Correlation effects in (111) bilayers of perovskite transition-metal oxides

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We investigate the correlation-induced Mott, magnetic and topological phase transitions in (111) bilayers of perovskite transition-metal oxides LaAuO₃ and SrIrO₃ for which the previous density functional theory calculations predicted topological insulating states. Using the dynamical-mean-field theory with realistic band structures and Coulomb interactions, LaAuO₃ bilayer is shown to be far away from a Mott insulating regime, and a topological-insulating state is robust. On the other hand, SrIrO₃ bilayer is on the verge of an orbital-selective topological Mott transition and turns to a trivial insulator by an antiferromagnetic ordering.

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Topological insulators (TIs) are novel quantum states of matter characterized by the nontrivial band topology due to the relativistic spin-orbit coupling (SOC) [1–7]. While the TI states are essentially single-particle phenomena, the interplay between the SOC and strong Coulomb interactions in electronic systems has gained considerable attention [8–11]. A variety of effects between these two couplings has been explored, such as TI states induced by Coulomb interactions [11,12], competition and/or cooperation between the two in a Bernevig-Hughes-Zhang (BHZ) model [13,14], and topological Kondo insulators [15,16]. However, until now physical realization for such correlation-related phenomena has been limited to an f electron system SmB₆ [19,21].

Here, we focus on d electron systems transition-metal oxides (TMOs) which cover a vast extension of novel phenomena due to the strong electron-electron and/or electron-lattice interactions, such as high-Tc superconductivity and colossal magnetoresistance effects [22]. The recent development in synthesizing artificial TMO heterostructures provides great tunability over fundamental physical parameters [23]. Once realized in TMOs, combining TIs with other novel states would allow us to study further novel phenomena utilizing proximity effects involving TIs [24,25]. In Ref. [26], based on the tight-binding modeling and subsequent density functional theory (DFT) calculations, bilayers of perovskite TMOs grown along the [111] crystallographic axis were proposed as possible candidates for two-dimensional topological insulators. However, the effect of electron-electron interactions was not addressed except for limiting cases. The correlation effects were investigated for nickelate (111) bilayer using Hartree-Fock approximations [27,28]. For iridate (111) bilayer, these effects were studied using a strong coupling approach from a Mott insulating side [29] and DFT+U and modified exchange potential [30].

In this paper, we investigate the correlation-induced phase transitions in (111) bilayers of SrIrO₃ (SIO) and LaAuO₃ (LAO) by means of the dynamical-mean-field theory (DMFT) [31]. It is shown that LAO bilayer is far away from Mott insulating and antiferromagnetic (AF) insulating regimes, and a TI state is robust. On the contrary, the correlation effect is significant for SIO bilayer, and an AF trivial insulating state is realized. This is induced by a relatively-narrow band width of near-Fermi-level states with the dominant $J_{\text{eff}} = 1/2$ character, which undergo an orbital-selective topological Mott transition when the magnetic ordering is suppressed.

For a realistic treatment on the band effects, we generate Wannier functions [32] from the outputs of DFT calculations reported previously [26]. The details of Wannier functions and comparison with DFT dispersions are presented in the supplementary information [33]. Correlation effects are dealt with using DMFT with the exact diagonalization impurity solver [34,35]. Here, the effective medium is approximated as a finite number of bath sites coupled to eigenstates of the local non-interacting Hamiltonian [35]. With the SOC and the trigonal crystal field, all degeneracies in d multiplet are lifted except for the Kramer’s degeneracy [26]. For a $t_{2g}$ system SIO there are three Kramer’s doublets, and for an $e_g$ system LAO there are two Kramer’s doublets. Thus, we introduce two bath sites per Kramer’s doublet for SIO and three bath sites per doublet for LAO [37]. Note that the highest energy doublet for SrIrO₃ bilayer has the strong $J_{\text{eff}} = 1/2$ character, while the other two doublets have the $J_{\text{eff}} = 3/2$ characters. As the typical gap amplitude of our systems is $\sim 0.01-0.1$ eV, we introduce temperature $T = 0.01$ eV and retain the lowest eigenstates of the interacting impurity Hamiltonians with Boltzmann factors larger than $10^{-5}$. The DMFT self-consistency condition is closed by updating the bath Green’s functions at each iteration by minimizing with a conjugate gradient algorithm a distance function that includes frequency dependence on the discrete Matsubara frequency $\omega_n = (2n+1)\pi T$ with a frequency weighting $1/|\omega_n|$ [38].
FIG. 1: (Color online) Bulk spectral function for (a) SrIrO$_3$ bilayer and (b) LaAuO$_3$ bilayer with $U = 2.0$ and $J = 0.2$ eV. Dotted lines show dispersion relations of non-interacting models.

Figure 1 shows the typical paramagnetic (PM) bulk spectral functions for (a) SIO bilayer and (b) LAO bilayer with $U = 2.0$ and $J = 0.2$ eV. The spectral functions are computed using the self-energy directly obtained on the real axis, where the delta function is broadened by using the logarithmic Gaussian function [39]. While both systems have similar bare band width $\sim 2$ eV, the effect of correlations is notably different. For SIO bilayer, two nearly-flat dispersions appear at the Fermi level. For LAO bilayer, the quasiparticle weights of two Kramers doublets (filled and open squares) show nearly-flat dispersions at the Fermi level $\omega = 0$. These bands are mainly coming from the $J_{\text{eff}} = 1/2$ states, but their effective mass is enhanced significantly due to the correlation effects and a large amount of spectral weight is transferred to higher energy regimes, forming the upper Hubbard band at $\sim 1$ eV and the lower Hubbard band at $\sim -0.3$ eV. The other states mainly from $J_{\text{eff}} = 3/2$ states are relatively unaffected, and their dispersion relations are nearly identical to those of non-interacting model with about 0.5 eV downshift and broadening due to the imaginary part of the self-energy. On the other hand, the spectral function for LAO bilayer is not modified from the non-interacting case except for the moderate band renormalization and broadening away from the Fermi level.

In order to see the nature of correlation-induced Mott transition, we plot the quasiparticle weight and the gap amplitude as a function of $U$ in Fig. 2. The quasiparticle weight of Kramers doublet $\alpha$ is evaluated from the self-energy at the lowest Matsubara frequency as $Z_{\alpha} = (1 - \Im \Sigma_{\alpha}(i\omega_0)/\omega_0)^{-1}$. To avoid the broadening of the spectral function by the imaginary part of the self-energy, the gap amplitude is evaluated from quasiparticle dispersions [40]. In both cases, the gap amplitude is monotonically decreased with increasing $U$ and does not show a transition to a high $U$ TI state in which the insulating gap increases with $U$ as reported for SIO bilayer in Ref. [30]. On the contrary, the gap amplitude is strongly correlated with the (smallest) quasiparticle weight, and they become zero simultaneously at Mott transitions. For SIO bilayer, one of three Kramers doublets with the $J_{\text{eff}} = 1/2$ character undergoes the Mott transition (filled squares) while the other two with the $J_{\text{eff}} = 3/2$ character do not (filled and open circles). This situation resembles that in the single-layer perovskite Sr$_2$IrO$_4$ [43]. But, as the transition in SIO bilayer accompanies the change in the band topology from a nontrivial one to a trivial one as confirmed from the fully gapped edge spectra in the latter [33], this transition is an orbital-selective topological Mott transition. For LAO bilayer, the quasiparticle weights of two Kramers doublets (filled and open squares) show nearly-identical $U$ dependence. Whether or not PM Mott insulating states of two systems support gapless “spinon” edge states, i.e. topological Mott insulators [41], is a very interesting question but remains beyond the scope of the current single-site DMFT.

We next study the effect of magnetic ordering. From both a weak-coupling approach and a strong-coupling approach [29], a Néel AF ordering is expected in SIO
bilayer. Our DFT calculation indeed found the Néel AF ordering with magnetic moments primarily lying perpendicular to the [111] plane. While the similar ordering is expected in LAO bilayer, our DFT calculations with $U$ up to 4 eV and $J/U$ up to ~0.2 did not find any magnetic ordering. This could be ascribed to the fact that the local moment formation is underestimated in DFT. Nevertheless, we assume the Néel AF ordering for both systems by taking the [111] axis, perpendicular to the plane, as the spin quantization axis and examine its stability.

Figure 3 shows the magnetic order parameters and the gap amplitude for (a) SIO and (b) LAO. As expected from the smaller critical $U$ for the Mott transition in SIO, the critical $U$ for an AF ordering is also small, $U \sim 0.5$ eV. In Fig. 3 (a), we also plot the magnetic order parameters obtained by generalized gradient approximation (GGA)+$U$. The critical $U$ for the AF ordering appears to be slightly smaller than the present DMFT result and much smaller than local density approximation +$U$ results in Ref. 30 where the AF ordering appears at $U > 3$ eV. It is noted that the DMFT order parameters are somewhat larger than those of GGA+$U$. In both the DMFT and GGA+$U$ results, the orbital moment and the spin moment are tilted with each other. In GGA+$U$, the spin moment is deviated from the [111] direction more strongly than the orbital moment, leading to the downturn of its [111] component at $U \gtrsim 2$ eV. As the spin quantization axis is fixed to the [111] axis in our DMFT calculations, the orbital moment is tilted. However its absolute value $|\mu_L|$ is about a factor 2 larger than the spin moment $\mu_S$ (about 3.8 in this case). This indicates the deviation of the spin symmetry from the SU(2) point due to the local trigonal field by which $a_{1g}$ (singlet) is lower than $e_g^2$ (doublet) when the SOC is suppressed. While our GGA+$U$ calculation did not find an AF ordering in LIO, the DMFT calculation did find the stable AF ordering at $U \gtrsim 2.1$ eV. As a Au$^{+3}$ ion has the $e_g^2$ electron configuration, DFT+$U$ requires very large $J$ to induce local moments $S = 1$. On the other hand, with finite $J$, $e_g^2$ states with $S = 1$ always have larger weights than those with $S = 0$. These weights increase with increasing $J$, and therefore a magnetic ordering can be induced more easily in DMFT.

In SIO bilayer, an AF Néel ordering was found to destroy the TI state immediately. This is because in a honeycomb lattice with the AF ordering, there is no combined symmetry with the time reversal by which the system remains invariant [15]. In LAO bilayer on the other hand, there appears a finite window for an AF TI where the gapless edge states and bulk magnetic ordering (time-reversal symmetry breaking) coexist. (See supplementary material for comparison between the AF TI and the AF trivial insulator.) This comes from the fact that the spin component perpendicular to the [111] plane is conserved in our $e_g$ electron model [26], and therefore LAO bilayer consists of two copies of Chern insulators with the opposite Chern number. The transition between the AF TI phase and the AF trivial insulating phase in LAO bilayer is found to be of the first order accompanied by a jump in the staggered magnetic moment. Thus, the gap closing is avoided. This transition could become a continuous one by including spatial correlations beyond the single-site DMFT. For instance, for a correlated BHZ model, a single-site DMFT calculation found a discontinuous transition from a PM TI phase to an AF trivial phase [14]. But a variational cluster approach found a continuous transition accompanied by the gap closing, leaving a finite parameter window for an AF TI phase [14].

Are SIO and LAO in AF trivial insulating phases or TI phases? If they are in AF trivial phases, can we turn them into TI phases by suppressing magnetic ordering? To answer these questions, we estimate realistic Coulomb interactions by using the constrained random phase approximation (cRPA) [23] [11] [12]. For SIO bilayer, we took the Slater parameters $F_{0,2,4}$ for Sr$_2$IrO$_4$ from Ref. 33 and deduced the intraorbital Coulomb interaction $U$ and the interorbital exchange and pair transfer $J$ as 2.232 eV and 0.202 eV, respectively, for the $\{xy,yz,zx\}$ basis. For LAO bilayer, we directly com-
puted these parameters for the $\{3z^2 - r^2, x^2 - y^2\}$ basis as $U = 1.80$ and $J = 0.225$ eV [33]. For both cases, the interorbital Coulomb interaction is given by $U' = U - 2J$. Note that $J/U \approx 0.1$ in both cases. These realistic parameter regimes are approximately indicated by shades in Fig. 3.

With the realistic parameters, we confirmed that an AF Néel ordered state is realized in SIO bilayer and a PM state in LAO bilayer. From the edge spectra, the AF SIO is an AF trivial insulator while the PM LAO is a TI (see Fig. 4). When an AF Néel order is suppressed, SIO bilayer turns into a PM TI not a PM Mott insulator. This situation is quite similar to that in Sr$_2$IrO$_4$ [33-34]. Since the gap amplitude of PM SIO bilayer is about 0.002 eV, which is an order of magnitude smaller than temperature $T = 0.01$ eV [see Fig. 2(a)], thermal broadening might hinder the TI nature and SIO bilayer would behave as a topological semimetal. In reality, finite spatial correlations may induce a pseudogap even without an AF long-range order in a PM phase of SIO bilayer with realistic Coulomb interactions. Whether or not such a pseudogap phase supports gapless electron or spinon edge modes remains a very interesting problem. A high pressure may push SIO bilayer back to a TI. Also, doping carriers into SIO bilayer may induce the unconventional superconductivity [35-36].

To summarize, we have investigated the correlation-induced phase transitions in (111) bilayers of perovskite TMOs SIO and LAO using the DMFT. SIO is found to be in the vicinity of an orbital-selective topological Mott transition, where only one band with the strong $J_{eff} = 1/2$ character shows the significant quasiparticle renormalization and undergoes “band insulator”–Mott insulator transition. As the effective band width of such a $J_{eff} = 1/2$-type band is narrow, SIO is unstable against a Néel AF ordering, resulting in an AFM insulator. On the contrary, LAO remains a correlated TI, far away from a Mott insulator and an AF trivial insulator.

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FIG. 4: (Color online) Edge spectral functions for 40-site thick zigzag slabs of (a) SrIrO$_3$ bilayer with $U = 2.232$ eV and $J = 0.202$ eV and (b) LaAuO$_3$ bilayer with $U = 1.80$ and $J = 0.225$ eV, $a$ is the nearest-neighbor distance projected on the [111] plane. Because of an AF ordering, SrIrO$_3$ bilayer does not support gapless edge modes.

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**Supplementary material: Correlation effects in (111) bilayers of perovskite transition-metal oxides**

### S1. DENSITY FUNCTIONAL THEORY

Computational details for the density functional theory (DFT) calculations are given in Ref. [26]. In this work, to obtain the Wannier functions, thinner supercells are adopted than in Ref. [26], consisting of 6 AO and 6 B layers along the [111] direction with (A,B) = (La,Al) or (Sr,Ti) with two adjacent B layers replaced by TM ions. We have checked that current DFT results agree with the previous ones.

Figure [S1] shows the resultant DFT dispersion relations (blue solid lines) and those of Wannier functions. With the finite spin-orbit coupling (SOC), SrIrO\(_3\) (111) bilayer was found to have tiny but finite magnetic moments, which prevent us from constructing maximally localized Wannier functions (MLWFs) with proper symmetries, whose dispersion relations are given by red dashed lines [Fig. [S1] (a), Wannier1]. To completely suppress the magnetism, we turned off the SOC and derived MLWFs and an effective tight-binding model. Then, the following atomic SOC is added to such a model Hamiltonian

\[ H_{SOC} = \frac{\lambda}{2} \sum_{\sigma} \sum_{\tau \tau'} \epsilon_{\tau \tau' \sigma''} d_{\tau \sigma} \sigma' d_{\tau' \sigma''}, \tag{S0.1} \]

where the following convention for the orbital index is used: \(|a\rangle = |d_{pz}\rangle, |b\rangle = |d_{zx}\rangle, |c\rangle = |d_{xy}\rangle\). \(\sigma\) with \(\tau = a, b, c\) is the Pauli matrix, and \(\epsilon_{\tau \tau' \sigma''}\) is the Levi-Civita antisymmetric tensor. Using \(\lambda\) as a fitting parameter,
we derive the model Hamiltonian for SrIrO$_3$ (111) bilayer. By matching the band gap, we optimized $\lambda$ as 0.36 eV. The resulting dispersion relations are given by green solid lines [Fig. S1 (a), Wannier2]. The data set Wannier2 is used in our realistic dynamical-mean-field theory calculation for SrIrO$_3$ (111) bilayer. While we did not have issues associated with magnetic moments for LaAuO$_3$ (111) bilayer, we found that the symmetry of Wannier functions is progressively reduced by iterations. To avoid this, we set Num_iter to 0 and derived “one-shot” Wannier functions including the SOC. The resultant dispersion relations are given by red dashed lines [Fig. S1 (b), Wannier]. This data set is used in our realistic dynamical-mean-field theory calculation for LaAuO$_3$ (111) bilayer.

The dispersion relations for 40-site thick zigzag slab of SrIrO$_3$ (111) bilayer using the Wannier2 parameter set are given in Fig. S1 (c). Corresponding results for 40-site thick zigzag slab of LaAuO$_3$ (111) bilayer are given in Fig. S1 (d). Slab dispersions confirm that both SrIrO$_3$ (111) bilayer and LaAuO$_3$ (111) bilayer are topological insulators with gapless edge modes crossing the Fermi level indicated by red lines.

Coulomb interactions for LaAuO$_3$ (111) bilayer is estimated by using the constrained random phase approximation (cRPA) [41] as described in Ref. [42]. For this purpose, we constructed a thinner supercell consisting of 2 LaAuO$_3$ and 1 LaAlO$_3$ layers along the [111] direction in which the local structure taken from the previous results with thicker LaAlO$_3$ layers. We used the density response code for Elk [42]. The polarization function was expanded in plane waves with an energy cutoff of 5 Ry and the total number of bands considered in the polarization calculation was set to 80.
FIG. S2: Edge spectral functions for 40-site thick zigzag slabs of (a) SrIrO$_3$ (111) bilayer with $U = 2.6$ eV and $J = 0.26$ eV and (b) LaAuO$_3$ bilayer with $U = 4.4$ eV and $J = 0.44$ eV without a magnetic ordering. These are in Mott insulating phases.

S2. MOTT PHASES

Here, we discuss the band topology of our (111) bilayers of TMOs in paramagnetic Mott insulating regimes. Figure S2 shows the edge spectral functions for 40-site thick zigzag slabs of (a) SrIrO$_3$ bilayer with $U = 2.6$ eV and $J = 0.26$ eV and (b) LaAuO$_3$ bilayer with $U = 4.4$ eV and $J = 0.44$ eV without a magnetic ordering. As seen in Fig. 2, these are in Mott insulating regimes. The absence of edge modes crossing the Fermi level at $\omega = 0$ indicates both are in trivial phases at least “electronically”. There is a possibility that the nontrivial band topology remains in spinon dispersions supporting bulk band gaps and gapless edge modes, i.e., topological Mott insulators, but the current dynamical-mean-field theory does not have access to such states.

S3. ANTIFERROMAGNETIC PHASES IN LaAuO$_3$ BILAYER

Here, we examine the nature of antiferromagnetic phases in LaAuO$_3$ bilayer. Figure S3 shows the edge spectral functions for 40-site thick zigzag slabs of antiferromagnetic LaAuO$_3$ (111) bilayer with (a) $U = 2.14$ eV and (b) $U = 2.16$ eV with $J/U = 0.1$. Because of the Néel antiferromagnetic ordering, the symmetry between $k > 0$ and $k < 0$ is broken. While the antiferromagnetic ordering breaks the time-reversal symmetry, the combined symmetry between the time reversal and mirror (mirror plane perpendicular to the zigzag direction) remains. As a result, each mode is two-fold degenerate. We see gapless edge modes at the Fermi level in (a) while not in (b), indicating the former is in...
FIG. S3: Edge spectral functions for 40-site thick zigzag slabs of LaAuO$_3$ (111) bilayer with (a) $U = 2.14$ eV and (b) $U = 2.16$ eV with $J/U = 0.1$. There are gapless edge modes at the Fermi level $\omega = 0$ in (a) while there is not in (b).

...a topologically nontrivial phase but the latter is in a trivial phase.