Thermodynamic properties and spin dynamics in the Kitaev spin liquid

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Department of Physics, Tokyo Institute of Technology

トポロジーが紡ぐ物質科学のフロンティア

Topological Materials Science

JN, T. Kaji, K. Matsuura, M. Udagawa, and Y. Motome, Phys. Rev. B **89**, 115125 (2014)

JN, M. Udagawa, and Y. Motome, Phys. Rev. Lett. **113**, 197205 (2014).

JN, M. Udagawa, and Y. Motome, Phys. Rev. B **92**, 115122 (2015).

JN, J. Knolle, D. L. Kovrizhin, Y. Motome, R. Moessner, Nat. Phys., **12**, 912 (2016).

J. Yoshitake, JN, and Y. Motome, Phys. Rev. Lett. **117**, 157203 (2016).

S.-H. Do et al., arXiv:1703.01081.

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Collaborators

敬称略

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- 宇田川　将文　(学習院理)
- 加藤　康之　(東大工)
- 紙屋　佳知　(理研)
- 吉竹　純基　(東大工)
- 加治　俊之　(元東大工)
- 松浦　慧介　(東大工→東大新領域)
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- D. Kovrizhin (*Univ. of Cambridge*)
- R. Moessner (*Max Plank Inst.*)

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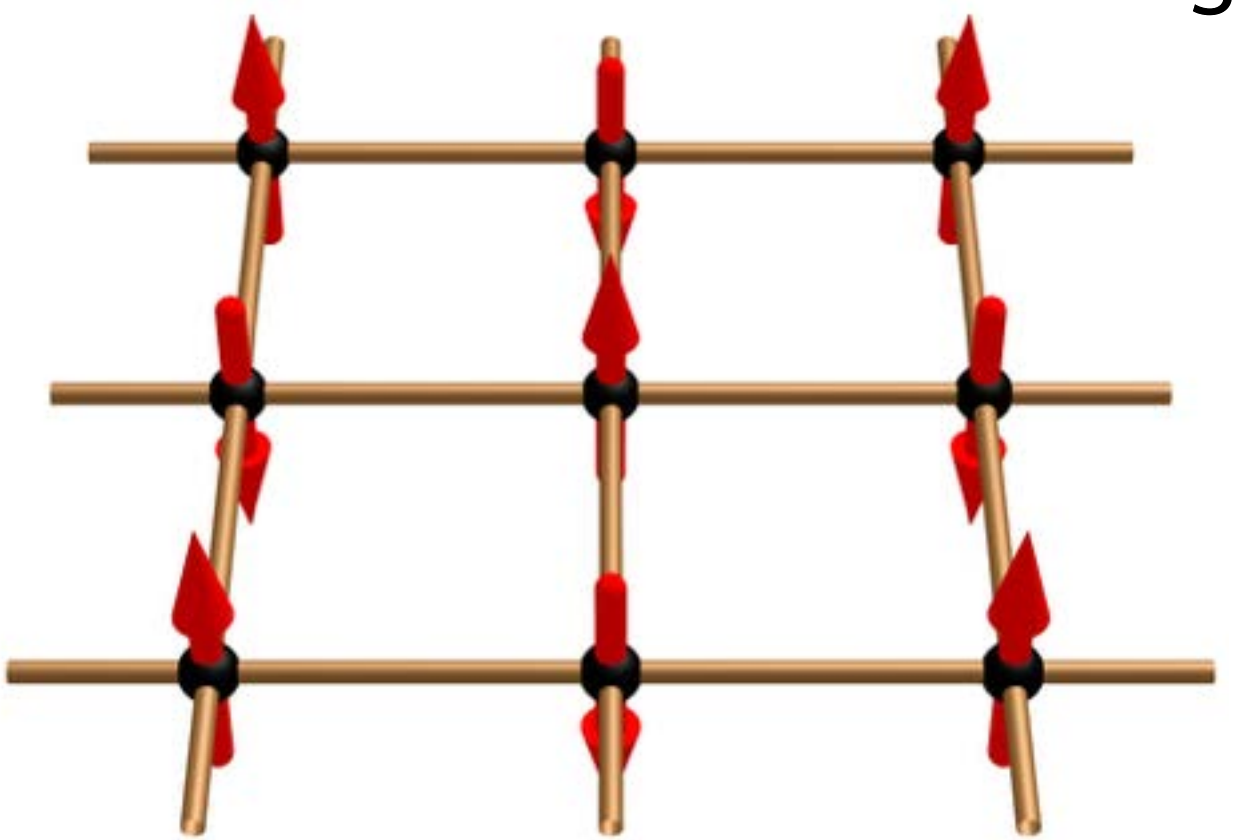
Magnetic interactions

Origin of magnetism: ***exchange interactions***



Spin lattice models (S=1/2)

- **Ising model**

$$\mathcal{H}_{\mathrm{Ising}} = J \sum_{\langle ij \rangle} S_i^z S_j^z = \frac{J}{4} \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z$$

- **Heisenberg model**

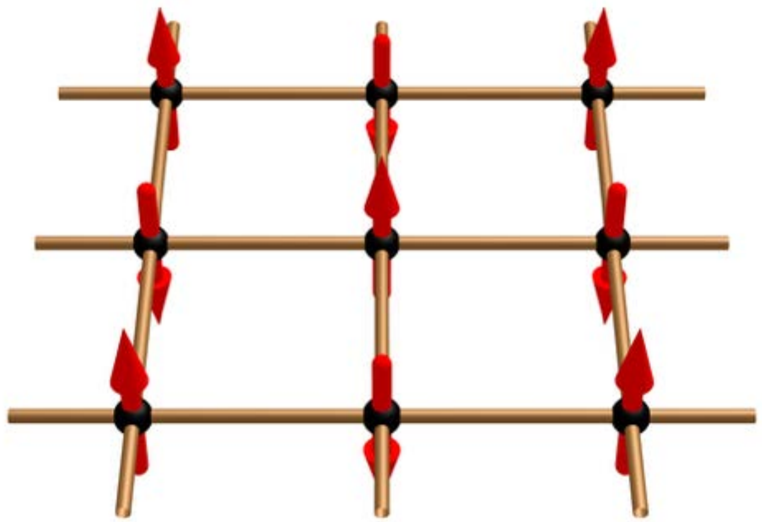
$$\mathcal{H}_{\mathrm{Heisenberg}} = J \sum_{\langle ij \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j = \frac{J}{4} \sum_{\langle ij \rangle} \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j$$

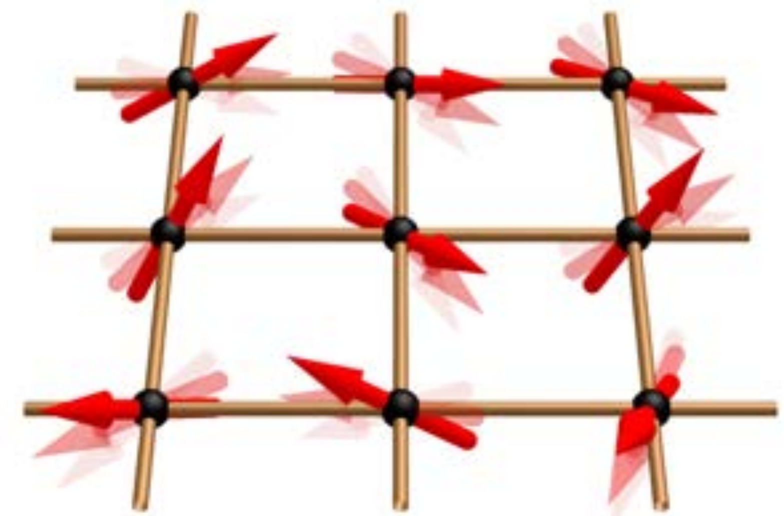
- **XY model …**

Here, we focus only on ***S*=1/2 spins**

- Anti-commutation of Pauli matrices: $\sigma^x \sigma^y = -\sigma^y \sigma^x$ etc.
- Square of Pauli matrices: $(\sigma^x)^2 = 1$ etc.
- Product of Pauli matrices: $\sigma^x \sigma^y \sigma^z = i$

Spontaneous symmetry breaking





Spontaneous symmetry breaking ⇐ ***Many-body effect***

quantum fluctuation??

- **Ising model** $$\mathcal{H}_{\text{Ising}} = J\sum_{\langle ij\rangle} S_i^z S_j^z = \frac{J}{4}\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z$$

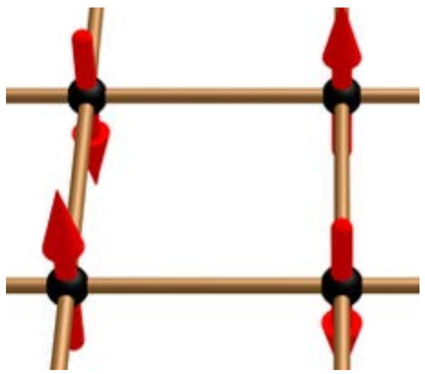
  $D > 1$ for finite T transition

- **Heisenberg model** $$\mathcal{H}_{\text{Heisenberg}} = J\sum_{\langle ij\rangle} \boldsymbol{S}_i\cdot\boldsymbol{S}_j = \frac{J}{4}\sum_{\langle ij\rangle}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j$$

  $D > 2$ for finite T transition

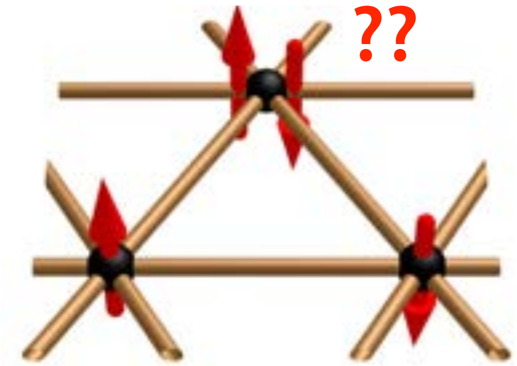
Geometrical frustration

All interactions are happy



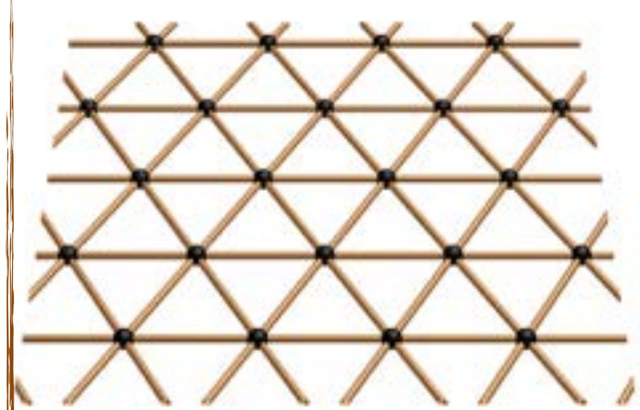
Antiferromagnetic interaction

frustrated magnetic interactions

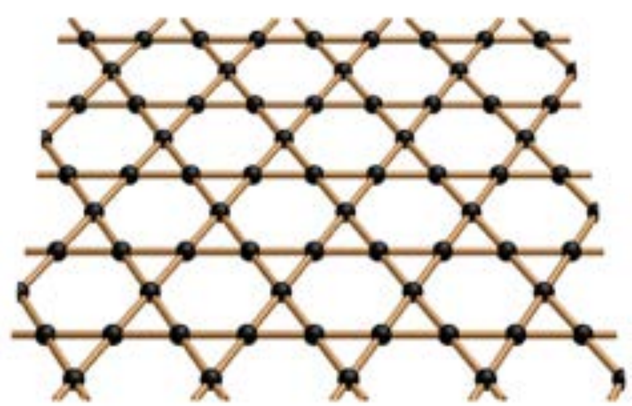


Magnetic ordering might be suppressed.

Geometrically frustrated lattices

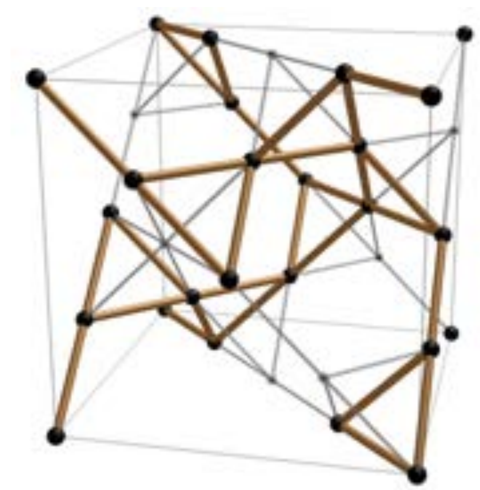


triangular lattice



kagomé lattice

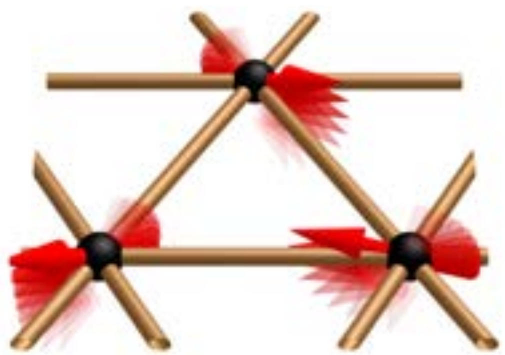
pyrochlore lattice



hyperkagomé lattice

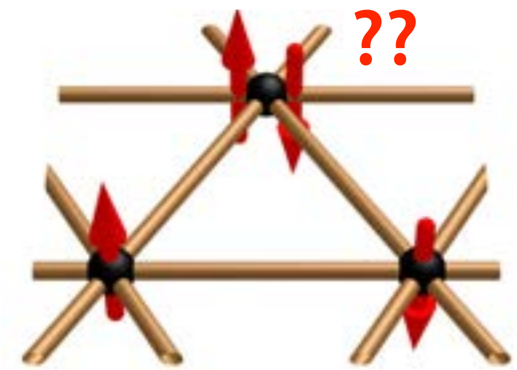
Suppression of ordering

Quantum fluctuation



+

frustrated magnetic interactions



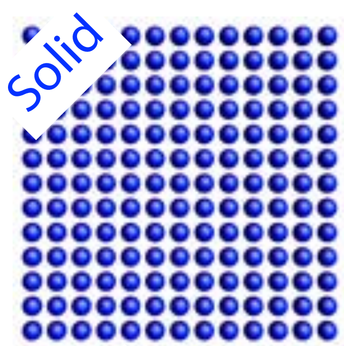
Quantum disordered state

Paramagnet

Analogy of liquid helium: not solidifying even at zero *T* due to strong quantum fluctuation?

→ **Quantum spin liquid (QSL)** ← ***Many-body effect***
Quantum fluctuation

Three states of matter:



Analogy to spin system:

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Quantum spin liquids

Anderson's suggestion in 1973

P. Anderson, Mater. Res. Bull. **8**, 153 (1973).

Ground state of the Heisenberg model on a triangular lattice

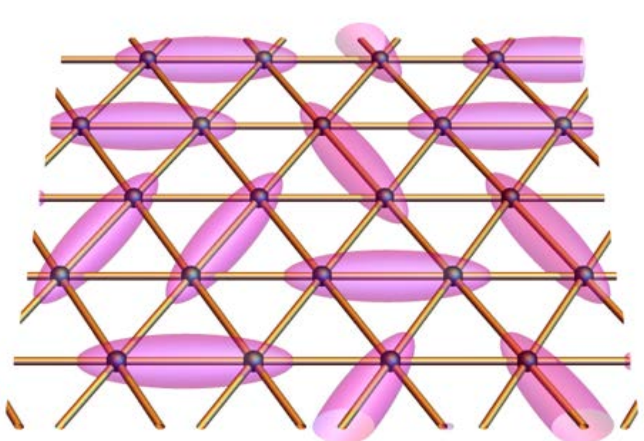
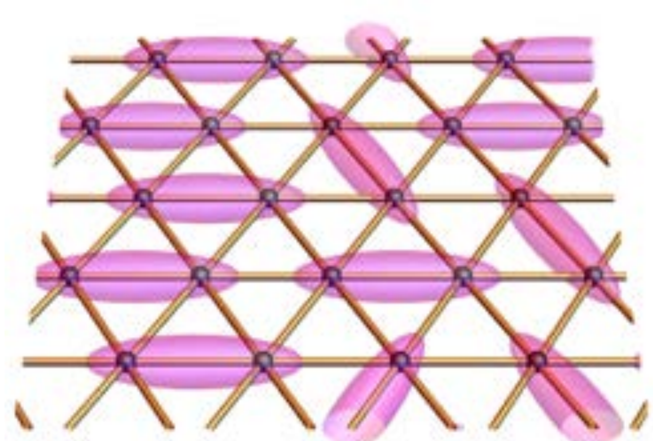
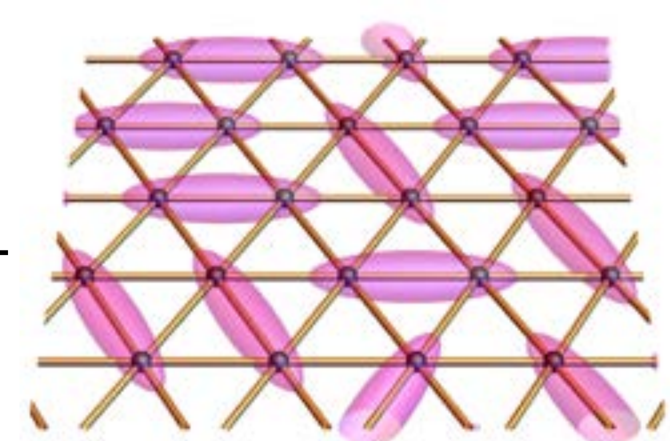
$$\mathcal{H}_{\mathrm{Heisenberg}} = J \sum_{\langle ij \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

Interaction energy $-\frac{J}{4}$

$|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$

Interaction energy $-\frac{3J}{4}$

Resonating valence bond (RVB) state: superposition of dimer coverings

$\Psi =$  $+$  $+$  $+ \ldots$

Definitions of QSLs

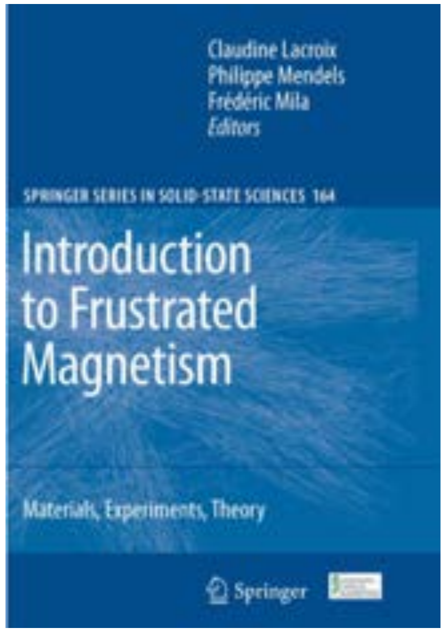
According to "Introduction to Frustrated Magnetism" (by G. Misguich)

16.2.1 Absence of Magnetic Long-Range Order (Definition 1)

Definition 1: *a quantum spin liquid is a state in which the spin–spin correlations, $\langle S_i^\alpha S_j^\beta \rangle$, decay to zero at large distances $|r_i - r_j| \to \infty$.*

16.2.2 Absence of Spontaneously Broken Symmetry (Definition 2)

Definition 2: *a quantum spin liquid is a state without any spontaneously broken symmetry.*



How to identify ?

Absence of evidence is not evidence of absence.

Positive identification

According to "Introduction to Frustrated Magnetism" (by G. Misguich)

16.2.1 Absence of Magnetic Long-Range Order (Definition 1)

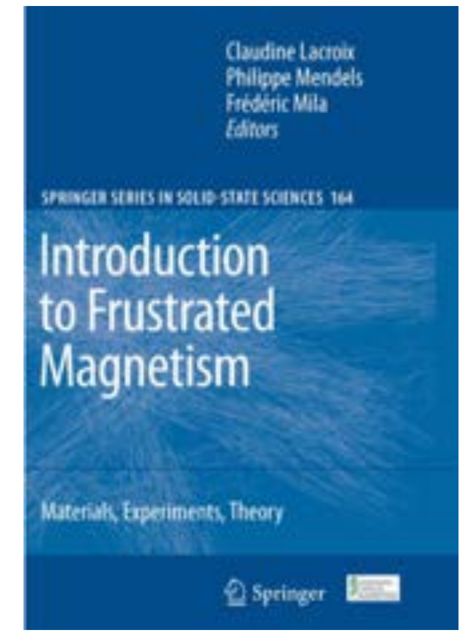
Definition 1: *a quantum spin liquid is a state in which the spin–spin correlations, $\langle S_i^\alpha S_j^\beta \rangle$, decay to zero at large distances $|r_i - r_j| \to \infty$.*

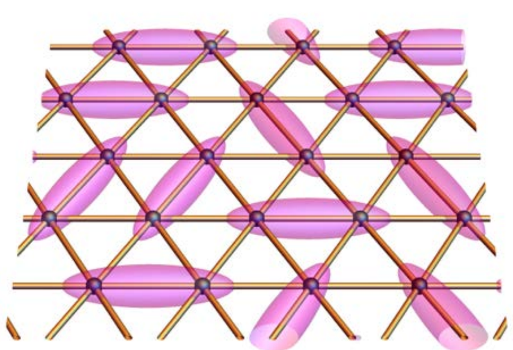
16.2.2 Absence of Spontaneously Broken Symmetry (Definition 2)

Definition 2: *a quantum spin liquid is a state without any spontaneously broken symmetry.*

16.2.3 Fractional Excitations (Definition 3)

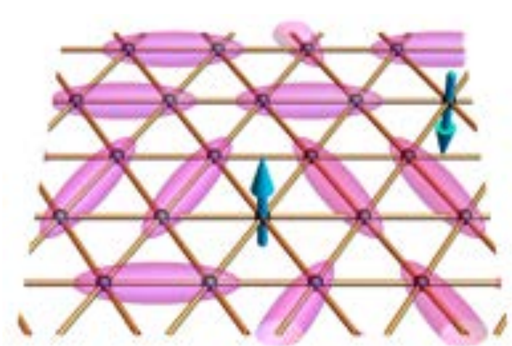
Definition 3: *a quantum spin liquid is a state with fractional excitations.*



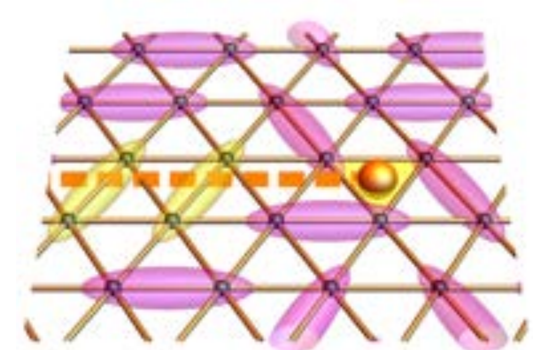


RVB ground state

Excitations in RVB



spinons



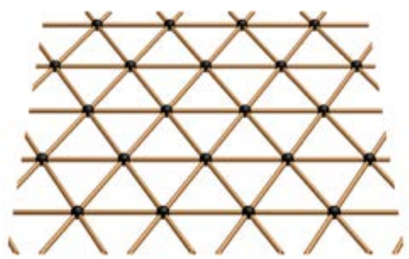
visons

Experimental works

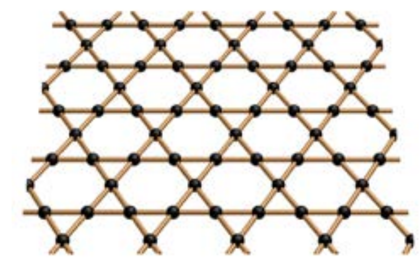
Fractional excitations : Characterization of QSLs

- Low-*T* behavior of C_v (*T-linear*)
- Transport
- Dynamical response (*continuum*)

Triangular systems

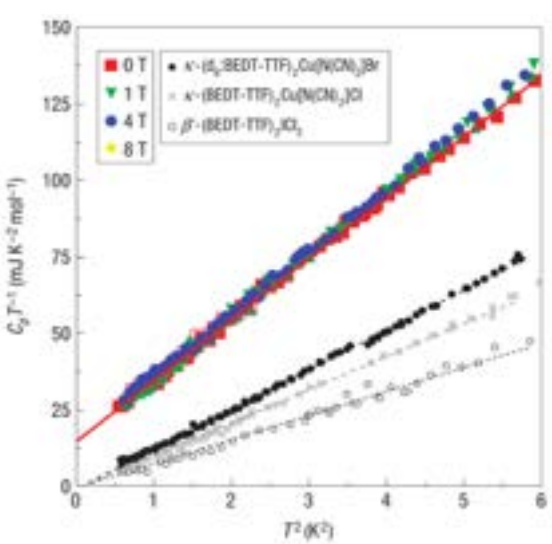


Kagomé systems



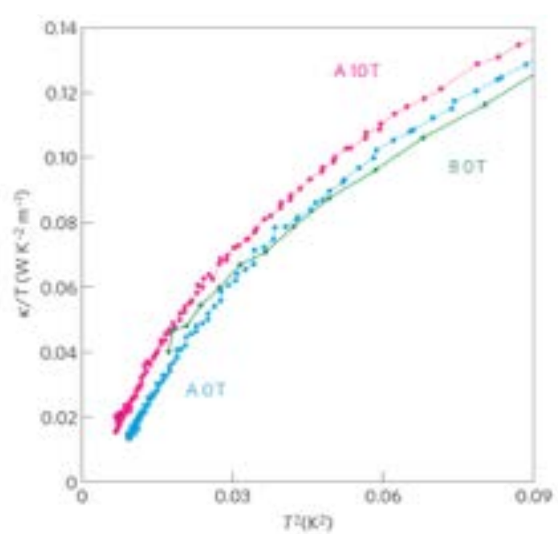
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

S. Yamashita et al., Nat. Phys. **4**, 459 (2008).



Specific heat

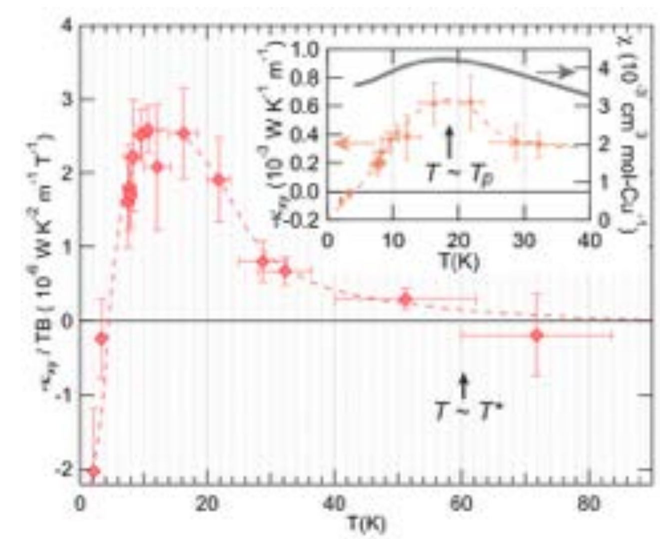
M. Yamashita et al., Nat. Phys. **5**, 44 (2009).



Thermal conductivity

volborthite

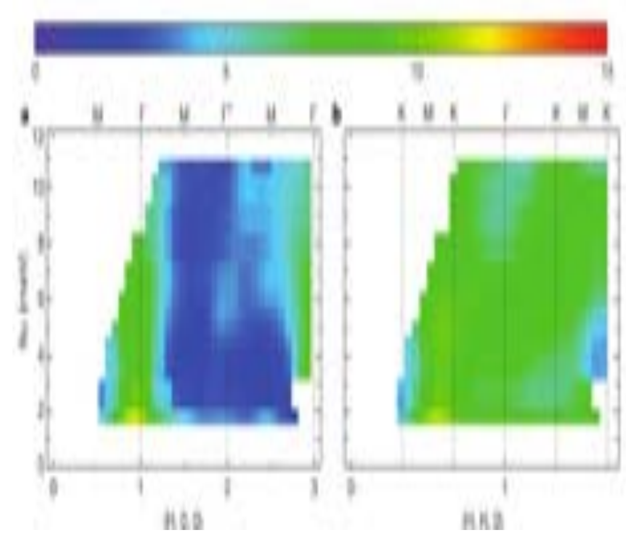
D. Watanabe *et al.*, Proc. Natl. Acad. Sci. 113, 8653 (2016).



Thermal Hall conductivity

herbertsmithite

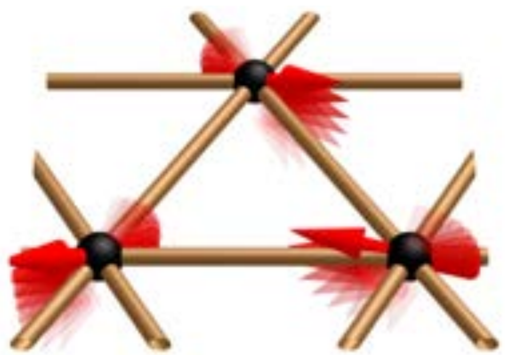
T.-H. Han *et al.*, Nature 492, 406 (2012).



Neutron scattering

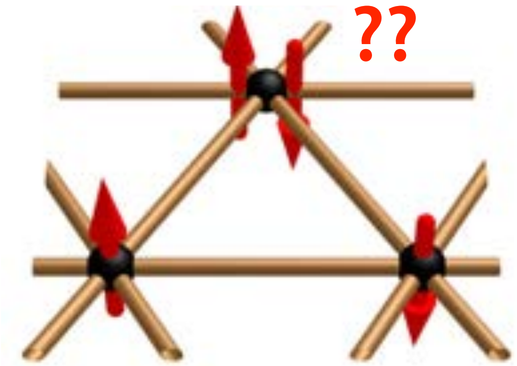
Numerical difficulty

Quantum fluctuation



\+

frustrated magnetic interactions



- Slave boson (fermion) mean-field theory
- Exact diagonalization
- Density Matrix Renormalization Group
- Variational Monte Carlo method
- Quantum Monte Carlo simulations etc.

Existence of QSL and its properties are still controversial.

<u>It is difficult to understand the properties even at *T*=0 !</u>

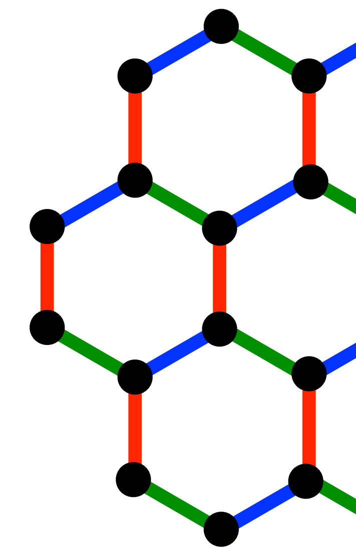
Finite temperature properties or spin dynamics
to compare experimental studies

Two breakthroughs

- Kitaev's quantum spin model — A. Kitaev, Annals of Physics **321**, 2 (2006).

Proposal of an exactly solvable model with QSL ground state

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

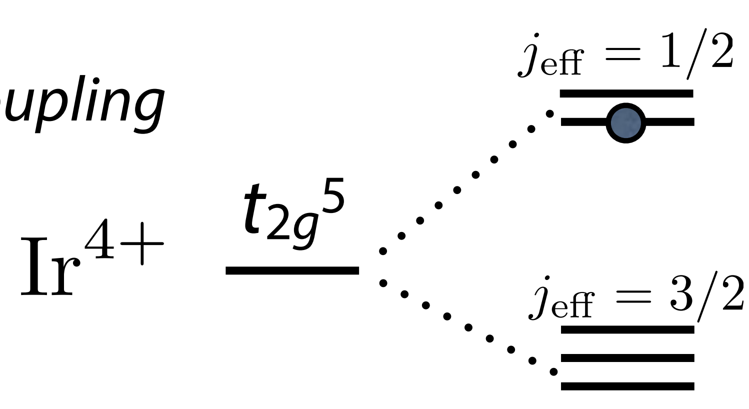


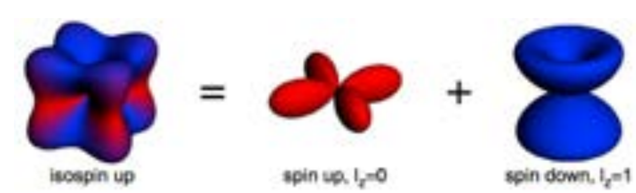
- Realization of Kitaev interaction in transition metal compounds

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

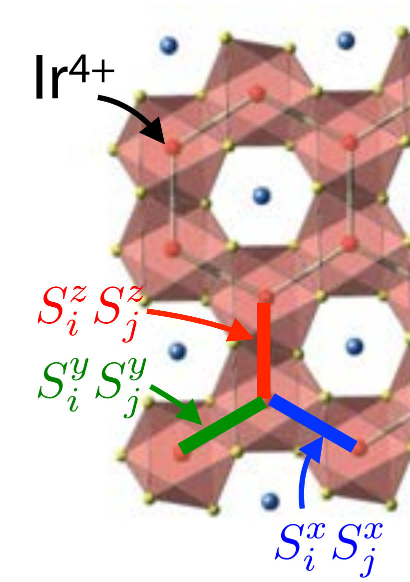
Proposal of candidate materials with the Kitaev interaction

Strong spin-orbit coupling





$j_{\rm eff}$=1/2 localized spin



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Contents

Intriguing features

- ***Simple*** interactions between *S*=1/2 spins
- ***Exact*** solvability
- ***Topological order***
- ***Fractional*** excitations: *emergent Majorana fermions* and *gauge fluxes*
- ***Quantum spin liquid***
- ***Topologically nontrivial*** Majorana fermion band (Majorana Chern insulator)
- Abelian / non-abelian anyons
- Methodological connection between spin model and fermion model
- Relevance to ***real materials***
- Relevance to ***topological quantum computation***

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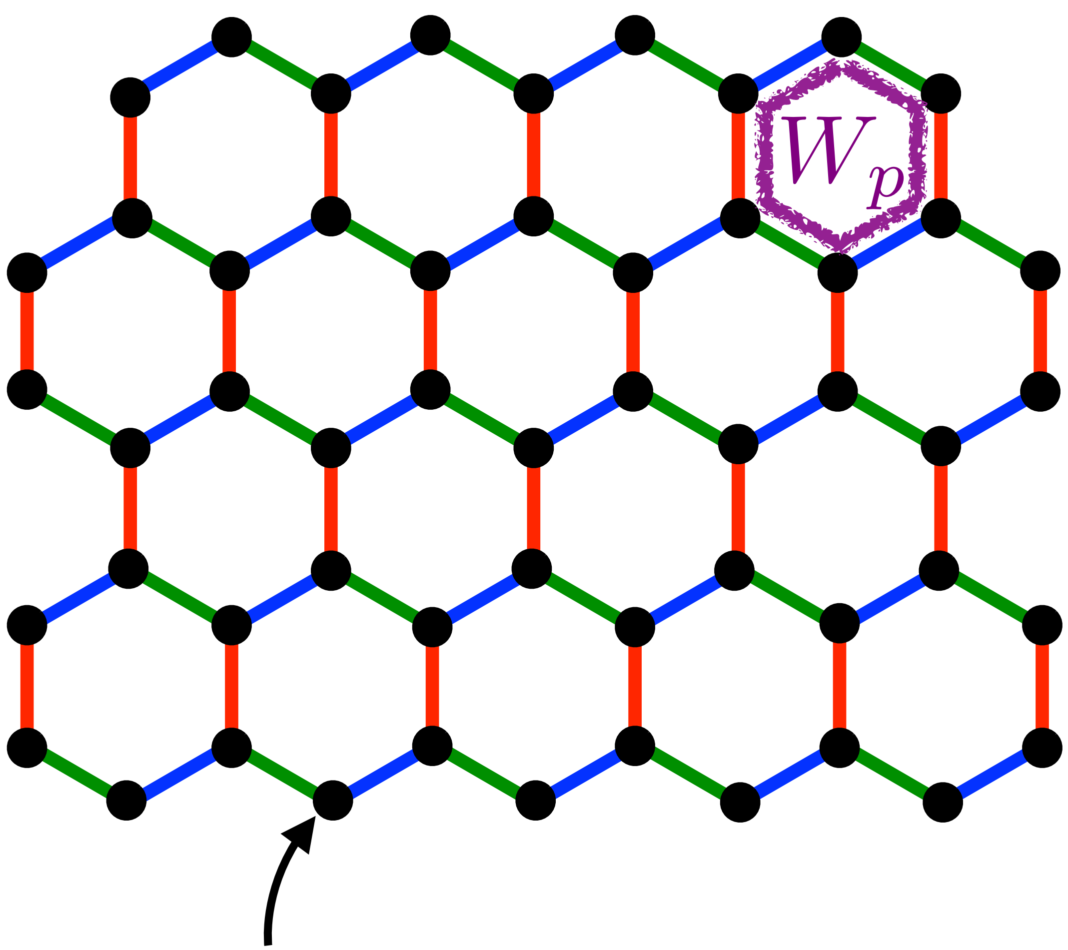
Kitaev model

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

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

Honeycomb lattice



S=1/2 spin

- Bond-dependent interaction

  ⇨ *Frustration*

  ⇨ *Novel ground state*

- Local conserved quantity W_p

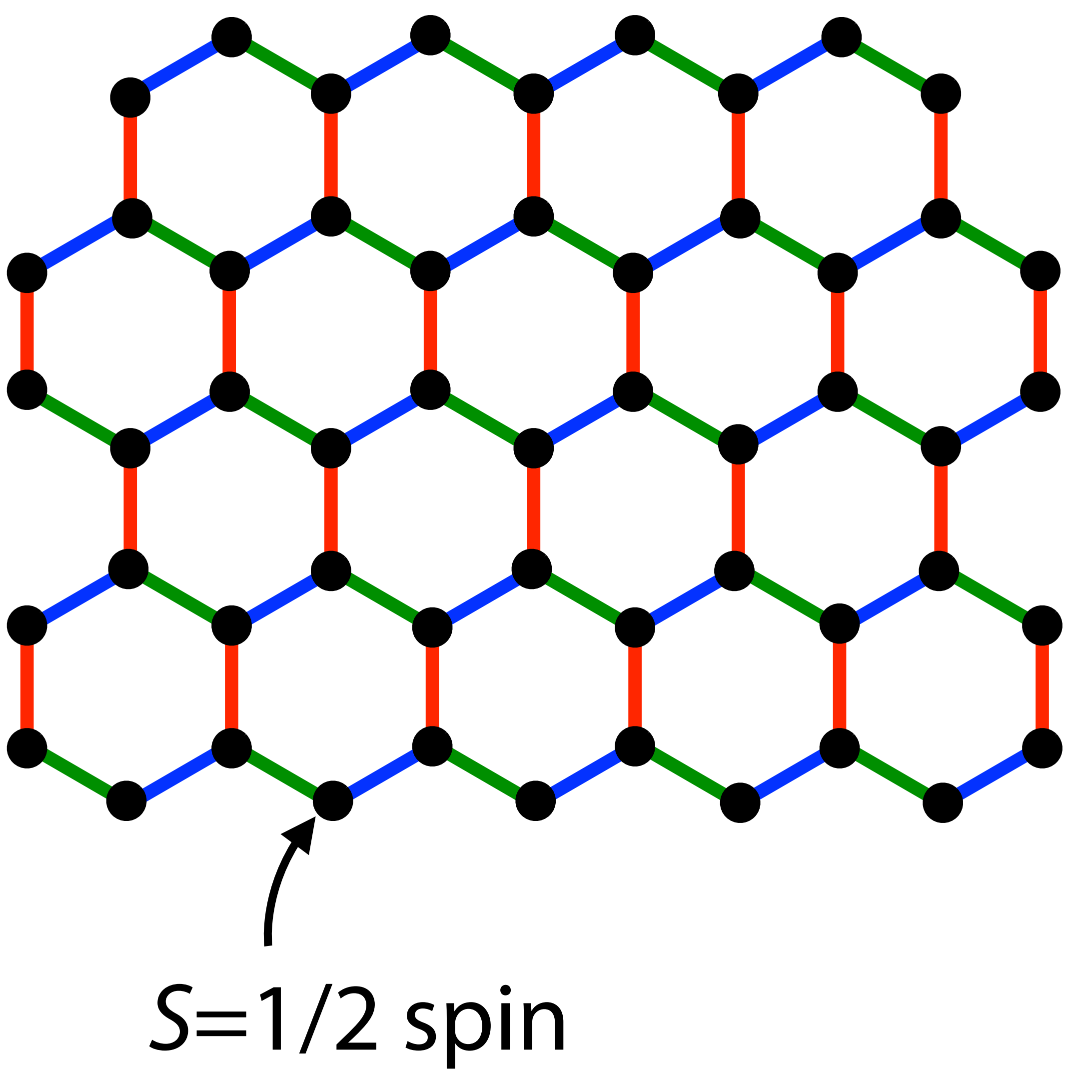
- **Ground state:**
  Quantum spin liquid (Exact solution)

Kitaev model

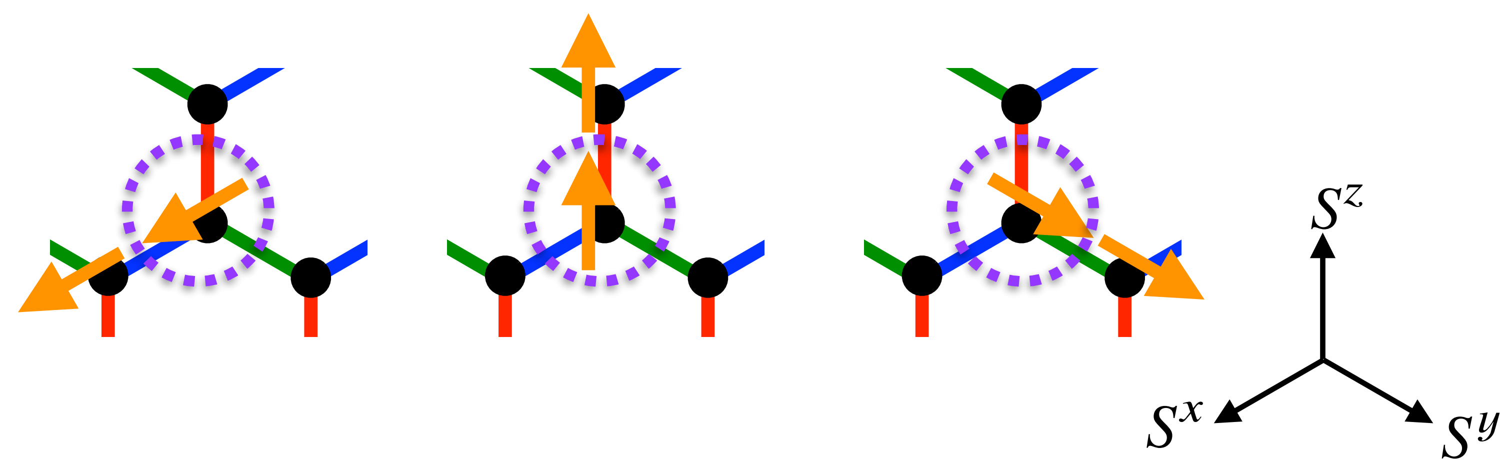
$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

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

Honeycomb lattice



- Bond-dependent interaction



All interaction energy can not be minimized simultaneously.

↓

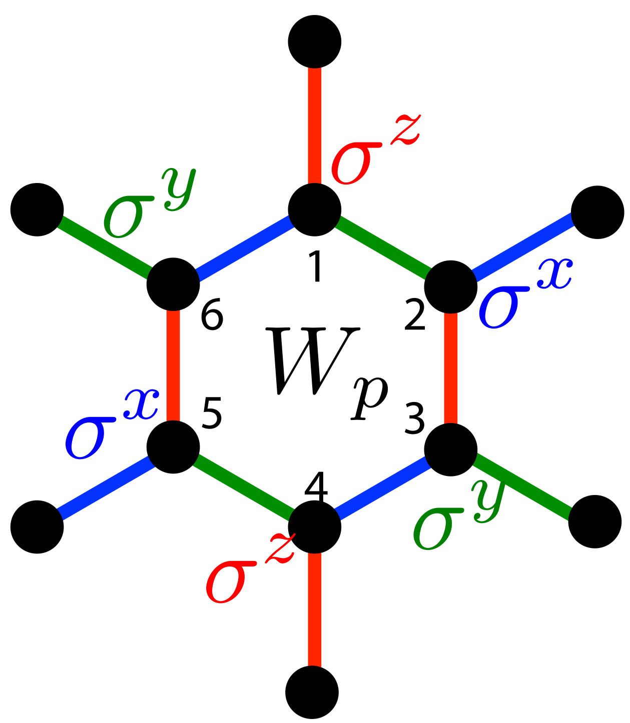
Frustration despite ferro-type interaction

Local conserved quantity

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

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

$$W_p = \sigma_1^z \sigma_2^x \sigma_3^y \sigma_4^z \sigma_5^x \sigma_6^y$$



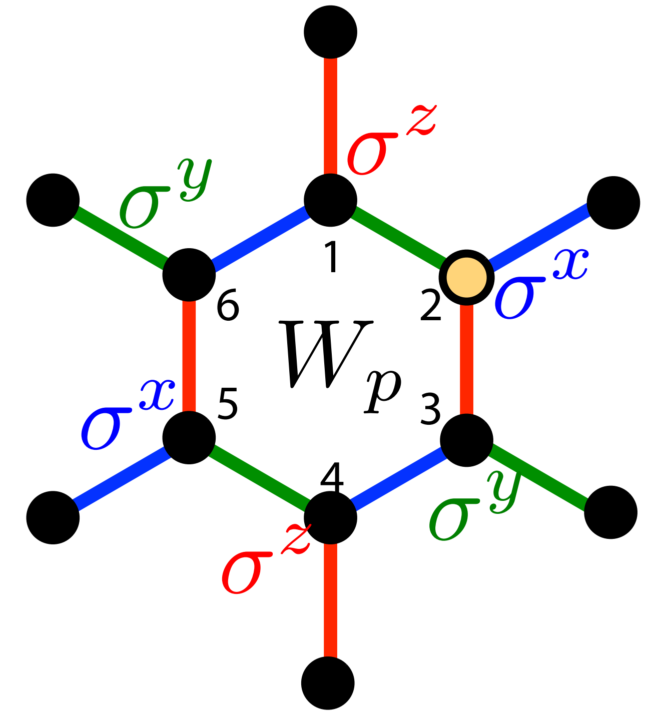
- $W_p^2 = 1$
- $[\mathcal{H}, W_p] = 0$
  since $[\sigma_1^y \sigma_2^y, W_p] = 0$
- $[W_p, W_{p'}] = 0 \qquad p \neq p'$

Eigenstates of Kitaev model are characterized by $\{W_p = \pm 1\}$

A. Kitaev, Annals of Physics **321**, 2 (2006).

Spin correlations

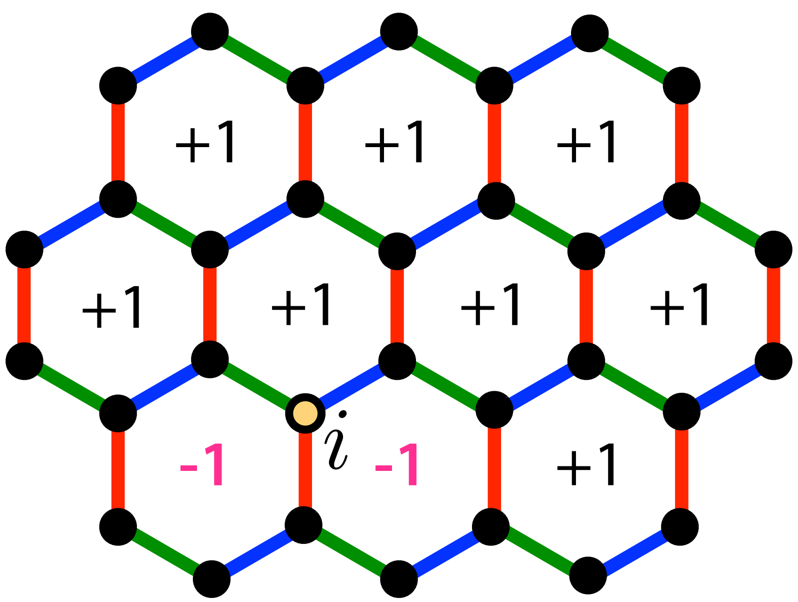
Ground state : all of W_p =+1



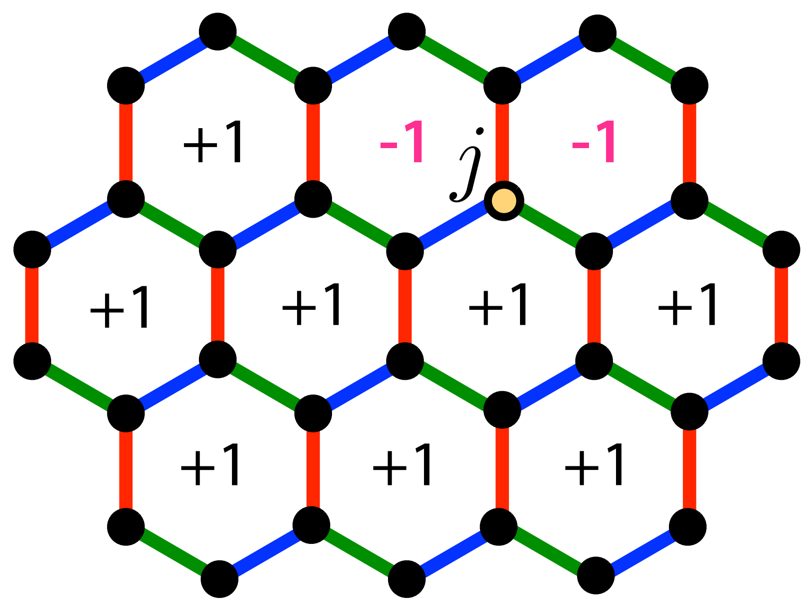
$$W_p \sigma_2^z = -\sigma_2^z W_p$$

$$\Rightarrow W_p |\sigma_2^z \Phi_0\rangle = -|\sigma_2^z \Phi_0\rangle$$

$|\sigma_i^z \Phi_0\rangle$



$|\sigma_j^z \Phi_0\rangle$



$$\langle \Phi_0 | \sigma_i^z \sigma_j^z | \Phi_0 \rangle$$
$$= \langle \sigma_i^z \Phi_0 | \sigma_j^z \Phi_0 \rangle$$
$$= 0$$

(except for NN bonds)

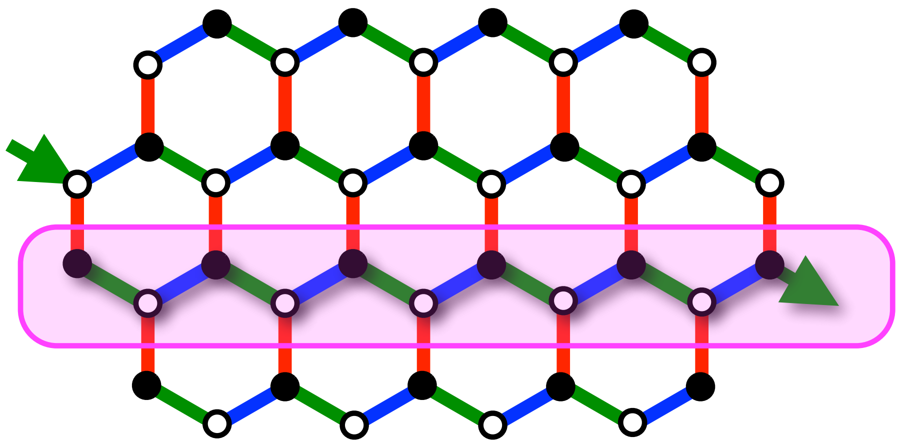
Anticommutation between spin and conserved quantity leads to the state without spin correlations. ⇒ ***Quantum spin liquid***

G. Baskaran, S. Mandal, and R. Shankar, Phys. Rev. Lett. **98**, 247201 (2007).

Jordan-Wigner transformation

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

Honeycomb lattice: a zigzag *xy* chain connected by *z*-bonds



Fermions: $a_i, \ a_i^\dagger$

Introducing *Majorana fermions*

$$c_i = a_i + a_i^\dagger$$

$$\bar{c}_i = (a_i - a_i^\dagger)/i$$

Majorana fermions: $\{c_i, c_j\} = 2\delta_{ij}$

$\rightarrow$ $c_i^2 = 1$

$[\bar{c}_i \bar{c}_j, \mathcal{H}] = 0 \qquad \eta_r^2 = 1$

$\eta_r \equiv i\bar{c}_i\bar{c}_j$: ***local conserved quantity***

Jordan-Wigner transformation

regarding the honeycomb lattice as *one open chain*

$$S_i^+ = (S_i^-)^\dagger = \prod_{i'=1}^{i-1} (1 - 2n_{i'}) a_i^\dagger \qquad S_i^z = a_i^\dagger a_i - \frac{1}{2}$$

H.-D. Chen and J. Hu, Phys. Rev. B 76, 193101 (2007).
X. Y. Feng, G.-M. Zhang, and T. Xiang, Phys. Rev. Lett. 98, 087204 (2007).
H.-D. Chen and Z. Nussinov, J. Phys. A Math. Theor. 41, 075001 (2008).

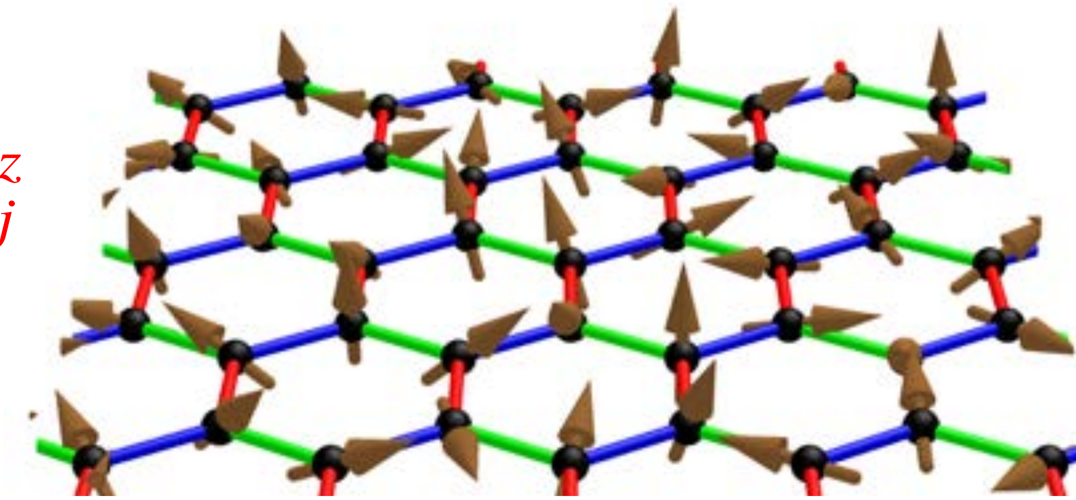
$$\mathcal{H} = \frac{iJ_x}{4} \sum_{\langle ij \rangle_x} c_i c_j - \frac{iJ_y}{4} \sum_{\langle ij \rangle_y} c_i c_j + \frac{J_z}{4} \sum_{\langle ij \rangle_z} \bar{c}_i \bar{c}_j c_i c_j$$

$$\mathcal{H} = \frac{iJ_x}{4} \sum_{\langle ij \rangle_x} c_i c_j - \frac{iJ_y}{4} \sum_{\langle ij \rangle_y} c_i c_j - \frac{iJ_z}{4} \sum_{\langle ij \rangle_z} \eta_r c_i c_j$$

Free Majorana fermion system

Quantum spin model

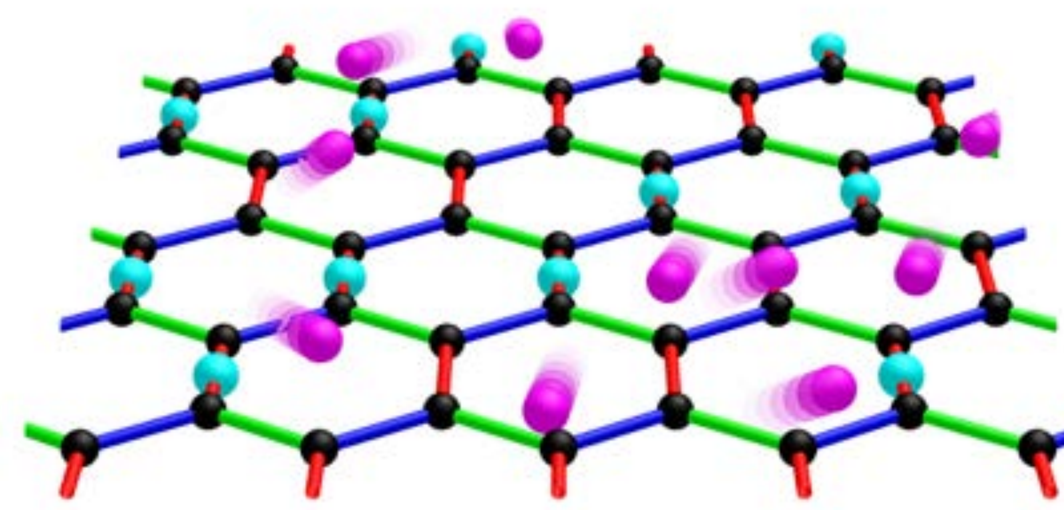
$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$



Jordan-Wigner transformation

Itinerant fermion model

$$\mathcal{H} = \frac{iJ_x}{4} \sum_{\langle ij \rangle_x} c_i c_j - \frac{iJ_y}{4} \sum_{\langle ij \rangle_y} c_i c_j - \frac{iJ_z}{4} \sum_{\langle ij \rangle_z} \eta_r c_i c_j$$



S_i →
- c_i : Itinerant Majorana
- $\bar{c}_i$: Localized Majorana

$$\eta_r = i\bar{c}_i \bar{c}_j$$

Free Majorana fermion system coupled with fluxes $W_p = \eta_r \eta_{r'}$

Ground state: all W_p=+1 → all η_r=+1

(flux-free state)

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

$$\mathcal{H} = \frac{iJ_x}{4} \sum_{\langle ij \rangle_x} c_i c_j - \frac{iJ_y}{4} \sum_{\langle ij \rangle_y} c_i c_j - \frac{iJ_z}{4} \sum_{\langle ij \rangle_z} c_i c_j$$ for flux-free state

Ground-state phase diagram

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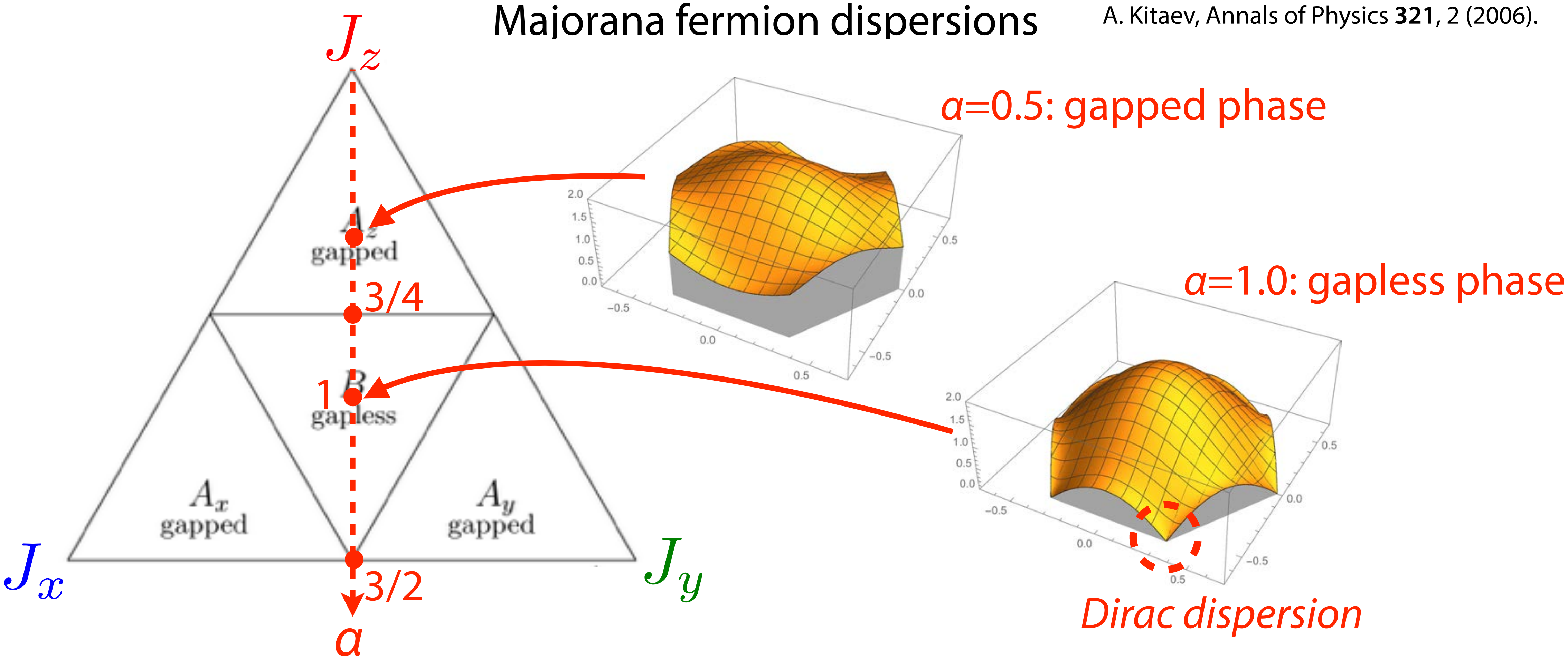
$$\mathcal{H} = \frac{iJ_x}{4}\sum_{\langle ij\rangle_x} c_i c_j - \frac{iJ_y}{4}\sum_{\langle ij\rangle_y} c_i c_j - \frac{iJ_z}{4}\sum_{\langle ij\rangle_z} c_i c_j$$ for flux-free state

$$\rightarrow \mathcal{H} = \sum_{\boldsymbol{k}} \varepsilon_{\boldsymbol{k}} \left(f_{\boldsymbol{k}}^{\dagger} f_{\boldsymbol{k}} - \frac{1}{2} \right)$$:free fermions

Majorana fermion dispersions

A. Kitaev, Annals of Physics **321**, 2 (2006).



- Phase diagram is depicted on a plane with $J_x + J_y + J_z = 1$.
- There are *gapped* and *gapless* **quantum spin liquids (QSLs)**.

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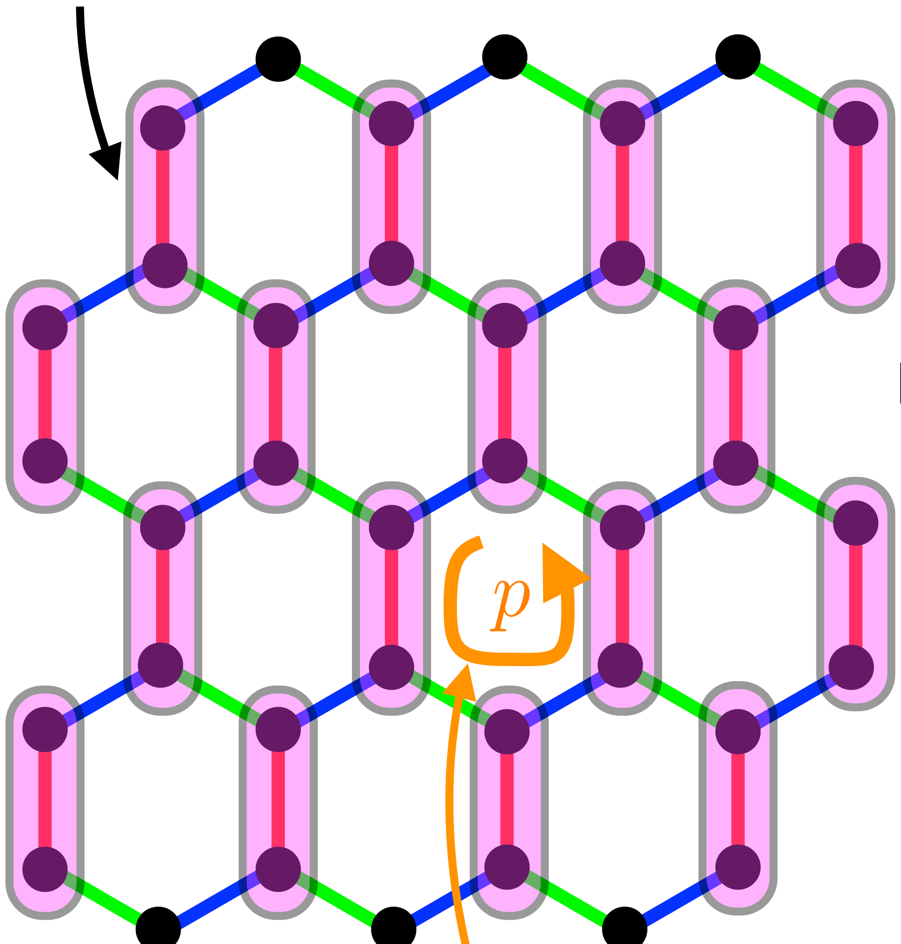
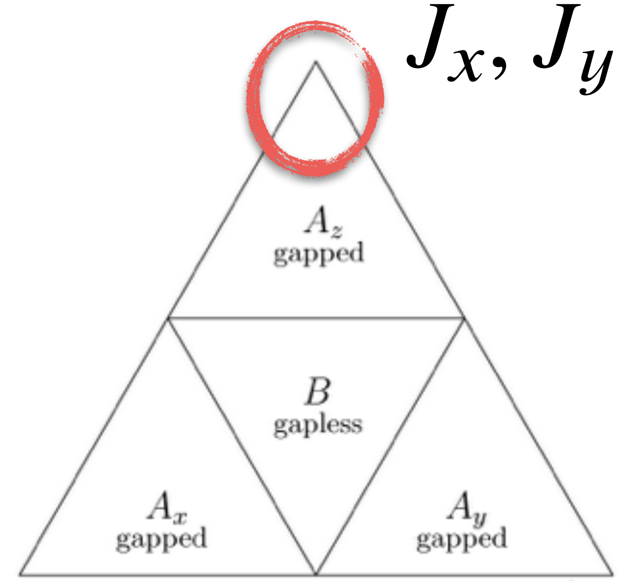
Toric code limit

$$J_x, J_y \ll J_z$$

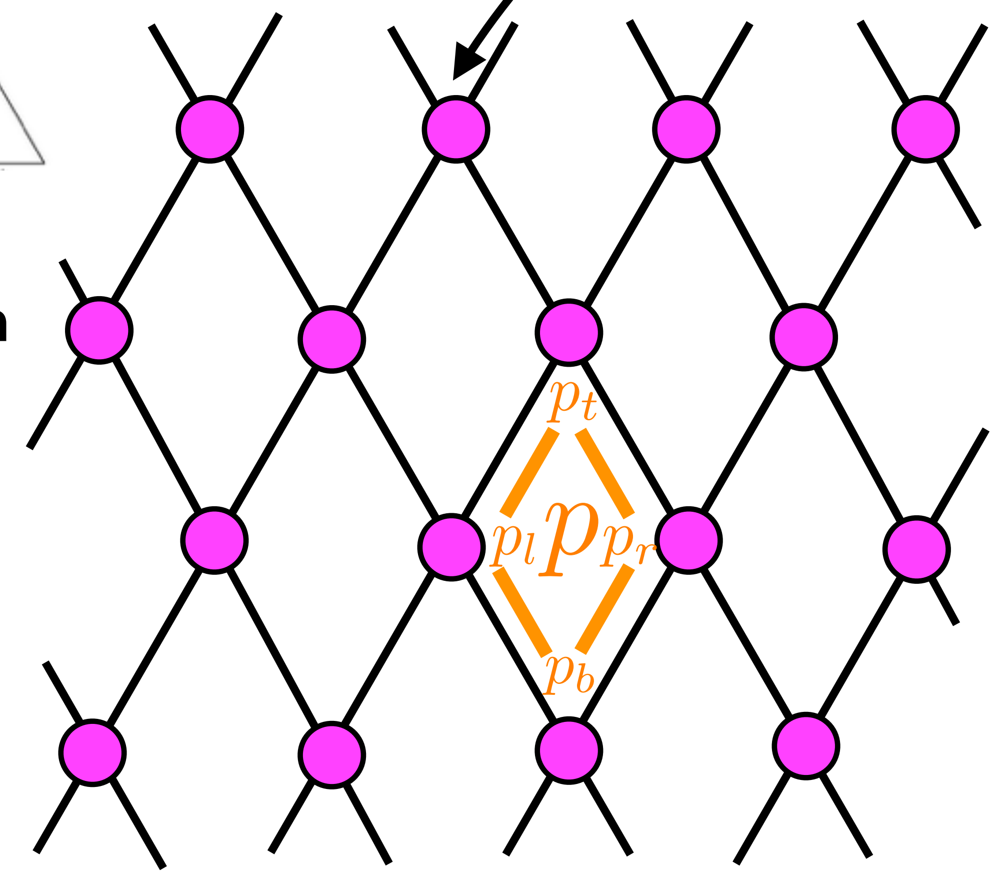
$$|\Uparrow\rangle = |\uparrow\uparrow\rangle$$
$$|\Downarrow\rangle = |\downarrow\downarrow\rangle$$





Perturbation expansion

$$\tau^z|\Uparrow\rangle = +|\Uparrow\rangle$$
$$\tau^z|\Downarrow\rangle = -|\Downarrow\rangle$$



Ring exchange-type interaction

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

perturbed term *unperturbed term*

$$\mathcal{H}_{\text{eff}} = -J_{\text{eff}} \sum_p \tau^z_{p_t} \tau^z_{p_b} \tau^y_{p_l} \tau^y_{p_r}$$

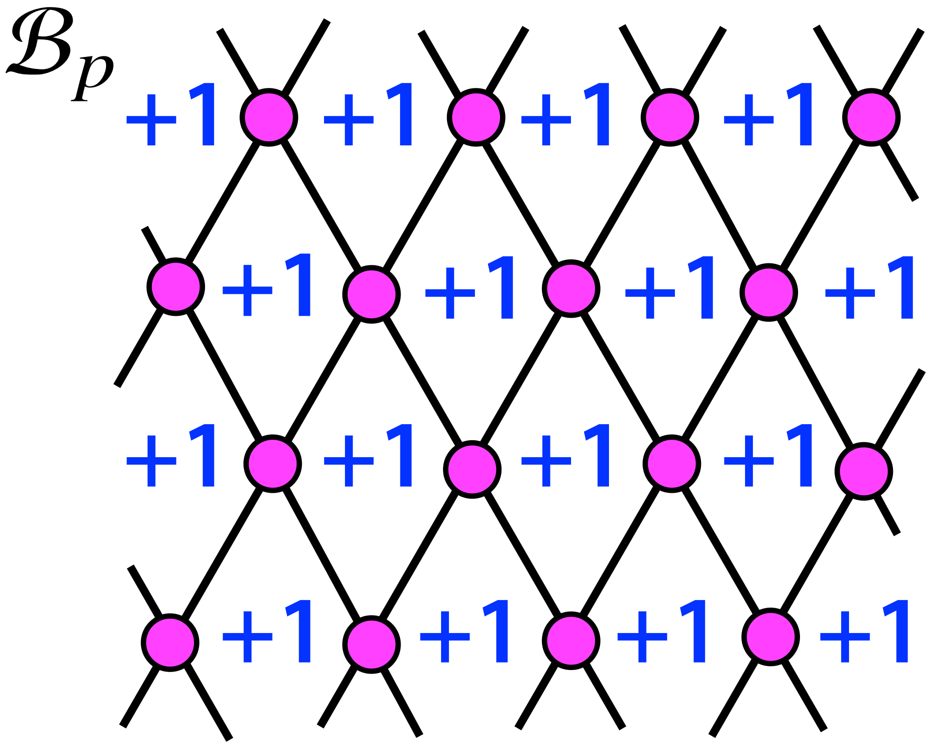
with $J_{\text{eff}} \propto J_x^2 J_y^2 / J_z^3$

Excited states in Toric code

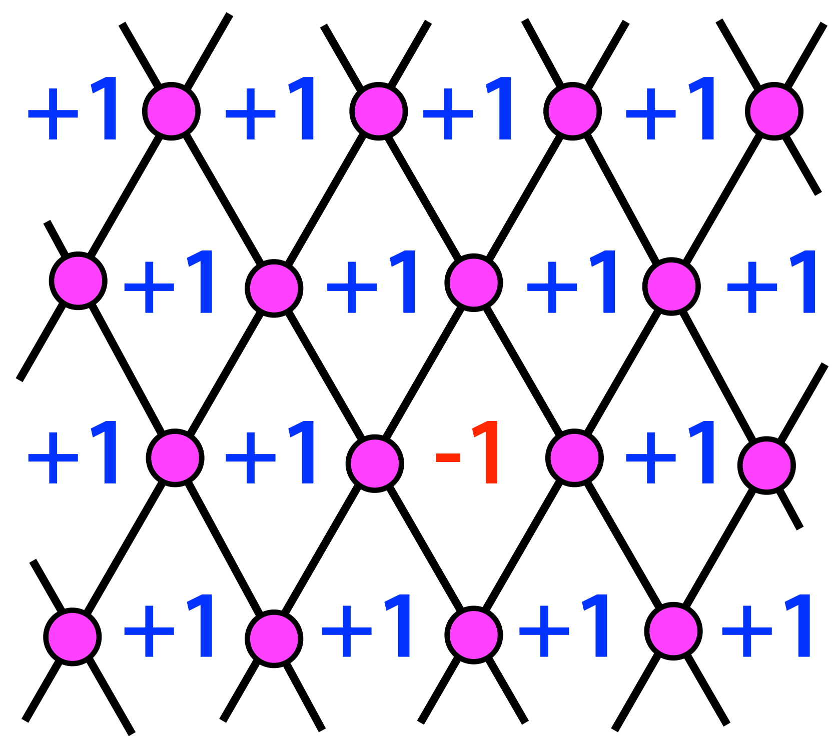
$$\mathcal{H}_{\rm eff} = -J_{\rm eff} \sum_p \mathcal{B}_p \qquad \text{with} \quad \mathcal{B}_p = \tau^z_{p_t}\tau^z_{p_b}\tau^y_{p_l}\tau^y_{p_r} = \mathcal{P}W_p\mathcal{P}$$

$$[\mathcal{H}_{\rm eff}, \mathcal{B}_p] = [\mathcal{B}_p, \mathcal{B}_{p'}] = 0 \qquad \mathcal{B}_p^2 = 1$$

Ground state



Excited state



No interactions between $\mathcal{B}_p$'s

Finite-*T* phase transition is not expected.

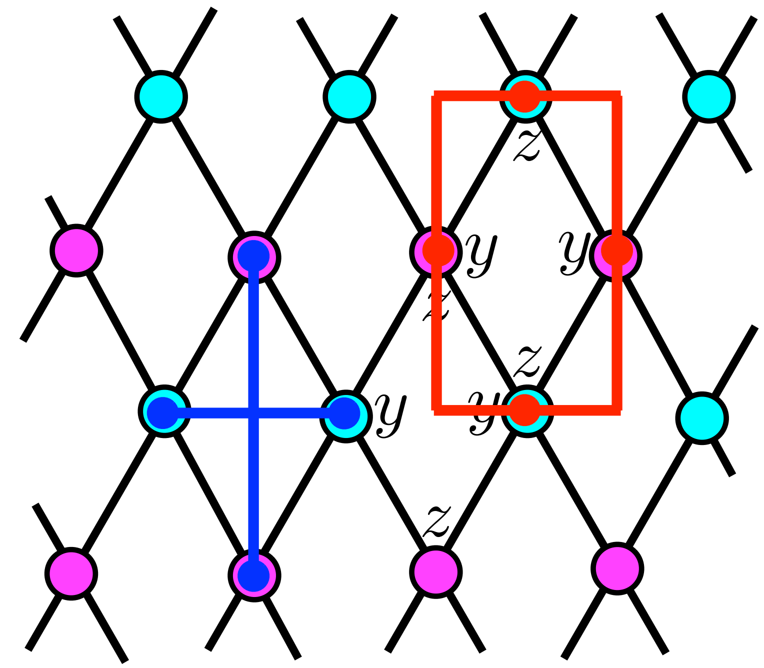
C. Castelnovo and C. Chamon, PRB76,184442(2007)

Z. Nussinov and G. Ortiz, Phys. Rev. B 77, 064302 (2008).

Another representation

$$\mathcal{H}_{\text{eff}} = -J_{\text{eff}} \sum_p \mathcal{B}_p$$

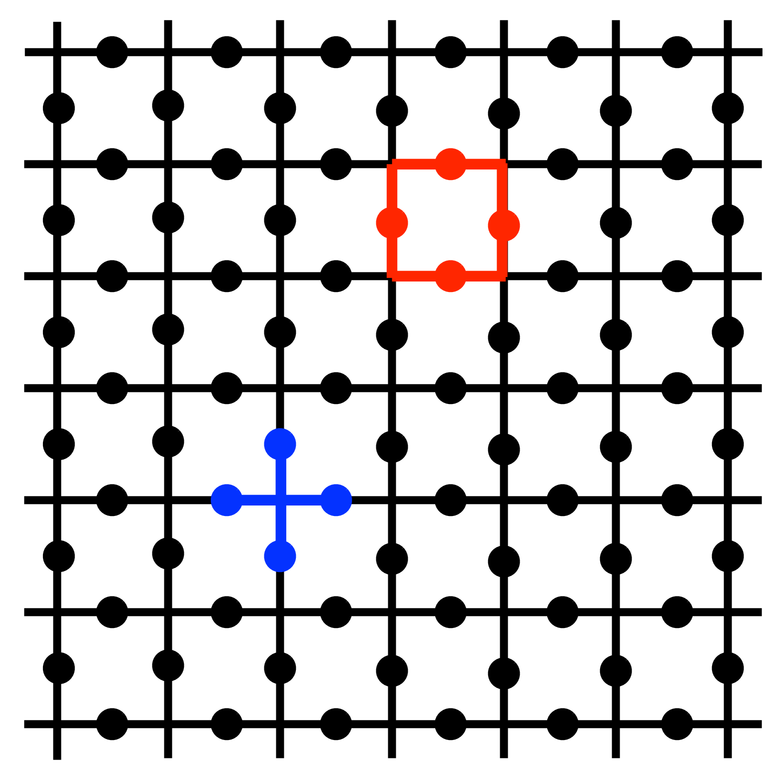
with $\mathcal{B}_p = \tau^z_{p_t} \tau^z_{p_b} \tau^y_{p_l} \tau^y_{p_r}$



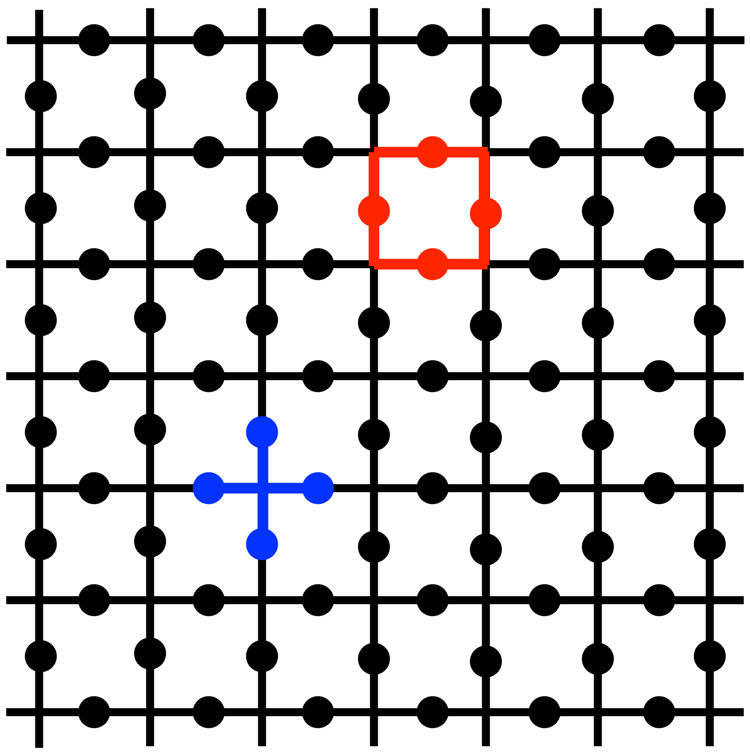
$$\mathcal{H}_{\text{toric}} = -\sum_s A_s - \sum_p B_p$$

$$A_s = \sigma^x_i \sigma^x_j \sigma^x_k \sigma^x_l$$

$$B_p = \sigma^z_i \sigma^z_j \sigma^z_k \sigma^z_l$$



Conserved quantities in Toric code



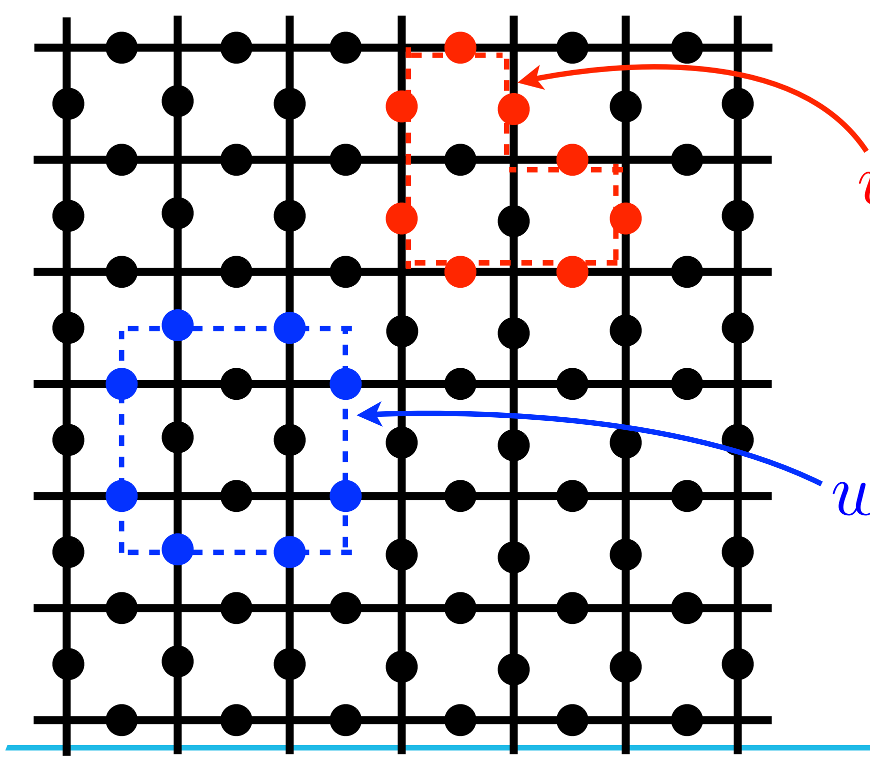
$$\mathcal{H}_{\rm toric} = -\sum_s A_s - \sum_p B_p$$

$$A_s = \sigma_i^x \sigma_j^x \sigma_k^x \sigma_l^x \qquad B_p = \sigma_i^z \sigma_j^z \sigma_k^z \sigma_l^z$$

$$[\mathcal{H}_{\rm toric}, A_s] = [\mathcal{H}_{\rm toric}, B_p] = 0$$

The ground state: $A_s = +1$

$B_p = +1$



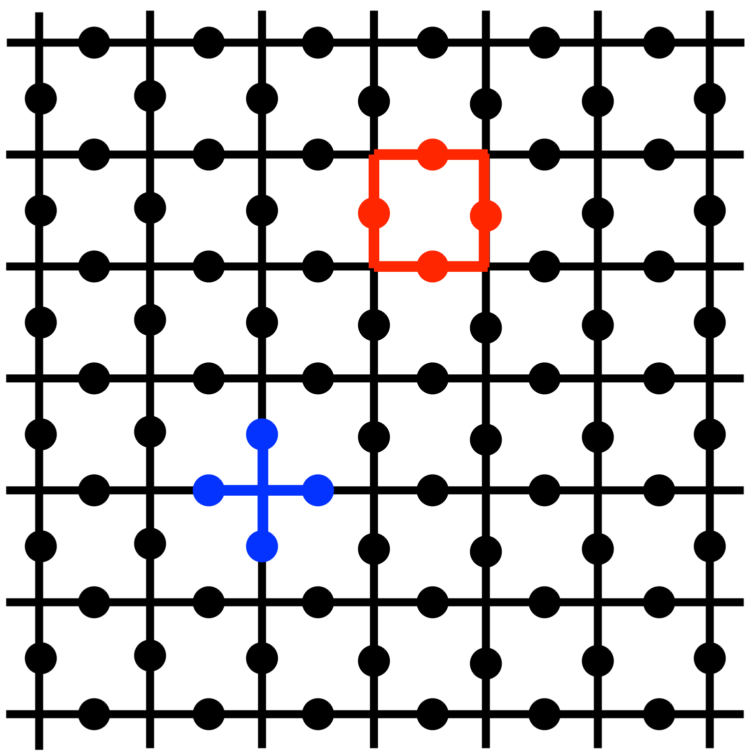
$$w_C^z = \prod_{i \in C} \sigma_i^z$$

$$w_C^x = \prod_{i \in C} \sigma_i^x$$

$$[\mathcal{H}_{\rm toric}, w_C^x] = [\mathcal{H}_{\rm toric}, w_{C'}^z] = 0$$

The Hamiltonian commutes with loop operators w^x, w^z

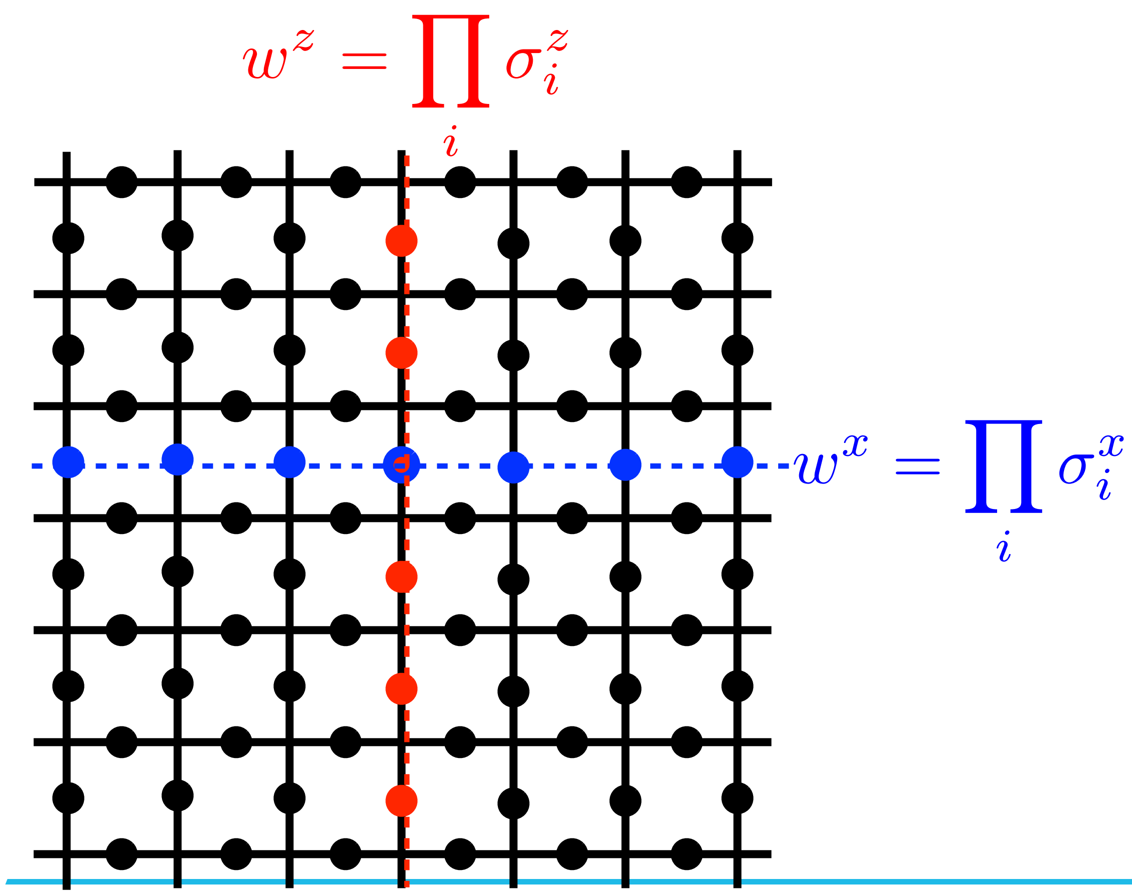
Topological degeneracy in Toric code



$$\mathcal{H}_{\rm toric} = -\sum_s A_s - \sum_p B_p$$

$$A_s = \sigma_i^x \sigma_j^x \sigma_k^x \sigma_l^x \qquad B_p = \sigma_i^z \sigma_j^z \sigma_k^z \sigma_l^z$$

$$[\mathcal{H}_{\rm toric}, A_s] = [\mathcal{H}_{\rm toric}, B_p] = 0$$



$$[\mathcal{H}_{\rm toric}, w^x] = [\mathcal{H}_{\rm toric}, w^z] = 0$$

$$w^x w^z = -w^z w^x$$

$|\Phi\rangle$: eigenstate of w_z with eigenvalue +1

and eigenstate of *H* with eigenvalue *E*

$$w^z w^x |\Phi\rangle = -w^x w^z |\Phi\rangle = -w^x |\Phi\rangle$$

$$\mathcal{H}_{\rm toric} w^x |\Phi\rangle = w^x E |\Phi\rangle$$

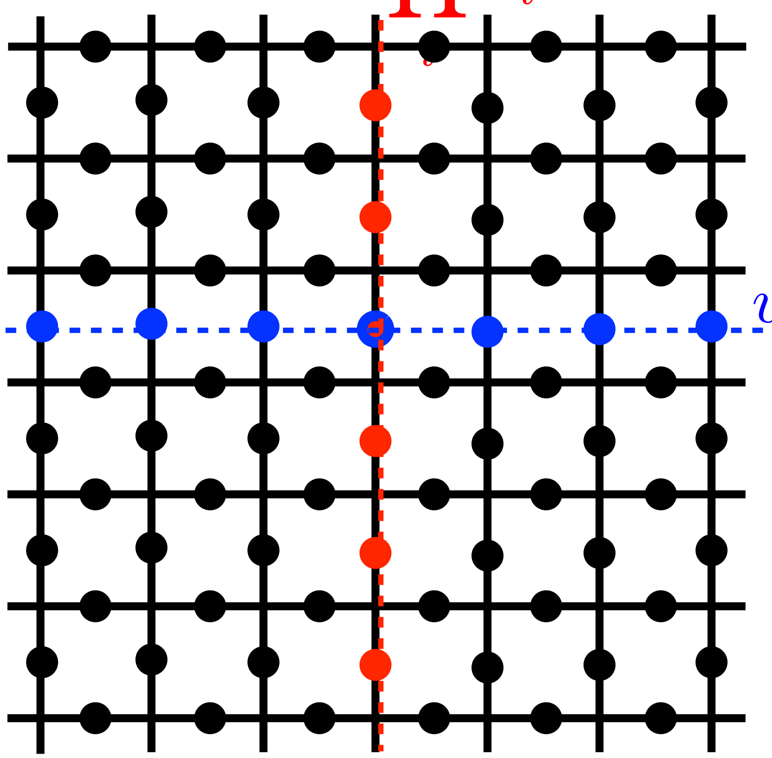
$w^x|\Phi\rangle$: eigenstate of w_z with eigenvalue -1

and eigenstate of *H* with eigenvalue *E*

topologically protected degeneracy

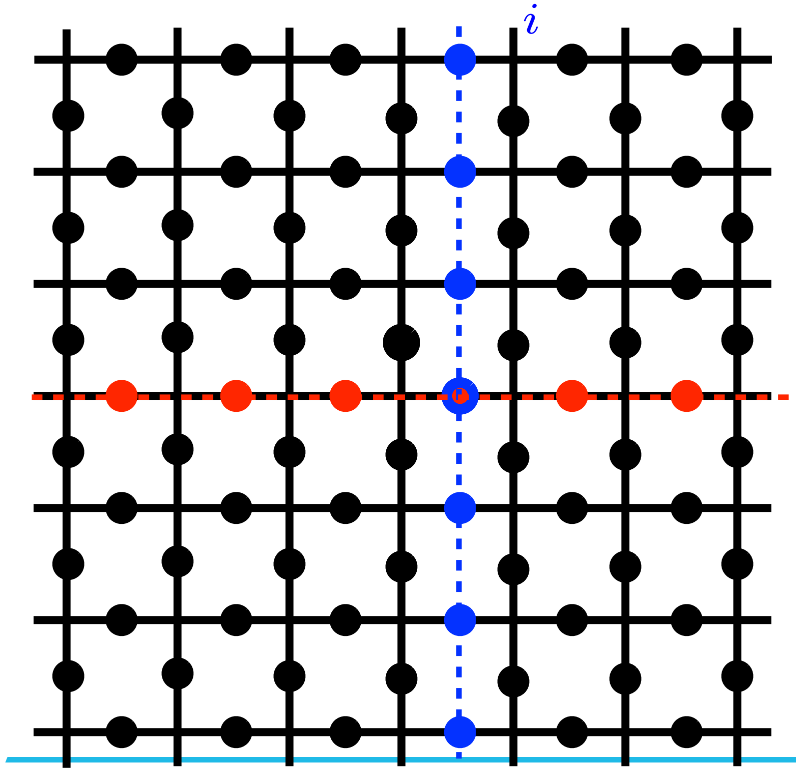
Topological degeneracy in Toric code

$$w^z = \prod \sigma_i^z$$



$$w^x = \prod_i \sigma_i^x$$

$$\tilde{w}^x = \prod_i \sigma_i^x$$



$$\tilde{w}^z = \prod_i \sigma_i^z$$

$$w^x w^z = -w^z w^x$$

$$\tilde{w}^x \tilde{w}^z = -\tilde{w}^z \tilde{w}^x$$

$$[w^z, \tilde{w}^z] = 0$$

	eigenvalue of w_z	eigenvalue of $\tilde{w}_z$
$\lvert\Phi\rangle$	+1	+1
$w^x\lvert\Phi\rangle$	-1	+1
$\tilde{w}^x\lvert\Phi\rangle$	+1	-1
$w^x\tilde{w}^x\lvert\Phi\rangle$	-1	-1

Four-fold topologically protected degeneracy

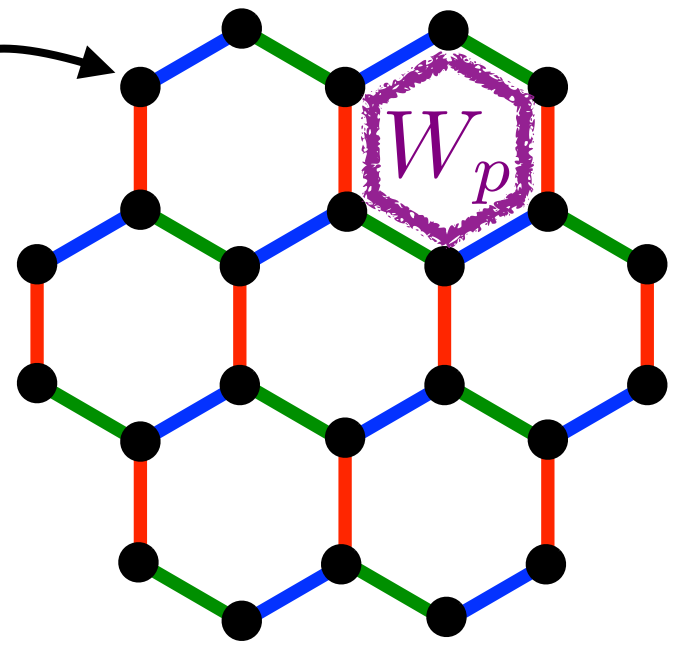
The simplest model with topological order

Kitaev model

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

A. Kitaev, Annals of Physics **321**, 2 (2006).

S=1/2 spin



☑ Bond-dependent interactions

→ ***frustration***

☑ Z_2 flux (conserved quantity) W_p on each plaquette

→ ground state: ***quantum spin liquid***

(Only NN interactions are finite)

☑ Free fermion system coupled with Z_2 variables

$$\mathcal{H} = \frac{iJ_x}{4} \sum_{\langle ij \rangle_x} c_i c_j - \frac{iJ_y}{4} \sum_{\langle ij \rangle_y} c_i c_j - \frac{iJ_z}{4} \sum_{\langle ij \rangle_z} \eta_r c_i c_j \qquad \eta_r = \pm 1$$

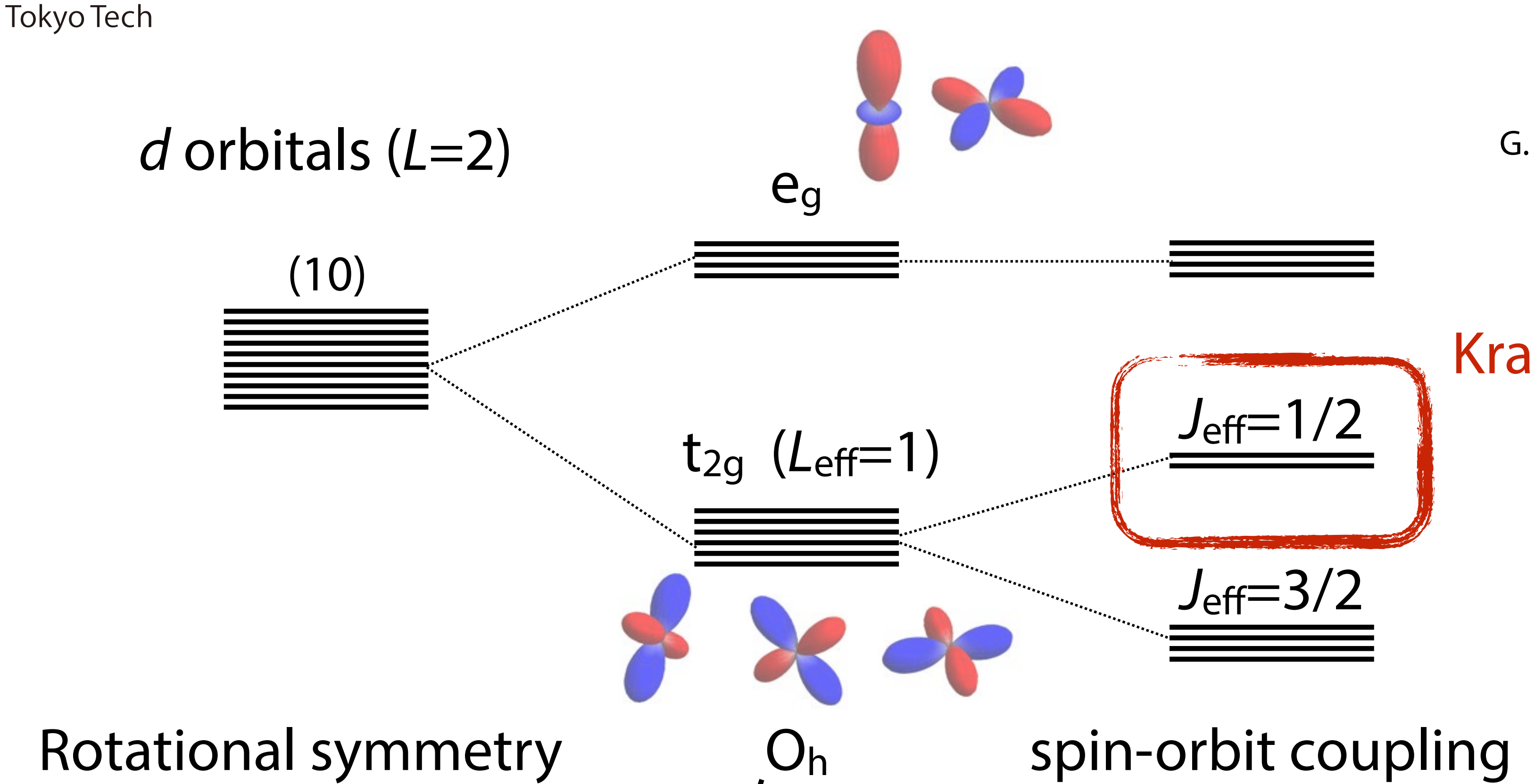
S_i → c_i : **Itinerant Majorana**

S_i → $\bar{c}_i$: **Localized Majorana**

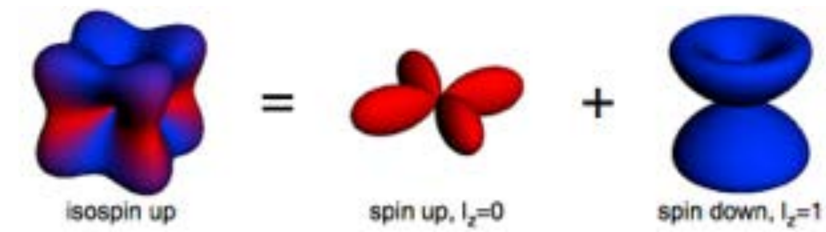
$$\eta_r = i\bar{c}_i\bar{c}_j$$

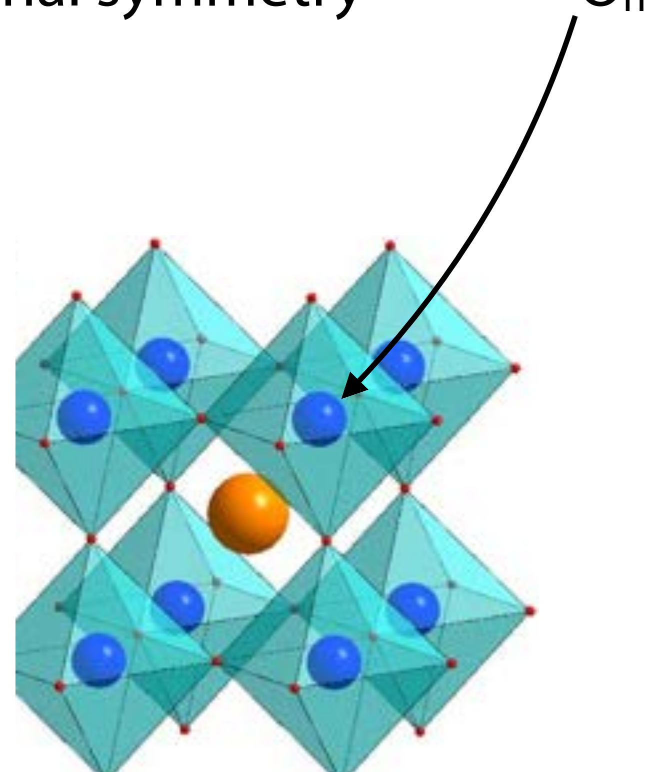
Relevance to real materials

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

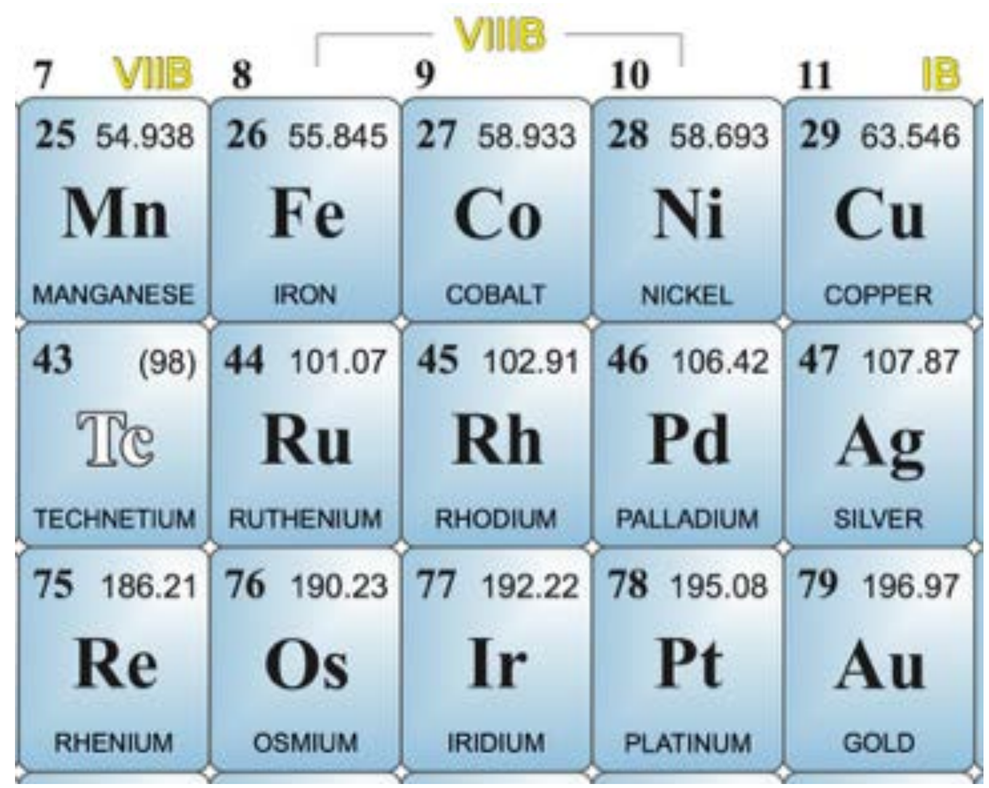


Kramers doublet





Perovskite-type structure

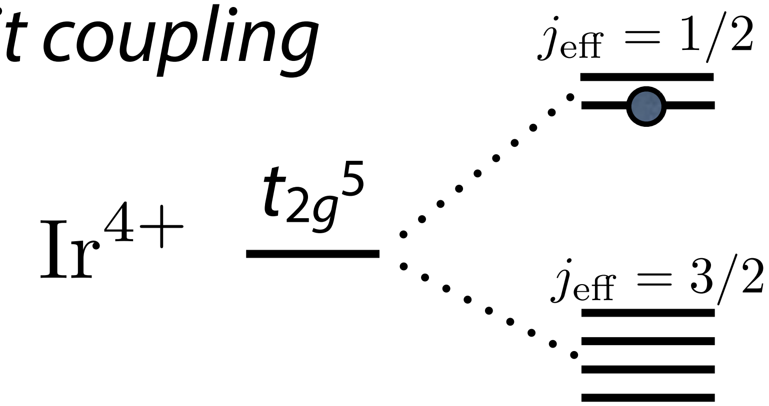


7 VIIB	8 VIIIB	9 VIIIB	10 VIIIB	11 IB
25 54.938 Mn MANGANESE	26 55.845 Fe IRON	27 58.933 Co COBALT	28 58.693 Ni NICKEL	29 63.546 Cu COPPER
43 (98) Tc TECHNETIUM	44 101.07 Ru RUTHENIUM	45 102.91 Rh RHODIUM	46 106.42 Pd PALLADIUM	47 107.87 Ag SILVER
75 186.21 Re RHENIUM	76 190.23 Os OSMIUM	77 192.22 Ir IRIDIUM	78 195.08 Pt PLATINUM	79 196.97 Au GOLD

Strong spin-orbit coupling

Realization of Kitaev QSLs

Strong spin-orbit coupling

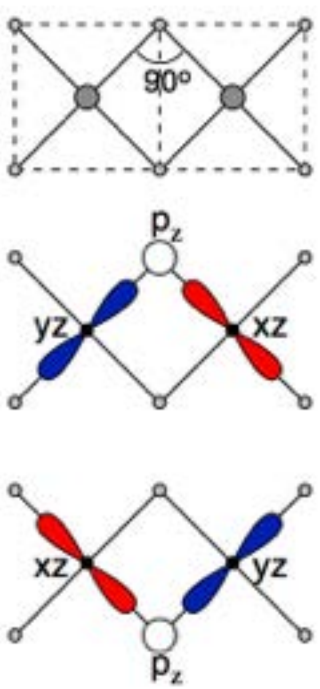


$j_{\rm eff}$=1/2 localized spin

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

Superexchange interaction

in ***edge sharing case***



dpd hopping on a *xy* plane

$-JS_i^zS_j^z$: *Ising interaction on xy plane*

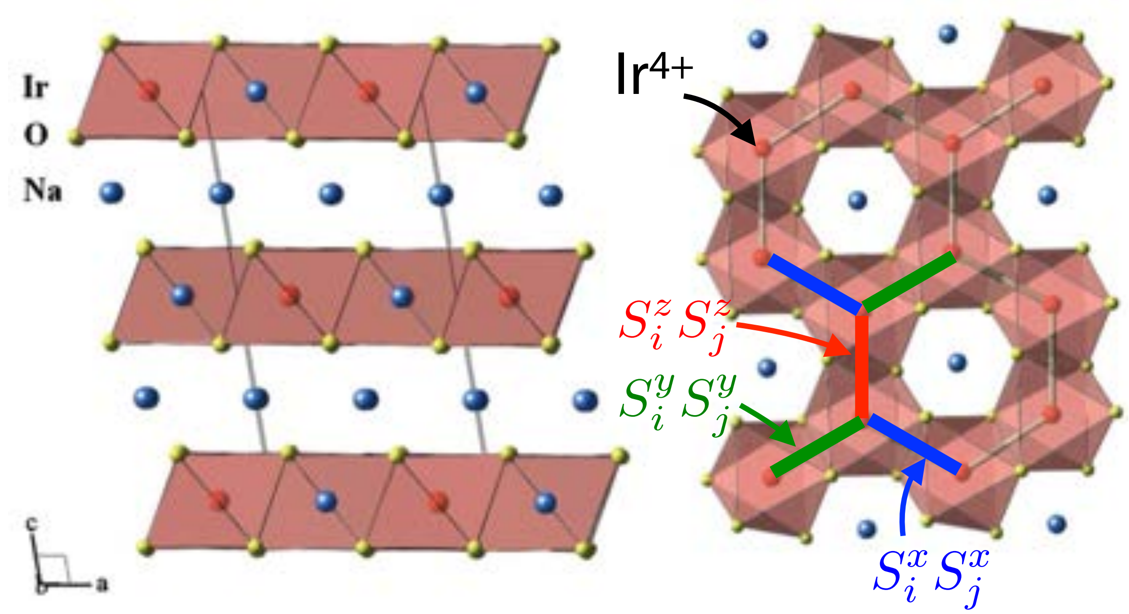
$-JS_i^xS_j^x$ *on yz plane*

$-JS_i^yS_j^y$ *on zx plane*

bond-dependent interaction due to orbital anisotropy

Iridate A_2IrO_3 (A=Li,Na)

Ruthenate α-$RuCl_3$



Realization of Kitaev QSLs

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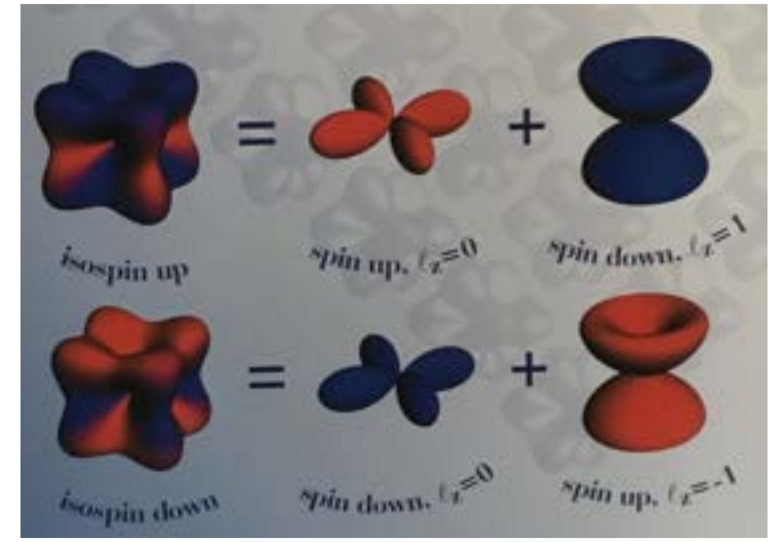
- Superexchange between $j_{\mathrm{eff}}=1/2$ spins
- Edge sharing of MO_6 octahedra

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

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z + J_{\mathrm{H}} \sum_{<ij>} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$



Taken in MPI Guest House

		Magnetic order	Heisenberg interaction	Kitaev interaction
☑ A_2IrO_3 (A=Li,Na)	Ir^{4+} $5d^5$	T_c~13K	~50K	~350K
☑ α-$RuCl_3$	Ru^{3+} $4d^5$	T_c~14K	~30K	~100K

A_2IrO_3:

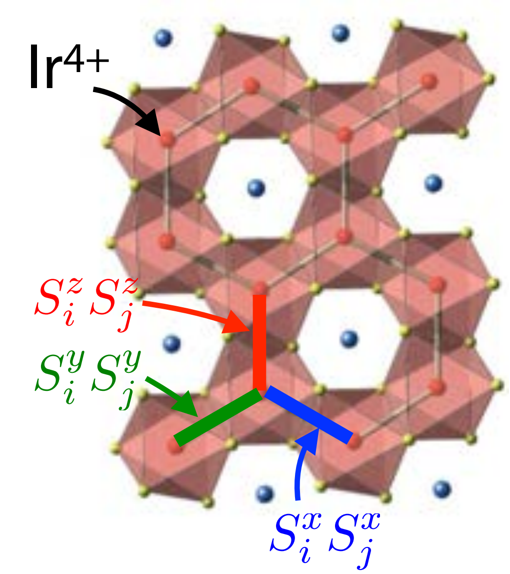
Y. Singh and P. Gegenwart, Phys. Rev. B **82**, 064412 (2010).
Y. Singh et. al., Phys. Rev. Lett. **108**, 127203 (2012).
R. Comin et. al., Phys. Rev. Lett. **109**, 266406 (2012).
S. K. Choi et. al., Phys. Rev. Lett. **108**, 127204 (2012).

Y. Yamaji et al., Phys. Rev. Lett. **113**, 107201 (2014).
K. Foyevtsova et al., Phys. Rev. B **88**, 035107 (2013).

α-$RuCl_3$:

K. W. Plumb et al., Phys. Rev. B. **90**, 041112 (2014).
Y. Kubota et al., Phys. Rev. B **91**, 094422 (2015).
L. J. Sandilands et al., Phys. Rev. Lett. **114**, 147201 (2015).
J. A. Sears, M. Songvilay et al., Phys. Rev. B **91**, 144420 (2015).
M. Majumder et al., Phys. Rev. B **91**, 180401(R) (2015).

A. Banerjee et al., Nat. Mater., nmat4604 (2016).

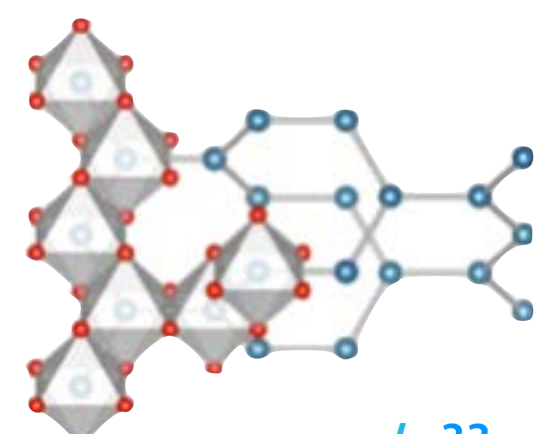


☑ β-Li_2IrO_3 hyperhoneycomb lattice T_c~38K

T. Takayama et al., Phys. Rev. Lett. **114**, 077202 (2015).
K. A. Modic et al., Nat. Commun. **5**, 4203 (2014).

(Heisenberg)/(Kitaev)~0.1-0.2

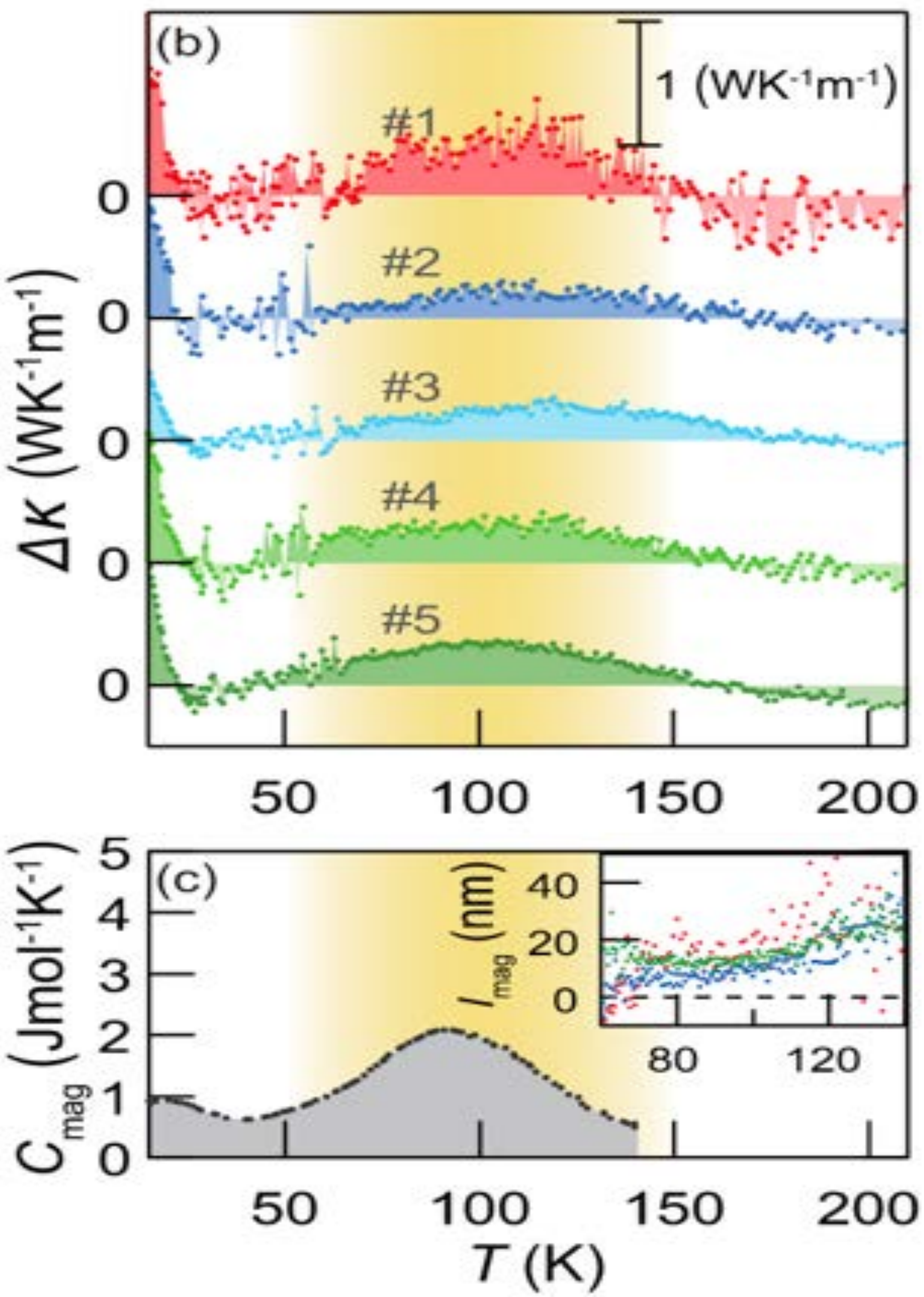
H.S. Kim et al., EPL **112**, 67004 (2016).



Kitaev term is dominant.

Dynamical response in α-$RuCl_3$

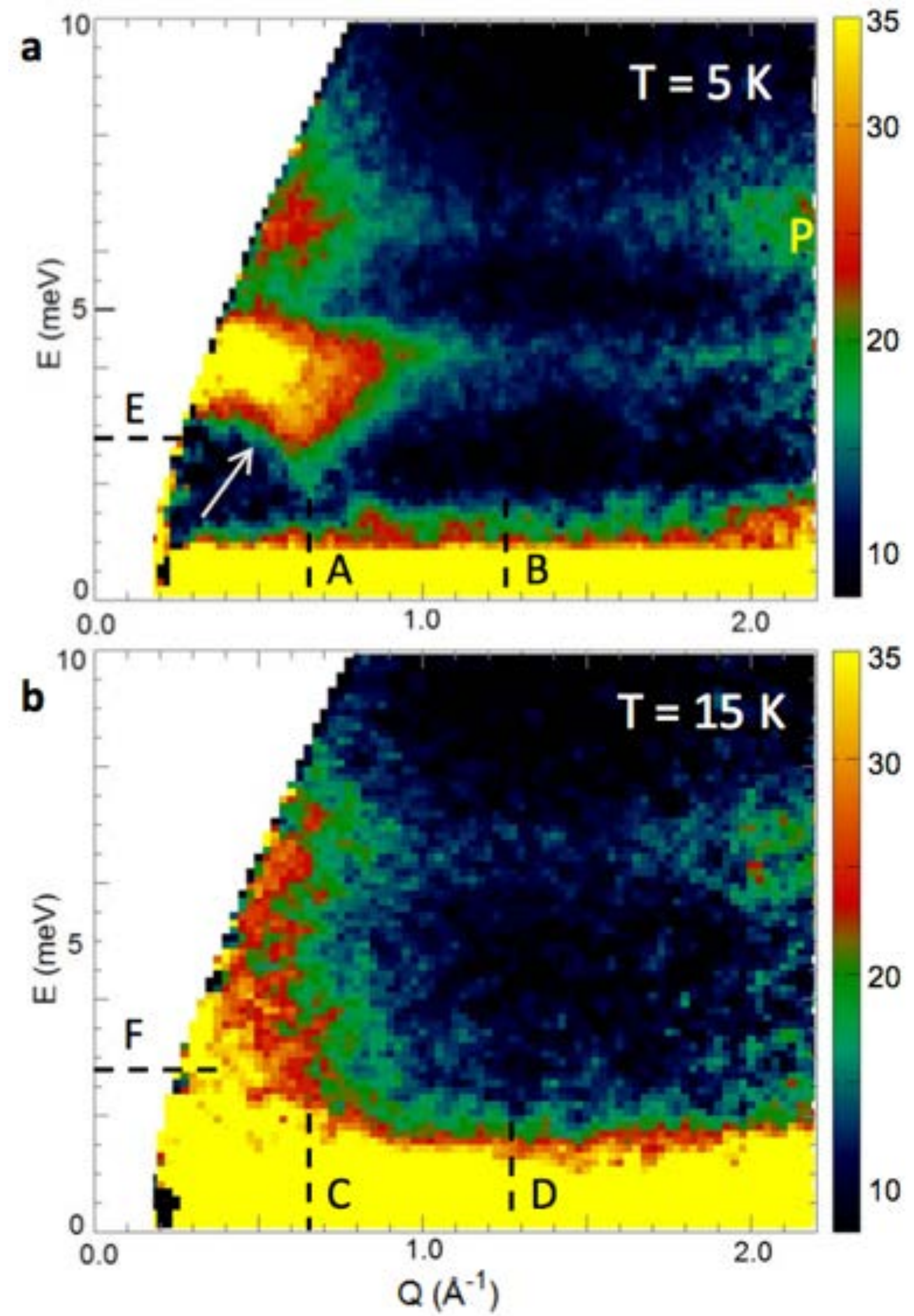
- Longitudinal thermal conductivity κ



D. Hirobe, M. Sato, Y. Shiomi, H. Tanaka, and E. Saitoh, Phys. Rev. B **95**, 241112 (2017).

- Inelastic neutron scattering

A. Banerjee et al., Nat. Mater., Nat. Mater. **15**, 733 (2016).



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Purpose

Thermodynamic properties in quantum spin liquids

- Fractionalization of spins
- Temperature dependence

Kitaev model: exactly solvable quantum spin model

- Exactly solvable quantum spin model
- Canonical model for QSLs

Developing new technique for quantum spin systems

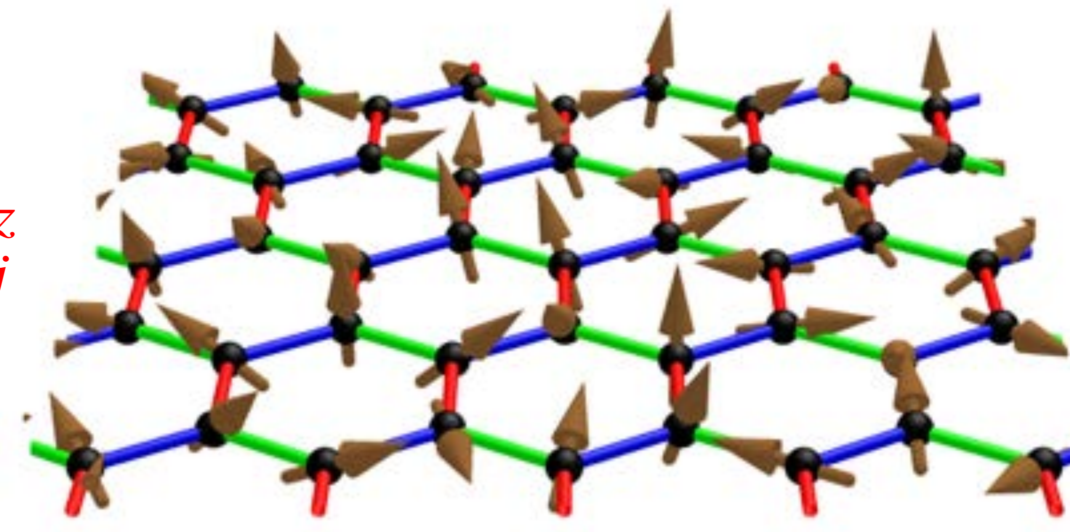
Tokyo Tech

Contents

Method

Quantum **spin model**

$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$



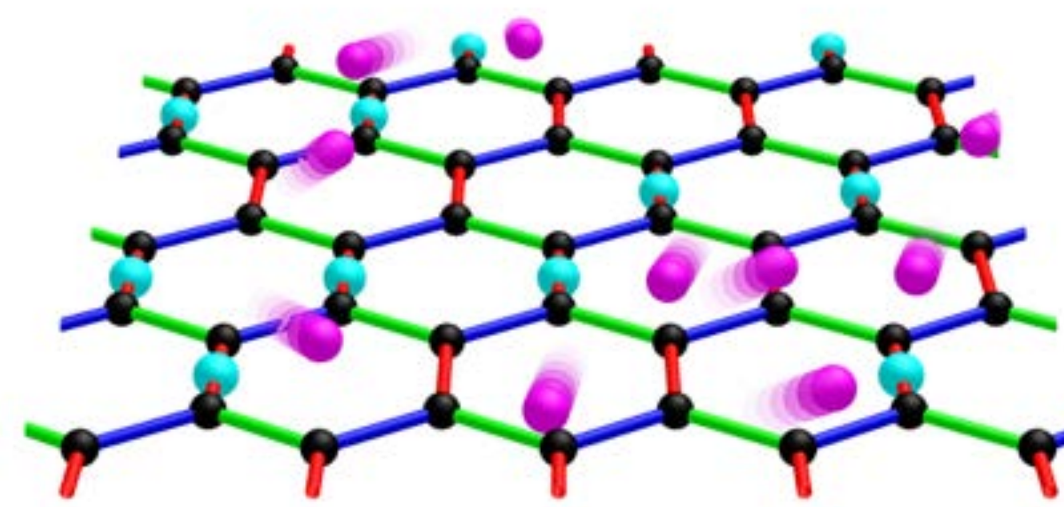
Jordan-Wigner transformation

Itinerant fermion **model**

$$\mathcal{H} = \frac{iJ_x}{4} \sum_{\langle ij \rangle_x} c_i c_j - \frac{iJ_y}{4} \sum_{\langle ij \rangle_y} c_i c_j - \frac{iJ_z}{4} \sum_{\langle ij \rangle_z} \eta_r c_i c_j$$

$$\eta_r = i\bar{c}_i \bar{c}_j$$

$\boldsymbol{S}_i$ →
- c_i : Itinerant Majorana
- $\bar{c}_i$: Localized Majorana



Free Majorana fermion system with thermally fluctuating fluxes $W_p = \eta_r \eta_{r'}$

- Sign problem-free "*Quantum*" Monte Carlo simulations

  Quantum nature of *S*=1/2 spins is fully taken into account!

- Simulations are *classical* and done for flipping Ising valuables η_r.

$$J_x = J_y = J_z = J$$

Monte Carlo simulation

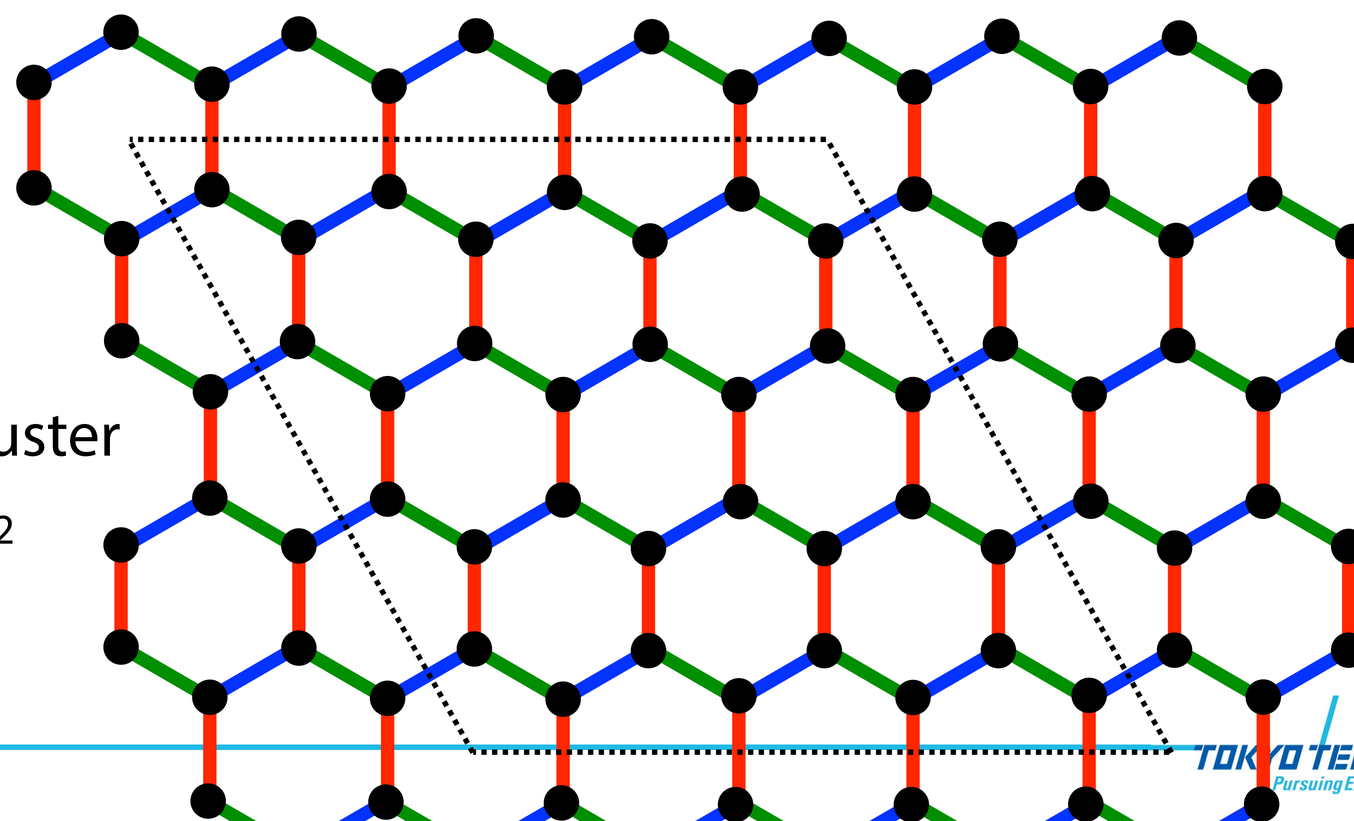
$$\mathcal{H} = \frac{iJ_x}{4}\sum_{\langle ij\rangle_x} c_i c_j - \frac{iJ_y}{4}\sum_{\langle ij\rangle_y} c_i c_j - \frac{iJ_z}{4}\sum_{\langle ij\rangle_z} \eta_r c_i c_j \qquad \eta_r = \pm 1$$

- Partition function: $Z = \mathrm{Tr}_{\{\eta_r\}}\mathrm{Tr}_{\{c_i\}} e^{-\beta\mathcal{H}} = \sum_{\{\eta_r=\pm1\}} e^{-\beta F_f(\{\eta_r\})}$

  $F_f(\{\eta_r\}) = -T \ln \mathrm{Tr}_{\{c_i\}} e^{-\beta\mathcal{H}(\{\eta_r\})}$ calculated by exact diagonalization

- $\{\eta_r\}$ are updated so as to reproduce the distribution $e^{-\beta F_f(\{\eta_r\})}$
- Sign-free "Quantum" Monte Carlo simulation for QSLs
- System size $2\mathrm{x}L^2$ up to L=20 (800sites)
- Only one energy scale:

  $J_x = J_y = J_z = J$

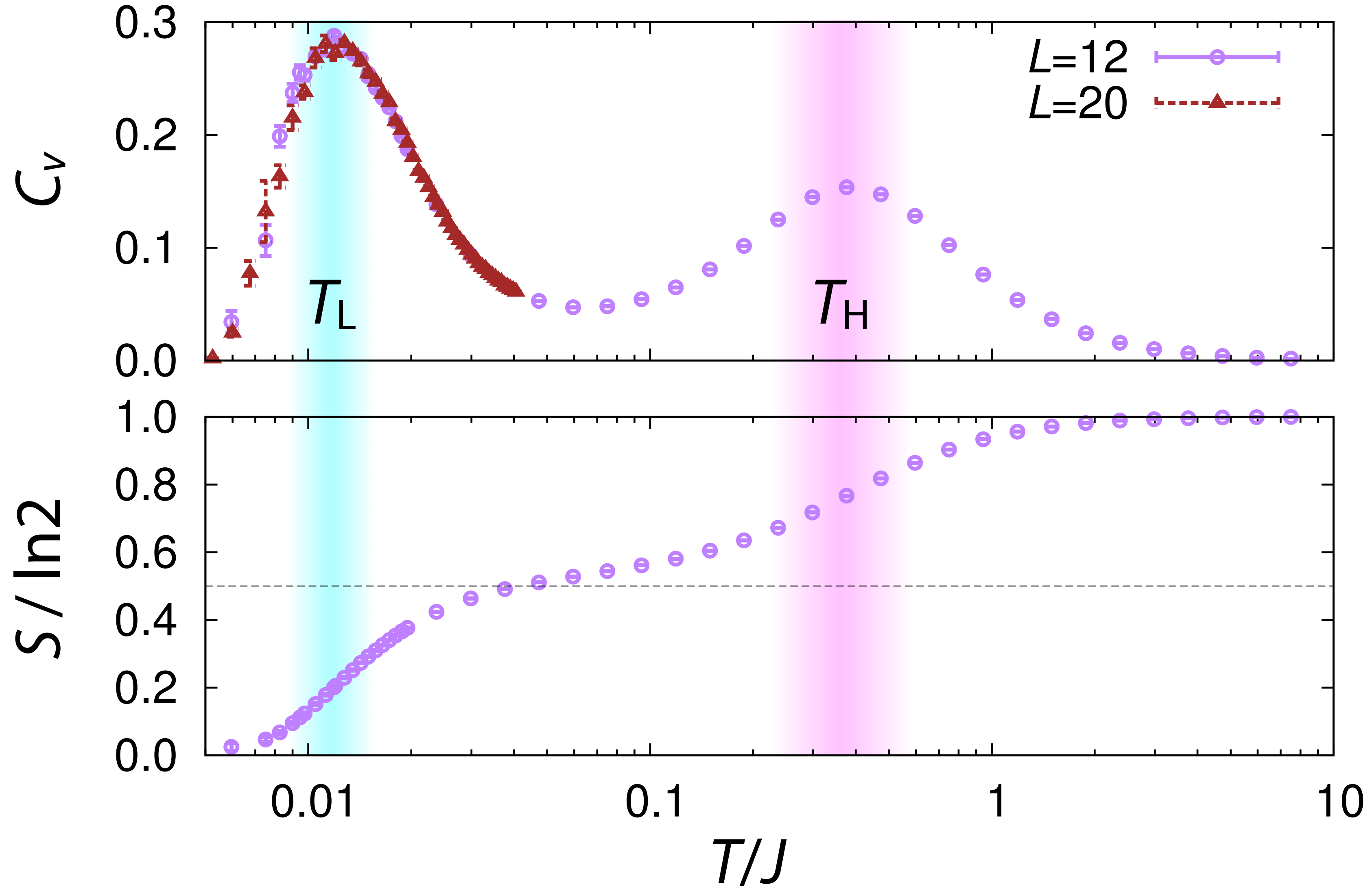


L=4 cluster

$N=2\mathrm{x}4^2$

Contents

Specific heat and entropy



Kitaev model：S=1/2 quantum spin model

Energy scale: only $J(=J_x=J_y=J_z)$

☑ Two peak structure

☑ A half of the entropy is released

Thermal fractionalization of

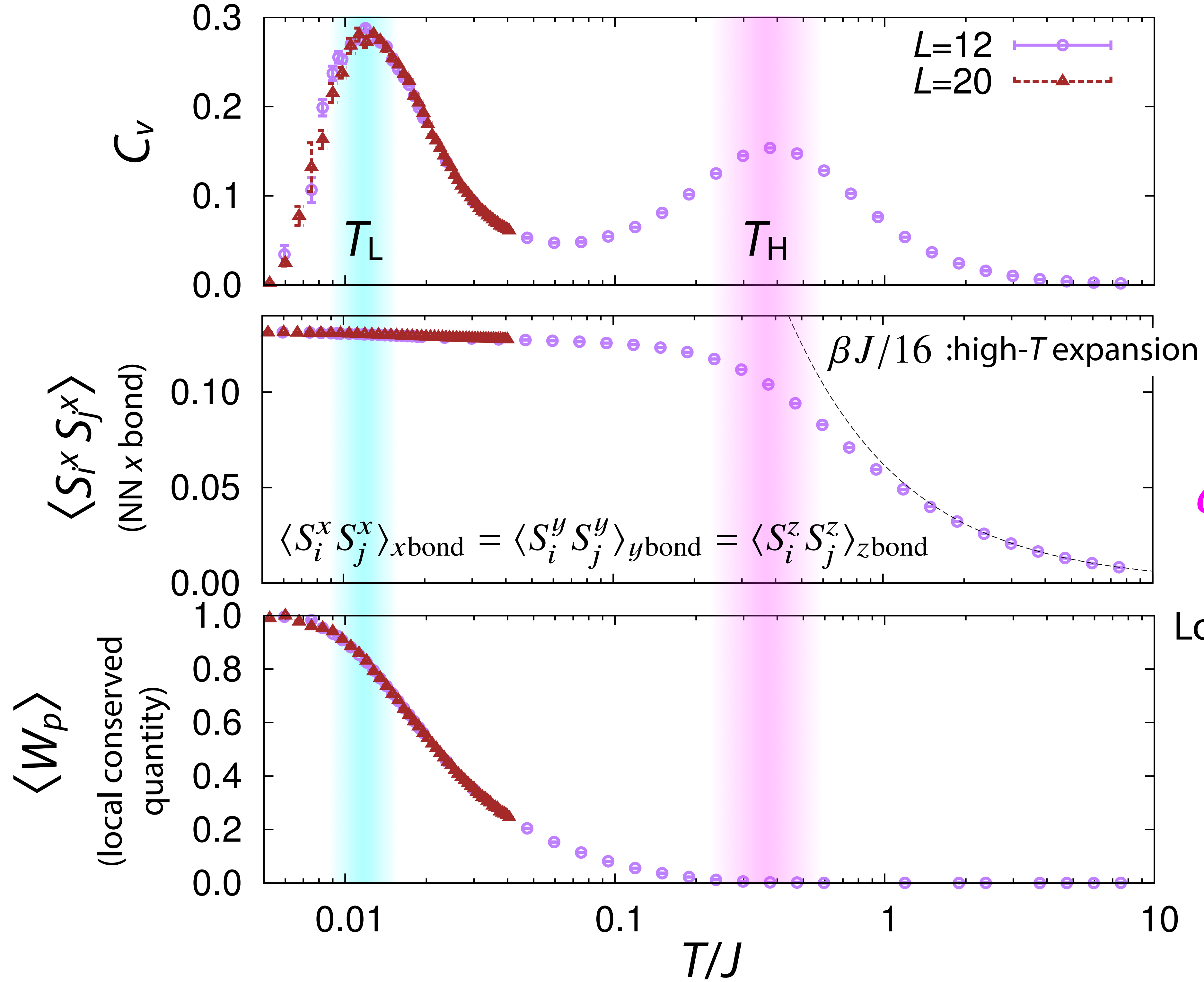
S=1/2 quantum spin

S_i → c_i : Itinerant Majorana

S_i → $\bar{c}_i$: Localized Majorana

JN, M. Udagawa, and Y. Motome: Phys. Rev. B **92**, 115122 (2015).

Fractionalization of spins



NN x bond

$$S_i^x S_j^x = -\frac{i}{4} c_i c_j$$

c_i : itinerant Majorana (matter Majorana)

Local conserved quantity

$$W_p = \prod_{r \in p} \eta_r$$

$$\eta_r \equiv i \bar{c}_b \bar{c}_w$$

$\bar{c}_i$: localized Majorana (flux Majorana)

S_i → c_i : itinerant Majorana ⟷ Entropy release at T_H ⟷ development of spin correlation

S_i → $\bar{c}_i$: localized Majorana ⟷ Entropy release at T_L ⟷ coherence of W_p

JN, M. Udagawa, and Y. Motome: Phys. Rev. B **92**, 115122 (2015).

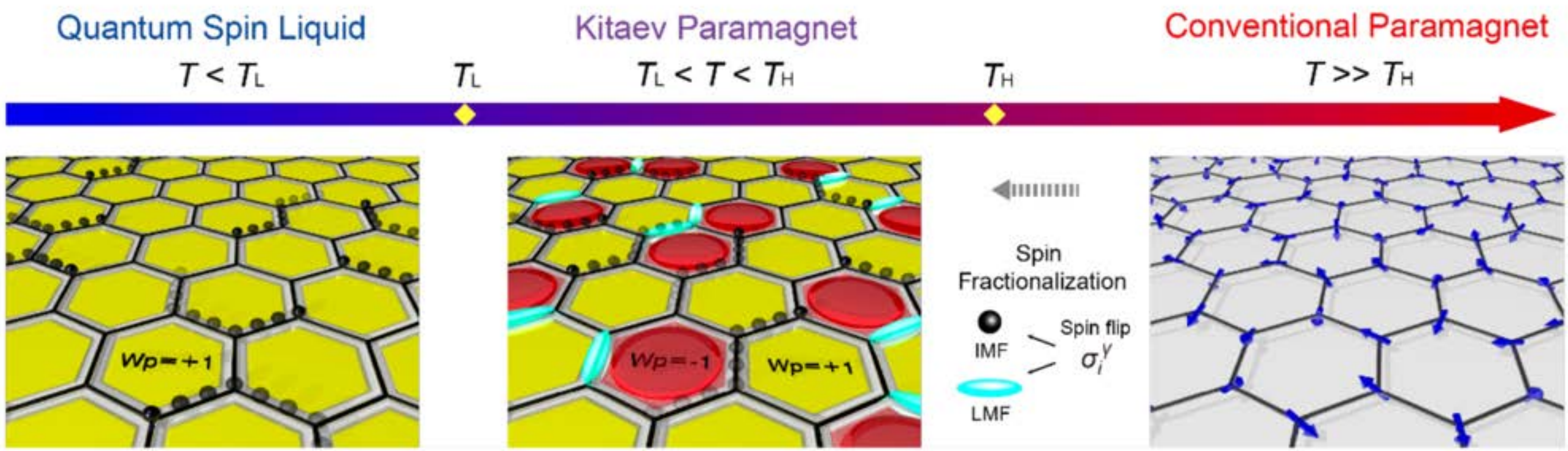
Schematic picture of T dependence

Freezing of Z_2 fluxes

(localized Majorana fermions)

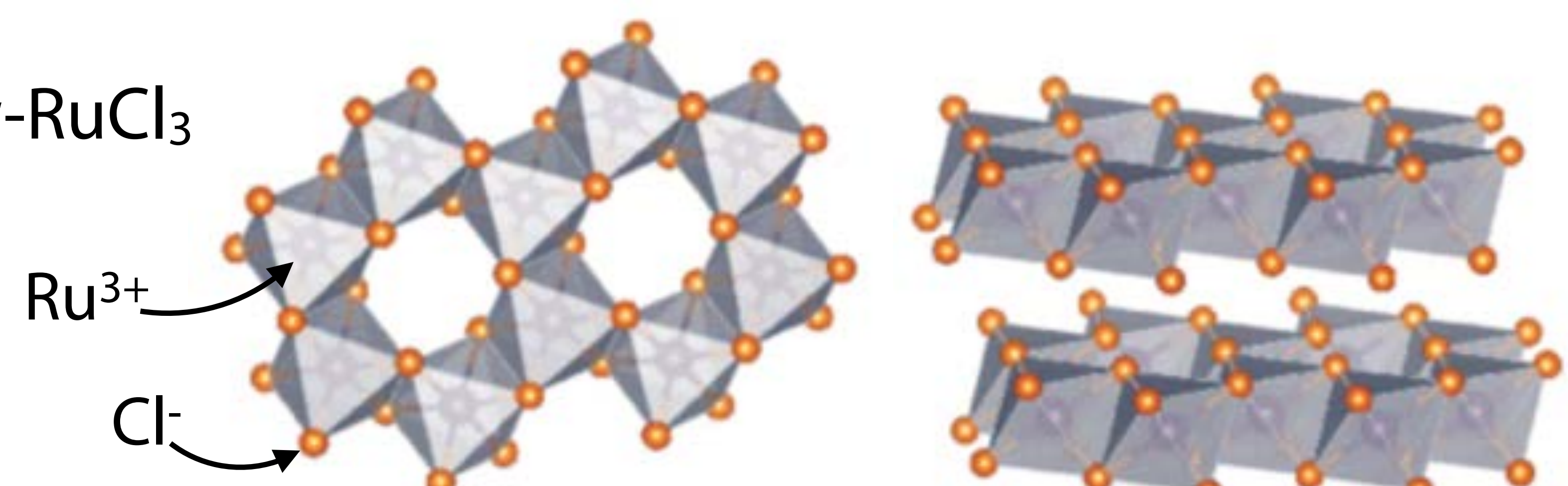
Formation of Fermi degeneracy of matter fermions

(itinerant Majorana fermions)



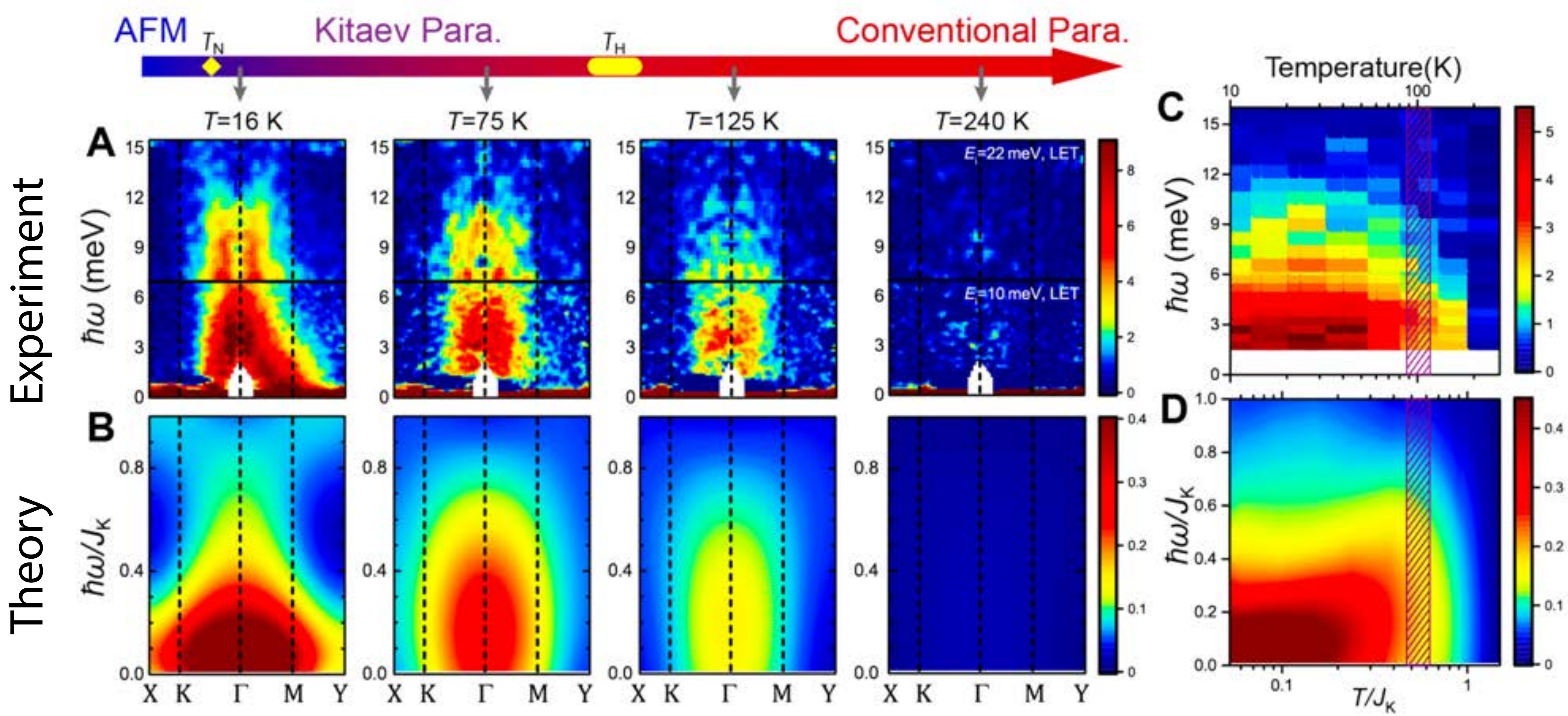
S.-H. Do et al., arXiv:1703.01081.

Candidate material: α-$RuCl_3$



Relevance to real materials

Spin dynamics measured by neutron scattering experiment in α-$RuCl_3$

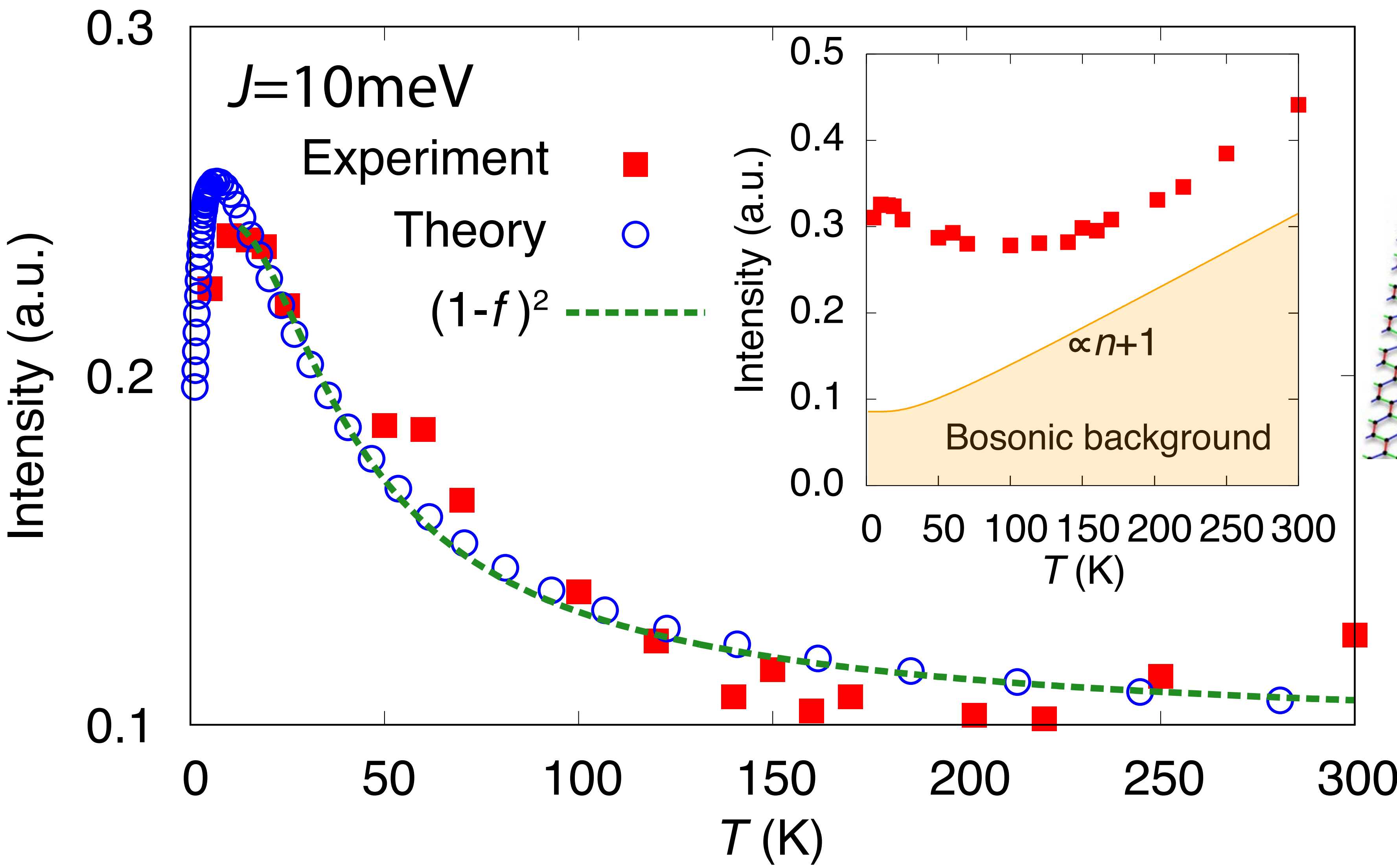


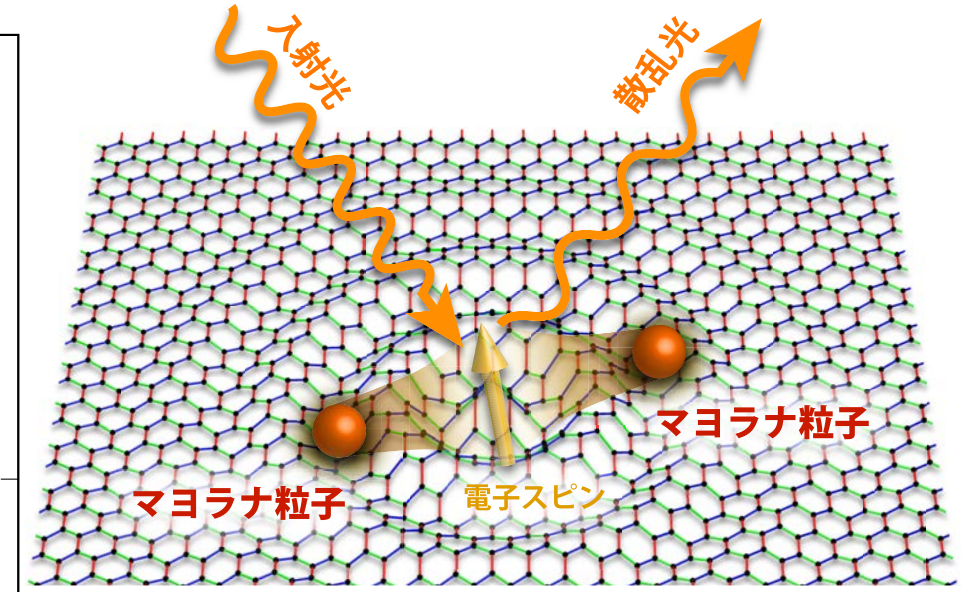
S.-H. Do et al., arXiv:1703.01081.

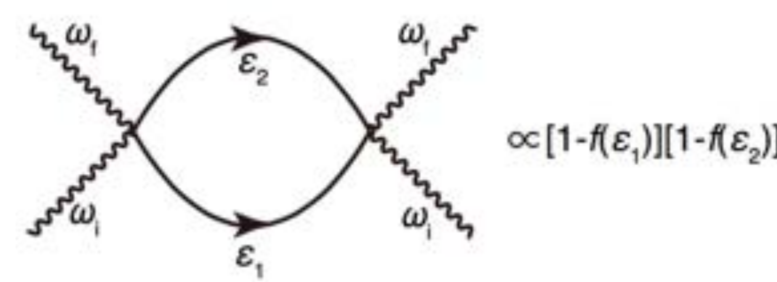
- Good agreement between the present theory and experimental results.
- Neutron scattering data are well reproduced by our theory.

Relevance to real materials

Magnetic Raman scattering







L. J. Sandilands et al., Phys. Rev. Lett. **114**, 147201 (2015).

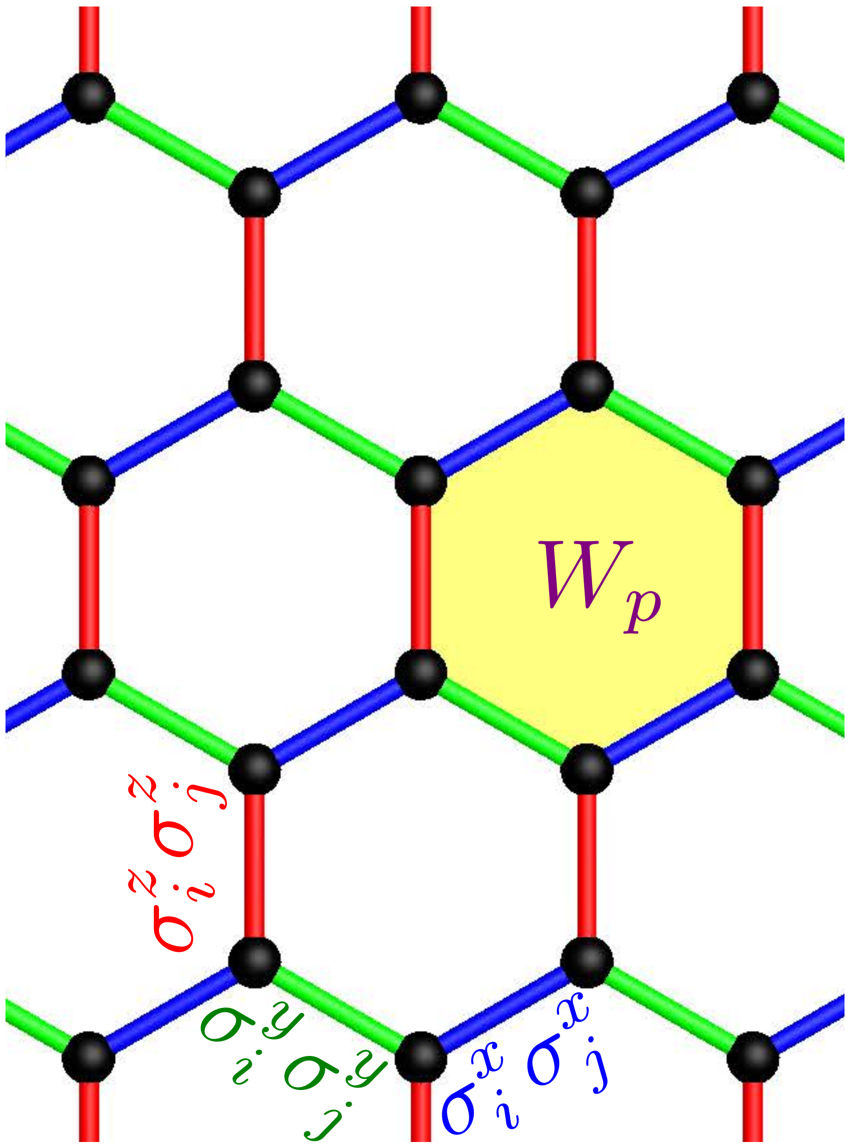
JN, J. Knolle, D. L. Kovrizhin, Y. Motome, R. Moessner, Nat. Phys., **12**, 912 (2016).

- Raman spectrum in *α*-$RuCl_3$
- Good agreement between the present theory and experimental results.
- Functional form indicates the existence of ***fractional fermionic excitations***
- This *T* dependence is also observed in β-, γ-Li_2IrO_3 A. Glamazda et al., Nat. Commun. 7, 12286 (2016).

Contents

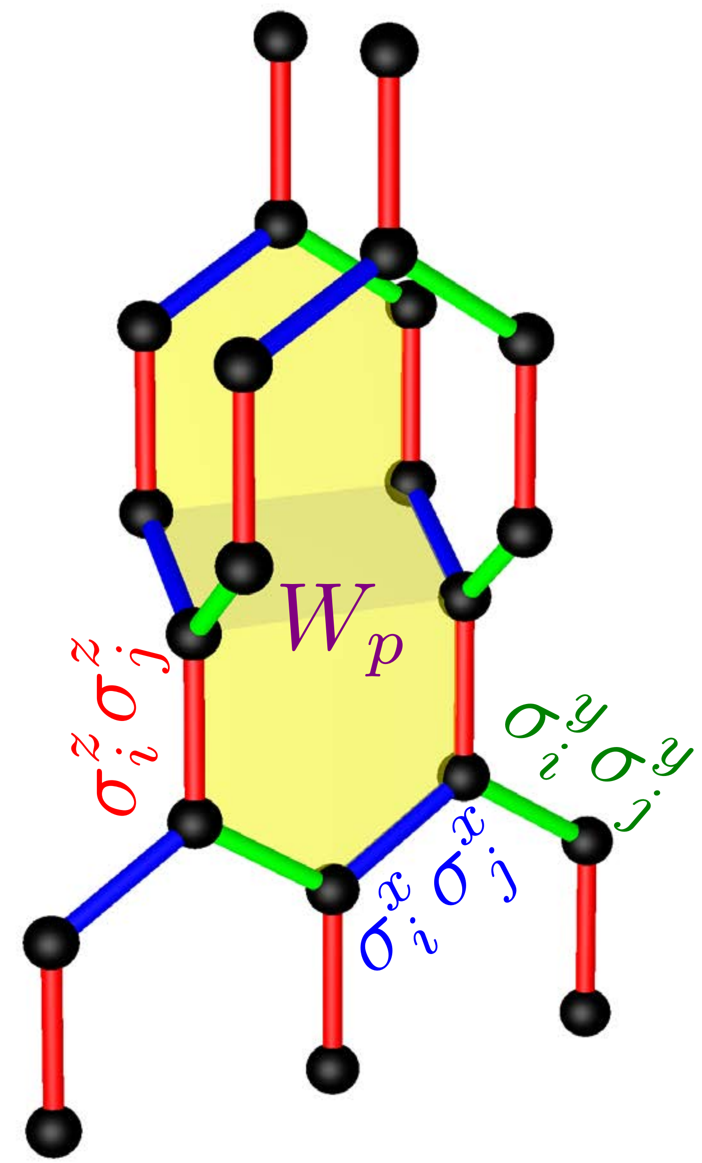
Extension to 3D

Honeycomb lattice



- ☑ A_2IrO_3 (A=Li,Na)
- ☑ α-$RuCl_3$

Hyperhoneycomb lattice



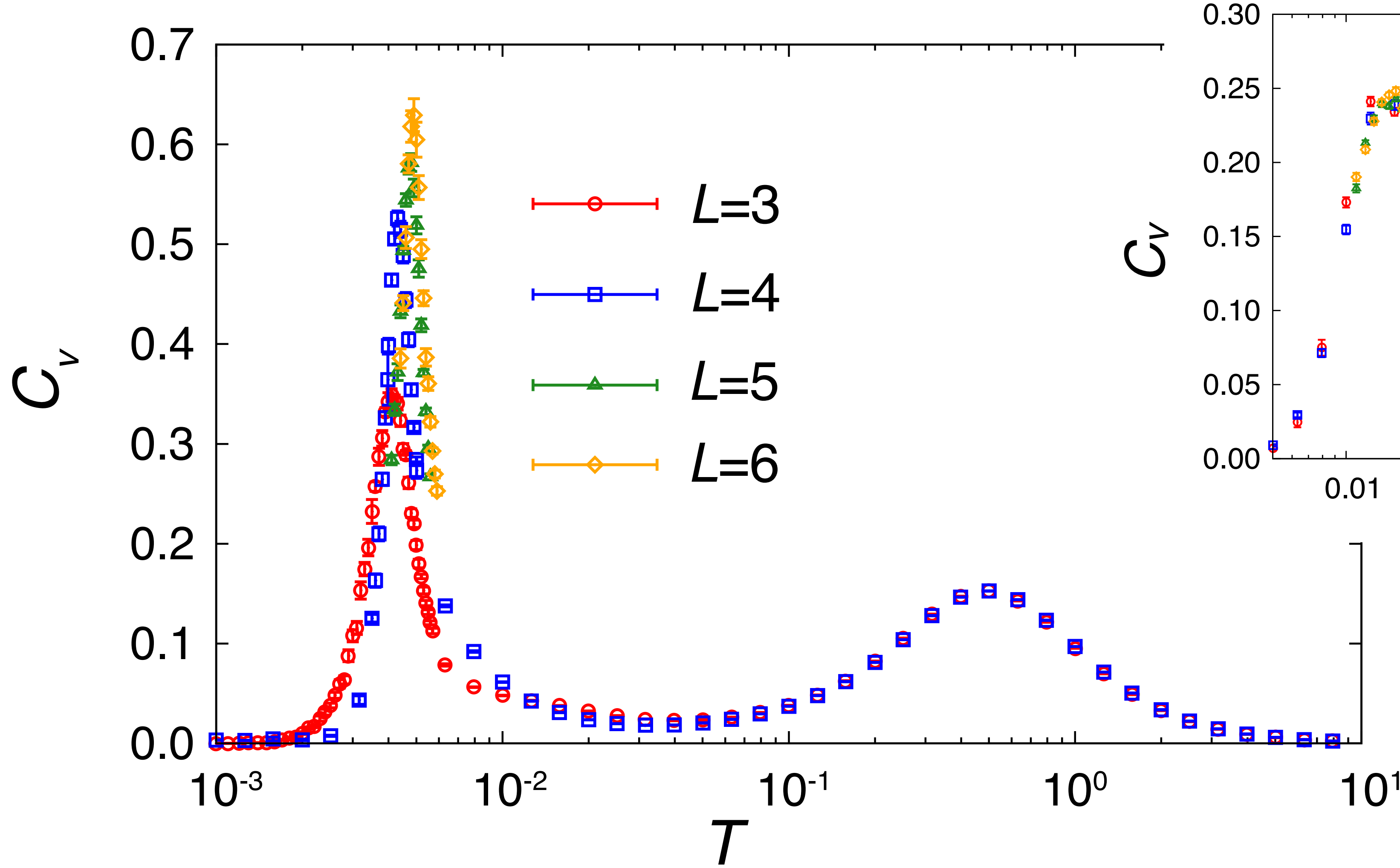
- ☑ β-Li_2IrO_3

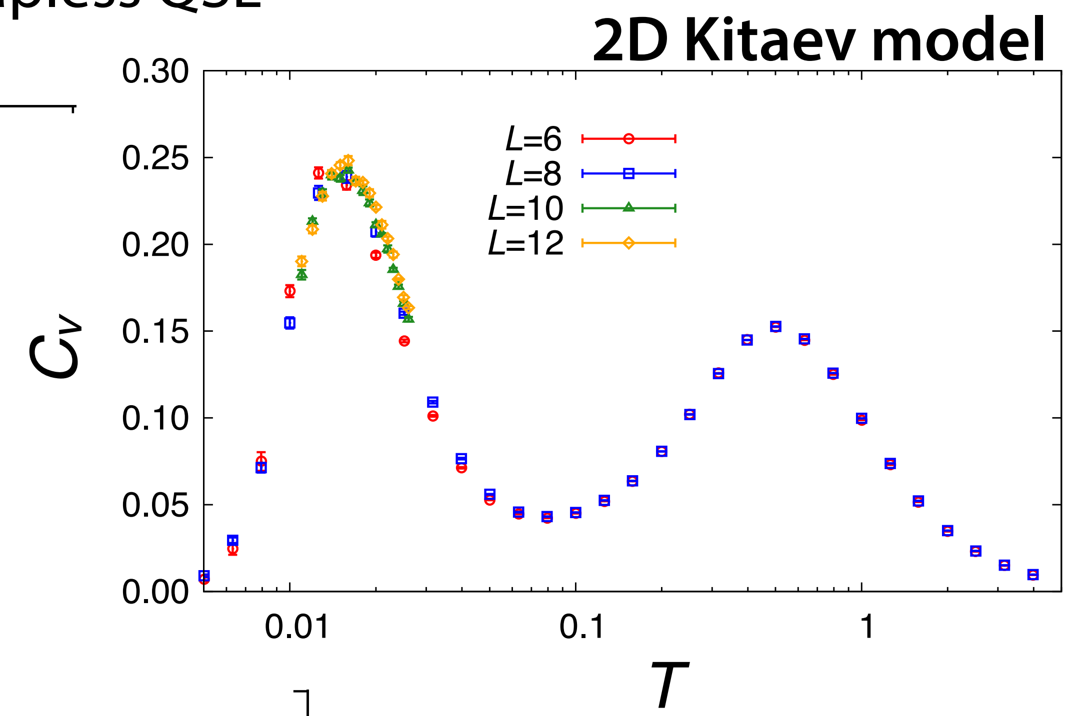
JN, M. Udagawa, and Y. Motome: Phys. Rev. Lett. **113**, 197205 (2014).

Specific heat

$$\mathcal{H} = -J_x \sum_{<ij>_x} \sigma_i^x \sigma_j^x - J_y \sum_{<ij>_y} \sigma_i^y \sigma_j^y - J_z \sum_{<ij>_z} \sigma_i^z \sigma_j^z$$

$J_x = J_y = J_z = 1/3$: ground state is gapless QSL





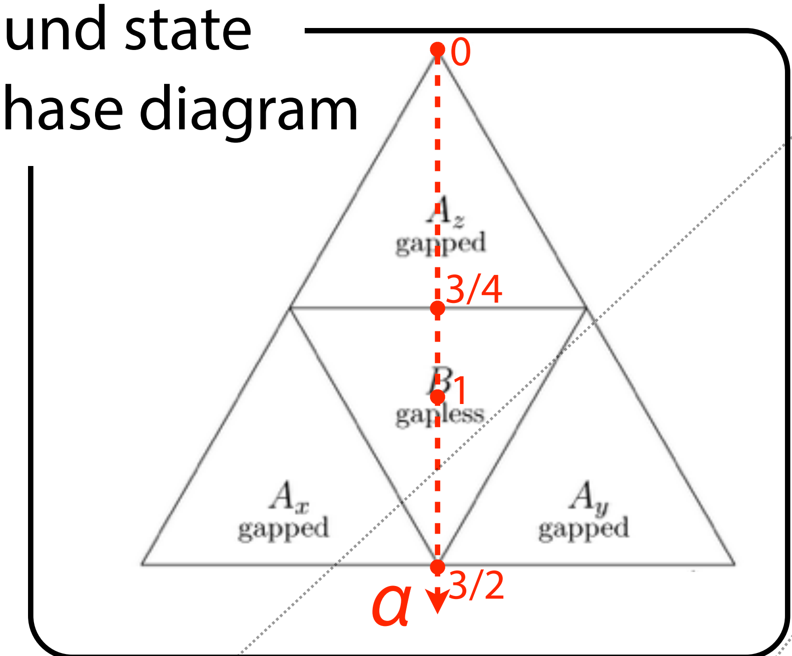
High temperature peak (Size independent)

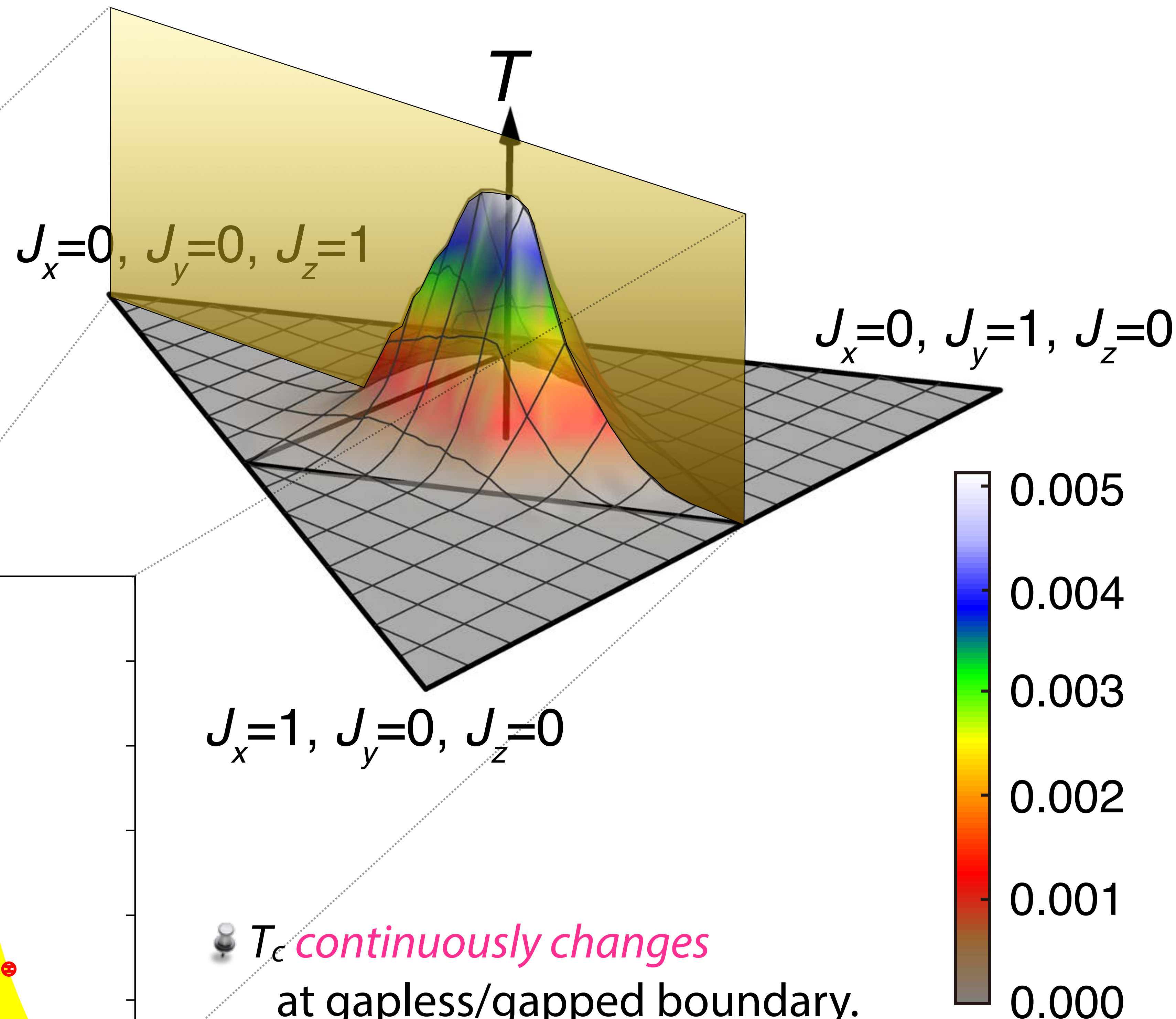
Low temperature peak (Size dependent)

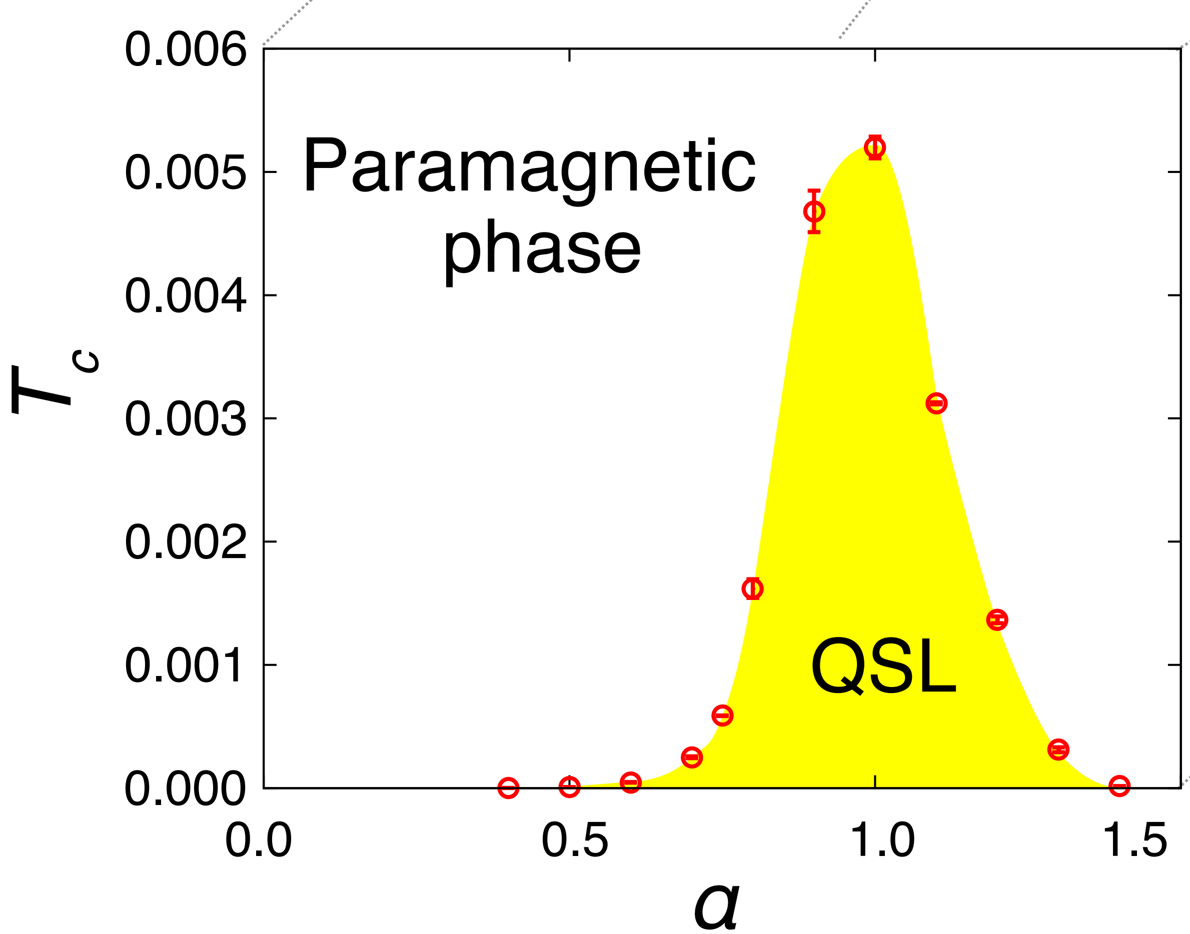
→ *Phase transition* (T_c~ 0.0052)

JN, M. Udagawa, and Y. Motome: Phys. Rev. Lett. **113**, 197205 (2014).

Phase transition







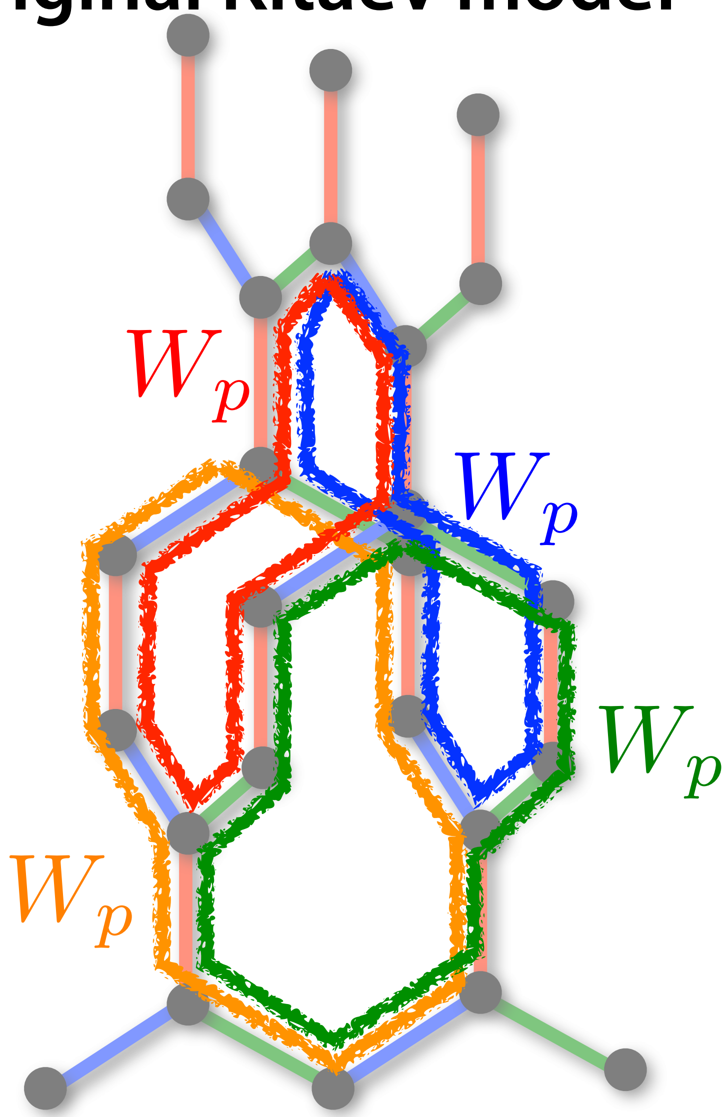
- T_c *continuously changes* at gapless/gapped boundary.
- T_c takes maximum at $J_x=J_y=J_z$.

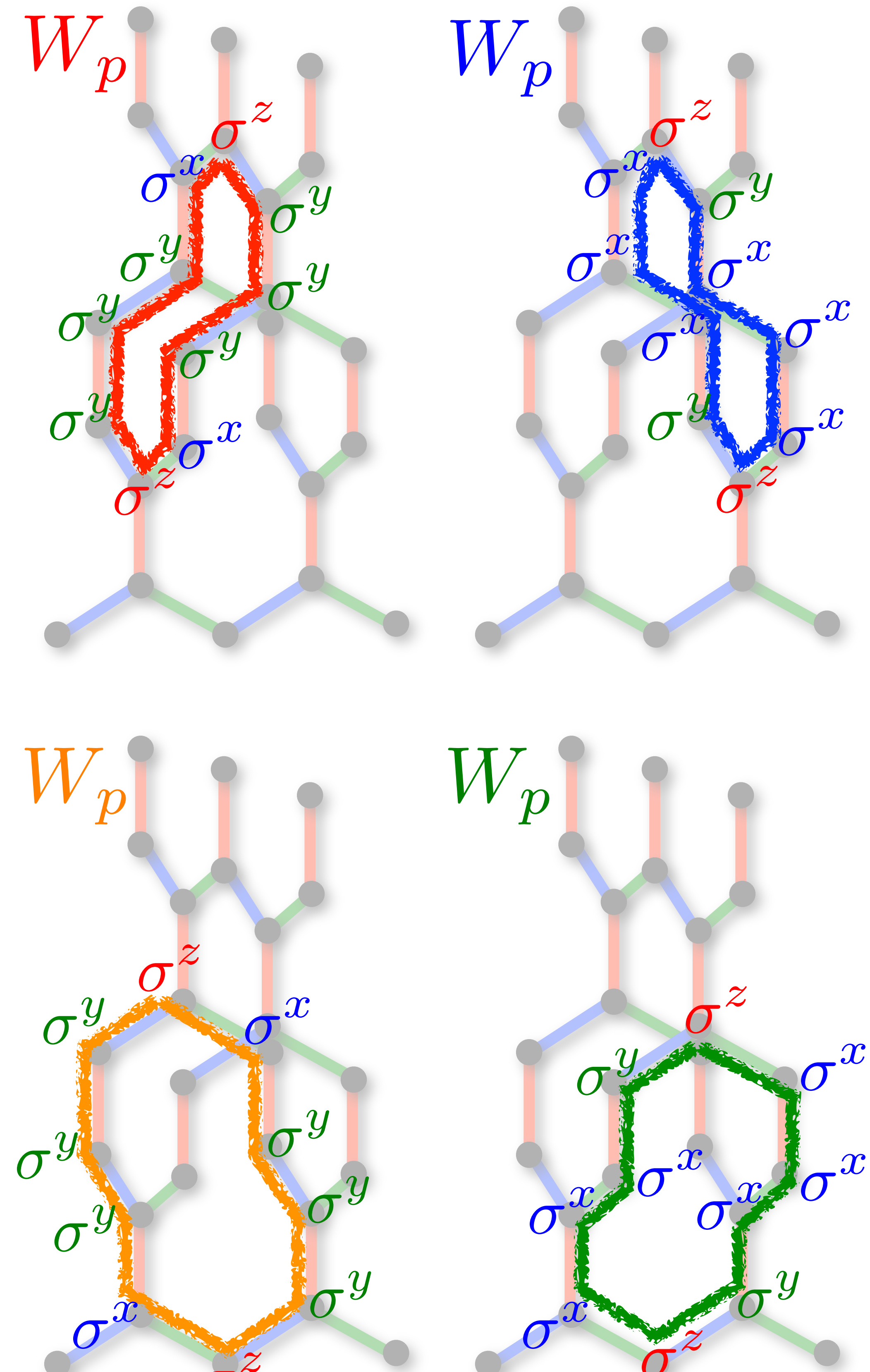
Frustration stabilizes the QSL.

JN, M. Udagawa, and Y. Motome: Phys. Rev. Lett. **113**, 197205 (2014).

Local constraints for fluxes

Original Kitaev model





Algebra of Pauli matrices

$$(\sigma^x)^2 = (\sigma^y)^2 = (\sigma^z)^2 = 1$$

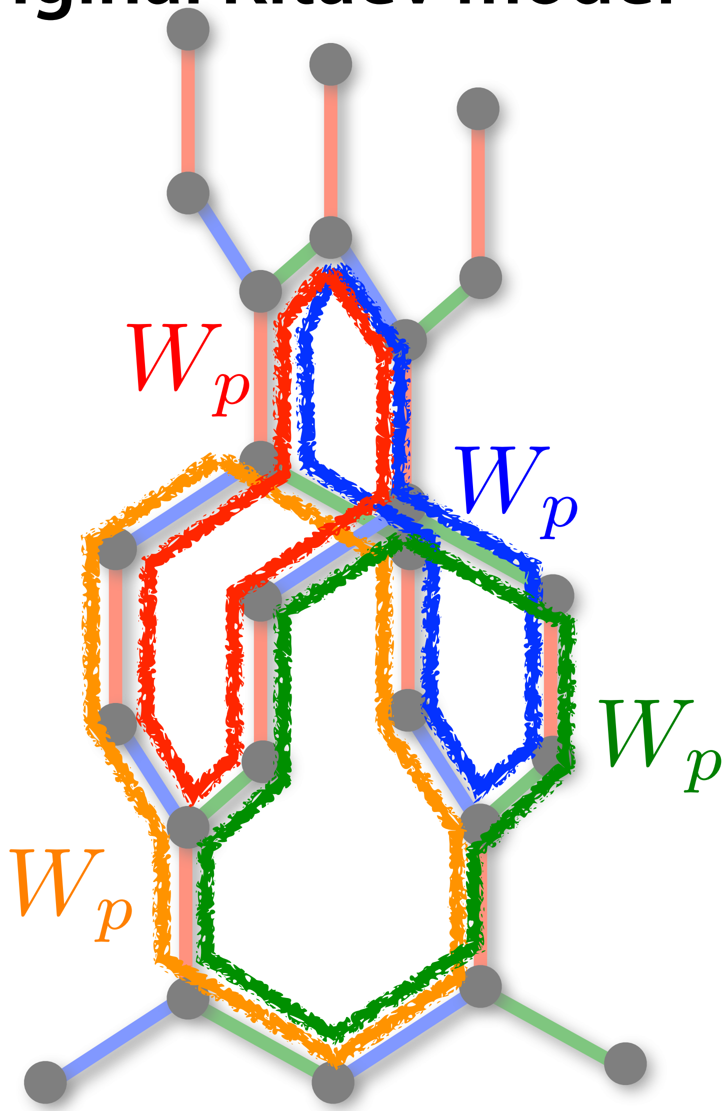
$$\sigma^x \sigma^y \sigma^z = i$$

$$\Rightarrow \quad W_p W_p W_p W_p = 1$$

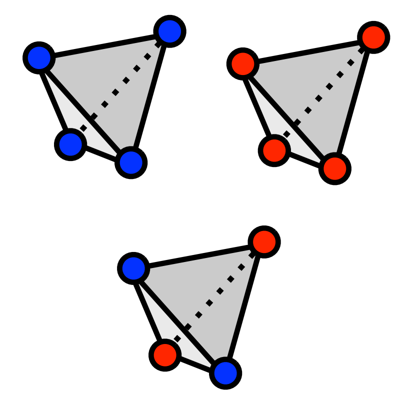
JN, M. Udagawa, and Y. Motome: Phys. Rev. Lett. **113**, 197205 (2014).

Nature of the phase transition

Original Kitaev model

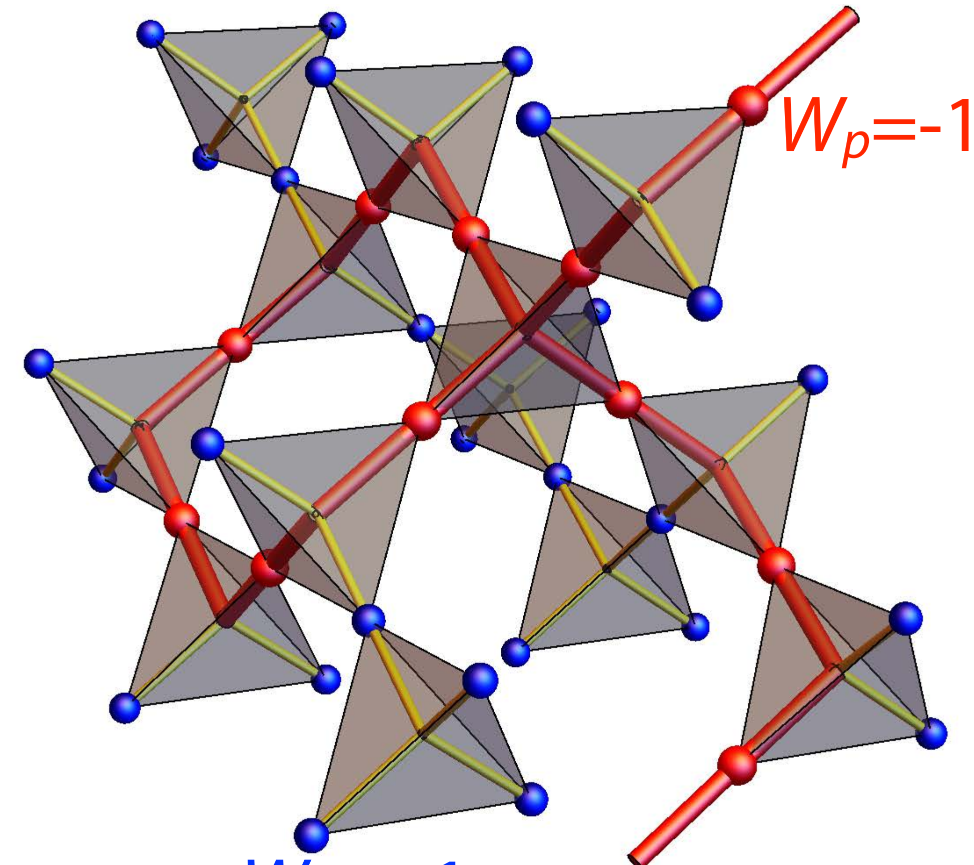


$$W_p W_p W_p W_p = 1$$



Number of $W_p = -1$ is even.

W_p form a pyrochlore lattice



Local constraint for W_p in the original Kitaev model originating from ***algebra of Pauli matrices***

"Quantum-effect-induced rigid constraints"

→ Flipped W_p form a loop. *cf.* Peierls argument

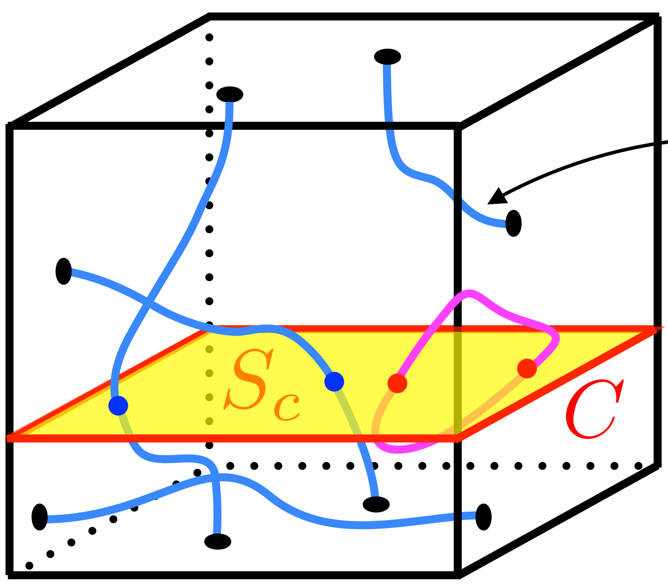
→ **Topological characterization** by W_p-loops.

JN, M. Udagawa, and Y. Motome: Phys. Rev. Lett. **113**, 197205 (2014).

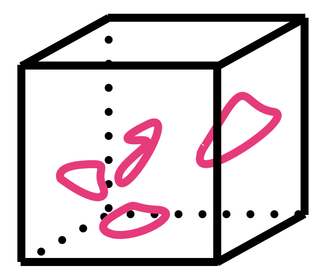
Loop excitation

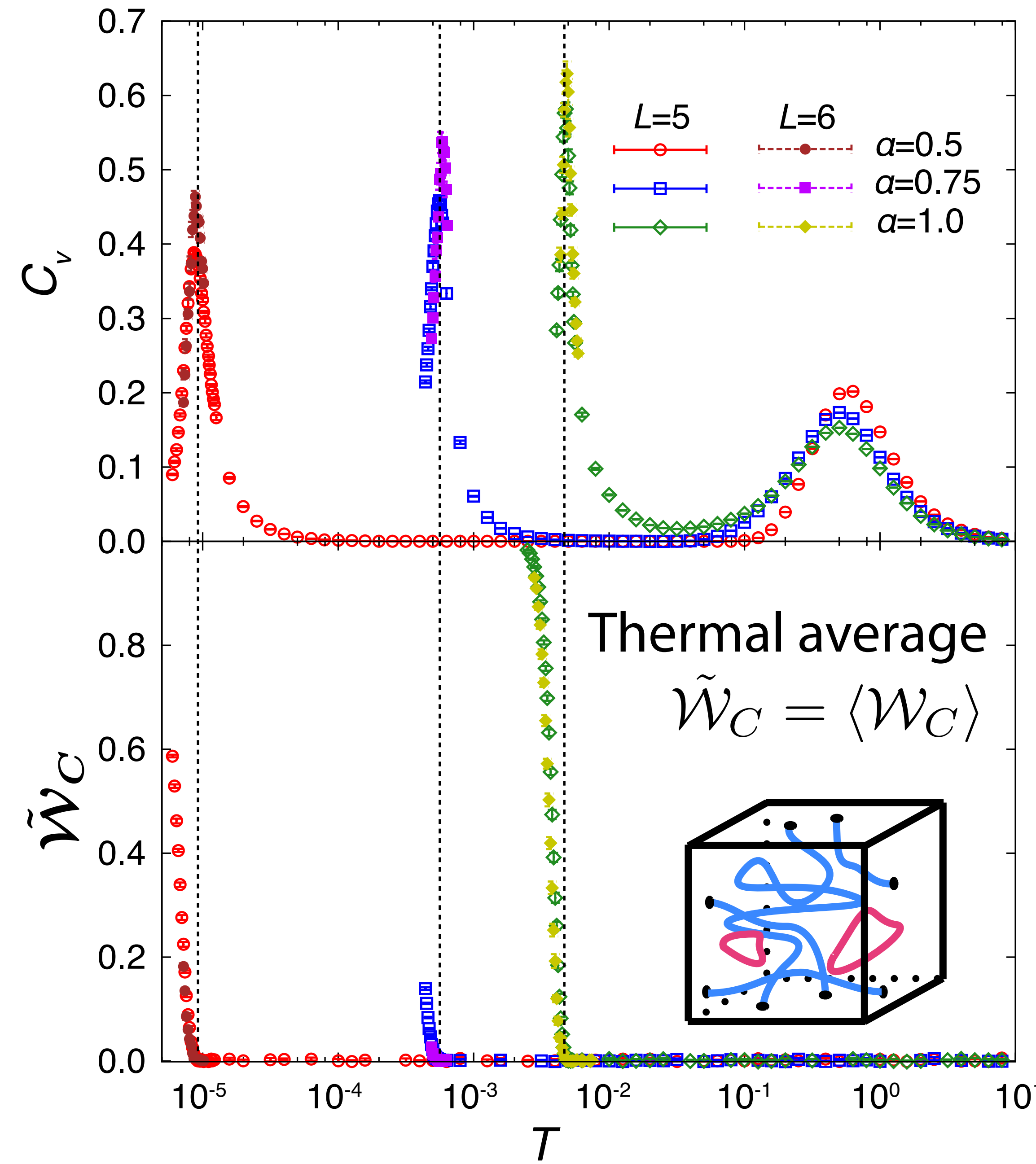
Parity of the intersection number between loops and a surface:

$$\mathcal{W}_C = \prod_{i \in C} \sigma_i^{l_i} = \prod_{p \in S_C} W_p \quad \text{(Conserved quantity)}$$



flipped $W_p = -1$





Thermal average

Extended loops : $\mathcal{W}_C = +1$ or -1 → $\tilde{\mathcal{W}}_C = 0$

Short loops : $\mathcal{W}_C = +1$ → $\tilde{\mathcal{W}}_C = 1$

$\tilde{\mathcal{W}}_C$ behaves like *an order parameter.*

JN, M. Udagawa, and Y. Motome: Phys. Rev. Lett. **113**, 197205 (2014).

Contents

Summary

Kitaev model: Canonical model with QSL ground state

Finite-temperature properties (static & dynamic)

- Static and dynamical quantities

  → Thermal fractionalization of spins

- Comparison with experiments

  → Good agreement with experiments

- Finite-temperature phase transition to QSL in 3D Kitaev model

  → Topological characterization