Spin-orbit coupled double perovskite bilayers: Magnetism, Chern bands, and quantum anomalous Hall insulators on the honeycomb lattice

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Recent experiments have demonstrated the controlled layer-by-layer growth of oxide heterostructures. This leads to the exciting prospect of tuning magnetism and topological states of correlated electrons in low dimensions. Here, we model \{111\}-grown bilayers of half-metallic double perovskites such as Sr\(_2\)FeMoO\(_6\), which form a buckled honeycomb lattice, with a triangular lattice of Fe moments interacting with spin-orbit coupled Mo electronic \(t_{2g}\) states. The combination of spin-orbit coupling, inter-orbital hybridization and symmetry-allowed trigonal distortion leads to a rich phase diagram with tunable ferromagnetic order, topological \(C = \pm 1, \pm 2\) Chern bands, and a \(C = \pm 2\) quantum anomalous Hall insulator regime. An effective two-band model of Zeeman-split \(j = 3/2\) states captures this emergence of \(C = \pm 2\) band topology.

**Introduction.** — The discovery of two-dimensional (2D) tunable and conducting electronic states at transition metal oxide interfaces \[^1\] holds great promise for oxide electronics \[^2\ ?,^3\]. The rapid experimental advances in oxide heterostructures \[^4\ ?,^8\] have led to the finding that these interface states host coexisting superconductivity and magnetism driven by electronic correlations \[^9\ ?,^11\]. Transport studies on epitaxial thin films of 5d transition metal oxides with strong spin-orbit coupling have revealed strain as a powerful tool to control their electronic properties \[^12\ ?,^13\]. These experiments have motivated significant work on understanding the interplay of correlations, quantum confinement, and spin orbit coupling in transition metal oxides such as cubic perovskites, pyrochlores, and oxide interfaces \[^14\ ?,^27\].

Recently, (LaNiO\(_3\))\(_m\)-(LaMnO\(_3\))\(_n\) oxide superlattices have been grown along the \{111\} direction \[^28\], and found to display an unusual exchange bias effect. The infinite \{111\} superlattice with alternating triangular layers of Ni and Mn ions corresponds to a “double” perovskite La\(_2\)NiMnO\(_6\). Such double perovskites (DPs) are complex oxides, A\(_2\)BB'O\(_6\), with transition metal ions B and B' residing on the two sublattices of a 3D cubic lattice \[^29\] as shown in Fig. 1(a). Theoretical calculations \[^30\] suggest multiferroic behavior in the series R\(_2\)NiMnO\(_6\) (R being a rare-earth element). Other important bulk DP materials include Sr\(_2\)FeMoO\(_6\) (SFMO) \[^31\], Ba\(_2\)FeReO\(_6\), and Sr\(_2\)FeReO\(_6\) \[^32\ ?,^33\] which support half-metallic ferrimagnetism \[^31\ ?,^33\] with potential for spintronic applications \[^44\ ?,^45\] as well as ferromagnetic Mott insulators like Sr\(_2\)CrOsO\(_6\) \[^46\ ?,^47\].

Motivated by these developments, we study the magnetism and spin-orbit coupled electronic bands in a \{111\} bilayer film of such DPs sandwiched between inert oxide band insulators, focusing on the 3d/4d material SFMO as a prototypical example. As shown in Fig. 1(b), such a \{111\} DP bilayer has Fe and Mo ions living on the two sublattices of a (buckled) honeycomb lattice. The system consists of spin-orbit coupled \(t_{2g}\) electrons on the triangular lattice formed by Mo, coupled to local moments on the triangular Fe lattice. Our central result is the emergence, in such a bilayer, of tunable \(C = \pm 1, \pm 2\) Chern bands and Chern insulators with a quantized anomalous Hall effect driven by spontaneous kinetic ferrimagnetism of Fe moments.

Our study of the magnetism and electronic states in the SFMO bilayer reveals the following. Among a large variety of magnetically ordered or disordered states we have examined, the ferromagnetically ordered state of the Fe moments has the lowest energy. This is consistent with experimental results on bulk SFMO \[^31\] and theoretical studies of bulk SFMO in the absence of spin-orbit coupling \[^35\]. The magnetic anisotropy arising from electronic energies is governed by the interplay of spin-orbit coupling, interorbital hybridization, and a symmetry-allowed trigonal distortion. Depending on parameters, this interplay is found to favor various orientations of the local moments. For the \{110\} orientation of magnetic order, we find electronic bands with Chern numbers \(C = \pm 1\). For the \{111\} ordered state, with Fe moments perpendicular to the bilayer, we find that the Mo \(t_{2g}\) electrons display bands with Chern numbers \(C = \pm 2\); we present an effective two-band triangular lattice model of

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**FIG. 1:** (a) Crystal structure of Sr\(_2\)FeMoO\(_6\). Arrows depict bulk ferrimagnetic ground state configuration of spins on the Fe and Mo sites. (b) \{111\} view of a bilayer, showing buckled honeycomb lattice with Fe and Mo ions on the two sublattices.
Zeeman-split $j = 3/2$ states which correctly captures the emergence of this nontrivial band topology. These bands have a direct gap, but typically overlap in energy leading to a metallic state. In the presence of a symmetry-allowed trigonal distortion, we find a regime of a $C = \pm 2$ Mn insulator, $CZ$, i.e., a quantum anomalous Hall insulator with a pair of chiral edge modes, having a gap $\sim 75K$.

**Model.** — Strong Hund’s coupling on Fe$^{3+}$ locks the 3d$^9$ electrons into a large $S_F = 5/2$ local moment, which we treat as a classical spin similar to Mn spins in the colossal magnetoresistive manganites [48]. The 4d electron on Mo$^{5+}$ hops on or off Fe, subject to a charge-transfer energy $\Delta$. Pauli exclusion on Fe forces the spin of the arriving electron to be antiparallel to the underlying Fe moment. This leads to kinetically stabilized ferromagnetic order of the Fe moments in bulk SFMO [34, 39]. However, previous work on SFMO has not considered the effect of quantum confinement or spin-orbit coupling on these electronic states. Here, we consider [111] bilayers which confines electrons to a two-dimensional (2D) honeycomb lattice geometry as shown in Fig. 1. The $t_{2g}$ orbitals on Mo act as effective $L=1$ angular momentum states, and experience local spin-orbit coupling $-\lambda \vec{L} \cdot \vec{S}$, with $\lambda > 0$, which should lead to a low energy $j = 3/2$ quartet and a high energy $j = 1/2$ doublet. Finally, the reduced symmetry of the honeycomb bilayer in a thin film grown along [111] permits a trigonal distortion [24] $H_{tri} = \chi \vec{n} \cdot (\vec{L} \times \vec{n})^2$, where $\vec{n}$ is a unit vector perpendicular to the bilayer: $\chi_{tri} > 0$ corresponds to compressing the Mo oxygen octahedral cage [49]. Incorporating these new ingredients, we arrive at the model Hamiltonian

$$H = \sum_{(ij),\ell,\sigma} \left[ t^{ij}_{\ell} g_{\ell}(j) d^{\dagger}_{i\ell \sigma} f_{j\ell} + \text{H.c.} \right] + \Delta \sum_{\ell} f^\dagger_{\ell} f_{\ell} + H_{tri}$$

$$+ \sum_{(ij),\ell,\sigma} \eta^{ij}_{\ell} d^{\dagger}_{i\ell \sigma} d_{j\ell \sigma} + i \frac{\lambda}{2} \sum_{i} \epsilon_{m\ell \sigma} \sigma_{\sigma} d^{\dagger}_{i\ell \sigma} d_{i \ell \sigma}.$$

(1)

Here $d$ ($f$) denotes electrons on Mo (Fe), $i$ labels sites, $\sigma$ is the spin label, $\ell = 1, 2, 3$ ($= yz, zx, xy$) is the orbital index, and $\epsilon$ is the totally antisymmetric tensor. With $F = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ denoting the Fe moment direction, Pauli exclusion leads to a single spin projection [38] (antiparallel to $F$) for electrons on Fe, with $g_r(j) = \frac{1}{2} e^{-i\phi_j/2}$ and $g_t(j) = -\cos \frac{\lambda}{2} e^{i\phi_j/2}$. Matrix elements $t^{ij}_{\ell}$ correspond to intra-orbital Mo-Fe hopings $t_{\ell}, t_{d}$, while $\eta^{ij}_{\ell}$ encodes Mo-Mo intra-orbital hopping amplitudes $t_{\ell}, t_{\ell}'$ and an inter-orbital hopping amplitude $t_m$ (see Supplementary Material for details of hopping processes). Such a model Hamiltonian, with strong spin-orbit coupling and $H_{tri} = 0$, has been previously shown [41] to successfully capture the phenomenology of the ferrimagnetic state of bulk Ba$_2$FeReO$_6$, quantitatively explaining its ab initio band dispersion [40], saturation magnetization [32, 51], the spin and orbital polarizations measured using X-ray magnetic circular dichroism (XMCD) [32], and neutron scattering data [40]. Our model captures the key energy scales in SFMO: (i) the implicit strong Hund’s coupling on Fe ($\sim 2\text{ eV}$), (ii) the Fe-Mo charge transfer energy ($\Delta \sim 0.5\text{ eV}$), (iii) the nearest neighbor intra-orbital Mo-Fe hopping which leads to electron itinerancy ($t_{ij}^\ell \sim 0.25\text{ eV}$), and (iv) the spin-orbit coupling on Mo ($\lambda \sim 0.12\text{ eV}$). Second neighbor intra-orbital and inter-orbital hoppings ($\eta_{ij}^{(\ell)} \sim 0.025\text{ eV}$) are much weaker. However, we retain them since they play a role in pinning the Fe moment direction in the ferrimagnetic state, leading to well-defined Chern bands and a nonzero ferromagnetic $T_c$ in 2D.

**Magnetism in the bilayer.** — The ground state of bulk SFMO is a ferrimagnet. In order to explore the magnetic structure of the [111] SFMO bilayer, we have computed the electronic energies of a wide range of magnetic configurations of Fe moments including (i) ferrimagnetic configurations with different spin orientations, (ii) period-2 stripe-like configurations with different spin and stripe orientations, and (iii) random configurations.

**FIG. 2:** (a) Ground state electronic energy, $E_{GS}$, per Fe spin in units of the Mo-Fe hopping $t_0 = 250\text{ meV}$ for different magnetic configurations of Fe moments including (i) ferrimagnetically ordered, (ii) stripe-like antiferromagnetically ordered, and (iii) random disordered states. For ordered states, the labels indicate the orientations of the magnetic moments. (b) $E_{GS}$ for the ferrimagnetic states plotted for different orientations of the Fe moments. (c) $E_{GS}$ for the ferromagnetic states for Fe moments pointing along high symmetry directions.
With no trigonal distortion, $\chi_{\text{tri}} = 0$, the six $\{110\}$ orientations with Fe moments lying in the bilayer plane have the lowest energy. As seen from Fig. 2(c), the other high symmetry orientations are higher in energy by $\delta E \sim 1\text{meV}$.

We have also explored the effect of trigonal distortion on the energy of different ferromagnetic orientations. For $\chi_{\text{tri}} < 0$, we expect $\hat{L} \parallel \hat{n}$ in order to minimize the energy. This favors the $\{111\}$ orientation of $\hat{L}$, and spin-orbit coupling then forces the spins to also point out of the bilayer plane. For $\chi_{\text{tri}} > 0$, it is energetically favorable to have $\hat{L} \perp \hat{n}$, so the $\{110\}$ orientations remain favorable. We have numerically confirmed these expectations. The combination of spin-orbit coupling and trigonal distortion can thus favor a variety of “Ising” or “clock” ferromagnetic ground states; we thus expect a nonzero magnetic $T_c$ even in the bilayer. For $\{111\}$ ordering, we expect an Ising transition temperature $T_c = \alpha S_F \sqrt{J_{\text{FF}} \delta E}$, where $\delta E$ is the anisotropy energy (i.e., the characteristic cost for rotating spins away from the Ising axis), and we estimate the numerical prefactor $\alpha \sim 4$ (see Supplementary Material for details). Using a typical anisotropy energy $\delta E \sim 0.2\text{meV}$, and setting $S_F = 5/2$ and $J_{\text{FF}} = 1.5\text{meV}$, this yields $T_c \sim 65\text{K}$, although $T_c$ might be much higher than this crude estimate suggests. When the ground state favors one of the six $\{110\}$ orientations, we expect a slightly reduced transition temperature, as well as an intermediate phase with power law magnetic order. These $T_c$ values are substantially lower than $T_c^{\text{bulk}} \sim 400\text{K}$ but still easily accessible in experiments. A full Monte Carlo study of the thermal fluctuation effects and finite temperature transitions will be discussed elsewhere. We next turn to the ground state electronic properties of this SFMO bilayer, focusing on the Berry curvature of electronic bands induced by the spontaneous ferromagnetism of Fe moments.

**Chern bands and phase diagram.** — In order to explore the dependence of band topology on the Hamiltonian parameters, for a filling of 1-electron per Mo, we have computed the magnetization direction and Chern number $C$ of the relevant low-lying bands as a function of the trigonal distortion parameter, $\chi_{\text{tri}}$, and the second neighbor interorbital hopping $t_m$. Even this limited exploration of the full parameter space yields a rich phase diagram, shown in Fig. 3, with several magnetic phases and emergent band topologies, illustrating that the $\{111\}$ grown DPs may be particularly useful systems to study topological phases of correlated oxide materials. We find that the electronic states show the following phases depending on the magnetization direction: (i) normal metal ($N,M$) where the lowest pair of bands overlap in energy and they are both topologically trivial; (ii) a normal insulator ($NI$) phase where a full gap opens up between these topologically trivial bands; (iii) A Chern metal ($CM$) where the lowest pair of bands have (opposite) nontrivial Chern numbers as indicated, yet overlap in energy leading to a metallic state with a non-quantized anomalous Hall response; (iv) a $C = \pm 2$ Chern insulator ($CI$) where a full gap opens up between the two lowest topologically nontrivial bands leading to a quantized anomalous Hall conductance $\sigma_{xy} = 2e^2/h$ and a pair of chiral edge modes. Fig. 4 shows the spectrum of the $CI$ state in a cylinder geometry, depicting a pair of chiral modes at each edge, which cross from the valence to the conduction band. We estimate the bulk gap of the $CI$ state to be $0.03t_c \sim 75\text{K}$.

**Effective model for $C = 2$ Chern bands.** — Chern bands with $C = 2$ are unusual [53][54] and differ from conven-
t m -0.05 explains the formation of C effective model thus captures our numerical results and find it leads to topologically trivial bands. This simple ω quantization axis along ˆ 3 j hopings and the inter-orbital hopping (Hamiltonian displays a skyrmion with winding number t details). For sufficiently large two Zeeman split states (see Supplementary Material for Mo. The two relevant lowest bands arise from the lowest point along 111. We project the Mo-Mo intra-orbital the states of neighboring Mo atoms, leading to Zeeman cally aligned Fe moments produce an exchange field on in Fig. 5(a)). At the atomic level, the ferromagneti- C the emergence of CI of bands remains separated from the higher bands. The CI(topological Landau levels or Hofstadter bands with 2 bands for t = 0, as we in- CI2 states on 111. As shown in Fig. 5(a), this leads to a wide swath of the phase diagram where the lowest two bands possess C = ±2. We find that this lowest pair of bands remains separated from the higher bands. The CI state then appears from opening up a full band gap between these lowest two nontrivial bands.

We next focus on constructing an effective model for the emergence of C = ±2 bands for χtri = 0, as we increase t m beyond a critical value (see trajectory shown in Fig. 3(a)). At the atomic level, the ferromagnetically aligned Fe moments produce an exchange field on the states of neighboring Mo atoms, leading to Zeeman splitting of the spin-orbit coupled j = 3/2 states on Mo. The two relevant lowest bands arise from the lowest Zeeman split j n = +3/2, +1/2 levels where j n = ˆ j · n and n is a unit vector along {111}. With the spin-quantization axis along n, these atomic wavefunctions are |j n = 3/2⟩ = 1/√3[(yz + ω |zx⟩ + ω2 |xy⟩)] ↑ and |j n = 1/2⟩ = −2√3/3[(yz) + |zx⟩ + |xy⟩] ↑ + 1/3[(yz) + ω |zx⟩ + ω2 |xy⟩] ↓ where ω = ei2π/3. We project the Mo-Mo intra-orbital hoppings and the inter-orbital hopping (t m) onto these two Zeeman split states (see Supplementary Material for details). For sufficiently large t m, the resulting two-band Hamiltonian displays a skyrmion with winding number 2 in the hexagonal Brillouin zone as shown in Fig. 3(b), thus accounting for C = ±2 Chern bands. For small t m, we find it leads to topologically trivial bands. This simple effective model thus captures our numerical results and explains the formation of C = ±2 bands.

Interestingly, we find a direct transition between the normal insulator and the CI with C = ±2 in our phase diagrams in Fig. 3(a) and Fig. 3(a). If such a transition occurred via a gap closing and reopening at the Brillouin zone corners, the critical theory would have massless Dirac fermions, with interactions being perturbatively irrelevant at the transition. Such transitions have also been discussed recently for integer quantum Hall plateau transitions of bosons 54. However, we find the gap closing at the NT-CI transition occurs at the zone center, leading to a quadratic band touching (with 2π Berry phase) at the critical point. Such a quadratic band touching point is protected by C6 symmetry in the noninteracting theory 55; interactions, which are marginally relevant, may lead to intermediate spontaneous nematic phases 56–58.

Discussion. — We have shown that simple double perovskite materials can exhibit a rich phase diagram with various ferromagnetic orders and band topologies in a bilayer grown along {111}. Such Chern bands emerging from half-metallic states have also been discussed recently at CrO2-TiO2 interfaces 22. Going beyond our simple model, the inclusion of electron-electron interaction effects on the Mo atom might expand the CI regime. Such correlation effects may be studied in the full model as well as the reduced two-band model of Chern bands which may be amenable to exact diagonalization studies. Such studies might also shed further light on the CI-NT transition and intermediate phases driven by a quadratic band touching instability. The broken inversion symmetry in the bilayer is expected to lead to a Rashba interaction; while the unusual topological phases we have uncovered are stable to a small Rashba coupling, a sufficiently strong Rashba interaction might drive spin spirals of Fe moments 15,27 and suppress the topological Chern bands. Further work is necessary to understand the competition between the Rashba and correlation effects. In future work, we also plan to study similar physics in bilayers of 5d-based double perovskites such as Ba2FeReO6 which have a d2 configuration and stronger spin-orbit coupling, which could stabilize more robust quantum anomalous Hall phases.

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a) t denotes by t, hopping terms are shown in Fig. 6(a)-(c) for intra-orbital as well as inter-orbital terms are allowed by symmetry, and we retain both processes. The intra-orbital t to the inter-orbital hopping, with coupling t, along \{34, 35, 38\}. We expect a similarly small interorbital hopping t, broken to a discrete symmetry, allowing for a nonzero \(t\). We have checked that the Chern bands are robust to slight variations in these hopping parameters and tuning of the spin orbit coupling strength.

The Ising axis over a correlated domain of area \(T\). We start from the isotropic 2D Heisenberg model, where the correlation length diverges as \(\xi(t) = \frac{t}{T}\). For weak Ising anisotropy \(\delta E\), the energy cost of misaligning moments away from the Ising axis over a correlated domain of area \(\xi^2(T)\) is \(\delta E \sim \xi^2(T)\). Equating this with \(T\) yields the temperature \(T\), which yields an estimate \(T\), in the case of the Ising ordered state along \{111\} which supports interesting \(C = \pm 2\) Chern bands.

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Next, staying at \(T \ll T\), we consider the effective domain wall energy cost of a \{111\}-\{\overline{1}1\} boundary in the presence of a typical anisotropy energy cost \(\delta E\) to turn the Fe spin away from the Ising axis. For small \(\delta E\), we can optimize the domain wall size \(\ell_{dw}\) by minimizing, with respect to \(\ell_{dw}\), the energy per unit length of a straight domain wall on the triangular lattice,

\[
\mathcal{E} = \frac{1}{2} J_F S_F^2 \left(\frac{\pi}{\ell_{dw}}\right)^2 \ell_{dw} + \frac{1}{2} (\delta E) \ell_{dw},
\]  

(2)

Parameters in the tight binding model.

We consider symmetry allowed nearest neighbor Mo-Fe intra-orbital hoppings. For next neighbor Mo-Mo hoppings, intra-orbital as well as inter-orbital terms are allowed by symmetry, and we retain both processes. The intra-orbital hopping terms are shown in Fig. 6(a)-(c) for \(d_{xz}, d_{yz}, d_{zz}\) orbitals. The two nearest neighbor intra-orbital hoppings are denoted by \(t_\pi\) and \(t_\delta\). The next-neighbor intra-orbital hoppings are denoted by \(t'\), and \(t''\). Finally, Fig. 6(d) depicts the inter-orbital hopping, with coupling \(t_m\), between different indicated orbitals on nearest pairs of Mo sites. In our computations, with \(t_\pi = 1\), we set \(t_\delta = -0.11\), \(t' = -0.09\), \(t'' = 0.1\), which are similar to values in the literature \[34, 35, 38\]. We expect a similarly small interorbital hopping \(t_m\sim -0.1t_\pi\). These hopping parameters provide a good description of the bulk properties; however, they might get slightly modified due to the trigonal distortion in the bilayer geometry. To simplify the exploration of the full multi-dimensional space of parameters, we fix the strength of the trigonal distortion \(\chi_{H}\) and the inter-orbital hopping term \(t_m\) keeping \(t_\delta, t', t''\) fixed. We fix the charge transfer energy \(\Delta = 2t_\pi\) \[38\], and the spin orbit coupling \(\lambda = 0.5t_\pi\). We fix \(t_\pi = 250meV\), close to values used in earlier studies \[34, 38\]. We have checked that the Chern bands are robust to slight variations in these hopping parameters and tuning of the spin orbit coupling strength.

Estimate of ferromagnetic \(T\).

In the absence of spin-orbit coupling, the Fe moments which form a ferromagnetic state on the triangular lattice can have moments pointing in any direction in spin-space. The resulting effective model for Fe moments will have Heisenberg symmetry, leading to \(T = 0\) for the 2D bilayer. With spin-orbit coupling, this Heisenberg symmetry is broken to a discrete symmetry, allowing for a nonzero \(T\). Below, we estimate \(T\) in the case of the Ising ordered state along \{111\} which supports interesting \(C = \pm 2\) Chern bands.

We start from the isotropic 2D Heisenberg model, where the correlation length diverges as \(\xi(T) \sim e^{2\pi\rho_s/T}\) \[31\], with the spin stiffness \(\rho_s \sim 2J_F S_F^2\). For weak Ising anisotropy \(\delta E\), the energy cost of misaligning moments away from the Ising axis over a correlated domain of area \(\xi^2(T)\) is \(\delta E \sim \xi^2(T)\). Equating this with \(T\) yields the temperature \(T\), below which we must view the Heisenberg model as an effective Ising model, where misalignment of spins away from the Ising axis is not thermally possible. Using \(J_F\) as 1.5meV, \(S_F = 5/2\), and \(\delta E \sim 0.2meV\), this yields an estimate \(T\) \(\sim 200K\), which is only logarithmically sensitive to \(\delta E\).

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\[
\mathcal{E} = \frac{1}{2} J_F S_F^2 \left(\frac{\pi}{\ell_{dw}}\right)^2 \ell_{dw} + \frac{1}{2} (\delta E) \ell_{dw},
\]  

(2)

Appendix

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where the first term is the gradient cost and the second term is the average ‘misalignment’ energy. This leads to 

\[ \ell_{dw} = \pi S_F \sqrt{2J_{FF}/\delta E}, \]

and to the optimal energy of the domain wall per unit length \( E_{opt} = \pi S_F \sqrt{2J_{FF}/\delta E}. \)

We can view \( E_{opt}/4 \) as being the exchange coupling \( J_{eff} \) of an effective 2D triangular lattice Ising model. The exact transition temperature of the Ising model on the 2D triangular lattice is \( T_c = 4J_{eff}/\ln 3 \approx 0.9E_{opt}. \) This yields a rough estimate \( T_c = \alpha S_F \sqrt{J_{FF}/\delta E}, \) with \( \alpha \approx 4. \) Using \( J_{FF} = 1.5\text{meV}, S_F = 5/2, \) and \( \delta E \approx 0.2\text{meV}, \) yields \( T_c \approx 65\text{K}. \)

The argument leading to this estimate appears to be internally consistent, with Ising-like moments forming at \( T \) approximately consistent with the experimentally inferred ordering at \( T_c < T_{\pi}. \)

In fact, our crude estimate might be significantly underestimating \( T_c, \) and the ordering is likely to occur at somewhat higher temperatures. Numerical studies of anisotropic Heisenberg models [62] find a much higher transition temperature, about 60% of the Ising model transition temperature, even for extremely weak anisotropies (\( \sim 10^{-3} \)). In our case, using this numerical result would suggest \( T_c \approx 220\text{K}. \) Thus magnetic ordering might occur quite close to the Heisenberg-Ising crossover temperature \( T_{\pi} \) estimated above.

**Effective two-orbital model of \( C = \pm 2 \) Chern bands.**

The spin-orbit coupled atomic wavefunctions corresponding to \( j = 3/2 \) states with projection \( j_n = 3/2, 1/2 \) are respectively given by

\[ |j_n=3/2\rangle = \frac{1}{\sqrt{3}}(|yz\rangle + \omega|zx\rangle + \omega^2|xy\rangle)|\uparrow\rangle, \quad (3) \]

and

\[ |j_n=1/2\rangle = -\frac{\sqrt{3}}{3}(|yz\rangle + |zx\rangle + |xy\rangle)|\uparrow\rangle + \frac{1}{3}(|yz\rangle + \omega|zx\rangle + \omega^2|xy\rangle)|\downarrow\rangle, \quad (4) \]

where \( \omega = e^{i2\pi/3}. \) Here \( j_n = \hat{j} \cdot \hat{n} \) with \( \hat{n} \) along \{111\}, and the Fe moments are assumed to point along \{111\}. Due to the Fe ordering, there is an effective Zeeman field experienced by the Mo sites which leads to a Zeeman splitting \( B_z \) between the \( j_n = 3/2 \) and \( j_n = 1/2 \) states. Since SFMO is half-metallic, we make the simplifying assumption that the relevant bands near the Fermi level are well described by considering only hopping of the \( \uparrow \) spins, and focusing on only the Mo sites due to the charge transfer energy \( \Delta = 2.5t_w \) which suppresses occupation on Fe sites. The Mo-Mo hopping has two dominant contributions: (i) the inter-orbital term \( t_m \) in the original Hamiltonian; (ii) an effective \( t'_m \) hopping, which includes the direct \( t' \) hopping between Mo-Mo as well as (possibly more significant) indirect Mo-Fe-Mo hoppings which can occur at \( O(t'_m^2/\Delta). \) These are schematically depicted in Fig. 7.

We can project both hopping processes onto the \( j_n = 3/2, 1/2 \) atomic states, which leads to a 2-orbital triangular lattice Hamiltonian. In momentum space, this takes the form

\[ H(k) = \begin{pmatrix}
-\frac{3}{2}(t'_{eff} - t_m)\gamma_k - B_z & \frac{2\sqrt{3}}{3\sqrt{3}}(t'_{eff} - \omega_t_m)\beta_k \\
\frac{2\sqrt{3}}{3\sqrt{3}}(t'_{eff} - \omega^2t_m)\beta^*_k & -\frac{4}{3}(t'_{eff} + 2t_m)\gamma_k
\end{pmatrix} \]

(5)
Let us define $\hat{a} = \hat{x}, \hat{b} = -\hat{x}/2 + \hat{y}\sqrt{3}/2, \hat{c} = -\hat{x}/2 - \hat{y}\sqrt{3}/2$. In terms of these, the matrix elements are given by $\gamma_k = \sum_{\delta} \cos k \cdot \hat{\delta}$ with $\hat{\delta} = \hat{a}, \hat{b}, \hat{c}$, and $\beta_k = \omega \cos k \cdot \hat{a} + \omega^2 \cos k \cdot \hat{b} + \cos k \cdot \hat{c}$. We expect $B_z \sim t'_\text{eff}$. Fixing $B_z, t'_\text{eff}$ and varying $t_m$ leads to a transition between (i) a topologically trivial state where both bands have Chern number zero and (ii) a topologically nontrivial state where bands have Chern numbers $C = \pm 2$. This topologically nontrivial state is characterized in momentum space by the development of a winding number 2 skyrmion texture as shown in Fig. 5(b) of the paper, where the arrows represent the ‘effective magnetic field’ direction in the $2 \times 2$ space of Eq. 5.