Spin transfer in ultrathin BiFeO$_3$ film under external electric field

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Abstract – First-principles calculations show that spin-up and spin-down carriers accumulate adjacent to the opposite surfaces of a BiFeO$_3$ (BFO) film upon applying an external bias. The spin carriers are equal in magnitude and opposite in direction, and spin-down carriers move in the direction opposite to the external electric field while spin-up ones move along the field direction. This novel spin transfer properties make the BFO film an intriguing candidate for application in spin capacitors and BFO-based multiferroic field-effect devices.

BiFeO$_3$ (BFO) has attracted much interest due to the coupling between antiferromagnetic (AFM) and ferroelectric order parameters at room temperature which drives a potential application in magnetic memory storage and emerging spintronics [1,2]. The electromechanical coupling across a morphotropic phase boundary makes it a good candidate for probe-based data storage [3]. In addition the diode effect, as well as the visible light photovoltaic effect, observed in BFO open the way for novel optoelectronics applications [4]. An electric-field controllable magnetization can be realized by producing a heterointerface composed by a BFO antiferromagnetic layer and a ferromagnetic layer, such as the BFO/La$_{0.7}$Sr$_{0.3}$MnO (LSMO) heterostructure [5,6], through the exchange-bias effect in the interface, instead of using the single-phase multiferroics which show a weak magnetoelectric coupling effect [7,8]. Surface property in BFO, specifically under external voltage, plays a crucial role in the orbital reconstruction proposed in the BFO/LSMO heterointerface, as well as in the BFO-based multifunctional devices. Moreover, we speculate that the charge transfer as well as the spin transfer also contribute to the BFO-based field-emission devices. Therefore the work function, as well as the spin density profile of BFO film under external electric field would give a transparent explanation for the transport properties in BFO-based heterostructures. As far as we know, associated works employing first-principles calculations have not been reported in the literature, although phase transition under compressive strain [9,10], as well as the Monte Carlo [11–13] and phase-field simulations under electric field (where Li and co-workers have revealed the nanoscale control of magnetoelectric coupling in strain engineered BFO films [14] and the switching of antiferromagnetic domains by mechanical stress [15]), have been carried out.

In this letter, we construct the rhombohedral $R3c$ structure (R phase) film with (001) surface in the hexagonal frame of reference [16], the bulk phase structure, and the tetragonal $P4mm$ structure (T phase) film with (001) surface, observed in the BFO film [17]. The slab model with nine and twelve atomic layers is used to simulate the ultrathin film for the P and R phase films, respectively [18–20]. The same depth is arranged for the vacuum region. The relaxation is carried out with the top and bottom two atomic layers being moved when the forces on the ions are less than 0.01 eV/Å. We adopt the local spin density approximation (LSDA) scheme and G-type AFM spin configuration [21–23] for the BFO film as we have used in our previous work [24–26]. We still include spin-orbit corrections and noncollinear magnetism. The external electric potential [27,28] has the form

$$V_{\text{ext}}(\mathbf{r}) = 4\pi m(r/r_m - 1/2), \quad 0 < r < r_m, \quad (1)$$

where $m$ is the surface dipole density of the slab, $r_m$ is the periodic length along the direction perpendicular to the surface. We set the external electric field varying from $-2$ to $2$ V/Å for the R phase film and from $-1$ to 1 V/Å for the T phase film since a field higher than that value causes large restoring forces and overshoots in the iteration process, the so-called “charge sloshing”.

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The work function (WF) is calculated as the difference between the averaged vacuum level ($\bar{\phi}$) and the Fermi energy ($E_F$) of the system. The WF for these two phases is shown in Fig. 1 as a function of the external bias. The WF of the T phase film without applying the electric field possesses the highest value compared with those under external electric field. There is no significant influence on the WF upon reversal of the external bias, and WFs exhibit a relatively lower value around 4.5 eV. On the contrary, the WF changes dramatically under an alternating external bias in the R phase. The WF experiences a sharp energy difference as the bias is reversed from $-1$ to $1$ V/Å, indicating a significant change in the surface electronic properties associated with the lattice distortions in the vicinity of the surface. We suggest this behavior is related to the structural change, as well as to the spin transfer properties under external electric field which will be discussed in the following section.

The electrostatic potential across the film averaged over the plane parallel to the surface with and without external electric field for the R phase and the T phase films is illustrated in Fig. 2 and Fig. 3, respectively. It can be seen clearly that the potential drop occurs in the vicinity of the film due to the external bias while the inside layers are less affected due to the screening effect. The potential distributes in a broad region and fluctuates dramatically near the surface with the external bias which is caused by the deviation of surface ions under the applied field. It is worth mentioning that contrary to the R phase film an internal field is observed in the T phase free-standing film without turning on the external bias. Moreover the potential distribution becomes narrower in the T phase with applying the electric field in contrast to that without applying the electric field. This behavior is attributed to the different charged layers in the T phase while the atomic layers in the R phase are neutral in nature.

We make further investigation to see the spin transfer properties and report the spin density profile for the R phase and the T phase films under electric field in Fig. 4 and Fig. 5, respectively. It is very interesting to see that spin-up electrons accumulate adjacent to the right plane along the direction of the electric field, while spin-down electrons move to the left plane opposite to the field direction in the R phase film. Moreover the magnetizations caused by the spin density in the two surfaces are equal in magnitude and opposite in direction. This partly reflects the fact that they will compensate each other and make the film remain neutral without electric field, while different spin carriers move to the opposite direction under external electric field. Although the accumulation of different spin carriers is again found in the T phase film, the magnetization adjacent to the surface caused by the spin carriers...
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Fig. 4: (Colour on-line) Distribution of magnetization across the R phase ultrathin BFO film averaged over the plane parallel to the film under electric field $E = 2$ V/Å.

Fig. 5: (Colour on-line) Distribution of magnetization across the T phase ultrathin BFO film averaged over the plane parallel to the film under electric field $E = 1$ V/Å.

is different in magnitude. The spin density doubles in the vicinity of the left surface in comparison with that close to the right surface, implying that the different spin carriers cannot be neutralized without applying the electric field. This behavior is attributed partly to the charged atomic layers in the T phase film, leading to spin polarization across the film, and this further implies that the polarization under electric field is also coupled to the spin transfer.

The spin carriers accumulating behavior applying external voltage is illustrated clearly in fig. 6. The amount of different spin carriers aggregating in the vicinity of the opposite surfaces is equal in the R phase film while this is not the case for the T phase film due to the charged internal layers, and this reflects the fact that the spin transfer is closely related to the lattice distortions as well as to the domain switching. The accumulation of spin-up and spin-down electrons adjacent to the opposite surfaces leads to spin density and magnetization near the surface. These meaningful and fantastic spin transfer properties observed in the R phase BFO film can be used in two applications: i) spin capacitors which accumulate the spin carriers instead of the opposite charges as in traditional capacitors [29]; and ii) BFO-based heterointerfaces which could tune the magnetization of the ferromagnetic layer by the external electric field through the exchange-bias effect [30,31]. The minimum electric field which is able to induce the change in charge density and spin transfer is about 0.001 V/Å [32].

In summary, ab initio calculations show that the R phase BFO film exhibits a significant change in the work function upon reversal of the external bias while this is not observed in the T phase film. This is related to the ionic structure, as well as to the spin density adjacent to the surface. Atomic layers in the T phase film are charged, leading to an internal potential and an unbalanced spin density near the opposite surfaces. Spin carriers accumulating behavior in the R phase film presents promising possibilities for spintronic applications.

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REFERENCES

[1] Lebeugle D., Colson C., Forget A., Viret M., Bataille A. M. and Gukasov A., Phys. Rev. Lett., 100 (2008) 227602.
[2] Fiebig M., J. Phys. D: Appl. Phys., 38 (2005) R123.
[3] Zeches R. J., Rosselli M. D., Zhang J. X., Hatt A. J., He Q., Yang C.-H., Kumar A., Wang C. H., Melville A., Adamo C., Sheng G., Chu Y.-H., Ihlefeld F. J., Erni R., Ederer C., Gopalan V., Chen L. Q., Schlam D. G., Spaldin N. A., Martin L. W. and Ramesh R., Science, 326 (2009) 977.
[4] Choi T., Lee S., Choi Y. J., Kiryukhin V. and Cheong S.-W., Science, 324 (2009) 63.
[5] Wu S. M., Cybart Shane A., Yu P., Rosselli M. D., Zhang J. X., Ramesh R. and Dynes R. C., Nat. Mater., 9 (2010) 756.
[6] Borisevich A. Y., Chang H. J., Huijben M., Oxley M. P., Okamoto S., Niranjan M. K., Burton J. D., Tsymbal E. Y., Chu Y. H., Yu P., Ramesh R., Kalinin S. V. and Pennycook S. J., Phys. Rev. Lett., 105 (2010) 087204.
[7] Ramesh R. and Spaldin N. A., Nat. Mater., 6 (2007) 21.
[8] Cheong S. W. and Mostovoy M., Nat. Mater., 6 (2007) 13.
[9] Prosandeev S., Kornev Igor A. and Bellaiche L., Phys. Rev. Lett., 107 (2011) 117602.
[10] Dupe B., Prosandeev S., Geneste G., Dkhil B. and Bellaiche L., Phys. Rev. Lett., 106 (2011) 237601.
[11] Lisenkov S., Rahmedov D. and Bellaiche L., Phys. Rev. Lett., 103 (2009) 047204.
[12] Yang Y., Li L. J. and Li J. Y., Appl. Phys. Lett., 98 (2011) 182905.
[13] Albrecht D., Lisenkov S., Ren Wei, Rahmedov D., Kornev Igor A. and Bellaiche L., Phys. Rev. B, 81 (2010) 140401(R).
[14] Liu Y. Y., Vasudev R. K., Pan K., Xie S. H., Liang W.-I., Kumar A., Jesse S., Chen Y.-C., Chu Y.-H., Nagarajan V., Kalinin S. V. and Li J. Y., Nanoscale, 4 (2012) 3175.
[15] Li L. J., Li J. Y., Shi Y. C. and Yen J. H., Appl. Phys. Lett., 93 (2008) 192506.
[16] Catalan G. and Scott J. F., Adv. Mater., 21 (2009) 2463.
[17] Wang J., Neaton J. B., Zheng H., Nagarajan V., Ogale S. B., Liu B., Viehland D., Vaithyanathan V., Schlam D. G., Waghmare U. V., Spaldin N. A., Rabe K. M., Wuttig M. and Ramesh R., Science, 299 (2003) 1719.
[18] Neugebauer J. and Scheffler M., Phys. Rev. B, 46 (1992) 16067.
[19] Feibelman J. P., Phys. Rev. B, 64 (2001) 125403.
[20] Che J. G. and Chan C. T., Phys. Rev. B, 67 (2003) 125411.
[21] Smith R. T., Achenbash G. D., Gerson R. and James W. J., J. Appl. Phys., 39 (1968) 70.
[22] Plumer M. L. and Walker M. B., J. Phys. C: Solid State Phys., 6 (1982) 68.
[23] Zhang J. T., Lu X. M., Zhou J., Sun H., Su J., Ju C. C., Huang F. Z. and Zhi J. S., Appl. Phys. Lett., 100 (2012) 242413.
[24] Feng H.-J., Phys. Rev. B, 46 (1992) 205426.
[25] Meyer B. and Vanderbilt David, Phys. Rev. B, 86 (2012) 014401.
[26] Bengtsson L., Phys. Rev. B, 69 (2004) 104404(R).
[27] Giri S., Patra M. and Majumdar S., J. Phys.: Condens. Matter, 23 (2011) 073201.
[28] Liu Y., Yao Y., Dong S., Yang S. and Li X., Phys. Rev. B, 86 (2012) 075113.
[29] Feng H.-J., Physica B, 412 (2013) 41.