基研研究会 量子多体系研究の新しい潮流
2014年12月17日

スピン液体とトポロジー
局所拘束条件と局所保存量の観点から

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Objective of this talk

quantum spin liquids = new state of matter in magnets (Mott insulators)

no long-range order down to T=0 due to quantum fluctuations

fundamental questions:

What is their intrinsic and unique nature?

Where are they realized? In which compounds/models?

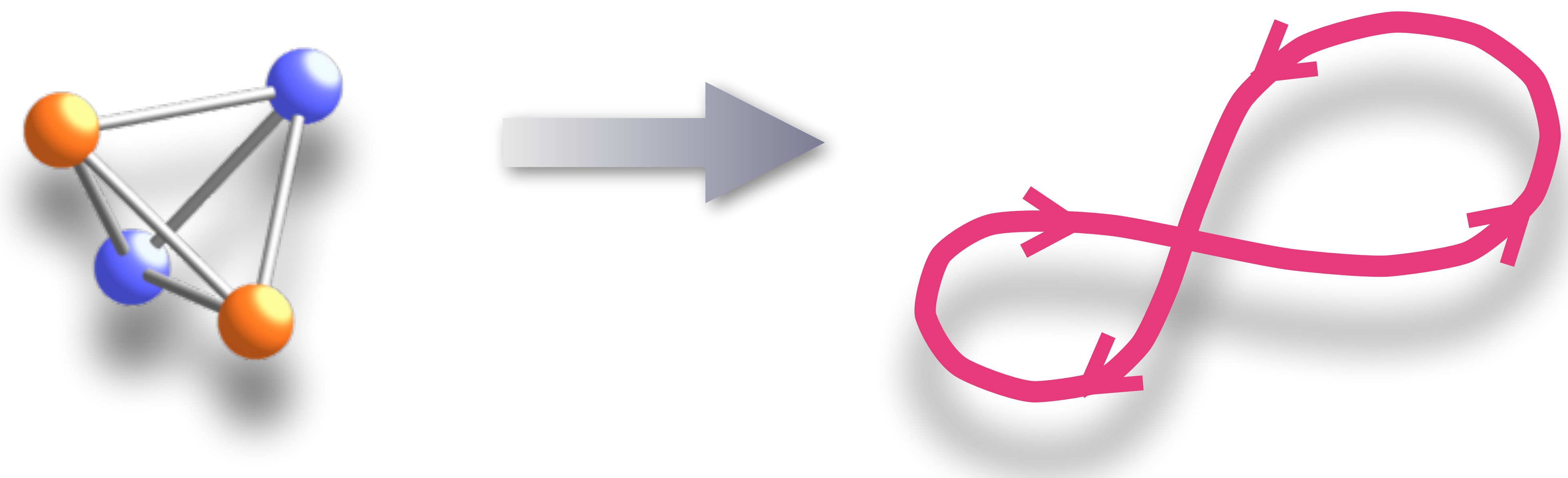
How to characterize them? How to distinguish them from paramagnets?

toward the answers to these questions:

- to review the present status from the author's biased view
- to present our recent findings on spin liquids
- to show some future directions

Message of this talk

local constraint | **emergent loop/flux**



- *topological nature*
- *exotic electronic and transport properties*
- *unconventional phase transitions*

Plan of this talk

- **What is spin liquid?**
  - classical and quantum theoretical examples
  - experimental candidates: spin ice, κ-ET salts, $Pr_2Ir_2O_7$, ...
- **Classical spin liquids**
  - spin ice and close-packed dimers
  - local constraint, Coulomb phase, unconventional phase transitions
- **Intermediate (hybrid)**
  - spin-charge coupling: transfer of peculiar spin textures to mobile electrons
  - loop liquid and scalar chiral liquid
- **Quantum spin liquids**
  - 3D Kitaev model: phase transition from paramagnet to spin liquid
  - topological aspect of the transition: proliferation of loops
- **Discussion and prospects**

What is spin liquid?

Classical example

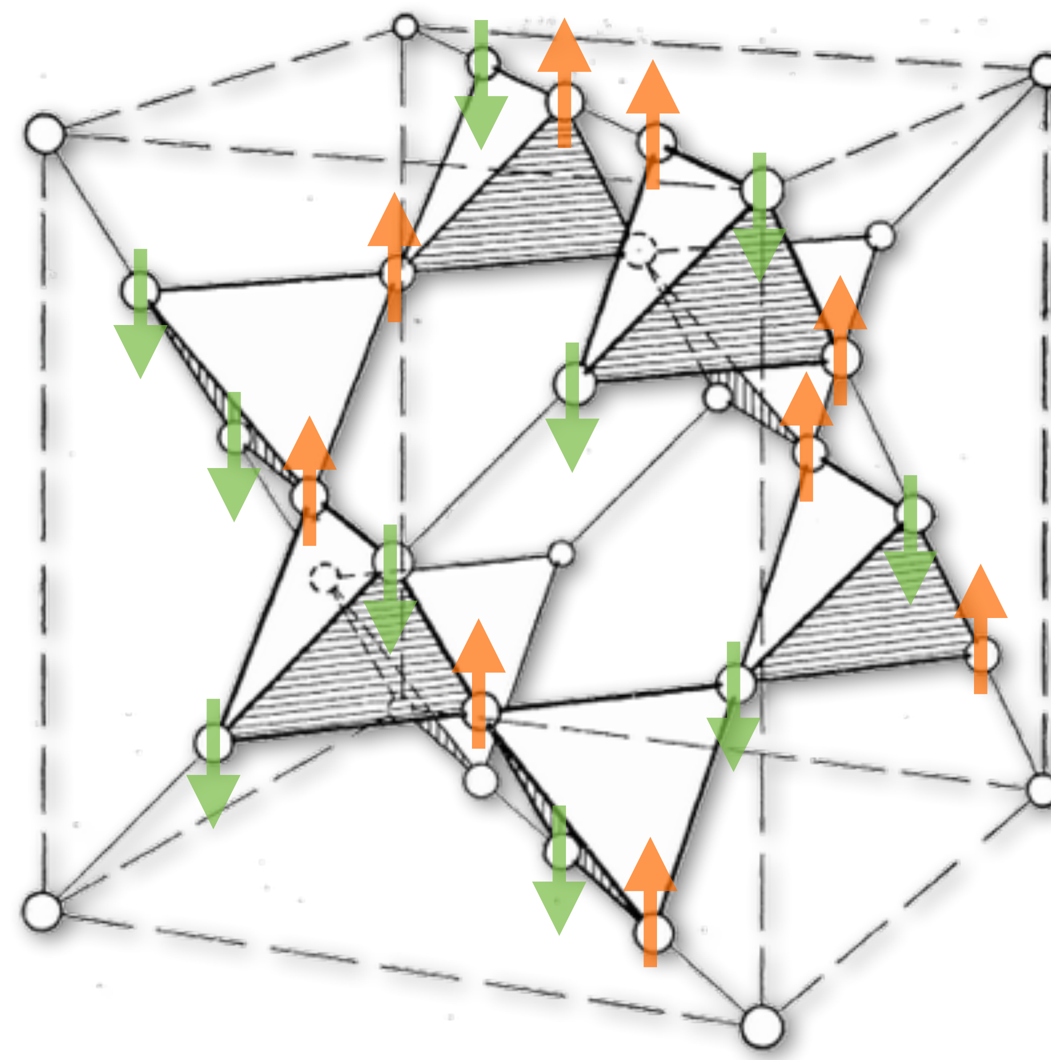
PHYSICAL REVIEW VOLUME 102, NUMBER 4 MAY 15, 1956

Ordering and Antiferromagnetism in Ferrites

P. W. Anderson
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received January 9, 1956)

- Verwey transition in magnetite Fe_3O_4
  - Fe^{3+}/Fe^{2+} on a pyrochlore lattice
  - Fe^{3+}=↑spin, Fe^{2+}=↓spin

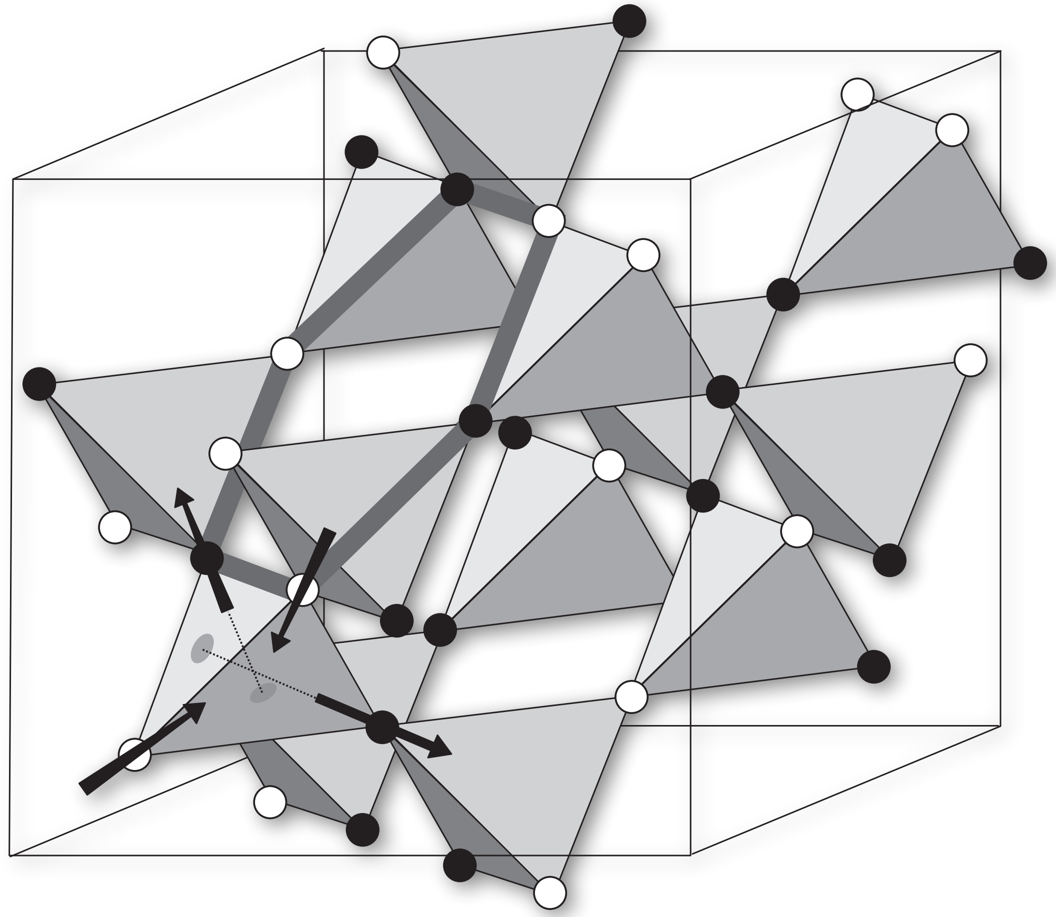
➡ pyrochlore Ising antiferromagnet
  - strong geometrical frustration
  - 2up-2down configuration is favored in each tetrahedron.
  - no long-range order down to $T=0$: macroscopic degeneracy (for nearest-neighbor interactions only)

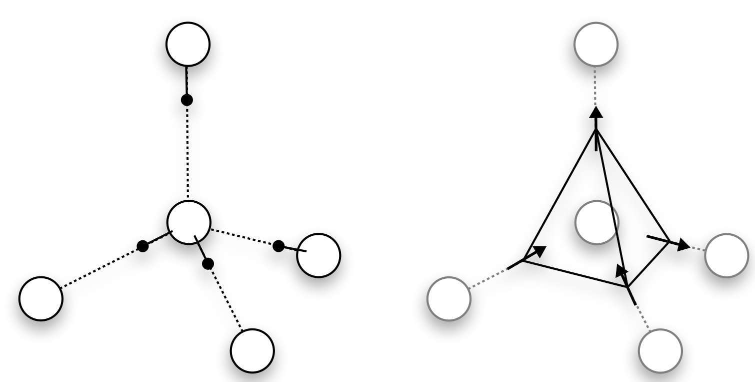


Classical (but modern) example

- spin ice model (nearest-neighbor interactions only)
  - ↑spin="in", ↓spin="out"
    - ➡ noncoplanar Ising spins on the pyrochlore lattice
  - strong geometrical frustration for *ferromagnetic* interactions
  - 2in-2out configuration is favored in each tetrahedron.
  - no long-range order down to T=0: macroscopic degeneracy

- magnetic analog of water ice
  - proton configurations = Ising spins with 2-in 2-out configurations

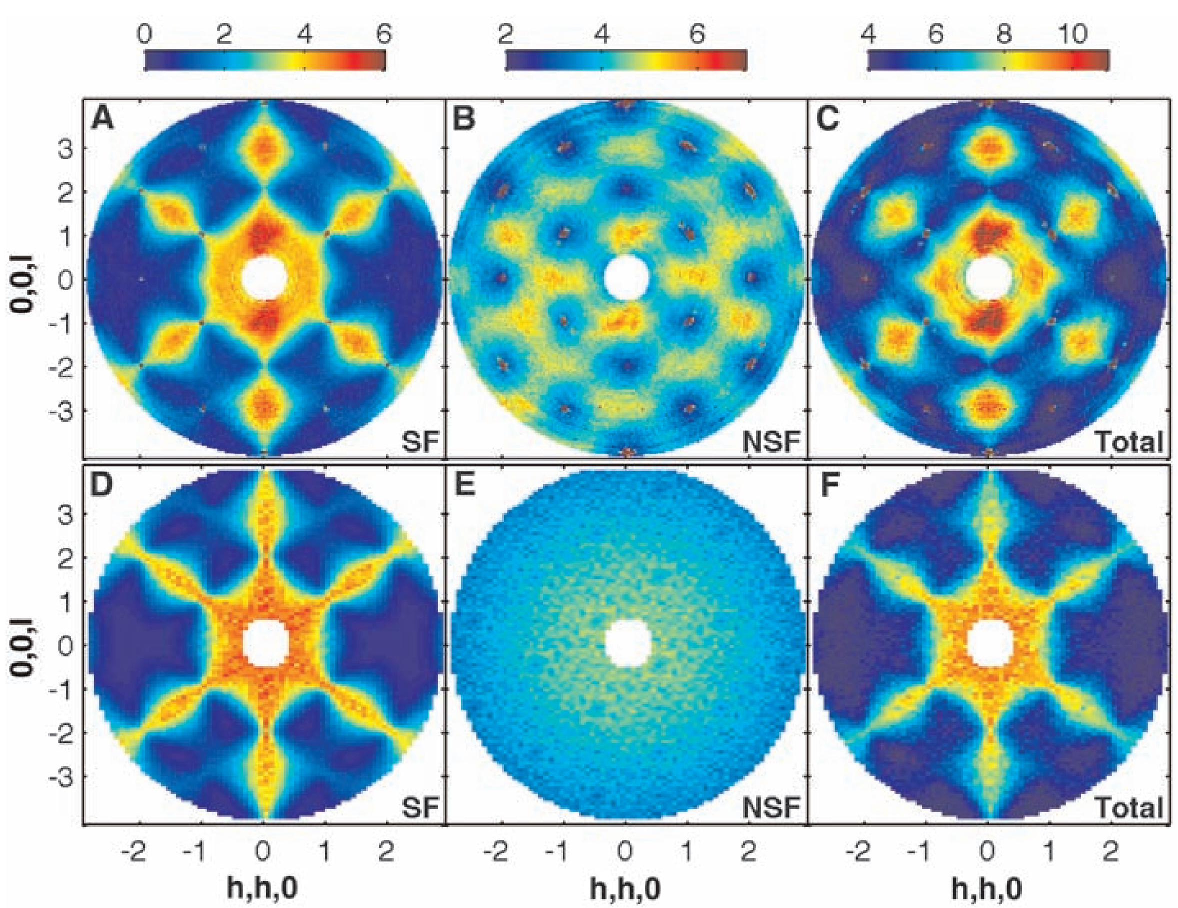




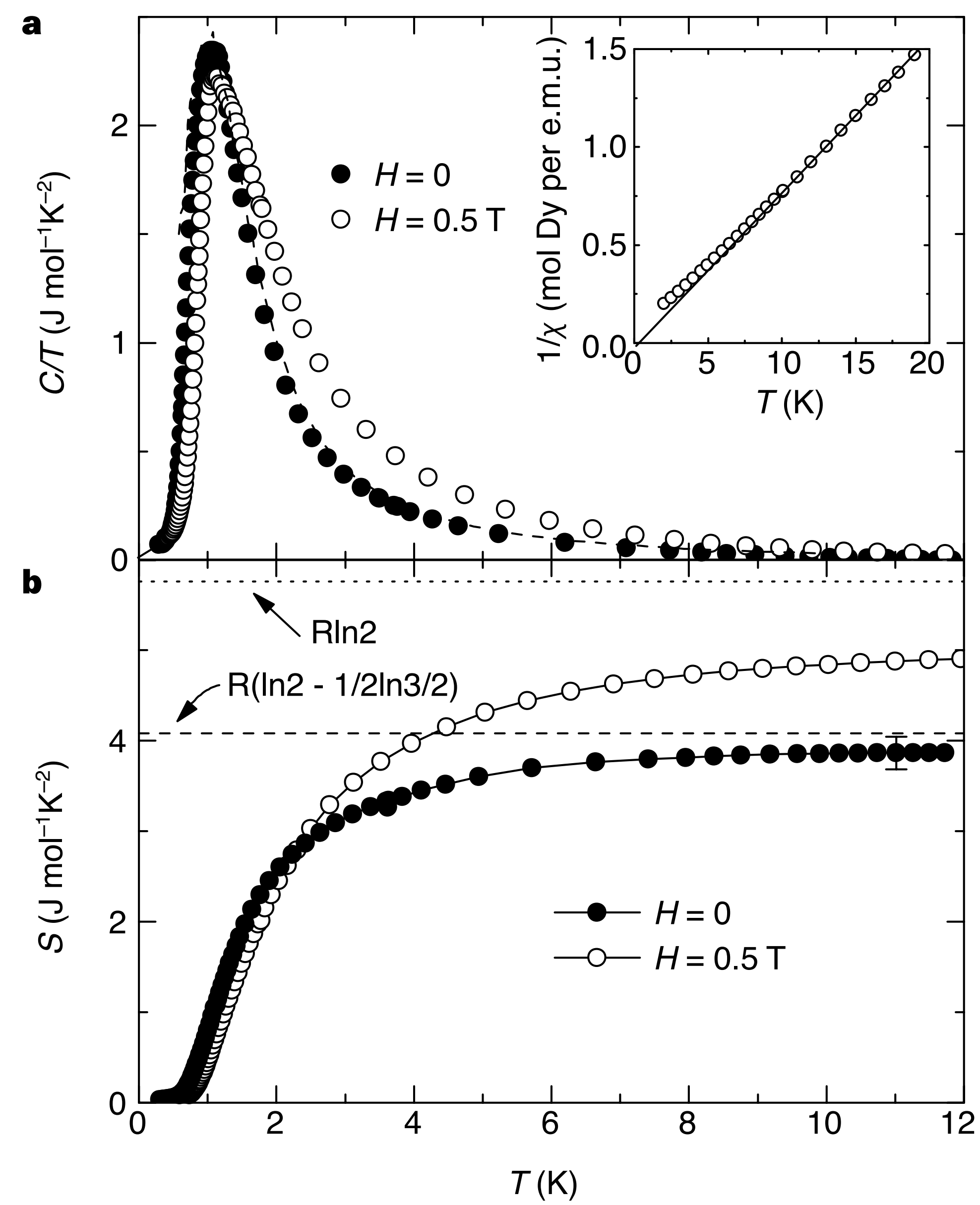
S. T. Bramwell and M. J. P Gingras, 2001

Experiment on spin ice

- rare-earth pyrochlore oxides: $Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$, $Ho_2Sn_2O_7$, ...
- no long-range order down to ~mK: only diffusive features in neutron scattering (T. Fennell *et al.*, 2009)



- residual entropy related to the macroscopic degeneracy (A. P. Ramirez *et al.*, 1999)



Quantum example (1)

- resonating valence bond (RVB) (P. W. Anderson, 1973)

$$|\Psi_{\mathrm{RVB}}\rangle = \sum |\text{singlet covering of the lattice}\rangle$$

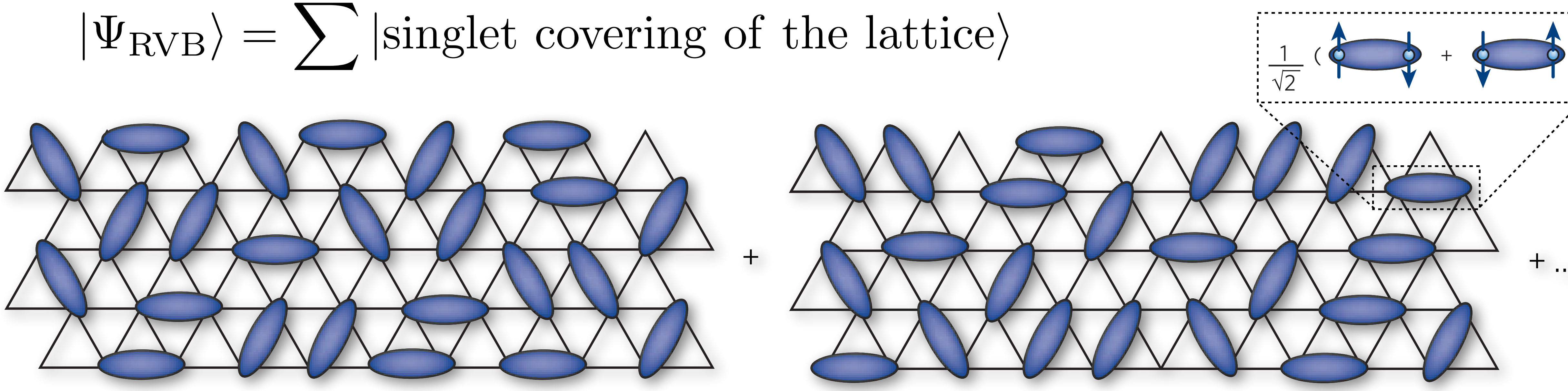
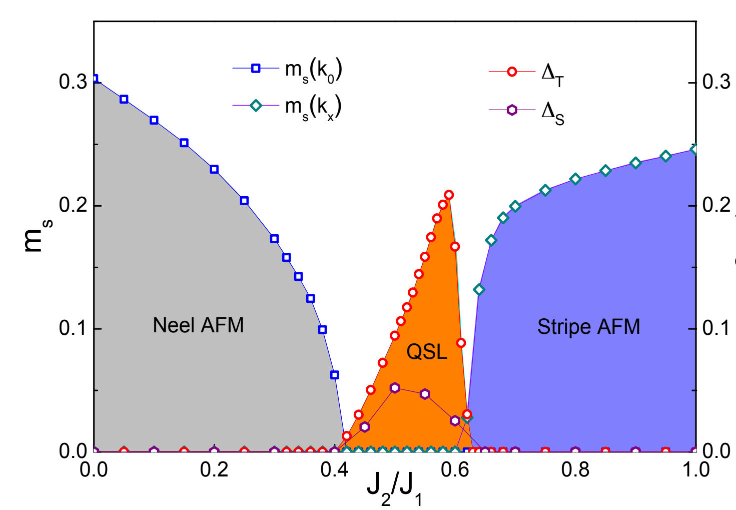


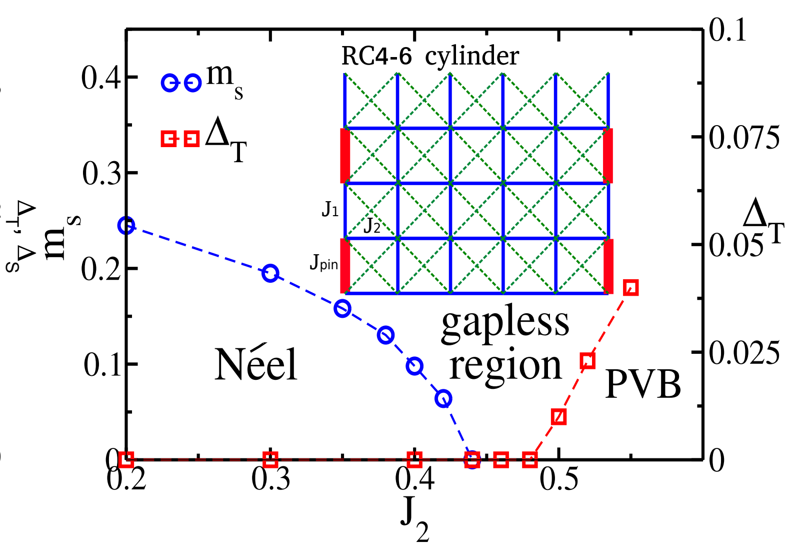
figure is taken and modified from L. Balents, 2010

- It is still controversial where such a state is realized.

  ex.) S=1/2 J_1-J_2 AF on a square lattice



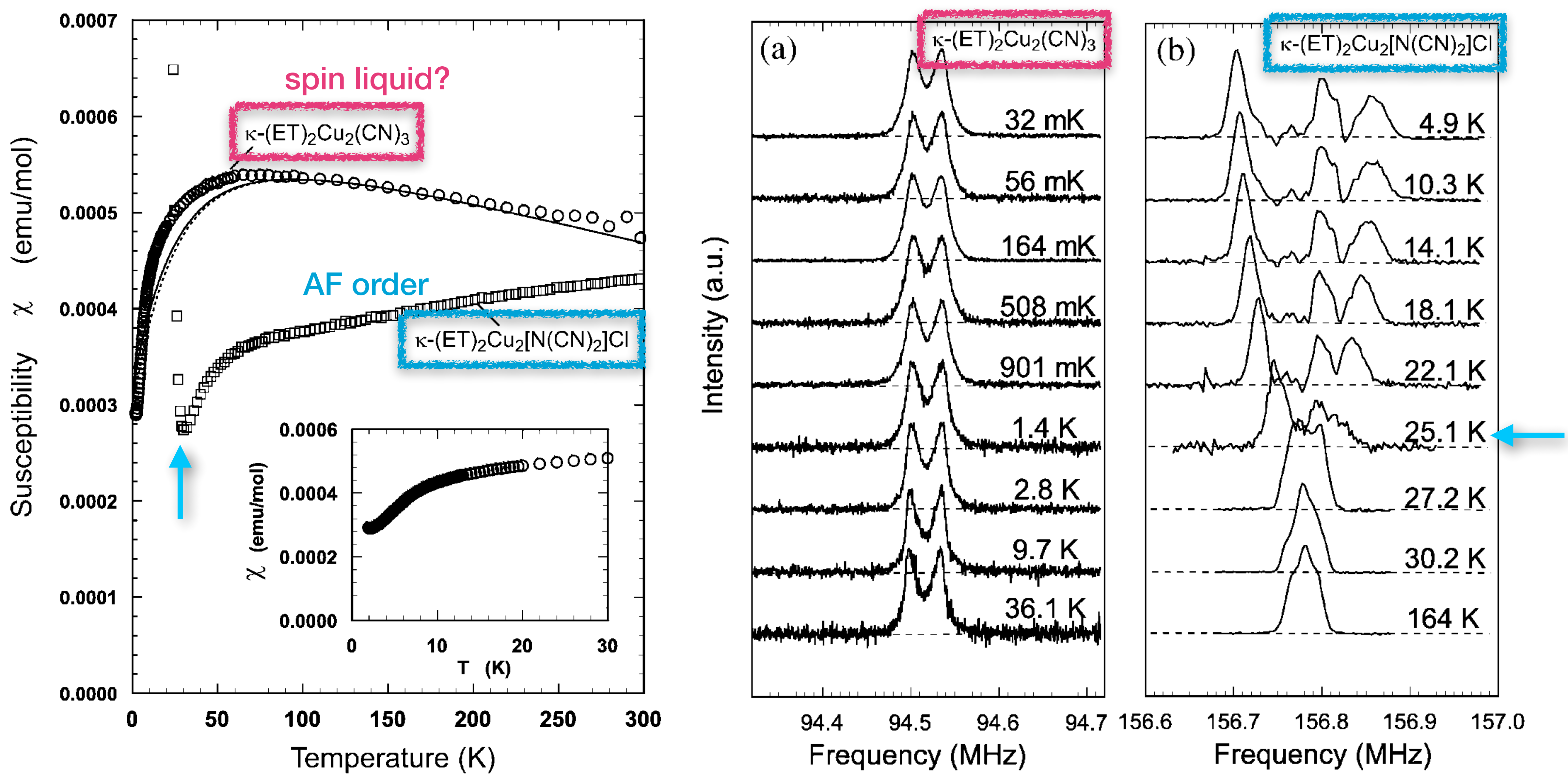
H.-C. Jiang *et al.*, 2012



S.-S. Gong *et al.*, 2014

Experimental candidate?

- organic conductor κ-$(ET)_2Cu_2(CN)_3$: S=1/2 spins on a triangular layers

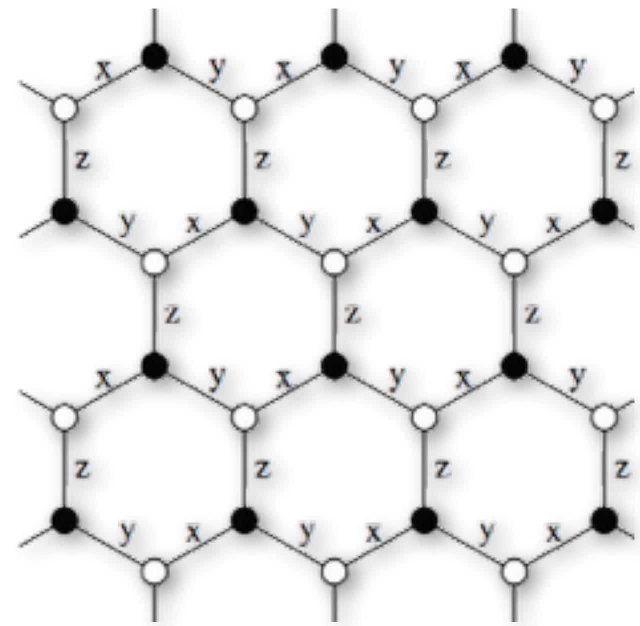


S. Shimizu *et al.*, 2003

Quantum example (2)

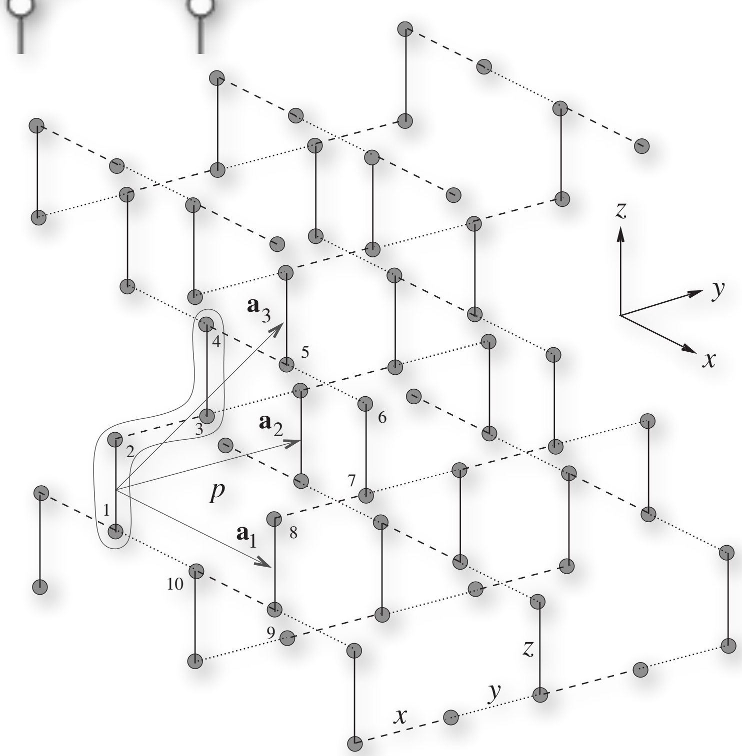
- Kitaev spin liquids: quantum spin liquids in the exact ground states

$$H = -J_x \sum_{\langle i,j\rangle_x} \sigma_i^x \sigma_j^x - J_y \sum_{\langle i,j\rangle_y} \sigma_i^y \sigma_j^y - J_z \sum_{\langle i,j\rangle_z} \sigma_i^z \sigma_j^z$$



  - original 2D honeycomb Kitaev model (A. Kitaev, 2006)
    - exactly soluble
    - both gapped and gapless spin liquids
    - nonzero correlations only for nearest-neighbors (G. Baskaran *et al.*, 2007)

  - extension to 3D hyperhoneycomb lattice (S. Mandal and N. Surendran, 2009)
    - also exactly soluble: the same ground-state phase diagram

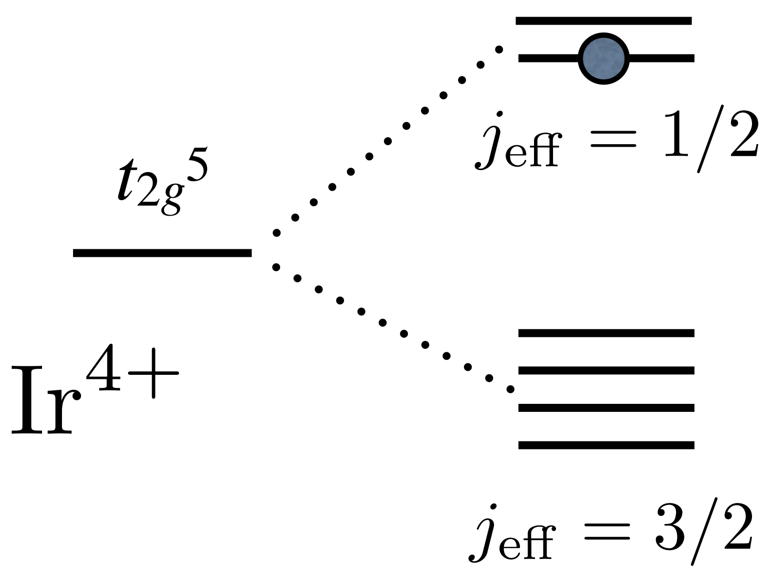


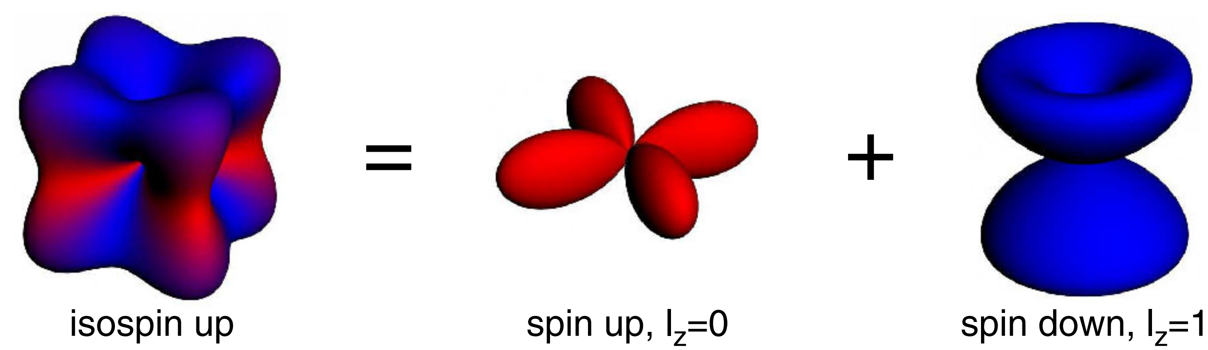
  - extensions to other lattice structures

    star lattices, hyperoctagon, etc.

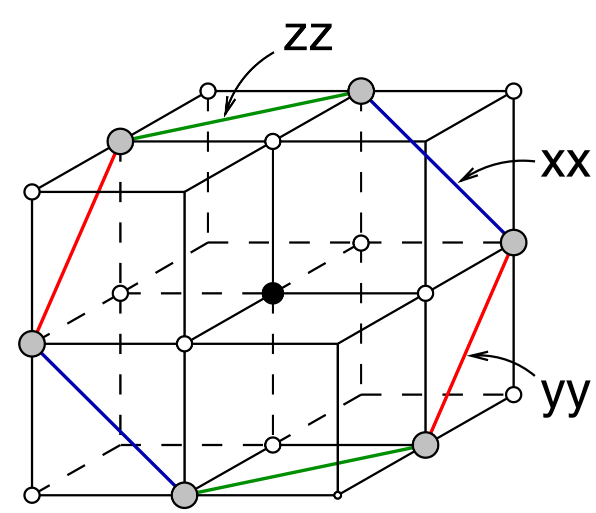
Experimental relevance?

- An effective interaction for partially-filled t_{2g} levels under strong spin-orbit coupling may become Kitaev type (G. Jackeli and G. Khaliullin, 2009).





j_{eff}=1/2 isospin



➡ experimental exploration of Kitaev spin liquids

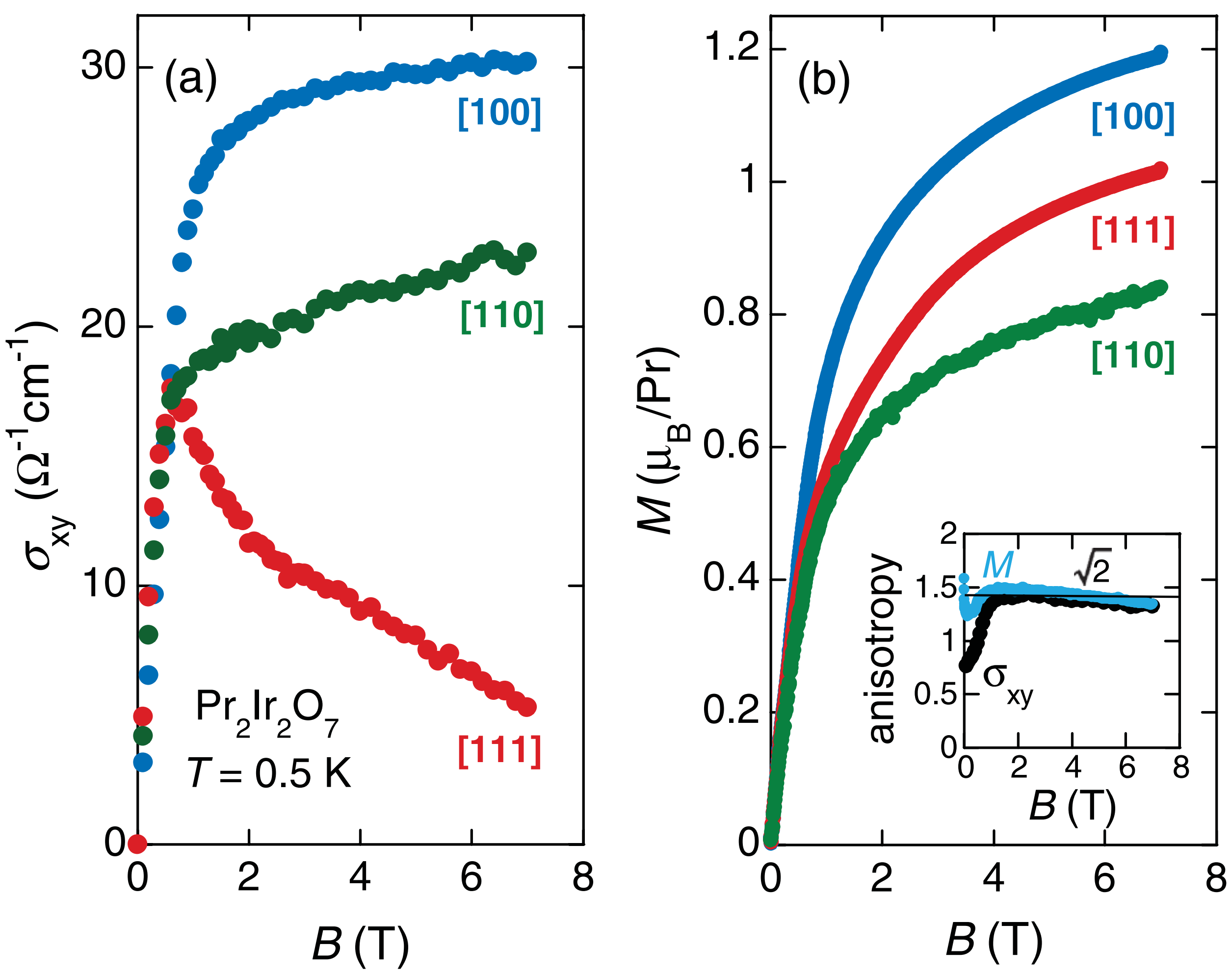
quasi-2D honeycomb compounds, Na_2IrO_3, Li_2IrO_3, ...
pyrochlore Ir_2O_4, hyperkagome $Na_4Ir_3O_8$,
hyperhoneycomb and harmonic honeycomb Li_2IrO_3, ...

no strong candidate yet (Most of them do show long-range ordering, presumably because of other interactions.)

Hybrid example

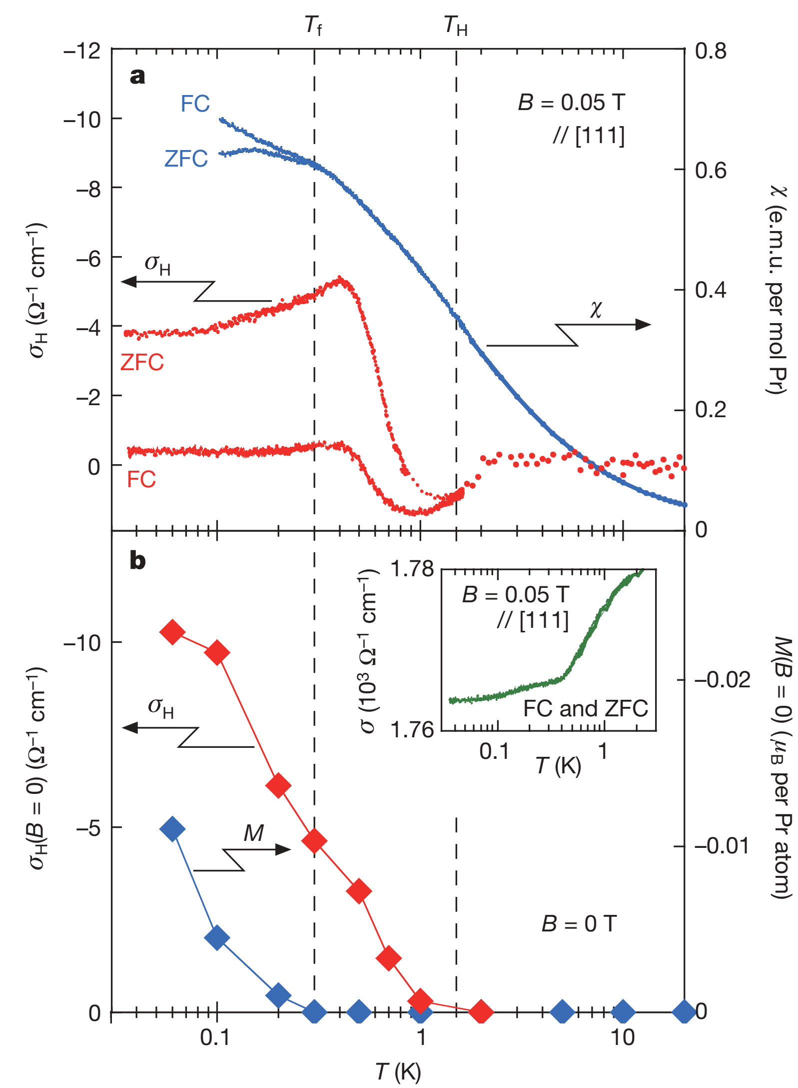
- $Pr_2Ir_2O_7$: spin ice (Pr 4*f* moments) + itinerant electrons (Ir 5*d* electrons)
  - Peculiar spin texture strongly affects the electronic and transport properties.

anomalous Hall effect dependent on the magnetic field direction



S. Nakatsuji *et al.*, 2006

spontaneous anomalous Hall effect



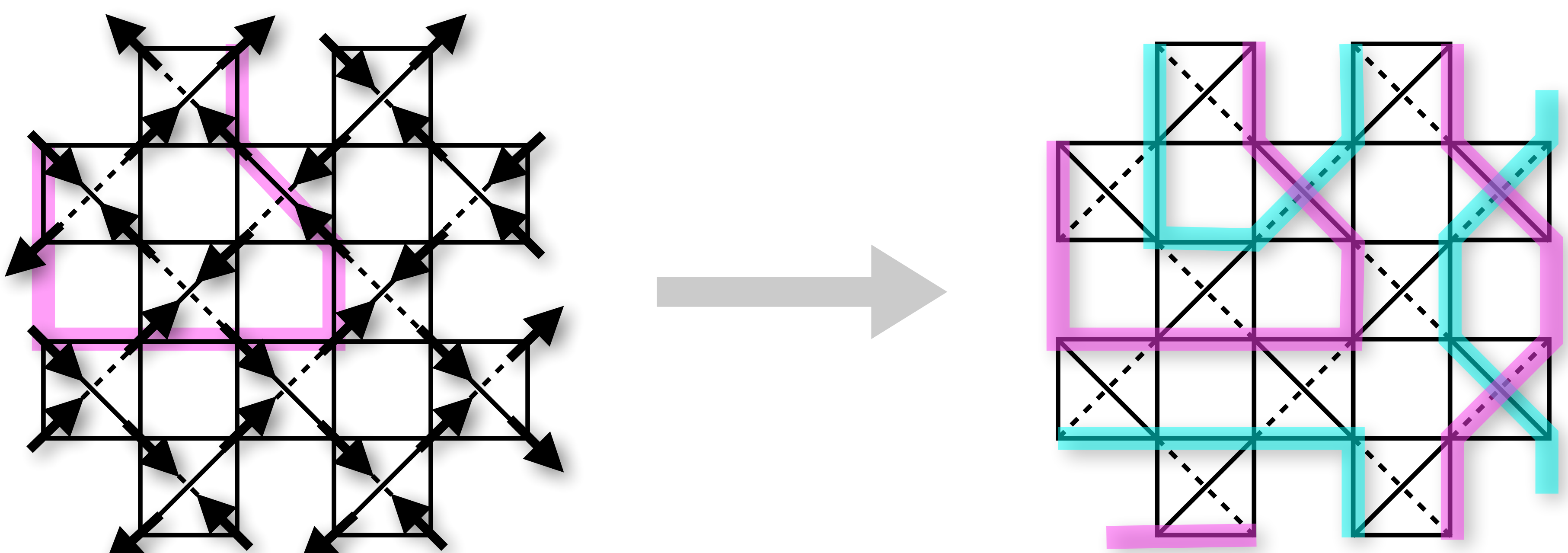
Y. Machida *et al.*, 2010

Classical spin liquids

spin ice and close-packed dimers

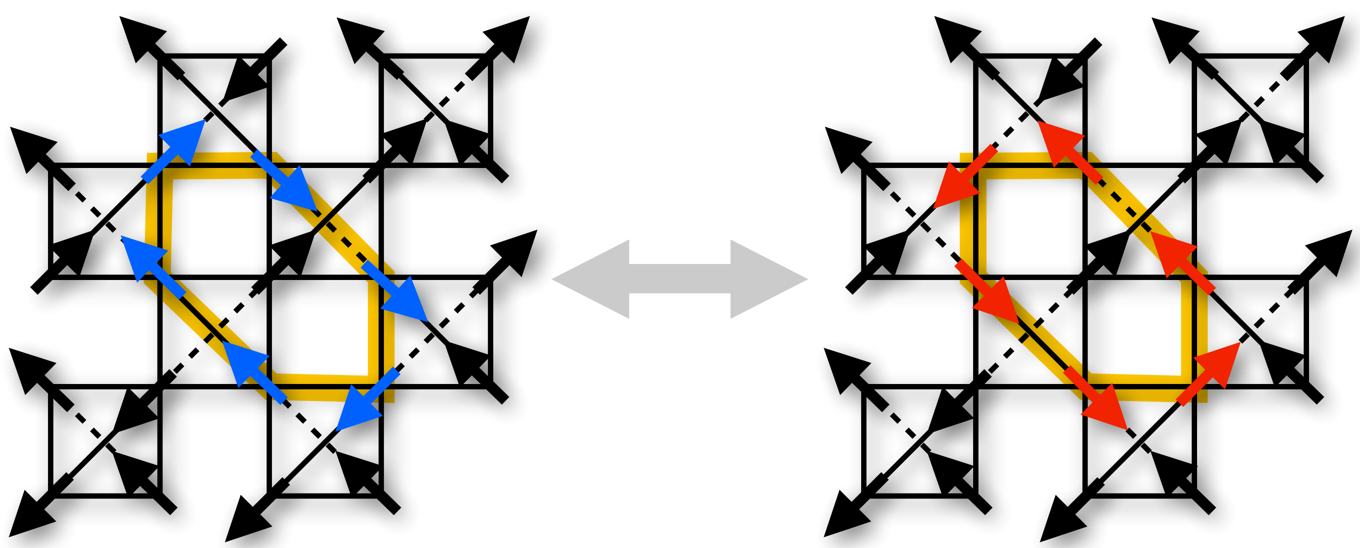
Spin ice

- local constraint = 2-in 2-out configuration in every tetrahedron (ice rule) (equivalent to six-vertex model)
  - macroscopic degeneracy: residual Pauling entropy ~30% of ln2
    - ➡ correlated disordered state dubbed as "Coulomb phase" (D. Huse *et al*., 2003; S. V. Isakov *et al.*, 2004; C. L. Henley, 2005)
  - ice rule = zero divergence condition
    - ➡ fictitious electromagnetic field, algebraic dipolar spin-spin correlations
- mapping from local spin configurations to <u>self-avoiding closed loops</u>



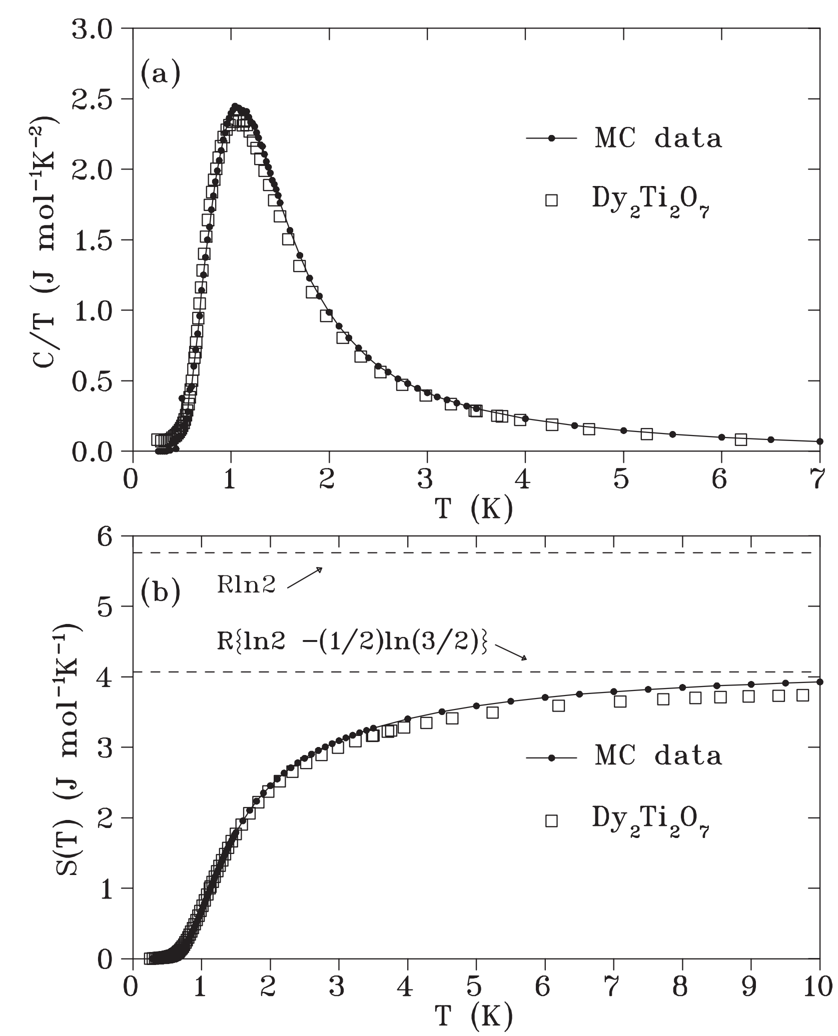
Spin ice model

- Any single spin flip makes 3-in 1-out/1-in 3-out pairs (monopoles), whose energy cost is O(*J*): strongly suppressed at low *T*<<*J*
- Global spin flips along the closed loops do not cost energy (zero modes), leaving the system within the 2-in 2-out manifold.

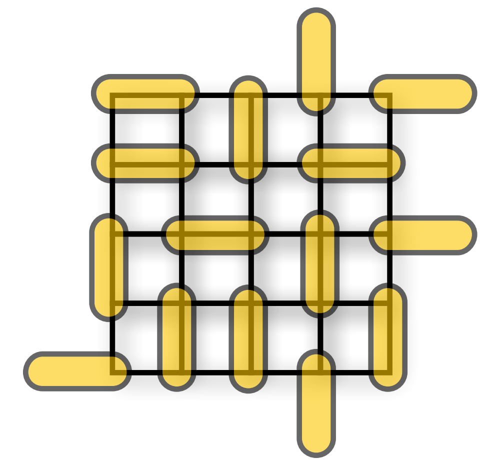


◉ efficient Monte Carlo simulation by using the global loop flips (R. G. Melko and M. J. P. Gingras, 2004)

no phase transition, just a crossover for the model with n.n. interactions only

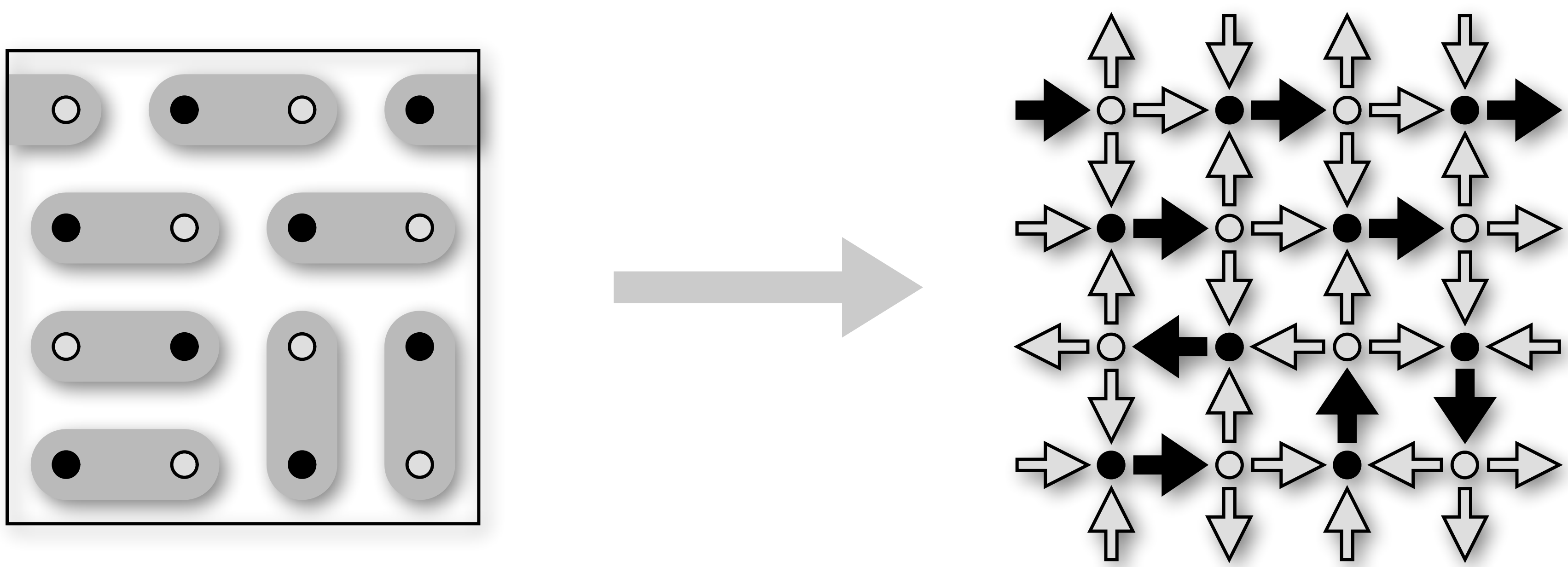


Close-packed dimers



- local constraint = every site belongs to a single dimer
  ➡ correlated disordered state dubbed as “Coulomb phase”:
  algebraic dimer-dimer correlations (D. Huse *et al*., 2003)

- mapping from dimers to fictitious field (D. Huse *et al.*, 2003)



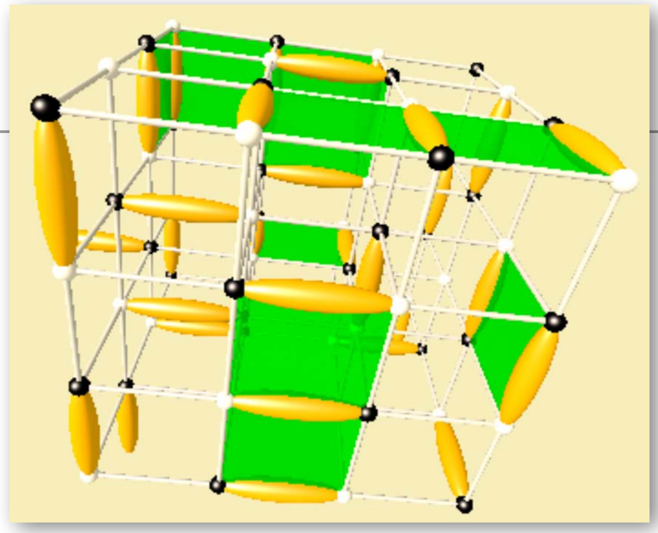
$n_\mu(\mathbf{r}) = 0 \text{ or } 1$ $\qquad$ $E_\mu(\mathbf{r}) = (-1)^{\mathbf{r}} n_\mu(\mathbf{r})$

➡ Coulomb phase = zero flux state of fictitious field (divergence free)

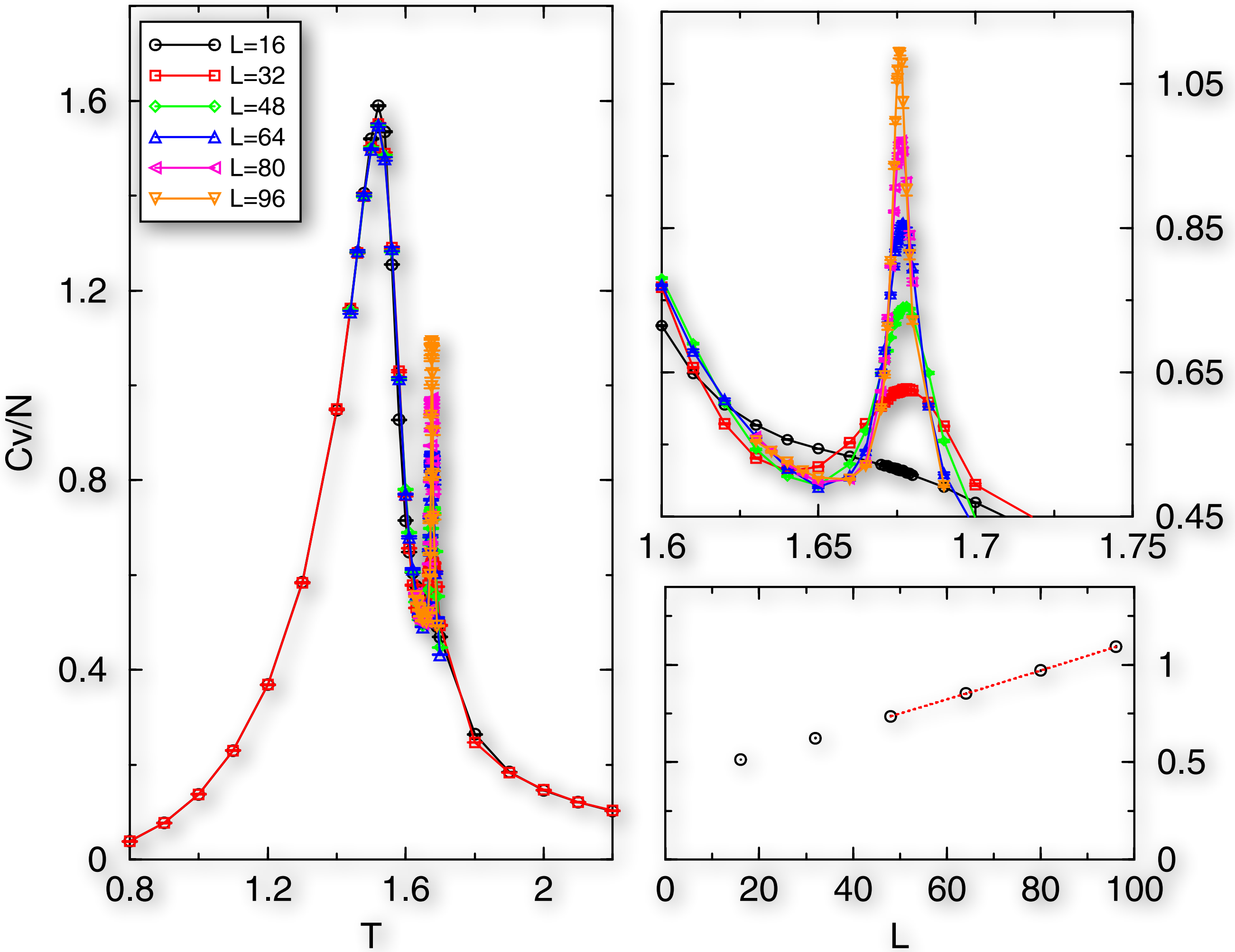
Close-packed dimer model

$$\mathcal{H} = -\sum_{\square}(n_{\parallel} + n_{=} + n_{/\!/})$$

$n_{\parallel} = 1$ for [image of a plaquette with two parallel dimers] otherwise 0, etc.

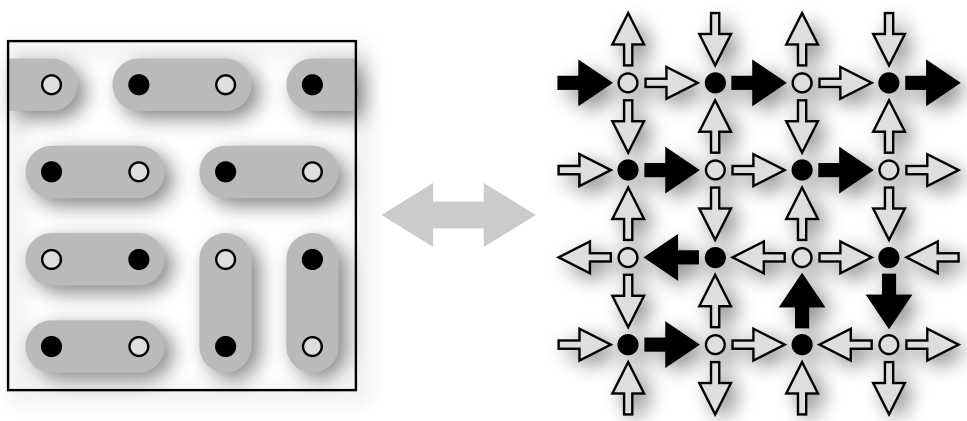


- low-T: columnar ordered phase
  translational and cubic symmetries are broken
- high-T limit: Coulomb phase with algebraic dimer-dimer correlations
  (D. Huse *et al.*, 2003)
- unconventional phase transition compatible with tricritical universality class (F. Alet *et al.*, 2006; D. Charrier and F. Alet. 2010)



Close-packed dimer model

- Coulomb phase = zero flux state of fictitious field (D. Huse *et al.*, 2003)

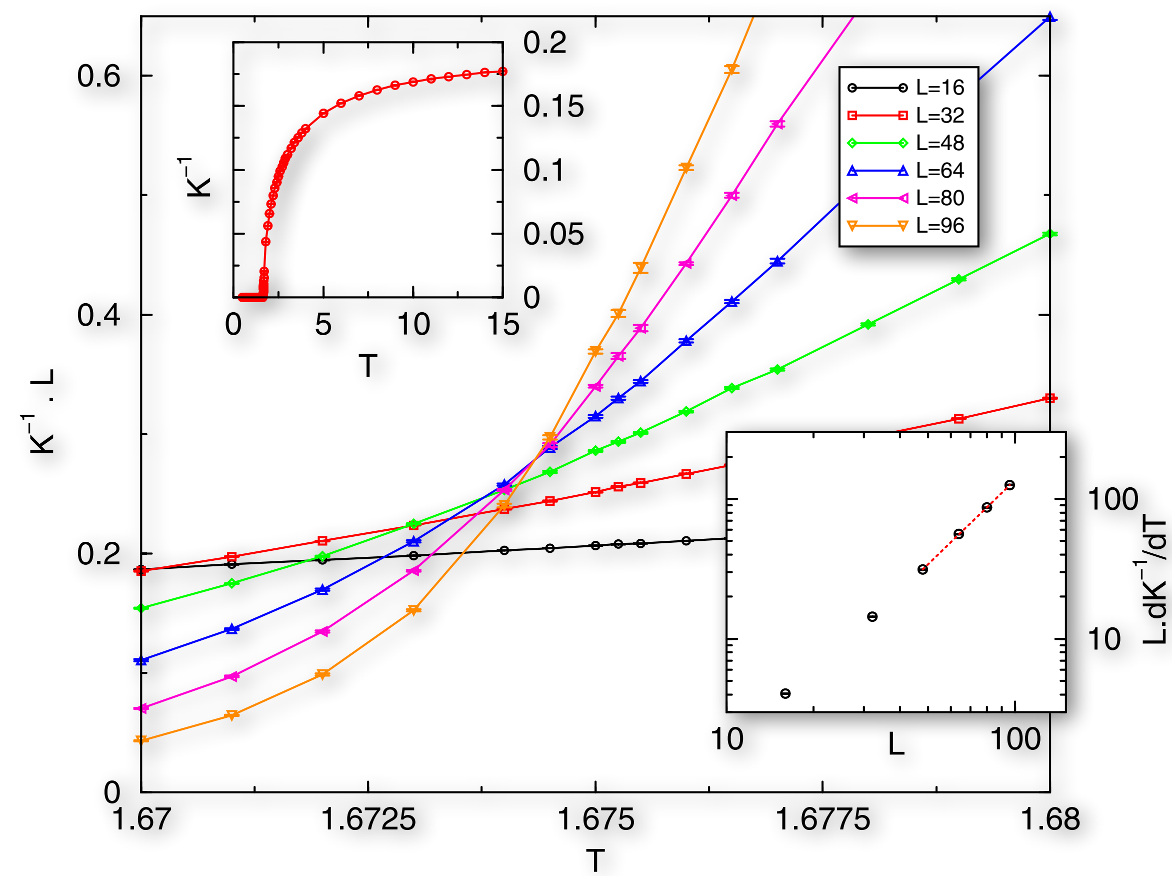


$n_\mu(\mathbf{r}) = 0 \text{ or } 1$ $\qquad$ $E_\mu(\mathbf{r}) = (-1)^{\mathbf{r}} n_\mu(\mathbf{r})$

➡ characterization by flux fluctuations

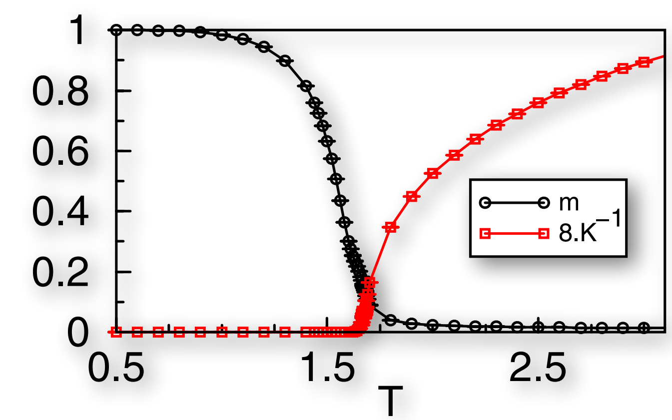
$$K^{-1} = \frac{\langle \phi^2 \rangle}{L} = \frac{1}{3L} \sum_{\mu=x,y,z} \langle \phi_\mu^2 \rangle$$

$$\phi = \int (-1)^{\mathbf{r}} \mathbf{n}(\mathbf{r}) \cdot d\mathbf{S}$$



F. Alet *et al.*, 2006

phase transition is signaled by
- columnar order parameter below T_c
- flux fluctuations for Coulomb phase above T_c

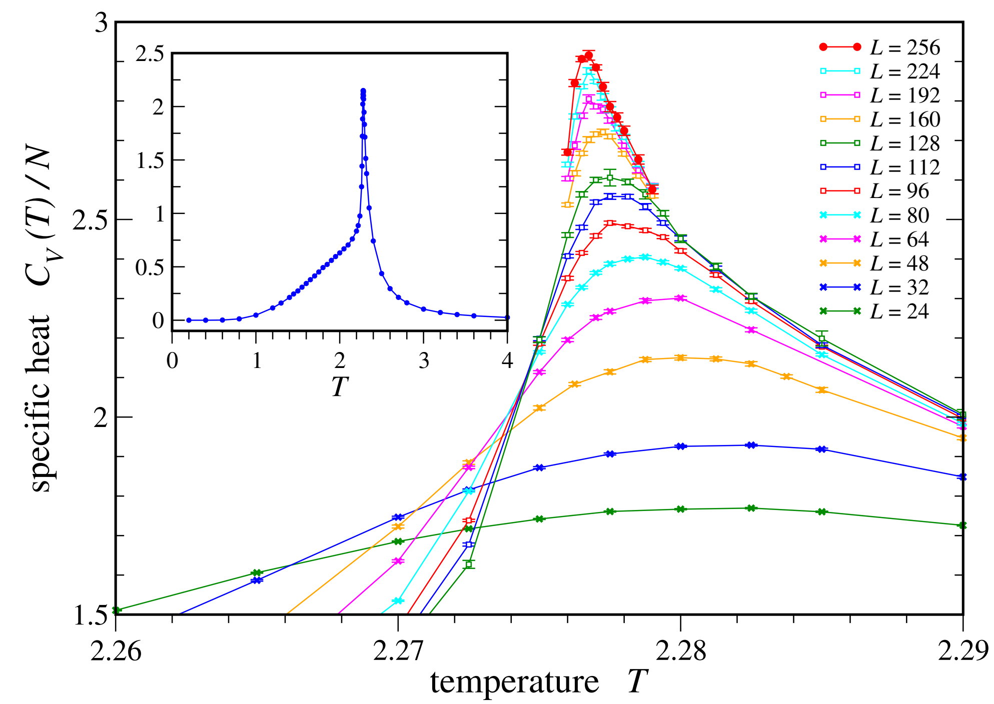


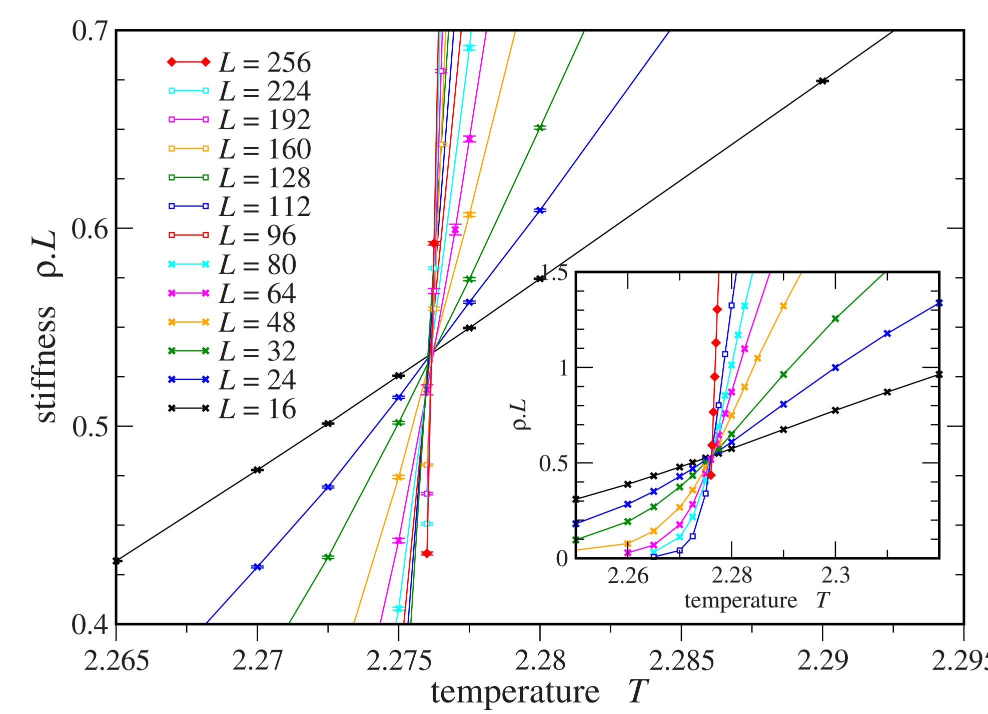
Close-packed dimer model: variants

$$\mathcal{H} = -\sum_{\square} n_{\parallel}^{\text{even}}$$

Translational and cubic symmetries are both already broken.

- **low-T: columnar "order"**
  no further breaking of the symmetry of the system
- **high-T: Coulomb phase with algebraic dimer-dimer correlations**
  non-zero flux fluctuations
- **phase transition compatible with 3D XY universality class** (G. Chen *et al.*, 2009)





Intermediate (hybrid)

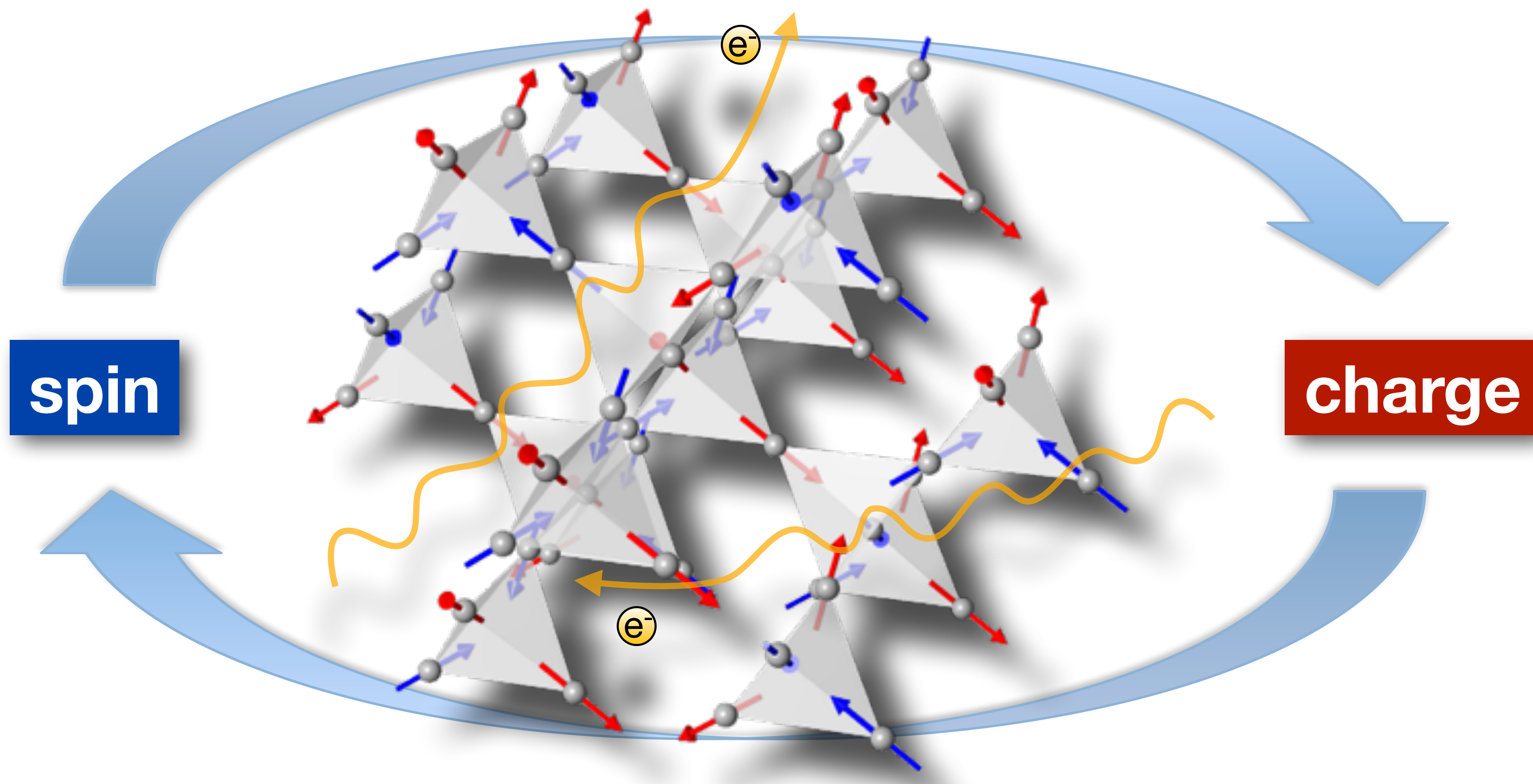
localized moments + itinerant electrons

loop liquid and scalar chiral liquid

Spin-charge coupling

internal field from peculiar magnetic texture

➡ exotic electronic and transport properties

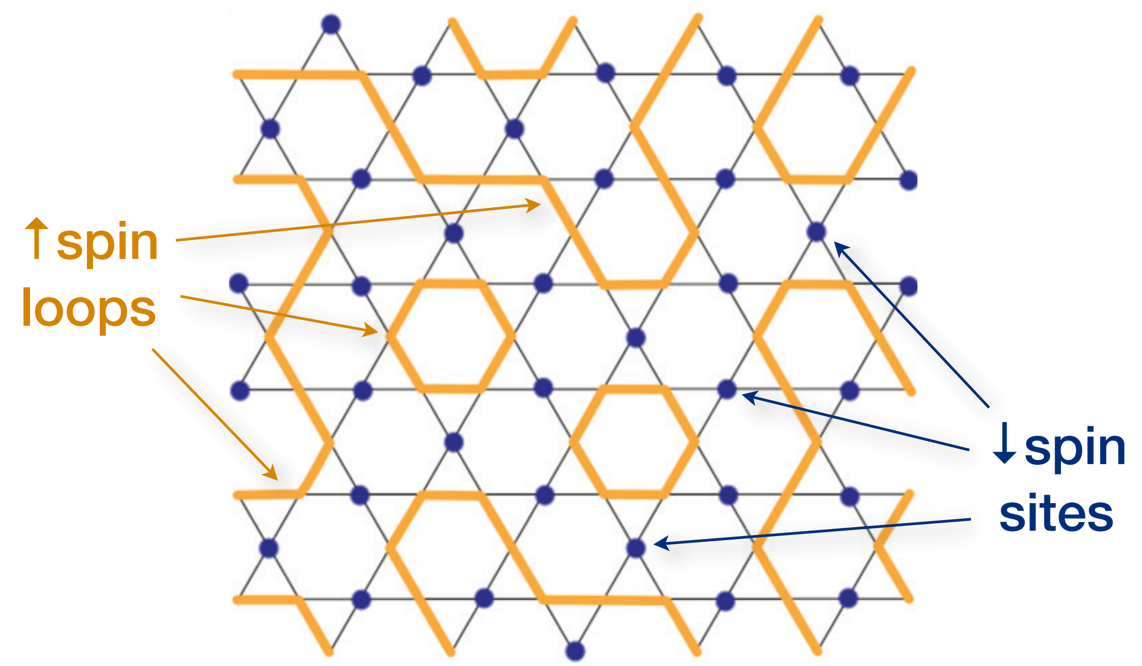


effective magnetic interactions

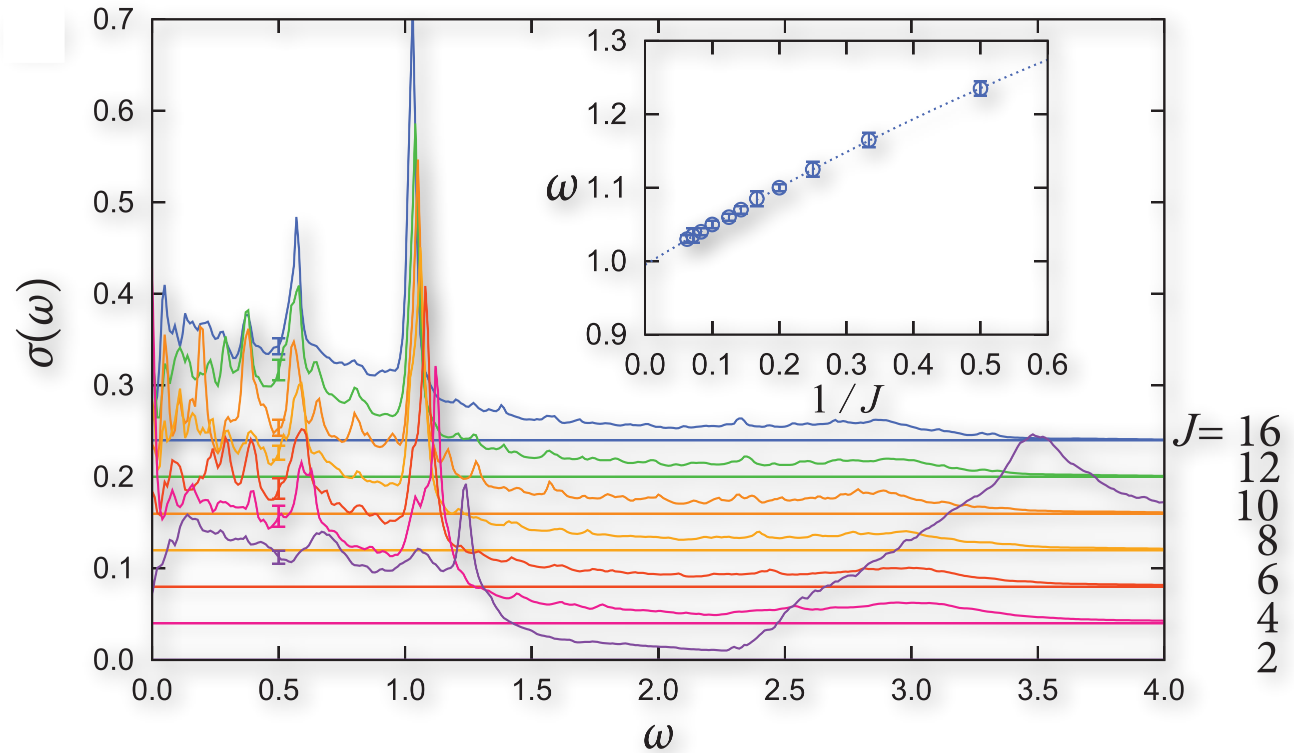
➡ reconstruction of magnetic structure

Loop liquid in kagome ice

- classical spin liquid state with 2up-1down local configuration (ferrimagnetic state)
  - ➡ ↑spin loops + isolated ↓spin sites

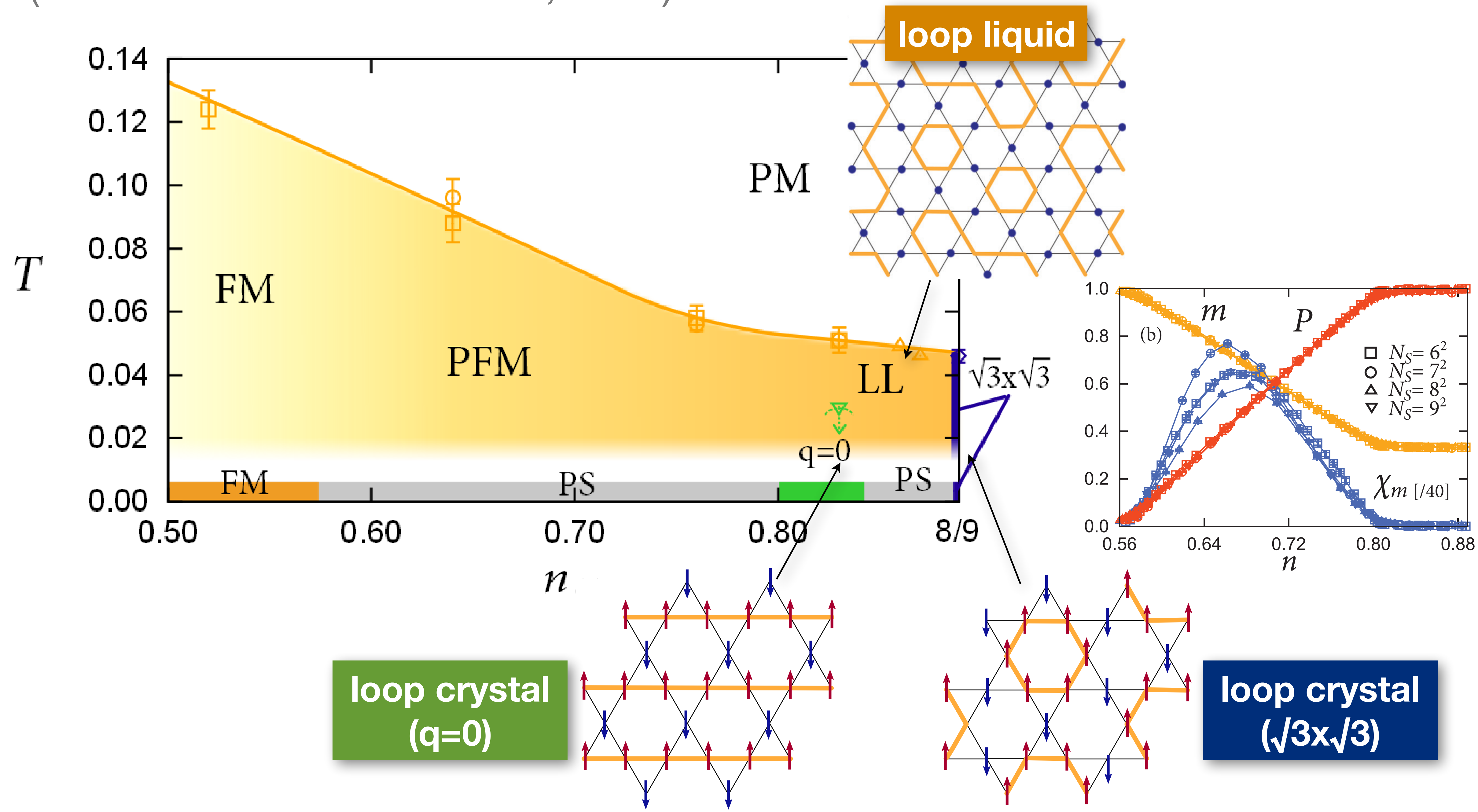


- Itinerant electrons come into the ↑spin loops to gain kinetic energy.
  - ➡ free electrons in closed 1D loops (J. Jaubert *et al.*, 2012)
  - ➡ resonating peaks in DOS and optical conductivity (H. Ishizuka and Y. Motome, 2013)



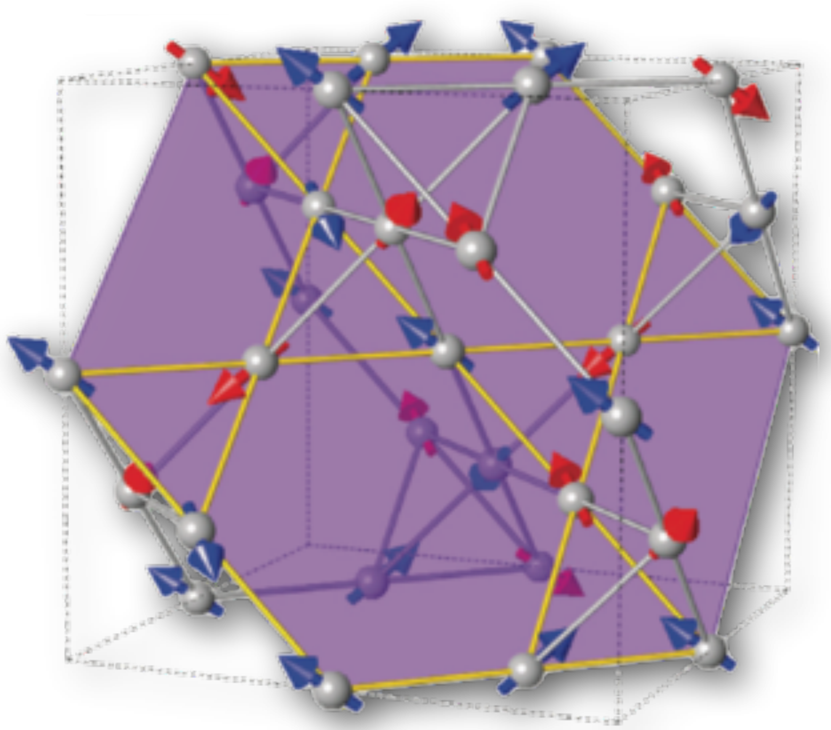
Loop liquid and crystals

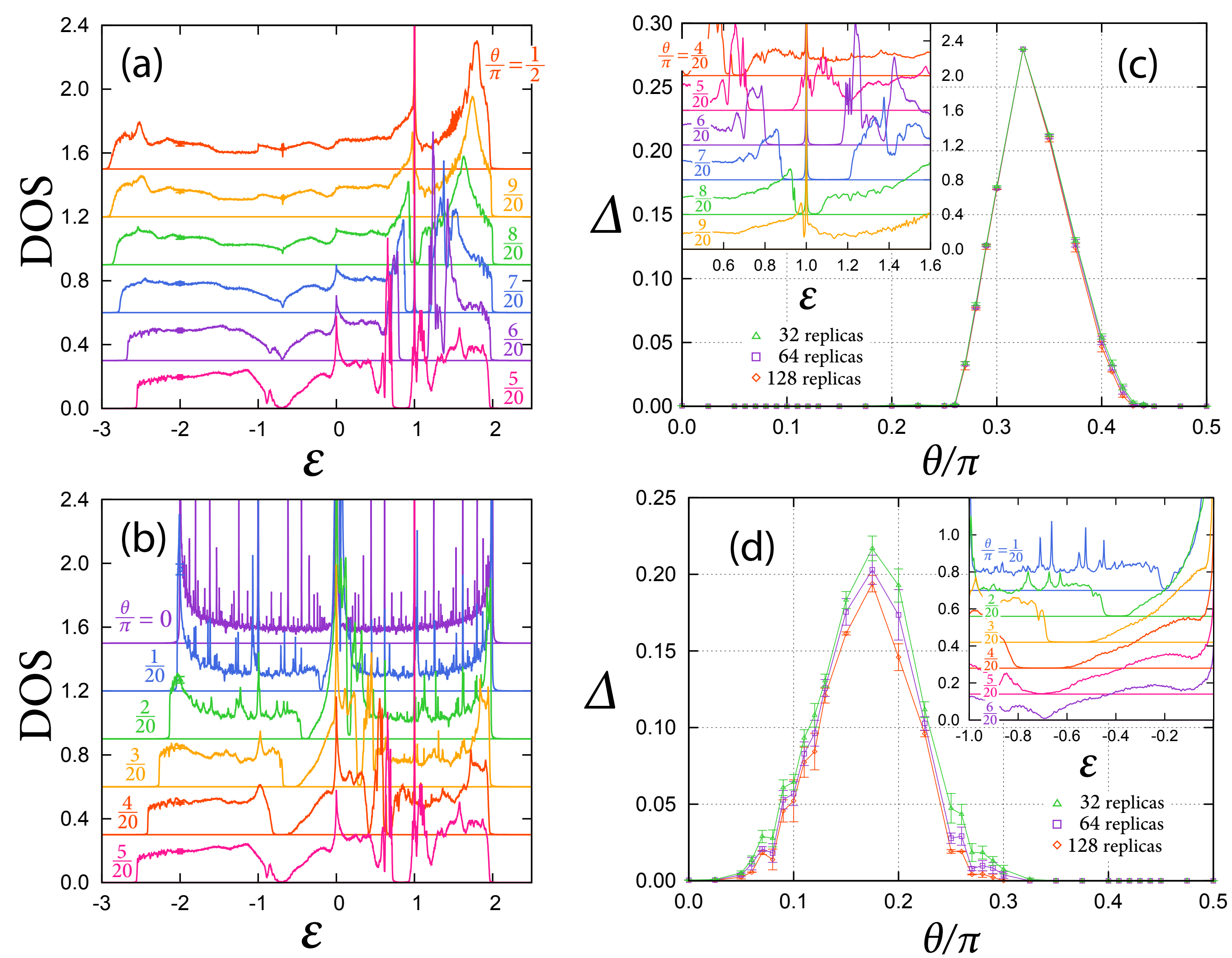
- Monte Carlo simulation for a Kondo lattice model with Ising spins
  (H. Ishizuka and Y. Motome, 2013)



Insulating kagome ice: scalar chiral liquid

- kagome ice with spin-ice type noncoplanar spin configuration: magnetically disordered but scalar chirality ordered $\sum_{\triangle}\langle \mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k)\rangle \neq 0$
  - ➡ Charge gap opens at *n*=1/3 and 2/3 (H. Ishizuka and Y. Motome, 2013; 2014)
  - ➡ quantum anomalous Hall effect (H. Ishizuka and Y. Motome, 2013)

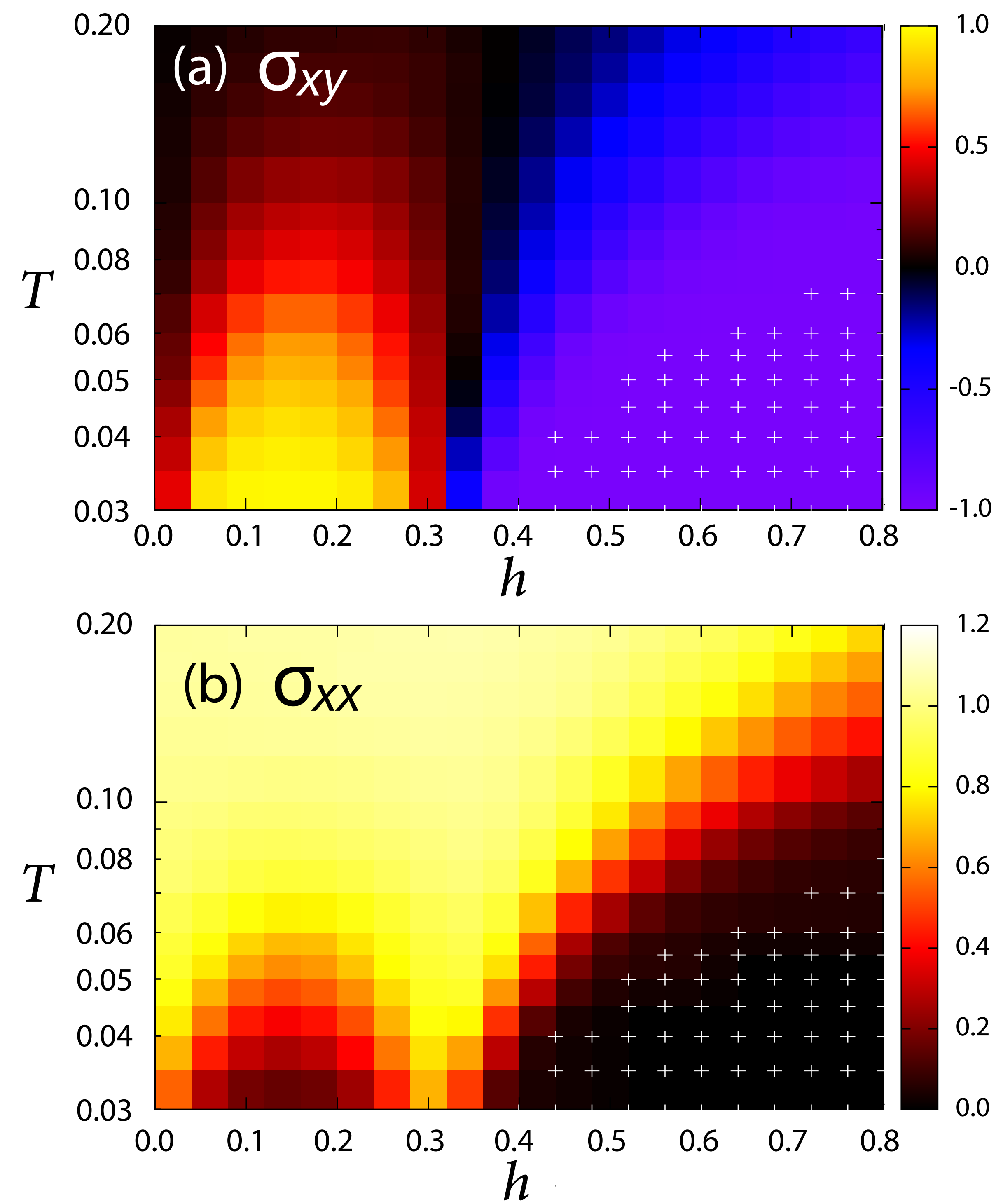




Insulating kagome ice: scalar chiral liquid

- Monte Carlo simulation for a Kondo lattice model with spin-ice type noncoplanar Ising spins ([111] plane of the pyrochlore spin ice)
  (H. Ishizuka and Y. Motome, 2013)

  - $0 \leq h \leq 0.3$: kagome ice insulator
    ➡ anomalous Hall effect with $\sigma_{xy} \sim +1$
  - $h \geq 0.3$: 3out insulator
    ➡ anomalous Hall effect with $\sigma_{xy} \sim -1$
  - critical point at $h \sim 0.3$?



Scalar chiral liquid on a triangular lattice

- scalar chiral order with 4-sublattice noncoplanar spin texture (I. Martin and C. D. Batista, 2008; Y. Akagi and Y. Motome, 2010)
  - ➡ Chern insulator at n=1/4 and 3/4
  - ➡ quantum Hall effect

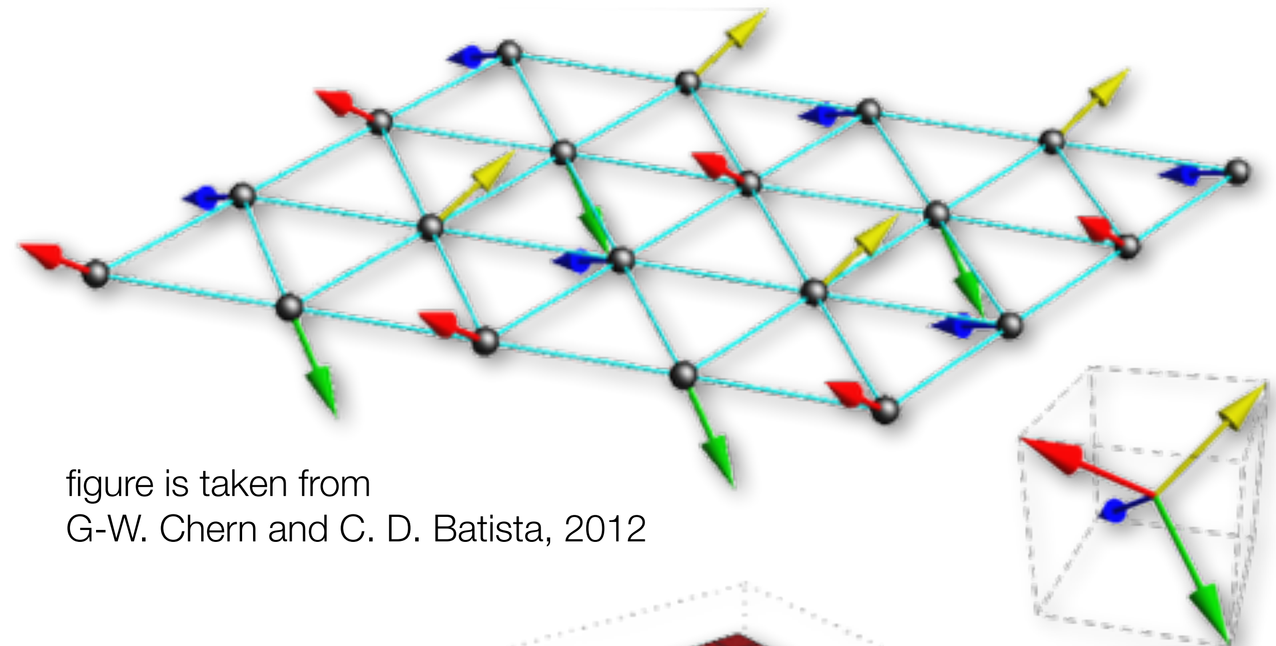
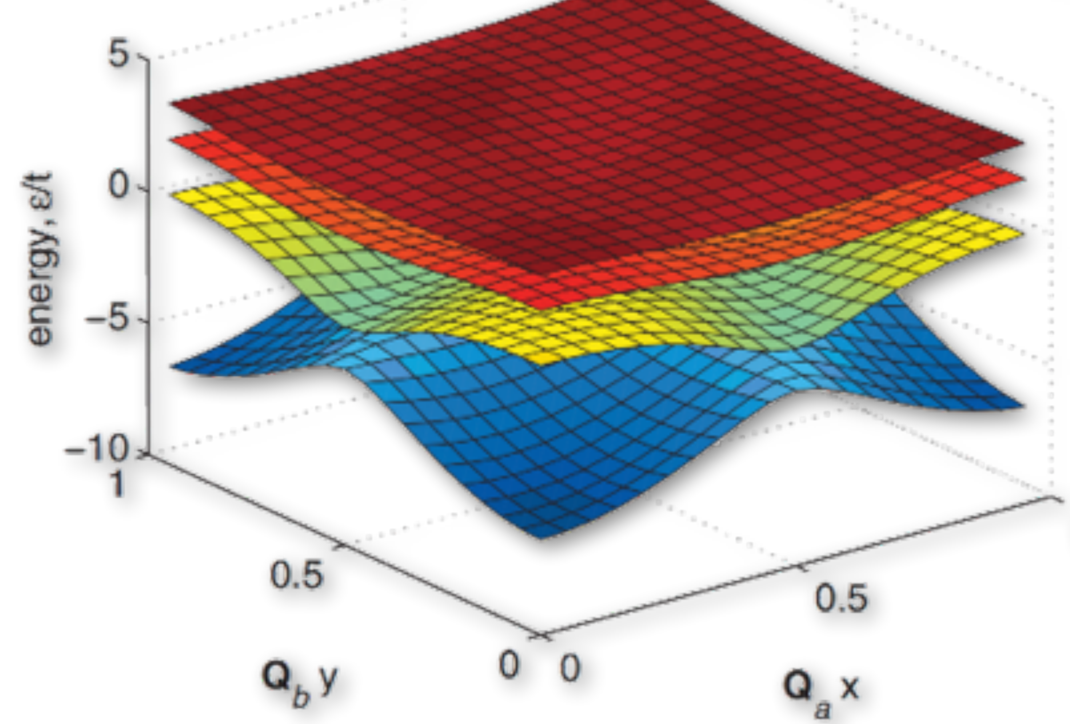


figure is taken from
G-W. Chern and C. D. Batista, 2012

- thermal fluctuations
  - ➡ scalar chiral liquid at nonzero T (Y. Kato *et al.*, 2010)

- quantum fluctuations
  - ➡ scalar chiral liquid at T=0? (Y. Akagi and Y. Motome, 2013; S. Jiang *et al*., 2014)



I. Martin and C. D. Batista, 2008

Quantum spin liquids

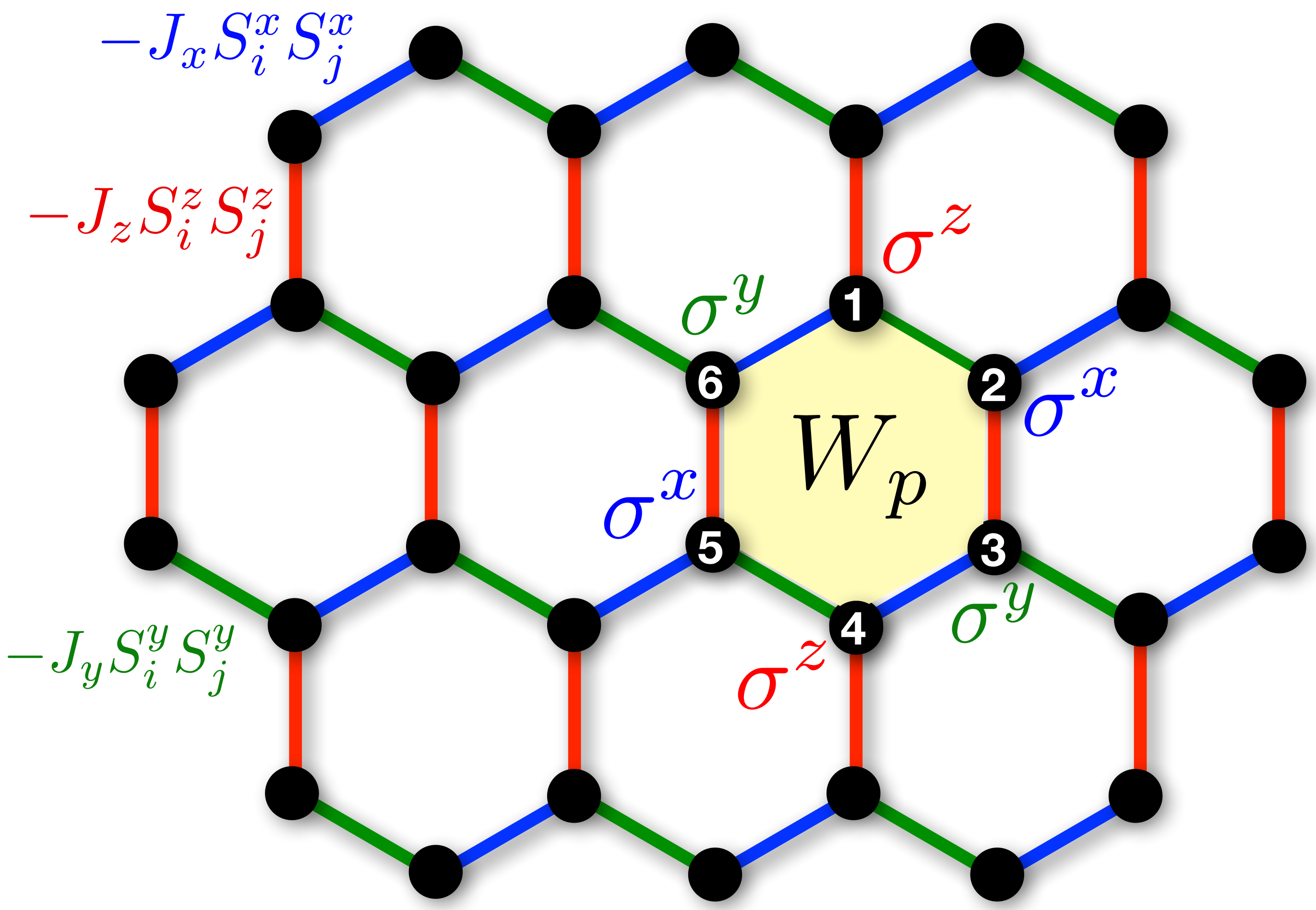
3D Kitaev model

- local conserved quantity
- local constraint from 3D lattice structure
- emergent loops: finite-T spin-liquid transition

Kitaev model

- S=1/2 quantum spin model on a 2D honeycomb lattice (A. Kitaev, 2006)

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



bond dependent interactions ➡ *frustration*

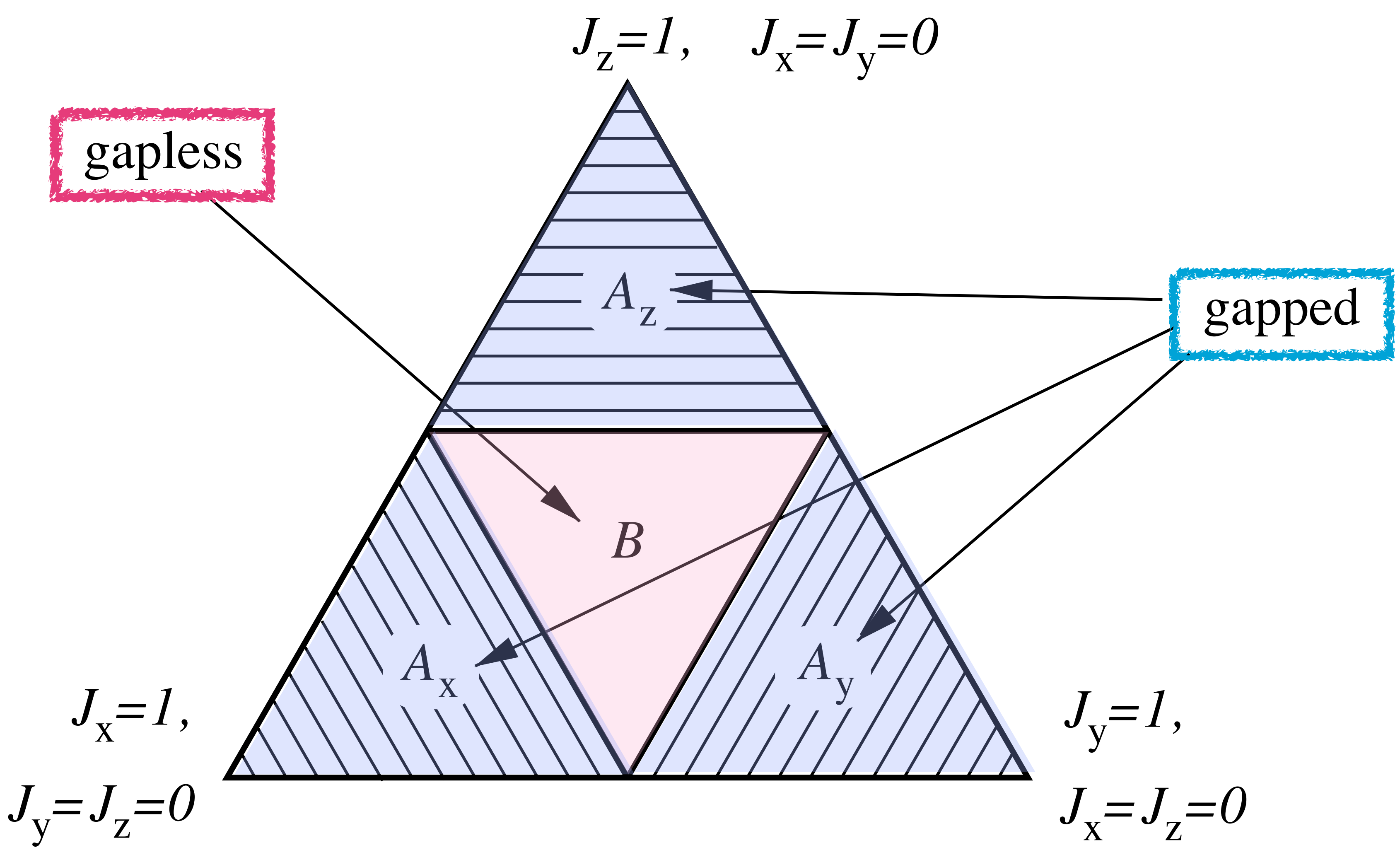
local conserved quantity

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

- ✓ $[\mathcal{H}, W_p] = 0$
- ✓ $[W_p, W_p'] = 0$ for $p \neq p'$
- ✓ $W_p^2 = 1$

Eigenstates are labelled by Z_2 variables $\{W_p = \pm 1\}$

Kitaev model: T=0 phase diagram



QSL ground states in the entire parameter region:
gapless and gapped QSLs depending on the anisotropy

topological order, extremely short-range spin correlation, non-abelian anyons, quantum computation, ...

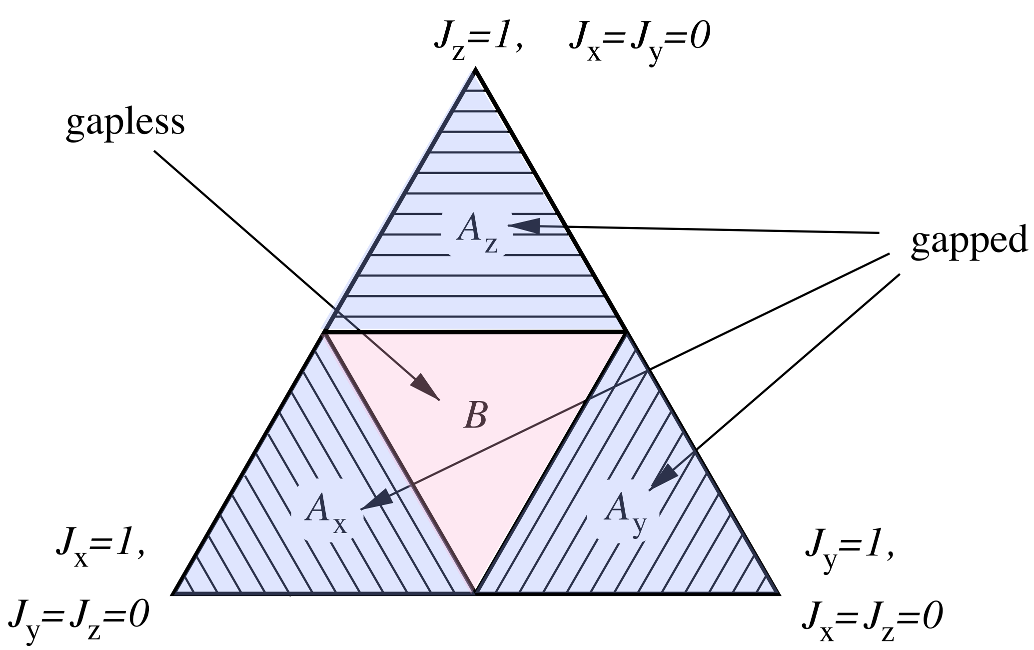
A. Kitaev, 2006; G. Baskaran, S. Mandal, and R. Shanker, 2007; C. Castelnovo and C. Chamon, 2007; Z. Nussinov and G. Ortiz, 2008, ...

3D extension of the Kitaev model

- 3D hyperhoneycomb lattice (S. Mandal and N. Surendran, 2009)

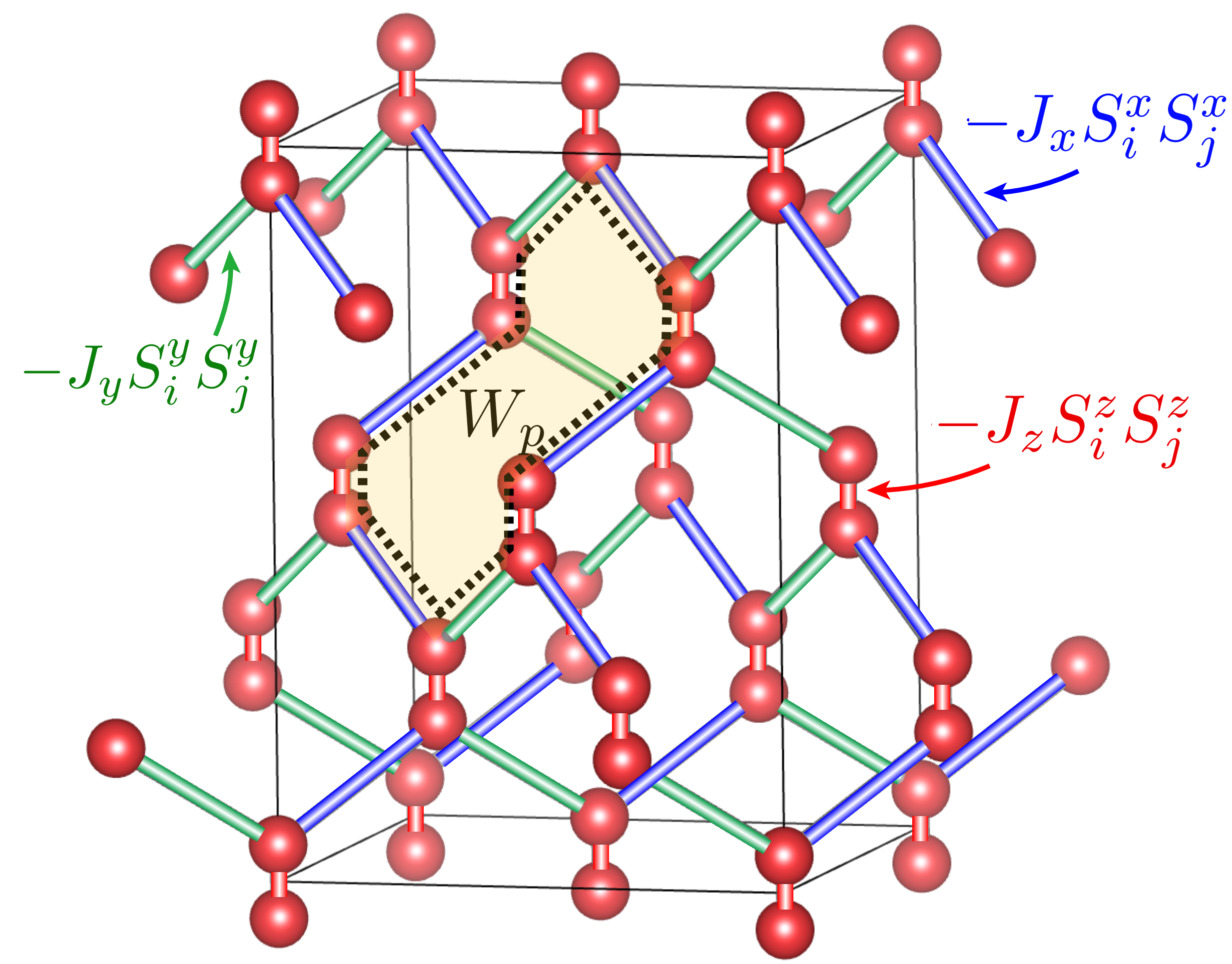
$$\mathcal{H} = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z$$

- The model inherits the solvability.



exactly the same T=0 phase diagram

QSL ground states in 3D



experiment: new Iridates β-Li_2IrO_3 and γ-Li_2IrO_3
(T. Takayama *et al.*, 2014; K. A. Modic *et al.*, 2014)

Method

- The conventional quantum Monte Carlo (QMC) methods on the basis of the world-line technique do not work because of the negative-sign problem:
  - Lattices are bipartite, but the interactions are frustrated.

- Our solution (J. Nasu, M. Udagawa, and Y. Motome, 2014):

interacting S=1/2 spin models

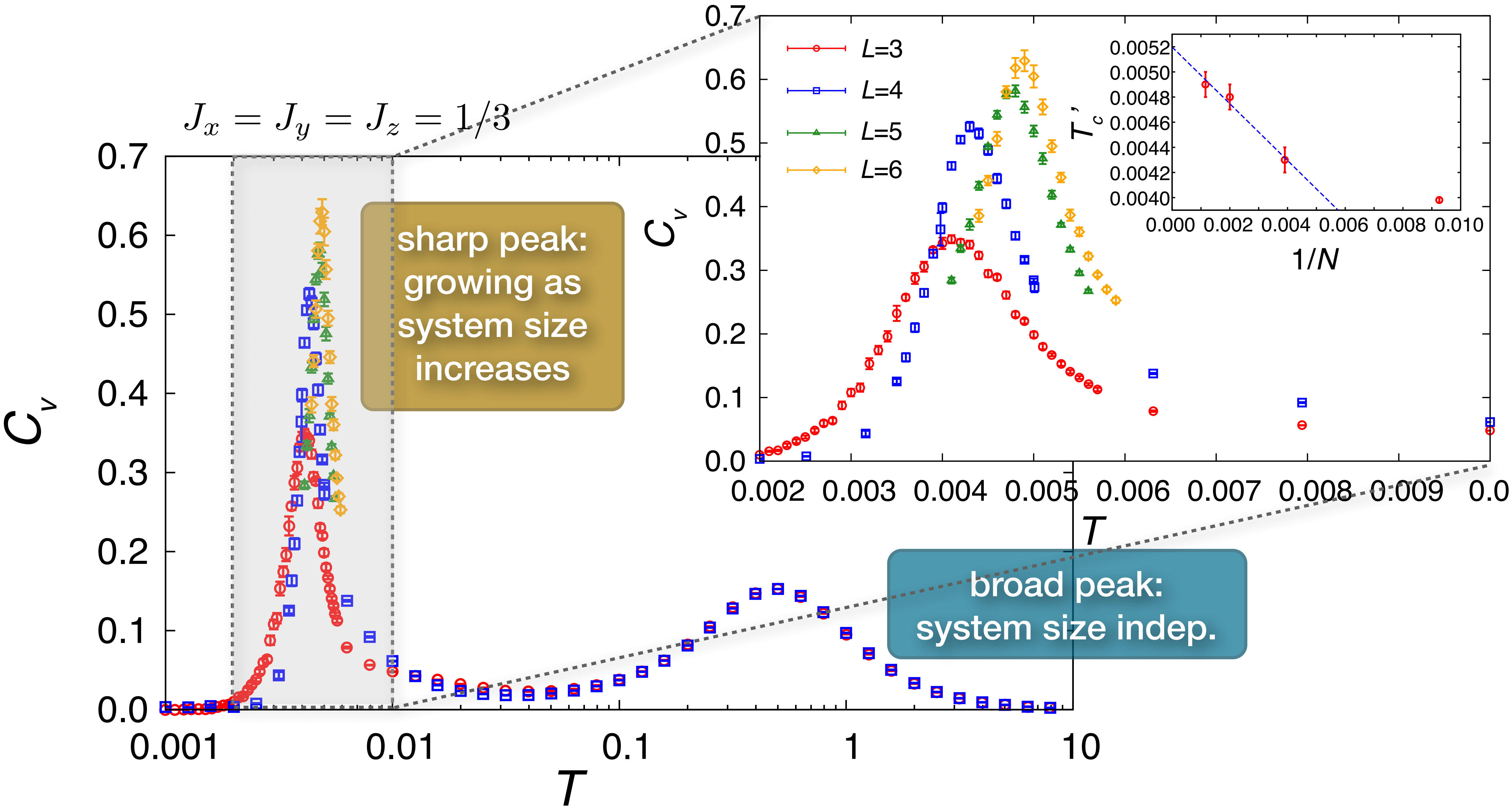
↓ Jordan-Wigner transformation
Majorana fermion representation

non-interacting Majorana fermions coupled to thermally-fluctuating Z_2 variables

↓

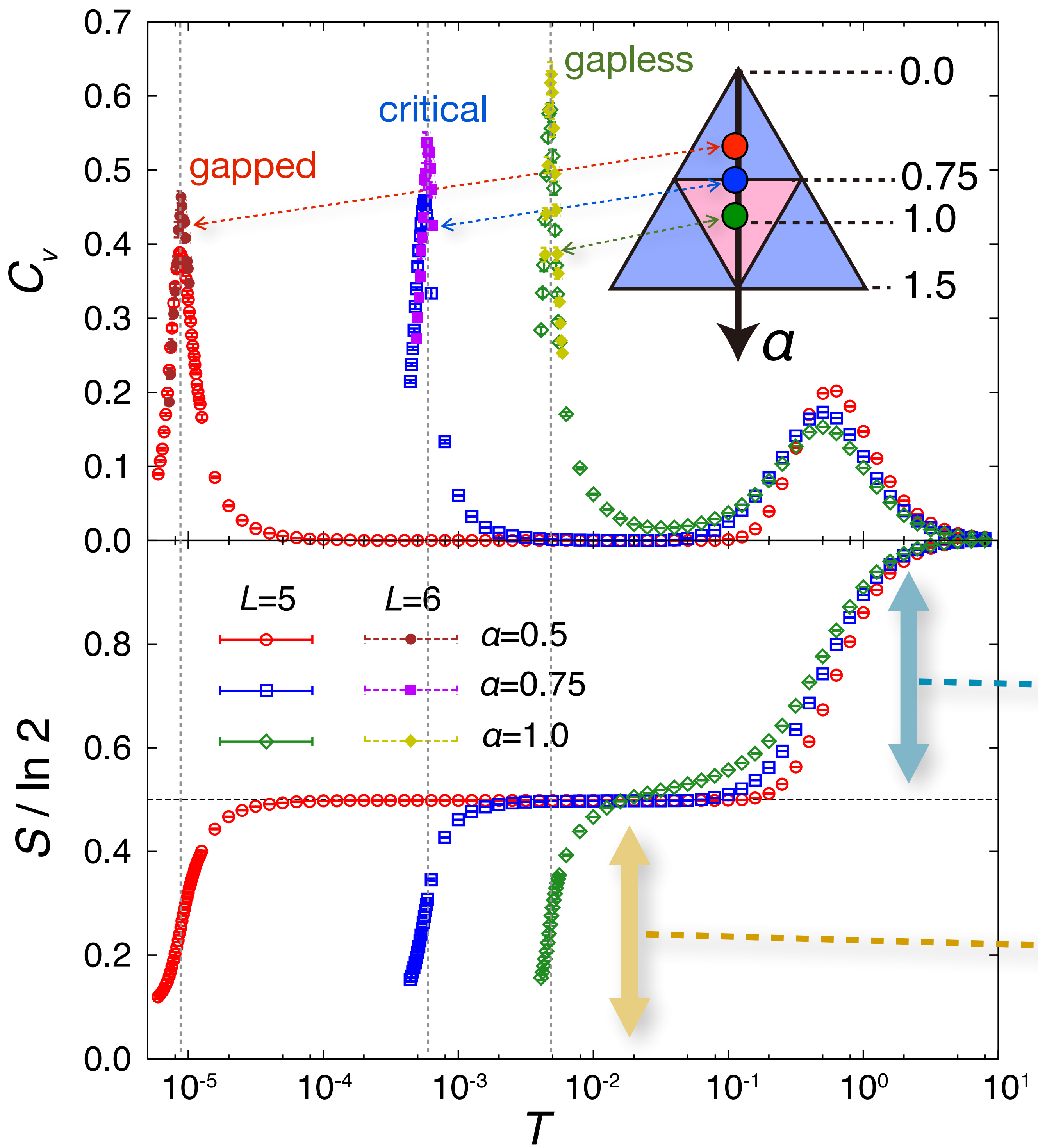
QMC free from negative-sign problem

Specific heat in the isotropic case



strong sign of phase transition at T_c~0.0052 (and crossover at T^*~0.5)

Separation of two energy scales



S=1/2 spin ➡ two Majorana fermions

$\mathbf{S}_i$ → c_i : free Majorana fermions
$\mathbf{S}_i$ → $\bar{c}_i$: Z_2 variables η_r

$$\mathcal{H} = \mathrm{i}J_x \sum_{x\text{ bonds}} c_w c_b - \mathrm{i}J_y \sum_{y\text{ bonds}} c_b c_w + J_z \sum_{z\text{ bonds}} \underbrace{\bar{c}_b \bar{c}_w}_{-\mathrm{i}\eta_r} c_b c_w$$

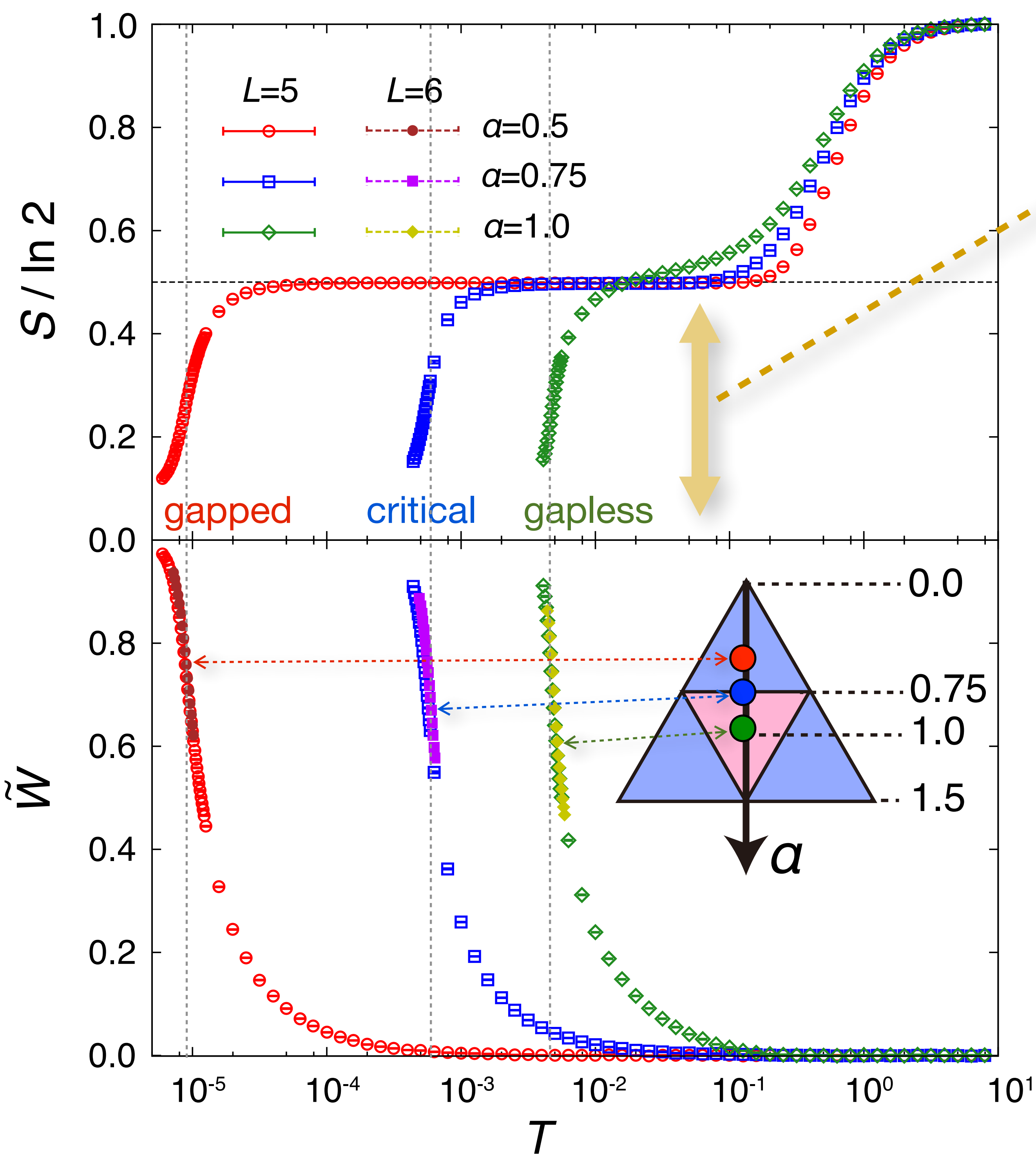
itinerant Majorana fermions c_i

independent of system sizes and anisotropy
➡ crossover at T^*

localized Majorana fermions $\bar{c}_i$

dependent of system sizes and anisotropy
➡ phase transition at T_c

Anomaly at T_c

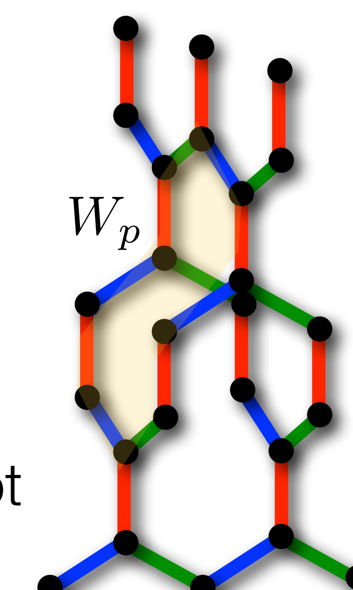


entropy release in localized Majorana fermions

coherent growth of local conserved quantity W_p

$$\tilde{W} = \frac{1}{N_p}\sum_p \langle W_p \rangle$$

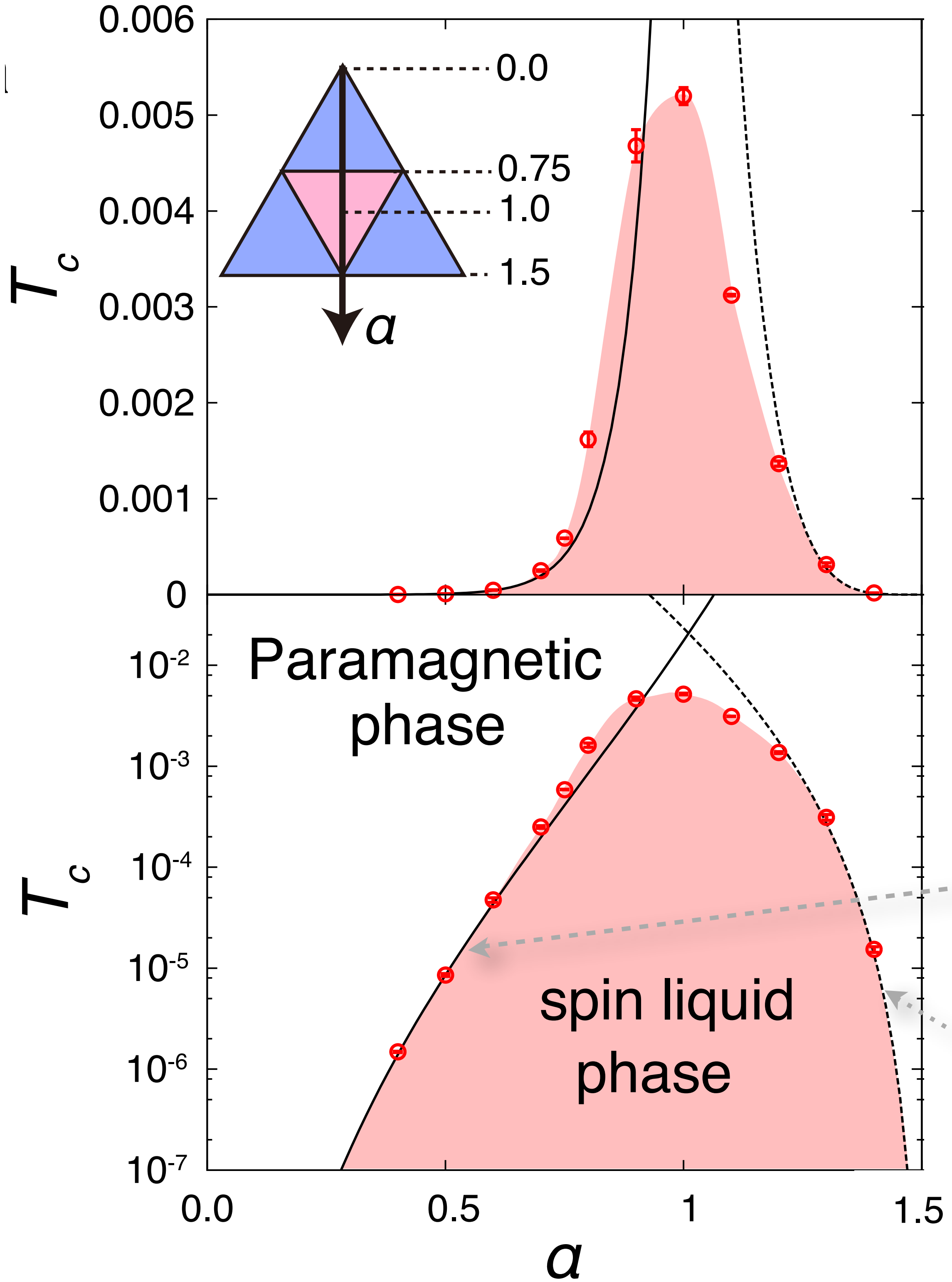
$$W_p = \prod_{r \in p} \eta_r$$



NB. $\tilde{W}$ is not an order parameter, as it does not become zero above T_c.

topological change in flipped W_p
(discussed later)

Finite-T phase diagram



- All low-T SL states are separated from high-T para by the phase transition.
  - Both gapped and gapless spin liquids remains as stable phases at finite T.
  - no adiabatic connection to para
- T_c is maximized for the isotropic case.
  - Frustration stabilized spin liquids.

- perturbation from the limit of $J_z >> J_x, J_y$ (3D toric code limit)

$$T_c = 1.925(1) \times \frac{7}{256} \frac{J^6}{J_z^5}$$

Monte Carlo estimate (later)

- perturbation from the limit of $J_z << J_x, J_y$ (weakly coupled 1D chains)

$$T_c \propto \frac{J_z^4}{J^3}$$

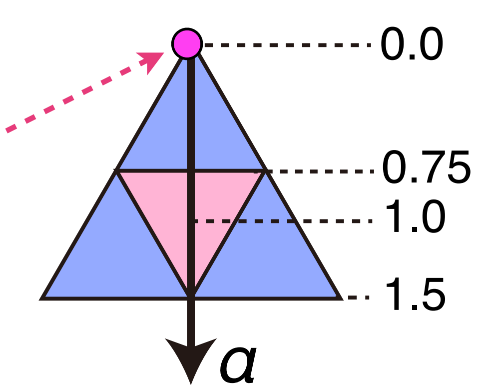
What is this phase transition?

- no anomalies in the local quantities, such as the conserved Z_2 variables W_p and nearest-neighbor spin-spin correlations

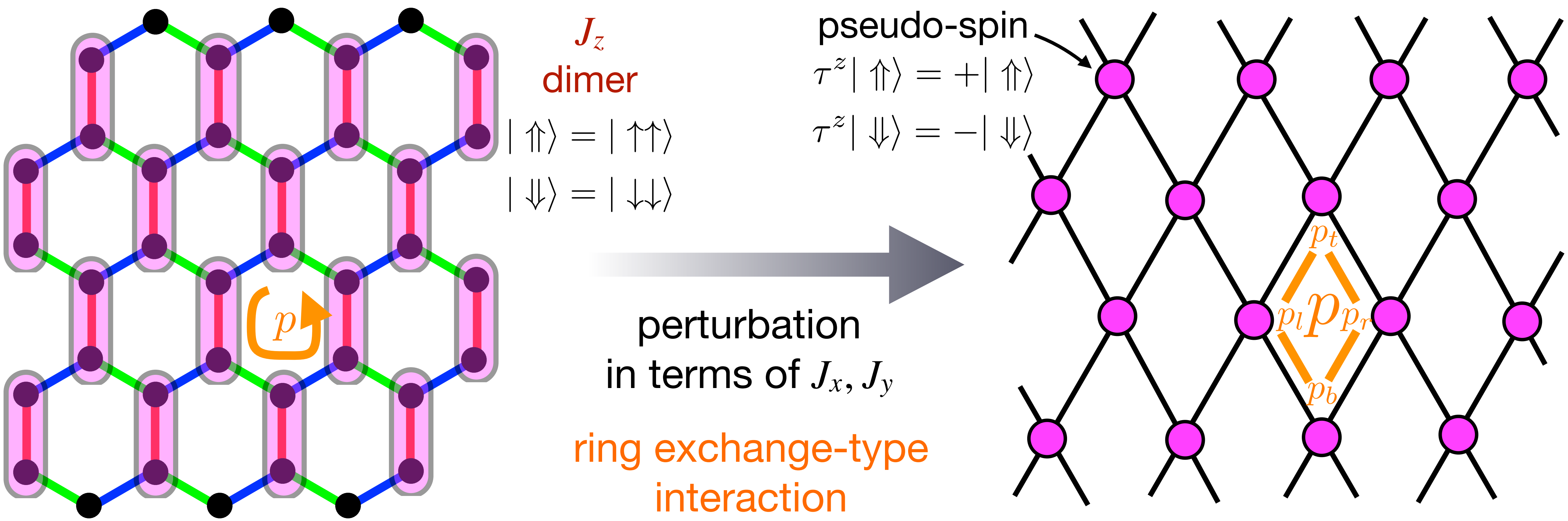
- topological transition?
  - controversy on the existence of quantum topological transition at a finite T (C. Castelnovo and C. Chamon, 2007; Z. Nussinov and G. Ortiz, 2008)

  ➡ discussed in the anisotropic limit (= toric code)

➡ What about our case?: close look into the anisotropic limit



Anisotropic limit of $J_z >> J_x, J_y$ in 2D (toric code)



- effective Ising-type model
  - eigenstates are labeled by $B_p = \pm 1$
  - QSL ground state with topological order
  - extremely short-range correlation
  - no phase transition at finite temperature

$$\mathcal{H}_{\text{eff}} = -J \sum_p B_p \qquad J \propto \frac{J_x^2 J_y^2}{J_z^3}$$

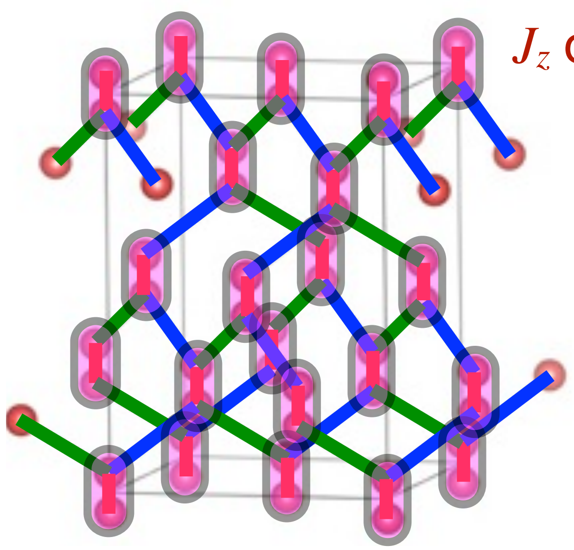
$$B_p = \tau_{p_t}^z \tau_{p_b}^z \tau_{p_l}^y \tau_{p_r}^y$$

$$[\mathcal{H}_{\text{eff}}, B_p] = [B_p, B_{p'}] = 0 \quad B_p^2 = 1$$

A. Kitaev, 2006

Anisotropic limit in 3D

hyperhoneycomb lattice

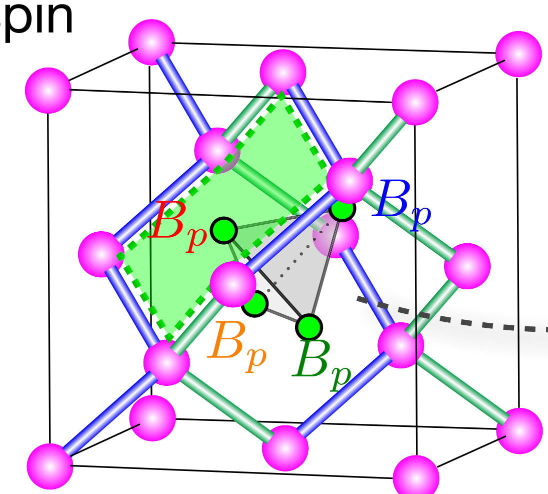


J_z dimer → pseudo-spin

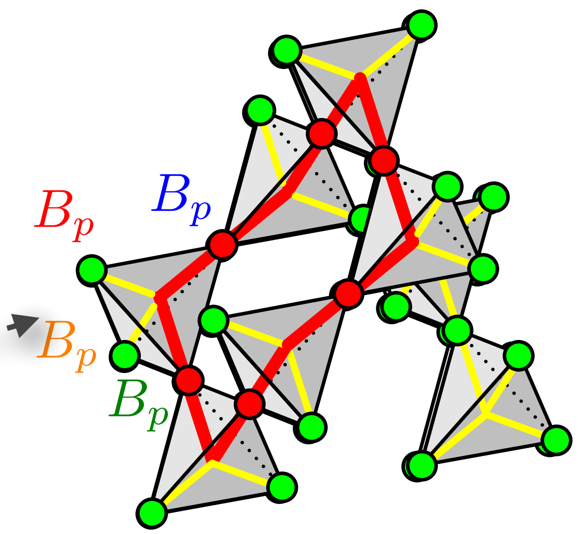
perturbation
in terms of J_x, J_y

ring exchange
-type interaction

tetrahedron of B_p → pyrochlore lattice of B_p



$B_p = \tau_1^z \tau_2^y \tau_3^x \tau_4^z \tau_5^y \tau_6^x$



- effective Ising-type model on a pyrochlore lattice with constraints

S. Mandal and N. Surendran, 2009; arXiv:1101.3718

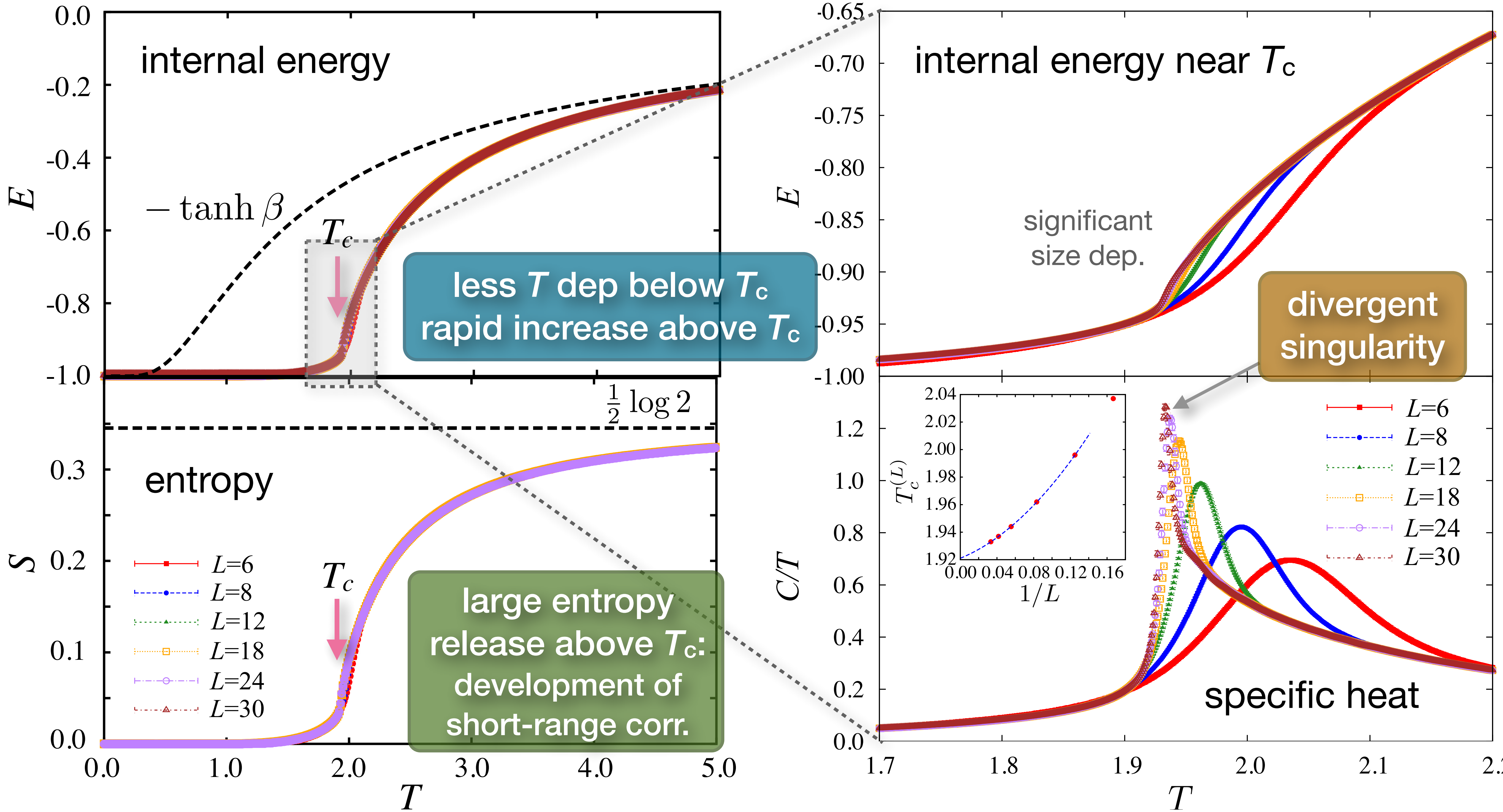
$$\mathcal{H}_{\rm eff} = -J_{\rm eff} \sum_p B_p \quad ; \; B_p = \pm 1, \; J_{\rm eff} = \frac{7}{256} \frac{J^6}{J_z^5} \; (J = J_x = J_y)$$

local constraints: $B_p B_p B_p B_p = 1$ for all tetrahedra + global constraints

(equivalent to 8-vertex model)

ground state: all B_p=+1
excited states: flipped B_p=-1 form closed loops to satisfy the local constraints

Results: Finite-T phase transition

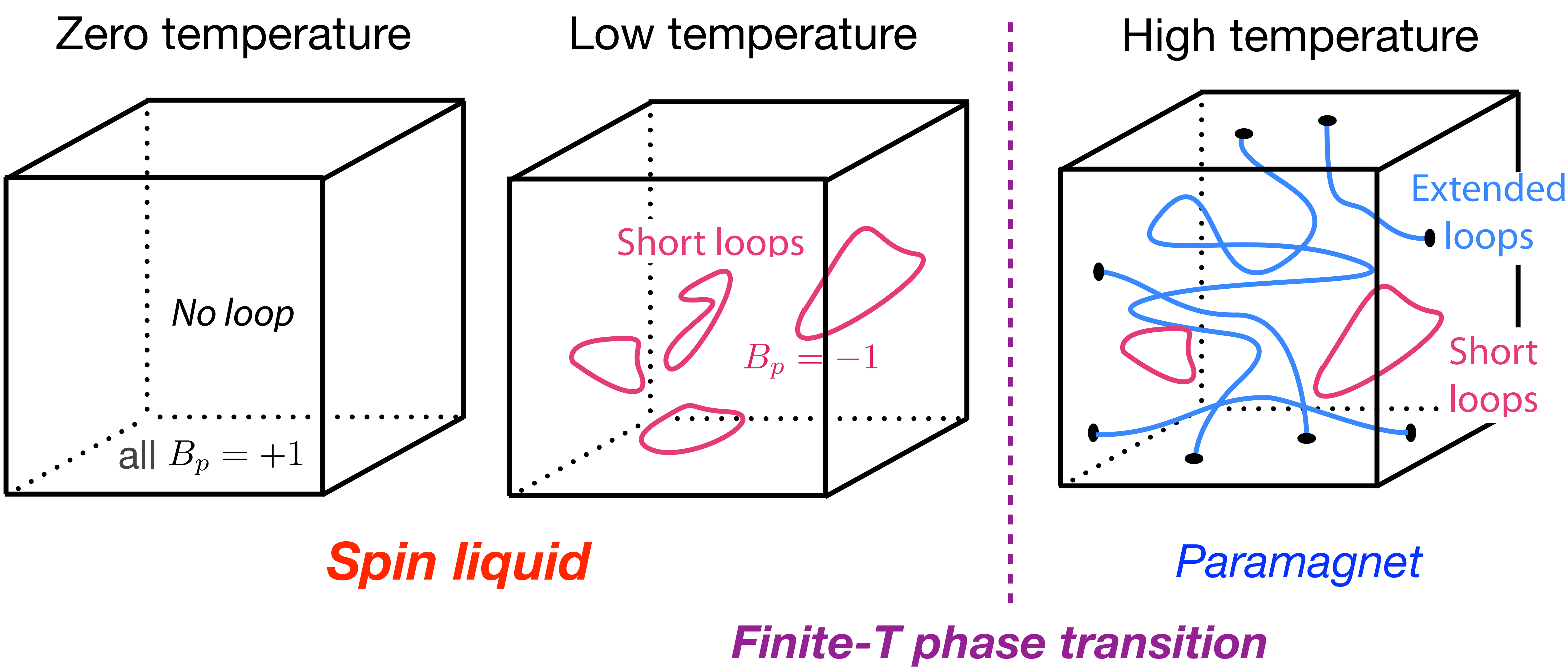


continuous phase transition at T_c=1.921(1) x J_{eff}

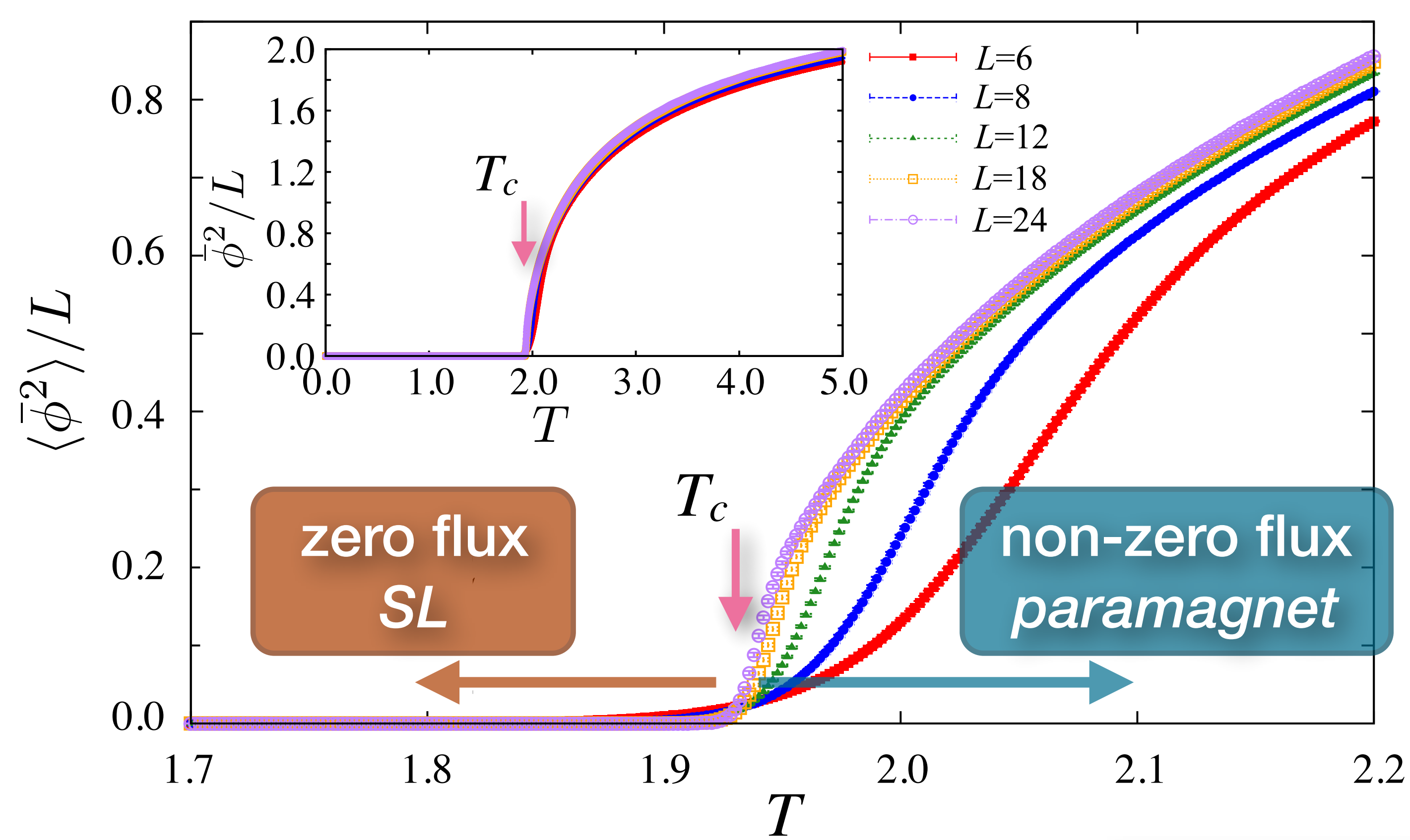
Topological point of view

- The transition is hard to characterize in terms local variables, as it apparently accompanies no symmetry breaking.

➡ characterization by loop degree of freedom (loops of flipped B_p)

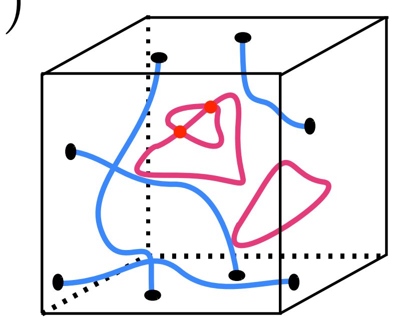


Results: characterization by flux density



$$\bar{\phi}^2/L = \frac{1}{L}\sum_i \sum_{\mu=1}^{3} (\phi_i^\mu)^2$$

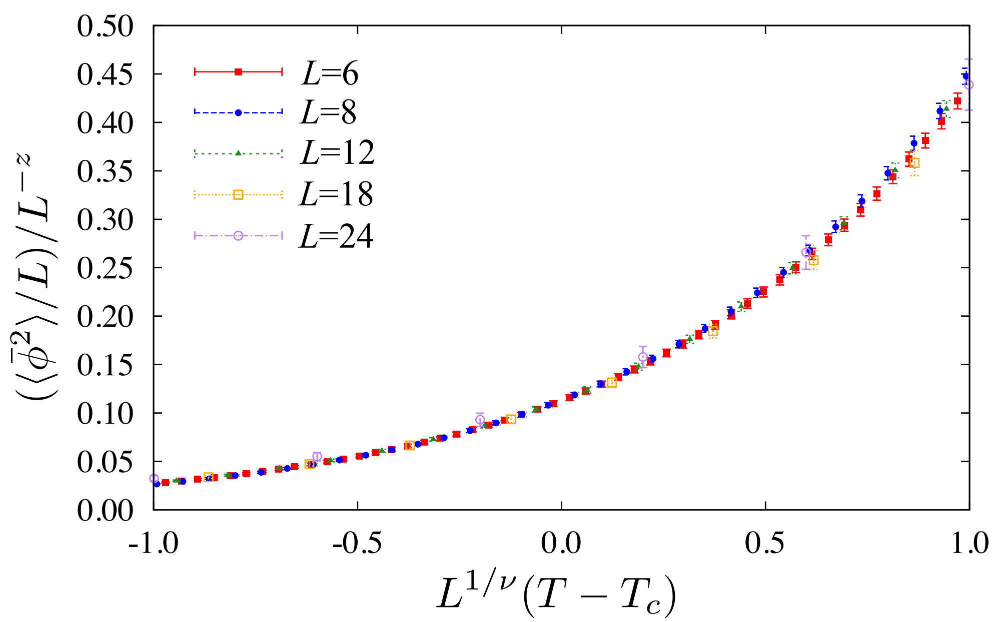
$$\phi_i^\mu = \frac{1}{L}\oint_{\mathcal{C}_i} \mathbf{a}_\mu \cdot d\mathbf{s}$$



path integral along a loop

primitive vector of pyrochlore

cf. flux density for close-packed classical dimer model (F. Alet *et al.*, 2006)



finite-size scaling: $\langle\bar{\phi}^2\rangle/L = L^{-z} f(L^{1/\nu}(T - T_c))$

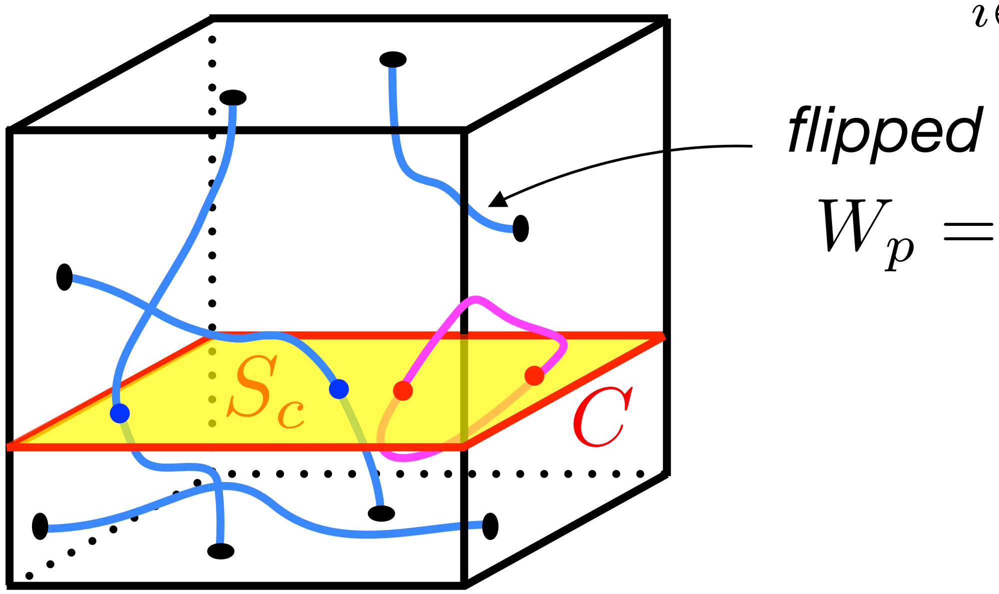
➡ $T_\mathrm{c} = 1.925(1),\ \nu = 0.60(5)\ [z = 1]$

consistent with the continuous transition at T_c
3D Ising universality class

NB. one-to-one correspondence to the 3D Ising model when omitting the global constraints

Topological viewpoint: back to the generic case

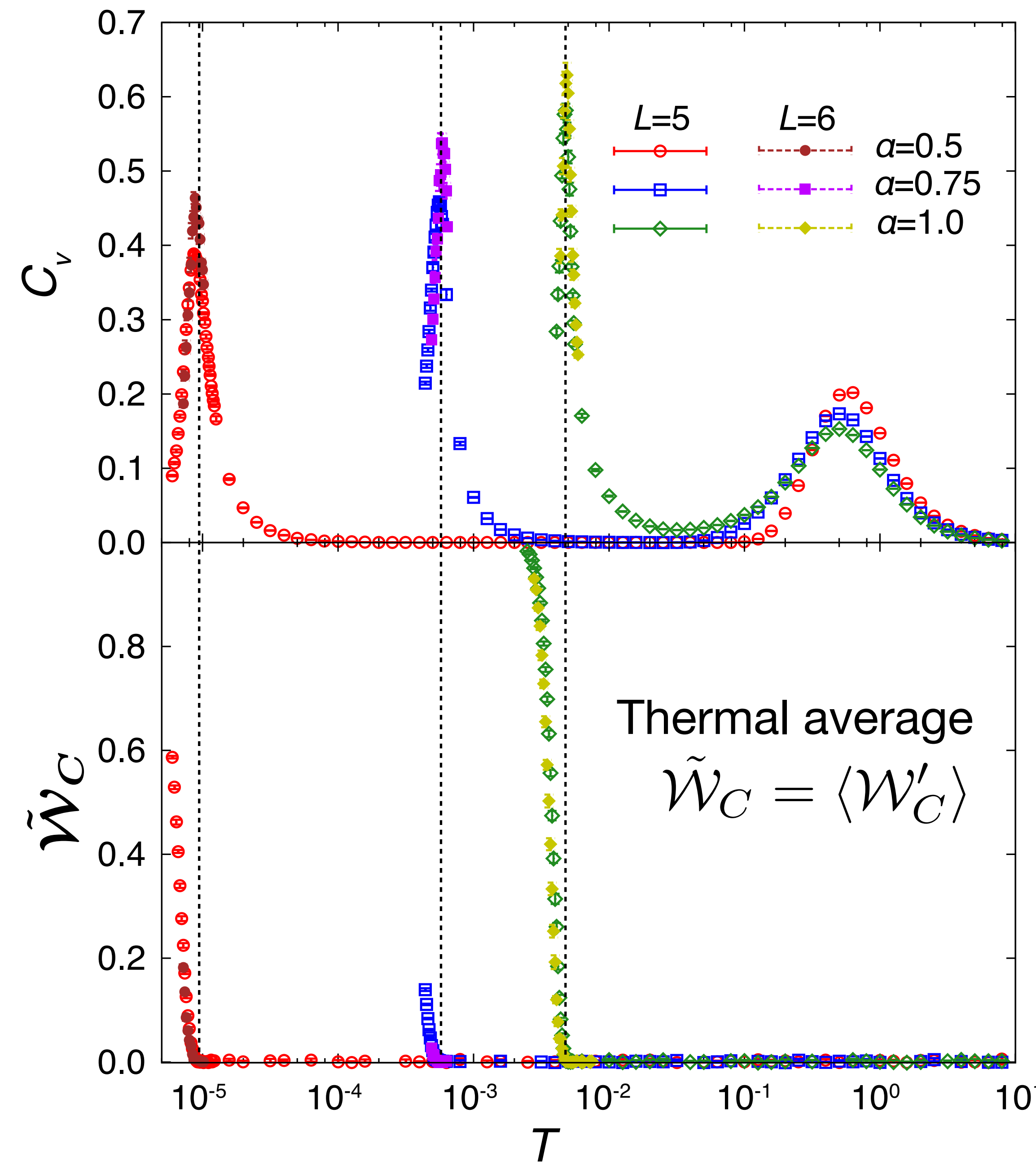
Loop operator (Wilson loop): $\mathcal{W}_C = \prod_{i \in C} \sigma_i^{l_i}$



$$\mathcal{W}_C = -\prod_{p \in S_C} W_p = -\prod_{r \in C} \eta_r \equiv -\mathcal{W}'_C$$

Extended loops : $\mathcal{W}_C = +1$ or -1 ➡ $\tilde{\mathcal{W}}_C = \langle \mathcal{W}'_C \rangle = 0$

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



The loop operator behaves like an order parameter.

Discussion

- spin ice (frustrated classical-spin antiferromagnets): local constraint by competing interactions
  - exact local constraint only at T=0
  - smearing out at finite T → no finite-T phase transition, just a crossover
- close-packed dimers: exact local constraint for all T (by hand)
  - unconventional phase transitions
    - zero flux state = columnar order of dimers
    - non-zero flux state = Coulomb phase
- 3D hyperhoneycomb Kitaev model: exact local constraint on local conserved quantities W_p for all T
  - phase transition between quantum spin liquid and paramagnet = proliferation of loops consisting of flipped conserved quantities W_p
  - The exact local constraint comes from S=1/2 algebra.

Prospects

- **further characterization of the phase transition in the quantum case**
  - hidden order? topological order? weak 1st order?
  - any difference between gapped and gapless regions?
  - how universal? specific to Kitaev models?

  cf. U(1) case (quantum spin ice): just a crossover? (Y. Kato and S. Onoda, preprint)

- **any classical correspondence?**
  - transition between Coulomb liquid and paramagnet
  - quantumness is necessary?

- **further interesting physics by transcription to mobile electrons**
  - quantum spin liquids + mobile electrons = ?