Title: Tuning multipolar orders and critical points in d-orbital Mott insulators

Speakers: Arun Paramekanti

Series: Quantum Matter

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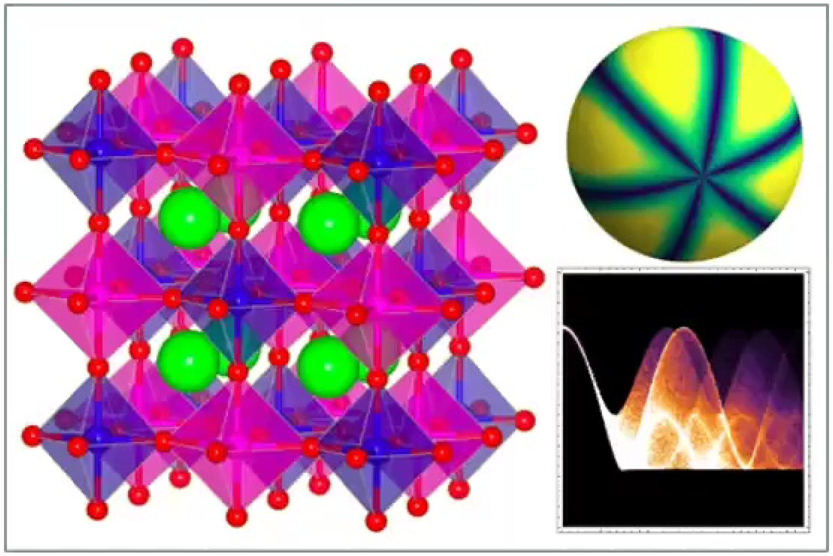
Abstract: Traditionally, magnetism in solids deals with ordering patterns of the electron magnetic dipole moment, as probed, for instance, via neutron diffraction. However, f-electron heavy fermion systems are well-known candidates for more complex forms of symmetry breaking, involving higher-order magnetic or electric multipoles. In this talk, I will discuss our recent theoretical proposal for Ising octupolar order in d-orbital systems, which appears to explain a wide range of experiments in certain 5d transition metal oxides with spin-orbit coupling. The proposed Ising ferro-octupolar order is shown to be linked to a type of orbital loop-current order. Deviations from cubic symmetry, via strain or surfaces, induces a transverse field on the octupolar order which can lead to surface quantum phase transitions, or transitions in thin films or in strained 3D crystals. We propose further experimental tests of our proposal.

Octupolar order and tuning of quantum criticality
Application to d-orbital Mott insulators



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(University of Toronto)

PI Quantum Matter Series
22 Nov 2021

UNIVERSITY OF TORONTO

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TATA INSTITUTE OF FUNDAMENTAL RESEARCH

Collaborators

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Arijit Halder

Bruce Gaulin

Dalini Maharaj

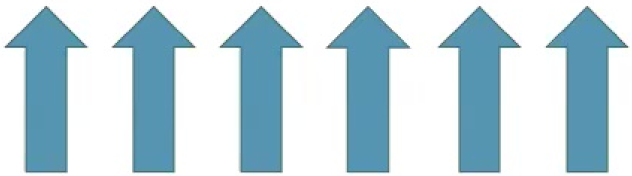
Graeme Luke

References

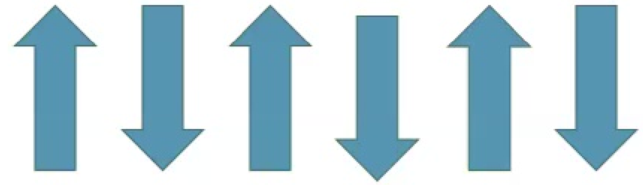
- D. D. Maharaj, G. Sala, M. B. Stone, E. Kermarrec, C. Ritter, F. Fauth, C. A. Marjerrison, J. E. Greedan, AP, Bruce Gaulin, ***PRL 124, 087206 (2020)***
- AP, D. D. Maharaj, Bruce Gaulin, ***PRB 101, 054439 (2020)***
- S. Voleti, D.D. Maharaj, Bruce Gaulin, Graeme Luke, AP, ***PRB 101, 155118 (2020)***
- S. Voleti, A. Halder, AP,***Phys. Rev. B 104, 174431 (2021)***

Multipolar orders

Dipolar order

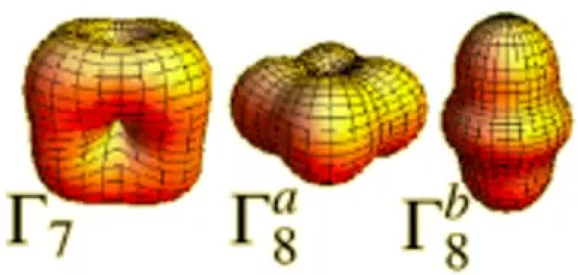


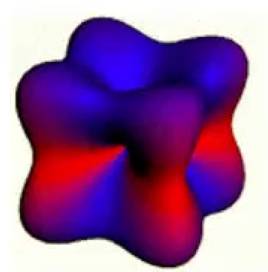
Ferromagnetism
Easily manifest



Antiferromagnetism
Probe: Neutron diffraction
Neutron: dipole interaction

Multipolar orders





SOC drives complex spin+orbital density distributions (mostly 5f heavy fermions)
"**Hidden order**": URu_2Si_2, NpO_2,...
(Palstra et al, PRL 1985; M.B. Maple, et al, PRL 1986;P. Chandra, et al, 2002; Haule, Kotliar, Nat. Phys 2009; Santini, et al, RMP 2009; Mydosh, Oppeneer, RMP 2011; Ikeda, et al, Nat. Phys. 2012)

5f heavy fermions v/s 5d transition metals

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

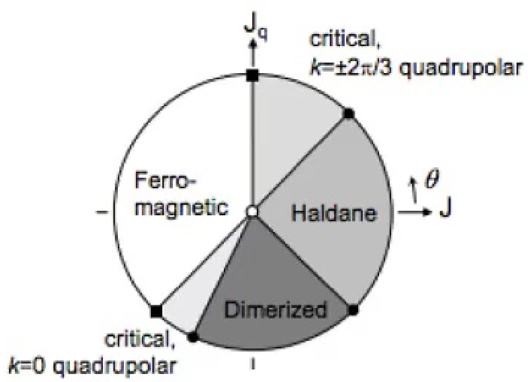
[write crystal field potential in terms of L]
SOC~ 0.4eV; Hund's coupling ~ 0.25eV

Can 5d systems also exhibit physics of heavy fermion systems?

Simple magnetic example: S=1 spin models

Local 3x3 Hermitian operators = 9

- *Identity*
- S_x, S_y, S_z *Dipoles*
- $S_a S_b + S_b S_a - \frac{2}{3} S(S+1)$ *Quadrupoles: Q_{ab}*



$$H = \sum_{i=1}^{n} \left(cos\theta \vec{S}_i \cdot \vec{S}_{i+1} + sin\theta (\vec{S}_i \cdot \vec{S}_{i+1})^2 \right)$$

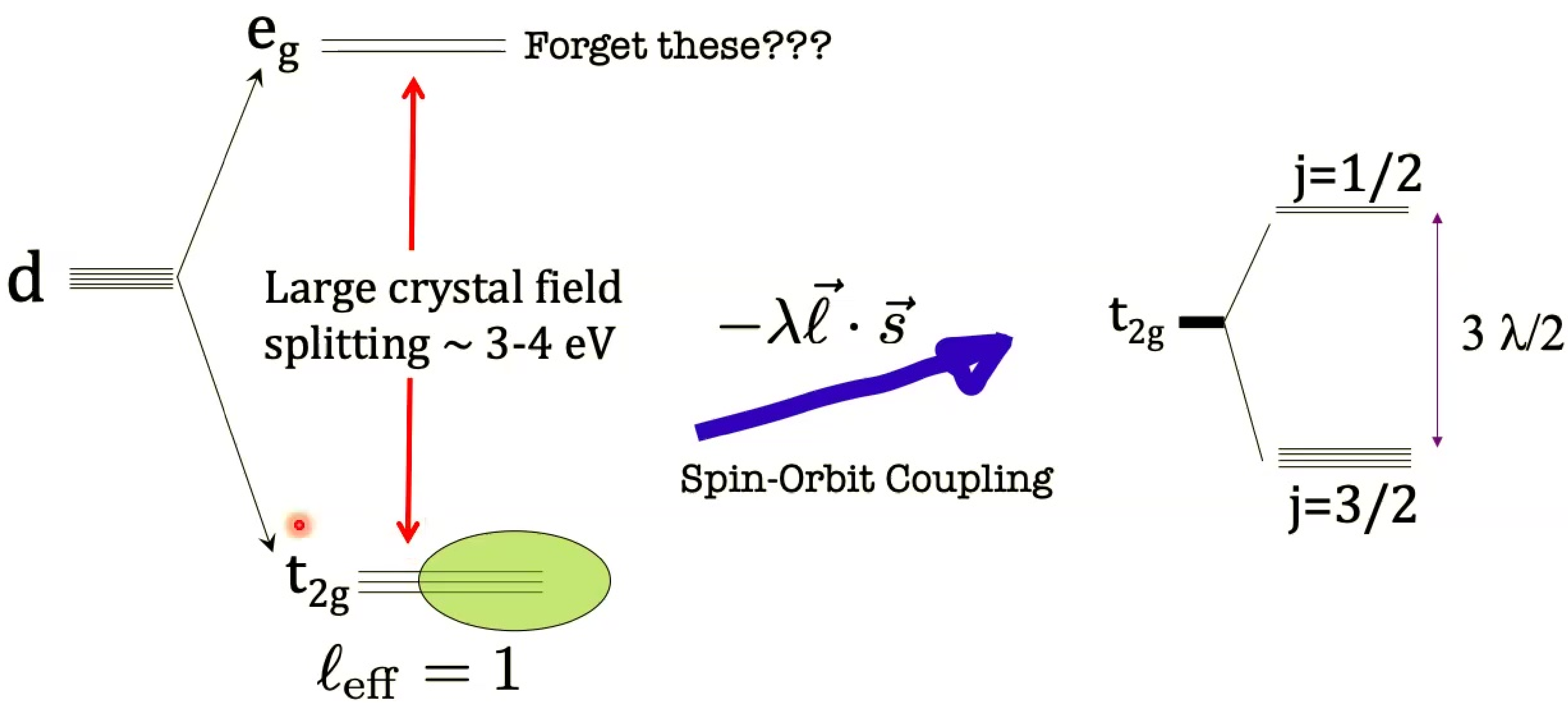
Chubukov, PRB 1991
Lauchli, Schmid, Trebst, PRB 2006

$NiGa_2S_4$?
Tsunetsugu, Arikawa JPSJ 2006; S. Bhattacharjee, Shenoy, Senthil, PRB 2006; Stoudenmire, Trebst, Balents, PRB 2009;

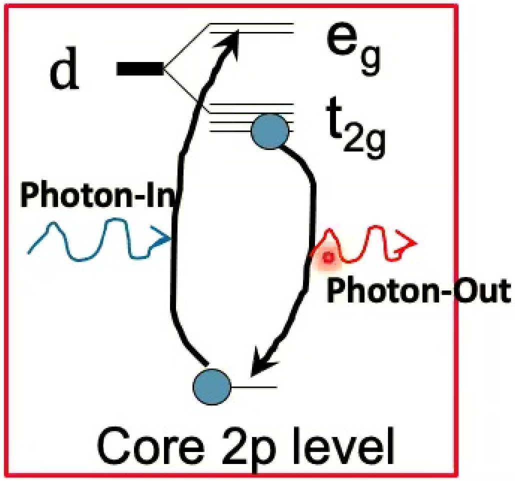
FeSe?
Wang, Hu, Nevidomskyy PRL 2016; Hu, et al, Q. Si, PRR 2020

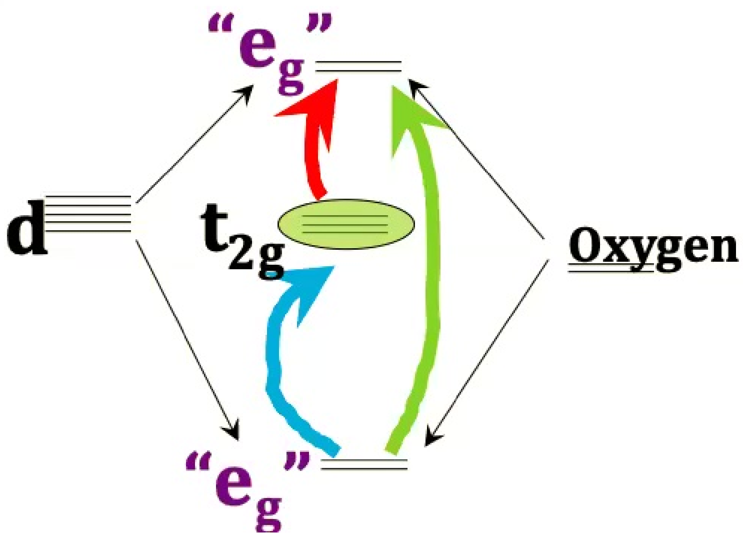
Multipolar order in d-orbital systems?

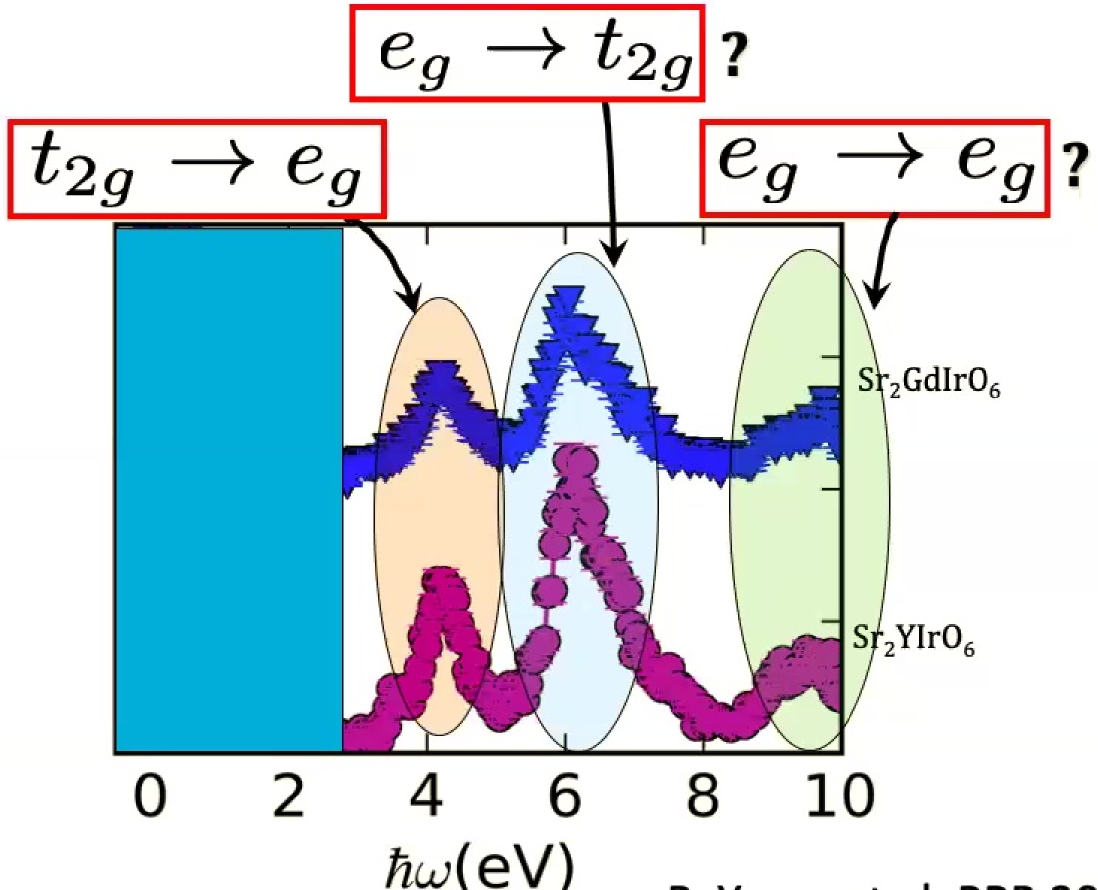
Atomic limit



Resonant Inelastic X-ray Scattering: Seeing the Crystal Field Levels



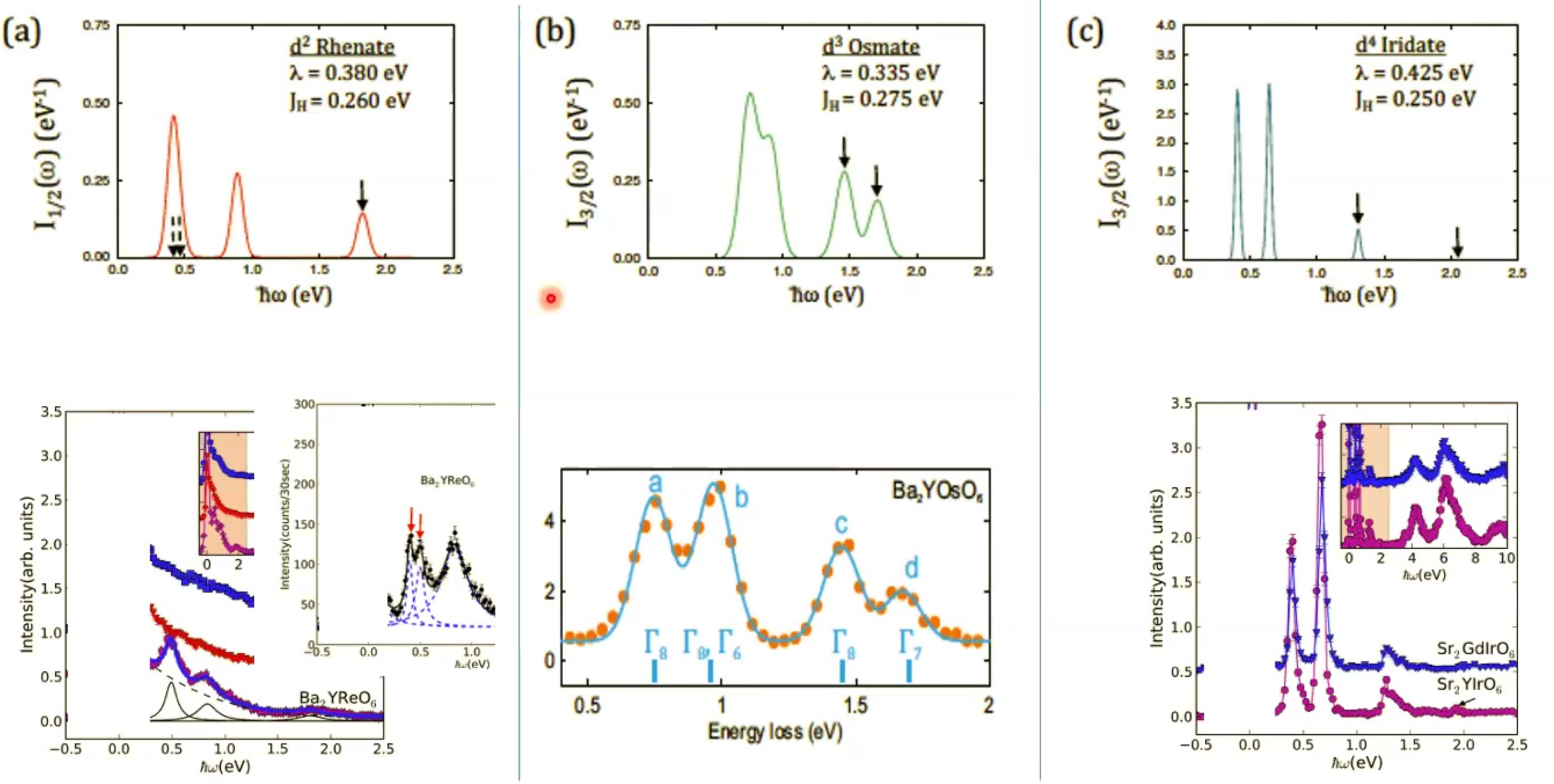




B. Yuan, et al, PRB 2017

AP et al, PRB 2018

Exact diagonalization for RIXS



A. Paramekanti, et al, PRB 2018

Ground state degeneracies (upon including interactions in t_{2g})

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 from spin-orbit entanglement

d^4 0-fold degenerate: J=0

Band insulator: Exciton condensation? J=1 triplons? Khaliullin PRL 2013; Chaloupka, Khaliullin PRL 2016

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

Kitaev-Gamma physics, "Twisted" Hubbard model, etc -- Jackeli, Khaliullin, PRL 2009; Jeff Rau, et al, PRL 2014; Wang, Senthil PRL 2011

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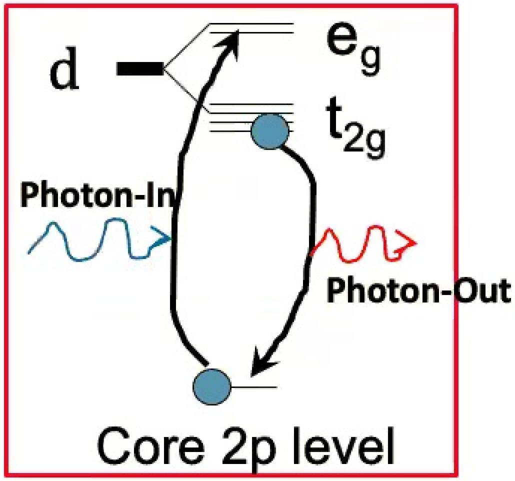
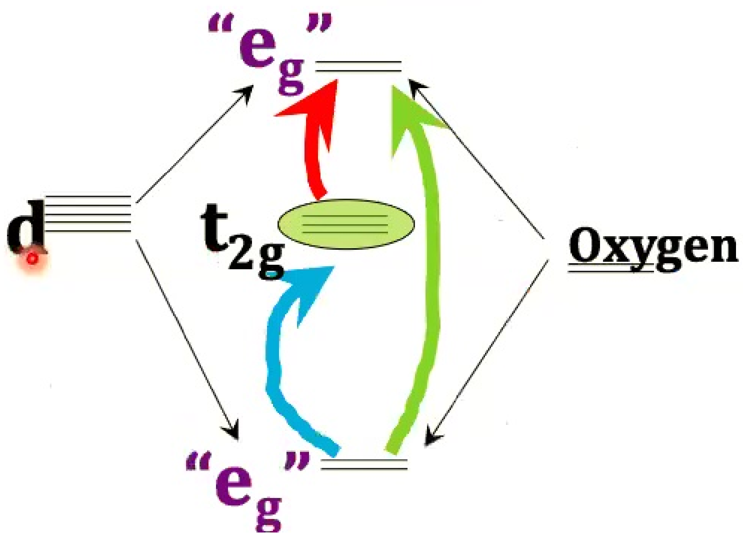
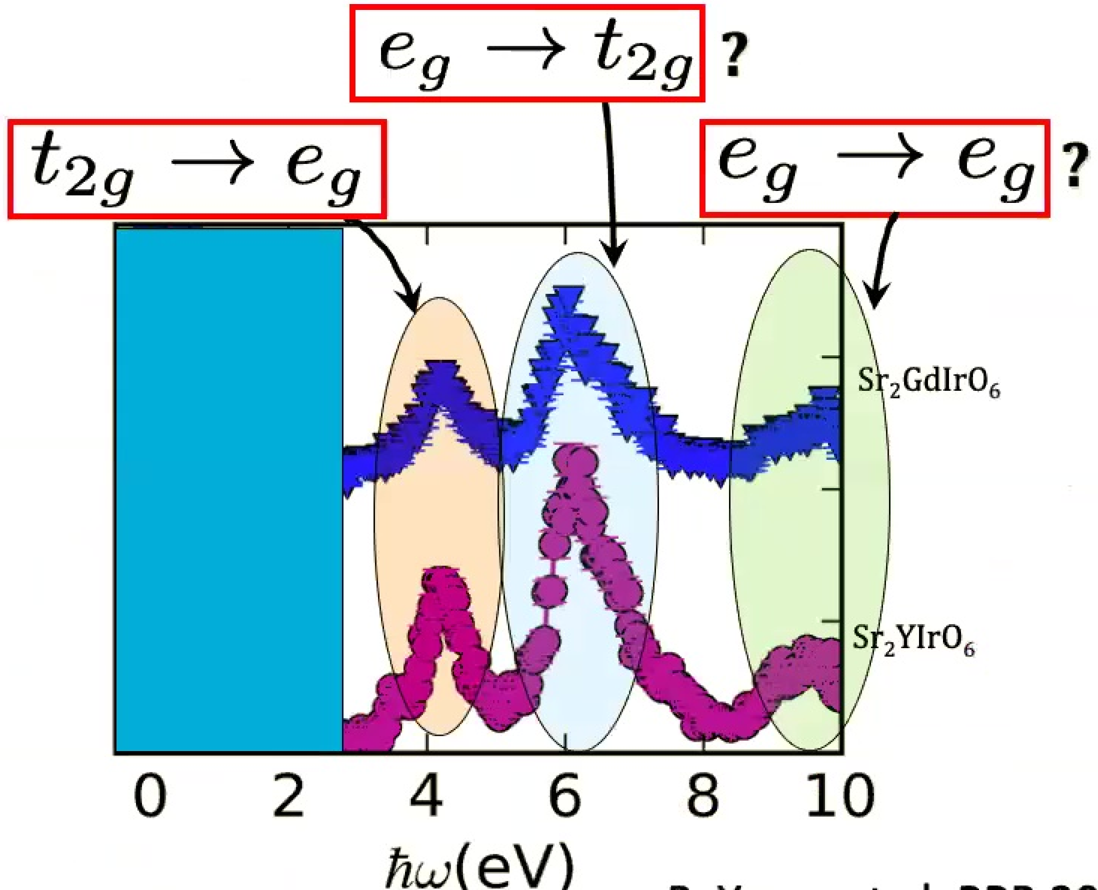


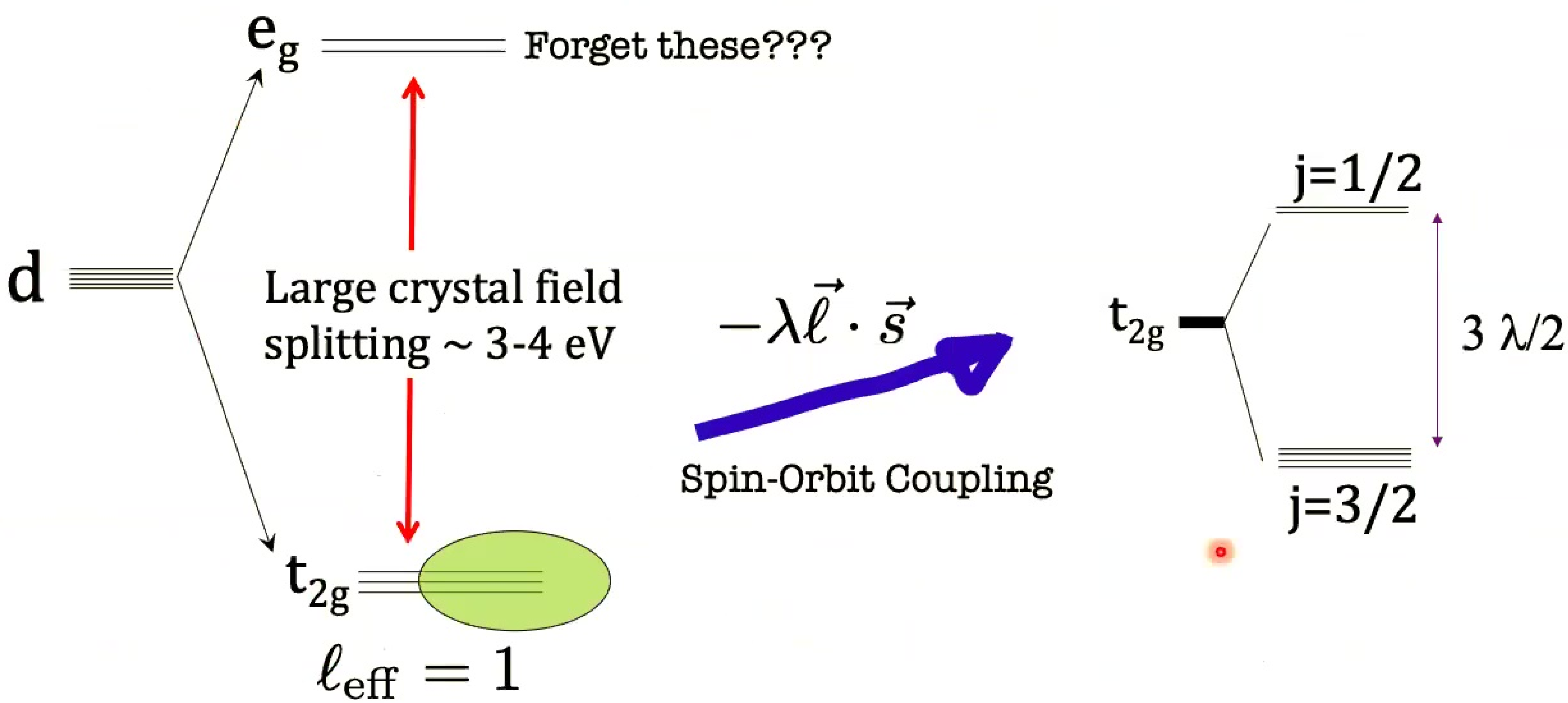




B. Yuan, et al, PRB 2017
AP et al, PRB 2018

Multipolar order in d-orbital systems?

Atomic limit



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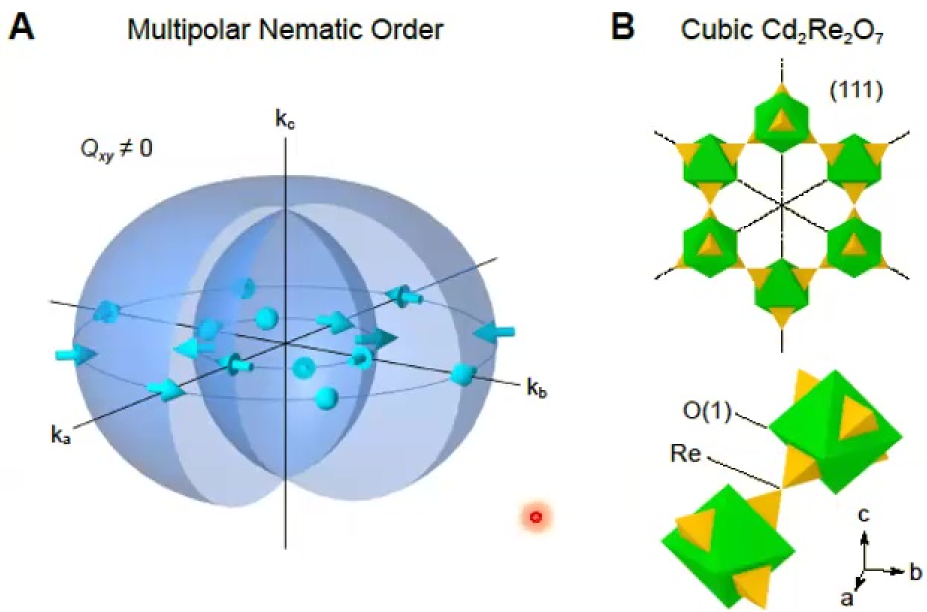
d^5 2-fold degenerate: $j=1/2$ → *Kitaev-Gamma physics, "Twisted" Hubbard model, etc -- Jackeli, Khaliullin, PRL 2009; Jeff Rau, et al, PRL 2014; Wang, Senthil PRL 2011*

Quadrupolar symmetry breaking in d-orbitals

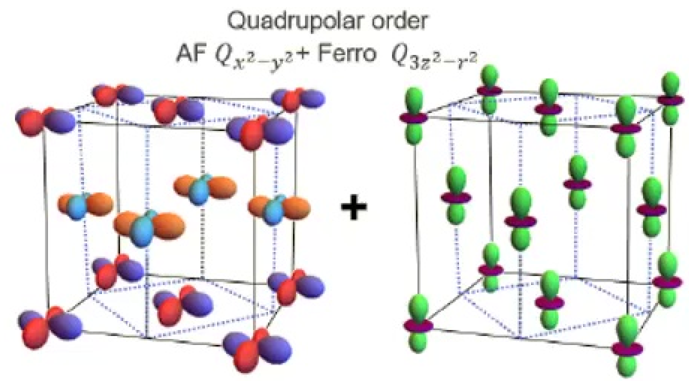


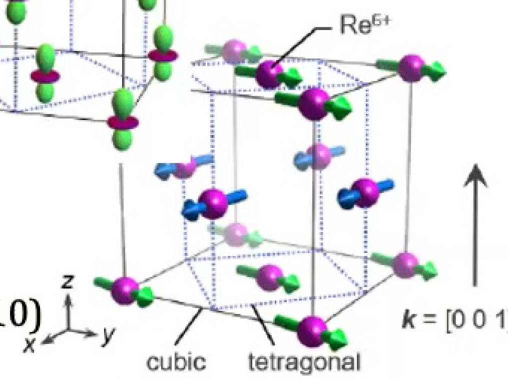
$$Q_{xy} \propto (k_x S_y + k_y S_x)$$

Parity-breaking quadrupolar order

Liang Fu, PRL (2015)

J. W. Harter, et al, Science (2017)





Pereira, Chen, Balents (PRB 2010)

Chen, Balents (PRB 2011)

Svoboda, W. Zhang, Randeria, Trivedi (PRB 2021)

L. Lu, et al, Nat. Comm. (2017): Ba2NaOsO6

D. Hirai, Z. Hiroi, JPSJ (2019): Ba2MgReO6

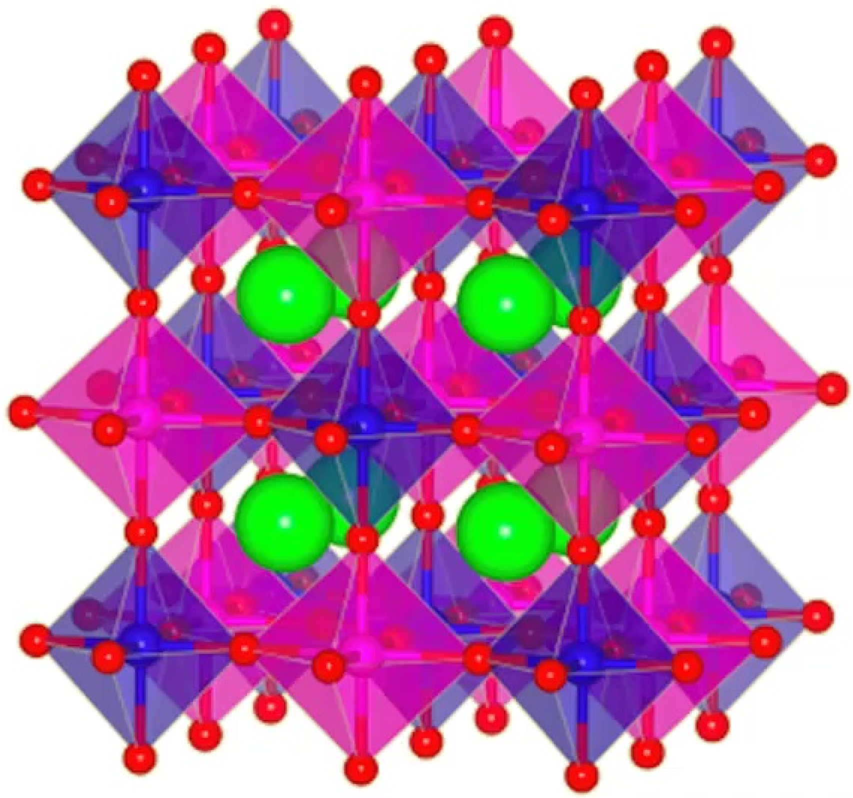
D. Hirai, et al, arXiv:2004.13928: Ba2MgReO6

D. Hirai, et al, Phys. Rev. Research 2, 022063 (2020)

TQ = 33K (seen from non-resonant XRD)

TN = 18K (seen from resonant XRD and polarization analysis)

"*Hidden order*" in Osmate Mott insulators

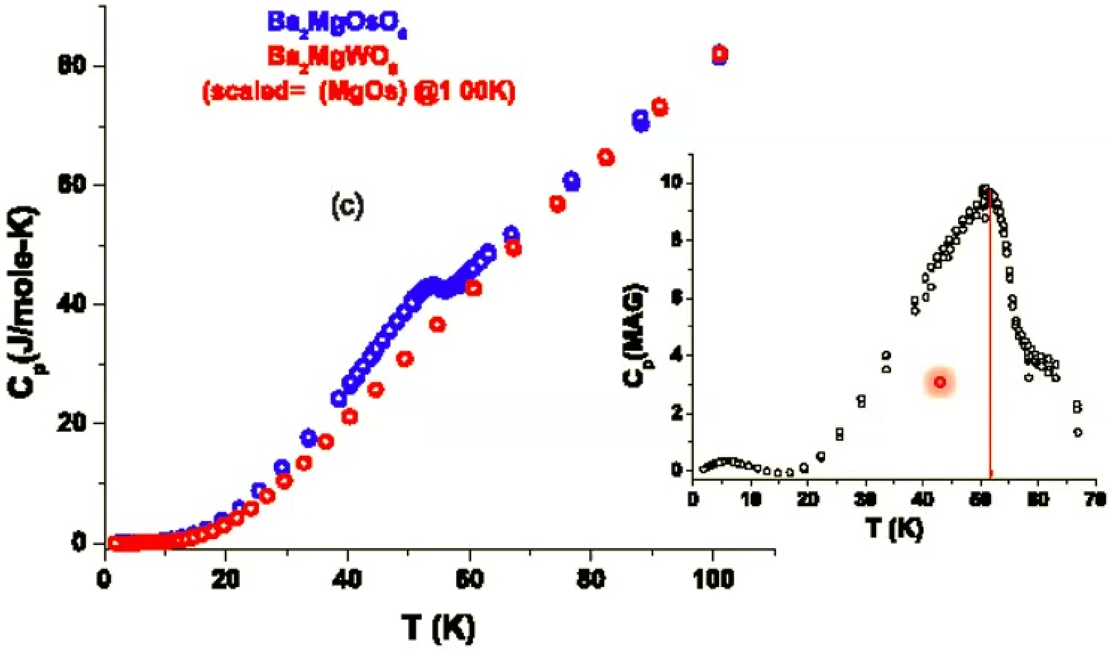


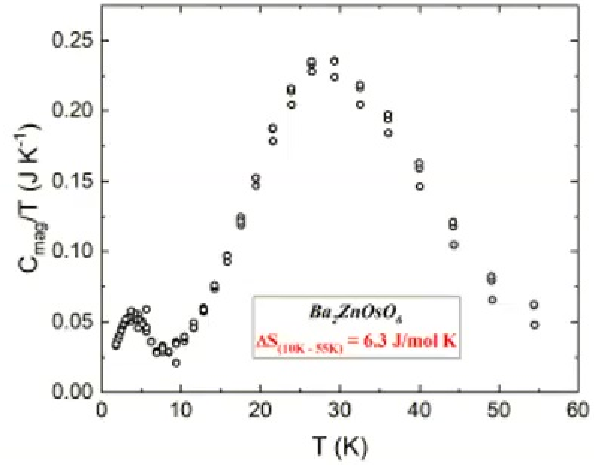
Ba_2MgOsO_6
Ba_2CaOsO_6
Ba_2ZnOsO_6

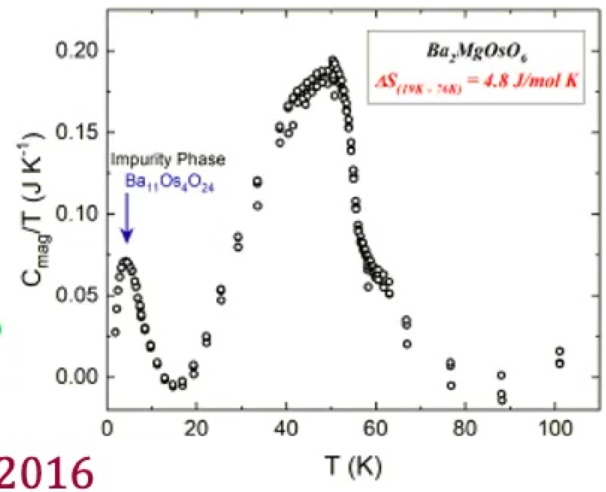
J=2?

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

Thermodynamics: Specific heat and entropy



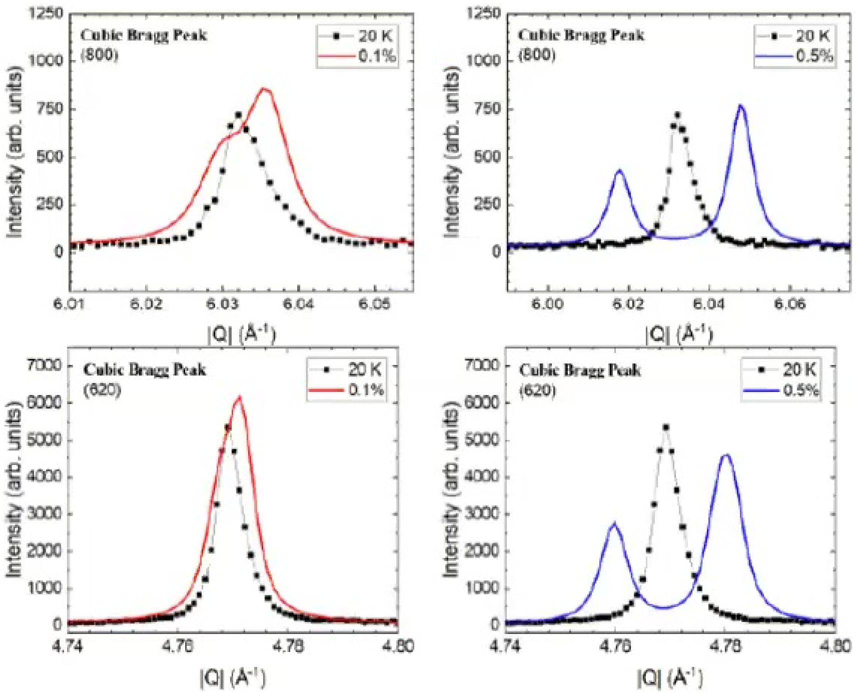




- Phase transition at T ~ 30-50K
- Entropy recovered << R ln (5): **Not J=2?**
- Rln(2) ~ 5.8 J/mol-K; Rln(3) ~ 9.1 J/mol-K

C. A. Marjerrison, et al, PRB 2016

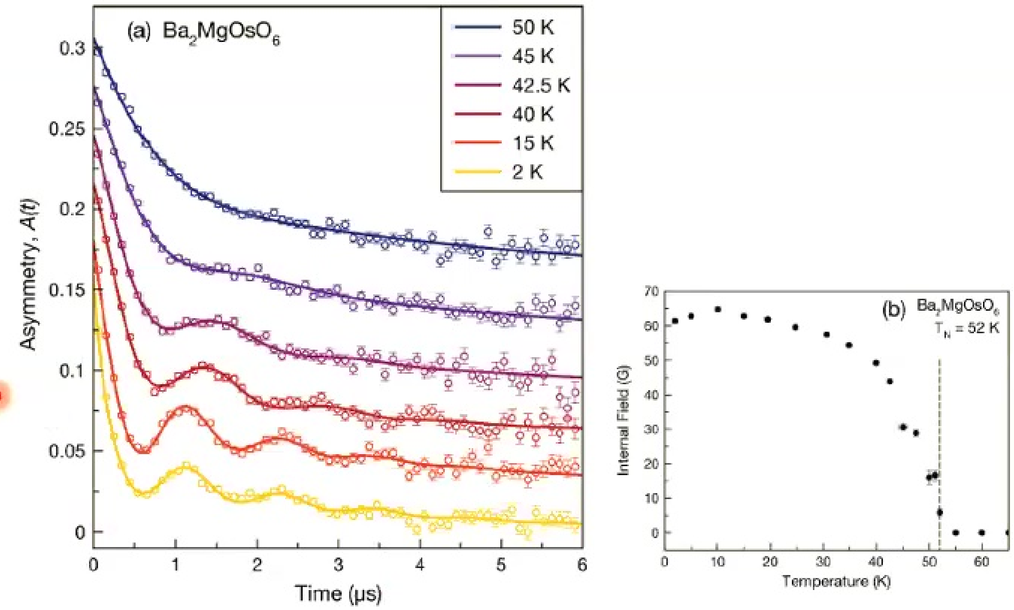
Xray diffraction



Cubic Fm$\bar{3}$m
(<0.1% distortion)

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

μSR



Signature of time-reversal
symmetry breaking

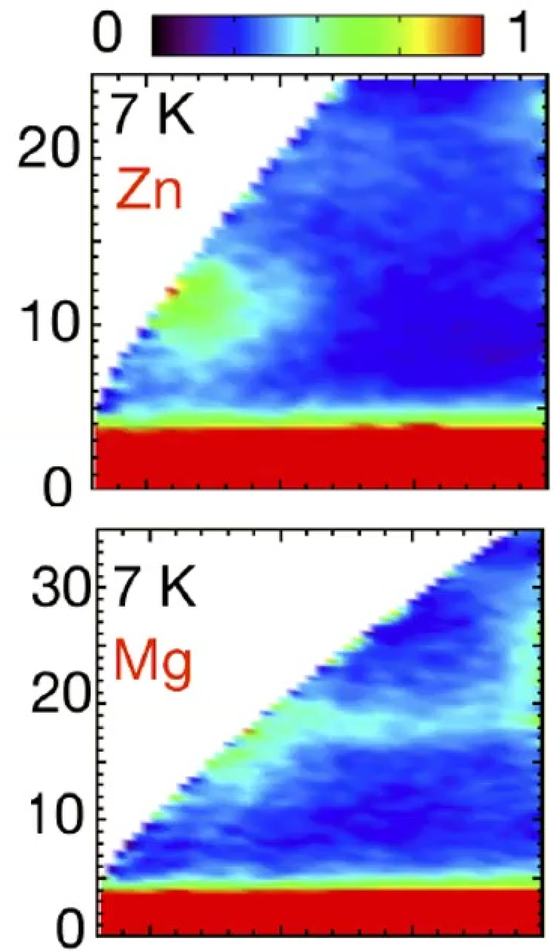
C. M. Thompson, et al, JPCM (2014)

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)



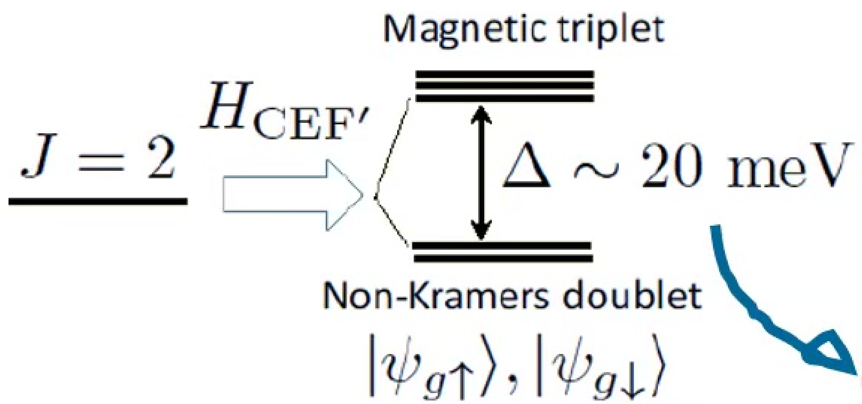
Puzzles

- ❑ 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 μSR
- ❑ 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



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

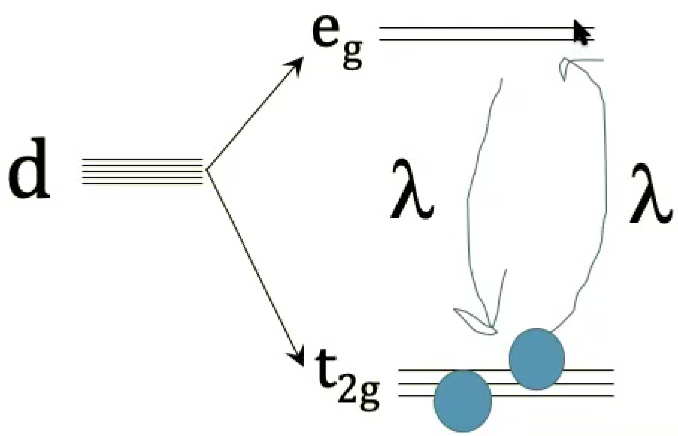
$$|\psi_{g,\downarrow}\rangle = \frac{1}{\sqrt{2}}(|2\rangle + |-2\rangle)$$

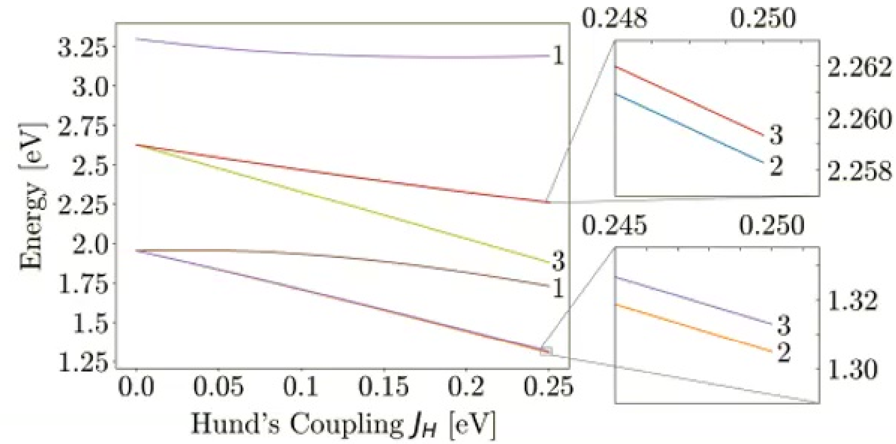
- ✓ Spin gap
- ✓ No dipole order
- ✓ Entropy: R ln(2)

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

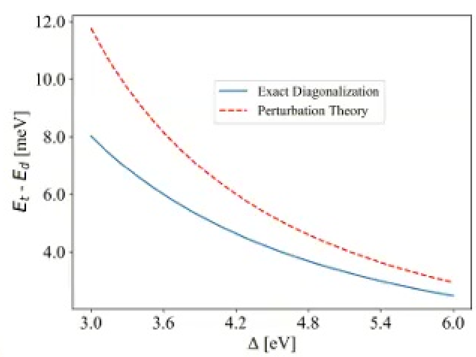
Cannot blindly drop e_g orbitals - crucial for low energy physics!

The correct "atomic limit"





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



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

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

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

Pseudospin components

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

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

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

"XY" Quadrupolar charge density

Ising magnetic octupole

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

<u>See also:</u>

4f Pr compounds: non-Kramers Γ_3 doublet

PrM_2Al_{20} Sakai, et al, JPSJ 2012; Taniguchi, et al, JPSJ 2016; Kittaka et al, JPSJ2020; …
Hattori, Tsunetsugu, JPSJ 2014; Tsunetsugu, et al, JPSJ 2021;
Freyer, et al, PRB 2018; S. B. Lee, et al, PRB 2018; Freyer et al, PRR 2018; Patri et al, Nat Comm 2019

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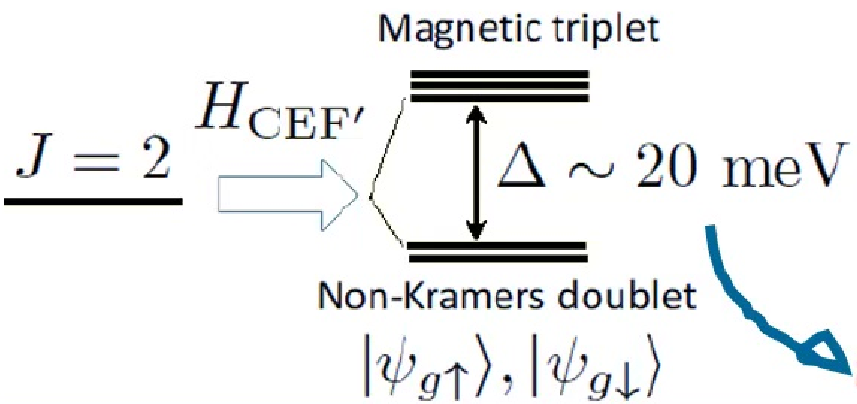

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- ✓ No dipole order
- ✓ Entropy: R ln(2)

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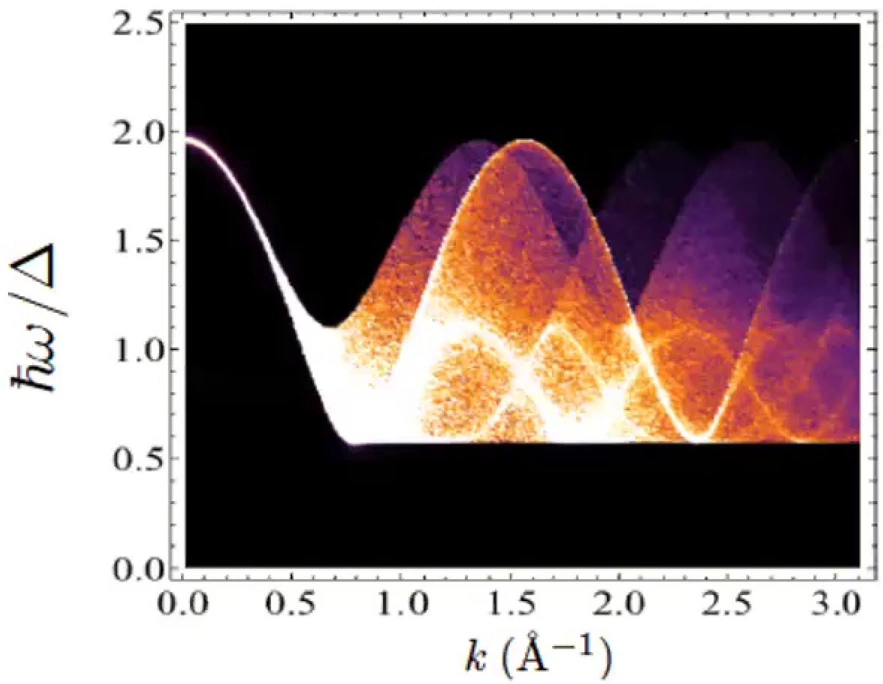
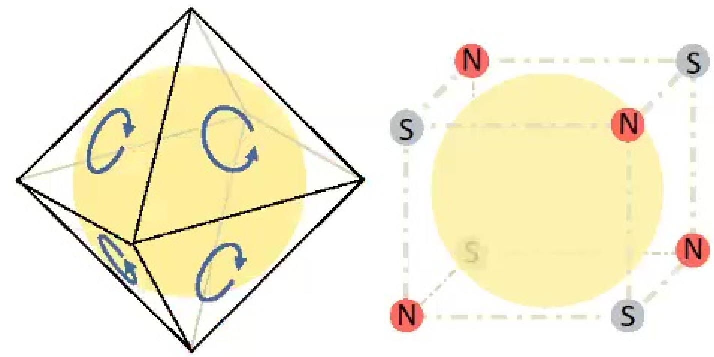
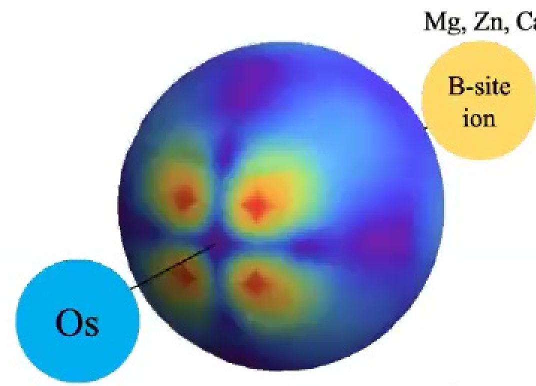
Freyer, et al, PRB 2018; S. B. Lee, et al, PRB 2018; Freyer et al, PRR 2018; Patri et al, Nat Comm 2019

Ferro-octupolar order: Neutron Scattering, μSR



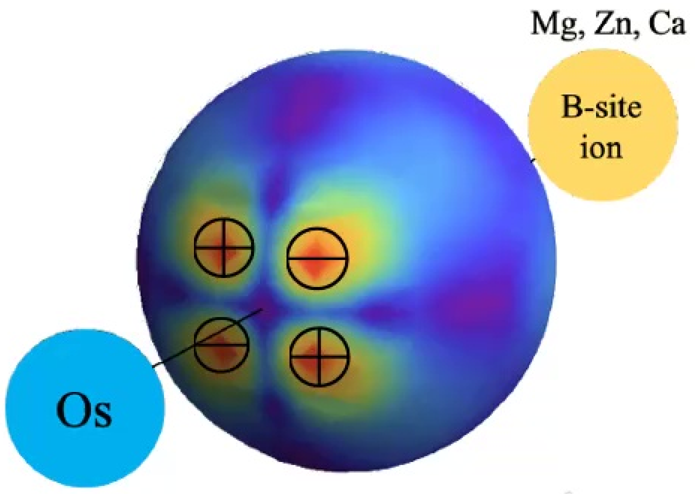




✓ Spin-gapped magnetic exciton

✓ Loop current order, B ~ 30G

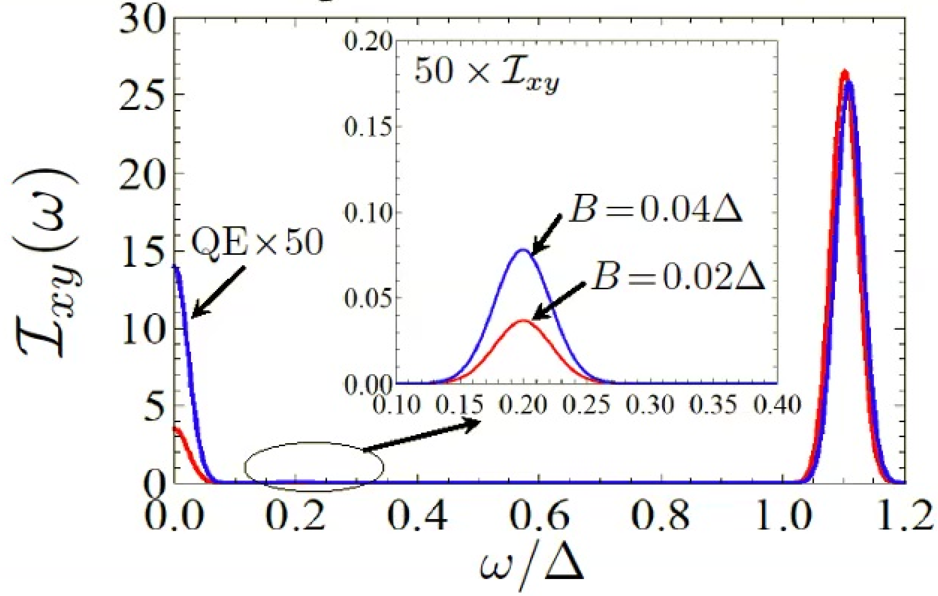
Predictions for NMR, Raman scattering



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

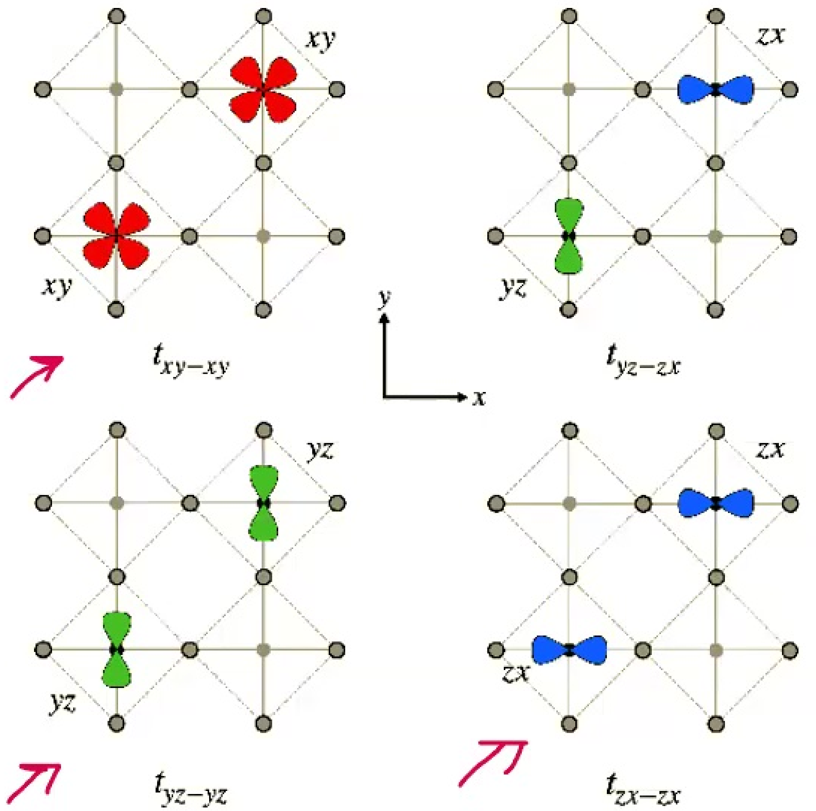
AP, D. D. Maharaj, Bruce Gaulin, PRB 2020
S. Voleti D. D. Maharaj, B. Gaulin, G. Luke, **AP**, PRB 2020

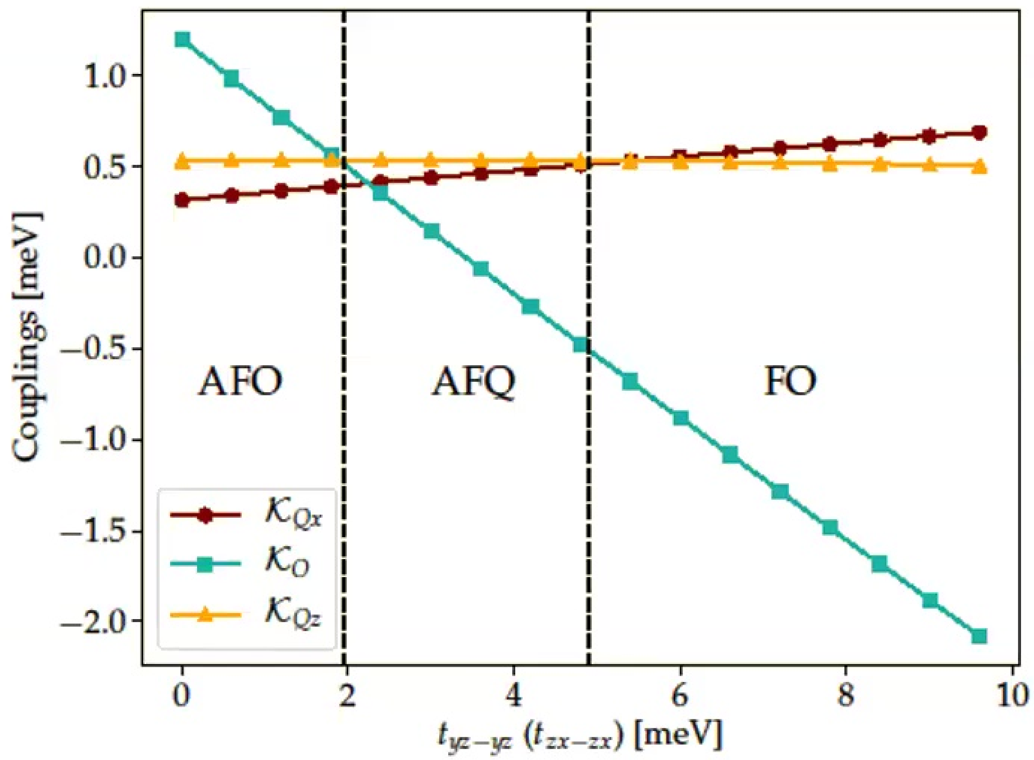
B_{2g} Raman scattering



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

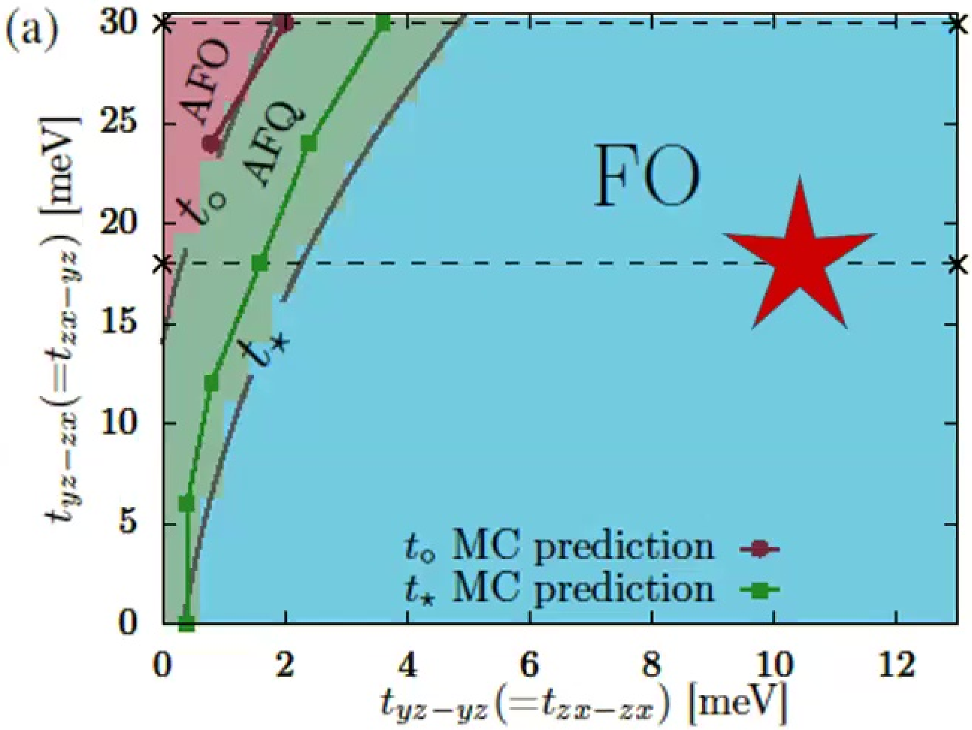
Two-site computation of exchange couplings

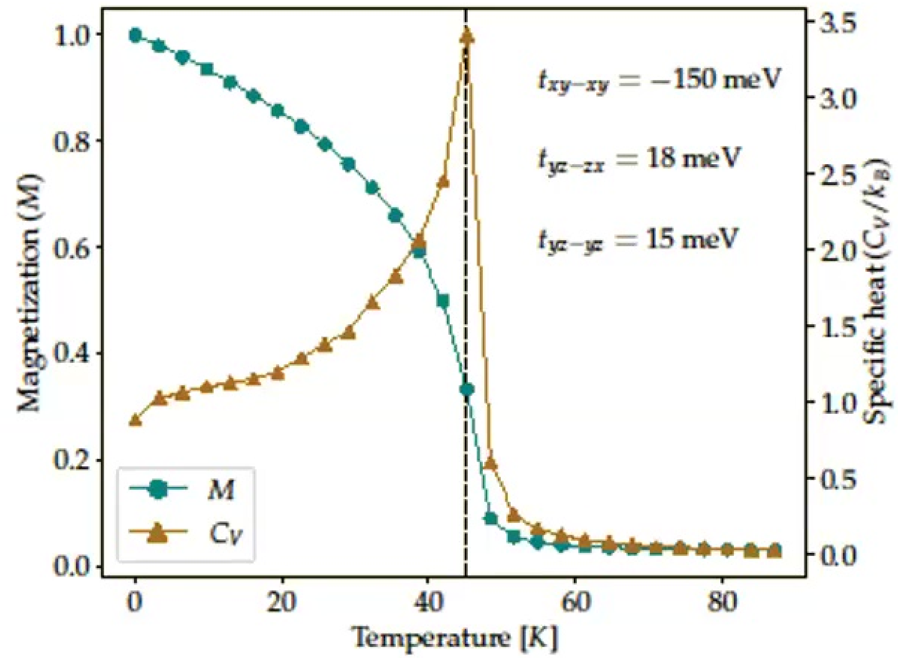




Sreekar Voleti, Arijit Halder, AP, Phys. Rev. B **104**, 174431 (2021)

Monte Carlo Phase diagram





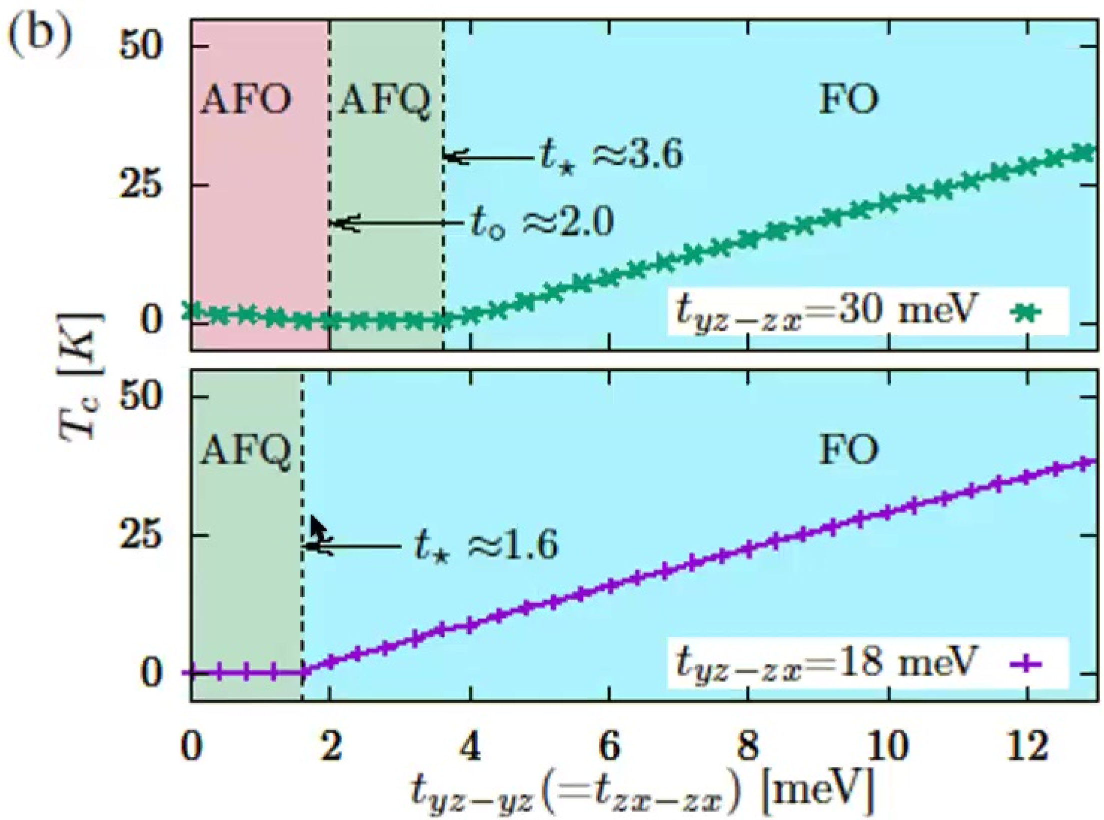
Sreekar Voleti, Arijit Halder, AP (arXiv: 2109.13266)

Agree: LDA+DMFT Pourovskii, Mosca, Franchini, arXiv: 2107.04493 (PRL, to appear)

Disagree: Lovesey, Khalyavin, PRB 102, 064407 (2020); Khaliullin, Churchill, Stavropoulos, Kee, PRR 3, 033163 (2021)

Fix: Churchill, Kee (arXiv:2109.08104)?

Monte Carlo Phase diagram

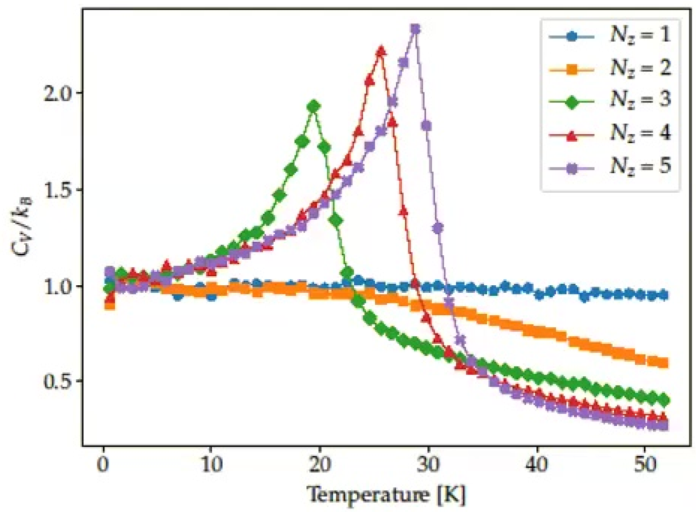


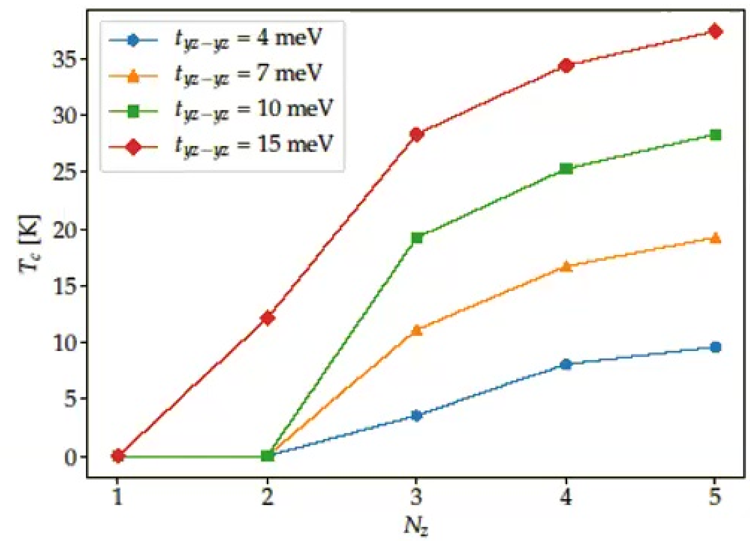
Sreekar Voleti, Arijit Halder, AP, Phys. Rev. B **104**, 174431 (2021)

Tuning octupolar quantum criticality
Impact of surfaces / thin films

Surfaces strongly break cubic symmetry – explore **thin films** or **surface QCPs**

Leads to large "field" on the τ_Z component = ***transverse field for octupolar order***





Sreekar Voleti, Arijit Halder, AP, Phys. Rev. B **104**, 174431 (2021)

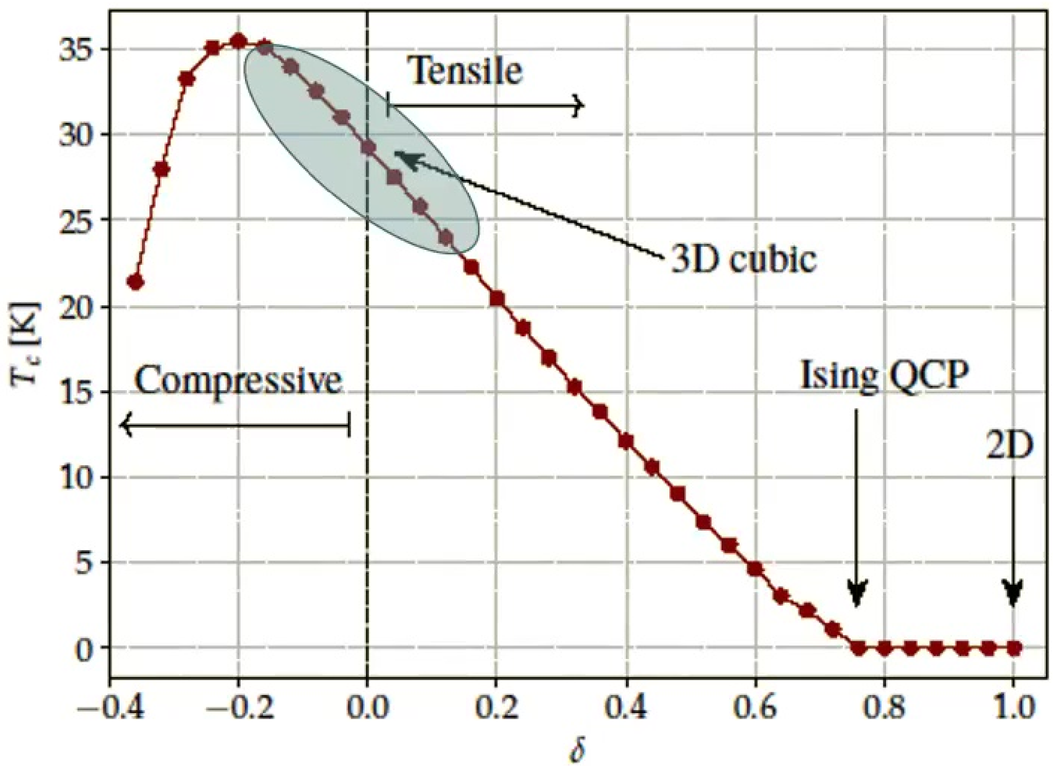
Cf: Quadrupoles -- Transverse fields via strain for tuning nematic criticality, A. Maharaj, et al, PNAS (2017)

Tuning octupolar quantum criticality
Impact of strain?

Let us scale the couplings involving z-direction by (1-δ)

Uniaxial strain / 2D limit

Strain also leads to "transverse field" for octupolar Ising order



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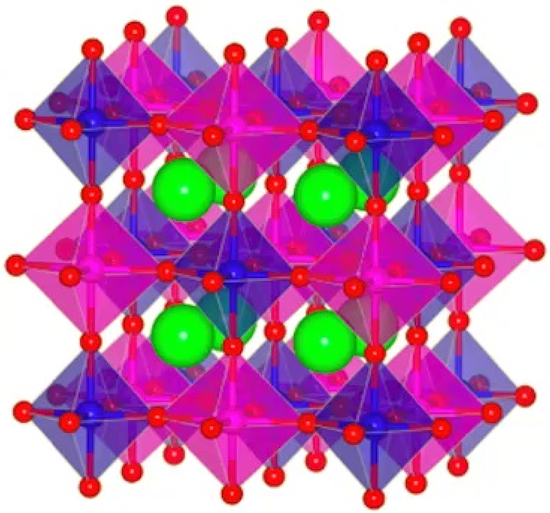
Summary

❖ Ba_2MgOsO_6
❖ Ba_2CaOsO_6
❖ Ba_2ZnOsO_6

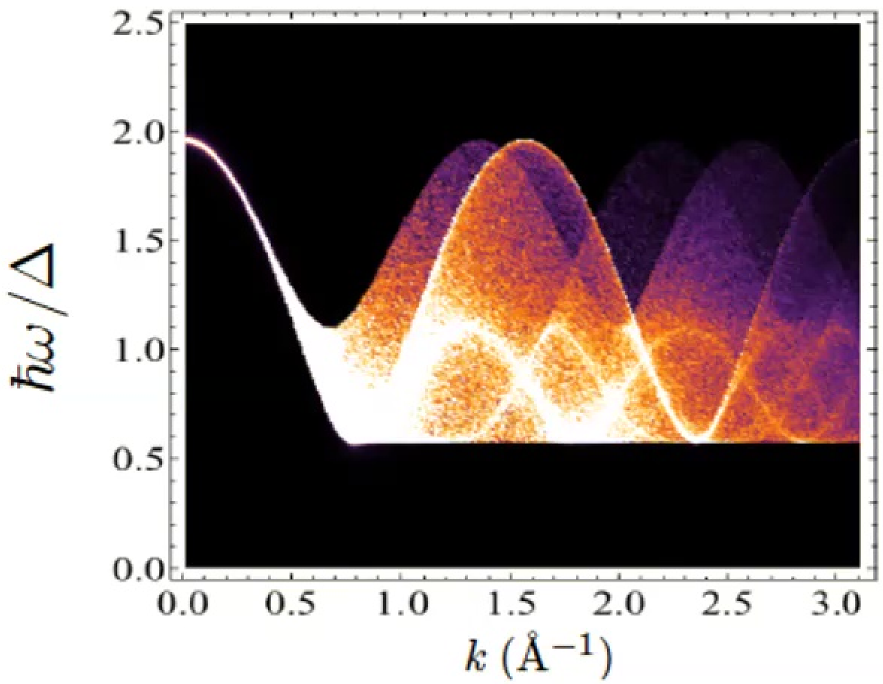


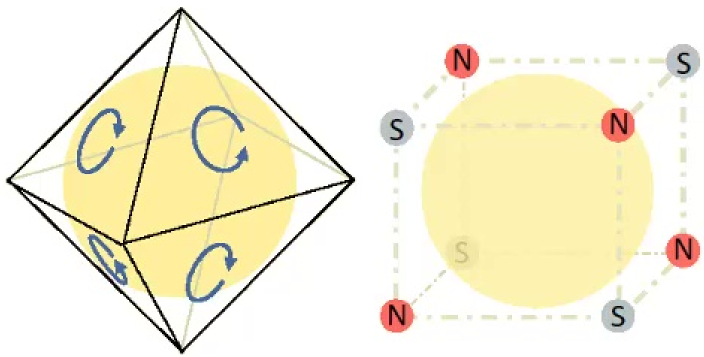
Discovery of d-orbital octupolar order

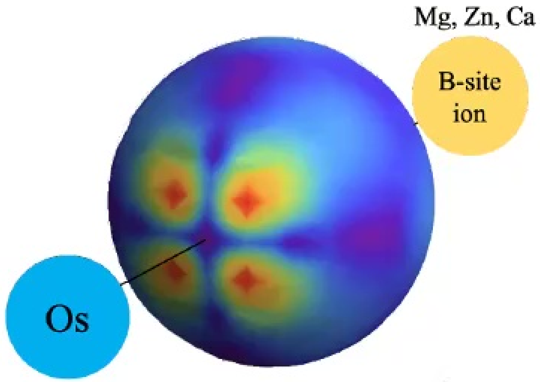
✓ Entropy recovered across T* ~ R ln(2)
✓ Remains cubic down to T << T*
✓ Time reversal breaking via μSR
✓ No sign of magnetic Bragg peaks
✓ Spin gap in neutron scattering ~ 10 meV
✓ Microscopic theory

❖ Predictions for Raman scattering / NMR
❖ Tuning 2d/3d Ising quantum criticality

Ferro-octupolar order: Neutron Scattering, μSR







✓ Spin-gapped magnetic exciton

✓ Loop current order, B ~ 30G