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## First-Principles Study of Effects of Boron on Magnetic Anisotropy in MgO/CoFe(B)/X (X=Ru, Ta) Systems

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We study the effects of boron on the magnetic anisotropy in the MgO/CoFe(B)/X (X=Ru, Ta) systems using relativistic first-principles calculations. It is found that the B atoms tend to be rejected from the CoFeB layer into the X underlayer. The system with no B atoms shows perpendicular magnetic anisotropy when the Fe atoms are adjacent to the MgO layer.

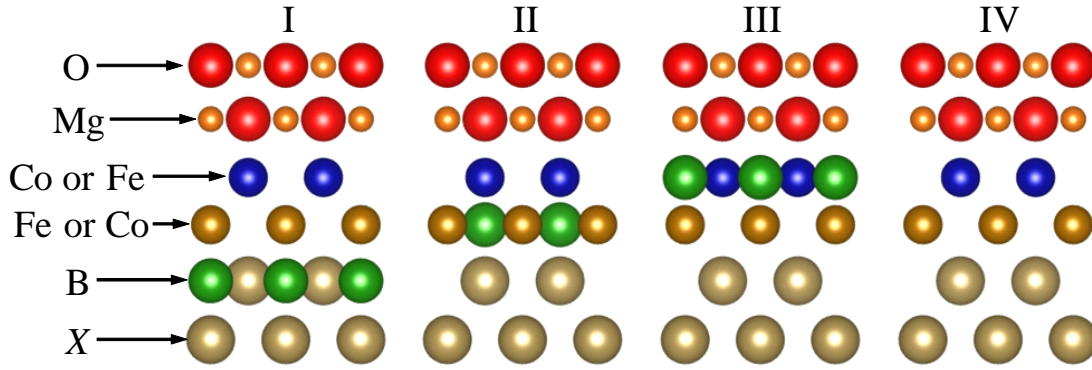
The MgO/CoFeB-based perpendicular magnetic tunnel junctions are a promising building block for future high-density memories. Ikeda et al. found that the Ta/Co<sub>20</sub>Fe<sub>60</sub>B<sub>20</sub>/MgO/Co<sub>20</sub>Fe<sub>60</sub>B<sub>20</sub>/Ta magnetic tunnel junctions show perpendicular magnetic anisotropy (PMA) when the thickness of the CoFeB layer is less than 1.5 nm, with high thermal stability at reduced dimension, low-current current-induced magnetization switching, and high tunnel magnetoresistance ratio all at the same time.<sup>1)</sup> The PMA in this system is due to the MgO/CoFeB interfacial magnetic anisotropy with negligible bulk crystalline anisotropy. Also the dependence of the PMA on B composition in MgO/(Co<sub>0.25</sub>Fe<sub>0.75</sub>)<sub>100-x</sub>B<sub>x</sub>/Ta stack structures was studied.<sup>2)</sup> The MgO/CoFeB interfacial magnetic anisotropy increases with decreasing B composition; the highest measured value of the interfacial magnetic anisotropy is 1.7 mJ/m<sup>2</sup> in the system with no B atoms, i.e., x=0. This is in agreement with the fact that the B atoms diffuse into the Ta underlayer upon annealing, resulting in the increase in the magnetization of the system.<sup>3)</sup> The reduction of B composition in the CoFeB layer as well as the removal of the B atoms from the MgO/CoFeB interface is important to the formation of a coherent MgO/CoFeB interface with a higher interfacial magnetic anisotropy. In this work, we study for the first time the effects of the B atoms on the magnetic anisotropy in the MgO/CoFe(B)/X(001) (X=Ru, Ta) systems using relativistic first-principles calculations.

We carried out all-electron calculations using the scalar relativistic full-potential linear-combination-of-atomic-orbitals (SFLCAO) method for structure optimization and the fully

relativistic full-potential linear-combination-of-atomic-orbitals (FFLCAO) method for calculating magnetic anisotropy energy (MAE);<sup>4-6)</sup> the SFLCAO and FFLCAO methods are both based on the density functional theory. We adopted the local spin density approximation (LSDA) using the Perdew-Wang parameterization of the Ceperley-Alder results as the exchange-correlation energy functional.<sup>7,8)</sup> The MAE was calculated as the total energy of the system with in-plane magnetization relative to that of the system with perpendicular magnetization, i.e.,  $\text{MAE} = E_{\text{tot}}^{[100]} - E_{\text{tot}}^{[001]}$ ; note that a positive MAE corresponds to the PMA. We do not consider the contribution of the shape magnetic anisotropy originated in the magnetic dipole-dipole interaction. The basis functions and the  $k$  points for the Brillouin-zone integration used in this work are of the same quality those used in our previous work.<sup>9)</sup>

In Fig. 1, we show the schematic diagram of the models of the MgO/CoFe(B)/X(001) systems studied in this work. The systems consist of the MgO layer, the CoFe(B) layer, and the  $X$  layer. To study the effects of the B atoms, we employ the four models denoted as I, II, III, and IV; the unit cell of each of the models I, II, and III contains 9 atoms including a single B atom while that of the model IV contains 8 atoms with no B atoms. Also, to study the effects of the difference in the stack of the Co and Fe atoms, we consider the following two types of stack, using for clarity a notation MgO||(*ferromagnetic layer*)|| $X$  to distinguish the systems with different stack of the Co and Fe atoms; one is the stack in which the Co and Fe atoms are adjacent respectively to the MgO and  $X$  layers, denoted as the MgO||CoFe(B)|| $X$  systems, while the other is the one in which the Fe and Co atoms are adjacent respectively to the MgO and  $X$  layers, denoted as the MgO||FeCo(B)|| $X$  systems. For all the models, we used the lattice constant of 2.979 Å, which is  $1/\sqrt{2}$  times the lattice constant of the bulk MgO crystal. In each model, the square-lattice layers of metal atoms are stacked alternately in a bcc-like structure to form octahedrons while the B atom in the unit cell is placed at the interstitial position inside an octahedron. The B atom is surrounded by single  $T$  ( $T = \text{Fe or Co}$ ) and five  $X$  atoms for the model I, five  $T$  and single  $X$  atoms for the model II, and single Mg and five  $T$  atoms for the model III. We do not consider the intermixing between  $T$  and  $X$  atoms at the CoFe(B)/ $X$  interface.

We now study the relative stability of the models I, II, III, and IV for each system. In Table I, we show the calculated total energies of the models I, II, and III relative to the sum of the total energy of the model IV and  $E_{\text{tot}}^{\text{XB}} - E_{\text{tot}}^{\text{X}}$ , where  $E_{\text{tot}}^{\text{XB}}$  and  $E_{\text{tot}}^{\text{X}}$  are the total energy of the square-lattice XB monolayer and that of the square-lattice  $X$  monolayer, respectively, supposing that the B atoms in the model IV are somewhere deep in the underlayer. It is found that the most stable is the model IV for each system; the models I, II, and III, each of which



**Fig. 1.** (Color online) Schematic diagram of the MgO/CoFeB/X(001) systems where  $X=\text{Ru, Ta}$ .

contains a single B atom in the unit cell, are less stable than the model IV, which contains no B atoms in the unit cell. This is in agreement with the experimental observation that the B atoms diffuse from the CoFeB layer into the Ta underlayer upon annealing.<sup>3)</sup> The next most stable is the model I for each system, also indicating that it is unfavorable for the B atoms to remain in the CoFeB layer. We thus conclude that in all the MgO/CoFeB/X systems the B atoms tend to be rejected from the CoFeB layer into the X underlayer.

**Table I.** Total energy (eV/cell) of models I, II, and III relative to corrected total energy of model IV.

System	I	II	III	IV
MgO  CoFe(B)  Ru	2.4	2.8	3.6	0
MgO  FeCo(B)  Ru	2.3	2.9	3.1	0
MgO  CoFe(B)  Ta	1.1	2.1	1.6	0
MgO  FeCo(B)  Ta	1.1	2.2	1.2	0

On the other hand, the relative stability between the models II and III depends on the underlayer, X. The model II is more stable than the model III for  $X=\text{Ru}$  while the model III is more stable than the model II for  $X=\text{Ta}$ ; the reason for this is that the interaction energy is larger for Ru and B than for Ta and B in the reaction  $X(\text{monolayer}) + \text{B}(\text{gas}) \rightarrow \text{XB}(\text{monolayer})$  by about 1 eV/XB. The result that the model III is more stable than the model II for  $X=\text{Ta}$  indicates that, as also found in the previous first-principles study,<sup>10)</sup> in the MgO/CoFeB/Ta systems with a thick CoFeB layer the B atoms can remain at the MgO/CoFeB interface, not diffusing into the Ta underlayer. This is consistent with the experimental observation that the B atoms remain at the MgO/CoFeB interface when the CoFeB layer is thicker

than 2 nm while they are rejected from the MgO/CoFeB interface into the Ta underlayer when the CoFeB layer is thinner.<sup>3)</sup>

**Table II.** MAE (meV/cell) of models I, II, III, and IV.

System	I	II	III	IV
MgO  CoFe(B)  Ru	0.9	-1.0	0.8	-0.1
MgO  FeCo(B)  Ru	0.8	-0.1	0.9	0.4
MgO  CoFe(B)  Ta	-2.3	-0.9	0.3	-0.1
MgO  FeCo(B)  Ta	-1.3	0.7	0.6	1.4

We next study the MAE of the systems. The results of calculations are shown in Table II. The measured interfacial contribution to the MAE in the MgO/Co<sub>0.25</sub>Fe<sub>0.75</sub>/Ta system is 1.7 mJ/m<sup>2</sup>,<sup>2)</sup> which corresponds to 0.85 meV/cell if we use the lattice constant of 2.979 Å. Our calculated MAE, 1.4 meV/cell, of the model IV of the MgO||FeCo(B)||Ta system is in reasonable agreement with the measured one. Also our result is in good agreement with the calculated MAE for the pure MgO/Fe interface, 2.93 erg/cm<sup>2</sup>, i.e., 1.6 meV/cell.<sup>11)</sup> On the contrary, our calculated MAE, -0.1 meV/cell, of the model IV of the MgO||CoFe(B)||Ta system is conflict with the measured one. Even the sign is opposite, i.e., negative. A possible reason for the disagreement is that in the model IV of the MgO||CoFe(B)||Ta system the transition metal atoms at the MgO interface are all Co atoms while in the MgO/Co<sub>0.25</sub>Fe<sub>0.75</sub>/Ta system, which is an Fe-rich system, they are predominantly Fe atoms. Thus, it is most likely that the interfacial PMA is due to the interaction between the Fe and O atoms at the interface of MgO-CoFeB stack structure as pointed out in the previous first-principles studies.<sup>11,12)</sup> It is worth mentioning that our calculated MAE of the model I with X=Ta is negative both for the MgO||CoFe(B)||Ta and MgO||FeCo(B)||Ta systems. This is in strong contradiction to the PMA observed in actual MgO/CoFe(B)/Ta systems. Our results indicate that the B atoms need to be rejected from the CoFeB layer into the Ta underlayer for the manifestation of a positive interfacial contribution to the MAE as found experimentally.<sup>2,3)</sup> We also believe that these results are not altered if we use a method beyond LSDA; although LSDA results in a significant error in some properties such as the orbital magnetization, the MAE calculated with LSDA for transition metal systems, e.g., CoPt, is in good agreement with experimentally measured MAE.<sup>13)</sup>

Finally, we compare the calculated magnetizations of the models IV of the MgO/CoFe(B)/X systems, i.e., MgO||CoFe||X or MgO||FeCo||X, with experimental ones. The

**Table III.** Spin and orbital magnetic moments ( $\mu_B$ ) of atoms in model IV of MgO/CoFe(B)/X system.

System	T1		T2		X	
	$M_{\text{spin}}$	$M_{\text{orb}}$	$M_{\text{spin}}$	$M_{\text{orb}}$	$M_{\text{spin}}$	$M_{\text{orb}}$
MgO  CoFe  Ru	1.57	0.07	2.75	0.07	0.36	-0.02
MgO  FeCo  Ru	2.59	0.07	1.63	0.09	0.10	0.00
MgO  CoFe  Ta	1.54	0.07	1.92	0.05	-0.28	0.05
MgO  FeCo  Ta	2.60	0.10	1.10	0.06	-0.15	0.05

magnetizations of the MgO||CoFe||Ta and MgO||FeCo||Ta systems estimated assuming the thickness of the ferromagnetic layer to be 2 (3) Å are 1800 (1200) and 1900 (1300) emu/cm<sup>3</sup>, respectively, in fair agreement with the measured magnetization of the MgO/Co<sub>0.25</sub>Fe<sub>0.75</sub>/Ta system, 1700-1800 emu/cm<sup>3</sup>.<sup>2)</sup> In Table III, we show our calculated spin and orbital magnetic moments of the atoms in the MgO||T1T2||X systems where T1 and T2 represent Fe or Co and X the underlayer atom adjacent to the ferromagnetic layer. The spin magnetic moments of the Co and Fe atoms in the MgO||CoFe||Ta system,  $M_{\text{spin}}^{\text{Co}}$  and  $M_{\text{spin}}^{\text{Fe}}$ , are  $M_{\text{spin}}^{\text{Co}}=1.54 \mu_B$  and  $M_{\text{spin}}^{\text{Fe}}=1.92 \mu_B$  while those in the MgO||FeCo||Ta system are  $M_{\text{spin}}^{\text{Fe}}=2.60 \mu_B$  and  $M_{\text{spin}}^{\text{Co}}=1.10 \mu_B$ . This shows that the magnetic moments of the T atoms are much smaller when they are adjacent to the Ta layer than adjacent to the MgO layer. Furthermore, it is found that the X atom adjacent to the ferromagnetic layer is magnetically polarized with non-negligible spin and orbital magnetizations. The magnetically polarized underlayer may play an important role in the PMA in the MgO/CoFe(B)/Ta system.<sup>14)</sup> This is an important issue to be studied theoretically in the future.

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