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Large magnetoelectric coupling in ferromagnetic/ferroelectric superlattices with asymmetric interfaces

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ABSTRACT

In this Letter, we investigate an attractive approach providing large and cumulative magnetoelectric coupling in ferromagnetic/ferroelectric superlattice by inserting a monolayer of different metal (ferromagnetic or normal metal) at one interface. Using first-principles calculations on $\text{FeM}/\text{PbTiO}_3$ ($\text{M}=\text{Ni}$, Pt and Ag) as a model system, the recorded high magnetoelectric coefficients are obtained in one unit cell with the insertion of Ni and Pt monolayer. A carefully analysis reveals that the large magnetoelectric effects are determined by combinations of several different mechanisms including interface bonding, spin-dependent screening, and distinct type of magnetic interactions.

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1. Introduction

The field of composite multiferroics has been developing vigorously due to the fact that the existing single-phase multiferroics have rather limited applications either because of their low Curie temperature or weak magnetoelectric (ME) coupling [1–4]. Recent advances in thin-film deposition techniques have allowed manufacturing epitaxial heterostructures with atomically abrupt interfaces. Current efforts focus on the so-called “interfacial magnetoelectricity” in which the ME coupling arises as an extrinsic effect at a ferromagnetic (FM)/ferroelectric (FE) interface, which can be fabricated in the form of multilayers of alternating FM and FE layers or vertically aligned columnar nanostructures [5–8].

Several types of coupling mechanisms giving rise to the ME effects at the FM/FE interfaces have been reported. Besides the elastic interaction mechanism, of which the ME coupling is exclusively mediated by strain across the interface between the piezoelectric and magnetostrictive (piezomagnetic) materials [9], the interface ME effects can be caused by pure electronic mechanisms, such as spin-dependent screening and interface bonding [4]. These two ME effects were first predicted from first-principles calculations [10–13] and then verified by experimental observations [14,15]. More recently, our work predicts a novel kind of ME effect at the Ni/ATiO_3 ($\text{A}=\text{Ba}$ and Pb) interfaces [16]. This ME effect is determined by the change of magnetic moments on Ni atoms near the interface, instead of the induced moments on the interfacial Ti atoms in the Fe/BaTiO_3 system [10], which is also distinguished from the spin-dependent screening mechanism [12]. The underlying physics is the $\text{pd}\sigma$ -type magnetic

interactions between Ni $3d$ and O $2p$ spins. Furthermore, in the Fe/PbTiO_3 system, we have demonstrated that the interface ME effect is determined by the combination of different ME coupling mechanisms [17], i.e., conjunction of the interface bonding and the spin-dependent screening. The combinations of different ME coupling mechanisms enhances the ME coefficient at the Fe/PbTiO_3 interface to be several times larger in magnitude than that of individual ME coupling mechanism.

Up to now, most of the theoretical investigations have concentrated on the FM/FE superlattices with identical termination interfaces. In these symmetric structures, the difference of magnetic moments between the two interfaces in one unit cell reflects the change which occurs at *one* interface if the polarization of FE reverses, while the overall induced magnetization will be canceled in a superlattice structure. On the other hand, in the asymmetric FM/FE structures with different interfaces, due to the broken inversion symmetry between the two interfaces in one unit cell, there would be a net magnetization in each unit cell and it can cumulate in a superlattice to achieve a large macroscopic magnetization. According to this guideline, a tri-component superlattice composed of a FM metal, FE, and normal metal (NM) was proposed to realize robust ME effect [18]. Using $\text{Fe}/\text{ATiO}_3/\text{Pt}$ ($\text{A}=\text{Ba}$ and Pb) superlattice as a model system, it is shown that a net and cumulative interfacial magnetization is induced in the FM metal near the FM/FE interface [19]. In this Letter, we put forward an alternative and attractive approach to realize the asymmetric structure in the FM/FE superlattice by inserting a monolayer of different metal (FM or NM) at one interface. We will demonstrate that our strategy is more efficient in enhancing the ME effects and will have potential applications in the field of multiferroics.

To examine the effect of inserting a monolayer metal at the interface, we use Fe/PbTiO_3 as a prototypical system, of which one interfacial Fe monolayer is replaced by another kind of metal M

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($M = \text{Ni, Pt and Ag}$). The polarization of the bulk $P4mm$ PbTiO_3 is 0.90 C/m^2 as calculated using Berry's phase method [20], while the magnetic moments of the bulk Fe is $2.13 \mu_B$ per atom. We terminate the PbTiO_3 with TiO_2 planes and place the metal atoms on top of oxygen. The in-plane lattice constant is fixed to the theoretical value calculated for the tetragonal bulk PbTiO_3 (3.890 \AA), which is in good agreement with the experimental lattice constant (3.895 \AA). The tetragonal supercell used in our study can be describe as $(\text{Fe}_2)_8-(\text{M}_2)_1-(\text{TiO}_2)_1-(\text{PbO}-\text{TiO}_2)_6$ (the subscripts indicate the number of atomic layers) which is illustrated in Fig. 1(a).

We use density functional theory (DFT) within the Perdew–Burke–Ernzerhof form [21] of generalized gradient approximation (GGA) as implemented in the VASP code and projector-augmented-wave pseudopotentials [22–24]. The energy cutoff of 500 eV is used for the plane-wave expansion. For atomic relaxations, integrals over the Brillouin-zone were replaced by sums on a $6 \times 6 \times 1$ Monkhorst-Pack k -point mesh [25] and the ions were relaxed until the Hellmann–Feynman forces are less than 20 meV/\AA . For subsequent electronic structure calculations, the grid was increased to $12 \times 12 \times 2$. The spin-orbit interaction is not included in our calculations.

To investigate the influence of M monolayer on the FE polarization, we show in Fig. 1(b–d) the displacements of Ti and Pb with respect to O atoms at each PbTiO_3 atomic plane as the FE polarization pointing to rightward. The inhomogeneous displacement across the PbTiO_3 film reflects the inhomogeneous polarization in the FE slab, which is the consequence of the asymmetry produced by the two interfaces [10,16,17,26]. In order to elucidate the influence of M monolayer on the relative displacements as the polarization direction reverses, both the displacements in case of M inserted at the left ($l = -4$) interface and that of the M layer inserted at right ($l = 4$) interface are illustrated. First, the asymmetric variations of the relative displacements are various with different M atoms. In contrast to the tiny change of the relative

displacements as the Ni monolayer inserted, the insertion of Pt and Ag monolayers induces more significant modifications in the relative displacements of Ti and Pb with respect to O atoms. Displacements of the two interfacial Ti atoms have large asymmetric change as the Pt monolayer inserted, while the insertion of Ag monolayer induces remarkable variations in the relative displacements of leftward Pb and rightward Ti atoms. Second, it is evident from Fig. 1(b–d) that the FE polarization decreases gradually as the M monolayer changes from Ni through Pt to Ag. To quantitatively estimate the spontaneous polarization, we construct the paraelectric (PE) state by averaging the atomic z -positions of the two opposite polarization states [27] as the M monolayer inserted at the same interface. Using the ionic displacements relative to the positions of the PE state and the Born effective charges obtained in bulk $P4mm$ PbTiO_3 , the average polarizations evaluated for $M = \text{Ni, Pt, and Ag}$ monolayer are $0.75, 0.67,$ and 0.49 C/m^2 , respectively. For the unmodified Fe/PbTiO_3 superlattice, our previous calculation [17] has shown that the average polarization is 0.77 C/m^2 . It is clear that the insertion of Ni and Pt monolayer has small influence on the FE polarization, while inserting a monolayer of Ag at the Fe/PbTiO_3 interface reduces the FE polarization remarkably. Obviously, the asymmetric changes of FE displacements and spontaneous polarizations are determined by the distinct electronic structure and interface bonding at the two interfaces as the M monolayer inserted.

The layer-resolved density of states (DOS) projected on d orbitals of the interface metal (Fe and M) atoms are shown in Fig. 2. We can see that the interface electronic structures are remarkably distinct from their corresponding bulk states. In case of the left interfacial Fe, the DOS are very similar for the three kinds of M monolayer inserted at the right interface. The aspect should be stressed is the significant enhanced weight of the Fe $3d$ minority-spin states near the Fermi energy. It varies with reversal of the FE polarization due to the spin-dependent screening and

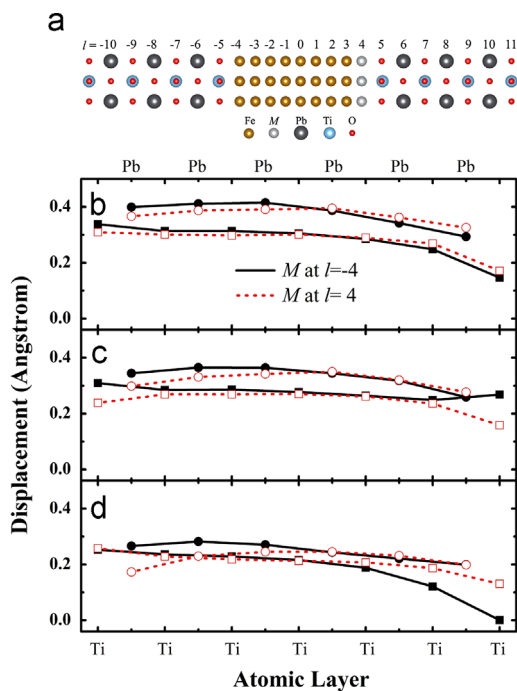


Fig. 1. (a) Atomic structure of the FeM/PbTiO_3 ($M = \text{Ni, Pt, and Ag}$) supercell. The index l denotes the l th atomic layer away from the central metal layer ($l = 0$), (b and d) correspond to displacements of cations (Ti and Pb) relative to oxygen in PbTiO_3 with the polarization direction pointing to right for $M = \text{Ni, Pt, and Ag}$, respectively. The square (circle) symbols mark the Ti–O₂ (Pb–O) displacements, while the red open (black solid) symbols indicate the displacements as the M layer located at $l = 4$ (-4). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

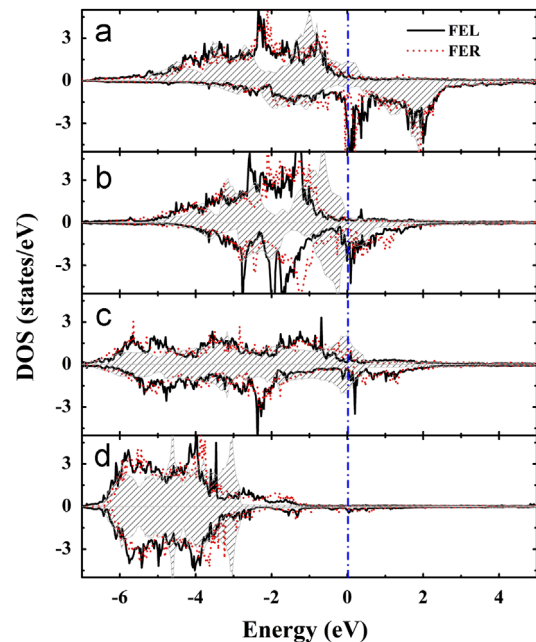


Fig. 2. Layer-projected DOS for d orbitals of the interfacial metal atoms in the two distinct ferroelectric states of PbTiO_3 . (a) Fe monolayer at the left interface ($l = -4$) in FeNi/PbTiO_3 . (b, c, and d) correspond to the Ni, Pt, and Ag monolayer at the right interface ($l = 4$), respectively. Black solid and red dotted curves refer to the FE polarization directions pointing to left (FEL) and right (FER), respectively. Gray shaded plots are their corresponding bulk DOS. The vertical dash-dotted line indicates the Fermi energy. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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