Magnetism and doping effects in spin-orbit coupled Mott insulators

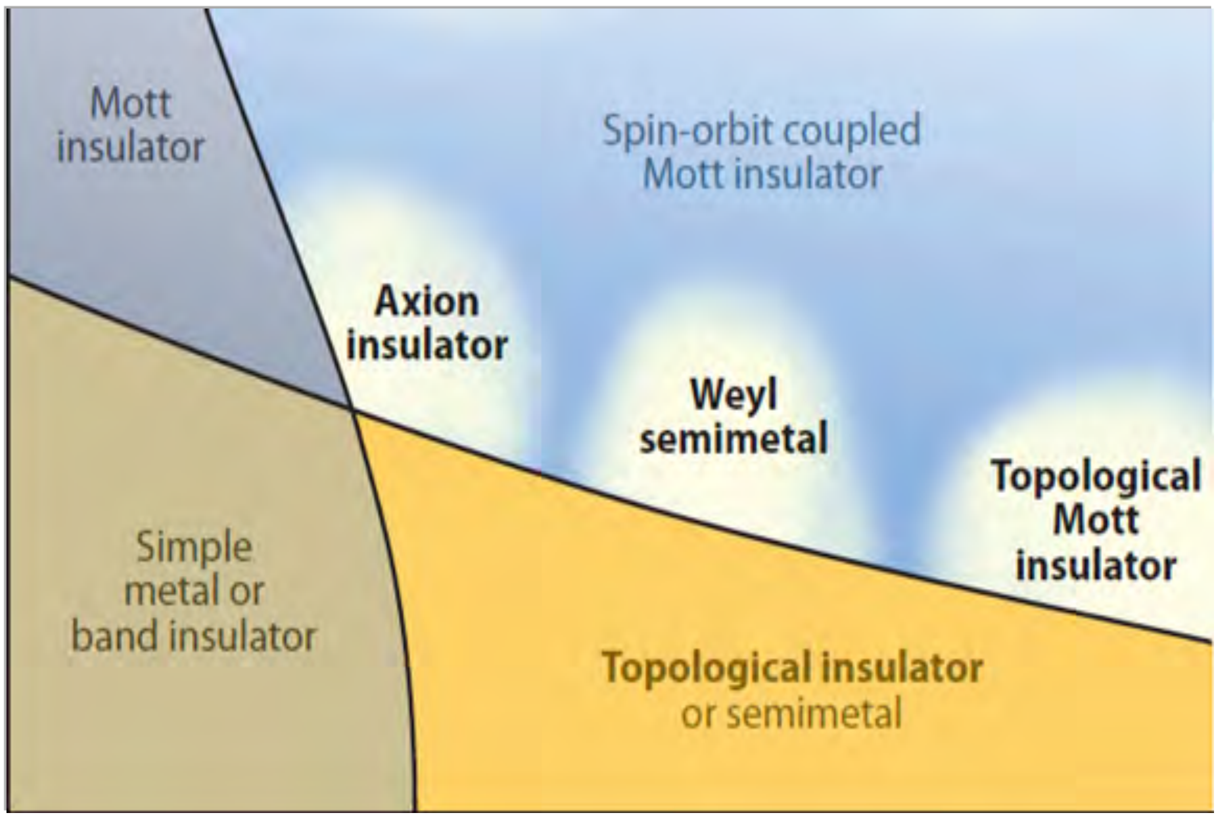
Giniyat Khaliullin

Max Planck Institute for Solid State Research, Stuttgart

Witczak-Krempa, Chen, Y-B.Kim, Balents (Annu. Rev. 2014)

Hubbard model with spin-orbit coupling:
unconventional magnets, insulators, metals

correlation U

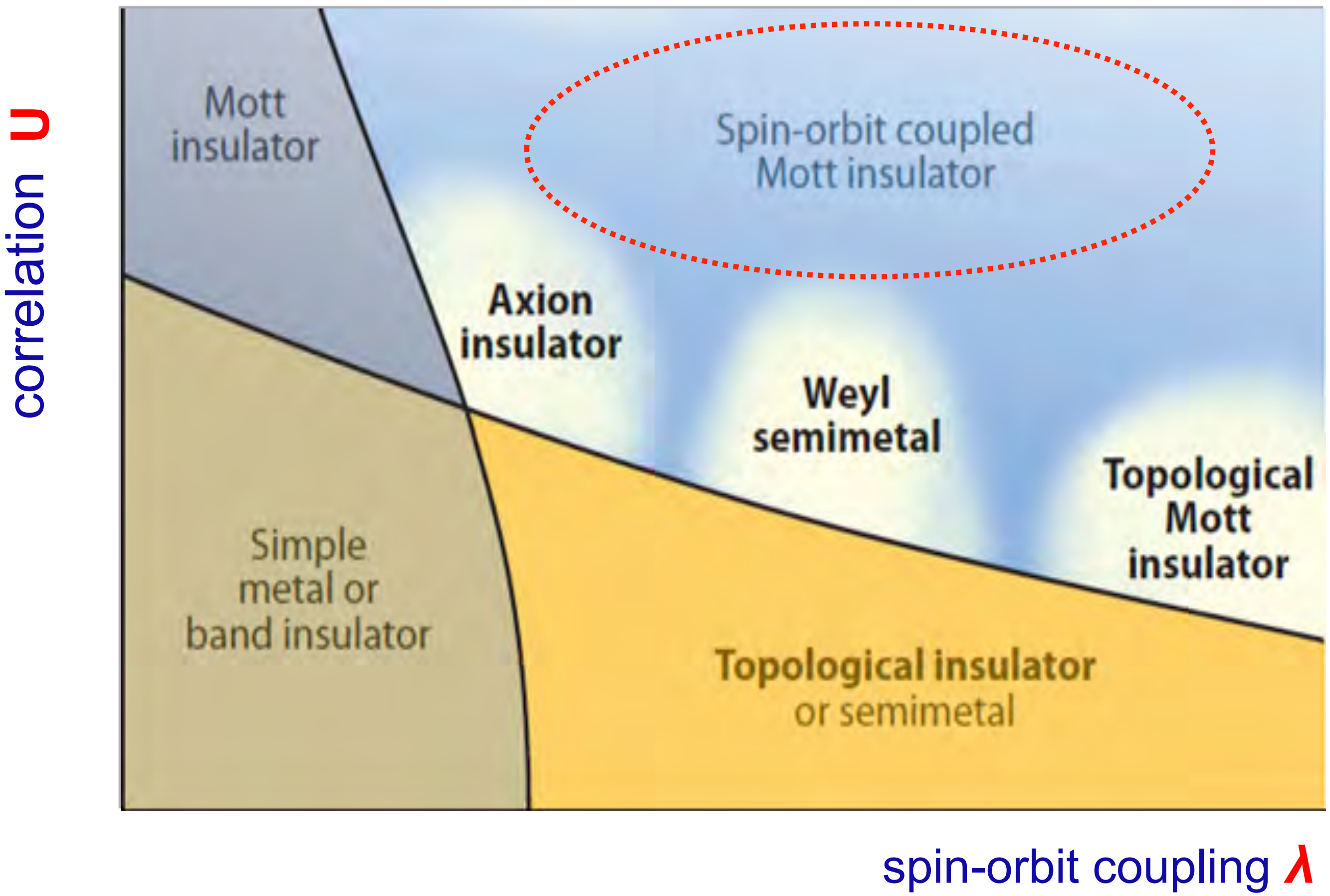


spin-orbit coupling λ

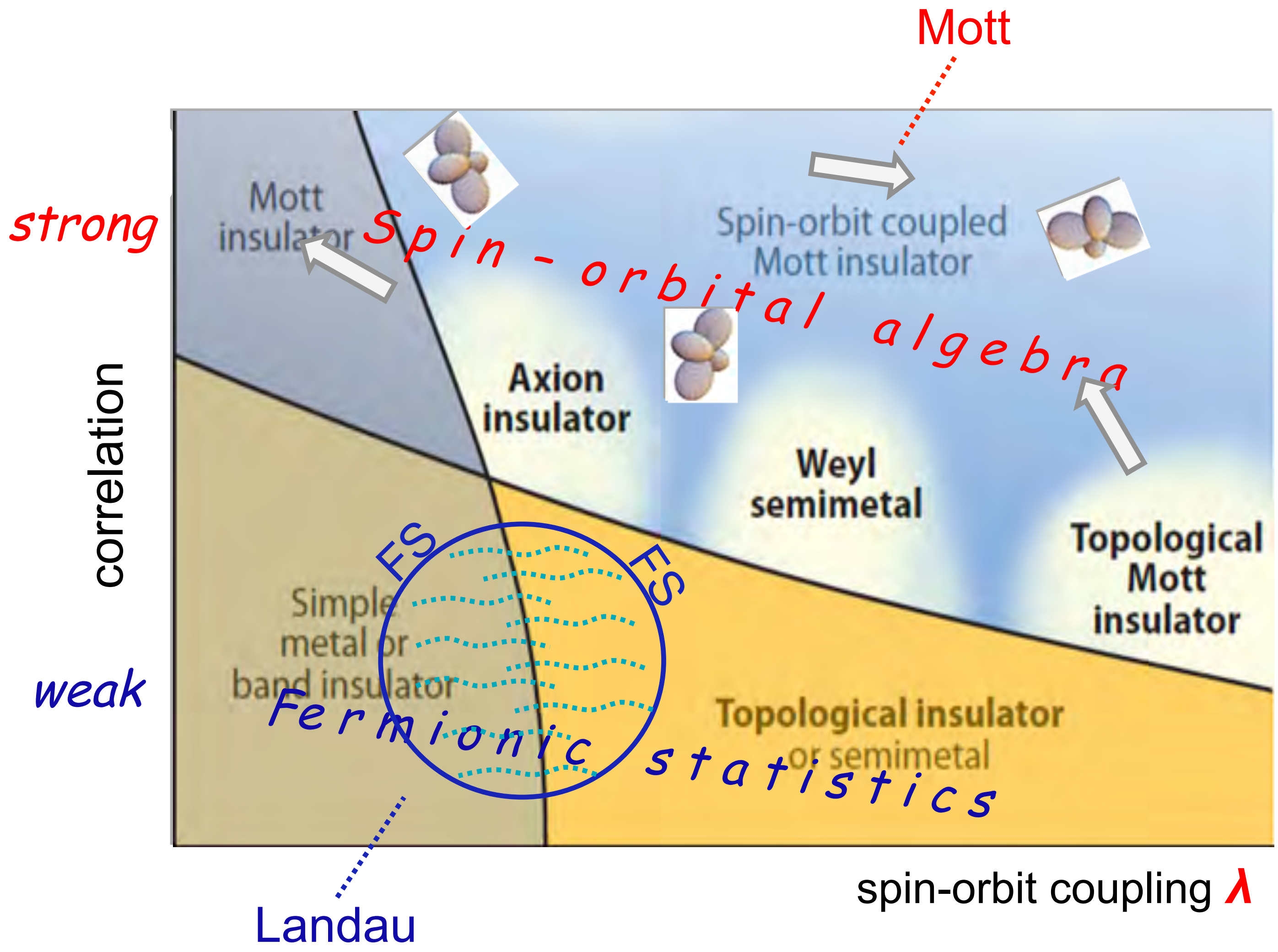
Witczak-Krempa, Chen, Y-B.Kim, Balents (Annu. Rev. 2014)

Hubbard model with spin-orbit coupling:

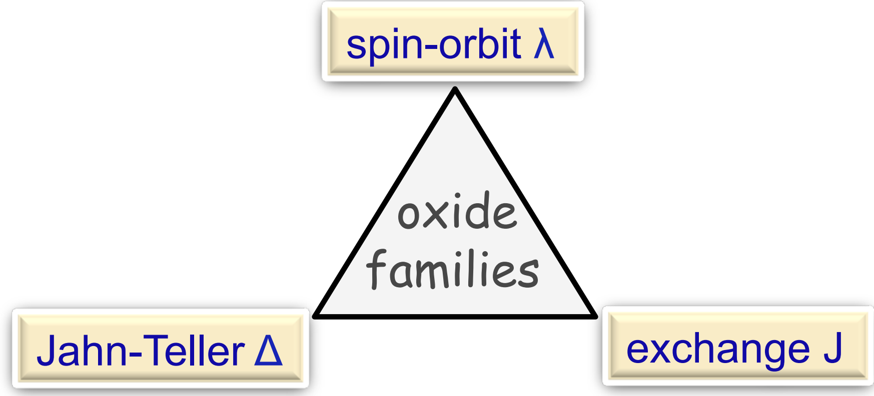
unconventional magnets, insulators, metals



Ionic multiplets *vs* Fermionic bands

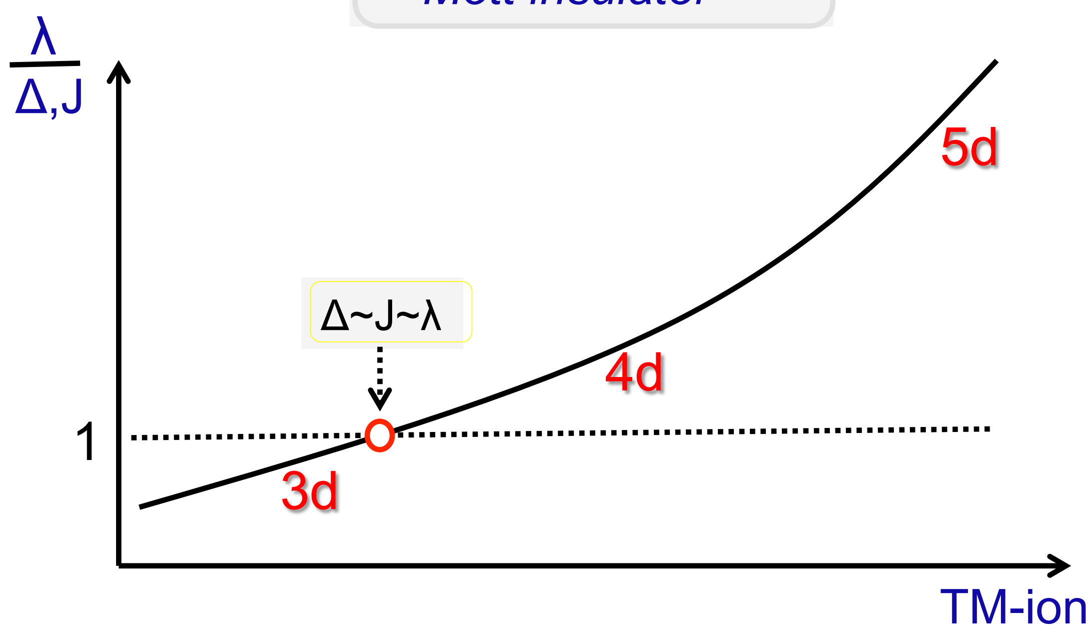


Three competitors:

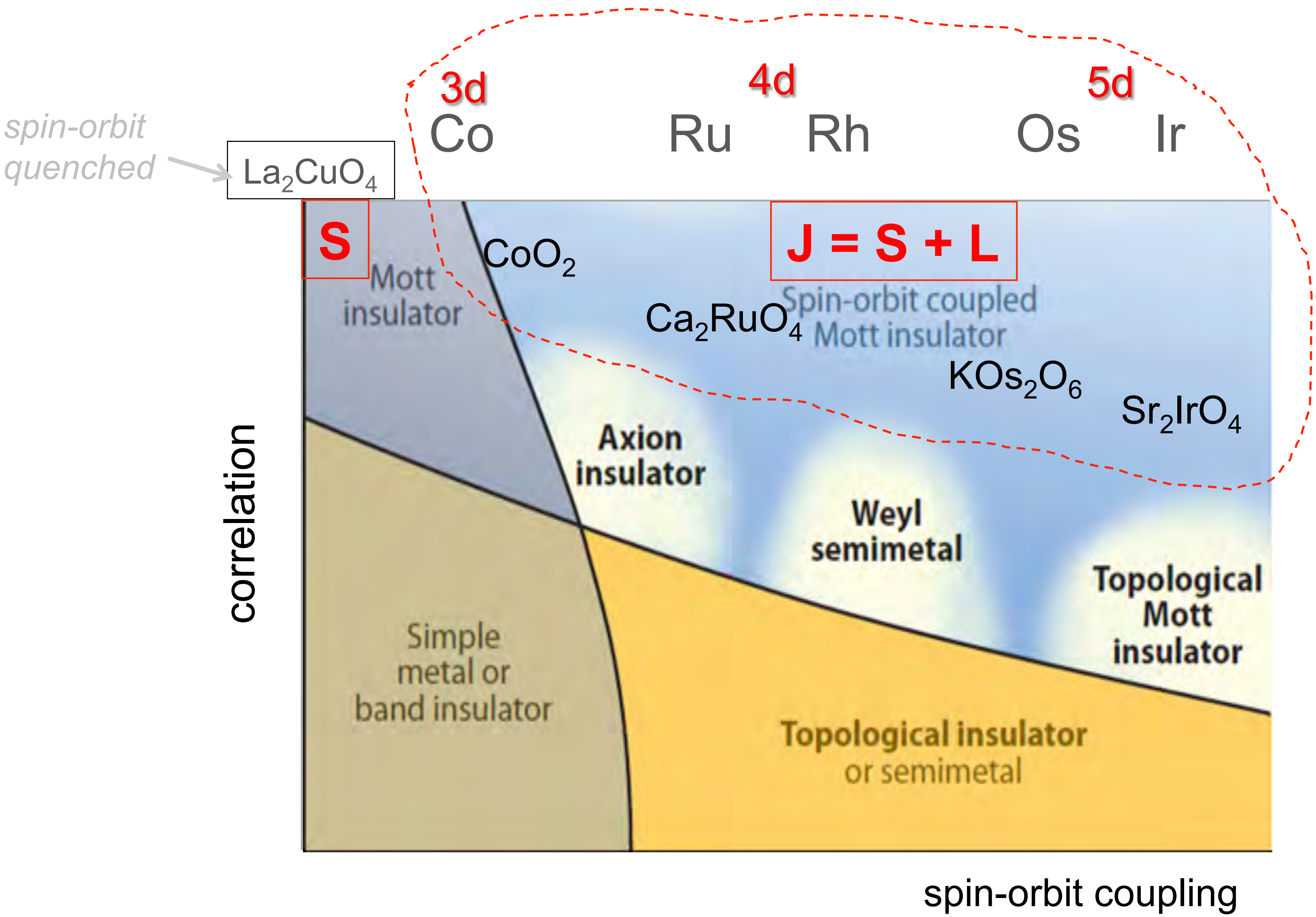


λ > Δ,J

„spin-orbit coupled"
Mott insulator



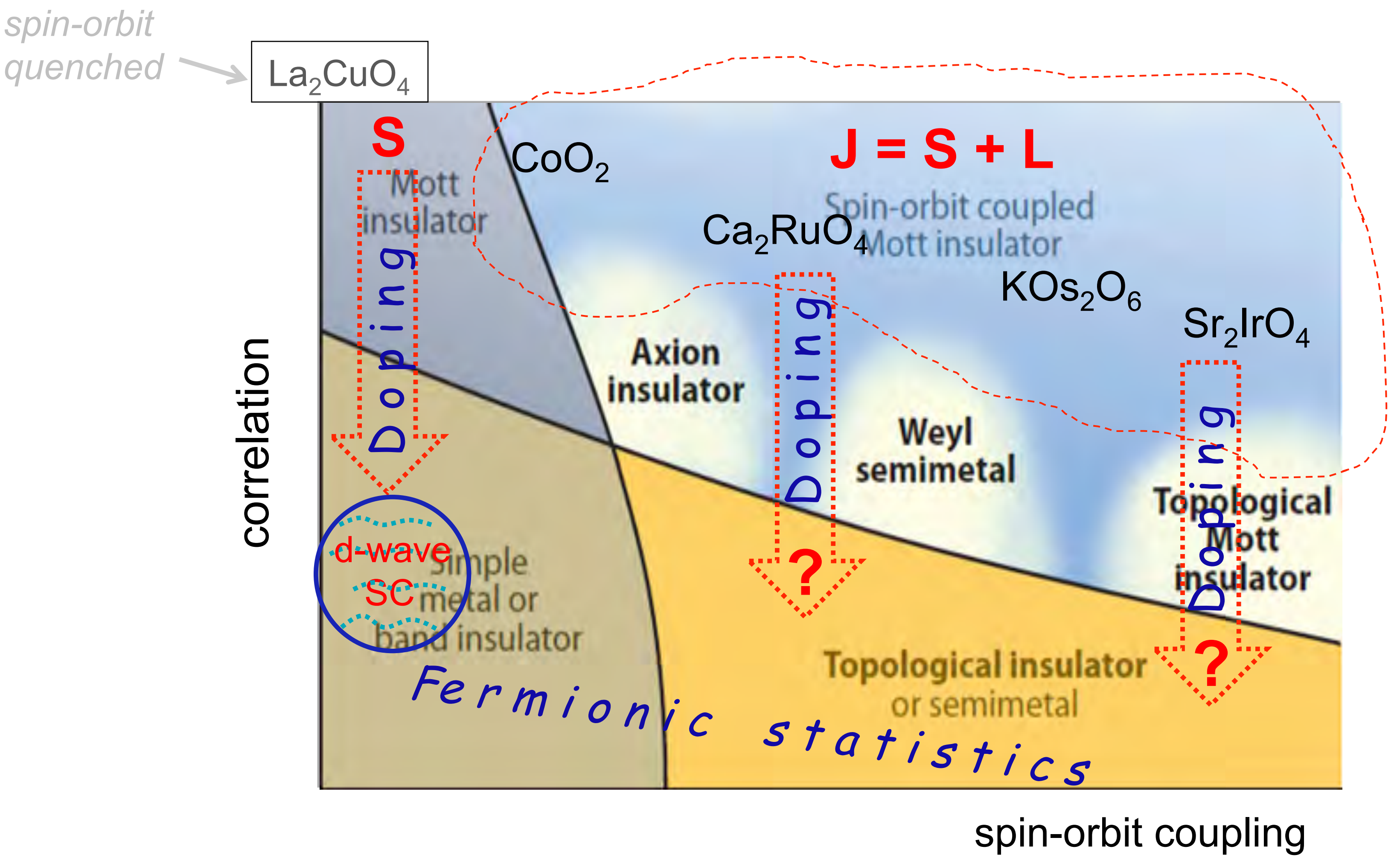
Spin-orbit coupled Mott insulators



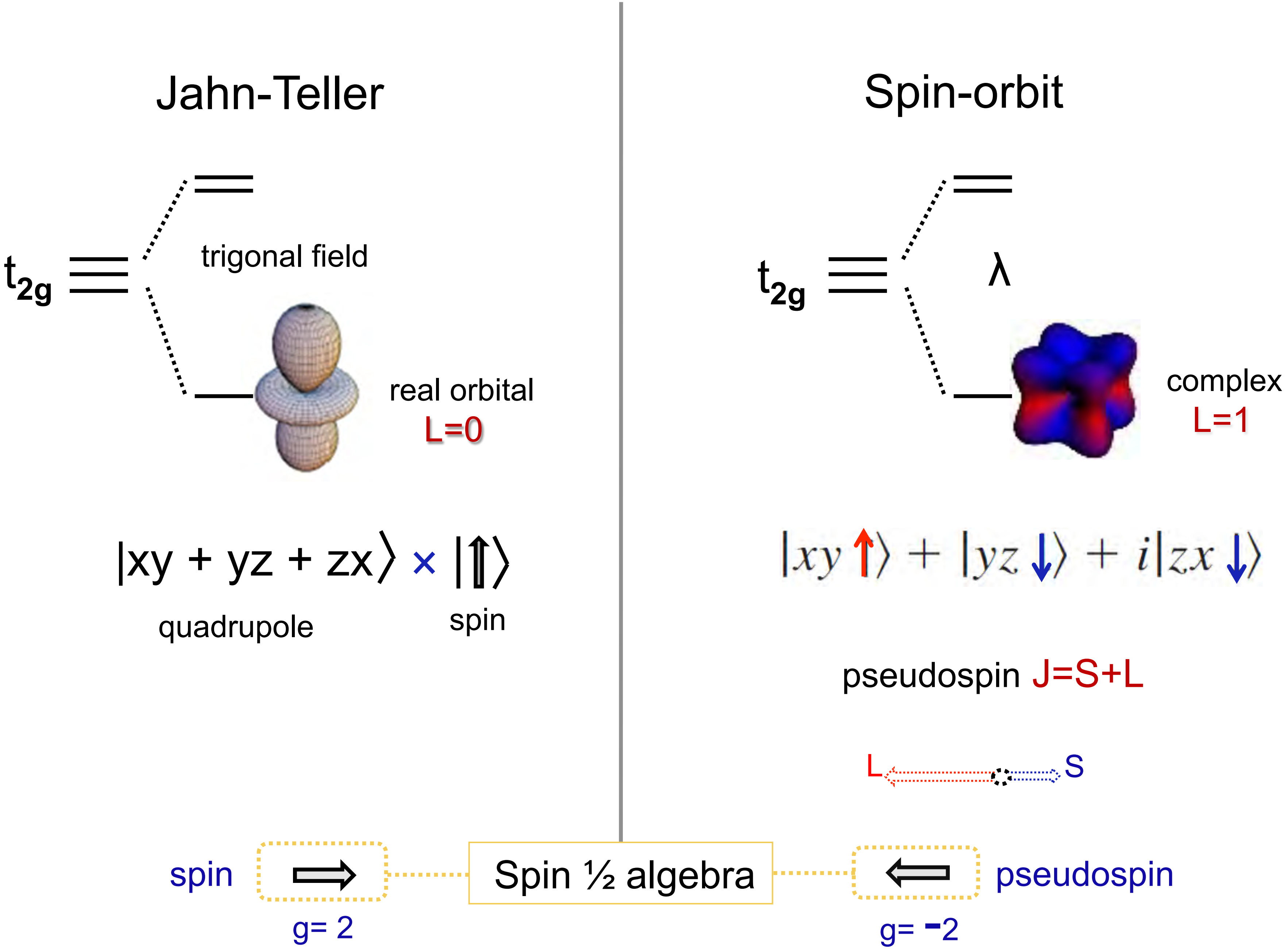
„unconventional“ J-magnetism

D O P I N G:

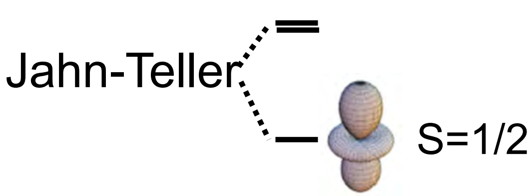
„unconventional“ metal & SC?

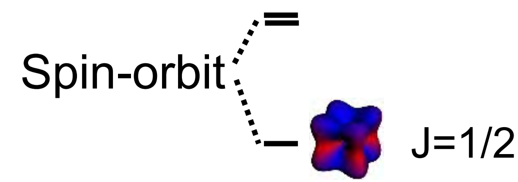


Lifting the orbital degeneracy

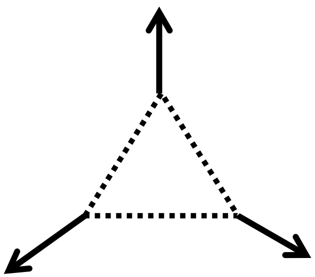


Magnetism



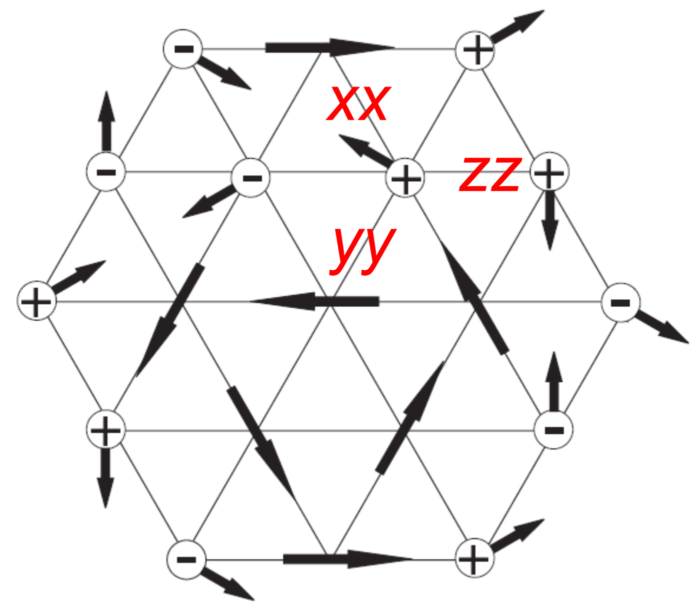


H = (SS)



coplanar,
single-Q

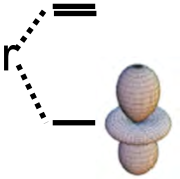
H= - (SS) + ISING$^{(\alpha)}$



large magnetic unit cell
non-coplanar, multi-Q
hosts spin vortex

GKh (PTPS, 2005)

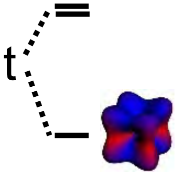
SC pairing



Spin-singlet d-wave SC

$|\uparrow\downarrow - \downarrow\uparrow\rangle$

Baskaran (2003)
Kumar, Shastry (2003)
Ogata (2003)
P. Lee et al.(2004)

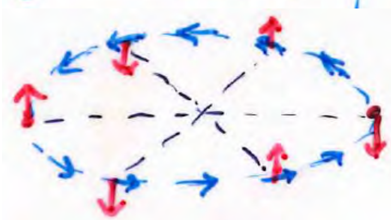


Pseudospin-triplet p-wave SC

$$\Delta_{tr} = \alpha_0 |\tilde{\uparrow}\tilde{\downarrow} + \tilde{\downarrow}\tilde{\uparrow}\rangle_{ij} + \alpha_1 e^{i\varphi_{ij}} |\tilde{\uparrow}\tilde{\uparrow}\rangle_{ij} + \alpha_1 e^{-i\varphi_{ij}} |\tilde{\downarrow}\tilde{\downarrow}\rangle_{ij}$$

nondegenerate

$\bar{d}$ -vector pattern on FS :



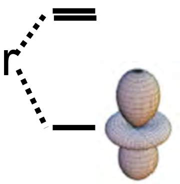
$d_x, d_y, d_z \neq 0$

Knight-shift finite in all directions

GKh, Koshibae, Maekawa (PRL 2004)
GKh (PTPS, 2005)

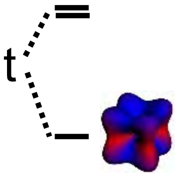
SC pairing

Jahn-Teller



S=1/2

Spin-orbit



J=1/2

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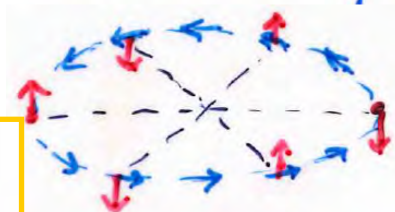
$\bar{d}$ -vector pattern on FS :



$d_x, d_y, d_z \neq 0$

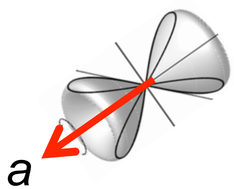
	3d	4d	5d
	Co^{4+}	Rh^{4+}	Ir^{4+}
λ :	80 meV	190 meV	380 meV

GKh, Koshibae, Maekawa (PRL 2004)
GKh (PTPS, 2005)

$Na_x(Co,Rh,Ir)O_2$

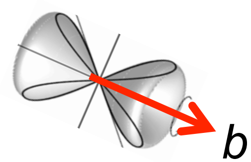
The origin of „unconventionality“: ORBITAL magnetism

Orbital moment **L** has a „shape“:



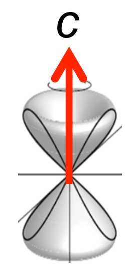
$L_x=1$

$x(y+iz)$



$L_y=1$

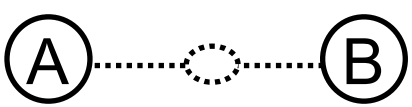
$y(z+ix)$



$L_z=1$

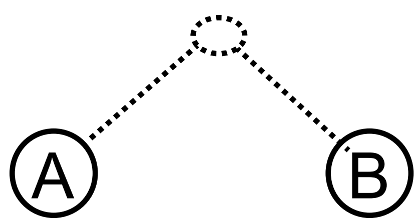
$z(x+iy)$

Hopping amplitude:



real

(inversion)



complex

(no inversion)

nontrivial band topology
Shitade *et al.* (2009)

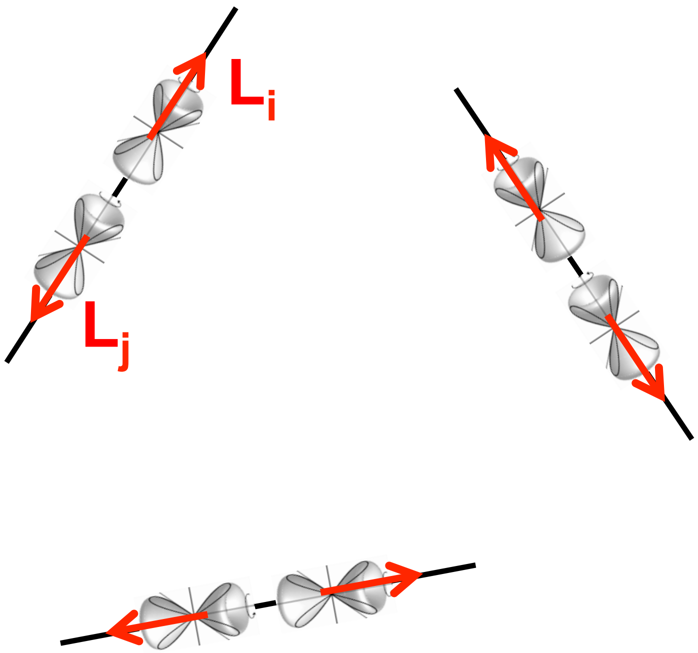
ORBITAL MAGNETISM

L-moment direction and chemical bonding:
one-to-one correspondence

L - moment interactions:

anisotropic, bond-dependent

„orbital frustration"



non-Heisenberg models:
pseudodipolar, biquadratic…

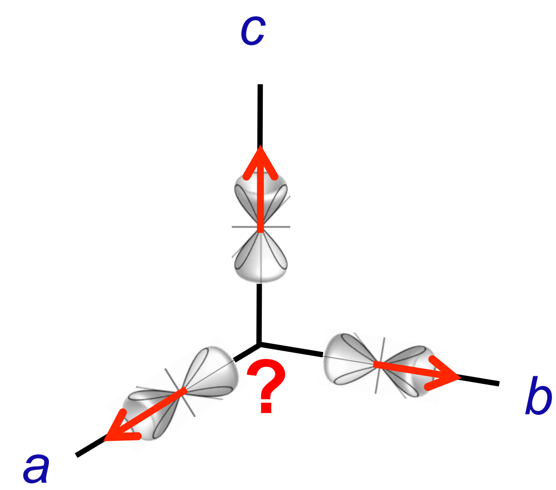
ORBITAL MAGNETISM

Orbital moment **L** interactions:

1) non-Heisenberg

2) bond-dependent

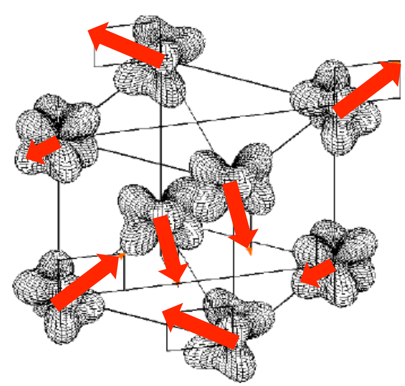
„orbital frustration"



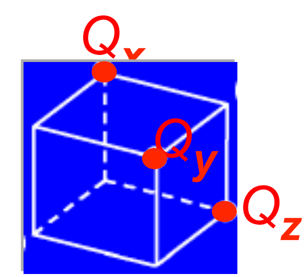
- Simple cubic lattice:

$$\mathcal{H}_{ij}^{(c)} = (L_i^x L_j^x)^2 + (L_i^y L_j^y)^2 + L_i^x L_i^y L_j^y L_j^x + L_i^y L_i^x L_j^x L_j^y$$

GKh & Okamoto (PRL 2002)

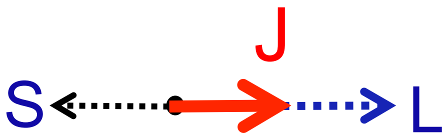


non-coplanar



multi-Q

no LRO at finite T



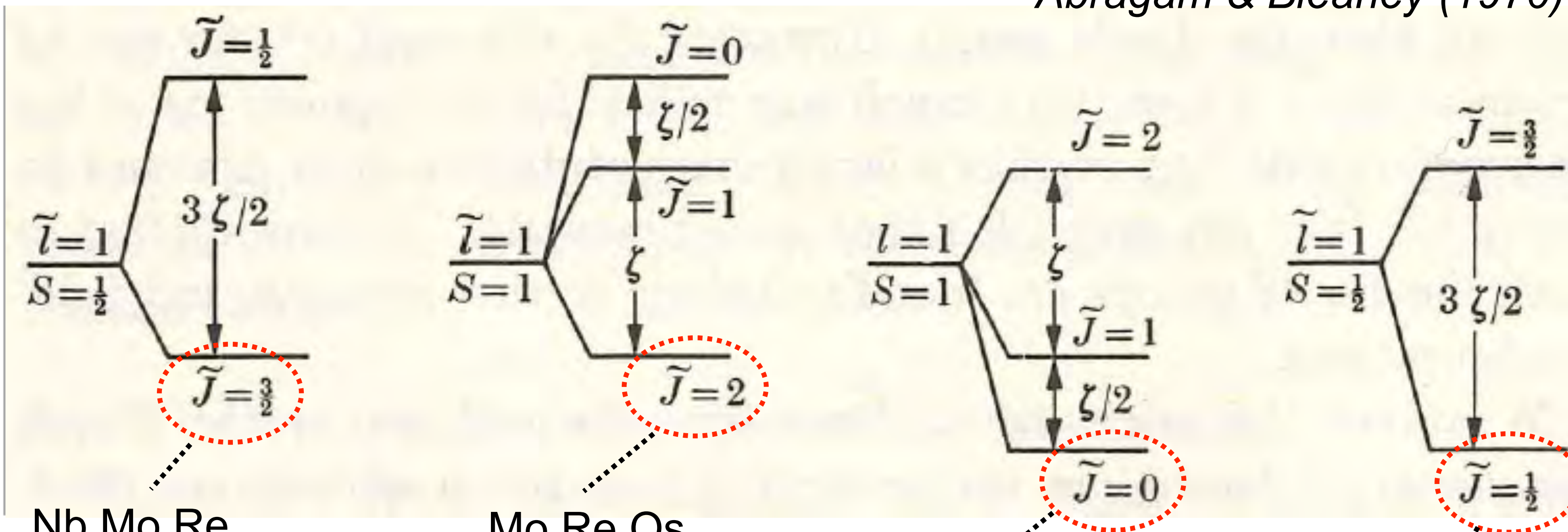
Pseudospin J=S+L inherits bond-dependent and frustrated nature of orbitals

⇩

„unconventional“ magnetism

Spin-orbit multiplels of TM-ions

Abragam & Bleaney (1970)



Nb,Mo,Re

Mo,Re,Os

large J multipoles:
spin nematic, „hidden" order

Ru,Re,Os
„nonmagnetic"

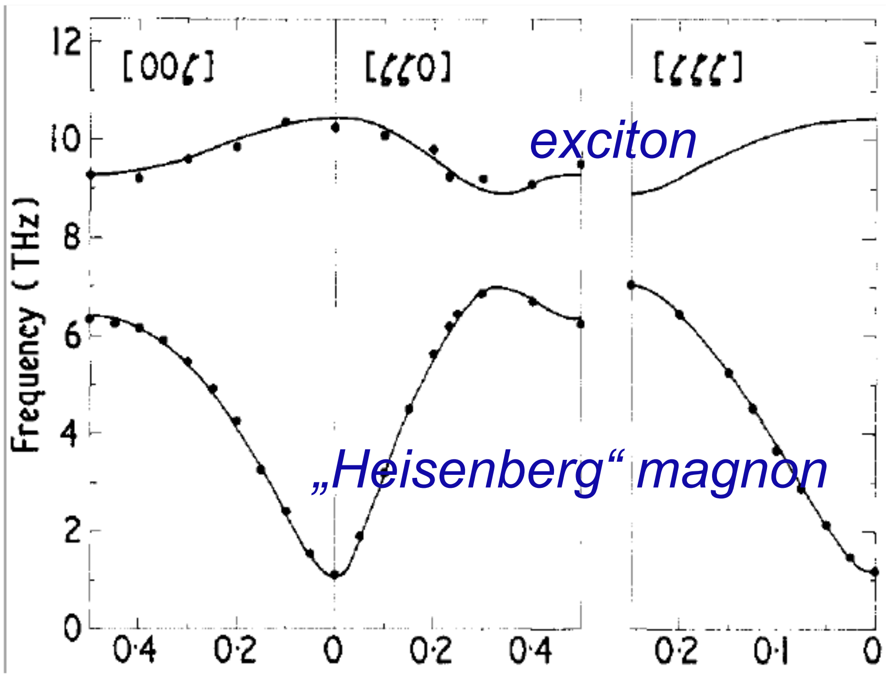
Co,Rh,Ir
quantum spin

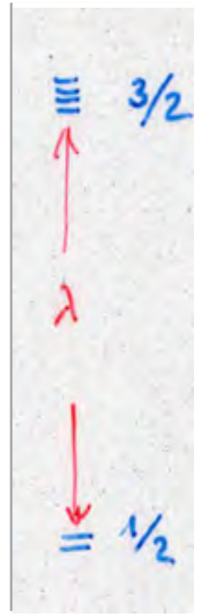
GS-degeneracy: *„pseudospin"* $\tilde{J}$

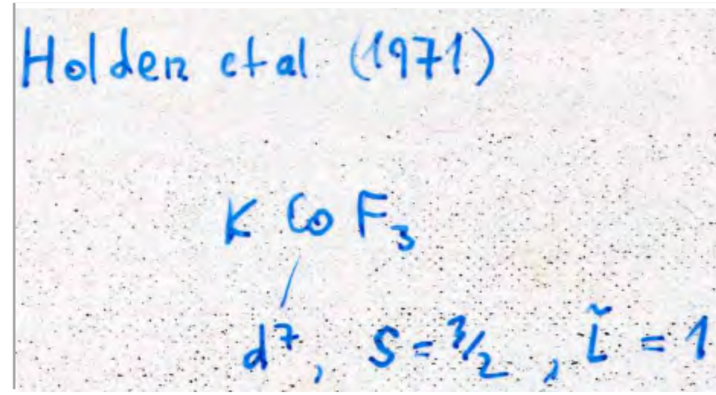
Kramers: *dipole, octupole,…*
non-Kramers: *quadrupole,…*

The „old“ pseudospin J=1/2

3d Co-fluoride, neutron scattering







Two branches split by spin-orbit:
magnon & exciton

from *3d* Co to *5d* Ir

B.J.Kim et al., …

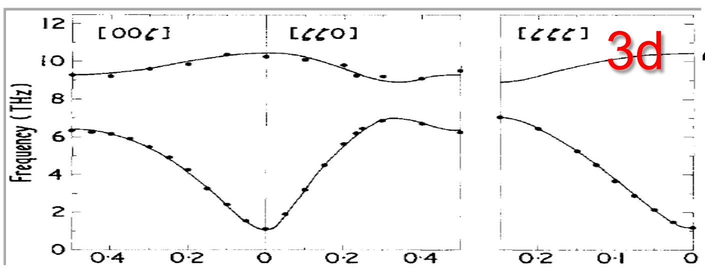
Exciton

5d 600 meV

× 10

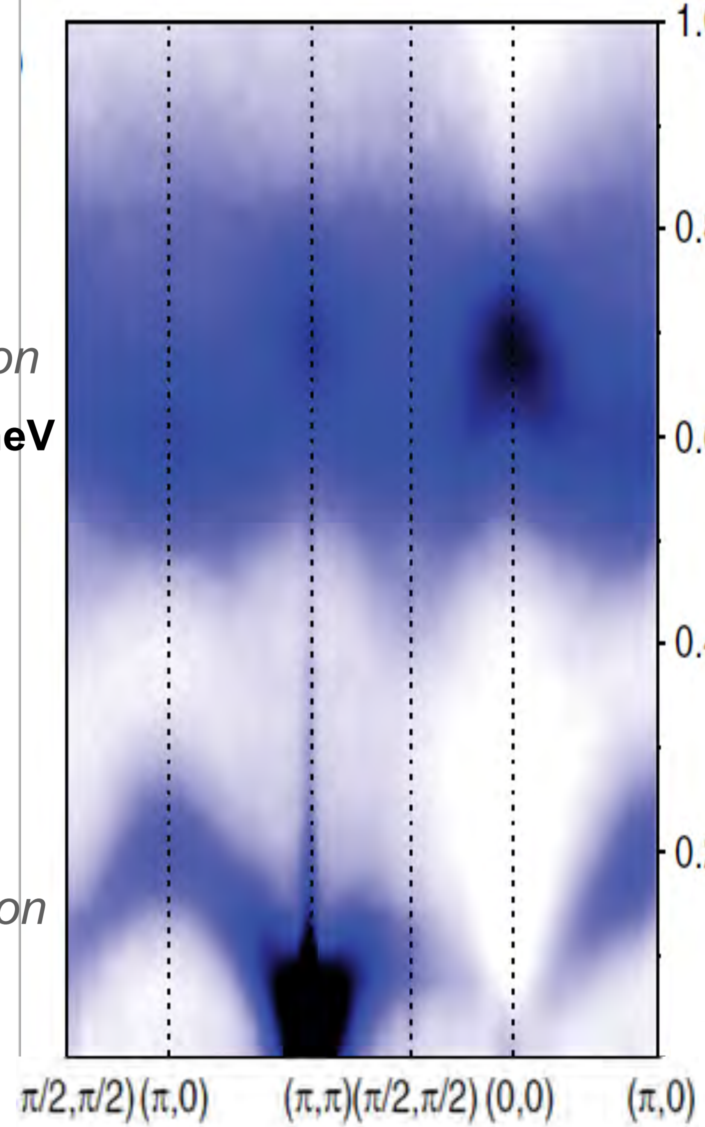
3d 40 meV

Magnon



K_2CoF_3

Holden *et al.* (1971)
neutron scatteting

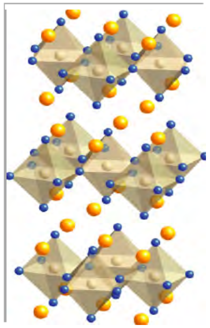


Sr_2IrO_4

Kim *et al.* (2012)
RIXS data

spin one-half 2D Mott systems similar to cuprates ?

d^1	d^5	d^7	d^9
t_{2g} electron	t_{2g} hole	e_g electron	e_g hole
Ti	Co	Ni	Cu
...	...	...	

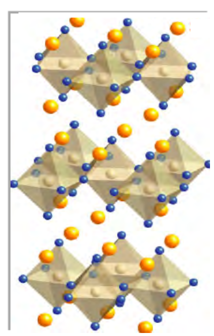


La_2CuO_4

J_{AF}=130 meV

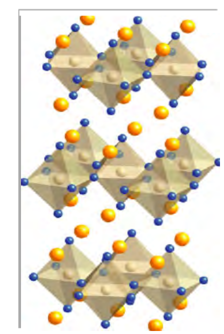
spin one-half 2D Mott systems similar to cuprates

d^1	d^5	d^7	d^9
t_{2g} electron	t_{2g} hole	e_g electron	e_g hole
	Ir		Cu



Sr_2IrO_4 T_N~240 K

Cuprate-like magnetism and SC?



La_2CuO_4

J_{AF}=130 meV

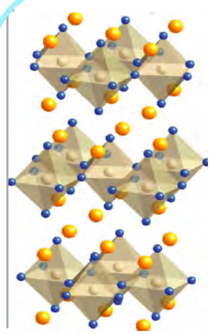
T_N~320 K

(Cava, Cao,… 1990's)

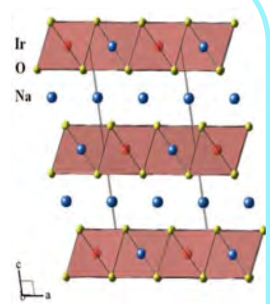
„..apart from cuprates, Sr_2IrO_4 would be the second S=1/2 2D AF "

spin one-half 2D Mott systems similar to cuprates

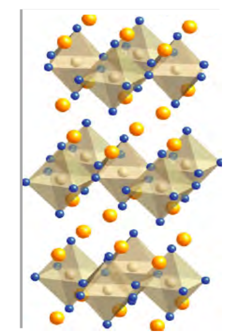
d^1	d^5	d^7	d^9
t_{2g} electron	t_{2g} hole	e_g electron	e_g hole
	Ir		Cu



Sr_2IrO_4



Na_2IrO_3



La_2CuO_4

t_{2g} ← *different orbitals* → e_g

spin-orbit coupling

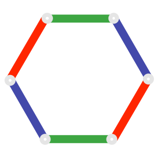
J=1/2 magnetism in iridates: *basic theory*

Jackeli, GKh (2009)
Chaloupka, Jackeli, GKh (2010, 2013)

Two-parameter Hamiltonian = Ising$^{(x,y,z)}$ + Heisenberg:

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

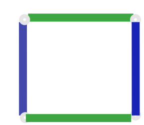
dominant in 90-bonding
(honeycomb Na_2IrO_3)



= *Kitaev model (2006)*

„Majorana world“ ?

dominant in 180-bonding
(Sr_2IrO_4 perovskite)



= *„cuprate“ model*

high-T_c SC ?

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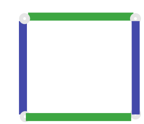

= *„cuprate“ model*
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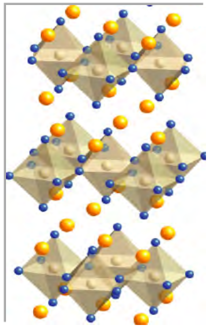
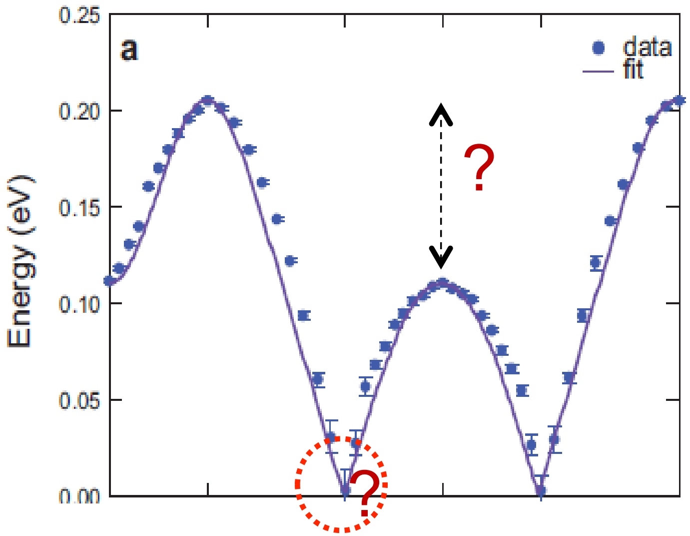


Sr_2IrO_4

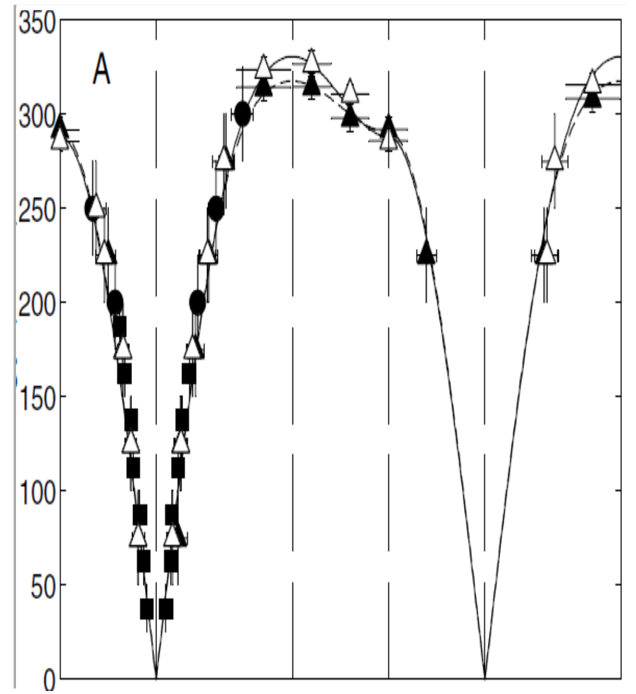
Pseudospin ½ in perovskites:

-theory predicts
„nearly" Heisenberg AF



Sr_2IrO_4
T_N~240 K

Kim et al.(2012)

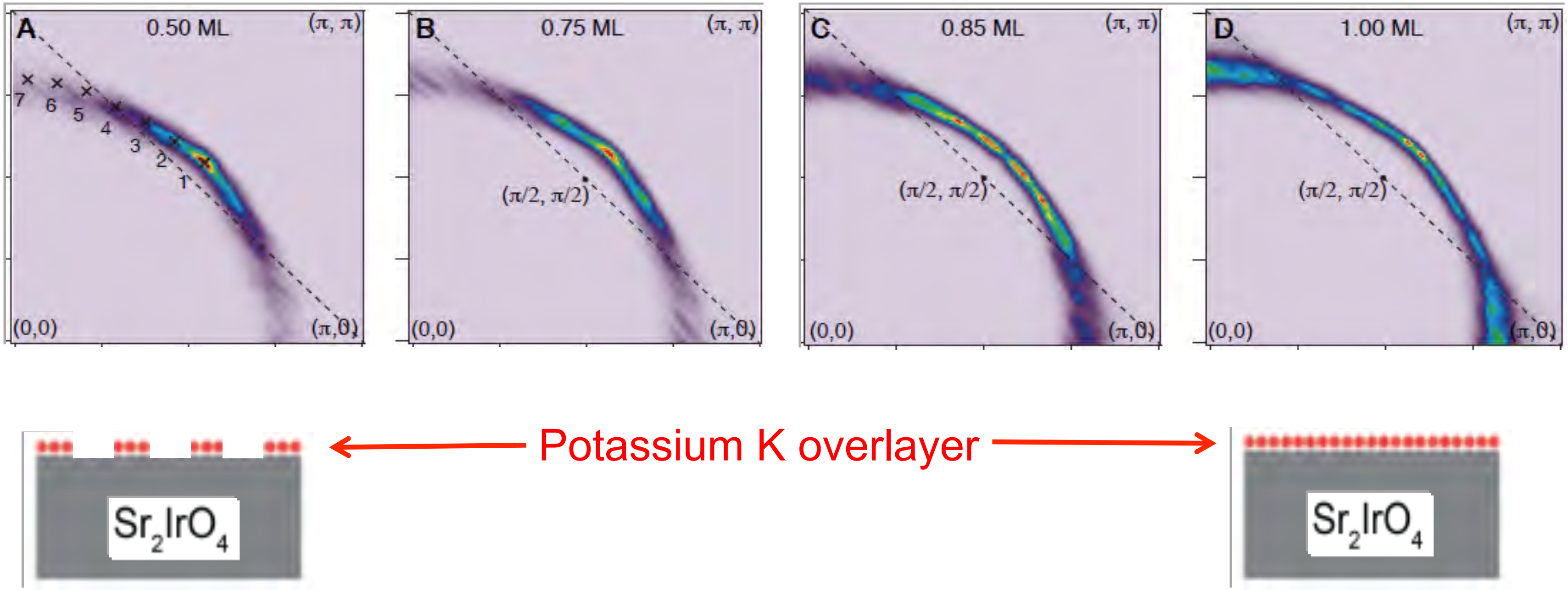


La_2CuO_4
T_N~320 K

Coldea et al.(2001)

Fermiology of electron doped Sr_2IrO_4

„Fermi-arcs“ at low doping *„normal“ FS*



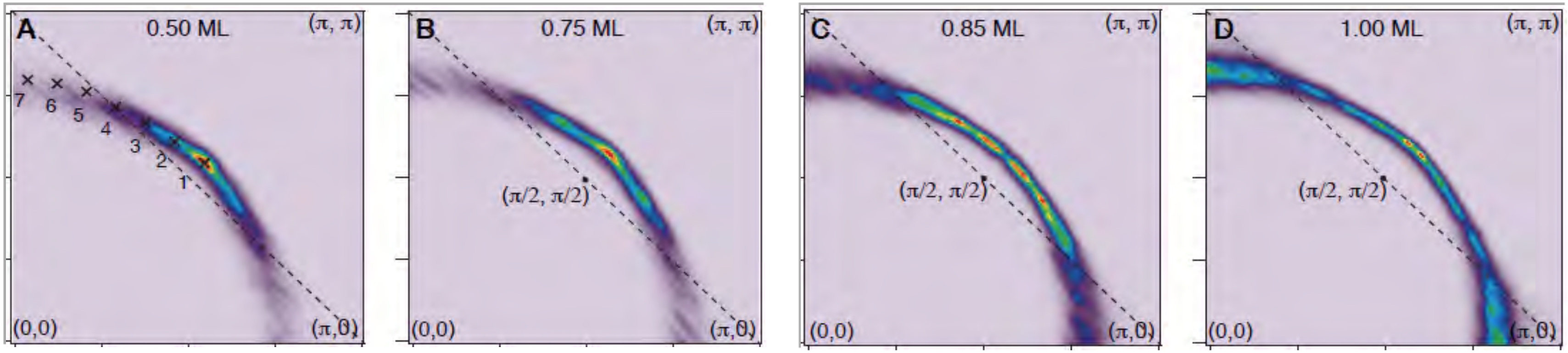
B.J. Kim *et al.* (Science 2014)

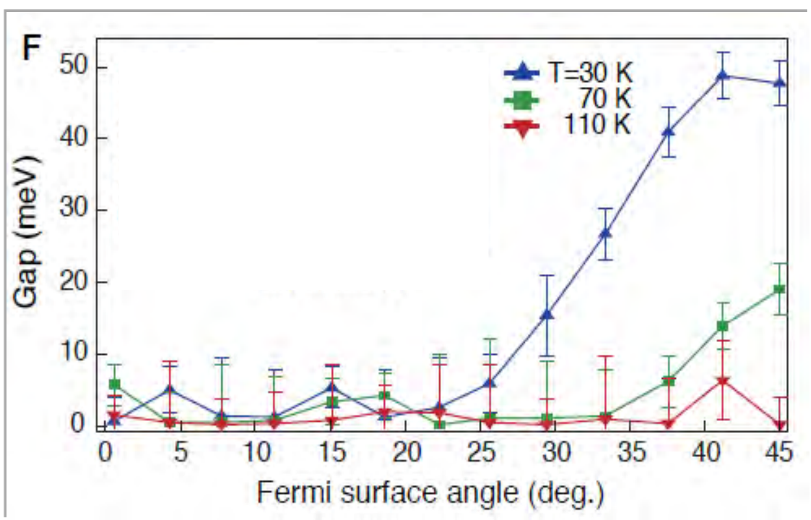
B.J. Kim et al. (Science 2014)

T-dependent „pseudogap" in Sr_2IrO_4

„Fermi-arcs" at low doping

„normal" FS





Pseudogap opens at low T

many-body effect !

…and closes at 110 K

Sr_2IrO_4 magnetism, fermiology & lattice: *same as in* La_2CuO_4

superconductivity?

YES

SUPER!
GREAT!

NO

„…find 10 differences…"

Sr_2IrO_4

La_2CuO_4

YES or NO? -- no definite answer yet…

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Chaloupka, Jackeli, GKh (2010, 2013)

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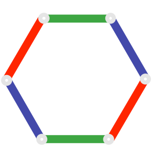
dominant in 90-bonding
(honeycomb Na_2IrO_3)

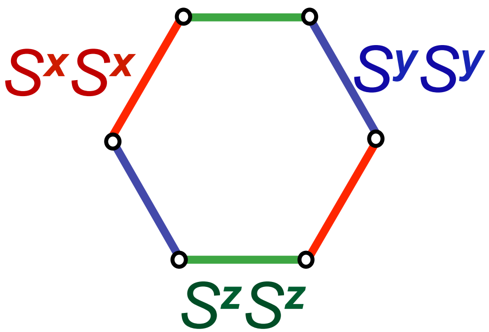


= *Kitaev model (2006)*
⇩
„Majorana world“ ?

dominant in 180-bonding
(Sr_2IrO_4 perovskite)

= *„cuprate“ model*
⇩
high-T_c SC ?

The Kitaev model

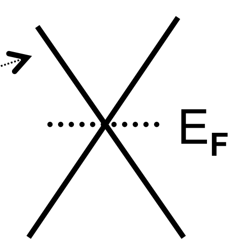


- Exactly solvable
- Short-range RVB, large spin gap
- Low-energy excitations:
  free Majorana fermions

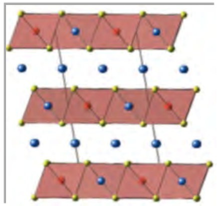
local & high-energy

$$S_\alpha = c \, f_\alpha$$

Itinerant free band
(uncharged)



Dirac cones



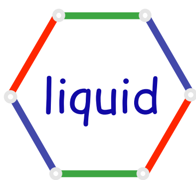
Na_2IrO_3

Honeycomb lattice:

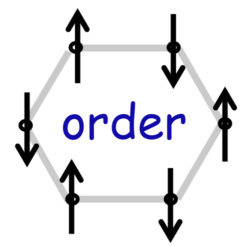
-- *Kitaev term is dominant*
but J is present as well

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

Kitaev model

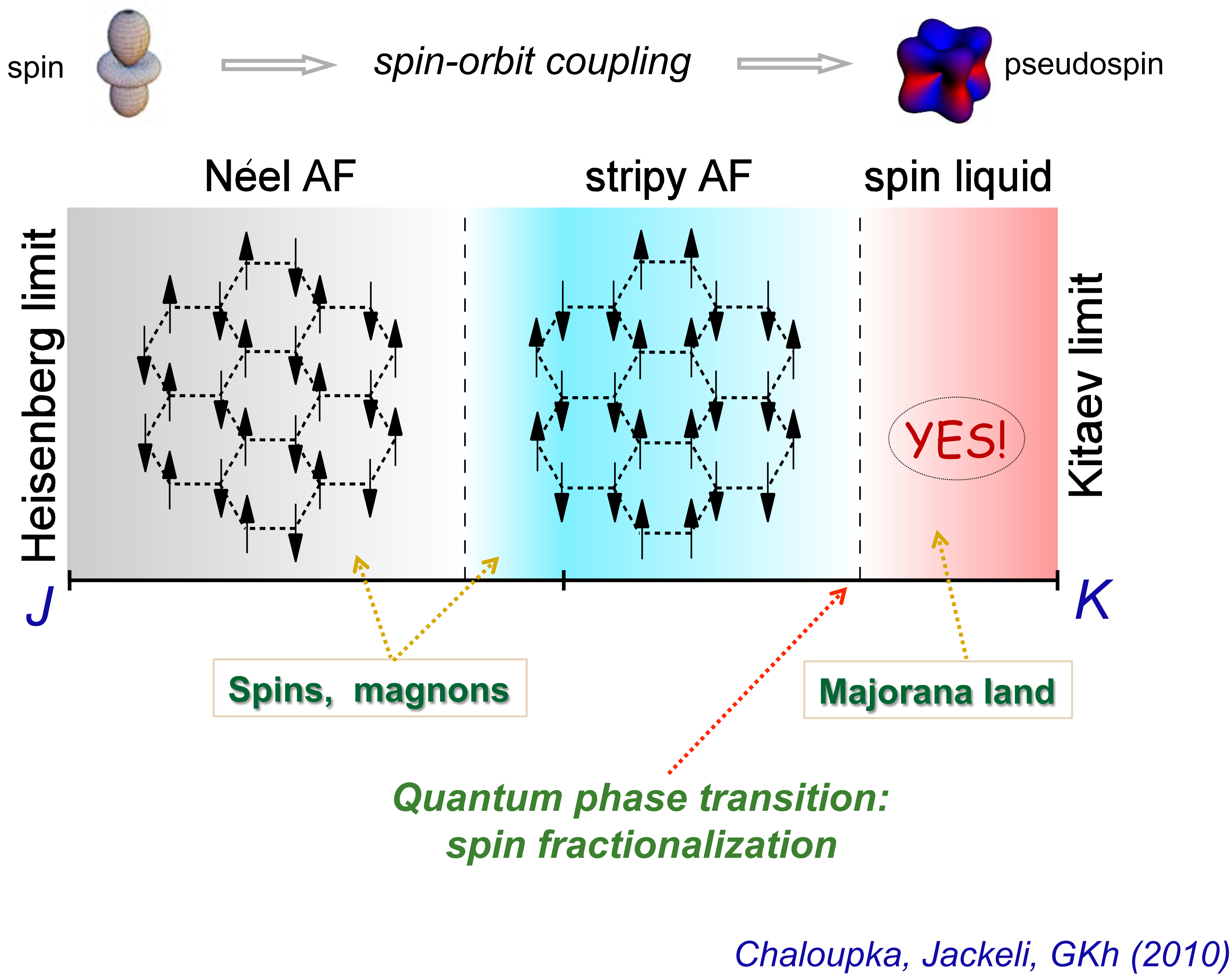


Heisenberg model



What is in between ?

-If „some liquid" is still left ?


spin
spin-orbit coupling
pseudospin
Néel AF
stripy AF
spin liquid
Heisenberg limit
Kitaev limit
YES!
J
K
Spins, magnons
Majorana land
Quantum phase transition:
spin fractionalization
Chaloupka, Jackeli, GKh (2010)

real world: $\mathbf{Na_2IrO_3}$

Exp.data

AM order T_N~15K

(1) Mag. bandwidth: 40 meV~30 T_N
(Gretarrson et al.)

(2) Intense q=0 scattering
(Gretarrson et al.; B.J. Kim et al.)

(3) SW gap is small < 2 meV
(Coldea et al.; B.J. Kim et al.)

Interactions:

strongly frustrated

non-Heisenberg

C3 symmetric

Kitaev-Heisenberg K-J model with large K > J

makes all these „for free“ but…

real world: $\mathbf{Na_2IrO_3}$

Exp.data

AM order T_N~15K

(1) Mag. bandwidth: 40 meV~30 T_N
(Gretarrson et al.)

(2) Intense q=0 scattering
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(3) SW gap is small < 2 meV
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Interactions:

strongly frustrated

non-Heisenberg

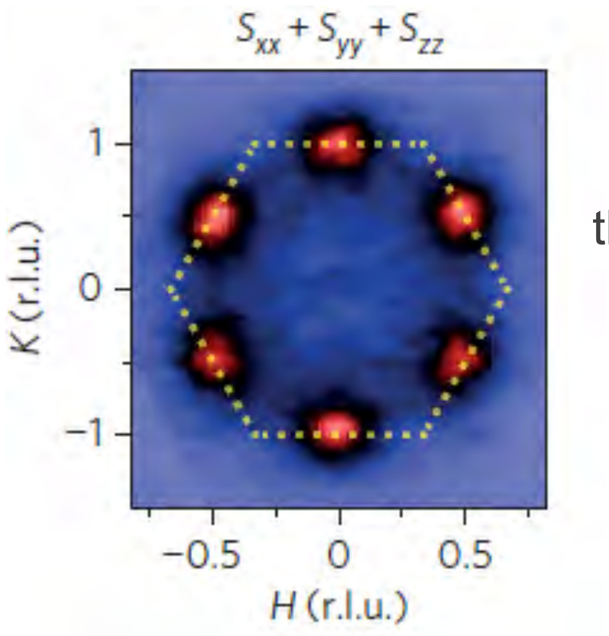
C3 symmetric

B.J. Kim et al., 2015:
Unusual moment direction away from symmetry axes

need more terms beyond Kitaev-Heisenberg model

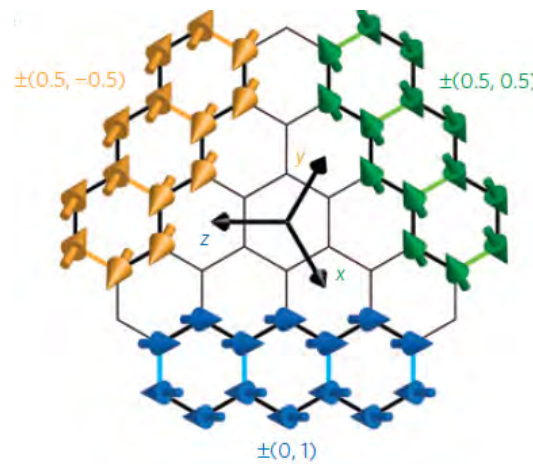
B.J. Kim et al., Nature Phys. 2015

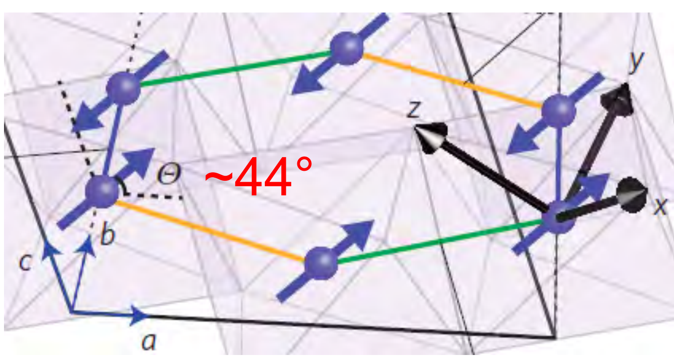
Diffuse magnetic X-ray scattering intensities above T_N



three equivalent zigzag quasielastic peaks

C3 symmetry





Unusual moment direction
away from symmetry axes

↓

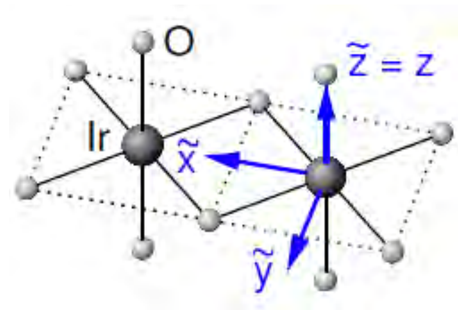
inconsistent with pure KH-model

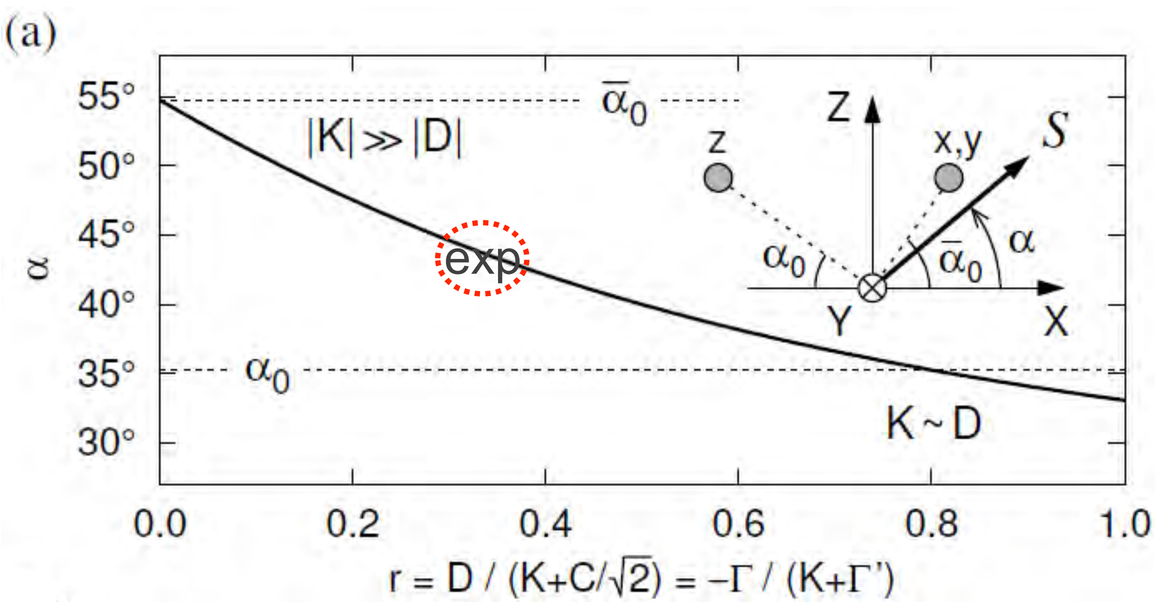
Phenomenological model („extended-KH“):

(H.Y. Kee et al., van den Brink et al, …)

$$\mathcal{H}_{\langle ij\rangle \parallel c} = J\,\mathbf{S}_i\cdot\mathbf{S}_j + K\,S_i^{\tilde z}S_j^{\tilde z} + D\,(S_i^{\tilde x}S_j^{\tilde x} - S_i^{\tilde y}S_j^{\tilde y}) + C\,(S_i^{\tilde y}S_j^{\tilde z} + S_i^{\tilde z}S_j^{\tilde y})$$

Two new terms: D and C





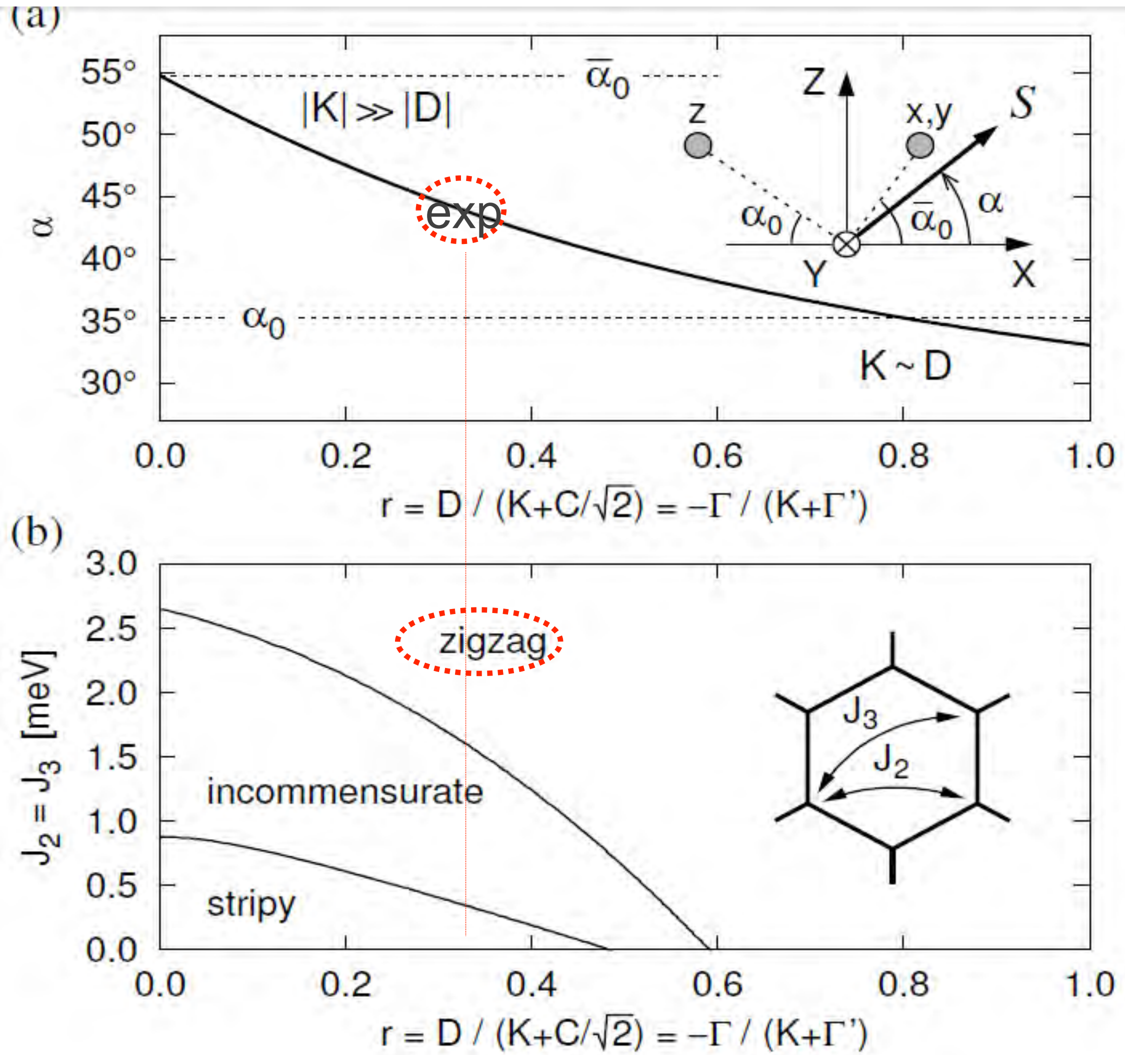
Chaloupka & GKh, condmat 2015

K,J,D,C model can make the moment angle 40°-45°.

However, zigzag is not then stable.
We need longer range interactions.

… adding $J_{2,3}$ *(Coldea et al. 2012)*

$$\mathcal{H}_{\langle ij\rangle \parallel c} = J\,\mathbf{S}_i \cdot \mathbf{S}_j + K\, S_i^{\tilde{z}} S_j^{\tilde{z}} + D\,(S_i^{\tilde{x}} S_j^{\tilde{x}} - S_i^{\tilde{y}} S_j^{\tilde{y}}) + C\,(S_i^{\tilde{y}} S_j^{\tilde{z}} + S_i^{\tilde{z}} S_j^{\tilde{y}}) + J_{2,3}(SS)$$



Sizable anisotropy D
&
long-range $J_{2,3}$

Right moment direction
&
zigzag order

PHYSICAL REVIEW B 88, 035107 (2013)

***Ab initio* analysis of the tight-binding parameters and magnetic interactions in Na_2IrO_3**

Kateryna Foyevtsova,[1] Harald O. Jeschke,[1] I. I. Mazin,[2] D. I. Khomskii,[3] and Roser Valentí[1]

⇩

... the nnKH model, is, apparently, inadequate.

Spatially extended, „quasimolecular“ orbitals: longer-range couplings $J_{2,3}$

PHYSICAL REVIEW B 88, 035107 (2013)

Ab initio analysis of the tight-binding parameters and magnetic interactions in Na_2IrO_3

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… the nnKH model, is, apparently, ~~inadequate.~~ *insufficient*

Yes, indeed

Spatially extended, „quasimolecular" orbitals: longer-range couplings $J_{2,3}$

Yes, indeed

Na_2IrO_3

(current status)

Data collected so far suggests that

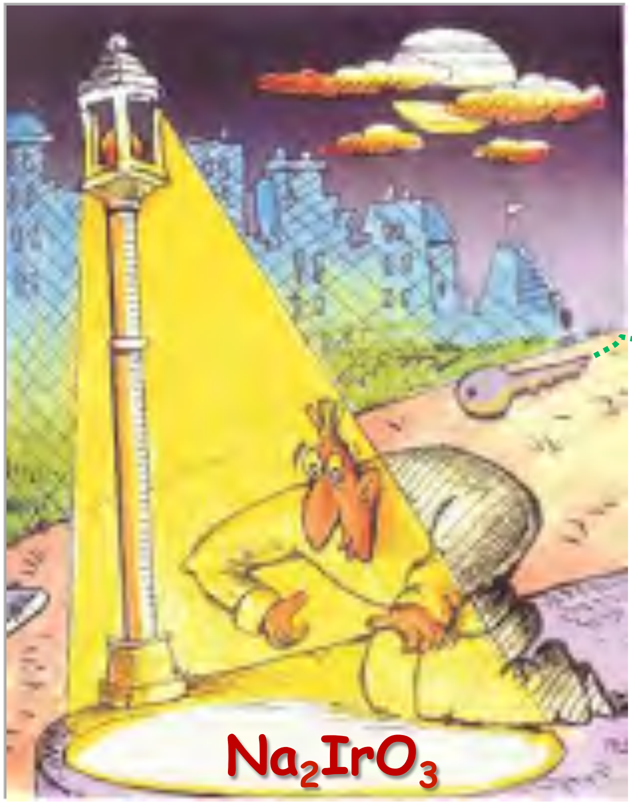
- Kitaev term seems to be dominant
- Other terms are substantial, yet to be sorted out

measure and fit,
measure and fit…

most wanted:
single-crystal
S(q,w)

for more details, see:
Chaloupka & GKh, condmat 2015

The streetlight effect



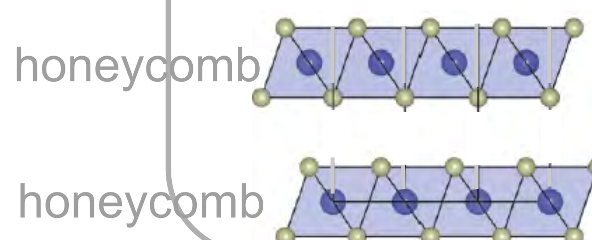
-Did you lose your Majoranas here?

-No, but the light is much better over here !

New candidate:

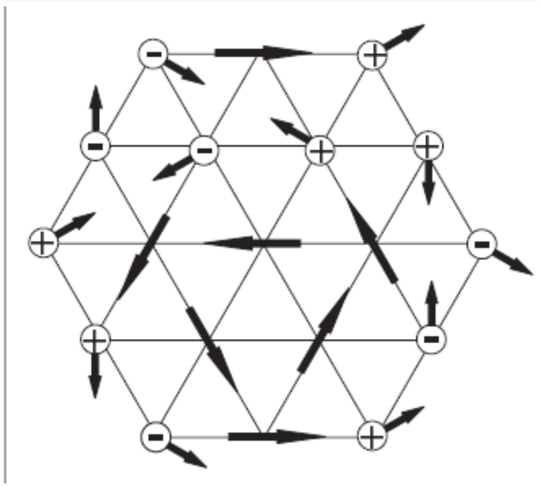
$RuCl_3$

Plumb et al, 2014



…and look for J=1/2 K-J model on other lattices

GKh (PTPS, 2005)

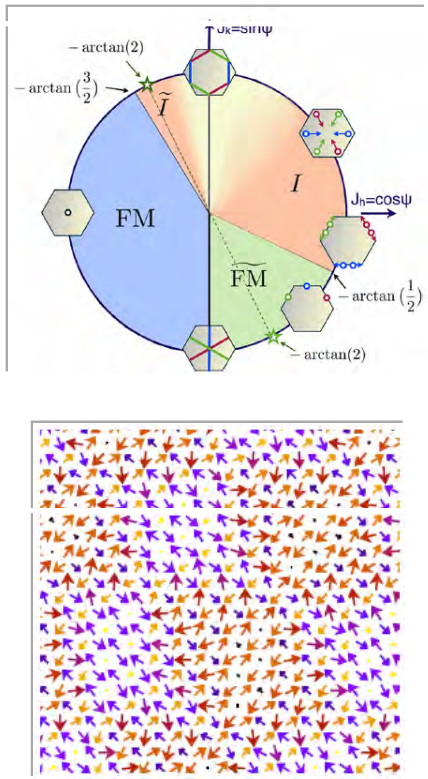


spin vortex condensate

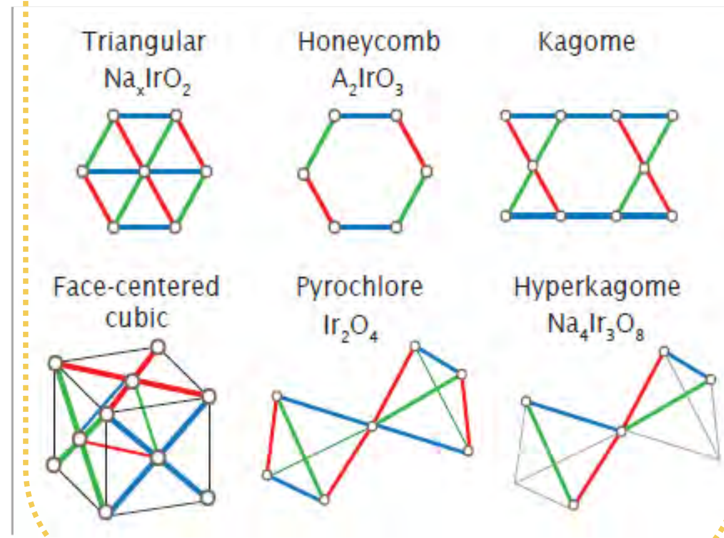
multi-Q order

„hidden" Goldstone

Rousochatzakis et al. (condmat, 2012)



Kimchi & Vishwanath (PRB, 2014)



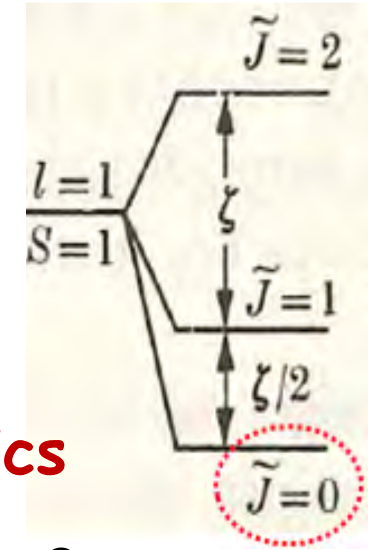
Pseudospin *AND* geometrical frustration:

- amplify the chances to find exotic states !

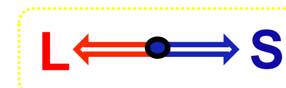
…farther away from the streetlight

d^5 Co, Rh, Ir

J=0 physics



d^4 Ru, Os,..



„nonmagnetic" Mott insulators

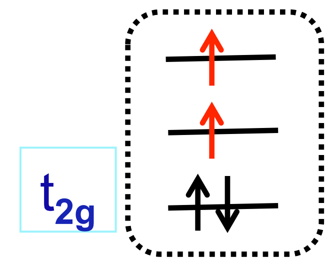
d^4 Mott insulators

Re^{3+}, Ru^{4+}, Os^{4+}, Ir^{5+}

4d, 5d electrons

1. Low-spin $S=1$

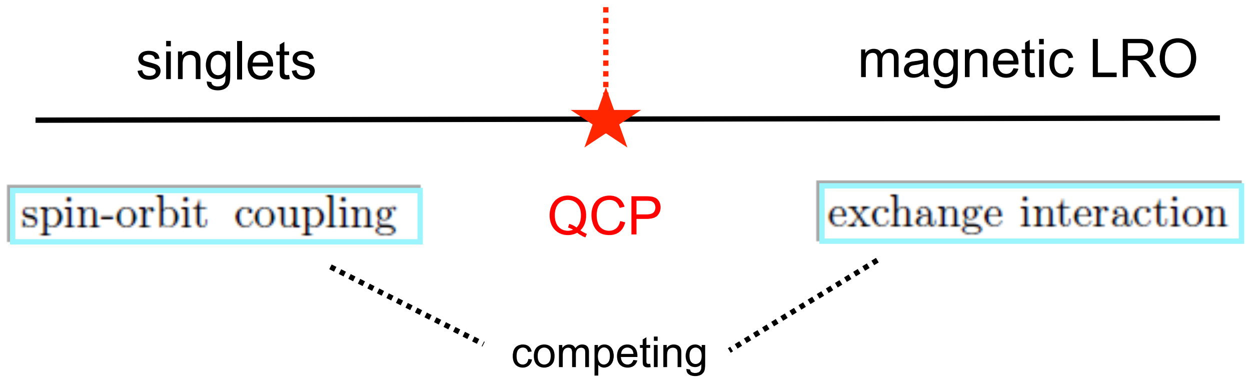
2. Unquenched $L=1$

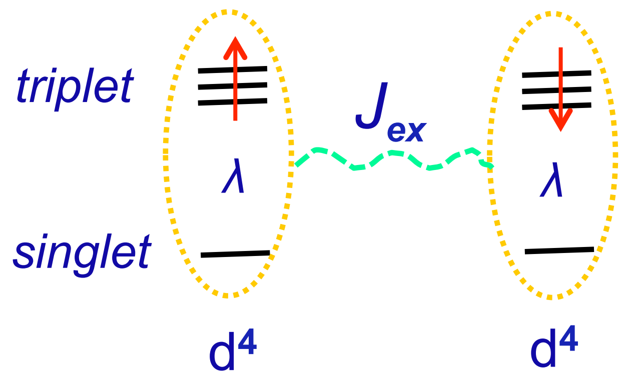


$\lambda(LS)$ ⇨ $J=S+L=0$

…no spin left to play with …

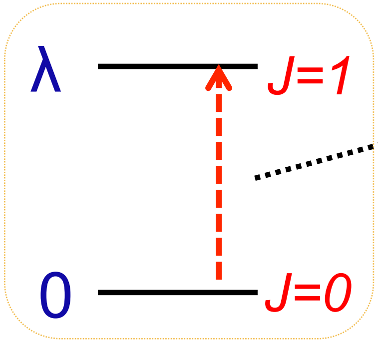
J=0 physics: *spin-orbit driven magnetic QCP*





d^4 ion: Van-Vleck magnetism

(i) *There are no „pre-existing“ moments*



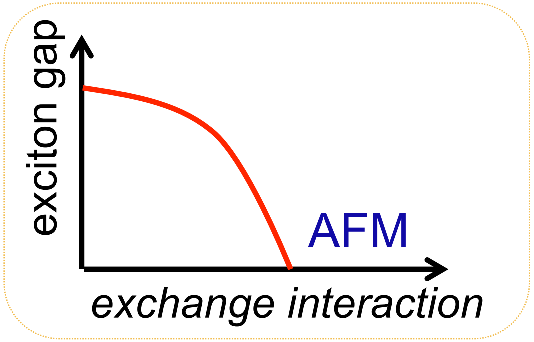
(ii) J=0 *to* J=1 *transition: off-diagonal magnetic moment* M=2S-L

(„spin-orbit exciton“)

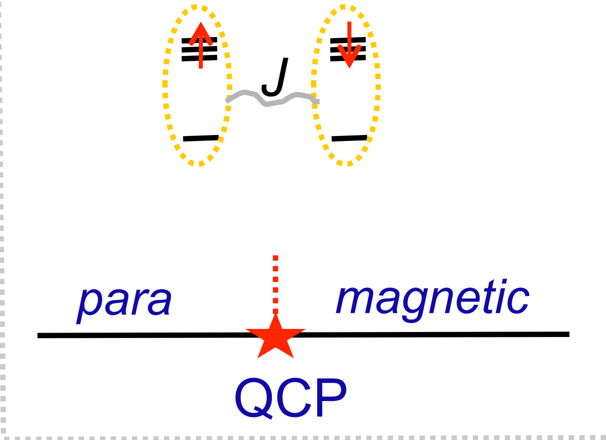
(iii) *Condensation of spin-orbit exciton*

⇩

„excitonic“ magnetism

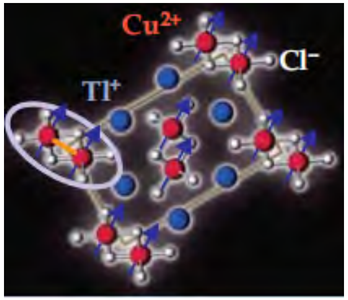


Singlet-triplet examples

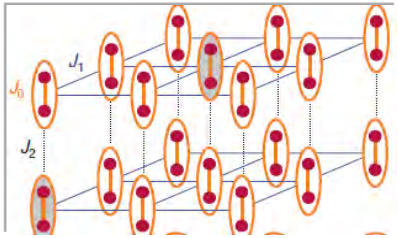


(A) Weakly coupled dimers

$TlCuCl_3$



Bilayer AF



THIERRY GIAMARCHI[1]*, CHRISTIAN RÜEGG[2]*
AND OLEG TCHERNYSHYOV[3]* *(Nat.Phys. 2009)*

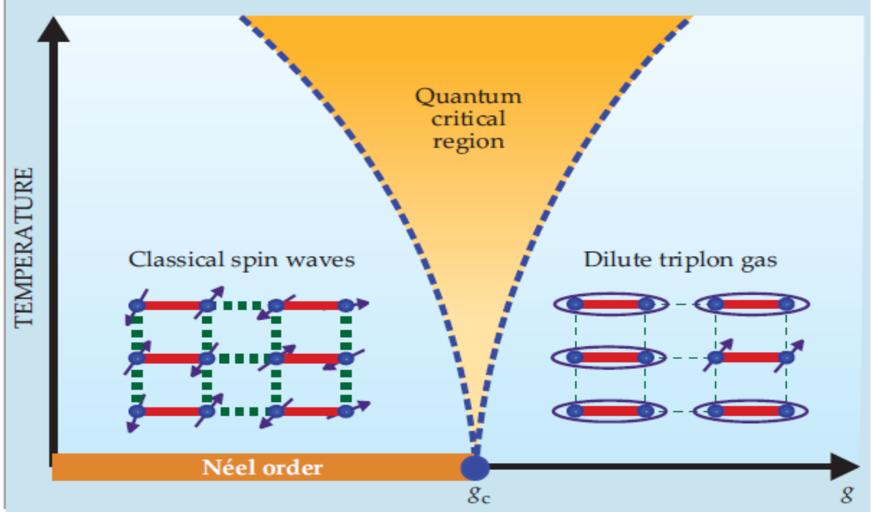
(B) 4f Pr compounds (broad literature since 1970's)

(C) e_g orbital $FeSc_2S_4$ *(Chen, Balents, Schnyder, 2009)*

(D) Spin-state-crossover in Fe-based SC (*Chaloupka & GKh, 2013)*

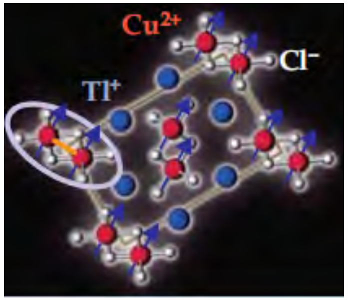
QUANTUM CRITICALITY

Sachdev, Keimer, Phys.Today, 2011



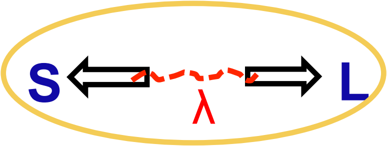
birthplace for „unconventional" physics

Inter-ionic dimers



-small energies
-special geometry

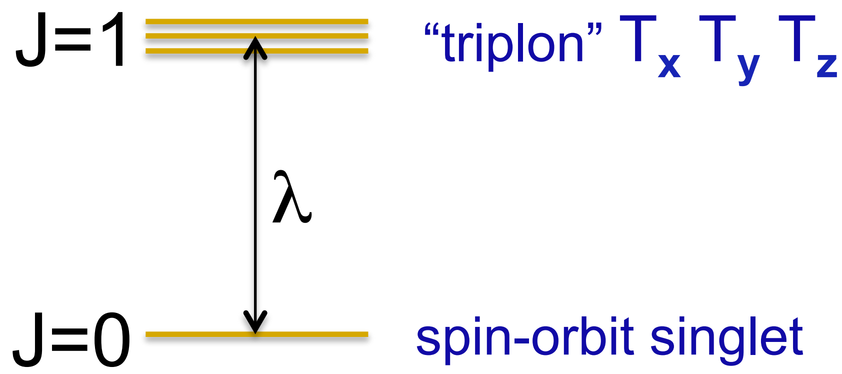
d^4 : Intra-ionic „dimer" made of S and L



1) energetic *(~100 meV)*
2) generic *(any lattice)*

GKh (2013)

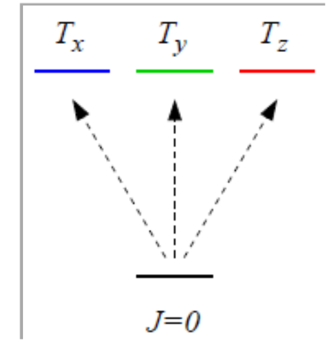
d^4 Mott insulator: single-ion states



d^4 Mott insulator: singlet-triplet model (180°)

GKh (2013)

(e.g. 214-perovskite)

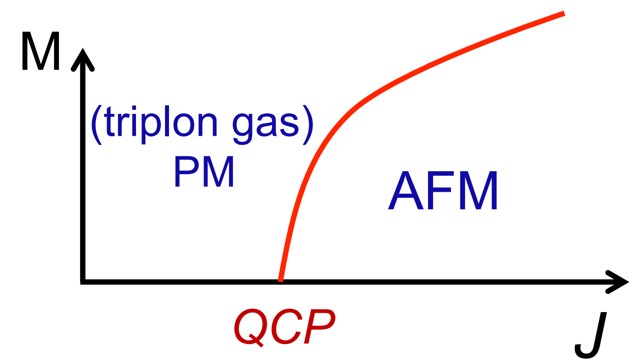


$$H = \lambda \sum_i n_i + J \frac{2}{9} \sum_{ij} [T_i^\dagger \cdot T_j - \frac{7}{16}(T_i \cdot T_j + H.c.)]$$

spin-orbit

exchange

S=1 boson



LRO moment: $M_z = 2\sqrt{6\rho(1-\rho)}$

$\rho = \frac{1}{2}(1-\tau^{-1})$

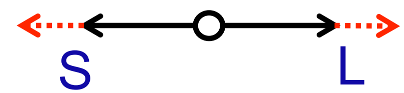
cond.density

$\tau = \kappa/\kappa_c > 1$

distance from QCP

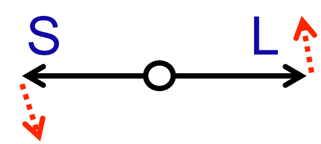
Excitations:

1. The amplitude mode
   changing the lengths of S & L



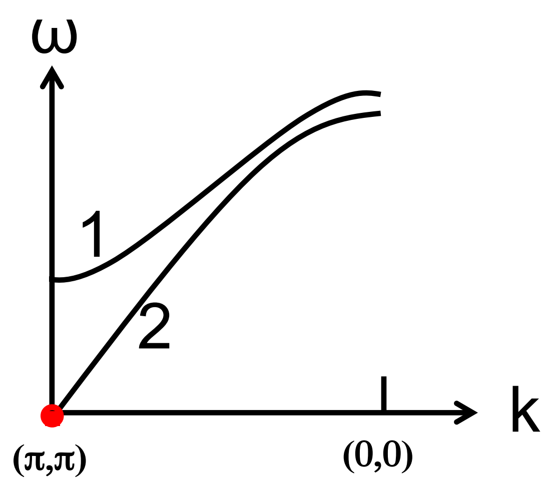
„Higgs“

2. The phase modes
   in-phase rotation of S & L



Goldstone

magnons in excitonic AF



Excitations:

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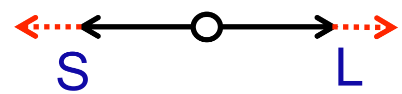

„Higgs“

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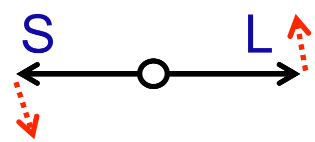
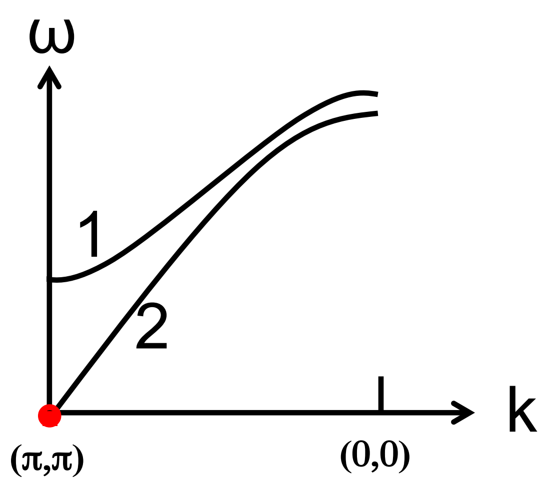
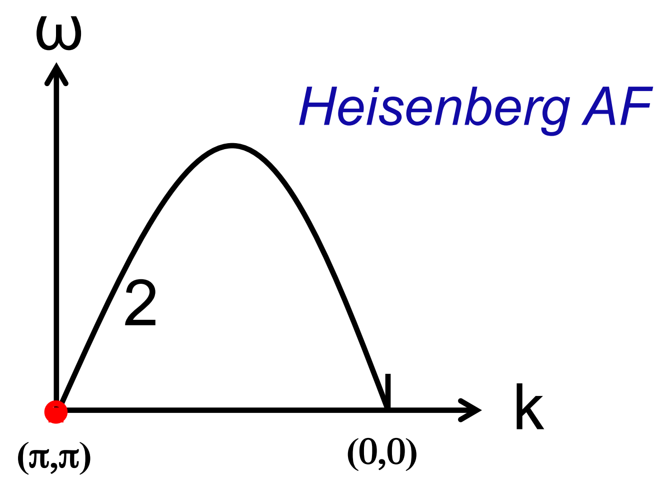


Goldstone

excitonic AF



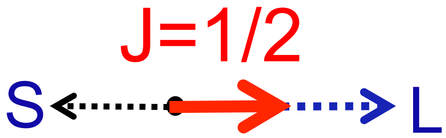


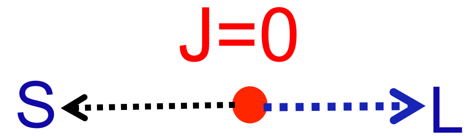
Van Vleck-type d^4 Mott insulators:
EXCITONIC magnetism

Candidate material:

Ca_2RuO_4

Summary





J=S+L inherits bond-dependent & frustrated nature of orbitals

„nonmagnetic" J=0 Mott insulators

unconventional magnetism

…unconventional SC?

Phys. Rev. Lett. **87**, 077202 (2001)

Spin-Orbit Coupling in the Mott Insulator Ca_2RuO_4

T. Mizokawa,[1,2] L. H. Tjeng,[2] G. A. Sawatzky,[2] G. Ghiringhelli,[3,4] O. Tjernberg,[3,5] N. B. Brookes,[3] H. Fukazawa,[6] S. Nakatsuji,[6] and Y. Maeno[6,7]

$\langle \sum_i l_{x'}(i) \cdot s_x(i) \rangle$ is about -0.28 ± 0.07, using $N_d = 4$. This value is comparable to that measured for CoO [16], where the orbital moment was found to be of the order of 1 μ_B. These measurements thus demonstrate that the

unquenched L-moment in ruthenates

PRL **112**, 127002 (2014)

Spin-Orbital Entanglement and the Breakdown of Singlets and Triplets in Sr_2RuO_4

C. N. Veenstra,[1] Z.-H. Zhu,[1] M. Raichle,[1] B. M. Ludbrook,[1] A. Nicolaou,[1,2,7] B. Slomski,[3,4] G. Landolt,[3,4] S. Kittaka,[5,6] Y. Maeno,[5] J. H. Dil,[3,4] I. S. Elfimov,[1,2] M. W. Haverkort,[1,2,7] and A. Damascelli[1,2,*]

$Singlet\text{-}triplet\ model\ H_{eff}(90°)$

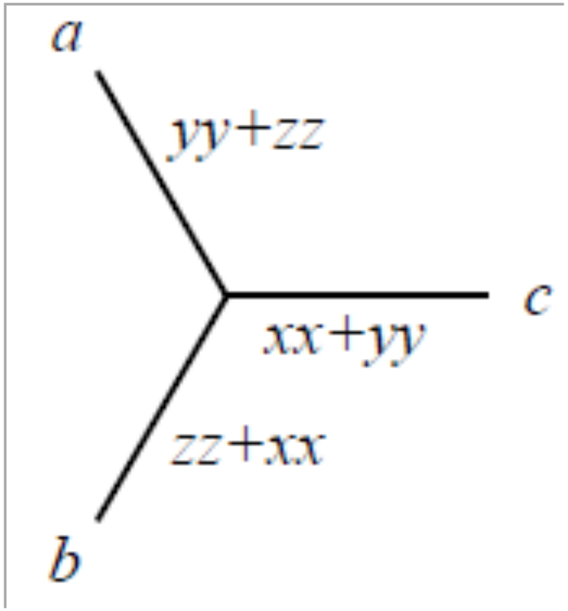
(triangular, honeycomb, kagome…)

c-bond exchange: $$\frac{2}{3}(T^{\dagger}_{ix}T_{jx} + T^{\dagger}_{iy}T_{jy}) - \frac{5}{6}(T_{ix}T_{jx} + T_{iy}T_{jy}) + H.c.,$$

hopping — pair-generation

x and y type bosons only involved

Bond-dependent „xy-model“:



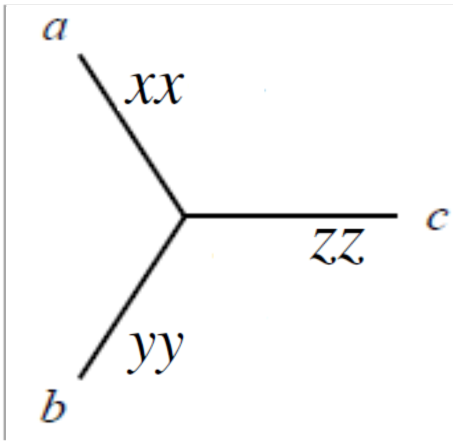
Exchange anisotropy, 90° bonding: d^4 vs d^5

(triangular, honeycomb, kagome…)

d^5 (Co,Rh,Ir)

bond-dependent Ising:

spin ½

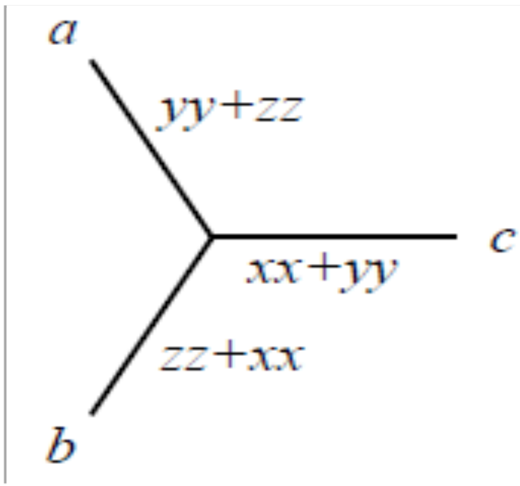


GKh (2005)
Chen & Balents (2008)
Jackeli & GKh (2009)

d^4 (Ru,Re,Os)

bond-dependent „xy“:

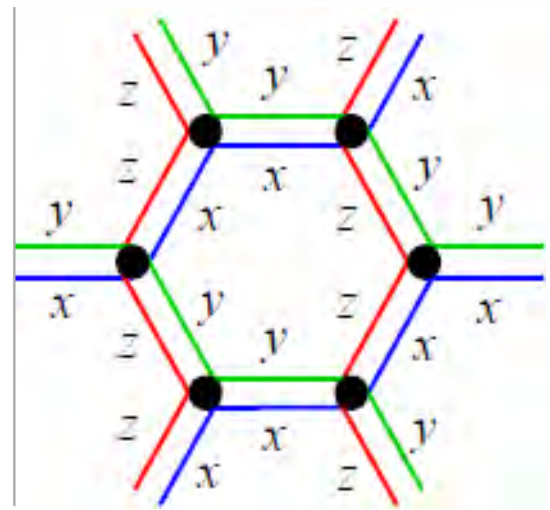
spin-one T boson



GKh (2013)

Honeycomb lattice: dimensionality reduction

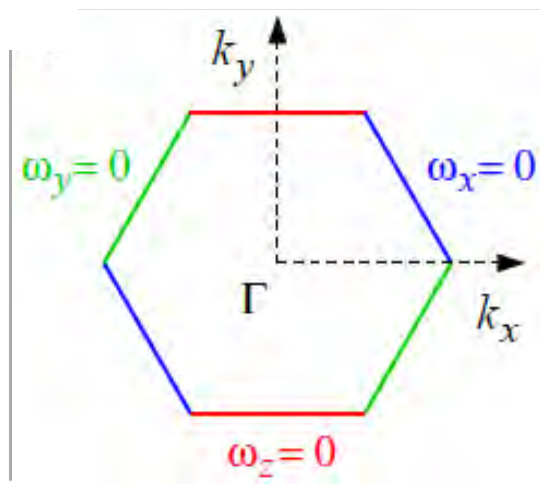
- *Each flavor T_x, T_y, T_z has its own zigzag to move along*



- *1D dispersion:*

$$\omega_z(\boldsymbol{k}) \equiv \omega_z(k_y) \simeq \lambda\sqrt{1 + (\kappa/\kappa_c)c_y}$$

- J=J_{crit} : *zero-energy lines*



unusual singlet-triplet model, yet to be solved…

VBS?
zigzag order?
spin nematic?
…