Spin-orbit Induced and Dynamics in Mott Insulating Iridates

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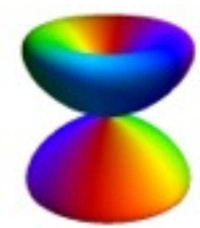
Max Planck Institute for Solid State Research

Universität Stuttgart

FMQ

GNSF



In collaboration with:

Theory:

G. Khaliullin (MPI Stuttgart) & J. Chaloupka (Uni Brno)

Experiment:

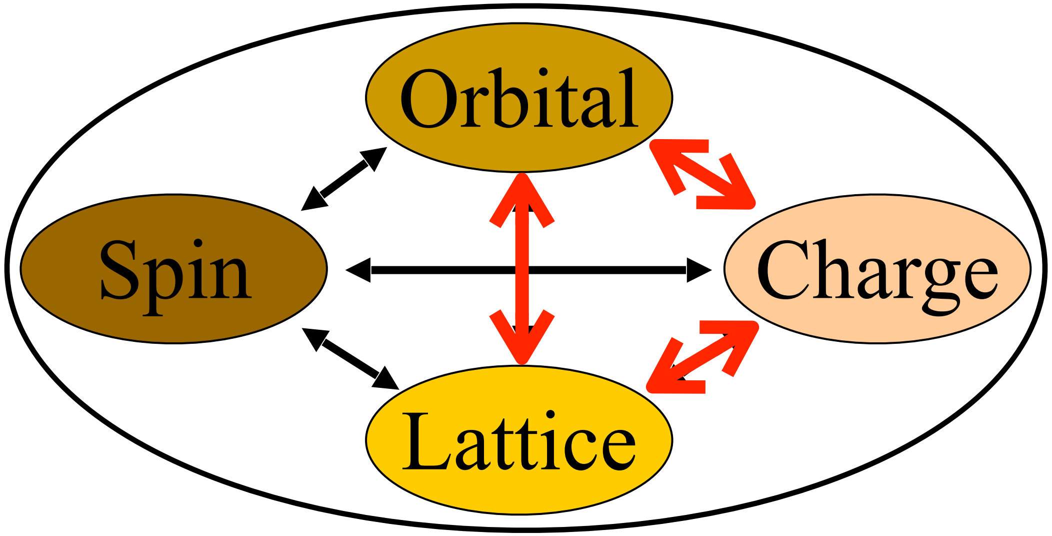
BJ. Kim (MPI Stuttgart),
V. Kataev (IFW Dresden) , H. Takagi (MPI Stuttgart & Uni Tokyo)

References:

GJ & G.Khaliullin, PRL 102, 017205 (2009)
103, 067205 (2009)
J.Chaloupka, GJ & G.Khaliullin, PRL 105, 027204 (2010)
110, 097204 (2013)
Kim et al, PRL 109, 037204 (2012)
109, 157402 (2012)
Bahr et al, PRB 180401(R) (2014)

Transition metal oxides: many challenges

- d- electrons are partially filled and not very extended
- Interactions dominate over kinetic energy
- Many degrees of freedom are at work

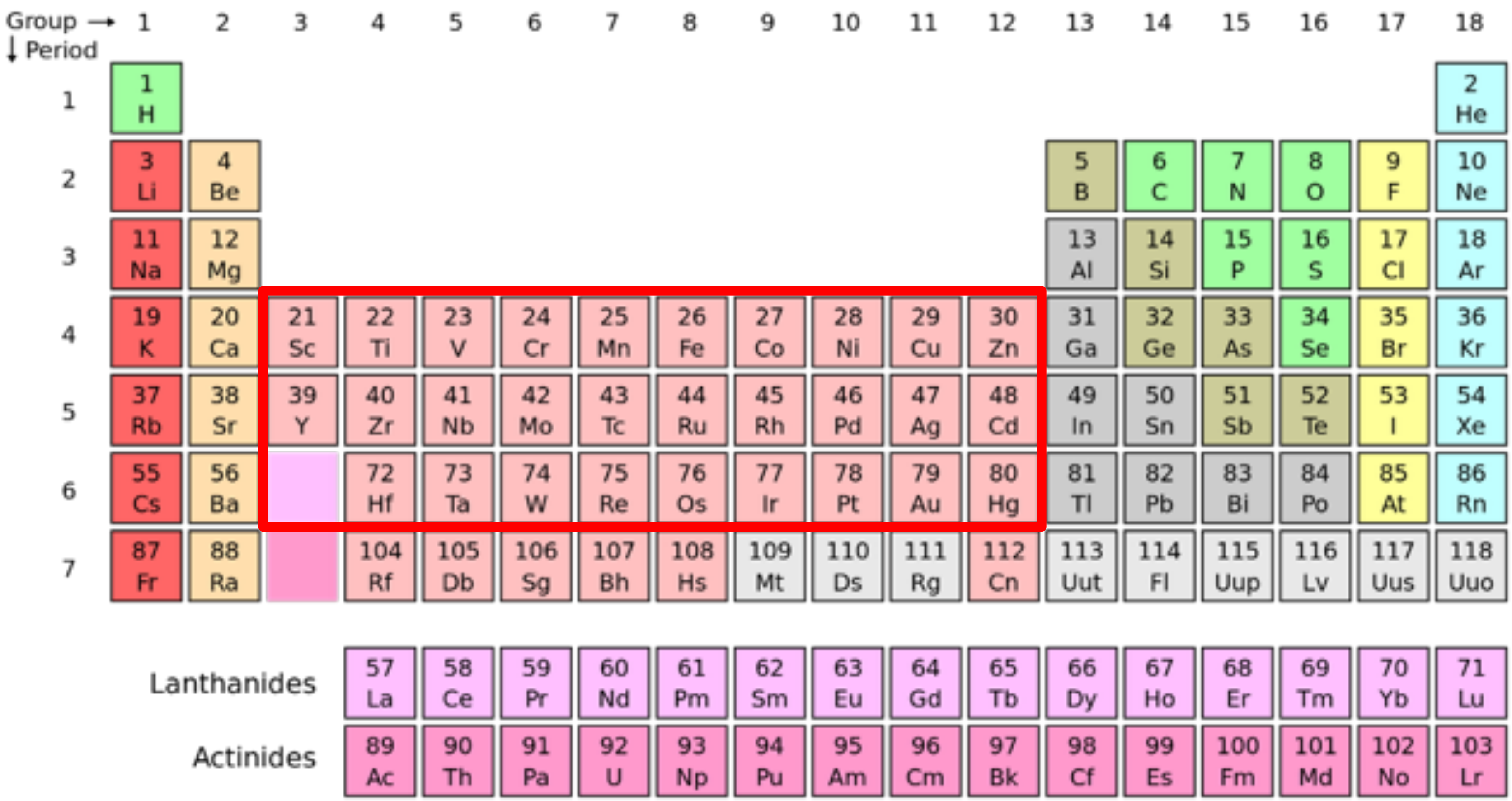


Metal to Magnetic Insulator

High-Tc Superconductivity
Colossal Magnetoresistance

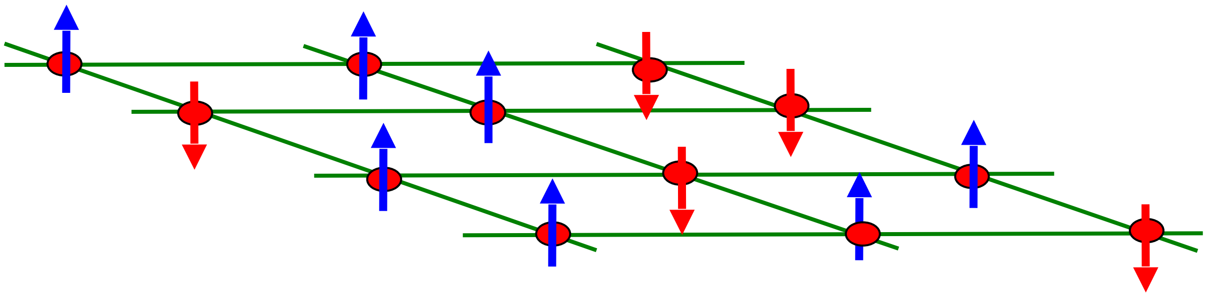
Unexpected variety of phases
and transitions between them

Most of the elements are Transition metals

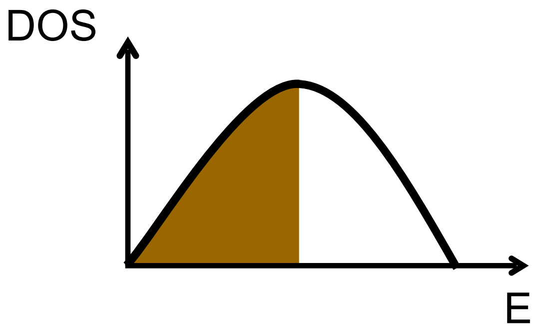


Mott Insulator

one electron per site

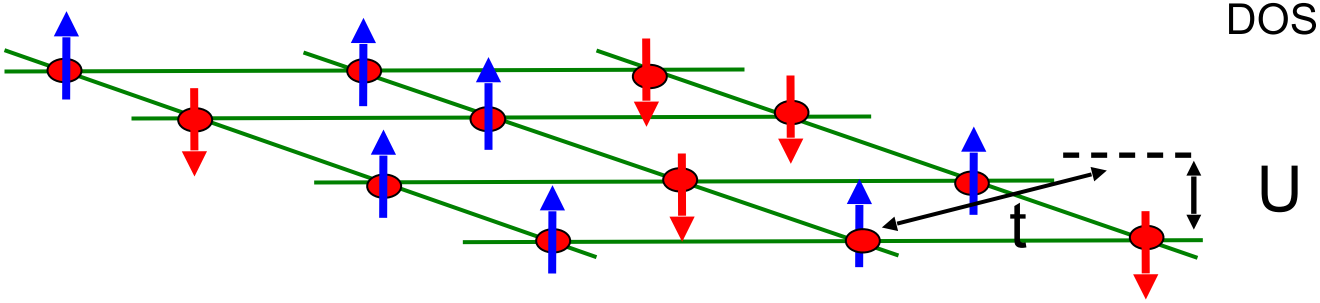


half-filled band: metal

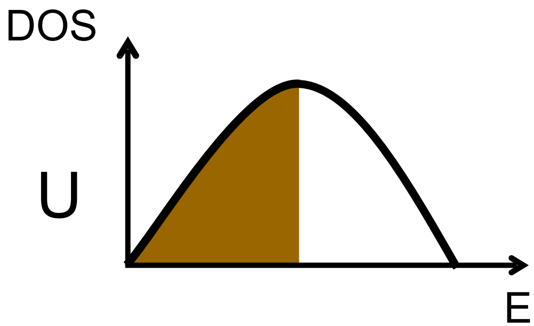


Mott Insulator

one electron per site

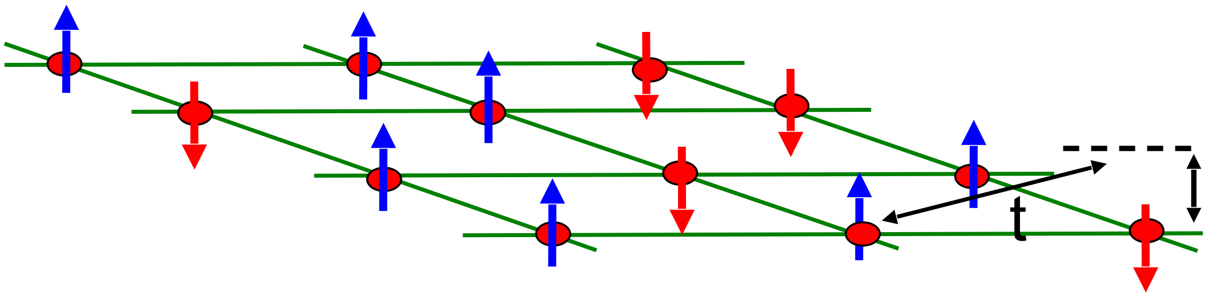


half-filled band: metal

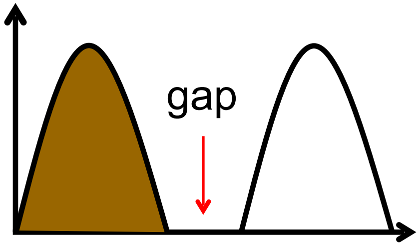


Mott Insulator

one electron per site



half-filled band: insulator

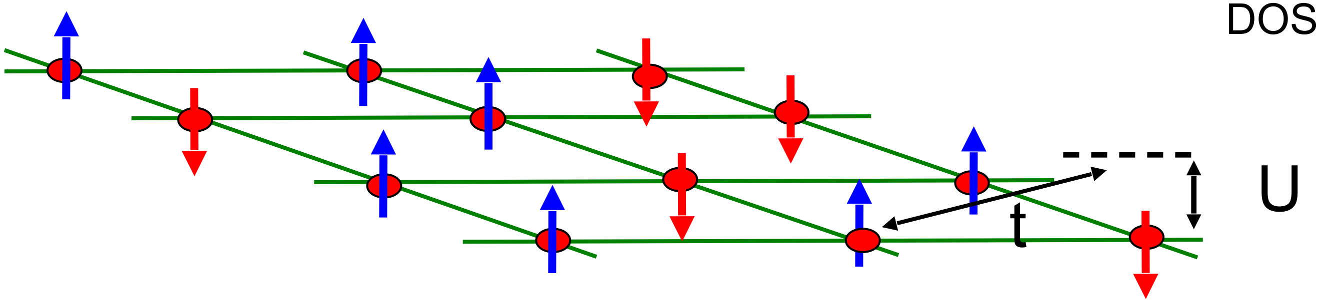


Sir Nevill Mott
(Nobel prize'77)

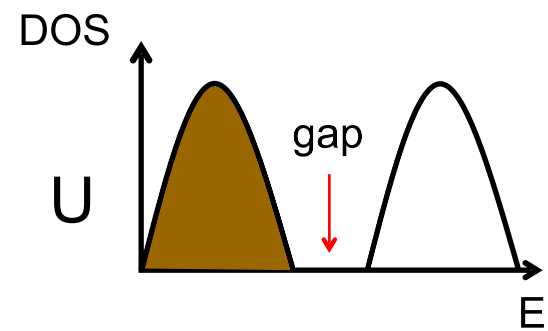
Coulomb repulsion U>>t Kinetic energy,
Charges become localized and gapped

Mott Insulator

one electron per site



half-filled band: insulator



Sir Nevill Mott
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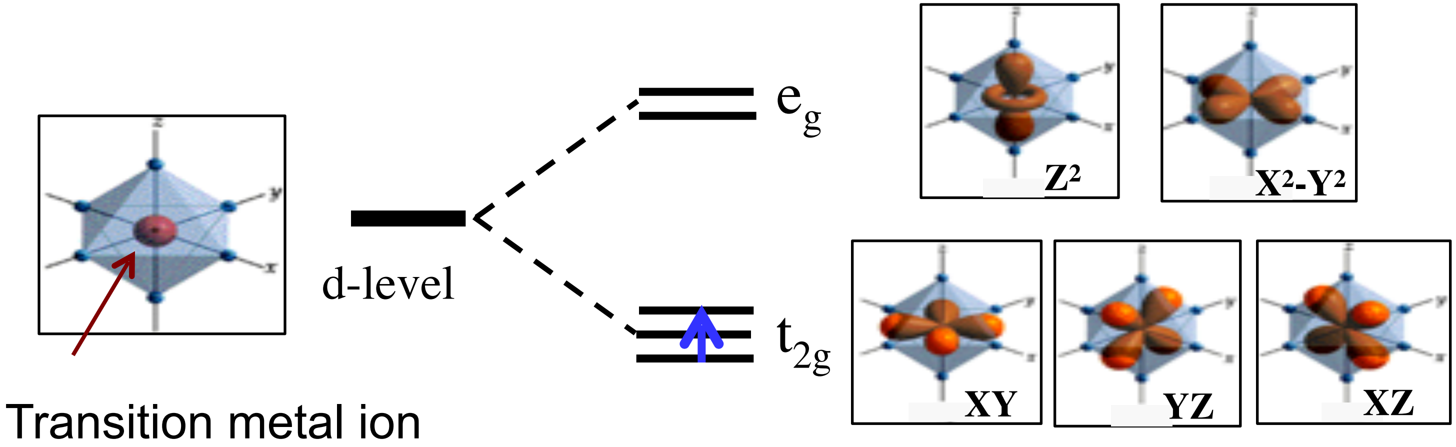
Coulomb repulsion U>>t Kinetic energy,
Charges become localized and gapped

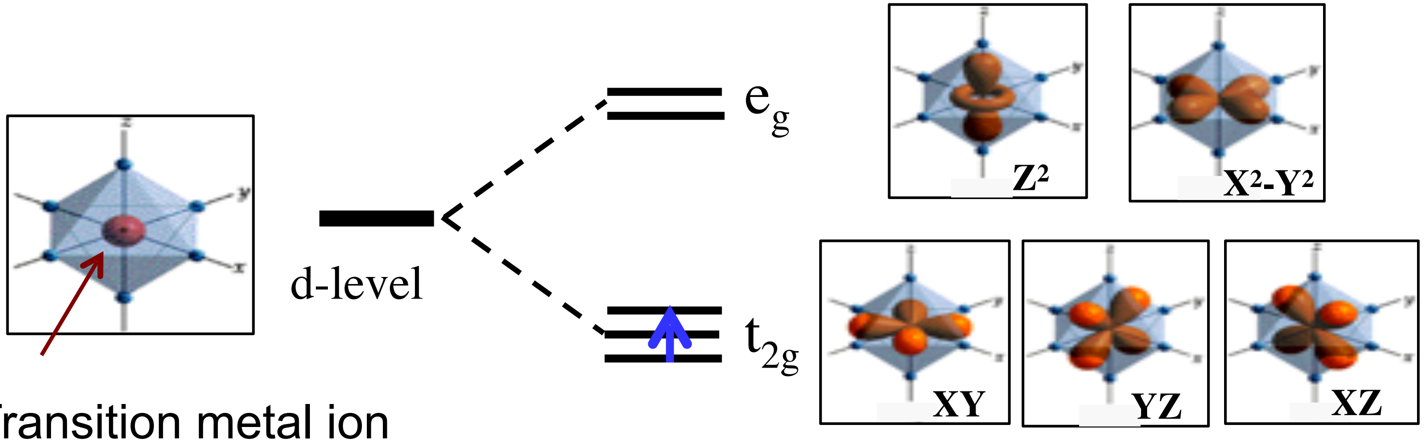
Werner Heisenberg
(Nobel prize'32)

Low energy degrees are magnetic:
Spins interacting via Heisenberg exchange

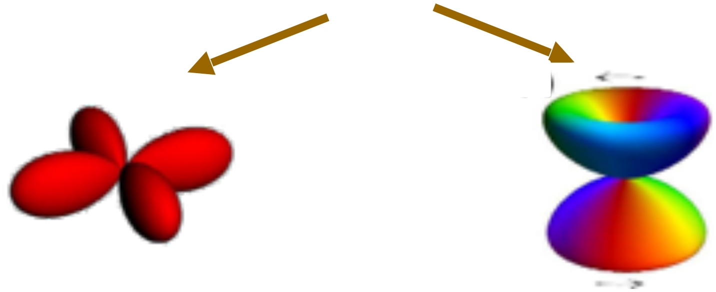
$$H = J \sum_{ij} \left[\vec{S}_i \vec{S}_j - 1/4 \right] \quad J = \frac{4t^2}{U}$$



d-level
e_g
t_2g
Z²
X²-Y²
XY
YZ
XZ
Transition metal ion



Unquenched angular momentum $L=-l_{eff}=1$

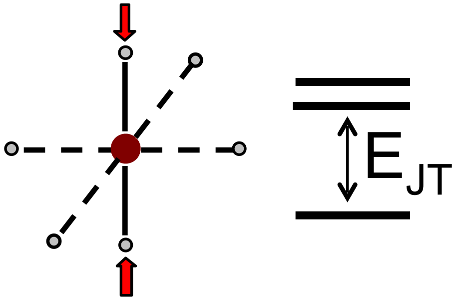


$|l_z\!=\!0\rangle \equiv |xy\rangle$

$|l_z\!=\!\pm1\rangle \equiv -\frac{1}{\sqrt{2}}(i|xz\rangle \pm |yz\rangle)$

Three different couplings & regimes in spin-orbital systems

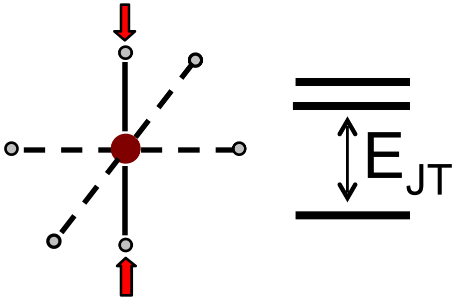
Jahn-Teller coupling- E_{JT}



orbitals are rigidly ordered:
spin-only Heisenberg model

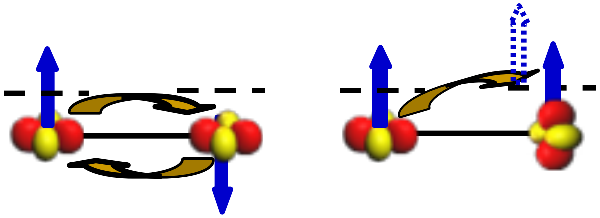
Three different couplings & regimes in spin-orbital systems

Jahn-Teller coupling- E_{JT}

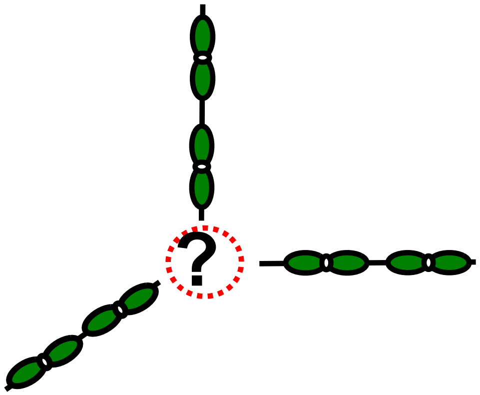


orbitals are rigidly ordered:
spin-only Heisenberg model

exchange interactions- J

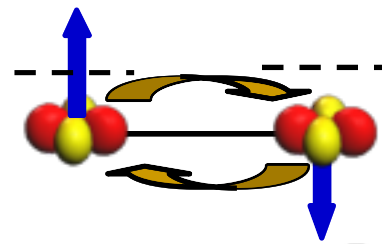


spin exchange depends on orbital occupancy:
directional character of orbitals induces frustration

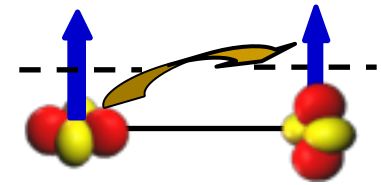


Coupled spin-orbital model

$$H = J_{\mathrm{AF}} \sum_{\langle ij \rangle} \left[\vec{S}_i \cdot \vec{S}_j / S^2 - 1 \right] O_{ij}^{\mathrm{OF}} - \sum_{\langle ij \rangle} \left[J_{\mathrm{O}} + J_{\mathrm{F}} \vec{S}_i \cdot \vec{S}_j / S^2 \right] O_{ij}^{\mathrm{OAF}}$$



AFM coupling ~ J



FM coupling ~ ηJ

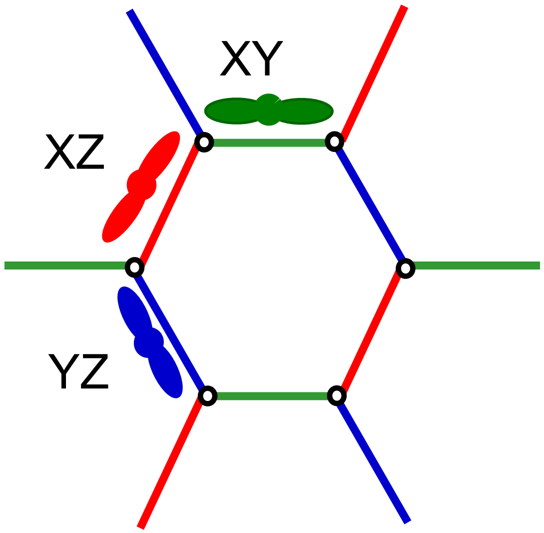
Exchange couplings:

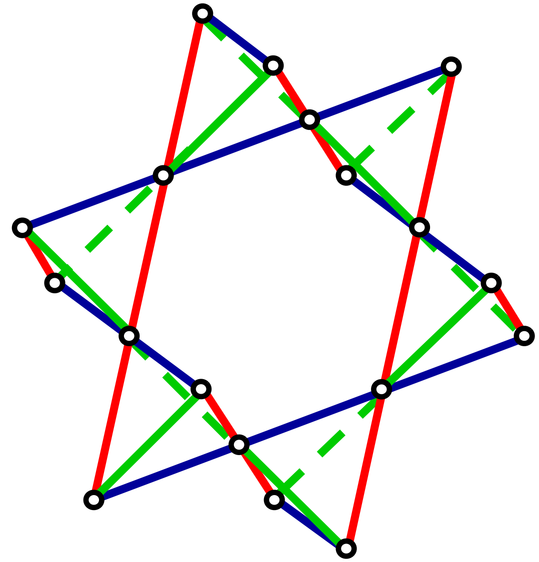
$$J_{\mathrm{AF}} \simeq J_{\mathrm{O}} \sim J = t^2/U$$

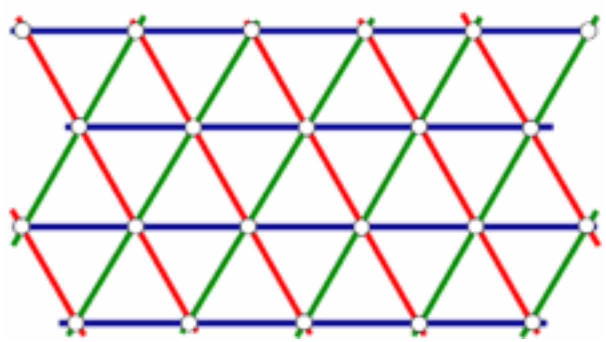
$$J_{\mathrm{F}} \sim \eta J, \quad \eta = J_{\mathrm{H}}/U \ll 1$$

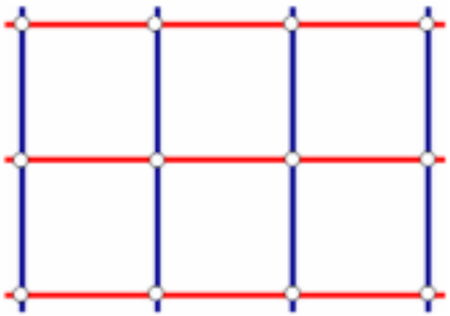
Orbital degrees are static Pott's-like!

Directional orbitals on a family of lattices





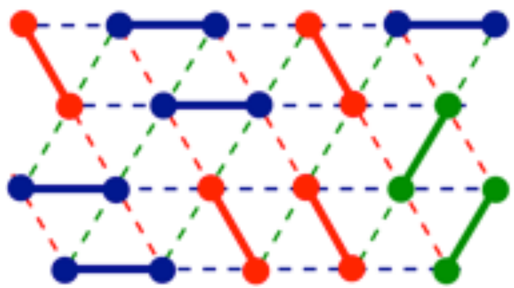


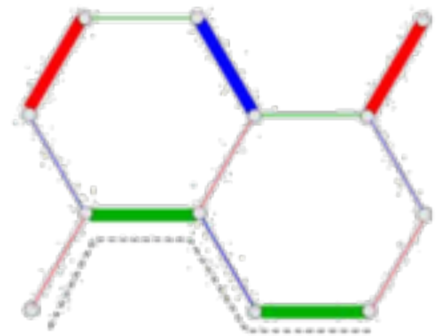


Ground state manifold: Hard core dimers

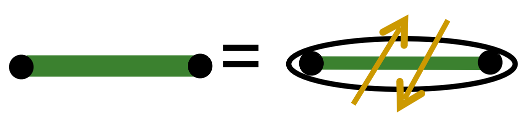
GJ & Ivanov PRB (2007)
GJ & Khomskii PRL (2008)

- Ground state manifold is spanned by hard-core dimer coverings





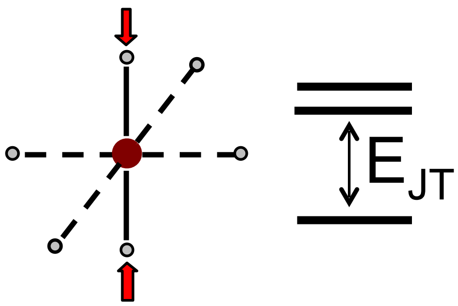
- Spins are bound into spin-singlet on dimer bonds: Spin gaped phase



- Extensive orientational degeneracy infinitely many ways of covering
  Different coverings are orthogonal due to orbitals

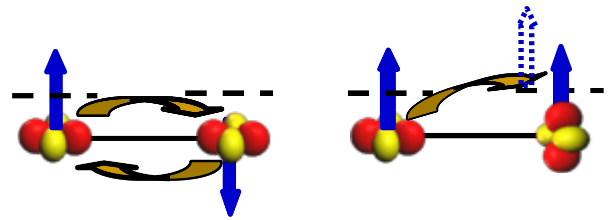
Three different couplings & regimes in spin-orbital systems

Jahn-Teller coupling- E_{JT}

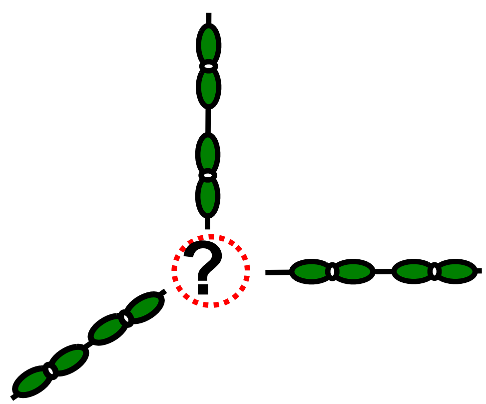


orbitals are rigidly ordered:
spin-only Heisenberg model

exchange interactions- J



spin exchange depends on orbital occupancy:
directional character of orbitals induces frustration



spin-orbit coupling-λ

$$\lambda \vec{l} \cdot \vec{s}$$

spins & orbitals locally entangled:
orbital frustration & directional character are converted to isospins

correlations ↑

21 **Sc** 44.9559 Scandium	22 **Ti** 47.867 Titanium	23 **V** 50.9415 Vanadium	24 **Cr** 51.9961 Chromium	25 **Mn** 54.938 Manganese	26 **Fe** 55.845 Iron	27 **Co** 58.9332 Cobalt	28 **Ni** 58.6934 Nickel	29 **Cu** 63.546 Copper	30 **Zn** 65.4089 Zinc
39 **Y** 88.9058 Yttrium	40 **Zr** 91.224 Zirconium	41 **Nb** 92.9064 Niobium	42 **Mo** 85.94 Molybdenum	43 **Tc** 98 Technetium	44 **Ru** 101.07 Ruthenium	45 **Rh** 102.9055 Rbodium	46 **Pd** 106.42 Palladium	47 **Ag** 107.8682 Silver	48 **Cd** 112.411 Cadmium
71 **Lu** 174.967 Lutetium	72 **Hf** 178.49 Hafnium	73 **Ta** 180.9497 Tantalum	74 **W** 183.84 Tungsten	75 **Re** 186.207 Rhenium	76 **Os** 190.23 Osmium	77 **Ir** 192.217 Iridium	78 **Pt** 195.084 Platinum	79 **Au** 196.9666 Gold	80 **Hg** 200.59 Mercury

spin-orbit ↓

correlations ↑

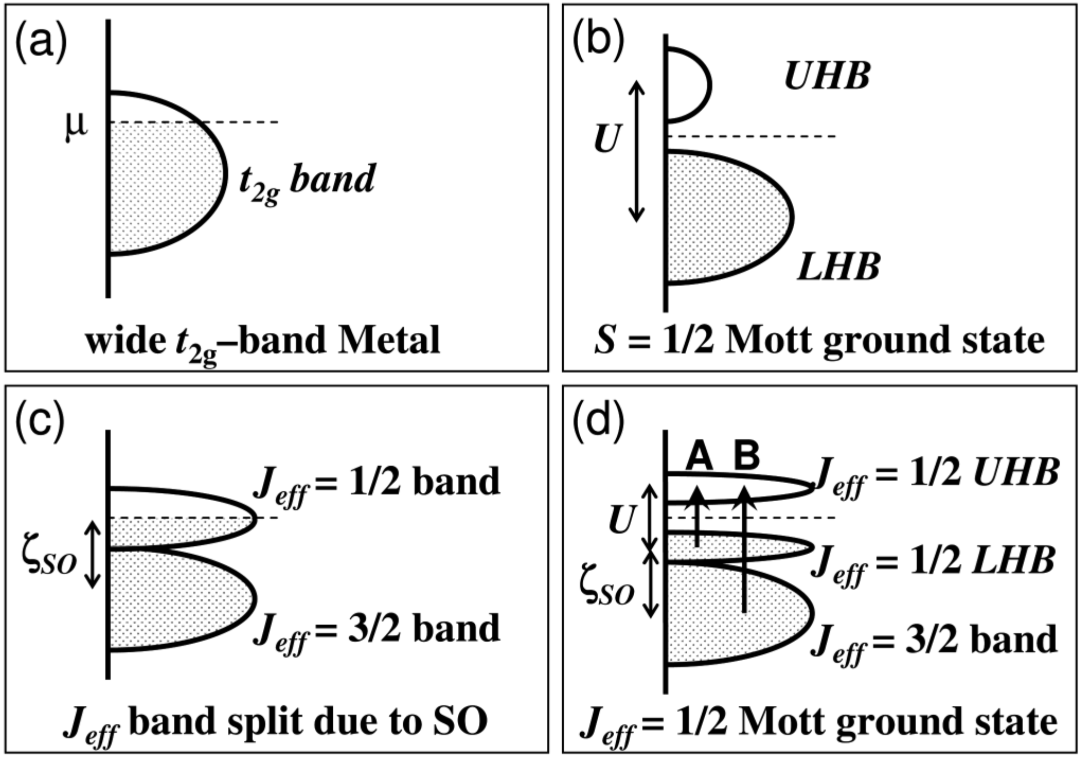
21	22	23	24	25	26	27	28	29	30
Sc	**Ti**	**V**	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Cu**	**Zn**
44.9559	47.867	50.9415	51.9961	54.938	55.845	58.9332	58.6934	63.546	65.4089
Scandium	Titanium	Vanadium	Chromium	Manganese	Iron	Cobalt	Nickel	Copper	Zinc
39	40	41	42	43	44	45	46	47	48
Y	**Zr**	**Nb**	**Mo**	**Tc**	**Ru**	**Rh**	**Pd**	**Ag**	**Cd**
88.9058	91.224	92.9064	85.94	98	101.07	102.9055	106.42	107.8682	112.411
Yttrium	Zirconium	Niobium	Molybdenum	Technetium	Ruthenium	Rhodium	Palladium	Silver	Cadmium
71	72	73	74	75	76	77	78	79	80
Lu	**Hf**	**Ta**	**W**	**Re**	**Os**	**Ir**	**Pt**	**Au**	**Hg**
174.967	178.49	180.9497	183.84	186.207	190.23	192.217	195.084	196.9666	200.59
Lutetium	Hafnium	Tantalum	Tungsten	Rhenium	Osmium	Iridium	Platinum	Gold	Mercury

spin-orbit ↓

with lucky atomic number 77

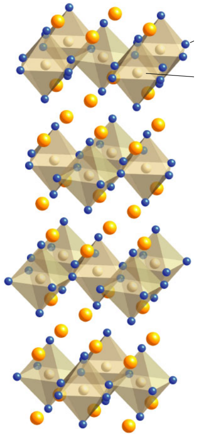
Novel $J_{\mathrm{eff}} = 1/2$ Mott State Induced by Relativistic Spin-Orbit Coupling in Sr_2IrO_4

B. J. Kim,[1] Hosub Jin,[1] S. J. Moon,[2] J.-Y. Kim,[3] B.-G. Park,[4] C. S. Leem,[5] Jaejun Yu,[1] T. W. Noh,[2] C. Kim,[5] S.-J. Oh,[1] J.-H. Park,[3,4,*] V. Durairaj,[6] G. Cao,[6] and E. Rotenberg[7]



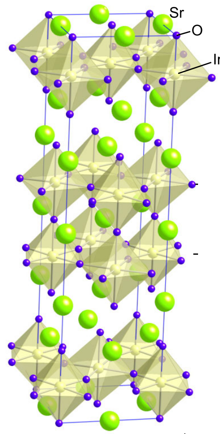
Spin-Orbit assisted Mott transition

'Zoo' of Iridate compounds

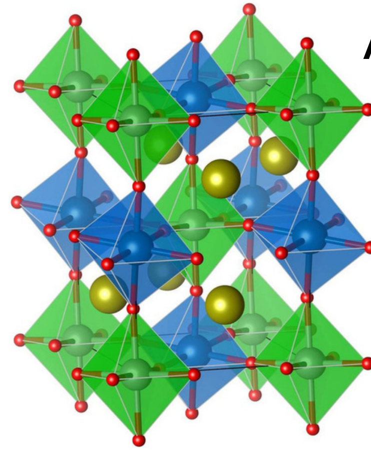


A_2IrO_4

Talk: Ashim K.Pamarik

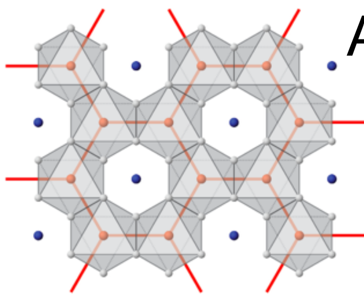


$A_3Ir_2O_7$



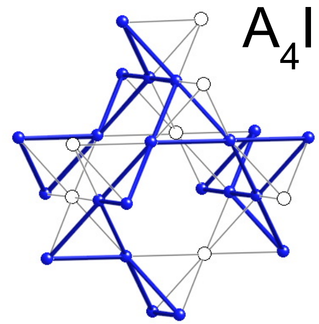
A_2BIrO_6

Talk: Arun Paremekanti



A_2IrO_3

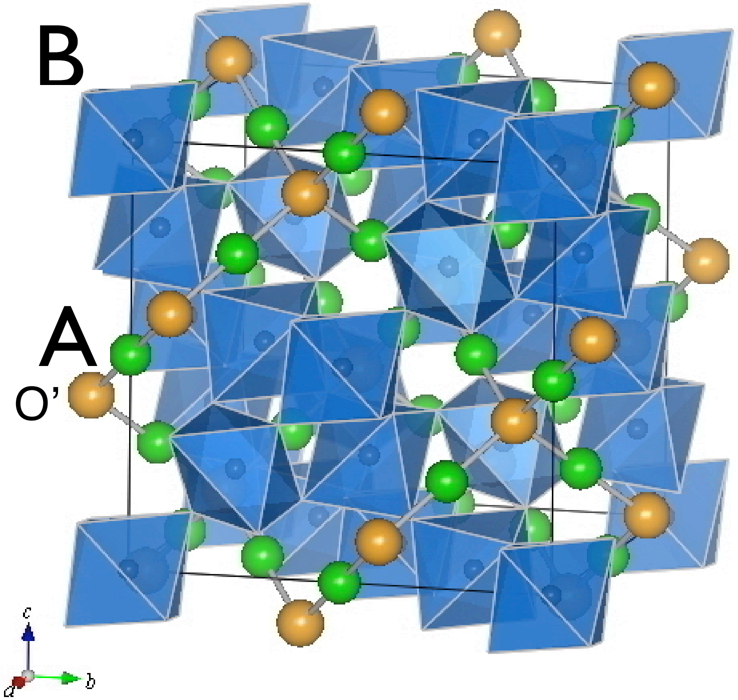
Talks: BJ Kim,
Yogesh Singh

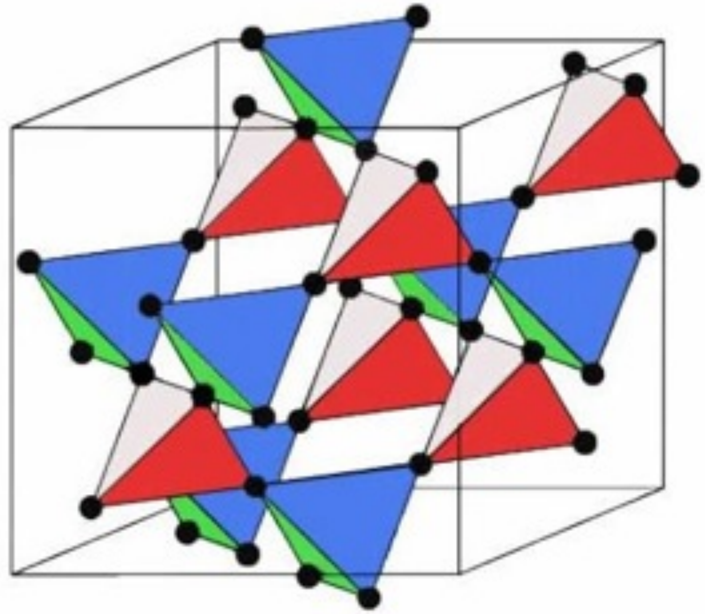


$A_4Ir_3O_8$

Talks: Philippe Mendels,
Yogesh Singh

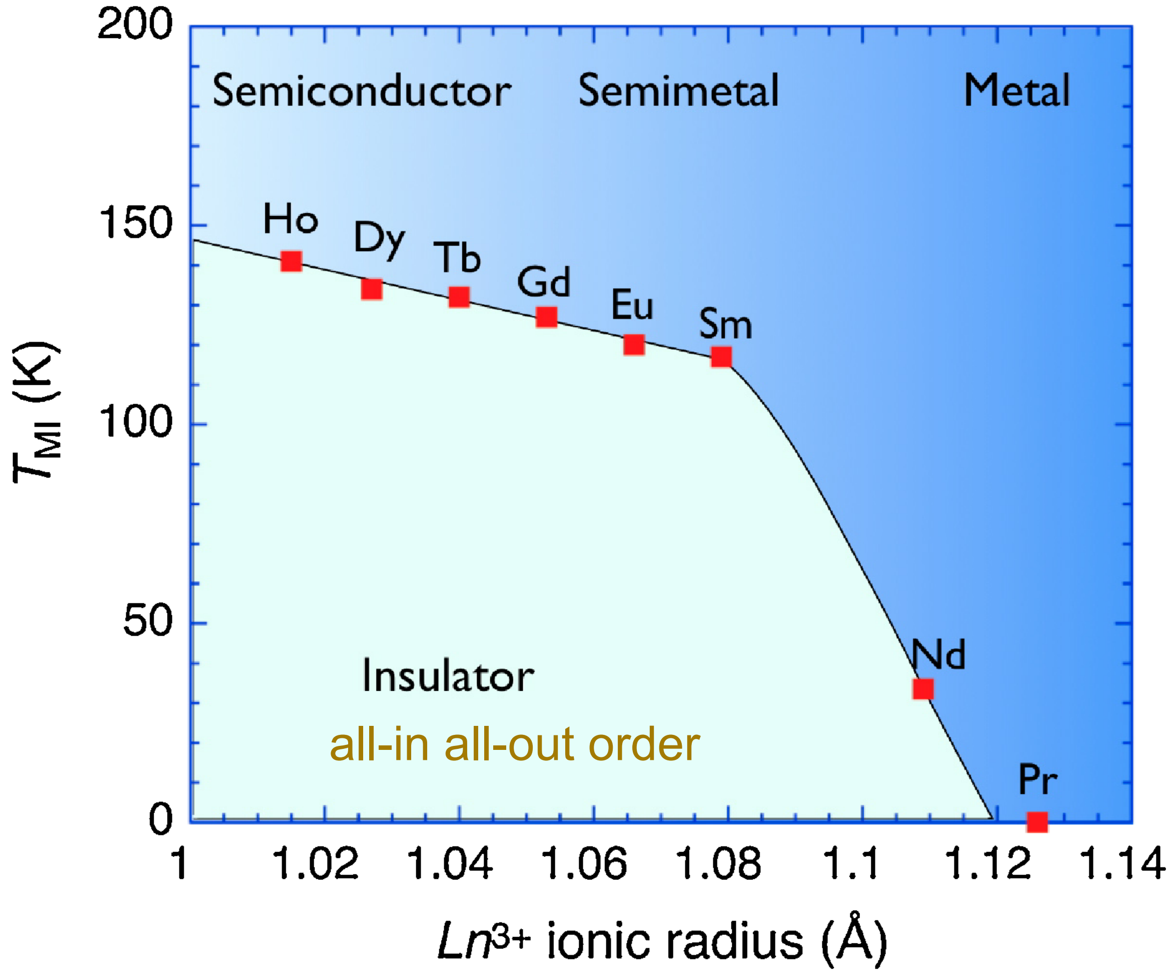
Pyrochlore Iridates $Sr_3Ir_2O_7$

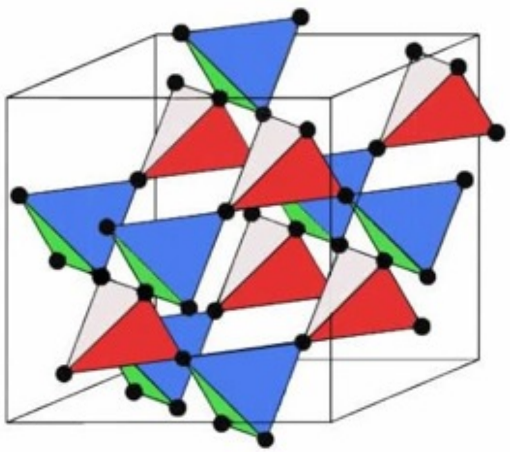




Pyrochlore Iridates $Sr_3Ir_2O_7$

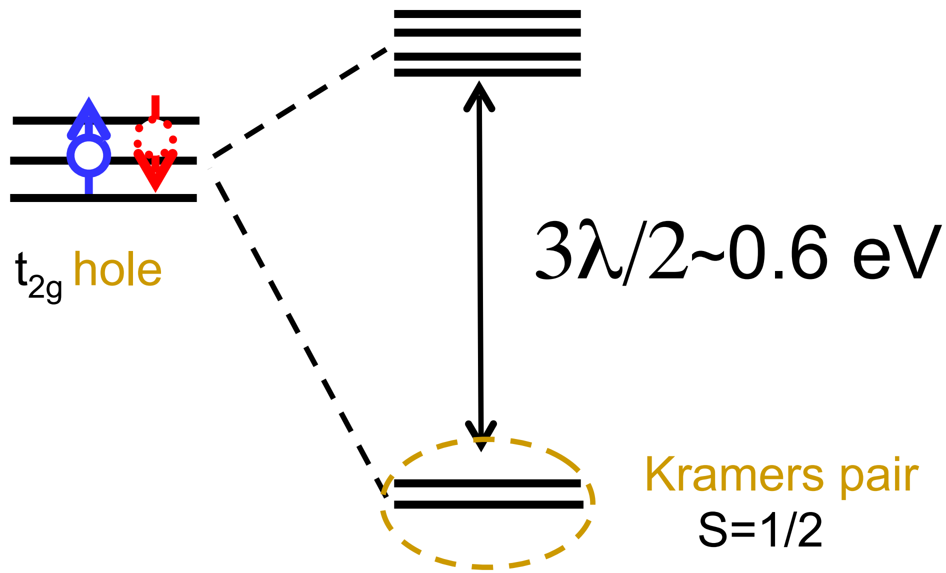
Matsuhira et al, JPSJ (2011)

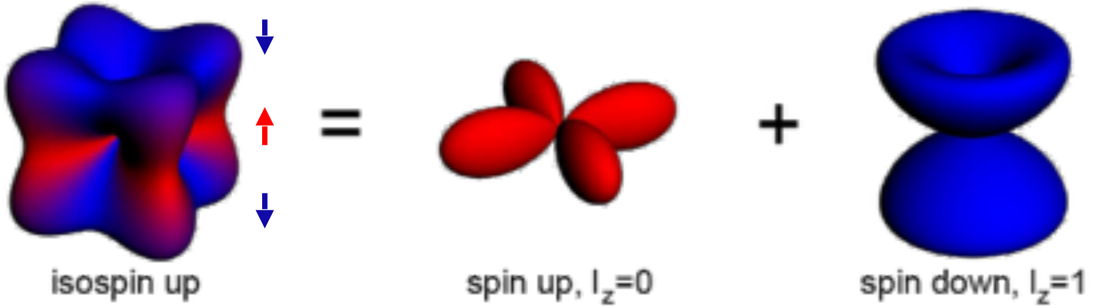




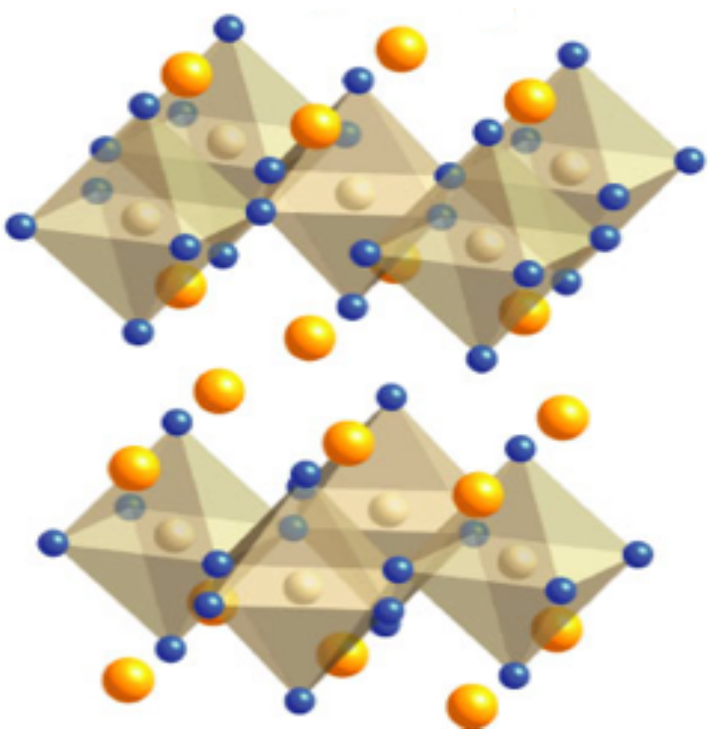
- single layer Sr_2IrO_4 : isotropic, nearly Heisenberg
- double layer $Sr_3Ir_2O_7$: anisotropic, nearly Ising
- hexagonal layered A_2IrO_3 : bond directional Ising
- Magnetically hidden octupolar order in Sr_2VO_4

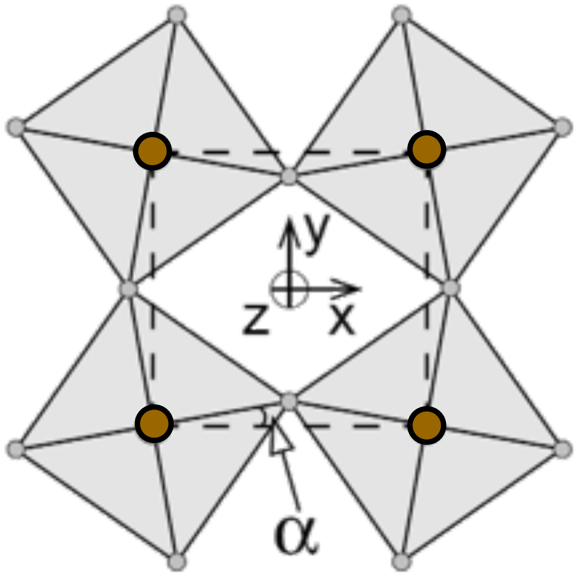
Low energy Kramers doublet of Ir^{4+}





Crystal structure of Sr_2IrO_4





Staggered rotation of octahedra around c-axis by $\alpha \sim 11^{o}$

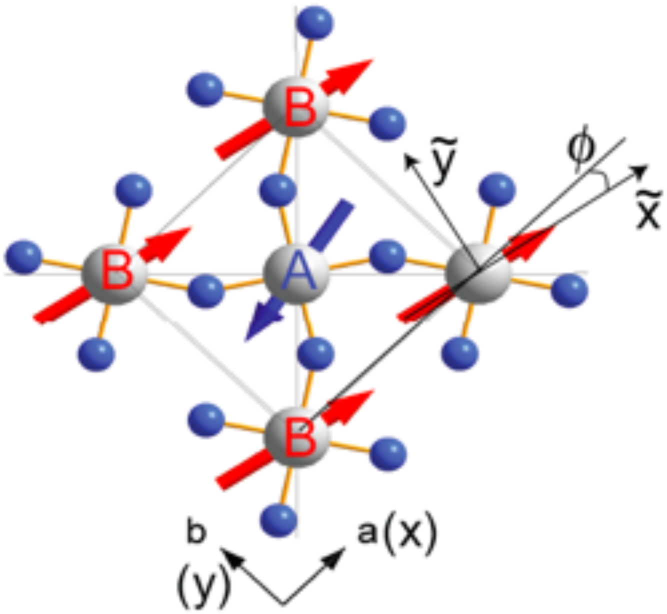
Magnetic structure of Sr_2IrO_4

Cao et al., PRB '98

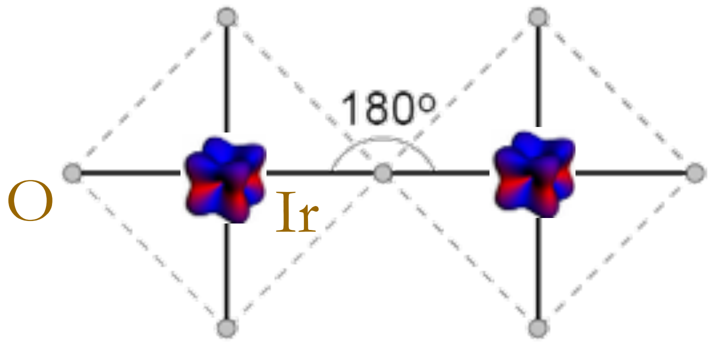
- FM moment M_{FM}~$0.1\mu_B$:
  too small for a saturated FM

  too large for weak a FM
  [e.g. in La_2CuO_4: $0.2x10^{-2}\mu_B$]

- Canted AFM in xy-plane ?
  Spin canting angle $\phi \sim \alpha$
  Spins rigidly follow rotation of octahedra



Super-exchange between Isospins

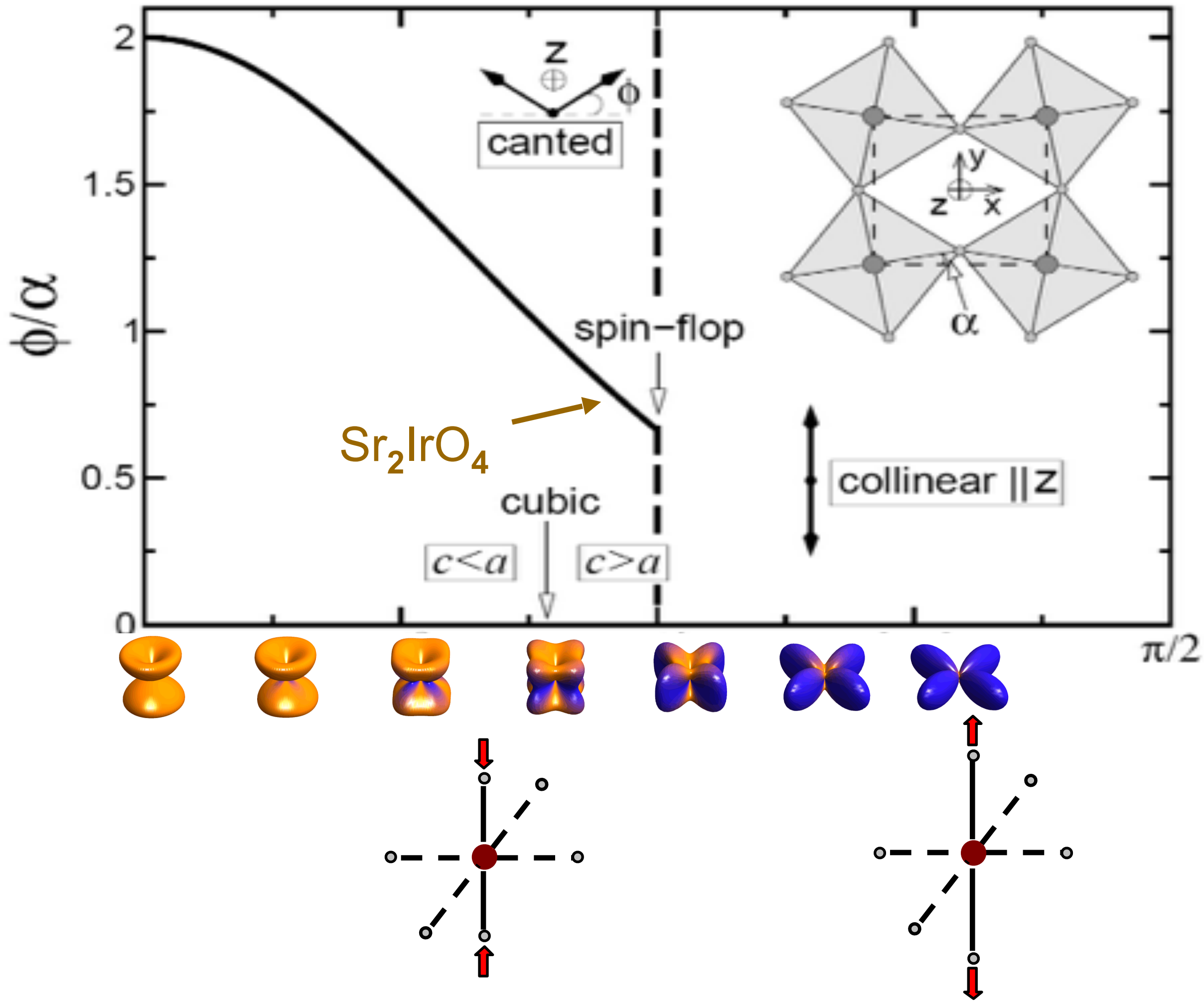


$$\mathcal{H}_{ij} = J_1 \vec{S}_i \cdot \vec{S}_j + J_2 (\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j)$$

AF-Heisenberg and weaker “dipolar”
+
DM induced by oct. rotations

Magnetism anisotropies are set by distortions

GJ & Khaliullin PRL (2009)



Phase-Sensitive Observation of a Spin-Orbital Mott State in Sr_2IrO_4

B. J. Kim,[1,2]* H. Ohsumi,[3] T. Komesu,[3] S. Sakai,[3,4] T. Morita,[3,5] H. Takagi,[1,2]* T. Arima[3,6]

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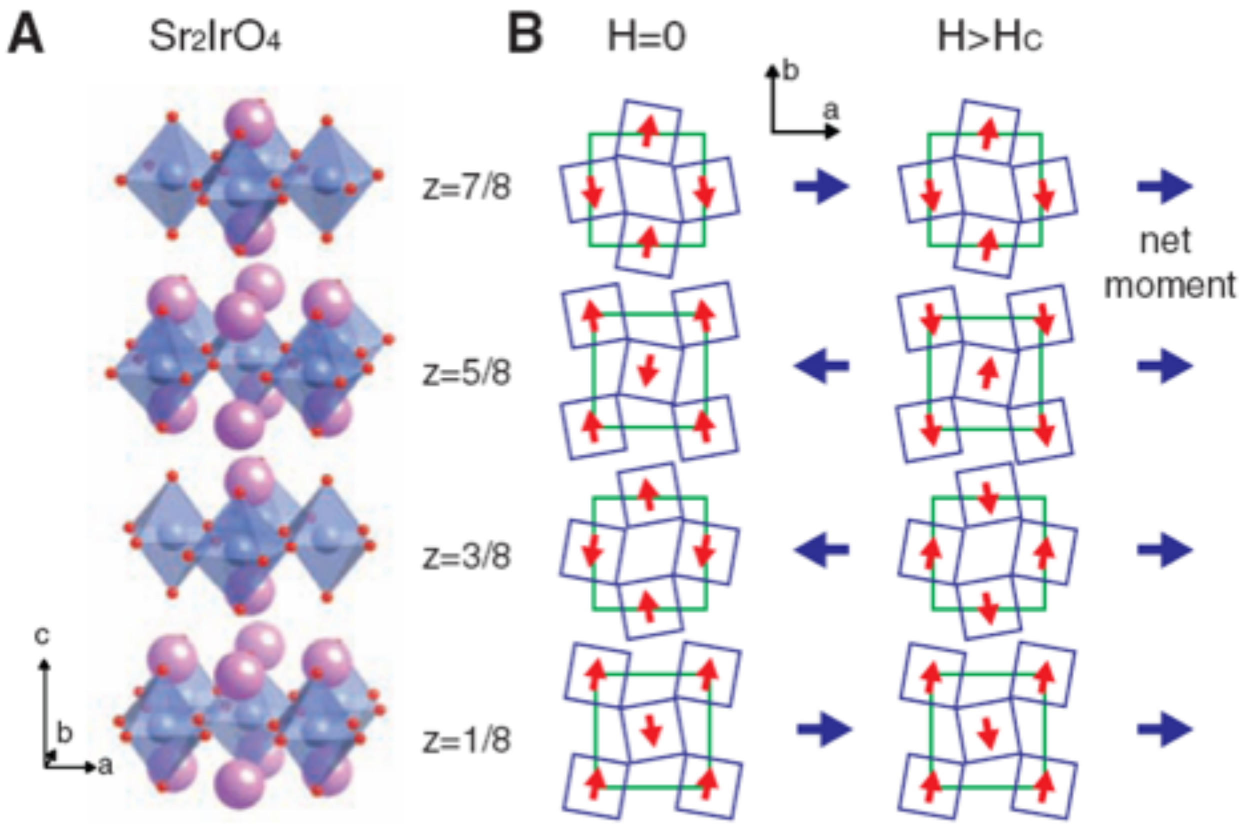
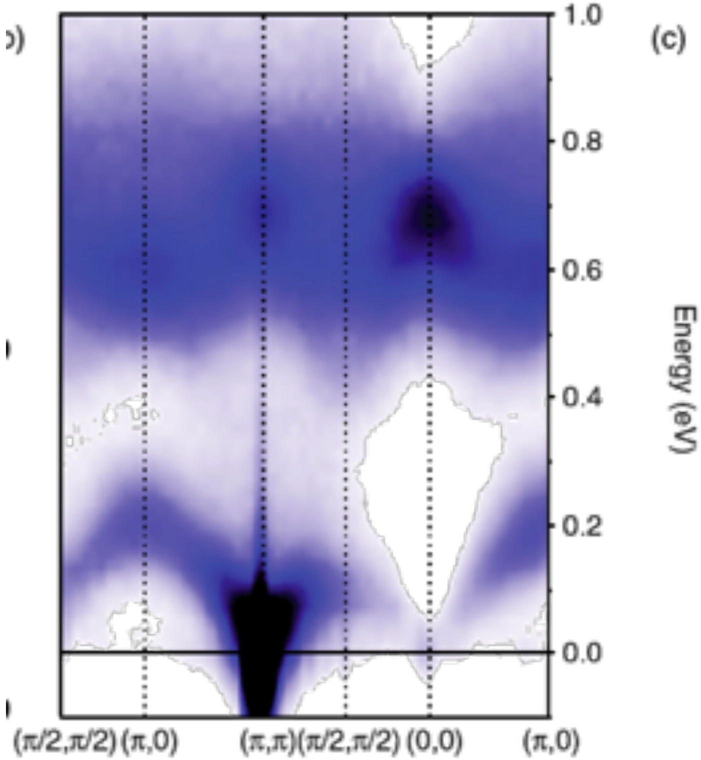
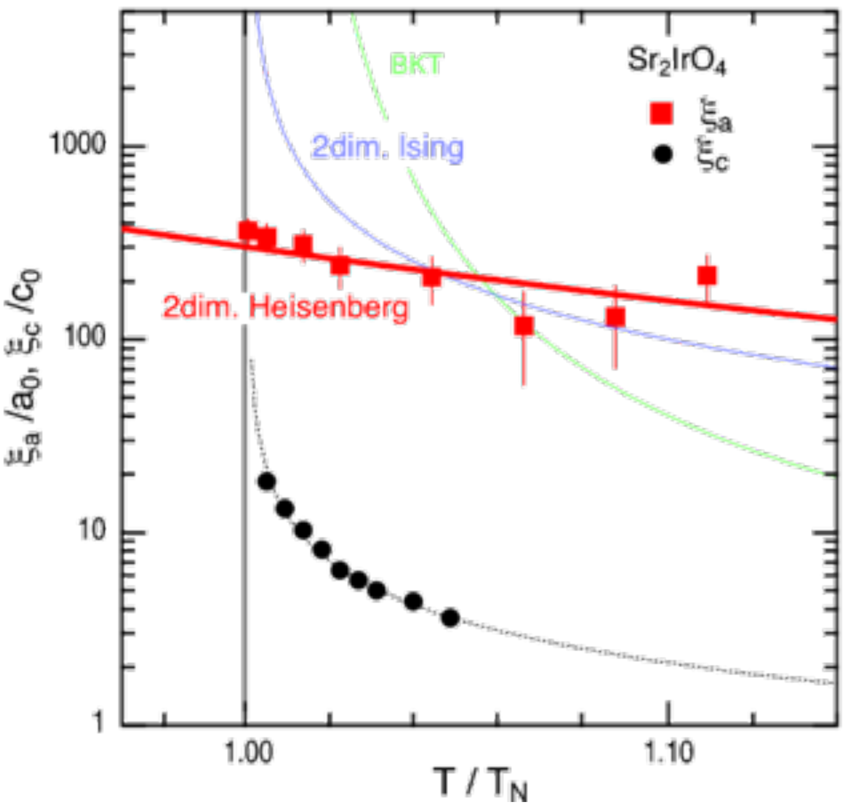


Heisenberg Magnetism of Sr_2IrO_4

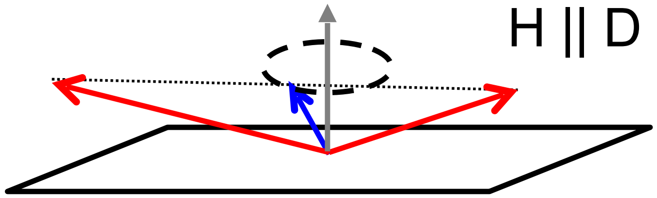


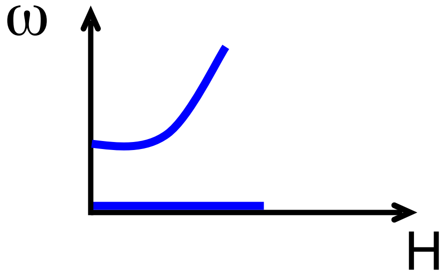
Kim et al, PRL (2012)

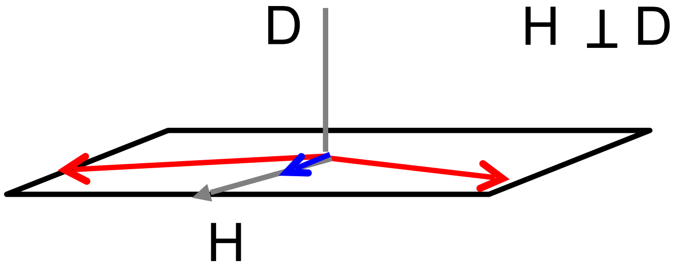


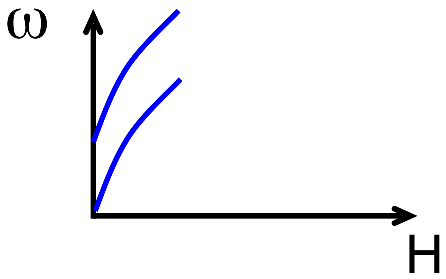
Fujiyama et al, PRL (2012)

Spin-Wave Modes



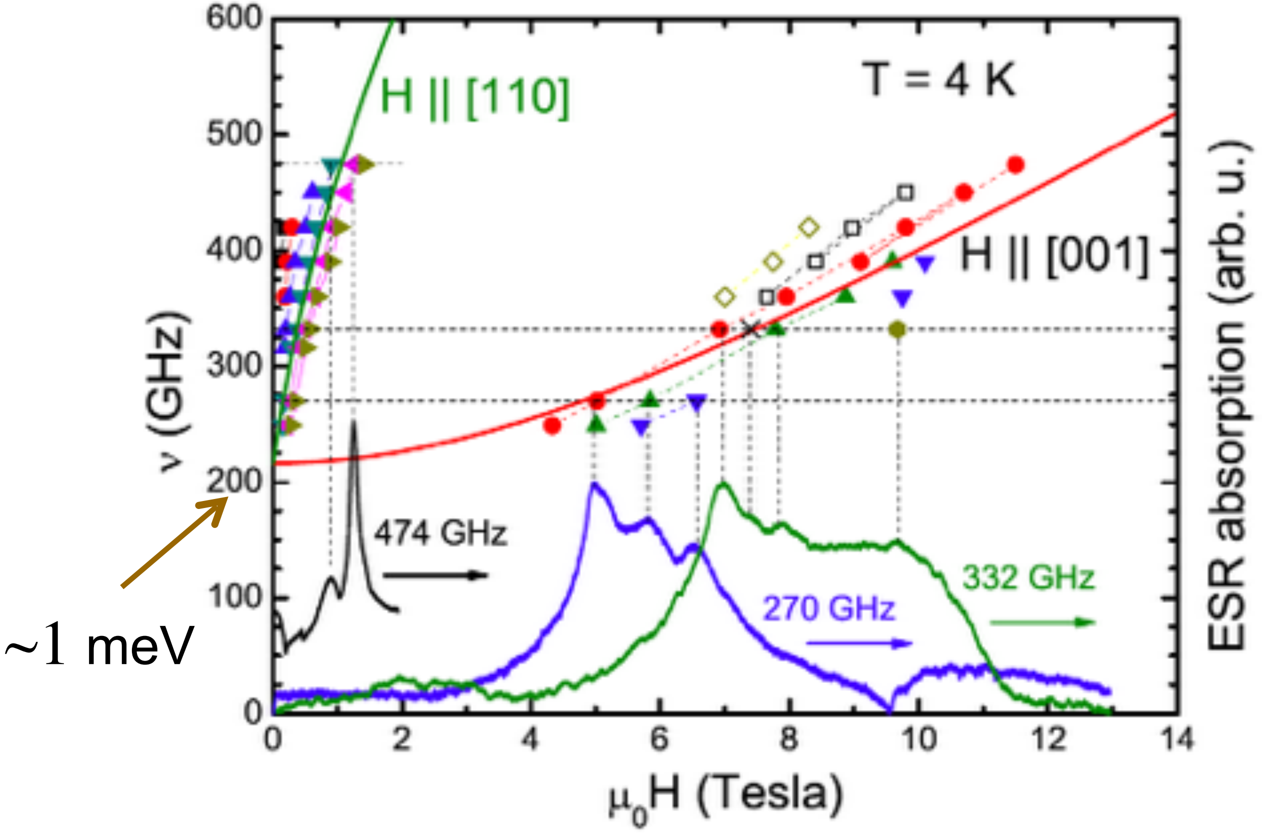






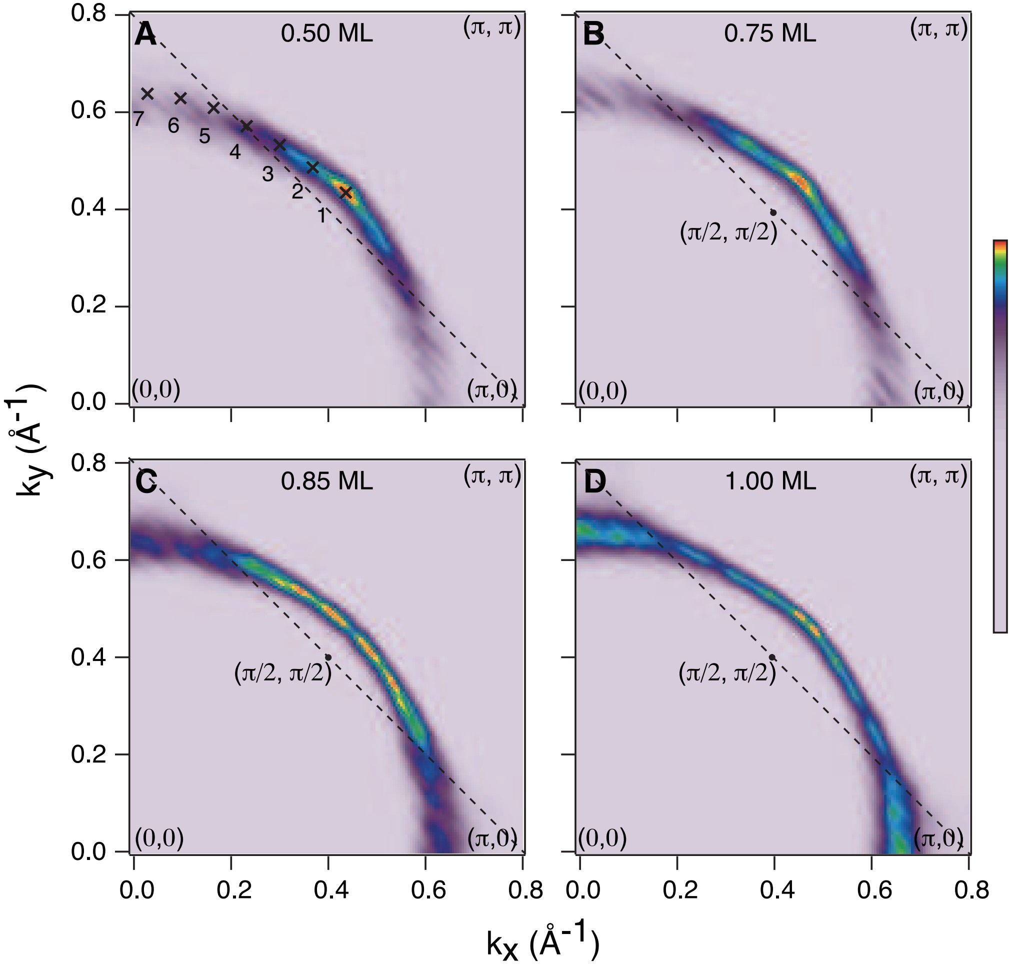
ESR spectra of Sr_2IrO_4

V.Kataev, GJ et al PRB (2014)



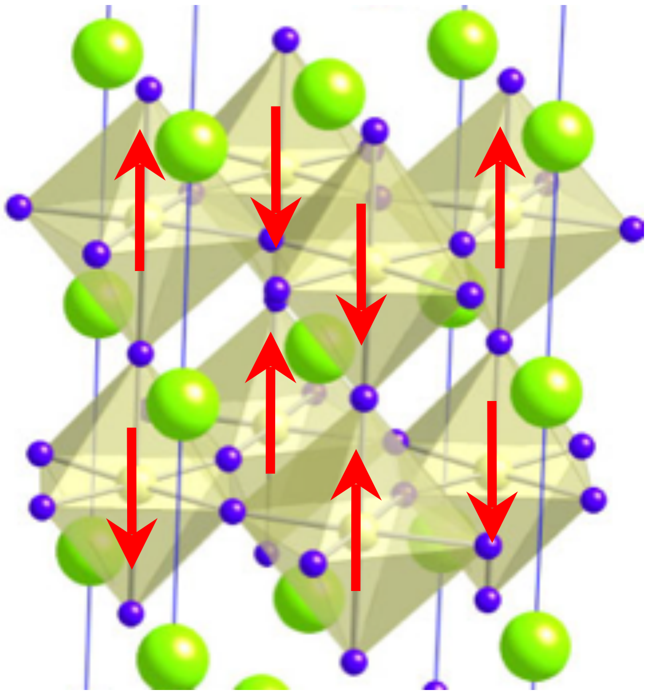
Fermi arcs in doped Sr_2IrO_4

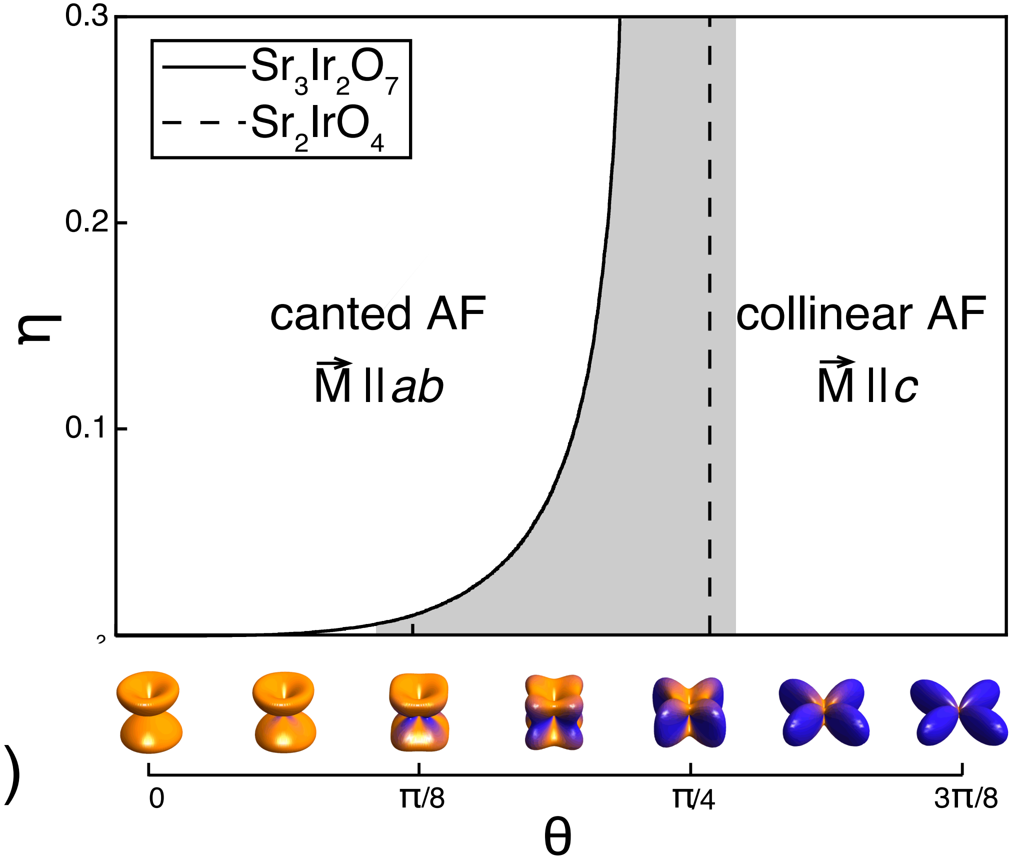
BJ Kim et al Science (2015)



- single layer Sr_2IrO_4 : isotropic, nearly Heisenberg
- double layer $Sr_3Ir_2O_7$: anisotropic, nearly Ising
- hexagonal layered A_2IrO_3 : bond directional Ising

Magnetism of $Sr_3Ir_2O_7$

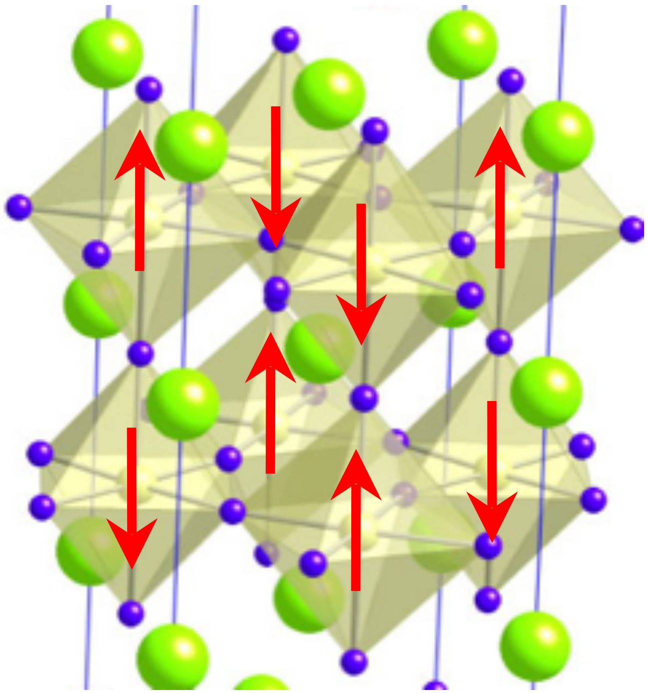




- *c* -axis AF (contrary to 214)

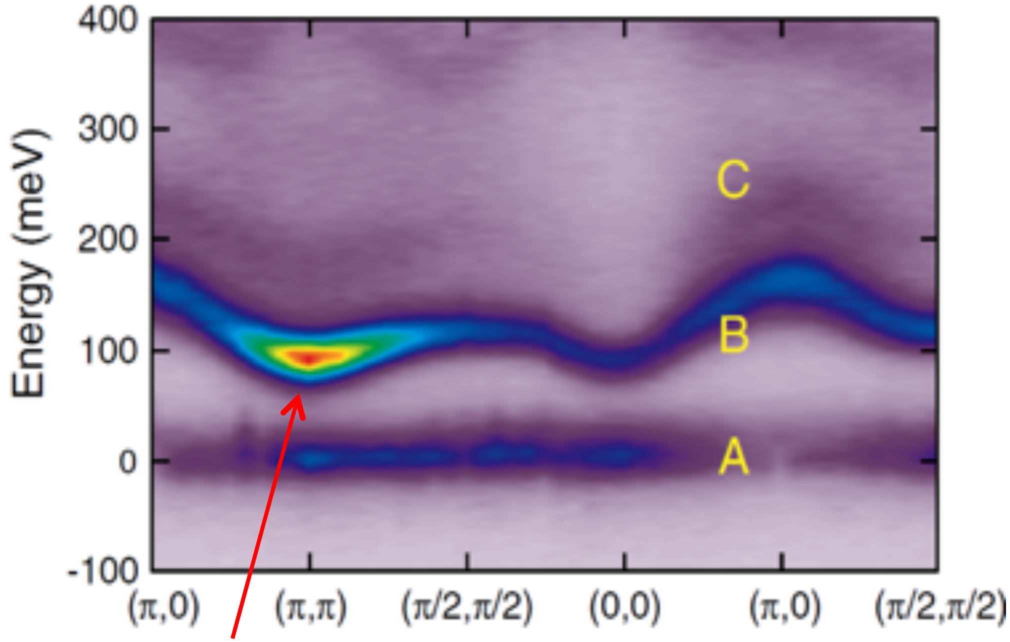
BJ Kim, GJ, et al, PRL (2012)

Magnetism of $Sr_3Ir_2O_7$



• *c* -axis AF (contrary to 214)

RIXS data

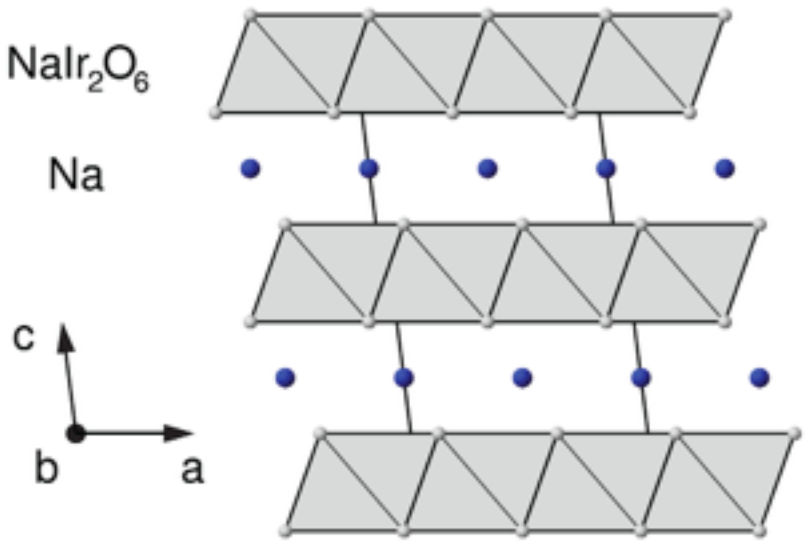


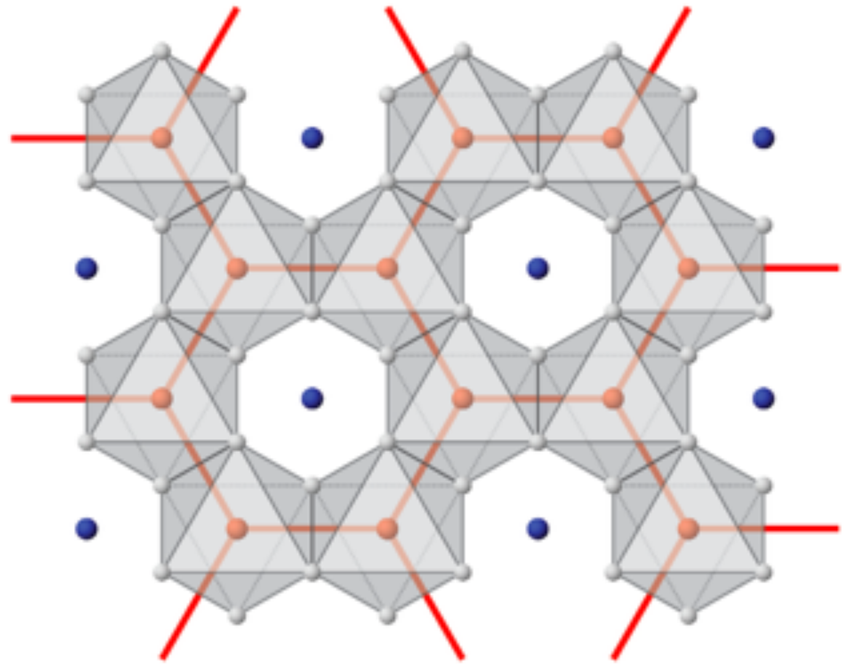
• Large magnon gap,
Ising like spin dynamics

BJ Kim, GJ, et al, PRL (2012)

- single layer Sr_2IrO_4 : isotropic, nearly Heisenberg
- double layer $Sr_3Ir_2O_7$: anisotropic, nearly Ising
- hexagonal layered A_2IrO_3 : bond directional Ising

Hexagonal Iridates A_2IrO_3 (A=Na, Li)





Hexagonal Iridates A_2IrO_3 (A=Na, Li)

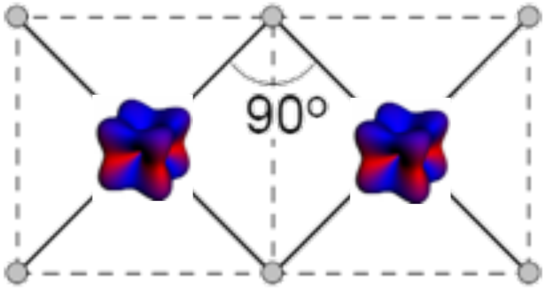
Two theory proposals based

GJ & G.Khaliullin, PRL'09
---
from Mott side:
as a candidate for a Quantum Spin Liquid

A.Shitade, H. Katsura, J. Kunes, XL Qi, SC Zhang, N.Nagaosa, PRL'09
---
from itinerant side:
as a candidate for a Topological Insulator and QSH effect

Superexchange Hamiltonian: 90°-bonds

GJ & G.Khaliullin PRL (2009)

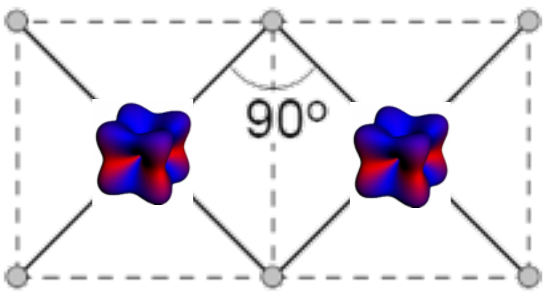


$$\mathcal{H}_{ij}^{(\gamma)} = J S_i^\gamma S_j^\gamma$$

Ising, γ-axis out of plane

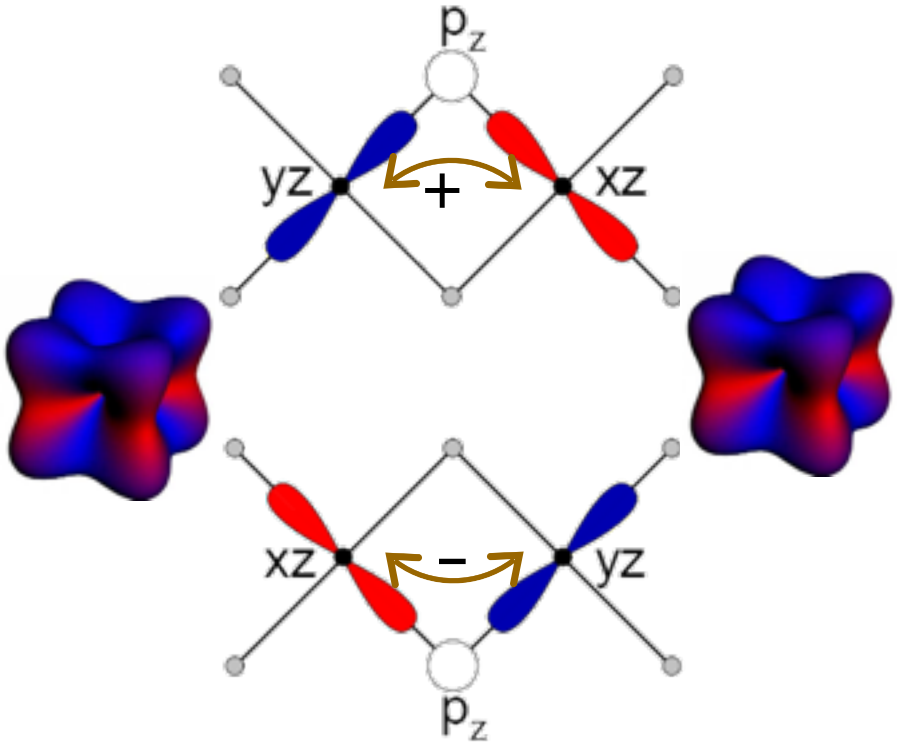
Superexchange Hamiltonian: 90°-bonds

GJ & G.Khaliullin PRL (2009)



$$\mathcal{H}_{ij}^{(\gamma)} = J S_i^{\gamma} S_j^{\gamma}$$

Ising, γ-axis out of plane

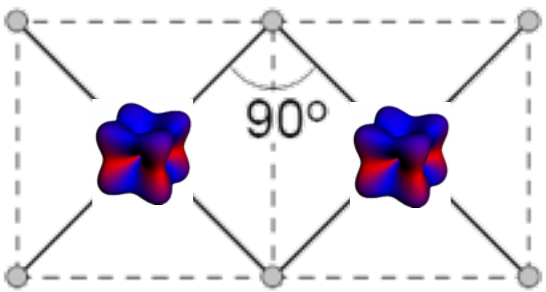


Destructive quantum interference between two channels

Heisenberg term is suppresed

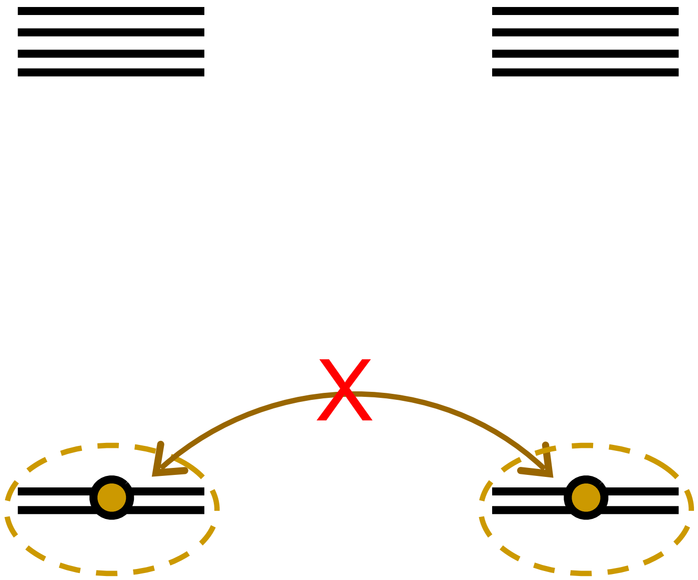
Superexchange Hamiltonian: 90°-bonds

GJ & G.Khaliullin PRL (2009)



$$\mathcal{H}_{ij}^{(\gamma)} = J S_i^\gamma S_j^\gamma$$

Ising, γ-axis out of plane



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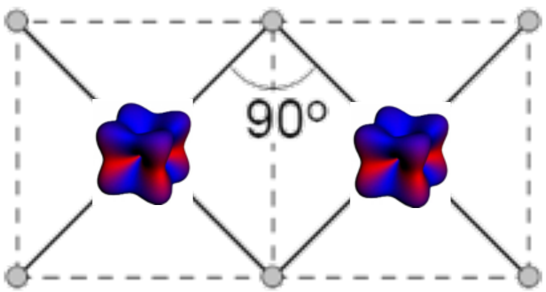

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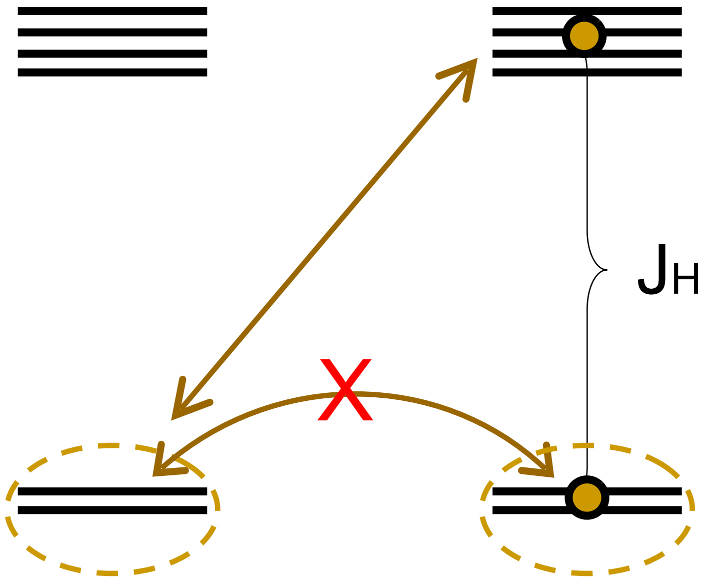
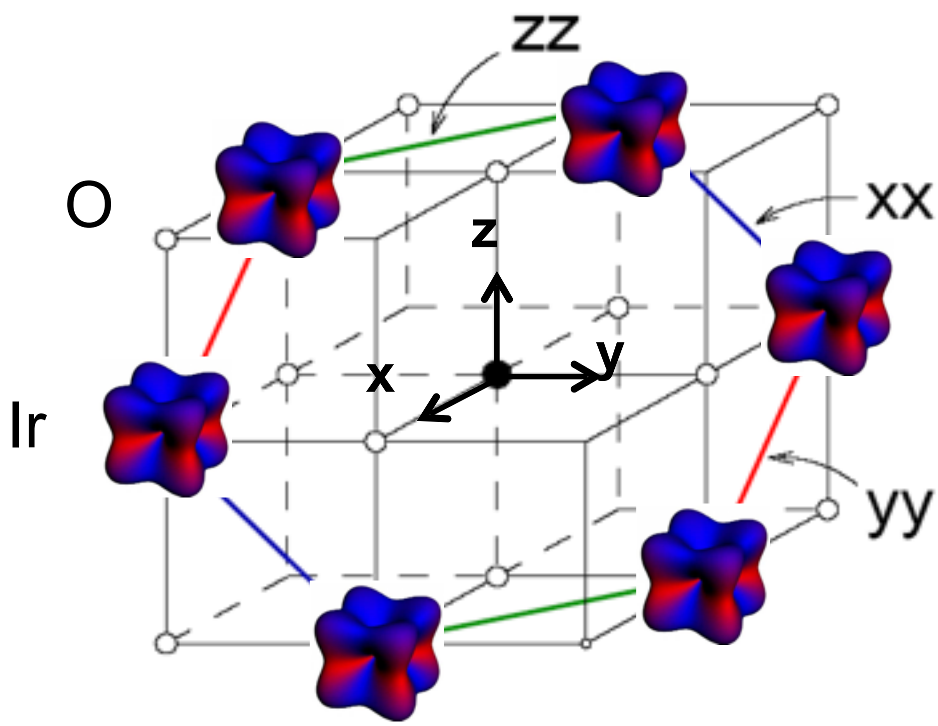
Ising, γ-axis out of plane



Destructive quantum interference between two channels

Heisenberg term is suppresed

Isospins on a Honeycomb layer of A_2IrO_3



$$\mathcal{H}_{ij}^{(\gamma)} = J S_i^\gamma S_j^\gamma$$

Kitaev Model

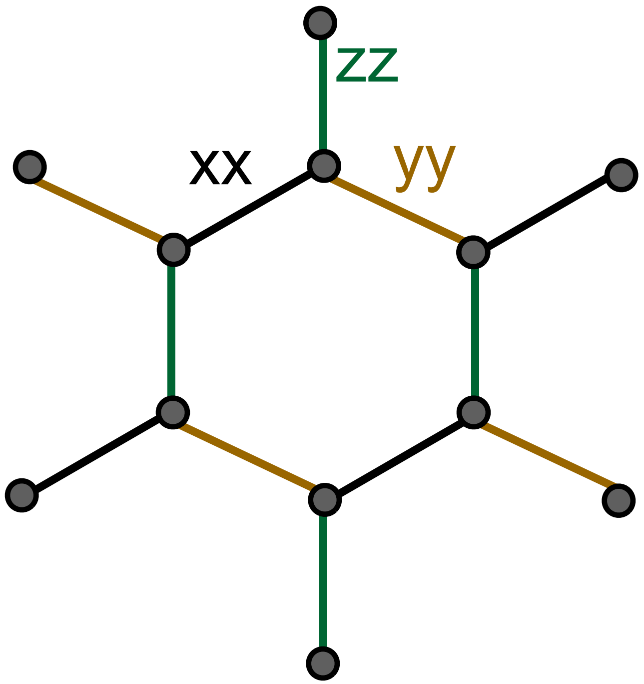
A.Kitaev, Ann. Phys'06

Kitaev Honeycomb Model

A. Kitaev, Ann. Phys'06

$$\mathcal{H}_{ij}^{(\gamma)} = J S_i^\gamma S_j^\gamma$$

- exactly soluble 2D quantum model



Kitaev Honeycomb Model

A. Kitaev, Ann. Phys'06

$$\mathcal{H}_{ij}^{(\gamma)} = J S_i^\gamma S_j^\gamma$$

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- spin liquid ground-state

Kitaev Honeycomb Model

A. Kitaev, Ann. Phys'06

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- Fractional excitations:
  Majorana Fermions, Dirac spectrum
- only NN two-spin correlations
  (Baskaran et al PRL'07)
- Exact dynamic spin structure factor
  (Knolle et al PRL'14)

Kitaev Honeycomb Model

A. Kitaev, Ann. Phys'06

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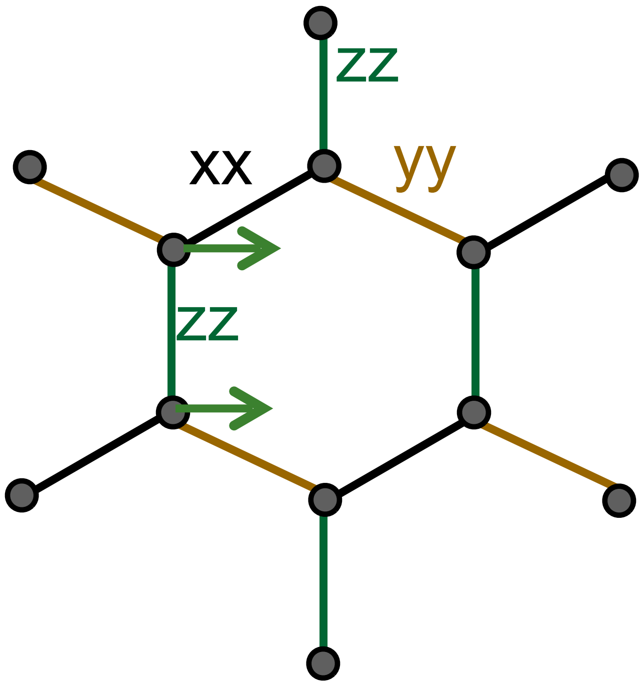

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A. Kitaev, Ann. Phys'06

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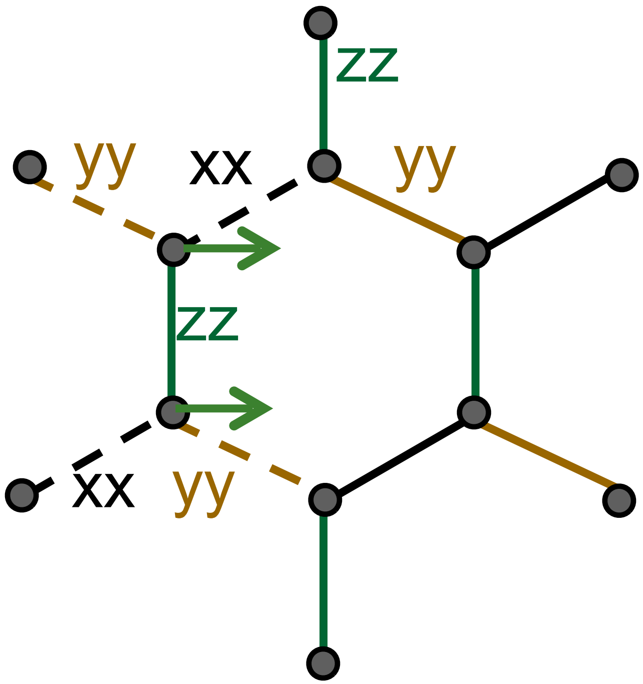
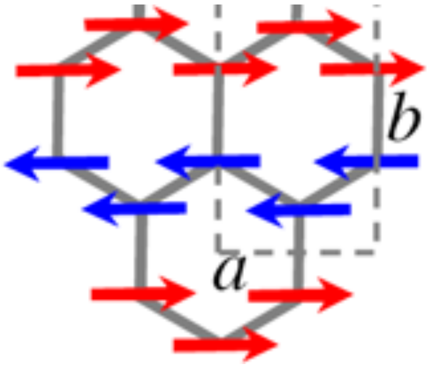


- exactly soluble 2D quantum model
- spin liquid ground-state
- Fractional excitations: Majorana Fermions, Dirac spectrum
- only NN two-spin correlations (Baskaran et al PRL'07)
- Exact dynamic spin structure factor (Knolle et al PRL'14)

Experimental Findings

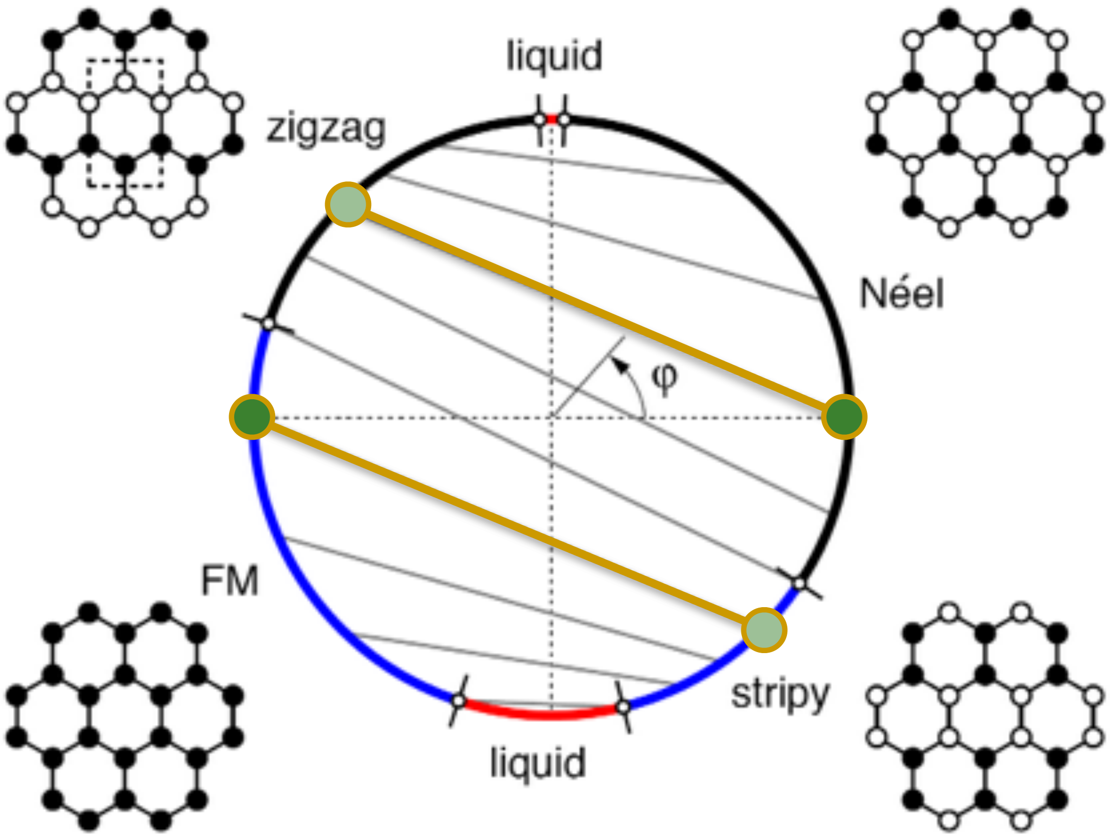
Susceptibility & Thermodynamics:
Singh and Gegenwart, PRB'10; Singh et al PRL'12
Ordering ~ 15K in both Na_2IrO_3 (θ=-125K) & Li_2IrO_3 (θ =-33K)

Zig-Zag order in Na_2IrO_3
XRS : Liu et al, PRB'11
INS: Choi, Coldea et al, PRL'12
N&XR Difraction: Ye et al PRB'12



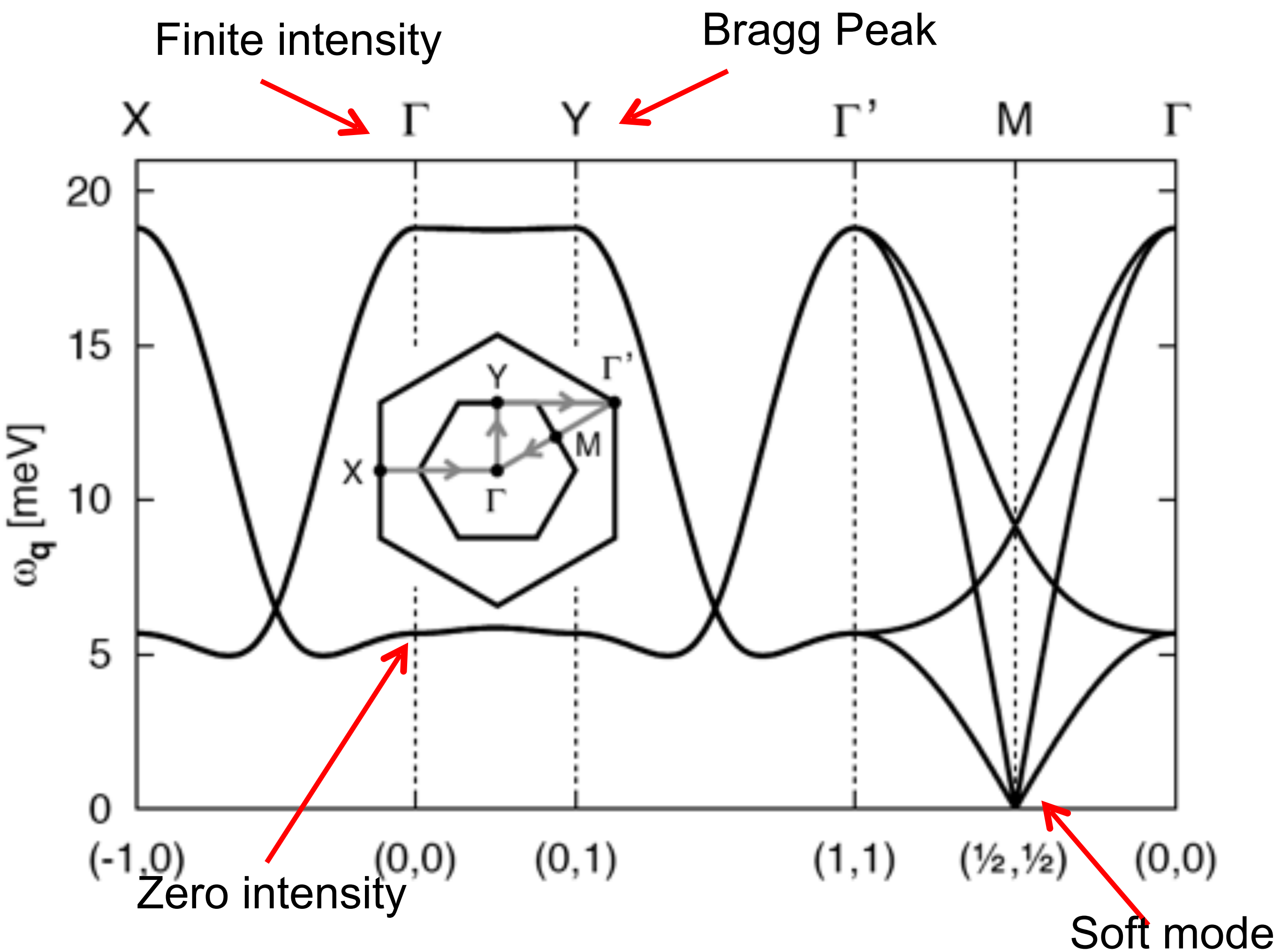
Phase Diagram of Kitaev-Heisenberg Model

$$\mathcal{H}_{ij}^{(\gamma)} = 2K\ S_i^\gamma S_j^\gamma + J\ \mathbf{S}_i \cdot \mathbf{S}_j \qquad K = A\sin\varphi \qquad J = A\cos\varphi$$



J.Chaloupka, GJ & G.Khaliullin PRL (2013)

Spin-wave dispersions of Zigzag phase

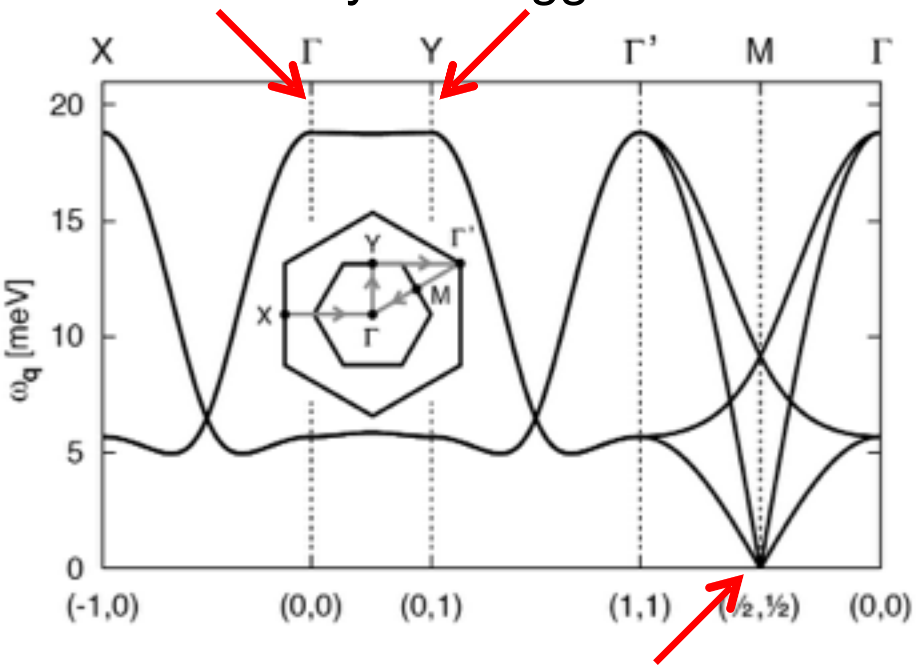


Spin-wave dispersions of Zigzag phase

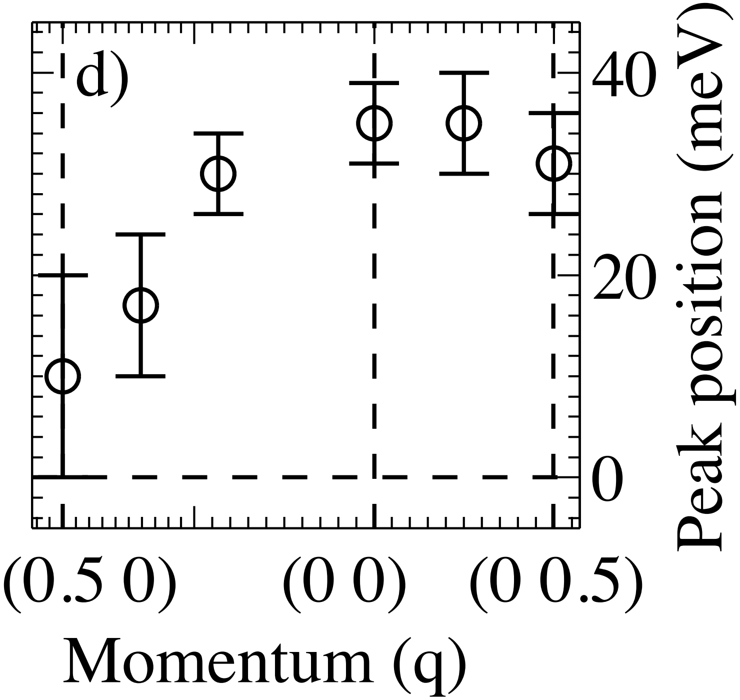
Chaloupka, GJ & Khaliullin PRL 2013

Gretarsson et al, PRB 2013

Finite intensity   Bragg Peak



Soft mode



further neighbor exchange also stabilise zigzag

PHYSICAL REVIEW B **84**, 180407(R) (2011)

Kitaev-Heisenberg-J_2-J_3 model for the iridates A_2IrO_3

Itamar Kimchi[1] and Yi-Zhuang You[1,2]

PRL **108**, 127204 (2012) PHYSICAL REVIEW LETTERS week ending 23 MARCH 2012

Spin Waves and Revised Crystal Structure of Honeycomb Iridate Na_2IrO_3

S. K. Choi,[1] R. Coldea,[1] A. N. Kolmogorov,[2] T. Lancaster,[1,*] I. I. Mazin,[3] S. J. Blundell,[1] P. G. Radaelli,[1] Yogesh Singh,[4,5] P. Gegenwart,[4] K. R. Choi,[6] S.-W. Cheong,[6,7] P. J. Baker,[8] C. Stock,[8] and J. Taylor[8]

PRL **108**, 127203 (2012) PHYSICAL REVIEW LETTERS week ending 23 MARCH 2012

Relevance of the Heisenberg-Kitaev Model for the Honeycomb Lattice Iridates A_2IrO_3

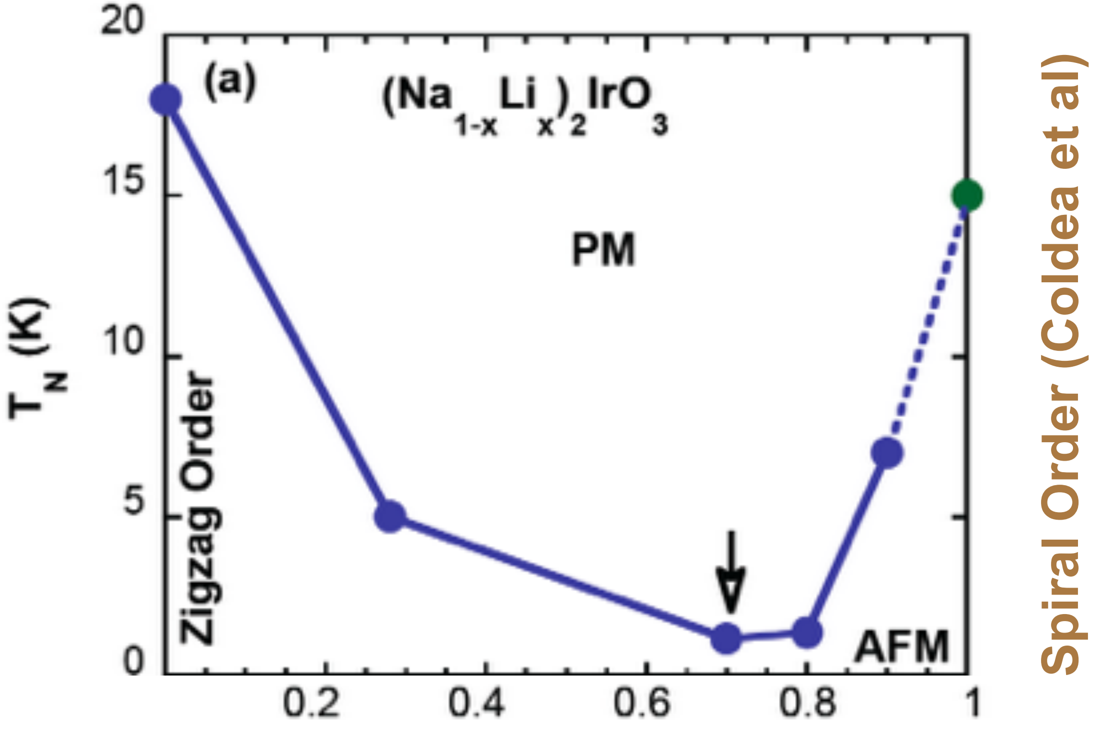
Yogesh Singh,[1,2] S. Manni,[2] J. Reuther,[3,4] T. Berlijn,[5,6] R. Thomale,[7] W. Ku,[5,6] S. Trebst,[8] and P. Gegenwart[2]

RAPID COMMUNICATIONS

PHYSICAL REVIEW B **88**, 220414(R) (2013)

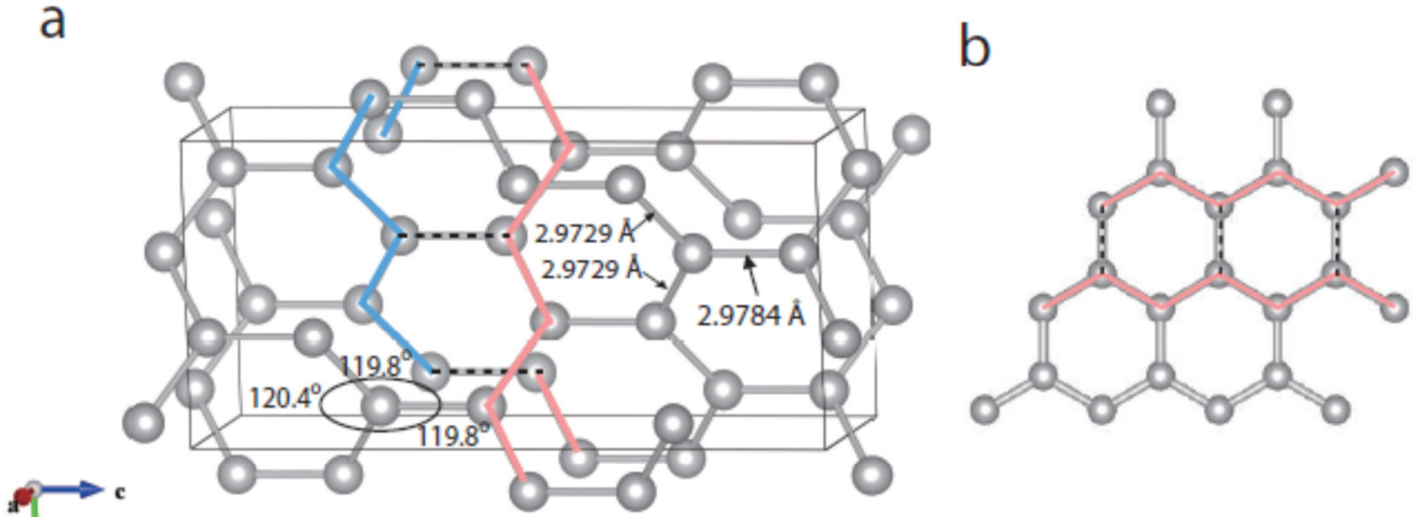
Evolution of magnetism in the single-crystal honeycomb iridates $(Na_{1-x}Li_x)_2IrO_3$

G. Cao,[1] T. F. Qi,[1] L. Li,[1] J. Terzic,[1] V. S. Cao,[1,2] S. J. Yuan,[1,3] M. Tovar,[1] G. Murthy,[1] and R. K. Kaul[1]



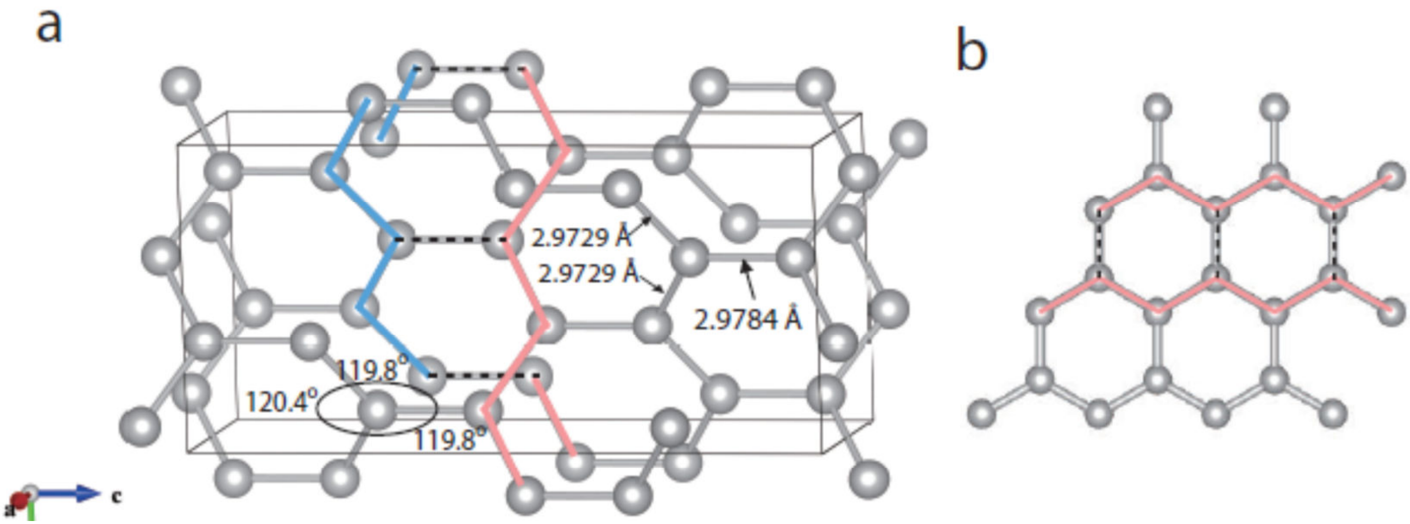
Hyper-Honeycomb β-Li_2IrO_3

T. Takayama et al, PRL'15

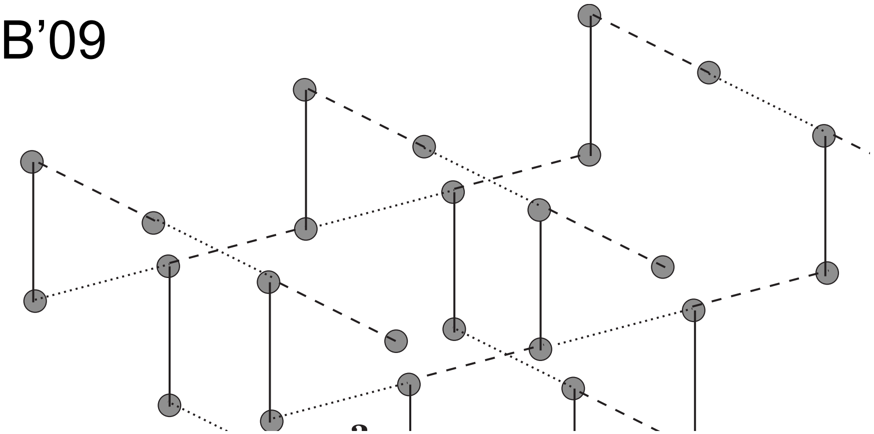


Hyper-Honeycomb β-Li_2IrO_3

T. Takayama et al, PRL'15

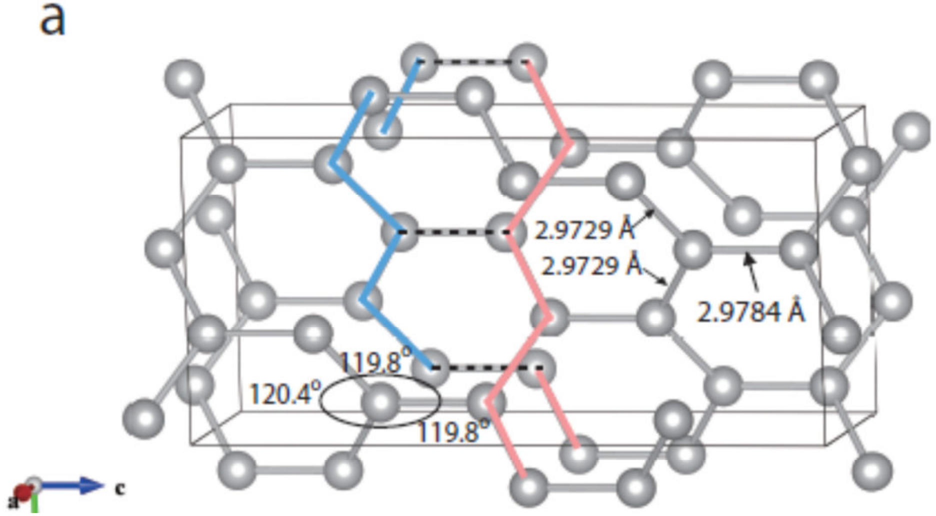


S.Mandal and N.Surendran, PRB'09

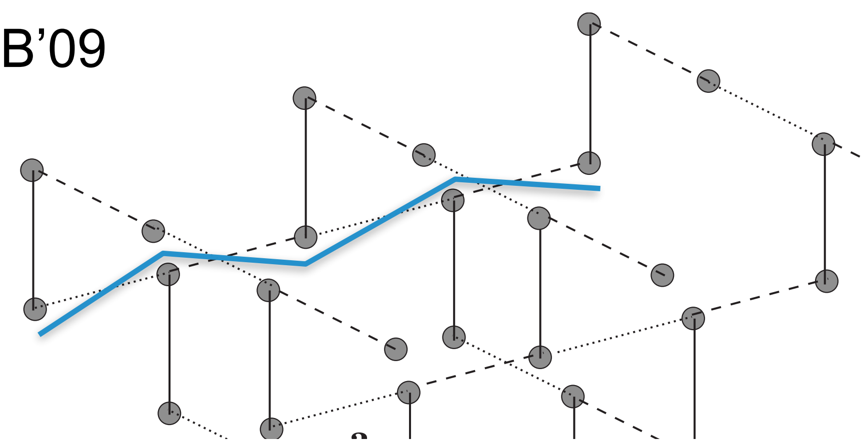


Hyper-Honeycomb β-Li_2IrO_3

T. Takayama et al, PRL’15

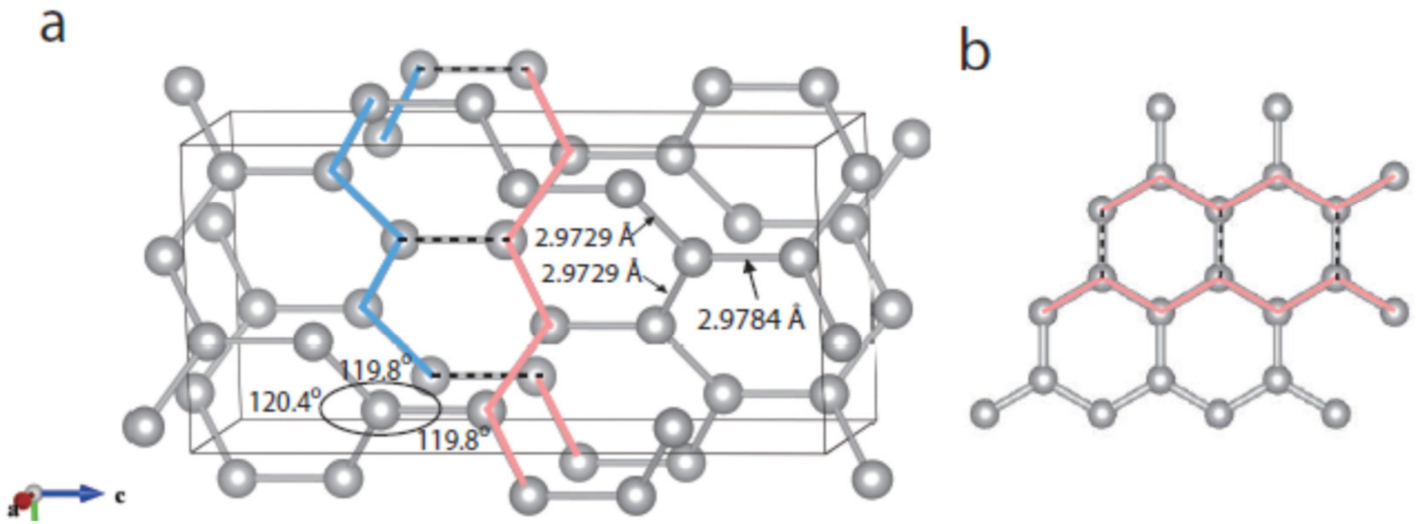


S.Mandal and N.Surendran, PRB’09

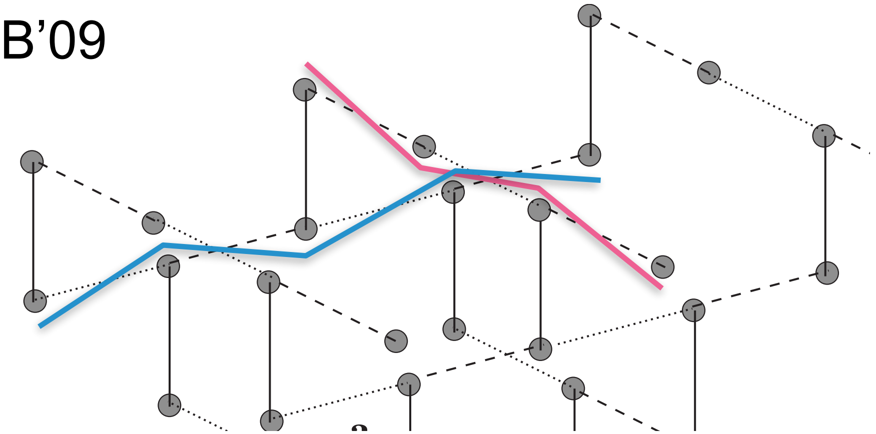


Hyper-Honeycomb β-Li_2IrO_3

T. Takayama et al, PRL'15



S.Mandal and N.Surendran, PRB'09

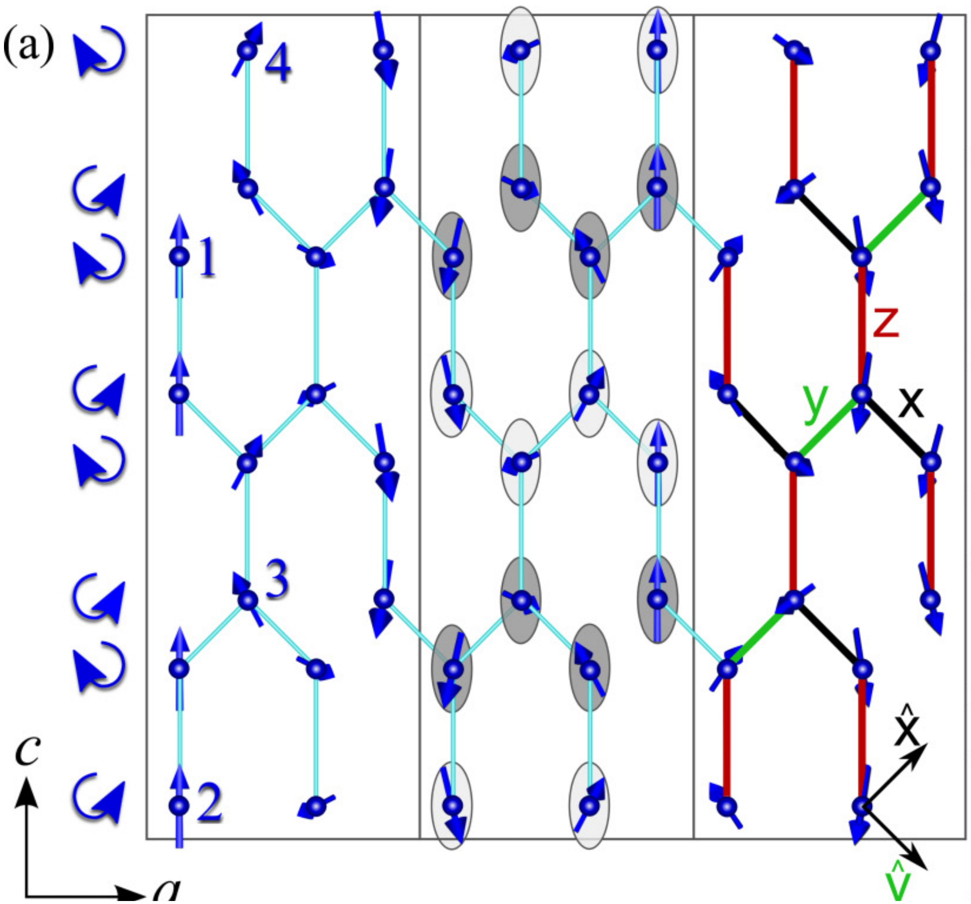


PHYSICAL REVIEW B **90**, 205116 (2014)

Unconventional magnetic order on the hyperhoneycomb Kitaev lattice in β-Li_2IrO_3: Full solution via magnetic resonant x-ray diffraction

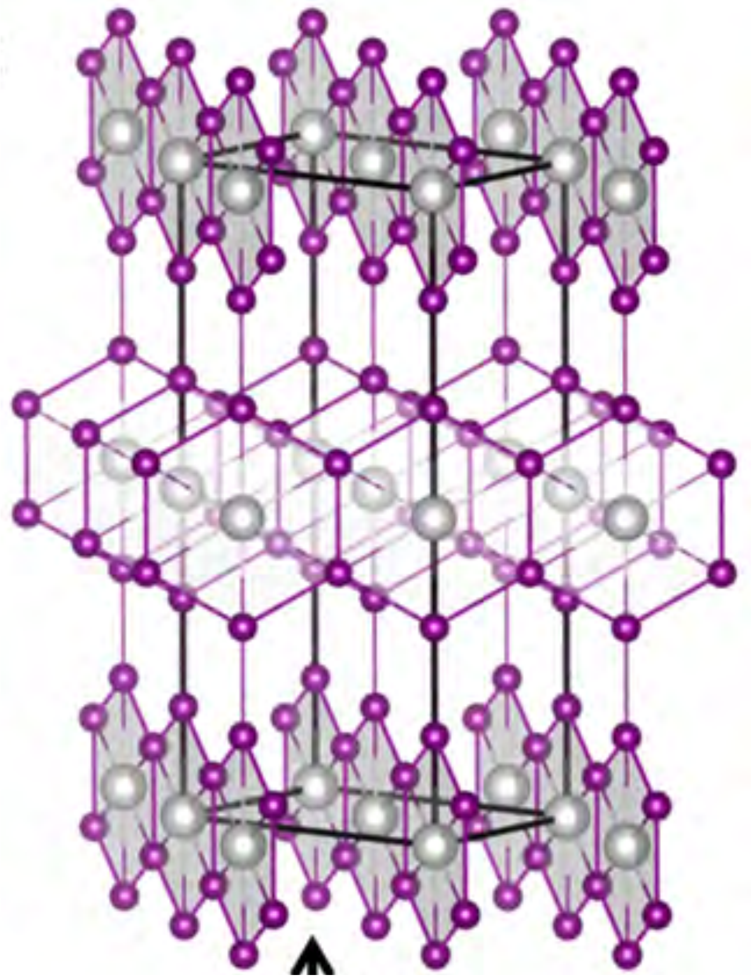
A. Biffin,[1] R. D. Johnson,[1] Sungkyun Choi,[1] F. Freund,[2] S. Manni,[2] A. Bombardi,[3] P. Manuel,[4] P. Gegenwart,[2] and R. Coldea[1]

(a)



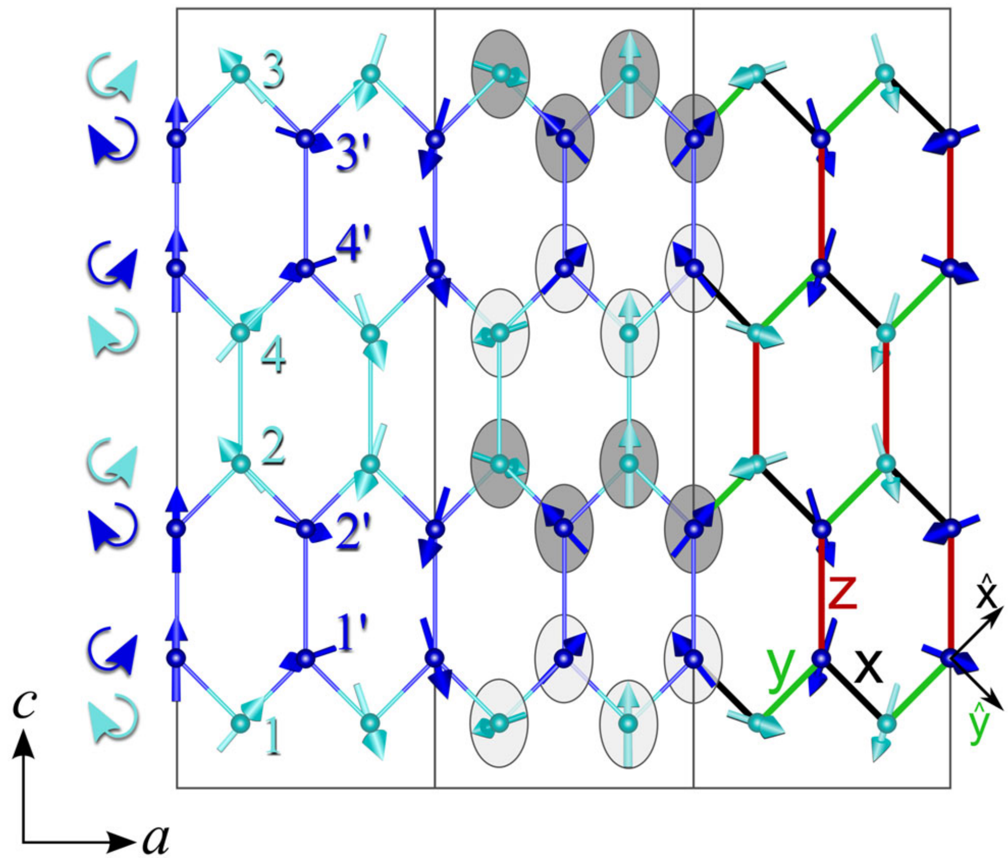
Harmonic-Honeycomb γ-Li_2IrO_3

Modic et al, Nature Comm.'15



Noncoplanar and Counterrotating Incommensurate Magnetic Order Stabilized by Kitaev Interactions in γ-Li_2IrO_3

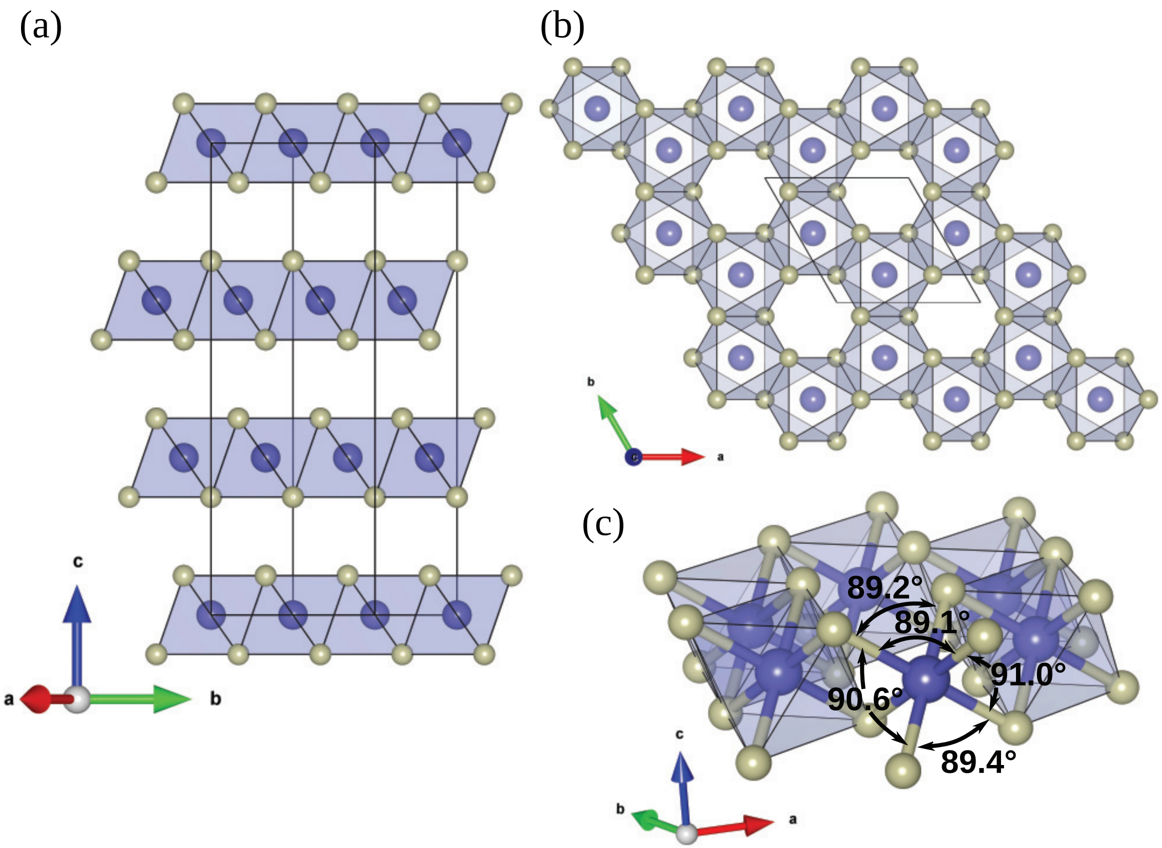
A. Biffin,[1] R. D. Johnson,[1] I. Kimchi,[2] R. Morris,[1] A. Bombardi,[3] J. G. Analytis,[2,4] A. Vishwanath,[2,4] and R. Coldea[1]



PHYSICAL REVIEW B **90**, 041112(R) (2014)

α-$RuCl_3$: A spin-orbit assisted Mott insulator on a honeycomb lattice

K. W. Plumb,[1] J. P. Clancy,[1] L. J. Sandilands,[1] V. Vijay Shankar,[1] Y. F. Hu,[2] K. S. Burch,[1,3]
Hae-Young Kee,[1,4] and Young-June Kim[1,*]



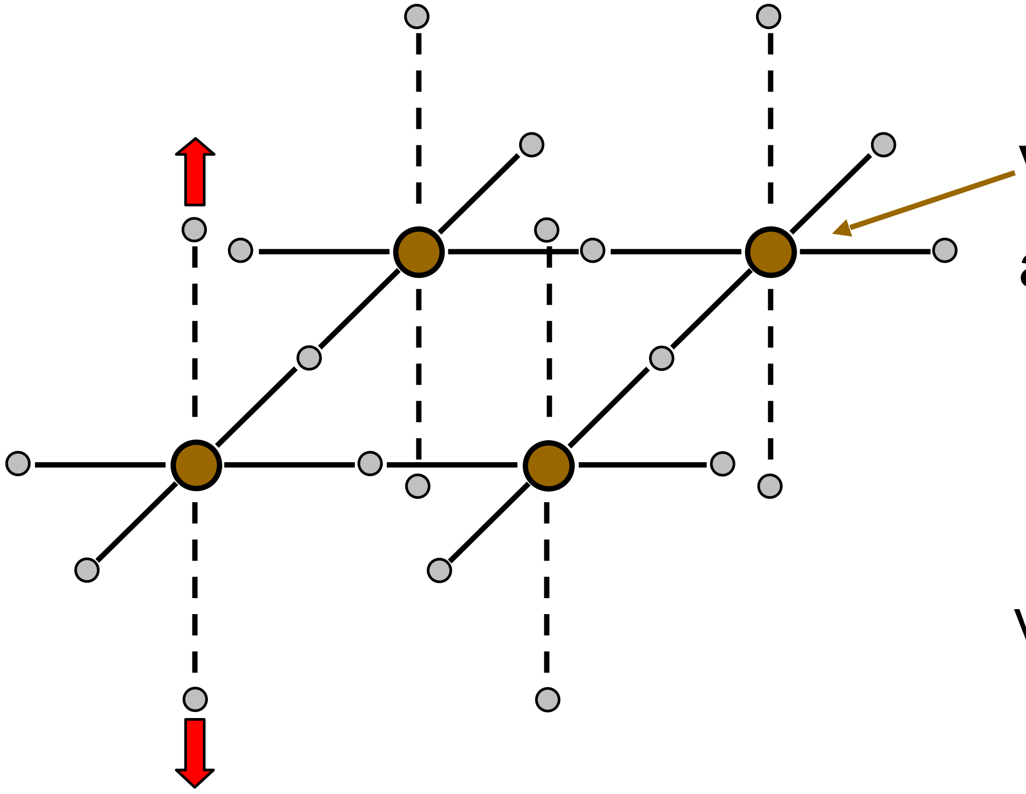
Optimistic Outlook

« IL EST BON DE SAVOIR QUE L'UTOPIE N'EST JAMAIS RIEN D'AUTRE QUE LA RÉALITÉ DE DEMAIN ET QUE LA RÉALITÉ D'AUJOURD'HUI ÉTAIT L'UTOPIE D'HIER »

LE CORBUSIER

Crystal structure of Sr_2VO_4

Zhou et al., PRL '07



V^{4+}, $3d^1$

analog of La_2CuO_4 $3d^9$

Tetragonal distortion:
Elongated along c-axis
V-O(1)=1.91A V-O(2)=1.94A

Sr_2VO_4

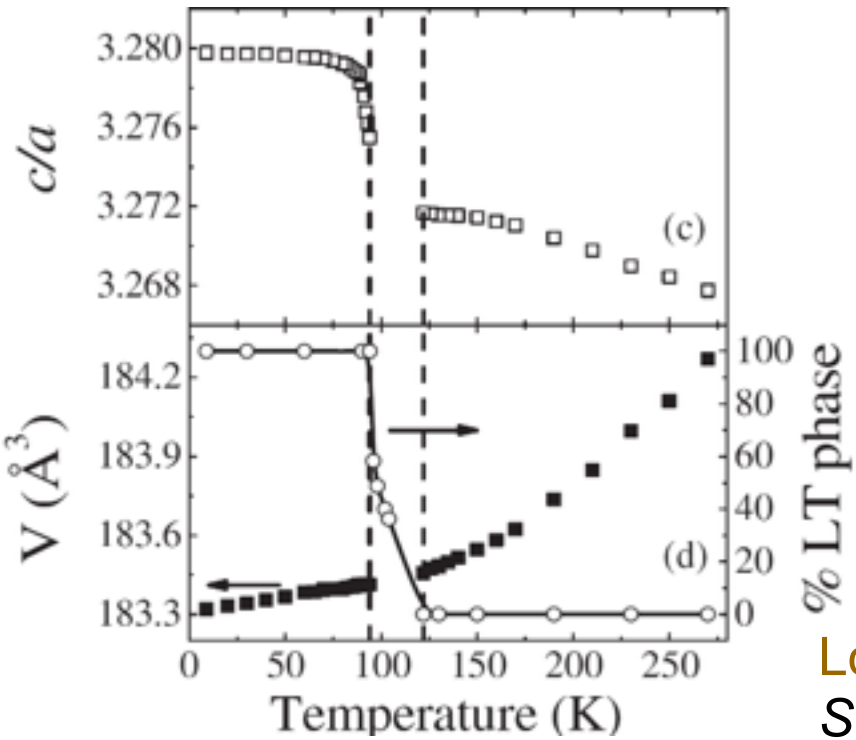
Cyrot et al., ‘90

of 7–5 K. The value of the Néel temperature is very sample dependent. On some samples we have measured a Néel temperature as high as 100 K. We have not been able to observe antiferromagnetism with polarized neutrons. However, we note that

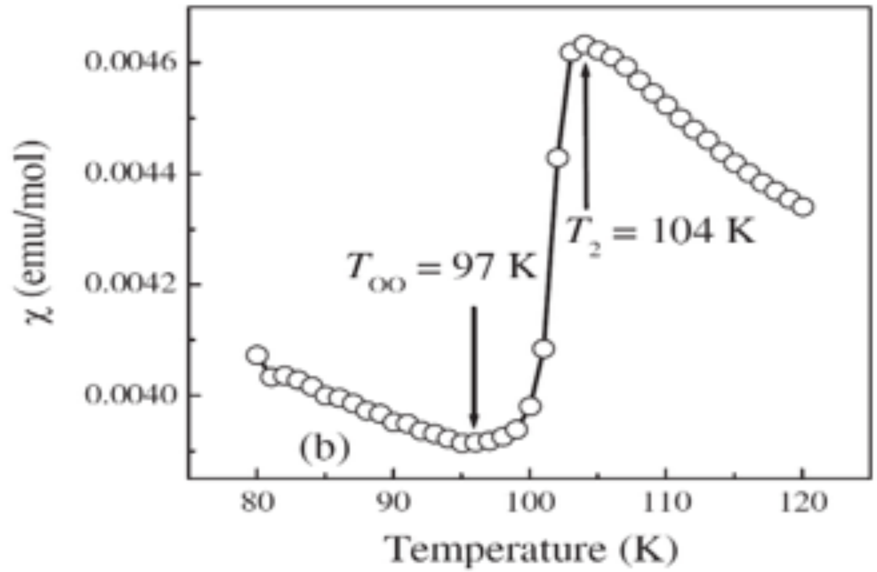
Phase transition in Sr_2VO_4

Zhou et al., PRL '07

Sharp increase of c/a
Structure remains tetragonal

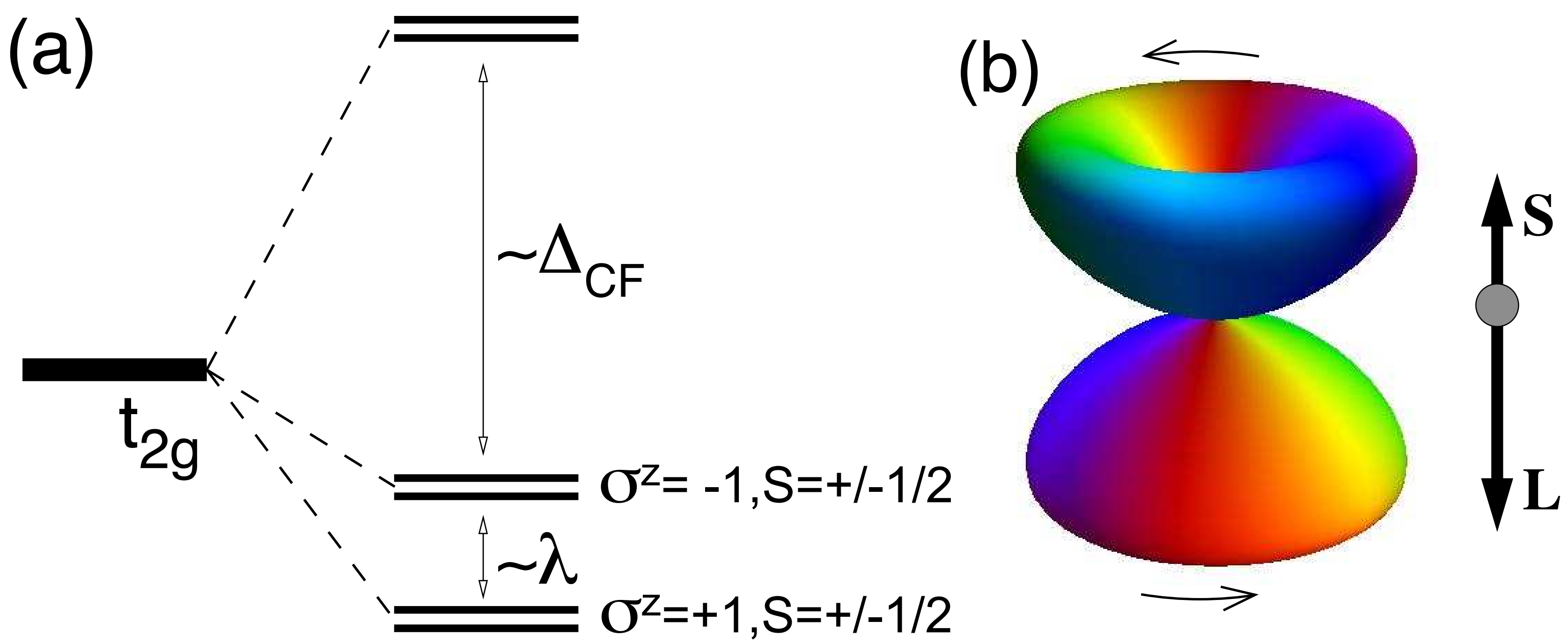


Sharp drop in spin susceptibility



Looks like 1st order magnetic transition…
So far no Bragg peaks seen by neutrons

Local Electronic Structure



Exchange coupling of Kramers doublets

GJ & G.Khaliullin PRL (2010)

$$\mathcal{H} = J \sum_{\langle ij \rangle} (\vec{s}_i \cdot \vec{s}_j + \frac{1}{4})(1 \pm \sigma_i^x)(1 \pm \sigma_j^x) - \frac{\delta}{2} \sum_i \sigma_i^z$$

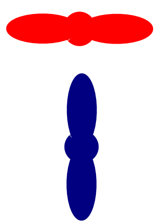
Isospins

Inter-doublet transitions
(non-negative, AFM coupling)

spin-orbit splitting between doublets

$\sigma^x = +1$

$\sigma^x = -1$



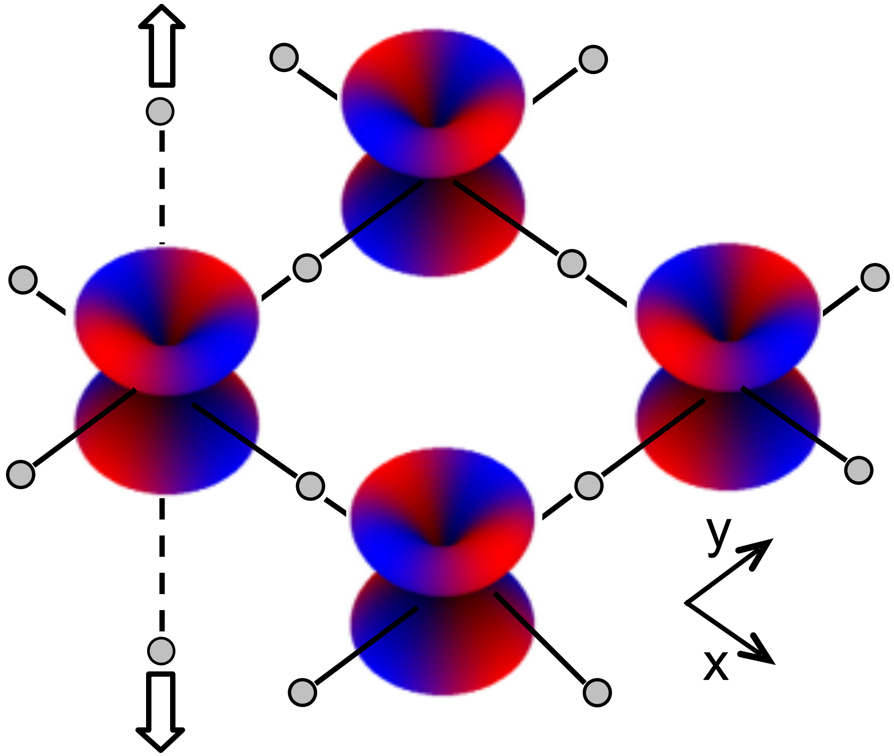
Exchange coupling of Kramers doublets

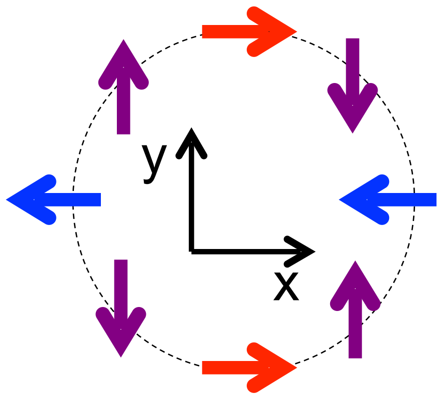
$$\mathcal{H} = J \sum_{\langle ij \rangle} (\vec{s}_i \cdot \vec{s}_j + \frac{1}{4})(1 \pm \sigma_i^x)(1 \pm \sigma_j^x) - \frac{\delta}{2} \sum_i \sigma_i^z$$

nearly zero,
only quantum
energy is gained

SO coupling
becomes relevant

Octupolar order

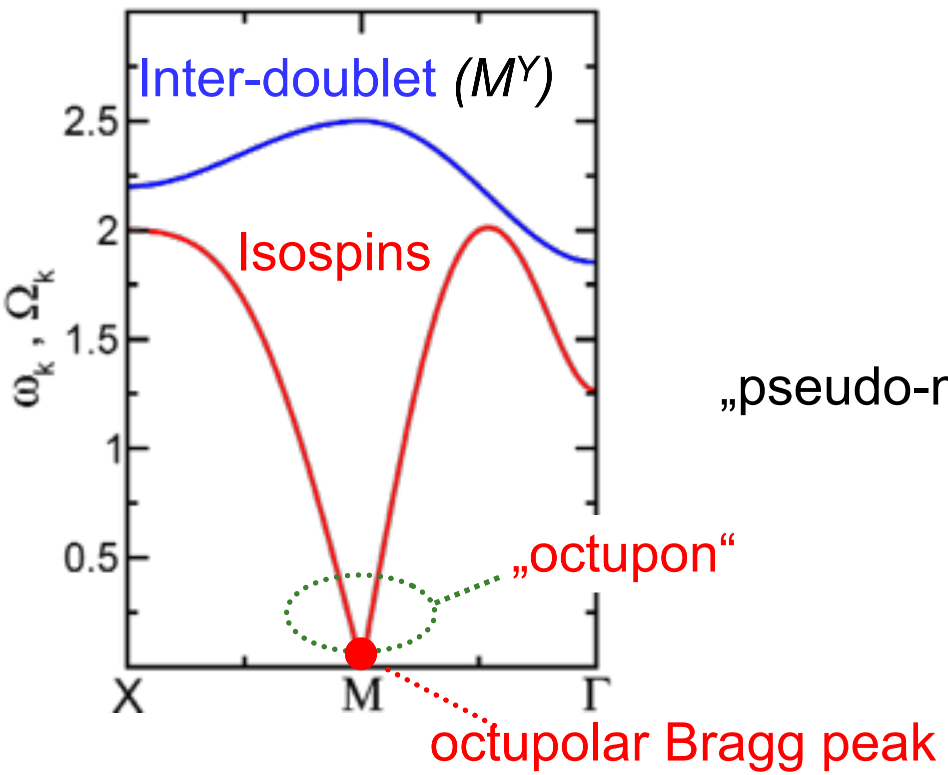


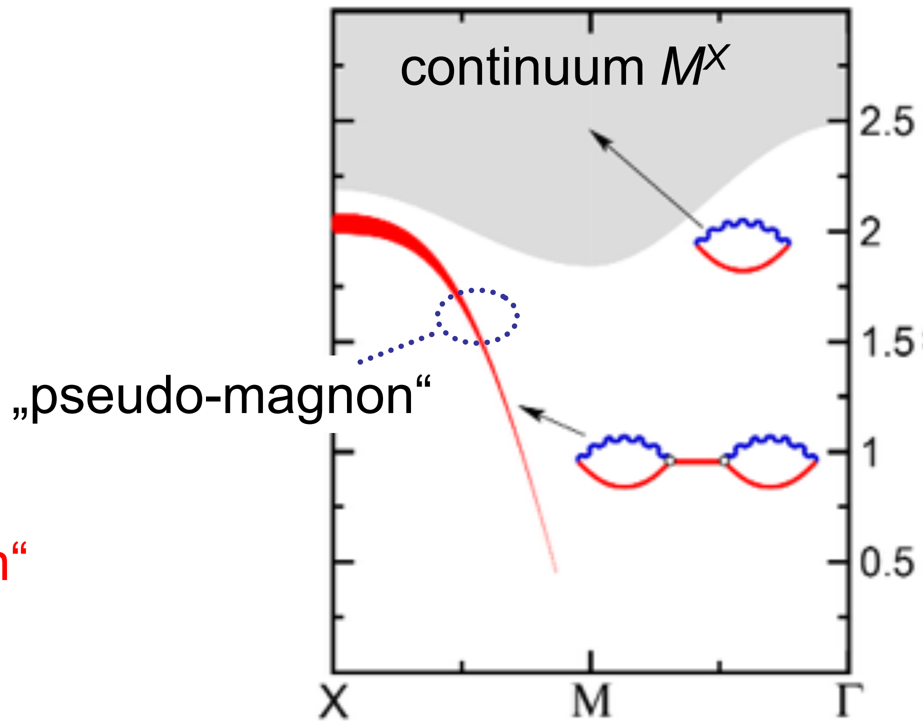


$$\langle \vec{S}_i \rangle = \langle \vec{l}_i \rangle = 0$$

$$\langle S^x[(l^x)^2 - (l^y)^2)] \rangle \neq 0$$

Elementary vs Magnetic Excitations





$$M_i^y = e^{\imath \mathbf{Q}\mathbf{R}_i}(a_i + a_i^\dagger) \quad M_i^z \simeq \imath(1-\kappa)e^{\imath \mathbf{Q}\mathbf{R}_i}(b_i - b_i^\dagger)$$

$$M_i^x \simeq (b_i + b_i^\dagger)(a_i + a_i^\dagger)$$

RAPID COMMUNICATIONS

PHYSICAL REVIEW B **89**, 020402(R) (2014)

Hidden magnetic order in Sr_2VO_4 clarified with μ^+SR

Jun Sugiyama,[1,*] Hiroshi Nozaki,[1] Izumi Umegaki,[1] Wataru Higemoto,[2] Eduardo J. Ansaldo,[3] Jess H. Brewer,[3,4] Hiroya Sakurai,[5] Ting-Hui Kao,[5,6] Hung-Duen Yang,[6] and Martin Månsson[7,8]

In order to elucidate the magnetic ground state of Sr_2VO_4, we have measured muon spin rotation and relaxation (μ^+SR) spectra of a powder sample in the temperature range between 1.8 and 140 K. As a result, we have clarified that the transition at 105 K is not magnetic but structural and/or electric in origin and found the appearance of static antiferromagnetic (AF) order below 8 K. Moreover, the distribution of the internal AF field was found to be very broad, even at the lowest temperature measured. These results are consistent with the formation of an orbital-stripe order with collinear AF order for the magnetic ground state of Sr_2VO_4.

RAPID COMMUNICATIONS

PHYSICAL REVIEW B **89**, 020402(R) (2014)

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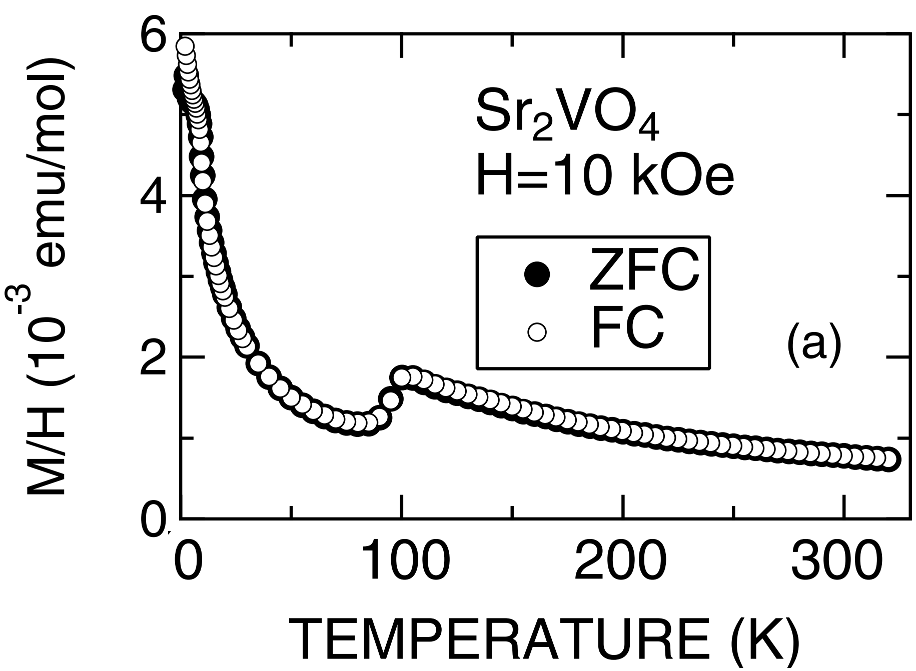
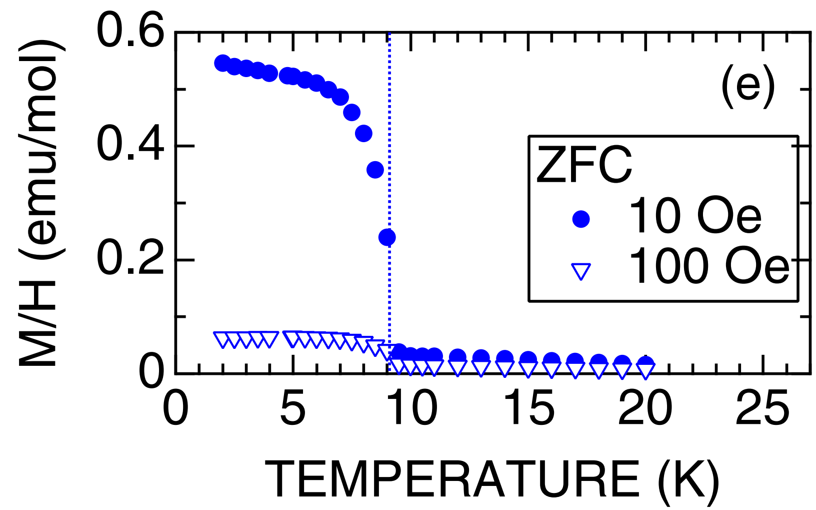


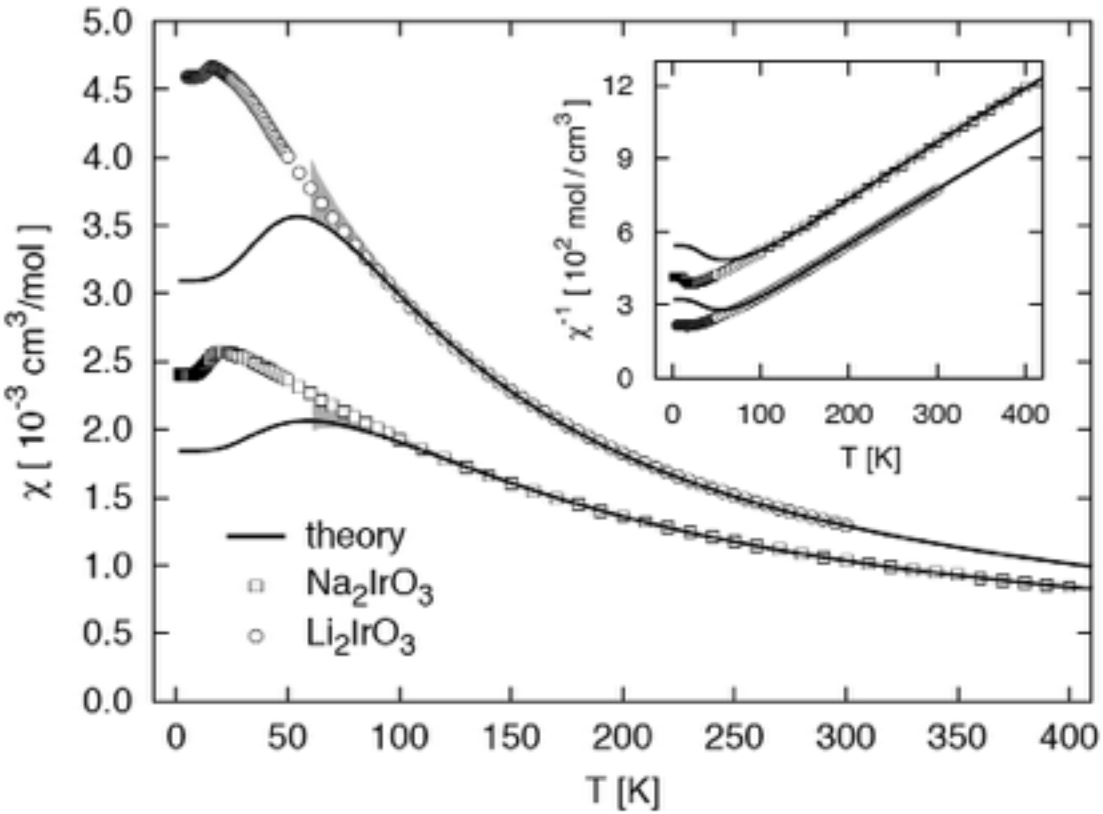


Optimistic Outlook

« IL EST BON DE SAVOIR QUE L'UTOPIE N'EST JAMAIS RIEN D'AUTRE QUE LA RÉALITÉ DE DEMAIN ET QUE LA RÉALITÉ D'AUJOURD'HUI ÉTAIT L'UTOPIE D'HIER »

LE CORBUSIER

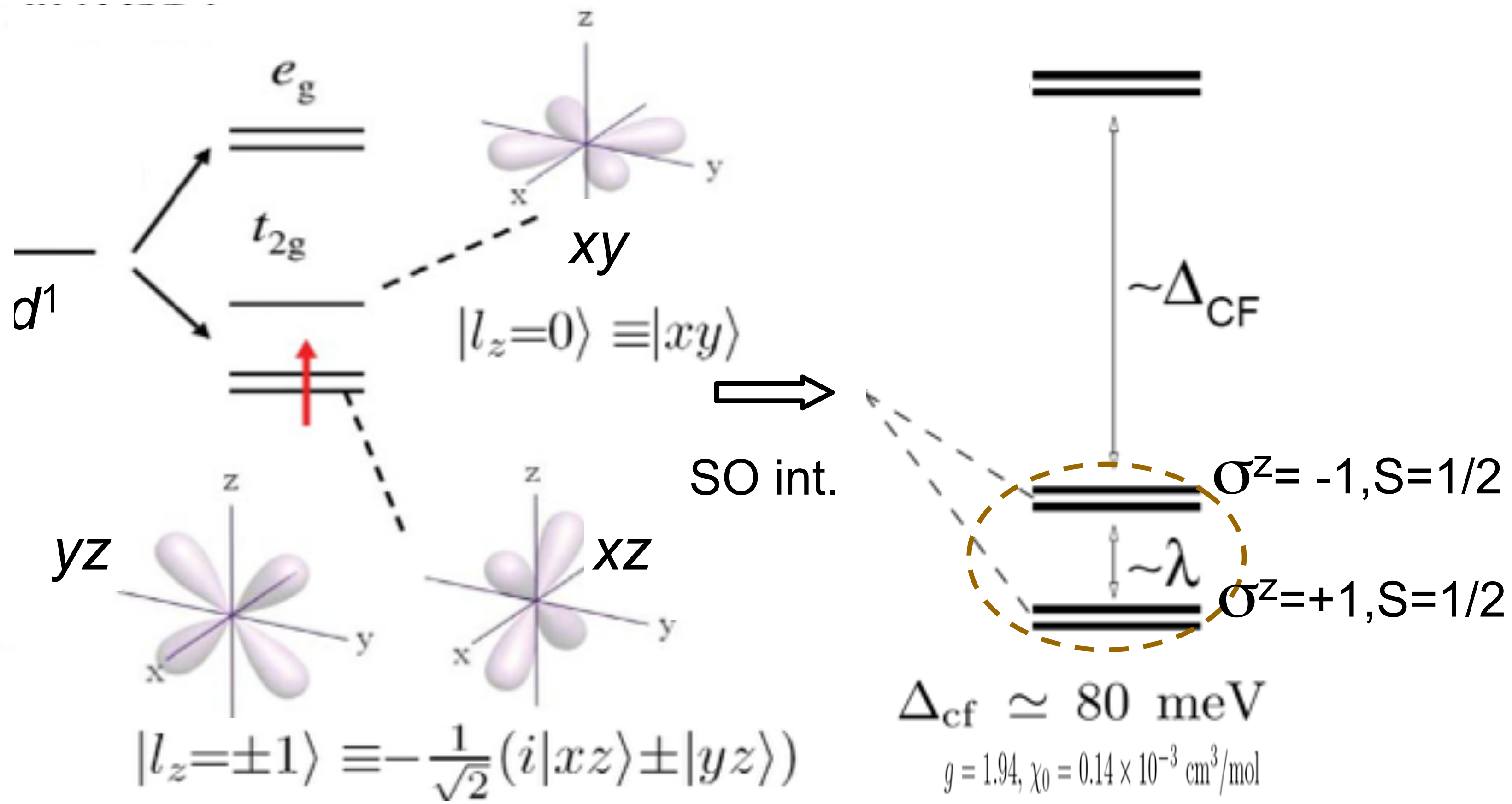
Susceptibly of Zigzag phase



$2K = 21.0\text{meV}, J = -4.0\text{meV} \quad g = 1.78 \;\; \chi_0 = 0.16 \times 10^{-3}\text{cm}^3/\text{mol}$

$2K = 15.7\text{meV}, J = -5.3\text{meV} \quad g = 1.94, \; \chi_0 = 0.14 \times 10^{-3} \; \text{cm}^3/\text{mol}$

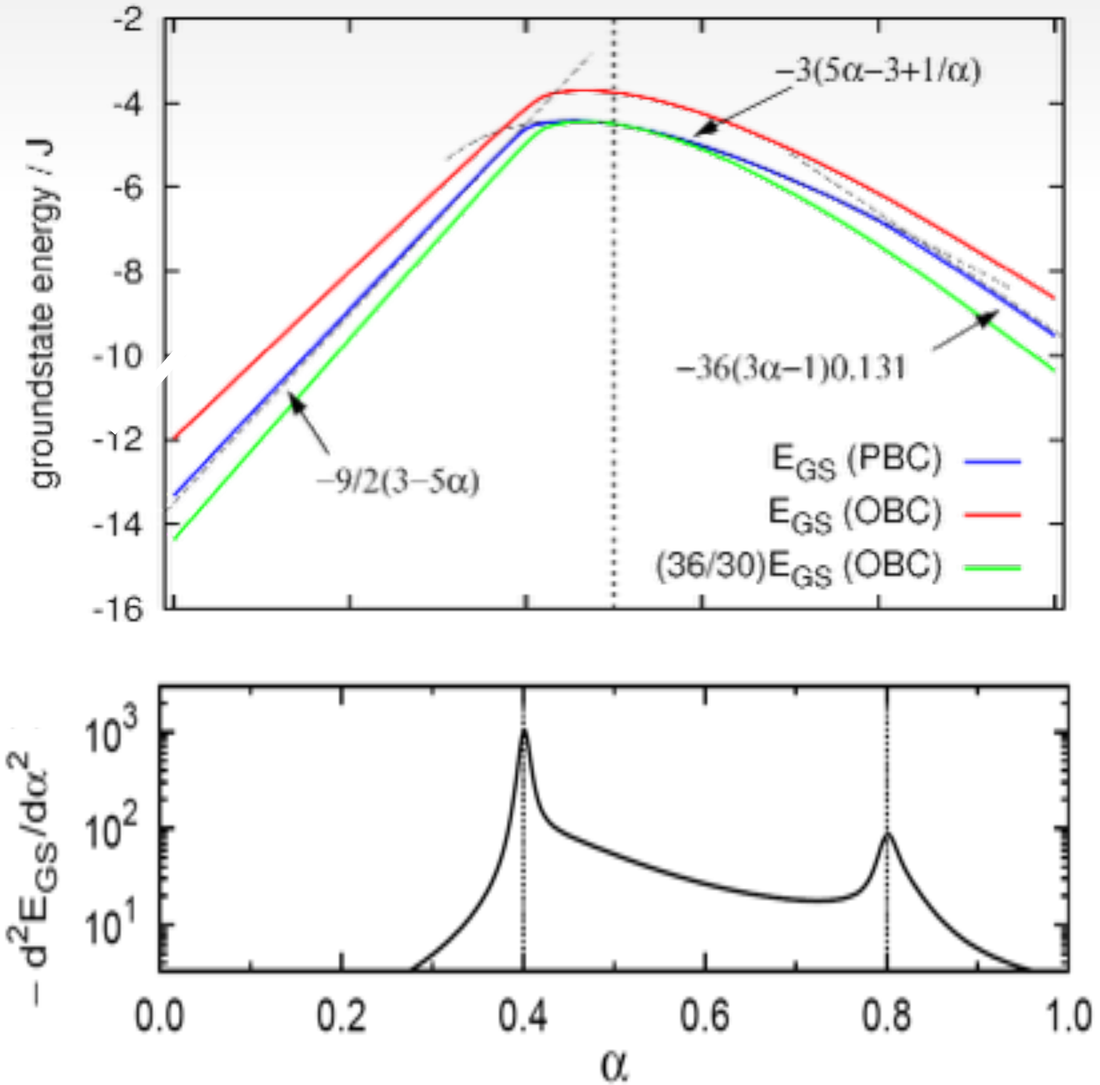
Local Electronic Structure



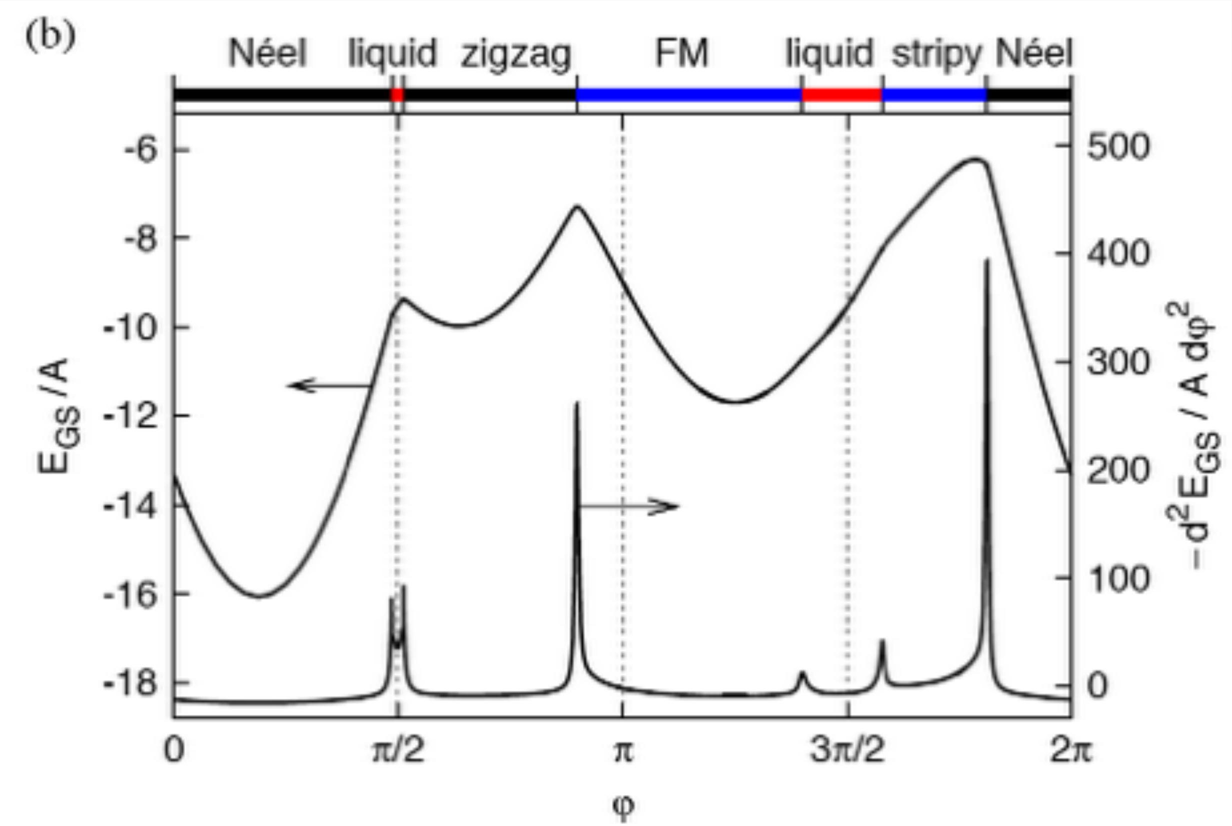
Tetragonal Field: Low energy quadruplet remains active

Spin-orbit: Quadruplet is split into Kramers doublets

Exact Diagonalization Results



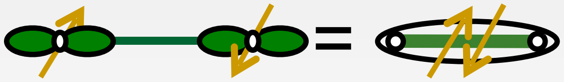
Exact Diagonalization Results



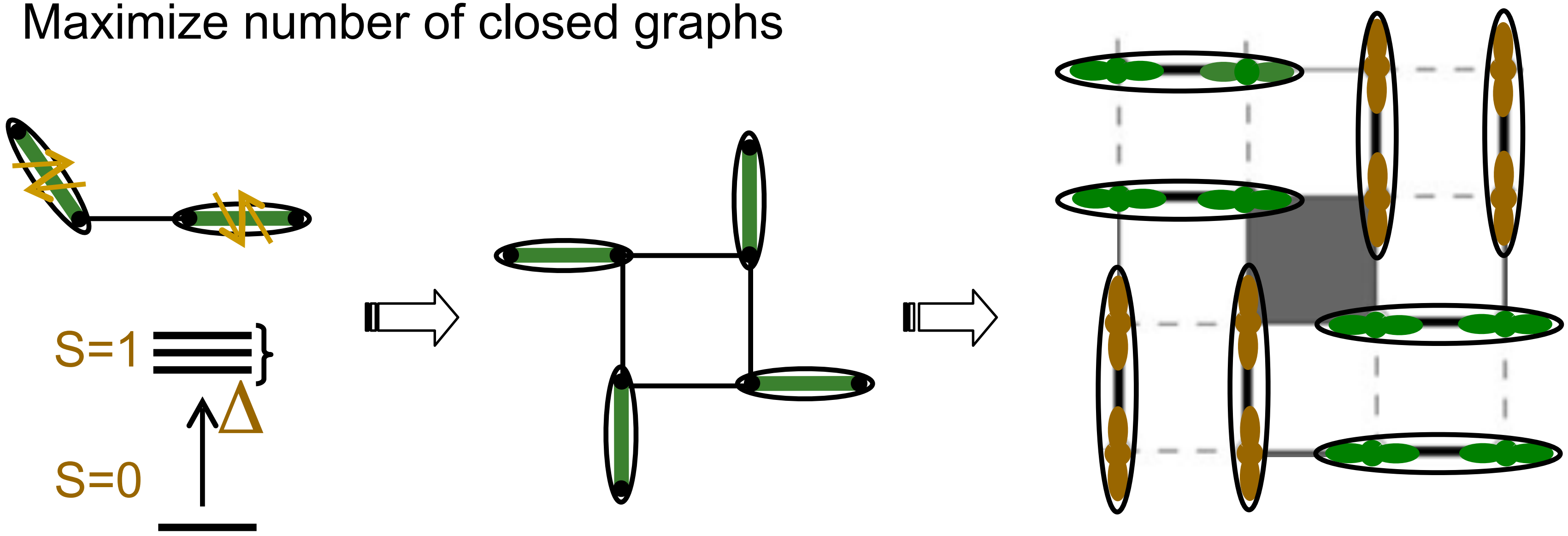
Dimer manifold & Valence bond crystal

GJ & D.Ivanov, PRB ‘07

- Spin-singlet on dimer bonds: Spin gap



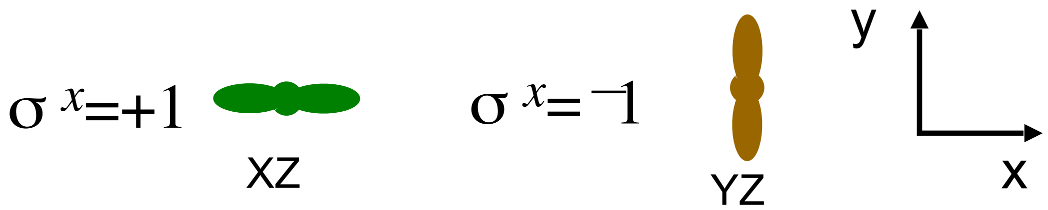
- Extensively degenerate GS manifold: hard-core dimer coverings of square lattice
- Order-out-of-disorder by triplet fluctuations: Maximize number of closed graphs

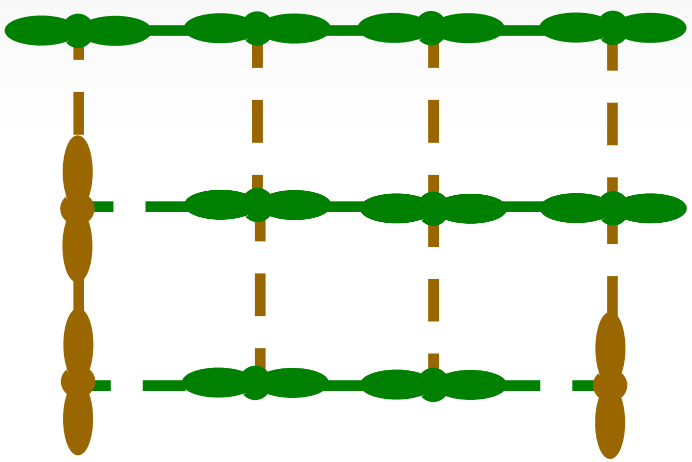


GJ & D.Ivanov, PRB '07

- Dimensionality reduction due to orbitals:
  Fragmentation in clusters of open/closed 1D AFM chains

$$\mathcal{H} = J \sum_{\langle ij \rangle} (\vec{s}_i \cdot \vec{s}_j + \frac{1}{4})(1 \pm \sigma_i^x)(1 \pm \sigma_j^x)$$

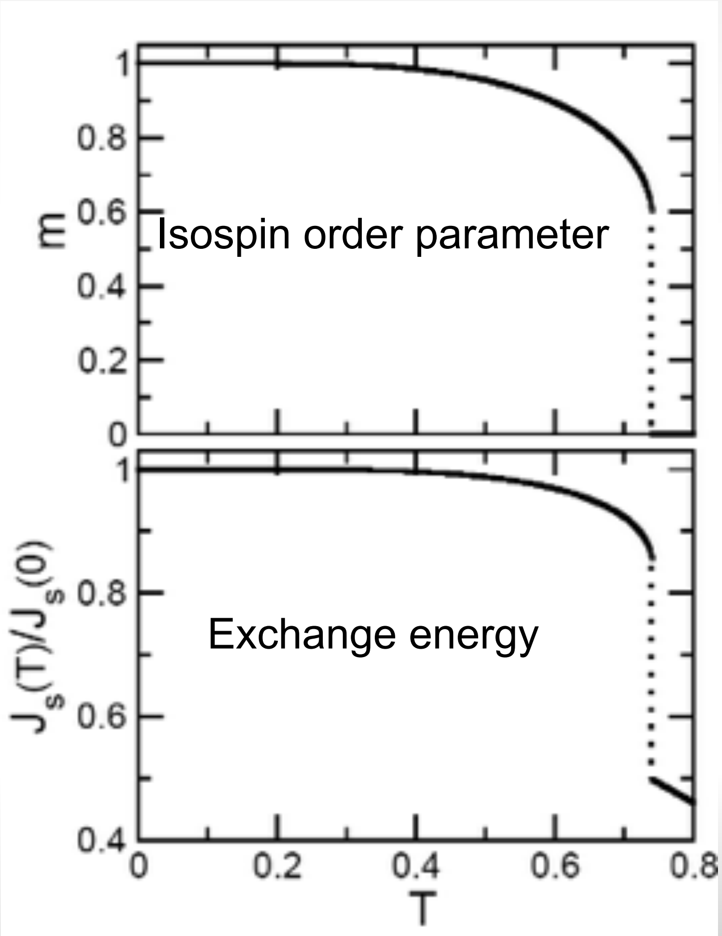




- Classical Neel state ($s_i s_j + 1/4 = 0$) has zero energy
- Shortest chains(=dimers) are favorable -> gain more quantum energy

Finite temperature transition in: Sr_2VO_4

The first order phase transition induced by an interplay between inter- and intra-doublet excitations.



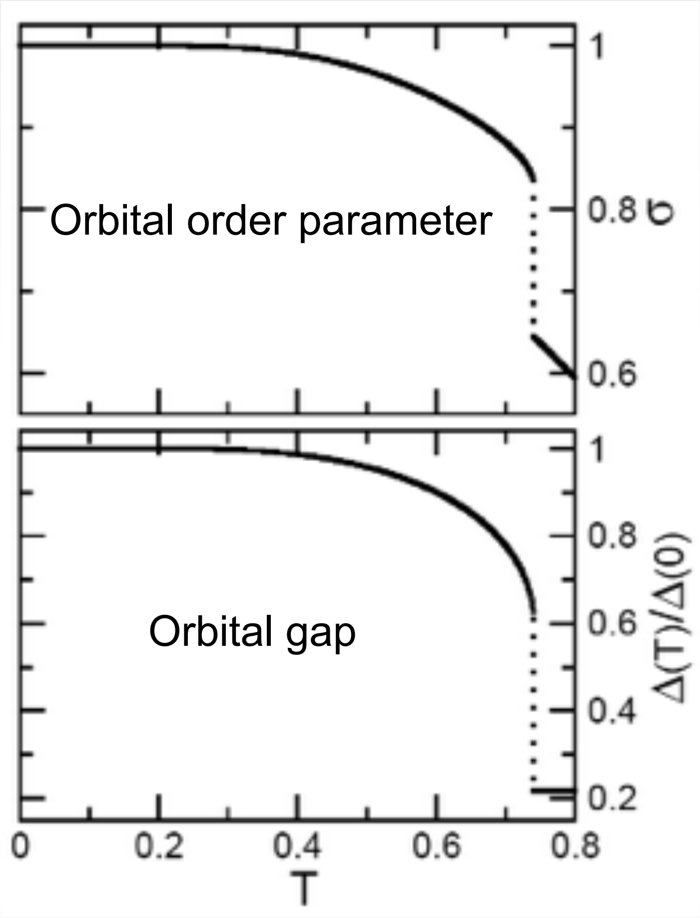
Isospin order parameter and exchange energy jump at the transition

⇓

Pronounced magnetoelastic effects explain an abrupt in-plane contraction of the crystal.

Jahn-Teller coupling acts in the same direction as magnetoelastic one amplifying the effect.

Finite temperature transition

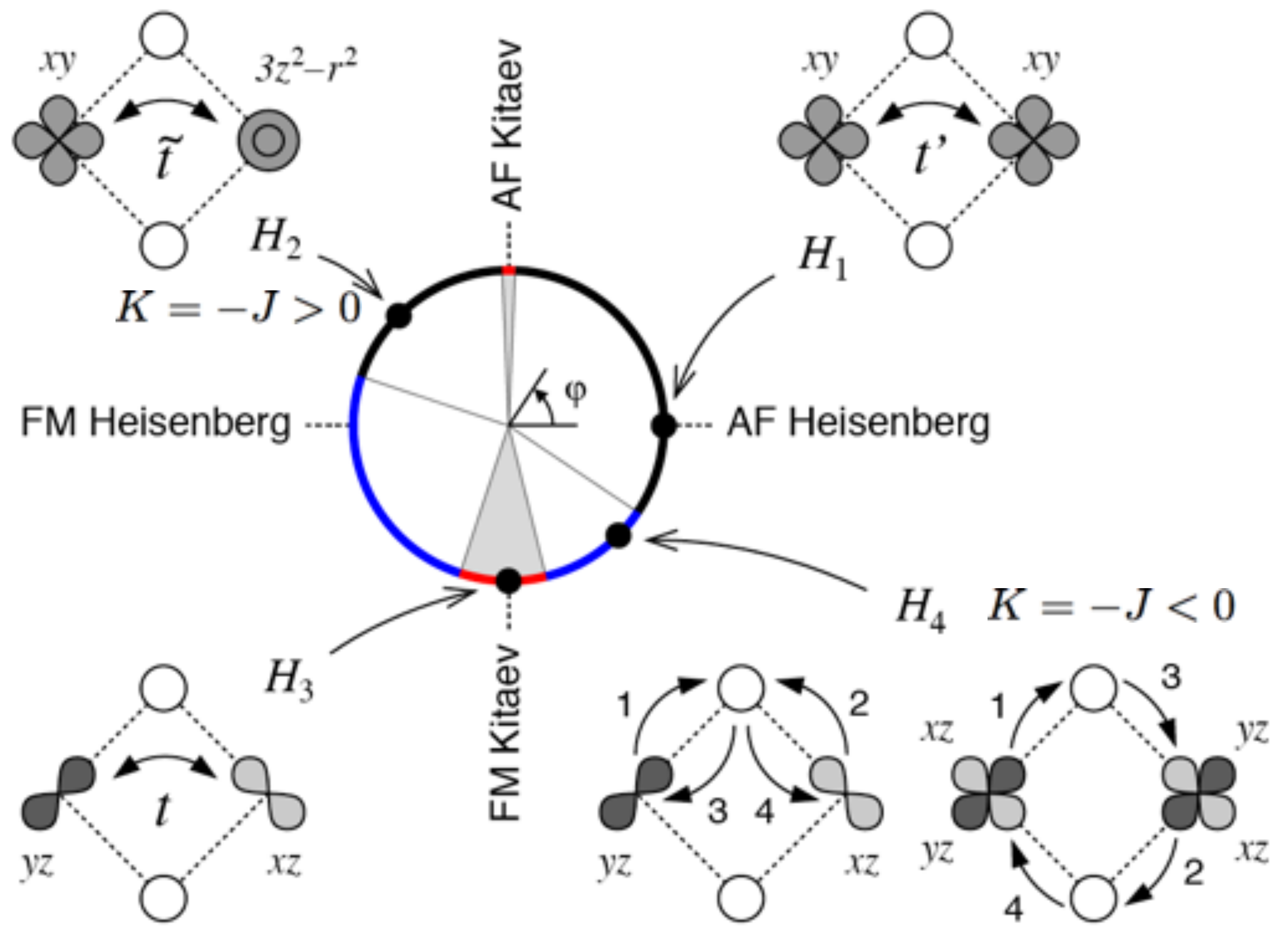


Abrupt enhancement of the population of low energy doublet

Its non-magnetic nature, vanishing g-factors, explains the drop of susceptibility

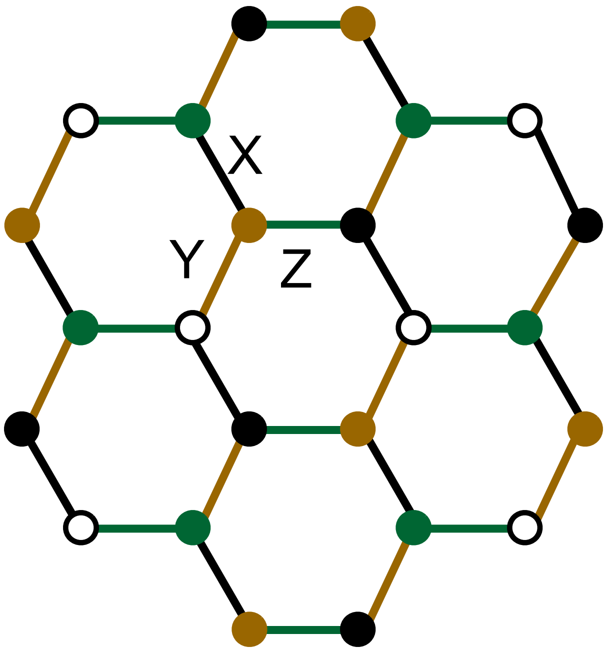
Examples of exchange processes

$$\mathcal{H}_{ij}^{(\gamma)} = 2K\ S_i^\gamma S_j^\gamma + J\ \mathbf{S}_i \cdot \mathbf{S}_j \qquad K = A\sin\varphi \qquad J = A\cos\varphi$$



Kitaev-Heisenberg Model

$$\mathcal{H}_{ij}^{(\gamma)} = 2K\ S_i^\gamma S_j^\gamma + J\ \mathbf{S}_i \cdot \mathbf{S}_j \qquad \tilde{\mathcal{H}}_{ij}^{(\gamma)} = 2[K+J]\ \tilde{S}_i^\gamma \tilde{S}_j^\gamma - J\ \tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j$$



- ○ original frame
- ● $(S^x, S^y, S^z) \rightarrow (S^x, -S^y, -S^z)$
- ● $(S^x, S^y, S^z) \rightarrow (-S^x, S^y, -S^z)$
- ● $(S^x, S^y, S^z) \rightarrow (-S^x, -S^y, S^z)$

$$\mathcal{H}_{ij}^{(z)} = S_i^x S_j^x + S_i^y S_j^y - S_i^z S_j^z \quad \Longrightarrow \quad \mathcal{H}_{ij}^{(\gamma)} = -\tilde{S}_i \cdot \tilde{S}_j$$

J.Chaloupka, GJ & G.Khaliullin PRL (2010)

further neighbor interactions explains zigzag phase

PHYSICAL REVIEW B **84**, 180407(R) (2011)

Kitaev-Heisenberg-J_2-J_3 model for the iridates A_2IrO_3

Itamar Kimchi[1] and Yi-Zhuang You[1,2]

PRL **108**, 127204 (2012) PHYSICAL REVIEW LETTERS week ending 23 MARCH 2012

Spin Waves and Revised Crystal Structure of Honeycomb Iridate Na_2IrO_3

S. K. Choi,[1] R. Coldea,[1] A. N. Kolmogorov,[2] T. Lancaster,[1,*] I. I. Mazin,[3] S. J. Blundell,[1] P. G. Radaelli,[1] Yogesh Singh,[4,5] P. Gegenwart,[4] K. R. Choi,[6] S.-W. Cheong,[6,7] P. J. Baker,[8] C. Stock,[8] and J. Taylor[8]

PRL **108**, 127203 (2012) PHYSICAL REVIEW LETTERS week ending 23 MARCH 2012

Relevance of the Heisenberg-Kitaev Model for the Honeycomb Lattice Iridates A_2IrO_3

Yogesh Singh,[1,2] S. Manni,[2] J. Reuther,[3,4] T. Berlijn,[5,6] R. Thomale,[7] W. Ku,[5,6] S. Trebst,[8] and P. Gegenwart[2]

PHYSICAL REVIEW B **87**, 220407(R) (2013)

Magnetic excitation spectrum of Na_2IrO_3 probed with resonant inelastic x-ray scattering

H. Gretarsson,[1] J. P. Clancy,[1] Yogesh Singh,[2] P. Gegenwart,[3] J. P. Hill,[4] Jungho Kim,[5] M. H. Upton,[5] A. H. Said,[5] D. Casa,[5] T. Gog,[5] and Young-June Kim[1,*]