Three-dimensional Stacking of Canted Antiferromagnetism and Pseudospin Current in Undoped Sr$_2$IrO$_4$: Symmetry Analysis and Microscopic Model Realization

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Recent optical second-harmonic generation experiments observed unexpected broken spatial symmetries in the undoped spin-orbit Mott insulator Sr$_2$IrO$_4$, leading to intensive debates on the nature of its ground state. We propose that it is a canted antiferromagnetism with a hidden order of circulating staggered pseudospin current. Symmetry analysis shows that a proper c-axis stacking of the canted antiferromagnetism and the pseudospin current lead to a magnetoelectric coexistence state that breaks the two-fold rotation, inversion, and time-reversal symmetries, consistent with experimental observations. We construct a three-dimensional Hubbard model with spin-orbit coupling for the five localized 5$d$ Wannier orbitals centered at Ir sites, and demonstrate the microscopic realization of the desired coexistence state in a wide range of band parameters via a combination of self-consistent Hartree-Fock and variational calculations.

I. INTRODUCTION

The layered square-lattice iridate Sr$_2$IrO$_4$ has been intensively studied since the discovery of the spin-orbit Mott insulator Sr$_2$IrO$_4$ [1,2], as a consequence of the interplay between spin-orbit coupling (SOC) and electron correlation [3–9]. The strong SOC of Ir atoms splits the $t_{2g}$ orbitals into a fully-occupied $J_{\text{eff}} = 3/2$ quartet and a half-filled $J_{\text{eff}} = 1/2$ doublet. The latter is then localized by an otherwise moderate electronic correlation, realizing a single-band pseudospin-1/2 Heisenberg antiferromagnet (AFM) on the quasi-two-dimensional square lattice [1], with strong exchange couplings $\sim J \sim 60$ meV [10]. This makes Sr$_2$IrO$_4$ a promising analog of the cuprates, and is thus expected to be another platform for unconventional superconductivity [11–14]. A remarkable range of cuprate phenomenology has been observed in both electron- and hole-doped Sr$_2$IrO$_4$, including Fermi surface pockets [15], Fermi arcs [16], pseudogaps [17, 18], and d-wave gaps [19, 20]. Whether a superconducting state exists as in the cuprates requires understanding thoroughly the correlated spin-orbit entangled electronic states observed in Sr$_2$IrO$_4$.

The ground state of the undoped Sr$_2$IrO$_4$ is of particular interest since it is the parent phase from which these novel spin-orbit entangled correlated states emerge. The electron correlation in the spin-orbit Mott state results in an insulating ground state with AFM long-range order. Neutron and resonant X-ray measurements reveal that the magnetic moments are aligned in the basal $ab$ plane, with their directions tracking the $\theta \sim 11^\circ$ staggered IrO$_6$ octahedra rotation about the $c$ axis due to strong SOC [21–25]. This gives rise to a net ferromagnetic (FM) moment along the $a$ axis, in addition to the AFM component along the $b$ axis. The net FM moment of each layer is shown to order in a $+-+-+$ pattern along the $c$ axis [2, 26], where $\pm$ refers to the direction the FM moment along the $a$ axis. A schematic illustration of the state is show in Fig. 1(a). This magnetic ground state, hereinafter denoted as $+-+-$ canted AFM (CAF), belongs to a centrosymmetric orthorhombic magnetic point group $2/m1'$ with spatial $C_{2z}$ rotation, inversion, and time-reversal symmetries [27]. Recent optical second-harmonic generation (SHG) experiments [27] reported evidence of unexpected breaking of spatial rotation and inversion symmetries, pointing to the existence of a symmetry-breaking hidden order. It is argued that the broken symmetries can be caused by loop-currents [27, 30] which were proposed to account for the pseudogap physics in the high-$T_c$ cuprates [31–33]. However, the oxygen 2$p$ states in Sr$_2$IrO$_4$ are much further away from ($\sim 3$ eV below) the Fermi level than those in the cuprates [1, 34], making it disadvantageous to develop the loop-currents that requires low-energy oxygen 2$p$ states. Furthermore, the experimental measurements [27, 30] suggest a magnetoelectric loop-current order that is ferroically stacked along the $c$ axis, which is incompatible with the recent observation [35] of an SHG signal that switches sign every two layers.

On the other hand, a different hidden order of circulating staggered (i.e., d-wave) pseudospin current (dPSCO) has been proposed [36] to describe the band dispersion and the pseudogap phenomena observed in the electron-doped Sr$_2$IrO$_4$. The dPSCO generates a d-wave spin-orbit density wave and gives rise to Fermi pockets and Fermi arcs in the nonmagnetic electron-doped Sr$_2$IrO$_4$, in good agreement with angle-resolved photoemission (ARPES) and scanning tunneling microscopy (STM) measurements [15, 17, 19]. It was argued that the dPSCO is already present in the insulating magnetic phase of the
undoped Sr$_2$IrO$_4$, responsible for the observed splitting of the bands at $(\pi, 0)$ whose two-fold degeneracy is otherwise protected by certain lattice symmetries. While describing remarkably well the highly unconventional quasiparticle properties observed in both the electron-doped and undoped Sr$_2$IrO$_4$, the $d$PSCO in Ref. was considered in the two-dimensional limit of a single IrO$_2$ layer. Further studies on the $c$-axis stacking of the dPSCO and the magnetic order in realistic three-dimensional systems are necessary in order to compare directly to the findings of the nonlinear optical experiments and the interpretation in terms of intracell loop-currents.

In this work, we discuss the symmetry properties of the $c$-axis stacking of CAF, dPSCO, and their coexistence, and study their microscopic realization in realistic three-dimensional models for undoped Sr$_2$IrO$_4$. The rest of the paper is organized as follows. In Sec. II, we perform symmetry analysis. We find that the particular coexistence state with $++--$ CAF and $\oplus\oplus\ominus\ominus$ dPSCO has the symmetries consistent with experimental observations in undoped Sr$_2$IrO$_4$. It is a magnetoelectric state that breaks the spatial two-fold rotation, inversion, and time-reversal symmetries. Considering all five $5d$ orbitals of the Ir atoms, a realistic three-dimensional tight-binding model including SOC (TB+SOC) and the structural distortion is constructed in Sec. IIIA, which describes faithfully the low-energy band structure of Sr$_2$IrO$_4$ with the structural distortion. The Hubbard interactions are introduced in Sec. IIIB and treated within the Hartree-Fock approximation to account for the effects of electron correlations that generate magnetism spontaneously. We obtain CAF phases with different $c$-axis stacking pattern self-consistently and compare their energies. The $++--$ CAF revealed in experiments is found to be energetically favored in a wide range of band parameters. In Sec. IIIC, the hidden dPSCO is considered phenomenologically by including a variational term in the Hamiltonian. We fix the stacking pattern of CAF to be $+-+$, and compare the energies of coexistence states with different $c$-axis stacking of dPSCO. The mostly favorable stacking pattern for dPSCO is found to be indeed the desirable $\oplus\ominus\ominus\ominus$, supporting the above-mentioned coexistence state as the ground state of undoped Sr$_2$IrO$_4$, with its symmetries consistent with experimental measurements. Discussions and summaries are presented in Sec. IV.

II. SYMMETRY ANALYSES

To be more precise, we denote the magnetic ground state of undoped Sr$_2$IrO$_4$ as $\{++--\}_a$ CAF, where the subscript $a$ specifies the direction of the net FM moment, since, in principle, it can be along either $a$ or $b$ axis. Fixing the net FM moment along $a = \{a, b\}$ axis, there are four possible relative stacking along the $c$ axis of the FM in-plane component of the moment in each of the four IrO$_2$ planes in a unit cell, i.e., $(++--)_a$, $(+-+-)_a$, $(+-+-)_a$, and $(+-++)_a$. It is easy to show that there is a one-to-one correspondence between states with FM moment along $a$ axis and those with FM along $b$ axis, by performing a $C_{4z}$ rotation along the $c$ axis and a lattice translation. Explicitly, the $(++--)_a$, $(+-+-)_a$, $(+-+-)_a$, and $(+-++)_a$ state are equivalent to, respectively, $(+-++)_b$, $(+-++)_b$, $(+-++)_b$, and $(+-++)_b$ state. The correspondence shall be verified numerically later in the microscopic model calculations presented in Sec. IIIB by comparing the state energies. Therefore, without loss generality, we restrict the direction of the FM moments to be along $a$ axis and drop the subscript for the canted AFM phases in the rest of the paper, unless otherwise noted.

(a) Symmetries of $c$-axis stacked CAF:

| stacking | $C_{2zz}$ | $M_z$ | $I$ | $T$ | $C'_{2zz}$ | $M'_z$ | $I'$ |
|----------|----------|-------|-----|-----|------------|--------|-----|
| ++--     | $\tau_{x}y_z$ | $\tau_{y}z_x$ | $\tau_{x}$ | $\tau_{y}z_x$ | 0 | $\tau_{x}$ | $\tau_{y}z_x$ |
| ++++     | $\tau_{x}y_z$ | $\tau_{y}z_x$ | $\tau_{y}z_x$ | $\tau_{y}z_x$ | 0 | $\tau_{y}z_x$ | $\tau_{y}z_x$ |
| +---     | $\tau_{x}y_z$ | $\tau_{y}z_x$ | $\tau_{y}z_x$ | $\tau_{y}z_x$ | 0 | $\tau_{y}z_x$ | $\tau_{y}z_x$ |

(b) Symmetries of $c$-axis stacked dPSCO:

| stacking | $C_{2zz}$ | $M_z$ | $I$ | $T$ | $C'_{2zz}$ | $M'_z$ | $I'$ |
|----------|----------|-------|-----|-----|------------|--------|-----|
| $\oplus\oplus\ominus\ominus$ | 0 | $\tau_{x}$ | $\tau_{x}$ | 0 | 0 | $\tau_{x}$ | $\tau_{x}$ |
| $\oplus\ominus\ominus\ominus$ | 0 | $\tau_{y}z_x$ | $\tau_{y}z_x$ | 0 | 0 | $\tau_{y}z_x$ | $\tau_{y}z_x$ |
| $\oplus\ominus\ominus\ominus$ | 0, $\tau_{x}y_z$ | $\tau_{x}z_y$ | $\tau_{x}z_y$ | 0, $\tau_{x}y_z$ | $\tau_{x}z_y$ | $\tau_{x}z_y$ |

TABLE I. Symmetries of $c$-axis stacked (a) CAF and (b) dPSCO. The table gives the lattice translation required for a state to recover itself after a symmetry operation of the magnetic space group $2/mI'$. Symbol $\times$ means such a lattice translation does not exist. Translation vector $\tau_z= (1/2, 0, 0)$, $\tau_{yz}=(0, 1/2, 1/2)$, and $\tau_{yz}= (1/2, 1/2, 1/2)$ in terms of the lattice constant of the conventional unit cell shown in Fig. 1. Note that the states listed in the last two rows of both (a) and (b) are invariant under a lattice translation of $\tau_{y,z}$, there are thus two possible lattice translations differed by $\tau_{x,z}$.

The symmetries of the CAF phases (without dPSCO) with different $c$-axis stacking are summarized in Table I(a), which gives the lattice translation, if exist, required for a state to recover itself after a symmetry operation of the magnetic point group $2/mI'$. Two-fold rotation around $z$-axis $C_{2zz}$, mirror reflection about $ab$-plane $M_z$, inversion $I$, time-reversal $T$, $C'_{2zz} = T C_{2zz}$, $M'_z = TM_z$, and $I' = TI$. The state does not have the corresponding symmetry if it could not recover itself by any lattice translation after a symmetry operation. Note that, because the magnetic moments are aligned in the basal $ab$ plane, without any $c$-axis component, the time-reversal operator $T$ transforms under the same irreducible representation as $C'_{2zz}$, and consequently, the operators $C'_{2zz}$.
\[ M'_2, \text{ and } I' \] are projected to identity \( E, I, \text{ and } M_2, \) respectively, as shown in Table I(a). The \(+--++\) and \(+++--\) CAF states share the same symmetries and belong to the centrosymmetric orthorhombic magnetic point group \( 2/m1' \). However, they are inequivalent in the presence of in-plane anisotropy \([33, 41]\), as will be shown in the microscopic model calculations presented in Sec. IIIB. The nonmagnetoelectric \(+++++\) CAF breaks \( \{C_{2z}, M_2, T, I'\} \), while the magnetoelectric \(+--++\) CAF breaks \( \{C_{2z}, I, T, M'_2\} \) symmetries. They belong to the magnetic point groups \( 2'/m' \) and \( 2/m \), respectively. It has been argued \([41]\) that both the \(+--++\) and \(+--++\) CAF can potentially explain the SHG experiment \([27]\) without invoking the loop-currents, and either of them might have been created by the laser pump used in the experiments. The possibility of laser-induced rearrangement of the magnetic stacking, however, has been ruled out by recent comprehensive measurements \([35]\), which show the magnetic stacking pattern is always \(+--++\) under the experimental condition before strong external field drives it to be \(+--++\).

| stacking | \( C_{2z} \) | \( M_2 \) | \( I \) | \( T \) |
|----------|---------|---------|--------|--------|
| +-- ++ / \( \oplus \ominus \ominus \ominus \) | x | x | ✓ | x |
| +-- ++ / \( \oplus \ominus \ominus \ominus \) | x | ✓ | x | x |
| +-- ++ / \( \oplus \ominus \ominus \ominus \) | ✓ | ✓ | ✓ | ✓ |
| +-- ++ / \( \oplus \ominus \ominus \ominus \) | ✓ | ✓ | ✓ | ✓ |
| ++-- / \( \ominus \oplus \ominus \ominus \) | x | ✓ | x | x |
| ++-- / \( \ominus \oplus \ominus \ominus \) | x | x | ✓ | x |
| ++-- / \( \ominus \oplus \ominus \ominus \) | x | x | ✓ | x |
| ++-- / \( \ominus \oplus \ominus \ominus \) | x | x | x | x |
| ++-- / \( \ominus \oplus \ominus \ominus \) | x | ✓ | x | x |
| ++-- / \( \ominus \oplus \ominus \ominus \) | x | x | x | x |
| ++-- / \( \ominus \ominus \ominus \ominus \) | x | ✓ | x | x |
| ++-- / \( \ominus \ominus \ominus \ominus \) | x | ✓ | x | x |
| ++-- / \( \ominus \ominus \ominus \ominus \) | x | x | x | x |

TABLE II. Symmetries of the coexistence states with possible \( c \)-axis stacking of CAF and dPSCO.

Before performing the symmetry analysis for the \( c \)-axis stacked hidden dPSCO, we define first the notation for its stacking pattern. The staggered \( \text{IrO}_6 \) octahedra rotation about the \( c \) axis results in two kinds of \( \text{Ir} \) sites, enclosed by octahedron rotated clockwise and anticlockwise, respectively, as shown in Fig. 1. The pseudospin moments on these two sublattices are represented, respectively, by red and green arrows. In a similar vein, the staggered rotation of \( \text{IrO}_6 \) octahedra gives rise to two kinds of \( \text{Ir} \) plaquettes, as depicted by the two blue squares in the \( z = 7/8 \) and \( z = 3/8 \) planes in Fig 1(a). The direction of the pseudospin currents around the two kinds of \( \text{Ir} \) plaquettes is denoted by, respectively, red and green symbols at the plaquette center, with \( \oplus/\ominus \) corresponding to anticlockwise/clockwise circulating pseudospin current. The stacking of the dPSCO is then characterized by the red symbols in each plane, from top to bottom. For instance, the stacking pattern for the dPSCO shown in Fig. 1(a) corresponds to \( \oplus \ominus \ominus \).

The symmetries of the nonmagnetic phases with possible \( c \)-axis stacked dPSCO are summarized in Table I(b). Since the dPSCO is invariant under the time-reversal operator \( T \), any operator is identical to its product with \( T \), \( e.g., C_{2z} = C'_{2z}, M_2 = M'_2, \) and \( I = I' \). As shown in Table I(b), the \( \oplus \ominus \ominus \), \( \oplus \ominus \ominus \), and \( \oplus \ominus \ominus \) dPSCO states have all the symmetries of the magnetic point group \( 2/m1' \), while the \( \oplus \ominus \ominus \) dPSCO breaks mirror reflection \( M_2 \) and inversion \( I \) but preserves the symmetries of two-fold rotation \( C_{2z} \) and time-reversal \( T \). It is important to note that all of the dPSCO states have the two-fold rotation and time-reversal symmetries. Thus none of them is able to describe the hidden order in \( \text{hole-doped Sr}_2\text{Ir}_{1-x}\text{Rh}_x\text{O}_4 \) observed by SHG and polarized neutron scattering measurements. We argue that the physics in the hole-doped \( \text{Sr}_2\text{Ir}_4 \) to be quite different than that on the electron-doped side. The \( \text{Rh} \) substitution \([44, 47]\) of the strongly spin-orbit coupled \( \text{Ir} \) in the \( \text{Ir}-\text{O} \) plane is very different than the electron doping by La substitution \([45, 50]\) in the off-plane charge reservoir layers or surface K doping \([16, 17, 19, 51]\). Furthermore, in the \( \text{IrO}_2 \) plane is very different than the electron doping substitution \([42–47]\) of the strongly spin-orbit coupled \( \text{Ir} \) has a different electronic structure than that of an electron and is more likely to involve higher pseudospin states \([52]\). We therefore leave the hole-doped \( \text{Sr}_2\text{Ir}_4 \) aside, and consider only the undoped and electron-doped \( \text{Sr}_2\text{Ir}_4 \). Their unconventional low-energy quasiparticle properties observed by ARPES and STM have been described successfully by the hiddenorder of dPSCO \([30]\). Our focus in this paper is to investigate the effects of dPSCO on the symmetry properties of the three-dimensional state, which enables a direct comparison to SHG and polarized neutron scattering experiments. At stoichiometry where these experiments have been conducted, the Néel temperature \( T_N \) and the hidden order transition temperature \( T_\text{h} \) are very close to each other and barely distinguishable, thus provide unambiguously only the symmetry information of the ground state, \( i.e., \) the coexistence state of CAF and hidden order.

The coexistence state has a symmetry only if there exist a lattice translation that simultaneously recovers both the CAF and the dPSCO states after the corresponding symmetry operation. Using the symmetries of the CAF and dPSCO summarized in Table I, the symmetries of the coexistence states are readily obtained, with the result given in Table II for all possible \( c \)-axis stacking patterns. Remarkably, there is one particular coexistence state, \( i.e., +-- ++ / \oplus \ominus \ominus \ominus \) with \(+-- ++\) CAF and \( \oplus \ominus \ominus \) dPSCO, whose magnetism and sym-
It is clear that only the structure in Fig. 1(c) can recover the original one by a simple lattice translation. Two-fold rotation and time-reversal symmetries while preserving the mirror reflection symmetry. These properties make this coexistence state a promising candidate for the ground state of undoped Sr$_2$IrO$_4$. Fig. 1(a) shows the structure of the CAF and dPSCO in the + − + / ⊕ ⊕ ⊕ coexistence state. The resultant structures upon applying two-fold rotation $C_{2z}$, mirror reflection $M_z$, inversion $I$, and time-reversal $T$ are shown explicitly in Fig. 1(b-e). It is clear that only the structure in Fig. 1(c) can recover the original structure in Fig. 1(a) after a lattice translation by $\tau_{yz}=\mathbf{0}$, while the other three structures could not recover that in Fig. 1(a) by any lattice translation.

### III. MICROSCOPIC MODELS

#### A. Three-dimensional TB+SOC model

The two-dimensional TB+SOC model constructed in Ref. 36

$$\mathcal{H}_0 = \sum_{i,j,\mu,\sigma} t_{ij}^{\mu,\sigma} d^\dagger_{i\mu\sigma} d_{j\nu\sigma} + \sum_{i,\mu,\sigma} \epsilon_{\mu} d^\dagger_{i\mu\sigma} d_{i\mu\sigma}$$

(1)

provides a faithful description of the DFT band structure downfolded to the five low-energy Ir 5$d$-electron orbitals, as shown in Fig. 2(d). Here, $d^\dagger_{i\mu\sigma}$ creates an electron with spin $\sigma$ at site $i$ in the $\mu$th orbital defined in the local coordinate that rotates with the IrO$_6$ octahedron, and $\mu = 1(d_{yZ}), 2(d_{zX}), 3(d_{XY}), 4(d_{XZ}-R_2), 5(d_{YZ}-Y_2)$. The crystalline electric field effects are taken into account in the on-site energy term $\epsilon_{\mu} = \sum_{i,j} D_{ij}\mathbf{R} \cdot \mathbf{S}_{ij}$, with a separation of $\Delta_c \equiv 10Dq \approx 3.4$ eV between the $t_{2g}$ and $e_g$ complexes. The strength of atomic SOC $\lambda_{SOC} = 357$ meV. The spin and orbital angular momentum operators, $\mathbf{S}$ and $\mathbf{L}$, have matrix elements, $S_{\sigma\sigma'} = \langle \sigma|\mathbf{S}|\sigma'\rangle$ and $L_{\mu\nu} = \langle \mu|\mathbf{L}|\nu\rangle$, given explicitly in Ref. 36. The spin-and-orbital-dependent complex hopping integrals $t_{ij}^{\mu,\sigma}$ between sites $i$ and $j$ in the realistic Sr$_2$IrO$_4$ with structural distortion are derived from those in the idealized Sr$_2$IrO$_4$ without structural distortion $t_{ij}^{\mu,\sigma}$ by transforming the $10 \times 10$ hopping matrix, $t_{ij} = R_i^\dagger t_{ij} R_j$. The operator $R_i = e^{-iL_z\theta_i} \otimes e^{iS_z\theta_i}$ amounts to a joint spatial rotation from the global $(x, y, z)$ to the local $(X, Y, Z)$ coordinates by $\theta_i$ and a spin rotaion by the same angle $\theta_i$. Note that there is a 45° rotation between the $x, y$ axis of the global coordinate defined in this Section and the $a, b$ axis used in Sec. II, as shown in the inset in Fig. 1. In the undistorted idealized systems, the hopping integrals $t_{ij}^{\mu,\sigma}$ are real, spin-independent, and given in Ref. 36 explicitly up to the fifth nearest neighbors in a IrO$_2$ layer.

To construct a realistic three-dimensional model for Sr$_2$IrO$_4$, in addition to the in-plane $t_{ij}$ given in Ref. 36, we include nonzero $I_{ij}$’s on the nearest neighbor (NN) inter-layer bonds, i.e., site $i$ and $j$ from two adjacent IrO$_2$ layers. Owing to the shift between adjacent IrO$_2$ ab-planes, as shown in Fig. 1(a), there are eight inter-layer NN sites for each Ir atom, four in the layer right above it and the other four in the layer right below it. The inter-layer hoppings can be limited to the $t_{2g}$ orbitals since the contribution from the $e_g$ orbitals to the low-energy states is negligible small. Furthermore, the inter-layer hoppings involving the planar orbital $d_{xy}$ are expected to be small. In fact, any significant inter-layer intraorbital hopping of the $d_{xy}$ orbital would split the bands around $-2$ eV below the Fermi level, clearly incompatible with the LDA band...
structures shown in Fig. 2(a). We thus consider only inter-layer hoppings involving the $d_{yz}$ and $d_{zx}$ orbitals, $t_z = (t_{z1}, t_{z2})$, where $t_{z1}$ and $t_{z2}$ denote, respectively, the intraorbital and interorbital hoppings. While the intraorbital hoppings are isotropic on the inter-layer NN bonds, the interorbital hoppings are anisotropic, taking values $\pm t_{z2}$ on the four inter-layer NN bonds parallel to the [1, ±1, 0] plane in the (x, y, z) global coordinates.

Figs. 2(b-f) show the electronic structures of the three-dimensional TB+SOC model with various inter-layer hoppings $t_z = (t_{z1}, t_{z2})$, in comparison to the LDA band structure plotted in Fig. 2(a). Note that the band structures for opposite inter-layer hoppings (i.e., $t_z \rightarrow -t_z$) are identical, and remain equivalent even in the presence of Hubbard interaction and dPSCO considered in the following subsections. We therefore fix the intraorbital $t_{z1}$ to be positive. Fig. 2(d) displays the band structure of the two-dimensional TB+SOC model without any inter-layer hopping, $t_z = 0$, and Fig. 2(e) and 2(f) show, respectively, the individual effects of the intraorbital $t_{z1}$ and interorbital $t_{z2}$ on the band structure. Clearly, $t_{z2}$ does little to the band structure, while $t_{z1}$ splits the bands and thus captures the essential interlayer features of the LDA bands displayed in Fig. 2(a). The splitting is about 4$t_{z1}$ at N point for the bands right below the Fermi level. Therefore, an intraorbital $t_{z1}$ of 20 meV would reproduce the ~78 meV band splitting in the LDA bands. The effects of the interorbital $t_{z2}$ on the band structures are negligible even in the presence of nonzero $t_{z1}$, as shown in Fig. 2(b) and 2(c). Consequently, $t_{z2}$ remains as a tunable band parameter that shall be determined later by the c-axis stacking of the magnetism.

B. Stacking of canted AFM

To investigate the magnetism in Sr$_2$IrO$_4$, we consider the three-dimensional five-orbital Hubbard model $H = H_A + H_U$, with the electron correlations described by the standard multiorbital Hubbard interactions

$$H_U = \sum_{i,\mu} \hat{n}_{i\mu}^+ \hat{n}_{i\mu} + (U' - J_H/2) \sum_{i,\mu < \nu} \hat{n}_{i\mu} \hat{n}_{i\nu} \quad (2)$$

$$- J_H \sum_{i,\mu \neq \nu} \mathbf{S}_{i\mu} \cdot \mathbf{S}_{i\nu} + J_H \sum_{i,\mu \neq \nu} d_{i\mu}^+ d_{i\nu} d_{i\nu}^+,$$

where $U$ and $U'$ are the local intraorbital and interorbital Coulomb repulsions and $J_H$ is the Hund’s rule coupling with the relation of $U = U' + 2J_H$. The interactions in Eq. (2) are treated within the Hartree-Fock approximations. In the presence of SOC, the Hartree and exchange self-energies induced by $H_U$ depend on the full spin-orbital-dependent density matrix $n_{\mu\sigma\sigma'}^{\mu\nu}$, which are determined self-consistently in the numerical calculations. Local physical quantities in the ground state can be expressed in terms of $n_{\mu\sigma\sigma'}^{\mu\nu}$, the local spin density $S_i^\sigma = \sum_{\mu,\sigma'} S_{i\mu\sigma}^\sigma n_{i\mu\sigma'}^{\mu\nu}$, and the local orbital angular momentum $L_i^\sigma = \sum_{\mu,\mu' \neq \sigma} S_{i\mu\sigma}^\sigma n_{i\mu\sigma'}^{\mu\nu} L_{\mu\nu}$. In all calculations presented in this paper, we choose $(U, J_H) = (1.2, 0.05)$ eV that, in the two-dimensional calculations [36], produces correctly the CAF as the magnetic ground state for the undoped Sr$_2$IrO$_4$, and the low-energy quasiparticle properties in good agreement with ARPES measurements [12].

We first verify numerically the one-to-one correspondence, discussed in the previous section, between the CAF states with FM moment along a axis and those with FM moment along b axis. The direction of the net FM moment can be pinned by choosing appropriate initial values for $n_{i\mu\sigma\sigma'}^{\mu\nu}$. In numerical calculations, an octant of the reduced Brillouin zone, corresponding to the conventional unit cell with eight Ir atoms shown in Fig. 1(a), is discretized evenly into $L_x \times L_y \times L_z$ k-points. We obtain these states self-consistently at various $L_x \times L_y \times L_z$ and compare their energies. Fig. 3(a) plots the energy difference between the $(+ - -)_a$ and $(+ + -)_b$ CAF states as a function of the in-plane k-point $L_x \times L_y$, with the inter-layer hopping fixed to be $t_z = (20, -20)$ meV. The energy difference is not sensitive to $L_x$, probably because the inter-layer hoppings $t_z$ are much smaller in amplitude than the in-plane hoppings. Except for the first two data points, the energy difference is less than 0.01 μeV per site, within the resolution of our numerical calculations.
We thus conclude that the (+−−+) and (+−−−) canted AFM states are equivalent, consistent with the symmetry analysis. The correspondences between other states are also verified numerically. To reduce the finite-size effect, we take \(L_x \times L_y \times L_z = 601 \times 600 \times 10\) in the rest of the paper.

In the absence of interorbital hopping, \(t_{z2} = 0\), the interorbital \(t_{z1}\) dependence of the state energy of +−−+, +−−−, and +−++ CAF is shown in Fig. 3(b), with respect to that of the +−++ CAF. Clearly, the +−−+ and +−++ CAF are identical in energy at any interorbital \(t_{z1}\), implying the absence of the in-plane anisotropy. It is thus equivalent for the net FM moment to align in either the \(a\)-axis or the \(b\)-axis, in the absence of \(t_{z2}\). The interorbital \(t_{z1}\) energetically favors the +−++ CAF, while disfavors mostly the +−−+ CAF. Fixing interorbital \(t_{z2} = 20\) meV by the \(\sim 78\) meV band splitting in LDA, Fig. 3(c) plots the energies of the CAF states as a function of interorbital \(t_{z2}\). While the +−−+ CAF is the most unfavored magnetic state on the positive \(t_{z2}\) side, there is a wide range on the negative side, \(t_{z2} \in (-38, -6)\) meV, where the +−−+ CAF becomes the lowest in energy, supporting the ground magnetic structure revealed in experiments [2, 20]. Furthermore, the +−++ CAF is higher in energy by about \(5\) µeV per site at \(t_{z2} = -20\) meV, which agrees remarkably well with the \(0.3\) T external magnetic field required in experiments to align all FM moment along one direction[2].

Within the pseudospin-only models [39, 54], it has been shown that the \(c\)-axis stacking of the static long-range magnetic order in the undoped \(Sr_2IrO_4\) is stabilized by the interplay of interlayer pseudospin couplings, including the first-nearest-interlayer interaction \(J_{1c}\), the second-nearest-interlayer interaction \(J_{2c}\), and the anisotropy \(\Delta_c\) comes from the anisotropic interlayer interaction[55]. In terms of the effective couplings between the net moments (\(S = L + 2S\), \(J_{1c} = 4J_{1c}S^2\sin^2 \theta\), \(J_{2c} = -J_{2c}S^2(\cos^2 \theta - \sin^2 \theta)\), and \(\delta_c = 4\Delta_cS^2\cos^2 \theta\), the energies of the +−−+, +−−−, +−++ and +−++ CAF states are, respectively, \(-\delta_c - J_{2c}\), \(-\delta_c - J_{2c}\), \(-J_{1c} + J_{2c}\), and \(-J_{1c} + J_{2c}\). In the CAF states obtained self-consistently at Hubbard interactions \((U, \bar{J}) = (1.2, 0.05)\) eV, the ordered pseudospin moment \(\vec{S} \simeq 0.67\) \(\mu_B\) and the canting angle \(\theta \simeq 22^\circ\). It is readily and instructive to extract, from the Hartree-Fock state energies given in Fig. 3(b) and 3(c), the values of \(J_{1c}, J_{2c}, \text{ and } \Delta_c\). The interlayer pseudospin couplings are plotted in Fig. 3(d) as a function of interorbital \(t_{z1}\) in the absence of interorbital \(t_{z2} = 0\), and in Fig. 3(e) as a function of interorbital \(t_{z2}\) with the interorbital hopping fixed to be \(t_{z1} = 20\) meV. Clearly, the interorbital \(t_{z1}\) does not generate any anisotropy \(\Delta_c\), while the interorbital \(t_{z2}\) produces an anisotropy linear in \(t_{z2}\). In the absence of \(t_{z2}\), the interorbital \(t_{z1}\) produces a \(J_{1c} \propto t_{z1}^2\) and a \(J_{2c} \propto t_{z1}^4\), as shown in Fig. 3(d). At a fixed nonzero interorbital \(t_{z1}\), the superexchange interactions \(J_{1c}\) and \(J_{2c}\) generated by interorbital \(t_{z2}\) can be well fitted by quadratic and quartic functions of \(t_{z2}\) respectively, as shown in Fig. 3(e). These behaviors are consistent with the fact that superexchange interactions \(J_{1c}\) and \(J_{2c}\) are generate by, respectively, the second-order and quadratic-order perturbations in the inter-layer hoppings. Interestingly, at \(t_{z2} = (-20)\) meV, the interlayer pseudospin couplings \((J_{1c}, J_{2c}, \Delta_c) = (21.6, 14.0, 16.6)\) \(\mu_eV\) are consistent with the values extracted from experiment [39].

Fig. 4 displays the band dispersions of the CAF states with different \(c\)-axis stackings. They are all AFM insulators with a similar overall structure. The quasiparticle band below the AFM gap has an eight-fold degeneracy at \(X\) point, four of them due to the folding along the \(c\) axis.
of the conventional unit cell and the other two are protected by the two-fold rotation symmetry $C_{2a}$ about the $a$ axis, along which the FM moment aligned. These eight bands behave differently along the $a$ axis, along which the FM moment aligned. These eight band structures. They remain degenerate in the $+$ state. Splitting into two branches in the $++$ CAF, split into three branches in the $+−−$ CAF, but split into two branches in the $+−+$ and $+++$ CAF states.

C. Stacking of dPSCO

The physical origin of the dPSCO is still under investigation [36], and out of the scope of the current paper. Therefore, unlike the CAF, its stacking pattern could not be determined self-consistently by including in the Hamiltonian an interaction term from which the dPSCO develops spontaneously. Instead, we determine its c-axis stacking via a variational approach. Explicitly, a variational term for the dPSCO, $H_{\Delta}$, is added to the Hamiltonian, $H = H_{0} + H_{U} + H_{\Delta}$, with

$$H_{\Delta} = i\Delta \sum_{i \in A, \sigma} \sum_{j = i + \delta} \eta_{i} \tau_{ij}^{\sigma} \left( \gamma_{i\sigma}^{\dagger} \gamma_{j\sigma} - \chi_{ij}^{\sigma} \right) + H.c.,$$

where the NN vector $\delta = \{ \pm \hat{x}, \pm \hat{y} \}$, the standard NN $d$-wave form factor $\tau_{ij} = (-1)^{i+y+j}$, and $\chi_{ij}^{\sigma} = \langle \gamma_{i\sigma}^{\dagger} \gamma_{j\sigma} \rangle$ whose presence ensures that the variational term $H_{\Delta}$ does not add an elastic part to the state energy. The operator $\gamma_{\sigma} = \frac{1}{\sqrt{\delta}}(i\sigma d_{yz,\sigma} + d_{zx,\sigma} + id_{xy,\sigma})$ annihilates the $J_{eff} = 1/2$ doublet in the quasiparticle excitations, $|J = 1/2, J_{z} = \pm 1/2 \rangle = \frac{1}{\sqrt{2}} |0 \rangle$. The c-axis stacking of dPSCO is then controlled by $\eta_{i}$ as it takes on values of $\pm 1$ for Ir site $i$ in different IrO$_2$ layers. For example, to generate the $\oplus \ominus \ominus \ominus$ stacking pattern for dPSCO, $\eta_{i}$ take the value of $+1, +1, -1,$ and $-1$, respectively, for lattice site $i$ in the four IrO$_2$ layers. We fix the the stacking pattern of CAF to be $+−−+$, and try to find the energetically preferred stacking pattern of dPSCO in the coexistence state.

The interlayer hoppings are chosen to be $t_{z} = (20, −20)$ meV where the $+−−$ CAF is the magnetic ground state. At a given strength of dPSCO, $\Delta$, we obtain the coexistence states of $+−−$ CAF with four possible c-axis stacked dPSCO self-consistently, and then compare their energies to find the preferred stacking pattern for the dPSCO. The energies of the coexistence states with $\oplus \ominus \ominus \ominus$, $\oplus \oplus \ominus$, and $\oplus \oplus \oplus \oplus$ dPSCO are plotted in Fig. 5(a) as a function of $\Delta$, with respect to that of the coexistence state with $\oplus \oplus \ominus \ominus$ dPSCO. It is clear that the $+−−+ / \oplus \ominus \ominus \ominus$ coexistence state is energetically favored over all other coexistence states and, as shown in Sec. II, its symmetry is compatible with available experimental observations on undoped Sr$_2$IrO$_4$ below the Néel temperature $T_{N}$.

The band dispersions of the four coexistence states with $+−−+$ CAF are plotted in Figs. 5(b-e) for $\Delta = 30$ meV. The dPSCO order breaks the $C_{2a}$ symmetry, splits the eight-fold degenerate band at $X$ point into two four-fold degenerate branches, giving rise to a band splitting $\sim 200$ meV at $X$ point. We note that the $C_{2a}$ symmetry is broken by the staggered tetragonal distortion of the IrO$_6$ octahedra at temperatures above $T_{\Omega}$ in undoped Sr$_2$IrO$_4$ [22,20]. However, without the important $d$-wave form factor, the tetragonal distortion is unable to capture the unconventional quasiparticle properties of Sr$_2$IrO$_4$ in both the undoped magnetic insulating phase and the electron-doped nonmagnetic phase.

IV. DISCUSSIONS AND SUMMARIES

The existence and the nature of a hidden order in Sr$_2$IrO$_4$ have been under intensive debate. After the observation of the anomalous SHG signal [27], alternative explanations without invoking loop currents were subse-
quently proposed by Matteo and Norman [41], including laser-induced rearrangement of the magnetic stacking and enhanced sensitivity to surface rather than bulk magnetism. Polarized neutron diffraction [28] and muon spin relaxation [29] measurements performed on undoped and hole-doped Sr$_2$IrO$_4$ revealed broken time-reversal symmetry below $T_N$. Meanwhile, a resonant X-ray scattering measurement [50] conducted on the electron-doped Sr$_2$IrO$_4$ has uncovered an incommensurate magnetic scattering in the pseudogap phase. These experimental observations support the idea that the pseudo-gap is associated with a symmetry-breaking hidden order. Recently, comprehensive experiments [35] conducted on undoped Sr$_2$IrO$_4$ have ruled out the possibility of laser-induced rearrangement of the magnetic stacking, and suggest that the surface-magnetization induced electric-dipole process in the SHG experiments can be strongly enhanced by SOC. However, the existence of a hidden order in Sr$_2$IrO$_4$ remains as a possible explanation for the experimental observations of symmetry breaking and unconventional quasiparticle excitations.

In this work, we have shown that the coexistence state of $+ - - -$ CAF and $\oplus \ominus \ominus$ dPSCO has all the symmetry properties compatible with the available experimental observations on the undoped Sr$_2$IrO$_4$ below the Néel temperature $T_N$. It is a magnetoelectric state that breaks the two-fold rotation $C_{2z}$, inversion $I$, and time-reversal $T$ symmetries. We then demonstrated its microscopic realization in a three-dimensional Hubbard model with spin-orbit coupling. Together with the fact that the highly unconventional quasiparticle properties observed in both the parent and electron-doped Sr$_2$IrO$_4$ can be described remarkably well by the dPSCO [32], the latter offers a promising candidate for the hidden order responsible for the pseudogap phase in the undoped and electron-doped iridates. The Néel temperature $T_N$ and the hidden order transition temperature $T_H$ are very close to each other in undoped Sr$_2$IrO$_4$. As a result, available experiments on undoped Sr$_2$IrO$_4$ could not tell us unambiguously the symmetry properties of the pseudogap phase. It is thus very desirable to carry out the optical SHG and neutron scattering experiments on the pseudogap phase in electron-doped Sr$_2$IrO$_4$. In the absence of magnetism, the dPSCO would preserve the spatial inversion and time-reversal symmetries, but lower the four-fold rotation symmetry to two-fold $C_{2z}$ due to the $d$-wave form factor.

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