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Upon reduction of the film thickness we observe a metal-insulator transition in epitaxially stabilized, spin-orbit-coupled SrIrO₃ ultrathin films. By comparison of the experimental electronic dispersions with density functional theory at various levels of complexity we identify the leading microscopic mechanisms, i.e., a dimensionality-induced readjustment of octahedral rotations, magnetism, and electronic correlations. The astonishing resemblance of the band structure in the two-dimensional limit to that of bulk SrIrO₃ opens new avenues to unconventional superconductivity by “clean” electron doping through electric field gating.

Although typically viewed as disparate properties, the interplay between strong spin-orbit coupling (SOC) and electronic correlations in high-Z 5d transition metal oxides can lead to exotic quantum states of matter like Kitaev spin liquids [1,2] and topological phases [3,4]. A prominent example is the Mott-insulating state found in the prototypical system Sr₂IrO₄ [5], which is promoted by the lifted orbital degeneracy of the t₂g manifold due to the entanglement of orbital and spin degrees of freedom in the presence of strong SOC [1]. Its quasi-two-dimensional (2D) layered-perovskite structure with corner-shared IrO₆ octahedra hosts a square lattice of antiferromagnetically coupled J_{eff} = 1/2 pseudospins reminiscent of the high-T_C superconducting cuprate parent materials [6]. Indeed, Sr₂IrO₄ is considered a promising candidate for exotic superconductivity since it reproduces much of the fermiology of hole-doped cuprates upon electron doping [7,8].

The strontium iridate Ruddlesden-Popper (RP) compounds Sr_{n+1}IrₙO_{3n+1} = [SrIrO₃]ₙ, SrO essentially consist of n SrIrO₃ perovskite layers, intercalated by SrO layers and laterally shifted against each other such that no Ir–O–Ir bonds persist between neighboring [SrIrO₃]ₙ blocks [9]. As one veers away from the quasi-2D limit (Sr₂IrO₄, n = 1) the Mott-insulating state breaks down as evidenced by the narrow-gap bilayer system Sr₂Ir₂O₅ hosting a collinear antiferromagnetic order [10,11] and the three-dimensional (n = ∞) semimetallic SrIrO₃ [12]. However, the instability of bulk Sr_{n+1}IrₙO_{3n+1} for n > 2 impedes a systematic investigation of the dimensionality-induced metal-insulator-transition (MIT). In an attempt to mimic the layered RP structure, a concurrent metal-insulator and magnetic transition has been observed in artificially tailored ([SrIrO₃]ₙ, SrTiO₃) superlattices, where the intercalated SrO layers were substituted with SrTiO₃ monolayers [13]. Noticeably, the different magnetic orders found in the bilayer analogues reflect the persisting non-negligible coupling between neighboring bilayers across the SrO/SrTiO₃ blocking layers. Indeed, both magnetism and resistivity have recently been shown to be affected by pronounced interlayer coupling in (SrIrO₃, [SrTiO₃]ₖ) superlattices [14].

In this Letter, we investigate the electronic and structural properties of epitaxially grown ultrathin perovskite SrIrO₃ films, which represent a considerably cleaner (or: better defined) approach to the two-dimensional limit than the superlattices. We observe the opening of a distinct charge gap at the chemical potential and concurrent changes in the film crystalline structure as a function of the film thickness. In a combined experimental and theoretical approach using soft-x-ray angle-resolved photoelectron spectroscopy (SX ARPES) and ab initio density functional theory (DFT and DFT + U) calculations we investigate the evolution of the electronic band structure across the film thickness-driven MIT. We thereby elucidate the complex, microscopic interplay between electronic correlations, structural degrees of freedom, magnetism, and dimensionality.

SrIrO₃ thin films were heteroepitaxially grown on TiO₂-terminated SrTiO₃ (001) substrates by pulsed laser deposition (PLD) from a polycrystalline SrIrO₃ target. The films adopt a pseudotetragonal perovskite structure with an in-plane lattice constant locked to that of SrTiO₃ (a = 3.905 Å) and an out-of-plane lattice constant of...
3.99 Å. Because of collective rotations of the IrO$_6$ octahedra ($a^+b^-b^-$ in Glazer notation with the $a$ axis along the [100] or [010] direction of the substrate [15,16]) the real-space unit cell is enlarged by $2 \times \sqrt{2} \times \sqrt{2}$ with respect to the tetragonal unit cell (for a thorough structural characterization, see the Supplemental Material [17]). SX-ARPES measurements were performed at 20 K at the ADRESS beam line of the Swiss Light Source, Paul Scherrer Institute [26,27]. Density functional theory calculations were performed by using the VASP ab initio simulation package [28] within the projector-augmented-plane-wave (PAW) method [29,30], using the generalized gradient approximation (GGA) as parametrized by the Perdew-Burke-Ernzerhof GGA functional [31]. Spin-orbit coupling was self-consistently included [32] and the Coulomb repulsion and exchange interaction $J$ of Ir $d$ orbitals were treated within the rotationally invariant DFT $+ U$ scheme of Liechtenstein, Anisimov, and Zaanen [33].

Figure 1 shows the experimental and theoretical band structures obtained from SX ARPES on a 9-unit-cell-(u.c.) thick metallic SrIrO$_3$ film and paramagnetic DFT(+U) calculations for the bulk material. As the starting point for the analysis of the photoemission data we consider a simplified tetragonal perovskite structure compressively strained to the SrTiO$_3$ substrate $(a_1/a_{STO} \approx 1.02)$ as depicted in Figs. 1(a) and 1(b). We investigate the combined effect of on-site Coulomb repulsion $U$ and exchange-coupling $J$ on the DFT(+U) band structure and Fermi surface topology. The resulting bands are projected onto a $J_{\text{eff}} = (1/2, 3/2)$ basis [1]. As seen in Figs. 1(c) and 1(e) DFT calculations already capture some of the low-energy spectral features. However, most noteworthy, they predict an Ir 5$d$ $e_y$ electron pocket around $\Gamma$ and an Ir $J_{\text{eff}} = 3/2$ hole pocket around the $A$ point, which are not seen in experiment. A way to reduce the discrepancy is to include short range Coulomb repulsion. Rather than looking for the lowest-energy solution within DFT + $U$, which would be a magnetic insulator at variance with experiment, we discard magnetism for this 9 u.c. film. In this framework, $U$ mainly acts to shift orbitals with different occupations relative to each other. We chose its value to match the position of the ARPES bands [17], thereby pushing the bands either above or below the chemical potential, whereupon the $e_y$ electrons are being predominantly transferred into the $J_{\text{eff}} = 3/2$ band, leaving the $J_{\text{eff}} = 1/2$ Luttinger volume relatively unchanged. It does not come as a surprise that the resulting values of $U$ (3.4) and $J$ (0.4 eV) are significantly larger than $ab$ initio estimates using the constraint random phase approximation (cRPA) [35,36]. A more accurate treatment of the many-body processes based on dynamical mean field theory (DMFT) would most likely account for the experimental features with smaller values of the interaction (for DMFT studies for Sr$_2$IrO$_4$ see Refs. [37,38]).

Despite the overall good agreement, a closer inspection of the DFT + $U$ band structure reveals subtle differences to the experimental data. In particular, the narrow band at the chemical potential between the $X$ and $M$ point and the spectral weight near the $R$ point are not captured in the DFT + $U$ calculations in Fig. 1(d). Indeed, previous ARPES studies using ultraviolet light have reported such narrow bands as a result of backfolding due to octahedral rotations [16], which introduce a periodic perturbation of

![Diagram](Image)

FIG. 1. (a) Real and (b) reciprocal space structure of strained, tetragonal SrIrO$_3$ without octahedral rotations [34]. (c),(d) $E$ vs $k$ dispersions along the high-symmetry lines $\Gamma - X - M - \Gamma$ and $Z - R - A - Z$ measured by SX ARPES ($h\nu = 745$ eV and $h\nu = 660$ eV, respectively) and compared to DFT + $U$ calculations. The band structure was calculated for the tetragonal setting and projected onto a $J_{\text{eff}} = (1/2, 3/2)$ basis with (c) $U = 0$ and $J = 0$ eV and (d) $U = 3.4$ and $J = 0.4$ eV. The introduction of a sizable on-site Coulomb repulsion significantly enhances the agreement between theory and experimental results. (e),(f) Fermi surface topology without (e) and with (f) on-site Coulomb repulsion $U$ and exchange coupling $J$. 

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setting and unfolded into the tetragonal Brillouin zone. The weighted, unfolded bands have a narrow

\[ E_{F} (eV) \]

\[ 0 \]

\[ 0.75 \]

\[ \frac{\varepsilon_{p}(k_{B}, E)}{\hbar^2} + \frac{\varepsilon_{s}(k_{B}, E)}{\hbar^2} \]

\[ \Gamma - X - M - \Gamma \]

\[ \Gamma - R - A - \Gamma \]

\[ \Gamma - Z - R - A - Z \]

\[ \Gamma - X - \Gamma \]

\[ \Gamma - Z - \Gamma \]

\[ \Gamma \]

\[ X \]

\[ M \]

\[ Z \]

\[ R \]

\[ (a) \]

\[ (b) \]

\[ (c) \]

FIG. 2. (a) Real-space lattice structure of SrIrO\(_{3}\) including octahedral rotations (\(a^+b^-b^-\) in Glazer notation with the \(a\) axis orthogonal to the film surface normal) and strain. The orthorhombic unit cell (blue) is enlarged by \(2 \times \sqrt{2} \times \sqrt{2}\) with respect to the tetragonal unit cell (black). (b) Reciprocal space structure of the orthorhombic (blue) and tetragonal (black) structure. (c) SX-ARPES band maps along the pseudotetragonal high-symmetry lines \(\Gamma - X - M - \Gamma\) and \(Z - R - A - Z\) in comparison to DFT + \(U\) band structure calculated in the orthorhombic setting and unfolded into the tetragonal Brillouin zone.

the crystal potential that enlarges (reduces) the real-space unit cell (Brillouin zone) as depicted in Figs. 2(a) and 2(b). In Fig. 2(c) we present the corresponding band structure unfolded into the original Brillouin zone, thereby taking into account the effect of this weak symmetry breaking by calculating the proper spectral weight distribution as described in Ref. [39] (represented by the size of the gray dots). The weighted, unfolded bands have a narrow bandwidth of \(\approx 400\) meV and resolve the aforementioned discrepancies between experiment and theory by exhibiting Fermi crossings around the \(X\) and \(R\) point. Note that the seemingly more pronounced backfolded band structure in ARPES measurements using He I light [16,40] may be due to different matrix elements and/or the inherently higher surface sensitivity as compared to photoemission in the soft-x-ray regime [41,42].

Epitaxially stabilized SrIrO\(_{3}\) thin films essentially exhibit bulk electronic and structural properties above a thickness of at least \(9\) u.c., i.e., paramagnetism and metallicity with a partially filled \(J_{eff} = 1/2\) band. Figure 3(a) shows photoemission spectra (He I, \(h\nu = 21.2\) eV) of SrIrO\(_{3}\) films of smaller thicknesses with \(m = 4, 3, 2, 1, 0\) u.c. (bare Nb: SrTiO\(_{3}\)). As expected in the three-dimensional limit thick films (\(m \geq 4\)) exhibit a metallic density of states with a pronounced Fermi-Dirac cutoff at the chemical potential. Intriguingly, at \(m = 3\) the Fermi cutoff disappears and upon further reduction of the film thickness a distinct charge gap opens. Hence, in analogy to the RP iridates the films undergo a MIT transition as a function of dimensionality as also observed in transport measurements [43]. As shown in the inset of Fig. 3(a) magnetic DFT + \(U\) calculations for \(m\) SrIrO\(_{3}\) layers on 4 SrTiO\(_{3}\) layers (denoted by \(m//4\)) similarly show a decreasing charge gap in the \(k\)-integrated density of states (DOS) as \(m\) is increased. Note, however, that in the presence of magnetic ordering the increasing film thickness alone does not trigger a transition from insulating to metallic in our calculations.

The photoemission gap opening is accompanied by a structural transition as inferred from low energy (LEED) and reflection high-energy electron diffraction (RHEED) from the film surface. As seen in Fig. 3(b), the surface periodicity, which reflects the complex rotational pattern of
IrO$_6$ octahedra within the film, changes at the threshold thickness. In atomically thin, insulating films only rotations about the surface normal prevail, whereas rotations about the in-plane directions are suppressed due to the corner-shared octahedral network between SrIrO$_3$ and the SrTiO$_3$ substrate [Fig. 3(c)], which is a cubic perovskite without octahedral rotations at room temperature. Analogously, the nonmetallic RP iridates Sr$_2$IrO$_4$ and Sr$_3$Ir$_2$O$_7$ exclusively exhibit octahedral rotations about the c axis (reducing their space group symmetry from $I4/mmm$ to $I4_1/acd$ and $Bbcb$, respectively) [11,44–46].

Figure 4(a) shows the SX-ARPES band map of a 1 u.c. SrIrO$_3$ film grown on Nb: SrTiO$_3$ in comparison to the DFT + $U$ band structure of a 1/4 SrIrO$_3$//SrTiO$_3$ slab. In excellent agreement with each other the experimental and theoretical data exhibit weakly dispersing bands with a good agreement with the experimental data exhibit weakly dispersing bands with excellent agreement with each other the experimental and theoretical data (smaller $W$) [36]. Second, the crystalline structure due to the IrO$_6$ rotations deviates from the rotational pattern of bulk SrIrO$_3$ in the two-dimensional limit, since the octahedral network with the cubic SrTiO$_3$ substrate imposes constraints upon the in-plane rotations. Finally, the onset of an insulating state is associated with magnetic correlations. The actual stabilization of a long-range ordered magnet is a very delicate issue, due to the sensitivity to the dimensionality and to the competition between in- and out-of-plane coupling [11]. The strong cooperative interplay between these degrees of freedom constitutes the complexity of the system. Specifically, octahedral rotations strongly affect the magnetic coupling in iridates due to pseudodipolar and Dzyaloshinskii-Moriya exchange interactions as evidenced by the locking of the Ir magnetic moments to the rotated oxygen octahedra [48]. In turn, the symmetry breaking due to octahedral rotations provides further spin-dependent hopping terms in the $J_{eff}$ basis that additionally increase the kinetic energy $W$ [49].

**FIG. 4.** (a) SX-ARPES band map of a SrIrO$_3$ monolayer grown on Nb: SrTiO$_3$ and DFT + $U$ band structure of a 1/4 SrIrO$_3$//SrTiO$_3$ slab along the pseudotetragonal high-symmetry line $\Gamma - X - M - \Gamma$. (b) DFT + $U$ $k$-integrated density of states (DOS) of the 1/4 slab. The antiferromagnetic solution exhibits a charge gap of $\approx 1$ eV, while the nonmagnetic solution is in a metallic state. (c) DFT + $U$ band structure calculation for bulk Sr$_2$IrO$_4$. 

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tendency is reflected in the (albeit small) resistivity drop at $T = 105$ K [50], where the SrTiO$_3$ substrate undergoes a structural transition involving $d^0d^0e^−$-rotations of the TiO$_6$ octahedra [51,52], which can induce in-plane tiltings in the SrIrO$_3$ film. On the other hand, changes in $U/W$ will affect the magnetic ordering by altering the required critical onsite Coulomb repulsion $U$ for the transition to an antiferromagnetic ground state [49].

The underlying reason for this extraordinary dimensionality dependence is the spatially three-dimensional $j_{\text{eff}} = 1/2$ Kramers doublet wave function, which results from the mixing of orbitals of different symmetries with the spin degrees of freedom due to the strong spin-orbit coupling in $5d$ systems. This is in stark contrast to typical $3d$ systems like the cuprates, where the planar $e_g$ orbitals host the $S = 1/2$ magnetic moments in the absence of strong spin-orbit coupling. With regard to Sr$_3$IrO$_4$ as a potential parent material for exotic superconductivity, the analogy between monolayer SrIrO$_3$ and bulk Sr$_3$IrO$_4$ may open a promising experimental avenue towards electron doping without the introduction of disorder through electrostatic or ion-liquid gating, possibly pushing the system into a novel, spin-orbit-driven superconducting phase.

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[42] Recently, a Dirac-like feature in the band structure of SrIrO$_3$ has been discussed in the literature. For more details related to our present work, see Supplemental Material, Sec. VIII.