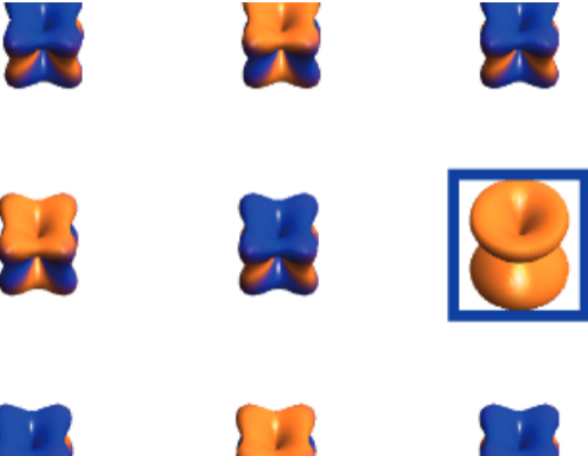




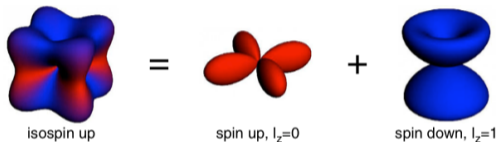
Universität Stuttgart



Maria  
Daghofer

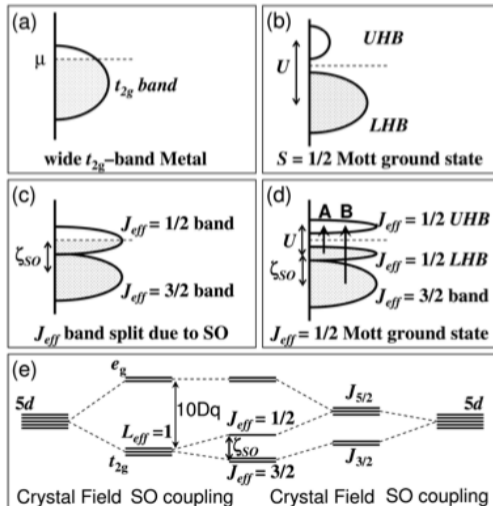
# Excitonic magnetism in $t_{2g}^4$ systems

# Spin-orbit coupling + correlations in iridates: 1 hole



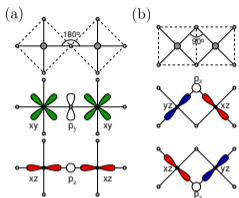
G. Jackeli and G. Khaliullin, PRL **102**, 017205 (2009)

- Three  $t_{2g}$  orbitals, two spins
- Spin-orbit coupling:  $j = \frac{1}{2}$  and  $j = \frac{3}{2}$  quartet
- Doublet has one hole

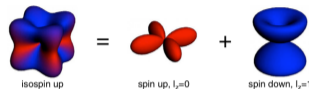


B. Kim *et al.*, PRL **101**, 076402 (2008)

# Spin-orbital Kitaev model

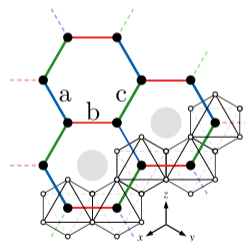


- $90^\circ$  bond angle: Anisotropic couplings crucial
- honeycomb  $\Rightarrow$  spin liquid
- variations thereon ...
- such bonds in  $A_2\text{IrO}_3$ ,  $\alpha\text{-RuCl}_3$ ,  $\text{H}_3\text{LiIr}_2\text{O}_6$



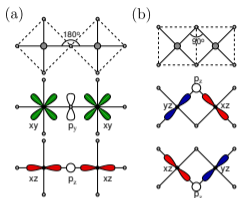
$$\mathcal{H} = J_K \sum_{\gamma} \sum_{\langle i,j \rangle \parallel \gamma} S_i^\gamma S_j^\gamma + J_H \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j + \Gamma \sum_{\gamma} \sum_{\langle i,j \rangle \parallel \gamma} (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \dots$$

G. Jackeli and G. Khaliullin, PRL **102**, 017205 (2009); J. Chaloupka, G. Jackeli, and G. Khaliullin, PRL **105**, 027204 (2010)

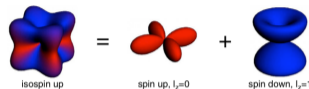


J.G. Rau *et al.*, PRL **112**, 077204 (2014)

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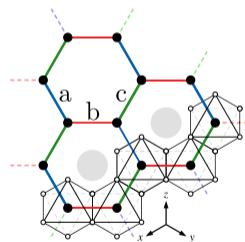


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G. Jackeli and G. Khaliullin, PRL **102**, 017205 (2009); J. Chaloupka, G. Jackeli, and G. Khaliullin, PRL **105**, 027204 (2010)



J.G. Rau *et al.*, PRL **112**, 077204 (2014)

$j = \frac{1}{2}$  on many 3D and 2D lattices: well understood and robust for square-lattice  $\text{Sr}_2\text{IrO}_4$

# Two holes with SOC

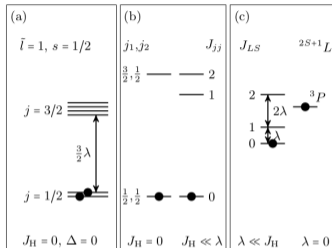
$\lambda \gg J_H$ :  $j$ - $j$  coupling

- two holes in  $j = \frac{1}{2}$ ,  
 $j = \frac{3}{2}$  filled
- band insulator
- $J_H$  affects  $j = \frac{3}{2}$

# Two holes with SOC: $jj$ vs. $LS$

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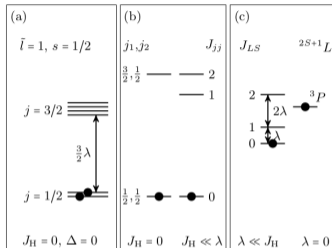
$J_H \gg \lambda$ :  $L$ - $S$  coupling

- two holes have  $S_{\text{tot}} = 1$  and  $L_{\text{tot}} = 1$
- $\lambda$  acts on degenerate  ${}^3P$
- $\lambda$  prefers  $J = 0$

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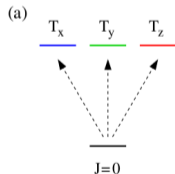
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$J_{LS} = 0$  scenario

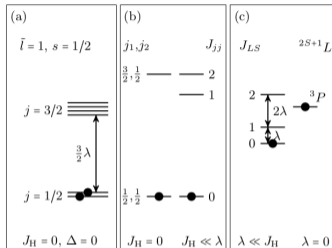
- ionic ground state  $J_{LS} = 0$
- superexchange can
  - induce triplet on both ions
  - let triplet and singlet exchange places
  - triplet  $\hat{=}$  three flavors of hard-core bosons
- a bit like coupled singlet dimers

G. Khaliullin, PRL  
111, 197201 (2013)

# Two holes with SOC: $jj$ vs. $LS$

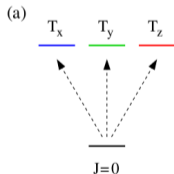
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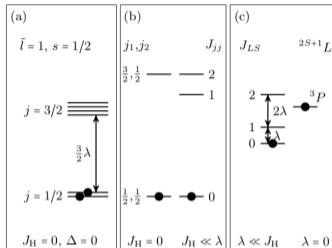
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111, 197201 (2013)



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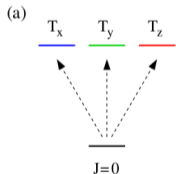
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Why would we want this?

Where might this be relevant?

G. Khaliullin, PRL  
111, 197201 (2013)

# Boson Kitaev-Heisenberg model

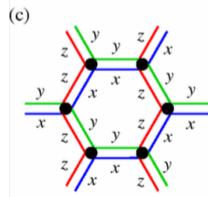
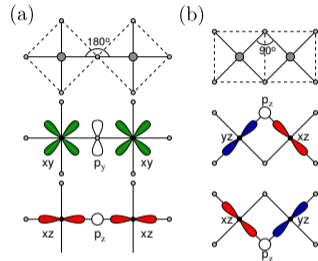
Two-triplon terms:

$$c_J, c_K, c_\Gamma \approx 1$$

$$\begin{aligned}
 H = & \lambda \sum_{i,\alpha} n_{i,\alpha} + J \sum_{\langle i,j \rangle} \left( \vec{T}_i^\dagger \vec{T}_j - c_J \vec{T}_i^\dagger \vec{T}_j^\dagger + \text{H. c.} \right) \\
 & + K \sum_{\alpha} \sum_{\langle i,j \rangle \parallel \alpha} \left( T_{i,\alpha}^\dagger T_{j,\alpha} - c_K T_{i,\alpha}^\dagger T_{j,\alpha}^\dagger + \text{H. c.} \right) \\
 & + \Gamma \sum_{\substack{\alpha \neq \beta \neq \gamma \\ \alpha \neq \gamma}} \sum_{\langle i,j \rangle \parallel \alpha} \left( T_{i,\beta}^\dagger T_{j,\gamma} - c_\Gamma T_{i,\beta}^\dagger T_{j,\gamma}^\dagger + \text{H. c.} \right)
 \end{aligned}$$

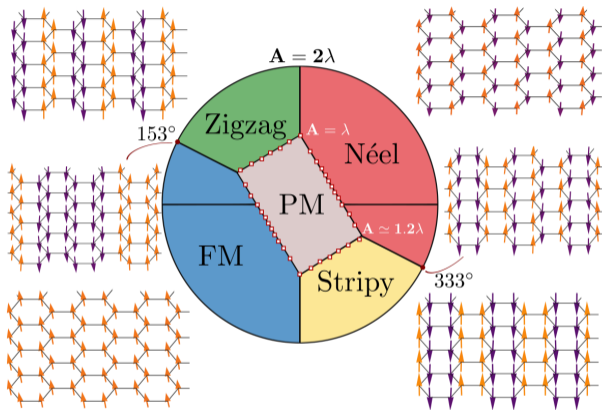
- Three- and four-triplon terms present
- Perturbation theory for  $J_{\text{Hund}} = 0$ :
  - only  $t$  via oxygen:  $0 < J = -K, \Gamma = 0$
  - only direct  $t'$ :  $0 < J \ll K, \Gamma = 0$
  - $\Gamma \propto tt'$
- $J_{\text{Hund}}$  promotes FM coupling  $J < 0$

C. Svoboda, M. Randeria, N. Trivedi, PRB **95**, 014409 (2017)



G. Khaliullin, PRL **111**, 197201 (2013)

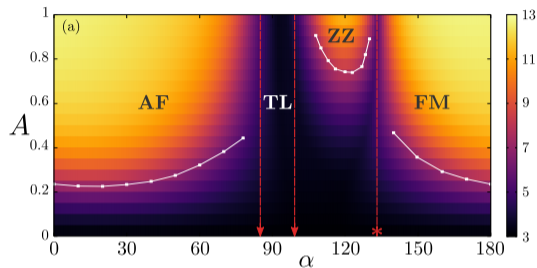
# Classical Phase diagram on honeycomb



- center: large SOC suppresses triplons
- rim: strong superexchange overcomes SOC
- $J = A \cos \alpha$
- $K = 2A \sin \alpha$
- $\Gamma = 0$

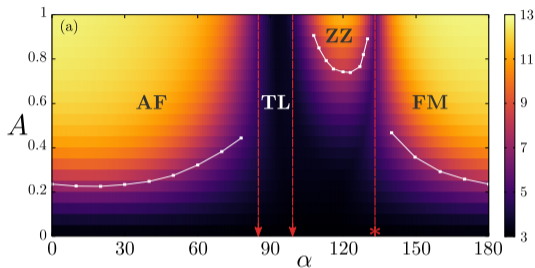
- Ordered phases are those of classical Kitaev-Heisenberg(- $\Gamma$ ) model.
- Non-magnetic region for dominant  $\lambda$
- Transition driven by  $T^\dagger T^\dagger$  terms

# Quantum Phase diagram from ED



- $J = A \cos \alpha$ ,  $K = 2A \sin \alpha$
- Large  $A \gg \lambda$ : more classical than  $j = \frac{1}{2}$  model  
D. Gotfryd *et al.*, PRB **95**, 024426 (2017)
- Keeps perfect symmetry  $\alpha \rightarrow \alpha + 180$
- 1<sup>st</sup>-order phase transitions at  $K = -J$

# Quantum Phase diagram from ED

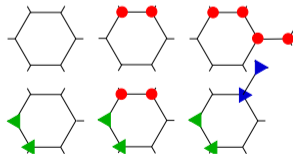


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Regime without magnetic order at  $J \approx 0$ :

- magnetic correlations strictly short range at Kitaev point
- no other order found
- gapped: lower energy than dimer coverings  $E < -K/2$
- non degenerate: order by disorder

P. S. Anisimov, F. Aust, G. Khaliullin, M. Daghofer, PRL **122**, 177201 (2019); J. Chaloupka and G. Khaliullin, arXiv:1910.00074



## Excitations of the $J = 0$ regime

$$\begin{aligned} H = & \lambda \sum_{i,\alpha} n_{i,\alpha} + J \sum_{\langle i,j \rangle} \left( \vec{T}_i^\dagger \vec{T}_j - c_J \vec{T}_i^\dagger \vec{T}_j^\dagger + \text{H. c.} \right) \\ & + K \sum_{\alpha} \sum_{\langle i,j \rangle \parallel \alpha} \left( T_{i,\alpha}^\dagger T_{j,\alpha} - c_K T_{i,\alpha}^\dagger T_{j,\alpha}^\dagger + \text{H. c.} \right) \\ & + \Gamma \sum_{\substack{\alpha \neq \beta \neq \gamma \\ \alpha \neq \gamma}} \sum_{\langle i,j \rangle \parallel \alpha} \left( T_{i,\beta}^\dagger T_{j,\gamma} - c_\Gamma T_{i,\beta}^\dagger T_{j,\gamma}^\dagger + \text{H. c.} \right), \end{aligned}$$

stick to  $\lambda \gg J, K, \Gamma \Rightarrow$  no magnetic order

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 \end{aligned}$$

stick to  $\lambda \gg J, K, \Gamma \Rightarrow$  no magnetic order

band structure for triplons:

- once a triplon has been created, it can hop
- bands are here topologically trivial
- non-trivial intersite-dimer triplons known

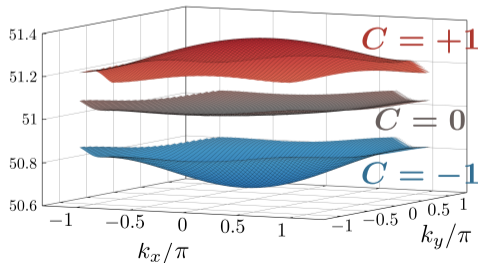
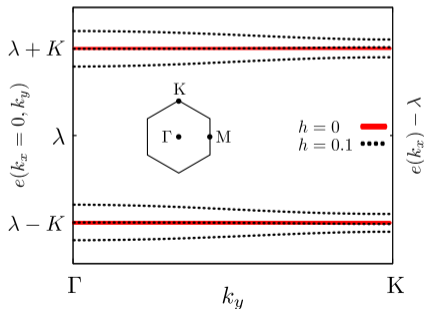
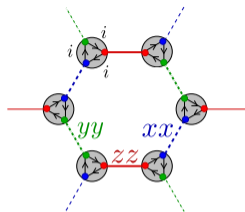
J. Romhányi *et al.*, Nat. Commun. **6**, 6805 (2015); P. A. McClarty *et al.*, Nat. Phys. **13**, 736 (2017);

D.G. Joshi and A.P.Schnyder, PRB **96**, 220405 (2017); **100**, 020407 (2019)

# Triplons non-trivial in magnetic field

$$M_{j,x} = -i\sqrt{6} (T_{j,x} - T_{j,x}^\dagger) + ig_J (T_{j,y}^\dagger T_{j,z} - T_{j,z}^\dagger T_{j,y})$$

$$\approx ig_J (T_{j,y}^\dagger T_{j,z} - T_{j,z}^\dagger T_{j,y})$$

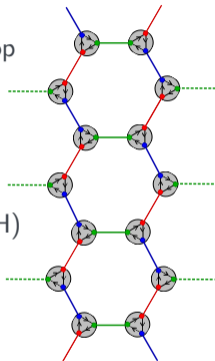




# Two kinds of Edge States

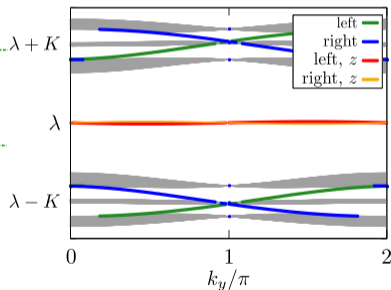
## states at $\lambda$

- on edge sites, green flavor can't hop
- topological of a different sort  
G. van Miert *et al.*, 2D Mat. **4**, 015023 (2017); D. G. Joshi and A. P. Schnyder, PRB **100**, 020407 (2019)
- related to Su-Shrieffer-Heeger (SSH) chain;  $\text{Ba}_2\text{CuSi}_2\text{O}_6\text{Cl}_2$ ?  
K. Nawa *et al.*, Nat. Comm., **10**, 2096 (2019)
- does not cross gap
- shifted with onsite energy

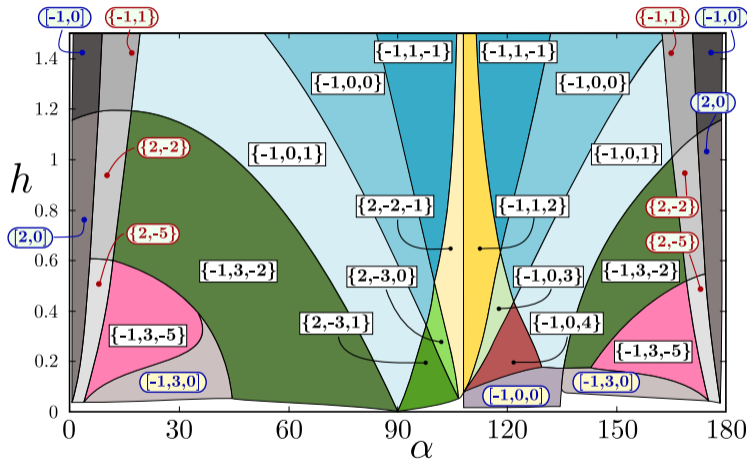


## states at $\lambda \pm K$

- due to Chern number of bands
- crosses nontrivial band gap
- dissipationless transport?



# Nontrivial triplon topology very robust



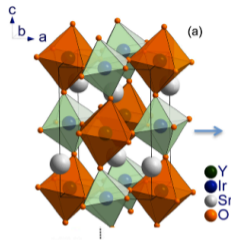
- $J = \cos \alpha$
- $K = \sin \alpha$
- $\vec{h} = (1, 1, 1) \cdot h$

P. S. Anisimov, F. Aust, G. Khaliullin, M. Daghofer, PRL 122, 177201 (2019)

$$\{-1,3,-2\} \rightarrow \{-1,3,-2,2,-3,1\}, \quad \{2,-5\} \rightarrow C_{1+2} = 2, \quad C_3 = -5, \quad C_4 = 5, \quad C_{5+6} = -2$$

$$[2,0] \rightarrow C_{1+2} = 2, \quad C_{3+4} = 0, \quad [1,-1,0] \rightarrow C_1 = 1, \quad C_2 = -1, \quad C_{3+4} = 0$$

# Where might $J = 0$ / $J = 1$ be relevant?



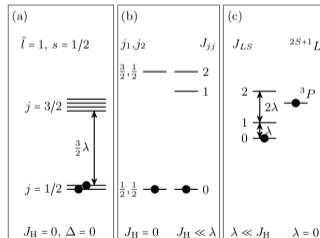
from G. Cau *et al.*, PRL **112**, 056402 (2014)

## Ir based double perovskites

- spin-orbit coupling (SOC) large for Ir
- Ir far apart
- likely nonmagnetic

K. Pajskr *et al.*, PRB **93**, 035129 (2016), S. Fuchs *et al.*, PRL **120**, 237204 (2018)

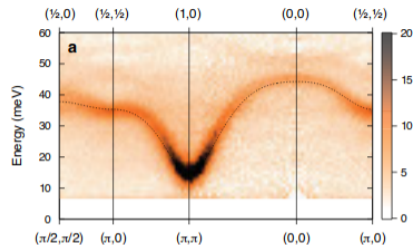
- very strong onsite singlet
- superexchange too weak to mix in triplet
- Picture applicable, but maybe a bit too robust



# Where might this be relevant?

$\text{Ca}_2\text{RuO}_4$

- AFM order
- excitations well explained with  $J = 0/J = 1$  scenario
- Esp. **maximum** at  $(0,0)$  rather than minimum



from A. Jain *et al.*, Nat. Phys. **13**, 633 (2017)

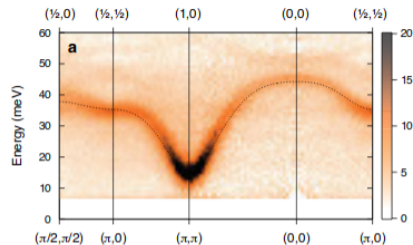
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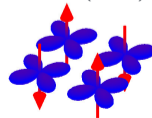
## Spin-and-Orbital vs. Spinorbital

- OTOH: spin and orbital also decent description  
T. Mizokawa *et al.*, PRL **87**, 077202 (2001); M. Cuoco *et al.*, PRB **74**, 195124 (2006)
- *ab-initio* treatment: strong orbital polarization  $\rightarrow$  spin  $S = 1$  good description  
G. Zhang and E. Pavarini PRB **95**, 075145 (2017); D. Sutter *et al.*, Nat. Comm. **8**, 15176 (2017)



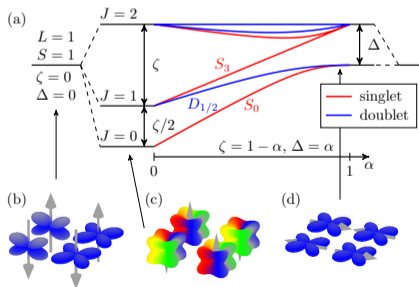
from A. Jain *et al.*, Nat. Phys. **13**, 633 (2017)

- correlations enhance SOC  $\rightarrow$  shows up in superexchange  
G. Zhang and E. Pavarini PRB **95**, 075145 (2017)



# Strongly correlated $t_{2g}$ model with SOC

- Model studies: investigation of excitonic vs. 'normal spin' magnetism
  - Variational cluster approach applied to model for electrons
  - Exact-diagonalization spectra of Kugel-Khomskii-type model for spins and orbitals
- $\text{Ca}_2\text{RuO}_4$ :
  - parameters from DFT (hopping, crystal field) and experiment (interactions, SOC)
  - one-particle spectra do not really show SOC, magnetic spectra do
  - excitonic despite orbital polarization



## $t_{2g}$ model on 2D square lattice

- Hopping (nearest neighbors or DFT derived)

$$H_{\text{kin}} = -t \sum_{\langle i,j \rangle, \sigma} c_{i,xy,\sigma}^\dagger c_{j,xy,\sigma} - t \left( \sum_{\langle i,j \rangle \parallel x, \sigma} c_{i,xz,\sigma}^\dagger c_{j,xz,\sigma} + \sum_{\langle i,j \rangle \parallel y, \sigma} c_{i,yz,\sigma}^\dagger c_{j,yz,\sigma} \right),$$

- tetragonal crystal field
- spin-orbit coupling

$$H_{\Delta} = -\Delta \sum_{i,\sigma} n_{i,xy,\sigma}$$

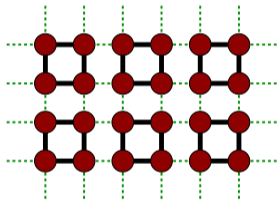
$$H_{\text{SOC}} = \zeta \sum_i \vec{l}_i \cdot \vec{s}_i = \frac{i\zeta}{2} \sum_i \sum_{\substack{\alpha,\beta,\gamma \\ \sigma,\sigma'}} \varepsilon_{\alpha\beta\gamma} \tau_{\sigma\sigma'}^\alpha c_{i,\beta,\sigma}^\dagger c_{i,\gamma,\sigma'}$$

- onsite Coulomb and Hund's coupling

$$H_{\text{int}} = U \sum_{i,\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + \frac{U'}{2} \sum_{i,\sigma} \sum_{\alpha \neq \beta} n_{i\alpha\sigma} n_{i\beta\bar{\sigma}} \\ + (U' - J_H) \sum_{i,\sigma} \sum_{\alpha > \beta} n_{i\alpha\sigma} n_{i\beta\sigma} - J_H \sum_{i,\alpha \neq \beta} (c_{i\alpha\uparrow}^\dagger c_{i\alpha\downarrow} c_{i,\beta\downarrow}^\dagger c_{i\beta\uparrow} - c_{i\alpha\uparrow}^\dagger c_{i\alpha\downarrow}^\dagger c_{i\beta\downarrow} c_{i\beta\uparrow})$$

# Variational Cluster Approximation

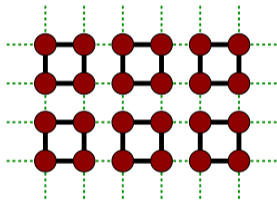
- Get free energy and one-particle Green function (GF) for small cluster  $\rightarrow$  self energy  $\Sigma$
- Plug  $\Sigma$  into GF of big system  $\rightarrow$  GF and grand potential of big system
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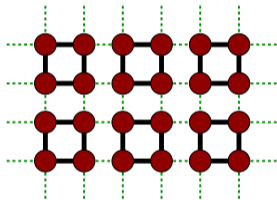


Symmetry-breaking field:  $h' \sum_i \Lambda_i e^{i\vec{Q}\vec{R}_i}$

- one-particle operator  $\Lambda_i$ , e.g.  $S_i^z$ ,  $L_i^x$ ,  $n_{i,xy}$
- $\vec{Q} = (0, 0), (\pi, \pi), (\pi, 0), (0, \pi)$  strongly restricted by cluster

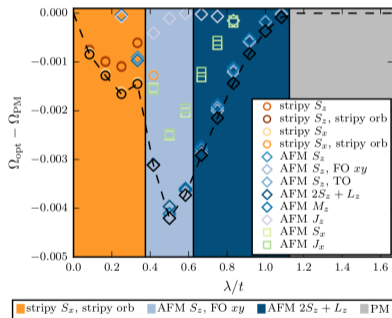
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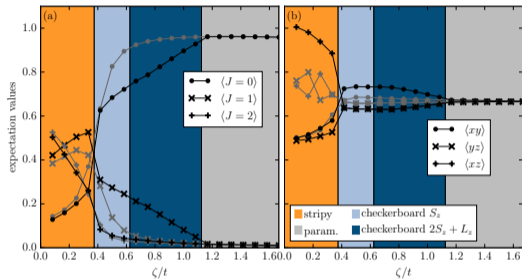
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- important:  $\vec{Q}$  and in-plane ( $x$ ) vs. out-of-plane ( $z$ )
- less important:  $\alpha$  in  $S_i^{x/z} + \alpha L_i^{x/z}$

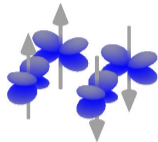
# SOC $\zeta$ reduces onsite degeneracy: $J_{LS} = 0$ / $J_{LS} = 1$ order



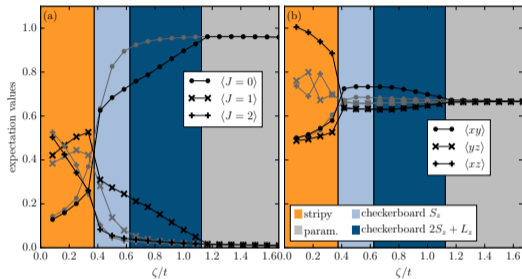
complex stripy pattern:

out of nine-fold  $L = 1$ ,  $S = 1$   
degeneracy

fits M. Cuoco *et al.*, PRB **74**, 195124  
(2006)



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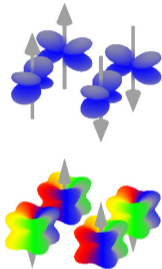


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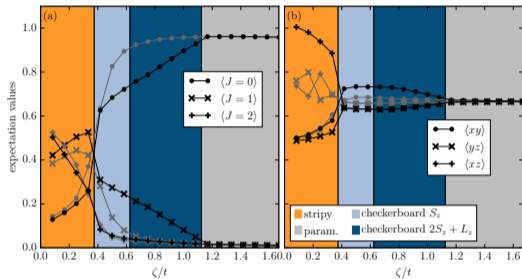
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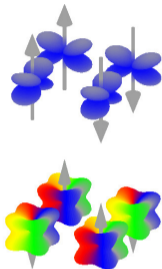


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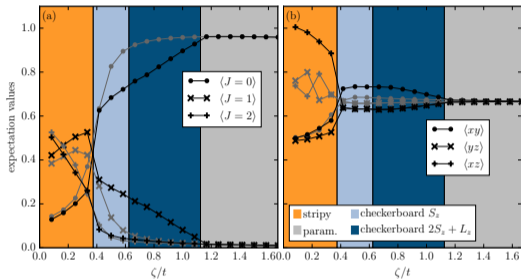
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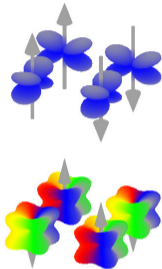
- grey symbols: no symmetry breaking  
 $J = 0$  stabilized by  $\zeta$ , soon dominates
- black symbols: optimized order  
weight shifted into  $J = 1$
- $J = 2$  unimportant

complex stripy pattern:

out of nine-fold  $L = 1$ ,  $S = 1$   
degeneracy

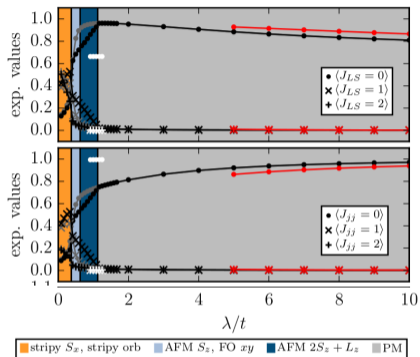
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# $L$ - $S$ to $j$ - $j$ : correlated to uncorrelated

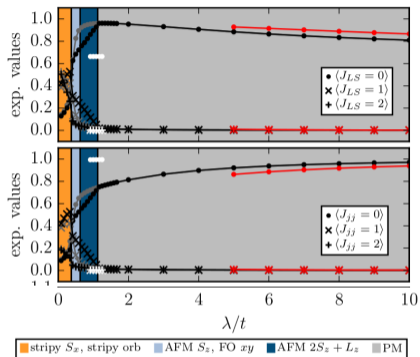


SOC  $\lambda \gg J_H$  Hund coupling:  $j$ - $j$

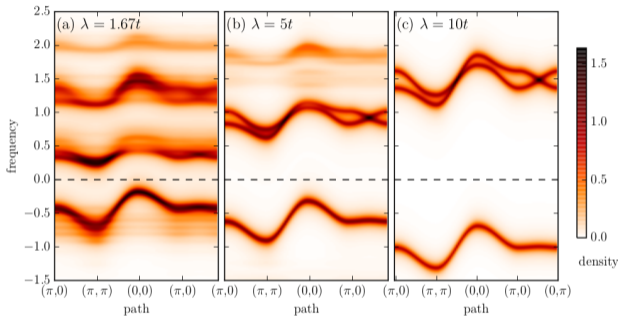
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white:  $J_H = 0$ , red:  $J_H = U/3$

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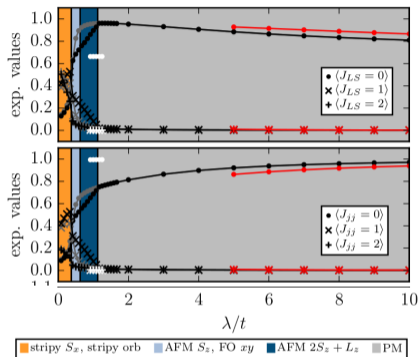
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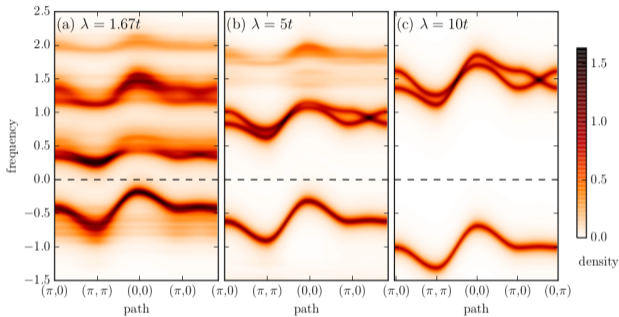
correlated bands split by  $J_H$   
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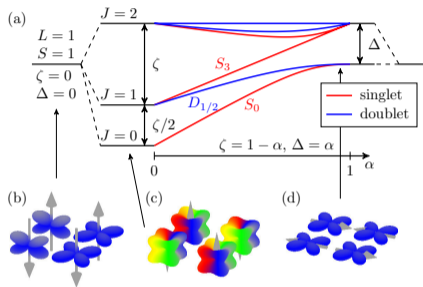
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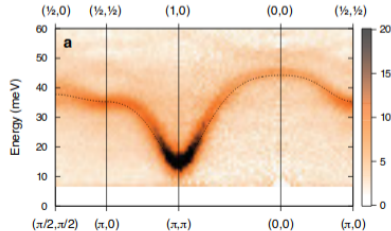
Sticking to correlated  $L$ - $S$  regime here.

# Crystal field vs. spin-orbit coupling



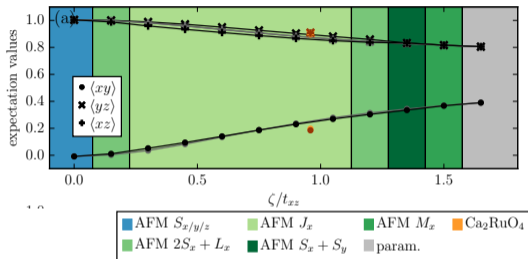
- $\Delta \approx 0$ : excitonic limit with  $J = 0/J = 1$
- $\zeta \approx 0$ : Spin-1 with doubly occ.  $xy$
- $\text{Ca}_2\text{RuO}_4$  somewhere in between
  - structural transition:  $xy$  almost doubly occupied (DFT+DMFT, ARPES)
  - Neutrons clearly show spin-orbit coupling

- Where in between is  $\text{Ca}_2\text{RuO}_4$ ?
- Orbitaly polarized  $S = 1$  with some spin-orbit corrections?
- Excitonic with some crystal-field corrections?



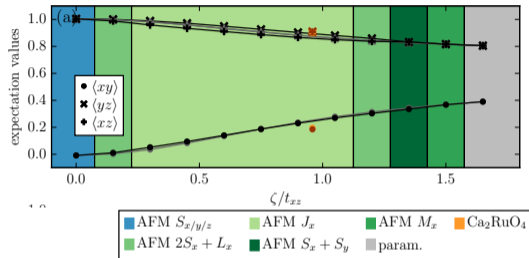
A. Jain *et al.*, Nat. Phys. **13**, 633 (2017)

# SOC in the presence of a Strong Crystal Field



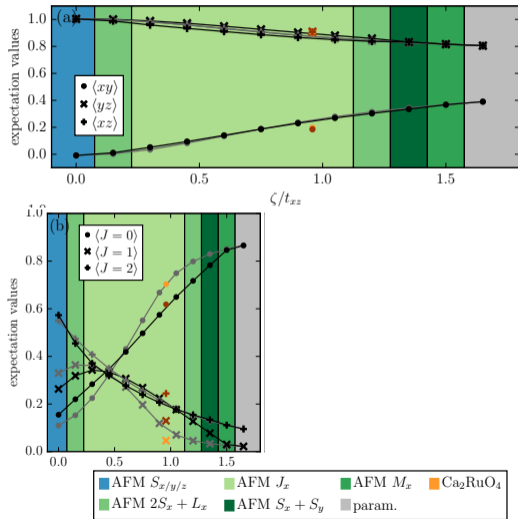
- SOC  $\zeta = 0$ : one hole on  $xz$ , one in  $yz \Rightarrow$  polarized  $S = 1$  with  $n_{xy}^h = 0$ ,  $n_{xz/yz}^h = 1$
- $\zeta \approx t$ : pretty polarized with  $n_{xy}^h \approx 0.2$ ,  $n_{xz/yz}^h \approx 0.9$

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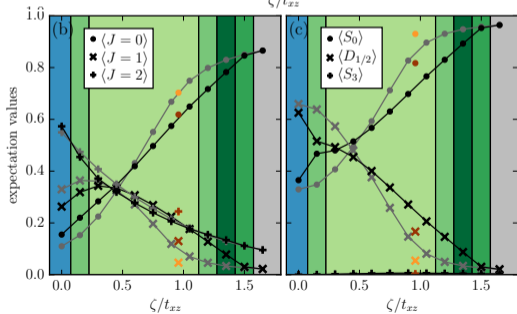
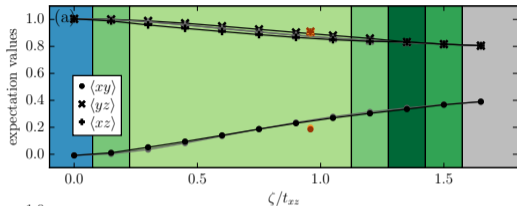
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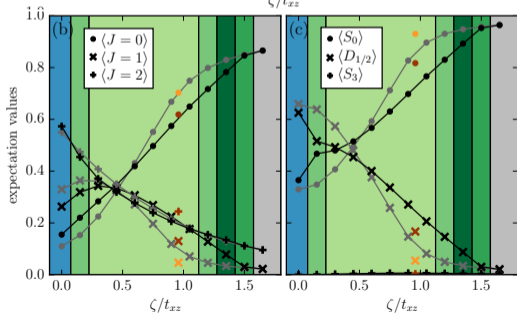
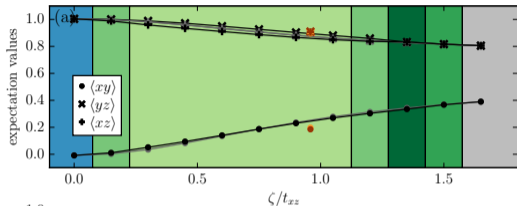
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■ AFM  $S_{x/y/z}$    
 ■ AFM  $J_x$    
 ■ AFM  $M_x$    
 ■ Ca<sub>2</sub>RuO<sub>4</sub>  
■ AFM  $S_x$    
 ■ AFM  $S_y$    
 ■ param. systems

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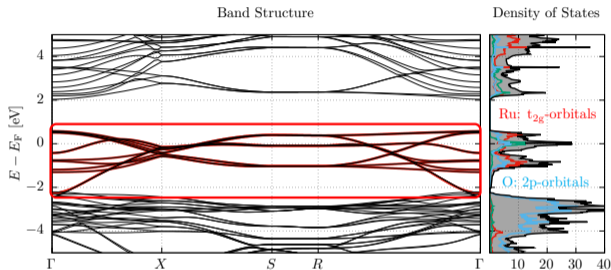


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# One-particle parameters for $\text{Ca}_2\text{RuO}_4$



- Wannier projection  $\Rightarrow$  get  $t$ 's and  $\Delta$   
suppl. mat. to J. Bertinshaw *et al.*, PRL **123**, 137204 (2019)
- get SOC  $\zeta \approx 0.13$  eV,  $U \approx 2$  eV and  $J_H \approx 0.34$  eV from H. Gretarsson *et al.*, PRB **100**, 045123 (2019)
- plug into VCA  $\Rightarrow$  red/orange symbols

$$\epsilon_{xy,xy}(\vec{k}) = -2t_{xy}^{\text{NN}}(\cos k_x + \cos k_y) - 4t_{xy}^{\text{NNN}} \cos k_x \cos k_y - \Delta,$$

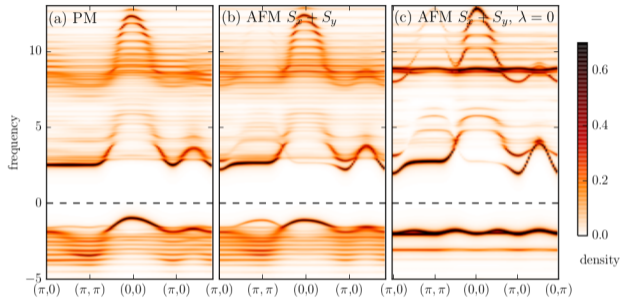
$$\epsilon_{xz,xz}(\vec{k}) = -2t_{xz}^{\text{NN}} \cos k_x, \quad \epsilon_{yz,yz}(\vec{k}) = -2t_{yz}^{\text{NN}} \cos k_y,$$

$$\text{and } \epsilon_{xz,xy}(\vec{k}) = -2t^o \cos k_x.$$

with  $t_{xy}^{\text{NN}} \approx 0.2$  eV,  $t_{xz}^{\text{NN}} = t_{yz}^{\text{NN}} \approx 0.14$  eV,  $t_{xy}^{\text{NNN}} \approx 0.09$  eV,  $t^o \approx 0.09$  eV

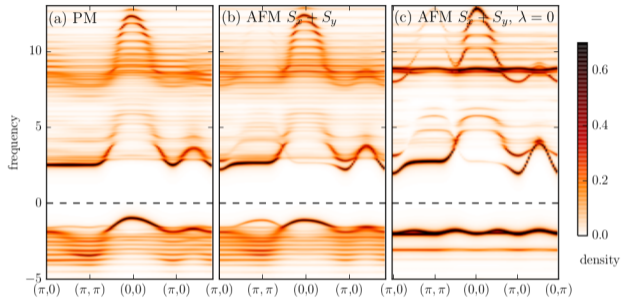


# SOC small correction to $n_{xy/yz/yz}$ and ARPES



- VCA: some changes due to  $\zeta$
  - ARPES discussed without much reference to SOC
- D. Sutter *et al.*, Nat. Comm. **8**, 15176 (2017); A. Kłosiński *et al.*, arXiv:1910.01605

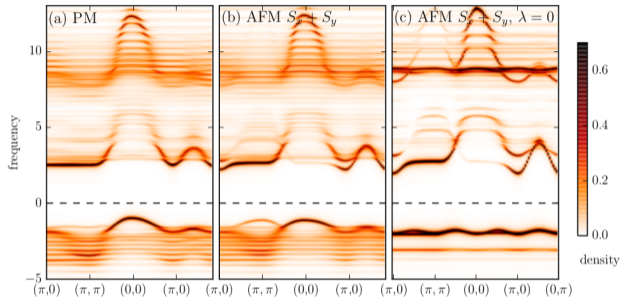
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What about magnetism?

# Magnetic Hamiltonian

- VCA:  $\vec{L} = 1$ ,  $\vec{S} = 1$  valid description (9 states/site)
- 2<sup>nd</sup> order perturbation theory  $\Rightarrow$  Kugel-Khomskii-type model  $H_{i,j} = H_{\vec{S}_i \cdot \vec{S}_j} + H_p + H_{\vec{T}_i \vec{T}_j}$

$$H_{\vec{S}_i \cdot \vec{S}_j} = J_{i,j} \left( \vec{S}_i \cdot \vec{S}_j - 1 \right) \otimes |T_i; T_j\rangle \langle T_i; T_j|, \quad J_{i,j} = \begin{cases} (t_\alpha^2 + t_\beta^2) \frac{U+J_H}{U(U+2J_H)} & \text{for } T_i = T_j = \gamma \\ \frac{t_\gamma^2(U+J_H)}{U(U+2J_H)} - \frac{J_H(t_\alpha^2+t_\beta^2)}{U(U-3J_H)} & \text{for } T_i = \alpha \neq T_j = \beta \text{ and } \alpha, \beta \neq \gamma \end{cases} \quad (1)$$

$$H_p = \left( \vec{S}_i \cdot \vec{S}_j - 1 \right) \left[ \sum_{\alpha \neq \beta} \frac{-t_\alpha t_\beta J_H}{U(U+2J_H)} |\alpha; \alpha\rangle \langle \beta; \beta| \right]. \quad (2)$$

$$H_{\vec{T}_i \vec{T}_j} = - \sum_{\beta \neq \gamma} \frac{t_\beta^2 + t_\gamma^2}{U - 3J_H} |\beta; \gamma\rangle \langle \beta; \gamma| + \sum_{\beta \neq \gamma} \frac{t_\beta t_\gamma}{U(U - 3J_H)} [2J_H + (U - J_H)(\vec{S}_i \cdot \vec{S}_j + 1)] \otimes |\beta; \gamma\rangle \langle \gamma; \beta|. \quad (3)$$

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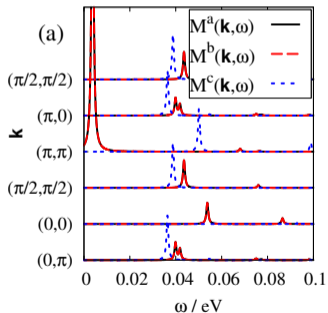
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- onsite part:  $H_{\text{ion}} = \frac{\zeta}{2} \vec{S} \vec{L} + \Delta(L^z)^2$

# Magnetic excitations spectra

$$M^\alpha(\vec{k}, \omega) = -\frac{1}{\pi} \Im \langle \phi_0 | M^\alpha(-\vec{k}) \frac{1}{\omega - H + \beta 0^+} M^\alpha(\vec{k}) | \phi_0 \rangle, \quad (4)$$

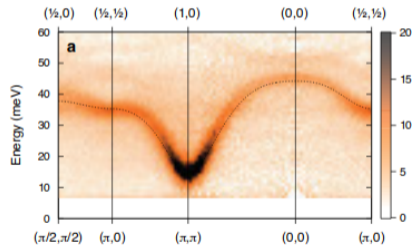
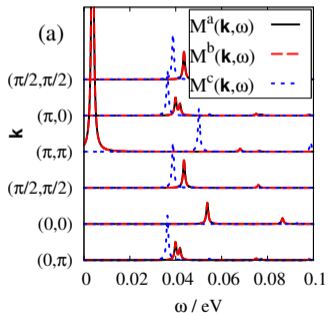
from ED for 8-site cluster, tetragonal '*ab-initio*' parameters ( $t^o = 0$ )



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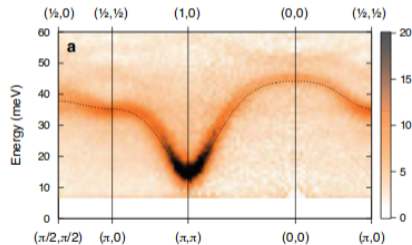
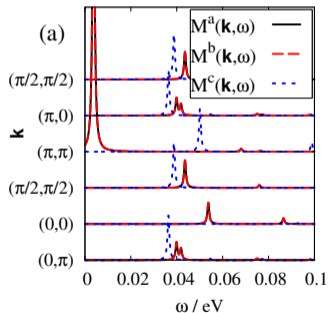


from A. Jain *et al.*, Nat. Phys. **13**, 633 (2017)

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from A. Jain *et al.*, Nat. Phys. **13**, 633 (2017)

good agreement, especially max at  $(0, 0)$ , no fitting parameters

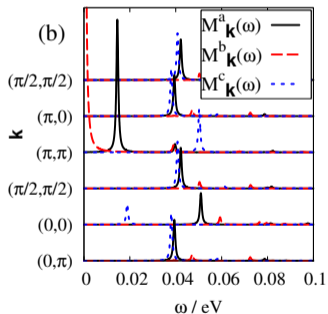


# Magnetic excitations spectra with in-plane anisotropy

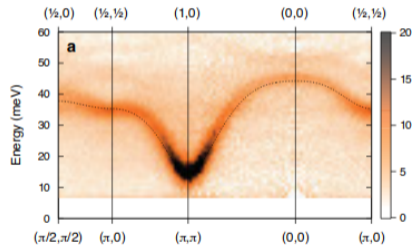
- before:  $t_{xy}^{\text{NN}} \approx 0.2$  eV,  $t_{xz}^{\text{NN}} = t_{yz}^{\text{NN}} \approx 0.14$  eV,  $t_{xy}^{\text{NNN}} \approx 0.09$  eV,  $\Delta \approx 0.25$  eV,  $t^o = 0$ ; finite  $t^o = 0.09$  made spin prefer  $b = (x - y)$  direction
- now: add  $\delta \frac{1}{2} (S^x - S^y)^2$  to mimic  $t^o$  and gap at  $(\pi, \pi)$

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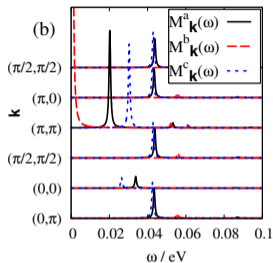
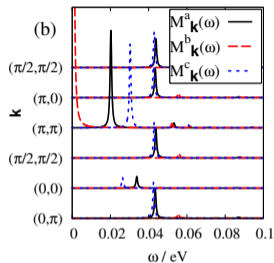
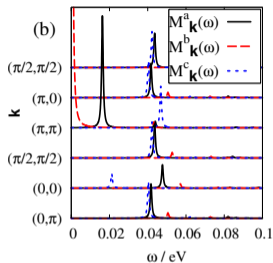
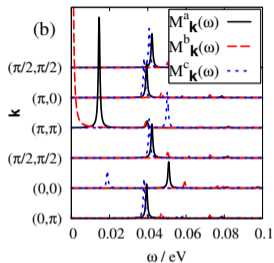


transverse modes + amplitude mode  $M^b$



from A. Jain *et al.*, Nat. Phys. **13**, 633 (2017)

# Max at (0,0) needs some (small) $n_{xy}^h$



- strong orbital polarization ( $n_{xz,yz}^h \approx 0.88$  vs.  $n_{xy}^h \approx 0.25$ )
- quite sensitive to  $n_{xy}^h$
- Second-order superexchange model + SOC +  $\Delta$  works rather well

# Thanks

- Pavel Anisimov, Friedemann Aust
- Teresa Feldmaier, Pascal Strobel
- Michael Schmid, Philipp Hansmann
- (many) discussions with Giniyat Khaliullin, George Jackeli, Hide Takagi

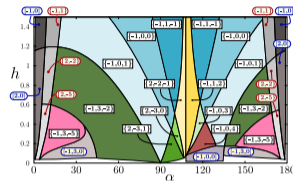


# Excitonic antiferromagnetism in $d^4$ systems

Triplons of honeycomb  $J = 0$  phase

- nontrivial triplons all over parameter space
- same ordered magnetic phases as  $j = \frac{1}{2}$ ; triplon liquid

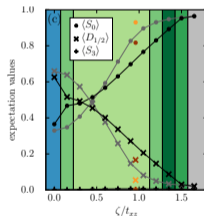
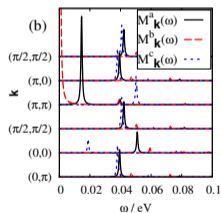
P. S. Anisimov, F. Aust, G. Khaliullin, M. Daghofer, PRL **122**, 177201 (2019)



Square-lattice models,  $\text{Ca}_2\text{RuO}_4$

- magnetic ordering via weight transfer  
 $J = 0 \rightarrow J = 1$
- valid for magnetic transition even at strong crystal fields/orbital polarization
- Kugel-Khomskii-like model + SOC +  $\Delta$  give neutron-scattering spectra

T. Feldmaier, P. Strobel, M. Schmid, P. Hansmann, M. Daghofer, arXiv:1910.13977



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