A Field Guide to Magnetic Frustration

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*Coming Soon

Outline

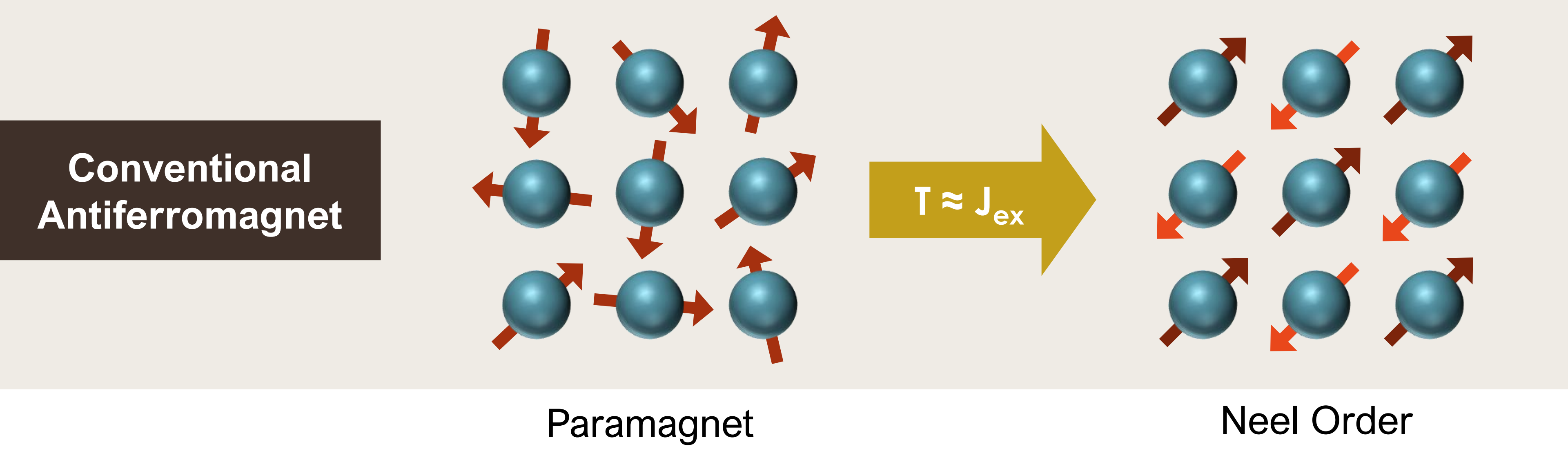
1. Introduction to magnetic frustration and its …
    - Origin
    - Effect
    - Detection

2. Frustration in the classical limit (dipolar spin ice)

3. Frustration in the quantum limit (quantum spin liquids)

4. Case Studies:
    - Proximate Kitaev spin liquid in α-$RuCl_3$
    - Phase competition in quantum XY pyrochlores

Magnetic Frustration: the inability of a magnet to satisfy all of its pairwise interactions simultaneously, resulting in the suppression of conventional magnetic order.



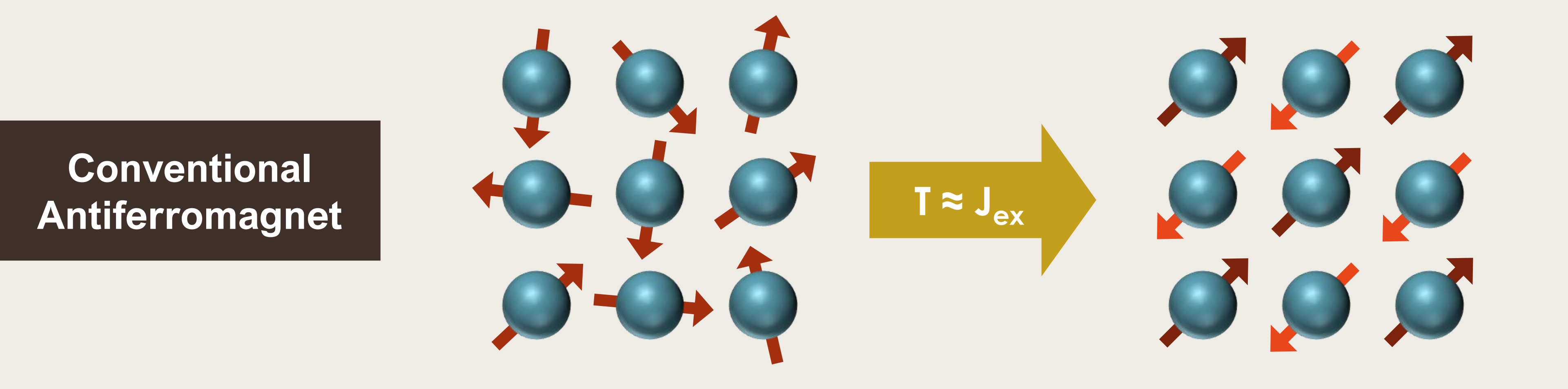
Conventional Antiferromagnet

$T \approx J_{ex}$

Paramagnet

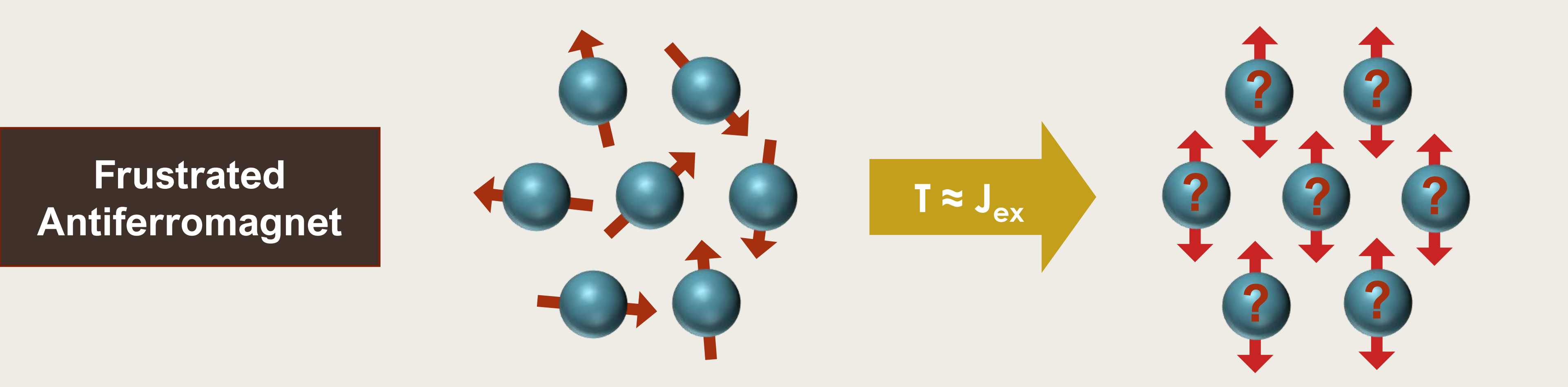
Neel Order

Magnetic Frustration: the inability of a magnet to satisfy all of its pairwise interactions simultaneously, resulting in the suppression of conventional magnetic order.



Conventional Antiferromagnet

$T \approx J_{ex}$

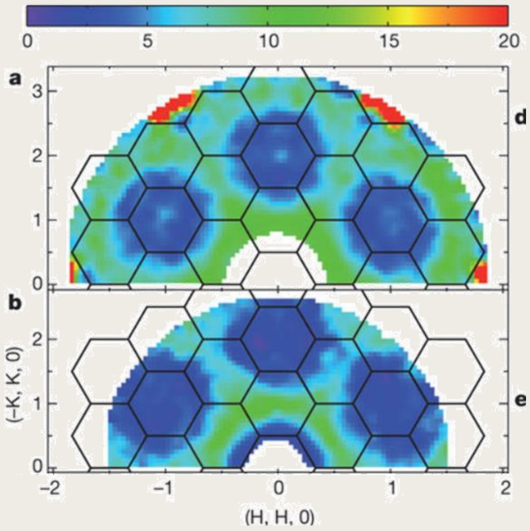


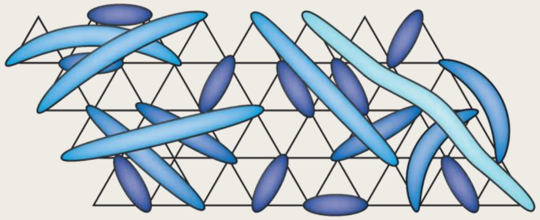
Frustrated Antiferromagnet

$T \approx J_{ex}$

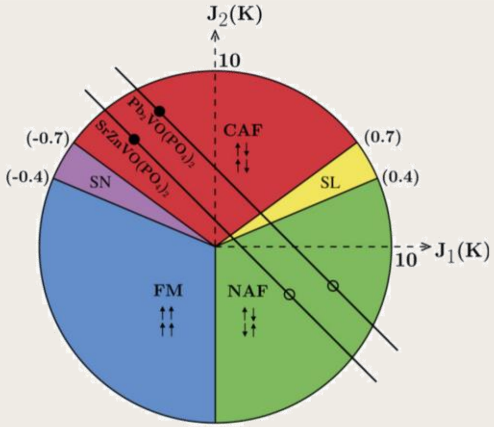
Big Picture: Why do we care about magnetic frustration?

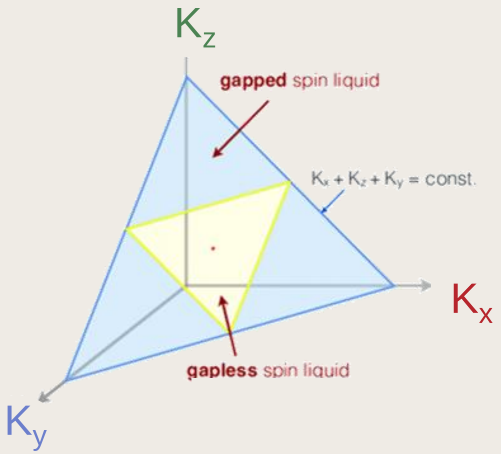
Exotic magnetic ground states



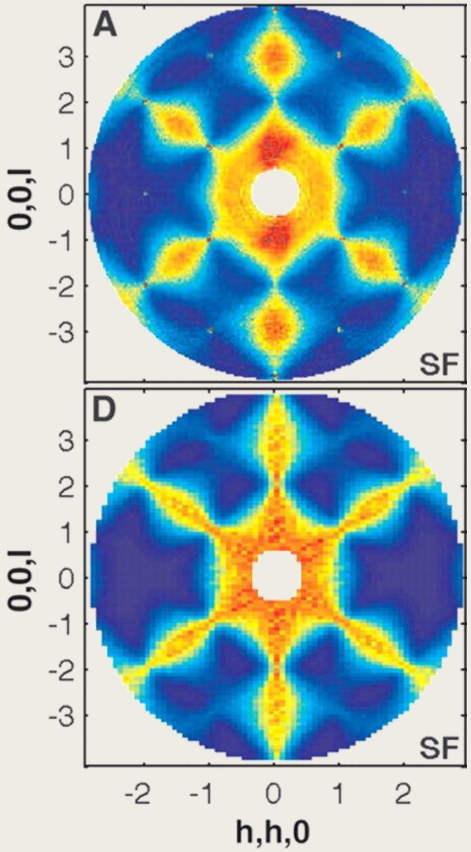


Tractable theoretical models





Connection between theory and experiment

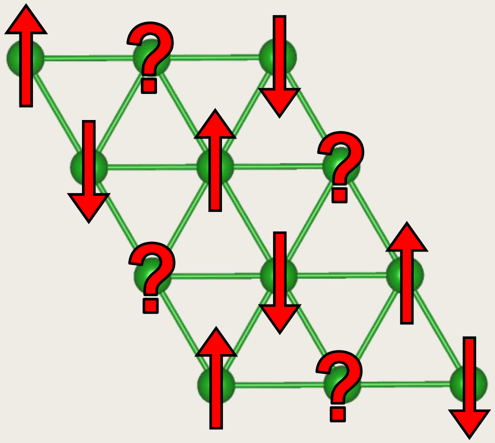


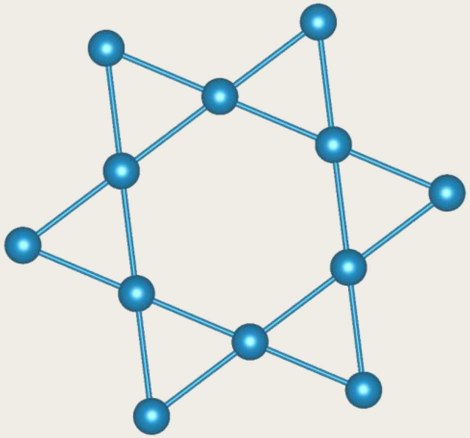
There are two main mechanisms of frustration:

1. **Lattice geometry**
2. Competing interactions

In 2D:

Triangular

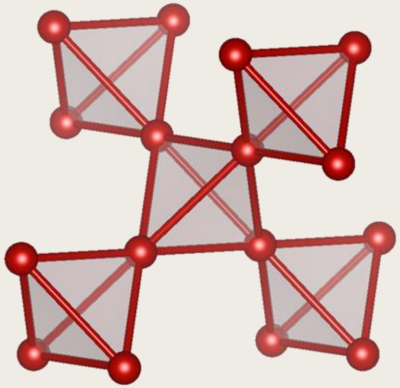


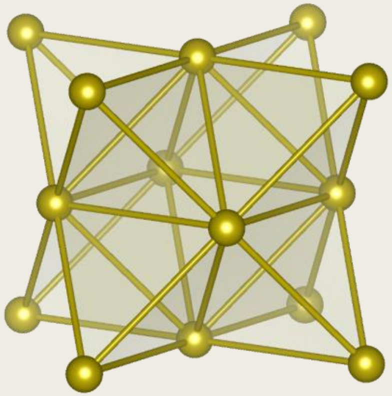


Kagome

In 3D:

Pyrochlore



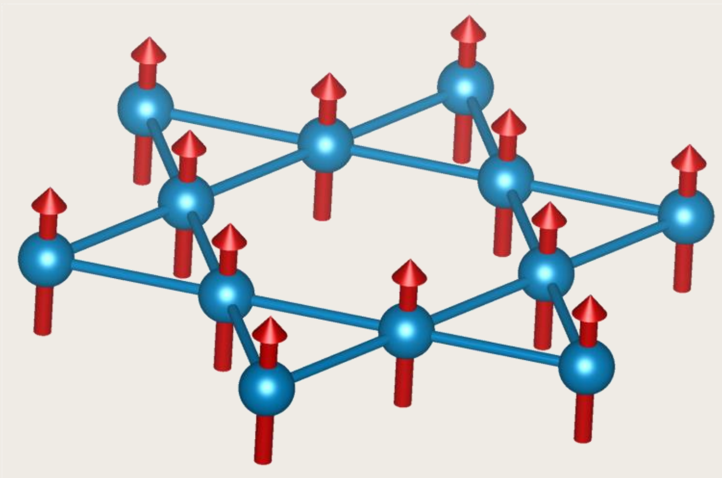


FCC

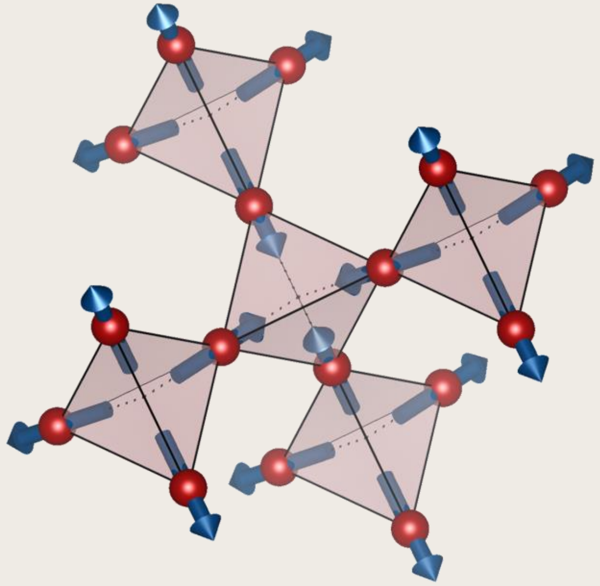
There are two main mechanisms of frustration:

1. **Lattice geometry**
2. Competing interactions

Important caveat: A frustration-prone lattice does not guarantee frustration in a real material. Depends on both the spin anisotropy and the exchange interactions (AFM or FM).



e.g. ferromagnetically coupled Ising spins on the Kagome lattice

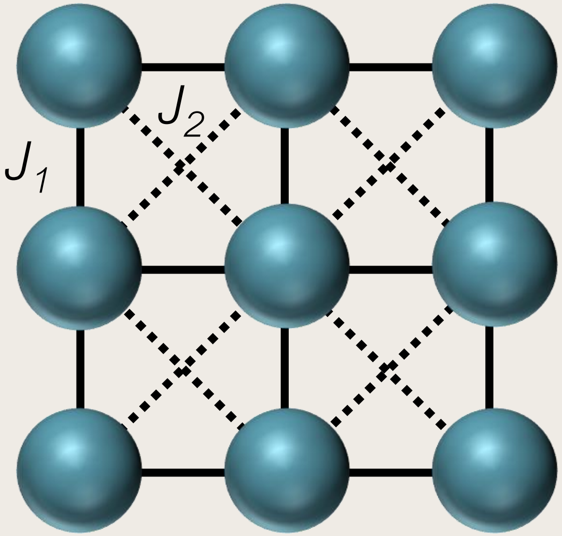


e.g. antiferromagnetically coupled Ising spins on the pyrochlore lattice

There are two main mechanisms of frustration:

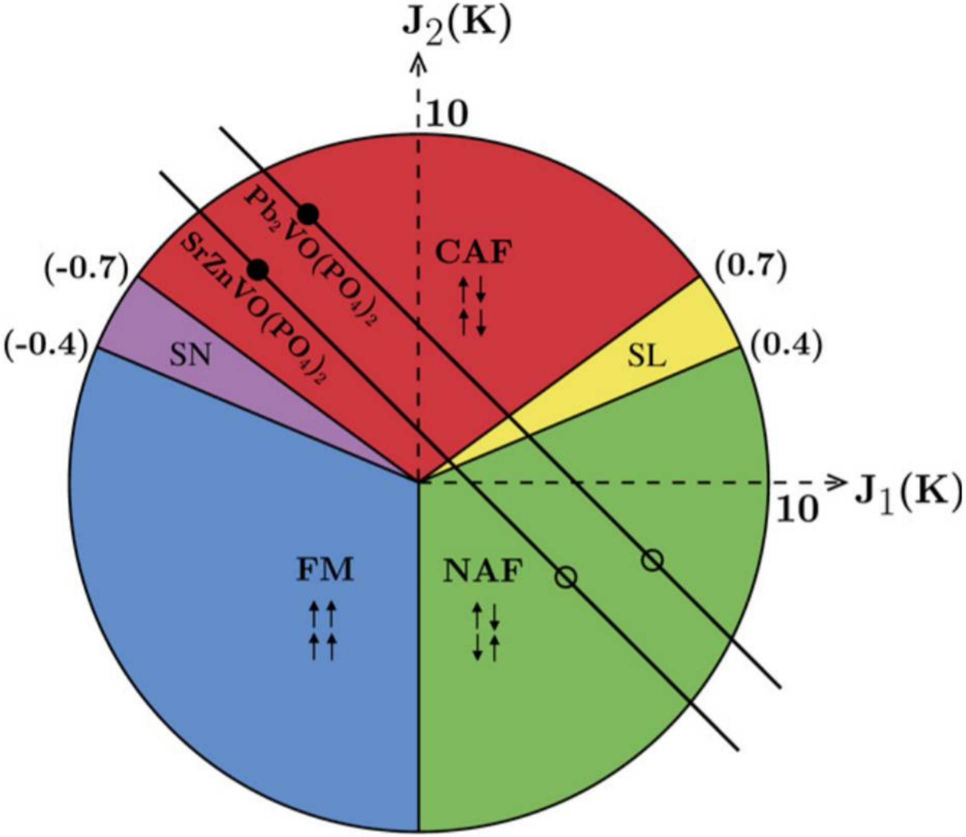
1. Lattice geometry
2. **Competing interactions**

J_1 - J_2 Frustrated Square Lattice



— J_1 n.n. exchange

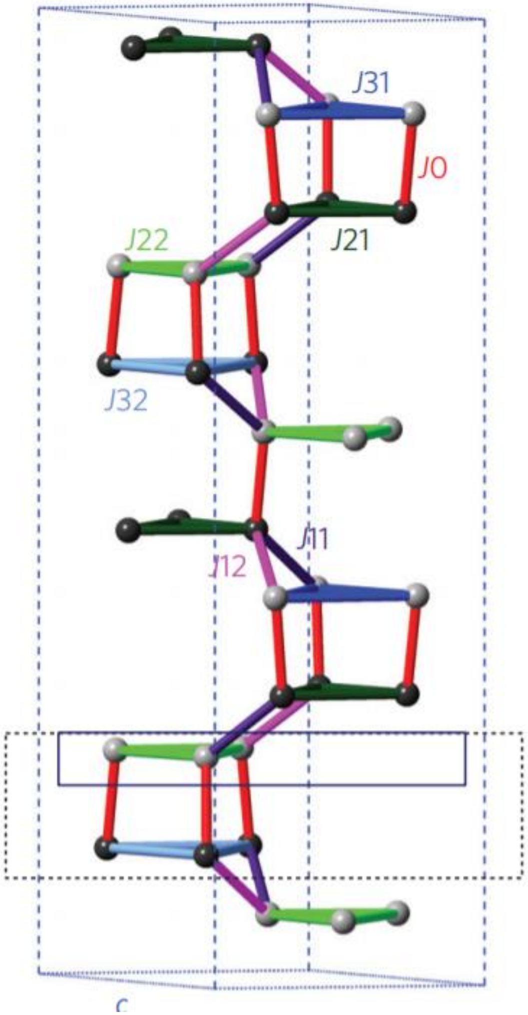
····· J_2 n.n.n. exchange



M. Skoulatos *et al.*, EPL 88 57005 (2009)

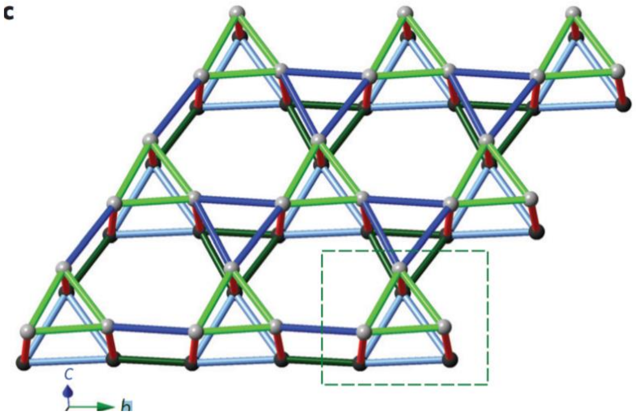
There are two main mechanisms of frustration:

1. Lattice geometry
2. **Competing interactions**



Things can get very messy.

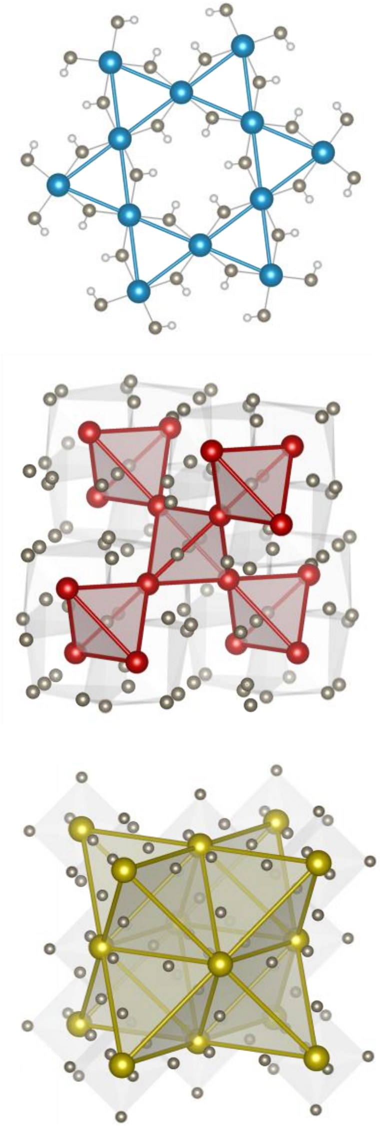
For example, in the spin liquid candidate $Ca_{10}Cr_7O_{28}$, which has a bilayer Kagome lattice, there are at least 5 relevant exchange couplings, some FM and some AFM.



Exchange	Coupling (meV)	Type
J0	−0.08(4)	FM
J11	0	
J12	0	
J21	−0.76(5)	FM
J22	−0.27(3)	FM
J31	0.09(2)	AFM
J32	0.11(3)	AFM
ΣJ	−0.91(17)	

Balz *et al.*, Nature Physics **12**, 942–949 (2016)

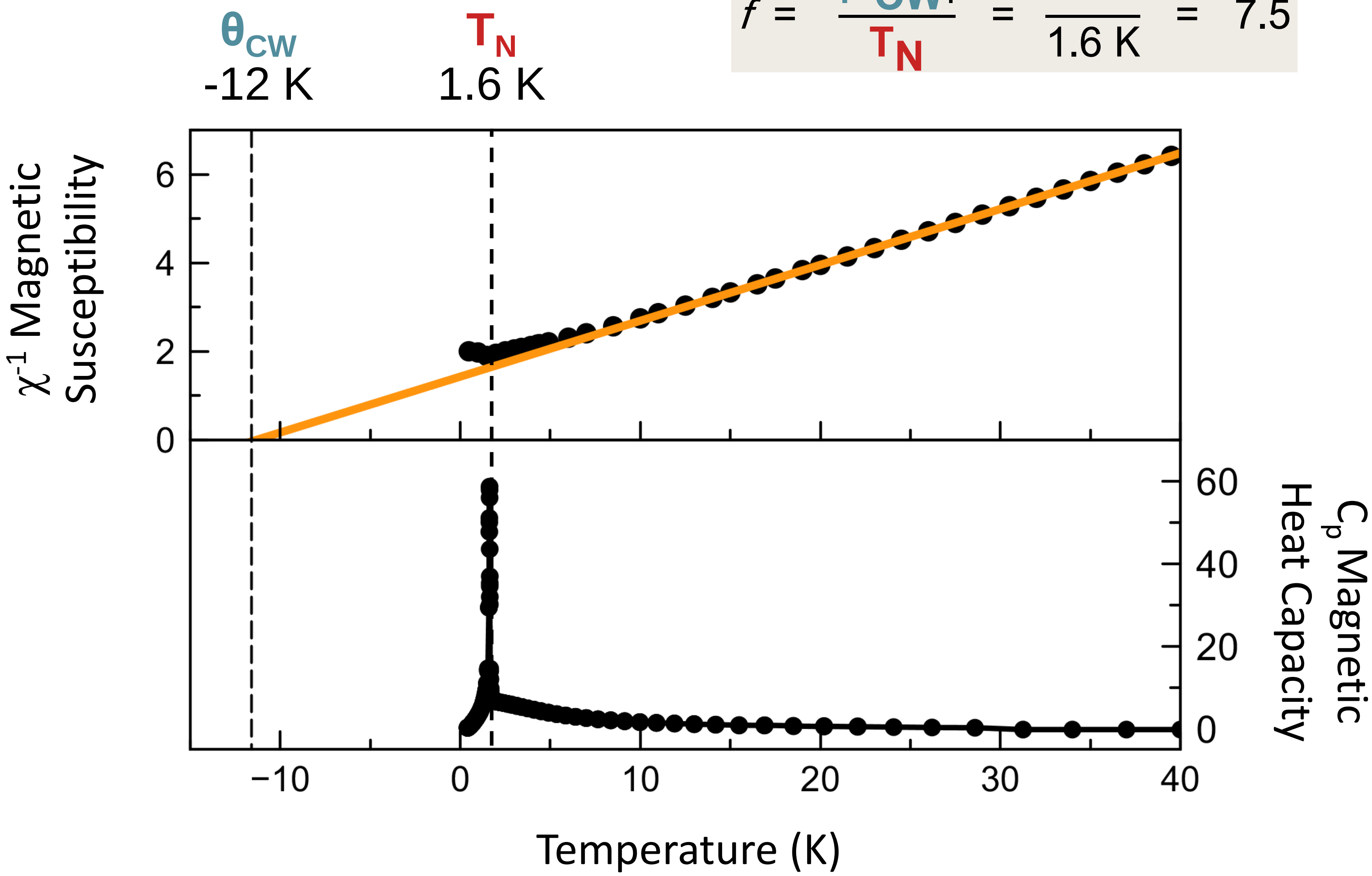
Wish list for an ideal magnetically frustrated material



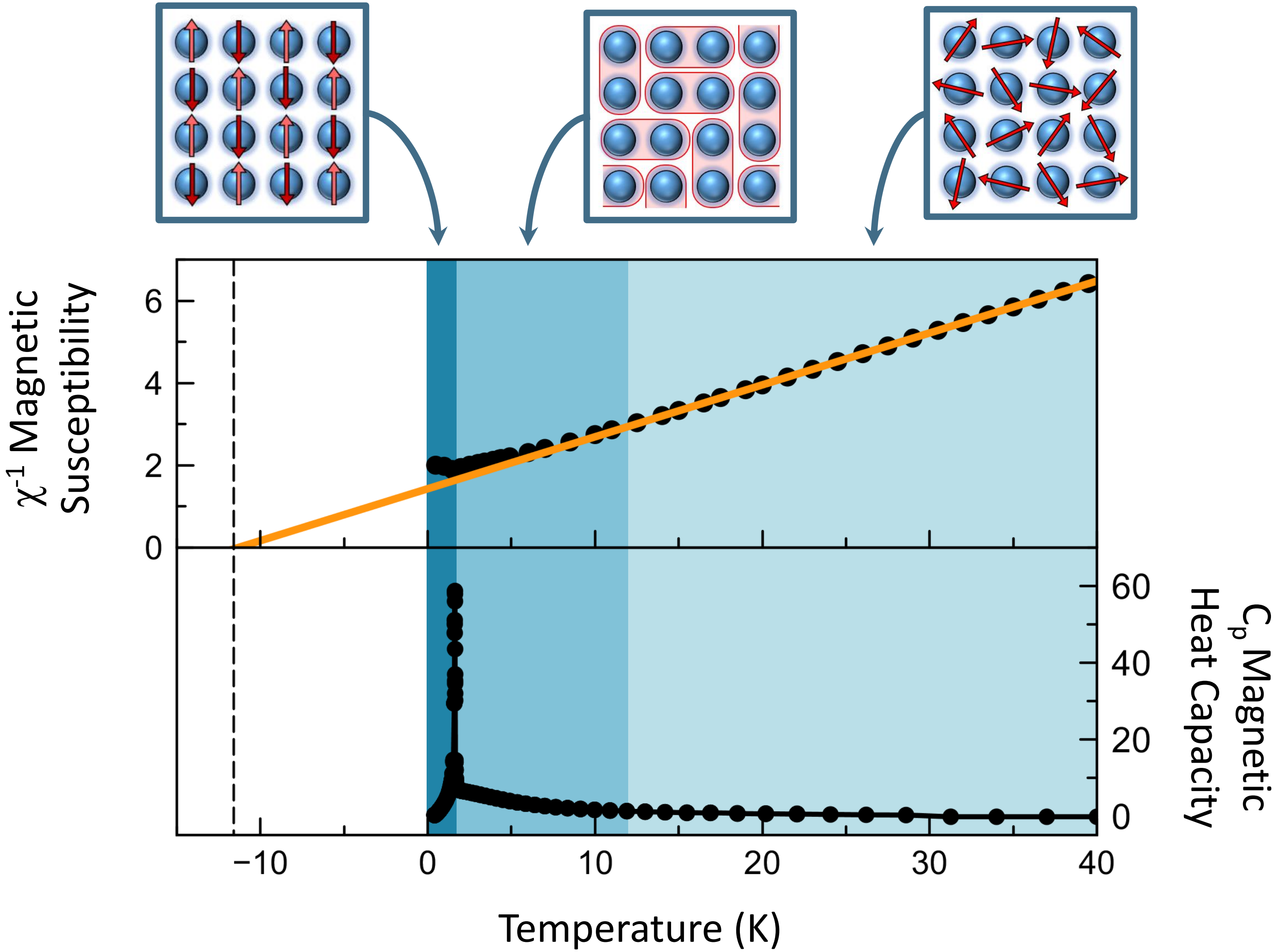
- Frustrated lattice occupied by a magnetic cation
- Only one magnetic site in the crystal structure
- Minimal chemical disorder
- **Can be grown as large single crystals** (enables anisotropic property measurements and scattering experiments)
- **Small spin ($S = \frac{1}{2}$) to maximize quantum fluctuations** (some exceptions)
- **Large spin-orbit coupling, $\lambda \sim Z^4$** (generates anisotropy)
- **Kramers' ion** (symmetry protected dipole moment)
- **Insulating** (don't want to worry about charge degrees of freedom)

We can use the frustration index, *f*, to assess if a material is frustrated. A system is empirically frustrated when *f* > 5.

$$f = \frac{|\theta_{CW}|}{T_N} = \frac{12\text{ K}}{1.6\text{ K}} = 7.5$$



For $\theta_{CW} < T < T_N$, the system is a cooperative paramagnet



How to study your new frustrated material

Does your material magnetically order? → pick your favourite probe!

- Peak in heat capacity
- Anomaly in susceptibility
- Bragg peaks in neutron diffraction
- Line splitting in NMR
- Oscillations in muon spin resonance

Are the spins static or dynamic?

- Longitudinal field response in muon spin resonance
- Frequency dependent ac susceptibility

Investigate the spin excitations!

- Inelastic neutron scattering
- Resonant inelastic x-ray scattering
- Thermal conductivity
- Electron spin resonance
- Terahertz spectroscopy

Connect with theory!

- Are there existing models that might be applicable to this material?
- What would a minimal Hamiltonian for this system look like?
- How should this ground state respond to magnetic field, pressure, chemical doping, disorder …

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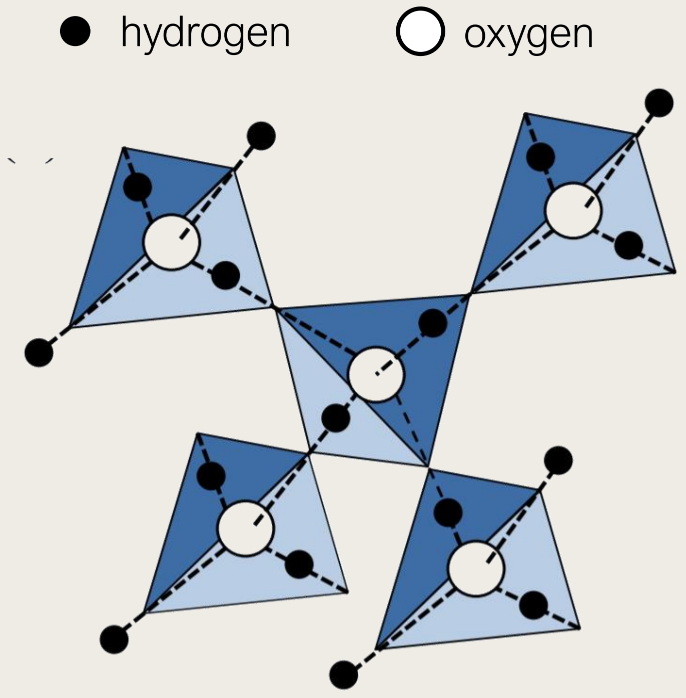
2. **Frustration in the classical limit (dipolar spin ice)**

3. Frustration in the quantum limit (quantum spin liquids)

4. Case Studies:
    - Proximate Kitaev spin liquid in α-$RuCl_3$
    - Phase competition in quantum XY pyrochlores

The pyrochlore $Dy_2Ti_2O_7$ has a dipolar spin ice ground state

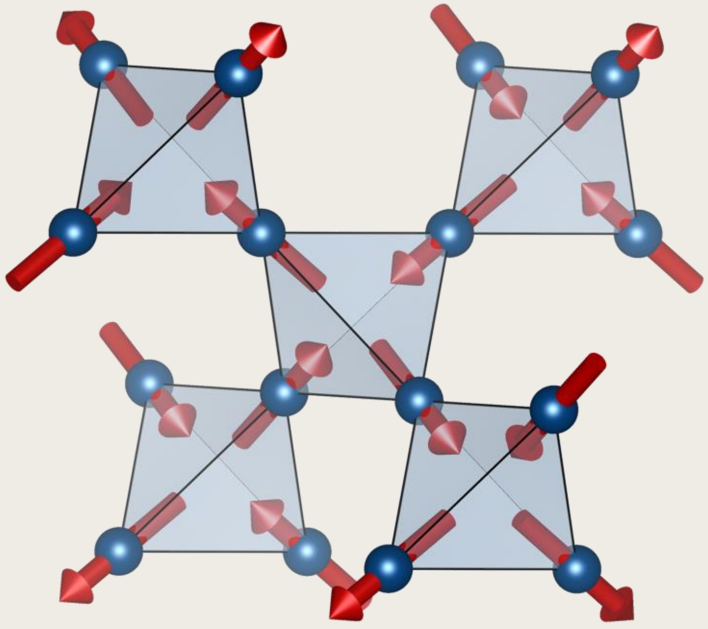
pyrochlore lattice + Ising anisotropy + net ferromagnetic interactions = spin ice



Water Ice

two-long/two-short

Dy^{3+}: $\mu_{eff} = 10\ \mu_B \rightarrow D_{nn}$ (FM) > J_{ex} (AFM)

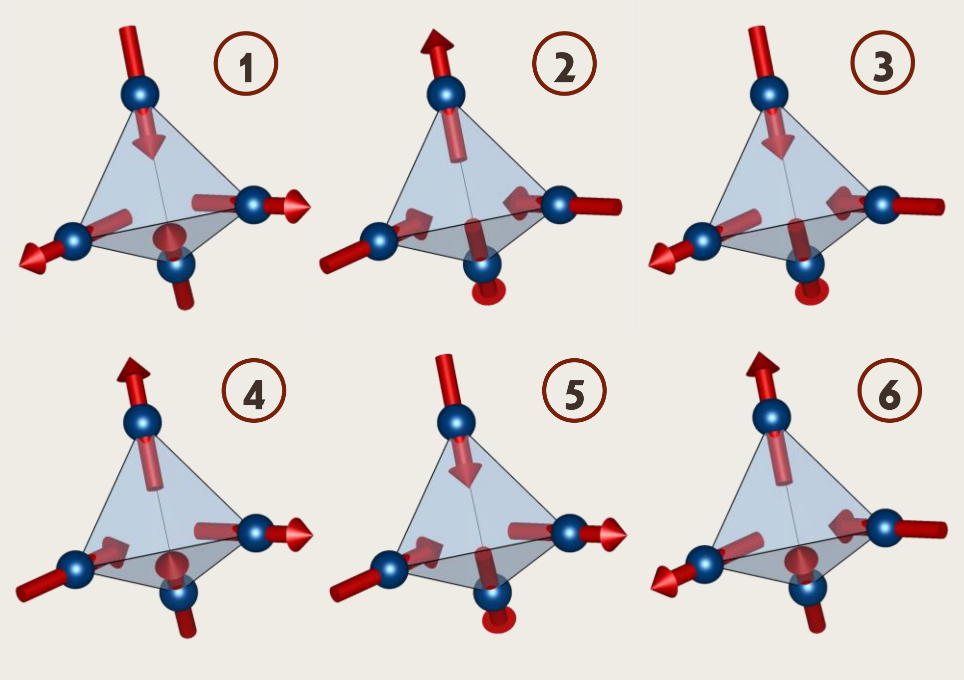


Spin Ice

two-in/two-out

n.b. Spin ice is a rare example of ferromagnetic frustration!

Frustration in spin ice originates from a macroscopic degeneracy

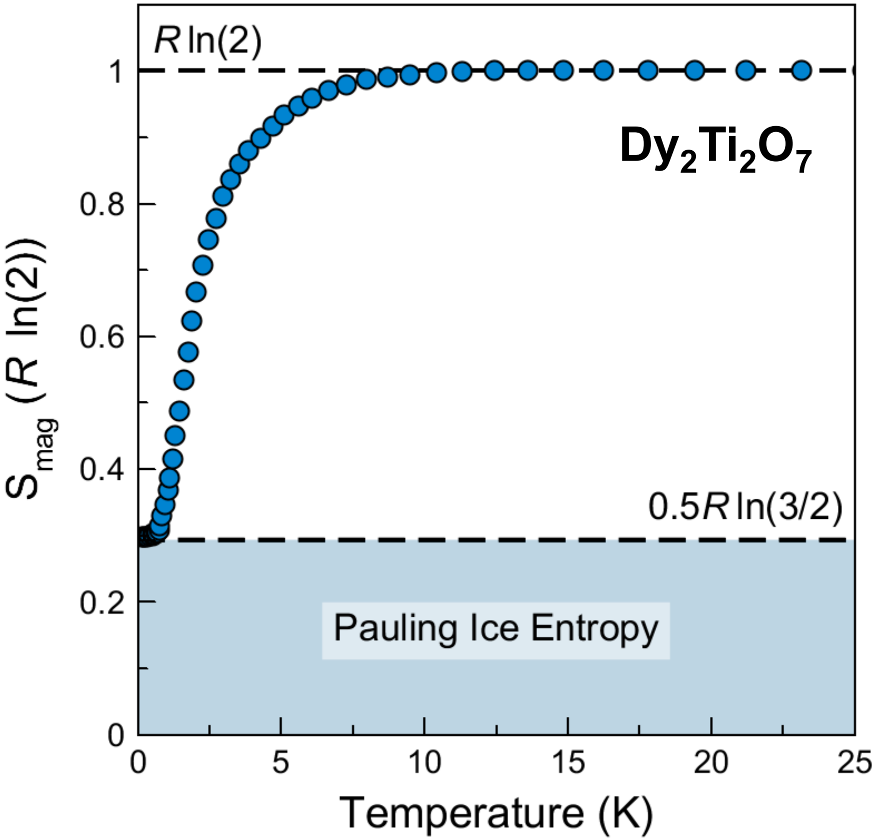


There are $2^4 = 16$ possible spin configurations for a single tetrahedron. Of these, 6 satisfy the two-in/two-out "ice rules".

For N spins and *N*/2 tetrahedra:

$$W = 2^N \cdot \left(\frac{6}{16}\right)^{\frac{N}{2}} = \left(\frac{3}{2}\right)^{\frac{N}{2}}$$

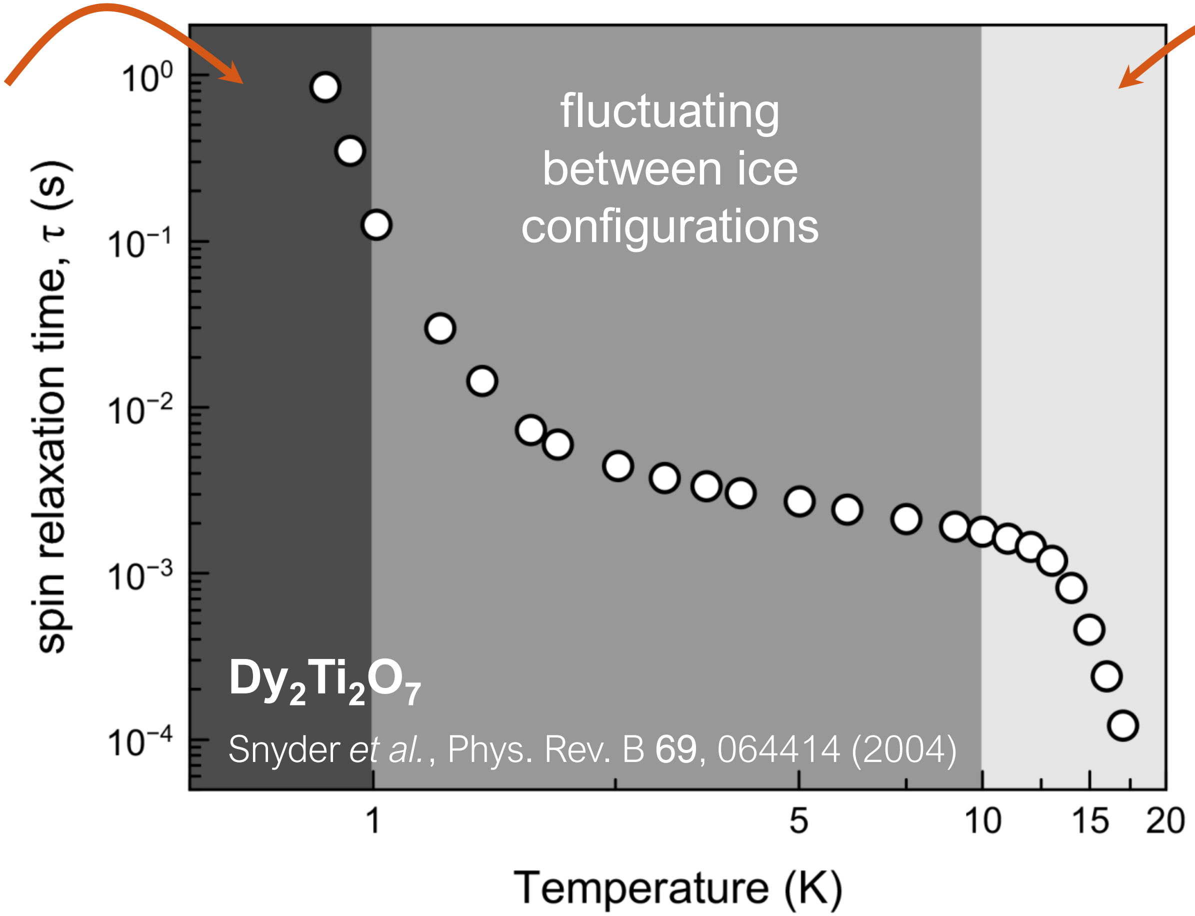
$$S = k_B \ln(W) = \frac{k_B N}{2} \ln\left(\frac{3}{2}\right)$$



Why is the spin ice state “classical”?

Quantum fluctuations are negligible* → spin freezing transition when thermal fluctuations become small

frozen into a single ice state, no fluctuations. NOT long range order



classical paramagnet

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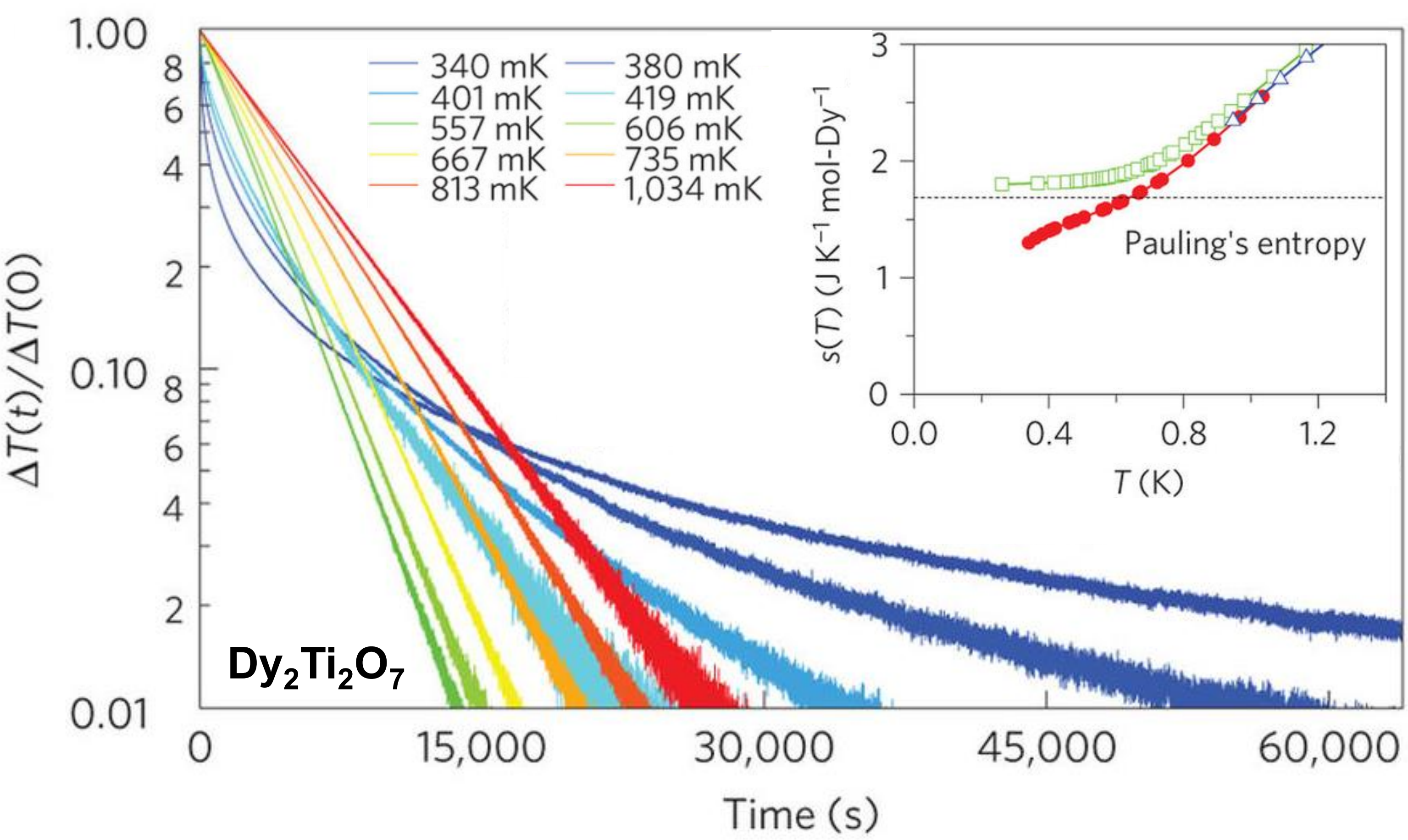
*Rau and Gingras “Magnitude of quantum effects in classical spin ices” PRB **92**, 144417 (2015)

In thermal equilibrium, the spin ice degeneracy is lifted

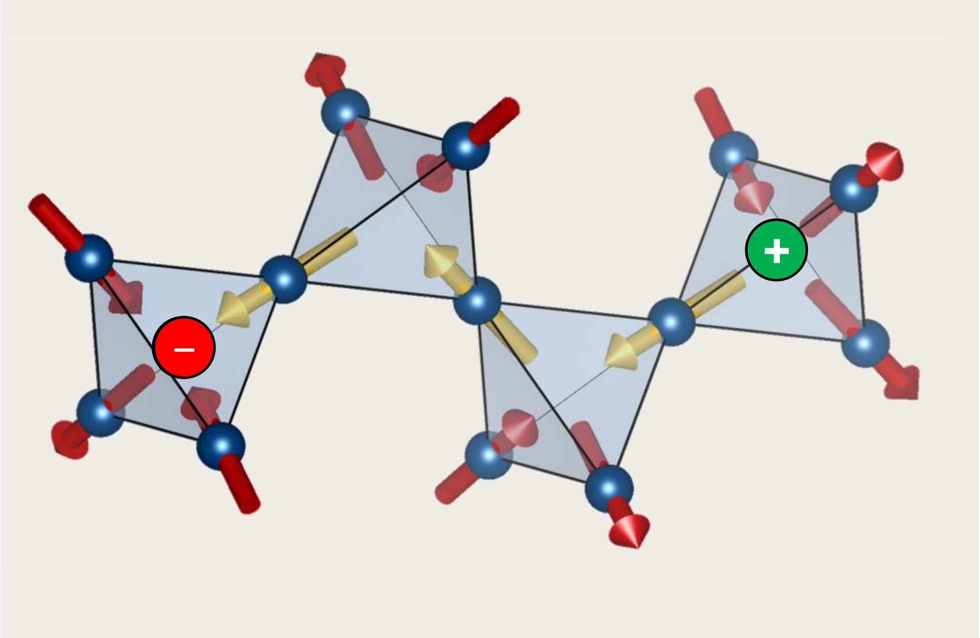
In equilibrium, $Dy_2Ti_2O_7$ would long range order at $T_C = 0.18$ K according to Monte Carlo simulations (Melko, PRL **87**, 067203 (2001))



Pomaranski *et al.*, Nature Physics **9**, 353-356 (2013)

The spin excitations in spin ice behave like magnetic monopoles!

A spin flip excitation in spin ice is analogous to creating a pair of monopoles, which can freely propagate away from one another.



The emergent magnetic monopoles in spin ice are connected via a Dirac string of flipped dipoles and interact via a magnetic Coulomb interaction

Castelnovo, Moessner, and Sondhi. Nature **451**, 42–45 (2008)

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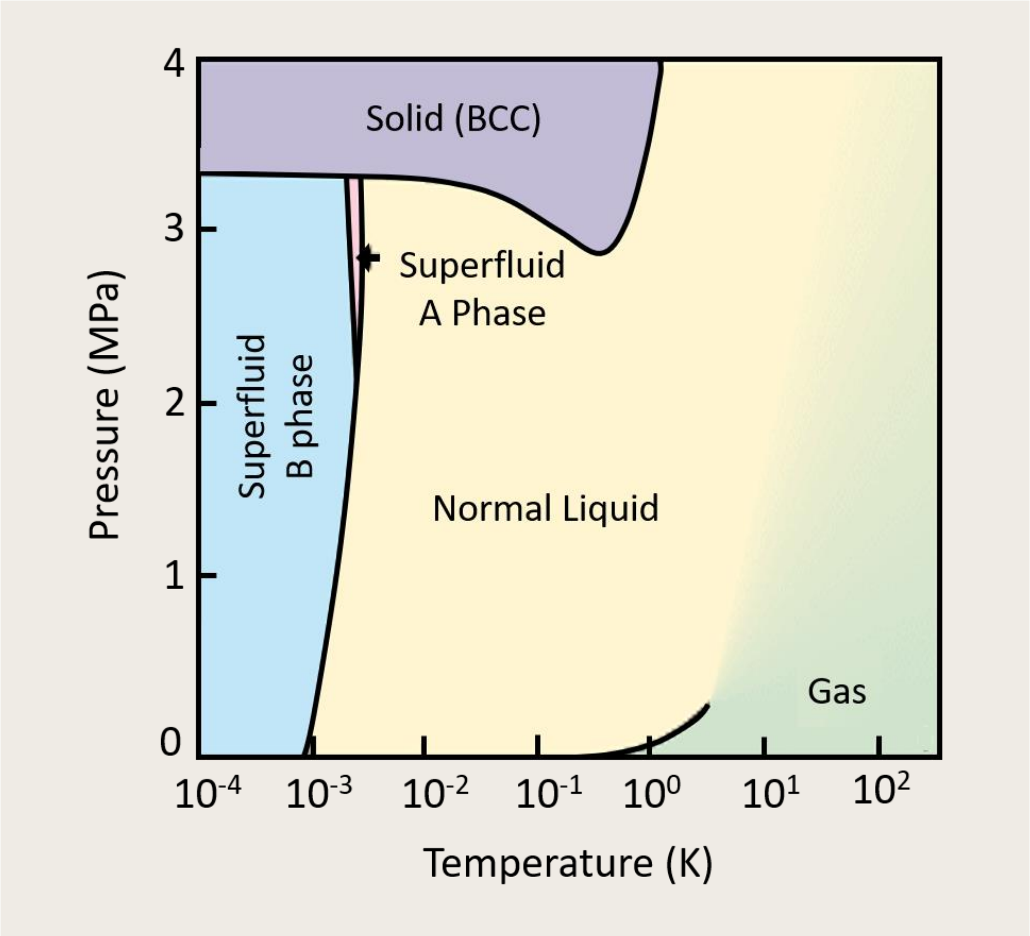
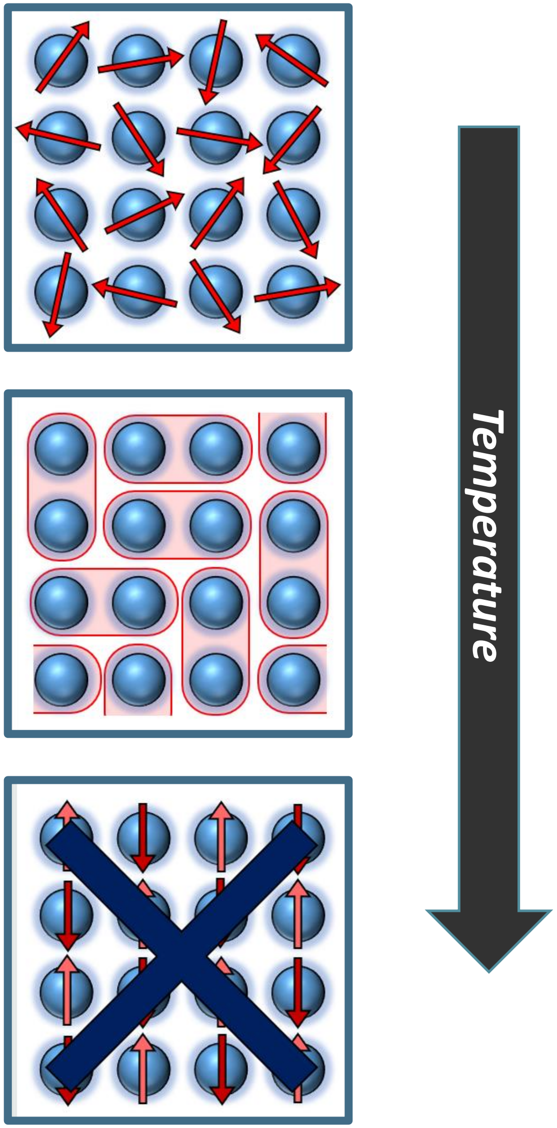
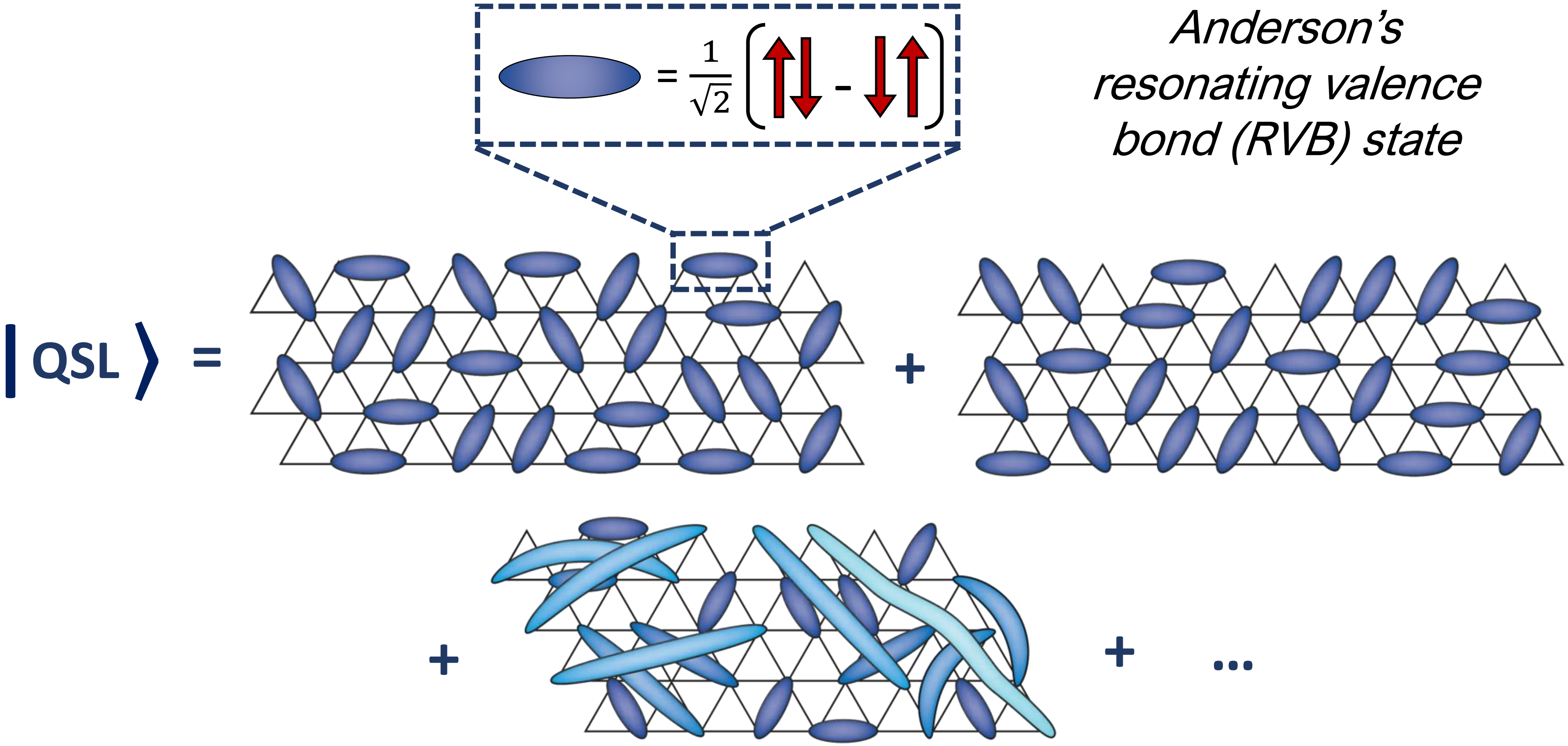
Spin liquids do not magnetically order or freeze down to 0 K

Analogy with He^3: fermion with large zero-point fluctuations, no solid phase at 1 atm.





Quantum spin liquids are a highly entangled state of matter

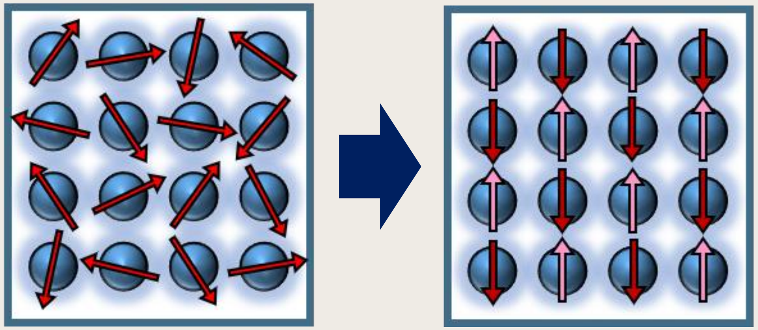


Anderson's resonating valence bond (RVB) state

Savary and Balents, *Quantum spin liquids: a review*
Reports on Progress in Physics 80.1 016502 (2016)

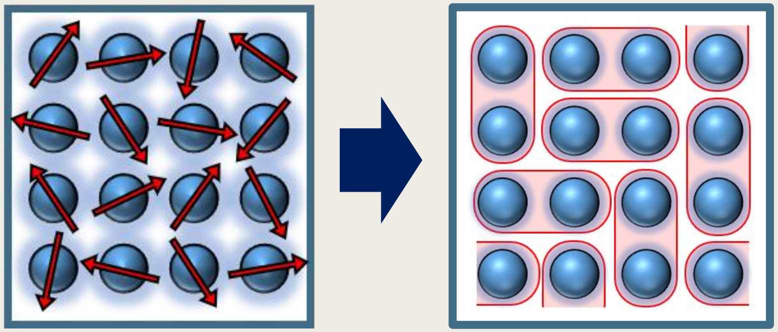
There is (currently) no experimental proof of a QSL state

Normal Ordered Magnet: Symmetry Breaking Transition



- ✓ Peak in heat capacity
- ✓ Cusp in susceptibility
- ✓ Magnetic Bragg peaks
- ✓ Oscillations in μSR

Quantum Spin Liquid: No Symmetry Breaking

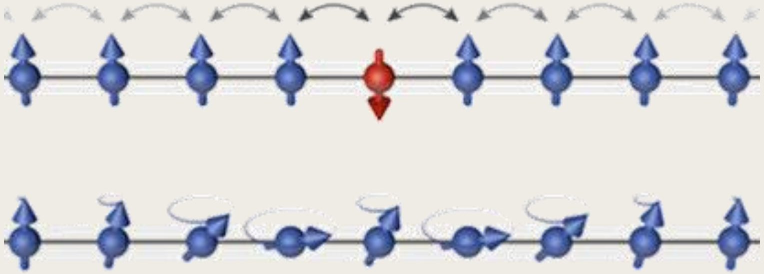


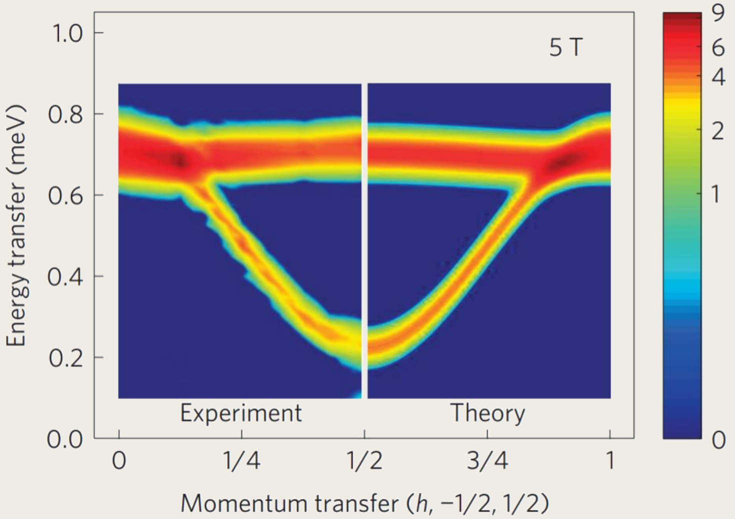
- ✕ No definitive experimental signature
- ✕ Absence of order and freezing
- ✕ Entanglement entropy?

No experimental smoking gun for a QSL state but fractionalized excitations a good starting point (for now).

Normal Ordered Magnet

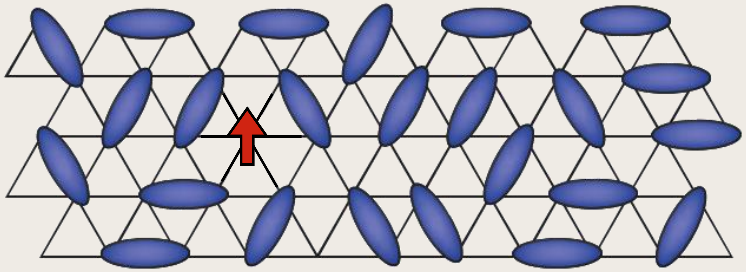
Magnon ($S = 1$) Excitations

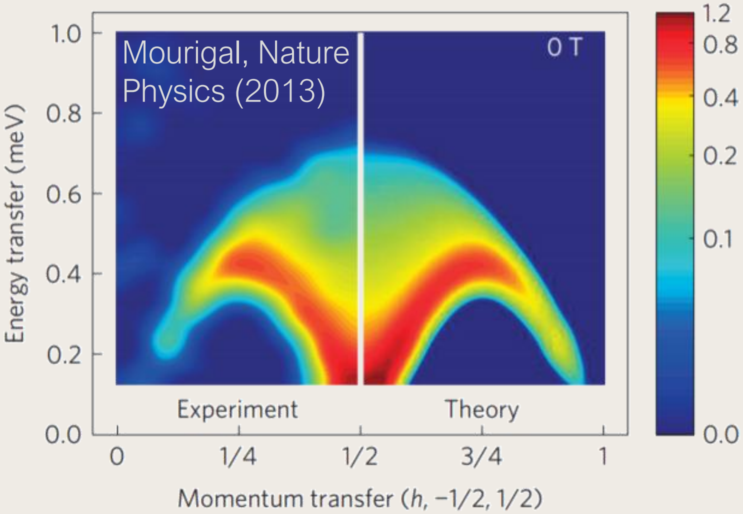




Quantum Spin Liquid

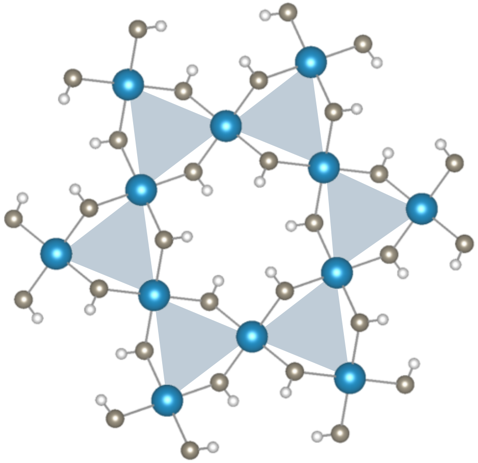
Fractional ($S = ½$) Excitations



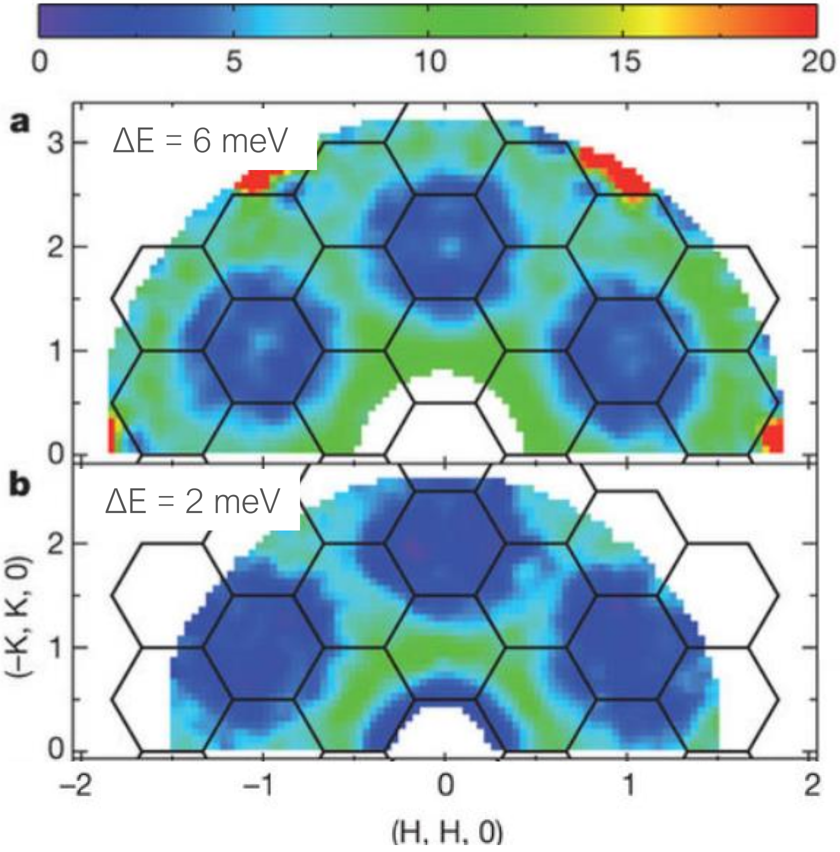


Mourigal, Nature Physics (2013)

Herbertsmithite is a strong quantum spin liquid candidate



Han et al., Nature **492**, 406–410 (2012).



Herbertsmithite has a "perfect" Kagome lattice of Cu^{2+} ($S = ½$).

There is a strong theoretical case that the ground state of an AFM $S = ½$ Heisenberg Kagome is a QSL.

Continuum of scattering indicates fractionalized excitations – gapped or gapless, role of disorder?

Theory for S = ½ Heisenberg Kagome AFM:
Yan, Huse, and White. Science, **333** 1173-1176 (2011).
Jiang, Wang, and Balents. Nat. Physics **8**, 902–905 (2012)

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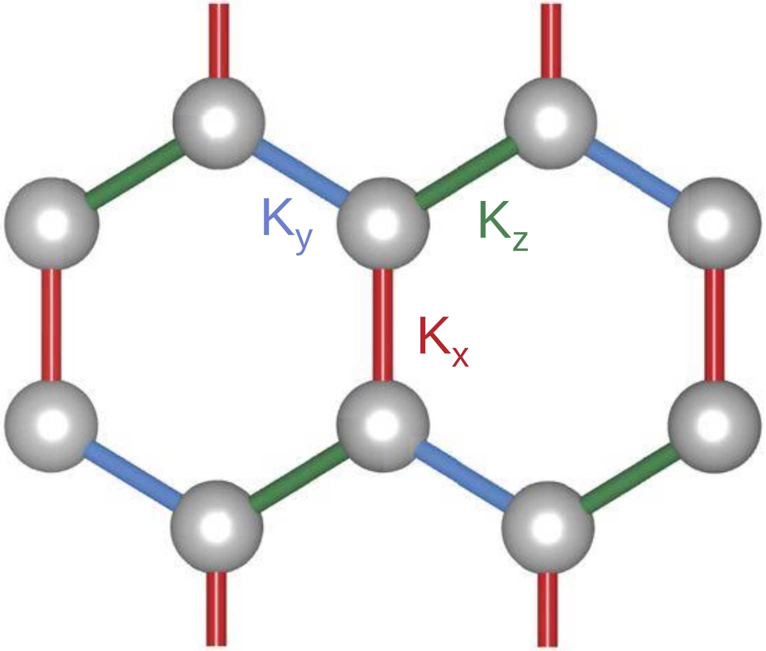
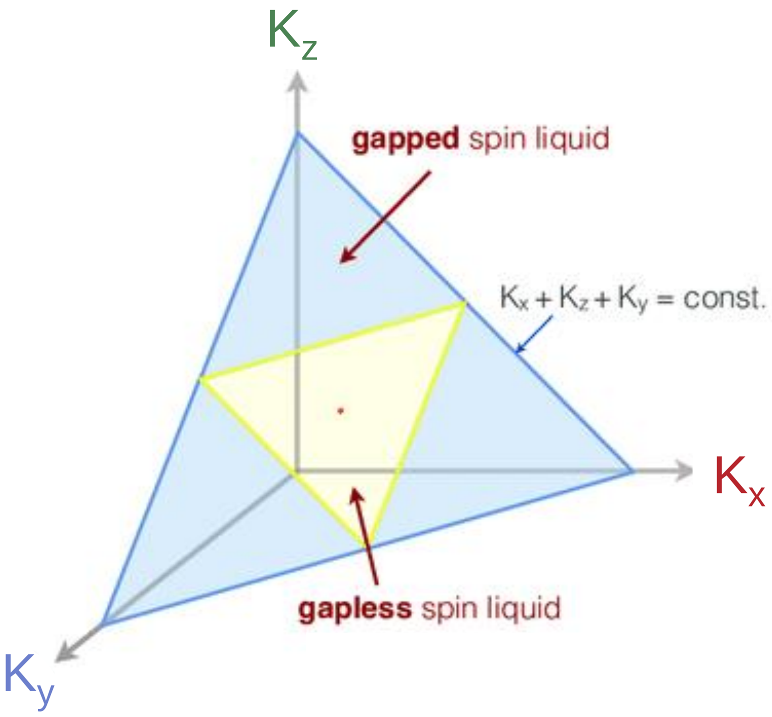
e.g. 1

The Kitaev model introduces bond-dependent Ising exchange on a Honeycomb lattice

$$H = K_x \sum_{x-\text{bonds}} S_i^x S_j^x + K_y \sum_{y-\text{bonds}} S_i^y S_j^y + K_z \sum_{z-\text{bonds}} S_i^z S_j^z$$





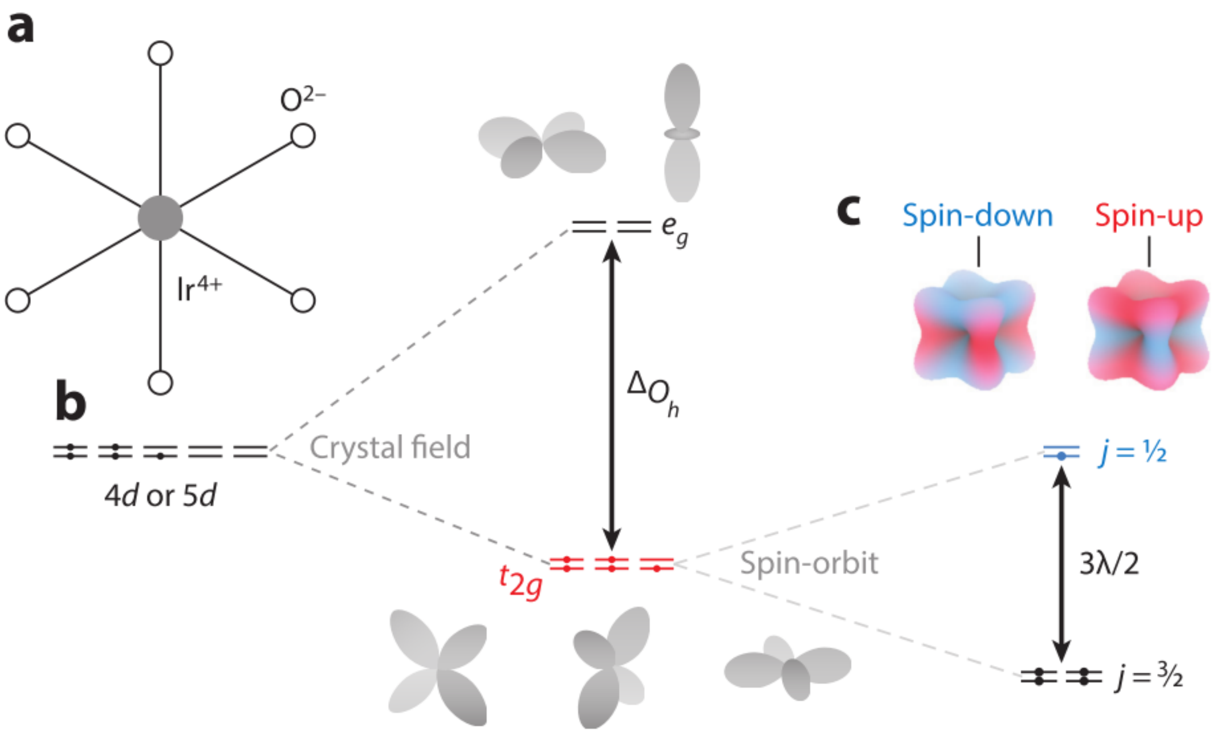
The Kitaev model is exactly solvable and gives both gapped and gapless spin liquid ground states!

Kitaev. Annals of Physics **321**, 2-111 (2006)
Trebst. arXiv:1701.07056 (2017)
Banerjee *et al.*, Science **356** 1055-1059 (2017)

e.g. 1

Kitaev-like exchange can actually be realized in some ions with large spin-orbit-coupling, such as Ir^{4+} and Ru^{3+}

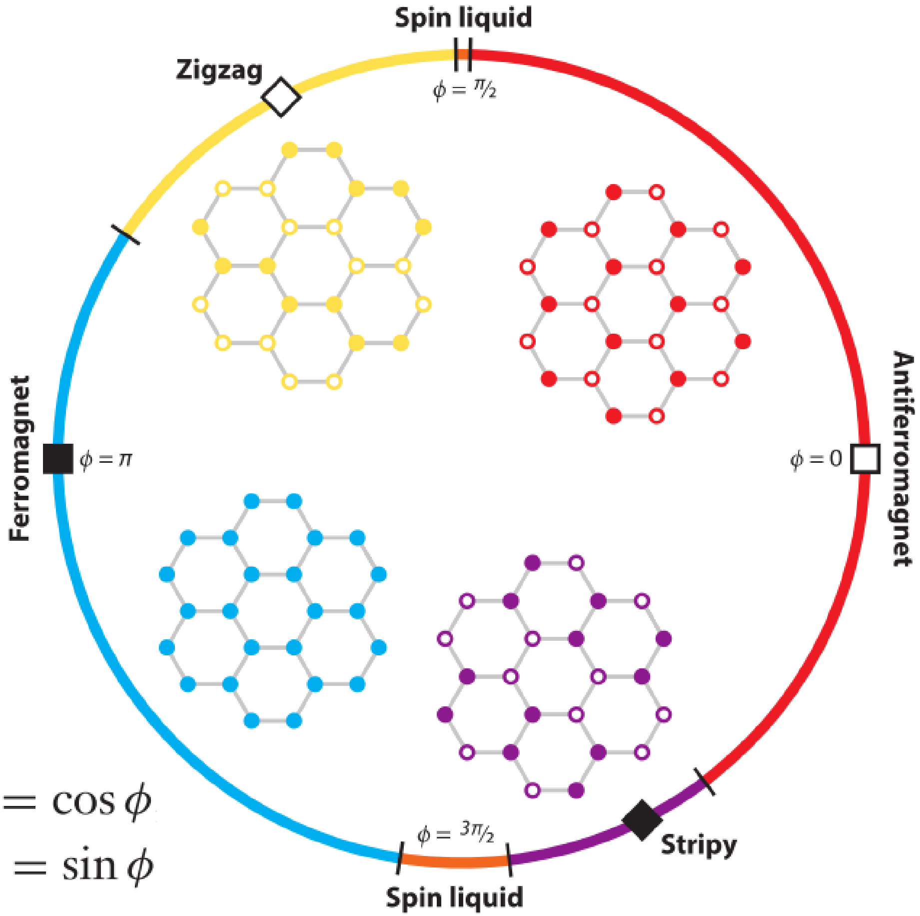
$$H = K_x \sum_{x-\text{bonds}} S_i^x S_j^x + K_y \sum_{y-\text{bonds}} S_i^y S_j^y + K_z \sum_{z-\text{bonds}} S_i^z S_j^z$$



Rau, Lee, and Kee. Ann. Rev. Cond. Matt Physics **7** 195-221 (2016)

e.g. 1

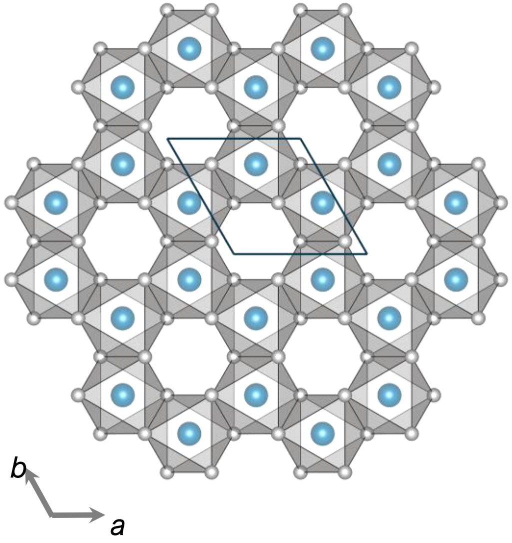
In materials with edge sharing octahedral lattices, Heisenberg exchange is suppressed and Kitaev interactions can dominate!



$$J = \cos\phi$$
$$K = \sin\phi$$

Heisenberg *Kitaev*

$$\mathbf{H} = J \sum_{ij} S_i S_j + K_\gamma \sum_{\gamma\text{-bonds}} S_i^\gamma S_j^\gamma$$

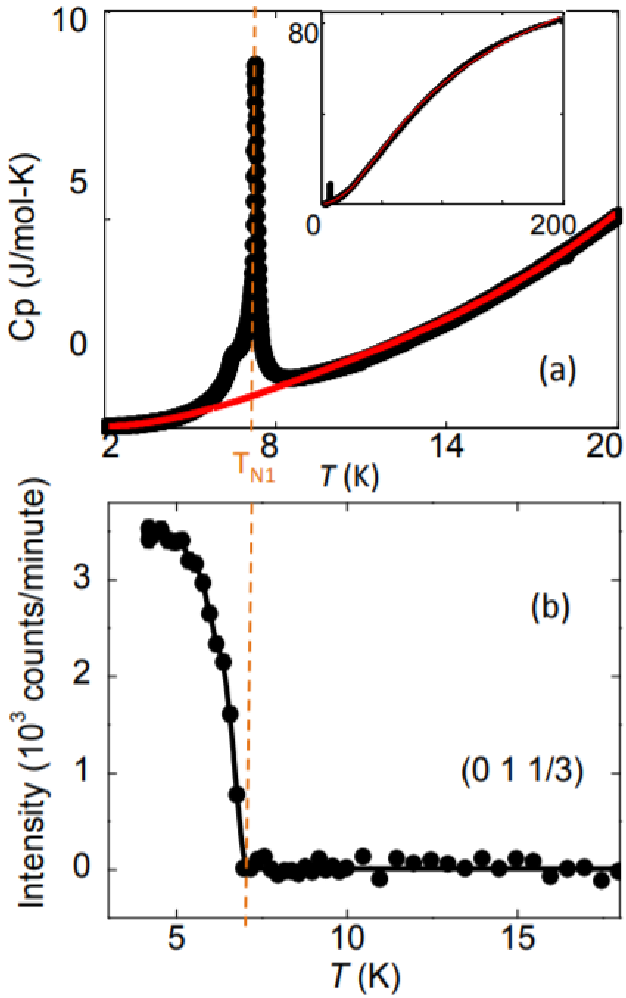


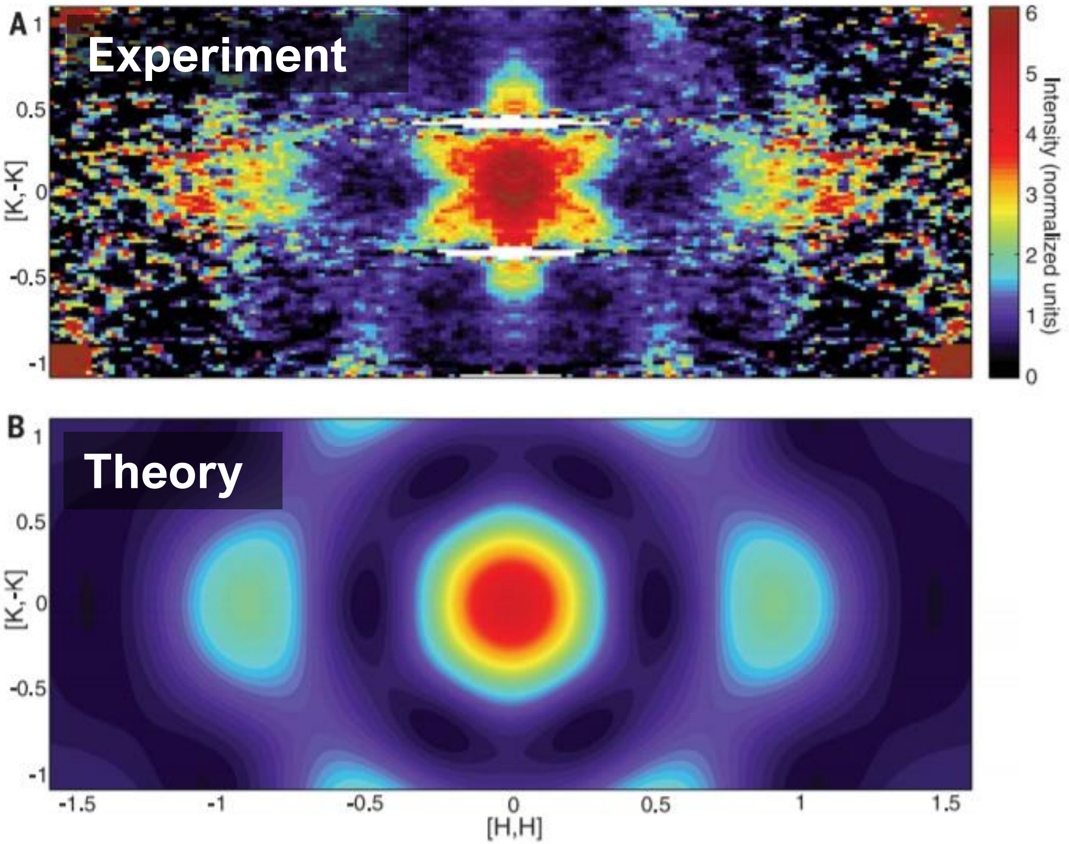
Examples: Na_2IrO_3, α-Li_2IrO_3, and α-$RuCl_3$

Jackeli and Khaliullin Phys. Rev. Lett. **102**, 017205 (2009)
Rau, Lee, and Kee. Ann. Rev. Cond. Matt Physics **7** 195-221 (2016)

***e.g.* 1**

Although α-RuCl$_3$ is not a spin liquid (T_N = 7 K), inelastic neutron scattering has revealed proximate Kitaev behavior

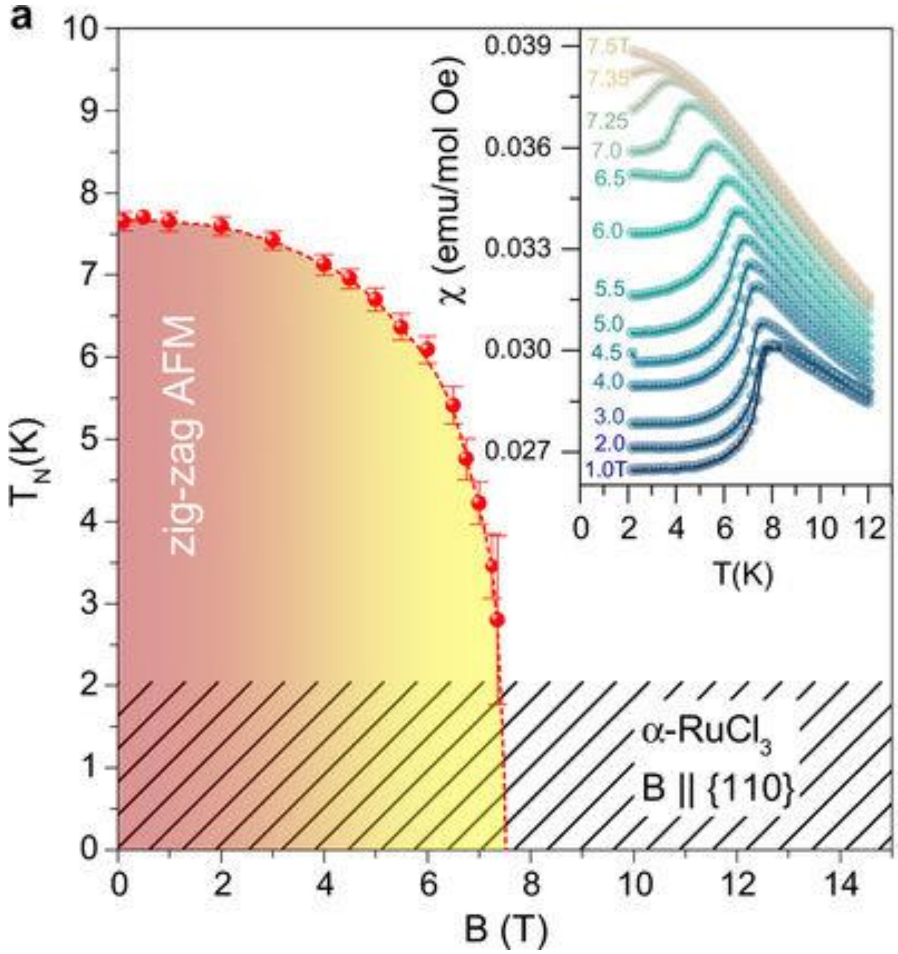


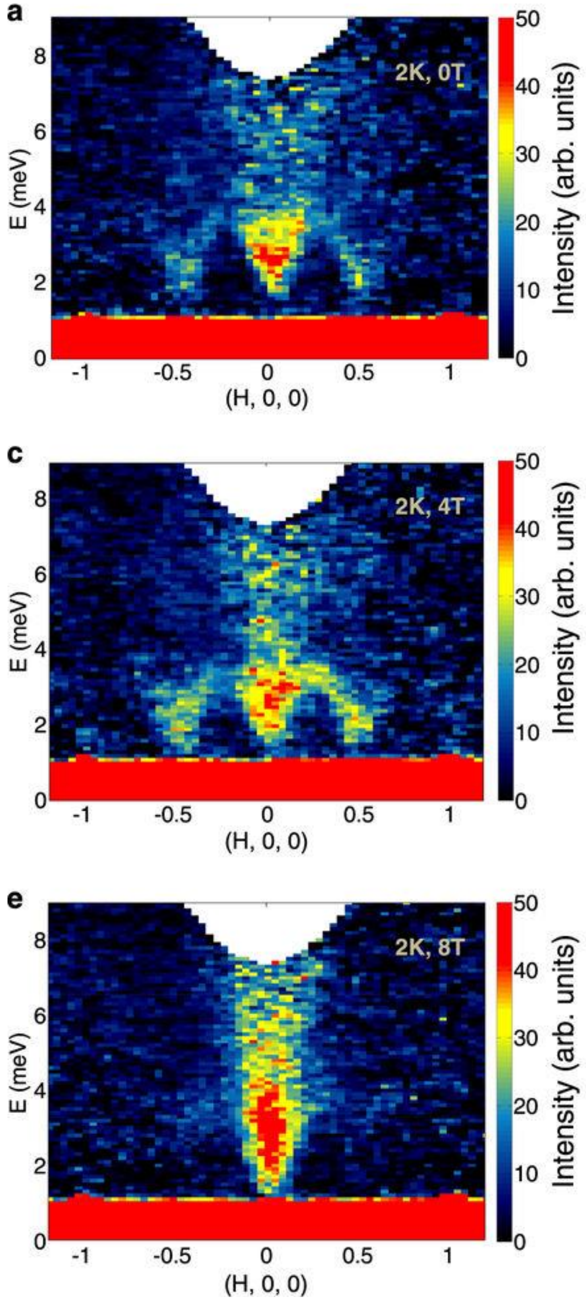


Cao *et al.*, Phys. Rev. B **93**, 134423 (2016)
Banerjee *et al.*, Science **356** 1055-1059 (2017)

e.g. 1

Recent experiments point to a field-induced QSL state in α-$RuCl_3$!

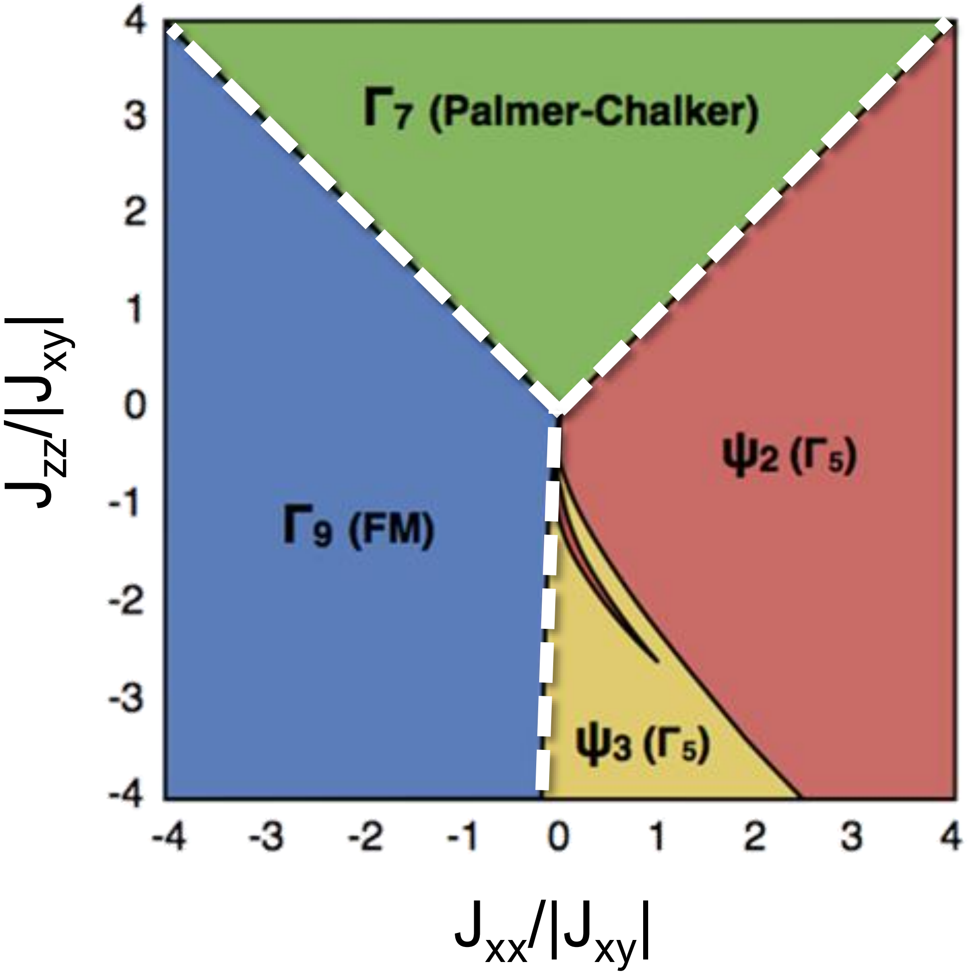




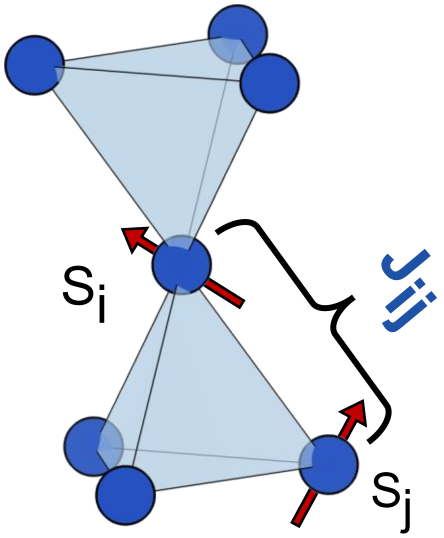
Banerjee *et al. npj Quantum Materials* **3**, *8 (2018)*.

e.g. 2

The XY pyrochlores inhabit a rich phase space. Strong phase competition occurs at the boundaries.



$$H = \sum S_i^a \mathbf{J_{ij}} S_j^b$$

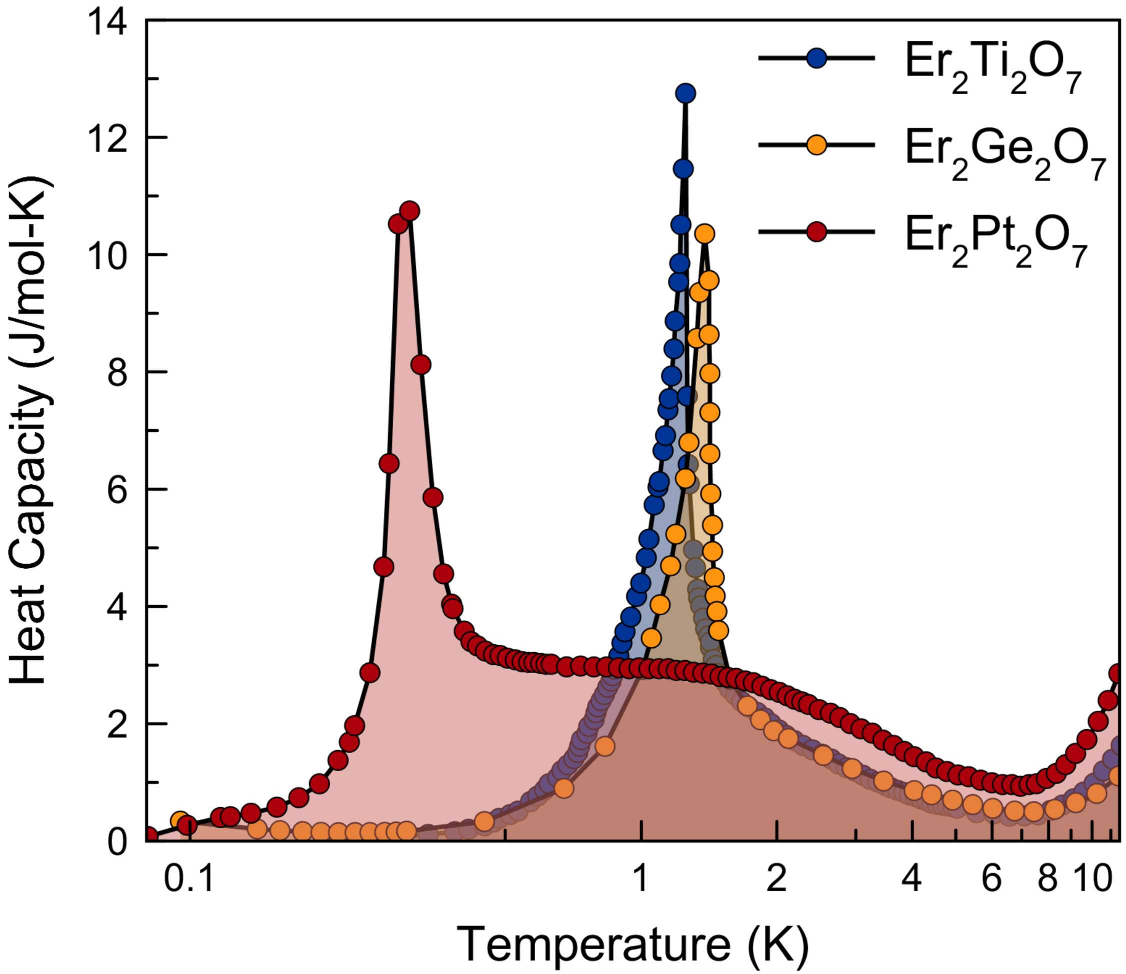


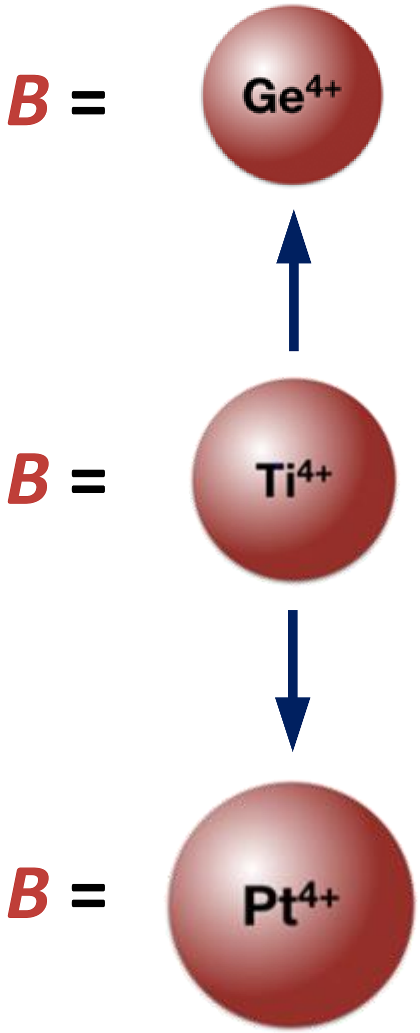
$$\mathbf{J_{ij}} = \begin{bmatrix} J_{zz} & 0 & 0 \\ 0 & J_{xx} & J_{xy} \\ 0 & J_{xy} & J_{xx} \end{bmatrix}$$

Yan *et al.*, PRB **95**, 094422 (2017).

e.g. 2

In the $Er_2B_2O_7$ XY pyrochlores, replacement of Ti by Pt increases the frustration index by 4x.

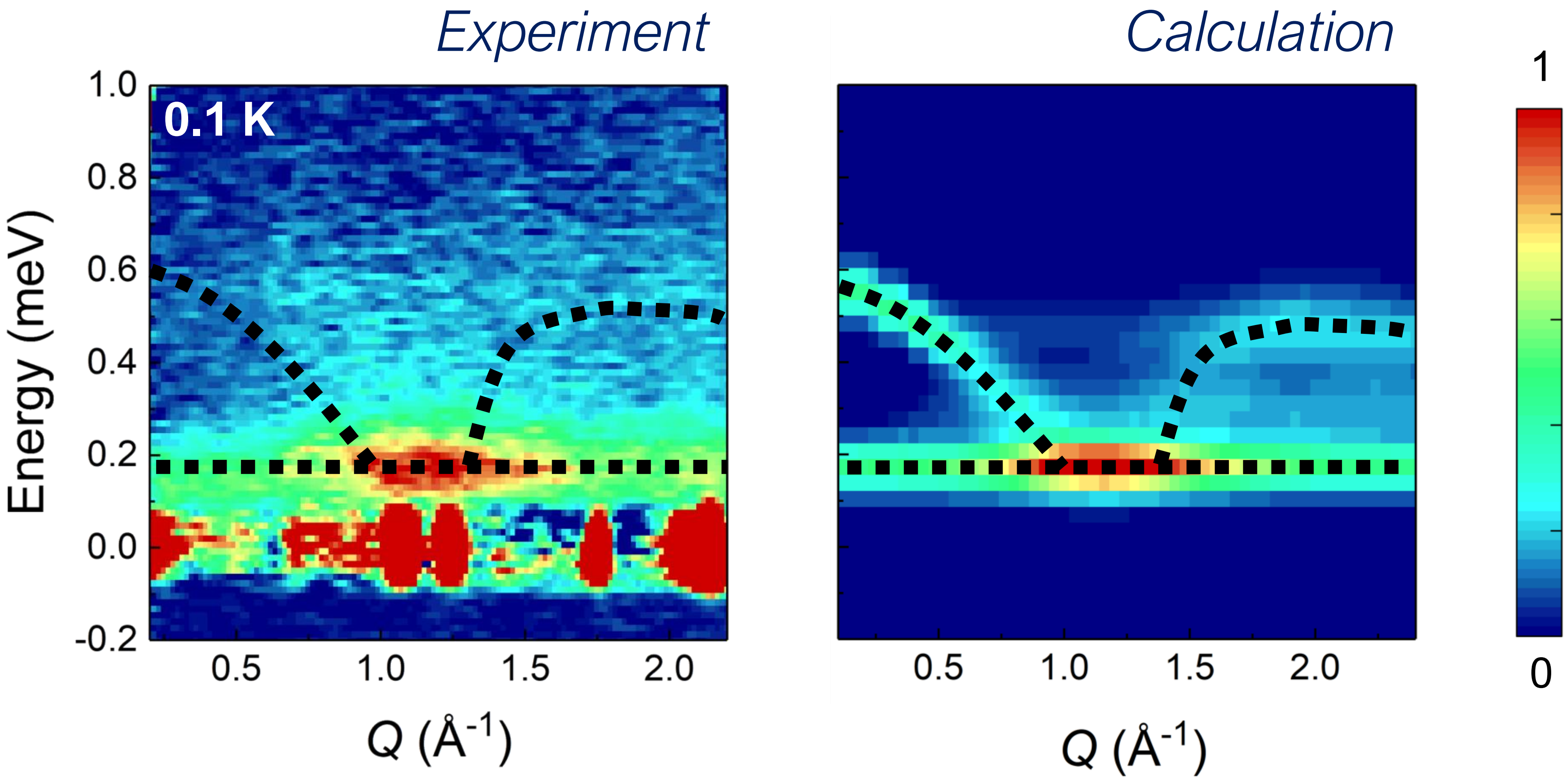




Hallas, Gaudet, and Gaulin. Ann. Rev. Cond. Matt. Physics 9 105-124 *(2018)*

e.g. 2

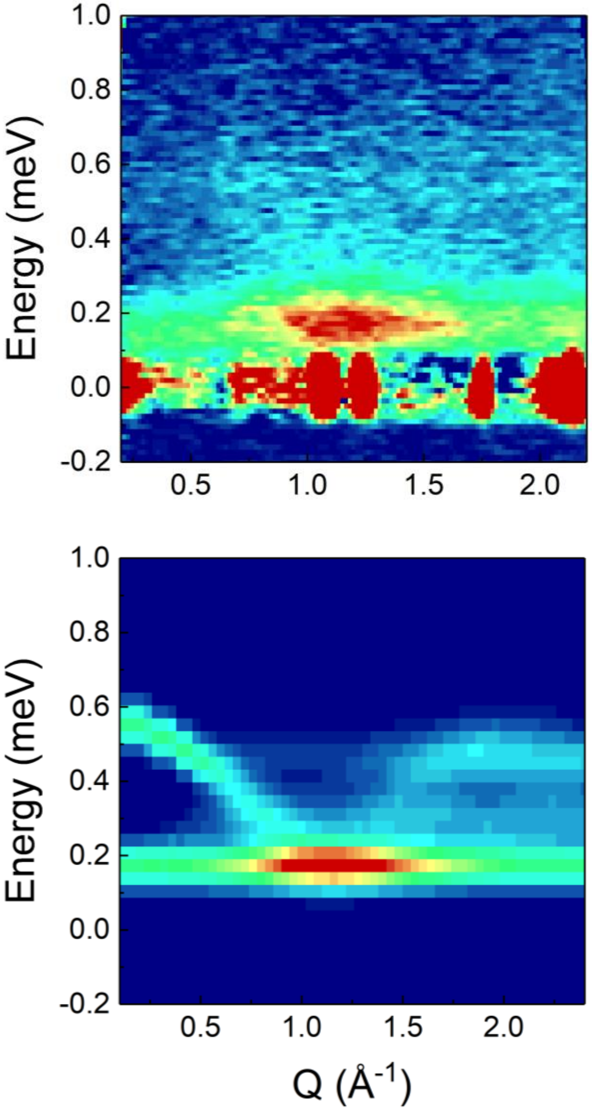
Inelastic neutron scattering measurements reveal a dispersive spin wave branch is missing in $Er_2Pt_2O_7$

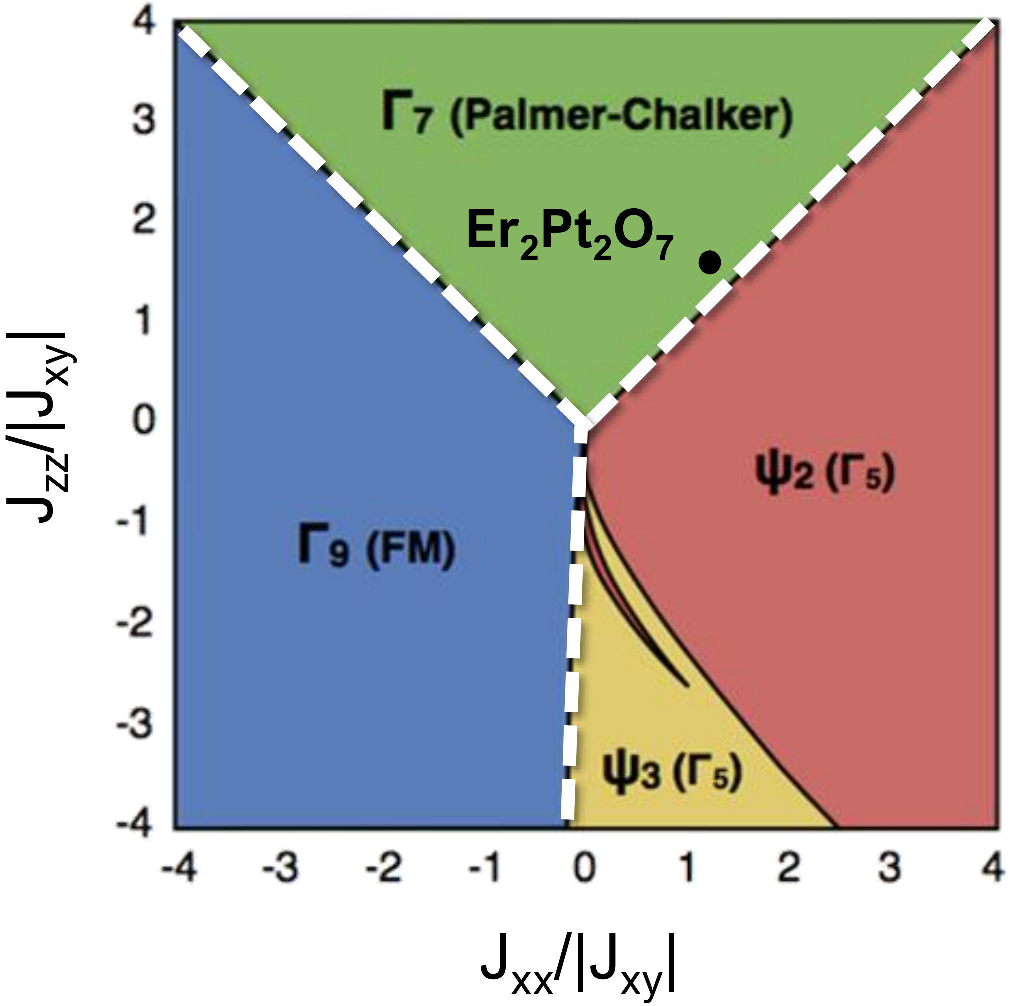


Hallas *et al.* Phys. Rev. Lett. **119**, 187201 (2017).

e.g. 2

$Er_2Pt_2O_7$ is in close proximity to a phase competition induced spin liquid state





Hallas *et al.* Phys. Rev. Lett. **119**, 187201 (2017).

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