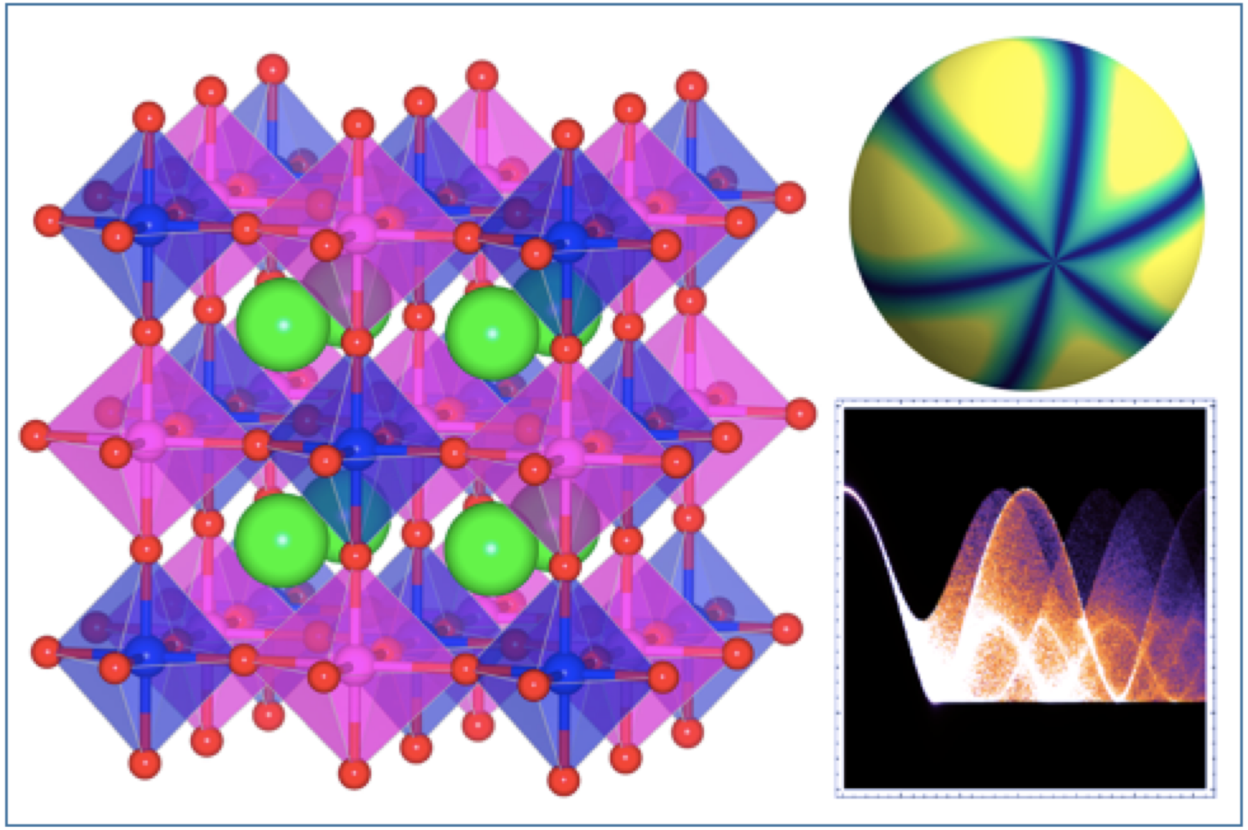
Octupolar order in a d-orbital Mott insulator



Arun Paramekanti
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KITP Multicomponent
7 Oct 2020

UNIVERSITY OF TORONTO

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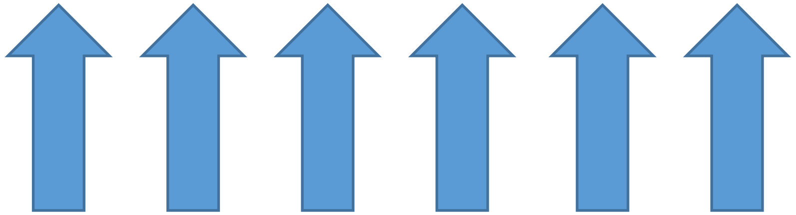
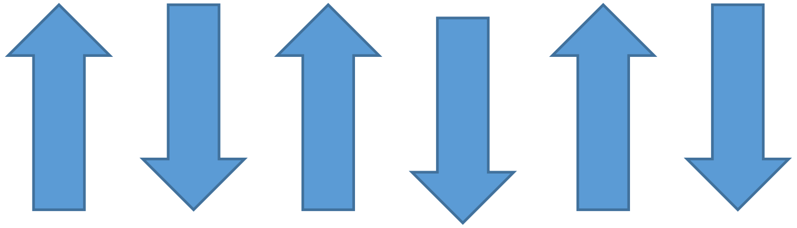
ICTS INTERNATIONAL CENTRE *for* THEORETICAL SCIENCES
TATA INSTITUTE OF FUNDAMENTAL RESEARCH

Multipolar orders

Dipolar order

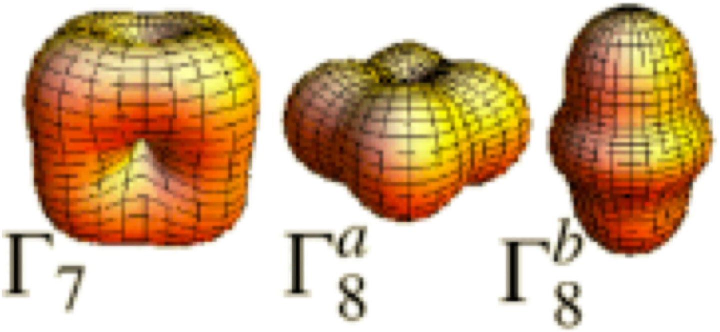


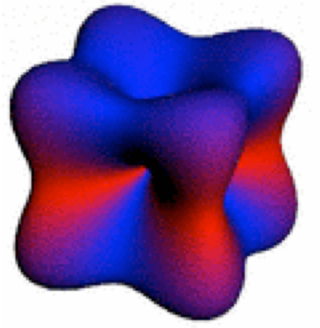
Ferromagnetism
Easily manifest



Antiferromagnetism
Probe: Neutron diffraction
Neutron: dipole interaction

Multipolar orders





SOC drives complex spin and orbital density distributions
"**Hidden order**": URu_2Si_2, NpO_2

Mostly 5f heavy fermions

Energy scales in 5f v/s 5d systems

5f (actinides)

Hubbard U ~ 1-2eV

Spin orbit coupling ~ 1-2eV

Hund's coupling ~ 0.5eV (?)

} Well-separated J levels

Crystal field splittings ~ 10-100meV

[write crystal field potential in terms of J]

5d systems

Hubbard U ~ 1-2eV

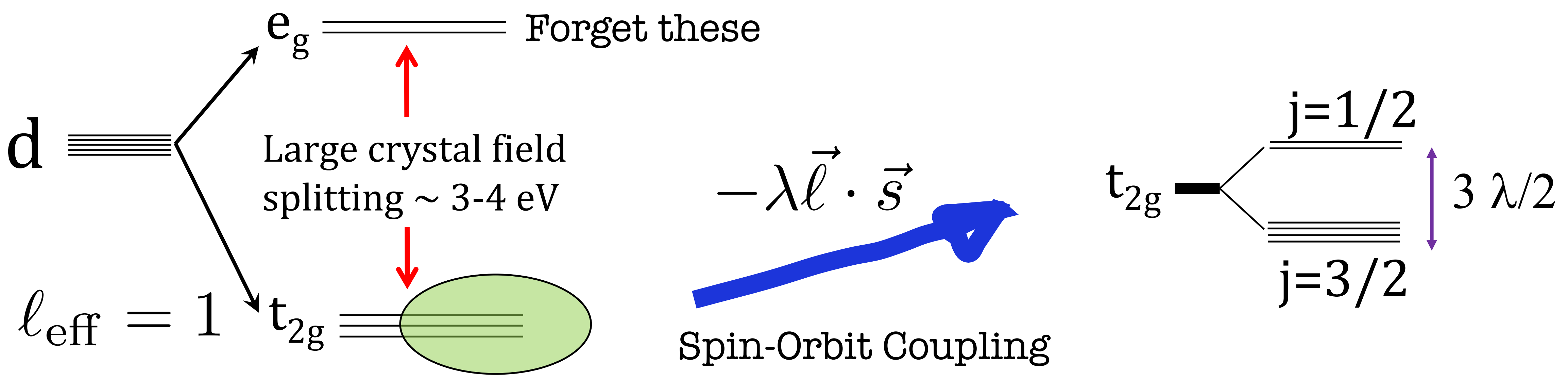
Crystal field splittings ~ 2-3eV

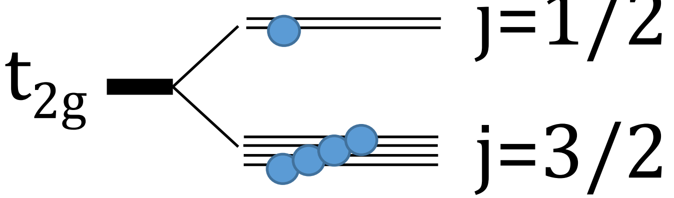
} Well-separated orbitals

[write crystal field potential in terms of L]

SOC~ 0.4eV; Hund's coupling ~ 0.25eV

d-orbitals: “atomic limit”





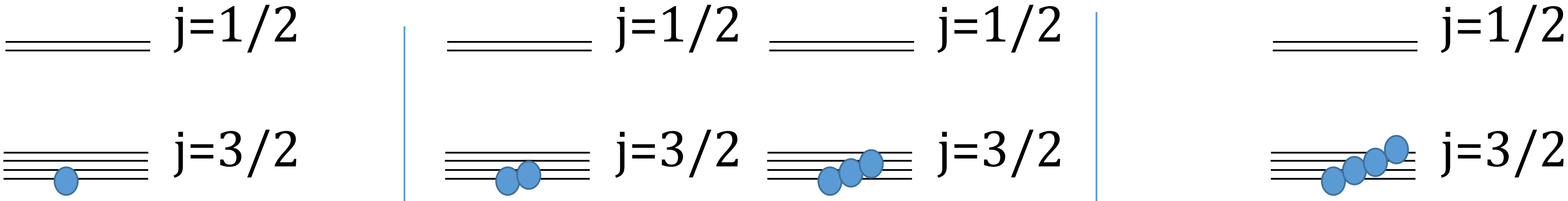
Sr2IrO4, Na2IrO3 , Li_2IrO_3

B. J. Kim, et al, PRL 2008
B. J. Kim, et al, Science 2009
Y. Singh, P. Gegenwart PRB 2010
G. Jackeli and G. Khaliullin, PRL 2009

Most well studied limit

- “Relativistic” Mott insulator
- Superconductivity in doped Sr_2IrO_4?
- Kitaev physics in edge-shared geometries

d-orbitals: "atomic limit"



Ba_2NaOsO_6, Ba_2MgReO_6

Chen, Pereira, Balents, PRB (2010)
C. Svoboda, Trivedi, Randeria (2017)
L. Lu, et al, Nat. Comm. (2017)
D. Hirai, Z. Hiroi, JPSJ (2019)
D. Hirai, et al, arXiv:2004.13928

Recent work

- ❖j=3/2 Mott insulator
- ❖Nnbr orbital repulsion?
- ❖Quadrupolar+Dipolar

This talk

D. D. Maharaj, et al, PRL (2020)
AP, et al, PRB (2020)
S. Voleti, et al, PRB (2020)

Ongoing work

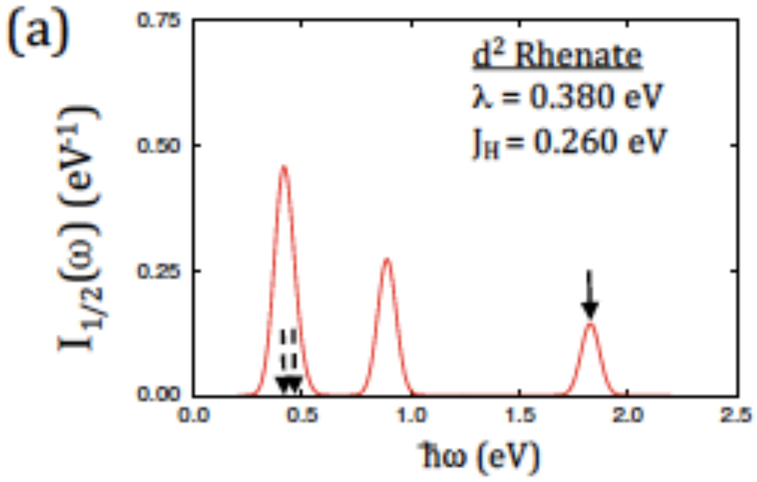
Ba_2YIrO_6, Sr_2YIrO_6

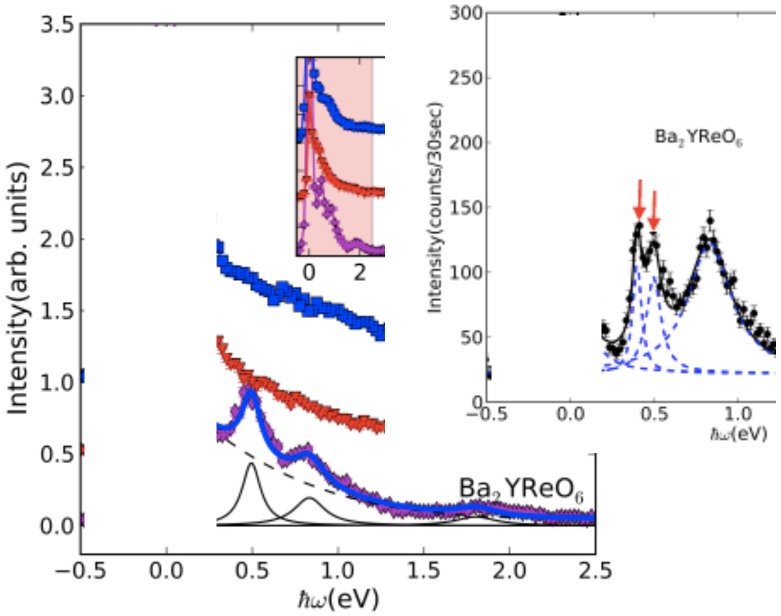
G. Khaliullin, PRL (2013)
G. Cao, et al, PRL (2014)
T. Dey, et al, Buchner, PRB (2016)
Svoboda, Randeria, Trivedi (2017)
A.Paramekanti, et al, PRB (2018)

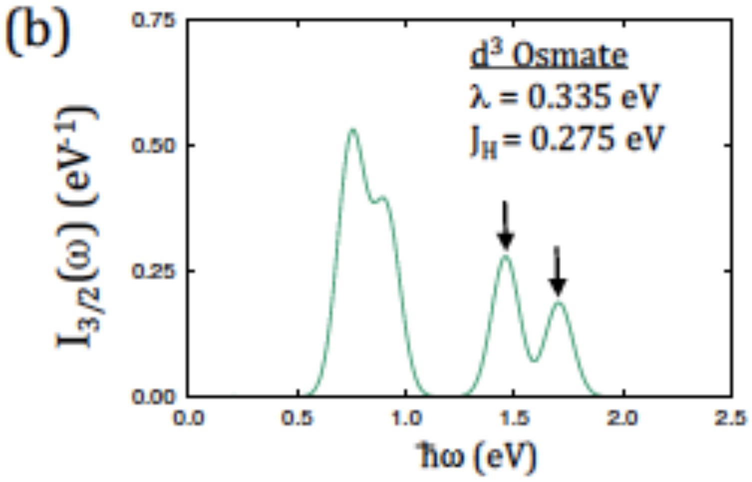
Recent work

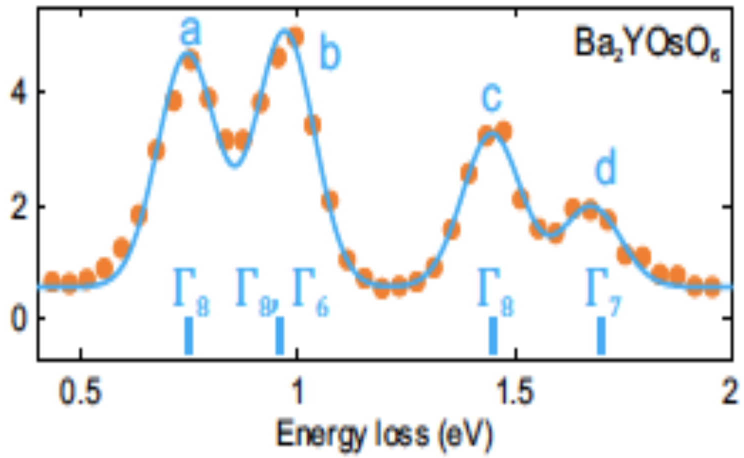
- ❖Fully occupied j=3/2 orbital
- ❖Mott insulator with J_{eff}=0
- ❖Excitonic magnetism?

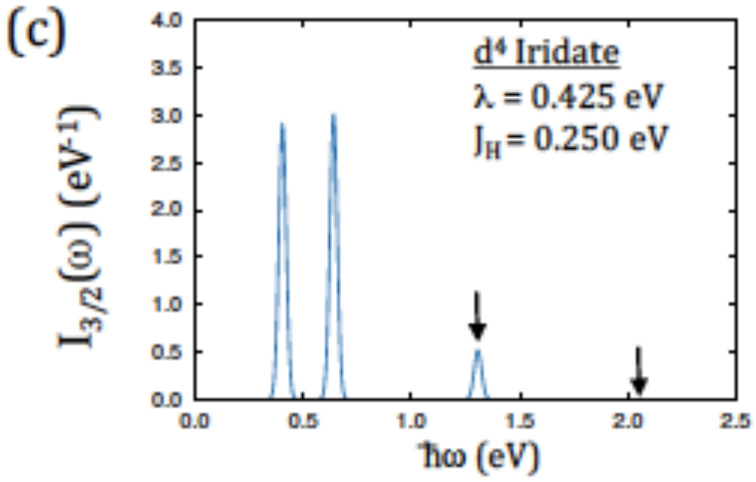
Exact diagonalization for RIXS

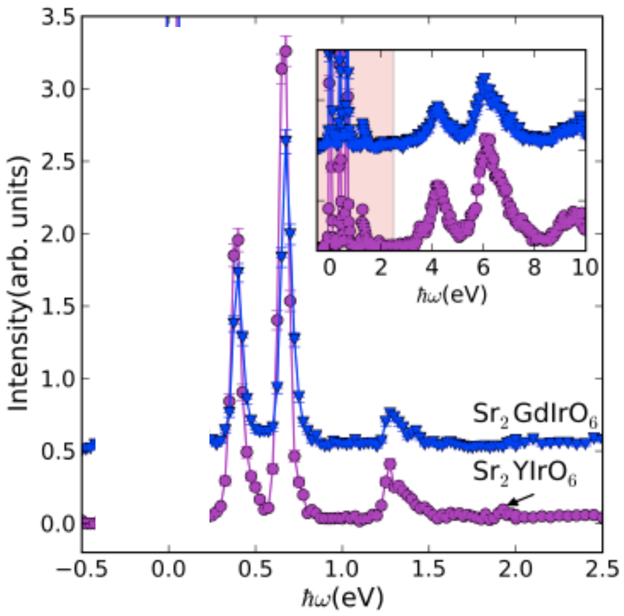












A. Paramekanti, et al, PRB 2018

Ground state degeneracies

d^1 4-fold degenerate: j=3/2

d^2 5-fold degenerate: J=2

d^3 4-fold degenerate: J=3/2

Candidates for multipolar orders

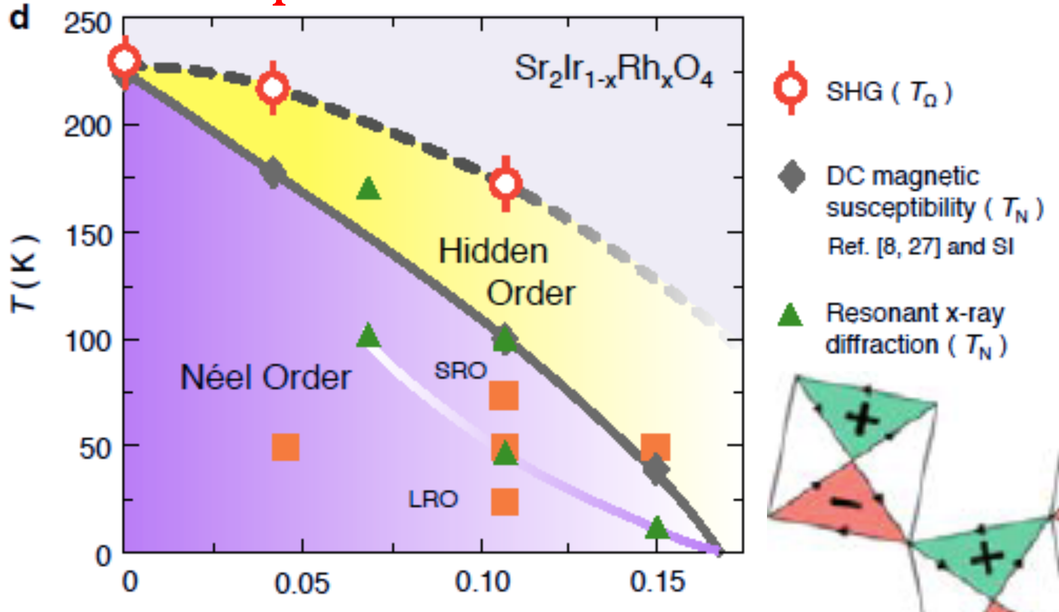
d^4 0-fold degenerate: J=0

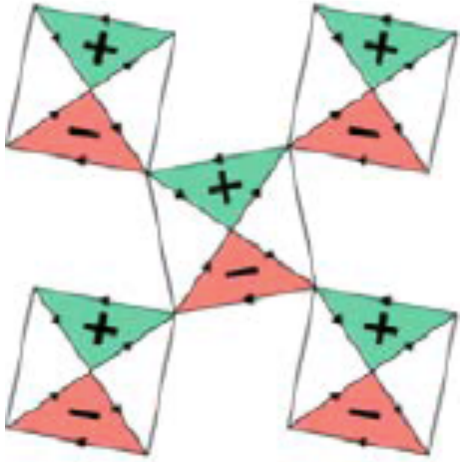
d^5 0-fold degenerate: j=1/2

Multipolar symmetry breaking in d-orbitals

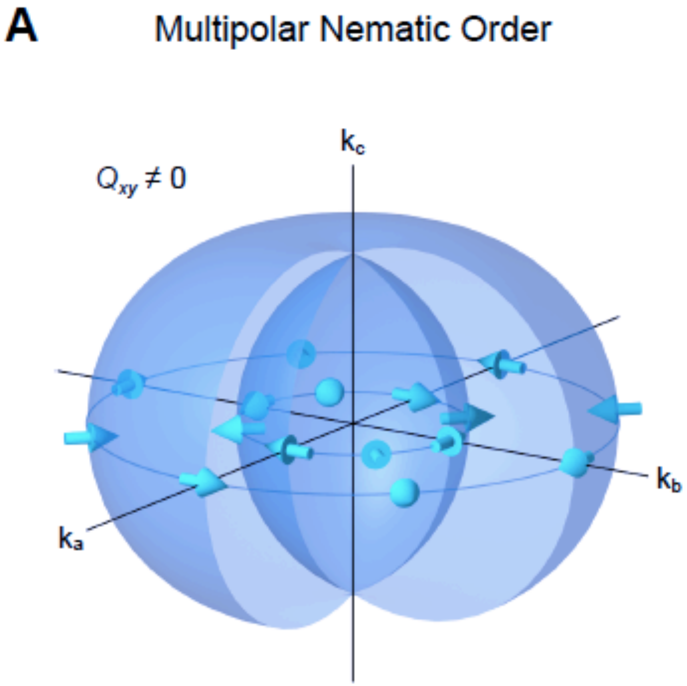
Inversion breaking -> SHG

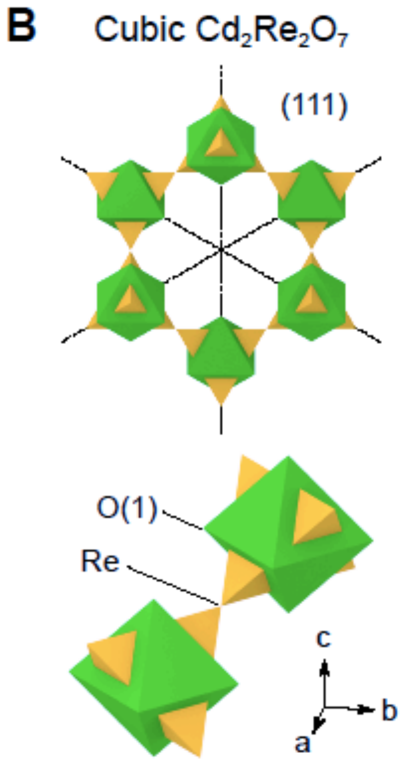
Parity-breaking transition
Toroidal moment?
Loop current order?





L. Zhao, et al,
Nat. Phys. (2016)





$$Q_{xy} \propto (k_x S_y + k_y S_x)$$

Parity-breaking quadrupolar order

Liang Fu, PRL (2015)
J. W. Harter, et al, Science (2017)

Quadrupolar+Dipolar ordering in d-orbitals

Rodrigo, Chen, Balents (PRB 2010)
Chen, Balents (PRB 2011)

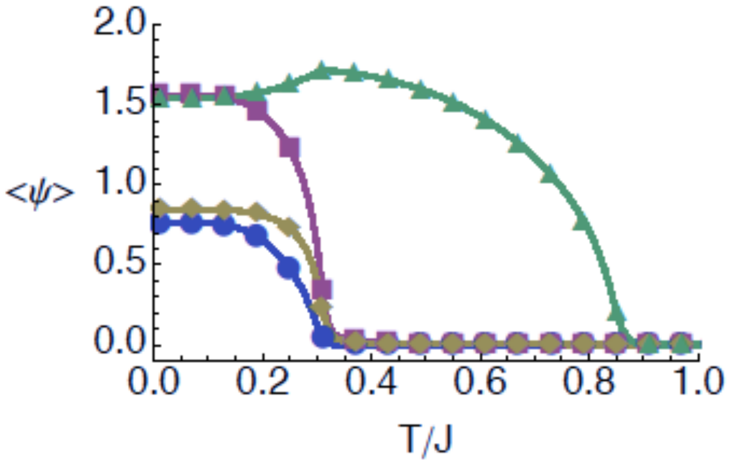
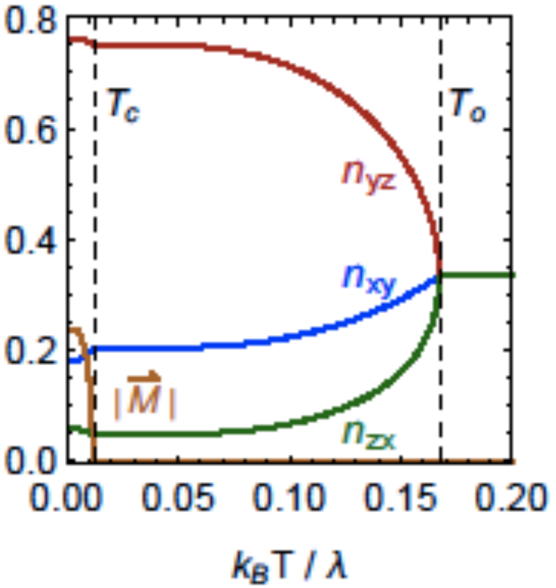
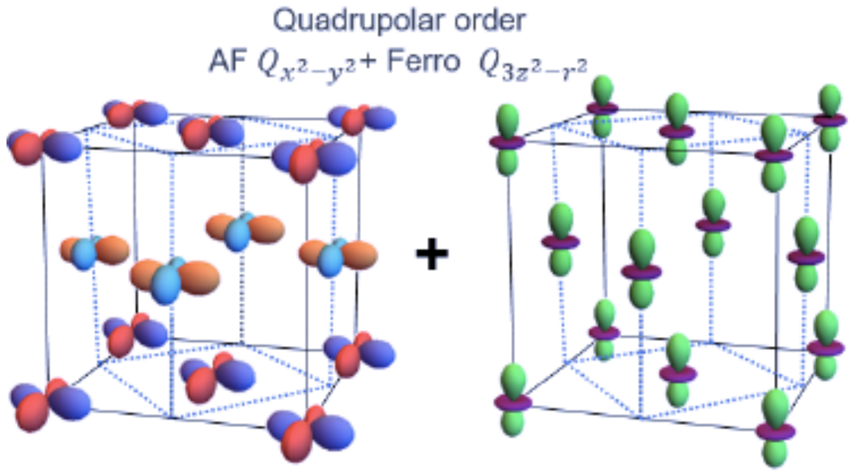
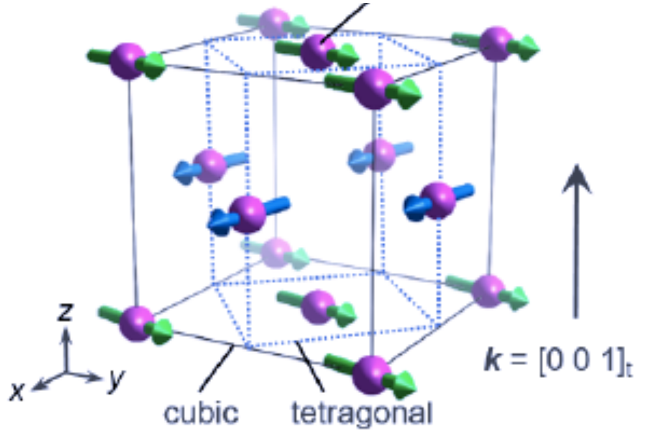


FIG. 9. (Color online) Temperature dependence of order parameters for $J'/J = 0.3$, $V/J = 0.3$. For these parameters, there is a continuous normal to quadrupolar transition, at $T/J \approx 0.85$, followed by a continuous quadrupolar to FM110 transition at $T/J \approx 0.33$. The four order parameters plotted are: squares (red online) $|\langle T_{A,\alpha} - T_{B,\alpha}\rangle|/2$, circles (blue online) $\langle Q_A^{3z^2} + Q_B^{3z^2}\rangle/2$, diamonds (yellow online) $|\langle j_A + j_B\rangle|/2$, and triangles (green online) $\langle Q_A^{x^2-y^2} - Q_B^{x^2-y^2}\rangle/2$.

Svoboda, Randeria, Trivedi
[+ W. Zhang] (arXiv:2017)





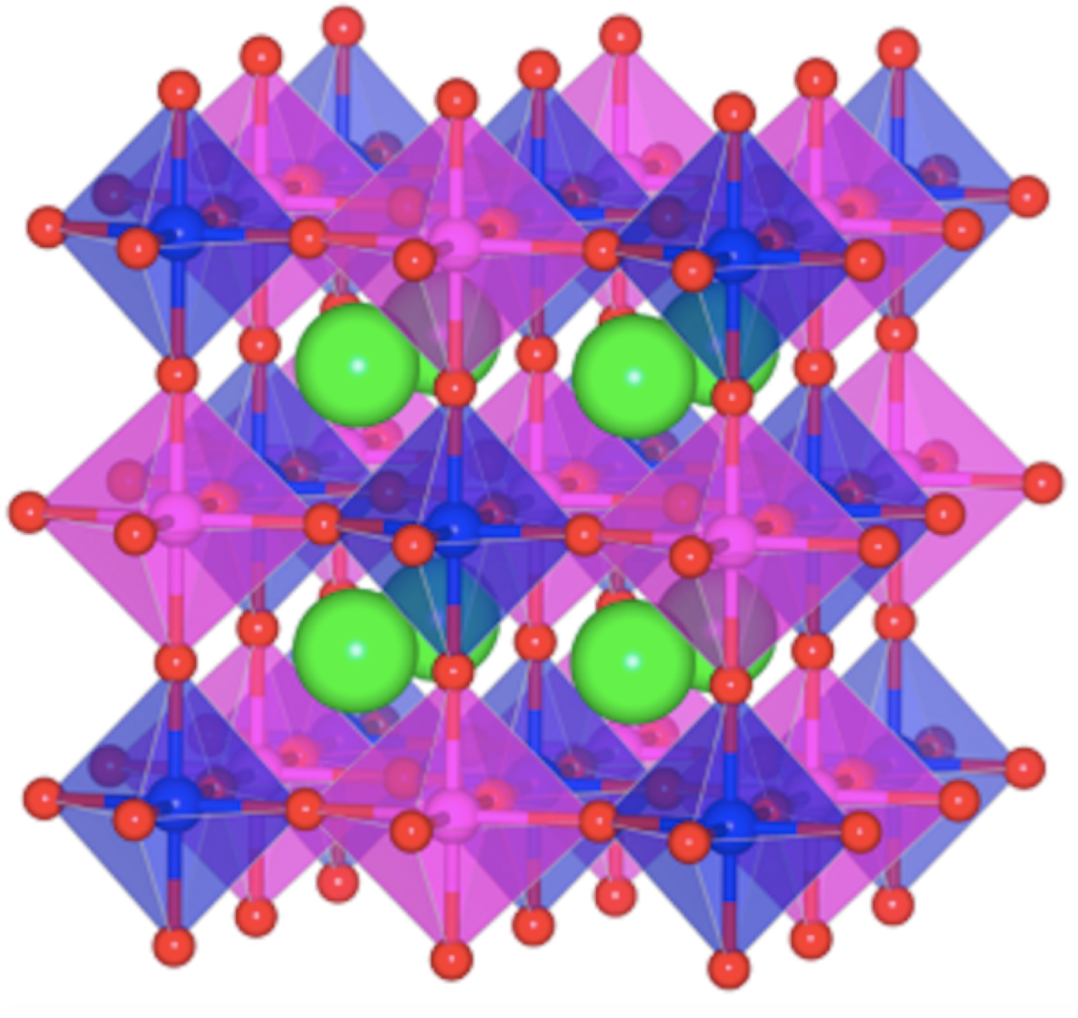


L. Lu, et al, Nat. Comm. (2017): Ba2NaOsO6
D. Hirai, Z. Hiroi, JPSJ (2019): Ba2MgReO6
D. Hirai, et al, arXiv:2004.13928: Ba2MgReO6

TQ = 33K (seen from non-resonant XRD)
TN = 18K (seen from resonant XRD and polarization analysis)

Ordered double perovskites with SOC

Mott insulator with J=2 spins



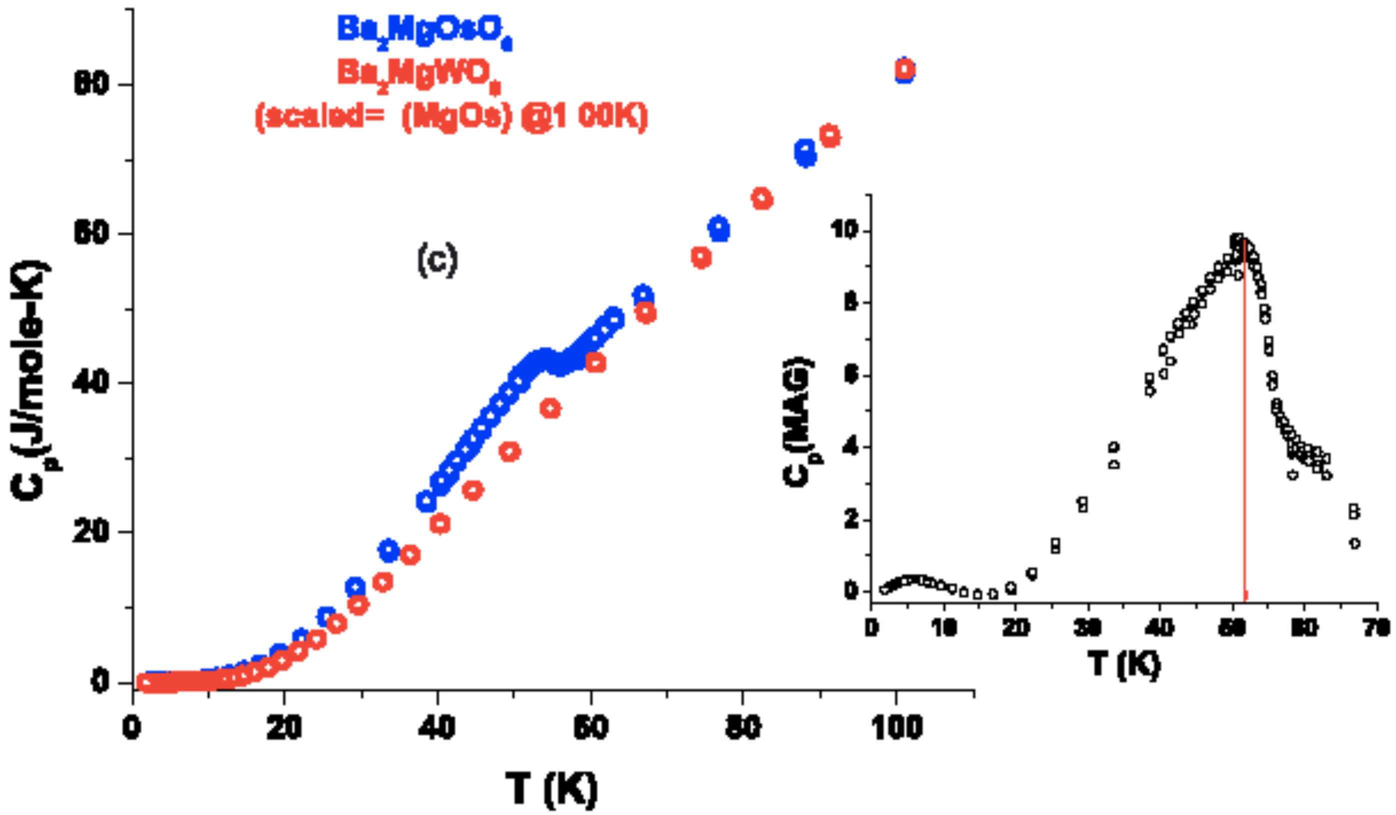
Ba_2Mg**Os**O_6

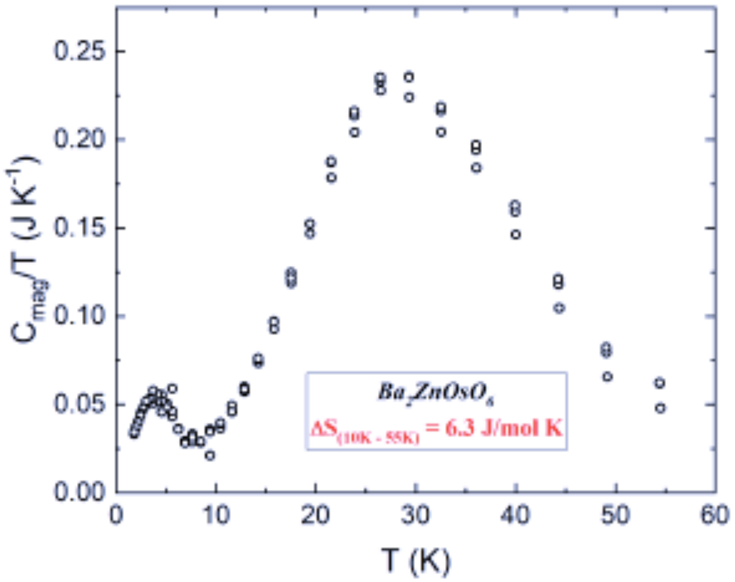
Ba_2Ca**Os**O_6

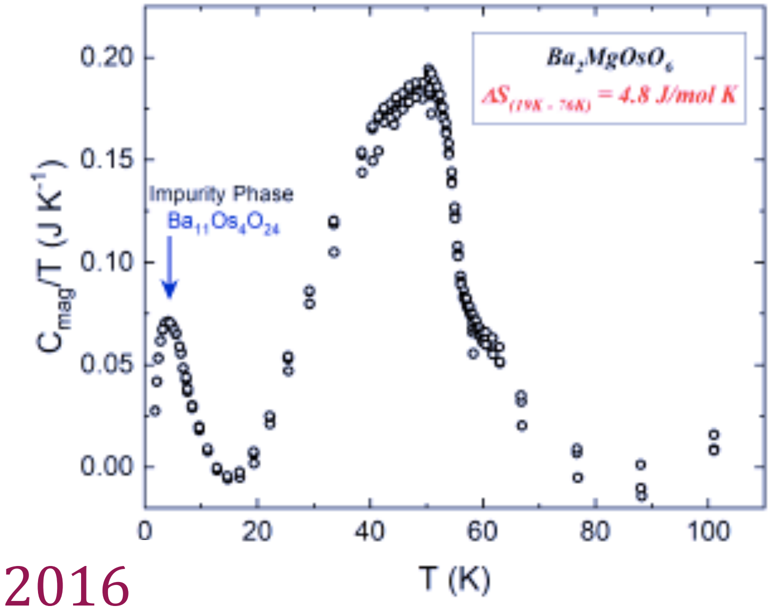
Ba_2Zn**Os**O_6

Os^{5+}:$5d^2$ in checkerboard pattern (FCC lattice)

Thermodynamics: Specific heat and entropy



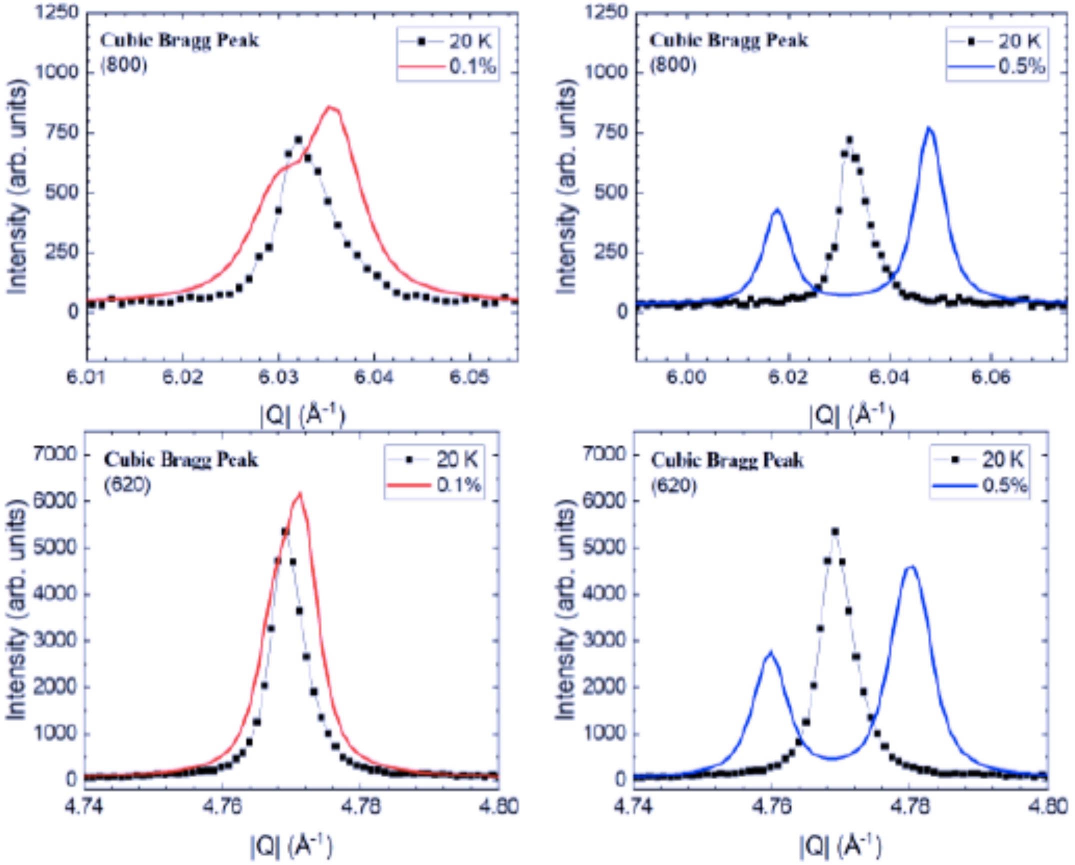




- Phase transition at T ~ 30-50K
- Entropy recovered << R ln (5): Not J=2?
- Rln(2) ~ 5.8 J/mol-K; Rln(3) ~ 9.1 J/mol-K

C. A. Marjerrison, et al, PRB 2016

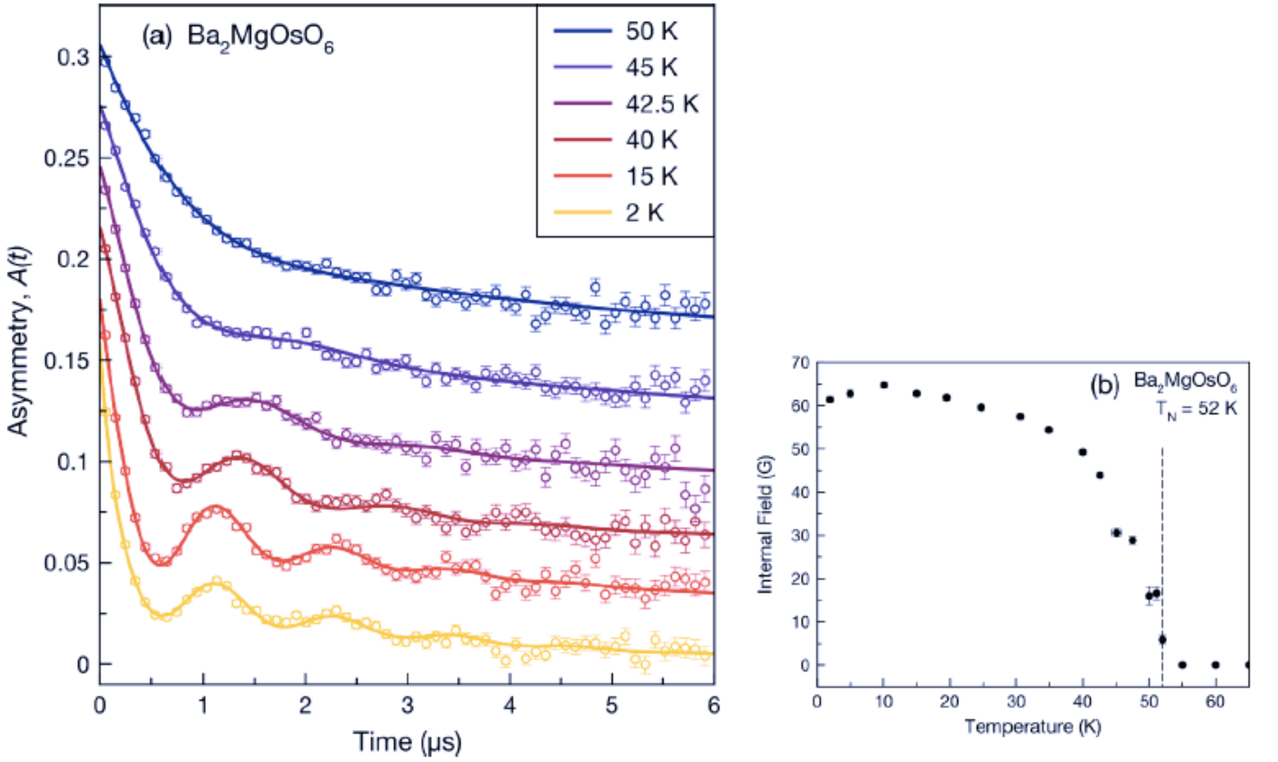
Xray diffraction



Perfectly cubic, Fm$\bar{3}$m
(<0.1% distortion)

D. D. Maharaj, et al, PRL (2020)

μSR



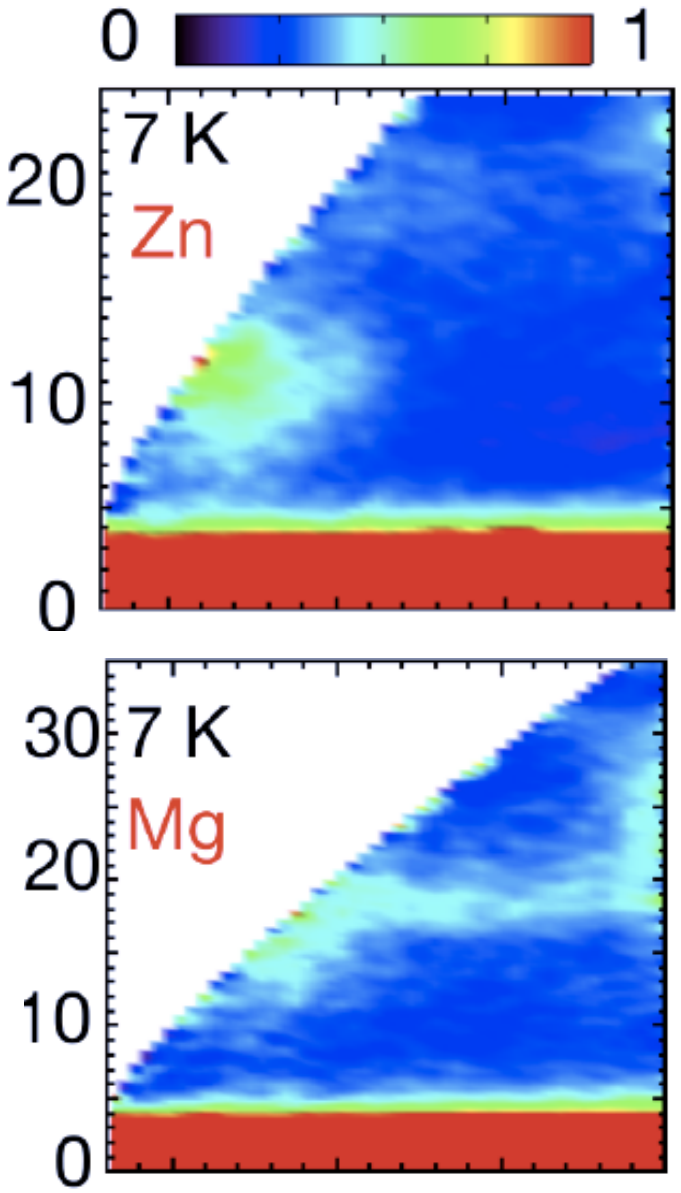
Signature of time-reversal symmetry breaking

C. M. Thompson, et al, JPCM (2014)

Susceptibility, neutron diffraction, neutron scattering

System	T^*	θ_{CW}	μ_{ord}
Ba_2CaOsO_6	49	−156.2(3)	$< 0.11\mu_B$
Ba_2MgOsO_6	51	−120(1)	$< 0.13\mu_B$
Ba_2ZnOsO_6	30	−149.0(4)	$< 0.06\mu_B$

- Moderate frustration
- No sign of ordered moment!
- Spin gap in inelastic scattering



D. D. Maharaj, et al, PRL (2020)

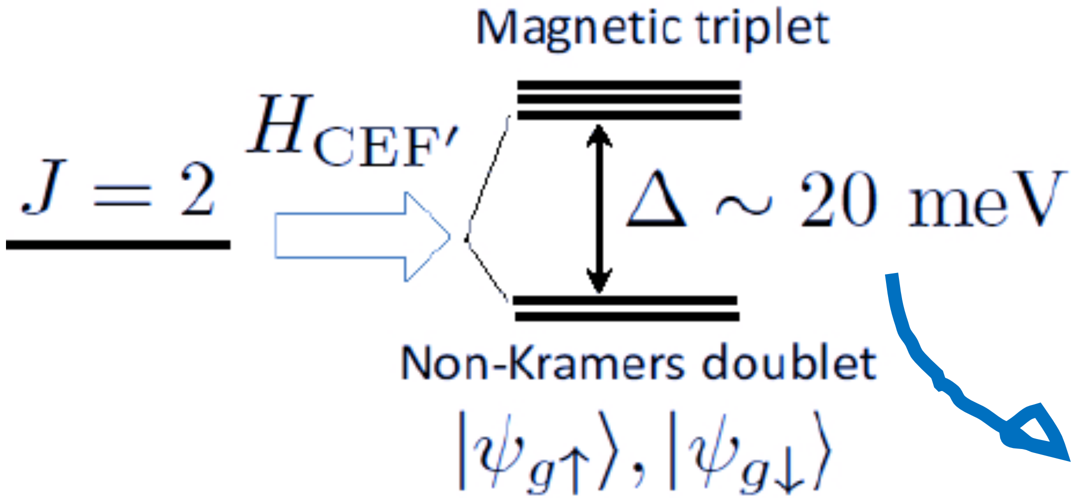
Puzzles

- ❑ Entropy recovered across T* << R ln(5)
- ❑ No sign of a second transition at higher T
- ❑ Remains cubic down to T << T*
- ❑ Time reversal breaking via mSR
- ❑ No sign of magnetic Bragg peaks
- ❑ Spin gap in neutron scattering ~ 10 meV

The correct "atomic limit"

New Idea:

- J=2 splits in octahedral environment
- Unresolved in RIXS



✓ Spin gap
✓ No dipole order
✓ Entropy: R ln(2)

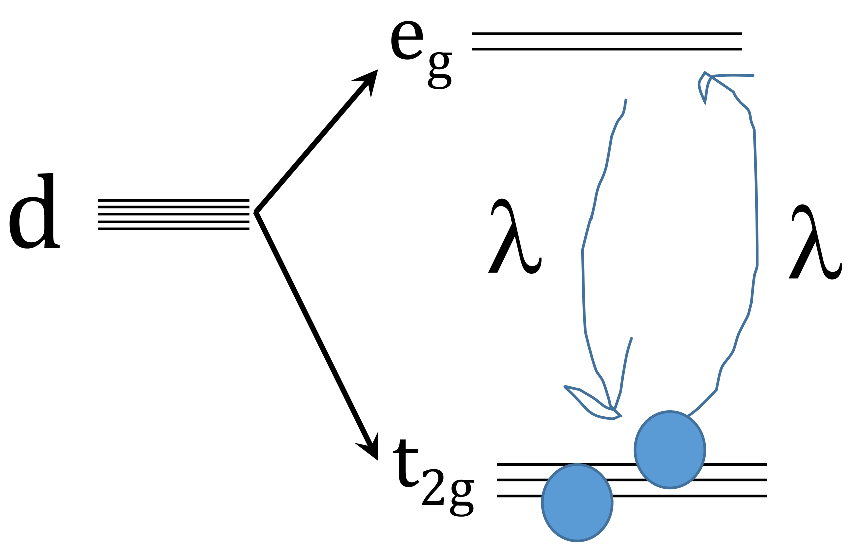
$$|\psi_{g,\uparrow}\rangle = |0\rangle$$

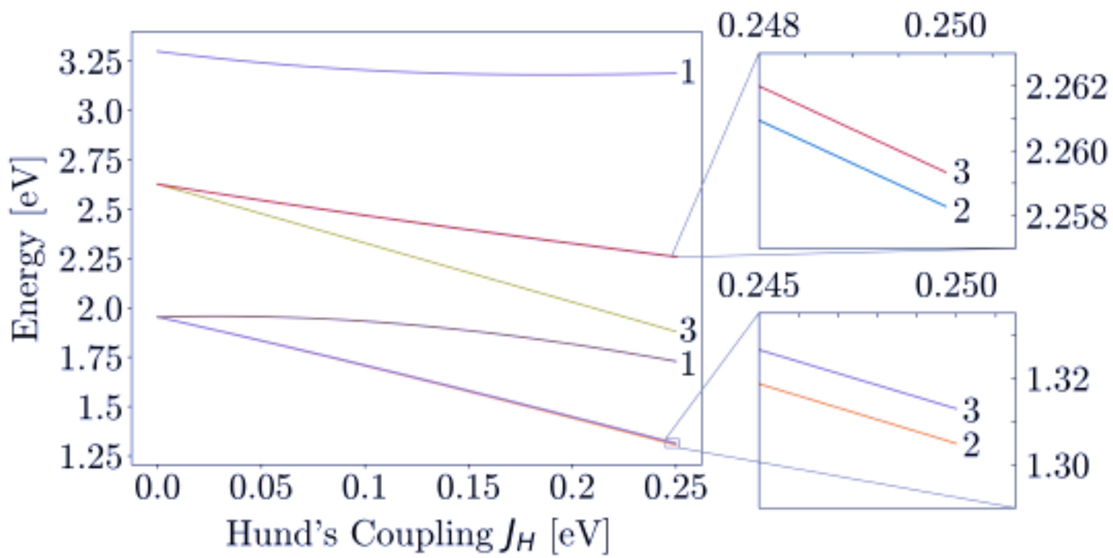
$$|\psi_{g,\downarrow}\rangle = \frac{1}{\sqrt{2}}(|2\rangle + |-2\rangle)$$

Splitting originates from t_{2g}-e_g coupling!

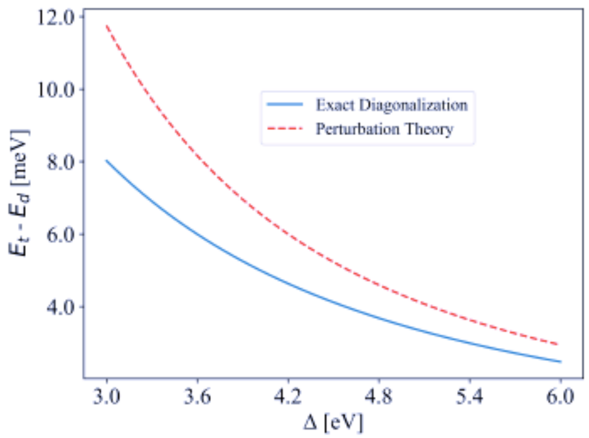
Cannot blindly drop e_g orbitals - Might be crucial for low energy physics!

The correct "atomic limit"





$$\Delta \sim O(J_H \lambda^2/V)$$



$$H_{\mathrm{CEF}} = -V_{\mathrm{eff}}(\mathcal{O}_{40} + 5\mathcal{O}_{44})$$

$$\mathcal{O}_{40} = 35J_z^4 - (30J(J+1) - 25)J_z^2 + 3J^2(J+1)^2 - 6J(J+1),$$

$$\mathcal{O}_{44} = \frac{1}{2}(J_+^4 + J_-^4).$$

Exchange interactions

$$\tau_x \propto J_x^2 - J_y^2$$

$$\tau_z \propto 3J_z^2 - J^2$$

"XY" Quadrupolar charge density

$$\tau_y \propto \mathrm{Sym}\,[J_x J_y J_z]$$

Ising magnetic octupole

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

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

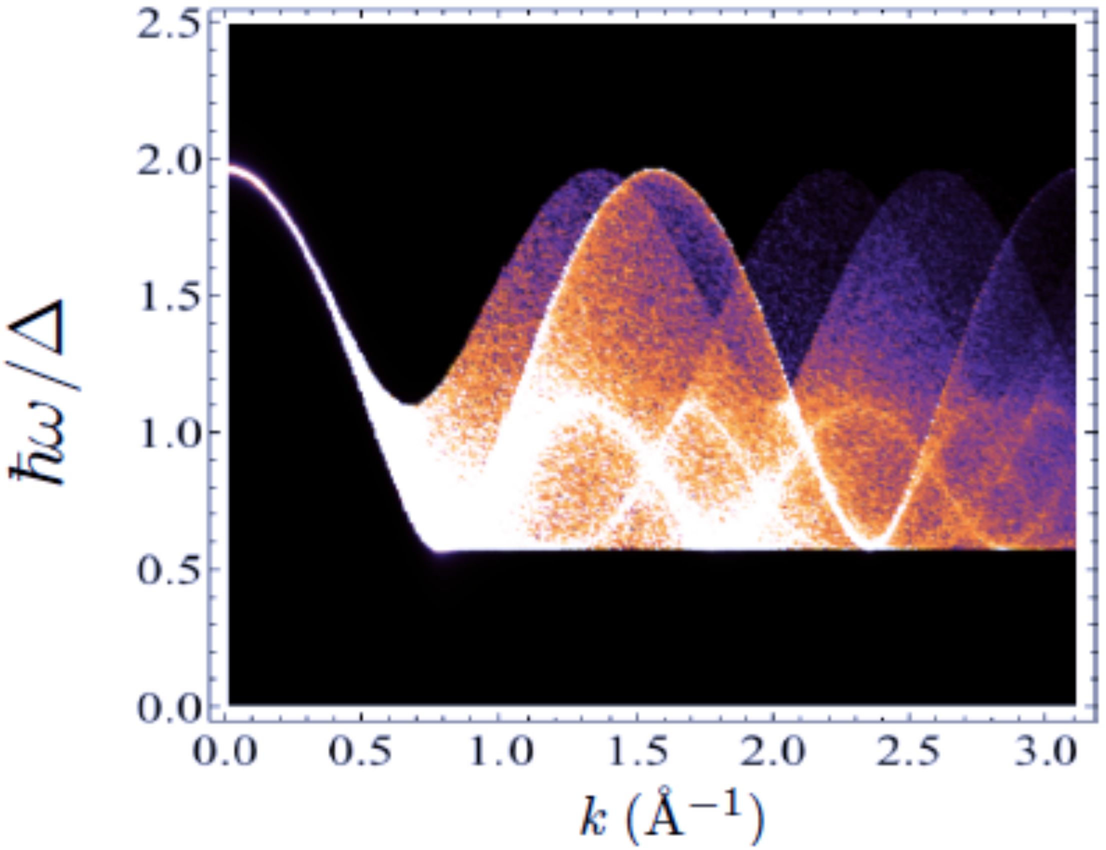
$$H^{\mathrm{Quad}}_{xy} = \sum_{\langle ij\rangle_{xy}} \left[(-\gamma_0 + 6\frac{\gamma_m \gamma_2}{\Delta})\tau_{ix}\tau_{jx} + (\gamma_1 - \frac{9}{8}\frac{\gamma_2^2}{\Delta})\tau_{iz}\tau_{jz} \right]$$

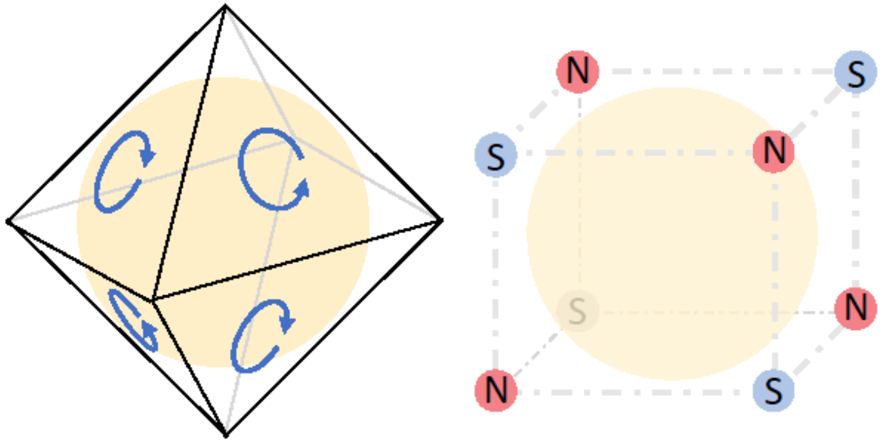
$$H^{(2)}_{\mathrm{Oct}} = -6\frac{\gamma_m \gamma_2}{\Delta} \sum_{\langle ij\rangle} \tau_{iy}\tau_{jy}$$

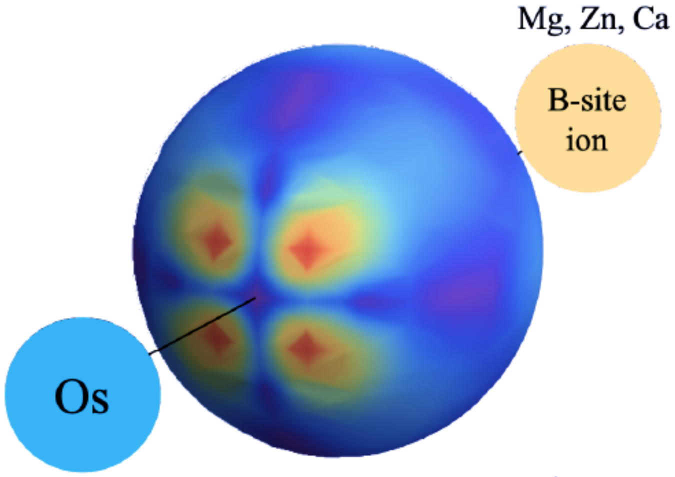
✓ Leads to ferro-octupolar coupling from cross-term

AP, D. D. Maharaj, Bruce Gaulin, PRB 2020

Ferro-octupolar order: Neutron Scattering, μSR



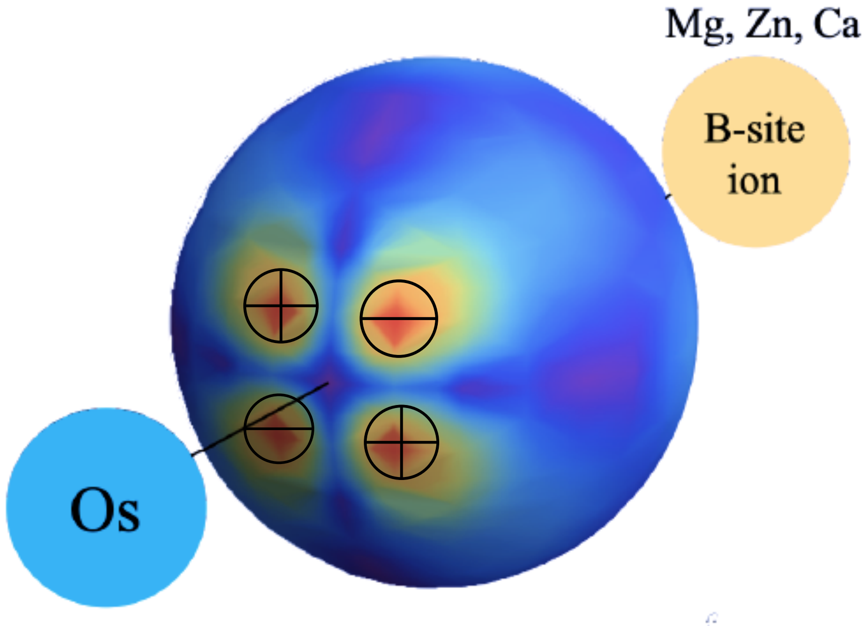


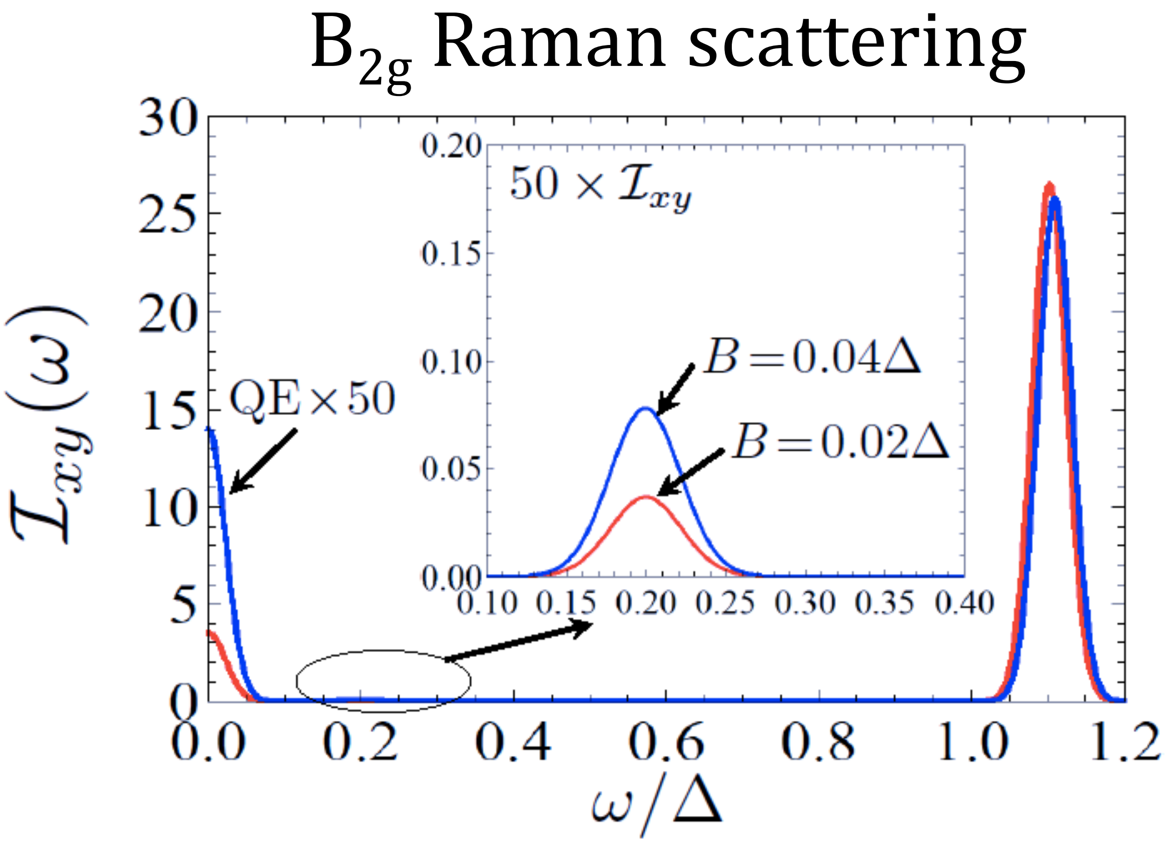


✓ Spin-gapped magnetic exciton

✓ Loop current order, B ~ 30G

Predictions for NMR, Raman scattering





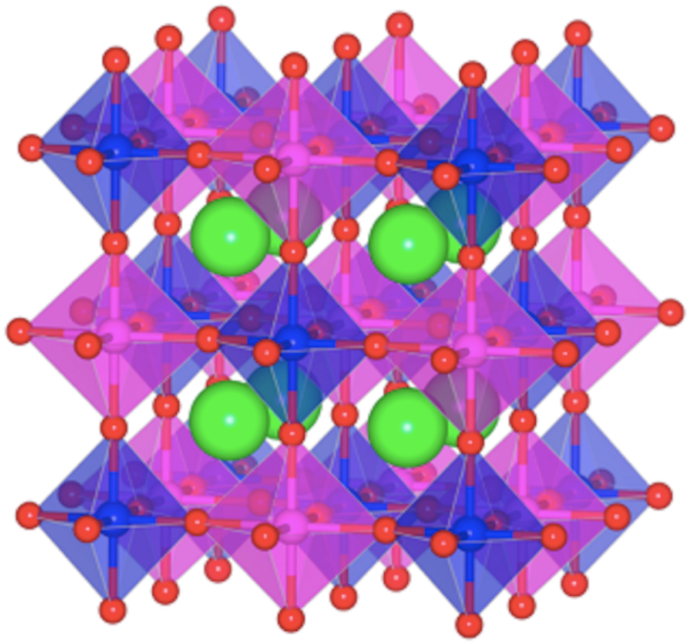
- Oxygen NMR should not see T* transition
- Break C4 symmetry -> NMR signal

- Field Bz induces Qxy quadrupolar order
- New quasielastic peak
- New transition between split doublet

AP, D. D. Maharaj, Bruce Gaulin, PRB 2020

S. Voleti, et al, PRB 2020

- Ba_2MgOsO_6
- Ba_2CaOsO_6
- Ba_2ZnOsO_6



Octupolar order

✓ Entropy recovered across T* ~ R ln(2)
✓ Remains cubic down to T << T*
✓ Time reversal breaking via mSR
✓ No sign of magnetic Bragg peaks
✓ Spin gap in neutron scattering ~ 10 meV

Old parallel: complex eg orbital order (J. van den Brink, D. Khomskii PRB 2001)

Collaborators and references

Bruce Gaulin

Dalini Maharaj

Sreekar Voleti

Graeme Luke

- D. D. Maharaj, G. Sala, M. B. Stone, E. Kermarrec, C. Ritter, F. Fauth, C. A. Marjerrison, J. E. Greedan, AP, Bruce Gaulin, PRL 124, 087206 (2020)
- AP, D. D. Maharaj, Bruce Gaulin, PRB 101, 054439 (2020)
- S. Voleti, D.D. Maharaj, Bruce Gaulin, Graeme Luke, AP, PRB 101, 155118 (2020)

Summary and open issues

- First candidate for d-orbital octupolar order?
- High Tc octupolar symmetry breaking
- Predictions for NMR and Raman scattering
- Elasticity (A. Patri, Nat Comm 2019; M. Sorensen, I. Fisher arXiv:2020)

- Can we dynamically “switch” the Ising order?
- Can doping lead to fluctuating loop currents?
- Are multipolar orders useful for anything?