Magnetism and doping effects in spin-orbit coupled Mott insulators

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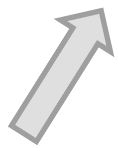
1. *Introduction:* spin-orbit coupled magnets
2. *d^5 ions:* perovskite & honeycomb iridates
3. *d^4 ions:* excitonic magnetism (ruthenates)

d-electron in TM compounds



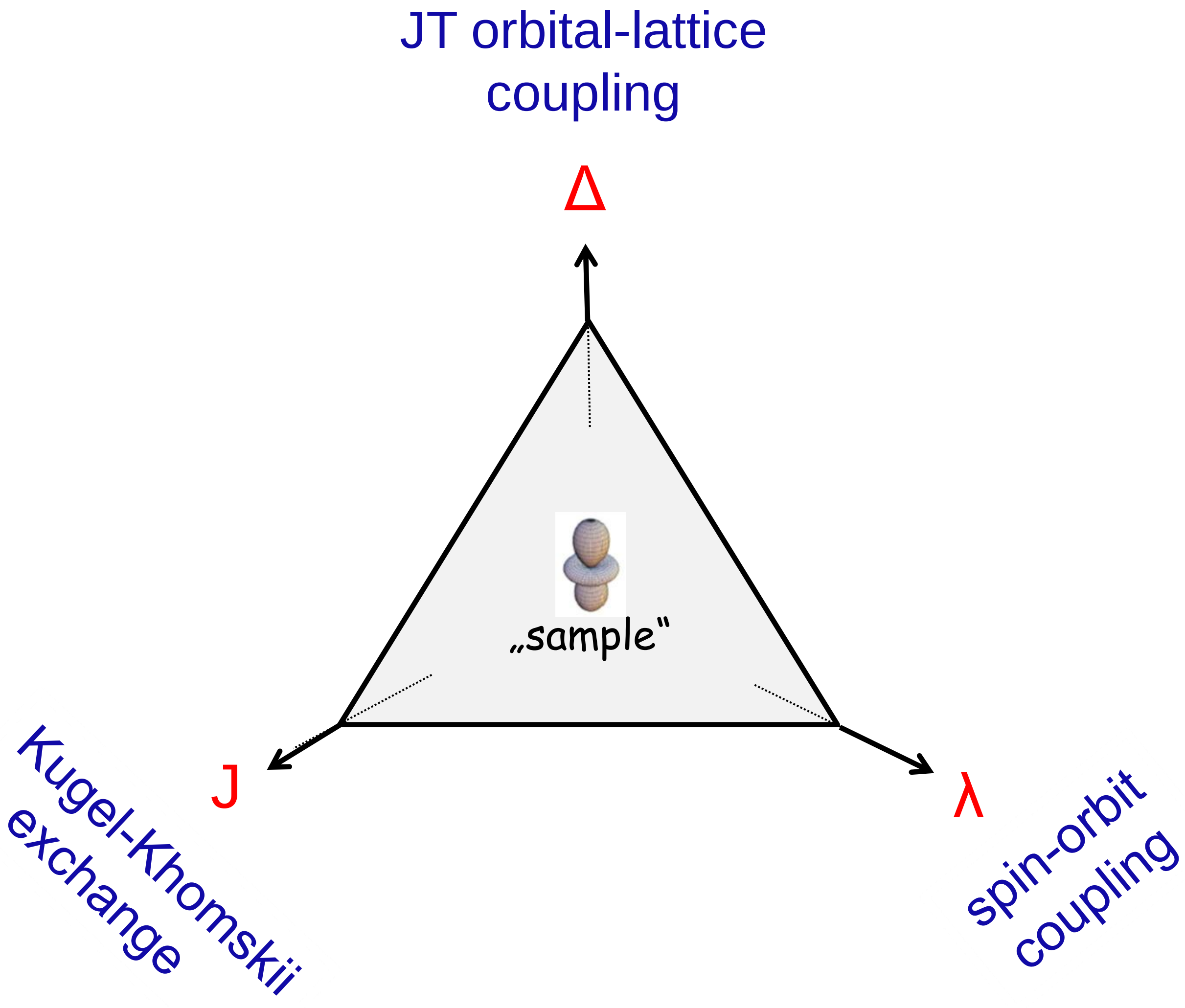
Magnetism and the Chemical Bond

BY JOHN B. GOODENOUGH

Interscience Publ. 1963

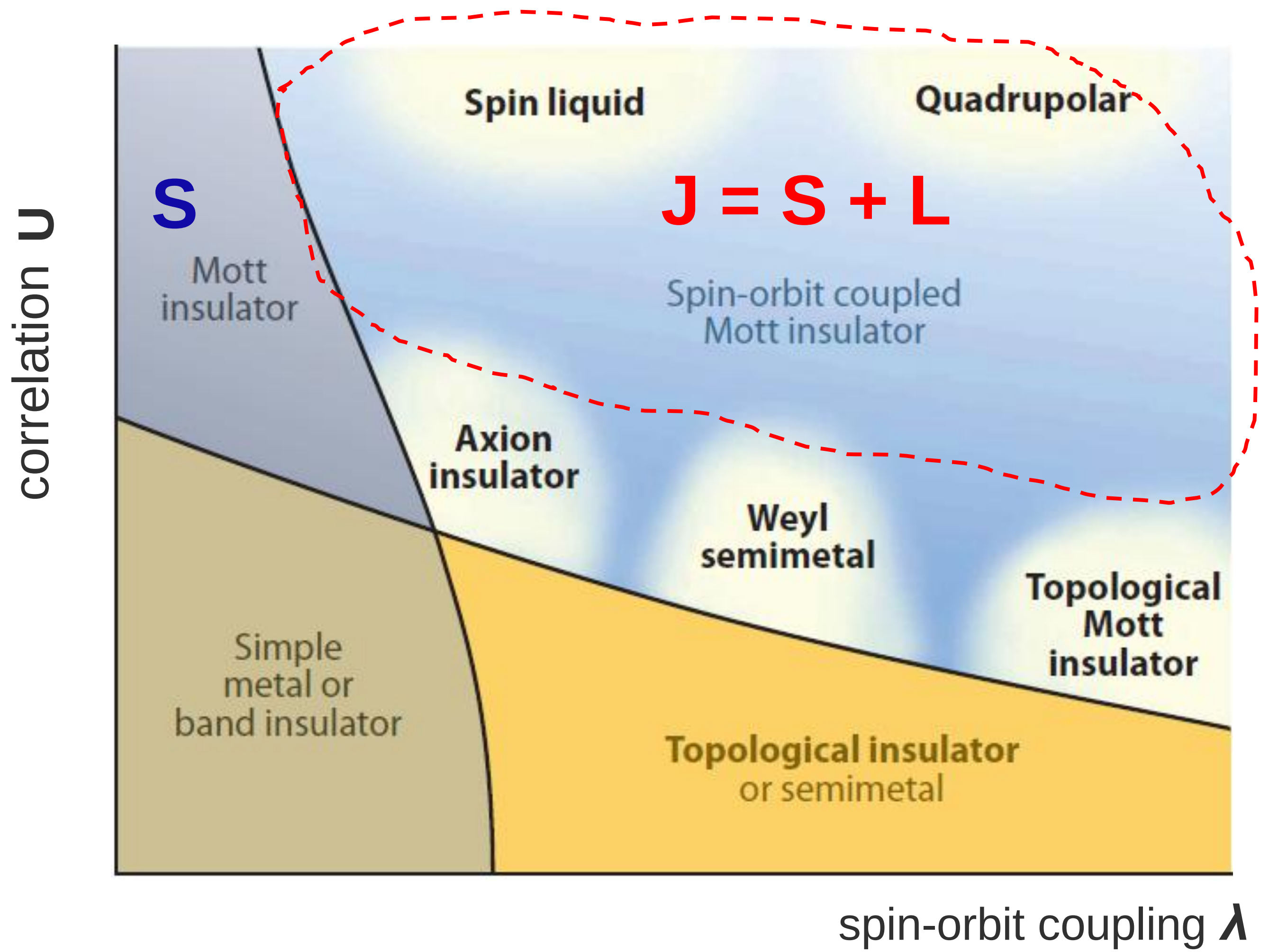
Goodenough-Kanamori
rules

Three competing interactions: Δ, J, λ (<< W~U)

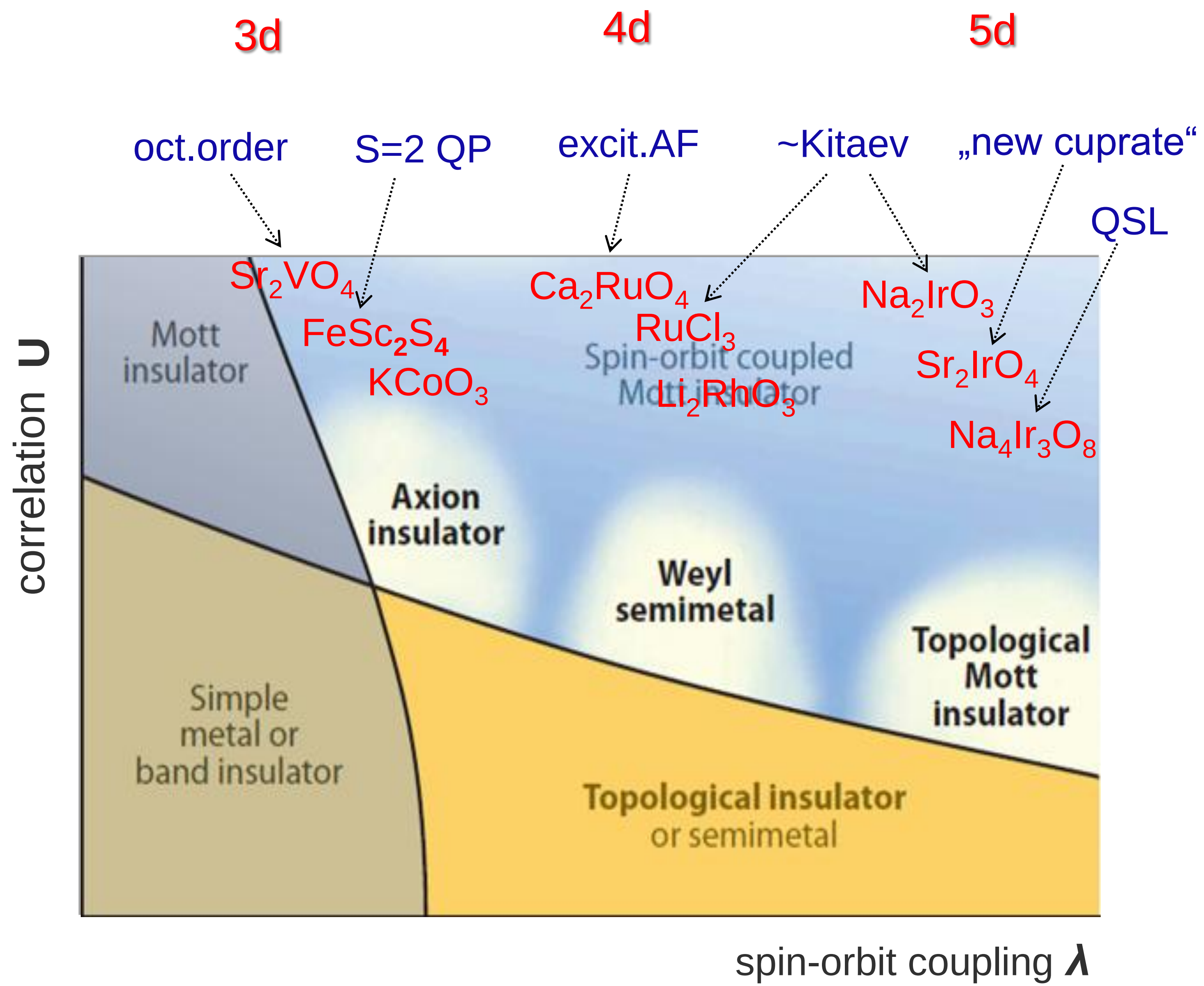


Witczak-Krempa, Chen, Y-B.Kim, Balents (2014)

Hubbard model with spin-orbit coupling

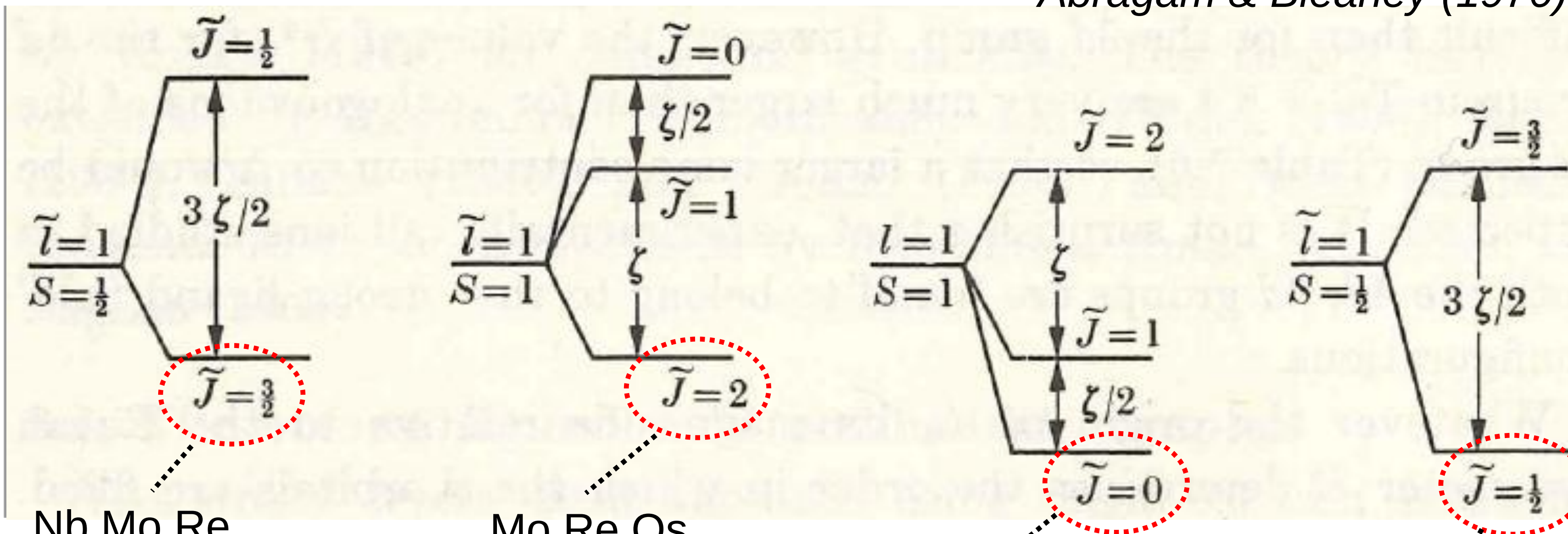


Spin-orbit magnets: some examples



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“

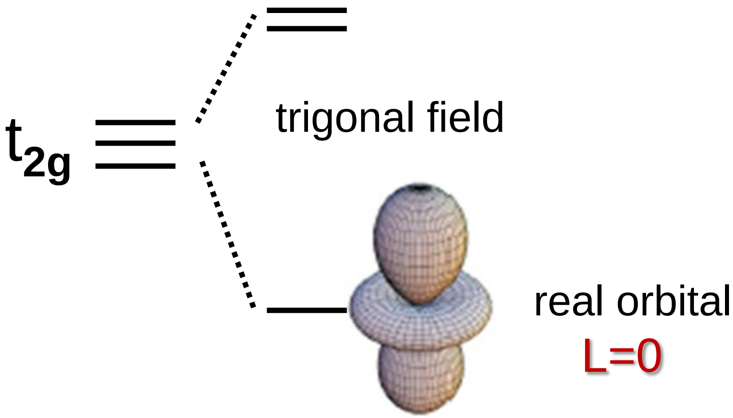
Co,Rh,Ir
quantum spin

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

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

Lifting the orbital degeneracy

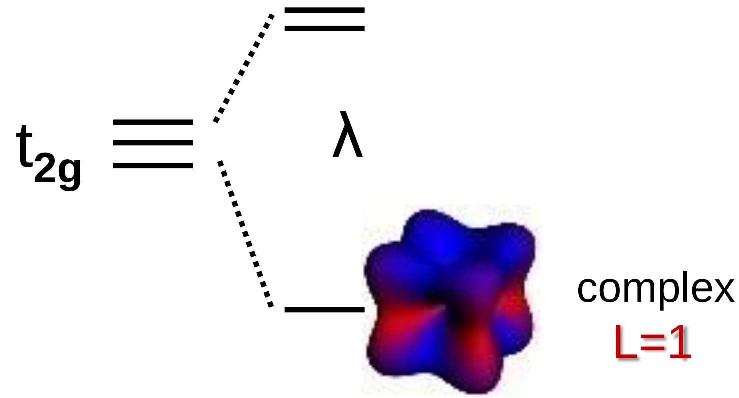
Jahn-Teller



$$|xy + yz + zx\rangle \times |\Uparrow\rangle$$

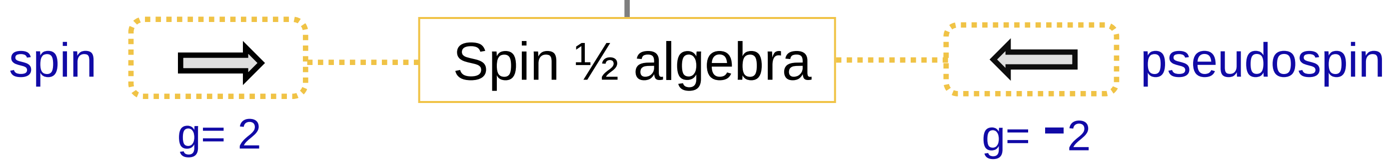
quadrupole — spin

Spin-orbit

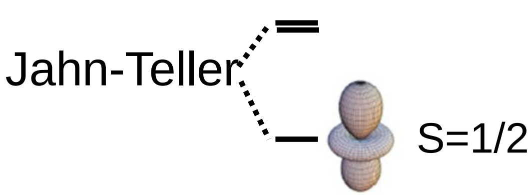


$$|xy \uparrow\rangle + |yz \downarrow\rangle + i|zx \downarrow\rangle$$

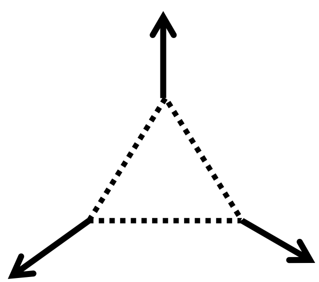
pseudospin J=S+L



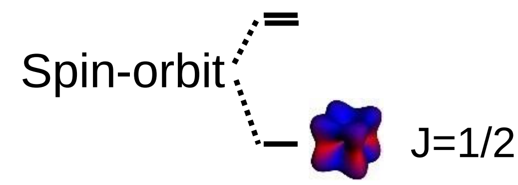
Magnetism



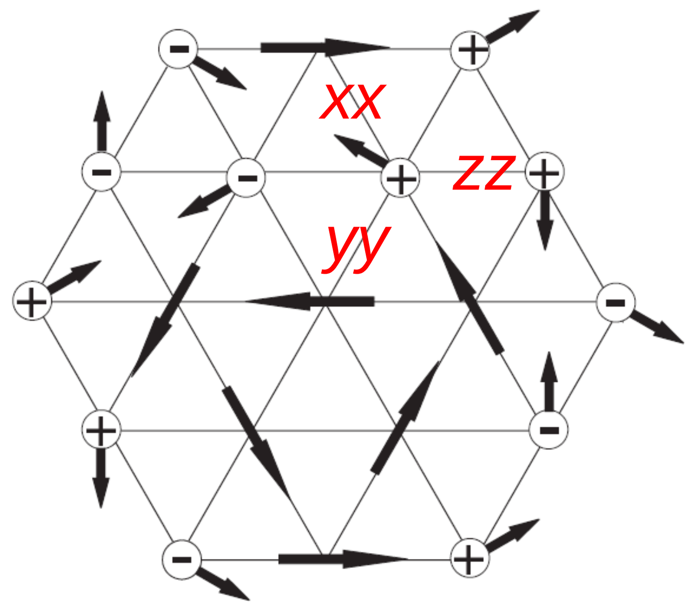
H = (SS)



coplanar,
single-Q



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



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

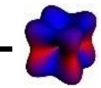
GKh (PTPS, 2005)

SC pairing

Jahn-Teller

S=1/2

Spin-orbit



J=1/2

Spin-singlet d-wave

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

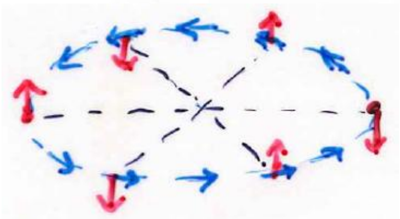
Baskaran (2003)
Kumar, Shastry (2003)
Ogata (2003)
P. Lee et al.(2004)

Pseudospin-triplet p+f wave

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

nondegenerate

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



$d_x , d_y , d_z \neq 0$

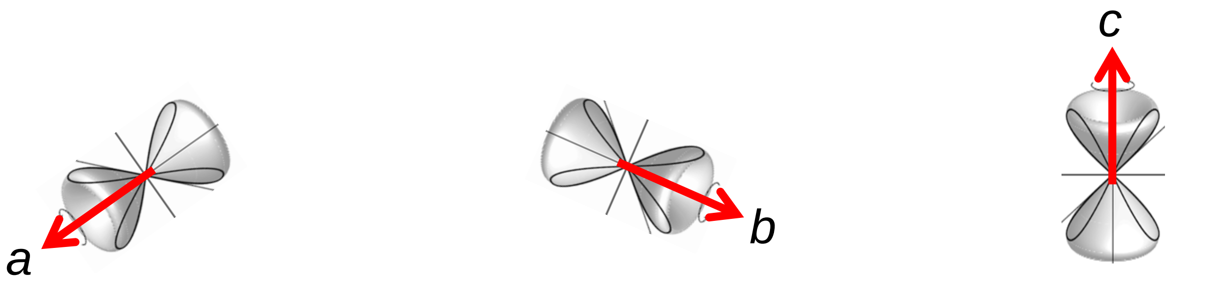
„ p + f “

Knight-shift finite in all directions

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

The origin of „unconventionality“: ORBITAL magnetism

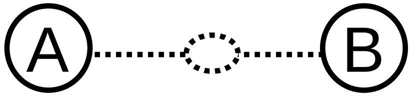
Orbital moment **L** has a „shape“:



$L_x=1$ $L_y=1$ $L_z=1$

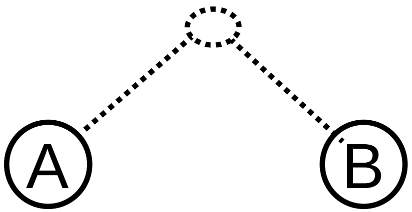
$x(y+iz)$ $y(z+ix)$ $z(x+iy)$

Hopping amplitude:



real

(inversion)



complex

(no inversion)

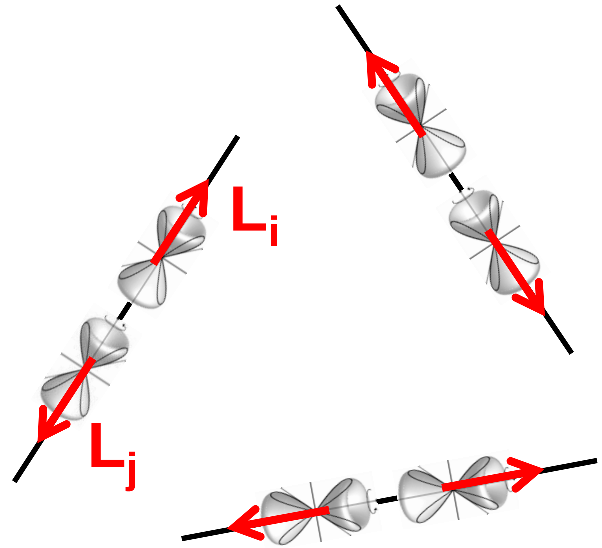
nontrivial band topology
Shitade *et al.* (2009)

ORBITAL MAGNETISM

Orbital moment **L** interactions:

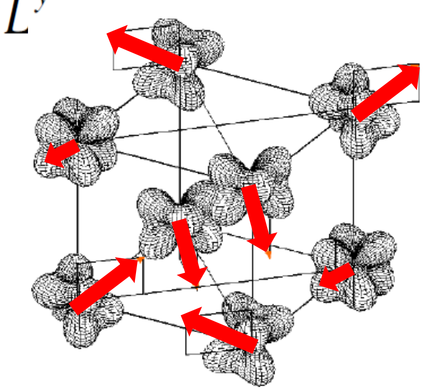
non-Heisenberg

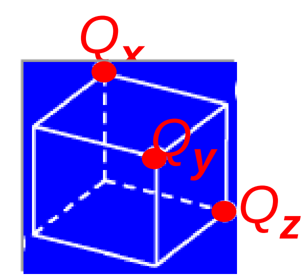
bond-dependent



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

cubic lattice:



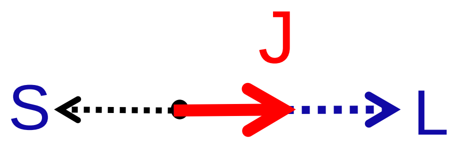


GKh & Okamoto (2002)

non-coplanar

multi-Q

no LRO at finite T



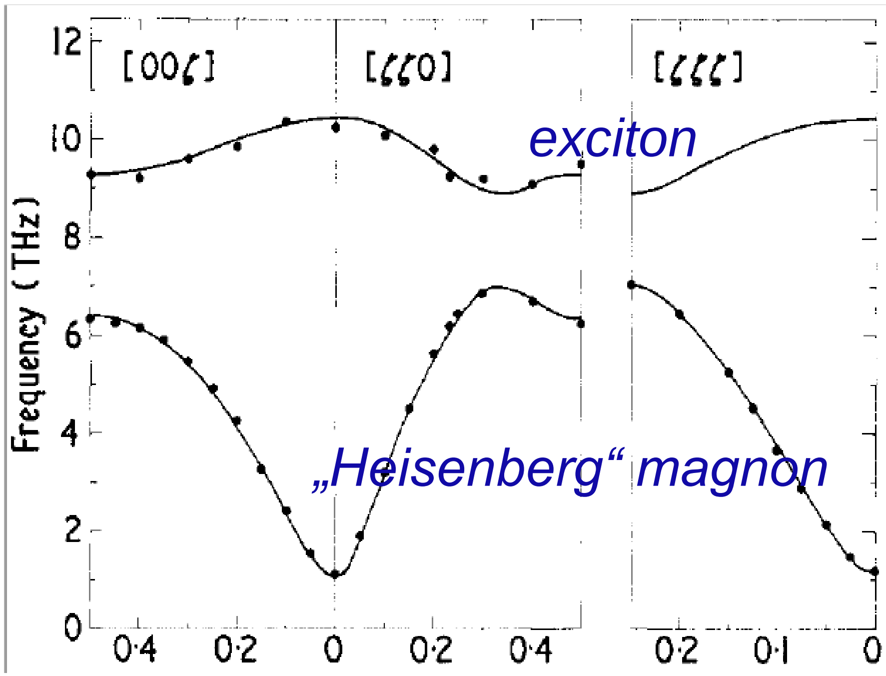
Pseudospin J=S+L inherits bond-dependent and frustrated nature of orbitals

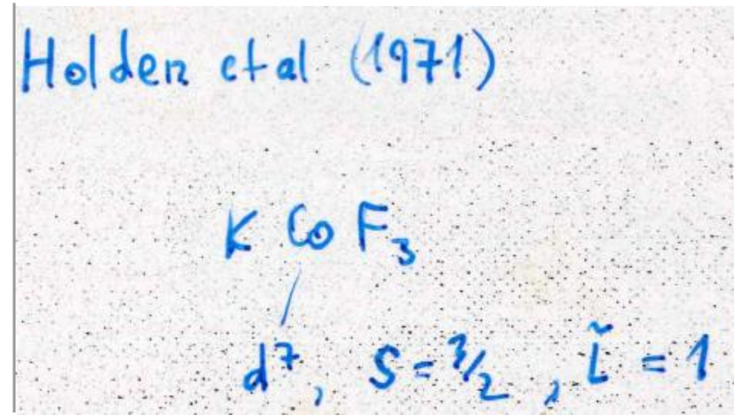
⇓

„unconventional“ magnetism

The „old“ pseudospin J=1/2

3d Co-fluoride, neutron scattering

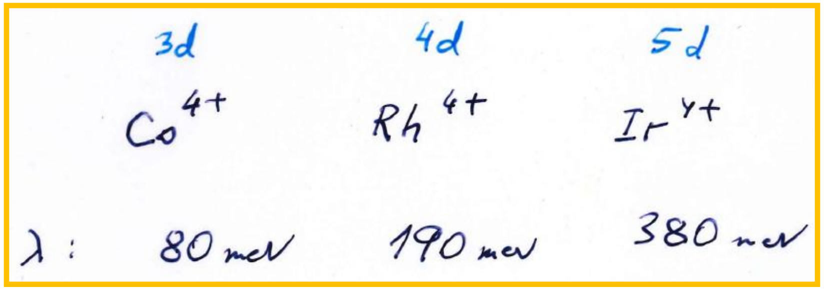


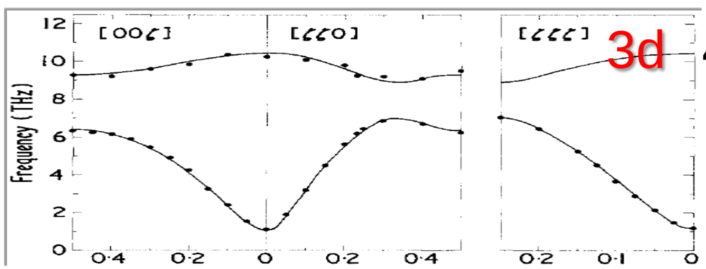


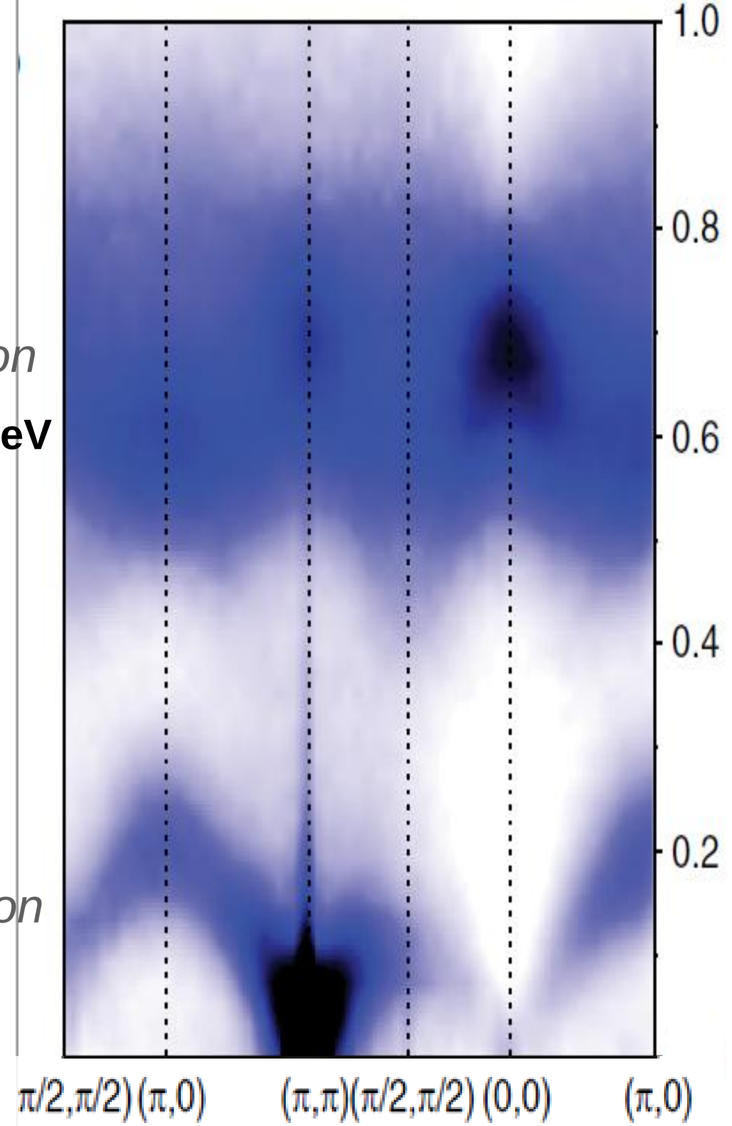
Two branches split by spin-orbit:
magnon & exciton

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

B.J.Kim, Takagi, …







$KCoF_3$

Holden *et al.* (1971)

neutron scatteting

Sr_2IrO_4

Kim *et al.* (2012)

RIXS data

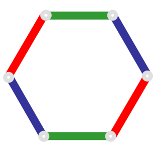
J=1/2 magnetism in iridates: *basic theory*

Jackeli & GKh (2009)

Two-parameter Hamiltonian = Kitaev + 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)

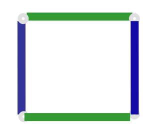


= *Kitaev model (2006)*

⇩

„Majorana world“ ?

dominant in 180-bonding
(Sr_2IrO_4 perovskite)



= *„cuprate“ model*

⇩

high-T_c SC ?

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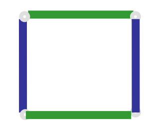

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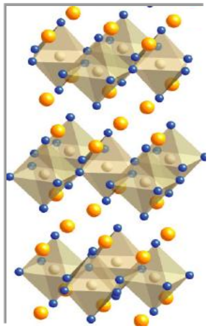
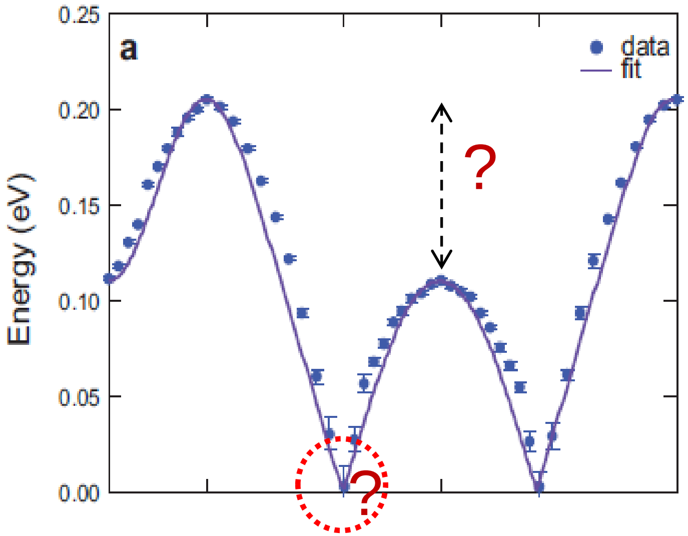
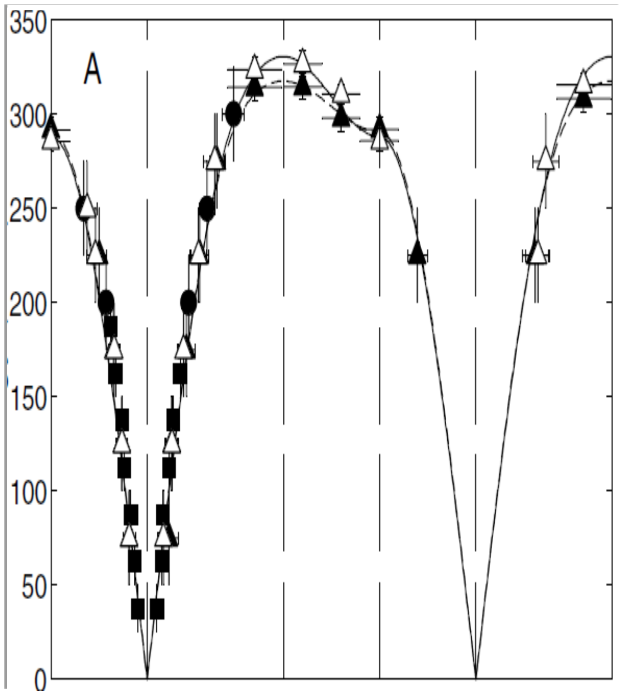


Sr_2IrO_4

Pseudospin ½ in perovskites:

-theory predicts
„nearly“ Heisenberg AF





Sr_2IrO_4
T_N~240 K

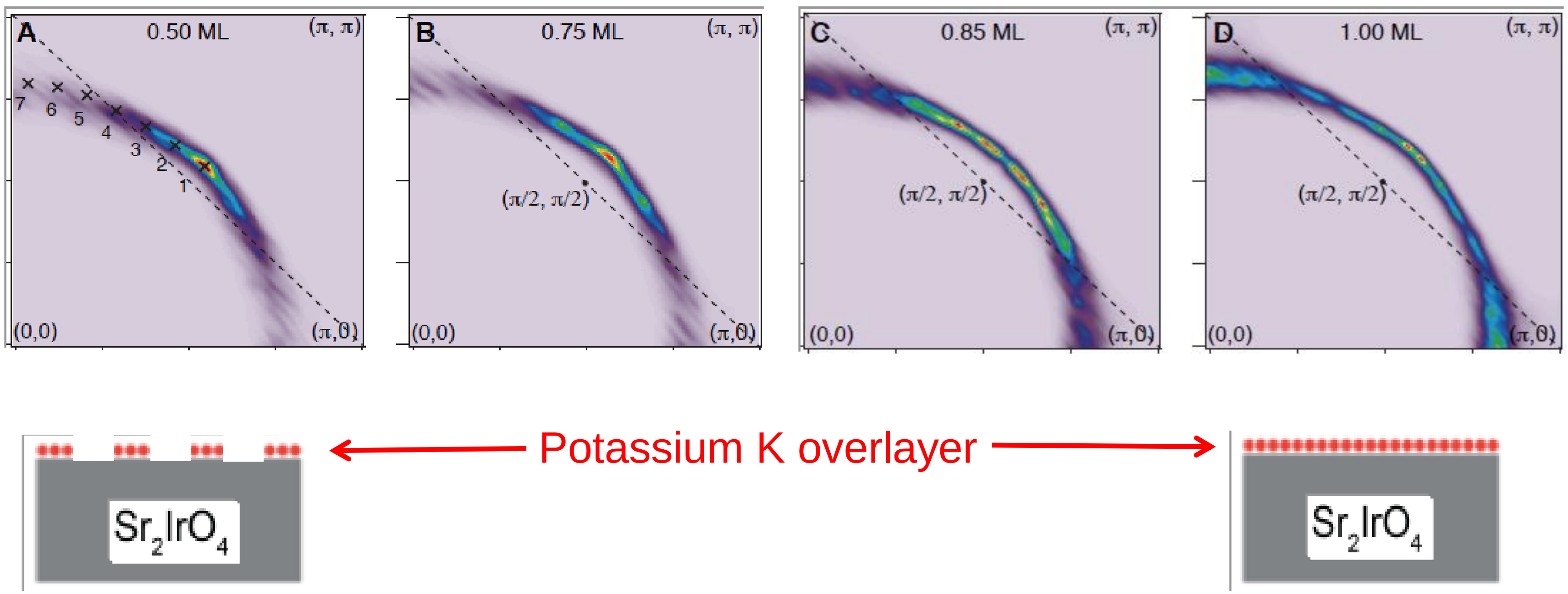
Kim et al.(2012)

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*

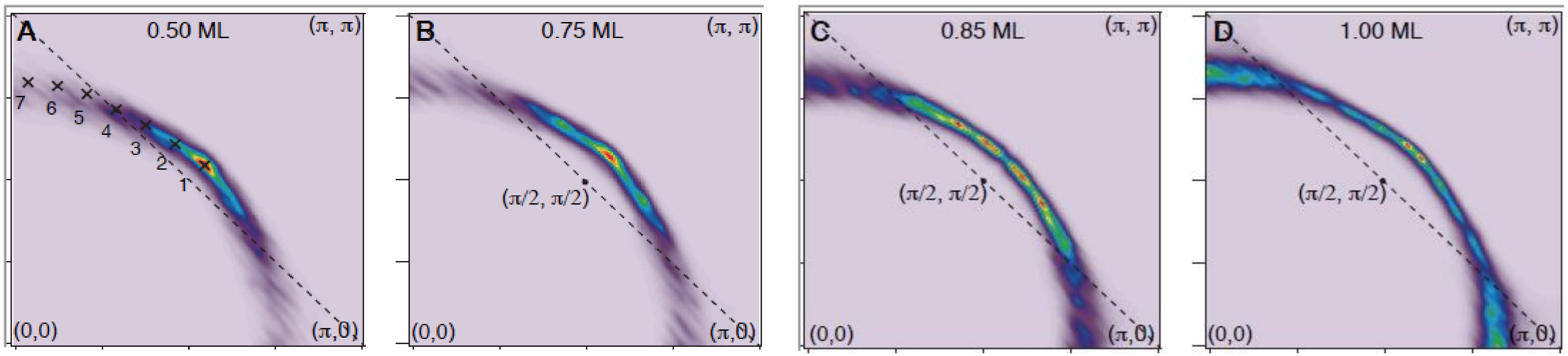


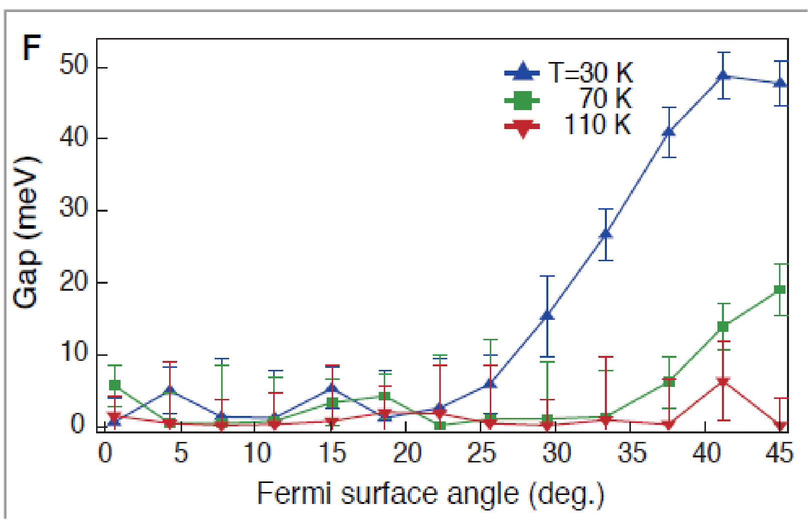
B.J. Kim *et al.* (Science 2014)

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?

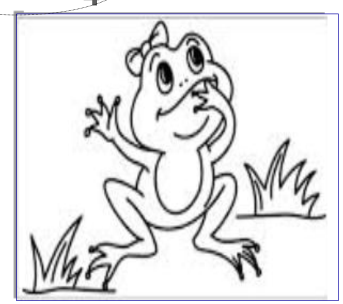
YES

SUPER!
GREAT!

NO

„...find 10 differences..."

Sr_2IrO_4



La_2CuO_4

YES or NO? -- no definite answer yet...

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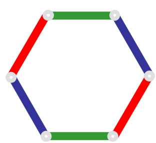

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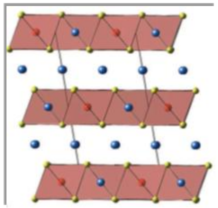
= *„cuprate“ model*

⇩

high-T_c SC ?

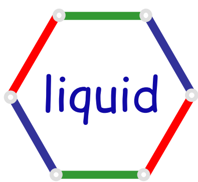


honeycomb lattice

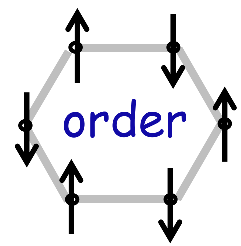
KH model

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

Kitaev model

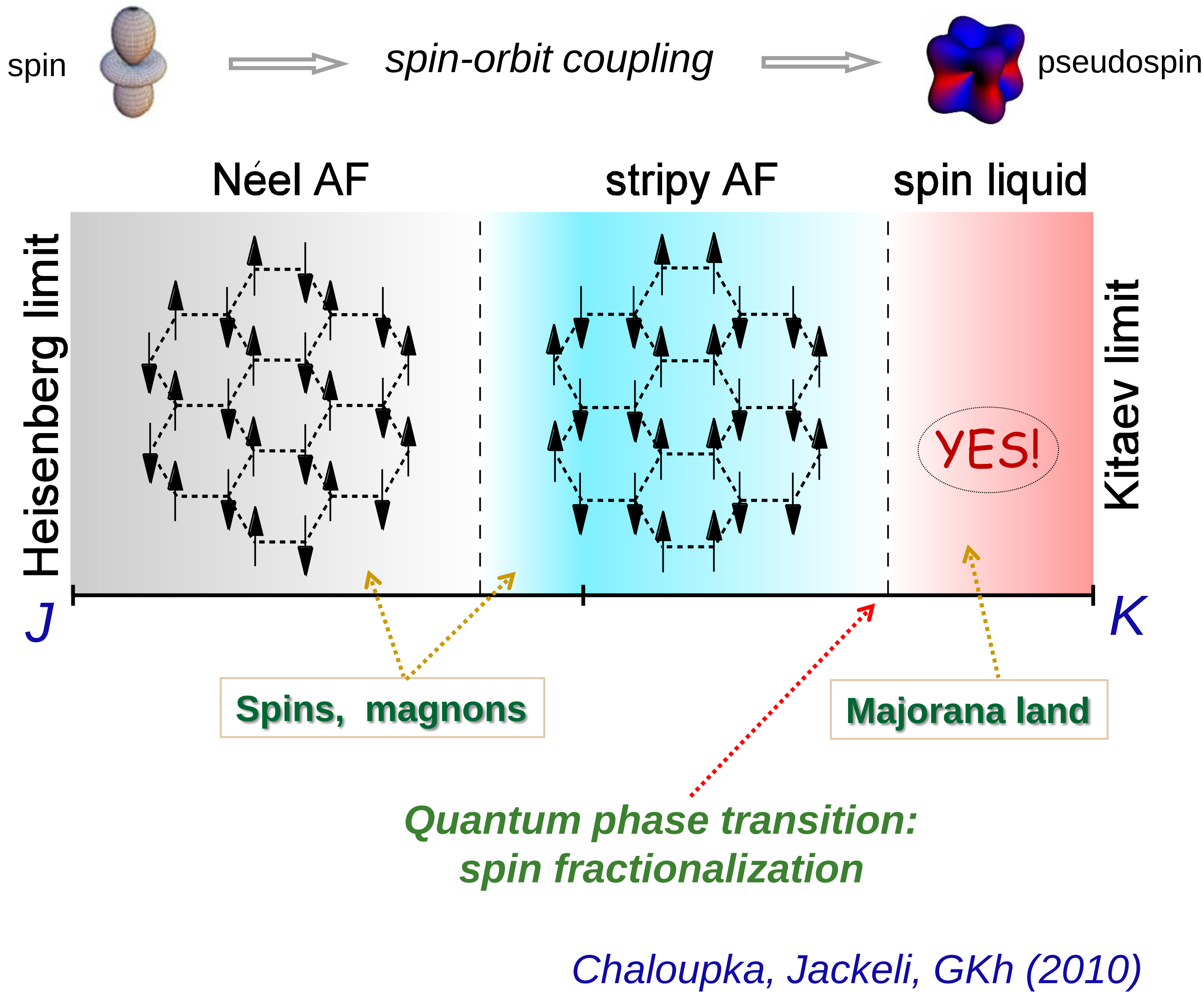


Heisenberg model



What is in between?

- if „some liquid“ 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: **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 model with large K > J

makes all these „for free“ but…

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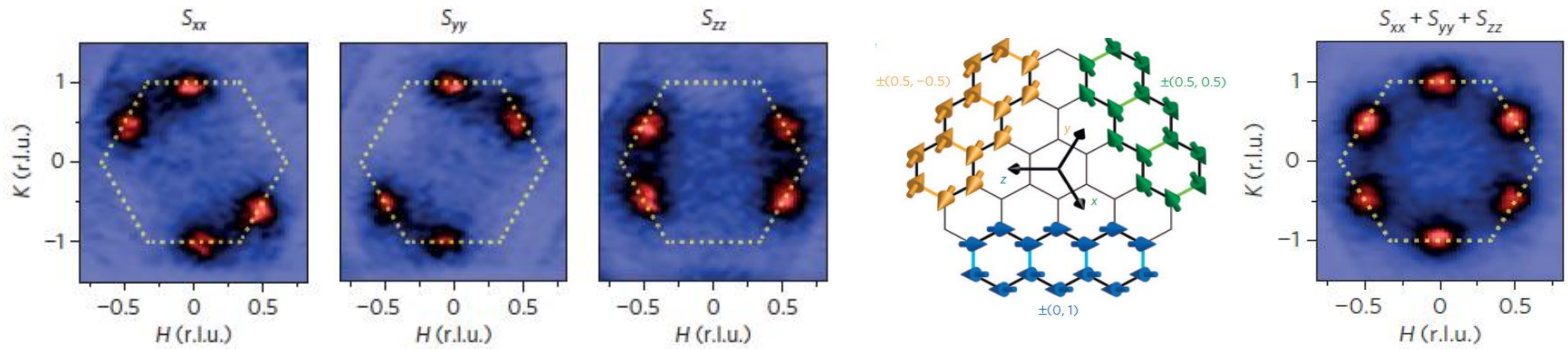
B.J. Kim et al. (2015): moment direction unexpected for KH model

Diffuse magnetic X-ray scattering intensities above T_N

B.J. Kim et al. (2015)

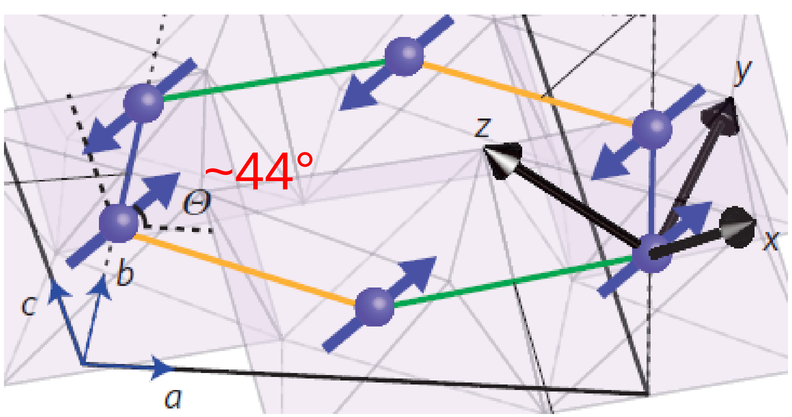


Each spin-component has its own Bragg-spot : non-Heisenberg

three equivalent zigzags

average

C3-symmetry involving both spin and lattice: natural for KH-model



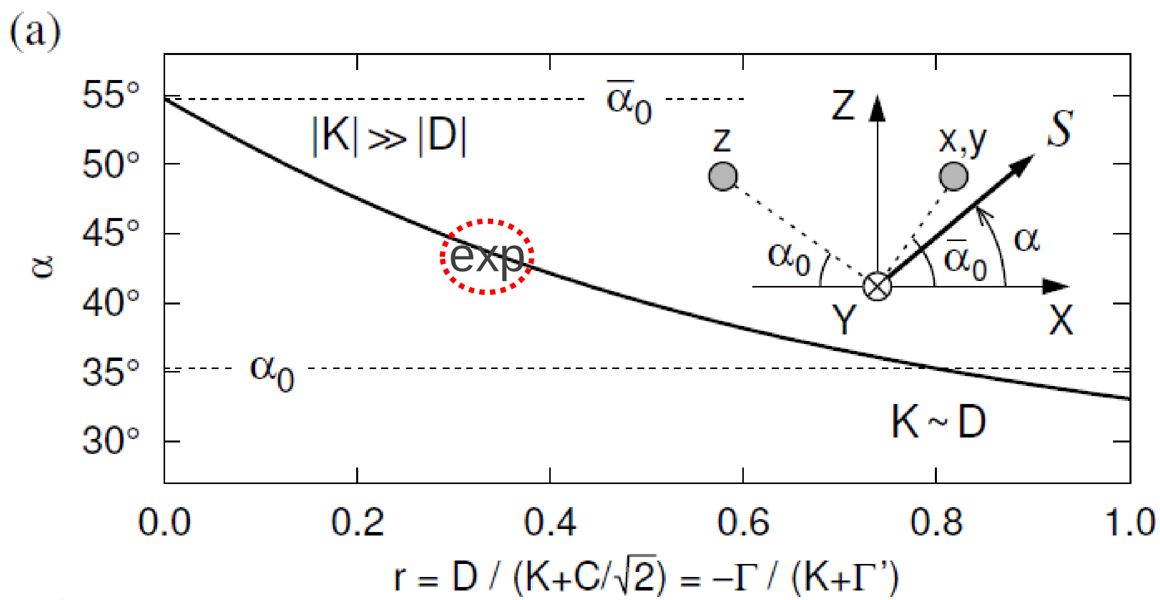
„Wrong“ easy axis:
spins look „nowhere“ away from the symmetry axes

NB: Kitaev term wants spin parallel to Ir-O bond

„Extended“ KH-model: *(H.-Y. Kee …; van den Brink …; Imada ...)*

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

Two new terms: D and C



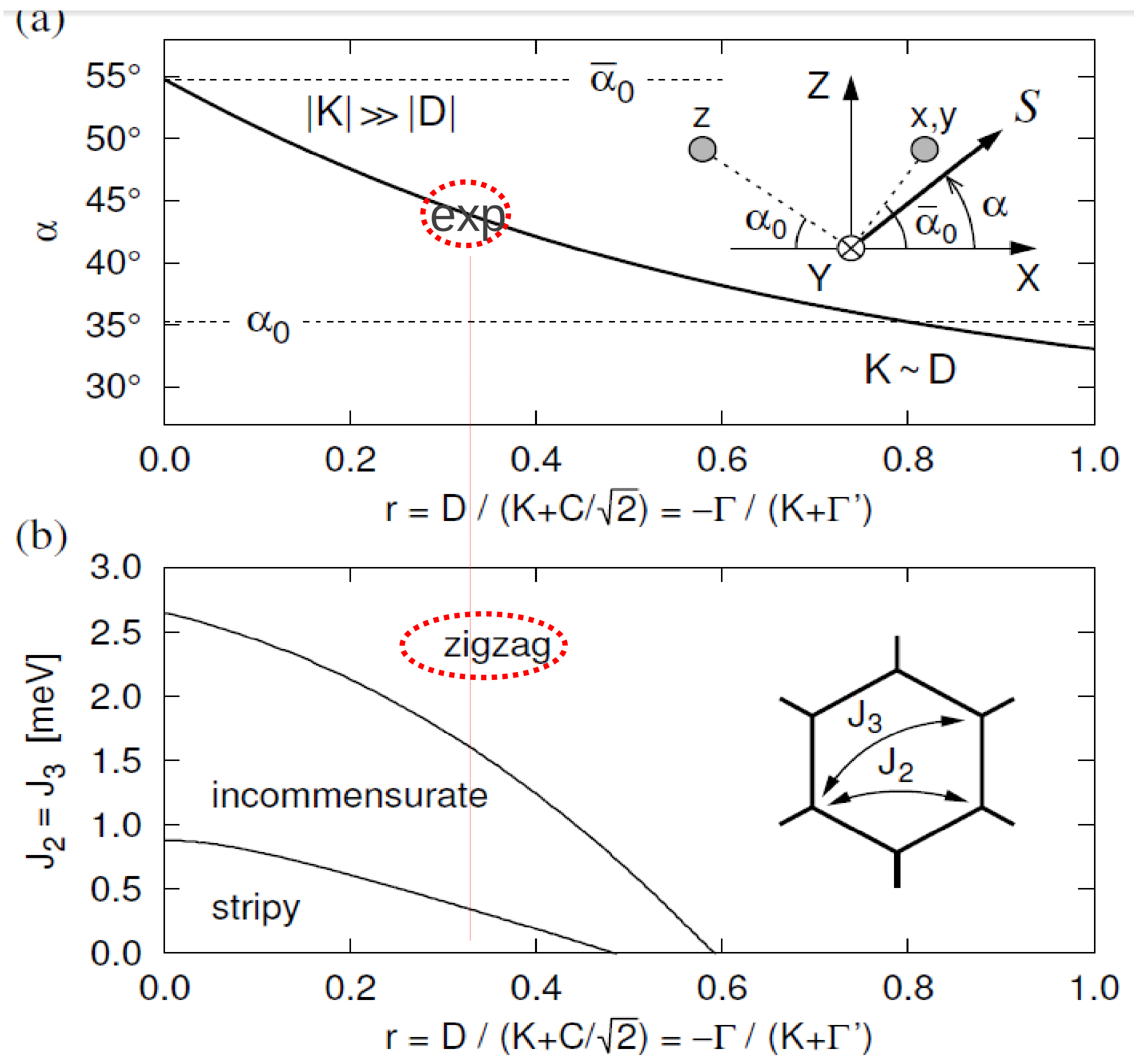
Chaloupka & GKh (2015)

Spin direction as a function of the departure from KH model

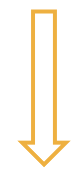
D and C make the right angle.
However, zigzag gets unstable. *We need longer range interactions.*

Coldea et al, 2012

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



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 $J_{2,3}$

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insufficient

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

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

yes, indeed

$(Na,Li)_2IrO_3$

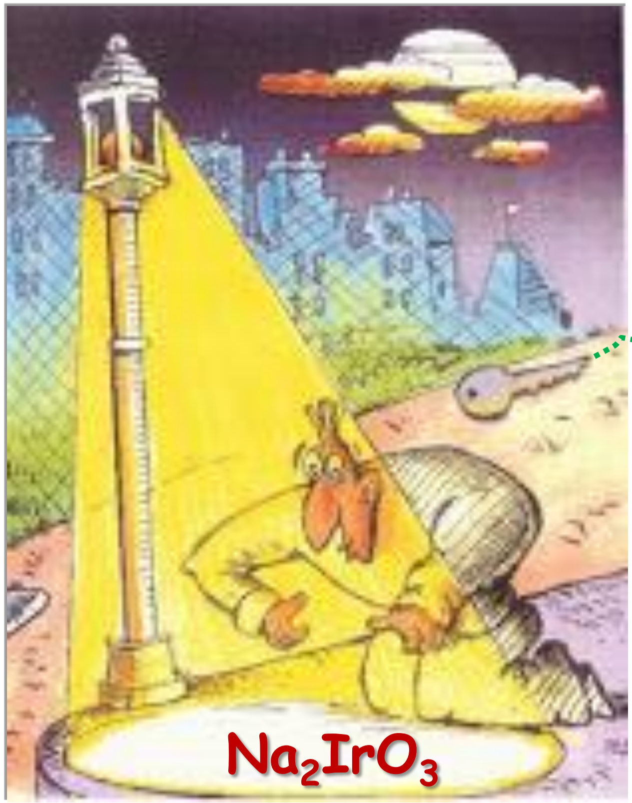
Data collected so far suggests that

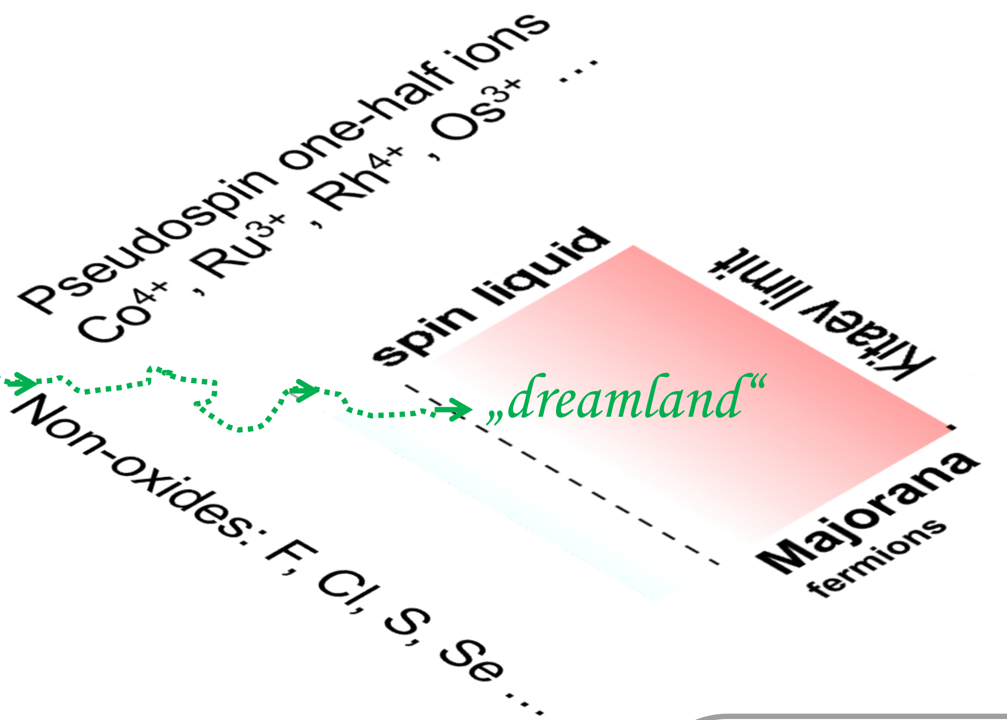
- Kitaev term seems to be dominant
- Other terms are substantial, yet to be sorted out

measure and fit,
measure and fit…

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

The streetlight effect





-Did you lose your Majoranas here?

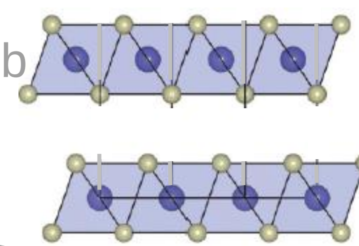
-No, but the light is much better over here !

New candidate:

$RuCl_3$

Plumb et al, 2014

honeycomb

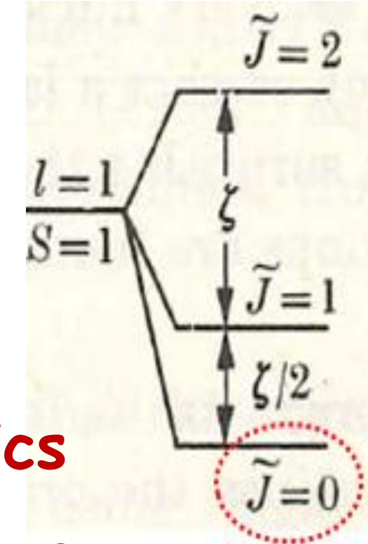


…farther away from the streetlight

J=1/2

d^5 Co, Rh, Ir

J=0 physics



d^4 Ru, Os,..

L ⟸●⟹ S

„nonmagnetic" Mott insulators

GKh (2013)
Meetei, Cole, Randeria, Trivedi (2015)

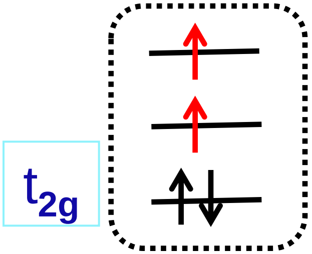
d^4 Mott insulators

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

4d, 5d electrons

1. Low-spin $S=1$

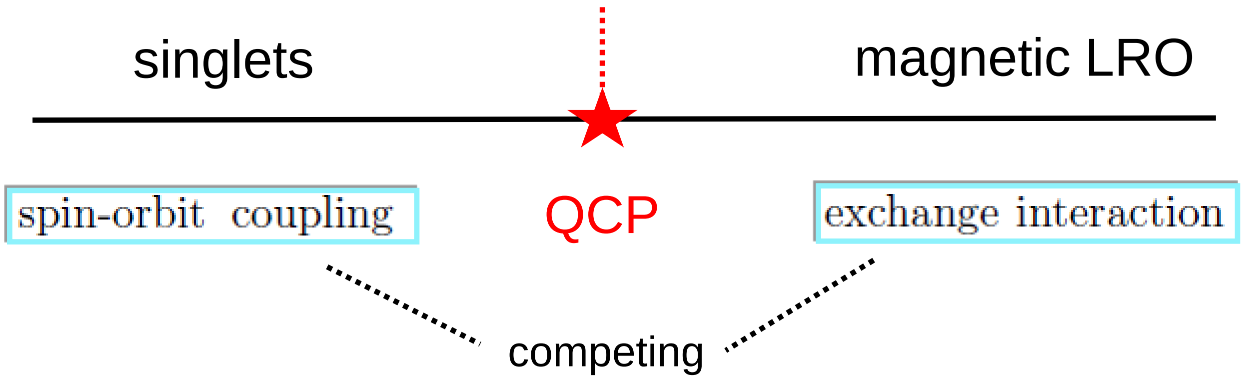
2. Unquenched $L=1$

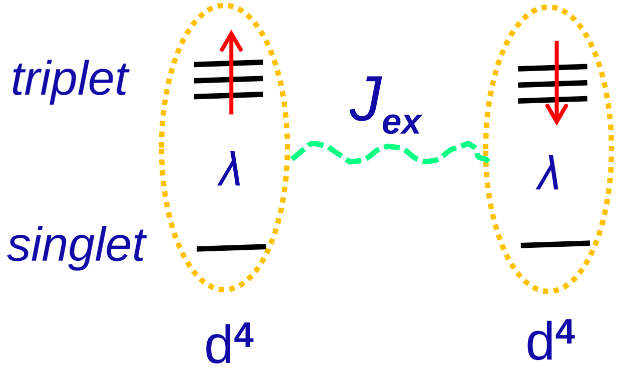


$\lambda(LS)$ ⇨ $J=S+L=0$

...no spin left to play with ...

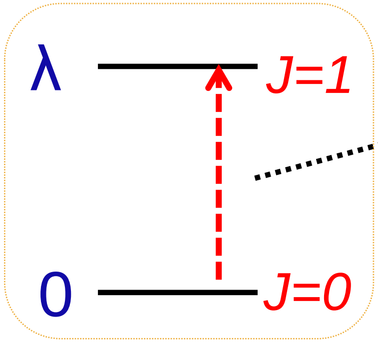
J=0 physics: *spin-orbit driven magnetic QCP*





d^4 ion: Van-Vleck magnetism

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



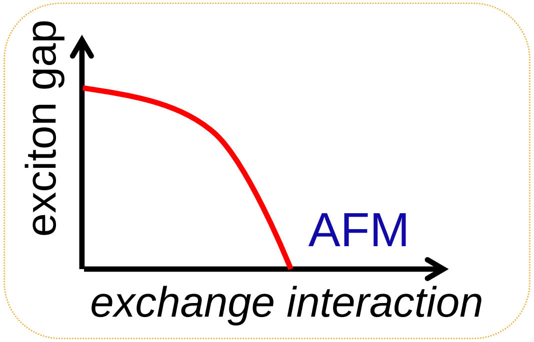
(ii) J=0 *to* J=1 *transition: off-diagonal magnetic moment* M=2S-L

(„spin-orbit exciton“)

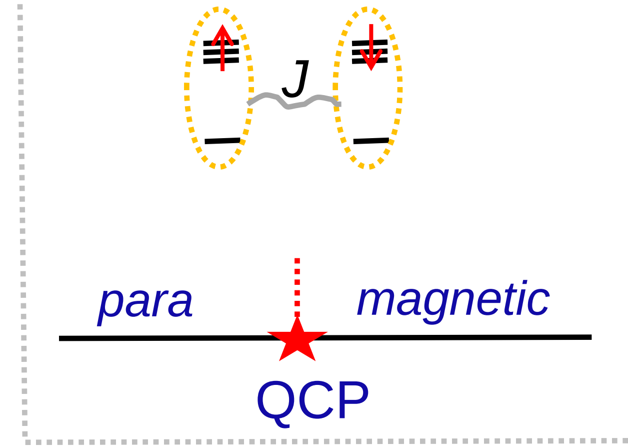
(iii) *Condensation of spin-orbit exciton*

⇩

„excitonic“ magnetism

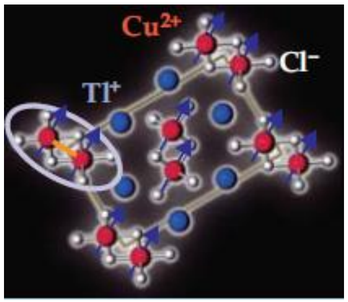


Singlet-triplet examples

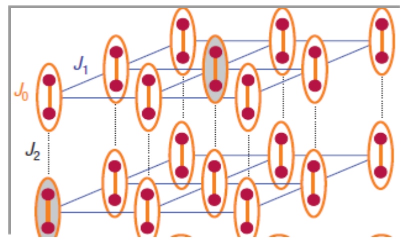


(A) Weakly coupled dimers

$TlCuCl_3$



Bilayer AF



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

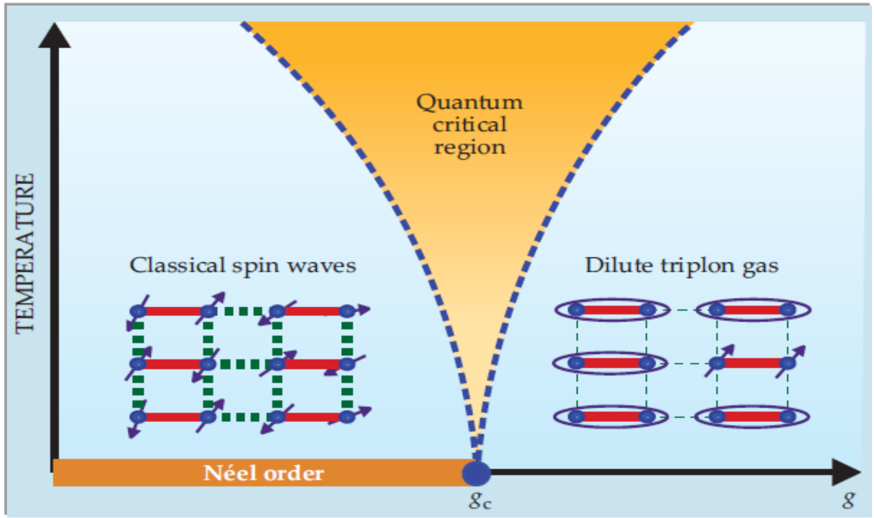
(B) 4f Pr compounds *(broad literature since 1970's)*

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

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

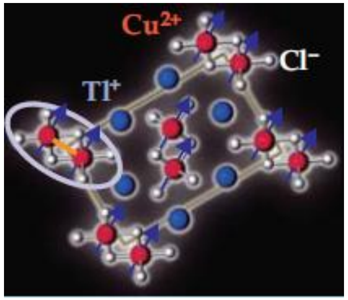
QUANTUM CRITICALITY

Sachdev, Keimer, Phys.Today, 2011



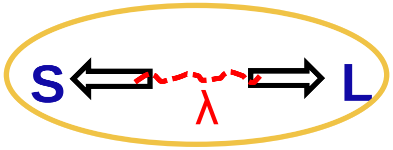
birthplace for „unconventional“ physics

Inter-ionic dimers



-small energies
-special geometry

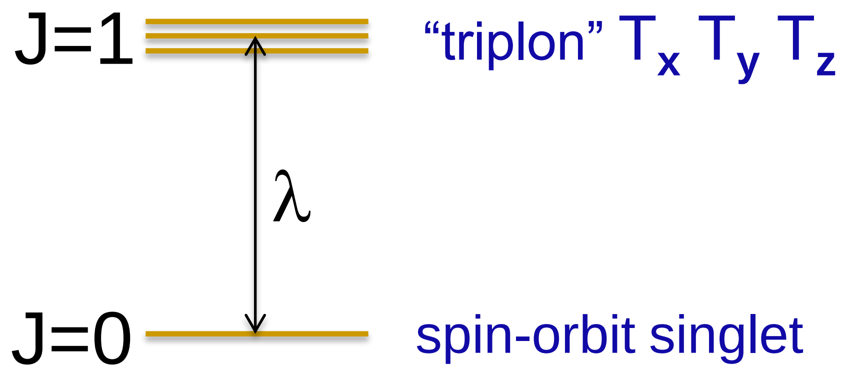
d^4 : *Intra-ionic „dimer“ made of S and L*



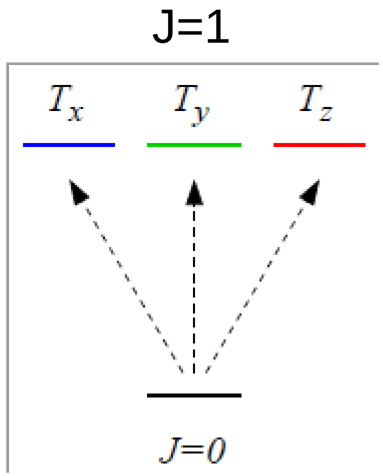
1) energetic *(~100 meV)*
2) generic *(any lattice)*

GKh (2013)

d^4 Mott insulator: single-ion states



express S and L via spin-one T-bosons



$$\boldsymbol{S} = -i\sqrt{\tfrac{2}{3}}(\boldsymbol{T} - \boldsymbol{T}^{\dagger}) + \tfrac{1}{2}\boldsymbol{J}$$

$$\boldsymbol{L} = i\sqrt{\tfrac{2}{3}}(\boldsymbol{T} - \boldsymbol{T}^{\dagger}) + \tfrac{1}{2}\boldsymbol{J}$$

magnetic moment M=2S-L:

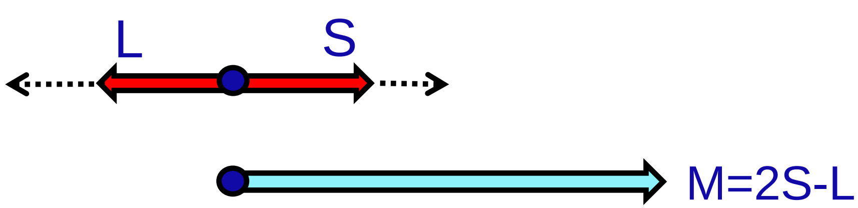
$$\boldsymbol{M} = -i\sqrt{6}\;(\boldsymbol{T} - \boldsymbol{T}^{\dagger}) + g_J \boldsymbol{J}$$

condensed

uncondensed

Bose condensation of T bosons:
finite L and S, finite M

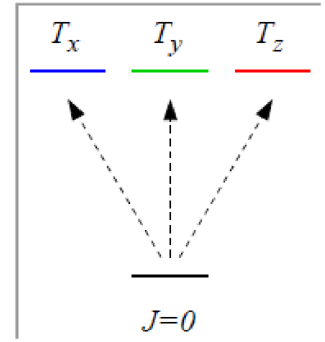
However, J=S+L=0 !



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

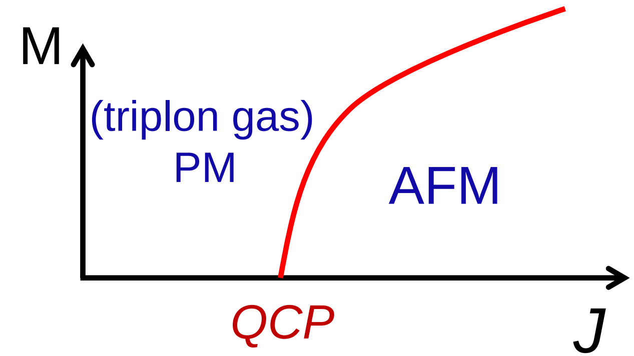
GKh (2013)

(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 (λ) — exchange (J) — S=1 boson (T)



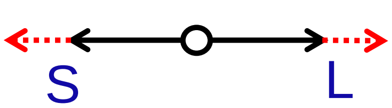
LRO moment: $M_z = 2\sqrt{6\rho(1-\rho)}$

$\rho = \frac{1}{2}(1 - \tau^{-1})$ — cond.density

$\tau = \kappa/\kappa_c > 1$ — distance from QCP

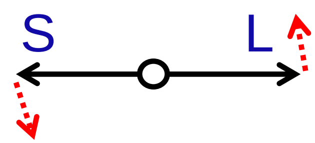
Excitations:

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



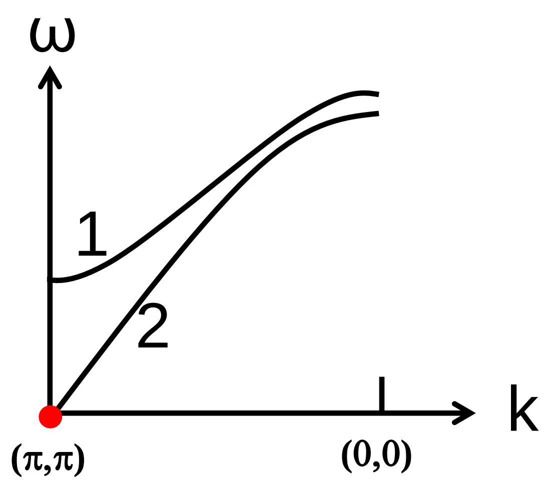
„Higgs“

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



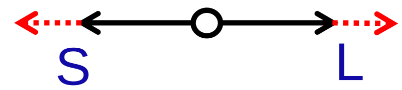
Goldstone

magnons in excitonic AF



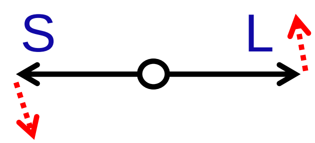
Excitations:

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



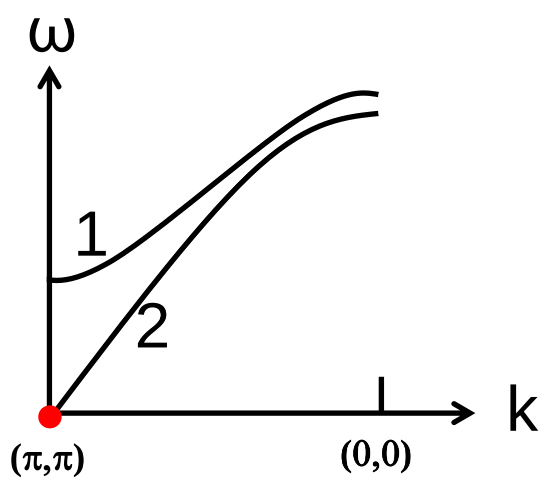
„Higgs“

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

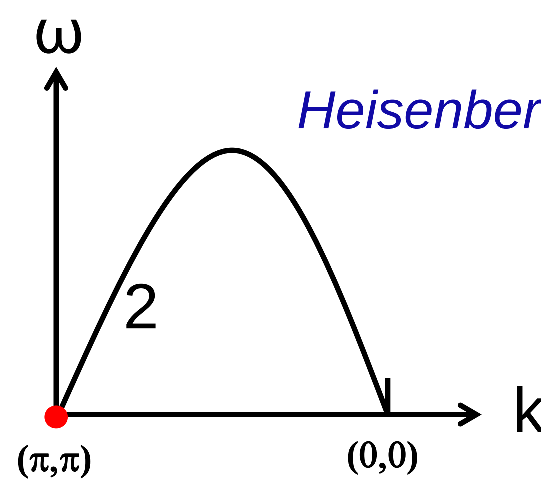


Goldstone

excitonic AF



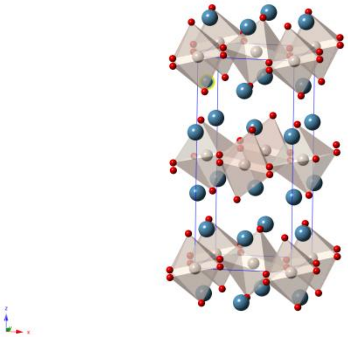
Heisenberg AF



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

Candidate material:

Ca_2RuO_4



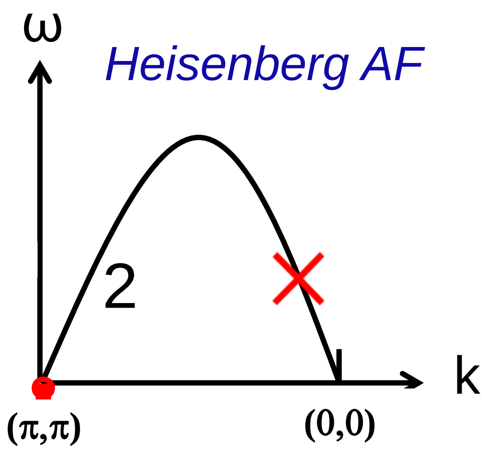
- Metal-to-insulator transition at 360 K
- Phase transition of unknown origin at 260 K *(Keimer et al. 2005)*
- Canted AF below T_N=110 K

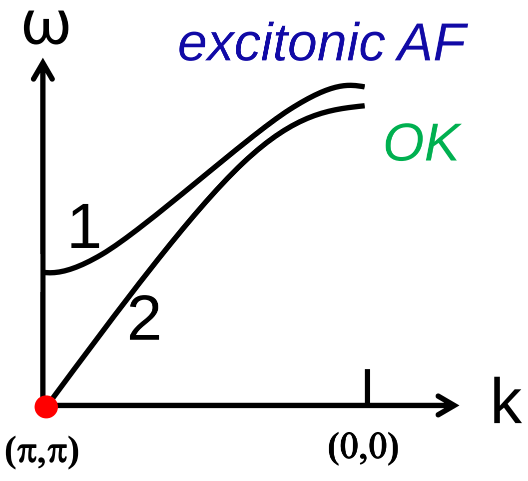
Ca_2RuO_4 single crystal, INS *(B.J. Kim et al, 2015)*

data

- ✓ far from Heisenberg
- ✓ strong SOC

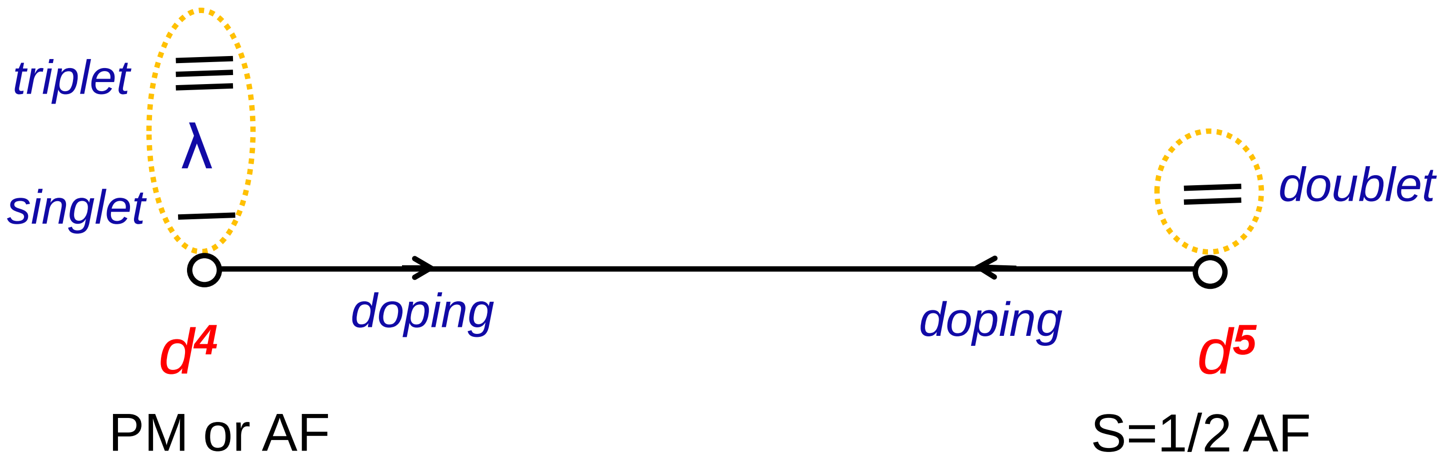
(to be published)



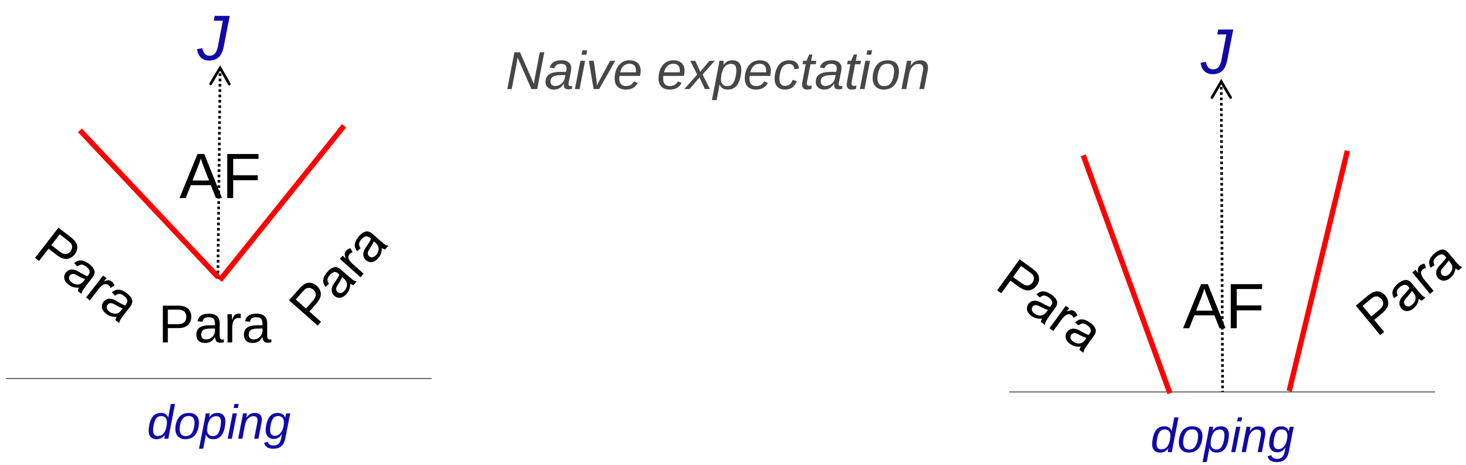


Doping of „nonmagnetic“ d⁴ Mott insulators

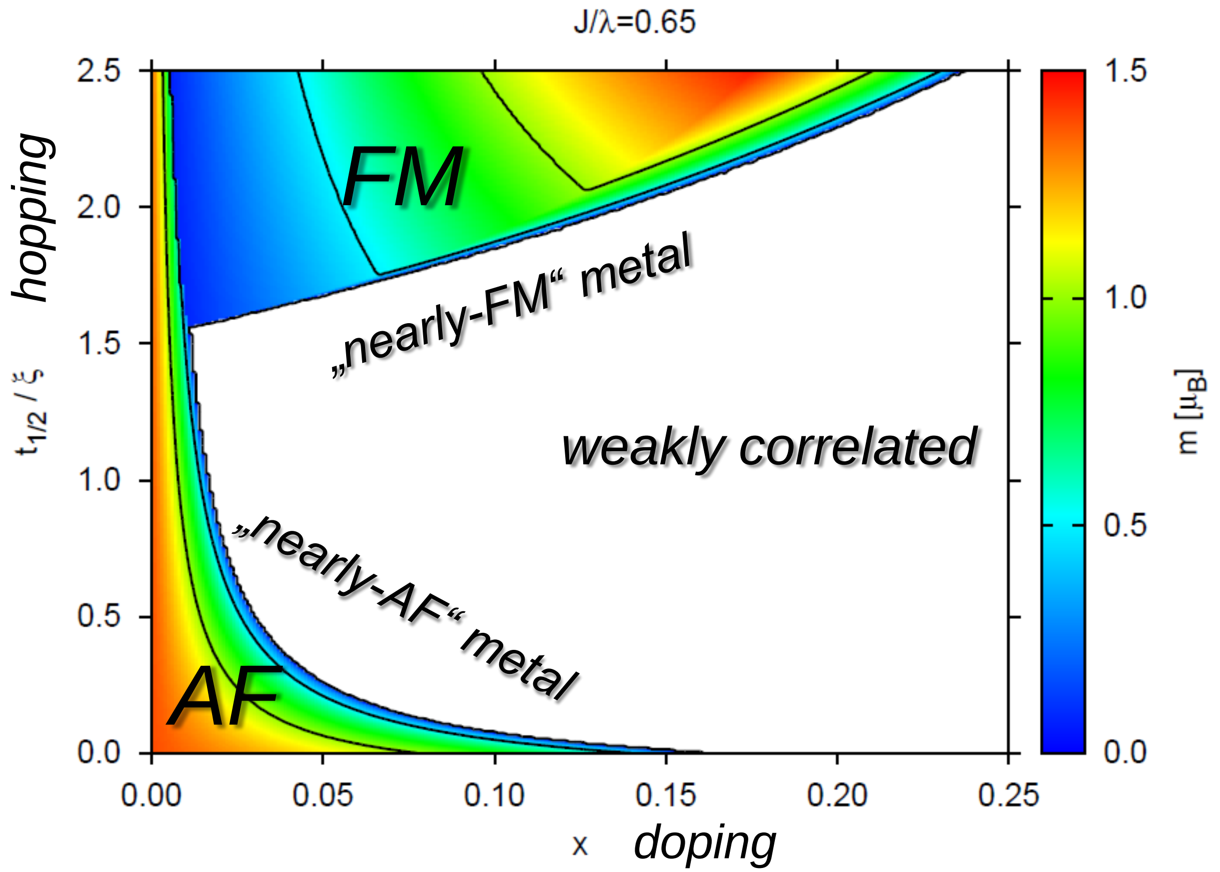
Chaloupka & GKh (unpublished)



Naive expectation



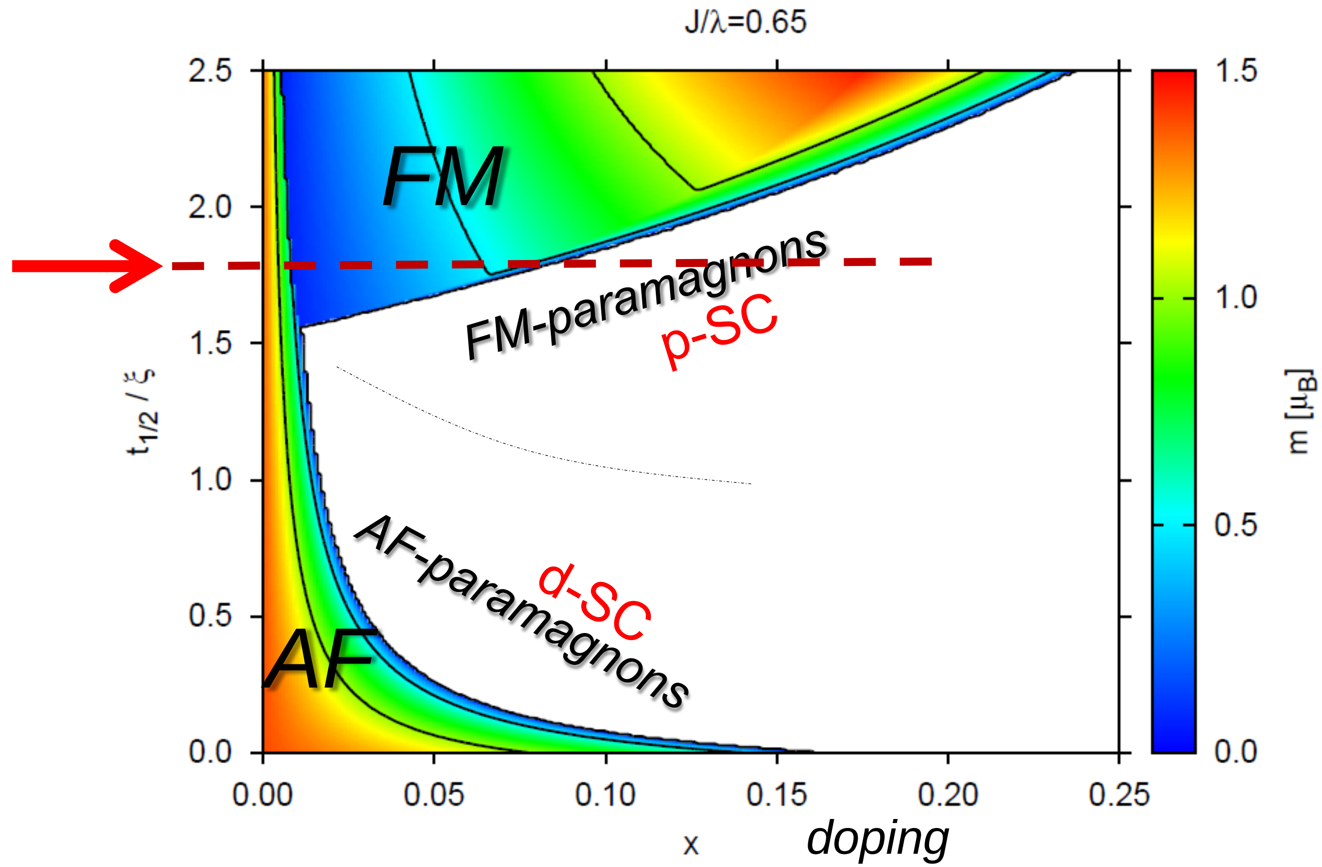
Phase diagram: doping x vs hopping t



AF --- nearly AF metal --- nearly FM metal --- FM

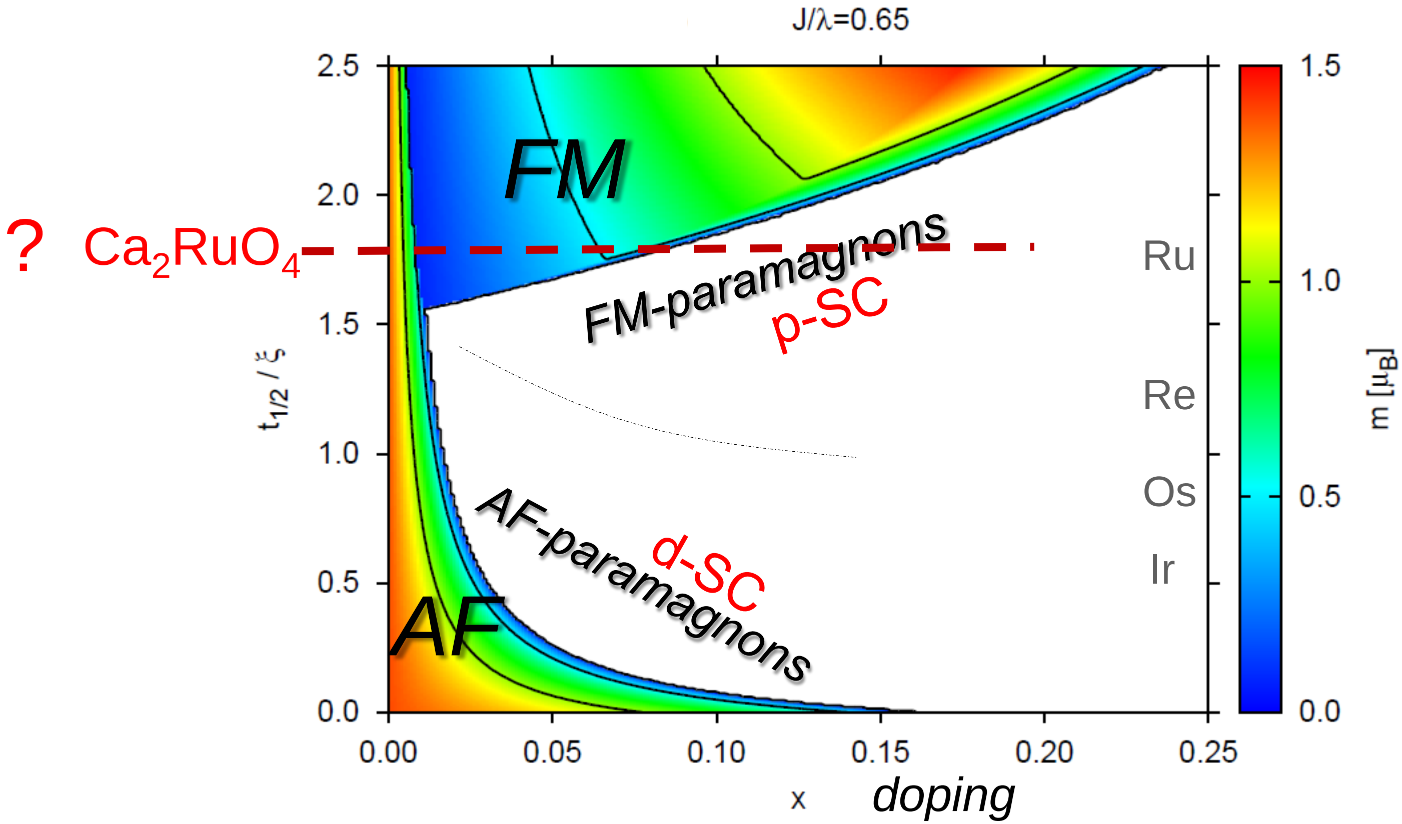
…all emerging out of the „nonmagnetic J=0“ state

Doping of „nonmagnetic“ d^4 Mott insulators



„Paramagnon-glue“: *triplet or singlet SC-pairing*

Doping of „nonmagnetic“ d^4 Mott insulators



„Sample“

t_{2g}^4 ions: Re^{3+}, Ru^{4+}, Os^{4+}, Ir^{5+}

Mott insulator to start with, better 2D

Spin-orbit comparable to hopping t

Singlet-triplet model $H_{eff}(90°)$

GKh (2013)

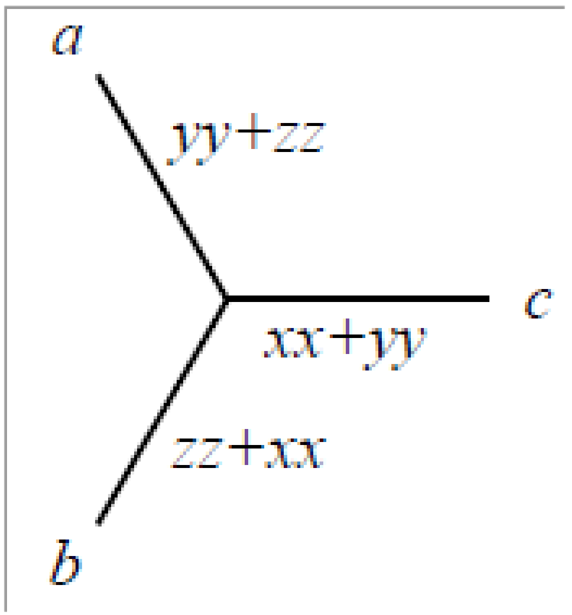
(triangular, honeycomb, kagome…)

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

hopping   pair-generation

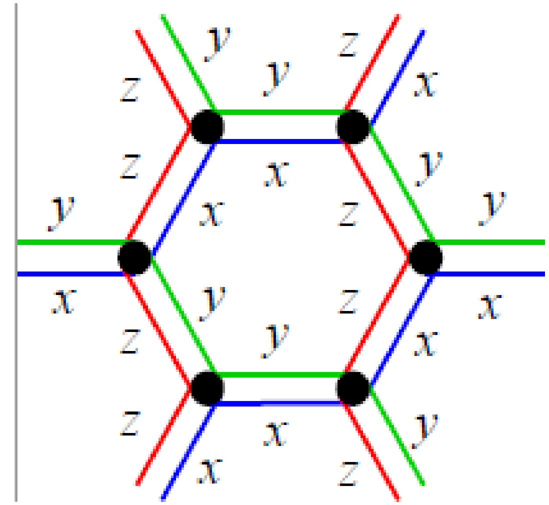
x and y type bosons only involved

Bond-dependent „xy-model“:



Honeycomb lattice: FLAT BANDS

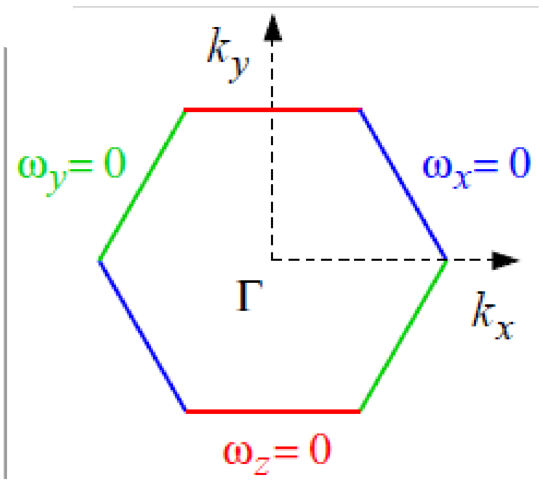
- *Each flavor T_x, T_y, T_z has its own zigzag to move along*



- *1D dispersion:*

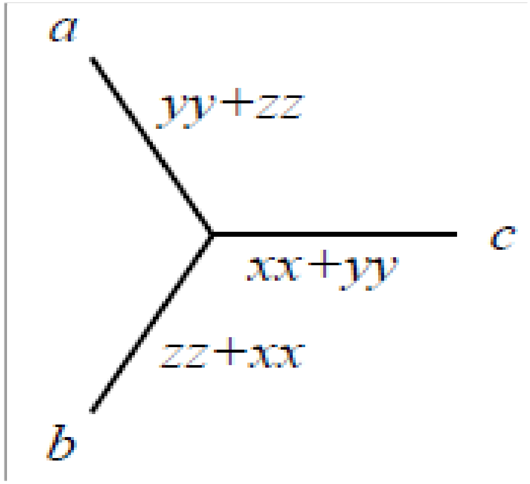
$$\omega_z(\boldsymbol{k}) \equiv \omega_z(k_y) \simeq \lambda\sqrt{1+(\kappa/\kappa_c)c_y}$$

- J=J$_{crit}$: *zero-energy lines*

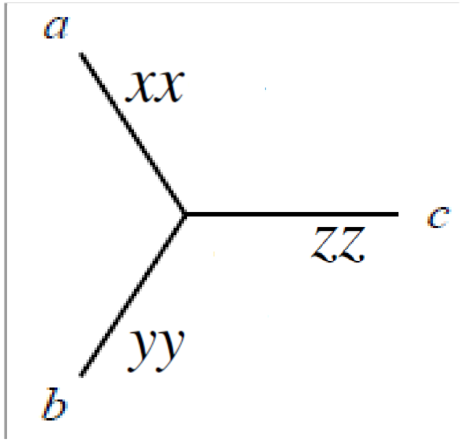


Boson exchange, two channels: t (via oxygen) vs t' (direct overlap)

Via oxygen t:
-bond-dependent „xy"



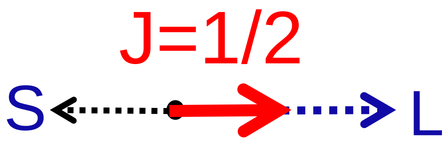
Direct overlap t':
-bond-dependent „Ising"

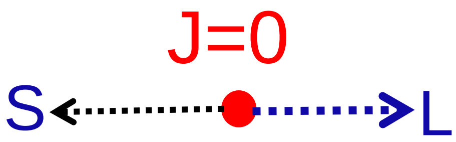


Unusual S=1 boson models

VBS?
nematic?
superfluid?

Summary





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

„nonmagnetic" J=0 Mott insulators

unconventional magnetism

…unconventional SC?