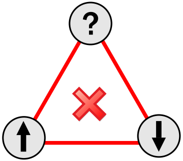
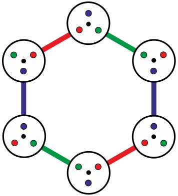
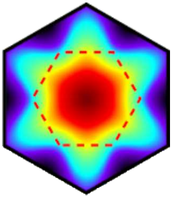
Frustrated magnetism

Alexander Tsirlin

Experimental Physics VI, Center for Electronic Correlations and Magnetism
University of Augsburg







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TRR80 Summer School
June 25, 2019

Frustrated magnetism

or everything you wanted to know about spin liquids but were afraid to ask

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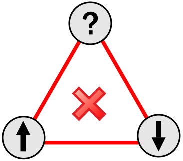

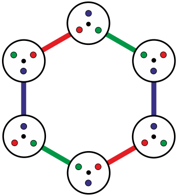

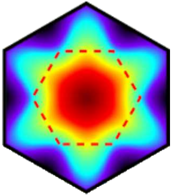

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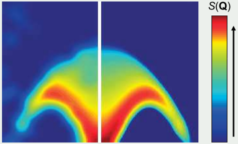
What is special?

Conventional and unconventional magnets

Spin liquids

Quantum effects and entanglement



How to identify?

Material classes

From proof-by-contradiction to proof-by-evidence

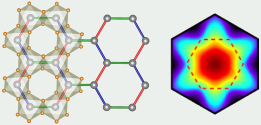
Continuous excitations and their detection

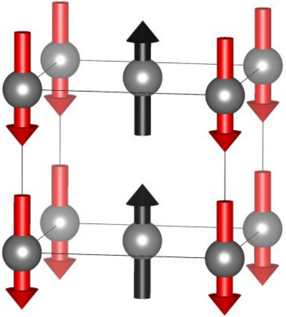
What to look for?

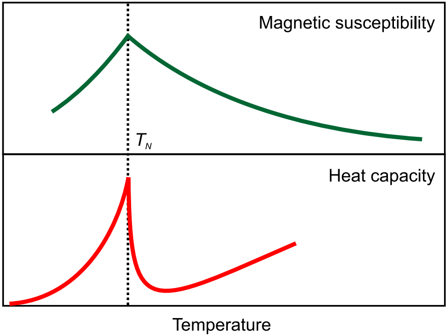
Quantum and classical spin liquids

Magnetic monopoles

Anyonic excitations

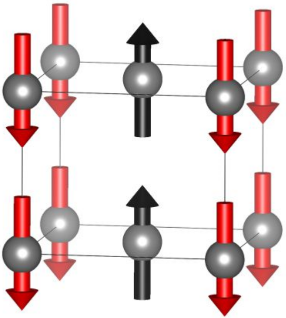


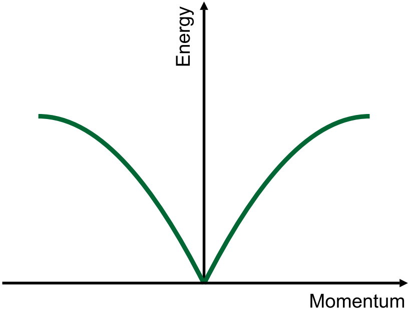




- Develop **magnetic order** below the characteristic Néel temperature T_N
- **Ordered state** can be represented by up- and down-spins (spin vectors)
- Spin-wave excitations (precessing spins)

Conventional magnets





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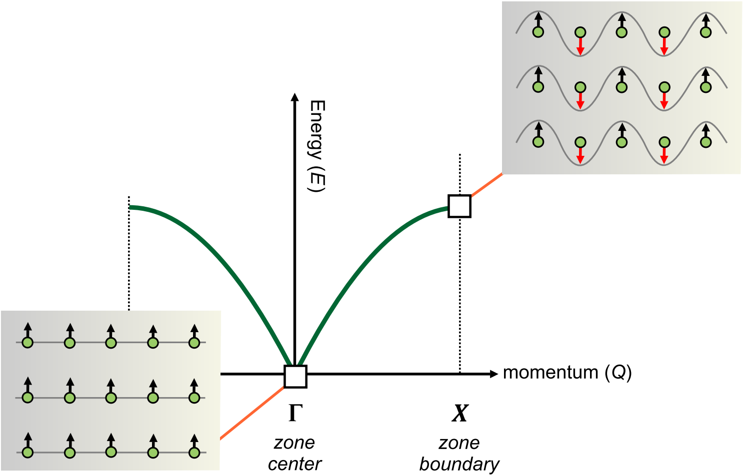
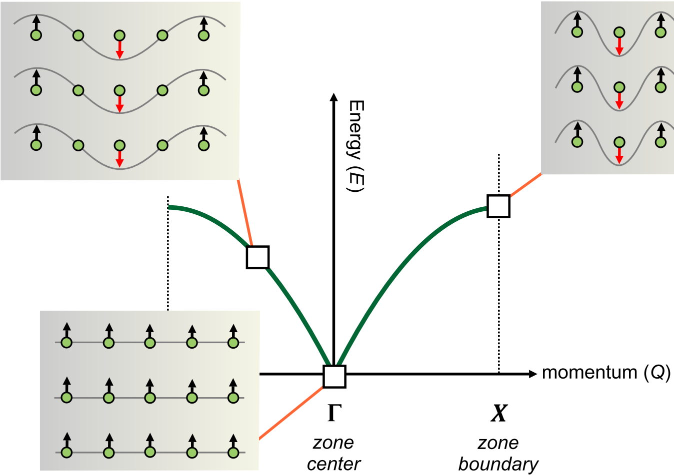


- Periodic atomic displacements form lattice waves (phonons)
- Each phonon is described by a dispersion relation between energy and momentum



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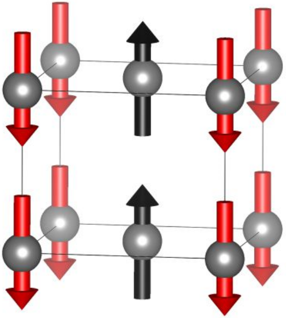
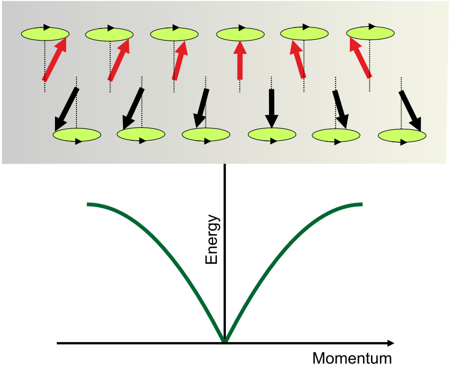
Conventional magnetic excitations





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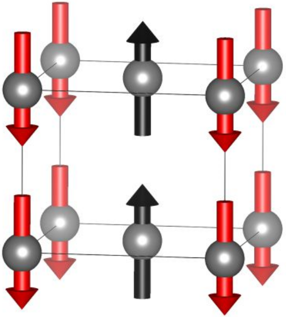

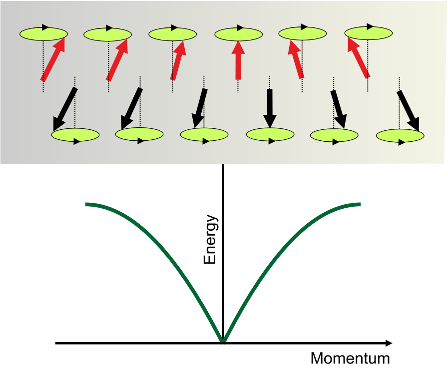

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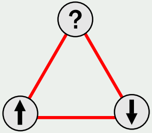
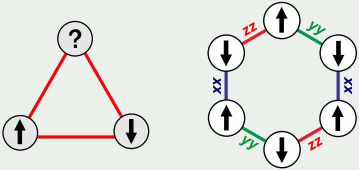
▶ **Frustrated magnets offer something less conventional**



geometrical frustration
competition on triangular loops



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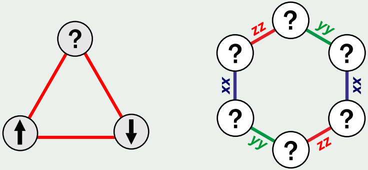

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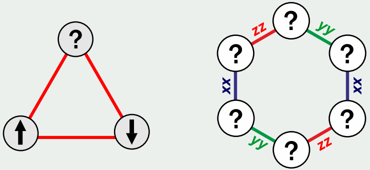
exchange frustration
incompatible easy-axis directions



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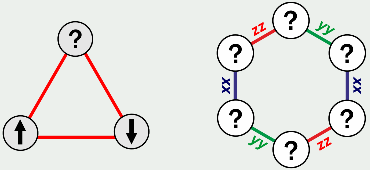
Spin liquid

No magnetic long-range order
Strong short-range correlations



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Spin liquid

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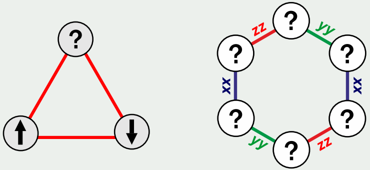
Ordinary liquids

freeze
upon cooling

$T < 0\,^{\circ}\mathrm{C}$

Frustration and spin liquids



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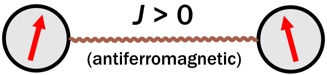
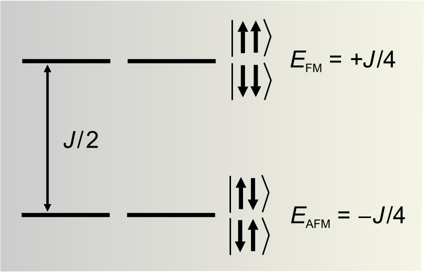
Quantum spin liquids are different and do not freeze

Spin dimer
(classical)
$\hat{H} = J\,\mathbf{S}_1\,\mathbf{S}_2$





Classical case

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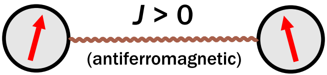

Spin dimer
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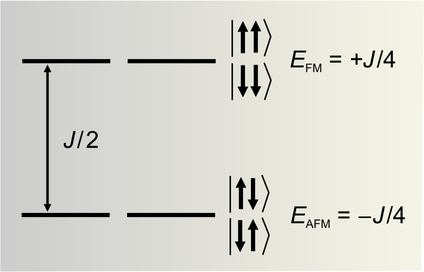

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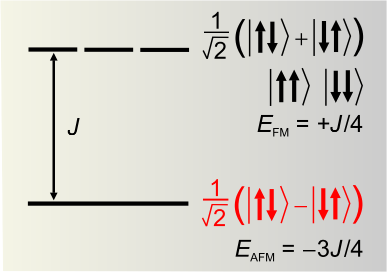

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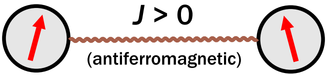

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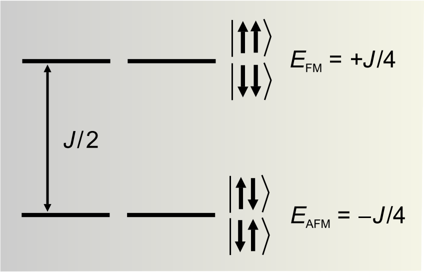

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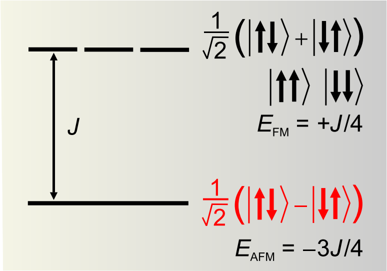
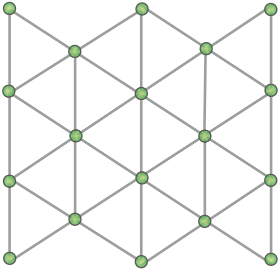
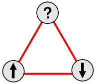


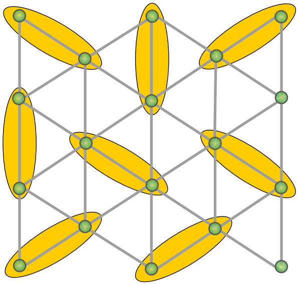
Quantum case

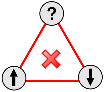
In quantum magnets, spins become entangled





- What happens in Heisenberg antiferromagnets with the triangular geometry?
- P.W. Anderson (1973): they form a **resonating-valence-bond (RVB) state**, a combination of all possible arrangements of spin singlets on the triangular lattice





Spin singlet (*valence bond*):


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

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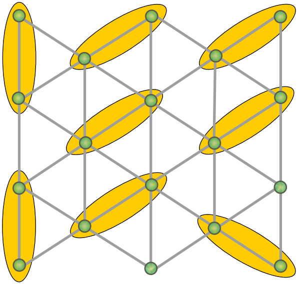

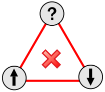

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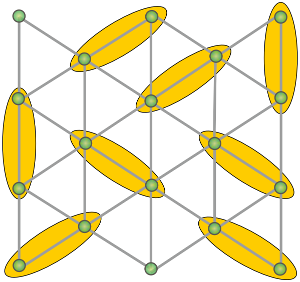
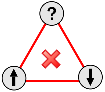
Novel states and their excitations





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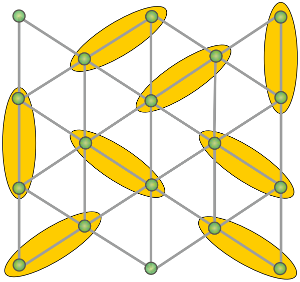

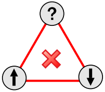

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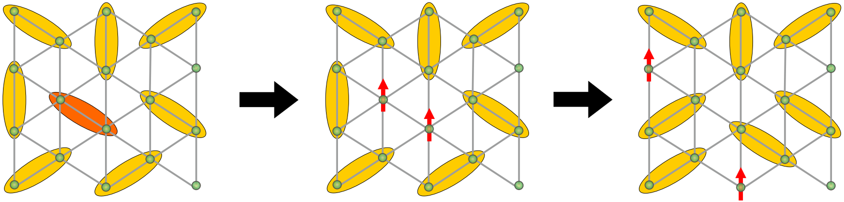
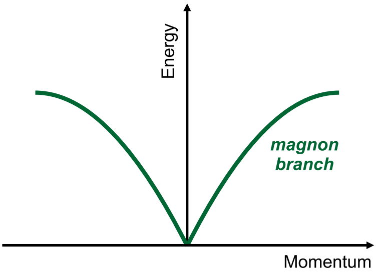


Two spins propagate independently, the $S = 1$ excitation breaks into two

Signatures of fractionalized excitations

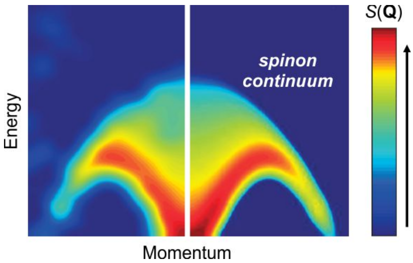
Conventional (ordered) magnet

$S = 1$ excitations, *magnons*



Unconventional (quantum) magnet

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



- Fractionalized excitations manifest themselves by broad spectral features and can be detected experimentally
- **This fractionalization is a fingerprint of quantum entanglement**

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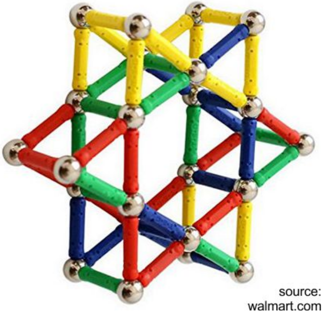
Realization: artificial systems



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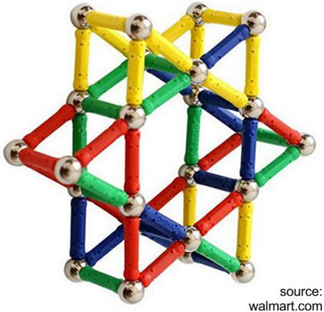
Realization: artificial systems



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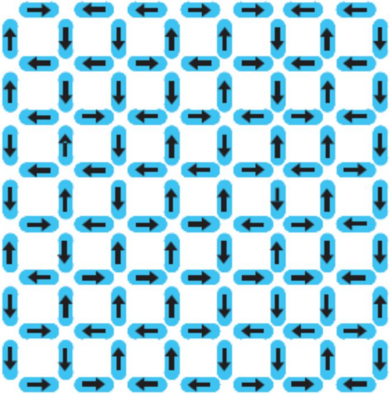
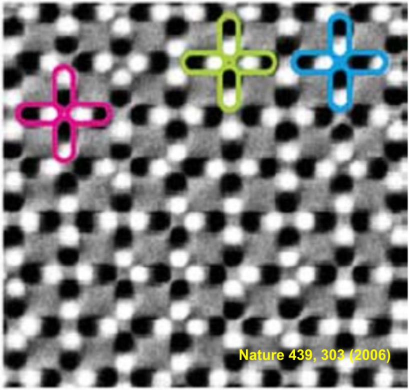
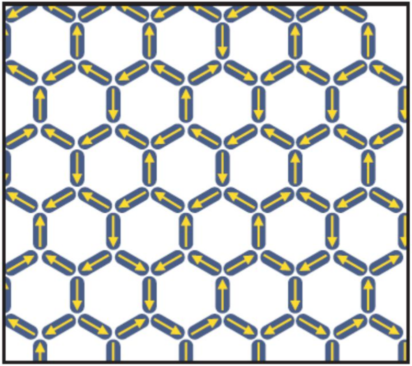
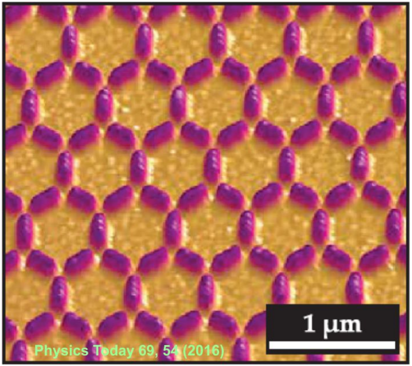



Nature 439, 303 (2006)

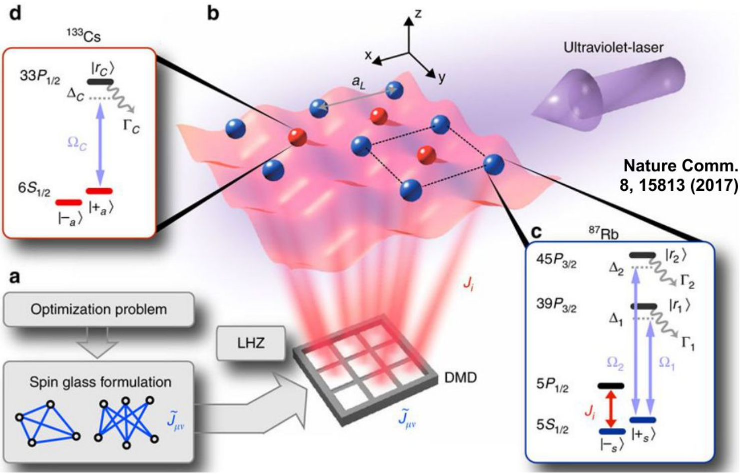




Large magnetic moments → no quantum effects

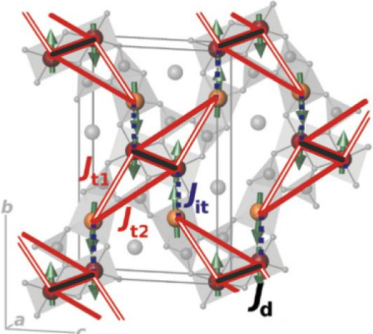
Further reading:

- I. Gilbert, C. Nisoli, P. Schiffer, *Physics Today* **69**, 54 (2016)
- C. Nisoli, R. Moessner, P. Schiffer, *Rev. Mod. Phys.* **85**, 1473 (2013)

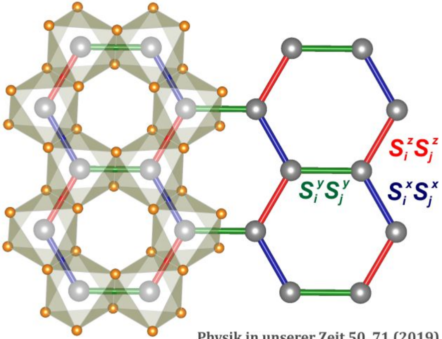


Plenty of interesting physics, but no spin liquids so far

Realization: synthetic and natural materials



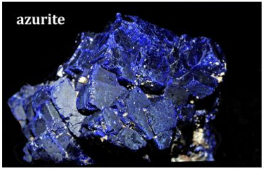
PRB 85, 064404 (2012)

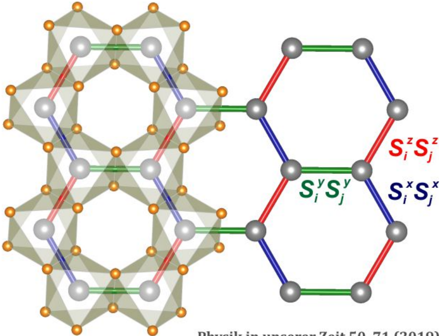


Physik in unserer Zeit 50, 71 (2019)

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

Realization: synthetic and natural materials





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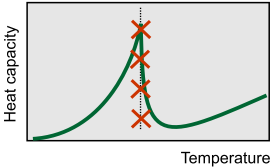
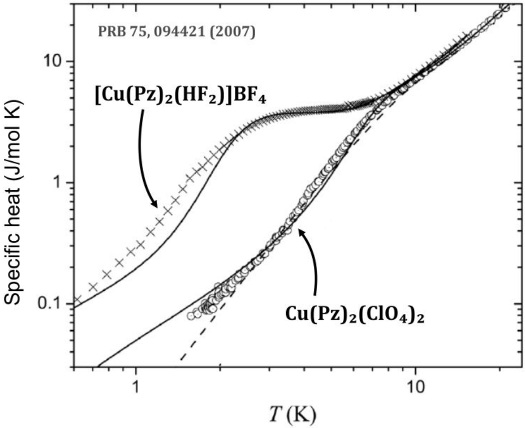
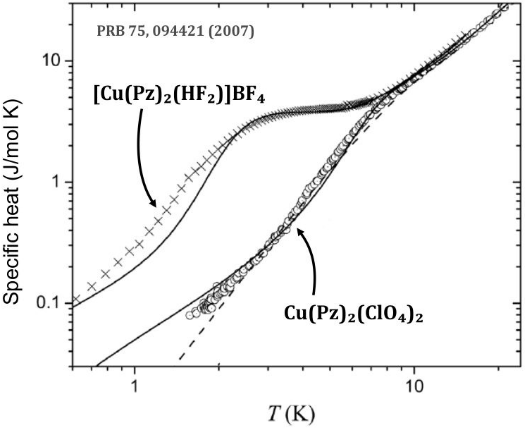
Stage 1

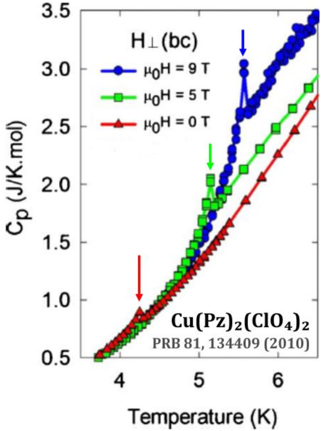
Absence of a magnetic transition indicates the spin liquid?

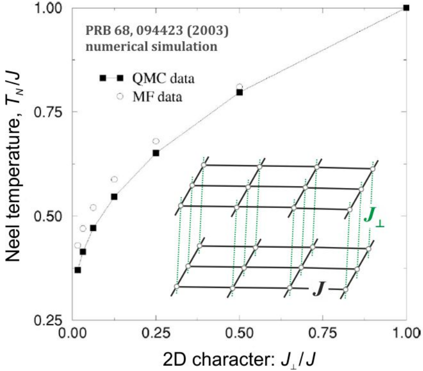


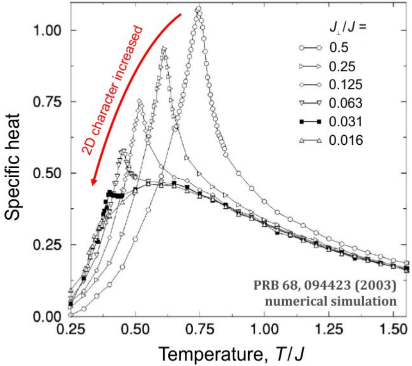
Absence of a magnetic transition



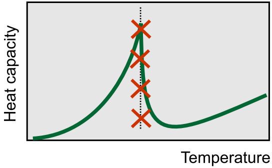

PRB 75, 094421 (2007)
Specific heat (J/mol K)
$[Cu(Pz)_2(HF_2)]BF_4$
$Cu(Pz)_2(ClO_4)_2$
T (K)


H⊥(bc)
μ_0H = 9 T
μ_0H = 5 T
μ_0H = 0 T
C_p (J/K.mol)
$Cu(Pz)_2(ClO_4)_2$
PRB 81, 134409 (2010)
Temperature (K)





In quantum magnets, thermodynamic signatures of magnetic ordering may be very weak and inconspicuous



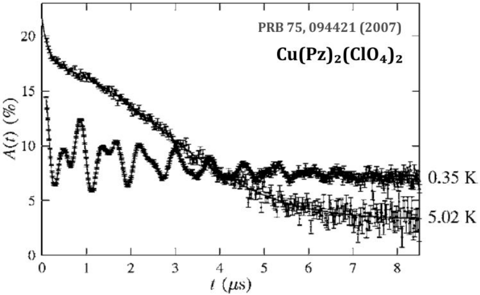
Stage 1

Absence of a magnetic transition indicates the spin liquid?

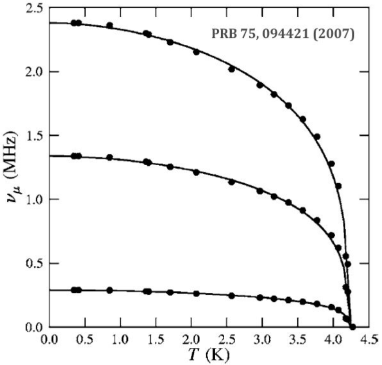
- **It may indicate that you did not look close enough**

Stage 2

Absence of local fields and presence of spin dynamics prove the spin-liquid formation?



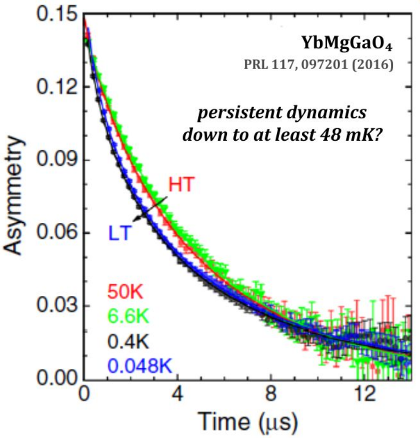
Oscillations indicate static local fields



Muons are able to say whether:

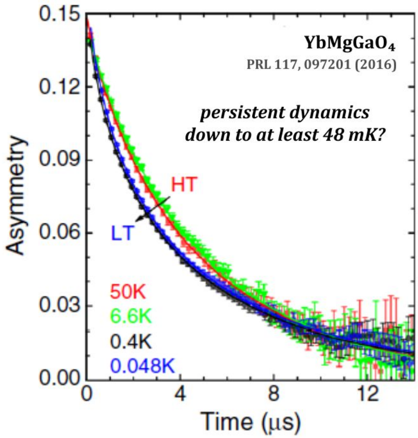
- your sample develops long-range order (discrete static fields)
- shows some other kind of static magnetism (spin glass)

μSR data

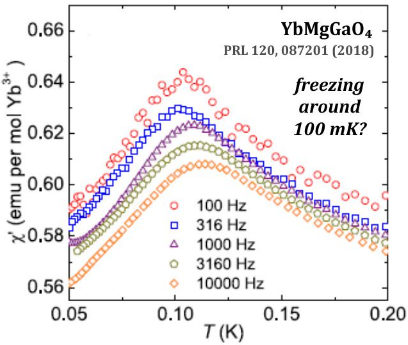

YbMgGaO4
PRL 117, 097201 (2016)
persistent dynamics
down to at least 48 mK?
HT
LT
50K
6.6K
0.4K
0.048K
Asymmetry
0.15
0.12
0.09
0.06
0.03
0.00
0
4
8
12
Time (μs)

Ambiguity of the muon data

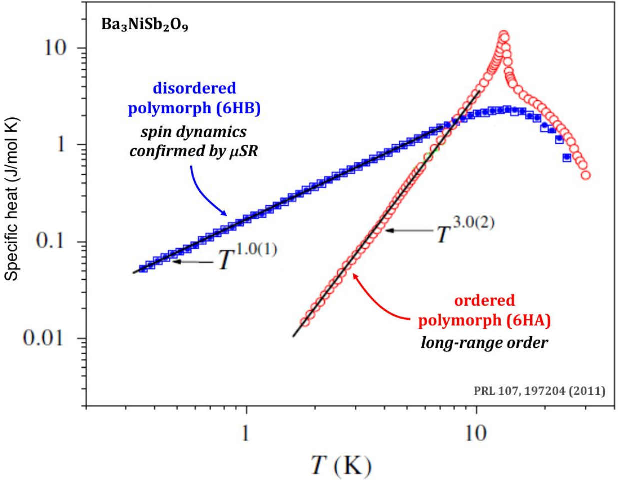
μSR data

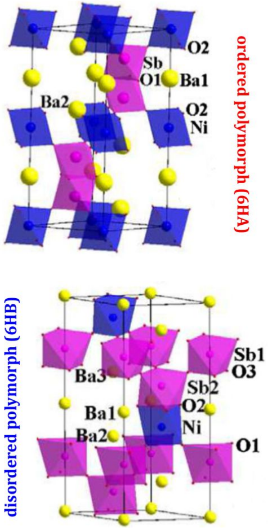


ac-susceptibility



Muons may see magnetism differently from other methods



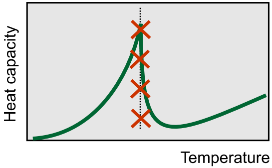


Spin dynamics is often accompanied by the structural disorder (and triggered by it?)

Stage 1

Absence of a magnetic transition indicates the spin liquid?

- **It may indicate that you did not look close enough**



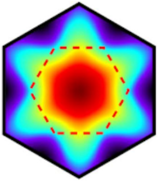
Stage 2

Absence of local fields and presence of spin dynamics prove the spin-liquid formation?

- **They prove disordered magnetism of some sort**

Stage 3

Unconventional excitations evidence the spin liquid



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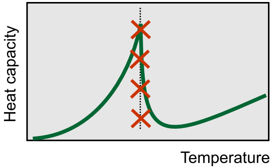


Stage 2

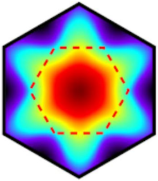
Absence of local fields and presence of spin dynamics prove the spin-liquid formation?

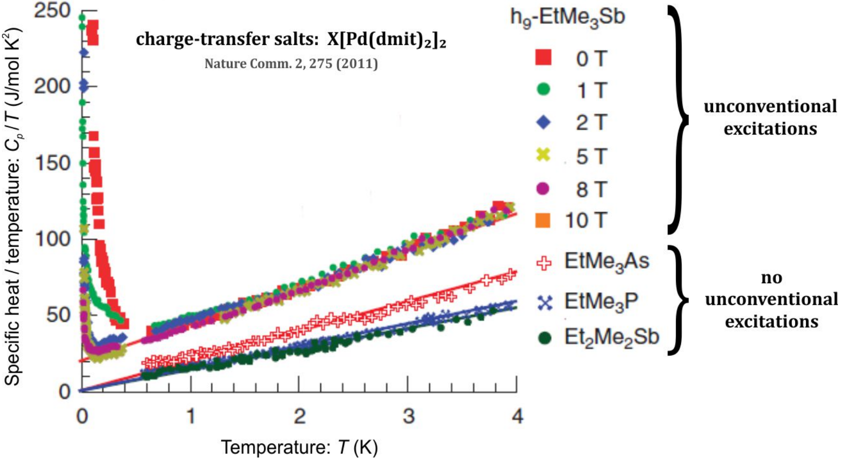
- **They prove disordered magnetism of some sort**

Stage 3

Unconventional excitations evidence the spin liquid

- **Yes, but how do we know they are unconventional?**

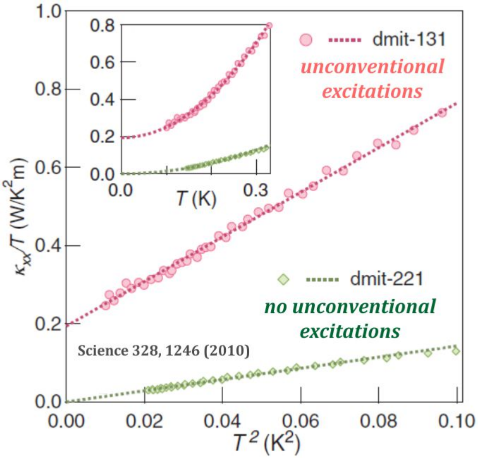




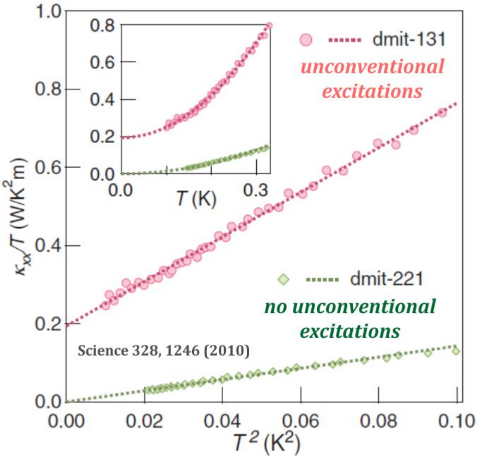
Linear term in the specific heat of an insulating material ($C_p \sim T$)
= non-zero intercept for C_p/T vs. T^2

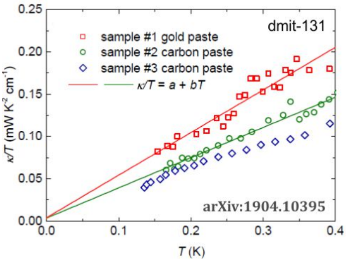
may be indicative of unconventional excitations

Unconventional excitations: thermal conductivity

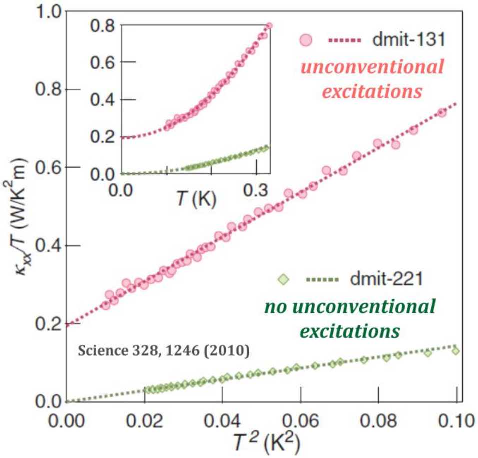


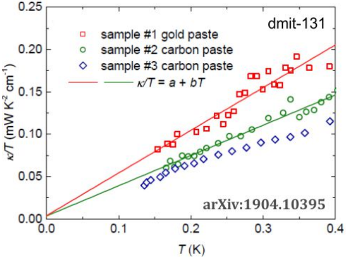
Unconventional excitations: thermal conductivity

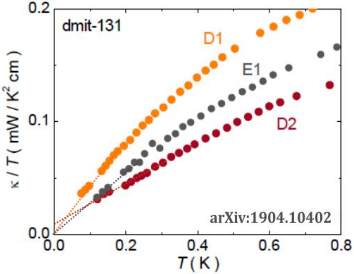




Unconventional excitations: thermal conductivity

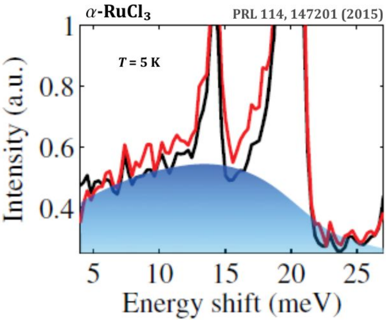




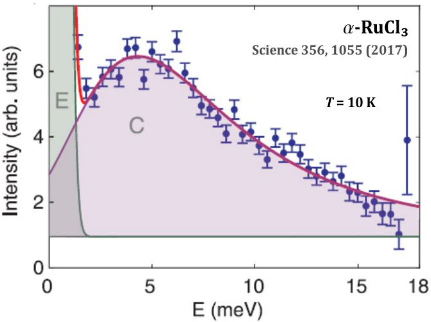


Thermal conductivity data may be ambiguous...

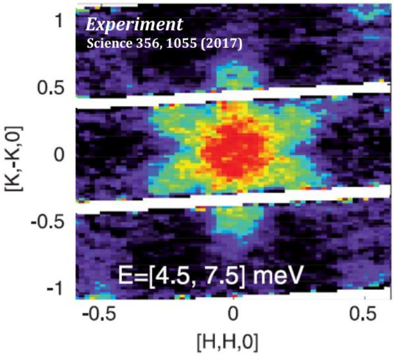
Raman scattering ($\mathbf{q} = 0$)

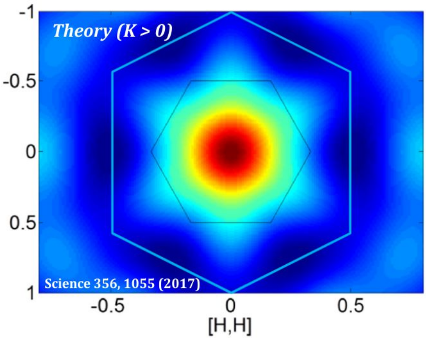


Inelastic neutron scattering (all $\mathbf{q}$'s)



Unconventional excitations are broadly distributed in energy and momentum, and manifest themselves by a broad spectral feature (*continuum*)

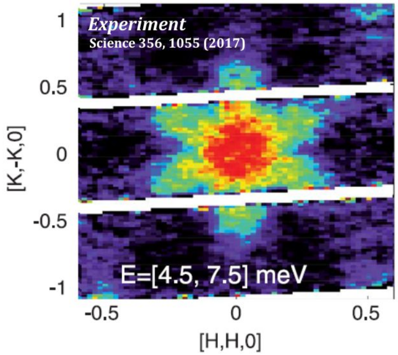


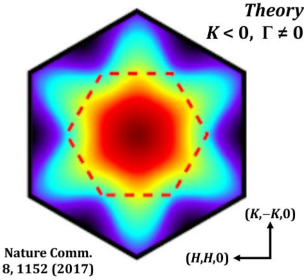


How to understand David's star?

- $K > 0$: exotic fractionalized excitations (spin-liquid scenario)
- $K < 0$ (and $\Gamma \neq 0$): conventional excitations

Unconventional excitations: spectroscopy





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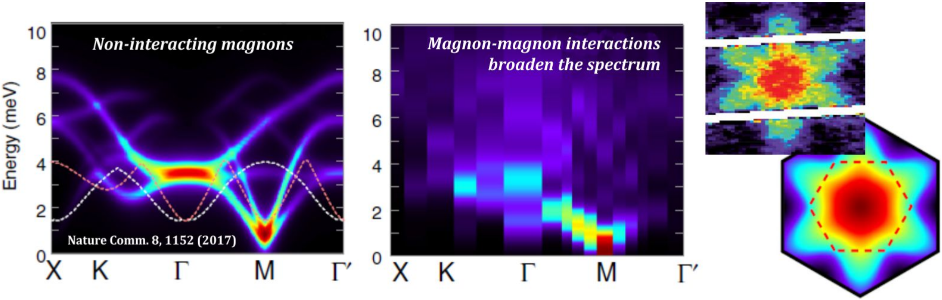
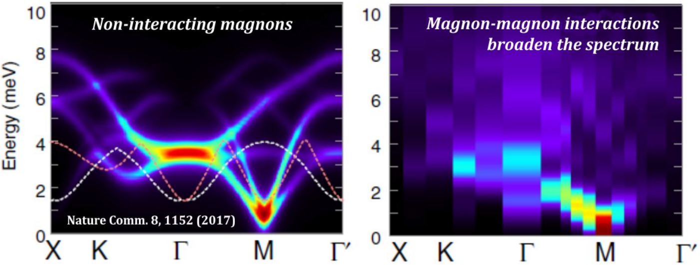
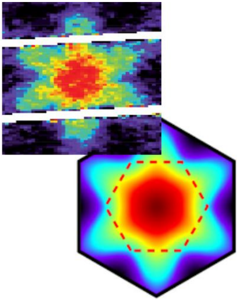


How to understand David's star?

- $K > 0$: exotic fractionalized excitations (spin-liquid scenario)
- $K < 0$ (and $\Gamma \neq 0$): conventional excitations (magnon breakdown)





How to understand David's star?

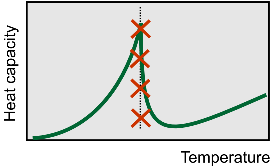
- $K > 0$: exotic fractionalized excitations (spin-liquid scenario)
- $K < 0$ (and $\Gamma \neq 0$): conventional excitations (magnon breakdown)

Even if you see a broad spectral feature, its meaning depends on the interpretation!

Stage 1

Absence of a magnetic transition indicates the spin liquid?

- **It may indicate that you did not look close enough**



Stage 2

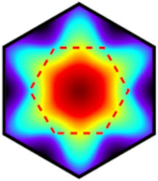
Absence of local fields and presence of spin dynamics prove the spin-liquid formation?

- **They prove disordered magnetism of some sort**

Stage 3

Unconventional excitations evidence the spin liquid

- Yes, but how do we know they are unconventional?
- **No unique experimental signature of a spin liquid exists, we have to cross-check the scenario by a variety of methods, and refer to "spin-liquid candidates" rather than "materials"**



What is special?

Conventional and unconventional magnets

Spin liquids

Quantum effects and entanglement

How to identify?

Material classes

From proof-by-contradiction to proof-by-evidence

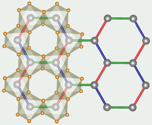
Continuous excitations and their detection

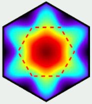
What to look for?

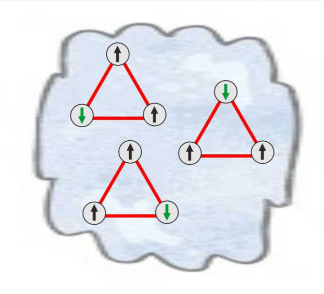
Quantum and classical spin liquids

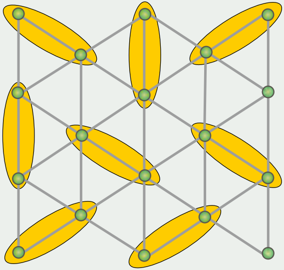
Magnetic monopoles

Anyonic excitations







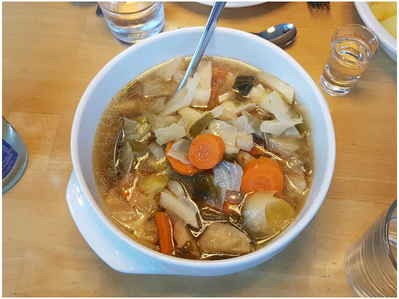


Classical spin liquid

Multiple classical states
having the same energy

Quantum spin liquid

Ground state is a superposition
of classical states



Classical soup

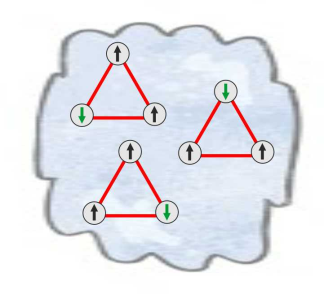
Quantum soup

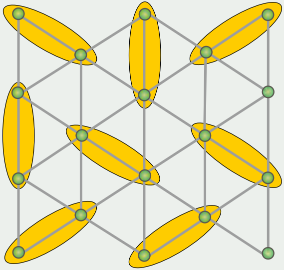
Classical spin liquid

Multiple classical states
having the same energy

Quantum spin liquid

Ground state is a superposition
of classical states





Classical spin liquid

Multiple classical states having the same energy

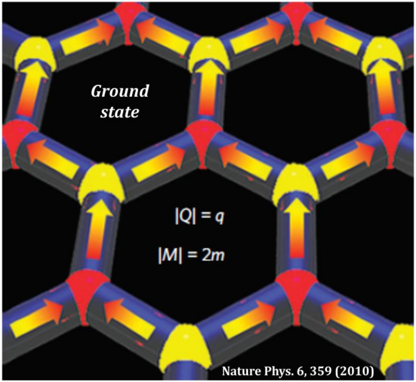
Only thermal fluctuations, **spins freeze at low T**

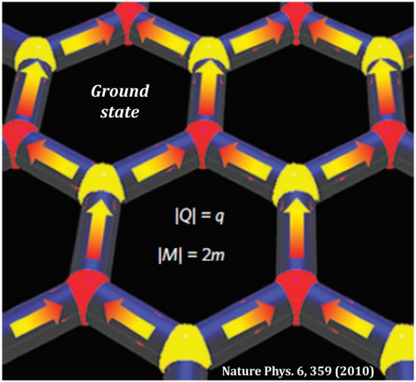
Quantum spin liquid

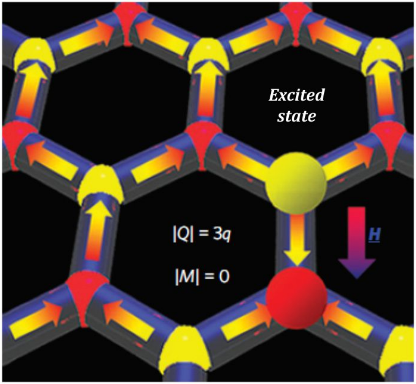
Ground state is a superposition of classical states

Quantum fluctuations **keep spins dynamic** down to 0 K

Classical spin liquids can be interesting too

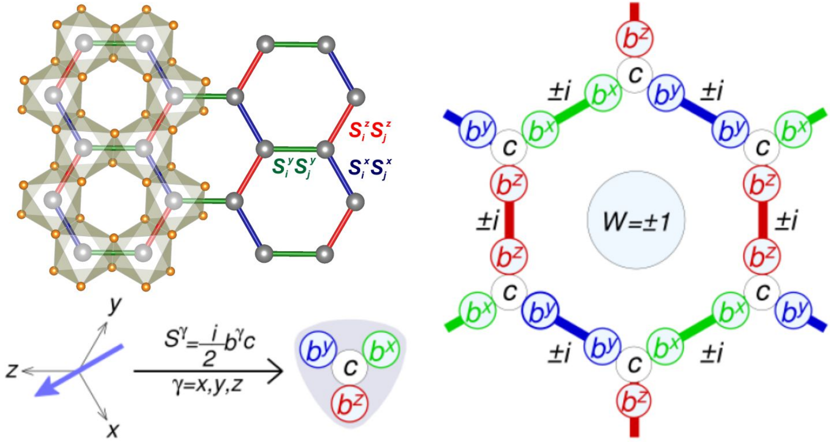

Ground state
|Q| = q
|M| = 2m
Nature Phys. 6, 359 (2010)





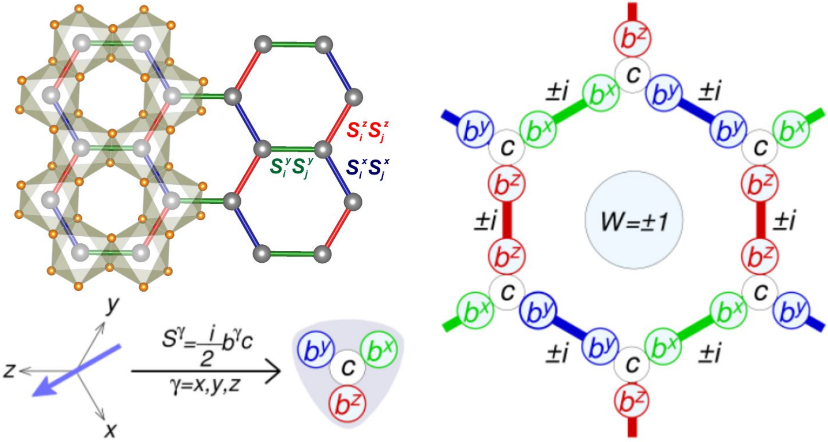
- Spin flip (excitation) generates two magnetic "charges" that can propagate independently
- Access to the physics of magnetic monopoles

▶ **What happens in the quantum case?**



- Different flavor of fractionalization: spin breaks down into Majorana fermions
- Excitations are represented by **anyons** – quasiparticles with an unusual statistics

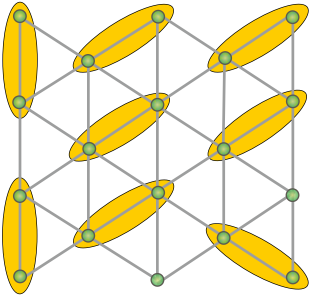
Anyonic excitations

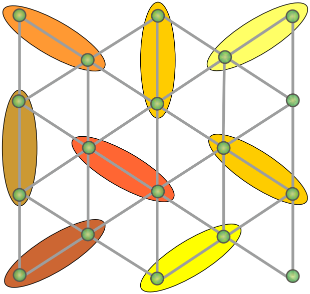


- Different flavor of fractionalization: spin breaks down into Majorana fermions
- Excitations are represented by **anyons** – quasiparticles with an unusual statistics

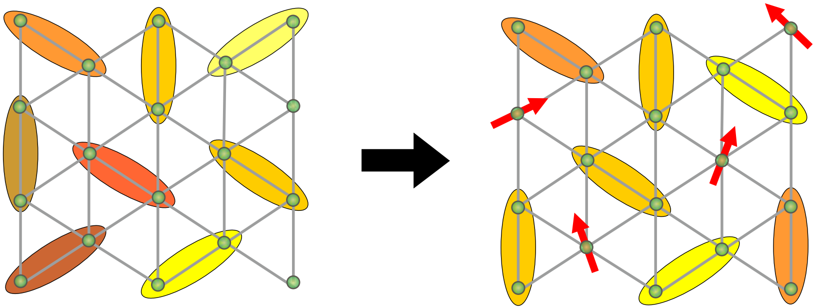
► **How to get there experimentally?**

New states cast by disorder



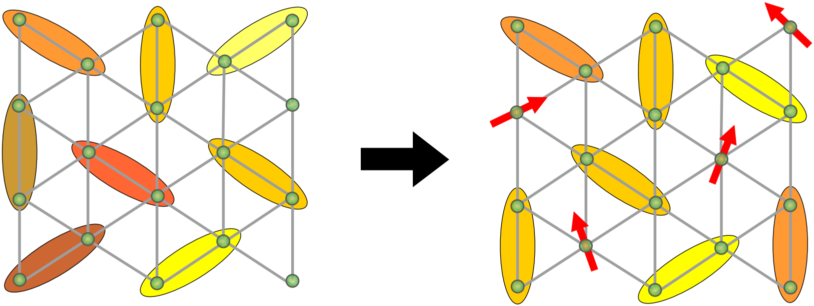


New states cast by disorder



- Structural disorder squeezes unpaired spins out of the valence-bond state
- A new way of getting spin-$\frac{1}{2}$ degrees of freedom

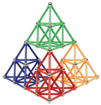
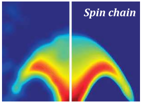
New states cast by disorder

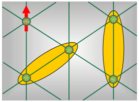
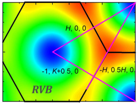


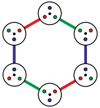
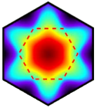
- Structural disorder squeezes unpaired spins out of the valence-bond state
- A new way of getting spin-$\frac{1}{2}$ degrees of freedom

► **How do these unpaired spins interact, and are their excitations exotic?**

Further reading

General / introductory:

- L. Balents, *Nature* **464**, 199 (2010)
- T. Imai and Y. Lee, *Physics Today* **69**, 30 (2016)
- A.A. Tsirlin and P. Gegenwart, *Physik in unserer Zeit* **50**, 71 (2019)
  English version available on request
- J. Knolle and R. Moessner, *Ann. Rev. Condens. Matter Phys.* **10**, 451 (2019)

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)
- M. Hermanns *et al.* *Ann. Rev. Condens. Matter Phys.* **9**, 17 (2018)
- J.G. Rau and M.J.P. Gingras, *Ann. Rev. Condens. Matter Phys.* **10**, 357 (2019)