$\mathfrak{Im}[\alpha$-Ru

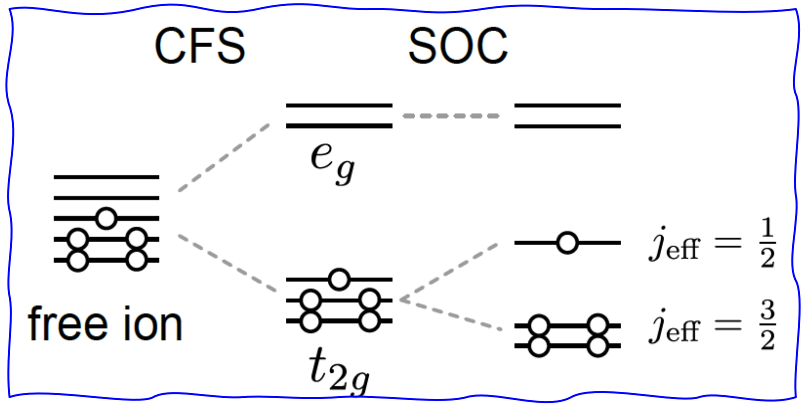
$\mathfrak{Re}[\alpha\text{-RuCl}_3]$

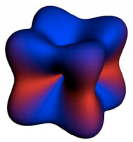
Pavel Maksimov

P. A. Maksimov and **SC**, "**Re**thinking α-$RuCl_3$", PR**Re**search **2**, 033011 (2020).

α-$RuCl_3$ essentials (and not)

Ru^{3+} [#44, year 1844, Kazan]
Carl Ernst Claus, Ruthenia;
$RuCl_3$ = 1845 (!);
(*) ESR, Kazan, 1944

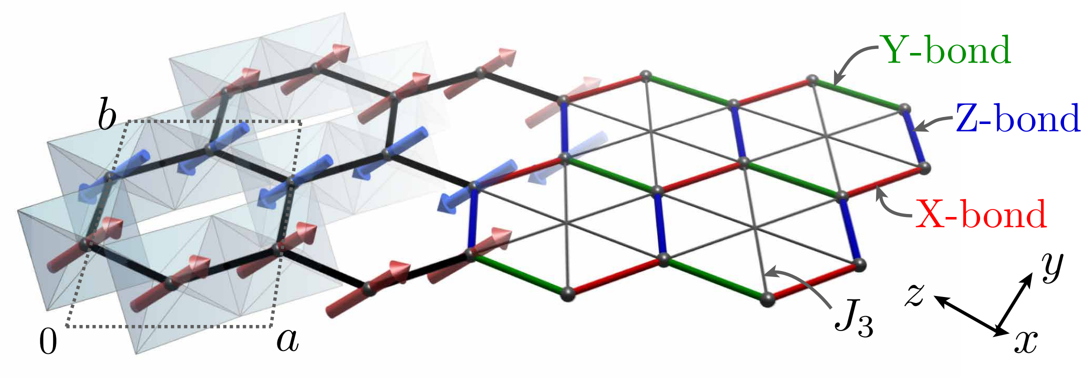


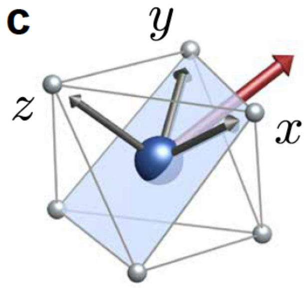


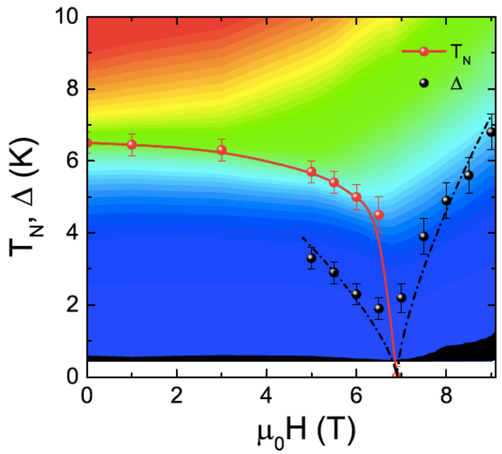
effective S=1/2

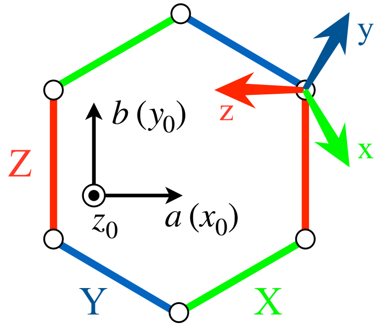
- honeycomb lattice
- octahedral* environment, Ru^{3+}, $\boldsymbol{J_{eff}}$=1/2
- zigzag order, tilted out of basal plane, $T_N \approx$ 7K
- in-plane* critical fields $H_{c,a} \approx H_{c,b} \approx$ 6-7 T

- (*) so-called **cubic axes** are used (Kitaev-explicit)









S. M. Winter *etal.*, JPCM **29**, 493002 (2017).

minimal **effective** model

approach:

- include all **symmetry-allowed** nearest-neighbor, add *minimal* non-NN terms
- use **phenomenology**

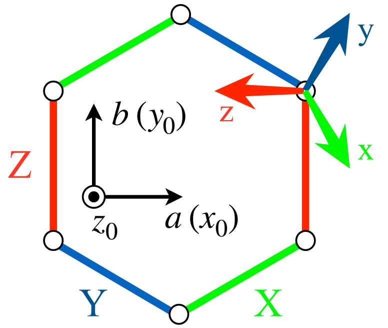
$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_1 + \hat{\mathcal{H}}_3 = \sum_{\langle ij \rangle} \mathbf{S}_i^{\mathrm{T}} \hat{\boldsymbol{J}}_{ij} \mathbf{S}_j + J_3 \sum_{\langle ij \rangle_3} \mathbf{S}_i \cdot \mathbf{S}_j$$

["generalized Kitaev" or J-K-Γ-Γ′-J_3 model,
≈ consensus]

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

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

- lattice symmetries* ⇛ **four** terms (+J_3) ⇛ **5D space**
- cubic axis parametrization of $\boldsymbol{J}_{ij}$ (exchange matrix)



this approach:

- ≠ but not ⊥ to DFT [DFT: many more terms, truncate, correlations?]
- ≠ but not ⊥ to downfoldings [superexchange expansions: perturbative]
- parameters of the effective model ≠ DFT parameters [effective resummation]

DF

guidance from DFT/dow

- overall values*
- hierarchy of terms*

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

$$\mathcal{H}_1 = \sum_{\langle ij \rangle_\gamma} \left\{ J \mathbf{S}_i \cdot \mathbf{S}_j + K \right.$$

prior work: combination of various approaches **and** phenomenologies

can do better:
need strong constraints

Reference	Method	K	Γ	Γ'	J	J_3
Banerjee et al. [22]	LSWT, INS fit	+7.0			-4.6	
Kim et al. [29]	DFT+t/U, $P3$	**-6.55**	**5.25**	-0.95	-1.53	
	DFT+SOC+t/U	**-8.21**	**4.16**	-0.93	-0.97	
	same+fixed lattice	-3.55	7.08	-0.54	**-2.76**	
	same+U+zigzag	+4.6	6.42	-0.04	**-3.5**	
Winter et al. [30]	DFT+ED, $C2$	**-6.67**	6.6	-0.87	-1.67	**2.8**
	same, $P3$	+7.6	8.4	+0.2	-5.5	**2.3**
Yadav et al. [24]	Quantum chemistry	**-5.6**	-0.87		+1.2	
Ran et al. [34]	LSWT, INS fit	**-6.8**	9.5			
Hou et al. [31]	DFT+t/U, $U=2.5$eV	-14.43	6.43		**-2.23**	**2.07**
	same, $U=3.0$eV	-12.23	**4.83**		**-1.93**	1.6
	same, $U=3.5$eV	**-10.67**	**3.8**		-1.73	1.27
Wang et al. [32]	DFT+t/U, $P3$	**-10.9**	6.1		-0.3	0.03
	same, $C2$	**-5.5**	7.6		+0.1	0.1
Winter et al. [35]	*Ab initio*+INS fit	**-5.0**	2.5		-0.5	0.5
Suzuki et al. [36]	ED, C_p fit	-24.41	**5.25**	-0.95	-1.53	
Cookmeyer et al. [37]	thermal Hall fit	**-5.0**	2.5		-0.5	0.11
Wu et al. [38]	LSWT, THz fit	-2.8	2.4		-0.35	0.34
Ozel et al. [39]	same, $K>0$	+1.15	2.92	+1.27	-0.95	
	same, $K<0$	-3.5	2.35		+0.46	
Eichstaedt et al. [33]	DFT+Wannier+t/U	-14.3	9.8	-2.23	-1.4	0.97
Sahasrabudhe et al. [42]	ED, Raman fit	**-10.0**	3.75		-0.75	0.75
Sears et al. [40]	Magnetization fit	**-10.0**	10.6	-0.9	**-2.7**	
Laurell et al. [41]	ED, C_p fit	-15.1	10.1	-0.12	-1.3	0.9

plan

I. “strong” constrains on the parameter space

II. consequences: better model(s)

III. more consequences: common features

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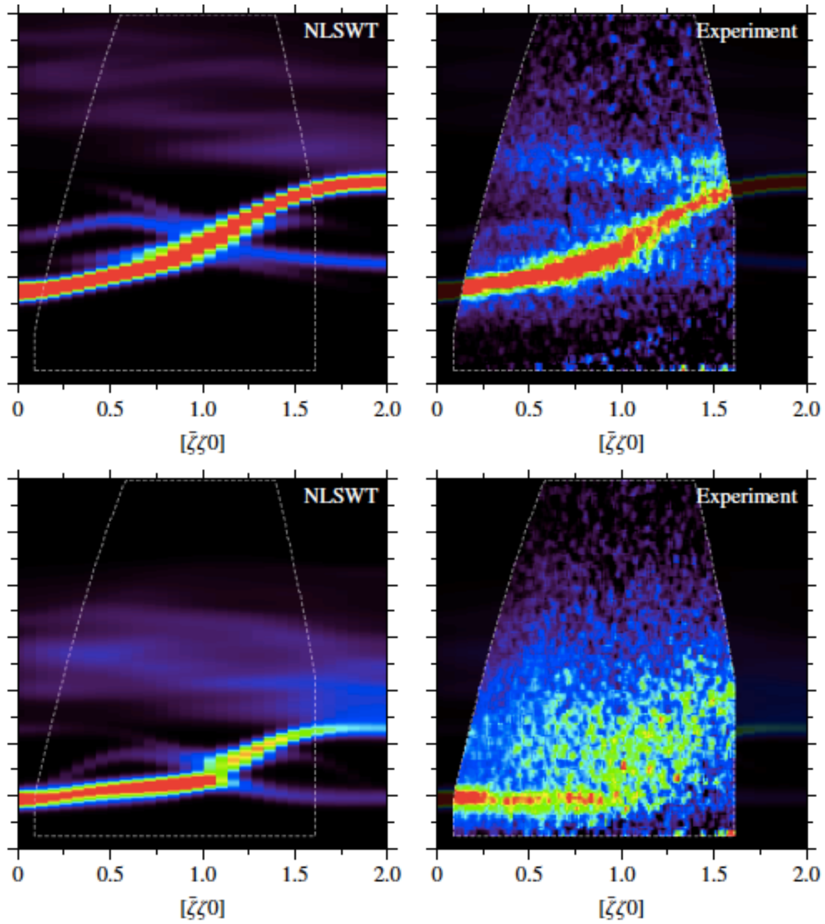
ideal world … [Radu Coldea version]

$Yb_2Ti_2O_7$, NLSWT
J. Rau *et al.*, PRB **100**, 033011 (2019).



- strong polarizing field
  ⇛ fit spin-flip dispersion in the “FM” state
  ⇛ **parameters**

- α-$RuCl_3$ ⇛ difficulties:
  -- fields somewhat too high
  -- neutron experiments limited
  -- fluctuations above H_c’s
  -- still, high-field regime is profoundly instructive

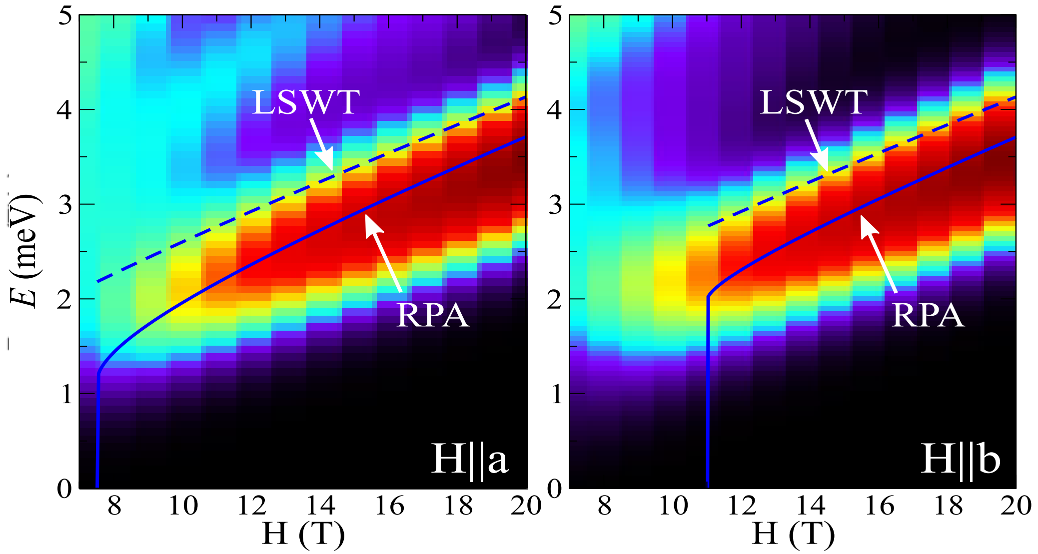
K. A. Ross *et al.*, 2011; J. D. Thompson *et al.*, 2017.

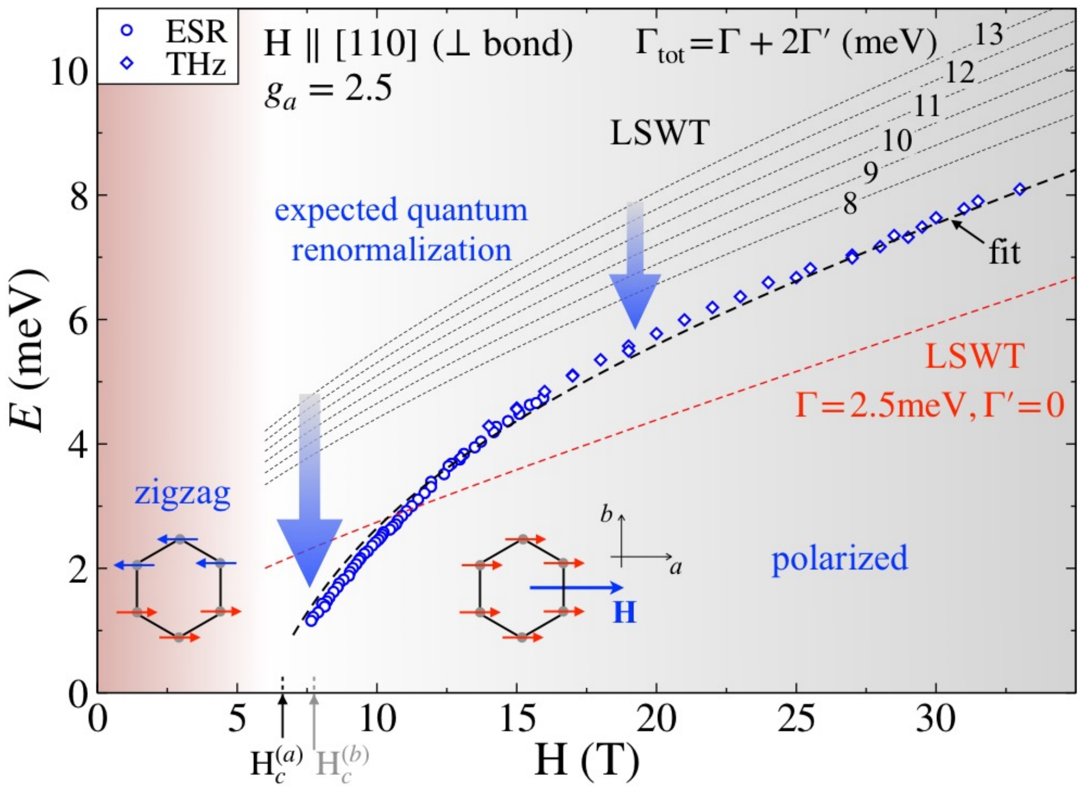
#1: ESR, THz, Raman (high field)

- strong in-plane field, probe **k=0** spin-flip excitation
- LSWT?
  -- $E_{\mathbf{k}=0}$ depends **only** on Γ and Γ′ [via $\Gamma_{tot}=\Gamma+2\Gamma'$]

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

  -- fluctuations renormalize $E_{\mathbf{k}=0}$ down [ED]
  -- **most** prior parameter choices [table] fail
- $\Gamma_{tot}=\Gamma+2\Gamma'$ **must be** at least 8 meV





- ⇛ **strong bounds on**
  $\Gamma_{tot} = \Gamma+2\Gamma' \gtrsim 8$ meV
  $\Gamma_{tot} = \Gamma+2\Gamma' \lesssim 13$ meV

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

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

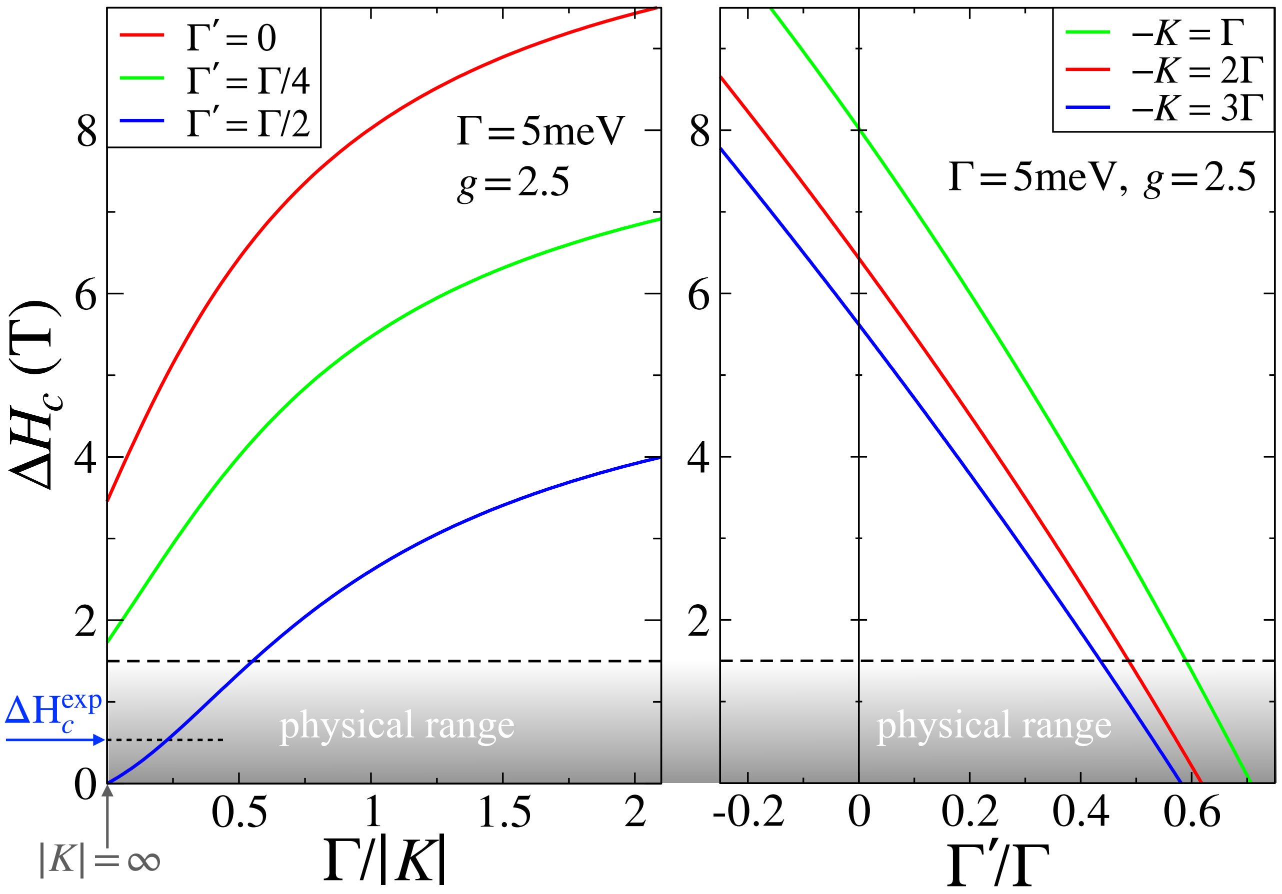
#2: [exp.] in-plane critical fields $H_{c,a} \approx H_{c,b}$

- $H_{c,a} \approx H_{c,b}$, what's a big deal? (if $g_a \approx g_b$)
- **not true** due to anisotropic exchanges: $H_{c,a} \neq H_{c,b}$ even if $g_a = g_b$

- LSWT? (1/S, small or is not affecting ΔH_c)
  -- ΔH_c depends **only** on K, Γ, and Γ′
- -- small ΔH_c is **impossible** to reconcile without Γ′ > 0 [!] and ≳ Γ/2
  -- **none** of the prior works predicted that

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

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



⇛ **Γ′ ≳ Γ/2**
⇛ **helps with large** Γ_{tot} = Γ+2Γ′ from the ESR

P. Lampen-Kelley *et al.*, 2018.

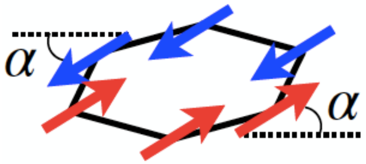
#3: critical fields $H_{c,a}$, $H_{c,b}$, #4: tilt angle α

- $H_{c,a}$ and $H_{c,b}$, by far the **strongest** dependence is on a combination of J and J_3: $J+3J_3$, ⇛ fixing $H_{c,a(b)}$ fixes $J+3J_3$

- #3 = strong constraint
- #4 = not too strong [exp]
- #5 = "soft" constraint, total spectral bandwidth W_0 helps with overall scale

+ checks for ZZ GS in H=0

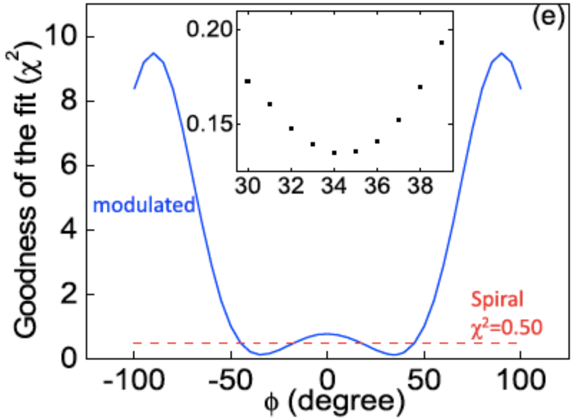
- (LSWT) out-of-plane tilt angle **α** also depends **only** on K, Γ, and Γ′
- -- experimentally, α ≈ 35°, ED suggest modest quantum corrections

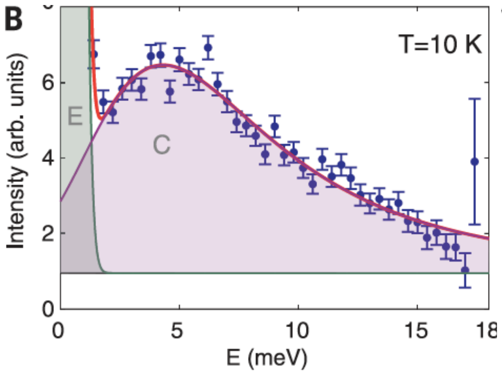


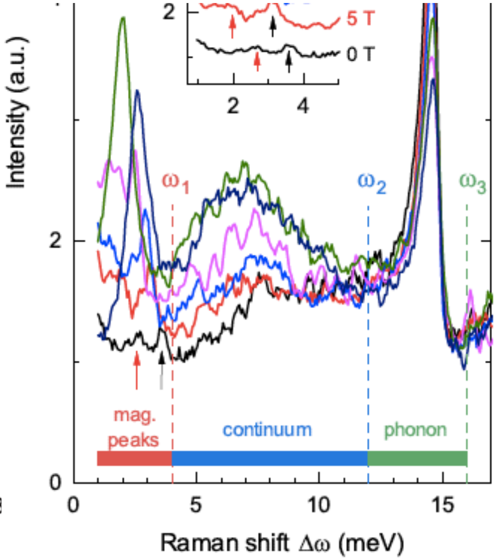
$$\tan 2\alpha = 4\sqrt{2}\,\frac{1+r}{7r-2}, \quad r = -\frac{\Gamma}{K+\Gamma'}$$

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

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

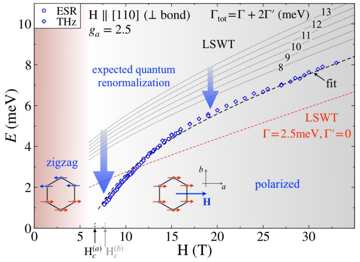


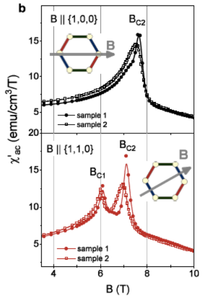


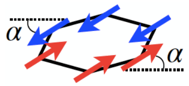


J. Chaloupka and G. Khaliullin, 2016. | H. B. Cao *etal.*, 2016. | A. Banerjee *etal.*, 2017, 2018; A. Sahasrabudhe *etal.*, 2019.

a taste of it … : "rigid constraints"





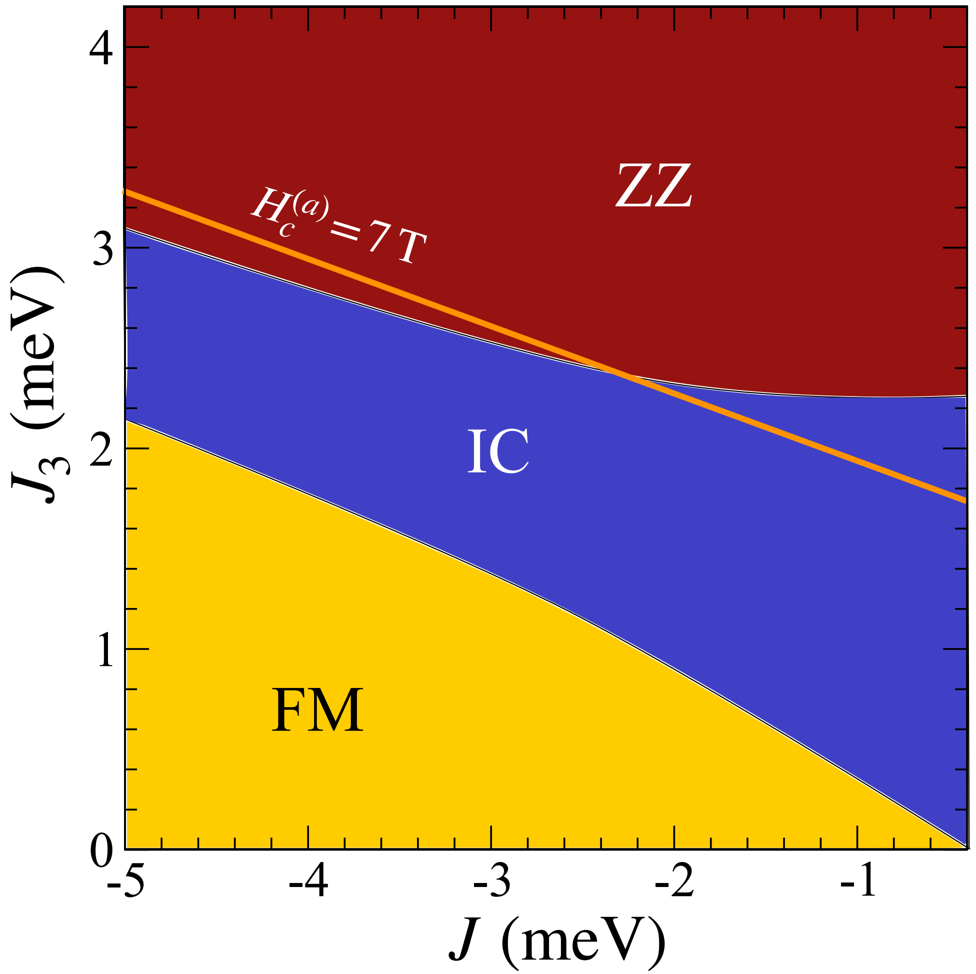


$$\{K, \Gamma, \Gamma'\} = \{-7.567, 4.276, 2.362\}\ \text{meV}$$

- **fix** [rigid constraints approach]:
- $\Gamma + 2\Gamma' = 9$ meV
- $\Delta H_c = 1$ T
- $\alpha = 35^\circ$
- $H_{c,a} = 7$ T

  $\Rrightarrow$ $J_{03} = J + 3J_3 = 4.768\ \text{meV}$

- 4 out of 5 parameters are fixed



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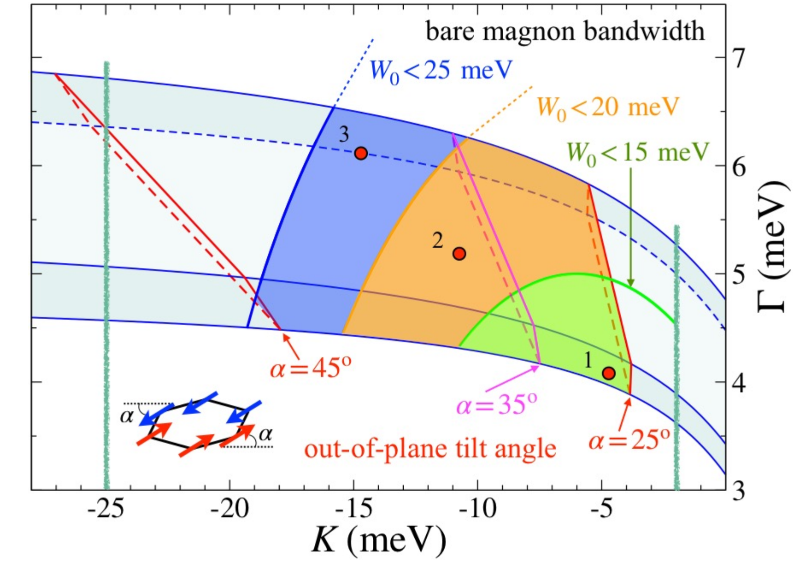
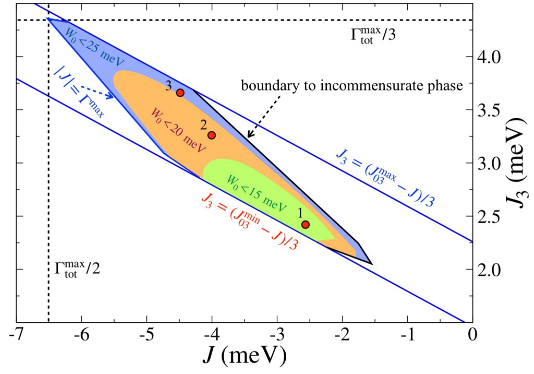
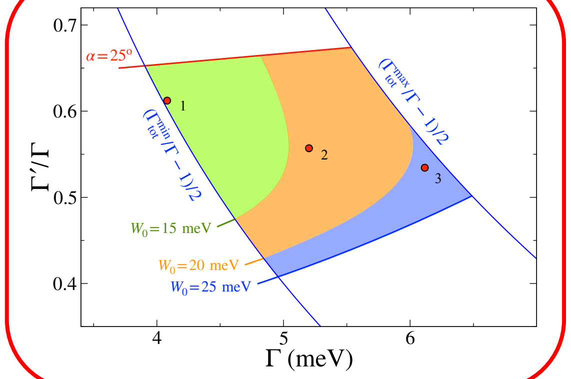
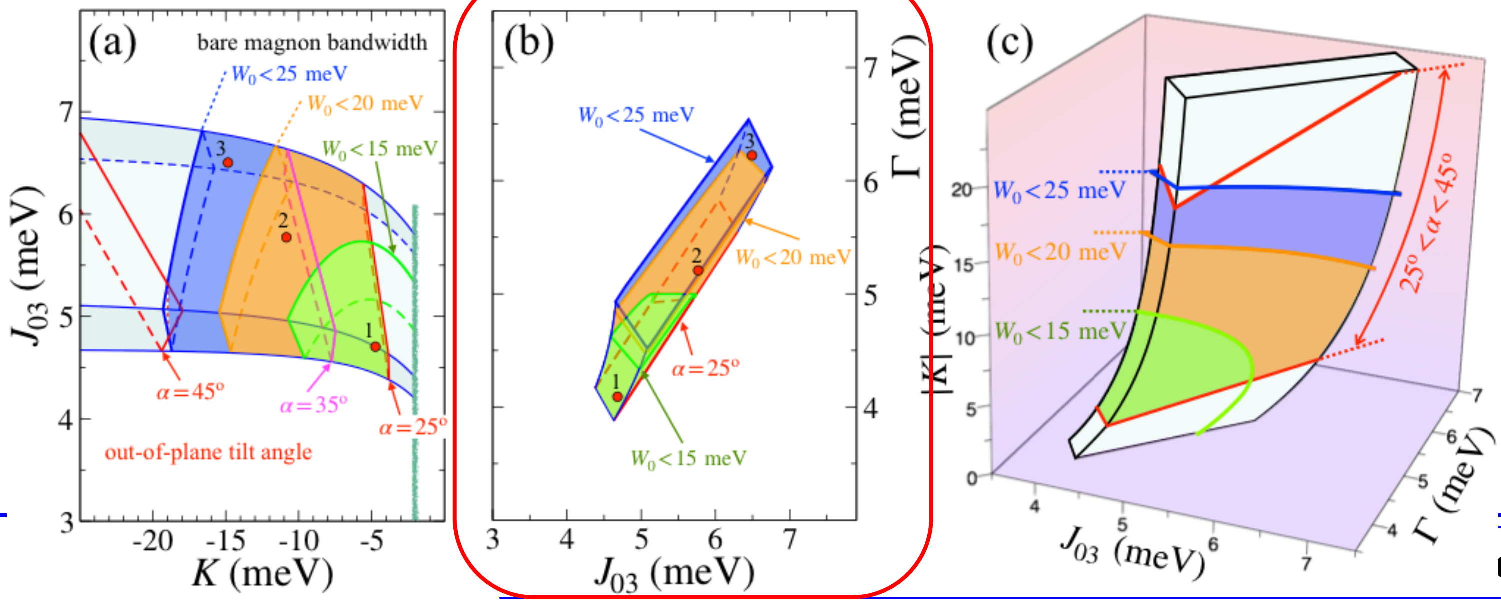
2D sections of 5D, varying constraints



- $\Gamma+2\Gamma' = 9–13$ meV
- $\Delta H_c = 0.5–1.5$ T
- $\alpha = 25^\circ – 45^\circ$
- $H_{c,a} = 7$ T
- spectral bandwidth W_0
  $W_0 < 15$ meV: "realistic"
  $W_0 < 20$ meV: "generous"
  $W_0 < 25$ meV: "outrageous"
- strongest bounds are on Γ, Γ′, and J_{03}





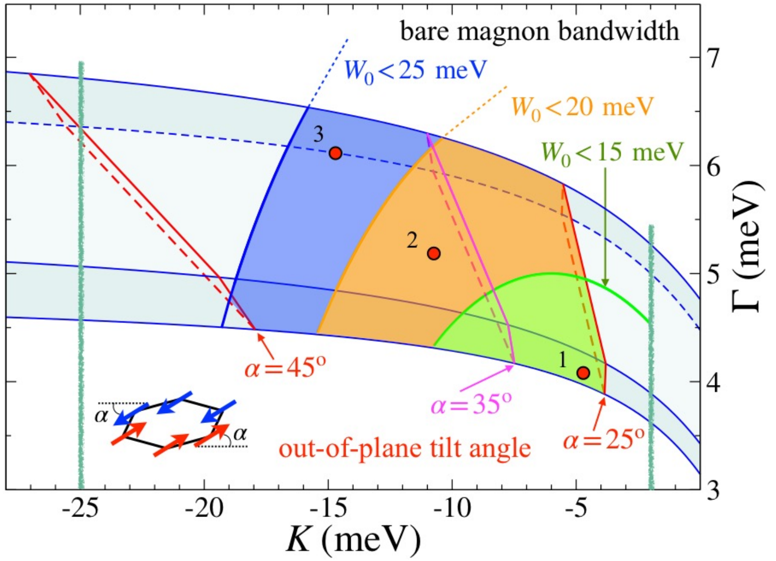


where are we?

- representative points from realistic, generous, and outrageous regions
- agree with some/most DFT guidance
- Γ' is the most significant difference
- **tight parameter space**, interrelated
- ranges do not do full justice

- **$K < 0$**, leading term ☑
- **$0 < \Gamma \lesssim |K|$,** ☑
- **$J < 0$**, subleading, ≈ ☑ but larger
- **$0 < J_3 \lesssim |J|$,** ≈ ☒ **$J_3 \approx |J|$**
- **$0 > \Gamma' \sim 0$,** ☒☒ **$\Gamma' \gtrsim \Gamma/2$**

	$(K,$	$\Gamma,$	$\Gamma',$	$J,$	$J_3)$	$\{\Gamma_{\text{tot}},$	$J_{03}\}$
Point 1:	(-4.8,	4.08,	2.5,	-2.56,	2.42)	{9.08,	4.70}
Point 2:	(-10.8,	5.2,	2.9,	-4.0,	3.26)	{11.0,	5.78}
Point 3:	(-14.8,	6.12,	3.28,	-4.48,	3.66)	{12.7,	6.50}
"realistic" range	**[-11,-3.8]**	**[3.9,5.0]**	**[2.2,3.1]**	**[-4.1,-2.1]**	**[2.3,3.1]**	**[9.0,11.4]**	**[4.4,5.7]**



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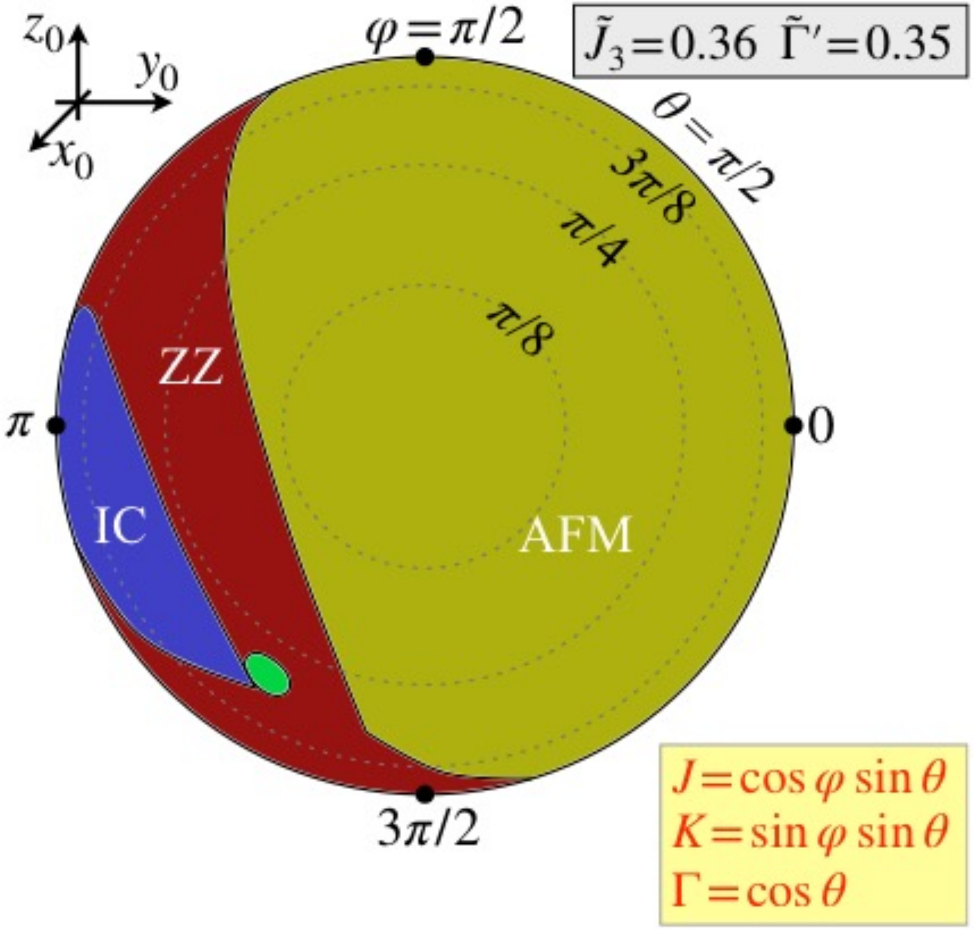
insights?: 2D cut of a 4D space

- fix Γ', and J_3 [5D ⇛ 3D]
- introduce global scale $\sqrt{J^2+K^2+\Gamma^2}$ ⇛ 2D
- α-$RuCl_3$ ⇛ ZZ in a proximity of IC phase

	$(K,$	$\Gamma,$	$\Gamma',$	$J,$	$J_3)$	$\{\Gamma_{\rm tot},\ J_{03}\}$
Point 1:	(-4.8,	4.08,	2.5,	-2.56,	2.42)	{9.08, 4.70}
Point 2:	(-10.8,	5.2,	2.9,	-4.0,	3.26)	{11.0, 5.78}
Point 3:	(-14.8,	6.12,	3.28,	-4.48,	3.66)	{12.7, 6.50}



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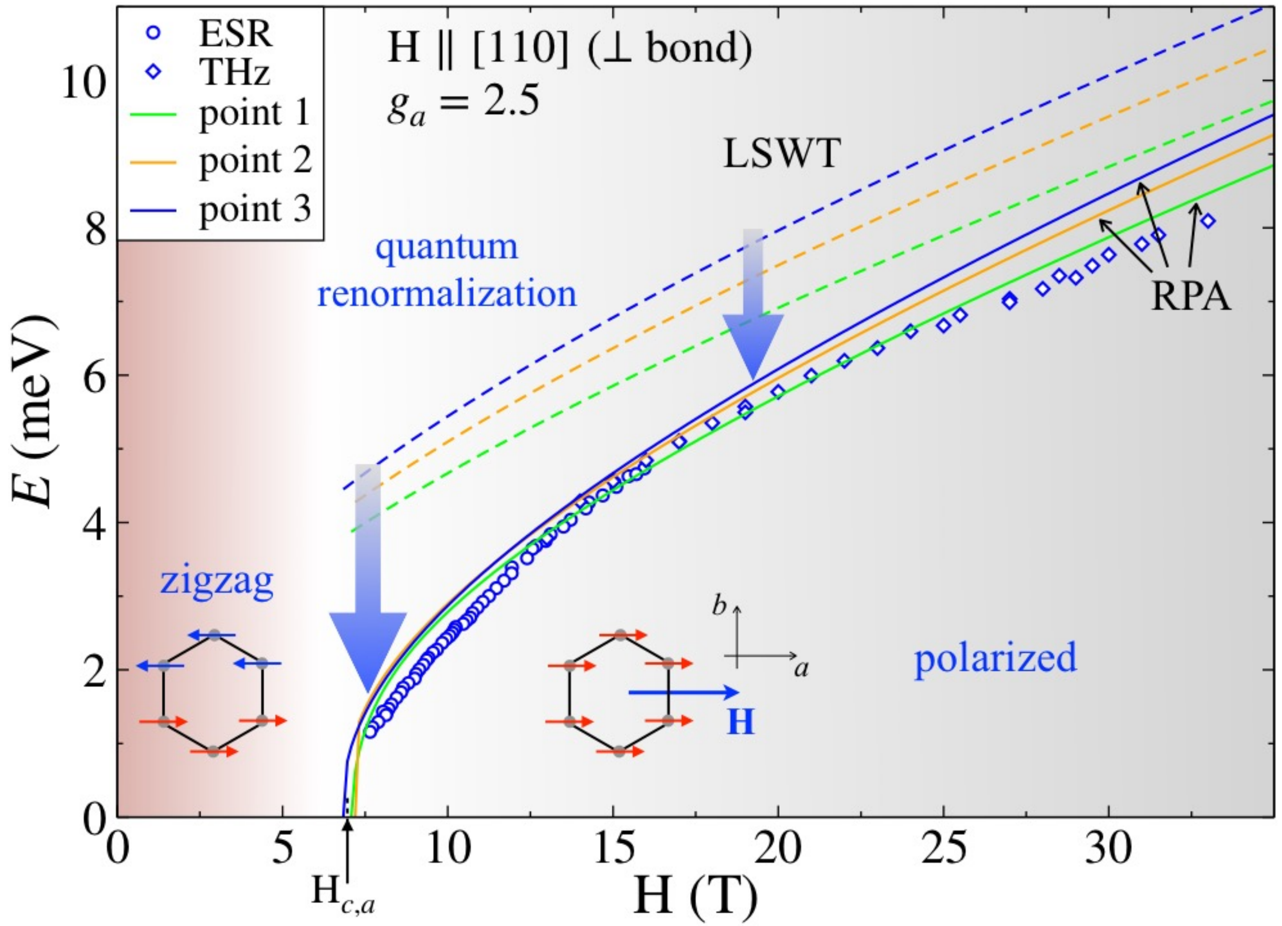
self-consistency, RPA

- proximity to the IC phase ⇛
  strong fluctuations:
  ordered moment in H=0, ⟨S⟩ = 0.22
- [in agreement with exp.]

	$\langle S \rangle$
Point 1	0.219
Point 2	0.220
Point 3	0.225



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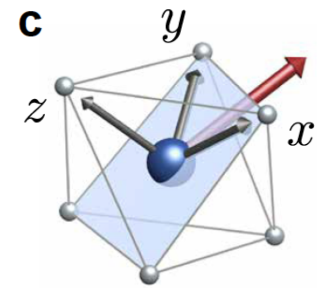
we are not in *K*ansas anymore …



- and, arguably, never been …
  ⇛ why do we need ***k***ubic axes? scaffolding that was never ideal…
- ⇛ use "**natural**" axes instead (honeycomb plane)

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

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

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

XXZ

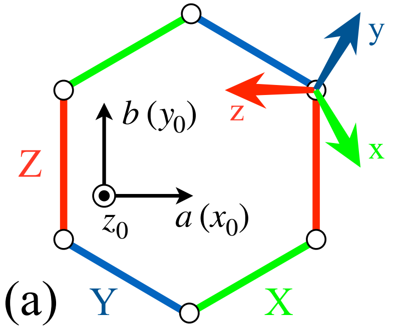
$$\tilde{\varphi}_\alpha = \{0, 2\pi/3, -2\pi/3\}$$

bond-dependent

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

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

-- ["ice-like"]



bond orientation is coupled to spin orientation

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parameter conversion and better model

$$J_1 = J + \frac{1}{3}(K - \Gamma - 2\Gamma'),$$

$$\Delta J_1 = J + \frac{1}{3}(K + 2\Gamma + 4\Gamma'),$$

$$2J_{\pm\pm} = -\frac{1}{3}(K + 2\Gamma - 2\Gamma'),$$

$$\sqrt{2}J_{z\pm} = \frac{2}{3}(K - \Gamma + \Gamma').$$

	$(J_1,$	$\Delta,$	$J_{\pm\pm},$	$J_{z\pm},$	$J_3)$
"ice" Point 1:	(-7.20,	-0.26,	0.3,	-3.0,	2.42)
"ice" Point 2:	(-11.3,	0.02,	1.0,	-6.2,	3.26)
"ice" Point 3:	(-13.6,	0.07,	1.5,	-8.3,	3.66)

- conversion table:
  $K, J < 0$, $\Gamma, \Gamma' > 0$
- J_1 ⇛ all add up, Δ ⇛ cancel out
- $J_{z\pm}$ ⇛ partially add up
- $J_{\pm\pm}$ ⇛ partially cancel
- ⇛ J_1 is the largest, $J_{z\pm}$ ⇛ second largest

⇛ neglect Δ, $J_{\pm\pm}$ is similar to $J_{z\pm}$, keep $J_{z\pm}$ only

altogether:

- α-$RuCl_3$ parameters imply a much simpler
  J_1-$J_{z\pm}$-J_3 model
- easy-plane FM J_1, AFM $J_3/|J_1| = 0.3{-}0.4$, and large anisotropic $J_{z\pm}/|J_1| = 0.5$
- [$J_{z\pm}$ yields spins' out-of-plane tilt]

P. A. Maksimov and **SC**, "**Re**thinking α-$RuCl_3$", PR**Re**search **2**, 033011 (2020).

italians, or back to the future …

NON-SIMPLE MAGNETIC ORDER FOR SIMPLE HAMILTONIANS

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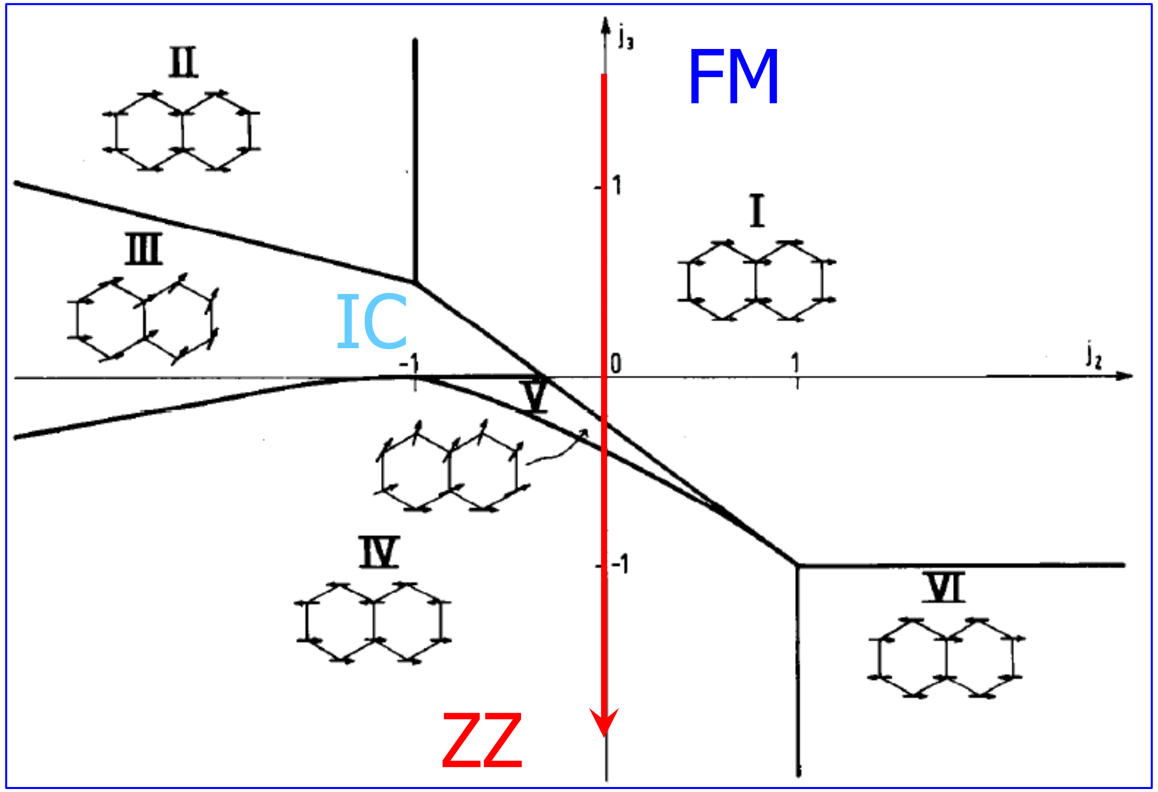
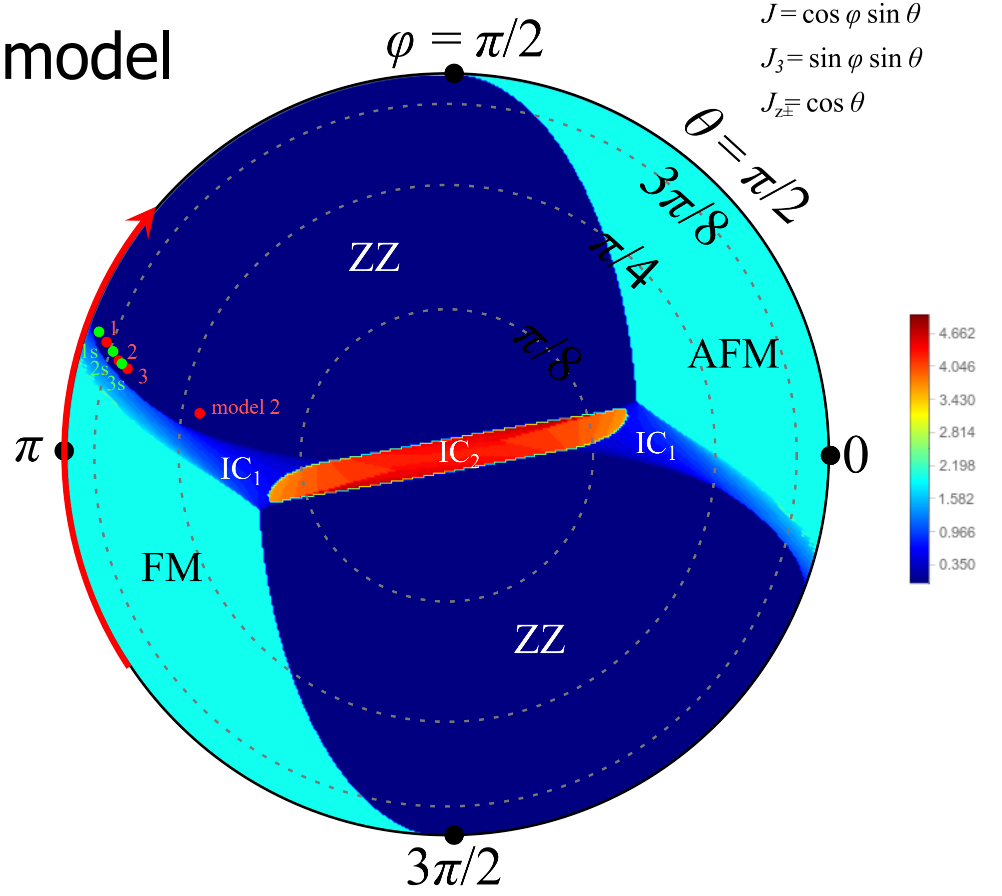
and
L. REATTO

Physica 97B (1979) 1-



J_1-$J_{z\pm}$-J_3 model

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

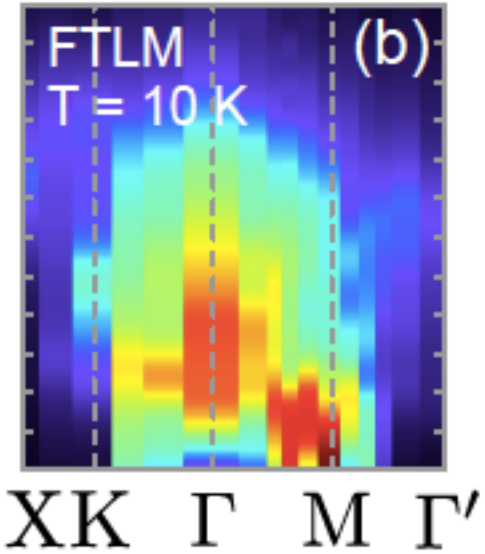


- α-$RuCl_3$ ⇏ is "just" a J_1-J_3 FM-AFM with a [strong] extra $J_{z\pm}$-term

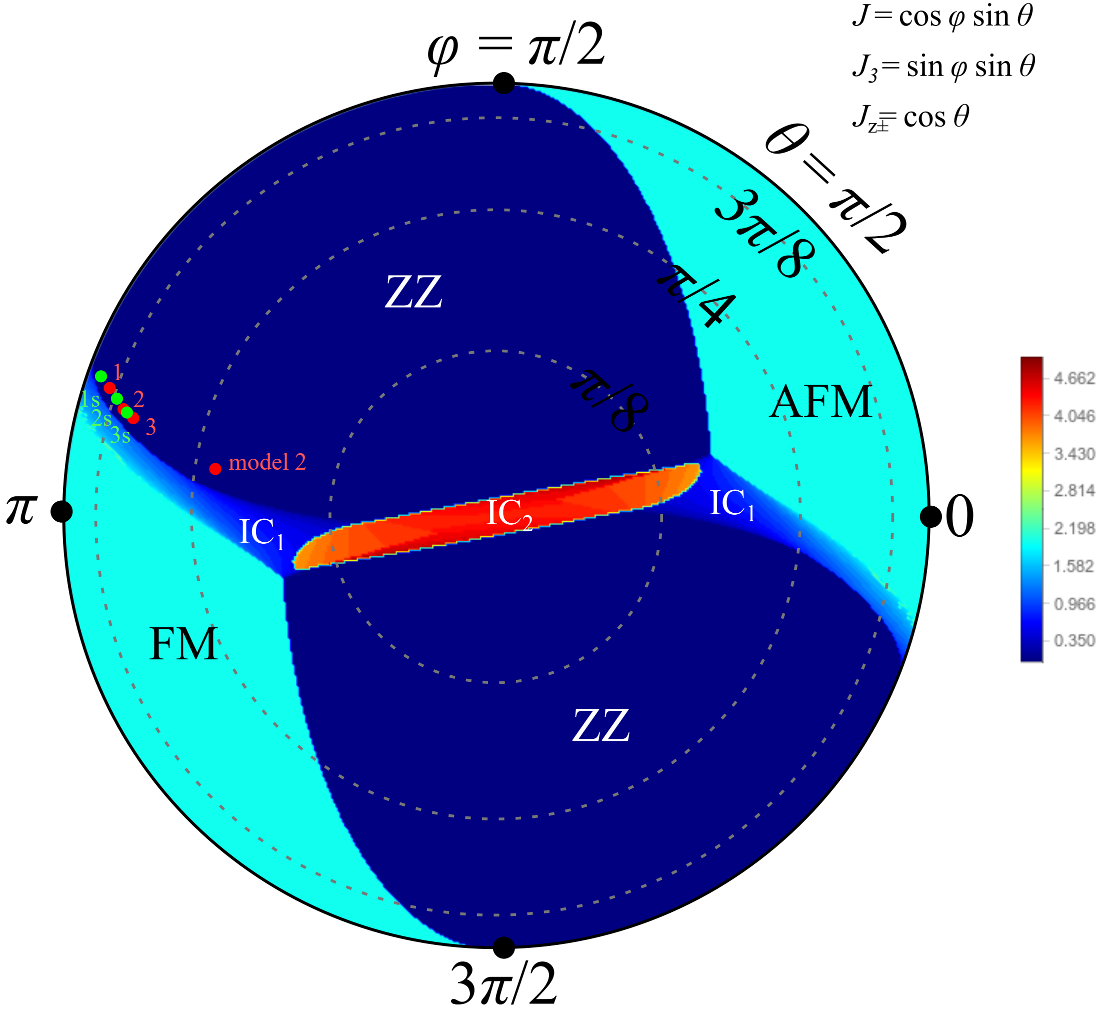
P. A. Maksimov and **SC**, "**Re**thinking α-$RuCl_3$", PR**Re**search **2**, 033011 (2020).

P. A. Maksimov, unpublished

ZZ proximity to IC that is near FM



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



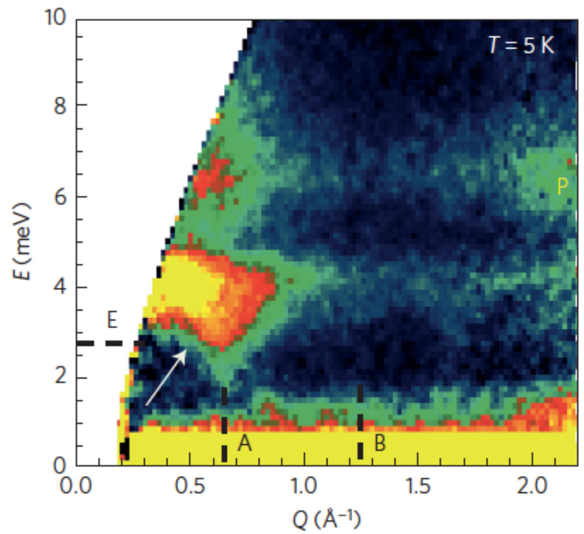
J_1-$J_{z\pm}$-J_3 model

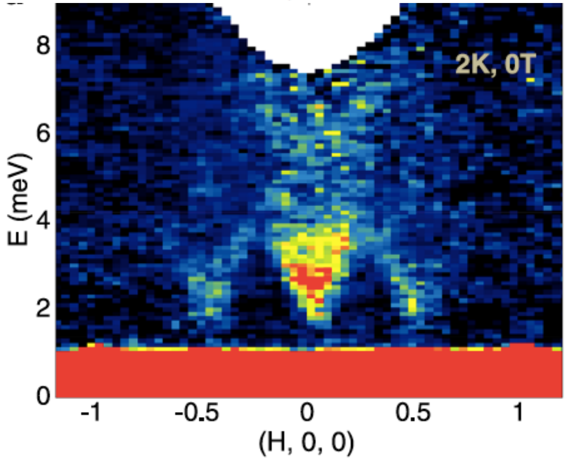
- refreshing perspective on α-$RuCl_3$
- similarity to J_1-J_3 is staggering
- offers a connection to a large body of work on J_1-J_2-J_3 models

P. A. Maksimov, unpublished

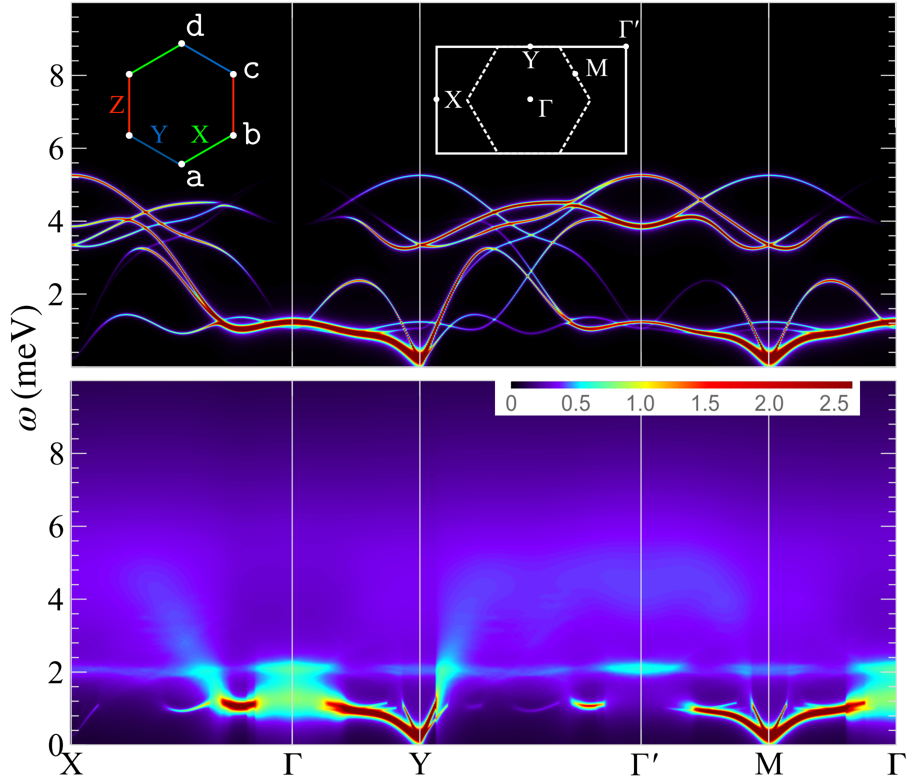
P. A. Maksimov and **SC**, "**Re**thinking α-$RuCl_3$", PR**Re**search **2**, 033011 (2020).

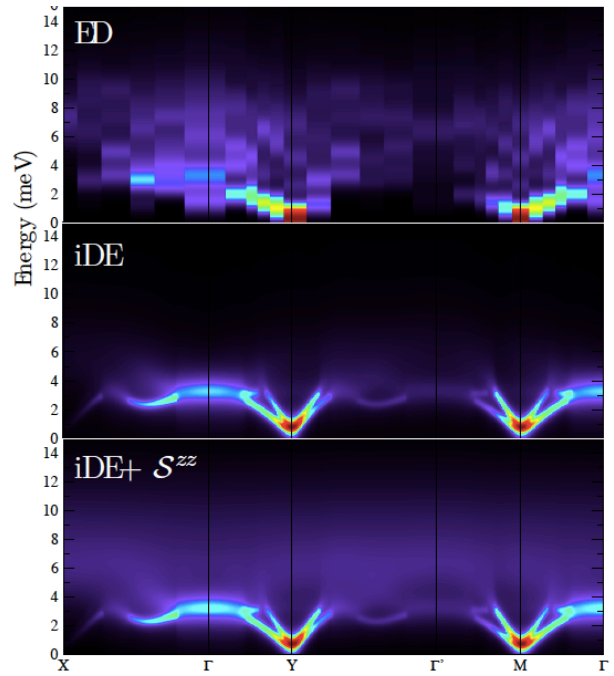
more consequences …





A. Banerjee *etal.*, 2016, 2018.





S. M. Winter *etal.*, 2017.

- S(**q**,ω), includes both strong decays and large continuum contribution
- decays for other model parameters, similarity with experiments

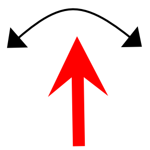
P. A. Maksimov and **SC**, "**Re**thinking α-$RuCl_3$", PR**Re**search **2**, 033011 (2020).

inevitable anharmonic terms

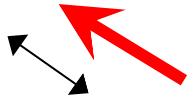
$$\widetilde{\boldsymbol{J}}_{ij} = \hat{\mathbf{R}}_i^{\mathrm{T}} \hat{\boldsymbol{J}}_{ij} \hat{\mathbf{R}}_j = \begin{pmatrix} \widetilde{J}_{ij}^{xx} & \widetilde{J}_{ij}^{xy} & \widetilde{J}_{ij}^{xz} \\ \widetilde{J}_{ij}^{yx} & \widetilde{J}_{ij}^{yy} & \widetilde{J}_{ij}^{yz} \\ \widetilde{J}_{ij}^{zx} & \widetilde{J}_{ij}^{zy} & \widetilde{J}_{ij}^{zz} \end{pmatrix}$$

$$\mathcal{H}_{\mathrm{od}} = \sum_{\langle ij \rangle} \left(\widetilde{J}_{ij}^{xz} \widetilde{S}_i^x \widetilde{S}_j^z + \widetilde{J}_{ij}^{yz} \widetilde{S}_i^y \widetilde{S}_j^z \right)$$

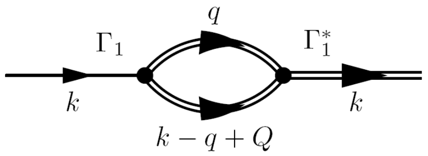
$$\mathcal{H}_3 = \sum_{\langle ij \rangle} \widetilde{V}_{ij} \left(a_i^\dagger a_j^\dagger a_j + \mathrm{H.c} \right)$$

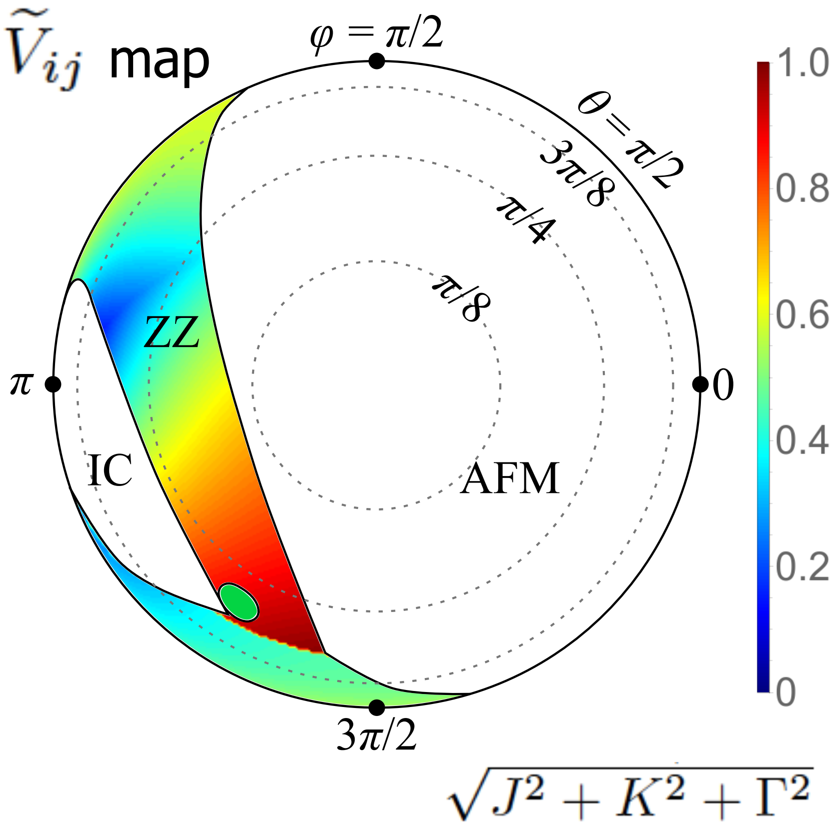


transverse
"one-magnon"



longitudinal
"two-magnon"





- general exchange matrix [due to SOC]
  ⇛ anisotropic exchanges couple **all** spin components
- inevitable magnon coupling ⇛ decays, continuum
- ⇛ key features:
  well-defined low-energy modes and broad continuum at higher energy
- ⇛ broad features in the spectrum **do not** require fine tuning of parameters
- the question is not "why?", but "why not?"

P. A. Maksimov and **SC**, "**Re**thinking α-$RuCl_3$", PR**Re**search **2**, 033011 (2020).

P. A. Maksimov, unpublished

an aftertaste of it …

☑ “strong” constrains on the parameter space of anisotropic-exchange systems can be inferred from phenomenology

☑ α-$RuCl_3$ is a ferro-antiferromagnet with an easy-plane FM J_1, AFM J_3, and large anisotropic $J_{z\pm}$. It is in a ZZ phase that is in a proximity of IC phase

☑ common features include broad continuum coexisting with well-defined modes at low energies

P. A. Maksimov and **SC**, “**Re**thinking α-$RuCl_3$”, PR**Re**search **2**, 033011 (2020).