Electric field-induced change in magnetocrystalline anisotropy in ferromagnetic transition-metal thin films

Kohji Nakamura1, Riki Shimabukuro1, Toru Akiyama1, Tomonori Ito1 and A. J. Freeman2

1 Department of Physics Engineering, Mie University, Tsu, Mie 514-8507, JAPAN
2 Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208, USA
E-mail: kohji@phen.mie-u.ac.jp

Abstract. In a search for promising ferromagnets exhibiting a giant magnetocrystalline anisotropy (MCA) change by an external electric field, the MCA in the Fe-Co(001) alloy monolayers is determined by means of full-potential linearized augmented plane-wave method. A large MCA change by the electric field is found to appear in the Fe0.75Co0.25 monolayer, and a reasonable origin from a band structural change in the minority-spin d states is obtained, where a position of Fermi level relative to the d band level is a key factor for designing the giant MCA change.

Interest in controlling and designing magnetic properties by an external electric field has increased for both basic and applied physics. Recently, the electric field-induced change in the magnetocrystalline anisotropy (MCA) in itinerant thin film ferromagnets has been demonstrated experimentally in thin film FePt and FePd with liquid interfaces,[1] and in ultrathin Fe/MgO and Fe/GaAs junctions.[2, 3] Motivated by these experimental findings, we also investigated the effects of an electric field on the MCA in transition-metal thin monolayers by means of the full-potential linearized augmented plane-wave (FLAPW) method,[4] and found that the MCA in an Fe(001) monolayer [but not in Co(001) and Ni(001) monolayers] is modified by the electric field through a change in band structure, in which the p-d hybridization near the Fermi level ($E_F$) plays a key role.

Here, in a search for promising ferromagnets exhibiting a giant MCA change by an electric field, we extend our calculations to the Fe-Co(001) alloy monolayer systems. Indeed, a large MCA change is found to appear in the Fe0.75Co0.25 monolayer, and a reasonable origin from a band structural change in the minority-spin d states is obtained, where the position of $E_F$ relative to the d band level is a key factor for designing the giant MCA change.

As models, the simple systems of free-standing Fe$_{1-x}$Co$_x$(001) monolayers ($x=0$, 0.25 and 0.5) with the lattice constant of 5.45 a.u. are employed. Although the effect of substrates might be of importance, our analysis described below may be valid for cases with ceramic substrates, since their substrates lack electronic states at $E_F$ which minimize the interface hybridization.[5]

Calculations are performed by the highly precise all-electron FLAPW method,[6, 7, 8] which treats film geometries, allowing a natural way to include an external electric field. The electric field potential applied along the surface normal (z-axis), $v_{ext} = F_{ext} z$, is expanded into...
Table 1. Calculated $E_{\text{MCA}}$ (in meV/atom) in an electric field of zero and 1 V/Å and their difference, $\Delta E_{\text{MCA}} = E_{\text{MCA}}(1 \text{ V/Å}) - E_{\text{MCA}}(0 \text{ V/Å})$, for Fe$_{1-x}$Co$_x$(001) monolayers ($x=0, 0.25$ and 0.5).

|          | zero field | 1 V/Å | $\Delta E_{\text{MCA}}$ |
|----------|------------|-------|--------------------------|
| Fe       | 0.19       | 0.02  | -0.17                    |
| Fe$_{0.75}$Co$_{0.25}$ | -1.52     | -0.82 | 0.70                     |
| Fe$_{0.50}$Co$_{0.50}$ | -1.74     | -1.57 | 0.17                     |

interstitial, muffin-tin (MT) spheres and vacuum regions,[4] where $F_{\text{ext}}$ and $z$ are external electric field and $z$-axis position, respectively. Having a Hamiltonian with the added $v_{\text{ext}}$, self-consistent calculations are first performed in the scalar relativistic approximation (SRA), i.e., excluding the spin-orbit coupling (SOC), based on the local spin density approximation (LSDA) using the von Barth-Hedin exchange-correlation.[9] LAPW functions with a cutoff of $|k + G| \leq 3.6$ a.u. and MT sphere radii of 2.3 a.u. for Fe and Co are used, where the angular momentum expansion inside the MT spheres is truncated at $\ell = 8$ for wave functions, charge and spin density and potential.

To determine the MCA, the second variational method[10, 11] for treating the SOC is performed by using the calculated eigenvectors in the SRA, and the MCA energy, $E_{\text{MCA}}$, is determined by the force theorem,[12, 13] which is defined as the energy eigenvalue difference for the magnetization oriented along the in-plane [100] and out-of-plane [001] directions. With 7,056 special $k$-points in the two-dimensional Brillouin zone (BZ), the $E_{\text{MCA}}$ was found to sufficiently suppress numerical fluctuations.

Table 1 summaries the calculated $E_{\text{MCA}}$ in an electric field of zero and 1 V/Å and their difference, $\Delta E_{\text{MCA}} = E_{\text{MCA}}(1 \text{ V/Å}) - E_{\text{MCA}}(0 \text{ V/Å})$, for the Fe$_{1-x}$Co$_x$(001) monolayers. For the Fe monolayer, as demonstrated previously,[4] the $E_{\text{MCA}}$ in zero field has positive value, indicating out-of-plane MCA, and the $E_{\text{MCA}}$ decreases by 0.17 meV/atom when the electric field is introduced. A maximum change in the $E_{\text{MCA}}$ by 0.7 meV/atom is found in the Fe$_{0.75}$Co$_{0.25}$ monolayer, although the $E_{\text{MCA}}$ in zero field has negative values and the $E_{\text{MCA}}$ increases when an electric field is introduced.

Furthermore, within a rigid band model, we analyze the behavior of the $E_{\text{MCA}}$ with respect to a variation of the number of valence electrons. The results are shown in Fig. 1 for the Fe and Fe$_{0.75}$Co$_{0.25}$ monolayers. For the Fe monolayer, the $E_{\text{MCA}}$ with a positive value at zero field decreases as the valence electron number increases, and a maximum change in the $E_{\text{MCA}}$ by an electric field is achieved at about 8.4 electrons. Thus, a tuning of the valence electron number (i.e., a position of Fermi level relative to the $d$ band level) is a key factor for designing the giant MCA change. Indeed, in the case of the Fe$_{0.75}$Co$_{0.25}$ monolayer, the behavior of the MCA is roughly identical to that in the Fe system, and the maximum change in the $E_{\text{MCA}}$ is confirmed to be at 8.25-8.3 electrons, corresponding to the number of valence electrons in the present Fe$_{0.75}$Co$_{0.25}$ system.

In order to discuss the origin of the MCA change, we calculated the partial density of states (DOS) in the MT spheres in the Fe and Fe$_{0.75}$Co$_{0.25}$ monolayers, which are shown in Fig. 2. In both monolayers, the DOS around $E_F$ arise from the minority-spin states, while the majority-spin DOS are almost fully occupied and are located from -1 to -4 eV below $E_F$. When an electric field is introduced, although the whole feature of the DOS does not alter much to that in zero field, a significant change in the DOS of the minority-spin state around $E_F$ is observed, as shown in the upper figures of Fig. 2.
Figure 1. Calculated MCA energy, $E_{\text{MCA}}$, as a function of the number of valence electrons for (a) Fe(001) and (b) Fe$_{0.75}$Co$_{0.25}$(001) monolayers. Dashed and solid lines represent results for an external electric field of zero and 1 V/Å values, respectively. The point at $N = 8$ in (a) [8.25 in (b)] corresponds to the number of valence electrons in the present Fe (Fe$_{0.75}$Co$_{0.25}$) system.

Figure 2. Density of states (DOS), $N$, in MT spheres in an electric field of zero (dotted lines) and 1 V/Å (solid lines) for (a) Fe(001) and (b) Fe$_{0.75}$Co$_{0.25}$(001) monolayers. Upper figures show the difference in the DOS, $\Delta N$, between 1 V/Å and zero field, which are further decomposed in momentum space. In both monolayers, strong $p$-$d$ hybridizations between the $d_{z^2}$ ($m = 0$) and $p_z$ ($m = 0$) orbitals, and between the $d_{xz,yz}$ ($m = \pm 1$) and $p_{x,y}$ ($m = \pm 1$) orbitals are observed due to the introduction of the electric filed, as pointed out previously.[4] According to perturbation theory,[14] the SOC between occupied and unoccupied $d$ states with the same (different) $m$ magnetic quantum number through the $L_z$ ($L_x$ and $L_y$) operator gives a positive (negative) contribution to the $E_{\text{MCA}}$. Thus, for the Fe monolayer, an enhancement of the DOS below $E_F$ and a depression above $E_F$ in both $d_{z^2}$ and $d_{xz,yz}$ bands may yield a negative contribution to the $\Delta E_{\text{MCA}}$, as mentioned in Table 1. In contrast, for the Fe$_{0.75}$Co$_{0.25}$ monolayer, since $E_F$ shifts above relative to the $d$ band level due to an increase in the number of electrons, $E_F$ lies between the enhanced regions in the DOS of the $d$ bands, which is emphasized in the Co DOS; this may lead to a positive contribution to the $\Delta E_{\text{MCA}}$.

In summary, we investigated the effects of an external electric field on the MCA in the Fe$_{1-x}$Co$_x$(001) alloy monolayers by means of the first principles FLAPW method, and found that the large enhancement/depression in the DOS of the $d$ bands around $E_F$ by the electric field yields the MCA change, where the position of $E_F$ relative to the $d$ band level is a key factor.
Figure 3. Difference in calculated partial density of states (DOS), $\Delta N$, between 1 V/Å and zero field for (a) Fe(001) and (b) Fe$_{0.75}$Co$_{0.25}$(001) monolayers, which are decomposed in momentum space. Only the DOS in the minority-spin states are drawn. $p_0$ and $p_{\pm 1}$ indicate $p_z$ and $p_{x,y}$ states, and $d_0$, $d_{\pm 1}$ and $d_{\pm 2}$ are $d_{x^2-y^2}$, $d_{xz,yz}$ and $d_{x^2-y^2}$ states, respectively.

for designing the giant MCA change.

Work at Mie University was supported by a Grant-in-Aid for Scientific Research (No. 20540334), and for computations performed at ISSP, University of Tokyo. Work at Northwestern University was supported by the U.S. Department of Energy (DE-FG02-88ER 45372).

References
[1] Weisheit M, Fähler S, Marty A, Souche Y, Poinsignon C and Givord D 2007 Science 315 349
[2] Maruyama T, Ohta K, Nozaki T, Shinjo T, Shiraishi M, Mizukami S, Ando Y and Suzuki Y 2009 Nature Nanotech. 4 158
[3] Ohta K, Maruyama T, Nozaki T, Shiraishi M, Shinjo T, Suzuki Y, Ha S-S, You C-Y, and Van Roy W 2009 Appl. Phys. Lett. 94 032501
[4] Nakamura K, Shimabukuro R, Fujiwara Y, Akiyama T, Ito T and Freeman A J 2009 Phys. Rev. Lett. 102 187201
[5] Freeman A J and Wu R 1991 J. Magn. Magn. Mater. 100 497 and references therein
[6] Wimmer E, Krakauer H, Weinert M and Freeman A J 1981 Phys. Rev. B 24 864
[7] Weinert M, Wimmer E and Freeman A J 1982 Phys. Rev. B 26 4571
[8] Nakamura K, Ito T, Freeman A J, Zhong L, and Fernandez-de-Castro J 2003 Phys. Rev. B 67 014420
[9] von Barth U and Hedin L 1972 J. Phys. C 5 1629.
[10] Wu R and Freeman A J 1999 J. Magn. Magn. Mater. 200 498
[11] Li C, Freeman A J, Jansen H J F and Fu C L 1990 Phys. Rev. B 42 5433
[12] Daalderop G H O, Kelly P J and Schuurmans M F H 1990 Phys. Rev. B 41 11919
[13] Wang X D, Wang D S, Wu R and Freeman A J 1996 J. Magn. Magn. Mater. 159 337
[14] Wang D S, Wu R and Freeman A J Phys. Rev. B 47 14932