Part 2:
3d transition metal oxides

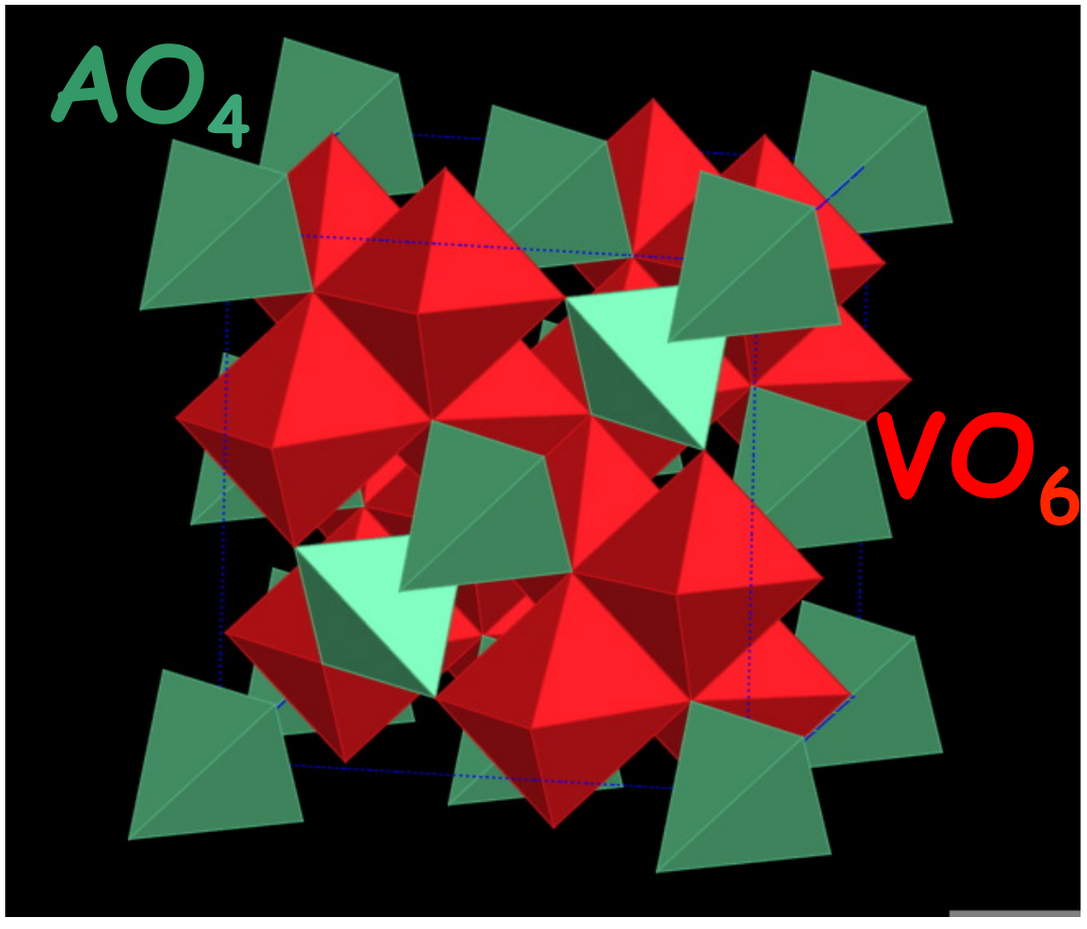
- ZnV_2O_4
- CaV_2O_4
- 1d spin-orbital chain

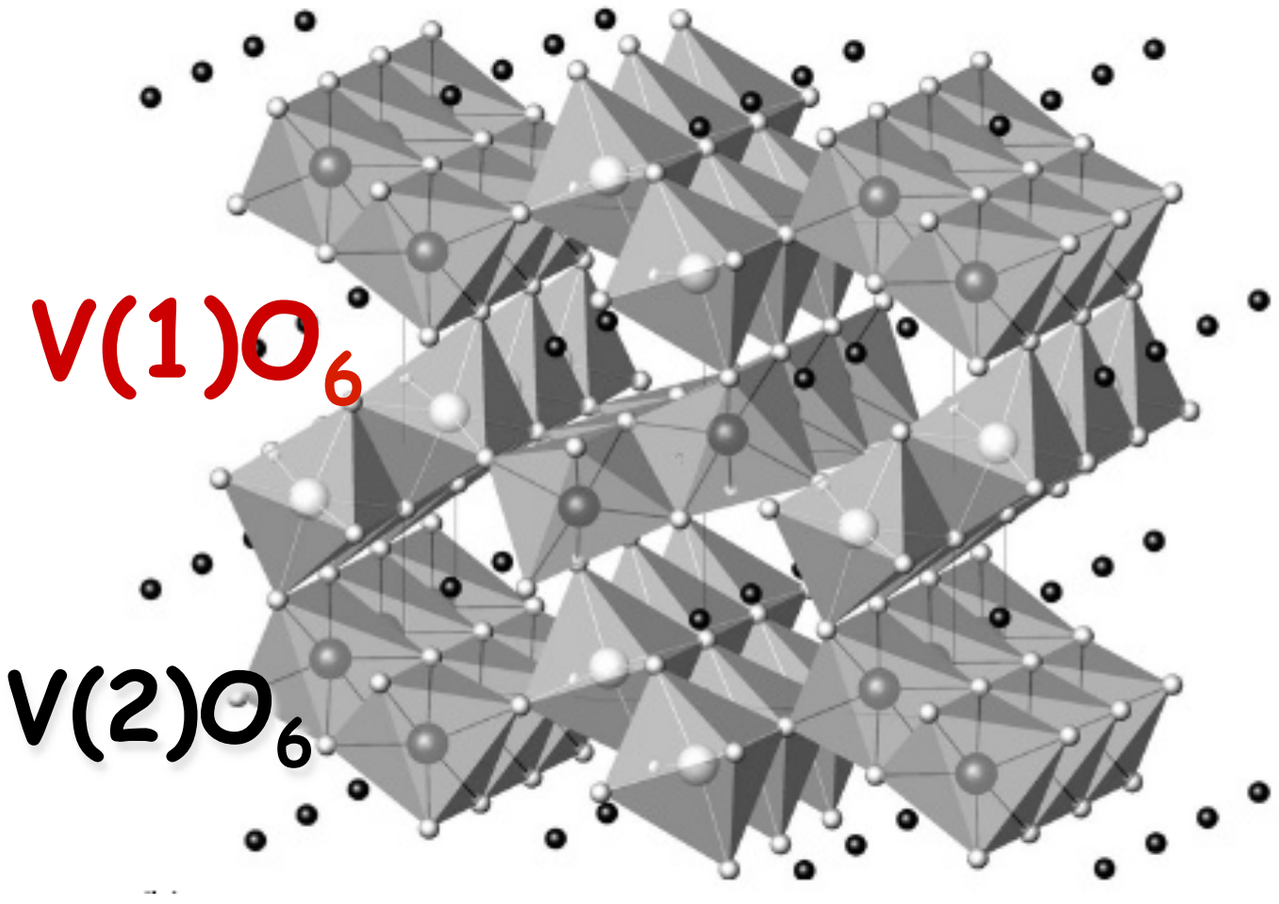
Spin-orbital physics in 3d transition metal oxides

Orbital degrees of freedom should incorporated in the super-exchange theory and the systems are described by means of effective Kugel-Khomskii spin-orbital models. In these models, magnetic and orbital orders are usually connected. The orbital order might be stabilized by lifting of orbital degeneracy either by lattice distortions, orbital interactions or spin orbit coupling.

Giniyat Khaliullin, Prog. Theor. Phys. Suppl. 160, 155 (2005)

Family of AV_2O_4

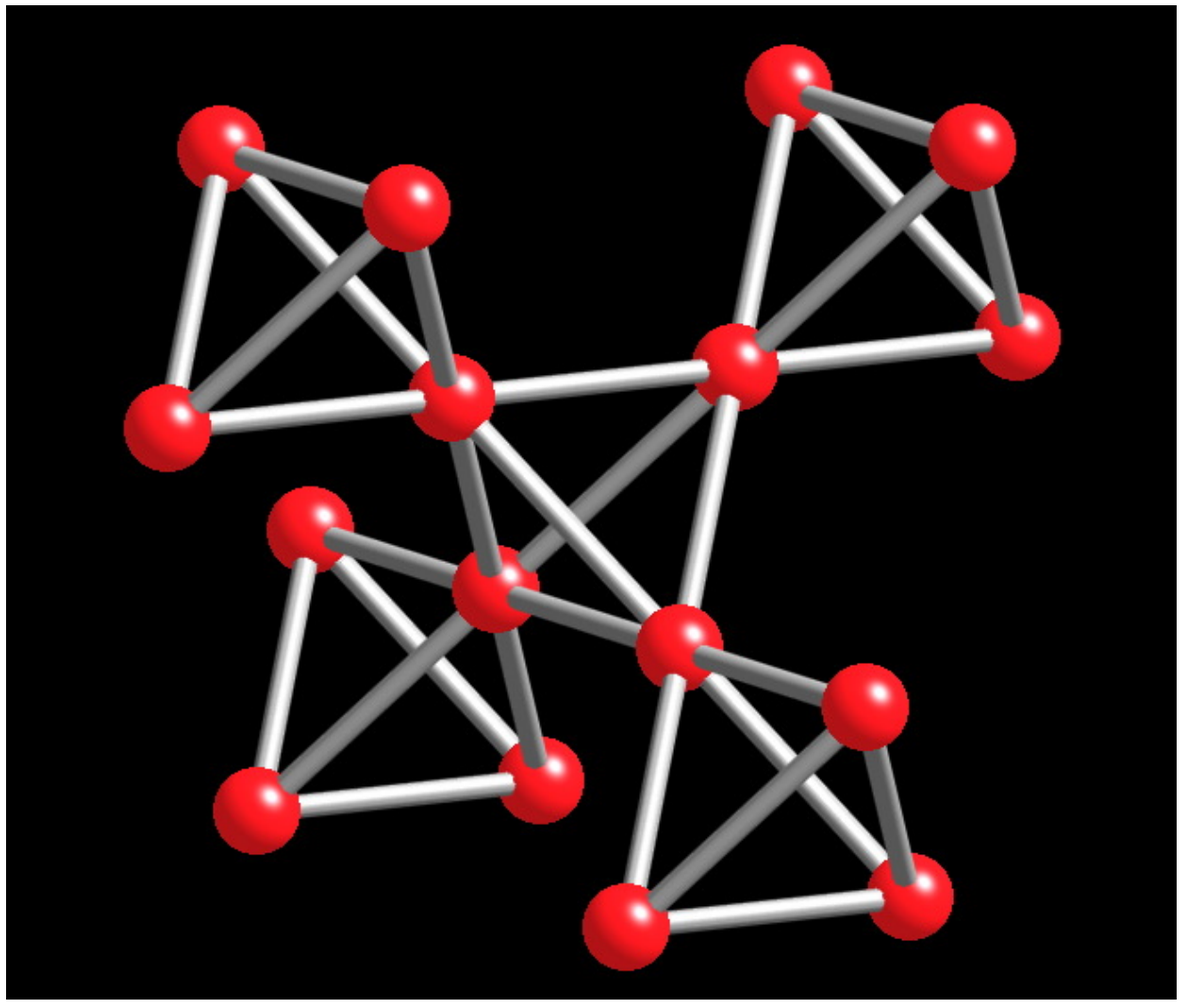


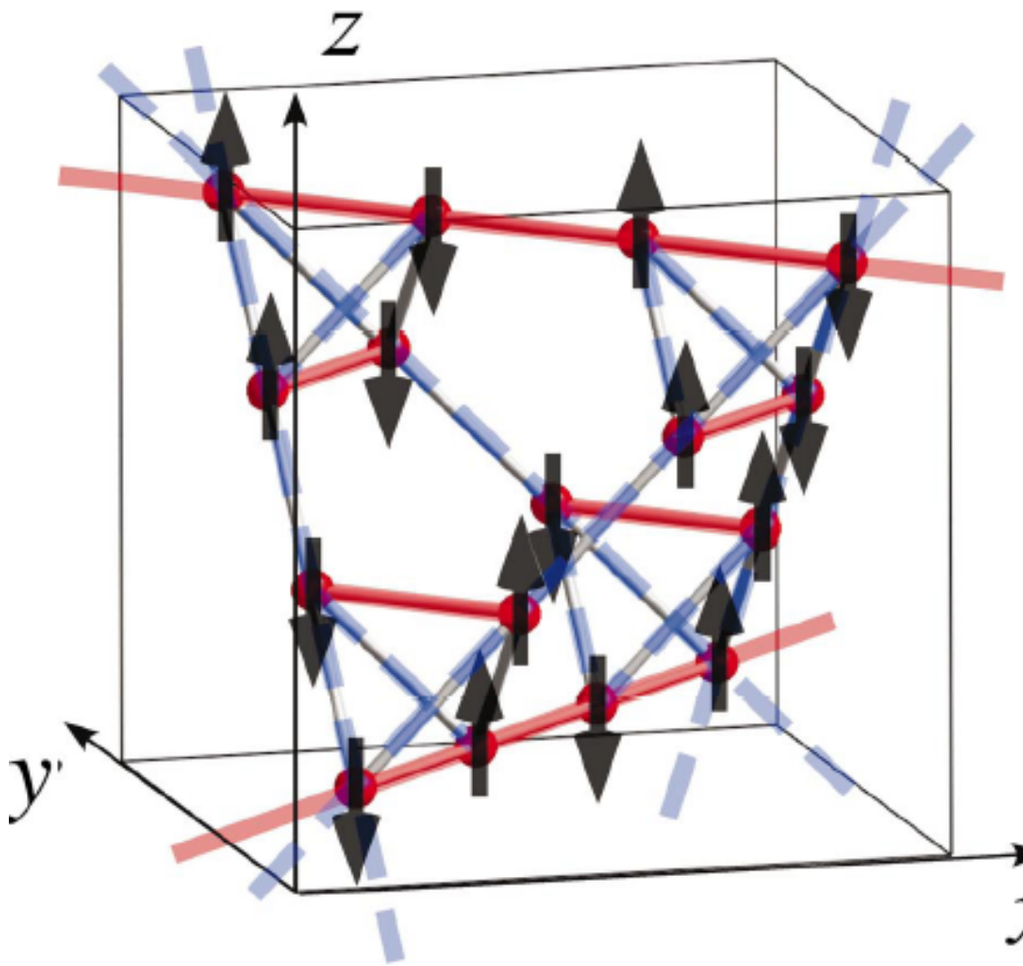


Spinels:
ZnV_2O_4
MnV_2O_4

Calcium-ferrite structure:
CaV_2O_4

Vanadium spinel: ZnV_2O_4





Structural (cubic-tetragonal): Ts=52K

Temperature-induced change in dimensionality, because of orbital degrees of freedom

Magnetic (para-antiferro): T_N=44K

AFM 1D order, consistent with a set of spin chains

Pyrochlore lattice

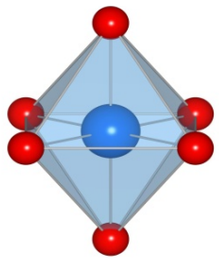
H. Mamiya *et al*, JAP 1997
M. Reehuis *et al*, EPJB 2003
S.-H. Lee *et al*, PRL 2004
E. M. Wheeler et al, PRB 2010
S.-H. Lee et al, PRL 2005

Single-ion properties of V^{3+}

t_{2g} states: $\mathbf{L} = -\alpha \mathbf{L}'$ $(\alpha \approx 1)$.

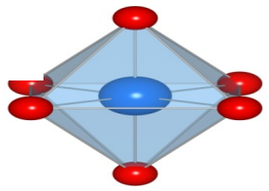
$\mathbf{J}' = \mathbf{S} + \mathbf{L}'$ with $S = 1$, $L' = 1$

$\Rightarrow$ Ground state $J' = 2$.



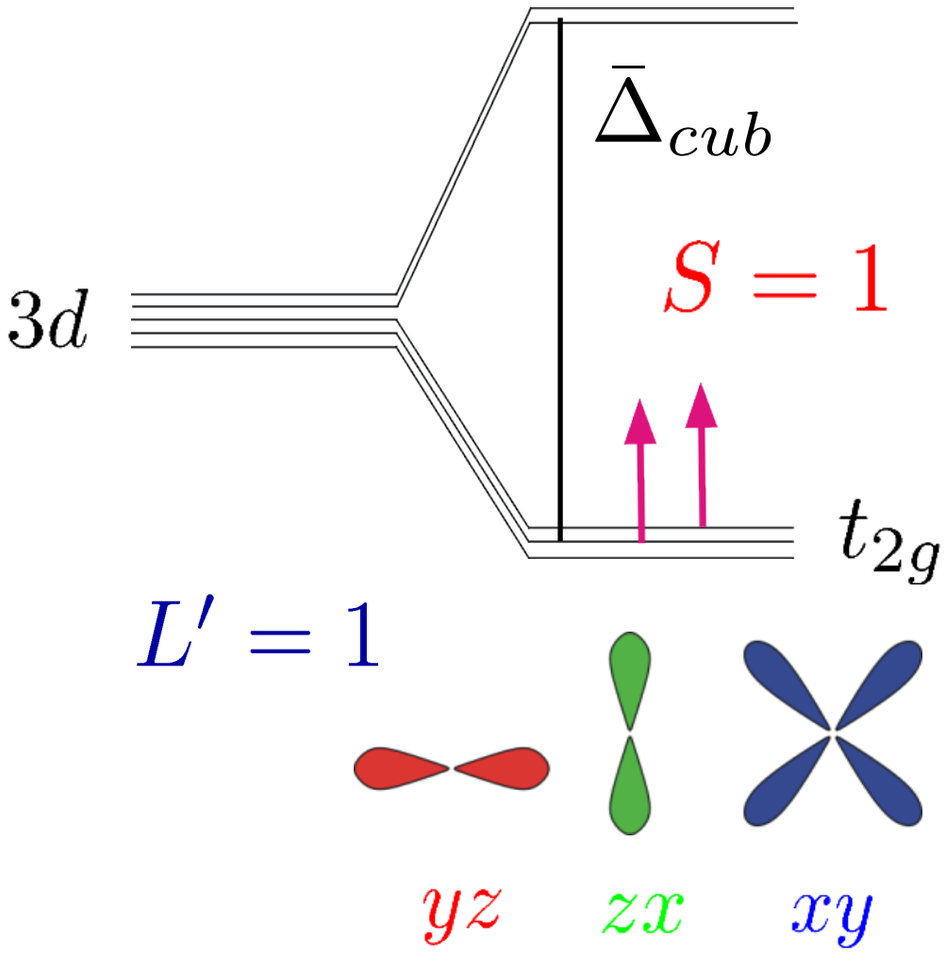
Cubic crystal field

$\bar{\Delta}_{cub} \simeq 2\,\mathrm{eV}$



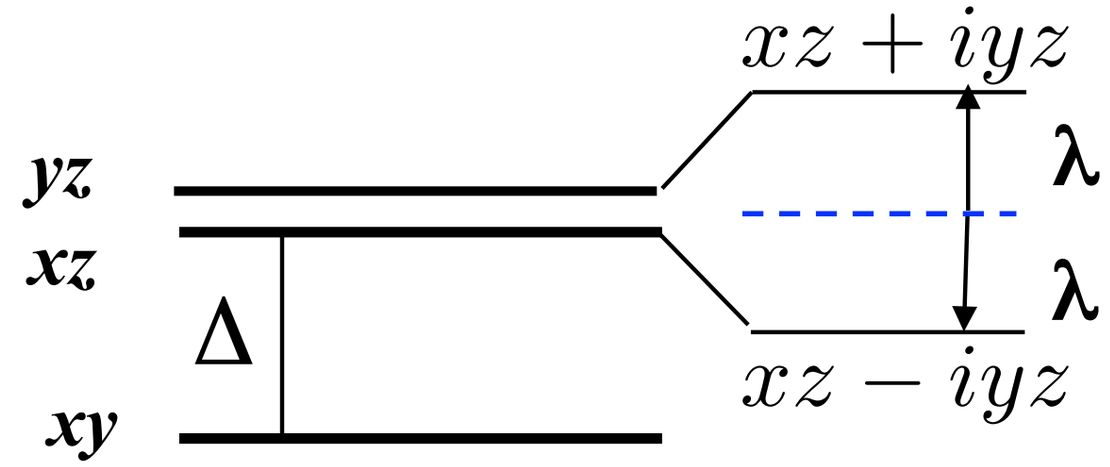
Tetragonal crystal field

$\Delta \simeq 200\,\mathrm{meV}$



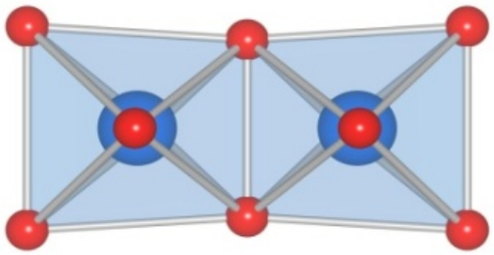
Spin-orbit coupling

$\lambda \simeq 20\mathrm{meV}$

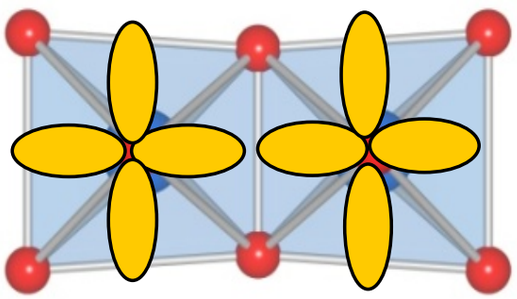


Interactions between t_{2g}-orbitals in AV_2O_4

- Octahedra are edge-sharing



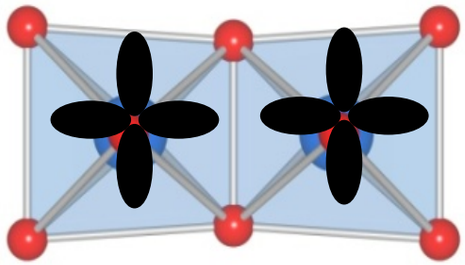
- Only ddσ overlap



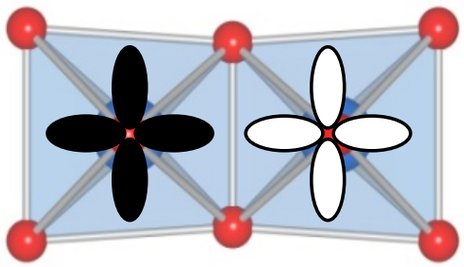
t≠0

Only direct hopping,
only diagonal hopping,
and only along "good" bond

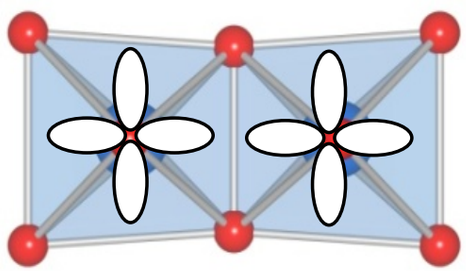
Static Potts-like orbital interactions
(only ddσ overlap)



AFM exchange

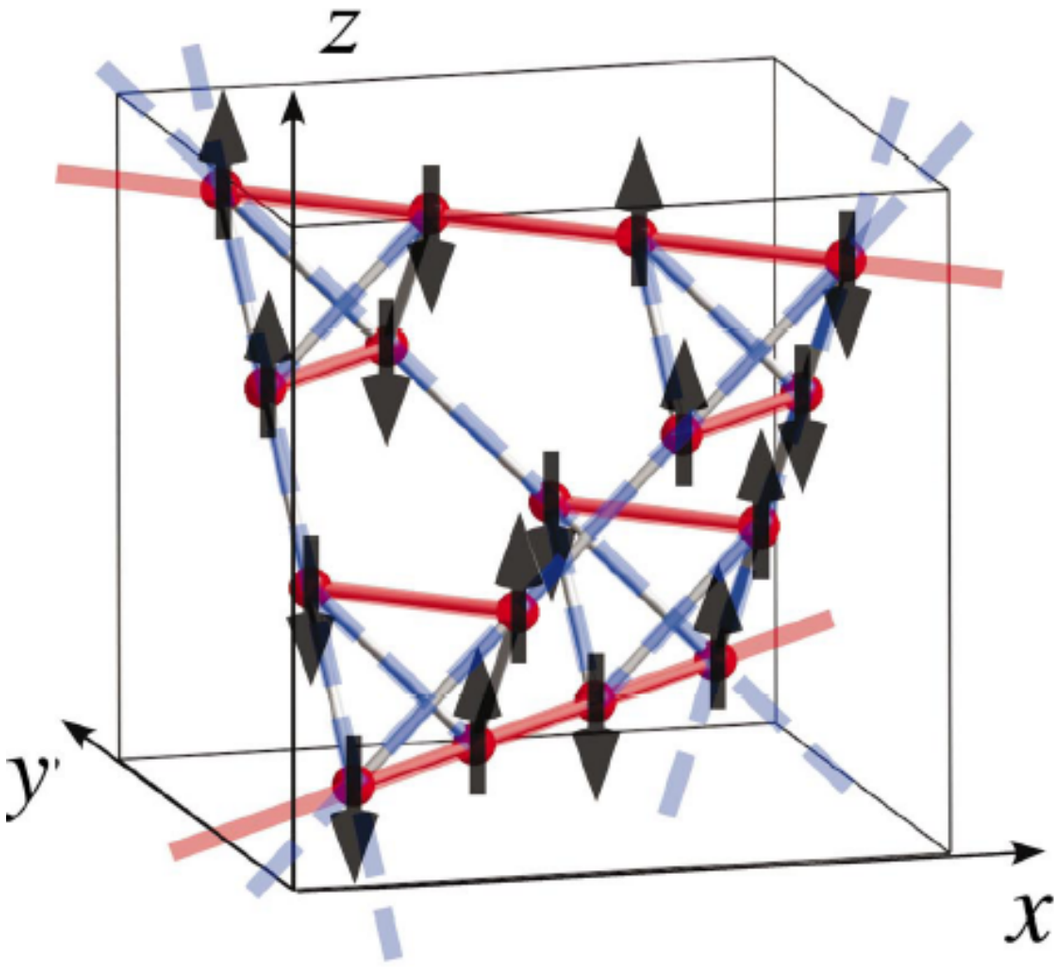


FM exchange



No exchange

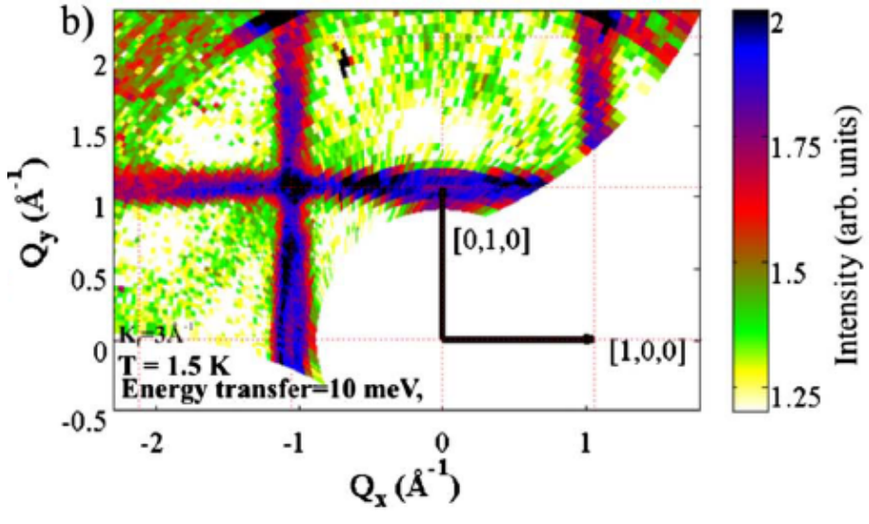
Magnetic order in ZnV_2O_4



Structural (cubic-tetragonal): Ts=52K

xy orbital is occupied

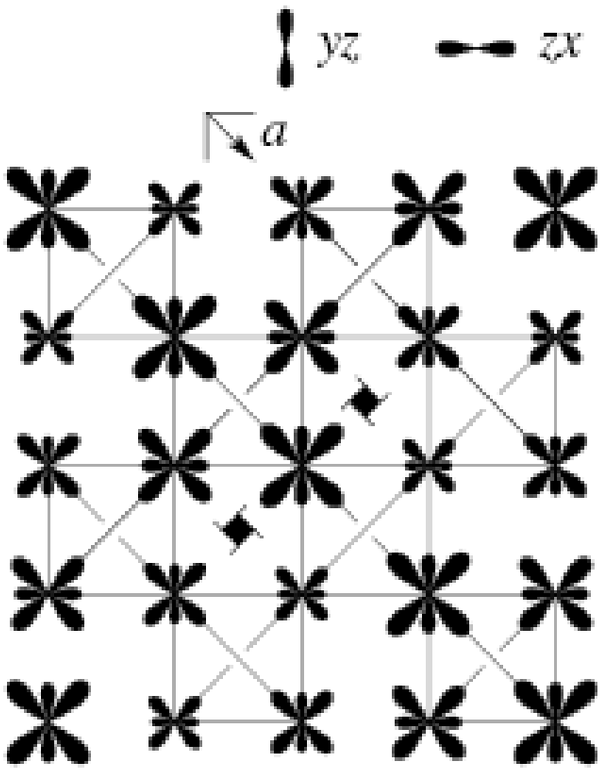
AFM chains along $(1,1,0)_{cub}$.



From E. M. Wheeler *et al*, PRB 2010

Orbital order in ZnV_2O_4

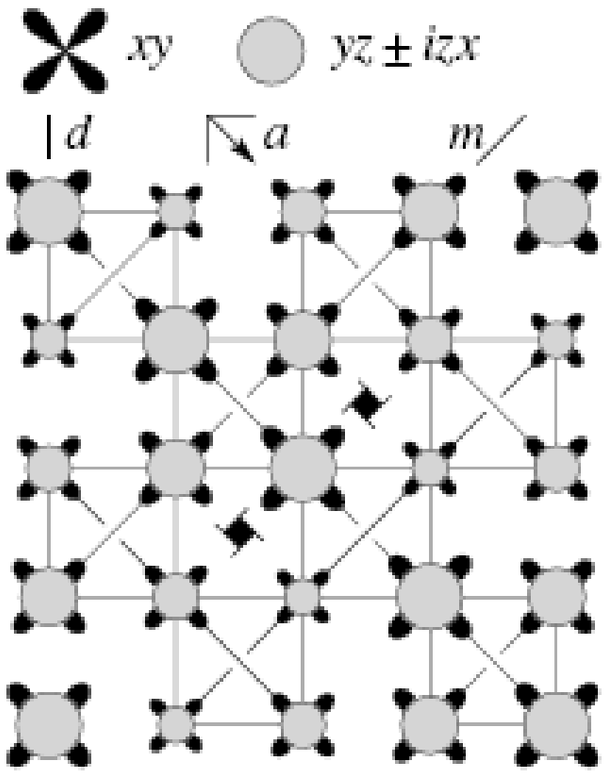
Real Orbital Order



(a $I4_1/a$

Y.Motome and H.Tsunetsugu
PRB 70, 184427

Complex Orbital Order

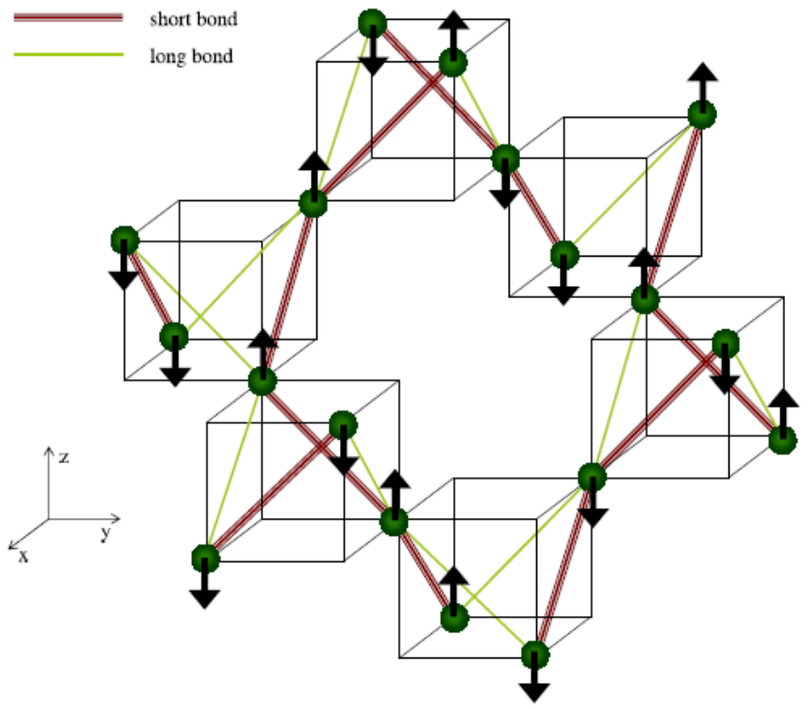


$I4_1/amd$

O.Tchernyshyov
PRL 93 157206

S. Di Matteo, G.Jackeli, NP,
PRB 72 020408(R)

Dimerized state

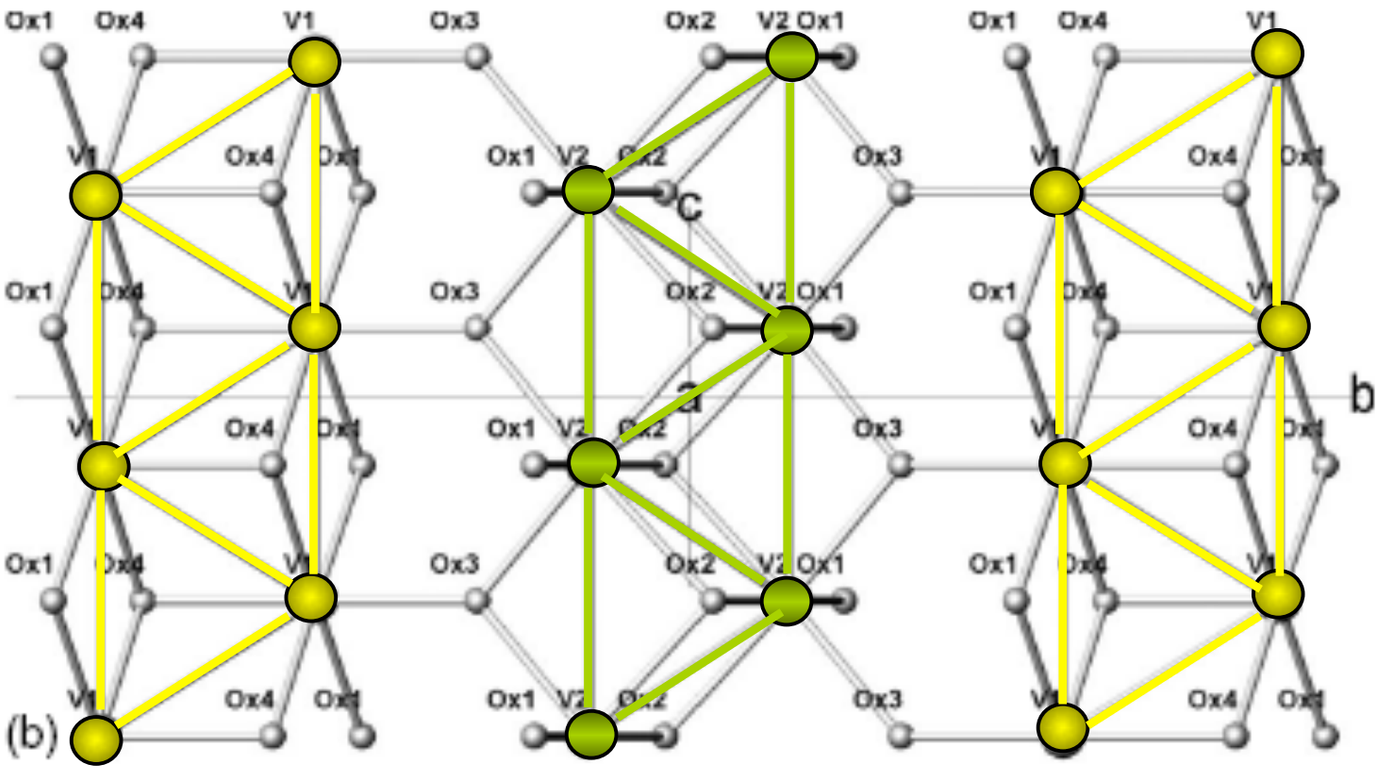


$P4_12_12_1$

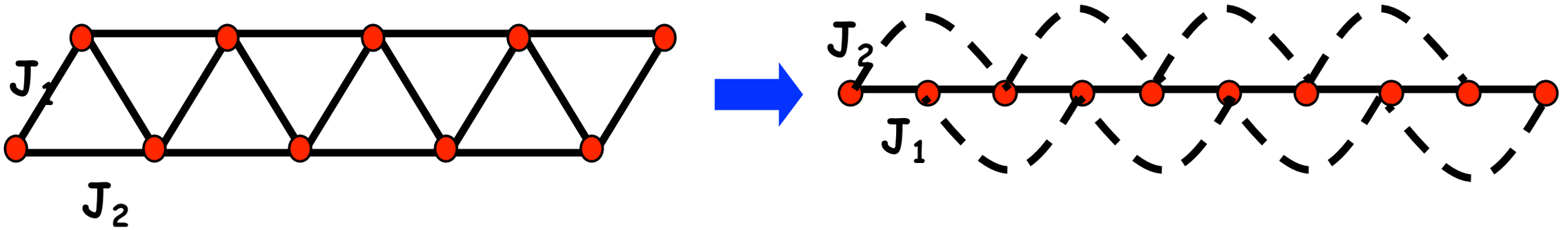
V. Pardo et al. PRL **101**, 256403

supported by SOC

Zigzag vanadium chains in CaV_2O_4



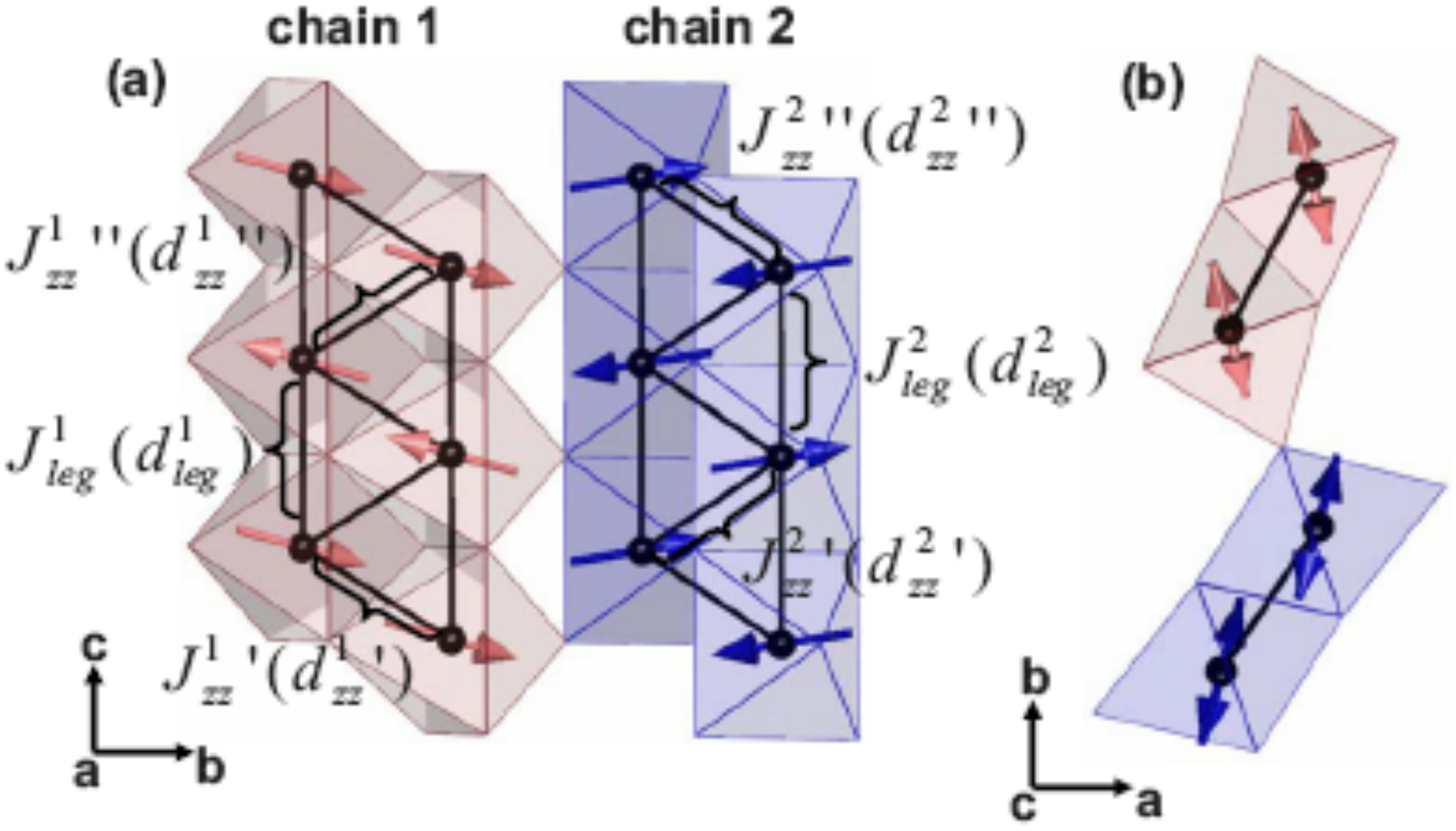
V^{3+} t_{2g}^2 $S = 1$



Zigzag spin-1 chain with comparable nn and nnn interactions + an easy-plane anisotropy of V^{3+} ions

Chiral spin liquid: a long-range chiral order + short range spin correlations

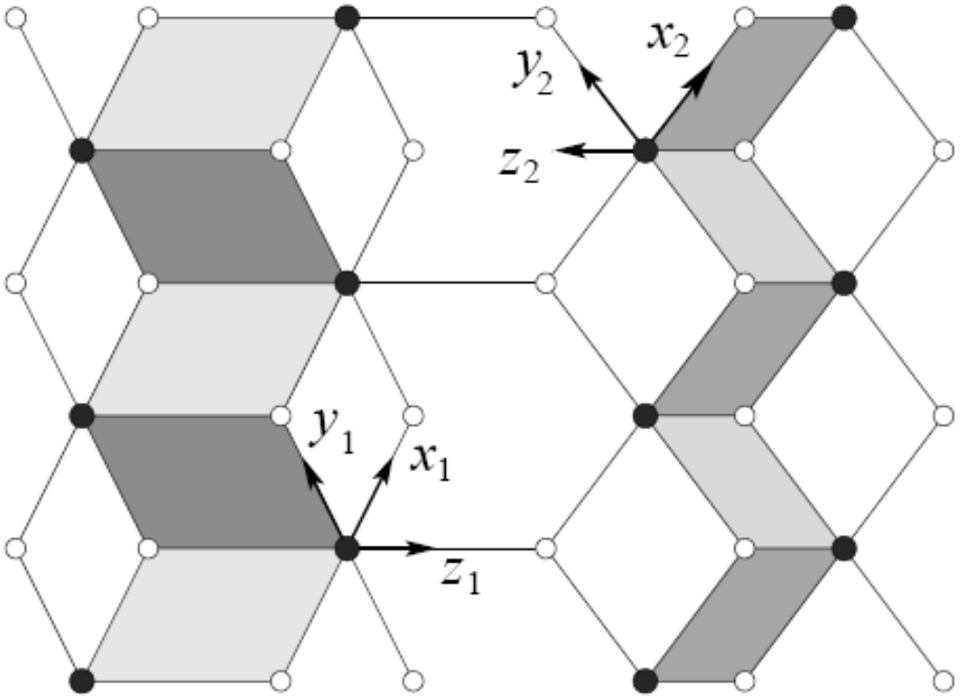
Magnetic order in CaV_2O_4

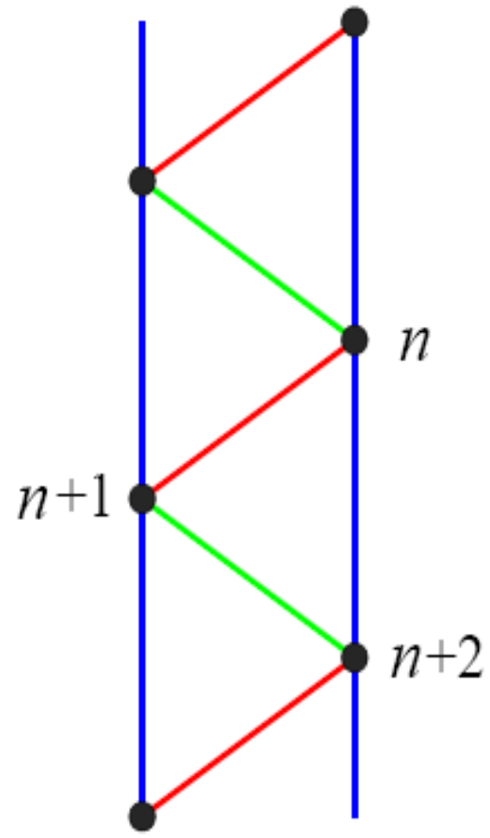


Two collinear antiferromagnetic spin chains canted on 19 degrees with respect to each other.

Pieper et al, PRB 2009, Niazi et al, PRB 2009

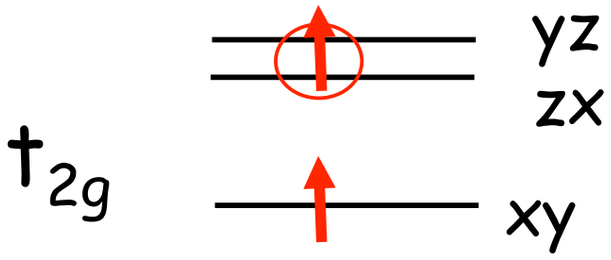
Spin-orbital zigzag chain





Degrees of freedom:

V^{3+} (d^2, $S=1$)



Strong AFM coupling of spins along blue rails. Interaction between spin chains is along zigzag bonds.

Orbital degrees of freedom are Ising variables. Orbital interaction is along red-green zigzag chain.

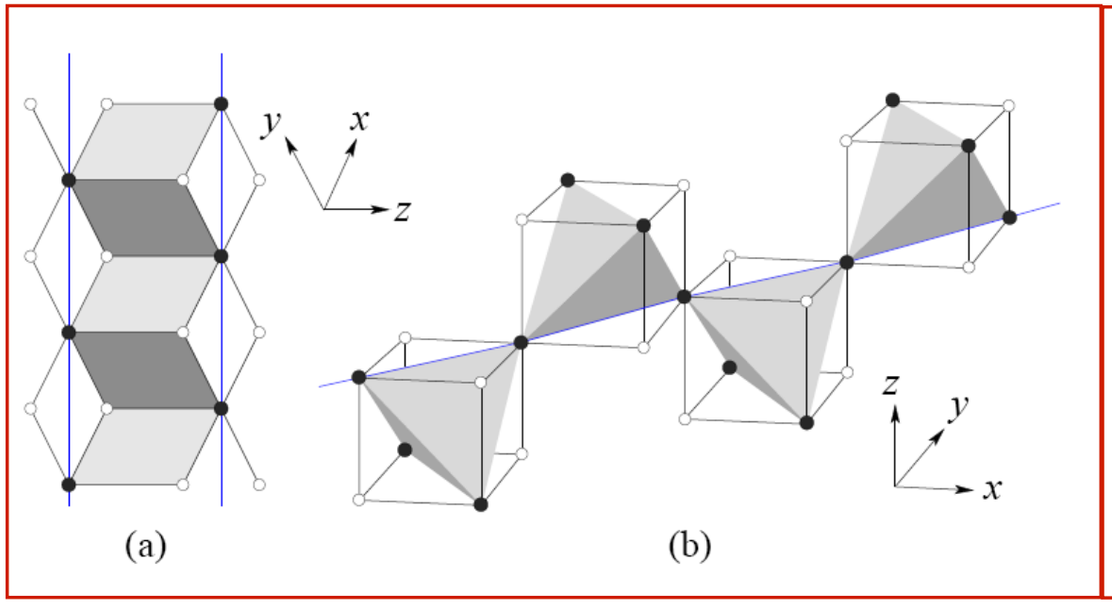
Chern, Perkins, PRB 2009

Motivations for the simplified description

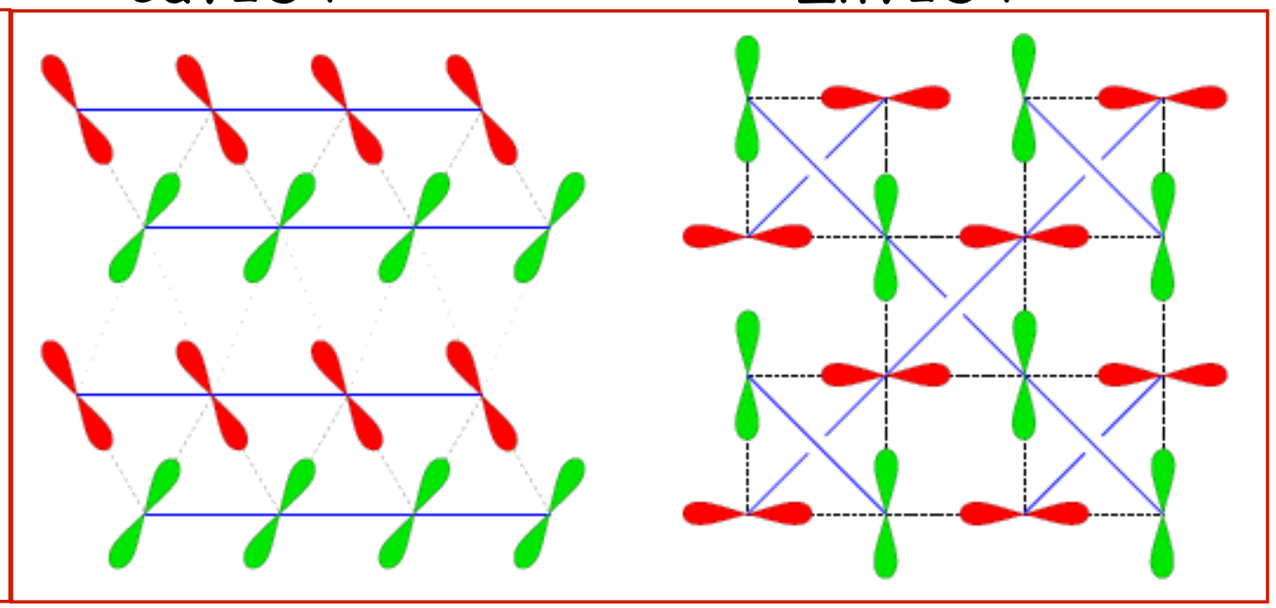
- ZnV2O4 and CaV2O4 are Mott insulators.
- Kugel-Khomskii-type spin-orbital Hamiltonian is a natural approach for describing magnetic and electronic properties.
- The orbital-dependent super-exchange and tetragonal crystal field (xy is always occupied) lead to a formation of 1D orthogonal single chains in ZnV2O4 and zigzag chains CaV2O4.
- Couplings between these spin-orbital chains are weak, but also geometrically frustrated. Thus, ZnV2O4 and CaV2O4 are essentially quasi-1D systems.
- A common feature shared by AV2O4 compounds is the presence of a relativistic spin-orbit interaction.
- Simple toy model: a S=1 Haldane chain and a AFM (FM) Ising chain locally coupled by spin-orbit coupling.

Toy model for quasi 1D vanadates

CaV2O4 ZnV2O4



CaV2O4 ZnV2O4



$$H = J\sum_n \mathbf{S}_n\cdot\mathbf{S}_{n+1} - K\sum_n \tau_n^z\tau_{n+1}^z - \lambda\sum_n \tau_n^x S_n^z.$$

Haldane spin chain

Orbital Ising chain

Spin-orbit coupling

Symmetry: $\mathrm{U}(1)\times Z_2\times Z_2$

Reduction of SU(2) to U(1)xZ2

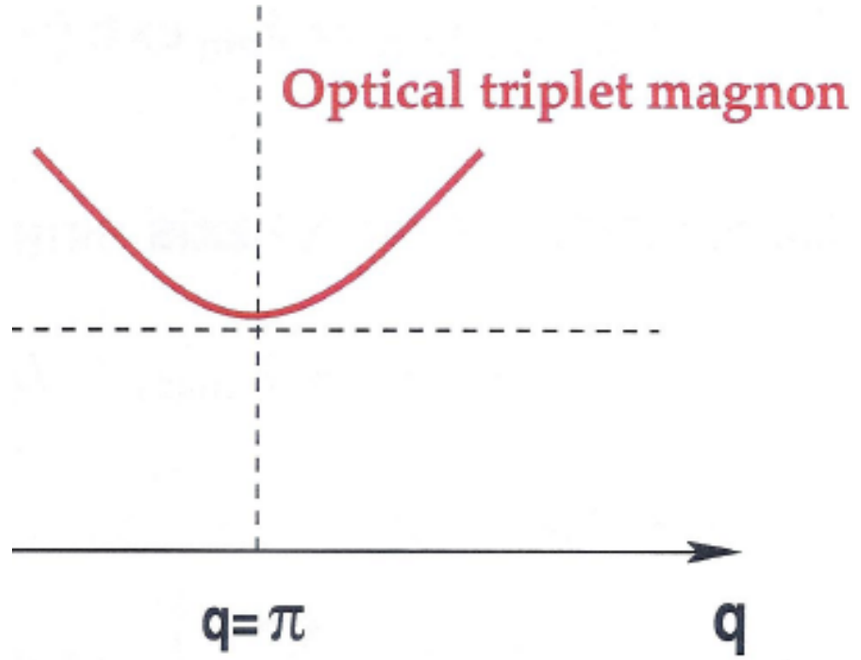
λ=0 limit

Spin-1 chain: a singlet ground state with triply degenerate magnon excitations

$$S^z_n = l^z_n + \tfrac{1}{2}\sum_k \phi_k\, e^{ikn}/\sqrt{L}$$

$$\phi_k = \sqrt{\frac{2v}{\omega_{\pi+k}}}\left(a^z_{\pi+k} + a^{z\dagger}_{-\pi-k}\right)$$

non-linear sigma model



F. D. M. Haldane,
Phys. Lett. 93A, 464 (1983)

S. R. White and D. A. Huse ,
PRB 48,3844 (1993)

I. Affleck ,
PRB 41, 6697 (1990)

Ising orbital chain: ferromagnetically ordered ground state with non-dispersive domain wall excitations .

$$\tau^z_n = \prod_{m<n}\left(2c^\dagger_m c_m - 1\right)\left(c_n + c^\dagger_n\right) \qquad \tau^x_n = 1 - 2c^\dagger_n c_n$$

Jordan-Wigner transformation

Finite λ – Perturbation theory

magnons

Jordan-Wigner fermions

$$H = \sum_q \omega_q a_q^\dagger a_q + \varepsilon_0 \sum_k c_k^\dagger c_k$$

$$+ \frac{2\lambda S}{\sqrt{L}} \sum_{k,q,q'}{}' \left[\cos\left(\frac{q+q'}{2}\right) \phi_k c_q^\dagger c_q + \frac{i}{2} \sin\left(\frac{q-q'}{2}\right) \phi_k (c_q^\dagger c_{q'}^\dagger - c_{q'} c_q) \right]$$

The one-loop corrections to the magnon self-energy

$\Pi_1 =$  $+$

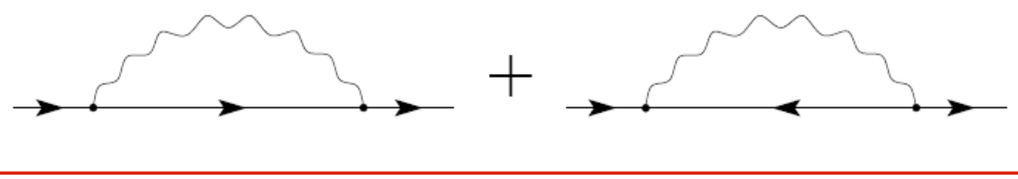
$$\Pi_1(\omega) = \frac{-4\lambda^2 S^2 \varepsilon_0}{4\varepsilon_0^2 - \omega^2}$$

$$\omega \ll K$$

$$\Delta_s \approx \Delta_0 - \frac{v\lambda^2}{4\Delta_0 K}$$

Magnon gap is renormalized

The one-loop corrections to the domain-wall self-energy

$\Sigma_1 =$  $+$

$$\Sigma_1(q) \approx -\frac{\lambda^2}{\Delta_0}\left(1 + e^{-\Delta_0/v}\cos 2q\right)$$

$$\varepsilon_q \approx 2K - \Sigma_1(q)$$

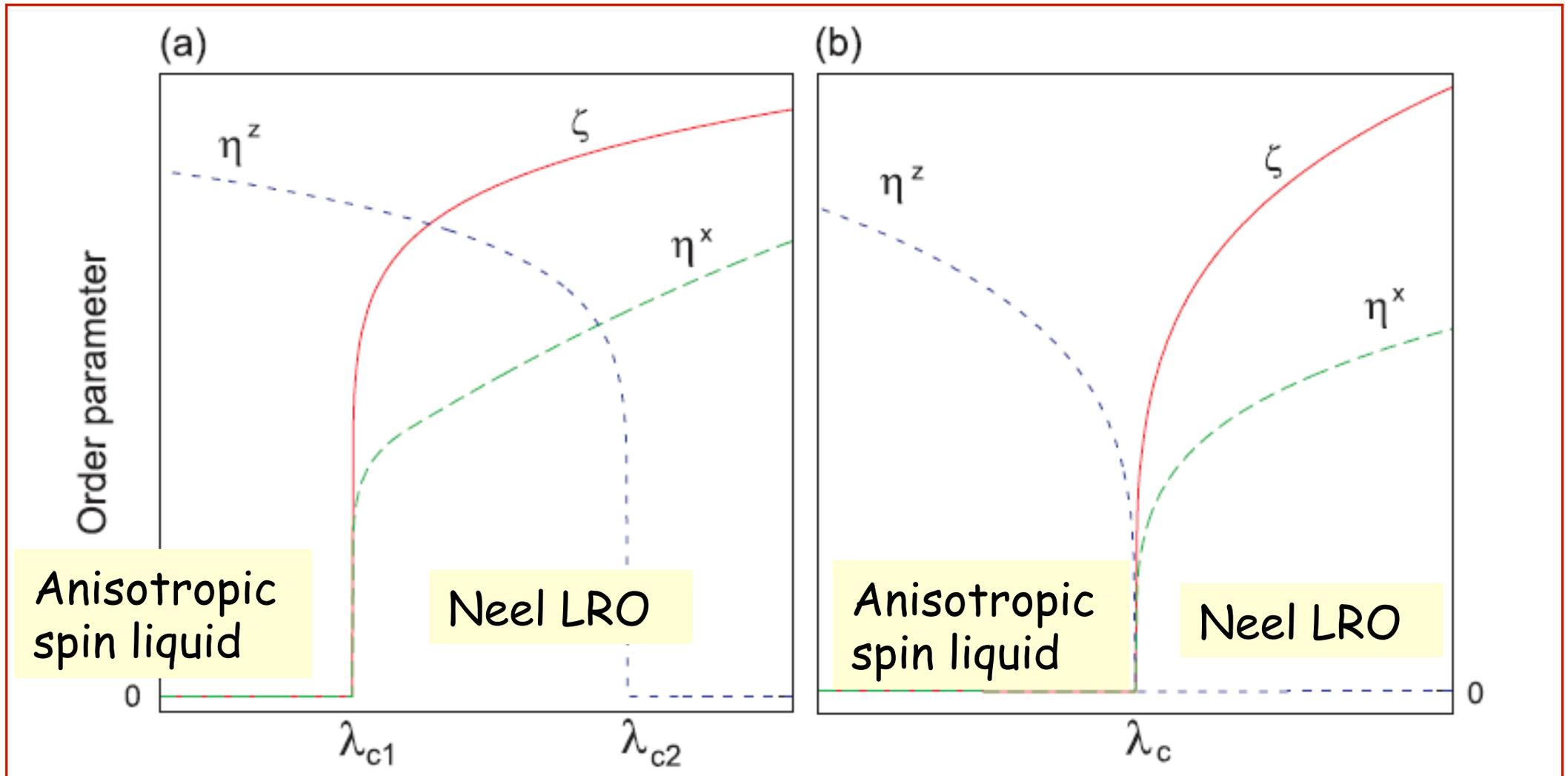
Domain-walls become mobile

Order parameters

$$(-1)^n \langle \tau_n^z \rangle \equiv \eta^z(\lambda)$$

$$(-1)^n \langle \tau_n^x \rangle \equiv \eta^x(\lambda)$$

$$\langle S_n^z \rangle = (-1)^n \zeta(\lambda)$$



$$K \gg \Delta_S$$

$$\Delta_S \gg K$$

physics of vanadium oxides on frustrated lattices

Entanglement of orbital and spin degrees …

Reduction of dimensionality due to orbital anisotropy…

Possibility for spin, orbital and spin-orbital liquids…

New models…

More details in

PHYSICAL REVIEW B 76, 214434 (2007)

Magnetic excitations in vanadium spinels

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[2]*Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA*
[3]*MPIPKS, Nöthnitzer Strasse 38, 01187 Dresden, Germany*
(Received 21 June 2007; revised manuscript received 25 September 2007; published 28 December 2007)

We study magnetic excitations in vanadium spinel oxides AV_2O_4 (A=Zn,Mg,Cd) using two models: the first one is a superexchange model for vanadium $S=1$ spins and the second one includes, in addition, spin-orbit coupling and crystal anisotropy. We show that the experimentally observed magnetic ordering can be obtained in both models; however, the orbital ordering is different with and without spin-orbit coupling and crystal anisotropy. We demonstrate that this difference strongly affects the spin-wave excitation spectrum above the magnetically ordered state, and argue that the neutron measurement of such dispersion is a way to distinguish between the two possible orbital orderings in AV_2O_4.

PHYSICAL REVIEW B 83, 205132 (2011)

Quantum phase transitions in a strongly entangled spin-orbital chain: A field-theoretical approach

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[2]*Andronikashvili Institute of Physics, Tamarashvili 6, 0177, Tbilisi, Georgia*
[3]*Center of Condensed Mater Physics, ITP, Ilia State University, 0162, Tbilisi, Georgia*
[4]*Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA*
(Received 5 January 2011; revised manuscript received 23 March 2011; published 26 May 2011)

Motivated by recent experiments on quasi-one-dimensional vanadium oxides, we study quantum phase transitions in a one-dimensional spin-orbital model describing a Haldane chain and a classical Ising chain locally coupled by the relativistic spin-orbit interaction. By employing a field-theoretical approach, we analyze the topology of the ground-state phase diagram and identify the nature of the phase transitions. In the strong coupling limit, a long-range Néel order of entangled spin and orbital angular momenta appears in the ground state. We find that, depending on the relative scales of the spin and orbital gaps, the linear chain follows two distinct routes to reach the Néel state. First, when the orbital exchange is the dominating energy scale, a two-stage ordering takes place in which the magnetic transition is followed by melting of the orbital Ising order; both transitions belong to the two-dimensional Ising universality class. In the opposite limit, the low-energy orbital modes undergo a continuous reordering transition which represents a line of Gaussian critical points. On this line the orbital degrees of freedom form a Tomonaga–Luttinger liquid. We argue that the emergence of the Gaussian criticality results from merging of the two Ising transitions in the strong hybridization region where the characteristic spin and orbital energy scales become comparable. Finally, we show that, due to the spin-orbit

PHYSICAL REVIEW B 82, 172408 (2010)

Quantum criticality of vanadium chains with strong relativistic spin-orbit interaction

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[2]*Andronikashvili Institute of Physics, Tamarashvili str. 6, 0177 Tbilisi, Georgia*
[3]*Ilia State University, Colokashvili Avenue 3-5, 0162 Tbilisi, Georgia*
(Received 12 October 2010; published 18 November 2010)

We study quantum phase transitions induced by the on-site spin-orbit interaction $\lambda \mathbf{L}\cdot\mathbf{S}$ in a toy model of vanadium chains. In the $\lambda \to 0$ limit, the decoupled spin and orbital sectors are described by a Haldane and an Ising chain, respectively. The gapped ground state is composed of a ferro-orbital order and a spin liquid with finite correlation lengths. In the opposite limit, strong spin-orbital entanglement results in a simultaneous spin and orbital-moment ordering, which can be viewed as an orbital liquid. Using a combination of analytical arguments and density-matrix renormalization-group calculation, we show that an intermediate phase, where the ferro-orbital state is accompanied by a spin Néel order, is bounded on both sides by Ising transition lines. Implications for vanadium compounds CaV_2O_4 and ZnV_2O_4 are also discussed.

PHYSICAL REVIEW B 80, 220405(R) (2009)

Model for frustrated spin-orbital chains as applied to CaV_2O_4

Gia-Wei Chern and Natalia Perkins
Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA
(Received 28 July 2009; published 10 December 2009)

Motivated by recent interest in quasi-one-dimensional compound CaV_2O_4, we study the ground states of a spin-orbital chain characterized by an Ising-like orbital Hamiltonian and frustrated interactions between $S=1$ spins. The on-site spin-orbit interaction and the Jahn-Teller effect compete with intersite superexchange leading to a rich phase diagram in which an antiferro-orbital phase is separated from the orbital paramagnet by a continuous Ising transition. Two distinct spin liquids depending on the underlying orbital order are found in the limit of small spin-orbit coupling. In the opposite limit, the zigzag chain behaves as a spin-2 chain with Ising anisotropy. The implications for CaV_2O_4 are discussed.

PRL 108, 247215 (2012) PHYSICAL REVIEW LETTERS week ending 15 JUNE 2012

Orbital Disorder Induced by Charge Fluctuations in Vanadium Spinels

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[1]*Theoretical Division, T4 and CNLS, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[3]*Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA*
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(Received 16 December 2011; published 15 June 2012)

Motivated by recent experiments on vanadium spinels, AV_2O_4, that show an increasing degree of electronic delocalization for smaller cation sizes, we study the evolution of orbital ordering (OO) between the strong and intermediate-coupling regimes of a multiorbital Hubbard Hamiltonian. The underlying magnetic ordering of the Mott insulating state leads to a rapid suppression of OO due to enhanced charge fluctuations along ferromagnetic bonds. Orbital double occupancy is rather low at the transition point indicating that the system is in the crossover region between strong and intermediate-coupling regimes when the orbital degrees of freedom become disordered.

PHYSICAL REVIEW B 72, 020408(R) (2005)

Orbital order in vanadium spinels

S. Di Matteo,[1,2] G. Jackeli,[3,*] and N. B. Perkins[1,4,5]
[1]*Laboratori Nazionali di Frascati INFN, via E. Fermi 40, C.P. 13, I-00044 Frascati (Roma), Italy*
[2]*Dipartimento di Fisica, Università di Roma III, via della Vasca Navale 84, I-00146 Roma, Italy*
[3]*Institute for Theoretical Physics, Ecole Polytechnique Fédérale de Lausanne, CH-1025, Lausanne, Switzerland*
[4]*Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Str. 38 01187 Dresden, Germany*
[5]*Bogoliubov Laboratory of Theoretical Physics, JINR, 141980, Dubna, Russia*
(Received 2 May 2005; published 25 July 2005)

Motivated by recent theoretical and experimental controversy, we present a theoretical study to clarify the orbital symmetry of the ground state of vanadium spinel oxides AV_2O_4 (A=Zn, Mg, Cd). The study is based on an effective Hamiltonian with spin-orbital superexchange interaction and a local spin-orbit coupling term. We construct a classical phase diagram and prove the complex orbital nature of the ground state. Remarkably, with our analysis we predict correctly also the coherent tetragonal flattening of oxygen octahedra. Finally, through analytical considerations as well as numerical *ab initio* simulations, we propose how to detect the predicted complex orbital ordering through vanadium K-edge resonant x-ray scattering.

PHYSICAL REVIEW B 80, 180409(R) (2009)

Large-J approach to strongly coupled spin-orbital systems

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(Received 21 August 2009; revised manuscript received 13 October 2009; published 11 November 2009)

We present an approach to study the ground-state and elementary excitations in compounds where spins and orbitals are entangled by on-site relativistic spin-orbit interaction. The appropriate degrees of freedom are localized states with an effective angular momentum J. We generalize J to arbitrary large values while maintaining the delicate spin-orbital entanglement. After projecting the intersite exchange interaction to the manifold of effective spins, a systematic $1/J$ expansion of the effective Hamiltonian is realized using the Holstein-Primakoff transformation. Applications to representative compounds Sr_2IrO_4 and particularly vanadium spinels AV_2O_4 are discussed.

PHYSICAL REVIEW B 81, 125127 (2010)

Quantum 120° model on pyrochlore lattice: Orbital ordering in MnV_2O_4

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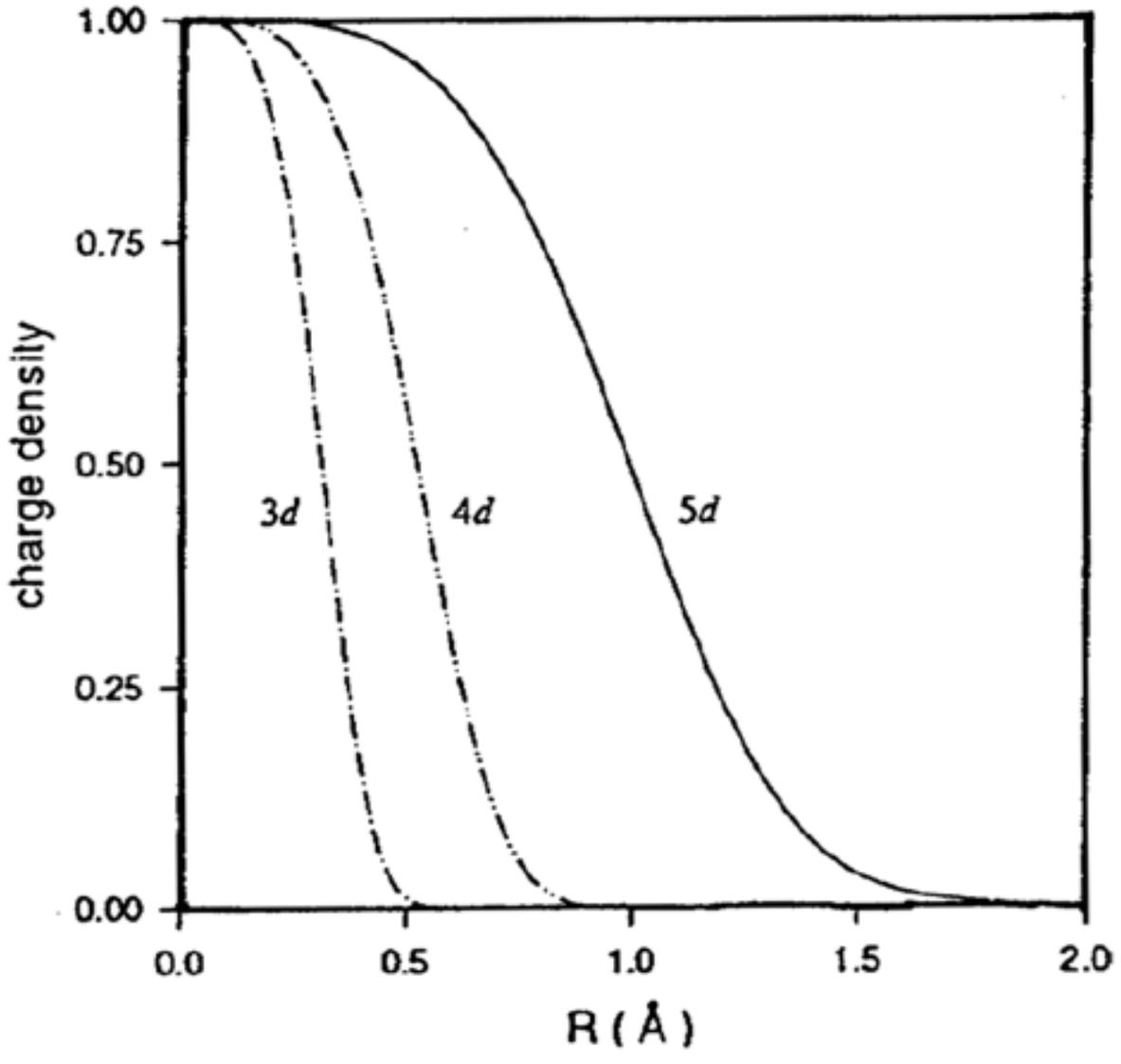
(Received 31 December 2009; published 26 March 2010)

We present an analytical model of orbital ordering in vanadium spinel MnV_2O_4. The model is based on recent first-principles calculation indicating a strong trigonal distortion at the vanadium sites of this compound [S. Sarkar, T. Maitra, R. Valentí, and T. Saha-Dasgupta Phys. Rev. Lett. **102**, 216405 (2009)]. At the single-ion level, the trigonal crystal field leaves a doubly degenerate atomic ground state and breaks the approximate rotational symmetry of t_{2g} orbitals. We find that the effective interaction between the low-energy doublets is described by a quantum antiferromagnetic 120° model on the pyrochlore lattice. We obtain the classical ground state and show its stability against quantum fluctuations. The corresponding orbital order consisting of two inequivalent orbital chains is consistent with the experimentally observed tetragonal symmetry. A periodic modulation of electron density function along orbital chains is shown to arise from the staggering of local trigonal axes. In the presence of orbital order, single-ion spin anisotropy arising from relativistic spin-orbit interaction stabilizes the experimentally observed orthogonal magnetic structure.

Part 3:
4d and 5d transition metal oxides

- spin-orbit assisted Mott insulator
- Kitaev interaction in real material
- Na_2IrO_3
- K_1-K_2 model

Is Mott insulating state possible for 5d-compounds?



5d orbitals are extended

Coulomb repulsion is screened

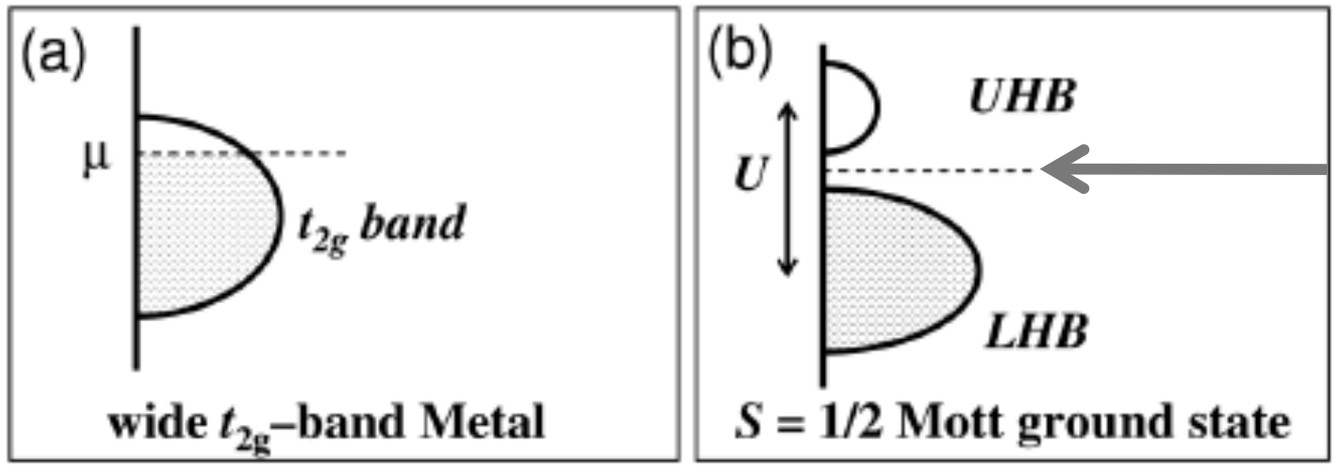
U: 0.5 – 2.5 eV

The Mott criteria
U ≈W
is difficult to satisfy

Iridates should be metals or band insulators

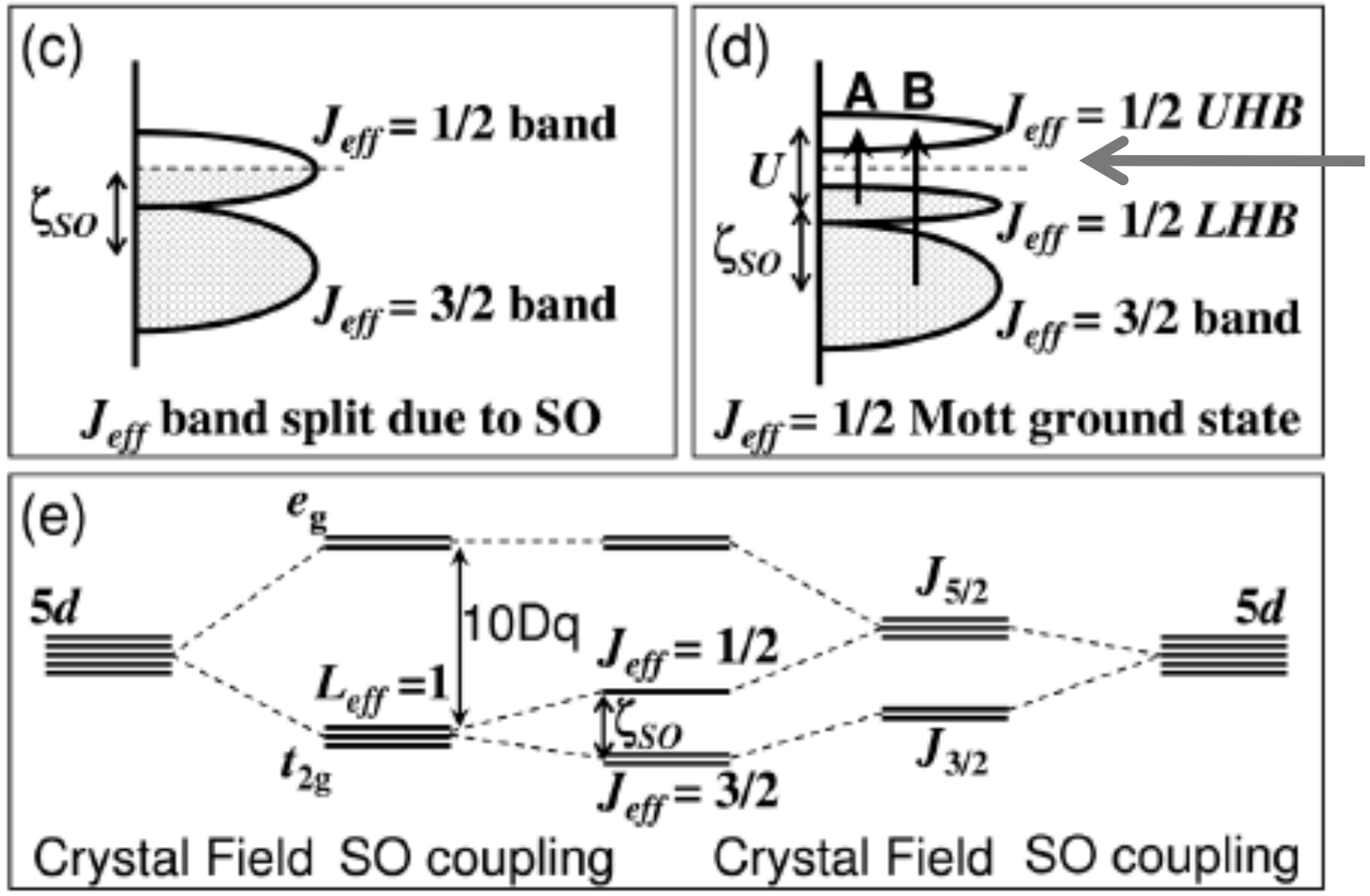
Iridates: spin-orbit assisted Mott insulator

Sr_2IrO_4

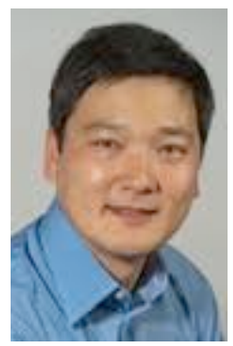


Unrealistically large U (~ 8-10 eV)

H_{SO}: two splited bands



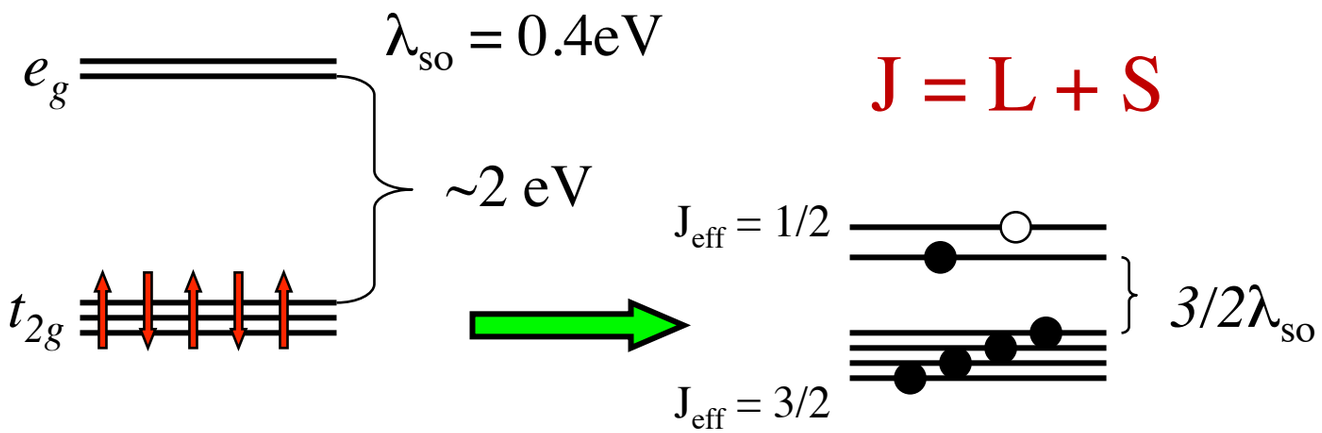
U~ W: effectively enhanced correlation effect



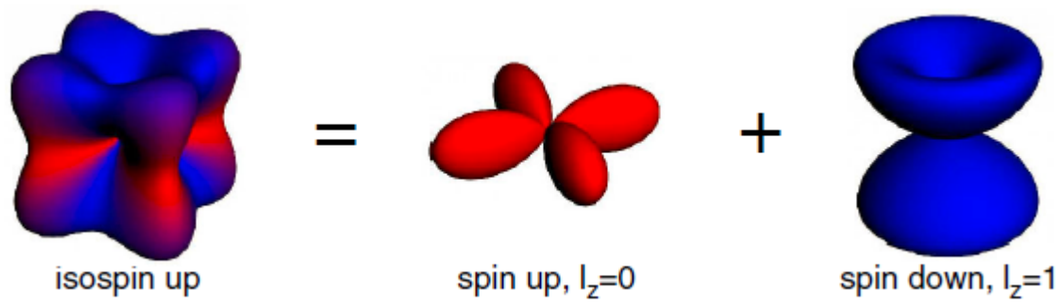
B. J. Kim et al., PRL 101 (2008) 076402

Pseudospins instead of spins in systems with strong SOC

Ir^{4+} - $5d^5$
Rh^{4+} - $4d^5$
Ru^{3+} - $4d^5$



$$J_{eff\,1/2} = \frac{1}{\sqrt{3}}\left(\left|xy,\pm 1/2\right\rangle \pm \left|yz,\mp 1/2\right\rangle + i\left|zx,\mp 1/2\right\rangle\right)$$



G. Jackeli and G. Khaliullin,
PRL 102, 017205 (2009)

Why $J_{eff} = 1/2$ magnets are interesting?

1) Complex phase *i*, coming from the contribution of orbital angular momentum into J, will manifest itself in magnetic coupling.

2) The form of magnetic interactions is no longer dictated by spin symmetry alone but is determined by the combination of spin and lattice symmetries.

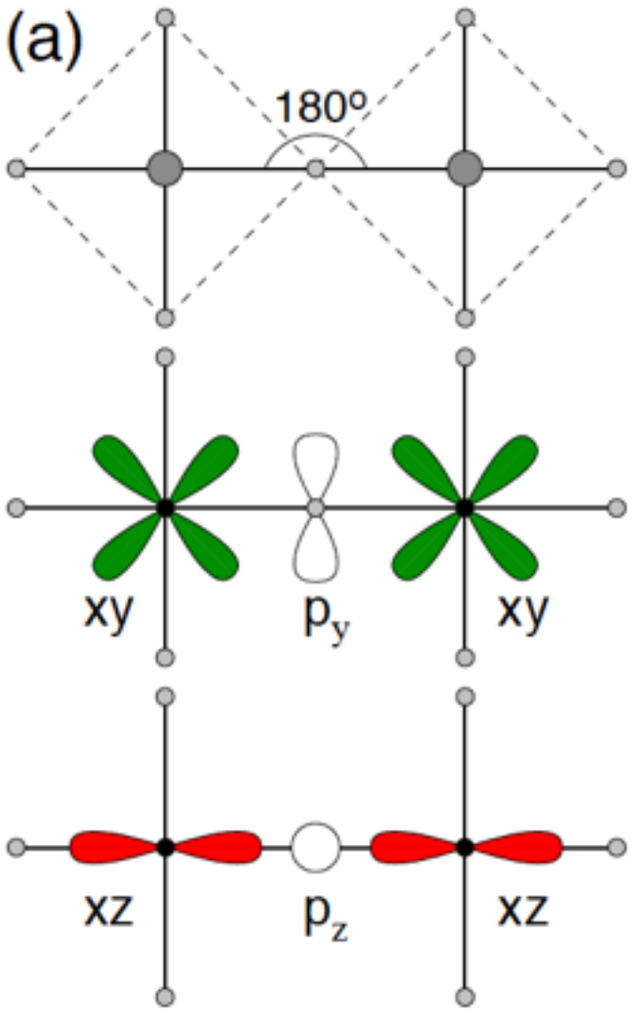
The form of the anisotropy term depends on the lattice.

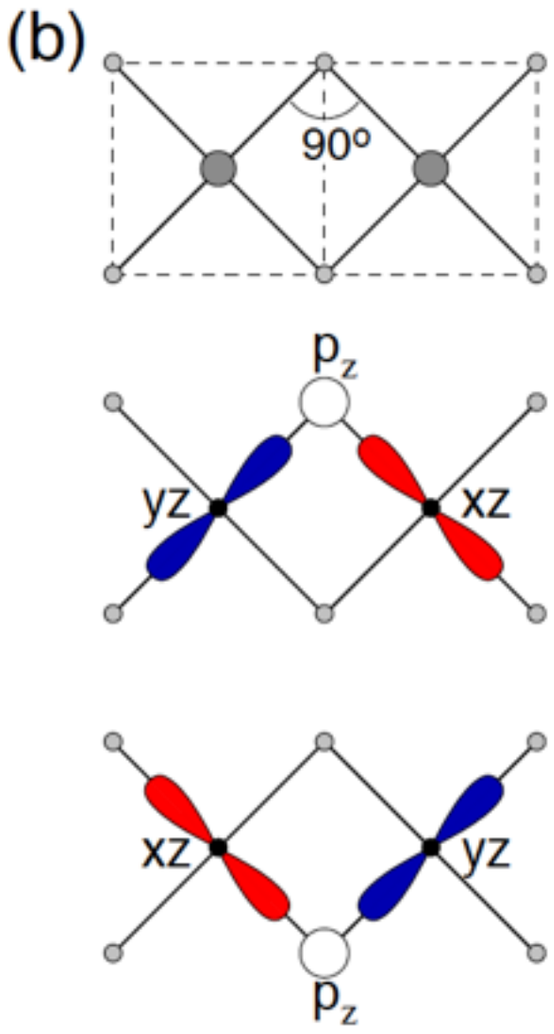
Dipole-dipole interaction

$$J\ (\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j)$$

Kitaev interaction

$$-JS_i^{\gamma} S_j^{\gamma}$$





Sr_2IrO_4

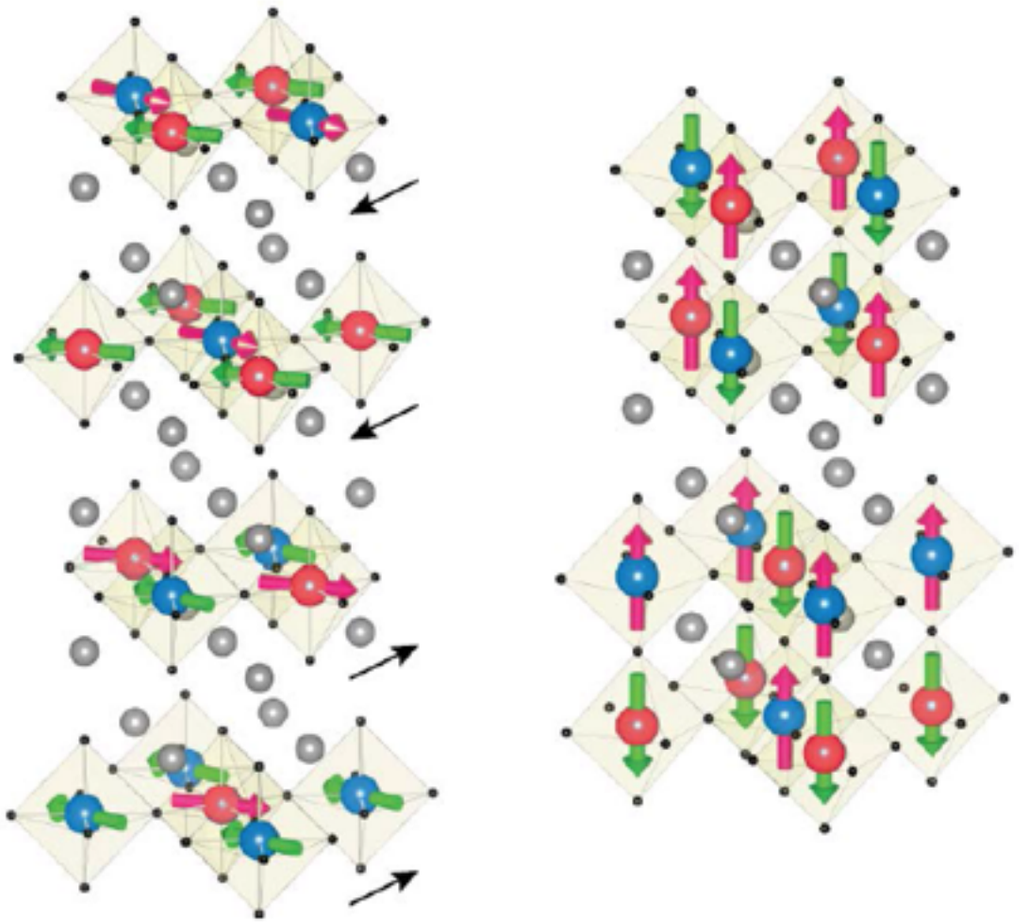
Na_2IrO_3
Li_2IrO_3

G. Jackeli and G. Khaliullin,
PRL 102, 017205 (2009)

"Emulating" High-Tc in iridates

Sr_2IrO_4 (n=1)

same crystal structure as La_2CuO_4



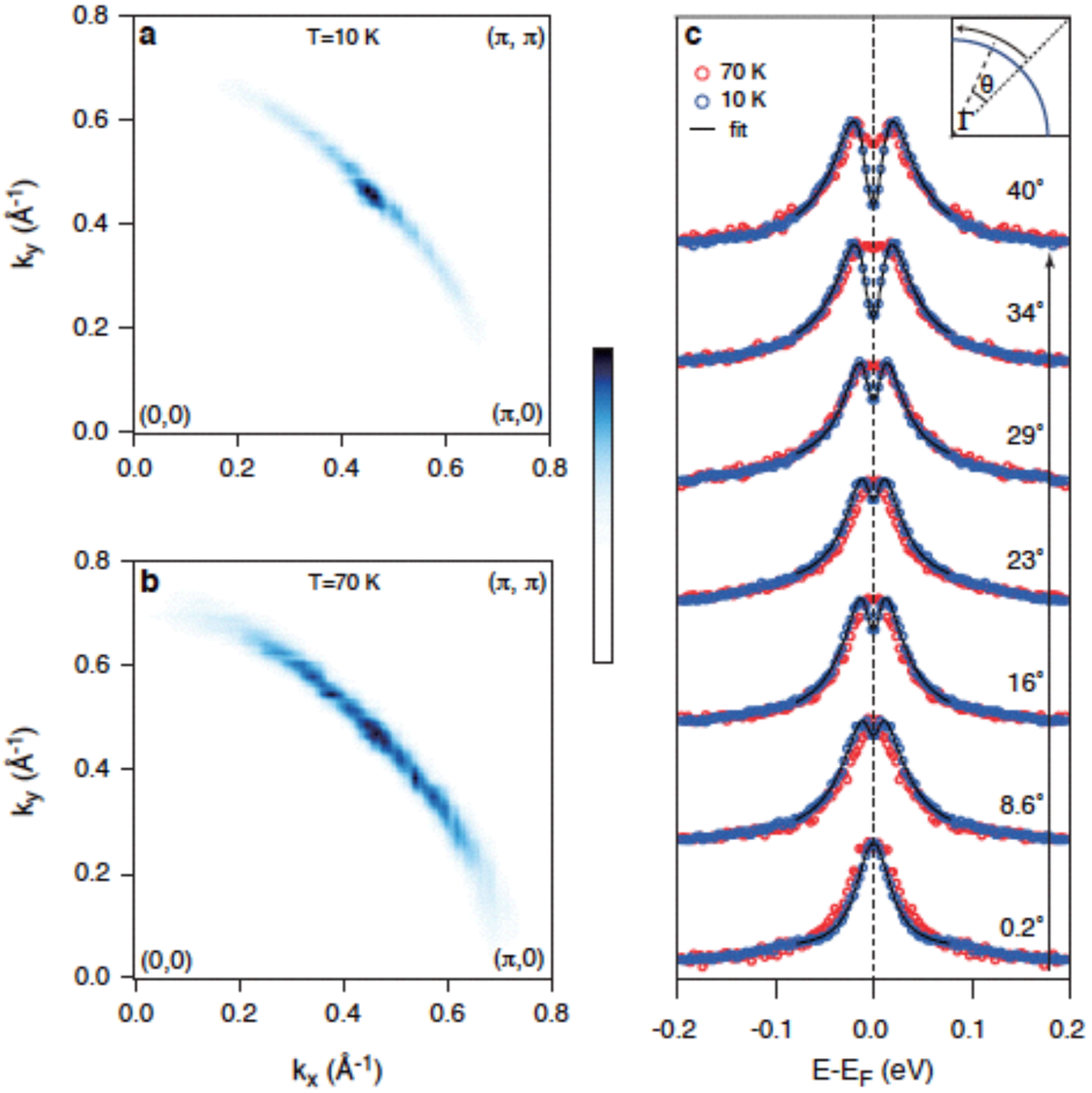
$Sr_3Ir_2O_7$ (n=2)

similar crystal structure as $Bi_2Sr_2CaCu_4O_8$

Any chance for superconductivity?

B.J. Kim group (2008-)

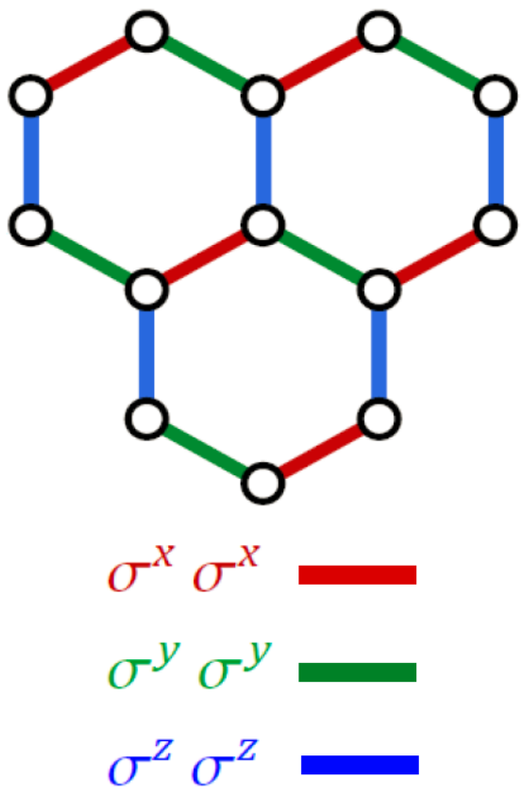
Low-temperature nodal Fermi surface and high-temperature Fermi arcs (ARPES)



B J Kim group 2015

Kitaev model on the honeycomb lattice

$$H = -\sum_{\langle jk \rangle} J_\alpha \, \sigma_j^\alpha \sigma_k^\alpha = -\sum_{\mathbf{r} \in A} \sum_{\alpha = x,y,z} J_\alpha \sigma_{\mathbf{r}}^\alpha \sigma_{\mathbf{r}+\mathbf{d}_\alpha}^\alpha$$



Exactly solvable 2D model

Spin liquid ground state

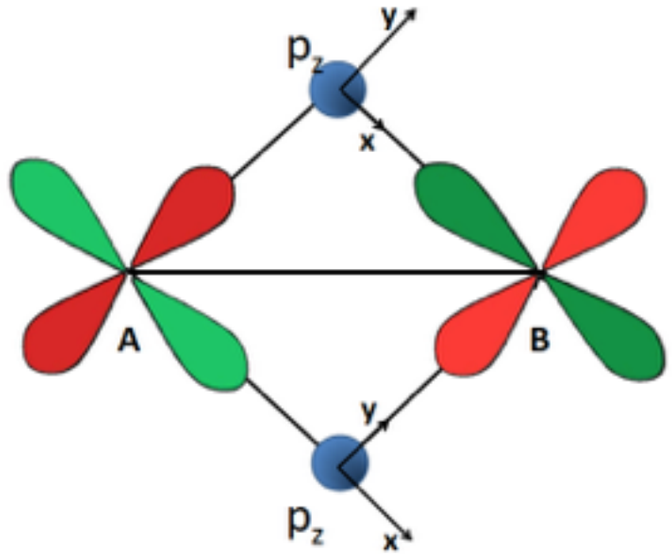
Fractionalized excitations

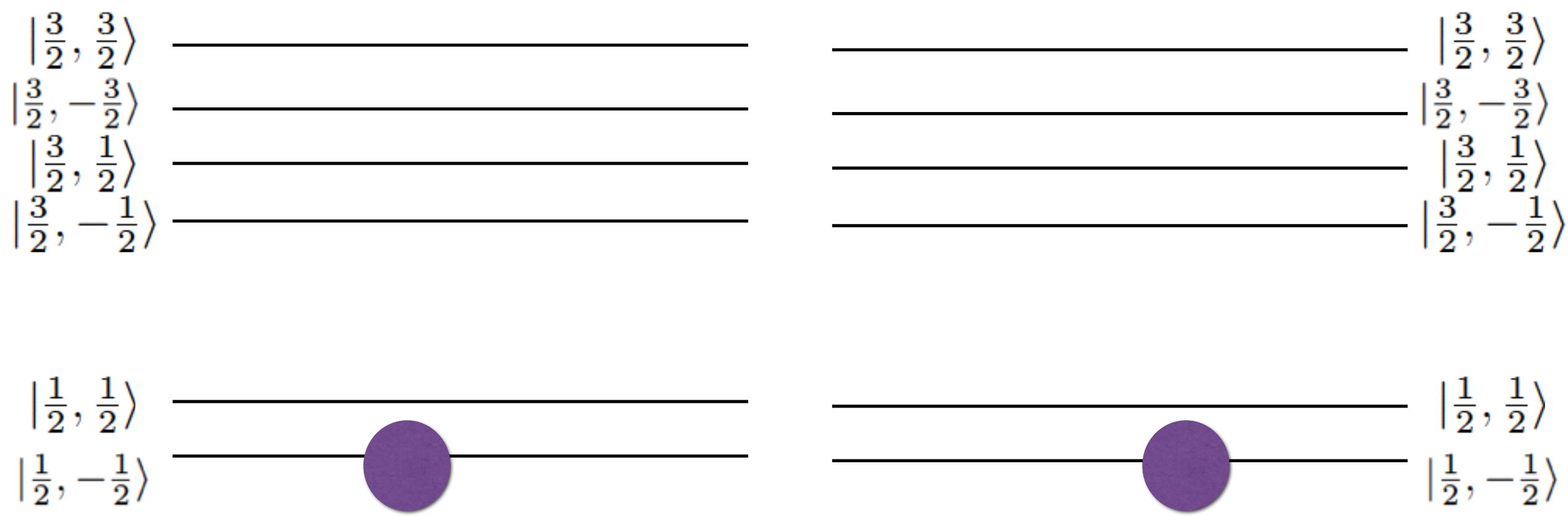
$$\boxed{\sigma_i^\alpha = i b_i^\alpha c_i}$$

$$u_{jk} = i b_j^\alpha b_k^\alpha$$

A. Kitaev, Annals of Physics **321**, 2 (2006)

Let’s derive Kitaev interaction



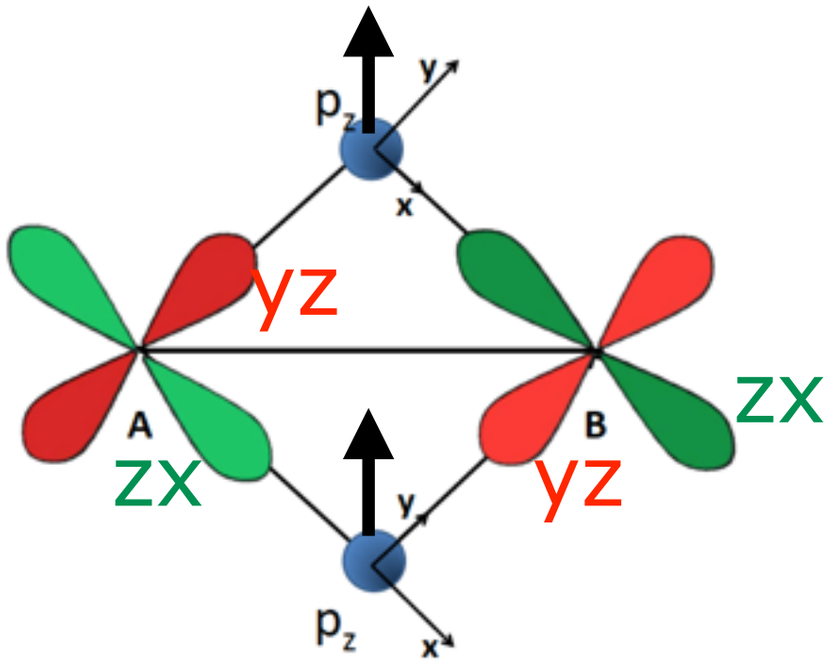


(j,j_z)-representation

$$\begin{pmatrix} \left|\frac{3}{2},\frac{3}{2}\right\rangle \\ \left|\frac{3}{2},\frac{1}{2}\right\rangle \\ \left|\frac{3}{2},\frac{-1}{2}\right\rangle \\ \left|\frac{3}{2},\frac{-3}{2}\right\rangle \\ \left|\frac{1}{2},\frac{1}{2}\right\rangle \\ \left|\frac{1}{2},\frac{-1}{2}\right\rangle \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{2}} & 0 & \frac{i}{\sqrt{2}} & 0 & 0 & 0 \\ 0 & \frac{1}{\sqrt{6}} & 0 & \frac{i}{\sqrt{6}} & \frac{2}{\sqrt{6}} & 0 \\ \frac{1}{\sqrt{6}} & 0 & \frac{-i}{\sqrt{6}} & 0 & 0 & \frac{2}{\sqrt{6}} \\ 0 & \frac{1}{\sqrt{2}} & 0 & \frac{-i}{\sqrt{2}} & 0 & 0 \\ 0 & \frac{1}{\sqrt{3}} & 0 & \frac{i}{\sqrt{3}} & \frac{-1}{\sqrt{3}} & 0 \\ \frac{-1}{\sqrt{3}} & 0 & \frac{i}{\sqrt{3}} & 0 & 0 & \frac{1}{\sqrt{3}} \end{pmatrix} \cdot \begin{pmatrix} |yz,\uparrow\rangle \\ |yz,\downarrow\rangle \\ |zx,\uparrow\rangle \\ |zx,\downarrow\rangle \\ |xy,\uparrow\rangle \\ |xy,\downarrow\rangle \end{pmatrix}$$

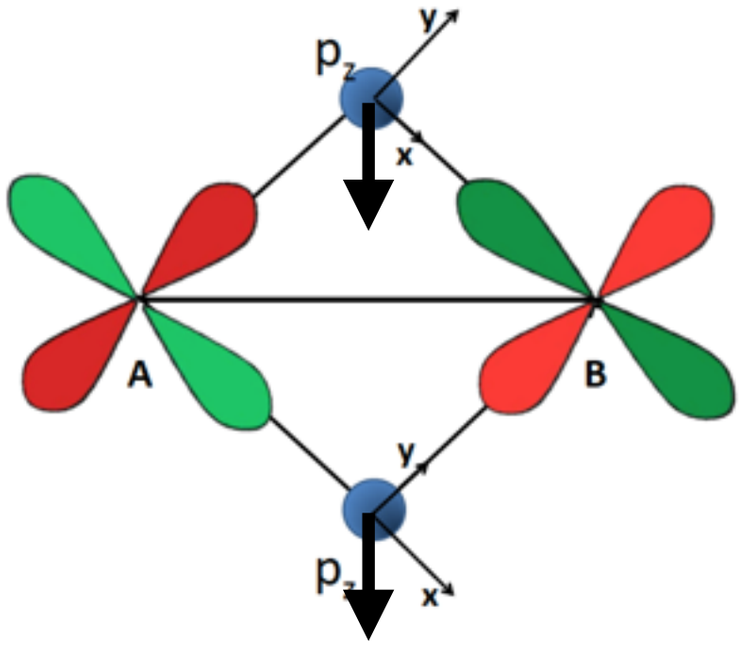
$$\mathrm{J}_{eff\frac{1}{2}} = |j,j_z\rangle = \left|\tfrac{1}{2},\pm\tfrac{1}{2}\right\rangle = \tfrac{1}{\sqrt{3}}(\left|xy,\pm\tfrac{1}{2}\right\rangle \pm \left|yz,\mp\tfrac{1}{2}\right\rangle + i\left|zx,\mp\tfrac{1}{2}\right\rangle)$$

$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,3/2\rangle = \frac{1}{\sqrt{2}}t$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,-3/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,-1/2\rangle = \frac{1}{\sqrt{6}}t$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|1/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|1/2,-1/2\rangle = -\frac{1}{\sqrt{3}}t$$



$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,3/2\rangle = \frac{i}{\sqrt{2}}t$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,-3/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,-1/2\rangle = -\frac{i}{\sqrt{6}}t$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|1/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|1/2,-1/2\rangle = -\frac{i}{\sqrt{3}}t$$

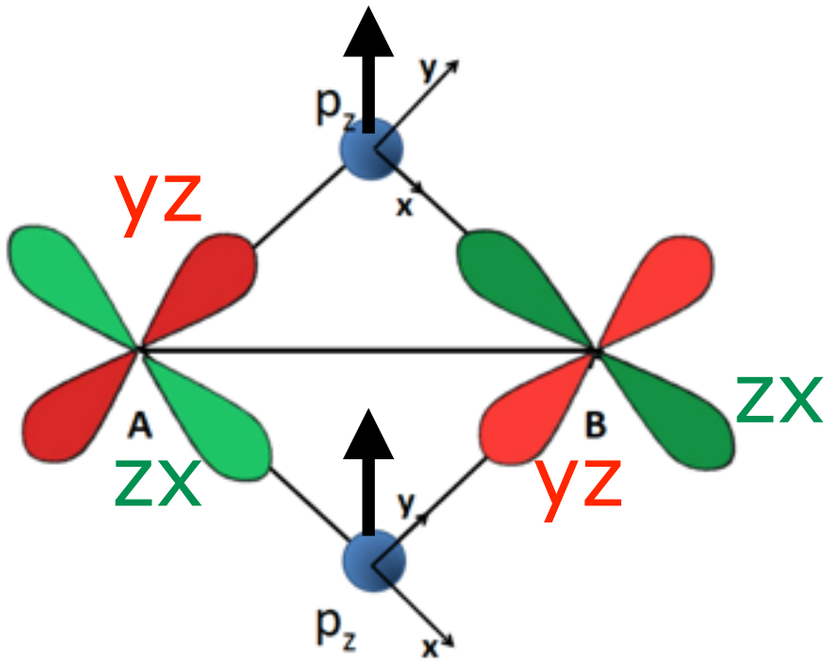
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,3/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,-3/2\rangle = \frac{1}{\sqrt{2}}t$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,1/2\rangle = \frac{1}{\sqrt{6}}t$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,-1/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|1/2,1/2\rangle = \frac{1}{\sqrt{3}}t$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|1/2,-1/2\rangle = 0$$



Upper path 1

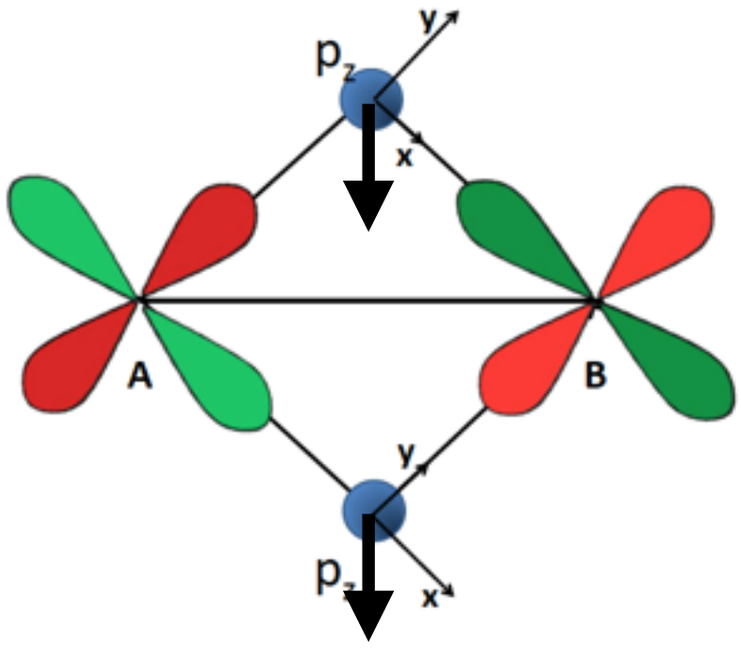
$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,3/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,-3/2\rangle = -\frac{i}{\sqrt{2}}t$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,1/2\rangle = \frac{i}{\sqrt{6}}t$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,-1/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|1/2,1/2\rangle = \frac{i}{\sqrt{3}}t$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|1/2,-1/2\rangle = 0$$

$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,3/2\rangle = \frac{i}{\sqrt{2}}t$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,-3/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|3/2,-1/2\rangle = -\frac{i}{\sqrt{6}}t$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|1/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{zx}|1/2,-1/2\rangle = -\frac{i}{\sqrt{3}}t$$



$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,3/2\rangle = \frac{1}{\sqrt{2}}t$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,-3/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|3/2,-1/2\rangle = \frac{1}{\sqrt{6}}t$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|1/2,1/2\rangle = 0$$
$$\langle p^z_\uparrow|\hat{T}_{yz}|1/2,-1/2\rangle = -\frac{1}{\sqrt{3}}t$$

$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,3/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,-3/2\rangle = -\frac{i}{\sqrt{2}}t$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,1/2\rangle = \frac{i}{\sqrt{6}}t$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|3/2,-1/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|1/2,1/2\rangle = \frac{i}{\sqrt{3}}t$$
$$\langle p^z_\downarrow|\hat{T}_{zx}|1/2,-1/2\rangle = 0$$



Low path 2

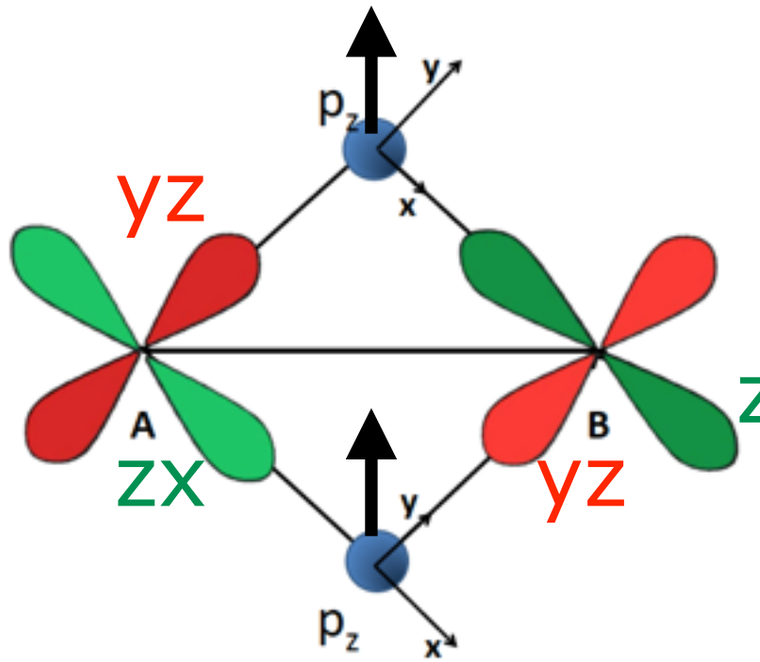
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,3/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,-3/2\rangle = \frac{1}{\sqrt{2}}t$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,1/2\rangle = \frac{1}{\sqrt{6}}t$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|3/2,-1/2\rangle = 0$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|1/2,1/2\rangle = \frac{1}{\sqrt{3}}t$$
$$\langle p^z_\downarrow|\hat{T}_{yz}|1/2,-1/2\rangle = 0$$

Upper path 1

$$\langle \frac{1}{2}, -\frac{1}{2} | \hat{T}_{yz} | p^z_\uparrow \rangle \langle p^z_\uparrow | \hat{T}_{zx} | \frac{1}{2}, -\frac{1}{2} \rangle = -\frac{\imath}{3} t^2$$

Lower path 2

$$\langle \frac{1}{2}, -\frac{1}{2} | \hat{T}_{xz} | p^z_\uparrow \rangle \langle p^z_\uparrow | \hat{T}_{yz} | \frac{1}{2}, -\frac{1}{2} \rangle = \frac{\imath}{3} t^2$$



Upper path 1 + Lower path 2=0

if only states $|\frac{1}{2}, \pm\frac{1}{2}\rangle$ are taken into account

Upper path 1 + Lower path 2

$$\langle \frac{1}{2}, -\frac{1}{2} | \hat{T}_{yz} | p^z_\uparrow \rangle \langle p^z_\uparrow | \hat{T}_{zx} | \frac{3}{2}, \frac{3}{2} \rangle + \langle \frac{1}{2}, -\frac{1}{2} | \hat{T}_{xz} | p^z_\uparrow \rangle \langle p^z_\uparrow | \hat{T}_{yz} | \frac{3}{2}, \frac{3}{2} \rangle = -\frac{2\imath}{\sqrt{6}}$$

same for spin down

Next step is a usual perturbation theory.

For the total exchange one gets:

$$J_K = \frac{(...)}{E_0 - E_{\uparrow\uparrow}} - \frac{(...)}{E_0 - E_{\uparrow\downarrow}} \sim E_{\uparrow\uparrow} - E_{\uparrow\downarrow} \sim J_H$$

This is the essence of the derivation of the Kitaev interaction

A bit more general approach…

Step 1: find single particle states from diagonalization of the single-ion Hamiltonian H=SOC + CF

$$H_{\lambda,\Delta} = \lambda \mathbf{S}\cdot\mathbf{L} + \Delta L^2_{[111]}$$

$$\hat{H} = \begin{pmatrix}
-\lambda & 0 & -\frac{(1-i)\Delta}{3\sqrt{6}} & 0 & \frac{(1+i)\Delta}{3\sqrt{2}} & \frac{i\Delta}{3}\sqrt{\frac{2}{3}} \\
0 & -\lambda & \frac{i\Delta}{3}\sqrt{\frac{2}{3}} & \frac{(1-i)\Delta}{3\sqrt{2}} & 0 & -\frac{(1+i)\Delta}{3\sqrt{6}} \\
-\frac{(1+i)\Delta}{3\sqrt{6}} & -\frac{i\Delta}{3}\sqrt{\frac{2}{3}} & \frac{\lambda}{2} & \frac{(1+i)\Delta}{3\sqrt{3}} & \frac{i\Delta}{3\sqrt{3}} & 0 \\
0 & \frac{(1+i)\Delta}{3\sqrt{2}} & \frac{(1-i)\Delta}{3\sqrt{3}} & \frac{\lambda}{2} & 0 & \frac{i\Delta}{3\sqrt{3}} \\
\frac{(1-i)\Delta}{3\sqrt{2}} & 0 & -\frac{i\Delta}{3\sqrt{3}} & 0 & \frac{\lambda}{2} & -\frac{(1+i)\Delta}{3\sqrt{3}} \\
-\frac{i\Delta}{3}\sqrt{\frac{2}{3}} & -\frac{(1-i)\Delta}{3\sqrt{6}} & 0 & -\frac{i\Delta}{3\sqrt{3}} & -\frac{(1-i)\Delta}{3\sqrt{3}} & \frac{\lambda}{2}
\end{pmatrix}$$

The basis we use is

$$\hat{J} = \{|\tfrac{1}{2},\tfrac{1}{2}\rangle, |\tfrac{1}{2},-\tfrac{1}{2}\rangle, |\tfrac{3}{2},\tfrac{3}{2}\rangle, |\tfrac{3}{2},\tfrac{1}{2}\rangle, |\tfrac{3}{2},-\tfrac{1}{2}\rangle, |\tfrac{3}{2},-\tfrac{3}{2}\rangle\}.$$

Diagonalization of single-ion Hamiltonian leads to three doublets at energies, corresponding to a single-hole states

$$E^{(5,6)} = \frac{\Delta}{3} + \frac{\lambda}{2}$$

$|\Phi_5\rangle$ and $|\Phi_6\rangle$

$$E^{(3,4)} = -\frac{\Delta}{6} - \frac{\lambda}{4} + \frac{1}{2}\sqrt{2\lambda^2 + (\Delta - \frac{\lambda}{2})^2}$$

$|\Phi_3\rangle$ and $|\Phi_4\rangle$

$$E^{(1,2)} = -\frac{\Delta}{6} - \frac{\lambda}{4} - \frac{1}{2}\sqrt{2\lambda^2 + (\Delta - \frac{\lambda}{2})^2}$$

$|\Phi_1\rangle$ and $|\Phi_2\rangle$

Since the Hamiltonian is time-reversal invariant, the ground-state of the single-ion single-hole is a Kramer's doublet, which can be described by a pseudospin-1/2.

Step 2: find two-hole states in the presence of interactions,spin-orbit coupling and crystal field interaction.

There are 15 partly degenerate two-hole states, which can be obtained by diagonalization of the full Hamiltonian:

$$H_{int+\lambda,\Delta} = H_{int} + H_{\lambda,\Delta}$$

$$\begin{aligned} H_{int} &= U_1 \sum_{i,\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + \frac{1}{2}(U_2 - J_H) \sum_{i,\sigma,\alpha\neq\alpha'} n_{i\alpha\sigma} n_{i\alpha'\sigma} \\ &+ U_2 \sum_{i,\alpha\neq\alpha'} n_{i\alpha\uparrow} n_{i\alpha'\downarrow} + J_H \sum_{i,\alpha\neq\alpha'} d^\dagger_{i\alpha\uparrow} d^\dagger_{i\alpha\downarrow} d_{i\alpha'\downarrow} d_{i\alpha'\uparrow} \\ &- J_H \sum_{i,\alpha\neq\alpha'} d^\dagger_{i\alpha\uparrow} d_{i\alpha\downarrow} d^\dagger_{i\alpha'\downarrow} d_{i\alpha'\uparrow} \ , \end{aligned}$$

For the basis we can use the product of single-hole states states:

$$
\begin{aligned}
|\oplus\oplus, 1\rangle &\equiv |\Phi_1\Phi_2\rangle \\
|\oplus\oplus, 2\rangle &\equiv |\Phi_1\Phi_3\rangle \\
|\oplus\oplus, 3\rangle &\equiv |\Phi_1\Phi_4\rangle \\
|\oplus\oplus, 4\rangle &\equiv |\Phi_1\Phi_5\rangle \\
|\oplus\oplus, 5\rangle &\equiv |\Phi_1\Phi_6\rangle \\
|\oplus\oplus, 6\rangle &\equiv |\Phi_2\Phi_3\rangle \\
|\oplus\oplus, 7\rangle &\equiv |\Phi_2\Phi_4\rangle \\
|\oplus\oplus, 8\rangle &\equiv |\Phi_2\Phi_5\rangle \\
|\oplus\oplus, 9\rangle &\equiv |\Phi_2\Phi_6\rangle \\
|\oplus\oplus, 10\rangle &\equiv |\Phi_3\Phi_4\rangle \\
|\oplus\oplus, 11\rangle &\equiv |\Phi_3\Phi_5\rangle \\
|\oplus\oplus, 12\rangle &\equiv |\Phi_3\Phi_6\rangle \\
|\oplus\oplus, 13\rangle &\equiv |\Phi_4\Phi_5\rangle \\
|\oplus\oplus, 14\rangle &\equiv |\Phi_4\Phi_6\rangle \\
|\oplus\oplus, 15\rangle &\equiv |\Phi_5\Phi_6\rangle
\end{aligned}
$$

diagonalization

$$
|D, \xi\rangle = \sum_{\mu=1}^{15} c_{\xi\mu} |\oplus\oplus, \mu\rangle
$$

Step 3: strong coupling approach. For nearest neighbor coupling, it is a second order perturbation theory expansion in the effective hopping parameters .

$$H_{ex,n,n'} = \sum_{\xi} \frac{1}{\epsilon_\xi} P H_{t,n,n'} Q_{\xi,n'} H_{t,n',n} P$$

$$P = \sum_{\sigma_n = \pm 1} |1/2, \sigma_n/2; n\rangle\langle n; 1/2, \sigma_n/2|$$

$$Q_{\xi,n'} = |D, \xi; n'\rangle\langle n'; D, \xi| = D^\dagger_{\xi,n'} D_{\xi,n'}$$

The projection operators onto two-hole intermediate states

The projection operators onto one-hole ground states

Step 4: write the super-exchange Hamiltonian can be written in terms of the magnetic degrees of freedom

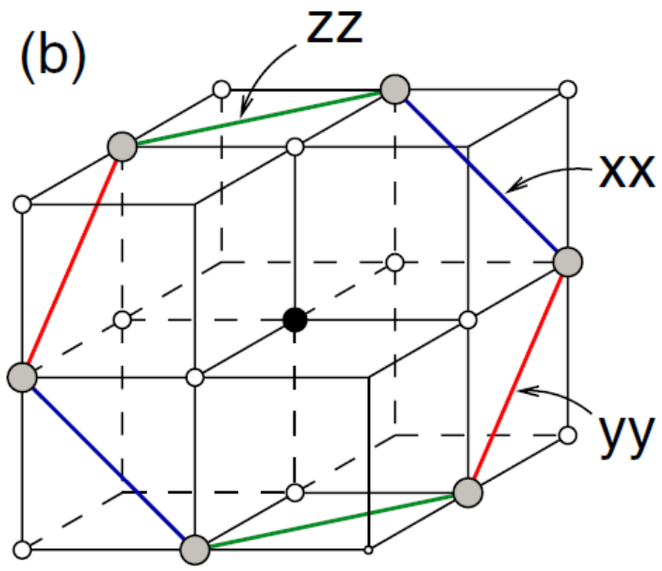
$$H_{\mathrm{ex},n,n'} = \sum_{\alpha\beta} \Xi^{\alpha\beta}_{n,n'} S^{\alpha}_{n} S^{\beta}_{n'}$$

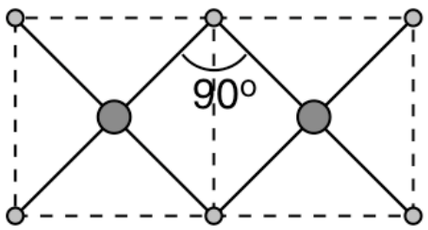
$$\Xi_{n,n'} = \begin{pmatrix} J^{x} & J^{xy} & J^{xz} \\ J^{yx} & J^{y} & J^{yz} \\ J^{zx} & J^{zy} & J^{z} \end{pmatrix}$$

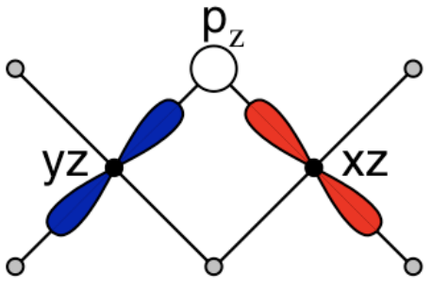
Goal achieved:

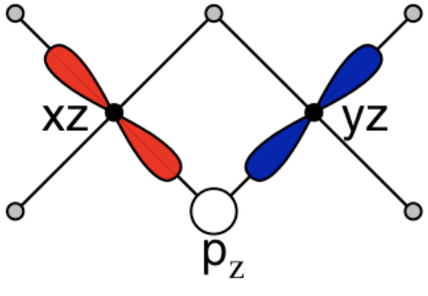
we find how the exchange coupling tensor depends on the microscopic parameters - CF distortion, Hund's coupling, Coulomb interaction and SO coupling.

Super-exchange in A_2IrO_3



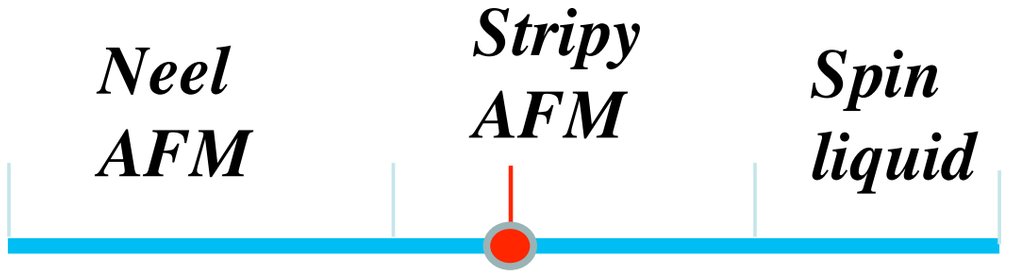


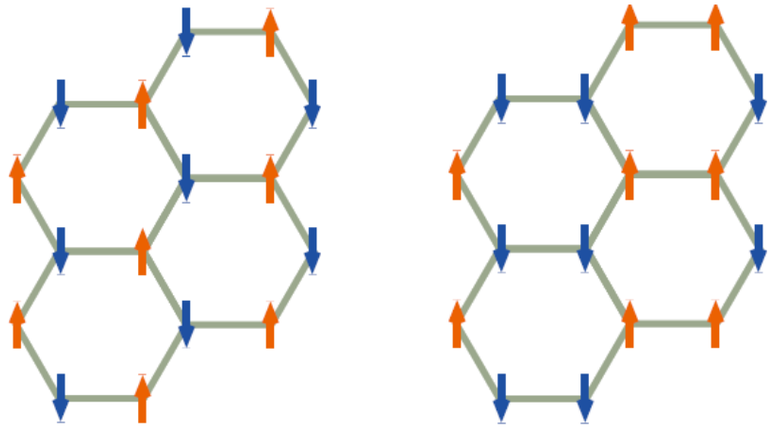




G. Jackeli and G. Khaliullin, PRL 102, 017205 (2009)

$$\mathcal{H} = -J_K \sum_{\langle ij \rangle_a} \hat{\sigma}_i^a \hat{\sigma}_j^a + J_H \sum_{\langle ij \rangle} \hat{\boldsymbol{\sigma}}_i \cdot \hat{\boldsymbol{\sigma}}_j$$

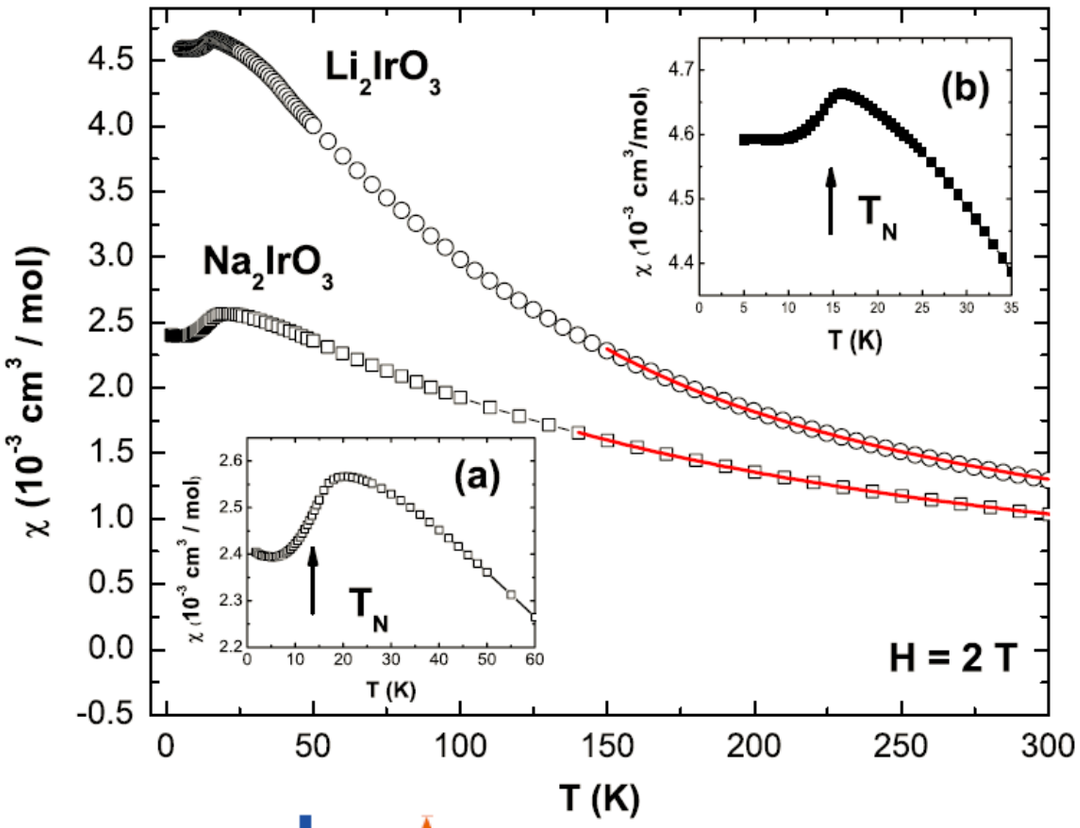


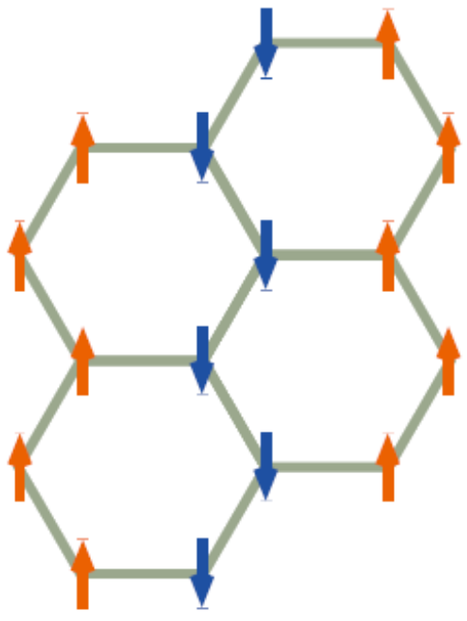


Kitaev spin liquid is stable against Heisenberg perturbations!

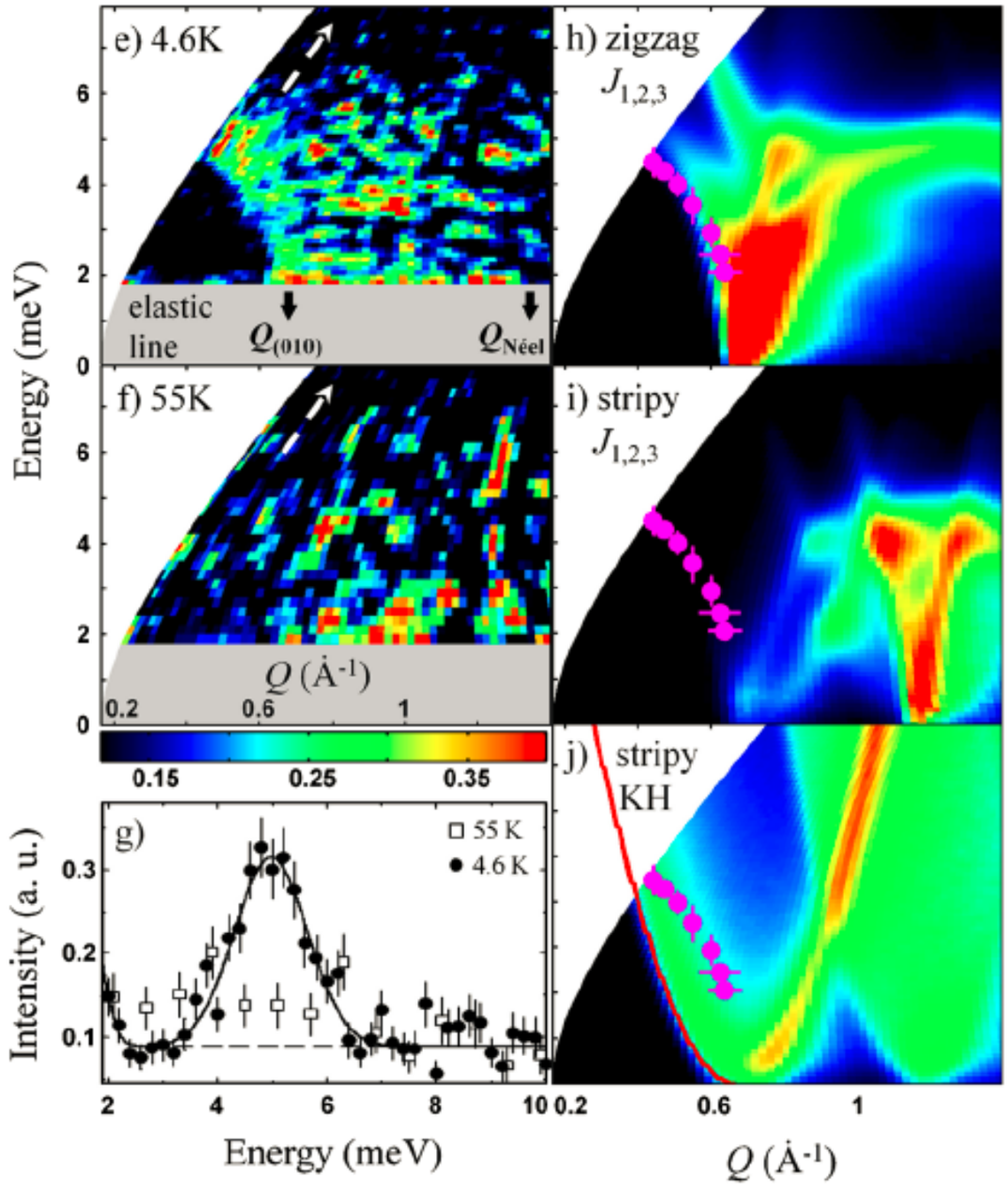
Na_2IrO_3 orders in AFM zigzag structure

Singh and Gegenwart, PRB 82, 064412 (2010);
Singh et al, PRL 108, 127203 (2012)



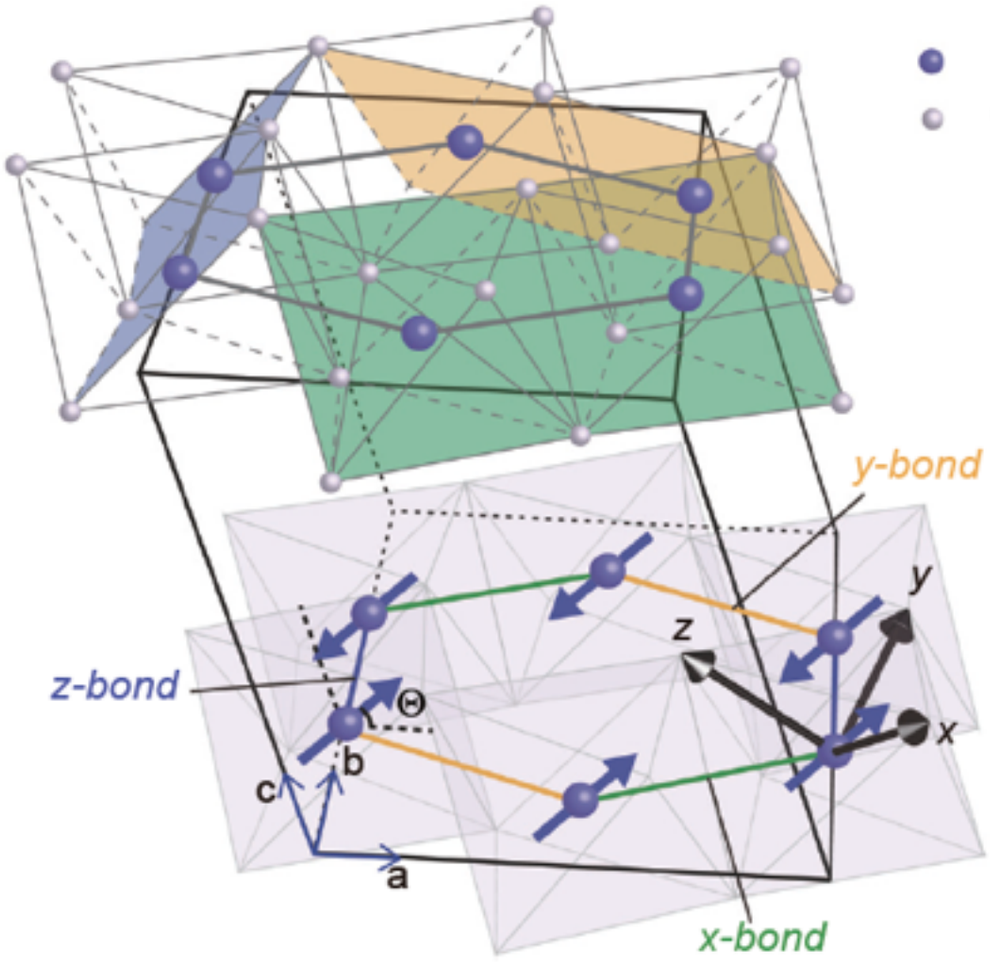


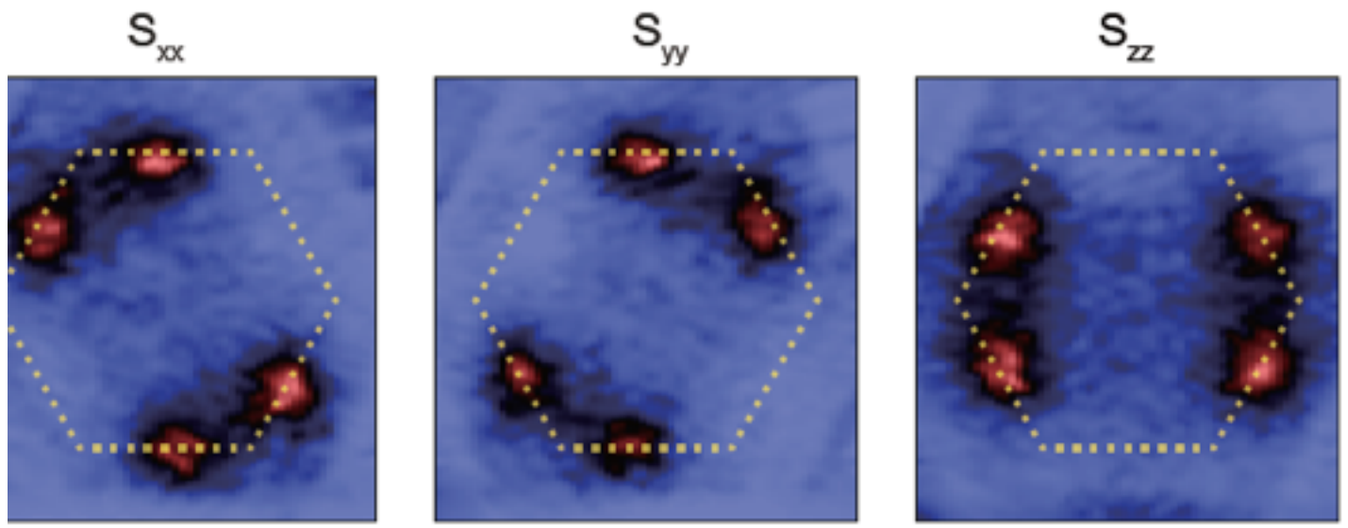
X. Liu et al, PRB 2011
Feng Ye et al, PRB 2012



S. K. Choi et al PRL 2012

Locking of the spin direction to the spatial orientation of the zigzag in Na_2IrO_3



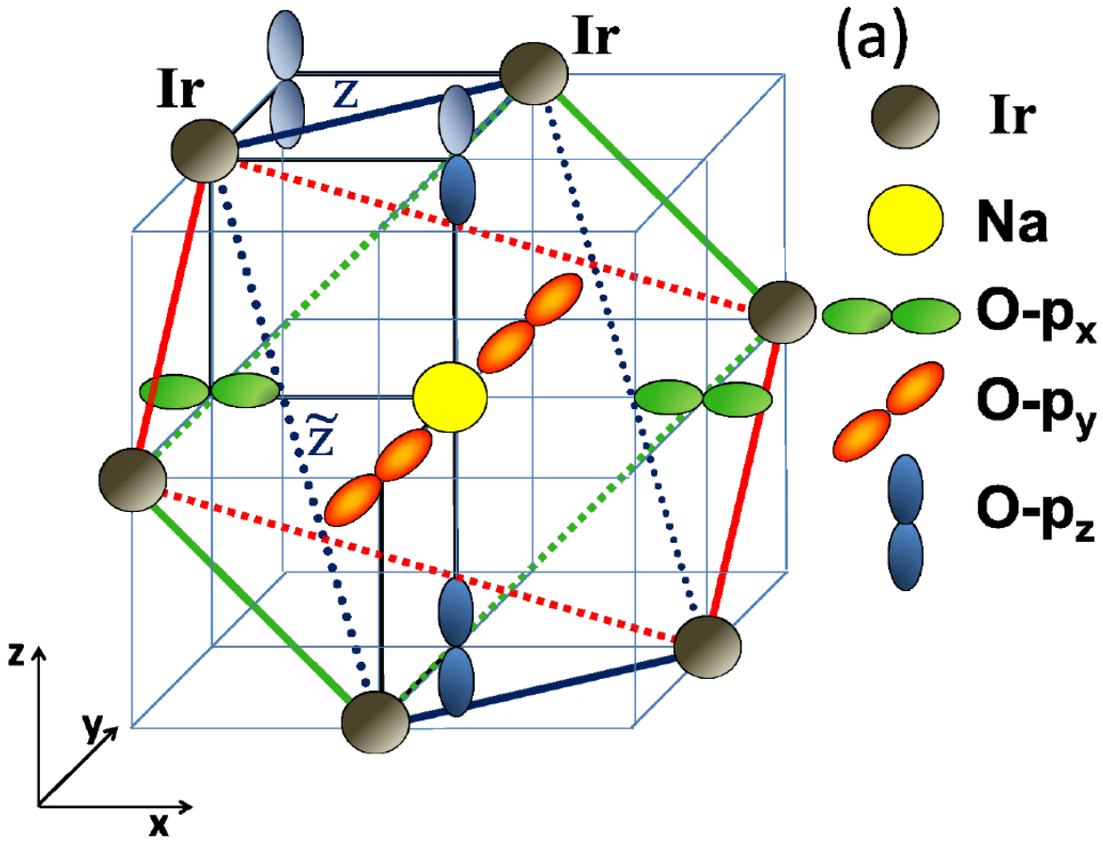


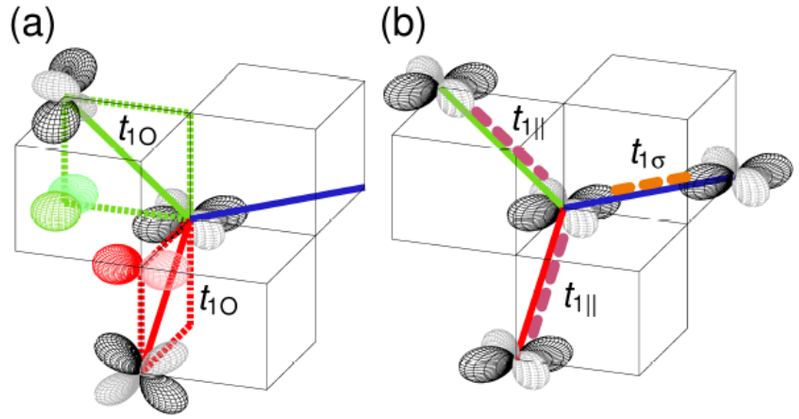
Spin-component-resolved equal-time correlations

Diffuse magnetic x-ray scattering

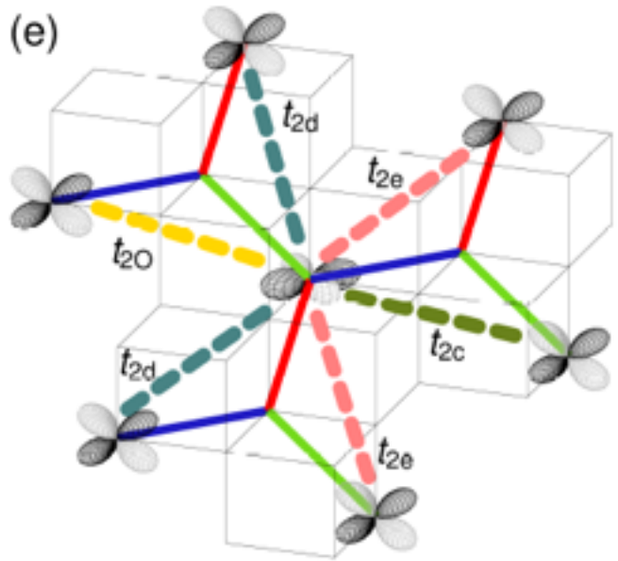
S.H.Chun et al, Nature Physics 2015

Revision of the super-exchange model for Na_2IrO_3





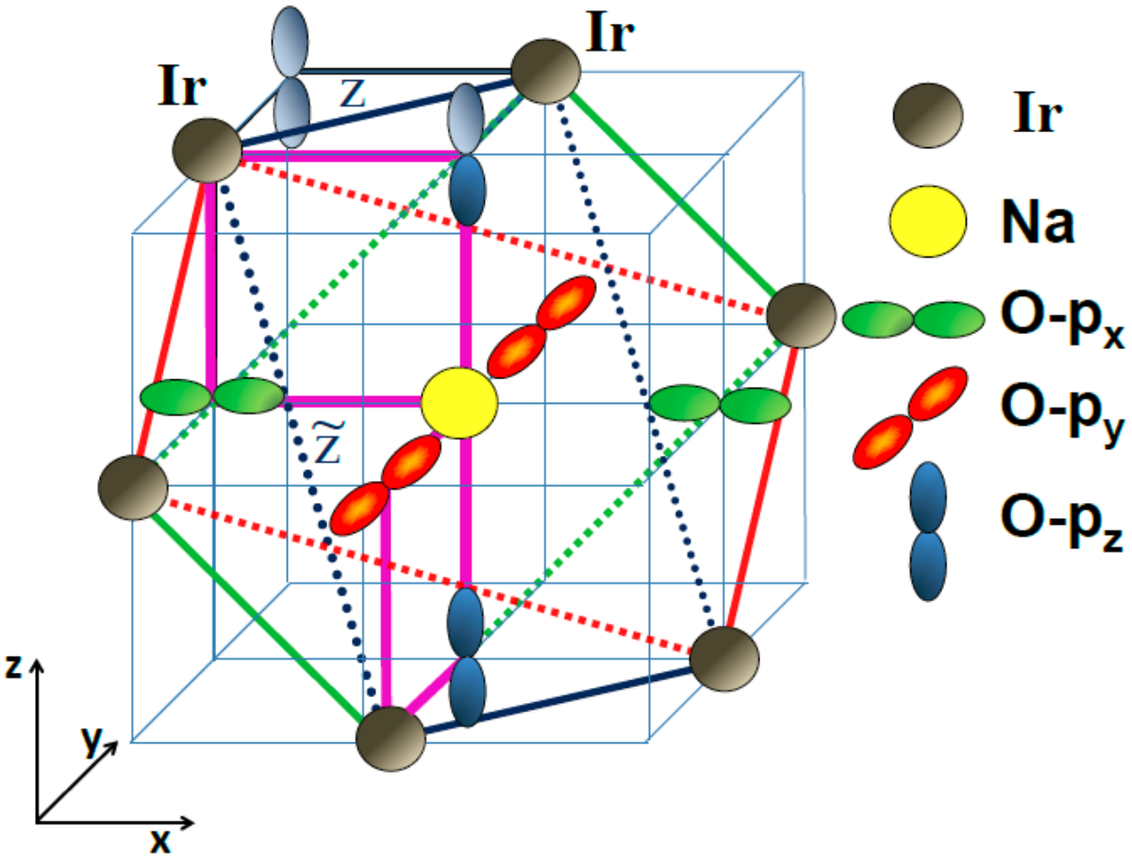
$$t_{1o} = 230 \text{ meV}$$
$$t_{1\sigma} = 67 \text{ meV}$$

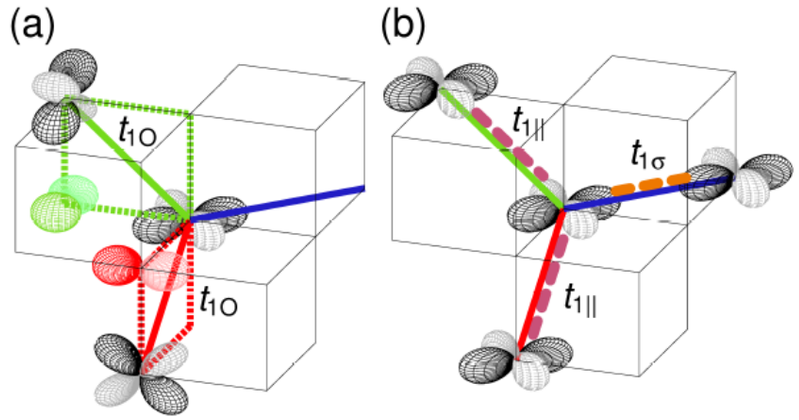


$$t_{2o} = 94.7 \text{ meV}$$

Kateryna Foyevtsova *et al*, PRB 2013

Revision of the super-exchange model for Na_2IrO_3





$$t_{1o} = 230 \text{ meV}$$
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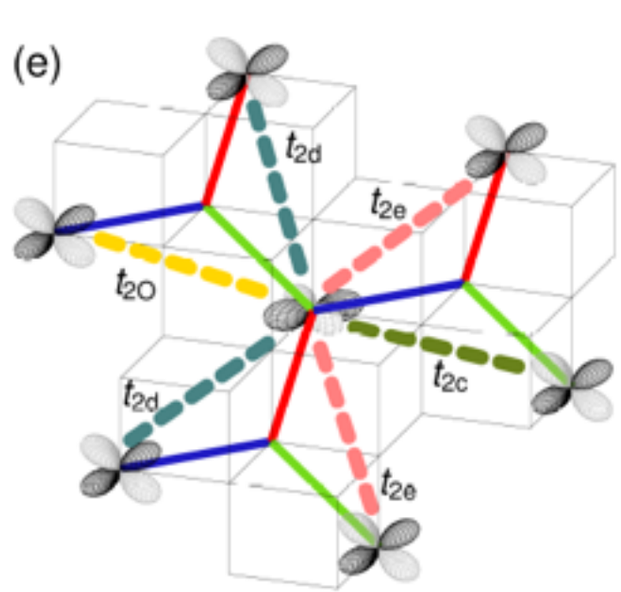


$$t_{2o} = 94.7 \text{ meV}$$

Kateryna Foyevtsova *et al*, PRB 2013

Second neighbors hopping

Path 1 : $\text{Ir}\,(Y) \rightarrow \text{O}\,(p_z) \rightarrow \text{Na}\,(s) \rightarrow \text{O}\,(p_z) \rightarrow \text{Ir}\,(X)$
Path 2 : $\text{Ir}\,(Y) \rightarrow \text{O}\,(p_z) \rightarrow \text{Na}\,(s)$
Path 3 : $\text{Ir}\,(Y) \rightarrow \text{O}\,(p_x) \rightarrow$
Path 4 : $\text{Ir}\,(Y) \rightarrow \text{O}\,(p_x) \rightarrow \text{Na}\,(s)$

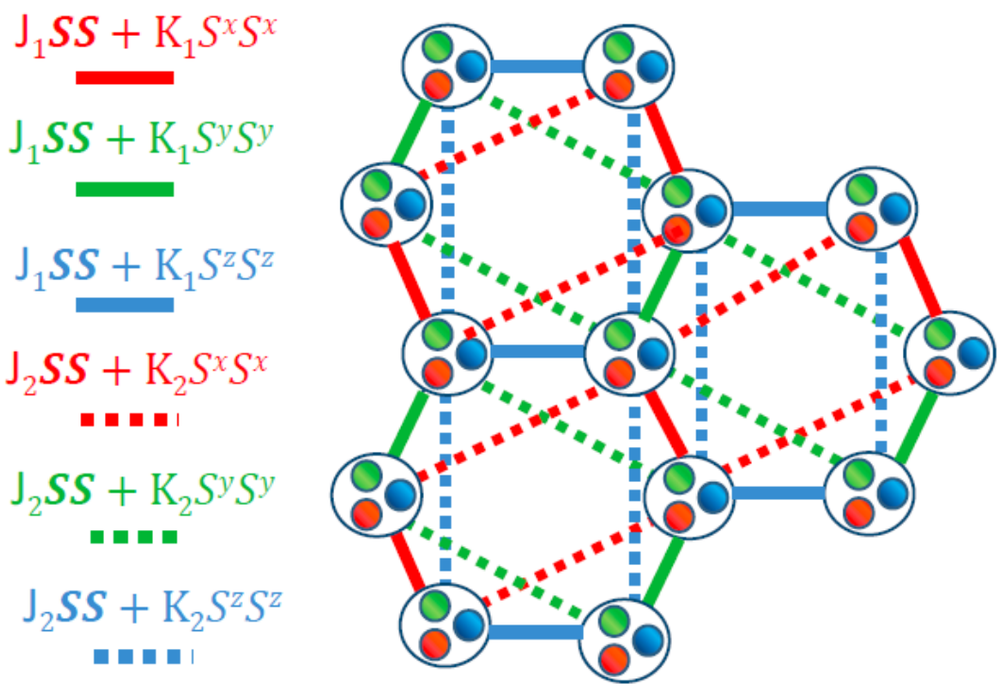
Kitaev interaction

$|X\rangle = |yz\rangle$, $|Y\rangle = |zx\rangle$ and $|Z\rangle = |xy\rangle$

J_1-K_1-J_2-K_2 model

$$\mathcal{H} = J_1 \sum_{\langle n,n' \rangle_\gamma} \mathbf{S}_n \mathbf{S}_{n'} + K_1 \sum_{\langle n,n' \rangle_\gamma} S_n^\gamma S_{n'}^\gamma$$

$$+ J_2 \sum_{\langle\langle n,n' \rangle\rangle_{\tilde{\gamma}}} \mathbf{S}_n \mathbf{S}_{n'} + K_2 \sum_{\langle\langle n,n' \rangle\rangle_{\tilde{\gamma}}} S_n^\gamma S_{n'}^{\tilde{\gamma}}$$



Na_2IrO_3

$$\Delta = 0.1 \text{ eV}, \lambda = 0.4 \text{ eV}$$
$$J_H = 0.3 \text{ eV}, U_2 = 1.8 \text{ eV}$$
$$t_{1o} = 230 \text{ meV}, t_d = 67 \text{ meV}$$
$$t_{2o} = 95 \text{ meV}$$

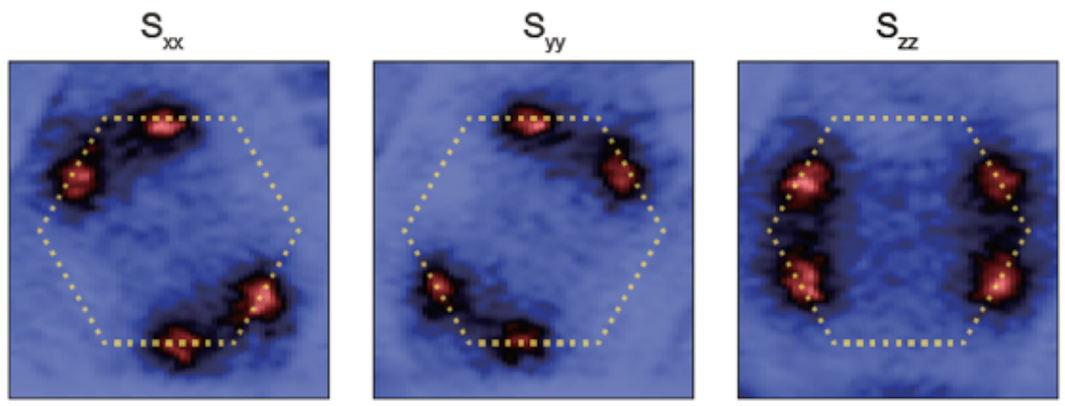
$$J_1 = 5.1\,\text{meV},\ K_1 = -14.8\,\text{meV}$$
$$J_2 = -4.5\,\text{meV},\ K_2 = 9\,\text{meV}$$

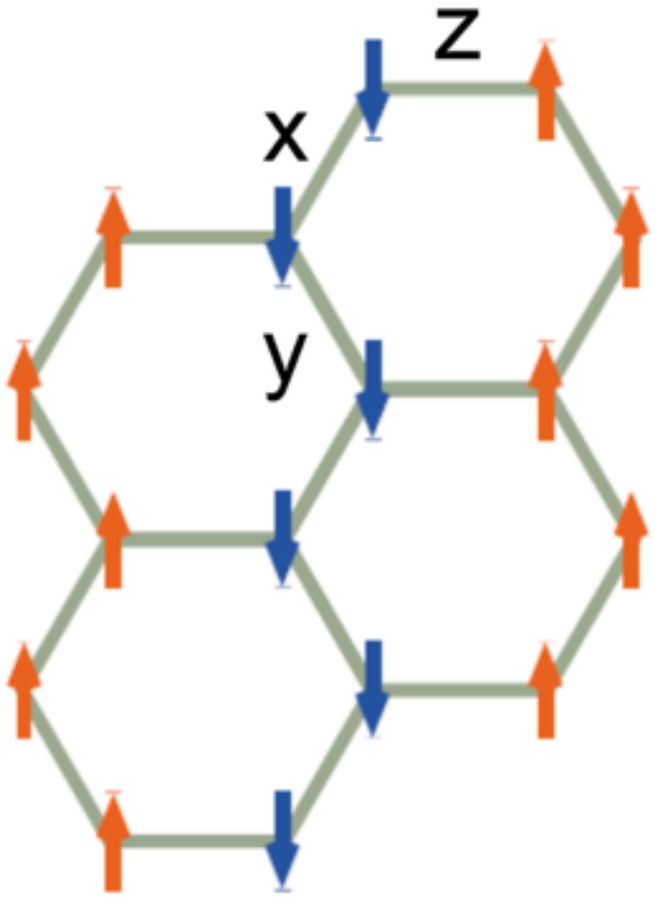
zigzag

$\theta_{CW} \approx$-98 K

Y.Sizyuk, C.Price, P. Wölfle, **N.B.P.**, PRB 2014

Locking of the spin direction to the spatial orientation of the zigzag in Na_2IrO_3



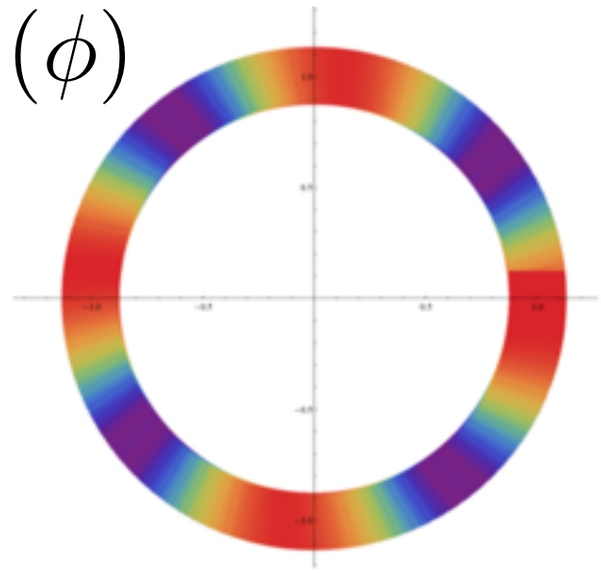


$$S = S_0 + S_{\rm fl}(\theta, \phi)$$

$$S_{\rm fl} = -\sum_{\boldsymbol{q}} \sum_{\kappa,\kappa'=A,B} \sum_{\mu,\mu'=0}^{2} \tilde{A}_{\boldsymbol{q}}^{\kappa\mu;\kappa'\mu'} \delta\phi_{-\boldsymbol{q}}^{\kappa\mu} \delta\phi_{\boldsymbol{q}}^{\kappa'\mu'}$$

$$\tilde{A}_{\boldsymbol{q}} = \begin{pmatrix} c_{11} & c_{12} & c_{13} & 1/(J_{\boldsymbol{q}}^x)^* & 0 & 0 \\ c_{12} & c_{22} & c_{23} & 0 & 1/(J_{\boldsymbol{q}}^y)^* & 0 \\ c_{13} & c_{23} & c_{33} & 0 & 0 & 1/(J_{\boldsymbol{q}}^z)^* \\ 1/J_{\boldsymbol{q}}^x & 0 & 0 & c_{11} & c_{12} & c_{13} \\ 0 & 1/J_{\boldsymbol{q}}^y & 0 & c_{12} & c_{22} & c_{23} \\ 0 & 0 & 1/J_{\boldsymbol{q}}^z & c_{13} & c_{23} & c_{33} \end{pmatrix}$$

$S_{\rm fl}(\phi)$



spin fluctuations select one of the diagonals in xy-plane

Y.Sizyuk, N.B.P, P. Wölfle 2015

Importance of Kitaev interactions for Na_2IrO_3

— the dominant interaction is nearest neighbor FM K_1 interaction;

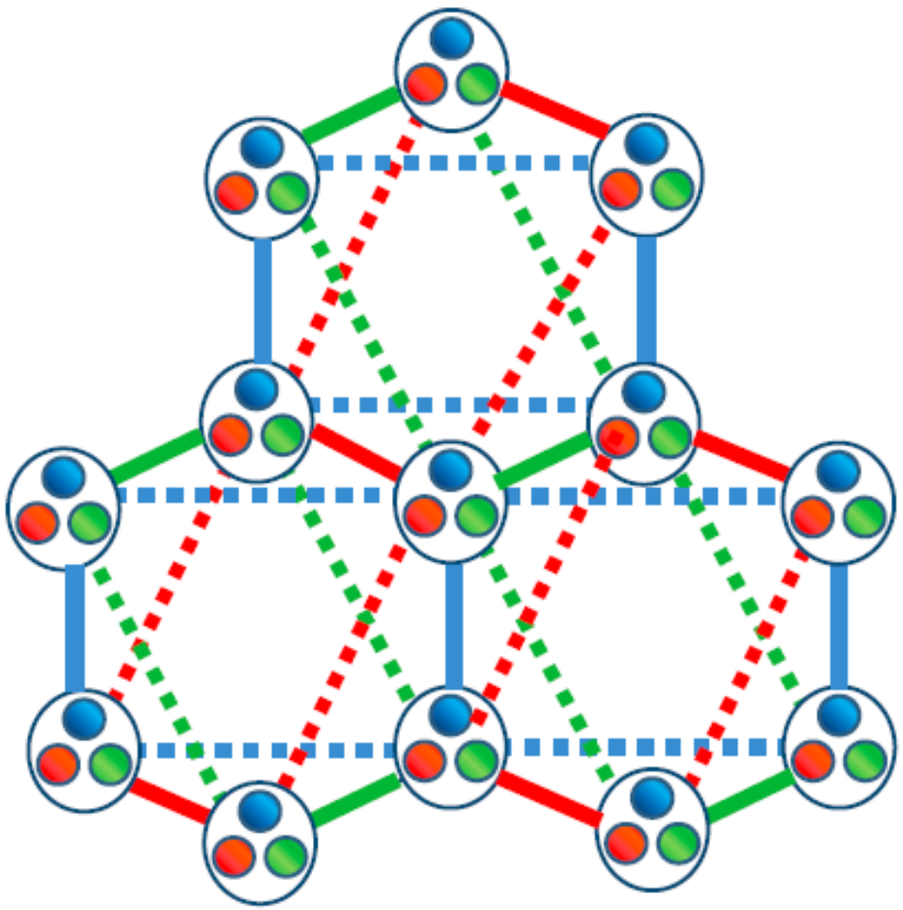
—AFM second neighbor K_2 coupling is the largest energy scale K_1;

— interplay of K_1 and K_2 stabilize the zigzag AFM state;

— K_2 provides a natural basis to account for the large and AFM Curie-Weiss temperature and also explains correct orientation of the spatial spin direction.

K_1-K_2 model on the honeycomb lattice

$$\mathcal{H} = K_1 \sum_{\langle ij \rangle} S_i^{\gamma_{ij}} S_j^{\gamma_{ij}} + K_2 \sum_{\ll ij \gg} S_i^{\lambda_{ij}} S_j^{\lambda_{ij}}$$



Talk by Ioannis Rousochatzakis on Saturday

More details in

PRL 109, 187201 (2012) PHYSICAL REVIEW LETTERS week ending 2 NOVEMBER 2012

Critical Properties of the Kitaev-Heisenberg Model

Craig C. Price and Natalia B. Perkins
Department of Physics, University of Wisconsin, 1150 University Avenue, Madison, Wisconsin 53706, USA
(Received 4 June 2012; published 2 November 2012)

We study the critical properties of the Kitaev-Heisenberg model on the honeycomb lattice at finite temperatures that might describe the physics of the quasi-two-dimensional compounds, Na_2IrO_3 and Li_2IrO_3. The model undergoes two phase transitions as a function of temperature. At low temperature, thermal fluctuations induce magnetic long-range order by the order-by-disorder mechanism. This magnetically ordered state with a spontaneously broken Z_6 symmetry persists up to a certain critical temperature. We find that there is an intermediate phase between the low-temperature, ordered phase and the high-temperature, disordered phase. Finite-sized scaling analysis suggests that the intermediate phase is a critical Kosterlitz-Thouless phase with continuously variable exponents. We argue that the intermediate phase has been observed above the low-temperature, magnetically ordered phase in Na_2IrO_3, and also, likely exists in Li_2IrO_3.

PHYSICAL REVIEW B 88, 024410 (2013)

Finite-temperature phase diagram of the classical Kitaev-Heisenberg model

Craig Price[1] and Natalia B. Perkins[2]
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[2]*Department of Physics, University of Wisconsin, 1150 University Avenue, Madison, Wisconsin 53706, USA*
(Received 2 May 2013; revised manuscript received 25 June 2013; published 15 July 2013)

We investigate the finite-temperature phase diagram of the classical Kitaev-Heisenberg model on the hexagonal lattice. Due to the anisotropy introduced by the Kitaev interaction, the model is magnetically ordered at low temperatures. The ordered phase is stabilized entropically by an order-by-disorder mechanism where thermal fluctuations of classical spins select collinear magnetic states in which magnetic moments point along one of the cubic directions. We find that there is an intermediate phase between the low-temperature ordered phase and the high-temperature disordered phase. We show that the intermediate phase is a critical Kosterlitz-Thouless phase exhibiting correlations of the order parameter that decay algebraically in space. Using finite-size-scaling analysis, we determine the boundaries of the critical phase with reasonable accuracy. We show that the Kitaev interaction plays a crucial role in understanding the finite-temperature properties of A_2IrO_3 systems.

PHYSICAL REVIEW B 90, 155126 (2014)

Importance of anisotropic exchange interactions in honeycomb iridates: Minimal model for zigzag antiferromagnetic order in Na_2IrO_3

Yuriy Sizyuk,[1,2] Craig Price,[3] Peter Wölfle,[1,4] and Natalia B. Perkins[1,2]
[1]*Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA*
[2]*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55116, USA*
[3]*Department of Physics, The Pennsylvania State University, 104 Davey Lab, University Park, Pennsylvania 16802, USA*
[4]*Institute for Condensed Matter Theory and Institute for Nanotechnology, Karlsruhe Institute of Technology, D-76128 Karlsruhe, Germany*
(Received 15 August 2014; revised manuscript received 2 October 2014; published 22 October 2014)

In this work, we investigate the microscopic nature of the magnetism in honeycomb iridium-based systems by performing a systematic study of how the effective magnetic interactions in these compounds depend on various electronic microscopic parameters. We show that the minimal model describing the magnetism in A_2IrO_3 includes both isotropic and anisotropic Kitaev-type spin-exchange interactions between nearest and next-nearest neighbor Ir ions, and that the magnitude of the Kitaev interaction between next-nearest neighbor Ir magnetic moments is comparable with nearest neighbor interactions. We also find that, while the Heisenberg and the Kitaev interactions between nearest neighbors are correspondingly antiferro- and ferromagnetic, they both change sign for the next-nearest neighbors. Using classical Monte Carlo simulations we examine the magnetic phase diagram of the derived super-exchange model. We find that the zigzag-type antiferromagnetic order occupies a large part of the phase diagram of this model and, for the ferromagnetic next-nearest neighbor Heisenberg interaction relevant for Na_2IrO_3, it can be stabilized at small and even at zero third nearest neighbor coupling. Our results suggest that a natural physical origin of the zigzag phase experimentally observed in Na_2IrO_3 is due to the interplay of the Kitaev anisotropic interactions between nearest and next-nearest neighbors.

PRL 113, 187201 (2014) PHYSICAL REVIEW LETTERS week ending 31 OCTOBER 2014

Raman Scattering Signatures of Kitaev Spin Liquids in A_2IrO_3 Iridates with A = Na or Li

J. Knolle,[1] Gia-Wei Chern,[2] D. L. Kovrizhin,[3] R. Moessner,[1] and N. B. Perkins[4,5]
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[2]*Center for Nonlinear Studies and Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[3]*T.C.M. Group, Cavendish Laboratory, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom, and RRC Kurchatov Institute, 1 Kurchatov Square, Moscow 123182, Russia*
[4]*Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA*
[5]*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55116, USA*
(Received 16 June 2014; published 28 October 2014)

We show how Raman spectroscopy can serve as a valuable tool for diagnosing quantum spin liquids (QSL). We find that the Raman response of the gapless QSL of the Kitaev-Heisenberg model exhibits signatures of spin fractionalization into Majorana fermions, which give rise to a broad signal reflecting their density of states, and Z_2 gauge fluxes, which also contribute a sharp feature. We discuss the current experimental situation and explore more generally the effect of breaking the integrability on response functions of Kitaev spin liquids.

PHYSICAL REVIEW B 89, 035143 (2014)

Interplay of many-body and single-particle interactions in iridates and rhodates

Natalia B. Perkins,[1] Yuriy Sizyuk,[1] and Peter Wölfle[1,2]
[1]*Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA*
[2]*Institute for Condensed Matter Theory and Institute for Nanotechnology, Karlsruhe Institute of Technology, D-76128 Karlsruhe, G*
(Received 4 November 2013; revised manuscript received 18 December 2013; published 29 January 2014)

Motivated by recent experiments exploring the spin-orbit-coupled magnetism in 4*d*- and 5*d*-band transition metal oxides, we study magnetic interactions in Ir- and Rh-based compounds. In these systems, the comparable strength of spin-orbit coupling, crystal-field splitting, and Coulomb and Hund's coupling leads to a rich variety of magnetic exchange interactions, leading to new types of ground states. Using a strong coupling approach, we derive effective low-energy superexchange Hamiltonians from the multiorbital Hubbard model by taking full account of the Coulomb and Hund's interactions in the intermediate states. We find that in the presence of strong SOC and lattice distortions the superexchange Hamiltonian contains various kinds of magnetic anisotropies. Here we are primarily interested in the magnetic properties of Sr_2IrO_4 and $Sr_2Ir_{1-x}Rh_xO_4$ compounds. We perform a systematic study of how magnetic interactions in these systems depend on the microscopic parameters and provide a thorough analysis of the resulting magnetic phase diagram. Comparison of our results with experimental data shows good agreement. Finally, we discuss the parameter space in which the spin-flop transition in Sr_2IrO_4, experimentally observed under pressure, can be realized.

PHYSICAL REVIEW B 92, 094439 (2015)

Theory of Raman response in three-dimensional Kitaev spin liquids: Application to β- and γ-Li_2IrO_3 compounds

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[1]*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455, USA*
[2]*Department of Physics, Cavendish Laboratory, JJ Thomson Avenue, Cambridge CB3 0HE, United Kingdom*
(Received 8 July 2015; published 25 September 2015)

We calculate the Raman response for the Kitaev spin model on the $\mathcal{H}$-0, $\mathcal{H}$-1, and $\mathcal{H}$-∞ harmonic honeycomb lattices. We identify several quantitative features in the Raman spectrum that are characteristic of the spin liquid phase. Unlike the dynamical structure factor, which probes both the Majorana spinons and flux excitations that emerge from spin fractionalization, the Raman spectrum in the Kitaev models directly probes a density of states of pairs of fractional, dispersing Majorana spinons. As a consequence, the Raman spectrum in all these models is gapless for sufficiently isotropic couplings, with a low-energy power law that results from the Fermi lines (or points) of the dispersing Majorana spinons. We show that the polarization dependence of the Raman spectrum contains crucial information about the symmetry of the ground state. We also discuss to what extent the features of the Raman response that we find reflect generic properties of the spin liquid phase, and comment on their possible relevance to α-, β-, and γ-Li_2IrO_3 compounds.

PHYSICAL REVIEW B 92, 155131 (2015)

Lifting mean-field degeneracies in anisotropic classical spin systems

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[2]*Condensed Matter Theory and Institute for Nanotechnology, Karlsruhe Institute of Technology, D-76128 Karlsruhe*
(Received 16 March 2015; revised manuscript received 10 September 2015; published 19 October 2015)

In this work, we propose a method for calculating the free energy of anisotropic classical spin systems. We use a Hubbard-Stratonovich transformation to express the partition function of a generic bilinear superexchange Hamiltonian in terms of a functional integral over classical time-independent fields. As an example, we consider an anisotropic spin-exchange Hamiltonian on the cubic lattice as is found for compounds with strongly correlated electrons in multiorbital bands and subject to strong spin-orbit interaction. We calculate the contribution of Gaussian spin fluctuations to the free energy. While the mean-field solution of ordered states for such systems usually has full rotational symmetry, we show here that the fluctuations lead to a pinning of the spontaneous magnetization along some preferred direction of the lattice.

PRL 114, 096404 (2015) PHYSICAL REVIEW LETTERS week ending 6 MARCH 2015

Structural Distortion-Induced Magnetoelastic Locking in Sr_2IrO_4 Revealed throu Nonlinear Optical Harmonic Generation

D. H. Torchinsky,[1,2] H. Chu,[1,3] L. Zhao,[1,2] N. B. Perkins,[4] Y. Sizyuk,[4] T. Qi,[5] G. Cao,[5] and D. Hsieh[1,
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(Received 10 July 2014; published 5 March 2015)

We report a global structural distortion in Sr_2IrO_4 using spatially resolved optical second and third harmonic generation rotational anisotropy measurements. A symmetry lowering from an $I4_1/acd$ to $I4_1/a$ space group is observed both above and below the Néel temperature that arises from a staggered tetragonal distortion of the oxygen octahedra. By studying an effective superexchange Hamiltonian that accounts for this lowered symmetry, we find that perfect locking between the octahedral rotation and magnetic moment canting angles can persist even in the presence of large noncubic local distortions. Our results explain the origin of the forbidden Bragg peaks recently observed in neutron diffraction experiments and reconcile the observations of strong tetragonal distortion and perfect magnetoelastic locking in Sr_2IrO_4.

Phase diagram and quantum order by disorder in the Kitaev K_1-K_2 honeycomb magnet

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[1]*School of Physics and Astronomy, University of Minnesota, Minneapolis, MN 55455, USA*
[2]*Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany*
[3]*Helmholtz-Zentrum Berlin für Materialien und Energie, 14109 Berlin, Germany*
[4]*Institute for Theoretical Physics, University of Würzburg, 97074 Würzburg, Germany*
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(Dated: July 1, 2015)

We show that the non-abelian Kitaev spin liquid on the honeycomb lattice is extremely fragile against the second neighbor Kitaev coupling K_2, which has been recently shown to be the dominant perturbation away from the nearest neighbor model in iridate Na_2IrO_3 and ruthenate $\alpha-RuCl_3$. This coupling explains naturally the zig-zag ordering in both compounds without introducing unphysically large long-range Heisenberg exchange terms. The minimal K_1-K_2 model that we present here hosts a number of unconventional aspects, such as the fundamentally different role of thermal and quantum fluctuations, which can be traced back to the principle that time reversal symmetry can only act globally in a quantum system.

Thank you