The case for octupolar order in $d$-orbital Mott insulators

A. Paramekanti,$^{1,2}$ D. D. Maharaj,$^3$ and B. D. Gaulin$^{3,4,5}$

$^1$Department of Physics, University of Toronto, 60 St. George Street, Toronto, ON, M5S 1A7 Canada
$^2$International Centre for Theoretical Sciences, Tata Institute of Fundamental Research, Bengaluru 560089, India
$^3$Department of Physics and Astronomy, McMaster University, Hamilton, ON L8S 4M1 Canada
$^4$Brockhouse Institute for Materials Research, McMaster University, Hamilton, ON L8S 4M1 Canada
$^5$Canadian Institute for Advanced Research, 661 University Ave., Toronto, ON M5G 1M1 Canada

(Dated: September 19, 2019)

Motivated by experimental and theoretical interest in realizing multipolar orders in $d$-orbital materials, we discuss the quantum magnetism of $J = 2$ ions on the face-centered cubic lattice which can be realized in spin-orbit coupled oxides with $5d^2$ transition metal ions. Based on the crystal field environment, we argue for a splitting of the $J = 2$ multiplet, leading to a low lying non-Kramers doublet which hosts quadrupolar and octupolar moments. We discuss a microscopic mechanism whereby the combined perturbative effects of orbital repulsion and antiferromagnetic Heisenberg spin interactions leads to ferro-octupolar coupling between neighboring sites, and stabilizes ferro-octupolar order. This same mechanism is also shown to disfavor quadrupolar ordering. We study spin dynamics in the ferro-octupolar state using a slave-boson approach, uncovering a gapped and dispersive magnetic exciton. For sufficiently strong magnetic exchange, the dispersive exciton can condense, leading to conventional type-I antiferromagnetic order which can preempt octupolar order. Our proposal for ferro-octupolar order, with specific results in the context of a model Hamiltonian, provides a comprehensive understanding of thermodynamics, $\mu$SR, X-ray diffraction, and inelastic neutron scattering measurements on a range of cubic $5d^2$ double perovskite materials including Ba$_2$ZnOsO$_6$, Ba$_2$CaOsO$_6$, and Ba$_2$MgOsO$_6$. Our proposal for exciton condensation leading to type-I magnetic ordering is argued to be relevant to materials such as Sr$_2$MgOsO$_6$.

Background overview: Ordered DP materials, with chemical formula $A_2BB'O_6$, are of great interest in the context of frustrated magnetism since the B and B’ sublattices individually form networks of edge-sharing tetrahedra. When only one of these ions (say B’) is magnetically active, it results in quantum magnetism on the face-centered cubic (FCC) lattice. Our work in this paper is directly motivated by a series of recent experiments on cubic double perovskite (DP) magnets, and we discuss how our results apply to these materials.

Multipolar symmetry-breaking orders have been extensively discussed in $f$-orbital based lanthanide and actinide compounds, which host ions where spin-orbit coupling (SOC) is a dominant energy scale [1]. For instance, the “hidden order” state of URu$_2$Si$_2$ has been proposed to host hexadecapolar symmetry breaking [2]. Another well-known example is cubic NpO$_2$ [3,6], where a large body of experiments have been reconciled in terms of a primary antiferro-triakontadipolar (rank-5 magnetic multipolar) symmetry breaking which drives secondary antiferro-quadrupolar order. In certain pyrochlore magnets, all-in-all-out magnetic order has been proposed as potential candidates to realize this antiferro-quadrupolar order. In certain pyrochlore magnets, antiferro-octupolar order has been proposed [3–6], where $d$-orbital candidates for hosting octupolar orders. Indeed, there appears to be no microscopic understanding of what are the key ingredients to potentially stabilize such octupolar phases. In this paper, we consider spin-orbit coupled Mott insulators having transition metal ions with total angular momentum $J = 2$. We show that such Mott insulators can exhibit competing multipolar orders, and discuss a microscopic mechanism which stabilizes a ferro-octupolar state on the face-centered cubic lattice. Our work in this paper is directly motivated by a series of recent experiments on cubic double perovskite (DP) magnets, and we discuss how our results apply to these materials.
these $t_{2g}$ states. For the most well-studied $d^9$ electronic configuration (e.g., for Ir$^{4+}$ or Ru$^{3+}$ ions), this results in a single hole in a $j_{\text{eff}} = 1/2$ state [18] [19]. Recent experimental and theoretical studies on the DP Ba$_2$CeIrO$_6$, which hosts such a $j_{\text{eff}} = 1/2$ Mott insulator on the FCC lattice, have found evidence of magnetic ordering with a strong frustration parameter, suggesting proximity to a quantum spin liquid state [20] [21]. Skipping to which clearly reveal the dominance of SOC over Hund’s finding magnetically ordered states with large spin gaps that 5\(d\) transition metal oxides bely this expectation, including specific heat, magnetic susceptibility, and include additional octahedral crystal field effects described by the Hamiltonian $H_{\text{CEF}} = -V_{\text{eff}}(O_{40} + 5O_{44})$. Here, the Steven’s operators are given by

\[
O_{40} = 35J_2^2 - (30J(J + 1) - 25)J_2^2 + 3J^2(J + 1)^2 - 6J(J + 1),
\]

\[
O_{44} = \frac{1}{2}(J_+^4 + J_-^4).
\]

The origin of $H_{\text{CEF}}$ might lie in an interaction-induced renormalization of the single-particle crystal field Hamiltonian. For $V_{\text{eff}} > 0$, this results in a non-Kramers ground state doublet, and an excited triplet with a gap $\Delta = 120V_{\text{eff}}$. This single-site model exhibits partial quenching of entropy upon cooling, from ln(5) per spin at high temperature to ln(2) at low temperature. These limiting values are indicated by dashed lines.

Finally, we turn to the topic of our work: $d^2$ ions with an effective $J_{\text{eff}} = 2$ angular momentum state. In this case, previous theoretical work has found intricate multipolar couplings as for $d^1$ filling, and broad swaths of quadrupolar orders in the phase diagram [22] [23] which may coexist with conventional dipolar magnetic order, or valence bond orders [24]. Indeed, recent experiments on $5d^2$ oxides, Ba$_2$NaOsO$_6$ with Os$^{7+}$ [25] [26] and Ba$_2$MgReO$_6$ with Re$^{6+}$ [27], have found clear evidence for multiple transitions associated with these distinct broken symmetries, with a higher temperature quadrupolar ordering transition followed by dipolar ordering at a lower temperature.

From coupling total $L = 1$ and $S = 1$ for two electrons, and including additional octahedral crystal field effects described by the Hamiltonian $H_{\text{CEF}} = -V_{\text{eff}}(O_{40} + 5O_{44})$. Here, the Steven’s operators are given by

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Working in the $|J_\pm = m\rangle$ basis, the ground state wavefunctions are

\[
|\psi_{g,\pm}\rangle = |0\rangle; \quad |\psi_{g,\mp}\rangle = \frac{1}{\sqrt{2}}(|2\rangle + |2\rangle)
\]

while the excited state wavefunctions are given by

\[
|\psi_{e,\pm}\rangle = |\pm 1\rangle; \quad |\psi_{e,0}\rangle = \frac{1}{\sqrt{2}}(|2\rangle - |2\rangle).
\]

The ground state manifold has vanishing matrix elements for the dipole operators $(J^z, J^\pm)$, precluding any dipolar order stemming from the low energy doublet manifold. However, $J$ can induce transitions between the ground doublet and the excited triplet, which will lead to a spin-gap $\Delta$ in the excitation spectrum. As discussed below, incorporating inter-site AF exchange would convert this local mode into a dispersing gapped ‘magnetic exciton’.

The spin gap also leads to a saturation of the low temperature magnetic susceptibility. Fig. 2 plots the inverse susceptibility $\chi^{-1}(T)$ showing its low $T$ saturation.

![Graph showing entropy per spin versus temperature](image)
At high temperature, \( T \gg \Delta \), we recover the Curie law \( \chi \sim J(J + 1)/3T \). However, there is a wide temperature range \( \Delta \leq T \leq 2\Delta \), where \( \chi^{-1}(T) \) can be fit to an apparent Curie-Weiss form over the limited range \( \Delta \lesssim T \lesssim 2\Delta \), with an apparent Curie-Weiss temperature \( \Theta_{\text{CW}}^{\text{app}} \sim 0.1\Delta \).

Examples of type-(I) interactions may be illustrated by considering a pair of neighboring sites in the \( xy \)-plane which will have interactions between the \( e_g \) quadrupolar charge densities \( (J^2_x - J^2_y) \) or \( (3J^2_z - J^2) \) at the two sites. These interactions may be directly projected into the doublet sector as

\[
H^{(1)\text{eff},xy}_{\text{eff},xy} = \sum_{\langle ij \rangle_{xy}} (-\gamma_0 \tau_{ix} \tau_{jx} + \gamma_1 \tau_{iz} \tau_{jz}),
\]

with \( \gamma_0, \gamma_1 > 0 \). (The effective Hamiltonian for nearest neighbors in other planes can be obtained using symmetry transformations.)

Examples of type-(II) interactions for a pair of neighboring spins in the \( xy \)-plane include the conventional AFM exchange \( \gamma_m \vec{J}_i \cdot \vec{J}_j \) with \( \gamma_m > 0 \), where \( \vec{J} \) denotes the \( J = 2 \) spin. In addition, they include \( t_{2g} \) quadrupolar interactions of the form \( \gamma_2 \rho_i \cdot \rho_j \) where \( \rho_i = (\rho_{i,x} \rho_{i,y} + \rho_{i,y} \rho_{i,z})/2 \) and \( \gamma_2 > 0 \). In this case, neither \( \vec{J} \) nor \( \rho_i \) have matrix elements in the low energy doublet space \( |L\rangle \), but they instead mix \( |L\rangle \) into the high energy triplet subspace \( |H\rangle \), with an energy cost \( 2\Delta \) since both sites \( (i, j) \) get excited into the triplet sector. We find that the effective Hamiltonian for such neighboring spins in the \( xy \)-plane is given, in second order perturbation theory, by

\[
H^{(2)\text{eff},xy}_{\text{eff},xy} = -\frac{1}{2\Delta} \sum_{\langle ij \rangle_{xy}} \left( \gamma_m \vec{J}_i \cdot \vec{J}_j + \gamma_2 \rho_{i,xy} \rho_{j,xy} \right)^2
\]

Projecting these operators to the doublet sector, we find

\[
H^{(2)\text{eff},xy}_{\text{eff},xy} = -\frac{1}{2\Delta} \sum_{\langle ij \rangle_{xy}} \left[ 12\gamma_m \gamma_2 \tau_{iy} \tau_{jy} + (6\gamma^2_m + \frac{9}{4}\gamma^2_2) \tau_{iz} \tau_{jz} \\
+ (6\gamma^2_m - 12\gamma_m \gamma_2) \tau_{ix} \tau_{jx} \right].
\]

This equation is one of the key results of our paper. The first term shows that the second order perturbation theory produces a ferro-octupolar coupling with strength \( 6\gamma_4 / \Delta \) from the cross-coupling of \( \gamma_2 \) and \( \gamma_m \). Furthermore, assuming a hierarchy \( \gamma_m \ll \gamma_2 \), we see that the net quadrupolar interaction, after including the terms in Eq. [5] involves direct and perturbative contributions which come with opposite signs,

\[
H_{xy}^{\text{Quad}} = \sum_{\langle ij \rangle_{xy}} \left[ (-\gamma_0 + 6\gamma^2_m / \Delta) \tau_{ix} \tau_{jx} + (\gamma_1 - \frac{9}{8}\gamma_2^2) \tau_{iz} \tau_{jz} \right].
\]

This partial cancellation may cause suppression of quadrupolar order, allowing for the ferro-octupolar coupling to dominate. Our derivation of these results is meant to be illustrative; further work is needed to obtain a complete microscopic theory, starting from an electronic hopping model with interactions, along the lines of calculations presented in Refs. [17] [31] [33] [38].

**Magnetic exciton dispersion:** In order to explore the impact of intersite couplings on the spin excitation...
spectrum in the ferro-octupolar ground state, which can be compared with neutron scattering results, we use a slave boson approach \[39–41\]. We define the ground and excited states of the low energy doublet via

\[
|\psi_{g,\sigma}\rangle = b_0^\dagger |\text{vac}\rangle \tag{9}
\]

\[
|\psi_{e,\alpha}\rangle = d_\alpha^\dagger |\text{vac}\rangle, \tag{10}
\]

where \(\sigma = \uparrow, \downarrow\), \(\alpha = 0, \pm\), and \(|\text{vac}\rangle\) denotes the boson vacuum. This requires a local constraint

\[
\sum_{\sigma = \pm} b_{\sigma}^\dagger b_\sigma + \sum_{\alpha = 0, \pm} d_\alpha^\dagger d_\alpha = 1. \tag{11}
\]

Excitations out of the low energy space contain at least one b-boson; we thus get

\[
J^+ = \sqrt{6}(b_+^\dagger d_- + d_+^\dagger b_\uparrow + \sqrt{2}(b_+^\dagger d_+ + d_+^\dagger b_\downarrow) \tag{12}
\]

\[
J^z = 2(d_0^\dagger b_\uparrow + b_\downarrow d_0) \tag{13}
\]

To model the dispersion of the gapped spin excitations, we consider a nearest-neighbor Heisenberg Hamiltonian \(\gamma_m \sum_{(ij)} \vec{J}_i \cdot \vec{J}_j\). We supplement this, in the ferro-octupolar symmetry broken phase, by a uniform octupolar Weiss field: \(-B_0 \sum_j \tau_{ijy}\). Here, \(\tau_{ijy} \equiv -i(b_\uparrow^\dagger b_\downarrow - b_\downarrow^\dagger b_\uparrow)\), and, without loss of generality, we can set \(B_0 > 0\).

The Weiss field favors a ground state Bose condensate \(b_\uparrow \approx 1/\sqrt{2}\) and \(b_\downarrow \approx i/\sqrt{2}\), resulting in the simplified expressions

\[
J^+ \approx 3(d_+^\dagger + d_-) - i(d_+ - d_-) \tag{14}
\]

\[
J^z \approx i\sqrt{2}(d_0^\dagger d_0) \tag{15}
\]

Using these and accounting for the local doublet-triplet gap, we transform to momentum space, so the full Hamiltonian for describing the magnetic excitons is given by

\[
H_{\text{exc}} = (\Delta + B_0) \sum_{\Omega \alpha} d_{k,\alpha}^\dagger d_{k,\alpha} + \frac{\gamma_m}{2} \sum_k \eta_k J^+_k J^-_k
\]

\[- \gamma_m \sum_{k} \eta_k (d_{k,0}^\dagger d_{-k,0} - d_{k,0} d_{-k,0}^\dagger) \tag{16}
\]

where \(J^+_k \equiv \sqrt{3}(d_{k,0}^\dagger + d_{-k,0}) - i(d_{k,0} - d_{-k,0})\), \(J^-_k \equiv (J^+_k)^\dagger\), and \(\eta_k = \sum_\delta e^{i \delta} \delta\) labelling the 12 nearest-neighbor vectors on the FCC lattice. This leads to a three-fold degenerate magnetic exciton with energy dispersion given by

\[
\lambda(k) = \sqrt{(\Delta + B_0)(\Delta + B_0 + 4\gamma_m \eta_k)} \tag{17}
\]

We find that the exciton energy \(\lambda(k)\) is largest at the \(\Gamma\) point, and is softest at the typical FCC lattice type-I AF ordering wavevector \(\mathbf{K}\).

We expect the exciton dispersion will have temperature dependence through the temperature dependence of the octupolar order parameter, which enters via the Weiss field \(B_0(T)\), softening somewhat as we heat upwards towards the octupolar ordering transition. A plot of the dispersion along a high symmetry path in the FCC Brillouin zone, for a choice of octupolar Weiss field \(B_0 = 0.2\Delta\), and two different choices for the Heisenberg coupling: (i) \(\gamma_m = 0.01\Delta\) (black) and (ii) \(\gamma_m = 0.05\Delta\) (blue). The exciton mode clearly softens with increasing \(\gamma_m\).

**Application to experiments:** The cubic oximates Ba\(_2\)MoO\(_6\) (with \(M = \text{Zn, Mg, Ca}\)) potentially provide a realization of \(J = 2\) ions on the FCC lattice. They all exhibit a single phase transition at \(T^* \sim 30-50\) K, across which the entropy release is only \(\sim \ln(2)\) per Os, suggesting that the full \(\ln(5)\) entropy is partially quenched for \(T \lesssim 100\) K \([42–44]\) without any phase transition. Indeed, the structure appears to be perfectly cubic, in the \(Fm\overline{3}m\) space group, at all temperatures; both neutron diffraction and high resolution XRD measurements find no signs of any non-cubic distortions \([15]\). This suggests that the entropy quenching above the phase transition at \(T^*\) must arise from cubic crystal field effects, as discussed in our theory with a non-Kramers ground state doublet.

Below the phase transition at \(T^*\), neutron diffraction sees no ordered moment, even for \(T \ll T^*\), instead placing tight upper bounds on the ordered dipolar moment, \(\lesssim 0.06-0.13\mu_B\), depending on the material \([45]\). At the same time, \(\mu SR\) measurements have found evidence for zero field oscillations, showing spontaneous breaking of time-reversal symmetry \([42]\). Since neutron diffraction appears to rule out dipolar magnetic order, and the cubic structure appears inconsistent with quadrupolar order, we argue that octupolar ordering within the doublet, which preserves cubic crystal symmetry and breaks time-reversal symmetry, provides the simplest explanation for the data. (Unlike for the \(\Gamma_5\) octupoles in \(\text{NpO}_2\) \([11]\), the octupolar \(\tau_y\) ordering here is not symmetry constrained to induce secondary quadrupolar order.)

Inelastic neutron scattering experiments \([15]\) find a spin gap \(\sim 10-15\) meV, which we interpret as arising from

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**FIG. 3.** Magnetic exciton dispersion \(\lambda(k)\) (in units of \(\Delta\)) along high symmetry path in the FCC lattice Brillouin zone, for a choice of octupolar Weiss field \(B_0 = 0.2\Delta\), and two different choices for the Heisenberg coupling: (i) \(\gamma_m = 0.01\Delta\) (black) and (ii) \(\gamma_m = 0.05\Delta\) (blue). The exciton mode clearly softens with increasing \(\gamma_m\).
although we have explored the detailed consequences for exchange $\gamma$ and site coupling is weaker than the on-site CEF splitting $\Delta$, we must estimate the octupolar coupling constant in Eq. 7. We do not have any microscopic symmetry breaking, we must estimate the octupolar coupling constant $\gamma_m$.

While the measured magnetic susceptibility \cite{12,14} for $T \leq 300$ K in these materials hints at a Curie-Weiss temperature scale $-150$ K, we have shown that the true $\Theta_{CW}$ must be shifted by $0.1\Delta$ due to the local spin gap, so we estimate $\Theta_{CW} \sim -120$ K; dividing this by $z J^2$, with the FCC coordination number $z = 12$ and moment size $J = 2$, we crudely estimate $\gamma_m \sim 0.25$ meV.

Next, in order to explain $T^*$ for the Ising octupolar symmetry breaking, we must estimate the octupolar coupling constant in Eq. 7. We do not have any microscopic estimate for $\gamma_2$. Assuming $\gamma_2 \ll \Delta$, so that this inter-site coupling is weaker than the on-site CEF splitting $\Delta$, if we set $\gamma_2 \sim 5$ meV, we find the Ising ferro-octupolar exchange $6\gamma_m \gamma_2/\Delta \sim 7$ K. Using a classical FCC Ising model to describe the ferrooctupolar order, the known results for the Ising transition temperature \cite{46}, lead us to estimate an ordering temperature $T^* \sim 70$ K, somewhat larger than the experimental result. (We note that although we have explored the detailed consequences for $\gamma_2 > 0$, we are unable to rule out the possibility that $\gamma_2 < 0$ which would favor antiferro-octupolar order. In this case, a larger value of $|\gamma_2| \sim 50$ meV would be necessary to explain the octupolar ordering temperature $T^*$; however, it is not then clear why the $\tau_x \tau_x$ coupling in Eq. 8 would not cause a leading quadrupolar instability.)

Turning to the measured exciton gap from inelastic neutron scattering, if we assume a Weiss field $B_0 \sim 2.5$ meV (which is $\sim T^*/2$), then using the above $\Delta, \gamma_m$, we find $\lambda(\mathbf{K}) \sim 25$ meV, larger than the measured exciton gap at $\mathbf{K}$. Choosing a larger $\gamma_m \sim 1$ meV leads to $\lambda(\mathbf{K}) \sim 14$ meV, in better agreement with the data. These uncertainties in $\gamma_m$ might reflect the possibility that other magnetic exchange terms could be important, beyond a single isotropic Heisenberg coupling. Fig. 4 shows the dynamical spin structure factor

\begin{equation}
S(k, \omega) \propto \sqrt{\frac{B_o + \Delta}{B_o + \Delta + 4\gamma_m \eta_k}} \delta(h\omega - \lambda(k)),
\end{equation}

plotted after powder averaging, and including the Os$^{6+}$ form factor. We find a high intensity gapped band in an energy window $\sim (0.5\Delta, \Delta)$, with the largest intensity concentrated at $k = \pi/a$, which corresponds to type-I ordering wavevector $\mathbf{K} = (\pi/a, 0, 0)$. We have assumed the Os-Os distance to be $a\sqrt{2}$, with $a = 4$ Å as the typical cubic lattice constant for such perovskite crystals.

We finally note that for a smaller gap $\Delta$ and stronger inter-site exchange, the octupolar order can coexist with dipolar order or even be totally preempted by Bose condensation of the magnetic exciton. The resulting conventional type-I AFM state can have a small ordered moment if it is close to the exciton condensation transition. We propose this scenario for Sr$_2$MgOsO$_6$ which appears to have a smaller $\Delta$ (based on its entropy) and a larger $\gamma_m$ (based on its Curie-Weiss temperature), and exhibits a type-I AFM ground state with an ordered moment $\sim 0.6\mu_B$, much smaller than the moment size $\sim 1.88\mu_B$ inferred from high temperature susceptibility measurements \cite{47}. While the tetragonal deformation in Sr$_2$MgOsO$_6$ splits the non-Kramers doublet, and partially the triplet, the magnetic exciton condensation proposed here might still be the dominant instability.

Summary: We have presented arguments in this work in favor of octupolar ordering of $J = 2$ ions on the FCC lattice which is relevant to a family of complex 5d$^2$ oxides, and identified a microscopic mechanism for generating ferro-octupolar coupling. Further theoretical and experimental work, perhaps using magnetostriction as discussed in Ref. 14, is needed to provide smoking gun signatures of the ferro-octupolar symmetry breaking, which may require single crystals of suitable sizes. It may also be useful to carry out more detailed microscopic calculations to compute the sign of the octupolar exchange; as noted above, the possibility of antiferro-octupolar order is not ruled out by our work. Another interesting experimental direction would be to apply pressure on the cubic DFs discussed here in an attempt to induce Bose condensation of the magnetic excitons. Finally, our finding

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig4.pdf}
\caption{Powder averaged dynamic spin structure factor (arbitrary units) including the Os$^{6+}$ form factor, showing the gapped magnetic exciton, as a function of momentum transfer $k$ (in Å$^{-1}$), for a lattice constant $a = 4$ Å and energy $\hbar\omega$ (in units of $\Delta$). We have set the Heisenberg exchange coupling $\gamma_m = 0.05\Delta$ and octupolar Weiss field $B_0 = 0.1\Delta$.}
\end{figure}
of a perturbative microscopic mechanism to inducing octupolar couplings via excited crystal field levels may be of potential importance in pinpointing analogous mechanisms in heavy fermion compounds.

ACKNOWLEDGMENTS

This work was supported by the Natural Sciences and Engineering Research Council of Canada. AP also acknowledges support from a Simons Foundation Targeted Grant to ICTS-TIFR.

* arump@physics.utoronto.ca

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