Local and Short-Range Magnetic Excitations

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Outline

Origins of unconventional magnetism

- antiferromagnetic interactions, low spin value, low dimensional
- Example spin-1/2 dimer antiferromagnet
- Example spin-1/2, antiferromagnetic chain

Origins of frustrated magnetism

- geometric frustration, competing interactions and anisotropy

Examples of frustrated magnets

- 2-Dimensional magnets e.g. Square, triangular, kagome, lattice
- 3-Dimensional magnets e.g. pyrochlore, spin ice and water ice

Unconventional Magnets

The Origins of Unconventional Magnetism

Quantum fluctuations suppress long-range magnetic order, spin-wave theory fails

- Quantum effects are most visible in magnets with
  - low spin values
  - antiferromagnetic exchange interactions
  - low-dimensional interactions
- Quantum effects give rise to exotic states and excitations

$$H = \sum_{n,m \neq n} H_{n,m}$$

$$H_{n,m} = J_{n,m} \mathbf{S}_n \mathbf{S}_m = J_{n,m} \left(S_n^x S_m^x + S_n^y S_m^y + S_n^z S_m^z \right)$$

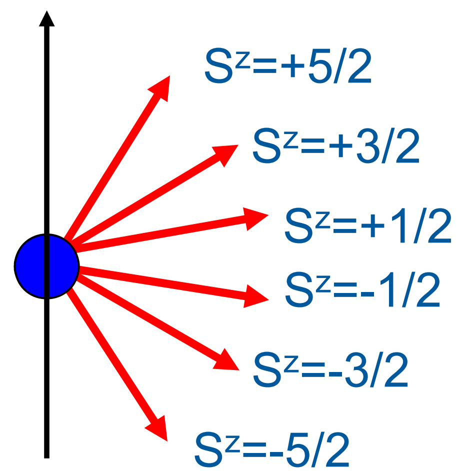
$$H_{n,m} = \boxed{J_{n,m} S_n^z S_m^z} + \boxed{J \left(S_n^+ S_m^- + S_n^- S_m^+ \right)}$$

Quantum Magnetism - Low Spin Value

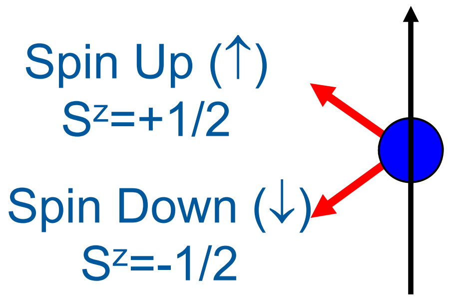
$$H_{n,m} = J_{n,m} S_n^z S_m^z + J\left(S_n^+ S_m^- + S_n^- S_m^+\right)$$

- Fluctuations have the largest effect for low spin values
- For S=1/2, changing S^z by 1 unit reverses the spin direction

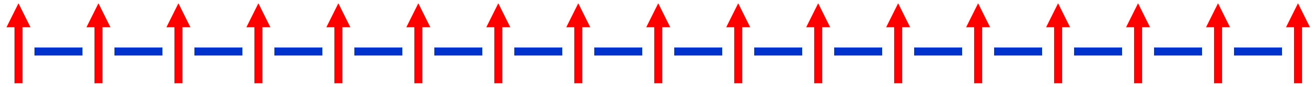
S=5/2
|S|^2=S(S+1)=35/4



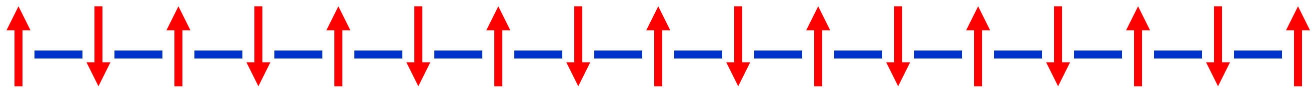
S=1/2
|S|^2=S(S+1)=3/4



Antiferromagnetic Exchange Interactions

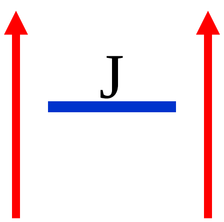


- Parallel spin alignment is an eigenstate of the Hamiltonian



- Antiparallel spin alignment (Néel state) is not an eigenstate of the Hamiltonian

J>0
ferromagnetic



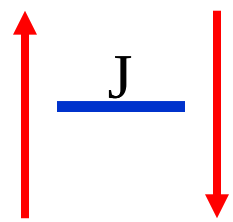
$S_1=1/2$ $S_2=1/2$
$S^z=1/2$ $S^z=1/2$

$$H_{1,2} = J\left(S_1^+ S_2^- + S_1^- S_2^+ + S_1^z S_2^z\right)$$

$$H_{1,2}\left|\uparrow_1\uparrow_2\right\rangle = J/4\left|\uparrow_1\uparrow_2\right\rangle$$

$$H_{1,2}\left|\uparrow_1\downarrow_2\right\rangle = -J/4\left|\uparrow_1\downarrow_2\right\rangle + J/4\left|\downarrow_1\uparrow_2\right\rangle$$

J>0
antiferromagnetic



$S_1=1/2$ $S_2=1/2$
$S^z=1/2$ $S^z=-1/2$

Low-Dimensional Interactions

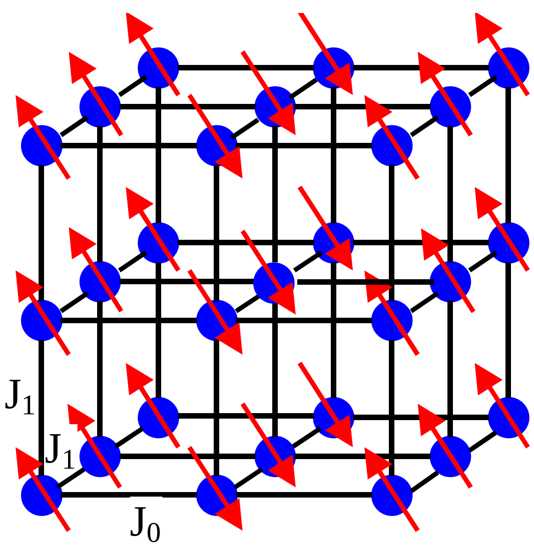
For 3-dimensional magnets each magnetic ion has six neighbours

For a 1-dimensional magnet there are only two neighbours

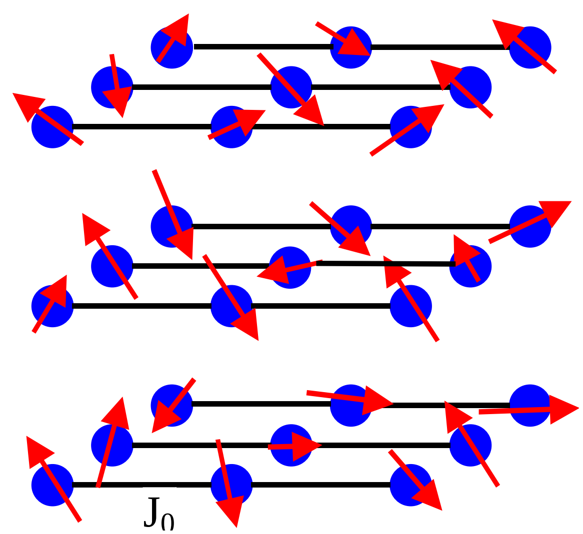
Neighbouring ions stabilize long-range order and reduce fluctuations

$$H_{n,m} = \boxed{J_{n,m} S_n^z S_m^z} + \boxed{J\left(S_n^+ S_m^- + S_n^- S_m^+\right)}$$

3D S=1/2



1D S=1/2



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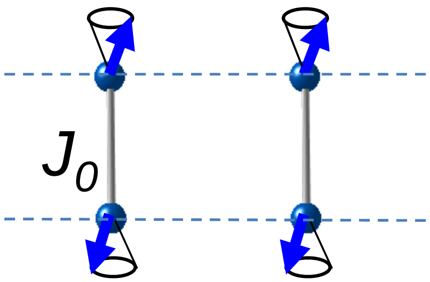
Zero Dimensional Quantum Magnets

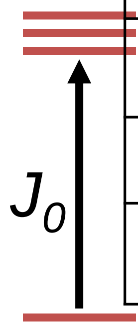
0-Dimensions - Spin-1/2, Dimer Antiferromagnets

Dimer Unit
S=1/2, $S^z = \pm 1/2$

$$H = J_0 \boldsymbol{S}_a . \boldsymbol{S}_b$$



Excitations

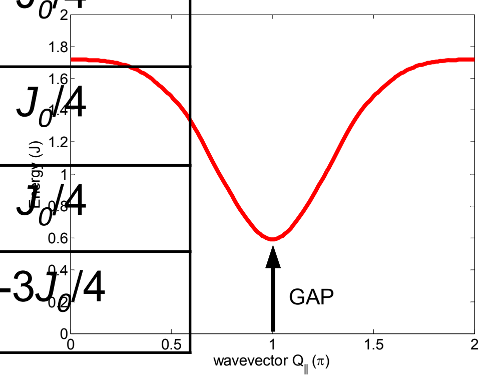


Eigenstates	S_T	S^z	Eigenvalue
$\vert 1\rangle = \vert\uparrow_1\uparrow_2\rangle$	1	1	$J_0/4$
$\vert 0\rangle = \frac{1}{\sqrt{2}}(\vert\uparrow_1\downarrow_2\rangle + \vert\downarrow_1\uparrow_2\rangle)$	1	0	$J_0/4$
$\vert -1\rangle = \vert\downarrow_1\downarrow_2\rangle$	1	-1	$J_0/4$
$\vert G\rangle = \frac{1}{\sqrt{2}}(\vert\uparrow_1\downarrow_2\rangle - \vert\downarrow_1\uparrow_2\rangle)$	0	0	$-3J_0/4$

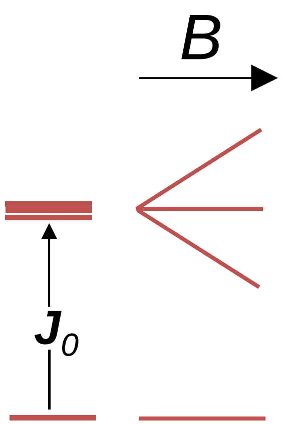
Triplet, S=1

Singlet, S=0

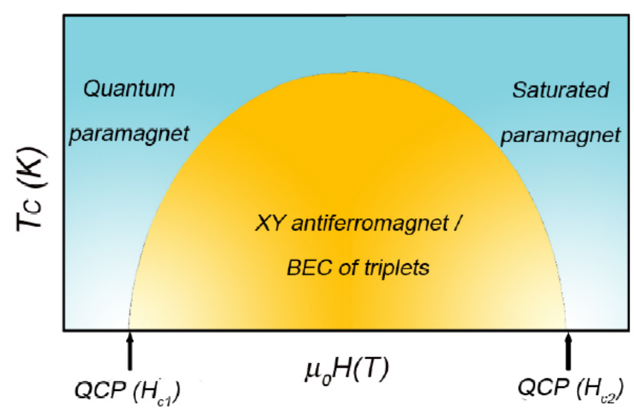
Interdimer coupling, gapped dispersive Mode



Zeeman Splitting in Field



Bose Einstein Condensation



Properties:

- Singlet ground state.
- Gapped 1-magnon mode
- 2-magnon continuum
- Bound modes.
- Bose Einstein condensation

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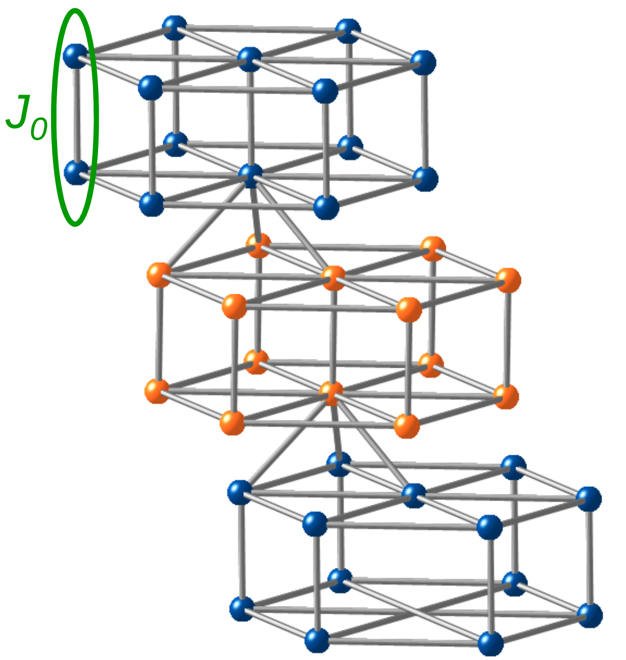
$Sr_3Cr_2O_8$ –Spin-1/2, Dimer AF

$Sr_3Cr_2O_8$ → Cr^{5+}, Spin-1/2.

$Sr_3Cr_2O_8$ is 3D network of dimers



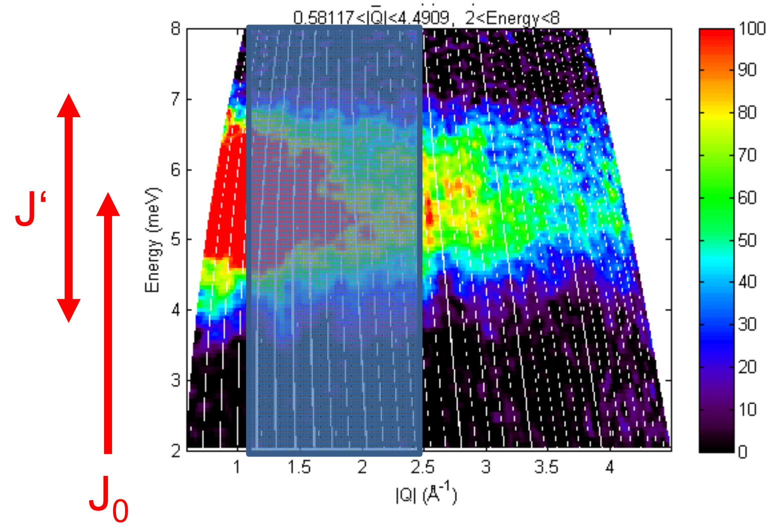
Dimer coupling is bilayer J_0

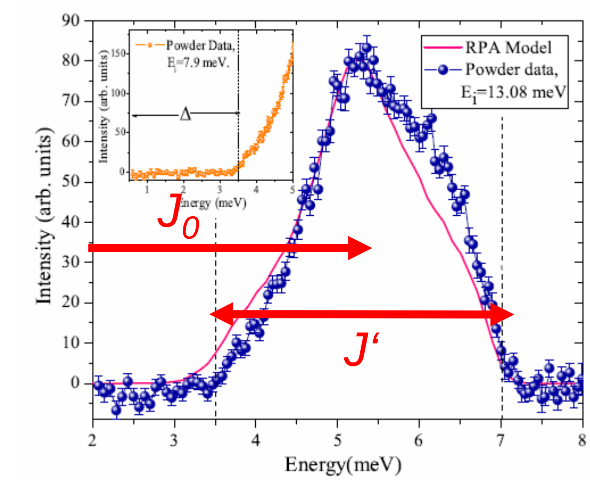
E_{gap}=3.4meV
E_{upper}=7.10meV

$E_{midband}$~J_0=5.5meV
$E_{bandwidth}$~J'=3.7meV

Powder inelastic neutron scattering NEAT, HZB

D.L. Quintero-Castro, et al
Phys. Rev. B. 81, 014415 (2010)





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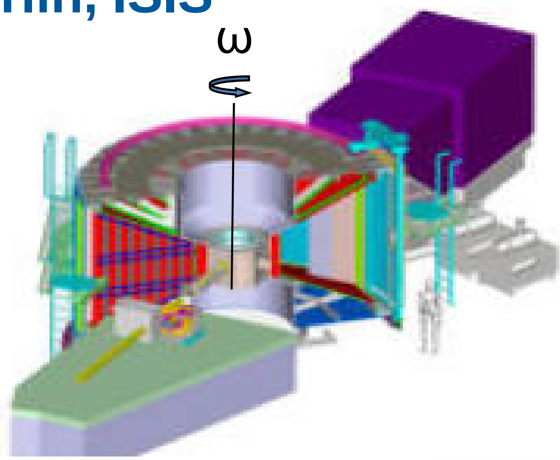
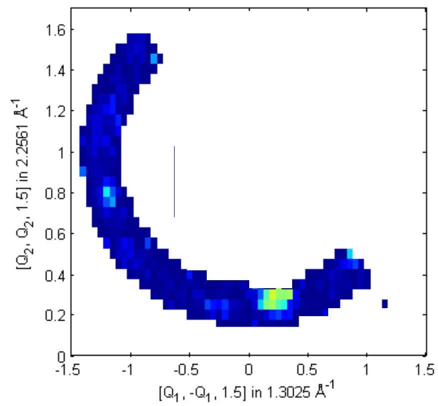
Single Crystal Inelastic Neutron Scattering

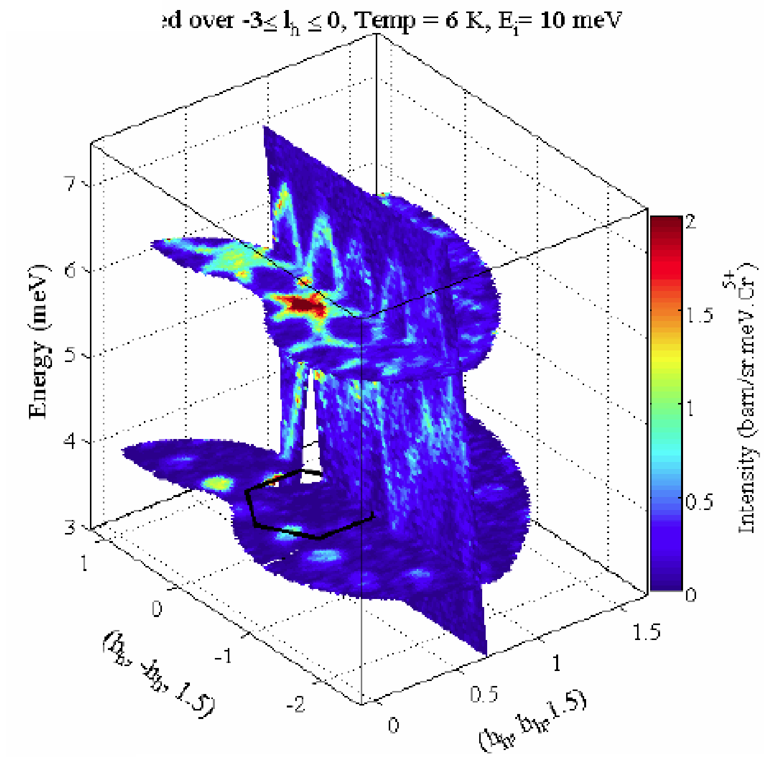
Merlin, ISIS



detectors:
180° horizontal
±30° vertical

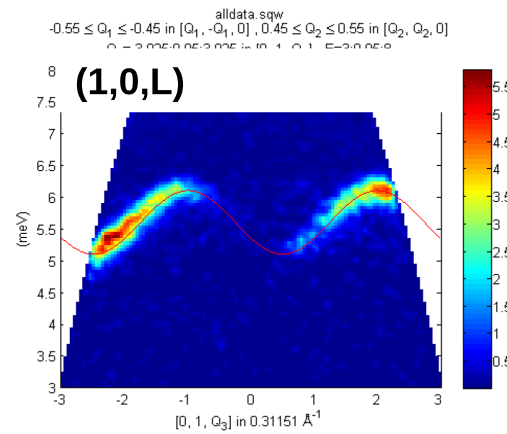
ω scans,
Range 70°
step=1°
2 hours per step.

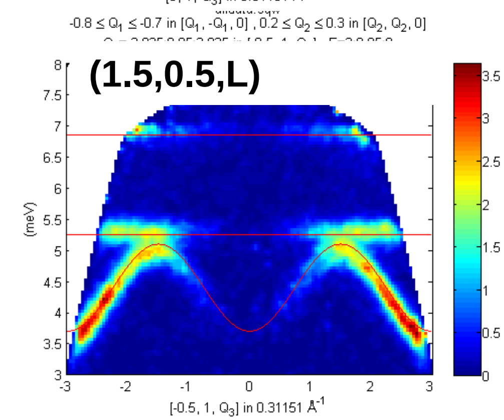




Individual scans combined to create a single file $S(Q_h, Q_k, Q_l, E)$.

large region of the energy and reciprocal space.





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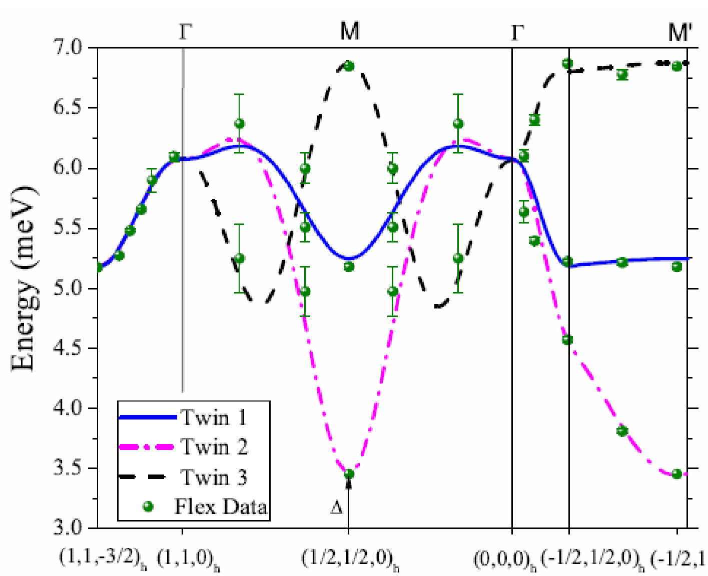
D.L. Quintero-Castro, et al Phy. Rev. B. 81, 014415 (2010)

Fitting to a Random Phase Approximation

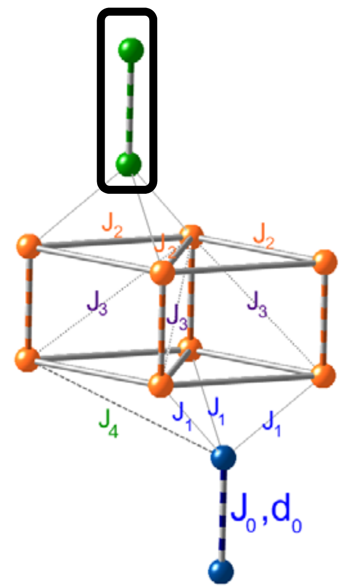
Extracted Dispersions



Random Phase Approximation

M. Kofu et al Phys. Rev. Lett. 102 037206 (2009)

$$\hbar\omega \cong \sqrt{J_0^2 + J_0\gamma(\mathbf{Q})} \qquad \gamma(\mathbf{Q}) = \sum_i J(\mathbf{R_i})e^{-i\mathbf{Q}\cdot\mathbf{R_i}}$$



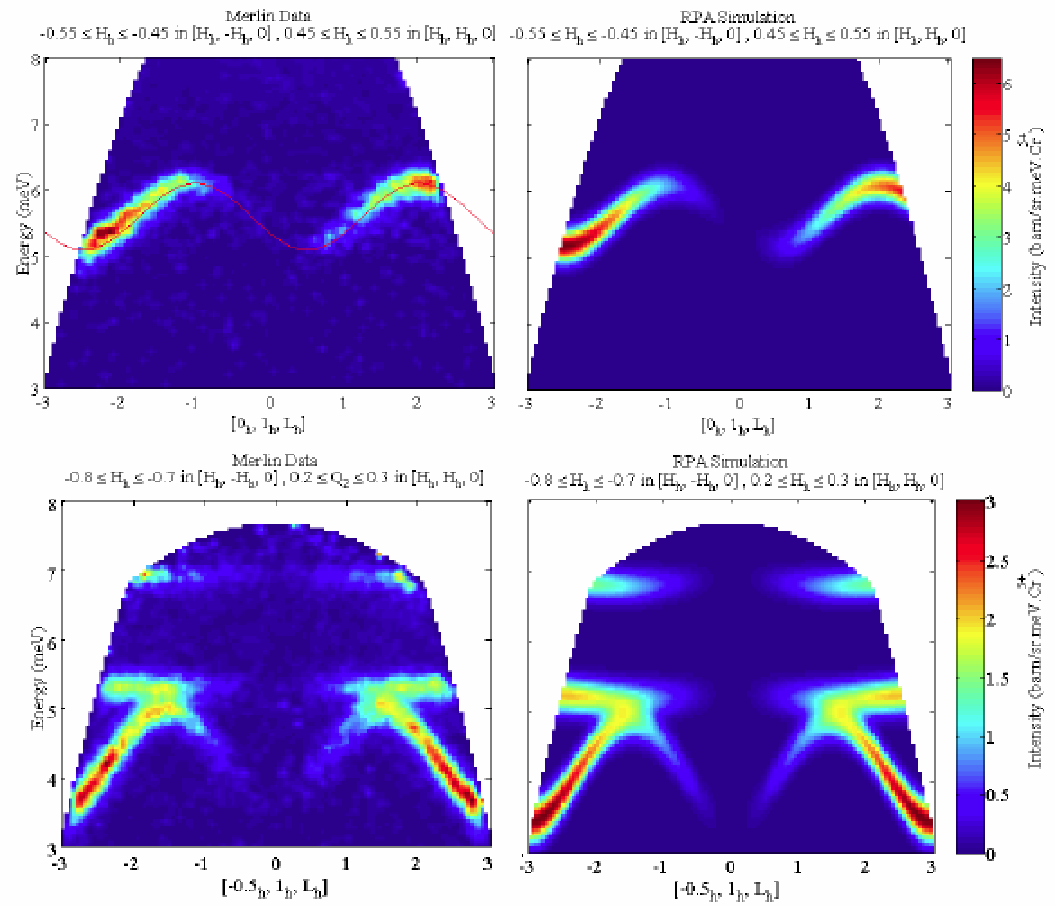
Constants	$Sr_3Cr_2O_8$
J_0	5.551(9)
J_1'	-0.04(1)
J_1''	0.24(1)
J_1'''	0.25(1)
$J_2' - J_3'$	0.751(9)
$J_2'' - J_3''$	-0.543(9)
$J_2''' - J_3'''$	-0.120(9)
J_4'	0.10(2)
J_4''	-0.05(1)
J_4'''	0.04(1)
J' =	$J' = 3.6(1)$
J'/J_0	$J'/J_0 = 0.6455$

Simulation and Data

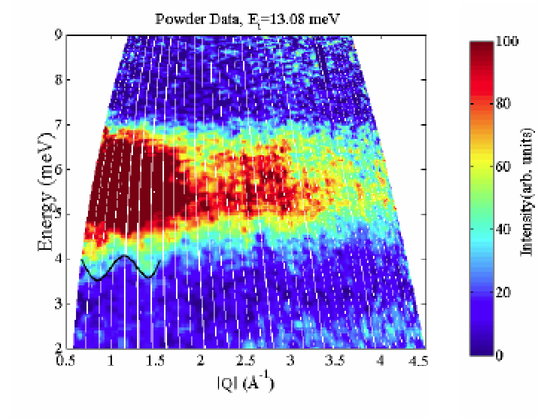
Neutron cross-section

$$\frac{d^2\sigma(\mathbf{Q},E)}{d\Omega dE} \approx \frac{|f_{Cr^{5+}}(|Q|)|^2\,(1-cos(\frac{2\pi\ell_h d_0}{c_h}))e^{(-(E-\hbar\omega)^2/\Delta E^2)}}{\hbar\omega(1-e^{E/k_BT})}$$

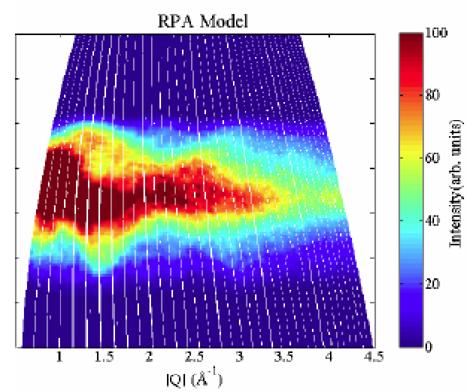
Data – single crystal simulation – single crystal



Data - Powder average:



Simulation - Powder average:

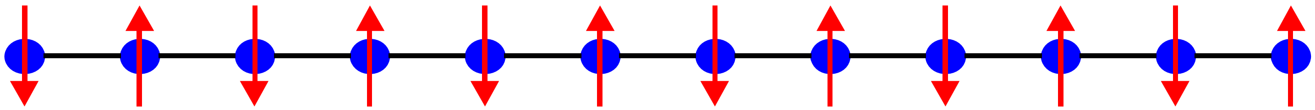


D. L. Quintero-Castro, B. Lake, E.M. Wheeler
Phy. Rev. B. 81, 014415 (2010)

Simulation of the TOF data with the fitted values interactions

One Dimensional Quantum Magnets

1D, S-½, Heisenberg, Antiferromagnet



$$H = J\sum_i \vec{S}_i \cdot \vec{S}_{i+1}$$

Bethe Ansatz

Hans Bethe
Bethe Ansatz
(1931)

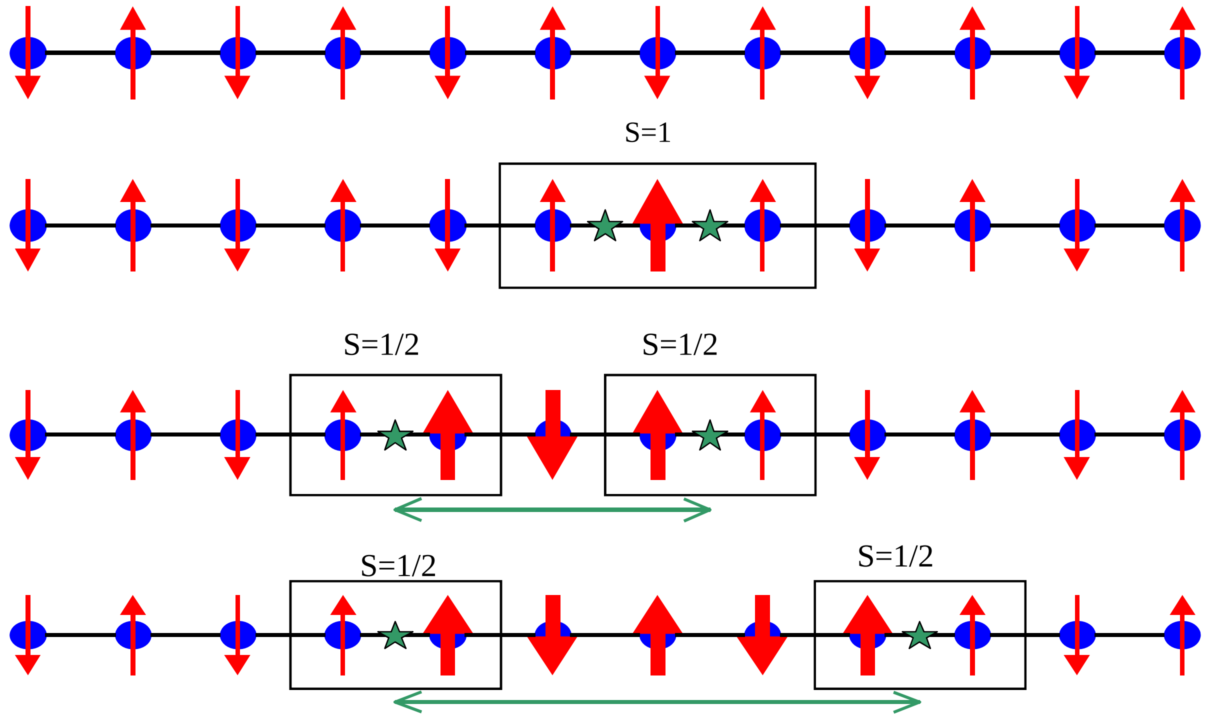
- Ground state has no long-range Néel order.
- Ground state consists of 50% spin-flip states
- Little physical insight into the quasi-particles.

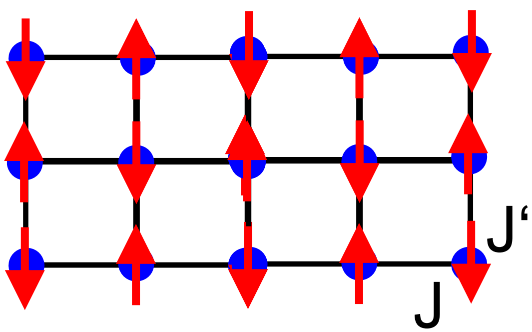
The Bethe Ansatz has been a long standing problem of theoretical condensed matter

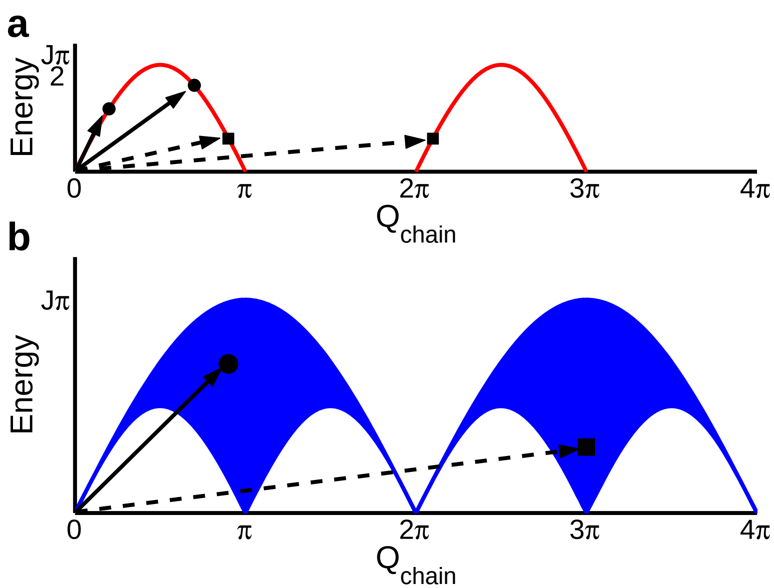
Spinons Excitations in the spin-1/2 AFM chain

Fadeev and Taktajan (1981)

The fundamental excitations are spinons not magnons.







<u>Spinons</u>

- Fractional spin-½ particles
- created in pairs
- spinon-pair continuum

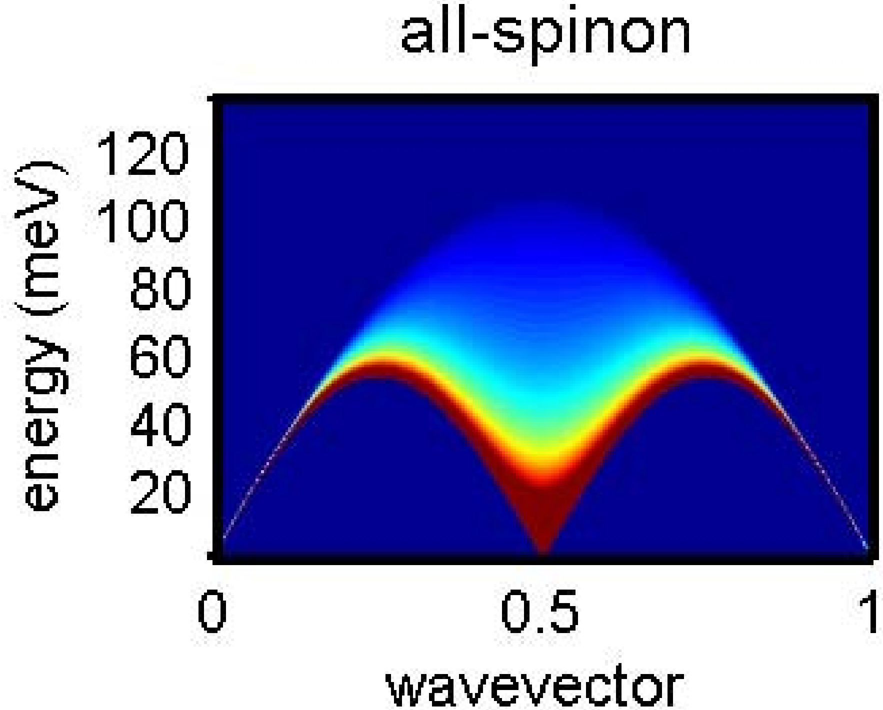
Solution of Bethe Ansatz

Several approximate theories have since been postulated for the spinon continuum of the spin-1/2 Heisenberg chain

- Müller Ansatz
- Luttinger Liquid Quantum Critical point

In 2006 J.-S. Caux and J.-M. Maillet solved the 1D, spin-1/2, Heisenberg, antiferromagnet, 75 years after the Bethe Ansatz was proposed.

J.-S. Caux,
R. Hagemans,
J. M. Maillet
(2006)



1D S-1/2 Heisenberg Antiferromagnetic - $KCuF_3$

Cu^{2+} ions S=1/2

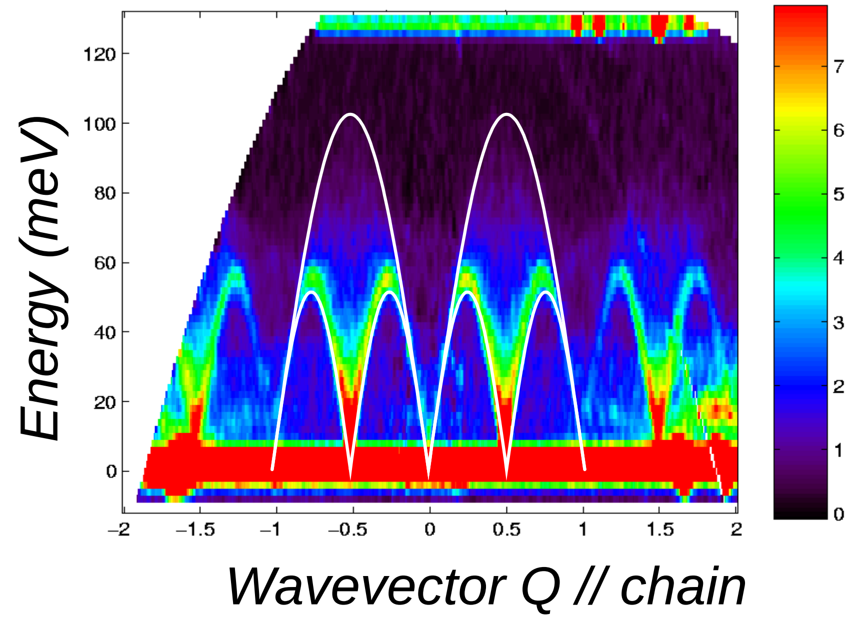
Antiferromagnetic chains, $J_{//}$= -34 meV

Weak interchain coupling, $J_{\perp}/J_{//}$ ~ 0.02

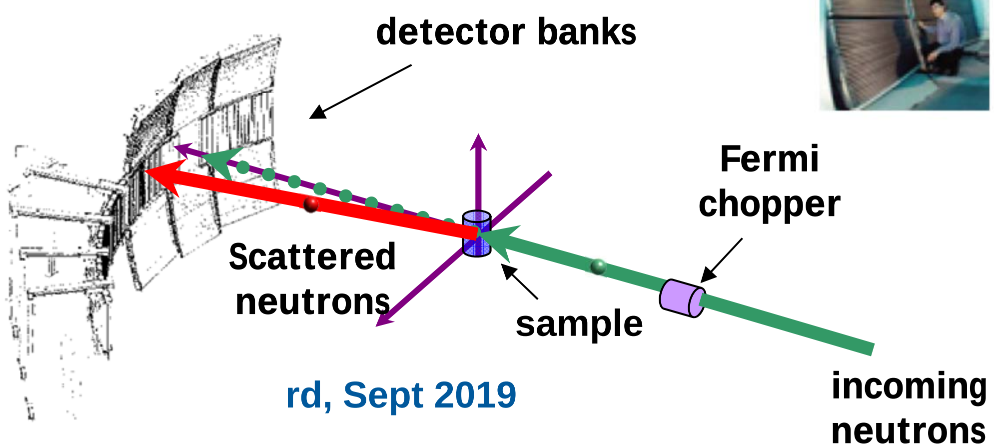
Antiferromagnetic order T_N ~ 39K

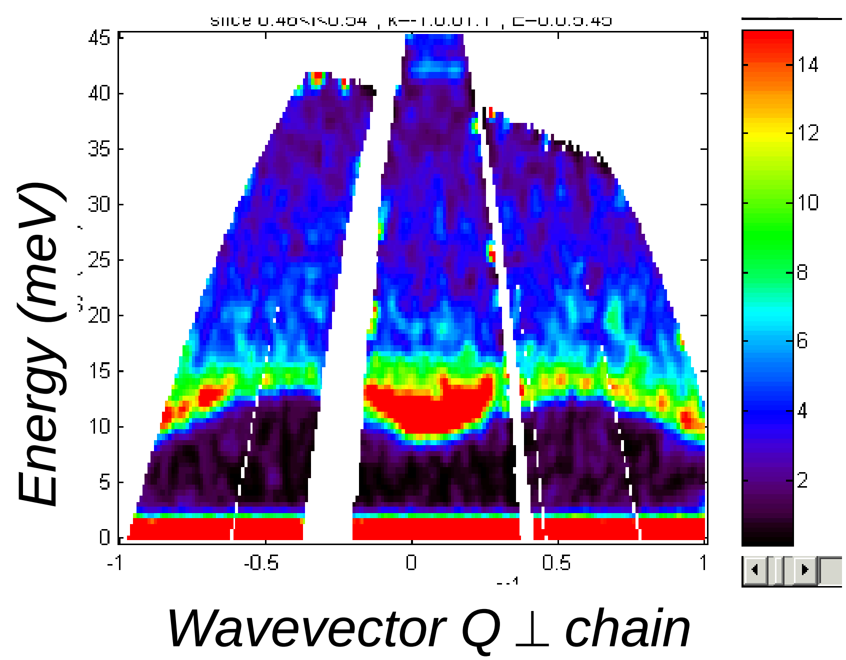
Only 50% of each spin is ordered

$$\hat{H} = J_{\|}\sum_{r}\vec{S}_{r,l}\cdot\vec{S}_{r+1,l} + J_{\perp}\sum_{l,\delta}\vec{S}_{r,l}\cdot\vec{S}_{r,l+\delta}$$



MAPS neutron spectrometer, ISIS



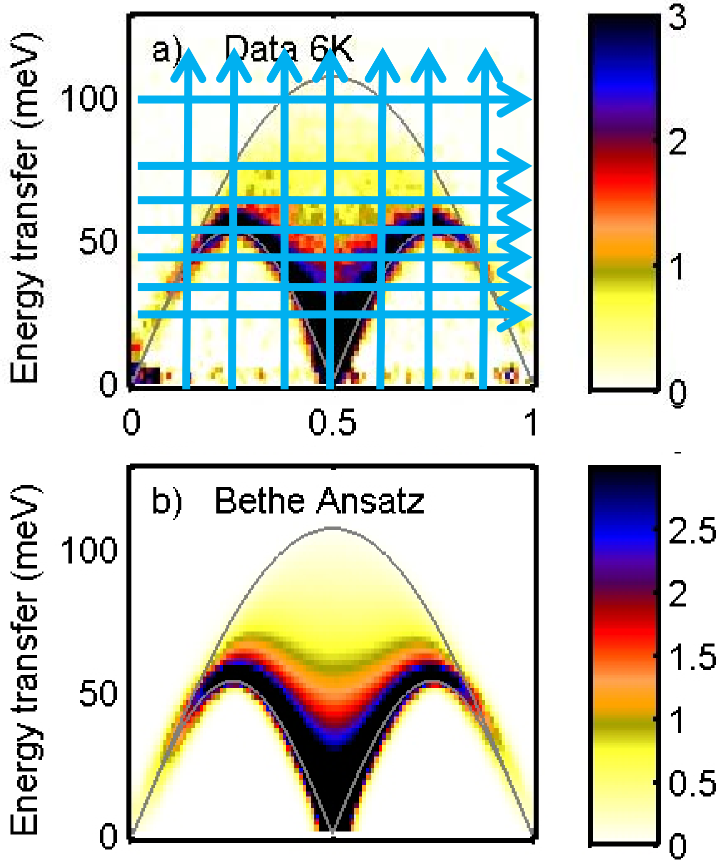


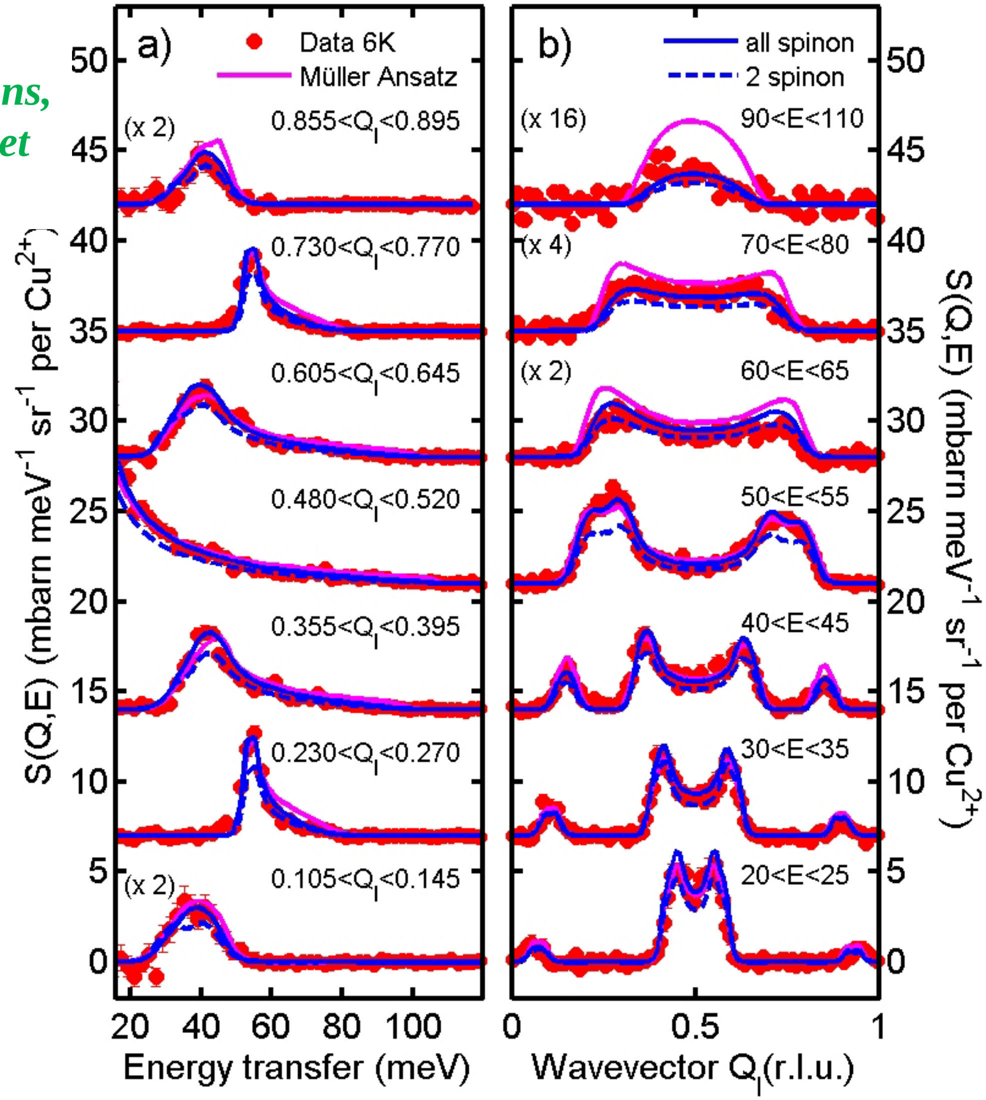
rd, Sept 2019

$KCuF_3$ compared to Bethe Ansatz, 2 and 4 spinons

Constant energy and constant-wavevector cuts compared to simulations

J.-S. Caux, R. Hagemans, J. M. Maillet (2006)





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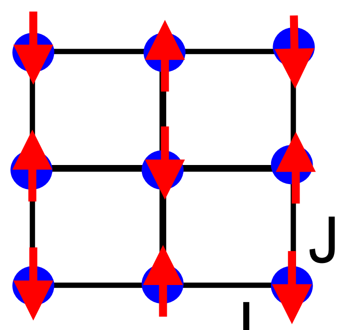
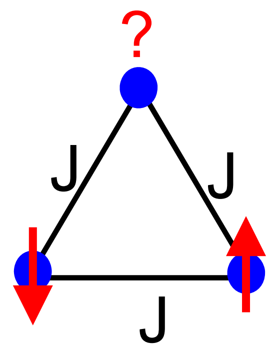
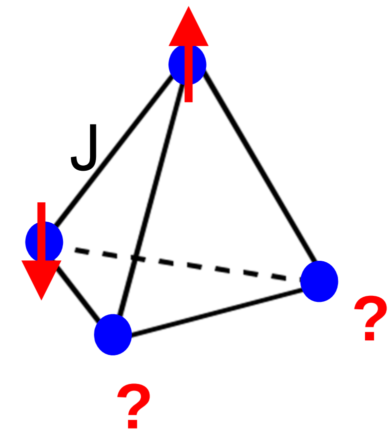
Frustrated Magnets

Geometrical Frustration

- Geometrical arrangements, e.g. triangular and tetrahedral geometries
- Antiferromagnetic interactions between 1st neighbour magnetic ions.

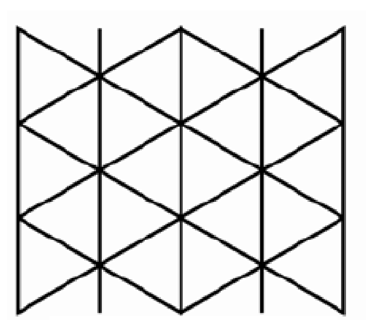




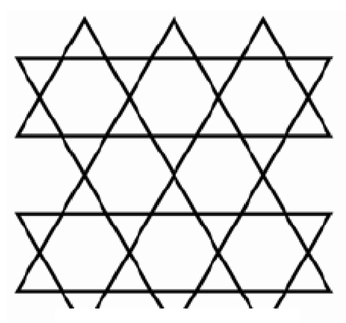


2-dimensional

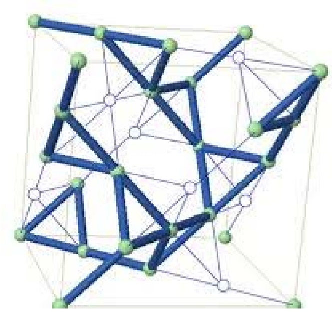
3-dimensional



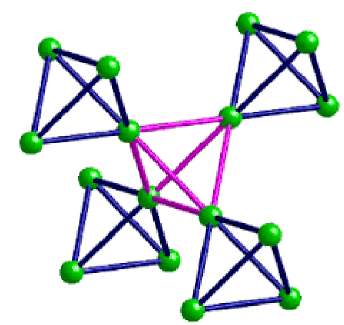
Triangular lattice



Kagome lattice



Hyperkagome lattice



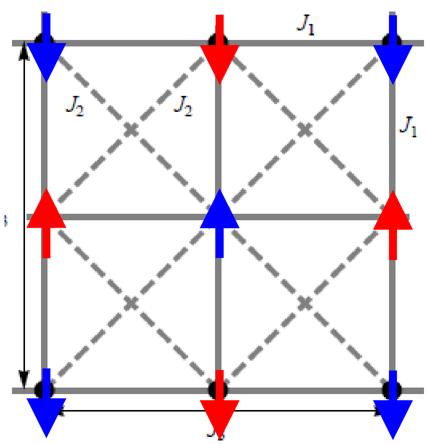
Pyrochlore lattice

B. Lake; Oxford, Sept 2019

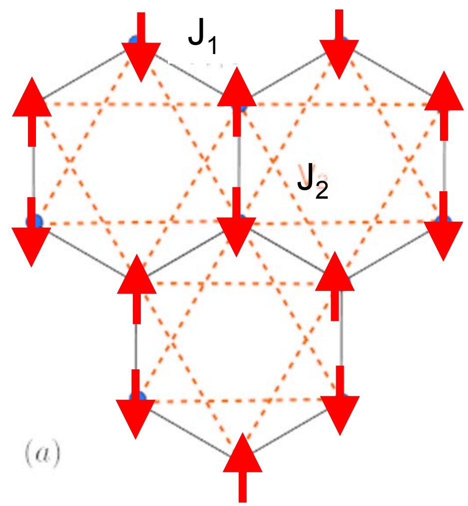
Frustration from competing interactions

- Second neighbour or further neighbour interactions compete with first neighbour interactions.
- The second neighbour interactions must be AFM

J_1=0
J_2=AFM

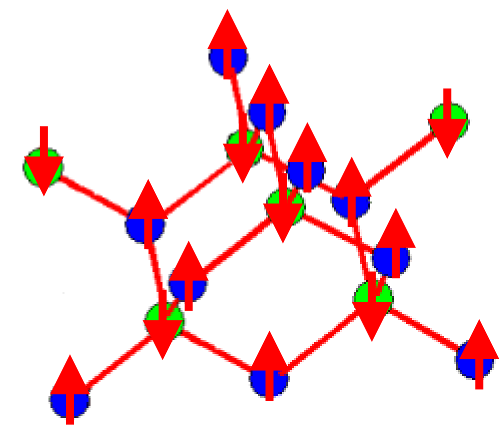


honeycomb



J_1=AFM
J_2=0

diamond



J_1=AFM
J_2=0

Frustration arising from anisotropy

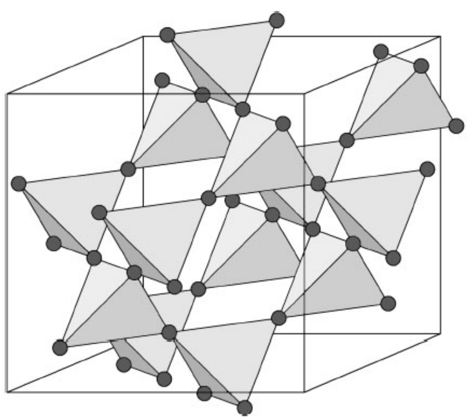
The pyrochlore lattice – corner-sharing tetrahedra

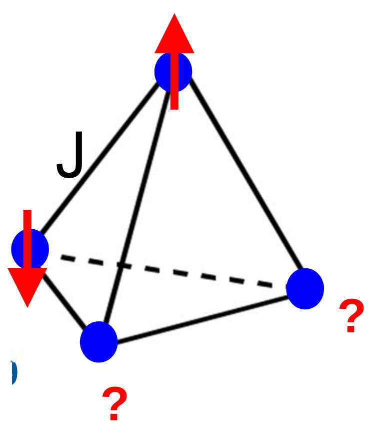
- No anisotropy & AFM interaction $\Rightarrow$ highly geometrically frustrated, no magnetic order

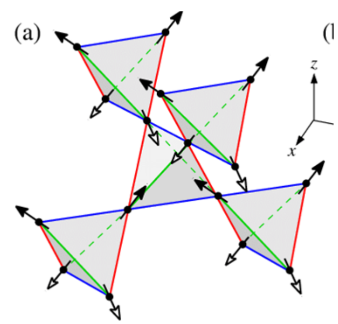
- Local 111 anisotropy & AFM interactions $\Rightarrow$ long-range magnetic order, all-in-all-out configuration! Unfrustrated

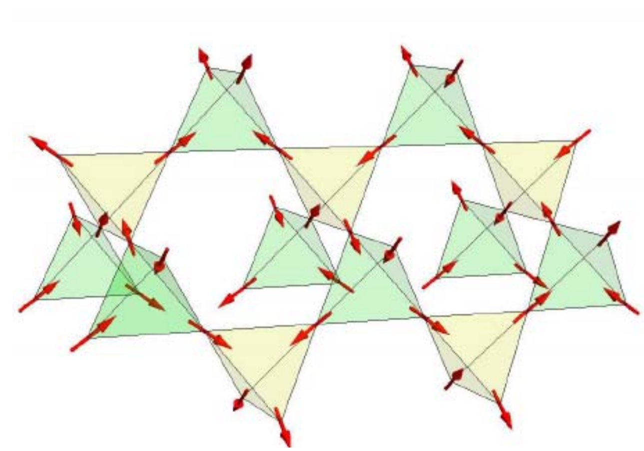
$$H = \sum_{n,m} -J_{n,m}\left[\varepsilon\left(\mathbf{S}_n^x\mathbf{S}_m^x + \mathbf{S}_n^y\mathbf{S}_m^y\right) + \mathbf{S}_n^z\mathbf{S}_m^z\right]$$

- Local 111 anisotropy & FM interactions $\Rightarrow$ 2-in-2-out on each tetrahedra, no unique ground state, famous spin ice with monopole excitations. Frustrated









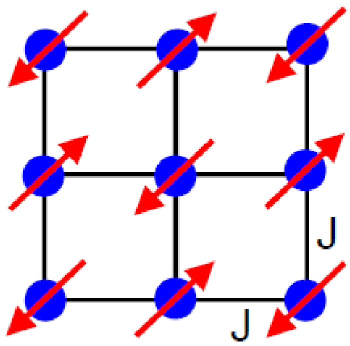
Quantum Magnets

Spin liquids,

- no local order, no static magnetism
- highly entangled, dynamic ground state
- topological order, spinon excitations

Two Dimensional Quantum Magnets

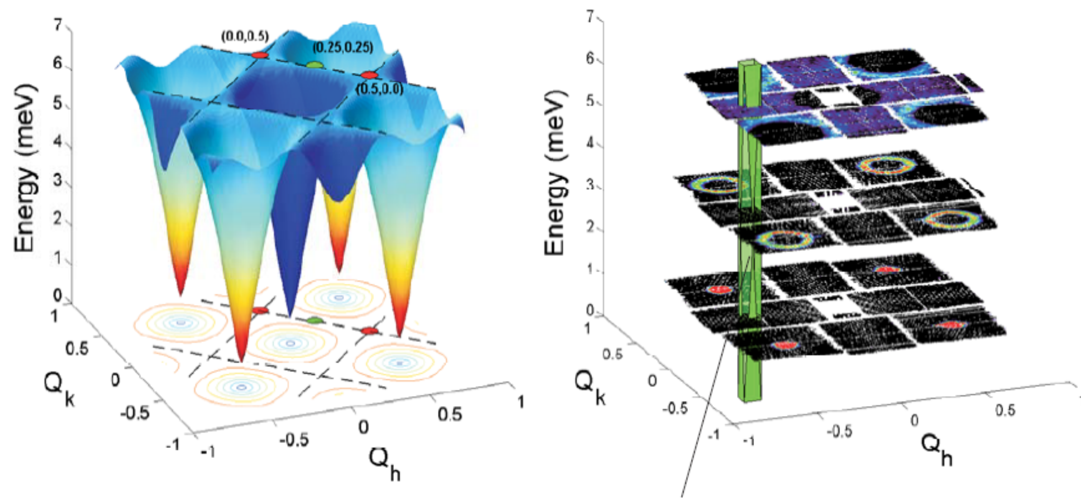
2-Dimensional Antiferromagnet - Square Lattice



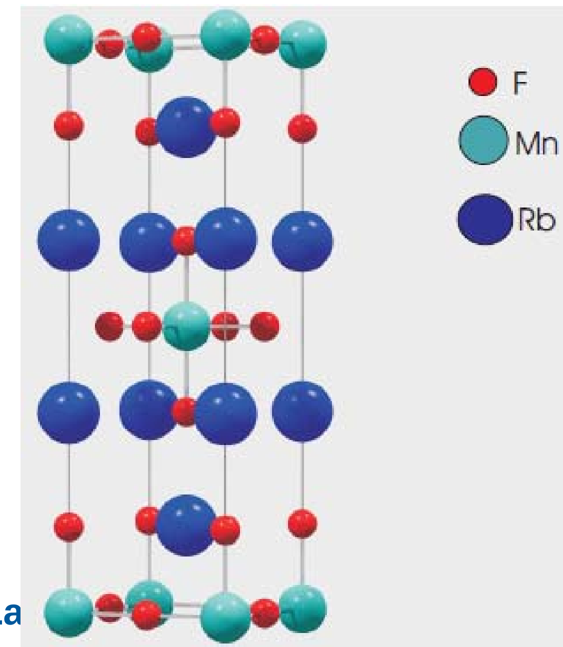
Ground state
long range order

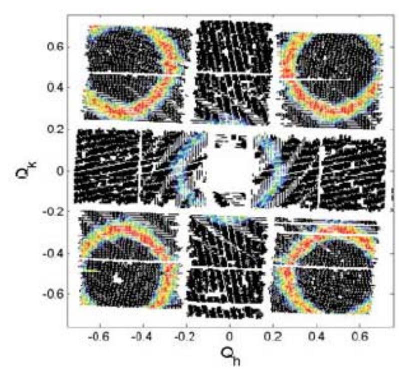
Excitations
Spin-waves

T Huberman et al J. Stat. Mech. (2008) P05017



Rb_2MnF_4
2-Dimensional Spin-5/2
Heisenberg Antiferromagnet



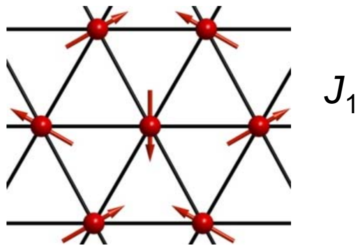


2-Dimensional Antiferromagnet - Triangular Lattice

Triangular Lattice

Ground state – long range order

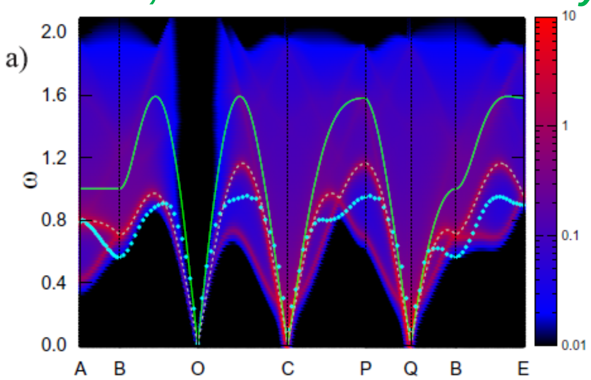
isotropic



$$\varphi = 120^\circ; \ \boldsymbol{k}_m = 1/3$$

Excitations

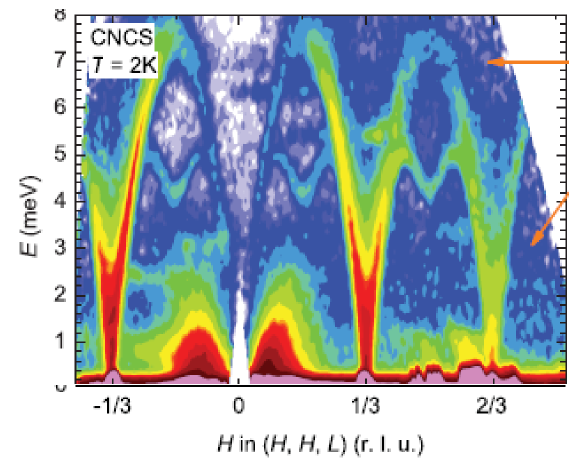
A Mezio, et al New Journal of Physics (2012)



renormalised and broadened compared to spin-wave theory

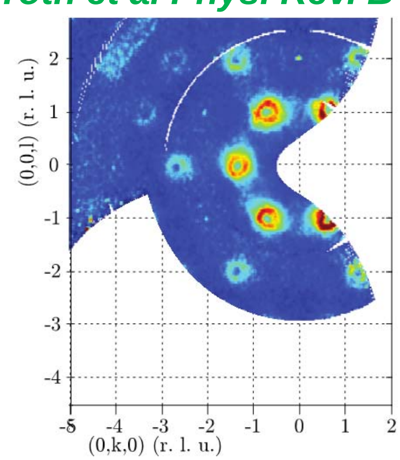
$CuCrO_2$ S-3/2, triangular lattice

M Frontzek et al Phys. Rev. B (2011)



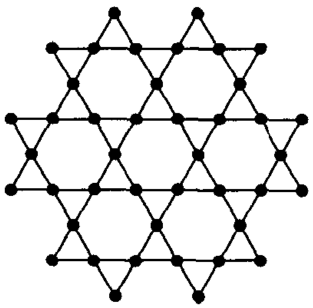
Alpha-Ca_2CrO_4 S-3/2, triangular lattice

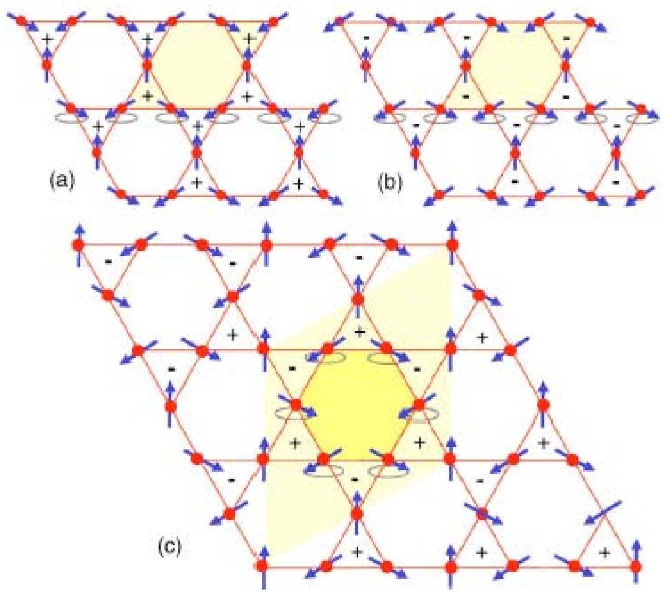
S Toth et al Phys. Rev. B (2011)



2-Dimensional Antiferromagnet - Kagome Lattice

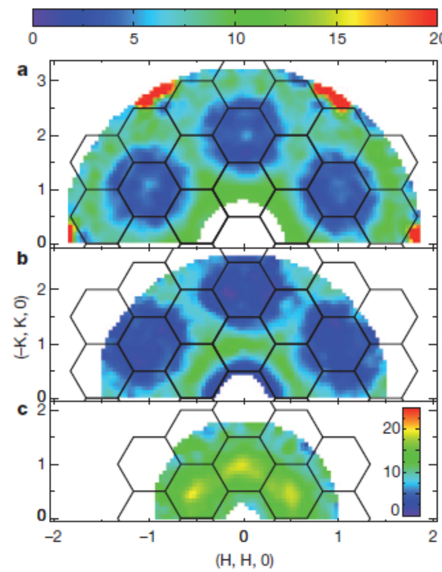
Kagome Lattice



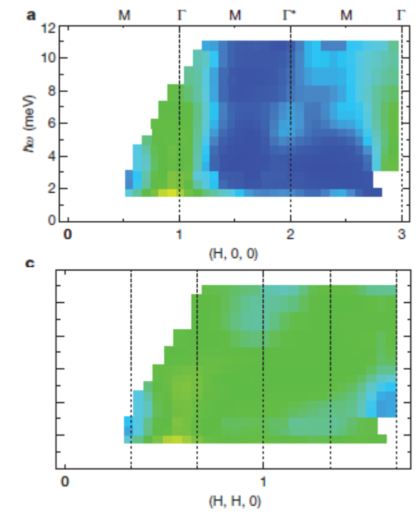


S-5/2
Long-range order
Spin-wave excitation

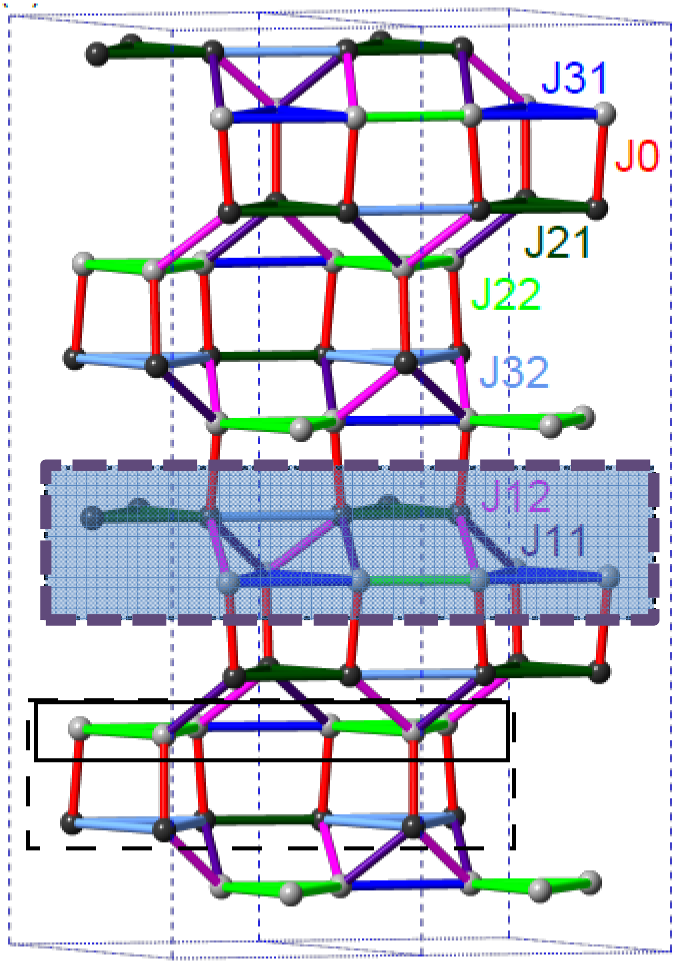
S-1/2
no order
diffuse spinon
excitations



e.g. Herbertsmithite
T.-H. Han
Nature 492, 406 (2012)



$Ca_{10}Cr_7O_{28}$ - Crystal structure



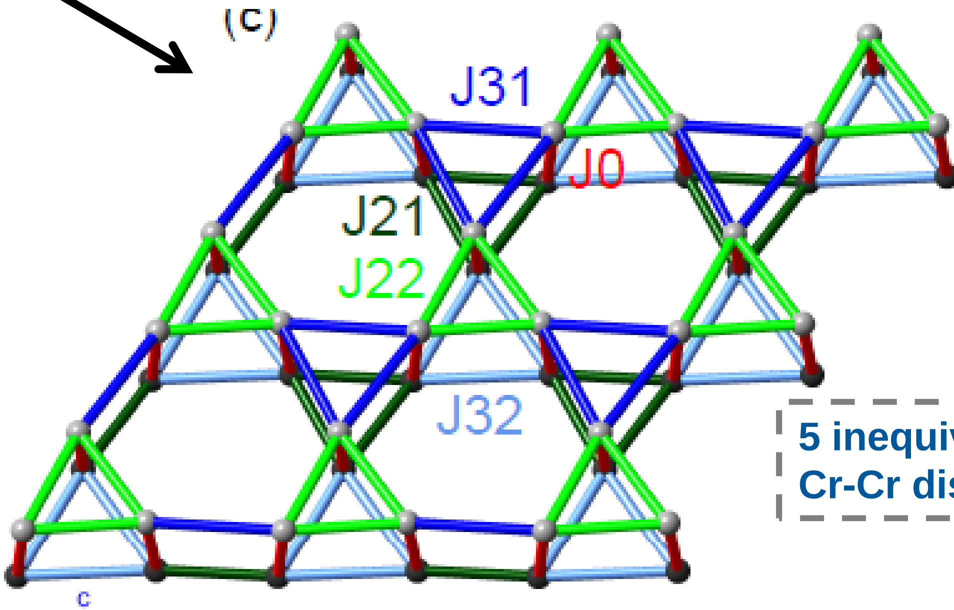
space group ***R*3*c***

D. Gyepesova, Acta Cryst. C69, 111 (2013)

- Cr^{5+} spin = ½ ions (1 electron in 3d-shell)
- 7 different exchange path in structure
- No long-range magnetic order

Kagome bilayer model

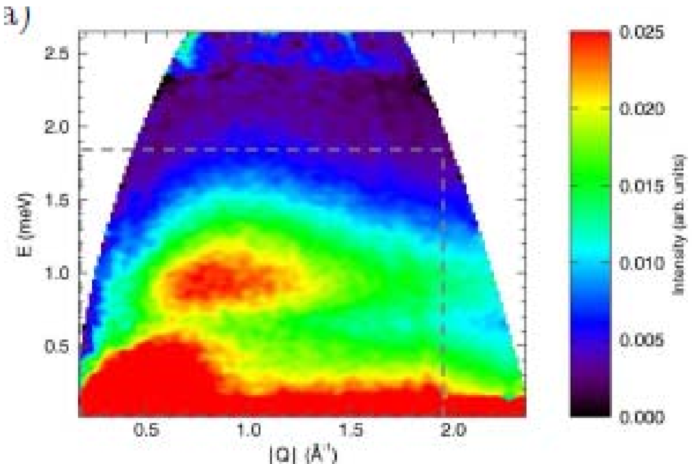
- *a-b* plane shows distorted kagome bilayers
- large blue and small green triangles alternate within each layer, and between layers



5 inequivalent Cr-Cr distances

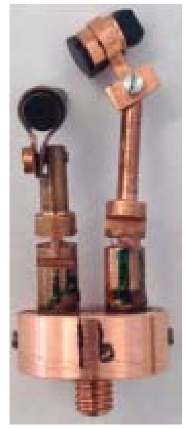
Inelastic Neutron Scattering – Zero Field

Powder; TOFTOF, FRM2; T=0.43K

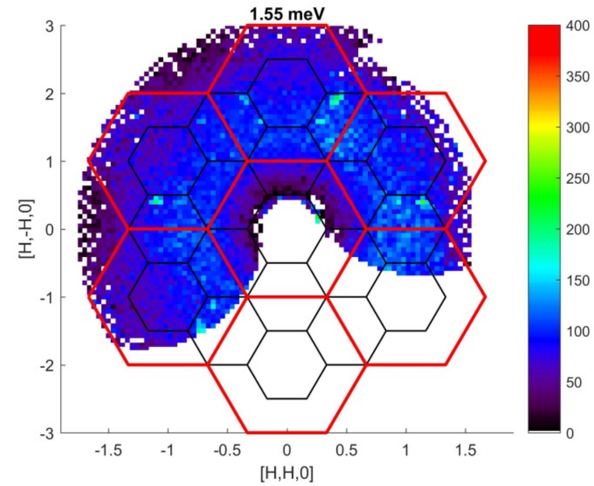


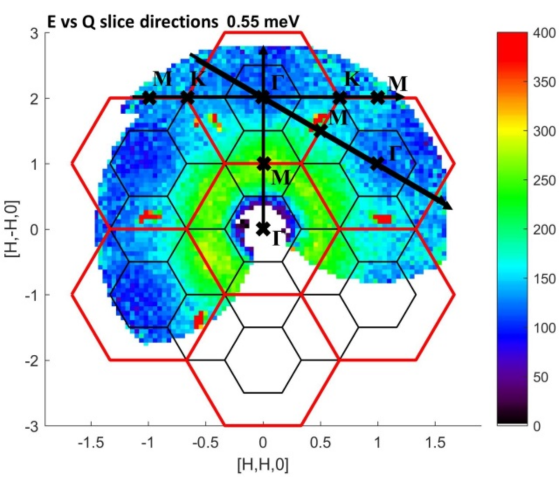
- Excitations to 1.6meV
- Two Bands of excitations

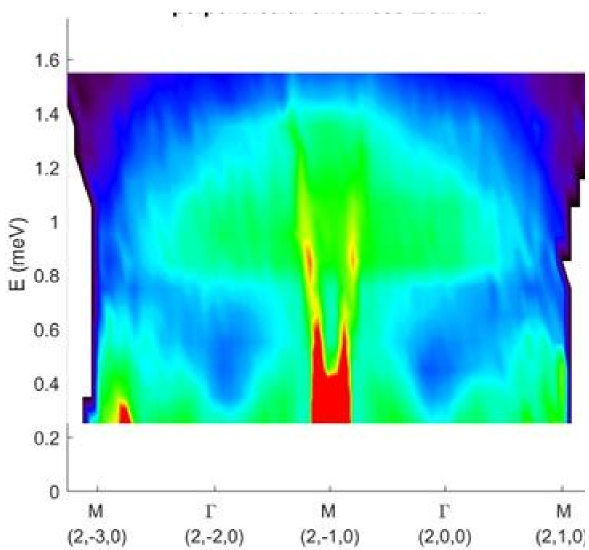
Single Crystals

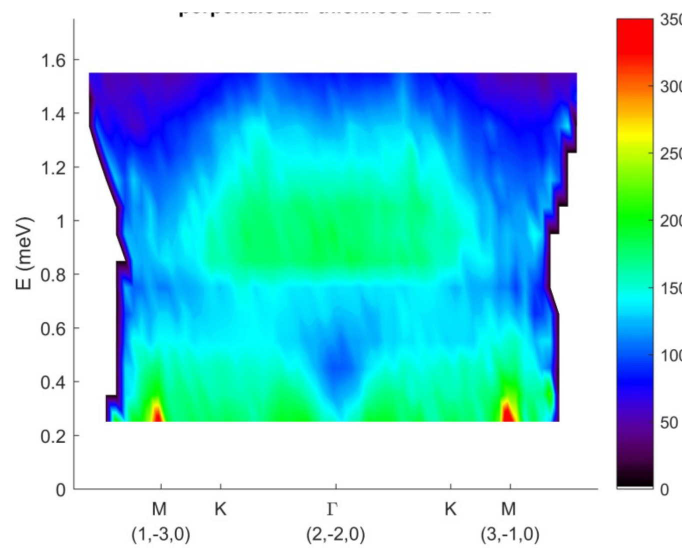


[H,K,0]; MACS, T=0.09K







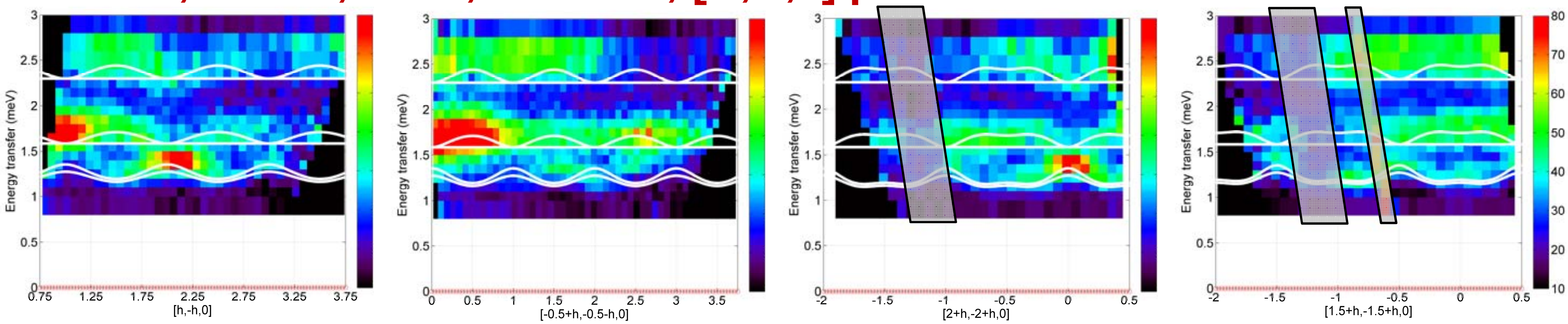


B. Lake; Oxford, Sept 2019

Broad diffuse scattering, no spin-waves

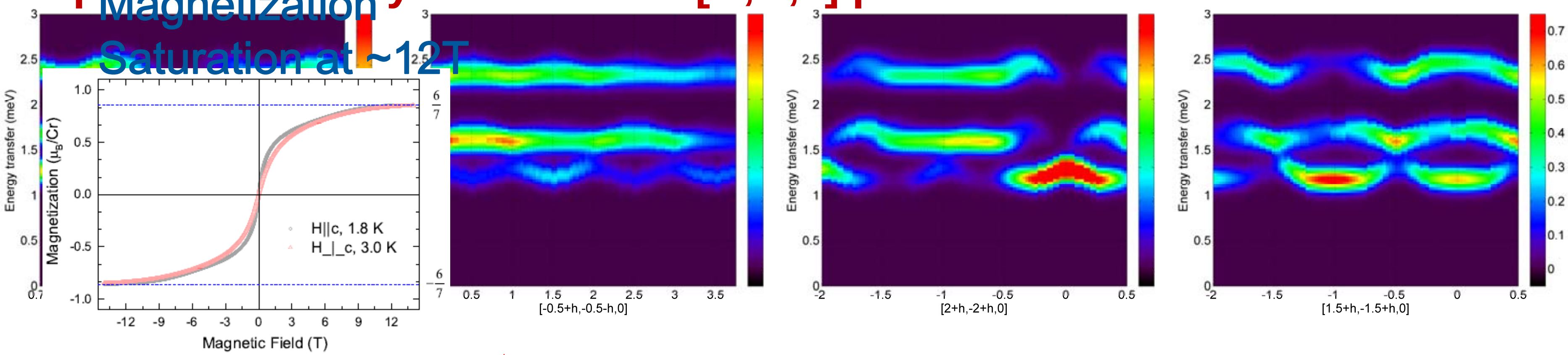
Inelastic Neutron Scattering – High Field

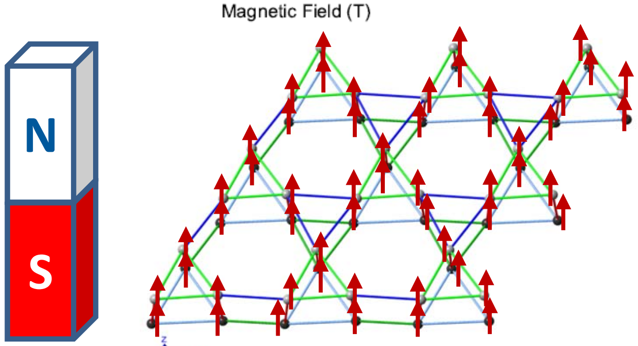
H=11T; MACS, NIST; T=0.09K; [H,K,0] plane



Spin wave theory calculations [H,K,0] plane

Magnetization
Saturation at ~12T





SpinW library, S. Toth and B. Lake, Journal of Physics: Cond. Mat. 27, 166002 (2015)

$Ca_{10}Cr_7O_{28}$ - Magnetic model - Exchange couplings

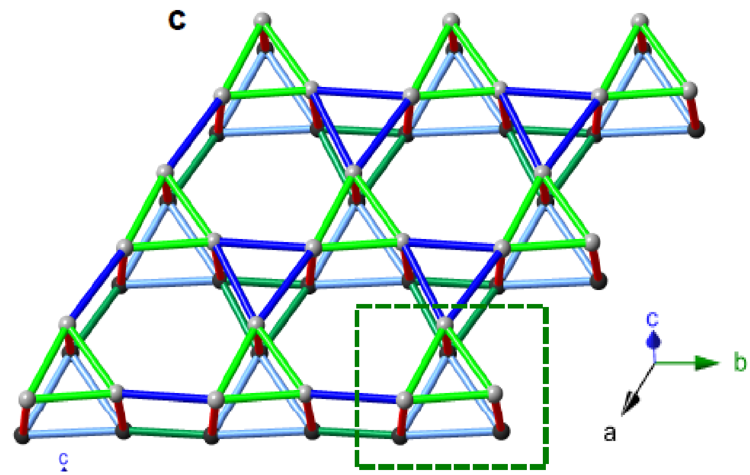
$$\mathcal{H} = J_{ij} \sum_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

exchange	coupling [meV]	type
J0	-0.08(4)	FM
J21	-0.76(5)	FM
J22	-0.27(3)	FM
J31	0.09(2)	AFM
J32	0.11(3)	AFM

} intrabilayer (J0)

} triangles (J21, J22)

} triangles (J31, J32)

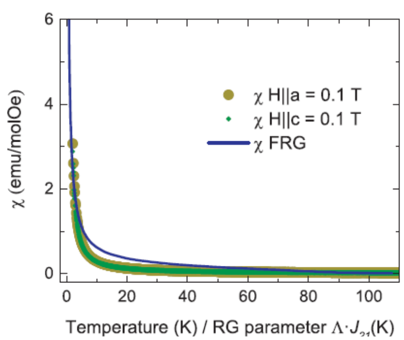


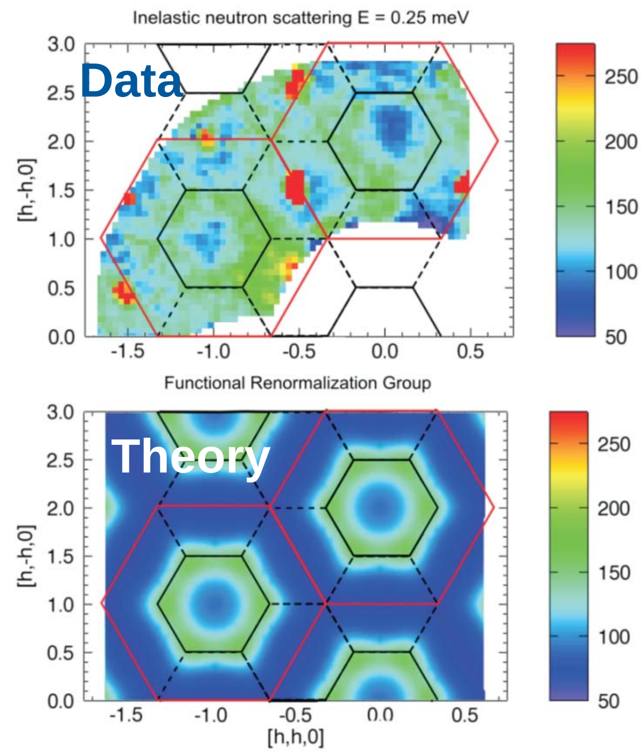
Pseudo-Fermion Functional Renormalisation Group

Using the Hamiltonian extracted from INS

⇒ Susceptibility shows no long-range order

⇒ Diffuse magnetic scattering





Non-ordered ground state, diffuse spinon scattering.
Reveals highly robust spin liquid state

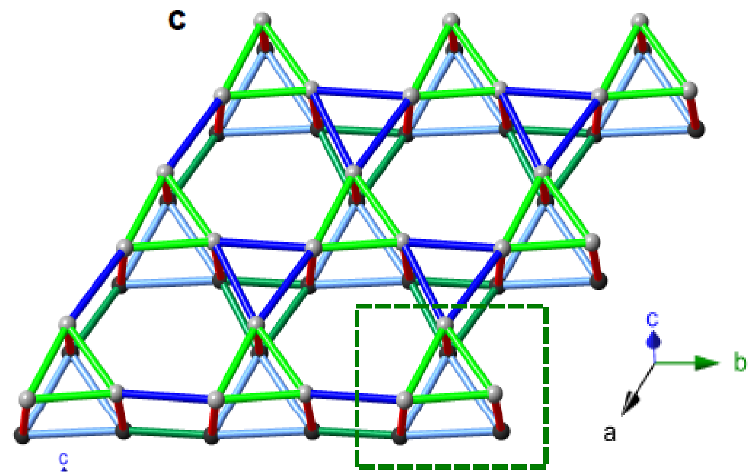
Why is $Ca_{10}Cr_7O_{28}$ a spin liquid?

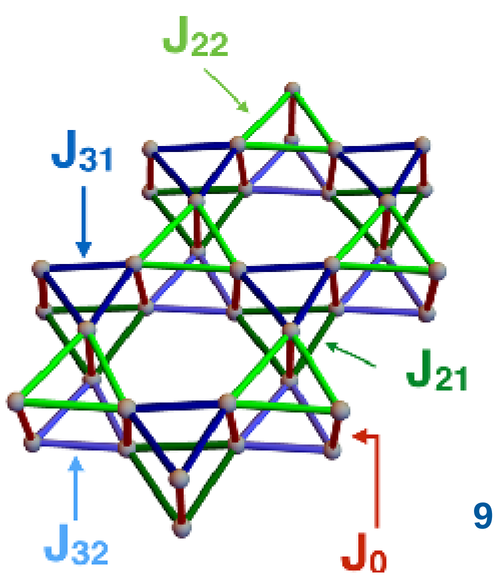
exchange	coupling [meV]	type
J0	-0.08(4)	FM
J21	-0.76(5)	FM
J22	-0.27(3)	FM
J31	0.09(2)	AFM
J32	0.11(3)	AFM

} intrabilayer

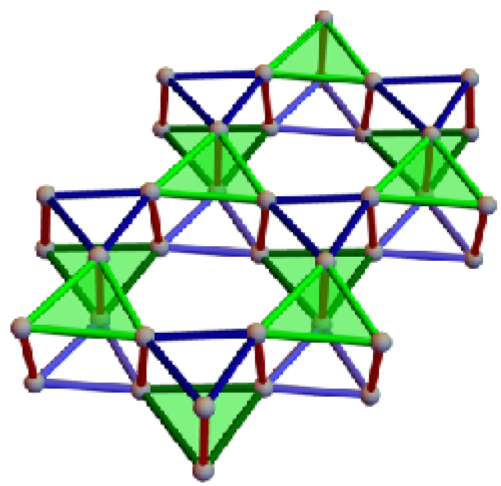
} triangles

} triangles





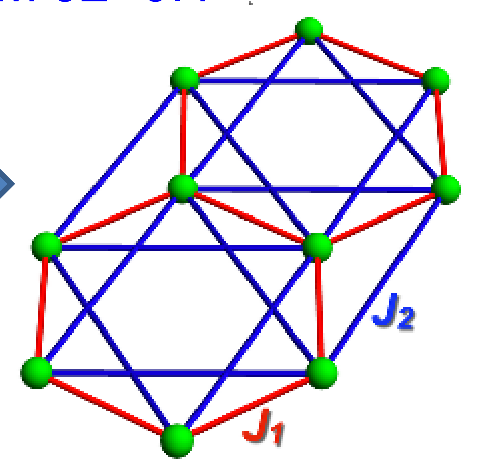
Strong FM interactions on alternating triangles



Effective S=3/2 honeycomb

FM J1=J0=-0.08

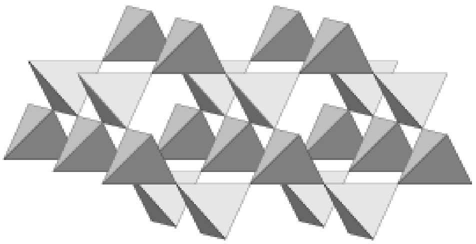
AFM J2=0.1

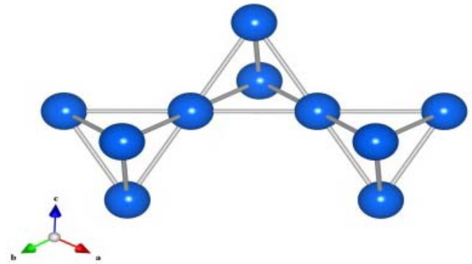


Three Dimensional Quantum Magnets

Frustrated 3-Dimensions Magnets – Pyrochlore Lattice

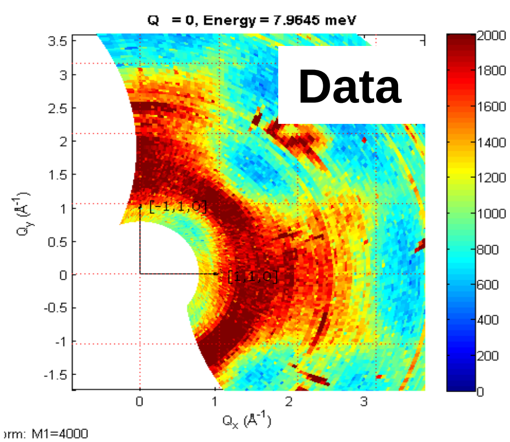
Pyrochlore Lattice – corner-sharing tetrahedra

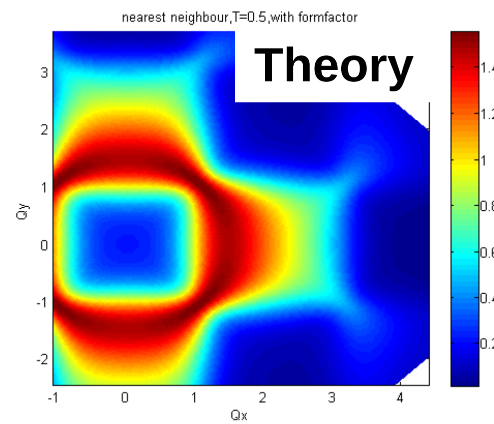




Interconnected chains
Antiferromagnetic J
3D frustration

MgV_2O_4, V^{3+} has spin-1



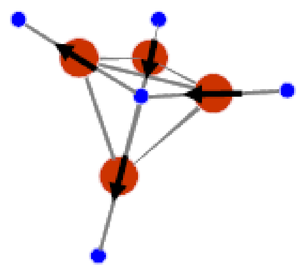


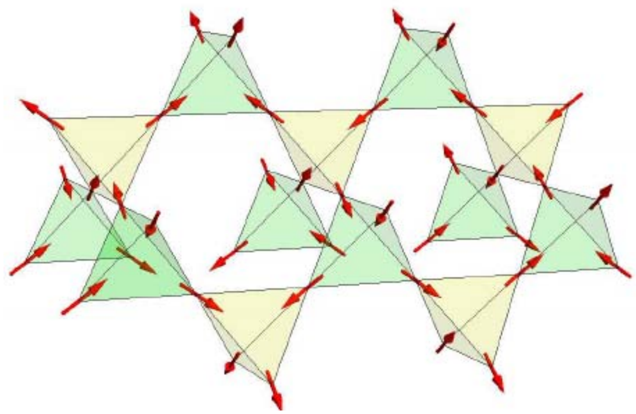
B. Lake; Oxf

Constant Energy E=8meV, IN20 with Flatcone
reveals broad diffuse scattering very different from spin-wave excitations

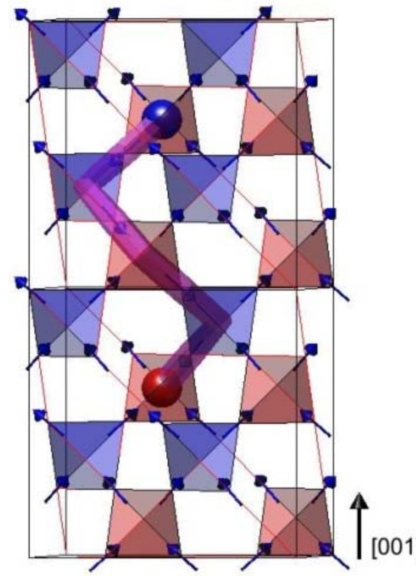
3-Dimensions - Pyrochlore Magnets

Spin Ice
- •Ferromagnetic interactions
- •Strong Ising anisotropy
- •Ice rules 2 in, 2 out

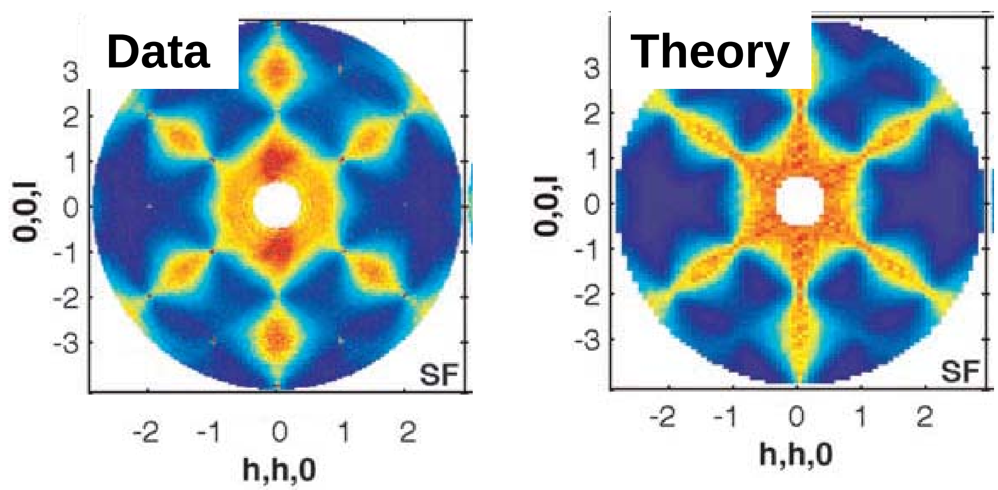




Ground state - topological order



Excitations - monopoles

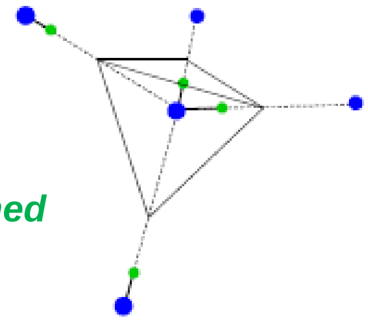


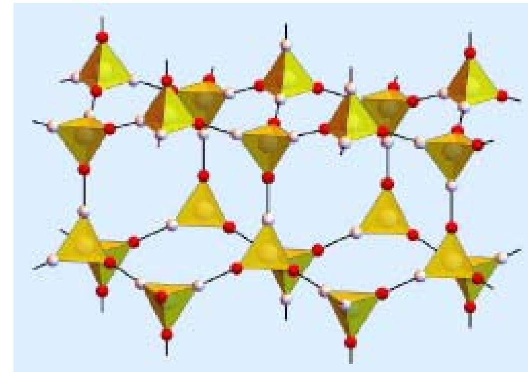
T. Fennell et al Science 326 415 (2009)
D.J.P. Morris, et al Science 326, 411 (2009)

Water Ice

Structural frustration e.g. water ice

- 2 Hydrogens in 2 out

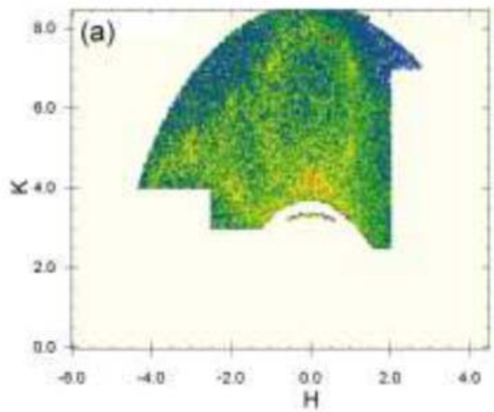


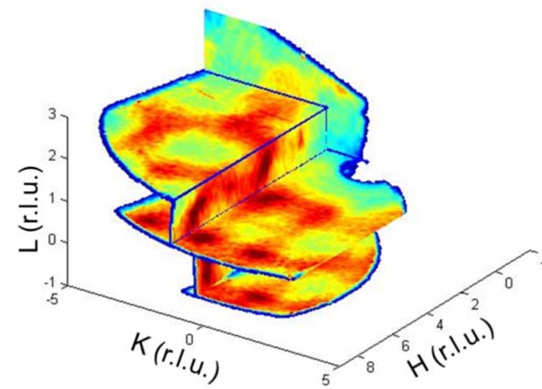


K. Siemensmeyer, J.-U. Hofmann, S. V. Isakov, B. Klemke, R. Moessner, J. P. Morris, to be published

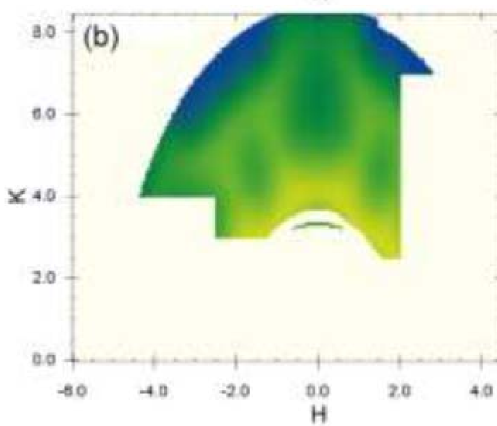
D_2O crystal

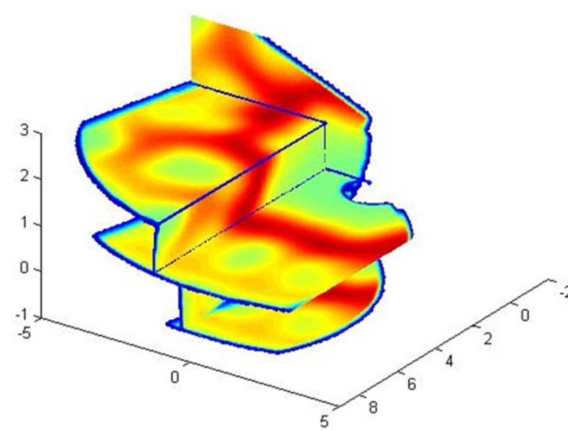
Neutron scattering of water ice





Simulation - Displacement of D^+ can be mapped onto a pseudo-spin





Accurate description with only one free parameter

Summary

Origins of unconventional magnetism

antiferromagnetic interactions, low spin value, low dimensional
Example spin-1/2 dimer antiferromagnet
Example spin-1/2, antiferromagnetic chain

Origins of frustrated magnetism

geometric frustration, competing interactions and anisotropy

Examples of frustrated magnets

2-Dimensional magnets e.g. Square, triangular, kagome, lattice
3-Dimensional magnets e.g. pyrochlore, spin ice and water ice