Pseudo-spin rotation symmetry breaking by Coulomb interaction terms in spin–orbit coupled systems

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Abstract

By transforming from the pure-spin-orbital (t_{2g}) basis to the spin-orbital entangled pseudo-spin-orbital basis, the pseudo-spin rotation symmetry of the different Coulomb interaction terms is investigated under SU(2) transformation in pseudo-spin space. While the Hubbard and density interaction terms are invariant, the Hund’s coupling and pair-hopping interaction terms explicitly break pseudo-spin rotation symmetry systematically. The form of the symmetry-breaking terms obtained from the transformation of the Coulomb interaction terms accounts for the easy x–y plane anisotropy and magnon gap for the out-of-plane mode, highlighting the importance of mixing with the nominally non-magnetic J = 3/2 sector, and providing a physically transparent approach for investigating magnetic ordering and anisotropy effects in perovskite (Sr_{2}IrO_{4}) and other d_{5} pseudo-spin compounds.

Keywords: spin–orbit coupling, Coulomb interaction terms, spin-orbital entanglement, magnetic ordering and anisotropy, magnon excitations, three-orbital model, Hund’s coupling (Some figures may appear in colour only in the online journal)

1. Introduction

Arising from a novel interplay between crystal field, spin–orbit coupling (SOC) and intermediate-strength Coulomb interactions, the emergent quantum states which essentially determine the electronic and magnetic properties of the iridium based transition-metal oxides involve correlated motion of electrons in spin-orbital entangled states [1–3]. In the spin–orbit Mott insulator Sr_{2}IrO_{4} with d_{5} configuration, electronic states near the Fermi energy have dominantly J = 1/2 character, and important magnetic properties such as in-plane canted antiferromagnetic (AFM) order and magnon excitations have been extensively discussed in terms of the effectively single pseudo orbital (J = 1/2) picture [4–7]. Finite-interaction and finite-SOC effects are responsible for the strong zone-boundary magnon dispersion measured in resonant inelastic x-ray scattering (RIXS) studies, highlighting the observable effect of mixing between J = 1/2 and 3/2 sectors [8].

The Dzyaloshinskii–Moriya (DM) and pseudo-dipolar (PD) anisotropic interactions in the J = 1/2 sector, although weakly affected by the tetragonal splitting [9], are not the source of true anisotropy in Sr_{2}IrO_{4}, as they yield pseudo-spin canting with no magnon gap due to compensation. True anisotropy has been ascribed to the Hund’s coupling term (J_{H}) using strong-coupling expansion (including virtual excitations to J = 3/2 states) and numerical self-consistent calculation [10–14]. While Coulomb interactions were considered within the pure-spin-orbital basis (t_{2g} orbitals, pure spins) in above approaches, their treatment within a pseudo-spin-orbital basis, and the role of weak magnetism in the other two pseudo orbitals (J = 3/2 sector) on the J_{H}-induced easy-plane magnetic anisotropy and magnon gap (∼40 meV), as measured in recent resonant inelastic x-ray scattering (RIXS) studies [15–17], have not been elucidated. Furthermore, the pseudo-spin-orbital based approach can allow for a unified study...
of both intra-orbital (magnon) and inter-orbital (spin–orbit exciton) excitations within a single formalism.

Magnetic anisotropy is generally associated with spin rotation symmetry breaking. Therefore, a general pseudo-spin rotation symmetry analysis of the different Coulomb interaction terms, treating all three pseudo orbitals on the same footing, can provide additional insight into the origin of true magnetic anisotropy in Sr$_2$IrO$_4$, arising from the interplay of spin-orbital entanglement and Coulomb interaction. Due to the spin-orbital entanglement, the same pseudo-spin rotation for all three pseudo orbitals ($l = 1, 2, 3$) corresponds to different pure-spin rotations for the three pure ($t_{2g}$) orbitals. This follows directly from the relation $\psi_\mu = \sigma_\mu \sum_\nu \epsilon_{\mu \nu} \psi_\nu$ between the fermionic field operators in the pure-spin-orbital basis ($\mu = yz, xz, xy$) and pseudo-spin-orbital basis ($l = 1, 2, 3$), where the Pauli matrices $\sigma_\mu = \sigma_x, \sigma_y, \sigma_z$ corresponding to the three pure orbitals $\mu = yz, xz, xy$. The same SU(2) transformation $\psi_\mu \rightarrow \psi'_\mu = [U]\psi_\mu$ for all three pseudo orbitals corresponds to different SU(2) transformations $\psi_\mu \rightarrow \psi''_\mu = [U]\psi_\mu$ for the three pure orbitals due to the pseudo-orbital dependence of $[U]\sigma_\mu [U]^\dagger \sigma_\mu$. Therefore, the question of how the different Coulomb interaction terms transform under the same pseudo-spin rotation for all three pseudo orbitals assumes importance. In other words, while all Coulomb interaction terms are invariant under the same pure-spin rotation for all three pure orbitals, does this invariance hold under same pseudospin-rotation in the pseudo-spin-orbital basis? Pseudo-spin rotation symmetry breaking by any Coulomb interaction term would imply true magnetic anisotropy and gapped magnon spectrum.

In this paper, we will show that while the Hubbard ($U$) and density ($U'$) interaction terms do preserve pseudo-spin rotation symmetry, the Hund’s coupling ($J_H$) and pair-hopping ($J_P$) interaction terms explicitly break this symmetry systematically. This symmetry breaking results in (on-site) anisotropic interactions between moments in the $J = 1/2$ and $J = 3/2$ sectors, highlighting the importance of the weak magnetism in the nominally filled $J = 3/2$ sector as well as the mixing between the two sectors. Magnetic anisotropy will not survive in the large SOC limit when the two sectors become effectively decoupled. The physically transparent approach for investigating magnetic ordering and anisotropy effects will be illustrated for the perovskite compound Sr$_2$IrO$_4$, where the sign of $J = 3/2$ sector moments directly yields easy $x$–$y$ plane anisotropy.

The structure of this paper is as below. After introducing the transformation between the pure-spin-orbital and pseudo-spin-orbital bases in section 2, a symmetry analysis of the individual Coulomb interaction terms is carried out in section 3, where the Hund’s coupling and pair hopping interaction terms are shown to explicitly break pseudo-spin rotation symmetry. The transformed Coulomb interaction terms are obtained in section 4, explicitly showing that the symmetry breaking terms account for the easy $x$–$y$ plane anisotropy in Sr$_2$IrO$_4$, as illustrated for the ground state energy in section 5, and for magnon excitations and anisotropy gap in section 6. Comparison with an independent $t_{2g}$ orbital based approach is discussed in section 7, which provides confirmation of the above symmetry analysis and also illustrates the significant simplification achieved in the pseudo-spin-orbital basis. After a critical comparison (section 8) of different but equivalent approaches for studying magnetic anisotropy in Sr$_2$IrO$_4$, some conclusions are finally presented in section 9.

2. Pseudo-spin-orbital basis

Due to large crystal-field splitting ($\sim 3$ eV) in the IrO$_6$ octahedra, low-energy physics in $d^9$ iridates is effectively described by projecting out the empty $e_g$ levels which are well above the $t_{2g}$ levels. Spin–orbit coupling (SOC) further splits the $t_{2g}$ states into $J = 1/2$ doublet ($m_f = \pm 1/2$) and $J = 3/2$ quartet ($m_f = \pm 1/2, \pm 3/2$), with an energy gap of $3\lambda/2$ (figure 1). Nominally, four of the five electrons fill the $J = 3/2$ states, leaving one electron for the $J = 1/2$ sector, rendering it magnetically active in the ground state.

In the $(yz \downarrow, xz \downarrow, xy \downarrow)$ basis, the SOC Hamiltonian becomes block diagonal with two $[3 \times 3]$ blocks. There is no mixing between the $(\downarrow, \downarrow)$ and $(\uparrow, \uparrow, \downarrow)$ sectors, which can therefore be treated as pseudo spins $\uparrow$ and $\downarrow$ for the three doubly degenerate eigenstates (pseudo orbitals). [6] Corresponding to the three Kramers pairs $(J, m_f)$ above, the pseudo-spin-orbital basis states $|l, \tau\rangle$ for the three pseudo orbitals ($l = 1, 2, 3$), with pseudo spins $(\tau = \uparrow, \downarrow)$ each, have the form:

$$|l = 1, \tau = \sigma\rangle = \frac{1}{\sqrt{3}} \left[ \begin{array}{c} \psi_{yz, \sigma} \pm i \psi_{xz, \bar{\sigma}} \pm i \psi_{xy, \sigma} \end{array} \right]$$

$$|l = 2, \tau = \sigma\rangle = \frac{1}{\sqrt{6}} \left[ \begin{array}{c} \psi_{yz, \sigma} \pm i \psi_{xz, \bar{\sigma}} \pm 2 \psi_{xy, \sigma} \end{array} \right]$$

$$|l = 3, \tau = \sigma\rangle = \frac{1}{\sqrt{2}} \left[ \begin{array}{c} \psi_{yz, \sigma} \pm i \psi_{xz, \bar{\sigma}} \end{array} \right]$$

where $\psi_{yz, \sigma}$, $\psi_{xz, \sigma}$, $|xy, \sigma\rangle$ are the $t_{2g}$ basis states and the signs $\pm$ correspond to spins $\sigma = \uparrow$ or $\downarrow$. The coherent superposition of different-symmetry $t_{2g}$ orbitals, with opposite spin polarization between $xz/yz$ and $xy$ levels implies spin-orbital entanglement, and also imparts unique extended 3D shape to the pseudo-orbitals $l = 1, 2, 3$, as shown in figure 1. The pseudo-spin dynamics in iridate heterostructures are gaining interest as their magnetic properties are much more sensitive to structural distortion compared to pure spin systems due to spin-orbital entanglement [18, 19].

Taking the conjugate to express the above basis transformation in terms of the $(l, \tau)$ and $(\mu, \sigma)$ states, and rewriting in terms of the corresponding fermionic field operators:

$$\psi_{l} = \begin{pmatrix} a_{l\uparrow} \\ a_{l\downarrow} \end{pmatrix} \quad \text{and} \quad \psi_{\mu} = \begin{pmatrix} a_{\mu\uparrow} \\ a_{\mu\downarrow} \end{pmatrix}$$

involve the annihilation operators for the pseudo orbitals ($l = 1, 2, 3$, $\tau = \uparrow, \downarrow$) and the $t_{2g}$ orbitals ($\mu = yz, xz, xy$, $\sigma = \uparrow, \downarrow$), we obtain (using Pauli matrices):

$$\psi_{l} = \frac{1}{\sqrt{3}} \left[ \sigma_x \psi_{yz} + \sigma_y \psi_{xz} + \sigma_z \psi_{xy} \right]$$
Kramers pairs along with their orbital shapes. The colors represent the weights of pure spins (↑-red, ↓-blue) in each pair.

\[ \psi_2 = \frac{1}{\sqrt{6}} \left[ \sigma_x \psi_{xz} + \sigma_y \psi_{x \pm z} - 2 \sigma_z \psi_{xy} \right] \]

\[ \psi_3 = \frac{1}{\sqrt{2}} \left[ \sigma_x \psi_{x \pm z} - \sigma_y \psi_{xz} \right]. \quad (3) \]

Inverting the above transformation yields the \( t_{2g} \) basis states represented in terms of the pseudo-spin-orbital basis states:

\[ \psi_{xz} = \sigma_x \left[ \frac{1}{\sqrt{3}} \psi_1 + \frac{1}{\sqrt{6}} \psi_2 + \frac{1}{\sqrt{2}} \psi_3 \right] \]

\[ \psi_{x \pm z} = \sigma_y \left[ \frac{1}{\sqrt{3}} \psi_1 + \frac{1}{\sqrt{6}} \psi_2 - \frac{1}{\sqrt{2}} \psi_3 \right] \]

\[ \psi_{xy} = \sigma_z \left[ \frac{1}{\sqrt{3}} \psi_1 - \frac{1}{\sqrt{2}} \psi_2 \right]. \quad (4) \]

The above equations are convenient for transforming the hopping and Coulomb interaction terms to the pseudo-spin-orbital basis, and can be expressed in the compact form:

\[ \psi_\mu = \sigma_\mu \sum_{l=1,2,3} c_{\mu l} \psi_l \quad (5) \]

where \( \sigma_\mu = \sigma_x, \sigma_y, \sigma_z \) for the three orbitals \( \mu = xz, x \pm z, xy \), respectively, and the (real) transformation coefficients \( c_{\mu l} \) are explicitly shown in equation (4).

### 3. Pseudo-spin rotation symmetry breaking

We consider the on-site Coulomb interaction terms in the \( t_{2g} \) basis (\( \mu, \nu = xz, x \pm z, xy \)):

\[ \mathcal{H}_{\text{int}} = \sum_{\mu \neq \nu} n_{\mu \uparrow} n_{\nu \downarrow} + U \sum_{\mu} n_{\mu \uparrow} n_{\mu \downarrow} + \left( U' - J_H \right) \left[ \sum_{\mu \neq \nu} \sigma_{\mu} \sigma_{\nu} \right] \]

\[ + \sum_{\mu \neq \nu} a_{\mu \downarrow} \sigma_{\mu} a_{\nu \uparrow} + U' \sum_{\mu} n_{\mu \uparrow} n_{\mu \downarrow} - J_H \sum_{\mu \neq \nu} S_{\mu \downarrow} \cdot S_{\nu \uparrow} \]

\[ + J_P \sum_{\mu \neq \nu} a_{\mu \downarrow} a_{\nu \uparrow} \]

\[ = \sum_{\mu \neq \nu} n_{\mu \uparrow} n_{\nu \downarrow} + U' \sum_{\mu} n_{\mu \uparrow} n_{\mu \downarrow} - J_H \sum_{\mu \neq \nu} S_{\mu \downarrow} \cdot S_{\nu \uparrow} \]

\[ + J_P \sum_{\mu \neq \nu} a_{\mu \downarrow} a_{\nu \uparrow} \quad (6) \]

including the intra-orbital (\( U \)) and inter-orbital (\( U' \)) density interaction terms, the Hund’s coupling term (\( J_H \)), and the pair hopping interaction term (\( J_P = J_{H} \)). Here \( a_{\mu \downarrow} \) and \( a_{\mu \uparrow} \) are the creation and annihilation operators for site \( \mu \), orbital \( \nu \), spin \( \sigma = \uparrow, \downarrow \), the density operator \( n_{\mu \sigma} = a_{\mu \sigma}^\dagger a_{\mu \sigma} \), the total density operator \( n_{\mu \sigma} = n_{\mu \uparrow} + n_{\mu \downarrow} = \psi_{\mu \sigma}^\dagger \psi_{\mu \sigma} \), and \( U'' = U' - J_H/2 \). All interaction terms above are SU(2) invariant and thus possess pure-spin rotation symmetry. In the following, we consider the transformation of individual Coulomb interaction terms to the pseudo-spin-orbital basis using equation (5), and examine their SU(2) transformation behavior in pseudo-spin space.

#### 3.1. Total density operator

For the total density operator (for site \( i \)), we obtain using equation (5):

\[ n_{\mu} = \psi_{\mu \uparrow}^\dagger \psi_{\mu \downarrow} = \sum_{l,m} c_{\mu l} c_{\mu m} \psi_{l \uparrow}^\dagger \psi_{m \downarrow} \quad (7) \]

where we have used \( \sigma_\mu^\dagger = \sigma_\mu \) and \( \sigma_\mu^2 = 1 \). Now, under the SU(2) transformation in pseudo-spin space (same for all three pseudo orbitals \( i \)):

\[ \psi_\mu \rightarrow \psi_{\mu i} = [U] \psi_\mu \quad (8) \]

the \( \psi_{\mu l} \psi_{m \downarrow} \) terms are invariant, and the total density operator is therefore SU(2) invariant. Therefore, the density interaction terms \( (U'') \) in equation (6) and the Hubbard interaction terms \( (U) \) (using \( n_{\uparrow} + n_{\downarrow} = n_{\uparrow} + 2n_{\downarrow} \)) are SU(2) invariant and possess spin rotation symmetry in pseudo-spin space.

#### 3.2. Pair hopping interaction term

For the pair hopping interaction term (for site \( i \)), we obtain:

\[ J_H \sum_{\mu \neq \nu} a_{\mu \downarrow} a_{\nu \uparrow} \psi_{\mu \sigma_i} \psi_{\nu \sigma_j} = J_H \sum_{\mu \neq \nu} a_{\mu \downarrow} a_{\nu \uparrow} a_{\mu \sigma_i} a_{\nu \sigma_j} \]

\[ = J_H \frac{1}{2} \left( a_{\mu \downarrow} a_{\nu \uparrow} + a_{\mu \uparrow} a_{\nu \downarrow} \right) \]

\[ \times \left( a_{\mu \sigma_i} a_{\nu \sigma_j} + a_{\mu \sigma_j} a_{\nu \sigma_i} \right) \]

\[ = J_H \left( \psi_{\mu \sigma_i} \psi_{\nu \sigma_j} \right)^2, \quad (9) \]

which is SU(2) invariant and possesses spin-rotation symmetry in pure-spin space. However, SU(2) invariance is lost in pseudo-spin space, as shown below. Again, using equation (5) to transform to the pseudo-spin-orbital basis, we obtain:

\[ \psi_{\mu l} \psi_{m \downarrow} = \sum_{l,m} c_{\mu l} c_{\mu m} \psi_{l \sigma_i} \psi_{m \sigma_j} = \sum_{l,m} c_{\mu l} c_{\mu m} \psi_{l \sigma_i}^\dagger \psi_{m \sigma_j} \]

where we have taken \( \mu = xz, \nu = x \pm z \) to illustrate the operations with Pauli matrices. Now, under the SU(2) transformation in pseudo-spin space (equation (8)), the last term in equation (10):

\[ \psi_{\mu}^\dagger (i \sigma_j) \psi_{m \downarrow} \rightarrow \psi_{\mu}^\dagger (i \sigma_j) [U] \psi_{m \downarrow} \neq \psi_{\mu}^\dagger (i \sigma_j) \psi_{m \downarrow}, \quad (11) \]
showing that $\psi_i^\dagger \psi_j$ is not SU(2) invariant. The pair-hopping interaction term therefore explicitly breaks pseudo-spin rotation symmetry.

3.3. Hund’s coupling term

For this term involving the pure-spin rotationally symmetric interaction $S_{\mu} \cdot S_{\nu}$, we consider the spin density operator (for site $i$), and obtain using equation (5):

$$2 S_{\mu} = \psi_i^\dagger \sigma \psi_i = \sum_{lm} c_{\mu l} c_{\nu m} \psi_i^\dagger (\sigma_{\mu l} \sigma_{\nu m}) \psi_m$$  \(12\)

which transforms under the SU(2) transformation (equation (8)) to:

$$2 S_{\mu} \rightarrow 2 S_{\mu}' = \psi_i^\dagger \sigma \psi_i = \sum_{lm} c_{\mu l} c_{\nu m} \psi_i^\dagger \left[ \left| U \right|^\dagger \sigma_{\mu l} \sigma_{\nu m} \left| U \right| \right] \psi_m.$$  \(13\)

We now consider the term in brackets above for the case $\sigma_{\mu l} = \sigma_x$ (yz orbital) and represent it in terms of a rotation operation in spin space:

$$\left[ \left| U \right|^\dagger \right] \left[ \begin{array}{ccc} \sigma_x & 0 & 0 \\ 0 & \sigma_y & 0 \\ 0 & 0 & \sigma_z \end{array} \right] \left[ \begin{array}{c} \sigma_x \sigma_z \end{array} \right] = \left[ \left| U \right|^\dagger \right] \left[ \begin{array}{ccc} \sigma_x & 0 & 0 \\ 0 & \sigma_y & 0 \\ 0 & 0 & \sigma_z \end{array} \right] \left[ \begin{array}{c} \sigma_x \sigma_z \end{array} \right]$$  \(14\)

where

$$R_U \left( \begin{array}{c} \sigma_x \\ \sigma_y \\ \sigma_z \end{array} \right) = \left( \begin{array}{c} \sigma_x' \\ \sigma_y' \\ \sigma_z' \end{array} \right) = \left[ \left| U \right|^\dagger \right] \left( \begin{array}{c} \sigma_x \\ \sigma_y \\ \sigma_z \end{array} \right)$$  \(15\)

shows the spin rotation by the rotation matrix $R_U$ corresponding to the SU(2) transformation $|U|$, and

$$R_\pi = \frac{1}{2} \left( \begin{array}{ccc} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{array} \right)$$  \(16\)

is the rotation matrix corresponding to $\pi$ rotation about the $x$ axis.

Similarly, for the $S_y$ operator with $\nu = y$ (xz orbital), we will obtain the product $R_\pi R(U)$. Therefore, $S_{\mu} \cdot S_{\nu}$ interaction term will yield the matrix product:

$$[R_\pi (U)] R_\pi R(U) = \tilde{R}(U) R_\pi R(U)$$  \(17\)

where $\tilde{R}(U)$ is the transpose of $R(U)$ and we have used $R_\pi = R_\pi (\pi)$ for the diagonal matrix. Finally, since

$$\tilde{R}(U) R_\pi (\pi) R(U) \neq R_\pi (\pi) R_\pi (\pi)$$  \(18\)

as $R_\pi (\pi) R_\pi (\pi) \neq 1$, the Hund’s coupling term $S_{\mu} \cdot S_{\nu}$ is not pseudo-spin SU(2) invariant and therefore does not possess pseudo-spin rotation symmetry.

The above symmetry analysis shows that the Hund’s coupling and pair hopping interaction terms explicitly break pseudo-spin rotation symmetry. It is important to note here that the spin rotation symmetry is broken systematically. In other words, it is broken for each term in $S_{\mu} \cdot S_{\nu}$ involving the summations over $(l, m)$ and $(l', m')$ for $S_{\mu}$ and $S_{\nu}$ in equation (12), and similarly for the pair hopping interaction term in equation (10).

4. Transformed Coulomb interaction terms

In the following, we illustrate the transformation of the different Coulomb interaction terms to the pseudo-spin-orbital basis, starting with $l = 1$ ($J = 1/2$) sector of the intra-pseudo-orbital interaction terms. Similar transformation to the $J$ basis has been discussed recently, focusing only on the density interaction terms [20]. Considering first the pair-hopping interaction term (equation (9)), and retaining only the $l = m = l' = m' = 1$ terms (indicated by $\rightarrow$ below), we obtain:

$$\frac{J_H}{2} \sum_{\mu \neq \nu} \langle \psi_i^\dagger \psi_j \rangle^2 = \frac{J_H}{2} \sum_{\mu \neq \nu} \left[ \sum_{lm} c_{\mu l} c_{\nu m} \psi_i^\dagger (\sigma_{\mu l} \sigma_{\nu m}) \psi_m \right] \times \left[ \sum_{l'm'} c_{\mu l'} c_{\nu m'} \psi_i^\dagger (\sigma_{\mu l'} \sigma_{\nu m'}) \psi_{m'} \right] \rightarrow J_H \left[ \begin{array}{ccc} c_{1z,1}^2 \sum_{l,m} \left\{ \psi_i^\dagger (U \psi_j) \right\} \end{array} \right]$$

$$= -4 J_H \left[ c_{\lambda 1}^2 \sum_{l,m} \left\{ \psi_i^\dagger (U \psi_j) \right\} \right]$$

Similarly, for the Hund’s coupling term in equation (6), we obtain:

$$-2 J_h \sum_{\mu < \nu} S_{\mu}^i S_{\nu}^i = -\frac{J_h}{2} \sum_{\mu < \nu} \left[ \sum_{lm} c_{\mu l} c_{\nu m} \psi_i^\dagger (\sigma_{\mu l} \sigma_{\nu m}) \psi_m \right] \times \left[ \sum_{l'm'} c_{\mu l'} c_{\nu m'} \psi_i^\dagger (\sigma_{\mu l'} \sigma_{\nu m'}) \psi_{m'} \right] \rightarrow -\frac{J_h}{2} \sum_{\mu < \nu} \left[ c_{\mu l} c_{\nu m} \psi_i^\dagger (U \psi_j) \right]$$

as the product $c_{\mu l} c_{\nu m}$ is identical for all three orbital pairs, and for the rotation matrices (equation (15)), we have $R_\pi R_y + R_y R_\pi + R_\pi R_z = -1$. Although the pair-hopping and
Hund’s coupling interaction terms generally break pseudo-spin rotation symmetry, within the magnetically active \( l = 1 \) (\( J = 1/2 \)) sector, they individually yield classically isotropic terms of the form \( S_i \cdot S_j \). This follows from the special symmetry within this sector, as reflected by the identical coefficients \( c_{\mu \nu} \) for all three orbitals \( \mu = y, z, x_y \) for \( l = 1 \) (equation (4)).

Finally, from the remaining \( J_H \) term (in the \( U'' \) term of equation (6)), again retaining only the \( l = m = \ell = m' = 1 \) term, we obtain (using equation (7) for site i):

\[
-J_H/2 \sum_{\mu < \nu} n_\mu n_\nu = -J_H/2 \sum_{\mu < \nu} \left( \sum_{l,m} c_{\mu \nu} c_{l,m} \psi^\dagger_1 \psi^\dagger_m \right) \\
\times \sum_{l',m'} c_{l',m'} c_{l,m} (\psi^\dagger_l \psi^\dagger_{l'}) (\psi^\dagger_m \psi^\dagger_{m'}) \\
\times \sum_{\mu < \nu} c_{l,m}^2 c_{l',m'}^2 (\psi^\dagger_l \psi^\dagger_{l'})^2 \\
= -J_H/2 \sum_{\mu < \nu} c_{l,m}^2 (n_{\mu 1} + n_{\nu 1})(n_{\mu 1} + n_{\nu 1}) \\
= 2 J_H/9 S_i \cdot S_i - J_H/3 n_i
\]  

(21)

where we have used \( n_{\mu 1}^2 = n_{\mu} \) and \( 2n_{\mu 1} n_{\nu 1} = n_i - (4/3) S_i \cdot S_i \). Collecting all the \( S_i \cdot S_i \) interaction terms resulting from the pair-hopping, Hund’s coupling, and density interaction terms corresponding to \( J_H \), as obtained in equations (19)–(21), yields an exact cancellation.

Similarly, considering the other two intra-pseudo-orbital cases (\( l = m = \ell = m' = 2, 3 \)) in the \( J = 3/2 \) sector, we obtain from the three explicitly \( J_H \) interaction terms:

\[
-J_H/2 \sum_{\mu < \nu} (\psi^\dagger_\mu \psi^\dagger_\nu)^2 \\
-4 J_H/9 S_i \cdot S_2 + J_H/3 S_2 \cdot S_2 - J_H S_2 \cdot S_3 \\
-J_H/18 S_i \cdot S_2 + J_H/3 S_2 \cdot S_2 + J_H/2 S_3 \cdot S_3 \\
-J_H S_2 \cdot S_3 \\
-J_H/2 \sum_{\mu < \nu} (\psi^\dagger_\mu \psi^\dagger_\nu)^3 \\
= J_H/6 S_2 \cdot S_2 + J_H/6 S_3 \cdot S_3 - J_H/4 (n_2 + n_3)
\]  

(22)

explicitly showing the symmetry breaking contributions \((S_2 \cdot S_2)\) from the pair-hopping and Hund’s coupling interaction terms, as expected from the SU(2) transformation analysis in section 3, in the absence of the special symmetry in the \( J = 3/2 \) sector. However, since \( S_2 \cdot S_2 = (1/4)(n_l + n_i) - 2n_{11} \), \((1/3) S_i \cdot S_i \) for all three components \( \alpha = x, y, z \) of \( S = 1/2 \) quantum spin operators \( S_\alpha = (1/2) \psi^\dagger \sigma_{\alpha} \psi \), there is no true magnetic anisotropy even if classically anisotropic terms such as \( S_2 \cdot S_2 \) are present, as in equation (22).

Substituting \( S_2 \cdot S_2 = (1/3) S_i \cdot S_i \) in equation (22) yields an exact cancellation of the three explicitly \( J_H \) interaction terms in the \( J = 3/2 \) sector also. Therefore, the Hubbard-like (or equivalently \( S_i \cdot S_i \)) intra-pseudo-orbital interaction terms in all three sectors result only from the \( U \) and \( U'' \) terms in equation (6). Using similar analysis as above, and dropping one-particle density terms as in equation (21), one obtains (for site i):

\[
U \sum_{\mu < \nu} n_\mu n_\nu + U'' \sum_{\mu < \nu} n_\mu n_\nu \sim \left( U + 2U'' \right) n_1 n_1 \\
+ \left( U + U'' \right) (n_2 n_2 + n_3 n_3) \\
\]

(23)

From the above discussion it follows that true magnetic anisotropy results only from the inter-orbital anisotropic interaction terms such as \( S_\alpha S_\beta \), with \( \ell \neq l \). Including inter-pseudo-orbital cases such as \( l = m, \ell = m' \) and \( l = m', \ell = m \) in equations (19) and (20), and keeping all interaction terms relevant for the present study (Hubbard, Hund’s coupling, and density), we obtain (for site i):

\[
H_{int}(i) = \left( U + 2U''/3 \right) n_{11} + \left( U + U''/2 \right) [n_{22} + n_{33}] \\
- \left( U - U''/3 \right) 2S_1 \cdot S_2 - \left( U - U'' - 2J_H \right) \]  

\[
\times [(2S_1 + S_3) \cdot S_1 + 2J_H(S_2^2 - S_1^2)] \\
+ \left( U + 5U''/6 - 3J_H \right) [n_2 n_2 + n_3 n_3] \\
+ \left( U + 11U''/12 - 6J_H \right) n_{23}
\]

(24)

where \( S_m = \psi^\dagger_m \psi_\mu \psi_\nu \psi_m \) and \( n_m = \psi^\dagger_m \psi_\mu \psi_\nu \psi_m \) are the spin and charge density operators. Using the spherical symmetry condition \((U' = U - 2J_H)\), the transformed interaction Hamiltonian (24) simplifies to:

\[
H_{int}(i) = \left( U - 4J_H/3 \right) n_{11} + (U - J_H) [n_{22} + n_{33}] \\
- 4J_H S_1 \cdot S_2 + 2J_H [S_2^2 - S_1^2] \\
+ \left( U - 13J_H/6 \right) [n_2 n_2 + n_3 n_3] \\
+ \left( U - 7J_H/3 \right) n_{23}
\]

(25)

In the above equation, the Hubbard-like terms \( U_m n_m n_m \) and \( U_\mu n_\mu n_\mu \) the Hund’s coupling-like term \( S_2 \cdot S_2 \), and the density terms \( n_m n_m \), are all invariant under pseudo-spin rotation and the corresponding SU(2) transformation \( \psi^\dagger_m \rightarrow \psi^\dagger_m \), therefore, only the interaction terms \( S_1^2 S_2^2 \) and \( S_1^2 S_3^2 \) between moments in the \( J = 1/2 \) and \( J = 3/2 \) sectors are responsible for the magneto-crystalline anisotropy in \( \text{Sr}_2\text{IrO}_4 \), highlighting the importance of the weak magnetism in the nominally filled \( J = 3/2 \) sector due to the mixing between the two sectors. Magnetic anisotropy will not survive in the large SOC limit when the two sectors become effectively decoupled. As shown below, an easy x-y plane anisotropy is obtained from
5. Easy x–y plane anisotropy

We consider the various interaction terms in equation (25) in the Hartree–Fock (HF) approximation, focussing on the staggered field terms corresponding to ($\pi$, $\pi$) ordered AF state on the square lattice. For general ordering direction with components $\Delta_z = (\Delta_x, \Delta_y, \Delta_t)$, the staggered field term for sector $l$ in the pseudo-orbital basis is given by:

$$\mathcal{H}_{\text{e}}(l) = \sum_{k, \alpha} \psi^{\dagger}_{k\alpha} (-s \mathbf{\tau} \cdot \mathbf{\Delta}_l) \psi_{k\alpha}$$

$$= \sum_{k, \alpha} -s \psi^{\dagger}_{k\alpha} \left( \Delta^x_l \psi^{\dagger}_{k\alpha} - i \Delta^y_l - i \Delta^z_l \right) \psi_{k\alpha}$$

(26)

where $\psi^{\dagger}_{k\alpha} = (a^{\dagger}_{k\alpha}, a^{\dagger}_{-k\alpha})$, $s = \pm 1$ for the two sublattices A/B, and the staggered field components $\Delta^{x,y,z}_l$ are self-consistently determined from:

$$2 \Delta^x_l = \delta U_l m_0^l + \frac{2J_H}{3} m_0^l + J_H (m_0^l - m_0^l) \delta \alpha_z$$

$$2 \Delta^y_l = \delta U_l m_0^l + \frac{2J_H}{3} m_0^l - J_H m_0^l \delta \alpha_y$$

$$2 \Delta^z_l = \delta U_l m_0^l + J_H m_0^l \delta \alpha_z$$

(27)

in terms of the staggered pseudo-spin magnetization components $m_0^{x,y,z}$. In practice, it is easier to choose set of $\Delta^{x,y,z}_l$ and self-consistently determine the Hubbard-like interaction strengths $U_l = U - \frac{1}{2} J_H$ and $U_l = U - J_H$ using equation (27). The interaction strengths are related by $U_{l=1,2,3} = U_{l=1} + J_H / 3$.

Transforming the staggered-field term back to the three-orbital basis ($\sigma|x,\sigma|y,\sigma|xy\rangle$), and including the crystal field, SOC and band terms, [6] we have considered $\mathcal{H}_{\text{HF}} = \mathcal{H}_{\text{SO}} + \mathcal{H}_{\text{band+ef}} + \mathcal{H}_{\text{a}}$ in our band structure and spin fluctuation analysis, where

$$\mathcal{H}_{\text{band+ef}} = \sum_{k, \sigma, \sigma'} \psi^{\dagger}_{k\sigma} \left[ \begin{array}{ccc} e_{k}^{\sigma} & 0 & 0 \\ 0 & e_{k}^{\sigma'} & 0 \\ 0 & 0 & e_{k}^{\sigma} + \epsilon_{k}^{\sigma'} \end{array} \right] \delta_{\sigma\sigma'}$$

$$+ \left( \begin{array}{ccc} e_{k}^{\sigma} & e_{k}^{\sigma} & e_{k}^{\sigma} \\ e_{k}^{\sigma} & e_{k}^{\sigma} & e_{k}^{\sigma} \\ e_{k}^{\sigma} & e_{k}^{\sigma} & e_{k}^{\sigma} \end{array} \right) \delta_{\sigma\sigma'} \right] \psi_{k\sigma\sigma'}$$

(28)

in the composite three-orbital, two-sublattice basis. The crystal field induced tetragonal splitting is included as the $xy$ orbital energy offset $\epsilon_{k}^{xy}$ from the degenerate $yz/xz$ orbitals. The different hopping terms in equation (28) connecting the same ($s = s'$) and opposite ($s \neq s'$) sublattice(s) are given by:

$$e_{k}^{xy} = -2t_1 \cos k_x + \cos k_y$$

$$e_{k}^{xy'} = -4t_2 \cos k_x \cos k_y - 2t_3 (2 \cos k_x + \cos k_y)$$

Here $t_1$, $t_2$, $t_3$ are respectively the first, second, and third neighbor hopping terms for the $xy$ orbital. For the $yz$ ($xz$) orbital, $t_4$ and $t_5$ are the NN hopping terms in $y$($x$) and $x$($y$) directions, respectively. Mixing between $xz$ and $yz$ orbitals is represented by the NN hopping term $t_m$. We have taken values of the tight-binding parameters ($t_1, t_2, t_3, t_4, t_5, t_m, \epsilon_{xy}, \lambda) = (1.0, 0.5, 0.25, 1.028, 0.167, 0.2, -0.7, 1.35)$ in units of $t_1$, where the energy scale $t_1 = 280$ meV. Using above parameters, the calculated electronic band structure shows AFM insulating state and mixing between pseudo-orbital sectors [6, 8].

For the band dispersion terms in equation (28) and henceforth, we have used the crystal (global) coordinate axes referred to as $x, y, z$ for convenience. The spin coordinate axes are chosen to align with the crystal axes. If required, a site-dependent spin rotation allows one to transform back to the local spin coordinate axes common with the octahedral axes due to SOC.

5.1. Canted AFM state

The octahedral-rotation-induced orbital mixing hopping term ($t_m$) between $yz$ and $xz$ orbitals generates PD ($\Delta S^x, \Delta S^y$) and DM [$\cdot (S \times S)$] anisotropic interactions in the strong coupling limit [8]. However, the AFM-state energy is invariant with respect to change of ordering direction from $z$ axis to $x$-$y$ plane provided spins are canted at the optimal canting angle, thus preserving the gapless Goldstone mode. Figure 2(a) shows the variation of AFM-state energy with canting angle ($\phi$) for ordering in the $x$-$y$ plane. The energy minimum at the optimal canting angle is exactly degenerate with the energy for $z$-direction ordering. This absence of true anisotropy in Sr$_2$IrO$_4$ due to octahedral rotation alone is consistent with the general gauge transformation analysis showing that the spin-dependent hopping terms arising from the orbital-mixing terms in the three-orbital model can be gauged away.

5.2. $J_{\mu}$-induced easy-plane anisotropy

The Hund’s-coupling-induced easy-plane magnetic anisotropy is explicitly shown in figure 2(b) by the variation of AFM-state energy with polar angle $\theta$ corresponding to staggered field orientation in the $x$–$z$ plane, with $\Delta_z = (\Delta + \Delta_{\mu}) \cos \theta$ and $\Delta_l = \Delta \sin \theta$. Here $\Delta$ represents the spin-rotationally-symmetric part ($U_l m_1 + \frac{2J_H}{3} m_2$)/2 of the staggered field and $\Delta_{\mu} = J_{\mu} (m_0^l - m_0^l)$/2 is the symmetry-breaking term, as seen from equation (27). We have taken $\Delta = 0.9, \Delta_{\mu} = -0.01$, and the orbital mixing hopping term $t_m$ has been set to zero for simplicity. Converting from energy per state as shown in figure 2 to simply energy per site ($\times$10 occupied states for each $k$), yields the magnetic anisotropy energy $E_{\text{HF}}(z) - E_{\text{HF}}(x) \approx 0.012$, which is comparable to the $\Delta_{\mu}$ magnitude. The simplified analysis presented in this section, with staggered field only for the $l = 1$ orbital, serves...
to explicitly illustrate the magnetic anisotropy features within our three-band-model calculation.

6. Magnon excitations and anisotropy gap

In view of the Hund’s-coupling-induced easy \( x-y \) plane anisotropy as discussed above, we consider the \( x \)-ordered AFM state. The magnon propagator corresponding to transverse spin fluctuations should therefore yield one gapless mode (y direction) and one gapped mode (z direction). Accordingly, we consider the time-ordered magnon propagator:

\[
\chi(q, \omega) = \int \frac{d\tau}{\Omega} \sum_{ij} e^{i\omega(\tau - \tau')} e^{-iq(\tau - \tau)}
\times \langle \Psi_0 | T[\hat{S}_{i,m}^\alpha(\tau) \hat{S}_{j,n}^\beta(\tau')] | \Psi_0 \rangle \tag{30}
\]

involving the transverse \( \alpha, \beta = y, z \) components of the pseudo-spin operators \( \hat{S}_{i,m}^\alpha \) and \( \hat{S}_{j,n}^\beta \) for pseudo orbitals \( m \) and \( n \) at lattice sites \( i \) and \( j \).

In the random phase approximation (RPA), the magnon propagator is obtained as:

\[
[\chi(q, \omega)] = \frac{[\chi^0(q, \omega)]}{1 - 2[H] \chi^0(q, \omega)} \tag{31}
\]

where the bare particle–hole propagator:

\[
[\chi^0(q, \omega)]_{ab}^\alpha = \frac{1}{4} \sum_k \left[ \frac{\langle \hat{S}_{k-q}^\alpha | \hat{S}_{k}^\beta \rangle}{E_k^\alpha - E_k - \omega - i\eta} \right]
+ \left[ \frac{\langle \hat{S}_{k-q}^\beta | \hat{S}_{k}^\alpha \rangle}{E_k^\beta - E_k - \omega - i\eta} \right] \tag{32}
\]

was evaluated in the composite spin-orbital-sublattice basis (2 spin components \( \alpha, \beta = y, z \) \( \otimes \) 3 pseudo orbitals \( m = 1, 2, 3 \) \( \otimes \) 2 sublattices \( x, s' = \text{A, B} \)) by integrating out the fermions in the \( (\pi, \pi) \) ordered state. Here \( E_k^\alpha \) and \( \varphi_k \) are the eigenvalues and eigenvectors of the Hamiltonian matrix in the pseudo-orbital basis, the indices \( a, b = 1, 6 \) correspond to the orbital-sublattice subspace, and the superscript \(+(-)\) refers to particle (hole) energies above (below) the Fermi energy. The amplitudes \( \varphi_{k\sigma}^m \) were obtained by projecting the \( k \) states in the three-orbital basis on to the pseudo-orbital basis \( |m, \tau, \uparrow, \downarrow\rangle \) corresponding to the \( J = 1/2 \) and 3/2 sector states, as given below:

\[
\varphi_{k\uparrow}^1 = \frac{1}{\sqrt{3}} \left( \phi_{k\uparrow}^x + 2 \phi_{k\uparrow}^y \right)
\]

\[
\varphi_{k\uparrow}^2 = \frac{1}{\sqrt{6}} \left( \phi_{k\uparrow}^x - \phi_{k\uparrow}^y - 2 \phi_{k\uparrow}^z \right)
\]

\[
\varphi_{k\uparrow}^3 = \frac{1}{\sqrt{2}} \left( \phi_{k\uparrow}^x + \phi_{k\uparrow}^y \right)
\]

in terms of the amplitudes \( \phi_{k\sigma}^m \) in the three-orbital basis \((\mu = yz, xz, xy)\).

The rotationally invariant Hubbard- and Hund’s-coupling-like terms having the form \( S_{i,m}^\alpha S_{j,m}^\beta \) are diagonal in spin components \((\alpha = \beta)\). The on-site Coulomb interaction terms are also diagonal in the sublattice basis \((s = s')\). The interaction matrix \([H]\) in equation (31) is therefore obtained as:

\[
[H] = \begin{pmatrix}
U_{\uparrow\uparrow} & \frac{2}{3} J_H & 0 \\
\frac{2}{3} J_H & U_{\uparrow\downarrow} & 0 \\
0 & 0 & U_{\downarrow\downarrow}
\end{pmatrix}
\delta_{\alpha\beta} \delta_{ss'}
\tag{34}
\]

in the pseudo-orbital basis. While the first interaction term above preserves spin rotation symmetry, the second interaction term (corresponding to the \( S_{i,m}^\alpha S_{j,m}^\beta \) terms in equation (25)) breaks rotation symmetry and is responsible for easy \( x-y \) plane anisotropy. The spin wave energies are calculated from the pole condition \( 1 - 2\xi_q(\omega) = 0 \) of equation (31), where \( \xi_q(\omega) \) are the eigenvalues of the \([H] \chi^0(q, \omega)\) matrix. The
The calculated magnon dispersion in the three-orbital model with staggered field in the $x$ direction. The easy $x$–$y$ plane anisotropy arising from Hund’s coupling results in one gapless mode and one gapped mode corresponding to transverse fluctuations in the $y$ and $z$ directions, respectively.

The electron fillings in the different pseudo orbitals are obtained as $n_l(1,2,3) \approx (1.064, 1.99, 1.946)$. Finite mixing between the $J = 1/2$ and $3/2$ sectors is reflected in the small deviations from ideal fillings $(1, 2, 2)$ and also in the very small magnetic moment values for $l = 2, 3$ as given above, which play a crucial role in the expression of magnetic anisotropy and magnon gap in view of the anisotropic $J_H$ interaction terms in equation (25). The values $\lambda = 0.38$ eV, $U = 0.93$ eV, and $J_H = 0.1$ eV taken above lie well within the estimated parameter range for Sr$_2$IrO$_4$ [21, 22].

We have investigated the crucial role of the small $J = 3/2$ sector magnetic moment on the magnon gap by studying the variation with SOC strength which effectively controls the mixing between $J = 1/2$ and $3/2$ sectors. Figure 4 shows that the magnon gap sharply increases with the dominant magnetic moment $|m_{z}^{\uparrow}|$, highlighting the finite-SOC effect on the experimentally observed out-of-plane magnon gap in Sr$_2$IrO$_4$. The opposite sign of the magnetic moment $m_{z}^{\downarrow}$ as compared to $m_{z}^{\uparrow}$ (due to spin-orbital entanglement) plays a vital role in the easy-plane anisotropy. It should be noted that the tetragonal splitting $\epsilon_{xy}$ weakly affects the magnon gap through the $J = 3/2$ sector magnetic moments.

The magnon gap in figure 3 is related to the anisotropy term $\Delta_{anis}$ (see discussion of figure 2), and we consider here an analytical expression relating the two which can be derived for a simple model. For the anisotropic Hubbard model [25], using the spin fluctuation analysis for the AFM state on the square lattice [26], we obtain:

$$\frac{\omega_{\text{gap}}}{\omega_{\text{max}}} = \sqrt{\left(\frac{U_{\text{anis}}}{t}\right)\left(\frac{U}{4t}\right)}$$

in the strong coupling limit ($U \gg t$), where $\omega_{\text{max}} = 2(4r^2/U)$, and $U_{\text{anis}} \approx 2\Delta_{anis}$ is the anisotropy term included in the Hubbard model. With $\Delta_{anis}/t = 0.01$ (as in figure 2(b) discussion) and $U/4t = 1$ for simplicity, the ratio is obtained as $\approx 1/7$, which is in good agreement with figure 3. It should be noted that a magnon gap of $\sim 4$ meV (one-tenth of that in figure 3) would correspond to an anisotropy term $\Delta_{anis}/t \sim 10^{-4}$ (one-hundredth of above), which is about 0.03 meV using the hopping energy scale $t_1 = 280$ meV.

7. Comparison with pure orbital based approach

A fully self-consistent approach within the pure $(t_{2g})$ orbital basis, wherein all orbital off-diagonal spin and charge condensates are included (along with the diagonal condensates) in the Coulomb interaction contributions, has been applied recently to the Ca$_2$RuO$_4$ compound with electron filling $n = 4$ to investigate the complex interplay due to intimately intertwined roles of SOC, Coulomb interactions, and structural distortions [23]. We discuss below the relevant results obtained by extending this approach to the Sr$_2$IrO$_4$ compound with $n = 5$.

Most importantly, we have explicitly confirmed the $J_H$ induced easy-plane anisotropy. The anisotropy is completely absent when $J_H = 0$ even for finite $\epsilon_{xy}$, and the fully self-consistent calculation (with octahedral rotation turned off for simplicity) yields degenerate solutions corresponding to arbitrary
reduced $mx$ is directly obtained from the orbital diagonal terms \[24\]. The all off-diagonal condensates in the pseudo orbital basis were finite and yield the Coulomb SOC renormalization, and pseudo-spincanting in the direction, which can be understood in terms of the induced DM interaction resulting from the interplay between octahedral tilting and SOC \[23\].

In terms of symmetry breaking, the above Coulomb-induced anisotropy term $\Gamma_1$ obtained via strong coupling expansion is equivalent to our local anisotropic interaction term (dominant $-2J_H S_z S_z$) derived using the transformation. The major difference is that while in reference \[17\] the $\Gamma_1$ value was treated as a fitting parameter in the 40 meV magnon gap analysis, in our work the explicit form of the anisotropic interaction term $[2J_H(S_z S_z - S_1 S_2)]$ has been derived, and the magnon gap is determined using the same microscopic Hamiltonian parameters which account for the high energy magnon feature (strong zone boundary dispersion) within a unified scheme. Furthermore, all physical terms are treated on the same footing, which is especially important for the weakly correlated 5d systems. \[8\] In general, the nature of magnetic anisotropy depends, through the sign and magnitude of the $J = 3/2$ sector moments $S_z$ and $S_z$, on the lattice, band structure, and magnetic ordering, which is of particular interest for the honeycomb lattice compounds Na$_2$IrO$_3$ and $\alpha$-RuCl$_3$.

### 8. Discussion

Finally, a critical comparison with other approaches for studying magnetic anisotropy in Sr$_2$IrO$_4$ is presented below. In reference \[10\], the IrO$_6$ octahedral rotation induced PD ($J,S_z S_z$) and DM $[D \cdot (S_1 \times S_2)]$ terms were considered as the dominant anisotrophic interactions, which were shown to be gauged out by a site-dependent rotation of the spin operators ($S \rightarrow S$), resulting in no true magnetic anisotropy in the absence of Hund’s coupling. After including the $J_H$ induced correction terms, the resulting anisotropic interaction terms were obtained as:

$$\mathcal{H}_{\text{anis}} = -\Gamma_1 S_z S_z \pm \Gamma_2 (S_z S_z - S_1 S_2)$$

in terms of the rotated spin operator $\tilde{S}$, where the $\pm$ sign corresponds to bond along the $x(y)$ direction. Presence of tetragonal distortion was found to affect the coefficient $\Gamma_1$.

Together with the Heisenberg AFM interaction, the first term with $\Gamma_1 > 0$ leads to easy $a - b$ plane magnetic ordering and magnon gap for out-of-plane fluctuations, whereas the much smaller easy-axis anisotropy and magnon gap for in-plane fluctuations were ascribed to the second term ($\Gamma_2 \ll \Gamma_1$). In reference \[17\], the large (40 meV) magnon gap measured for out-of-plane fluctuations was explained in terms of the above $J_H$ induced anisotropy term $\Gamma_1$, although $J_H$ was not explicitly mentioned in the very brief discussion, as the major focus was on resolving the small ($\sim$3 meV) magnon gap corresponding to the easy-axis anisotropy and basal-plane fluctuations via high-resolution RIXS and inelastic neutron scattering (INS). The proposed mechanism for the easy-axis anisotropy involve interlayer coupling and orthorhombic distortion which are beyond the scope of this work.

For the easy-plane anisotropy, our conclusions are in agreement with the above analysis, including no true anisotropy in the absence of $J_H$, and the tetragonal distortion term only affecting the magnitude and not being the source of anisotropy. In terms of symmetry breaking, the above $J_H$ induced easy-plane anisotropy term $\Gamma_1$ obtained via strong coupling expansion is equivalent to our local anisotropic interaction term (dominantly $-2J_H S_z S_z$) derived using the transformation. The major difference is that while in reference \[17\] the $\Gamma_1$ value was treated as a fitting parameter in the 40 meV magnon gap analysis, in our work the explicit form of the anisotropic interaction term $[2J_H(S_z S_z - S_1 S_2)]$ has been derived, and the magnon gap is determined using the same microscopic Hamiltonian parameters which account for the high energy magnon feature (strong zone boundary dispersion) within a unified scheme. Furthermore, all physical terms are treated on the same footing, which is especially important for the weakly correlated 5d systems. \[8\] In general, the nature of magnetic anisotropy depends, through the sign and magnitude of the $J = 3/2$ sector moments $S_z$ and $S_z$, on the lattice, band structure, and magnetic ordering, which is of particular interest for the honeycomb lattice compounds Na$_2$IrO$_3$ and $\alpha$-RuCl$_3$.

### 9. Conclusion

While all Coulomb interaction terms are invariant under same pure-spin rotation for all three pure ($t_{2g}$) orbitals, the Hund’s coupling and pair hopping interaction terms were shown to explicitly break pseudo-spin rotation symmetry systematically due to the spin-orbital entanglement. Transformation of the various Coulomb interaction terms to the pseudo-spin-orbital basis formed by the $J = 1/2$ and $3/2$ states therefore provides a physically transparent approach for investigating

### Table 1. Self consistently determined magnetization and density values for the three pure ($\mu = yz, xz, xy$) and three pseudo ($l = 1, 2, 3$) orbitals on the two sublattices ($s = A/B$), showing the simplified structure in the pseudo orbital basis.

| $\mu$ (s) | $m_\mu^s$ | $m_\mu^a$ | $m_\mu^l$ | $m_\mu^n$ | $l$ (s) | $m_l^s$ | $m_l^a$ | $m_l^n$ | $n_l$ |
|-----------|-----------|-----------|-----------|-----------|---------|-----------|-----------|-----------|-------|
| $yz$ (A/B) | ±0.24 | 0.0 | 0.0 | 1.62 | 1 (A/B) | ±0.69 | 0.0 | 0.0 | 1.04 |
| $xz$ (A/B) | ±0.24 | 0.0 | 0.0 | 1.62 | 2 (A/B) | ±0.006 | 0.0 | 0.0 | 1.99 |
| $xy$ (A/B) | ±0.18 | 0.0 | 0.0 | 1.75 | 3 (A/B) | ±0.023 | 0.0 | 0.0 | 1.97 |
magnetic ordering and anisotropy effects in the perovskite
(Sr$_2$IrO$_4$) and other $d^5$ pseudo-spin compounds. Explicitly pseudo-spin symmetry-breaking terms were obtained (dominantly $-2J_0S_i^zS_j^z$), resulting in easy $x$--$y$ plane anisotropy and magnon gap for the out-of-plane mode, highlighting the importance of mixing with the nominally non-magnetic $J = 3/2$ sector in determining the magnetic properties of Sr$_2$IrO$_4$.

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