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Tailoring magnetic anisotropy at the ferromagnetic/ferroelectric interface

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It is predicted that magnetic anisotropy of a thin magnetic film may be affected by the polarization of a ferroelectric material. Using a Fe/BaTiO₃ bilayer as a representative model and performing first-principles calculations, we demonstrate that a reversal of the electric polarization of BaTiO₃ produces a sizable change in magnetic anisotropy energy of Fe films. Tailoring the magnetic anisotropy of a nanomagnet by an adjacent ferroelectric material may yield entirely new device concepts, such as electric-field controlled magnetic data storage. © 2008 American Institute of Physics. [DOI: 10.1063/1.2901879]

Nanomagnetic materials are playing an increasingly important role in modern technologies spanning the range from information processing and storage to spin electronics, communication devices, electrical power generation, and nanobiosensing.¹ One of the properties that make them so attractive is magnetic anisotropy, which aligns the magnetization along a preferred spatial direction. As pointed out by Van Vleck in 1930s,² the physical origin of the magnetic anisotropy is the spin-orbit coupling. The magnitude of the magnetocrystalline anisotropy of single phase bulk magnetic materials with cubic symmetry, e.g., Fe, Co, and Ni, is generally quite small due to the quenching of the orbital angular momentum by the crystal field. In thin films and multilayers, however, the reduced symmetry of surfaces and interfaces yields an additional anisotropy effect.³ As a result, the magnetocrystalline anisotropy can be enhanced by orders of magnitude in thin films,⁴ interfaces,⁵ and superlattices,⁶ overcoming the magnetostatic shape anisotropy which has a tendency to align the magnetization in the plane of a film or a multilayer. Therefore, high anisotropy materials, especially the thin ferromagnetic films, are widely utilized in modern perpendicular magnetic recording technology, which can achieve a storage density of 1 Tbits/in.² much greater than that of the traditional longitudinal magnetic recording.⁷

The large coercivity of the high anisotropy materials requires, however, high magnetic field to “write” bit information on them, thus, significantly restricting the modern recording technology for portable or ultrahigh-speed processing devices. In the last decade, various methods have been proposed to solve this problem, e.g., heat-assisted recording⁸ or the application of anisotropy-graded⁹ and exchange-coupled media.¹⁰

In this study, we explore an alternative way to tailor the anisotropy of such materials. Our strategy is based on the application of the magnetoelectric effect^{11–13} between adjacent ferromagnetic and ferroelectric films. In the previous study,¹⁴ we found that it is possible to change the magnetization at the ferromagnet/ferroelectric interface by reversing

the electric polarization of the ferroelectric. The magnetization change is not, however, the only consequence of the ferroelectric switching. Indeed, several other important properties may also be altered with polarization. Among them, electron tunneling through a ferroelectric^{15,16} or magneto-electric barrier¹⁷ and the magnetic anisotropy of the ferromagnet immersed in an electrolyte.¹⁸ Here, we demonstrate that the magnetic anisotropy of the ferromagnetic film can be altered by switching the polarization of the adjacent ferroelectric through applied bias voltage. The effect occurs due to the change of the electronic structure at the interface region, which is produced by ferroelectric displacements and mediated by interface bonding. This phenomenon is different from the magnetoelectric effect caused by magnetostriction associated with the strain induced by piezoelectricity^{19–21} and the direct effect of an electric field on surface states.²² As stated earlier, this approach has not only great potential in magnetic data storage or electric-field controlled random access memory,¹² but also the underlying physics itself is intriguing and attractive.

To illustrate the significance of this phenomenon, we construct a Fe/BaTiO₃ bilayer as our model system, where BaTiO₃ is a ferroelectric material with a large dielectric constant and Fe is a prototypical ferromagnetic metal. bcc Fe and perovskite BaTiO₃ have a very good match of the lattice constants (a mismatch is only about 1.4%) making them suitable for epitaxial growth of Fe/BaTiO₃. We build up supercells by aligning the body centered cubic iron [110] axis and the [100] axis of the BaTiO₃ [Fig. 1(a)]. The in-plane lattice constant is fixed to be the experimental value of the bulk BaTiO₃ (3.991 Å).

We calculate the magnetocrystalline anisotropy energy (MAE) as the difference between the total energies for the magnetization perpendicular (001) and parallel (100) to the interface by performing total energy density-functional calculations for two opposite polarization orientations of BaTiO₃.^{23,24} In particular calculations, we consider a monolayer Fe film on top of a relatively thick BaTiO₃ layer. The ferroelectric film in this heterostructure is about 66 Å thick, well exceeding the critical thickness for ferroelectricity (see, e.g., Ref. 25 for discussion) and demonstrating almost bulk-

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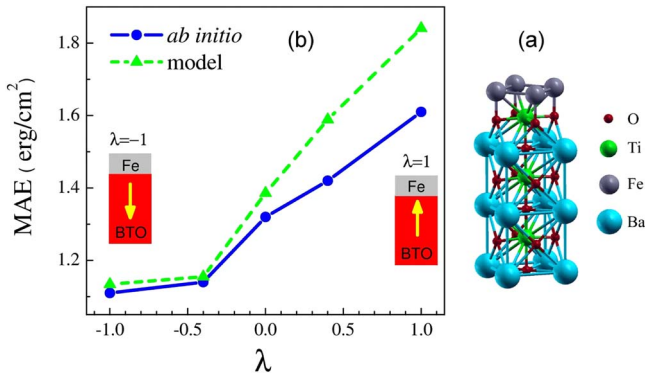


FIG. 1. (Color online) Results of calculations of MAE. (a) Unit cell of a monolayer Fe film on BaTiO₃ used in the calculation. The vacuum layer between each slice is 20 Å thick. (b) MAE as a function of a polarization scaling factor λ (b). Here, $\lambda=1$ and $\lambda=-1$ correspond to the spontaneous polarization up and down, respectively. Circles: MAE obtained using *ab initio* calculations. Triangles: MAE obtained by a model calculation based on Eq. (2). In the latter, the effective spin-orbit coupling parameter ξ for Fe is taken to be 35 meV.

like properties.¹⁴ To reduce the highly intensive computational efforts to a manageable level, we only use three unit cells of BaTiO₃ in the slice structure shown in Fig. 1(a). It is a reasonable approximation because MAE is a short-ranged intrinsic property of ferromagnetic materials.²⁶ This is supported by our test calculations which show that the MAE difference between structures with three and four unit cells of BaTiO₃ is less than 3%. The Fe monolayer is relaxed for two opposite polarization orientations in BaTiO₃ to ensure equilibrium position of Fe atoms. Our test results on the MAE of Fe, Co, Ni monolayers, and some transition CuAu(I)-type materials agree well with previous theoretical and experimental data.^{27,28}

To ensure the stability of our calculations, we check to see if MAE monotonically changes as polarization is continuously varied from up to down orientation. This is done by linearly interpolating ionic displacements between the spontaneously polarized up and down states with a scaling factor λ and repeat the MAE calculations. Figure 1(b) shows that the MAE monotonically changes as the polarization of BaTiO₃ gradually changes from the “down” to “up” state, thereby confirming the stability of our calculations.

Calculated values of the magnetocrystalline anisotropy energy K per unit area are presented in Table I. We notice that the MAE of the BaTiO₃/Fe multilayer is of the same magnitude as that of the freestanding Fe monolayer and has a tendency to align the magnetization normal to the film plane, which is reflected in the positive values of K . However, there exists apparent difference between the MAEs for the opposite ferroelectric polarization orientations. The up

TABLE I. Calculated magnetocrystalline anisotropy constant $K=(E^{(100)}-E^{(001)})/\text{area}$ for a monolayer Fe film on BaTiO₃. K is presented for two opposite polarization orientations n_p and is given in units of mJ/m² (or erg/cm²). The result for a freestanding Fe monolayer is given for comparison.

Structure	n_p	K_u	ΔK_u ($\uparrow-\downarrow$)
(BaTiO ₃) ₃ /Fe ₂	\uparrow	1.62	0.51
	\downarrow	1.11	
Fe Monolayer		1.76	

TABLE II. Calculated spin and orbital magnetic moments (M_S , M_L , in units of μ_B) of Fe and interfacial Ti atoms at the Fe/BaTiO₃ interface. Results are listed for different magnetization orientations n_M and ferroelectric polarization orientations n_p . $\Delta M_L=M_L(100)-M_L(001)$ is the anisotropy of the orbital moment. The results of monolayer Fe on the same square lattice are also listed for comparison.

n_p	Atom	n_M	M_S	M_L	ΔM_L	
\uparrow	Fe	(100)	3.033	0.073	-0.035	
		(001)	3.032	0.108		
	Ti	(100)	-0.416	0.012	0.009	
		(001)	-0.416	0.003		
	\downarrow	Fe	(100)	3.096	0.082	-0.021
			(001)	3.095	0.103	
Ti		(100)	-0.232	0.012	0.013	
		(001)	-0.232	-0.001		
Fe monolayer		(100)	3.192	0.114	-0.039	
		(001)	3.192	0.153		

(\uparrow) polarization pointing toward the interface has larger MAE than down (\downarrow) polarization pointing away from the interface and the relative MAE change, $|\Delta E_{\downarrow}-\Delta E_{\uparrow}|/\Delta E_{\uparrow}$, is as large as 46%.

An important point in anisotropic studies is the relationship between MAE and orbital angular momentum as determined by the spin-orbital Hamiltonian $H=\xi\mathbf{L}\cdot\mathbf{S}$, where ξ is the spin-orbit coupling parameter and \mathbf{L} and \mathbf{S} are orbital and spin angular momentum operators, respectively. Using second-order perturbation theory and neglecting spin-flip terms, the energy change due to the spin-orbit coupling can be written as follows:²⁹

$$\delta E \approx -\frac{\xi}{4}\hat{S}[\langle L \rangle - 2\langle L^{\uparrow} \rangle], \quad (1)$$

where $\langle L^{\uparrow} \rangle$ is the expectation value of the orbital momentum operator for the majority-spin electrons. When the majority-spin d bands are fully occupied so that their orbital moments vanish, the spin-orbit interaction energy is then proportional to $\langle L \rangle$. In this case, the MAE of a ferromagnetic film has a form³⁰

$$E^{\text{MAE}} = \delta E^{(100)} - \delta E^{(001)} = -\frac{\xi}{4}(S_x\langle L \rangle_x - S_z\langle L \rangle_z). \quad (2)$$

Similar to a freestanding Fe monolayer,³¹ we find that in our system, the majority-spin d bands of Fe are nearly fully occupied so that we can use Eq. (2) to estimate the MAE.

When the polarization changes from down to up states, the bonding between Fe and Ti atoms becomes stronger, i.e., the hybridization between the minority-spin d states of Fe and Ti atoms increases.¹⁴ This leads to the reduction of the interfacial Fe magnetic moments and increment of the induced magnetic moments on adjacent Ti atoms, as well as the changes of the orbital momentum components L_x and L_z of Fe 3d electrons. In addition, the anisotropy of the orbital moment term $\Delta M_L=M_L(100)-M_L(001)$ increases faster than the decrease of the spin moments, thus, the net effect is the increase of the MAE, as the polarization gradually changes from down to up states. Using Eq. (2) and the calculated spin and orbital magnetic moments of Fe (Table II), we obtained a series of MAEs and plotted them in Fig. 1(b) to be compared with the first-principles results. The good agreement between the band structure and model calculations shows

that the MAE change caused by the ferroelectric switching is largely due to the Fe $3d$ orbital moment anisotropy change. The difference between the *ab initio* and model calculation results in Fig. 1(b) is due to the approximations made in the model calculation which neglects spin-flip terms and contributions from Ti layer.

Tailoring magnetic anisotropy of a nanomagnet by an adjacent ferroelectric material may be used for magnetic moment switching by external electric field. We see from Table I that magnetoelectric effect alters the magnetocrystalline anisotropy of an Fe monolayer by about 50%. For thicker ferromagnetic films, the ferroelectric polarization reversal is expected to alter magnetocrystalline anisotropy of the ferromagnet/ferroelectric interface (K_{int}) in a similar manner as it does on a monolayer film. In addition to the magnetocrystalline anisotropy, the total magnetic anisotropy energy per unit area of the film involves the magnetostatic shape anisotropy energy $K_m = -2\pi M^2 t$, where M is the magnetization and t is the film thickness. For the Fe monolayer with the magnetic dipole moment of $3\mu_B$, $K_m = -0.29$ erg/cm². The shape anisotropy favors in-plane alignment of magnetization, whereas positive K_{int} favors out-of-plane alignment. Thus, with the magnetoelectric control of magnetocrystalline anisotropy and thickness dependent shape anisotropy, it may be possible to design ferromagnetic films with the anisotropy switchable between in plane and out of plane. Also, the magnetoelectric control of MAE may be helpful with the problem of writing on high coercivity perpendicular media mentioned in the introduction.

In conclusion, we have shown that the magnetoelectric effect at the Fe/BaTiO₃ interface can produce significant changes in the magnetocrystalline anisotropy of the ferromagnet. We therefore hope that our theoretical predictions will stimulate experimental studies of these and similar ferromagnetic/ferroelectric heterostructures to observe the effect of ferroelectric polarization switching on magnetic anisotropy.

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