Orbital Frustration and Entanglement with Spin and Lattice degrees of freedom

Department of Physics
Tohoku University

Sumio Ishihara

TOHOKU UNIVERSITY

Emergent Quantum Phases in Condensed Matter (EQPCM)
ISSP Univ. of Tokyo, June 3-21, 2013

Outline

Outline

Introduction
Ring exchange interaction in orbital model
Orbital 120 model in honeycomb lattice
Entanglement of Orbital-Spin-Lattice in honeycomb lattce
Summary

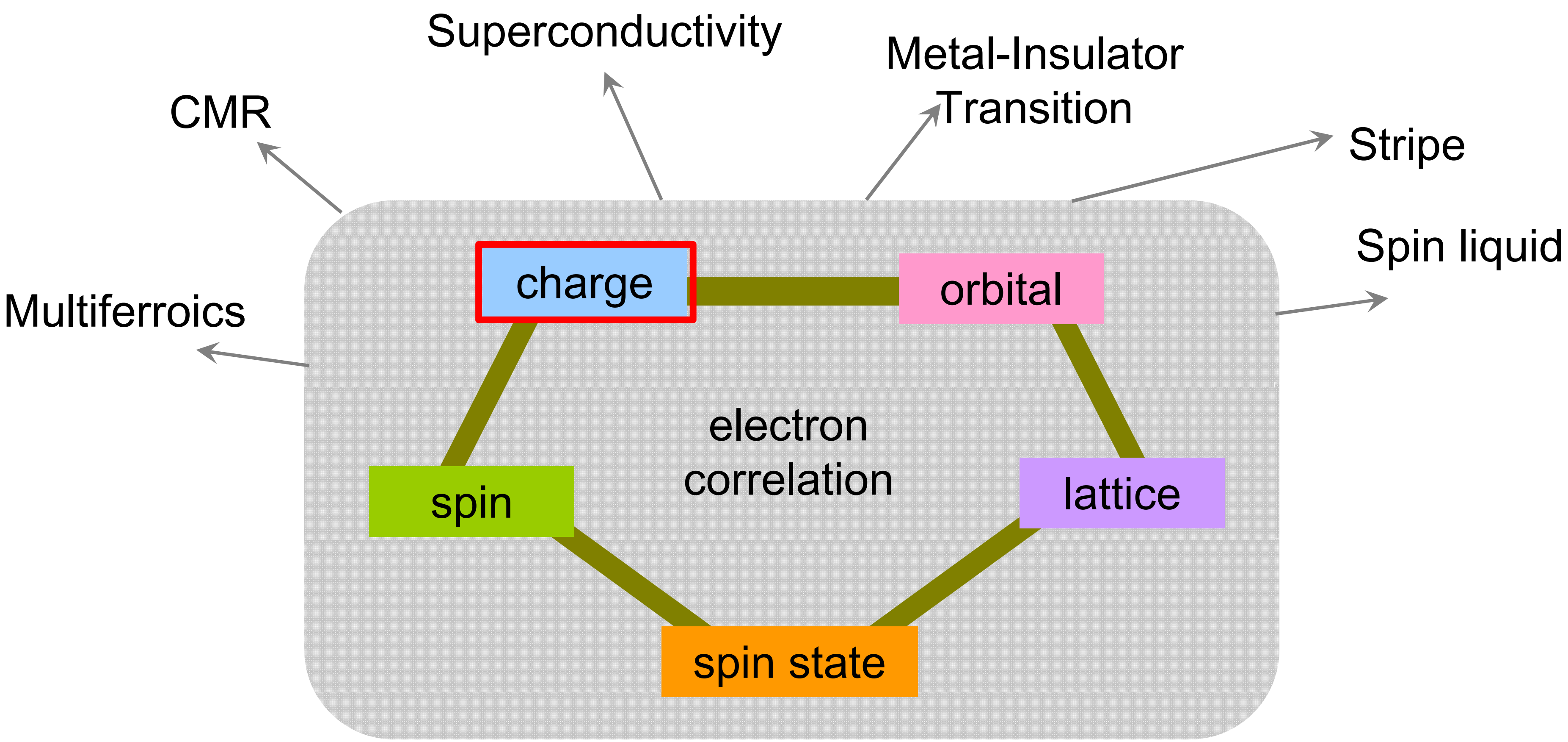
Collaborators

J. Nasu (Tohoku -> Tokyo), T. Tanaka (Tohoku)

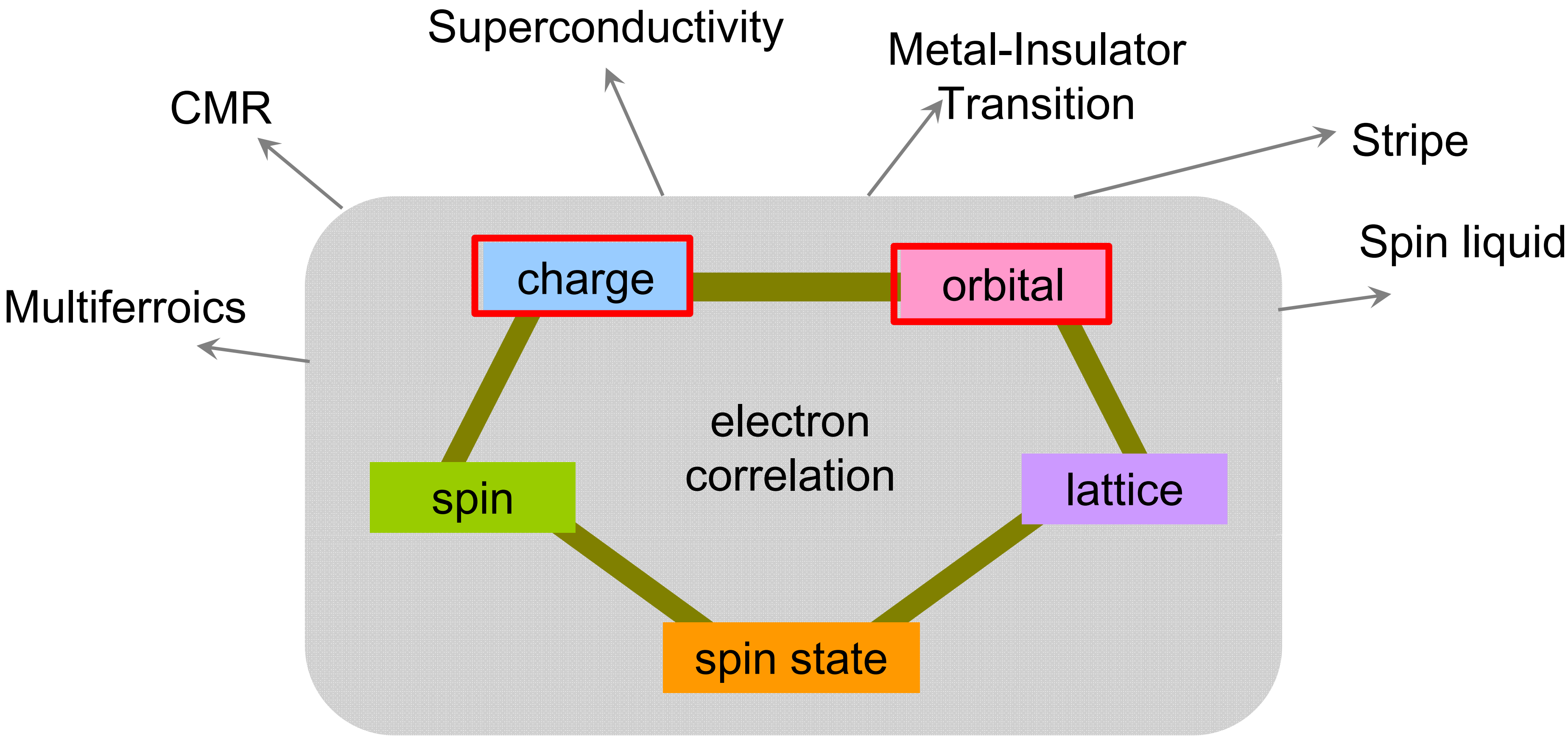
References

T. Tanaka, M. Matsumoto and SI	PRL **95**, 267204 ('05)
T. Tanaka and SI	PRL **98** 256402 ('07)
T. Tanaka and SI	PRB **78** 153106 ('08)
A. Nagano, M. Naka, J. Nasu, & SI,	PRL **99,** 217202 ('07)
M. Naka, A. Nagano, & SI,	PRB **77**, 224441 ('08)
J. Nasu, A. Nagano, M. Naka & SI,	PRB **78**, 024416 ('08)
J. Nasu & SI	JPSJ **80**, 033704 ('11)
J. Nasu and SI	EPL **97**, 27002 ('12)
J. Nasu, S. Todo, and SI	PRB **85**, 205141 ('12)
J. Nasu, and SI	arXiv: 12090239

Correlated system with multi-degrees



Orbital Physics



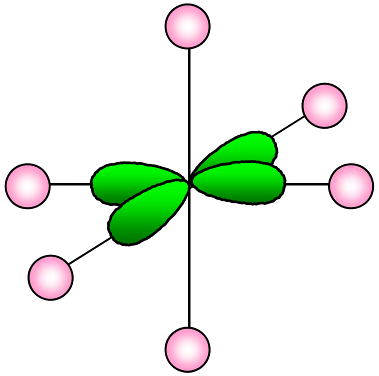
Kurogo ➡ Main actor, Hidden Boss

Orbital degree of freedom

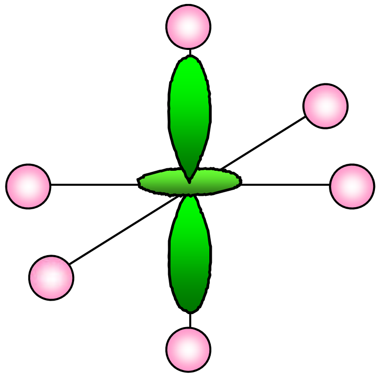
d orbital

$l=2$

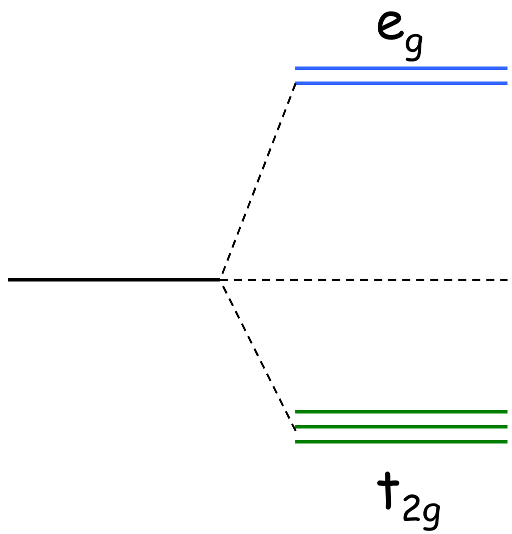
$2l+1=5$

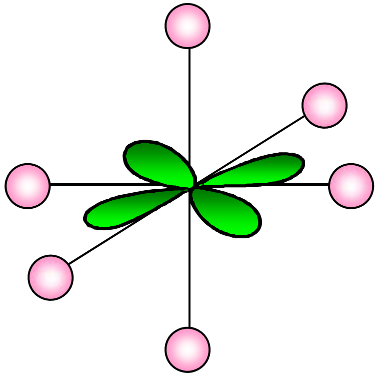


d(x^2-y^2)

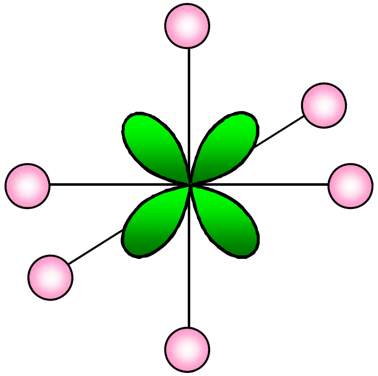


d($3z^2-r^2$)

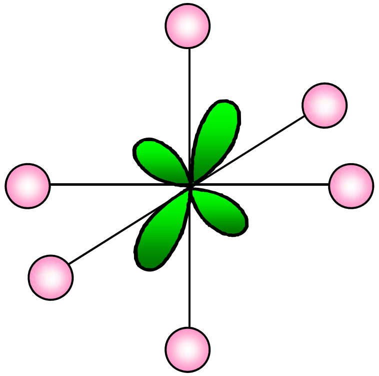




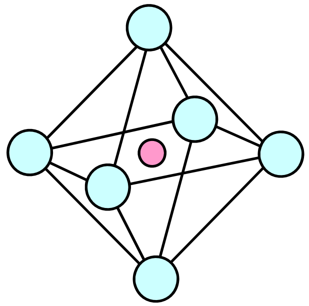
d(xy)



d(yz)

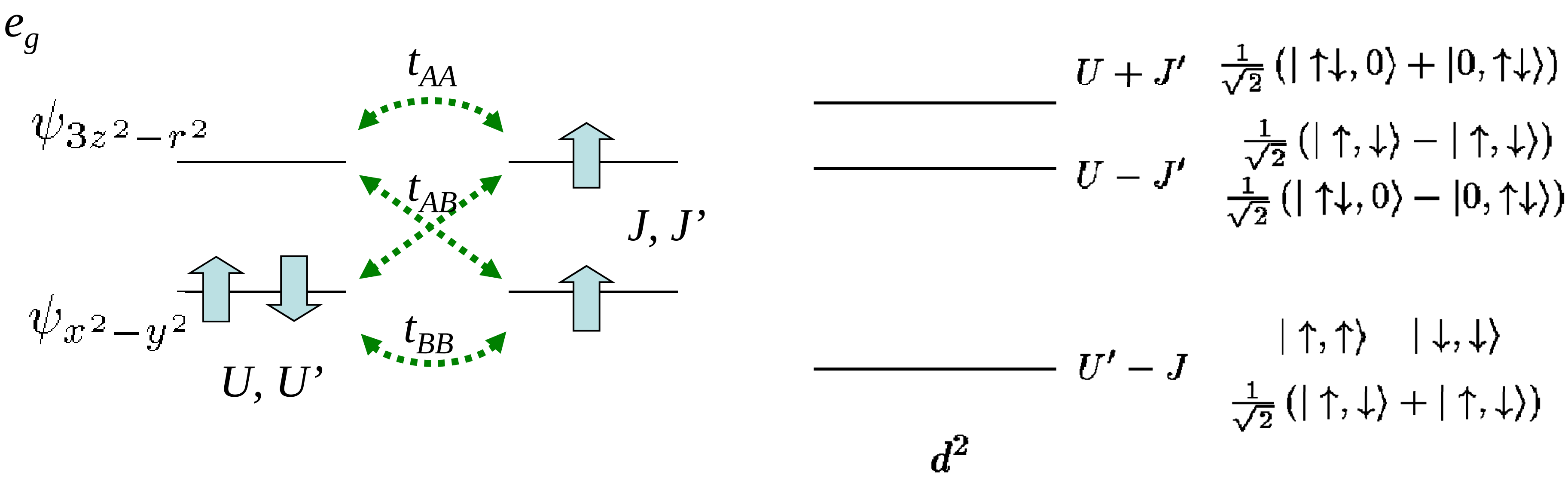


d(zx)

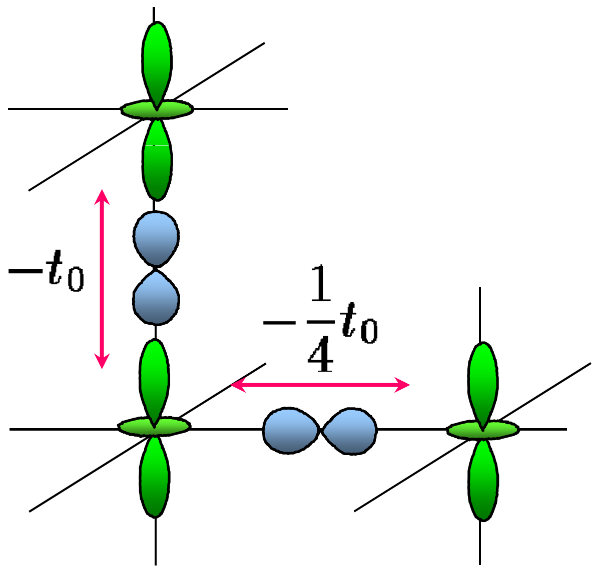


Multi-orbital Hubbard model

$$
\begin{aligned}
H &= \sum_{\langle ij \rangle \gamma \sigma} t_{\gamma\gamma'} \left(c^{\dagger}_{i\gamma\sigma} c_{j\gamma'\sigma} + H.c. \right) \\
&+ U \sum_{i\gamma} n_{i\gamma\uparrow} n_{i\gamma\downarrow} + U' \sum_{i\sigma\sigma'} n_{iA\sigma} n_{iB\sigma'} \\
&- J \sum_{i\sigma\sigma'} c^{\dagger}_{iA\sigma} c_{iB\sigma} c^{\dagger}_{iB\sigma'} c_{iA\sigma'} - J' \sum_{i\gamma} c^{\dagger}_{i\gamma\uparrow} c_{i\bar{\gamma}\uparrow} c^{\dagger}_{i\gamma\downarrow} c_{i\bar{\gamma}\downarrow},
\end{aligned}
$$

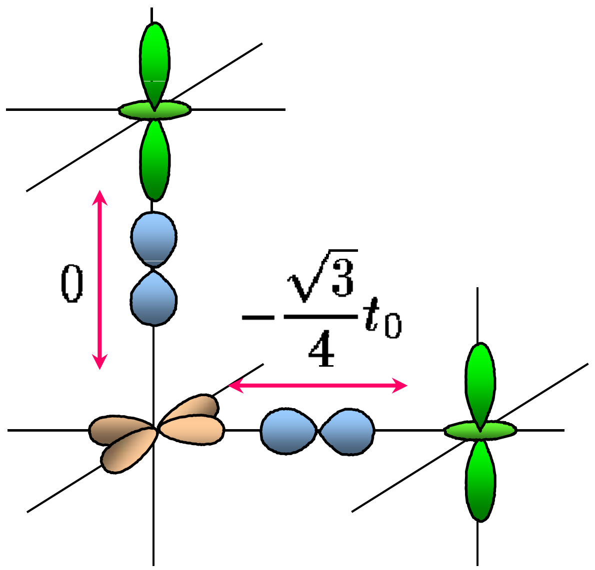


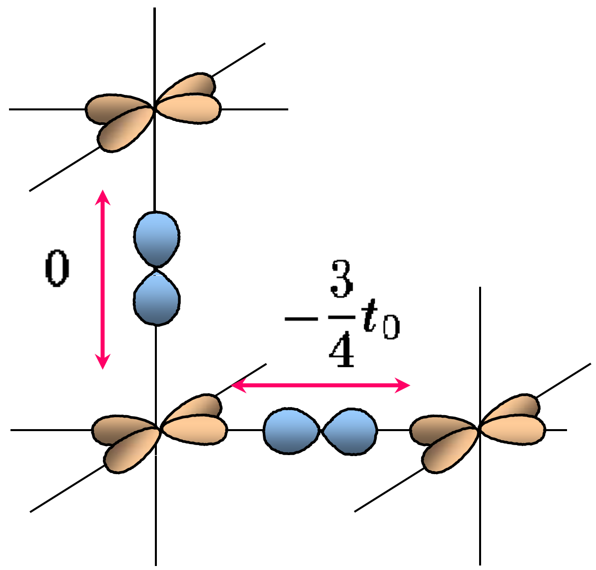
Electron transfer



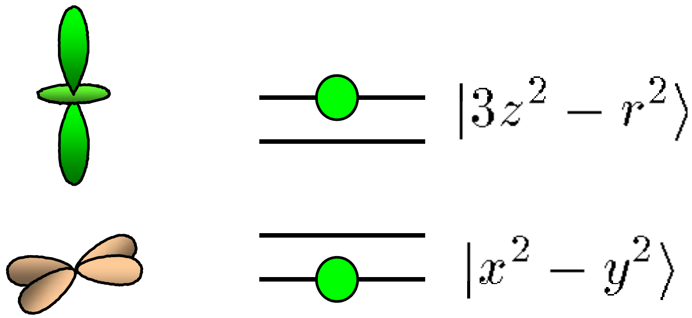
Through O2p orbital

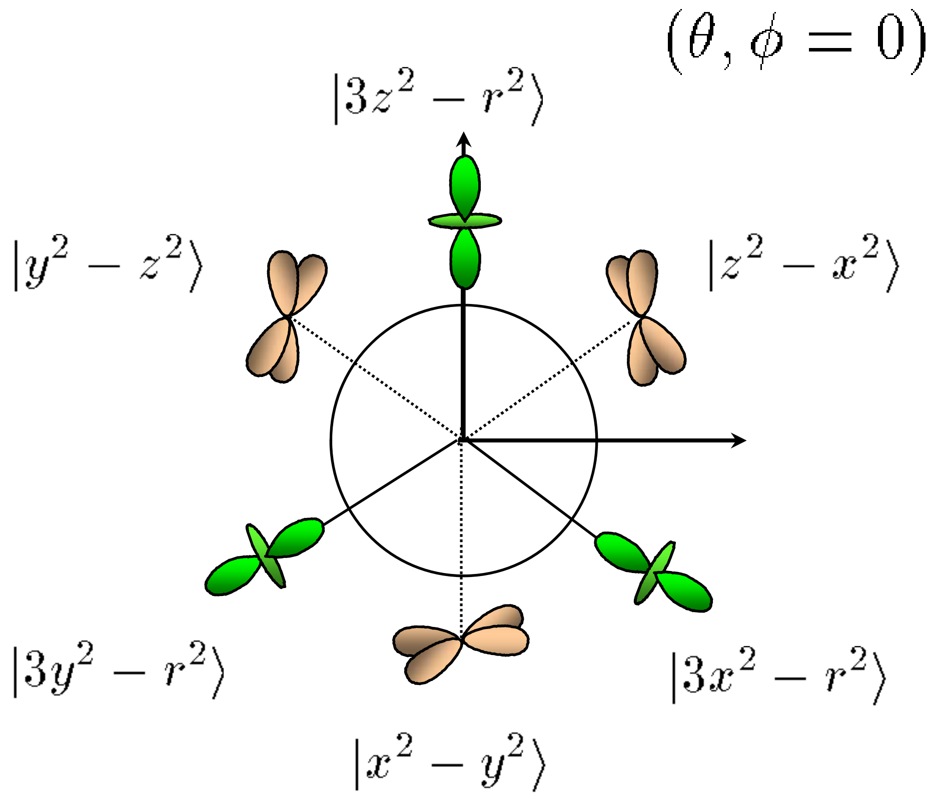
NN e_g orbital





Pseudo-spin



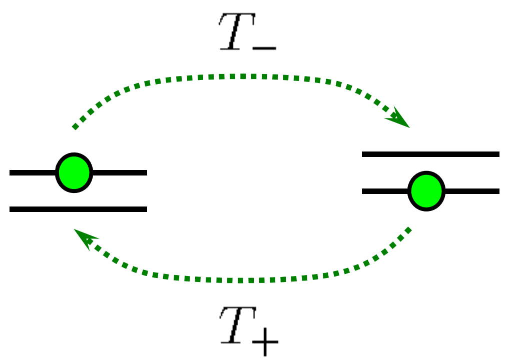


$$|\theta, \phi\rangle = \cos\left(\frac{\theta}{2}\right)|3z^2 - r^2\rangle + e^{i\phi}\sin\left(\frac{\theta}{2}\right)|x^2 - y^2\rangle$$

Pseudo-spin

$$\vec{T}_i = \frac{1}{2}\sum_{\gamma_1\gamma_2\sigma} c^\dagger_{i\gamma_1\sigma}\vec{\sigma}_{\gamma_1\gamma_2}c_{i\gamma_2\sigma}$$

$T_z = +\frac{1}{2}$



$T_z = -\frac{1}{2}$

$$E_g \times E_g = A_{1g} + E_g + A_{2g}$$

Electric monopole

T_0

Electric quadrupole

$T_x \; T_z$

$|3z^2 - r^2\rangle$

$|x^2 - y^2\rangle$

Magnetic monopole

T_y

$$\frac{1}{\sqrt{2}}\left(|3z^2 - r^2\rangle + i|x^2 - y^2\rangle\right)$$

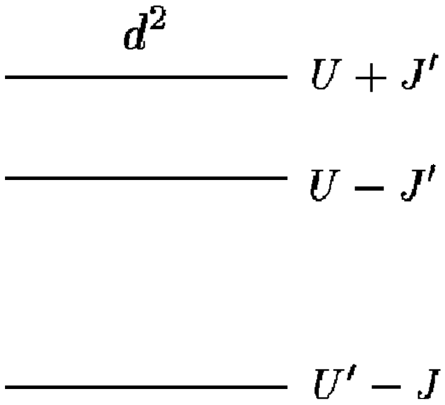
Spin-Orbital model

$$
\begin{aligned}
H = & -2J_1 \sum_{\langle ij \rangle} \left(\frac{3}{4} + \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{4} - \tau_i^l \tau_j^l \right) \\
& -2J_2 \sum_{\langle ij \rangle} \left(\frac{1}{4} - \vec{S}_i \cdot \vec{S}_j \right) \left[\left(\frac{1}{4} - \tau_i^l \tau_j^l \right) + \left(\frac{1}{2} + \tau_i^l \right) \left(\frac{1}{2} + \tau_j^l \right) \right] \\
& -2J_3 \sum_{\langle ij \rangle} \left(\frac{1}{4} - \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{2} + \tau_i^l \right) \left(\frac{1}{2} + \tau_j^l \right)
\end{aligned}
$$

$$\tau_i^l = \cos\left(\frac{2\pi n_l}{3}\right) T_i^z + \sin\left(\frac{2\pi n_l}{3}\right) T_i^x$$

$(n_x, n_y, n_z) = (1, 2, 3)$ l: $i\ j$ bond direction

$$J_1 = \frac{t_0^2}{U' - J} \qquad J_2 = \frac{t_0^2}{U - J'} \qquad J_3 = \frac{t_0^2}{U + J'}$$



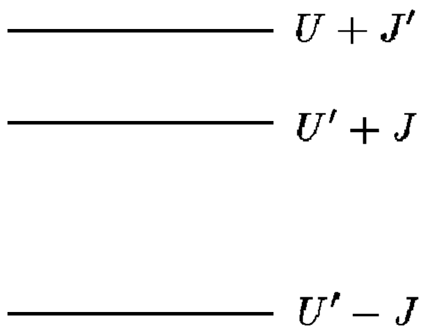
K. I. Kugel, and D. I. Khomskii,
Sov. Phys. Usp. 25, 231 ('82).

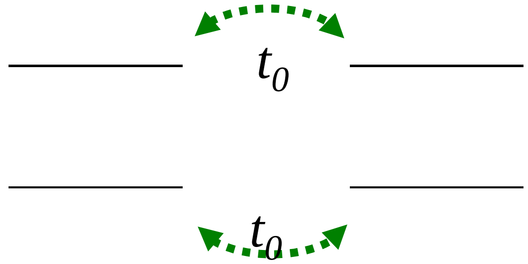
Spin-Orbital model

$$t_{\gamma_1\gamma_2} = \delta_{\gamma_1\gamma_2} t_0 \quad J_2 = J_3$$

SU(2) ×SU(2) spin-orbital model

$$\begin{aligned} H &= -2J_1 \sum_{\langle ij \rangle} \left(\frac{3}{4} + \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{4} - \vec{T}_i \cdot \vec{T}_j \right) \\ &\quad -2J_2 \sum_{\langle ij \rangle} \left(\frac{1}{4} - \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{3}{4} + \vec{T}_i \cdot \vec{T}_j \right) \end{aligned}$$





$$t_{\gamma_1\gamma_2} = \delta_{\gamma_1\gamma_2} t_0 \quad J_1 = J_2 = J_3 \; (J = J' = 0)$$

SU(4) spin-orbital model

$$H = 2J_1 \sum_{\langle ij \rangle} \left(\frac{1}{4} + \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{4} + \vec{T}_i \cdot \vec{T}_j \right) + const.$$

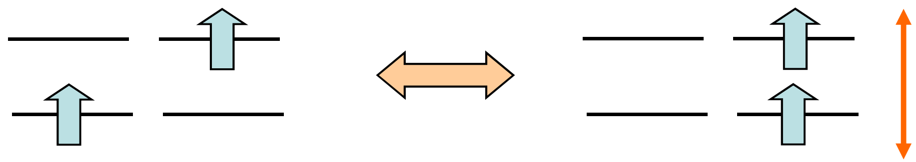
Spin-Orbital model

SU(2) ×*SU*(2) spin-orbital model

$$J_1 = \frac{t_0^2}{U' - J} > J_2 = J_3 = \frac{t_0^2}{U + J}$$

$$H = -2J_1 \sum_{\langle ij \rangle} \left(\frac{3}{4} + \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{4} - \vec{T}_i \cdot \vec{T}_j \right)$$

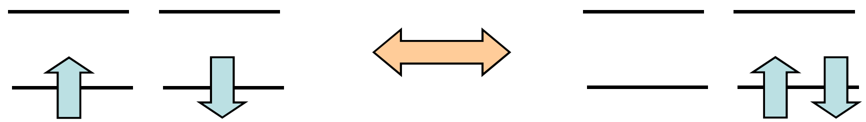
Spin triplet Orbital singlet



Hund's coupling

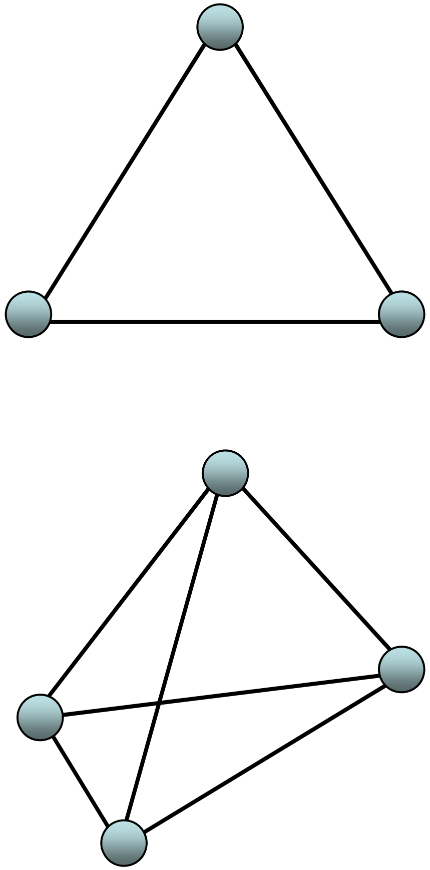
$$-2J_2 \sum_{\langle ij \rangle} \left(\frac{1}{4} - \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{3}{4} + \vec{T}_i \cdot \vec{T}_j \right)$$

Spin singlet Orbital triplet

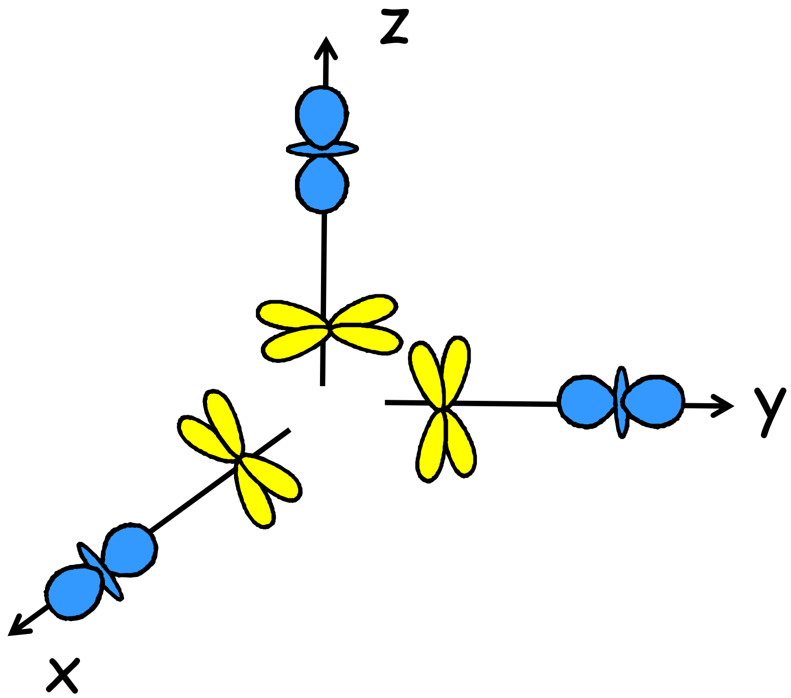


Orbital & frustration

Geometrical frustration



Intrinsic orbital frustration



Even without geometrical frustration

Orbital models

Spin-Orbital model

$\vec{T}$

Pseudo-spin (S=1/2)
for doubly degenerate orbital

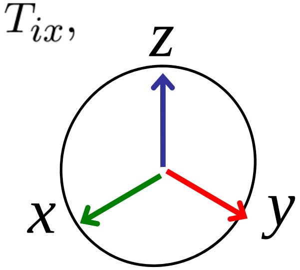
Kugel-Khomskii model

$$\mathcal{H}_{SE} = -2J_1 \sum_{\langle ij \rangle} \left(\frac{3}{4} + \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{4} - \tau_i^l \tau_j^l \right)$$
$$-2J_2 \sum_{\langle ij \rangle} \left(\frac{1}{4} - \vec{S}_i \cdot \vec{S}_j \right) \left[\left(\frac{1}{4} - \tau_i^l \tau_j^l \right) + 2 \left(\frac{1}{2} + \tau_i^l \right) \left(\frac{1}{2} + \tau_j^l \right) \right]$$

Orbital only model

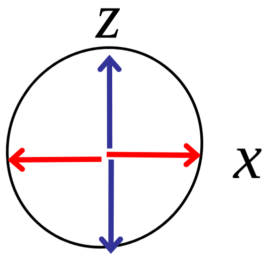
120° model

$$\mathcal{H} = 2J \sum_{\langle ij \rangle} \tau_i^l \tau_j^l \qquad \tau_i^l = \cos\left(\frac{2\pi}{3} n_l\right) T_{iz} + \sin\left(\frac{2\pi}{3} n_l\right) T_{ix},$$



Orbital compass model

$$\mathcal{H} = 2J \sum_{\langle ij \rangle} T_i^l T_j^l$$



120° model in a cubic lattice

$$\mathcal{H} = 2J \sum_{\langle ij \rangle} \tau_i^l \tau_j^l$$

$$\tau_i^l = \cos\left(\frac{2\pi}{3} n_l\right) T_{iz} + \sin\left(\frac{2\pi}{3} n_l\right) T_{ix},$$

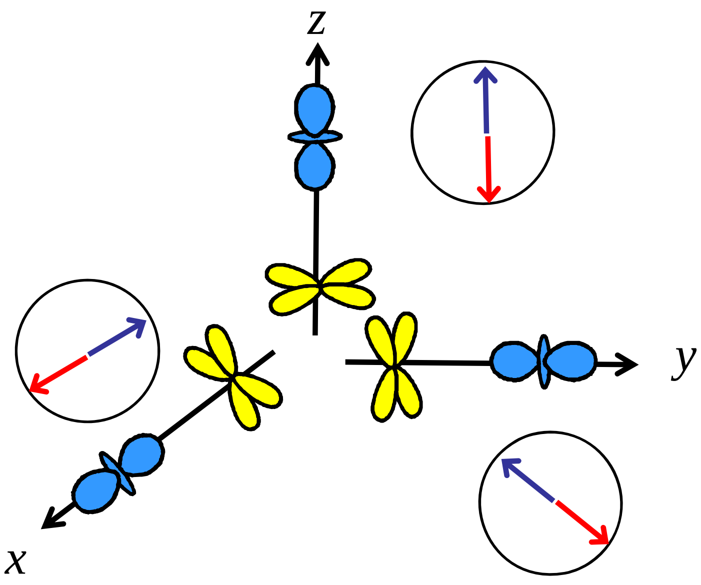
$l(= x, y, z)$: bond direction

$$(n_x, n_y, n_z) = (1, 2, 3)$$

$$\begin{cases} T_{iz} T_{jz} & l = z \\ \left[-\frac{1}{2} T_{iz} + \frac{\sqrt{3}}{2} T_{ix}\right] \left[-\frac{1}{2} T_{jz} + \frac{\sqrt{3}}{2} T_{jx}\right] & l = x \\ \left[-\frac{1}{2} T_{iz} - \frac{\sqrt{3}}{2} T_{ix}\right] \left[-\frac{1}{2} T_{jz} - \frac{\sqrt{3}}{2} T_{jx}\right] & l = y \end{cases}$$

Interaction explicitly depends on bond direction

eg orbitals in a cubic lattice

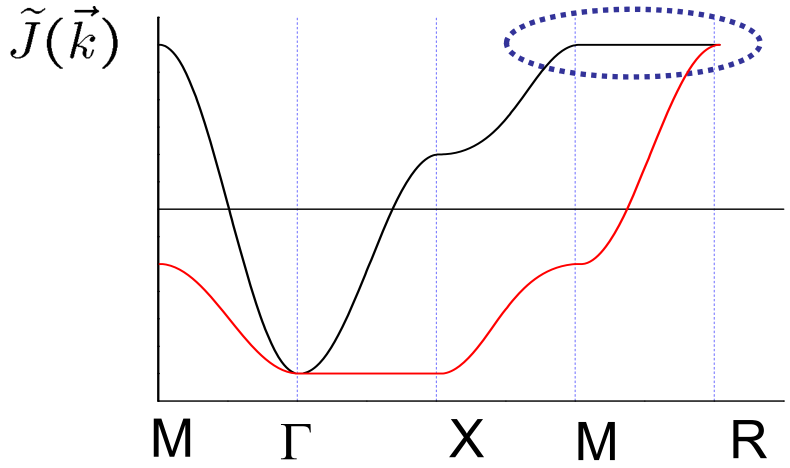


A kind of frustration

Orbital 120° model in a cubic lattice

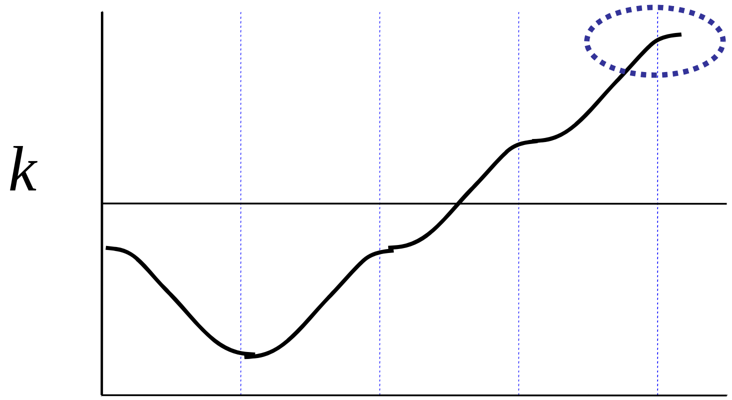
Interaction in momentum space

e_g orbital model



$$\tilde{J}(\vec{k}) = J\Big[-c_x - c_y - c_z \pm \sqrt{c_x^2 + c_y^2 + c_z^2 - c_x c_y - c_y c_z - c_z c_x}\Big]$$

Heisenberg model



$$J(\vec{k}) = J\left\{c_x + c_y + c_z\right\}$$

$$c_l = \cos(ak_l)$$

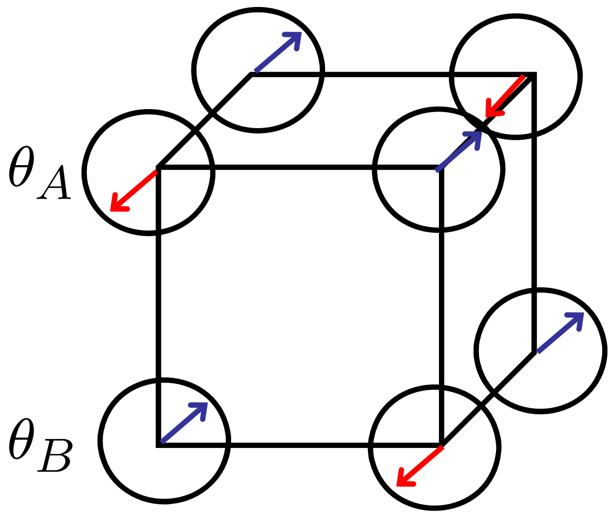
Orbital configuration is not determined uniquely in classical ground state
(like frustrated spin systems)

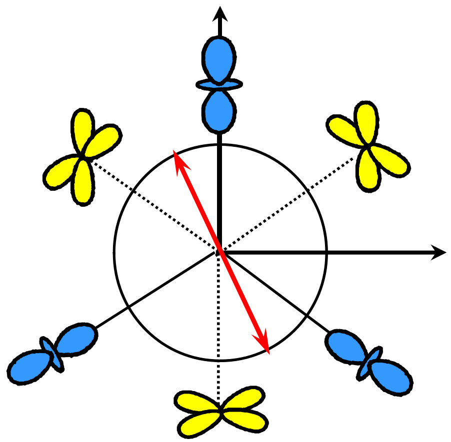
Degeneracy in classical ground state

A macroscopic degeneracy in classical GS

[1] continuous staggered states

Feiner et al PRL(97)
Khaliullin et al. PRB(97)
Ishihara et al. PRB (00)
Kubo et al. JPSJ (02)
Nussinov et al. EPL(04)





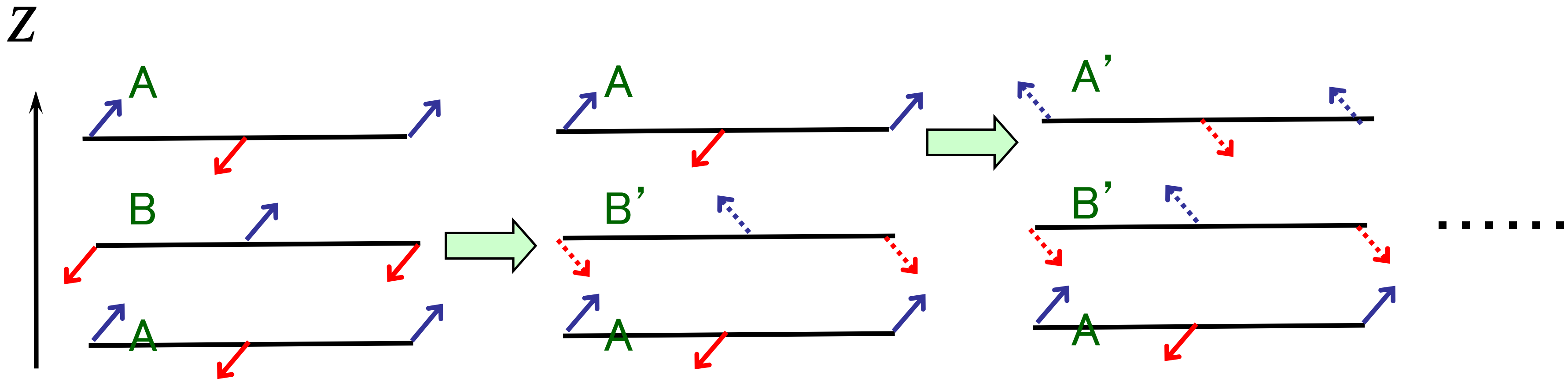
$$(\theta_A/\theta_B) = (\theta/\theta+\pi) \quad \forall\theta$$

No continuous symmetry
in Hamiltonian

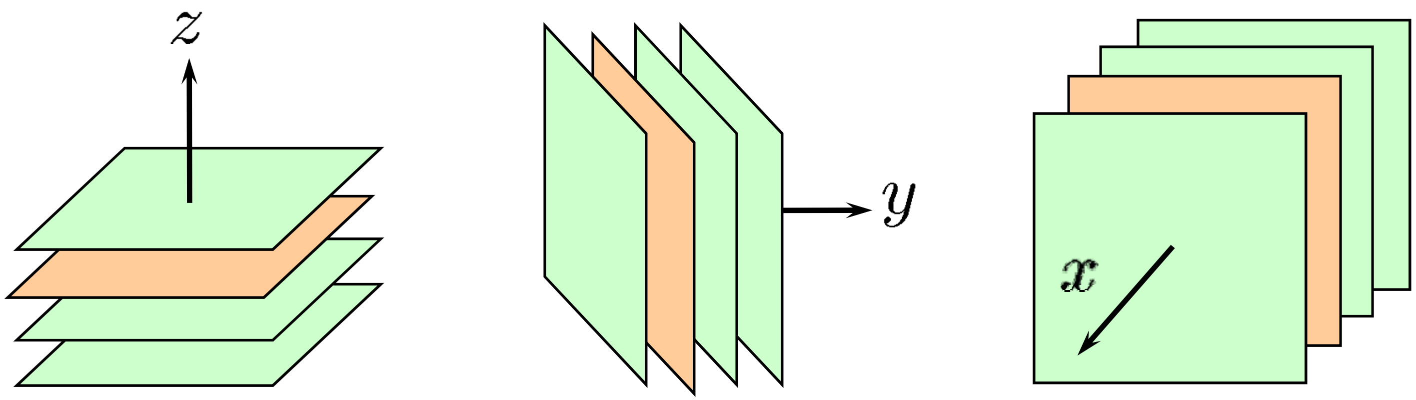
Degeneracy in classical ground state

[2] staking degenerate states

$$\mathcal{H}(\mathrm{z-direction}) \sim 2JT_i^zT_j^z$$

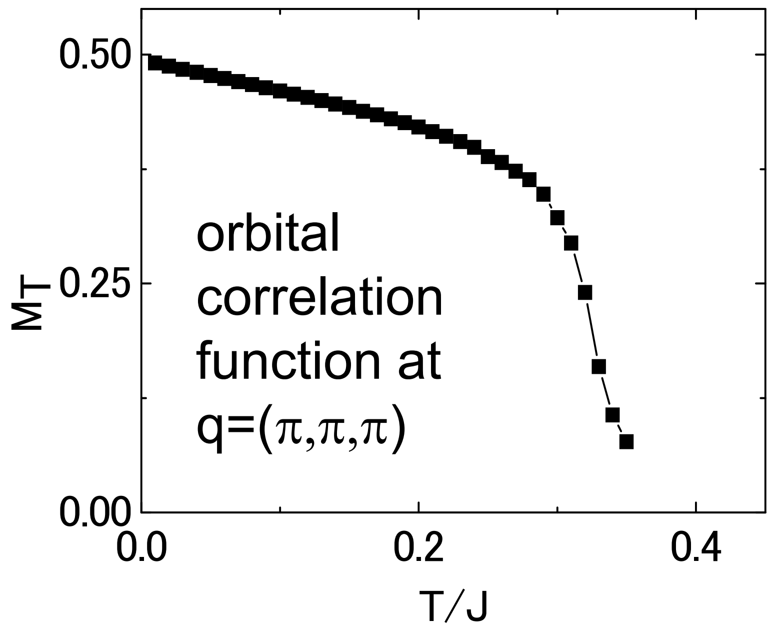


$T^x \rightarrow -T^x$ $T^x \rightarrow -T^x$



Order by fluctuation

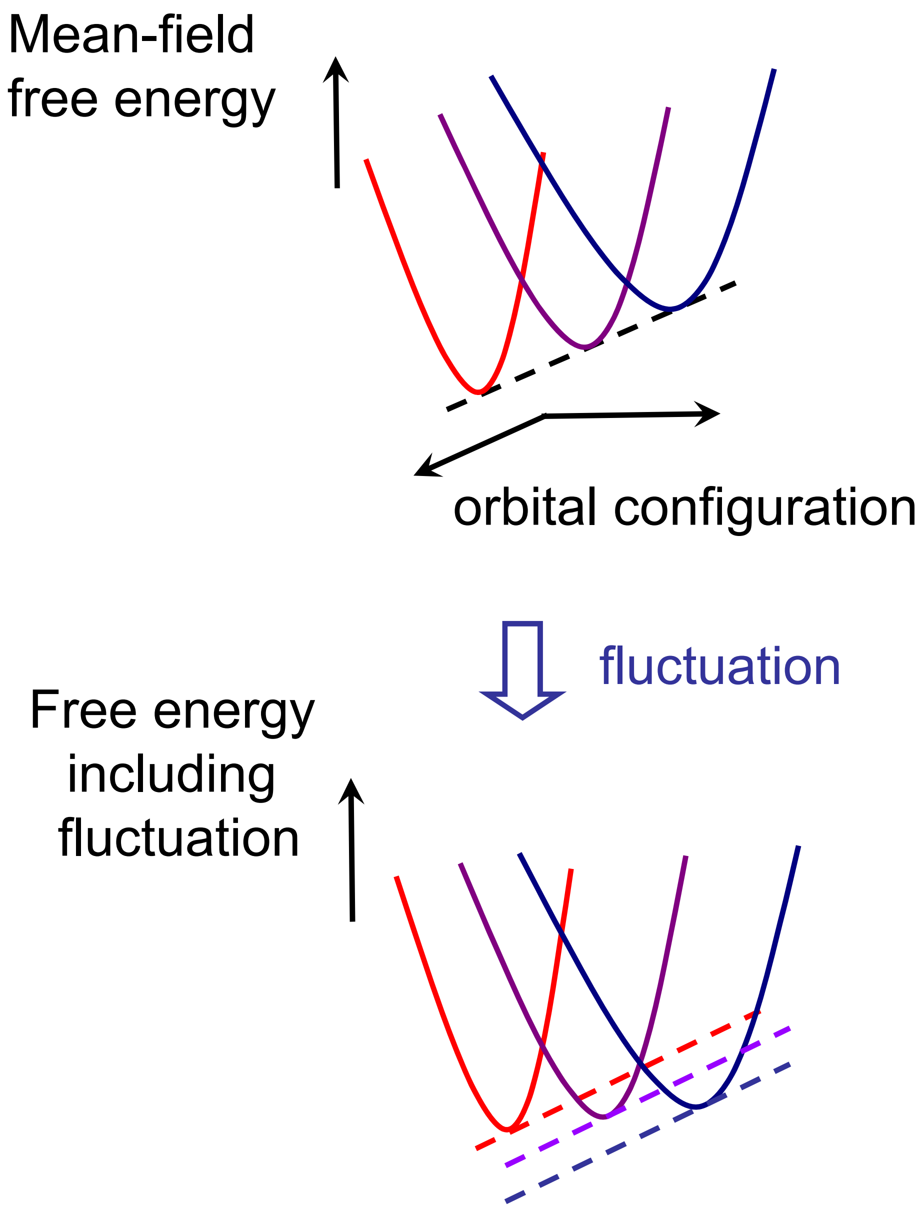
Classical Monte Carlo
T. Tanaka & SI, PRB ('09)



Thermal/Quantum fluctuation

⇩

Long-range orbital order

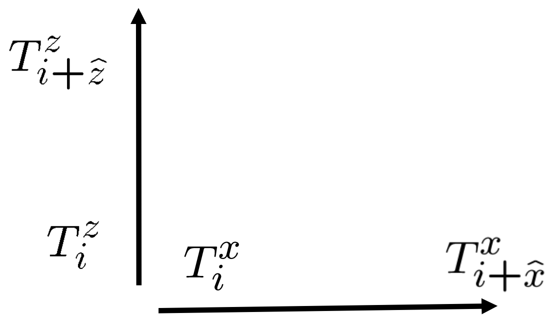


Feiner et al (97), Khaliullin et al. (97)
SI et al. (00), Kubo et al. (02), Nussinov et al. (04)

Compass model in a square lattice

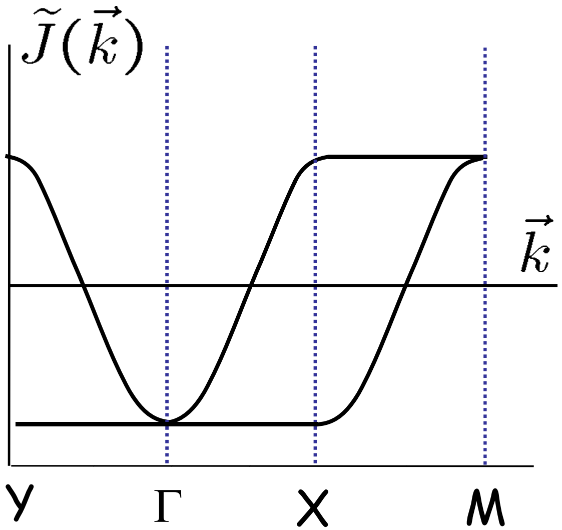
“Orbital compass model” in a 2-dim. square lattice

$$\mathcal{H} = J \sum_i \left(T_i^x T_{i+\hat{x}}^x + T_i^z T_{i+\hat{z}}^z \right)$$



Kugel-Khomskii JETP (73),
Khomskii-Mostovoy J. Phys (03)
Oles Gr.

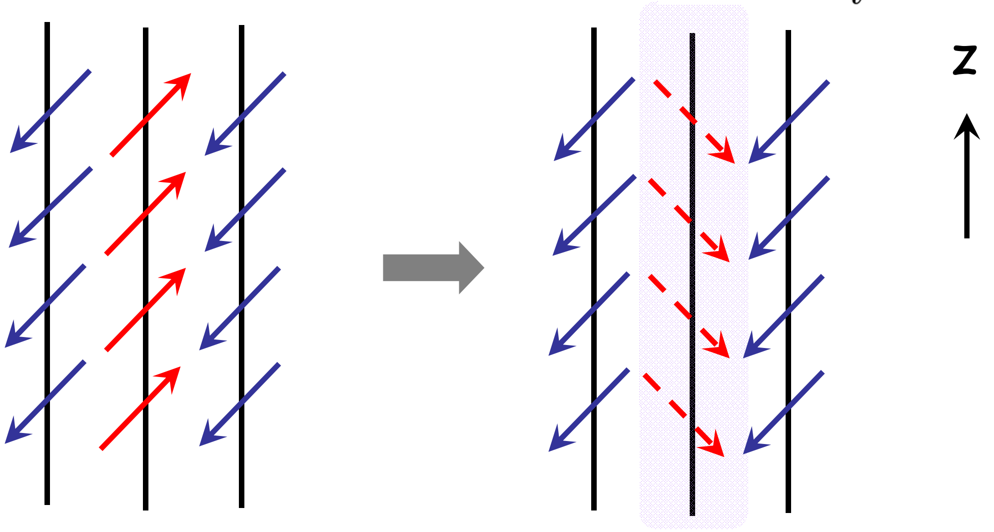
Momentum dependence of orbital interaction



Continuous classical GS degeneracy

Symmetry of Compass model

$$\mathcal{H} = J\sum_i \left(T_i^x T_{i+\hat{x}}^x + T_i^z T_{i+\hat{z}}^z\right)$$

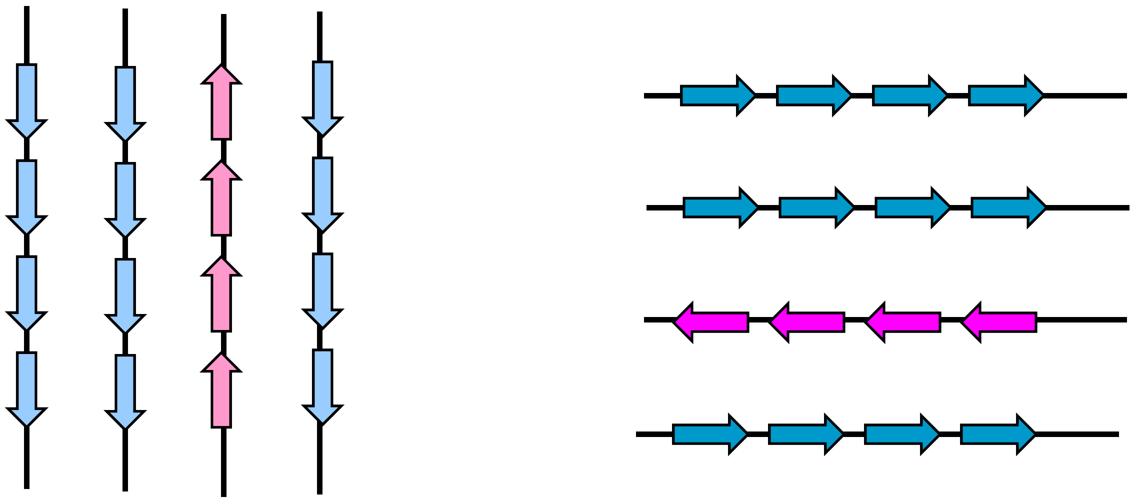


Hamiltonian is invariant
under the transformation of $T^z \to -T^z$ in each column

Mishra et al PRL(04), Nussinov et al. EPL(04),
Dorier et al. PRB('05), Doucot et al. PRB(05)

Conventional orbital order does not appear
(generalized Elitzur's theorem)

Directional order



Directional order

: T^x (T^z) correlation along x (z) direction

$$D = N^{-1} \sum_i \left(T_i^x T_{i+\hat{x}}^x - T_i^z T_{i+\hat{z}}^z \right)$$

Mishra et al PRL(04) Classical
Dorier et al. PRB('05) T=0
Doucot et al. PRB(05) T=0

Present talk

Orbital 120°model
in a cubic lattice

Classical GS degeneracy
Order by disorder

Ring exchange interaction
in orbital 120 °model

Orbital 120 °model
in honeycomb lattice

Effect of
dynamical Jahn-Teller
and spin liquid

Orbital excitation

Ring-exchange interaction

in orbital 120 mode

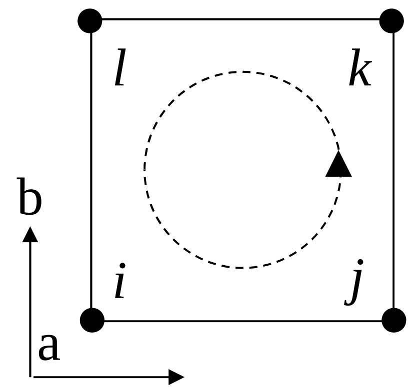
Ring exchange interaction

e_g orbital model
in a cubic lattice

a macroscopic number of degeneracy
↓
LRO by fluctuation

Ring exchange Interaction

Long range interaction

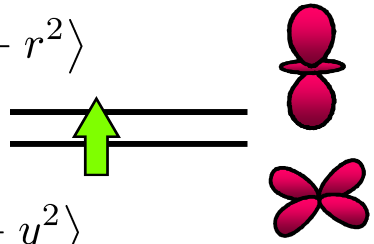


^{3}He
High-Tc cuprates
Spin ladder
Triangular magnet
SU(4) model
Deconfined criticality

Close to MIT
(Nickelates
Manganite with pressure)

Spin-less e_g orbital Hubbard model

$$|u\rangle = |3z^2 - r^2\rangle$$

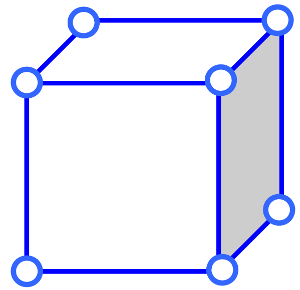


$$|v\rangle = |x^2 - y^2\rangle$$

$$H = \sum_{<ij>_l} \sum_{\mu\mu'=u,v} \left[t_l^{\mu\mu'} c_{i\mu}^\dagger c_{j\mu'} + h.c. \right] + U \sum_i n_{iu} n_{iv}$$
$$= H_t + H_U$$

perturbation up to O(t^4)

$$H_{\text{eff}} = H_2 + H_4$$



Cubic lattice
Half filling
Spin less

2nd order

$$H_J = H_2 = J \sum_i \left(\tau_i^x \tau_{i+\hat{x}}^x + \tau_i^y \tau_{i+\hat{y}}^y + \tau_i^z \tau_{i+\hat{z}}^z \right)$$

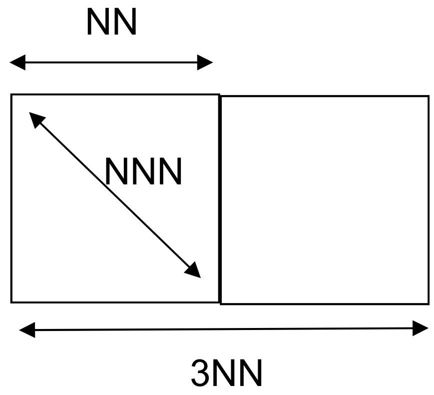
$$\begin{cases} \tau_i^x = -\frac{1}{2} T_i^z - \frac{\sqrt{3}}{2} T_i^x \\ \tau_i^y = -\frac{1}{2} T_i^z + \frac{\sqrt{3}}{2} T_i^x \\ \tau_i^z = T_i^z \end{cases}$$

$d_{3z^2-r^2}$	$d_{x^2-y^2}$
$T^z \rightarrow +\frac{1}{2}$	$T^z \rightarrow -\frac{1}{2}$

Ring exchange interaction

4th order term

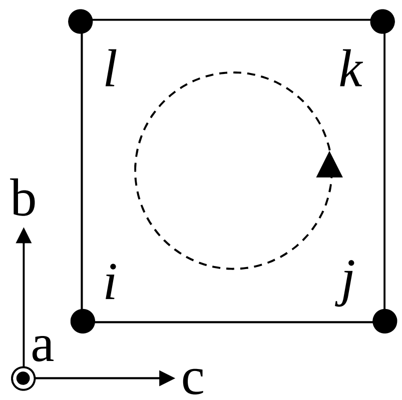
$$
\begin{aligned}
\mathcal{H}_4 \;=\;& K_{NN} \sum_{\langle ij \rangle_a} \left(\tau_i^a \tau_j^a - \bar{\tau}_i^a \bar{\tau}_j^a - T_i^y T_j^y \right) \\
+\;& K_{NNN} \sum_{\langle ij \rangle_a}{}' \left(\tau_i^a \tau_j^a - 5 \bar{\tau}_i^a \bar{\tau}_j^a + \frac{1}{2} T_i^y T_j^y \right) \\
+\;& K_{3NN} \sum_{\langle ij \rangle_a}{}'' \tau_i^a \tau_j^a + \mathcal{H}_R
\end{aligned}
$$



$K_{NN} = 3t^4/(2U^3)$ $K_{NNN} = 3t^4/(4U^3)$ $K_{3NN} = 4t^4/U^3$

Ring exchange interaction

$$\mathcal{H}_R = K_R \sum_{[ijkl]_a} \frac{1}{2} \left(\tau_i^{a+} \tau_j^{a-} \tau_k^{a+} \tau_l^{a-} + H.c. \right)$$



$$\tau_i^{\pm a} = \tau_i^a \pm i(\sqrt{3}/2) T_i^y$$

$$K_R = 40t^4/U^3$$

$$r_R = \sqrt{K_R/(20J)}$$

$r_R = 0.1 - 0.2$ $LaMnO_3$

$r_R = 0.4 - 0.5$ $LaNiO_3$

Ring exchange v.s. NN exchange

c.f. f-electron system (Kuramoto, Shiba)
doped manganite (Khomskii, Shiba, Nagaosa)

Ring exchange interaction

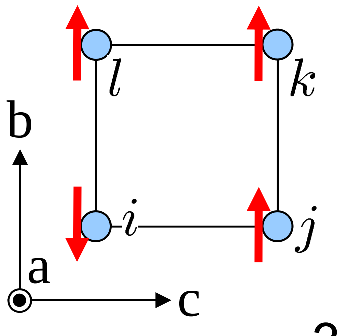
$$\mathcal{H}_R = K_R \sum_{[ijkl]_a} \frac{1}{2}\left(\tau_i^{a+}\tau_j^{a-}\tau_k^{a+}\tau_l^{a-} + H.c.\right)$$

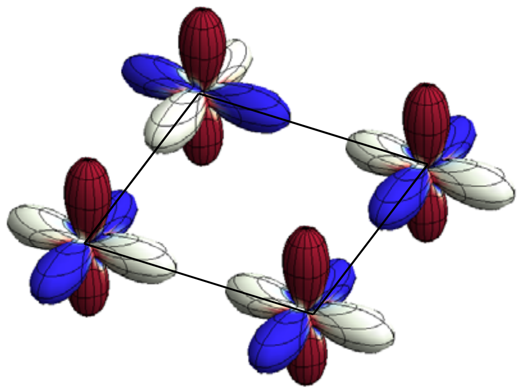
$$T^y \propto \overline{l_x l_y l_z}$$ A_{2g} Magnetic octupole

$$|3z^2 - r^2|\rangle + i|x^2 - y^2\rangle$$

$$\tau_i^{\pm a} = \tau_i^a \pm i(\sqrt{3}/2)T_i^y$$

$$K_R = 40t^4/U^3$$





3-up 1-down

2nd order NN exchange

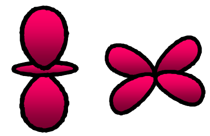
$$H_J = H_2 = J\sum_i \left(\tau_i^x\tau_{i+\hat{x}}^x + \tau_i^y\tau_{i+\hat{y}}^y + \tau_i^z\tau_{i+\hat{z}}^z\right)$$

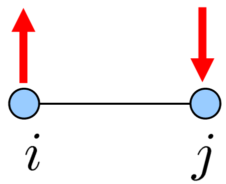
$$\begin{cases} \tau_i^x = -\frac{1}{2}T_i^z - \frac{\sqrt{3}}{2}T_i^x \\ \tau_i^y = -\frac{1}{2}T_i^z + \frac{\sqrt{3}}{2}T_i^x \\ \tau_i^z = T_i^z \end{cases}$$

$$T^z \propto 3l_z^2 - \boldsymbol{l}^2$$

$$T^x \propto l_x^2 - l_y^2$$

E_g Electric quadrupole





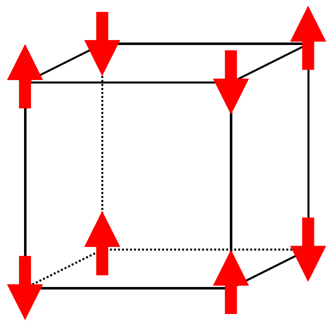
AF

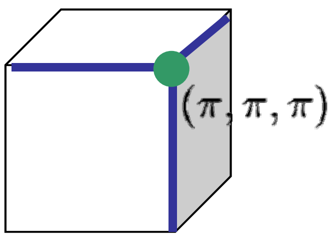
Phase diagram (classical)

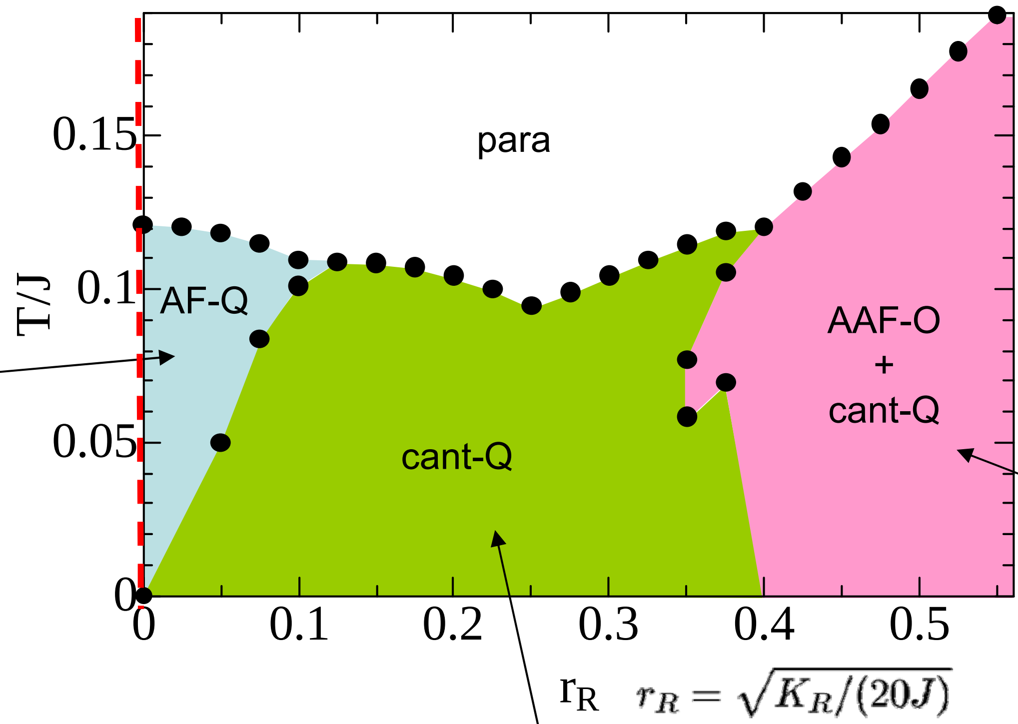
$$H_{\text{eff}} = H_2 + H_4$$

Classical Monte Carlo
Mean field
10*10*10 cluster
Wang-Landau method

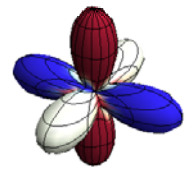
Antiferro Quadrupole



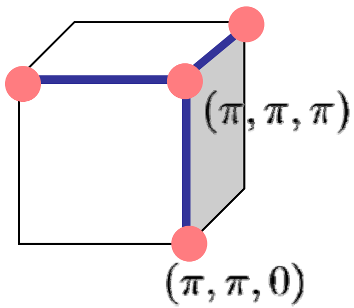


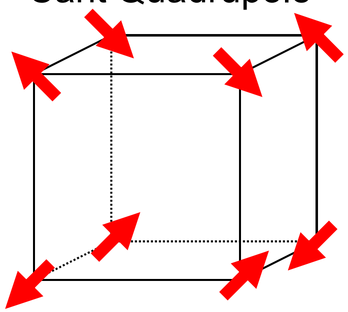


Octupole
+
Qudrupole



Cant Quadrupole

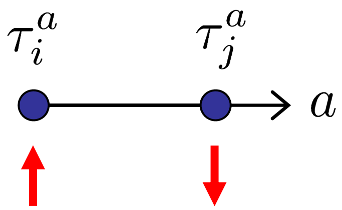


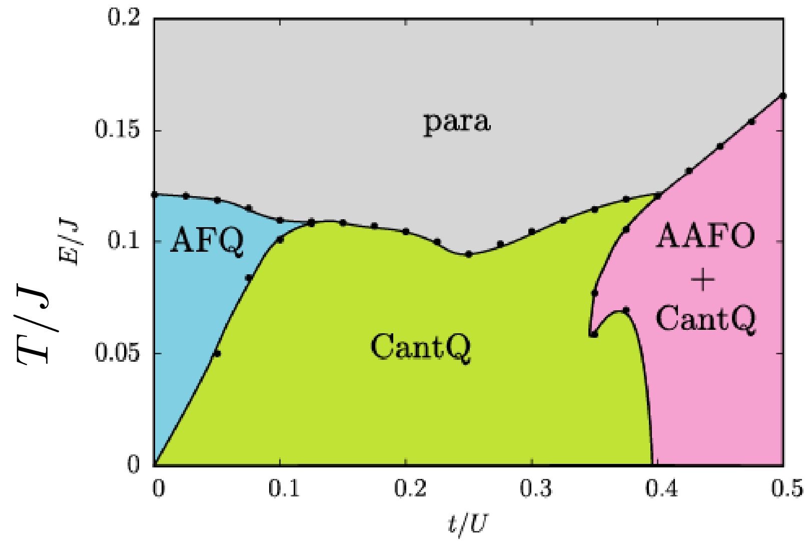


Ring exchange
lifts the degeneracy
in a different way
from NN exchange

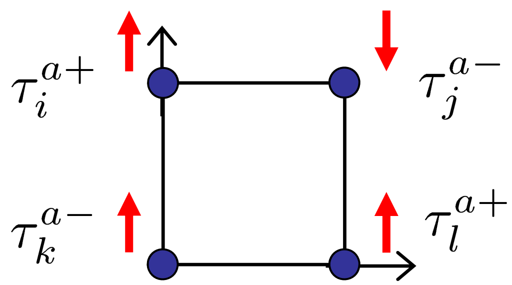
Octupole order

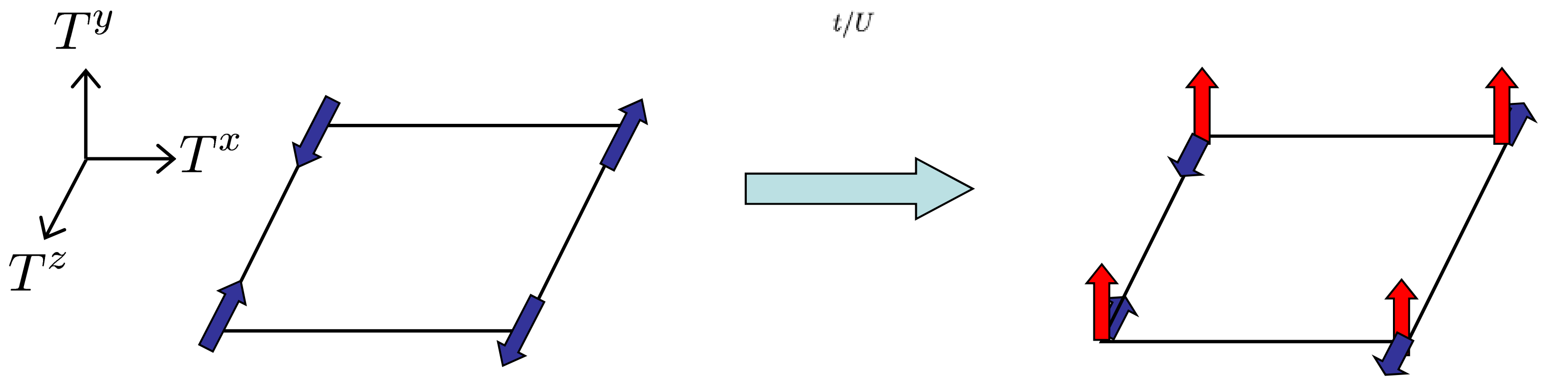
AF





1-up 3-down
1-down 3-up





Strong competition between H_J & H_K on the Tx-Tz plane

Ferro T_y order

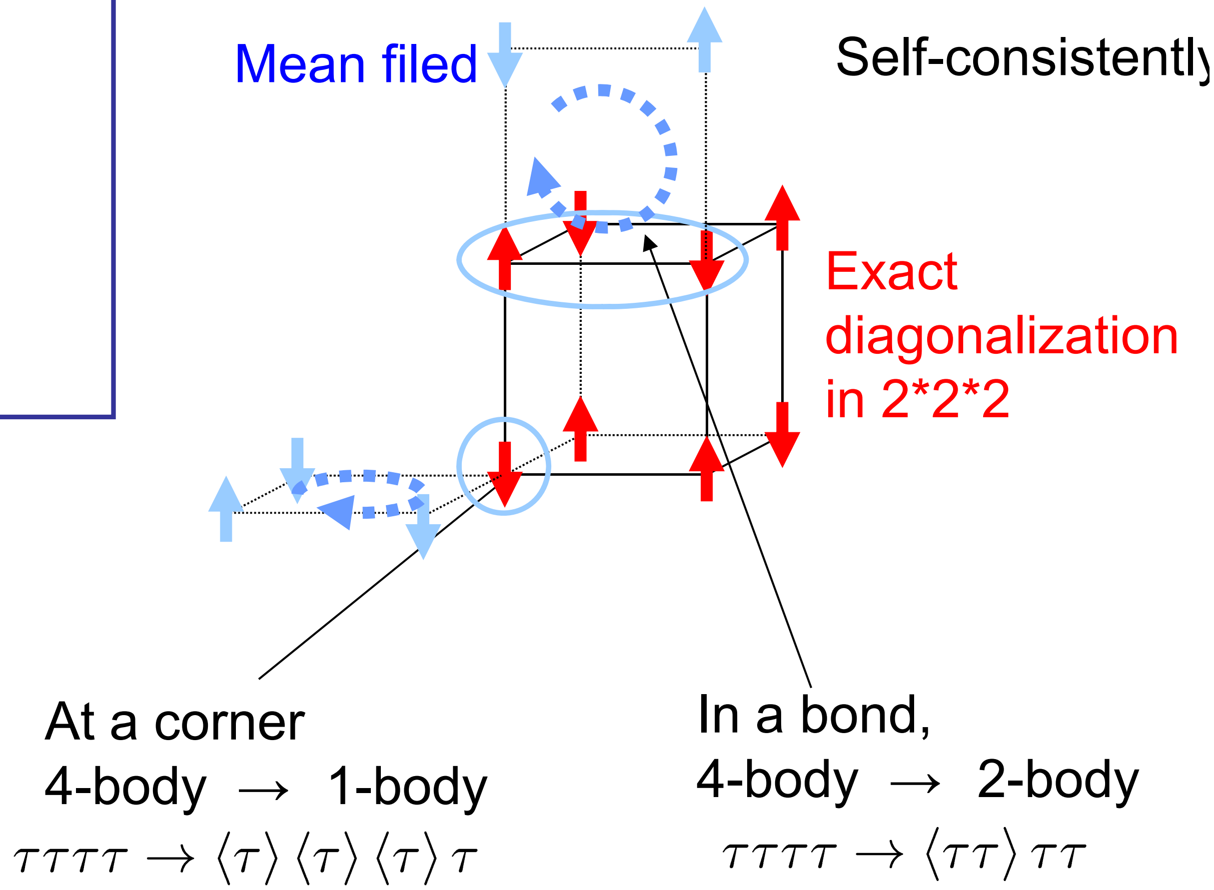
$$\frac{3}{4}K(T_i^z T_j^z T_k^y T_l^y + \cdots)$$

Antiferro Ferro

Quantum orbital state

- Extended Bethe method
- Exact diagonalization (Lanczos)
- Spin wave (zero-point energy)

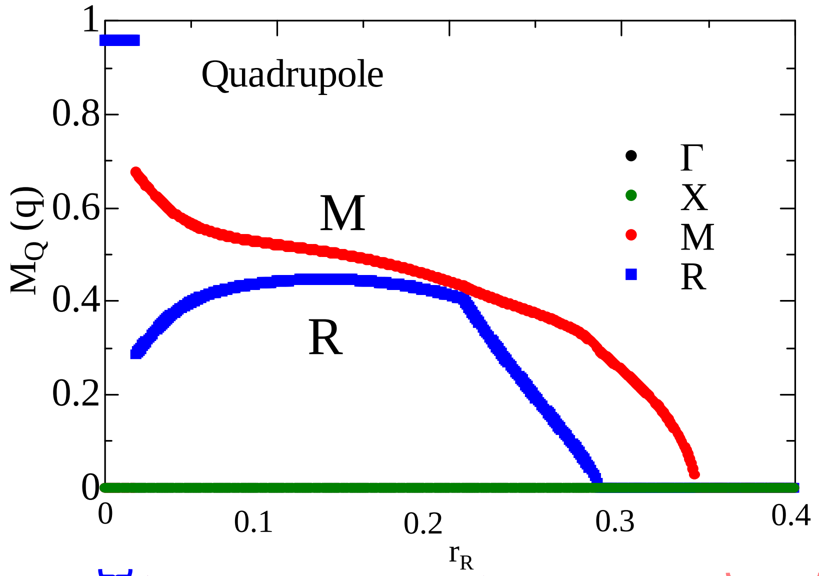
Extended Bethe approximation



Quantum orbital state

Extended Bethe method
(T=0)

Quadrupole (Tx,Tz) ordered moment

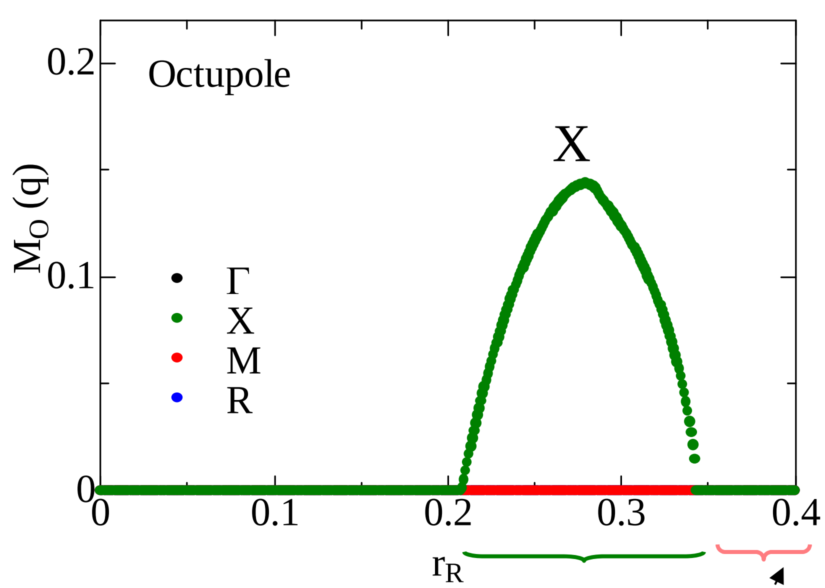


AF

Cant

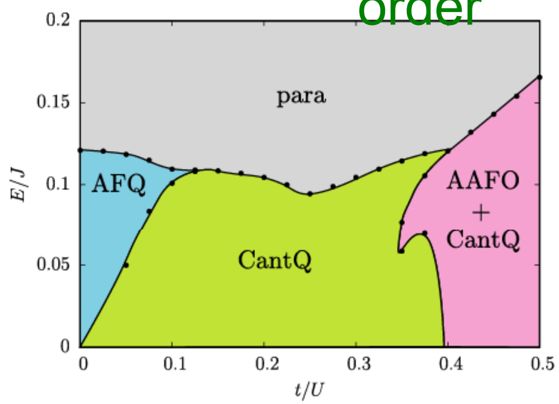
Octupole order

Octupole (Ty) ordered moment



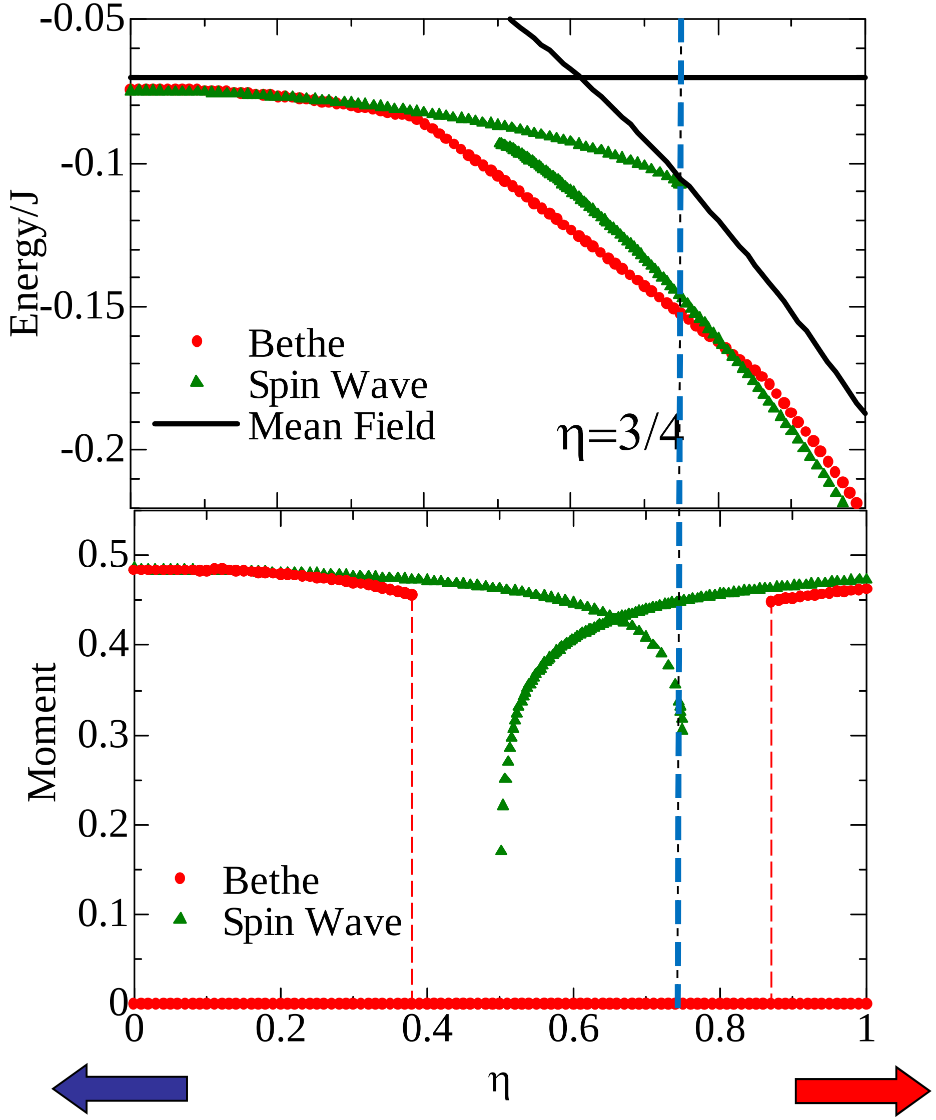
Octupole order

quantum para (?)



classical

Quantum orbital state



Quadrupole

Octupole

Only ring exchange term
(No NN exchange)

$$\mathcal{H}_R = K_R \sum_{[ijkl]_a} \frac{1}{2}\left(\tau_i^{a+}\tau_j^{a-}\tau_k^{a+}\tau_l^{a-} + H.c.\right)$$

$$\tau_i^{\pm a} = \tau_i^a \pm i(\sqrt{3}/2)T_i^y$$

T_i^x, T_i^z

Quadrupole Octupole

$$\tau_i^{\pm a}(\eta) = \tau_i^a \pm i\sqrt{\eta}T_i^y$$

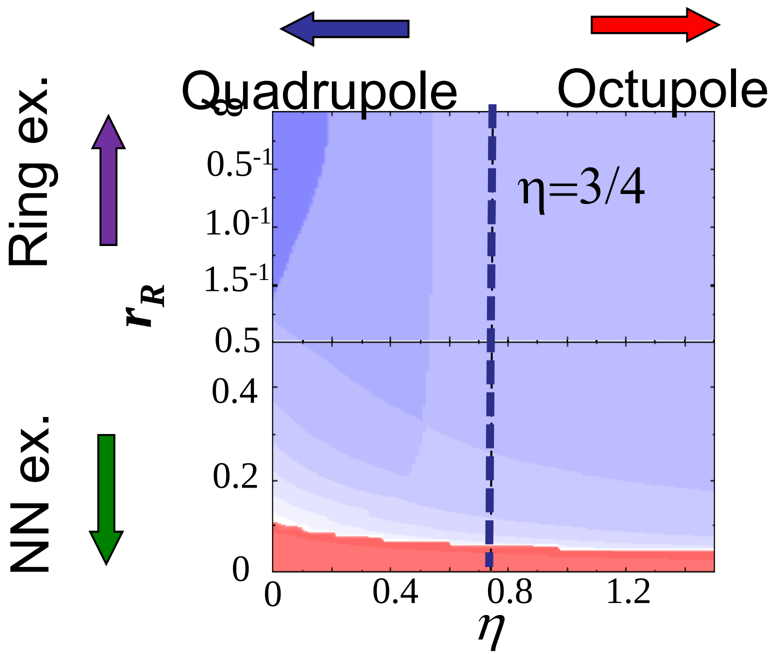
octupole
v.s.
quadrupole

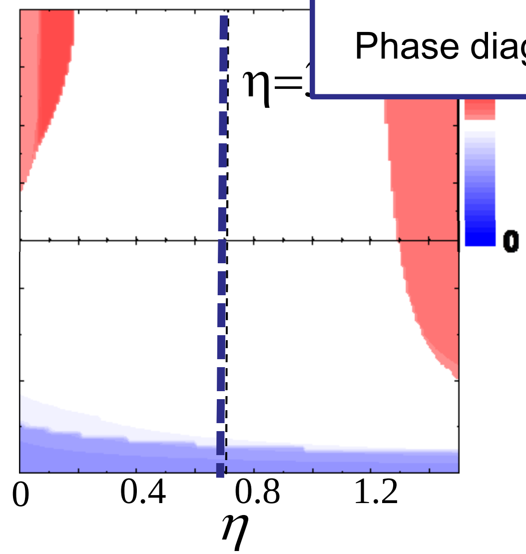
Suppression of ordered moment

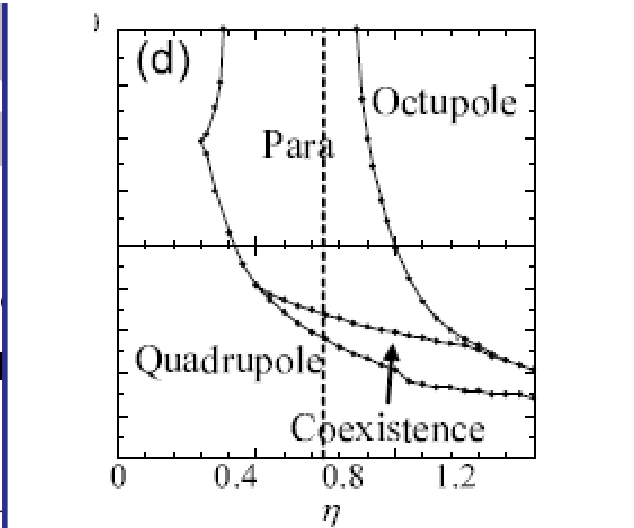
Phase diagram (quantun

2-body
correlation function

Plaquett
correlation fu







Phase diagram by Bethe method

Ring exchange v.s. NN exchange

2-body correlation func.

$$K_Q(\mathbf{q}) = 4N^2 \sum_{ij} \langle T_i^x T_j^x + T_i^z T_j^z \rangle e^{\mathbf{q}\cdot(\mathbf{r}_i - \mathbf{r}_j)}$$

Octupole v.s. Quadrupole

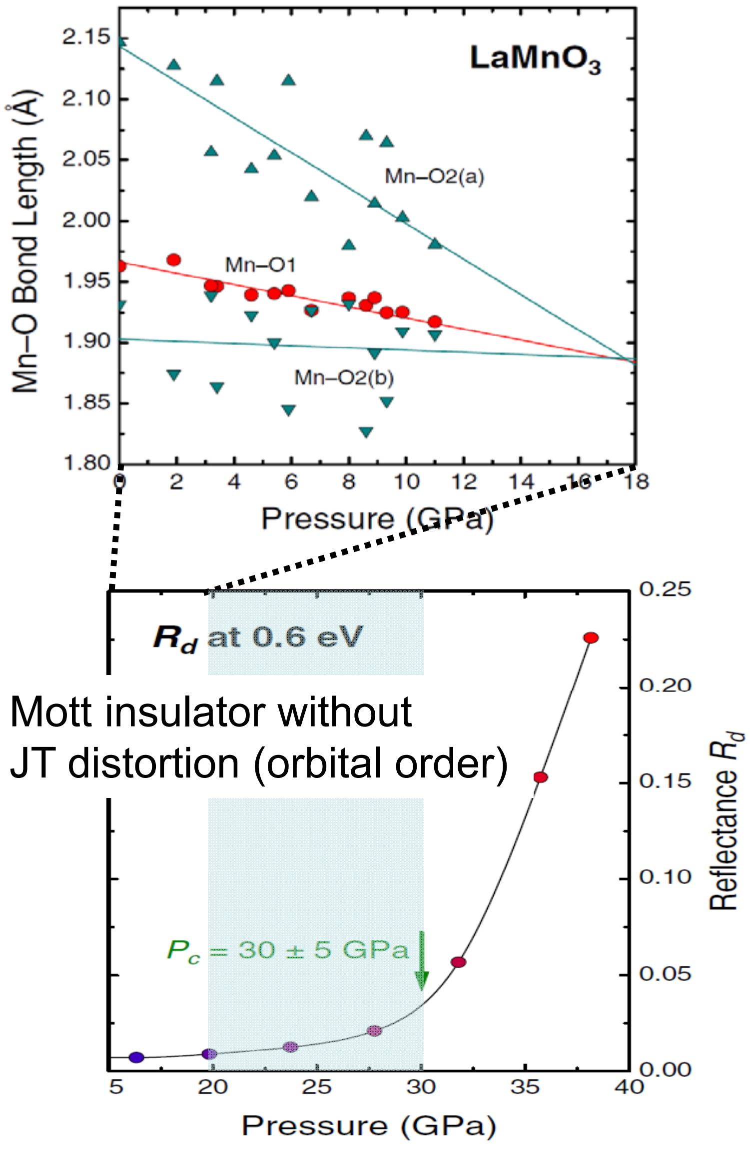
Plaquette 4-body correlation func.

$$P^\alpha = \frac{1}{6N} \sum_{[ijkl]} (1 - 16 \langle T_i^\alpha T_j^\alpha T_k^\alpha T_l^\alpha \rangle)$$

⇩

Suppression of ordered moment

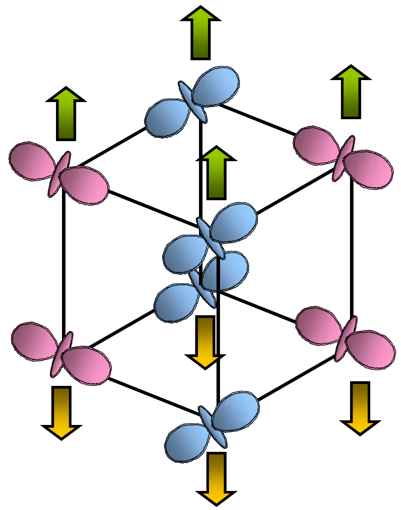
Implication for experiments



Mott insulator without
JT distortion (orbital order)

I. Loa, et.al., Phys. Rev. Lett. **87**, 125501 (2001)

$LaMnO_3$
(a parent of CMR material)



Orbital order with
JT distortion
T(OO) >> T_N

Pressure effect
Enhancement of Ring Ex.

Orbital 120 mode

on a honeycomb lattice

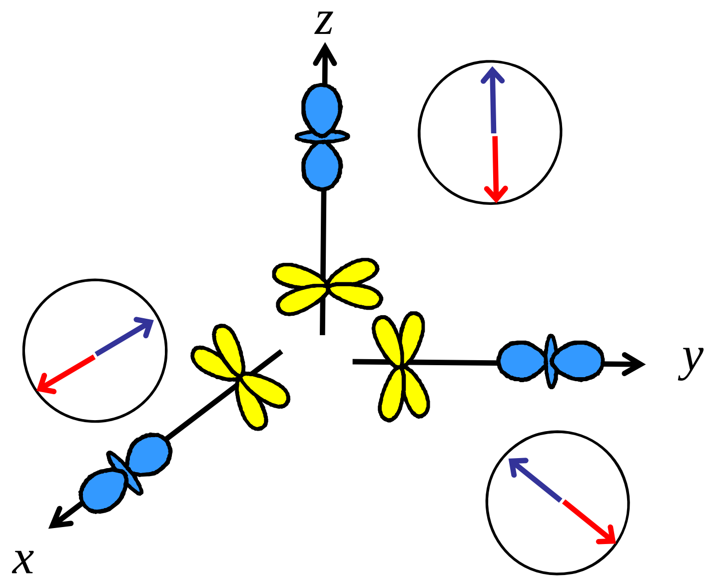
120° model in a cubic lattice

$$\mathcal{H} = 2J \sum_{\langle ij \rangle} \tau_i^l \tau_j^l$$

$$\tau_i^l = \cos\left(\frac{2\pi}{3} n_l\right) T_{iz} + \sin\left(\frac{2\pi}{3} n_l\right) T_{ix},$$

$l(= x, y, z)$: bond direction

$$(n_x, n_y, n_z) = (1, 2, 3)$$



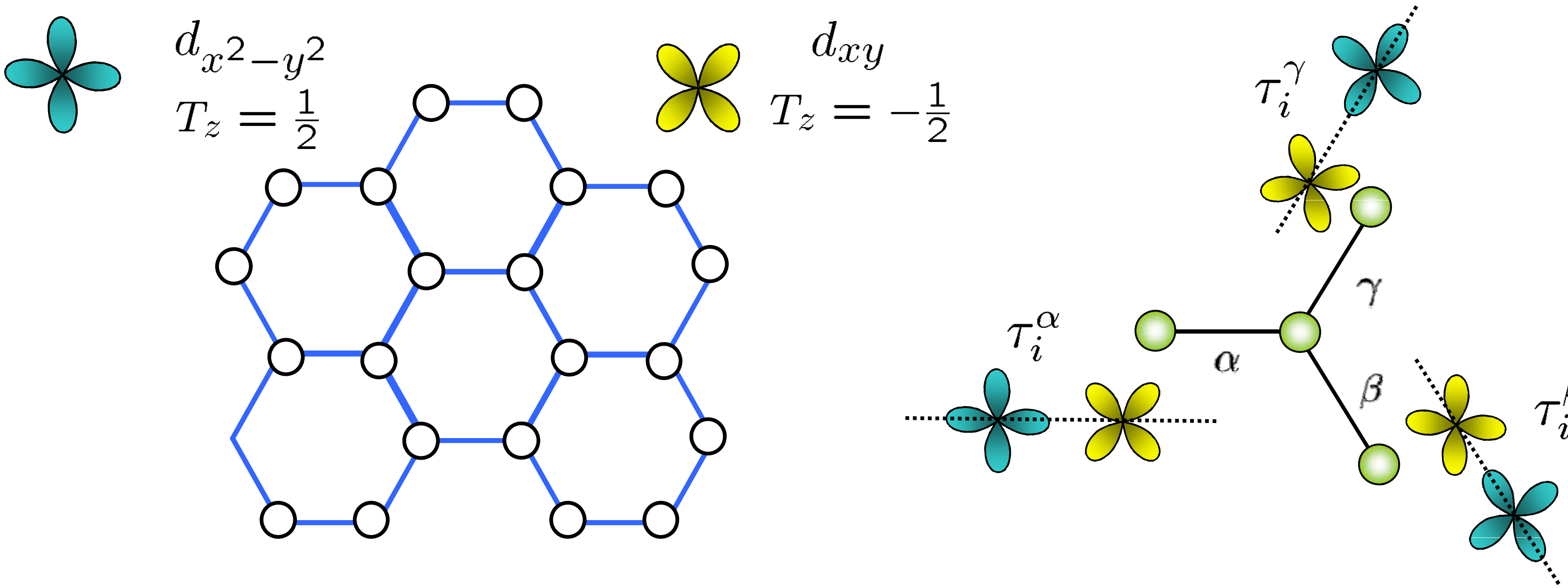
$$\begin{cases} T_{iz}T_{jz} & l = z \\ \left[-\frac{1}{2}T_{iz} + \frac{\sqrt{3}}{2}T_{ix}\right]\left[-\frac{1}{2}T_{jz} + \frac{\sqrt{3}}{2}T_{jx}\right] & l = x \\ \left[-\frac{1}{2}T_{iz} - \frac{\sqrt{3}}{2}T_{ix}\right]\left[-\frac{1}{2}T_{jz} - \frac{\sqrt{3}}{2}T_{jx}\right] & l = y \end{cases}$$

A kind of frustration

Interaction explicitly depends on bond direction

Honeycomb lattice 120 degree model

Doubly-degenerate orbital on a honeycomb lattice



$$\mathcal{H} = \frac{1}{2} J \sum_i \left(\frac{3}{4} + \tau_i^\alpha \tau_{i+\delta_\alpha}^\alpha + \tau_i^\beta \tau_{i+\delta_\beta}^\beta + \tau_i^\gamma \tau_{i+\delta_\gamma}^\gamma \right)$$

$J < 0$

$$\tau_i^l = \cos\left(\frac{2n_l\pi}{3} + \frac{\pi}{2}\right) T_i^z + \sin\left(\frac{2n_l\pi}{3} + \frac{\pi}{2}\right) T_i^x$$

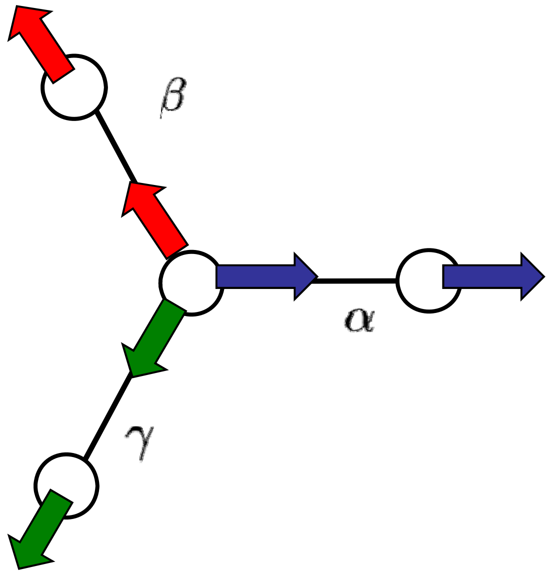
$$(n_\alpha, n_\beta, n_\gamma) = (1, 2, 3)$$

Honeycomb lattice orbital model

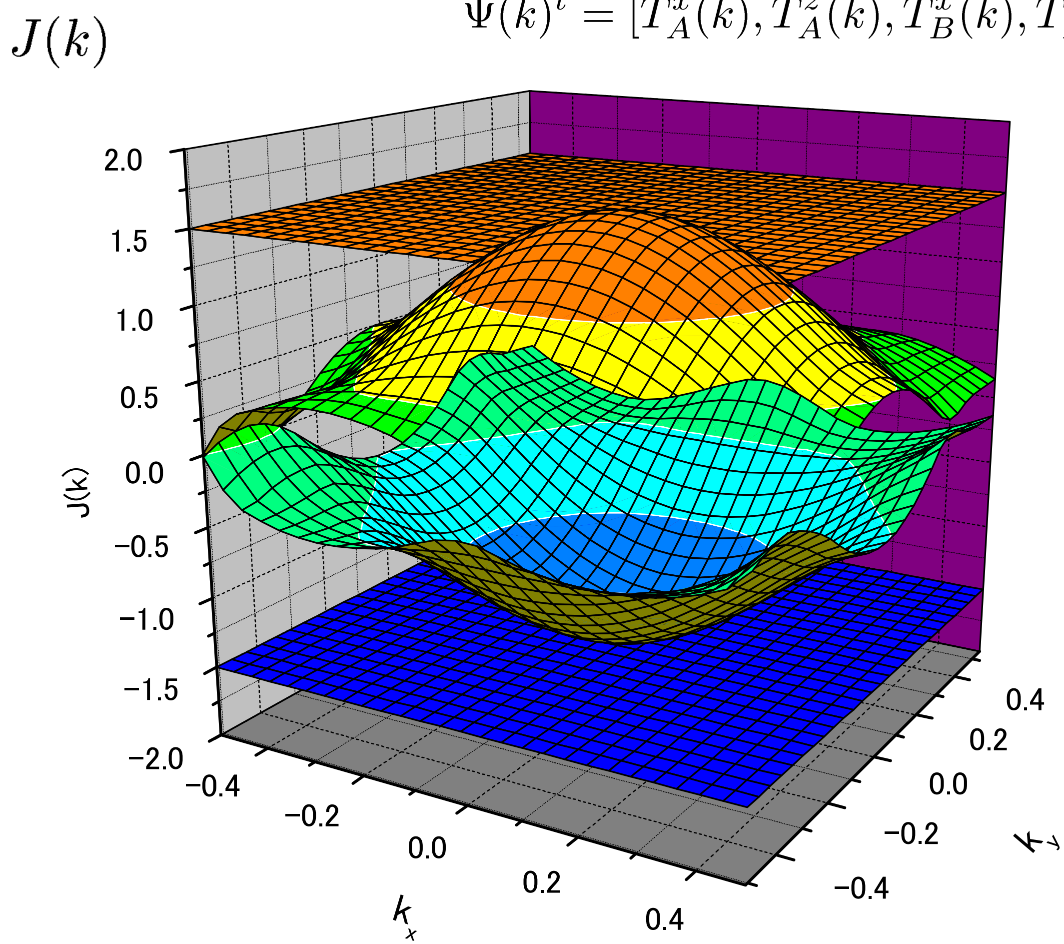
Interaction in momentum space

$$\mathcal{H} = \frac{J}{2}\sum_{\vec{k}} \Psi(\vec{k})^t \hat{J}(\vec{k}) \Psi(\vec{k})$$

$$\Psi(\vec{k})^t = [T_A^x(\vec{k}), T_A^z(\vec{k}), T_B^x(\vec{k}), T_B^z(\vec{k})]$$



An intrinsic frustration
for orbital pseudo-spin

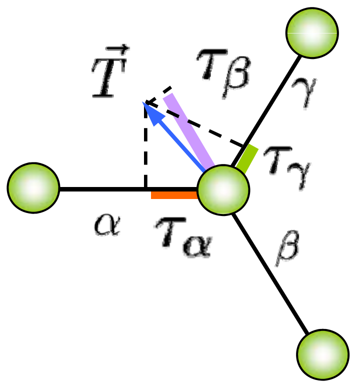


Flat dispersion

Classical ground state

$$\mathcal{H} = \frac{J}{2} \sum_{i \in A, l} \left(\tau_i^l - \tau_{i+\hat{l}}^l \right)^2 - \frac{3}{16} JN$$

τ_i^l : a projection component along l



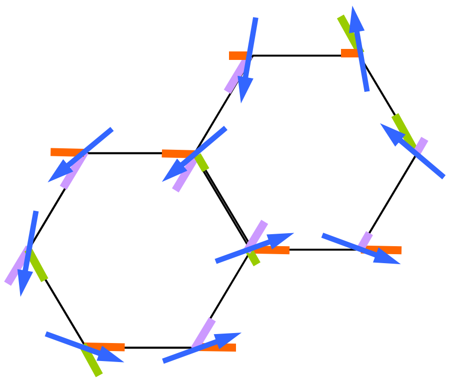
"τ=τ rule"

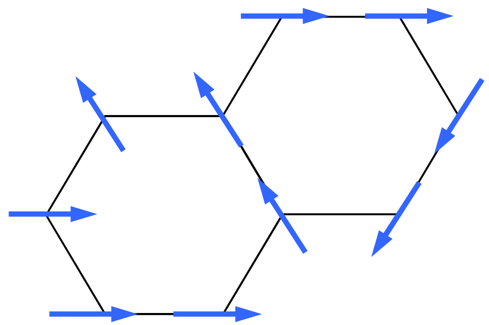
Classical ground states:

$$\tau_i^l = \tau_j^l$$

for all NN bonds

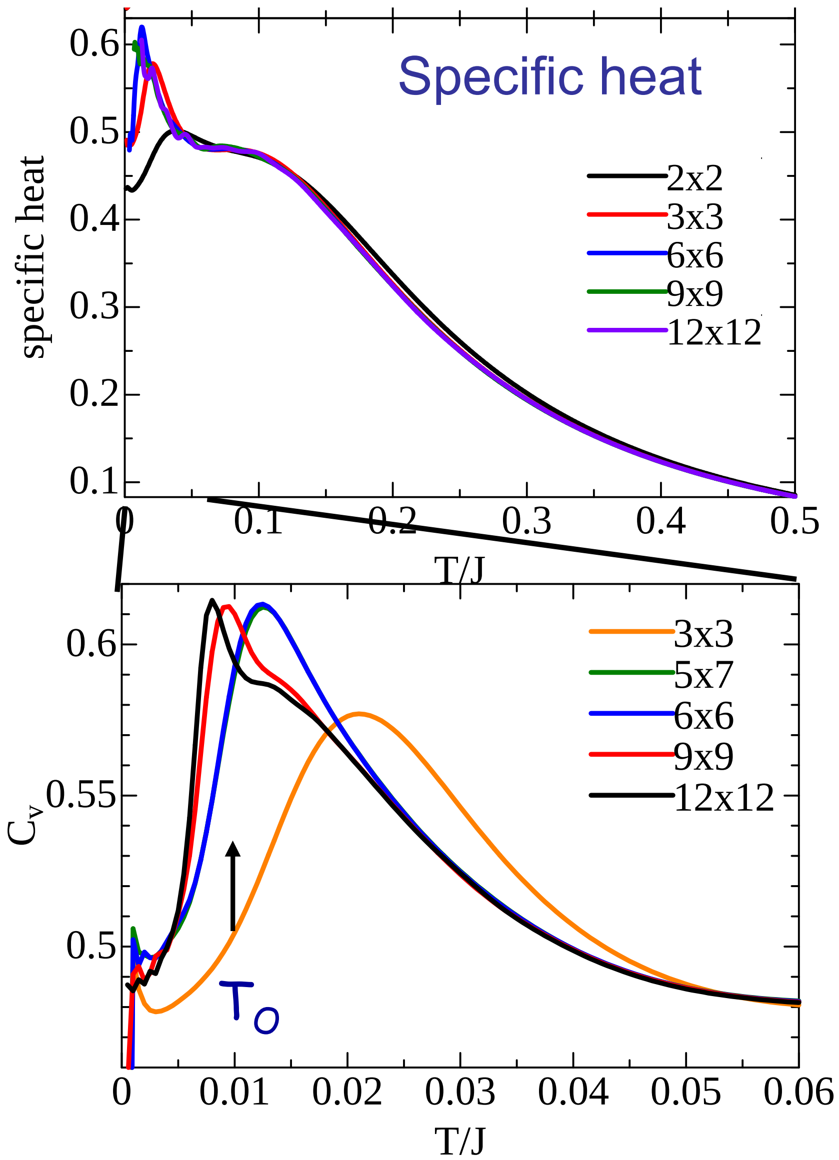
A macroscopic number of degeneracy in classical configurations





. . .

Classical state at finite T



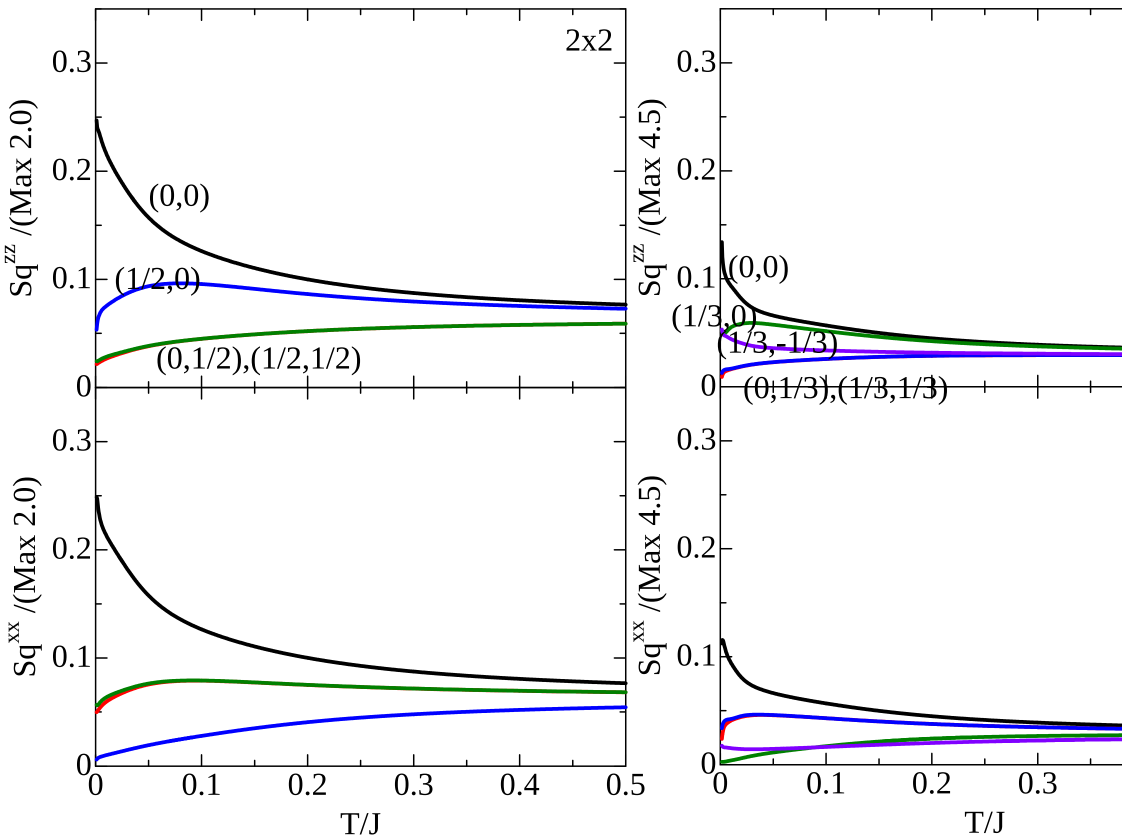
Multi-Canonical MC
(Classical)
Orbital Model
N=2×2 − 12×12

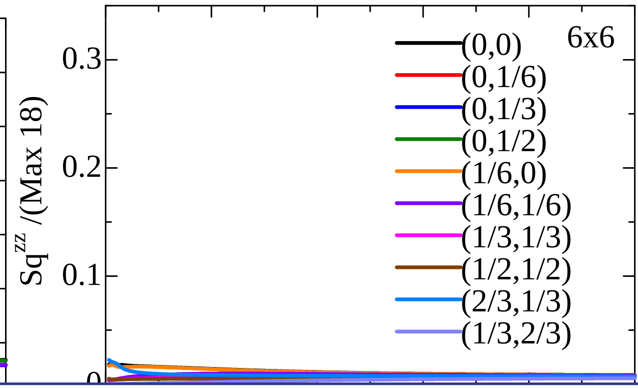
A peak in specific heat
at very low $T << T_{MF}=3J/4$

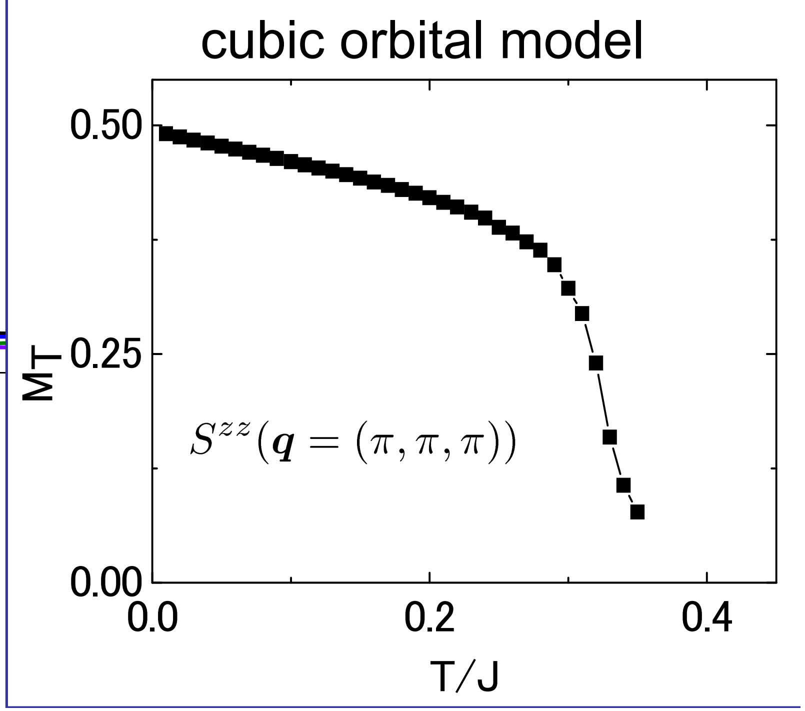
Correlation function (classical)

Orbital correlation function

$$S^{zz}(\boldsymbol{q}) = \frac{1}{N}\sum_{ij}\langle T_i^z T_j^z\rangle e^{i\boldsymbol{q}(\boldsymbol{r}_i-\boldsymbol{r}_j)}$$



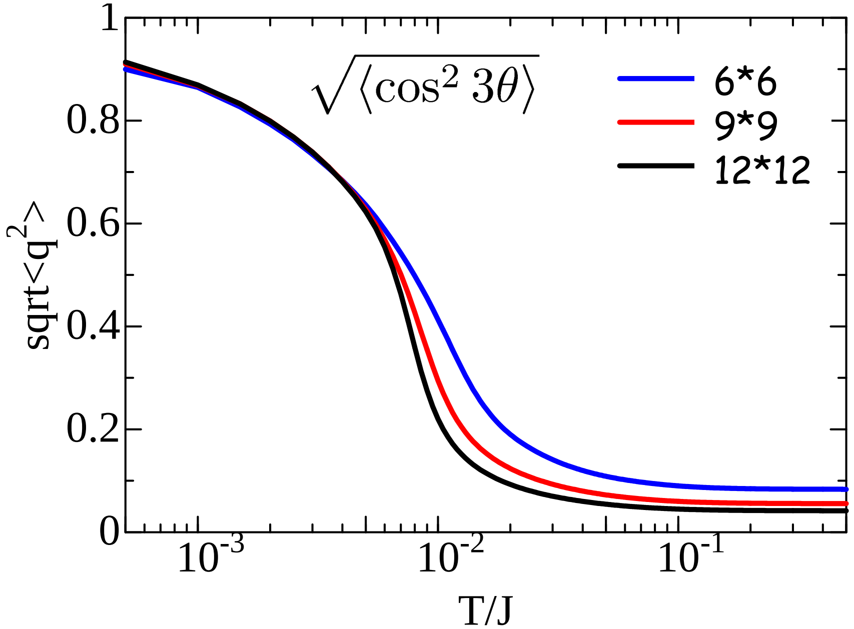


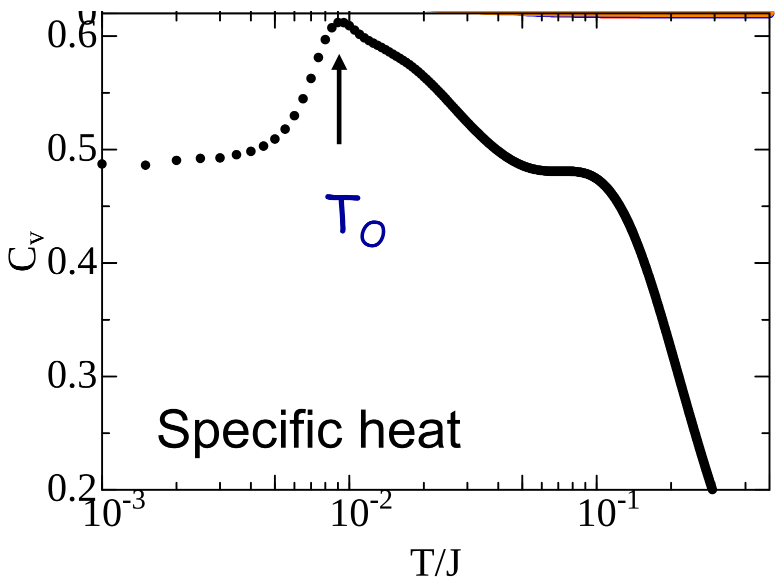


No order by thermal fluctuation

Also No KT transition, No directional order

A kind of angle order





Multi-Canonical MC (Classical)

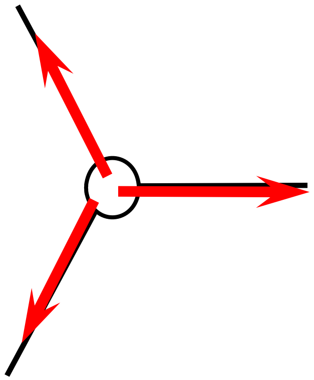
Order Parameter (?) $\langle\cos 3\theta\rangle$

$$\langle\cos^2 3\theta\rangle = \sqrt{\langle\left(\frac{1}{N}\sum_i \cos 3\theta_i\right)^2\rangle}$$

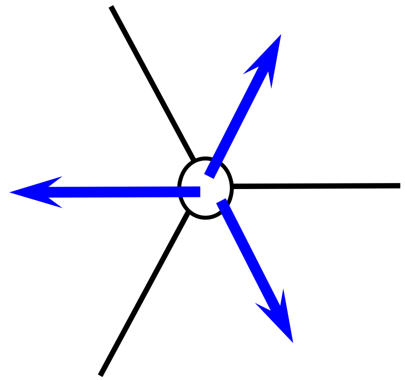
Orbital angle is fixed at $\cos 3\theta = 1$ or $\cos 3\theta = -1$ below T_O

A kind of angle order

$\langle \cos 3\theta \rangle$



OR



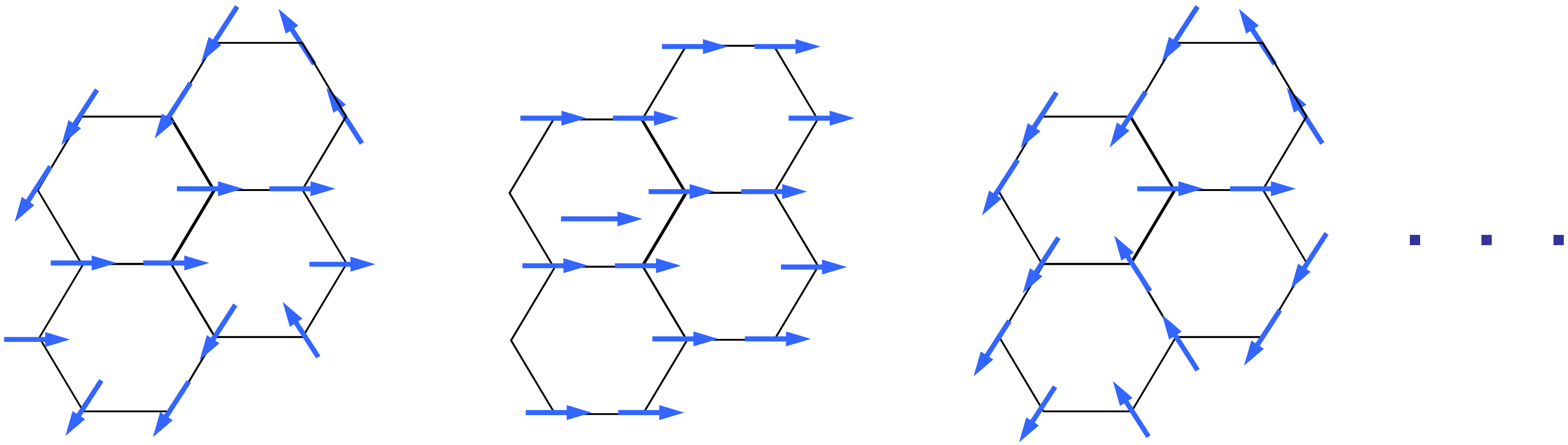
$\cos 3\theta = 1$ at all sites

$\theta = 0 \text{ or} 2\pi/3 \text{ or } 4\pi/3$

$\cos 3\theta = -1$ at all sites

$\theta = \pi \text{ or} 1\pi/3 \text{ or } 5\pi/3$

$\cos 3\theta = 1$



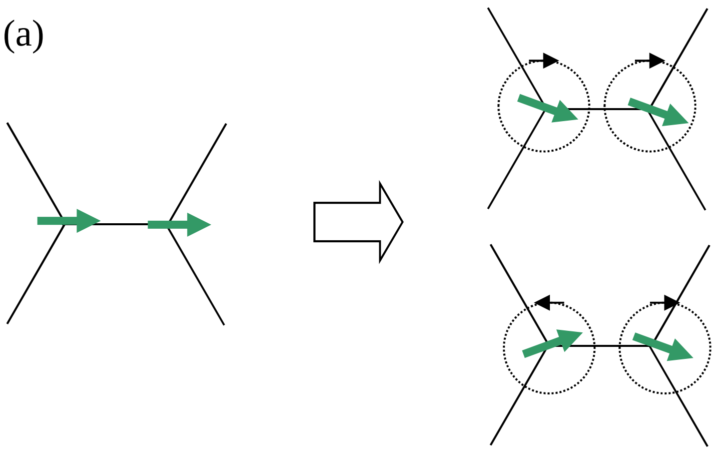
A macroscopic number of degenerate configurations still remain

Why cos3θ=+1 / -1 ?

Fluctuation with keeping
a condition $\tau_i^l = \tau_j^l$ rule

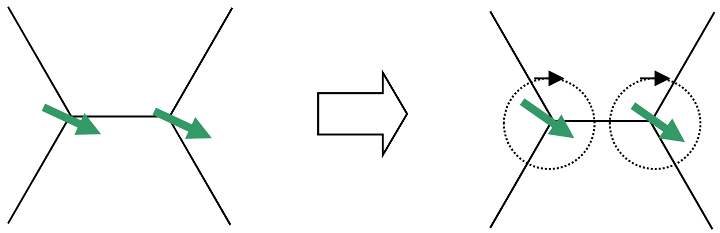
$\cos 3\theta = \pm 1$

(a)



$\cos 3\theta \neq \pm 1$

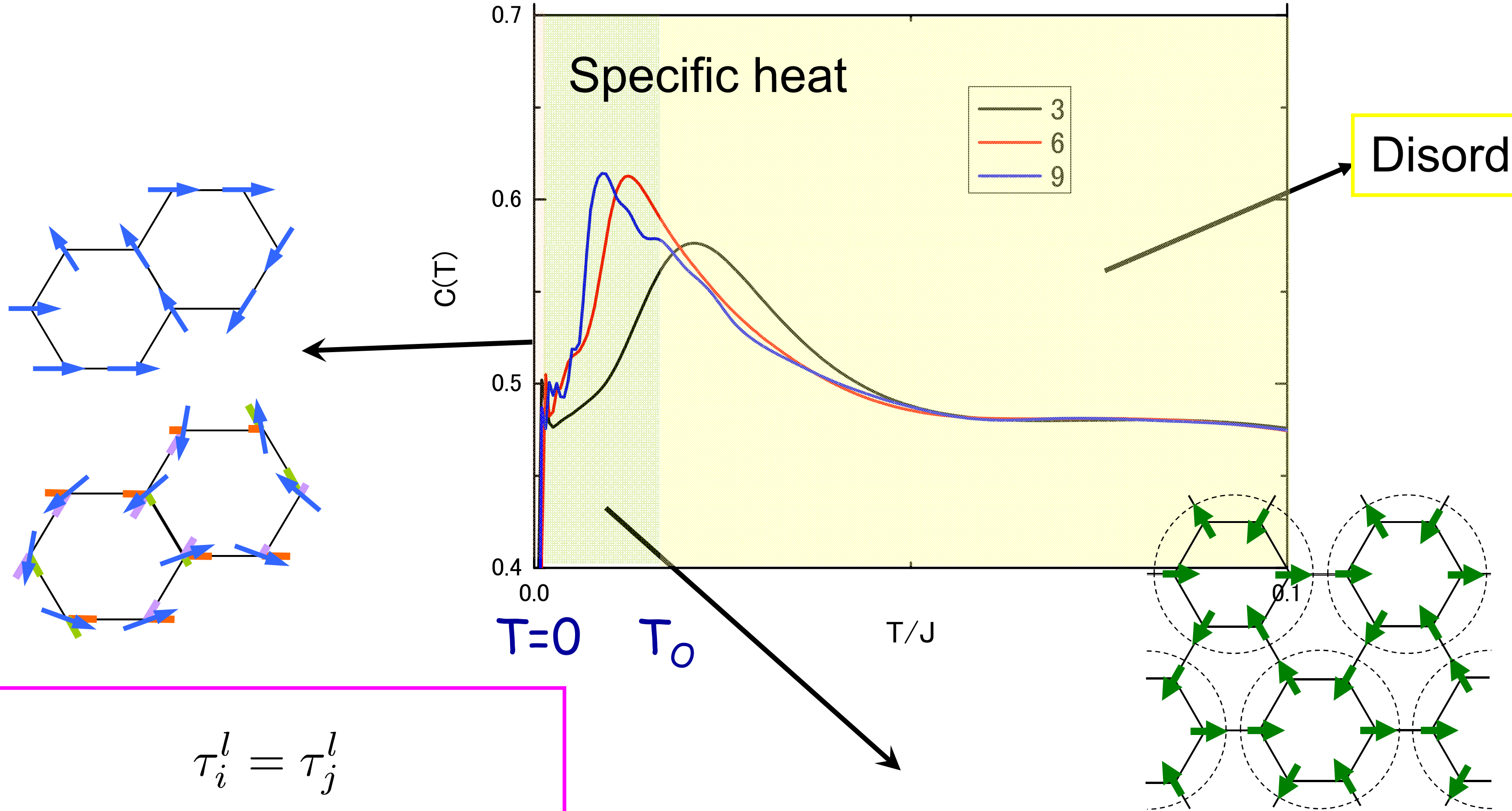
(b)



low-lying fluctuation around
$\cos 3\theta = \pm 1$

⇒ entropy gain in finite T

Classical phase diagram



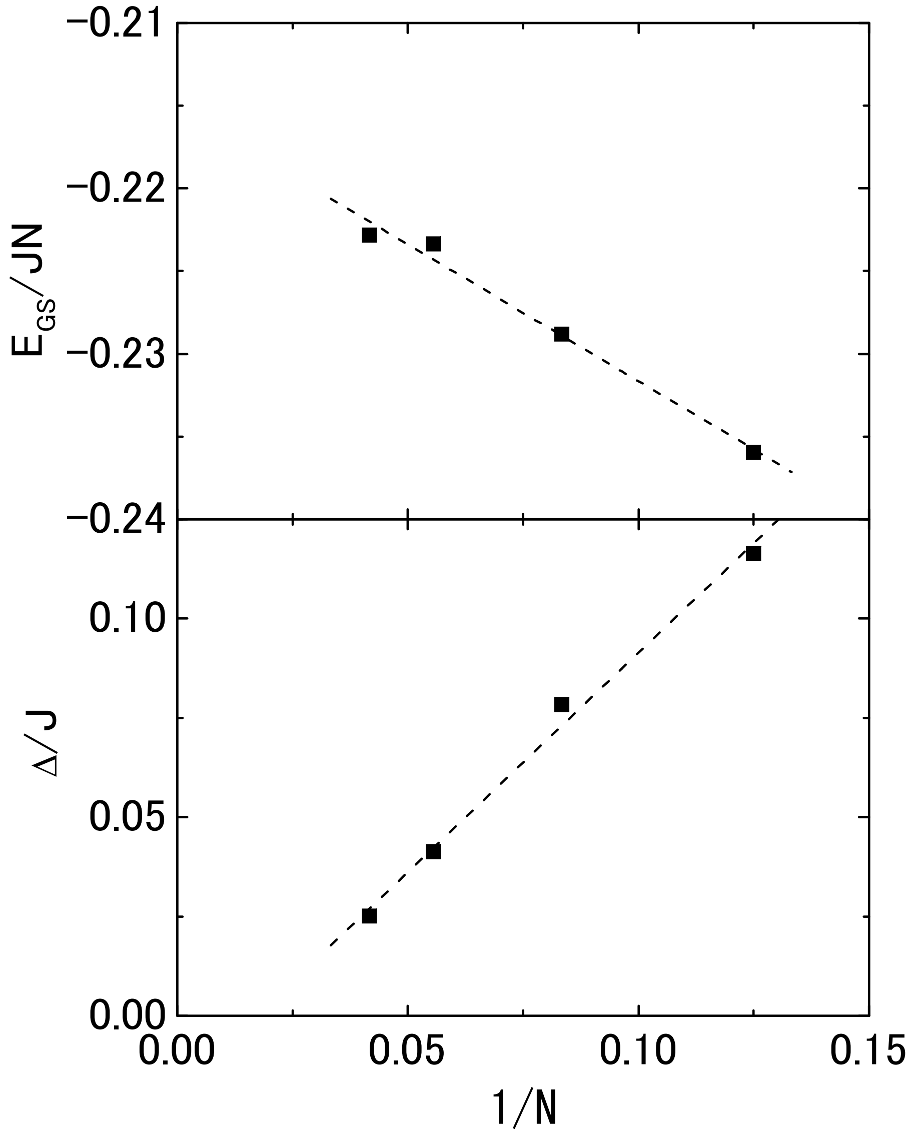
$\tau_i^l = \tau_j^l$

large degenerate states

$\cos 3\theta = \pm 1$ for all sites

still large degeneracy

Quantum state at T=0



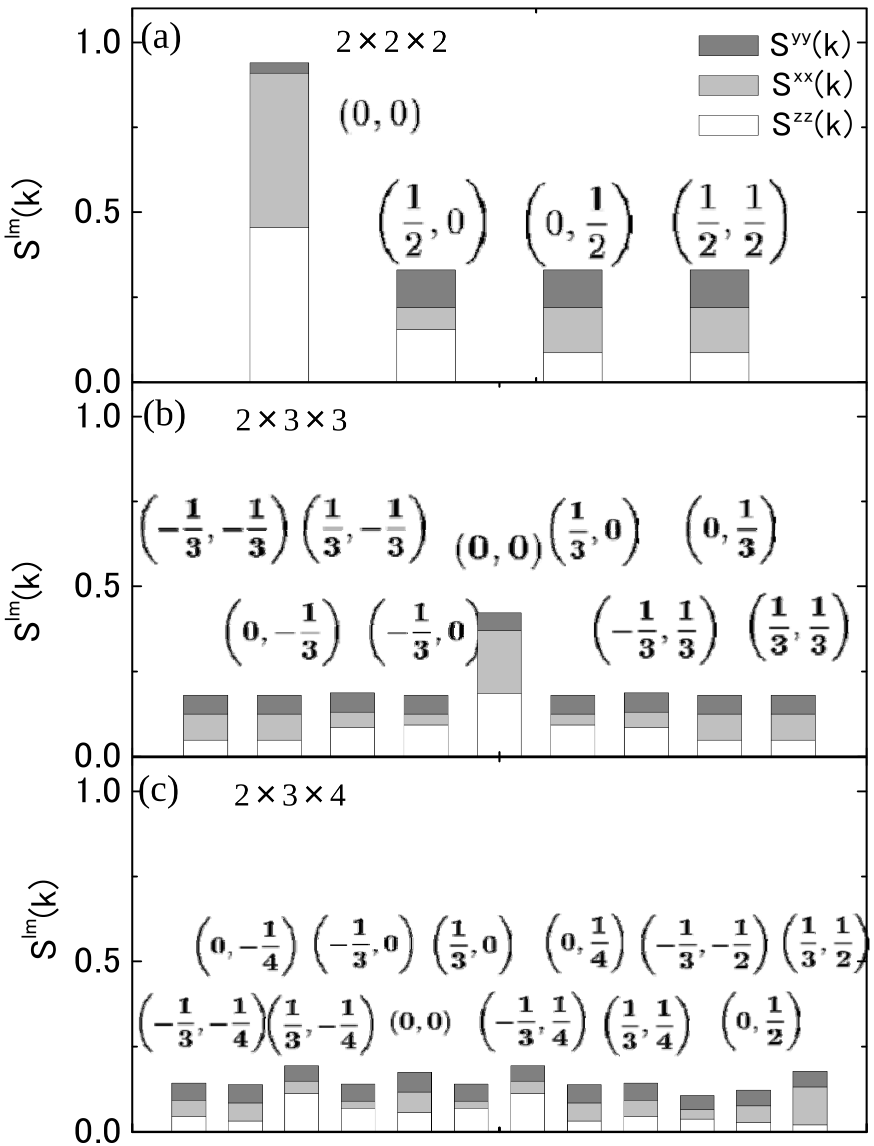
Lanczos method

2×2×2、2×2×3、
2×3×3、2×3×4
site clusters

$$\Delta = E(1st) - E(GS)$$

Gapless or degenerate state

Quantum state at T=0



Exact diagonalization
in finite cluster
by
Lanczos method

$$S^{zz}(\boldsymbol{q}) = \frac{1}{N}\sum_{ij} T_i^z T_j^z e^{i\boldsymbol{q}(\boldsymbol{r}_i - \boldsymbol{r}_j)}$$

No possibility of conventional order by quantum fluctuation

Variational approach

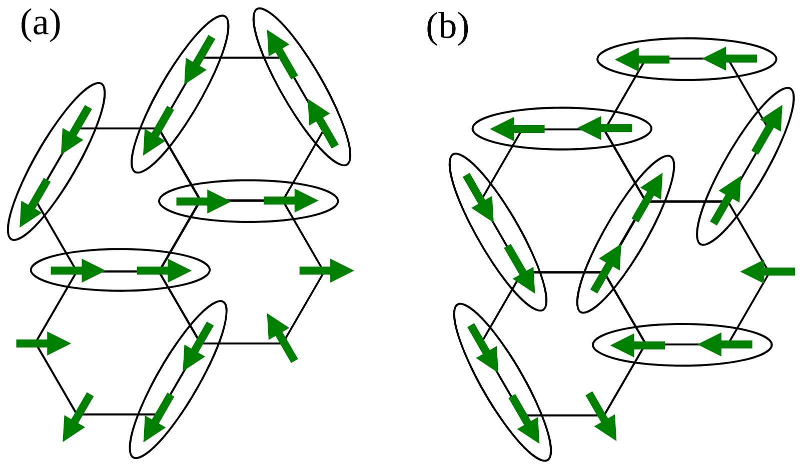
Honeycomb lattice is covered by NN bonds
with the minimum bond energy

trial wave function

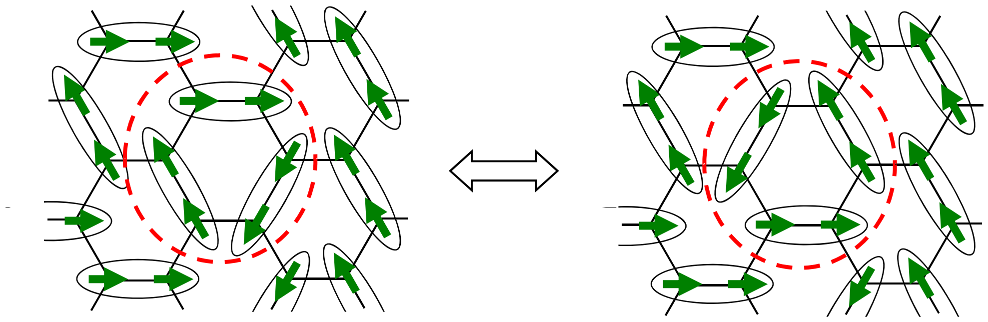
$$|\Psi^{(+)}\rangle = \mathcal{N}\sum_{l}\mathcal{A}_l\left\{|\psi_l^{(\uparrow)}\rangle + |\psi_l^{(\downarrow)}\rangle\right\},$$

$$|\psi_l^{(\uparrow)}\rangle = \prod_{\langle ij\rangle_l} U(\phi_\eta)_{\langle ij\rangle_l}|\uparrow\cdots\uparrow\rangle.$$

$$U(\phi_\eta)_{\langle ij\rangle_l} = \exp\left[-i\phi_\eta\left(T_i^y + T_j^y\right)\right],$$



Quantum Resonance

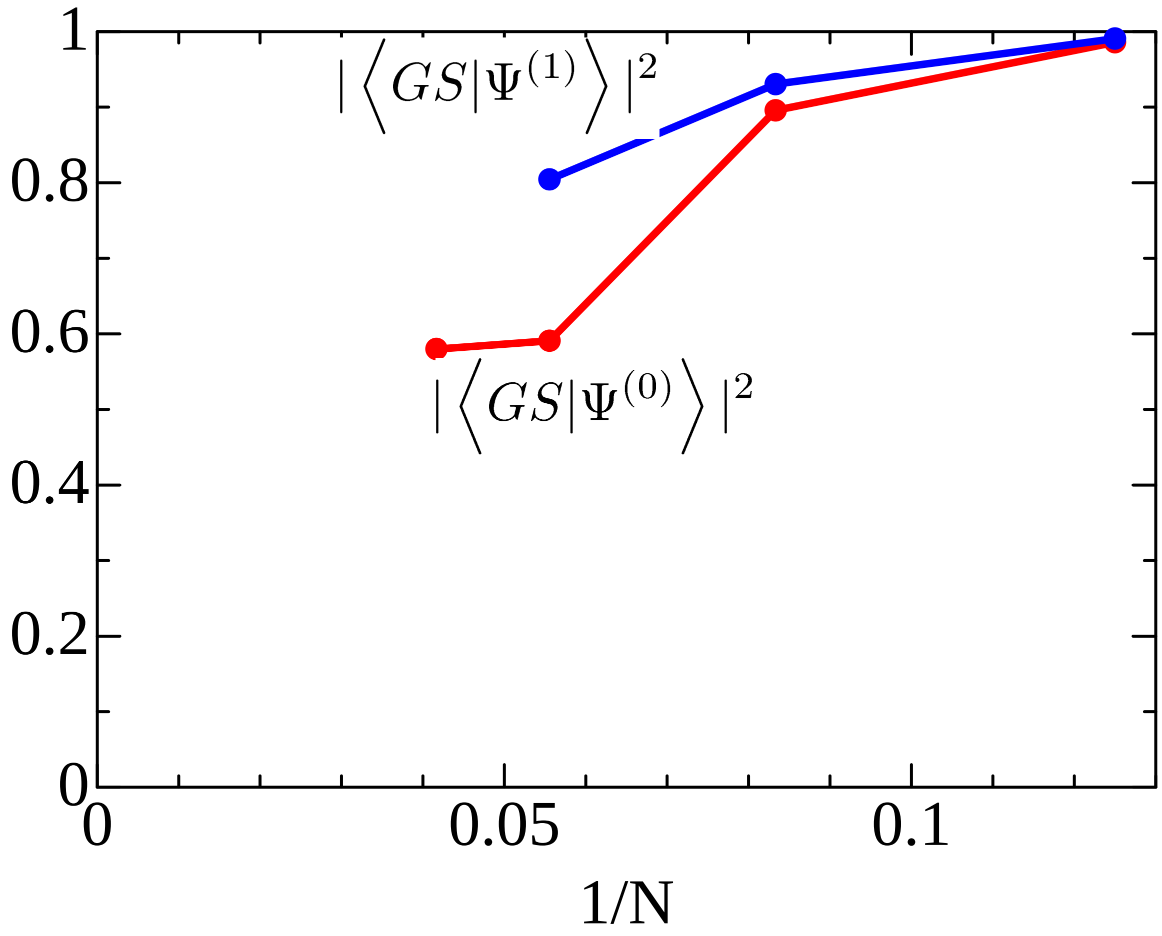


Resonance energy:

～10% of energy gain
of quantum effect

Variational approach

Overlap between the trial w.f. and GS w.f. by Lanczos



$\left|\Psi^{(0)}\right\rangle$ for $\mathcal{A}_l \equiv 1$

$\left|\Psi^{(1)}\right\rangle$ $\mathcal{A}_l$ is optimized

Dynamical JT effect on

Honeycomb lattice spin-orbital model

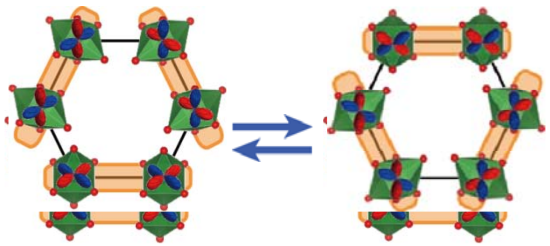
Quantum Spin Liquid State

No long range magnetic order
down to low temperatures

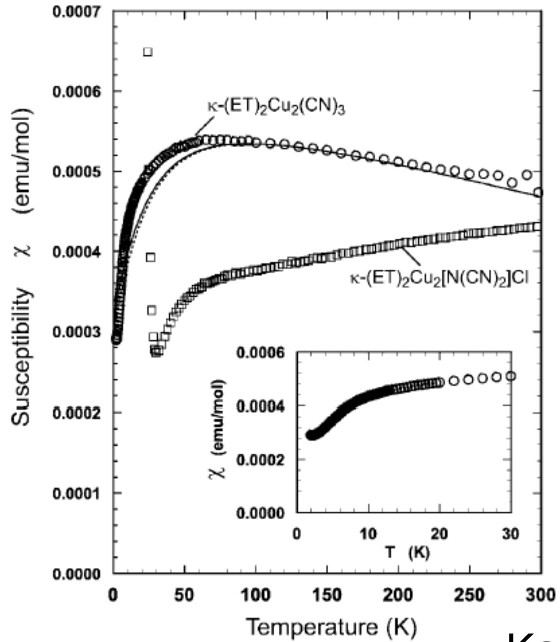
- One-dimensional spin chain
  No LRO at finite Temperature
  S=1/2   S=1(Haldane)

- Geometrical Frustration
  e.g. 2dim. triangular lattice

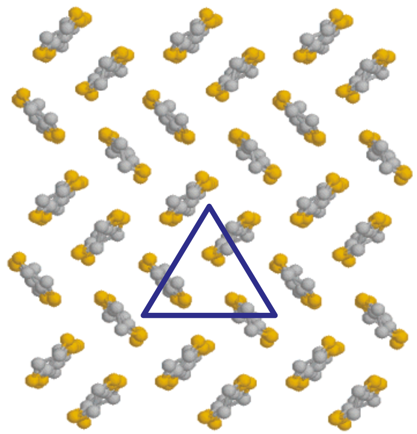
- A possibility of spin liquid in Spin-Orbital system
  with Dynamical Jahn-Teller effect



κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$



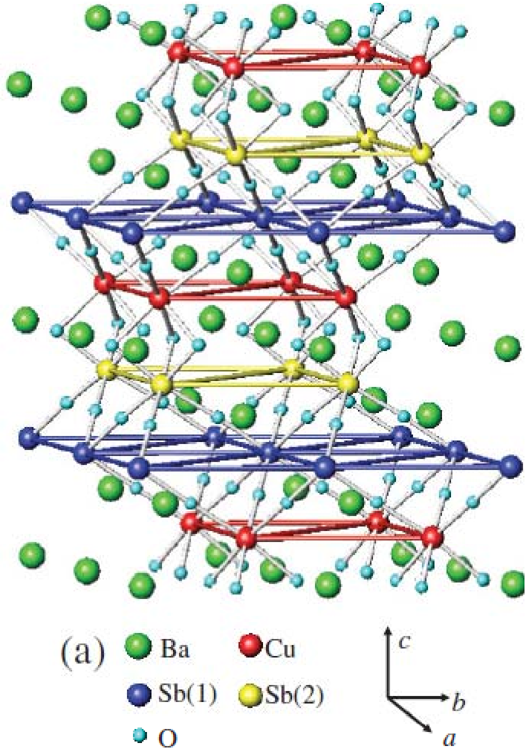
Kanoda Gr.

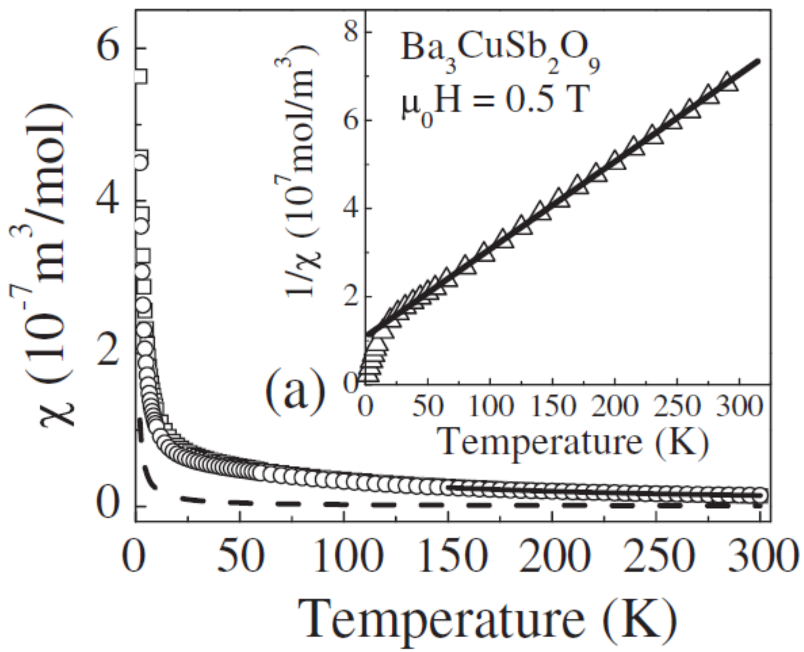


$Ba_3CuSb_2O_9$

Spin liquid state in the *S*=1/2 triangular lattice $Ba_3CuSb_2O_9$

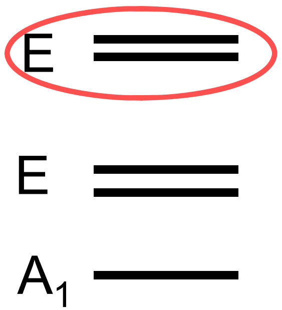
No LRO T>0.2K





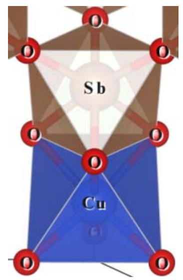
H. D. Zhou et al. PRL106, 147204 (2011)

Orbital degeneracy



$Cu^{2+}(d^9)$

C_{3v}

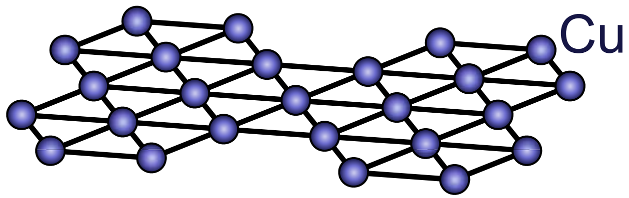


Nakatsuji et al.
Science 336, 559 (2012)

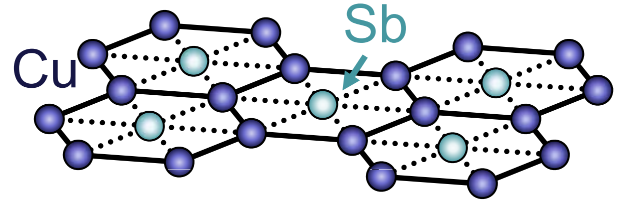
$Ba_3CuSb_2O_9$

Nakatsuji, Sawa, Hagiwara, Wakabayashi et al. Science 336, 559 (2012)

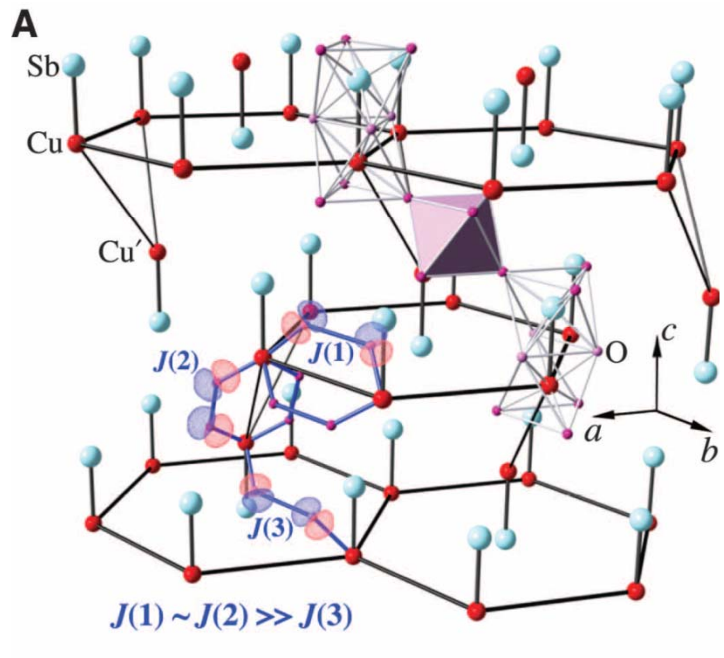
triangle



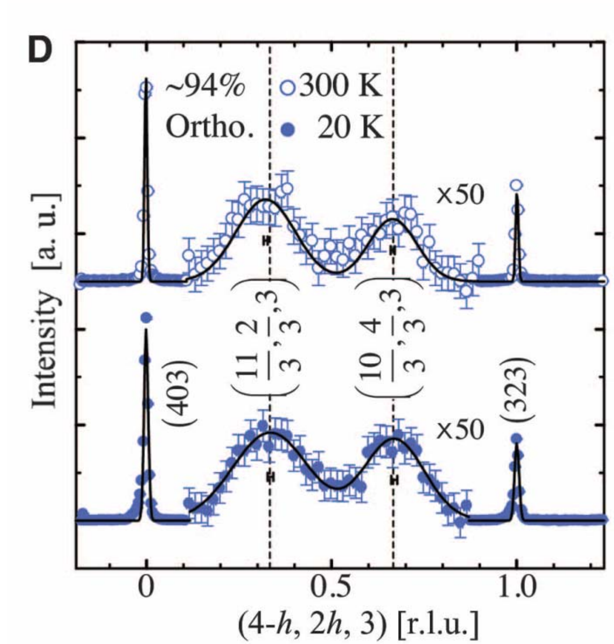
honeycomb



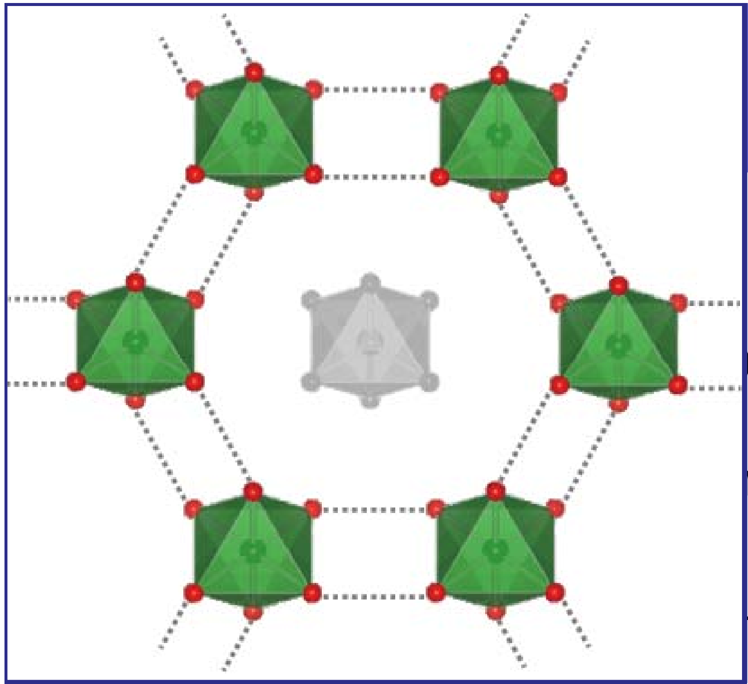
(Spin liquid w/o frustration ?)



X-ray diffraction

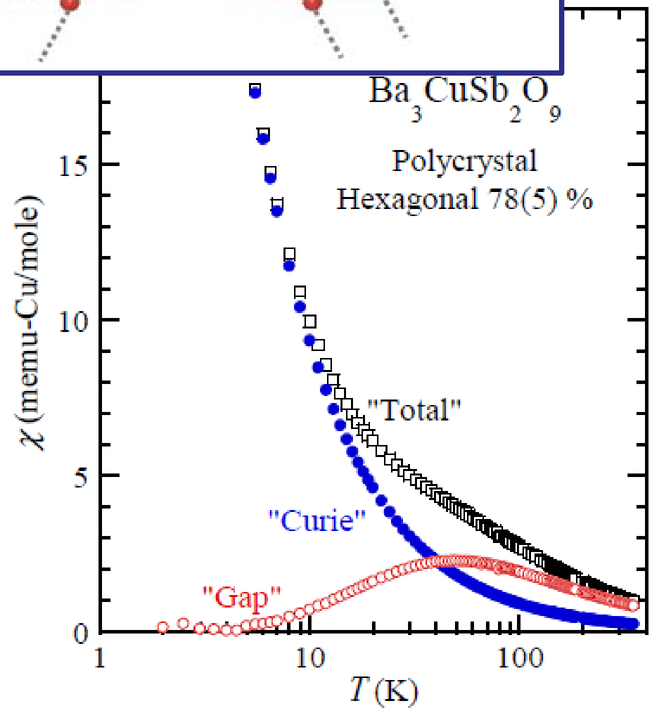


$Ba_3CuSb_2O_9$



)mK

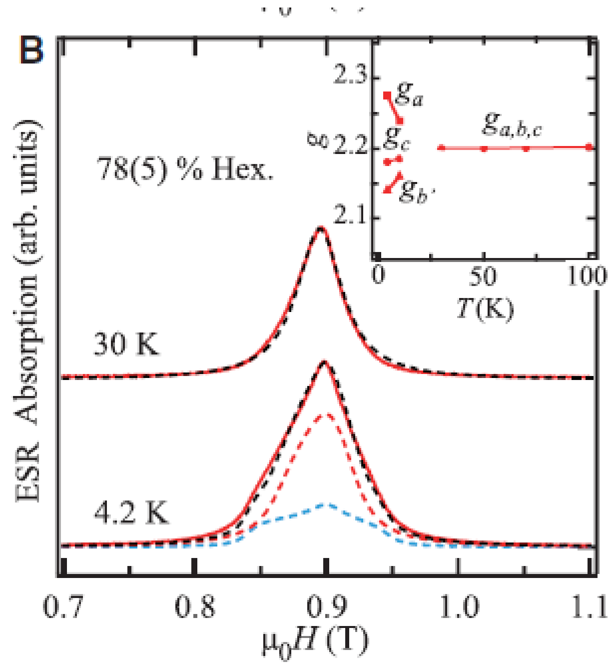
ent (~50K)



ESR

Nearly isotropic g-factor

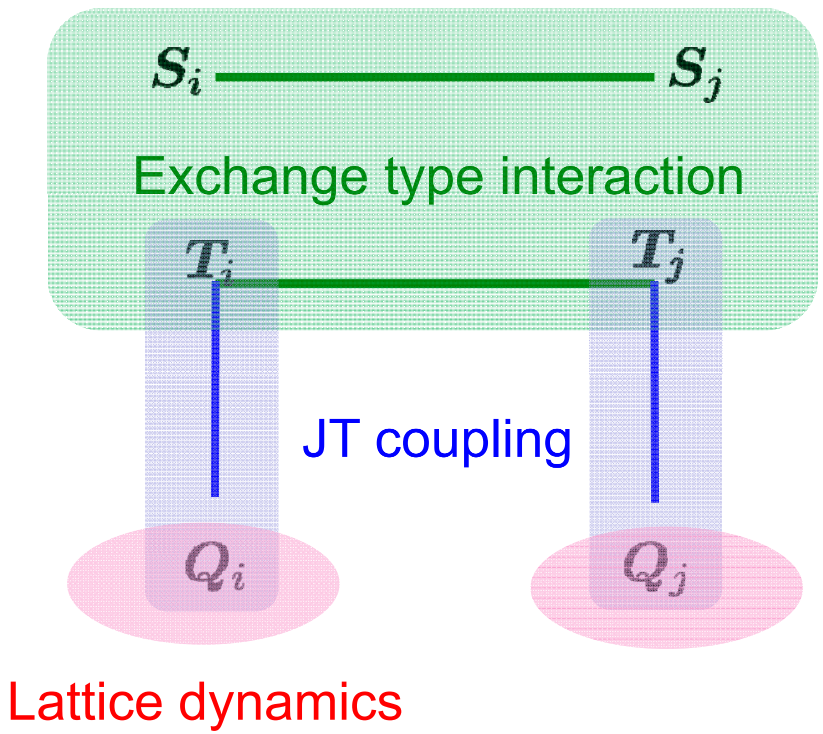
No/weak static JT distortion
(Dynamical JT ?)



But.
orbital freezing in EXAFS

Nakatsuji, Sawa, Hagiwara, Wakabayashi et al. Science 336, 559 (2012)

Orbital –Spin + Dynamical JT system

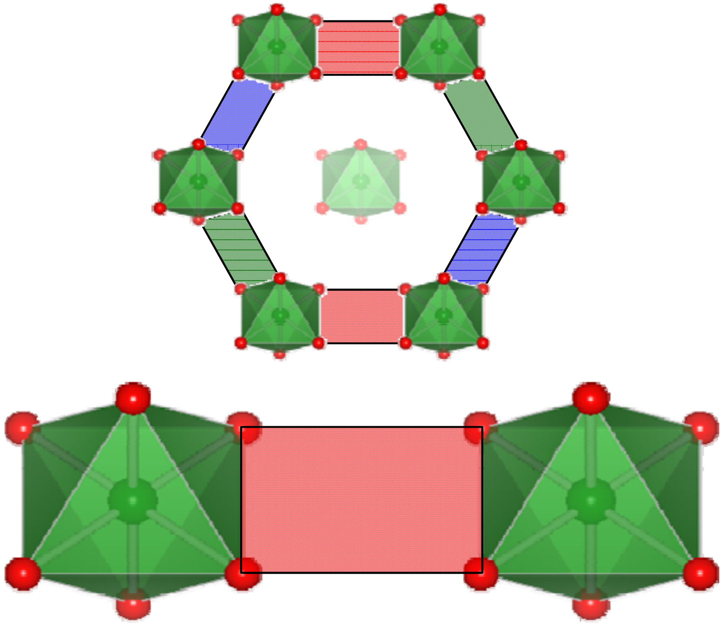


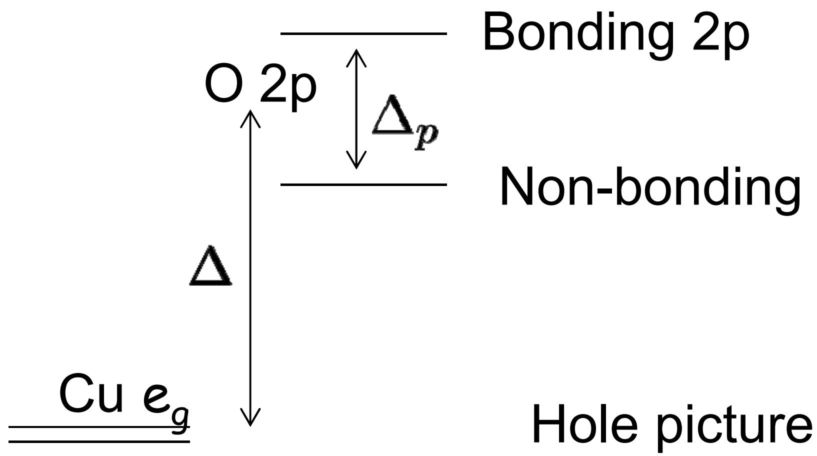
✓ Inter-site Exchange Interaction
v.s.
Local Dynamical JT Effect

✓ A possible scenario for spin liquid in $Ba_3CuSb_2O_9$

$$H = H_{\mathbf{exch}} + H_{\mathbf{JT}}$$

Spin-orbital Superexchange





dp type model Hamiltonian

$$H = \sum_{<ij>} d_i^\dagger p_j + h.c. + \Delta \sum n_p + U_d \sum n_\uparrow^d n_\downarrow^d + U', J, J' + \cdots$$

Perturabation for electron transfer

⇨ Kugel-Khomskii type Hamiltonian

Super-exchange interaction

Kugel-Khomskii type Hamiltonian

$$H_{\rm exch} = \sum_{<ij>_\gamma} \left(H_{dd}^{ij;\gamma} + H_{dpd}^{ij;\gamma}\right) \sim \sum_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j)(T_i T_j)$$

$$\begin{cases} \tau_i^x = -\frac{1}{2}T_i^z - \frac{\sqrt{3}}{2}T_i^x \\ \tau_i^y = -\frac{1}{2}T_i^z + \frac{\sqrt{3}}{2}T_i^x \\ \tau_i^z = T_i^z \end{cases}$$

$$\begin{cases} \bar{\tau}_i^x = -\frac{1}{2}T_i^x + \frac{\sqrt{3}}{2}T_i^z \\ \bar{\tau}_i^y = -\frac{1}{2}T_i^x - \frac{\sqrt{3}}{2}T_i^z \\ \bar{\tau}_i^z = T_i^x \end{cases}$$

$$H_{dd}^{ij;\gamma} = -A_d\left(\frac{5}{4} - 5\tau_i^\gamma\tau_j^\gamma + 3\bar{\tau}_i^\gamma\bar{\tau}_j^\gamma + 3T_i^yT_j^y\right)P_{ij}^T$$

$$- B_d\left(\frac{5}{2} \mp 2\tau_i^\gamma \mp 2\tau_j^\gamma - 6T_i^yT_j^y\right)P_{ij}^S$$

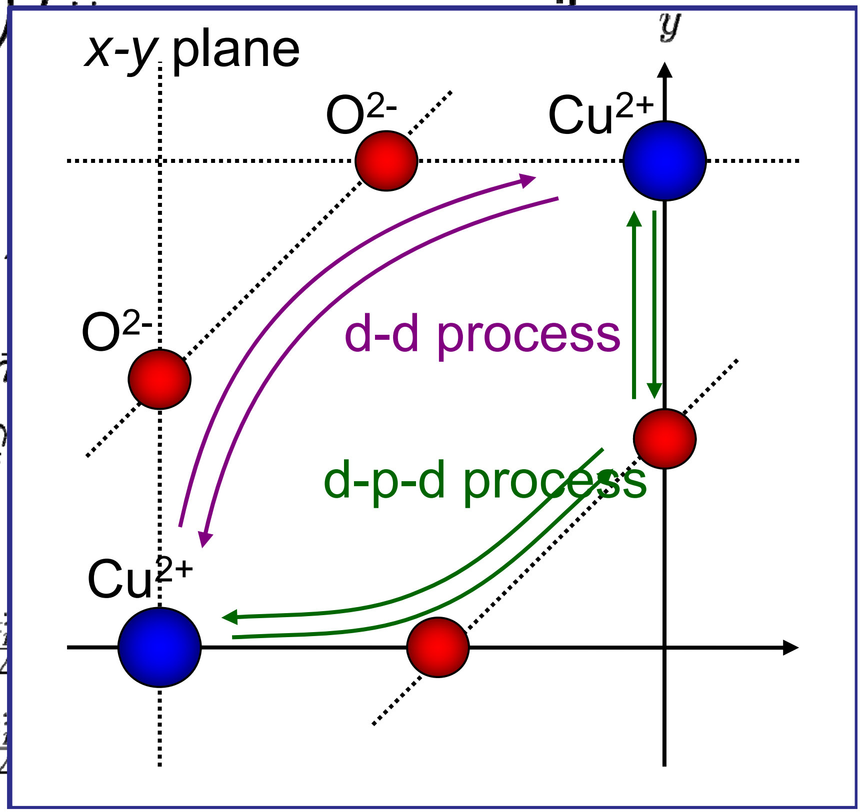
$$- C_d\left(\frac{5}{4} \mp 2\tau_i^\gamma \mp 2\tau_j^\gamma + 5\tau_i^\gamma\tau_j^\gamma - 3\bar{\tau}_i^\gamma \ldots$$

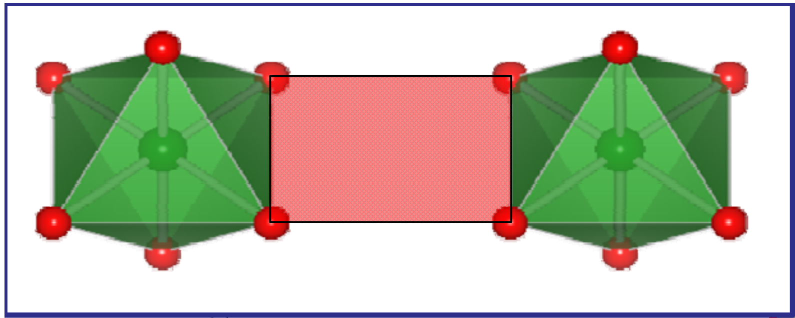
$$H_{dpd}^{ij;\gamma} = -A_p\left(4 \mp 4\tau_i^\gamma \mp 4\tau_j^\gamma + 4\tau_i^\gamma\tau_j^\gamma - 12\bar{\tau}_i^\gamma \ldots$$

$$- (B_p + 2C_p)\left(4 \mp 4\tau_i^\gamma \mp 4\tau_j^\gamma + 4\tau_i^\gamma\tau_j^\gamma \ldots$$

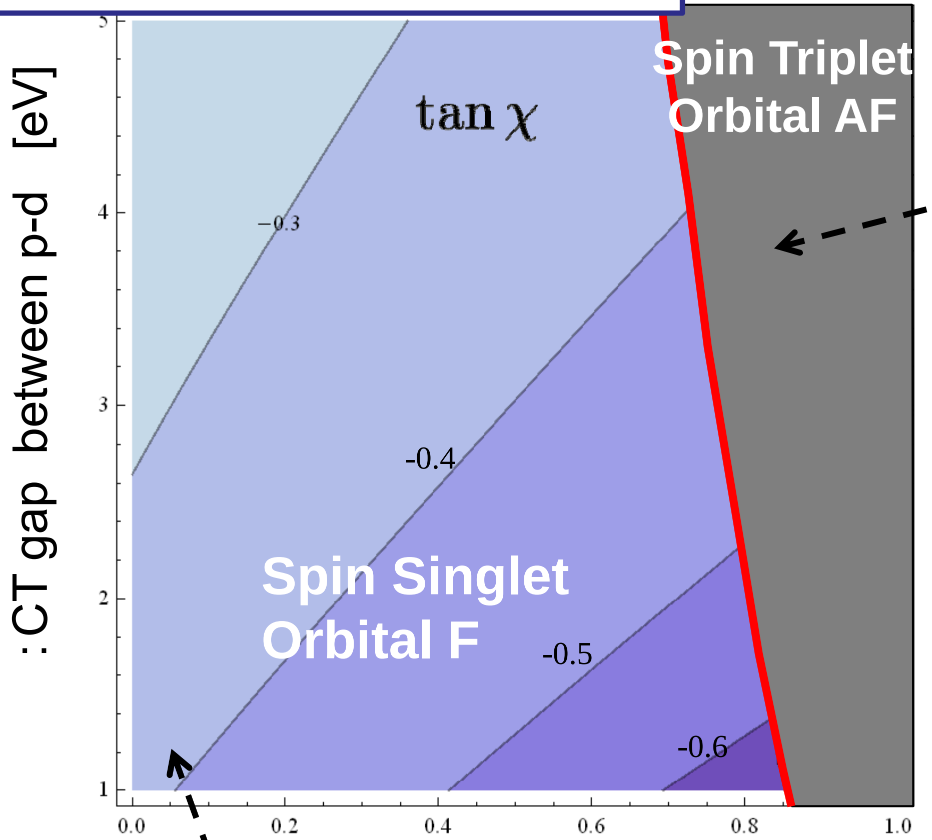
$$A_d = \frac{t_p^2 t_d^4}{\Delta^4}\frac{1}{U_d' - J_d} \qquad B_d = \frac{t_p^2 t_d^4}{\Delta^4}\frac{1}{U_d' + J_d} \qquad C_d = \ldots$$

$$A_p = \frac{t_p'^2 t_d^4}{\Delta^4}\frac{1}{U_p' - J_p + 2\Delta} \qquad B_p = \frac{t_p'^2 t_d^4}{\Delta^4}\frac{1}{U_p' + J_p + 2\Delta} \qquad C_p = \ldots$$





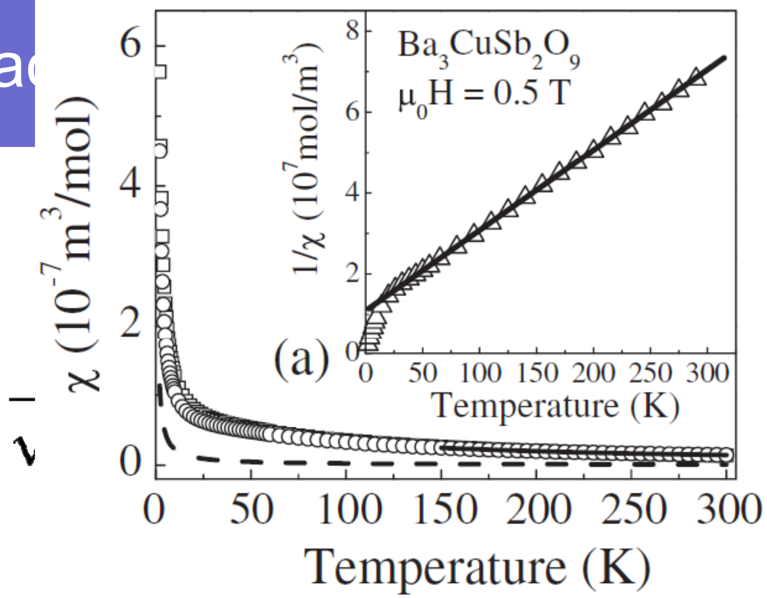
change Interac



$$x = J_d/U'_d = J_p/U'_p = J'_p/U'_p$$

Hund coupling

$$(\cos\chi|uu\rangle + \sin\chi|vv\rangle)_T|\mathrm{Singlet}\rangle_S$$



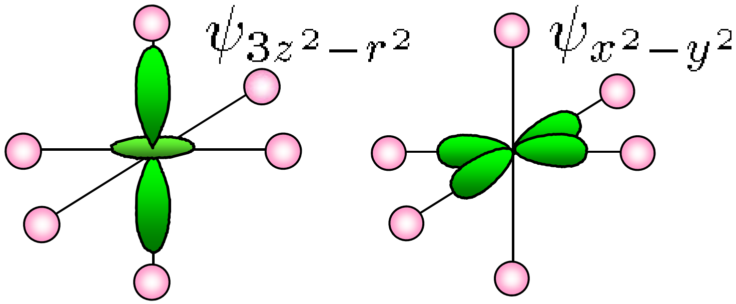
Spin: AF Orbital : F
in a wide region

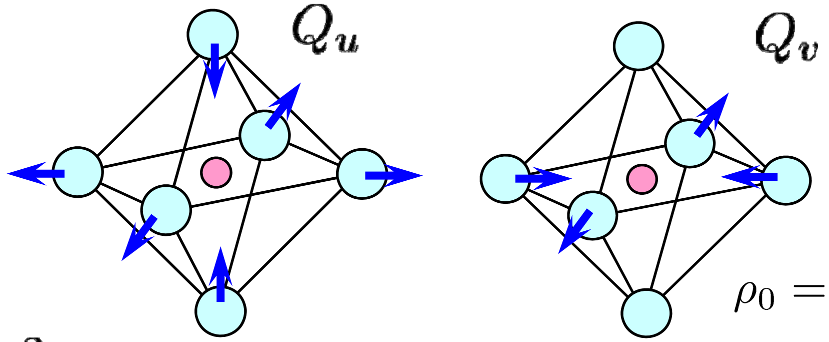
Consistent with
positive Weiss constant

⇕

Spin: F Orbital : AF
in a usual corner share bond
(e.g. Perovskite)

E ×e Jahn-Teller effect



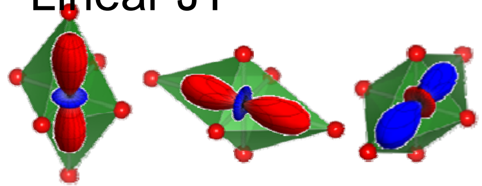


$$\rho_0 = \frac{A}{M\omega^2}$$

$$H_{\mathrm{JT}} = -\frac{\hbar^2}{2M}\left(\frac{\partial^2}{\partial Q_u^2} + \frac{\partial^2}{\partial Q_v^2}\right) + \frac{M\omega^2}{2}(Q_u^2 + Q_v^2) + A(\sigma^x Q_v - \sigma^z Q_u)$$

Kinetic　　Linear potential

Linear JT



$+B\left(Q_u^3 - 3Q_v^2 Q_u\right)$ Anharmonic potential

$E_{JT} \sim M\omega\rho_0^2/2$ JT energy gain (0.1-1eV)

$J_{AH} \sim B\rho_0^3$ Anharmonic potential energy (1-10meV)

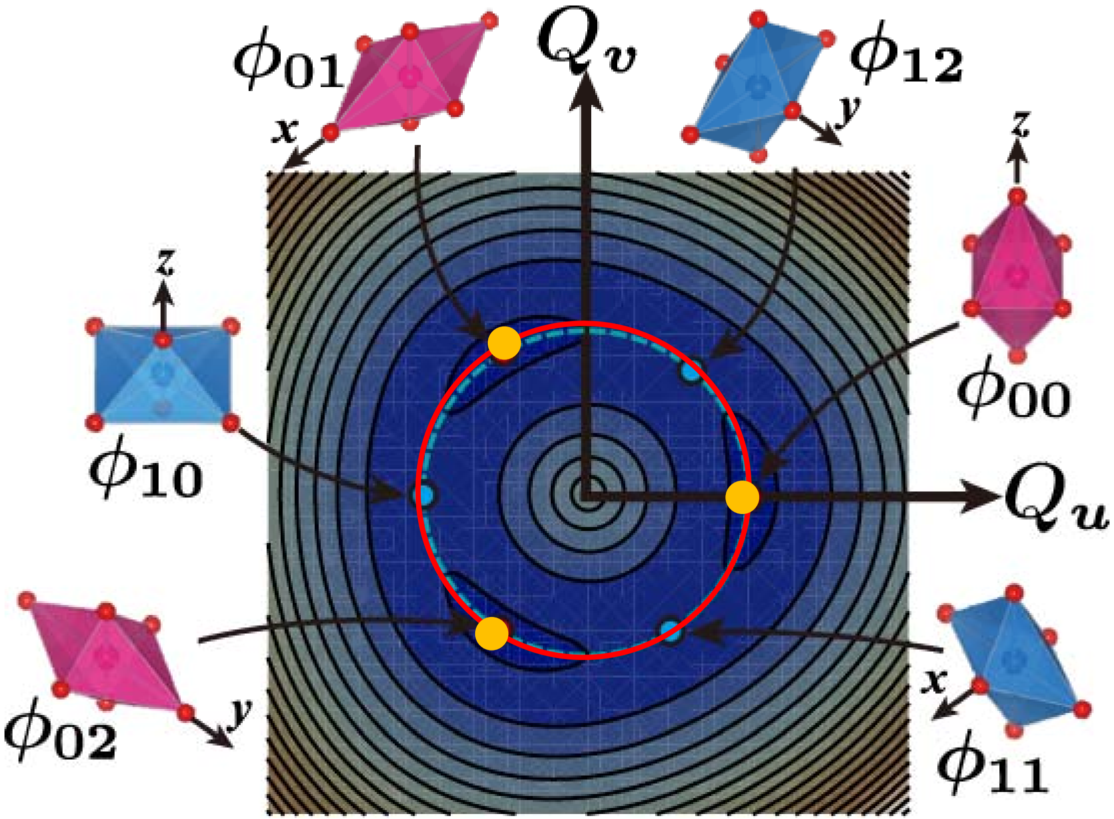
$J_{DJT} \sim (\hbar\omega)^2/E_{JT}^2$ dJT energy gain (1-10meV)

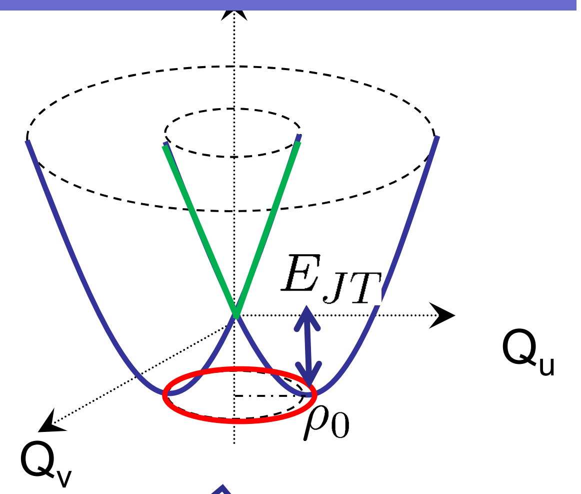
$J_{SE} \sim O(t_{pd}^4 t_{pp}^2/(U\Delta^4))$

Inter-site exchange (1-10meV)

Dynamic Jahn-Teller effect

Lower adiabatic potential with anisotropy





$$\mathcal{H}_{\rm rot} = -(2M\rho_0^2)^{-1}\partial^2/(\partial\theta^2) + B\rho_0^3\cos 3\theta$$

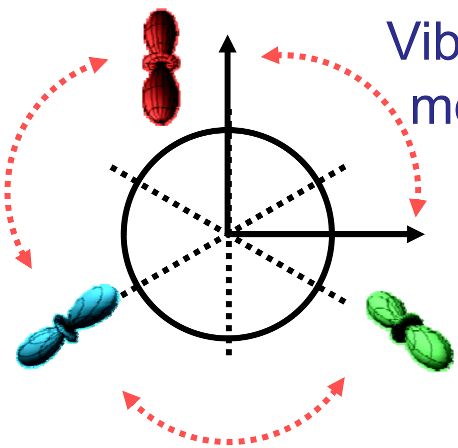
Rotational Mode

Tunnel between
3-potential minima

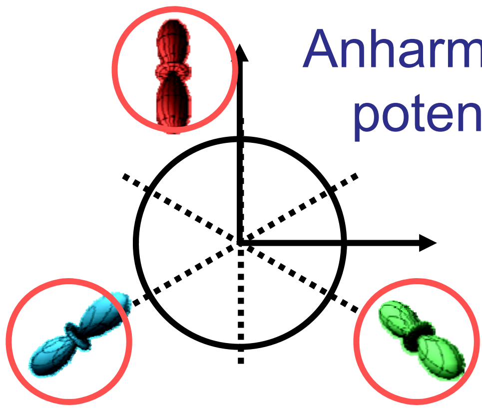
Hamiltonian for low-lying vibronic states

Effective Hamiltonian for the lowest 6 vibronic states

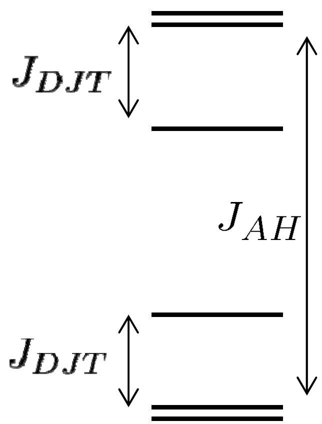
$$H_{JT} = \frac{1}{2}\sigma^z \left(J_{DJT} A + J_{AH} B\right)$$



Vibronic motion



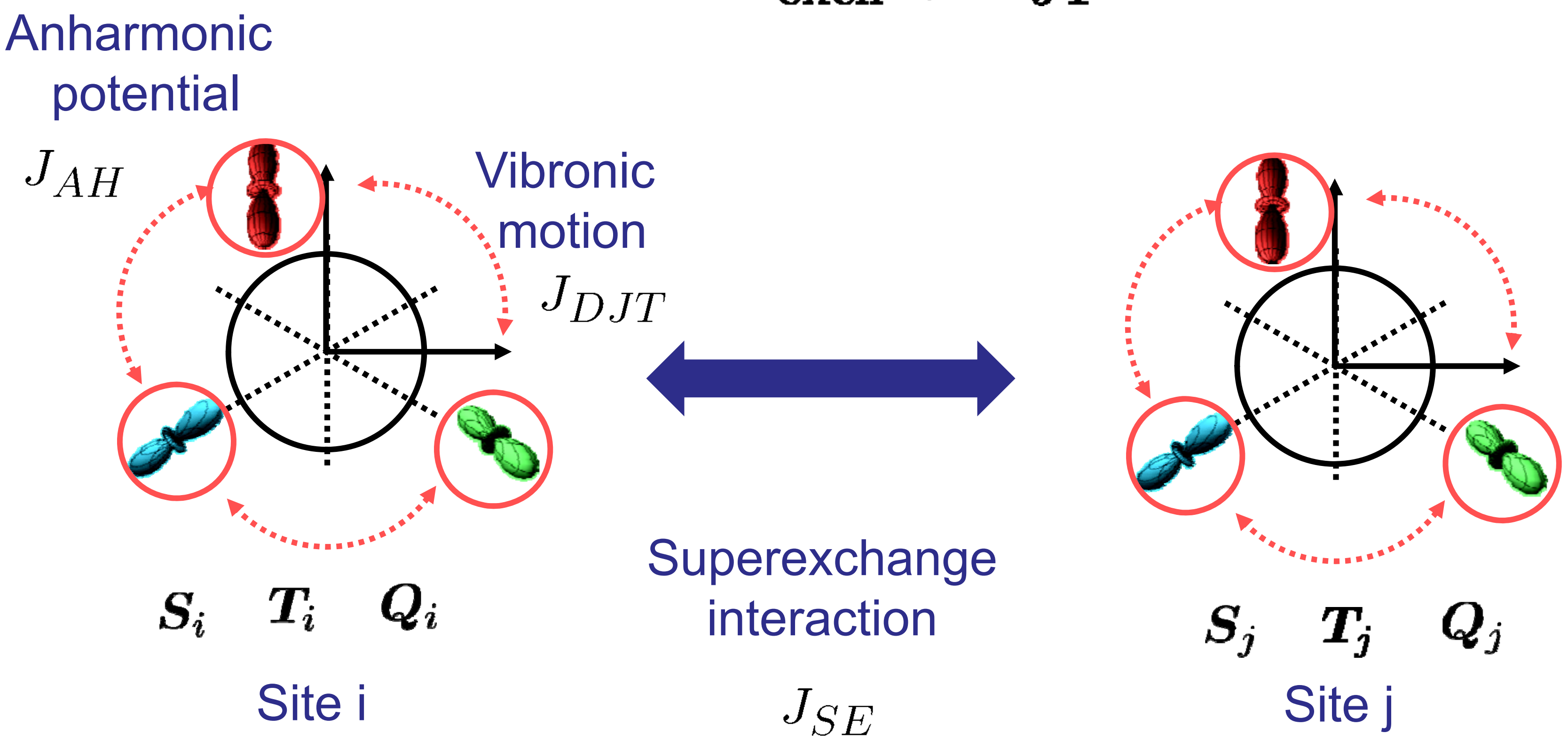
Anharmonic potential



$$A = \begin{pmatrix} 0 & 1 & 1 \\ 1 & 0 & 1 \\ 1 & 1 & 0 \end{pmatrix} \qquad B = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

Superexchange v.s. dJT

$$H = H_{\mathbf{exch}} + H_{\mathbf{JT}}$$



Local v.s. Inter-site

c.f. Kondo v.s. RKKY

Method

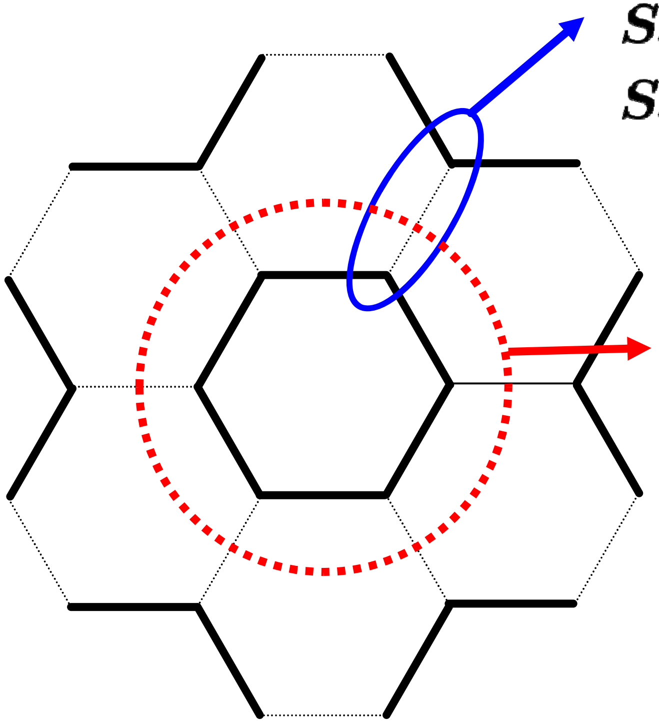
Exact-diagonalization
+
Mean field (MF) approx.

$$H = H_{\mathbf{exch}} + H_{\mathbf{JT}}$$

Mean field approx.

$$\boldsymbol{S}_i \cdot \boldsymbol{S}_j \to \boldsymbol{S}_i \cdot \langle \boldsymbol{S}_j \rangle$$

$$\boldsymbol{S}_i \cdot \boldsymbol{S}_j \tau_i \tau_j \to \tau_i \boldsymbol{S}_i \cdot \langle \boldsymbol{S}_j \tau_j \rangle \text{etc}$$



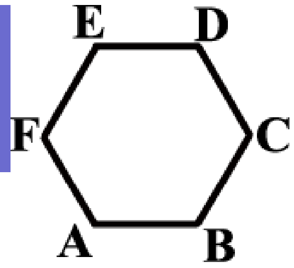
Exact diagonalization

Mean fields

Also
QMC+MF method

Further
$H_{\mathbf{exch}}$ is analyzed by Exact Dagonalization, & MF method

Spin & Orbital State



$J_{SE}/J_{AH} = 0.15 : fix$

Neel-type spin moment

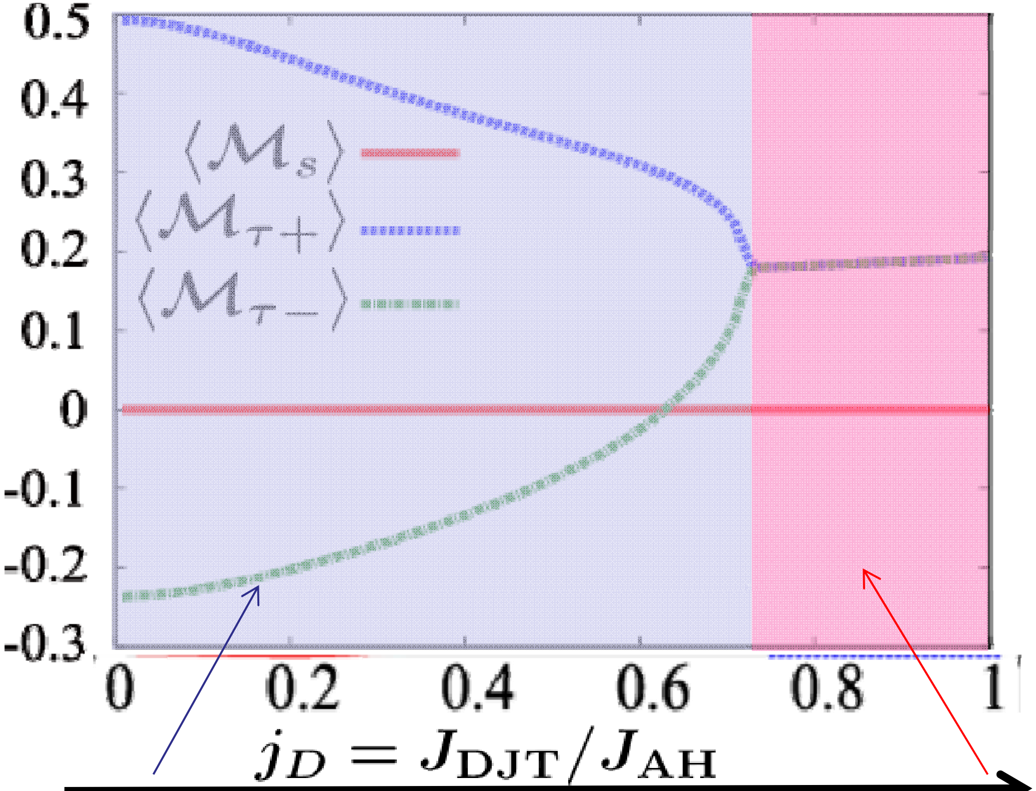
$$\mathcal{M}_s = \frac{1}{6}\sum_i (-1)^i S_i^z$$

3-hold orbital ordered moment (+)

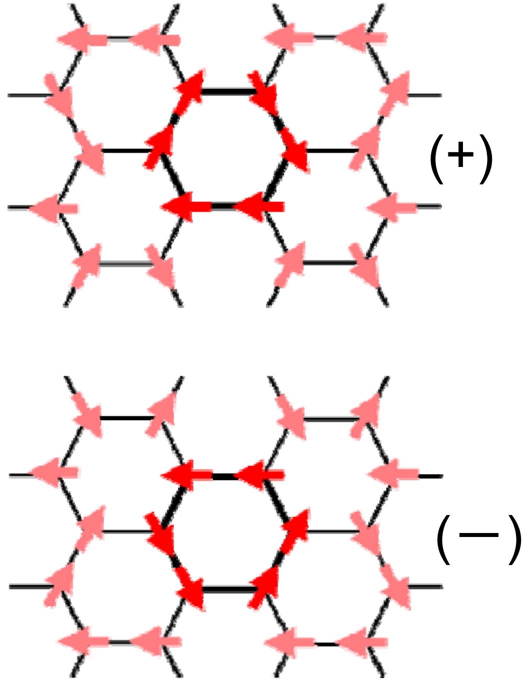
$$\mathcal{M}_{\tau+} = -\frac{1}{6}(\tau_{\rm A}^z + \tau_{\rm B}^z + \tau_{\rm C}^x + \tau_{\rm D}^x + \tau_{\rm E}^y + \tau_{\rm F}^y)$$

3-hold orbital ordered moment (-)

$$\mathcal{M}_{\tau-} = -\frac{1}{6}(\tau_{\rm A}^x + \tau_{\rm B}^y + \tau_{\rm C}^y + \tau_{\rm D}^z + \tau_{\rm E}^z + \tau_{\rm F}^x)$$



Orbital pseudospin configuration



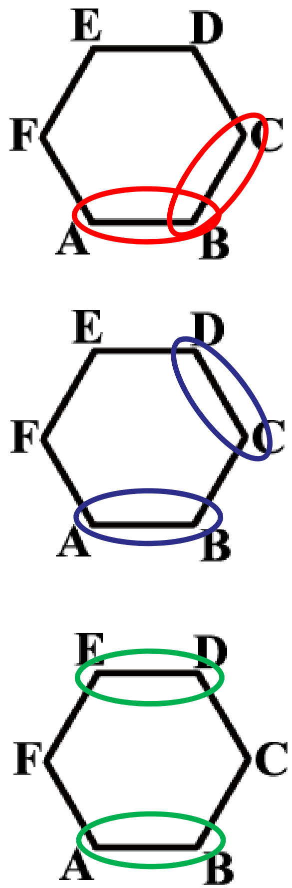
(+)-type 3-hold orbital order

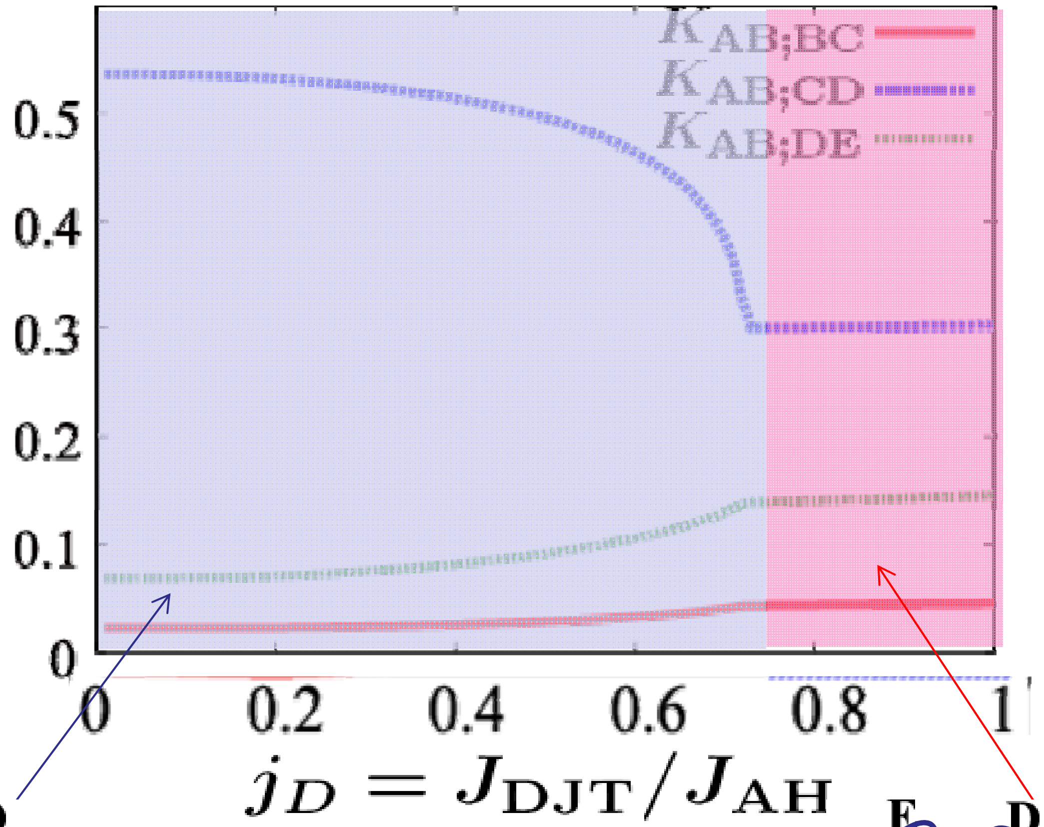
Vibronic Motion

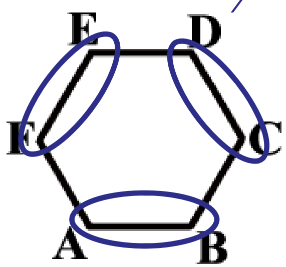
Superposition of (+) & (-) orbital orders

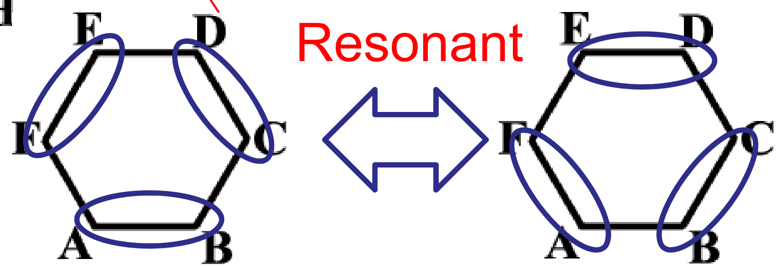
Spin & Orbital State

Bond correlation function $K_{ij;kl} = \langle (\mathbf{S_i} \cdot \mathbf{S_j})(\mathbf{S_k} \cdot \mathbf{S_l}) \rangle$









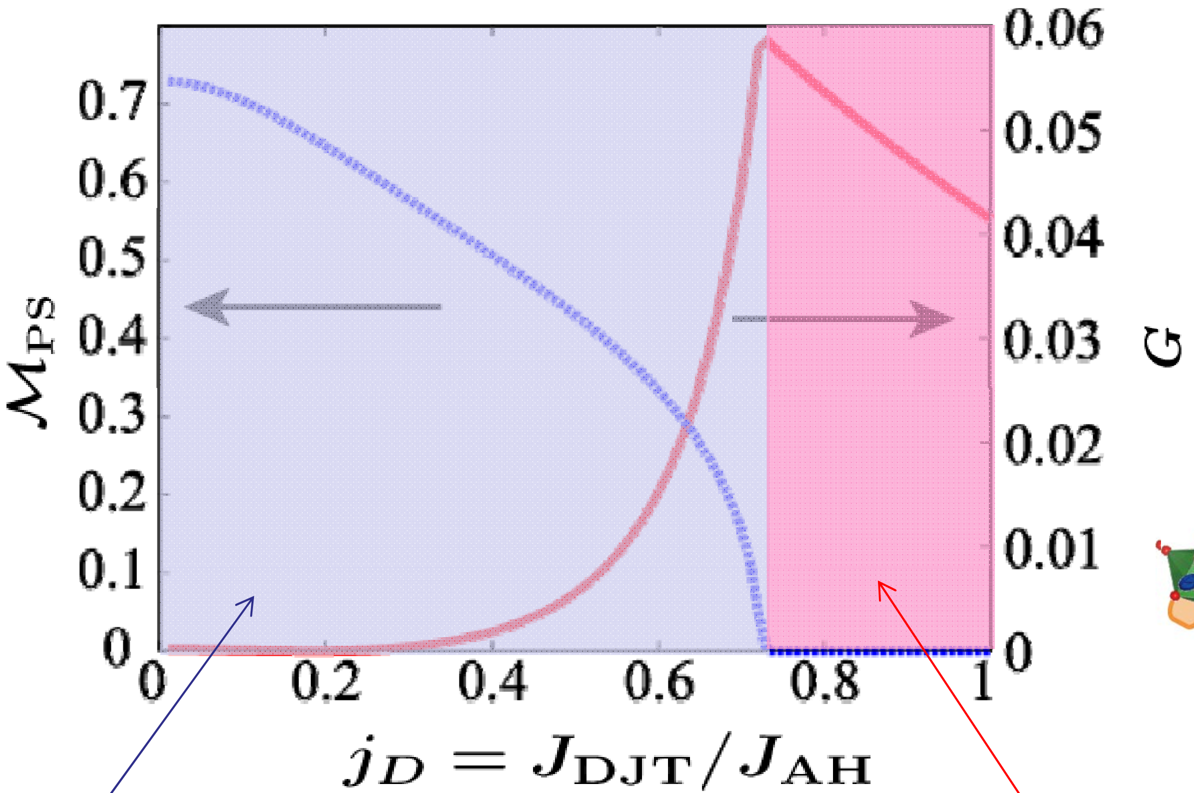
Spin & Orbital State

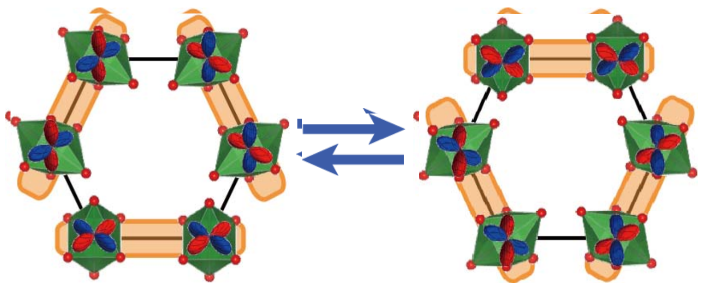
Spin-orbital entanglement

$$G = [\frac{1}{6}\sum_{\langle ij \rangle_l} G^l_{ij}]^2$$

$$G^l_{ij} = 16[\langle (\mathbf{S_i}\cdot\mathbf{S_j})(\tau^l_\mathbf{i}\tau^l_\mathbf{j})\rangle - \langle \mathbf{S_i}\cdot\mathbf{S_j}\rangle\langle \tau^l_\mathbf{i}\tau^l_\mathbf{j}\rangle]$$

3-hold orbital order parameter $\mathcal{M}_{\rm PS} \equiv \langle \mathcal{M}_{\tau+} - \mathcal{M}_{\tau-} \rangle$

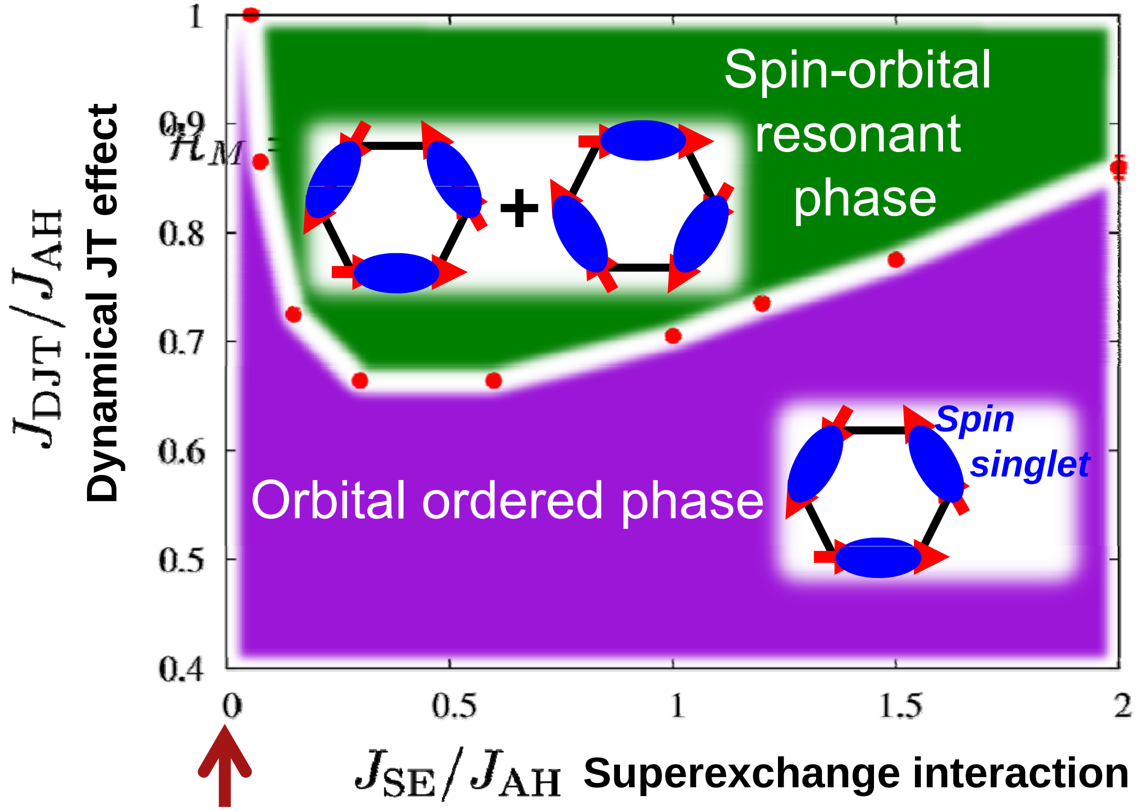




Spin-orbital disentangled orbital order

Spin-orbital entangled quantum state

Superexchage v.s. dJT



Spin-orbital system decoupled from lattice vibration

Spin-orbital system with reduced orbital moments

Competition between Dynamical JT and superexchange interaction

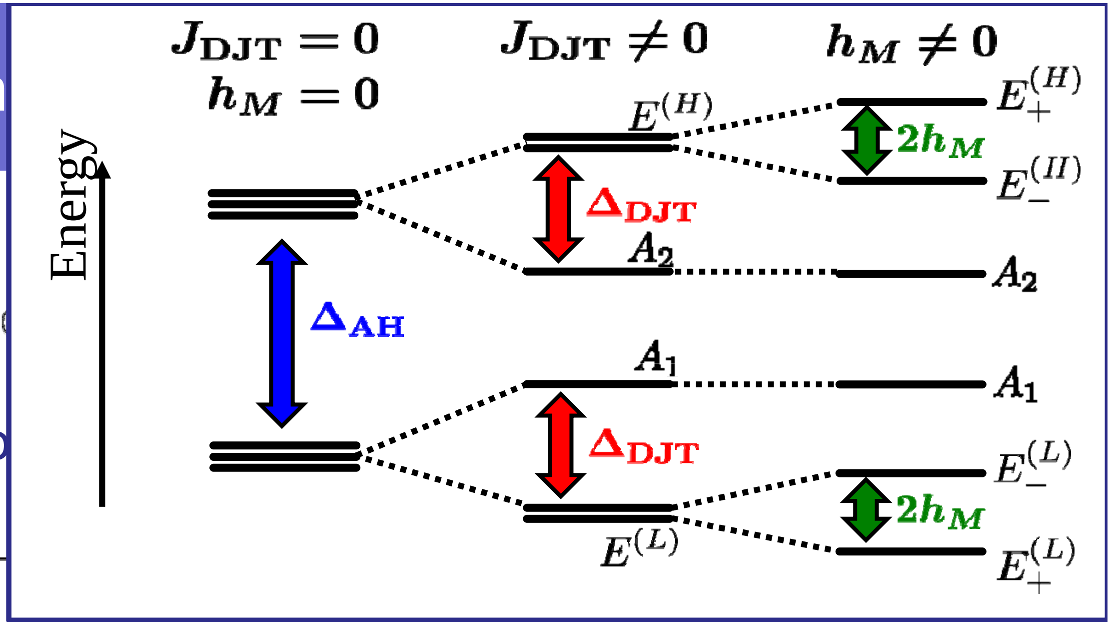
Spin-orbital resonant state

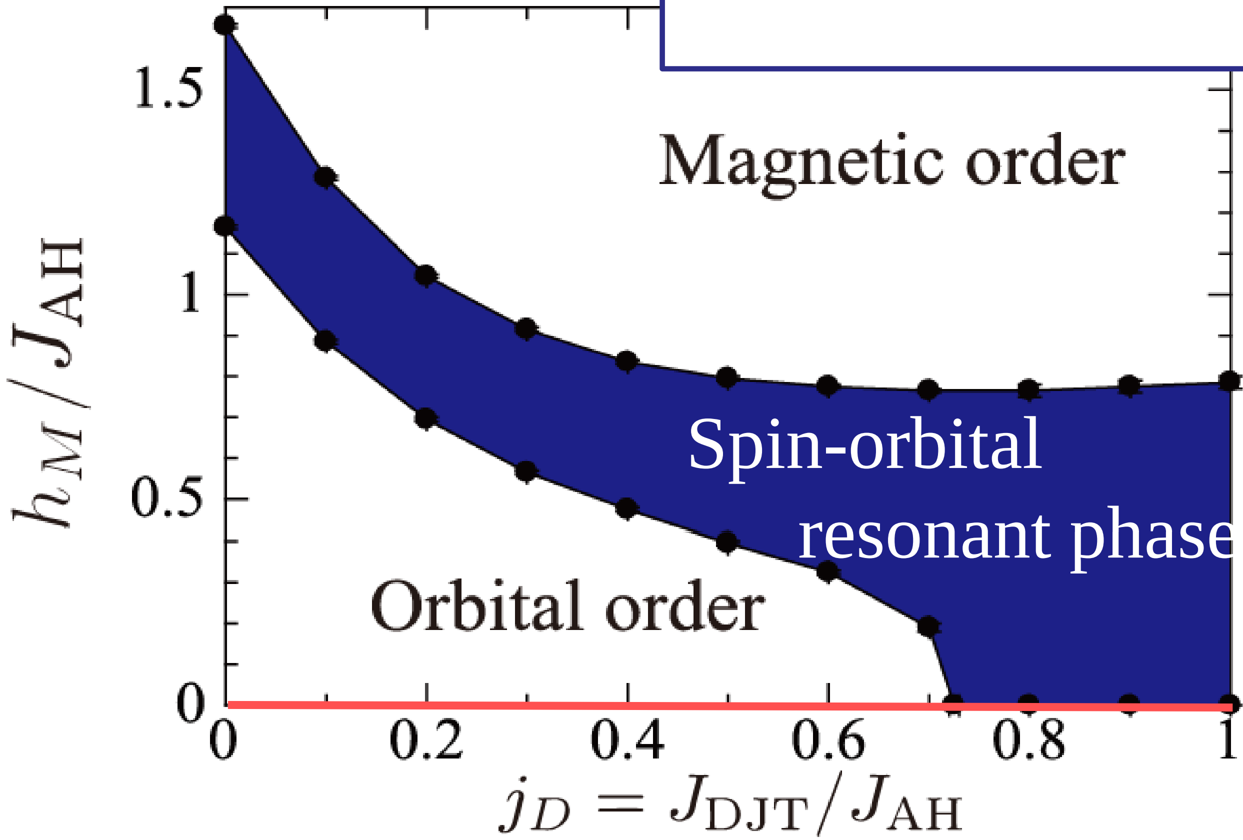
Connection

Artificial external

$\mathcal{H}_M = -h_M \sum_i \sigma_i^z$

$h_M \to \infty$ AFM ho



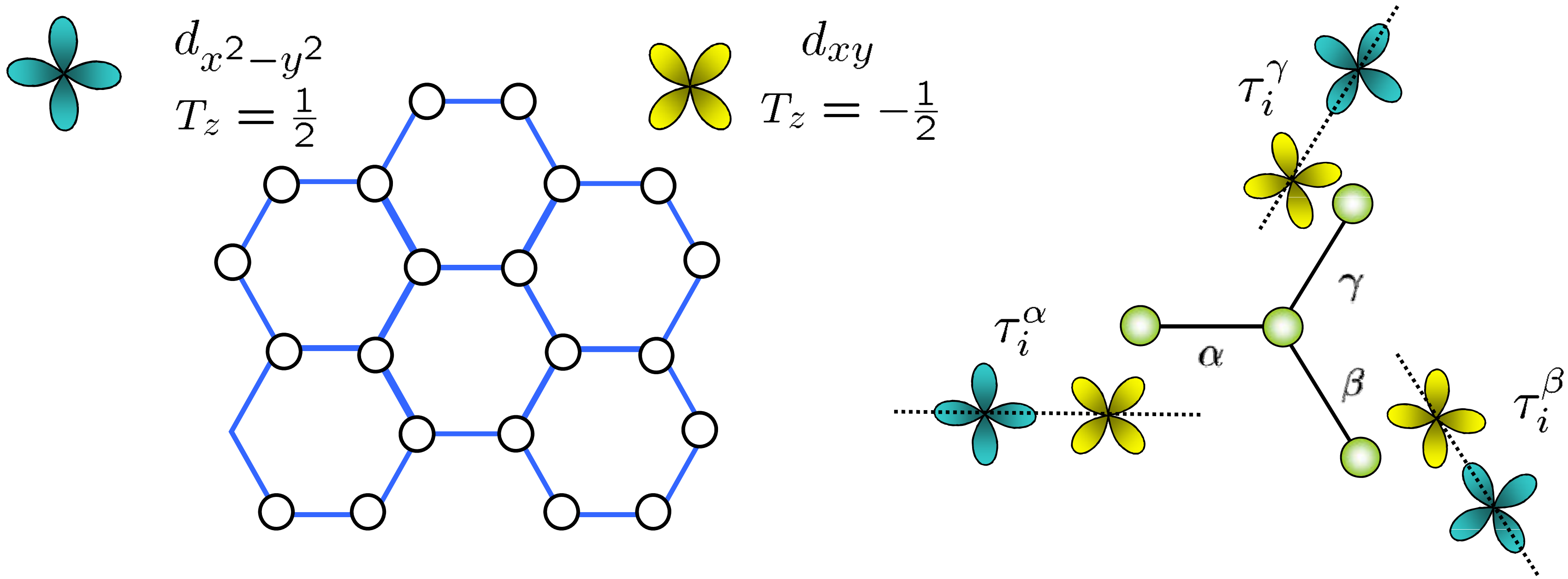


Heisenberg model without orbital degree of freedom

Original model (spin-orbital-lattice)

Honeycomb lattice 120 degree orbital model

Doubly-degenerate orbital on a honeycomb lattice



$$\mathcal{H} = \frac{1}{2} J \sum_i \left(\frac{3}{4} + \tau_i^\alpha \tau_{i+\delta_\alpha}^\alpha + \tau_i^\beta \tau_{i+\delta_\beta}^\beta + \tau_i^\gamma \tau_{i+\delta_\gamma}^\gamma \right)$$

$J < 0$

$$\tau_i^l = \cos\left(\frac{2n_l\pi}{3} + \frac{\pi}{2}\right) T_i^z + \sin\left(\frac{2n_l\pi}{3} + \frac{\pi}{2}\right) T_i^x$$

$$(n_\alpha, n_\beta, n_\gamma) = (1, 2, 3)$$

Variational approach

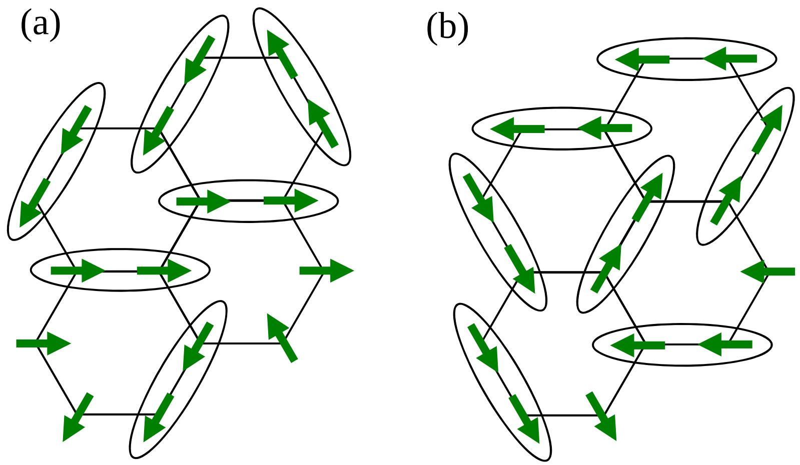
Honeycomb lattice is covered by NN bonds
with the minimum bond energy

trial wave function

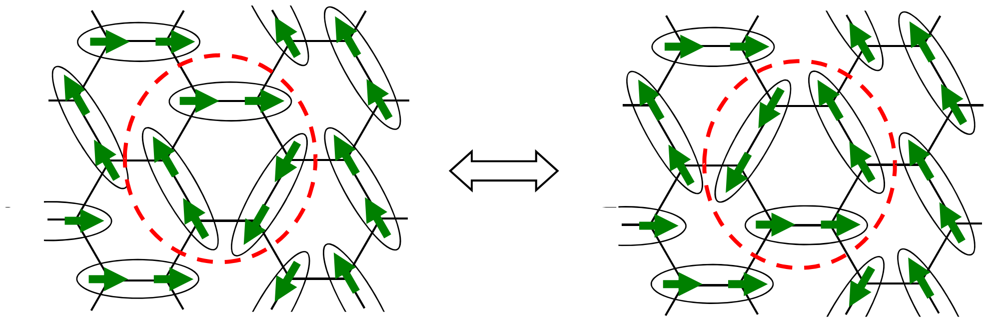
$$|\Psi^{(+)}\rangle = \mathcal{N} \sum_l \mathcal{A}_l \left\{ |\psi_l^{(\uparrow)}\rangle + |\psi_l^{(\downarrow)}\rangle \right\},$$

$$|\psi_l^{(\uparrow)}\rangle = \prod_{\langle ij \rangle_l} U(\phi_\eta)_{\langle ij \rangle_l} | \uparrow \cdots \uparrow \rangle.$$

$$U(\phi_\eta)_{\langle ij \rangle_l} = \exp\left[-i\phi_\eta \left(T_i^y + T_j^y \right) \right],$$



Quantum Resonance



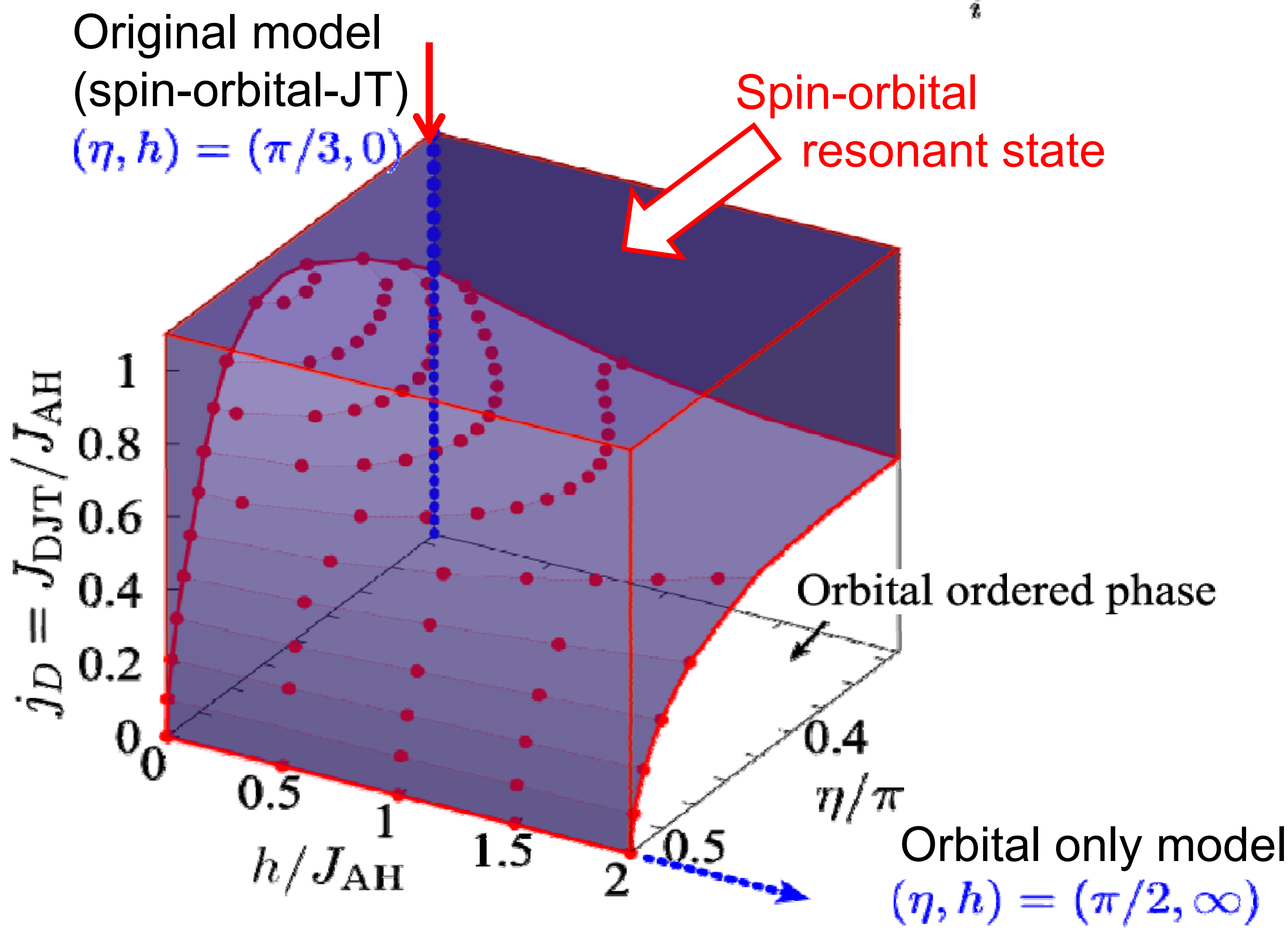
Resonance energy:

～10% of energy gain
of quantum effect

Connection to orbital-only model

Generalization of electron transfer $t_{pd} \to t_{pd}(\eta)$

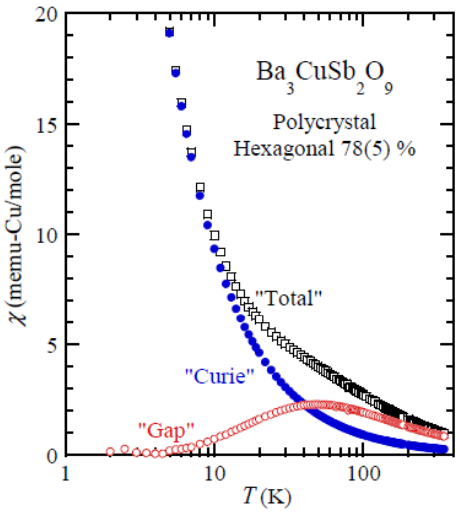
Staggered magnetic field $\mathcal{H}_h = -h \sum_i (-1)^i S_i^z$



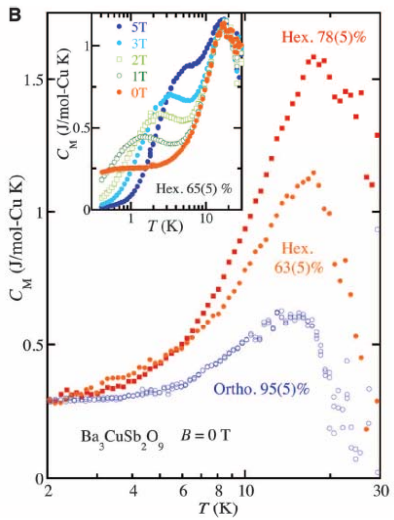
Discussion for $Ba_3CuSb_2O_9$

Low energy excitation + Gapped excitation

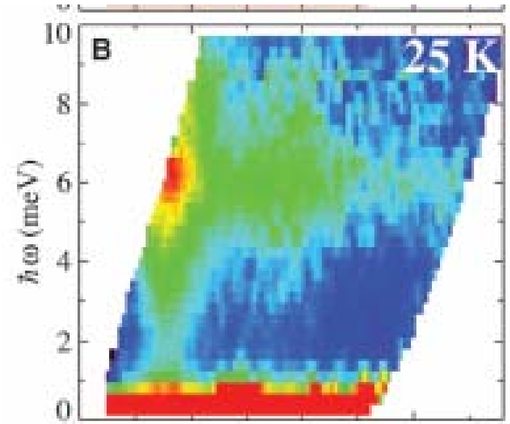
- Susceptibility
  $\chi \sim \chi(\text{free}) + \chi(\text{gap})$



- Specific heat
  low energy + gapped parts



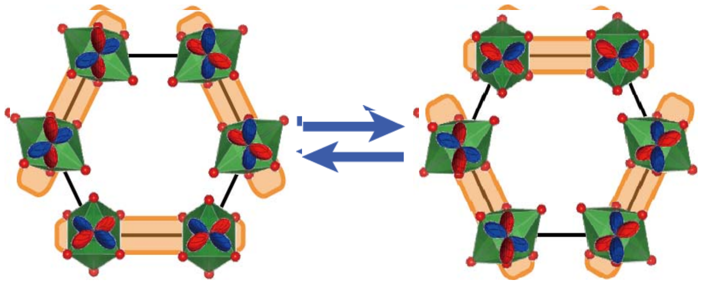
- Inelastic Neutron scattering
  Gapped excitation

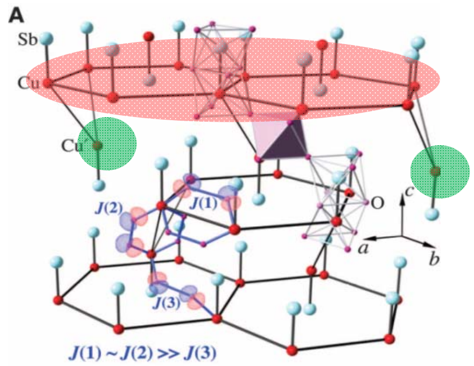


Nakatsuji et al.
Science 336, 559 (2012)

Spin-orbital resonance

Gapped spin liquid





Orbital Excitation “Orbiton”
&
INS as an experimental probe

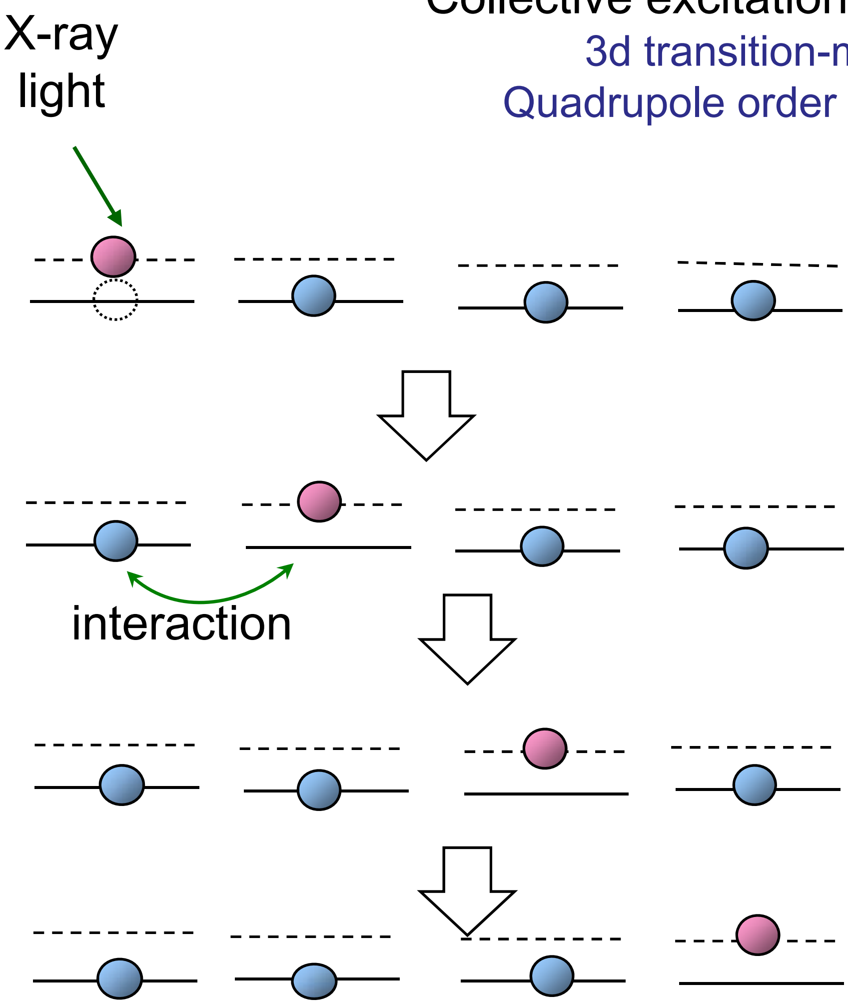
Orbiton

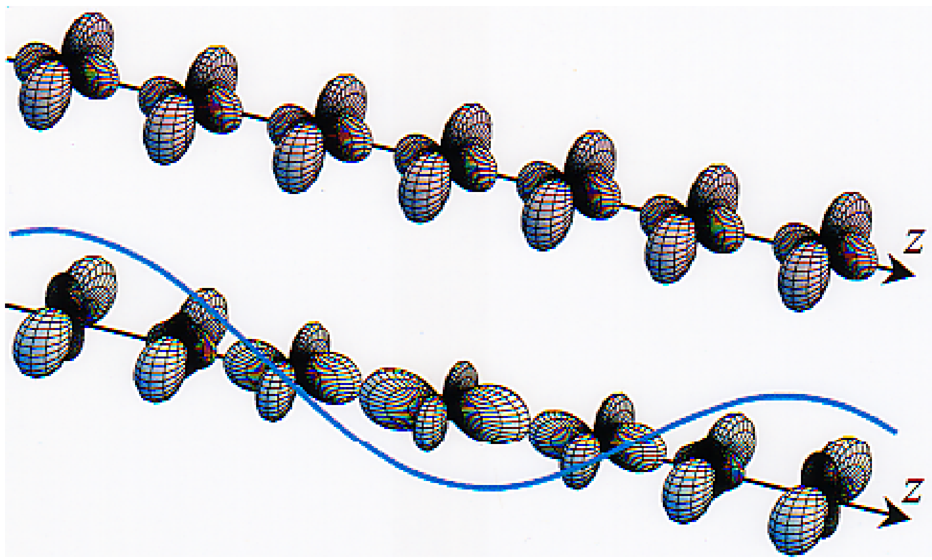
Orbital wave (orbiton)

Collective excitation in orbital ordered state

3d transition-metal compounds

Quadrupole order in 4f electron systems

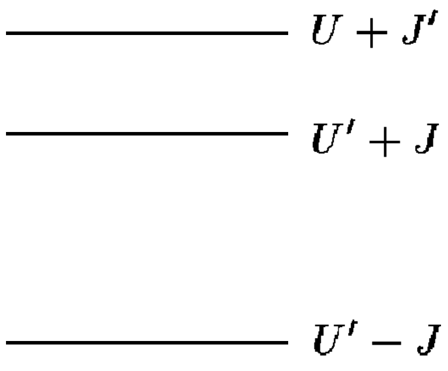




M. Cyrot and C. Lyon-Caen, J. Phys. (Paris) 36, 253 (1975)

Orbiton

Spin-orbitla model

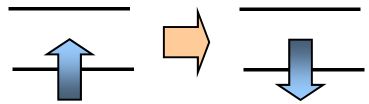


$$H = -2J_1 \sum_{\langle ij \rangle} \left(\frac{3}{4} + \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{4} - \tau_i^l \tau_j^l \right)$$
$$-2J_2 \sum_{\langle ij \rangle} \left(\frac{1}{4} - \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{3}{4} + \tau_i^l \tau_j^l + \tau_i^l + \tau_j^l \right)$$

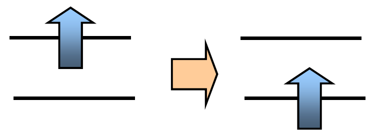
Horstein-Primakoff trans.

$$T_i^z = \frac{1}{2} - t_i^\dagger t_i$$
$$T_i^+ = \left(1 - t_i^\dagger t_i\right)^{1/2} t_i$$
$$T_i^- = t_i^\dagger \left(1 - t_i^\dagger t_i\right)^{1/2}$$

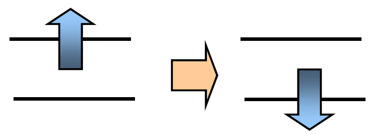
magnon $s_i^\dagger \ s_i$



orbiton $t_i^\dagger \ t_i$

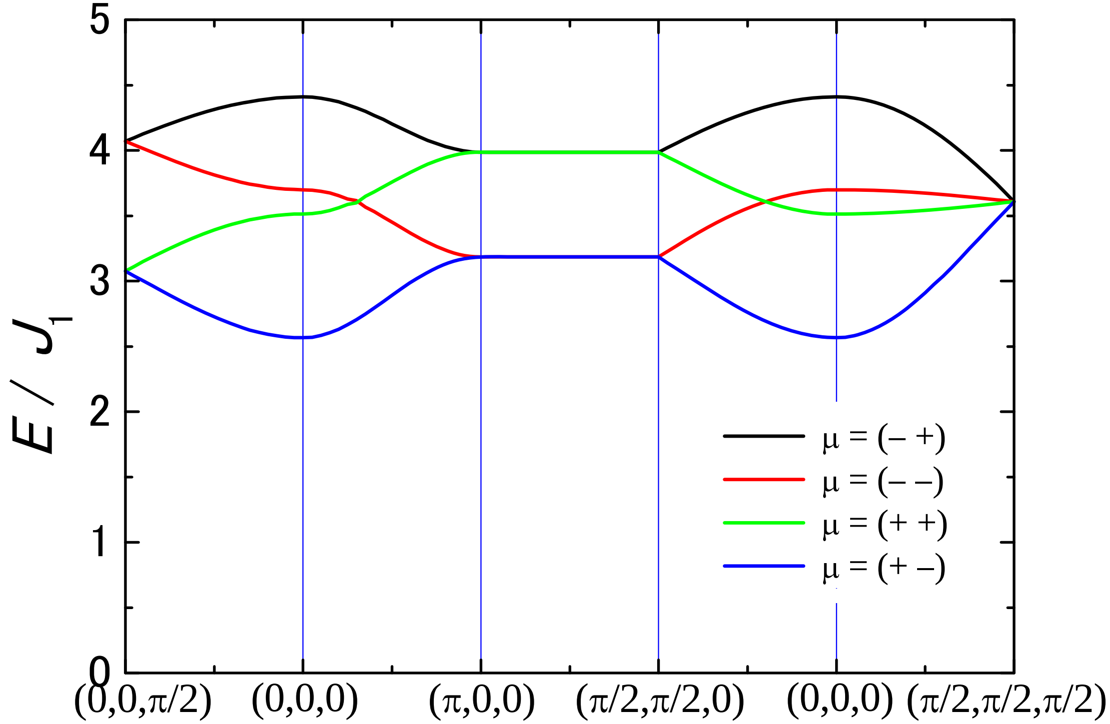


orbiton-magnon $t_i^\dagger s_i^\dagger \quad t_i s_i \quad t_i^\dagger s_i \quad t_i s_i^\dagger$



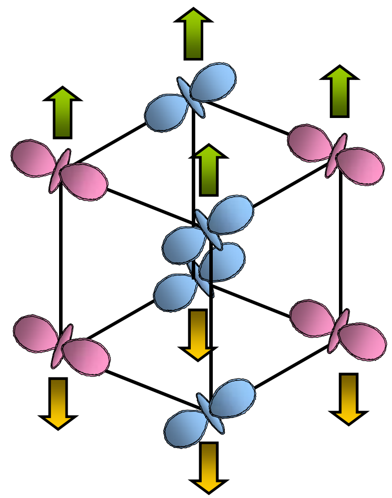
Orbiton dispersion relation

LaMnO$_3$
-a mother compound of CMR material -



4-modes
(4-inequivalent Mn sites)

Gap-full excitation
(No SU(2) or O(2))

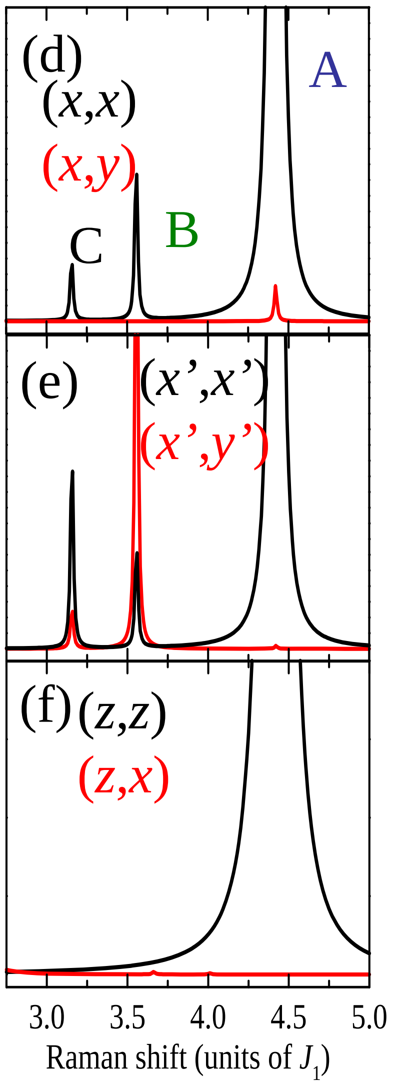


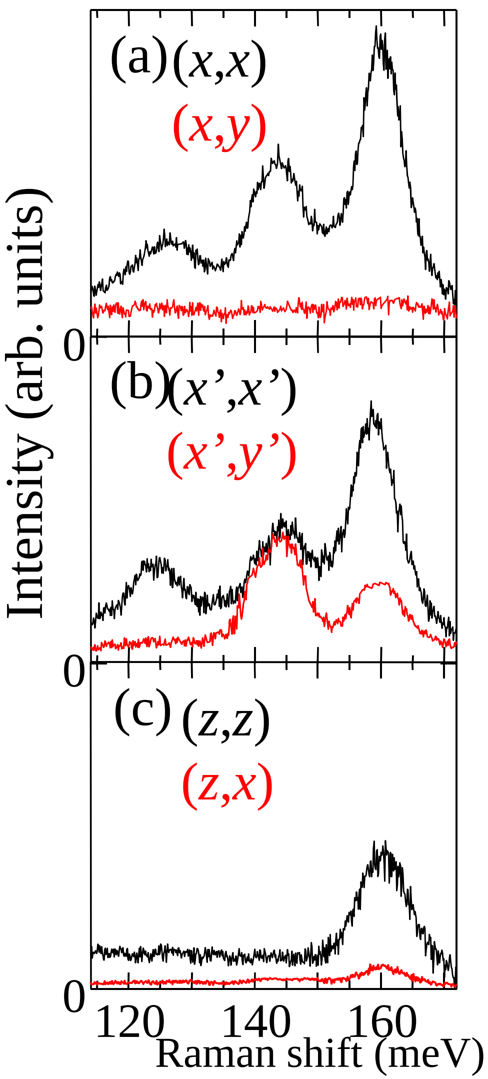
S. Ishihara, J. Inoue, S. Maekawa
Phys. Rev. B 55, 8280 ('97).

Orbiton by Ramman scattering

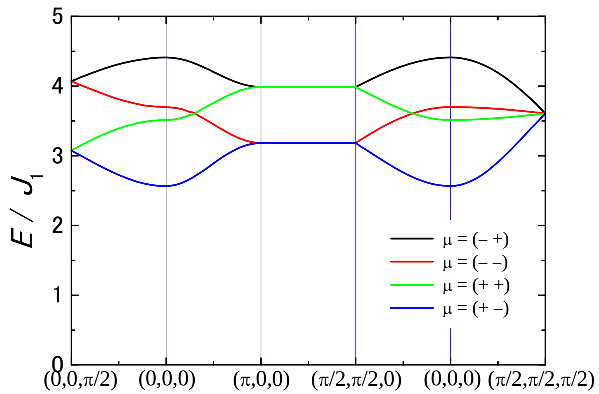
Theory

Experiment





$LaMnO_3$



E. Saitoh et al.
Nature 410 180 ('01)

(Multi-phonon ?)

tal + Dynamical

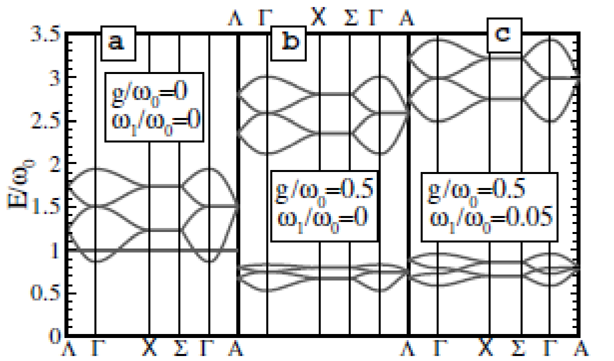


FIG. 2. Orbiton and phonon dispersion, neglecting dynamical effects due to the *e-p* coupling; (a) without *e-p* coupling *g* and without bare phonon dispersion, (b) $g/\omega_0 = 1/2$, no bare phonon dispersion, and (c) $g/\omega_0 = 1/2$, finite bare phonon dispersion. The points of high symmetry in the Brillouin zone correspond to those of Ref. [13].

Effects of

JT coupling

mics (Dynamical JT,

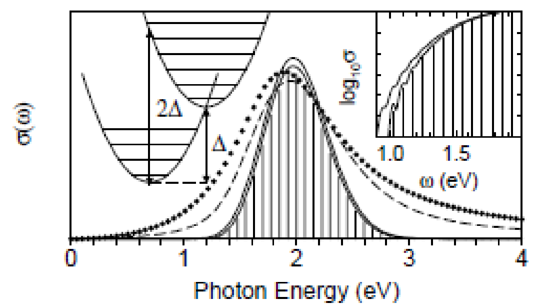


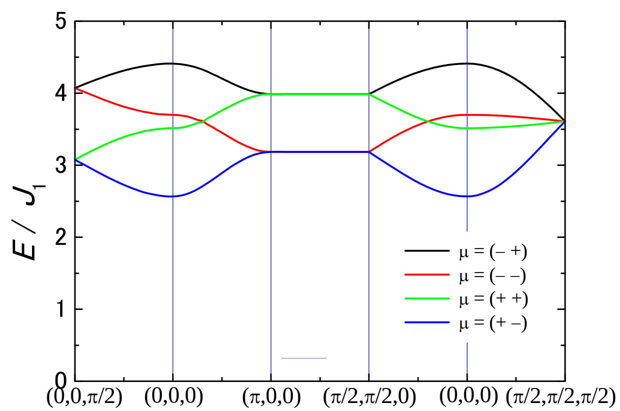
FIG. 2. Optical conductivity of $LaMnO_3$. The points are the lowest Lorentzian oscillator fit by Jung *et al.* [16] to their data. The dashed curve is a $T = 0$ sum of convolved Lorentzians centered at the vibrational replicas shown as vertical bars; the solid curves are $T = 0$ (lower) and $T = 300$ K (upper) sums of convolved Gaussians, also shown in the inset on a logarithmic scale. Tick marks in the inset denote decades.

Vibronic excitation (cooperative JT problem)

SI et al. Phys. Rev. B 62, 2338 ('00)

Frozen JT distortion

(OK for ω(orbiton) > ω(phonon))



V. Pere ... 18 (01)

Orbital ex ... nge int.)

Fu ... cs

Orbital – Lattice coupling

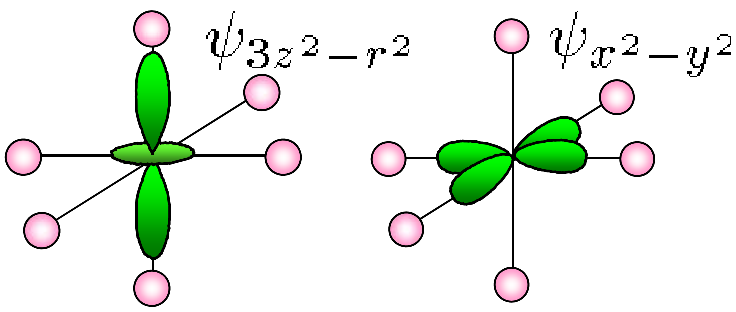
$$H_J = -2J_1 \sum_{\langle ij \rangle} \left(\frac{3}{4} + \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{1}{4} - \tau_i^l \tau_j^l \right)$$
$$-2J_2 \sum_{\langle ij \rangle} \left(\frac{1}{4} - \vec{S}_i \cdot \vec{S}_j \right) \left(\frac{3}{4} + \tau_i^l \tau_j^l + \tau_i^l + \tau_j^l \right)$$

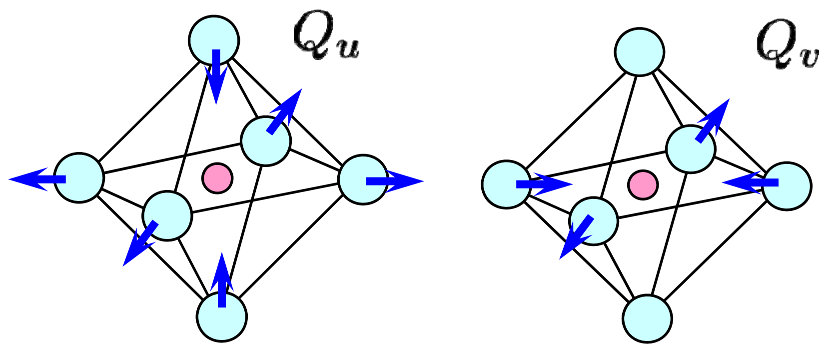
Exchange interaction

$$\vec{S}_i \cdot \vec{S}_j \rightarrow \langle \vec{S}_i \cdot \vec{S}_j \rangle$$

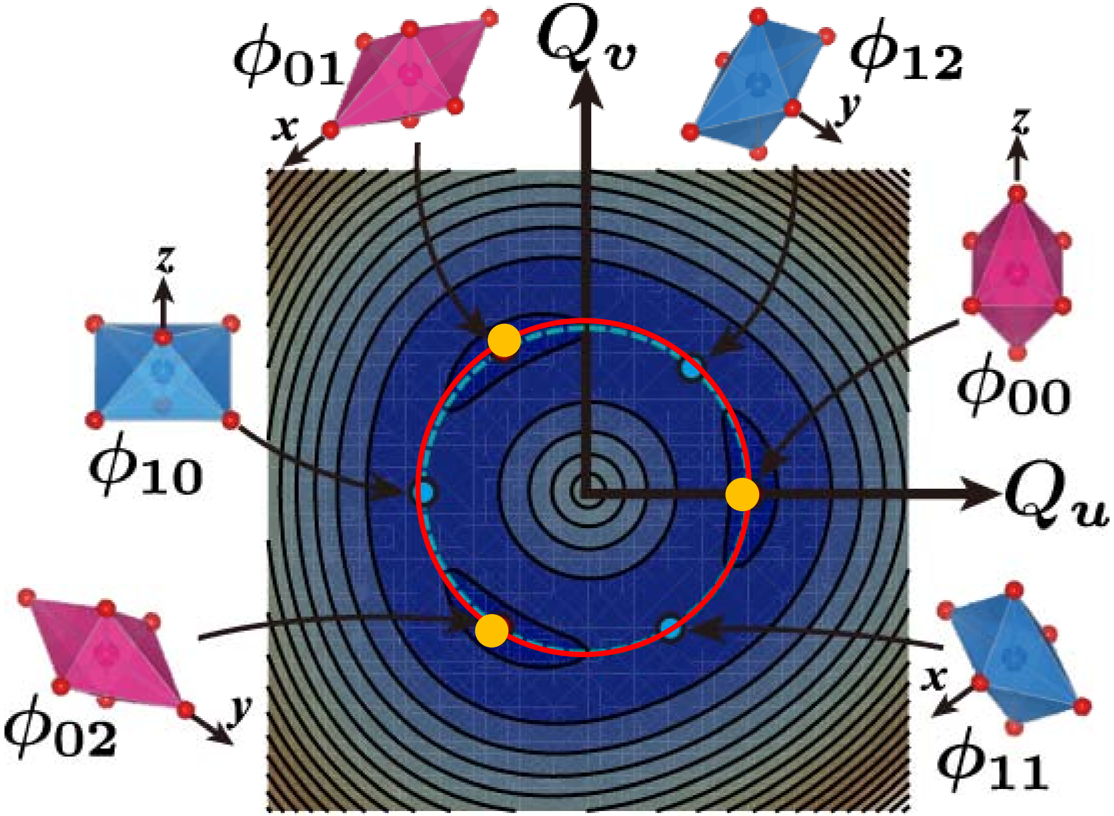
$$H_{\mathrm{JT}} = -\frac{\hbar^2}{2M} \left(\frac{\partial^2}{\partial Q_u^2} + \frac{\partial^2}{\partial Q_v^2} \right) + \frac{M\omega^2}{2} (Q_u^2 + Q_v^2) + A(\sigma^x Q_v - \sigma^z Q_u)$$

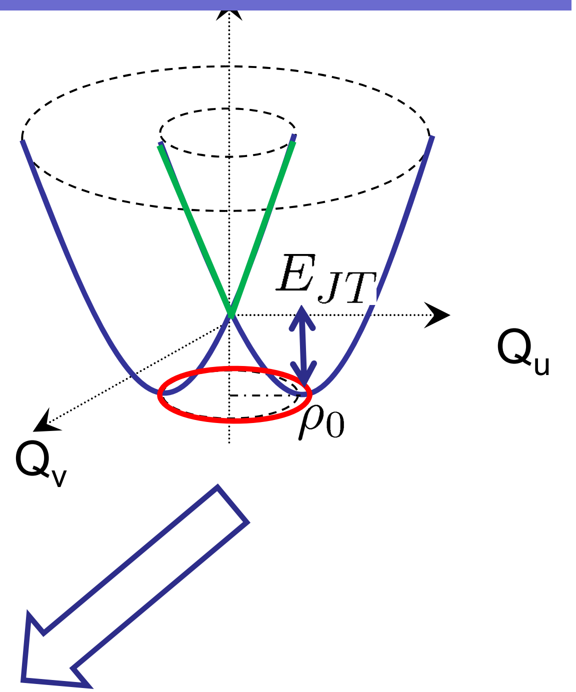
Kinetic | Lattice potential | JT interaction



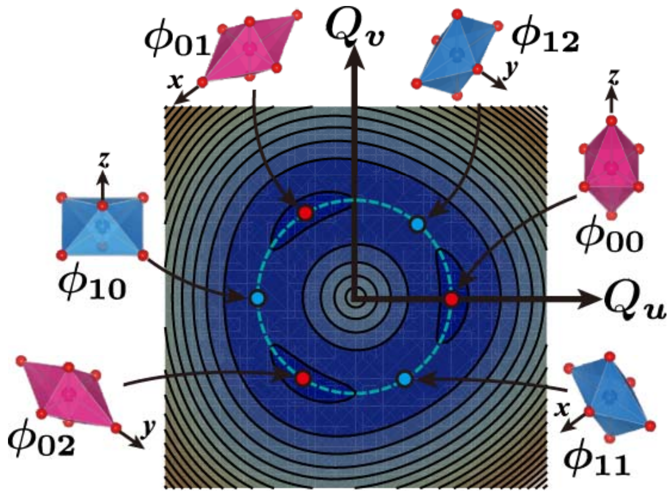


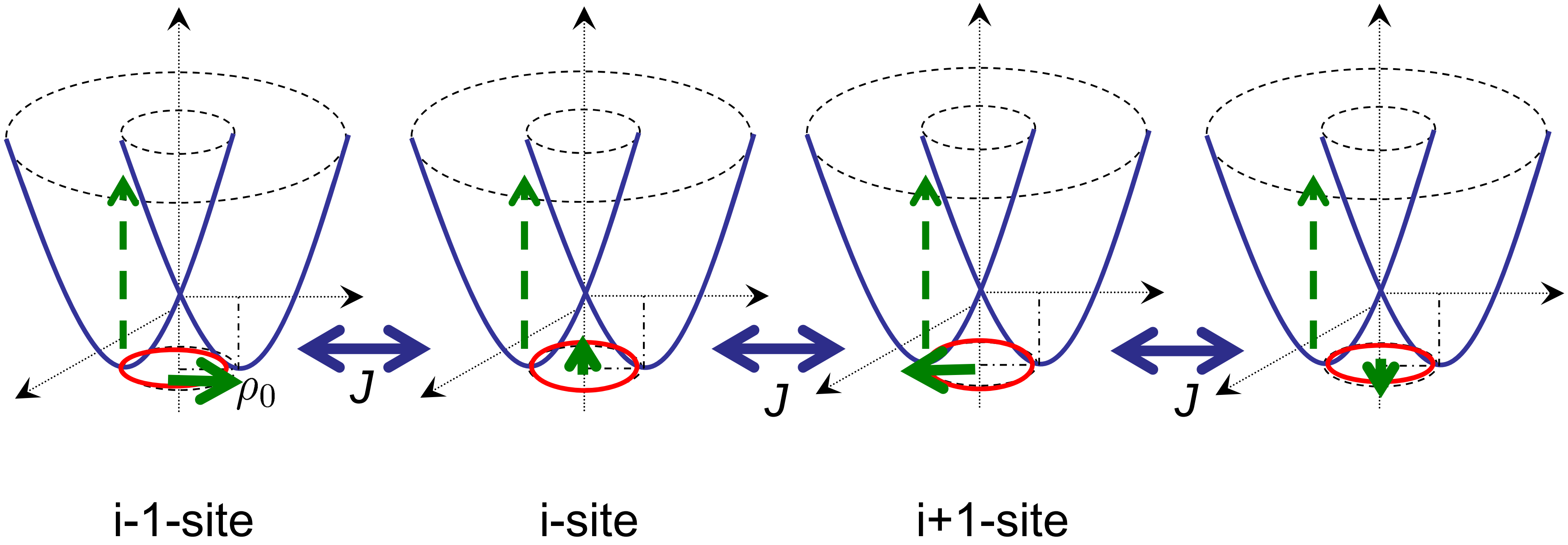
Dynamic Jahn-Teller effect





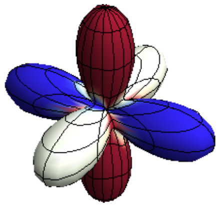
Vibronic collective mode





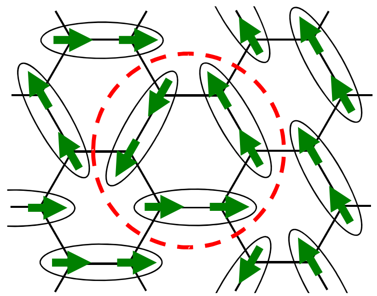
Summary

"Orbital-Frustration-Entanglement"



- Ring exchange interaction in cubic orbital 120models

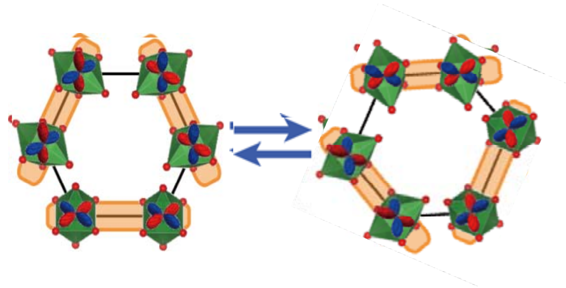
  Magnetic quadrupole order



- Honeycomb lattice orbital 120models

  Possibility of quantum orbital state

- DJT effect in honeycomb lattice spin-orbital model



  Spin-Orbital resonant state
  Implication to Ba3Sb2CuO9

- Orbital excitation

  Low energy collective
  vibronic mode