Introduction to Neutron Scattering

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Outline

Conventional magnets

Long-range magnetic order and spin-wave excitations

Neutron scattering concepts

Neutron properties, neutron sources,
neutron scattering triangles and cross sections

Neutron scattering techniques

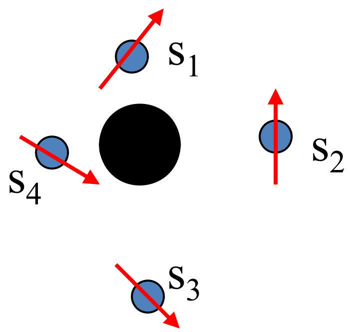
Neutron diffraction, inelastic neutron scattering TAS TOF

Spin-waves

Calculations and measurements

Conventional Magnets

Conventional Magnetism – Magnetic Moments

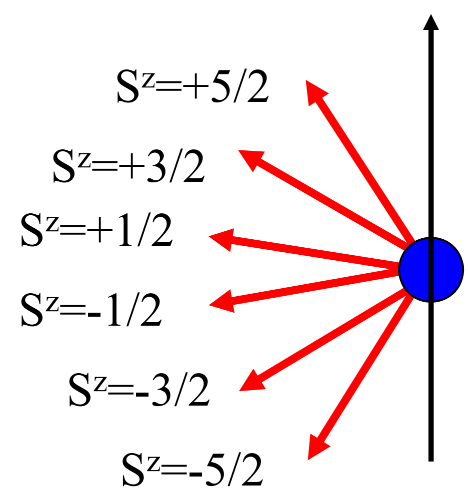


$S = s_1 + s_2 + s_3 + s_4 + \ldots$

- Electrons possess spin and orbital angular momenta (*s* and ***l***).
- ***S*** and ***L*** for an ion can be determined by summing the electronic ***s*** and ***l*** of the unpaired electrons
- The ionic magnetic moment is $\boldsymbol{m} = g_s \mu_B \boldsymbol{S} + \mu_B \boldsymbol{L}$
.

The Mn^{2+} ion, $S=5/2$

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



- ***S*** is the quantum number associated with the angular momentum ***S***.
- ***S*** is restricted to take on discrete values either integer or half integer.

Conventional Magnetism - Exchange Interactions

Heisenberg interactions

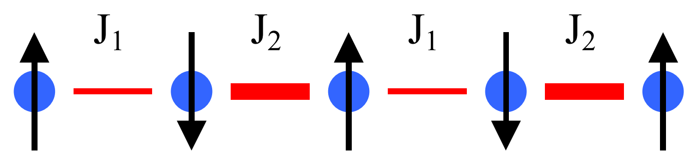
$$H=\sum_{n,m} J_{n,m}\mathbf{S}_n.\mathbf{S}_m$$

J < 0 ferromagnetic
J > 0 antiferromagnetic

3D magnet

$|J_1|=|J_2|=|J_3|=|J_4|$

e.g. $RbMnF_3$



1D magnet

$|J_1|=|J_2|$, $J_3=J_4=0$

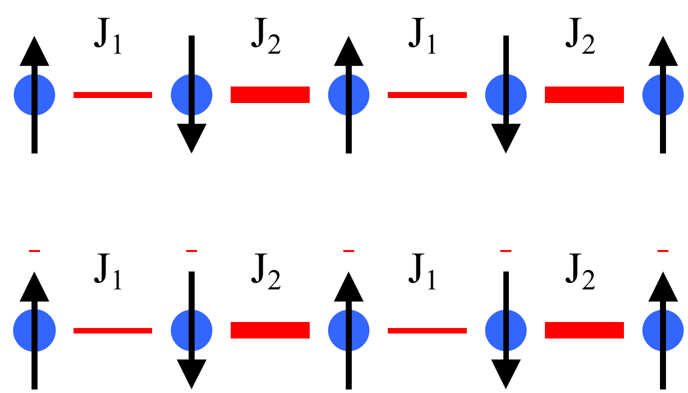
e.g. $KCuF_3$

2D magnet

$|J_1|=|J_2|=|J_3|$, $J_4=0$

e.g. La_2CuO_4

and CFTD



1D alternating magnet

$|J_1|\neq|J_2|$, $J_3=J_4=0$

e.g. $CuGeO_3$ and $CuWO_4$

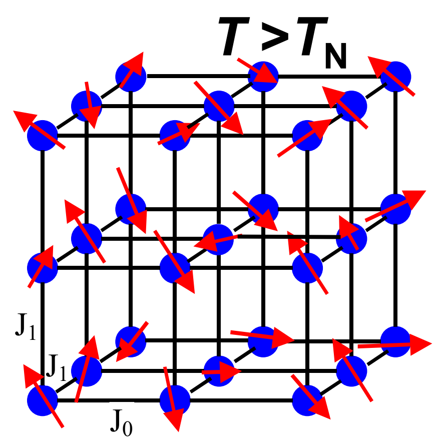
Anisotropic interactions

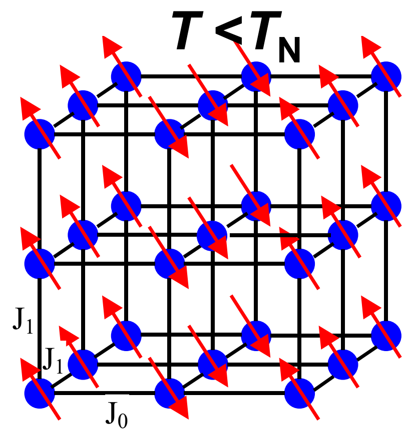
$$H=\sum_{n,m} -J_{n,m}\left[\varepsilon\left(\mathbf{S}_n^x\mathbf{S}_m^x+\mathbf{S}_n^y\mathbf{S}_m^y\right)+\mathbf{S}_n^z\mathbf{S}_m^z\right]$$

Conventional Antiferromagnets

Real Space

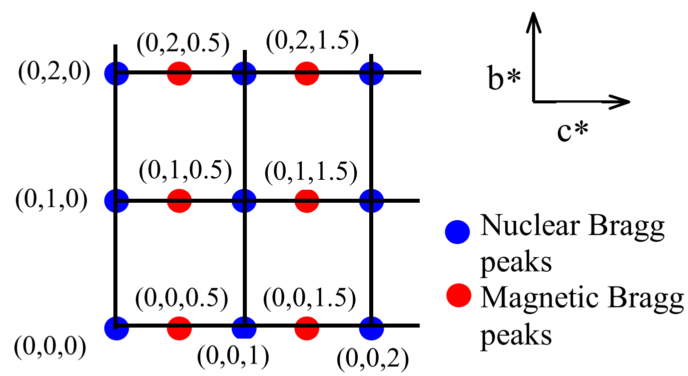
- Long-range magnetic order on cooling as thermal fluctuations weaken

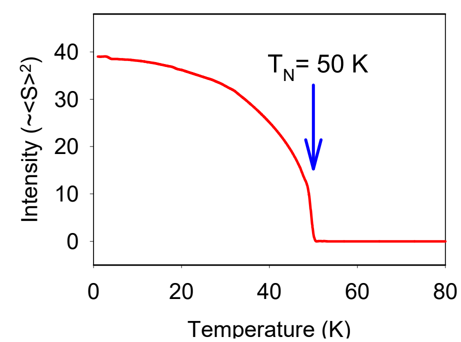




Reciprocal Space

- Magnetic Bragg peaks appear below the transition temperatures and grow as a function of temperature



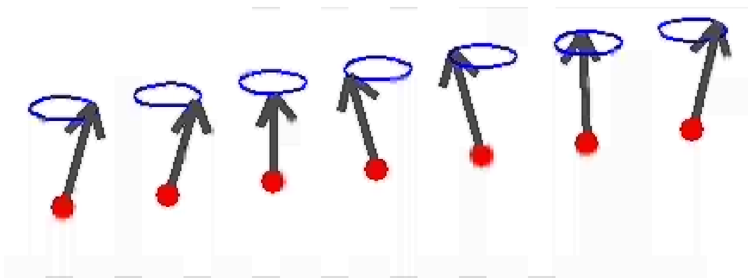


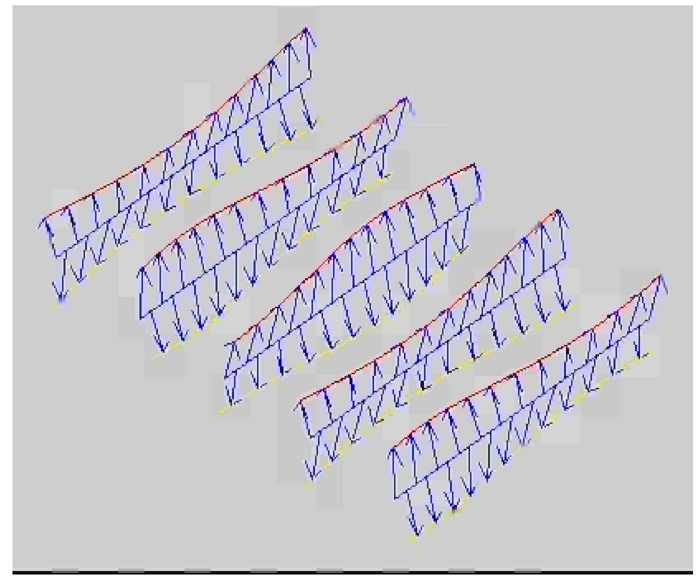
B. Lake; TMS, Aug 2022

Magnetic Excitations

Real Space

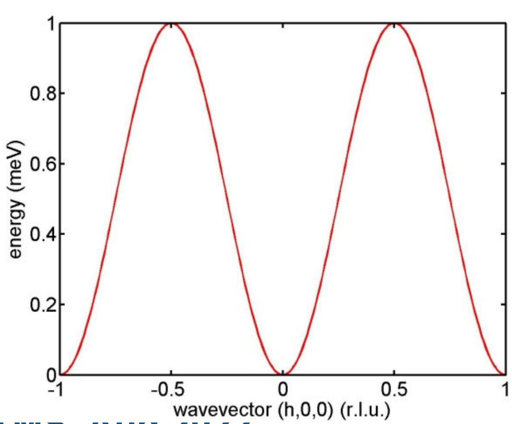
- Spin-waves are the collective motion of spins, about an ordered ground state (similar to phonons)

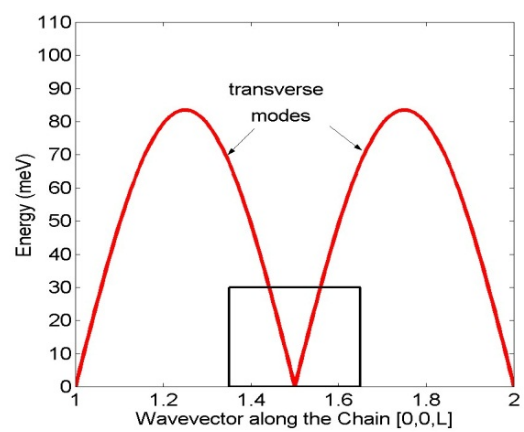




Reciprocal Space

- Observed as a well defined dispersion in energy and wavevector





Neutron Scattering Concepts

Basic Properties of the Neutron

The neutron is a nuclear particle

The neutron has a mass similar to the proton

– m_n=1.675x10^{-27}kg

The neutron is electrically neutral

– Charge = 0

The neutron has spin angular momentum

– S_n=1/2

The neutron has a magnetic moment (antiparallel to spin)

– $\mu_n = \gamma\mu_N$; γ=-1.913; $\mu_N = e\hbar/m_p$;

Neutron lifetime

– τ=886s (~15 minutes)

The Neutron – both wave and particle

- In neutron scattering experiments neutrons behave like particles when they are created, like waves when they scatter, and again like particles when they are detected.

- Momentum is $p=m_n v$, and in quantum mechanics it is related to wavevector k, by $p=\hbar k$ (units of k are Å^{-1})

$$v = \frac{\hbar}{m_n}k; \quad k = \frac{m_n}{\hbar}v$$

- A particle has a de Broglie wavelength λ (=$2\pi/k$) (units Å)

$$\lambda = \frac{2\pi}{k} = \frac{2\pi\hbar}{m_n v}$$

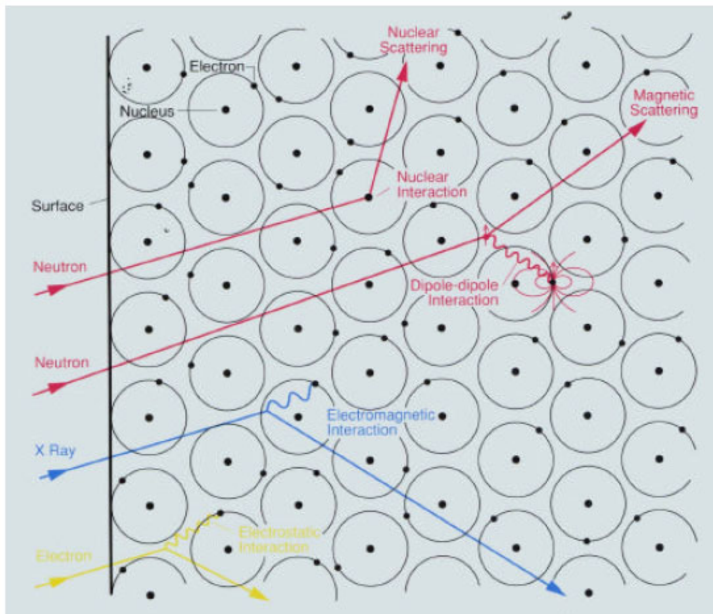
- kinetic energy E (units eV, meV where 1eV=$1.6\text{x}10^{-19}$)

$$E = \frac{1}{2}m_n v^2 = \frac{\hbar^2 k^2}{2m_n}$$

	v [ms^{-1}]	λ^{-1} [Å^{-1}]	k [Å^{-1}]	$\sqrt{E}$ [meV$^{1/2}$]
v [ms^{-1}]	1	2.528×10^{-4}	1.588×10^{-3}	2.286×10^{-3}
λ^{-1} [Å^{-1}]	3956	1	6.283	9.045
k [Å^{-1}]	629.6	0.1592	1	1.440
$\sqrt{E}$ [meV$^{1/2}$]	437.4	0.1106	0.6947	1

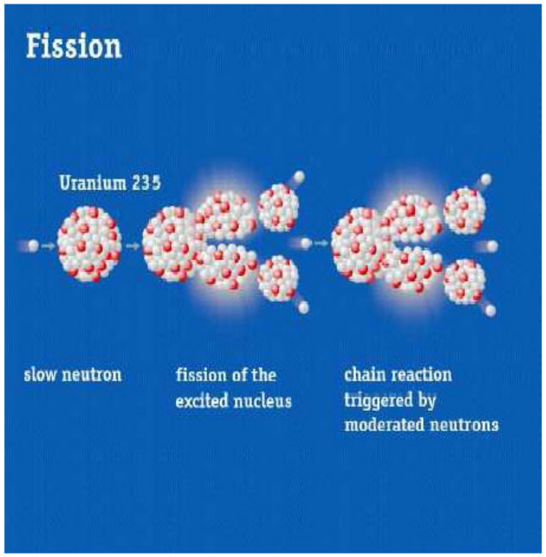
The Neutron – Interactions with Matter

- The neutron interacts with matter in two ways,
  - with nuclei via the strong nuclear force (very short range ~fm 10^{-15}m)
  - with magnetic moments via dipole-dipole coupling, they are able to ‘see’ the unpaired electrons in the material – magnetic neutron scattering



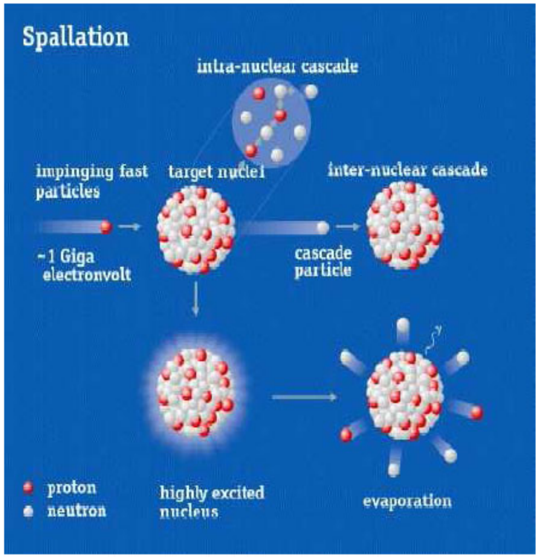
Sources of Neutrons

Fission. A continuous flux of neutrons is produced in the core of a conventional fission reactor. Research reactors with compact cores are used rather than the more abundant nuclear power plants.



$$n + {}^{235}_{92}\mathrm{U} \rightarrow {}^{236}_{92}\mathrm{U} \rightarrow {}^{144}_{56}\mathrm{Ba} + {}^{89}_{36}\mathrm{Kr} + 3n + 177\,MeV$$

Spallation. The spallation target is made from a heavy metal. Protons hitting nuclei in the target trigger an intra-nuclear cascade, placing individual nuclei into a highly excited state. The nuclei then release energy by evaporating nucleons (mainly neutrons), some of which will leave the target, while others go on to trigger further reactions. Each proton delivered to the target results in approximately 15-20 neutrons.



E

The Advantages of Neutrons

Inelastic scattering

Neutrons are able to measure excitations within materials. Neutrons are particularly suitable because both their energy and wavelength can be simultaneously matched to the sample's energy and length scale.

	Temp, T, (K)	Energy $E=k_BT$, (meV)	Wavelength, λ, (Å)
Cold	1-120	0.1-10	4-30
Thermal	60-1000	5-100	1-4
Hot	1000-6000	100-500	0.4-1

Thermal neutrons which have a wavelength (~2 Å) similar to inter-atomic distances also have an energy (20 meV) similar to elementary excitations in solids. Allowing simultaneous information on the structure and dynamics of materials to be obtained.

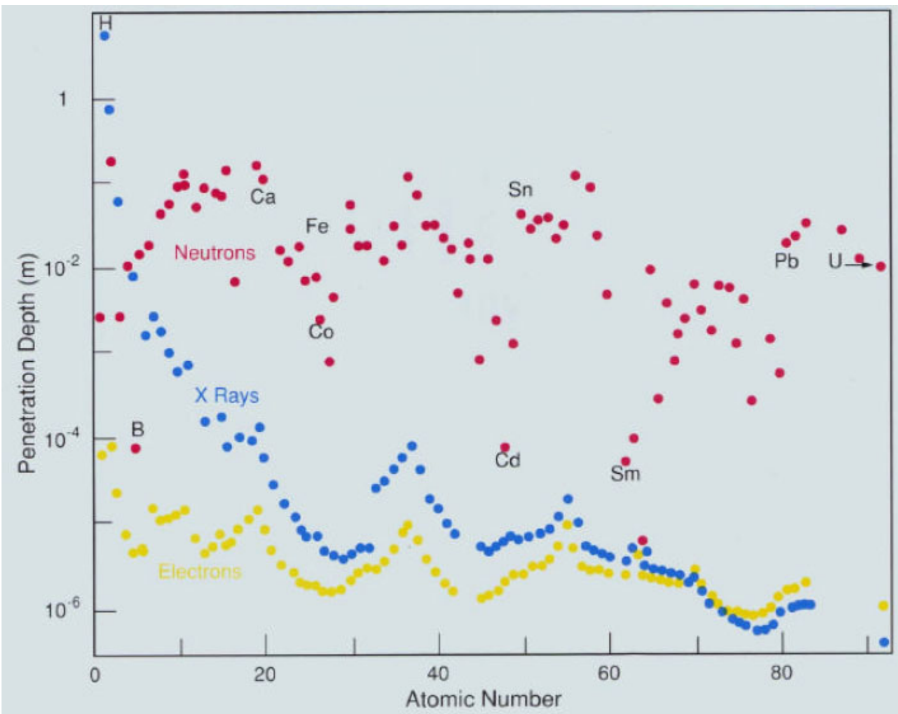
Energy at λ ~ 2 Å

Neutrons	~ 20 meV
Electrons	~ 38 eV
X-ray	~ 6 keV

The Advantages of Neutrons

Weakly interacting probe

The interactions between neutrons and solids is weak, so that neutrons in most cases probe the bulk of the sample, and not only its surface (as is often the case with x-rays and electrons). In addition, quantitative comparisons between neutron scattering data and theoretical models are possible.



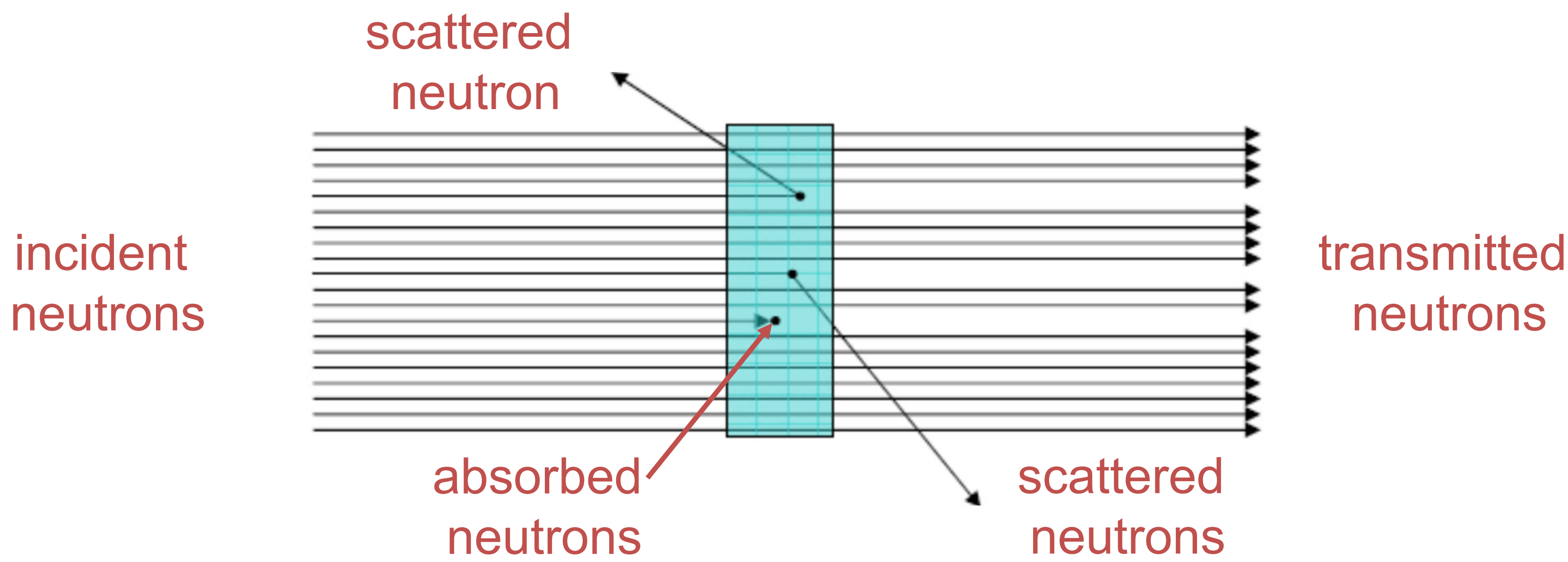
Electrons: very strong interaction with charge dominates

X-rays, light: strong interaction with charge, many orders of magnitude weaker with magnetic moments

Neutrons: weak and comparable strength interactions with nuclei and magnetic fields

How Neutrons Probe

What happens to the neutron?

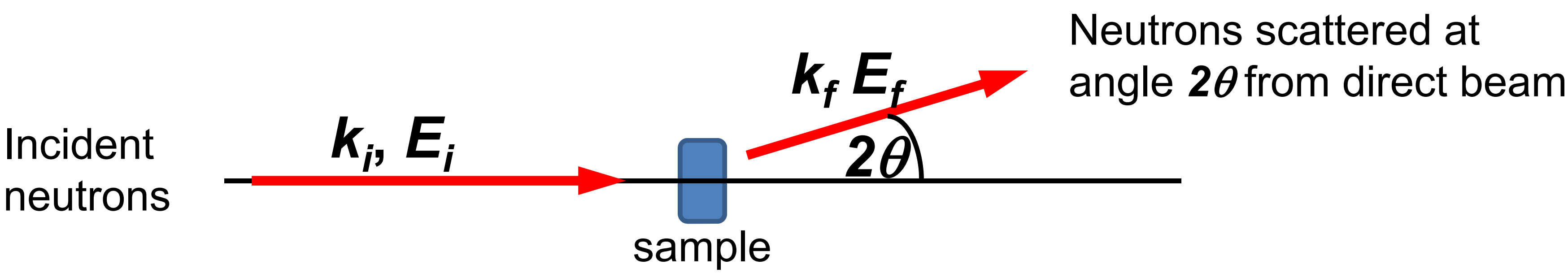


When Neutrons are incident on a sample they can be transmitted, scattered, or absorbed by the sample. We measure the scattered or transmitted beams to obtain information.

Information from transmitted neutron beam
a) Real space imaging

Information from the scattered neutron beam
b) Interference (scattering density distribution $\rho(R)$)

Scattered Neutrons – Basic Concepts



- Neutrons are scattered by the sample, the scattered pattern is a specific function of 2θ characteristic of the sample.
- During the scattering process the neutron energy is either ***unchanged*** or it ***gains or loses energy*** to the sample.
  - The atom can recoil during the collision with the neutron in which case the neutron loses energy and the sample gains energy (eg a phonon).
  - Alternatively if the atom is already moving e.g. a phonon vibration, it gives this energy to the neutron, the neutron gains energy and the sample loses energy.

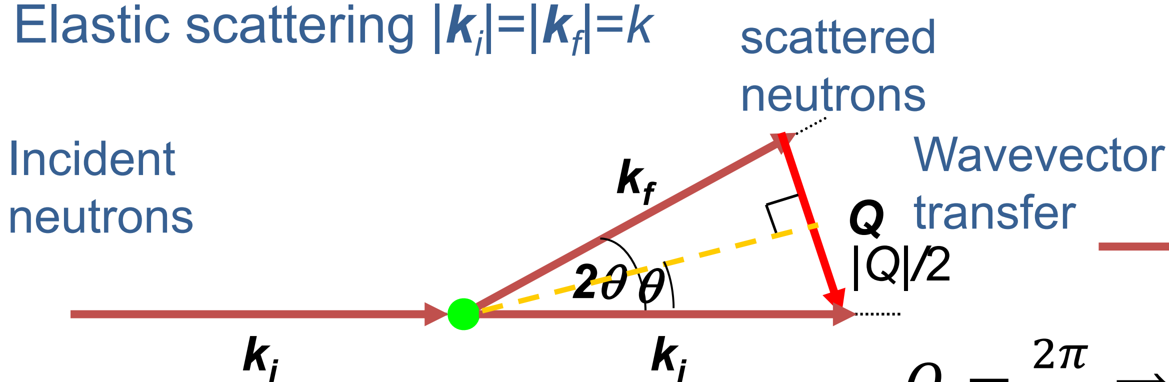
Elastic neutron scattering is when the neutron energy is unchanged. $E_i=E_f$

Inelastic scattering is when the neutron gains or loses energy, $E_i \neq E_f$

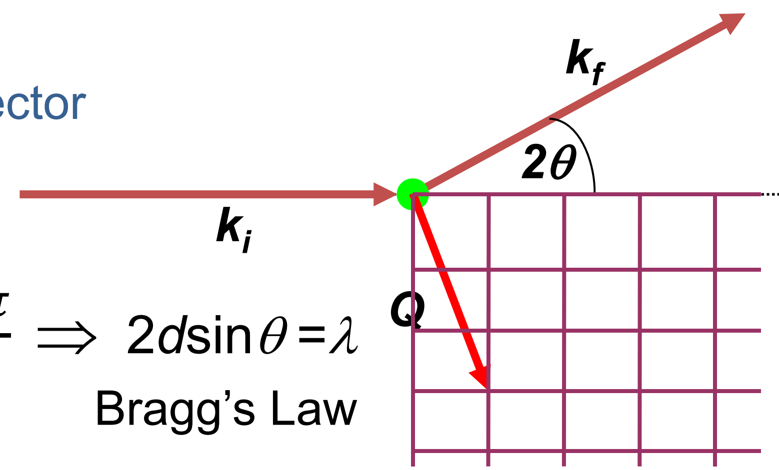
Scattering triangles – Elastic Scattering

- The total energy and momentum are conserved. The total energy lost by the neutron ($\hbar\omega$) equals the energy gained by the sample.
- Energy conservation gives $$E_i - E_f = \frac{1}{2}mv_i^2 - \frac{1}{2}mv_f^2 = \frac{1}{2m}\hbar^2\left(k_i^2 - k_f^2\right) = \hbar\omega$$
- Momentum conservation gives $\hbar\boldsymbol{Q} = \hbar(\boldsymbol{k}_i - \boldsymbol{k}_f)$.
  where $\hbar\boldsymbol{Q}$ is the sample momentum
- ***Q*** is known as the scattering vector $\boldsymbol{Q} = \boldsymbol{k}_i - \boldsymbol{k}_f$
- For elastic scattering the modulus of the wavevectors are equal $|\boldsymbol{k}_i| = |\boldsymbol{k}_f|$ (although they point in different directions)
- The angle 2θ is known as the scattering angle

Elastic scattering $|\boldsymbol{k}_i|=|\boldsymbol{k}_f|=k$



$$\sin\theta = \frac{Q/2}{k} \Rightarrow Q = 2k\sin\theta = \frac{4\pi\sin\theta}{\lambda}$$



$$Q = \frac{2\pi}{d} \Rightarrow 2d\sin\theta = \lambda$$

Bragg's Law

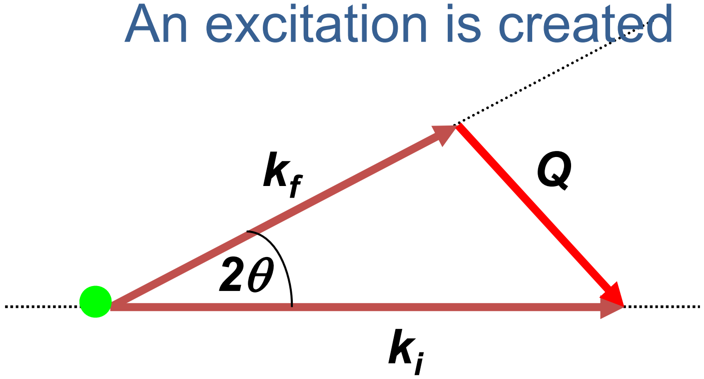
Scattering triangles – Inelastic scattering

- Conservation of energy and momentum

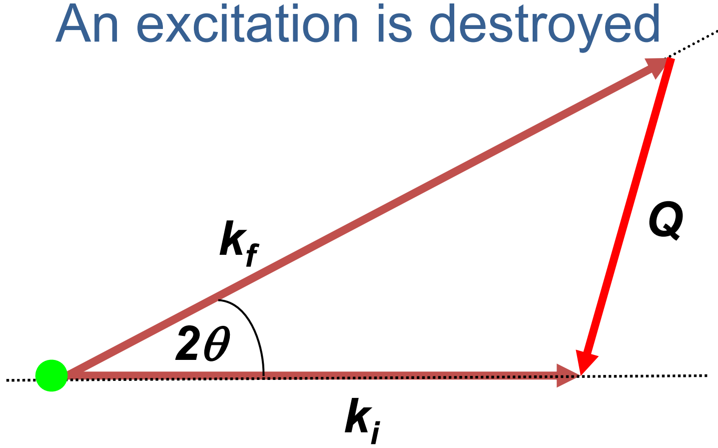
$$E_i - E_f = \frac{1}{2}mv_i^2 - \frac{1}{2}mv_f^2 = \frac{1}{2m}\hbar^2\left(k_i^2 - k_f^2\right) = \hbar\omega \qquad \boldsymbol{Q} = \boldsymbol{k}_i - \boldsymbol{k}_f$$

- For elastic scattering the modulus of the wavevectors are not equal $|k_i| \neq |k_f|$
- Inelastic Scattering triangles

Neutron loses energy
An excitation is created



Neutron gains energy
An excitation is destroyed



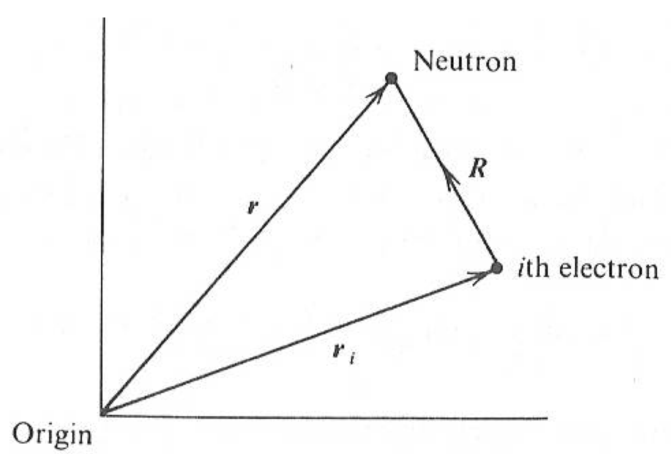
The Magnetic Cross-section

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{\rho_i, s_i} p_{\lambda_i} p_{s_i} \sum_{\rho_f, s_f} \left| \left\langle \boldsymbol{k}_f s_f \rho_f \left| V \right| \boldsymbol{k}_i s_i \rho_i \right\rangle \right|^2 \delta\left(E_{\rho_i} - E_{\rho_f} + \hbar\omega\right) \qquad E = \hbar\omega = \frac{\hbar^2}{2m}\left(k_i^2 - k_f^2\right)$$

V - the magnetic interaction between neutron and electrons

The electrons in an atom possess spin and orbital angular momentum, both of which give rise to an effective magnetic field. The neutrons interact with this field because they possess a spin moment

The interaction between a neutron at point **R** away from an electron with momentum ***l*** and spin ***s*** is



$$V_{magnetic} = -\boldsymbol{\mu}_n . \boldsymbol{B} = \frac{-\mu_0 \gamma \mu_N 2\mu_B}{4\pi} \sum_j \boldsymbol{\sigma} . \left\{ curl\left(\frac{\boldsymbol{s}_j \times \hat{\boldsymbol{R}}_j}{R^2}\right) + \frac{1}{\hbar}\left(\frac{\boldsymbol{l}_j \times \hat{\boldsymbol{R}}_j}{R^2}\right)\right\}$$

$$V_{nuclear} = \frac{2\pi\hbar}{m} \sum_j b_j \delta\left(\boldsymbol{r} - \boldsymbol{r}_j\right)$$

The Magnetic Cross-section

Cross section for spin only scattering by ions

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)=\frac{(\gamma r_0)^2}{2\pi\hbar}\frac{k_f}{k_i}\left[F(\boldsymbol{Q})\right]^2\exp\langle -2W\rangle\sum_{\alpha,\beta}\left(\delta_{\alpha,\beta}-\hat{Q}_\alpha\hat{Q}_\beta\right)S^{\alpha\beta}(\boldsymbol{Q},\omega)$$

$$S^{\alpha\beta}(\boldsymbol{Q},\omega)=\sum_{r_i}\sum_{r_j}\exp\left(i\boldsymbol{Q}.\left(\boldsymbol{r}_i-\boldsymbol{r}_j\right)\right)\int_{-\infty}^{\infty}\left\langle S_{r_i}^{\alpha}(0)S_{r_j}^{\beta}(t)\right\rangle\exp(i\omega t)\,dt$$

For elastic neutron scattering it becomes

$$\left(\frac{d\sigma}{d\Omega}\right)=\frac{(\gamma r_0)^2}{2\pi\hbar}\frac{k_f}{k_i}\left[F(\boldsymbol{Q})\right]^2\exp\langle -2W\rangle\sum_{\alpha,\beta}\left(\delta_{\alpha,\beta}-\hat{Q}_\alpha\hat{Q}_\beta\right)\sum_{r_i}\sum_{r_j}\exp\left(i\boldsymbol{Q}.\left(\boldsymbol{r}_i-\boldsymbol{r}_j\right)\right)\left\langle S_{r_i}^{\alpha}S_{r_j}^{\beta}\right\rangle$$

$F(\boldsymbol{Q})$ Magnetic form factor which reduces intensity with increasing wavevector

$\exp\langle -2W\rangle$ Debye-Waller factor which reduces intensity with increasing temperature

$\left(\delta_{\alpha,\beta}-\hat{Q}_\alpha\hat{Q}_\beta\right)$ polarisation factor which ensures only components of spin perpendicular to Q are observed

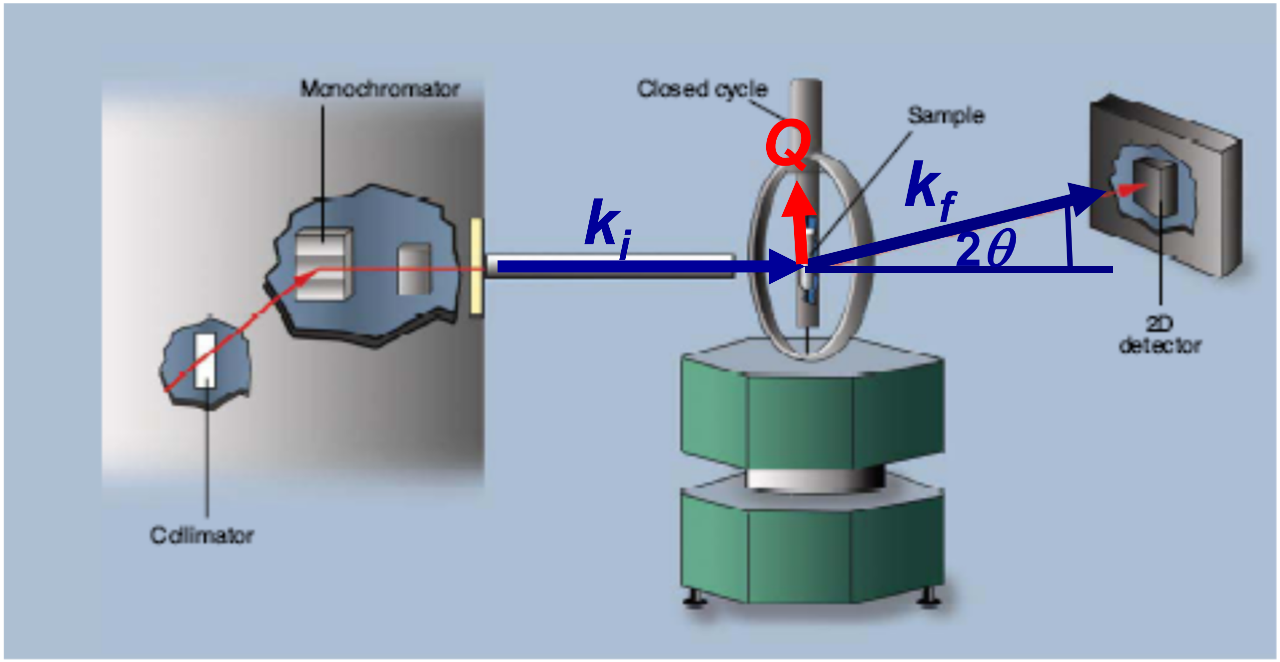
$\left\langle S_{r_i}^{\alpha}(0)S_{r_j}^{\beta}(t)\right\rangle$ is the spin-spin correlation function which describes how two spins separated in distance and time a related

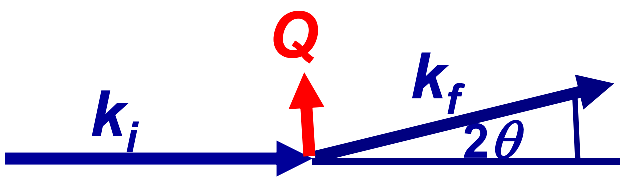
Neutron Scattering Techniques:

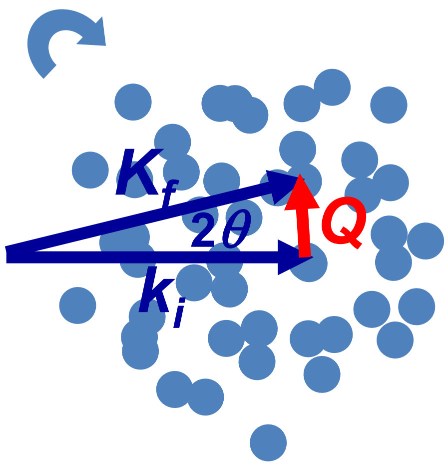
Neutron Diffraction

Neutron Diffraction - Single-Crystal Diffractometer

Fixed wavelength λ

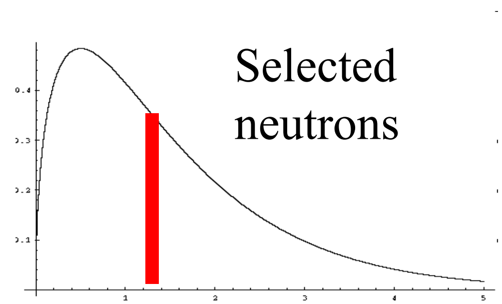


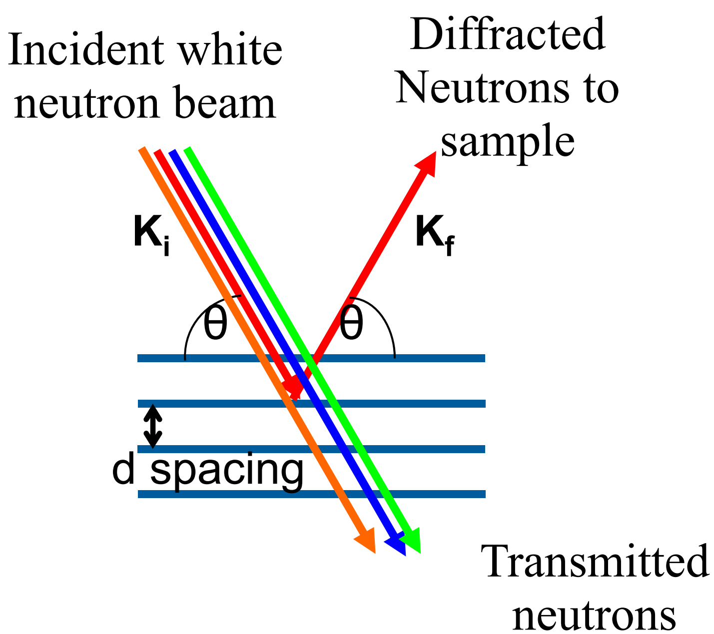




Monochromator

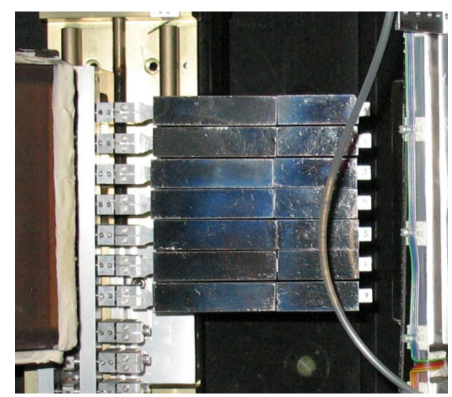
The monochromator is a crystalline material and selects a single wavelength from the white neutron beam of the reactor/spallation source by Bragg scattering where the scattering angle is chosen to select λ.





$$2d \sin\theta = n\lambda$$
$$n = 1,2,3\ldots.$$

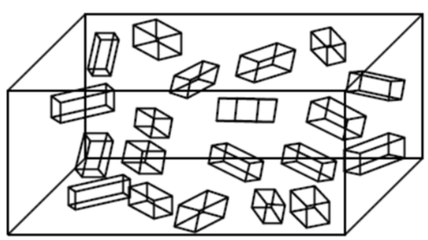
Vertically focusing monochromator

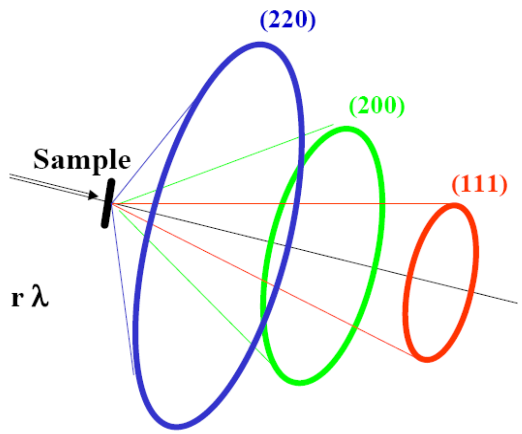


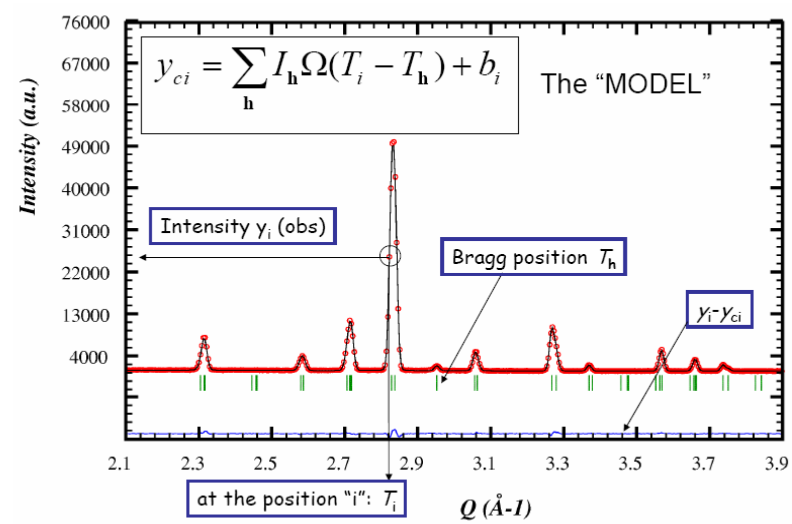
from graphite, Copper, Germanium, blades can be focused

Neutron Diffraction - Powders

- A powder consists of many very small single crystals or crystallites
- All orientations are present.
- Typical volume 1 -10μm





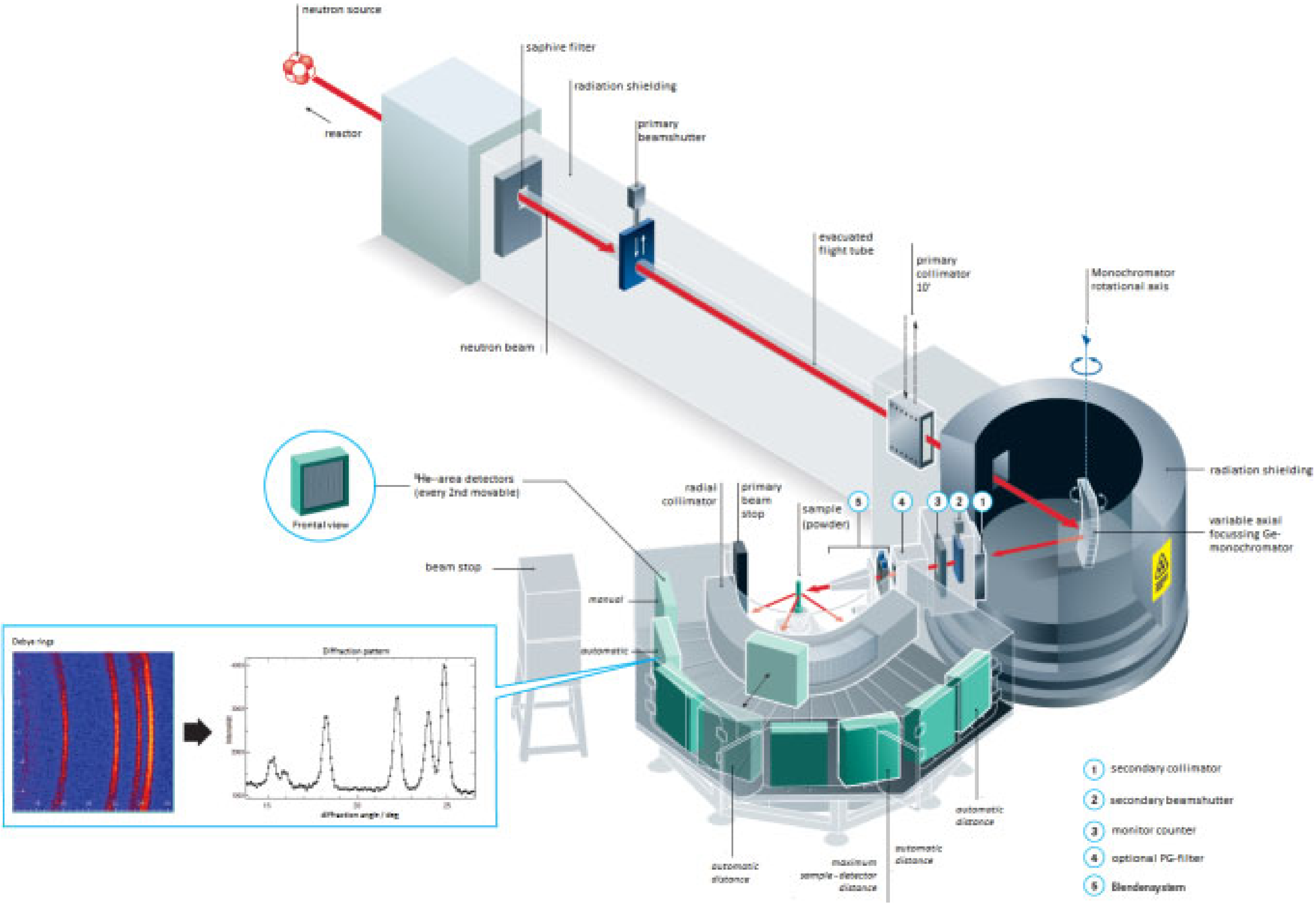


- Neutrons of fixed wavelength λ are scattered into Debye-Scherrer cones of fixed 2θ.
- A scan of either Q or 2θ can be used to measure all the Bragg peaks d= λ/(2sin2θ)
- A powder is measured using a powder diffractometer

Rietveld refinement –

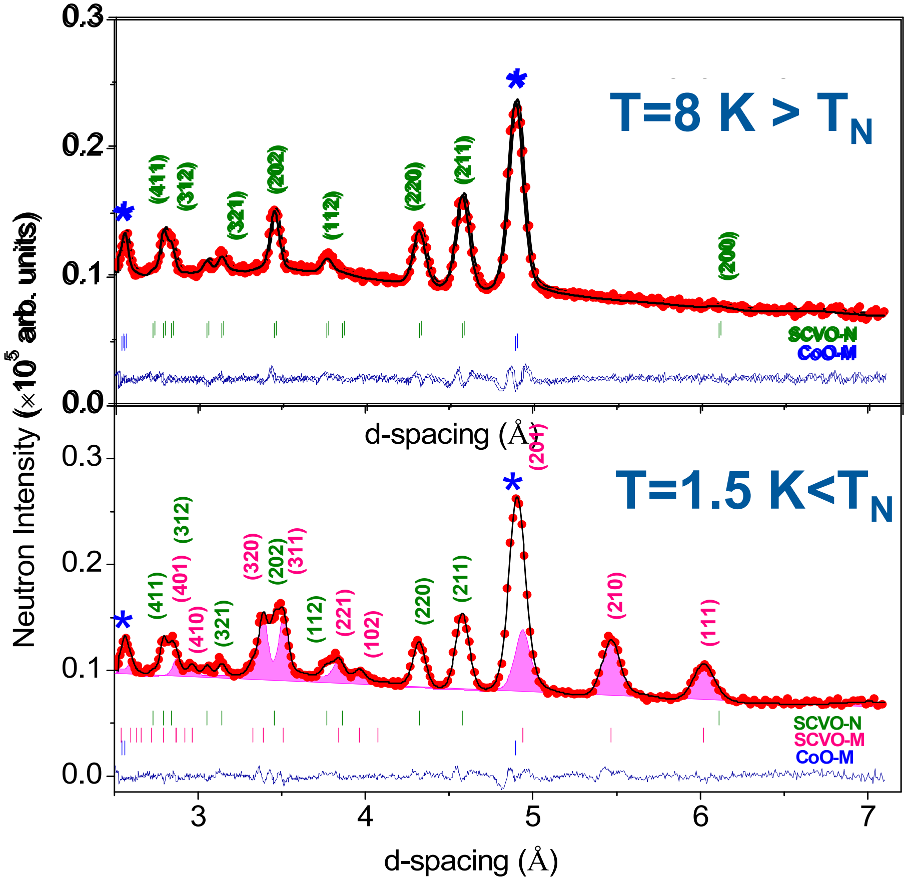
- the powder pattern of a model structure is calculated and compared to the data
- The model is varied iteratively until it matches the data

Neutron Diffraction - Powder Diffractometer



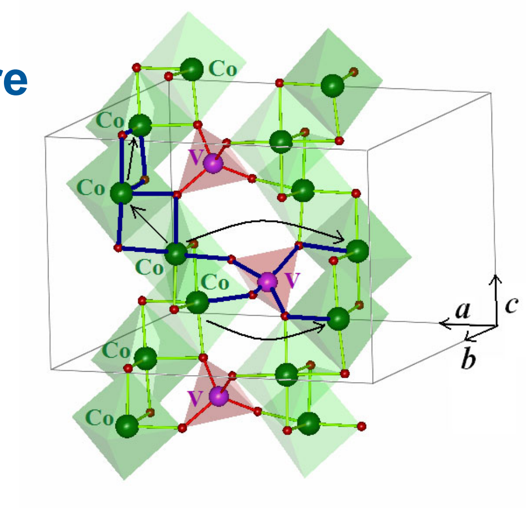
Neutron Diffraction of $SrCo_2V_2O_8$ T_N=5.2K

Neutron Diffraction

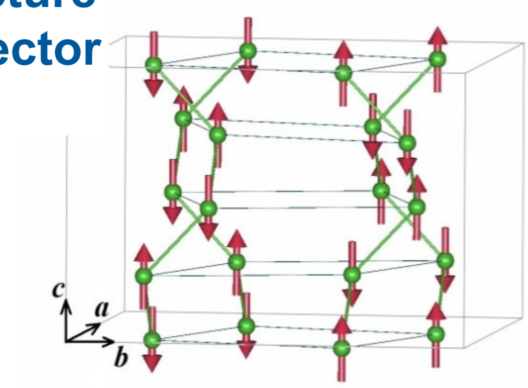


A. K. Bera, B. Lake et. al.,
Phys. Rev. B 89, 094402 (2014)

Crystal Structure
Space Group
$I4_1cd$



Magnetic structure
Propagation vector
k = (0 0 1)

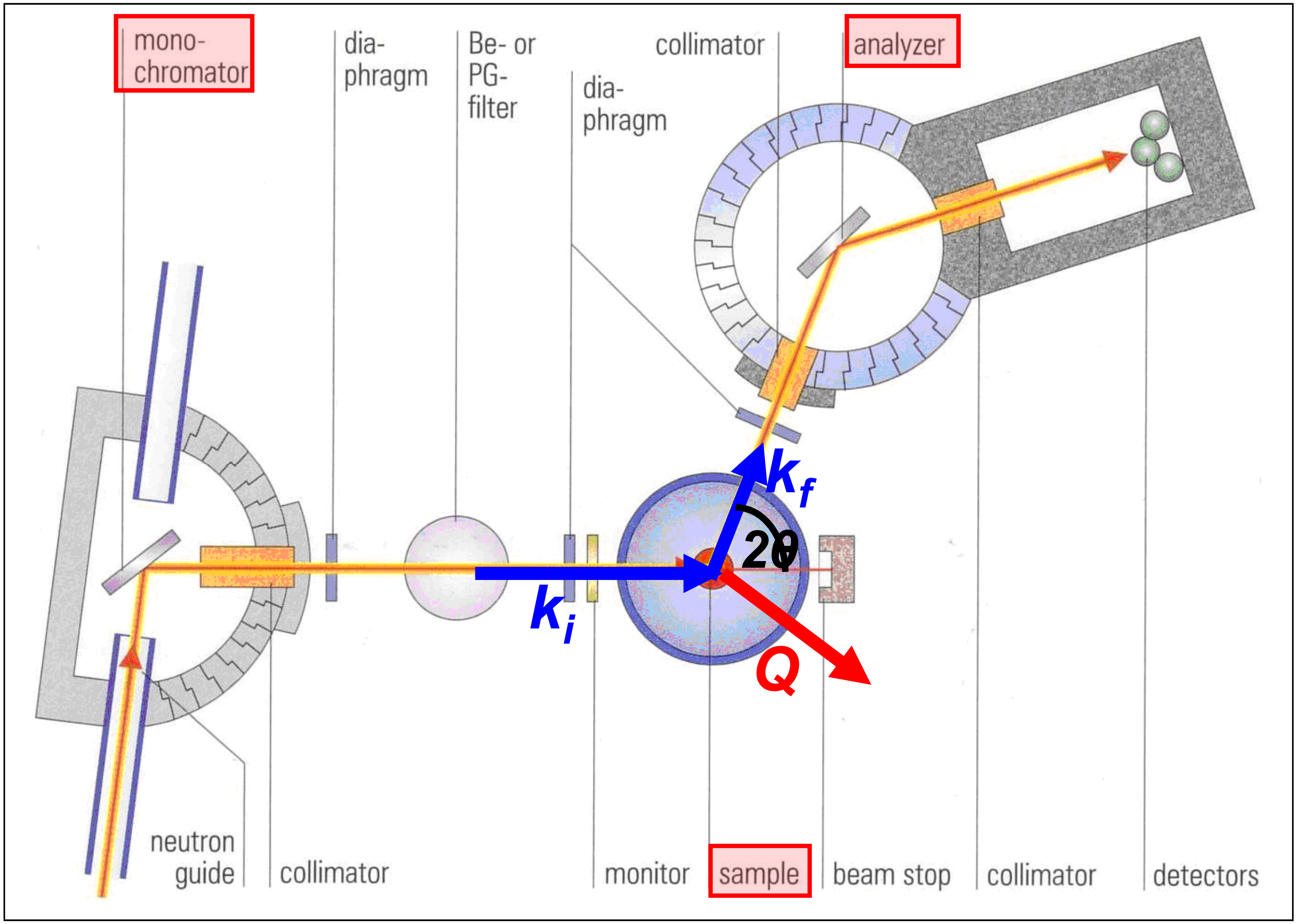


- AFM chains along *c* axis.
- Antiferro-/ferromagnetic along *a/b* axis
- Spins along c axis

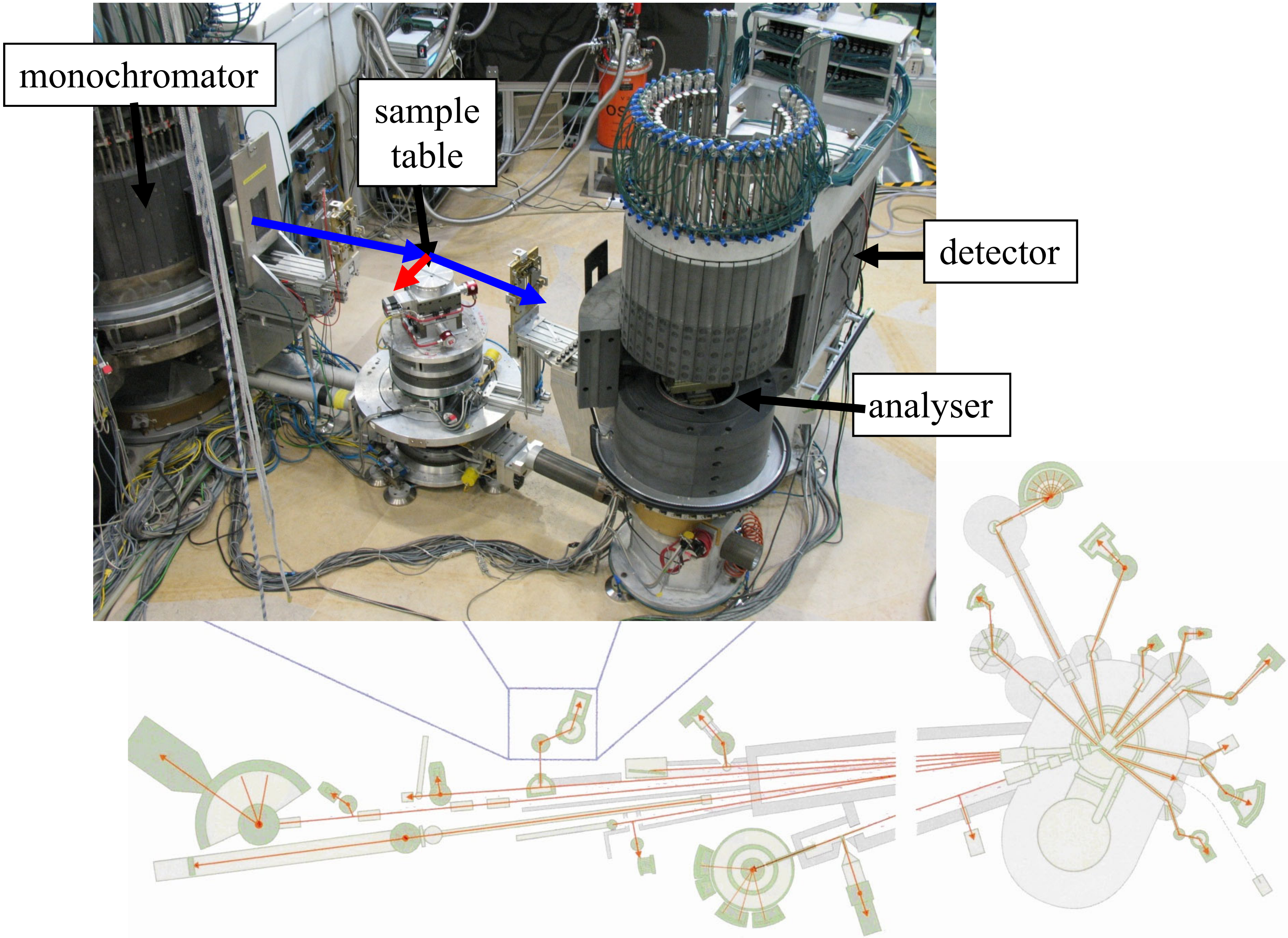
Neutron Scattering Techniques:

Inelastic Neutron Scattering

Inelastic Neutron Scattering - Triple Axis Spectrometer

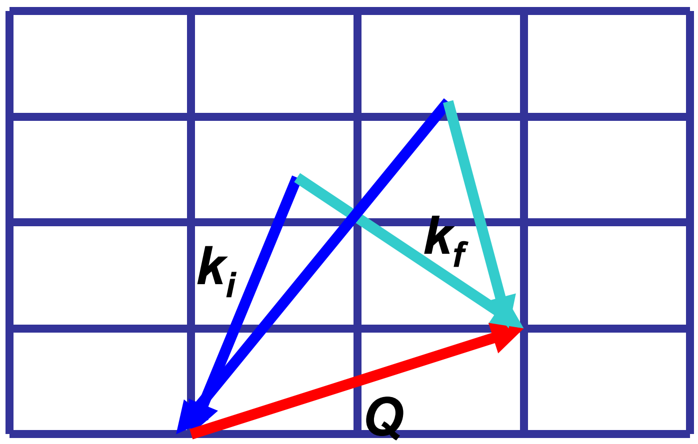


Inelastic Neutron Scattering - Triple Axis

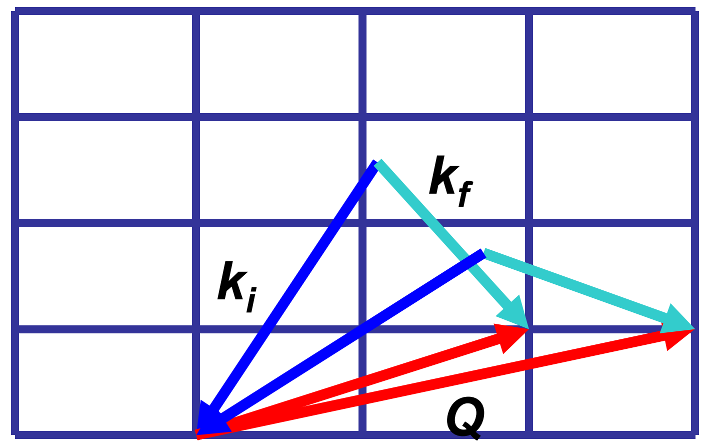


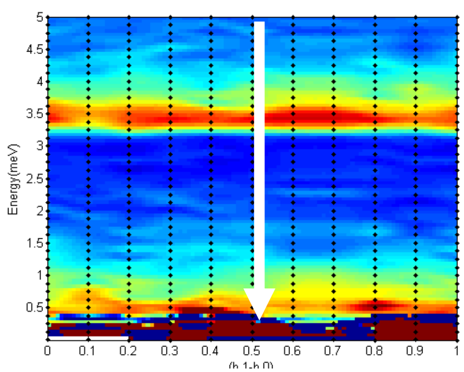
Inelastic Neutron Scattering - Triple Axis Spectrometer

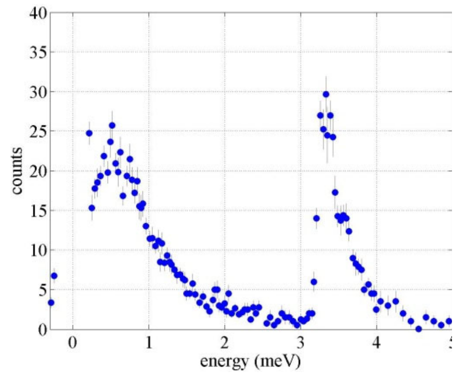
Keep wavevector transfer constant and, scan energy transfer.

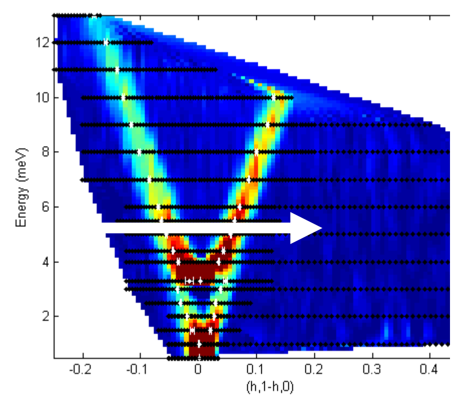


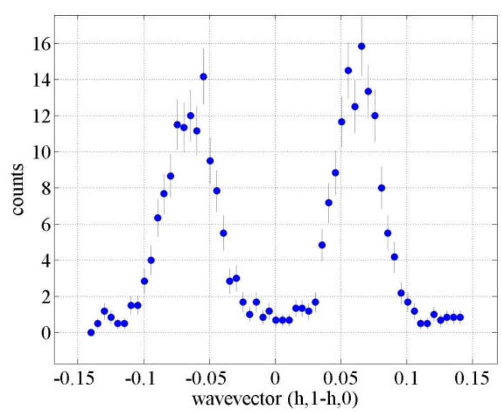
Keep energy transfer constant and, scan wavevector transfer.







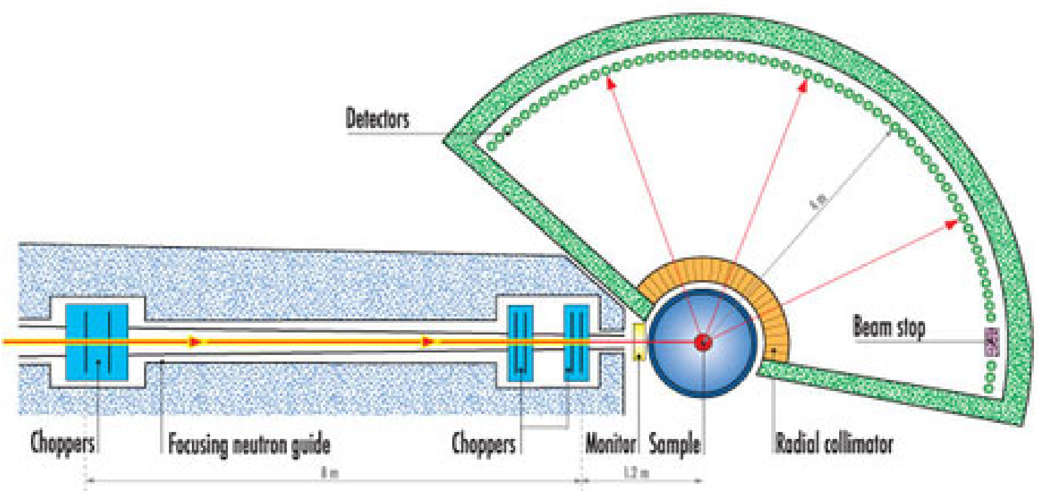


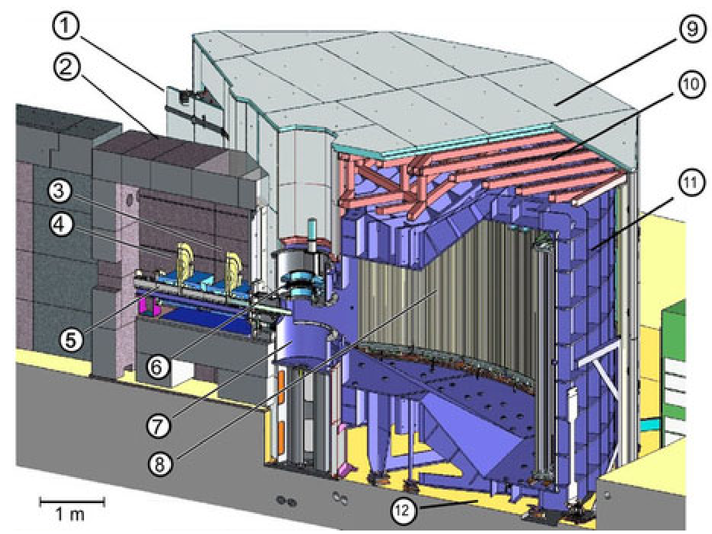


B. Lake; TMS, Aug 2022

Inelastic Neutron Scattering - Time of Flight Spectrometer

Time and distance are used to calculate the initial and final neutron velocity and therefore energy. This is achieved by cutting the incident beam into pulses to give an initial time and incident energy





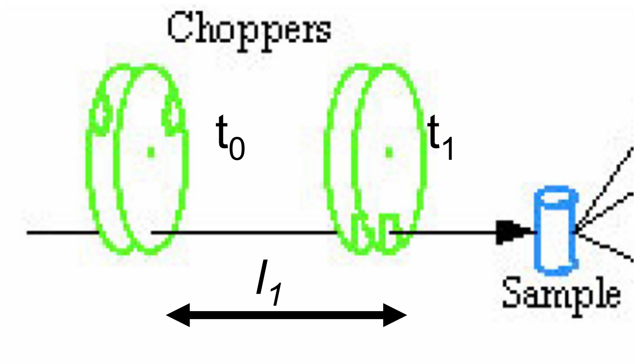
IN5, ILL

Inelastic Neutron Scattering - Time of Flight Spectrometer

The neutron beam is cut into pulses of neutrons using rotating disk choppers.

I[st] chopper lets neutrons through once per revolution and sets initial time t_0

2[nd] chopper rotates at the same rate and opens at a specific time later. The phase is chosen to select initial neutrons of a specific velocity and energy.



$$v_i = \frac{l_1}{(t_1 - t_0)}$$

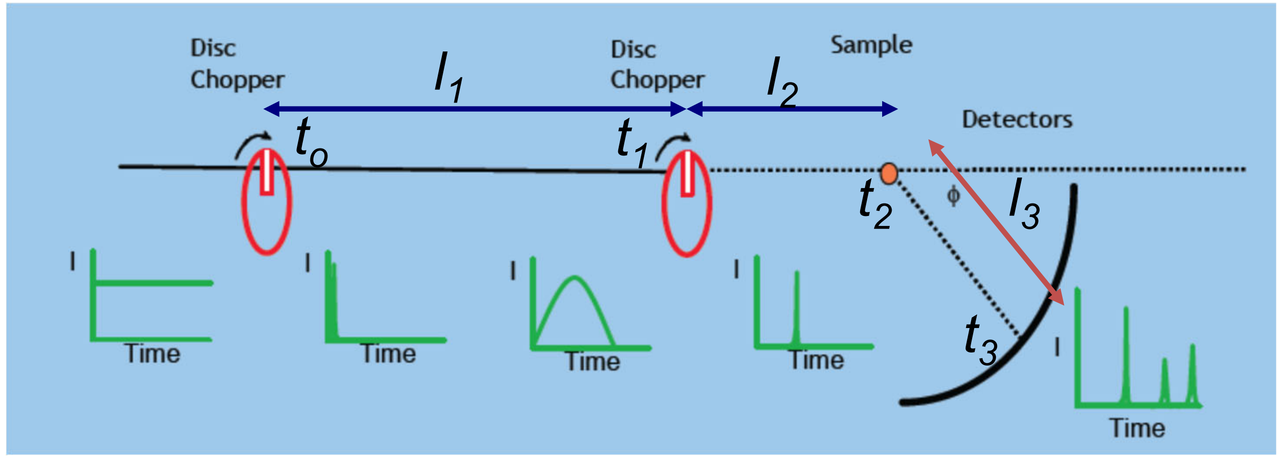
$$E_i = \frac{mv^2}{2} = \frac{ml_1^2}{2(t_1 - t_0)^2}$$

After scattering at the sample the detector again measures time as well as number of neutrons, thus the velocity and energy of the scattered neutrons is known.

Inelastic Neutron Scattering - Time of Flight Spectrometer

$$E_i = \frac{m l_1^2}{2(t_1 - t_0)^2}$$

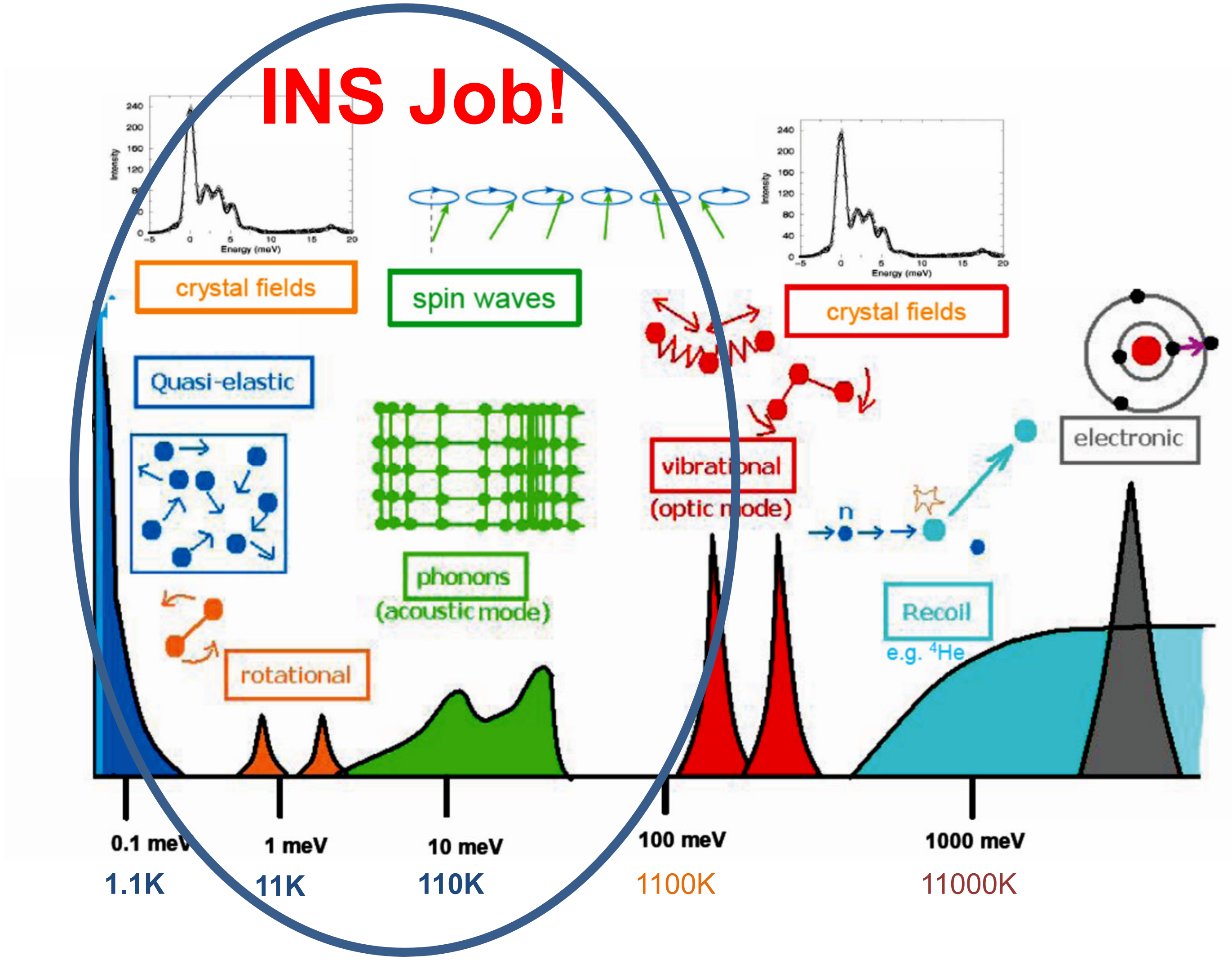
$$E_f = \frac{m (l_3)^2}{2(t_3 - t_2)^2}$$



- First chopper sets the initial time.
- Second chopper sets the initial energy
- Detectors measure final time and energy.

Spin-Wave Excitations

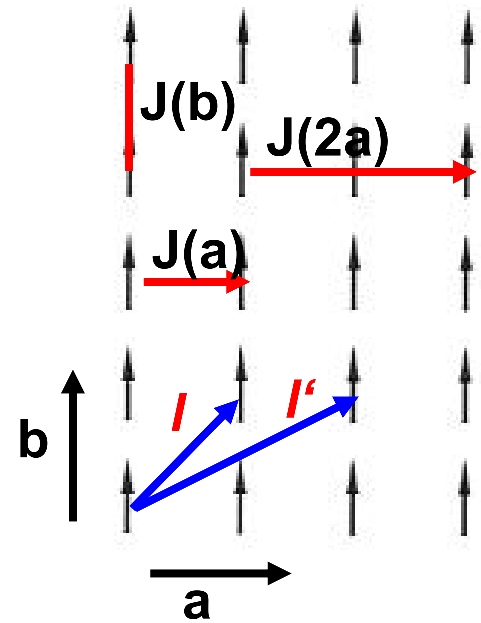
Excitations in Condensed Matter



Spin-Waves

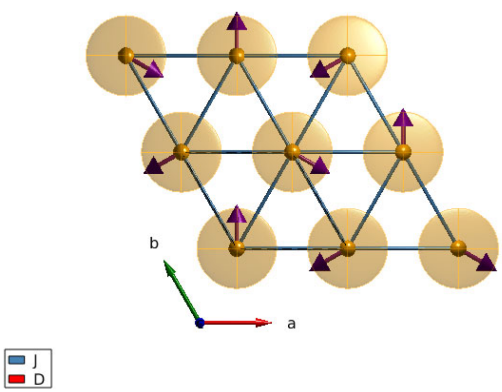
magnetic materials with Hamiltonians such as

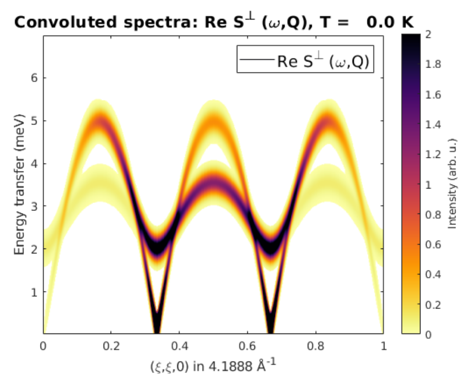
$$H = -\sum_{l,l'} J(\boldsymbol{l}-\boldsymbol{l}')\boldsymbol{S}_{l}.\boldsymbol{S}_{l'}$$



Assumption of fully aligned ground state
Excitations are fluctuations about this order

They can be calculated by diagonalizing the Hamiltonian to find eigenstates and eigenvalues.





Spin-waves have quantum spin number S=1 and have a well-defined energy as a function of wavevector

http://spinw.org/

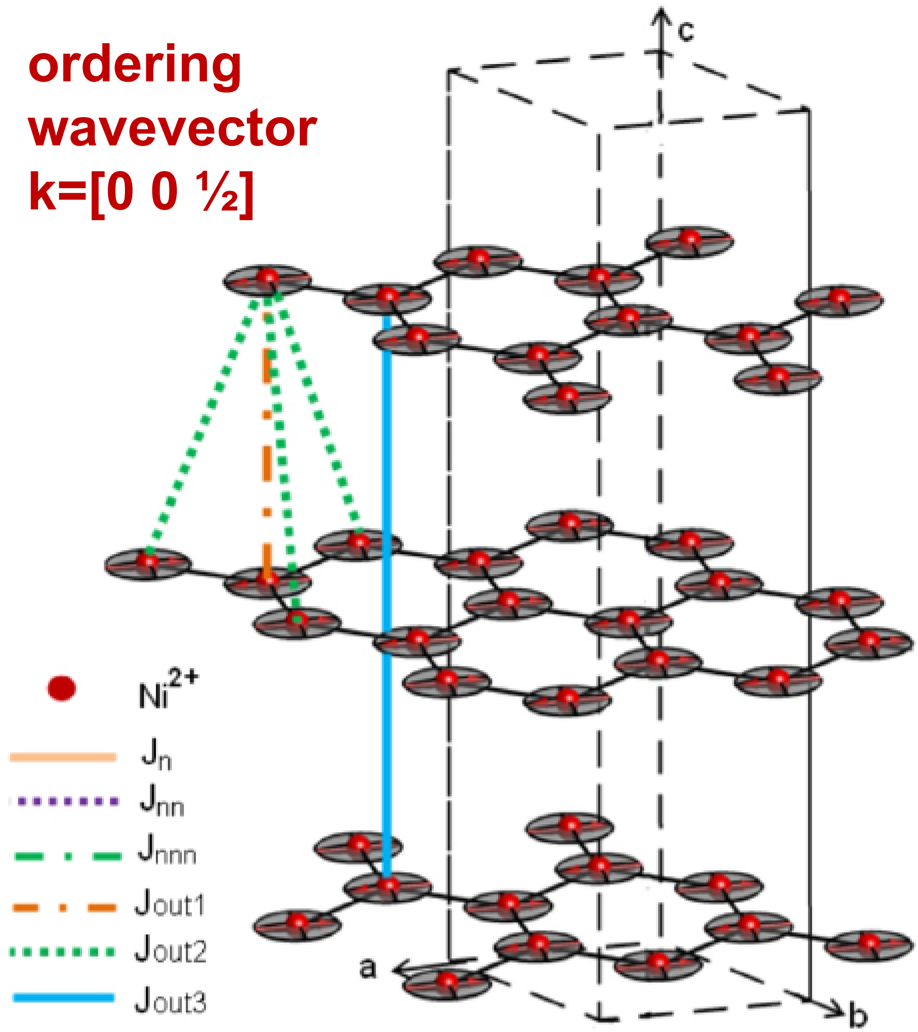
S. Toth and B. Lake,
B. La *J. Phys. Condens. Matter 27, 166002 (2014)*

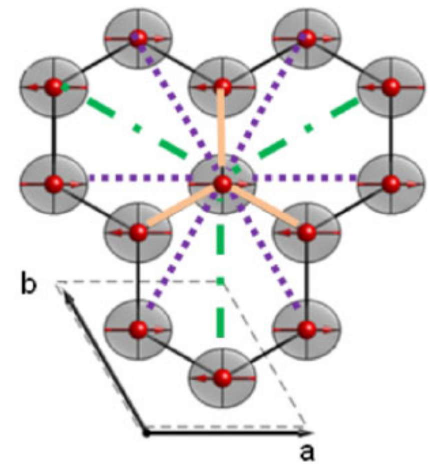
Spin
By Sandor Toth

Spin-Waves in $BaNi_2V_2O_8$

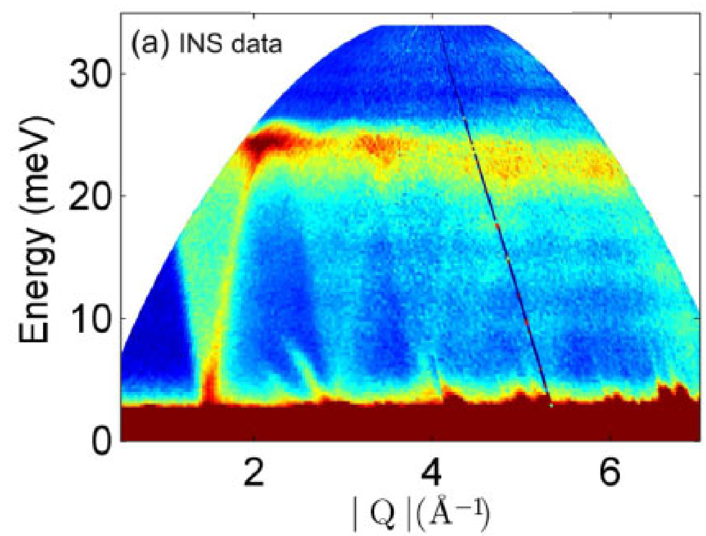
$$H = \sum_{i>j} J_n \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} J_{nn} \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} J_{nnn} \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} J_{out} \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} D_{EP} \cdot {S_i^c}^2 + \sum_{i>j} D_{EA} \cdot {S_i^a}^2$$

ordering wavevector k=[0 0 ½]



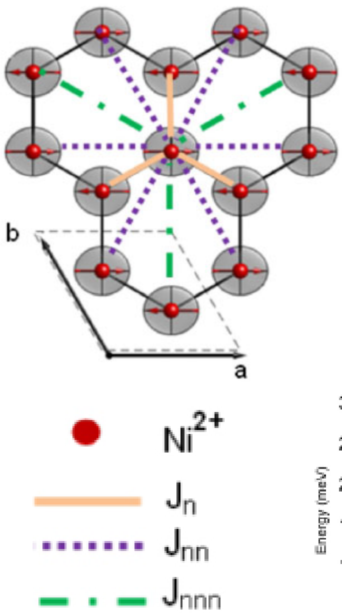


TOF, MERLIN, ISIS

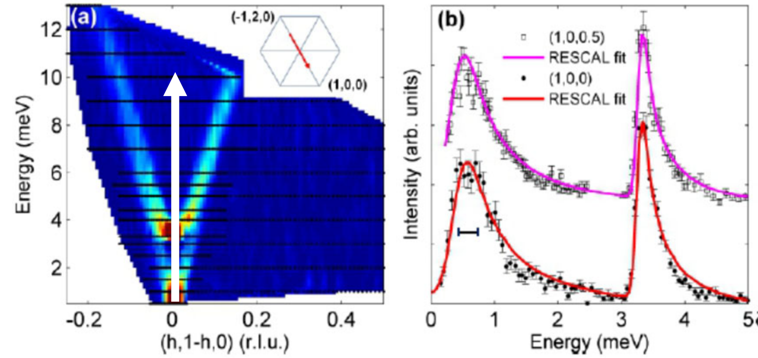


B. Lake; TMS, Aug 2022

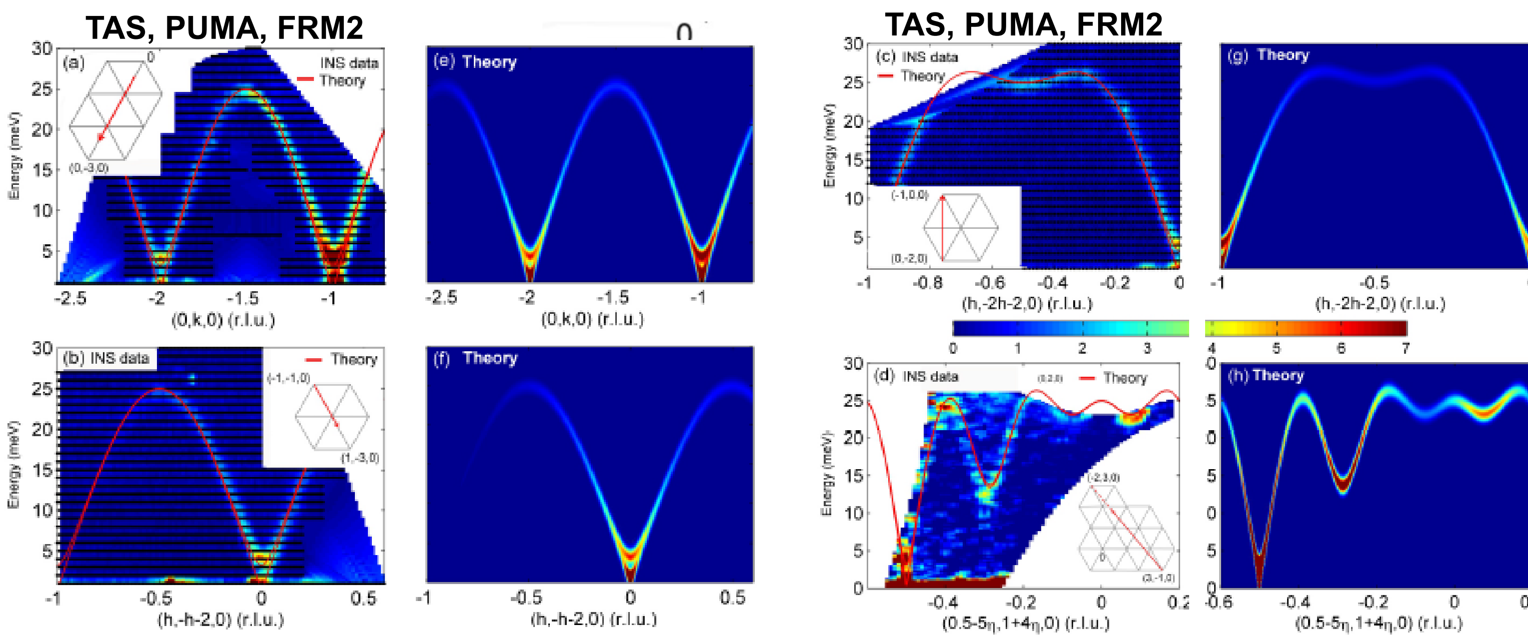
Spin-Waves in $BaNi_2V_2O_8$



$$H=\sum_{i>j}J_n\cdot S_i\cdot S_j+\sum_{i>j}J_{nn}\cdot S_i\cdot S_j+\sum_{i>j}J_{nnn}\cdot S_i\cdot S_j+\sum_{i>j}J_{out}\cdot S_i\cdot S_j+\sum_{i>j}D_{EP}\cdot S_i^{c^2}+\sum_{i>j}D_{EA}\cdot S_i^{a^2}$$



k=[0 0 ½]



1st neighbor 10.9meV< J_n <11.8meV

2nd neighbor 1.1meV< J_{nn} <0.65meV

3rd neighbors -0.1meV< J_{nnn} <0.4meV

Interplane coupling J_{out} <0.0001meV

Easy-plane anisotropy 0.8< D_{EP} <0.73

Easy-axis anisotropy -0.0011< D_{EA} <-0.0009

Summary

Conventional magnets

Long-range magnetic order and spin-wave excitations

Neutron scattering concepts

Neutron propteries, neutron sources,
neutron scattering triangles and cross sections

Neutron scattering techniques

Neutron diffracion, inelastic neutron scattering TAS TOF

Spin-waves

Calculations are measurements

Next Lecture

Unconventional magnets and neutron scattering

Neutron scattering as a tool to study quantum magnets

Bella Lake
Helmholtz Zentrum Berlin, Germany
Berlin Technical University, Germany

Outline

Quantum magnets

Neutron scattering study of

Example 1 Zero-dimensional quantum magnet

Example 2 One-dimensional quantum magnet

Frustrated magnets

Neutron scattering study of

Example 3 Two-dimensional quantum magnet

Quantum Magnets

The Origins of Quantum 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

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

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

Quantum Magnets Characterised by

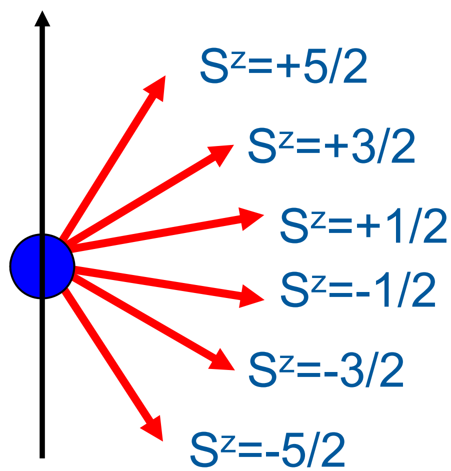
- $T_N << T_{CW}$ and $<S> < S$, or no order.
- The excitations are broadened and renormalised
- Unusual quantum numbers, new theoretical approaches

Quantum Magnetism - Low Spin Value

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

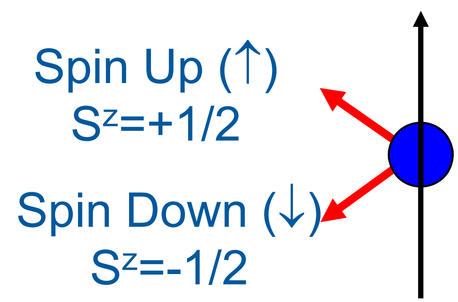
S=5/2

$|\mathbf{S}|^2 = S(S+1) = 35/4$

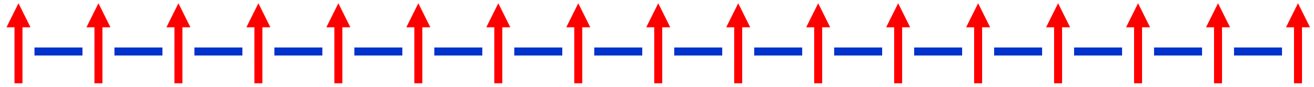


S=1/2

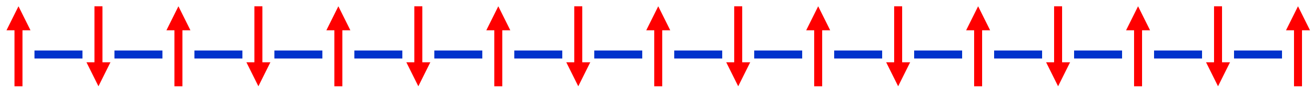
$|\mathbf{S}|^2 = S(S+1) = 3/4$



Antiferromagnetic Exchange Interactions

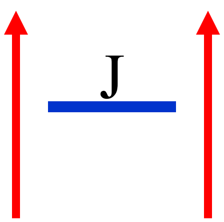


- Parallel spin alignment is an eigenstate of the Hamiltonian



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

J>0
ferromagnetic



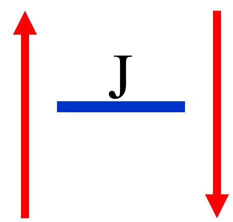
$S_1=1/2$ $S_2=1/2$
$S^z=1/2$ $S^z=1/2$

$$H_{1,2}=J\left(S_1^+S_2^-+S_1^-S_2^++S_1^zS_2^z\right)$$

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

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

J>0
antiferromagnetic



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

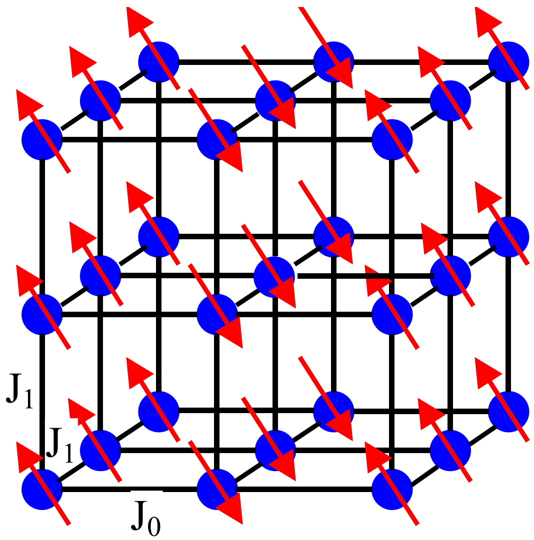
Low-Dimensional Interactions

For 3-dimensional each magnetic ion has 6 neighbours

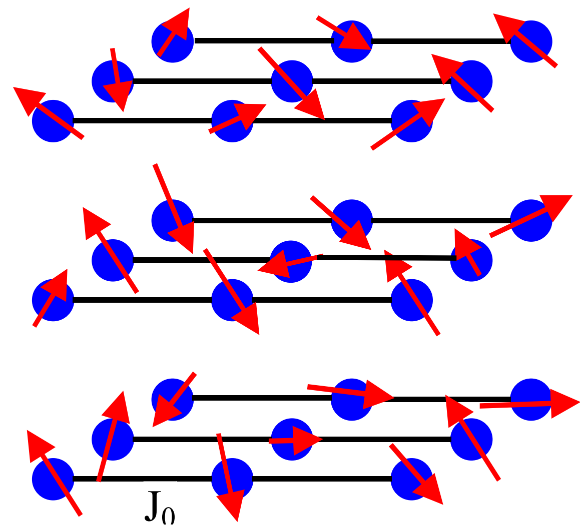
For a 1-dimensional there are only 2 neighbours

Neighbouring ions stabilize long-range order and reduce fluctuations

3D S=1/2



1D S=1/2



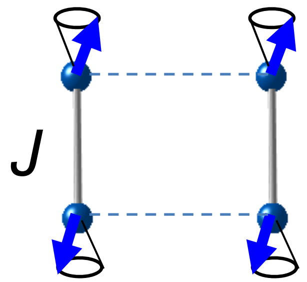
Example 1
Zero Dimensional
Quantum Magnets

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

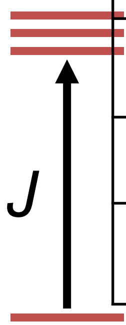
Dimer Unit

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

$$\hat{H} = J\hat{S}^a \cdot \hat{S}^b$$



Excitations

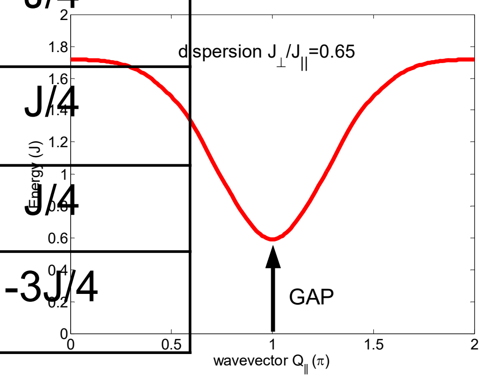


Eigenstates	S_T	S^z	Eigenvalue
$\|1,+1\rangle = \|\uparrow_1\uparrow_2\rangle$	1	1	J/4
$\|1,0\rangle = \frac{1}{\sqrt{2}}(\|\uparrow_1\downarrow_2\rangle + \|\downarrow_1\uparrow_2\rangle)$	1	0	J/4
$\|1,-1\rangle = \|\downarrow_1\downarrow_2\rangle$	1	-1	J/4
$\|G\rangle = \frac{1}{\sqrt{2}}(\|\uparrow_1\downarrow_2\rangle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

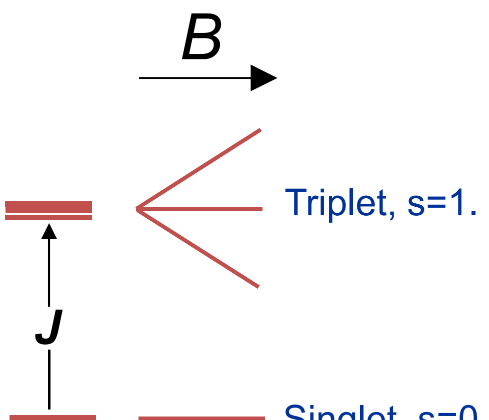
Triplet, S=1

Singlet, S=0

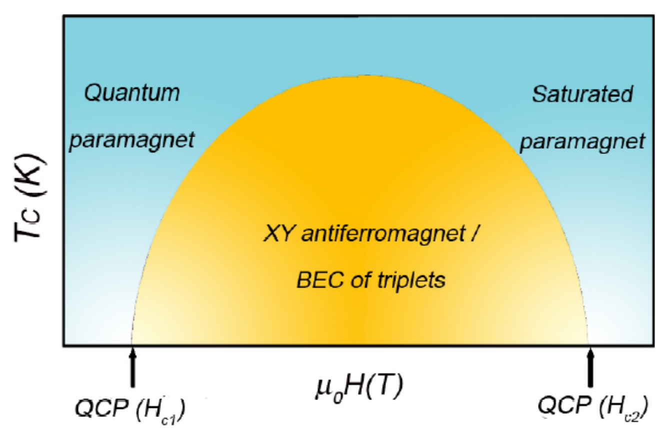
Interdimer coupling, gapped dispersive Mode



Zeeman Splitting in Field



Bose Einstein Condensation



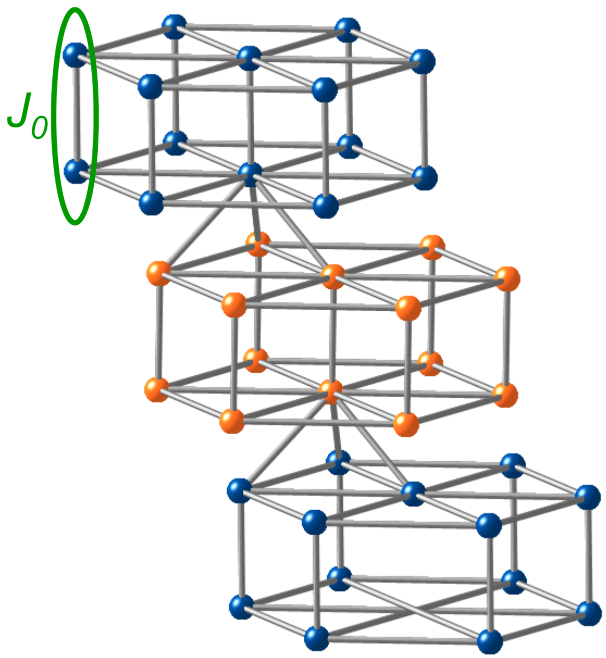
Properties:

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

$Sr_3Cr_2O_8$ –Spin-1/2, Dimer AF

$Sr_3Cr_2O_8$ → Cr^{5+}, Spin-1/2.

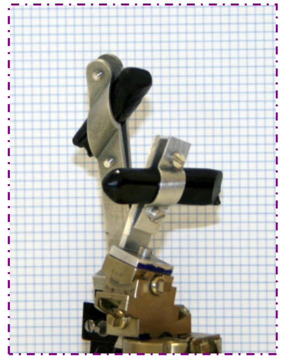
Space group - R-3m



Dimer coupling is bilayer J_0

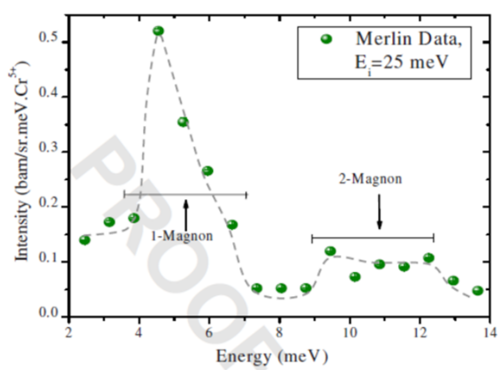
$Sr_3Cr_2O_8$ is 3D network of dimers

B. Lake; TMS, Aug 2022

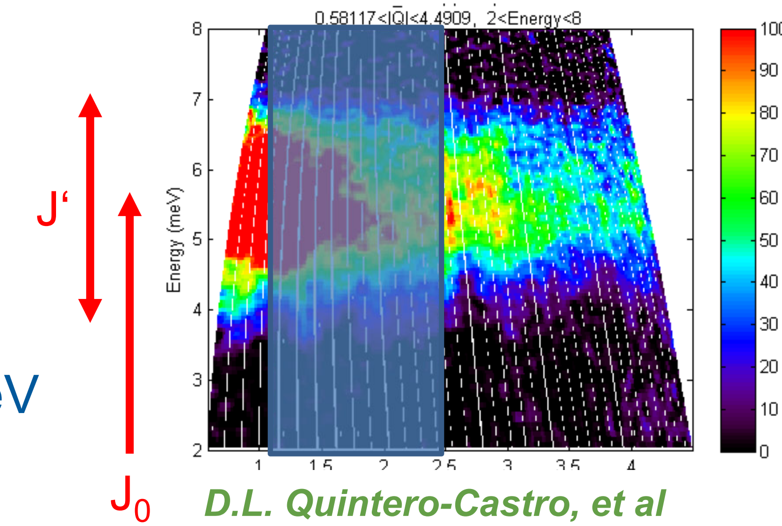


E_{gap}=3.4meV
E_{upper}=7.10meV

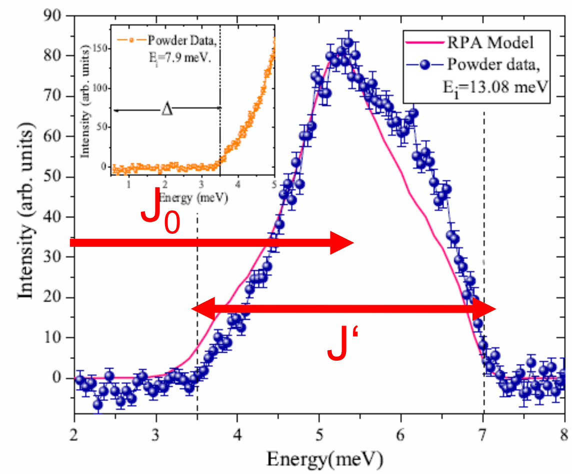
$E_{midband}$~J_0=5.5meV
$E_{bandwidth}$~J‘=3.7meV



Powder inelastic neutron scattering

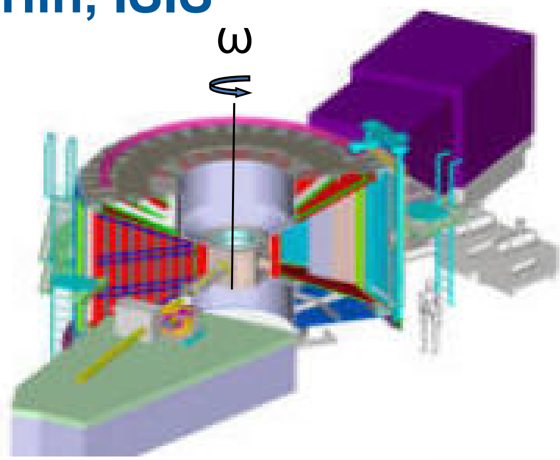


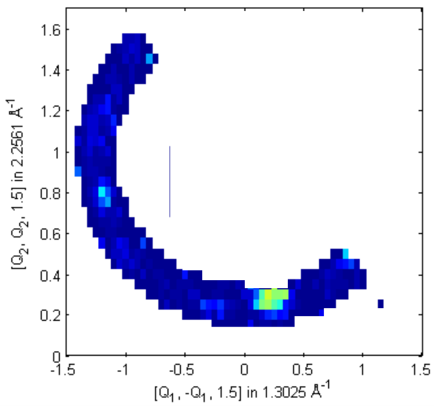
D.L. Quintero-Castro, et al Phy. Rev. B. 81, 014415 (2010)



Single Crystal Inelastic Neutron Scattering

Merlin, ISIS



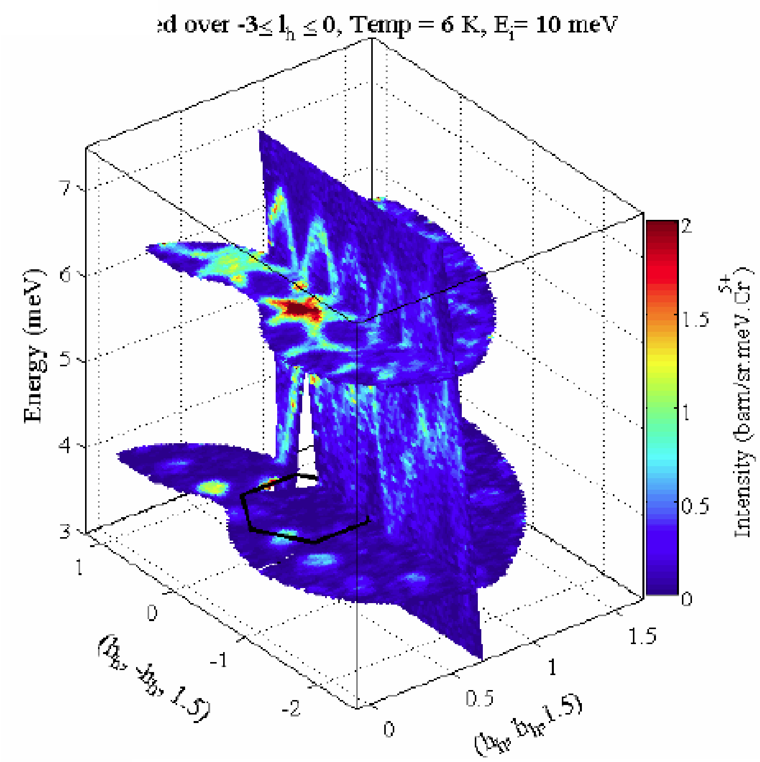


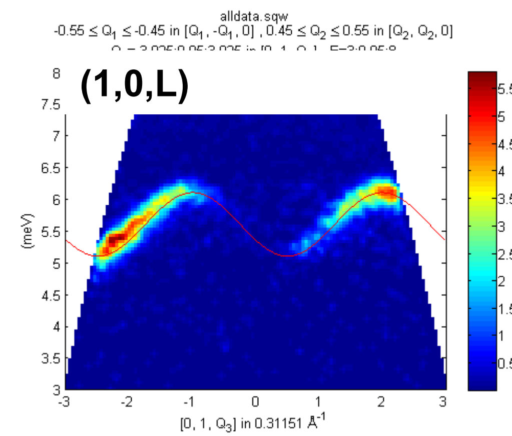
Individual scans combined to create a single file $S(Q_h, Q_k, Q_l, E)$.

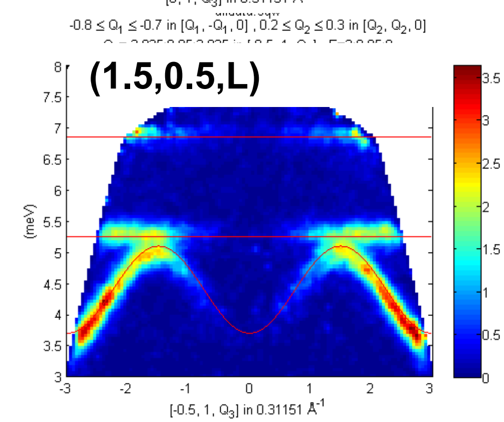
large region of the energy and reciprocal space.

detectors:
180° horizontal
±30° vertical

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





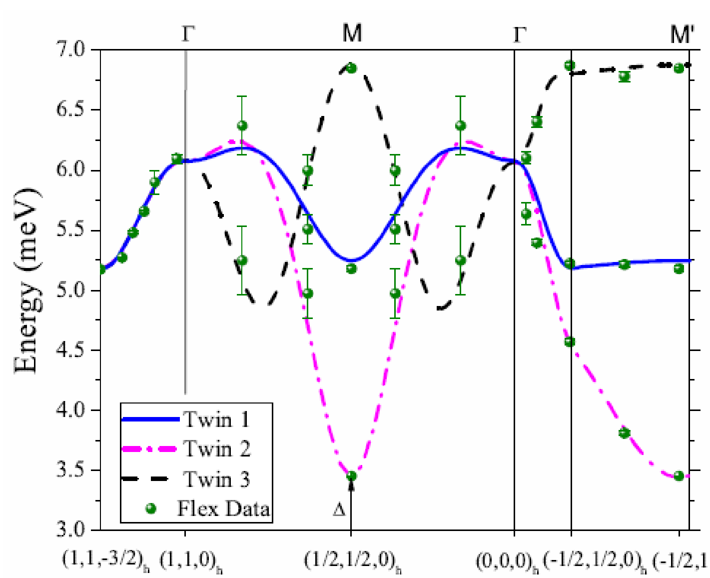


B. Lake; TMS, Aug 2022

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

Fitting to a Random Phase Approximation

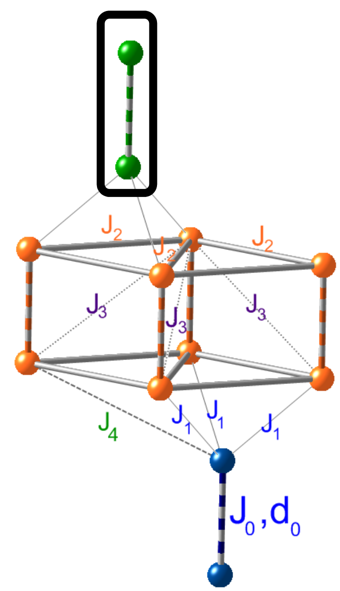
Extracted Dispersions



Random Phase Approximation

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

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



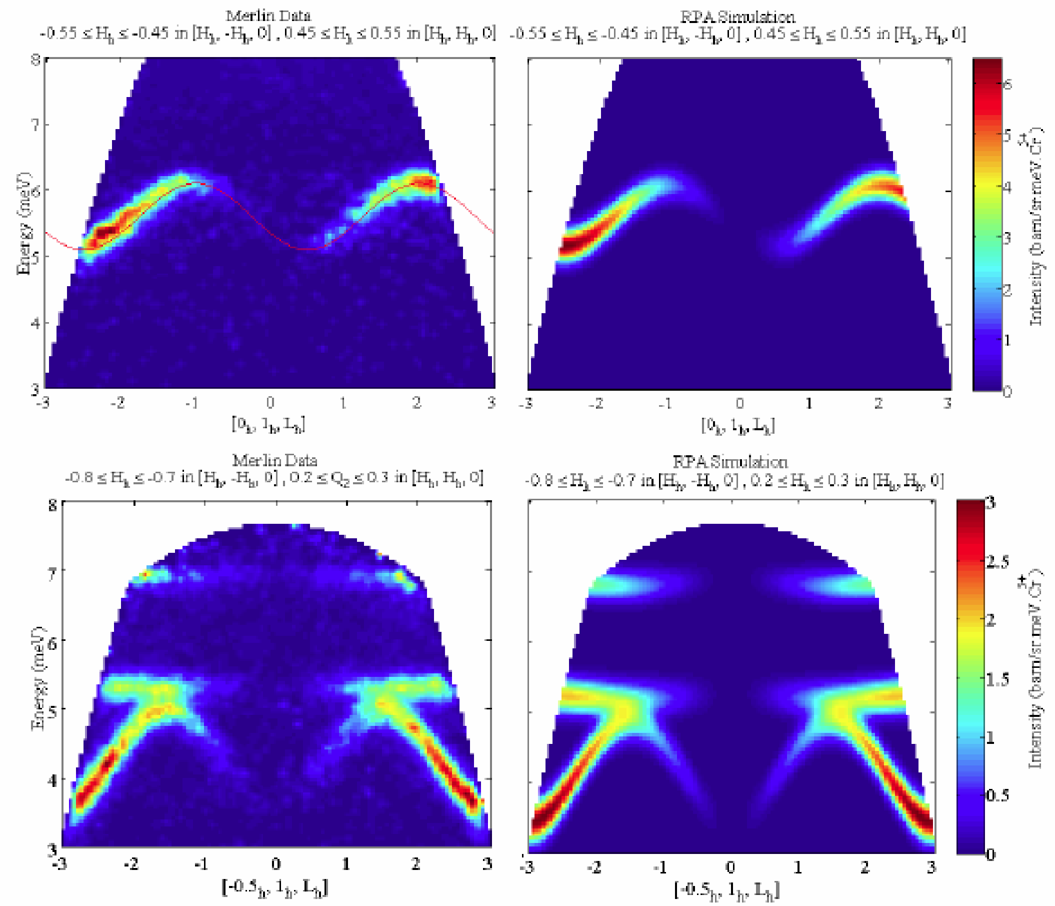
Constants	$Sr_3Cr_2O_8$
J_0	5.551(9)
J_1'	-0.04(1)
J_1''	0.24(1)
J_1'''	0.25(1)
$J_2' - J_3'$	0.751(9)
$J_2'' - J_3''$	-0.543(9)
$J_2''' - J_3'''$	-0.120(9)
J_4'	0.10(2)
J_4''	-0.05(1)
J_4'''	0.04(1)
J' =	$J' = 3.6(1)$
J'/J_0	$J'/J_0 = 0.6455$

Simulation and Data

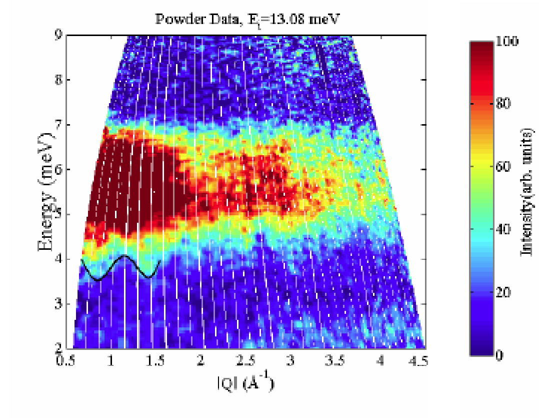
Neutron cross-section

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

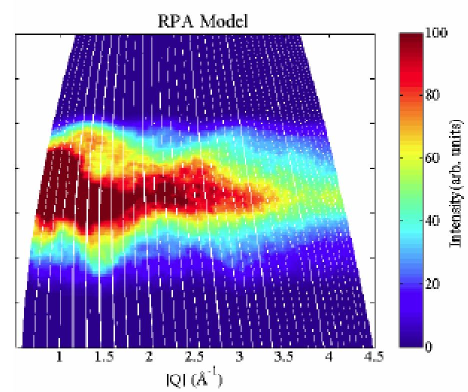
Data – single crystal simulation – single crystal



Data - Powder average:



Simulation - Powder average:

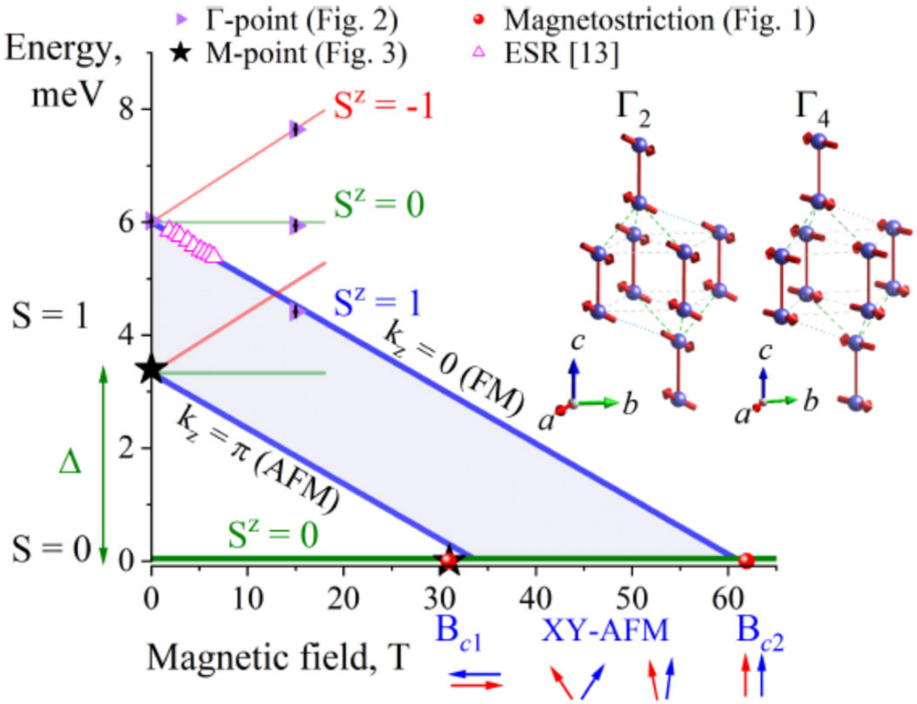


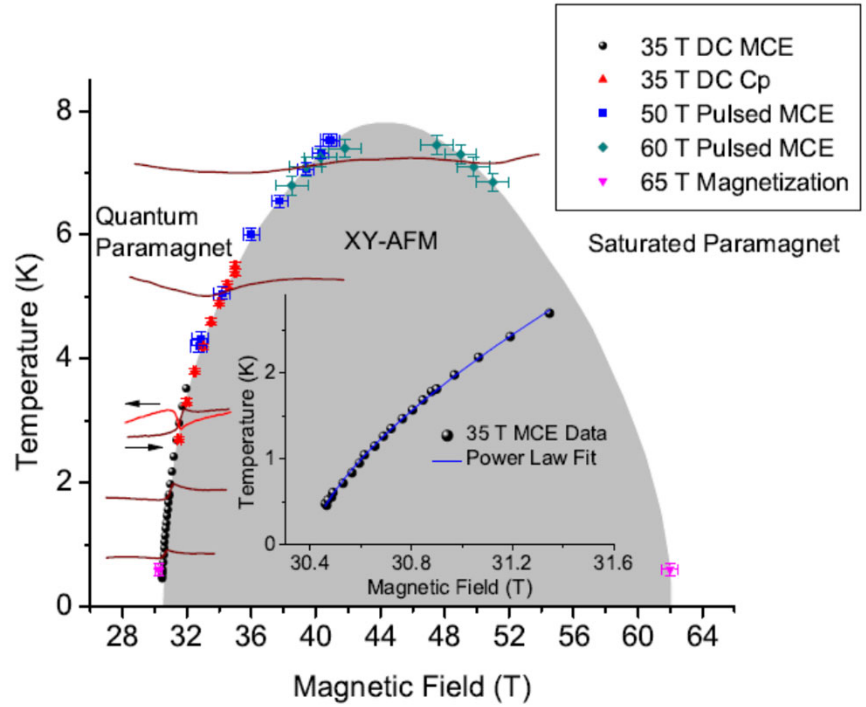
D. L. Quintero-Castro, B. Lake, E.M. Wheeler
Phy. Rev. B. 81, 014415 (2010)

Simulation of the TOF data with the fitted values interactions

Bose-Einstein Condensation in $Sr_3Cr_2O_8$

Zeeman Splitting in Field





A.A. Aczel, Y. Kohama, C. Marcenat, F. Weickert, O.E. Ayala-Valenzuela, M. Jaime, R.D. McDonald, S.D. Selesnic, H.A. Dabkowska, G.M. Luke PRL 103, 207203 (2009)

$$T_c(H) = A(H - H_{c1})^{\upsilon}$$

Critical field $H_{c1} = 30.40$ T

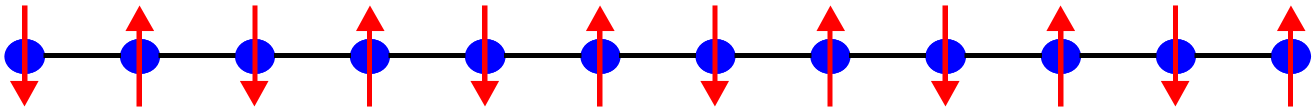
Critical exponent $\upsilon = 0.65(2)$

3D BEC universality class $\upsilon = 2/3$

Bose-Einstein Condensation confirmed

Example 2
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)

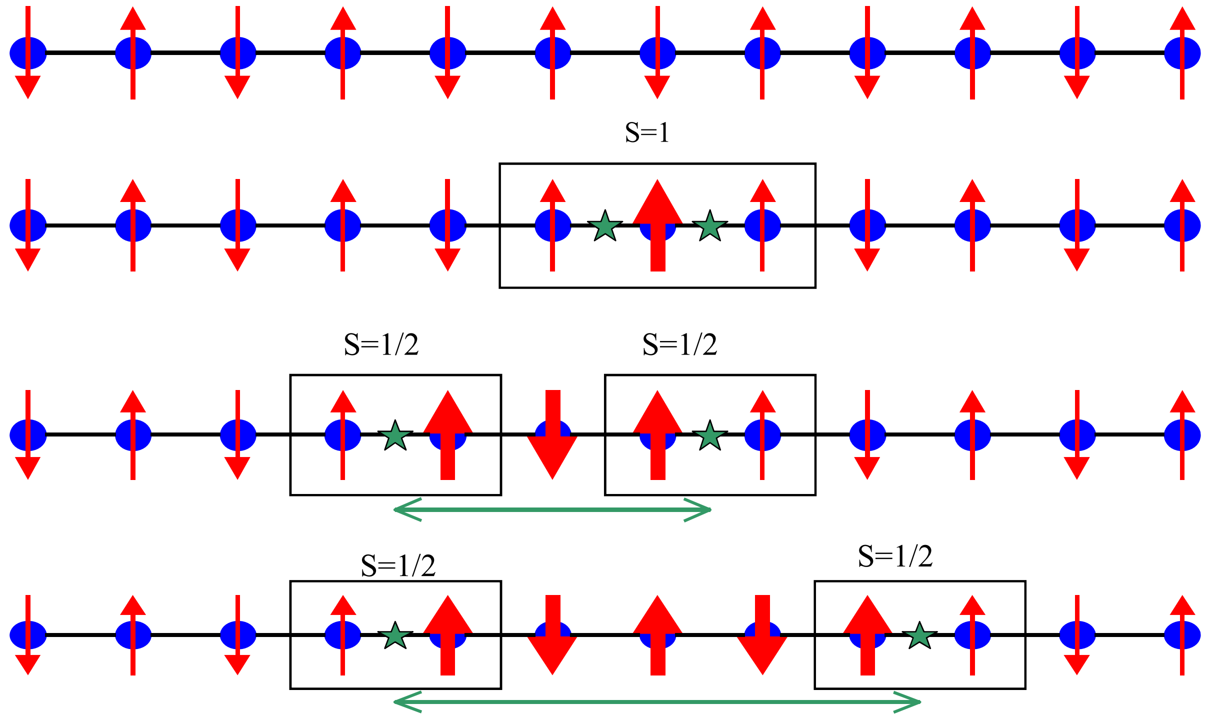
- Ground state has no long-range Néel order.
- Ground state consists of 50% spin-flip states
- All combinations must be considered.
- Little physical insight into the quasi-particles.

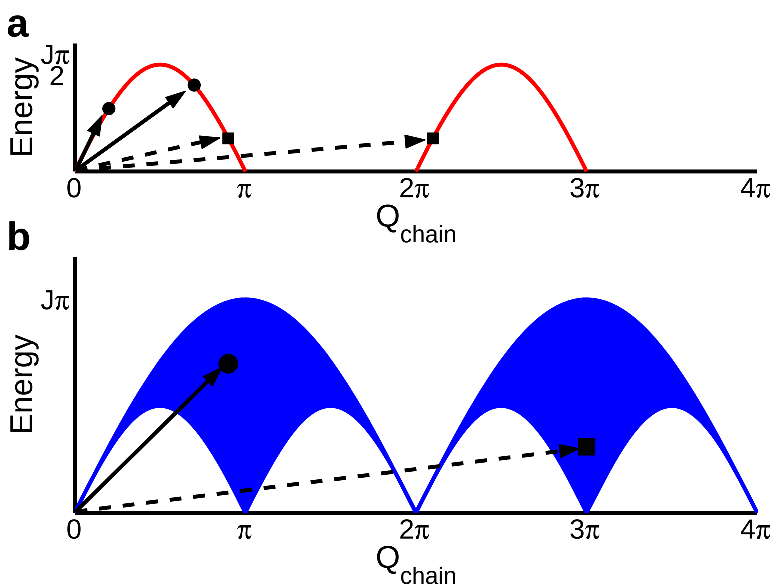
The Bethe Ansatz has been a long standing problem of theoretical condensed matter

Spinons Excitations

Fadeev and Taktajan (1981)

The fundamental excitations are spinons not magnons.





Spinons

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

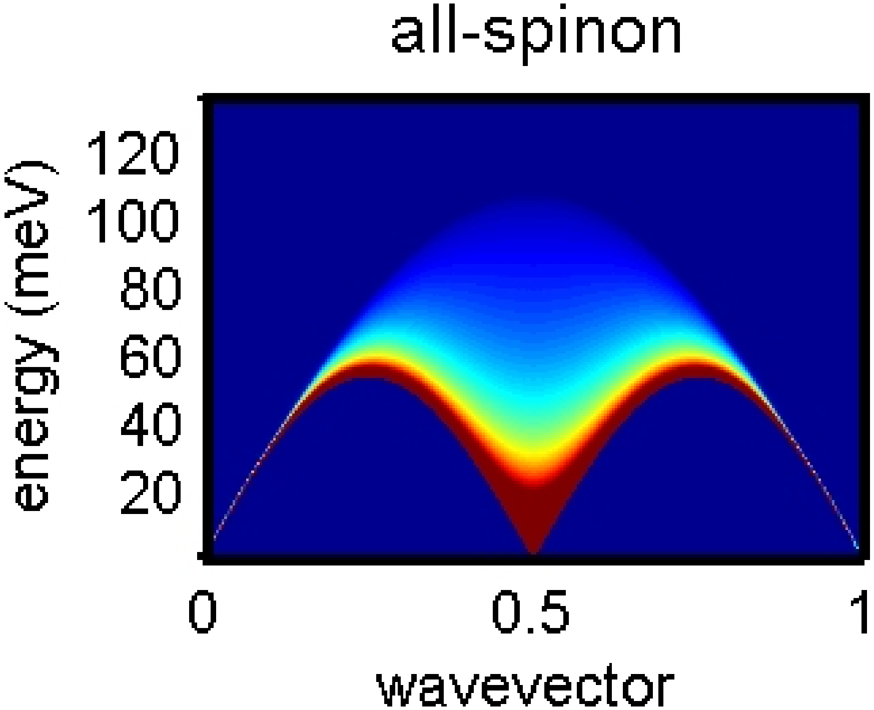
Solution of Bethe Ansatz

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

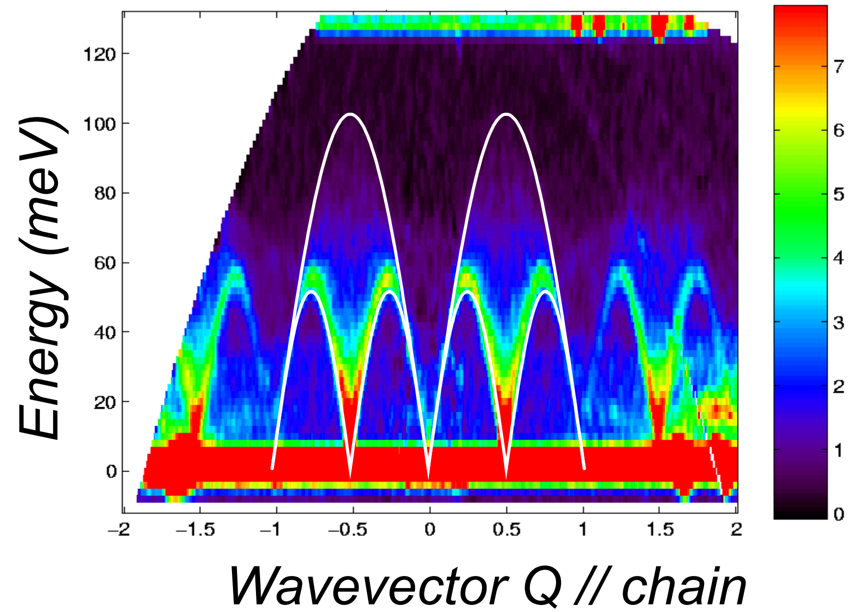
Antiferromagnetic chains, $J_{//}$= -34 meV

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

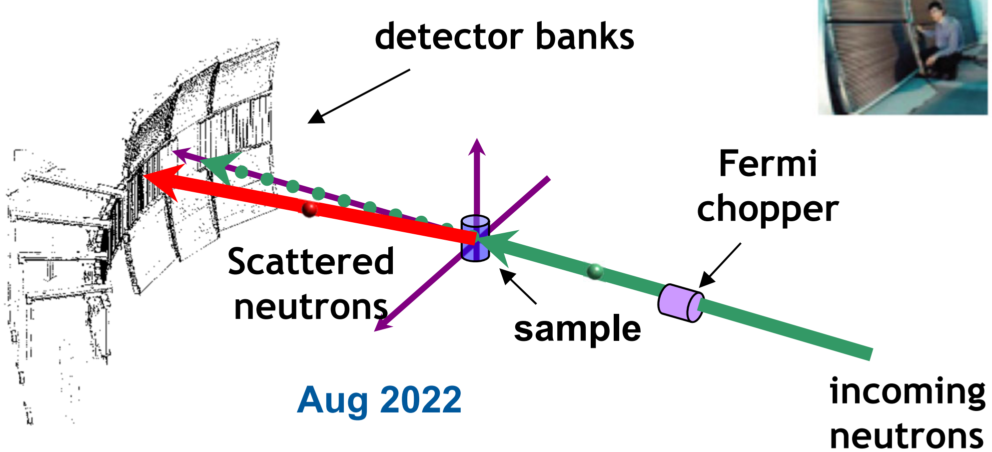
Antiferromagnetic order T_N ~ 39K

Only 50% of each spin is ordered

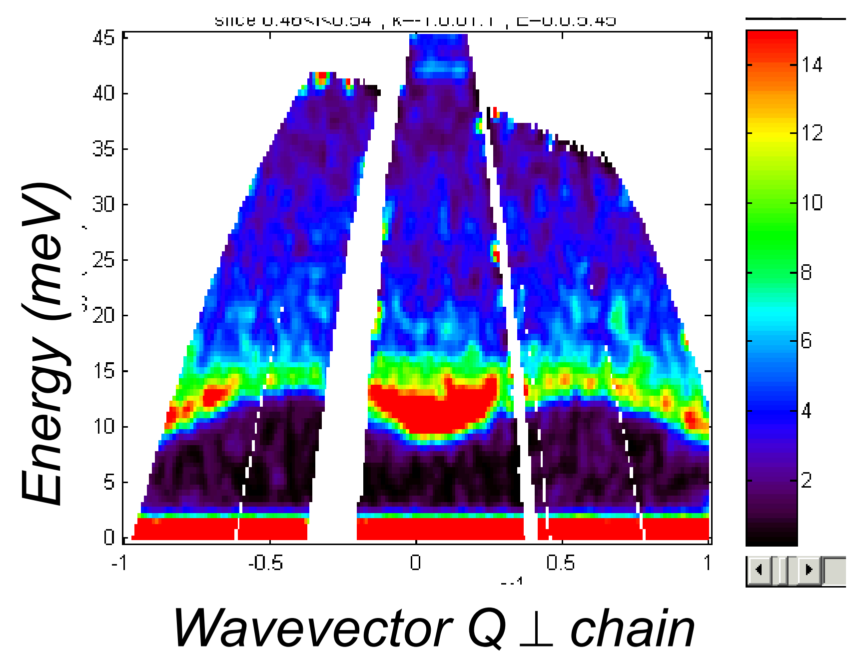
$$\hat{H} = J_{\parallel} \sum_{r} \vec{S}_{r,l} \cdot \vec{S}_{r+1,l} + J_{\perp} \sum_{l,\delta} \vec{S}_{r,l} \cdot \vec{S}_{r,l+\delta}$$



MAPS neutron spectrometer, ISIS



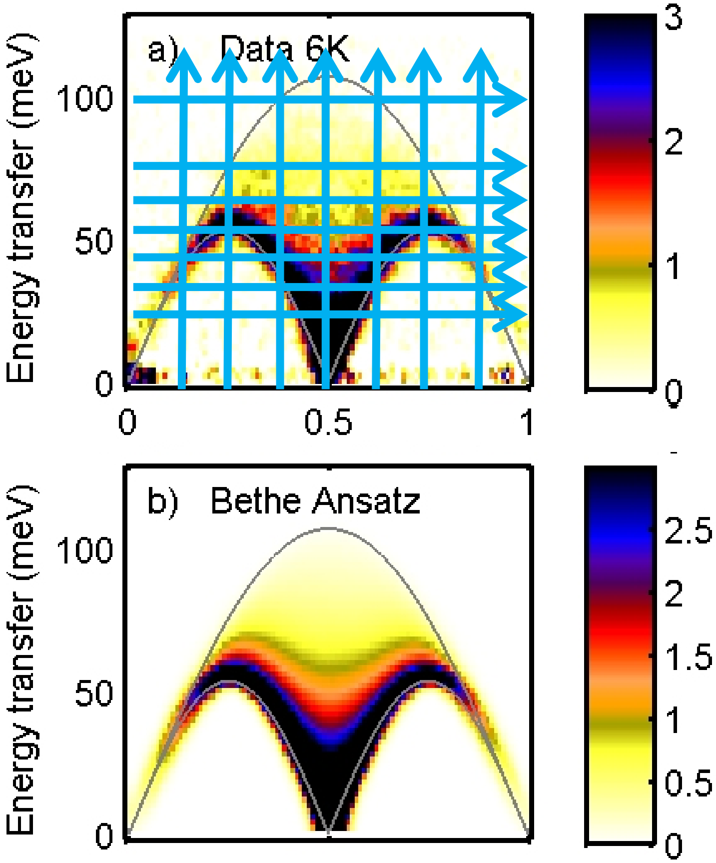
Aug 2022

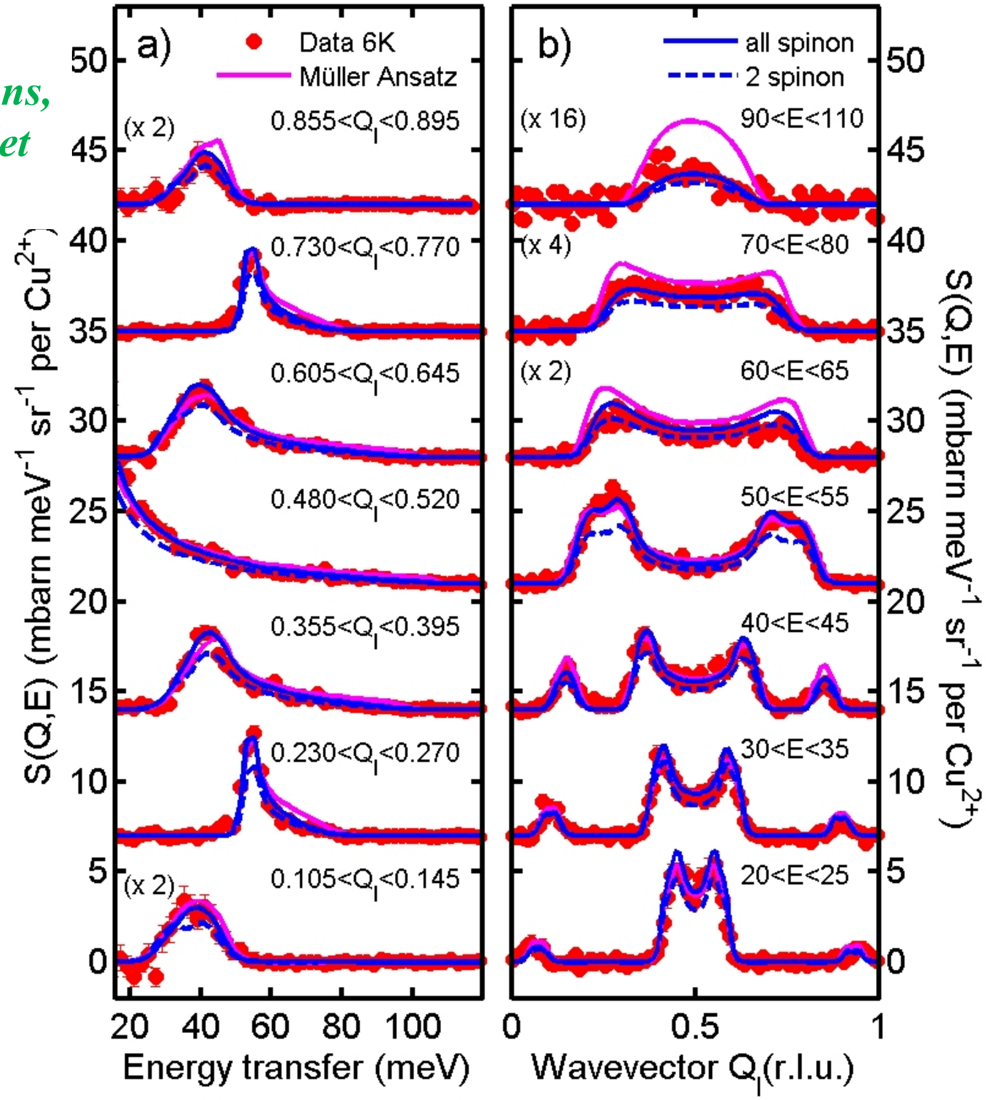


KCuF$_3$ compared to Bethe Ansatz, 2 and 4 spinons

Constant energy and constant-wavevector cuts compared to simulations

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



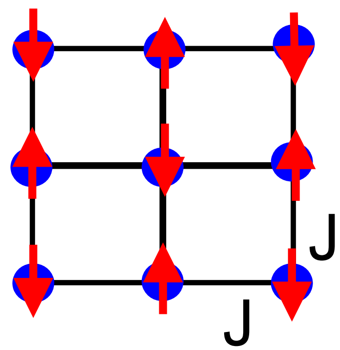


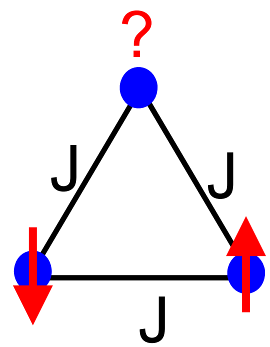
B. Lake et al, Phys. Rev. Lett. (2013)

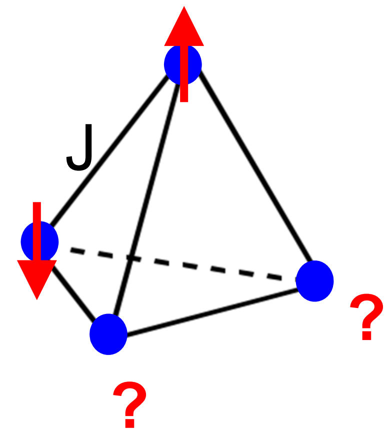
Frustrated magnets

Geometrical Frustration

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

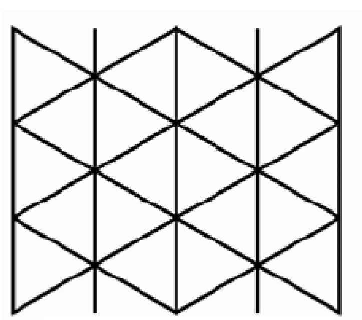




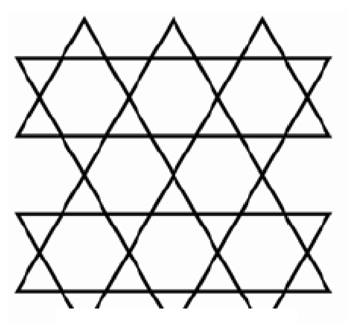


2-dimensional

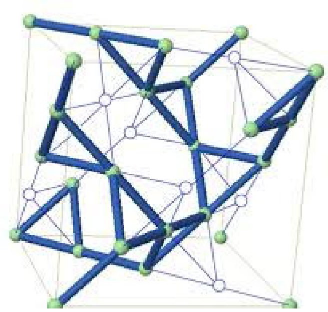
3-dimensional



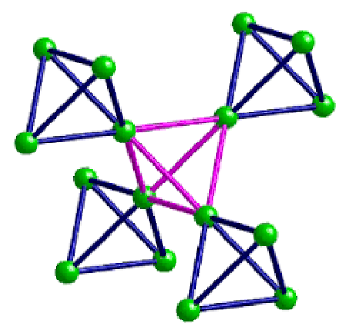
Triangular lattice



Kagome lattice



Hyperkagome lattice

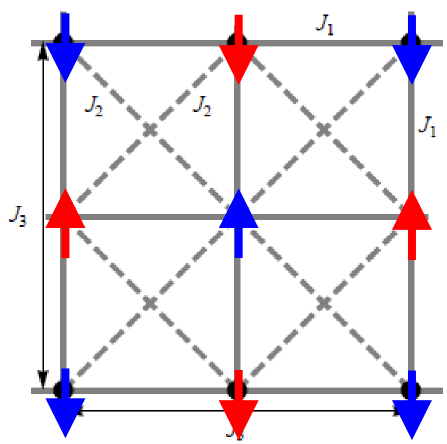


Pyrochlore lattice

Frustration from competing interactions

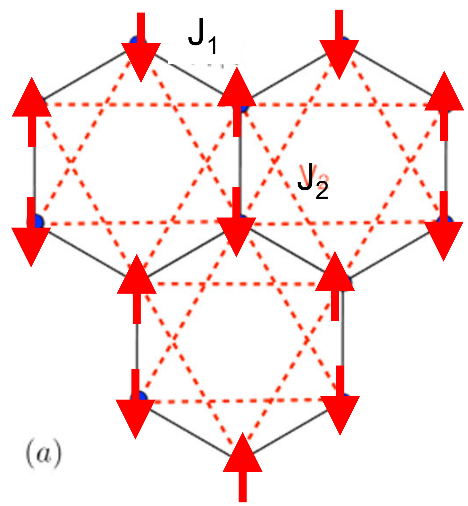
- Second neighbour or further neighbour interactions compete with first neighbour interactions.
- The second neighbour interactions must be AFM

square



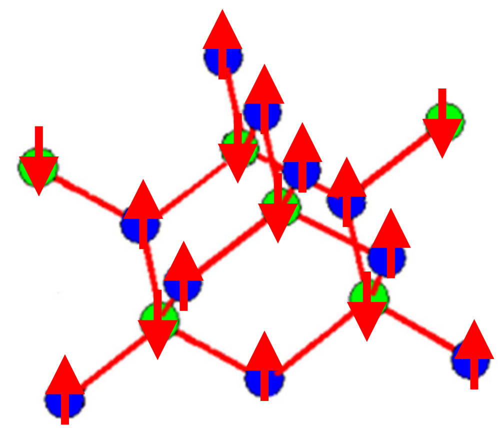
J_1=0
J_2=AFM

honeycomb



J_1=AFM
J_2=0

diamond

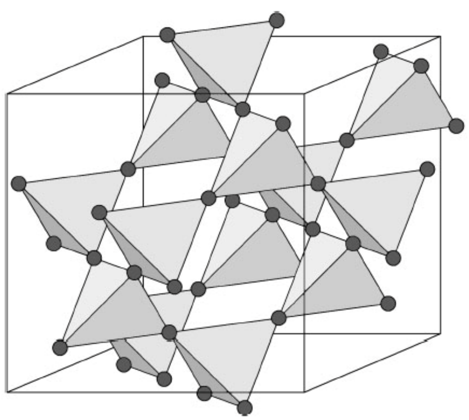


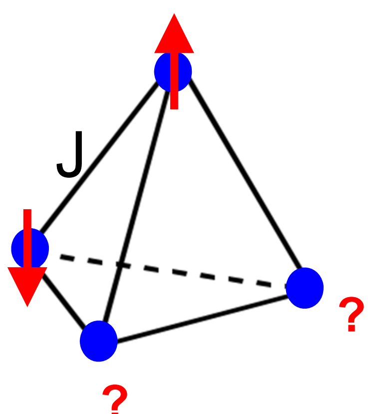
J_1=AFM
J_2=0

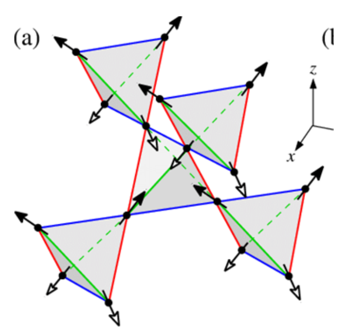
Frustration arising from anisotropy

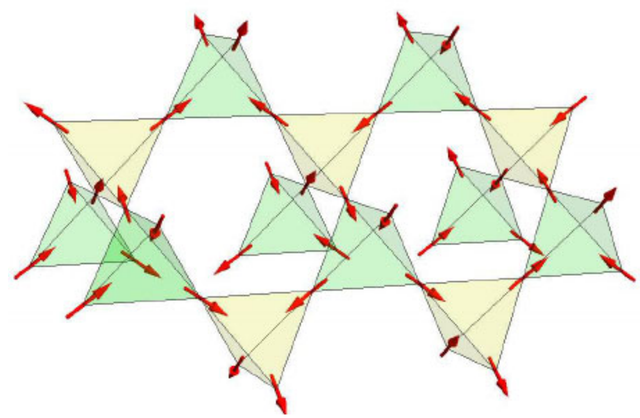
The pyrochlore lattice – corner-sharing tetrahedra

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









Spin liquid

Very strong frustration can induce the spin liquid ground state

No long-range magnetic order or static magnetism even at T=0K

The excitations are not magnons or spin-waves (S=1), but spinons which fractional quantum numbers (S=1/2)

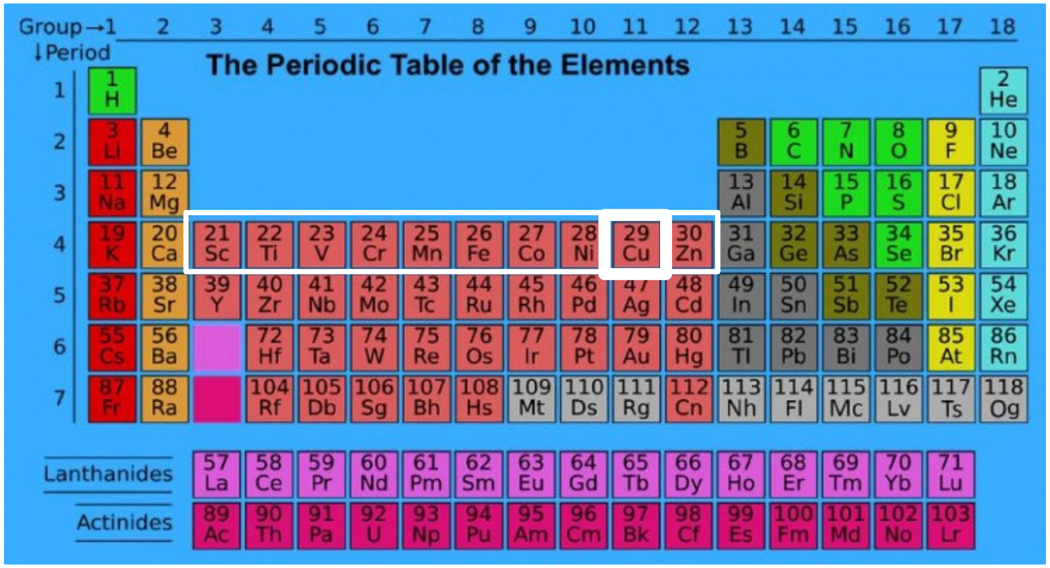
Spin liquids,

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

Physical realisations of frustrated magnets

Physical realisations of frustrated magnets

Light transition metal ions (3d-shell) have quenched orbitals due to strong crystal field. The magnetic moment is due to the spin only and is isotropic, many ions have several valences and the spin depends on the valence.
Quantum spin (S=1/2) is particularly interesting.



<u>Copper</u>
3d-shell
5 orbital x 2 spins = 10 electrons
4s-shell
1 orbital x 2 spins = 2 electrons

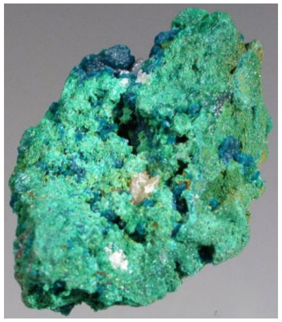
Cu [Ar] ↑↓ ↑↓ ↑↓ ↑↓ ↑↓ ($3d^{10}$) ↑ ($4s^1$)

Cu^+ [Ar] ↑↓ ↑↓ ↑↓ ↑↓ ↑↓ ($3d^{10}$) _ ($4s^0$) **S=0**

Cu^{2+} [Ar] ↑↓ ↑↓ ↑↓ ↑↓ ↑ ($3d^9$) _ ($4s^0$) **S=1/2**

Herbertsmithite - realisation of quantum kagome magnet

$ZnCu_3(OH)_6Cl_2$



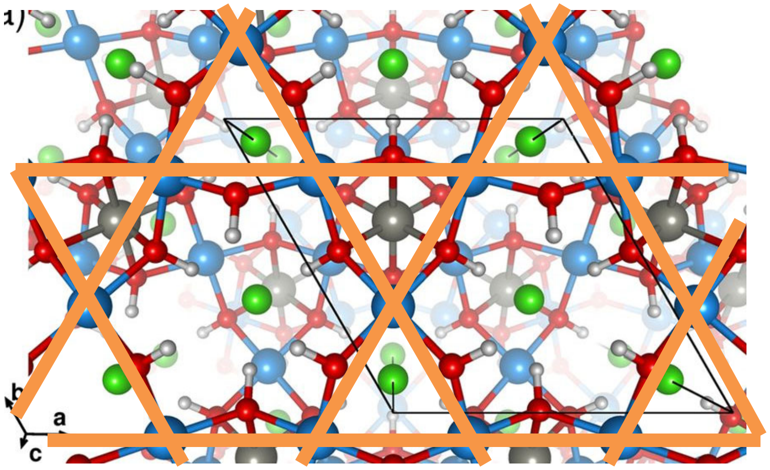
Zn valence is +2, O valence is -2, H valence is -1, Cl valence is +1

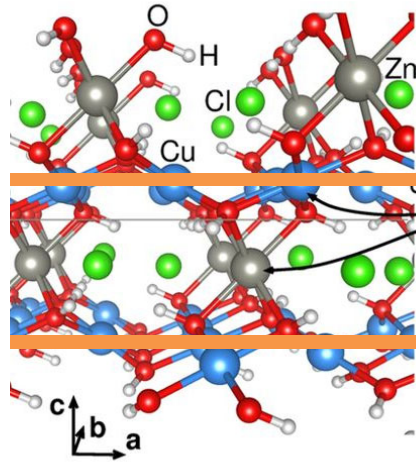
$$1x(+2)[Zn] + 3x(V_{cu})[Cu] + 6x(-2)[O] + 6x(+1)[H] + 2x(-1)[Cl] = 0$$

$$3V_{Cu} – 6 = 0$$

$$V_{Cu} = +2$$

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

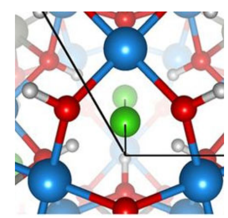




The space group is trigonal $R\bar{3}m$ Cu(blue) O(Red) H(white) Zn(grey), Cl(green)

- 3-fold symmetry gives kagome lattice of Cu^{2+} ions
- Large distance between planes - 2D magnetism

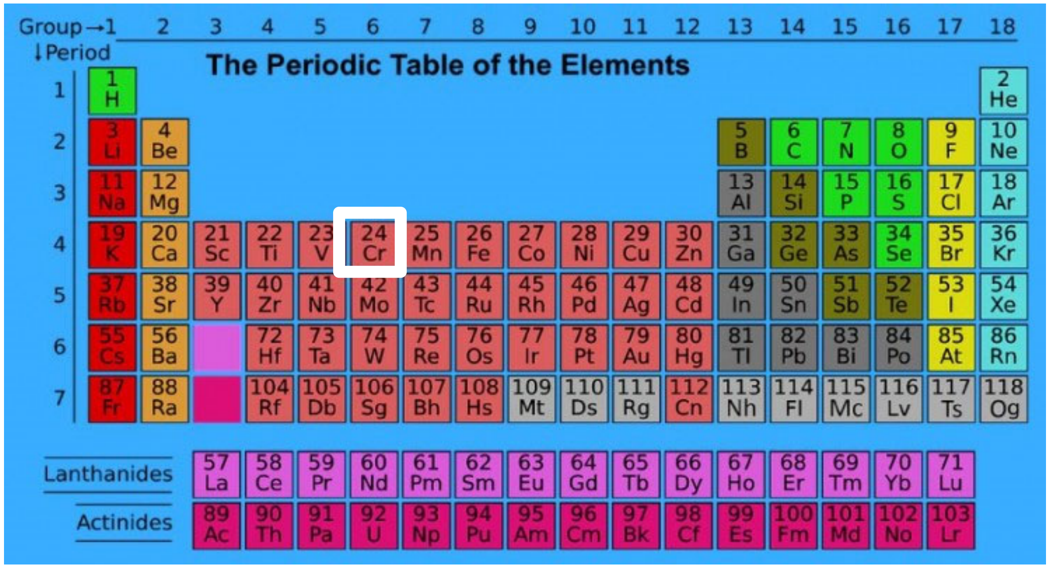
Superexchange via O^{2-} ions Cu^{2+} - O^{2-} - Cu^{2+} bonds ~180°
Goodenough-Kanamori-Anderson rules give AFM interactions



Possible quantum spin liquid but Cu/Zn disorder

$CuCrO_2$ realisation of a triangular lattice

$CuCrO_2$ $\quad 1xV_{Cu}[Cu] + 1xV_{Cr}[Cr] + 2x(-2)[2O] = 0$ $\quad V_{Cu} + V_{Cr} = +4$



Cr [Ar] ↑ ↑ ↑ ↑ ↑ $3d^5$ ↑ $4s^1$

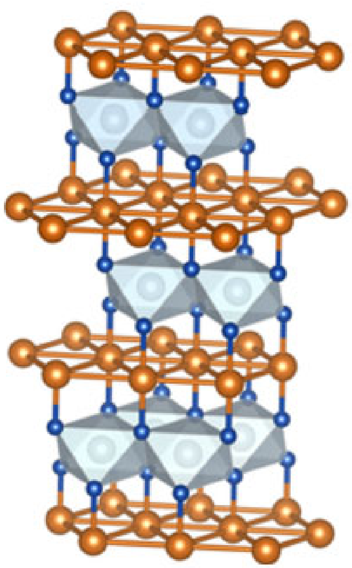
Chromium takes valence 3+

Cr^{3+} [Ar] ↑ ↑ ↑ $3d^3$ — $4s^0$ **S=3/2**

Hund's first rule, maxmize the spin

$V_{Cu} = +4 - (+3) = +1$

Cu must take the valence +1 **S=0**



The space group is trigonal $\bar{3}m$

The Cr^{3+} ions form a triangular lattice well separated by non-magnetic copper ions

They are coupled by direct exchange interactions between half occupied t_{2g} orbitals which are therefore AFM

Good example of triangular lattice AFM, but additional 2nd neighbour coupling.

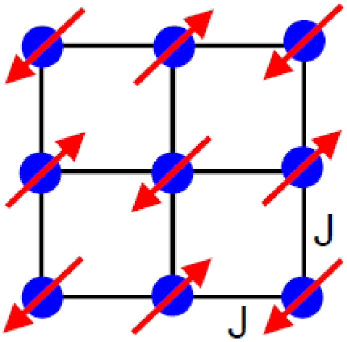
Experimental methods for frustrated magnets

Susceptibility
can show signs of suppressed ordering transition T_N,<<T_{CW} and enhanced correlations above T_N this can also be a sign of low dimensionality which is also interesting

Heat capacity,
suppressed order, magnetic entropy continues to be released above T_N evidence of low lying energy levels

Example 3
Two Dimensional
Quantum Magnets

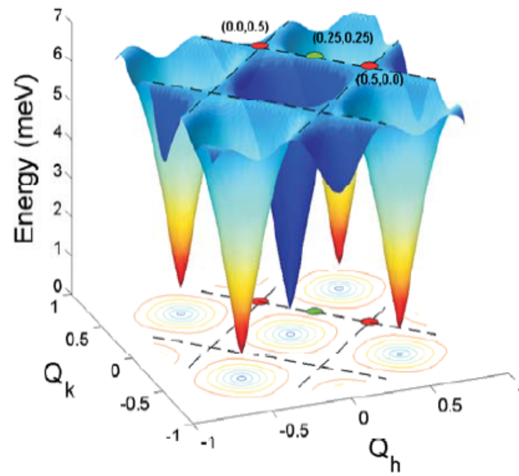
2-Dimensional Antiferromagnet - Square Lattice

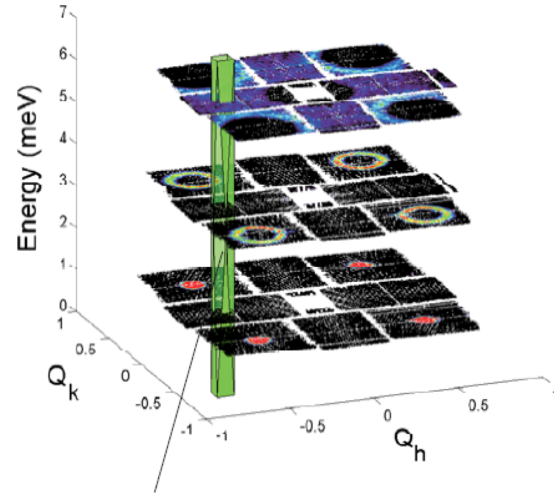


Ground state
long range order

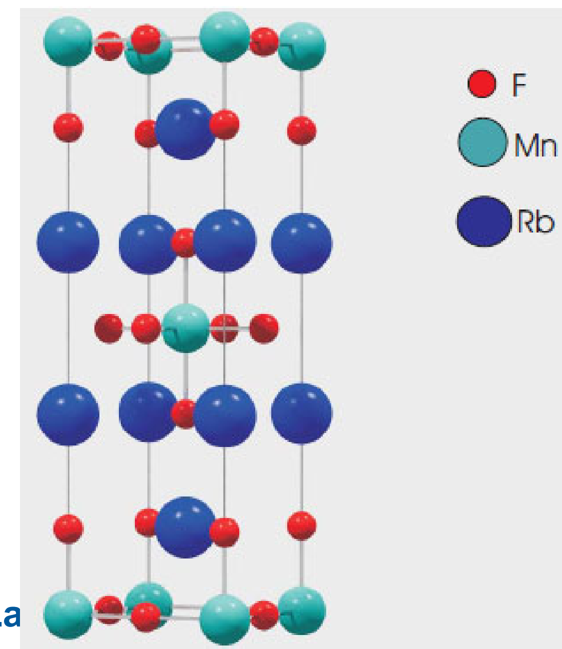
Excitations
Spin-waves

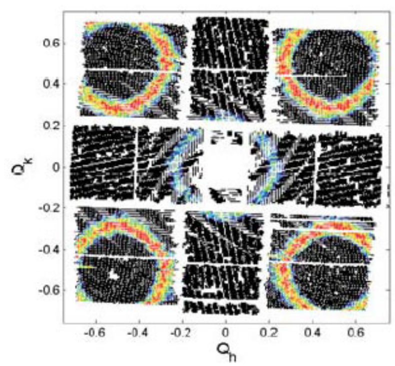
T Huberman et al J. Stat. Mech. (2008) P05017

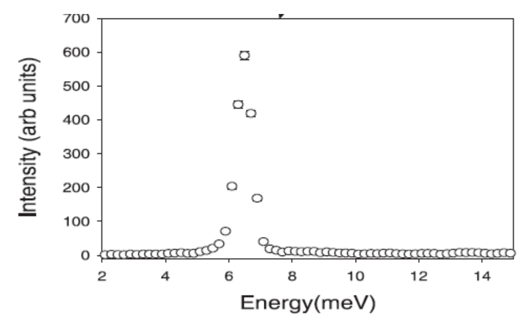




Rb_2MnF_4
2-Dimensional Spin-5/2
Heisenberg Antiferromagnet





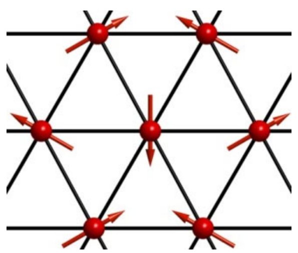


B. La

2-Dimensional Antiferromagnet - Triangular Lattice

Triangular Lattice
Ground state – long range order

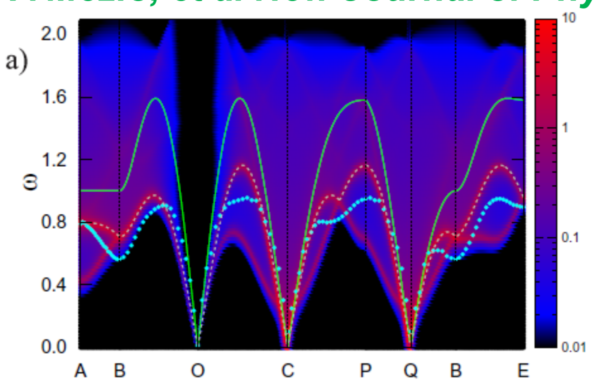
isotropic



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

Excitations

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

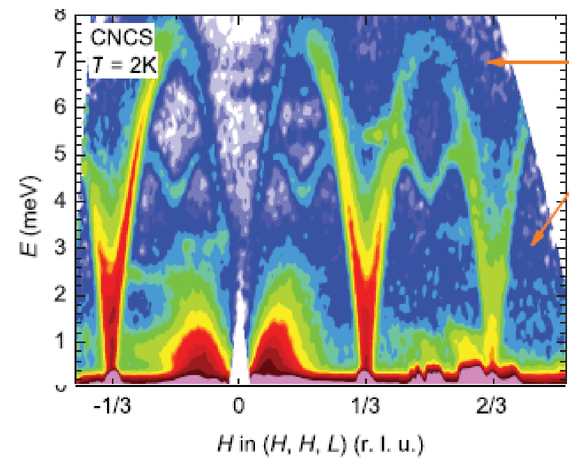


renormalised and broadened compared to spin-wave theory

B. Lake; TMS, Aug 2022

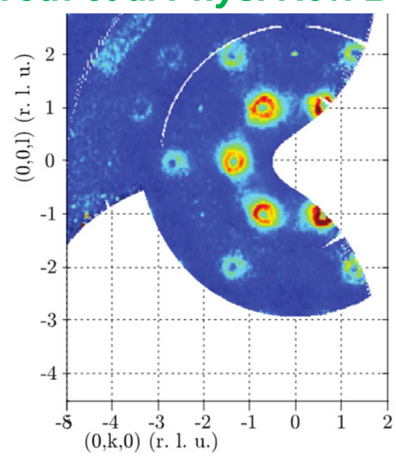
$CuCrO_2$ S-3/2, triangular lattice

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



Alpha-Ca_2CrO_4 S-3/2, triangular lattice

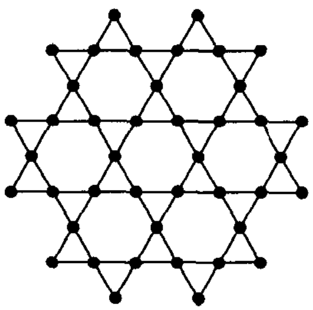
S Toth et al Phys. Rev. B (2011)

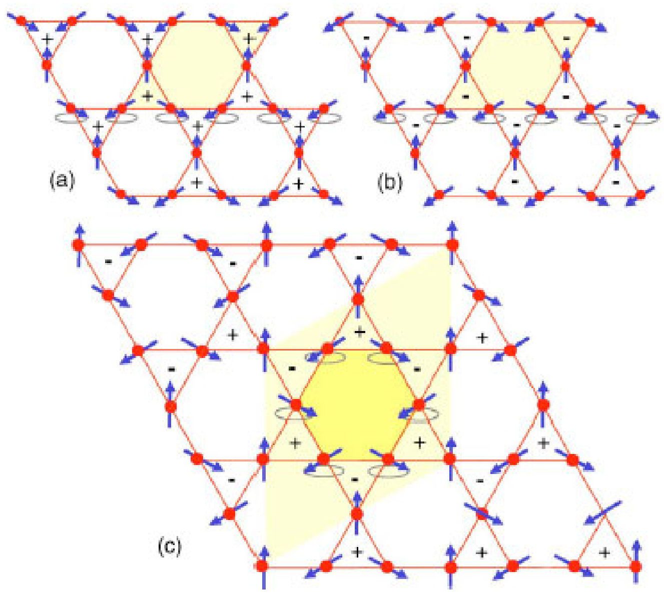


Ideal S-1/2, triangular antiferromagnet, $Ba_3CoSb_2O_9$ H. Tanaka et al

2-Dimensional Antiferromagnet - Kagome Lattice

Kagome Lattice





S-1/2

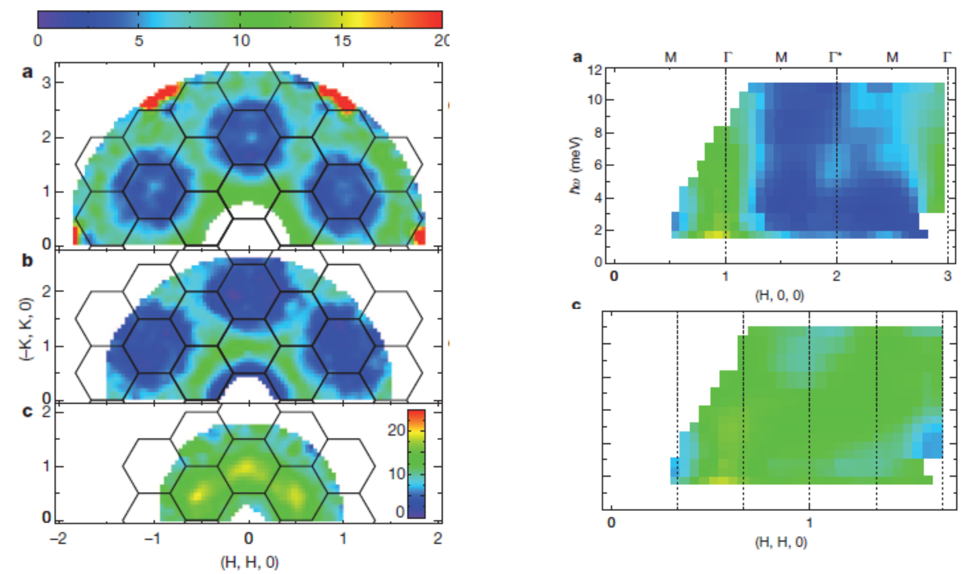
no order

diffuse excitations

e.g. Herbertsmithite

T.-H. Han

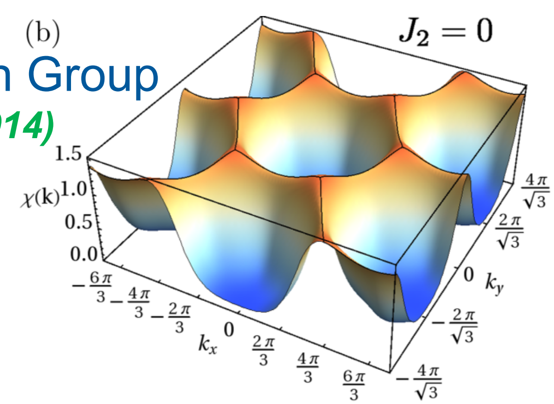
Nature 492, 406 (2012)



Pseudo-Fermion

Functional Renormalisation Group

R. Suttner, et al Phys. Rev. B (2014)



S-5/2

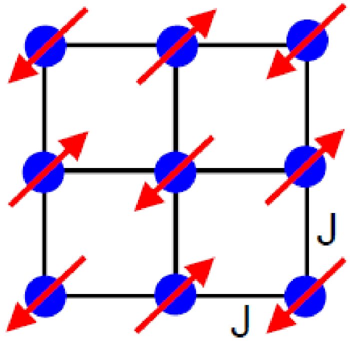
Long-range order

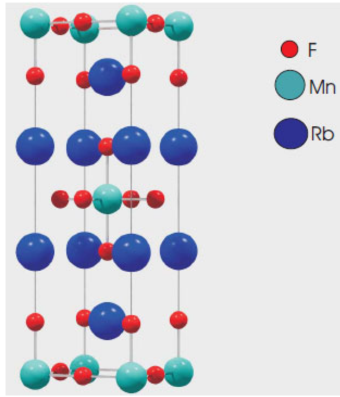
Spin-wave excitation

B. L

2-Dimensional Antiferromagnets

Square Lattice

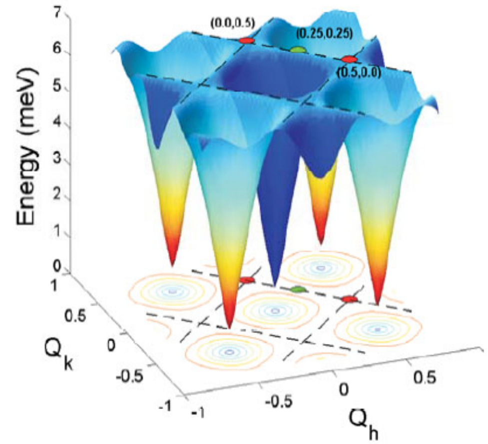




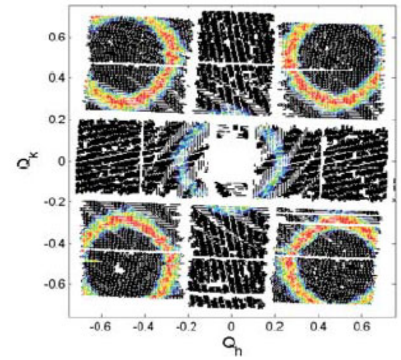
e.g. Rb_2MnF_4

S-5/2 Heisenberg Antiferromagnet

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



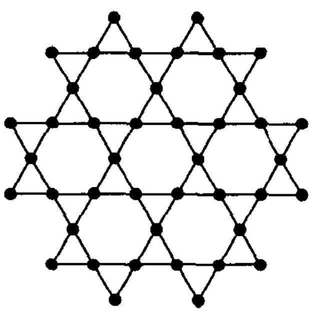
Long-range magnetic order



Spin-waves

Kagome Lattice

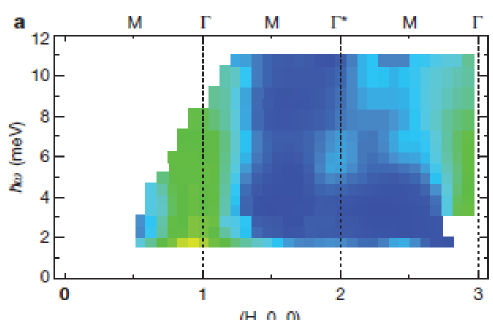
corner-sharing triangles



e.g. Herbertsmithite

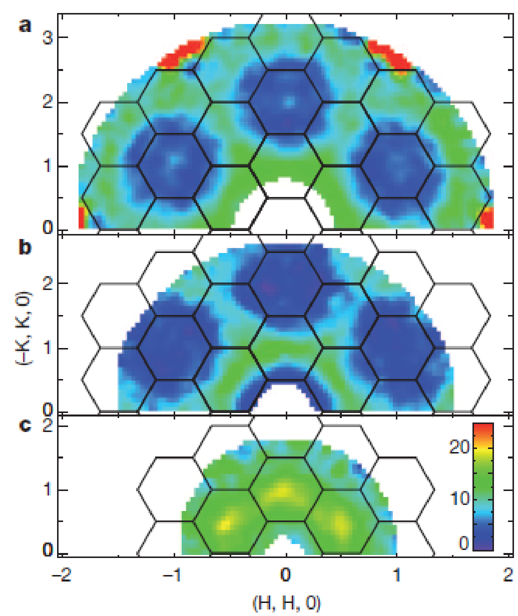
S-1/2 Heisenberg AFM

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



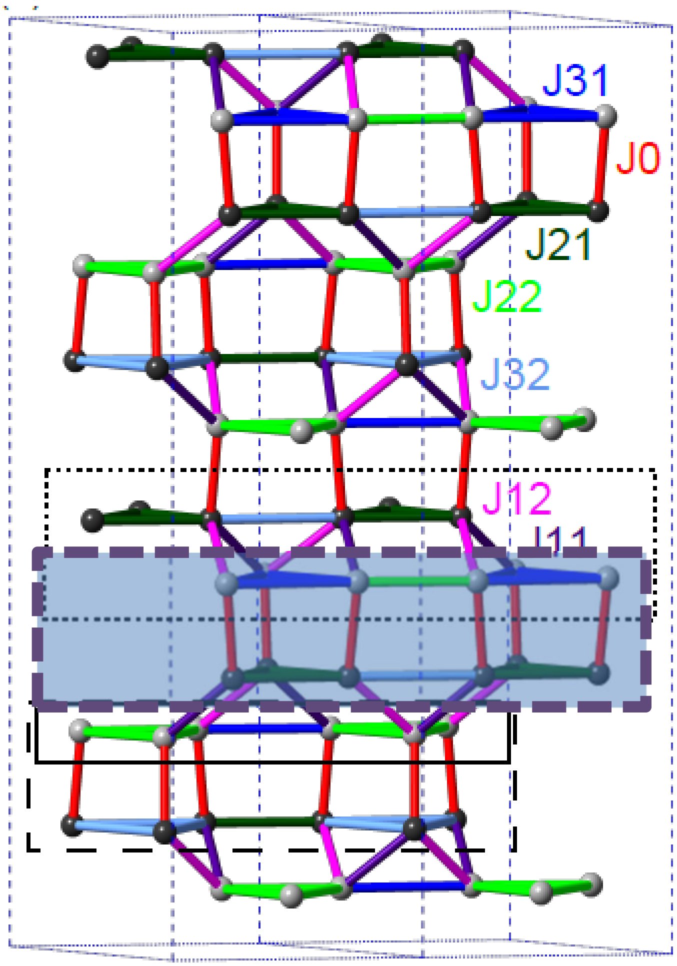
No long-range order

spinon excitations



B. Lake; TMS, Aug 2022

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



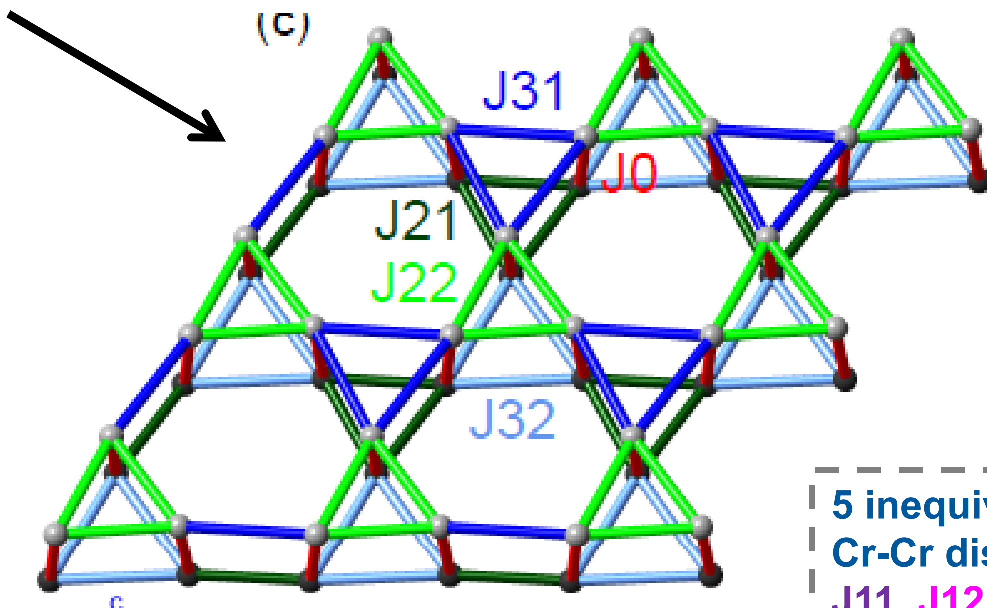
space group *R3c*

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

- Cr^{5+} spin = ½ ions
- 7 different exchange path in structure

Kagome bilayer model

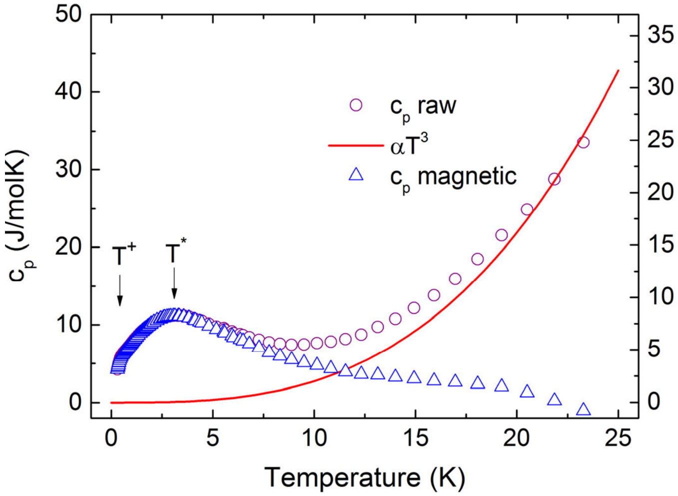
- *a-b* plane shows distorted kagome bilayers
- large blue and small green triangles alternate within and between layers

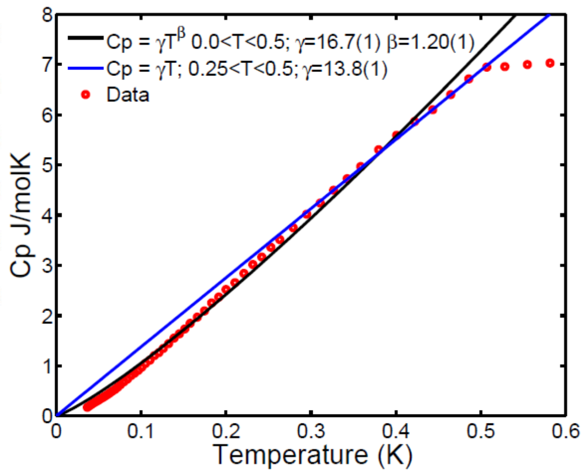


5 inequivalent Cr-Cr distances
J11, J12 = 0

$Ca_{10}Cr_7O_{28}$ - Bulk properties

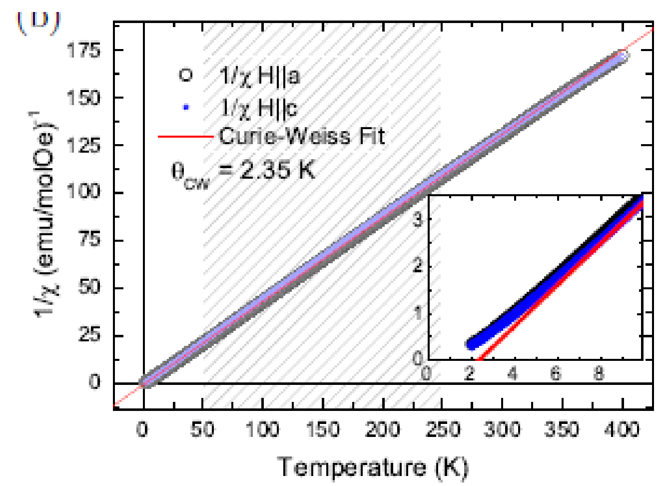
Specific heat





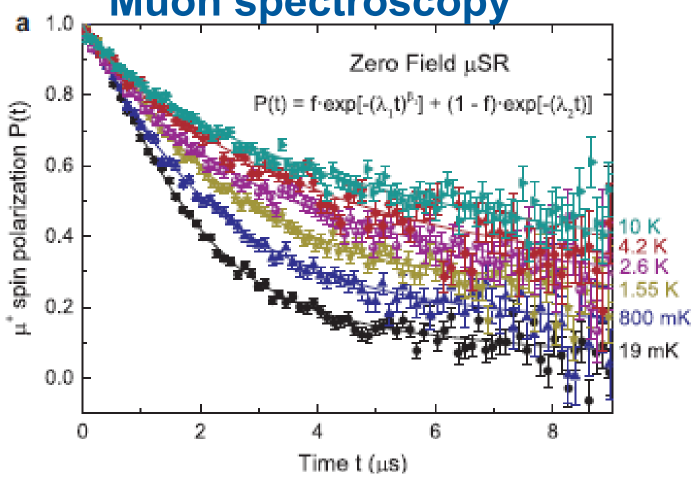
• No order down to 50 mK

DC Susceptibility

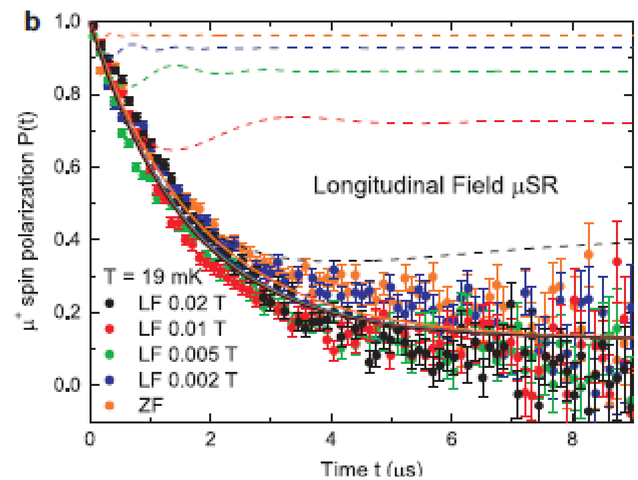


• Isotropic interactions
• FM & AFM interactions

Muon spectroscopy



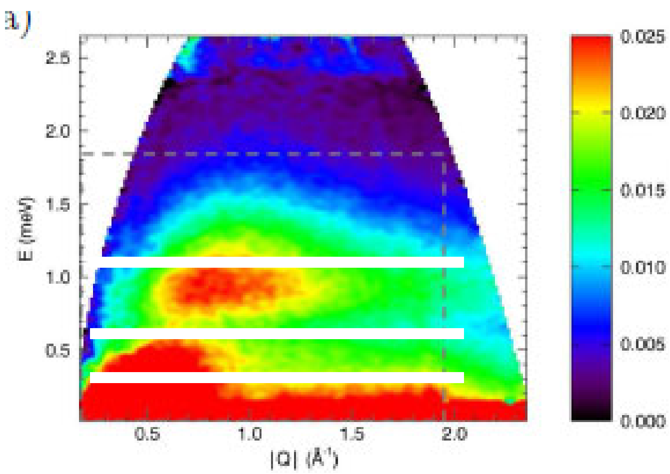
•No oscillations down to 19mK
•No residual polarization at long times
⇒ No static magnetism



•Relaxation persists in longitudinal field
•1T required to overcome relaxation
⇒ dynamical ground state

Inelastic Neutron Scattering – Zero Field

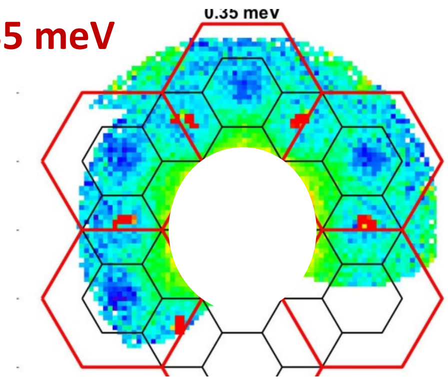
TOFTOF Powder; T=0.43K



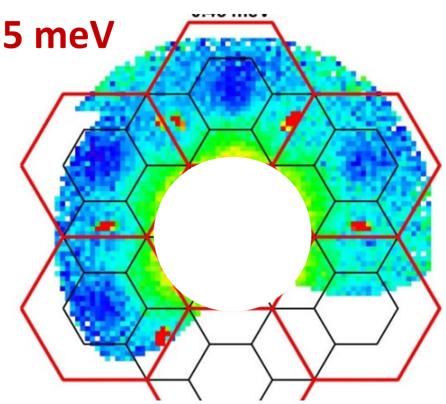
- Excitations to 1.6meV
- Two Bands of excitations
- Gap smaller than 0.1meV

MACS Single Crystals

E = 0.35 meV



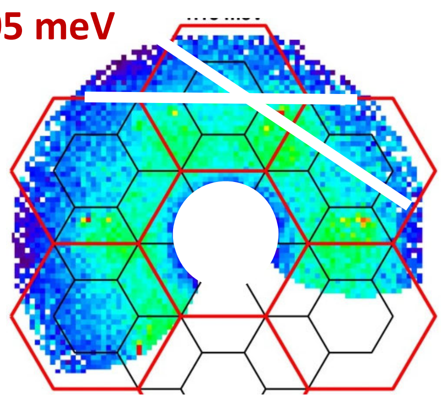
E = 0.55 meV

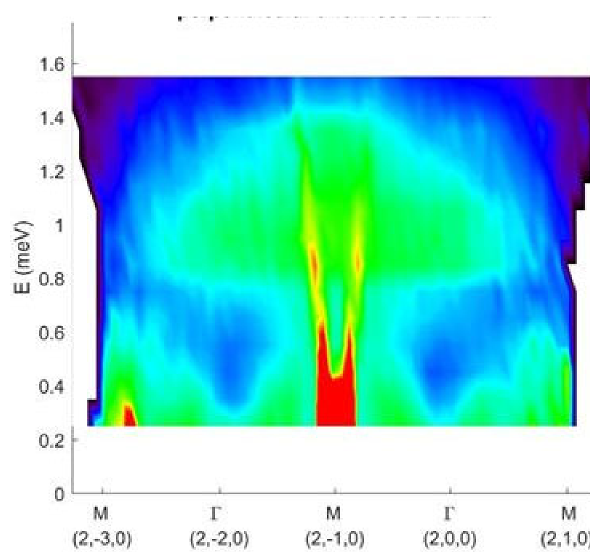


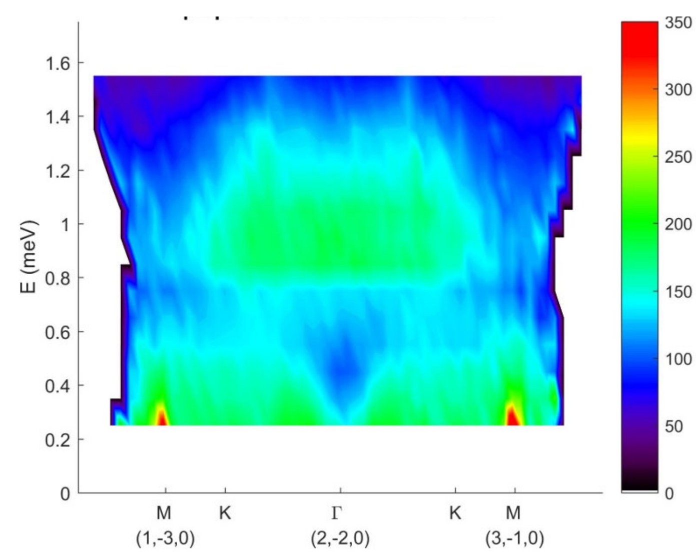
Broad diffuse scattering

No spin-waves?

E = 1.05 meV



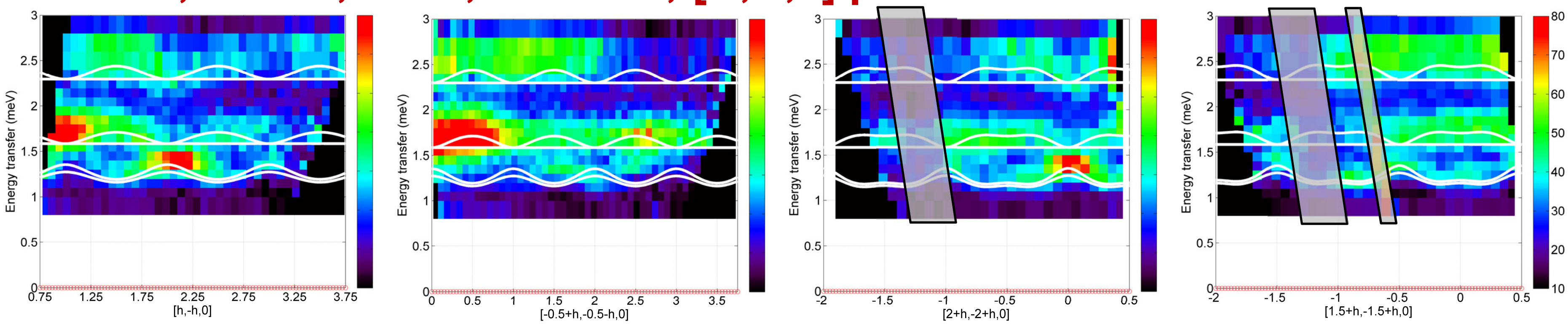




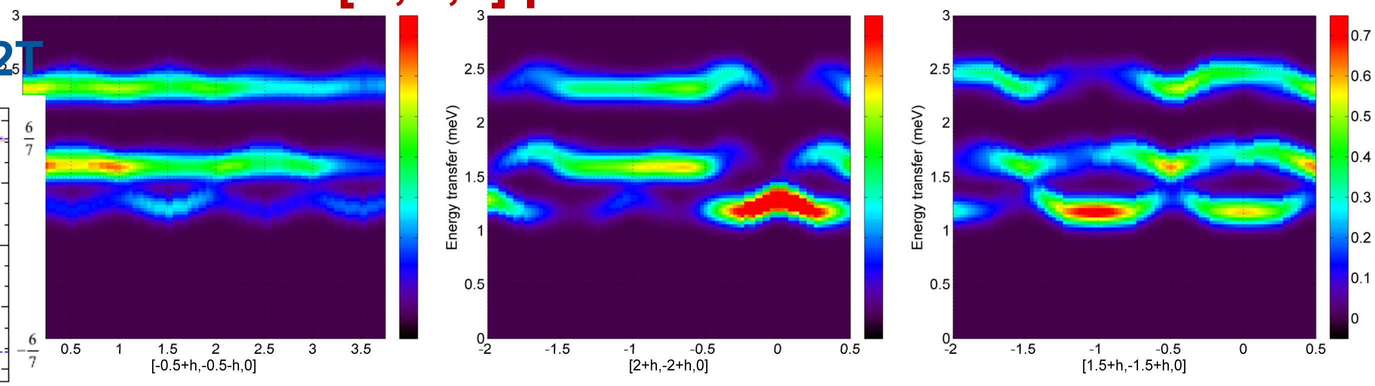
B. Lake; TMS, Aug 2022

Inelastic Neutron Scattering – High Field

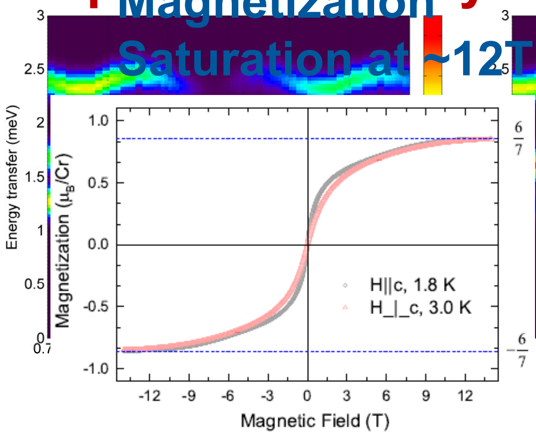
H=11T; MACS, NIST; T=0.09K; [H,K,0] plane

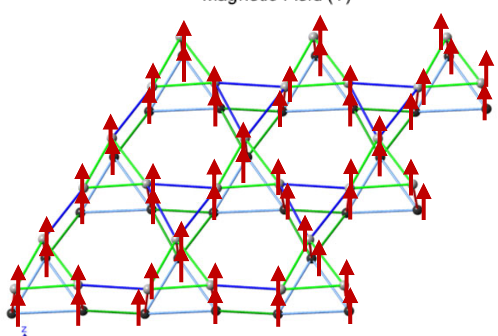


Spin wave theory calculations [H,K,0] plane



Magnetization Saturation at ~12T





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

Hamiltonian of $Ca_{10}Cr_7O_{28}$

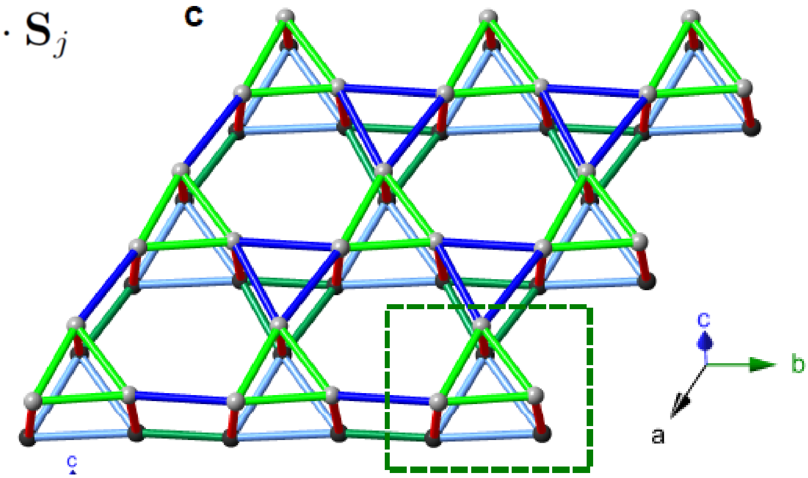
$$\mathcal{H} = \sum_{i,j} J_{i,j}\, \mathbf{S}_i \cdot \mathbf{S}_j$$

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)

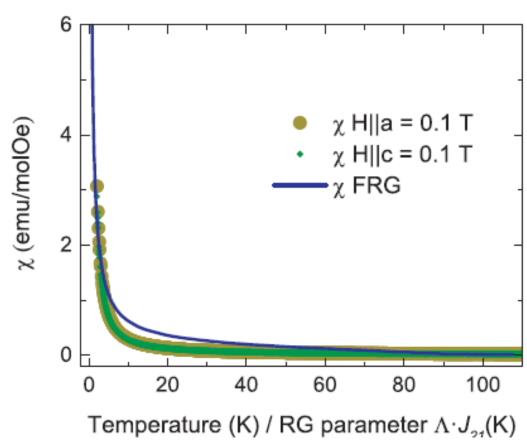


Breathing Kagome Bilayers

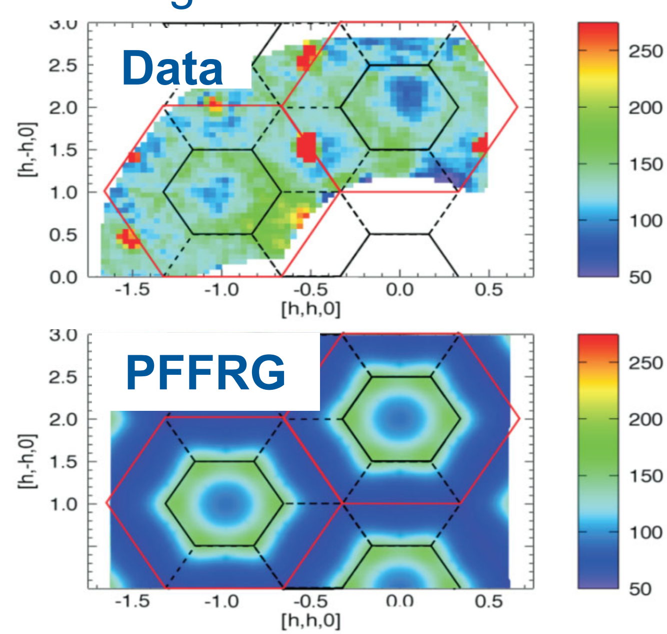
Pseudo-Fermion functional renormalisation group

Using the Hamiltonian extracted from INS

DC Susceptibility



Magnetic structure factor



Theory confirms - No long-range magnetic order, diffuse scattering

Why is $Ca_{10}Cr_7O_{28}$ frustrated?

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

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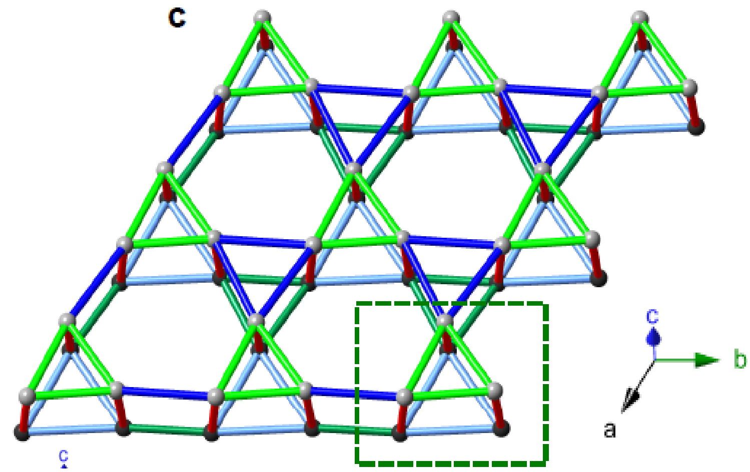
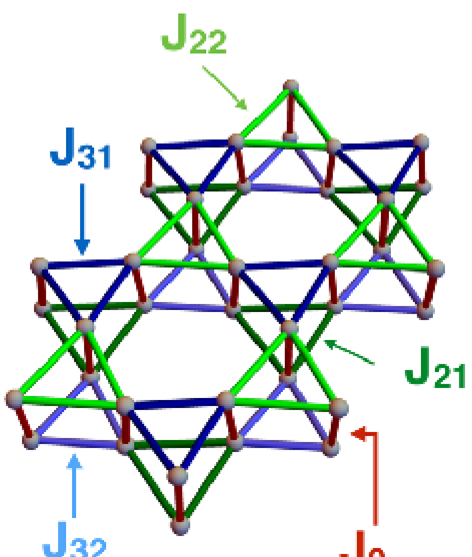
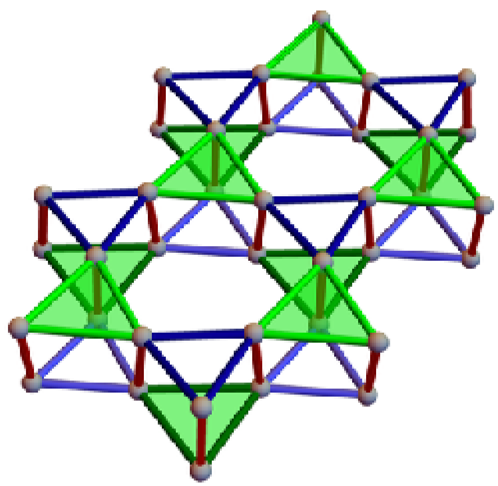
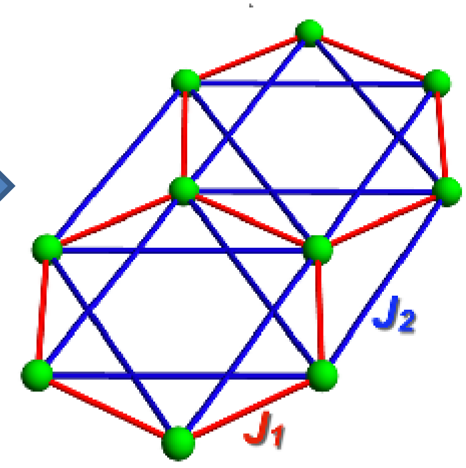




Strong FM interactions on alternating triangles



Effective S=3/2 honeycomb
FM J1=J0=-0.08
AFM J2=0.1



Outline

Quantum magnets

Neutron scattering study of

Example 1 Zero-dimensional quantum magnet

Example 2 One-dimensional quantum magnet

Frustrated magnets

Neutron scattering study of

Example 3 Two-dimensional quantum magnet