

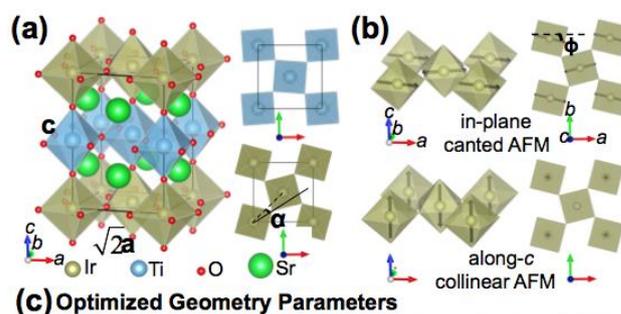
Project Title:
Theoretical Modeling of Surface/Interface States in Topological Materials
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1. Introduction

The discovery of novel effective total angular momentum $J_{\text{eff}} = 1/2$ states in Sr_2IrO_4 by Kim et al. is like a knocking stone dropped into a peaceful pond. The inherently large spin-orbit coupling (SOC) of 5d orbitals in Ir ion reorganizes the t_{2g} states into so called $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ states, provided that the local crystal field separates unoccupied e_g states and partially occupied t_{2g} states (five electrons per Ir ion) in a much larger energy scale. Soon after the discovery of the J_{eff} states, Moon et al. have established a general picture of the electronic states for Ruddlesden-Popper series $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ ($n = 1, 2, \infty$) based on the interplay among SOC, electron correlations (U), and band width (W). As W becomes larger with increasing the number n of perovskite layers, the series turns from insulating ($n = 1, 2$) to metallic ($n = \infty$) state. Although the insulating Ir oxides, Sr_2IrO_4 (hereafter 214) and $\text{Sr}_3\text{Ir}_2\text{O}_7$ (hereafter 327), have promoted a large amount of research, the metallic compound, SrIrO_3 (hereafter 113), has not received much attention. This is presumably because on the experimental side, it requires high pressure and high temperature to synthesize 113, and on the theoretical side, 113 has three-dimensional (3D) perovskite structure while the other 214 and 327 compounds are in two-dimensional (2D) perovskite structures. The available experimental results suggest that 113 is a bad metal close to the metal-insulator transition boundary, as manifested by the low-temperature upturn in resistivity.

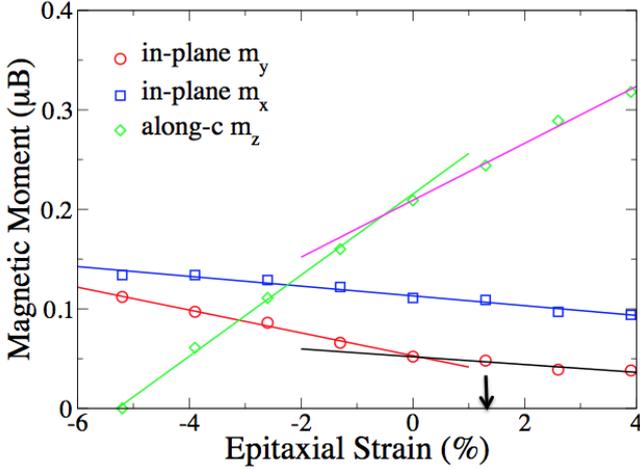
Recently, Matsuno and coworkers have fabricated the (001) $[\text{SrIrO}_3]_m/[\text{SrTiO}_3]_1$ (hereafter SIO/STO) superlattice (SL) and found a signature of metal-insulator transition as the number m of SrIrO_3 layers decreases below 4. Thus building

superstructures such as SL is another effective way to tip the metallic state in 113 bulk to an insulating state. It has been suggested that the magnetic structure is a canted in-plane antiferromagnetic (AFM) order with a weak ferromagnetic moment (see Fig.1(b)) for a single 113 layer ($m = 1$). This in-plane canted AFM structure is the same ground state as the one for 214 bulk. Thus it is interesting to make a comparable study between SIO/STO SL and 214 bulk. Indeed, both experimental and theoretical studies suggest that SIO/STO SL with a single 113 layer ($m=1$) is similar to 214 bulk in the electronic and magnetic structure. By the same token, it is also natural to expect that SL with $m > 1$ would be comparable to $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ ($n > 1$), following Moon's picture. The story would come to the end if SIO/STO SL were simply an artificial duplicate material of the corresponding bulk. As will be shown below, the detailed analysis reveals distinct properties between the SL and bulk compounds due to the intrinsic structural difference.



Based on first principles calculations, we report the electronic and magnetic properties under various lattice distortion for the SL with $m = 1$ ($[\text{SrIrO}_3]_1/[\text{SrTiO}_3]_1$). Considering two different magnetic structures, we show that in both cases the

magnetic moment depends differently on the compressive or tensile strain. The band structure is also found to be sensitive to local distortion. We demonstrate that the band gap in the $J_{\text{eff}} = 1/2$ based bands can be closed in certain condition to induce an insulator-metal transition.



2. Calculation method

In the present study, a $[(\text{SrIrO}_3)_m, \text{SrTiO}_3]$ superlattice was constructed and first principles calculations were performed to investigate the intriguing metal-insulator transition (MIT) with respect of decreasing m . Calculations were carried out by Vienna Ab initio Simulation Package (VASP) and Full Potential Linearized Augmented Wave code (ELK). To study the electronic properties of superlattice, local density approximation (LDA) + U + spin orbit coupling (SOC) calculations of SrIrO_3 bulk was first carried out to test the validity of results. As Ir^{4+} has a $5d^5$ configuration, the SOC is large (~ 0.5 eV) and correlation should be small due to the fact of extended feature of $5d$ orbitals. We choose U to be 1eV (We also tested other values later). All the calculations were performed on Massively Parallelized PC and Memory sharing UPC clusters of RICC.

3. Result

The structure crystallizes to a tetragonal lattice, as shown in Fig. 1(a). The lattice constants obtained are $a=3.83 \text{ \AA}$ and $c=7.76 \text{ \AA}$, which are in consistent with the experimental value. To determine the ground state magnetic structure, we calculate the

total energy of two different magnetic structures, the canted in-plane AFM and along- c collinear AFM structures, as shown in Fig. 1(b). The ground state turns out to be canted AFM with a weak ferromagnetic moment along either a or b axis, which is in good agreement with experiments [6] and previous calculations. Due to the correlation U , a band gap opens between the $J_{\text{eff}}=1/2$ based states (see Fig. 3(a)). In the following subsections, we examine in detail how the magnetic and electronic structure changes under strain or local octahedron distortions.

To study strain effects, we systematically change the lattice constant in ab -plane from 3.75 \AA to 3.95 \AA , and relax c -axis lattice constant as well as the internal atomic coordinates for a given in-plane lattice constant until the optimized structure is obtained. The optimized c -axis lattice constant decreases under tensile strain and increases under compressive strain, which is in consistent with expectation. The IrO_6 (TiO_6) octahedron is distorted locally in an antiferroelectric way, i.e., rotating along the c -axis in phase (out of phase), instead of simply altering the Ir-O (Ti-O) bond length, as shown in Fig. 1(a).

As shown in Fig. 2, the magnetic moments for the two different magnetic structures exhibit different dependence of compressive or tensile strain. Because the canted AFM structure has a weak ferromagnetic moment, we decompose the local magnetic moment to m_y (along the AFM direction) and m_x (along the canted ferromagnetic direction). We also plot in Fig. 2 the magnetic moment (m_z) for the collinear AFM structure along c direction. We find that both m_y and m_x increase with increasing the compressive strain while m_z enhances with increasing the tensile strain. The different dependence is due to the resulting tetragonal distortion by either compressive or tensile strain. The compressive strain shifts the d_{xy} orbital level ($E_{d_{xy}}$) higher in energy than the d_{yz} and d_{zx} levels ($E_{d_{yz/xz}}$), and thus $\Delta = E_{d_{xy}} - E_{d_{yz/xz}}$ is positive. On the other hand, the tensile strain shifts

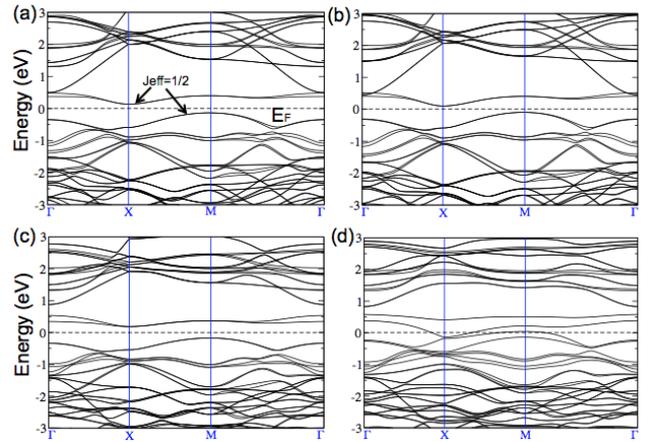
$E_{d_{xy}}$ lower than $E_{d_{yz/xz}}$, and thus Δ is negative. A positive Δ favors the canted AFM structure while a negative Δ favors the collinear AFM structure.

An unexpected behavior is found in the strain dependence of the magnetic moments. As shown in Fig. 2, m_x for the canted AFM structure seems to vary smoothly under compressive or tensile strain. However, m_y for the canted AFM structure and m_z for the collinear AFM structure exhibit different strain dependence for the compressive and tensile cases. We have also examined the strain dependence of α_{Ir} and α_{Ti} and found that α_{Ti} is correlated to the unexpected behavior of the magnetic moments m_y and m_z . The reason why α_{Ti} is relevant to the magnetic moments can be understood in the electronic band structures. In Fig. 3(b), we show the electronic band structure for SL ($m = 1$) under compressive strain. The bands with the $j_{\text{eff}} = 1/2$ character and the bands with the Ti-3d character already mix with each other above Fermi level at around 6 eV at the Γ point. This indicates that the electron hopping between Ti-3d and $j_{\text{eff}} = 1/2$ states should play a role to influence Ir magnetic moment via the effective exchange interaction.

The local lattice distortion affects the electronic band structure and sometimes induces novel electronic phases. Fig. 3(a) shows the electronic band structure for SL ($m = 1$) with the optimized structure. The band gap opens between the upper and lower $J_{\text{eff}} = 1/2$ based bands, which is in good agreement with the previous reports. The bands immediately above the upper $J_{\text{eff}} = 1/2$ based bands (around 0.5 eV at the Γ point) have Ti-3d character, indicating that the Ti-3d bands are close to the upper $J_{\text{eff}} = 1/2$ based bands. This behavior is more pronounced when the compressive strain is applied, as shown in Fig. 3(b), where the upper $J_{\text{eff}} = 1/2$ based bands are mixed with the bands originated from Ti-3d at the Γ point around 0.5 eV above the Fermi energy (E_F).

Next, we increase the rotation of local IrO_6 and TiO_6 octahedra both in-phase (IrO_6) and out-of-phase (TiO_6) and examine how the electronic

band structure evolves. The electronic band structure obtained is shown in Fig. 3(c), corresponding to $\alpha_{Ir} = \alpha_{Ti} = 16^\circ$, which is larger than the optimized values $\alpha_{Ir} = 14.5^\circ$ and $\alpha_{Ti} = 3.5^\circ$. Due to the enhanced rotational distortion, the band mixing between the upper $J_{\text{eff}} = 1/2$ based bands and the Ti-3d based bands disappears. Now if we adjust the local ab-plane Ir-O (Ti-O) bond length to be inequivalent between the nearest neighbor octahedrons, the band structure changes dramatically, especially in the $J_{\text{eff}} = 1/2$ based bands, as shown in Fig. 3(d). The almost doubly degenerate upper $J_{\text{eff}} = 1/2$ bands along the X-M line are lifted and one of these bands is pushed downward to cross the Fermi energy. This unexpected insulator-metal transition demonstrates that the $J_{\text{eff}} = 1/2$ based bands of the SIO/STO SL with $m = 1$ are very sensitive to local distortion.



4. Conclusion

We have performed first principles calculations on the SIO/STO superlattice with several approximate schemes including LDA, LDA+U, LDA+SOC, and LDA+SOC+U. We have demonstrated that the magnetic and electronic structure is very sensitive to the lattice distortion. We have found that the magnetic moments follow the different linear dependence under the compressive or tensile strain. The electronic band structure indicates that the magnetic structure is influenced by the band mixing between $J_{\text{eff}} = 1/2$ based bands and the Ti-3d based bands above E_F . The degree of this mixing can be tuned by the strain.

Indeed, we have found that the band mixing increases (decreases) by the compressive strain (by the tensile strain or by increasing the local rotation of IrO_6 and TiO_6 octahedron). We have also shown that in certain conditions, the band gap can be closed between the $J_{\text{eff}} = 1/2$ based bands, e.g., by increasing the local rotation with an inequivalent in-plane Ir-O (Ti-O) bond length. These unusually sensitive electronic and magnetic properties to the lattice distortion urgently require more careful and intensive research in 5d materials with the strong SOC.

5. Future work

Density functional theory based calculations are very efficient on RICC. We will continue working on first principle calculations of topological phase realization of SrIrO_3 based interface or superlattice. Meanwhile, to investigate the effect of electron correlation, we will also try to perform LDA+DMFT calculation.