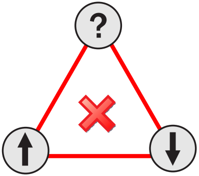
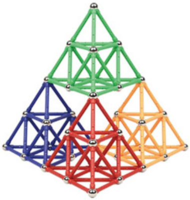
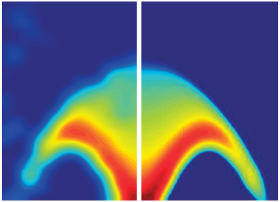
Quantum spin liquids: where to find them, and how?

Alexander Tsirlin

Felix Bloch Institute of Solid-State Physics, Leipzig University







TRR360 Young Researcher Forum
November 20, 2024

UNIVERSITÄT LEIPZIG

Where to search?

How to detect?

Where to search?

How to detect?

Where to search exactly?

How to ensure it is really a spin liquid?

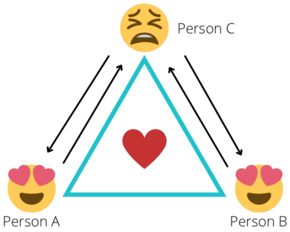
Where?...

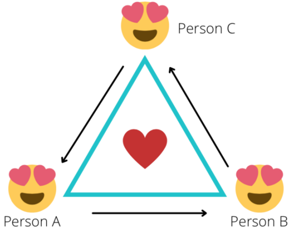
How?...

Where?...

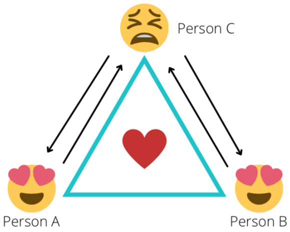
How?...

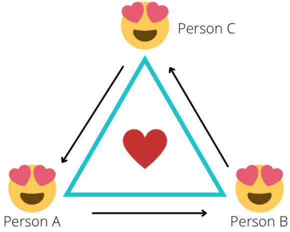
Triangle? Not this one...





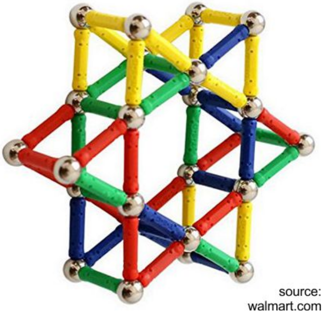
Triangle? Not this one...

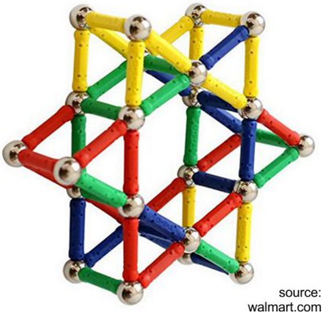




a lot of
frustration too...

Image credits: Britton Perelman (fair use), Mpelletier1 and Joan Lanfranco (CC-BY-SA)


source:
walmart.com



source: walmart.com

Magnastix Educational Magnetic Sticks Building Blocks Toys - Brain Toys, Family Fun for all Ages

★★★★★ Write a review SmaritLLC

This item is no longer available

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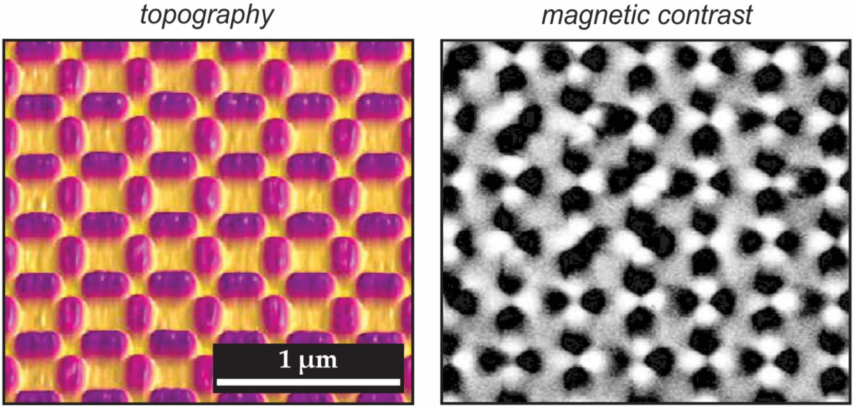
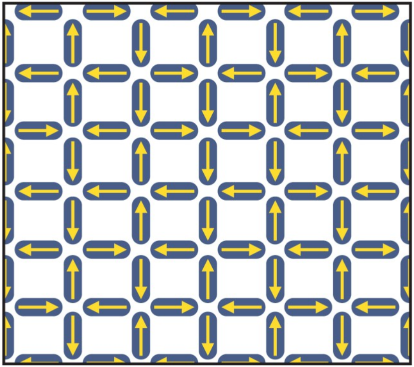
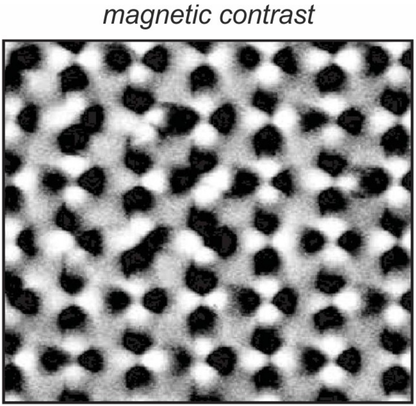


Nanosized islands made of a ferromagnetic Ni-Fe alloy

Large moments $\longrightarrow$ strong dipole-dipole interactions

Phys. Today 69(7), 54 (2016)





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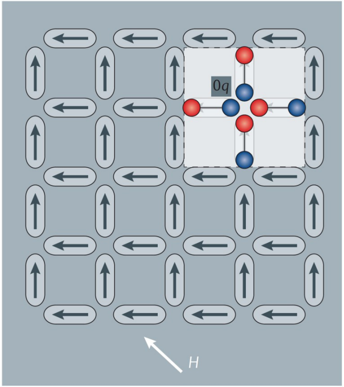
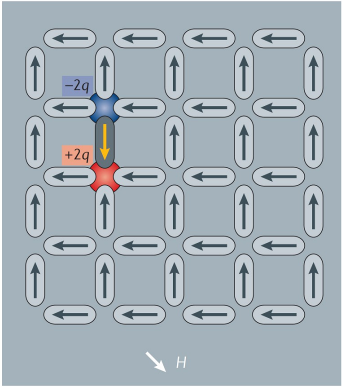
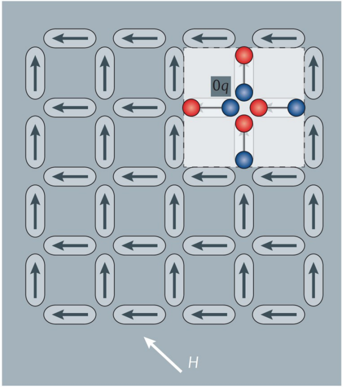
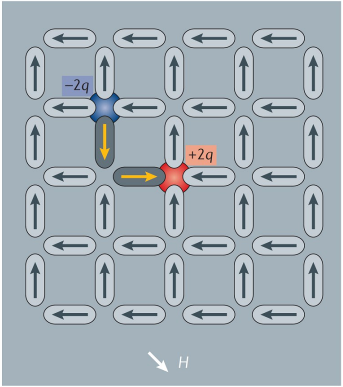
Nature Rev. Phys. 2, 13 (2020)



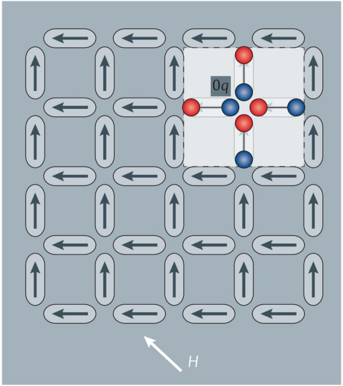


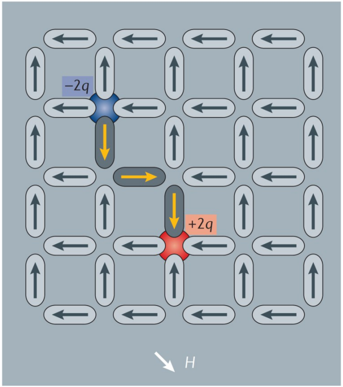
Nature Rev. Phys. 2, 13 (2020)



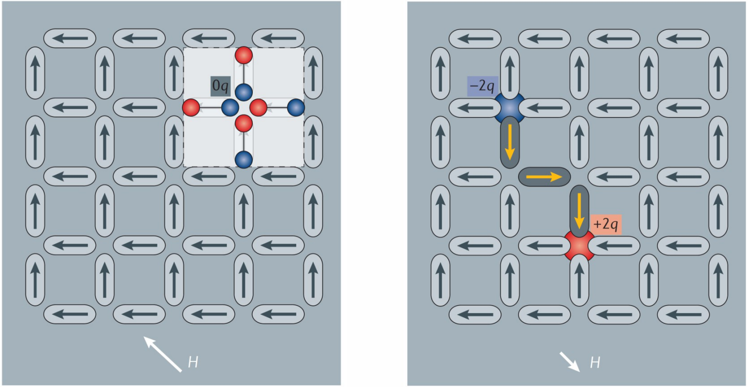


Nature Rev. Phys. 2, 13 (2020)





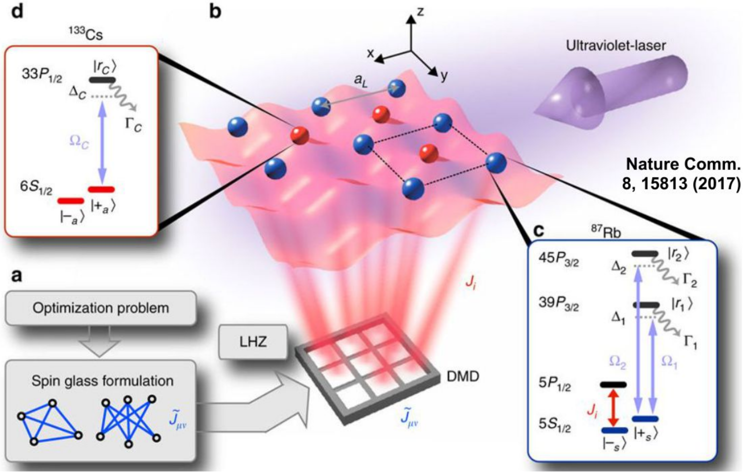
Nature Rev. Phys. 2, 13 (2020)



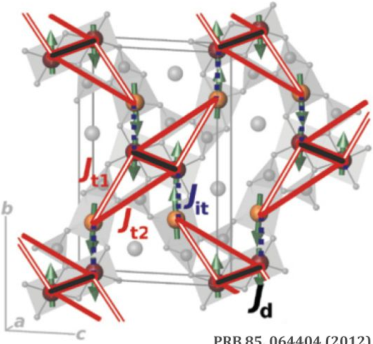
Large magnetic moments → no quantum effects

Further reading:

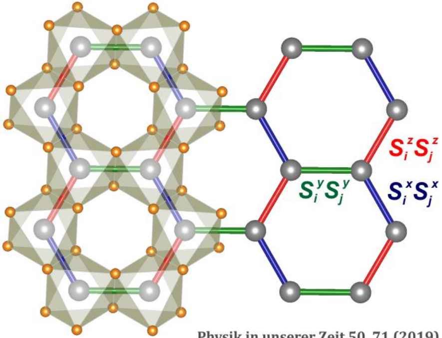
- I. Gilbert, C. Nisoli, P. Schiffer, *Physics Today* **69**, 54 (2016)
- S. Skjœrvø *et al.* *Nature Rev. Phys.* **2**, 13 (2020)



Plenty of interesting physics, but no spin liquids so far

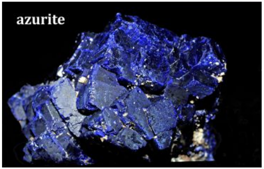


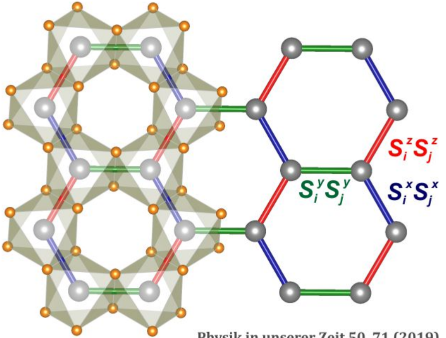
PRB 85, 064404 (2012)



Physik in unserer Zeit 50, 71 (2019)

Crystals of magnetic compounds are hitherto
the best experimental realization of frustrated magnets,
including quantum-spin-liquid candidates





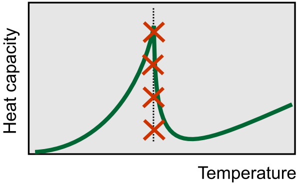
Physik in unserer Zeit 50, 71 (2019)

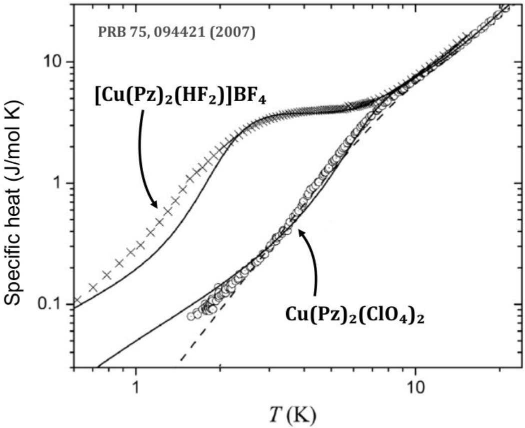
Crystals of magnetic compounds are hitherto
the best experimental realization of frustrated magnets,
including quantum-spin-liquid candidates

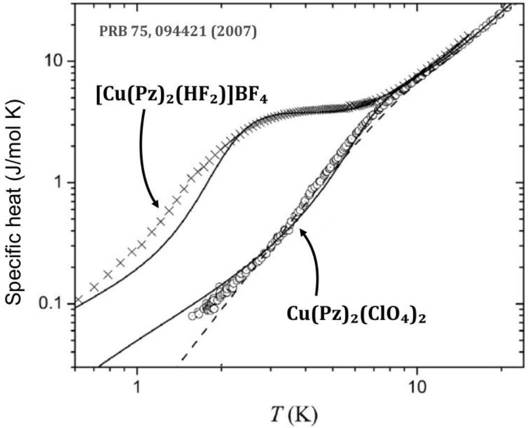
How to detect the spin liquid?

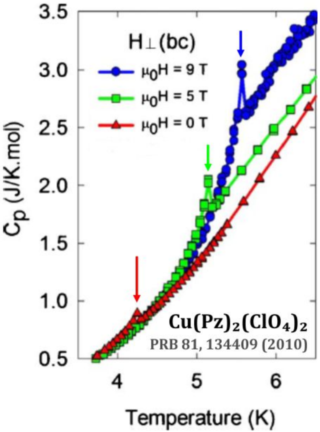
Stage 1

absence of magnetic order

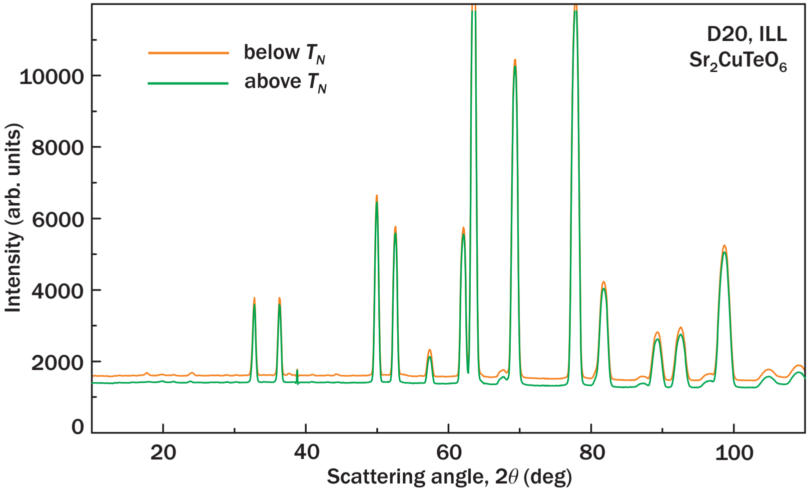


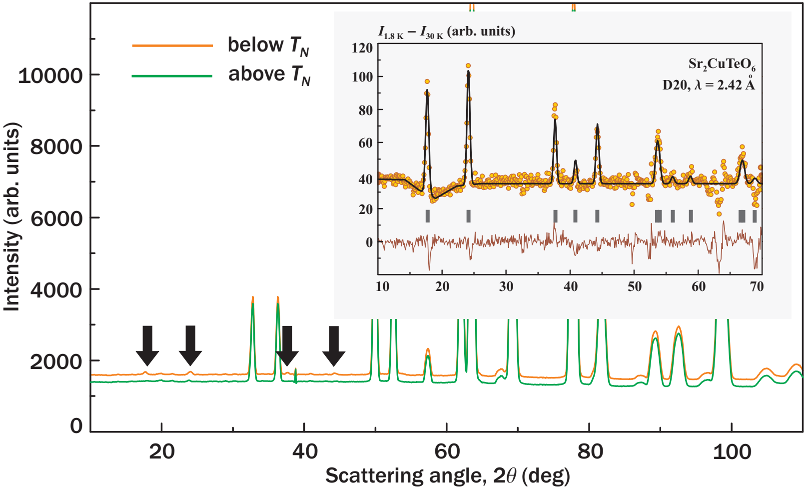

PRB 75, 094421 (2007)
[Cu(Pz)$_2$(HF$_2$)]BF$_4$
Cu(Pz)$_2$(ClO$_4$)$_2$
Specific heat (J/mol K)
10
1
0.1
1
10
T (K)





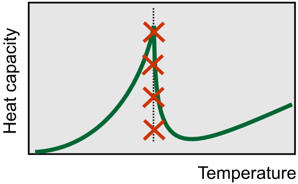
Specific heat anomalies may be weak...


below T_N
above T_N
D20, ILL
Sr_2CuTeO_6
Intensity (arb. units)
10000
8000
6000
4000
2000
0
20
40
60
80
100
Scattering angle, 2θ (deg)

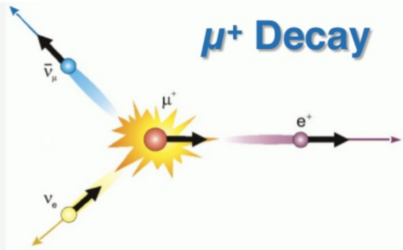


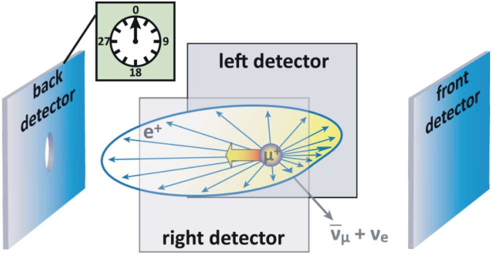
Magnetic Bragg peaks can be **very weak**, because $I \sim \mu_m^2$

Stage 1
absence of magnetic order



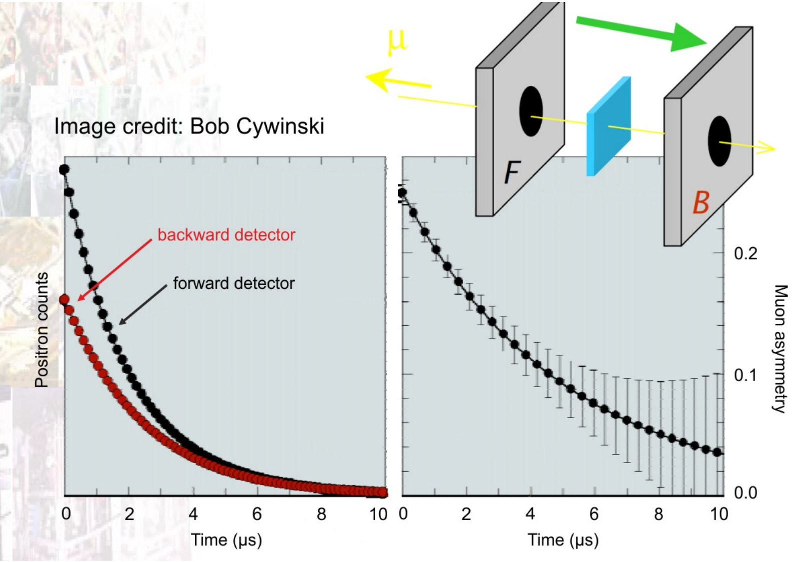
Stage 2
persistent spin dynamics



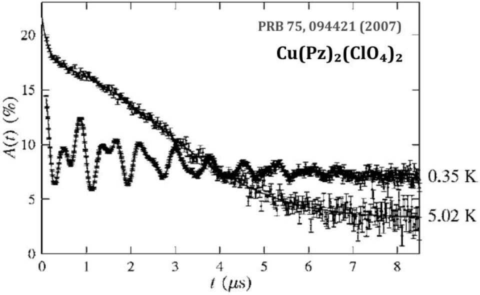


- What happens: positron follows the direction of muon spin
- Measure: asymmetry of the positron emission
- Extract: local magnetization

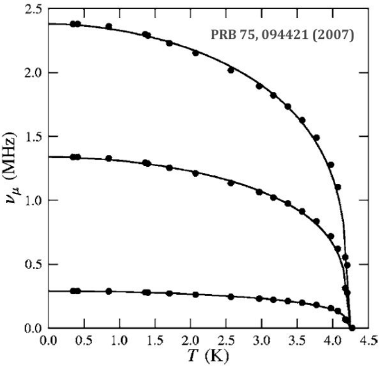
Image credit: J.H. Brewer, slides at Int. Conference on Hyperfine interactions (2014)



Further reading: A. Hillier *et al. Nature Rev. Methods Primer* **2**, 4 (2022)



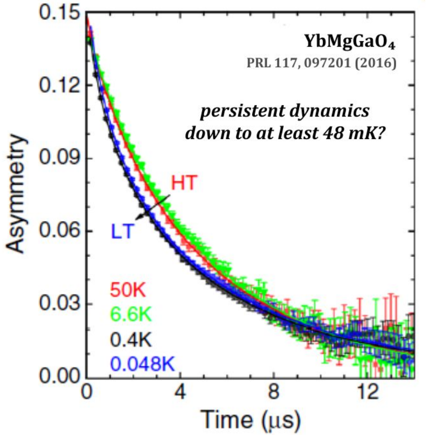
Oscillations indicate static local fields



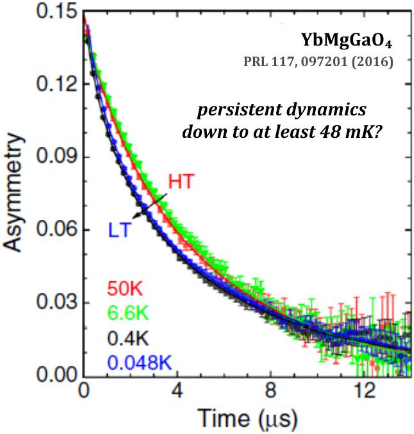
Muons are able to say whether:

- your sample develops long-range order (discrete static fields)
- shows some other kind of static magnetism (spin glass)

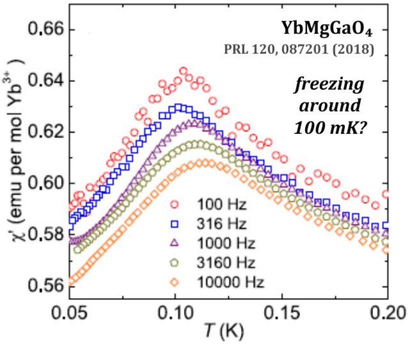
μSR data


YbMgGaO₄
PRL 117, 097201 (2016)
persistent dynamics
down to at least 48 mK?
HT
LT
50K
6.6K
0.4K
0.048K
Asymmetry
Time (μs)
0.15
0.12
0.09
0.06
0.03
0.00
0
4
8
12

μSR data



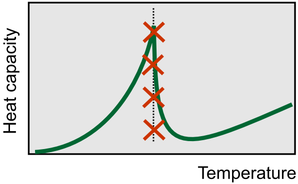
ac-susceptibility



Muons may see magnetism differently from other methods

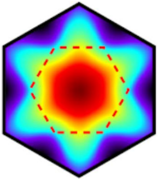
Three stages of experimental research

Stage 1
absence of magnetic order



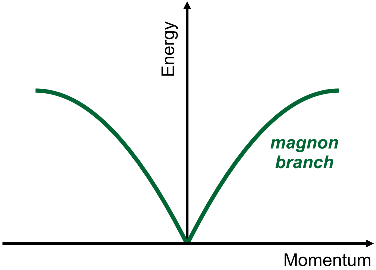
Stage 2
persistent spin dynamics

Stage 3
evidence of unconventional excitations



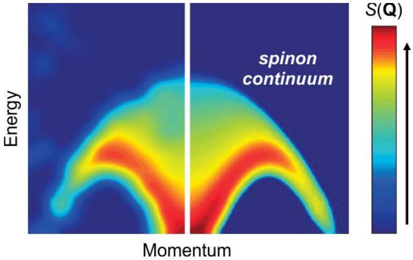
Conventional (ordered) magnet

$S = 1$ excitations, *magnons*



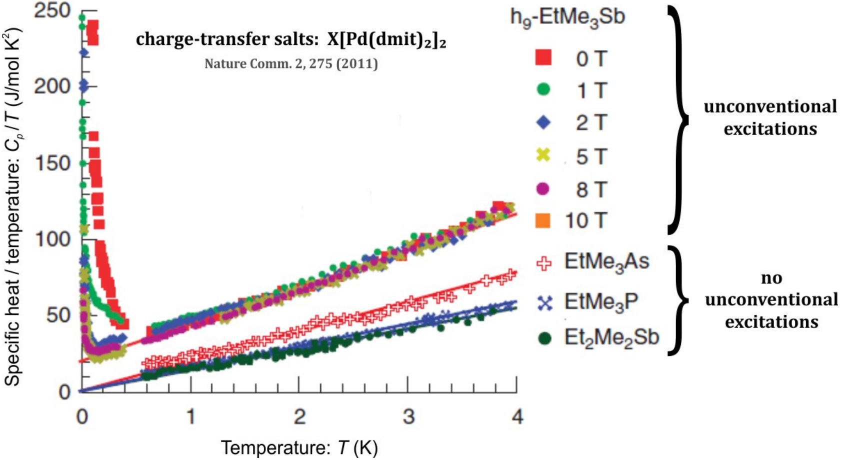
Unconventional (quantum) magnet

$S = \frac{1}{2}$ excitations, *spinons*

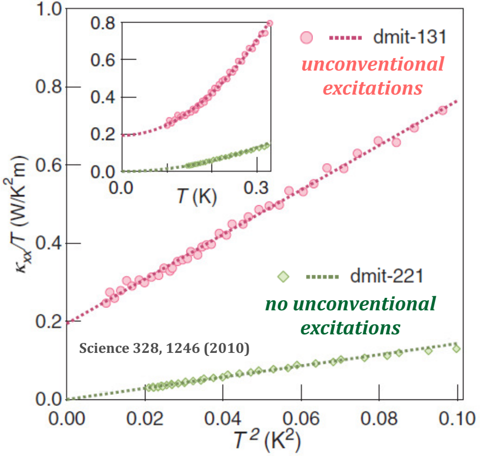


Fractionalized excitations manifest themselves by broad spectral features

Image from M. Mourigal *et al.* Nature Phys. 9, 435 (2013)

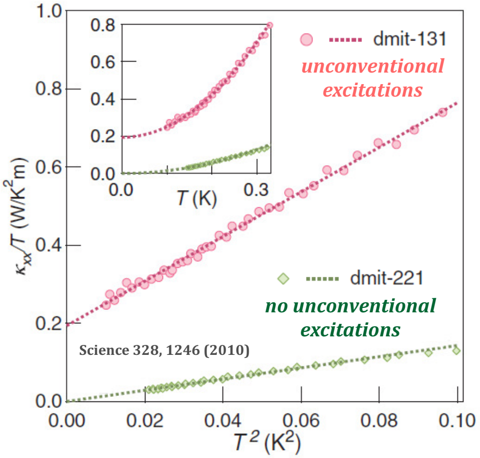


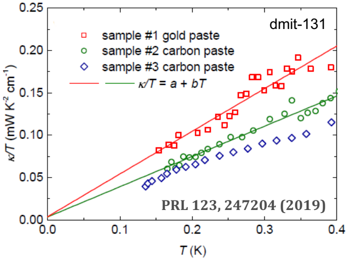
In insulators, **linear term** in the specific heat ($C_p \sim T$) may be indicative of unconventional excitations

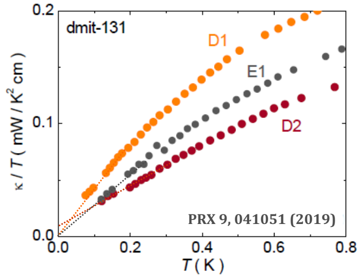


1.0
0.8
0.6
0.4
0.2
0.0
0.00
0.02
0.04
0.06
0.08
0.10
T^2 (K^2)
κ_{xx}/T (W/K^{2}m)
0.8
0.6
0.4
0.2
0.0
0.0
0.3
T (K)
dmit-131
unconventional excitations
dmit-221
no unconventional excitations
Science 328, 1246 (2010)

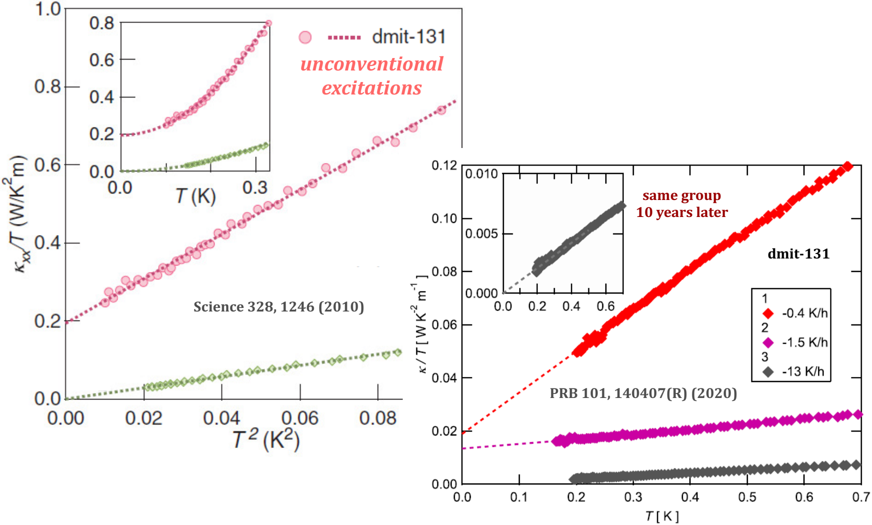
Unconventional excitations: thermal conductivity





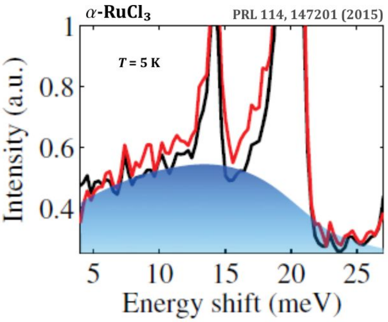


Unconventional excitations: thermal conductivity

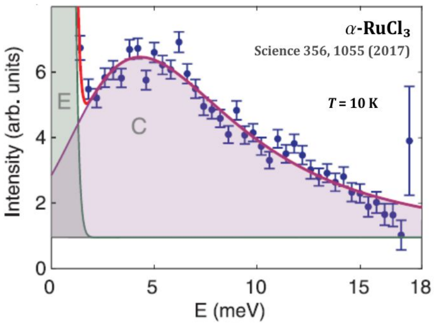


Thermal conductivity data may be ambiguous...

Raman scattering ($\mathbf{q} = 0$)



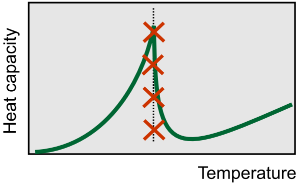
Inelastic neutron scattering (all $\mathbf{q}$'s)



Excitation **continuum** manifests

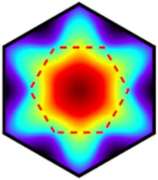
unconventional nature of the system

Stage 1
absence of magnetic order



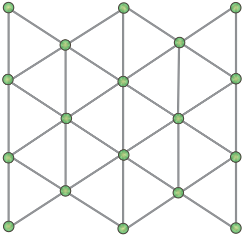
Stage 2
persistent spin dynamics

Stage 3
evidence of unconventional excitations



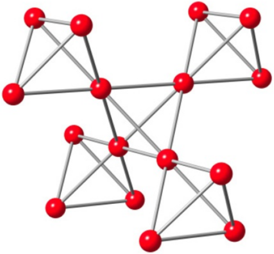
Where to search for the spin liquid?

(part 2)

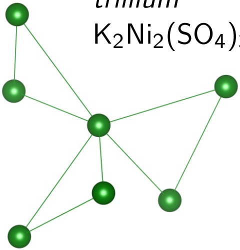


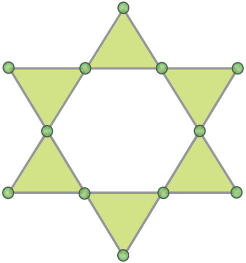
triangular / $NaYbO_2$

pyrochlore
$NaCaCo_2F_7$

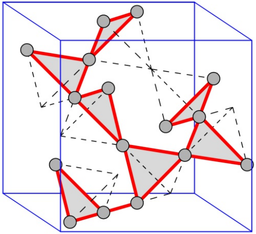


trillium
$K_2Ni_2(SO_4)_3$





kagome
herbertsmithite



hyperkagome / $PbCuTe_2O_6$

Cu-based kagome minerals:

- herbertsmithite
- tondiite
- kapellasite
- volborthite
- barlowite (Zn-doped)
- claringbullite (Zn-doped)

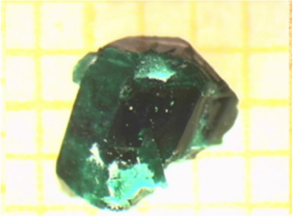
and perhaps more
hidden under the ground...

Cu-based kagome minerals:

- herbertsmithite
- tondiite
- kapellasite
- volborthite
- barlowite (Zn-doped)
- claringbullite (Zn-doped)

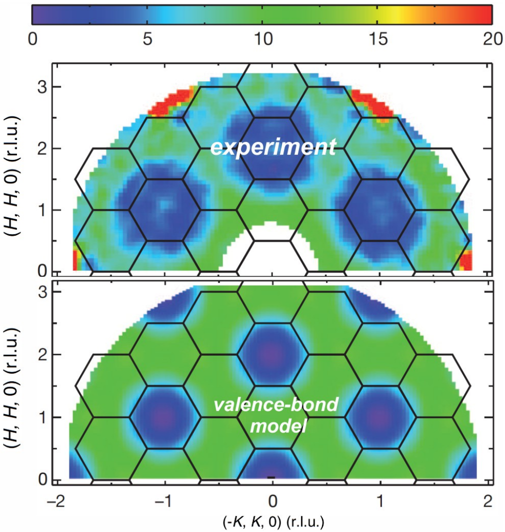
and perhaps more
hidden under the ground...

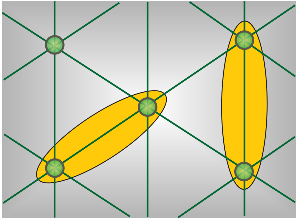
natural sample



synthetic sample

Kagome (herbertsmithite)





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

"valence bond"

Most of the spectrum:

excitations of nearest-neighbor singlets

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

Cu-based kagome minerals:

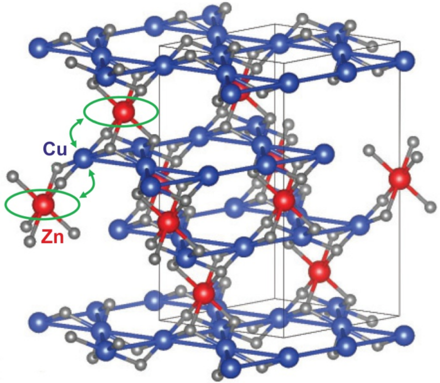
- herbertsmithite
- tondiite
- kapellasite
- volborthite
- barlowite (Zn-doped)

...all look like spin liquids

Synthetic compounds:

- $YCu_3(OH)_6Cl_3$
- $CaCu_3(OH)_6Cl_2$
- $KCu_3As_2O_7(OH)_3$

...are all magnetically ordered



Cu/Zn site mixing
(*randomness*)
is it relevant?

Image from T.-H. Han *et al.* Phys. Rev. B 94, 060409 (2016)

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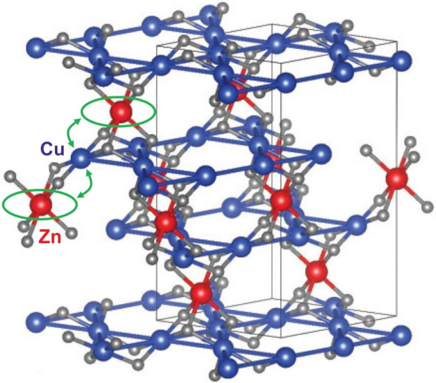
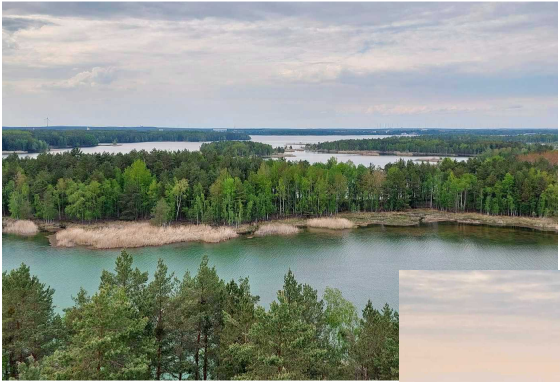


Cu/Zn site mixing
(*randomness*)
is it crucial?

Image from T.-H. Han *et al.* Phys. Rev. B 94, 060409 (2016)

Lausitzer Seenland
near Senftenberg

How do we know it is liquid?



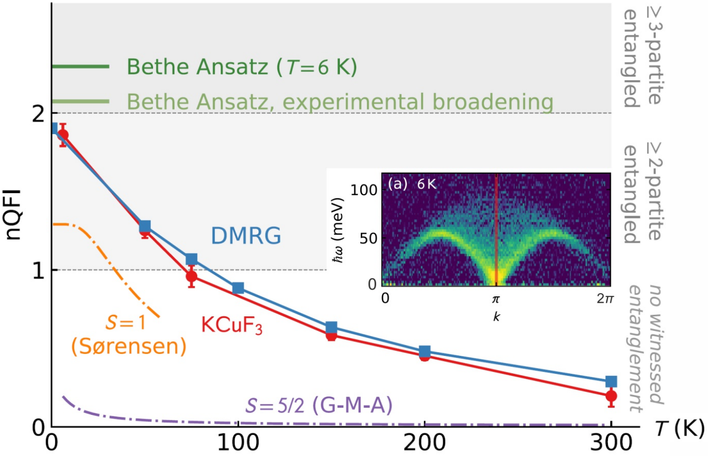
Lausitzer Seenland
near Senftenberg

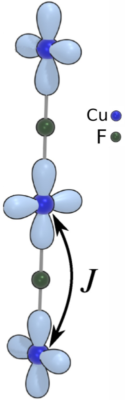
lake or solar panels?

How to detect the spin liquid?

(part 2)

A. Scheie *et al.*
PRB 103, 224434 (2021)

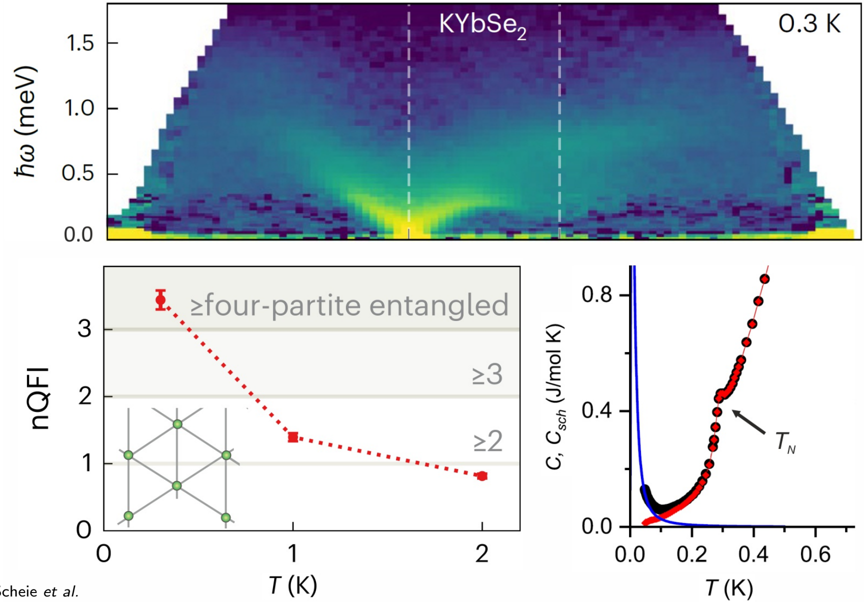




Quantum Fisher information

$$\mathrm{nQFI}(T) = \frac{\hbar}{3\pi S^2}\int_0^\infty d(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_B T}\right)\chi''(\hbar\omega, T)$$

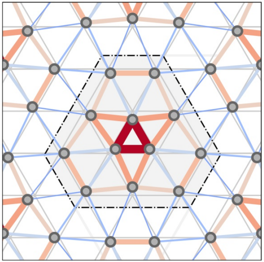
Witness of entanglement: proximate spin liquid



A. Scheie *et al.*
Nature Phys. 20, 74 (2024)

1. Genuine vs. randomness "spin liquid": **distinguishable** by quantum Fisher information?

2. Is quantum spin liquid **stable** against weak perturbations?



U.F.P. Seifert *et al.* Nature Comm. 15, 7110 (2024)

Further reading

General / introductory:

- T. Imai and Y. Lee, *Physics Today* **69**, 30 (2016)
- A.A. Tsirlin and P. Gegenwart, *Physik in unserer Zeit* **50**, 71 (2019)
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- J. Knolle and R. Moessner, *Ann. Rev. Condens. Matter Phys.* **10**, 451 (2019)
- X.-G. Wen, *Science* **363**, eaal3099 (2019)
- C. Broholm *et al. Science* **367**, eaay0668 (2020)

Technical (and more theoretical):

- L. Savary and L. Balents, *Rep. Prog. Phys.* **80**, 016502 (2017)
- Y. Zhou *et al. Rev. Mod. Phys.* **89**, 025003 (2017)

Models and materials:

- S. Winter *et al. J. Phys.: Condens. Matter* **29**, 493002 (2017)
- J.G. Rau and M.J.P. Gingras, *Ann. Rev. Condens. Matter Phys.* **10**, 357 (2019)
- J.R. Chamorro, T.M. McQueen, T.T. Tran, *Chem. Rev.* **121**, 2898 (2021)