Title: Spin Liquids in Frustrated Magnets

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Abstract: Frustrated magnets are materials in which localized magnetic moments, or spins, interact through competing exchange interactions that cannot be simultaneously satisfied, giving rise to a large degeneracy of the system ground state. Under certain conditions, this can lead to the formation of fluid-like states of matter, so-called spin liquids, in which the constituent spins are highly correlated but still fluctuate strongly down to a temperature of absolute zero. The fluctuations of the spins in a spin liquid can be classical or quantum and show remarkable collective phenomena such as emergent gauge fields and fractional particle excitations. This exotic behaviour is now being uncovered in the laboratory, providing insight into the properties of spin liquids and challenges to the theoretical description of these materials.

Spin Liquids in Frustrated Magnets

Leon Balents, KITP

PI Colloquium, 4/2010

Outline

- Liquids and spin liquids
- Spin ice: a classical spin liquid
- Quantum spin liquids
- How we look for them and what we've found so far

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- **Liquids and spin liquids**
- Spin ice: a classical spin liquid
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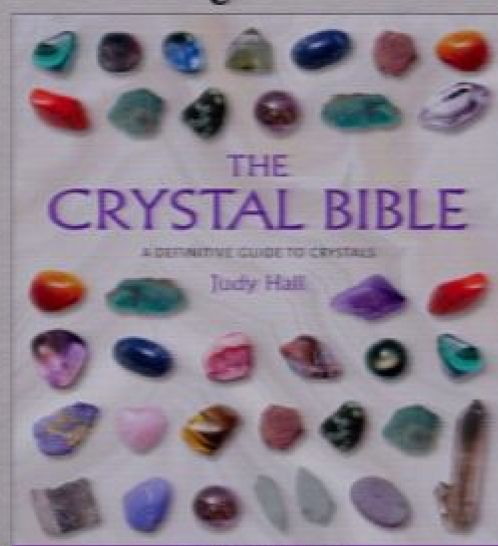
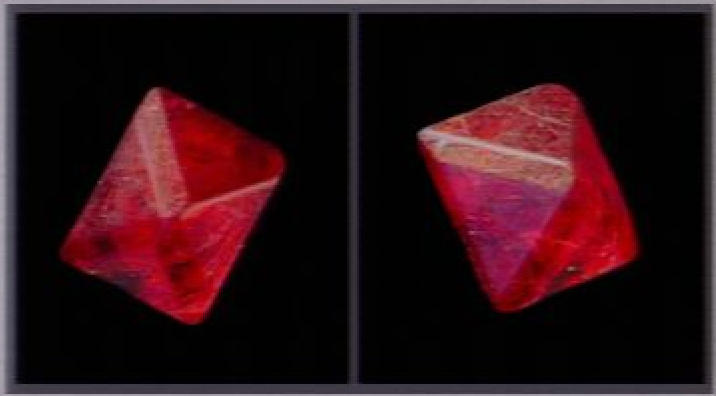
Why liquids?

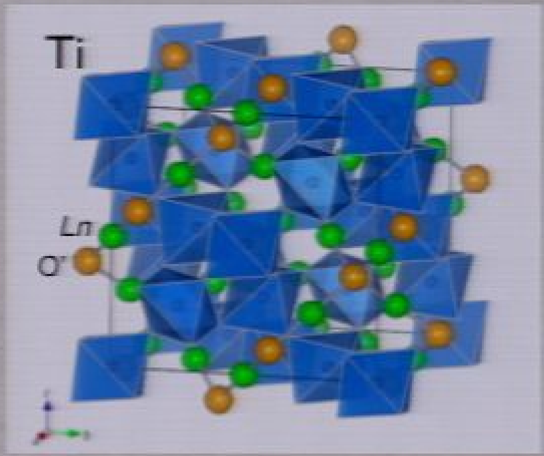
- Aren't solids infinitely more interesting?



Solids

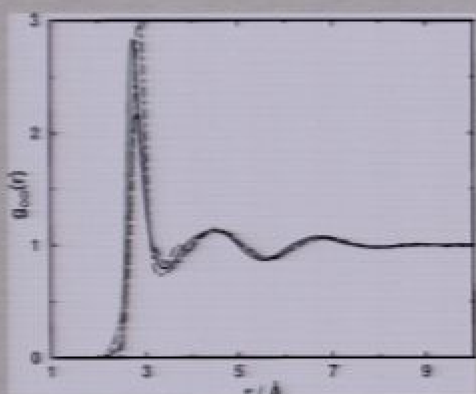


- Detailed structure description is possible - nature is very rich!
- Can classify state by *symmetry* and *order parameters* - e.g. 230 space groups

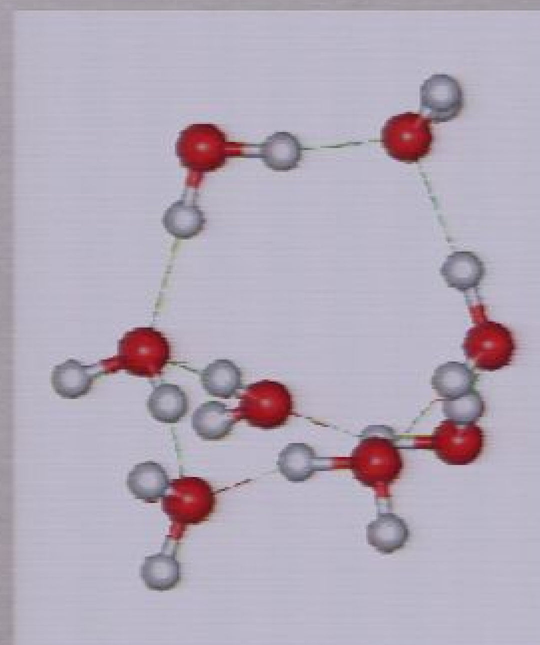


LIQUIDS ARE MORE SUBTLE

- Liquids, e.g. water (*especially water)* contain strong dynamical correlations at short distances, which are not captured by any broken symmetry or static picture



radial distribution function of O in water



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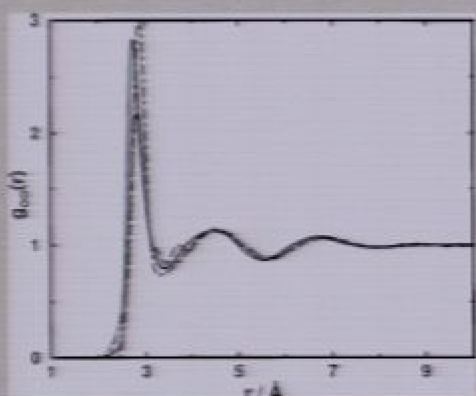

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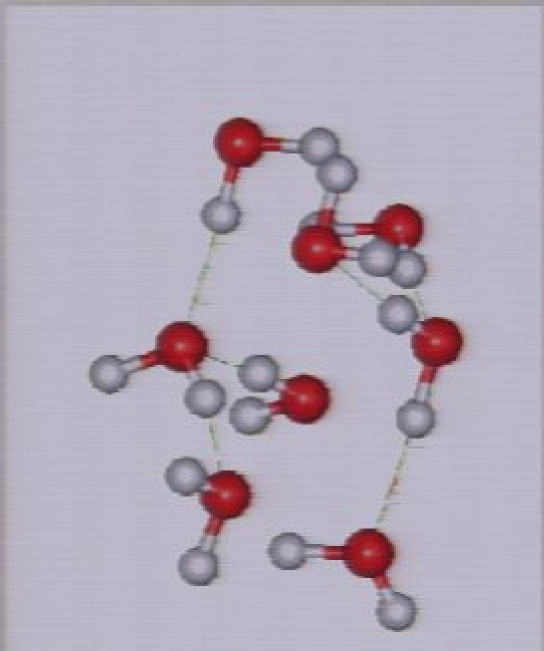

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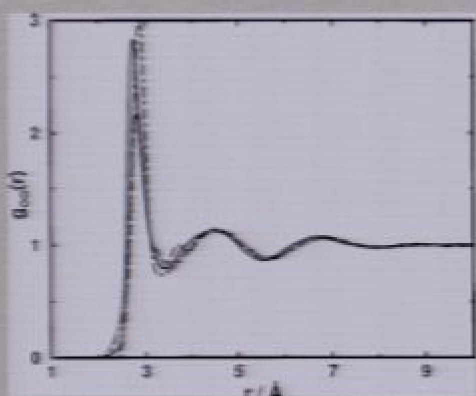

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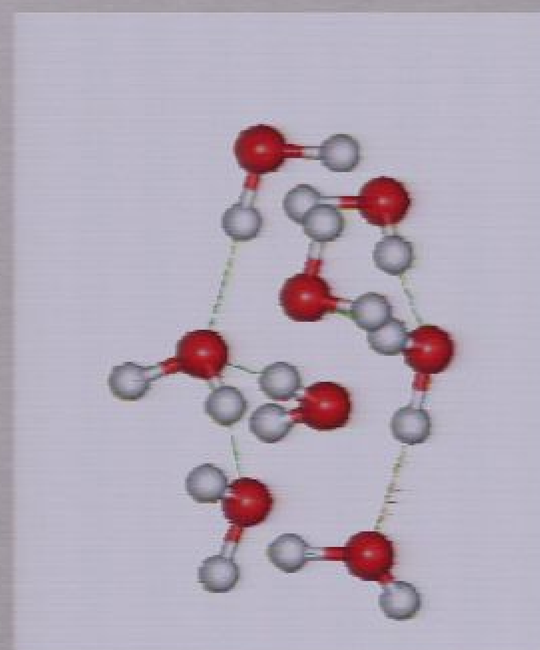

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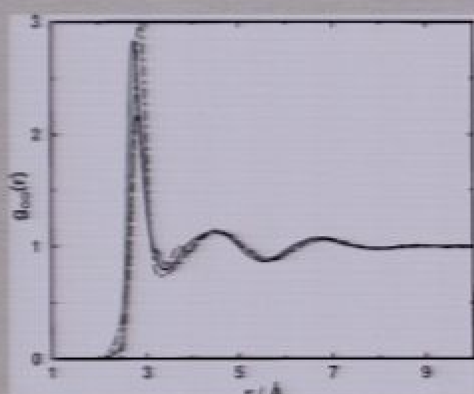

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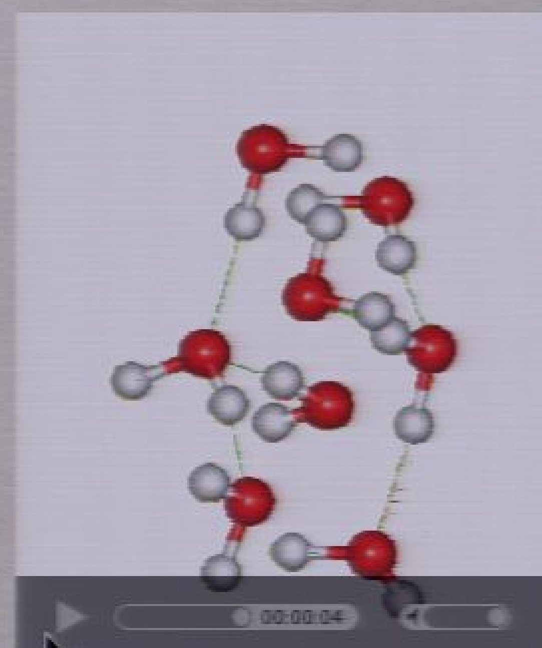

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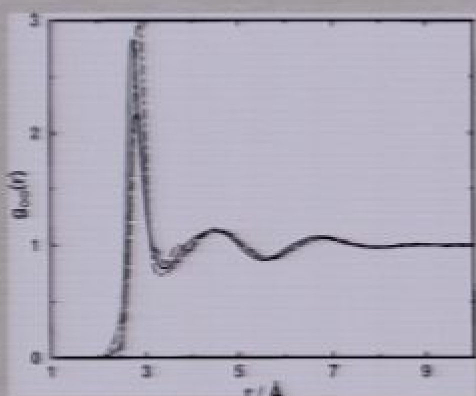

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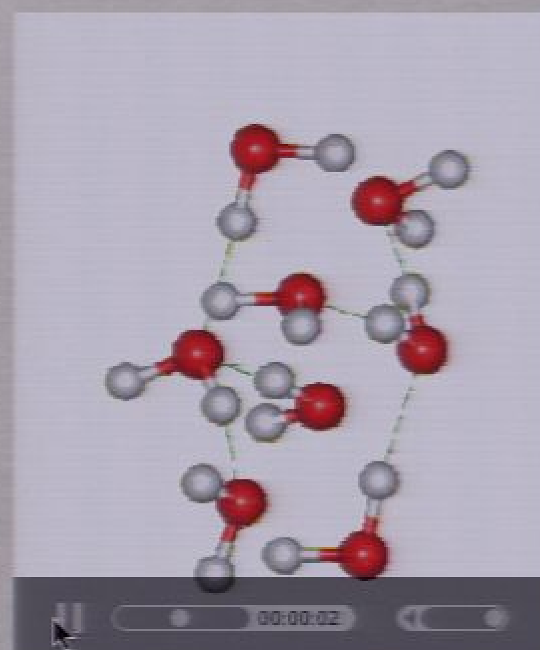

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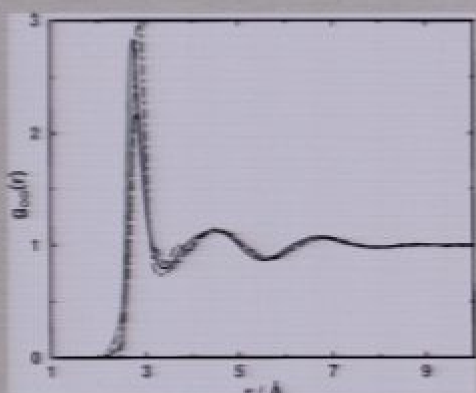

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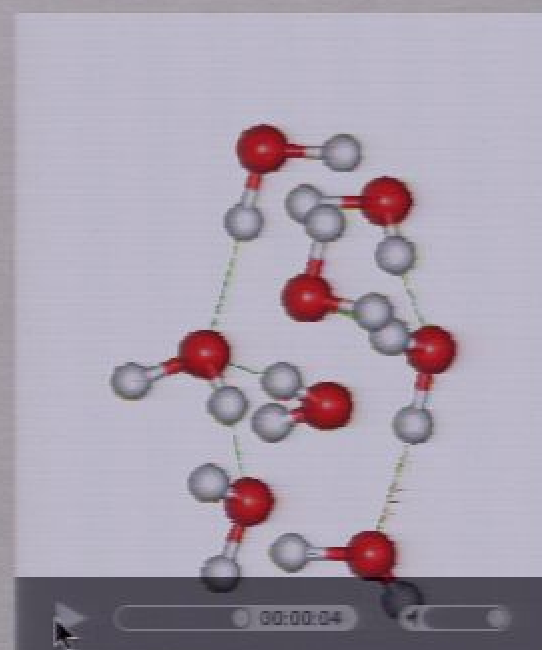
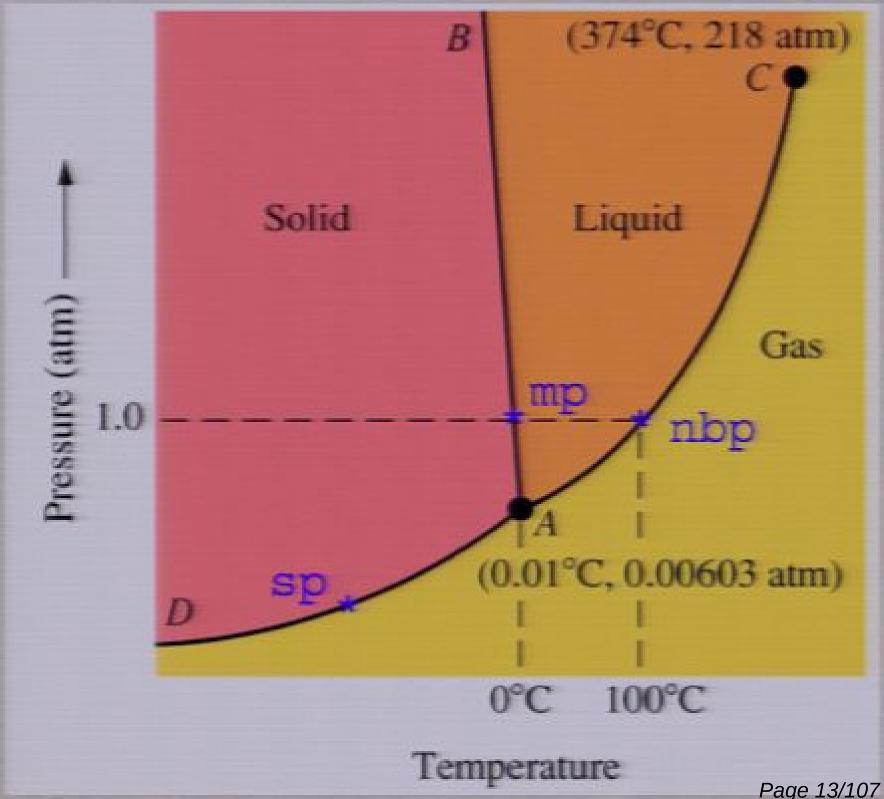


LIQUID = GAS?

- Classical liquids are continuously connected to gases - *not* a distinct phase of matter
  - but still interesting



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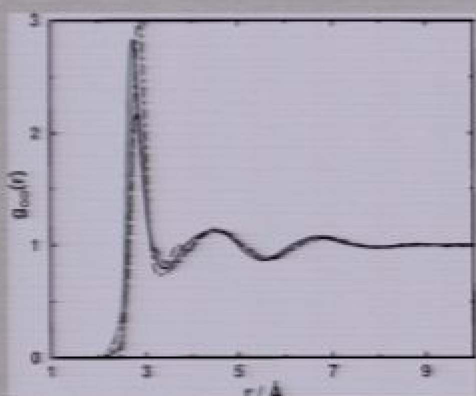

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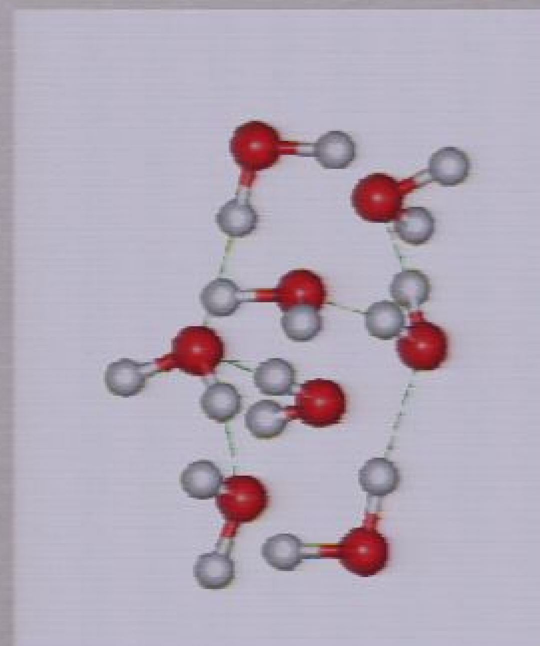

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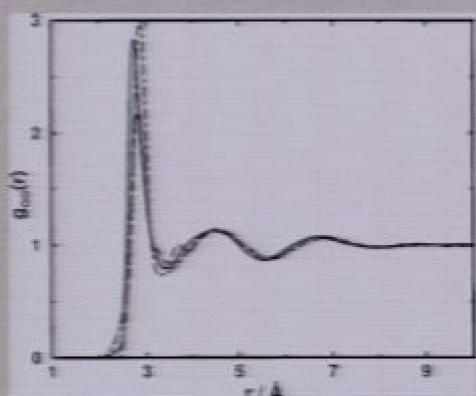

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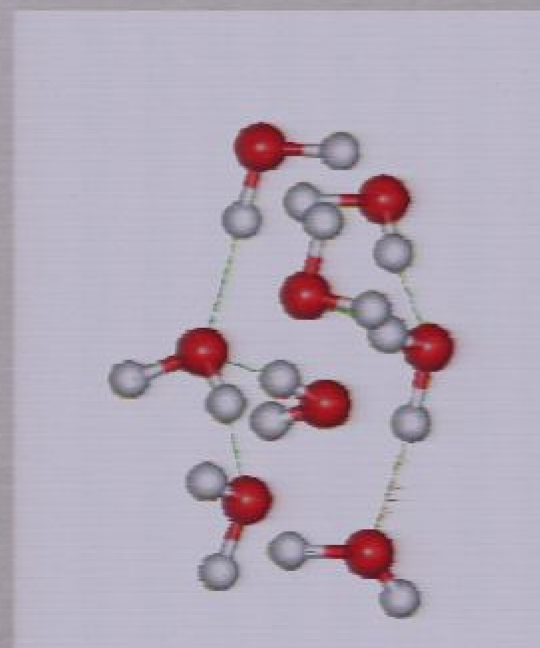

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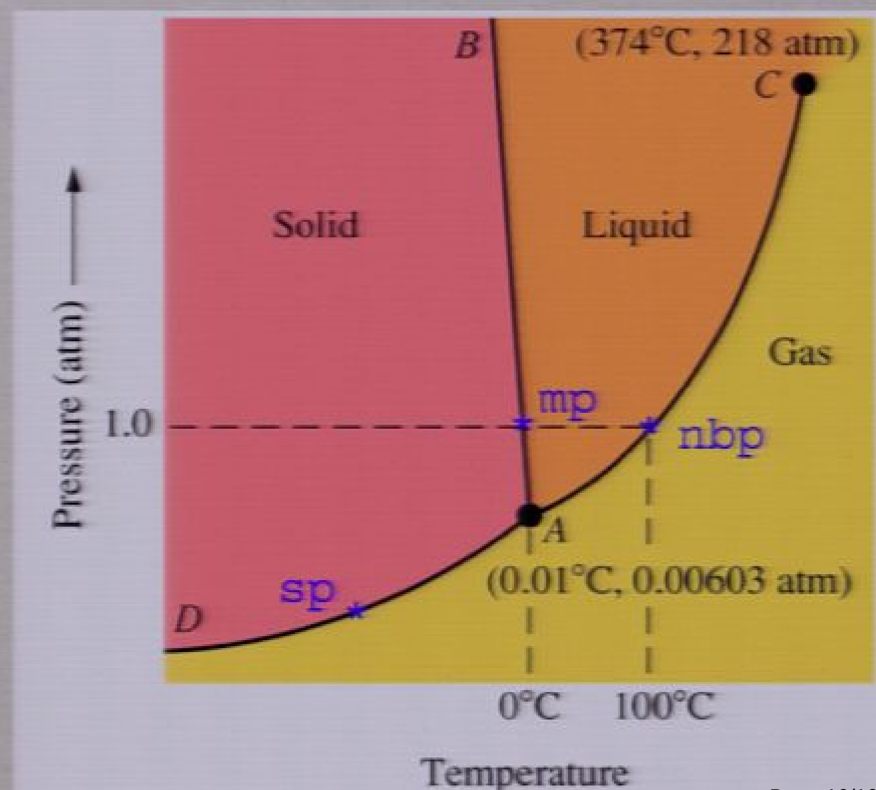
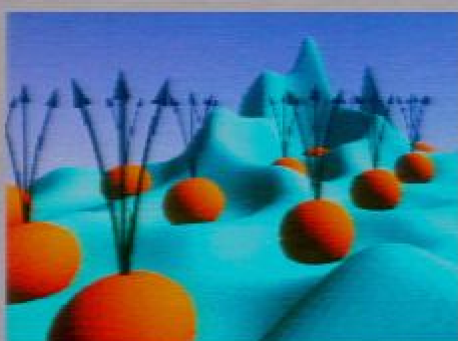
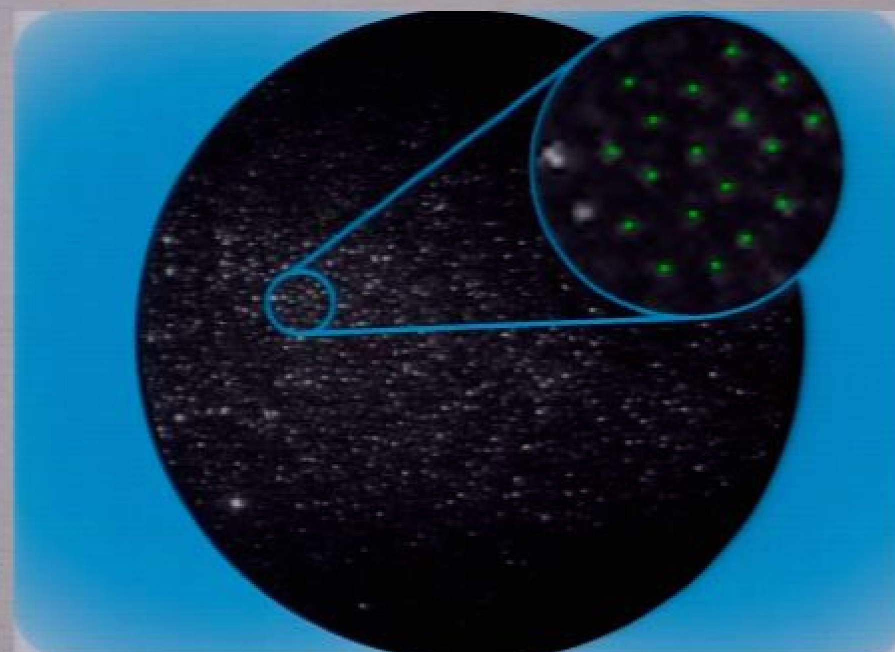


Quantum Liquids

- Quantum liquids *can* be distinct phases of matter



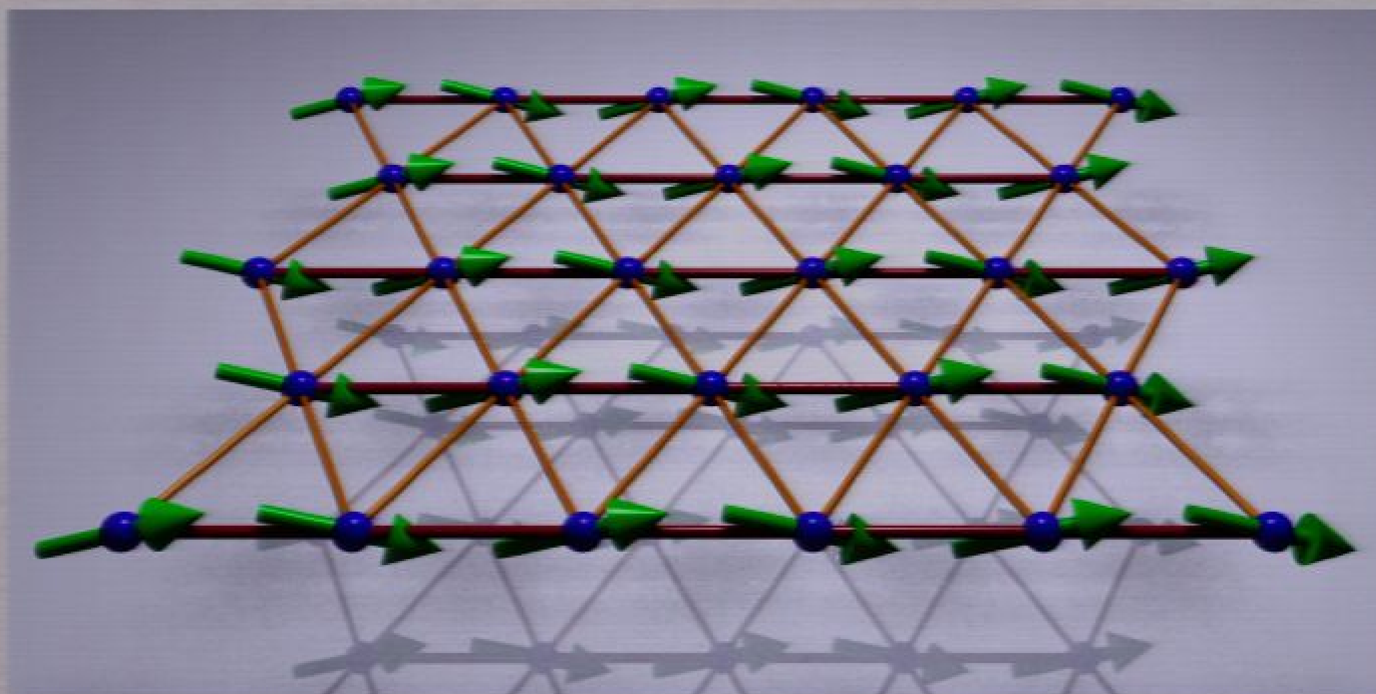
FQHE



Superfluid helium
(a more subtle broken symmetry)

Spins

- These phases (and more!) have analogies in magnetic materials

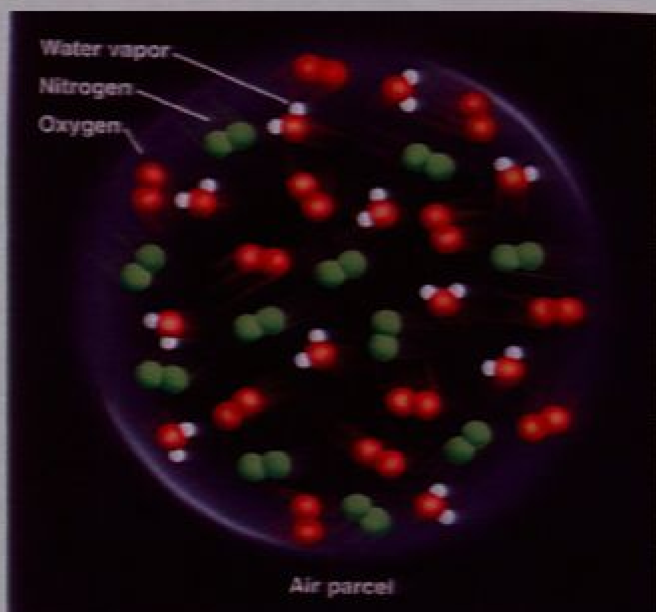


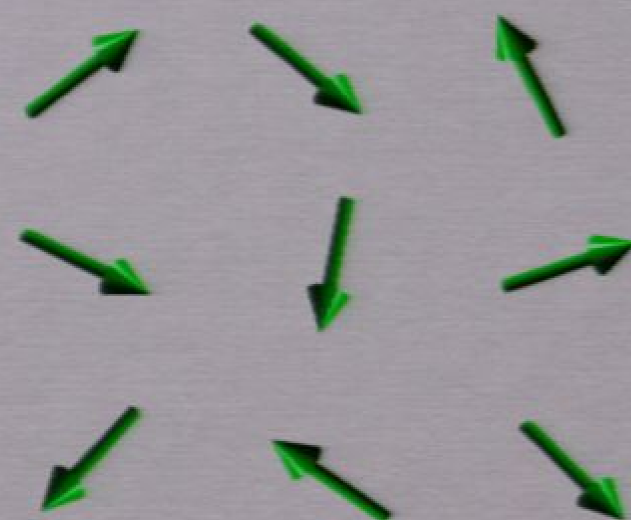
Spin Analogy

- Molecule ⇒ spin

Spin Analogy

- Gas ⇒ paramagnet



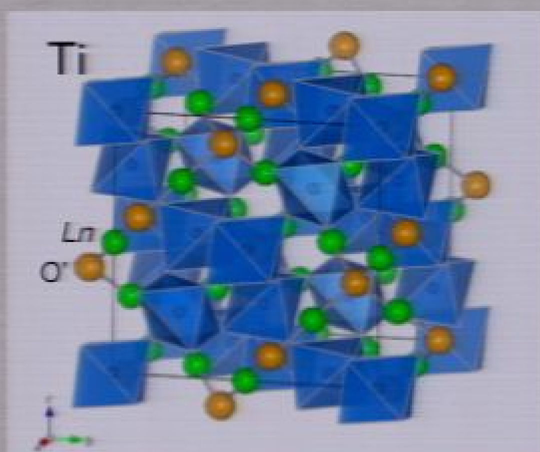


independent molecules

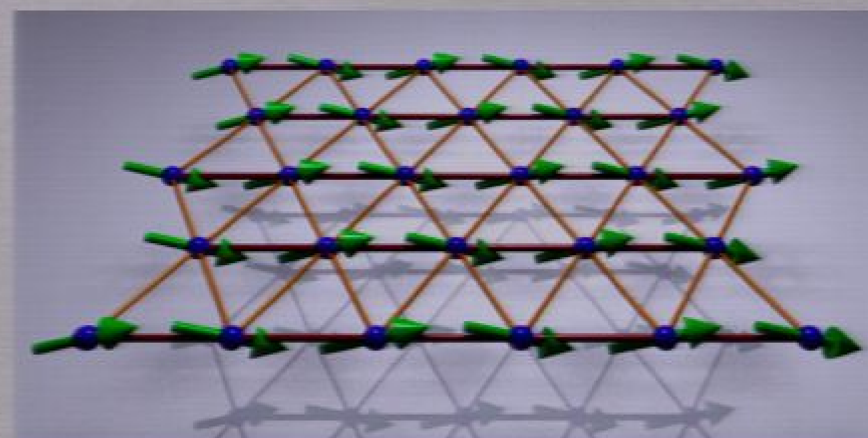
independent spins

Spin Analogy

- Solid ⇒ (anti)-ferromagnet



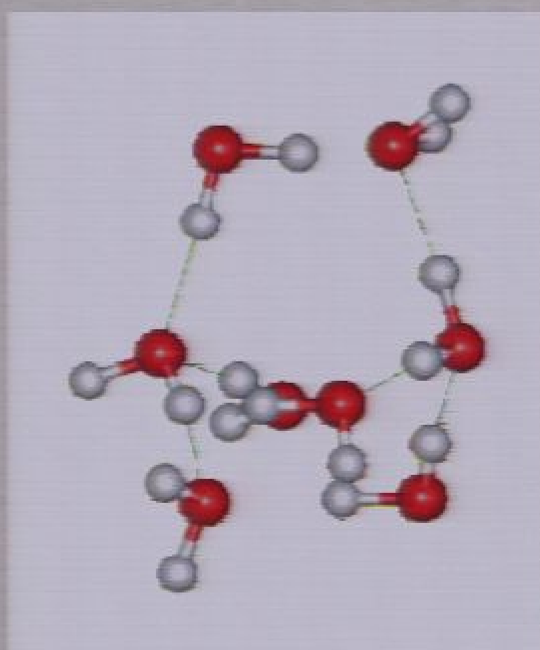
atoms fixed



Spins static

Spin liquids?

- Liquid ⇒ "spin liquid" = "cooperative paramagnet"



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What makes spins correlated but not ordered?

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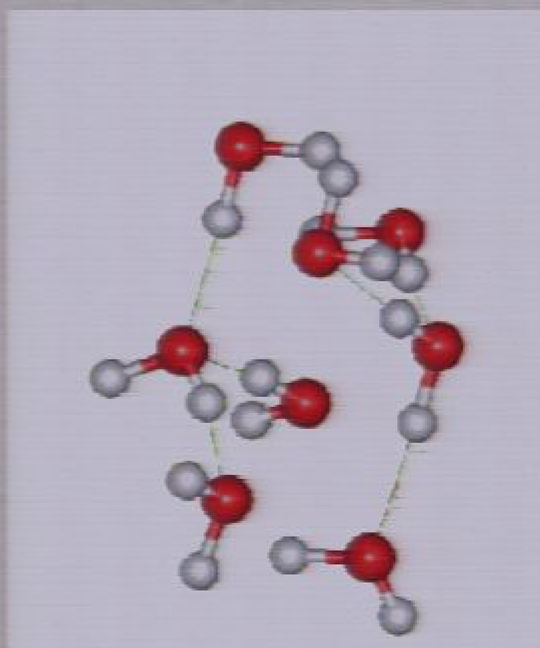

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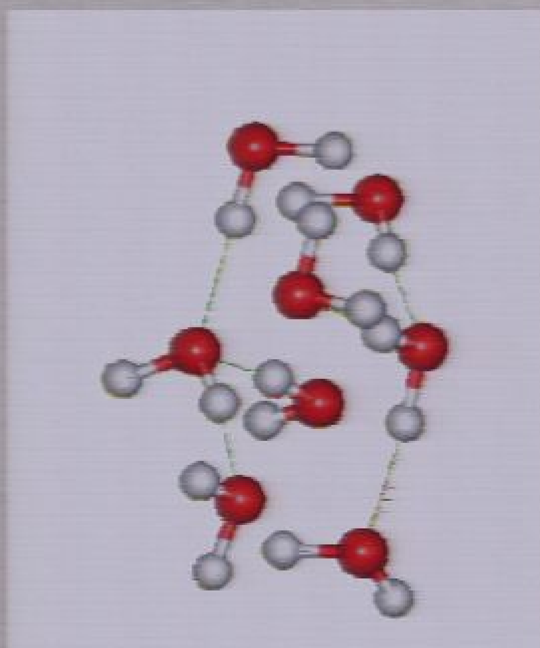

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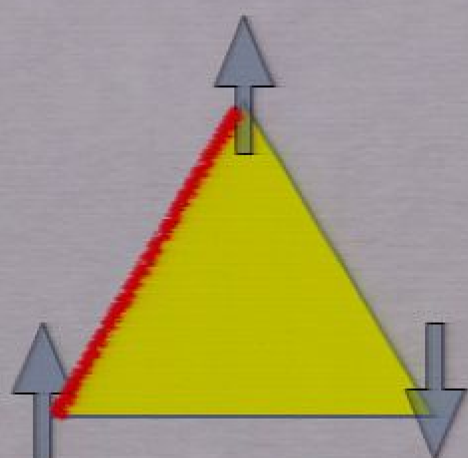
What makes spins correlated but not ordered?

FRUSTRATION

- Simplest idea: pairwise exchange interactions cannot be simultaneously satisfied

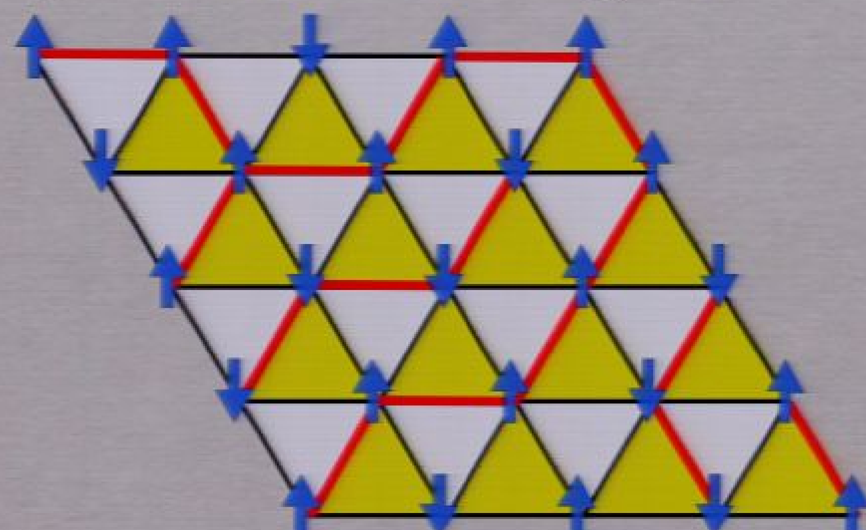


"geometric frustration"

- But this is a bit simplistic, and overstates the problem

Degeneracy

- Ideally: frustration induces ground state degeneracy, and in spin liquid, spins fluctuate within those ground states
- e.g. triangular lattice Ising antiferromagnet

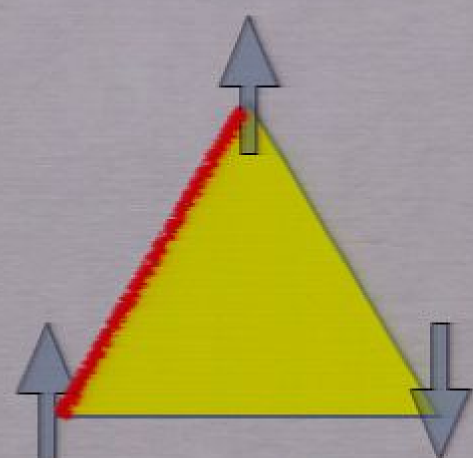


1 frustrated bond per triangle

Wannier (1950): $\Omega = e^{S/k_B}$ $S \approx 0.34 N k_B$

Frustration

- Simplest idea: pairwise exchange interactions cannot be simultaneously satisfied



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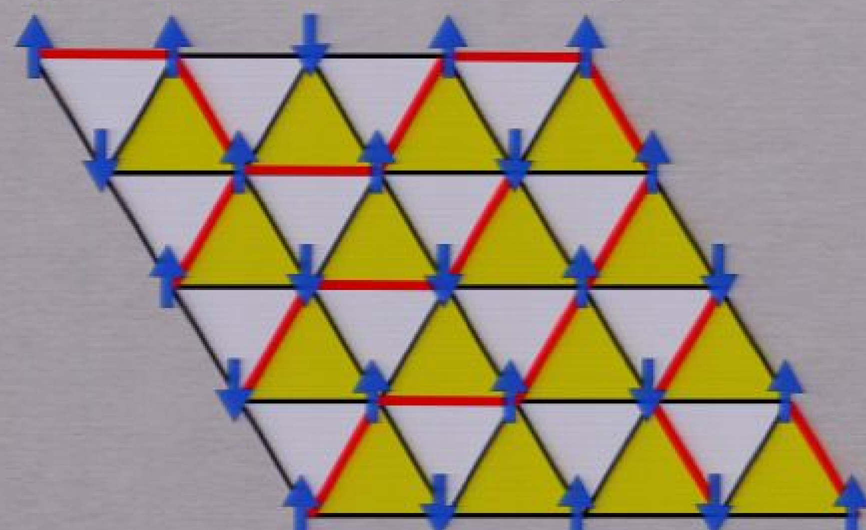

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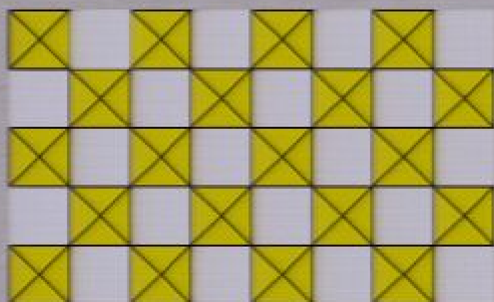
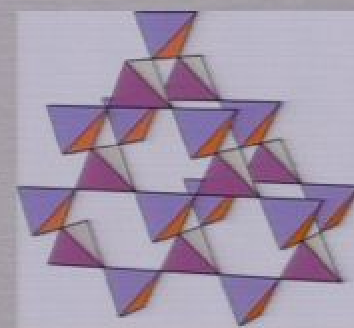
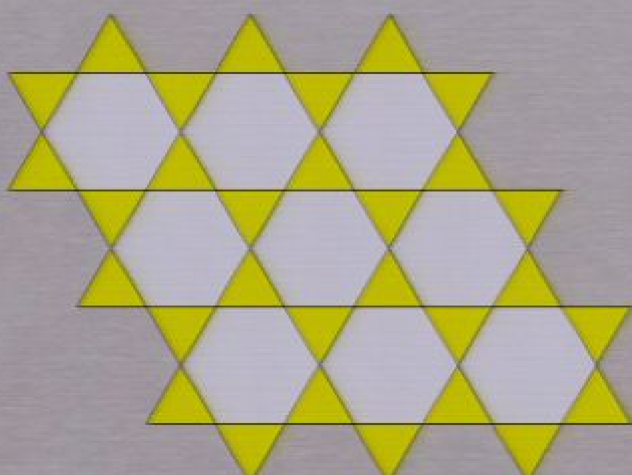
OTHER LATTICES



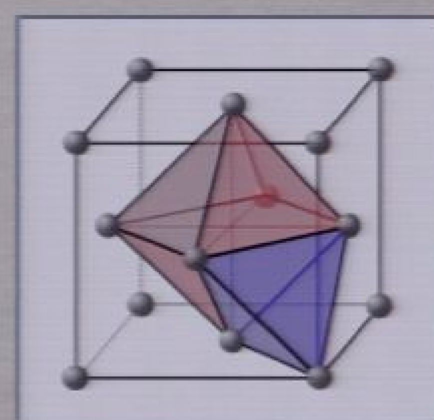
checkerboard $S \sim 0.216\ N\ k_B$



pyrochlore $S \sim 0.203\ N\ k_B$



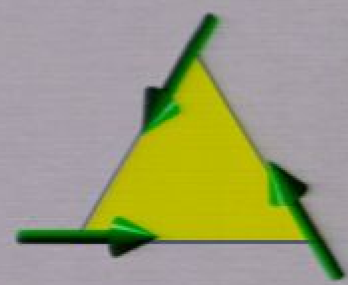
kagome $S \sim 0.5\ N\ k_B$



FCC: $S \sim c\ N^{1/3}\ k_B$

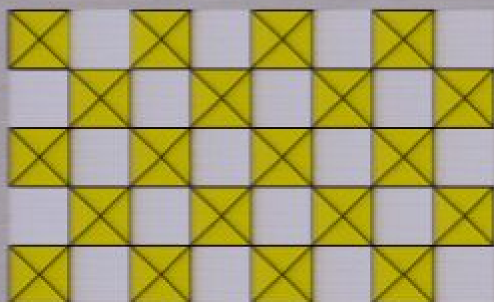
Complications

- Heisenberg spins



- More complex interactions
- Quantum effects: level repulsion avoids degeneracies
    - a true Quantum Spin Liquid is a more subtle thing

Other lattices



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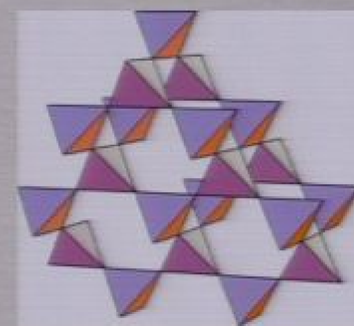

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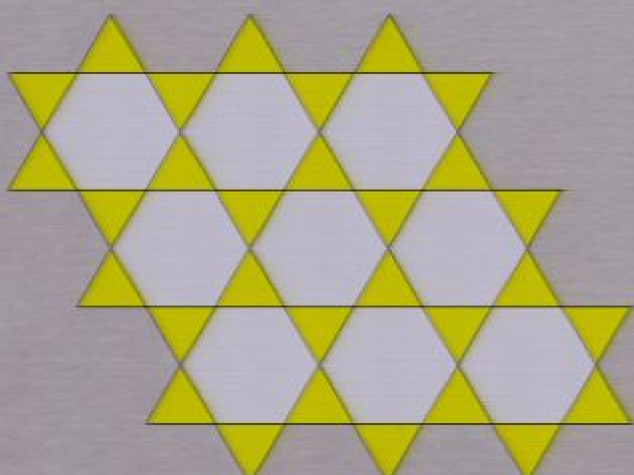

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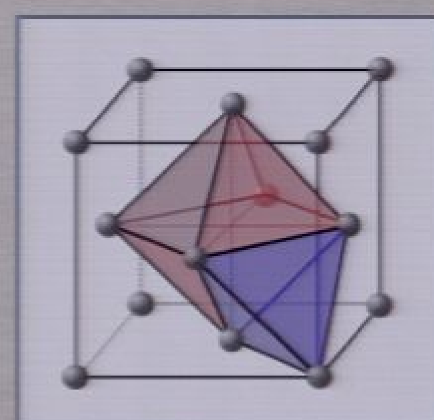

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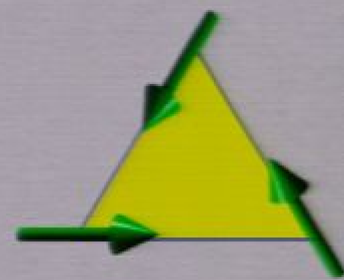

- Heisenberg spins
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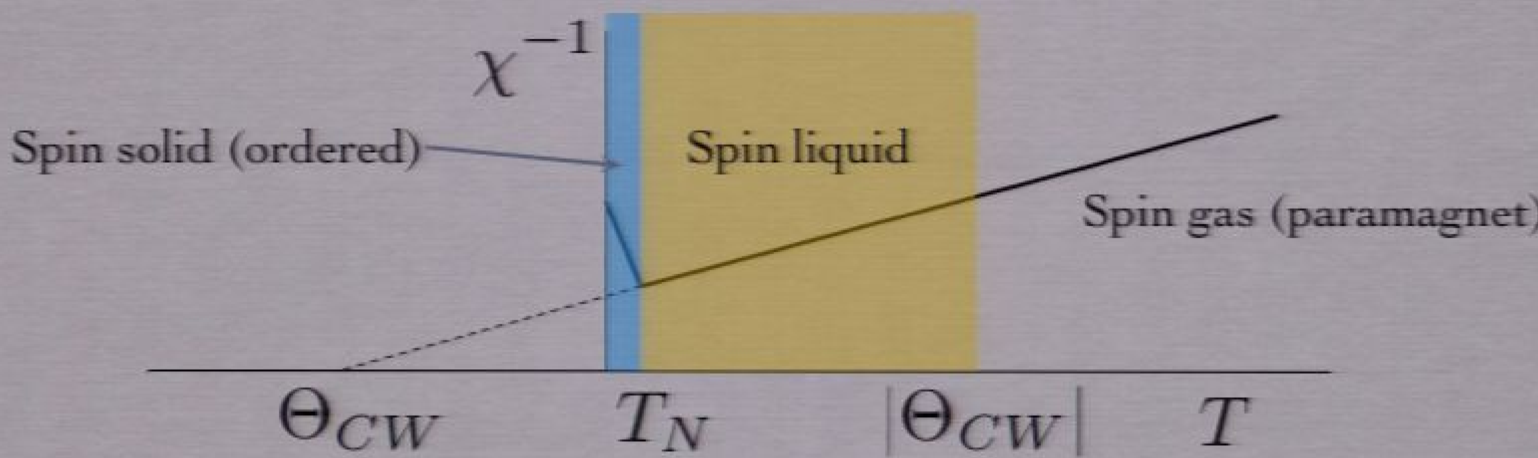
EMPIRICAL MEASURE



- Local moments: Curie-Weiss law at high T

$$\chi \sim \frac{A}{T - \Theta_{CW}}$$

- Frustration/fluctuation parameter: $f = |\Theta_{CW}|/T_N$

- $f>>1$: wide regime $T_N < T < |\Theta_{CW}|$

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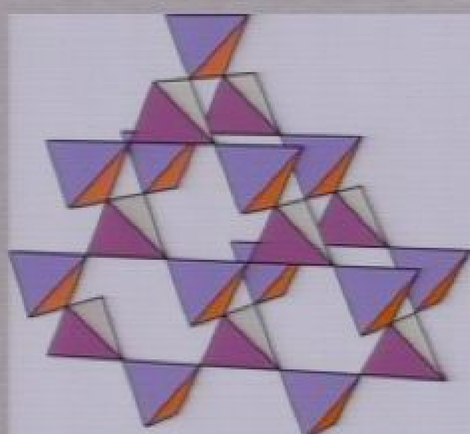
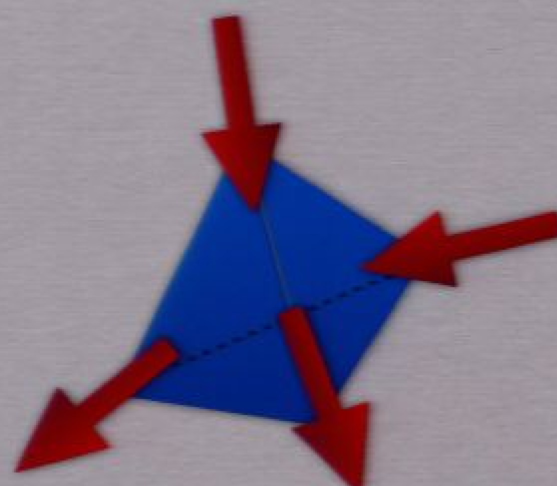
Spin Ice (for theorists)

- In certain rare earth magnets, e.g. $Ho_2Ti_2O_7$, spins behave like classical vectors of fixed length, on the sites of a pyrochlore lattice, oriented along *local* easy axes

$$\vec{S}_i = \hat{e}_i \sigma_i$$





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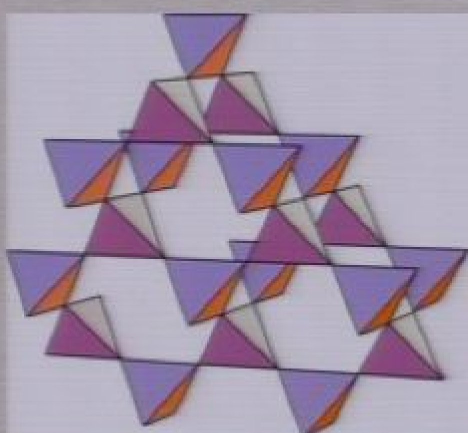

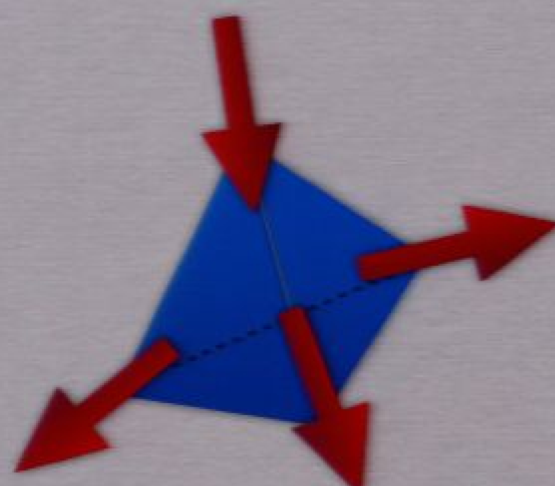

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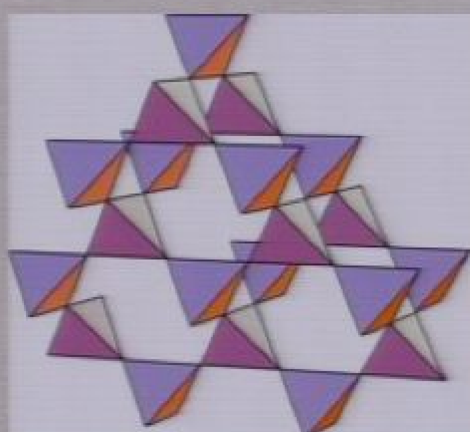

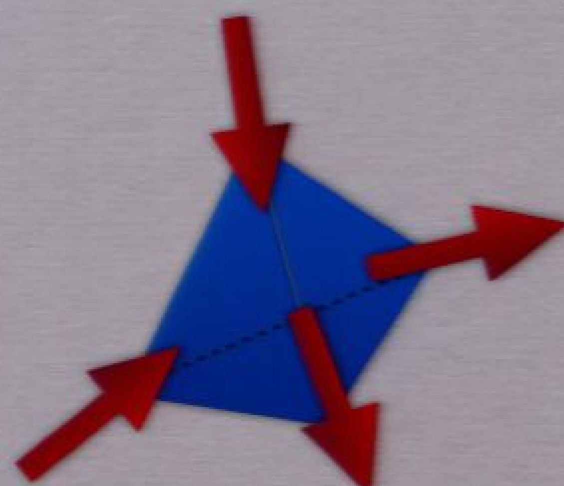

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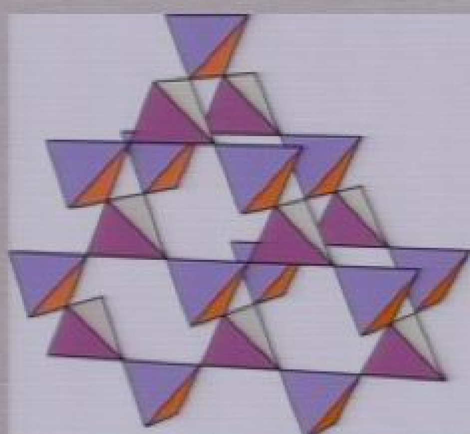
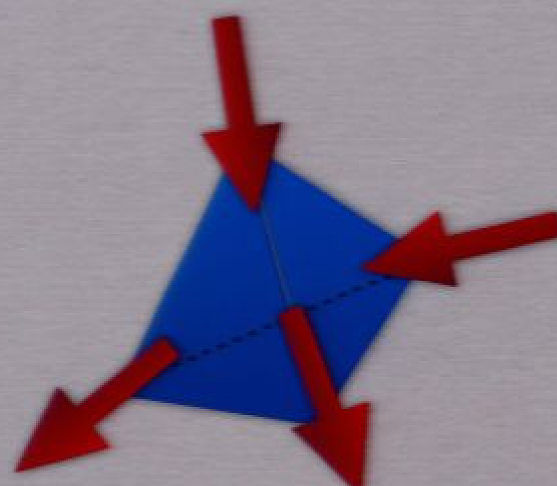


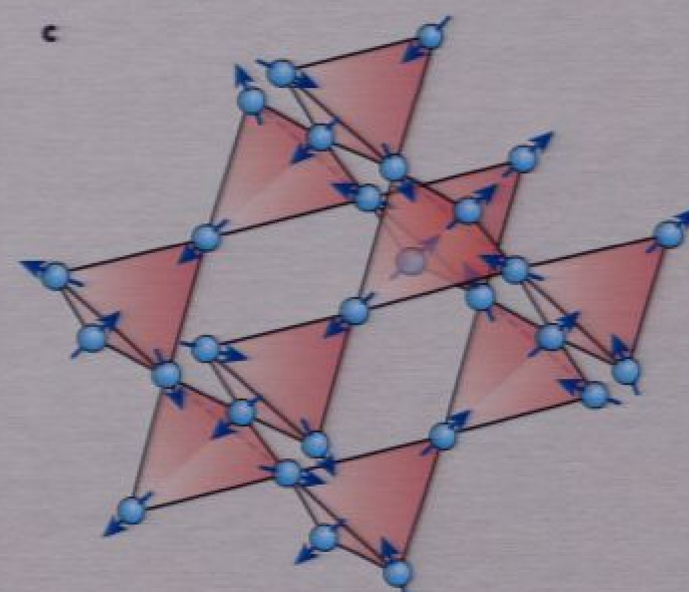


Spin Ice (for theorists)

- Exchange (due largely to dipolar interactions) is *ferromagnetic*
  - Prefers "2 in - 2 out" states

$$-J\vec{S}_i \cdot \vec{S}_j = \frac{J}{3}\sigma_i\sigma_j$$

same as Ising antiferromagnet



"ice rules"

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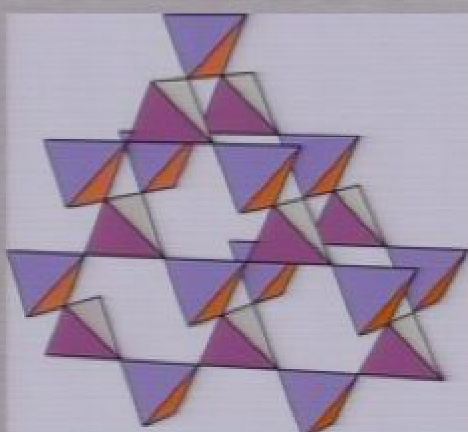
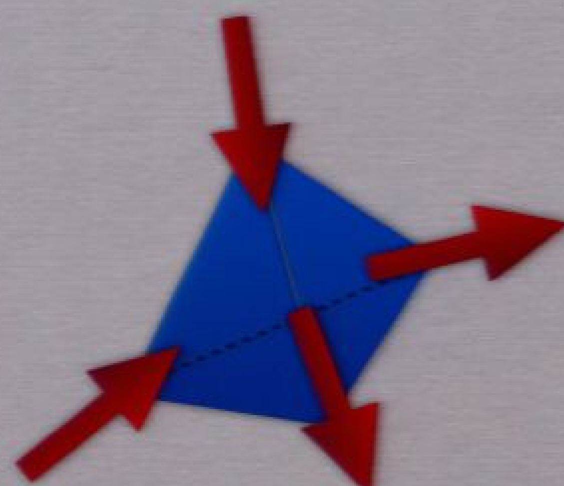
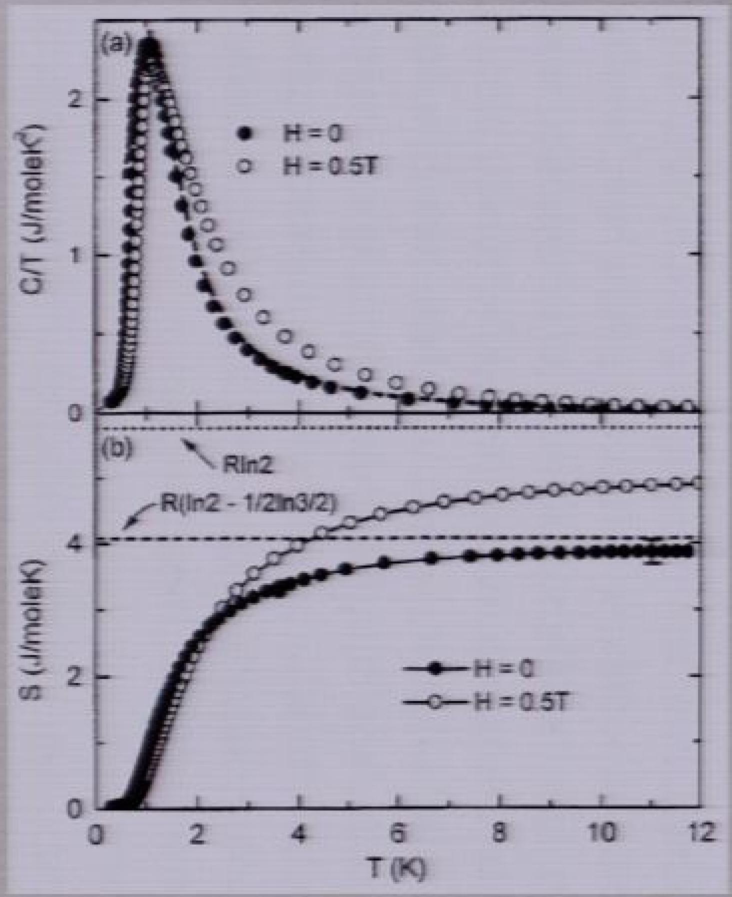




Entropy

- The integrated specific heat of $Dy_2Ti_2O_7$ showed explicitly that the entropy did not vanish at low temperature
  - quantitative agreement with Pauling's 1935 estimate



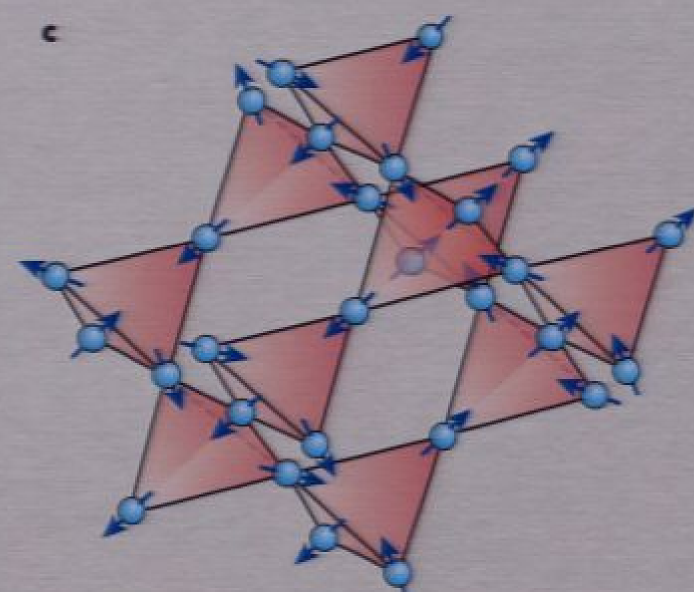
A.P. Ramirez *et al*, 1999

Spin Ice (for theorists)

- Exchange (due largely to dipolar interactions) is *ferromagnetic*
  - Prefers "2 in - 2 out" states

$$-J\vec{S}_i \cdot \vec{S}_j = \frac{J}{3}\sigma_i\sigma_j$$

same as Ising antiferromagnet



"ice rules"

ENTROPY

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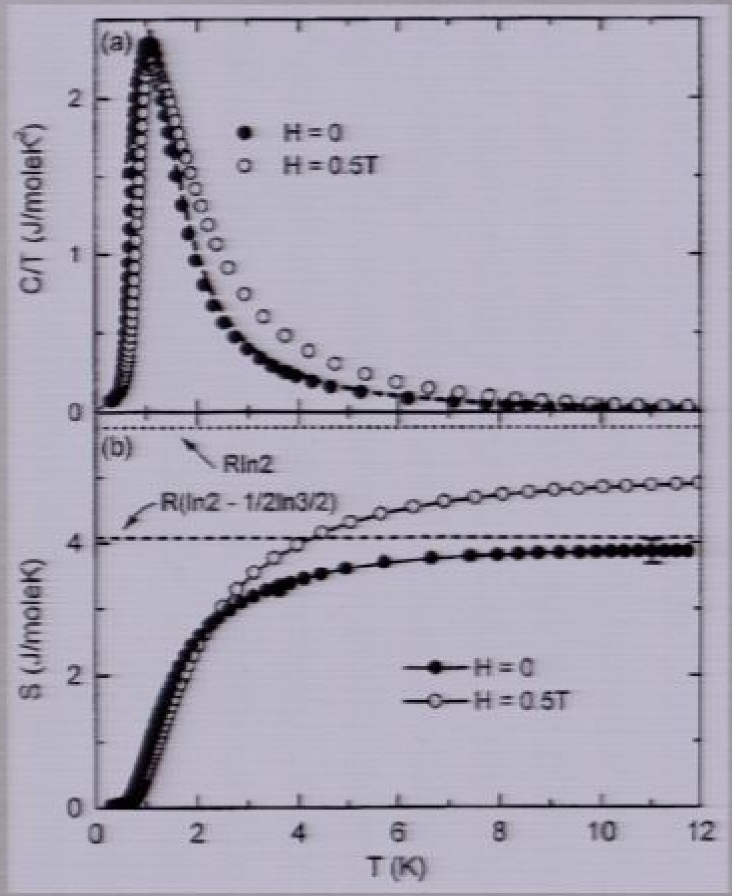
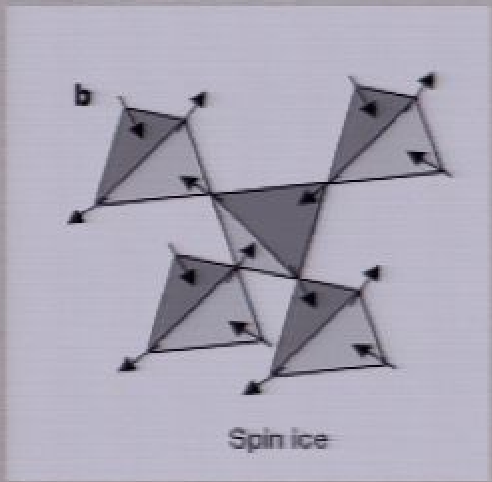


A.P. Ramirez *et al*, 1999

Spin liquid physics

- The spin liquid fluctuations are a form of "artificial magnetostatics" (classical)
  - ice rules: divergence free condition



$$\vec{S} \sim \vec{b}$$

$$\vec{\nabla} \cdot \vec{b} = 0$$

Power law correlations

- Effective theory

$$H_{\text{eff}} = \int d^3r\, \frac{c}{2}|\vec{b}|^2$$

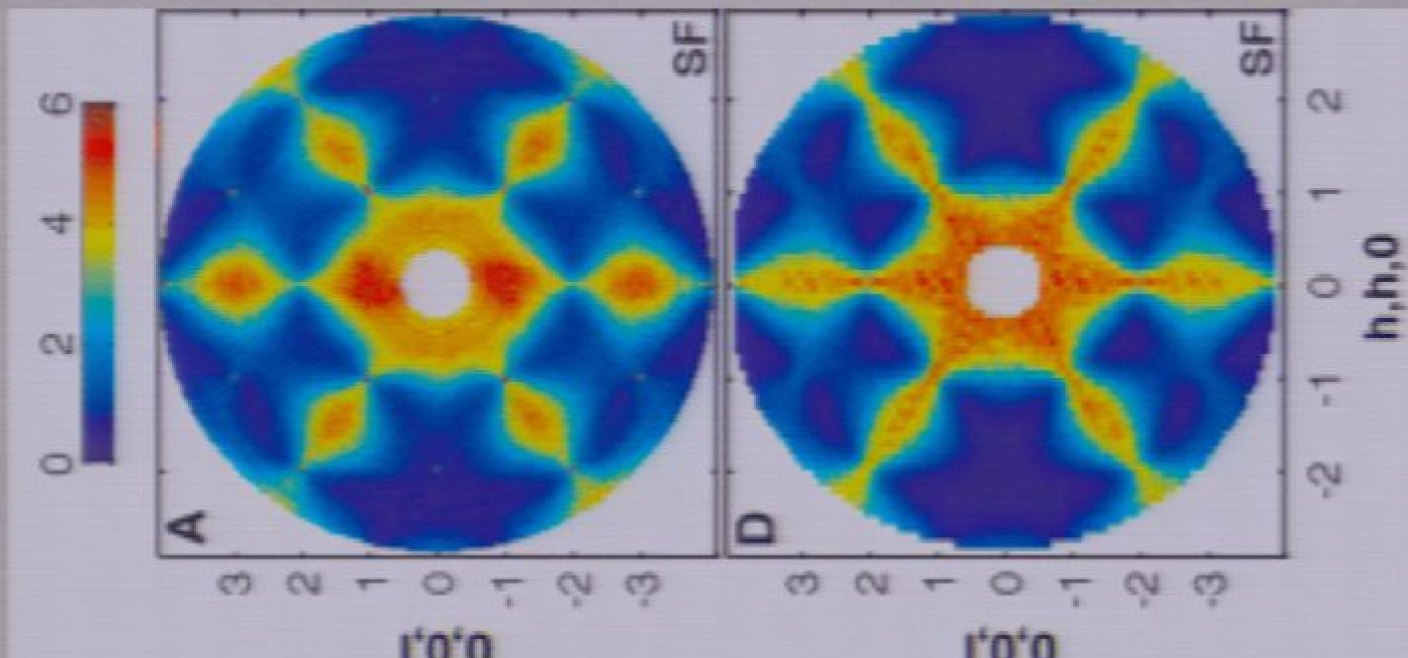
- Using vector potential b = ∇×a

$$\langle b_\mu(-k) b_\nu(k) \rangle = \frac{1}{c}\left(\delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2}\right)$$

- This is directly proportional to the static magnetic structure factor measured in a neutron experiment

$$S(K_{200} + k) \sim \frac{k_y^2 + k_z^2}{k^2}$$

PINCH POINTS IN $Ho_2Ti_2O_7$



experiment theory

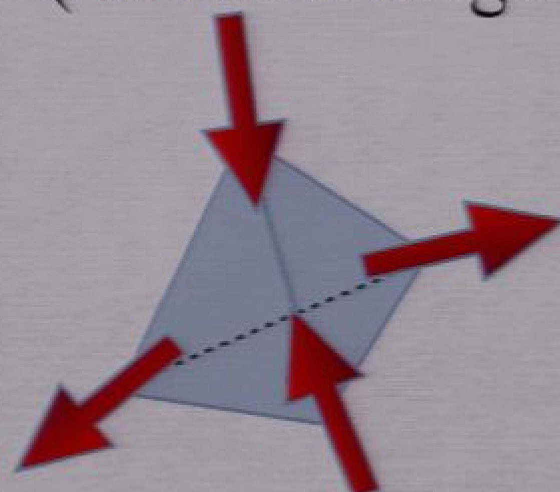
. Fennell *et al*, 2009

$$S(K_{200} + k) \sim \frac{k_y^2 + k_z^2}{k^2}$$ vanishes along lines

Magnetic monopoles

Castelnovo *et al*, 2008

- Defect tetrahedra are sources and sinks of "magnetic" flux (and real magnetization)



$$\vec{M} \propto \vec{b}$$

- It is a somewhat non-local object
  - Must flip a semi-infinite string of spins to create a single monopole

Magnetic monopoles

Castelnovo *et al*, 2008

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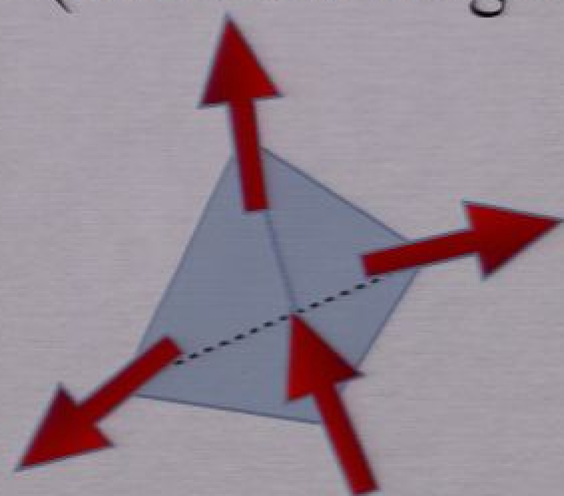

div b = 1

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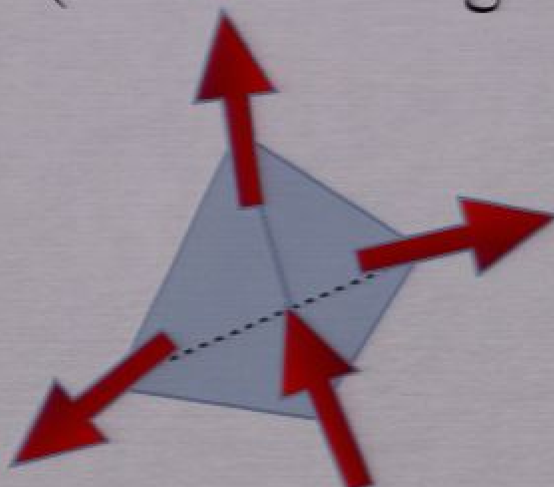

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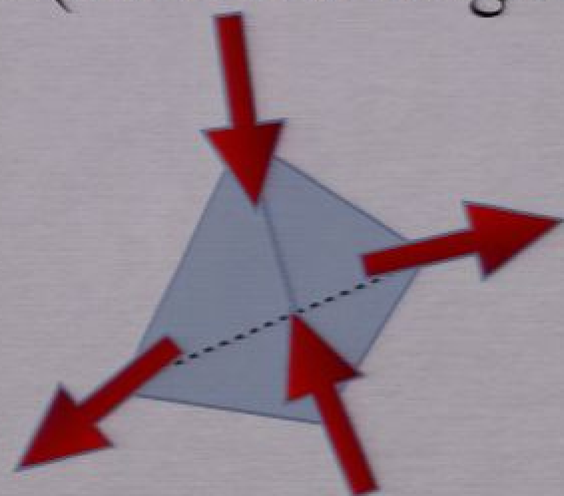

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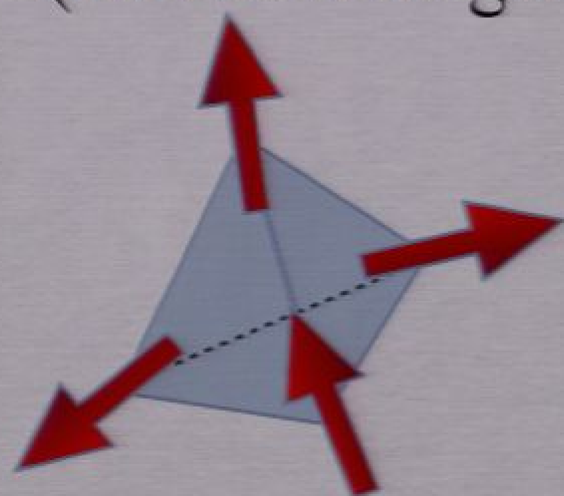

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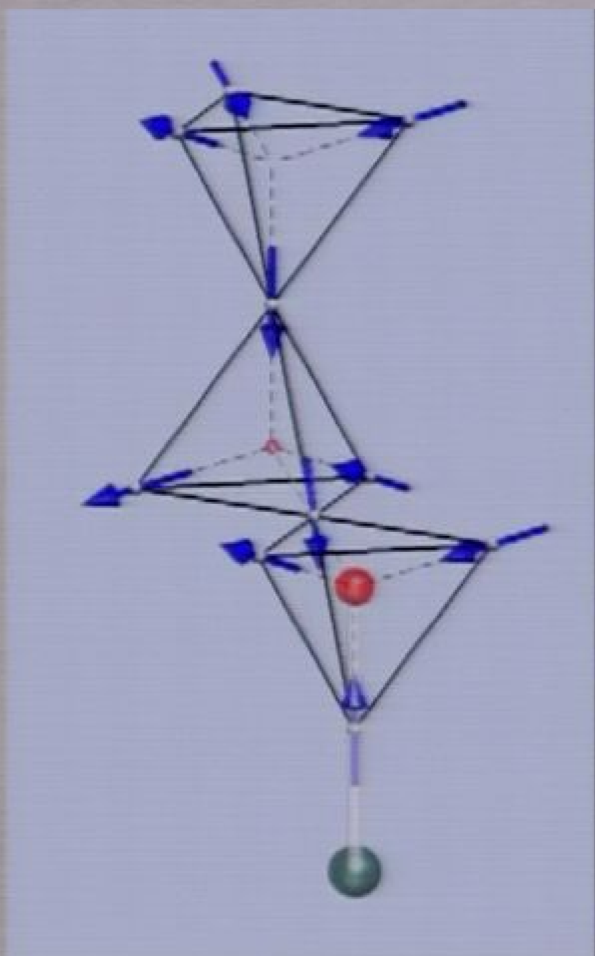
STRING



stolen (by somebody else on youtube) from Steve Bramwell

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  - this should be spoiled at low temperature by corrections to H
- Once created, the monopole can move by single spin flips

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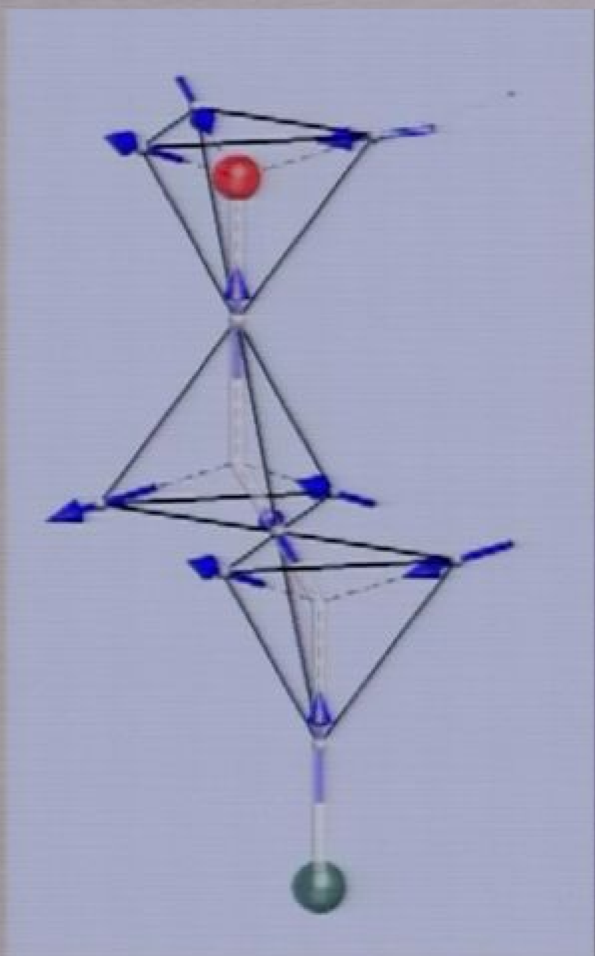

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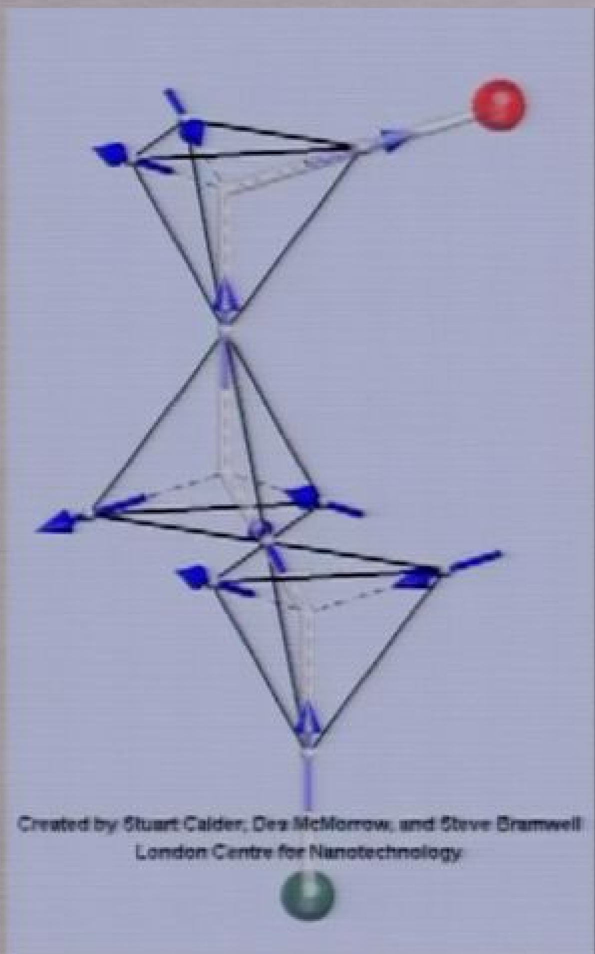

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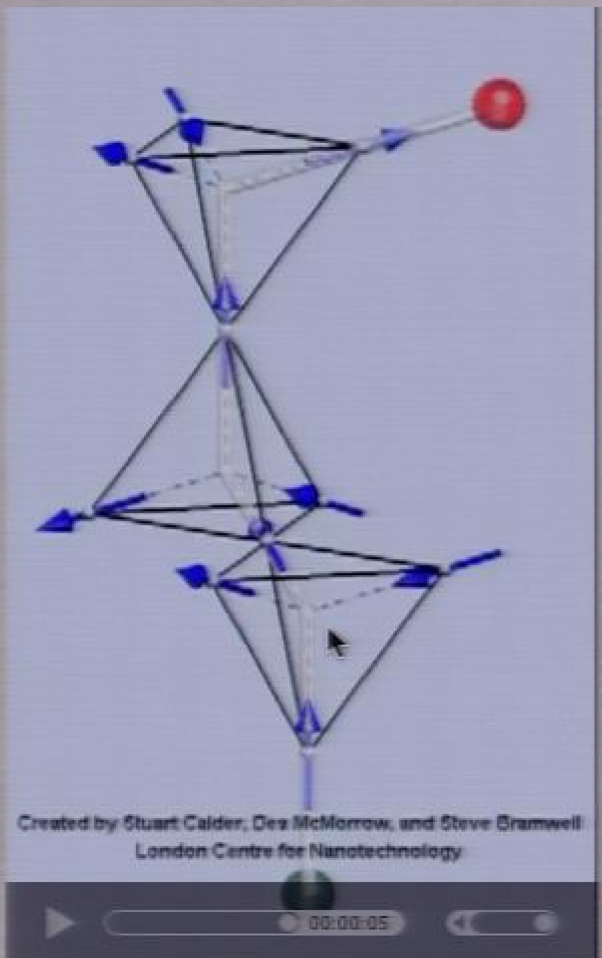

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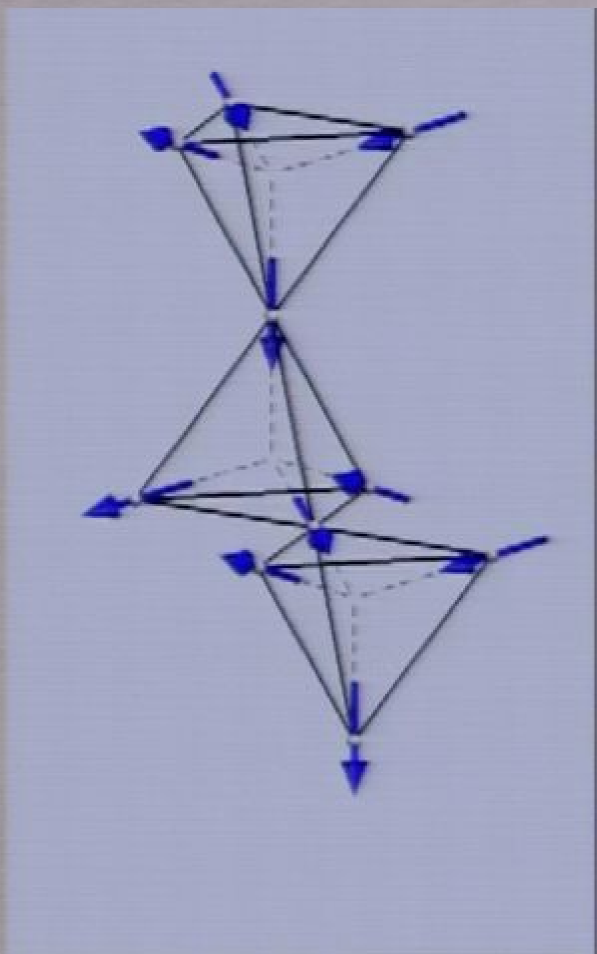

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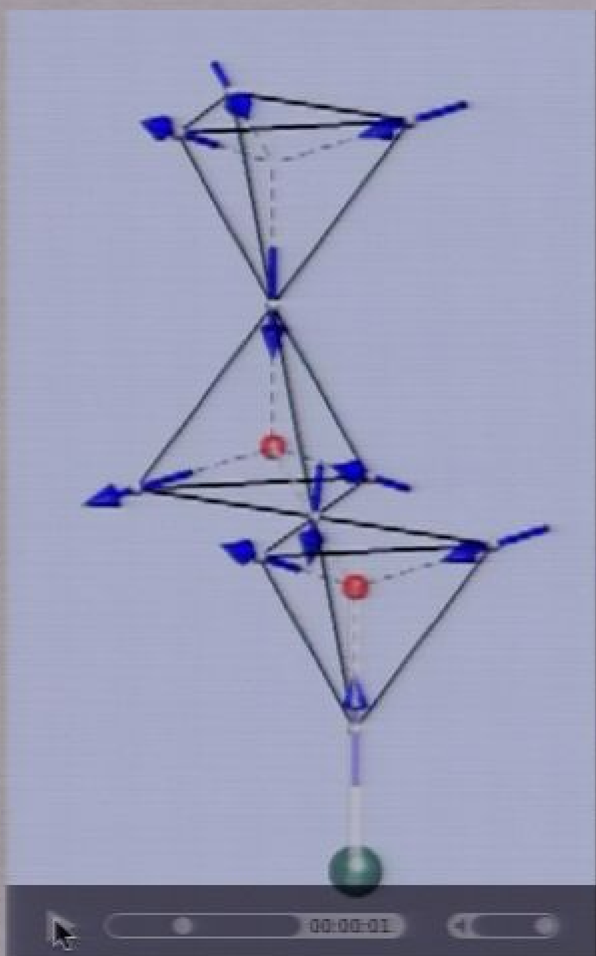

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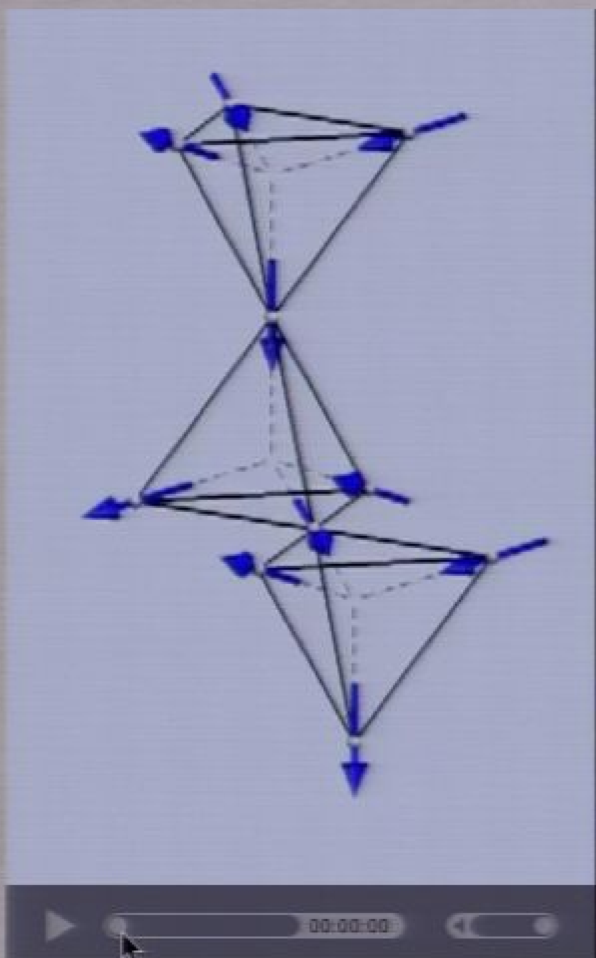

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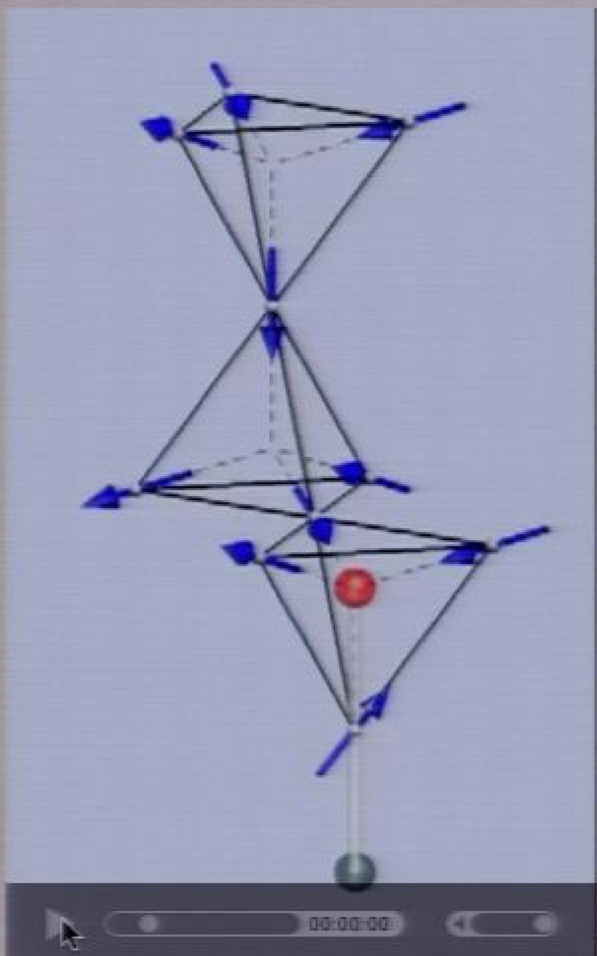

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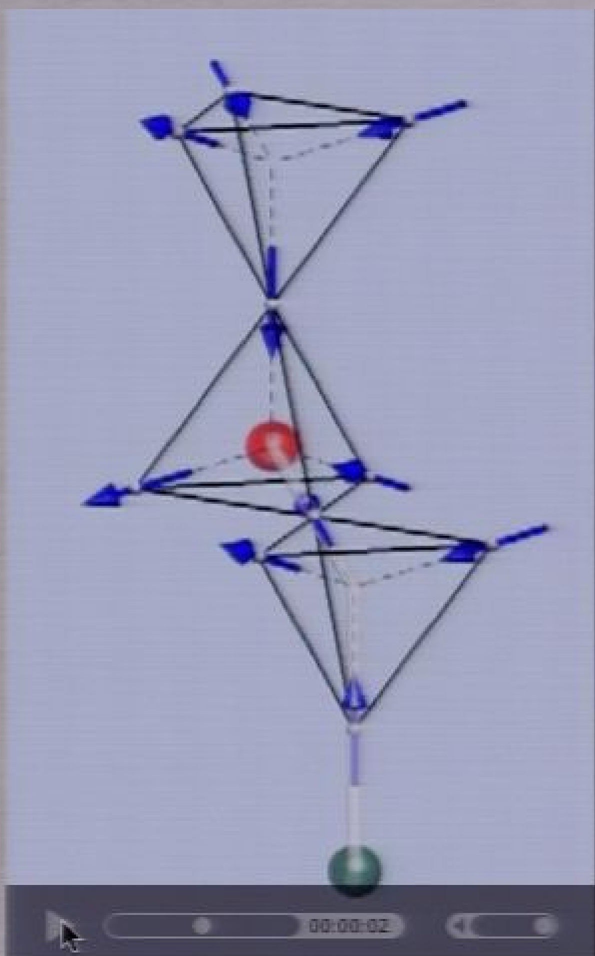

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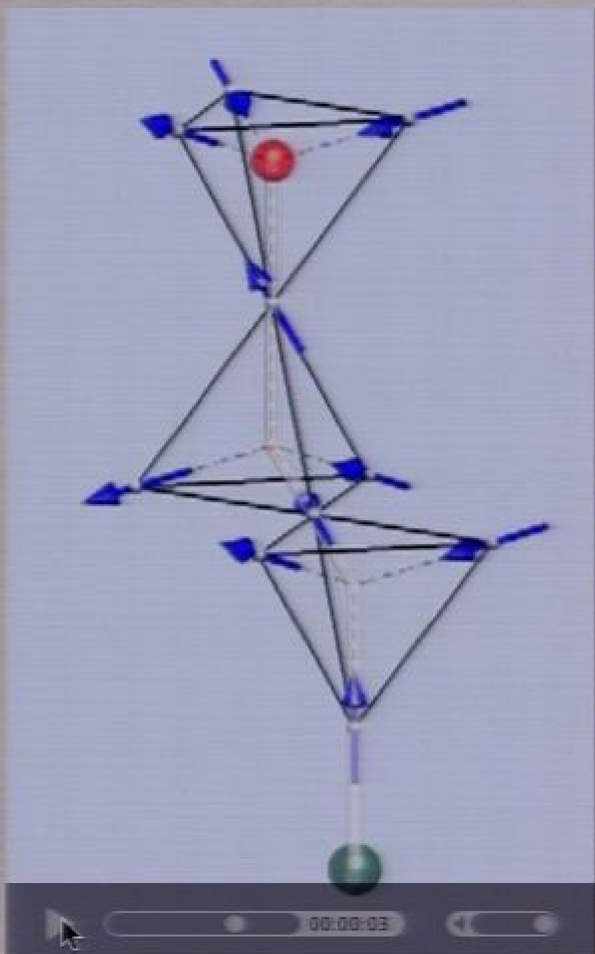

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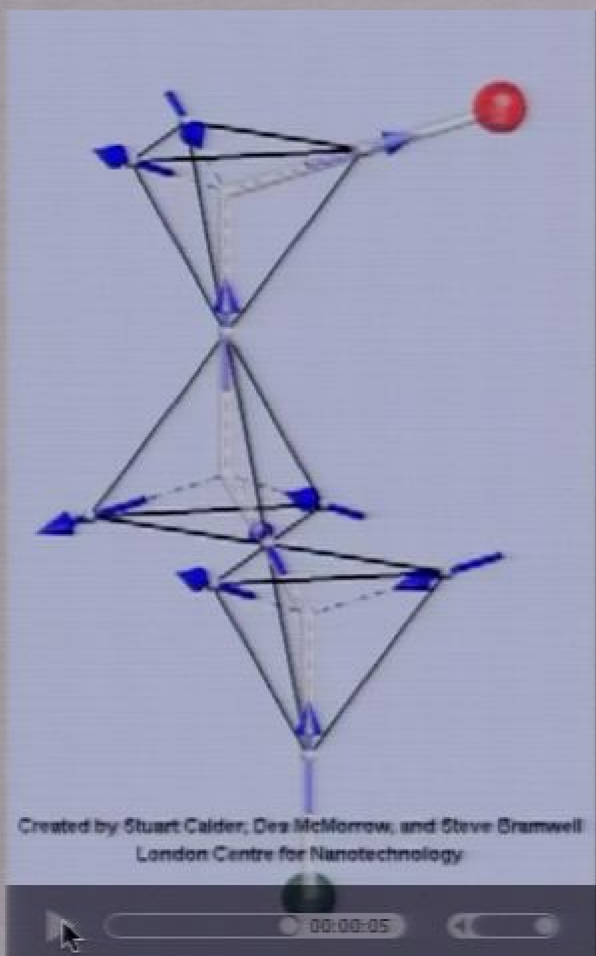

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Experimental evidence for monopoles

- Careful study of quasi-activation behavior of magnetization relaxation rate (Jaubert +Holdsworth, 2009)
  - measures the energy of a monopole
- Magnetic "Wien" effect (Bramwell et al, 2009)
  - measures a monopole's magnetic charge
- Several neutron measurements see "strings" in applied fields
- I suggest you talk to Michel Gingras to learn more about these experiments

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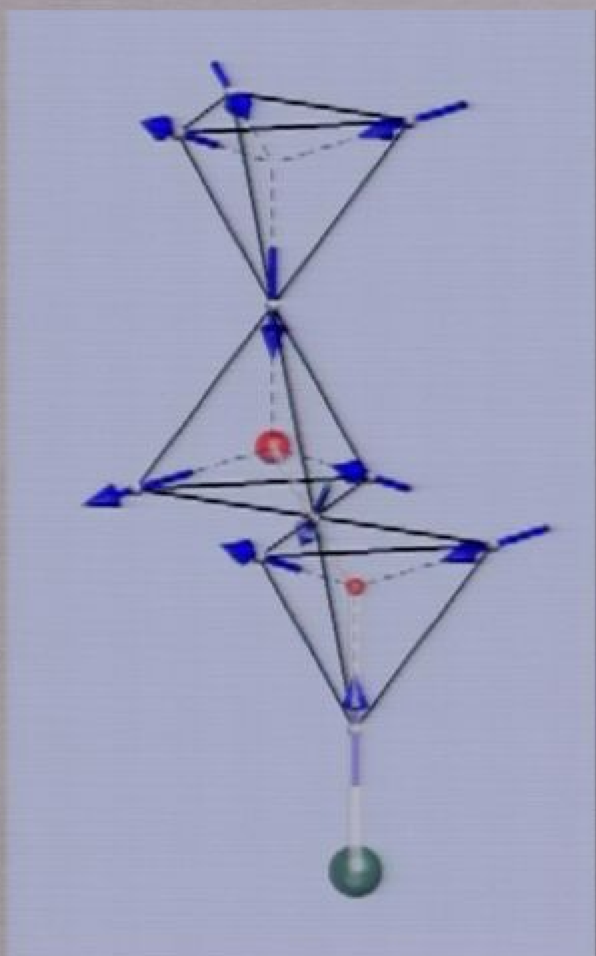

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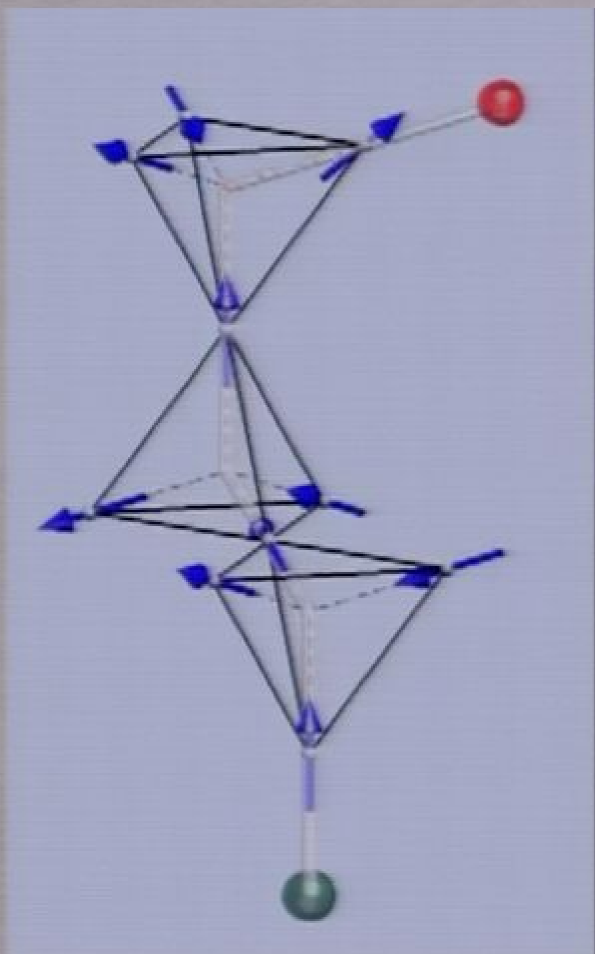

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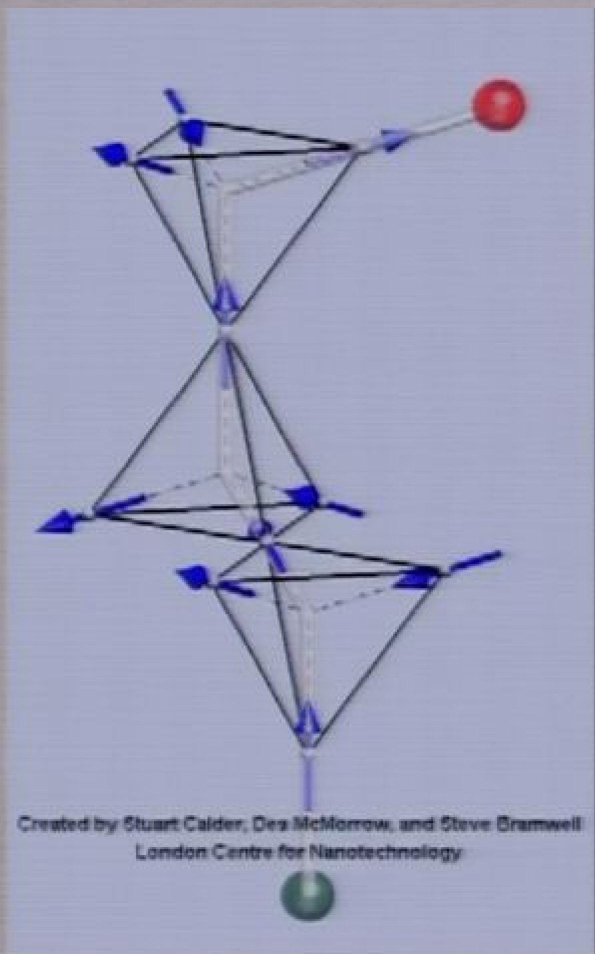

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Outline

- Liquids and spin liquids
- Spin ice: a classical spin liquid
- **Quantum spin liquids**
- How we look for them and what we've found so far

Quantum Spin Liquids

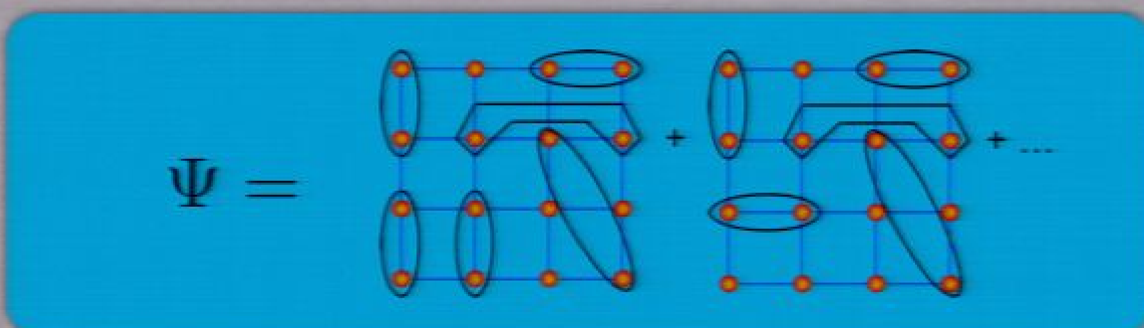
- In a classical spin liquid (spin ice), spins eventually freeze at low temperature
  - c.f. R. Melko et al: spin ice *should* order, but falls out of equilibrium
  - eventually, the strings become "visible" and monopoles are confined
- In a true *quantum* spin liquid, spins fluctuate even at T=0, and there are no observable strings

Why QSLs?

- Fascinating exotic states of matter
  - fractional quantum numbers and fractional (even non-abelian) statistics
  - emergent gauge fields
  - non-trivial interacting field theories
- Natural progenitors of superconductivity

RVB States

- Anderson (73): ground states of quantum magnets might be approximated by superpositions of singlet "valence bonds"

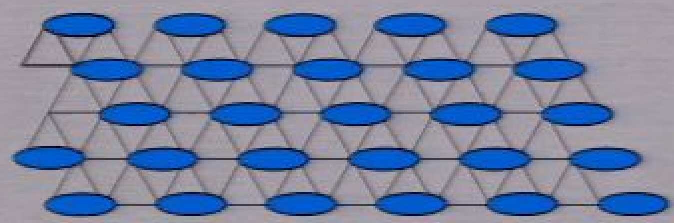


- Valence bond = singlet

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

VB STATES

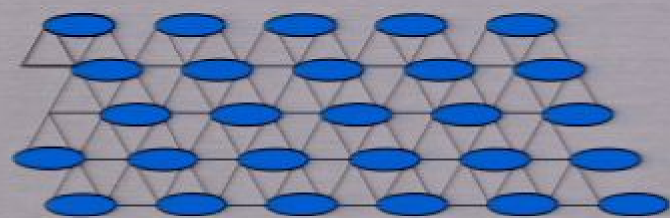
VBS



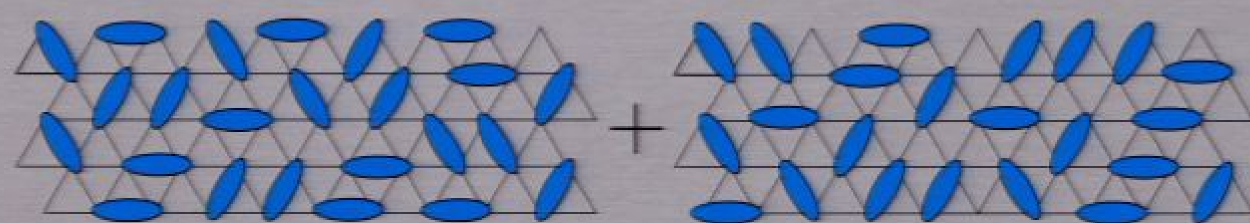
not a spin liquid

VB STATES

VBS



Short-range RVB

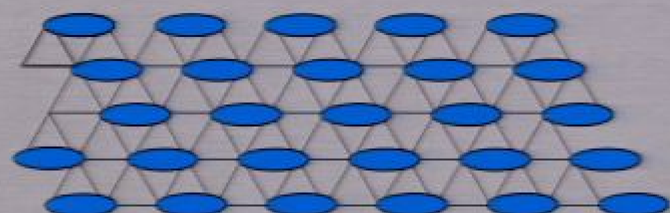


+ ...

a QSL with an energy gap to break a singlet

VB STATES

VBS



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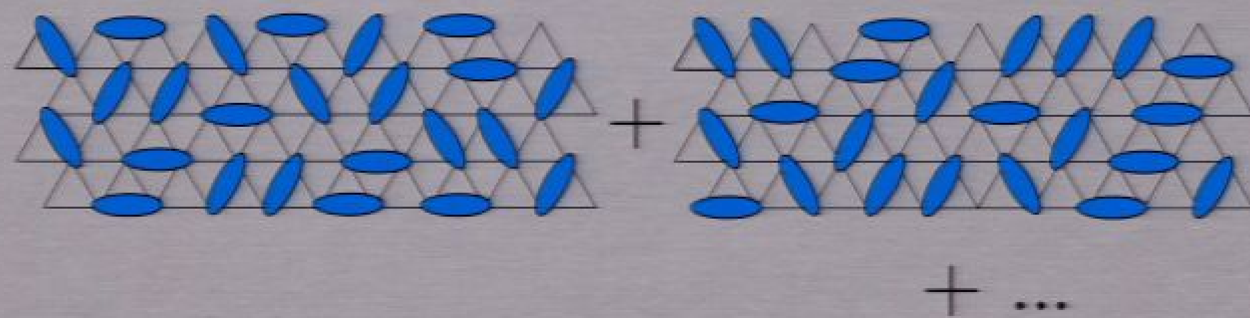
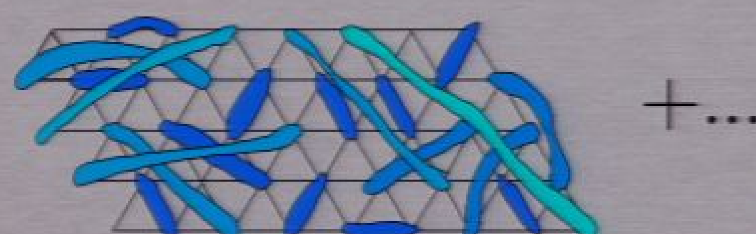


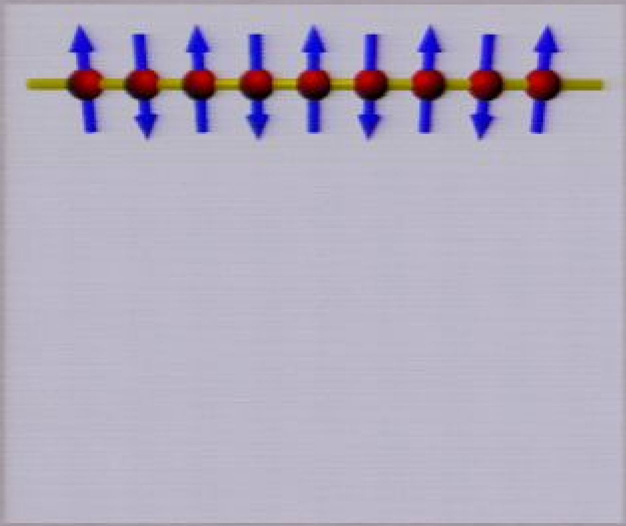
Long-range RVB



gapless spin excitations

Spinons

- QSLs generically support "spinons", neutral particles with S=1/2



- In 1d, the spinon is a domain wall or soliton
- It has in this sense a "string", but this does not confine the spinon because the string's boundary is just its endpoint

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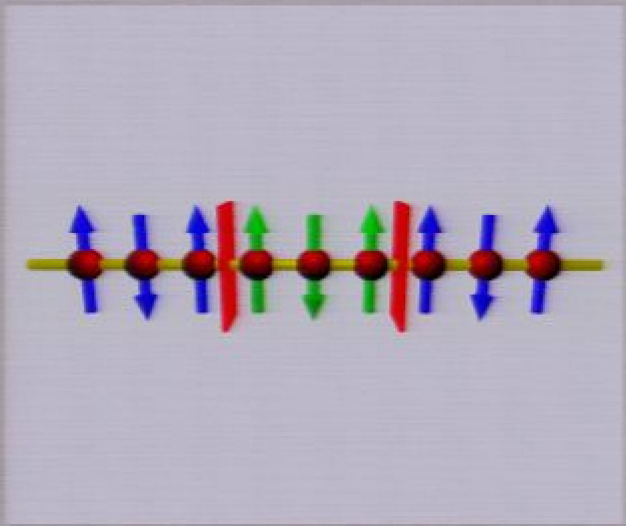

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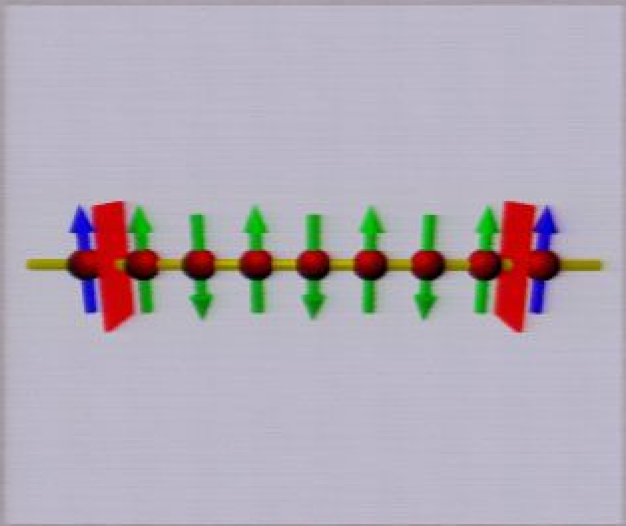

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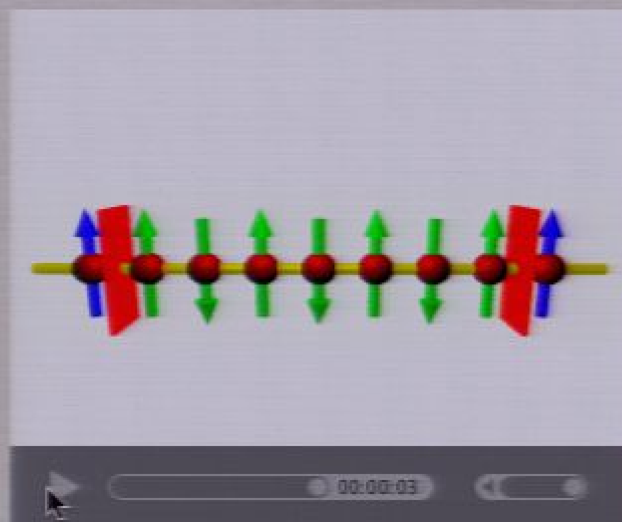

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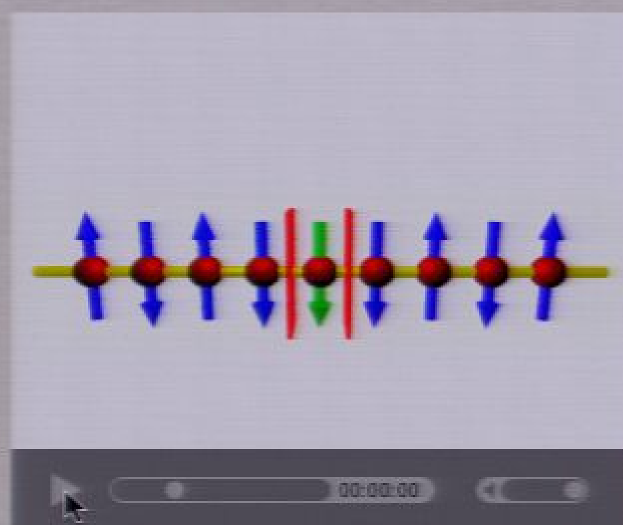

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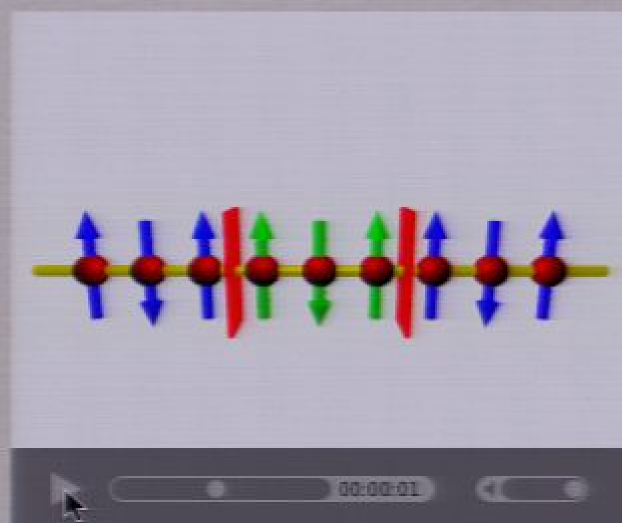

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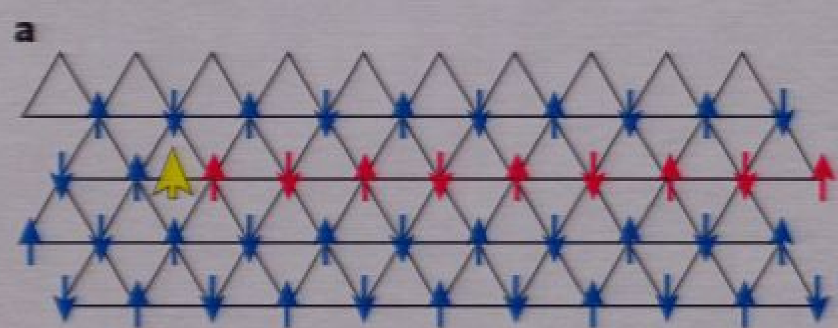
Spinons

- In d>1, any *observable* string costs divergent energy



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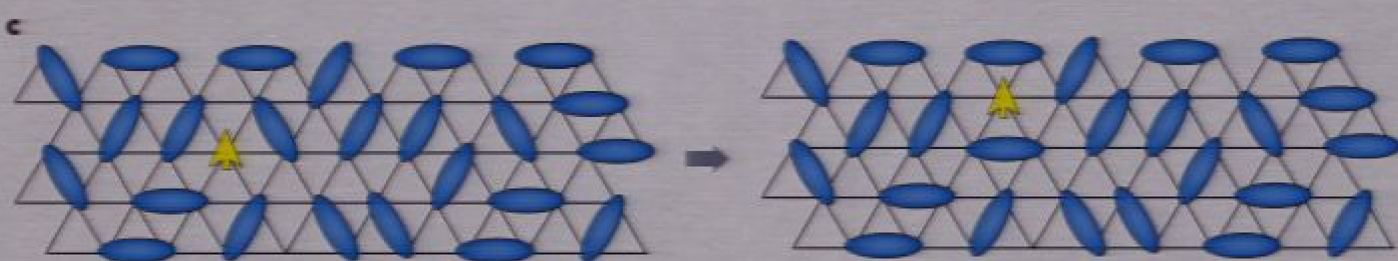


- If the ground state is a *superposition* of many states, the string need not be observable, if motion of the string simply reshuffles states in the superposition

Slave particles

- Gutzwiller-type variational wavefunction uses a reference Hamiltonian

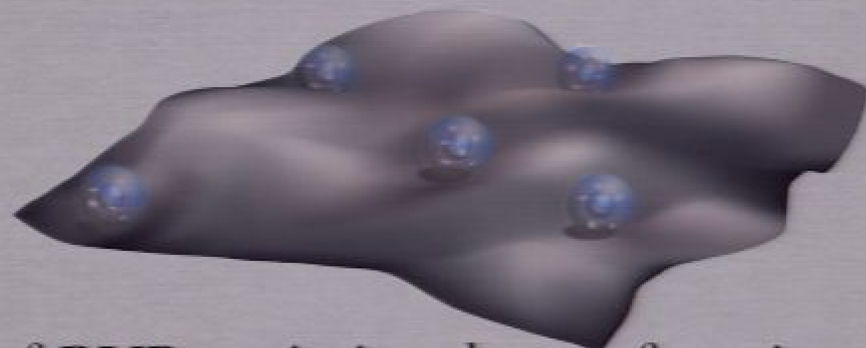
$$H_{ref} = \sum_{ij} \left[t_{ij} c^\dagger_{i\alpha} c_{j\alpha} + \text{h.c.} + \Delta_{ij} c^\dagger_{i\uparrow} c^\dagger_{j\downarrow} + \text{h.c.} \right]$$

- Project

$$|\Psi_{var}\rangle = \prod_i \hat{P}_{n_i=1} |\Psi_{ref}\rangle$$

- Gauge transformations of reference state leave physical state invariant
  - this is believed to be reflected in *emergent gauge fields* in the QSL phases: U(1), Z_2, ...

The "Landscape"



- The space of RVB variational wavefunctions is vast
- The number of distinct Quantum Spin Liquid (QSL) *phases* is also huge
  - e.g. X.G. Wen has classified *hundreds* of different QSL states all with the same symmetry on the square lattice (and this is *not* a complete list!)
  - This makes it difficult to compare all of the states
- Many QSLs are described by non-trivial interacting QFTs, which are themselves not well understood

Outline

- Liquids and spin liquids
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- Quantum spin liquids
- **How we look for them and what we've found so far**

The paradox

- There seem to be so many QSLs in theory
- But *no* clear demonstrations in experiment
  - probably thousands of quantum antiferromagnets have been studied experimentally and nearly all of them order magnetically

Where to look?

- Materials with
  - S=1/2 spins
  - Frustration
  - Other sources of fluctuations, e.g. proximity to Mott transition (where the electrons become delocalized)

What to look for

- $1/f = T_c = 0$: no ordering (magnetic or otherwise!)
- No spin freezing (hysteresis, NMR, μSR)
- Structure of low energy excitations
  - $\chi(T)$, $C_v(T)$, $1/T_1$, $\varkappa$, inelastic neutrons
  - theoretical guidance helpful!
- Smoking gun?

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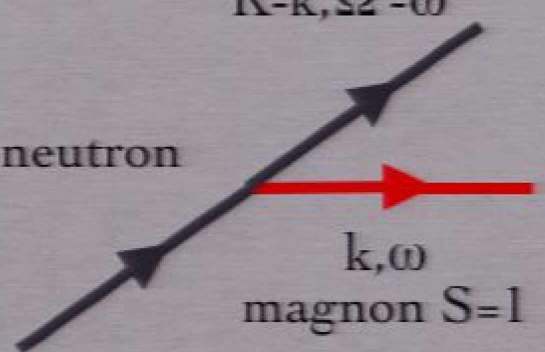
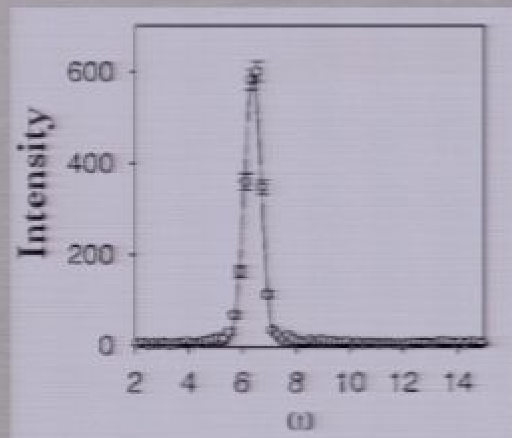
Seeing Spinons

- A proof of principle: 1d spinons have been observed in several materials by neutron scattering
- Basic idea



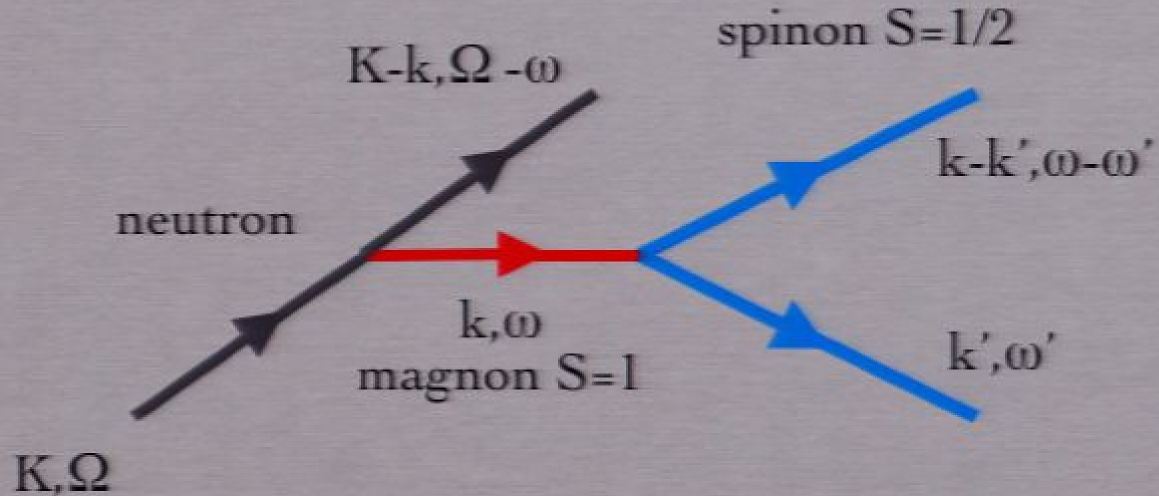


Line shape in Rb_2MnF_4

leads to a sharp peak at $\omega = \varepsilon(k)$

Seeing Spinons

- A proof of principle: 1d spinons have been observed in several materials by neutron scattering
- Basic idea



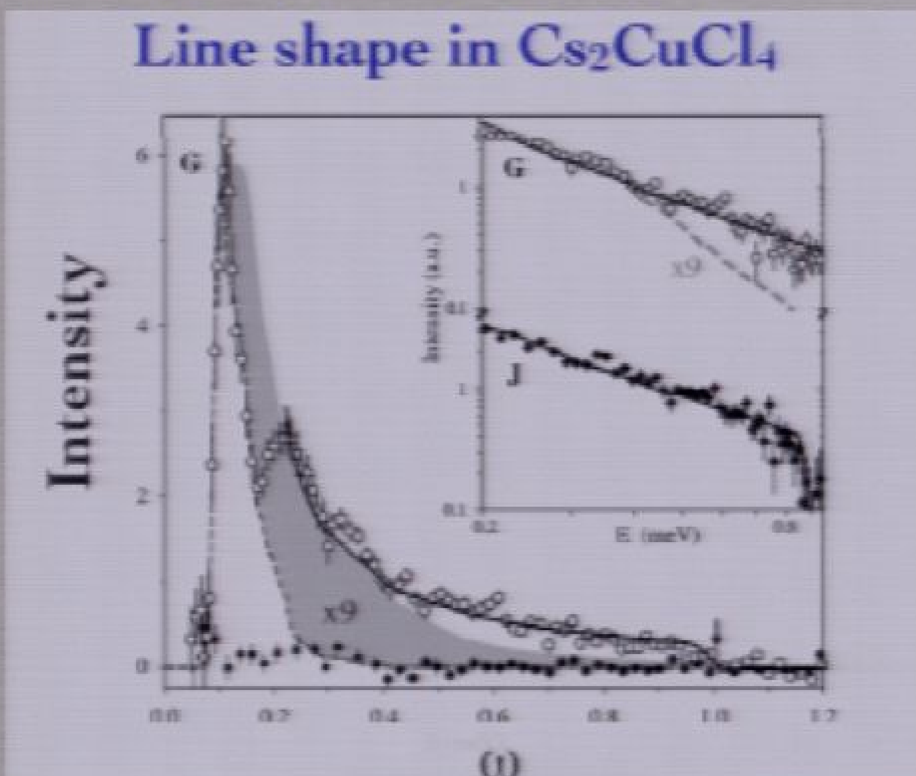
broad peak with $\omega=\varepsilon(k')+\varepsilon(k-k')$

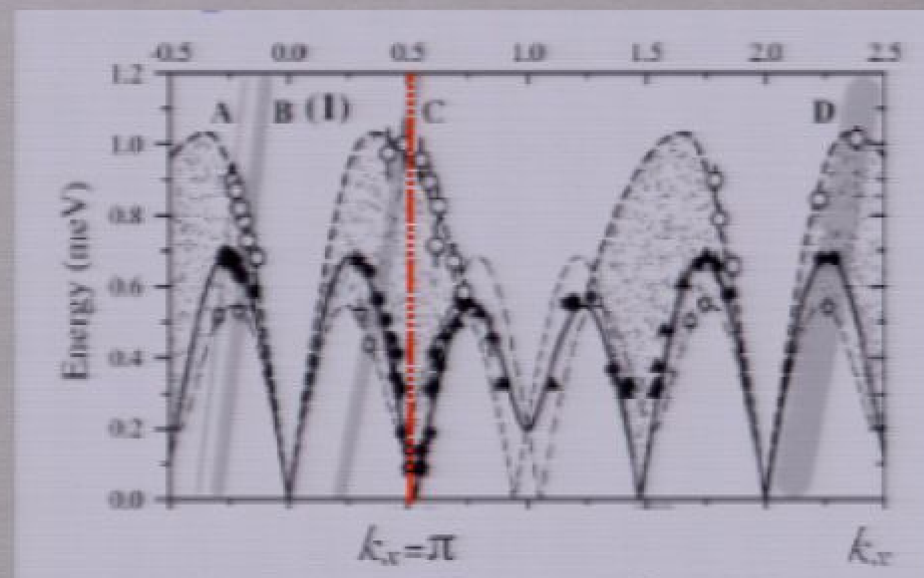
Cs_2CuCl_4

Oleg Starykh

Masanori Kohno

- "Power law" fits well to free spinon result
  - Fit determines normalization





R. Coldea

Oleg Starykh

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Masanori Kohno

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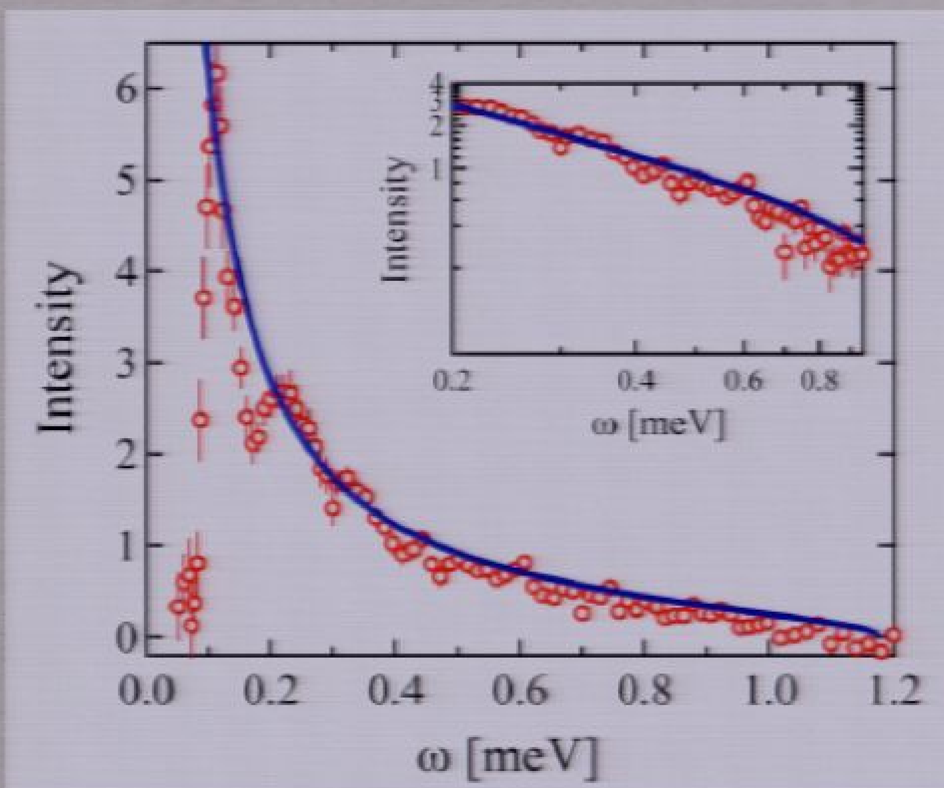
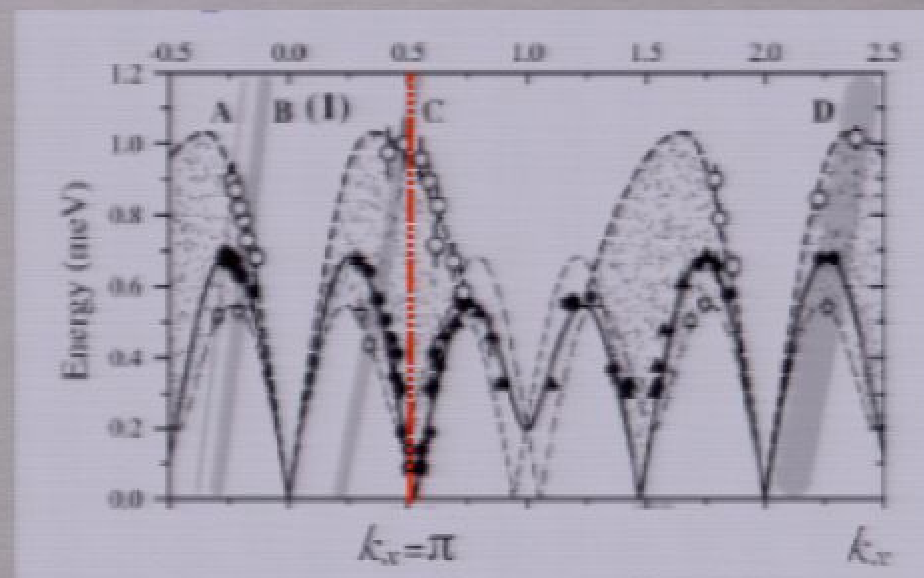
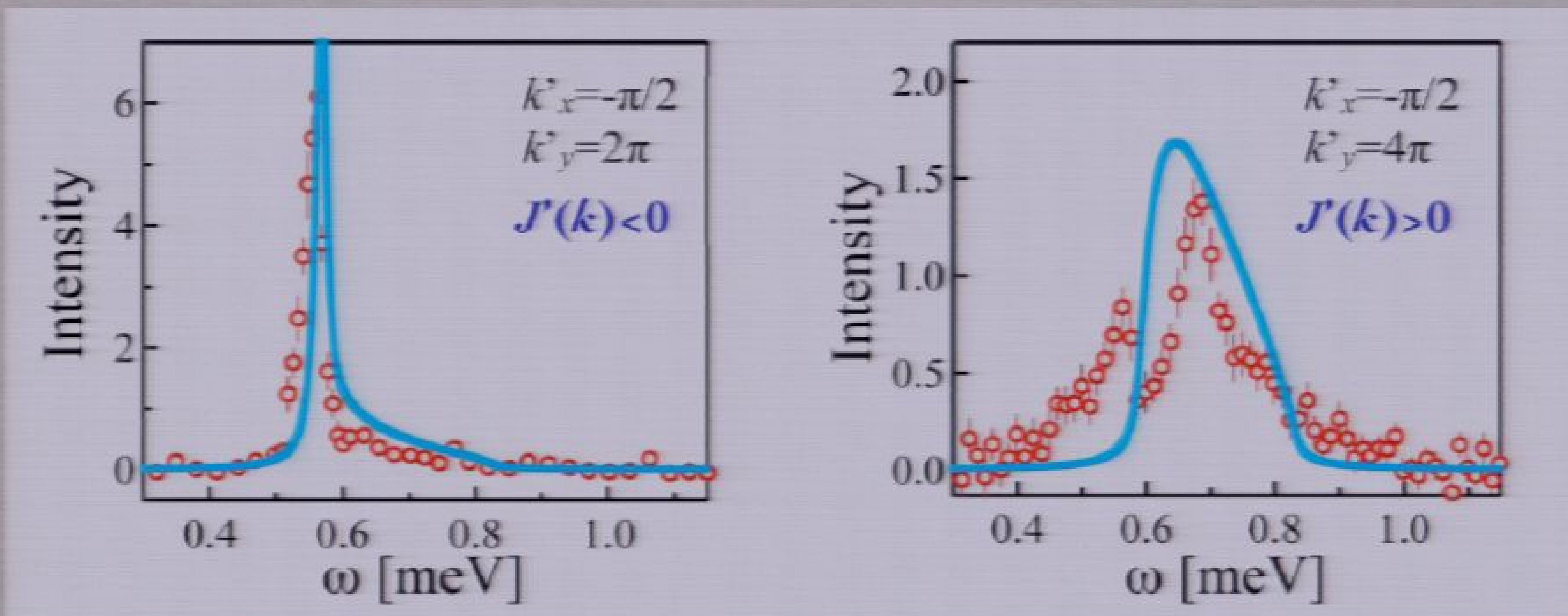




R. Coldea

Spinon interactions

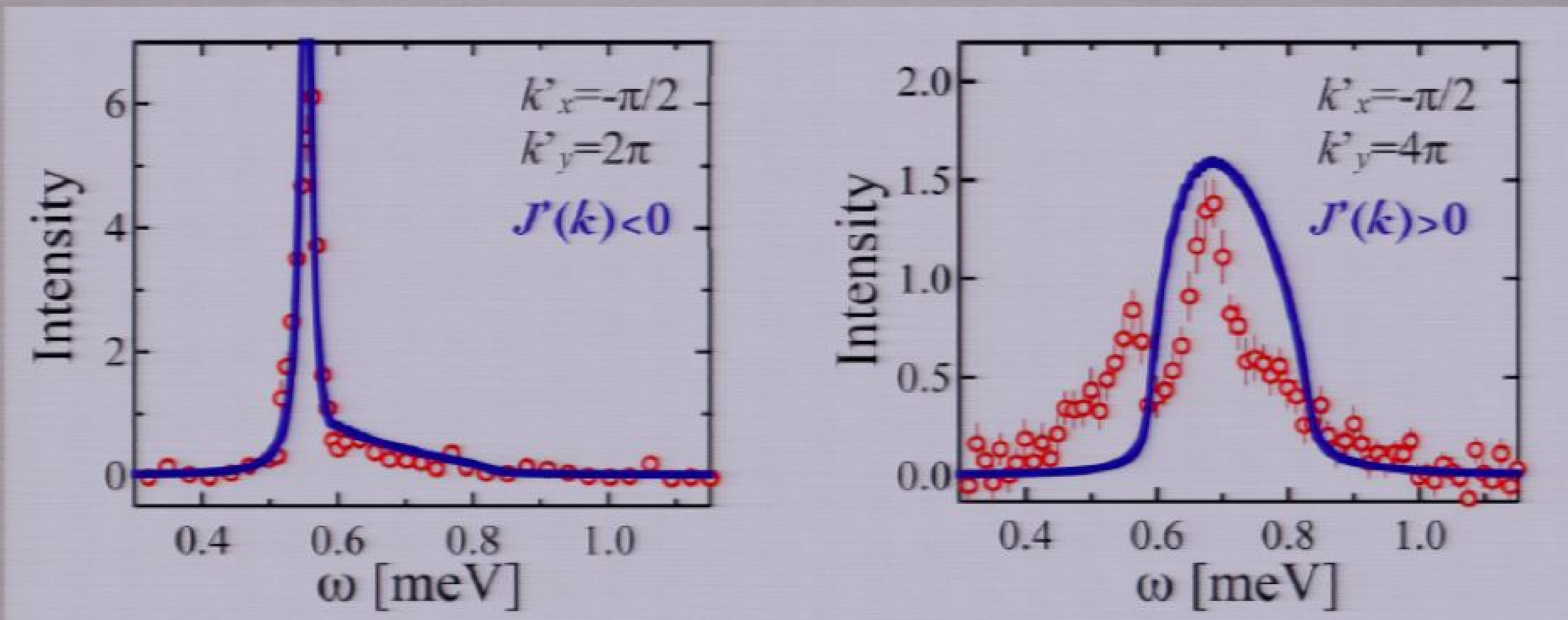
- Depending upon k, spinons may be bound or not



- Curves: 2-spinon theory w/ experimental resolution

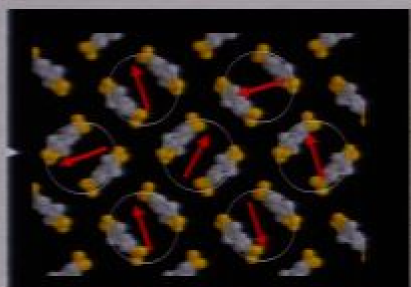
Spinon interactions

- Depending upon k, spinons may be bound or not



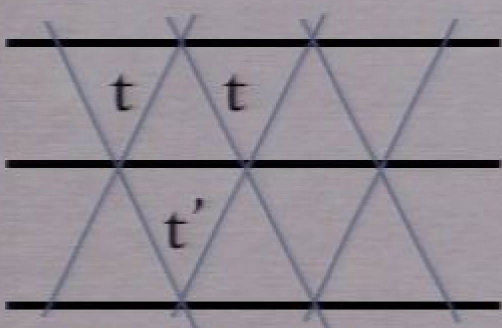
- Curves: 4-spinon RPA w/ experimental resolution

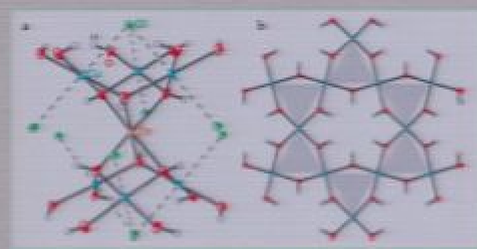
D>1 QSL MATERIALS



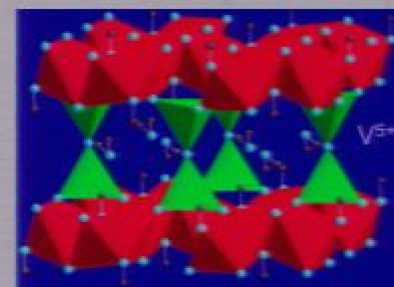
$\varkappa$-(BEDTTTF)$_2$Cu$_2$(CN)$_3$

EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

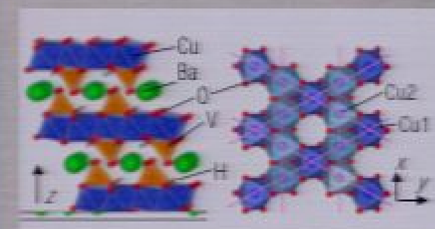




herbertsmithite



volborthite



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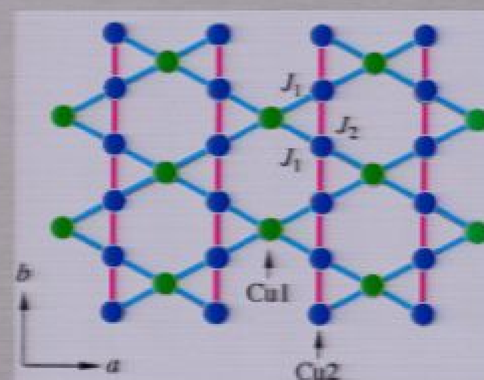
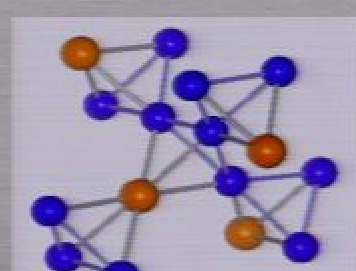
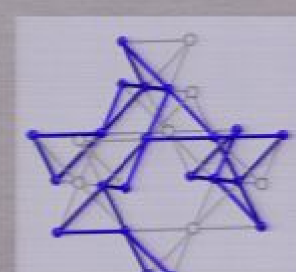
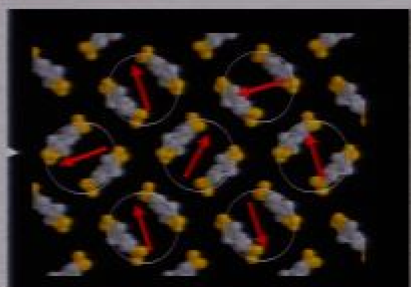






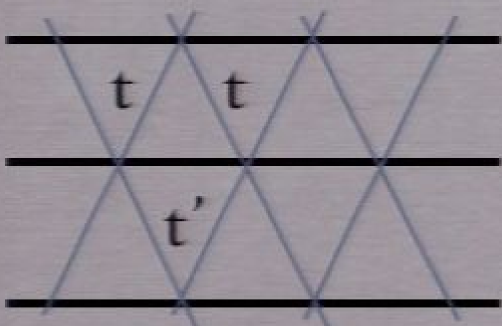
Na$_4$Ir$_3$O$_8$

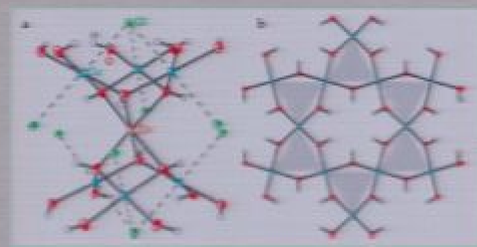
D>1 QSL MATERIALS



κ-$(BEDTTTF)_2Cu_2(CN)_3$

$EtMe_3Sb[Pd(dmit)_2]_2$





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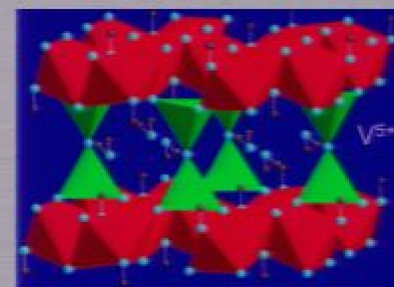

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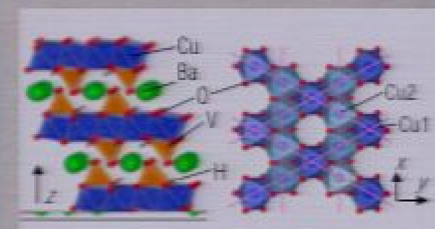

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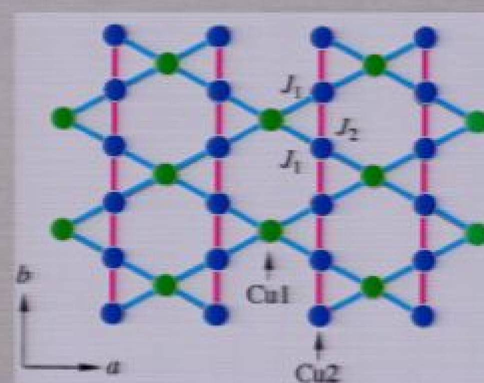
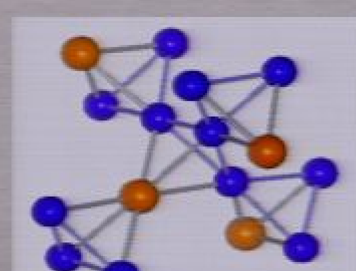
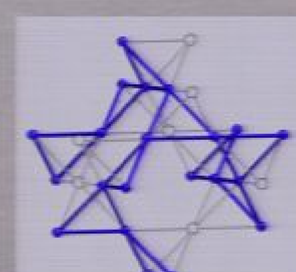


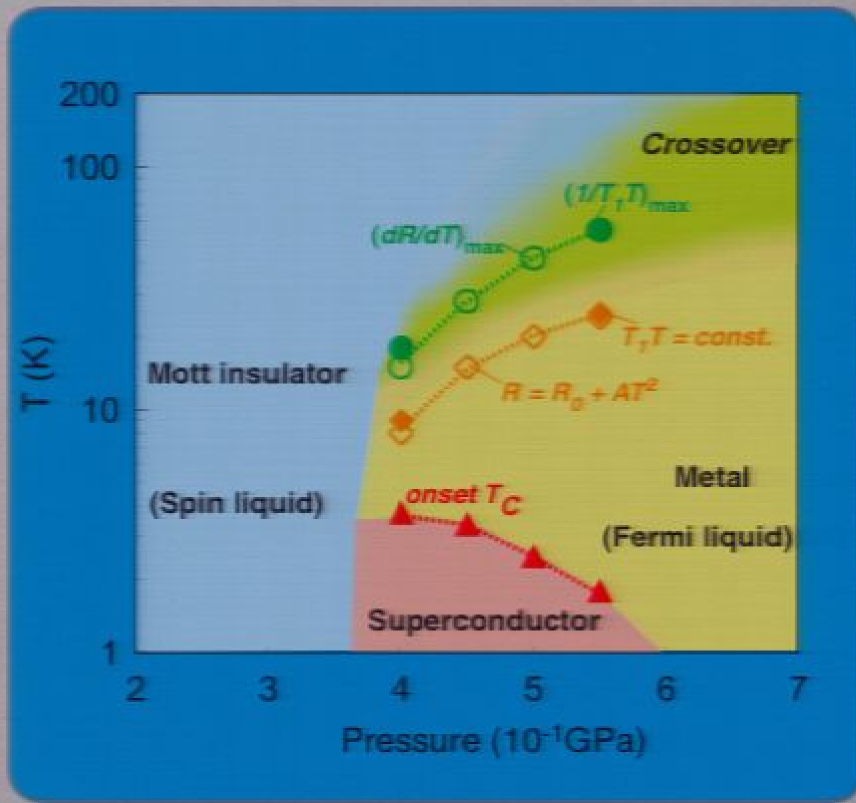




$Na_4Ir_3O_8$

Organics

- Materials are proximate to a Mott transition
  - "weak" insulators
- Charge fluctuations are expected to make QSL behavior more likely



κ-(BED-TTTF)$_2$Cu$_2$(CN)$_3$

K. Kanoda group (2003-)

Theory: Organics

- RVB/QSL state:
  - Motrunich, Lee+Lee: (2005) "uniform RVB"
  - this is a kind of RVB state with very many (maybe a maximal number of?) long-range VBs
  - It is described by a **"Fermi sea" of spinons** coupled to a U(1) gauge field
- Good variational energy for triangular lattice Hubbard model
- How does it fit with experiments?

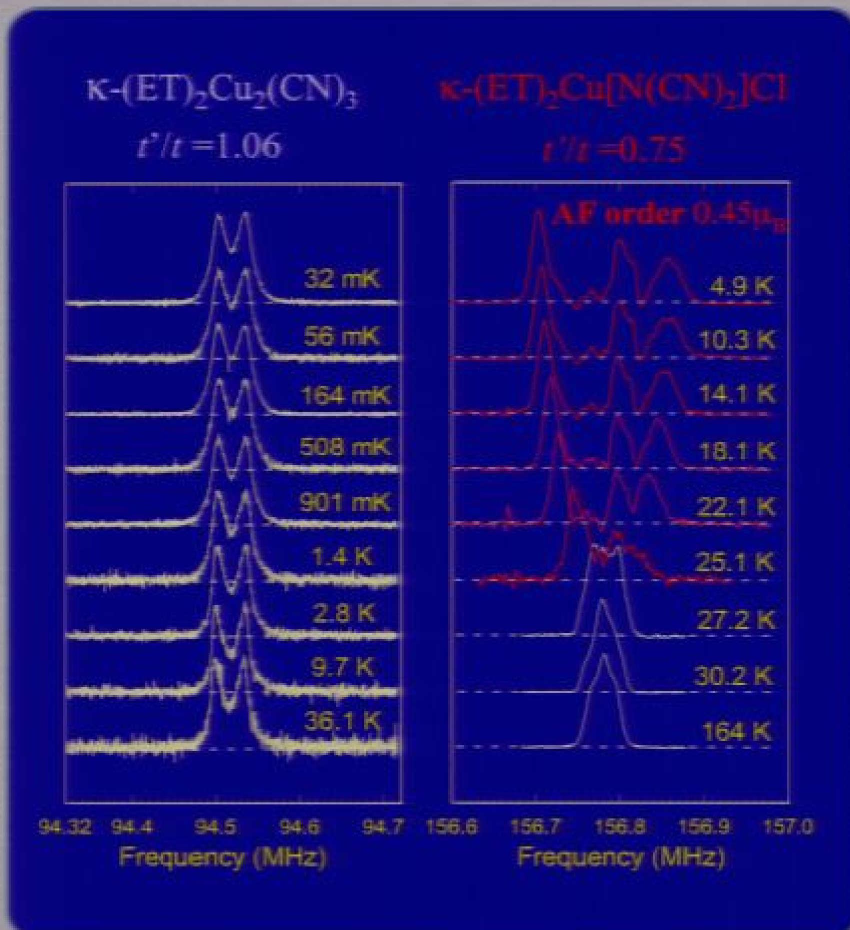
Circumstantial Evidence

- No ordering ✔
- Large T=0 susceptibility ✔
- Power-law $1/T_1$ ✔
    - but it's not clear the actual power works

NMR LINESHAPES

^{1}H NMR

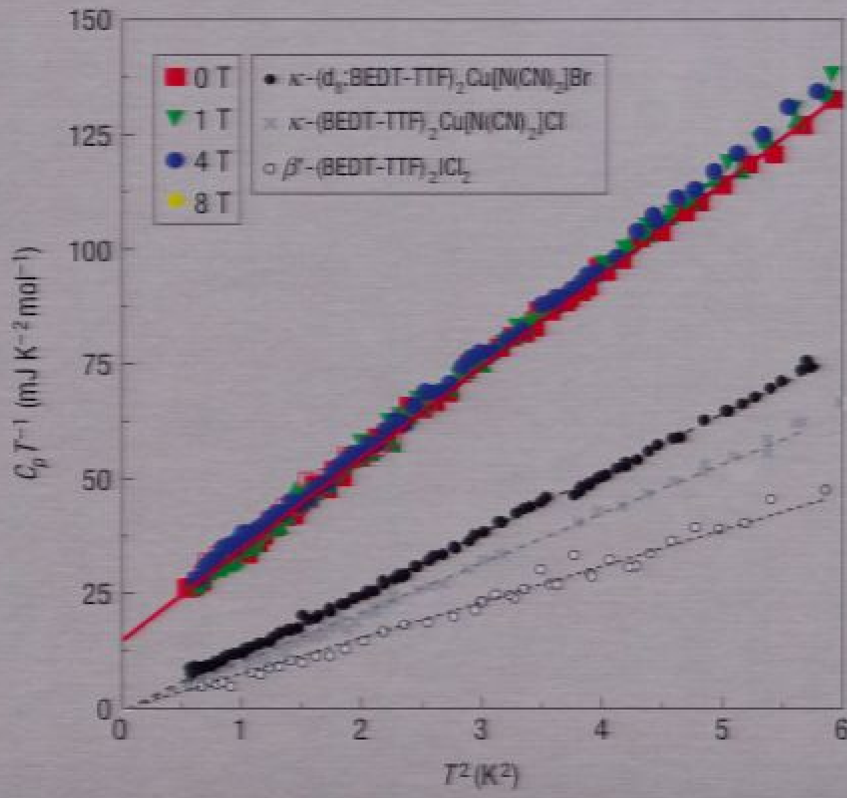
Y. Shimizu *et al*, 2003



Evidence for lack of static moments: f> 1000!

Specific Heat

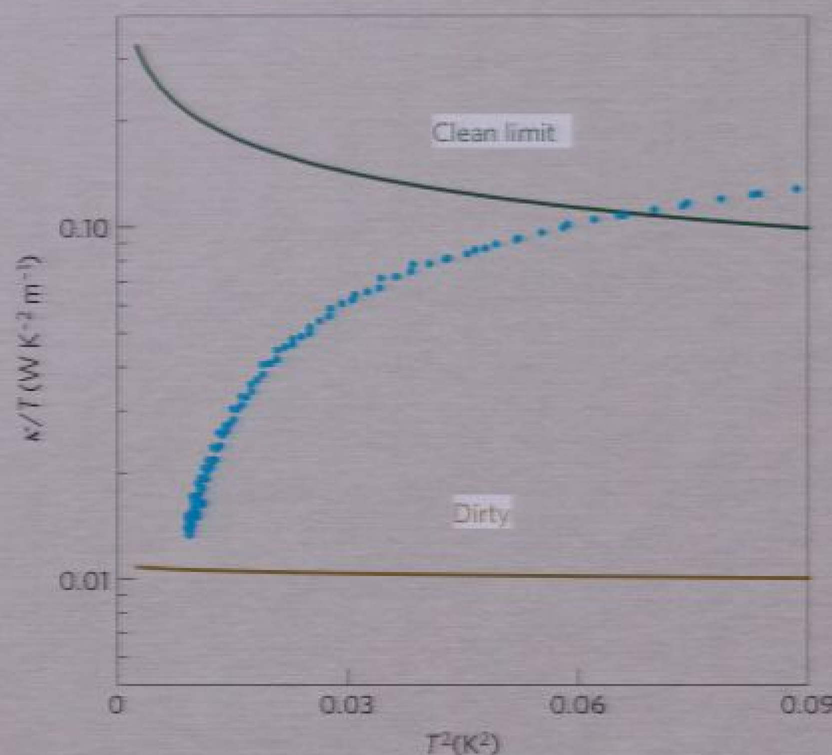
- Linear specific heat as expected for *free fermions* - approximately as expected for QSL - but very strange for an insulator



S. Yamashita *et al*, 2008

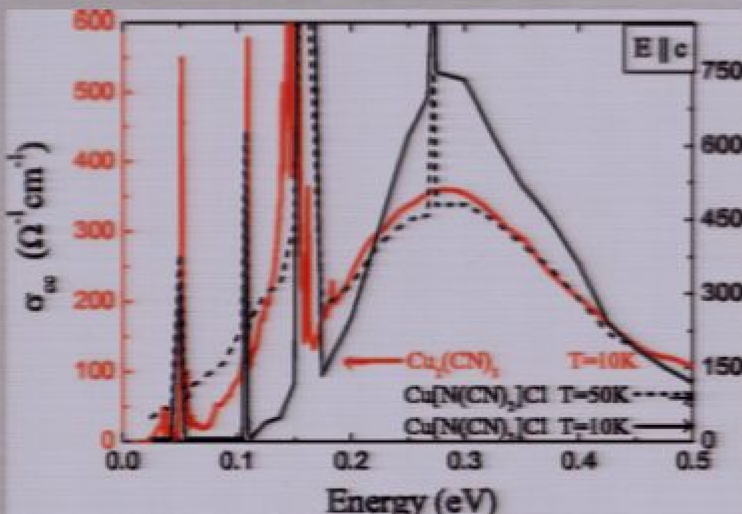
Challenges

- Thermal conductivity shows a gap
- Not consistent with uniform RVB state
- Consistency with specific heat?
- Different behavior seen for other organic very recently

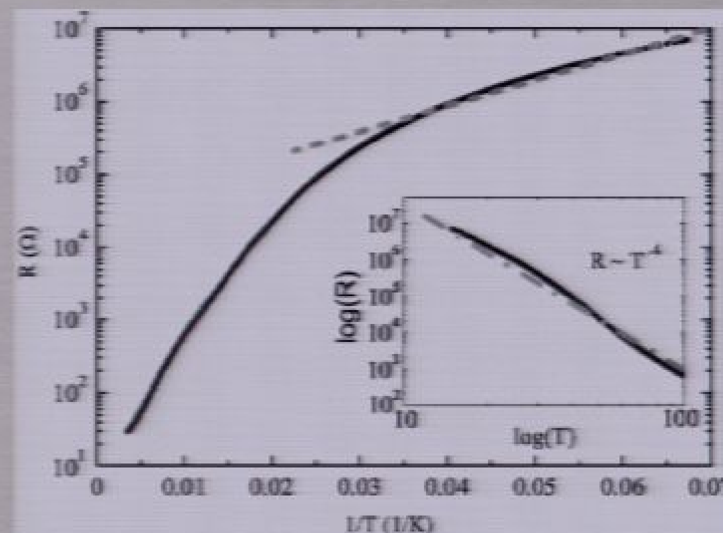


M. Yamashita et al, 2008

Challenges



very small or no optical gap (pseudogap)

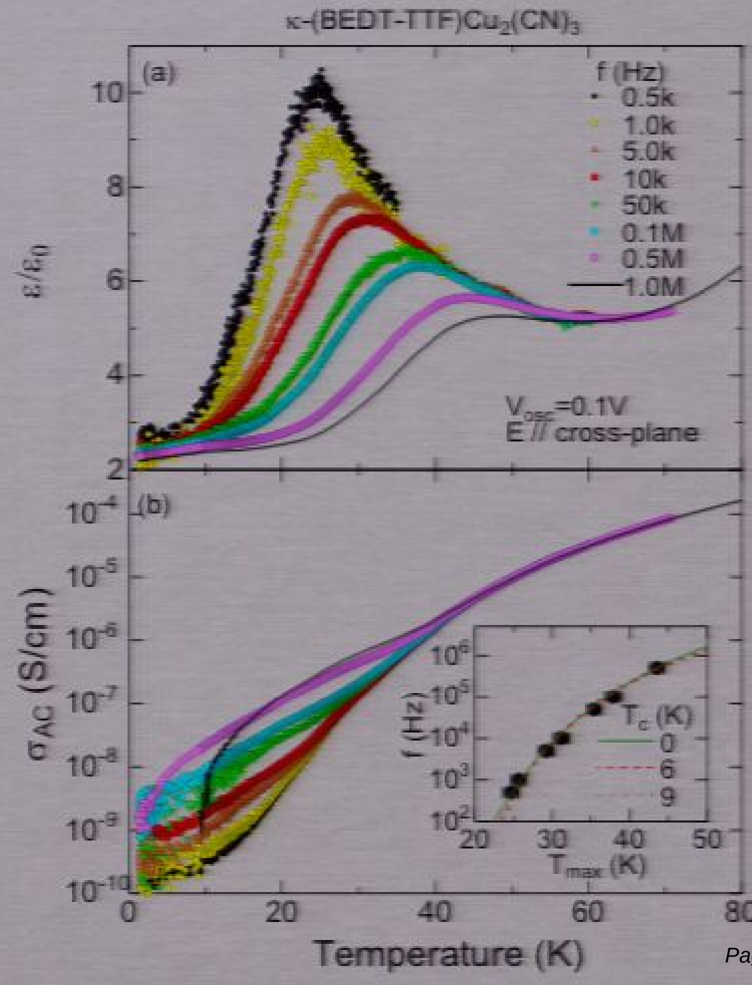


non-activated resistivity (or gap <15 meV)

Kézsmárki *et al*, 2006

Challenges

- Dramatic dielectric anomalies observed at T<60K
- Points to molecular dipoles in individual organic "dimers" - not taken into account by RVB theory



M. Abdel-Jawad *et al.* 2010

The Smoking Gun

- Can we devise an experiment which convincingly shows the presence of exotic excitations directly?
  - maybe inelastic single crystal neutrons - they do see spinons in 1d
  - the "Senthil experiment" to see *visons* (cannot be done on most materials)
  - Can you see $2k_F$ oscillations somehow in a Mott insulator?
  - something more clever?

Conclusions

- Frustrated magnets provide a rich variety of phenomena including a number of promising new quantum spin liquid candidates

- For QSLs, what is needed is a combined effort of innovative experimental and theoretical work, with attention of the latter paid to the former!