

Heisenberg-Kitaev physics in magnetic fields

Lukas Janssen

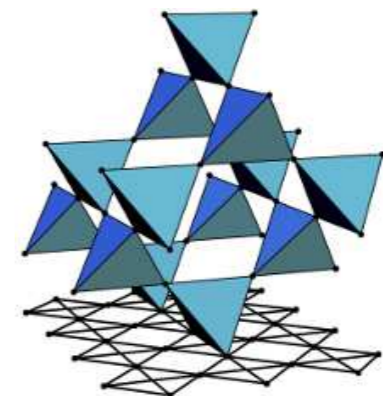
&

Eric Andrade, Matthias Vojta

L.J., E. Andrade, and M. Vojta, Phys. Rev. Lett. **117**, 277202 (2016)

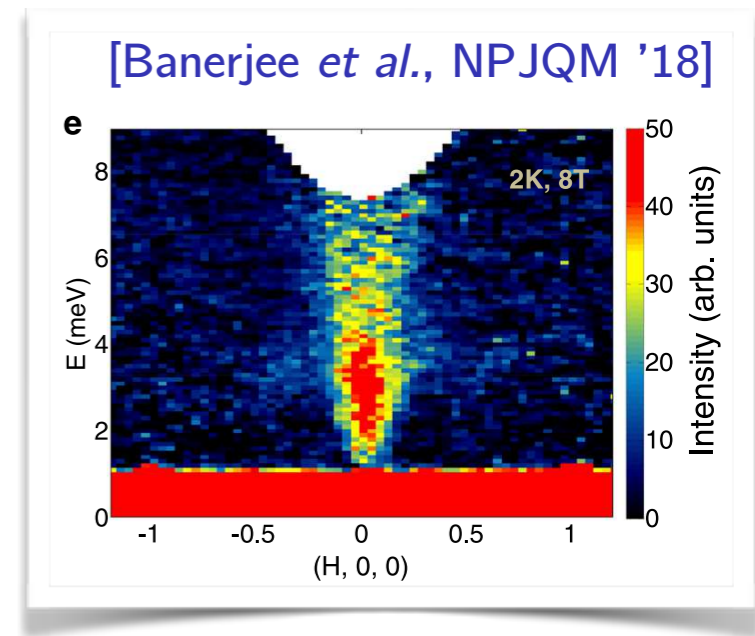
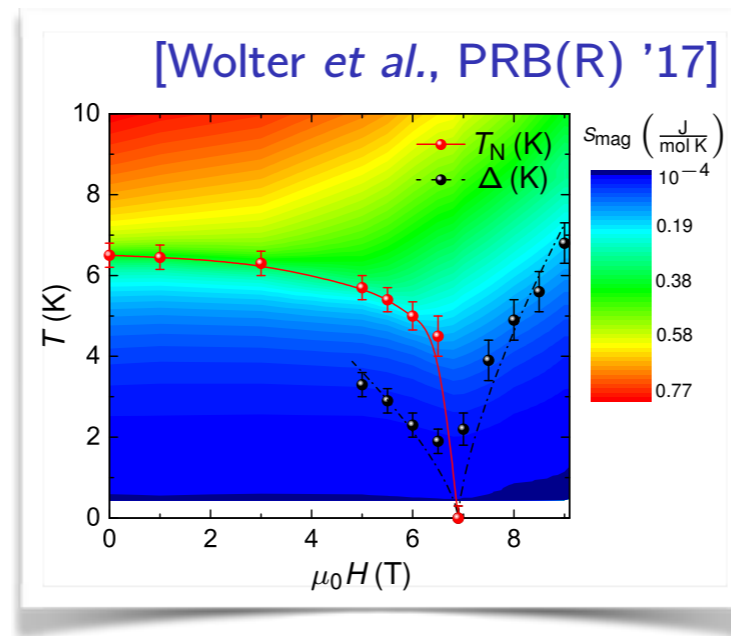
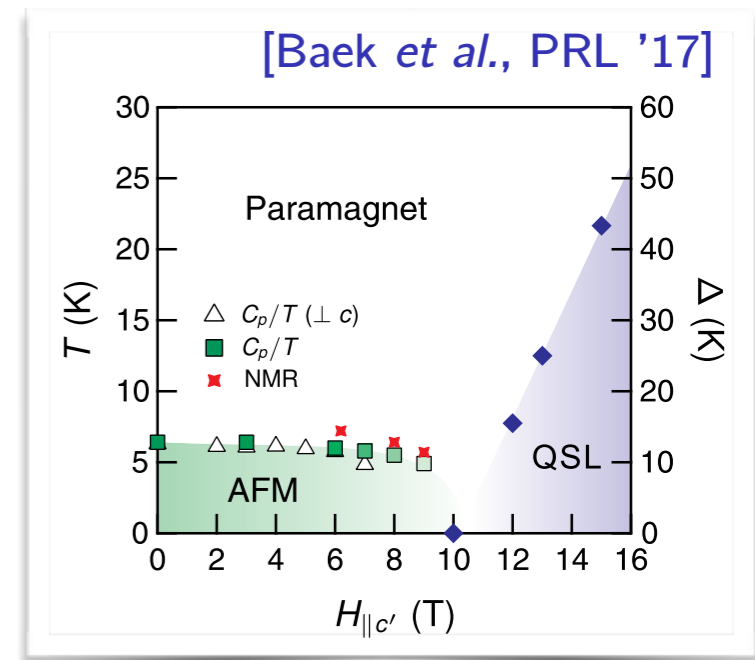
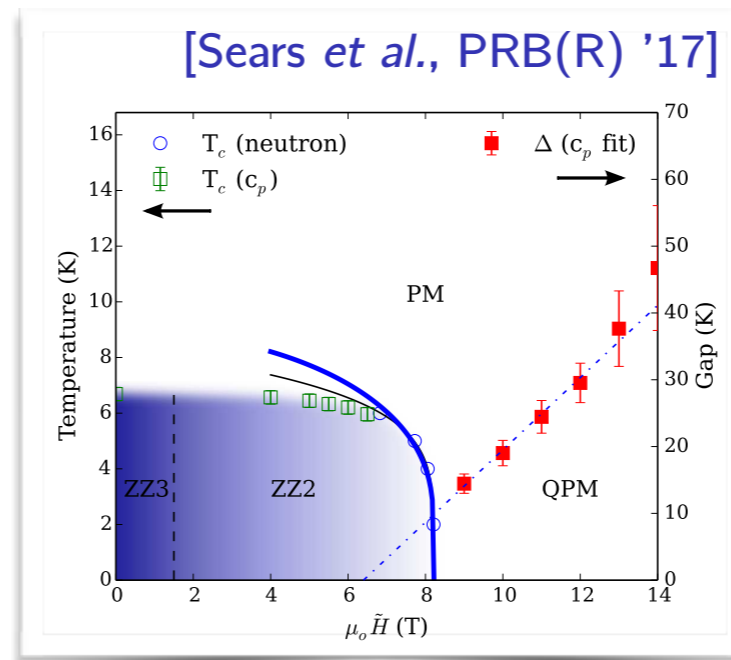
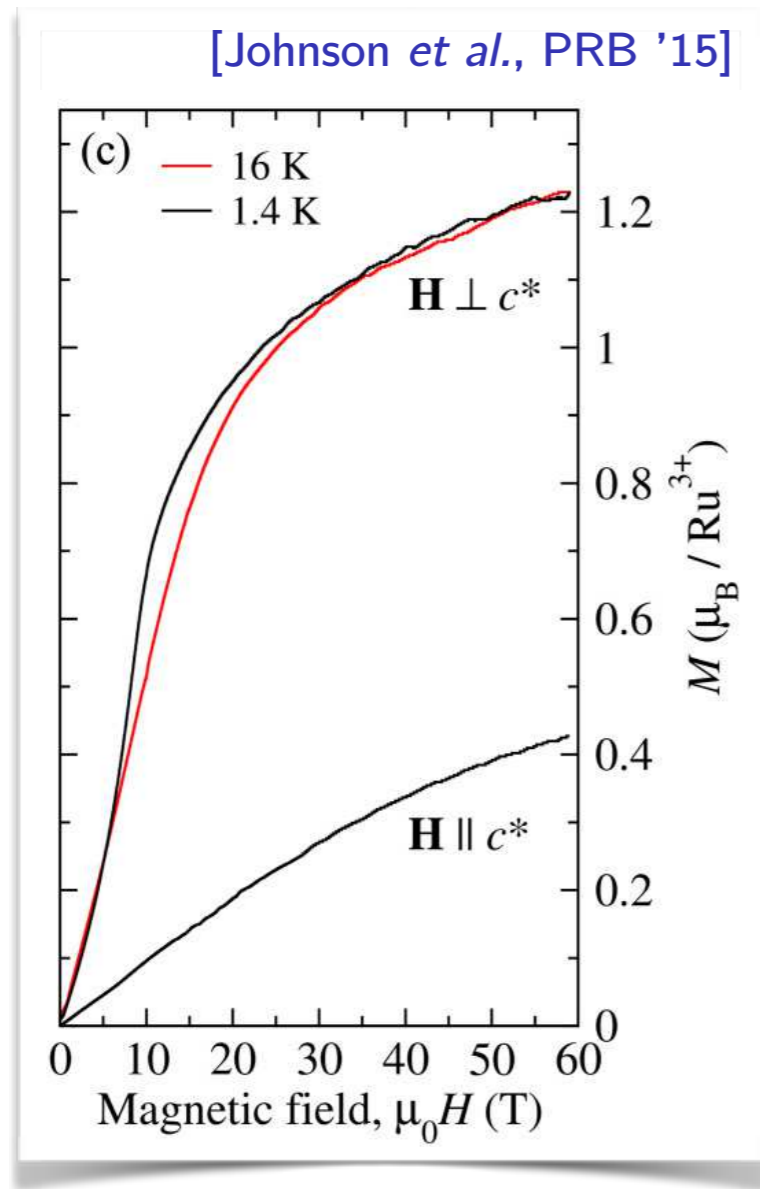
L.J., E. Andrade, and M. Vojta, Phys. Rev. B **96**, 064430 (2017)

A. Wolter, L. Corredor, L.J., *et al.*, Phys. Rev. B **96**, 041405(R) (2017)



SFB 1143

α -RuCl₃ in field:



Questions:

- Reason for anisotropy?
- Nature of field-induced phases?
- Appropriate spin model?

... g factors or intrinsic?

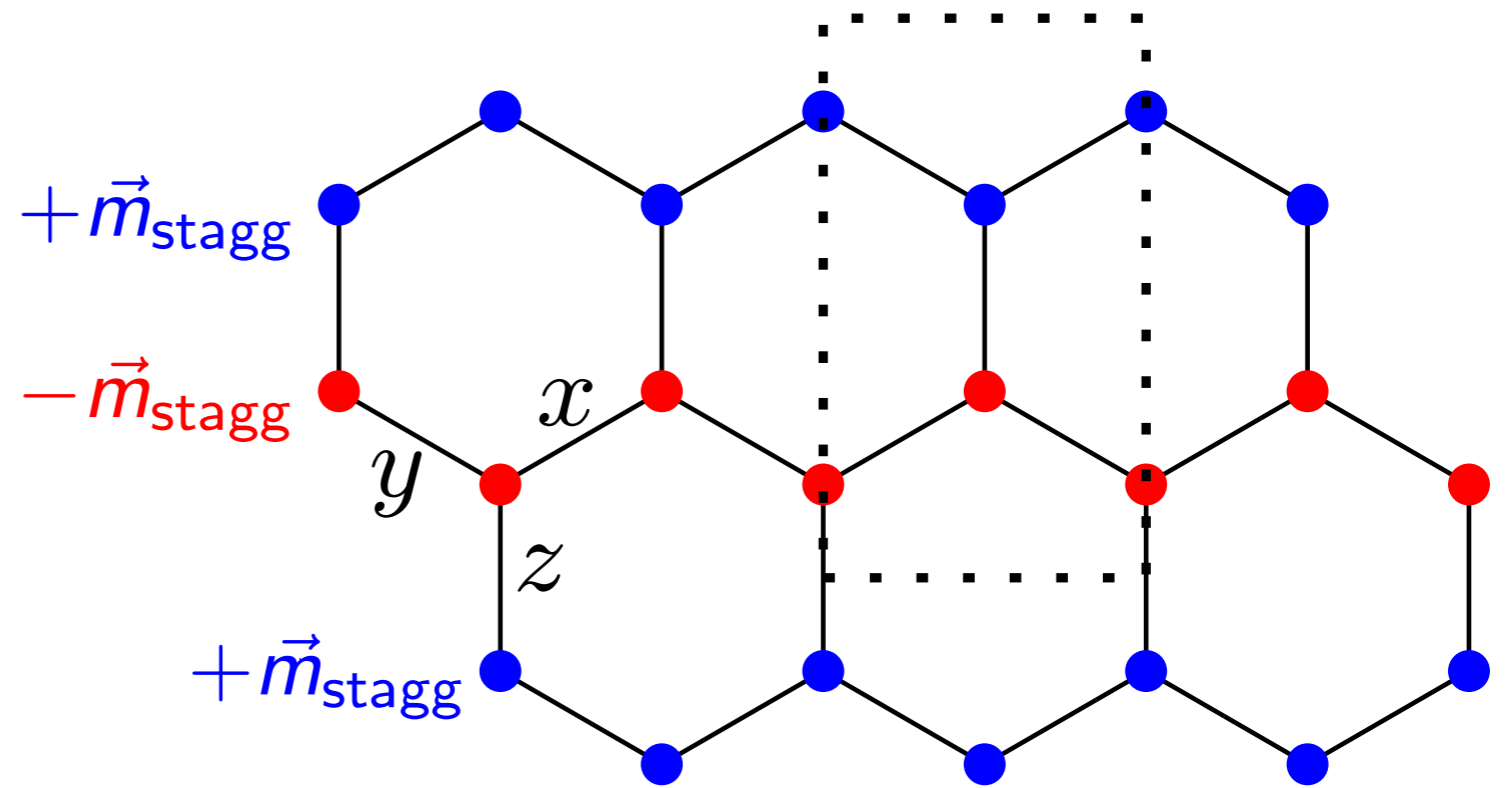
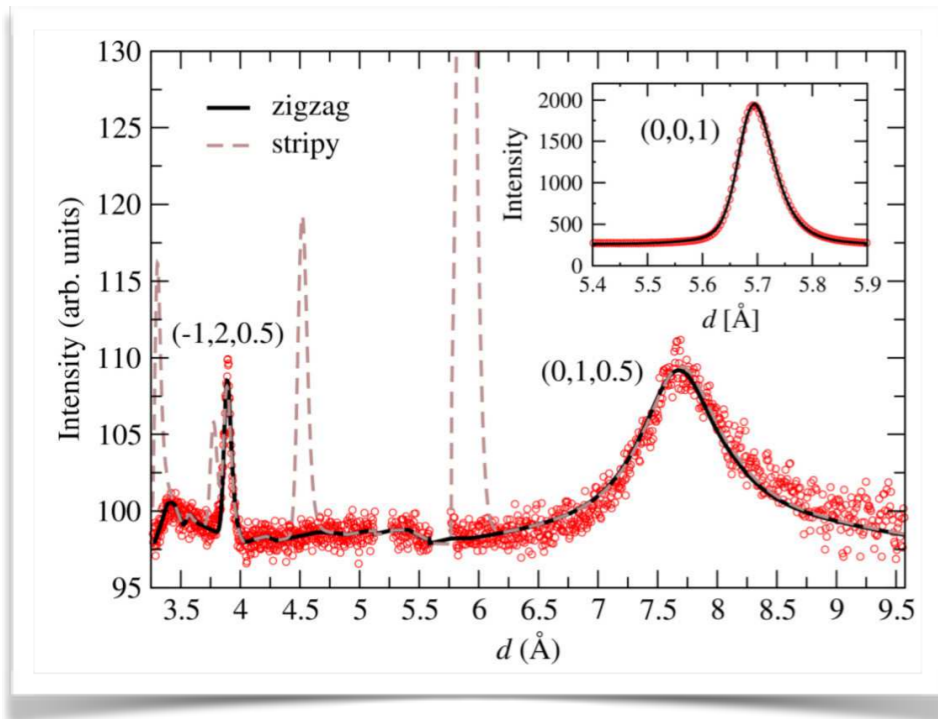
... quantum paramagnet/topological spin liquid?

... sign of K_1 , role of J_3 , Γ_1 ?

α -RuCl₃ in zero field: Zigzag antiferromagnet

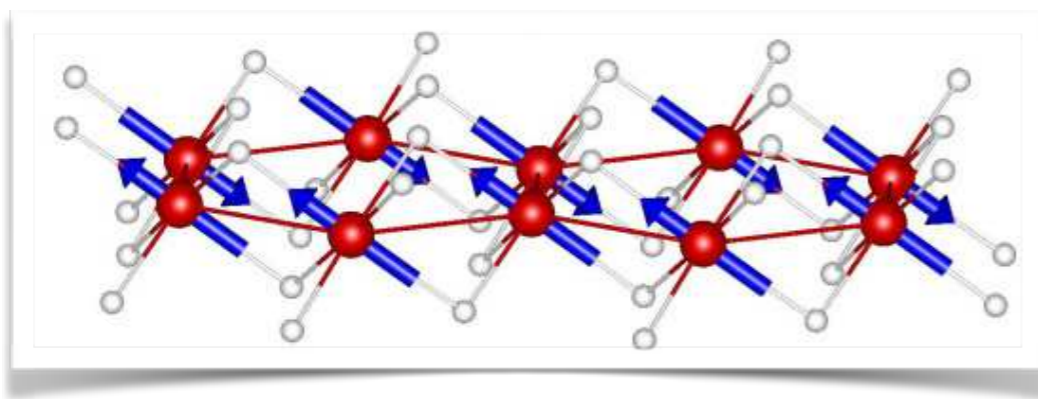
Neutron diffraction:

[Johnson *et al.*, PRB '15]



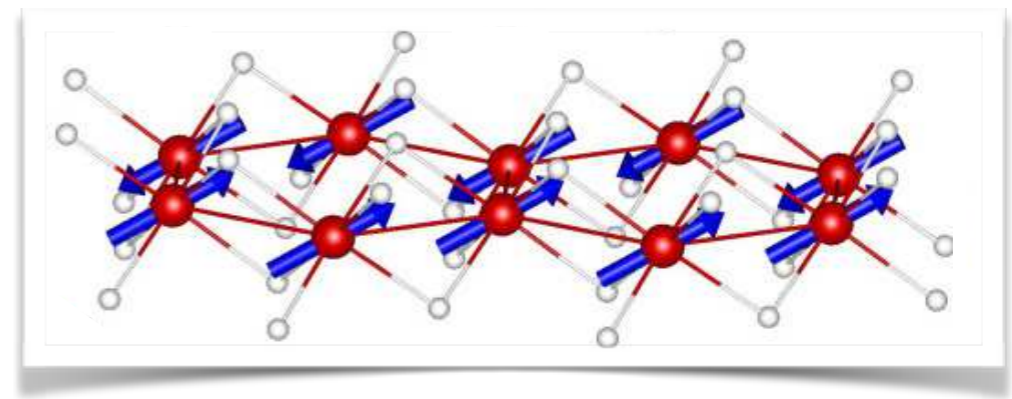
Moment direction:

[Cao *et al.*, PRB '16]



$$\vec{S} \parallel \pm[001] \in ac^*, \quad \angle(\vec{S}, a) \simeq -35^\circ$$

or



$$\vec{S} \parallel \pm[xxz] \in ac^*, \quad \angle(\vec{S}, a) \simeq +35^\circ$$

Mechanisms to stabilize zigzag

Extended Heisenberg-Kitaev models:

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right] \\ + \sum_{\langle\langle ij \rangle\rangle} \left(J_2 \vec{S}_i \cdot \vec{S}_j + K_2 S_i^\gamma S_j^\gamma \right) + \sum_{\langle\langle\langle ij \rangle\rangle\rangle} J_3 \vec{S}_i \cdot \vec{S}_j - \vec{h} \cdot \sum_i \vec{S}_i$$

... neglect trigonal distortion

Set	Material	J_1 (meV)	K_1 (meV)	Γ_1 (meV)	J_2 (meV)	K_2 (meV)	J_3 (meV)	Method	Ref.	Year
1	α -RuCl ₃	-4.6	+7.0	-	-	-	-	Fit to neutron scattering	[35,36]	2016
1'	Na ₂ IrO ₃	-4.0	+10.5	-	-	-	-	Fit to susceptibility and neutron scattering	[30]	2013
1 + Γ	α -RuCl ₃	-12	+17	+12	-	-	-	DFT + t/U expansion	[44]	2015
2	Na ₂ IrO ₃	0	-17	0	0	-	+6.8	DFT + exact diagonalization	[32]	2016
2 + Γ	Na ₂ IrO ₃	+3	-17	+1	-3	+6	+1	DFT + t/U expansion, direction of moments	[40,45]	2016
(2 + Γ)'	Na ₂ IrO ₃	+3	-17.5	+1	+5	-	+5	MRCI, fit to θ_{CW}	[47]	2014
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Three scenarios:

(1) Antiferromagnetic K_1 , ferromagnetic J_1

[Chaloupka *et al.*, PRL '13]

Mechanisms to stabilize zigzag

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[Chaloupka *et al.*, PRL '13]

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[Winter *et al.*, PRB '16]

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(2) Ferromagnetic K_1 , antiferromagnetic J_3

[Winter *et al.*, PRB '16]

[Katukuri *et al.*, NJP '14; ...]

(3) Ferromagnetic K_1 , positive Γ_1

[Rau *et al.*, PRL '14; Ran *et al.*, PRL '17]

... we will show that finite $J_1 < 0$ is also needed

Mechanisms to stabilize zigzag

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[Rau *et al.*, PRL '14; Ran *et al.*, PRL '17]

... we will show that finite $J_1 < 0$ is also needed

This talk: Magnetic-field behavior allows to **distinguish** scenarios!

Warm-up: SU(2) Heisenberg antiferromagnet in field

Minimal model:

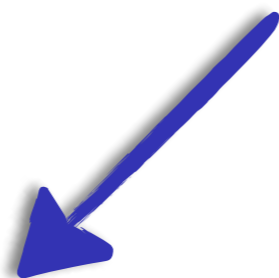
$$\mathcal{H} = \sum_{\langle ij \rangle} J_1 \vec{S}_i \cdot \vec{S}_j - \vec{h} \cdot \sum_i \vec{S}_i$$



Warm-up: SU(2) Heisenberg antiferromagnet in field

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$$|\vec{h}| = 0$$

Warm-up: SU(2) Heisenberg antiferromagnet in field

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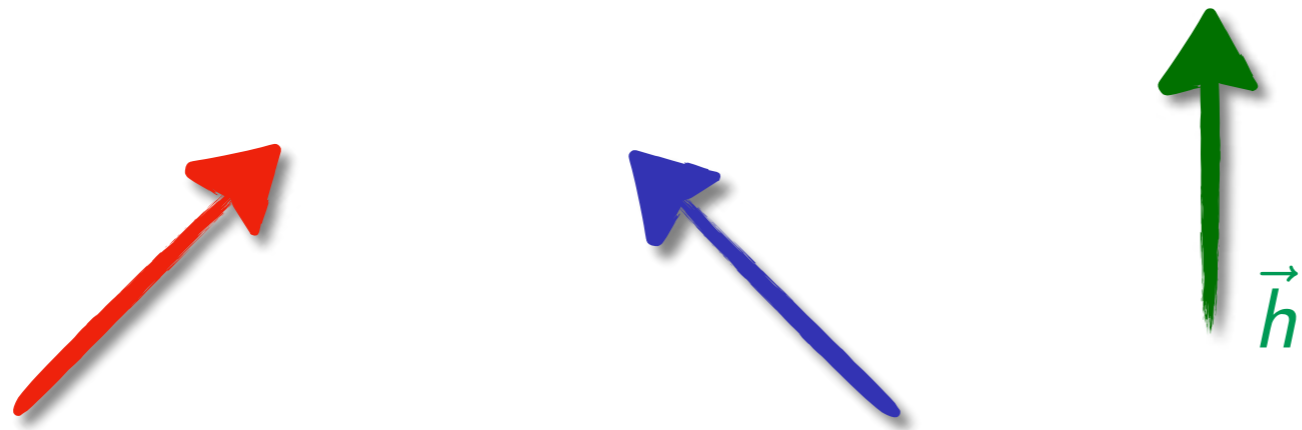
$$0 < |\vec{h}| \ll J_1 S$$

$\vec{S}_i \perp \vec{h}$ (largest susceptibility)

Warm-up: SU(2) Heisenberg antiferromagnet in field

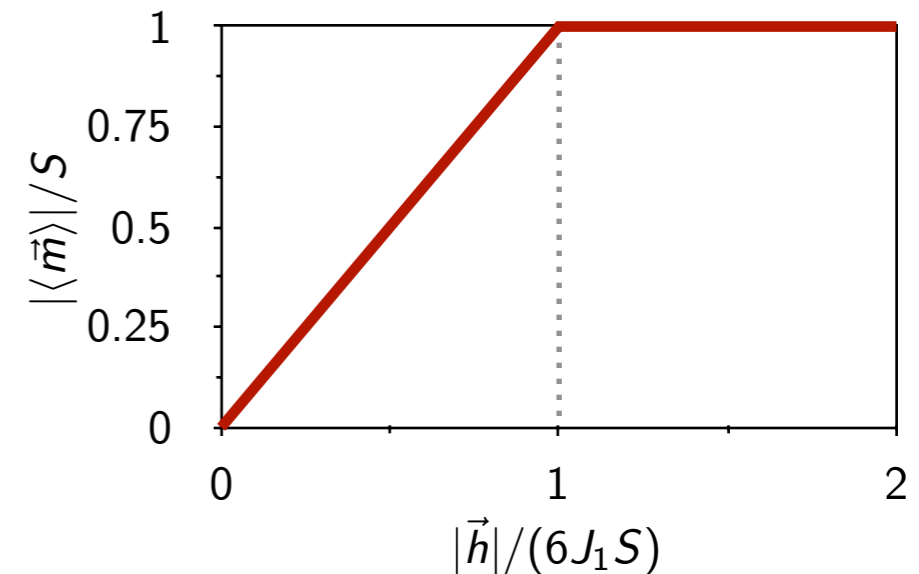
Minimal model:

$$\mathcal{H} = \sum_{\langle ij \rangle} J_1 \vec{S}_i \cdot \vec{S}_j - \vec{h} \cdot \sum_i \vec{S}_i$$



$$0 < |\vec{h}| < 6J_1 S$$

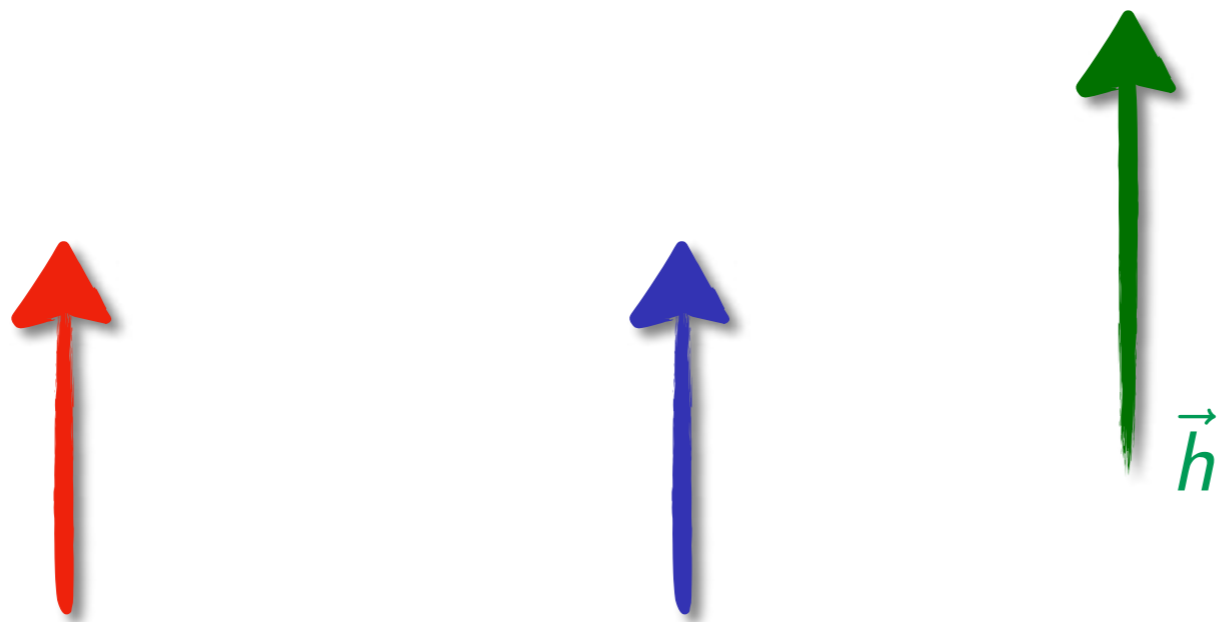
“homogenous” canting



Warm-up: SU(2) Heisenberg antiferromagnet in field

Minimal model:

$$\mathcal{H} = \sum_{\langle ij \rangle} J_1 \vec{S}_i \cdot \vec{S}_j - \vec{h} \cdot \sum_i \vec{S}_i$$

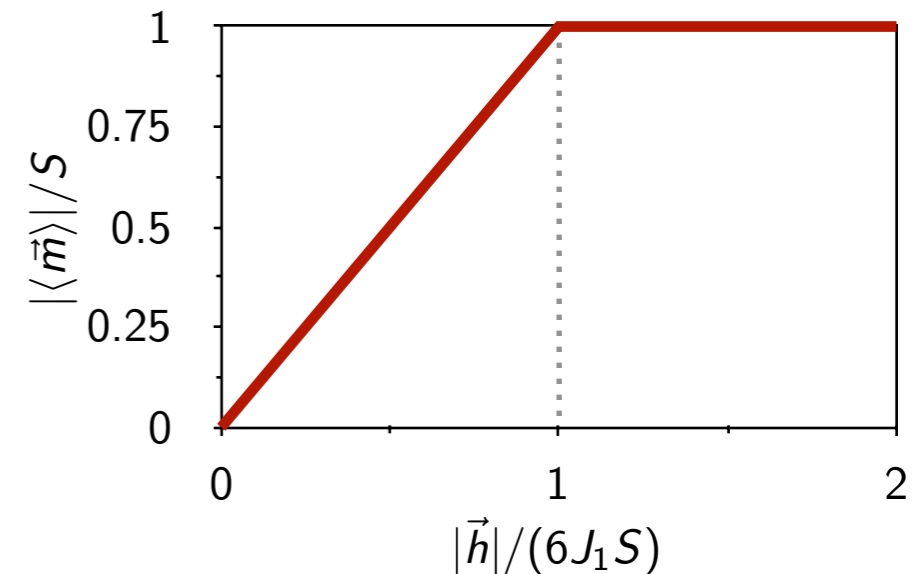


$$6J_1 S \leq |\vec{h}|$$

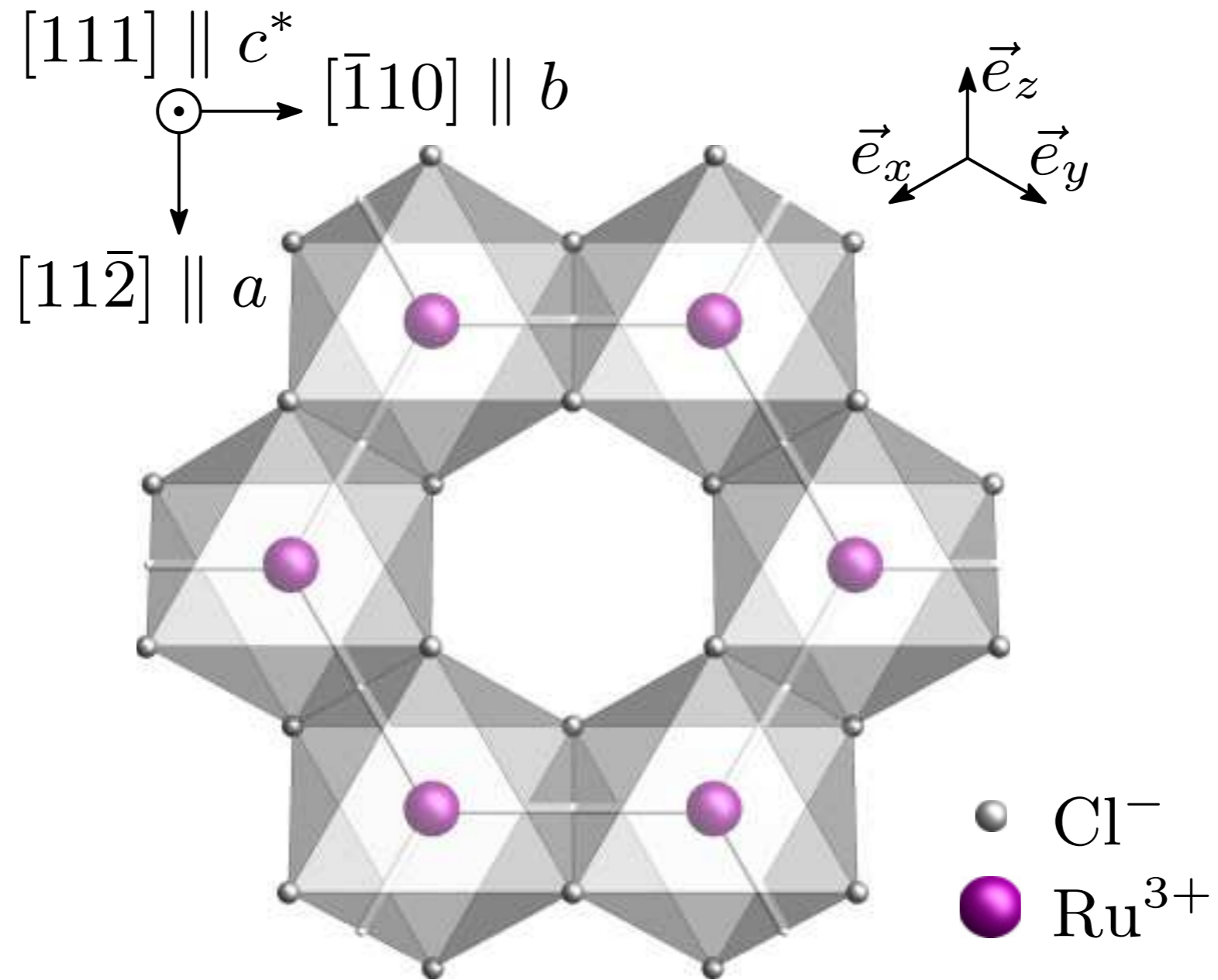
fully polarized

$$|\langle \vec{m} \rangle| / S = 1$$

... since one-magnon state eigenstate of \mathcal{H}



α -RuCl₃: Field directions



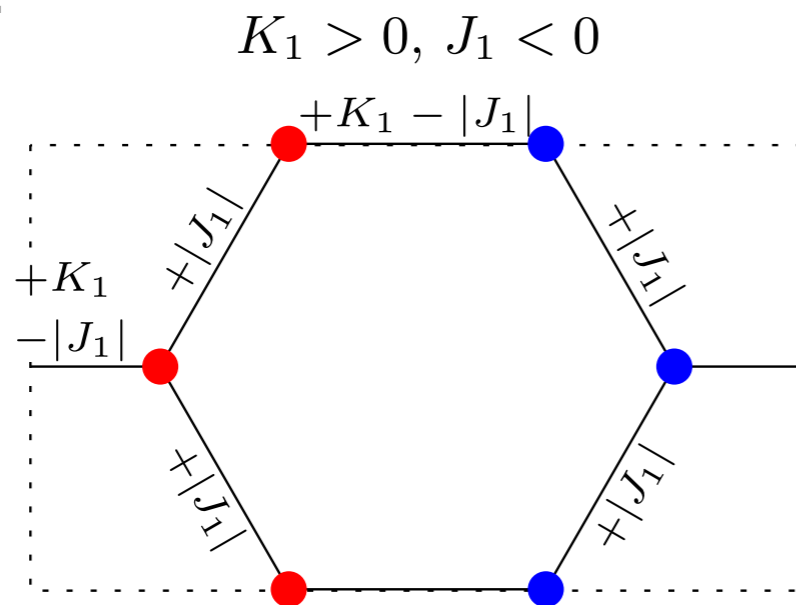
Scenario 1: Antiferromagnetic K_1 , ferromagnetic J_1

Minimal model:

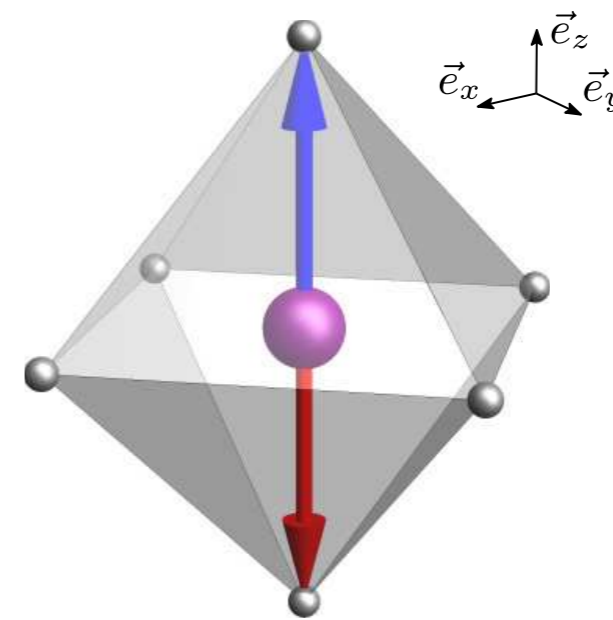
[Chaloupka *et al.*, PRL '10; PRL '13]

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma \right]$$

Zero field:



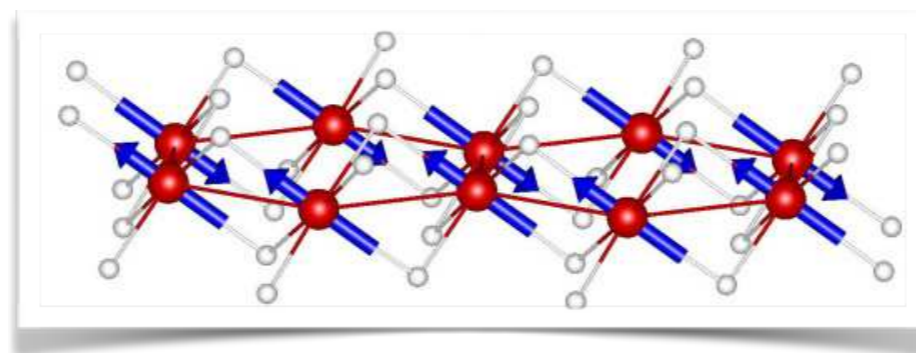
“cubic-axes zigzag”



$\vec{S} \parallel \pm \vec{e}_z$

... along Ru-Cl bonds

... consistent with one of the two options in α - RuCl_3 :

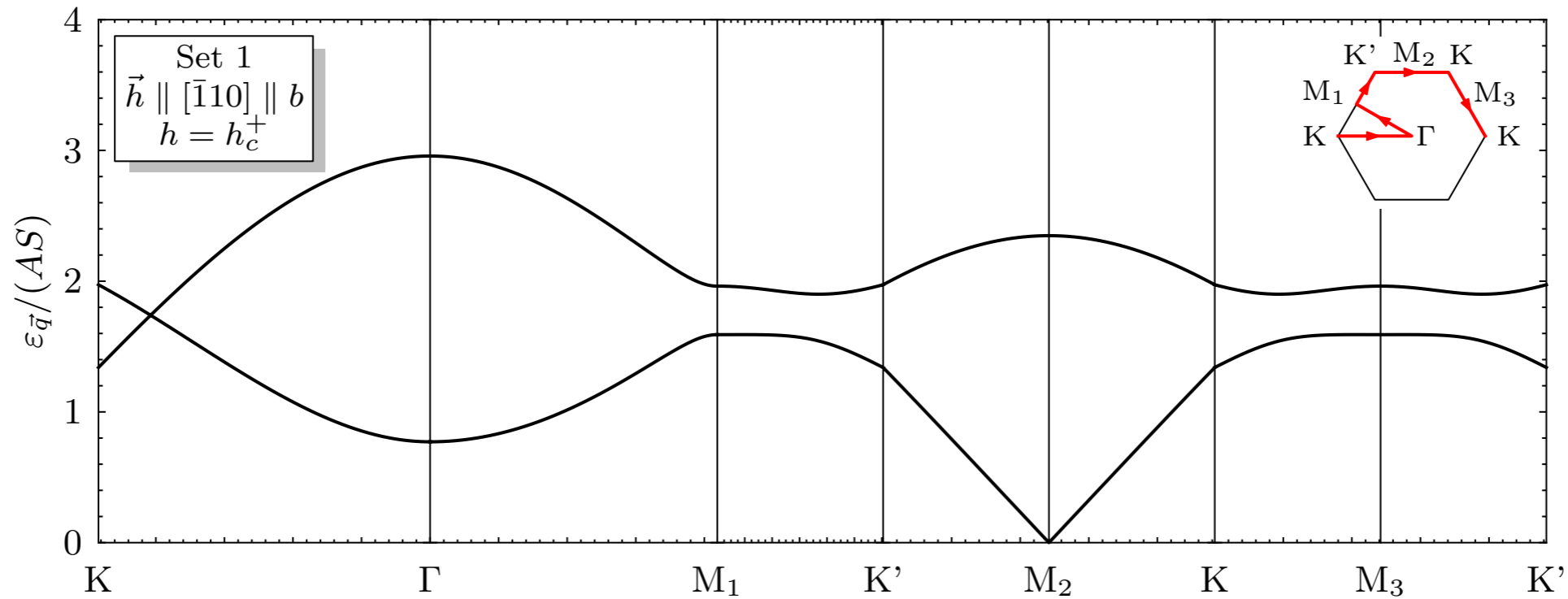


[Cao *et al.*, PRB '16]

Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [\bar{1}10] \parallel b$

... along Ru-Ru bonds

Spin-wave spectrum in high-field phase:



$(J_1, K_1) = (-4.6, +7.0)$ meV

[Banerjee *et al.*, Nat. Mat. '16]

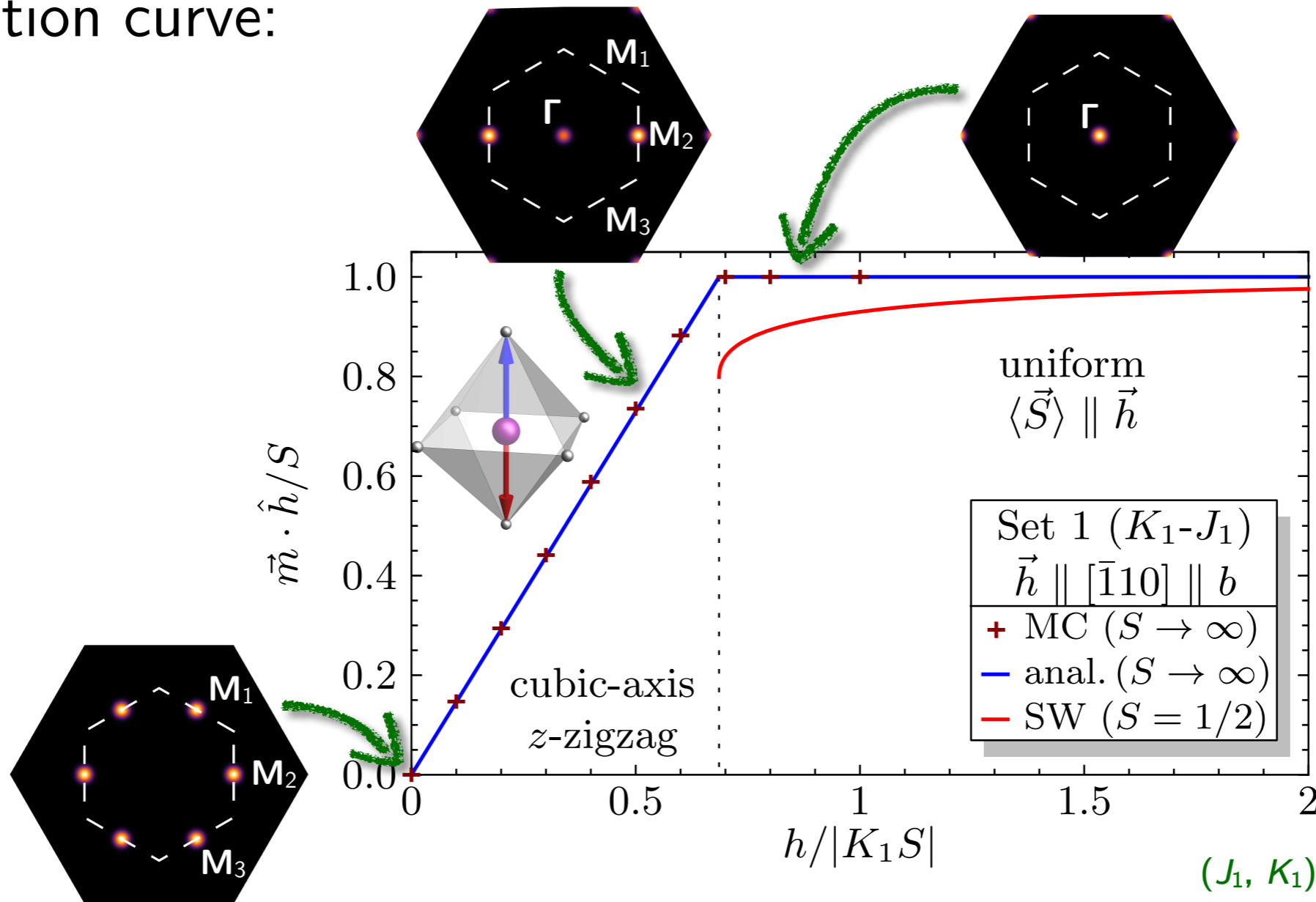
... gap closes precisely at z zigzag ordering wavevector $\vec{Q} = \mathbf{M}_2$

... consistent with a **direct continuous** transition to canted zigzag

Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [\bar{1}10] \parallel b$

... along Ru-Ru bonds

Magnetization curve:

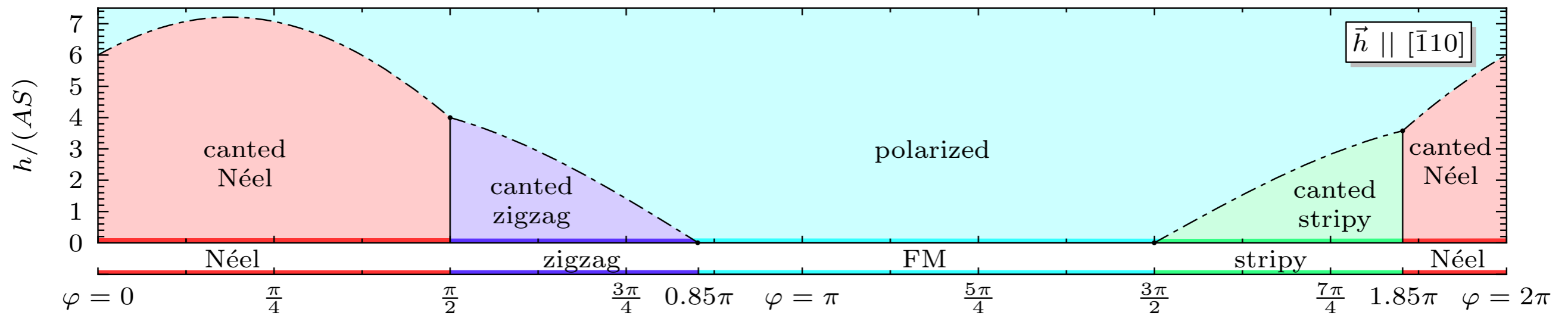


$(J_1, K_1) = (-4.6, +7.0)$ meV

... but: high-field state not fully polarized: $|\langle \vec{m} \rangle| / S < 1$

Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [\bar{1}10] \parallel b$

Classical phase diagram in $[\bar{1}10]$ field:



$$J_1 = A \cos \varphi$$

$$K_1 = 2A \sin \varphi$$

Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

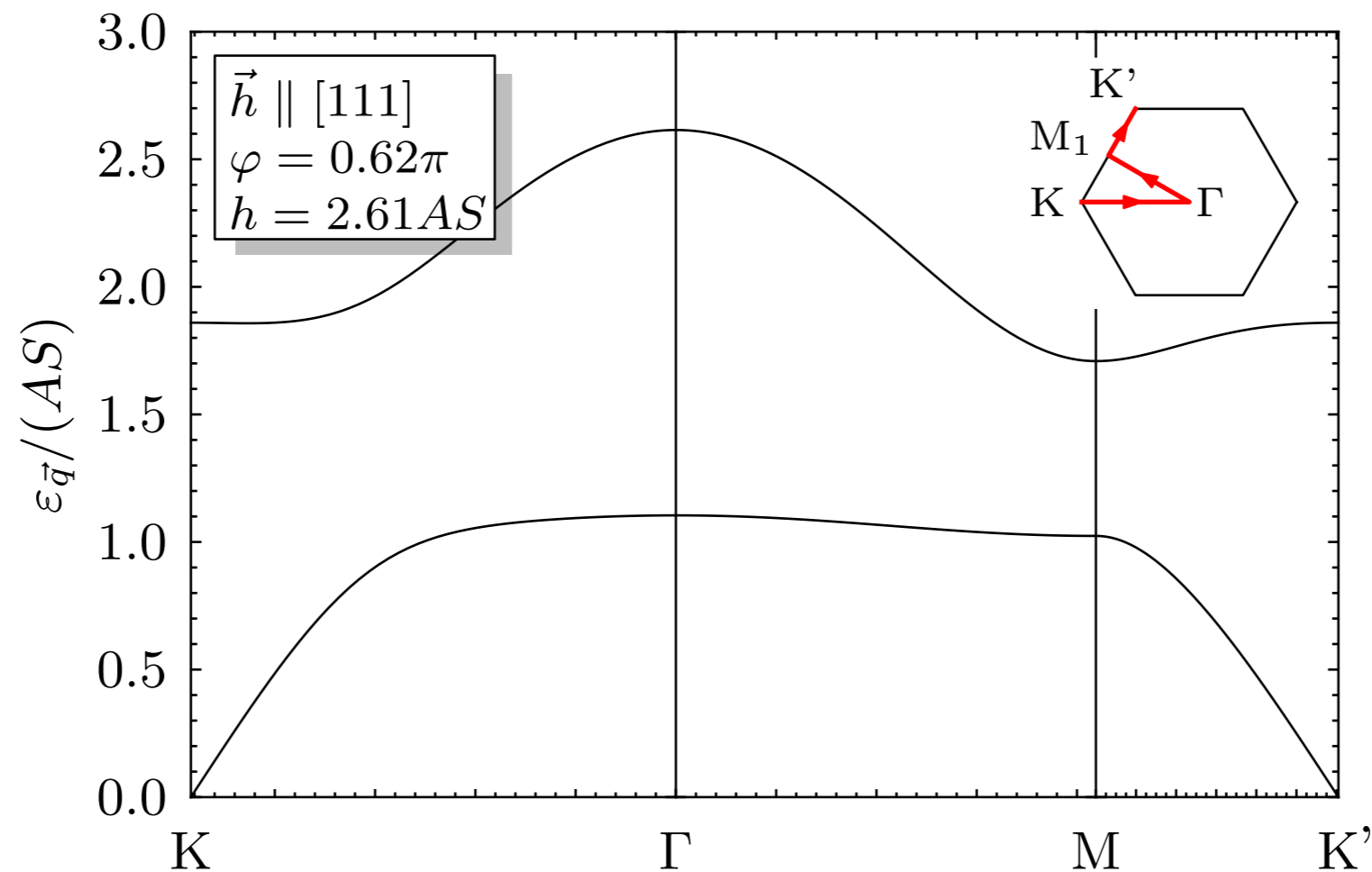
Zero field: \vec{S}_i along cubic axes

... "cubic-axes zigzag"

\Rightarrow simple canting impossible!

... canted zigzag will compete with other states

Spin-wave spectrum in high-field phase:



... magnon gap closes at **K** points!

... different from zigzag ordering wavevector

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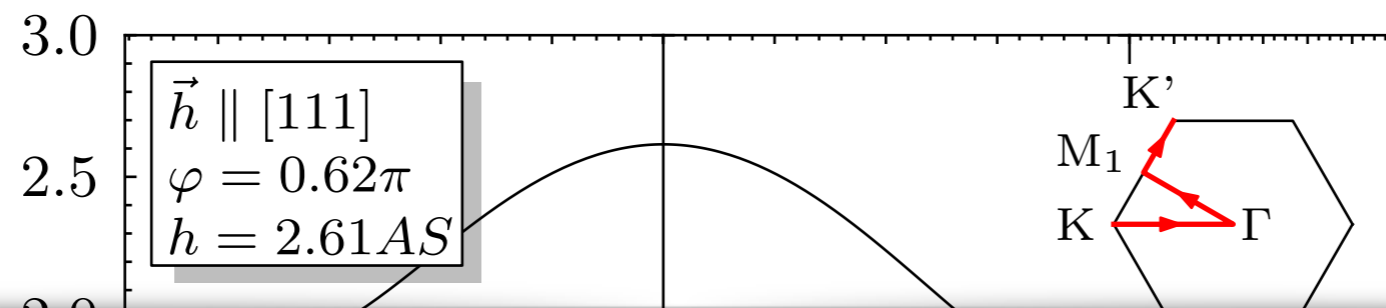
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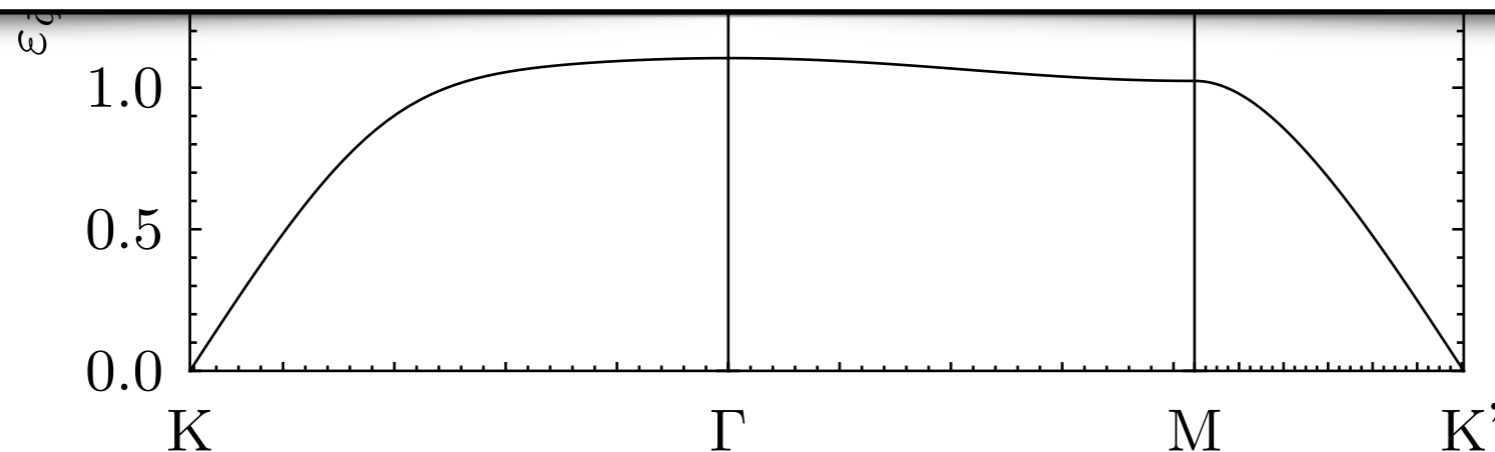
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... canted zigzag will compete with other states

Spin-wave spectrum in high-field phase:



X Continuous transition to canted zigzag impossible!



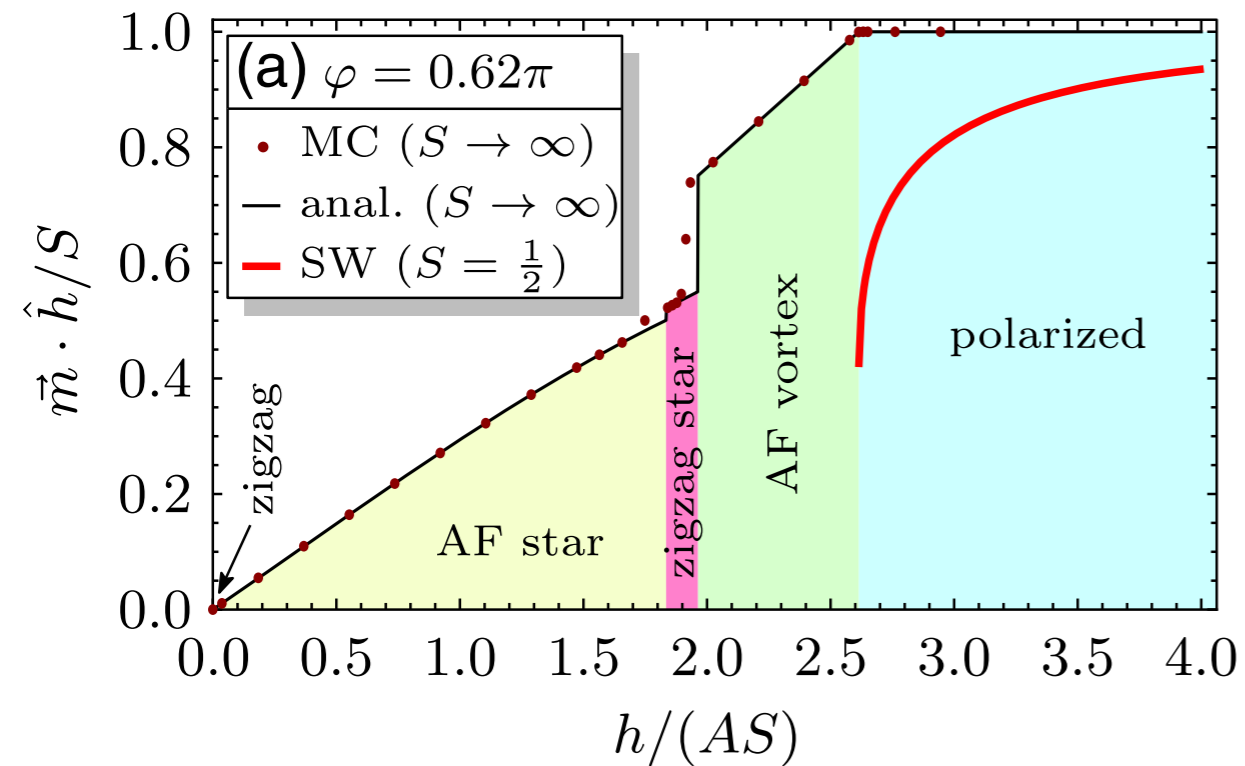
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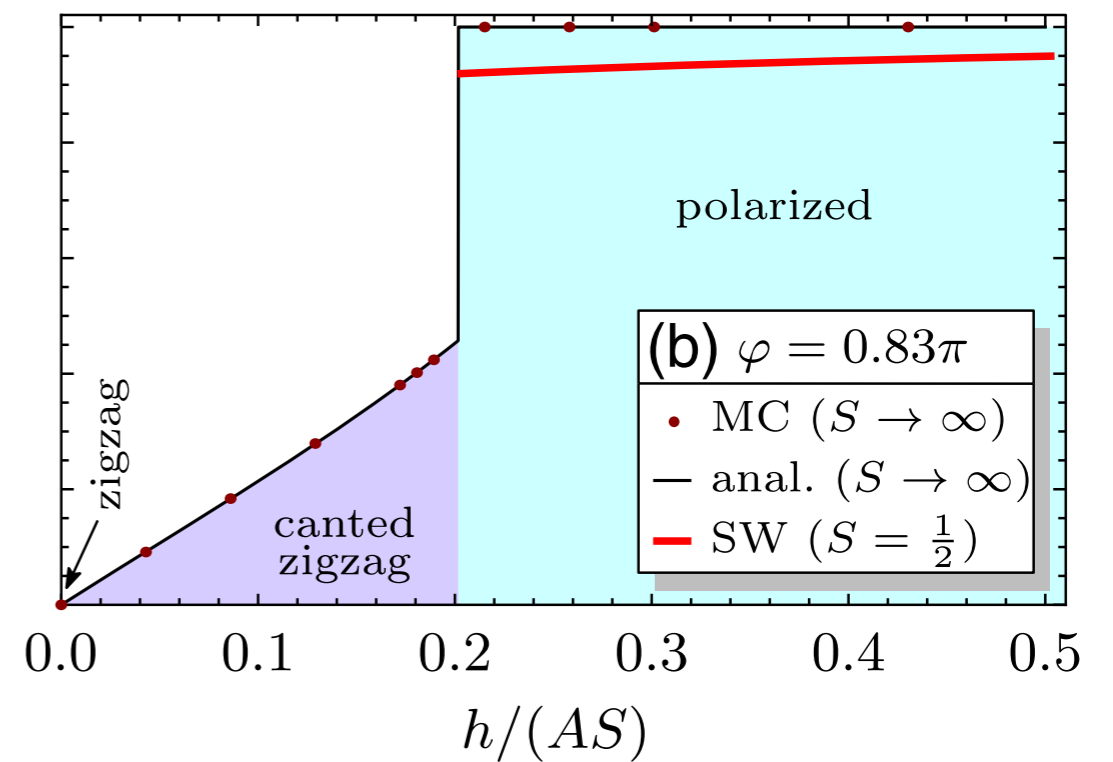
$$K_1 > -2J_1 :$$

intermediate phases



$$-2J_1 > K_1 > -J_1 :$$

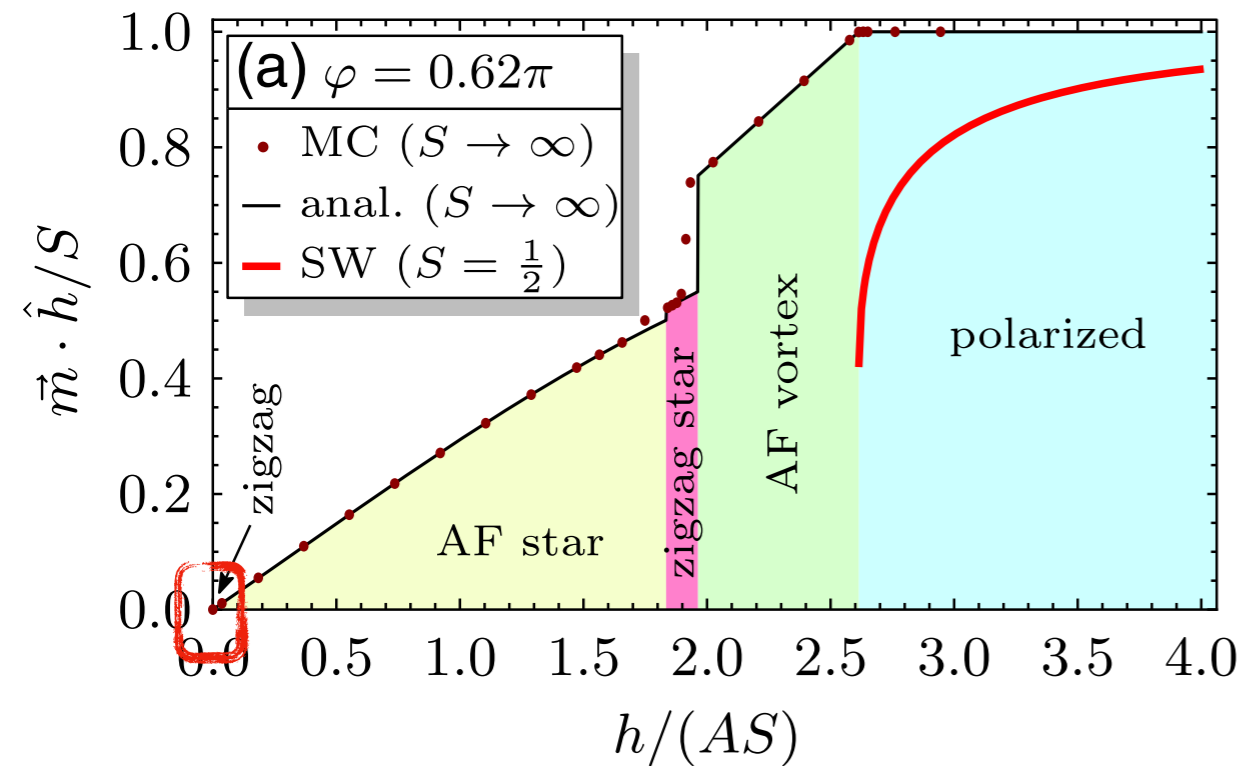
first-order transition



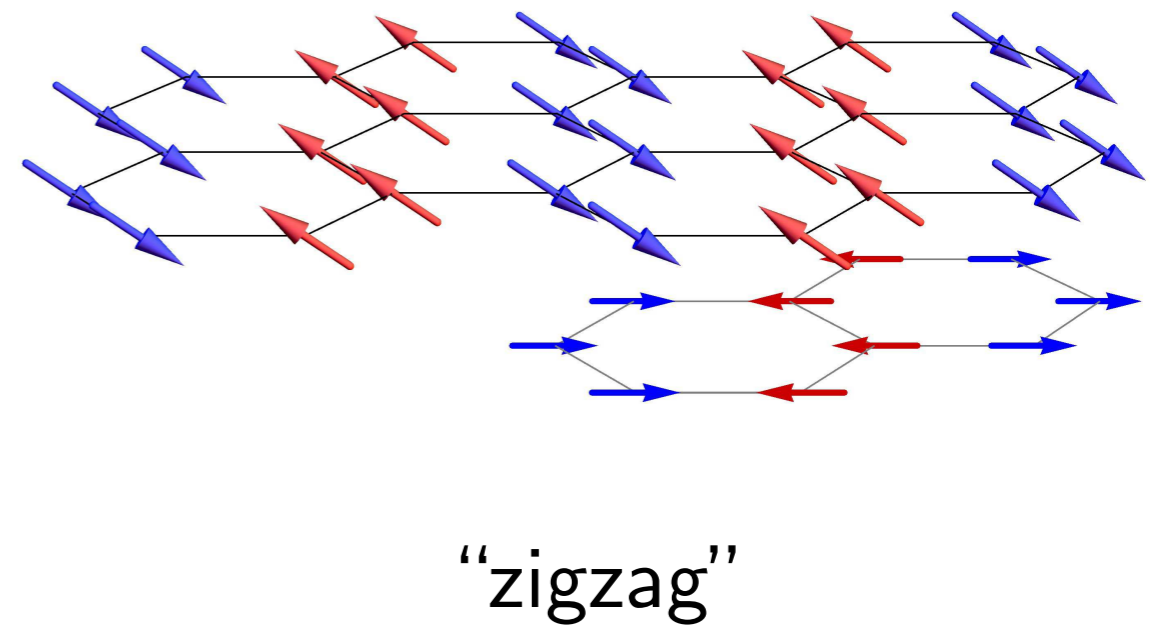
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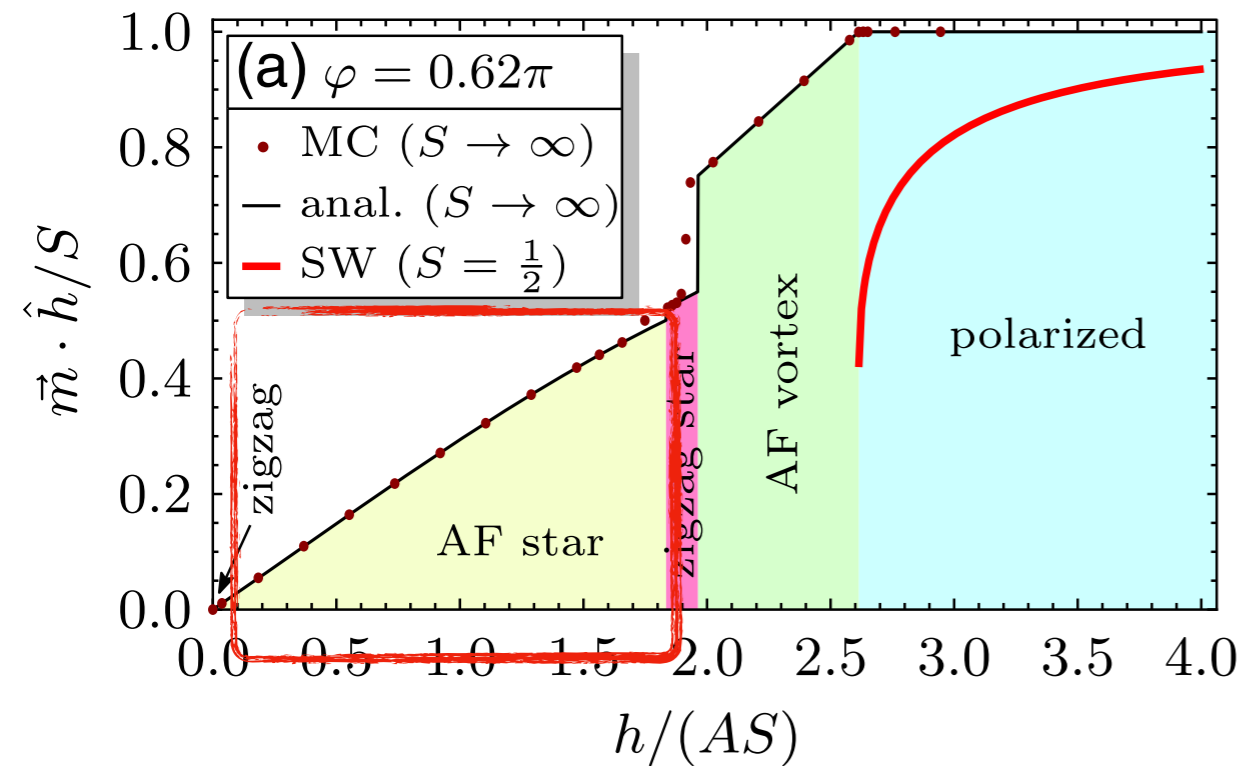
$h = 0$:



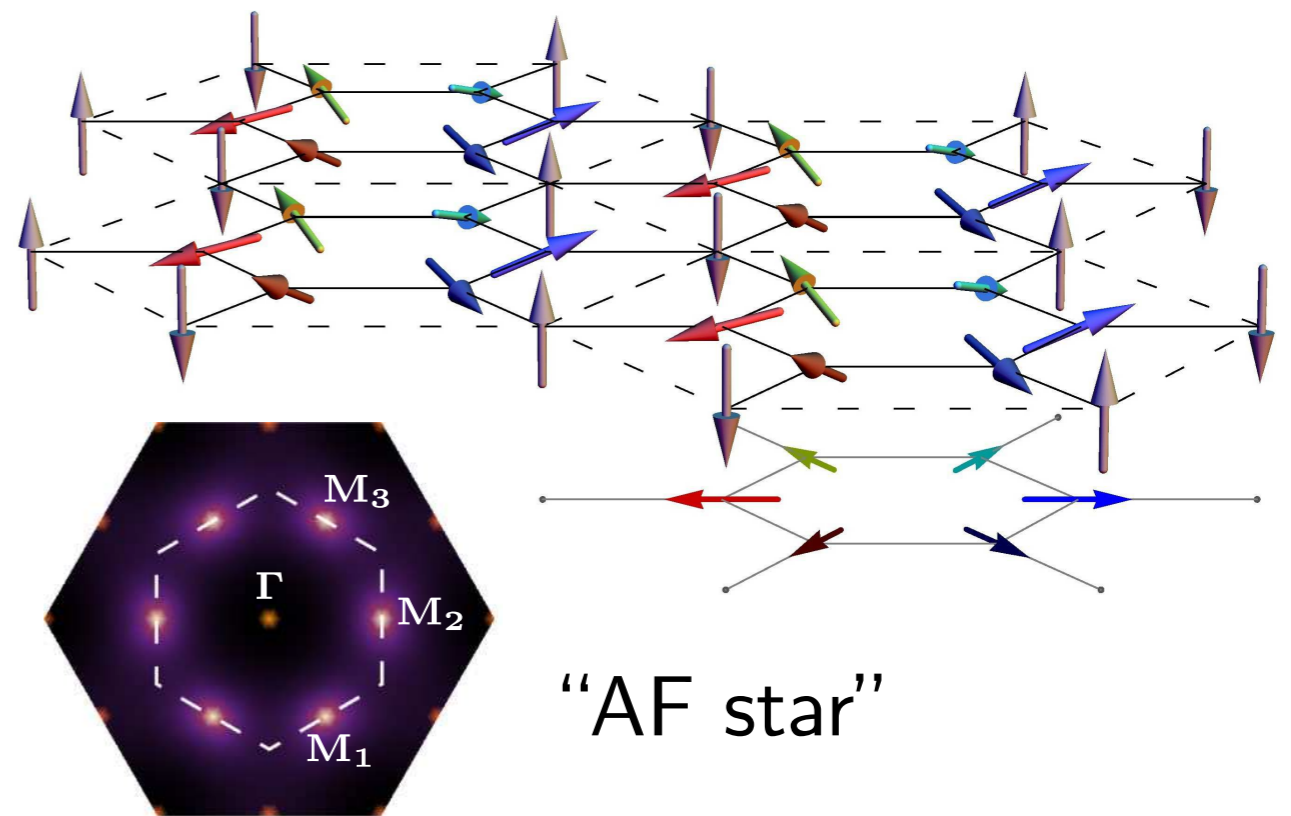
Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

$$K_1 > -2J_1 :$$

intermediate phases



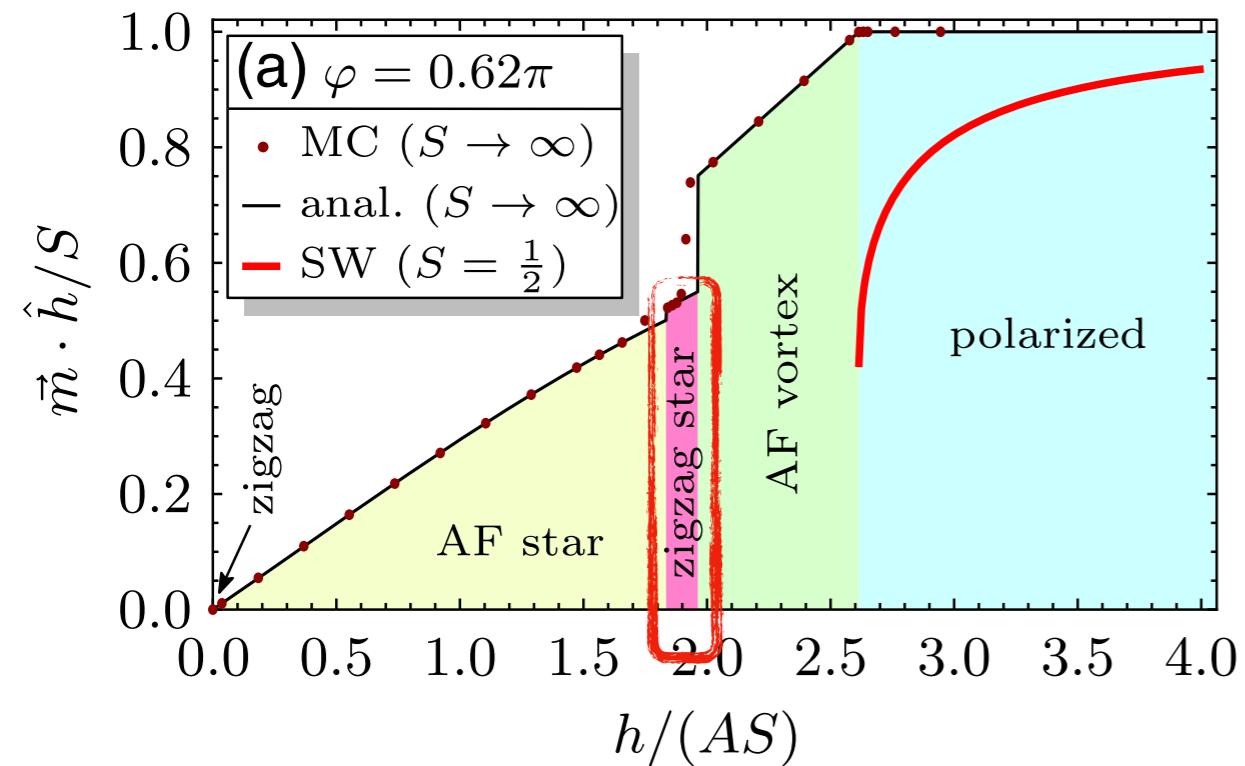
$$0 < h/(AS) < 1.8:$$



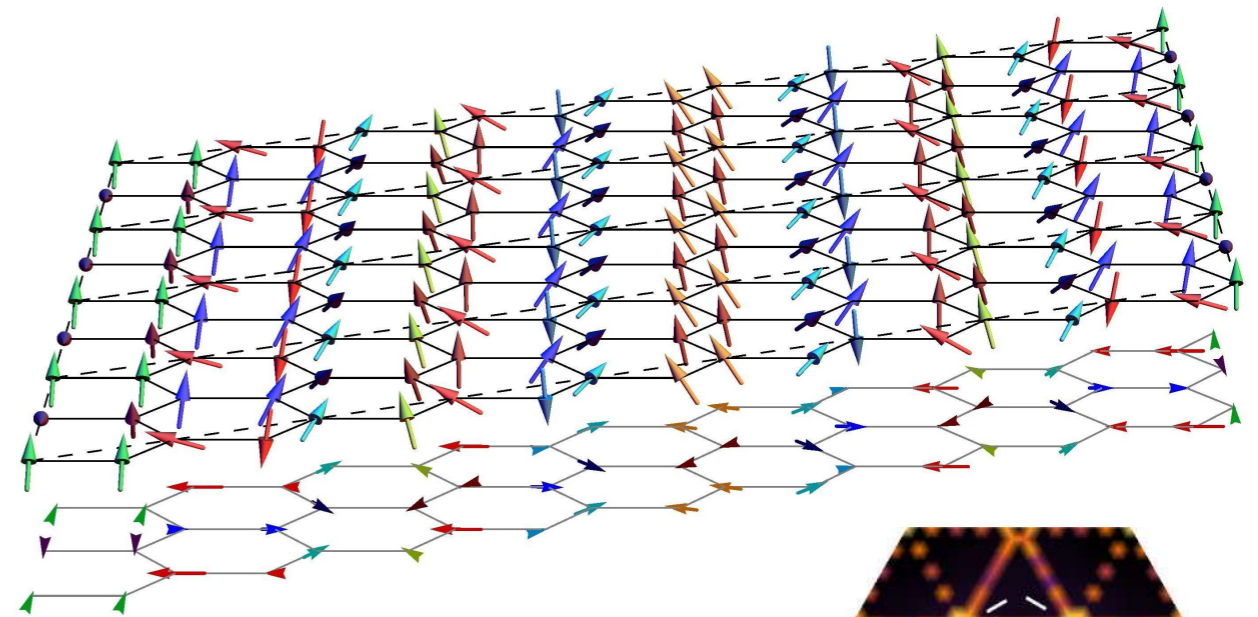
Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

$$K_1 > -2J_1 :$$

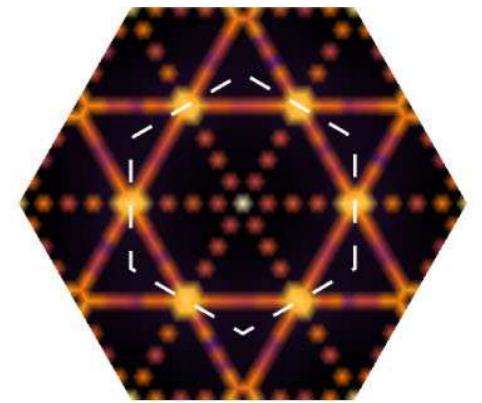
intermediate phases



$$1.8 < h/(AS) < 2.0:$$



“ziggzag star”

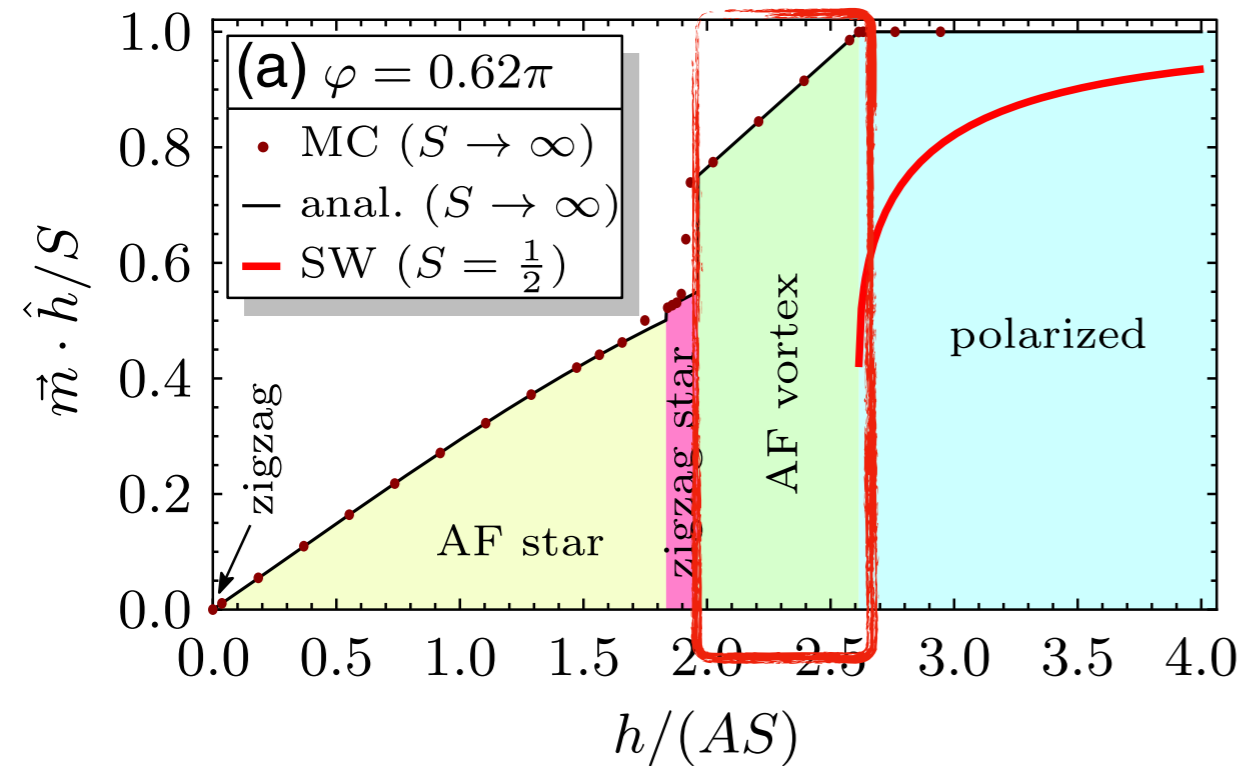


... cf. [Chern *et al.*, PRB '17]

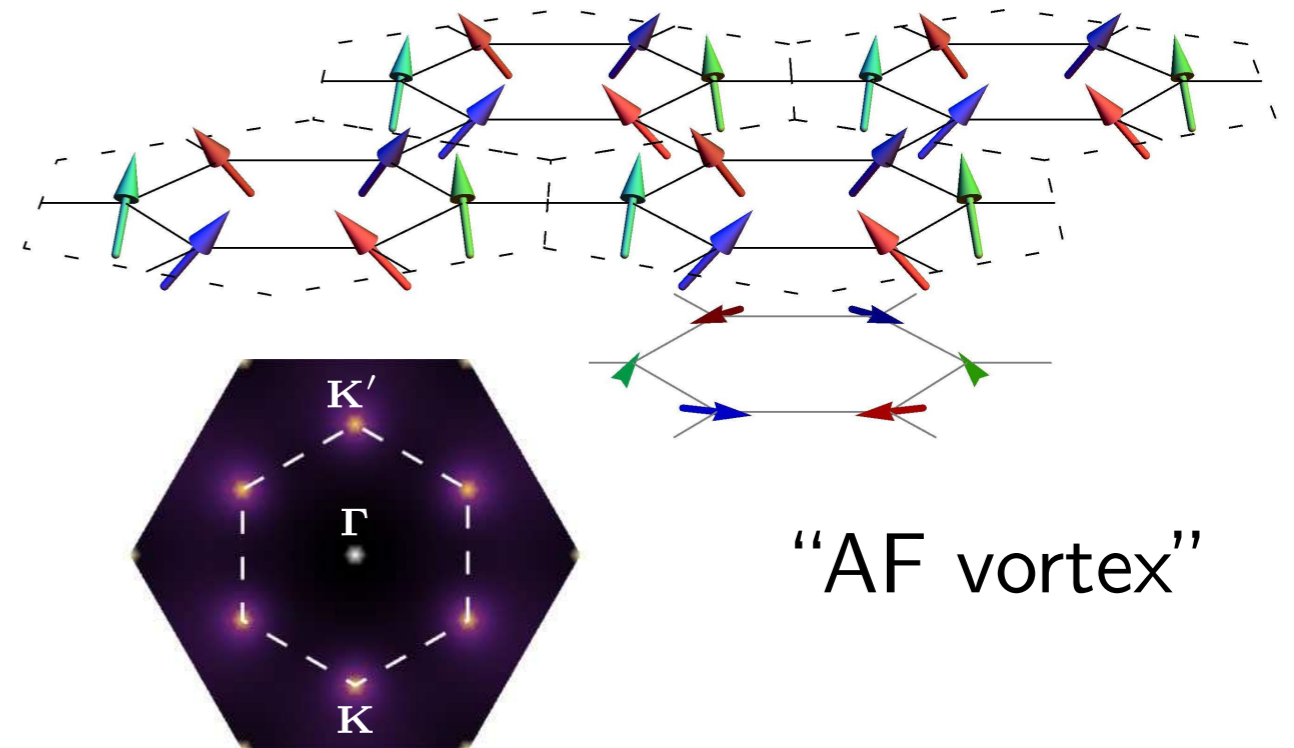
Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

$$K_1 > -2J_1 :$$

intermediate phases



$$2.0 < h/(AS) < 2.6:$$

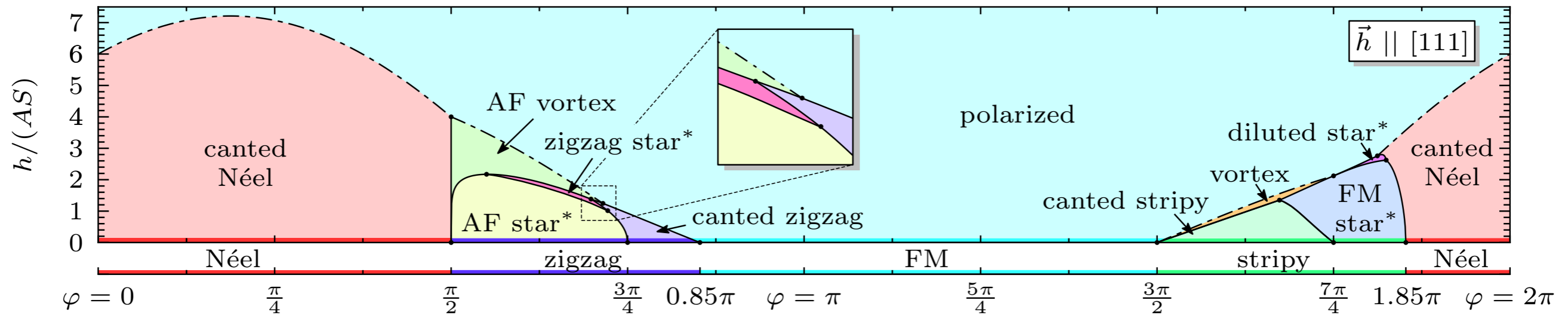


... consistent with spin-wave analysis

Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

Classical phase diagram in $[111]$ field:

[LJ, Andrade, Vojta, PRL '16]



$$J_1 = A \cos \varphi$$

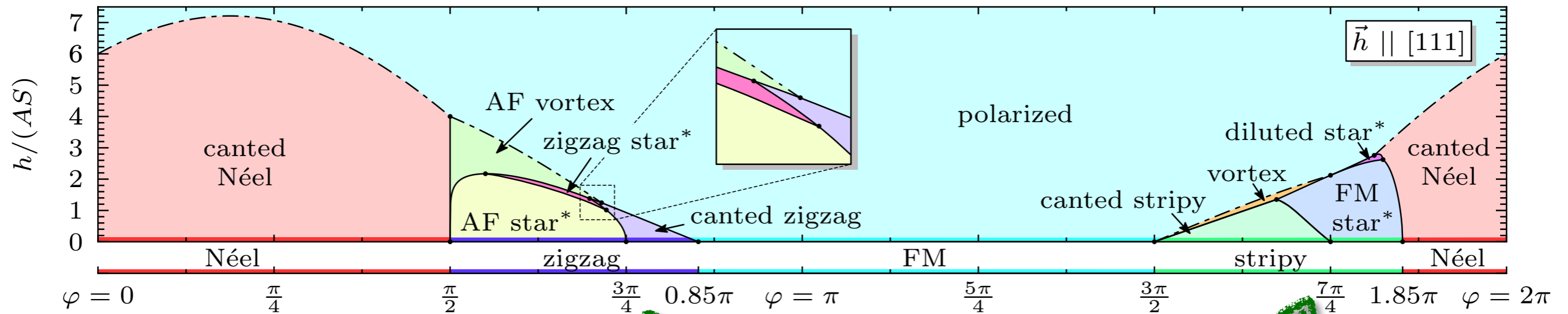
$$K_1 = 2A \sin \varphi$$

... similarly complicated phase diagram for $[11\bar{2}]$ direction (a axis)

Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

Classical phase diagram in $[111]$ field:

[LJ, Andrade, Vojta, PRL '16]



zigzag/stripy & "star"
 degenerate @ "Klein" points
 \Rightarrow intermediate phases also for $S = 1/2$

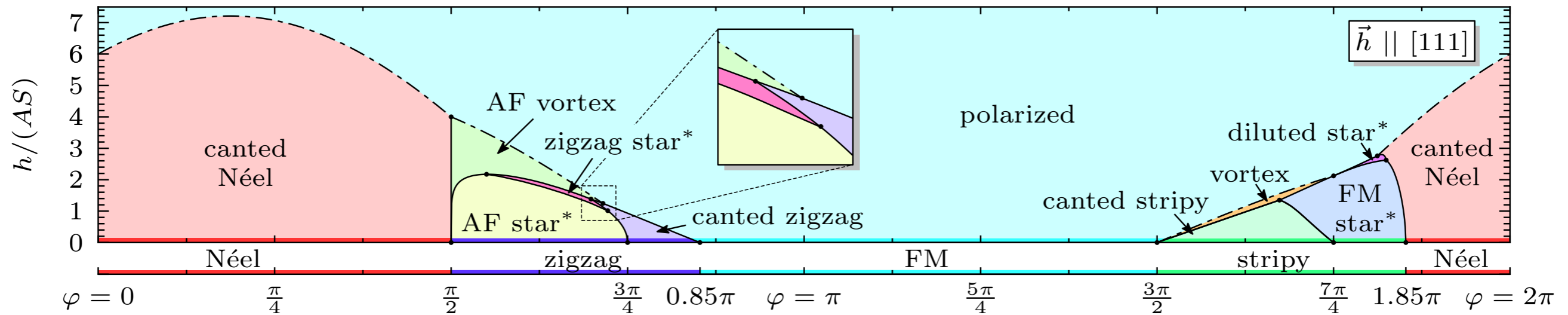
$$J_1 = A \cos \varphi$$

$$K_1 = 2A \sin \varphi$$

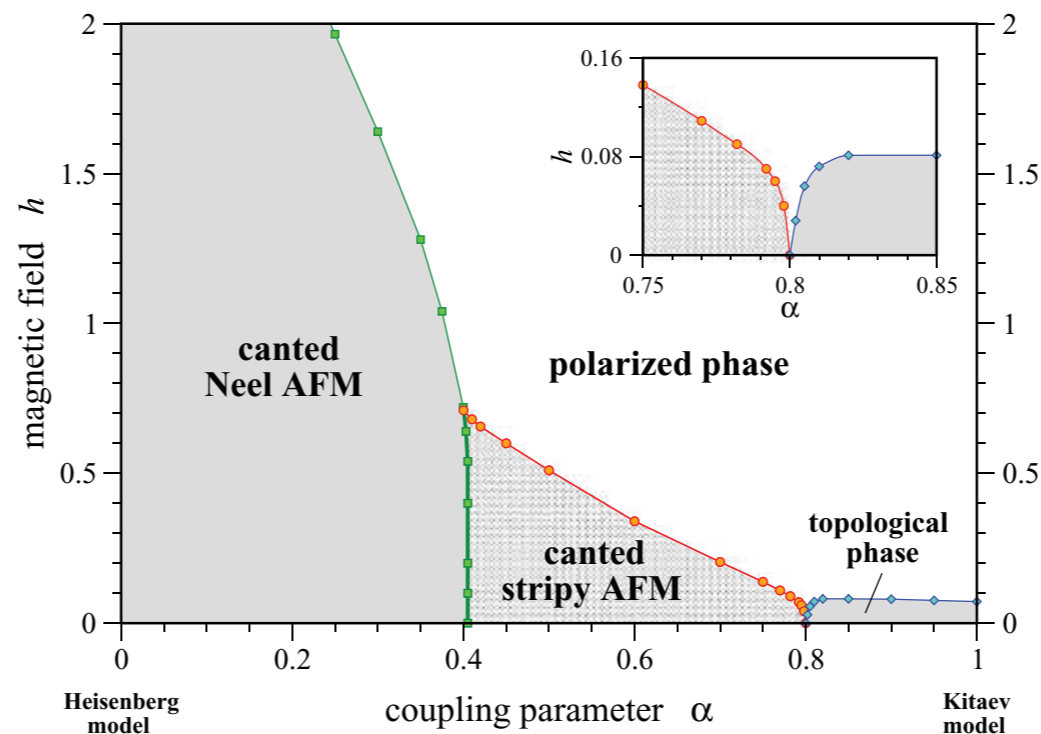
Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

Classical phase diagram in $[111]$ field:

[LJ, Andrade, Vojta, PRL '16]



DMRG:



$$J_1 = 1 - \alpha$$

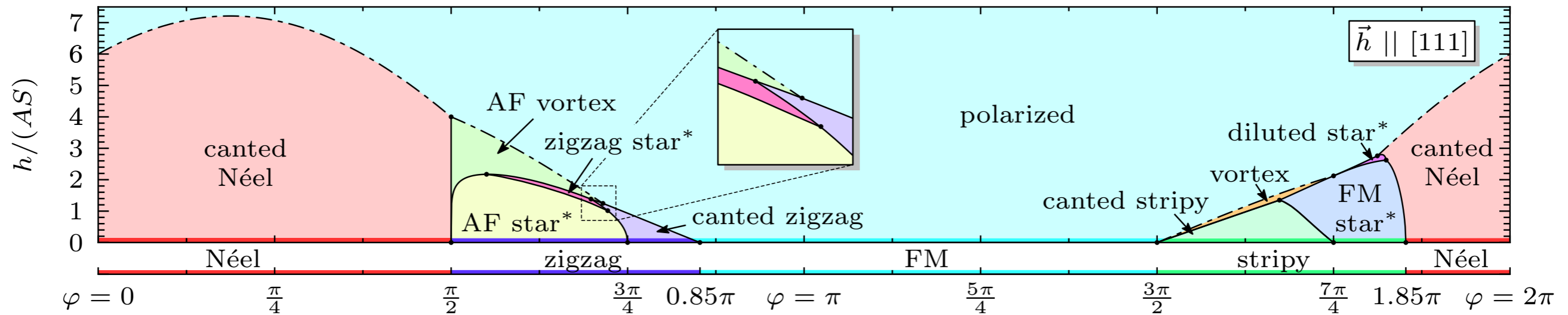
$$K_1 = -2\alpha$$

[Jiang, Gu, Qi, Trebst, PRB '11]

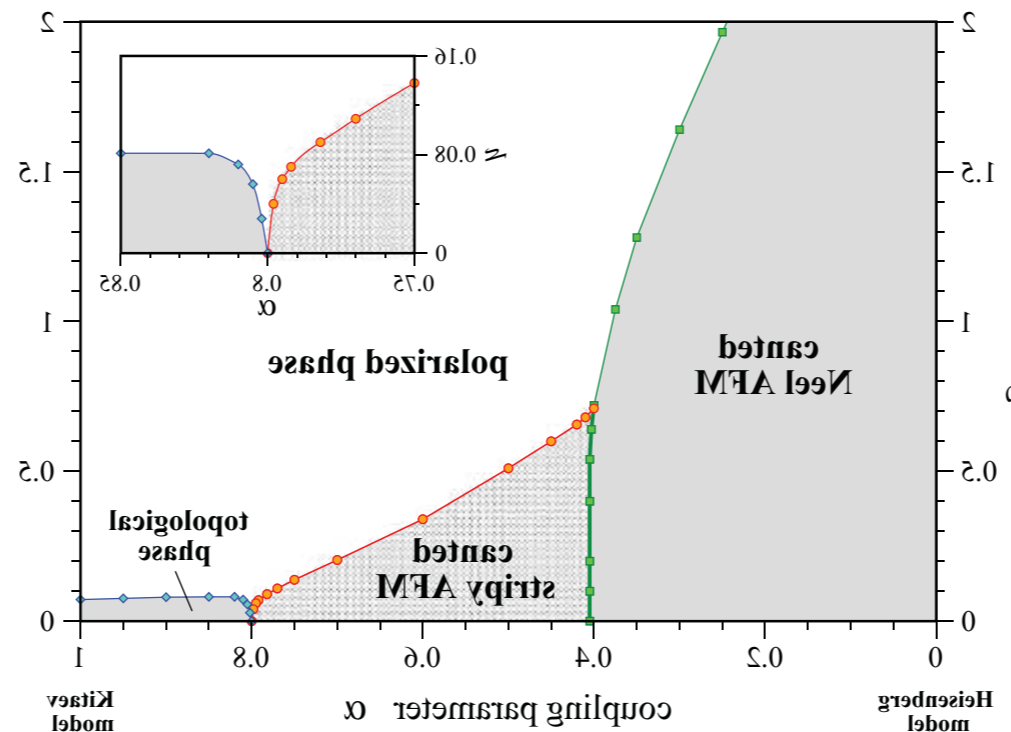
Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

Classical phase diagram in [111] field:

[LJ, Andrade, Vojta, PRL '16]



DMRG:



$$J_1 = 1 - \alpha$$

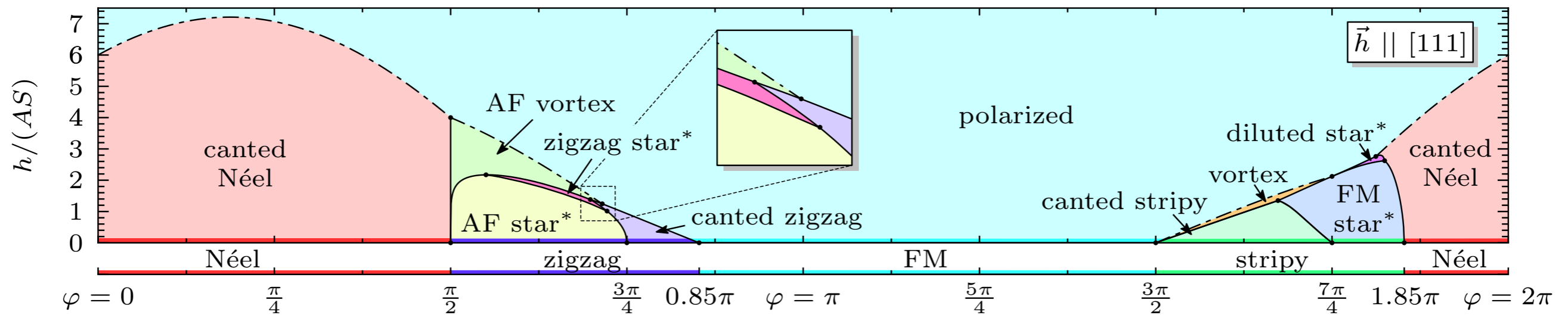
$$K_1 = -2\alpha$$

[Jiang, Gu, Qi, Trebst, PRB '11]

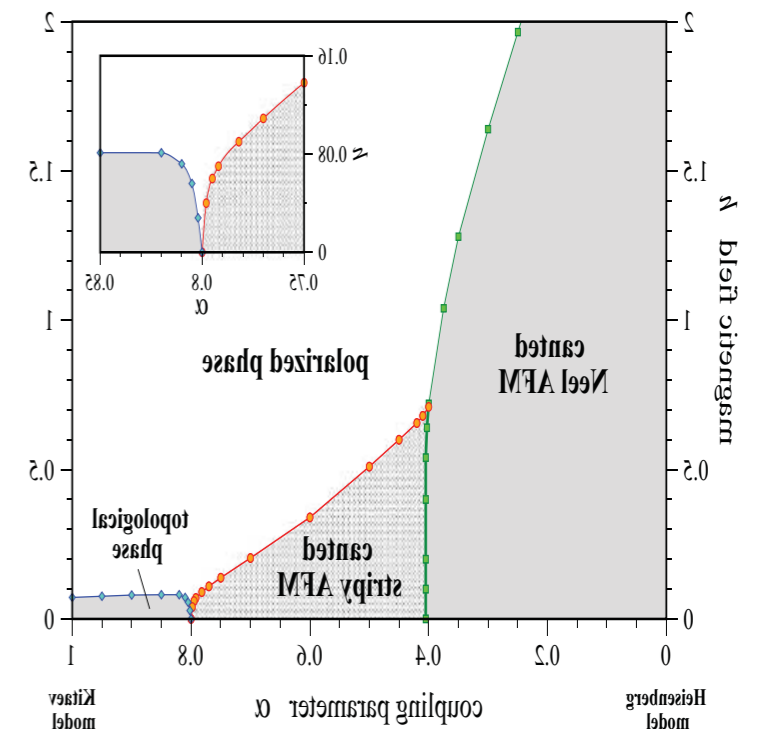
Scenario 1 ($K_1 > 0, J_1 < 0$) in field: $\vec{h} \parallel [111] \parallel c^*$

Classical phase diagram in $[111]$ field:

[LJ, Andrade, Vojta, PRL '16]



DMRG:



[Jiang, Gu, Qi, Trebst, PRB '11]

Summary: Scenario 1 ($K_1 > 0, J_1 < 0$)

Magnetization process ...

... strongly **anisotropic**

... typically no simple canting

... exotic **intermediate** phases

... but only weak angle dependence of susceptibility and h_c

... does not explain anisotropy in α -RuCl₃

... unless field along high-symmetry direction

... large unit cell, multi- \vec{Q} , vortex

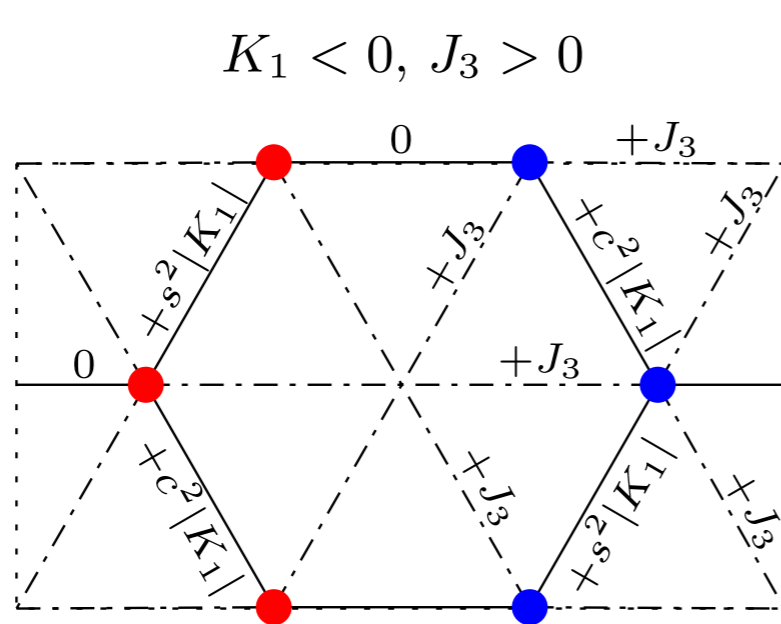
Scenario 2: Ferromagnetic K_1 , antiferromagnetic J_3

Minimal model:

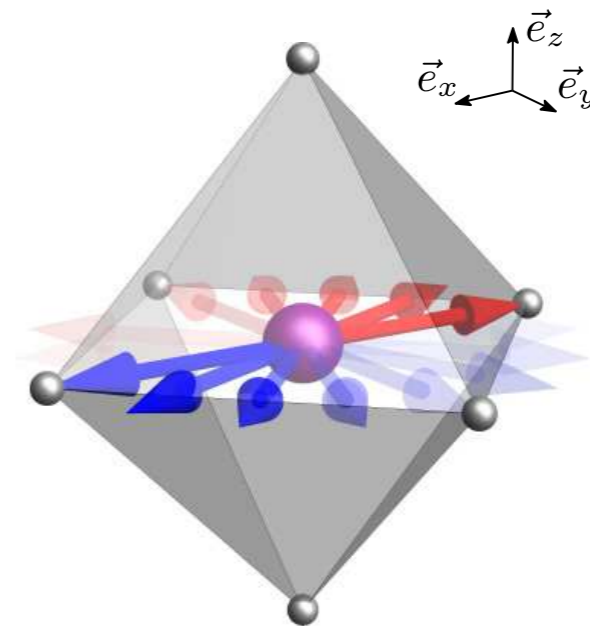
[Winter *et al.*, PRB '16]

$$\mathcal{H} = \sum_{\langle ij \rangle} K_1 S_i^\gamma S_j^\gamma + \sum_{\langle\langle ij \rangle\rangle} J_3 \vec{S}_i \cdot \vec{S}_j$$

Zero field:



“cubic-planes zigzag”



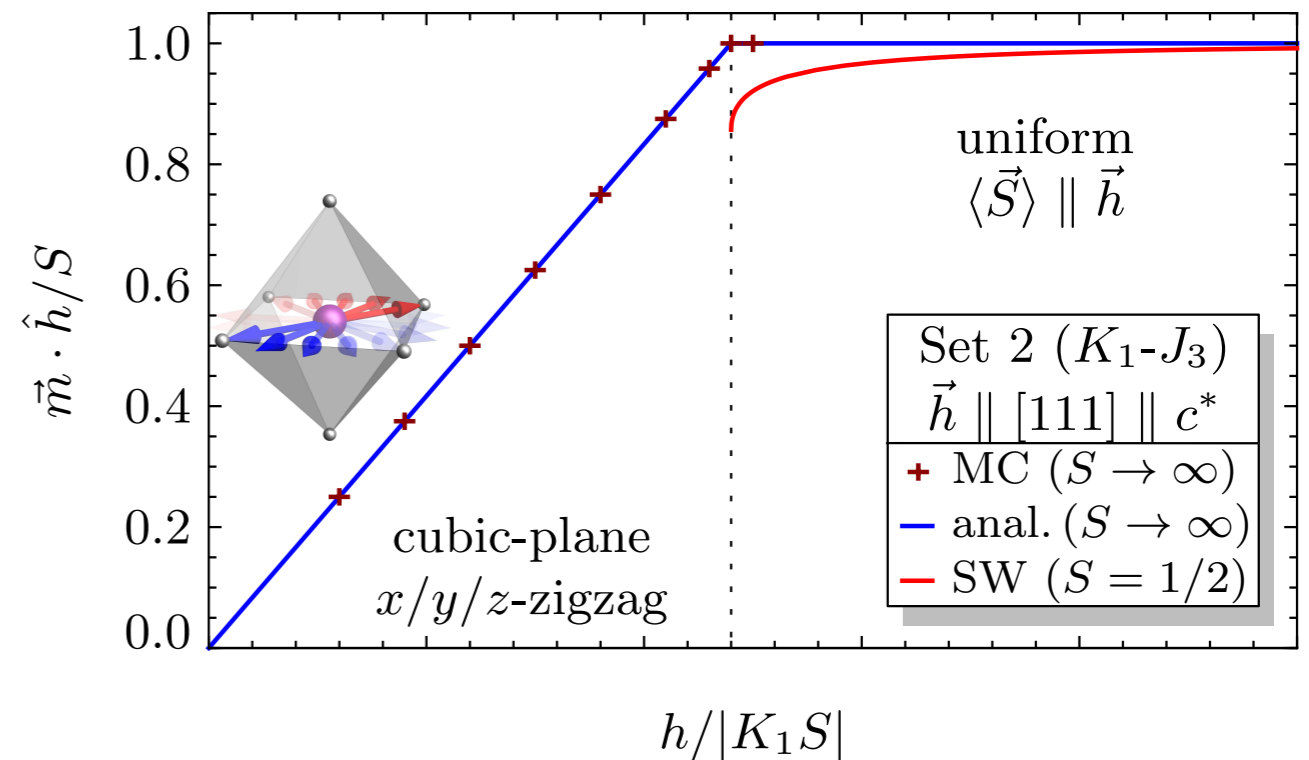
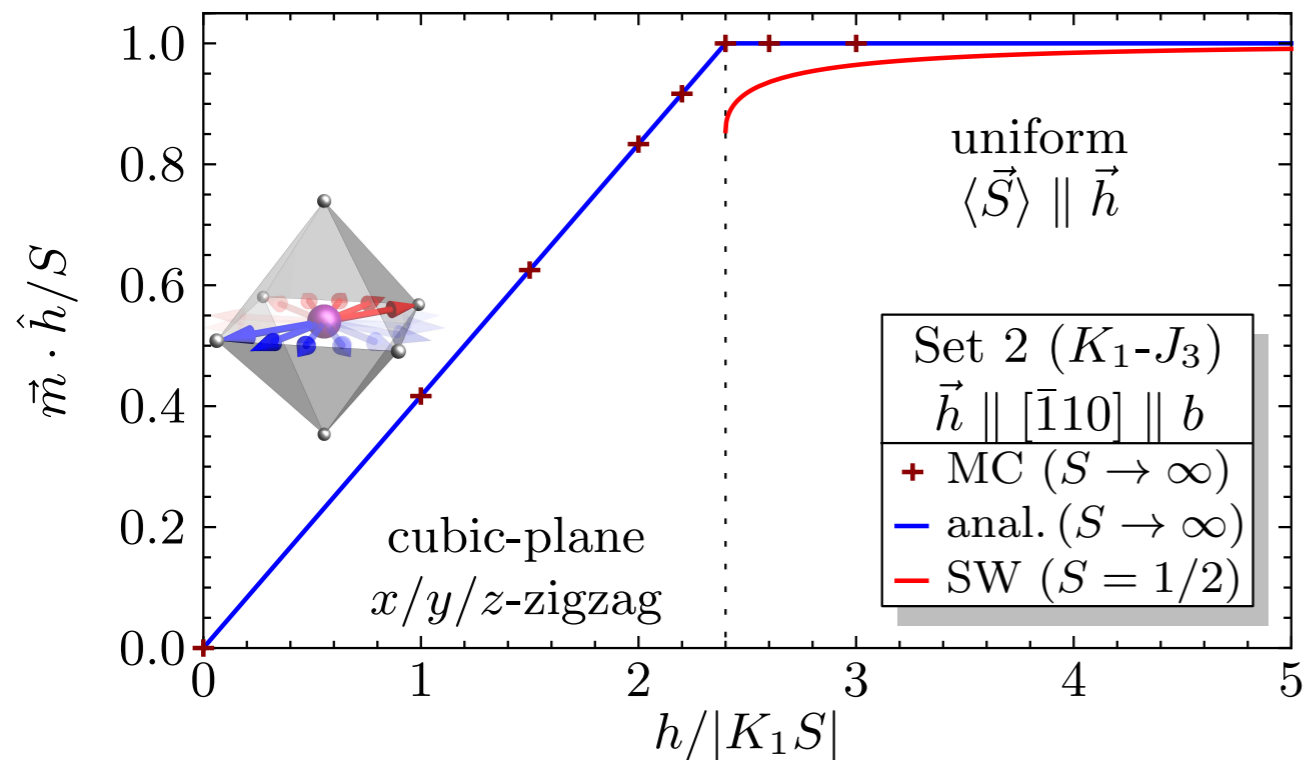
$$\vec{S} \parallel \pm(c\vec{e}_x + s\vec{e}_y), \quad c^2 + s^2 = 1$$

... i.e., within Ru_2Cl_2 planes

... degeneracy will be lifted in real materials

Scenario 2 ($K_1 < 0, J_3 > 0$) in field

For $\Gamma_1 = 0$:



$(K_1, J_3) = (-17, +6.8)$ meV

[Winter *et al.*, PRB '16]

... classical magnetization curve **independent** of field direction!

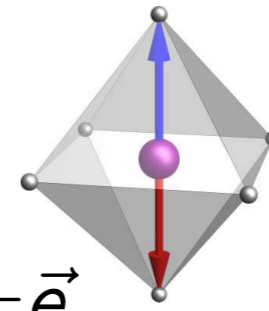
... and angle dependence of quantum corrections very weak

Scenario 2 ($K_1 < 0, J_3 > 0$) in field: Perturbations

$\mathbb{Z}_3 \times O(2)$ degeneracy lifted by:

(i) quantum fluctuations:

$$\vec{S}_i \parallel \pm \vec{e}_x, \pm \vec{e}_y, \pm \vec{e}_z$$

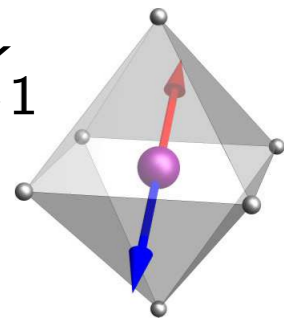


[Sizyuk *et al.*, PRB '16]
[Chaloupka *et al.*, PRB '16]

“cubic-axes zigzag”

(ii) off-diagonal $\Gamma_1 > 0$:

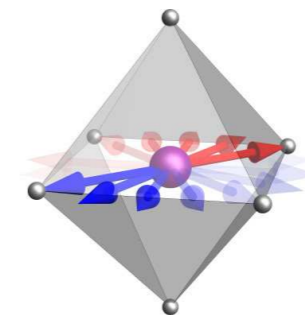
$$\vec{S}_i \parallel \begin{cases} \pm[110] & \text{or symmetry-equivalent,} \\ \pm[11\bar{1}] & \text{or symmetry-equivalent,} \end{cases} \quad \begin{array}{l} \text{if } 0 < \Gamma_1 \ll K_1 \\ \text{if } \Gamma_1 \gg K_1 \end{array}$$



“face-center zigzag”

(iii) magnetic field:

$$\vec{S}_i \perp \vec{h}$$



“cubic-plane zigzag”

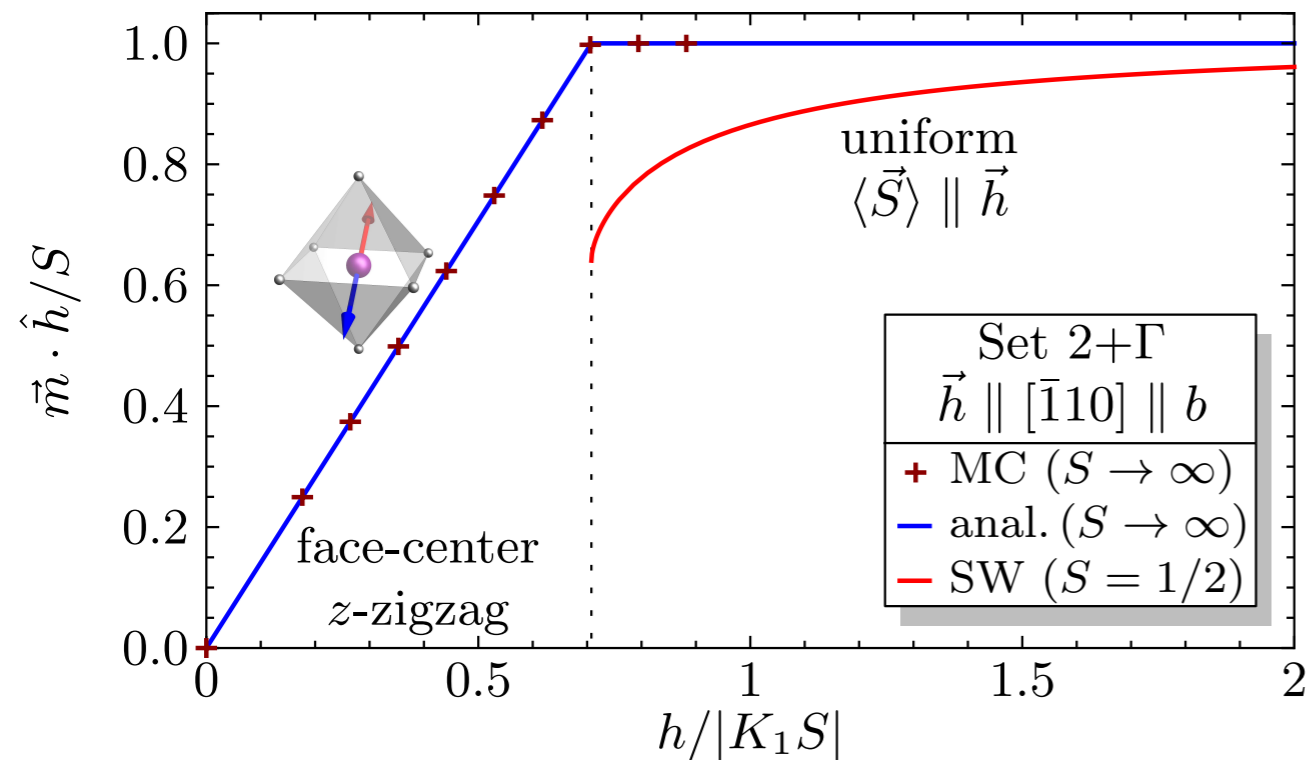
\Rightarrow **competition** between different effects

... can be tuned by h

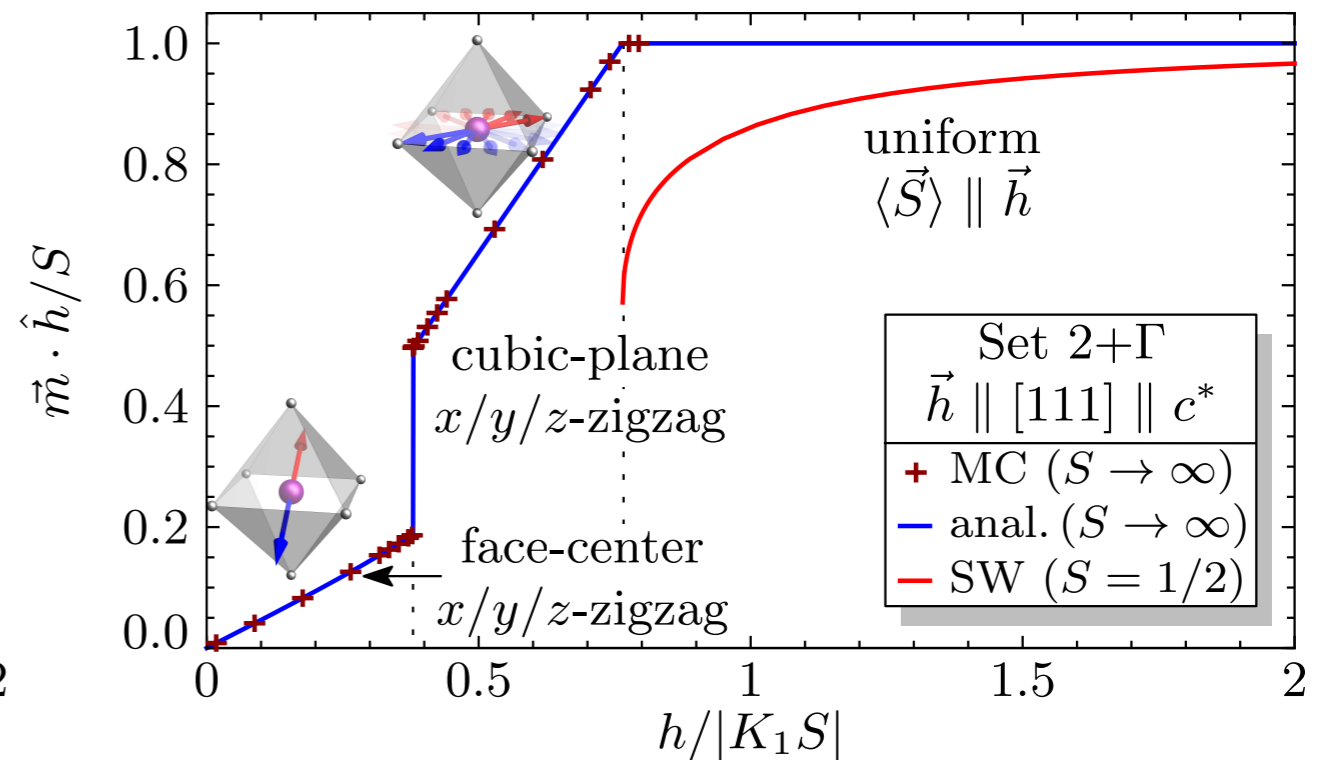
Scenario 2 ($K_1 < 0, J_3 > 0$) in field: Perturbations

Metamagnetic **transitions** between canted **zigzag** states:

$$\vec{h} \parallel [\bar{1}10] \parallel b$$



$$\vec{h} \parallel [111] \parallel c^*$$



$$(J_1, K_1, \Gamma_1, J_2, K_2, J_3) = (+3, -17, +1, -3, +6, +1) \text{ meV}$$

[Sizyuk *et al.*, PRB '16]

Summary: Scenario 2 ($K_1 < 0$, $J_3 > 0$)

Magnetization process ...

... mostly **isotropic**

... very weak angle dependence of susceptibility and h_c

... does not explain anisotropy in α -RuCl₃

... **transitions** between different canted **zigzag** states possible

... for some field directions

... **no** exotic intermediate phases

... high-field transition **continuous**

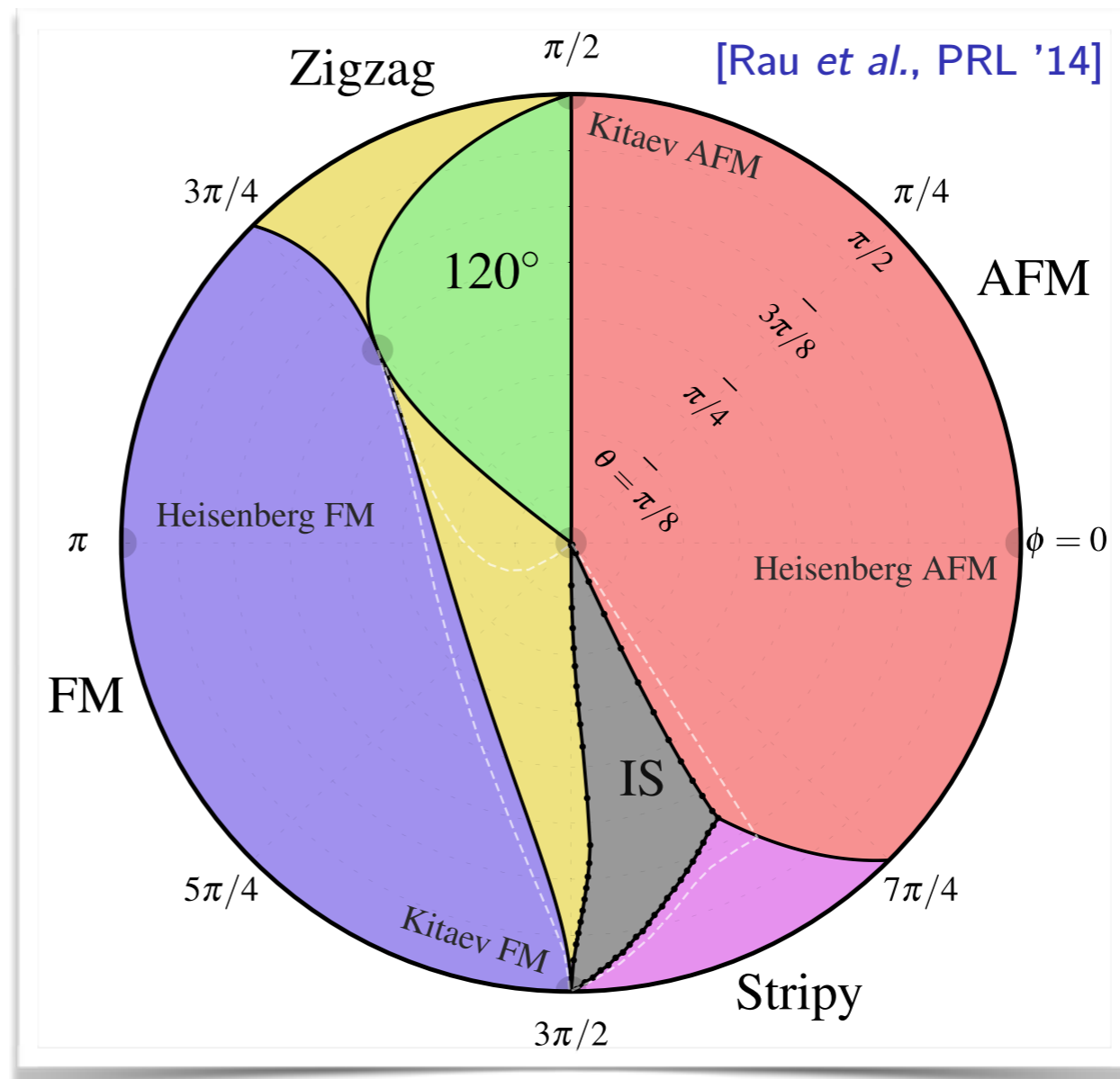
Scenario 3: Ferromagnetic K_1 , positive Γ_1

Minimal model:

[Ran et al., PRL '17]

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right]$$

Zero-field phase diagram?



Scenario 3: Ferromagnetic K_1 , positive Γ_1

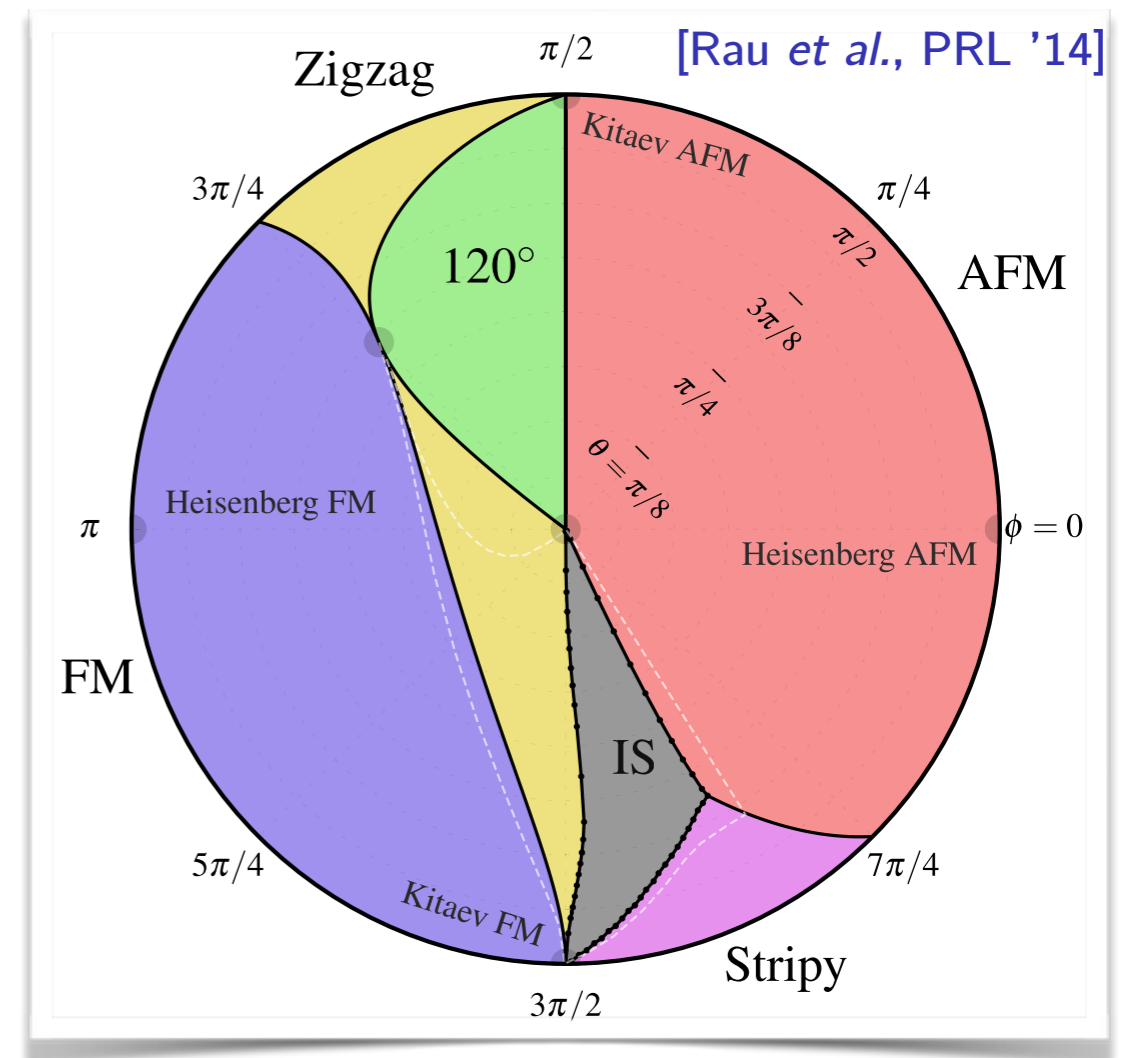
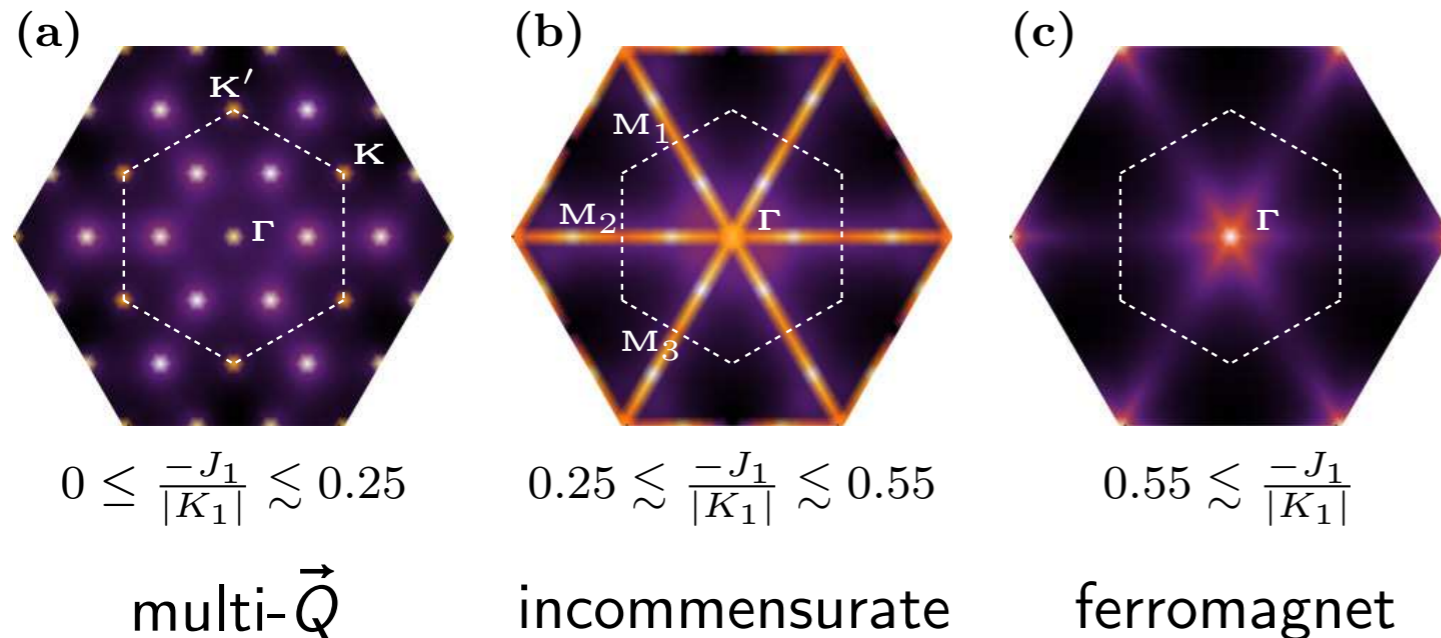
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Zero-field phase diagram?

$(K_1, \Gamma_1) = (-6.8, +9.5)$ meV
[Ran et al., PRL '17]



... failure of Luttinger-Tisza

Scenario 3: Ferromagnetic K_1 , positive Γ_1

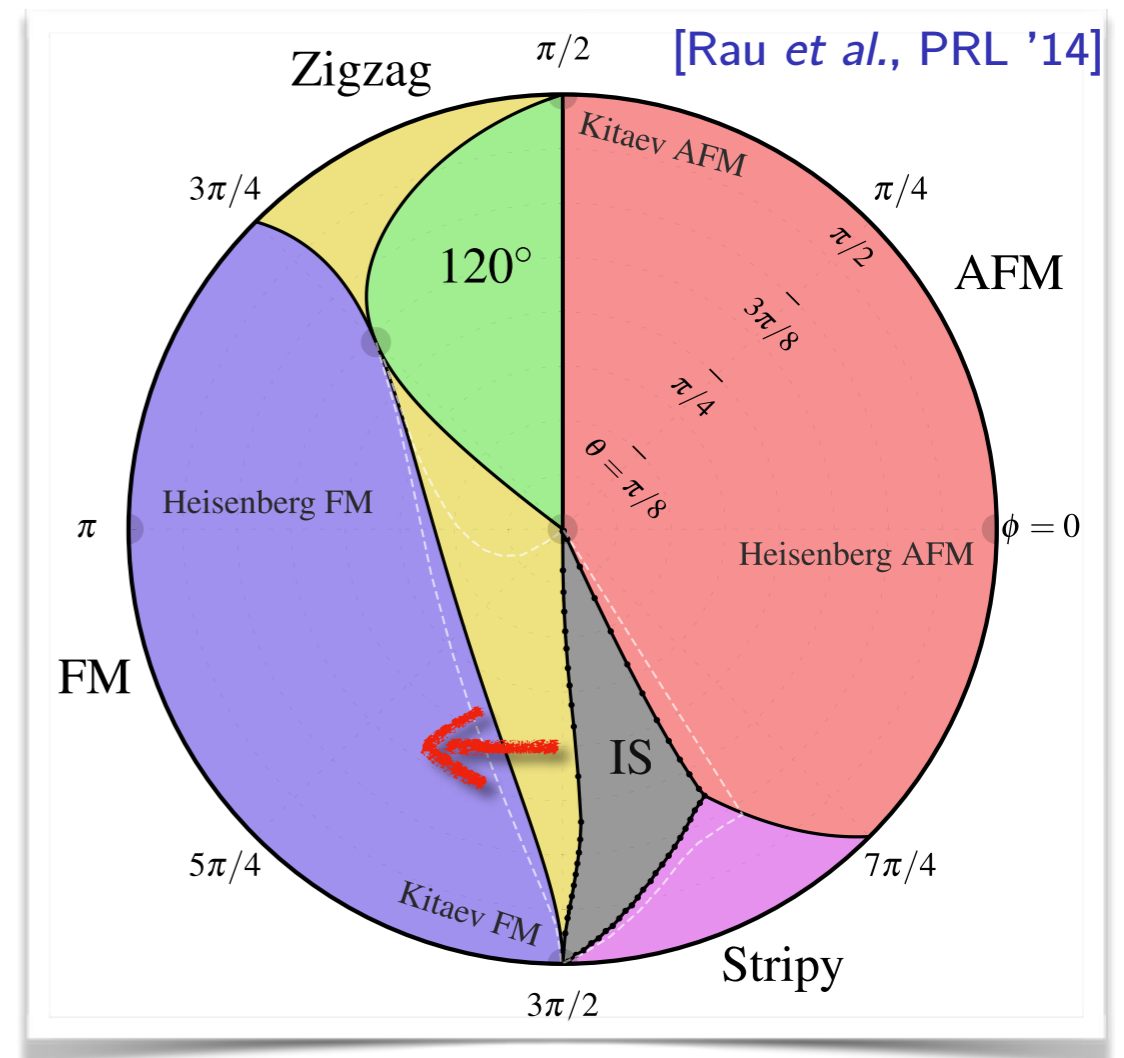
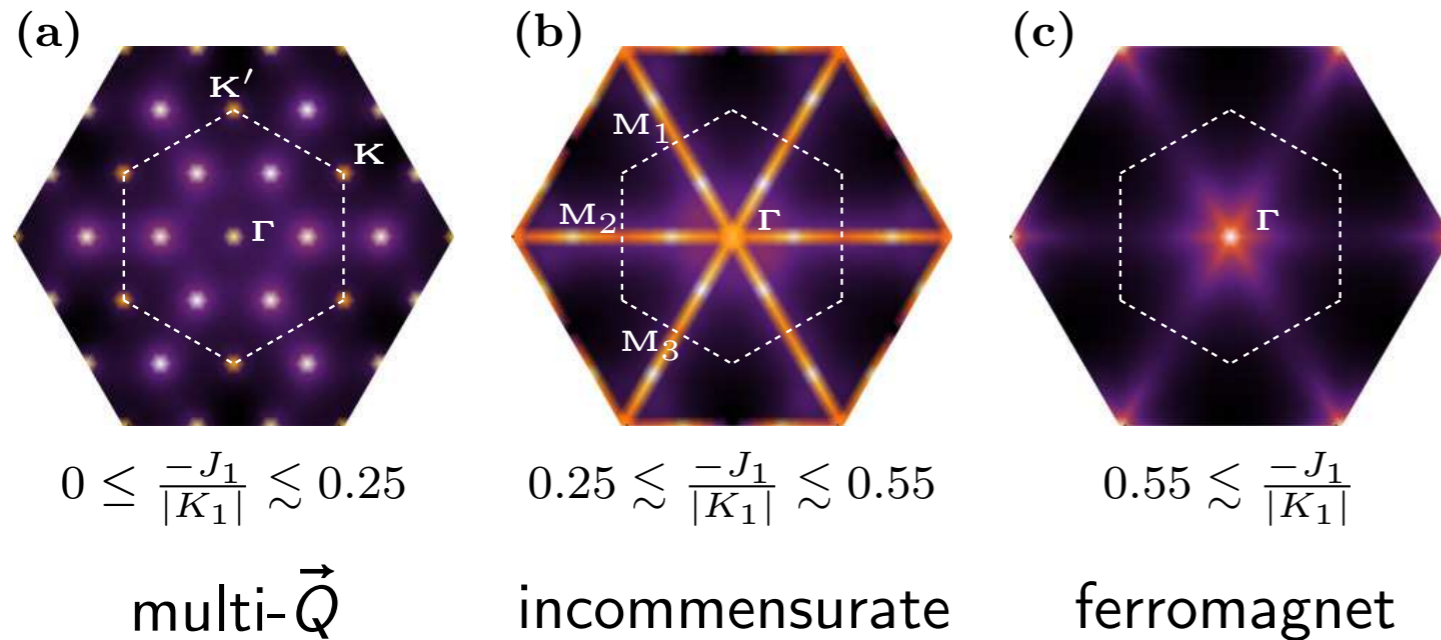
Minimal model:

[Ran et al., PRL '17]

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right]$$

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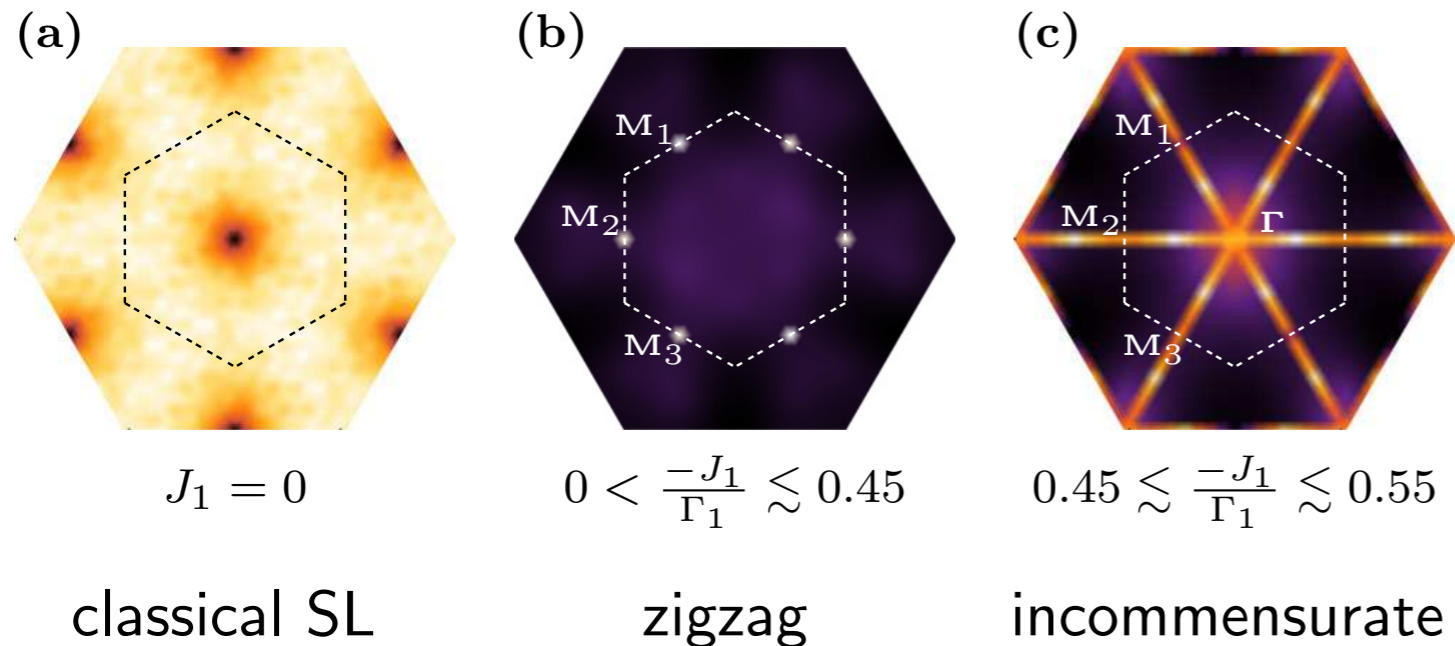
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Minimal model:

[Ran et al., PRL '17]

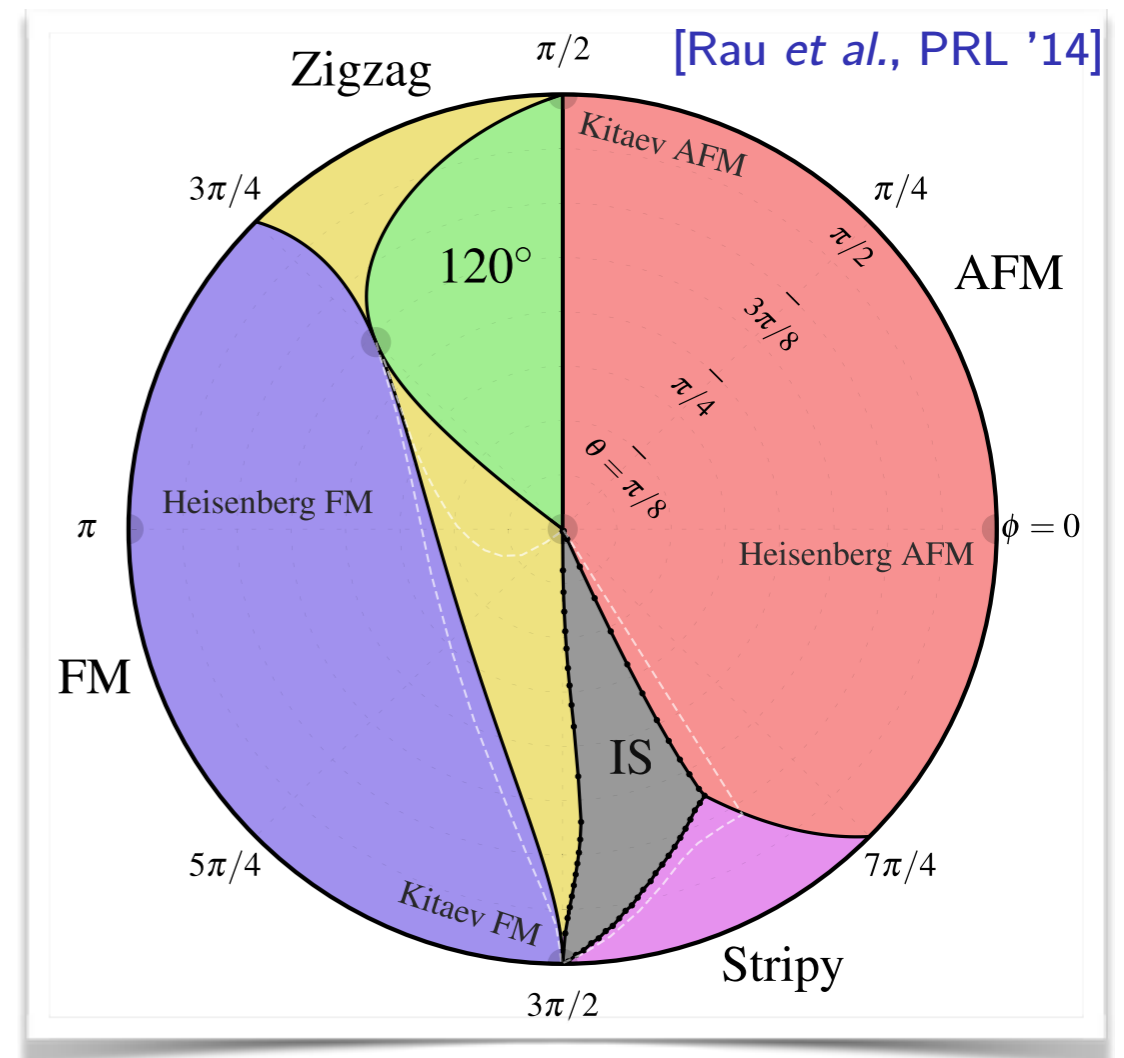
$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right]$$

Zero-field phase diagram?



[Rousochatzakis & Perkins, PRL '17]

... **zigzag** can be recovered for $\Gamma_1 \gg |K_1|$



... failure of Luttinger-Tisza

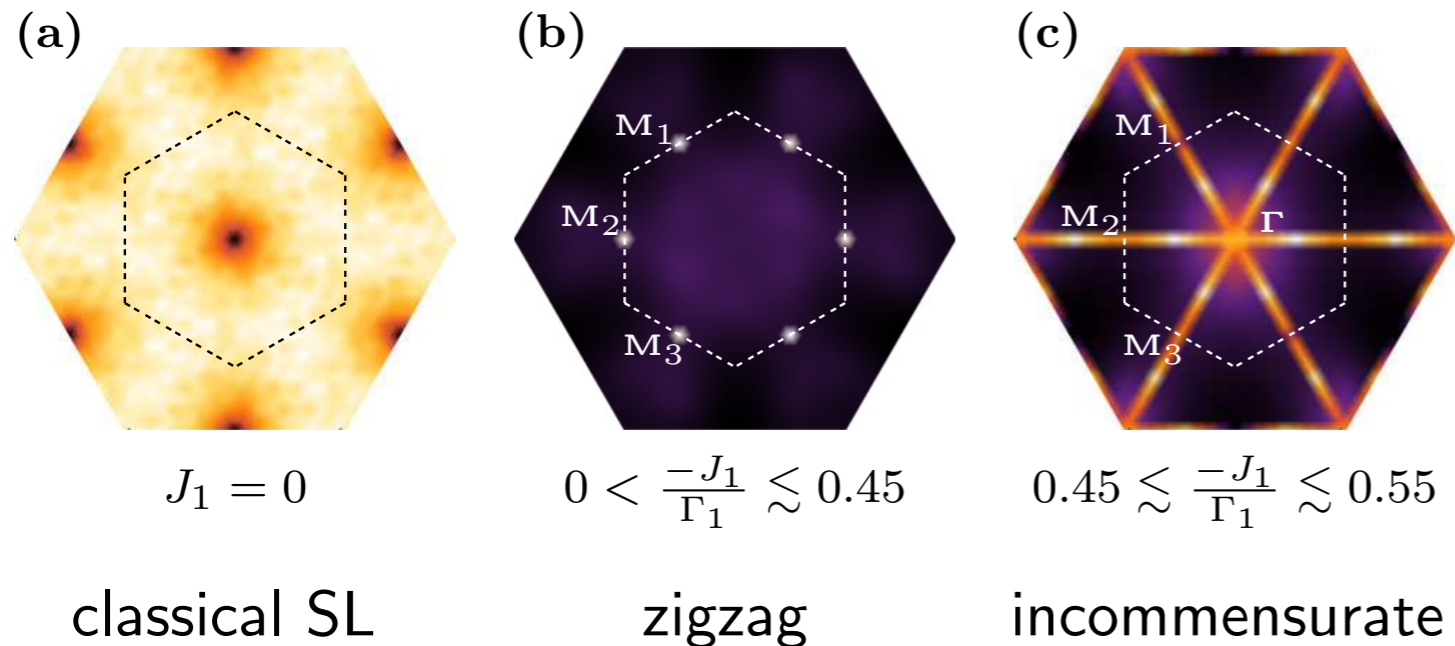
Scenario 3: Ferromagnetic K_1 , positive Γ_1

Minimal model:

[Ran et al., PRL '17]

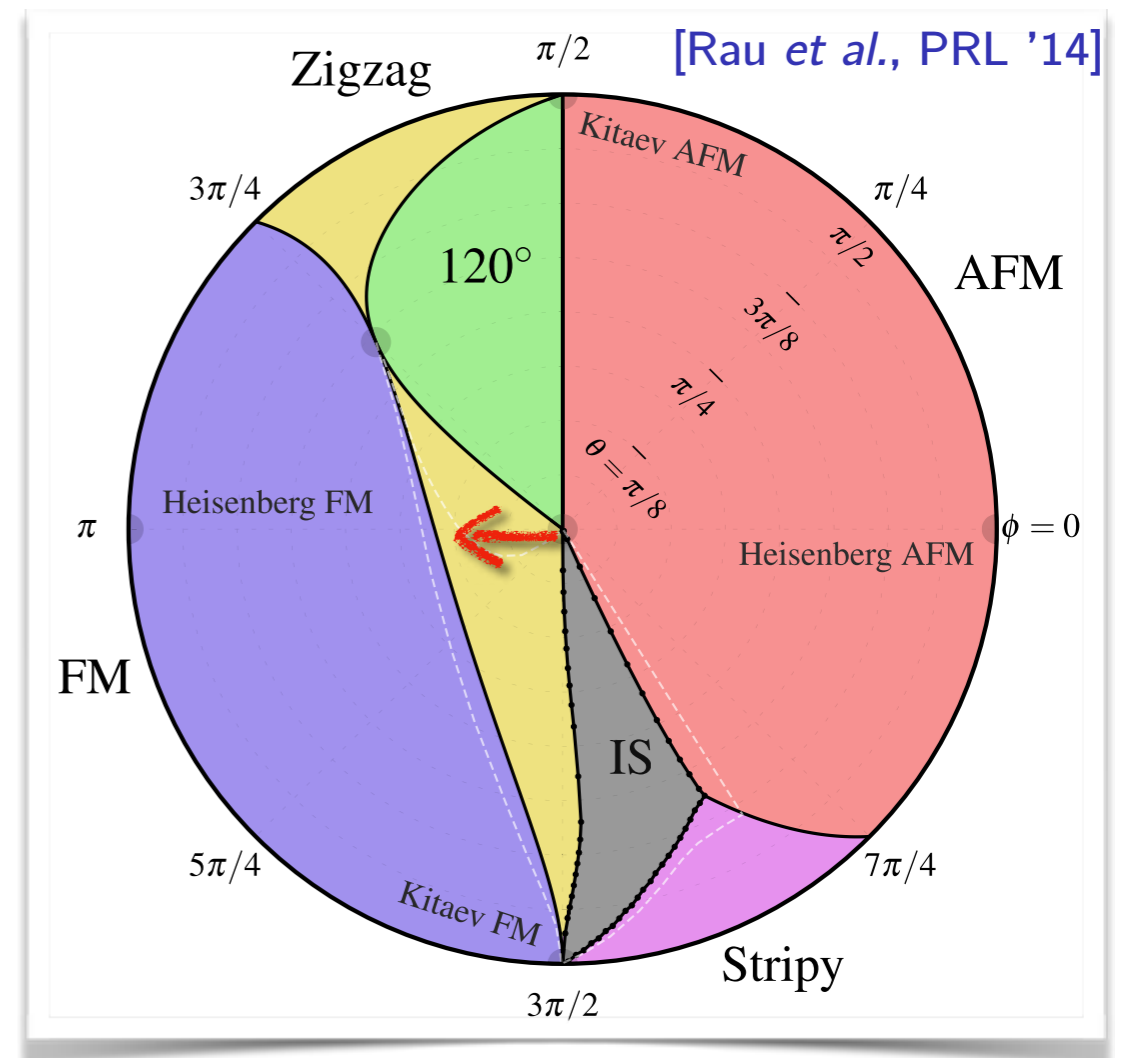
$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right]$$

Zero-field phase diagram?



[Rousochatzakis & Perkins, PRL '17]

... **zigzag** can be recovered for $\Gamma_1 \gg |K_1|$



... failure of Luttinger-Tisza

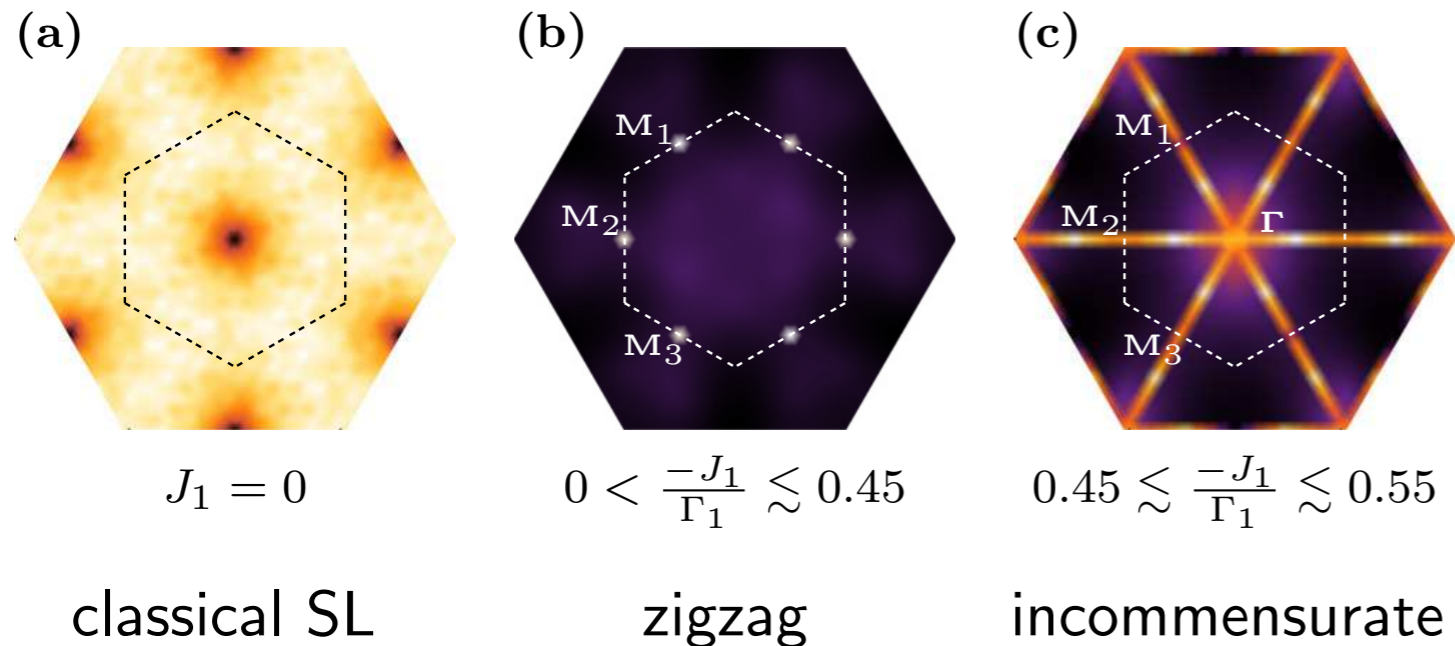
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Minimal model:

[Ran et al., PRL '17]

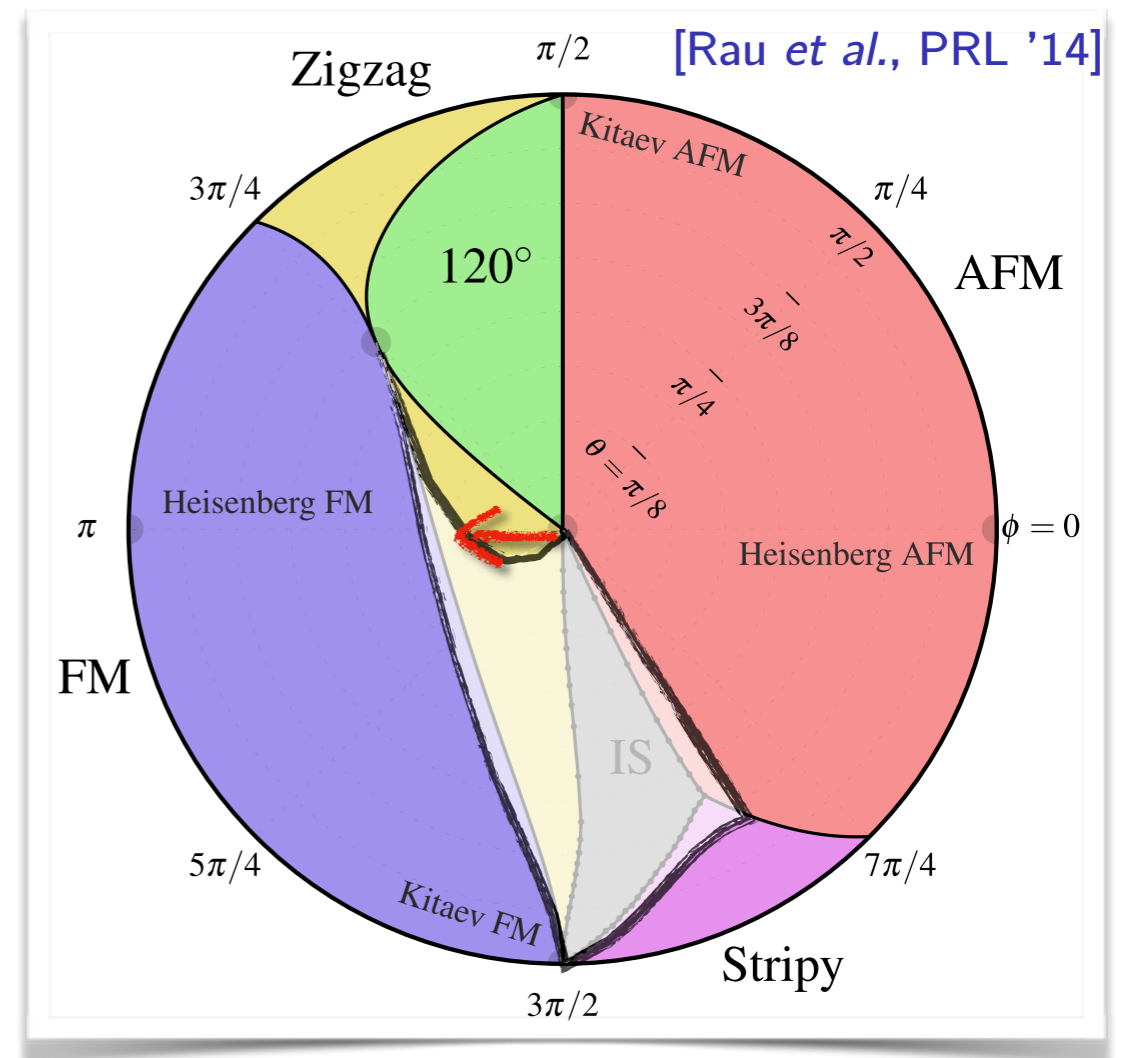
$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right]$$

Zero-field phase diagram?



[Rousochatzakis & Perkins, PRL '17]

... **zigzag** can be recovered for $\Gamma_1 \gg |K_1|$



... failure of Luttinger-Tisza

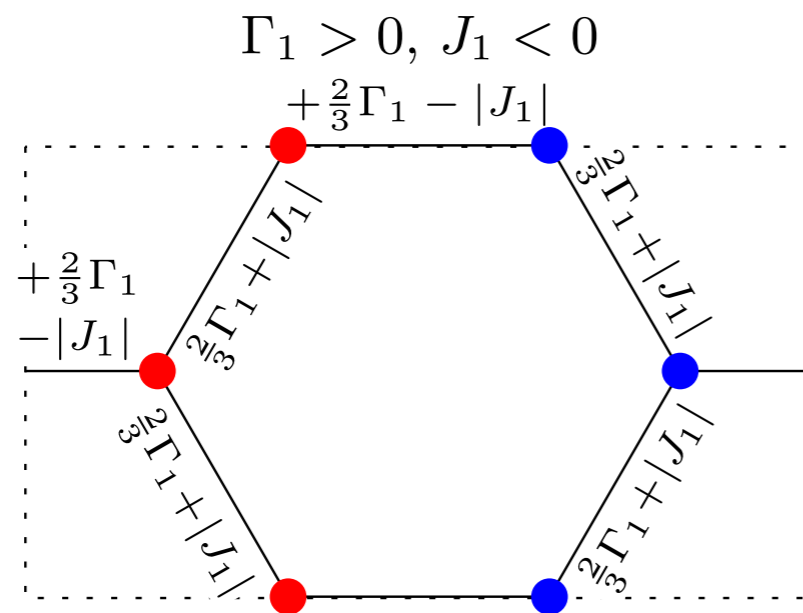
Scenario 3: Ferromagnetic K_1 , J_1 , positive Γ_1

Minimal model:

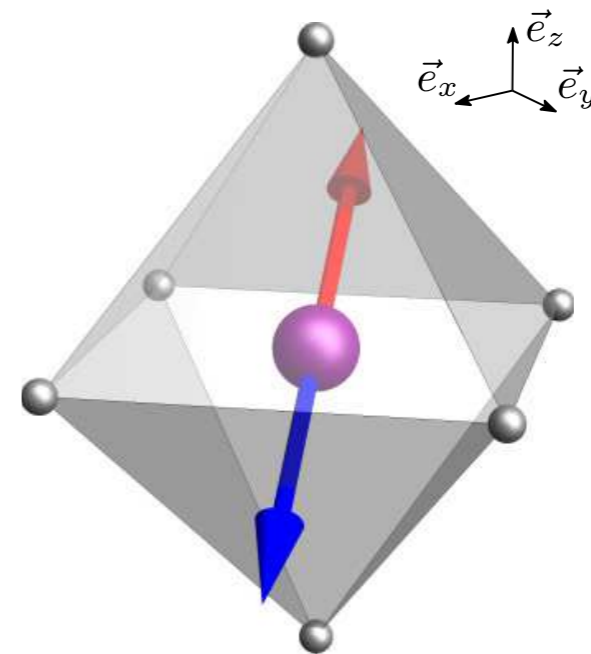
$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1 \vec{S}_i \cdot \vec{S}_j + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right]$$

Zero field:

... i.e., take the limit $K_1/\Gamma_1 \rightarrow 0$



“face-center zigzag”



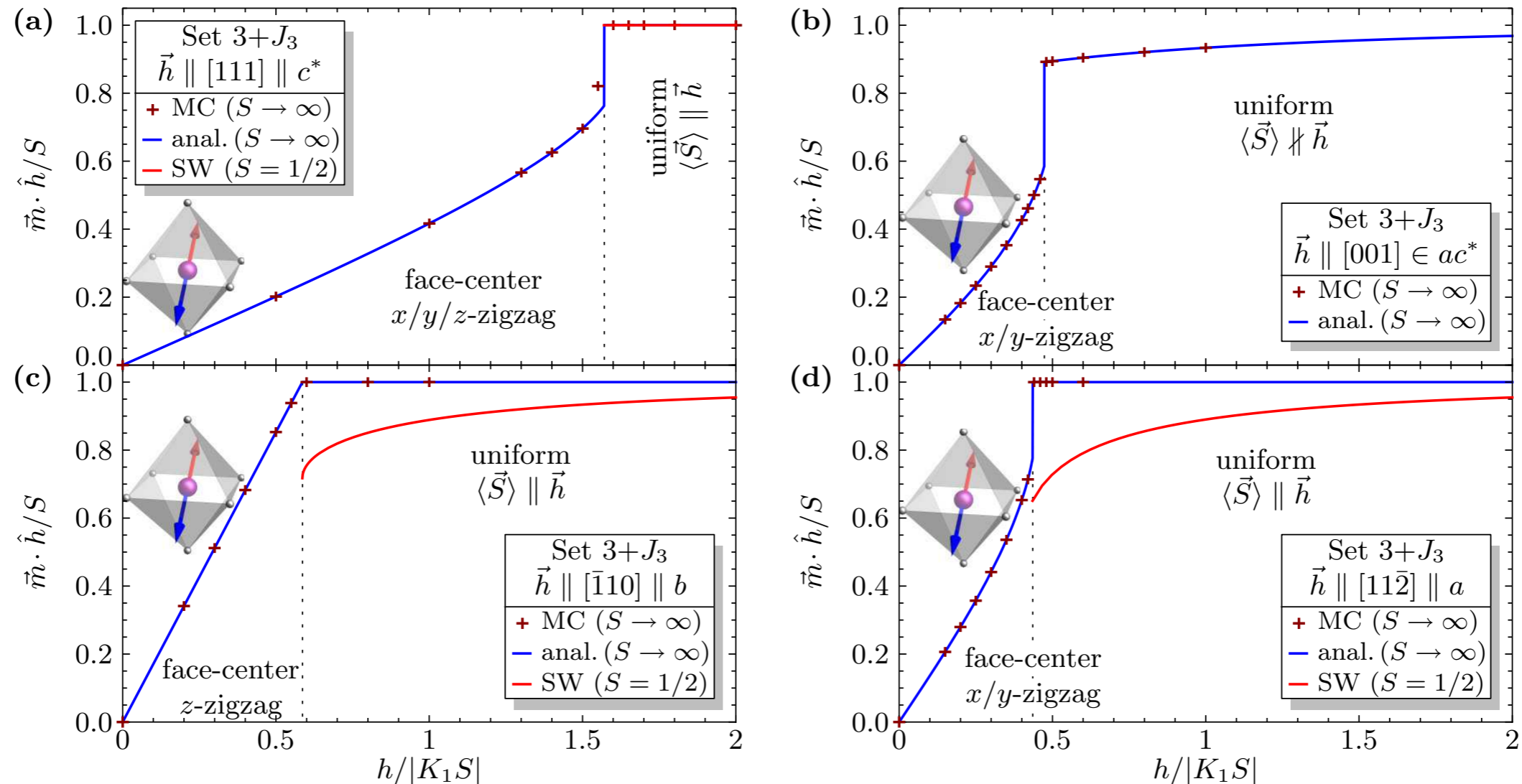
$$\vec{S} \parallel \pm [11\bar{1}]$$

... towards faces of RuCl_6 octahedra

Scenario 3: Ferromagnetic K_1 , J_1 , positive Γ_1

$(J_1, K_1, \Gamma_1, J_3) = (-0.5, -5.0, +2.5, +0.5)$ meV

[Winter *et al.*, Nat. Comm. '17]



Observations:

(1) Out-of-plane h_c much larger than in-plane h_c

... as in α -RuCl₃

(2) $|\langle \vec{m}_{\parallel} \rangle| / S < 1$ in polarized phase already in classical limit

Scenario 3 ($K_1, J_1 < 0, \Gamma_1 > 0$): Consequences of strong Γ_1

Toy model:

$$\mathcal{H} = \sum_{\langle ij \rangle} \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)$$

In polarized state $\vec{S}_i \equiv S$

$$\langle E_0 / (NS^2) \rangle = \begin{cases} +\Gamma_1 & \text{if } \vec{S}_i \parallel [111] \parallel c^* \\ -\Gamma_1/2 & \text{if } \vec{S}_i \parallel [\bar{1}10] \parallel b \\ -\Gamma_1/2 & \text{if } \vec{S}_i \parallel [11\bar{2}] \parallel a \end{cases}$$

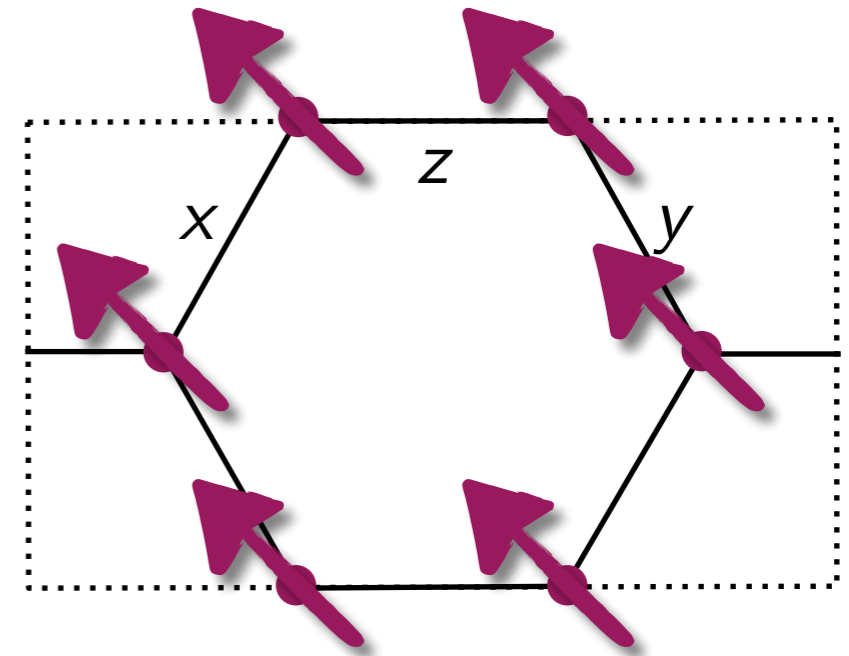
... similarly: components of \vec{S}_i in out-of-plane vs. in-plane direction

$\Rightarrow \Gamma_1 > 0$ means ...

... “antiferromagnetic” for **out-of-plane** spins

... “ferromagnetic” for **in-plane** spins

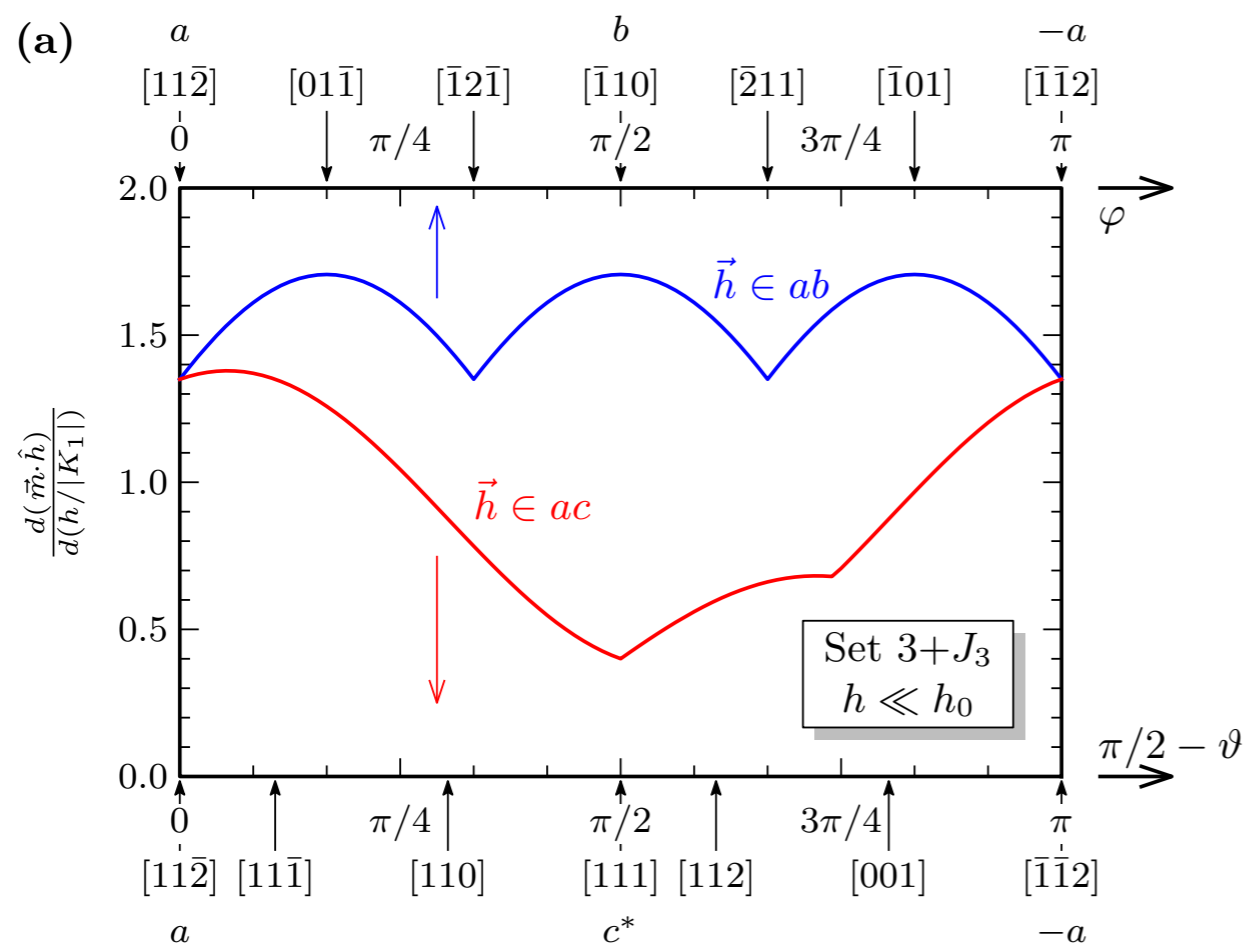
... naturally explains in-plane vs. out-of-plane anisotropy



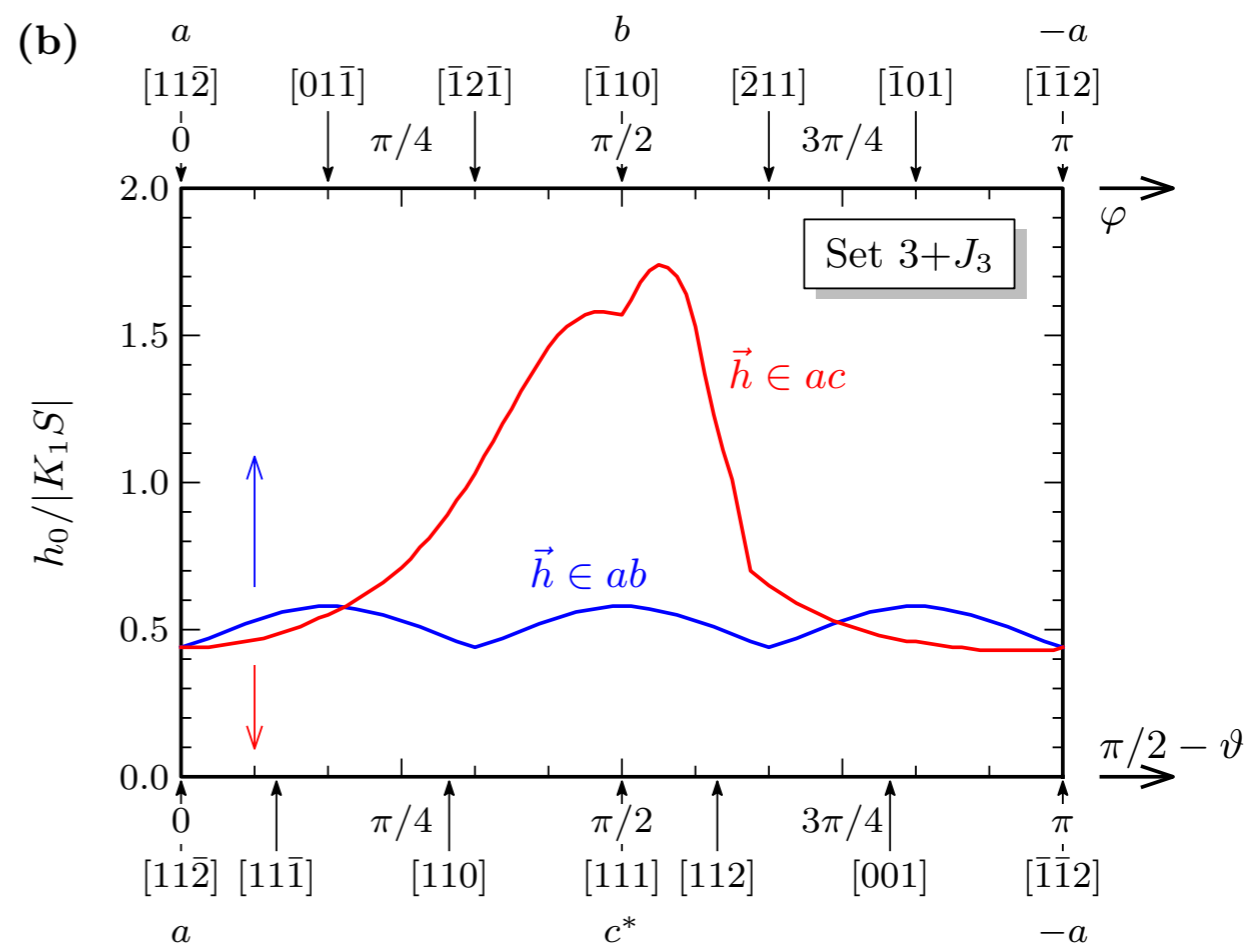
Scenario 3 ($K_1, J_1 < 0, \Gamma_1 > 0$): Field-angle dependence

[LJ, Andrade, Vojta, PRB '17]

Susceptibility:



Critical field h_c :



$(J_1, K_1, \Gamma_1, J_3) = (-0.5, -5.0, +2.5, +0.5)$ meV

... immediately testable in α -RuCl₃

Scenario 3 ($K_1, J_1 < 0, \Gamma_1 > 0$): High-field transversal magnetization

High fields: expect ground state **w/o** spontaneous symmetry breaking

... i.e., expect high-field state to be adiabatically connected to fully polarized state

If $\Gamma_1 = 0$: $S_i^x \mapsto -S_i^x, \quad S_i^y \mapsto -S_i^y, \quad S_i^z \mapsto -S_i^z$

... is an accidental higher symmetry

For $\vec{h} \parallel [001]$:

(a) $\Gamma_1 = 0$: $\langle \vec{S}_i \rangle \parallel [001] \Rightarrow |\langle \vec{m}_{\parallel} \rangle| / S = 1$

(b) $\Gamma_1 \neq 0$: $\langle \vec{S}_i \rangle \parallel [xxz]$ with $x \neq 0$ not forbidden

\Rightarrow finite $\langle \vec{m}_{\perp} \rangle \perp \vec{h}$ possible

\vec{m}_{\perp} is a direct measure of Γ_1

... independent of K_1, J_1, J_3, \dots

$$|\langle \vec{m}_{\perp} \rangle| / |\langle \vec{m}_{\parallel} \rangle| \simeq 0.51$$

at $h = h_c^+$ for $(J_1, K_1, \Gamma_1, J_3) = (-0.5, -5.0, +2.5, +0.5)$ meV

Summary: Scenario 3 ($K_1, J_1 < 0, \Gamma_1 > 0$)

Magnetization process ...

... strongly **anisotropic**

... χ_{ab} much larger than χ_c

... even without g -factor anisotropy

... $\chi_{ab}/\chi_c \approx 3 \dots 4$ for $(J_1, K_1, \Gamma_1, J_3) = (-0.5, -5.0, +7.0, +0.5)$ meV

... consistent with small trigonal distortion [Agrestini *et al.*, PRB(R) '17]

... high-field state with transversal magnetization

... for field in intermediate direction, e.g., [001]

Conclusions:

Magnetization process crucially dependent on zigzag stabilization:

- Scenario 1 ($K_1 > 0, J_1 < 0$)
 - ▶ Weak anisotropy in χ and h_c
 - ▶ Various exotic intermediate phases
- Scenario 2 ($K_1 < 0, J_3 > 0$)
 - ▶ Nearly no anisotropy in χ and h_c
 - ▶ Metamagnetic transitions between different canted zigzag
- Scenario 3 ($K_1, J_1 < 0, \Gamma_1 > 0$)
 - ▶ Strong anisotropy in χ and h_c ... natural explanation for anisotropy in α -RuCl₃
 - ▶ Finite transversal magnetization in high-field state ... possibly measurable?
 - ▶ Intermediate phases?

α -RuCl₃: expect strong $\Gamma_1 > 0, K_1 < 0$, and small $J_3 > 0$