by addition of Pt shown in figure 4. This difference from the systematic variation of other 5d elements would be attributed to the quite different lattice parameters of Co–Pt (figure 2), because the MAE is generally very sensitive to the crystal structure [7, 19].

In the present work, we have clarified the very unique effects of 5d elements on the MAE

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of hcp-Co. In order to explain the present results theoretically, sophisticated first-principle calculations would be required, because the band splitting due to the spin–orbit coupling responsible for the MAE is two or three orders smaller than the band width. One may say that the behaviour of the MAE in figures 4 and 6 can be explained by the so-called band-filling effect within the framework of the rigid band scheme. This picture might be applicable to the case for fixed x where the lattice parameters are almost independent of 5d element species except Pt. For each x, at a first glance, the monotonic decrease of A_2 and increase of A_4 with n in figure 6 may be understood by this band-filling effect. However, this argument seems to be inappropriate for the 5d element concentration dependence of the MAE, because the lattice parameters depend greatly on the concentration x, as shown in figure 2. According to the recent first-principles calculations by Daalderop *et al* [6] and Sakuma [7, 19], the rigid band scheme is obviously inappropriate for the MAE of the ordered 3d–5d alloys with AuCu structure, such as FeIr, FePt, CoIr, and CoPt.

In summary, we have investigated the MAE of Co-5d transition element alloys systematically, and have found that the MAE varies significantly depending on the 5d element species. The early 5d element W enhances the first anisotropy constant K_1 and decreases the second constant K_2 . The 5d element with larger valence electron number *n* decreases K_1 and changes its sign from positive to negative, accompanied by the enhancement of K_2 . For Pt, however, the behaviour of the MAE is different from the systematic trend of other 5d elements, because the lattice parameters vary quite differently from those of other elements.

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