The Saga of α-$RuCl_3$. Models. Parameters. Phase Diagrams.

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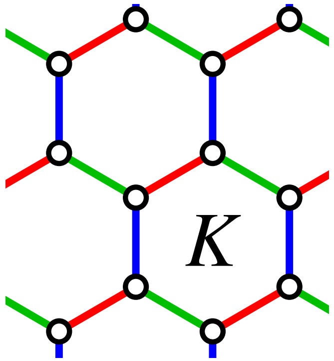
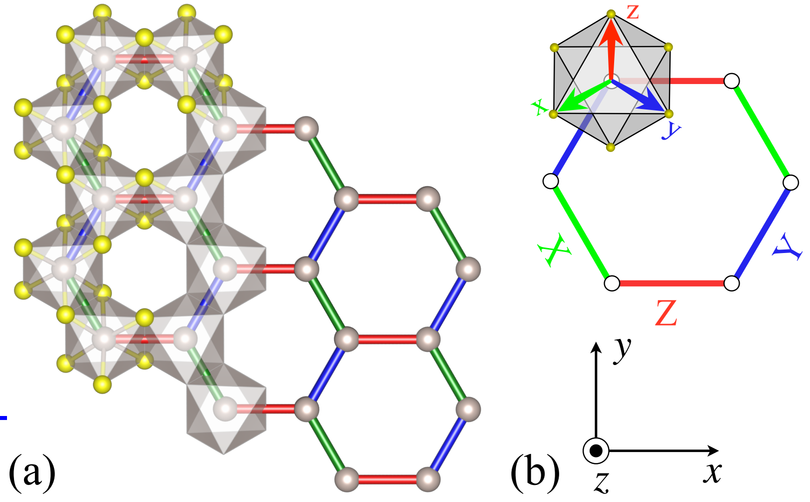
$$\hat{\mathcal{H}} = \sum_{\langle ij \rangle} \mathbf{S}_i^{\mathrm{T}} \hat{\boldsymbol{J}}_{ij} \mathbf{S}_j$$

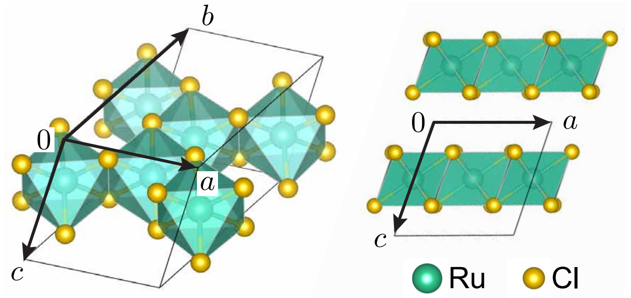
hunt for Kitaev and beyond…



anisotropic-exchange

α-$RuCl_3$





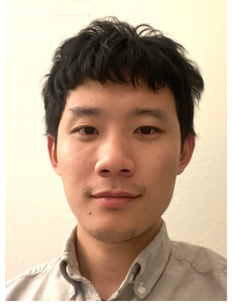
G. Jackeli and G. Khaliullin, PRL **102**, 017205 (2009).

Pavel Maksimov

LT + analytics

Marius Moeller

ED



Shengtao Jiang

DMRG

Roser Valenti

Steven White

P. A. Maksimov and **SC**, *Rethinking* α-$RuCl_3$, PRR **2**, 033011 (2020).

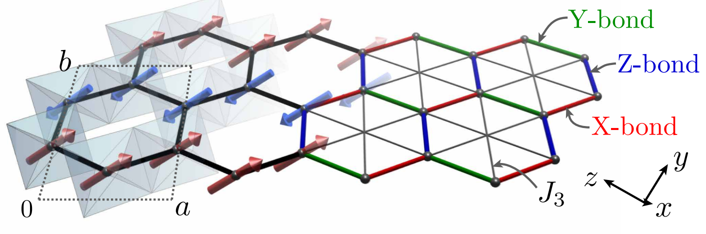
MM, PM, SJ, SW, RV, and **SC**, (unpublished).

part 1:

better parameters?

effective model

- ⇛ generalized Kitaev or J-K-Γ-Γ′(-J_3) model, ≈ **consensus**



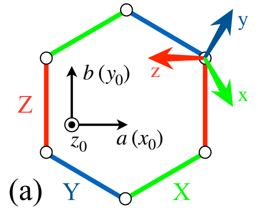
$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_1 + \hat{\mathcal{H}}_3 = \sum_{\langle ij\rangle} \mathbf{S}_i^{\mathrm{T}} \hat{\boldsymbol{J}}_{ij} \mathbf{S}_j + J_3 \sum_{\langle ij\rangle_3} \mathbf{S}_i \cdot \mathbf{S}_j$$

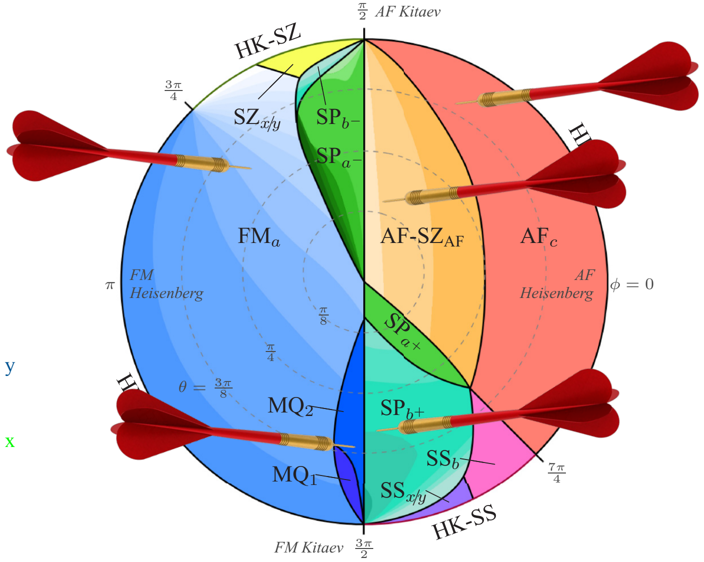
$$\gamma = \{X, Y, Z\}$$

$$\mathcal{H}_1 = \sum_{\langle ij\rangle_\gamma} \Big\{ J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right) + \Gamma'\left(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma\right)\Big\}$$

- how to constrain parameters?
  ⇛ use **phenomenology**
  ⇛ effects that would not be there if not for the anisotropic terms?

- (*) note: K=Γ=Γ′=0 ⇛ J_1-J_3 FM-AFM (Heisenberg) model



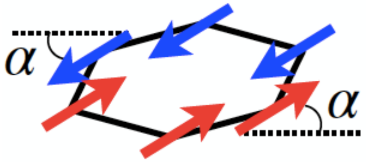


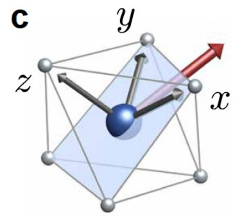
P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

MM, PM, SJ, SW, RV, and **SC**, (unpublished).

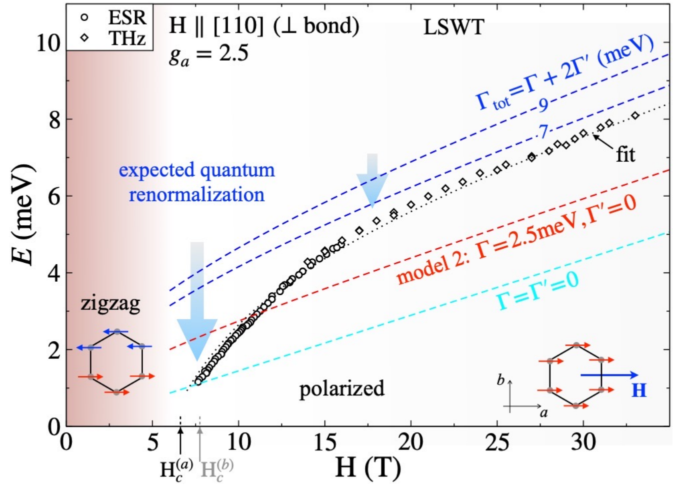
phenomenological constraints

- B=0, zigzag order, **tilted** out of plane of Ru^{3+} ions [$\alpha \approx 35^\circ$]

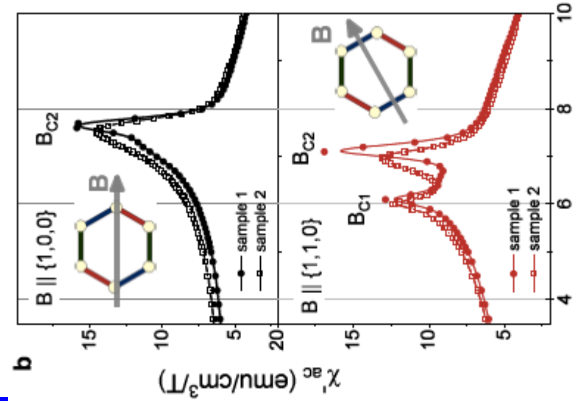




- high field **k**=0, spin-flip (ESR, Raman) ⇛ **non-linear vs H**



- in-plane critical field are **nearly equal**: $H_{c,a} \approx H_{c,b} \approx$ 6-7 T



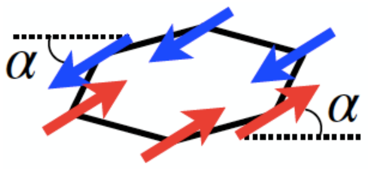
Winter *etal.*, 2017. | Sears *etal.*, 2020. | Wolter *etal.*, 2017. | Sahasrabudhe *etal.*, 2019. | P. Lampen-Kelley *etal.*, 2018.

other ways ...

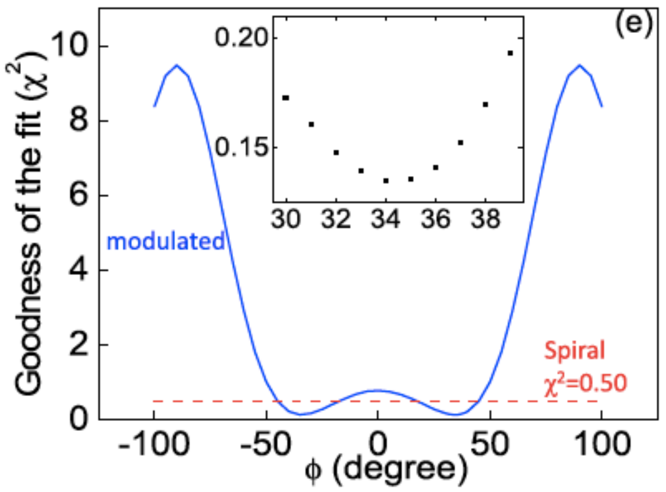
Reference	Method	K	Γ	Γ'	α (°)	Γ+2Γ'	ΔH (T)
Kim et al. [29]	DFT+t/U, $P3$	-6.55	5.25	-0.95	**36.6**	3.35	9.64
	DFT+SOC+t/U	-8.21	4.16	-0.93	40.9	2.3	7.03
	same+fixed lattice	-3.55	7.08	-0.54	28.4	6.01	14.4
Winter et al. [30]	DFT+ED, $C2$	-6.67	6.6	-0.87	**34.4**	4.87	12.2
Ran et al. [34]	LSWT, INS fit	-6.8	9.5		**30.1**	**9.5**	16.6
Hou et al. [31]	DFT+t/U, U=2.5eV	-14.4	6.43		41.1	6.43	7.96
	same, U=3.0eV	-12.2	4.83		42.2	4.83	5.74
	same, U=3.5eV	-10.7	3.8		43.2	3.8	4.36
Wang et al. [32]	DFT+t/U, $P3$	-10.9	6.1		38.9	6.1	8.15
	same, $C2$	-5.5	7.6		**30.2**	**7.6**	13.3
Winter et al. [35]	*Ab initio*+INS fit	-5.0	2.5		40.0	2.5	3.22
Suzuki et al. [36]	ED, C_p fit	-24.4	5.25	-0.95	47.3	3.35	6.76
Cookmeyer et al. [37]	thermal Hall fit	-5.0	2.5		40.0	2.5	3.22
Wu et al. [38]	LSWT, THz fit	-2.8	2.4		**34.6**	2.4	3.68
Ozel et al. [39]	same	-3.5	2.35		**37.0**	2.35	3.34
Eichstaedt et al. [33]	DFT+Wannier+t/U	-14.3	9.8	-2.23	38.3	5.33	18.1
Sahasrabudhe et al.[42]	ED, Raman fit	-10.0	3.75		42.7	3.75	4.38
Sears et al. [40]	Magnetization fit	-10.0	10.6	-0.9	**33.4**	**8.8**	19.0
		-10.0	8.8		**34.3**	**8.8**	13.6
Laurell et al. [41]	ED, C_p fit	-15.1	10.1	-0.12	**37.2**	**9.86**	14.6
Suzuki et al. [43]	RIXS	-5.0	2.5	+0.1	39.8	2.7	3.03
Kaib et al. [44]	GGA+U	-10.1	9.35	-0.73	**34.5**	**7.89**	16.0
Andrade et al. [45]	χ	-6.6	6.6		**33.1**	6.6	10.6
Janssen et al. [46]	LSWT+3D	-10.0	5.0		40.0	5.0	6.43
Li et al. [47]	C_m, χ	-25.0	7.5	-0.5	44.8	6.5	9.03
Ran et al. [48]	polarized INS	-7.2	5.6		**35.6**	5.6	8.33
Samarakoon et al. [49]	Machine learning, INS	-5.3	0.15		**36.4**	0.15	**0.11**
Liu et al. [50]	downfolding	-5.0	2.8	+0.7	**37.3**	4.2	2.37

#1: tilt angle α

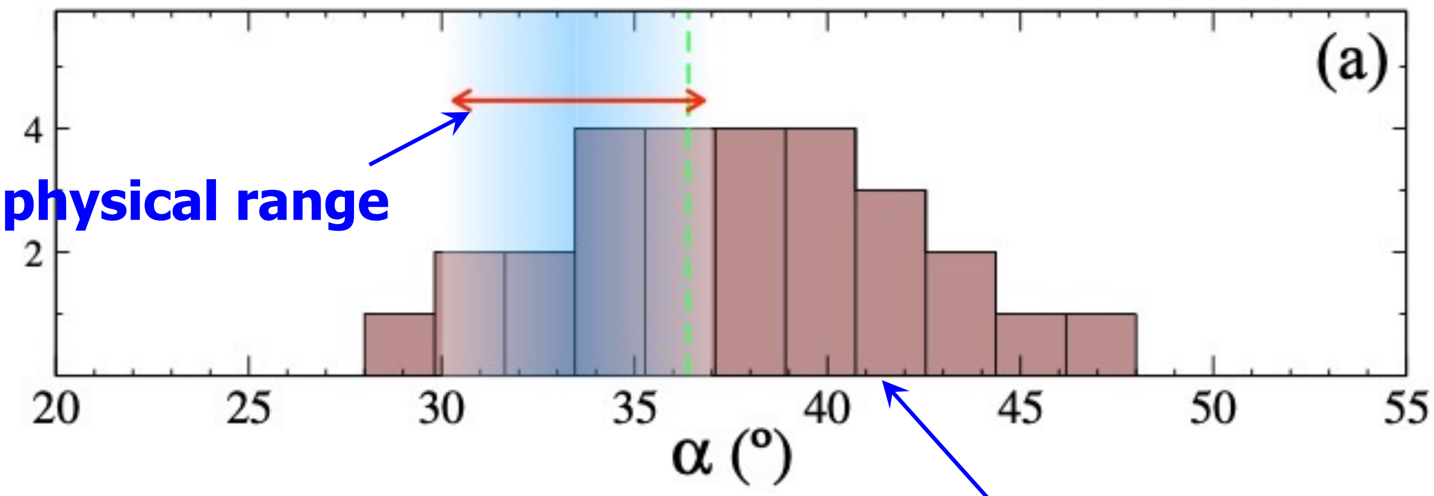
- (classical) tilt angle **α** depends **only** on K, Γ, and Γ′
- -- experiments: α ≈ 32°..35°, ED suggest modest quantum corrections



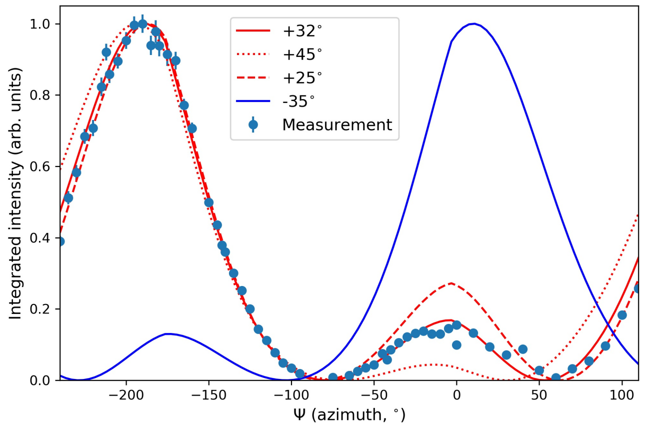
$$\tan 2\alpha = 4\sqrt{2}\cdot\frac{\Gamma - K - \Gamma'}{7\Gamma + 2K + 2\Gamma'}$$



physical range



histogram from the table



- ⇛ physical range
  30° ≲ α ≲ 37°

J. Chaloupka and G. Khaliullin, 2016; H. B. Cao *etal.*, 2016; Sears *etal.*, 2020.

MM, PM, SJ, SW, RV, and **SC**, (unpublished).

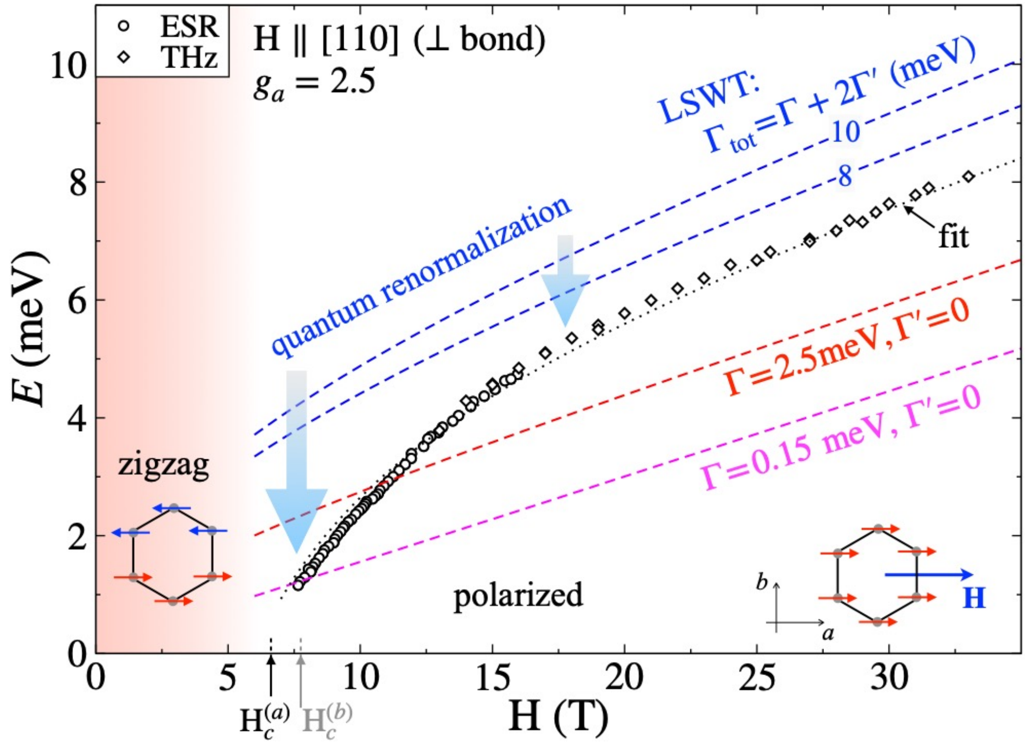
#2: ESR, THz, Raman (**k**=0 spin-flip)

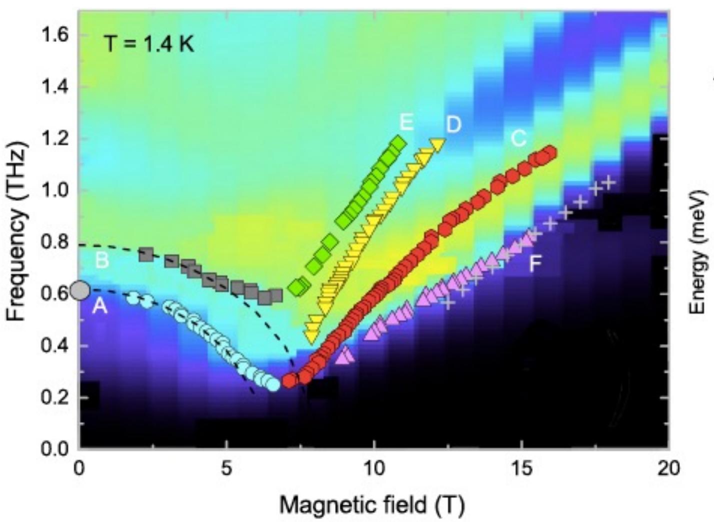
- strong in-plane field, **k=0 spin-flip excitation**
- LSWT: $E_{\mathbf{k}=0}$ depends **only** on Γ and Γ′

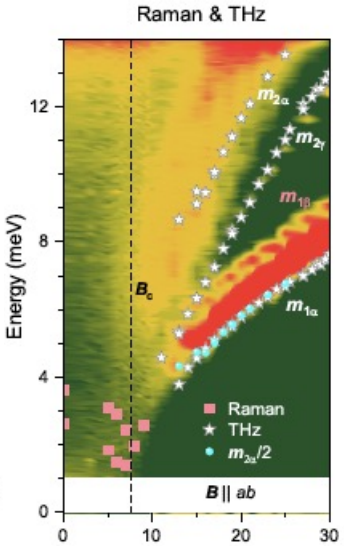
$$\varepsilon_0^{(0)} = \sqrt{h\left(h + 3S(\Gamma + 2\Gamma')\right)}, \qquad h = g\mu_B H$$

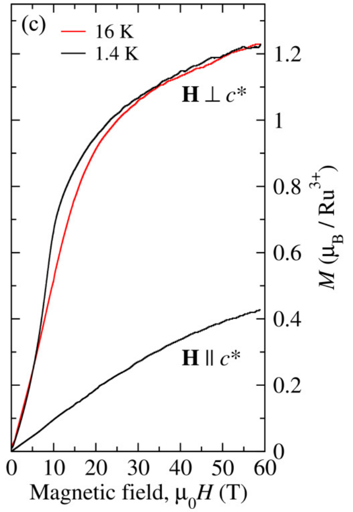
-- fluctuations renormalize $E_{\mathbf{k}=0}$ **down** [RPA, ED]

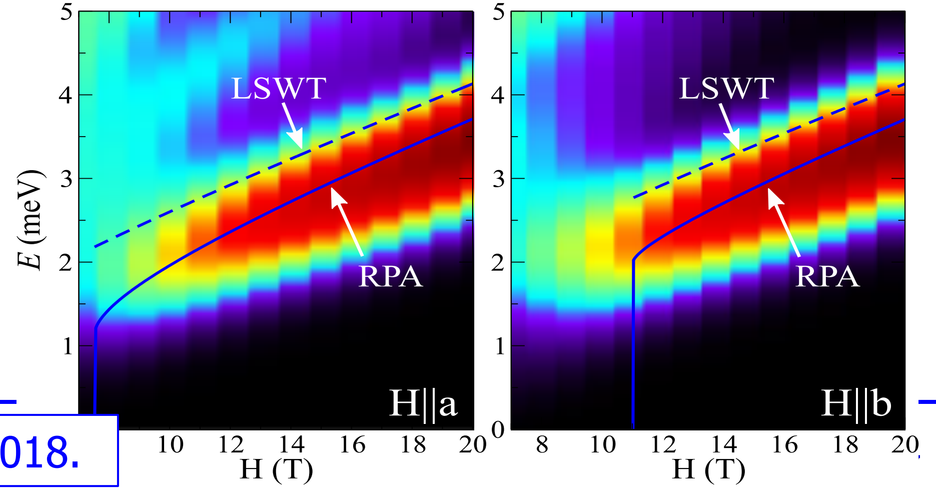
- ⇛ strong upper and lower bounds on 7.5 meV ≲ **Γ+2Γ′** ≲ 10 meV











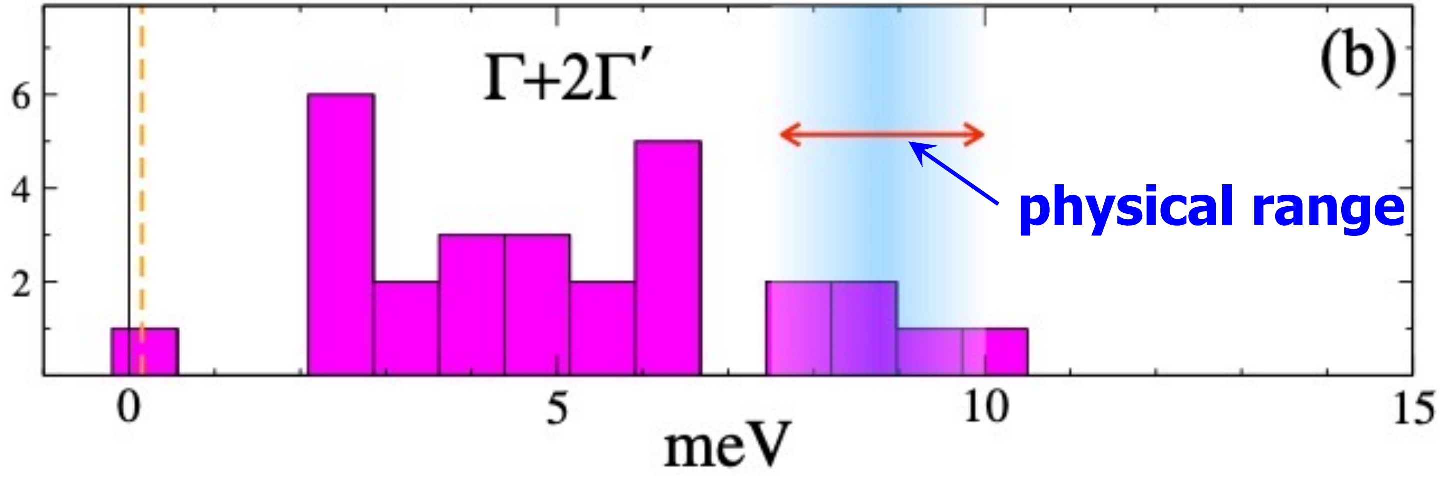
A. N. Ponomaryov *etal.*, 2017; A. Sahasrabudhe *etal.*, 2019.

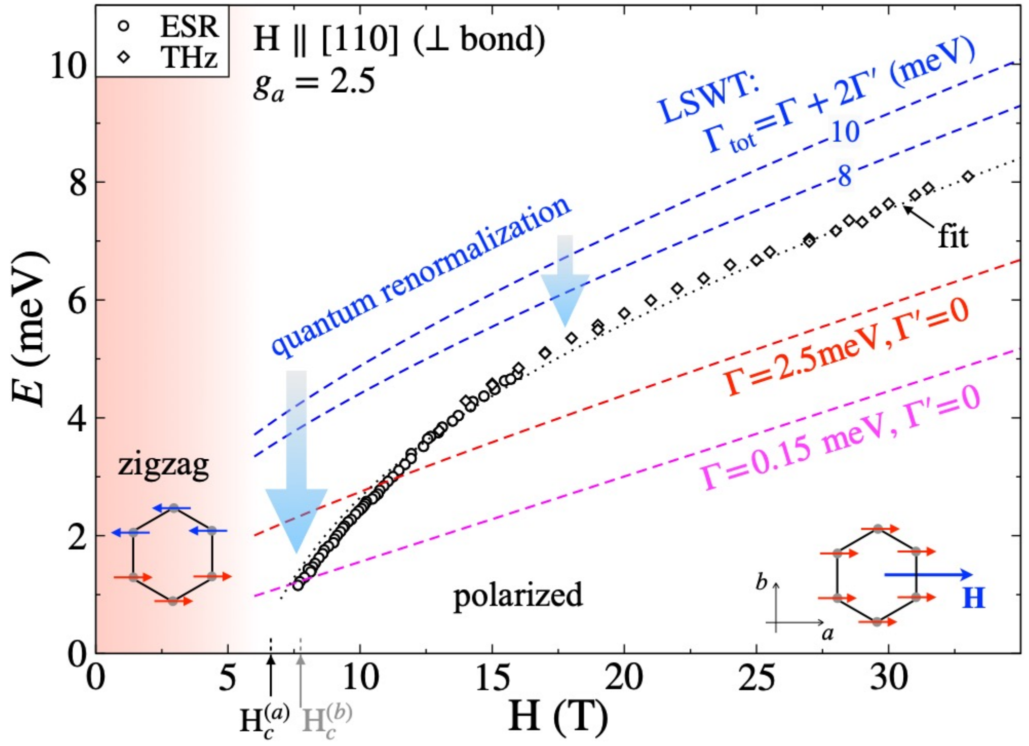
S. M. Winter *etal.*, 2018.

#2: "ESR parameter"

$$\varepsilon_0^{(0)} = \sqrt{h\left(h + \boxed{3S(\Gamma + 2\Gamma')}\right)}, \qquad h = g\mu_B H$$

○ ⇛ physical range
7.5 meV ≲ **Γ+2Γ′** ≲ 10 meV





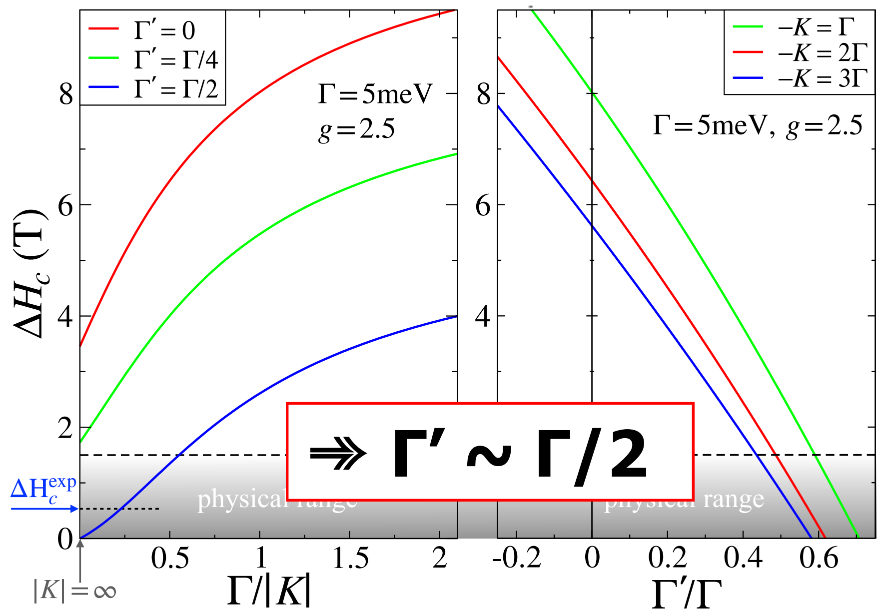
P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

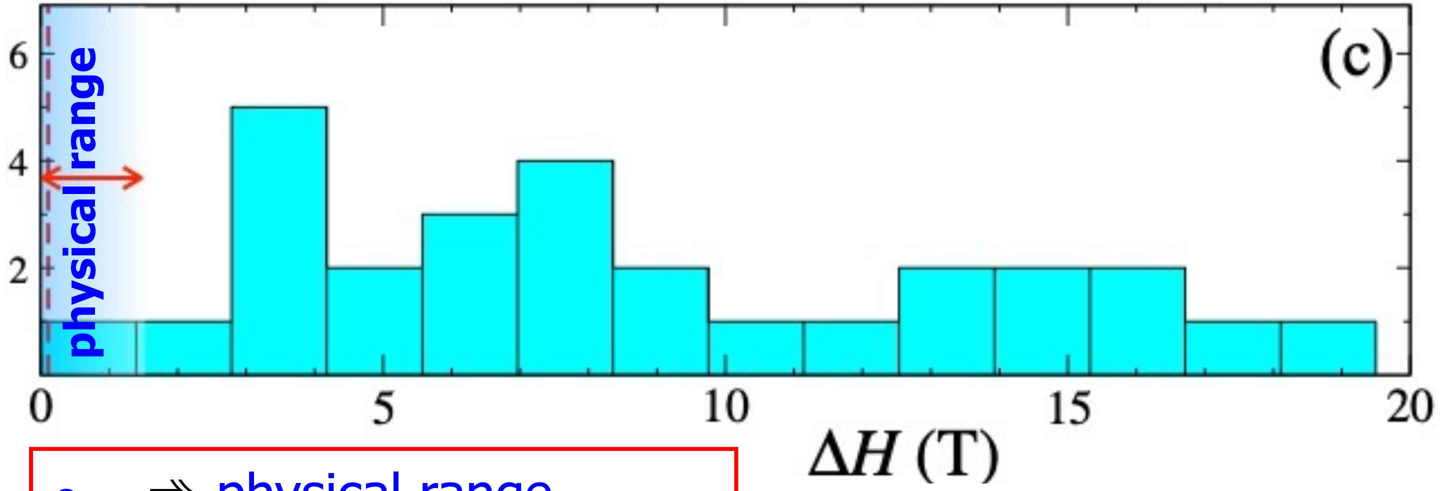
MM, PM, SJ, SW, RV, and **SC**, (unpublished).

#3: in-plane critical fields $H_{c,a} \approx H_{c,b}$

- how is $H_{c,a} \approx H_{c,b}$??
- LSWT: $\Delta H_c = H_{c,a} - H_{c,b}$ depends **only** on K, Γ, and Γ′ !
- -- quantum fluctuations? ⇛ small ΔH_c will stay small
- -- is **impossible** to reconcile without **Γ′ ~ Γ/2 > 0**

previous works: **underutilized** parameter space ⇛ **Γ′ ~ Γ/2**





- ⇛ physical range
  $0\ \mathrm{T} \lesssim \Delta H_c \lesssim 1.5\ \mathrm{T}$

$$h_c^{(a)} = J + 3J_3 + \frac{1}{12}(5K - 5\Gamma - 16\Gamma') + \frac{1}{12}\sqrt{(K + 5\Gamma + 4\Gamma')^2 + 24(K - \Gamma + \Gamma')^2},$$

$$h_c^{(b)} = J + 3J_3 + \frac{1}{4}(2K - \Gamma - 6\Gamma') + \frac{1}{12}\sqrt{(2K + 7\Gamma + 2\Gamma')^2 + 32(K - \Gamma + \Gamma')^2},$$

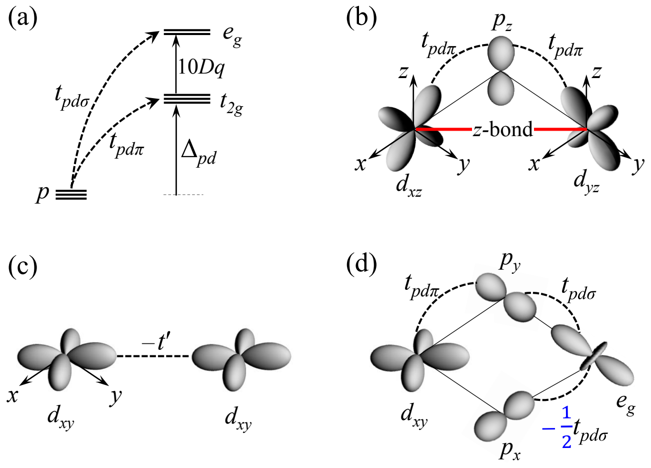
P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

P. Lampen-Kelley *etal.*, 2018.

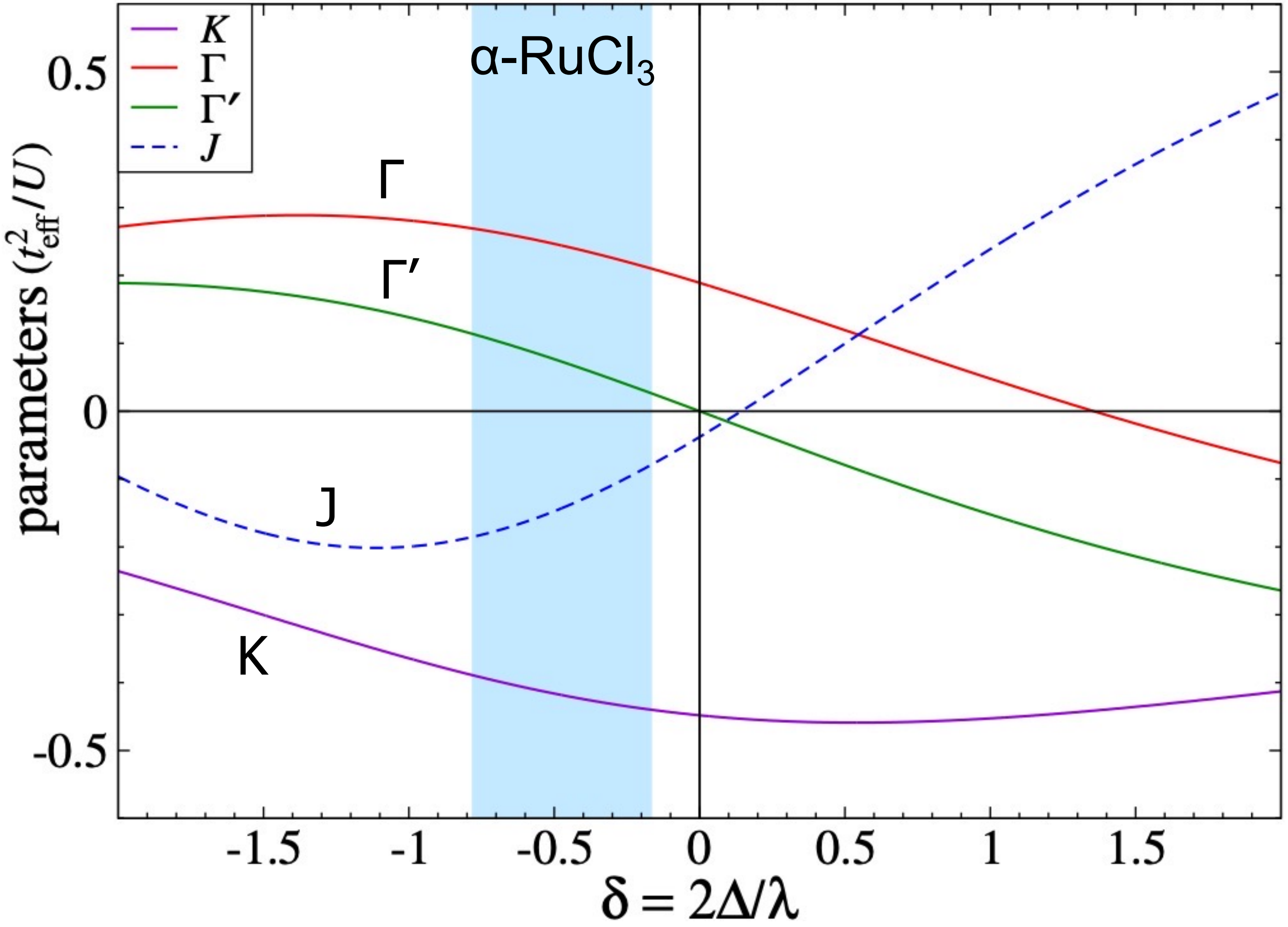
MM, PM, SJ, SW, RV, and **SC**, (unpublished).

present from Giniyat…

- ***"t/U"*** –downfolding/projection



Γ′>0, not small!

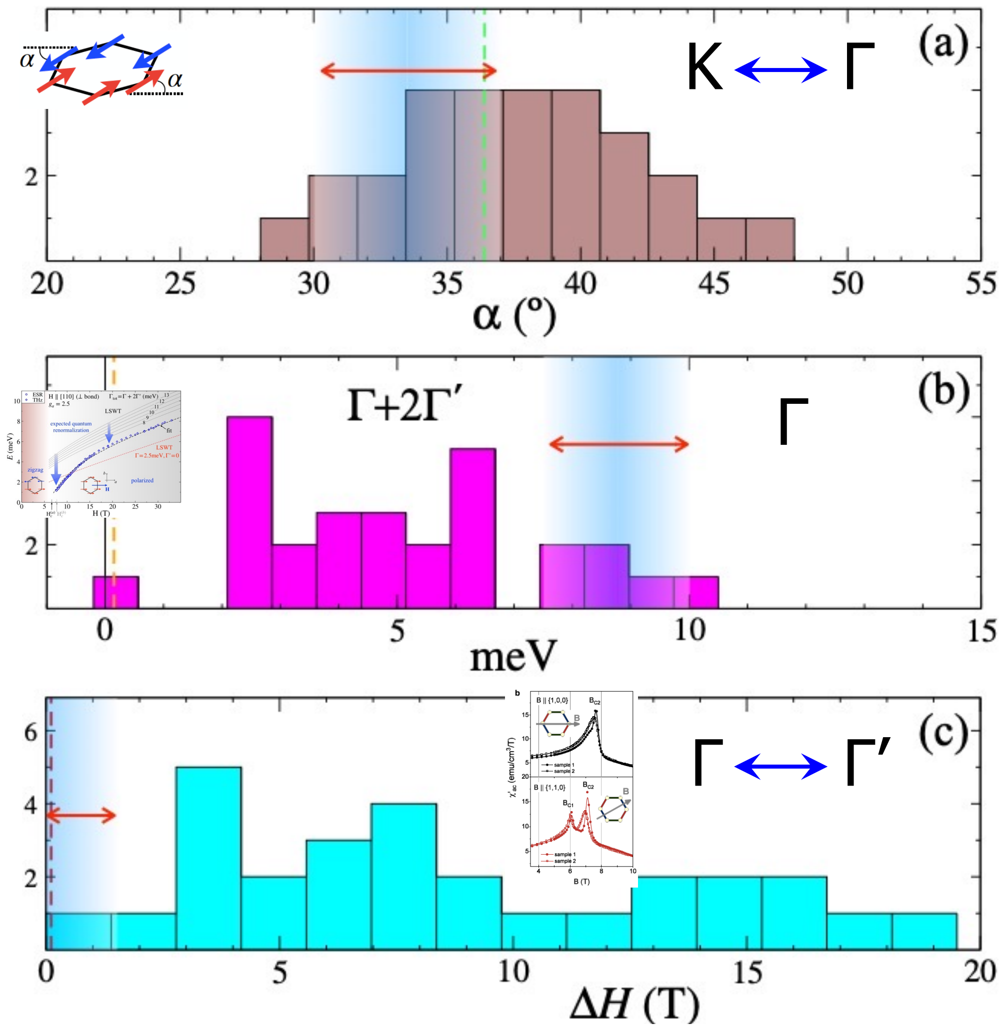


Liu, Chaloupka, and Khaliullin, PRB (2022)

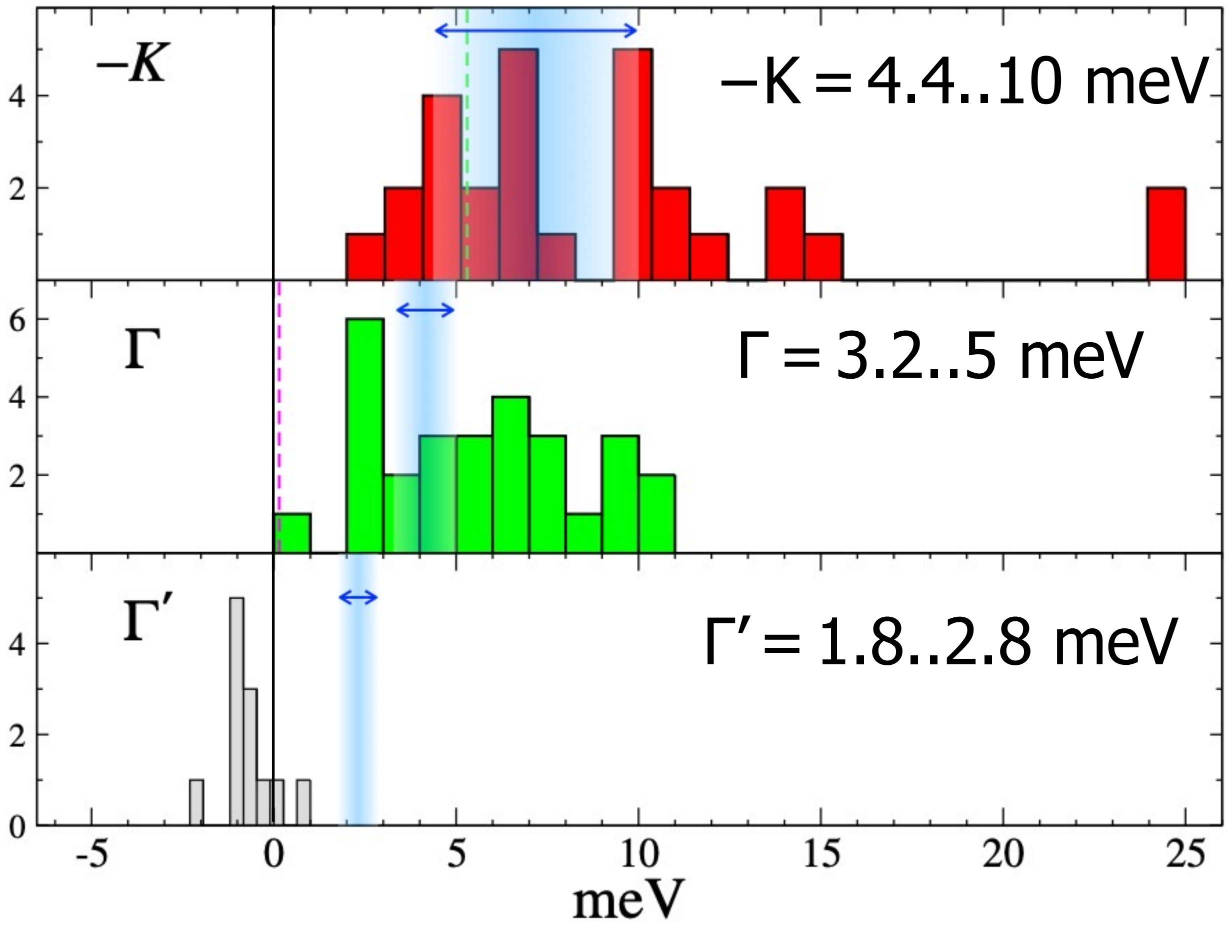
MM, PM, SJ, SW, RV, and **SC**, (unpublished).

3 constraints, 3 parameters

- $\Gamma+2\Gamma' = 7.5-10$ meV; $\Delta H_c = 0-1.5$ T; $\alpha = 30^\circ-37^\circ$ ⇛ strongest bounds are on Γ and Γ′



⇛



P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

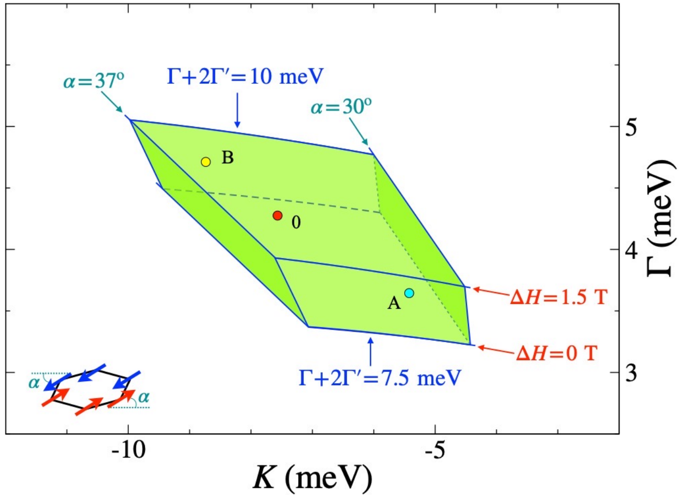
MM, PM, SJ, SW, RV, and **SC**, (unpublished).

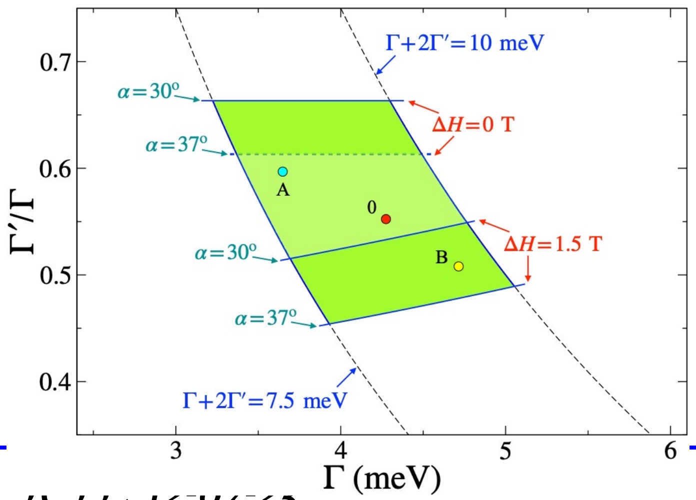
point #0: J-J_3 phase diagram

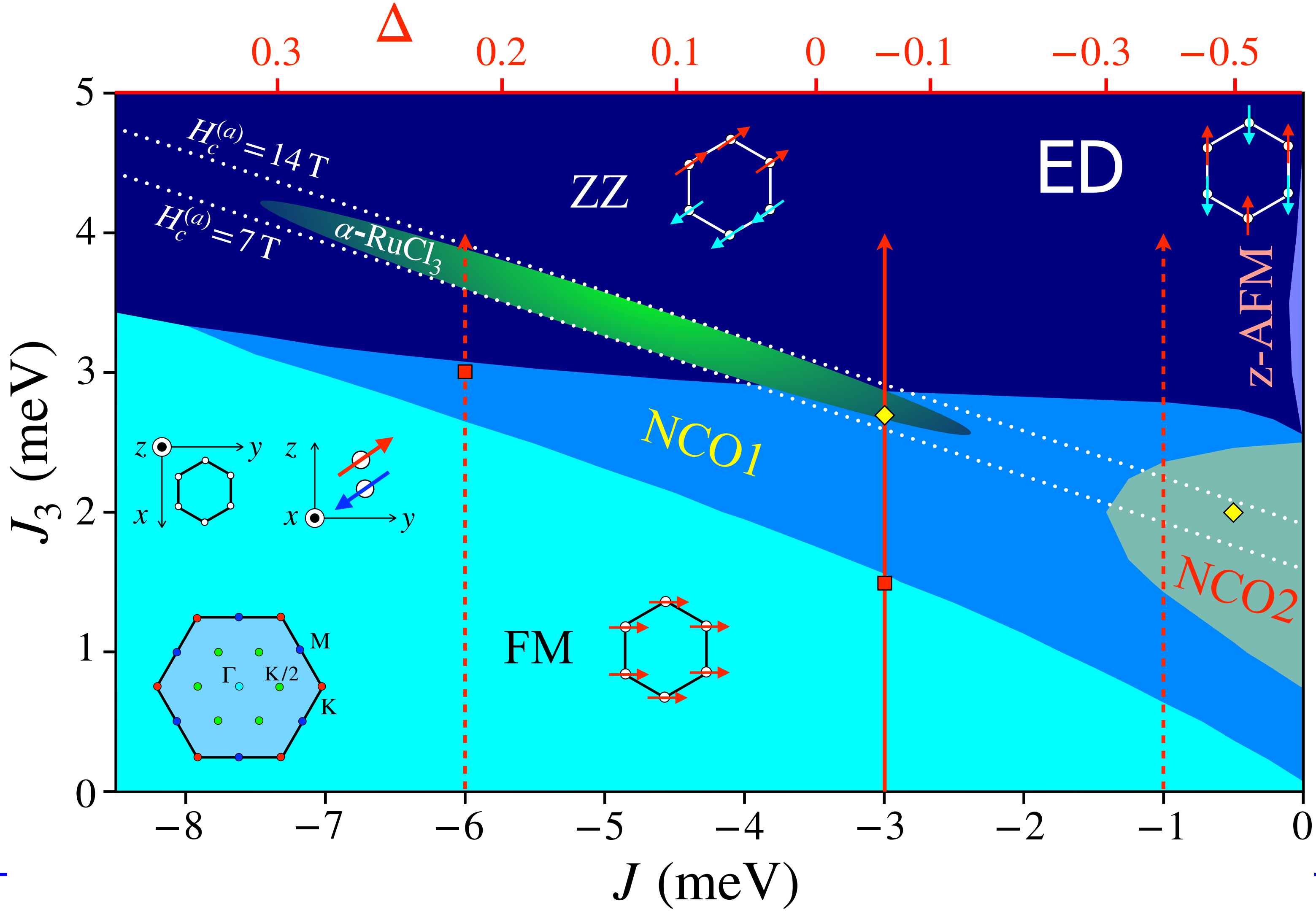
$\Gamma + 2\Gamma' = 9$ meV, $\Delta H_c = 0.8$ T, $\alpha = 35°$

$\{K, \Gamma, \Gamma'\} = \{-7.567, 4.276, 2.362\}$ meV

$$\mathcal{H}_1 = \sum_{\langle ij\rangle_\gamma} \left\{ J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right) + \Gamma'\left(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma\right)\right\}$$



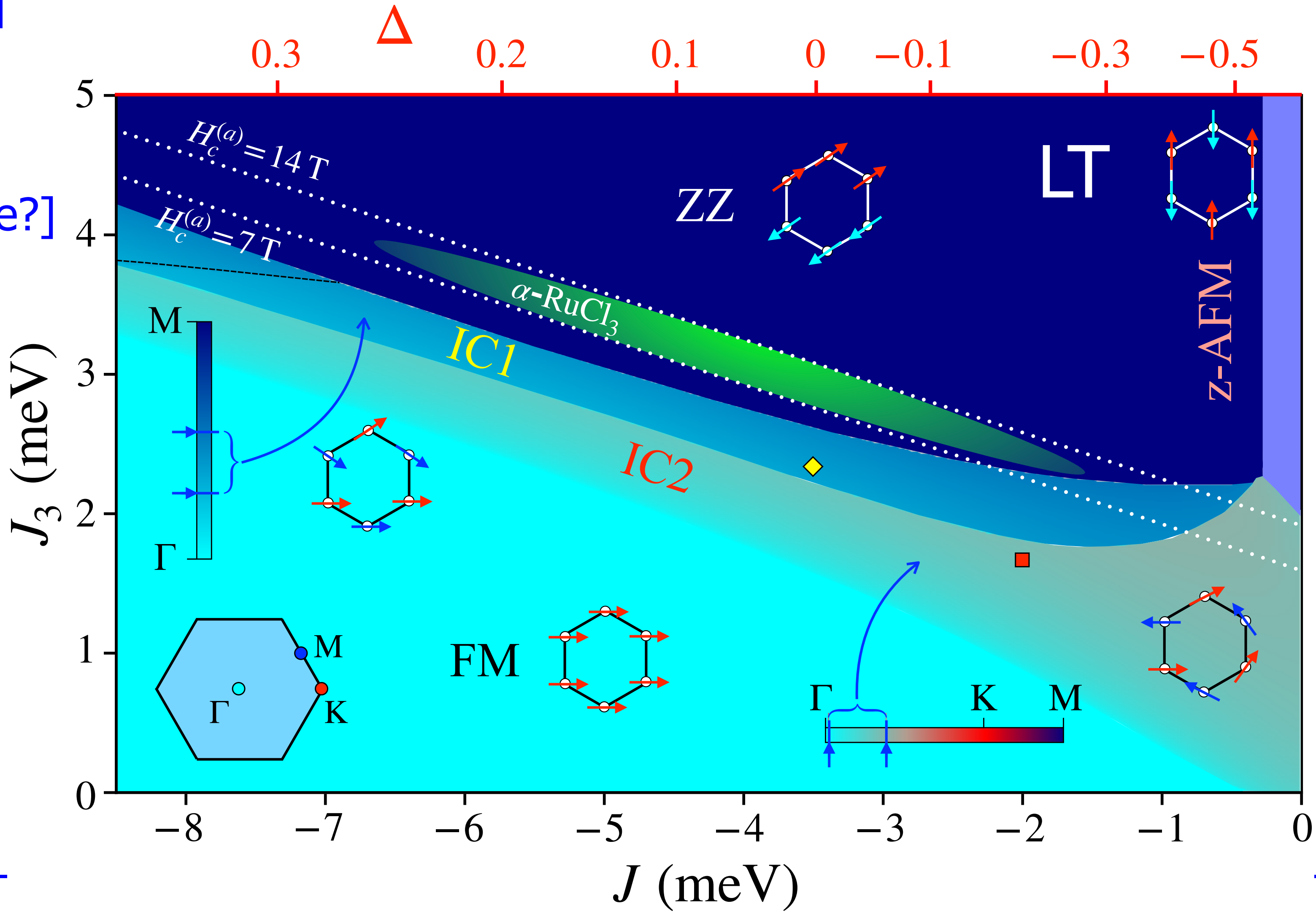




main point

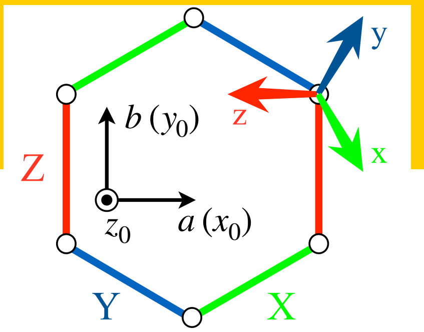
- ☑ phenomenology ⇛
  strong constraints on α-$RuCl_3$ model

- ☑ α-$RuCl_3$ is proximate to an
  incommensurate phase [what type?]



part B:
better axes??

XXZ-$J_{\pm\pm}$-$J_{z\pm}$ model virtues?



$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_1 + \hat{\mathcal{H}}_3 = \sum_{\langle ij\rangle} \mathbf{S}_i^{\mathrm{T}} \hat{\boldsymbol{J}}_{ij} \mathbf{S}_j + J_3 \sum_{\langle ij\rangle_3} \mathbf{S}_i \cdot \mathbf{S}_j$$

$$\mathcal{H}_1 = \sum_{\langle ij\rangle_\gamma} \Big\{ J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma\big(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\big) + \Gamma'\big(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma\big)\Big\}$$

$$\tilde{\varphi}_\alpha = \{0, 2\pi/3, -2\pi/3\}$$

$$\mathcal{H}_1 = \sum_{\langle ij\rangle} \Big\{ \mathrm{J}_1\big[\Delta S_i^z S_j^z + S_i^x S_j^x + S_i^y S_j^y\big]$$

XXZ

$$- 2\mathrm{J}_{\pm\pm}\big[\cos\tilde{\varphi}_\alpha \left(S_i^x S_j^x - S_i^y S_j^y\right) - \sin\tilde{\varphi}_\alpha \left(S_i^x S_j^y + S_i^y S_j^x\right)\big]$$

$$- \mathrm{J}_{z\pm}\big[\cos\tilde{\varphi}_\alpha \left(S_i^x S_j^z + S_i^z S_j^x\right) + \sin\tilde{\varphi}_\alpha \left(S_i^y S_j^z + S_i^z S_j^y\right)\big]\Big\}$$

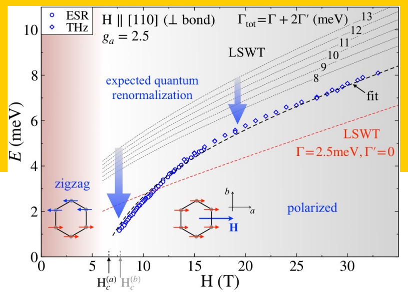
bond-dependent

- exchange matrix **not invariant** under axis transformation
- ⇛ more **intuitive** terms and quantities; fewer bond-dependent terms
- ⇛ connection to other models in frustrated magnetism
- ⇛ **fewer terms**?, simpler model?

P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

MM, PM, SJ, SW, RV, and **SC**, (unpublished).

intuitive quantities…

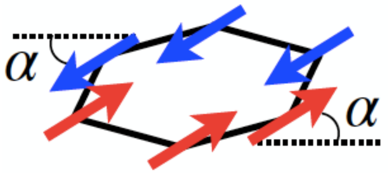


- ESR gap, $E_{\mathbf{k}=0}$, for **k=0** spin-flip excitation

$$\varepsilon_0^{(0)} = \sqrt{h\big(h+3S(\Gamma+2\Gamma')\big)}, \quad \Rightarrow \quad \varepsilon_0^{(0)} = \sqrt{h\big(h-3S\mathrm{J}_1(1-\Delta)\big)}$$

$$h = g\mu_B H$$

- ⇒ Γ+2Γ′ is **just** a complicated way of writing **easy-plane anisotropy**
- tilt angle



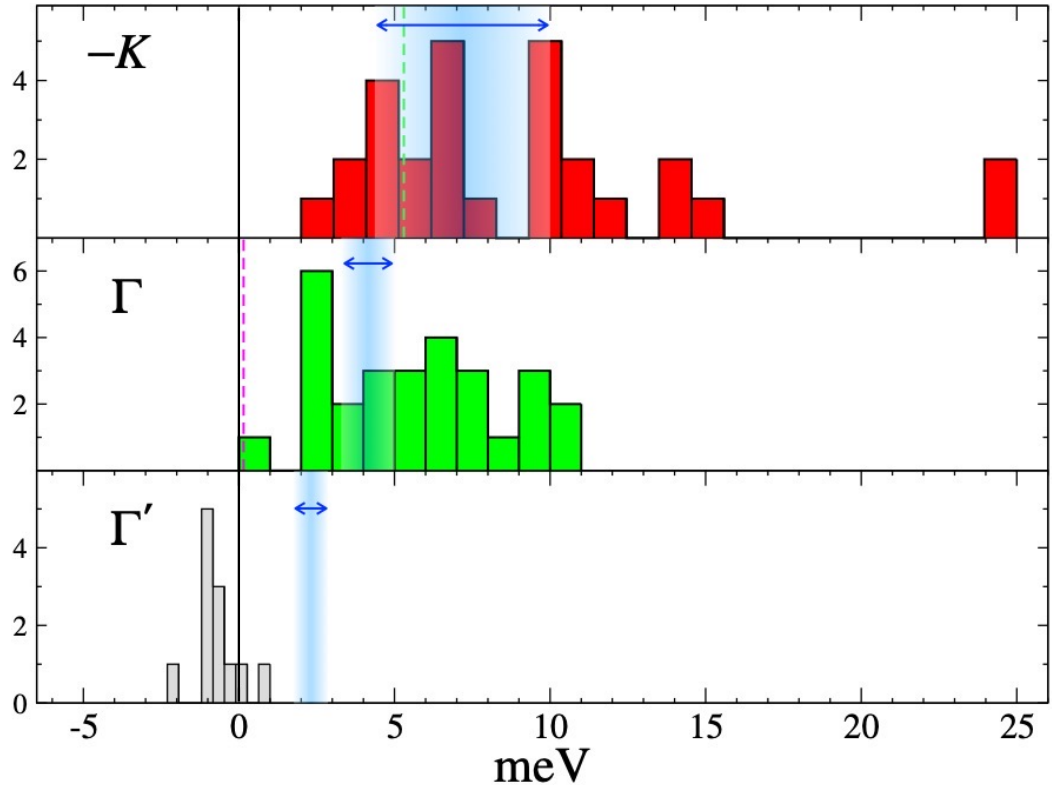
$$\tan 2\alpha = 4\sqrt{2}\cdot\frac{\Gamma-K-\Gamma'}{7\Gamma+2K+2\Gamma'} \quad \Rightarrow \quad \tan 2\alpha = \frac{4\mathrm{J}_{z\pm}}{\mathrm{J}_1(1-\Delta)+4\mathrm{J}_{\pm\pm}}$$

- ⇒ **key** term: $\mathrm{J}_{z\pm}$ (**naturally** yields the out-of-plane tilt of spins)

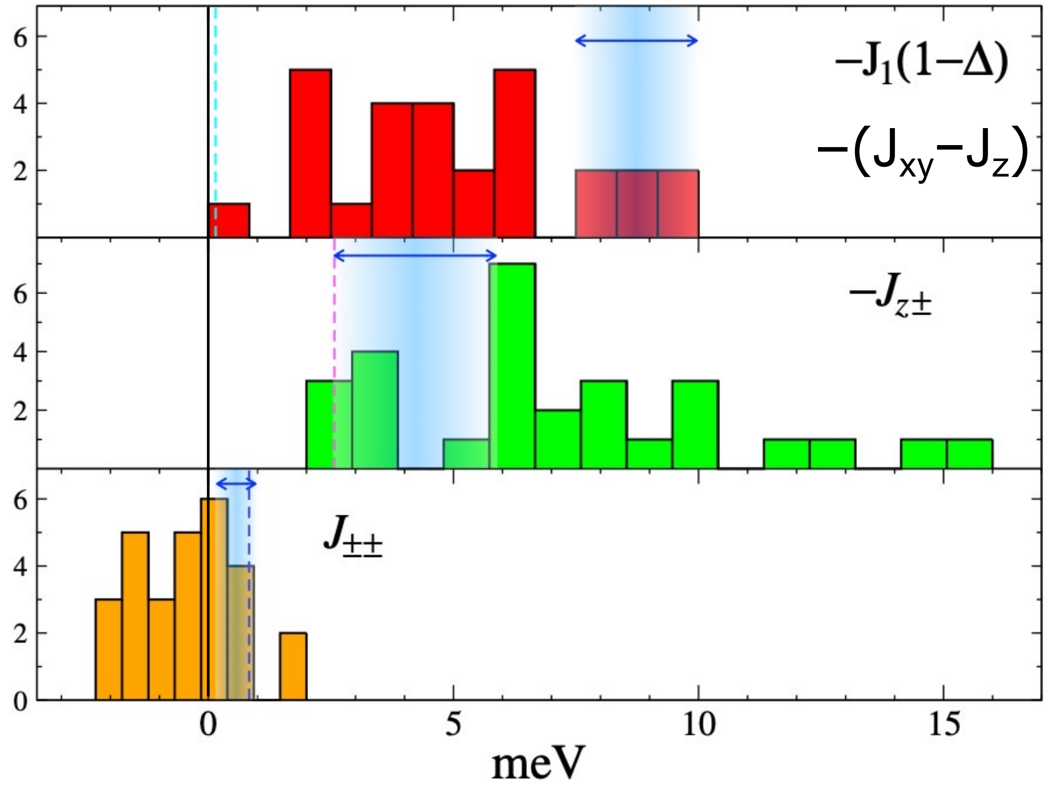
$$\mathcal{H}_1 = \sum_{\langle ij\rangle}\Big\{\mathrm{J}_1\big[\Delta S_i^z S_j^z + S_i^x S_j^x + S_i^y S_j^y\big] - 2\mathrm{J}_{\pm\pm}\big[\cos\tilde{\varphi}_\alpha\,(S_i^x S_j^x - S_i^y S_j^y) - \sin\tilde{\varphi}_\alpha\,(S_i^x S_j^y + S_i^y S_j^x)\big] - \mathrm{J}_{z\pm}\big[\cos\tilde{\varphi}_\alpha\,(S_i^x S_j^z + S_i^z S_j^x) + \sin\tilde{\varphi}_\alpha\,(S_i^y S_j^z + S_i^z S_j^y)\big]\Big\}$$

MM, PM, SJ, SW, RV, and **SC**, (unpublished).

K-J-Γ-Γ' ⇛ J_1-Δ-[XXZ]-$J_{\pm\pm}$-$J_{z\pm}$ translation



⇛



$$J_1 = J + \frac{1}{3}(K - \Gamma - 2\Gamma'),$$
$$\Delta J_1 = J + \frac{1}{3}(K + 2\Gamma + 4\Gamma'),$$
$$2J_{\pm\pm} = -\frac{1}{3}(K + 2\Gamma - 2\Gamma'),$$
$$\sqrt{2}J_{z\pm} = \frac{2}{3}(K - \Gamma + \Gamma').$$

- **conversion** of $K, J < 0$, $\Gamma > 0$
- J_1, $J_{z\pm}$ ⇛ **add up,**
- Δ, $J_{\pm\pm}$ ⇛ **cancel out**

⇛ simplification: $\Delta = J_z/J_{xy} \approx 0$,

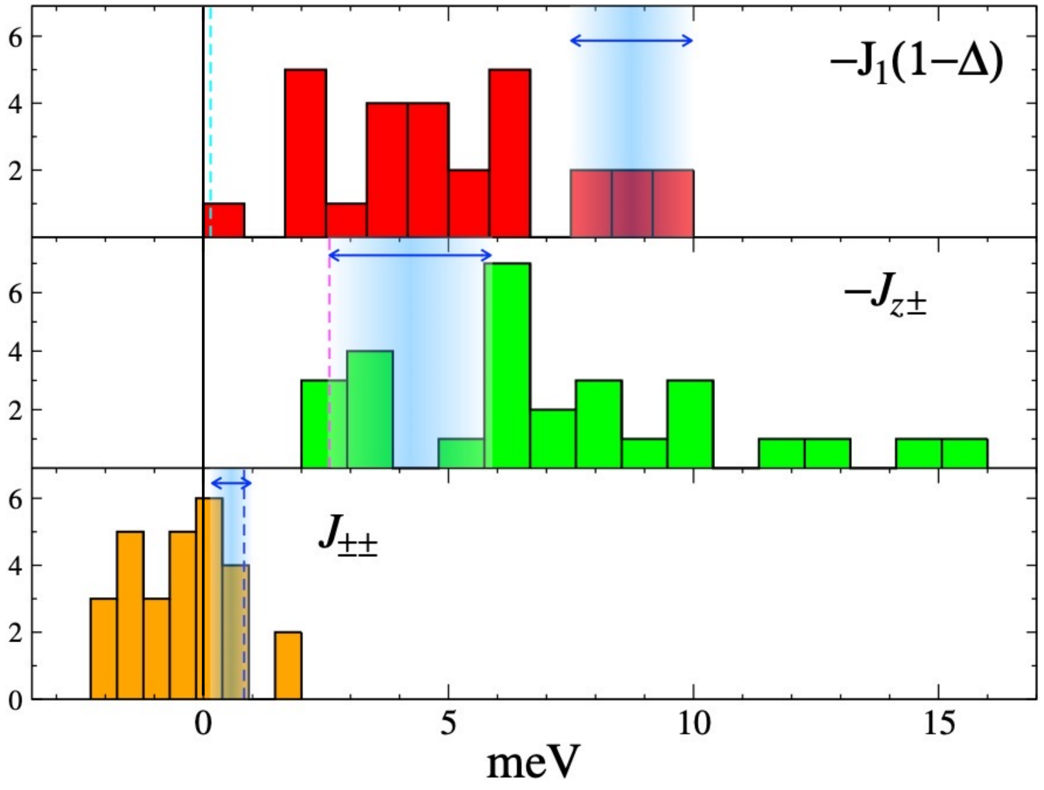
⇛ can neglect $J_{\pm\pm}$

P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

MM, PM, SJ, SW, RV, and **SC**, (unpublished).

fewer parameters…

$$\mathcal{H}_1 = \sum_{\langle ij \rangle} \Big\{ \mathrm{J}_1 \big[\Delta S_i^z S_j^z + S_i^x S_j^x + S_i^y S_j^y \big]$$
$$- 2\mathrm{J}_{\pm\pm} \big[\cos\tilde{\varphi}_\alpha \left(S_i^x S_j^x - S_i^y S_j^y \right) - \sin\tilde{\varphi}_\alpha \left(S_i^x S_j^y + S_i^y S_j^x \right) \big]$$
$$- \mathrm{J}_{z\pm} \big[\cos\tilde{\varphi}_\alpha \left(S_i^x S_j^z + S_i^z S_j^x \right) + \sin\tilde{\varphi}_\alpha \left(S_i^y S_j^z + S_i^z S_j^y \right) \big] \Big\}$$



many prior attempts to infer α-$RuCl_3$ parameters:

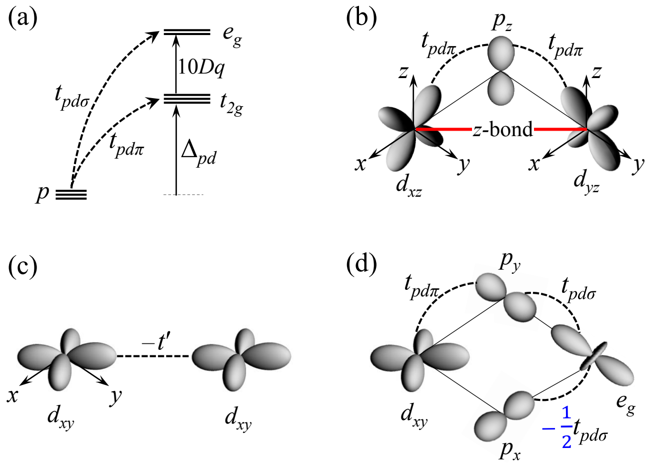
- implied J_1-$J_{z\pm}$-J_3 model with **easy-plane** FM J_1, AFM J_3, + large $J_{z\pm}$

P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

MM, PM, SJ, SW, RV, and **SC**, (unpublished).

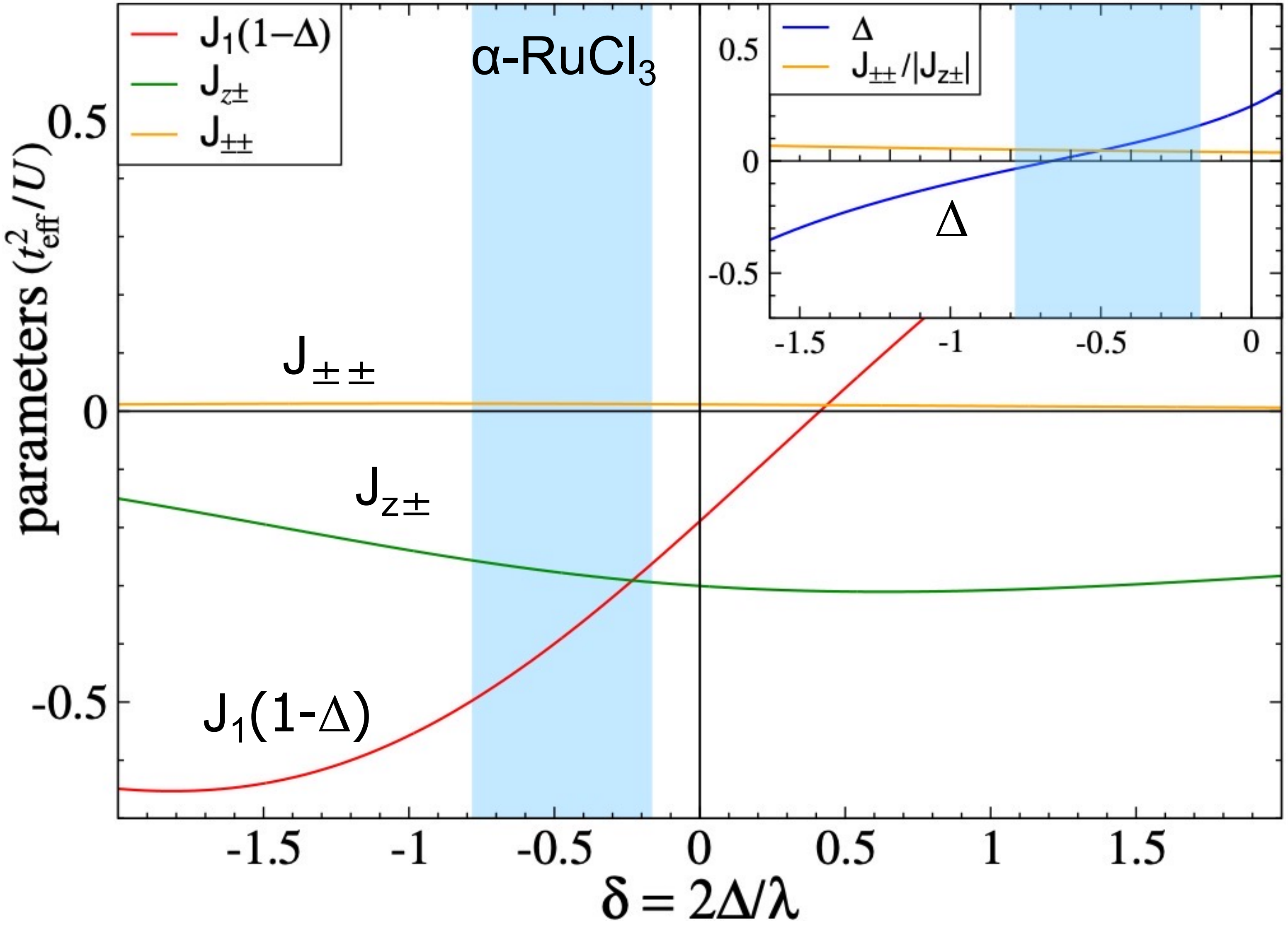
present from Giniyat…

- ***"t/U"***–downfolding/projection



$\Delta \approx 0$!

$J_{\pm\pm} \approx 0$!

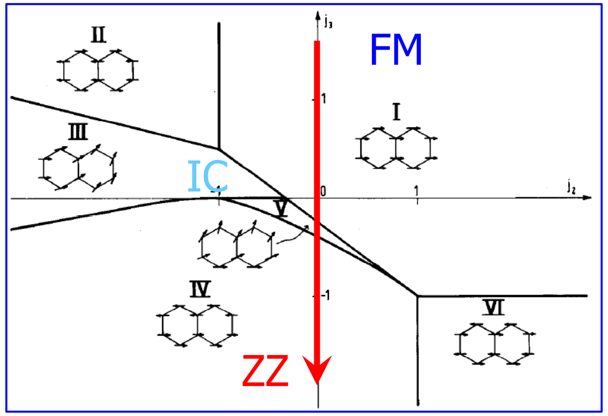


Liu, Chaloupka, and Khaliullin, PRB (2022)

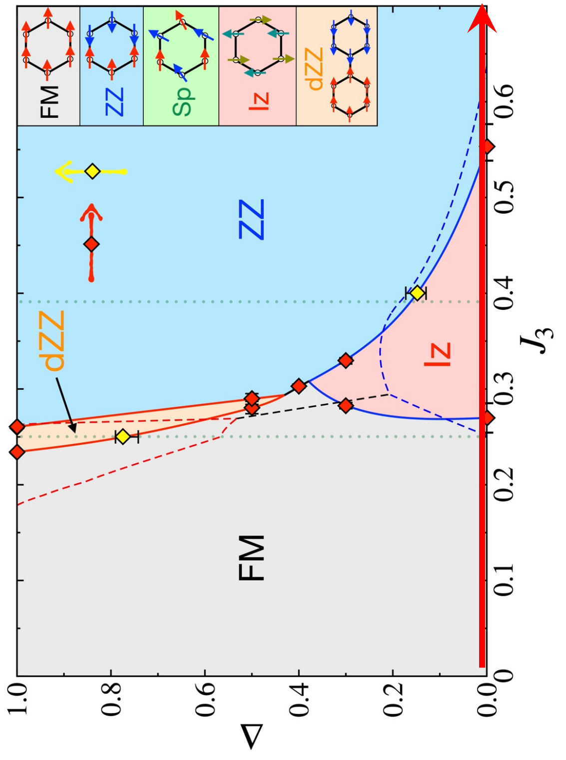
MM, PM, SJ, SW, RV, and **SC**, (unpublished).

J_1-$J_{z\pm}$-J_3 (Δ_1=0) model, phase diagram?

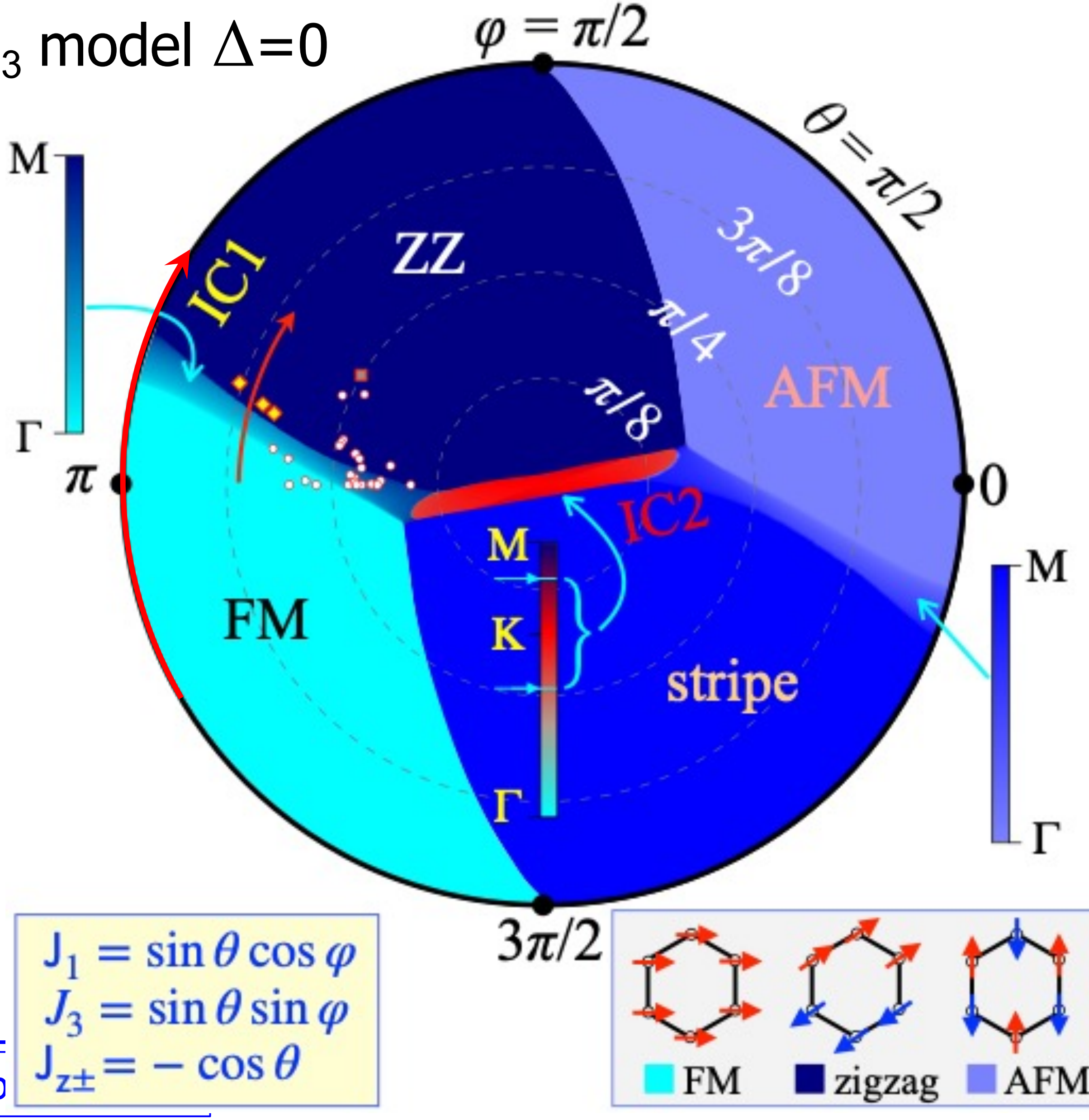
○ α-$RuCl_3$ ⇛ is a J_1-J_3 FM-AFM **enriched** by a strong $J_{z\pm}$-term



Rastelli *etal*, Physica 97B (1979).



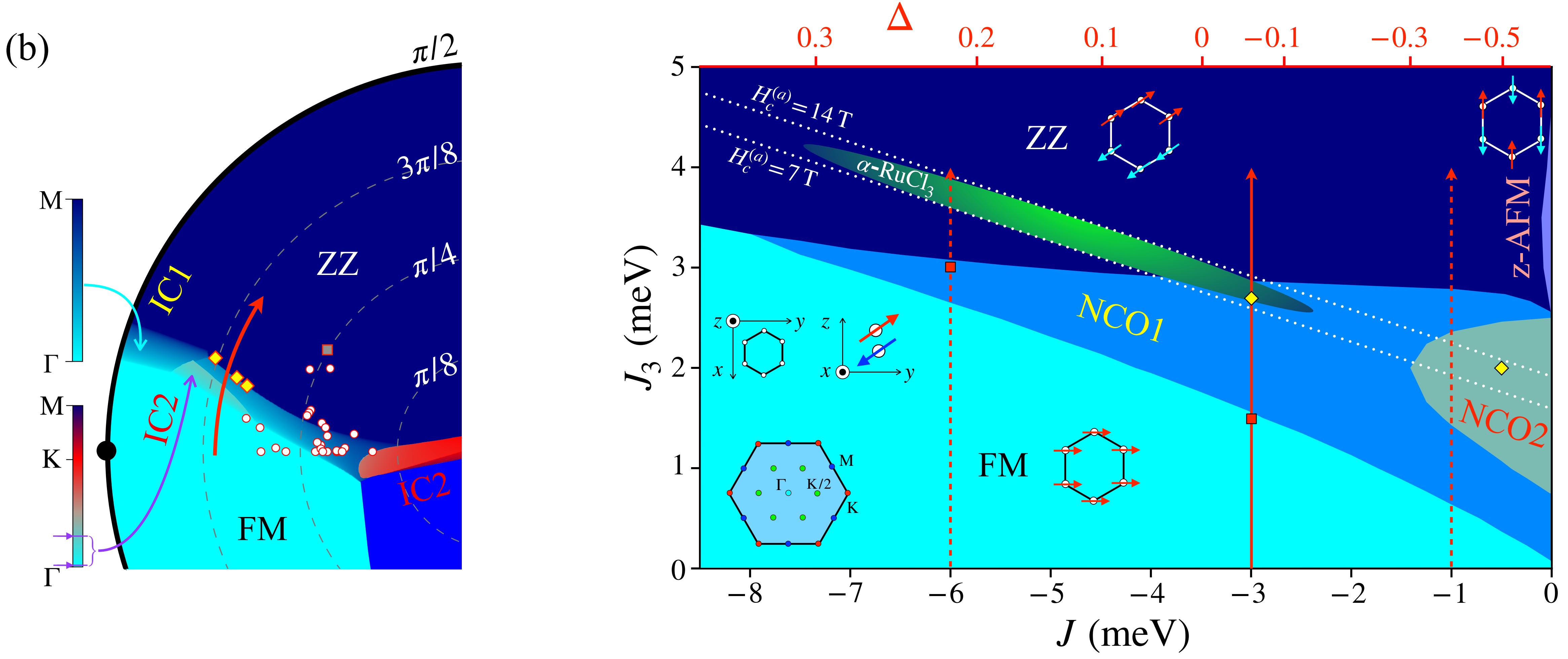
J_1-$J_{z\pm}$-J_3 model Δ=0



Jiang *etal*, PRB (2023).

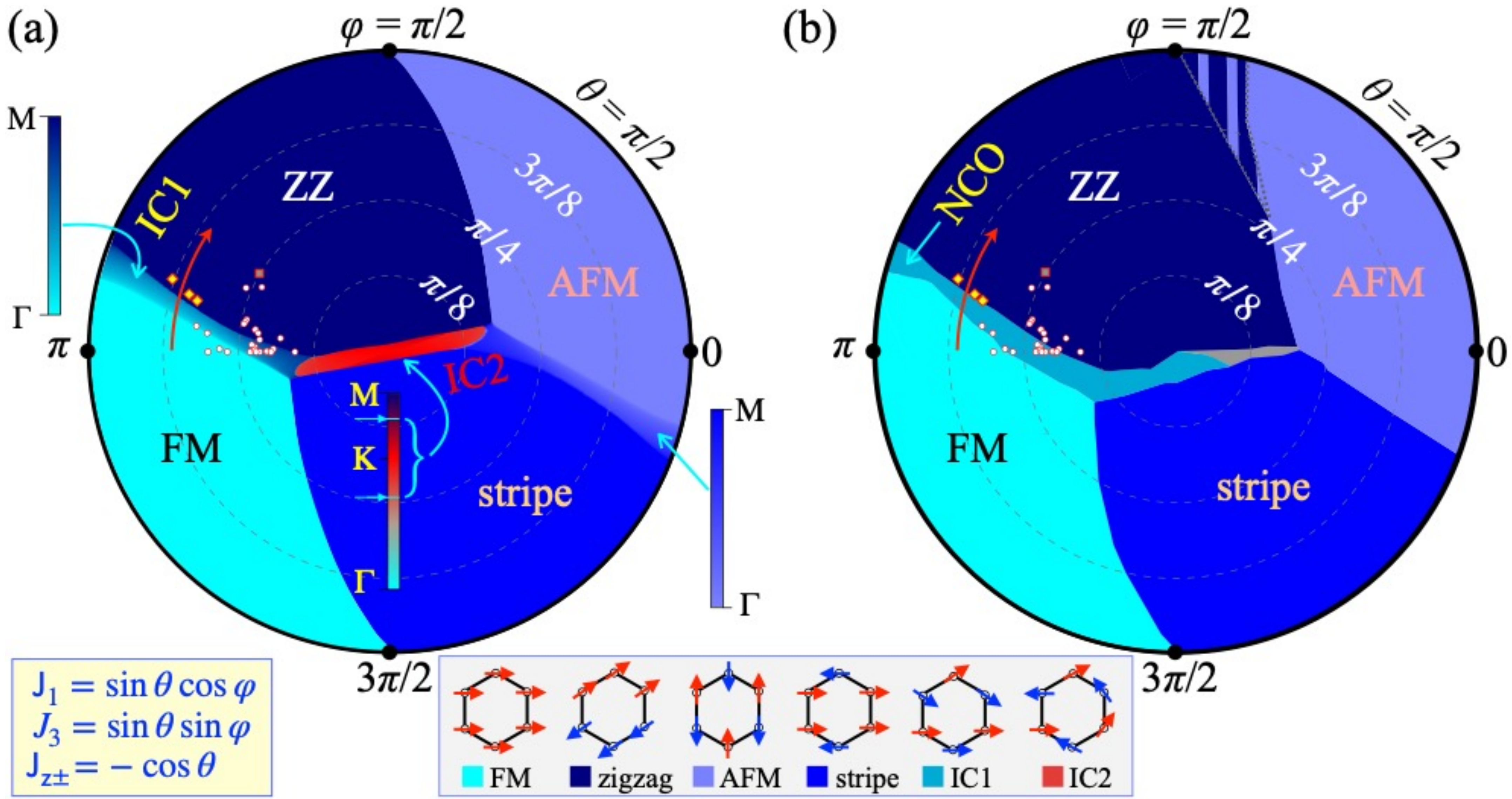
MM, PM, SJ, SW, RV, and **SC**, (unp

where are we? ZZ near IC, next to FM phase



MM, PM, SJ, SW, RV, and **SC**, (unpublished).

LT vs ED

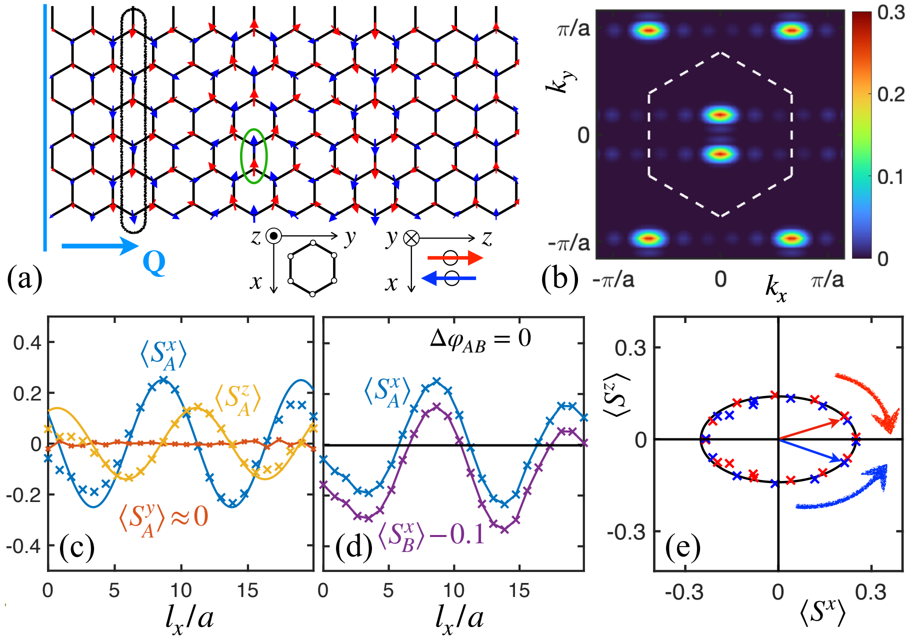


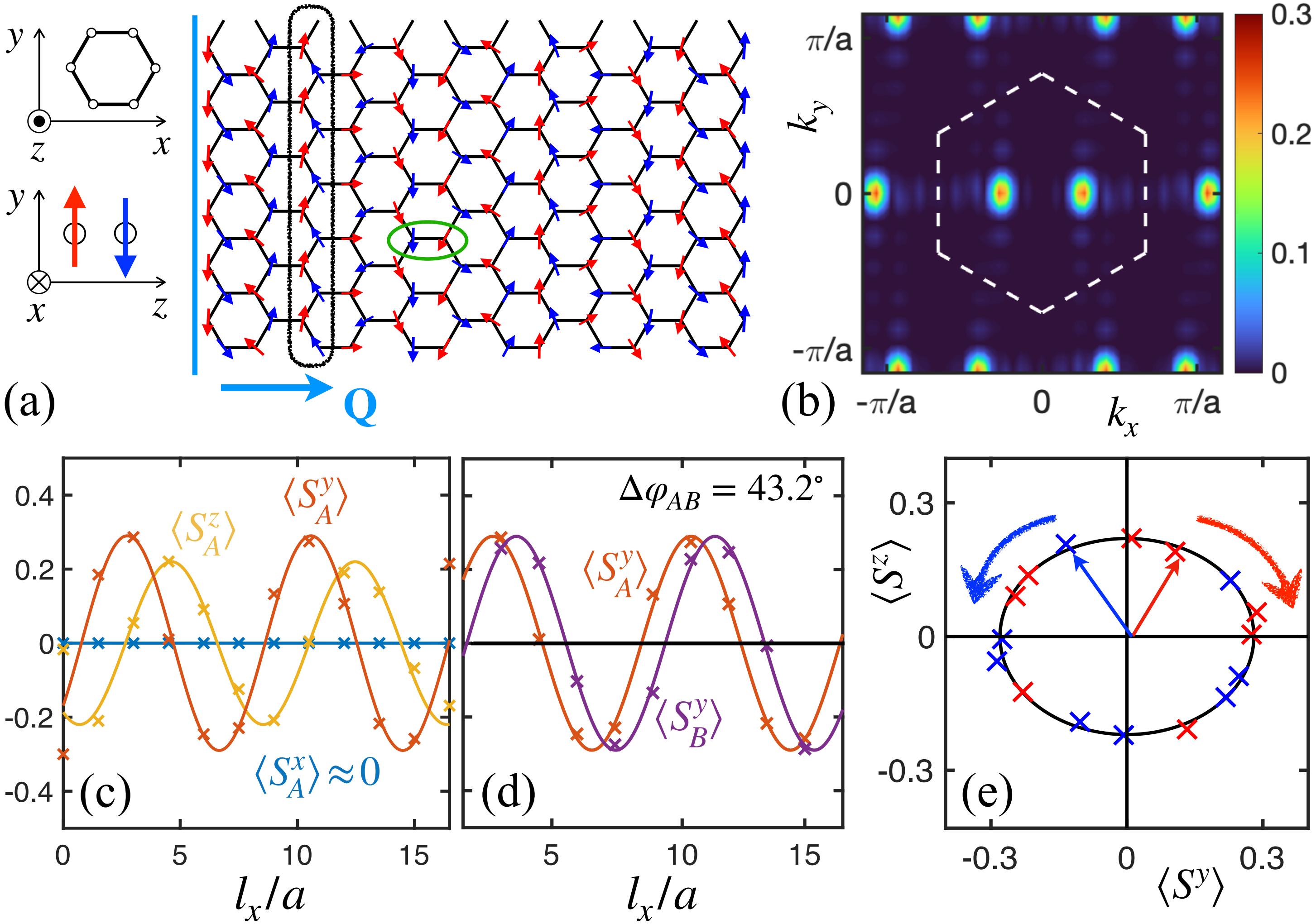
$J_1 = \sin\theta\cos\varphi$
$J_3 = \sin\theta\sin\varphi$
$J_{z\pm} = -\cos\theta$

MM, PM, SJ, SW, RV, and **SC**, (unpublished).

IC phases? DMRG: counter-rotating spirals!

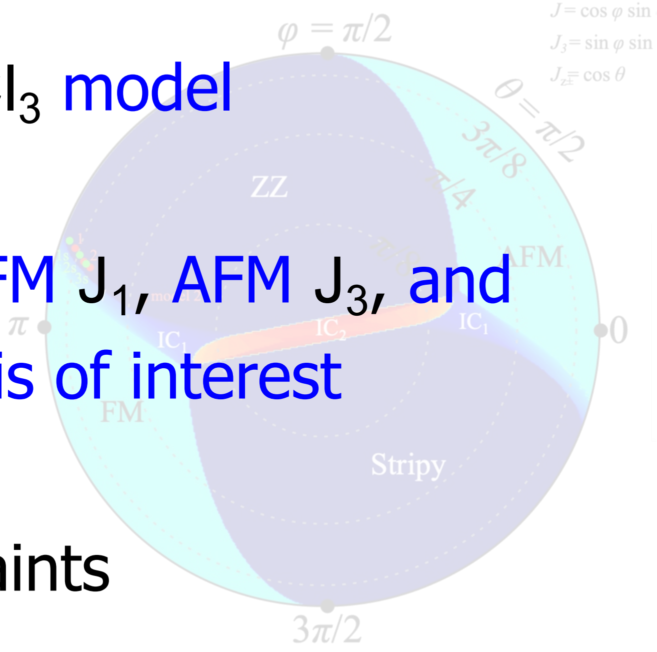
- ☑ two types of IC phases ⇛ orientations of the ordering vector, phase shift, deformation
- ☑ in a remarkable agreement with LT





MM, PM, SJ, SW, RV, and **SC**, (unpublished).

summary



- ☑ there is a better, more intuitive parametrization of the α-$RuCl_3$ model

- ☑ α-$RuCl_3$ is a **ferro-antiferromagnet** with an **easy-plane** FM J_1, AFM J_3, and large anisotropic $J_{z\pm}$. Proximity of the ZZ phase to IC phase is of interest

- ☑ parameters yield adequate phenomenology ⇛ strong constraints

- ☑ ICs = counter-rotating helices

P. A. Maksimov and **SC**, PRR **2**, 033011 (2020).

MM, PM, SJ, SW, RV, and **SC**, (unpublished).