SPIN-ORBIT INTERACTION IN FRUSTRATED MAGNETIC SYSTEMS:
APPLICATION TO DOUBLE PEROVSKITES

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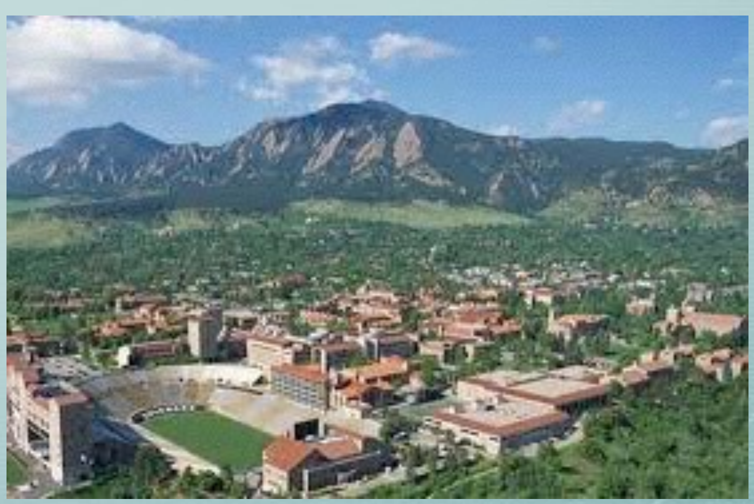


Collaborators

* **Rodrigo Pereira** (University of Sao Paulo, Brazil) for an early work
* **Leon Balents** (KITP, UCSB)

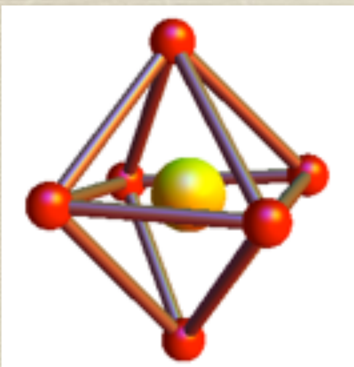
Chen, Pereira, Balents, PRB 2010
Chen, Balents (in preparation)

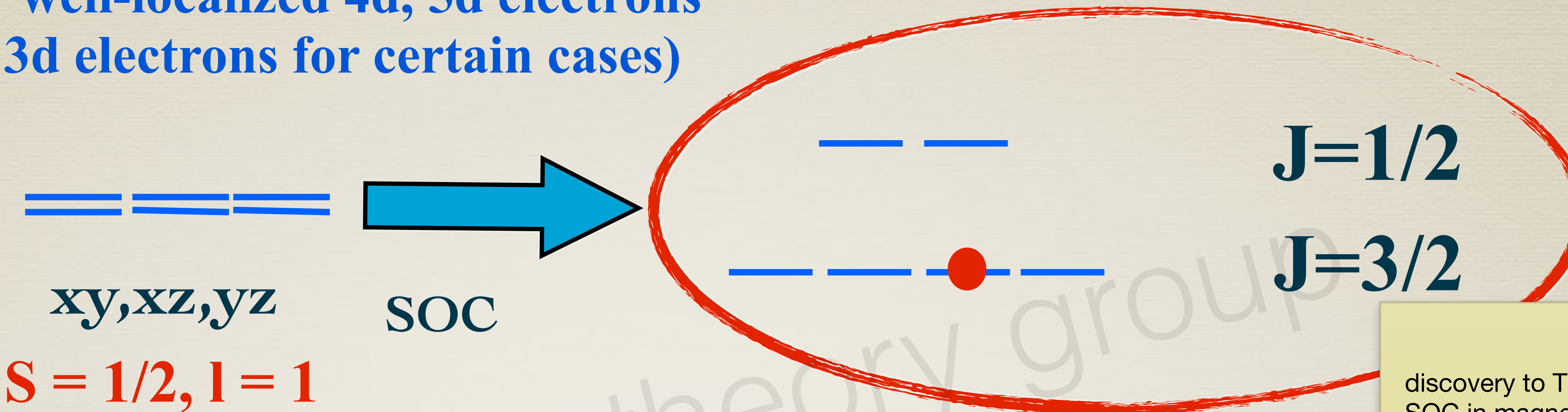
OUTLINE

* Introduction: Spin-orbit coupling and ordered double perovskites
* Ground state phases
* Finite temperature phases
* Experiments and discussion

Motivation: Spin-orbit interaction on degenerate **t2g** manifold

Consider well-localized 4d, 5d electrons (maybe 3d electrons for certain cases)



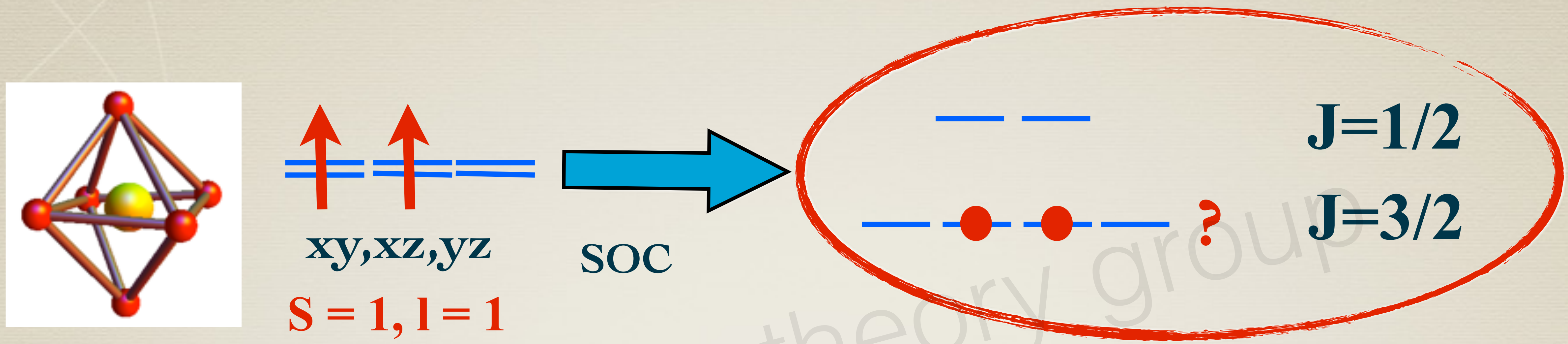


$$H_{SOC} = -\lambda \mathbf{l} \cdot \mathbf{S}$$

electron # per site	Local spin state	some compounds	Talks
1	J=3/2	next few sides	this talk

discovery to TI,
SOC in magnetic system
the systems described are systems with heavy magnetic ions, 4d or 5d. Consider an octahedron

Spin-orbit interaction on degenerate **t2g** manifold



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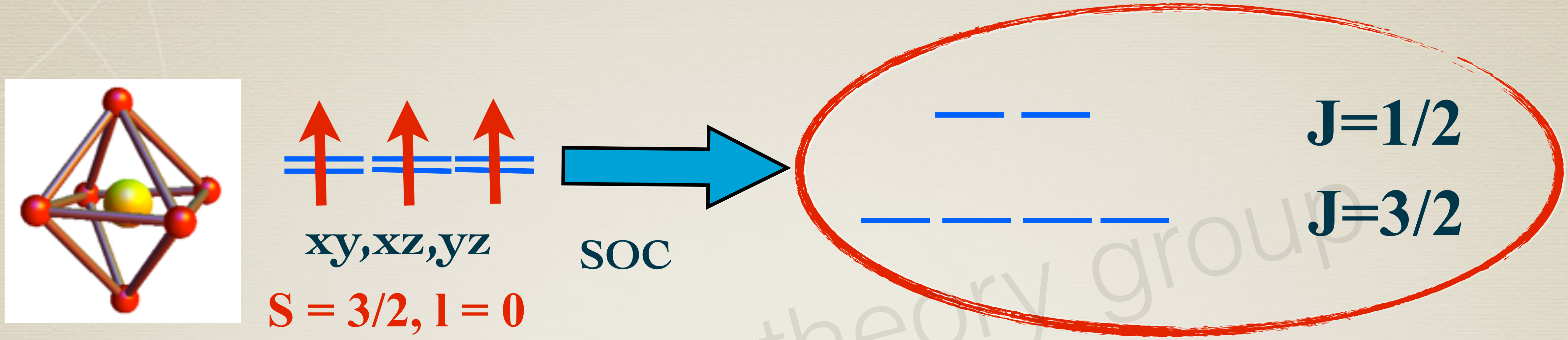
Spin-orbit interaction on degenerate **t2g** manifold



$$H_{SOC} = -\lambda \mathbf{l} \cdot \mathbf{S}$$

electron # per site	Local spin state	some compounds	Talks of the conference
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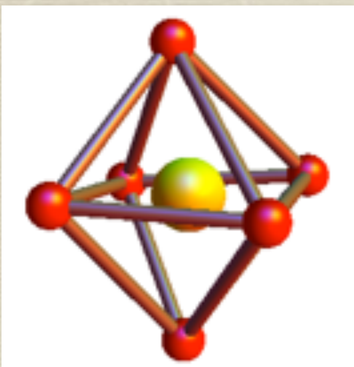
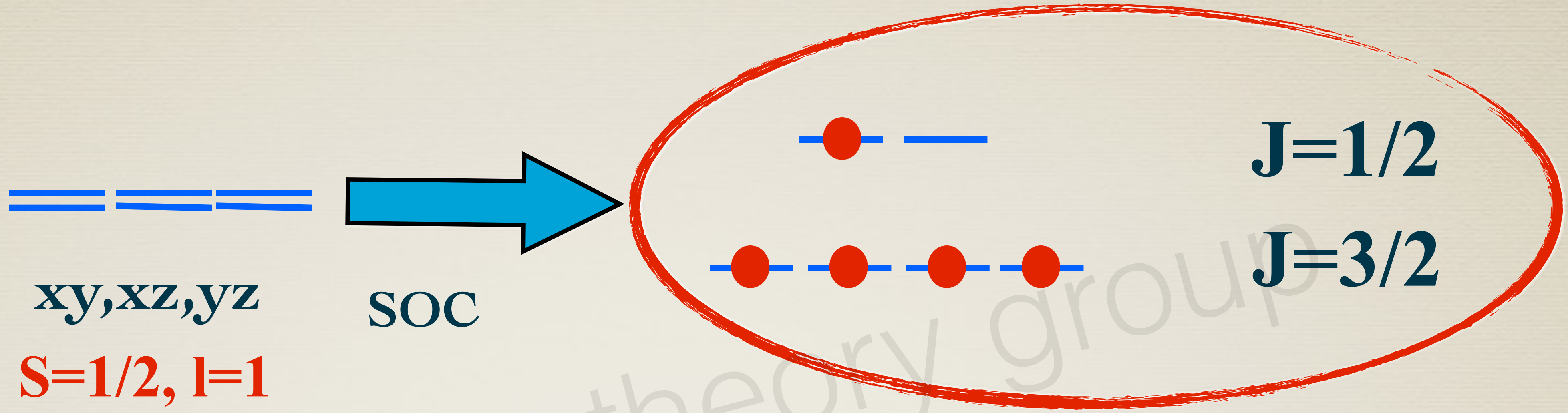
3	S=3/2	SCGO, $ZnCr_2O_4$, etc	Nenert
4	J=0		

Spin-orbit interaction on degenerate **t2g** manifold





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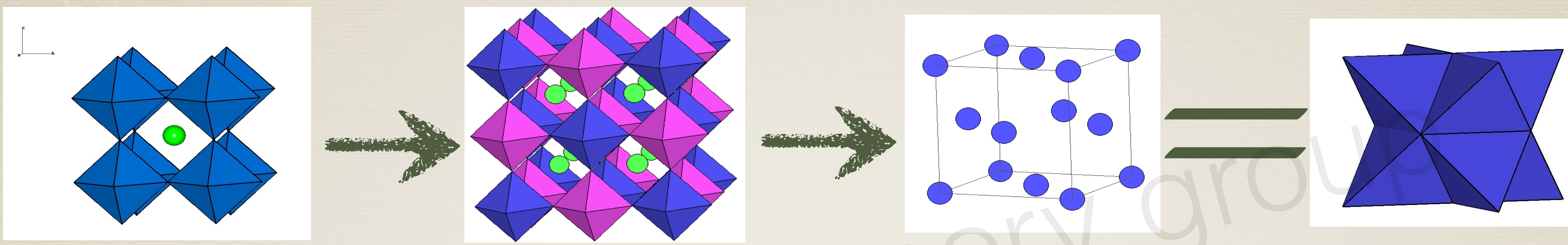
5	J=1/2	Iridates	Takayama, Jiang

Ordered double perovskites

FCC ordered double perovskites $A_2BB'O_6$



Interplay between geometrical frustration and strong SOC

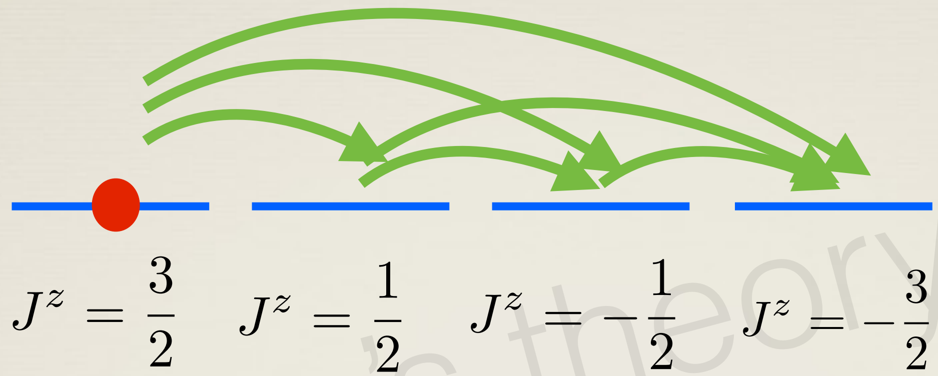
Compound	B' config.	crystal structure	θ_{CW}	$\mu_{\mathrm{eff}}(\mu_B)$	magnetic transition	frustration parameter f
Ba_2YMoO_6	$Mo^{5+}(4d^1)$	cubic	-91K	1.34	PM down to 2K	$f \gtrsim 45$
Ba_2YMoO_6	$Mo^{5+}(4d^1)$	cubic	-160K	1.40	PM down to 2K	$f \gtrsim 80$
Ba_2YMoO_6	$Mo^{5+}(4d^1)$	cubic	-219K	1.72	PM down to 2K	$f \gtrsim 100$
La_2LiMoO_6	Mo^{5+} (4d^1)	monoclinic	-45K	1.42	PM to 2K	$f \gtrsim 20$
Sr_2MgReO_6	$Re^{6+}(5d^1)$	tetragonal	-426K	1.72	spin glass, $T_G \sim 50$K	$f \gtrsim 8$
Sr_2CaReO_6	$Re^{6+}(5d^1)$	monoclinic	-443K	1.659	spin glass, $T_G \sim 14$K	$f \gtrsim 30$
Ba_2CaReO_6	$Re^{6+}(5d^1)$	cubic to tetragonal (at $T \sim 120$K)	-38.8K	0.744	AFM $T_c = 15.4$K	$f \sim 2$
Ba_2LiOsO_6	$Os^{7+}(5d^1)$	cubic	-40.48K	0.733	AFM $T_c \sim 8$K	$f \gtrsim 5$
Ba_2NaOsO_6	$Os^{7+}(5d^1)$	cubic	-32.45K	0.677	FM $T_c \sim 8$K	$f \gtrsim 4$
Ba_2NaOsO_6	$Os^{7+}(5d^1)$	cubic	~ -10K	~ 0.6	FM $T_c = 6.8$K	$f \gtrsim 4$

One electron per site: Chen, Pereira and Balents,PRB 2010

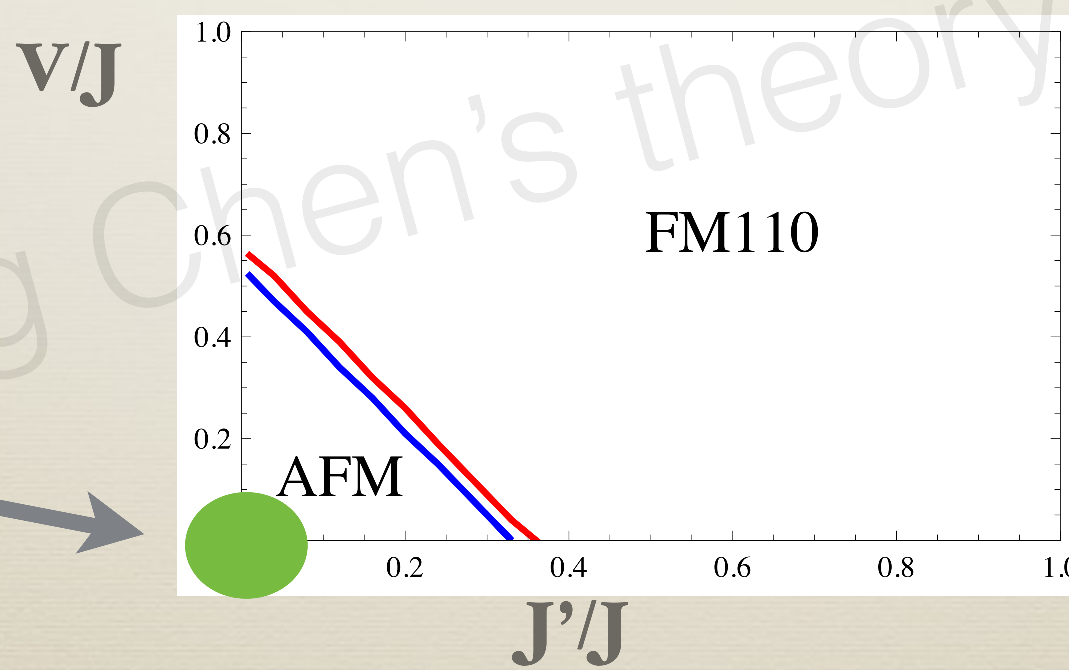
Erickson, et al PRL 2007
M. A. de Vries, et al PRL (2010).
T. Aharen, et al PRB (2010).
E. J. Cussen, et al, Chem. Mater(2006).
C. R.Wiebe,et al PRB (2003).
C. Wiebe, et al PRB (2002).
K. Yamamura,et al JSSC (2006).

Predictions for 1 electron case: J=3/2

$$H \sim \mathbf{J}_i \cdot \mathbf{J}_j + J_i^2 J_j^2 + J_i^3 J_j^3 + J_i J_j^3 + J_i^3 J_j$$



Enhanced quantum fluctuation!



Possible QSL

Might be relevant for
Ba_2YMoO_6

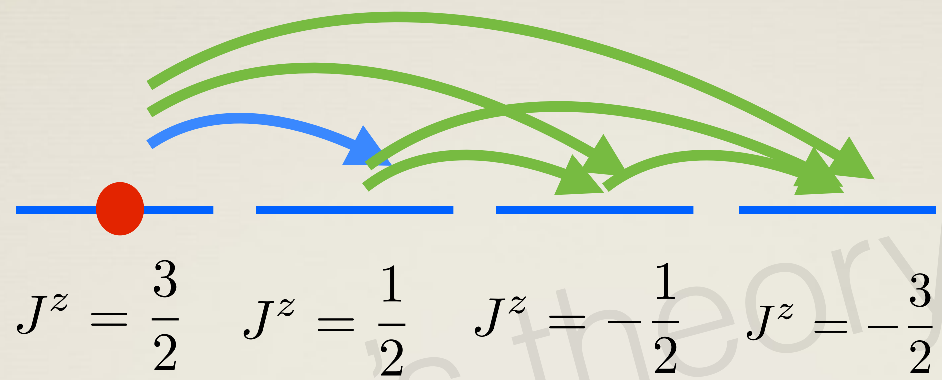
J is AFM exchange, J' is FM exchange, V is electric quadrupolar interaction

Chen, et al, PRB 2010

Predictions for 1 electron case: J=3/2

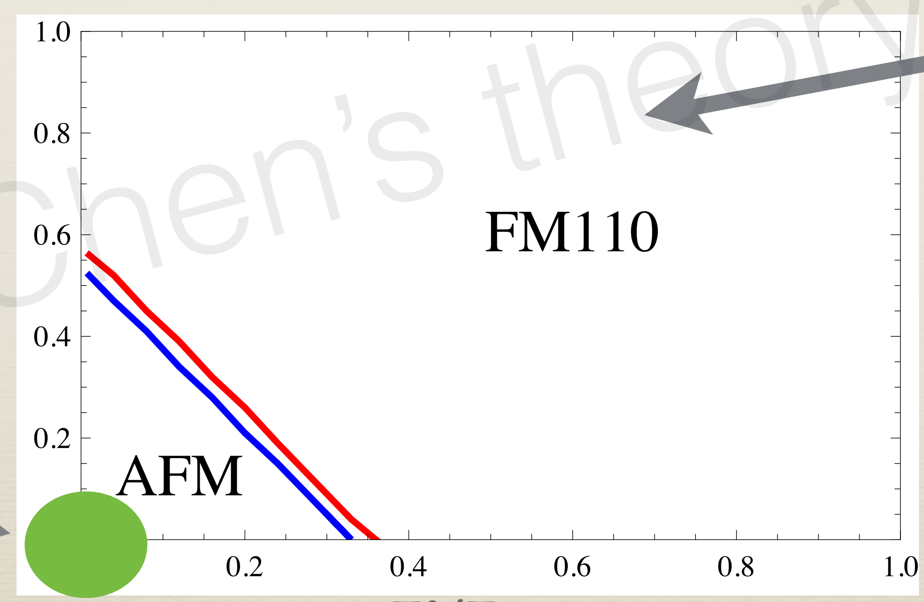
$$H \sim \mathbf{J}_i \cdot \mathbf{J}_j + J_i^2 J_j^2 + J_i^3 J_j^3 + J_i J_j^3 + J_i^3 J_j$$

non-heisenberg like hamiltonian;
biquadratic, bicubic, magnetic quadruopole, octupole moment
enhanced quantum fluctuation;
linear spin wave theory



Enhanced quantum fluctuation!

V/J



J'/J

Magnetic multipole order:
Octupole, Quadrupole

$$\langle J_i^\mu J_i^\nu J_i^\rho \rangle, \quad \langle J_i^\mu J_i^\nu \rangle$$

FM state is observed in
Ba2NaOsO6

Possible QSL

Might be relevant for
Ba2YMoO6

J is AFM exchange, J' is FM exchange, V is electric quadrupolar interaction

Chen, et al, PRB 2010
Carlo, et al, ArXiv 1105.3457
Aharen, et al, PRB 2010
M. A. de Vries, et al PRL (2010)
Erickson, et al PRL 2007

2 electron case: **J=2**

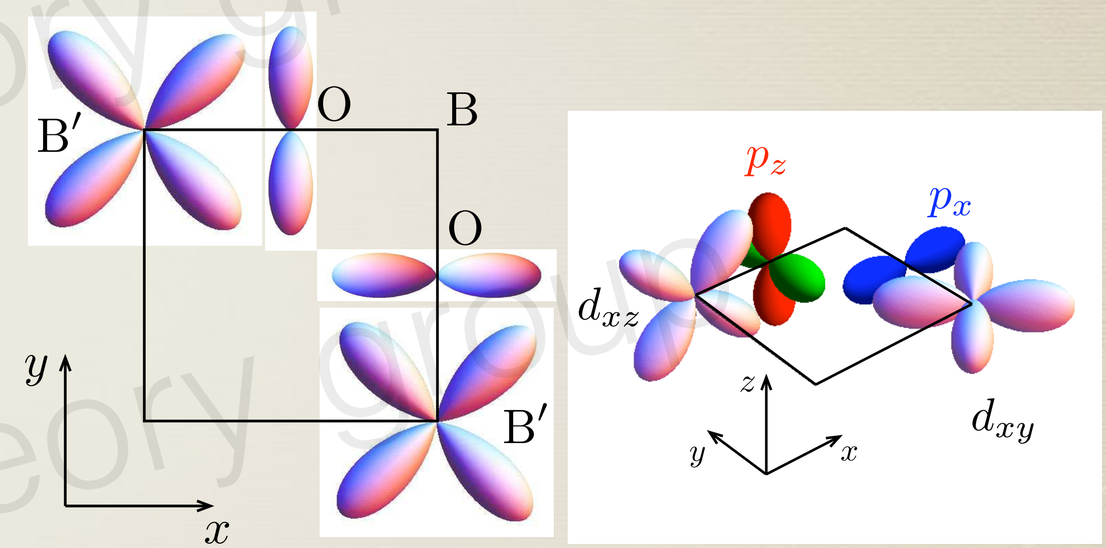
Re^{5+}, Os^{6+} in Ba_2CaOsO_6, La_2LiReO_6, Ba_2YReO_6

Exchange interaction

$$\mathcal{H}^{\rm XY}_{\text{ex-1}} = J\sum_{\langle ij\rangle}\Big(\boldsymbol{S}_{i,xy}\cdot\boldsymbol{S}_{j,xy} - \frac{1}{4}\,n_{i,xy}n_{j,xy}\Big)$$

$$\mathcal{H}^{\rm XY}_{\text{ex-2}} = -J'\sum_{\langle ij\rangle}\Big[\boldsymbol{S}_{i,xy}\cdot(\boldsymbol{S}_{j,yz}+\boldsymbol{S}_{j,xz}) + \frac{3}{4}\,n_{i,xy}(n_{j,yz}+n_{j,xz}) + \langle i\leftrightarrow j\rangle\Big]$$

AFM and FM exchange path



$S_{i,xy}$ and $n_{i,xy}$ denote the electron spin residing on xy orbital and orbital occupation number for single electron xy orbital at site i, respectively.

Electric quadrupolar interaction: non-trivial after projection

$$\mathcal{H}^{\rm XY}_{\rm quad} = \sum_{\langle ij\rangle\in XY}\Big[-\frac{4V}{3}(n_{i,xz}-n_{i,yz})(n_{j,xz}-n_{j,yz}) + \frac{9V}{4}n_{i,xy}n_{j,xy}\Big]$$

Experiments: Aharen, et al PRB 81,064436, (2010)
Yamamura, et al JSSC 179 (2006) 605–612

2 electron case: J=2

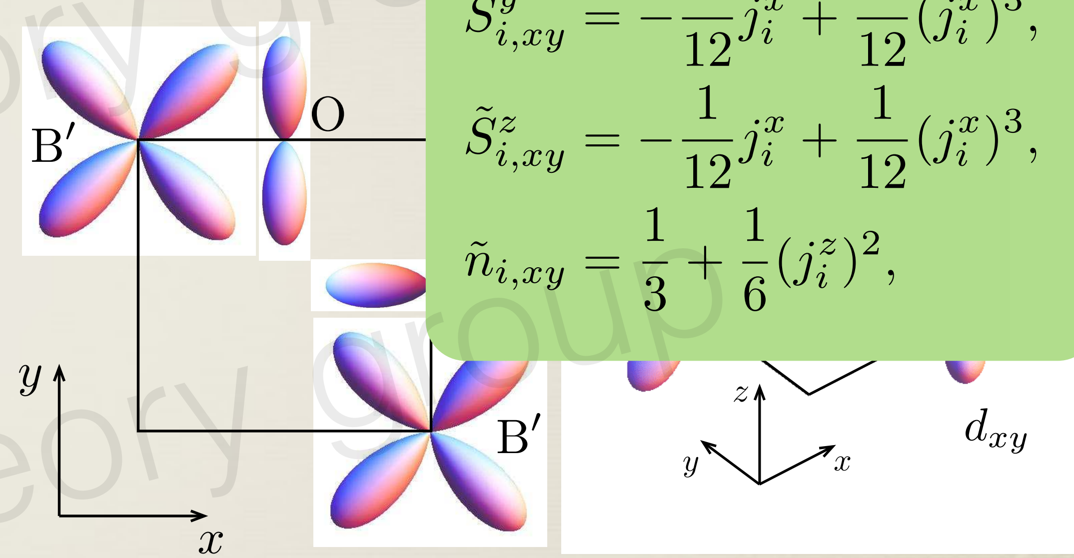
Re^{5+}, Os^{6+} in Ba_2CaOsO_6, La_2LiReO_6, Ba_2YReO_6

Projection to J=2

$$\tilde{S}^x_{i,xy} = -\frac{1}{12} j^x_i + \frac{1}{12}(j^x_i)^3,$$
$$\tilde{S}^y_{i,xy} = -\frac{1}{12} j^x_i + \frac{1}{12}(j^x_i)^3,$$
$$\tilde{S}^z_{i,xy} = -\frac{1}{12} j^x_i + \frac{1}{12}(j^x_i)^3,$$
$$\tilde{n}_{i,xy} = \frac{1}{3} + \frac{1}{6}(j^z_i)^2,$$

Exchange interaction

AFM and



$$\mathcal{H}^{\mathrm{XY}}_{\text{ex-1}} = J \sum_{\langle ij \rangle} \left(\boldsymbol{S}_{i,xy} \cdot \boldsymbol{S}_{j,xy} - \frac{1}{4}\, n_{i,xy} n_{j,xy} \right)$$

$$\mathcal{H}^{\mathrm{XY}}_{\text{ex-2}} = -J' \sum_{\langle ij \rangle} \Big[\boldsymbol{S}_{i,xy} \cdot (\boldsymbol{S}_{j,yz} + \boldsymbol{S}_{j,xz}) + \frac{3}{4}\, n_{i,xy}(n_{j,yz} + n_{j,xz}) + \langle i \leftrightarrow j \rangle \Big]$$

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Frustration comes from anisotropic nature of orbital orientation.

Experiments: Aharen, et al PRB 81,064436, (2010)
Yamamura, et al JSSC 179 (2006) 605–612

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$$\tilde{n}_{i,xy} = \frac{1}{3} + \frac{1}{6}(j^z_i)^2,$$

Exchange interaction

$$\mathcal{H}^{\rm XY}_{\text{ex-1}} = J \sum_{\langle ij \rangle} \Big(\boldsymbol{S}_{i,xy} \cdot \boldsymbol{S}_{j,xy} - \frac{1}{4} n_{i,xy} n_{j,xy} \Big)$$
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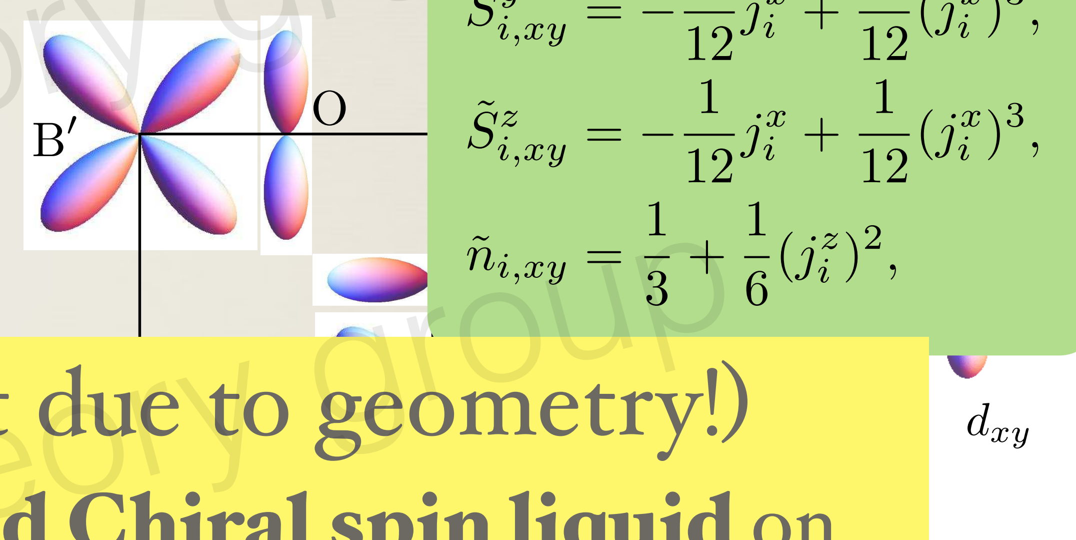
AFM and



Intrinsic Frustration (not due to geometry!)

e.g. **SU(N) Heisenberg model and Chiral spin liquid** on square lattice and spin ladders (see **my poster**)

occupation number for single electron xy orbital at site i, respectively.

Electric quadrupolar interaction: non-trivial after projection

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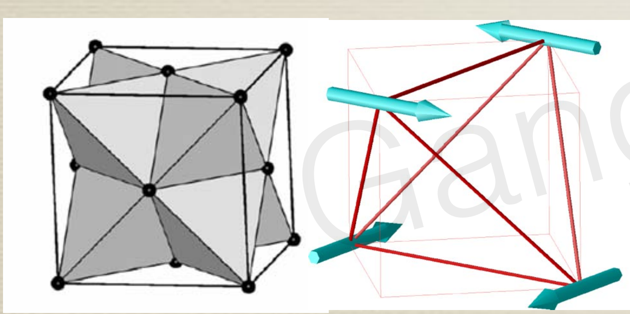
Yamamura, et al JSSC 179 (2006) 605–612

Ground state phase diagram: J=2

- Uniform state: FM111
- 2-sublattice state(P=2Pi[001]): AFM100, FM110, "$*$" phase, Quadrupole phase
- Four-sublattice AFM state: Δ



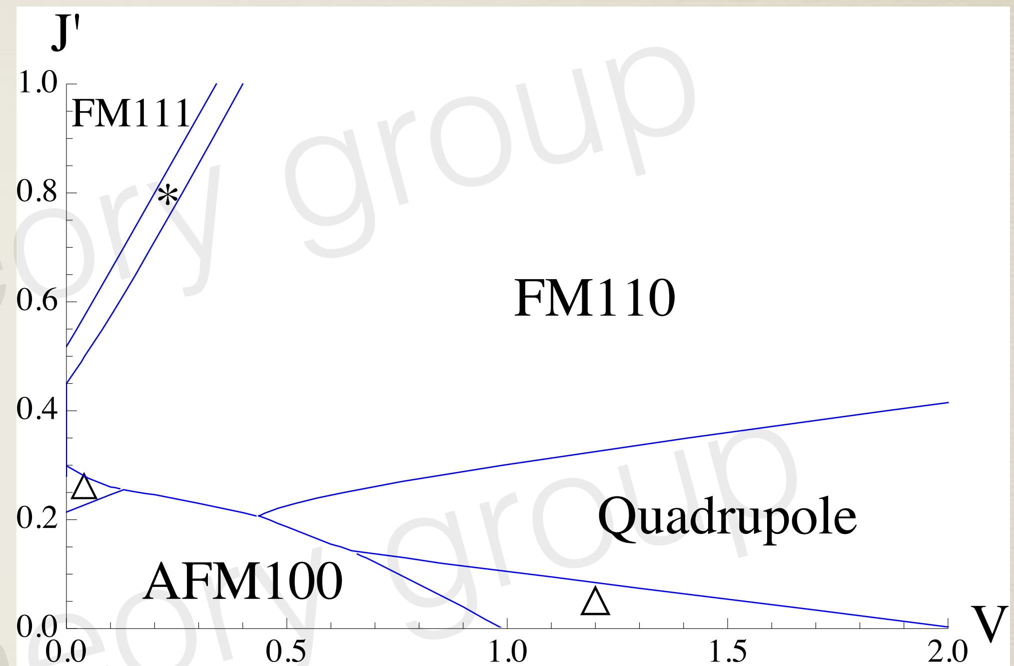


FIG. 1. (Color online) Ground state phase diagram. "Δ" phase is a four-sublattice AFM phase; "$*$" phase is an intermediate ferromagnetic phase between FM110 and FM111 phase. $J = 1$ in the phase diagram.

orbital-orbital interaction favors a non-collinear spin structure

These variational states are found to be stable by linear flavor wave theory.

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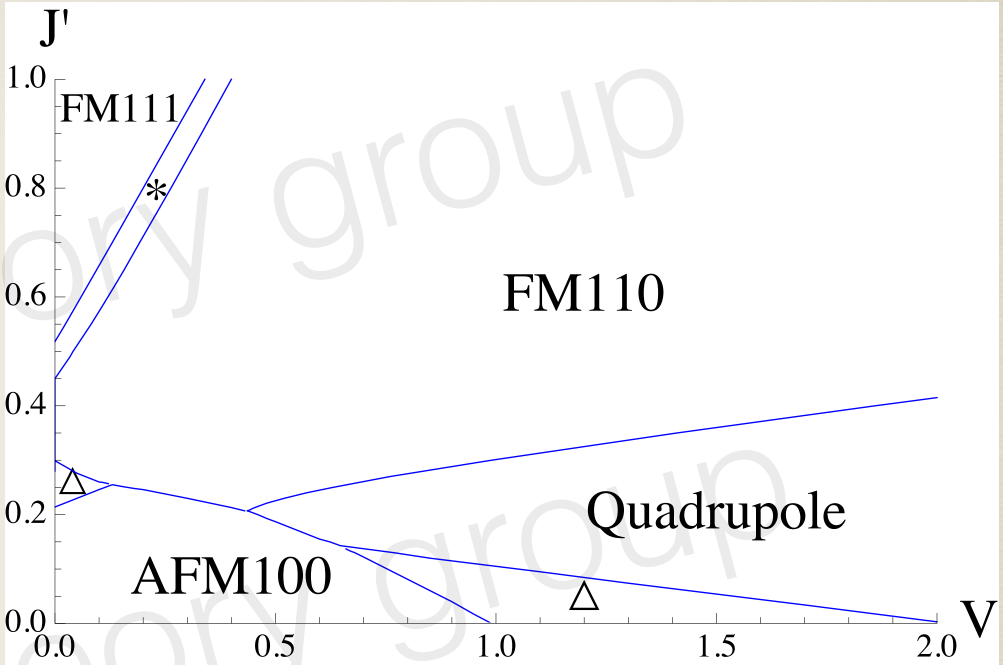

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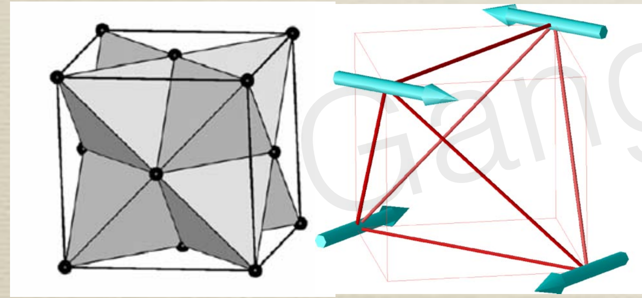

e.g. the wavefunction of A/B sublattice of the Quadrupole phase is

$$|A\rangle = \frac{1}{2}|j^z = 2\rangle + \frac{1}{\sqrt{2}}|j^z = 0\rangle + \frac{1}{2}|j^z = -2\rangle$$

$$|B\rangle = \frac{1}{2}|j^z = 2\rangle - \frac{1}{\sqrt{2}}|j^z = 0\rangle + \frac{1}{2}|j^z = -2\rangle$$

Preserve time reversal symmetry, no magnetic dipolar order but a **spin nematic order**!

Magnetic multipole order: dipole, quadrupole, octupole!

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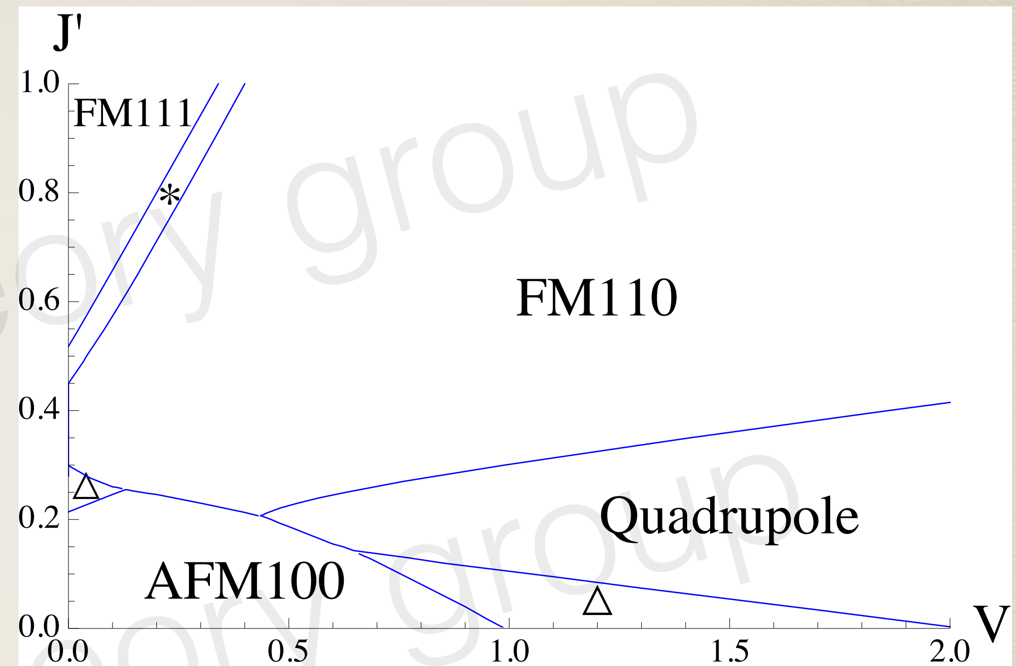

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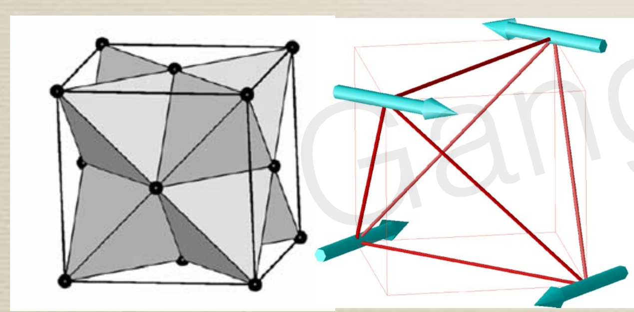

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Preserve time reversal symmetry, no magnetic dipolar order but a **spin nematic order**!

Also seen in $NiGa_2S_4$!

Magnetic multipole order: dipole, quadrupole, octupole!

Gang Chen’s theory group

Finite temperature phases

* Numerical calculation from MFT suggests that there exists a **spin-nematic phase** (which preserves time reversal symmetry) in the intermediate-temperature for some region of the phase diagram.

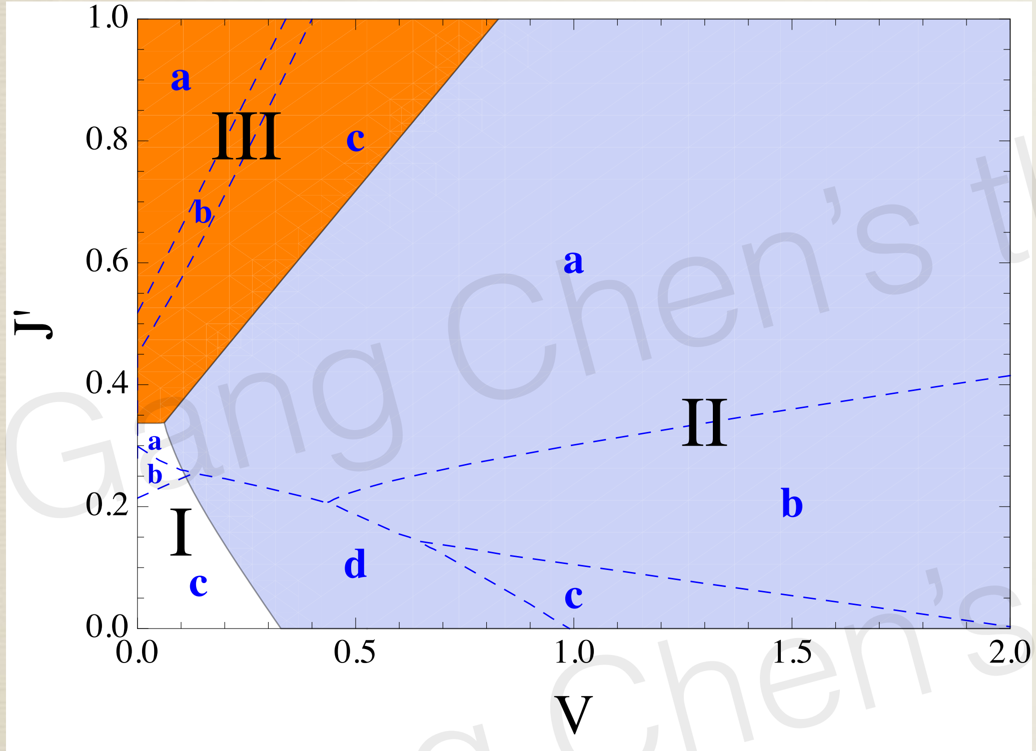
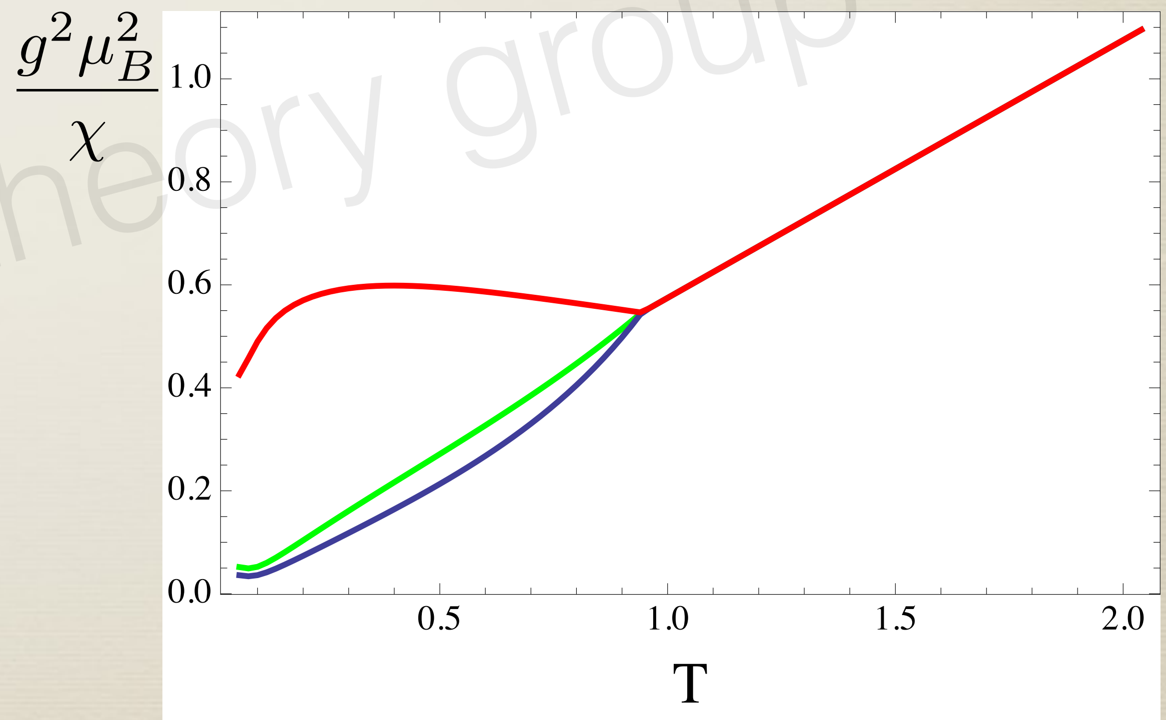


FIG. 3. (Color online) Finite temperature phase diagram. Region I (white) corresponds to a thermal transition from/to paramagnetic phase at $T_m(\mathbf{p} = 2\pi(001))$, region II (blue) corresponds to a thermal transition from/to paramagnetic phase at $T_Q(\mathbf{p} = 2\pi(001))$, region III (red) corresponds to a thermal transition from/to paramagnetic phase at $T_m(\mathbf{p} = \mathbf{0})$. Dashed curves are phase boundaries of the ground state phases taken from Fig. 1. "a, b, c, d" label the low temperature phases of each region. $J = 1$ in the phase diagram.

10 subregions in total!

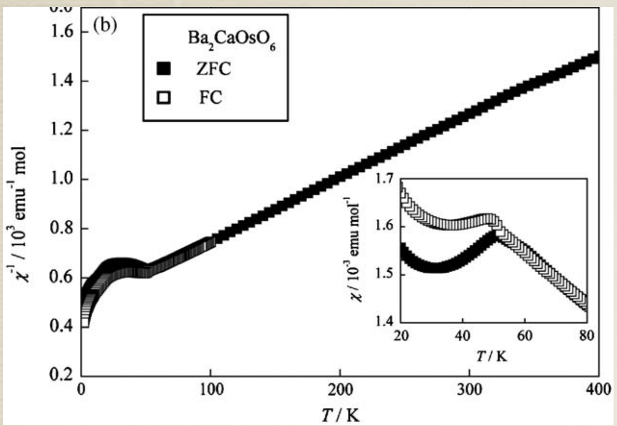
Region II (light blue): **spin nematic phase** at intermediate temperatures

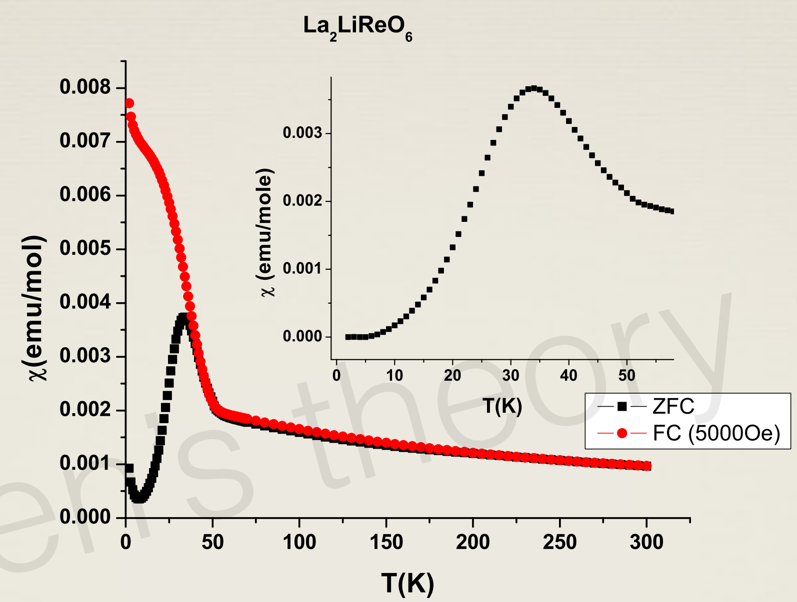


(b) $J' = 0.2J, V = 0.55J$ in II_b

Chen and Balents, (in preparation)

Experiments





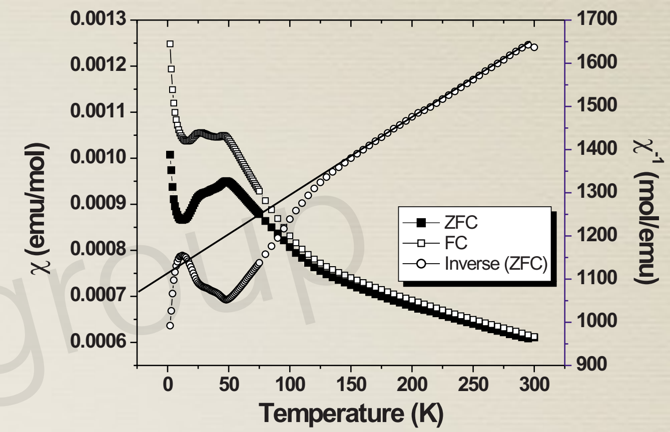
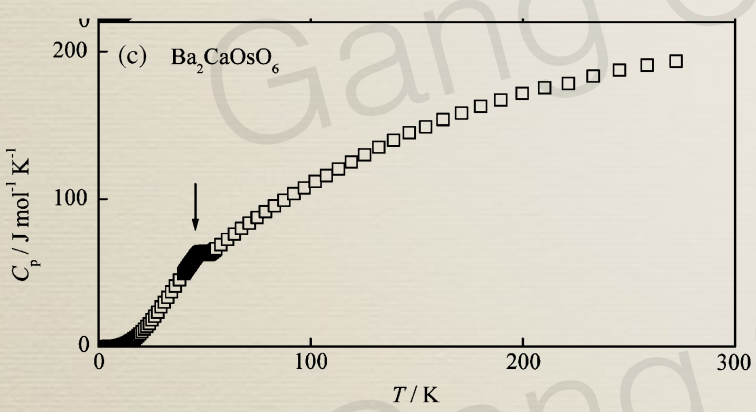
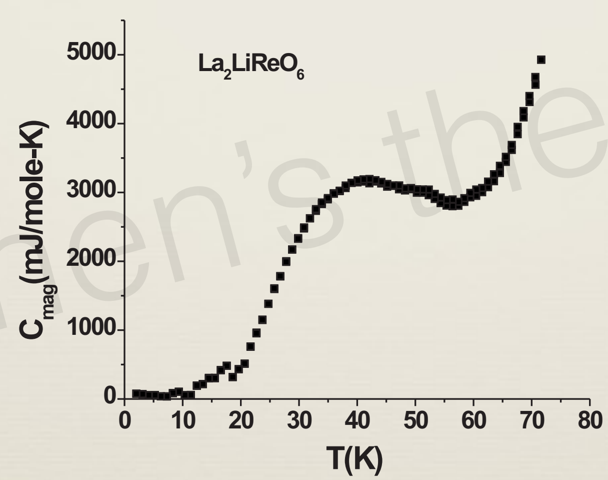
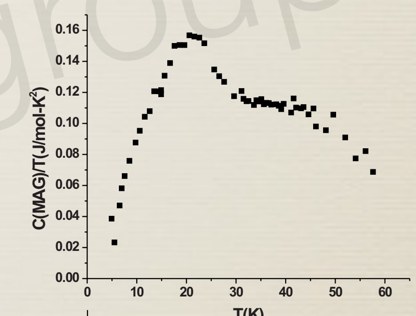
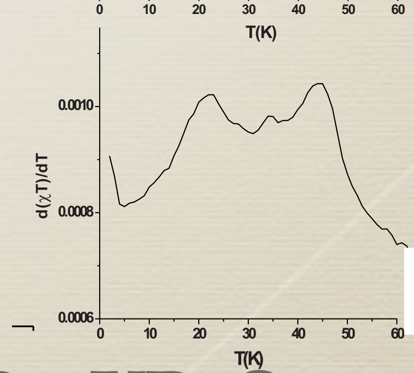


FIG. 12. (Color online) The magnetic susceptibility of Ba_2YReO_6. A Curie-Weiss fit of the inverse susceptibility data, solid line, yields the parameters C=0.554(5) (emu/mole K) and θ=−616(7) K.









Ba_2CaOsO_6

Single AFM phase transition corresponds to phases in region I

La_2LiReO_6 and Ba_2YReO_6

Seem to suggest two thermal transitions: two Curie regime!

Experiments: Aharen, et al PRB 81,064436, (2010)
Yamamura, et al JSSC 179 (2006) 605–612

Summary

- We study a geometrically frustrated magnetic system with strong SOC
- We find various zero temperature ground states and finite temperature phases in ordered double perovskites
- Single crystal samples and NMR+Neutron are required to identify these phases.