Electric field effects on magnetocrystalline anisotropy in ferromagnetic Fe monolayers

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Abstract

The magnetocrystalline anisotropy (MCA) for an Fe monolayer on MgO substrate (Fe/MgO) and that sandwiched by MgO (MgO/Fe/MgO) is investigated by means of the first principles full-potential linearized augmented plane-wave method, and the effects of an external electric field on the MCA is discussed. In both systems, the MCA is found to be modified through a change in the *d* band structure around Fermi level by the external electric field.

Key words: magnetocrystalline anisotropy, electric field effect, Fe monolayer, first principles calculations

1. Introduction

Controlling magnetic properties by an external electric field is one key challenge in modern magnetic physics and technology, such as the use of magnetoelectric multiferroics.[1, 2] Recently, even in ferromagnetic metallic films FePt and FePb with liquid interfaces,[3] a modification in the coercivity i.e., the magnetocrystalline anisotropy (MCA), with the application of an external electric field was experimentally observed. Moreover, a direct switching of the magnetization by an applied voltage was demonstrated for ultrathin films Fe and $Fe_{0.8}Co_{0.2}$ on MgO(001) substrates.[4, 5] Motivated by these experimental findings, we also investigated the effects of an external electric field on the MCA in free-standing transition-metal monolayers by means of the full-potential linearized augmented plane-wave (FLAPW) method,[6] 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 the *d* band structure, in which the *p*-*d* hybridization near Fermi level (E_F) plays a key role.

In the present work, we extend our Fe monolayer calculations by including the MgO substrates in order to treat more realistic systems. Indeed, our calculations demonstrate that the MCA in the Fe monolayers even included the MgO substrates is modified by an external electric field. Thus, the Fe monolayers on the MgO substrates would be promising candidates exhibiting the MCA modification by the external electric field.

2. Model and method

We considered two models of Fe monolayers, an Fe monolayer on three-atomic-layer MgO substrate (Fe/MgO) and an Fe monolayer sandwiched by the three-atomic-layers MgO (MgO/Fe/MgO), where the in-plane lattice constant of the Fe monolayers is assumed to match that of the MgO(001) since the assumed lattice constant is very close to that in the bulk Fe (within a lattice mismatch of 4 %). For the atomic interfacial structures, the Fe atoms place on the top of the O atoms, and the Fe-MgO interlayer distance is assumed to be the relaxed structure parameters determined by the atomic force FLAPW calculations.[7]

Calculations were performed by the film FLAPW method,[8, 9, 10] 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 interstitial, muffin-tin (MT) spheres and vacuum regions, where F_{ext} and z are external electric field and z-axis position, respectively.[6] Having a Hamiltonian with the added the v_{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.[11] LAPW functions with a cutoff of $|\mathbf{k} + \mathbf{G}| \leq 3.9$ a.u. and MT sphere radii used for Fe, Mg, and O atoms are 2.30, 2.15. and 1.80 a.u., respectively, where the angular momentum expansion inside the MT spheres is truncated at ℓ=8 for wave functions, charge and spin density and potential.

To determine the MCA, the second variational method[12] for treating the SOC is performed by using the calculated eigenvectors in the SRA, and the MCA energy, E_{MCA}, is determined by the force theorem,[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 6,400 special k-points in the two-dimensional Brillouin zone (BZ), the E_{MCA} was found to sufficiently suppress numerical fluctuations.

3. Results and discussion

First, we present results of the electronic structures of the Fe monolayers in the Fe/MgO and MgO/Fe/MgO. Figure 1 shows the density of states (DOS) in the MT spheres of the Fe and O atoms at the interfaces in zero field, in which results for a free-standing Fe monolayer[6] are also shown in the figure. For the free-standing Fe monolayer, the DOS around E_F arises mainly from the minority-spin state while the majority-spin state is almost fully occupied and is located from -1 to -4 eV below E_F. In

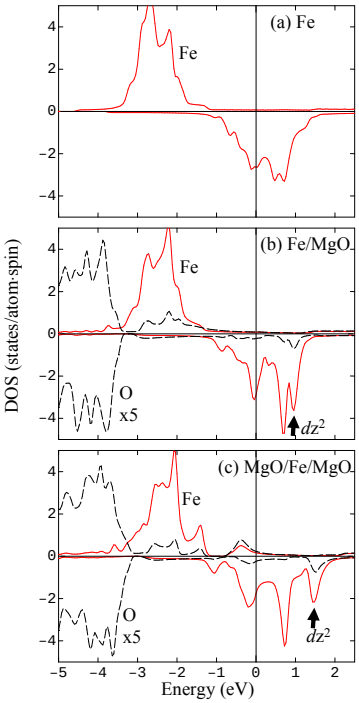

Figure 1: Density of states (DOS) in the MT spheres of Fe (solid lines) and O (dashed lines) atoms in an external electric field of zero for (a) free-standing Fe monolayer, (b) Fe/MgO and (c) MgO/Fe/MgO. The DOS of the O atom is multiplied by 5, and arrows indicate anti-bonding d_{z^2} bands of the minority-spin state.

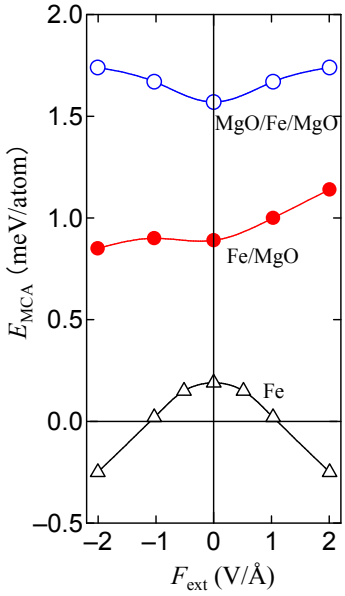

Figure 2: Calculated MCA energy, $E_{\rm MCA}$, as a function of the external electric field, $F_{\rm ext}$, for free-standing Fe monolayer (open triangles), Fe/MgO (closed circles), and MgO/Fe/MgO (open circles).

the Fe/MgO and MgO/Fe/MgO, although the overall the DOS is similar compared to that in the free-standing Fe monolayer, the major peaks of the Fe d states are found to correlate with those of the neighbor O p orbitals, where the Fe d_{z^2} bands can hybridize with the O p_z orbitals. Importantly, this hybridization pushes the anti-bonding d_{z^2} bands of the minority-spin state up above $E_{\rm F}$ as indicated by arrows in Fig. 1 (b) and (c).

Calculated $E_{\rm MCA}$ as a function of the external electric field is summarized in Fig. 2. For the free-standing Fe monolayer, the $E_{\rm MCA}$ in zero field has positive value, indicating out-of-plane MCA. When the electric field is introduced, the $E_{\rm MCA}$ significantly decreases and becomes negative as the electric field is larger than about 1 V/Å, in which the $E_{\rm MCA}$ show an even function of the electric field due to a symmetric geometry with a z-reflection. In contrast, for the Fe/MgO and MgO/Fe/MgO, the $E_{\rm MCA}$ in zero field shows large out-of-plane MCA, and the $E_{\rm MCA}$ further increases when the electric field increases. It also notes that for the Fe/MgO, an asymmetric dependence on the electric field appears due to a breaking of z-reflection of the system, but the MCA modification in negative field has rather a less-dependence on the electric field.

Within a rigid band model, furthermore, we analyze the behavior of the $E_{\rm MCA}$ with respect to a variation of the number of valence electrons. Results are shown in Fig. 3 for the free-standing Fe monolayer, Fe/MgO, and MgO/Fe/MgO. For all systems, the $E_{\rm MCA}$ of the positive values at zero field decreases when the number of valence electrons increases, and those show almost same trend since the MgO lacks electronic states at $E_{\rm F}$ which minimizes the interface hybridization. Moreover, a maximum change in the $E_{\rm MCA}$ by the electric field is achieved around N=8.4~8.5, and so a tuning of the number of valence electrons may be a key factor for designing a giant MCA modification.

To understand the MCA modification, the band structures and the MCA energy contributions, $E_{\rm MCA}(\mathbf{k})$, along high-symmetry directions are calculated, and shown in Fig. 4. For the free-standing Fe monolayer, as pointed out previously,[6] the MCA originates mainly from the SOC between the minority-spin d bands crossing $E_{\rm F}$. At around $\bar{\Gamma}$, the SOC interaction between the occupied d_{z^2} (band 1) state at -0.05 eV and the unoccupied $d_{xz,yz}$ (bands 5 and 5*) state at 0.7 eV yields a negative $E_{\rm MCA}(\mathbf{k})$; at around $\bar{\rm M}$, the SOC interaction between the occupied $d_{xz,yz}$ (bands 5 and 5*) state at -0.1 eV and the unoccupied d_{z^2} (band 1) state at 0.7 eV also makes for a negative $E_{\rm MCA}(\mathbf{k})$. In most of the BZ except around $\bar{\Gamma}$ and $\bar{\rm M}$, however, the contribution is mostly positive from the SOC between the occupied and unoccupied $d_{xz,yz}$ states, which leads to the positive $E_{\rm MCA}$ value of the out-of-plane MCA. When the electric field is introduced, although the overall the band structure and the $E_{\rm MCA}(\mathbf{k})$ are similar compared to that in zero field, a significant difference is observed; e.g., two intersecting bands 1 and 5 above $E_{\rm F}$ at $\frac{1}{3}(\bar{\Gamma}$-$\bar{\rm M})$ hybridize with each other and are split by about 0.2 eV; at around $\frac{3}{5}(\bar{\rm X}$-$\bar{\Gamma})$, the lowered band is almost pushed down below $E_{\rm F}$ by the electric field. These changes give rise to a negative contribution of the $E_{\rm MCA}(\mathbf{k})$[6] [arrows 1 and 2 in Fig. 4 (a)]. On contrary, at around $\bar{\rm M}$, a small shift of the bands 5 and 5* in lower energy gives rise to a positive contribution of the $E_{\rm MCA}(\mathbf{k})$ (arrows 3 and 4).

Now, consider the Fe/MgO and MgO/Fe/MgO. In zero field,

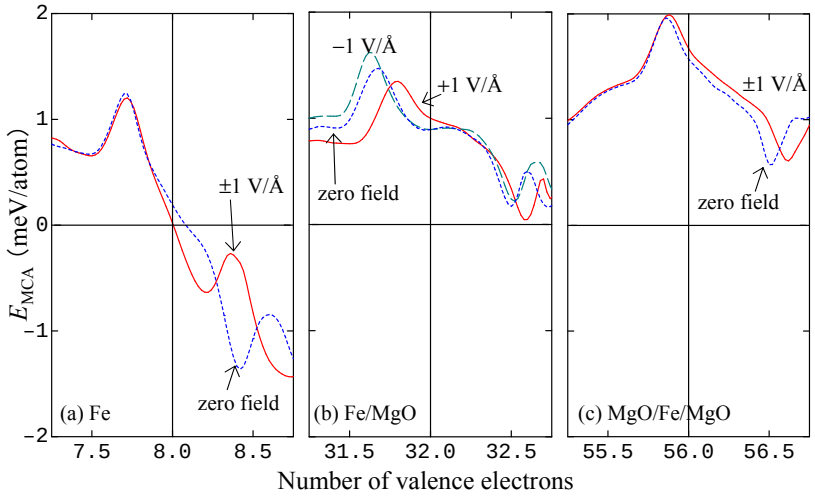

Figure 3: Calculated MCA energy, E_{MCA}, as a function of the number of valence electrons for (a) free-standing Fe monolayer, (b) Fe/MgO, and (c) Mg/Fe/MgO. Dotted, solid, and dashed lines represent results in an external electric field of zero, 1 V/Å, and -1 V/Å, respectively.

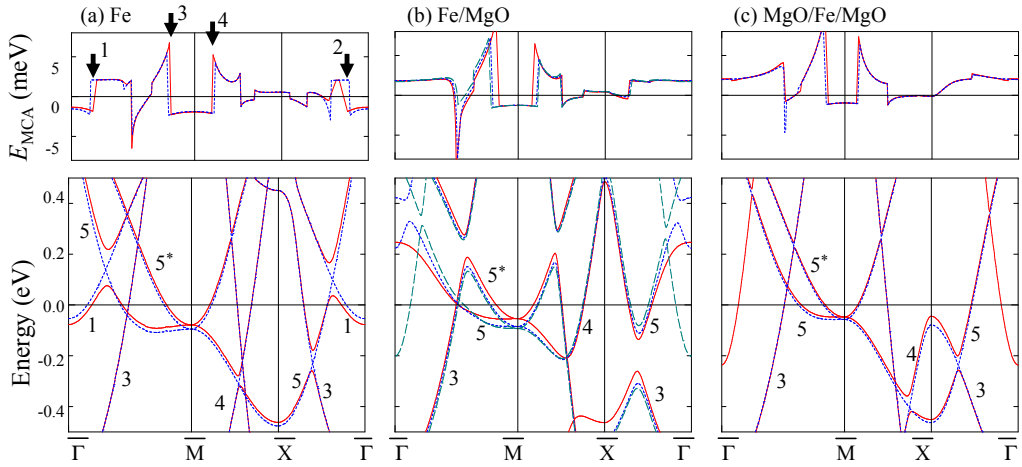

Figure 4: Calculated minority-spin band structures and MCA energy contribution, $E_{MCA}(\mathbf{k})$, along high-symmetry directions in zero field (dotted line), 1 V/Å (solid line), and -1 V/Å (dashed lines) for (a) free-standing Fe monolayer, (b) Fe/MgO, and (c) Mg/Fe/MgO. The reference energy ($E = 0$) places Fermi energy, E_F. Arrows in (a) indicate modification in the $E_{MCA}(\mathbf{k})$ by an external electric field. Bands 1, 3, and 4 stand for d_{z^2}, $d_{x^2-y^2}$, and d_{xy} states, respectively, and bands 5 and 5^* are $d_{xz,yz}$ states.

the whole feature of the *d* band structure does not alter much to that in the free-standing Fe monolayer. However, since the band 1 (d_{z^2}) is now pushed up above E_F by the Fe-O hybridization as pointed out in Fig. 1, the $E_{MCA}(\mathbf{k})$ at around $\bar{\Gamma}$ turns out to be positive. This indicates that the out-of-plane MCA in the Fe/MgO and MgO/Fe/MgO is enhanced compared to that in free-standing Fe monolayer. Moreover, since there is no longer band the intersects between bands 1 and 5 at $\frac{1}{3}(\bar{\Gamma}$-$\bar{M})$ and $\frac{3}{5}(\bar{X}$-$\bar{\Gamma})$, no modification in the $E_{MCA}(\mathbf{k})$ by the electric field appears [such as denoted by arrows 1 and 2 in Fig. 4 (a)]. Thus, the positive contribution around $\bar{M}$ by the electric field becomes dominant so as to increase the E_{MCA} when the electric field increases.

4. summary

We investigated the MCA for the Fe/MgO and MgO/Fe/MgO, and discussed the electric field-induced MCA modification. In both systems, the MCA is found to be modified by an electric field, which is caused by the change in the *d* band structure around E_F. Moreover, the position of E_F relative to the *d* band level may be a key factor for designing a large MCA modification.

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