# Quantum Spin Liquids:
# Signatures of Fractionalization

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*Bar-Ilan, Zoom Seminar, July 2, 2020*

# Quantum Spin Liquids: Signatures of Fractionalization

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Physics Department
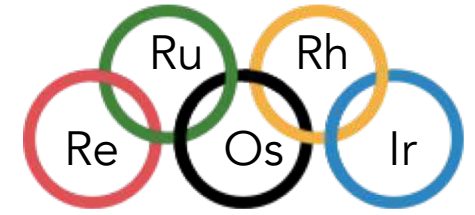The Ohio State University

trivedi.15@osu.edu
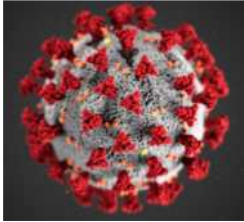
http://trivediresearch.org.ohio-state.edu/

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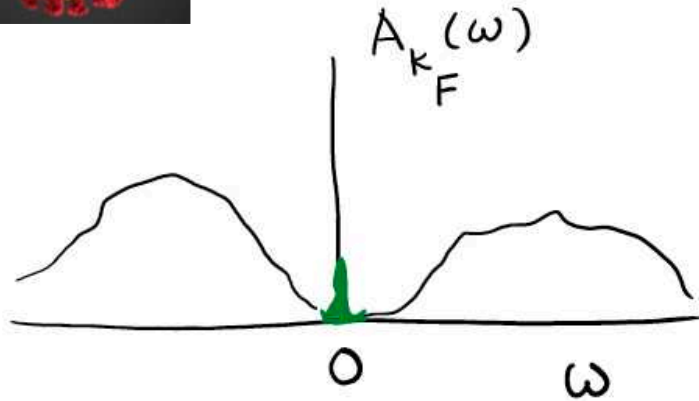
Center of Emergent Materials
NSF MRSEC – DMR

She will talk about "Quantum Spin Liquids: Signatures of Fractionalization Nandini".
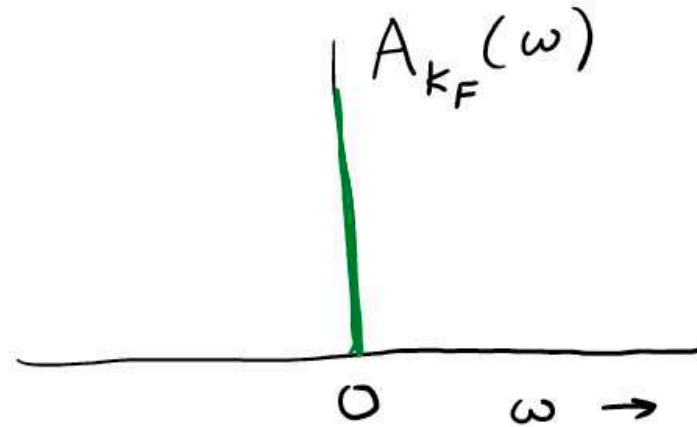
BC

$A_{k_F}(\omega)$

O

$\omega$

fractionalized

~ today

$A_{k_F}(\omega)$

O

$\omega \rightarrow$

non interacting
quasi particle

$$\int_{-\infty}^{\infty} A_k(\omega)\, d\omega = 1$$

# Roadmap

➢ Big picture

➢ 2D: Kitaev Model
❖ Discovery of a new gapless QSL with a spinon Fermi surface

❖ Spectrum of 1 spin flip and 2 spin flip excitations

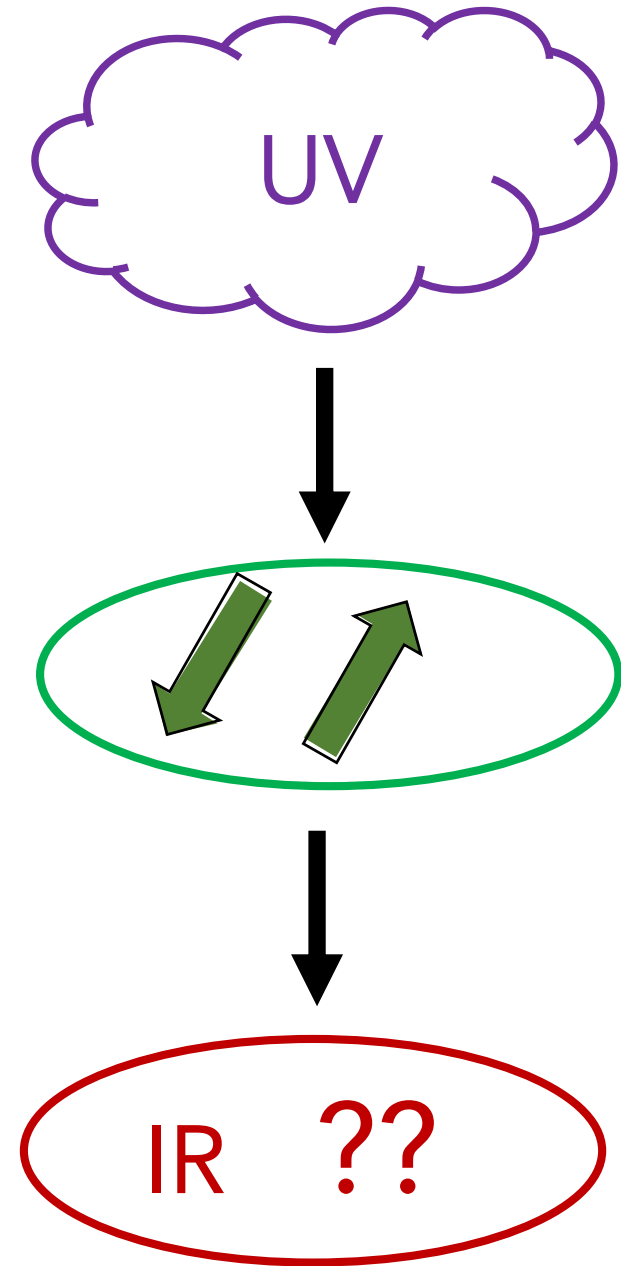➢ QSL Materials

➢ How do you detect a QSL?
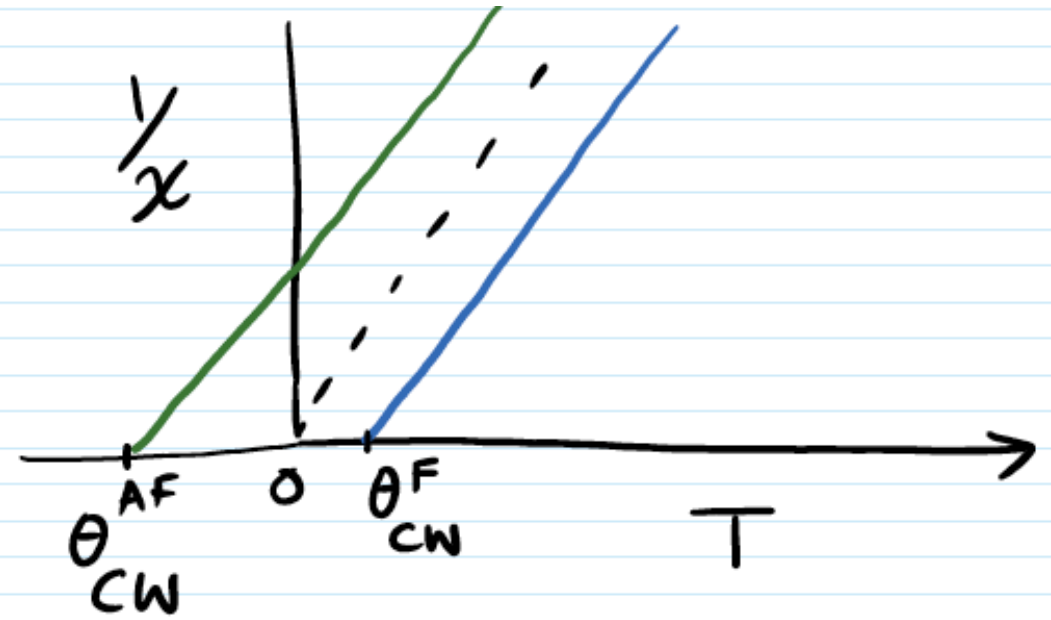
➢ Going forward….
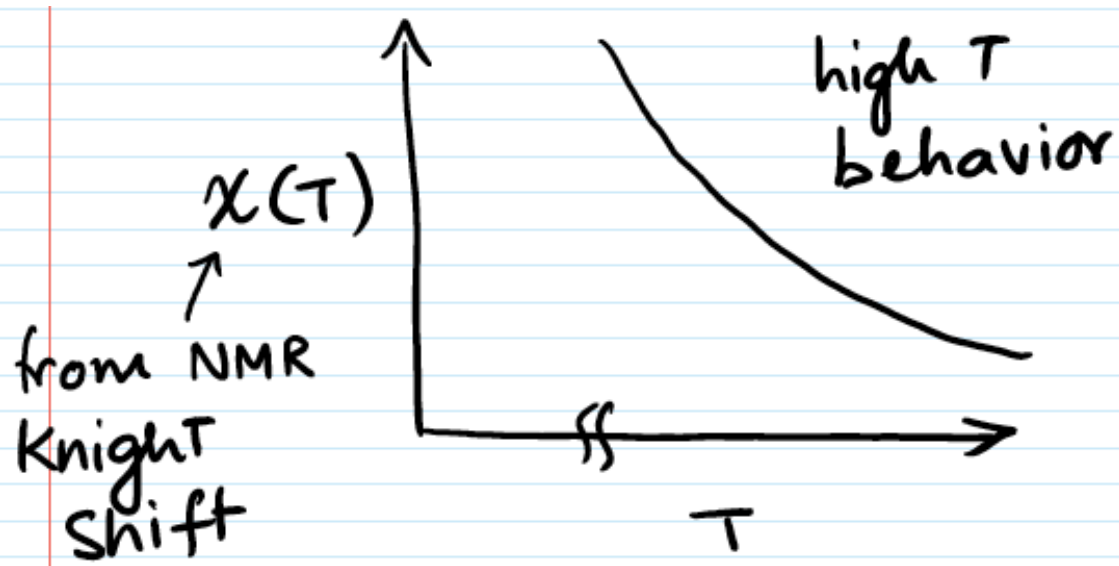
# Big Picture

What is a QSL?
Why is it interesting?  Important?

# First Signatures of a QSL
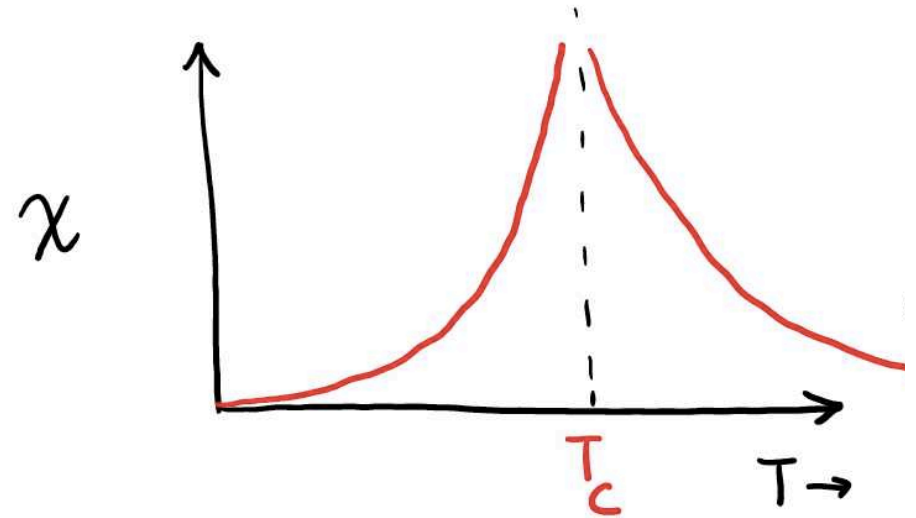
# *First* signatures of a QSL: large frustration parameter
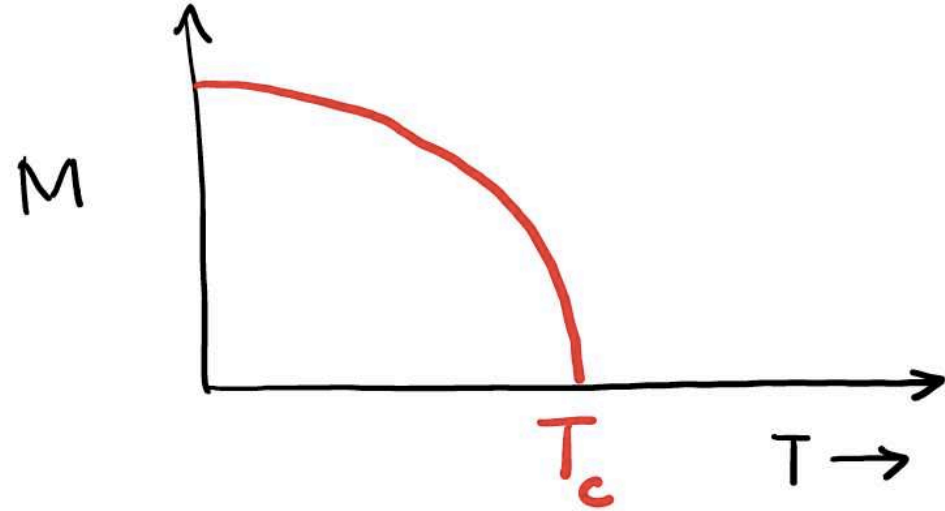


$$\chi(T) = \frac{C}{T - \theta_{cw}}$$

# Spontaneously broken time reversal symmetry

$$\mathcal{H} = J \sum_{\langle ij \rangle} S_i^z S_j^z \qquad S_i = \pm \frac{1}{2}$$
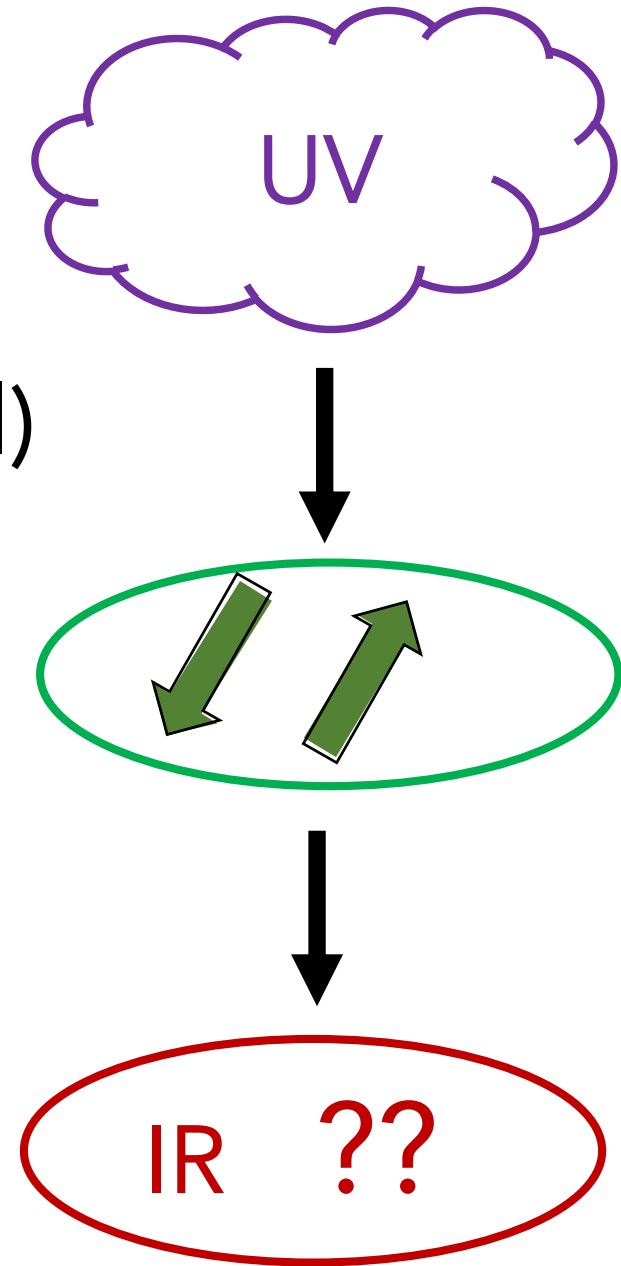
$$M = \langle S_i^z \rangle$$



$$f = \frac{|\theta_{cw}|}{T_c}$$

# First Signatures of a QSL

o **Mott insulator**
(odd number of electrons in unit cell)

o **Local moments**
(typically $S=1/2$ or $J_{eff}=1/2$)

o **Strongly interacting moments**
$\theta_{CW} \sim 100$ K

o **No magnetic ordering**
$f = \theta / Tc \sim 10^4$

UV

IR ??

# First Signatures of a QSL

*So why is a paramagnet interesting?*

○ Mott insulator
      (odd number of electrons in unit cell)
○ Local moments
      (typically $S=1/2$ or $J_{eff}=1/2$)
○ Strongly interacting moments
      $\theta \sim 100$ K
○ No magnetic ordering
      $f = \theta / Tc \sim 10^4$

# Quantum Matter

❑ Landau paradigm: spontaneously broken symmetry→
local order parameter m
bosonic excitations: magnons (for continuous spins)

❑ Topological Paradigm

"IQHE"
Topological Insulators
Topological Superconductors
Topological Weyl and Dirac Semimetals
Topological magnons

"FQHE"
Quantum Spin Liquids
Possess Topological Order

- Ground state degeneracy
- Long range entanglement
- Fractionalized Excitations

Review: Savary and Balents, Repts. on
Progress in Physics  80, 016502 (2017)

Wen, X.-G. (1989) PRB 40, 7387
Wen, X.-G. and Niu, Q. (1990) PRB 41, 9377

# Quantum Matter

❑ Landau paradigm: spontaneously broken symmetry→
        local order parameter m
        bosonic excitations: magnons (for continuous spins)

❑ Topological Paradigm

"IQHE"

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Topological Superconductors
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"FQHE"
Quantum Spin Liquids
Possess Topological Order

- Ground state degeneracy
- Long range entanglement
- Fractionalized Excitations

Important for storing information non-locally;
robust against decoherence
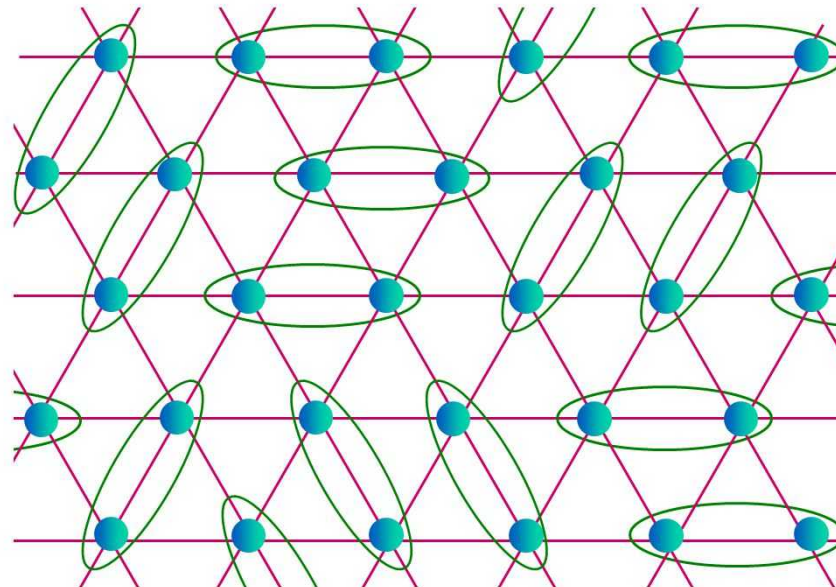
Singlet
or valence bond

$$= \frac{1}{\sqrt{2}} \left( |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right)$$

valence
bond solid

Picture of a QSL

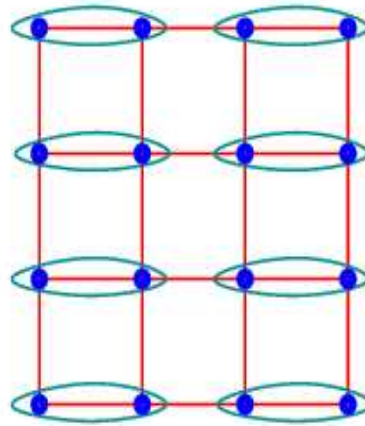Resonating valence bond
-- candidate quantum spin liquid

Anderson 1973
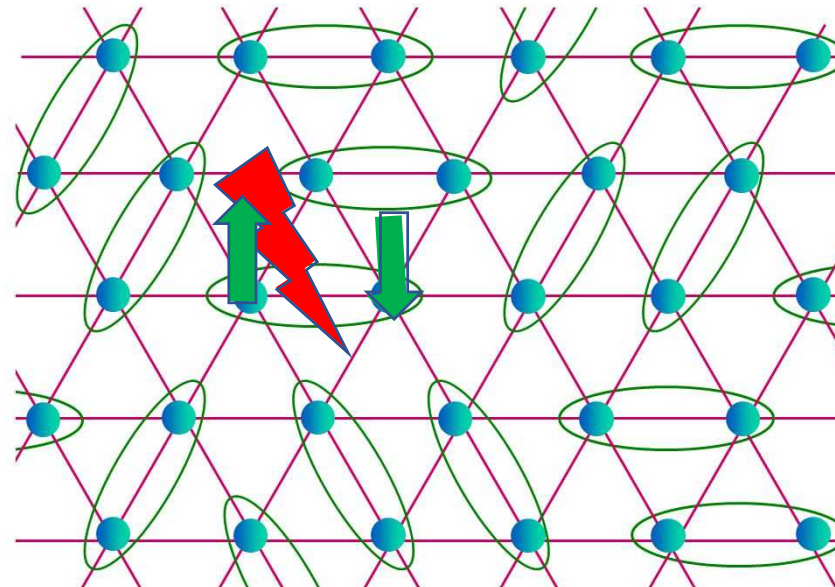
## Excitations of a QSL

Singlet
or valence bond

$$= \frac{1}{\sqrt{2}} \left( |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right)$$

Contrast with
ordered magnet:
Magnons: S=1 (bosons)

valence
bond solid

Resonating valence bond
-- candidate quantum spin liquid

Anderson 1973

deconfined
S=1/2
spinons

Fermionic
excitations

# Why would a bunch of interacting spins not order at T=0 ?

(1)  Low spin

(2)  Low dimensionality

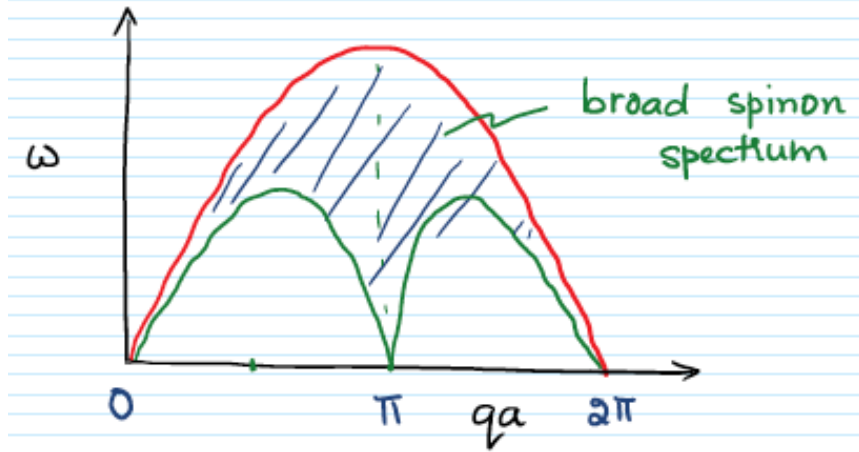(3)  Frustration
[Geometric, Interactions, …]

Quantum Fluctuations

# Fractionalization of excitations in quantum spin liquids
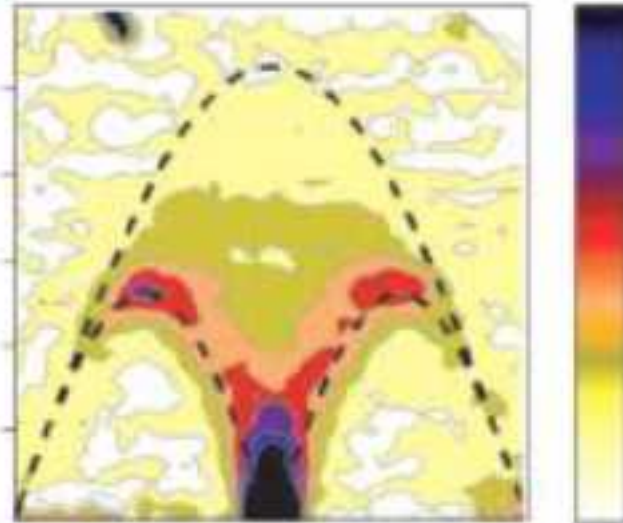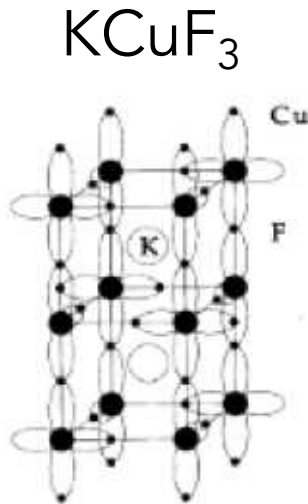
# 1d Quantum Spin Liquid

Fractionalized S=1 magnons (bosons) into two S=1/2 neutral spinons (fermions)

$|RVB\rangle = \{|\cdots\rangle$

$+ |\cdots\rangle$

$+ \cdots \}$

linear superposition of all possible singlet coverings $\rightarrow$ Spin liquid

broad spinon spectrum

$\omega$

$0 \qquad \pi \quad qa \quad 2\pi$

Inelastic neutron scattering S(q,w)

$KCuF_3$



Lake et al Nat. Mat. 4, 329 (2005)

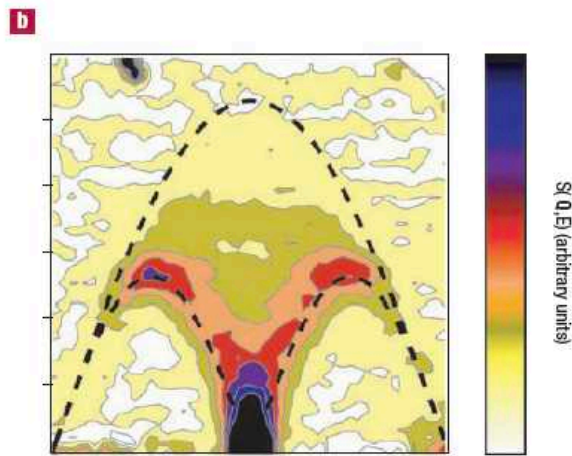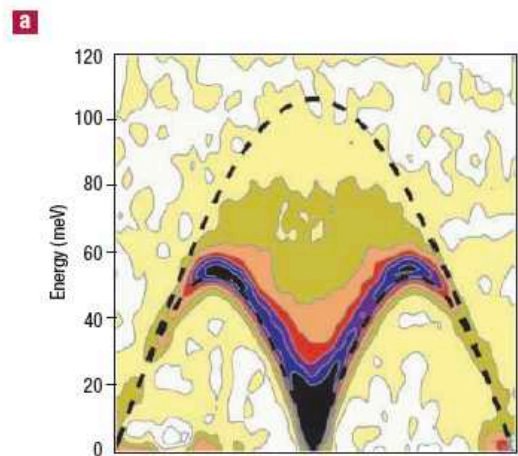$$\omega_\ell(q) = \pi \frac{J}{2} |\sin qa|$$

$$\omega_u(q) = \pi J \left| \sin \frac{qa}{2} \right|$$

compare with $\omega_{c\ell}(q) = 2J |\sin qa|$
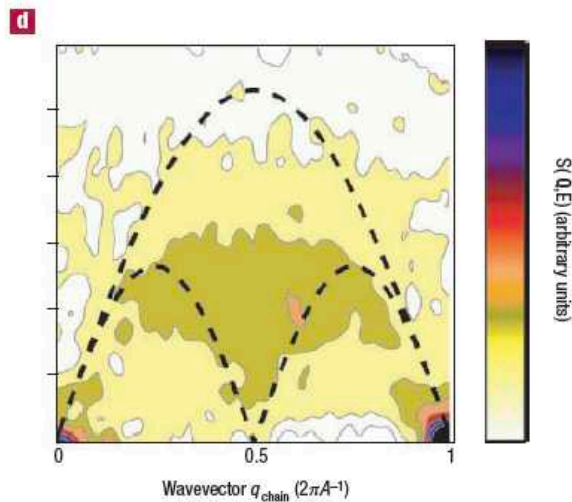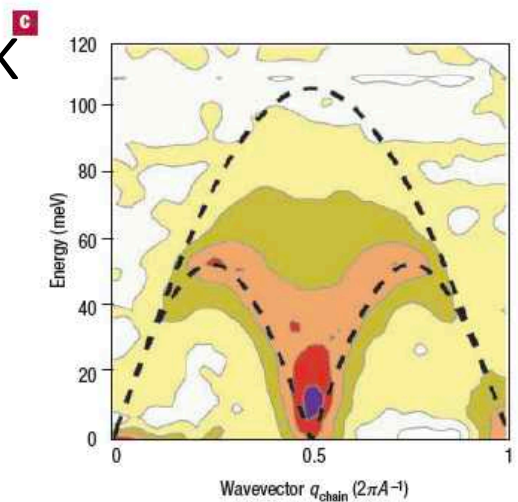
Broad spectrum indicates fractionalization of magnons

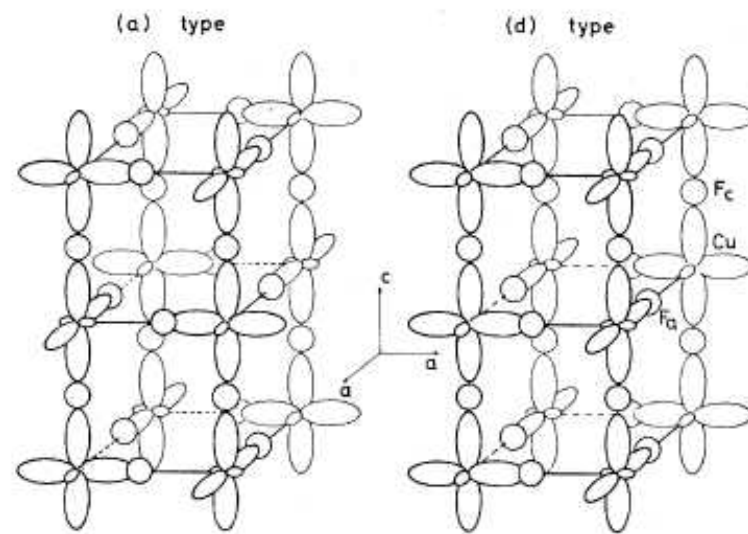Ordering~ 10K

T=6K

T=50 K

T=150 K

T=300 K



FIG. 1. Schematics of type (a) and (d) structures observed in KCuF$_3$.

$J \sim 340\ K$

$J_\perp = -16\ K$

Black dotted: multi-spinon continuum predicted at T=0 (Muller ansatz equation)

# Roadmap

➤ Big picture

➤ 2D: Kitaev Model
❖    Discovery of a new gapless QSL with a spinon Fermi surface

❖    Spectrum of 1 spin flip and 2 spin flip excitations

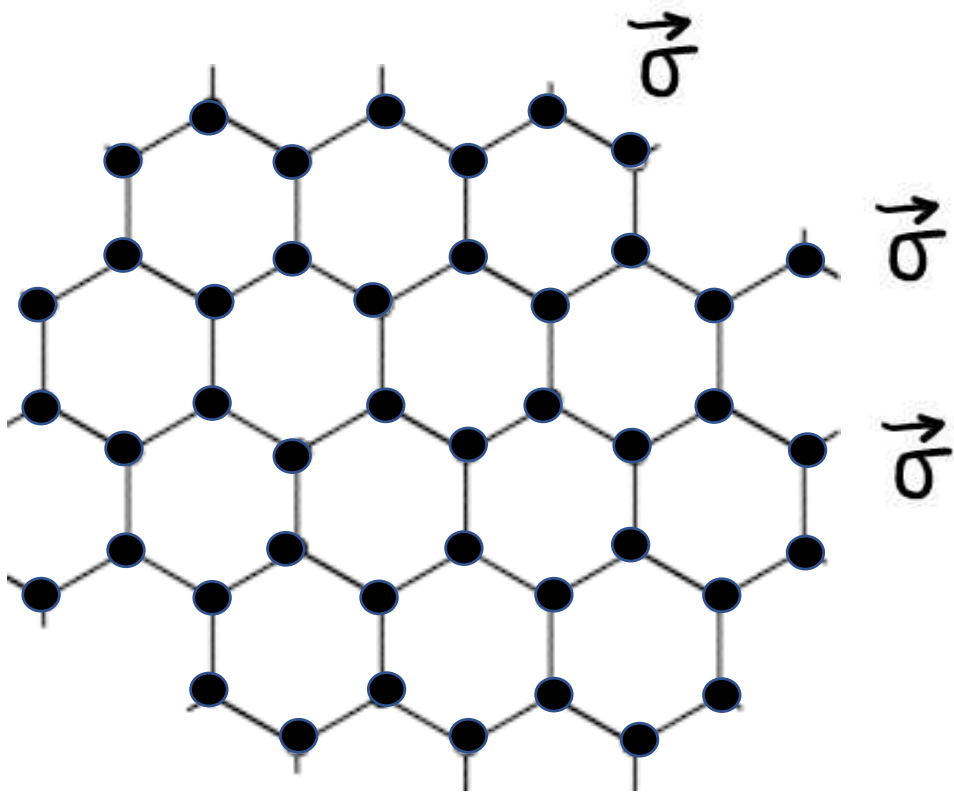➤ QSL Materials

➤ How do you detect a QSL?

➤ Going forward….

# Kitaev Model

$\vec{\sigma}$
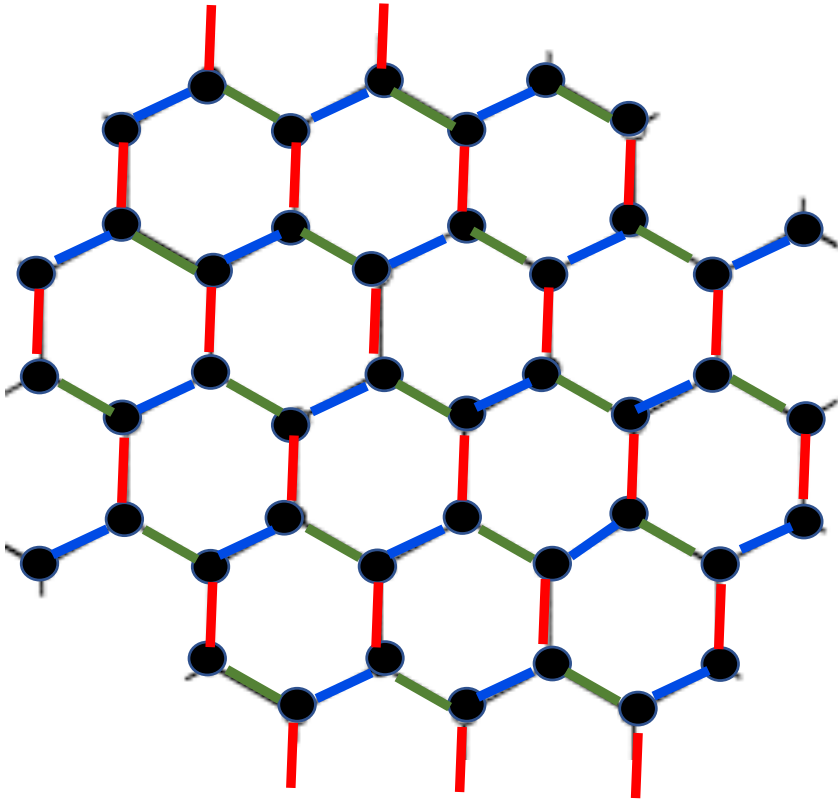
$\vec{\sigma}$

$\vec{\sigma}$
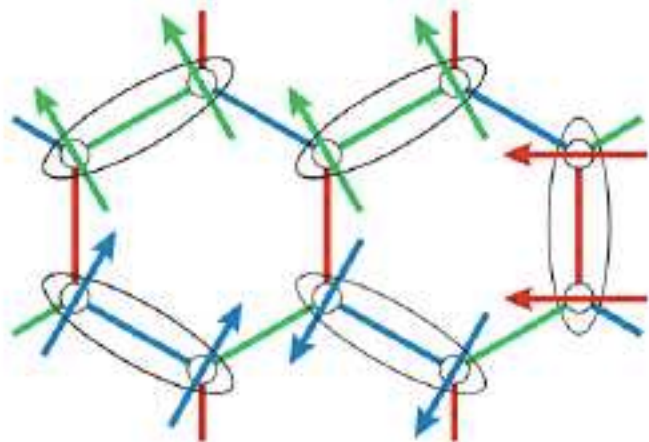
put qubit on each site

Honey comb lattice
Bipartite lattice; no geometric frustration

# Kitaev Model: bond-dependent interactions
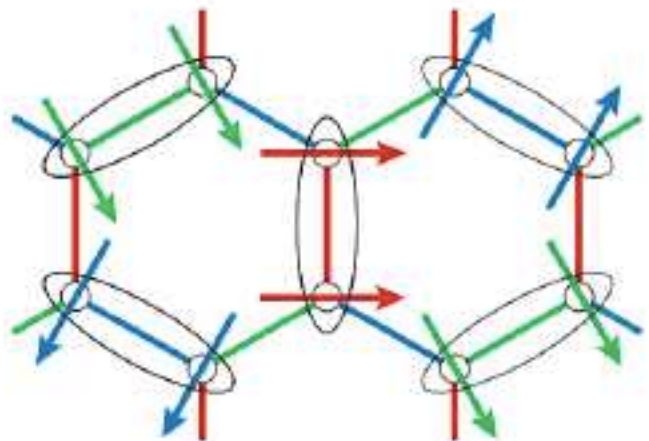


$$\mathcal{H} = K\left[\sum_{\langle ij\rangle \in x} \sigma_i^x \sigma_j^x + \sum_{\langle ij\rangle \in y} \sigma_i^y \sigma_j^y + \sum_{\langle ij\rangle \in z} \sigma_i^z \sigma_j^z\right]$$
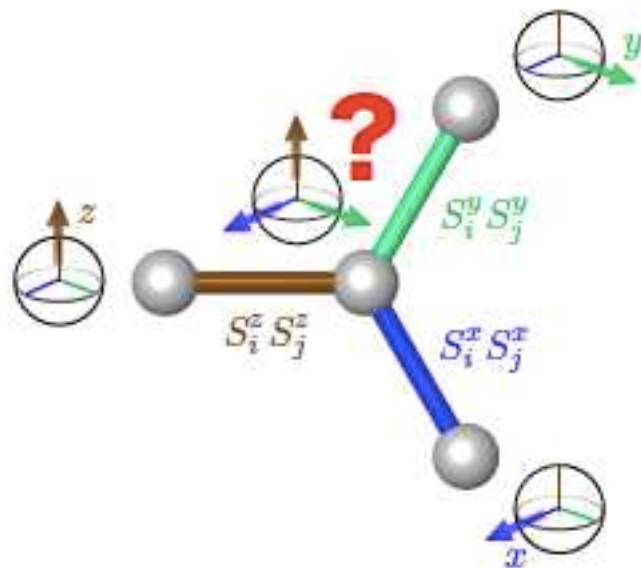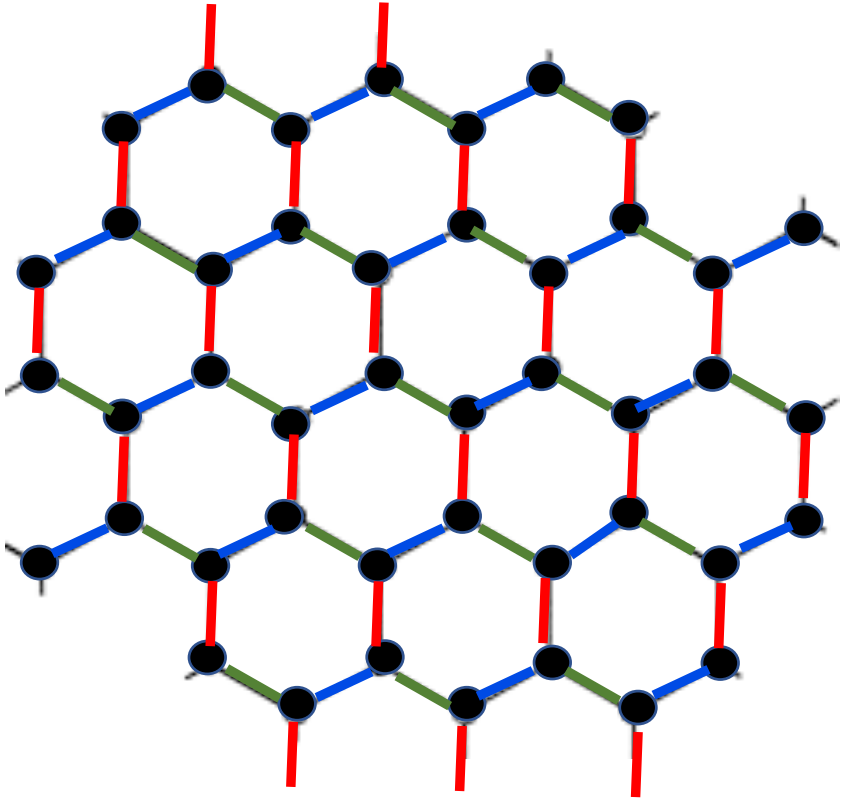
# Kitaev Model: bond-dependent interactions



$$\mathcal{H} = K\left[\sum_{\langle ij\rangle \in x} \sigma_i^x \sigma_j^x + \sum_{\langle ij\rangle \in y} \sigma_i^y \sigma_j^y + \sum_{\langle ij\rangle \in z} \sigma_i^z \sigma_j^z\right]$$

$S_i^y S_j^y$

$S_i^z S_j^z$

$S_i^x S_j^x$

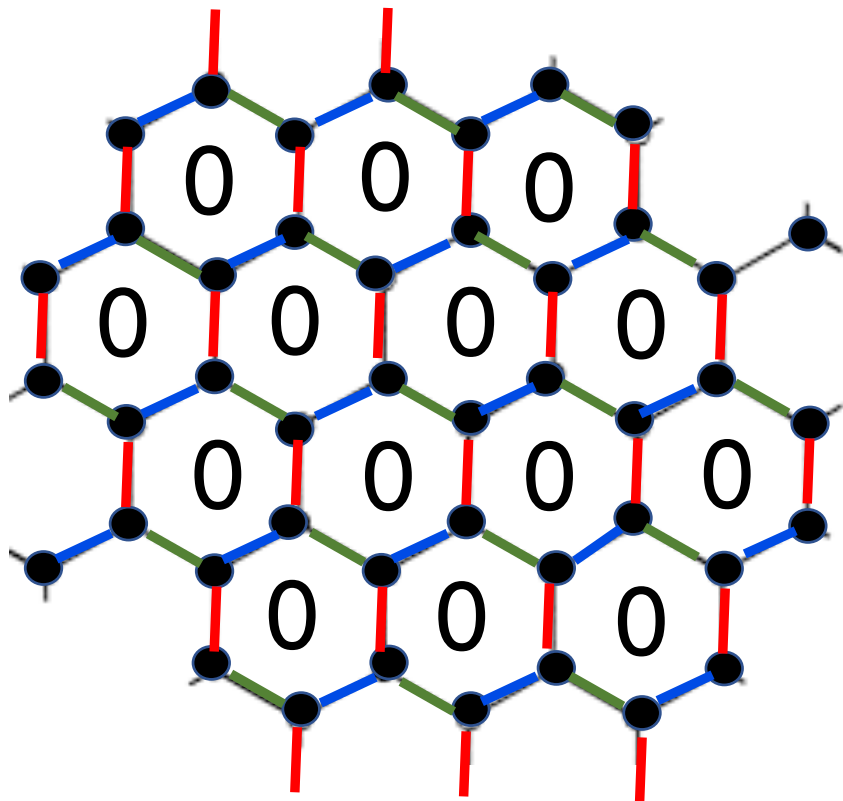# Kitaev Model: bond-dependent interactions



$$\mathcal{H} = K \left[ \sum_{\langle ij \rangle \in x} \sigma_i^x \sigma_j^x + \sum_{\langle ij \rangle \in y} \sigma_i^y \sigma_j^y + \sum_{\langle ij \rangle \in z} \sigma_i^z \sigma_j^z \right]$$
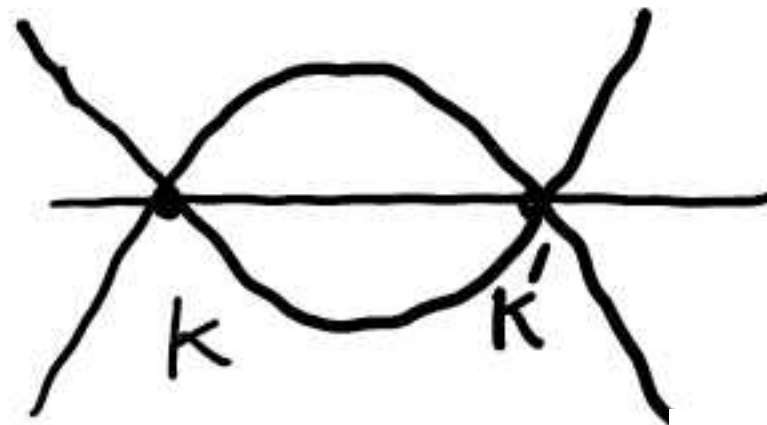
Parton construction:

$$\sigma^\alpha = i \, b^\alpha c$$

$$\mathcal{H} = K \frac{i}{2} \sum_{\langle ij \rangle} \hat{u}_{ij} \, c_i c_j$$

# Ground State:



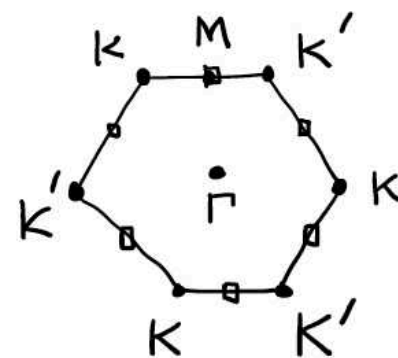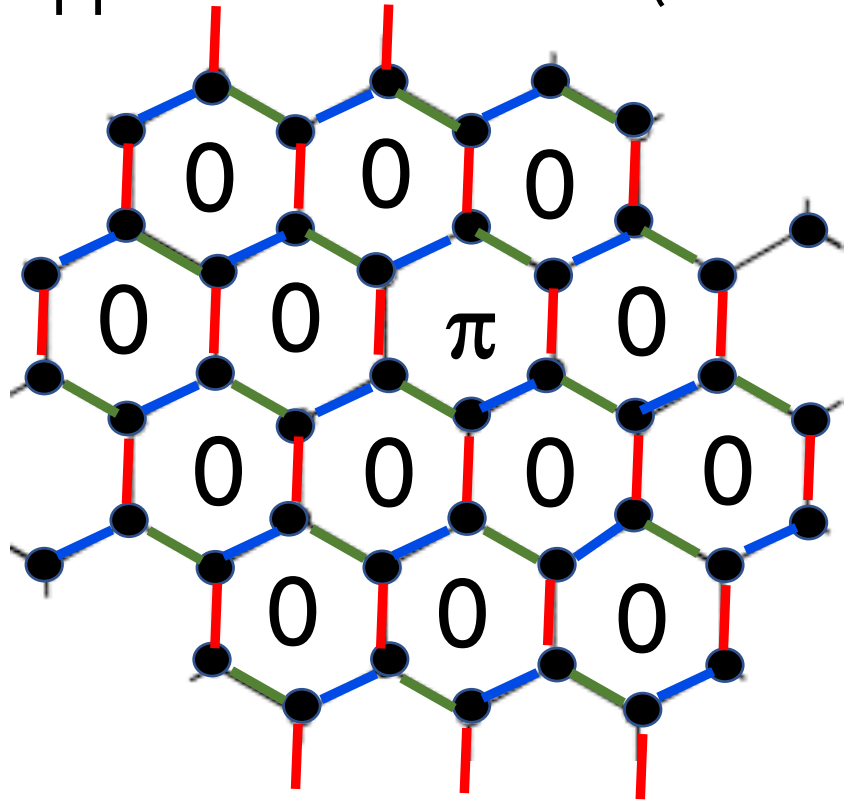All plaquettes have zero flux



c-majorana
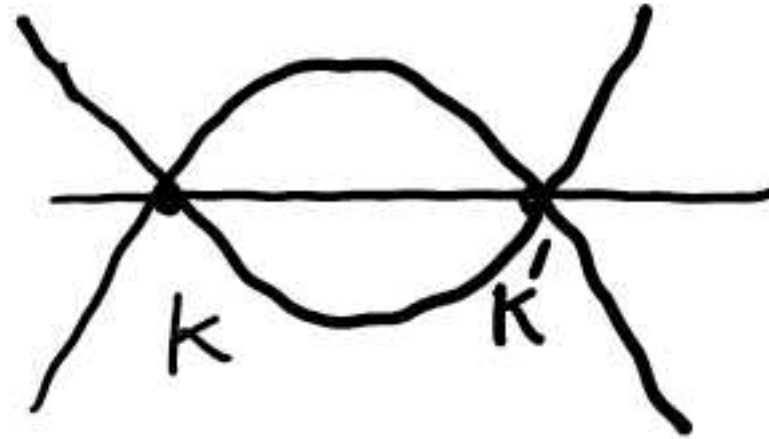fermions have a
Dirac dispersion

# Excitations:

(1)Gapped flux excitation (visons)



(2) Gapless majorana fermions

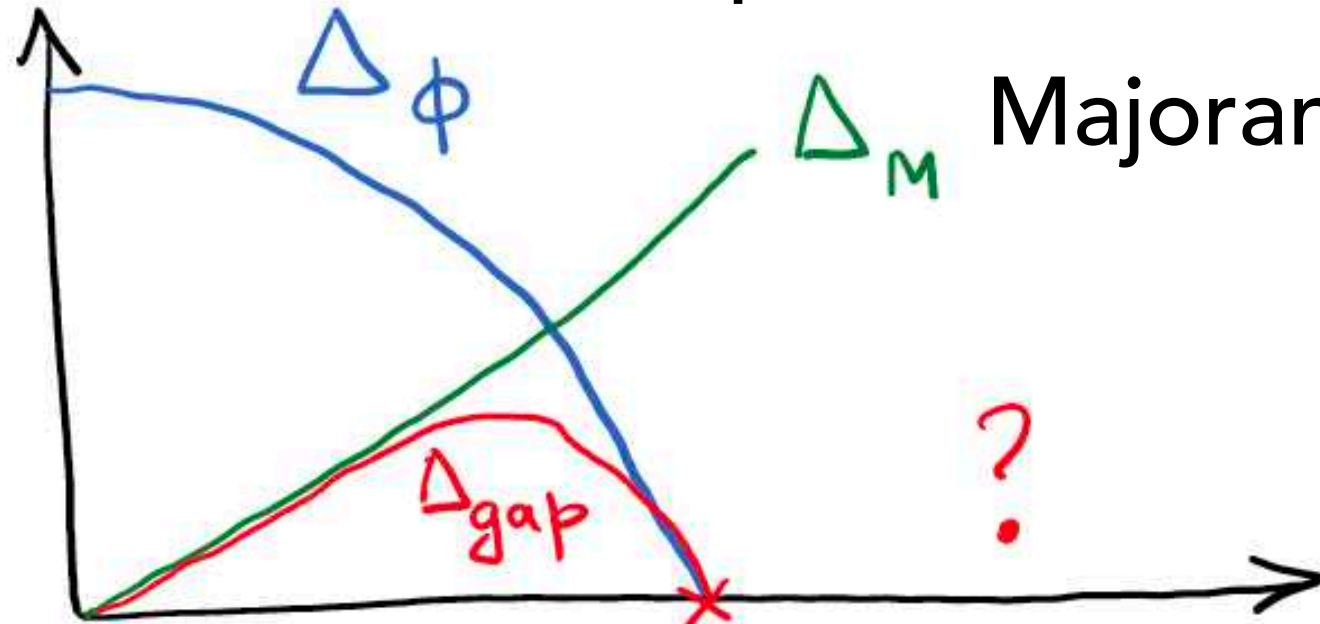

# Gapless $Z_2$ Quantum Spin Liquid

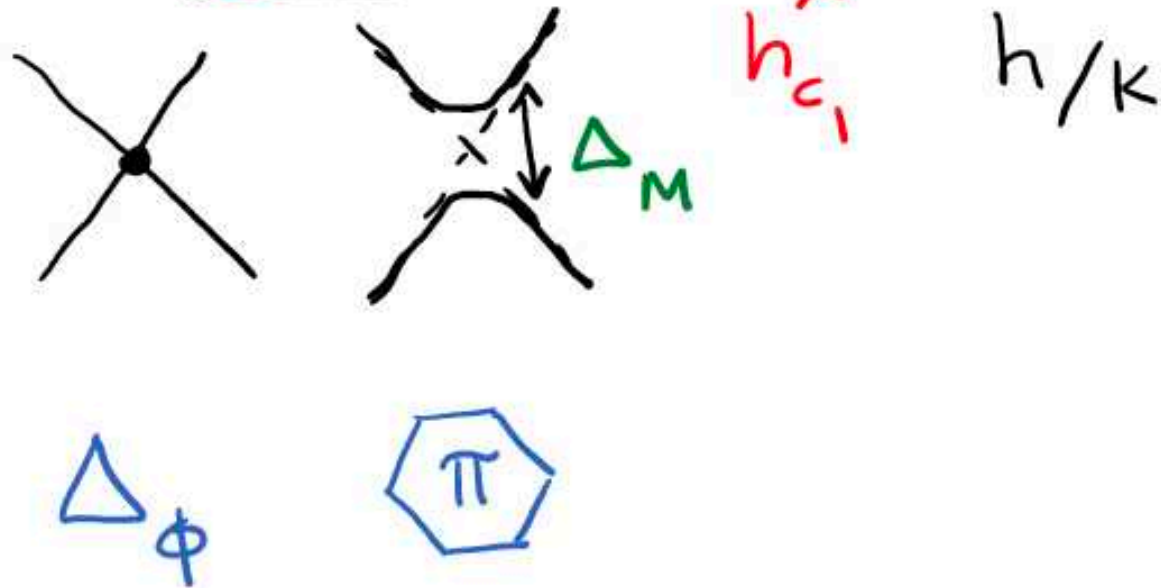# Now add a magnetic field…

$$H = H_K + h \sum_{i\alpha} S_i^\alpha$$

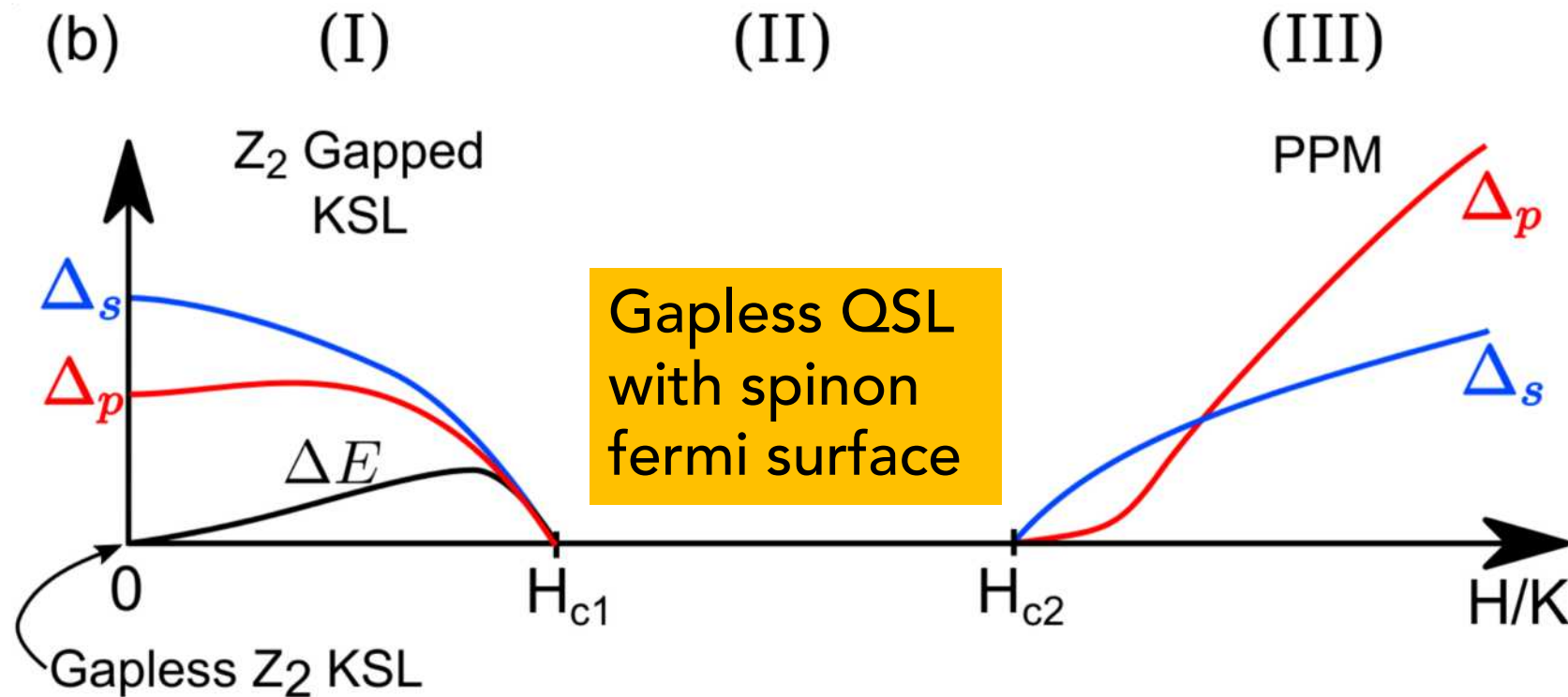Focus on here:
AF Kitaev interactions and field along $h||[111]$

Flux Gap

Majorana Gap

$\Delta_\phi$

$\Delta_M$

?

$\Delta_{gap}$

$h_{c_1}$

$h/K$

$\Delta_M$

$\Delta_\phi$ π

# Our Main Results: Kitaev Model in a Magnetic Field



(b)    (I)    (II)    (III)

$Z_2$ Gapped KSL

PPM

$\Delta_p$

$\Delta_s$

$\Delta_s$

$\Delta_p$

Gapless QSL with spinon fermi surface

$\Delta E$

0    $H_{c1}$    $H_{c2}$    H/K

Gapless $Z_2$ KSL

$\Delta_s$: single spin flip energy
$\Delta_p$: 2-spin flip energy

Results based on exact diagonalization ED and
Density matrix renormalization group (DMRG)

David Ronquillo    Adu Vengal    Nirav Patel    Subhasree Pradhan

Field-orientation-dependent spin dynamics of the Kitaev honeycomb model
Phys. Rev. B 99, 140413 (2019)

Magnetic field induced intermediate gapless spin-liquid phase with a spinon Fermi surface
PNAS 201821406 (2019)

Two-Magnon Bound States in the Kitaev Model in a [111]-Field
PRB 101, 180401 (2020)

Related work:
Z. Zhu, et al., Phys. Rev. B 97, 241110 (2018)
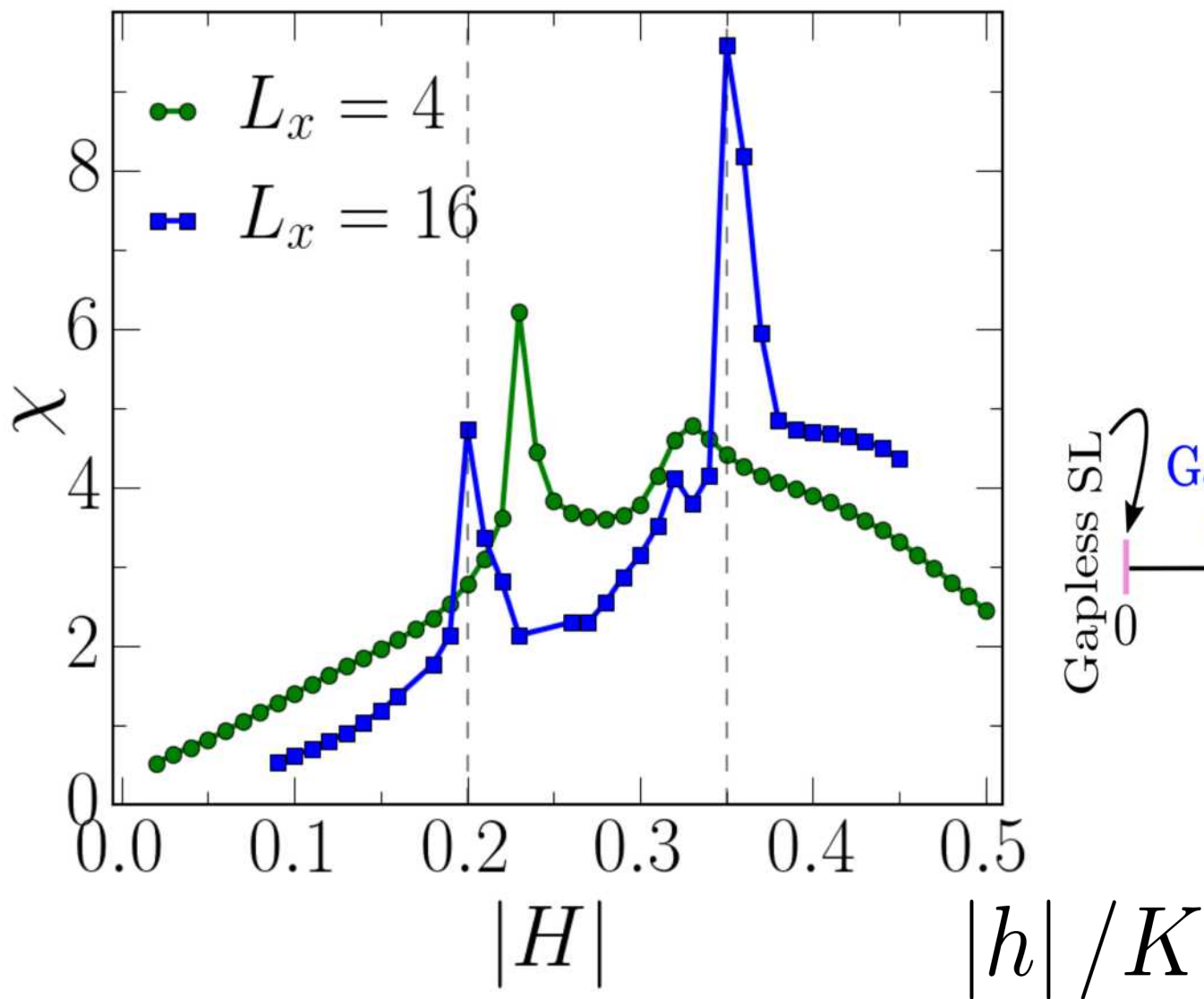M. Gohlke, et al., Phys. Rev. B 98, 014418 (2018)
C. Hickey and S. Trebst, Nat. Comm. 10, 530 (2019)
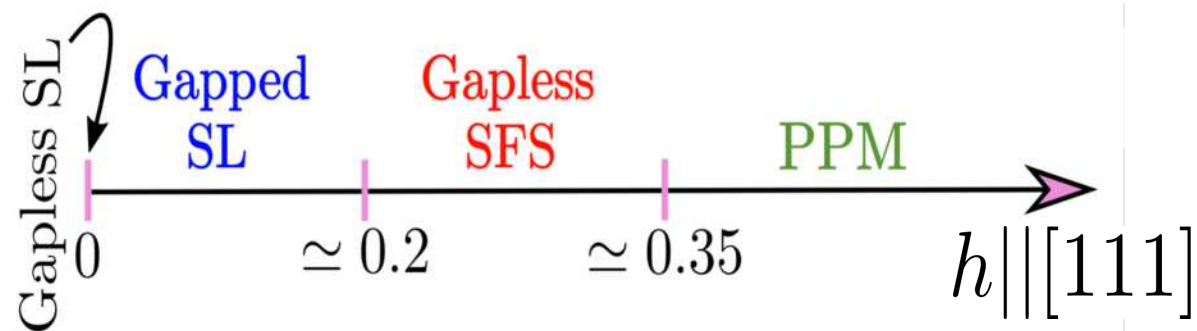H.C. Jiang et al. arXiv 1809.08247
Y. Motome and J. Nasu, JPSJ 89, 012002 (2020)

# Evidence for TWO phase transitions

# Kitaev Model + Magnetic field: $h\|[111]$
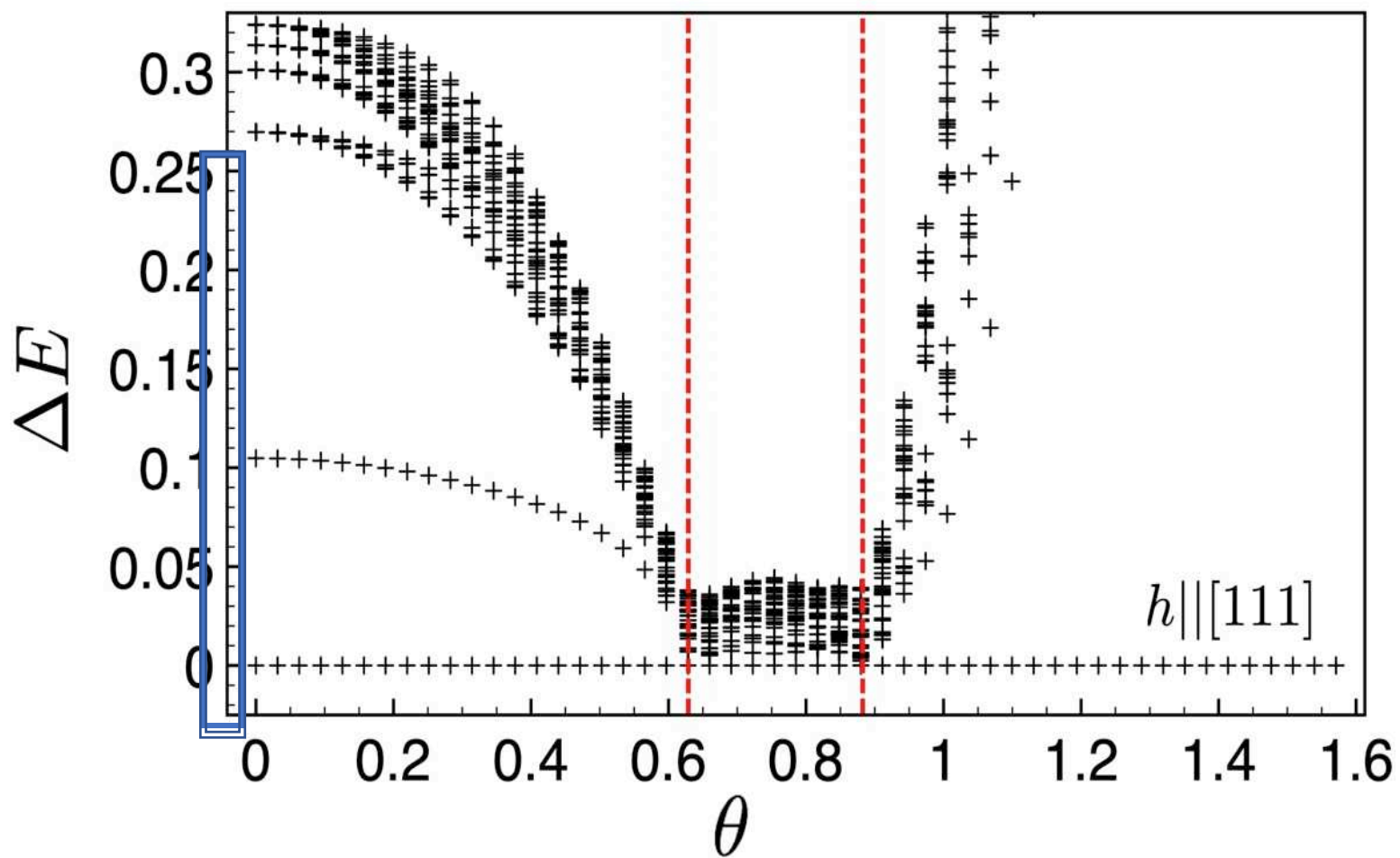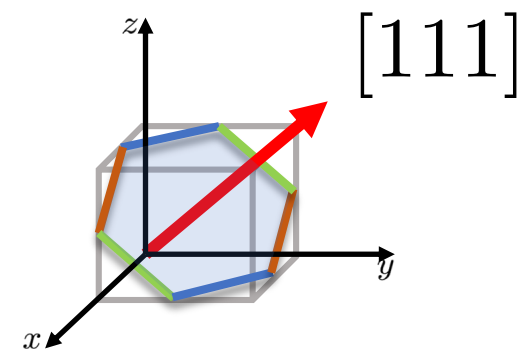## susceptibility



$$H = H_K + h \sum_{i\alpha} S_i^\alpha$$

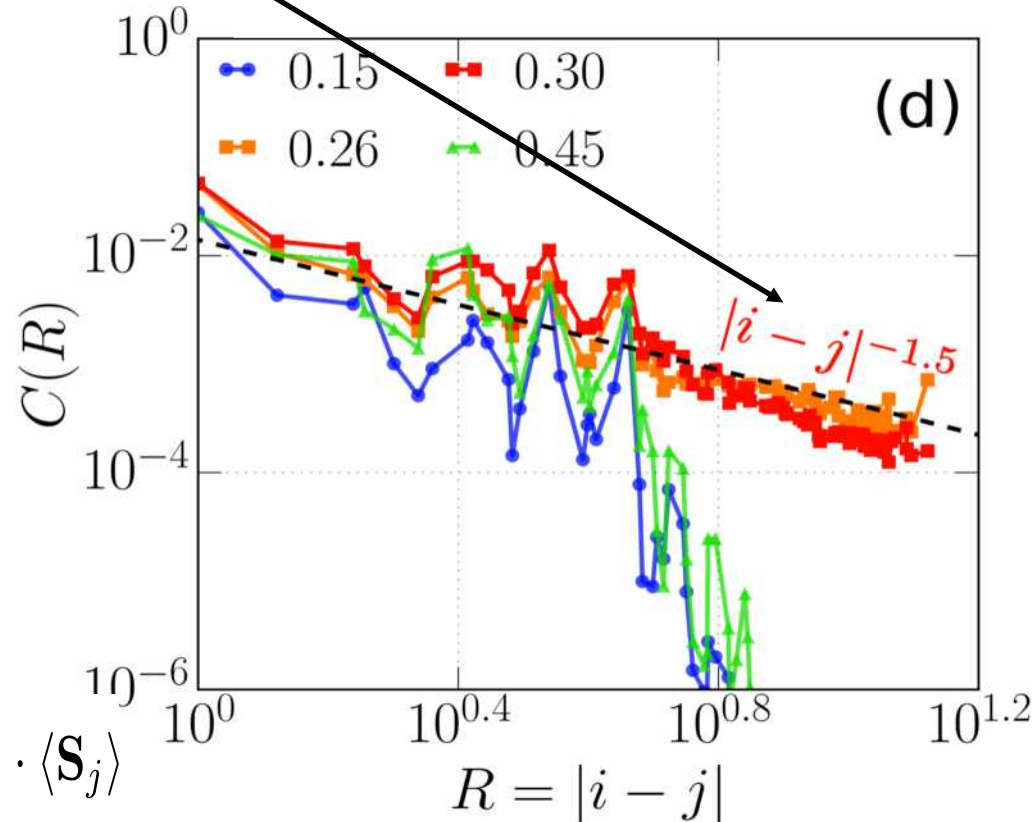Density Matrix Renormalization Group calculations with 160 spins
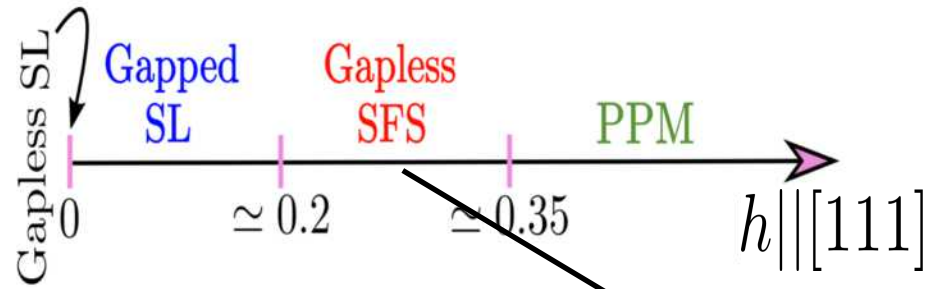
# Evidence for gapless intermediate phase

# Energy Spectra in a field



$h||[111]$

$[111]$

$\theta \sim |\vec{h}|/K$

# Spin-Spin Correlations in Intermediate phase



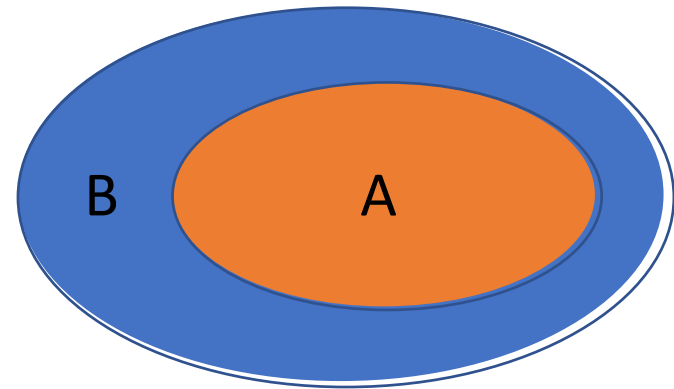Distinct power law decay of real-space spin-spin correlations!

$$C(R) = \frac{1}{N_R} \sum_{R=|i-j|} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle - \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle$$

# Evidence for QSL

# Entanglement Entropy for a Gapped QSL

$$\rho_A \equiv \mathrm{Tr}_B(\rho)$$

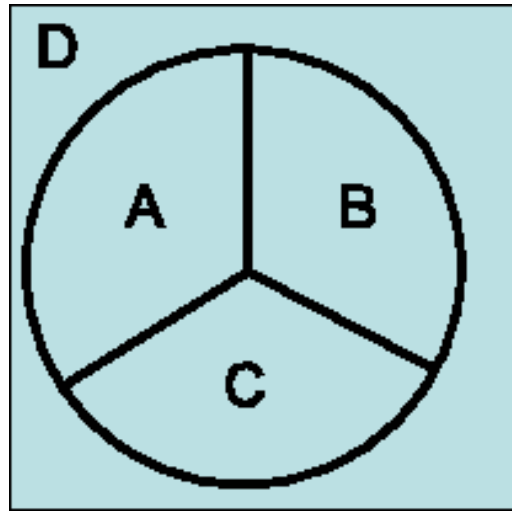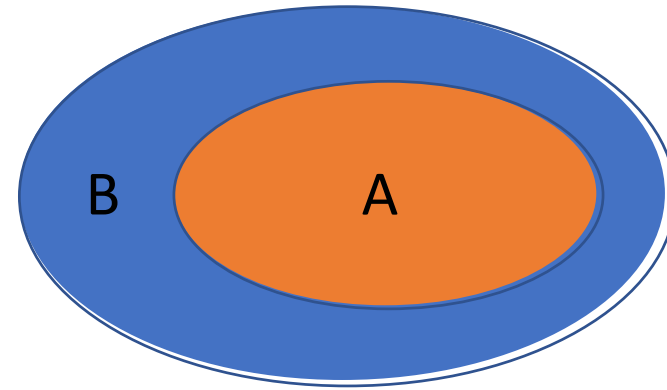$$S_A = -\mathrm{Tr}\rho_A \log \rho_A$$



$$S_A \sim \alpha L$$
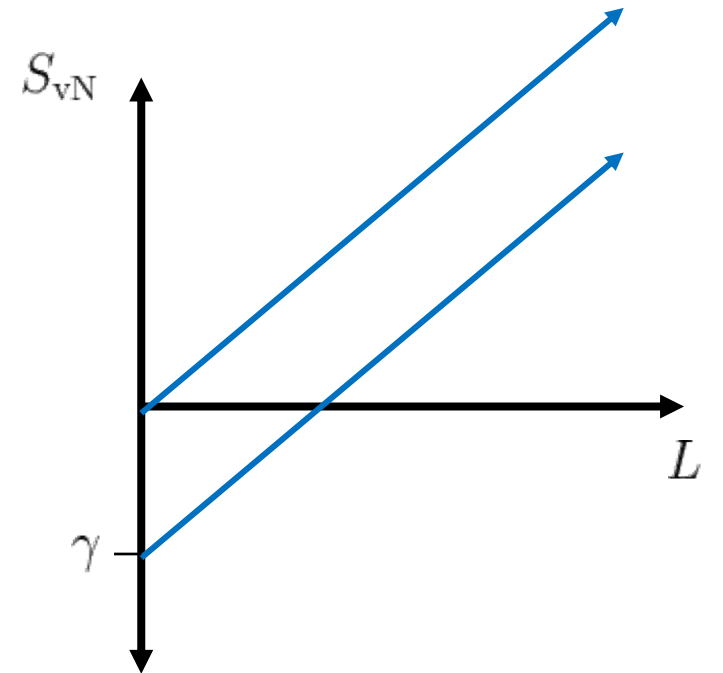
"Area Law" Entanglement in a gapped system

# Topological Entanglement Entropy $\gamma$



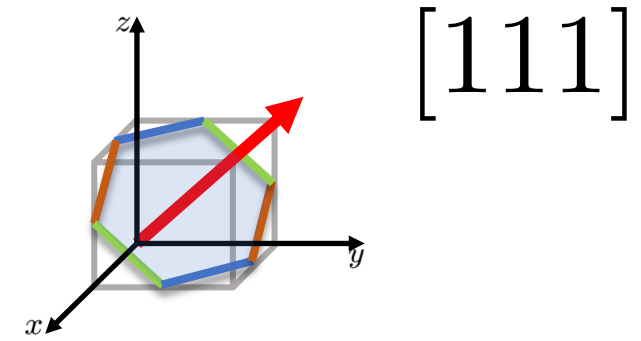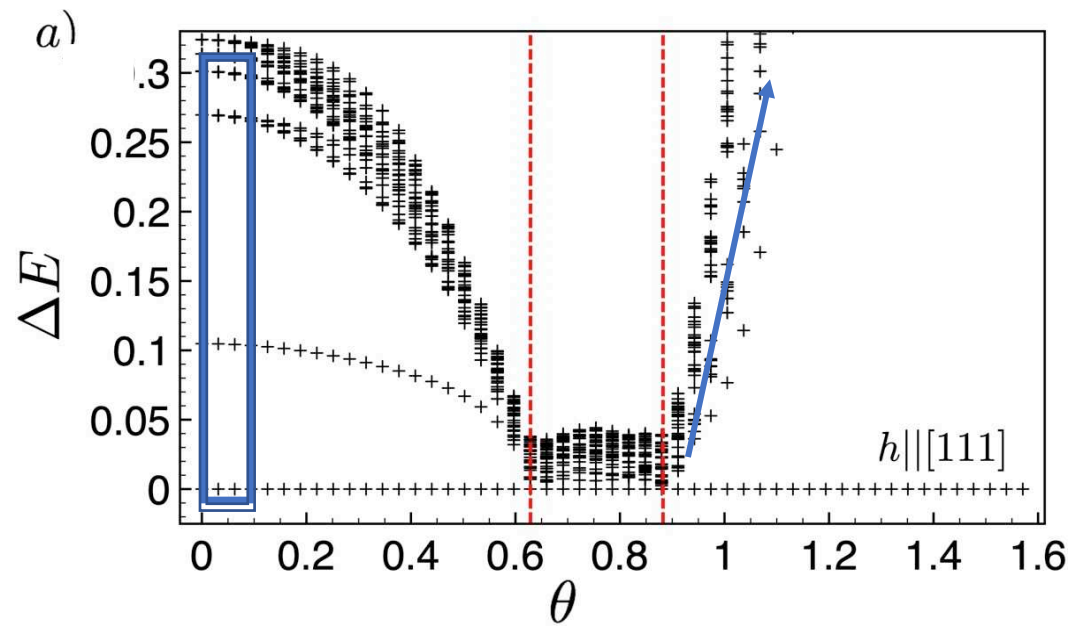$$S_A \sim \alpha L - \gamma$$

with $\gamma > 0$.

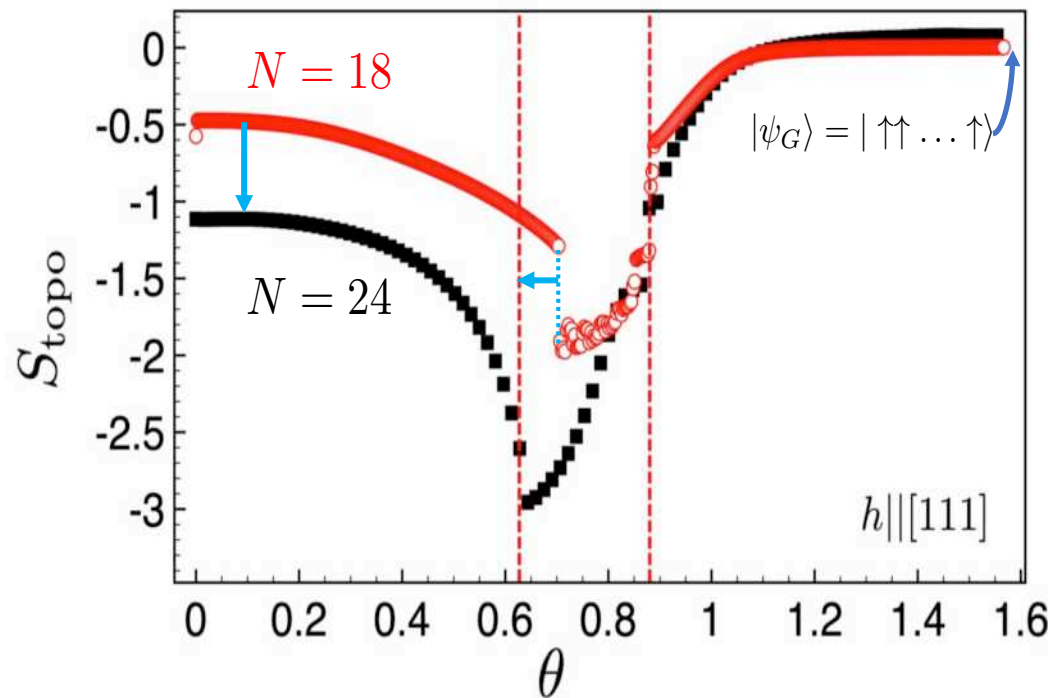Kitaev-Preskill Construction
to extract $\gamma$

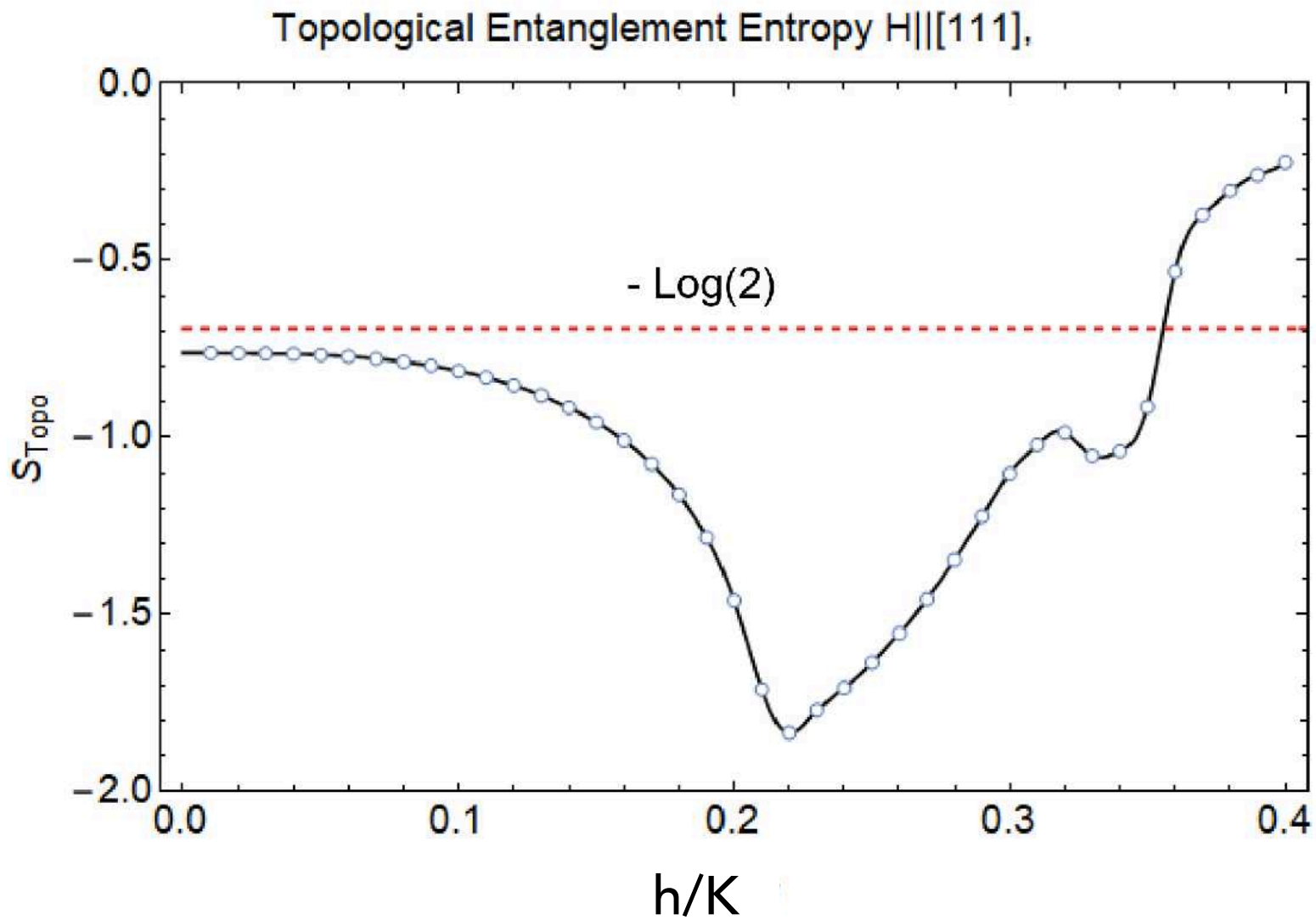$$S_{\text{topo}} = S_A + S_B + S_C - S_{AB} - S_{BC} - S_{CA} + S_{ABC}$$

Energy
Spectra



$$\theta \sim |\vec{h}|/K$$

TEE



Ronquillo, Vengal, Trivedi,
PRB 99, 140413(R) (2019)

Topological Entanglement Entropy H∥[111],

$-\text{Log}(2)$

$S_{Topo}$

h/K

Topological entanglement entropy: Information resource

Ian Osbourne

Finite $\gamma$ implies the existence of topological order

$\rightarrow$ long range entanglement structure
$\rightarrow$ Quantum dimension of excitations

# Gapped Non abelian KSL

Vacuum $\quad 1 \sim d_1 = 1$

Fermion $\quad \epsilon \sim d_\epsilon = 1$ $\quad$ abelian

Vortex $\quad v \sim d_v = \sqrt{2} > 1$ $\quad$ Non abelian

$$\rightarrow D = \sqrt{d_1^2 + d_\epsilon^2 + d_v^2}$$
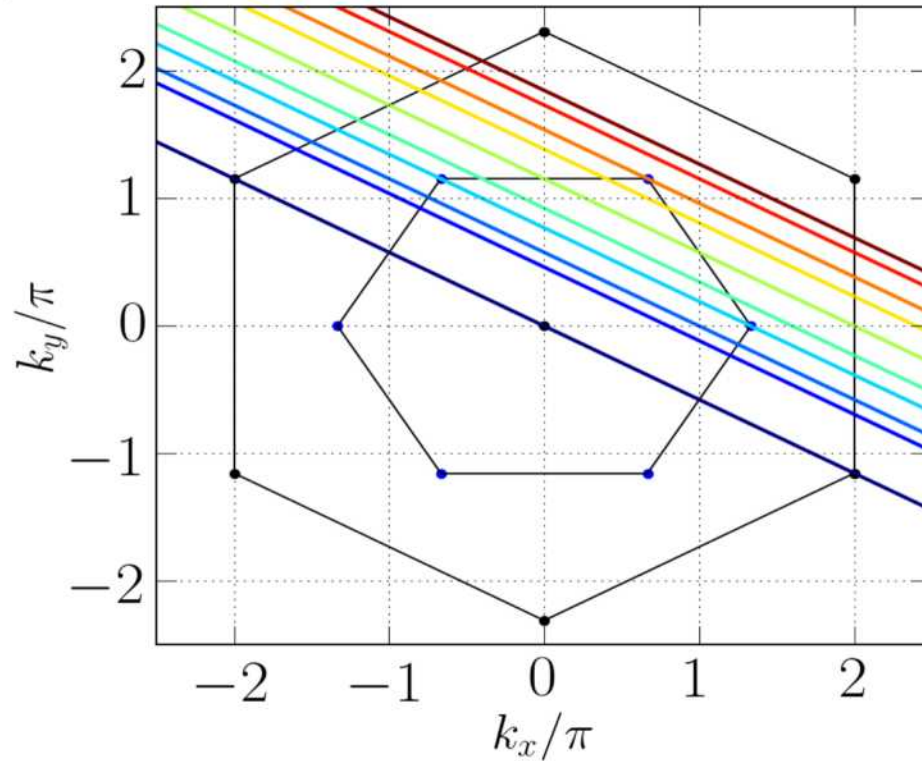
$$= \sqrt{1 + 1 + 2}$$

$$= 2$$

$$\rightarrow \gamma = \log D = \log 2$$

# Evidence for spinon Fermi surface

# Kitaev Model: spin structure factor



Brillouin Zone: Momenta cuts
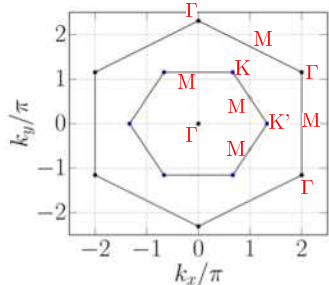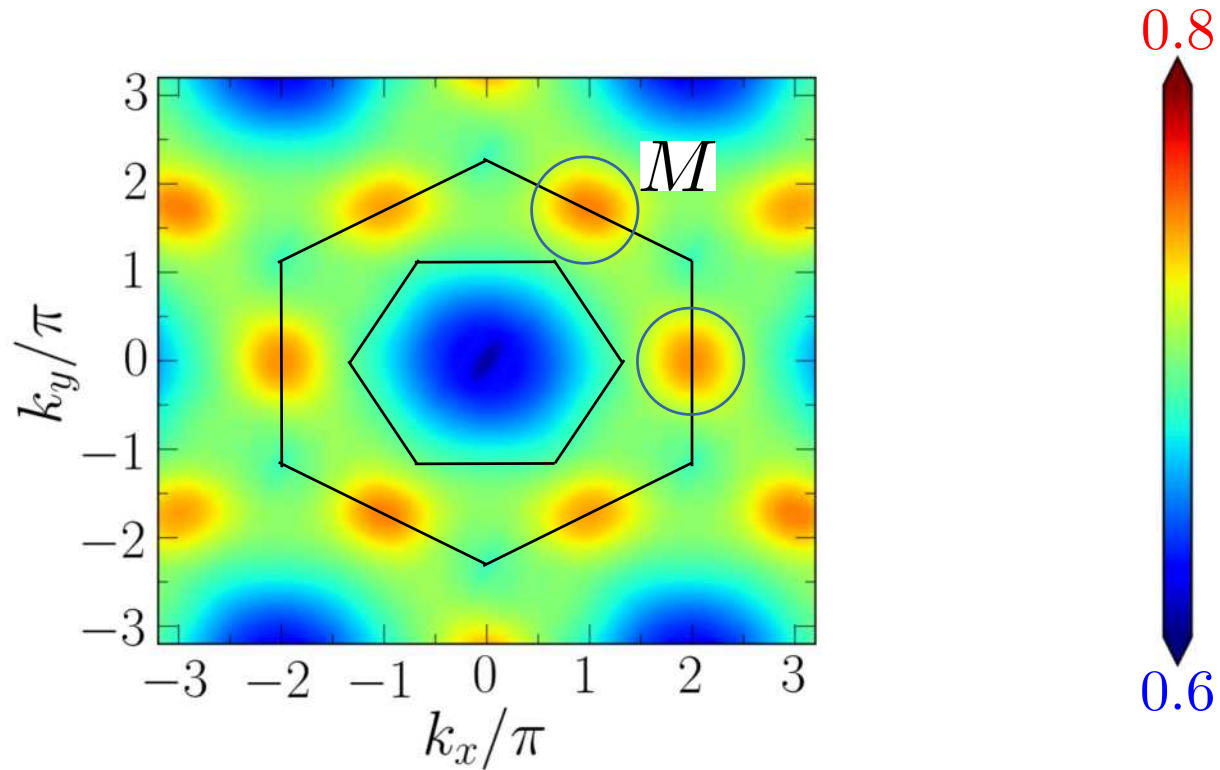
$$H_K = K \sum_{\langle ij \rangle} S_i^x S_j^x + S_i^y S_j^y + S_i^z S_j^z$$
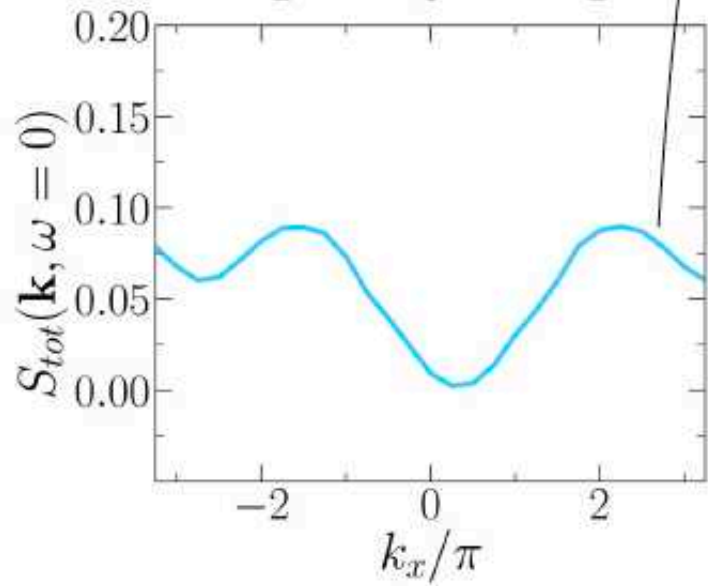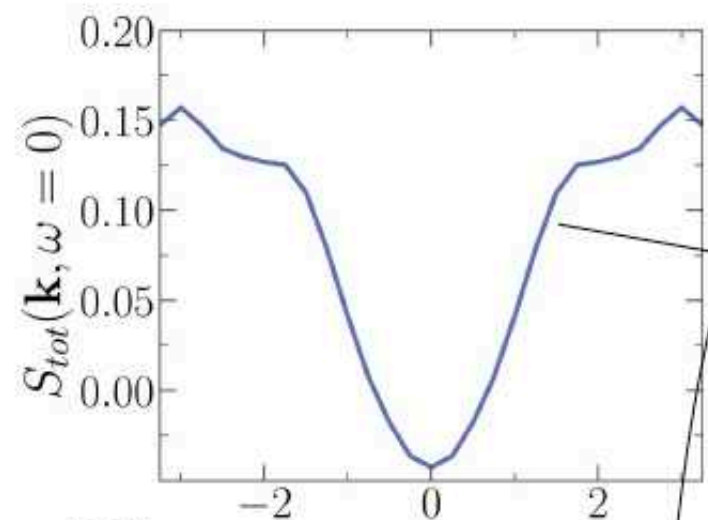
DMRG++ Open Source:
https://web.ornl.gov/~gz1/dmrgPlusPlus/
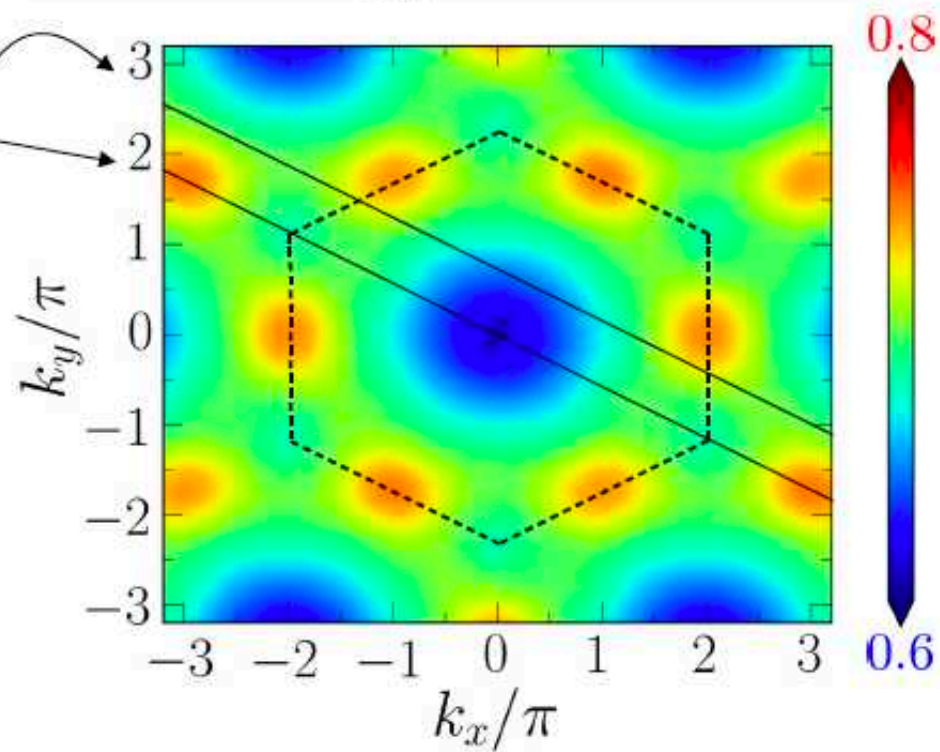
# Structure Factor S(k) – Intermediate phase



$$S_{\gamma\gamma'}(\mathbf{k}) = \frac{1}{L^2} \sum_{i\in\gamma, j\in\gamma'} e^{-i\mathbf{k}\cdot\mathbf{r_{ij}}} \left[ \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle - \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle \right]$$

$$S_{tot} = \sum_{\gamma\gamma'} S_{\gamma\gamma'} \qquad S(k) = \begin{bmatrix} S_{AA} & S_{AB} \\ S_{BA} & S_{BB} \end{bmatrix}$$
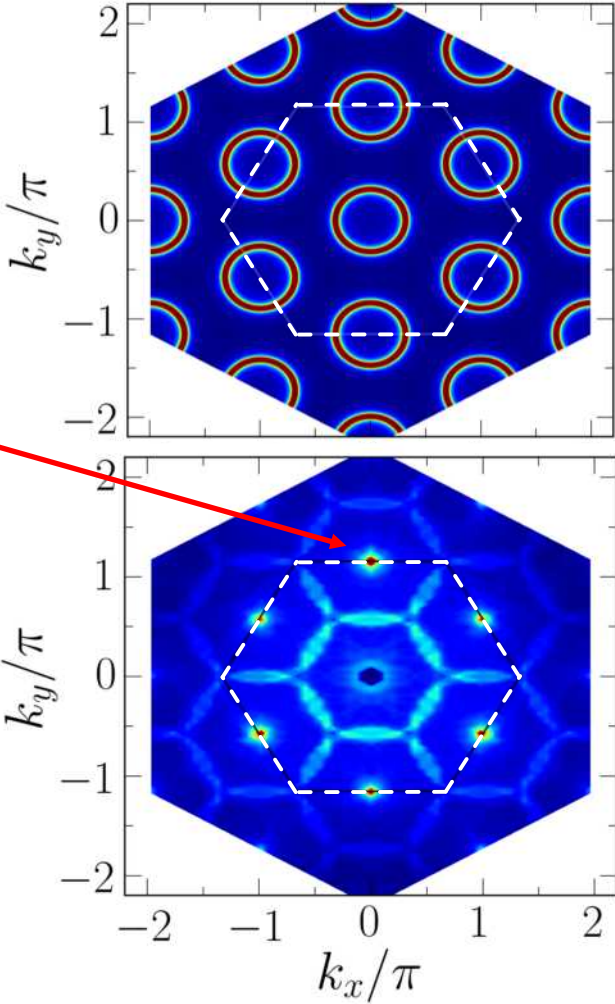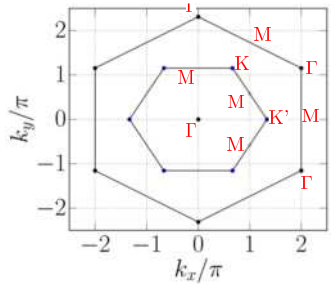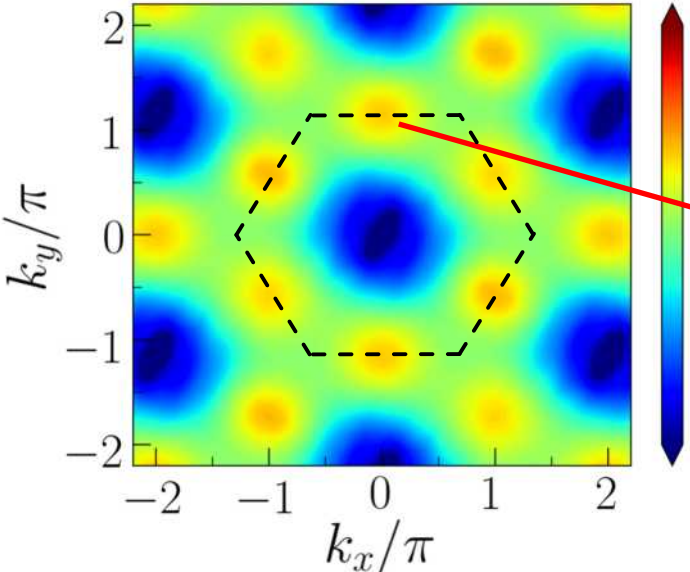
$$S_{tot}(\mathbf{k}) \propto \int_0^\infty S_{tot}(\mathbf{k}, \omega) d\omega$$
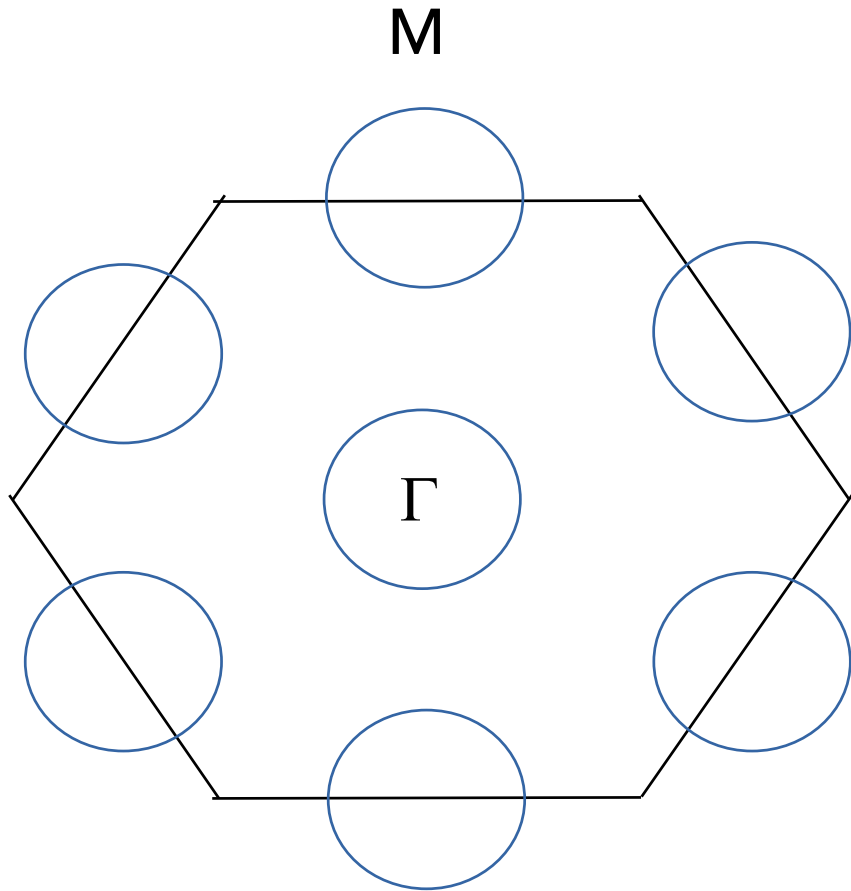
# S(k) → spinon Fermi surface

DMRG Results



$A(\mathbf{k}, \omega = 0)$

Conjectured
Fermi
surface

$\omega = 0$

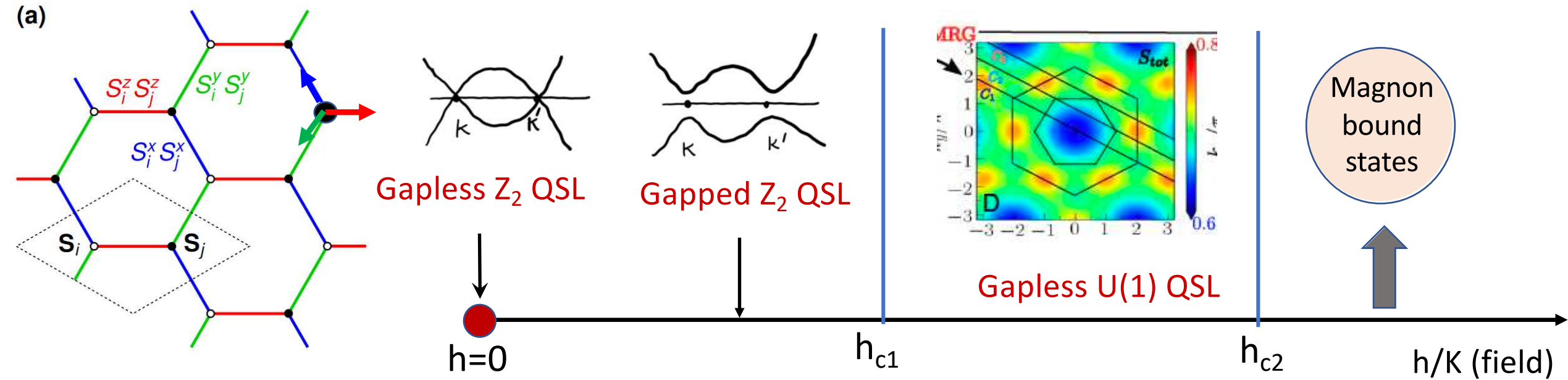$\Im(\mathbf{k}) = \sum_{q} A(\mathbf{k} + \mathbf{q}) A(\mathbf{q})$

# "Fermi Surface" of spinons in a Mott insulator!

M



Test using VMC on projected wave function

Singularities at all the M points related by C3 Rotations and Translations

# Predictions for Kitaev magnets



(a)

$S_i^z S_j^z$   $S_i^y S_j^y$

$S_i^x S_j^x$

$S_i$   $S_j$

Gapless $Z_2$ QSL

Gapped $Z_2$ QSL

Gapless U(1) QSL

Magnon bound states

h=0

$h_{c1}$

$h_{c2}$

h/K (field)

Ronquillo, Vengal, Trivedi, PRB **99**, 140413 (2019)
Patel & Trivedi, *PNAS* **116**, 12199 (2019)
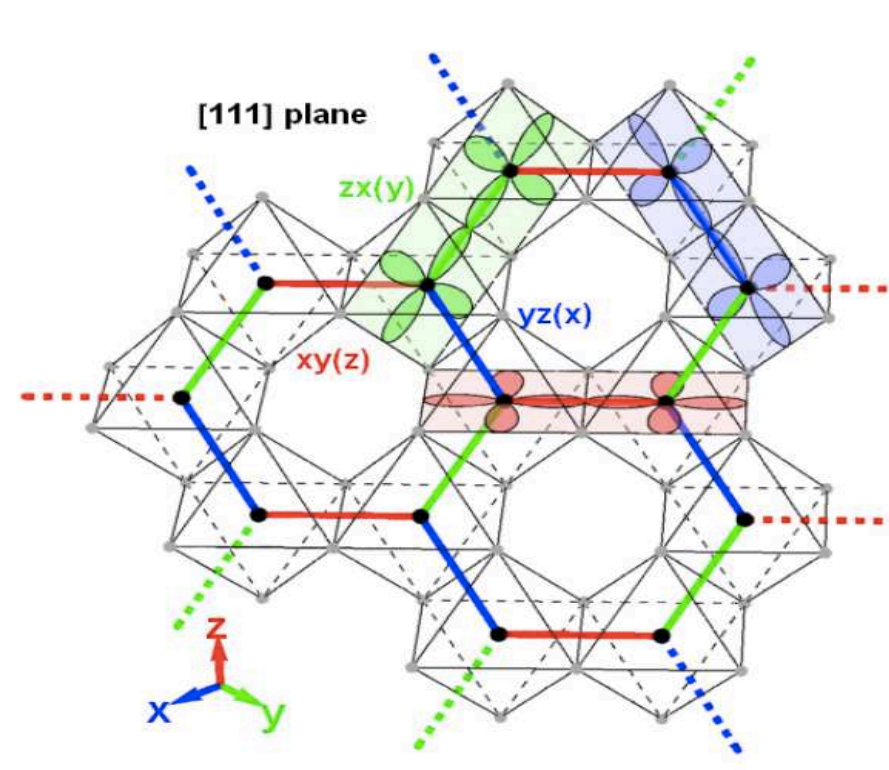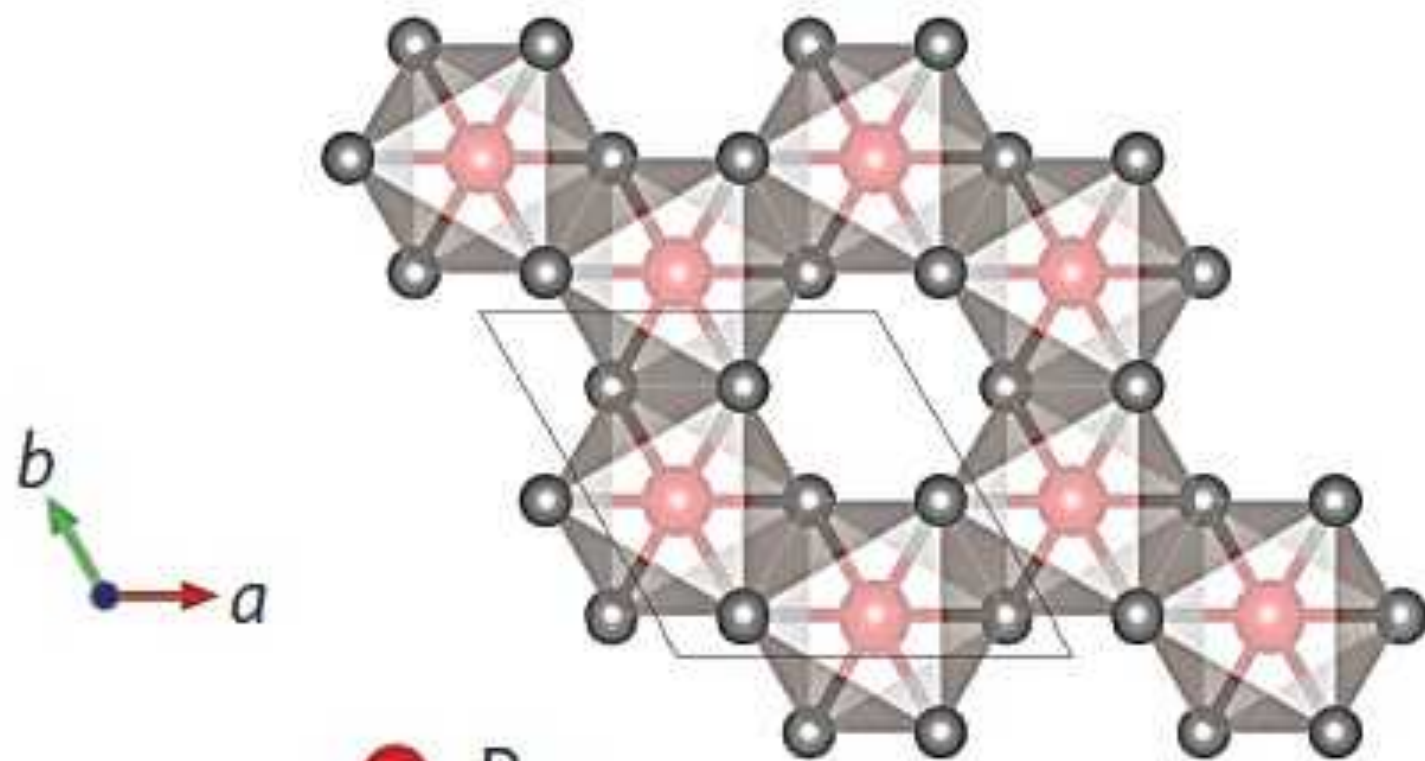Pradhan, Patel, Trivedi, PRB 101, 180401 (2020)

# Roadmap

➢ Big picture

➢ 2D: Kitaev Model
❖ Discovery of a new gapless QSL with a spinon Fermi surface

❖ Spectrum of 1 spin flip and 2 spin flip excitations

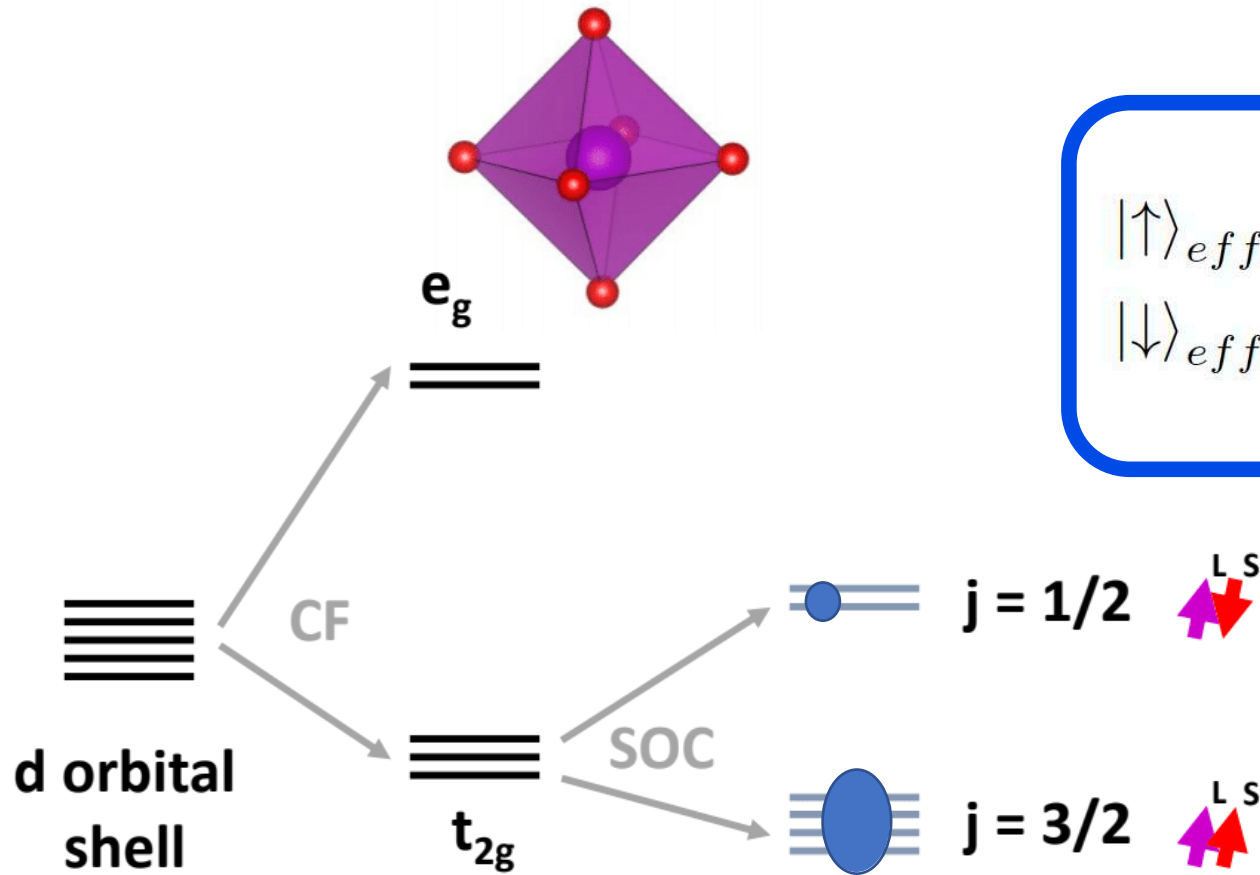➢ QSL Materials

➢ How do you detect a QSL?

➢ Going forward….

# Main ingredients for Kitaev materials

α–RuCl₃
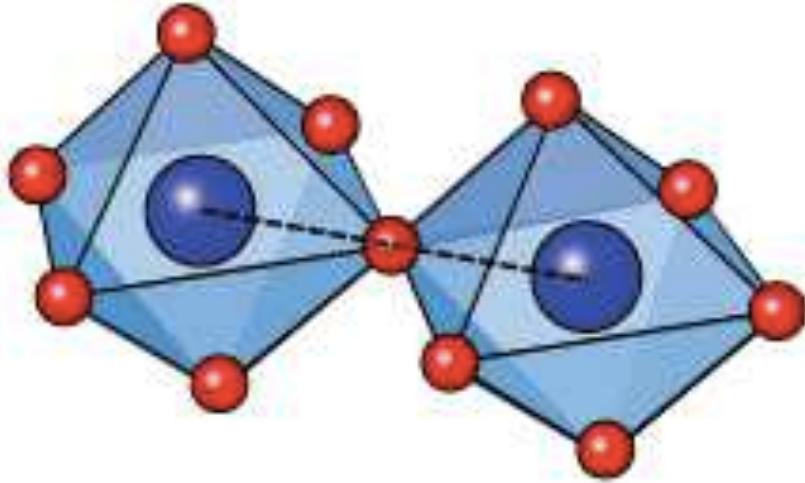
Ru
Cl

# Spin-Orbit coupled Mott Insulators

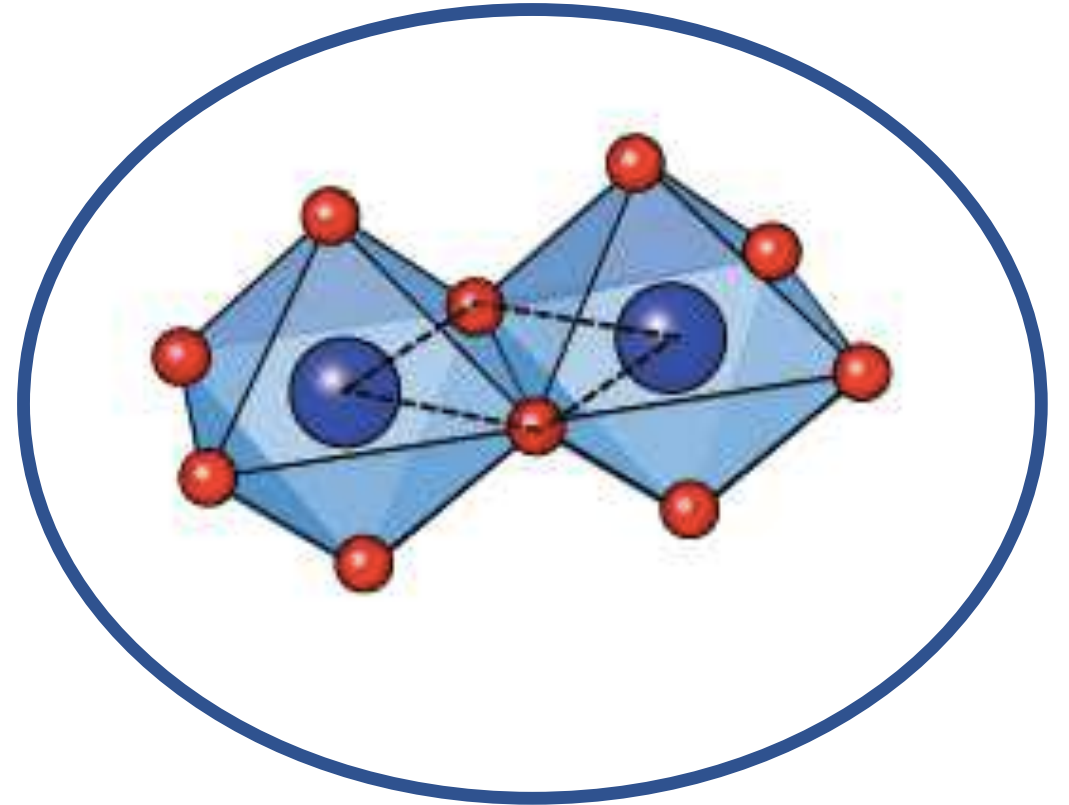| Mo | Tc | **Ru** | Rh |
|----|----|--------|-----|
| W | **Re** | **Os** | **Ir** |



$$|\uparrow\rangle_{eff} \sim i\,|zx,\downarrow\rangle + |yz,\downarrow\rangle + |xy,\uparrow\rangle$$
$$|\downarrow\rangle_{eff} \sim -i\,|zx,\uparrow\rangle + |yz,\uparrow\rangle - |xy,\downarrow\rangle$$

$e_g$

CF

d orbital shell
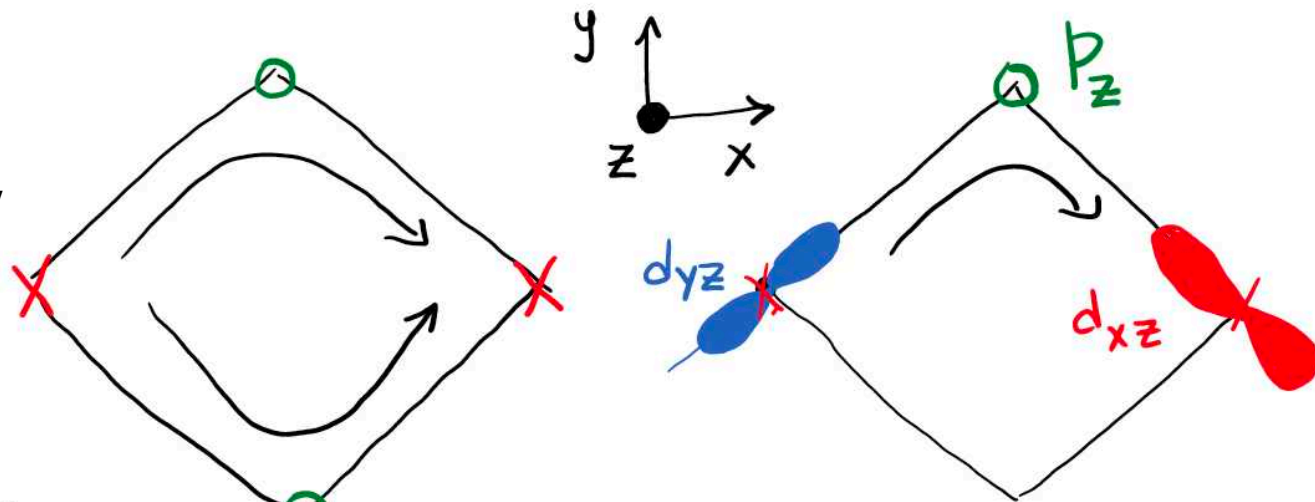
$t_{2g}$

SOC

$j = 1/2$

$j = 3/2$
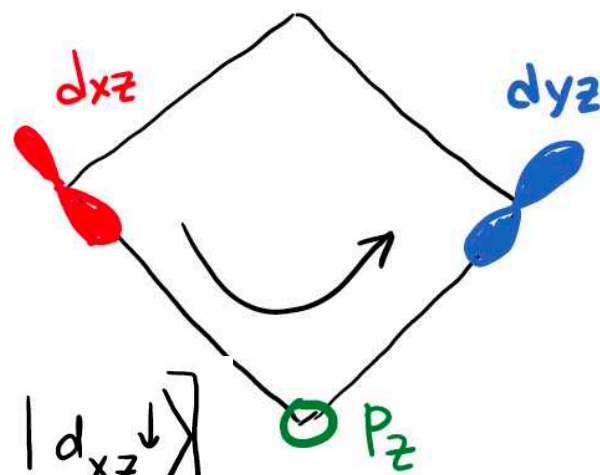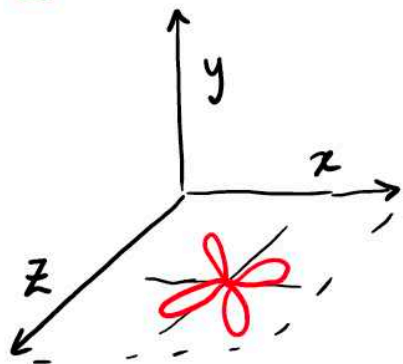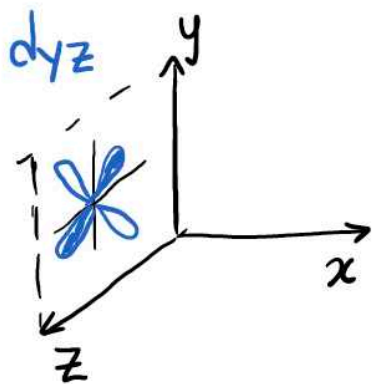
I: corner-sharing II: edge-sharing

Destructive interference between the two pathways generates bond-dependent interactions

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



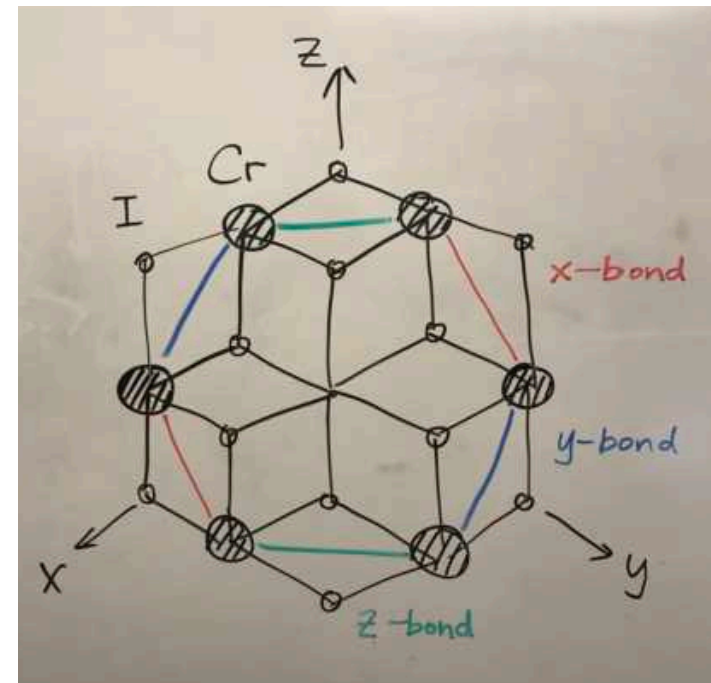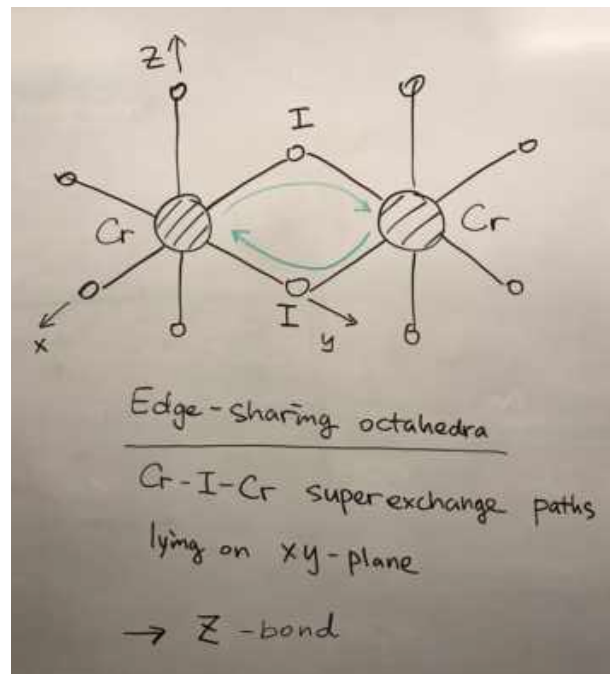hopping via $P_z$ orbital on ligand changes $d_{yz} \to d_{xz}$

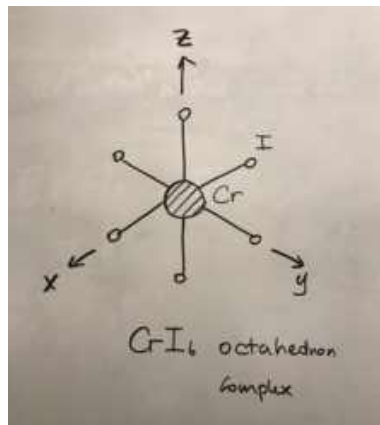$d_{xz} \longrightarrow d_{yz}$

$$| J_z = +\tfrac{1}{2} \rangle = \frac{1}{\sqrt{3}} \left[ |d_{xy}\uparrow\rangle + |d_{yz}\downarrow\rangle + i |d_{xz}\downarrow\rangle \right]$$

# Crystal structure of α-RuCl$_3$
→ candidate Kitaev material



CrI$_6$ octahedron complex



Edge-sharing octahedra

Cr-I-Cr super exchange paths lying on xy-plane
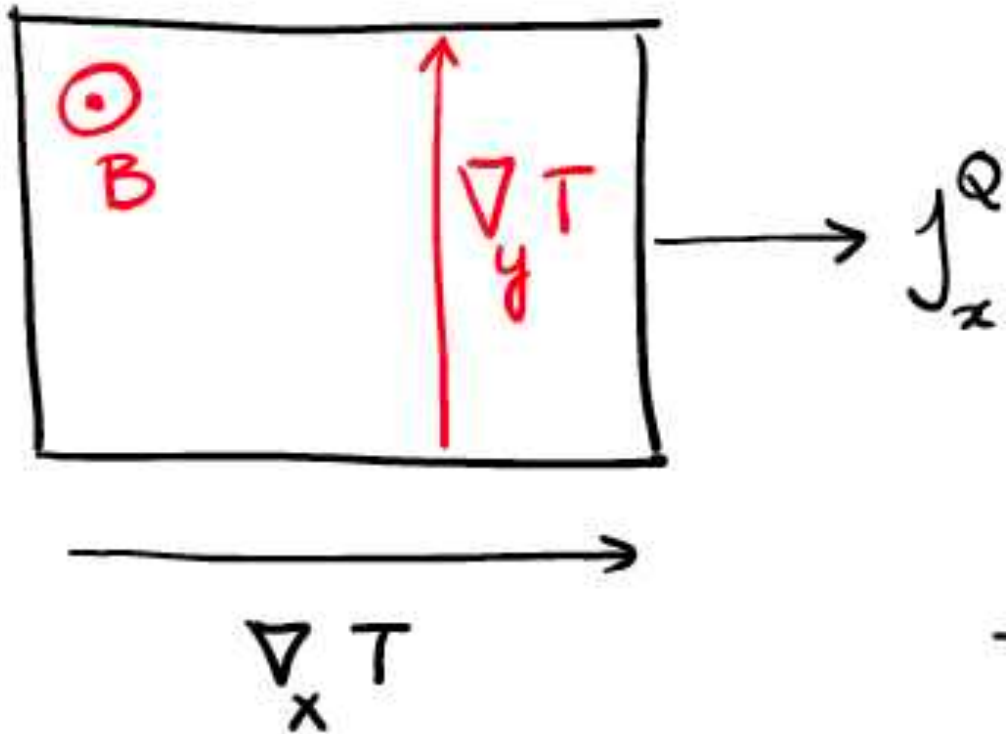
→ z-bond



x-bond

y-bond

z-bond

# Roadmap

➢ Big picture

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❖    Spectrum of 1 spin flip and 2 spin flip excitations

➢ QSL Materials

➢ How do you detect a QSL?

➢ Going forward….
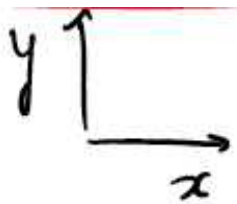
# Thermal Hall Conductivity



$$\vec{J}^Q = \overleftrightarrow{K}(-\vec{\nabla}T)$$

$$\overleftrightarrow{K} = \begin{pmatrix} K_{xx} & K_{xy} \\ -K_{xy} & K_{xx} \end{pmatrix}$$

$J_y^Q = 0$
(impose condition)

**Material :** $\alpha - RuCl_3 \simeq$ Kitaev Magnet

$$J_K \simeq \underline{80\ K}$$



For $S = 1/2$

1 Tesla $\simeq$ 1 K

$3K < T < 5K$

$$K_{xy} = \left(\frac{1}{2}\right) K_Q$$

$$K_Q = \frac{\pi^2}{6} k_B^2/\hbar$$

Material :  $\alpha - RuCl_3$  $\simeq$  Kitaev Magnet

$$J_K \simeq \underline{80\ K}$$

7K

T

zig-
zag

"QSL"

7T

$H_{||}$

What object carries heat?
Electron? Spin? Phonon?
Majorana edge mode

$x$

$$-\frac{\pi}{12}\frac{k_B^2}{\hbar}$$

$\frac{k_{xy}}{T}$

7T    9T

$3K < T < 5K$

$H_{||}$ (T.)

$$k_{xy} = \left(\frac{1}{2}\right) k_Q$$

$$k_Q = \frac{\pi^2}{6}\frac{k_B^2}{\hbar}$$

# Signatures of a QSL: *quantized* thermal Hall conductance
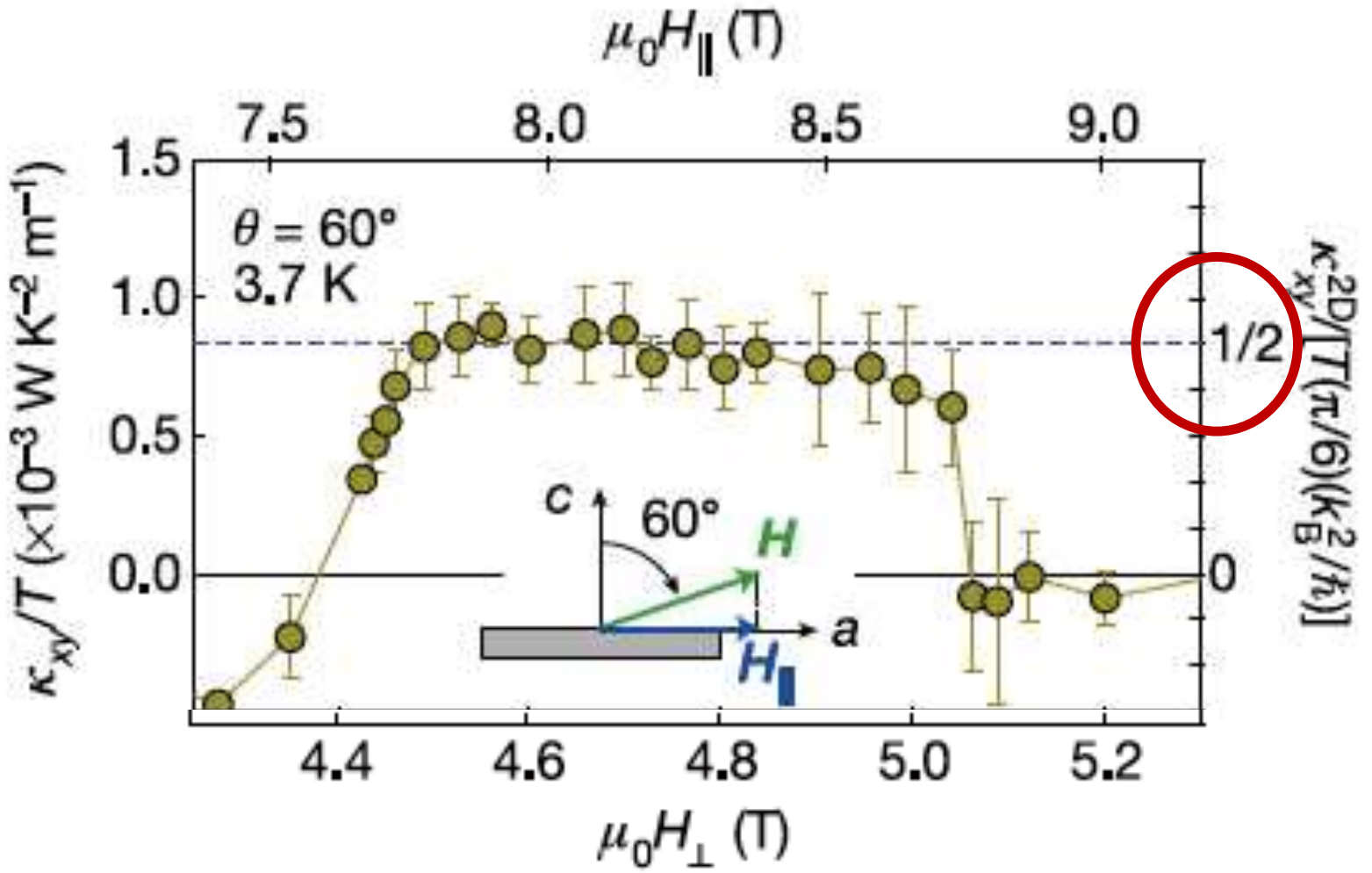


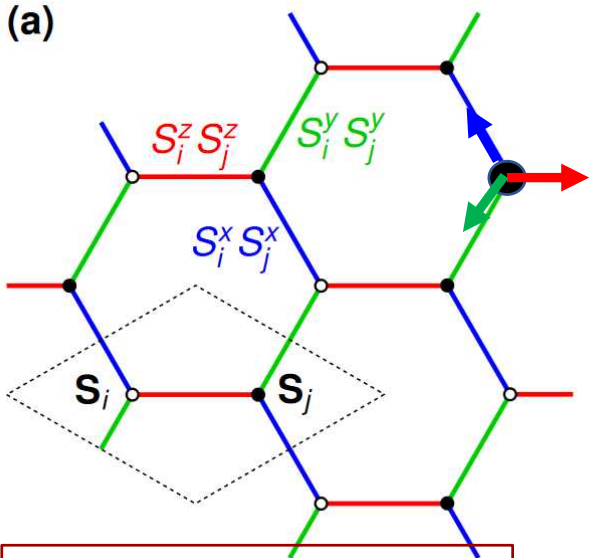$$\kappa_{xy}^{2D} = \left[ \frac{1}{2} \right] \frac{\pi}{6} \frac{k_B^2}{\hbar} T$$

$$\kappa_{xy}^{2D} = \kappa_{xy} d$$

Y. Kasahara, T. Ohnishi, Y. Mizukami, O. Tanaka, Sixiao Ma, K. Sugii, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, T. Shibauchi & Y. Matsuda, Nature 559, 227 (2018).
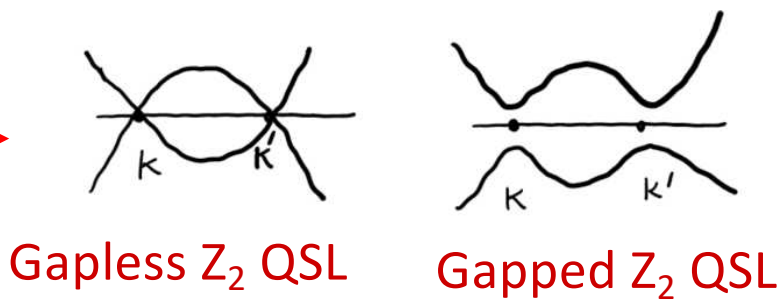
# Predictions for Kitaev magnets



**(a)**

$S_i^z S_j^z$    $S_i^y S_j^y$

$S_i^x S_j^x$

$S_i$    $S_j$

Gapless $Z_2$ QSL     Gapped $Z_2$ QSL

Gapless U(1) QSL

Magnon bound states

h=0                 $h_{c1}$         $h_{c2}$     h/K (field)

**Frustration from bond-dependent interactions**

Kitaev (2006)

Jackeli, Khaliullin (2009)

$$\kappa_{xy}^{2D} = \left[\frac{1}{2}\right] \frac{\pi}{6} \frac{k_B^2}{\hbar} T$$

cf: FQHE    $\sigma_{xy} = \frac{1}{3}\frac{e^2}{h}$

**Fermi surface of neutral, gapless spinons in an insulator!**

$\kappa_{xx} \sim T$

**Chiral spinon edge mode → Quantized thermal Hall conductance**

Ronquillo, Vengal, Trivedi, PRB **99**, 140413 (2019)
Patel & Trivedi, *PNAS* **116**,12199 (2019)
Pradhan, Patel, Trivedi, PRB 101, 180401 (2020)

# Going forward…..

1. Predictions for Raman scattering to observe magnon bound states

2. Spin and heat transport

3. Observation of neutral spinon Fermi surfaces

4. Doped QSLs → ??