Orbital Physics

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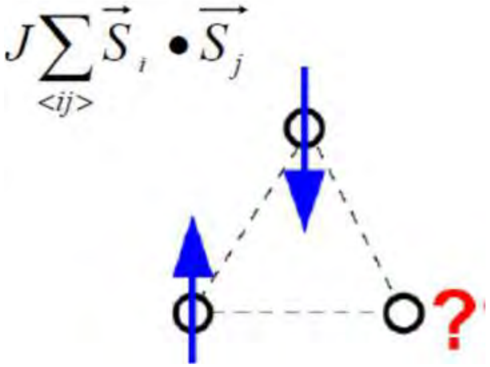

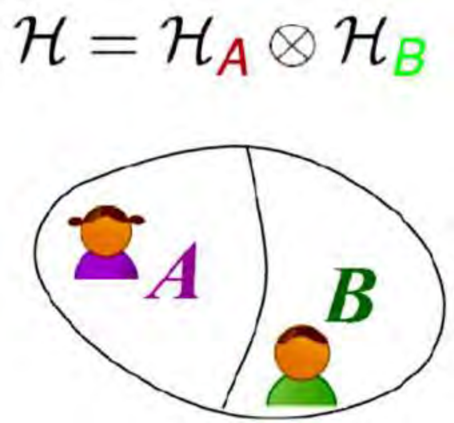

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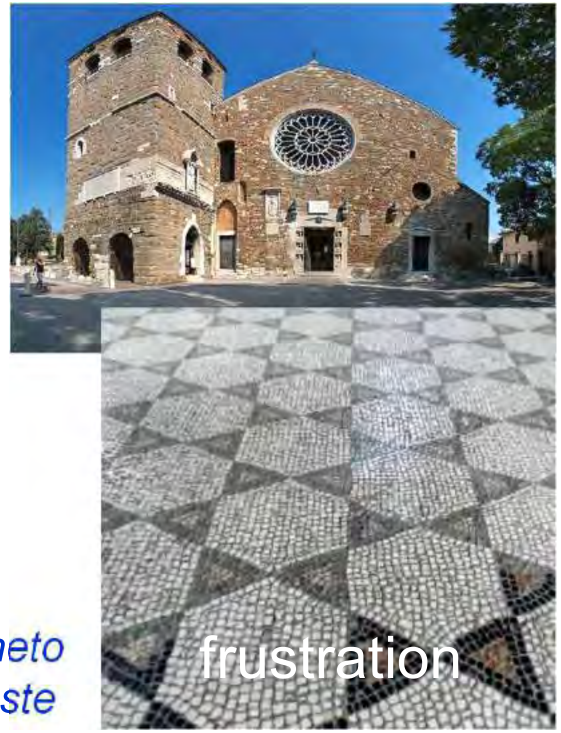
Outline

- **Orbital models: intrinsic quantum frustration**
- **Spin-orbital superexchange: entanglement**
- **Construction of Kugel-Khomskii model: $KCuF_3$**
- **Spin-orbital model for $LaMnO_3$**
- **Spin-orbital models with t_{2g} orbitals; orbital fluctuations**
- **Fractionalization of orbitons; double exchange; perspective**



Mosaic from 12th century, Veneto
Cattedrale di San Giusto, Trieste

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HIPPOCRATICA CIVITAS STUDIUM SALERNI

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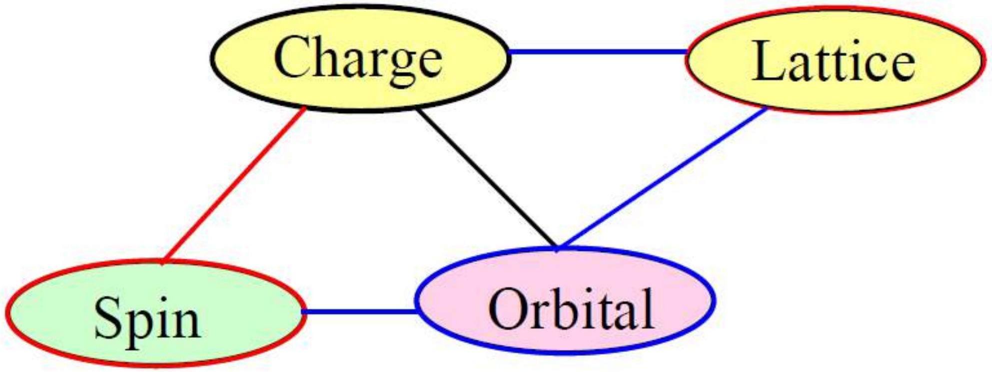
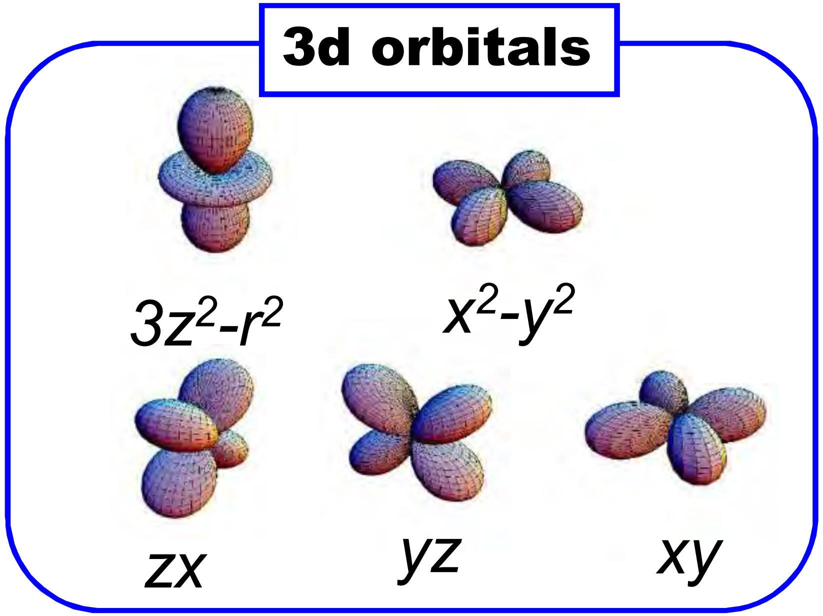
Interplay of **spin, orbital**, charge, lattice degree of freedom



correlated insulators



orbitals are quantum

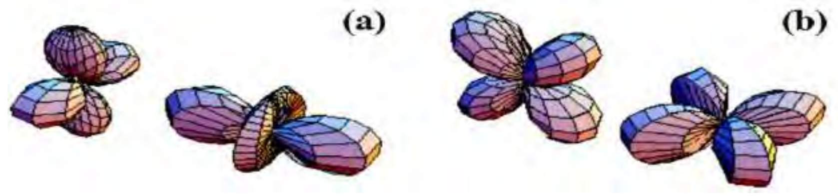
- Superconductivity
- Colossal magnetoresistance (CMR)
- Charge and orbital ordering
- Non-Fermi liquid

Intrinsic frustration of orbital interactions

SU(2) symmetry: $H_{spin} = J \sum_{<ij>} \vec{S}_i \bullet \vec{S}_j$

***e_g systems* (*d^7,d^9*)** *z*-like active

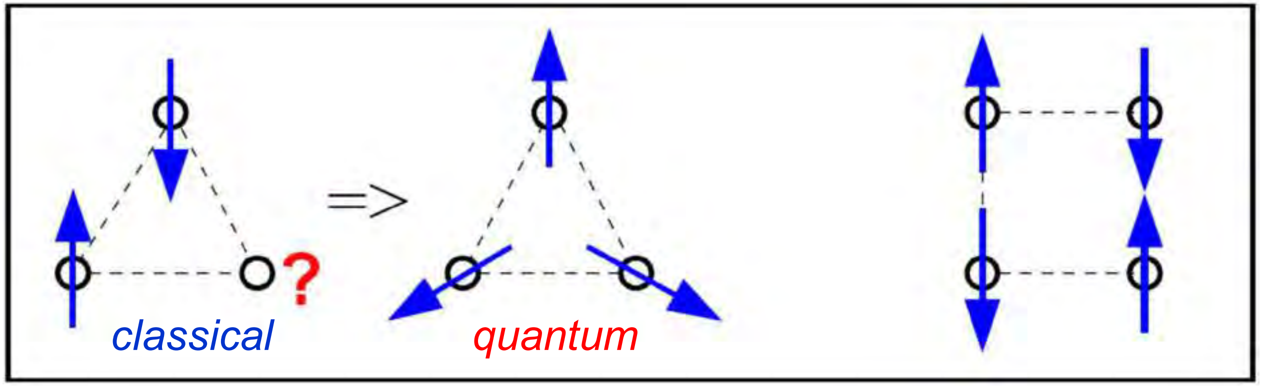
t_{2g} systems (***d^1***, ***d^2, d^4***) two active orbitals along each axis, e.g. *zx* and *xy* along *a* axis



e_g orbitals $\qquad$ t_{2g} orbitals

Orbital interactions are directional !

frustrated kagome lattice



frustrated triangular lattice $\qquad$ order out of disorder $\qquad$ square lattice: no frustration

Cubic symmetry of directional orbital interactions: $H_{orb} = J \sum_{<ij>} T_i^{(\gamma)} T_j^{(\gamma)}$

Interaction depends on the bond direction => ***frustration*** on a square lattice

At large U: from Hubbard to the t-J model

$$H = -t\sum_{\langle ij\rangle,\sigma}(c_{i\sigma}^{\dagger}c_{j\sigma} + \text{H.c.}) + U\sum_{i} n_{i\uparrow}n_{i\downarrow}$$

Canonical transformation
in the regime of $t << U$

Perturbation:
processes P_1HP_2 and P_2HP_1

charge excitations => superexchange

$$d_i^m d_j^m \rightleftharpoons d_i^{m+1} d_j^{m-1}$$

$$P_1HP_1 = t\sum_{ij\sigma}{}'(1-n_{i-\sigma})a_{i\sigma}^{\dagger}a_{j\sigma}(1-n_{j-\sigma})$$

$$P_1HP_2 = t\sum_{ij\sigma}{}'(1-n_{i-\sigma})a_{i\sigma}^{\dagger}a_{j\sigma}n_{j-\sigma},$$

$$P_2HP_1 = t\sum_{ij\sigma}{}'n_{i-\sigma}a_{i\sigma}^{\dagger}a_{j\sigma}(1-n_{j-\sigma}),$$

$$P_2HP_2 = t\sum_{ij\sigma}{}'n_{i-\sigma}a_{i\sigma}^{\dagger}a_{j\sigma}n_{j-\sigma} + U\sum_{i}n_{i\uparrow}n_{i\downarrow}$$

$$\mathscr{H}(\epsilon) = e^{-i\epsilon S}H(\epsilon)e^{i\epsilon S} \quad \text{with} \quad H_1 + i[H_0, S] = 0$$

$$\tilde{H} = \tilde{H}(\epsilon = 1) = H_0 + \tfrac{1}{2}i[H_1, S]$$

(Erik Koch)

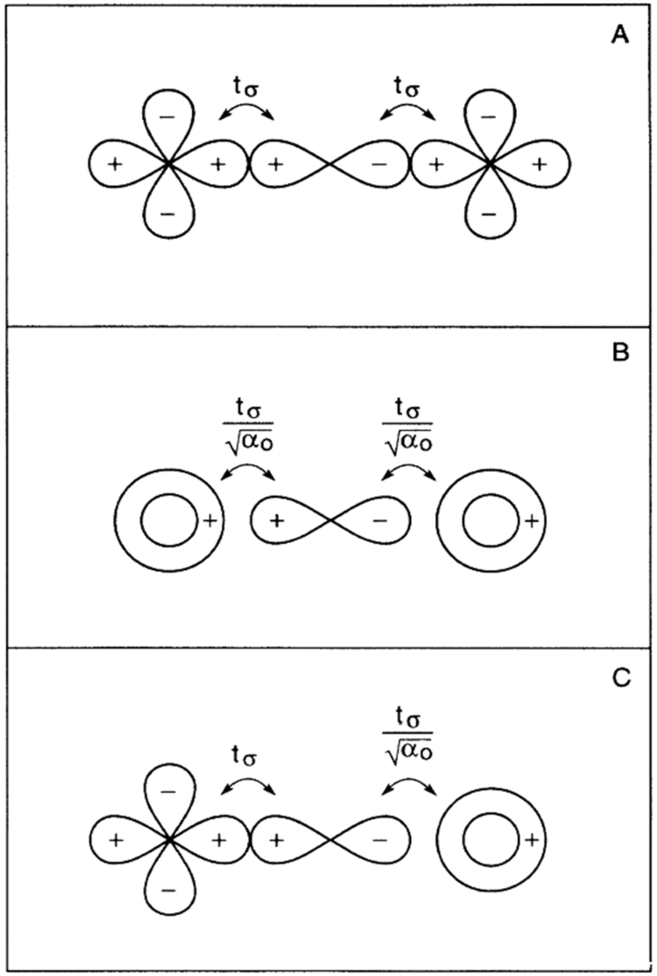
$$H = -t\sum_{\langle ij\rangle,\sigma}(\tilde{c}_{i\sigma}^{\dagger}\tilde{c}_{j\sigma} + \text{H.c.}) + J\sum_{\langle ij\rangle}\left(\mathbf{S}_i\cdot\mathbf{S}_j - \frac{n_i n_j}{4}\right)$$

kinetic exchange

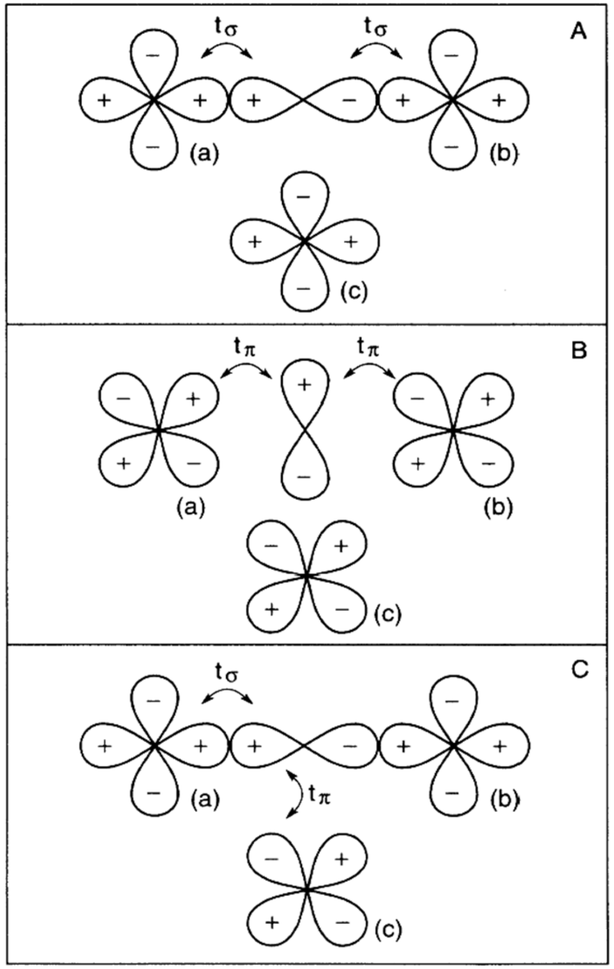
$$J = 4t^2/U$$

[K.A. Chao, J. Spałek, A.M. Oleś, J. Phys. C **10**, L271 (1977)]

Toward superexchange model: hybridization and *d-d* hopping *t*



Effective hopping
t via oxygen
3d-2p-3d
=>
bandwidth W



e_g orbitals

strong

JT coupling

weak

t_{2g} orbitals

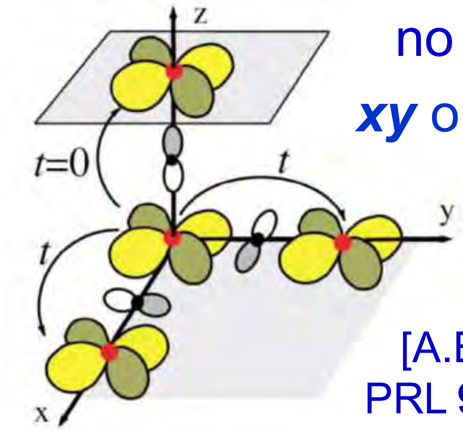
[J. Zaanen and AMO, PRB **48**, 7197 (1993)]

Hopping and orbital superexchange for t_{2g}

In ***t_{2g}* systems** (***d^1,d^2, …***) two states

are active along each cubic axis,
e.g. *yz* & *zx* for the axis *c* –

$$H_t(t_{2g}) = -t \sum_{\alpha} \sum_{\langle ij \rangle \| \gamma \neq \alpha} a^\dagger_{i\alpha\sigma} a_{j\alpha\sigma}$$



no hopping ||*c*
xy orbital called ***c***

[A.B. Harris *et al*., PRL **91**, 087206 (03)]

We introduce convenient notation

$$\boxed{|a\rangle \equiv |yz\rangle, \qquad |b\rangle \equiv |zx\rangle, \qquad |c\rangle \equiv |xy\rangle}$$

Orbital interactions have cubic symmetry

they are described by **quantum** operators:

$$\vec{T}_i = \{T_i^x, T_i^y, T_i^z\} \qquad T_i^x = \tfrac{1}{2}\sigma_i^x,\ T_i^y = \tfrac{1}{2}\sigma_i^y,\ T_i^z = \tfrac{1}{2}\sigma_i^z.$$

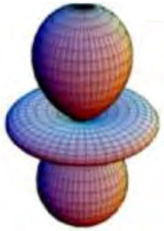
Scalar product $\vec{T}_i \cdot \vec{T}_j$ but for J_H>0 also other terms breaking the „SU(2)" symmetry

Hopping for e_g orbitals

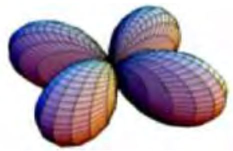
Hamiltonian for e_g electrons couples two directional e_g-orbitals

Real basis:

$$H_t(e_g) = -t \sum_{\alpha} \sum_{\langle ij \rangle \| \alpha, \sigma} a^{\dagger}_{i\zeta_\alpha \sigma} a_{j\zeta_\alpha \sigma}$$



$$|z\rangle \equiv \frac{1}{\sqrt{6}}(3z^2 - r^2), \qquad |\bar{z}\rangle \equiv \frac{1}{\sqrt{2}}(x^2 - y^2)$$



$$H_t^{\uparrow}(e_g) = -\frac{1}{4} t \sum_{\langle ij \rangle \| ab} \left[3a^{\dagger}_{i\bar{z}} a_{j\bar{z}} + a^{\dagger}_{iz} a_{jz} \mp \sqrt{3} \left(a^{\dagger}_{i\bar{z}} a_{jz} + a^{\dagger}_{iz} a_{j\bar{z}} \right) \right] - t \sum_{\langle ij \rangle \| c} a^{\dagger}_{iz} a_{jz}$$

complex e_g orbitals $\quad |j+\rangle = \frac{1}{\sqrt{2}}(|jz\rangle - i|j\bar{z}\rangle), \qquad |j-\rangle = \frac{1}{\sqrt{2}}(|jz\rangle + i|j\bar{z}\rangle)$

e_g electrons with only one spin flavor $\sigma = \uparrow$ (FM manganites)

$$\mathcal{H}^{\uparrow}(e_g) = -\frac{1}{2} t \sum_{\alpha} \sum_{\langle ij \rangle \| \alpha} \left[\left(a^{\dagger}_{i+} a_{j+} + a^{\dagger}_{i-} a_{j-} \right) + \gamma \left(e^{-i\chi_\alpha} a^{\dagger}_{i+} a_{j-} + e^{+i\chi_\alpha} a^{\dagger}_{i-} a_{j+} \right) \right]$$

with $\chi_a = +2\pi/3$, $\chi_b = -2\pi/3$, and $\chi_c = 0$ $\quad$ has cubic symmetry

Correl17 $\quad$ with interaction $\quad \bar{U} \sum_i n_{i+} n_{i-}$ => *orbital Hubbard model*

Intraatomic Coulomb interactions for 3*d* orbitals

$$\begin{aligned} H_{int} &= U\sum_{i\alpha} n_{i\alpha\uparrow}n_{i\alpha\downarrow} + \sum_{i,\alpha<\beta}\left(U_{\alpha\beta}-\frac{1}{2}J_{\alpha\beta}\right) n_{i\alpha}n_{i\beta} - 2\sum_{i,\alpha<\beta} J_{\alpha\beta}\,\vec{S}_{i\alpha}\cdot\vec{S}_{i\beta} \\ &+ \sum_{i,\alpha<\beta} J_{\alpha\beta}\left(a^{\dagger}_{i\alpha\uparrow}a^{\dagger}_{i\alpha\downarrow}a_{i\beta\downarrow}a_{i\beta\uparrow} + a^{\dagger}_{i\beta\uparrow}a^{\dagger}_{i\beta\downarrow}a_{i\alpha\downarrow}a_{i\alpha\uparrow}\right). \end{aligned}$$

$$\boxed{U = U_{\alpha\beta} + 2J_{\alpha\beta}}$$

rotational invariance

Intraorbital Coulomb $\boxed{U = A + 4B + 3C}$

On-site interorbital exchange elements $J_{\alpha\beta}$ for 3d orbitals

$3d$ orbital	xy	yz	zx	x^2-y^2	$3z^2-r^2$
xy	0	$3B+C$	$3B+C$	C	$4B+C$
yz	$3B+C$	0	$3B+C$	$3B+C$	$B+C$
zx	$3B+C$	$3B+C$	0	$3B+C$	$B+C$
x^2-y^2	C	$3B+C$	$3B+C$	0	$4B+C$
$3z^2-r^2$	$4B+C$	$B+C$	$B+C$	$4B+C$	0

Hund's exchange

$$\begin{aligned} J_H^t &= 3B + C, \\ J_H^e &= 4B + C. \end{aligned}$$

Orbital operators: e_g

define:

$$
\begin{aligned}
|i\vartheta\rangle &= \cos(\vartheta/2)\,|iz\rangle - \sin(\vartheta/2)\,|i\bar{z}\rangle \\
|i\bar{\vartheta}\rangle &= \sin(\vartheta/2)\,|iz\rangle + \cos(\vartheta/2)\,|i\bar{z}\rangle
\end{aligned}
$$

For angles $\vartheta = \pm 4\pi/3$ one finds equivalent pairs $\{|i\zeta_a\rangle, |i\xi_a\rangle\}$ and $\{|i\zeta_b\rangle, |i\xi_b\rangle\}$

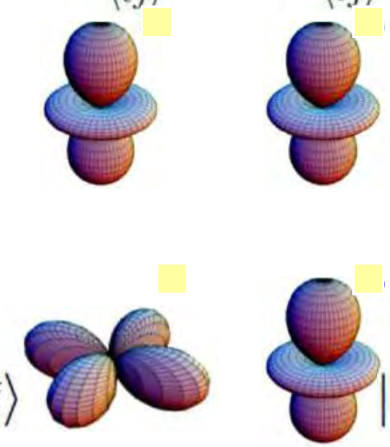
Local projection operators:

$$
\mathcal{P}^{\gamma}_{i\zeta} = |i\zeta_\gamma\rangle\langle i\zeta_\gamma| = \left(\frac{1}{2} + \tau_i^{(\gamma)}\right), \qquad \mathcal{P}^{\gamma}_{i\xi} = |i\xi_\gamma\rangle\langle i\xi_\gamma| = \left(\frac{1}{2} - \tau_i^{(\gamma)}\right)
$$

$$
\tau_i^{(\gamma)} \equiv \frac{1}{2}\left(|i\zeta_\gamma\rangle\langle i\zeta_\gamma| - |i\xi_\gamma\rangle\langle i\xi_\gamma|\right)
$$

$$
\boxed{\tau_i^{(a)} = -\frac{1}{4}\left(\sigma_i^z - \sqrt{3}\sigma_i^x\right), \qquad \tau_i^{(b)} = -\frac{1}{4}\left(\sigma_i^z + \sqrt{3}\sigma_i^x\right), \qquad \tau_i^{(c)} = \frac{1}{2}\sigma_i^z}
$$

c axis: $\mathcal{P}^{(\gamma)}_{\langle ij\rangle}$ $\mathcal{Q}^{(\gamma)}_{\langle ij\rangle}$



$|i\bar{z}\rangle$ $|iz\rangle$

Bond projection operators:

$$
\begin{aligned}
\mathcal{P}^{(\gamma)}_{\langle ij\rangle} &\equiv \left(\frac{1}{2} + \tau_i^{(\gamma)}\right)\left(\frac{1}{2} - \tau_j^{(\gamma)}\right) + \left(\frac{1}{2} - \tau_i^{(\gamma)}\right)\left(\frac{1}{2} + \tau_j^{(\gamma)}\right), \\
\mathcal{Q}^{(\gamma)}_{\langle ij\rangle} &\equiv 2\left(\frac{1}{2} + \tau_i^{(\gamma)}\right)\left(\frac{1}{2} + \tau_j^{(\gamma)}\right).
\end{aligned}
$$

Orbital superexchange for e_g orbitals

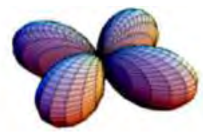
The hopping t couples two directional orbitals $\{|i\zeta_\gamma\rangle, |j\zeta_\gamma\rangle\}$

Consider $d_i^m d_j^m \rightleftharpoons d_i^{m+1} d_j^{m-1}$ high spin (S=1) excitations:

$$\varepsilon_1 \equiv E_1(d^{m+1}) + E_0(d^{m-1}) - 2E_0(d^m) = U - 3J_H = \bar{U}$$

effective U

$|\zeta_\gamma\rangle$



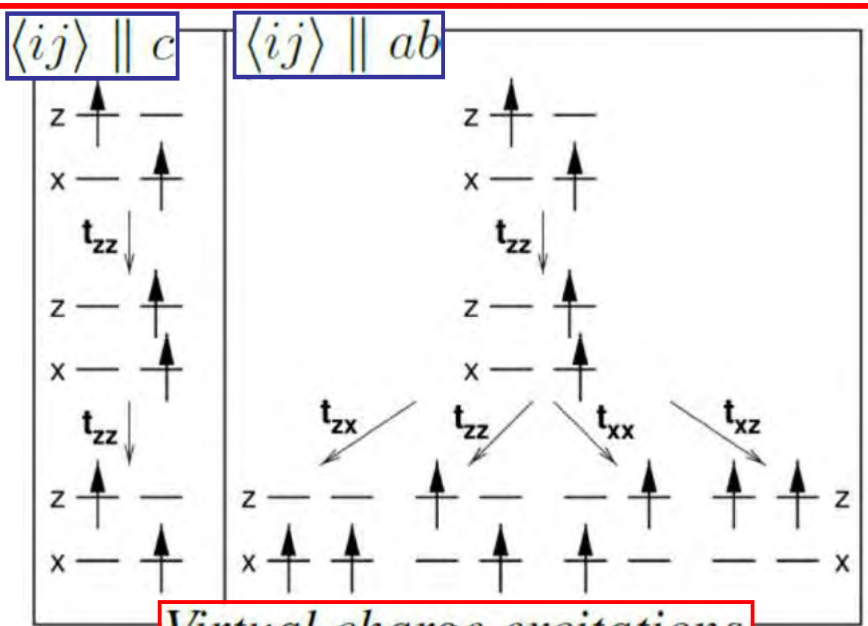
$|\xi_\gamma\rangle$

$$\boxed{\mathcal{H}^{\uparrow}(e_g) = -\frac{t^2}{\varepsilon_1} \sum_{\langle ij\rangle \parallel \gamma} \mathcal{P}^{(\gamma)}_{\langle ij\rangle}}$$

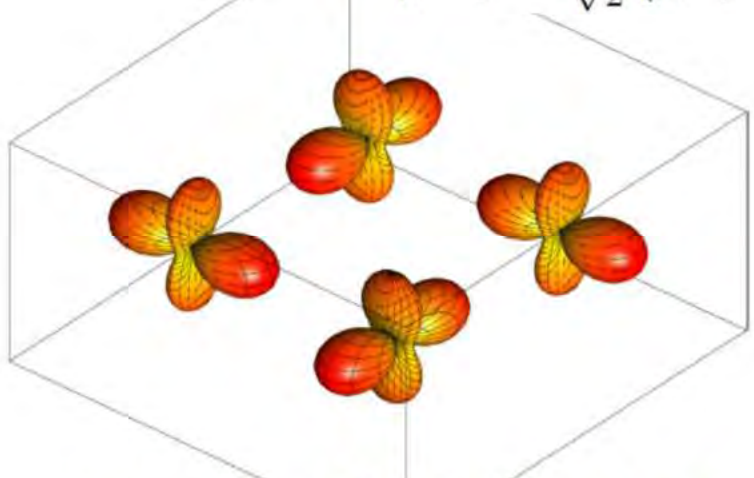
$$\boxed{\mathcal{H}^{\uparrow}(e_g) = \frac{1}{2} J r_1 \sum_{\langle ij\rangle \parallel \gamma} \left(\tau_i^{(\gamma)} \tau_j^{(\gamma)} - \frac{1}{4} \right) + E_z \sum_i \tau_i^{(c)}}$$

$$|i+\rangle = \frac{1}{\sqrt{2}}\left(|iz\rangle + |i\bar{z}\rangle\right)$$

$$|j-\rangle = \frac{1}{\sqrt{2}}\left(|jz\rangle - |j\bar{z}\rangle\right)$$



Virtual charge excitations



AO state of $|\mathrm{AO}\pm\rangle$ *orbitals*

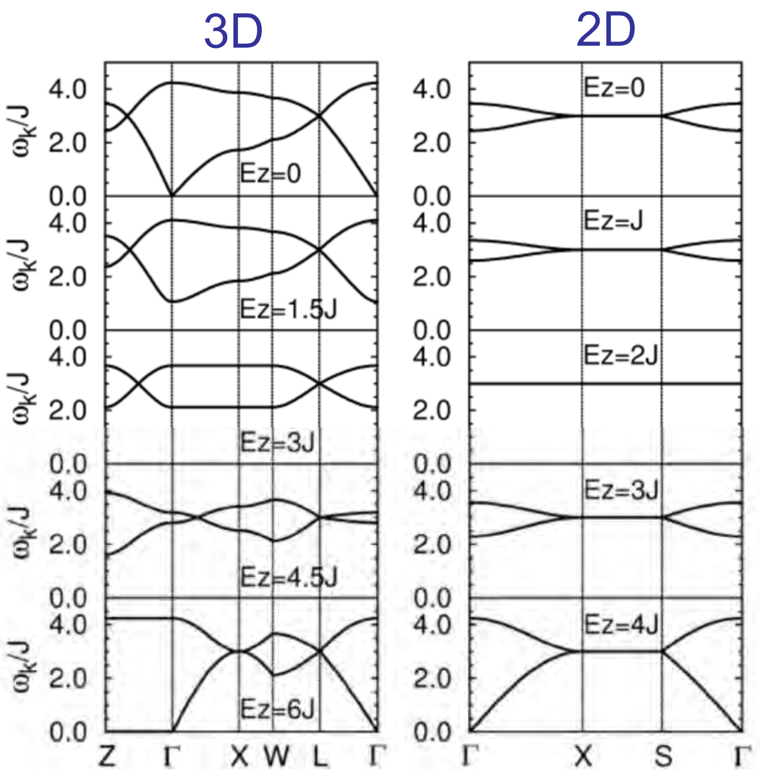
Orbital waves and gap for e_g orbitals

$$|\Phi_0\rangle = \prod_{i\in A} |i\theta_A\rangle \prod_{j\in B} |j\theta_B\rangle$$

$$\mathcal{H}^{\uparrow}(e_g) = \frac{1}{2} J r_1 \sum_{\langle ij\rangle \| \gamma} \left(\tau_i^{(\gamma)} \tau_j^{(\gamma)} - \frac{1}{4} \right) + E_z \sum_i \tau_i^{(c)}$$

$$\begin{aligned} |i\theta_A\rangle &= \cos(\theta/2)\,|iz\rangle + \sin(\theta/2)\,|ix\rangle, \\ |j\theta_B\rangle &= \cos(\theta/2)\,|jz\rangle - \sin(\theta/2)\,|jx\rangle \end{aligned}$$

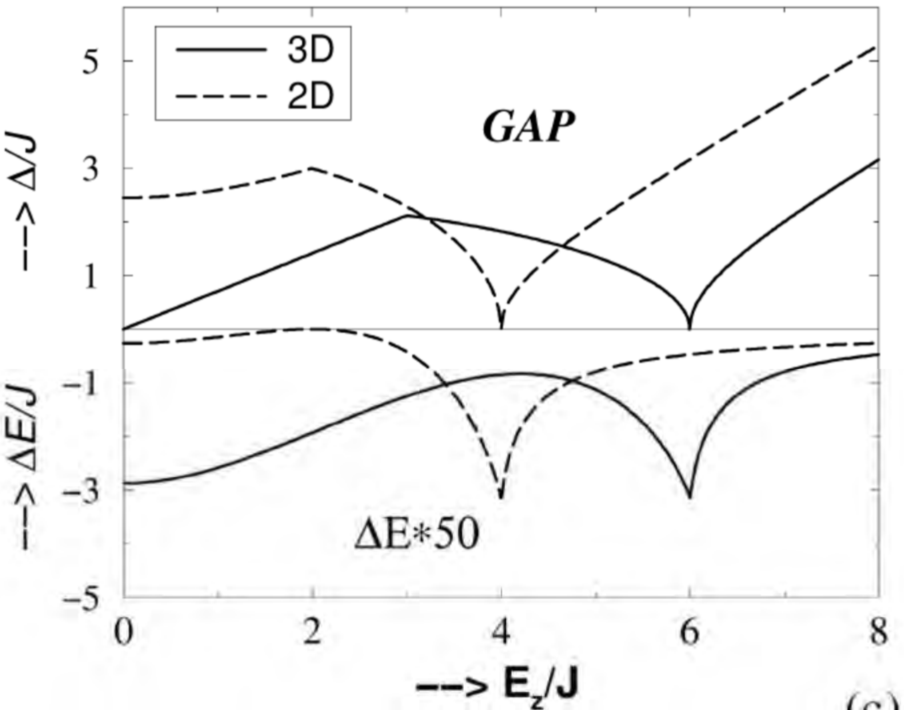
Two sublattices – orbital waves as for a quantum antiferromagnet



(a-b)

Orbital-wave excitations

The gap vanishes at $E_z = zJ$ and in 3D for $E_z = 0$



(c)

Quantum corrections are small

[J. van den Brink et al., PRB **59**, 6795 (99)]

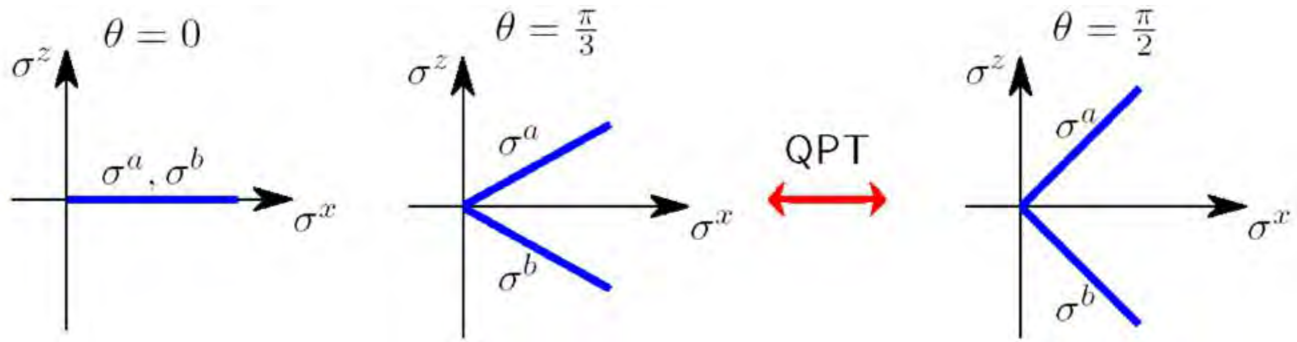
2D model with increasing frustration: from Ising to compass

From Ising to compass:

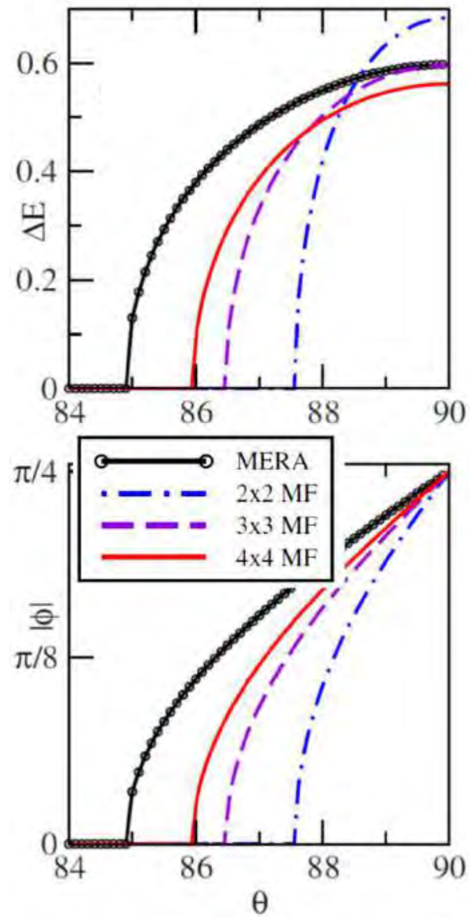
$$\mathcal{H}(\theta) = -J_{\rm cm} \sum_{\{ij\}\in ab} \left\{ \sigma^a_{ij}(\theta)\sigma^a_{i+1,j}(\theta) + \sigma^b_{ij}(\theta)\sigma^b_{i,j+1}(\theta) \right\}$$

$$\begin{aligned} \sigma^a_{ij} &= \cos(\theta/2)\ \sigma^x_{ij} + \sin(\theta/2)\ \sigma^z_{ij} \\ \sigma^b_{ij} &= \cos(\theta/2)\ \sigma^x_{ij} - \sin(\theta/2)\ \sigma^z_{ij} \end{aligned}$$

$$\Delta E(\theta) = \left| \langle \sigma^a_{ij}(\theta)\sigma^a_{i+1,j}(\theta)\rangle - \langle \sigma^b_{ij}(\theta)\sigma^b_{i,j+1}(\theta)\rangle \right|$$



Ising **e_g orbital** **compass**



A QPT from **2D Ising** order to **1D compass nematic order** at $\theta \sim 85$ degrees

$$\phi = \arctan\left(\frac{M^z}{M^x}\right)$$

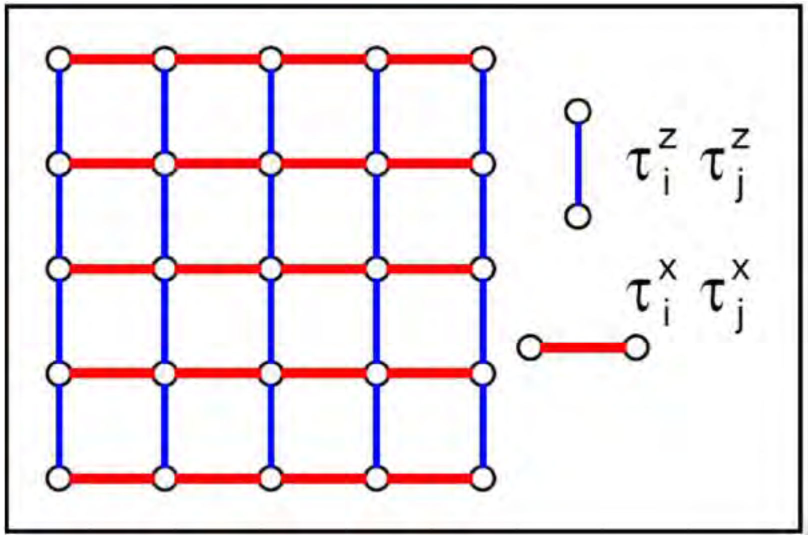
2D compass

$$\mathcal{H}(\pi/2) = -J_{\rm cm} \sum_{\langle ij\rangle \| a} \sigma^x_{ij}\sigma^x_{i+1,j} - J_{\rm cm} \sum_{\langle ij\rangle \| b} \sigma^z_{ij}\sigma^z_{i,j+1}$$

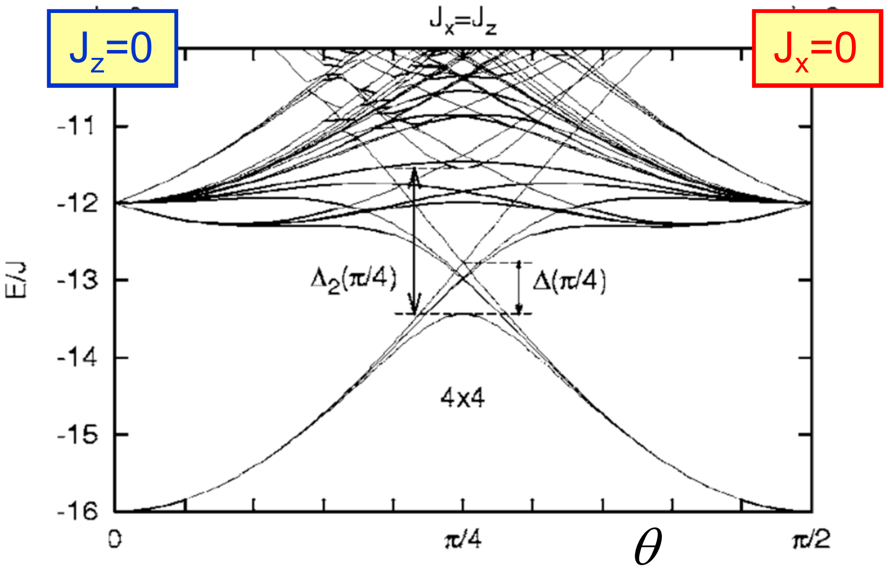
Frustration in the 2D quantum compass model

Orbital (pseudospin) model with competing Ising-like interactions:

$$H = -\sum_{r} \left(J_x \tau_r^x \tau_{r+e_x}^x + J_z \tau_r^z \tau_{r+e_z}^z \right)$$



Artist's view of the *LxL* cluster
for the 2D compass model (*L*=4)



Low-energy states for the 4*x*4
cluster for $J_x = J\cos\theta$, $J_z = J\sin\theta$

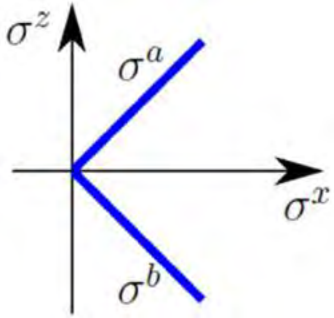
High $2x2^L$ degeneracy at the isotropic J_x=J_z point

Model for quantum qubits => quantum computation

Correl17

[J. Dornier, F. Becca, F. Mila, PRB **72**, 024448 (05)]

Order at finite T in 2D compass model



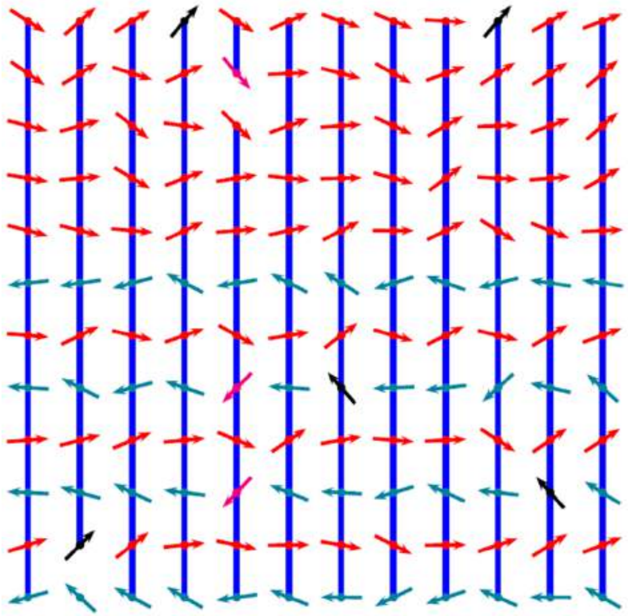
$$H = -\frac{1}{4} J_x \sum_j X_j X_{j+e_a} - \frac{1}{4} J_z \sum_j Z_j Z_{j+e_b}$$

compass model

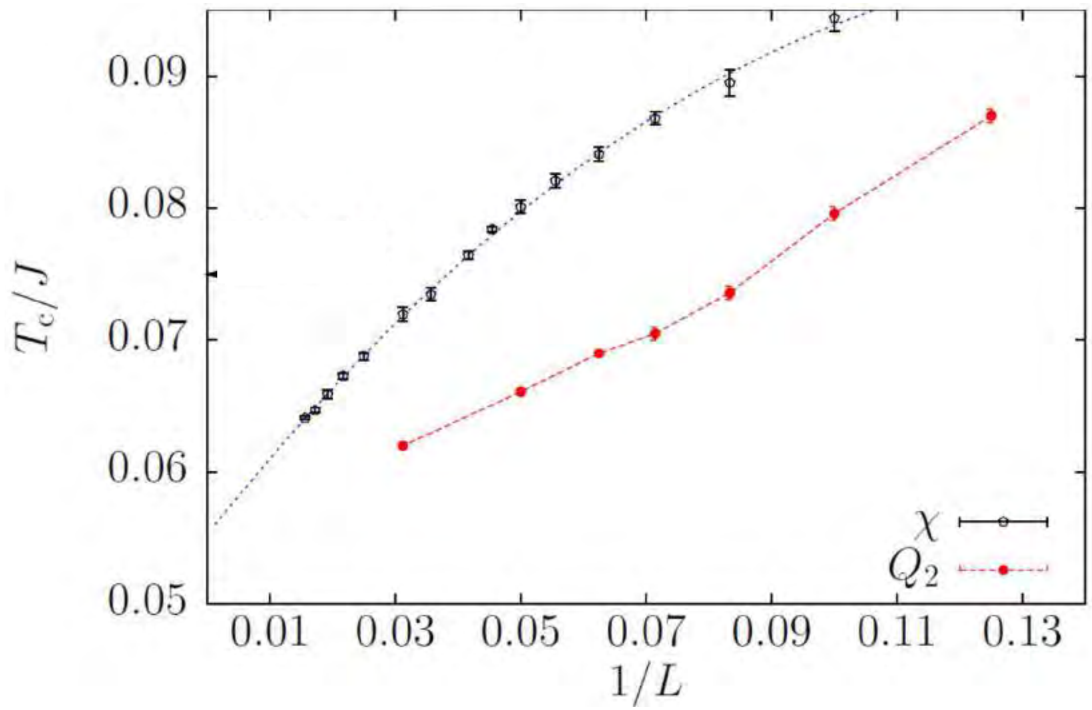
$X_j \equiv \sigma_j^x$ and $Z_j \equiv \sigma_j^z$ are Pauli matrices

$$Q \equiv |\langle Q_j \rangle| = |\langle X_j X_{j+e_a} - Z_j Z_{j+e_b} \rangle|$$

nematic order

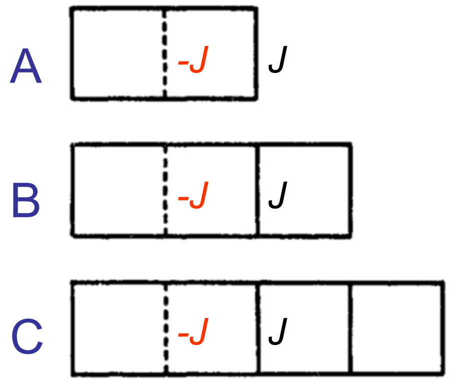


Ordered **nematic phase** in
2D FM classical compass model
CMC for 12x12 lattice at T =0.1J



Interpolation in the 2D compass model from QMC

[S. Wenzel and W. Janke, PRB **78**, 064402 (08)]



Classical vs quantum for 2D frustrated models

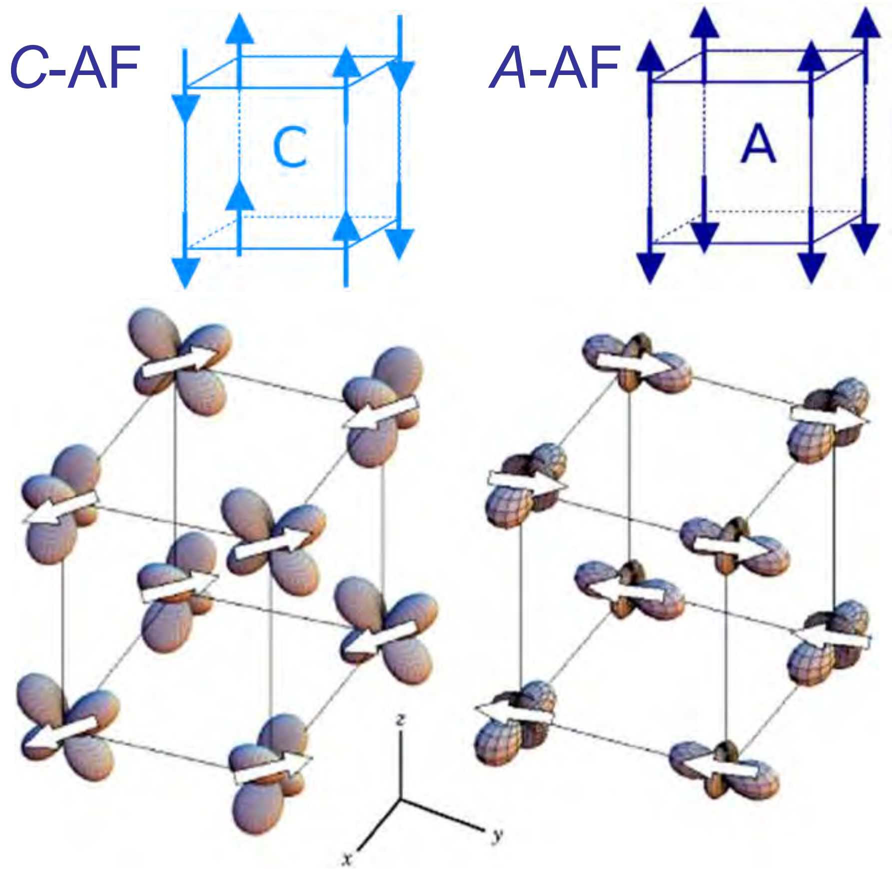
$$\boxed{\frac{1}{4}\sigma_i^z\sigma_j^z} \Rightarrow \boxed{T_c^{\text{Ising}} = \frac{1}{2\log(1+\sqrt{2})} \approx 0.567296.}$$

TABLE I. The critical temperature $\mathcal{T}_c$ and the type of order for the classical and quantum models on a square lattice: Ising model, $\frac{1}{2}$ and $\frac{2}{3}$ frustrated Ising [29], fully frustrated Villain model [30], e_g orbital model [23] and 2D compass model [22].

2D model	order	$\mathcal{T}_c/J$	method	interactions
Ising	2D	0.567296	exact	Onsager
$\frac{1}{2}$ frustrated	2D	0.410	exact	C
$\frac{2}{3}$ frustrated	2D	0.342	exact	B
Villain	—	0.0	exact	A
e_g orbital	2D	0.3566 ± 0.0001	T>0 tensor network	$\propto \frac{3}{16}\sigma_i^x\sigma_j^x$
compass	nematic	0.0606 ± 0.0004		$\frac{1}{4}\sigma_i^z\sigma_j^z$

Spin-orbital physics

AF phases with some **FM** bonds



$LaVO_3$ — t_{2g} orbitals

$LaMnO_3$ — e_g orbitals

Frustration can be removed

Simplest case:
Mott insulators with
spin-orbital order

manganites, nickelates,
vanadates, titanates, …

Goodenough-Kanamori rules:

AO order supports **FM** spin order

FO order supports **AF** spin order

Are these rules sufficient?

Qualitative changes due to
spin-orbital entanglement

Inventors of spin-orbital physics at Blois (06)

[4] K.I. Kugel and D.I. Khomskii, Sov. Phys. Usp. **25**, 231 (1982)

Degenerate Hubbard model & charge excitations (*t<<U*)

Two parameters: ***U*** – intraorbital Coulomb interaction, ***J_H*** – Hund's exchange

$$H_{\text{int}} = U\sum_{i\alpha} n_{i\alpha\uparrow}n_{i\alpha\downarrow} + (U - \tfrac{5}{2}J_H)\sum_{i,\alpha<\beta} n_{i\alpha}n_{i\beta} - 2J_H\sum_{i,\alpha<\beta}\vec{S}_{i\alpha}\cdot\vec{S}_{i\beta}$$
$$+ J_H\sum_{i,\alpha<\beta}(d^{+}_{i\alpha\uparrow}d^{+}_{i\alpha\downarrow}d_{i\beta\downarrow}d_{i\beta\uparrow} + d^{+}_{i\beta\uparrow}d^{+}_{i\beta\downarrow}d_{i\alpha\downarrow}d_{i\alpha\uparrow})$$

[A.M. Oleś, PRB **28**, 327 (1983)]

In a Mott insulator (t<<U) superexchange follows from charge excitations

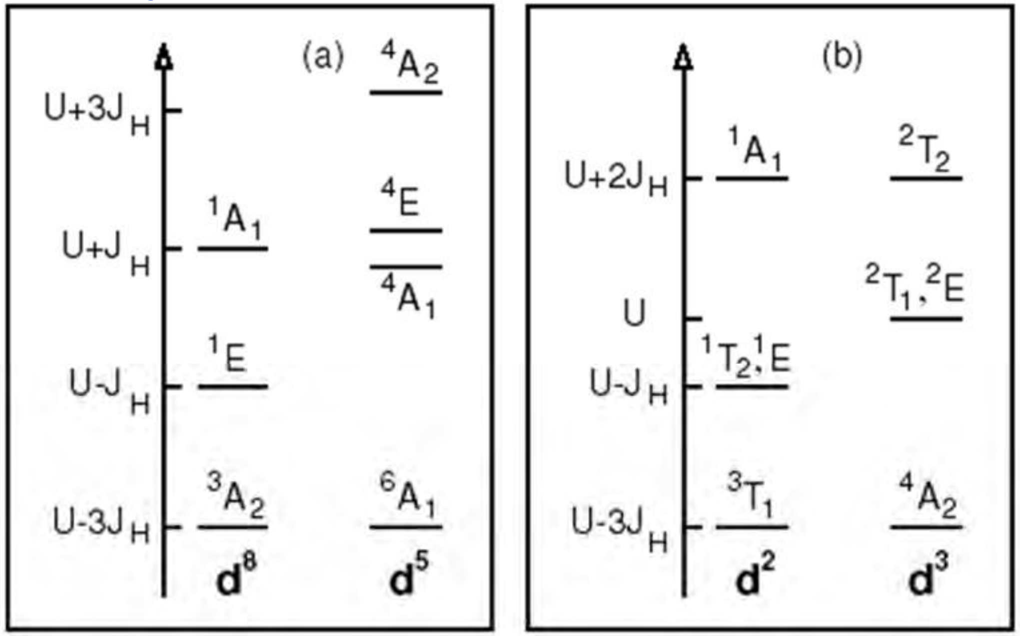
$$d_i^m d_j^m \rightleftharpoons d_i^{m+1} d_j^{m-1}$$

single parameter:

$\eta = J_H/U$

Correl17

e_g systems t_{2g} systems



LS

HS

FIG. 1: Energies of $d_i^m d_j^m \to d_i^{m+1} d_j^{m-1}$ charge excitations

Low energy Hamiltonian: **Spin-orbital superexchange** ($t<<U$)

Two parameters: ***U*** – intraorbital Coulomb interaction, J_H – Hund's exchange

At large $U >> t$ ($J=4t^2/U$):

$\eta = J_H/U$

charge excitation $\varepsilon_n = E_n(d^{m+1}) + E_0(d^{m-1}) - 2E_0(d^m)$

$P_{\langle ij\rangle}(\mathcal{S})$ is the projection on the total spin $\mathcal{S} = S \pm \frac{1}{2}$

$\mathcal{O}^{\gamma}_{\langle ij\rangle}$ is the projection operator on the orbital state

spin interactions: **SU(2) symmetry**

=> superexchange

$$\mathcal{H} = -\sum_n \frac{t^2}{\varepsilon_n} \sum_{\langle ij\rangle \| \gamma} P_{\langle ij\rangle}(\mathcal{S})\, \mathcal{O}^{\gamma}_{\langle ij\rangle}$$

$$\mathcal{H}_J = J \sum_{\gamma} \sum_{\langle ij\rangle \| \gamma} \left\{ \hat{\mathcal{K}}^{(\gamma)}_{ij} \left(\vec{S}_i \cdot \vec{S}_j + S^2 \right) + \hat{\mathcal{N}}^{(\gamma)}_{ij} \right\}$$

spin-orbital model

contains orbital operators $\hat{\mathcal{K}}^{(\gamma)}_{ij}$ and $\hat{\mathcal{N}}^{(\gamma)}_{ij}$ of **cubic symmetry** (γ =a,b,c)

Averaging over orbital (dis)ordered state => anisotropic ***spin model***:

$$H_s = J_c \sum_{\langle ij\rangle_c} S_i \cdot S_j + J_{ab} \sum_{\langle ij\rangle_{ab}} S_i \cdot S_j$$

$$J_\gamma \equiv \left\langle J^{(\gamma)}_{ij} \right\rangle$$

Here spin and orbital operators are **disentangled**

Kugel-Khomskii model

$$d_i^9 d_j^9 \rightleftharpoons d_i^{10} d_j^8$$

We follow the general scheme:

$$\mathcal{H} = -\sum_n \frac{t^2}{\varepsilon_n} \sum_{\langle ij\rangle \parallel \gamma} P_{\langle ij\rangle}(\mathcal{S})\, \mathcal{O}^{\gamma}_{\langle ij\rangle}$$

$$\begin{aligned}\mathcal{H}(d^9) \;=\; \sum_{\gamma}\sum_{\langle ij\rangle\parallel\gamma} \Bigg\{ & -\frac{t^2}{U-3J_H}\left(\vec{S}_i\cdot\vec{S}_j+\frac{3}{4}\right)\mathcal{P}^{(\gamma)}_{\langle ij\rangle} + \frac{t^2}{U-J_H}\left(\vec{S}_i\cdot\vec{S}_j-\frac{1}{4}\right)\mathcal{P}^{(\gamma)}_{\langle ij\rangle} \\ & + \left(\frac{t^2}{U-J_H}+\frac{t^2}{U+J_H}\right)\left(\vec{S}_i\cdot\vec{S}_j-\frac{1}{4}\right)\mathcal{Q}^{(\gamma)}_{\langle ij\rangle}\Bigg\} + E_z\sum_i \tau_i^c. \qquad (44)\end{aligned}$$

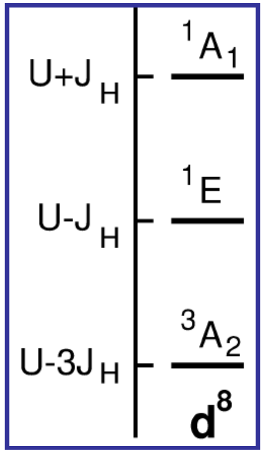
KK model

crystal field $\propto E_z$

with

$$\begin{aligned}\mathcal{P}^{(\gamma)}_{\langle ij\rangle} &\equiv \left(\frac{1}{2}+\tau_i^{(\gamma)}\right)\left(\frac{1}{2}-\tau_j^{(\gamma)}\right)+\left(\frac{1}{2}-\tau_i^{(\gamma)}\right)\left(\frac{1}{2}+\tau_j^{(\gamma)}\right), \\ \mathcal{Q}^{(\gamma)}_{\langle ij\rangle} &\equiv 2\left(\frac{1}{2}+\tau_i^{(\gamma)}\right)\left(\frac{1}{2}+\tau_j^{(\gamma)}\right).\end{aligned}$$

ε_n

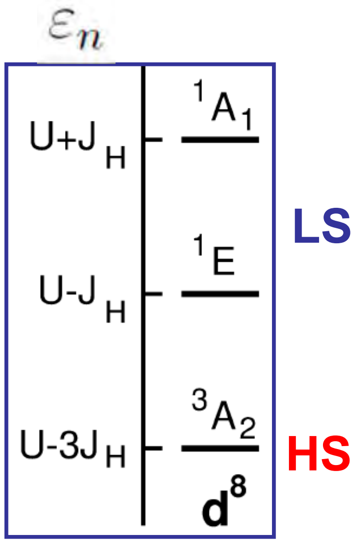


charge excitation			spin state		orbital state	
n	type	ε_n	$\mathcal{S}$	$P_{\langle ij\rangle}(\mathcal{S})$	orbitals on $\langle ij\rangle \parallel \gamma$	projection
1	HS	$U-3J_H$	1	$\left(\vec{S}_i\cdot\vec{S}_j+\frac{3}{4}\right)$	$\lvert i\zeta_\gamma\rangle\lvert j\xi_\gamma\rangle\;(\lvert i\xi_\gamma\rangle\lvert j\zeta_\gamma\rangle)$	$\mathcal{P}^{(\gamma)}_{\langle ij\rangle}$
2	LS	$U-J_H$	0	$-\left(\vec{S}_i\cdot\vec{S}_j-\frac{1}{4}\right)$	$\lvert i\zeta_\gamma\rangle\lvert j\xi_\gamma\rangle\;(\lvert i\xi_\gamma\rangle\lvert j\zeta_\gamma\rangle)$	$\mathcal{P}^{(\gamma)}_{\langle ij\rangle}$
3	LS	$U-J_H$	0	$-\left(\vec{S}_i\cdot\vec{S}_j-\frac{1}{4}\right)$	$\lvert i\zeta_\gamma\rangle\lvert j\zeta_\gamma\rangle$	$\mathcal{Q}^{(\gamma)}_{\langle ij\rangle}$
4	LS	$U+J_H$	0	$-\left(\vec{S}_i\cdot\vec{S}_j-\frac{1}{4}\right)$	$\lvert i\zeta_\gamma\rangle\lvert j\zeta_\gamma\rangle$	$\mathcal{Q}^{(\gamma)}_{\langle ij\rangle}$

Kugel-Khomskii model

Equidistant multiplet structure for d^8 ions

$\eta = J_H/U$



Charge excitations fully characterized by:

$$r_1 = \frac{1}{1-3\eta}, \qquad r_2 = r_3 = \frac{1}{1-\eta}, \qquad r_4 = \frac{1}{1+\eta}$$

$$J = 4t^2/U$$

$$\mathcal{H}(d^9) = \frac{1}{2}J\sum_\gamma \sum_{\langle ij\rangle\parallel\gamma}\left\{\left[-r_1\left(\vec{S}_i\cdot\vec{S}_j+\frac{3}{4}\right)+r_2\left(\vec{S}_i\cdot\vec{S}_j-\frac{1}{4}\right)\right]\left(\frac{1}{4}-\tau_i^{(\gamma)}\tau_j^{(\gamma)}\right)\right.$$
$$\left.+(r_3+r_4)\left(\vec{S}_i\cdot\vec{S}_j-\frac{1}{4}\right)\left(\tau_i^{(\gamma)}+\frac{1}{2}\right)\left(\tau_j^{(\gamma)}+\frac{1}{2}\right)\right\}+E_z\sum_i \tau_i^c.$$

KK model

Experimental observations:

K_2CuF_4 — the FM spin phase

$KCuF_3$ finite Hund's exchange η favors AO order stabilizing A-AF

$KCuF_3$ exhibits spinon excitations for $T > T_N$

Correl17

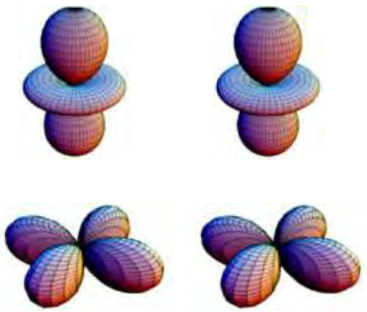
In this regime behaves as the 1D quantum antiferromagnet

Which kind of spin-orbital order ?

Strong anisotropy for ***G*-AF**

$|z\rangle$

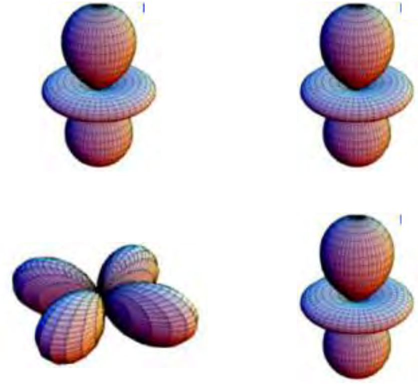
$|\bar{z}\rangle$



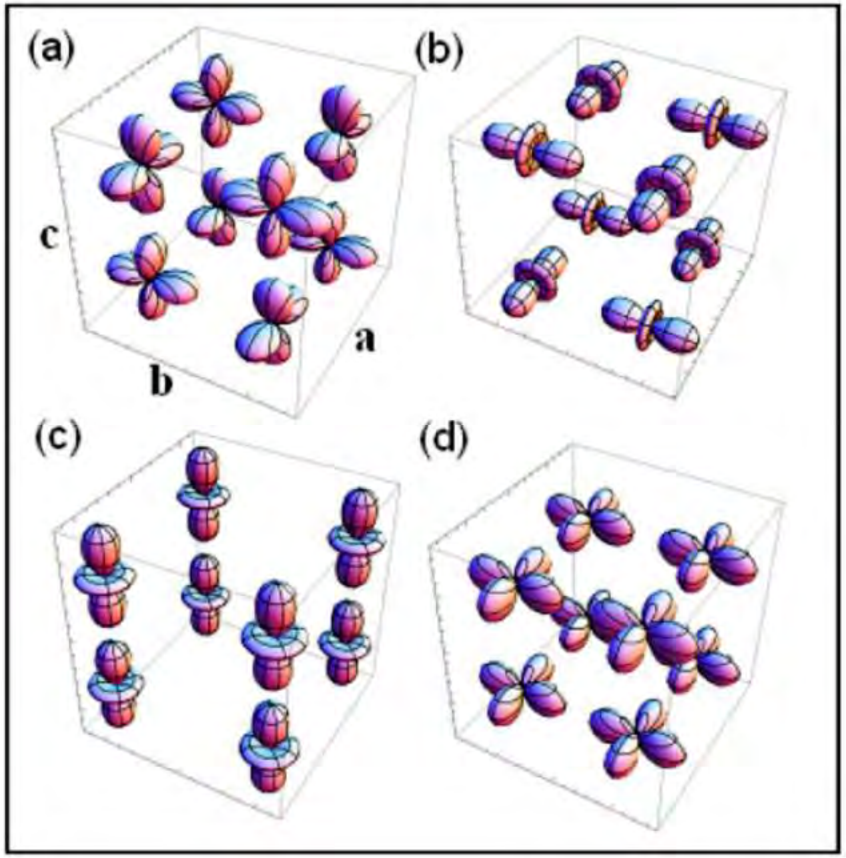
$$J^{z}_{ab} = \frac{1}{16} J$$

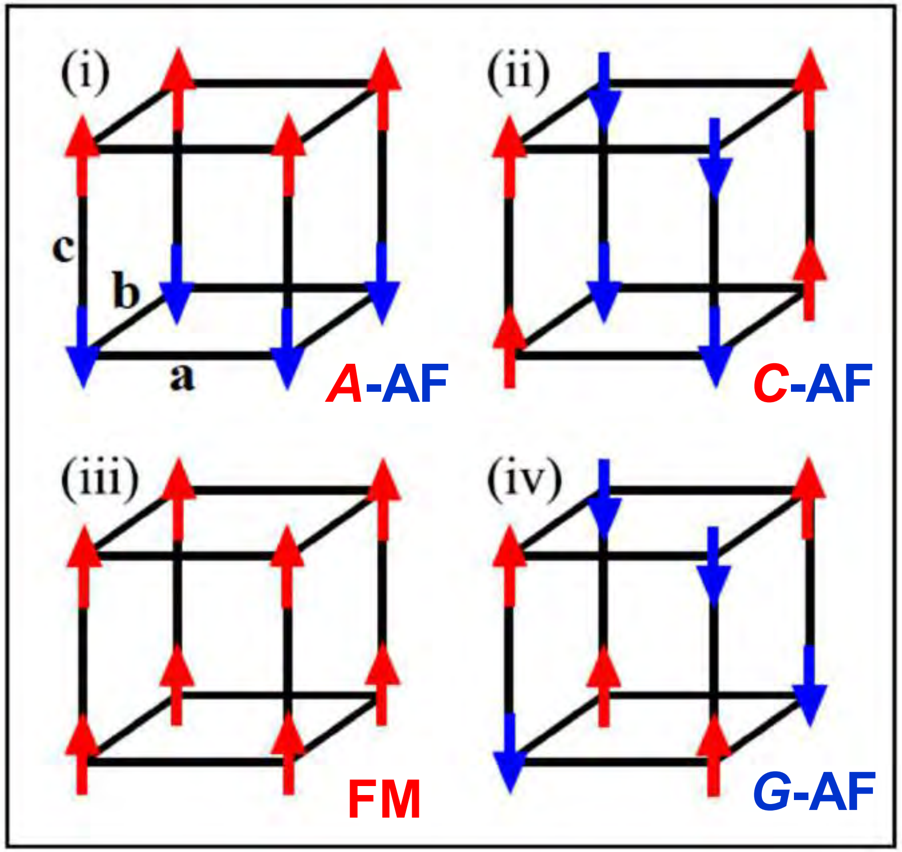
$$J^{\bar{z}}_{ab} = \frac{9}{16} J$$

spin exchange



FM **AF**





Mean field analysis of spin-orbital order

Two-sublattice ground state: $$|\Phi_0\rangle = \prod_{i\in A} |i\theta_A\rangle \prod_{j\in B} |j\theta_B\rangle$$

$$\begin{aligned} |i\theta_A\rangle &= \cos(\theta/2)\,|iz\rangle + \sin(\theta/2)\,|ix\rangle, \\ |j\theta_B\rangle &= \cos(\theta/2)\,|jz\rangle - \sin(\theta/2)\,|jx\rangle \end{aligned}$$

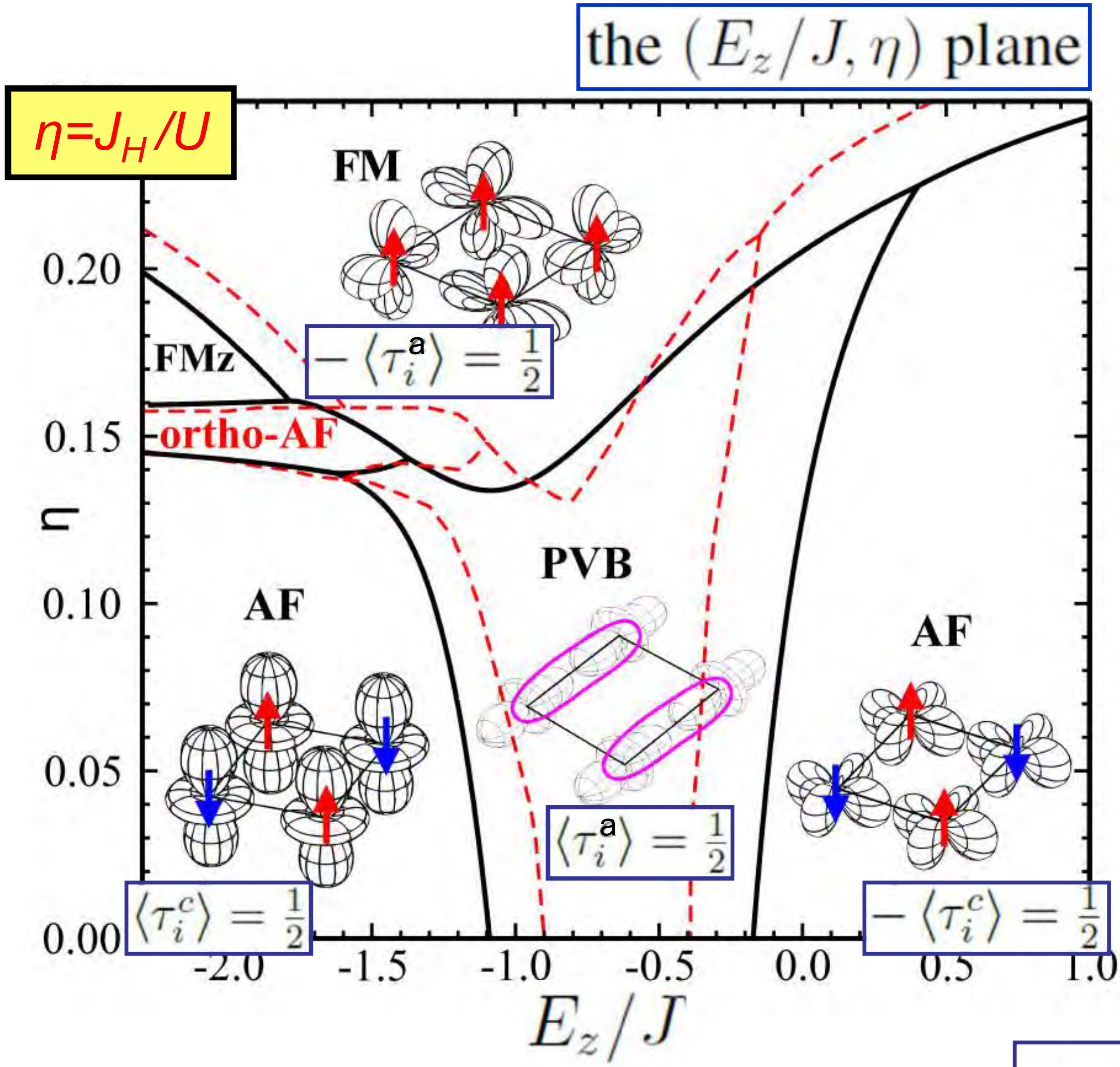
Averages of the orbital projection operators

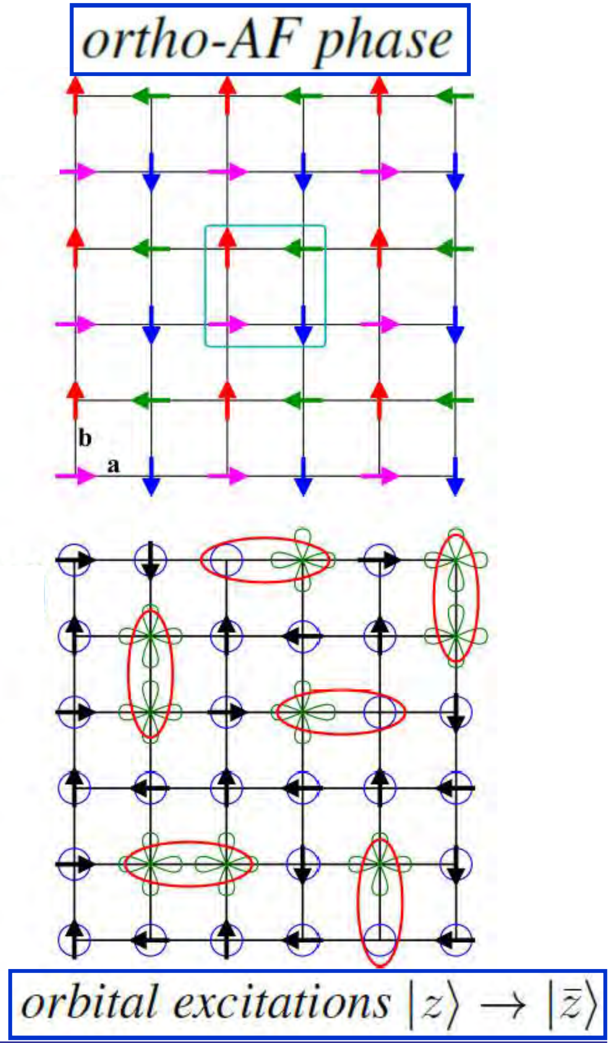
operator	average	ab	c
$\mathcal{Q}^{(\gamma)}_{\langle ij\rangle}$	$2\left\langle\left(\frac{1}{2}-\tau_i^{(\gamma)}\right)\left(\frac{1}{2}-\tau_j^{(\gamma)}\right)\right\rangle$	$\frac{1}{2}\left(\frac{1}{2}-\cos\theta\right)^2$	$\frac{1}{2}\left(1+\cos\theta\right)^2$
$\mathcal{P}^{(\gamma)}_{\langle ij\rangle}$	$\left\langle\frac{1}{4}-\tau_i^{(\gamma)}\tau_j^{(\gamma)}\right\rangle$	$\frac{1}{4}\left(\frac{3}{4}+\sin^2\theta\right)$	$\frac{1}{4}\sin^2\theta$
$\mathcal{R}^{(\gamma)}_{\langle ij\rangle}$	$2\left\langle\left(\frac{1}{2}+\tau_i^{(\gamma)}\right)\left(\frac{1}{2}+\tau_j^{(\gamma)}\right)\right\rangle$	$\frac{1}{2}\left(\frac{1}{2}+\cos\theta\right)^2$	$\frac{1}{2}\left(1-\cos\theta\right)^2$

anisotropic exchange constants

$$\begin{aligned} J_c &= \frac{1}{8}J\Big\{-r_1\sin^2\theta + (r_2+r_3)(1+\cos\theta) + r_4(1+\cos\theta)^2\Big\}, \\ J_{ab} &= \frac{1}{8}J\left\{-r_1\left(\frac{3}{4}+\sin^2\theta\right) + (r_2+r_3)\left(1-\frac{1}{2}\cos\theta\right) + r_4\left(\frac{1}{2}-\cos\theta\right)^2\right\} \end{aligned}$$

Phase diagram: 2D Kugel-Khomskii model





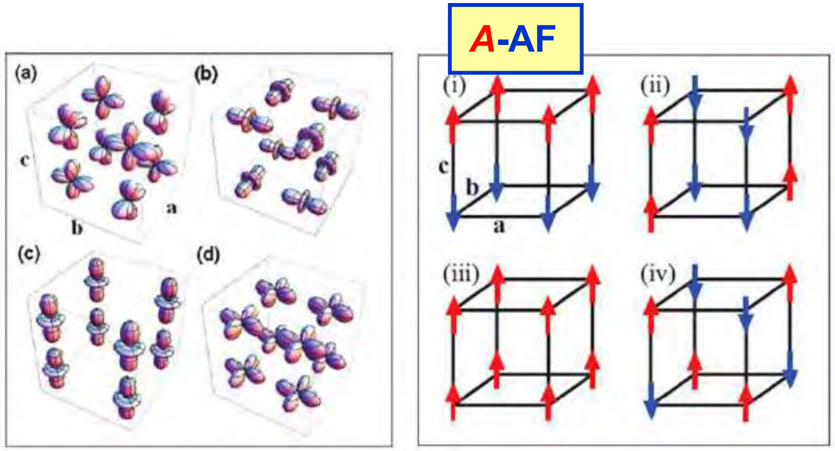
$$H_s \simeq J\left\{N\varepsilon_0 + H_s^{(1)} + H_s^{(2)} + H_s^{(3)}\right\}$$

Much weaker AF interactions between holes in $|z\rangle$ orbitals than in $|\bar{z}\rangle$

in mean field approximation a quantum critical point $Q_{2D} = (-0.5, 0)$

Spin-orbital entanglement near the QCP

Example: Kugel-Khomskii (KK) model (d^9)

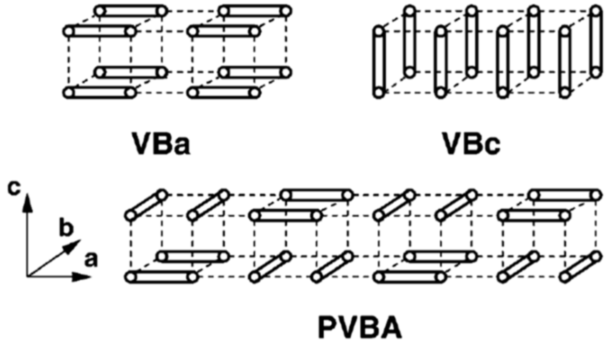


e_g orbitals T=1/2 **spins S=1/2**

Parameters: (1) E_z/J – e_g orbital splitting
(2) J_H/U – Hund´s exchange

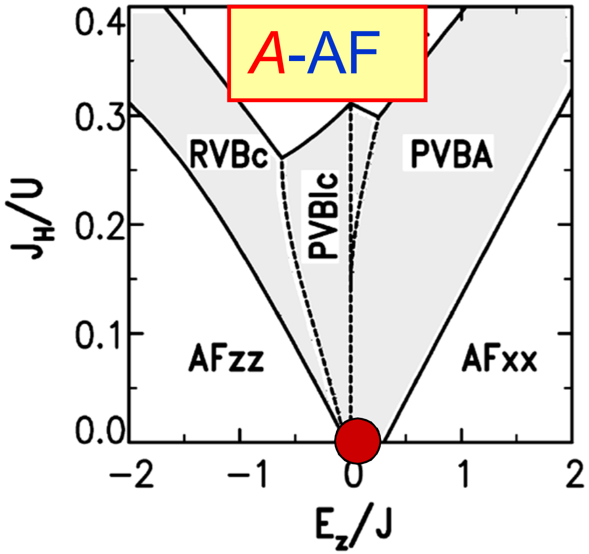
Quantum critical point: $\boxed{Q_{3D} = (0,0)}$

Entanglement near the QCP ?



Spin disordered states

***A*-AF** phase in $KCuF_3$

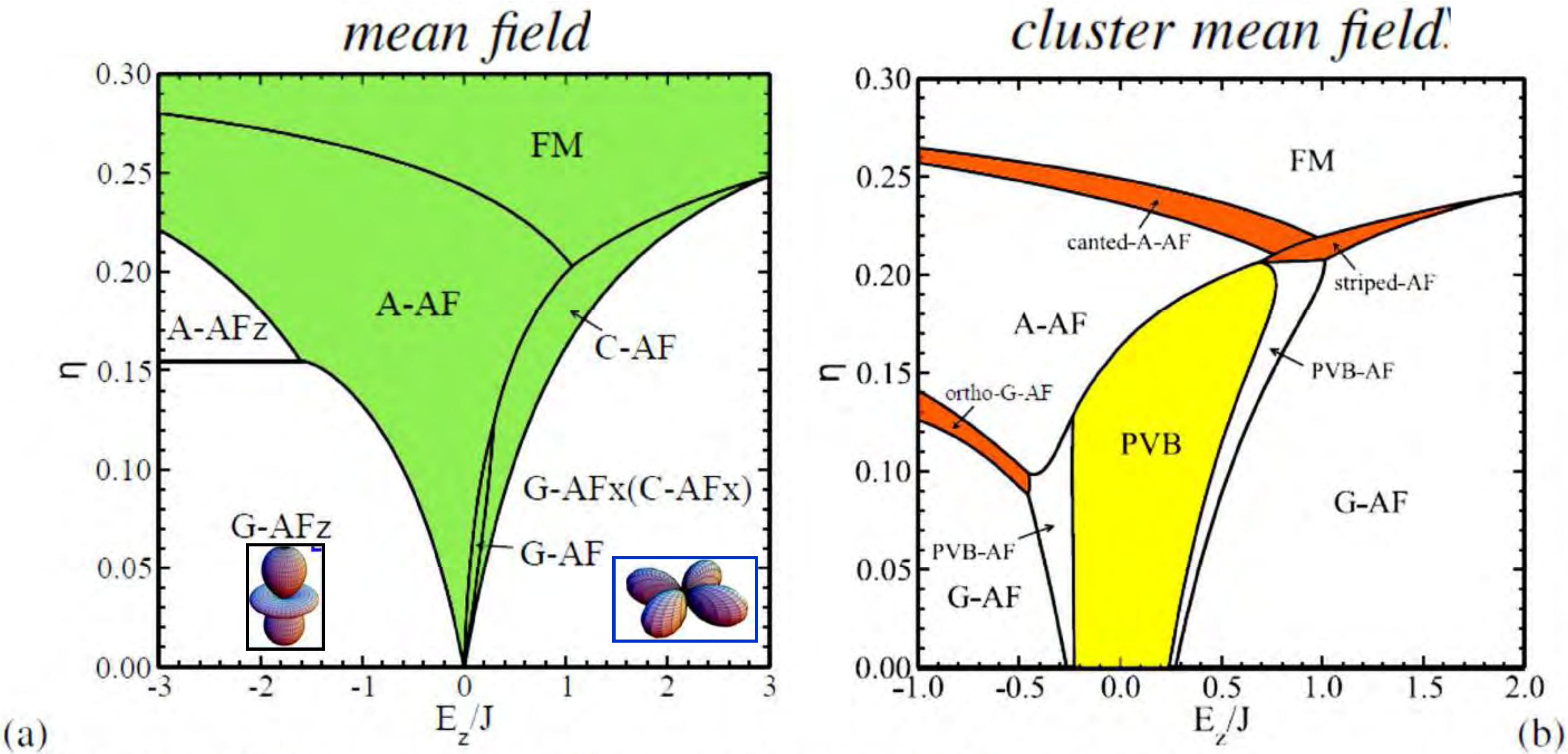


Phase diagram of the d^9 model

[L.F. Feiner, AMO, J. Zaanen, PRL **78**, 2799 (97)]

Phase diagram: 3D Kugel-Khomskii model

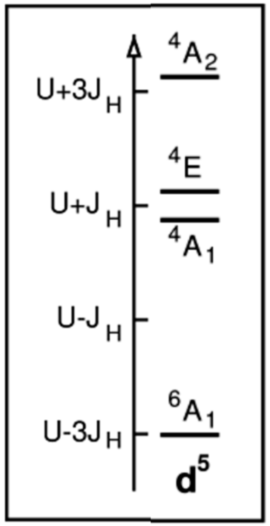
the $(E_z/J, \eta)$ plane



in mean field approximation a quantum critical point $Q_{3D} = (0, 0)$

Exotic magnetic order arises at boarder lines between phases AF due to spin-orbital entanglement

Spin-orbital model for $LaMnO_3$



Weak AF superexchange due to t_{2g} electrons

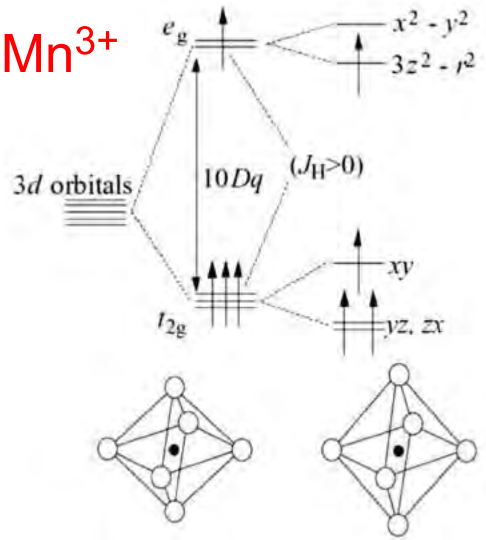
$$\mathcal{H}_t = \frac{1}{8} J\beta r_t \left(\vec{S}_i \cdot \vec{S}_j - 4\right)$$

(i) the HS ($\mathcal{S}=\frac{5}{2}$) $^6A_1$ state, and (ii) the LS ($\mathcal{S}=\frac{3}{2}$) states: $^4A_1$, ^4E ($^4E_\epsilon$, $^4E_\theta$), and $^4A_2$

$\varepsilon(^4A_1)=U+\frac{3}{4}J_H$, $\varepsilon(^4E)=U+\frac{5}{4}J_H$ (twice), and $\varepsilon(^4A_2)=U+\frac{13}{4}J_H$

$$\mathcal{H}_e = \frac{1}{16}\sum_\gamma \sum_{\langle ij\rangle\parallel\gamma} \left\{ -\frac{8}{5}\frac{t^2}{\varepsilon(^6A_1)}\left(\vec{S}_i\cdot\vec{S}_j+6\right)\mathcal{P}^{(\gamma)}_{\langle ij\rangle} + \left[\frac{t^2}{\varepsilon(^4E)}+\frac{3}{5}\frac{t^2}{\varepsilon(^4A_1)}\right]\left(\vec{S}_i\cdot\vec{S}_j-4\right)\mathcal{P}^{(\gamma)}_{\langle ij\rangle} + \left[\frac{t^2}{\varepsilon(^4E)}+\frac{t^2}{\varepsilon(^4A_2)}\right]\left(\vec{S}_i\cdot\vec{S}_j-4\right)\mathcal{Q}^{(\gamma)}_{\langle ij\rangle}\right\} + E_z\sum_i \tau_i^c. \quad (49)$$

Mn^{3+}



Bond projection operators:

$$\mathcal{P}^{(\gamma)}_{\langle ij\rangle} \equiv \left(\frac{1}{2}+\tau_i^{(\gamma)}\right)\left(\frac{1}{2}-\tau_j^{(\gamma)}\right)+\left(\frac{1}{2}-\tau_i^{(\gamma)}\right)\left(\frac{1}{2}+\tau_j^{(\gamma)}\right),$$

$$\mathcal{Q}^{(\gamma)}_{\langle ij\rangle} \equiv 2\left(\frac{1}{2}+\tau_i^{(\gamma)}\right)\left(\frac{1}{2}+\tau_j^{(\gamma)}\right).$$

crystal field $\propto E_z$

The spin-orbital part is similar to $KCuF_3$ (S=2)

[L.F. Feiner and AMO, PRB **59**, 3295 (99)]

Spin-orbital physics: optical spectral weights

Structure of the spin-orbital model:

$$\mathcal{H} = J \sum_n \sum_{\langle ij \rangle \| \gamma} H_n^{(\gamma)}(ij)$$

Terms originate from charge excitations to multiplet states *n*

$$\varepsilon_n = E_n(d^{m+1}) + E_0(d^{m-1}) - 2E_0(d^m)$$

Spectral weight for an excitation at energy ω_n

$$\frac{a_0 \hbar^2}{e^2} \int_0^\infty \sigma_n^{(\gamma)}(\omega) d\omega = \frac{\pi}{2} K_n^{(\gamma)}$$

These weights are found from the superexchange terms:

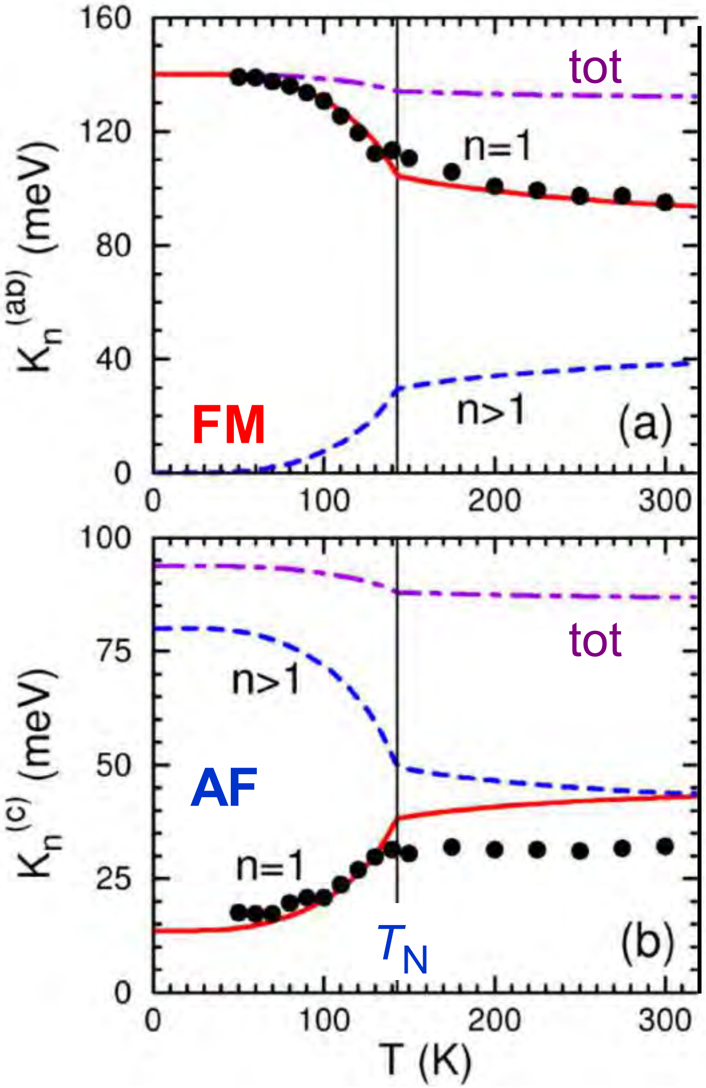
$$K_n^{(\gamma)} = -2J \left\langle H_n^{(\gamma)}(ij) \right\rangle$$

for excitation at ω_n

$$K^{(\gamma)} = -2J \sum_n \left\langle H_n^{(\gamma)}(ij) \right\rangle$$

total weight

Optical spectral weights for $LaMnO_3$



spin-orbital model

$$\mathcal{H} = J \sum_n \sum_{\langle ij \rangle \| \gamma} H_n^{(\gamma)}(ij)$$

spectral weight for excitation at energy ω_n

$$K_n^{(\gamma)} = -2J \left\langle H_n^{(\gamma)}(ij) \right\rangle$$

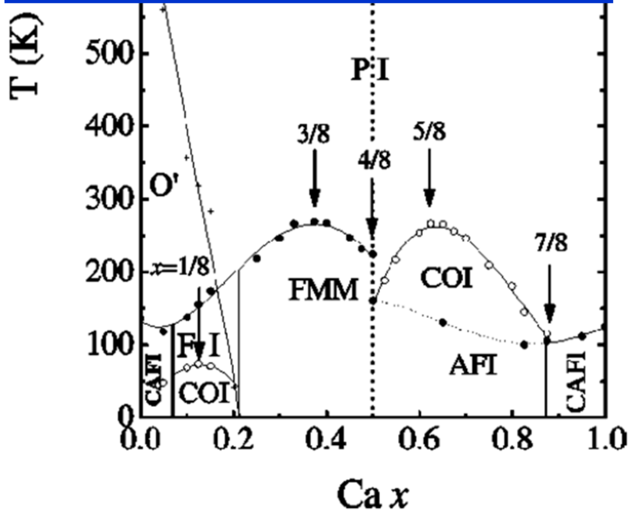
Theory reproduces the spectral weights for low energy ω_1 high-spin excitations

Spin and orbital correlations are here disentangled

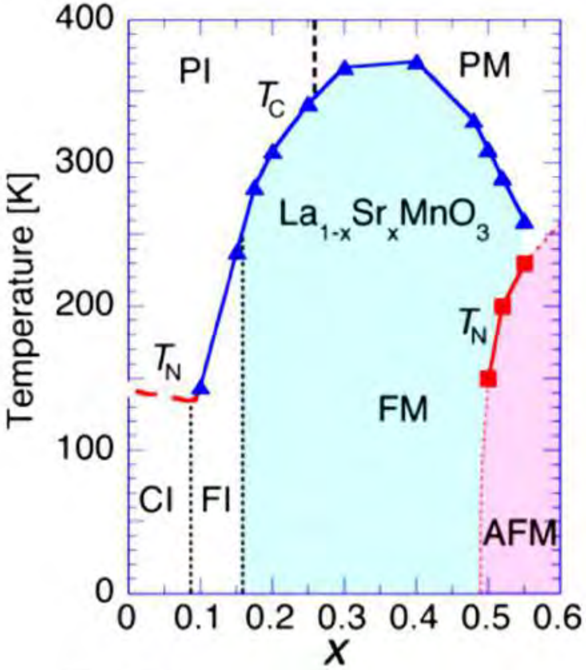
Note: $S = 2$

Colossal magnetoresistance in manganites

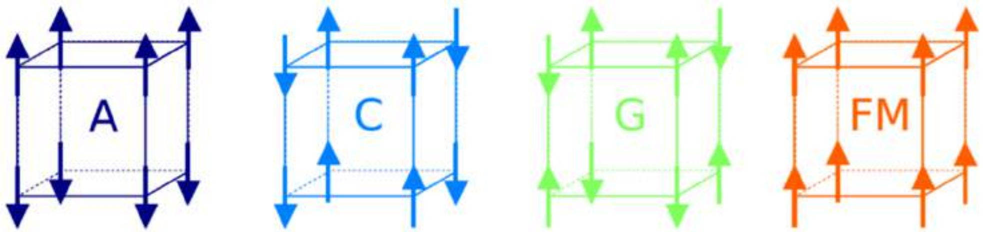
Phase diagrams $La_{1-x}A_xMnO_3$



[Sang-Wook Cheong (98)]



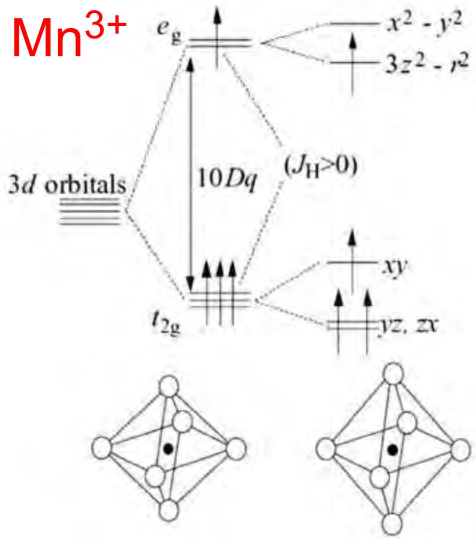
[Y. Tokura, RPP **67**, 797 (06)]



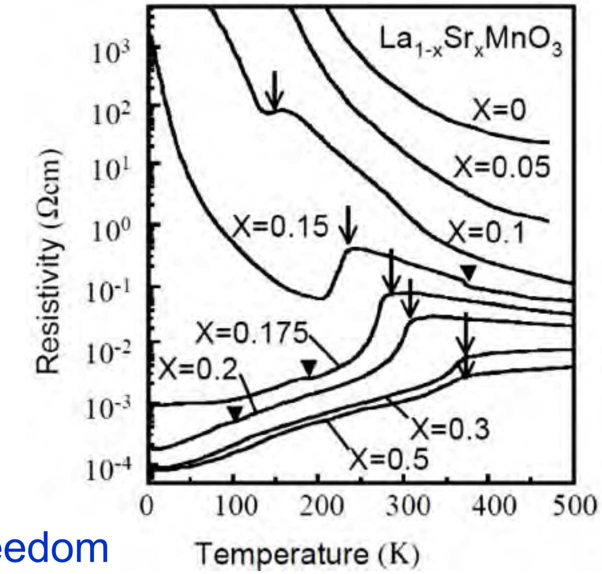
Possible magnetic phases in doped manganites

[M. Stier and W. Nolting, PRB **78**, 144425 (08)]

Mn^{3+}



Orbital and charge degree of freedom



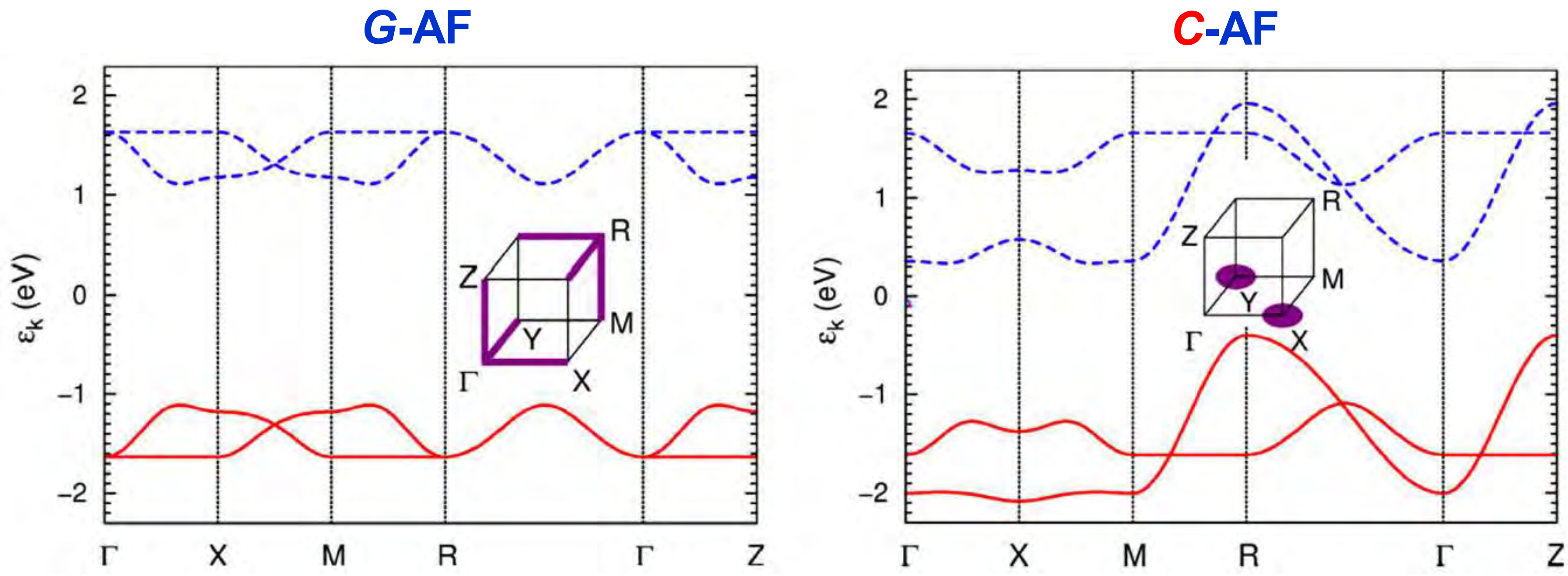
Theory has to include strong correlations of e_g electrons

Avoiding electron correlations: electron doped $Sr_{1-x}La_xMnO_3$

Kondo-like model for (few) noninteracting e_g electrons :

$$\mathcal{H} = -\sum_{ij,\alpha\beta,\sigma} t^{ij}_{\alpha\beta} a^{\dagger}_{i\alpha\sigma} a_{j\beta\sigma} - 2J_H \sum_i \vec{S}_i \cdot \vec{s}_i + J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - gu \sum_i (n_{iz} - n_{i\bar{z}}) + \frac{1}{2} N K u^2$$

$$u \equiv 2(c-a)/(c+a) \quad \textit{Parameters: } t = 0.4\,eV,\ J_H = 0.74\,eV,\ g = 3\,eV.$$



$$\Gamma = (0,0,0),\ X = (\pi,0,0),\ M = (\pi,\pi,0),\ R = (\pi,\pi,\pi),\ Z = (0,0,\pi)$$

C-AF phase stabilized by double exchange for $x > 0.02$

Degenerate Hubbard model & charge excitations ($t \ll U$)

Two parameters: ***U*** – intraorbital Coulomb interaction, ***J_H*** – Hund's exchange

$$H_{\mathrm{int}} = U \sum_{i\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + (U - \tfrac{5}{2} J_H) \sum_{i,\alpha<\beta} n_{i\alpha} n_{i\beta} - 2J_H \sum_{i,\alpha<\beta} \vec{S}_{i\alpha} \cdot \vec{S}_{i\beta}$$
$$+ J_H \sum_{i,\alpha<\beta} (d^{+}_{i\alpha\uparrow} d^{+}_{i\alpha\downarrow} d_{i\beta\downarrow} d_{i\beta\uparrow} + d^{+}_{i\beta\uparrow} d^{+}_{i\beta\downarrow} d_{i\alpha\downarrow} d_{i\alpha\uparrow})$$

In a Mott insulator (t<<U) superexchange follows from charge excitations

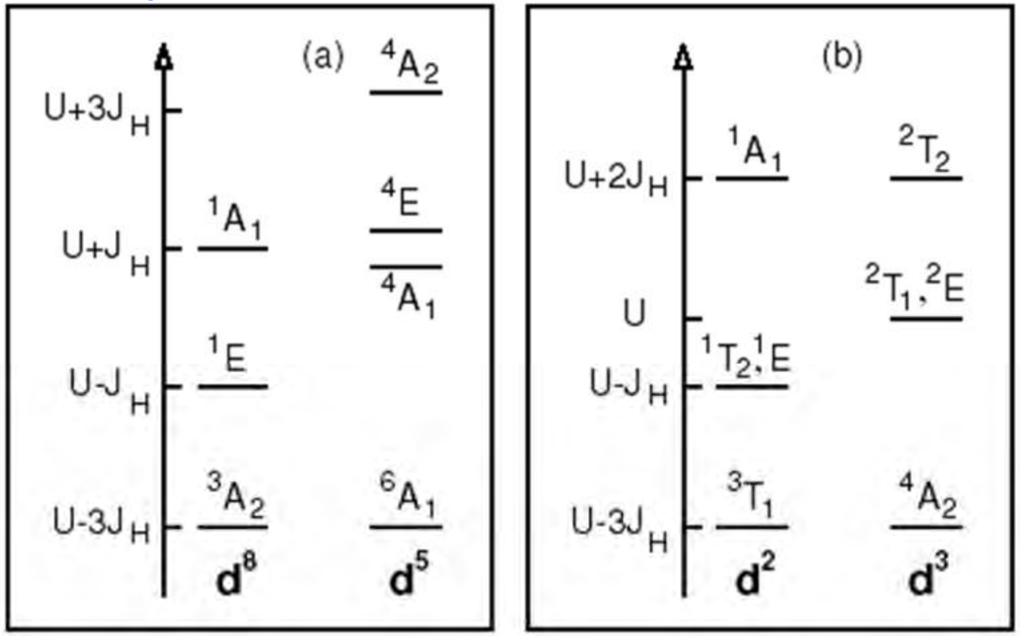
$$d_i^m d_j^m \rightleftharpoons d_i^{m+1} d_j^{m-1}$$

single parameter:

$\eta = J_H / U$

Correl17

e_g systems $\qquad$ t_{2g} systems



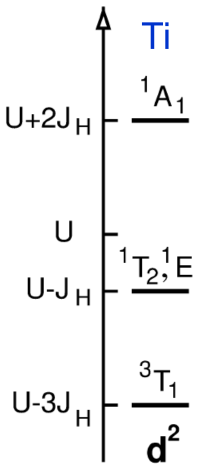
LS

HS

FIG. 1: Energies of $d_i^m d_j^m \to d_i^{m+1} d_j^{m-1}$ charge excitations

$$d_i^1 d_j^1 \rightleftharpoons d_i^2 d_j^0.$$

Spin-orbital superexchange in *R*TiO_3



$$|a\rangle \equiv |yz\rangle, \qquad |b\rangle \equiv |zx\rangle, \qquad |c\rangle \equiv |xy\rangle$$

constraint: $n_{ia} + n_{ib} + n_{ic} = 1$

Multiplet structure =>

$$r_1 = \frac{1}{1-3\eta}, \qquad r_2 = \frac{1}{1-\eta}, \qquad r_3 = \frac{1}{1+2\eta}$$

superexchange

$$\mathcal{H} = J \sum_n \sum_{\langle ij\rangle \parallel \gamma} H_n^{(\gamma)}(ij)$$

$$\begin{aligned}
H_1^{(\gamma)} &= \frac{1}{2} J r_1 \left(\vec{S}_i \cdot \vec{S}_j + \frac{3}{4}\right)\left(A_{ij}^{(\gamma)} - \frac{1}{2} n_{ij}^{(\gamma)}\right), \\
H_2^{(\gamma)} &= \frac{1}{2} J r_2 \left(\vec{S}_i \cdot \vec{S}_j - \frac{1}{4}\right)\left(A_{ij}^{(\gamma)} - \frac{2}{3} B_{ij}^{(\gamma)} + \frac{1}{2} n_{ij}^{(\gamma)}\right), \\
H_3^{(\gamma)} &= \frac{1}{3} J r_3 \left(\vec{S}_i \cdot \vec{S}_j - \frac{1}{4}\right) B_{ij}^{(\gamma)},
\end{aligned}$$

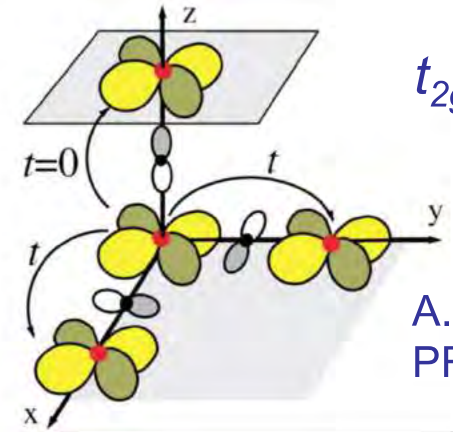
orbital fluctuations

$$A_{ij}^{(\gamma)} = 2\left(\vec{\tau}_i \cdot \vec{\tau}_j + \frac{1}{4} n_i n_j\right)^{(\gamma)}, \quad B_{ij}^{(\gamma)} = 2\left(\vec{\tau}_i \otimes \vec{\tau}_j + \frac{1}{4} n_i n_j\right)^{(\gamma)}, \quad n_{ij}^{(\gamma)} = n_i^{(\gamma)} + n_j^{(\gamma)}$$

$$\vec{\tau}_i \otimes \vec{\tau}_j = \tau_i^x \tau_j^x - \tau_i^y \tau_j^y + \tau_i^z \tau_j^z$$

Nonconservation of orbital flavor
=> spin-orbital liquid ?

Spin-Orbital Model for RVO$_3$ (R=La,Y, …)



t_{2g} hopping

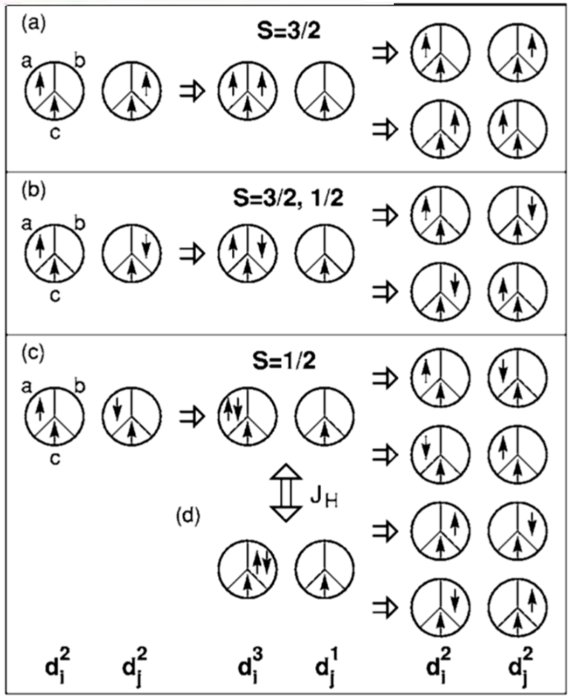
A.B. Harris *et al*., PRL **91**, 087206 (03)

$t_{2g}{}^2$ configurations of V^{3+} ions with S =1 spins
each orbital is inactive along one axis

$$|a\rangle \equiv |yz\rangle, \quad |b\rangle \equiv |xz\rangle, \quad |c\rangle \equiv |xy\rangle,$$

For $T<T_s$ *xy* orbitals are occupied:

$$n_{ic} \simeq 1, \quad n_{ia} + n_{ib} \simeq 1$$



orbital fluctuations

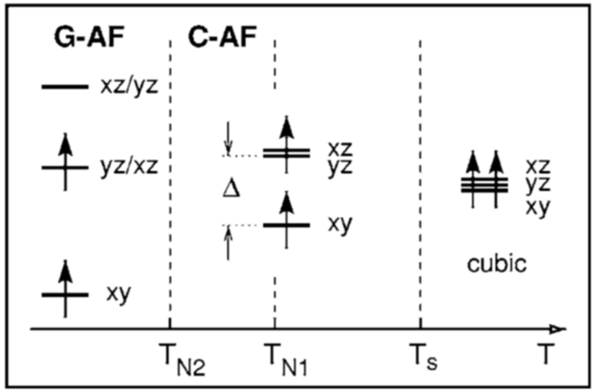
spin fluctuations

spin fluct.

orbital fluct.

spin-orbital fluctuations

charge excitations $d_i^2 d_j^2 \rightarrow d_i^3 d_j^1 \rightarrow d_i^2 d_j^2$



Energies of t_{2g} orbitals in YVO$_3$

Superexchange for t<<U (at J_H=0):

$$\boxed{\mathcal{H}_0 = \frac{1}{2} J \sum_{\langle ij \rangle \parallel \gamma} (\vec{S}_i \cdot \vec{S}_j + 1) \left(\vec{\tau}_i \cdot \vec{\tau}_j + \frac{1}{4} n_i n_j \right)^{(\gamma)}}$$

[G. Khaliullin, P. Horsch, AMO, PRL **86**, 3879 (01)]

Spin-orbital superexchange in $R\mathrm{VO}_3$

In t_{2g} **systems** (d^1,d^2) two states are active, e.g. *yz* i *zx* for the axis *c* – they are described by **quantum** operators:

$$\vec{\tau}_i = \{ \tau_i^x \ , \ \tau_i^y \ , \ \tau_i^z \ \}$$

Scalar product $\vec{\tau}_i \cdot \vec{\tau}_j$ but for η>0 also:

$$\boxed{\vec{\tau}_i \otimes \vec{\tau}_j = \tau_i^x \tau_j^x - \tau_i^y \tau_j^y + \tau_i^z \tau_j^z}$$

Local constraints:

$$n_{ic} \simeq 1, \quad n_{ia} + n_{ib} \simeq 1$$

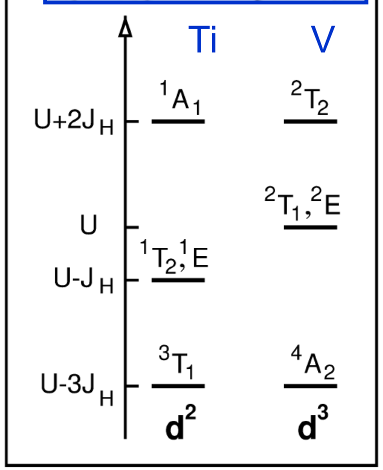
c orbitals occupied

{***a***,***b***} – orbital degree of freedom

Orbital interactions have cubic symmetry

Orbital SU(2) symmetry is broken !

$$\boxed{d_i^2 d_j^2 \rightleftharpoons d_i^3 d_j^1}$$



Spin-orbit superexchange in $R\mathrm{VO}_3$ d^2 (S = 1):

$$\boxed{\mathcal{H}_0 = \frac{1}{2} J \sum_{\langle ij \rangle \parallel \gamma} (\vec{S}_i \cdot \vec{S}_j + 1) \left(\vec{\tau}_i \cdot \vec{\tau}_j + \frac{1}{4} n_i n_j \right)^{(\gamma)}}$$

Multiplet structure =>

$$\boxed{r_1 = \frac{1}{1-3\eta}, \qquad r_3 = \frac{1}{1+2\eta}}$$

Entanglement expected !

Strong orbital fluctuations in *C*-**AF** phase ($LaVO_3$)

{*a*,*b*}={*yz*,*zx*} **fluctuations** on bonds <*ij*> || *c* axis $\quad n_{ic} \simeq 1, \quad n_{ia} + n_{ib} \simeq 1$

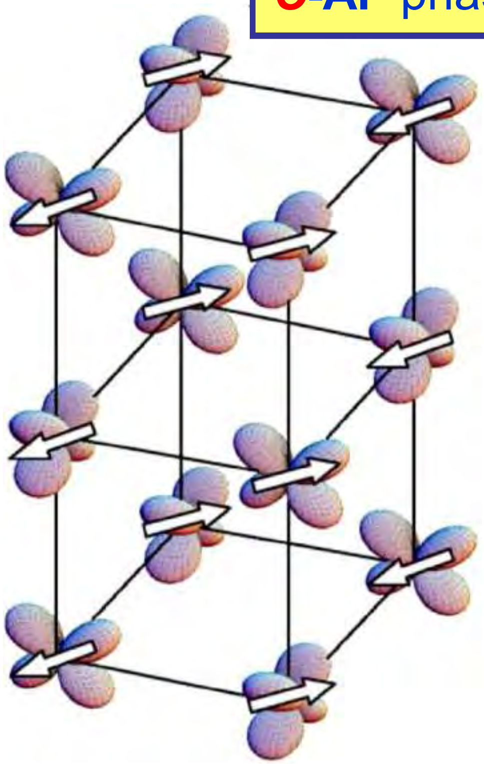
⇒ finite value of **FM** $-J_c>0$ for $\eta=0$ (**no Hund's mechanism!**)

⇒ similar values of exchange **AF** J_{ab} and **FM** $-J_c$ for $\eta=0.13$

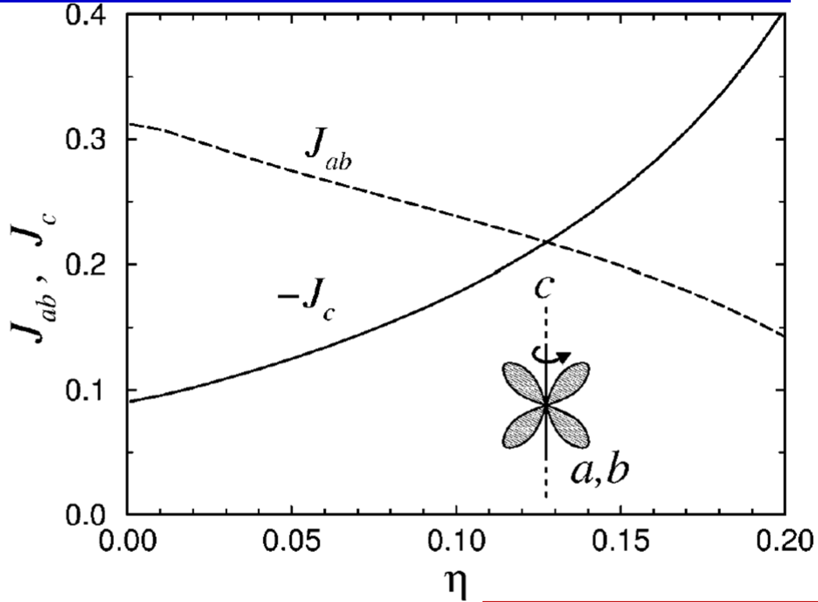
$$J_c = \frac{1}{2}J\left\{\eta r_1 - (r_1 - \eta r_1 - \eta r_3)\left(\tfrac{1}{4} + \langle\vec{\tau}_i\cdot\vec{\tau}_j\rangle\right) - 2\eta r_3\langle\tau_i^y\tau_j^y\rangle\right\}$$

$$J_{ab} = \frac{1}{4}J\left\{1 - \eta r_1 - \eta r_3 + (r_1 - \eta r_1 - \eta r_3)\left(\tfrac{1}{4} + \langle\tau_i^z\tau_j^z\rangle\right)\right\},$$

C-**AF** phase



Exchange constants in *C*-**AF** phase for increasing Hund's exchange η



[G. Khaliullin P. Horsch, AMO, PRL **86**, 3879 (01)]

$\eta=J_H/U=0.13$

Spin-orbital superexchange in $R\mathrm{VO}_3$: spectral weights

cubic symmetry broken by ***C*-AF**

$$K_n^{(\gamma)} = -2J\left\langle H_n^{(\gamma)}(ij)\right\rangle$$

orbital fluctuations:

$$H_1^{(c)}(ij) = -\frac{1}{3}Jr_1\left(\vec{S}_i\cdot\vec{S}_j+2\right)\left(\tfrac{1}{4}-\vec{\tau}_i\cdot\vec{\tau}_j\right)$$

$$H_2^{(c)}(ij) = -\frac{1}{12}J\left(1-\vec{S}_i\cdot\vec{S}_j\right)\left(\tfrac{7}{4}-\tau_i^z\tau_j^z-\tau_i^x\tau_j^x+5\tau_i^y\tau_j^y\right)$$

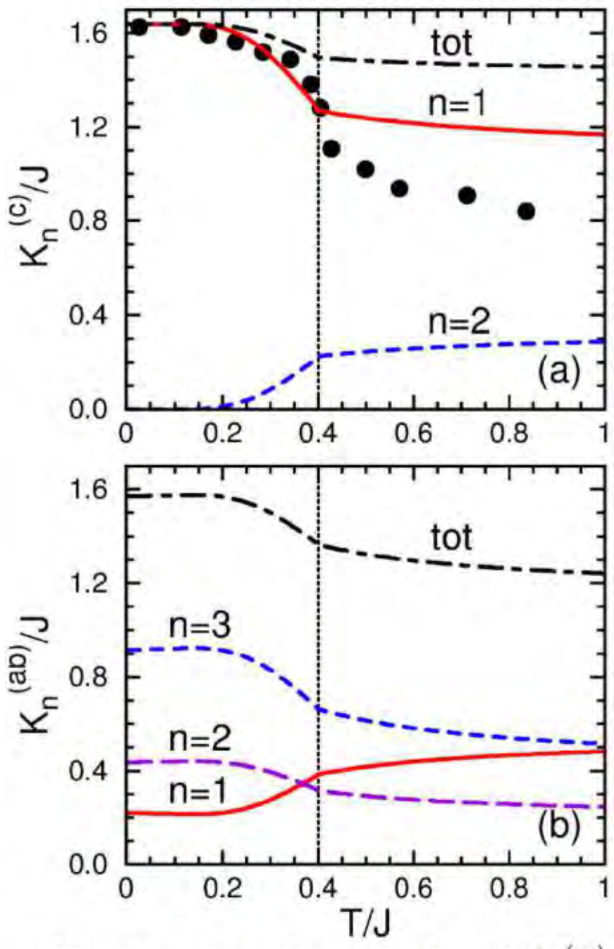
$$H_3^{(c)}(ij) = -\frac{1}{4}Jr\left(1-\vec{S}_i\cdot\vec{S}_j\right)\left(\tfrac{1}{4}+\tau_i^z\tau_j^z+\tau_i^x\tau_j^x-\tau_i^y\tau_j^y\right)$$

more classical:

$$H_1^{(ab)}(ij) = -\frac{1}{6}Jr_1\left(\vec{S}_i\cdot\vec{S}_j+2\right)\left(\tfrac{1}{4}-\tau_i^z\tau_j^z\right)$$

$$H_2^{(ab)}(ij) = -\frac{1}{8}J\left(1-\vec{S}_i\cdot\vec{S}_j\right)\left(\tfrac{19}{12}\mp\tfrac{1}{2}\tau_i^z\mp\tfrac{1}{2}\tau_j^z-\tfrac{1}{3}\tau_i^z\tau_j^z\right)$$

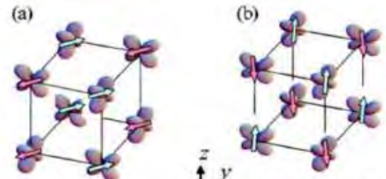
$$H_3^{(ab)}(ij) = -\frac{1}{8}Jr\left(1-\vec{S}_i\cdot\vec{S}_j\right)\left(\tfrac{5}{4}\mp\tfrac{1}{2}\tau_i^z\mp\tfrac{1}{2}\tau_j^z+\tau_i^z\tau_j^z\right)$$



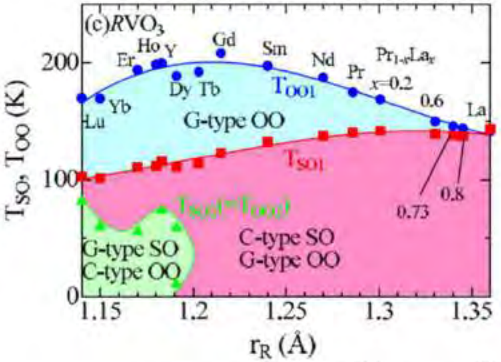
Kinetic energies per bond $K_n^{(\gamma)}$

Spin-orbital entanglement along the *c* axis

Spin and orbital phase transitions in RVO_3



C-AF phase　　**G-AF** phase



$$u = (b - a)/a$$

T_{N1} modified due to entanglement !

Orbital-lattice coupling $\propto \langle \tau^x \rangle$ is crucial

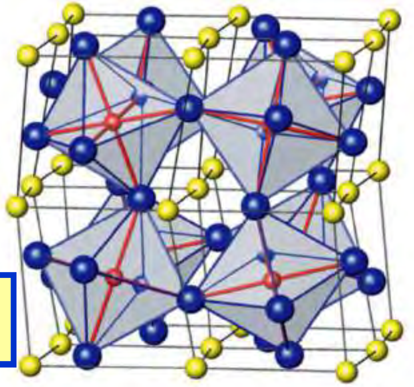
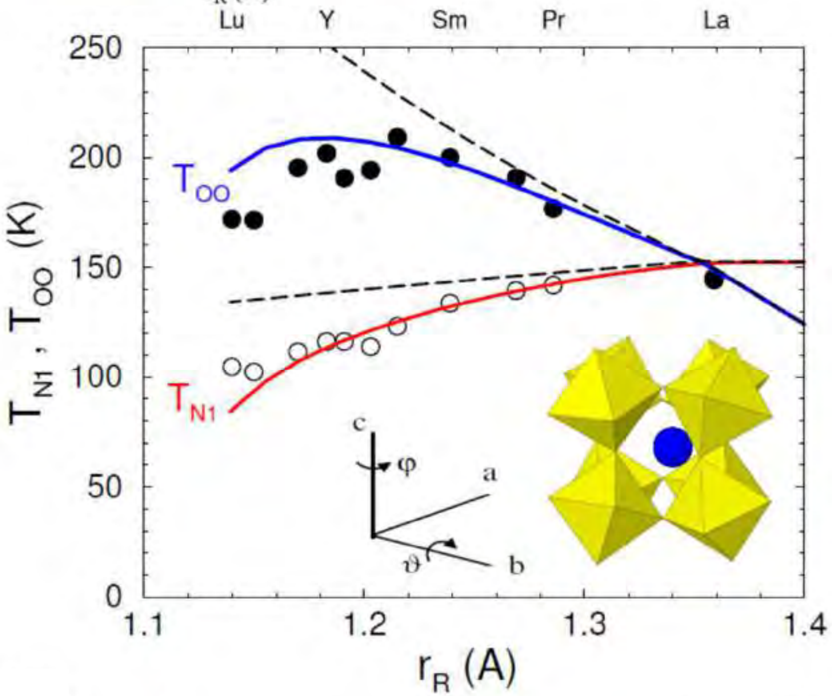
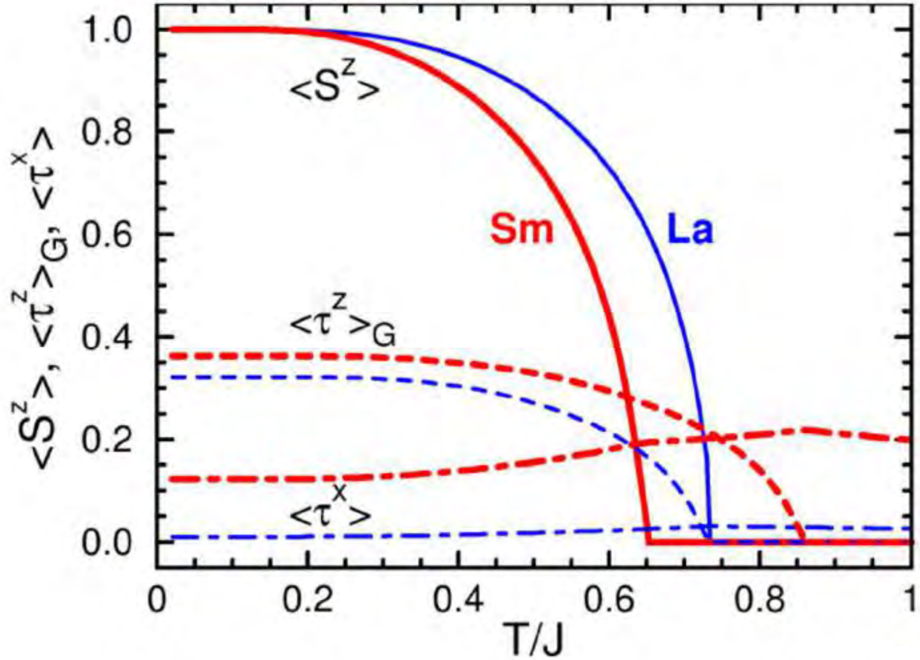


Figure 2. Crystal structure of a $GdFeO_3$-distorted ABO_3



Phase transitions in the vanadium perovskites RVO_3



spin $\langle S_i^z \rangle$ *(solid) and G-type orbital* $\langle \tau_i^z \rangle_G$ *(dashed) and the transverse orbital polarization* $\langle \tau_i^x \rangle$ *(dashed-dotted lines)*

Goodenough-Kanamori rules

AO order supports **FM** spin order

FO order supports **AF** spin order

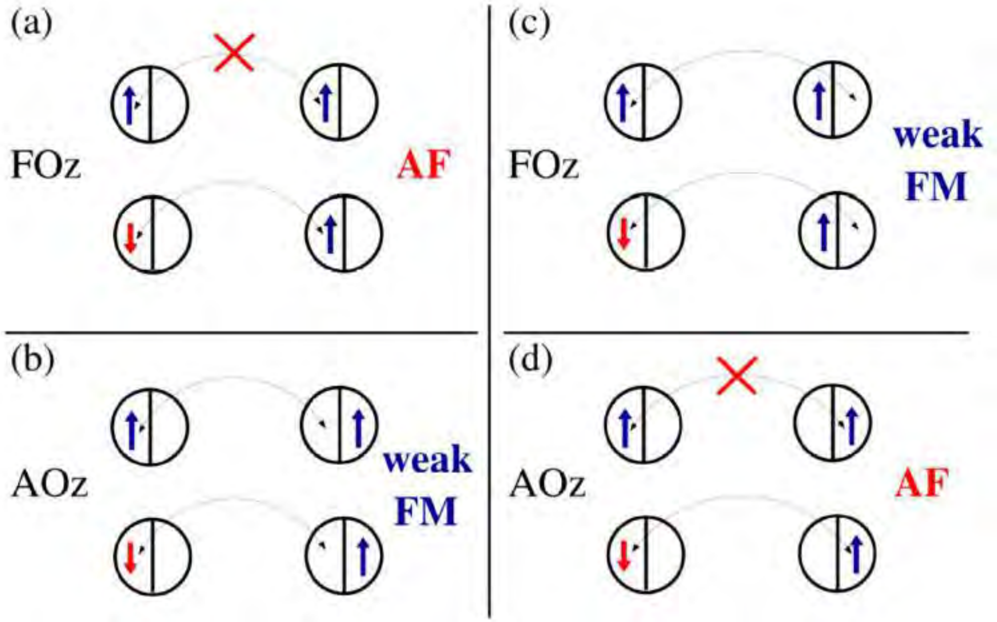
e_g orbitals

why ?

$\{|i\zeta_\gamma\rangle, |j\xi_\gamma\rangle\}$ -- **FM** order wins $\propto \eta$

$\{|i\zeta_\gamma\rangle, |j\zeta_\gamma\rangle\}$ -- only **AF** terms

intraorbital hopping *t* interorbital hopping *t'*



complementarity
for orbital conserving hopping *t*

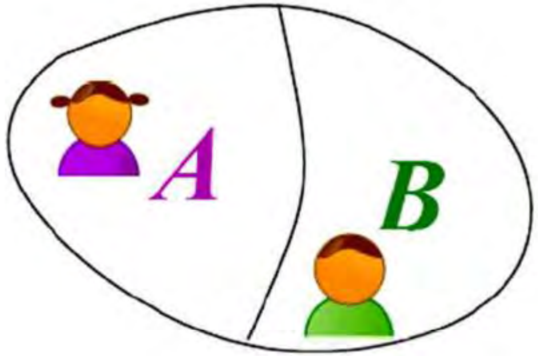
frustration
by interorbital hopping *t'*
$\Rightarrow$ **FM/FO** or **AF/AO**
are possible

Exotic spin orders possible in certain situations

Entanglement entropy (Bipartite)

- Two subsystems: ***A*** and ***B***
- Wave function: $\psi_{AB} = \sum_{mn} C_{mn} \psi_A^{(m)} \psi_B^{(n)}$
- Product state: $C_{mn} = C_m C_n$
- Entangled state: $C_{mn} \neq C_m C_n$
- Taking trace over ***B*** leads to

$\mathcal{H} = \mathcal{H}_A \otimes \mathcal{H}_B$



$$\rho_A^{(0)} = \mathrm{Tr}_B |\Psi_0\rangle\langle\Psi_0|$$

Entanglement is measured by von Neumann entropy in the ground state

$$S_{\mathrm{vN}}^0 \equiv -\mathrm{Tr}_A\{\rho_A^{(0)} \log_2 \rho_A^{(0)}\}$$

Here ***A*** **and** ***B*** **are spin and orbital** degrees of freedom of the system
i.e., the boundary involves the entire system

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J. Phys.: Condens. Matter **24** (2012) 313201 (28pp) doi:10.1088/0953-8984/24/31/313201

TOPICAL REVIEW

Fingerprints of spin–orbital entanglement in transition metal oxides

Andrzej M Oleś

Contents

Example: triangular lattice

$d_i^1 d_j^1 \rightleftharpoons d_i^2 d_j^0$ charge excitations in $NaTiO_2$ are possible due to:

(i) the effective hopping $t = t_{pd}^2/\Delta$

(ii) direct hopping t'

$\Rightarrow$ spin-orbital model

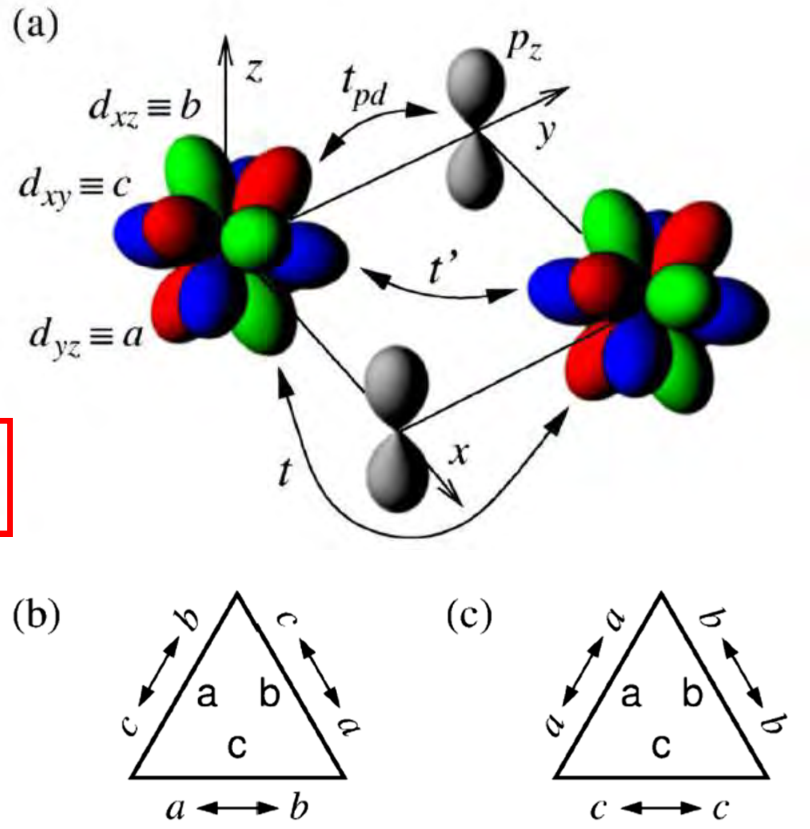
$$\boxed{\mathcal{H} = J\left\{(1-\alpha)\,\mathcal{H}_s + \sqrt{(1-\alpha)\alpha}\,\mathcal{H}_m + \alpha\,\mathcal{H}_d\right\}}$$

$$\boxed{\alpha = \frac{t'^2}{t^2 + t'^2}}$$

α interpolates between

the superexchange $\mathcal{H}_s$ $(\alpha = 0)$

kinetic exchange $\mathcal{H}_d$ $(\alpha = 1)$



t orbitals interchanged

t' orbital conserving

[B. Normand and AMO, PRB **78**, 094427 (08)]

Example: entangled states in a free hexagon

$$\mathcal{H} = J\left\{(1-\alpha)\,\mathcal{H}_s + \sqrt{(1-\alpha)\alpha}\,\mathcal{H}_m + \alpha\,\mathcal{H}_d\right\}$$

correlation functions for a bond $\langle ij \rangle$

$$S_{ij} \equiv \frac{1}{d}\sum_n \left\langle n|\vec{S}_i\cdot\vec{S}_j|n\right\rangle ,$$

$$T_{ij} \equiv \frac{1}{d}\sum_n \left\langle n|(\vec{T}_i\cdot\vec{T}_j)^{(\gamma)}|n\right\rangle ,$$

$$C_{ij} \equiv \frac{1}{d}\sum_n \left\langle n|(\vec{S}_i\cdot\vec{S}_j - S_{ij})(\vec{T}_i\cdot\vec{T}_j - T_{ij})^{(\gamma)}|n\right\rangle$$

C_{ij} is a local measure of entanglement

In the superexchange model ($\alpha = 0$)

$n_{1c} = 1$ as the c orbital is active along both bonds

RVB hexagon $S_{ij} = -0.4671$

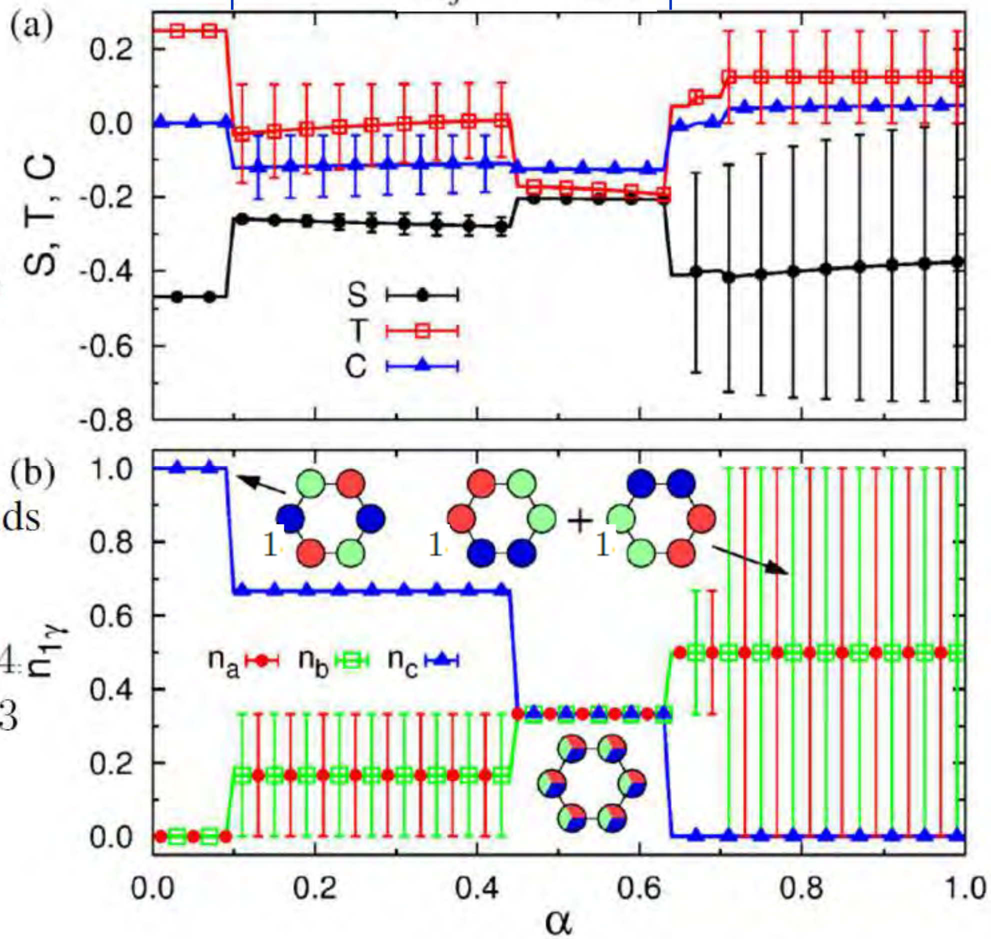
entanglement $C_{ij} \simeq -0.12$ for $0.10 < \alpha < 0.44$

$C_{ij} \simeq -0.13 \qquad 0.44 < \alpha < 0.63$

Here spins and orbitals cannot be decoupled

Weak entanglement is found for $\alpha > 0.63$

entangled $C_{ij} < -0.10$



Ground state for a free hexagon as a function of α

[J. Chaloupka and AMO, PRB **83**, 094406 (11)]

Evolution of orbital densities in a free hexagon

$$\mathcal{H} = J\left\{(1-\alpha)\,\mathcal{H}_s + \sqrt{(1-\alpha)\alpha}\,\mathcal{H}_m + \alpha\,\mathcal{H}_d\right\}$$

In the superexchange model ($\alpha = 0$)

RVB hexagon

precisely one orbital at each site

$n_{1c} = 1$ as the c orbital is active along both bonds

$0.10 < \alpha < 0.44$

$n_{1c} = 2/3 \quad n_{1a} = n_{1b} = 1/6$

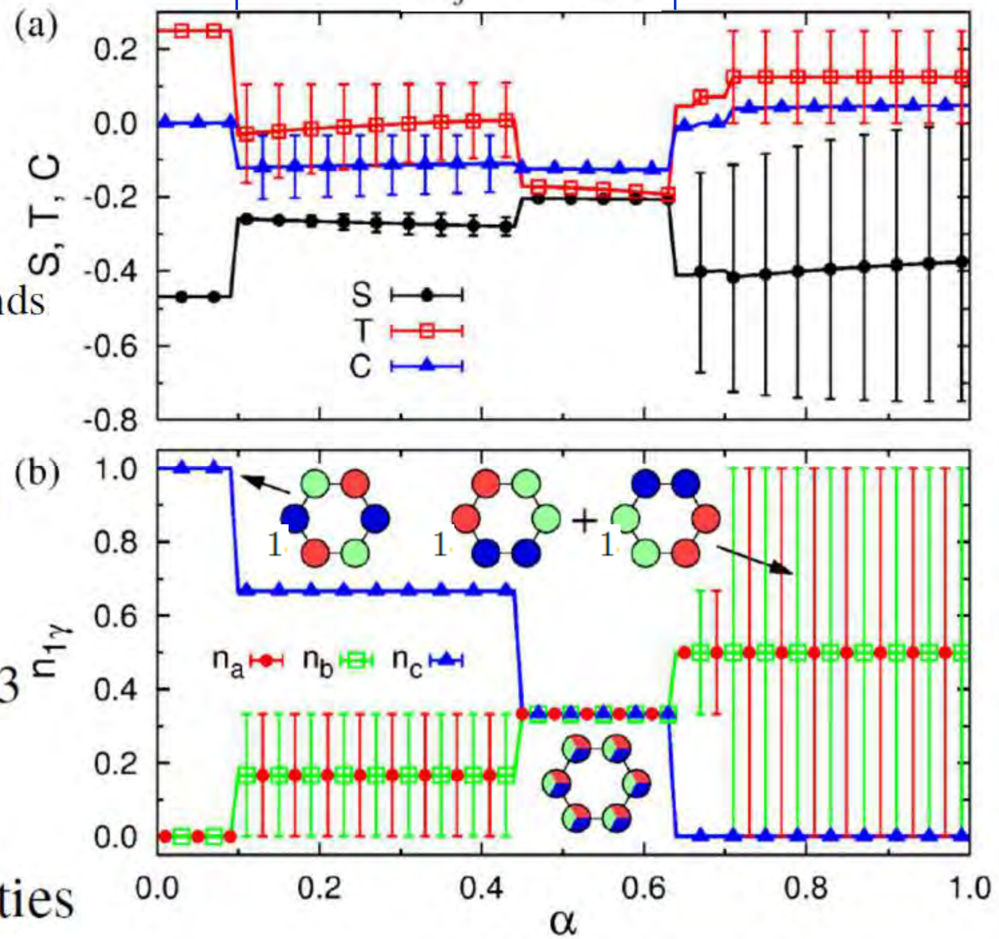
$0.44 < \alpha < 0.63$

orbitals contribute equally and $n_{1\gamma} = 1/3$

$0.63 < \alpha$

two distributions of the orbital densities

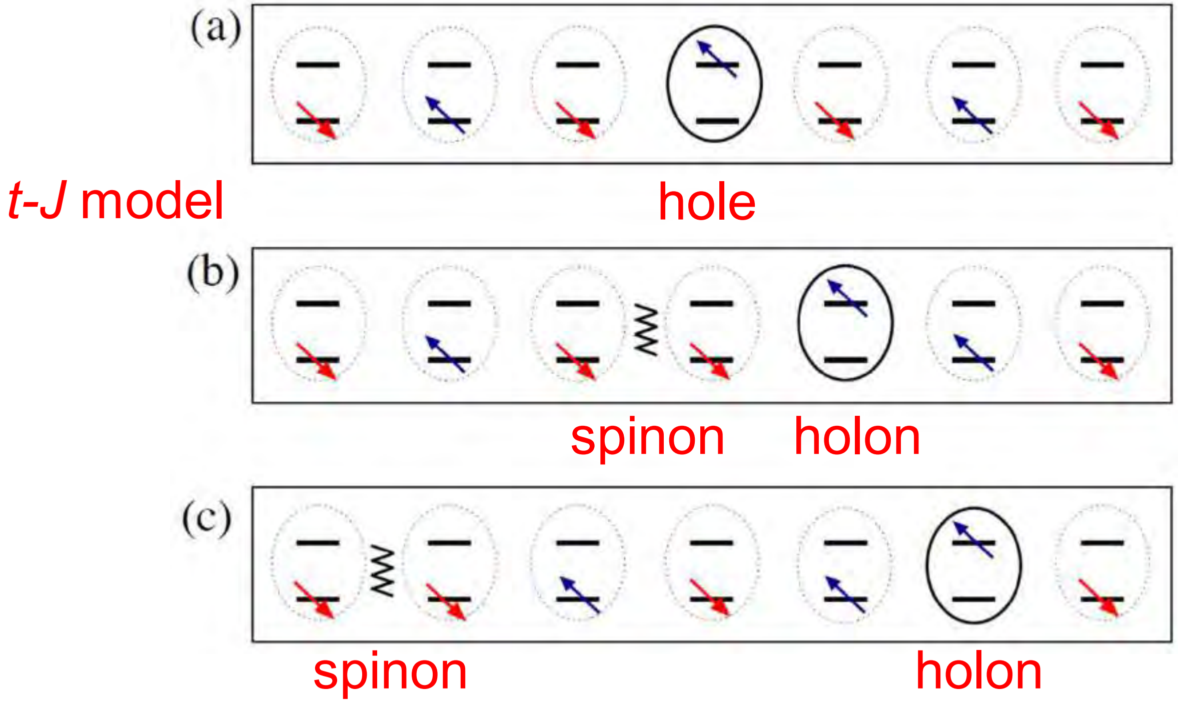
entangled $C_{ij} < -0.10$



Ground state for a free hexagon as a function of α

[J. Chaloupka and AMO, PRB **83**, 094406 (11)]

Fractionalization of orbital excitations: **AF/FO**



orbiton is excited

orbiton moves and creates a spinon

orbiton propagating with hopping $t = J/2$

In a 1D spin-orbital model the orbiton fractionalizes into a freely propagating spinon and orbiton, in analogy to spinon and holon in spin *t-J* model

[K. Wohlfeld *et al*., PRL **107**, 127201 (11)]

t-*J*-like model for ferromagnetic manganites

Double exchange gives **FM** coupling proportional to the kinetic energy

for $La_{1-x}Sr_xMnO_3$ $\boxed{J_{\rm DE} = \frac{1}{2z\mathcal{S}^2}\left|\left\langle \tilde{H}_t^{\uparrow}(e_g)\right\rangle\right|}$

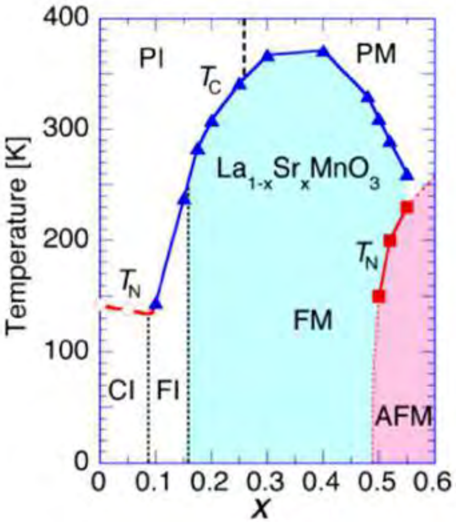
between average spins $\boxed{2\mathcal{S} = 4 - x}$

the frustrating AF superexchange $J_{\rm SE}$ reduces the **FM** coupling $J_{\rm DE}$

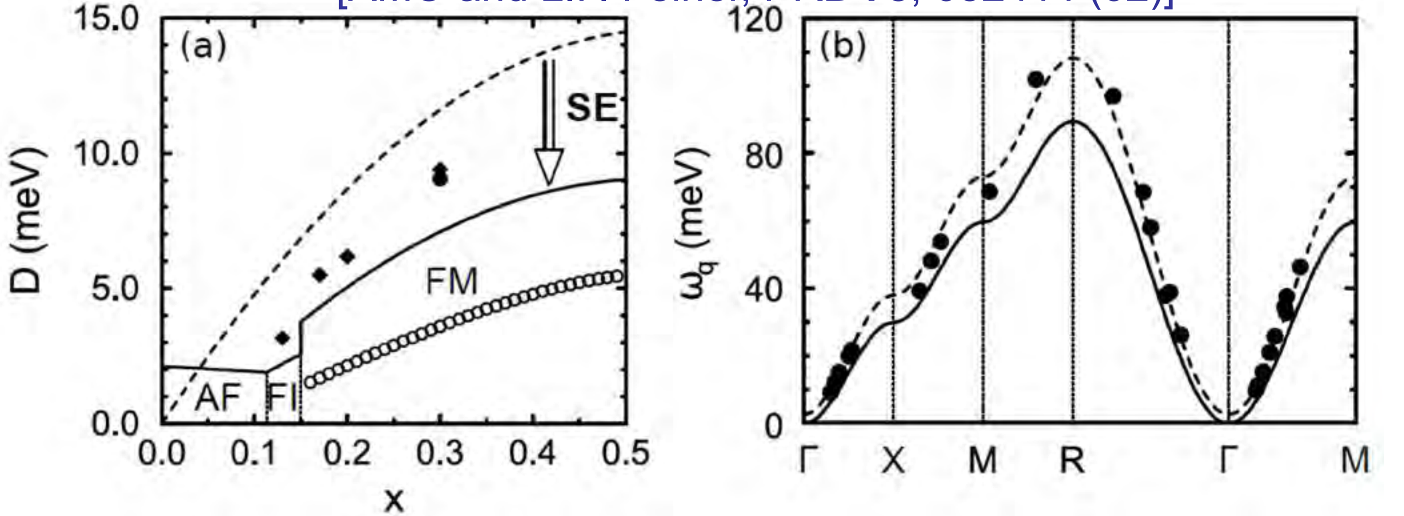
Derivation with Schwinger bosons at doping *x*

magnon dispersion $\boxed{\omega_{\vec{q}} = (J_{\rm DE} - J_{\rm SE})\, z\mathcal{S}^2(1-\gamma_{\vec{q}})}$ where $\gamma_{\vec{q}} = \frac{1}{z}\sum_{\vec{\delta}} e^{i\vec{q}\cdot\vec{\delta}}$

[AMO and L.F. Feiner, PRB **78**, 052414 (02)]



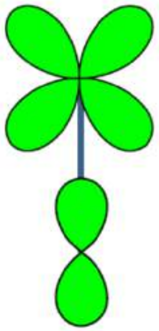
[Y. Tokura, RPP **67**, 797 (06)]



spin-wave stiffness D (solid line) points for $La_{1-x}Sr_xMnO_3$ (diamonds) and $La_{0.7}Pb_{0.3}MnO_3$ (circle)

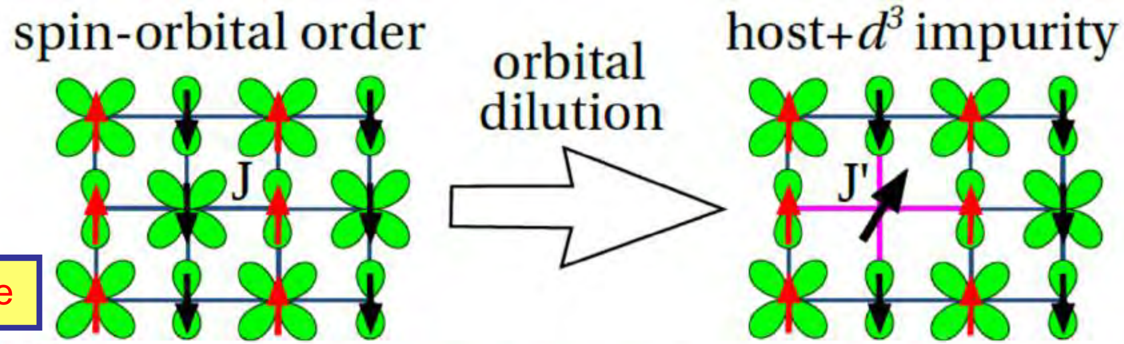
$\omega_{\vec{q}}$ obtained at $x = 0.30$ (solid line) points for $La_{0.7}Pb_{0.3}MnO_3$

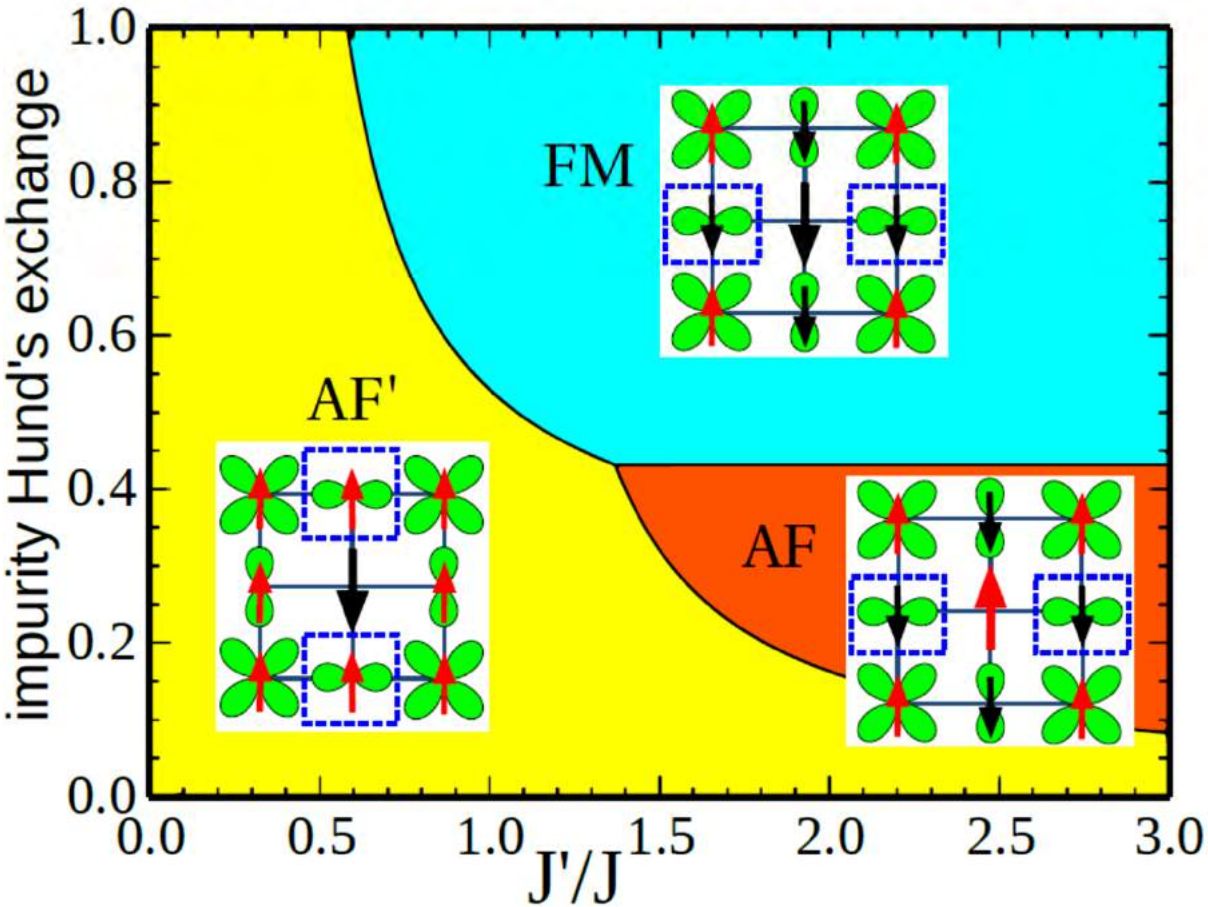
Orbital dilution in d^4 ($S = 1$) by d^3 impurities ($S = 3/2$)



Orbital degree of freedom

C-AF phase





Futuristic electronic devices may rely on properties that are highly sensitive to magnetic and orbital order

spintronics - orbitronics

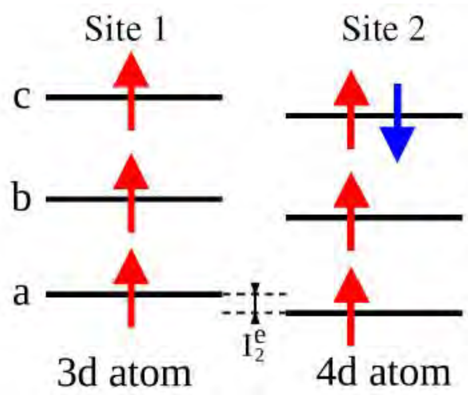
[W. Brzezicki, AMO, M. Cuoco, PRX **5**, 011037 (15)]

Correl17

Parameters for a hybrid 3*d*-4*d* bond

Mismatch potential renormalized by Coulomb *U*'s and Hund's J_H's

$$\boxed{\Delta = I_e + 3(U_1 - U_2) - 4(J_1^H - J_2^H)}$$



S=3/2 **S=1**

Schematic view of a 3*d*-4*d* bond with a doublon in the host at orbital *c*

$$J_{\text{imp}} = \frac{t^2}{4\Delta} \qquad \eta_{\text{imp}} = \frac{J_1^H}{\Delta}$$

$$J_{\text{host}} = \frac{4t_h^2}{U_2} \qquad \eta_{\text{host}} = \frac{J_2^H}{U_2}$$

Parameters to characterize the impurity:

$$\boxed{J_{\text{imp}}/J_{\text{host}}} \qquad \boxed{\eta_{\text{imp}} = \frac{J_1^H}{\Delta}}$$

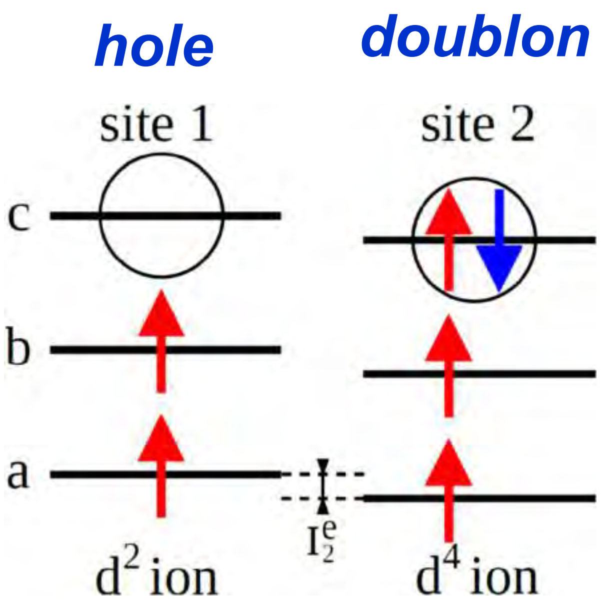
superexchange — Hund's exchange at 3*d*

[W. Brzezicki, AMO, M. Cuoco, PRX **5**, 011037 (15)]

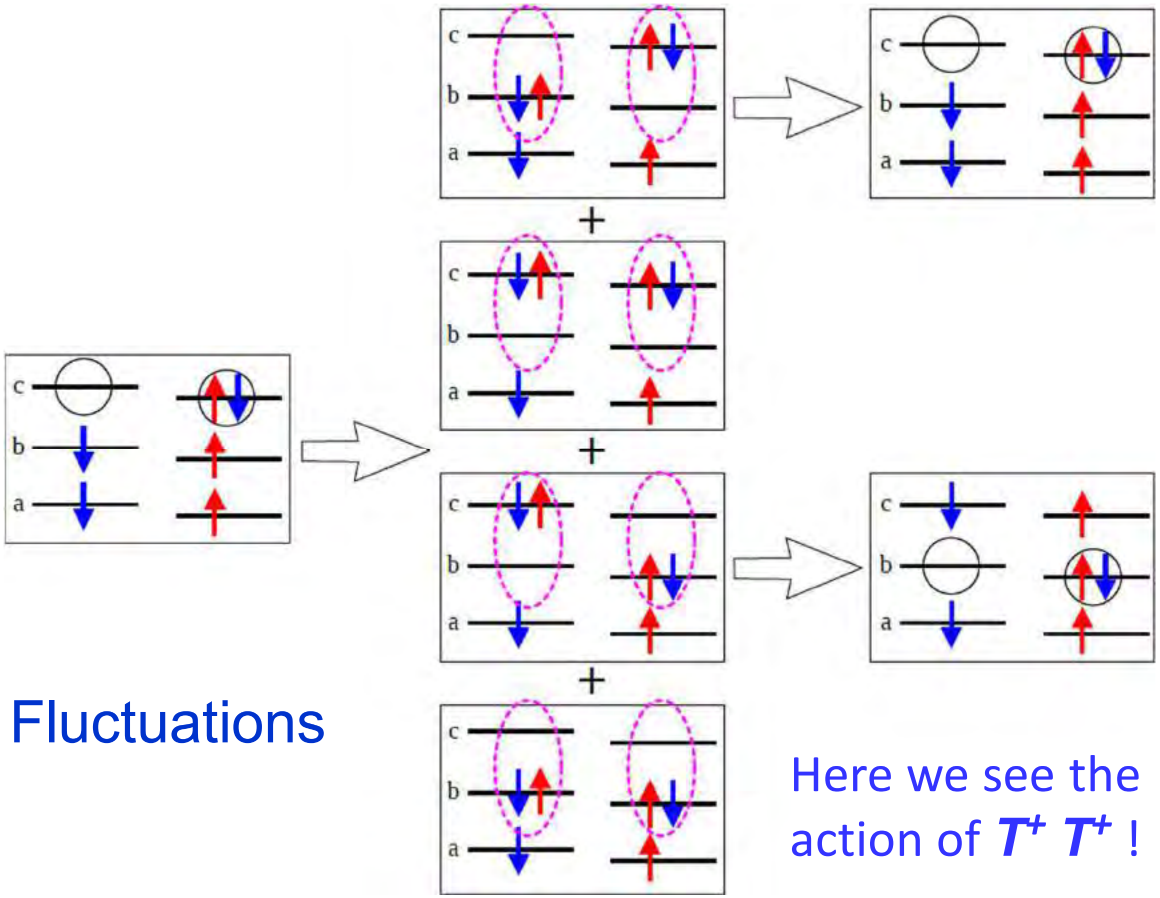
Enhanced fluctuations for d^4-d^2 hybrids (charge dilution)

We solve a problem of two sites in the second order perturbation expansion in hopping to get a **spin-orbital bond Hamiltonian**.

$$|a\rangle \equiv |yz\rangle\,, \quad |b\rangle \equiv |xz\rangle\,, \quad |c\rangle \equiv |xy\rangle$$



Ground state



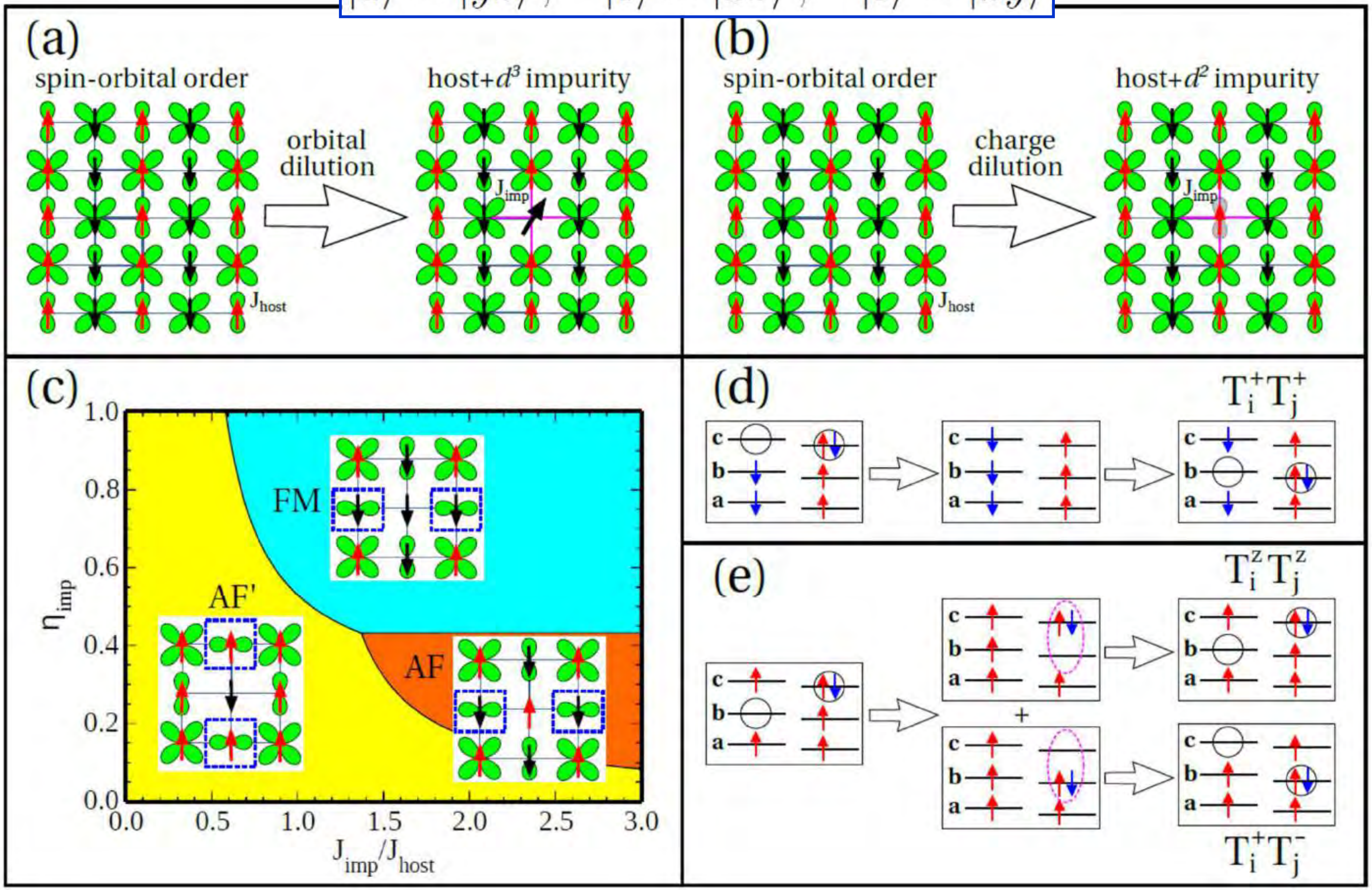
Fluctuations

Here we see the action of $T^+\, T^+$!

Correl17

Enhanced fluctuations for d^4-d^2 hybrids (charge dilution)

$$|a\rangle \equiv |yz\rangle\,, \quad |b\rangle \equiv |xz\rangle\,, \quad |c\rangle \equiv |xy\rangle$$



Frustration & entanglement: orbital and spin-orbital

1. Orbital models: frustration => orbital order at $T < T_c$
2. Features of spin-orbital superexchange models
3. Different properties of systems with e_g and t_{2g} orbitals
4. Spin-orbital entanglement
5. Doped systems: double exchange & novel phases

Quantum Goodenough-Kanamori rules for exchange bonds:

$$\left[\begin{array}{c}\text{spin triplet}\\ \text{orbital singlet}\end{array}\right] \Longleftrightarrow \left[\begin{array}{c}\text{spin singlet}\\ \text{orbital triplet}\end{array}\right]$$

Complementary correlations are dynamical !

Challenge: excitations in spin-orbital systems ?

Thank you for your kind attention !

Correl17

http://sces.if.uj.edu.pl/