Electrically controlled magnetism in iron thin film

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Abstract. The coupling of ferroelectric and ferromagnetic order parameters in low dimension materials has been of great interest due to the potential spintronic applications. In this report, by employing first-principles density-functional-theory calculation without including spin-orbital coupling, it is demonstrated that the magnetization of iron thin film increases in proportion to the external electric field and it prefers to be perpendicular to the film plane. This intriguing property stems from the spin-dependent screening of the electric field which leads to a spin imbalance of the excess surface charge. The result is in agreement with previous publications. We suggest that the electrically controlled magnetism might be used in advanced magnetic and electronic devices.

1. Introduction
Physics behind the coupling between magnetism and electricity in heterostructures are magnetoelectric (ME) effects which support to control magnetization of a material via applying a static electric field. In general, ME effects involve other related phenomena such as electrically controlled exchange bias, magnetoanisotropy, spin transport, etc. [1, 2] which play an important role to manipulate spin orientations and couplings [3, 4, 5]. The mechanism of the ME effects might stem from two mechanism schemes. The first one is that the external electric field displaces the ions away from their equilibrium positions, thereby altering the exchange-correlation and the electronic spin interactions which lead to the change of magnetism of materials. The second reason relates to the composite multiferroic materials, piezoelectric strain in the ferroelectric constituent in which magnetostriction is responsible for the changes in the magnetic properties of the ferromagnetic constituent [6, 7, 8].

In this report, we employed first-principles density functional theory to study the direct effect of an external electric field on the electronic and magnetic properties of iron nano-thin films. It will be shown that the magnetization of the thin film depends on the applied electric field. This originates from electronic properties. The results, therefore, stimulate the control of magnetism of nano thin film by using electrostatic electric field.

2. Crystal structure and computational details
To examine the effect of the electric field on the magnetization of Fe thin film, we carried out first-principles density functional theory calculations [9, 10] in the generalized gradient approximation for exchange and correlation, as implemented within Vienna Ab-Initio Simulation Package VASP [11, 12]. In the calculation, a Monkhorst-Pack k-point grid sampling [13] with $10 \times 10 \times 1 k$ points in the first Brillouin zone and wavefunction cut-off of 500 eV have been used.
to obtain sufficient numerical accuracy.

The crystal structure of Fe thin film used in this study is body centered cubic (BCC) with space symmetry group Im-3m which is formed by cutting the crystal along the plane (001). The configuration includes seven conventional cells separated by five equivalent-vacuum cells, as shown in FIG. 1. The in-plane lattice constant of the configuration is kept to be the optimal value obtained from the relaxation of bulk calculation. The structure is then relaxed in the absence of the electric field along \( z \) direction to obtain the lowest energy, i.e. the most stable configuration. An external electric field is then introduced by the dipole layer method such that it is perpendicular to the thin film (see Fig. 1) [14]. The magnitude of \( \vec{E} \) increases from 0 to 0.4 V/Å.

3. Results and Discussion
For the computational time, it is found that for the case of \( \vec{E} \neq 0 \), the convergence is slower drastically. This is due to the relaxation of Fe ions at lattice sites to the equilibrium positions.

![Figure 1. Crystal structure of Fe thin film placed in an electric field.](image)

![Figure 2. (Color online) Induced charges (a) and magnetic moments (b) for the external electric field strength \( E = 0, 0.1, 0.2, 0.3, \) and 0.4 V/Å.](image)
When an electric field is applied, as of the classical electromagnetic picture, the electrons in the thin film are exerted and redistributed in the entire thin film. The change in the charge along the thickness is very small. When the change of the thin-film system as a function of the z coordinate along the thickness of the thin film for various electric fields is displayed in the same figure, its variation cannot be distinguished. Therefore, we take the charge of the case $\vec{E} = 0$ as the reference charge and study its variation around this value, i.e. the induced charge due to the electric field. The dependence of this charge variation, $\Delta \text{Charge}(\vec{E}) = \text{Charge}(\vec{E} \neq 0) - \text{Charge}(\vec{E} = 0)$, is calculated as a function of z coordinate along the normal of thin-film surface. The calculated results are shown in FIG. 2(a). The horizontal axis shows the coordinate along the direction of the electric field. The vertical axis shows the induced charge. The tiny circles on the horizontal axis represent Fe ions. As can be seen, the induced charge is accumulated on the surface as expected. The accumulation of electrons distributes such that it screens the electric field from leaking into the thin film. The larger the electric field, the greater the screening electron density. Interestingly, this induced charge density is spin-dependent. To substantiate, the induced magnetic moments at each ion site have been computed and analyzed, 

$$\delta m = \mu_B \int_{V_m} (\rho_{\uparrow}(\vec{E}) + \rho_{\uparrow}(0) - \rho_{\downarrow}(\vec{E}) - \rho_{\downarrow}(0)),$$

where $V_m$ is atomic muffin-tin volume. The calculated results are represented in FIG. 2(b). As can be seen, the magnetic moment at the surface significantly increases. This also implies the change of spin polarization of the screened charges at the surface, i.e. the induced charges are spin-dependent. This feature is crucial to determine the magnetic properties of the system due to the electric field.

The calculated results of the dependence of the magnetic moments, $m$, unit in $\mu_B$, of the ions on the surface as a function of the electric field strength $E$, are represented in FIG. 3. The circles represent the results calculated directly from density functional theory at different values of $E$. The straight line presents the linear line which fits the calculated results according to the model proposed below (see Eq. (3)). It can be seen that the magnetic moment of the iron atom increases linearly with the electric field. The origin of this magnetization stems from the dependence of the induced charge density on the spin polarization, i.e. minority spin and majority spin. Induced charges that accumulate on the surface of thin film to screen the electric field from entering the thin film depend strongly on the direction of spin polarization of the electron as pointed out above. This is because the majority and minority electron spins have different Fermi wave vectors. Therefore, electrons in the crystal with different spin polarizations are coupled differently [5, 15, 16]. This linear dependence explains the straight line in FIG. 3. This prediction is consistent with previously published results in which the spin orbital coupling

Figure 3. (Color online) Magnetic moment of Fe as a function of electric field strength.
was included [5].

The results can be understood by a simple model which relates to the change in the total density of states (DOS) under the electric field. In FIG. 4, the calculated DOS for $\vec{E} = 0$ together with the change in DOS with $E=0.4$ V/Å showing in the panel have been displayed. As can be seen, the change in DOS is very small. Thus, it, $D(\epsilon)$, is considered to be fixed whereas the Fermi energy is slightly shifted under the electric field. This shift is proportional to the magnitude of $E$. The magnetic moment is determined by the difference of the majority and minority spin polarizations, i.e.

$$m = \mu_B (n_\uparrow - n_\downarrow) \quad (2)$$

where $n_{\uparrow, \downarrow} = \int_{\epsilon_{\uparrow, \downarrow}}^{\epsilon_{F}} f(\epsilon) D(\epsilon, \vec{E} \neq 0) d\epsilon$, are the occupied numbers for majority and minority spins, respectively; $\mu_B$ the Bohr magneton, $\epsilon_{\uparrow, \downarrow}$ the Fermi energies and $f$ the Fermi-Dirac distribution function. Taylor’s expansion applied to $D(\epsilon)$ for $\epsilon$ around Fermi energy and taking integral lead to

$$n_{\uparrow, \downarrow} = \int_{\epsilon_{\uparrow, \downarrow}}^{\epsilon_{F}} f(\epsilon) D(\epsilon) d\epsilon \pm \beta D_F E, \quad (3)$$

where $E = |\vec{E}|$, $D(\epsilon) \equiv D(\epsilon, \vec{E} = 0)$ and $D_F \equiv D(\epsilon = \epsilon_F, \vec{E} = 0)$, i.e. density of states at Fermi energy with absence of the electric field. It is noted that without electric field the configuration has finite magnetic moment. From Eqs. (2) and (3), the induced magnetic moments $\delta m$ are obtained

$$\delta m = 2\beta D_F E, \quad (4)$$

where $\beta$ is a proportional coefficient that determines how the Fermi energy perturbed from its value in the case with absence of electric field; and $D_F$ is the density of states at Fermi energy. This explains why the magnetic moment can be linearized shown in FIG. 3. This prediction is in accordance with other calculations published previously [3, 16].

To study the stability of spin polarization, the noncollinear calculation has been performed. The magnetic moment is oriented along the in-plane ($x$ axis) and out of plane ($z$ axis) directions.
for the total-energy ($T_E$) calculation. The calculated results are presented in FIG. 5. As can be seen, the energy difference between the two configurations is about 1 meV (per unit supercell, see FIG. 1). It implies that the magnetic moment aligning perpendicular to the thin film is more stable. Together with modulated magnetization above, this is an advantage to apply for new magnetic recording technology.

4. Conclusions
First-principles calculation on the surface magnetization of iron thin film shows that under an external electric field up to 0.4 V/Å, the induced charges accumulate on the surface of the thin film to screen the electric field. The induced-charge density is spin-dependent. It is responsible for the change of the magnetic property of the thin film of which the magnetization increases linearly with the electric-field strength. This feature can be understood that Fermi energies corresponding to different spin polarizations are shifted oppositely. It was also found that the FM state with an easy axis of magnetization perpendicular to the thin-film plane is stable. This notable change in magnetization suggests the possibility of control of magnetism of thin film only using an external electric field. The results are promising for the development of electric field-controlled magnetic data storage and spintronic technology.

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References
[1] Niranjan M K, Duan C G, Jaswal S S and Tsymbal E Y 2010 Electric field effect on magnetization at the Fe/MgO(001) interface Appl. Phys. Lett. 96 222504
[2] Ohno H, Chiba D, Matsukura F, Oniya T, Abe E, Dietl T, Ohno Y and Ohtani K 2000 Electric-field control of ferromagnetism Nature 408 9446
[3] Kanai S, Yamanouchi M, Ikeda S, Nakatan Y, Matsukura F and Ohno H 2012 Electric field-induced magnetization reversal in a perpendicular-anisotropy CoFeB-MgO magnetic tunnel junction Appl. Phys. Lett. 101 122403
[4] Tran Van Quang 2017 Electric-field modification of magnetism in a free-standing palladium ultrathin film 42th Natl. Conf. Theor. Physics, Can Tho, Vietnam
[5] Duan C G, Velev J P, Sabirianov R F, Zhu Z, Chu J, Jaswal S S and Tsymbal E Y 2008 Surface magnetoelectric effect in ferromagnetic metal films Phys. Rev. Lett. 101 137201
[6] Velev J P, Jaswal S S and Tsymbal E Y 2011 Multi-ferroic and magnetoelectric materials and interfaces Philos. Trans. R. Soc. A Math. Phys. Eng. Sci. 369 306997
[7] Duan C, Velev J P, Sabirianov R F, Mei W N, Jaswal S S and Tsymbal E Y 2015 Tailoring magnetic anisotropy at the ferromagnetic/ferroelectric interface Appl. Phys. Lett. 92 122905
[8] Quang T Van, Kim H and Miyoung K 2014 Ab initio investigation on the magnetization of Pd thin films Int. Symp. Magn. Magn. Mater., Korea
[9] Kohn W and Sham L J 1965 Self-Consistent Equations Including Exchange and Correlation Effects Phys. Rev. 140 A11338
[10] Hohenberg P and Kohn W 1964 Inhomogeneous Electron gas Phys. Rev. 136 B864-871
[11] Kresse G and Furthmüller J 1996 Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set Phys. Rev. B 54 1116986
[12] Kresse G and Joubert D 1999 From ultrasoft pseudopotentials to the projector augmented-wave method Phys. Rev. B 59 175875
[13] Monkhorst H and Pack J 1976 Special points for Brillouin zone integrations Phys. Rev. B 13 518892
[14] Neugebauer J and Scheffler M 1992 Adsorbate-substrate and adsorbate-adsorbate interactions of Na and K adlayers on Al(111) Phys. Rev. B 46 1606780
[15] Zhang S 1999 Spin-Dependent Surface Screening in Ferromagnets and Magnetic Tunnel Junctions Phys. Rev. Lett. 83 6403
[16] Chu Y H, Martin L W, Holcomb M B, Gajek M, Han S J, He Q, Balke N, Yang C H, Lee D, Hu W, Zhan Q, Yang P L, Fraile-Rodriguez A, Scholl A, Wang S X and Ramesh R 2008 Electric-field control of local ferromagnetism using a magnetoelectric multiferroic Nat. Mater. 7 47882