Emergent interlayer magnetic order via strain-induced orthorhombic distortion in the 5*d* Mott insulator Sr₂IrO₄

S. Shrestha¹, M. Krautloher², M. Zhu³, J. Kim¹, J. Hwang³, J. Kim⁴, J.-W. Kim⁴, B. Keimer², and A. Seo¹

¹Department of Physics and Astronomy, University of Kentucky, Lexington, KY 40506, USA

²Max-Planck-Institut für Festkörperforschung, D-70569 Stuttgart, Germany ³Department of Materials Science and Engineering, The Ohio State University, Columbus, Ohio 43210, USA

⁴Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA

Abstract

We report a La₂CuO₄-like interlayer antiferromagnetic order in Sr₂IrO₄ films with large orthorhombic distortion (> 1.5%). The biaxial lattice strain in epitaxial heterostructures of Sr₂IrO₄/Ca₃Ru₂O₇ lowers the crystal symmetry of Sr₂IrO₄ from tetragonal (*C*₄) to orthorhombic (*C*₂), guiding the Ir 5*d J*_{eff} = 1/2 pseudospin moment parallel to the elongated *b*-axis via magnetic anisotropy. From resonant X-ray scattering experiments, we observed an antiferromagnetic order in the orthorhombic Sr₂IrO₄ film whose interlayer stacking pattern is inverted from that of the tetragonal Sr₂IrO₄ crystal. This interlayer stacking is similar to that of the orthorhombic La₂CuO₄, implying that the asymmetric interlayer exchange interactions between *a*- and *b*-directions exceed the anisotropic interlayer pseudo-dipolar interaction. Our result suggests that strain-induced distortion can provide a delicate knob for tuning the long-range magnetic order in quasi-twodimensional systems by evoking the competition between the interlayer exchange coupling and the pseudo-dipolar interaction. Transition metal oxides are a featured condensed matter system for realizing emergent electronic and magnetic phases due to competing interactions and strong electron correlation. Layered oxides with the quasi-two-dimensional K₂NiF₄-type structure are of particular interest since they include superconducting high- T_c cuprates [1,2], spin/charge/orbital ordered manganates [3], spin-orbit coupled iridates [4,5], and so forth. Despite their diverse physical properties, the spin Hamiltonian of the system is commonly described by

$$H_{ij} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \boldsymbol{\Gamma}_{ij} \cdot \mathbf{S}_j \qquad \text{Eq.1}$$

, where the first term is the Heisenberg exchange interaction, the second term is the antisymmetric Dzyaloshinskii-Moriya (DM) interaction, and the third term is the pseudo-dipolar interaction with a symmetric traceless second rank tensor Γ_{ij} , respectively, between two sites *i* and *j* [6]. Because of the quasi-two-dimensional nature of the layered oxides, in-plane interactions are often considered important in theoretical models [7]. This approximation is justified because the Heisenberg exchange interaction within the basal plane is a few orders of magnitude stronger than any interlayer interactions. Nevertheless, it is noteworthy that many layered-oxides exhibit long-range magnetic order with well-defined Néel temperature (T_N), which is not allowed in an ideal 2D Heisenberg spin system with continuous symmetry [8]. The interlayer long-range magnetic order is determined by interlayer Hamiltonian defined as:

$$H_{\langle i,j \rangle //a} = J^{a}_{out} \left(\mathbf{S}_{i} \cdot \mathbf{S}_{j} \right) + \Gamma^{a}_{out} \left(\mathbf{S}_{i}^{a} \mathbf{S}_{j}^{a} - \mathbf{S}_{i}^{b} \mathbf{S}_{j}^{b} \right)$$
$$H_{\langle i,j \rangle //b} = J^{b}_{out} \left(\mathbf{S}_{i} \cdot \mathbf{S}_{j} \right) + \Gamma^{b}_{out} \left(\mathbf{S}_{i}^{b} \mathbf{S}_{j}^{b} - \mathbf{S}_{i}^{a} \mathbf{S}_{j}^{a} \right)$$
Eq.2

, where J_{out}^a and J_{out}^b are the nearest interlayer exchange interactions and Γ_{out}^a and Γ_{out}^b are the anisotropic interlayer pseudo-dipolar interactions along the crystallographic *a*- and *b*-directions,

respectively, as shown in Fig 1(a). We define j_c and δ_c as the energies from the nearest interlayer exchange interaction and the anisotropic interlayer pseudo-dipolar interaction, respectively:

$$j_{c} \equiv J_{out}^{a} \left(\mathbf{S}_{i} \cdot \mathbf{S}_{j} \right) + J_{out}^{b} \left(\mathbf{S}_{i} \cdot \mathbf{S}_{j} \right), \qquad \delta_{c} \equiv \Gamma_{out}^{a} \left(\mathbf{S}_{i}^{a} \mathbf{S}_{j}^{a} - \mathbf{S}_{i}^{b} \mathbf{S}_{j}^{b} \right) + \Gamma_{out}^{b} \left(\mathbf{S}_{i}^{b} \mathbf{S}_{j}^{b} - \mathbf{S}_{i}^{a} \mathbf{S}_{j}^{a} \right). \qquad \text{Eq.3}$$

Two distinct antiferromagnetic (AFM) interlayer stacking patterns are found in the K₂NiF₄type oxides depending on j_c and δ_c . For example, tetragonal La₂NiO₄ and Sr₂IrO₄ crystals show the AFM-1 type [9-13] whereas orthorhombic La₂CuO₄ crystals exhibit the AFM-2 type [14-17], as shown in Fig. 1(b). La₂CoO₄ crystals present both types depending upon their phase transitions between tetragonal and orthorhombic structures [18]. On the other hand, by applying external stimuli such as pressure, the frustrated interlayer interaction leads to the suppression of the interlayer magnetic order, enhancing the quantum fluctuation that can open a path to quantum paramagnetic phases [19].



Fig 1. (a) A schematic crystal structure of K₂NiF₄-type oxides. The interlayer exchange interactions J_{out}^a and J_{out}^b share the same interaction paths with the interlayer pseudo-dipolar interactions Γ_{out}^a and Γ_{out}^b , respectively. (b) Schematic diagrams of two different long-range antiferromagnetic (AFM) stacking patterns in the K₂NiF₄-type oxides. The AFM-1 type is observed in tetragonal Sr₂IrO₄ and La₂NiO₄ crystals whereas the AFM-2 type is observed in orthorhombic La₂CuO₄ crystals.

In the tetragonal structure, the AFM stacking order is correlated with anisotropic interlayer pseudo-dipolar interaction since the interlayer exchange interaction cancels out, i.e., AFM-1 for δ_c < 0 and AFM-2 for δ_c > 0 [12]. On the other hand, the interlayer exchange interaction is also an important factor to determine the stacking order in the orthorhombic structure. Therefore, one may conjecture that AFM-2 type order can be induced when orthorhombic distortion is induced in a tetragonal system. However, recent experiments by Kim *et al.* showed that the AFM-1 type order remained in the Sr₂IrO₄ crystals even under orthorhombic distortion using a piezoelectric strain device [20]. Hence, the interlayer AFM stacking order would be irrelevant to the crystal structure or the magnitude of the orthorhombic distortion (~0.03%) was not large enough to stabilize the AFM-2 type stacking pattern in the Sr₂IrO₄ crystals. It is formidable to figure out which one is correct because conventional strain/stress device approaches using bulk crystals cannot exert much larger external pressure than the magnitude used in the studies of Ref. [20].

In this *Letter*, we report that the long-range AFM-2 stacking order emerges in the layered iridate Sr₂IrO₄ thin films when substantially large (> 1.5%) orthorhombic distortion is applied by biaxial strain. Such a large magnitude of orthorhombic distortion is realized by constructing epitaxial thin film heterostructures of Sr₂IrO₄/Ca₃Ru₂O₇ with excellent structural coherence and atomically sharp interfaces. We observed clear magnetic Bragg reflections from the Ir L-edge resonant X-ray scattering (RXS) experiments, indicating that the AFM-2 type stacking order is stabilized when $|j_e|$ exceeds $|\delta_e|$. Our result confirms that the long-range AFM stacking order of the quasi-two-dimensional oxides is correlated with their crystal structures. It is noteworthy that our experimental finding provides a useful approach for enhancing the magnetic frustrations (i.e., magnetic quantum fluctuations) of various two-dimensional magnetic systems via utilizing the subtle competition between the interlayer exchange interaction and the pseudo-dipolar interaction.



Fig 2. (a) A schematic diagram of a Sr₂IrO₄/Ca₃Ru₂O₇ heterostructure. The red and blue arrows represent compressive and tensile strain directions and magnitudes. **(b)** X-ray diffraction (0 0 L) scan of a Sr₂IrO₄/Ca₃Ru₂O₇ heterostructure. The (0 0 12) and (0 0 18) peaks from the Sr₂IrO₄ thin film are visible. The asterisks [*] indicate the peaks from the Ca₃Ru₂O₇ single crystal substrate. **(c)** High-resolution Z-contrast STEM images of a Sr₂IrO₄/Ca₃Ru₂O₇ heterostructure for two different cross-sectional directions. The red triangles mark the atomically sharp interface between Sr₂IrO₄ and Ca₃Ru₂O₇. The scale bar is 1 nm.

We conceived $Sr_2IrO_4/Ca_3Ru_2O_7$ heterostructures to study the effect of orthorhombic distortion to the AFM order because of the large anisotropic lattice mismatch between the tetragonal Sr_2IrO_4 and the orthorhombic $Ca_3Ru_2O_7$, i.e., -2.22% and +0.45% along the *a*- and *b*-directions, respectively, as shown in Fig. 2(a). The heterostructure samples were synthesized by depositing 24-nm-thick Sr_2IrO_4 epitaxial thin-films on $Ca_3Ru_2O_7$ (001) single crystals with cleaved surfaces using pulsed laser deposition (PLD). The PLD conditions are a laser fluence of 1.2 J/cm², a substrate temperature of 700°C, and an oxygen partial pressure of 10 mTorr, which are consistent with Ref. [21]. Figure 2(b) shows X-ray diffraction with well-defined (0 0 l) peaks and Kiessig fringes from a Sr₂IrO₄/Ca₃Ru₂O₇ heterostructure, indicating a high-quality thin film with a thickness of ~24 nm. The structural coherence length of ~29 nm, obtained from the FWHM of the (0 0 20) peak is comparable to the thickness of the Sr₂IrO₄ thin film (Fig. S1(c)). The H and K scans near (2 0 20) and (0 2 20)-reflections show that the Sr₂IrO₄ thin films are fully strained along both in-plane directions (Figs. S1(a) and (b)). Large in-plane anisotropy with -2.22% compressive strain along the *a*-axis and +0.45% tensile strain along the *b*-axis is observed in these thin films (Table I) which correspond to the significantly large orthorhombic distortion ($\alpha \equiv (b-a)/(b+a)$) of +1.58% at low temperature (6 K). Considering Young's modulus of ~130 GPa of Sr₂IrO₄ crystals [22], this compressive strain corresponds to an effective uniaxial pressure of ~ 2.9 GPa along the *a*-axis, which is far above the accessible range of conventional strain/stress approaches [23]. This anisotropic strain is expected to lower the crystal symmetry of Sr₂IrO₄ from tetragonal to orthorhombic, as shown in Fig. 2(a). Structural Bragg peaks of (1 2 19) and (1 2 21) that are relevant to the octahedral rotation along the *c*-axis [24,25] were not observed from our samples (Fig. S1(d)), implying that the octahedral rotational pattern is substantially modified in these Sr₂IrO₄/Ca₃Ru₂O₇ heterostructures presumably due to the large orthorhombic distortion. Figure 2(c) shows high-resolution Z-contrast scanning transmission electron microscopy (STEM) data of the cross-sections of the heterostructures. Note that an atomically sharp interface between Sr₂IrO₄ and Ca₃Ru₂O₇ is observed without any noticeable misfit dislocations or atomic inter-mixing even under such a large orthorhombic distortion.

	a (Å)	<i>b</i> (Å)	c (Å)	α(%) ^a	Eaa (%)	Е <i>bb</i> (%)	Есс (%)	V (Å ³)
Ca ₃ Ru ₂ O ₇ substrate 8 K [26]	5.367	5.536	19.521					
Ca3Ru2O7 substrate 296 K [26]	5.378	5.523	19.587					
Sr2IrO4 thin films 6 K	5.364(4)	5.536(4)	25.92(2)	+1.58	-2.21(7)	+0.93(7)	+0.47(8)	769.9
Sr2IrO4 thin films 296 K	5.378(4)	5.523(4)	25.94(2)	+1.33	-2.22(9)	+0.44(8)	+0.61(8)	770.5

Table I: Lattice constants and strain values of Sr₂IrO₄ (001) epitaxial thin films grown on Ca₃Ru₂O₇ (001) single crystals.

^aValues are obtained using $\alpha \equiv (b-a) / (b+a) \times 100(\%)$



Fig 3. (a) Normalized integrated intensities of the Ir L-edge (1 0 24) magnetic Bragg peaks of a Sr₂IrO₄/Ca₃Ru₂O₇ heterostructure as a function of azimuthal angle (Ψ) with σ - π polarization channel. The solid (red) line represents a theoretical calculation for the $J_{eff} = \frac{1}{2}$ pseudospin magnetic moment (\vec{m}) parallel to the crystallographic *b*-axis, as shown in the schematic diagram. $\Psi = 0$ is defined as the crystallographic *a*-axis lies in the scattering plane. The inset shows the same magnetic Bragg peak intensities at selected azimuthal angles. (b) Normalized integrated intensities of the (1 0 24) magnetic Bragg peaks as a function of temperature. The solid (red) line is a power-law fit using equation $I \propto [1 - \frac{T}{T_N}]^{2\beta}$, where T_N and β are the Néel temperature (~222 K) and the critical exponent, respectively. (Inset) Normalized integrated intensity vs. the reduced temperature ($1 - \frac{T}{T_N}$) in the logarithmic scale. The estimated critical exponent $\beta = 0.22$ is below 0.23 that corresponds to the 2D XY*h*₄ universality model [33, 34].

Resonant X-ray scattering experiments reveal an unprecedented AFM-2 stacking order, which has not been observed from any Sr₂IrO₄ crystals or thin films. Using resonant X-ray near the Ir L₃ edge ($\hbar\omega = 11.217$ keV as shown in Fig. S3(f)), we observed a clear (1 0 24) magnetic Bragg peak at 6 K with a magnetic coherence length of ~ 26 nm along the out-of-plane direction (Fig. S3(b)), comparable to the out-of-plane structural coherence length of ~29 nm (Fig. S1(c)). Figure 3(a) shows the azimuthal angle (Ψ) dependence of the (1 0 24) magnetic Bragg peak (see Fig. S4(a)) in which the sample is rotated about the scattering vector with $\Psi = 0$ defined such that the crystallographic *a*-axis lies in the scattering plane as shown in Fig. S3(e). We used the $\sigma - \pi$ polarization channel of the incoming and outgoing X-rays because the magnetic Bragg peak intensities follow the relation $I \propto |\vec{m}| (\sigma_i \times \sigma'_0)|^2$, where σ_i and σ'_0 are the incoming and outgoing polarizations of photons and \vec{m} is the $J_{eff} = 1/2$ pseudo-spin magnetic moment [27,28]. We observed that the azimuthal angle dependence of the integrated peak intensity, obtained by fitting the Gaussian peak shape of L-scans, follows the theoretical calculation for $\vec{m} \not | \vec{b}$. This observation indicates that the magnetic moment is along the crystallographic *b*-axis without any secondary magnetic domains of $\vec{m} /\!\!/ \vec{a}$. Note that this observation of $\vec{m} /\!\!/ \vec{b}$ is consistent with the result of Ref. [20], in which the orthorhombic distortion of ~0.03% is applied using uniaxial compressive strain along the *a*-axis of a Sr₂IrO₄ single crystal. These two results confirm that the uniaxial magnetic anisotropy due to orthorhombic distortion is crucial in determining the direction of the magnetic moment with respect to the crystallographic axis [29].

Figure 3(b) shows that the integrated intensity of the magnetic Bragg peak (see Fig. S4(b)) decreases with increasing temperature, suggesting the long-range AFM order below $T_N = 222$ K. Note that this value of T_N is comparable to that of Sr₂IrO₄ single crystals (< 230 K) [30-32] despite the orthorhombic distortion. The integrated intensity follows the power-law scaling function $I(t) \approx |t|^{2\beta}$ with the critical exponent $\beta = 0.22$, where $t \equiv 1 - \frac{T}{T_N}$ is the reduced temperature, as shown in the inset of Fig. 3(b). The critical exponent (0.22), corresponding to the 2D XY*h*₄ universality class similar to the bulk crystal, seems reasonable for this quasi-2D system with weak interlayer coupling [33-35].

Full (0 1 L) and (1 0 L) scans over the wide range of L from 15 to 25 reciprocal lattice units show clear (1 0 4*n*) and (0 1 4*n*+2) magnetic Bragg peaks with n = 4, 5, and 6, as shown in Fig. 4(a). Additional strong structural Bragg reflections from the Ca₃Ru₂O₇ crystal are indicated by asterisks (*). Note that these magnetic Bragg peaks, i.e., (1 0 4*n*) and (0 1 4*n*+2) with $\vec{m} // \vec{b}$, are the ones expected in the AFM-2 type stacking order [12] which is observed in orthorhombic systems such as La₂CuO₄ [14-16]. In tetragonal Sr₂IrO₄ crystals, the AFM-2 type stacking is energetically $2|\delta_c|$ higher than the AFM-1 type stacking, i.e., (1 0 4*n*+2) and (0 1 4*n*) with $\vec{m} // \vec{b}$ [12].



Fig 4. (a) Ir L-edge X-ray (1 0 L) and (0 1 L) scans of a Sr₂IrO₄/Ca₃Ru₂O₇ heterostructure at 6 K. (1 0 16), (1 0 20), (1 0 24), (0 1 18), and (0 1 22) magnetic Bragg peaks are observed. The asterisks [*] indicate the peaks from the Ca₃Ru₂O₇ single crystal substrate. **(b)** A schematic phase diagram of AFM stacking orders of Sr₂IrO₄ as a function of orthorhombic distortion. When $\alpha = 0$, i.e., a tetragonal structure, equivalent twin domains (AFM-1) can coexist. With increasing the orthorhombic distortion (α) up to the critical point (α_c), the magnetic domains are just de-twined. With larger orthorhombic distortion than α_c , the AFM-2 type stacking order is stabilized, exhibiting the magnetic moment parallel to the *b*-axis and the magnetic Bragg peaks of (1 0 4*n*) and (0 1 4*n*+2).

Our experimentally finding not only confirms that the interlayer AFM order is correlated with crystal symmetry but also leads us to further understand that the competition between j_c and δ_c in determining the long-range AFM order in the system, as schematically shown in Fig. 4(b):

- 1) The orthorhombic distortion (α) = 0, tetragonal Sr₂IrO₄ crystals have isotropic J_{out} , i.e., $J_{out}^{a} = J_{out}^{b}$, resulting in $j_{c} = 0$ for a G-type AFM order. Two equivalent AFM domains with the AFM-1 type stacking order with $\vec{m} /\!\!/ \vec{a}$ and $\vec{m} /\!\!/ \vec{b}$ are stabilized with the energy of δ_{c} < 0. They correspond to *uudd* and *lrrl* stacking order discussed in Ref. [12].
- 0 < α < α_c, where α_c is a critical value, as the tetragonal structure of Sr₂IrO₄ becomes an orthorhombic structure, the AFM-1 type order with m // b is stabilized over the other phase with m // a due to the pseudospin-lattice interaction within the IrO₂ basal planes. This transition was experimentally observed with α ≈ 0.03% [20]. J_{out} is no longer isotropic, i.e., J^a_{out} > J^b_{out}, resulting in non-zero j_c. Nevertheless, the interlayer pseudo-dipolar interaction remains dominant since |j_c| < |δ_c|.
- 3) α > α_c, the interlayer exchange interaction becomes dominant over the pseudo-dipolar interaction, i.e., j_c + δ_c < 0 with |j_c| > |δ_c|, making the AFM-2 type stacking order stabilized. Using the tight-binding approximation, we can estimate the value of j_c to be around -0.9 μeV (α = 1.6%), whose magnitude is larger than the estimated value of δ_c = 0.3 μeV.
- 4) $\alpha \approx \alpha_c$, both AFM-1 and AFM-2 type stacking orders can coexist due to $|j_c| \approx |\delta_c|$, meaning that the system has a large interlayer magnetic frustration and consequently enhances the magnetic quantum fluctuation. By assuming $\delta_c = 0.3 \ \mu eV$ (Ref. [12]) and linear extrapolation between our experimental values, α_c is estimated to be around +0.7%. It would be a formidable task to experimentally study the interlayer magnetic frustration near

the critical value. However, applying a gradual strain to obtain continuous orthorhombic distortion in freestanding nanomembranes [36] could provide an accessible approach.

Our result suggests that the competition between interlayer exchange interaction and pseudodipolar interaction provides an intriguing tuning parameter to stabilize the long-range magnetic stacking order in quasi-2D systems. We have observed a distinct La₂CuO₄-like AFM stacking order in Sr₂IrO₄ by enhancing $|j_c|$ over $|\delta_c|$ via large orthorhombic distortion. In addition to opening exciting questions on the detailed nature of the competition between interlayer interactions, these results raise the prospects of tuning the anisotropic coupling between spins into exotic quantum phases.

Acknowledgment

We acknowledge the support of National Science Foundation Grants No. DMR-1454200, DMR-2011876, and DMR-2104296 for sample synthesis and characterization. This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility, operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357. Electron microscopy was performed at the Center for Electron Microscopy and Analysis at the Ohio State University. B.K. acknowledges financial support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – Projektnummer 107745057 - TRR 80.

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