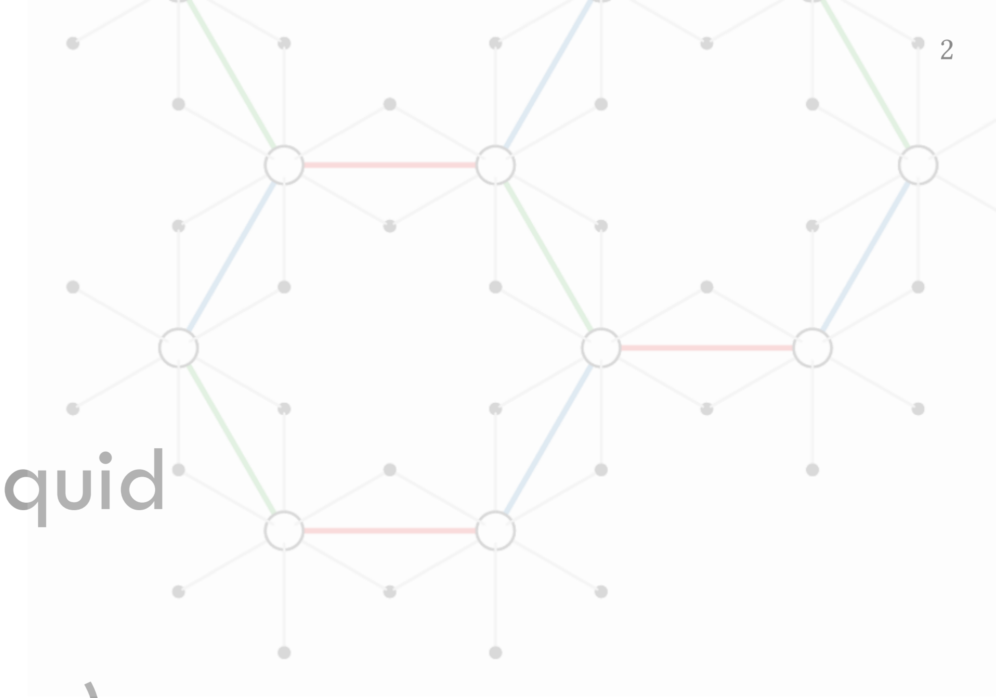
Tutorial:
Physics of the Kitaev Model and its Realization in Kitaev Materials

Jeffrey G. Rau

University of Windsor

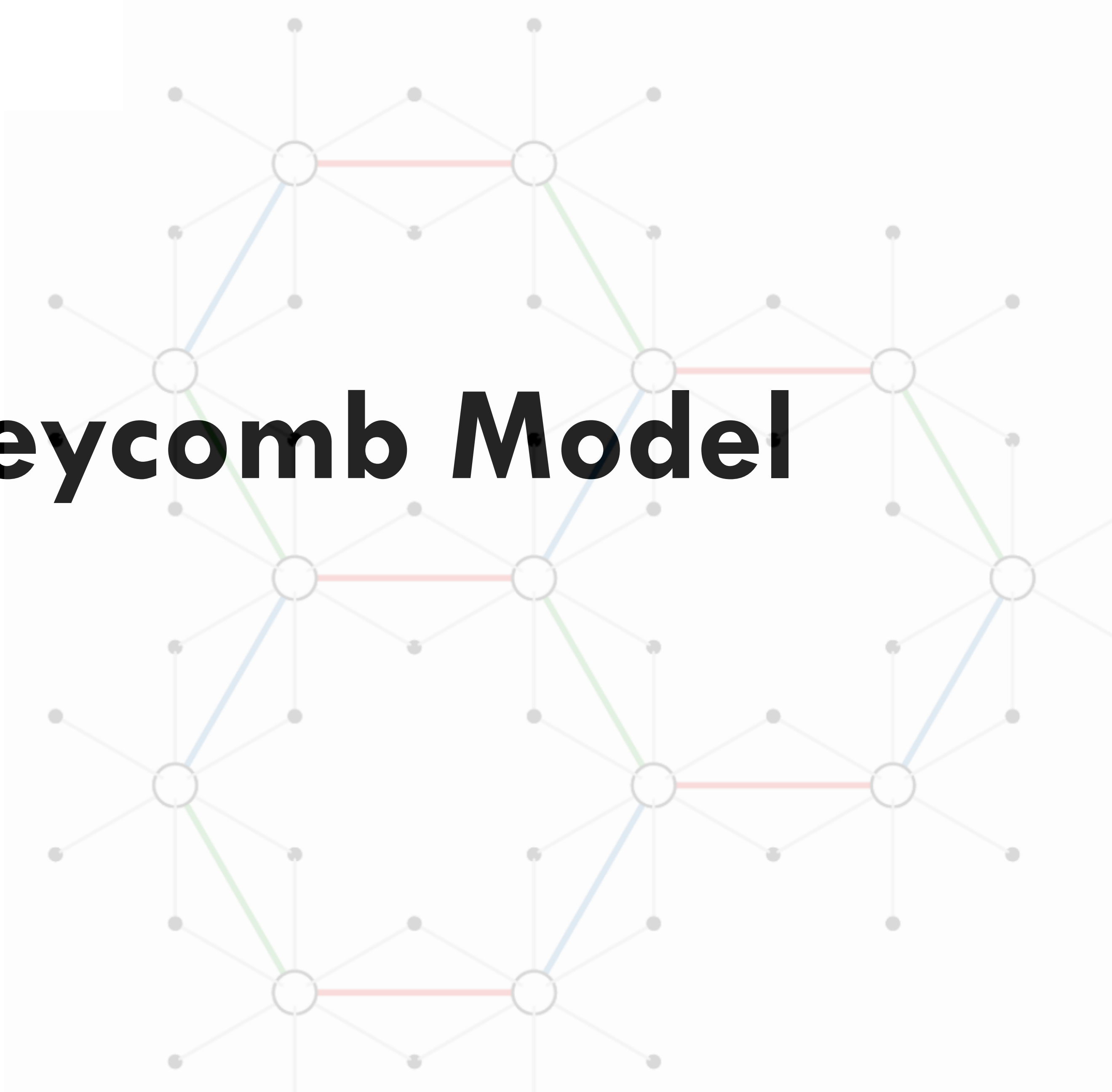
University of Windsor

waiting for the conference on HFM, MPI-PKS, January 27, 2021



1. Kitaev’s honeycomb model
   i. Definition & Solution
   ii. Properties of the Kitaev Spin Liquid
   iii. Effect of a Magnetic Field
   iv. Generalizations (3D, disorder, …)
2. Kitaev *materials*
   i. Jackeli-Khalliulin mechanism
   ii. Perturbations
   iii. $RuCl_3$

Kitaev’s Honeycomb Model



Kitaev's Honeycomb Model

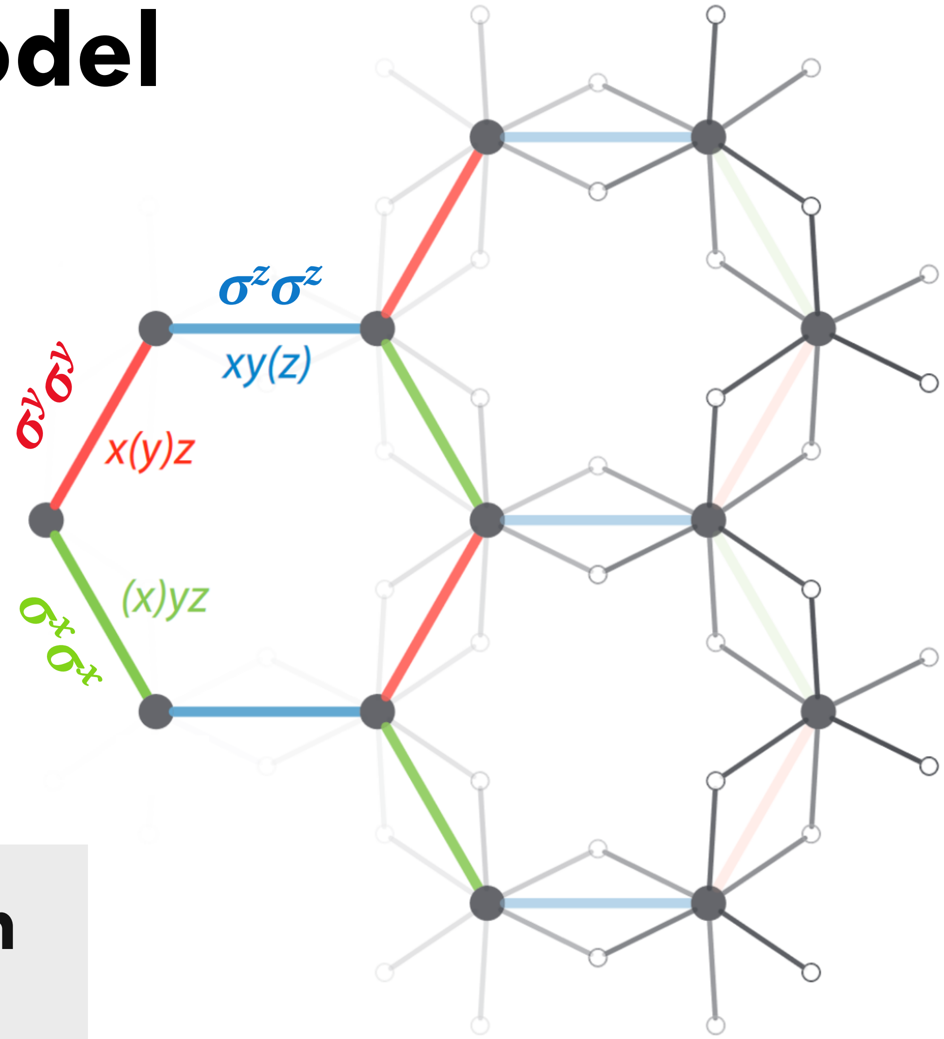
- Frustrated spin-1/2 model on honeycomb lattice

$$-J\sum_{\langle ij\rangle_\gamma}\sigma_i^\gamma\sigma_j^\gamma$$

Two-spin interactions only

- Frustration by *interactions* not geometry

Exactly solvable of a **quantum spin liquid** with *emergent Majorana fermion excitations*



Kitaev, Ann. Phys. ***321***, *2 (2006)*

Plaquette symmetries

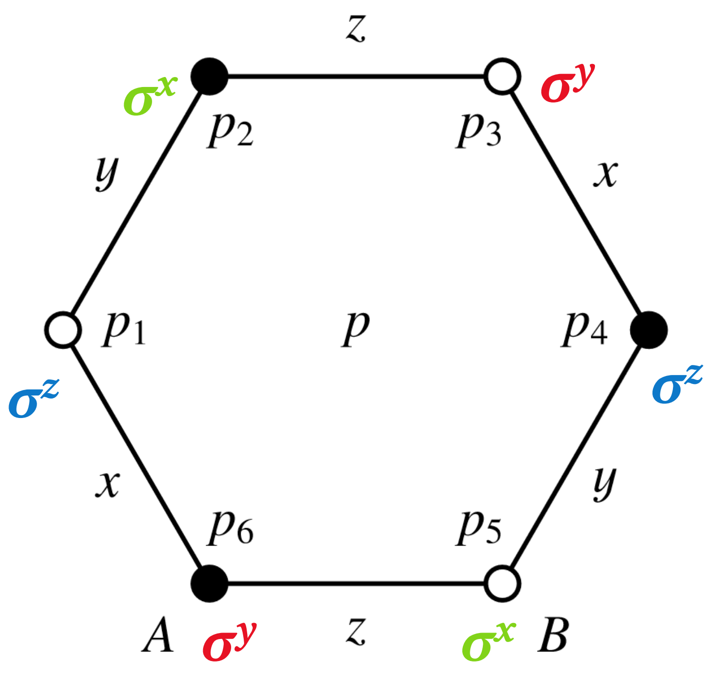
- *Infinite* number of conserved quantities

$$W_p = \sigma^z_{p_1}\sigma^x_{p_2}\sigma^y_{p_3}\sigma^z_{p_4}\sigma^x_{p_5}\sigma^y_{p_6}$$

- Commute with Hamiltonian *and* each other

$$[H, W_p] = 0 \qquad [W_p, W_{p'}] = 0$$

- Eigenvalues +1, -1:
  - *$2^{N/2}$ sectors each of size $2^{N/2}$*



For N sites, there are N/2 plaquettes

Kitaev, Ann. Phys. **321**, 2 (2006)

Absence of magnetic order

- Plaquette symmetries imply **no magnetic order**

$$\{\sigma_i^\mu, W_p\} = 0$$

Anti-commutation relation

there exists

- *Elitzur's theorem:* Can't spontaneously break local symmetries

$$\langle \boldsymbol{\sigma}_i \rangle = 0$$

- Also valid for higher-*S* Kitaev models

$$\langle \Psi_0 | \sigma_i^\mu | \Psi_0 \rangle$$

$W_p^2 = 1$

$$\langle \Psi_0 | \sigma_i^\mu W_p^2 | \Psi_0 \rangle$$

$\{\sigma_i^\mu, W_p\} = 0$

$$-\langle \Psi_0 | W_p \sigma_i^\mu W_p | \Psi_0 \rangle$$

Eigenstate of plaquette operators

$$-\langle \Psi_0 | \sigma_i^\mu | \Psi_0 \rangle$$

Exact solution: Plan

$D_i \equiv b_i^x b_i^y b_i^z c_i = 1$

$U_{ij} \equiv i b_i^\gamma b_j^\gamma$

$$-J \sum_{\langle ij \rangle_\gamma} \sigma_i^\gamma \sigma_j^\gamma \quad \rightarrow \quad iJ \sum_{\langle ij \rangle_\gamma} \left(i b_i^\gamma b_j^\gamma \right) c_i c_j \quad \rightarrow \quad iJ \sum_{\langle ij \rangle_\gamma} U_{ij} c_i c_j$$

$\boldsymbol{\sigma}_i \equiv i \boldsymbol{b}_i c_i$

$u_{ij} = \pm 1$

$$H[u] \equiv J \sum_{\langle ij \rangle_\gamma} i u_{ij} c_i c_j$$

$u_{ij} = +1$

$$H_0 = J \sum_{\langle ij \rangle_\gamma} i c_i c_j$$

Free fermions **(solvable)**

$w_p = u_{p_1 p_2} u_{p_2 p_3} u_{p_3 p_4} u_{p_4 p_5} u_{p_5 p_6} u_{p_6 p_1}$

Kitaev, Ann. Phys. ***321****, 2 (2006)*

Majorana representation

- Highly suggestive: *2^N/2* states per sector, *Majorana fermions?*

$$\boldsymbol{\sigma}_i \equiv i\boldsymbol{b}_i c_i \qquad \boldsymbol{b}_i \equiv (b_i^x, b_i^y, b_i^z)$$

- Represent spin-1/2 as *four* Majoranas, subject to *constraint*

$$D_i \equiv b_i^x b_i^y b_i^z c_i = 1$$

- Satisfy the anti-commutation relations for for Majorana fermions

$$\{c_i, c_j\} = 2\delta_{ij}$$

$$\{c_i, \boldsymbol{b}_j\} = 0$$

$$\{b_i^\mu, b_j^\nu\} = 2\delta_{ij}\delta_{\mu\nu}$$

Relation to SU(2) slave fermions?

- How does the relate to the "usual" representation:

$$\boldsymbol{\sigma}_i = \boldsymbol{f}_i^\dagger \boldsymbol{\sigma} \boldsymbol{f}_i$$

Complex *fermions*

- With constraint: $\boldsymbol{f}_i^\dagger \boldsymbol{f}_i = 1$

- **Equivalent**; just a *change of basis*

$$c = \frac{1}{\sqrt{2}}(f_\uparrow + f_\uparrow^\dagger)$$

$$b^x = \frac{1}{i\sqrt{2}}(f_\downarrow - f_\downarrow^\dagger)$$

$$b^y = -\frac{1}{\sqrt{2}}(f_\downarrow + f_\downarrow^\dagger)$$

$$b^z = \frac{1}{i\sqrt{2}}(f_\uparrow - f_\uparrow^\dagger)$$

One possible way to express Majoranas in terms of complex fermions

Burnell & Nayak, Phys. Rev. B ***84****, 125125 (2011);*
You et al, Phys. Rev. B ***86****, 085145 (2012)*

Hamiltonian in terms of Majoranas

- Substitute these in to Kitaev model:

$$\tilde{H} = iJ \sum_{\langle ij \rangle_\gamma} \left(i b_i^\gamma b_j^\gamma \right) c_i c_j$$

*Defined in **extended** space, need to impose constraint*

- If we can solve *this*, and get ground state $|\tilde{\Psi}_0\rangle$ then just need to *project* into physical subspace

*Really, **any** eigenstate*

$$|\Psi_0\rangle = P\,|\tilde{\Psi}_0\rangle$$

Ground state of Kitaev model

Imposes constraint $D_i \equiv b_i^x b_i^y b_i^z c_i = 1$

Link operators and Z_2 gauge structure

- To solve this, notice that the operators

$$U_{ij} \equiv i b_i^\gamma b_j^\gamma$$

- Commute with the Hamiltonian *and* with each other: **definite value in energy eigenstate**

$$[\tilde{H}, U_{ij}] = 0$$

$$[U_{ij}, U_{lk}] = 0$$

$$U_{ij}^2 = 1$$

*Really, **any** eigenstate*

$$U_{ij} |\tilde{\Psi}_0\rangle = u_{ij} |\tilde{\Psi}_0\rangle$$

- Two possible values: $u_{ij} = \pm 1$

Defines a Z_2 **gauge field** for the c Majorana fermions

Z_2 Flux Operators

Under gauge transformation:

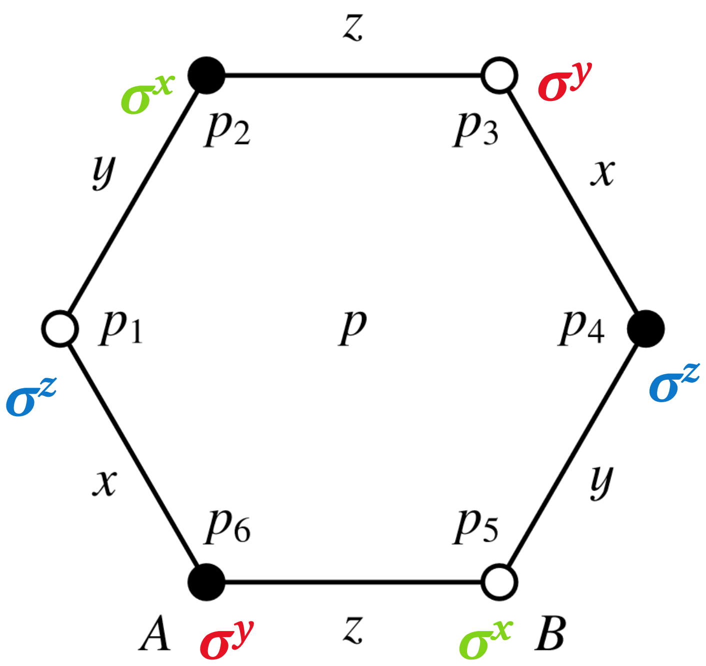
Preserves spin-operators
$\sigma_i \equiv i b_i c_i$

$z_i = \pm 1$

$$c_i \rightarrow z_i c_i$$
$$b_i \rightarrow z_i b_i$$

$$u_{ij} \rightarrow z_i z_j u_{ij}$$

$$W_p = \sigma^z_{p_1}\sigma^x_{p_2}\sigma^y_{p_3}\sigma^z_{p_4}\sigma^x_{p_5}\sigma^y_{p_6}$$



- What are the associated **Z_2 flux** operators?

$$w_p = u_{p_1p_2}u_{p_2p_3}u_{p_3p_4}u_{p_4p_5}u_{p_5p_6}u_{p_6p_1}$$

Product of link variables around hexagon

$$W_p\,|\tilde{\Psi}_0\rangle = w_p\,|\tilde{\Psi}_0\rangle$$

± 1

- *Gauge invariant* quantities

Flux sectors

- Gauge field is **static**: fluxes (and links) have *fixed* values
- Each of the $2^{N/2}$ choices of u_{ij} defines **flux sector**

$$H[u] \equiv J \sum_{\langle ij \rangle_\gamma} i u_{ij} c_i c_j$$

Independent "block" of Hamiltonian

Size of block = $2^{N/2}$

- Each flux sector is a *free fermion* problem! (efficiently solvable)

Cost is $O(N^3)$

Ground state? Need to find flux sector with *lowest possible energy.*

Ground state flux sector & Lieb's Theorem

- Could brute force minimize; instead can use **Lieb's theorem:**

Ground sector state is **flux-free**

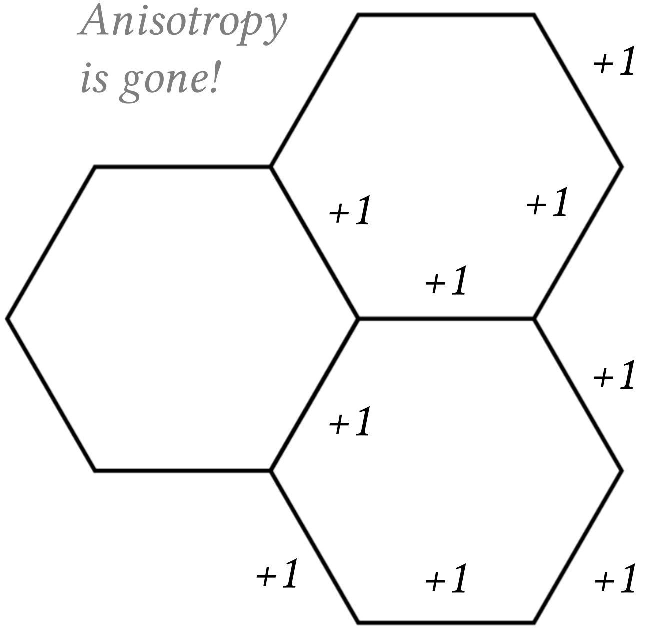
Depends on lattice structure

Simplest gauge choice

$$u_{ij} = +1$$

- Description is *free Majoranas* hopping on honeycomb lattice

Anisotropy is gone!



$$H_0 = J \sum_{\langle ij \rangle_\gamma} i c_i c_j$$

Lieb, Phys. Rev. Lett. ***73,*** *2158 (1994)*

Solution in flux-free sector

$$c_{\boldsymbol{r},\alpha} = \frac{1}{\sqrt{N}} \sum_{\boldsymbol{k}} e^{i\boldsymbol{k}\cdot\boldsymbol{r}} c_{\boldsymbol{k},\alpha}$$

- Now problem is simple: *Fourier transform, then diagonalize*

$$H_0 = J \sum_{\langle ij \rangle_\gamma} i c_i c_j = \frac{1}{2} \sum_{\boldsymbol{k}>0} (c_{-\boldsymbol{k},A} \; c_{-\boldsymbol{k},B}) \begin{pmatrix} 0 & f(\boldsymbol{k}) \\ f(\boldsymbol{k})^* & 0 \end{pmatrix} \begin{pmatrix} c_{\boldsymbol{k},A} \\ c_{\boldsymbol{k},B} \end{pmatrix}$$

$$f(\boldsymbol{k}) \equiv 2iJ\left(1 + e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_1} + e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_2}\right)$$

- Final dispersion has two bands:

$$\epsilon(\boldsymbol{k}) \equiv \pm |f(\boldsymbol{k})|$$

- Defines the ground state wave-function

We are done!

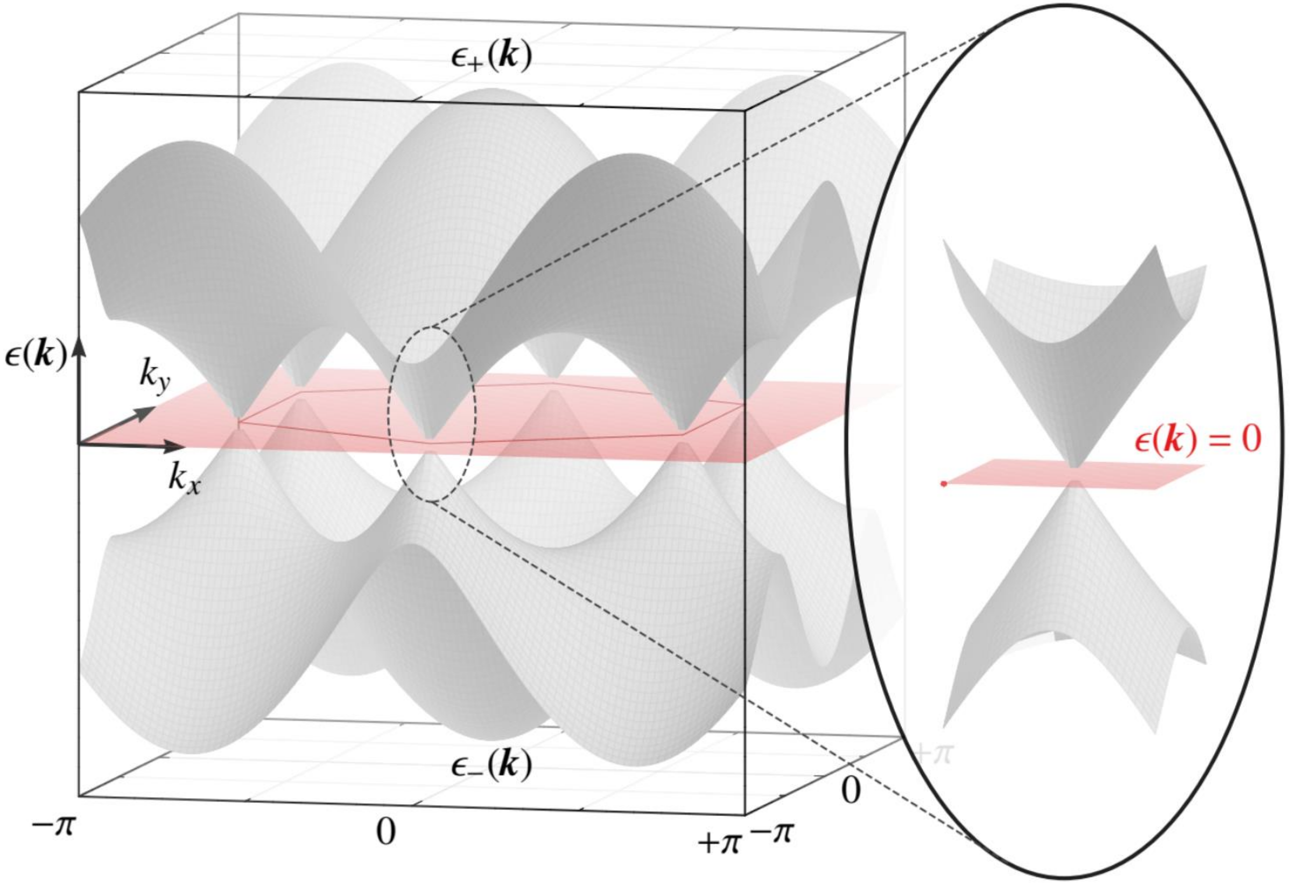
Flux-free spectrum

- What does the dispersion look like?

$$\epsilon(\boldsymbol{k}) \equiv \pm|f(\boldsymbol{k})|$$

$$f(\boldsymbol{k}) \equiv 2iJ\left(1 + e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_1} + e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_2}\right)$$

- **Dirac cones** near the corners o the Brillouin zone
- Same spectrum as graphene



Stable to (symmetric) perturbations

$$\epsilon(\boldsymbol{K} + \boldsymbol{q}) \approx \pm v|\boldsymbol{q}|$$

Kitaev, Ann. Phys. ***321****, 2 (2006)*

Anisotropic Kitaev model

- We solved the **isotropic** Kitaev model; can change coupling on bonds

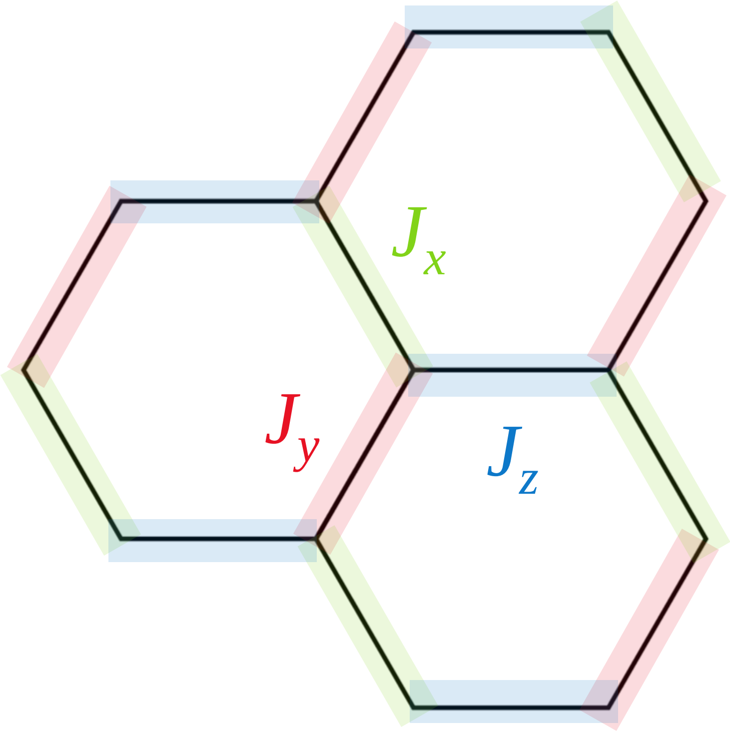
J_x, J_y, J_z

$$J\sum_{\langle ij\rangle_\gamma}\sigma_i^\gamma\sigma_j^\gamma \rightarrow \sum_{\langle ij\rangle_\gamma} J_\gamma \sigma_i^\gamma\sigma_j^\gamma$$

Different on different bonds

Exact solution proceeds *identically*

1. Plaquette symmetries
2. Link operators, gauge field
3. Flux sectors, Lieb's theorem



What changes?

Spectrum in flux-free sector

Anisotropic Kitaev Model (cont.)

- Still Majoranas hopping on honeycomb lattice, but now *bond-dependent*

$$f(\boldsymbol{k}) = iJ(1 + e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_1} + e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_2}) \rightarrow i(J_z + J_y e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_1} + J_x e^{-i\boldsymbol{k}\cdot\boldsymbol{a}_2})$$

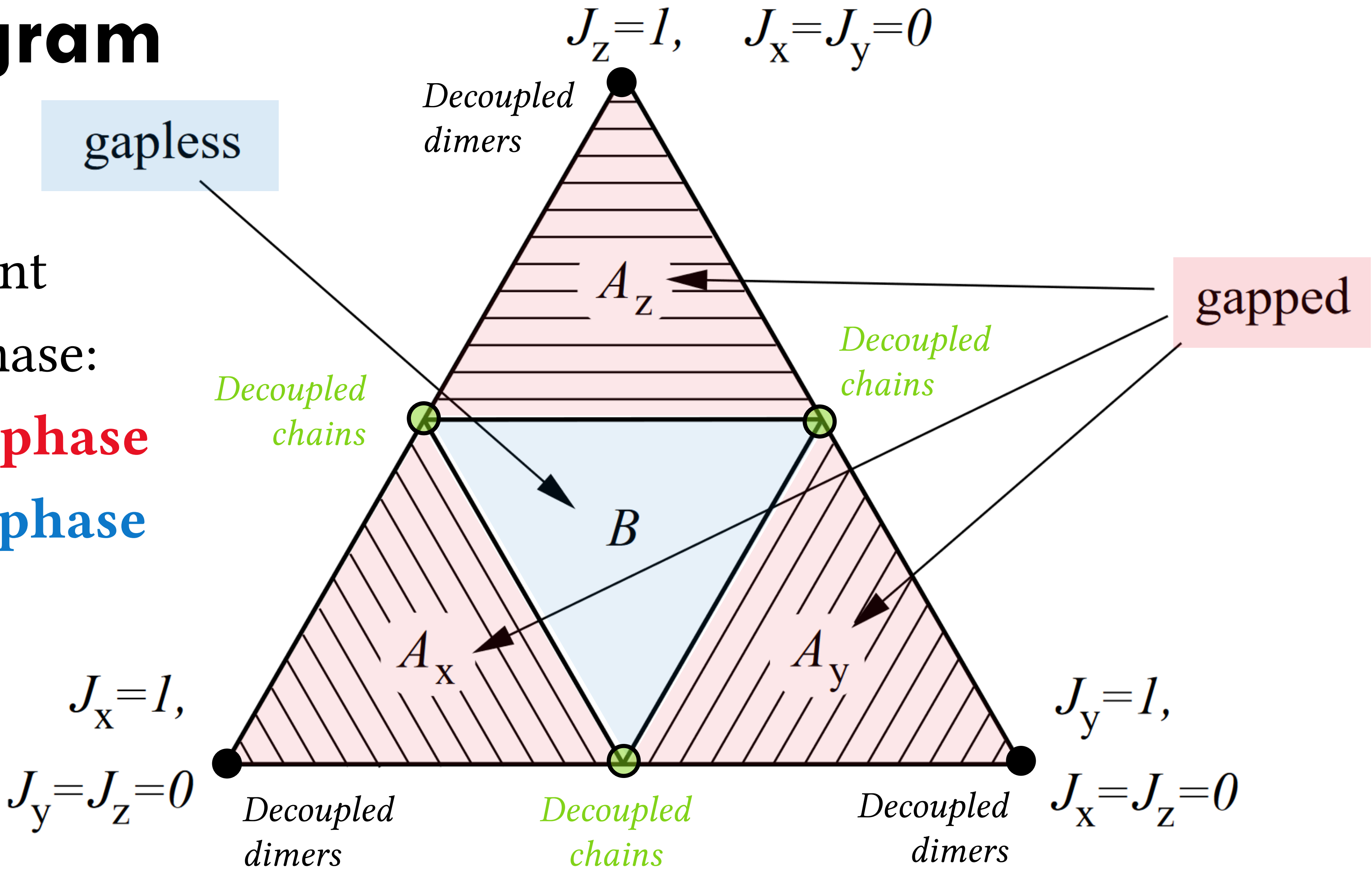
- Spectrum is still given by $\epsilon(\boldsymbol{k}) \equiv \pm|f(\boldsymbol{k})|$

How does this spectrum change when $J_x \neq J_y \neq J_z$?

Phase diagram

- *Sign* unimportant
- *Two* types of phase:

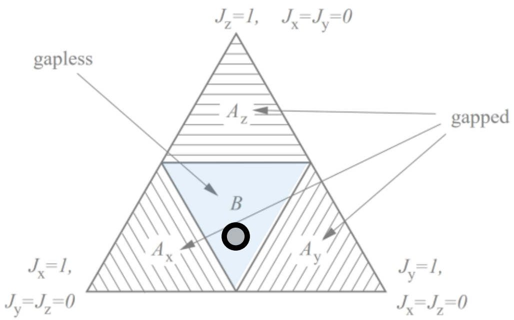
1. **Gapped "A" phase**
2. **Gapless "B" phase**



$$J_x + J_y + J_z = 1$$

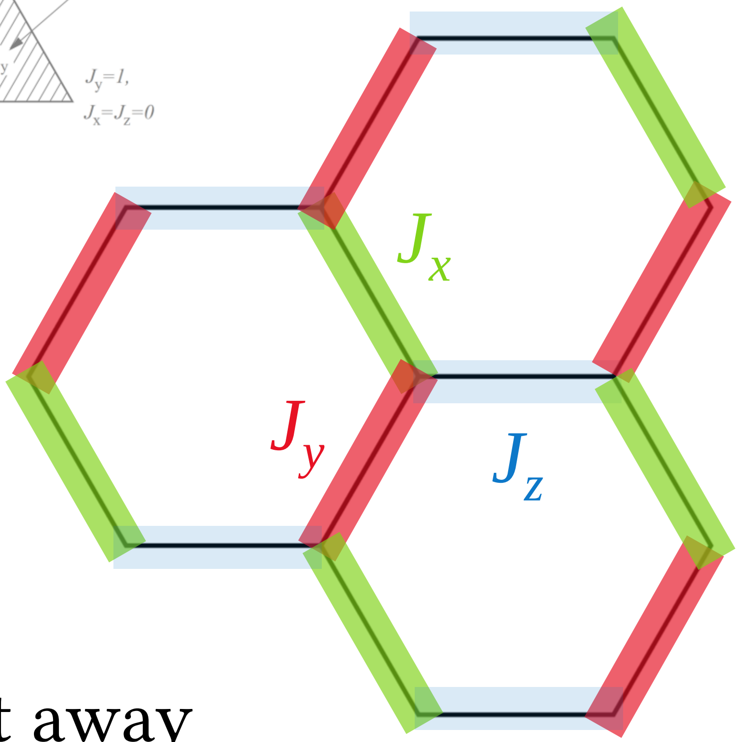
Kitaev, Ann. Phys. ***321****, 2 (2006)*

Gapless "B" Phase



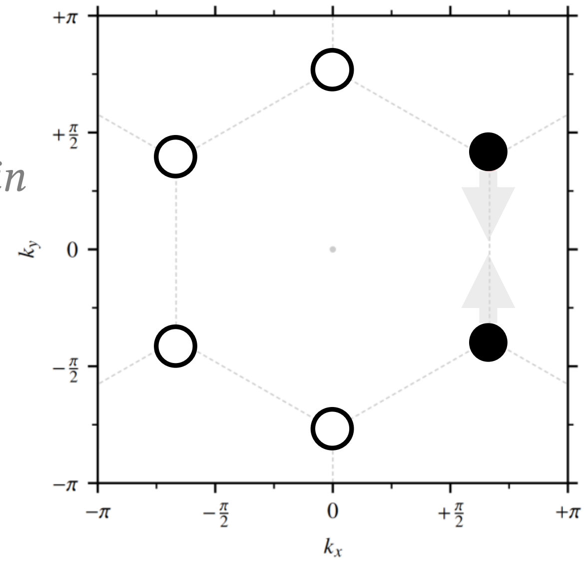
- **Isotropic phase** belongs to the "B" phase
- Small changes in couplings *don't* lift the Dirac cones

Symmetry protected!



$J_x, J_y \gtrsim J_z$

Brillouin zone



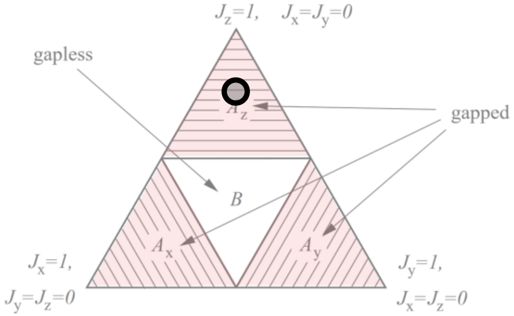
Dirac Cone

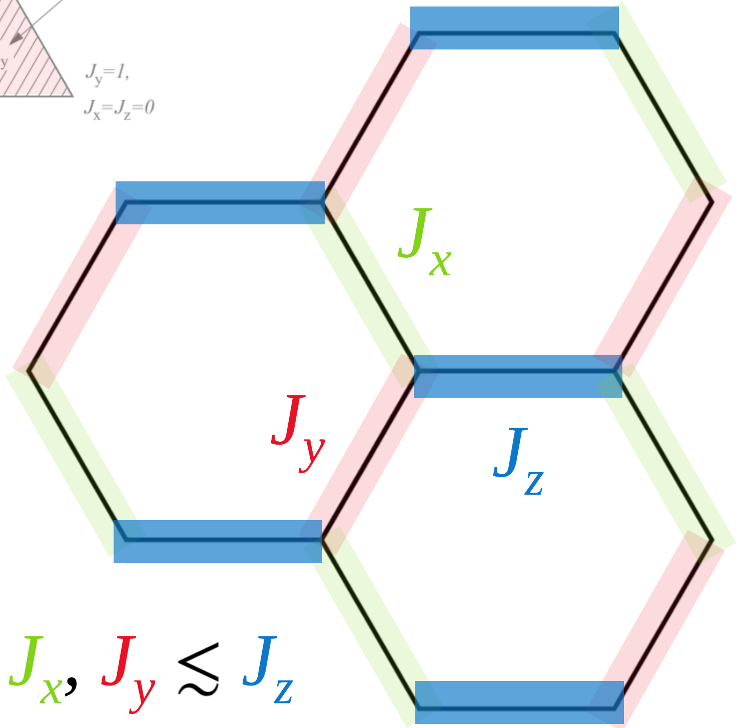
Dirac Cone

- **Dirac cones** shift away from the corners of the Brillouin zone

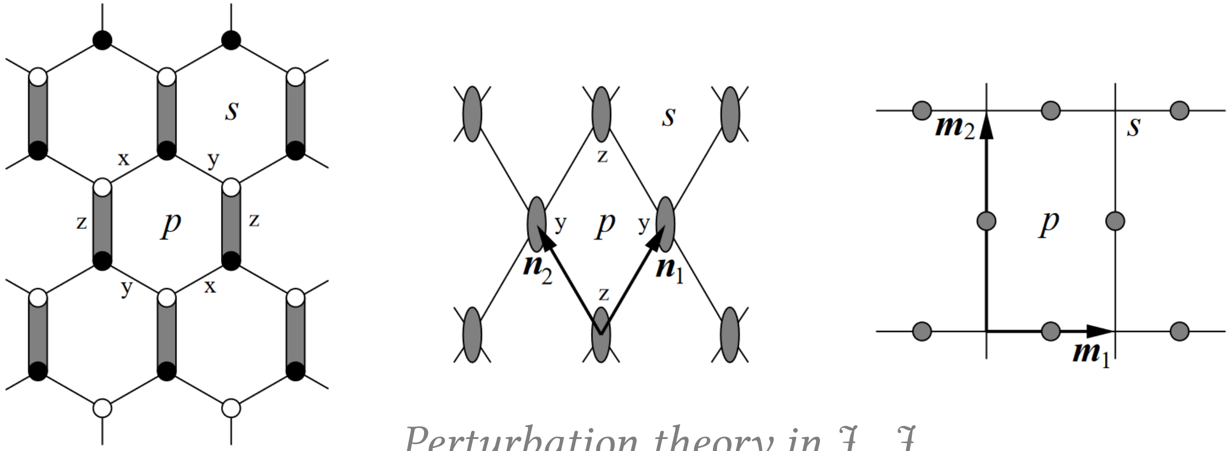
Gapped “A” Phase

- Make it anisotropic enough the **Dirac cones meet**
- They then *annihilate*, opening a **gap** in the spectrum
- Can be mapped to *toric code* model





$J_x, J_y \lesssim J_z$

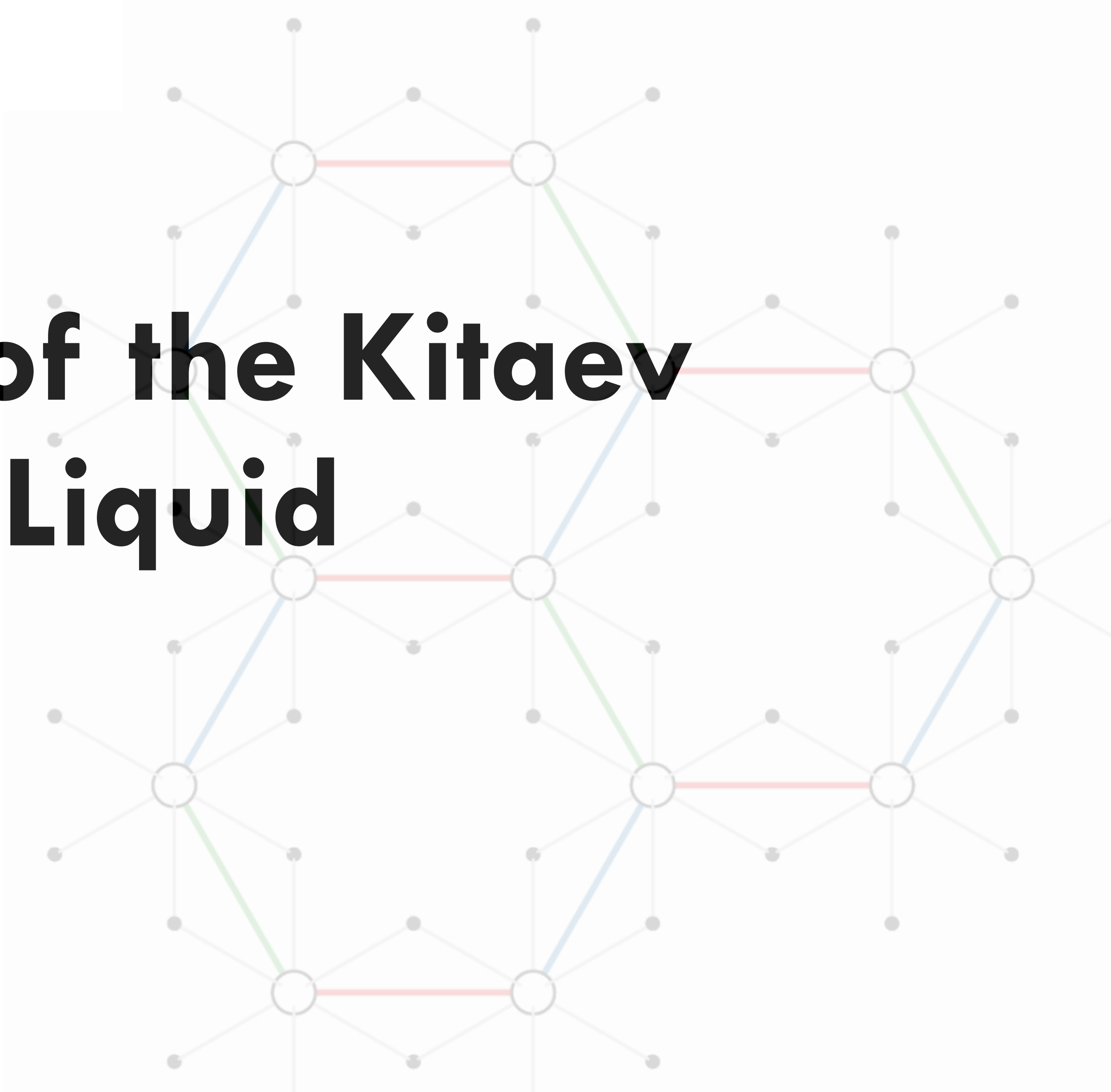


Perturbation theory in J_x, J_y

$$H_{\text{eff}} = -\frac{J_x^2 J_y^2}{16|J_z|^3} \sum_p Q_p$$

$$Q_p = \sigma^y_{\text{left}(p)} \sigma^y_{\text{right}(p)} \sigma^z_{\text{up}(p)} \sigma^z_{\text{down}(p)}$$

Kitaev, Ann. Phys. ***321***, *2 (2006)*



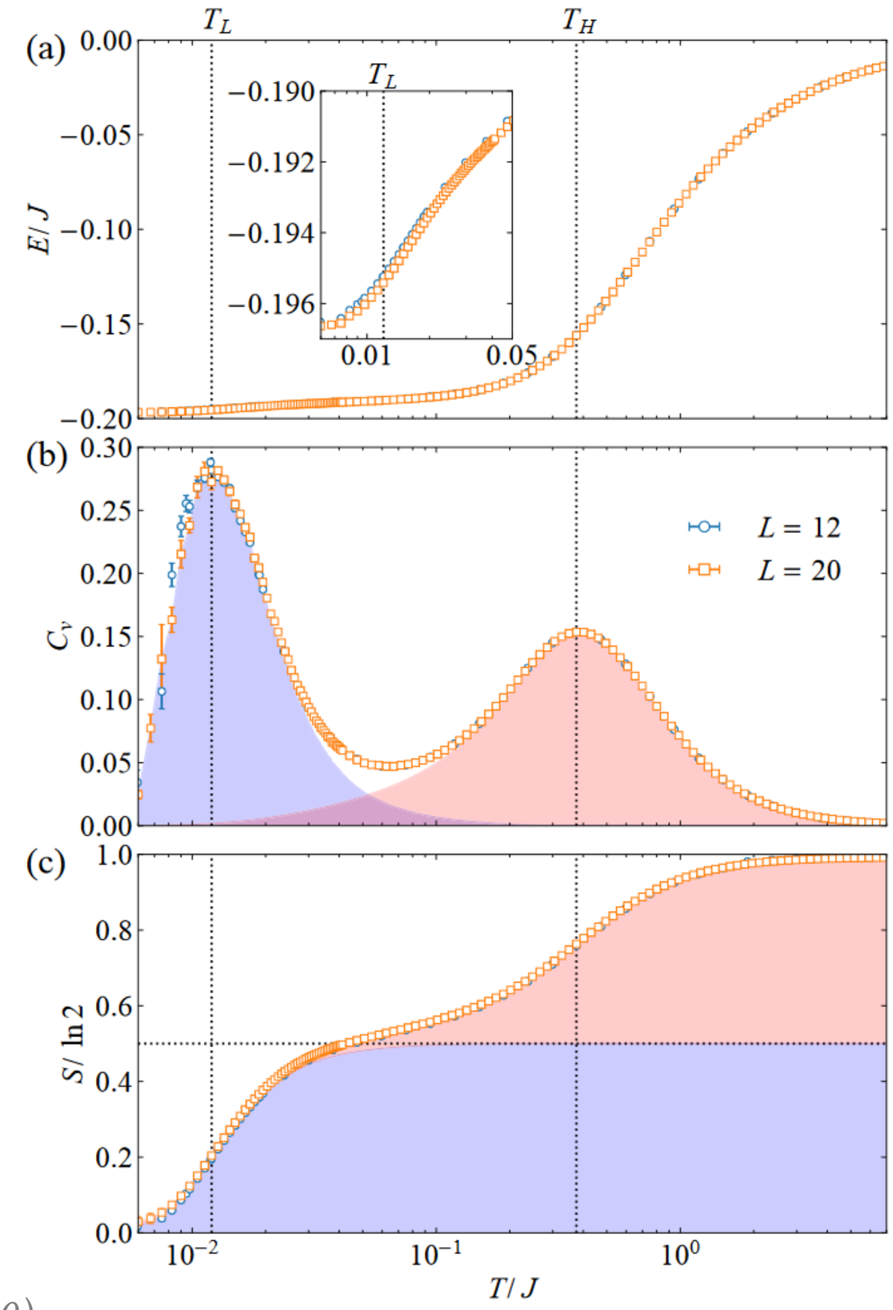
Properties of the Kitaev Spin Liquid

Thermodynamics:

- Structure from exact solution allows for Monte Carlo simulation at *finite temperature*

Roughly: Sample flux sectors, by solving fermionic problem in each sector

- *Note:* Practically uses Jordan-Wigner form of solution



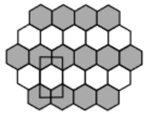
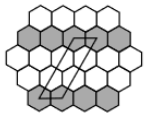
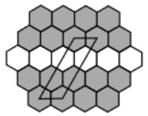
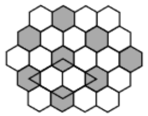
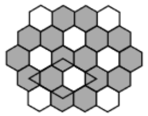
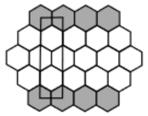
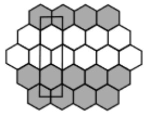
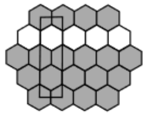
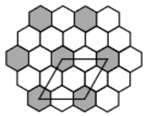
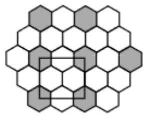
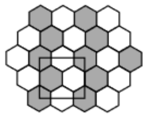
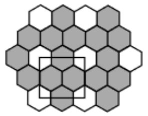
Nasu et al, Phys. Rev. ***92*** *115122 (2015); Motome & Nasu, JPSJ* ***89*** *012002 (2020)*

Excitations

$E_{\text{vortex}} \approx 0.1536, \quad \Delta E\Big($  $\Big) \approx -0.04, \quad \Delta E\Big($  $\Big) \approx -0.07.$

- Two classes of excitations
  1. **Majorana excitations:** Governed by dispersion in that flux sector
  2. **Flux Excitations:** *Add* non-zero fluxes to system

- *Intertwined:* Majoranas depends on the flux sector, flux sector energy depends on Majoranas

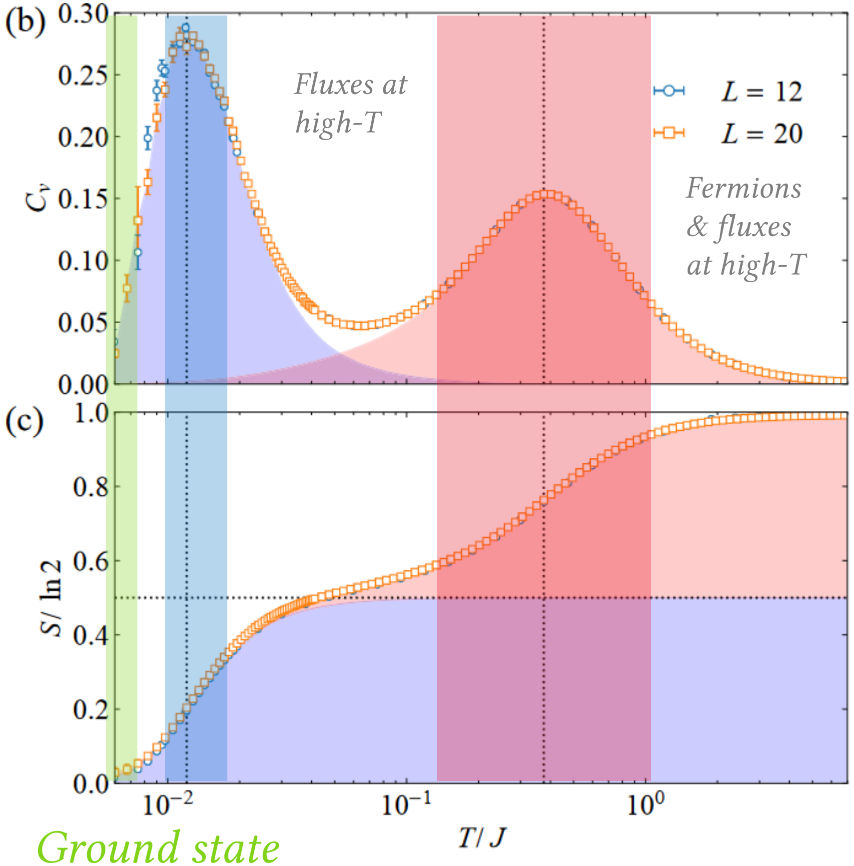
	Phase	Vortex density	Energy per ⬡ and per vortex
1		$\frac{1}{1}$	0.067 0.067
2		$\frac{1}{2}$	0.052 0.104
3		$\frac{1}{3}$	0.041 0.124
4		$\frac{2}{3}$	0.054 0.081
5		$\frac{1}{3}$	0.026 0.078
6		$\frac{2}{3}$	0.060 0.090
7		$\frac{1}{4}$	0.034 0.136
8		$\frac{2}{4}$	0.042 0.085
9		$\frac{3}{4}$	0.059 0.078
10		$\frac{1}{4}$	0.042 0.167
11		$\frac{3}{4}$	0.074 0.099
12		$\frac{1}{4}$	0.025 0.101
13		$\frac{2}{4}$	0.046 0.092
14		$\frac{3}{4}$	0.072 0.096

Kitaev, Ann. Phys. ***321****, 2 (2006)*

Thermodynamics (cont.):

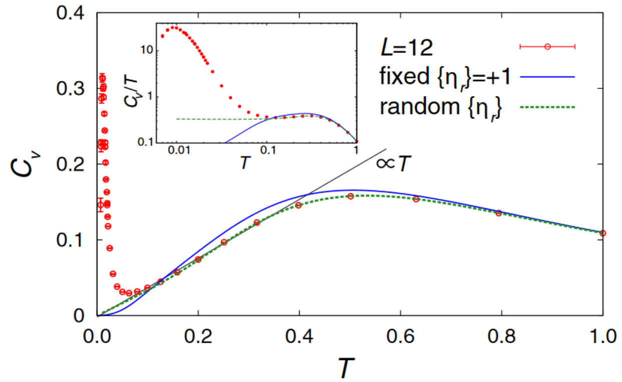
- Can understand in terms of two energy scales:
  1. **Fermionic scale:** Spins have fractionalized into Majoranas, fluxes are *disordered* ~ *O(J)*
  2. **Flux scale:** Flux excitations no longer populated, settle into flux-free sector ~ *O(flux gap)*
- At *each* of these, release ~ log(2)/2 entropy per spin

Flux Scale　　*Fermionic Scale*



Ground state physics

Looks like Majoranas in random flux background



Nasu et al, Phys. Rev. ***92*** *115122 (2015); Motome & Nasu, JPSJ* ***89*** *012002 (2020)*

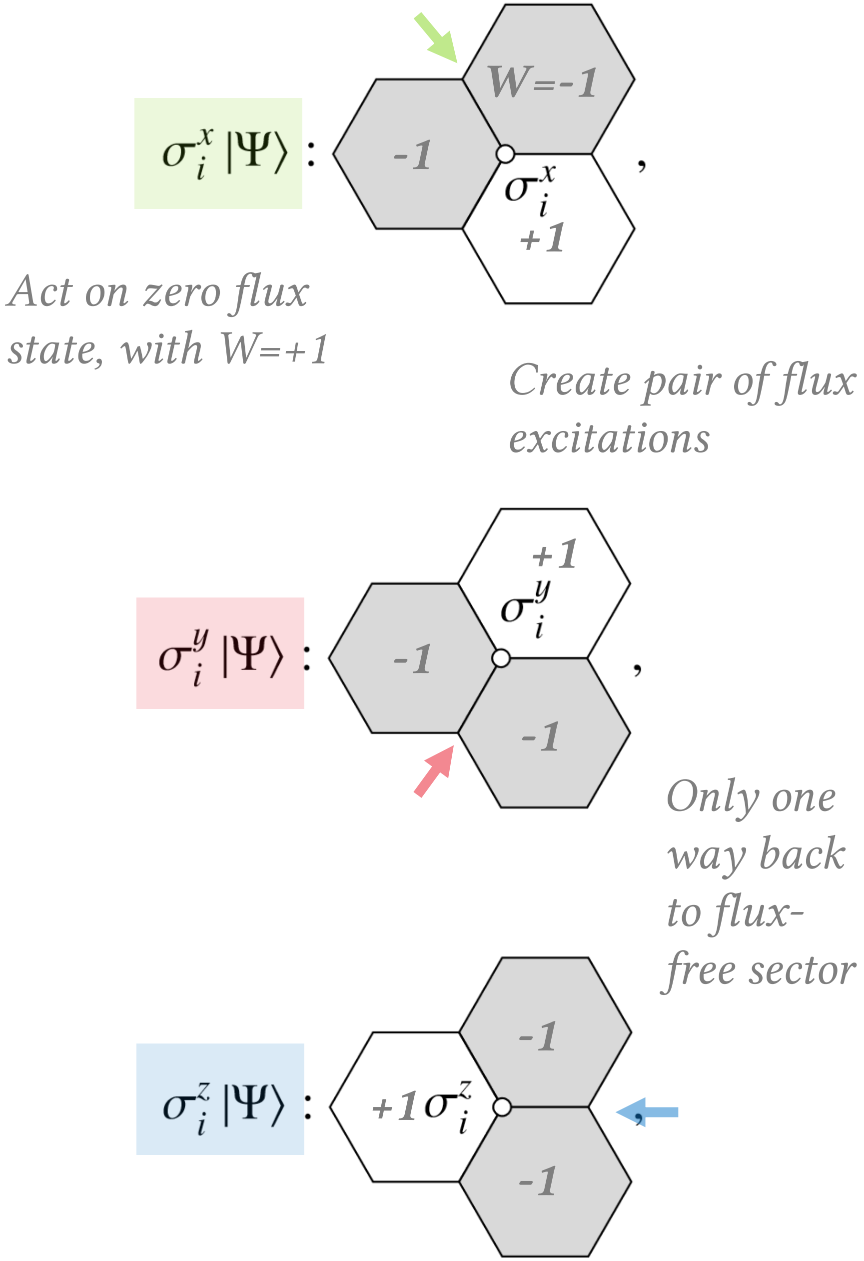
Spin correlations:

- *Static* spin-spin correlations are **ultra-short range**

$$\langle \sigma_i^\gamma \sigma_j^\gamma \rangle = \begin{cases} \neq 0, & \langle ij \rangle \in \gamma \\ = 0, & \langle ij \rangle \notin \gamma \end{cases}$$

- Consequence of *plaquette symmetries*
- At isotropic point? *single correlation function*
- Also holds for **dynamical** correlator

$$\langle \sigma_i^\gamma(t) \sigma_j^\gamma \rangle$$



Baskaran et al, Phys. Rev. Lett. ***98****, 247201 (2007)*

Dynamics?

- Can compute from exact solution; **hard**, must deal with two-flux excitations

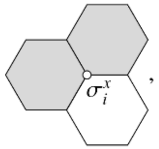
Remove flux pair + c-fermion — *Evolve with fluxes* — *Add flux pair + c-fermion*

$$\langle \sigma_i^\gamma(t)\sigma_j^\gamma\rangle = e^{iE_0t}\langle\Psi_0|\sigma_i^\gamma e^{-iHt}\sigma_j^\gamma|\Psi_0\rangle$$

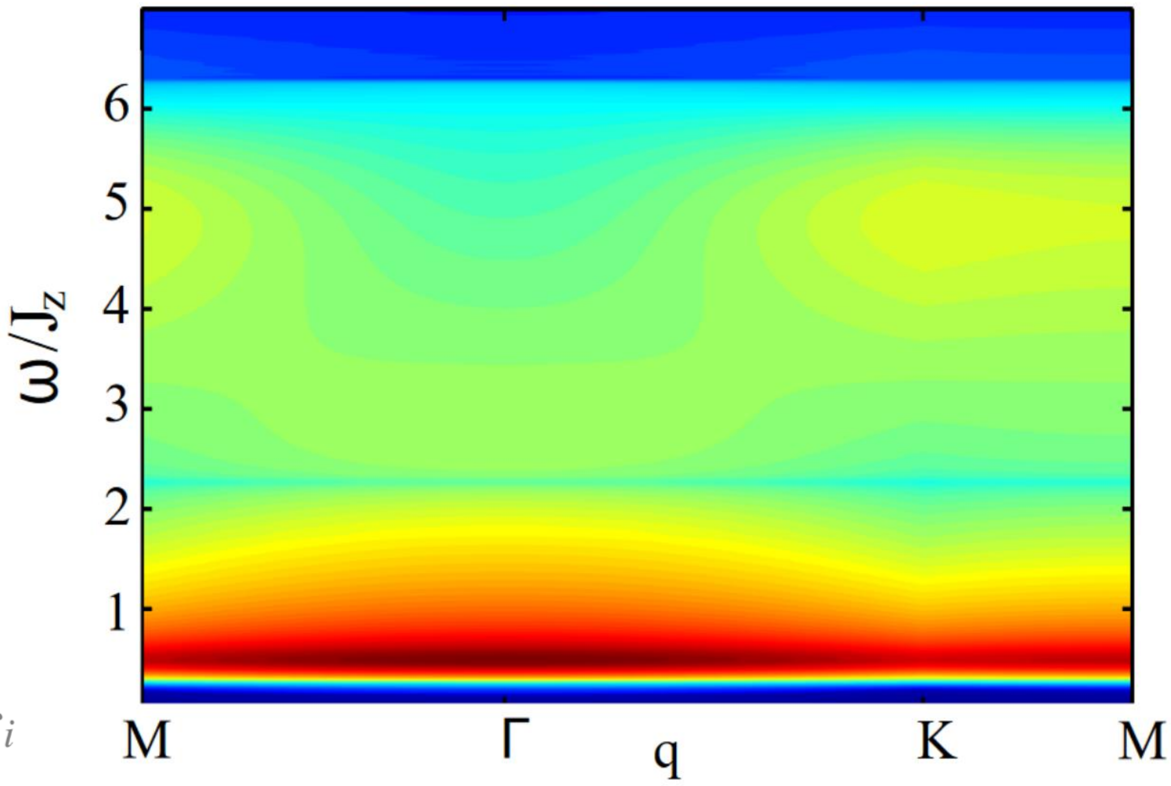
$\sigma_i \equiv i b_i c_i$

$$= e^{iE_0t}\langle\tilde\Psi_0|c_i e^{-iH[u_{\text{pair}}]t} c_j|\tilde\Psi_0\rangle$$

Sector with pair of fluxes



- Related to *X-ray edge problem*



$$S(\boldsymbol{q},\omega) \propto \sum_\gamma \sum_{ij} \int dt\, e^{i\boldsymbol{q}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)-i\omega t}\langle\sigma_i^\gamma(t)\sigma_j^\gamma\rangle$$

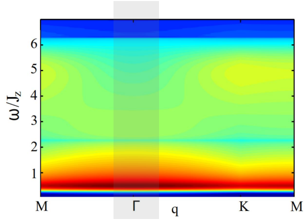
Fourier-transform of spin-spin correlator

Knolle et al, Phys. Rev. Lett ***112*** *207203 (2014)*

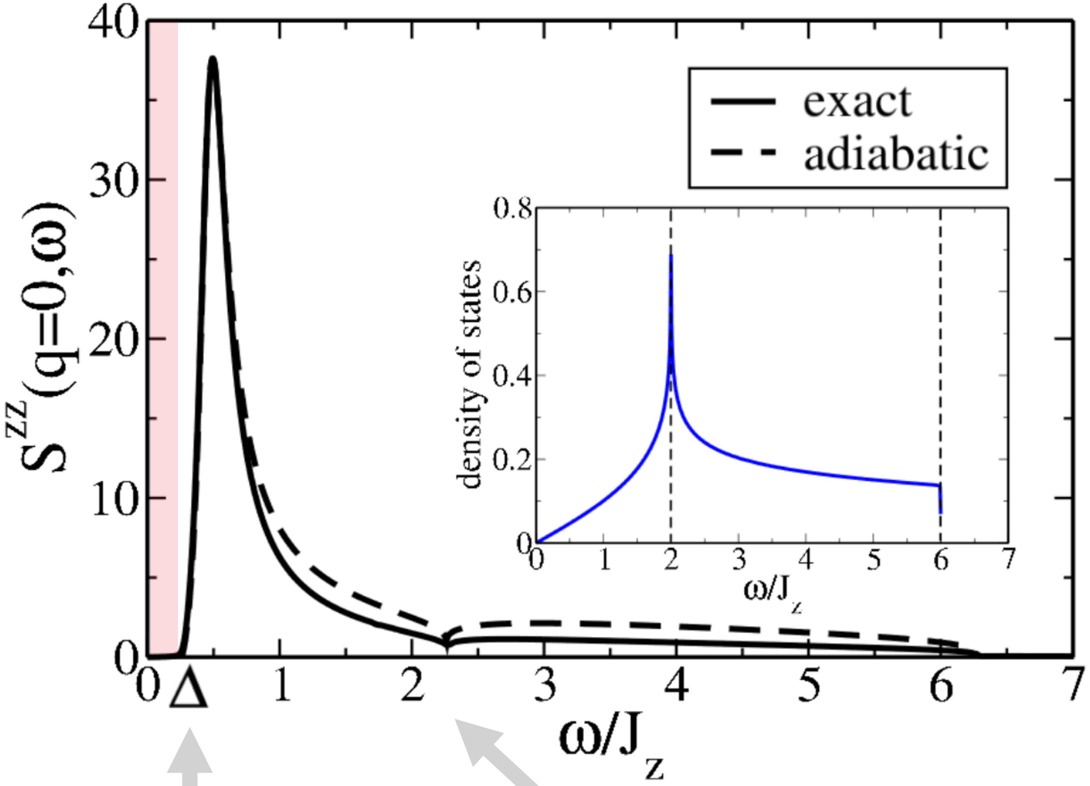
Dynamics (cont.):

- Dirac cones *not* directly visible, no flux change
- Clear **gap** corresponding to energy cost to create pair of flux excitations
- **Continuum** of intensity going out energies of ~*O(J)*

Energy scale of Majorana dispersion



No response



Gap to creating flux pairs

Continuum of Majorana excitations

Knolle et al, Phys. Rev. Lett **112** 207203 (2014)

Effect of a Magnetic Field

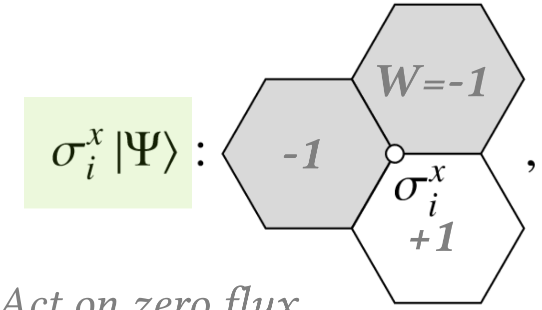
Effect of a magnetic field

- Application of magnetic field gaps out Majorana fermions

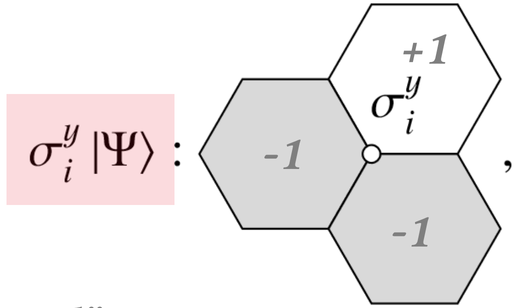
$$-J\sum_{\langle ij\rangle_\gamma}\sigma_i^\gamma\sigma_j^\gamma - \sum_i \boldsymbol{h}\cdot\boldsymbol{\sigma}_i$$

Couples to magnetization operator

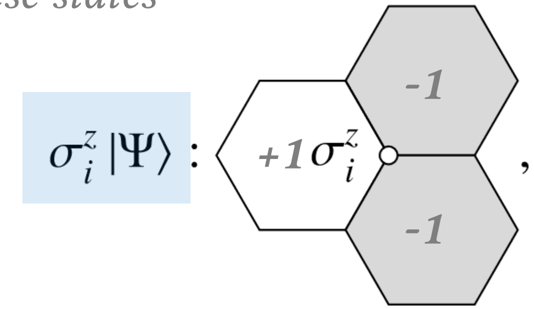
- *No longer exactly solvable*
- *Individual* spin operators change flux sector
- **Can do (quasi-) degenerate perturbation theory within zero flux sector**



Act on zero flux state, with W=+1



"Virtual" processes involve these states



Kitaev, Ann. Phys. ***321****, 2 (2006)*

Second-order corrections

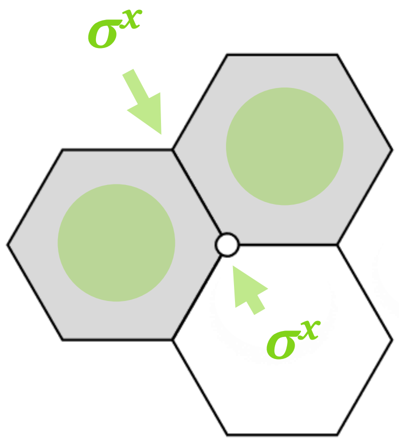
Second order in field, generates *renormalization of Majorana hoppings*

- Does *not* explicitly break time-reversal
- Gives *finite* susceptibility at *T=0*
- Isotropic model preserved for [111] field

Energy required to excite flux pair

$$H_{\text{eff}} = -\sum_{\langle ij\rangle_\gamma}\left(J + \frac{2h_\gamma^2}{\Delta}\right) P_0 \sigma_i^\gamma \sigma_j^\gamma P_0 \quad + \text{const.}$$

Project into zero-flux sector

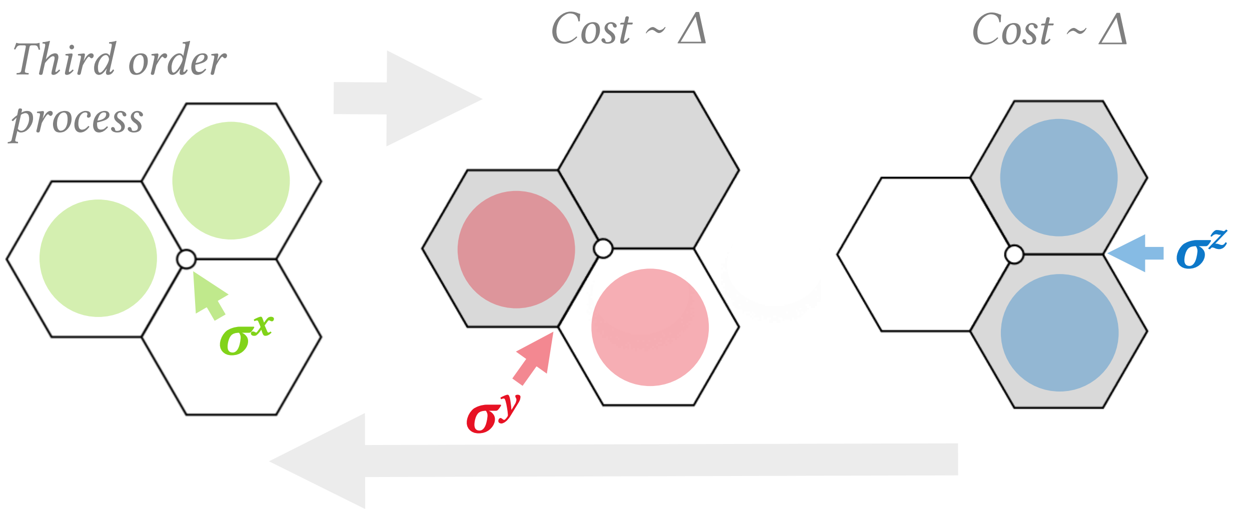


A. Willans, Thesis (2010); Kitaev, Ann. Phys. ***321****, 2 (2006)*

Third-order corrections

- At *next* order?
- Most important piece at *third*-order: **generates a three-spin interaction term**
- *Explicitly* breaks time-reversal symmetry

What does this do?



Energy required to excite flux pair

$$H_{\rm eff} = -\sum_{\langle ij\rangle_\gamma}\left(J + \frac{2h_\gamma^2}{\Delta}\right) P_0\sigma_i^\gamma\sigma_j^\gamma P_0$$

Project into zero-flux sector

Still solvable!

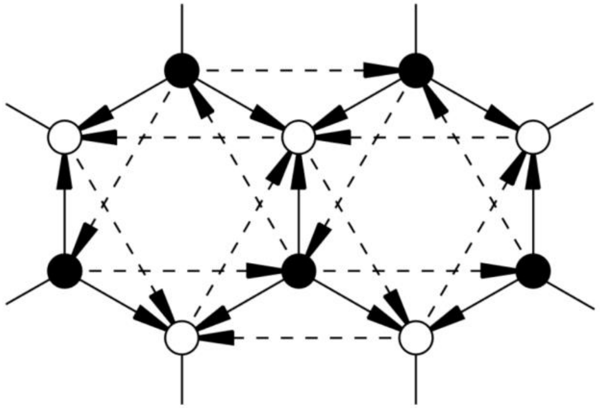
$$-\frac{6h_xh_yh_z}{\Delta^2}\sum_{i,j,k}\left[P_0\sigma_i^x\sigma_j^y\sigma_k^z P_0\right]$$

Kitaev, Ann. Phys. ***321****, 2 (2006)*

Effect of a magnetic field (cont.)

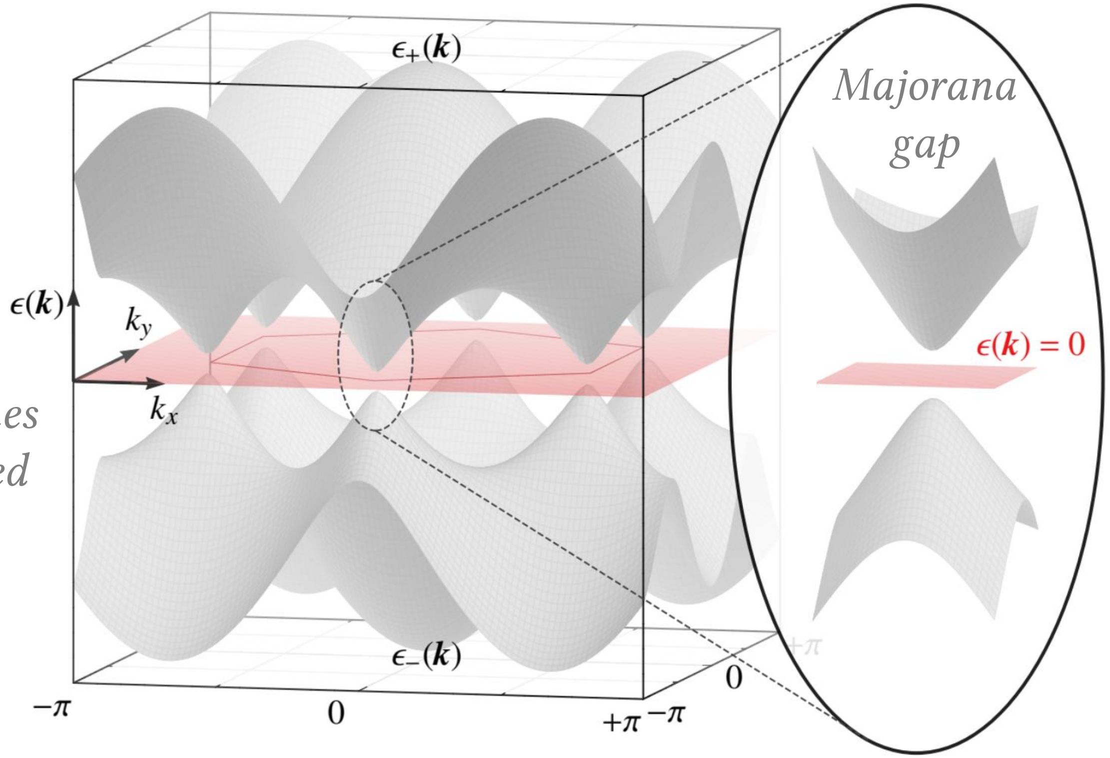
$$\Delta \sim \frac{h_x h_y h_z}{J^2}$$

- Third-order term appears as *second-neighbour hopping*



- *Identical* in form to Haldane-type model
- *Topological bands;* ***chiral Majorana edge modes***

Dirac cones are gapped out



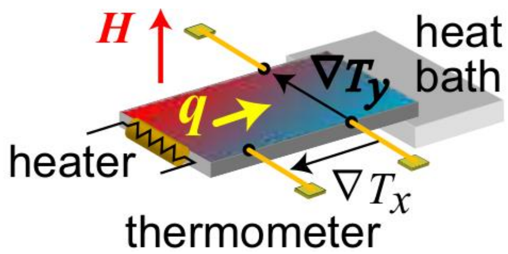
Spectrum near cones

$$\varepsilon(\mathbf{q}) \approx \pm\sqrt{3J^2\,|\delta\mathbf{q}|^2 + \Delta^2}$$

Majorana "mass"

Haldane, Phys. Rev. Lett. ***61*** *2015 (1988); Kitaev, Ann. Phys.* ***321****, 2 (2006)*

Thermal Hall Effect



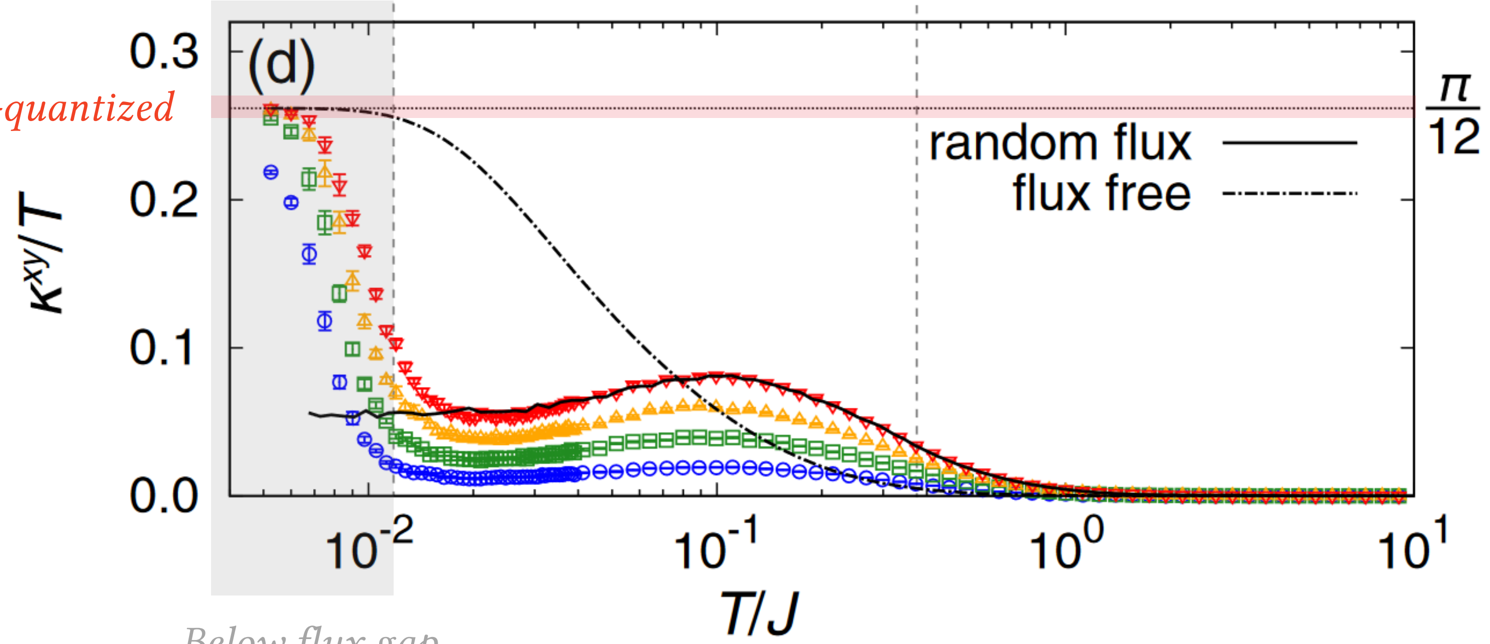
- Chiral edge modes give *half*-quantized thermal Hall effect

$$\frac{\kappa_{xy}}{T} = q\frac{\pi}{6}\frac{k_B^2}{\hbar}$$

(Chiral) central charge of edge modes

From QMC simulations

Half-quantized



Below flux gap

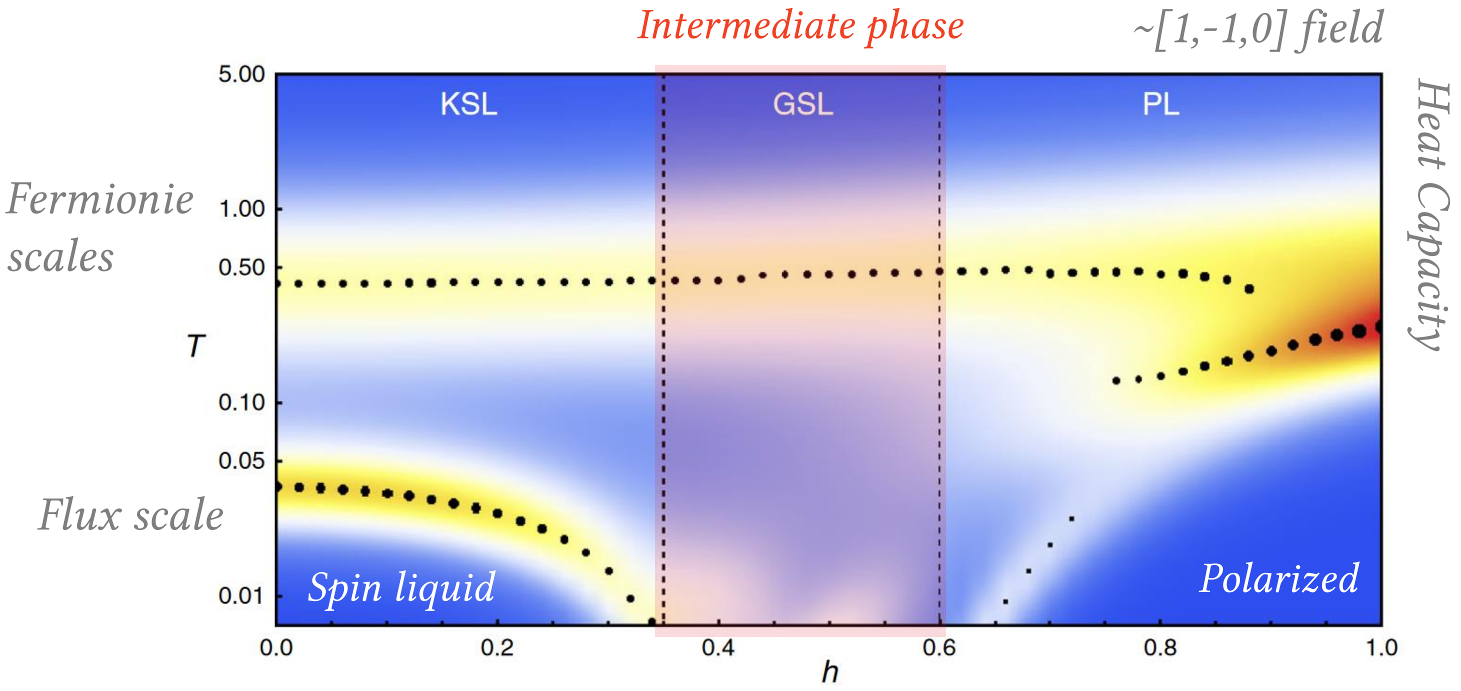
- For Majoranas *q=1/2*

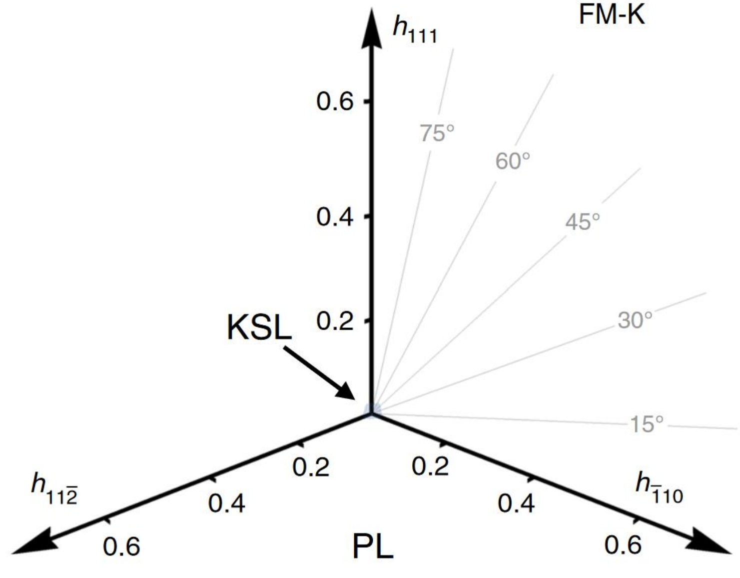
"Half" degree of freedom

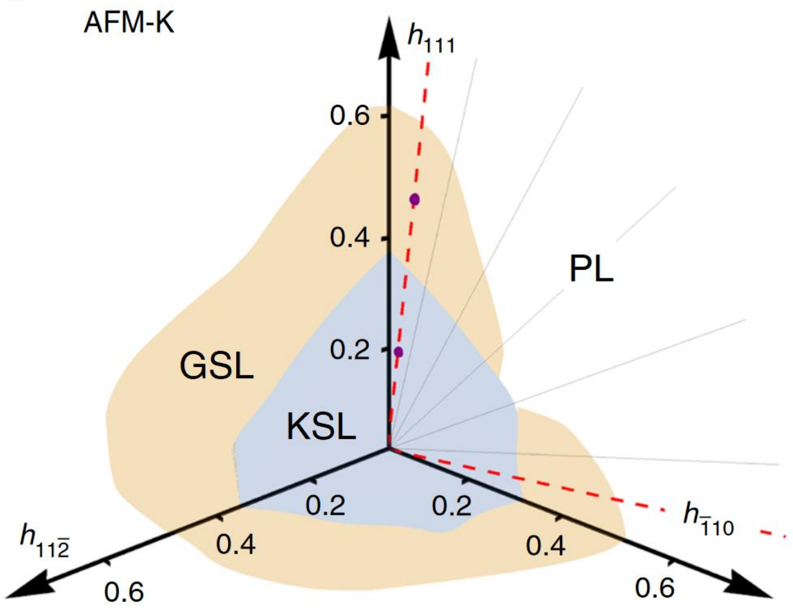
Nasu et al, Phys. Rev. Lett. **119**, 127204 (2017); Kitaev, Ann. Phys. **321**, 2 (2006)

Larger fields?

- *FM Kitaev?* Quickly to *polarized phase*
- *AF Kitaev?* Larger region, **intermediate phase (?)**







Hickey & Trebst, Nat. Comm. **10**, 530 (2019)

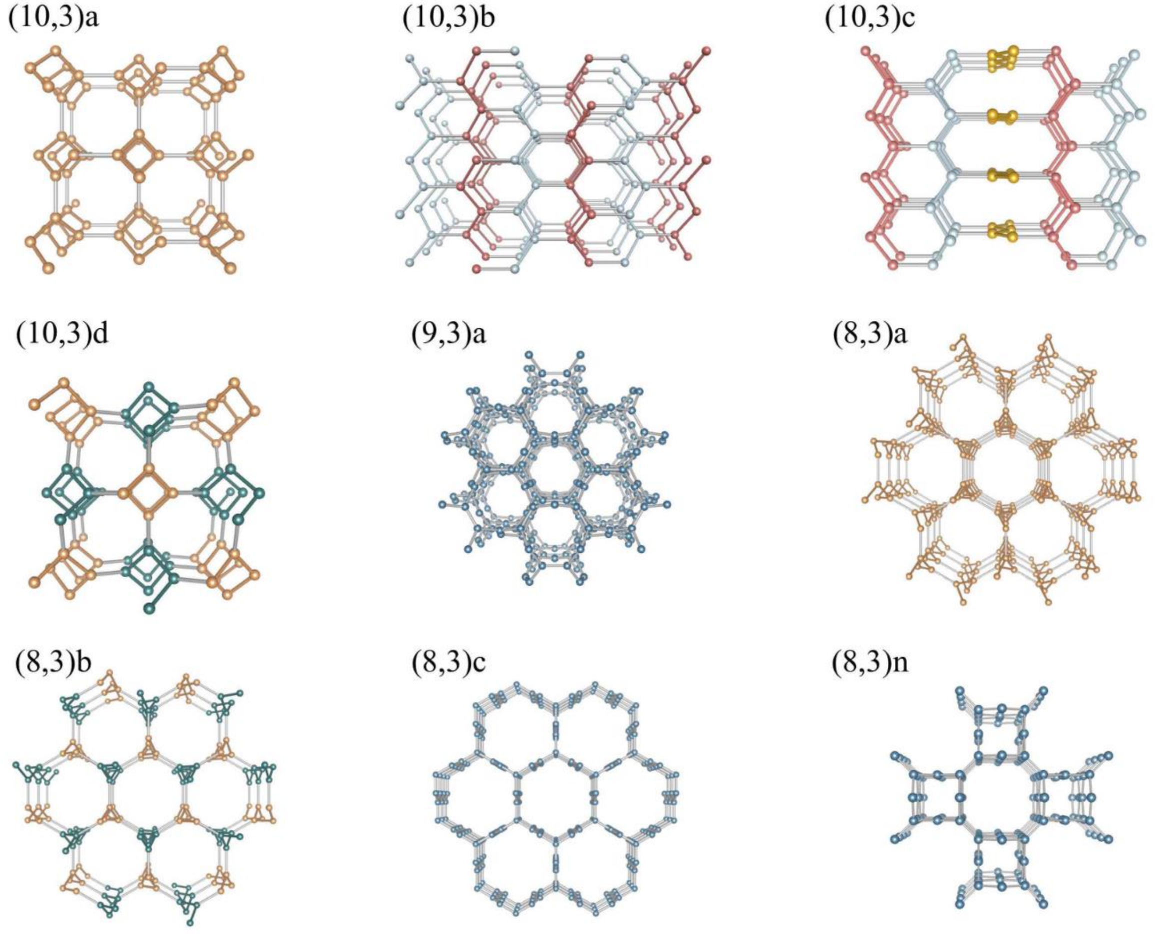
Generalizations

Three-dimensional Kitaev models

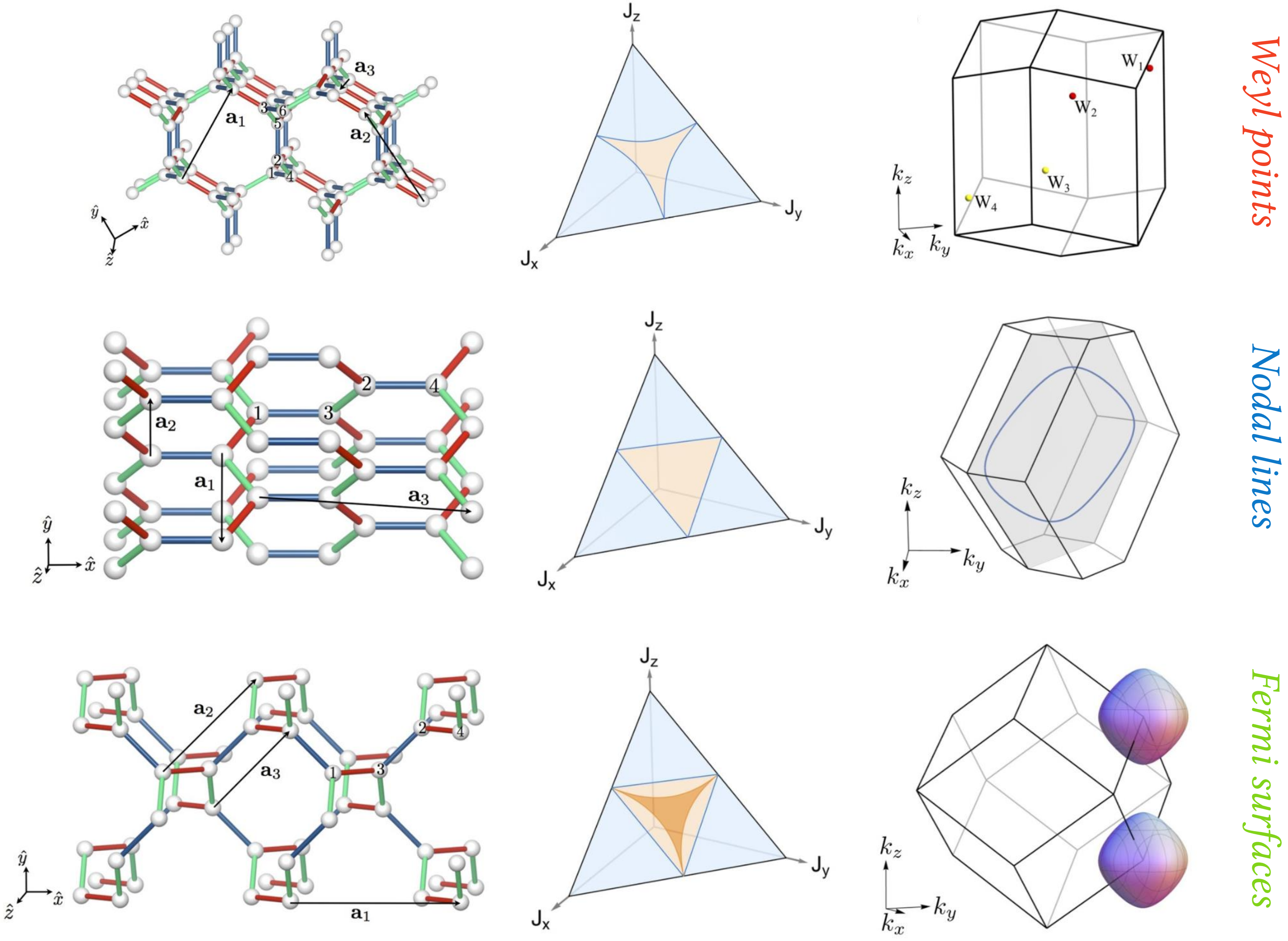
- Solvable on many tri-coordinated lattices – two *and* three dimensional
  - Star lattice (2D)
  - Hyperhoneycomb
  - Hyperoctagon
  - Stripy-honeycomb …

- Derivation is *mostly* identical

All of three bond types



Trebst, Kitaev Materials 1701.07056 (Review)



- Variety of ground state flux-sectors (*not necessarily flux-free*)
- Variety of Majorana spectra
  1. Weyl points
  2. Nodal Lines
  3. Majorana-Fermi surfaces*

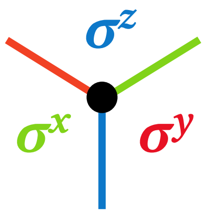
**Can be unstable to interactions*

*O'Brien et al, Phys. Rev. B **93** 085101 (2016)*

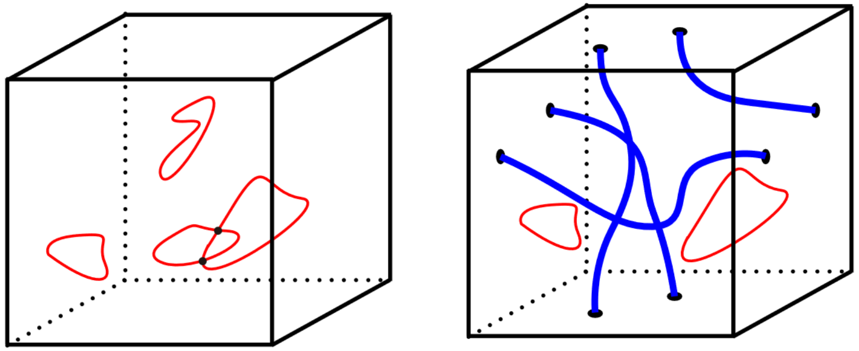
Thermal phase transitions

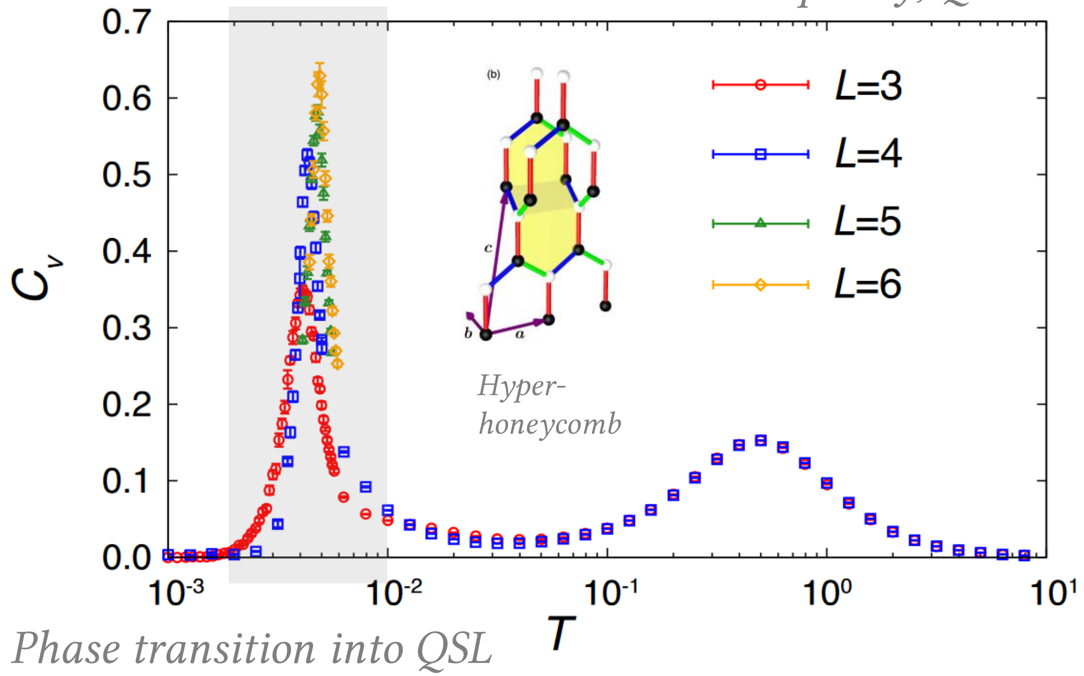
- Additional constraints on *plaquette operators*

$$\prod_{p \in \text{volume}} W_p = 1$$



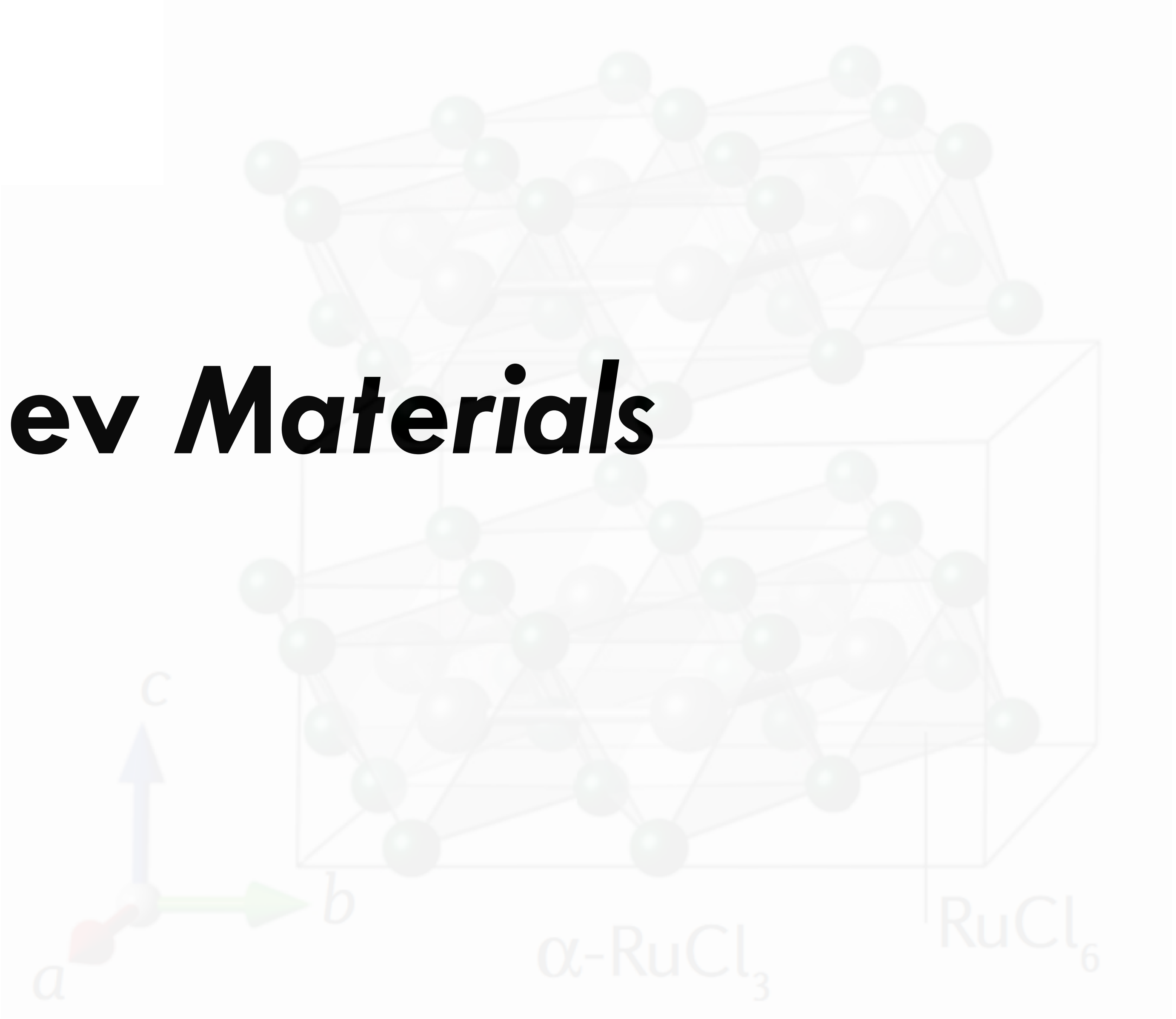
- Flux excitations form *loops*
- Confinement-deconfinement transition at finite temperature





Nasu et al, Phys. Rev. Lett. ***113*** *197205 (2014)*

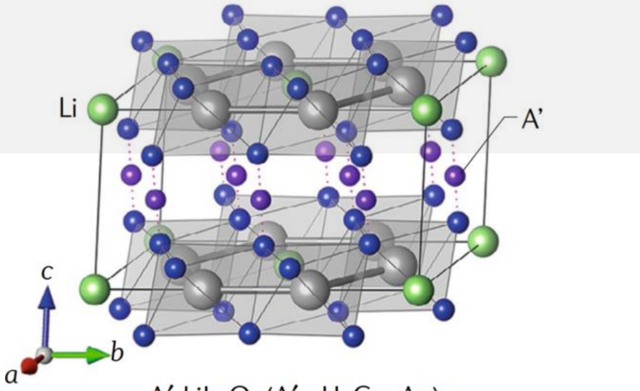
Kitaev *Materials*



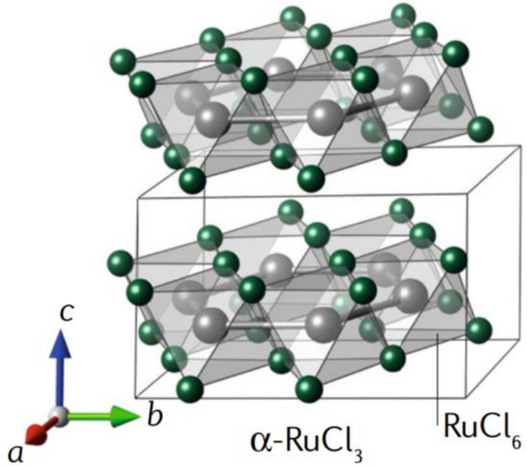
Kitaev *Materials*

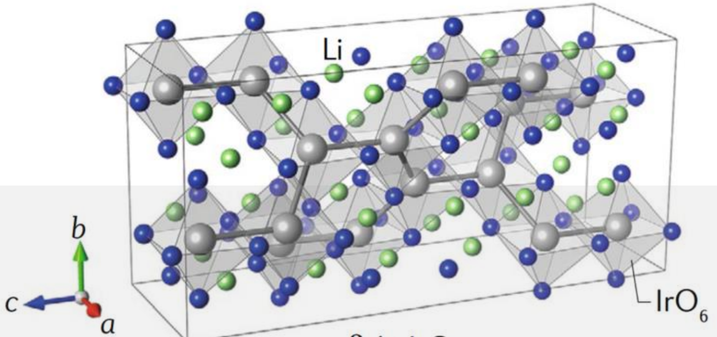
Growing family where Kitaev interaction is believed to be *dominant*:

1. **α-$RuCl_3$**
2. Na_2IrO_3
3. α-Li_2IrO_3, β-Li_2IrO_3, γ-Li_2IrO_3
4. $H_3LiIr_2O_6$
5. Cu_2IrO_3
6. $Cu_3LiIr_2O_6$, $Ag_3LiIr_2O_6$, …



$A'_3LiIr_2O_6$ (A' = H, Cu, Ag)



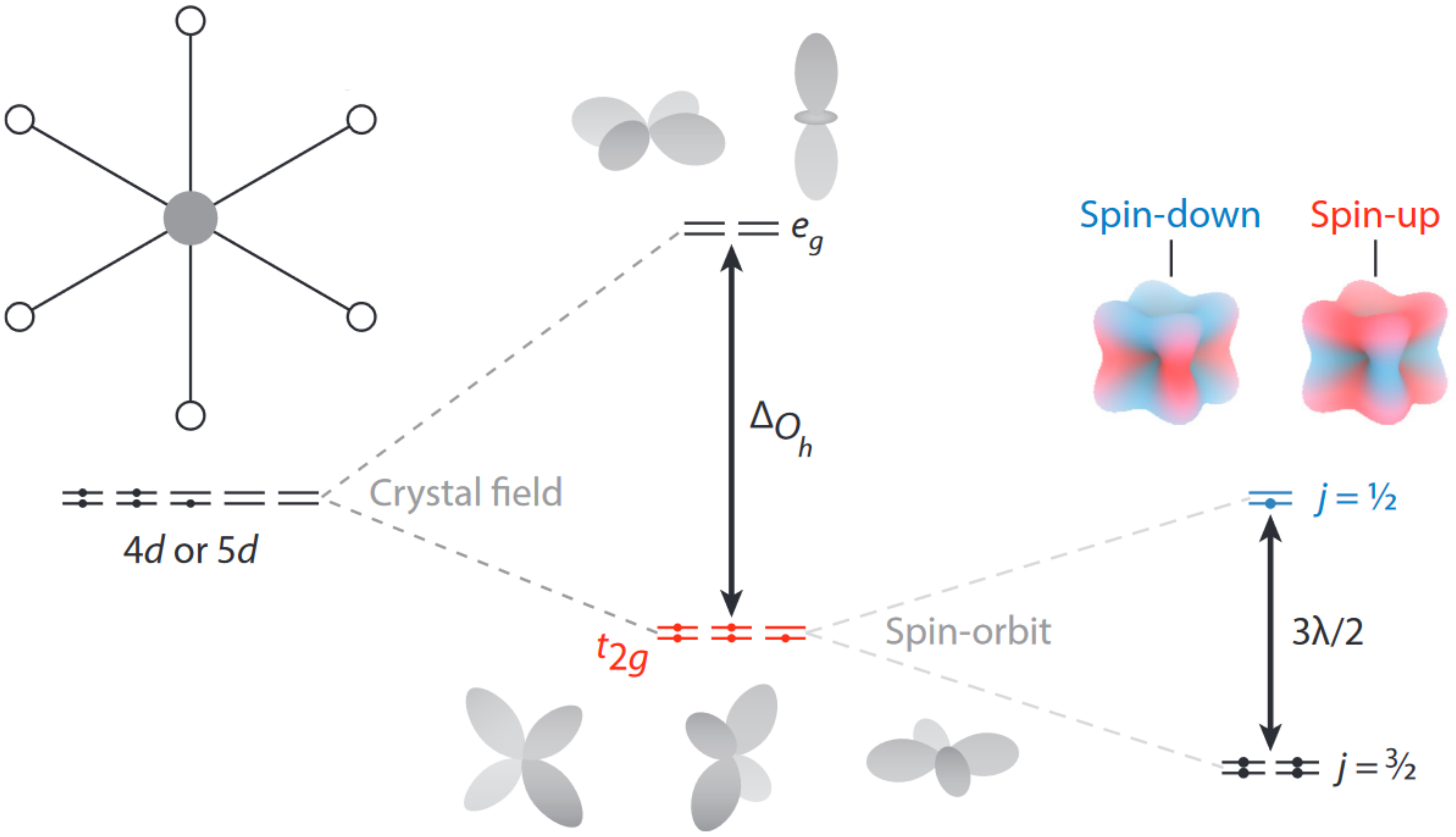


β-Li_2IrO_3

Takagi et al., Nature Reviews Physics ***1****, 264–280 (2019)*

J_{eff} = ½ Magnetism

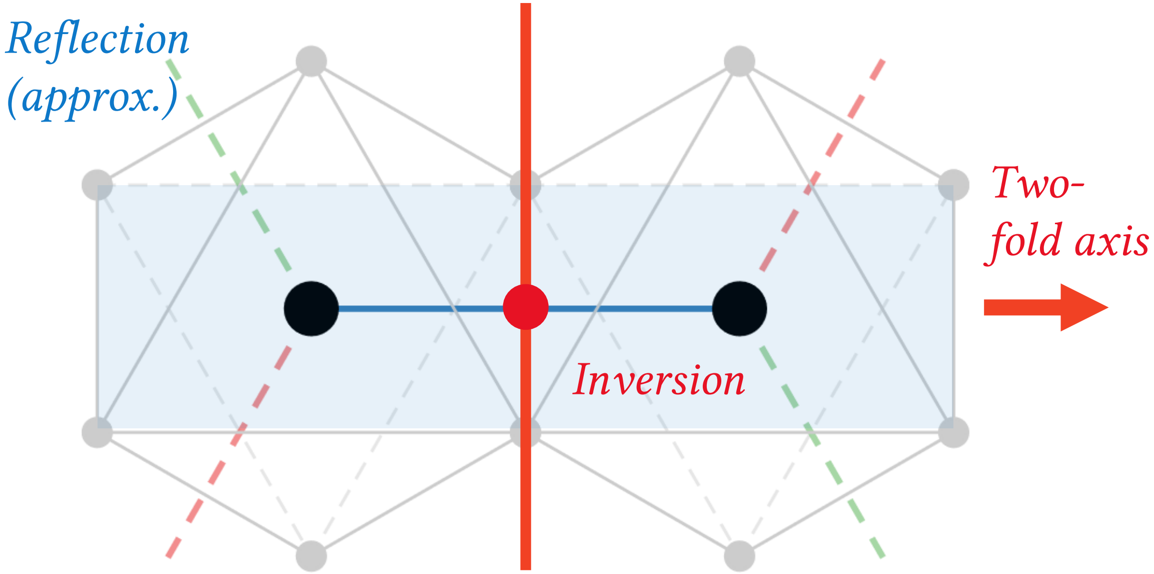
- Partially filled *d*-shell with strong spin-orbit coupling
- Low-lying *half-filled* doublet
- Doublet states strongly mix *spin* and *orbital* degrees of freedom



Rau, *Lee & Kee, Ann. Rev. Cond. Mat. 7, 195-221 (2016)*

Symmetry allowed exchanges

- *Edge*-shared octahedra
- Bond symmetries constrain exchanges: ***Four allowed***



Heisenberg | *Kitaev* | *Symmetric Off-diagonal*

$$J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^z S_j^z + \Gamma(S_i^x S_j^y + S_i^y S_j^x)$$

$$+\Gamma'(S_i^x S_j^z + S_i^z S_j^x + S_i^y S_j^z + S_i^z S_j^y)$$

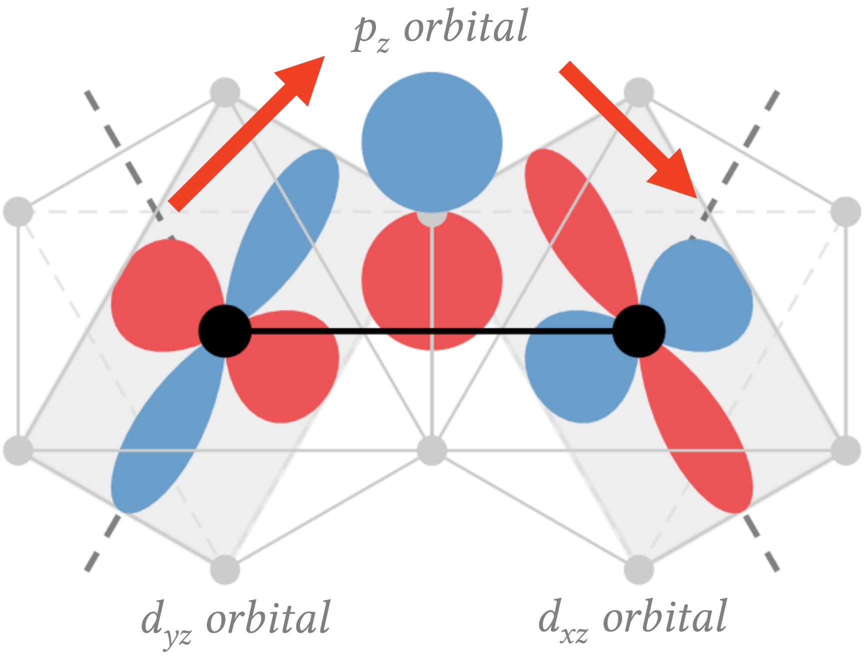
Symmetric Off-diagonal

*Only if **pair** of **ideal** octahedra*

Katakuri et al., New. J. Phys. ***16,*** *013056 (2014)*
Rau**, Lee & Kee, Phys. Rev. Lett.* ***112, *077204 (2014)*

Jackeli-Khaliullin Mechanism

- Exchange known *not* be generic in reasonable limit
- *Ligand* mediated hopping is dominant
- *Ferromagnetic* **Kitaev interaction** is leading exchange



Is this the dominant piece?

Hund's Coupling

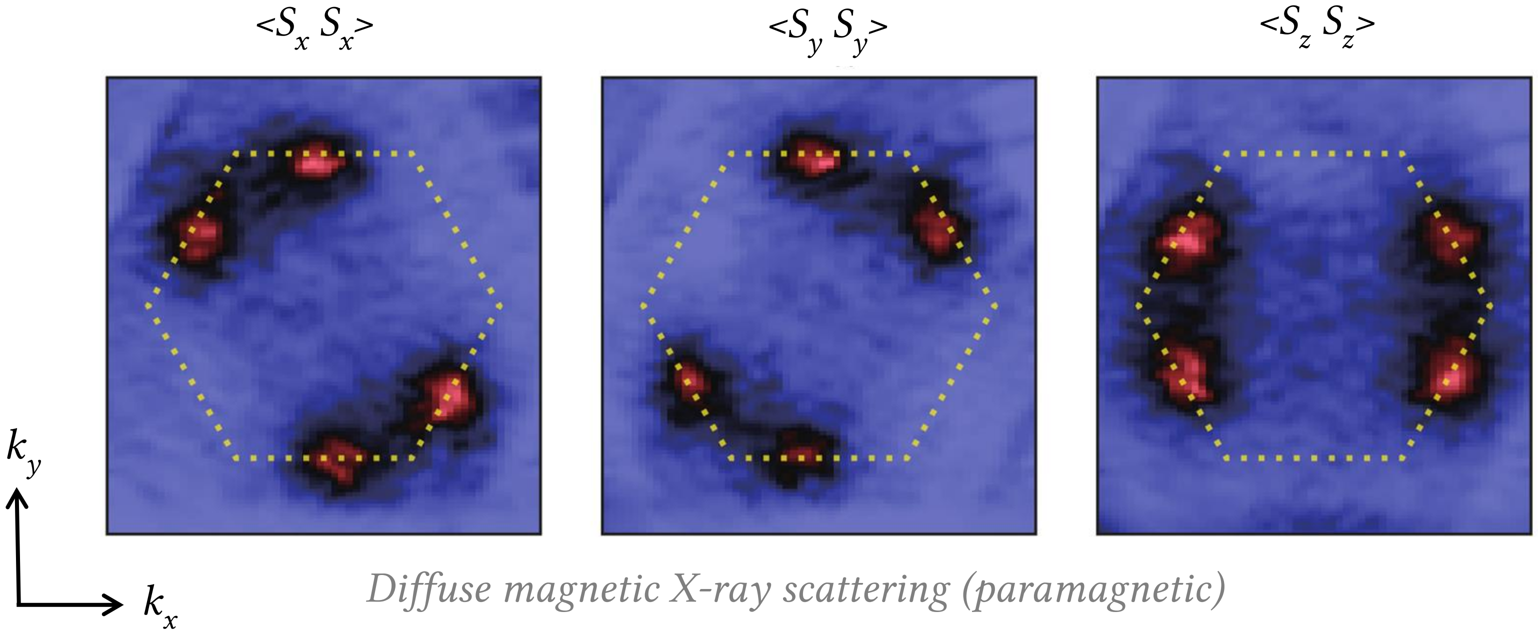
$$-\frac{8t^2 J_H}{3U} S_i^z S_j^z$$

Ferromagnetic Kitaev

Jackeli & Khaliullin, Phys. Rev. Lett. ***102****, 017205 (2009)*

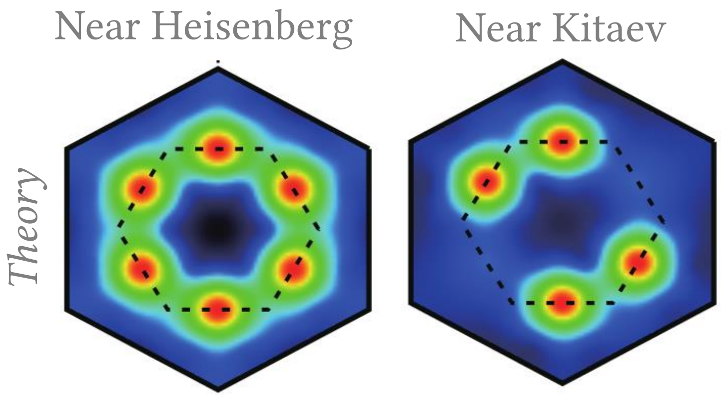
Evidence for Kitaev Exchange

Honeycomb iridate Na_2IrO_3



Diffuse magnetic X-ray scattering (paramagnetic)

- Implication of strong Kitaev interactions
- **Spin** and **spatial** orientation strongly correlated



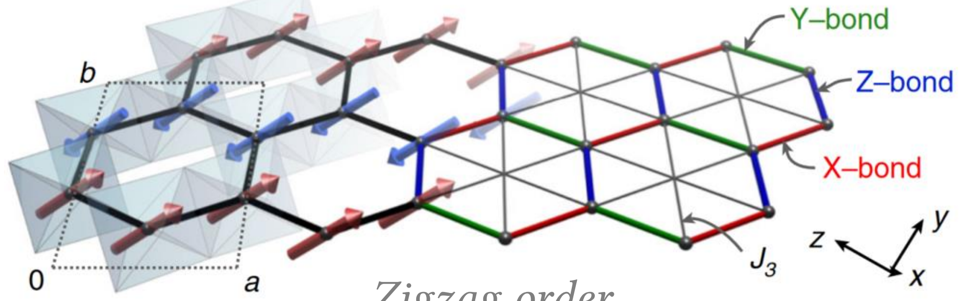
S.H. Chun et al, Nature Physics ***11****, 462 (2015)*

… unfortunately, nearly all *order*

- Most Kitaev materials *do not realize* the Kitaev spin liquid ground state
- **Magnetically order** at low temperatures
- Either *"zigzag"* or *"incommensurate spiral"* orderings have been seen

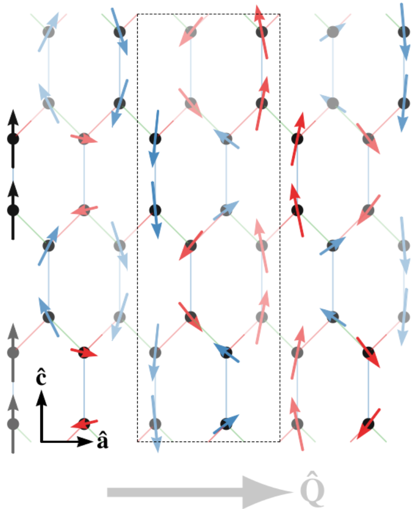
What is the cause?

$RuCl_3$, Na_2IrO_3



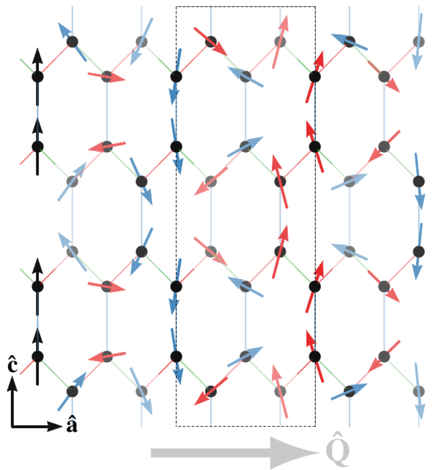
Zigzag order

β-Li_2IrO_3



Spiral order

γ-Li_2IrO_3



Spiral order

- More processes: *Direct* d_{xy}-d_{xy} overlap

***Two* processes**:

1. There and back via d_{xy} -d_{xy}
2. There via d_{xy} –d_{xy}, but *back* via oxygen-mediated route

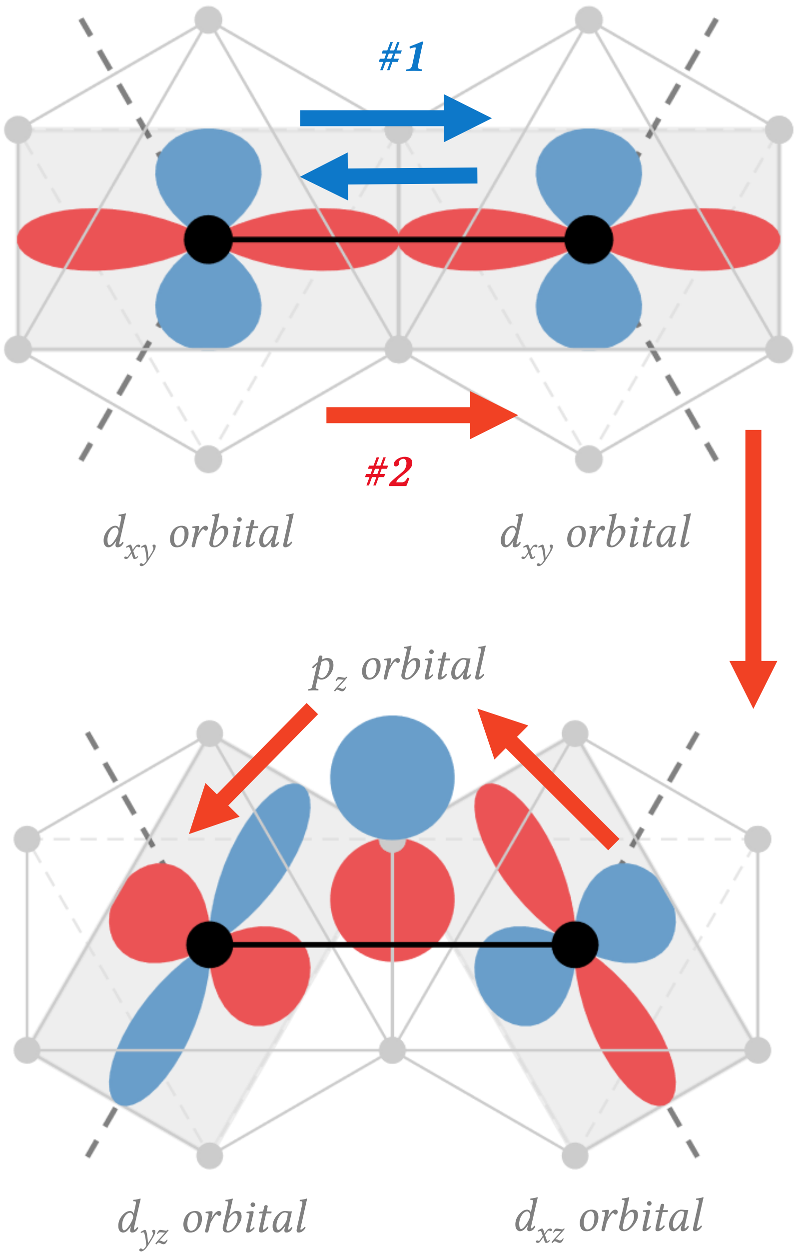
***Process #1*:**

Only Heisenberg exchange

***Process #2*:**

With Heisenberg, …

Symmetric off-diagonal exchange



Chaloupka, Jackeli & Khaliullin, Phys. Rev. Lett. ***105****, 027204 (2010); Rau, Lee & Kee, Phys. Rev. Lett.* ***112****, 077204 (2014)*

Generic model?

Generically expect Heisenberg **and** symmetric off-diagonal exchange when beyond the Kitaev limit

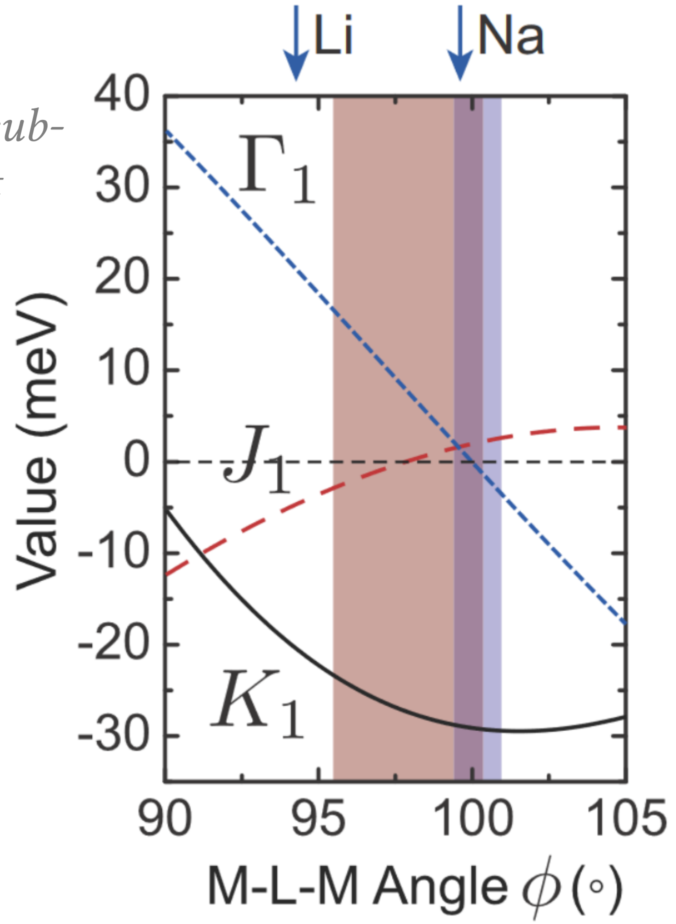
- Microscopic calculations suggest that in many Kitaev materials:

$\Gamma > 0$ and *sub-dominant*

*Not as large as Kitaev exchange, leading **non-Kitaev** interaction*

Sign expected from super-exchange process

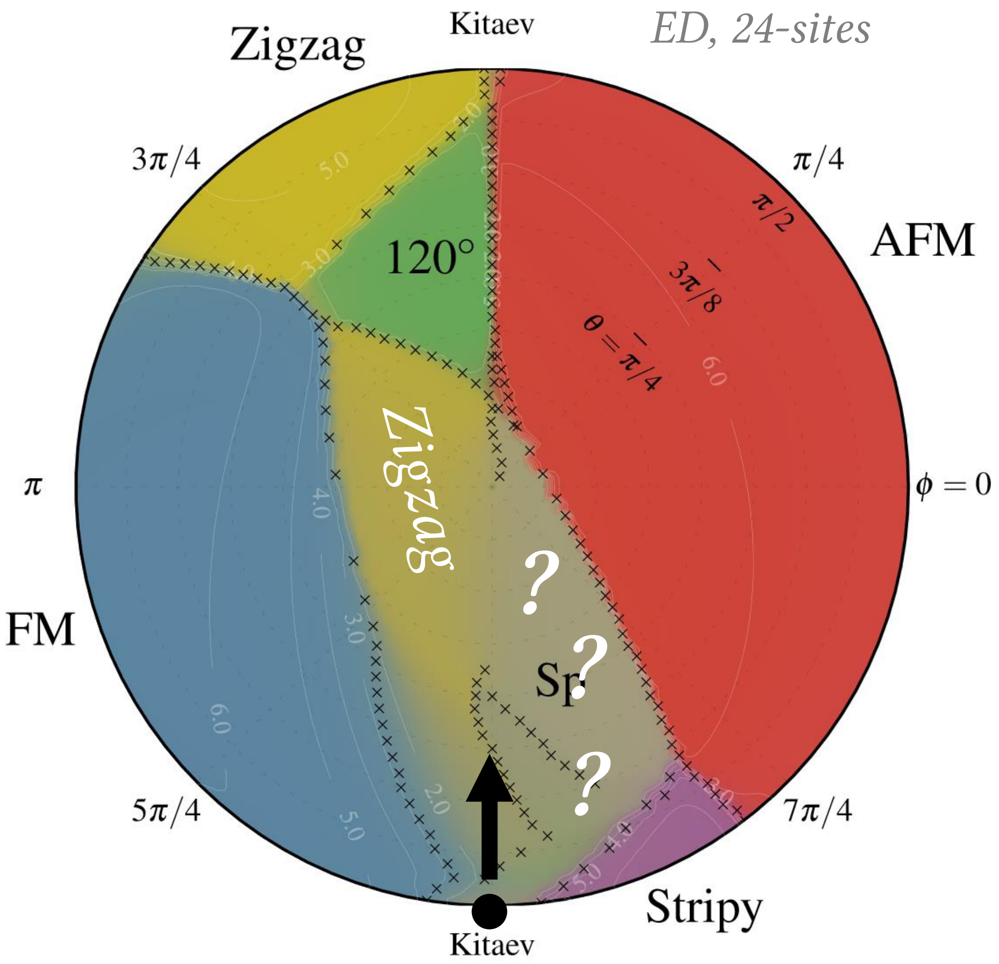
Leading sub-dominant term



*Winter et al., Phys. Rev. B **93**, 214431 (2016)*

Effect on Kitaev spin liquid

- Kitaev spin liquid occupies relatively *small* region
- Small positive Γ pushes it into **zigzag** phase *or* into *poorly-characterized region*
    - **Spiral? Spin liquid? Nematic?**

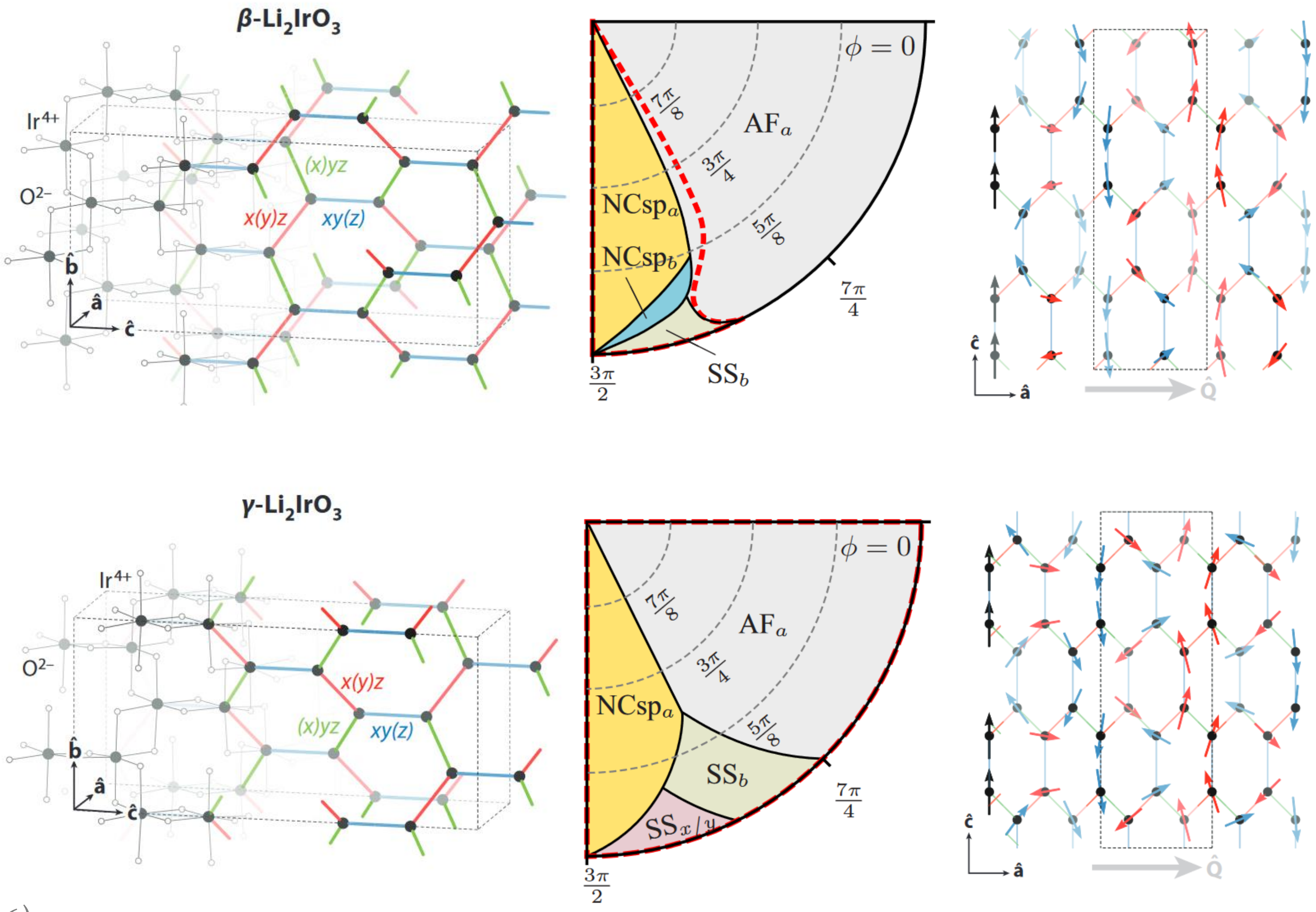


Contains experimentally observed *ordered* phases:

1. Zigzag ($RuCl_3$, Na_2IrO_3)
2. Incommensurate (Li_2IrO_3)

Incommensurate phases in 3D iridates

- *Mostly* successful in explaining ordering pattern in hyper- and harmonic-honeycombs
- Complex *counter rotating incommensurate spirals*
- **Appear near FM Kitaev limit with positive Γ**



*Lee, **Rau**, Kim, Physical Review B **93**, 184420 (2016)*

Importance for $RuCl_3$?

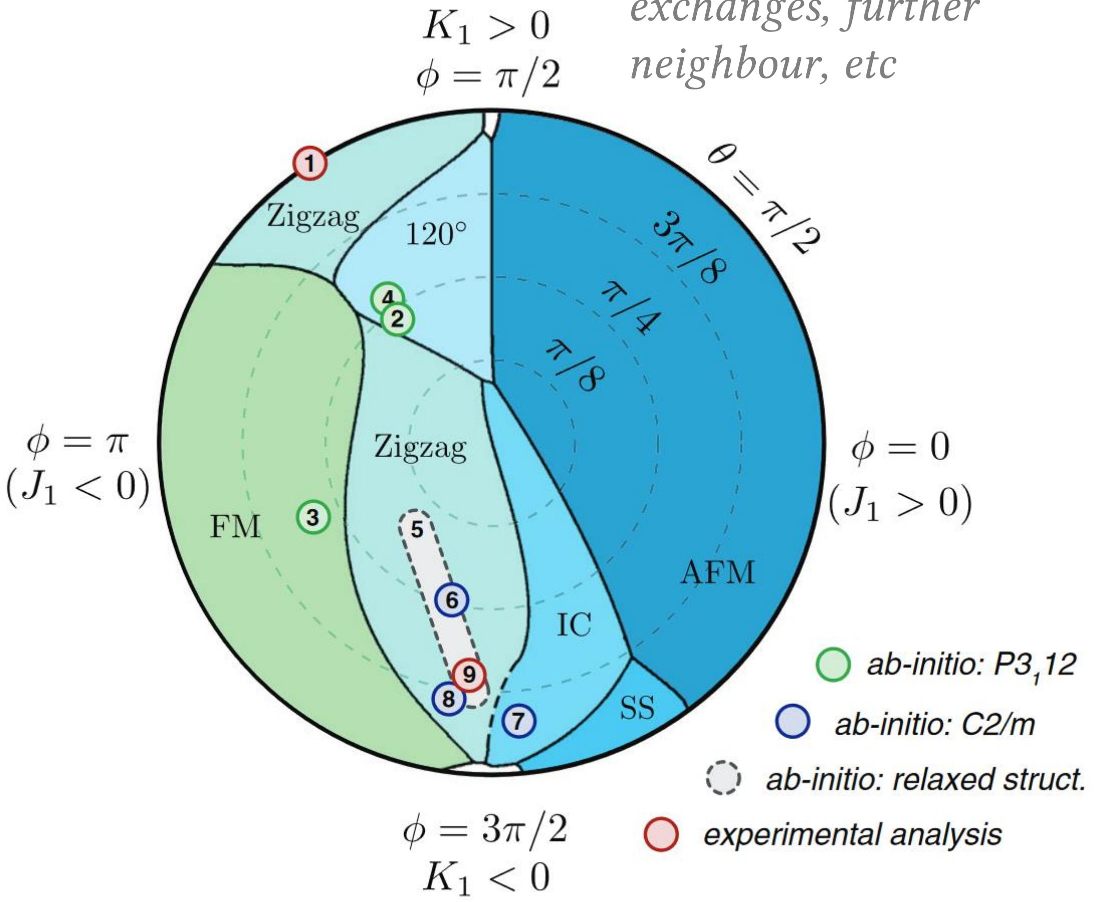
- Ab-initio find large, positive Γ
- Sometimes even *comparable* to the Kitaev exchange

Kitaev

Method	Structure	J_1	K_1	Γ_1
Exp. An. [166]	—	−4.6	+7.0	—
Pert. Theo. [149]	$P3_112$	−3.5	+4.6	+6.4
QC (2-site) [41]	$P3_112$	−1.2	-0.5	+1.0
ED (6-site) [45]	$P3_112$	−5.5	+7.6	+8.4
Pert. Theo. [149]	Relaxed	−2.8/ − 0.7	−9.1/ − 3.0	+3.7/+ 7.3
ED (6-site) [45]	$C2/m$	−1.7	−6.7	+6.6
QC (2-site) [41]	$C2/m$	+0.7	−5.1	+1.2
DFT [180]	$C2/m$	−1.8	−10.6	+3.8
Exp. An. [181]	—	−0.5	−5.0	+2.5

Symm. Off-Diag.

Includes more (realistic) exchanges, further neighbour, etc



Winter et al., *J. Phys. Cond. Mat.* **29**, 493002 (2017)

Maximov & Chernyshev, Phys. Rev. Research 2, 033011 (2020)

Reference	Method	K	Γ	Γ'	J	J_3	$\Gamma+2\Gamma'$	$J+3J_3$
Banerjee *et al.* [22]	LSWT, INS fit	+7.0			−4.6			−4.6
Kim *et al.* [29]	DFT+t/U, $P3$	**−6.55**	**5.25**	−0.95	−1.53		3.35	−1.53
	DFT+SOC+t/U	**−8.21**	**4.16**	−0.93	−0.97		2.3	−0.97
	Same+fixed lattice	−3.55	7.08	−0.54	**−2.76**		6.01	−2.76
	Same+U + zigzag	+4.6	6.42	−0.04	**−3.5**		6.34	−3.5
Winter *et al.* [30]	DFT+ED, $C2$	**−6.67**	6.6	−0.87	−1.67	**2.8**	4.87	6.73
	Same, $P3$	+7.6	8.4	+0.2	−5.5	**2.3**	**8.8**	+1.4
Yadav *et al.* [24]	Quantum chemistry	**−5.6**	−0.87		+1.2		−0.87	+1.2
Ran *et al.* [34]	LSWT, INS fit	**−6.8**	9.5				**9.5**	
	DFT+t/U, $U=2.5$ eV	−14.43	6.43		**−2.23**	**2.07**	6.43	+3.97
Hou *et al.* [31]	Same, $U=3.0$ eV	−12.23	**4.83**		**−1.93**	1.6	4.83	+2.87
	Same, $U=3.5$ eV	**−10.67**	**3.8**		−1.73	1.27	3.8	+2.07
Wang *et al.* [32]	DFT+t/U, $P3$	**−10.9**	6.1		−0.3	0.03	6.1	−0.21
	Same, $C2$	**−5.5**	7.6		+0.1	0.1	7.6	+0.4
Winter *et al.* [35]	*Ab initio* + INS fit	**−5.0**	2.5		−0.5	0.5	2.5	+1.0
Suzuki *et al.* [36]	ED, C_p fit	−24.41	**5.25**	−0.95	−1.53		3.35	−1.53
Cookmeyer *et al.* [37]	Thermal Hall fit	**−5.0**	2.5		−0.5	0.11	2.5	−0.16
Wu *et al.* [38]	LSWT, THz fit	−2.8	2.4		−0.35	0.34	2.4	+0.67
Ozel *et al.* [39]	Same, $K>0$	+1.15	2.92	+1.27	−0.95		5.45	−0.95
	Same, $K<0$	−3.5	2.35		+0.46		2.35	+0.46
Eichstaedt *et al.* [33]	DFT+Wannier+t/U	−14.3	9.8	−2.23	−1.4	0.97	5.33	+1.5
Sahasrabudhe *et al.* [42]	ED, Raman fit	**−10.0**	3.75		−0.75	0.75	3.75	1.5
Sears *et al.* [40]	Magnetization fit	**−10.0**	10.6	−0.9	**−2.7**		**8.8**	−2.7
Laurell *et al.* [41]	ED, C_p fit	−15.1	10.1	−0.12	−1.3	0.9	**9.86**	+1.4
This work	**“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]**
	Point 1	**−4.8**	**4.08**	**2.5**	**−2.56**	**2.42**	**9.08**	**4.7**
	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	6.5

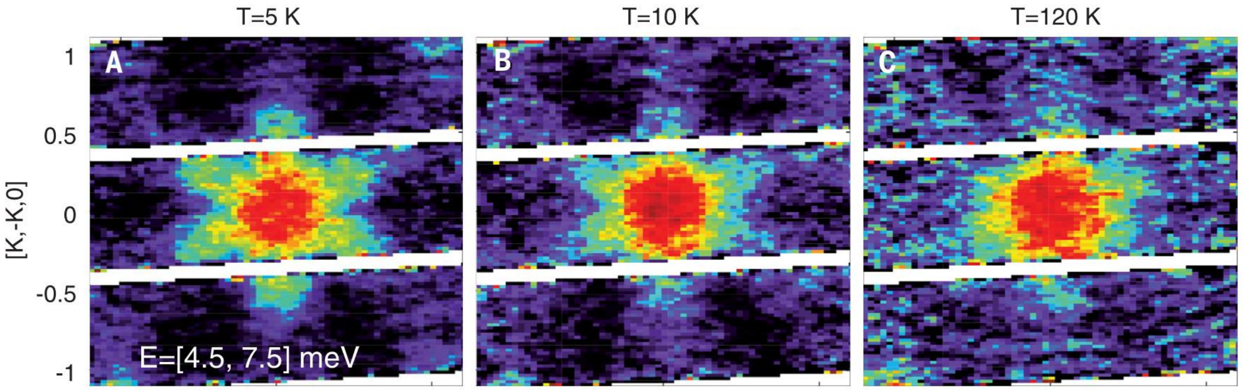
Full model more complicated

*Not **just** ab-initio methods*

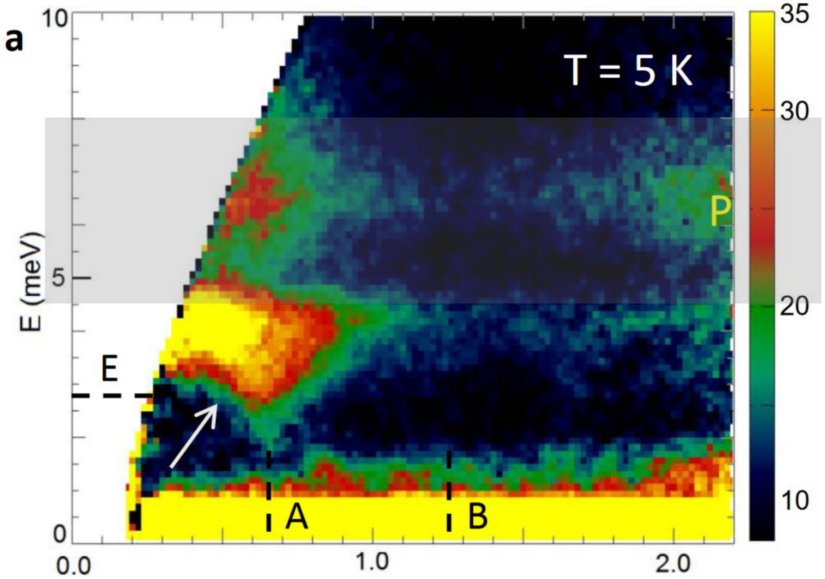
…

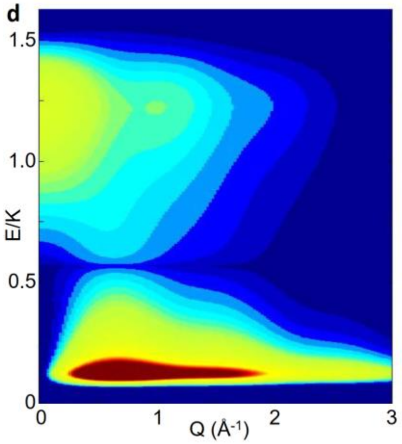
Remnants of the spin liquid?

- Some indications of Kitaev-like features in *high-energy excitations* in $RuCl_3$
- Indicating some **proximity** to the spin liquid phase?



High-energy feature





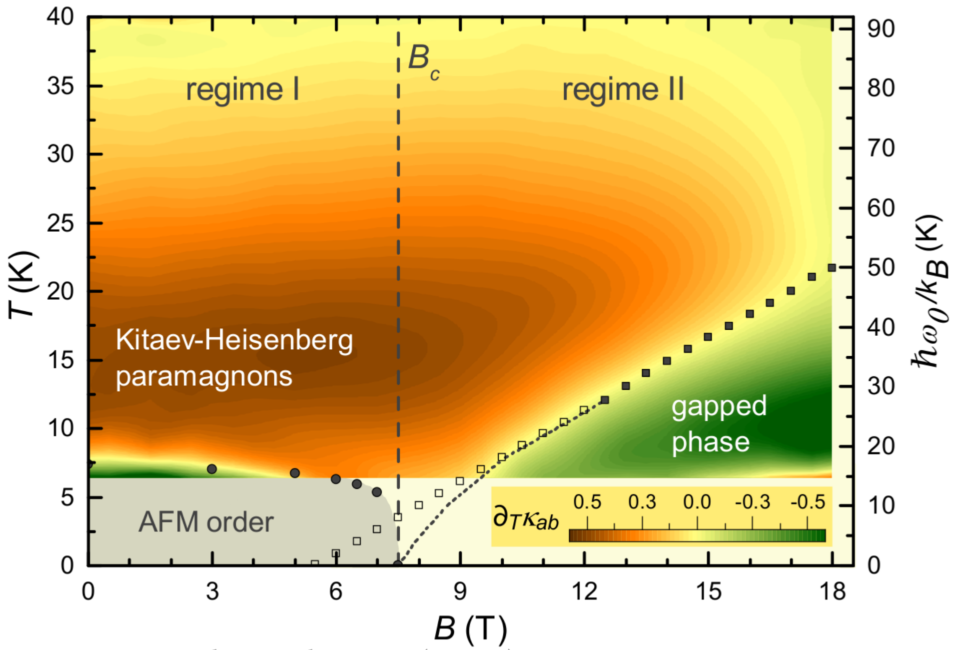
AF Kitaev model (theory)

Banerjee et al, Nat. Mat. ***15*** *733 (2015);*
Banerjee et al, Science ***356*** *1055 (2017)*

Towards Kitaev by applied field?

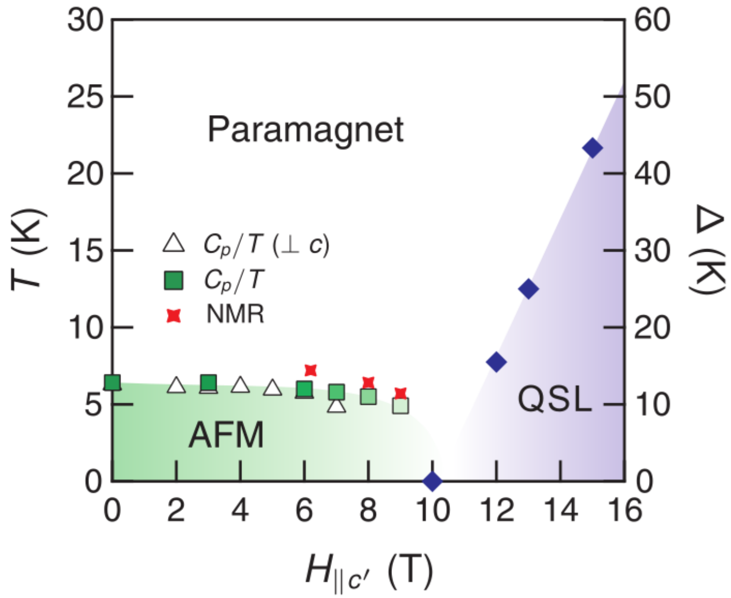
- Can suppress ordering *quickly* with **in-plane** applied magnetic field

Thermal conductivity, in-plane field



Hentrich et al, PRL (2018)

NMR & specific heat, tilted field

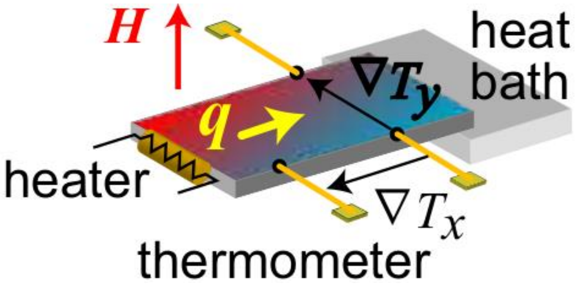


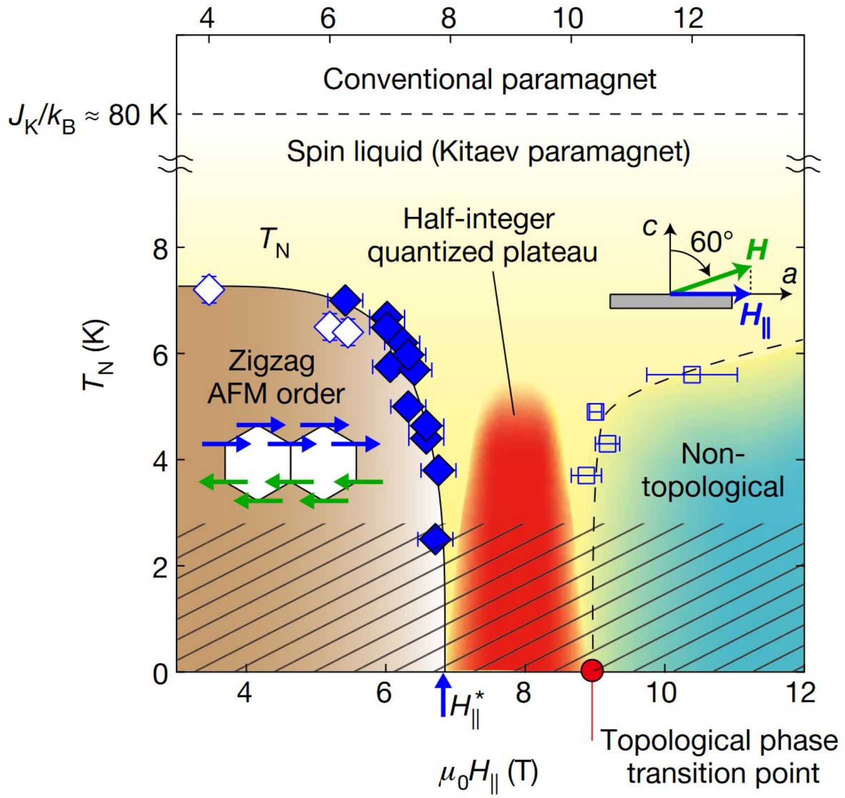
Baek et al, PRL (2017)

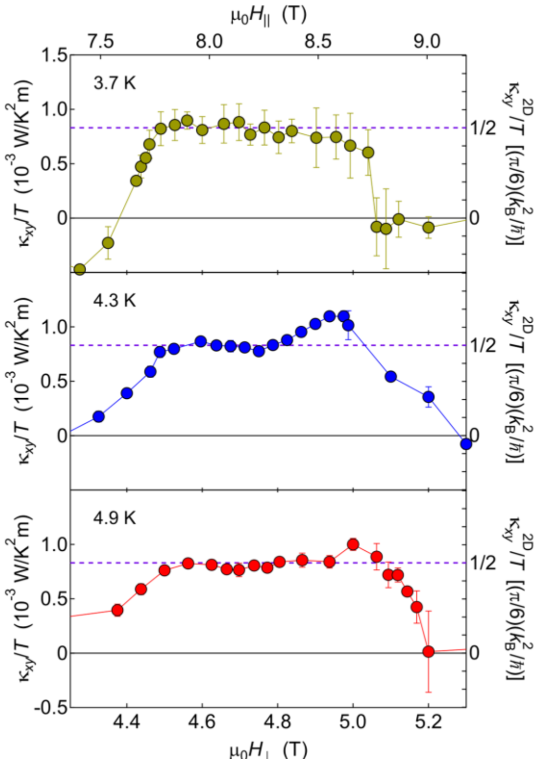
Some evidence for an **intermediate** phase once order dies off …

Chiral Majorana Edge Modes?

- **Thermal Hall** is *quantized* at low temperature in intermediate phase







$$\frac{\kappa_{xy}}{T} = q\frac{\pi}{6}\frac{k_B^2}{\hbar}$$

(Chiral) central charge of edge modes

- Quantized *half-integral* value: one (chiral) *Majorana* edge mode?

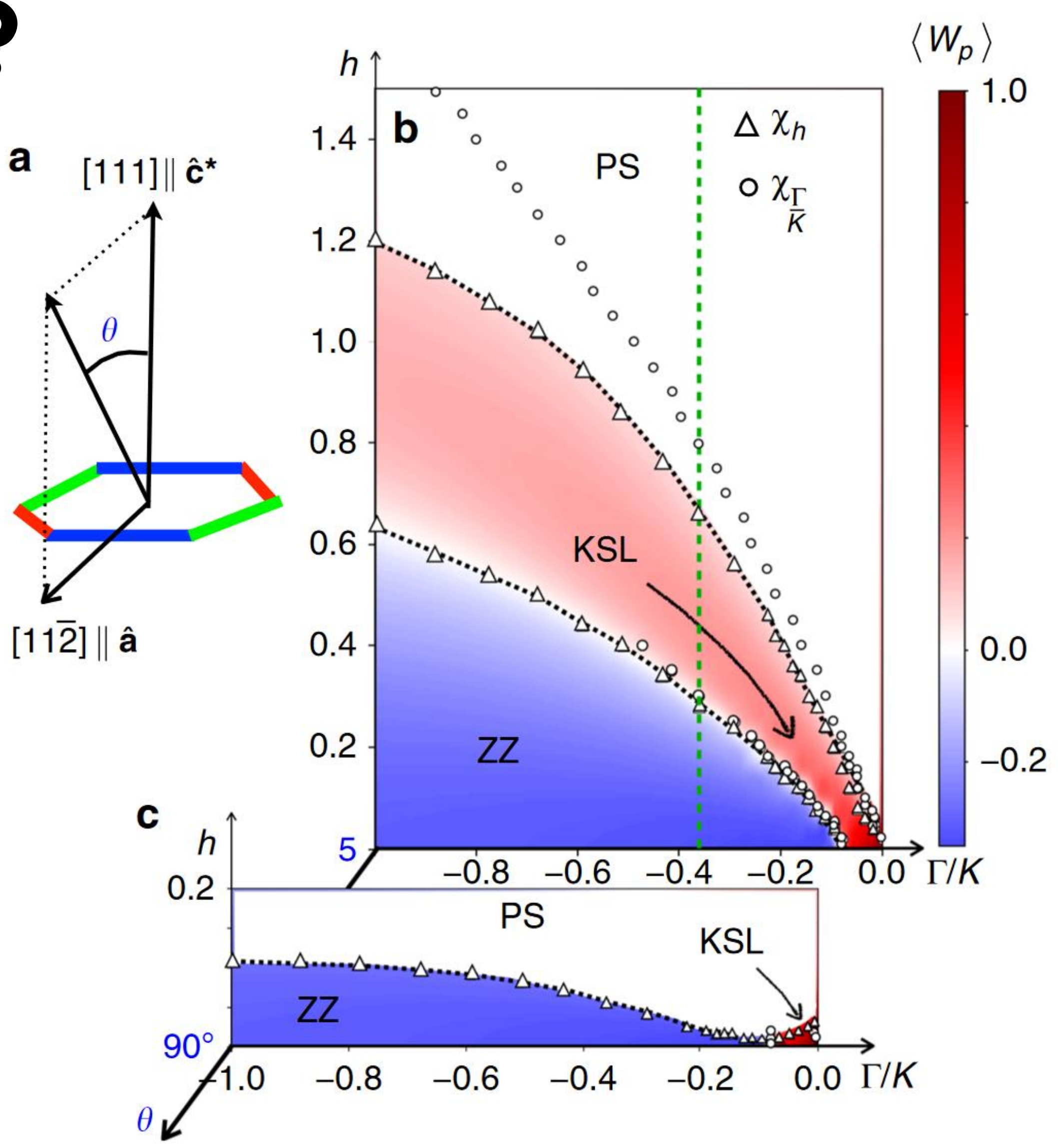
Kasahara et al, Nature (2018)

Subdominant exchanges?

- Perturb spin liquid with sub-dominant exchanges, **add field**
- Spin liquid can *re-emerge* at finite **titled** field

Proof of principle: Kitaev spin liquid **can** re-emerge in applied field

- *Other* explanations?



Gordon et al, Nat. Comm. **10** 2470 (2019)

Summary

Kitaev's honeycomb model (& generalizations):

- Exactly solvable models of **Z_2 quantum spin liquids**
- Explicit demonstration of *fractionalization* of spins into *Majorana fermions*
- Very rich thermodynamic & dynamical properties and under an applied magnetic field *Chiral Majorana edge modes*

Kitaev *Materials*:

- Growing family of materials with **Kitaev as dominant exchange** $RuCl_3$, Na_2IrO_3, Li_2IrO_3, …
- *Potential* to **realize** Kitaev's spin liquid in solid-state systems *Half-quantized thermal Hall effect in $RuCl_3$?*

Thank you for your attention