**Ab initio study of the intrinsic exchange bias at the SrRuO$_3$/SrMnO$_3$ interface**

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In a recent publication [S. Dong et al., Phys. Rev. Lett. 103, 127201 (2009)], two (related) mechanisms were proposed to understand the intrinsic exchange bias present in oxides heterostructures involving G-type antiferromagnetic perovskites. The first mechanism is driven by the Dzyaloshinskii-Moriya interaction, which is a spin-orbit coupling effect. The second is induced by the ferroelectric polarization, and it is only active in heterostructures involving multiferroics. Using the SrRuO$_3$/SrMnO$_3$ superlattice as a model system, density-functional calculations are here performed to verify the two proposals. This proof-of-principle calculation provides convincing evidence that qualitatively supports both proposals.

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1. INTRODUCTION

The exchange bias (EB) effect is known to be present in a wide range of magnetic composites. This effect has been extensively used in a variety of magnetic storage and sensor devices.$^{1-6}$ In principle, the physical origin of the EB is the magnetic coupling at the interface between antiferromagnetic (AFM) and ferromagnetic (FM) (or ferrimagnetic) materials. However, a more fundamental microscopic theoretical understanding of the subtle EB effect has not been developed in spite of the fact that the EB phenomenon has been known for more than half a century and used in industry for decades. In many textbooks of magnetism, the EB is described as induced by spin-pinning effects at the FM/AFM interface. An uncompensated AFM interface, which means that there is a net magnetic moment at the interface on the AFM side, is often invoked to illustrate how the pinning may work, as sketched in Fig. 1(a). These uncompensated magnetic moments provide a bias field to the neighboring FM moments via the exchange coupling across the interface. This ideal scenario is very clear with regards to the possible cause of the effect, but it appears incomplete since it actually fails in several real cases.$^7$ $^9$

To understand the deficiencies of the uncompensated AFM scenario, consider the case of AFM interfaces that are optimally uncompensated, as in Fig. 1(a). An example is provided by the (111) surface of G-type AFM materials. In these cases, a large EB would be expected. However, such an effect has not been observed in real materials with these characteristics.$^7$ In contrast, several compensated AFM interfaces, as illustrated in Figs. 1(b) and 1(c), do display an EB effect, that may be even larger than for the "uncompensated" interface of the same materials.$^7$ To solve these puzzles, several possible mechanisms have been proposed in the past decades. Extrinsic factors are often considered, such as interface roughness,$^8$ spin canting near the interface,$^{10}$ frozen interfacial and domain pinning,$^{11,12}$ and others. For more details, the readers are referred to the theoretical review by Kiwi that summarizes several previous models.$^7$ In most of these models, a small "frozen" uncompensation of the AFM moments near the interface remains the common ingredient for the explanation of the bias field, despite the different origins of this uncompensation.

In recent years, considerable attention and research have been devoted to the study of the exchange bias in magnetic oxides heterostructures.$^{13,14}$ In particular, when a multiferroic material participates as one of the two components at the interface, the exchange bias can then be tuned by electric fields,$^{15-21}$ an effect that is difficult to understand with only purely magnetic arguments. Motivated by these experimental studies, some of the present authors have recently proposed two (related) mechanisms to understand the EB effects in magnetic oxides heterostructures.$^{22}$ These mechanisms are based on the Dzyaloshinskii-Moriya (DM) interaction and on the ferroelectric (FE) polarization, with the later only active in the heterostructures that involve multiferroics. These two mechanisms are quite different from previous models used to address the EB effect, since they do not rely on the existence of uncompensated AFM moments anymore.

The DM interaction was first proposed over half a century ago (two years after the observation of the EB effect) to explain the existence of weak ferromagnetism in some AFM materials.$^{23,24}$ The DM interaction originates in the spin-orbit coupling (SOC) effect caused by relativistic corrections to superexchange. This DM interaction can be written as $\vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$, where $\vec{S}_i$ and $\vec{S}_j$ are nearest-neighbor spins. The vector $\vec{D}_{ij}$ obeys the asymmetric rule $\vec{D}_{ij} = - \vec{D}_{ji}$. In transitional metal oxides, $\vec{D}_{ij}$ is determined by the bending of the $M$-O-$M'$ bond (where $M'$s denote transition metal ions and O the oxygen ion). From symmetry considerations,
are noncollinear, namely, the magnetic easy axes of the FM and AFM moments are collinear. In (c), the FM and AFM magnetic moments state. (b) and (c) Fully compensated AFM interfaces, as they occur (001) surface of A-type AFM state or (111) surface of G-type AFM respectively. Local magnetic moments are marked as arrows. (a) A

FIG. 1. (Color online) Sketch of possible FM-AFM interfaces. The upper/lower (blue/red) layers denote FM/AFM components, respectively. Local magnetic moments are marked as arrows. (a) A fully uncompensated AFM interface, as it occurs for example in an (001) surface of A-type AFM state or (111) surface of G-type AFM state. (b) and (c) Fully compensated AFM interfaces, as they occur in a G-type AFM state. In (a) and (b), the FM and AFM magnetic moments are collinear. In (c), the FM and AFM magnetic moments are noncollinear, namely, the magnetic easy axes of the FM and AFM materials are different.

the direction of $\vec{D}_{ij}$ should be perpendicular to the plane spanned by the bended $M_i$-O-$M_j$ bond. The intensity of $|\vec{D}_{ij}|$ is proportional to the oxygen displacements from the middle point of $M_i$-$M_j$. Thus, if the $M_i$-O-$M_j$ bond is straight, namely, a 180° angle bond, then $\vec{D}_{ij}$ becomes zero.

According to the formula for the DM interaction, the presence of a noncollinear spin pair is a key ingredient. However, these spin arrangements are not very common as ground states in bulk materials since most spin states are typically governed by strong superexchange interactions that often favor collinear states. However, it is quite natural to have noncollinear spin pairs across the interfaces in heterostructures, due to the different magnetic easy axes of the two components.

In bulks of several perovskites, there are intrinsic and collective tiltings and rotations of the oxygen octahedra, which are often generically called the “GdFeO$_3$-type” distortions. In several oxide thin films and heterostructures, these octahedral tiltings and rotations are still present, although the distortion modes may change with respect to those that exist in the bulk limit due to the lattice mismatch and substrate strain or stress. A crucial aspect of our arguments is that due to these collective octahedral distortions, the $M_i$-O-$M_j$ bonds are bent in a staggered manner, giving rise to the staggered arrangement of $\vec{D}$ vectors shown in Fig. 2(b). According to our recent model investigations, a uniform bias field emerges from the combination of the staggered $\vec{D}$ vectors and the staggered G-type AFM spin order.

Furthermore, if one component has an (in-plane) FE polarization which induces a uniform displacement between cations and anions, the angles of the $M_i$-O-$M_j$ bonds become staggered, as sketched in Figs. 2(b) and 2(c). As a result, the intensities of the exchange couplings are modulated, which also induces a uniform bias field when in the presence of the G-type AFM spin order.

Therefore these two mechanisms rely on the crystal distortions, the spin-orbit coupling, and the spin-lattice coupling, all of which are intrinsic in oxide heterostructures. However, from the experimental information available up to date, it remains difficult to reach a definitive conclusion about the validity of the proposed mechanisms, particularly considering the complexity of real oxide heterostructures. Thus it is very important to analyze further our proposal by considering other theoretical perspectives to judge how robust our ideas actually are.

II. MODEL SYSTEM AND METHOD

To verify the two proposals for the EB effect discussed above, here an approach complementary to that followed in the original publication will be pursued, namely, the ab initio density-functional theory (DFT) simulation will be employed. It should be remarked that although the DFT methods are becoming more and more powerful in several branches of condensed matter physics and materials science, it is rare to find DFT applications in the field of exchange bias. The reason is that in most previous scenarios proposed to understand the EB effect, large length scales are involved, and the phenomenon is described as induced by magnetic domains and metastable states. These subtle effects are believed to be beyond the ability of current DFT techniques. However, the two mechanisms proposed in Ref. are intrinsic and they are supposed to occur at the atomic scale, which allows to investigate their existence employing a DFT methodology.

In this publication, the model system is chosen to be a SrRuO$_3$/SrMnO$_3$ superlattice. This system is ideal for our purposes for several reasons: (1) it has been known from experiments that the Mn spins lie in-plane and form a G-type AFM order, while the Ru spins point out-of-plane and form a FM order. (2) All layers (SrO, RuO$_2$, MnO$_2$) are nonpolar, which excludes the possible transfer of charge due to the polar catastrophe. Since it is expected that the Fermi energy of metallic SrRuO$_3$ lies within the gap band of insulating SrMnO$_3$, then charge transfer due to different work functions is not expected to be of relevance either. (3) The A-site cations are both Sr$^{2+}$, which provides a unique interfacial termination, and the EB has indeed been observed experimentally in these superlattices.

The DFT calculations were here performed based on the projected augmented wave (PAW) pseudopotentials using the Vienna ab initio simulation package (VASP). The valence states include 4$s$$4p$$5$s$, 3$p$$4s$$3d$, 4$p$$5$s$ and 2$s$$2p$ for Sr, Mn, Ru, and O, respectively. The electron-electron interaction is described using the generalized gradient approximation (GGA) method.

A superlattice structure (SrRuO$_3$)$_2$-(SrMnO$_3$)$_2$ (totally 40 atoms) is stacked along the [001] axis, as shown in Fig. 2(a). The initial atomic positions are set as in the bulk arrangement for SrRuO$_3$ and the in-plane crystal lattice constants are set as 3.905 Å to match the widely used SrTiO$_3$ substrate. Then, the out-of-plane lattice constant is relaxed. And the atomic positions of the superlattices are also fully optimized as the Hellman-Feynman forces are converged to less than 0.01 eV/Å. This optimization and electronic self-consistency are performed under a nonmagnetic state.

After the optimization of the atomic positions, different magnetic profiles are applied into the system. The initial magnetic moments are ±3 for Mn (G-type AFM) and 2 for Ru (FM). The options in the codes to include the noncollinear
O-Ru bond angles of oxygen octahedra are obvious to the eye, giving rise to bended pseudocubic comparison. In-plane lattice constants \( b \) are directed by \( \vec{F} \) polarization \( \vec{E} \) nearest-neighbor pairs are staggered. The then the bond angles become modulated: \( (\text{drawn by VESTA}) \).\(^{31}\) The space group is \( Pmc2_1 \) (orthorhombic).\(^{90}\) All calculations have been carried out for Ru-O-Ru; the out-of-plane bond angles are 173° for Mn-O-Mn and 156° for Ru-O-Ru; the out-of-plane bond angles are 173.8° for Mn-O-Mn, 167.7° for Ru-O-Ru, and 170.4° for Ru-O-Mn. It is clear that the bonds in SrMnO\(_3\) are very close to straight, while the bonds in SrRuO\(_3\) are more bending, implying here that this bending of the bonds is due to SrRuO\(_3\), in agreement with the arguments presented above. Here, the tilting angle of the octahedra in SrRuO\(_3\) (180° - 167.7 = 12.3°) is close to, but slightly different from, the previous DFT result (10.5°) on a pure FM SrRuO\(_3\) film on SrTiO\(_3\).\(^{35}\) Considering the differences between the studied systems and the differences in the details of the calculations, this observed nice agreement between our present results and the previous ones suggests that our approach is reasonable.

Let us consider first the collinear spin state without the SOC. The output local magnetic moments are 2.56 ± 0.05 \( \mu_B \) for Mn and 1.42 ± 0.01 \( \mu_B \) for Ru, which are close to the experimental data and to previous DFT calculations (with LSDA or LSDA+U) in the corresponding bulk materials.\(^{46–48}\) These magnetic moments suggest the robust stability of the G-type AFM + FM spin configurations in the (SrRuO\(_3\))\(_2\)-(SrMnO\(_3\))\(_2\) superlattice.\(^{49}\) The weak disproportions (±0.05 for Mn and ±0.01 for Ru) arise from the different possible bonds [Mn(↑)-O-Ru(↑)] versus Mn(↓)-O-Ru(↑), where ↑ and ↓ denote the directions of the spins.

The energy difference (\( \Delta E \)) is only 0.058 meV, which is already far below the expected precision of VASP, since even the default accuracy for electronic minimization is 0.1 meV in VASP. Thus it is clear that these two states before and after the Ru’s spin flipping must be degenerate, or in other words no bias effect is present. This degeneracy is certainly to be expected considering the presence of independent inversion symmetries in the spin space and crystal space.

### III. RESULTS AND DISCUSSION

The optimized lattice is shown in Fig. 2(a). When in the bulk, SrMnO\(_3\) is cubic,\(^{43}\) while SrRuO\(_3\) is orthorhombic.\(^{44}\) Thus the oxygen octahedral distortion in the SrRuO\(_3\)-SrMnO\(_3\) superlattice (on SrTiO\(_3\)) is inherited from SrRuO\(_3\). Here all Mn-O-Mn, Mn-O-Ru, and Ru-O-Ru bonds are bending: the in-plane bond angles are 176.3° for Mn-O-Mn and 156.2° for Ru-O-Ru; the out-of-plane bond angles are 173.8° for Mn-O-Mn, 167.7° for Ru-O-Ru, and 170.4° for Ru-O-Mn. It is clear that the bonds in SrMnO\(_3\) are very close to straight, while the bonds in SrRuO\(_3\) are more bending, implying here that this bending of the bonds is due to SrRuO\(_3\), in agreement with the arguments presented above. Here, the tilting angle of the octahedra in SrRuO\(_3\) (180° - 167.7 = 12.3°) is close to, but slightly different from, the previous DFT result (10.5°) on a pure FM SrRuO\(_3\) film on SrTiO\(_3\).\(^{35}\) Considering the differences between the studied systems and the differences in the details of the calculations, this observed nice agreement between our present results and the previous ones suggests that our approach is reasonable.

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#### A. DM-driven EB

To verify the previously proposed DM-driven EB mechanism, the noncollinear spins are now considered. The initial spins of Ru (\( \vec{S}_{Ru} \)) point out-of-plane while the spins of Mn (\( \vec{S}_{Mn} \)) are in-plane, as observed in the real heterostructures.\(^{32–36}\) Furthermore, the Mn spins are rotated within the \( x\)-\( y \) plane for comparison.

At first, the SOC is kept “disabled” in the calculations (thus no DM interaction is possible in principle). As listed in Table I, the \( \Delta E \)'s are 0.231 and 0.264 meV when \( \vec{S}_{Mn} \)’s are along the \( x \) and \( y \) axes, 0.0016 and –0.023 meV when the \( \vec{S}_{Mn} \)’s are along the two diagonal directions, and both 0.169 meV when the \( \vec{S}_{Mn} \)’s are along \( (\pm 2, \pm 1, 0) \) and \( (\pm 2, \mp 1, 0) \). These small \( \Delta E \)'s remain below the valid precision of VASP, implying again no
The observed small but non-negligible $\Delta E$’s are certainly not due to the inaccuracy in the calculations, which should be below 1 meV according to the estimations discussed before, but they arise from a bias field that originates in the SOC. Even more interestingly, $\Delta E$ reaches a maximum (absolute) value $\sim 10.125$ meV when $\vec{S}_{\text{Mn}}$ is along the [110] direction but it is almost zero (0.339 meV) when $\vec{S}_{\text{Mn}}$ is along the [110] direction in the pseudocubic frame. This nontrivial behavior exactly agrees with the $\vec{D}$'s vector directions, namely, here the $\vec{D}$'s across the interfaces are along the [110] direction, since the interfacial oxygens’ (in-plane) displacement is along the [110] direction, as found from the optimized crystal structure. Note that since SrRuO$_3$ is a FM metal with itinerant magnetic moments, the DM formula $\vec{D} \cdot (\vec{S}_{\text{Ru}} \times \vec{S}_{\text{Mn}})$, which is based on localized moments may be inaccurate quantitatively. Even though, our DFT results confirm a qualitative agreement with DM ideas from the anisotropy of $\Delta E$.

To further confirm the presence of a DM contribution, the $\vec{S}_{\text{Ru}}$ spins are also initialized to be in plane. However, in this case, $\Delta E$ is almost zero no matter whether $\vec{S}_{\text{Ru}}||\vec{S}_{\text{Mn}}$ or $\vec{S}_{\text{Ru}} \perp \vec{S}_{\text{Mn}}$, as shown in the last two lines of Table II. Thus these DFT data provide clear qualitative evidence that the bias field is in the form of $\vec{D} \cdot (\vec{S}_{\text{Mn}} \times \vec{S}_{\text{Ru}})$ and $\vec{D}$ is along the [110] direction of the pseudocubic frame.

The maximum value of the bias field driven by the DM interaction is found to be about 2.5 meV per Mn-O-Ru bond (here, totally four Mn-O-Ru bonds across two interfaces due to the periodic boundary conditions), which is larger than our previous estimation$^{22}$ by two orders of magnitude. There are two reasons for finding such a prominent bias field. The

Table I. DFT calculations with noncollinear spins but with the SOC disabled. The $\vec{S}_{\text{Mn}}$ and $\vec{S}_{\text{Ru}}$ vectors shown here are the initial magnetic moments, which will be reduced to the self-consistent values: $\sim 2.5–2.6\mu_B$ for Mn and $\sim 1.4\mu_B$ for Ru along their initial directions. The small canting components are $\sim 0.2\mu_B$ (out-of-plane) for $\vec{S}_{\text{Mn}}$ and $\sim 0.1\mu_B$ (in-plane) for $\vec{S}_{\text{Ru}}$, as shown in Fig. 3(a).

<table>
<thead>
<tr>
<th>$\vec{S}_{\text{Mn}}$</th>
<th>$\vec{S}_{\text{Ru}}$</th>
<th>$\Delta E$(meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(\pm 3,0,0)$</td>
<td>$(0,0,2)$</td>
<td>0.231</td>
</tr>
<tr>
<td>$(0,\pm 3,0)$</td>
<td>$(0,0,2)$</td>
<td>0.264</td>
</tr>
<tr>
<td>$(\pm 2,\pm 2,0)$</td>
<td>$(0,0,2)$</td>
<td>0.016</td>
</tr>
<tr>
<td>$(\pm 2,\pm 1,0)$</td>
<td>$(0,0,2)$</td>
<td>0.169</td>
</tr>
<tr>
<td>$(\pm 2,\mp 1,0)$</td>
<td>$(0,0,2)$</td>
<td>0.169</td>
</tr>
</tbody>
</table>

The observed small but non-negligible $\Delta E$’s are certainly not due to the inaccuracy in the calculations, which should be below 1 meV according to the estimations discussed before, but they arise from a bias field that originates in the SOC. Even more interestingly, $\Delta E$ reaches a maximum (absolute) value $\sim 10.125$ meV when $\vec{S}_{\text{Mn}}$ is along the [110] direction but it is almost zero (0.339 meV) when $\vec{S}_{\text{Mn}}$ is along the [110] direction in the pseudocubic frame. This nontrivial behavior exactly agrees with the $\vec{D}$’s vector directions, namely, here the $\vec{D}$’s across the interfaces are along the [110] direction, since the interfacial oxygens’ (in-plane) displacement is along the [110] direction, as found from the optimized crystal structure. Note that since SrRuO$_3$ is a FM metal with itinerant magnetic moments, the DM formula $\vec{D} \cdot (\vec{S}_{\text{Mn}} \times \vec{S}_{\text{Ru}})$, which is based on localized moments may be inaccurate quantitatively. Even though, our DFT results confirm a qualitative agreement with DM ideas from the anisotropy of $\Delta E$.

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most important one is that in our previous estimation a very small bond bending ($\delta\gamma$) was assumed, which was clearly an underestimation for the present (SrRuO$_3$)$_2$/SrMnO$_3$ case [$\approx10^\circ$, as shown in Fig. 2(b)]. Another possible reason for the discrepancy may be induced by the Ru cations because the SOC is disabled. The results are summarized in Table III.

TABLE III. DFT calculations with interfacial FE polarizations. Here only collinear spins are considered, namely the SOC is disabled. The $\delta_{\text{Ma}}$ and $\delta_{\text{Ru}}$ are input magnetic moments. The output ones are $\sim2.5$--$2.6\mu_\text{B}$ for Mn and $\sim1.4\mu_\text{B}$ for Ru.

<table>
<thead>
<tr>
<th>$\delta_{\text{Ma}}$</th>
<th>$\delta_{\text{Ru}}$</th>
<th>$\delta_\gamma$ (Å)</th>
<th>$\Delta E$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\pm3$</td>
<td>2</td>
<td>$(0.005,0.005,0)$</td>
<td>16.037</td>
</tr>
<tr>
<td>$\pm3$</td>
<td>2</td>
<td>$(-0.005,0.005,0)$</td>
<td>16.486</td>
</tr>
</tbody>
</table>

B. FE-driven EB

To fully verify our previous proposals, it is now necessary to check the bias field that is driven by the FE polarizations. However, here neither SrRuO$_3$ nor SrMnO$_3$ is a FE material. Then, to test this FE mechanism it is necessary to apply an “artificial” polarization at the interfaces. For this purpose, the oxygen anions between Ru and Mn are slightly displaced ($\delta_\gamma$) uniformly from their optimized positions to mimic an in-plane polarization. These displacements produce modulated Ru-O-Mn bonds $[\gamma = 171.6^\circ$ and $\beta = 168.5^\circ$ when $\delta_\gamma = (0.005,0.005,0)$Å, while the original one is $\gamma = 170.4^\circ$], as shown in Figs. 2(b)–2(d). Although these displacements are imposed manually, their result, namely, the presence of modulated bonds, had already been confirmed in our previous DFT optimized analysis of La$_{3/3}$Sr$_{1/4}$MnO$_3$/BiFeO$_3$ superlattices since BiFeO$_3$ is a multiferroic material (for more details, see the supplementary material of Ref. 22). Since the proposed FE-driven EB does not require noncollinear spin configurations, here only collinear patterns are considered and the SOC is disabled. The results are summarized in Table III.

From the results shown above, it is obvious that the in-plane FE polarization at interfaces can indeed induce an EB field via the modulation of normal exchanges (note that the total effect on the DM interaction from such a bond angle modulation will be canceled at the first order approximation, and thus can be neglected especially when the weak intensity of the DM interaction is considered). The bias field is about $4$ meV per Ru-O-Mn bond and the FE polarization corresponding to $|\delta_\gamma| = 0.005\sqrt{2}$ Å is about $3.5$ mC/m$^2$. Although to be expected, it is very interesting to note that this EB field can be switched between positive and negative values by flipping the FE polarization using electrical methods, which can provide hints to understand the interfacial magnetoelectricity in BiFeO$_3$-based multiferroic heterostructures. It should be pointed out that the exchange striction effect could also induce such a modulation of the bond angles, namely, the bond angle between a parallel spins’ pair is different from that between an antiparallel spins’ pair, e.g., in the multiferroic orthorhombic HoMnO$_3$ which is driven by the nonrelativistic exchange ($J$) between spins. This bond modulation can be easily observed if the lattice is optimized under a collinear G-AFM plus FM configuration: the $\text{Ru(}\uparrow\text{-}O\text{-Mn(}\uparrow\text{)}$ [or $\text{Ru(}\downarrow\text{-}O\text{-Mn(}\downarrow\text{)}$] becomes 164.5° while the $\text{Ru(}\uparrow\text{-}O\text{-Mn(}\downarrow\text{)}$ [or $\text{Ru(}\downarrow\text{-}O\text{-Mn(}\uparrow\text{)}$] becomes 168.3°. However, in principle, this bond modulation would be switched following the switching of FM moments, implying the absence of a bias effect although the coercive field might be increased. Thus only the FE polarization can give rise to a robust bond modulation with an origin independent of the exchange striction. Of course, the (inverse effect of) exchange striction can affect the underlying FE polarization when switching the FM moments, which is also an important issue for interfacial magnetoelectricity.

IV. CONCLUSION

Summarizing, in this investigation, the $ab$ $initio$ method has been used to verify previous proposals that were presented to rationalized the exchange bias effect in compensated antiferromagnets. The DFT calculation discussed here indeed provides qualitative evidence that both the Dzyaloshinskii-Moriya interaction and the ferroelectric polarization can induce exchange bias effects in perovskites heterostructures. Their common ingredient is the intrinsic bending of the bonds across the interface, which is induced by the oxygen octahedra rotations and tiltings. It can be argued that the proposed mechanisms originate from “uncompensated exchanges,” namely, the exchanges ($\hat{D}$ and $J$) are no longer uniform but modulated following the period of the antiferromagnetic order. The “uncompensated exchanges” emphasized in this work have usually been ignored in the previous models for the EB effect that mainly rely on uncompensated moments at the AFM interfaces.

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Due to the two interfaces arising from the periodic boundary conditions, the displacements of oxygens at the upper and lower interfaces should be opposite.

Many spin configurations, including (1) full FM; (2) full G-type AFM (Ru); (3) full A-type AFM; (4) full C-type AFM; (5) C-type AFM (Mn); (6) G-type AFM (Mn) + A-type (Ru); (7) G-type AFM (Mn) + A-type (Ru), have been tested, all of which have higher energies than the G-type AFM (Mn) + FM (Ru) configuration. However, an A-type AFM (Mn) + FM (Ru) configuration has even lower energy. This is due to the strong Ru-O-Mn exchange coupling because here the SrMnO₃ portion is too thin: two Ru-Mn interfaces for two Mn layers. In real heterostructures, the magnetic moments in SrMnO₃ are indeed not be affected qualitatively.

Due to the two interfaces arising from the periodic boundary conditions, the displacements of oxygens at the upper and lower interfaces should be opposite.


108. Many spin configurations, including (1) full FM; (2) full G-type AFM; (3) full A-type AFM; (4) full C-type AFM; (5) C-type AFM (Mn) + A-type AFM (Ru); (6) C-type AFM (Mn) + FM (Ru); (7) G-type AFM (Mn) + A-type (Ru), have been tested, all of which have higher energies than the G-type AFM (Mn) + FM (Ru) configuration. However, an A-type AFM (Mn) + FM (Ru) configuration has even lower energy. This is due to the strong Ru-O-Mn exchange coupling because here the SrMnO₃ portion is too thin: two Ru-Mn interfaces for two Mn layers. In real heterostructures, the magnetic moments in SrMnO₃ are indeed canting from the ideal G-type AFM when there are only two Mn layers, but this arrangement will return to the G-type AFM state when SrMnO₃ is thick enough. Even with these caveats, since a G-type AFM + FM configuration can be stabilized in the model system, the conclusion of our proof-of-principle calculations will not be affected qualitatively.