Spin-orbit coupling induced staggered-to-entangled orbital order transition in a three-orbital model for Sr$_2$CrO$_4$

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With octahedrally coordinated $t_{2g}$ orbitals which are active at filling $n = 2$, the Sr$_2$CrO$_4$ compound exhibits rich interplay of spin-orbital physics with tetragonal distortion induced crystal field tuning by external agent such as pressure. Considering both reversed and restored crystal field regimes, collective spin-orbital excitations are investigated in the antiferromagnetic (AFM) state of a realistic three-orbital model using the generalized self consistent + fluctuation approach including spin-orbit coupling (SOC). Important effects of SOC and Coulomb interaction induced orbital mixing terms are highlighted, including staggered-to-entangled orbital order transition. Behavior of the calculated energy scales of collective excitations with crystal field is in striking similarity to that of the transition temperatures with pressure as obtained from susceptibility and resistivity anomalies in high-pressure studies.
I. INTRODUCTION

Among layered perovskite structured 3d transition metal compounds such as Sr$_2$MO$_4$, with $n = 1, 2, 3$ electrons in the $t_{2g}$ sector for M=V,Cr,Mn, the Sr$_2$CrO$_4$ compound presents a promising case for investigating effects of spin-orbit coupling (SOC) and Coulomb interaction induced orbital mixing terms in view of the rich spin-orbital physics exhibited by this compound, as discussed below. Among other members of the Ruddlesden-Popper (RP) series Sr$_{n+1}$Cr$_n$O$_{3n+1}$, the cubic perovskite SrCrO$_3$ ($n = \infty$) is known to exhibit weak antiferromagnetic (AFM) order accompanied with small tetragonal distortion and staggered $yz/xz$ orbital order due to the $(xy)^1(yz,xz)^1$ electronic configuration. The AFM state in the bilayer compound Sr$_3$Cr$_2$O$_7$ ($n = 2$) is also of a similar nature. With $n = 2$ also in the cubic perovskite LaVO$_3$ exhibiting active orbital degree of freedom, inter-site spin-orbital entanglement was found to play an important role in magnetic and orbital ordering temperature, optical spectra, and magnon spectra anomalies.

It is only recently that pure samples of $\alpha$-Sr$_2$CrO$_4$ have been synthesized successfully in bulk, and uncovering the interesting magnetic and orbital physics has attracted considerable interest. Magnetic susceptibility and specific heat measurements, as well as neutron scattering and muon spin rotation studies, have indicated phase transitions at temperatures $\sim$ 110 K and 140 K. A high-pressure study of Sr$_2$CrO$_4$ has been carried out recently using magnetic susceptibility and electrical resistivity measurements up to 14 GPa pressure. The evolution of the 110 K and 140 K transitions was investigated with pressure in the $P \lesssim 3$ GPa regime where the transition temperatures were identified based on $\chi - T$ anomalies. Similar successive transitions in temperature based on $\rho - T$ anomalies were also identified in the $P \gtrsim 6$ GPa regime where the crystal field is restored.

With a K$_2$NiF$_4$-type crystal structure, the CrO$_6$ octahedra in $\alpha$-Sr$_2$CrO$_4$ are elongated along $c$ axis. The octahedral crystal field partially lifts the five-fold degeneracy of $3d$ levels, resulting in $e_g$ and $t_{2g}$ sets. Due to octahedral elongation, the low-lying $t_{2g}$ set is conventionally expected to further split into low-lying degenerate $yz, xz$ orbitals and higher-energy $xy$ orbital. However, the $xy$ orbital was found to be low-lying instead in the more recent DFT study. This counter-intuitive reversal of the usual crystal field effect was attributed to the negative charge-transfer gap associated with Cr-$3d$ and O-$2p$ orbitals. Because of the competing ionic and covalent bonding interactions, the $t_{2g}$ set was projected...
to become degenerate only for highly elongated CrO$_6$ octahedra, whereas for the realistic elongation in $\alpha$-Sr$_2$CrO$_4$, the $xy$ orbital was found to be relatively lower in energy.

Theoretically, the spin-orbital physics in Sr$_2$CrO$_4$ is yet to be fully understood as only few studies have been carried out. These include: density functional theory (DFT) based calculations,\textsuperscript{13-16} tight-binding model fitting to DFT band structure and real-space Hartree-Fock (HF) approximation, supplemented by density-matrix renormalization group (DMRG) study.\textsuperscript{16} The Cr$^{4+}$ ion has 3$d^2$ configuration, and with one electron in the low-lying $xy$ orbital, the system is rendered orbitally active by the remaining one electron for the degenerate $yz, xz$ orbitals. Staggered ($\pi, \pi$) magnetic and orbital order was obtained in the HF study for large on-site Coulomb interaction. However, the study was carried out for small system size and the full range of crystal field splitting was not considered. The low-energy collective spin-orbital excitations remain largely unexplored both experimentally as well as theoretically. Understanding the observed behavior of the transition temperatures in high pressure studies of Sr$_2$CrO$_4$, where the conventional crystal field is restored, also remains elusive.

Several important physical effects have not been considered in earlier theoretical works. These include: SOC induced magnetic anisotropy, Coulomb interaction induced orbital mixing terms, orbital moments, and Coulomb SOC renormalization. Remarkably, the orbital mixing terms are generated spontaneously from the Coulomb interaction terms, even in the absence of usual sources (SOC, octahedral tilting/rotation). In view of the crystal field tuning by pressure, and the peculiar insulator-insulator transition in the crossover region between the reversed and restored crystal field regimes,\textsuperscript{11,15} investigation of the orbital mixing effect is of crucial importance to understand how the insulating gap is protected even as the $xy$ level crosses the $yz, xz$ levels.

Due to the rich interplay expected between SOC, crystal field splitting, Coulomb interaction, and spin-orbital ordering, investigation of collective spin-orbital excitations is of particular interest. This requires a generalized approach in which all physical elements are treated on the same footing. For this purpose, we will employ the generalized self consistent and fluctuation approach for a unified investigation of magnetic and orbital ordering, SOC induced magnetic anisotropy effects, as well as of the various collective excitation modes (magnon, orbiton, spin-orbiton) over the full crystal field regime of interest for the Sr$_2$CrO$_4$ compound. This approach was recently used to investigate spin-orbital fluctuations in several 4$d$ and 5$d$ compounds (NaOsO$_3$, Ca$_2$RuO$_4$, Sr$_2$IrO$_4$) with $n = 3, 4, 5$ $t_{2g}$ electrons.\textsuperscript{17,18}
The structure of this paper is as below. The three-orbital model including hopping, crystal field, Coulomb interaction, and SOC terms is introduced in Sec. II, and the generalized self-consistent + fluctuation approach is reviewed in Sec. III. Results of the self-consistent calculation are presented in Sections IV and V for the staggered and entangled orbital orders, including crystal field driven magnetic reorientation transition, SOC induced anisotropic magnetic interactions, electronic band structure, and spectral functions for the collective excitations (magnon, orbiton, and spin-orbiton). Important features of our calculated results, including comparison of the behavior of excitation energy scales with crystal field and that of the measured transition temperatures with pressure, are discussed in Sec. VI. Finally, conclusions are presented in Sec. VII.

II. THREE-ORBITAL MODEL WITH SOC AND COULOMB INTERACTIONS

In the three-orbital ($\mu = yz, xz, xy$), two-spin ($\sigma = \uparrow, \downarrow$) basis defined with respect to a common spin-orbital coordinate axes along the planar Cr-O-Cr directions, we consider the Hamiltonian $H = H_{\text{band}} + H_{\text{cf}} + H_{\text{int}} + H_{\text{SOC}}$ within the $t_{2g}$ manifold. The band and crystal field terms have been discussed earlier, and are briefly summarized below.

The first, second, and third neighbor hopping terms for the $xy$ orbital are represented by $t_1, t_2, t_3$, respectively. For the $yz$ ($xz$) orbital, $t_4$ and $t_5$ are the nearest-neighbor (NN) hopping terms in $y$ ($x$) and $x$ ($y$) directions, respectively, corresponding to $\pi$ and $\delta$ orbital overlaps. The $xy$ orbital energy offset $\epsilon_{xy}$ (relative to the degenerate $yz/xz$ orbitals) represents the effective crystal field splitting, including the octahedral distortion (elongation/flattening) effect. We have taken hopping parameter values: $(t_1, t_2, t_3, t_4, t_5) = (-1.0, 0.3, 0, -1.0, 0.2)$, and considered the range $-1 \lesssim \epsilon_{xy} \lesssim +1$, all in units of the realistic hopping energy scale $|t_1| = 250 \text{ meV}$. As there is no experimental evidence for octahedral rotation/tilting in $\text{Sr}_2\text{CrO}_4$, the orbital mixing hopping terms $t_{m1, m2, m3}$ have been set to zero.

For the on-site Coulomb interaction terms in the $t_{2g}$ basis ($\mu, \nu = yz, xz, xy$), we consider:

$$
H_{\text{int}} = U \sum_{i,\mu} n_{i\mu\uparrow} n_{i\mu\downarrow} + U' \sum_{i,\mu<\nu,\sigma} n_{i\mu\sigma} n_{i\nu\bar{\sigma}} + (U' - J_H) \sum_{i,\mu<\nu,\sigma} n_{i\mu\sigma} n_{i\nu\bar{\sigma}}
$$

$$
+ J_H \sum_{i,\mu<\nu} a_{i\mu\uparrow} a_{i\nu\downarrow} a_{i\mu\downarrow} a_{i\nu\uparrow} + J_P \sum_{i,\mu<\nu} a_{i\mu\uparrow} a_{i\mu\downarrow} a_{i\nu\downarrow} a_{i\nu\uparrow}
$$

$$
= U \sum_{i,\mu} n_{i\mu\uparrow} n_{i\mu\downarrow} + U' \sum_{i,\mu<\nu} n_{i\mu\uparrow} n_{i\nu\downarrow} - 2J_H \sum_{i,\mu<\nu} S_{i\mu} S_{i\nu} + J_P \sum_{i,\mu<\nu} a_{i\mu\uparrow} a_{i\mu\downarrow} a_{i\nu\downarrow} a_{i\nu\uparrow} \quad (1)
$$
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 ($J_P = J_H$), with $U'' \equiv U' - J_H/2 = U - 5J_H/2$ from the spherical symmetry condition $U' = U - 2J_H$. Here $a_{i\mu\sigma}^\dagger$ and $a_{i\mu\sigma}$ are the electron creation and annihilation operators for site $i$, orbital $\mu$, spin $\sigma = \uparrow, \downarrow$. The density operator $n_{i\mu\sigma} = a_{i\mu\sigma}^\dagger a_{i\mu\sigma}$, total density operator $n_{i\mu} = n_{i\mu\uparrow} + n_{i\mu\downarrow} = \psi_{i\mu}^\dagger \psi_{i\mu}$, and spin density operator $S_{i\mu} = \psi_{i\mu}^\dagger \sigma_{i\mu} \psi_{i\mu}$ in terms of the electron field operator $\psi_{i\mu}^\dagger = (a_{i\mu\uparrow}^\dagger a_{i\mu\downarrow})$. All interaction terms above are SU(2) invariant and thus possess spin rotation symmetry.

Finally, we consider the spin-space representation:

$$H_{SOC}(i) = -\lambda \mathbf{L} \cdot \mathbf{S} = -\lambda (L_z S_z + L_x S_x + L_y S_y)$$

$$= \left[ \left( \psi_{yz\uparrow}^\dagger \psi_{yz\downarrow}^\dagger \right) \left( i\sigma_z \lambda/2 \right) \left( \psi_{xz\uparrow} \psi_{xz\downarrow} \right) + \left( \psi_{xz\uparrow}^\dagger \psi_{xz\downarrow}^\dagger \right) \left( i\sigma_z \lambda/2 \right) \left( \psi_{xy\uparrow} \psi_{xy\downarrow} \right) \right] + \text{H.c.}$$

for the bare spin-orbit coupling term, which explicitly breaks SU(2) spin rotation symmetry and therefore generates anisotropic magnetic interactions from its interplay with other Hamiltonian terms. In the following, we will consider $U$ in the range 8-12 (2-3 eV), $J_H/U$ between 1/10 to 1/6, and bare SOC values between 0.1 to 0.2, all in the energy scale unit $|t_1|=250$ meV, which yields the realistic range $\lambda = 25$ to 50 meV for 3$d$ elements.

### III. GENERALIZED SELF CONSISTENT + FLUCTUATION APPROACH

The generalized self consistent approach including all orbital diagonal and off-diagonal spin and charge condensates has been applied recently to the NaOsO$_3$, Ca$_2$RuO$_4$, and Sr$_2$IrO$_4$ compounds, illustrating the rich interplay between different physical elements. The coupling of orbital moments to weak orbital fields and the interaction-induced SOC renormalization highlight the role of orbital off-diagonal condensates on the emergent orbital and spin-orbital physics.

Resulting from orbital off-diagonal (OOD) spin and charge condensates, the additional contributions of the Coulomb interaction terms (Eq. 1) included in the generalized self consistent approach are:

$$[H_{HF}^{\text{int}}]_{\text{OOD}} = \sum_{i,\mu<\nu} \psi_{i\mu}^\dagger \left( -\sigma \cdot \Delta_{i\mu\nu} + \mathcal{E}_{i\mu\nu} 1 \right) \psi_{i\nu} + \text{H.c.}$$

(3)
where the orbital off-diagonal spin and charge fields are self-consistently determined from:

$$\Delta_{i\mu\nu} = \left( \frac{U''}{2} + \frac{J}{4} \right) \langle \sigma_{i\mu\nu} \rangle + \left( \frac{J_P}{2} \right) \langle \sigma_{i\mu\nu} \rangle$$

$$\mathcal{E}_{i\mu\nu} = \left( -\frac{U''}{2} + \frac{3J}{4} \right) \langle n_{i\mu\nu} \rangle + \left( \frac{J_P}{2} \right) \langle n_{i\mu\nu} \rangle$$

in terms of the corresponding condensates $\langle \sigma_{i\mu\nu} \rangle \equiv \langle \psi_{i\mu}^\dagger \sigma \psi_{i\nu} \rangle$ and $\langle n_{i\mu\nu} \rangle \equiv \langle \psi_{i\mu}^\dagger 1 \psi_{i\nu} \rangle$. The orbital mixing terms above explicitly preserve spin rotation symmetry, and are generally finite due to orbital mixing induced by SOC or octahedral tilting/rotation. In the following, we will see that these terms can also be generated spontaneously.

The orbital moments and Coulomb interaction induced SOC renormalization are calculated from the orbital off-diagonal charge and spin condensates:

$$\langle L_\alpha \rangle = -i \left[ \langle \psi_{\mu}^\dagger \psi_{\nu} \rangle - \langle \psi_{\mu}^\dagger \psi_{\nu} \rangle^* \right] = 2 \text{Im} \langle \psi_{\mu}^\dagger \psi_{\nu} \rangle$$

$$\lambda^\text{int}_\alpha = (U'' - J_H/2) \text{Im} \langle \psi_{\mu}^\dagger \sigma_\alpha \psi_{\nu} \rangle$$

where the orbital pair $(\mu, \nu)$ corresponds to the component $\alpha = x, y, z$. The last equation yields the Coulomb renormalized SOC values $\lambda_\alpha = \lambda + \lambda^\text{int}_\alpha$ where $\lambda$ is the bare SOC value.

Since all generalized spin $\langle \psi_{\mu}^\dagger \sigma \psi_{\nu} \rangle$ and charge $\langle \psi_{\mu}^\dagger \psi_{\nu} \rangle$ condensates are included in the generalized self consistent approach, the fluctuation propagator must also be defined in terms of the generalized operators. We therefore consider the time-ordered generalized fluctuation propagator:

$$[\chi(q, \omega)] = \int dt \sum_i e^{i\omega(t-t')} e^{-i q \cdot (r_i - r_j)} \times \langle \Psi_0 | T[\sigma^\alpha_{\mu\nu}(i, t) \sigma^\alpha_{\mu'\nu'}(j, t')] | \Psi_0 \rangle$$

in the self-consistent AFM ground state $|\Psi_0 \rangle$. The generalized spin-charge operators at lattice sites $i, j$ are defined as $\sigma^\alpha_{\mu\nu} = \psi_{\mu}^\dagger \sigma^\alpha \psi_{\nu}$, which include both orbital diagonal $(\mu = \nu)$ and off-diagonal $(\mu \neq \nu)$ cases, as well as the spin $(\alpha = x, y, z)$ and charge $(\alpha = c)$ operators, with $\sigma^\alpha$ defined as Pauli matrices for $\alpha = x, y, z$ and unit matrix for $\alpha = c$.

The generalized fluctuation propagator in the random phase approximation (RPA) was investigated recently for several $4d$ and $5d$ compounds with electron fillings $n = 3, 4, 5$ in the $t_{2g}$ sector. Due to the active $yz/yz$ orbital degree of freedom in the Sr$_2$CrO$_4$ compound in the $\epsilon_{xy} \sim -1$ regime (where $n_{yz} + n_{xz} = 1$), investigation of collective excitations in this $n = 2$ case is of special importance. Since the generalized spin and charge operators $\psi_{\mu}^\dagger \sigma^\alpha \psi_{\nu}$ include spin $(\mu = \nu, \alpha = x, y, z)$, orbital $(\mu \neq \nu, \alpha = c)$, and spin-orbital $(\mu \neq \nu, \alpha = x, y, z)$
cases, the spectral function of the fluctuation propagator:

$$A_q(\omega) = \frac{1}{\pi} \text{Im} \text{Tr}[\chi(q,\omega)]_{\text{RPA}}$$  \hspace{1cm} (7)$$

provides information about the collective excitations (magnon, orbiton, and spin-orbiton), where the character is determined from the basis resolved contributions in the composite $\mu\nu\alpha$ basis. Orbiton and spin-orbiton modes correspond to same-spin and spin-flip particle-hole excitations, respectively, involving different orbitals.

In the following, we will show that there exists a critical SOC value $\lambda^*$ (weakly $U$ and $J_H$ dependent) which determines the nature of the orbital ordering in the $\epsilon_{xy} \sim -1$ regime where $n_{yz} + n_{xz} \sim 1$. The AFM ordering is accompanied with either staggered orbital order (for $\lambda < \lambda^*$) or with entangled orbital order (for $\lambda > \lambda^*$). Results of the self consistent calculation for these two cases are discussed in the next two sections. The calculated $\lambda^*$ values (between 0.1 (25 meV) and 0.2 (50 meV)) lie within the realistic SOC range for 3d elements.

### IV. RESULTS - I ($\lambda < \lambda^*$)

#### A. Magnetic reorientation transition

Results of the generalized self consistent calculation are shown in Fig. [1]. Throughout this section we will consider parameter values: $U = 8$, $J_H = U/6$, and $\lambda = 0.1$, unless otherwise mentioned. Besides the spin magnetic moments in the AFM state, the orbital magnetic moments and Coulomb renormalized SOC values are also shown for different $\epsilon_{xy}$ values. The staggered magnetization values shown in Fig. [1(b)] refer to orbital-summed $z$ and $x - y$ plane components:

$$m_z = \sum_{\mu} m_{\mu} = \sum_{\mu} (n_{\mu}^\uparrow - n_{\mu}^\downarrow)_A = \sum_{\mu} (n_{\mu}^\downarrow - n_{\mu}^\uparrow)_B$$

$$m_{x-y} = \sum_{\mu} [(m_{\mu}^x)^2 + (m_{\mu}^y)^2]^{1/2}_{A/B}$$  \hspace{1cm} (8)$$

in the AFM state. The planar orbital moment and renormalized SOC components $\langle L_{x-y} \rangle$ and $\lambda_{x-y}$ were also evaluated as above for $m_{x-y}$ (without the orbital sum). The magnetic and orbital moment are seen to undergo a reorientation transition. The composite spin-orbital structure for the two limiting cases $\epsilon_{xy} \sim \pm 1$ is illustrated in Fig. [2] and the salient features are discussed below.
FIG. 1: (a) The total and differential electron occupancies in the \( yz/xz \) sector, showing strong proclivity for staggered \((\pi, \pi)\) structure when \( n_{yz} + n_{xz} \sim 1 \). Axial and planar magnetic moments (b) and orbital moments (c), showing the magnetic reorientation transition induced by the crystal field term \( \epsilon_{xy} \). The ordering direction switches from \( z \) (for \( \epsilon_{xy} \sim -1 \)) to \( x - y \) plane (for \( \epsilon_{xy} \sim +1 \)). (d) The Coulomb renormalized SOC values show similar behavior.

B. Staggered orbital + axial (z) AFM order

For \( \epsilon_{xy} \sim -1 \) (reversed crystal field), the \( xy \) band is pulled down, resulting in nominally \( (xy)^1(yz,xz)^1 \) electron occupancy. Due to maximal instability when \( n_{yz} + n_{xz} \sim 1 \), the \( yz,xz \) sector develops staggered \((\pi, \pi)\) orbital ordering, which is stabilized by the density interaction term \( U'' \). Equivalence between attributing this structure either to density interaction or to Jahn-Teller effect has been reviewed earlier in the context of manganites with two \( e_g \) orbitals and one electron per site.\(^{21}\)

The AFM order is stabilized by AFM interactions between \( xy \) moments resulting from the \( xy \) orbital electron hopping; the \( yz,xz \) moments order accordingly due to Hund’s coupling. The AFM order is frustrated by the preferred FM order of \( yz,xz \) moments.\(^{22}\) The energy gain due to virtual electron hopping \([\sim t_4^2/(U' - J_H)]\) is greater for parallel neighboring spins since the doubly occupied intermediate state has lower energy due to Hund’s coupling. This
frustration (relative energy gain for parallel vs. antiparallel spins \( \sim t_4^2 J_H/U^2 \)) is important when \( U \) is not large and results in low magnon energy. Due to SOC induced magnetic anisotropy, the magnetic and orbital moments are oriented along the \( z \) direction.

### C. Planar \((x-y)\) AFM order

For \( \epsilon_{xy} \sim +1 \) (conventional crystal field), the \( xy \) band is pushed up, aided by the Coulomb renormalization of crystal field splitting, resulting in nominally \( (xy)^0(yz,xz)^2 \) electron occupancy. The AFM order is dominantly stabilized by AFM interaction between \( xz \) (\( yz \)) moments in \( x \) (\( y \)) direction only. Due to the SOC induced easy-plane magnetic anisotropy, the \( yz,xz \) magnetic moments as well as the orbital moments order in the \( x - y \) plane. Both orders are oriented along azimuthal angle \( \phi = \pi/4 \) within the \( x - y \) plane.

The obtained ordering direction of magnetic and orbital moments reflects the magnetic anisotropy effect resulting from an interplay between SOC induced anisotropic interactions, orbital electron occupancy, and Heisenberg interactions. This interplay is discussed below.
D. SOC induced anisotropic interactions

The SOC induced magnetic anisotropy provides insight into how the onset of staggered $yz/xz$ ordering (for $\epsilon_{xy} \sim -1$) is responsible for reorienting the AFM order along the $z$ direction from the easy-plane orientation when both $yz,xz$ orbitals are occupied (for $\epsilon_{xy} \sim +1$). In analogy with the strong-coupling analysis for the spin-dependent hopping terms $i\sigma\cdot t_{ij}^{\alpha}$, strong-coupling expansion for the bare SOC terms $-\lambda \sum_{\alpha=x,y,z} L_{\alpha} S_{\alpha}$ to second order in $\lambda$ yields the anisotropic diagonal (AD) intra-site interactions (for site $i$):

$$[H_{\text{eff}}^{(2)}]_{\text{AD}}(i) = \frac{4(\lambda/2)^2}{U} \left[ S_{yz}^z S_{xz}^z - (S_{yz}^x S_{xz}^x + S_{yz}^y S_{xz}^y) \right] + \frac{4(\lambda/2)^2}{U} \left[ S_{xz}^x S_{xy}^x - (S_{xz}^y S_{xy}^y + S_{xz}^z S_{xy}^z) \right] + \frac{4(\lambda/2)^2}{U} \left[ S_{xy}^y S_{yz}^y - (S_{xy}^x S_{yz}^x + S_{xy}^z S_{yz}^z) \right]$$

(9)

between the $yz,xz,xy$ moments. Here all three orbitals have been assumed to be nominally half-filled in this general analysis.

Now, for $\epsilon_{xy} \sim +1$, when only $yz,xz$ moments are present, only the first term in Eq. (9) is operative, which directly yields preferential $x-y$ plane ordering (easy-plane anisotropy) for parallel $yz,xz$ moments (enforced by Hund’s coupling). On the other hand, for $\epsilon_{xy} \sim -1$, when only $xy$ and $yz$ moments are present on A sublattice (say) and only $xy$ and $xz$ moments on B sublattice, only the third and second terms are operative, respectively. There is no frustration if the moments are oriented along $z$ direction, whereas if moments are oriented along $x$ direction on A sublattice and along $y$ direction on B sublattice, the AFM order is frustrated. The resulting easy ($z$) axis anisotropy thus involves a crucial interplay between the SOC induced anisotropic interactions, orbital occupancy, Hund’s coupling, and the AFM Heisenberg interactions.

For the planar ordering case (for $\epsilon_{xy} \sim +1$), the continuous symmetry for in-plane spin rotation is further reduced to $C_4$ symmetry due to a weak easy-axis anisotropy (along the $45^\circ$ directions) resulting from the weak anisotropic interactions (second and third terms in Eq. 9) due to the small $xy$ moment in this regime. The above qualitative analysis for the SOC induced magnetic anisotropy and ordering directions in the two opposite $\epsilon_{xy}$ regimes is confirmed by the generalized self consistent calculations described earlier.

For the easy-plane anisotropy case ($\epsilon_{xy} \sim +1$), Fig. 3 shows the evolution of the axial ($m_z$) and planar ($m_{x-y}$) sublattice magnetization components with iterations in the self
FIG. 3: Variation of the axial ($m_z$) and planar ($m_{x-y}$) sublattice magnetization components (identical for both $yz, xz$ orbitals) with iterations in the (i) standard (dashed line) and (ii) generalized (solid line) self consistent calculations. Starting with nearly-$z$ initial direction, the AFM ordering direction approaches the $x - y$ plane. Here bare SOC value $\lambda = 0.1, U = 8, \epsilon_{xy} = +1$.

consistent calculation, starting with nearly-$z$ initial ordering direction. The extremely low magnetic anisotropy energy scale $\lambda^2/U$ is reflected in the extremely slow approach of the AFM ordering direction to the $x - y$ plane. The approach is relatively much faster in the generalized (compared to the standard) self consistent calculation due to additional (but indirect) SOC-induced contribution to magnetic anisotropy from the Coulomb orbital mixing terms, showing effectively enhanced SOC. Other spin-rotationally-symmetric terms such as the crystal field term $\epsilon_{xy} n_{xy}$ also yield indirect SOC-induced contribution to magnetic anisotropy, as seen from the significant magnon gap dependence on $\epsilon_{xy}$ for the NaOsO$_3$ compound.$^{18}$

E. Electronic band structure

For the two ordering direction cases, the orbital resolved electronic band structure calculated in the self-consistent AFM state is shown in Fig. 4. In both cases, the large gaps across the Fermi energy $E_F$ are due to Hubbard $U$ between opposite-spin bands for the three orbitals. The small gaps between same-spin bands are due to: (a) staggered $yz/xz$ ordering and (b) crystal field term $\epsilon_{xy}$. 
Role of orbital mixing terms on electronic band structure

We now highlight the important role of Coulomb interaction induced orbital mixing terms (Eqs. [31]). When OOD condensates and SOC are neglected (standard HF approach), bands for the three orbitals are completely decoupled due to absence of orbital mixing. For \( \epsilon_{xy} = 0.5 \), the \( xy \) band lies just above the \( yz, xz \) bands as seen in Fig. 4(a). However, when OOD condensates are included (generalized HF approach), the band structure [Fig. 5(b)] shows a significant insulating gap induced by the Coulomb orbital mixing terms. Also, part of the \( xy \) band is now seen to lie below the \( yz, xz \) bands.

FIG. 5: Orbital resolved electronic band structure in the self-consistent state: (a) without and (b) with the orbital off-diagonal condensates included. SOC is set to zero in (a) and weak SOC has negligible effect in (b). Here \( \epsilon_{xy} = 0.5 \).
FIG. 6: Low-energy part of the spectral function in the axial ordering case for several $\epsilon_{xy}$ values, showing magnon (below 20 meV) and two orbiton modes. While energy of the lower orbiton mode (involving $yz, xz$ orbitals) remains unchanged, energy of the upper orbiton mode (involving $xy$ and $yz/xz$ orbitals) decreases with decreasing crystal field splitting. Here $J_H = U/5.5$.

With $\epsilon_{xy}$ decreasing further, $xy$ orbital weight is progressively transferred below the insulating gap, until the entire $xy$ band is shifted below the Fermi energy [Fig. 4(a)], while the insulating gap remains finite. This $xy$ orbital spectral weight transfer across the insulating gap with varying $\epsilon_{xy}$ is similar to that obtained for the Ca$_2$RuO$_4$ compound.$^{17}$

It is important to note that the orbital mixing induced insulating gap and the electronic band structure [Fig. 5(b)] are nearly unchanged in the weak SOC regime. This is because the OOD condensates are generated spontaneously from the Coulomb interaction terms. In the absence of SOC, these condensates are purely real, and acquire imaginary part (rotation in the complex plane) when SOC is introduced, resulting in finite orbital moments and SOC renormalization (Eq. 5). Here, the role of SOC is crucial, because finite circulating orbital and spin-orbital currents are generated due to spin-orbital entanglement, resulting in finite orbital and spin-orbital moments.

F. Collective excitations

In the axial ordering case, low-energy part of the spectral function calculated for several $\epsilon_{xy}$ values shows (Fig. 6) magnon modes (below 20 meV) and two orbiton modes. As expected, energy of the lower orbiton mode (not involving $xy$ orbital) remains constant, while that of the upper orbiton mode (involving $xy$ hole) decreases from 160 meV to 40 meV with decreasing crystal field splitting. Slightly higher $J_H$ value was taken here in order to separate the lower orbiton and magnon modes for clarity, which are otherwise nearly
FIG. 7: The spectral function calculated in the planar ordering case, showing magnon and orbiton modes for several $\epsilon_{xy}$ values. While energy of the orbiton mode (involving $xy$ orbital) increases with crystal field, the magnon mode energy increases only slightly. (b) shows magnon-orbiton entanglement when energy of the two modes are comparable.

overlap in energy for $J_H = U/6$.

In addition, there are three spin-orbiton modes ($\sim 400, 500, 625$ meV for $\epsilon_{xy} = -0.5$) and high-energy magnon mode ($\sim 650$ meV) which reflects the cost of out-of-phase spin fluctuations of different orbitals due to Hund’s coupling. The small magnon gap (8 meV) reflects the SOC induced easy ($z$) axis anisotropy and finite energy cost for transverse ($x, y$) spin fluctuations. The low magnon mode energy is due to frustration and competing (FM) order preferred by $yz, xz$ moments. Consequently, magnon energy increases with decreasing $J_H$, for which the orbiton mode energy decreases due to enhanced interaction term $U'' - J_H/2$, which lowers the orbiton mode energy in the usual resonant scattering mechanism.

In the planar ordering case, the calculated spectral function is shown in Fig. 7 for several $\epsilon_{xy}$ values. Again, the most significant feature is that while energy of the orbiton mode increases with $\epsilon_{xy}$ due to increasing particle-hole excitation energy, the magnon mode energy increases only slightly. The orbiton modes arise from particle-hole excitations involving $xy$ (particle) and $yz, xz$ (hole) orbitals, and therefore correspondng to $L_x, L_y$ fluctuations. The prominent gapped magnon mode (b) shows a gap of 25 meV at $q = (\pi, \pi)$ for out-of-plane spin fluctuations. The gap for the in-plane magnon mode (b) is about 3 meV. When the $xy$ band is pushed up by increasing $\epsilon_{xy}$, the orbiton modes shift to higher energy (c) and disentangle from the magnon modes, which are now reduced to conventional magnons in two-dimensional quantum Heisenberg AFM model with effective spin $S = 1$ constituted by Hund’s coupled $yz, xz$ moments.
V. RESULTS - II ($\lambda > \lambda^*$)

A. Entangled orbital order

In the reversed crystal field regime ($\epsilon_{xy} \sim -1$), where the $yz/xz$ sector orbital degree of freedom is maximal since $n_{yz} + n_{xz} \sim 1$, we obtain a transition from staggered to entangled orbital order at a critical SOC value ($\lambda^*$) which is $U$ and $J_H$ dependent. For $\lambda > \lambda^*$, the self-consistent calculation shows that the staggered orbital order (S OO) decreases to zero with iterations, and the self-consistent state corresponds to the entangled orbital order (EOO) with $n_{yz} = n_{xz}$, as illustrated in Fig. 8(a). The optimal entanglement in the $yz/xz$ sector is reflected by the saturated orbital moment $\langle L_z \rangle = 1$ for $\lambda > \lambda^*$, as shown in Fig. 8(b).

To understand the nature of the EOO, it is convenient to consider the entangled eigenstates of the SOC + crystal field Hamiltonian:

$$
\psi_1 = \frac{1}{\sqrt{2+\alpha^2}}[\sigma_x \psi_{yz} + \sigma_y \psi_{xz} + \alpha \sigma_z \psi_{xy}]
$$

$$
\psi_2 = \frac{1}{\sqrt{2+\beta^2}}[\sigma_x \psi_{yz} + \sigma_y \psi_{xz} - \beta \sigma_z \psi_{xy}]
$$

$$
\psi_3 = \frac{1}{\sqrt{2}}[\sigma_x \psi_{yz} - \sigma_y \psi_{xz}]
$$

(10)

where $\psi_{\mu} = (a_{\mu\uparrow}, a_{\mu\downarrow})^T$ are the electron field operators and the coefficients $\alpha, \beta$ are given by:

$$
\alpha = \frac{-1 + \zeta + \sqrt{9 + \zeta^2 - 2\zeta}}{2}
$$

$$
\beta = \frac{1 - \zeta + \sqrt{9 + \zeta^2 - 2\zeta}}{2}
$$

(11)

in terms of the parameter $\zeta = 2\epsilon_{xy}/\lambda$. In the limit $\zeta \to -\infty$, where $\alpha \approx 0$ and $\beta \approx |\zeta|$, the $\zeta$-dependent eigenstates reduce to:

$$
\psi_1 \approx \frac{1}{\sqrt{2}}[\sigma_x \psi_{yz} + \sigma_y \psi_{xz}]
$$

$$
\psi_2 \approx -\sigma_z \psi_{xy}.
$$

(12)

Thus, for $-\epsilon_{xy} \gg \lambda$, the eigenstates $\psi_1$ and $\psi_2$ have dominantly $yz/xz$ and $xy$ orbital characters, respectively, as shown in Fig. 8(c). Similarly, in the limit $\zeta \to +\infty$ ($\epsilon_{xy} \gg \lambda$), the results for $\psi_1, \psi_2$ are simply interchanged (except for $\psi_2$ sign). Also, $\psi_2$ and $\psi_3$ are the two lowest-energy eigenstates over the entire $\epsilon_{xy}$ range.

Including the AFM order with EOO corresponds to the occupation of $\psi_2$ and $\psi_3$ states with pseudo-spin $\tau = \uparrow$ on A and $\tau = \downarrow$ on B sublattices. The opposite pseudo-spin cases
FIG. 8: (a) Schematic representation of the distinct self-consistent states obtained at $\epsilon_{xy} = -0.75$ for different bare SOC values. The various transitions obtained are: disappearance of staggered orbital order (IV to I and III to II) when SOC is increased beyond critical value $\lambda^*$, magnetic reorientation due to SOC induced anisotropy (IV to III and I to II) when a small perturbation $\delta m_z$ (AFM) is included, and reorientation + disappearance of SOO (IV to II) when both $\delta m_z$ and increased SOC are included. (b) The saturation of orbital moment $\langle L_z \rangle = 1$ for $\lambda > \lambda^*$ reflects the optimal entanglement of $yz, xz$ orbitals in the entangled orbital order. (c) The orbital shapes of the SOC eigenstates. (d) Variation of the critical SOC value ($\lambda^*$) with $U$ showing the phase boundary between SOO and EOO orders for $\epsilon_{xy} = -0.75$ and $J_H = U/6$.

for $\psi_2$ and $\psi_3$ along with remaining $\psi_1$ doublet with $\tau = \uparrow, \downarrow$ are the four unoccupied states above the Fermi energy. This picture is confirmed by the electronic band structure (Fig. 10) which shows the six emergent bands.

Fig. 8(d) shows the variation of the critical SOC value with $U$ for fixed Hund’s coupling ($J_H = U/6$). The critical SOC value weakly decreases with increasing $U$ (which suppresses band effects) and with decreasing $J_H$ (not shown), because both band effects (hopping terms...
FIG. 9: (a) The total and differential electron occupancies in the $yz/xz$ sector show the entangled orbital order ($n_{yz} - n_{xz} = 0$ even when $n_{yz} + n_{xz} = 1$). (b) Axial and planar magnetic moments and (c) orbital moments, showing the magnetic reorientation transition. The orbital moments (c) and Coulomb renormalized SOC values (d) are stronger in comparison to the SOO case.

mix different $J$ sectors) and Hund’s coupling (which tends to align moments of different orbitals) are detrimental to the SOC induced entanglement. The critical SOC value remains unaffected by the crystal field value.

In the following, we will consider the parameter set $U = 12$ (3 eV), $J_H = U/6$, bare SOC value $\lambda = 0.2$ (above critical SOC), and $\epsilon_{xy} = -0.75$ unless otherwise indicated, with the hopping parameters and energy scale ($|t_1| = 250$ meV) same as earlier. Results of the generalized self consistent calculation are shown in Fig. 9 for the crystal field range $-1 \leq \epsilon_{xy} \leq +1$. As expected, the orbital moments and renormalized SOC values are stronger compared to the SOO case (Fig. 1) due to enhanced SOC induced orbital entanglement.

The calculated electronic band structure and magnetic excitation spectral function in the AFM state with EOO are shown in Fig. 10. The band structure is much simpler in this case with only six doubly degenerate bands and no fine splitting between $yz/xz$ bands as in the SOO case (Fig. 4(a)). The three doubly degenerate bands formed near the Fermi energy are derived from the three entangled states discussed above. Similarly, three bands for opposite spin are formed at much higher energy due to the Hubbard interaction $U$. The insulating gap in this case is produced by the Coulomb orbital mixing terms, while it was
FIG. 10: (a) The orbital resolved electronic band structure and (b) low energy part of spectral function calculated in the AFM state with EOO.

the staggered orbital field which was responsible in the SOO case.

The lowest energy collective excitation (∼40 meV) in Fig. 10(b) corresponds to the $yz/xz$ orbiton mode involving particle-hole excitation between bands originating from the entangled $\psi_1$ and $\psi_3$ states discussed above. The second mode (∼90 meV) with dominantly $xy$ character is the magnon mode corresponding to transverse spin fluctuations away from the easy $z$ axis. The large magnon gap (∼60 meV) reflects the strong SOC-induced magnetic anisotropy. The third pair (∼200 meV) are orbiton modes corresponding to p-h excitations involving $yz/xz$ (particle) and $xy$ (hole) bands originating from the $\psi_1$ and $\psi_2$ states.

When SOC strength approaches the critical value, the $yz/xz$ orbiton mode becomes gapless due to the degeneracy between staggered and entangled orbital orders, as shown in Fig. 11(a). Fluctuations which change the orbital order from entangled to staggered, or

FIG. 11: (a) The spectral function calculated in the AFM state with EOO shows the gapless lower orbiton mode at the critical SOC value $\lambda^* = 0.13$, reflecting the degeneracy with SOO case. (b) Variation of both orbiton and magnon gap energies at $\mathbf{q} = 0$ with bare SOC strength.
vice versa, cost no energy for $q = 0$. For $\lambda > \lambda^*$, the enhanced energy lowering from the Coulomb orbital mixing terms shifts the balance in favor of EOO, whereas the staggered orbital field provides the dominant energy lowering for $\lambda < \lambda^*$. This accounts for the orbiton gap increasing from zero on both sides of $\lambda^*$, as seen in Fig. 11(b) which shows the variation of the orbiton and magnon gaps with bare SOC value. As expected, the calculated magnon gap properly goes to zero with SOC.

The Hund’s coupling only weakly affects the collective excitation energy scales. Fig. 12 shows the variation of the calculated excitation energies at $q = (\pi/2, \pi/2)$ with $\epsilon_{xy}$ in the AFM state with EOO for three $J_H$ values. In all cases, the lower orbiton energy remains flat over the entire $\epsilon_{xy}$ range and the magnon energy is flat over a broad range ($\epsilon_{xy} \leq -0.5$). Energy of the upper orbiton (involving $xy$ hole) increases linearly with $|\epsilon_{xy}|$ for higher $J_H$ values.

Finally, we consider the extremely weak SOC regime ($\lambda \sim 0.1$) in order to highlight the strong contrast with the no SOC case. Fig. 13 shows the spectral functions for the (a)

![FIG. 12: Variation of the different collective excitation energies at $(\pi/2, \pi/2)$ with crystal field in the AFM state with EOO for several $J_H$ values.

![FIG. 13: The spectral function of collective excitations calculated for the SOO case with SOC values: (a) $\lambda = 0.01$ and (c) $\lambda = 0.1$. (b) Variation of magnon and orbiton mode energy scales with bare SOC strength. Here $\epsilon_{xy} = -0.75$.](image)
FIG. 14: The spectral functions calculated for the planar AFM order, showing magnon and orbiton modes for several $\epsilon_{xy}$ values: (a) 0.5, (b) 0.75, and (c) 1.0.

essentially no SOC and (c) extremely weak SOC cases. The energy of the magnon mode is lowest in (a) but increases significantly above the orbiton mode in (c). The variation of magnon and orbiton mode energy scales ($\omega_q$ for $q = (\pi/2, \pi/2)$) with SOC strength is shown in (b). Even in the SOO case, there is strong enhancement of the magnon energy scale with SOC, which is driven by the increasing magnon gap (Fig. 11). This shows that the extremely weak SOC case is remarkably different from the no SOC case.

**B. Planar AFM order**

Turning now to collective excitations in the planar AFM ordering case ($\epsilon_{xy} > 0$), the calculated spectral functions are shown in Fig. 14 for several $\epsilon_{xy}$ values. The lower energy modes are the magnon modes involving the magnetically active $yz, xz$ orbitals, showing nearly gapless and strongly gapped modes corresponding to in-plane and out-of-plane spin fluctuations due to the easy-plane anisotropy. The higher energy modes (energy increasing with $\epsilon_{xy}$) are the orbiton modes involving $xy$ (particle) and $yz/xz$ (hole) excitations.

We now discuss the estimation of Néel temperature $T_N$ from the magnon energy scale within the picture of thermal excitation of magnons as driving the demagnetization. From Fig. 14(c), the magnon energy scale $\omega_{\text{max}} \sim 70 \text{ meV}$ for $\epsilon_{xy} = +1$ corresponding to the high pressure side. Next, taking advantage of the well-studied finite-temperature spin dynamics including interlayer hopping and magnetic anisotropy effects, qualitatively similar magnon dispersion in the cuprate compound La$_2$CuO$_4$ with $T_N \sim 400 \text{ K}$ and $\omega_{\text{max}} \sim 300 \text{ meV}$ and neglecting differences in interlayer hopping and magnetic anisotropy gaps which only weakly (logarithmically) affect the spin dynamics, we obtain $T_N \sim 90 \text{ K}$ which is
close to the transition temperature $T_N \sim 120$ K for the chromate compound as seen on the high-pressure side of the $P - T$ diagram.\[11\]

VI. DISCUSSION

Orbital moments $\langle L_\alpha \rangle$ and spin-orbital moments $\langle L_\alpha S_\beta \rangle$ (where $\alpha, \beta = x, y, z$) are generated by circulating orbital and spin-orbital currents which are finite only in presence of SOC induced spin-orbital entanglement. Extremely weak SOC in strongly correlated 3$d$ transition metal systems is usually neglected in theoretical studies, and therefore consideration of orbital and spin-orbital moments is naturally excluded. Also generally neglected are the Coulomb orbital mixing terms as in Eqs. 3, 4 which are generated in the generalized HF approach. Furthermore, in studies where the staggered $yz/xz$ ordering is considered in terms of the Jahn-Teller effect only, the Coulomb renormalization of crystal field splitting is not considered due to neglect of the inter-orbital density interaction term $U''$.

The most dramatic effect of SOC seen in our calculation for the Sr$_2$CrO$_4$ compound is the staggered-to-entangled orbital order transition, where the $yz/xz$ orbiton mode becomes gapless. The extremely weak critical SOC value ($\lambda^* = 0.13$ for $U = 12$) lies in the realistic range for 3$d$ elements. Other important consequences of SOC include: strong orbital and spin-orbital moments, magnetic anisotropy induced reorientation transition, finite magnon gap, and strong magnon-orbiton entanglement. Besides enhanced magnetic anisotropy, orbital mixing effects (due to both SOC and Coulomb interaction terms) result in insulating gap in the electronic band structure and transfer of $xy$ orbital spectral weight across the insulating gap with changing $\epsilon_{xy}$.

Our study of collective excitations provides insight into the observed behavior of the two transition temperatures with pressure which tunes the crystal field. In the high-pressure study,\[11\] while the temperature of the 140 K transition (at ambient pressure) was found to decrease linearly with pressure, that of the 110 K transition remains nearly unchanged. Both transitions (obtained from $\chi - T$ anomalies) disappear near 3 GPa where their temperatures meet. A similar but inverted behavior is obtained in the higher pressure regime, where the two transition temperatures (obtained from $\rho - T$ anomalies) approach each other with decreasing pressure and meet near 8 GPa. In addition, a peculiar insulator-to-insulator transition was obtained at around 5 GPa between the two pressure regimes. Disappearance
FIG. 15: Variation of the two excitation energy scales with $\epsilon_{xy}$ in the two regions I and II. The in-between shaded region is where the $xy$ orbital electron spectral weight transfer across the insulating gap is active. Here bare SOC value $\lambda = 0.2$, $U = 12$, and $J_H = U/6$. These features closely track the observed behaviour of the two transition temperatures and the insulator-insulator transition found in the high-pressure study.\textsuperscript{11}

of the 140 K transition was attributed to restoration of the conventional crystal field in a consequent DFT study.\textsuperscript{15}

Towards a qualitative comparison with results of the high-pressure study, Fig. 15 shows the behavior of the relevant excitation energy scales — lower energy orbiton mode ($\omega_1$) involving $yz/xz$ orbitals in region I, and magnon mode ($\omega_2$) and higher energy orbiton mode ($\omega_3$) in both regions I and II. The variation of the calculated energy scales with $\epsilon_{xy}$ in the two regions is in striking similarity to the behavior of the two transition temperatures with pressure in the $P \lesssim 3$ GPa and $P \gtrsim 6$ GPa regimes, respectively. The above picture is consistent with the thermal excitation of low-energy collective modes driving the phase transitions. Also shown is the variation of $xy$ orbital electron density $n_{xy}$. The shaded $\epsilon_{xy}$ region, where active spectral weight transfer (see. Fig. 14) causes significant change in the orbital character of states across the insulating gap, corresponds to the insulator-insulator transition found in the high pressure study.

In the previous section, the Néel temperature was estimated from the magnon energy scale in the positive $\epsilon_{xy}$ regime. We now consider finite-temperature effects on the EOO in the negative $\epsilon_{xy}$ regime (assuming that $\lambda > \lambda^*$), where the lowest-energy mode is the $yz/xz$
orbiton mode (Fig. 15) involving excitations between the entangled \(|yz\) \pm i|xz\rangle\) states across the Fermi energy. Resulting from the thermal excitation of this orbiton mode, the lowest temperature effects will therefore be associated with orbital dynamics and reduction of orbital (\(\langle L_z \rangle\)) and spin-orbital (\(\langle L_z S_z \rangle\)) moments, leading to loss of entangled orbital order and disentanglement transition. Due to effectively reduced spin-orbital correlations, the dominantly \(xy\)-like magnon mode energy will be significantly reduced, as in Fig. 13 for decreasing spin-orbit coupling, resulting in strong thermal demagnetisation and concomitant magnetic disordering (Néel) transition. The disentanglement and magnetic disordering transitions approximately coincide in this proposed picture.

The \(xy - yz/xz\) orbiton mode energy (\(\sim 150\) meV at \(\epsilon_{xy} = -0.5\)) will also get downward renormalised above the Néel temperature where only short-range AFM order survives. The neighbouring spin twisting away from AFM order will enhance the hopping effect and thus slightly reduce the local \(xy\) moment and the polarising field term \(-\sigma.\Delta_{xy}\). The \(xy\) (hole) band will therefore go up, leading to lowered orbiton energy. Further studies are clearly required to explore this complex regime involving rich finite-temperature induced coupled spin-orbital physics.

VII. CONCLUSIONS

Including SOC and Coulomb interaction induced orbital mixing terms in the generalized self consistent approach results in rich spin-orbital physics in Sr\(_2\)CrO\(_4\). Due to the active \(yz/xz\) orbital degree of freedom in the reversed crystal field (\(\epsilon_{xy} \sim -1\)) regime, even extremely small SOC has important consequences such as strong orbital and spin-orbital moments, magnetic reorientation transition and magnon gap due to magnetic anisotropy, and staggered-to-entangled orbital order transition. The behavior of the low-energy magnon and orbiton excitation energy scales with the crystal field term \(\epsilon_{xy}\) is in striking similarity to the observed behavior of transition temperatures with pressure found in the high-pressure study, supporting the picture of thermal excitation of these collective modes driving the finite-temperature spin and orbital dynamics.
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Z. H. Zhu, W. Hu, C. A. Occhialini, J. Li, J. Pelliciari, C. S. Nelson, M. R. Norman, Q. Si, and R. Comin, *Néel and Stripe Ordering from Spin-Orbital Entanglement in $\alpha$-Sr$_2$CrO$_4$, arXiv:1906.04194v2* (2021).

T. Yamauchi, T. Shimazu, D. N.-Hamane, and H. Sakurai, *Contrasting Pressure-Induced Metalization Processes in Layered-Perovskites, $\alpha$-Sr$_2$MO$_4$ (M=V, Cr)*, Phys. Rev. Lett. 123, 156601 (2019).

T. Baikie, Z. Ahmad, M. Srinivasan, A. Maignan, S. S. Praman, and T. J. White, *The Crystallographic and Magnetic Characteristics of Sr$_2$CrO$_4$ (K$_2$NiF$_4$-type) and Sr$_{10}$(CrO$_4$)$_6$F$_2$ (apatite-type)*, J. Solid State Chem. 180, 1538 (2007).

H. Weng, Y. Kawazoe, X. Wan, and J. Dong, *Electronic Structure and Optical Properties of Layered Perovskites Sr$_2$MO$_4$ (M=Ti, V, Cr, and Mn): An ab initio Study*, Phys. Rev. B 74, 205112 (2006).

T. Ishikawa, T. Toriyama, T. Konishi, H. Sakurai, and Y. Ohta, *Reversed Crystal-Field Splitting and Spin-Orbital Ordering in $\alpha$-Sr$_2$CrO$_4$*, J. Phys. Soc. Jpn. 86, 033701 (2017).

R. Takahashi, T. Yamaguchi, K. Sugimoto, T. Yamauchi, H. Sakurai, and Y. Ohta, *Pressure-Induced Restoration of the Reversed Crystal-Field Splitting in $\alpha$-Sr$_2$CrO$_4$*, JPS Conf. Proc. 30, 011026 (2020).

B. Pandey, Y. Zhang, N. Kaushal, R. Soni, L.-F. Lin, W.-J. Hu, G. Alvarez, and E. Dagotto, *Origin of the Magnetic and Orbital Ordering in $\alpha$-Sr$_2$CrO$_4$*, Phys. Rev. B 103, 045115 (2021).

S. Mohapatra and A. Singh, *Magnetic Reorientation Transition in a Three Orbital Model for Ca$_2$RuO$_4$ — Interplay of Spin-Orbit Coupling, Tetragonal Distortion, and Coulomb Interactions*, J. Phys.: Condens. Matter 32, 485805 (2020).

S. Mohapatra and A. Singh, *Coupled Spin-Orbital Fluctuations in a Three Orbital Model for 4d and 5d Oxides with Electron Fillings $n=3, 4, 5$ — Application to NaOsO$_3$, Ca$_2$RuO$_4$, and Sr$_2$IrO$_4$*, J. Phys.: Condens. Matter 33, 345803 (2021).

J. Stöhr and H. C. Siegmann, *Magnetism: From Fundamentals to Nanoscale Dynamics*, Springer, 2006.

S. Mohapatra, R. Kundu, A. Dubey, D. Dutta, and A. Singh, *Role of Orbital off-diagonal Spin and Charge Condensates in a Three Orbital Model for Ca$_2$RuO$_4$ — Coulomb Renormalized Spin-Orbit Coupling, Orbital Moment, and Tunable Magnetic Order*, J. Magn. Magn. Mater 537, 168172 (2021).
21. D. K. Singh, B. Kamble, and A. Singh, *Spin-Charge and Spin-Orbital Coupling Effects in Ferromagnetic Manganites*, J. Phys.: Condens. Matter **22**, 396001 (2010).

22. L. M. Roth, *Simple Narrow-Band Model of Ferromagnetism Due to Intra-Atomic Exchange*, Phys. Rev. **149**, 306 (1966).

23. S. Mohapatra and A. Singh, *Spin Waves and Stability of Zigzag Order in the Hubbard Model with Spin-Dependent Hopping Terms: Application to the Honeycomb Lattice Compounds Na$_2$IrO$_3$ and α-RuCl$_3$*, J. Magn. Magn. Mater **479**, 229 (2019).

24. A. Singh, Z. Tešanović, H. Tang, G. Xiao, C. L. Chien, and J. C. Walker, *Magnetic Dynamics in Copper-Oxide Based Antiferromagnets: The Role of Interlayer Coupling*, Phys. Rev. Lett. **64**, 2571 (1990).

25. A. Singh and Z. Tešanović, *Magnetic Dynamics in La$_2$CuO$_4$ with Interlayer Coupling and Anisotropy Gaps*, Phys. Rev. B **43**, 11445-11447 (1991).

26. A. Singh and P. Goswami, *Spin-Wave Spectrum in La$_2$CuO$_4$ — Double Occupancy and Competing Interaction Effects*, Phys. Rev. B **66**, 092402 (2002).