# **Frustrated Magnetism & Spin Liquids:**
## *Quantum Materials Summer School*

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# Three questions:

1. What *is* a spin liquid?

2. How to *stabilize* a spin liquid?

3. How to *detect* a spin liquid?

*What can go wrong?*

# What is a spin liquid?

- *Broad* sense:

[1]*Doesn't spontaneously break any symmetries*

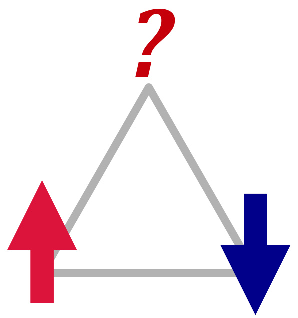
> Magnet that *doesn't order*[1] down to zero temperature **and** is *distinct*[2] *from a trivial paramagnet*[3]

[2]*Not "smoothly connected"*

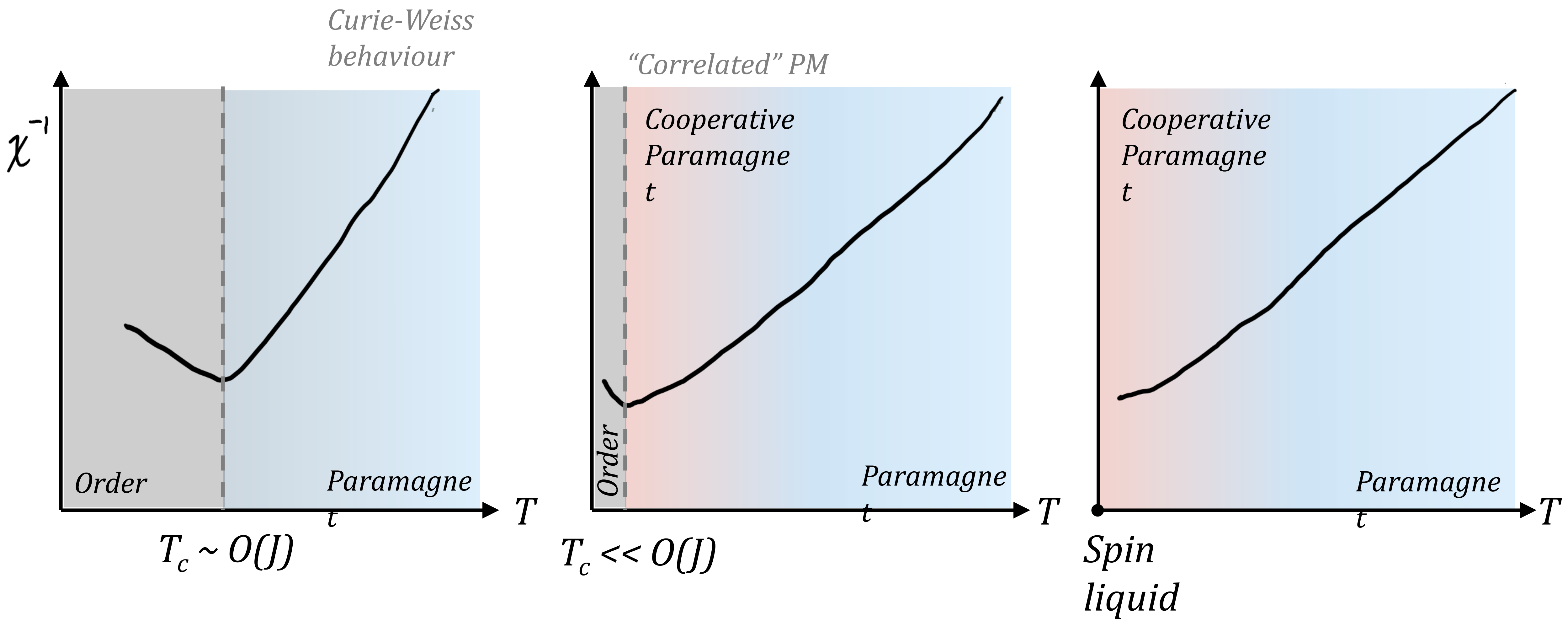
[3]*Has some kind of "topological order"*

Valence bond solid? ***No***
Frozen product state due to disorder? ***No***
One dimension? ***Complicated***

- Typically *highly frustrated*
- Broad *cooperative paramagnet* regime, *well below* characteristic scale




*No choice to satisfy* ***all*** *bonds*




Unfrustrated

Frustrated

*Highly* Frustrated?

# Why are they interesting?

## *Fractionalized* excitations

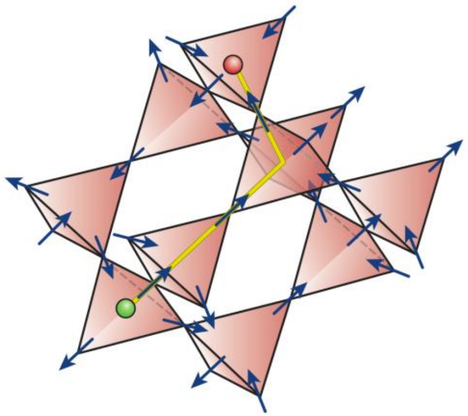
Excitations *split* into new independent parts

## Emergent *gauge* theories

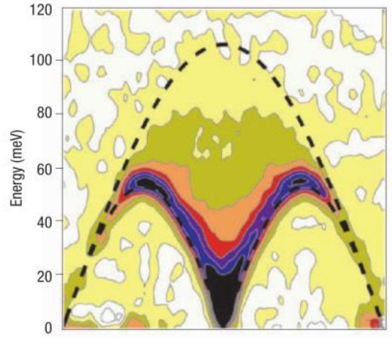
Realizations of electromagnetism, complete with *new* photon

## *Topological* order

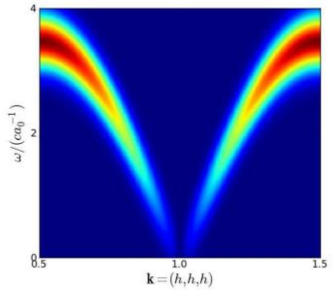
Long-range quantum entangled ground states



*Magnetic monopoles in spin ice*

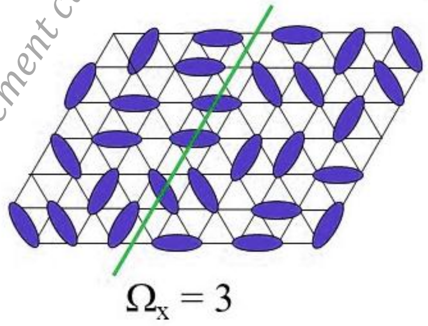


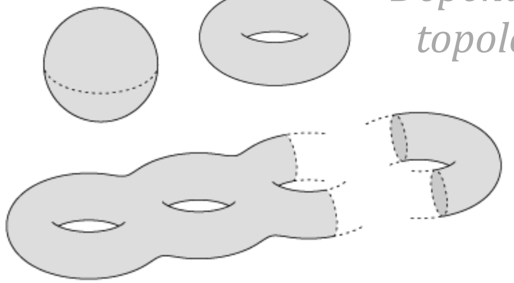

*Spinons in a spin chain*




*Prediction for emergent photon in quantum spin ice*

*Entanglement cut*

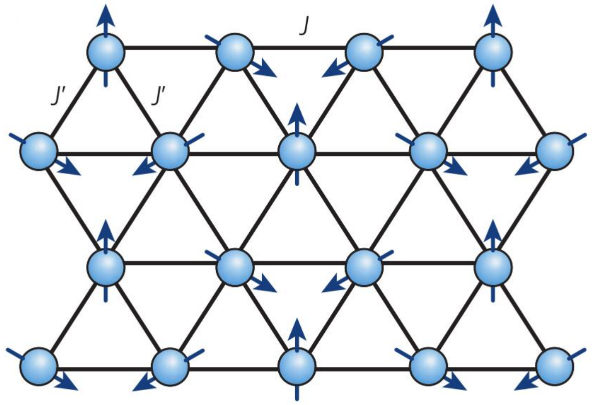


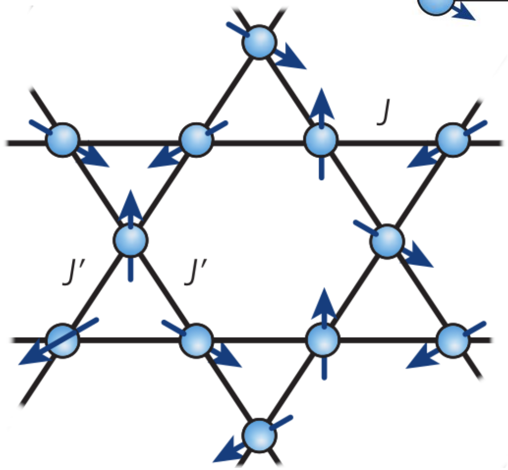



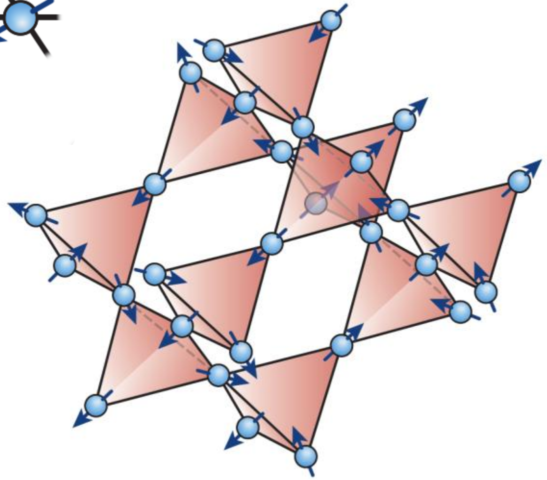
*Depends on topology*

# What kind of models are *known* to have spin liquid ground states?

*Lots of triangles*



- **Classical models**
  - Triangular Ising AFM
  - Pyrochlore Heisenberg AFM
  - Spin ice, …

*Extensive ground state manifolds*

*Quantum*

- **Exactly solvable models**
  - Toric code,
  - Kitaev's honeycomb model
  - String-net models, …

*Hand-crafted interactions*

- **Non-solvable models**
  - Kagome anti-ferromagnet
  - Quantum spin ice
  - $J_1$-$J_2$ models, …

*Numerical (mostly)*

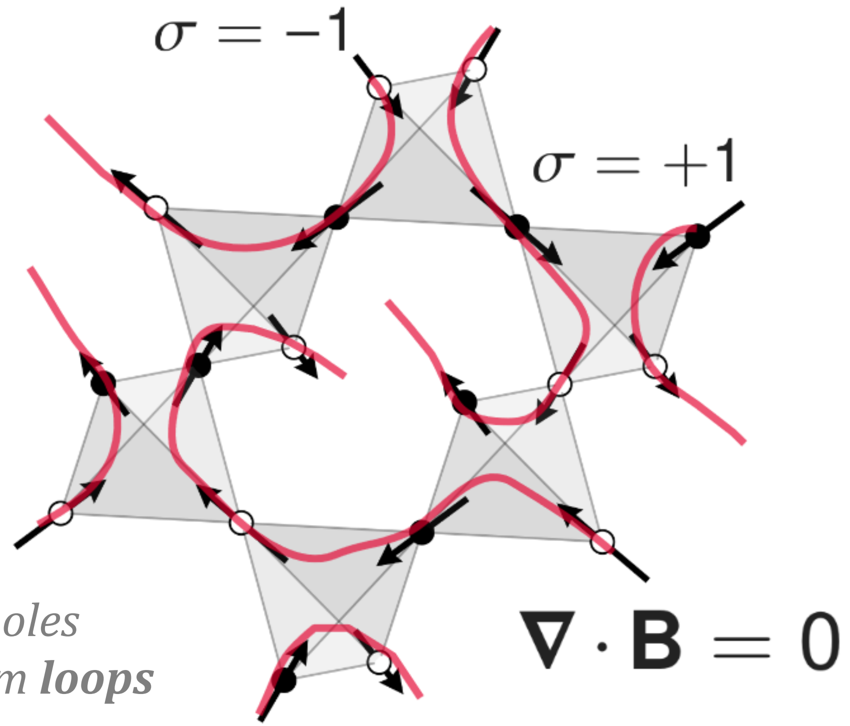
# Example: *Classical* Spin Ice

- Simplest realization:

*Ising model on pyrochlore lattice*

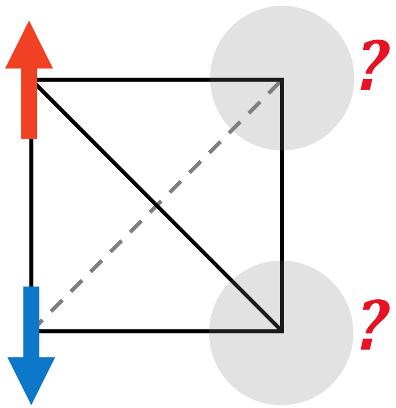
*Anti-ferromagnetic exchange*

$$E = J \sum_{\langle ij \rangle} \sigma_i \sigma_j$$



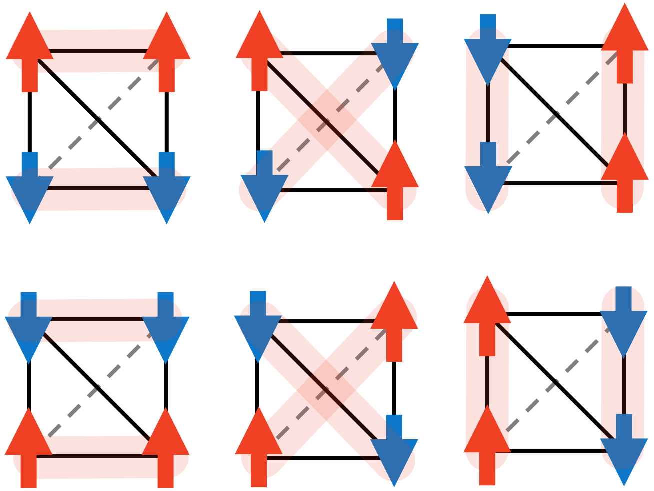

*Dipoles form* ***loops***

*Corner-sharing tetrahedra*




*Tetrahedron*

*"Two in / two out" rule*



*Six ground states per tetrahedron*

- Extensive ground state degeneracy
- **Classical spin liquid**

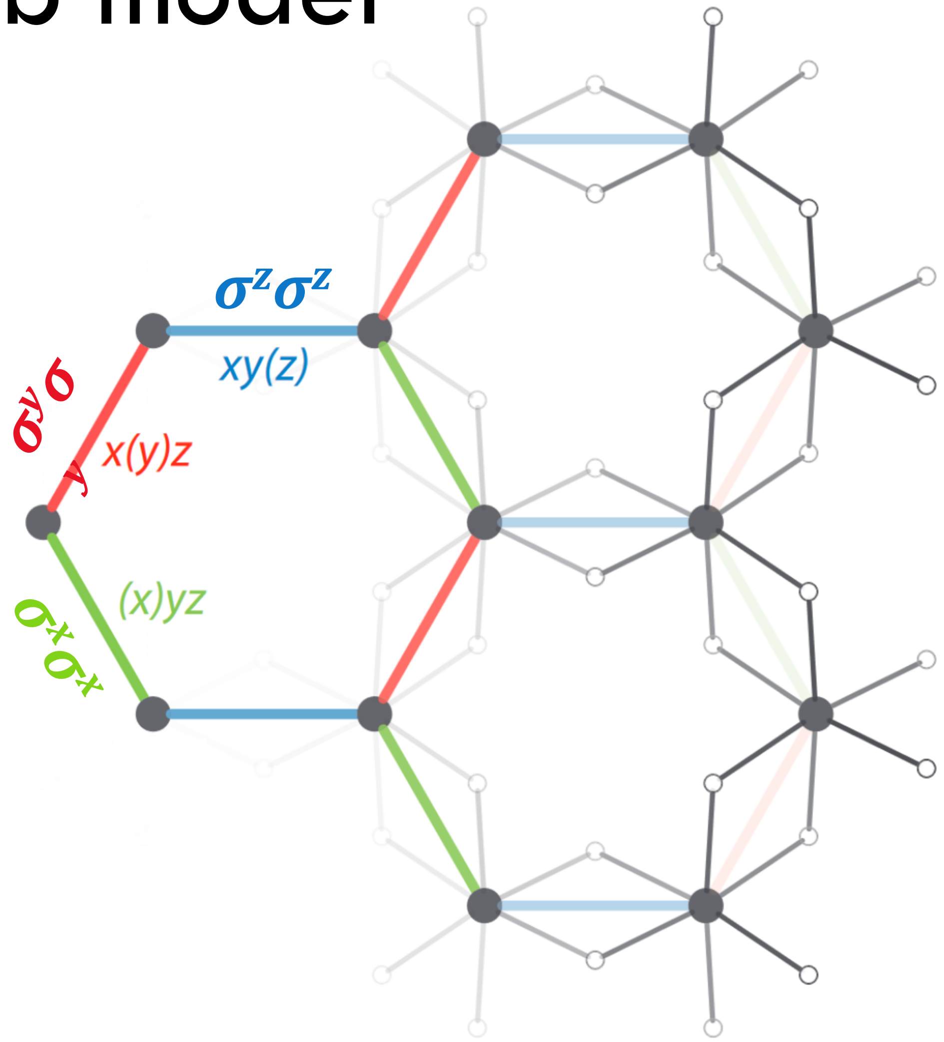
# **Example:** Kitaev's Honeycomb model

- Frustrated spin-1/2 model on honeycomb lattice

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

*Two-spin interaction s only*

- Frustration by *interactions* not geometry




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

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

# **Example:** *Quantum* spin ice

- Simplest realization:

$$H = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z - J_{\pm} \sum_{\langle ij \rangle} \left( S_i^+ S_j^- + \text{h.c.} \right)$$

*Classical spin ice model*

*Term that induces quantum fluctuations*

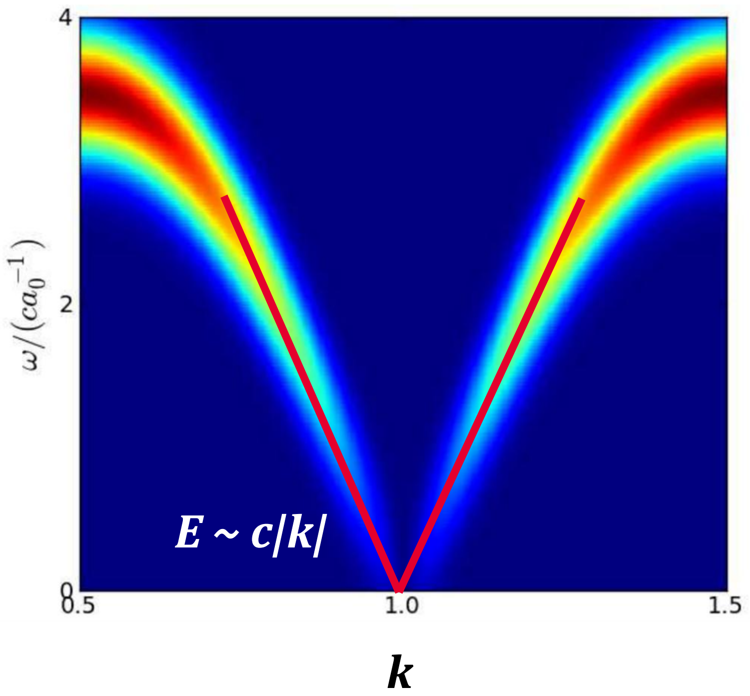
- Effective model:

*Perturbative in quantum part*

$$-\frac{12 J_{\pm}^3}{J_{zz}^2} \sum_{\text{hexagons}} P_{\text{ice}} \left( S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + \text{h.c.} \right) P_{\text{ice}}$$

*Can map to U(1) lattice gauge theory;* ***solve numerically***

*Emergent photon excitation*




***Classical SI:*** **emergent magnetostatics**

***Quantum SI:*** **emergent electrodynamics**

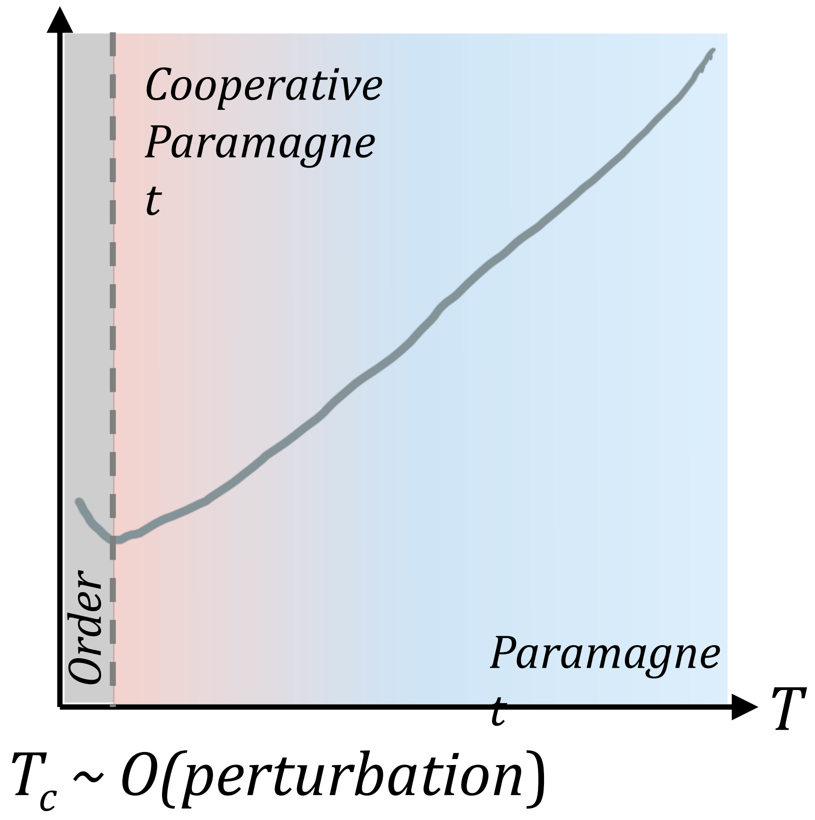
*Hermele et al., Phys. Rev. B* ***69*** *064404 (2004), Benton et al, Phys. Rev. B* ***86*** *075154 (2012)*

# Stability?

*Classical* spin liquids are **unstable to small perturbations,** always *"fine-tuned"*

- "Third-law": Can't have finite entropy density *generically*
- Perturbations that lift degeneracy set ordering scale

*Typical scenario*




**Instability *can* be toward *quantum* spin liquid**

# Stability?

**Stability is possible!**

- Kitaev? *Time-reversal symmetry*
- Quantum spin ice? ***Any*** *perturbation*

- Still need to worry about **energy scales**

*Effective model of QSI*

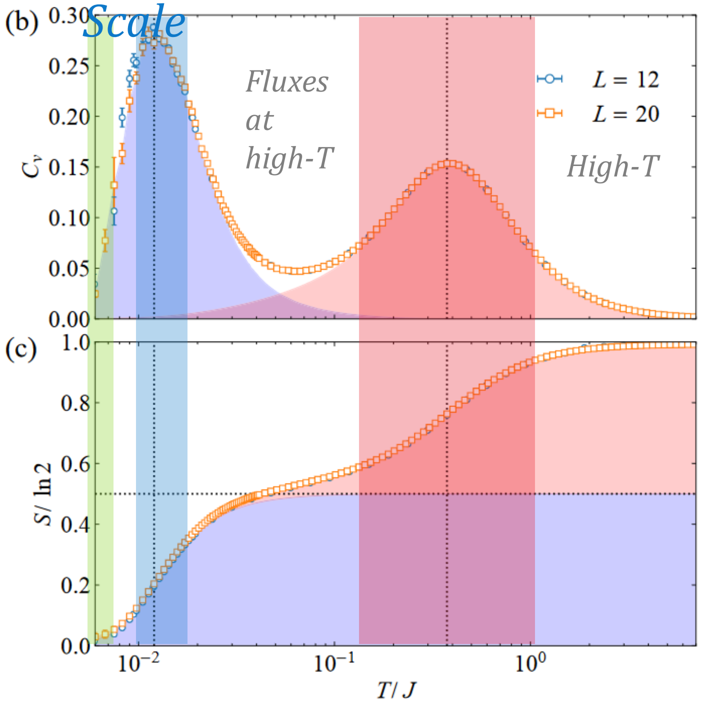
$$-\frac{12J_{\pm}^3}{J_{zz}^2}\sum_{\text{hexagons}} P_{\text{ice}}\left(S_1^+S_2^-S_3^+S_4^-S_5^+S_6^- + \text{h.c.}\right)P_{\text{ice}}$$

*Temperature/perturbations must be compared to* ***this***

*Kato & Onoda, Phys. Rev. Lett.* ***115*** *077202 (2015);*
*Motome & Nasu, JPSJ* ***89*** *012002 (2020)*

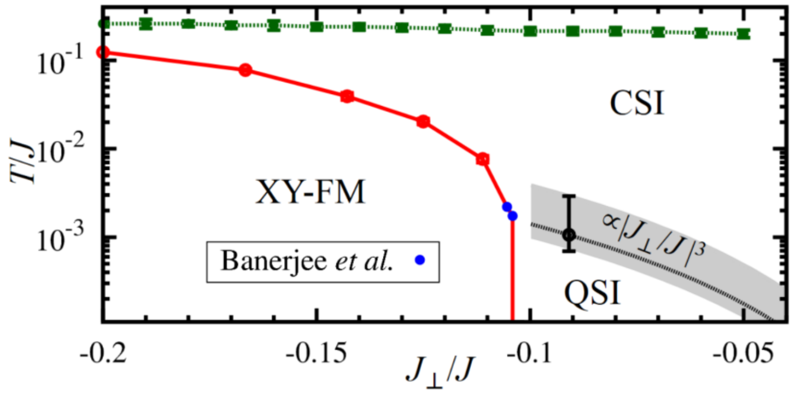
*Flux Scale* *Fermionic Scale*

*Ground state physics*




*Thermodynamics of Kitaev Model*

*Phase diagram of QSI*




… temperatures *order of magnitude* or two **smaller than**

# Signatures of spin liquids

- Lack of magnetic order
- Shows broad excitation spectrum

*Fractionalization ?*

- Still *dynamic* at very low temperature
- Topological response

*e.g. Emergent photon, quantized gravitational response, …*

*Is disorder playing a role?*

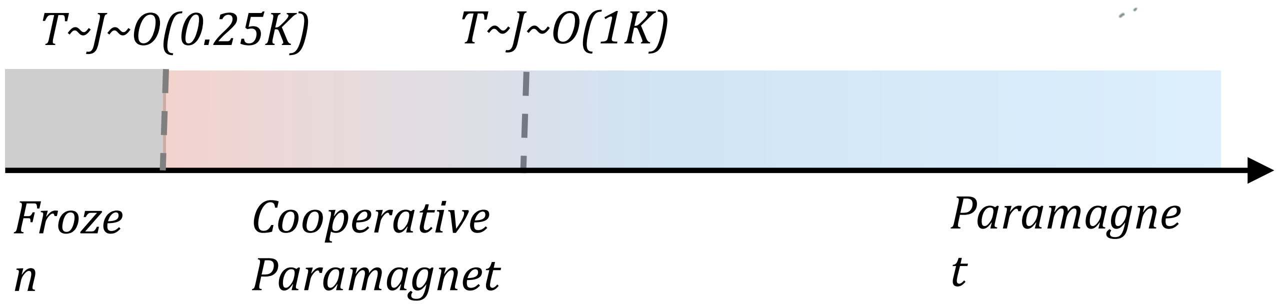
*Is temperature/energy low enough?*

*Conventional route?*

# Example: $(Dy,Ho)_2Ti_2O_7$

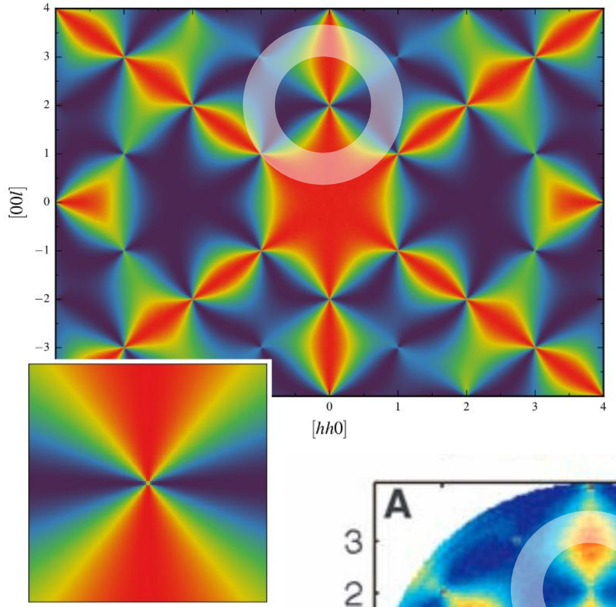
*Additional terms*

$$J_{\rm nn}\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z + \frac{3D_{\rm nn}r_{\rm nn}^3}{5}\sum_{i<j}\left[\frac{\hat{\mathbf{z}}_i\cdot\hat{\mathbf{z}}_j}{|\mathbf{r}_{ij}|^3} - \frac{3\left(\hat{\mathbf{z}}_i\cdot\hat{\mathbf{r}}_{ij}\right)\left(\hat{\mathbf{z}}_j\cdot\hat{\mathbf{r}}_{ij}\right)}{|\mathbf{r}_{ij}|^3}\right]\sigma_i^z\sigma_j^z$$

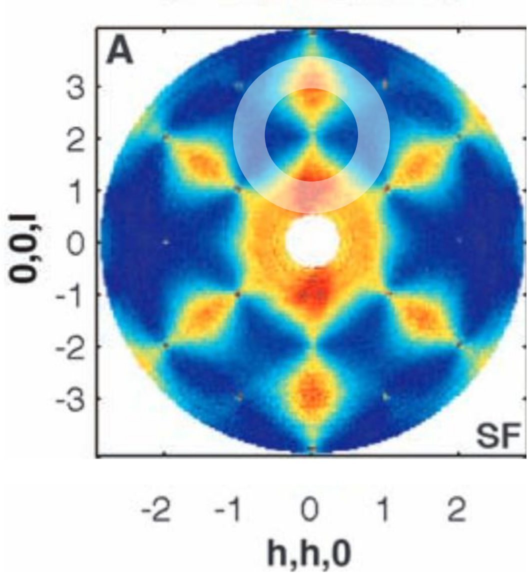



- *Fine-tuned*; further neighbour/dipole interactions **lift degeneracy**
- *Freezes* before order and/or quantum effects

*Result from simplest version of th...*




**"Pinch-point"**

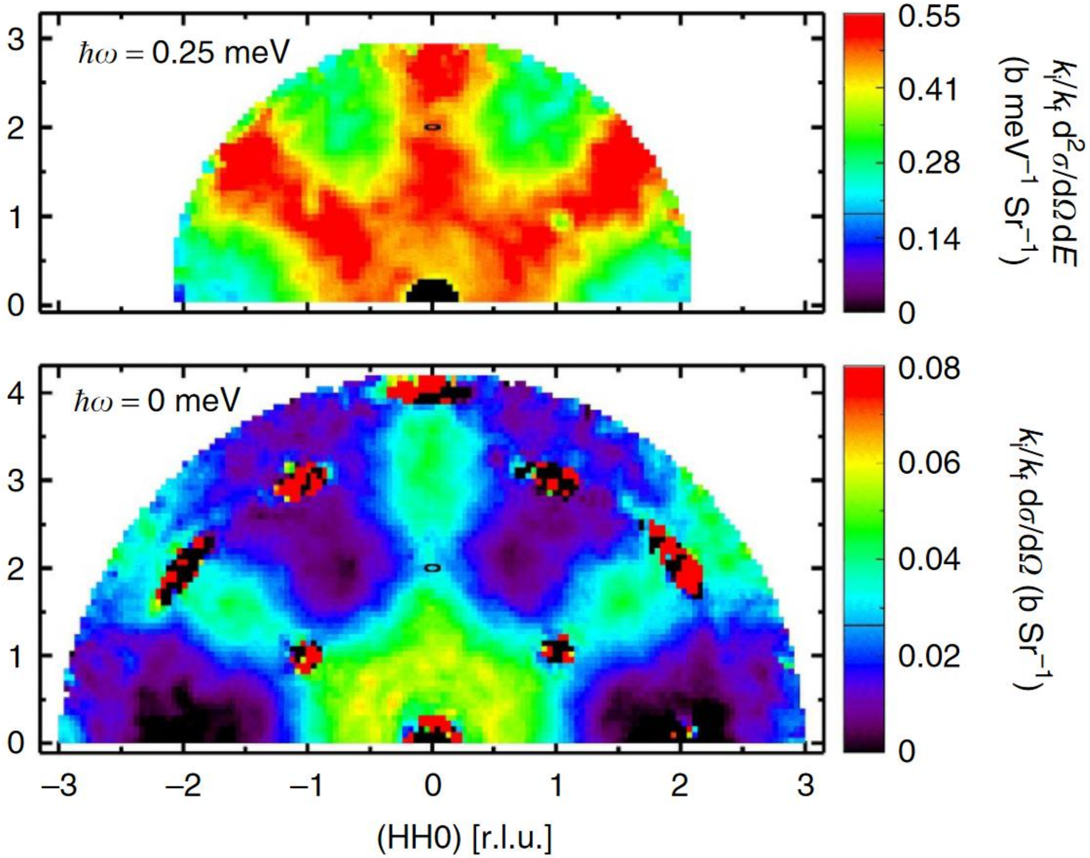



*Diffuse neutron scattering on $Ho_2Ti_2O_7$*

*Fennell et al., Science **326**, 415-417*

# **Example:** $Pr_2Zr_2O_7$

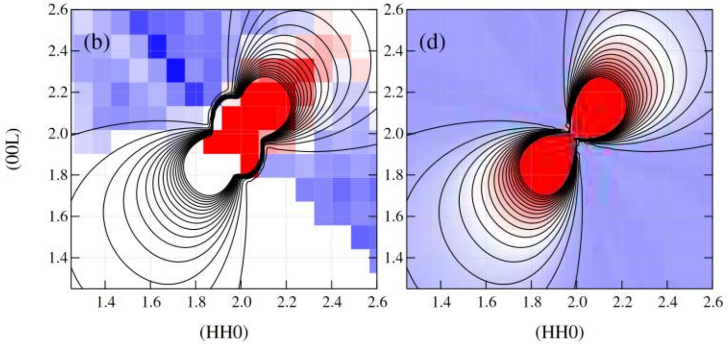
- Example of *quantum spin ice?*

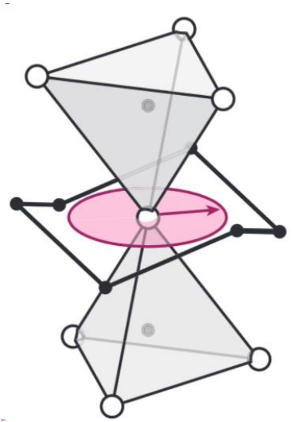



*Spatial correlations (still) need detail explanation*

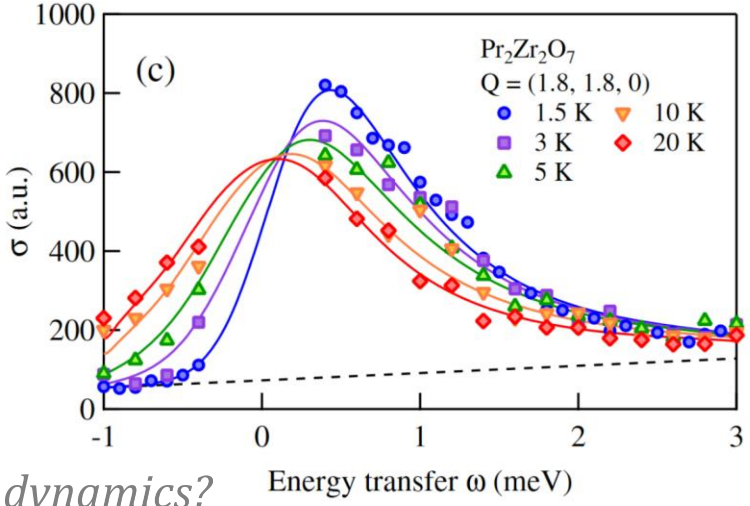
- Evidence for significant *structural disorder*

*Diffuse (structural) neutron scattering*



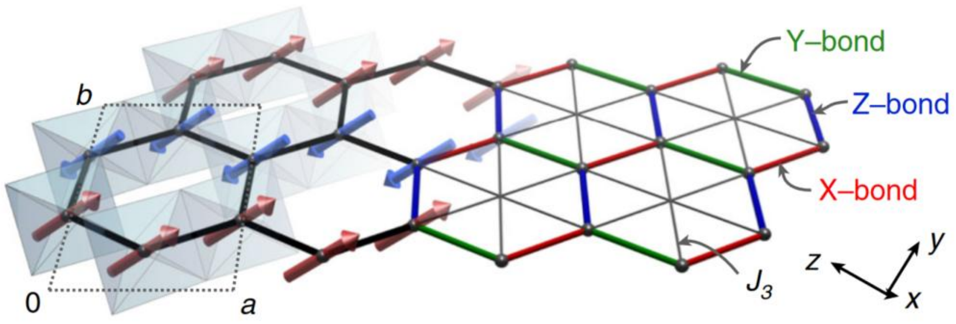
*"Off-centering" of Pr ion*



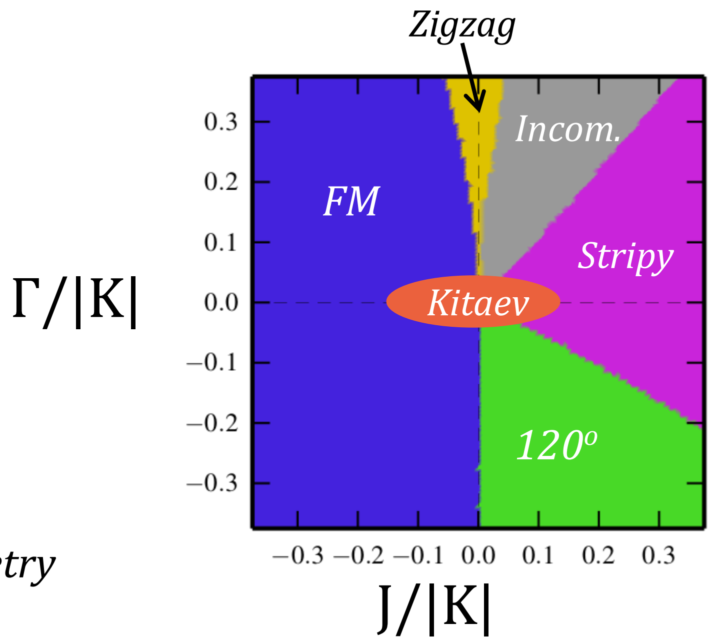

*Explains dynamics?*

*Kimura et al, Nat. Comm.* ***4*** *1934 (2013), Martin et al, Phys. Rev. X* ***7*** *041028 (2017)*

# **Example:** $RuCl_3$




- Kitaev spin liquid is *stable,* **but …**
- … sub-dominant perturbations large enough to **destroy the spin liquid**



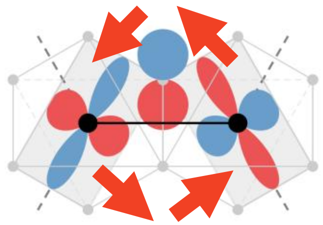

$$\sum_{\langle ij\rangle\in\alpha\beta(\gamma)} \left[ J\mathbf{S}_i\cdot\mathbf{S}_j + K S_i^{\gamma} S_j^{\gamma} + \Gamma\left(S_i^{\alpha} S_j^{\beta} + S_i^{\beta} S_j^{\alpha}\right)\right]$$

*Jackeli/Khaliullin*

*From direct d-d overlap*

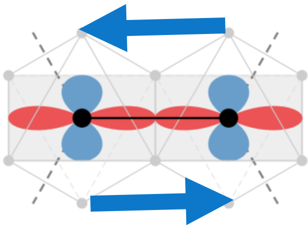
*Cross-term*

***Generic*** *symmetry allowed model*



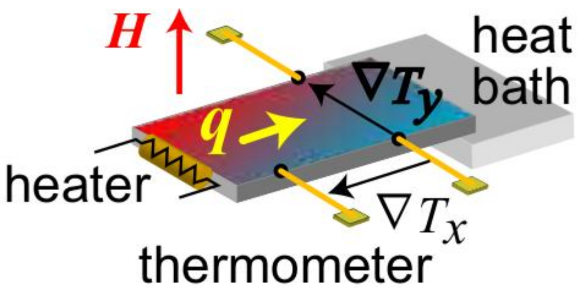
*Ligand mediated*

*Direct overlap*



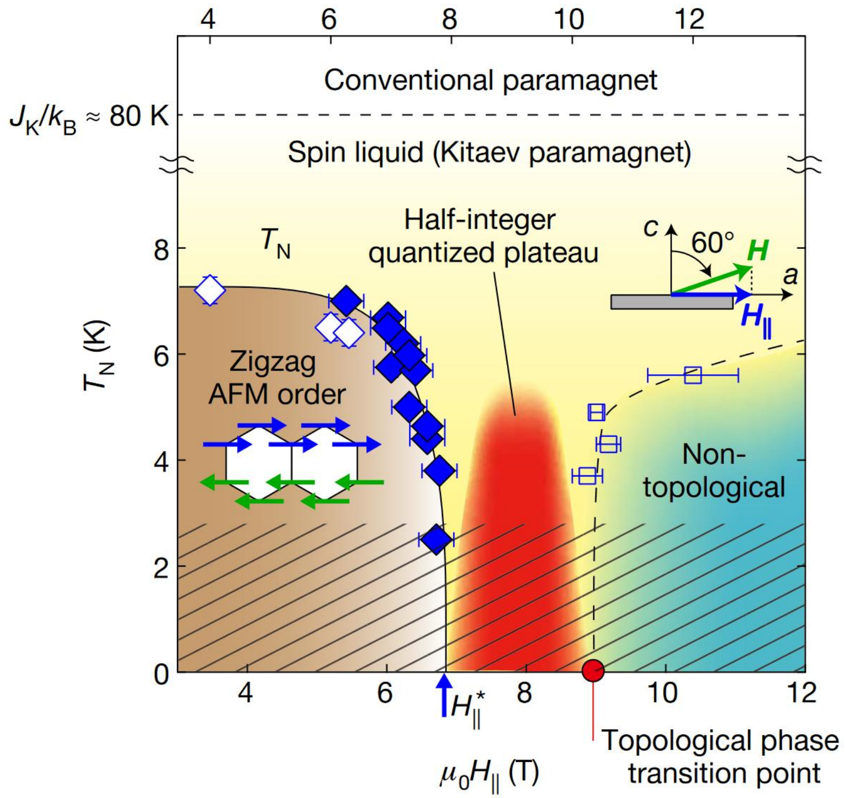
*Katakuri et al., New. J. Phys.* ***16,*** *013056 (2014)*
*Rau, Lee & Kee, Phys. Rev. Lett.* ***112,*** *077204 (2014)*

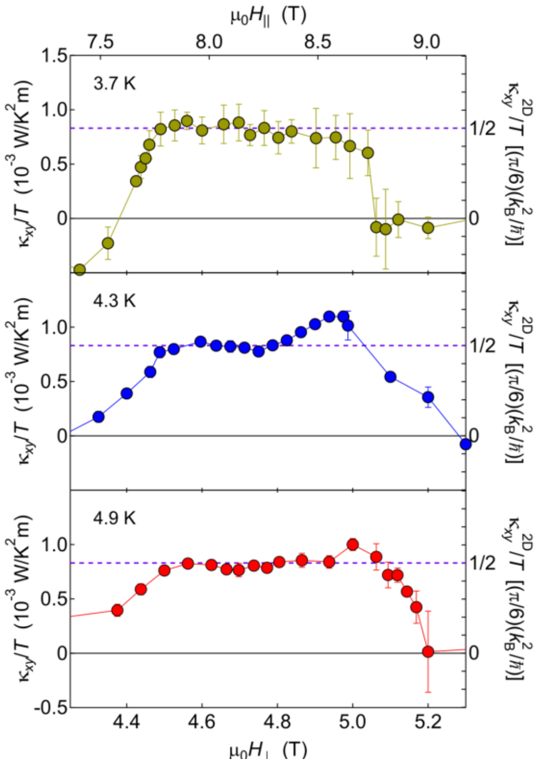
# **Example:** $RuCl_3$ in (tilted) magnetic field




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

*(Chiral) central charge of edge mod*




- Conventional explanation? ***Hard***, since *half-quantized*
- This experiment has *not yet been (independently) reproduced*

*Kasahara et al, Nature (2018)*

# Three ~~questions~~ "answers"

1. What *is* a spin liquid?

   → Magnet that *doesn't order* down to zero temperature **and** is *distinct from a trivial paramagnet*

2. How to *stabilize* a spin liquid?

   → Look for highly frustrated models (e.g. extensive degeneracy), minimize any perturbations

3. How to *detect* a spin liquid?

   → Go to low enough energy, be *mindful* of disorder, look for fractionalized excitations and/or topological responses