# Generic spin model for the honeycomb iridates with trigonal distortion

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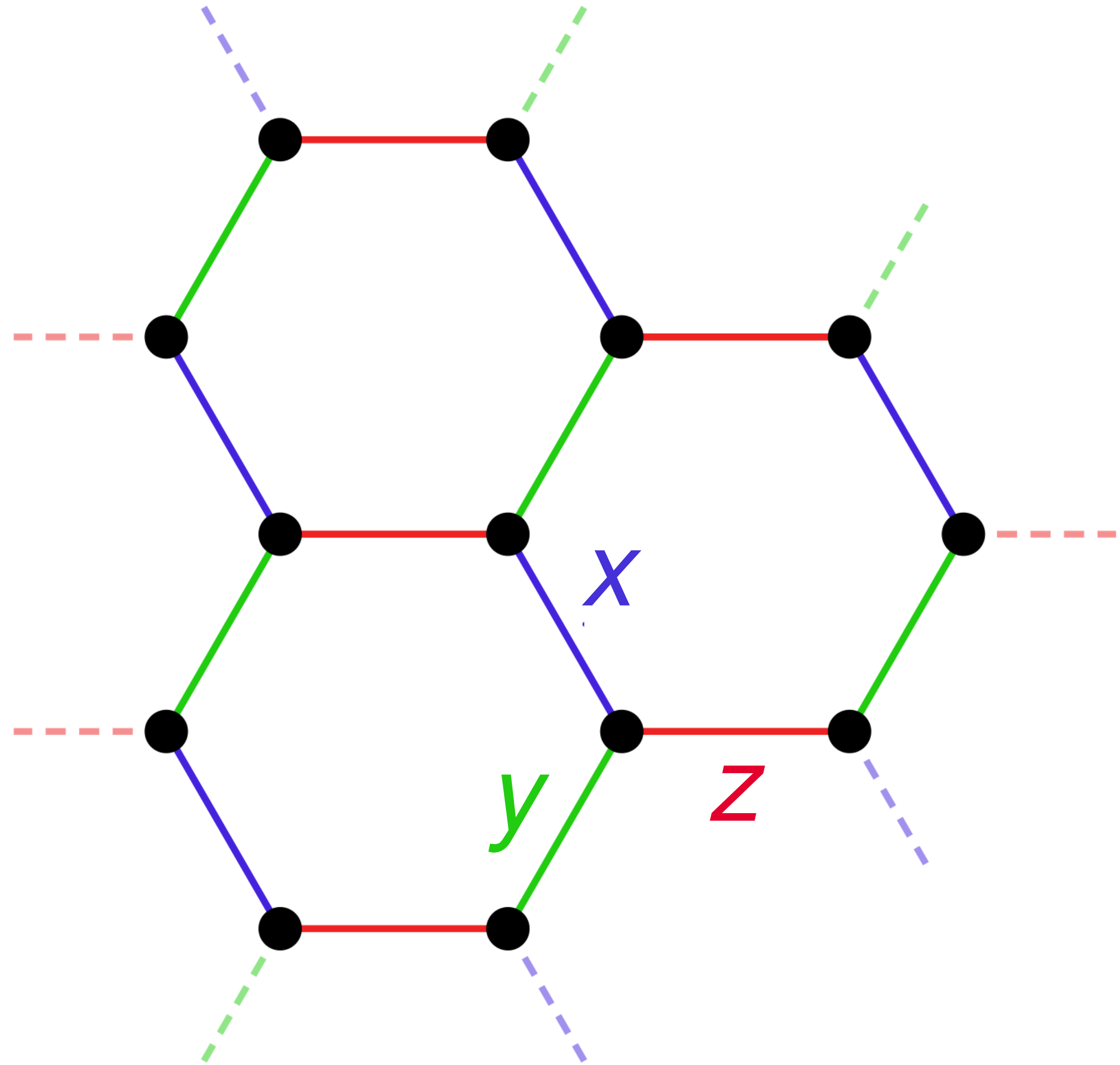
# Kitaev's honeycomb model

- Spin-½ model
- Highly *anisotropic* in spin and space
- Frustrated by anisotropy, not geometry

Kitaev Exchange

$$K \sum_{\langle ij \rangle \in \gamma} S_i^\gamma S_j^\gamma$$

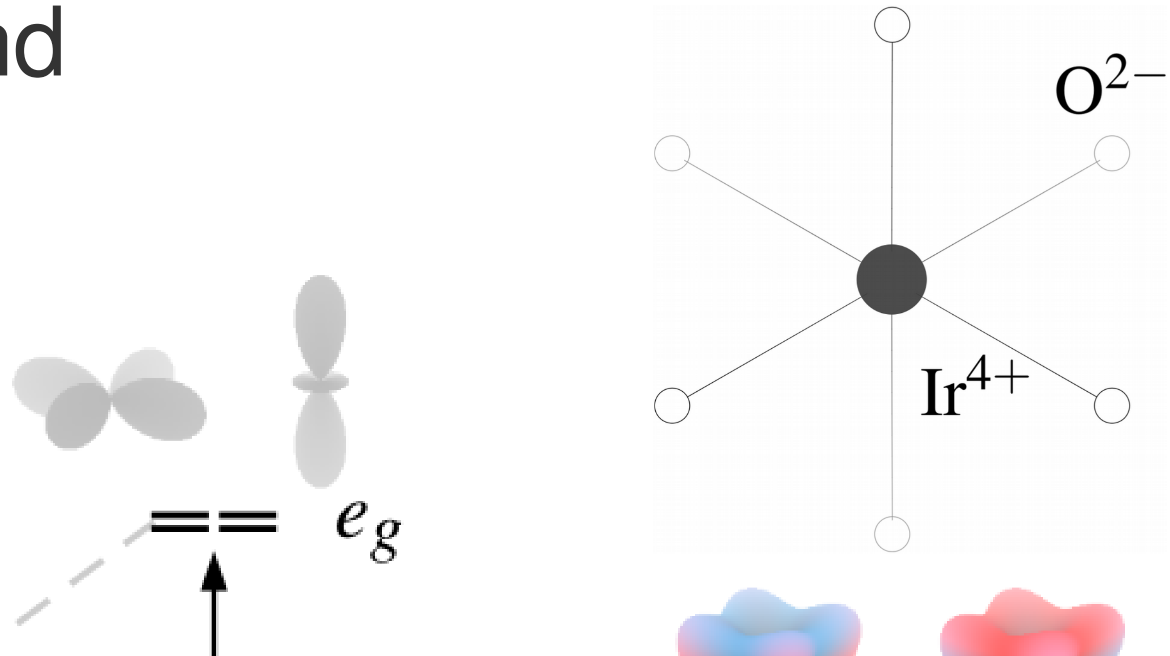


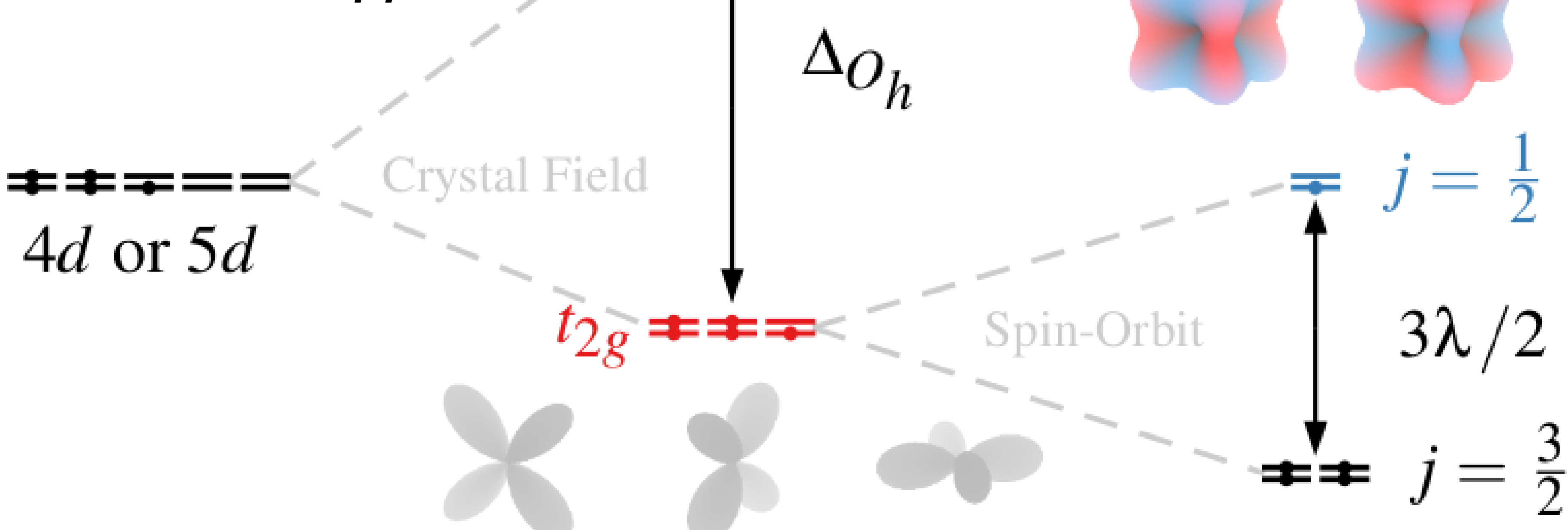

**Exactly solvable with spin liquid ground state**

A. Kitaev, Ann. Phys. (2005)

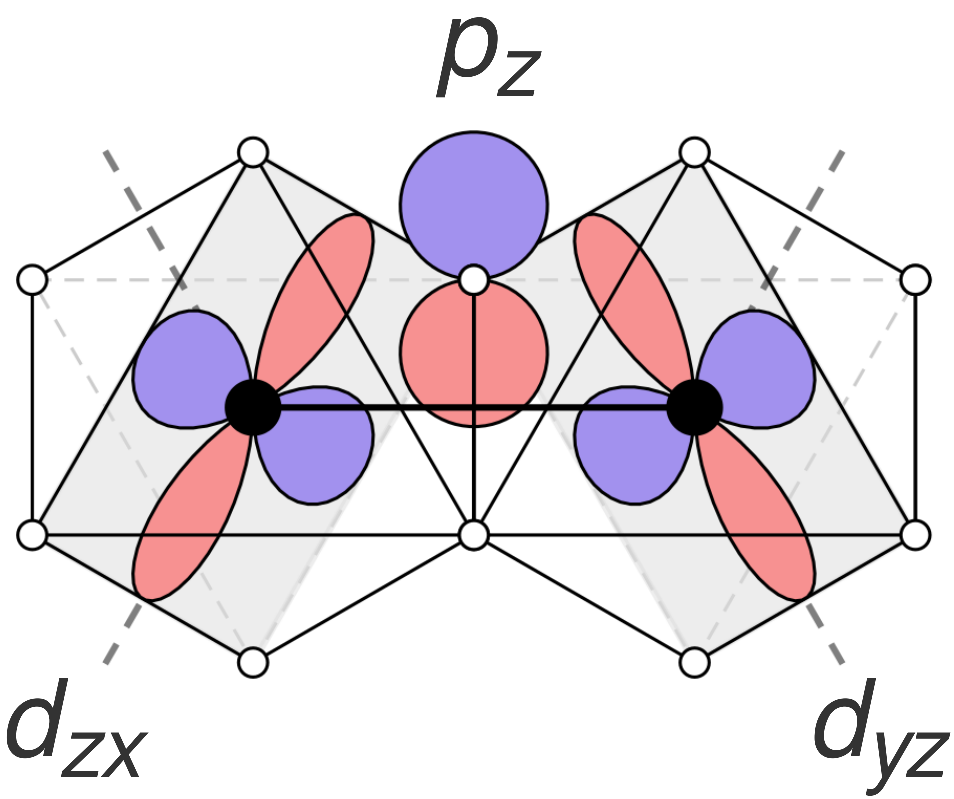
# Atomic Physics

- Crystal field and spin-orbit give $j = \frac{1}{2}$ states
- Interactions: Hubbard $U$, Hund's $J_H$



# Spin-orbit Mott insulators




- Edge-shared oxygen octahedra
- Effective $j=½$ spin model
- Considers only oxygen mediated exchange

*Ferromagnetic*

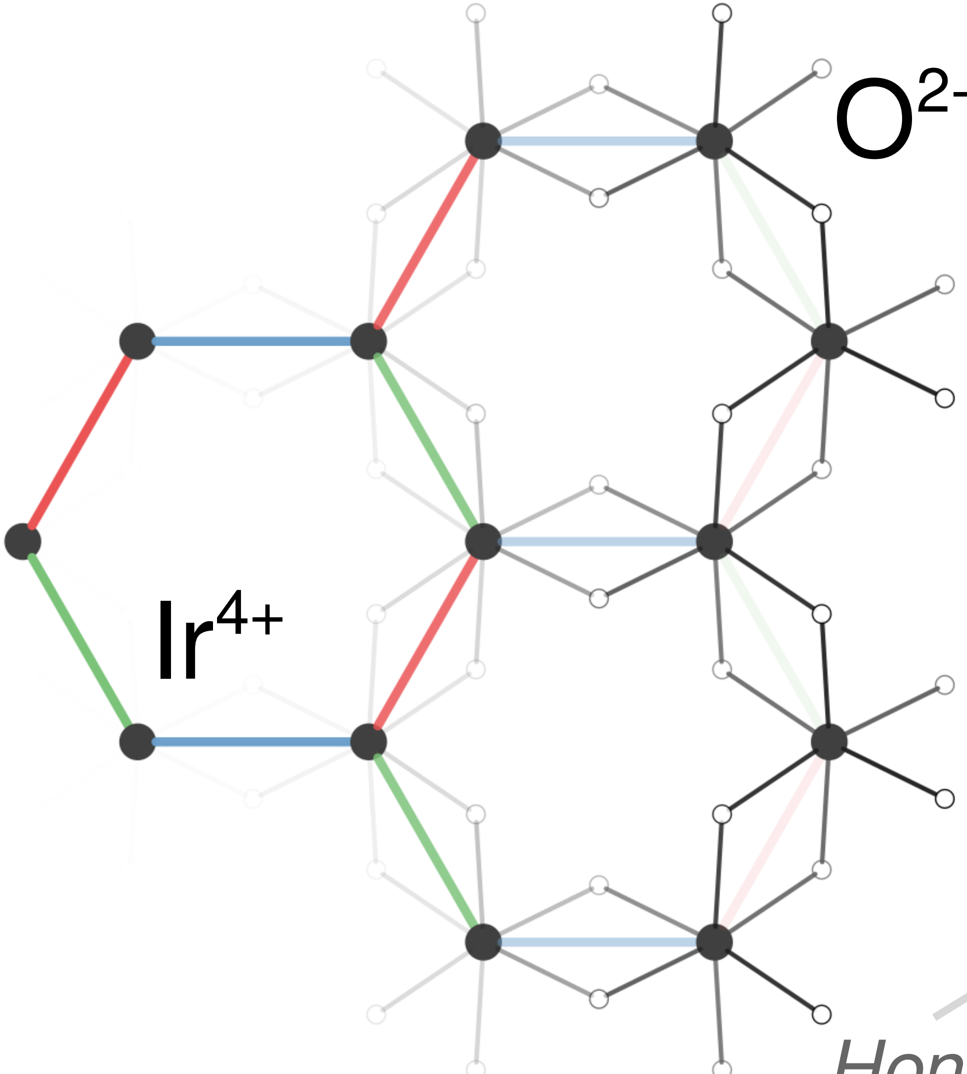
$$K \sim -\frac{8J_H}{3U^2}\left(\frac{t_{pd\pi}^2}{\Delta_{pd}}\right)^2$$

*Hund's coupling*

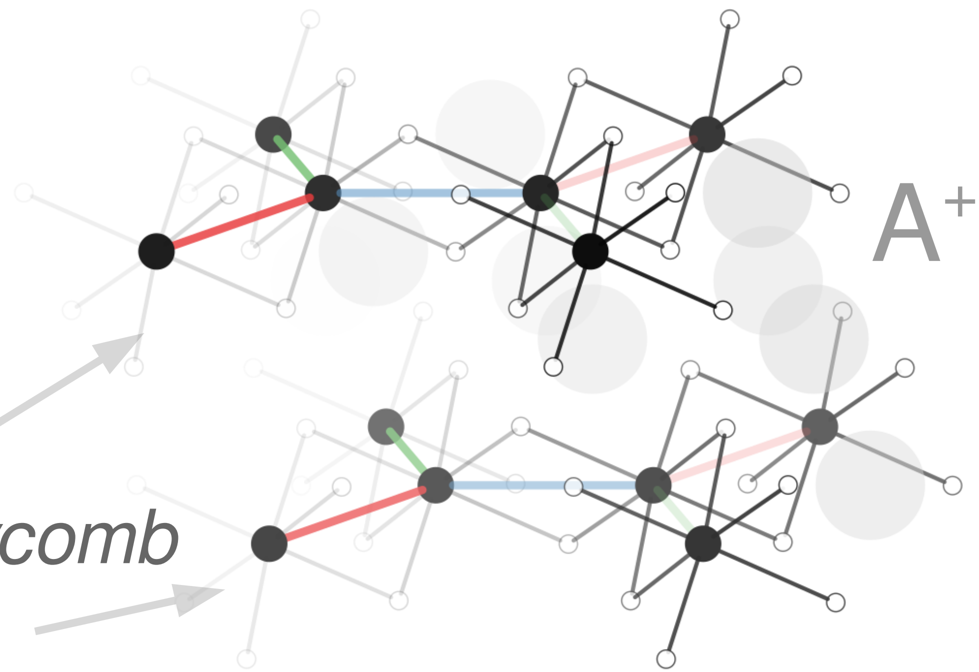
**Candidate materials?**

G. Jackeli & G. Khalliliuin, Phys. Rev. Lett. (2009)

# $Na_2IrO_3$ & α-$Li_2IrO_3$




- Strong Mott insulators
- Layered – Quasi-2D
- Edge-shared $O^{2-}$ octahedra about $Ir^{4+}$

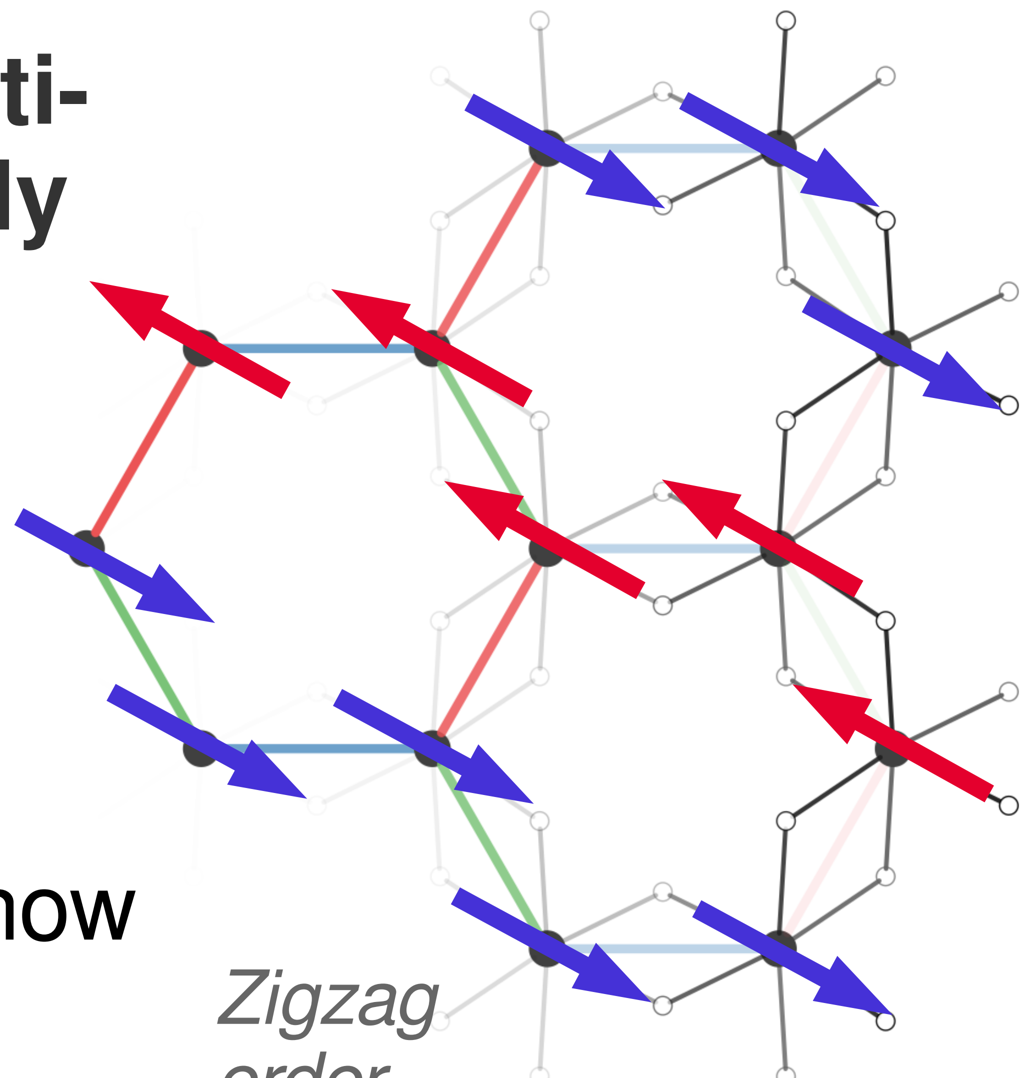



Y. Singh & P. Gegenwart, Phys. Rev. B (2010)

# Magnetic Ordering

- Both materials **anti-ferromagnetically order** near ~15K
- $Na_2IrO_3 \rightarrow$ **Zigzag** ordering pattern
- $Li_2IrO_3 \rightarrow$ Spiral order?

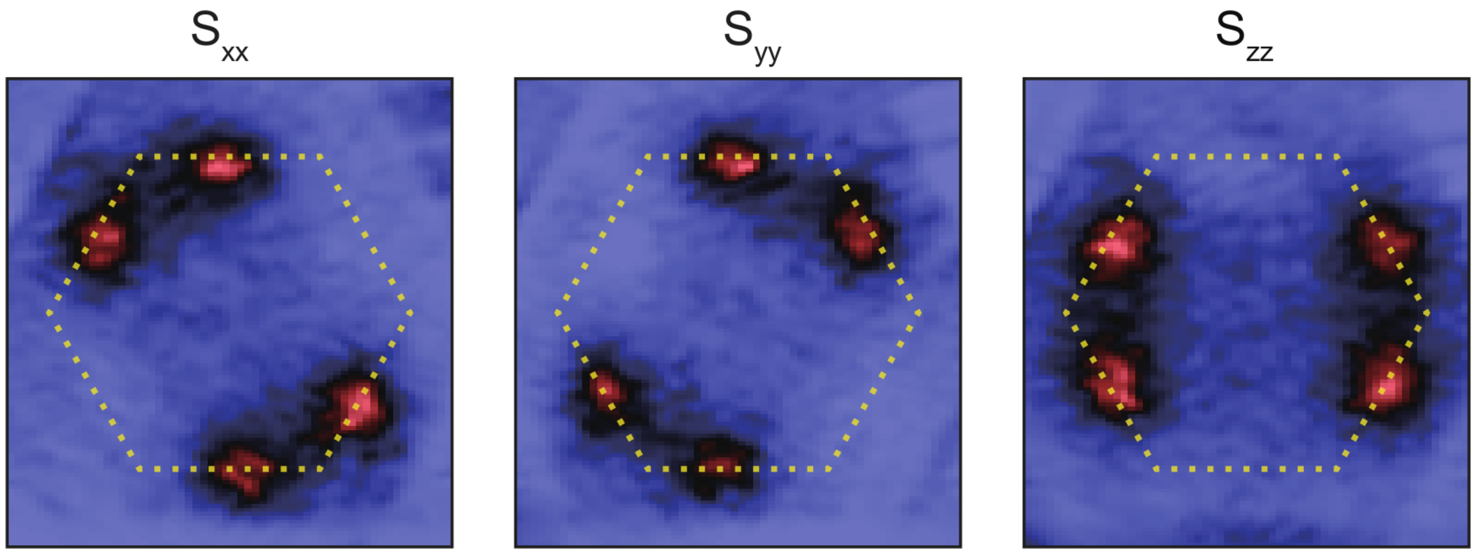
What else do we know about the zigzag order?



*Zigzag order*

Y. Singh et al, Phys. Rev. Lett. (2011); X. Liu et al, Phys. Rev. B (2011); F. Ye et al, Phys. Rev. B (2012)

# Diffuse scattering - $Na_2IrO_3$



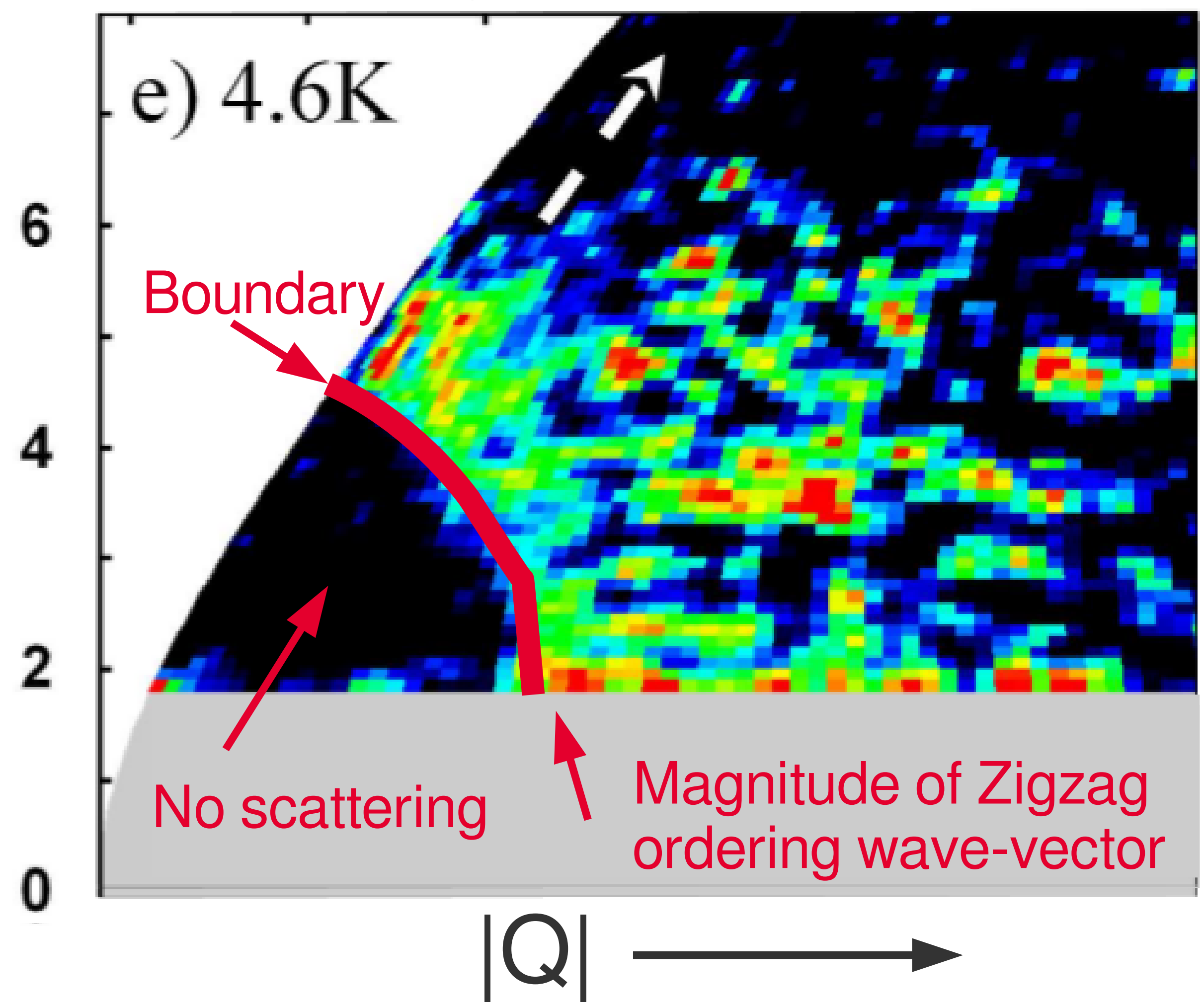

S.H. Chun, Nature Physics (2015)

- Above ordering transition – short range correlations
- Strong correlation between wave-vector and spin component
- **Strong anisotropic interactions**

# Neutron scattering - $Na_2IrO_3$

- Absence of scattering at low-Q
- Scattering down to at least 1 meV - **small gap**
- Concave boundary

Power-averaged inelastic cross-section



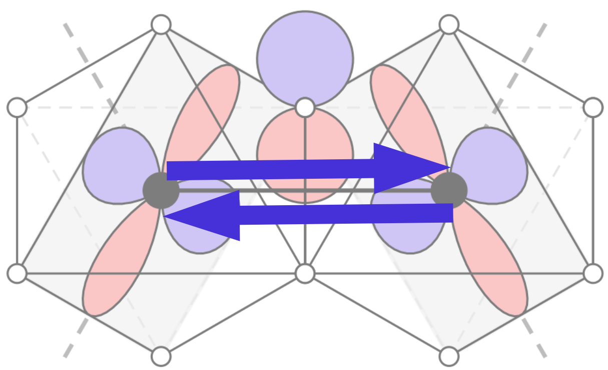

**Some of the only information on the low-energy structure of the zigzag state**

S.K. Choi et al, Phys. Rev. Lett. 108, 127204 (2012)

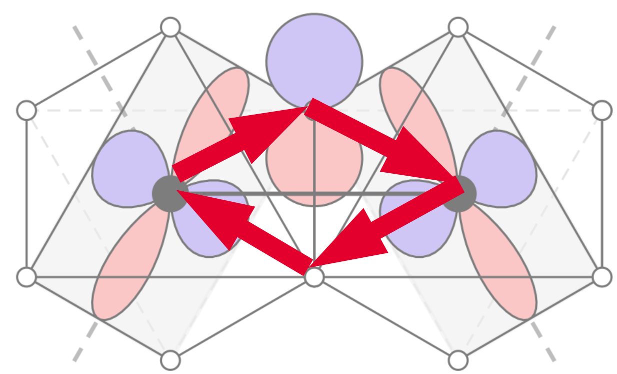
# Heisenberg-Kitaev Model

- Heisenberg interaction in addition to Kitaev

$$\sum_{\langle ij\rangle \in \gamma} \left( J \vec{S}_i \cdot \vec{S}_j + K S_i^{\gamma} S_j^{\gamma} \right)$$



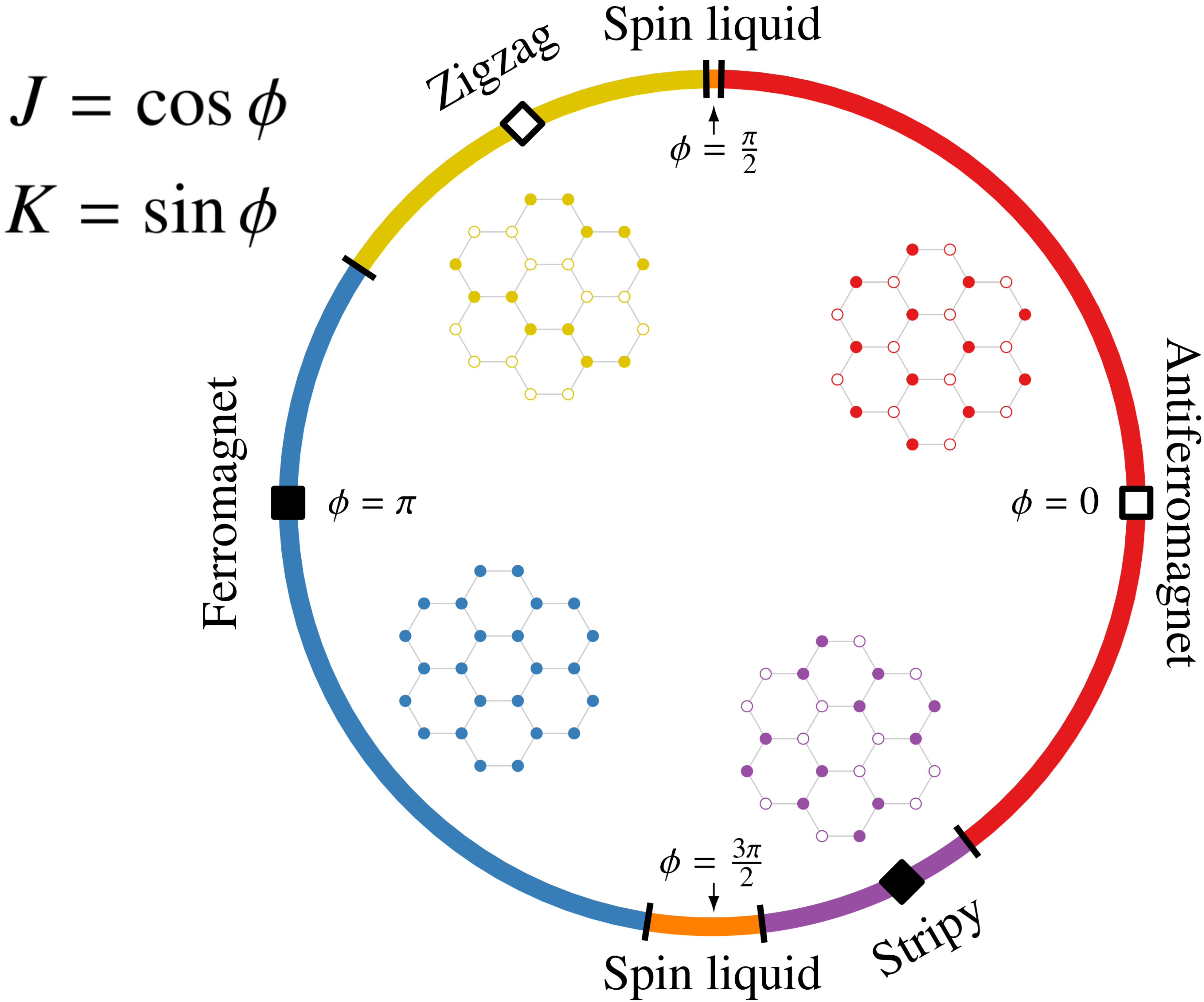
*J from direct d-d*



*K oxygen-mediated*

- Phase diagram of this model well-established; classically, exact diagonalization, DMRG, …

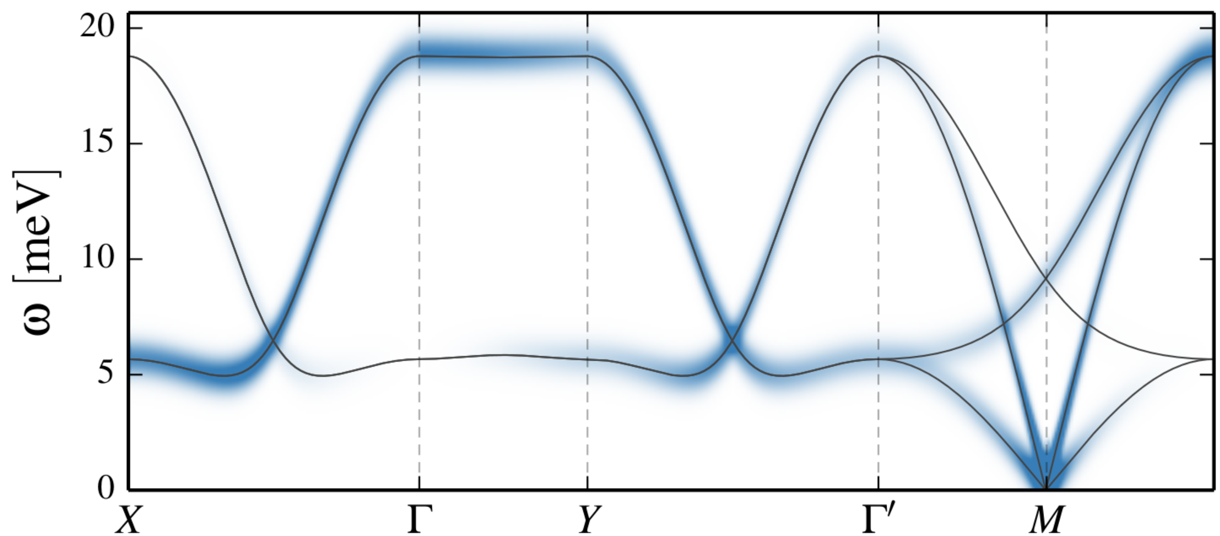
J. Chaloupka, G. Jackeli & G. Khalliliuin, Phys. Rev. Lett. (2010), ibid (2013);Okamoto, Phys. Rev. Lett. (2013); H.C.Jiang et al Phys. Rev. B (2011)



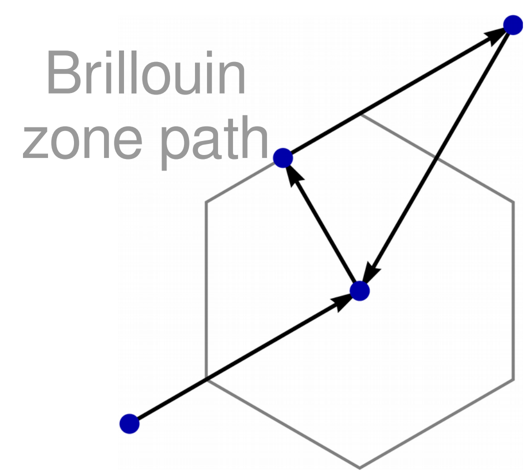
$J = \cos\phi$
$K = \sin\phi$
Zigzag
Spin liquid
$\phi = \frac{\pi}{2}$
Ferromagnet
$\phi = \pi$
$\phi = 0$
Antiferromagnet
$\phi = \frac{3\pi}{2}$
Spin liquid
Stripy

# HK-Model: Properties

- Roughly in agreement with INS and RIXS data; nearly gapless with large energy scale
- **Kitaev coupling must be near AFK limit**




- **Accidental** classical degeneracy – gapless spin-waves




J. Chaloupka, G. Jackeli & G. Khalliliuin, Phys. Rev. Lett (2013)

# HKModel: Problems

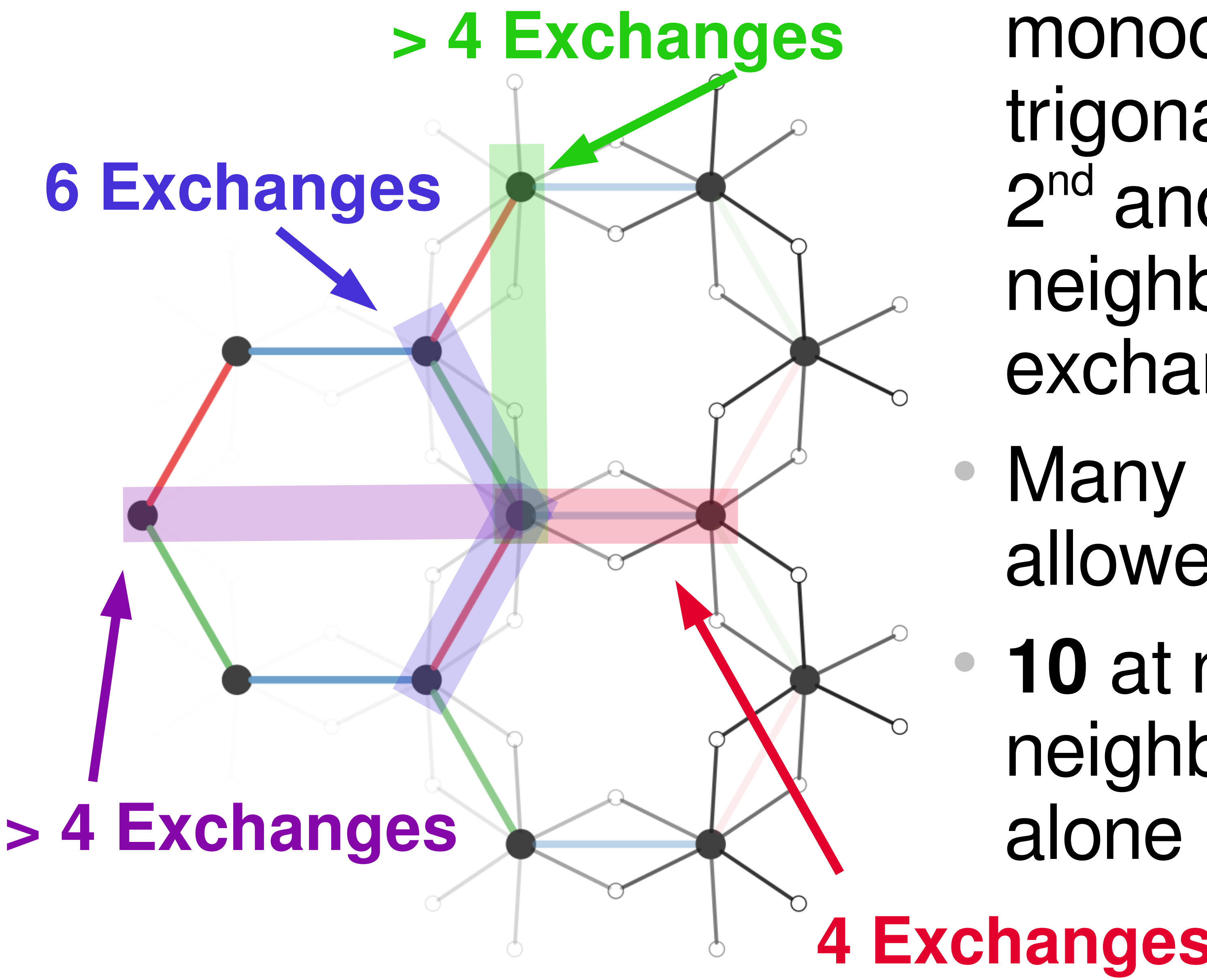
**Problem:** Oxygen mediated hopping dominates so K has wrong sign – Zigzag hard to get in HK model

**Some proposed solutions:**

- Strong $e_g$-$t_{2g}$ contribution to K to change sign
- Large further neighbour $J_2$ and $J_3$,
- Monoclinic distortions
- Second neighbour Kitaev, …

Chaloupka et al. Phys. Rev. Lett.(2010), (2013); Kimchi & You, Phys. Rev. B (2011), Yamaji et a. Phys. Rev. Lett. (2014), Katakuri et al, N. J. Phys. (2014), Sizyuk et al, Phys. Rev. B (2014), etc

# Generalities




- Structure has monoclinic, trigonal distortion, 2nd and 3rd neighbour exchange
- Many symmetry allowed channels
- **10** at nearest neighbour level alone

Yamaji et al, Phys. Rev. Lett (2014)

# Plan of attack

- Won't attempt full microscopic treatment
- Let's try the simplest possible consistent model and see how far it can take us
- **Questions:**

  **Can one find a zigzag phase?**

  **Can one explain the small excitation gap given strong anisotropy?**
- We'll start with **no** monoclinic distortion, **no** trigonal distortion; **nearest neighbour exchanges only – what's the model?**

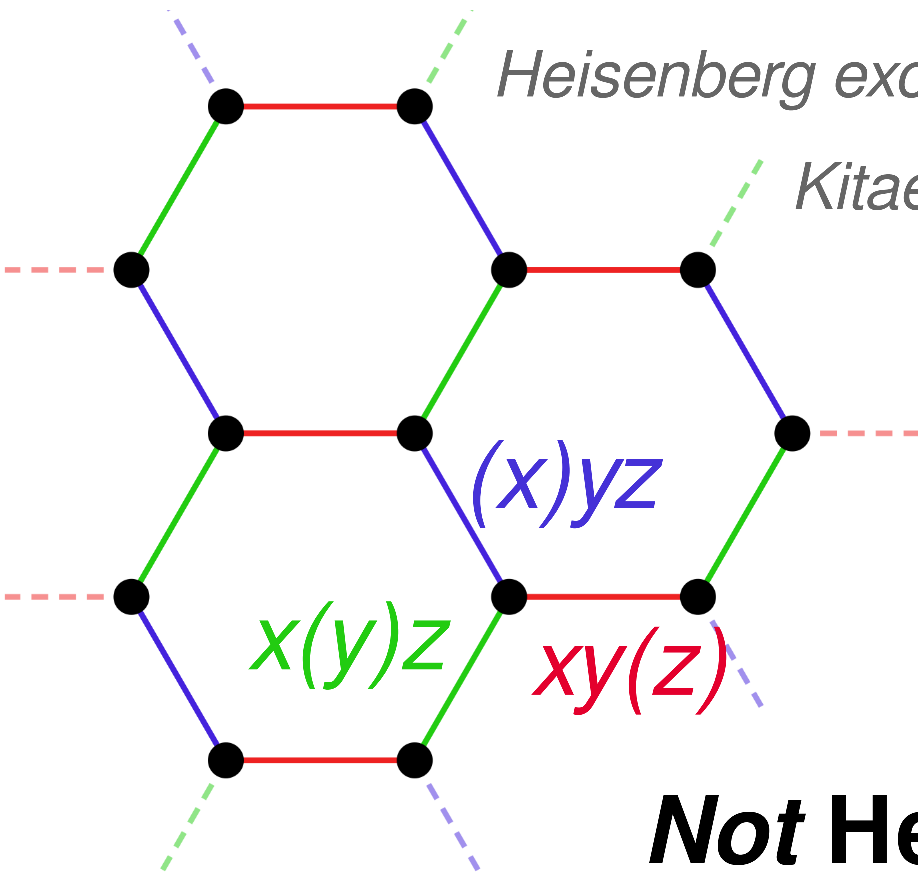
# Heisenberg-Kitaev-Γ Model

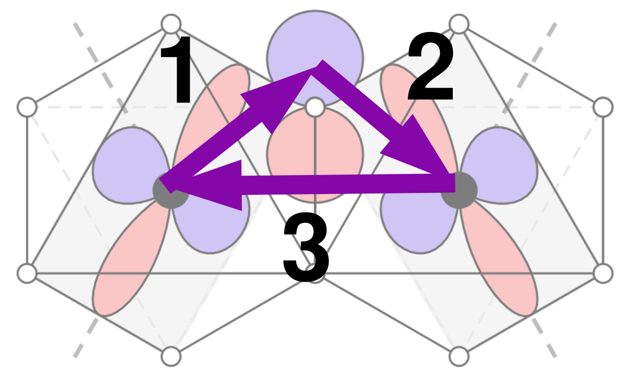
$$H = \sum_{\langle ij \rangle \in \alpha\beta(\gamma)} \left[ J\vec{S}_i \cdot \vec{S}_j + K S_i^\gamma S_j^\gamma + \Gamma \left( S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \right) \right]$$

*Heisenberg exchange*

*Kitaev exchange*

*Symmetric off-diagonal exchange*



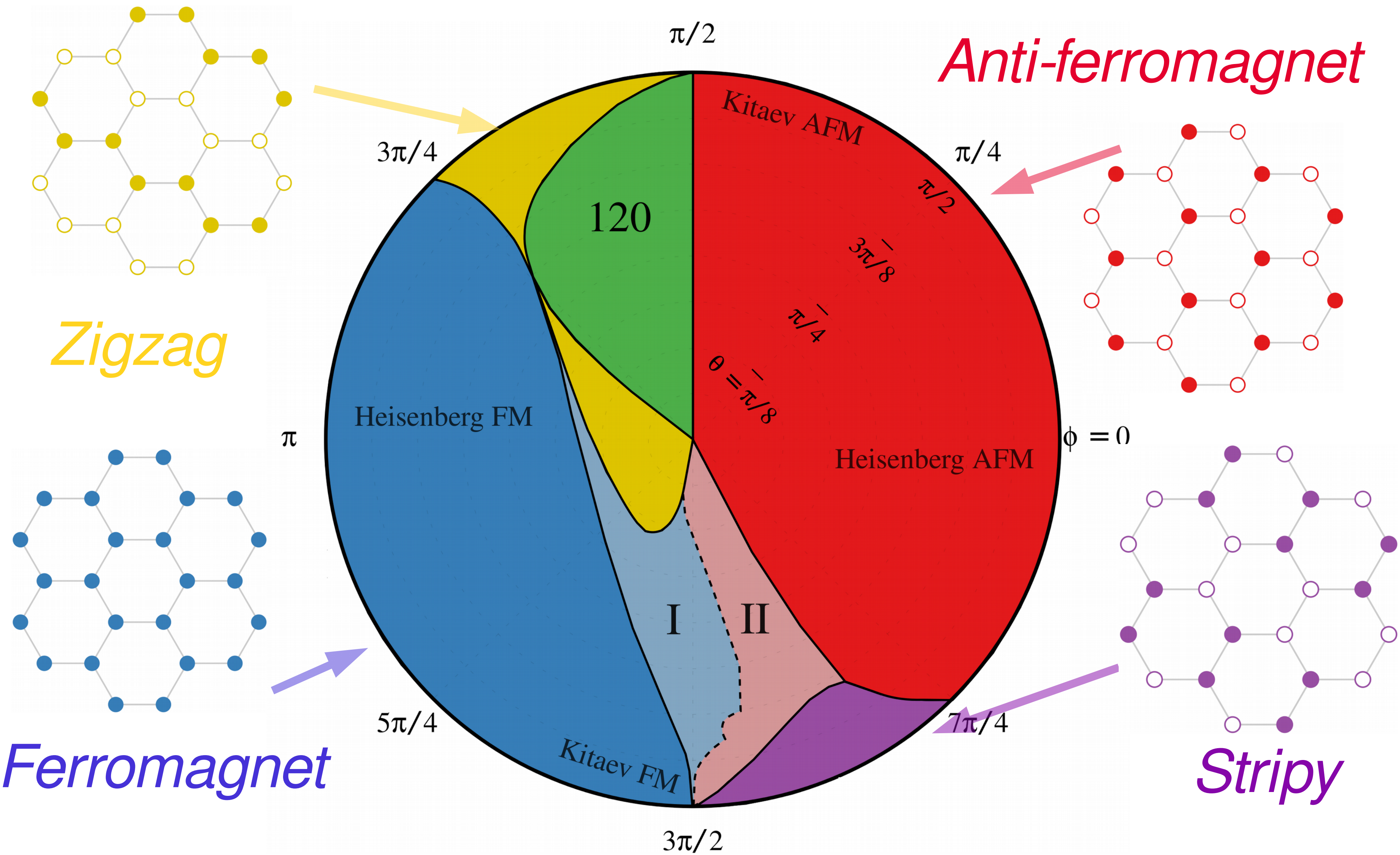




*Γ is cross-term*

***Not* Heisenberg-Kitaev model**

J. G. Rau, E. K. H. Lee & H. Y. Kee Phys. Rev. Lett. 112, 077204 (2014)

# Classical Phase Diagram Γ>0

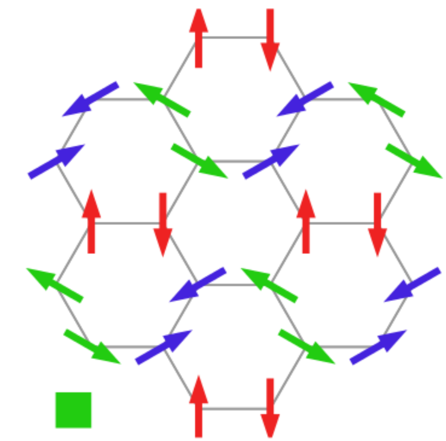


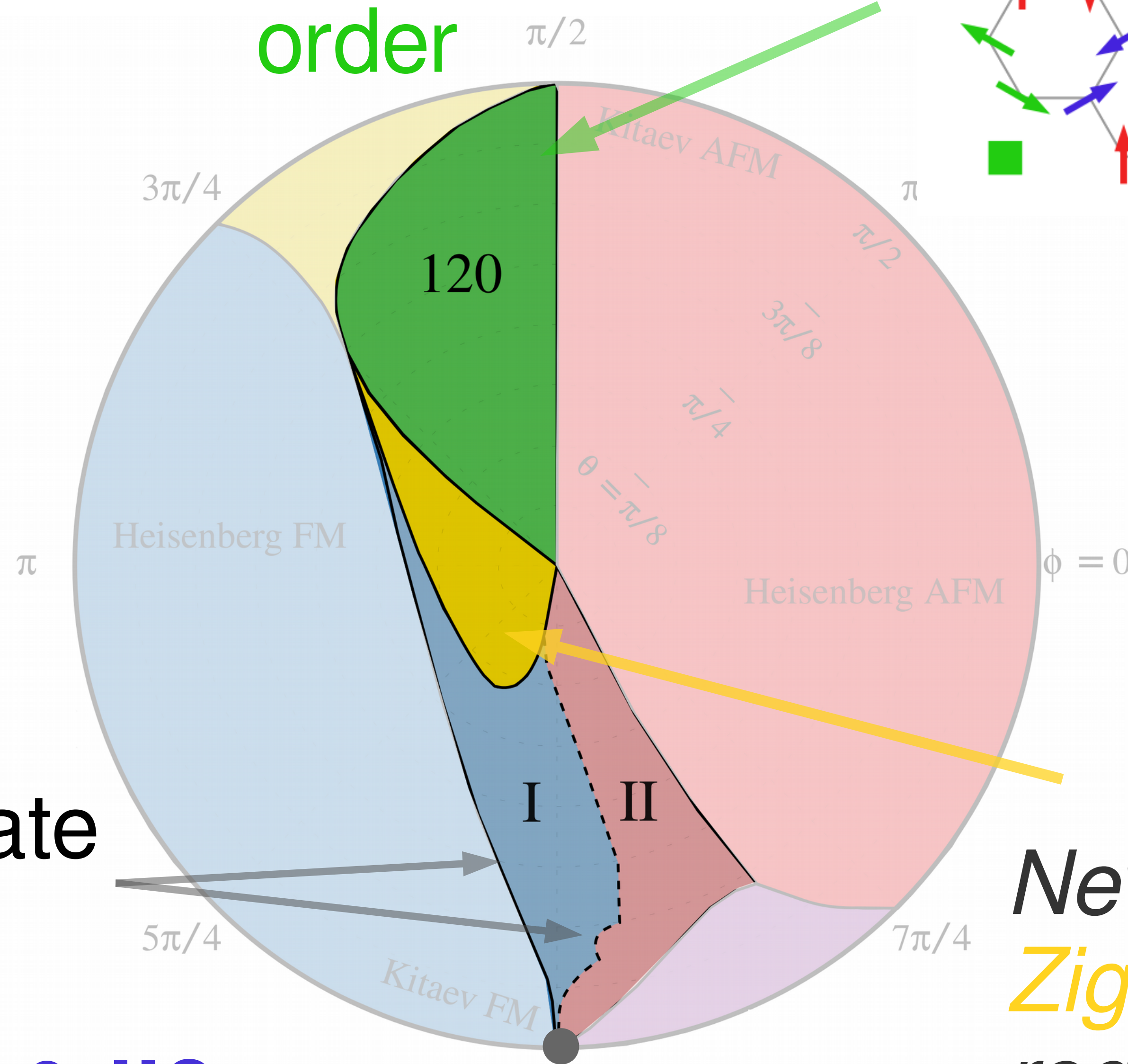

$$J = \sin\theta\cos\phi, \qquad K = \sin\theta\sin\phi, \qquad \Gamma = \cos\theta$$

# New Phases

- Two new phases beyond HK model
- **New Zigzag** away from FK limit

Three sublattice, 120° order






Incommensurate spiral orders

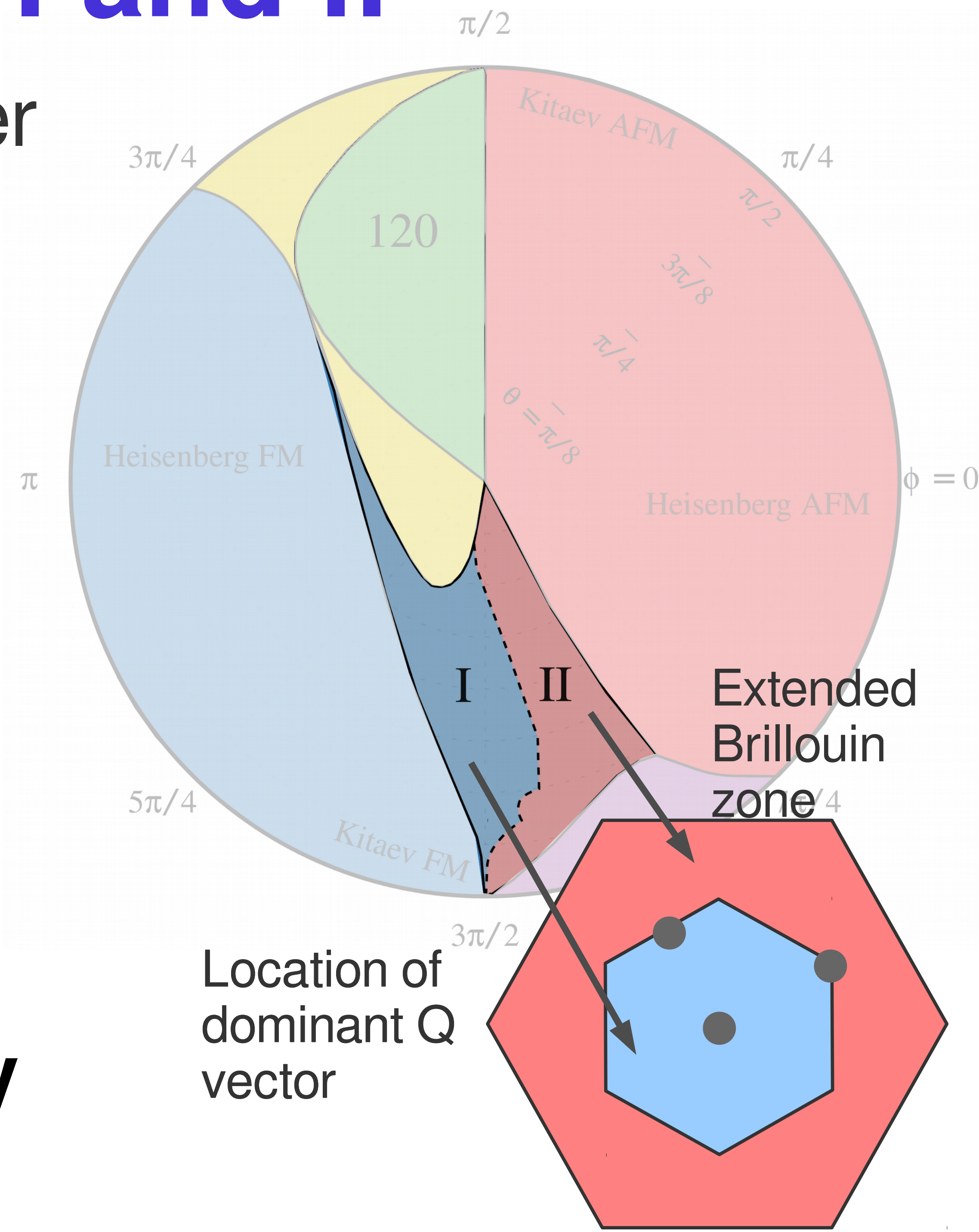
*New Zigzag region*

*FK limit*

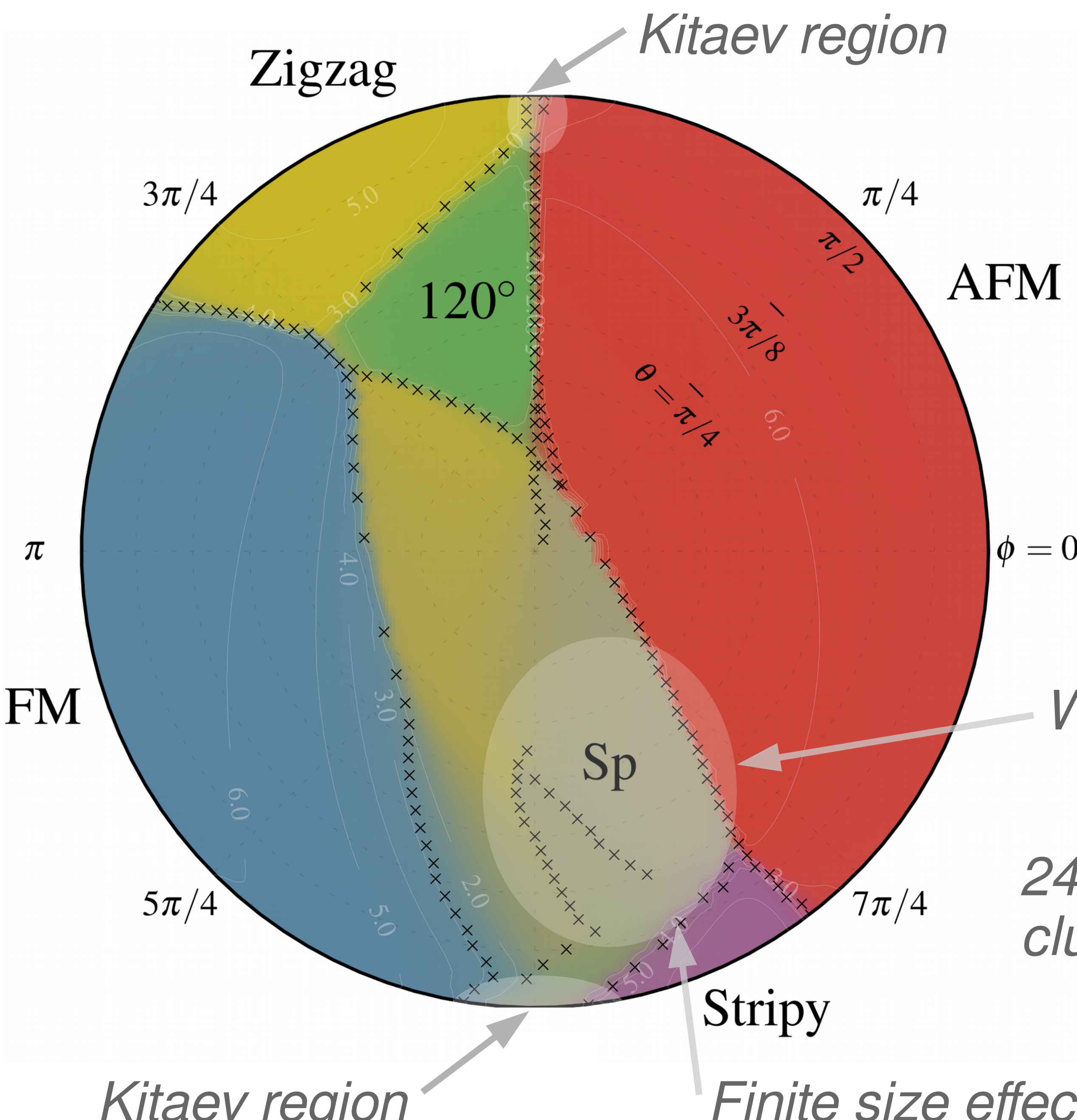
**Phase I & II?**

# Spiral phases: I and II

- Track largest Fourier component
- Dominant wave-vector changes ~ smoothly
- **Not single-Q**
- **Phase I**: in 1st BZ
- **Phase II**: not in 1st BZ
- Phase I energy **very close to zigzag**

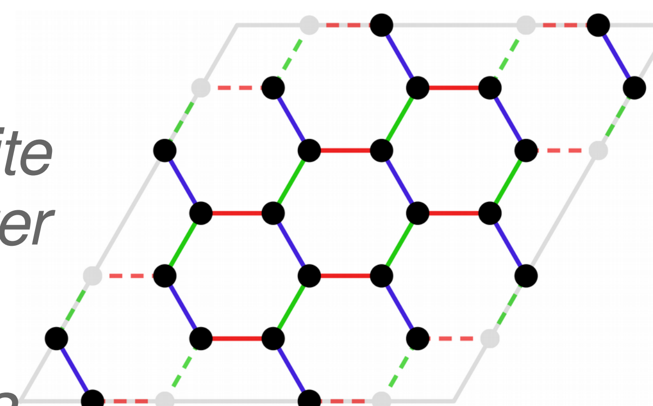


# Exact Diagonalization: Γ>0




- Identify phases using static structure factor
- *Sp* phase? $S_Q$ maximum at *K/2*

*24 site cluster*



# Trigonal Distortion?

- $Na_2IrO_3$ has trigonal **compression** along [111], out of plane
- Introduces another **off-diagonal exchange**

$$\Gamma' \sum_{\langle ij\rangle \in \alpha\beta(\gamma)} \left[S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\beta\right]$$

Positive for trigonal compression

$$\Gamma' = -\frac{8J_H}{9}\left[\frac{\sqrt{2}\theta\left((t_2+t_3)^2+4t_2^2\right)}{2(U-3J_H)(U-J_H)}\right]$$

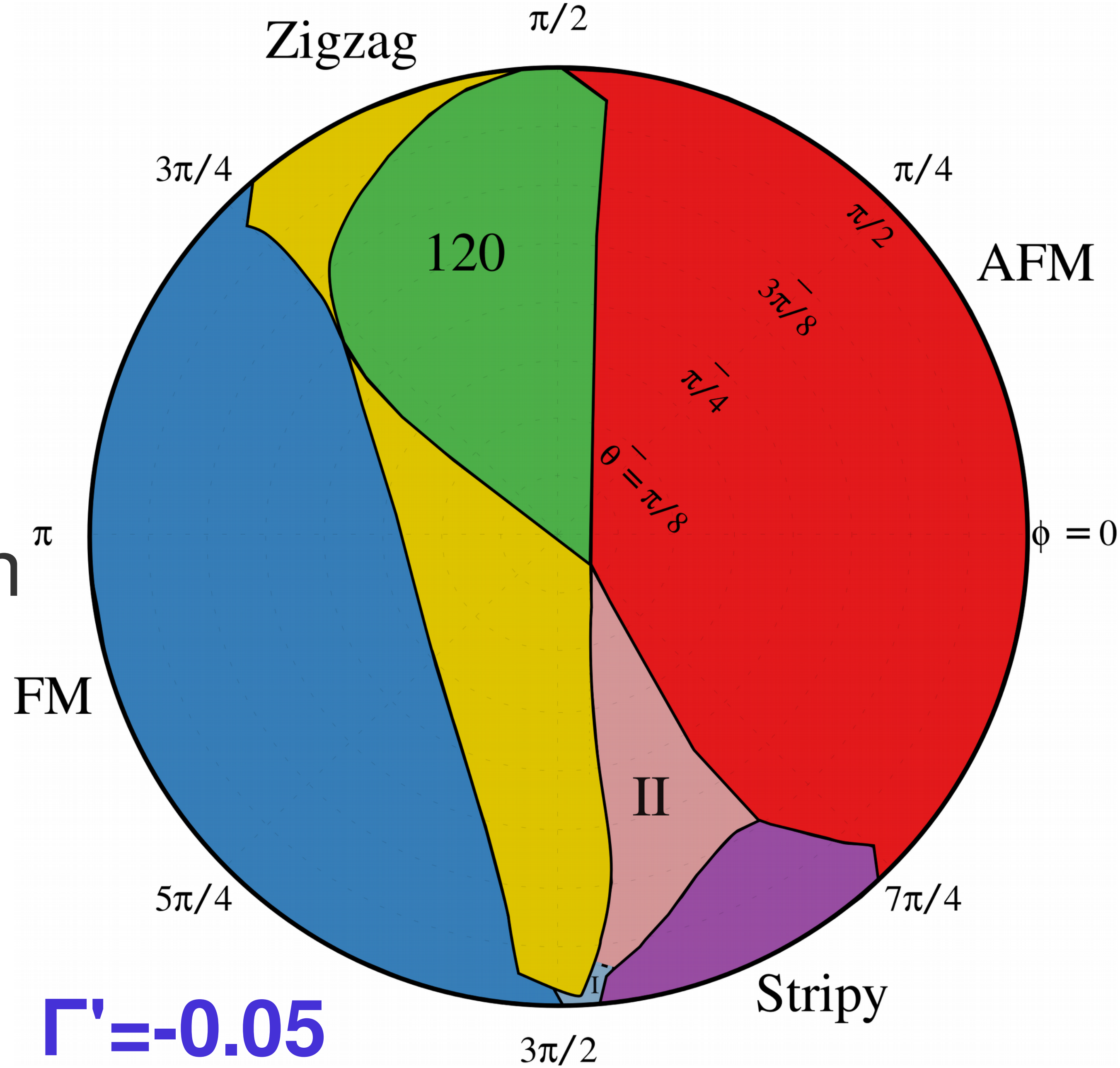
Leading term for small trigonal distortion

Negative for trigonal compression

**Will take $\Gamma' < 0$ for $Na_2IrO_3$**

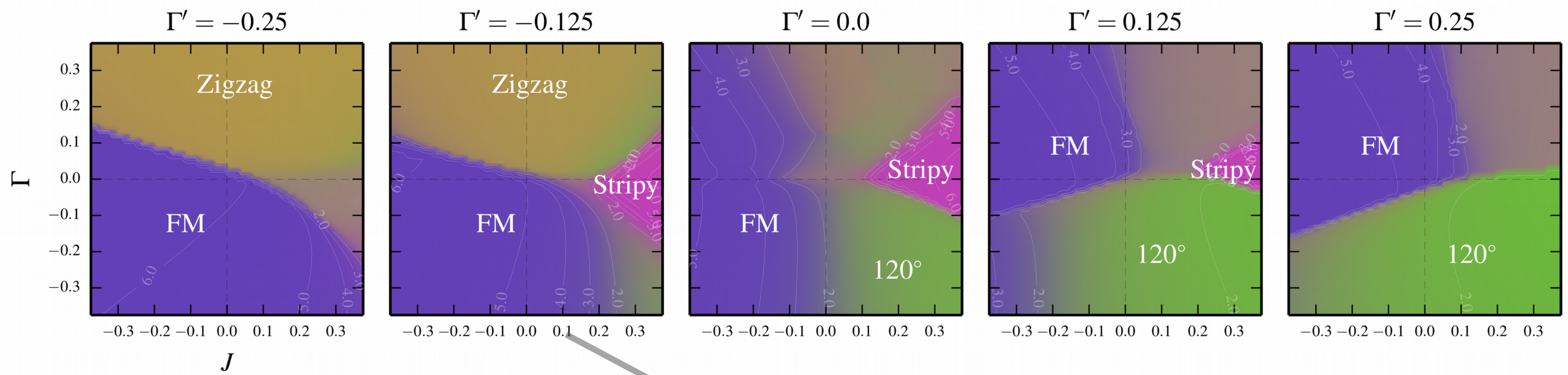
# Effects of Γ'<0 (classical)

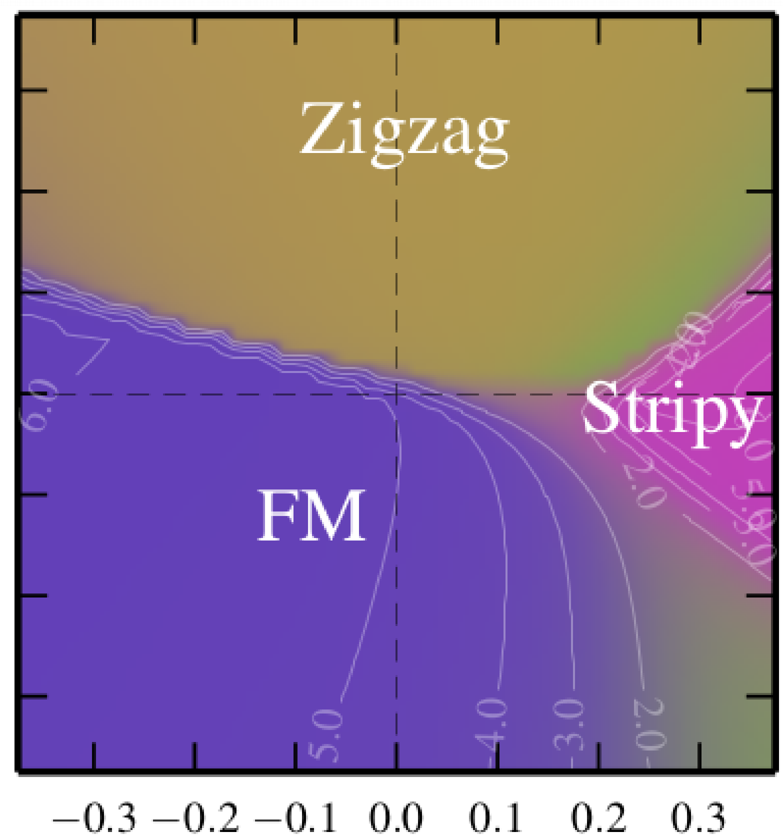
- Phase I **sensitive** to trigonal distortion
- **Large zigzag phase** for even small values
- **Zigzag close to FK limit**




Γ'=-0.05

# Effects of Γ'<0 (quantum)




Fixed scale so |K|=1

- Focus on FK limit
- Zigzag correlations increasing

**Beyond ordering?**

# Spin waves with Γ and Γ'

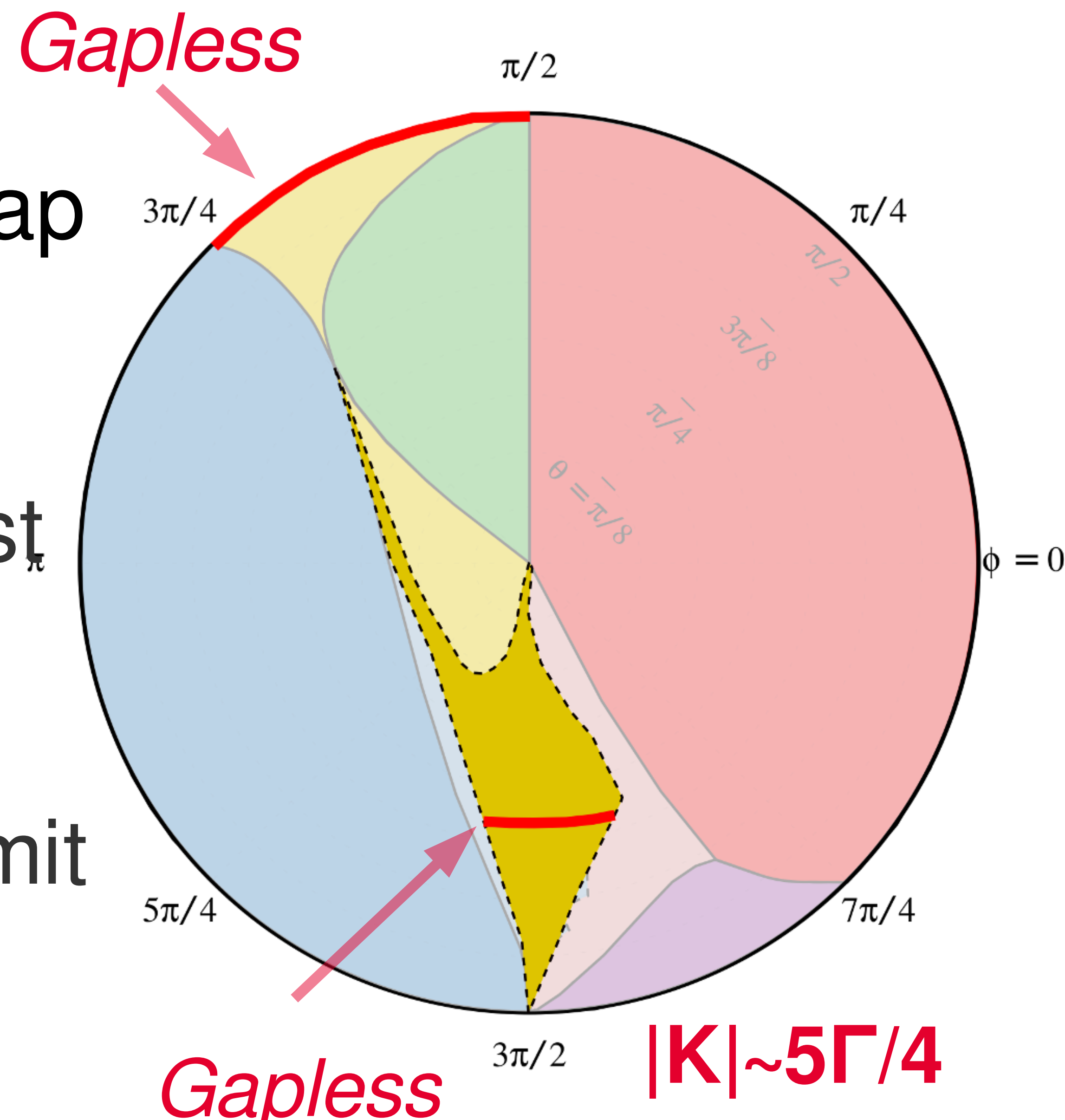
- Accidental classical degeneracy of HK model **spoiled**
- Gap ~ |Γ|,|Γ'|
- Microscopics: no reason for Γ to be that small
- Spin waves generically gapped

- Experimental input: excitation spectrum has low energy excitations <1-2 meV
- Overall scale large; RIXS excitations at ~30 meV

**How to reconcile?**

# Spin wave gap in zigzag phase

- Consider Γ'=0 and look at **spin-wave gap in zigzag state**
- Zigzag phase ***metastable*** for most of phase I region
- **Two gapless regimes**: the HK-limit & a large Γ region near FK limit



# What's going on?

- Highly anisotropic, **shouldn't have gap**
- Duality in J-K-Γ-Γ' model – C2 spin rotation about [111]

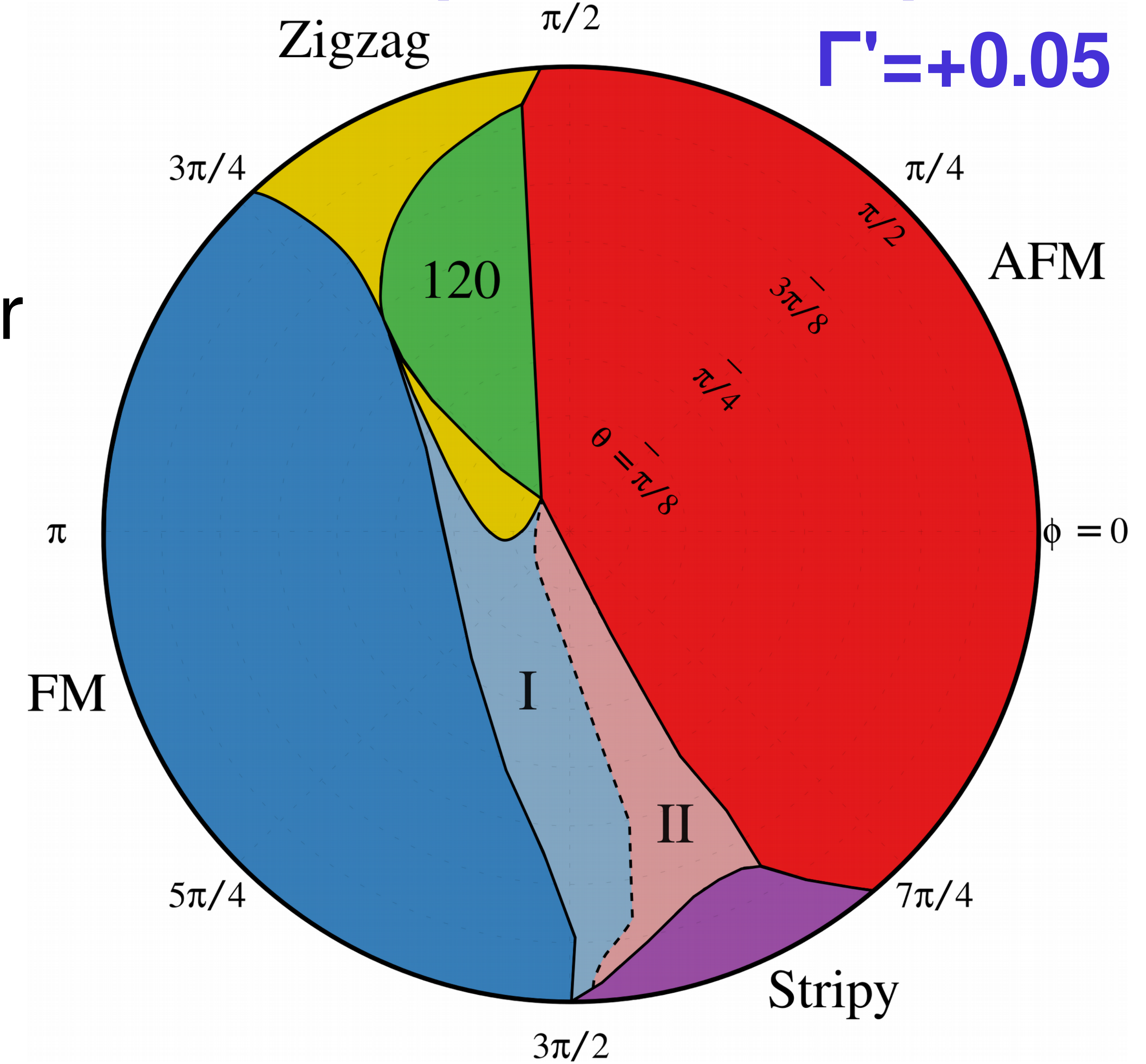
$$\begin{pmatrix} J \\ K \\ \Gamma \\ \Gamma' \end{pmatrix}' = \begin{pmatrix} 1 & +\frac{4}{9} & -\frac{4}{9} & +\frac{4}{9} \\ 0 & -\frac{1}{3} & +\frac{4}{3} & -\frac{4}{3} \\ 0 & +\frac{4}{9} & +\frac{5}{9} & +\frac{4}{9} \\ 0 & -\frac{2}{9} & +\frac{2}{9} & +\frac{7}{9} \end{pmatrix} \begin{pmatrix} J \\ K \\ \Gamma \\ \Gamma' \end{pmatrix}$$

- **Gapless regime dual to HK model** with accidental degeneracy
- 'Solvable' dual point with zigzag order

Chaloupka & Khalliliulin, PRB (2015)

# Effects of Γ'>0 (classical)

- Phase I is **stable**
- Incommensurate spirals persist
- Trigonal distortion is **smaller** in Li2IrO3

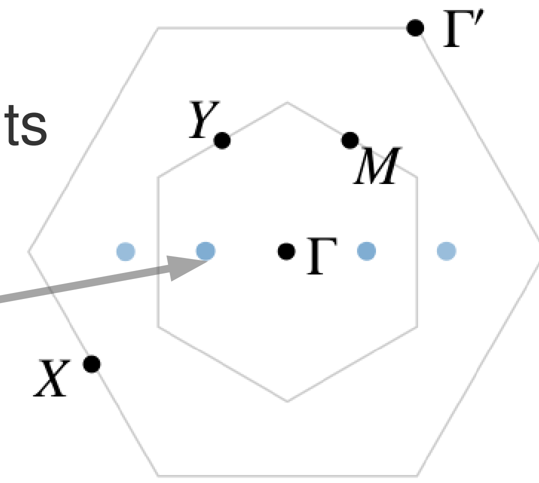


# From Phase I: 2M/3 Phase

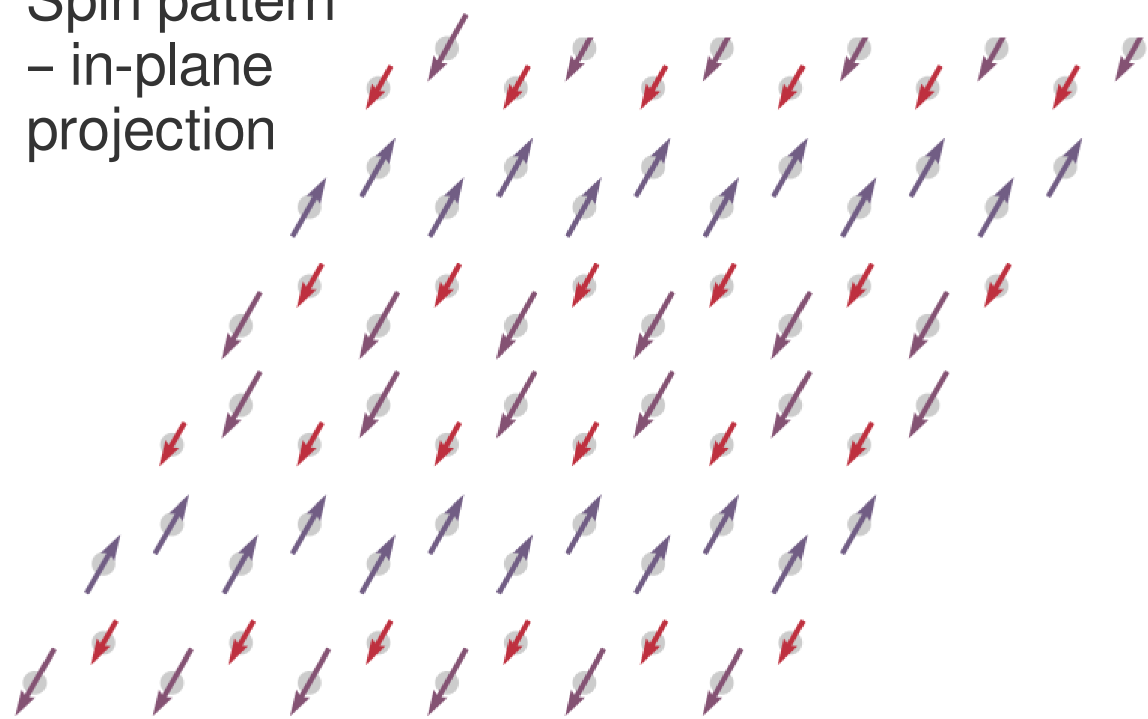
- Look at phase I near Γ'=0
- Dominant wave-vector ~2M/3, commensurate
- Three-fold, not single-Q state
- Ferrimagnetic (~0.2/site)
- $Q_{exp} \sim 0.41$ Å$^{-1}$ ?

Fourier components

2M/3 component




Spin pattern – in-plane projection



**2M/3~0.4Å$^{-1}$**

S.K. Choi, APS March Meeting Talk (2014)

# Summary

- **Trigonal compression can stabilize a zigzag phase near the FK-limit** within a nearest neighbour model
- Parameter regime with **small spin-wave gap at large anisotropy**
- Zigzag phase near FK limit competes with incommensurate spiral phase – **related to $Li_2IrO_3$?**


For details see:

J. G. Rau & H. Y. Kee, arxiv:1408.4811 (2014)

J. G. Rau, E. K. H. Lee & H. Y. Kee, Phys. Rev. Lett. **112**, 077204 (2014)