Measuring Spin-Waves

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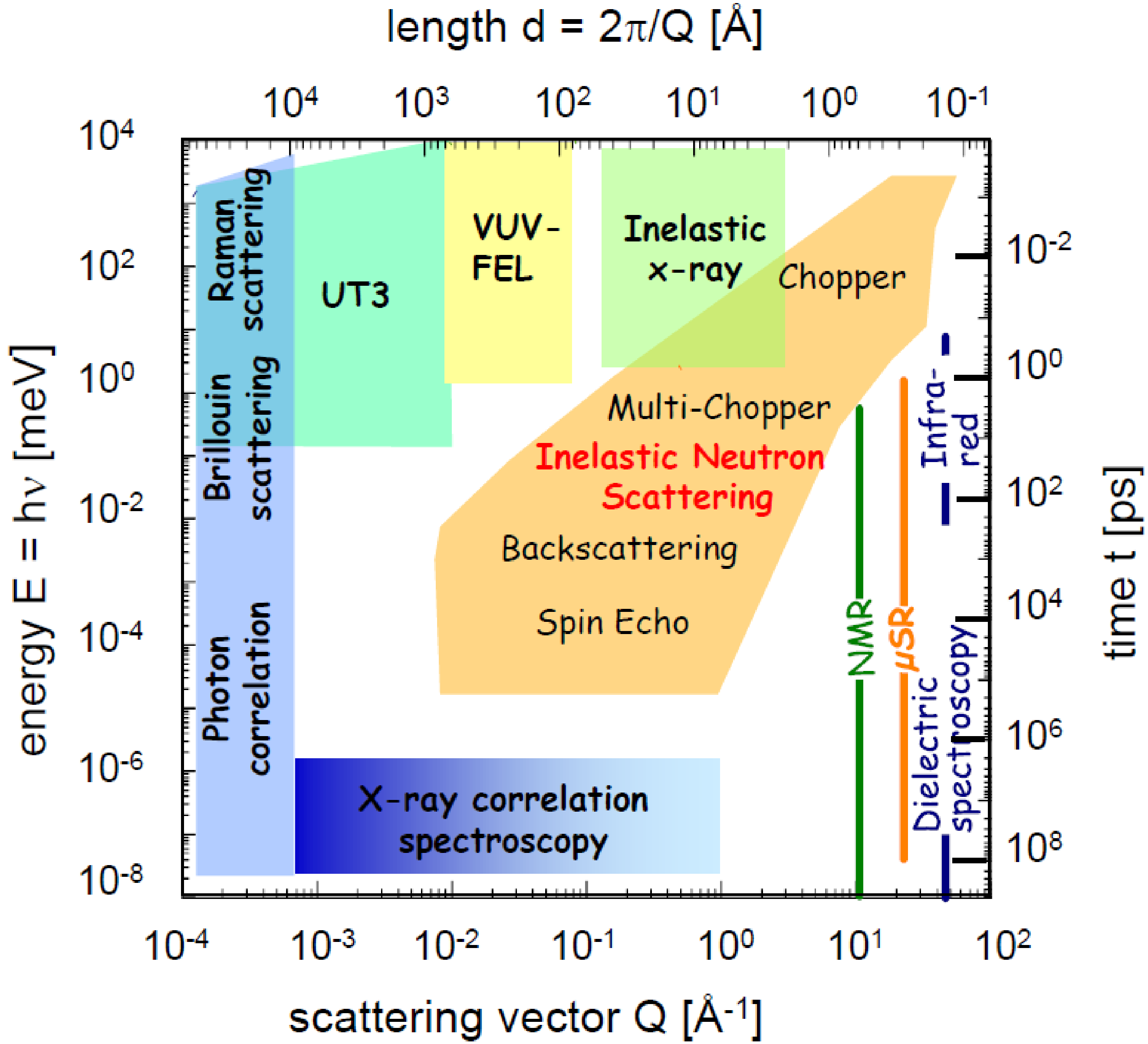
Outline

- Conventional Magnets
  - long-range magnetic order, spin-wave excitations
- Inelastic Magnetic Neutron Scattering Cross-Section
- Measuring spin-waves
  - Triple-axis and time-of-flight spectrometers
- Example

Techniques for measuring excitations



Conventional Magnets

Conventional Magnetism - Exchange Interactions

Heisenberg interactions

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

3D magnet

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

e.g. $RbMnF_3$

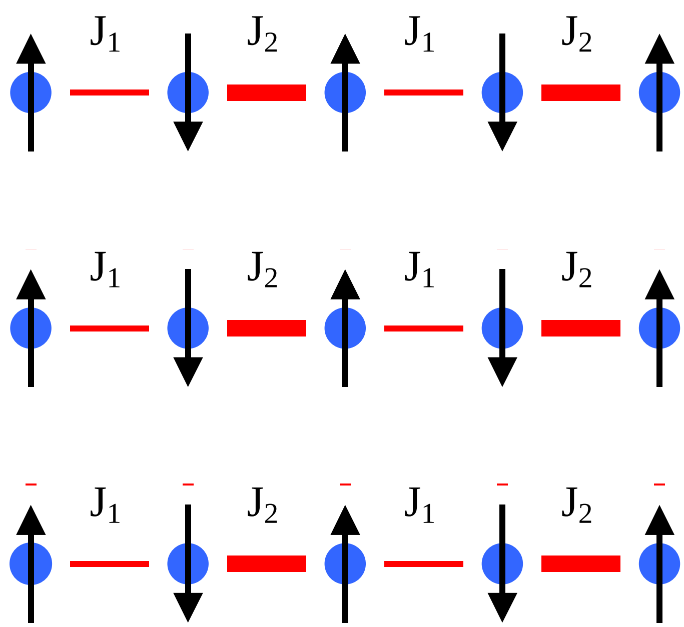
$J < 0$ ferromagnetic

$J > 0$ antiferromagnetic

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

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

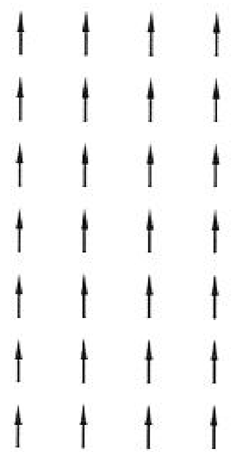
e.g. $CuGeO_3$ and $CuWO_4$

Anisotropic interactions

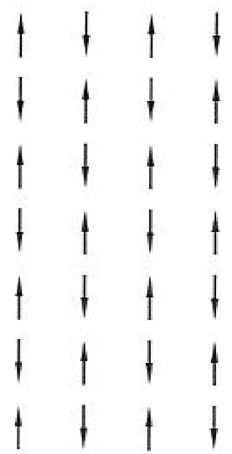
Conventional Magnetism - Ordered Ground State

Exchange interactions between magnetic ions often lead to long-range order in the ground state.

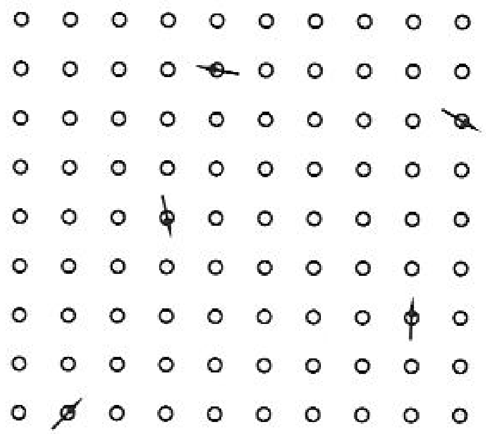
ferromagnet



antiferromagnet

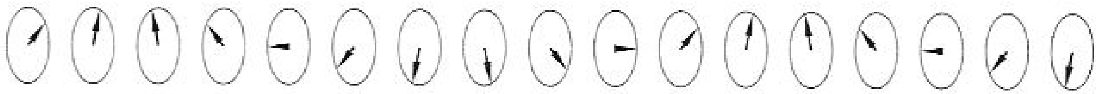


spin glass



spiral magnet

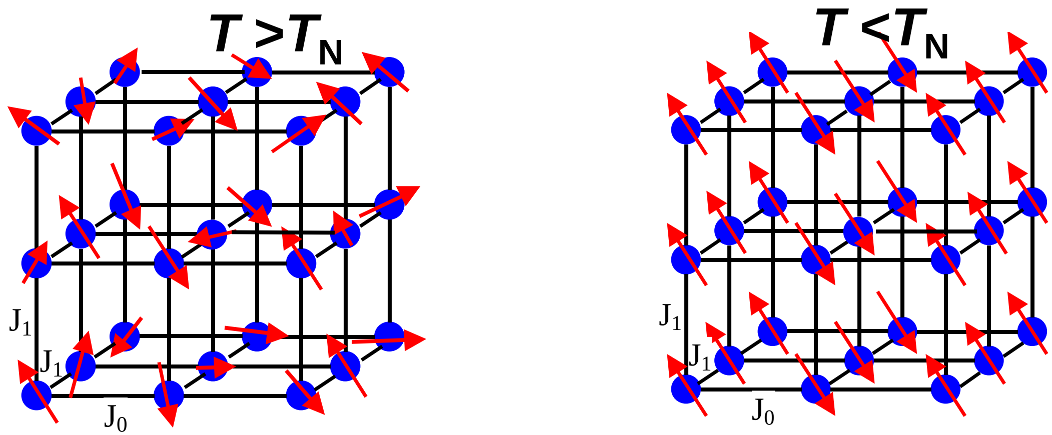
helical magnet



Conventional Magnet - Long-range magnetic order

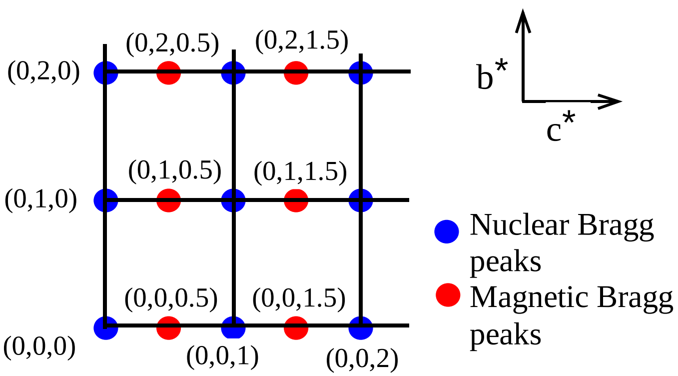
Real Space

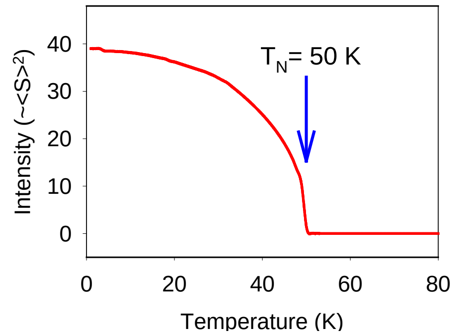
- Long-range magnetic order on cooling as thermal fluctuations weaken



Reciprocal Space

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



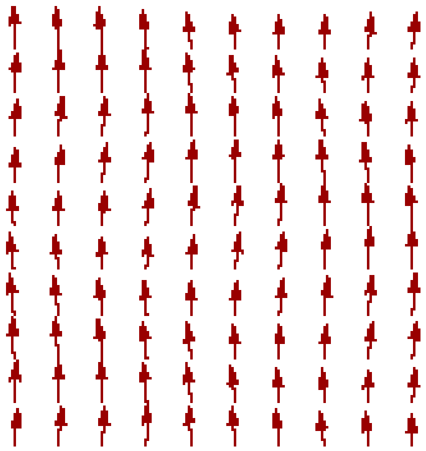


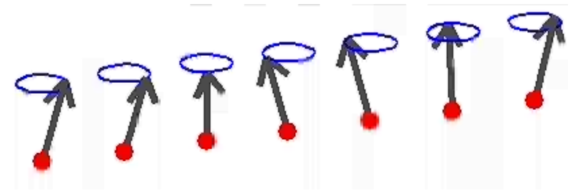
B. Lake; Oxford, Sept 2019

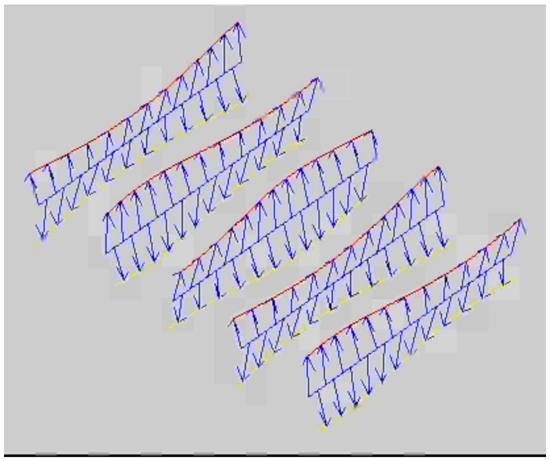
Magnetic Excitations – Spin-Waves

Real Space

Collective motion of spins about an ordered ground state

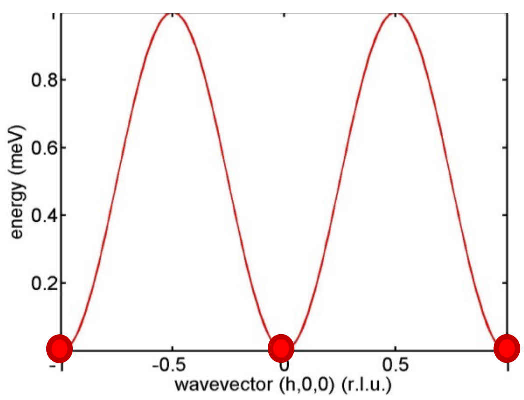


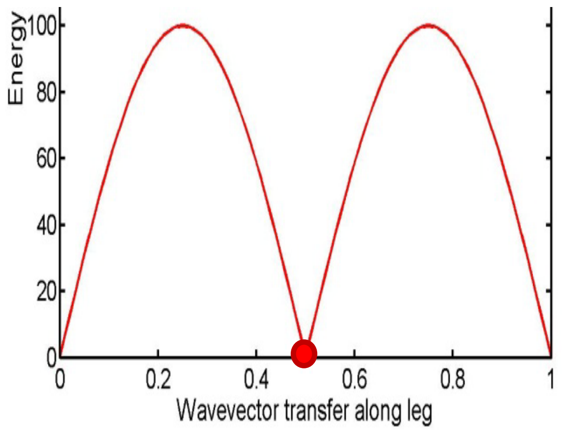




Reciprocal Space

Well-defined dispersion in energy and wavevector





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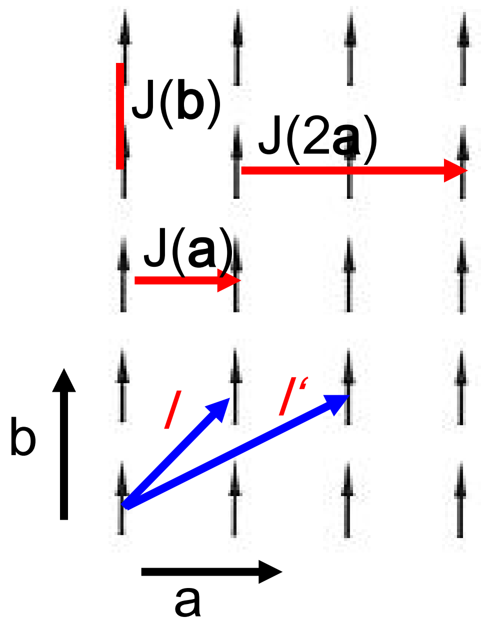
Spin-Wave Theory

The Hamiltonian assuming
Heisenberg exchange interactions

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



Assumption of fully aligned ground state

Excitations are fluctuations about this ground state

Aim to diagonalize the Hamiltonian, find eigenstates and eigenvalues.

The Hamiltonian is put through a series of transformations

1. Ladder operators S^+, S^-, S^z
2. Holstein-Primakoff operators, acting on spin deviations
3. Fourier transform of Holstein-Primakoff operators
4. Bogliubov transformation for antiferromagnets and complex magnets

Spin-Wave Theory

SpinW

By Sandor Toth

http://spinw.org/

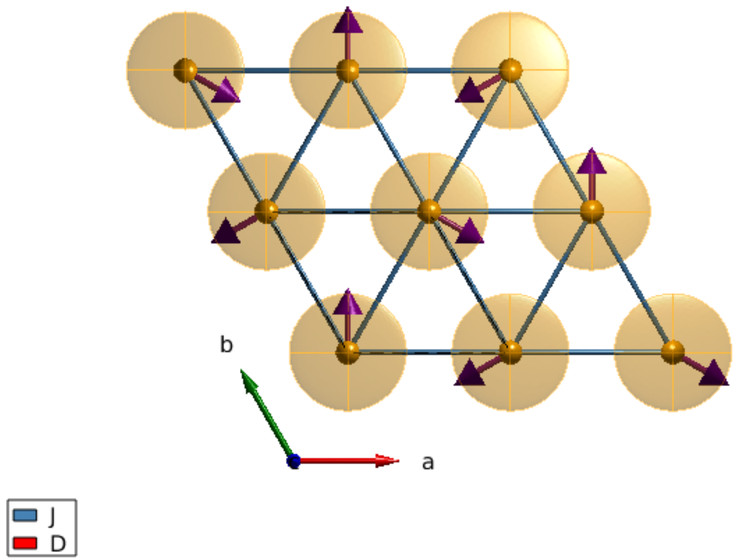
S. Toth and B. Lake,
J. Phys. Condens. Matter 27, 166002 (2014)

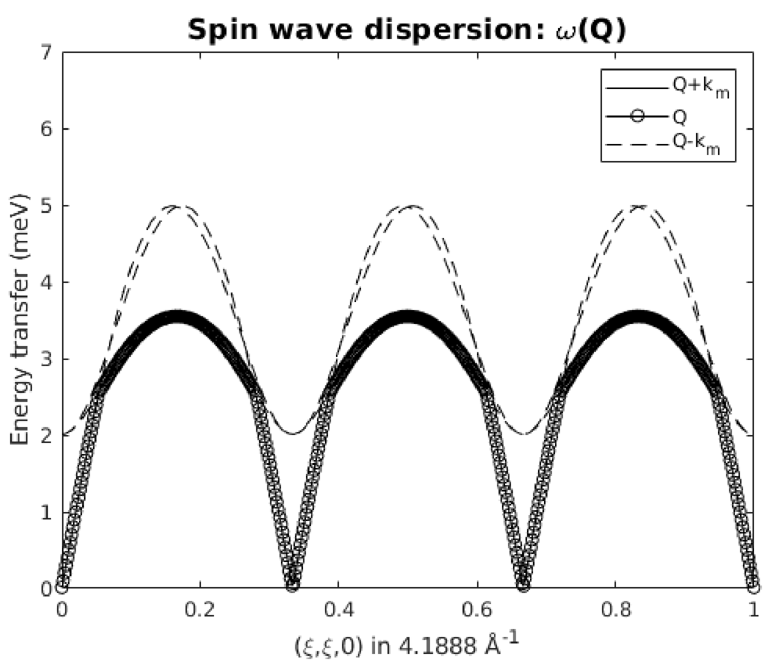
- Spin-waves are characterised by quantum spin number S=1.
- Spin-waves have a well-defined energy as a function of wavevector
- For several magetic ions per unit cell it is necessary to define several sublattices
- The number of spin-wave branches equals the number, *n*, of magnetic ions, 1 acoustic branch and (*n*-1) optic branches.
- Spin-wave theory can also describe helical structures, in which case a rotating coordinate frame can be used.
- Single-ion and exchange anisotropies can also be included.
- Spin-wave models are used to extract value of the exchange interactions

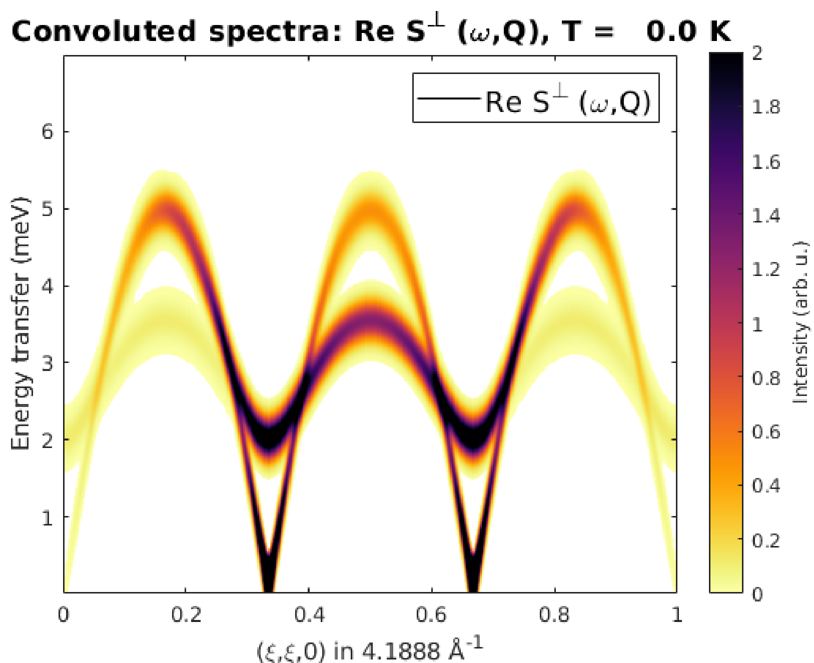
Spin-Wave Theory

Triangular lattice antiferromagnet with easy plane anisotropy

$$H = \sum_{n,m} J_{n,m} \mathbf{S}_n . \mathbf{S}_m + \sum_n D_n \left(\mathbf{S}_n^z \right)^2$$







Inelastic Magnetic Neutron Scattering Cross-Section

Basic Properties of the Neutron

- The neutron has spin angular momentum S_n=1/2
- And magnetic moment $\mu_n = \gamma\mu_N$; γ=-1.913; $\mu_N = e\hbar/m_p$;
- Momentum is $p=m_n v$, and is $p=\hbar k$ (k units Å^{-1})

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

- Its de Broglie wavelength λ (=2π/k) (units Å)

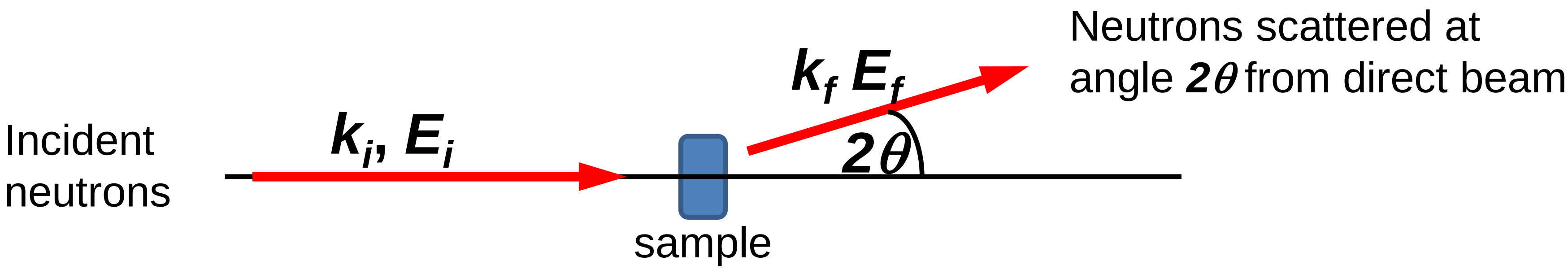
$$\lambda = \frac{2\pi}{k} = \frac{2\pi\hbar}{m_n v}$$

- kinetic energy E (meV where 1eV=1.6x10^{-19})

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

Values of v, λ, k and E are all related

Scattered Neutrons – Differential Neutron Cross-section



- Neutrons are scattered by the sample, the scattered pattern is a function of 2θ characteristic of the sample.
- During scattering 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 spin-wave).
  - Alternatively if the spins are already moving e.g. a spin-wave, it gives this energy to the neutron, the neutron gains energy and the sample loses energy.

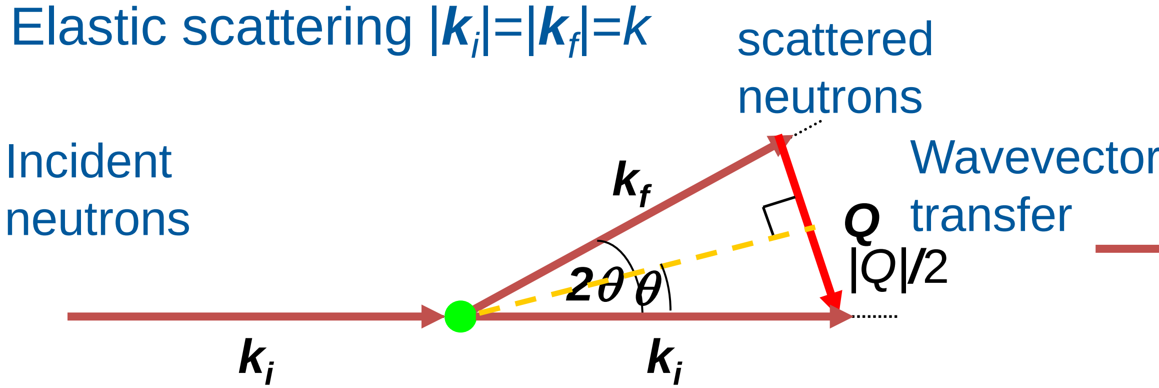
Elastic neutron scattering is when the neutron energy is unchanged. $E_i = E_f$

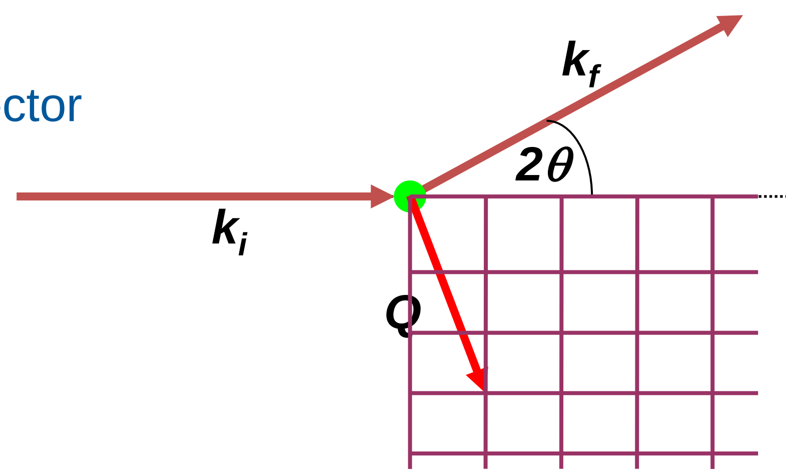
Inelastic scattering is when the neutron gains or loses energy, $E_i \neq E_f$

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





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

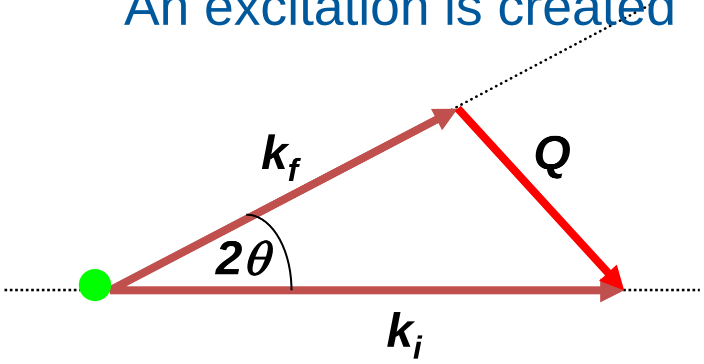
Scattering triangles – Inelastic scattering

- Conservation of energy and momentum

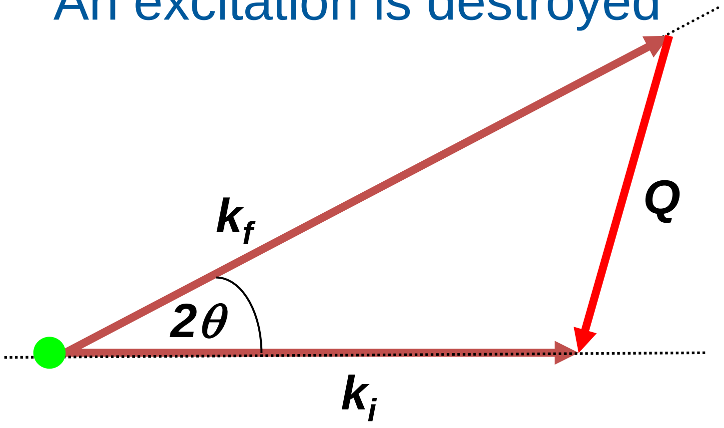
$$E_i - E_f = \frac{1}{2}mv_i^2 - \frac{1}{2}mv_f^2 = \frac{1}{2m}\hbar^2\left(k_i^2 - k_f^2\right) = \hbar\omega \qquad \boldsymbol{Q} = \boldsymbol{k}_i - \boldsymbol{k}_f$$

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

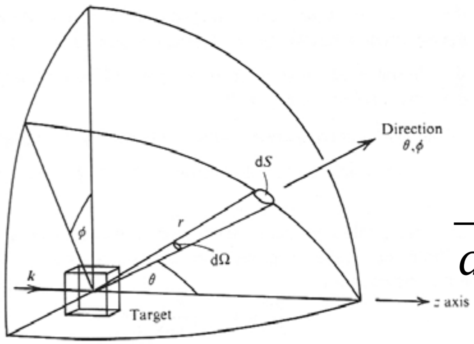
Neutron loses energy
An excitation is created



Neutron gains energy
An excitation is destroyed



Differential Neutron Scattering Cross-Section



$$\frac{d^2\sigma}{d\Omega dE} = \begin{array}{l}\text{number of neutrons scattered per second} \\ \text{into solid angle } d\Omega \text{ and } dE \;/\; \Phi\, d\Omega\, dE\end{array}$$

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

Probability of being in the initial state

Matrix element for moving from initial to final state

Energy conservation

$$E = \hbar\omega = \frac{\hbar^2}{2m}\left(k_i^2 - k_f^2\right)$$

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

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

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

The Magnetic Cross-section

Inelastic cross section for spin only scattering by ions

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

Dynamical structure factor

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

$F(\mathbf{Q})$ 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

Distinguishing Phonons and Magnons with Neutrons

Wavevector-dependence

- Phonon excitations have high intensity at large |Q| and when **Q** is parallel to the mode of vibration
- Magnetic excitations have high intensity at low |Q| and when **Q** is perpendicular to the magnetic moment direction

Temperature dependence

- Phonon excitations become stronger as temperature increases
- Magnetic excitations become weaker as temperature increases

Measuring
Spin-Waves

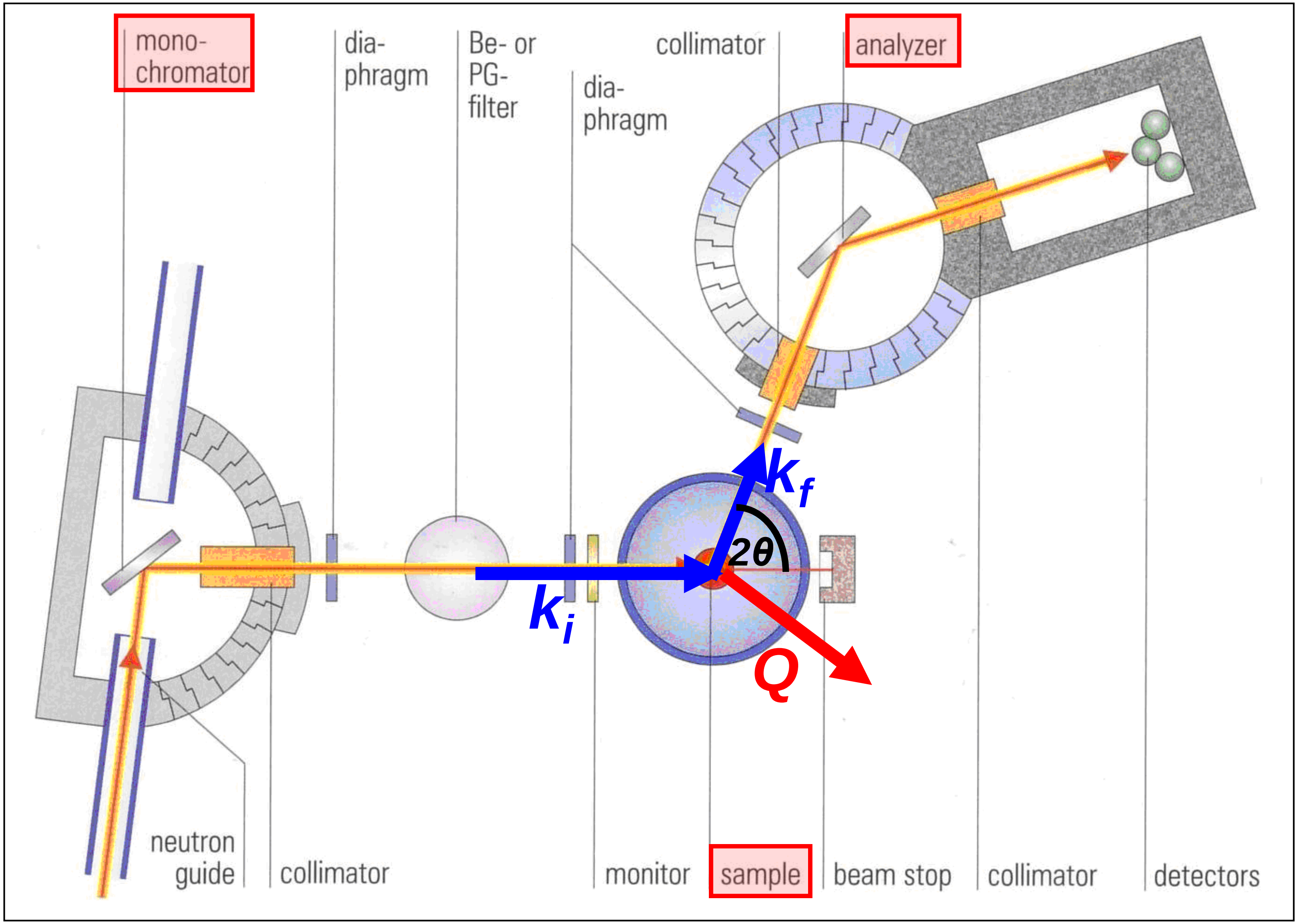
Instruments for Measuring Inelastic Scattering

Inelastic neutron scattering
-both the initial and final neutron energy must be known

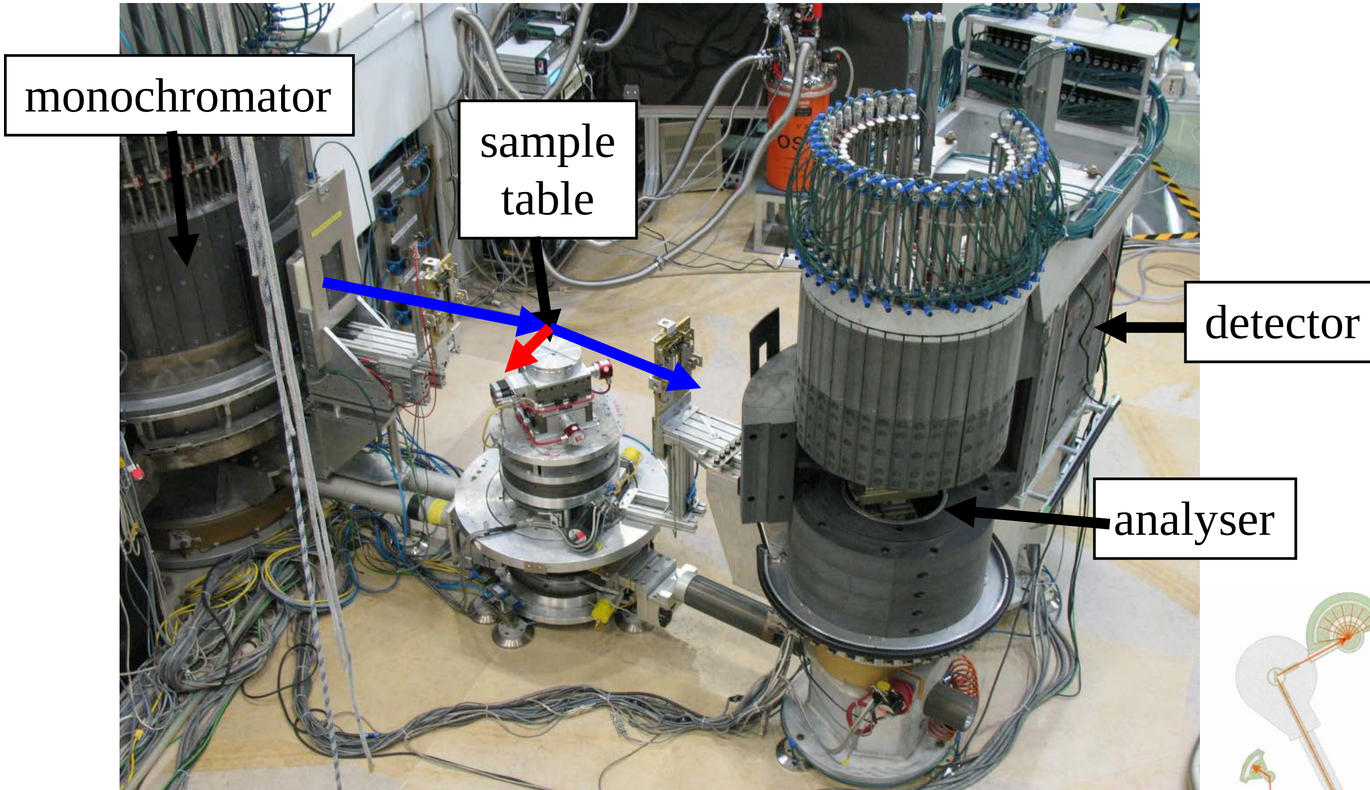
Triple-axis spectrometer
The initial and final neutron energies can be selected or measured using monochromator and analyser crystals where the wavelength of the neutrons is determined by the scattering angle.

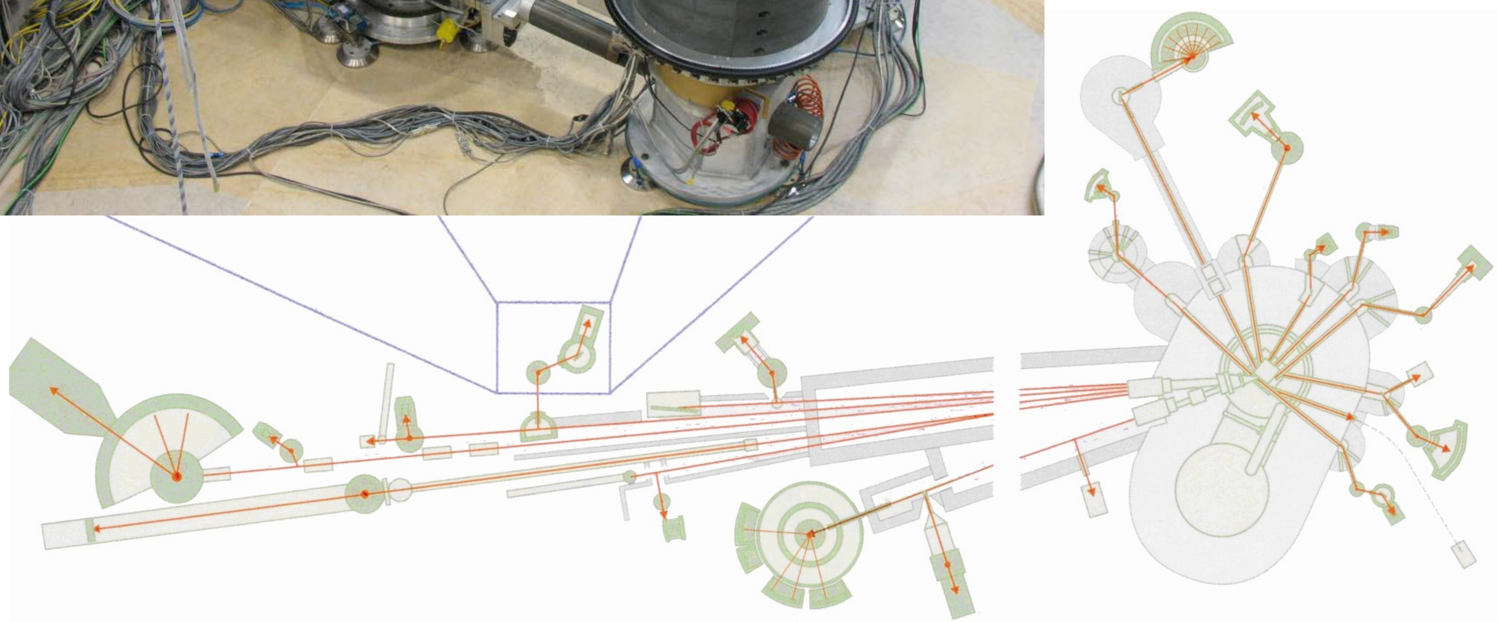
Time-of-flight Spectrometer.
The initial and final energies are selected or measured using the time it takes the neutron to travel through spectrometer to the detector from this the velocity and hence kinetic energy are deduced.

The Triple Axis Spectrometer - Layout



The Triple Axis Spectrometer – V2/FLEX, HZB



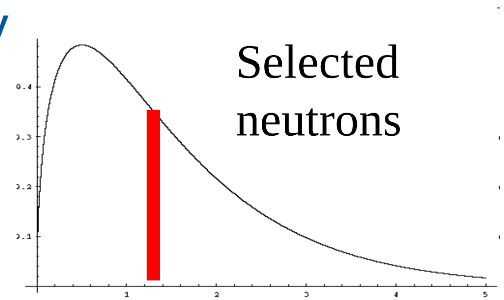


The Triple Axis Spectrometer – Monochromator Analyser

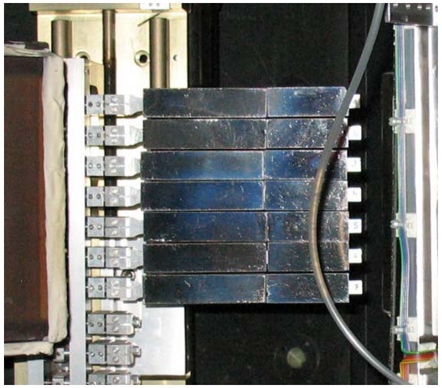
The monochromator is a crystalline material and selects a single wavelength from the white neutron beam of the reactor/spallation source by Bragg scattering where the scattering angle is chosen to select λ. The analyser measures the final neutron energy

$$2d \sin\theta = n\lambda$$

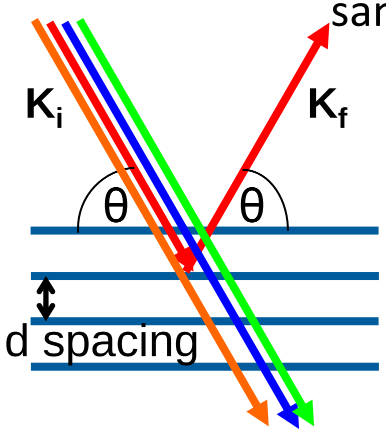
$$n = 1, 2, 3 \ldots$$

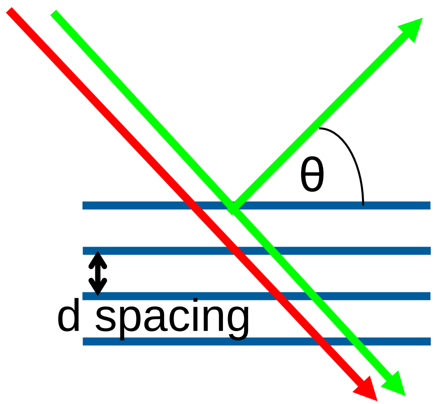


Vertically focusing monochromator

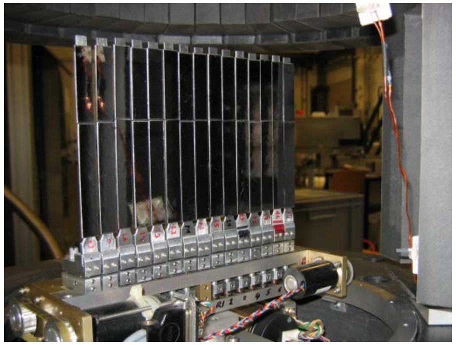


from graphite, Copper, Germanium, blades can be focused





Horizontally focusing analyser



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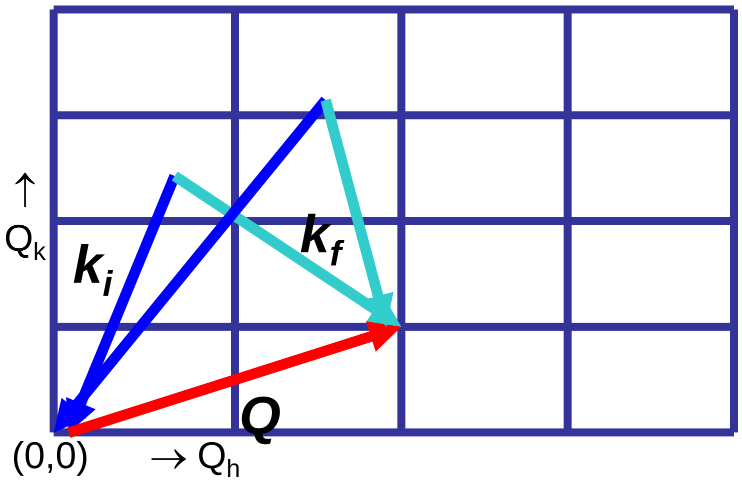
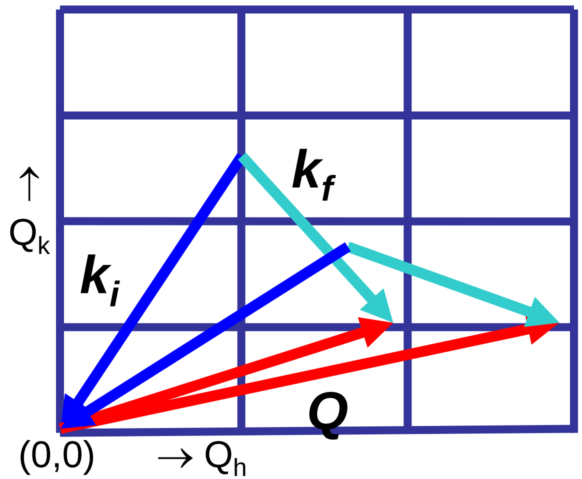
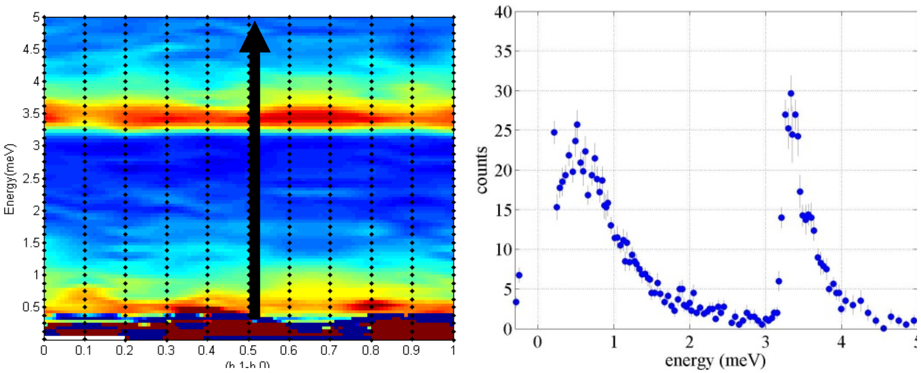
The Triple Axis Spectrometer – Measurements

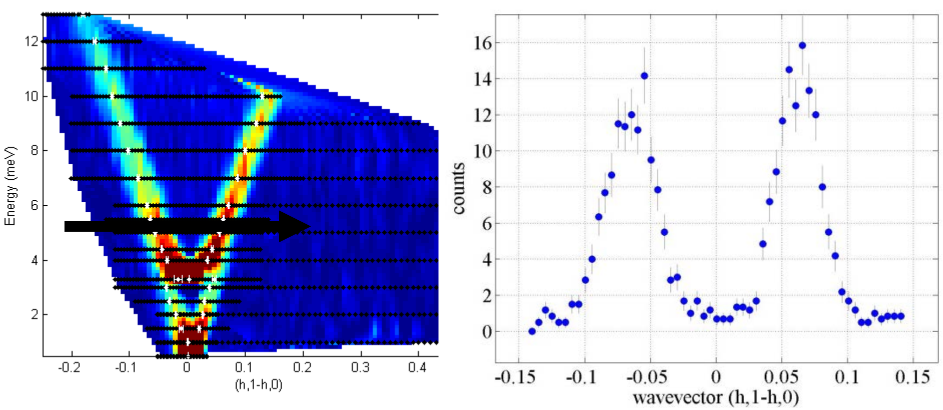
Keep wavevector transfer constant and scan energy transfer.



Keep energy transfer constant and scan wavevector transfer.







Triple Axis Spectrometer – Pros and Cons

Advantages

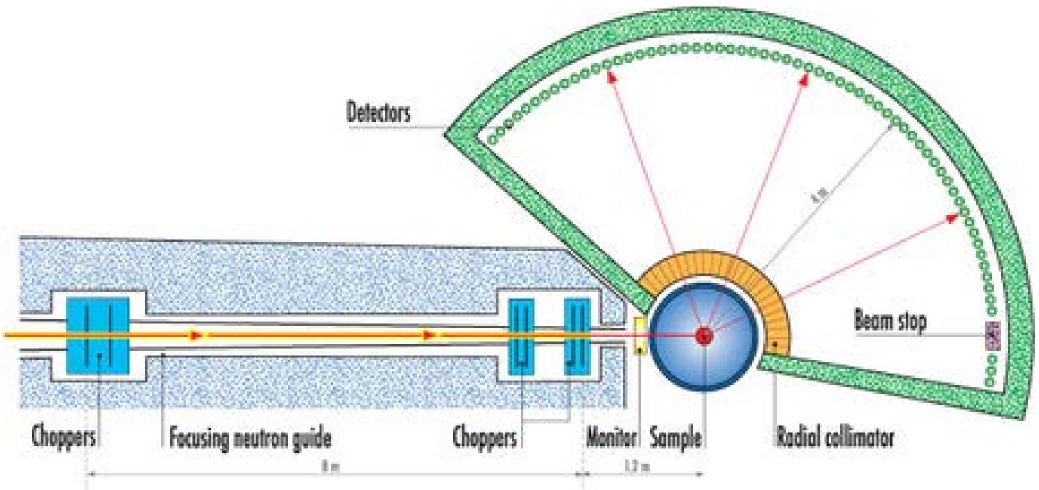
- Can focus all intensity on a specific point in reciprocal space
- Can make measurements along high-symmetry directions
- Can use focusing and other ‘tricks’ to improve the signal/noise ratio
- Can use polarisation analysis to separate magnetic and phonon signals

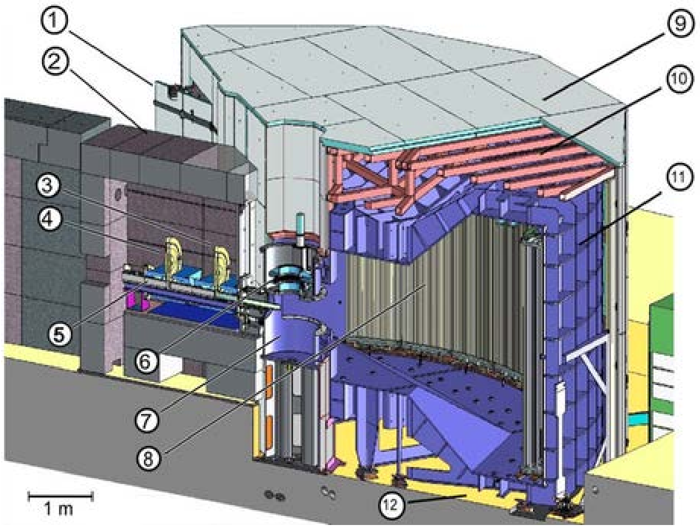
Disadvantages

- Technique is slow and requires some expert knowledge
- Use of monochromator and analyser crystals gives rise to possible higher-order effects that are known as “spurions”
- With measurements restricted to high-symmetry directions it is possible that unexpected signal might be missed

Time of Flight Spectrometer – Layout of V3/NEAT

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





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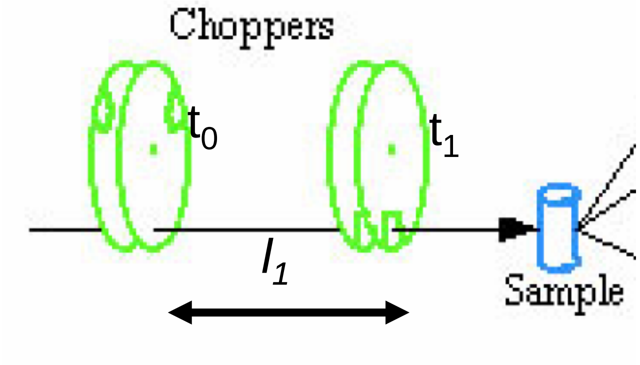
Time of Flight Spectrometer - Choppers

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

Ist chopper rotates and lets neutrons through once per revolution and sets initial time t_0

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



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

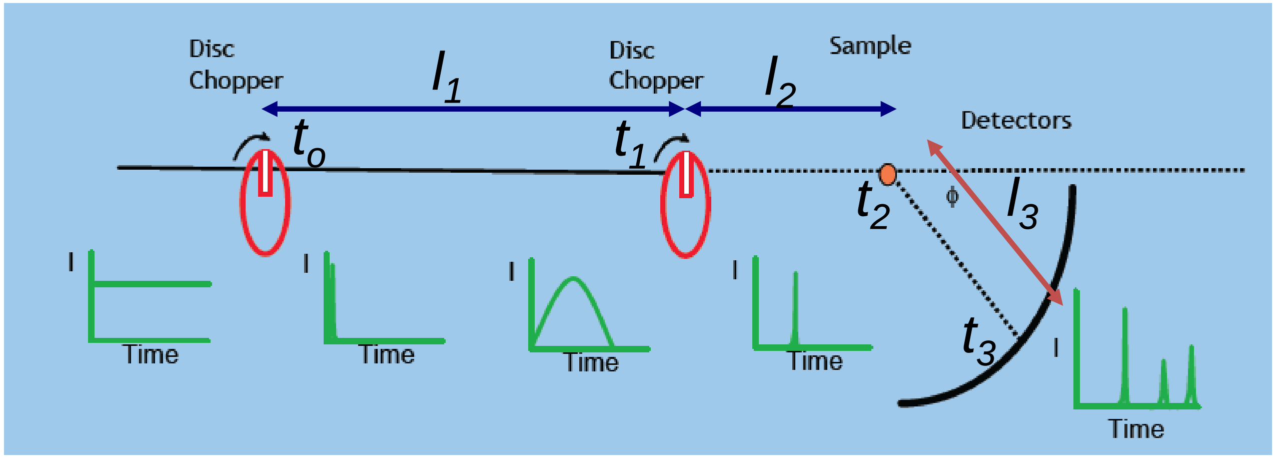
$$E_i = \frac{m v_i^2}{2} = \frac{m l_1^2}{2(t_1 - t_0)^2}$$

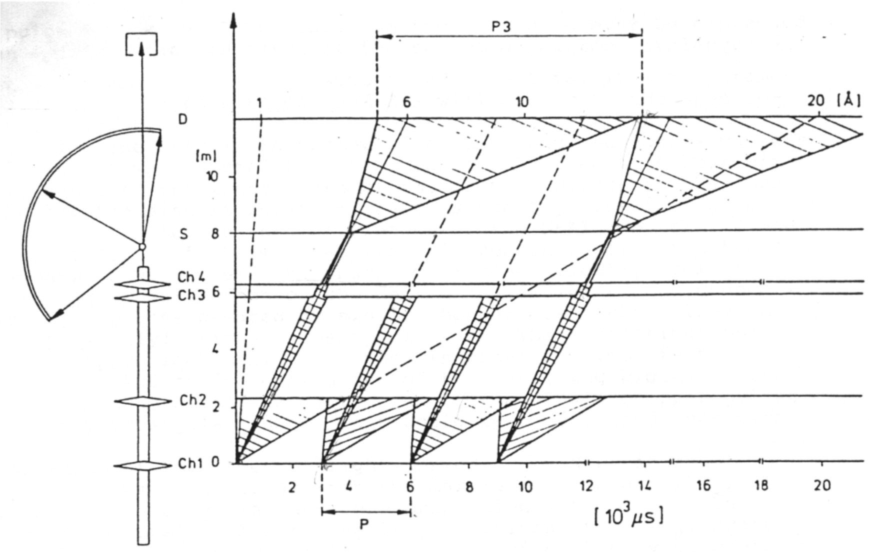
After scattering at the sample the detector again measures time as well as number of neutrons, thus the velocity and energy of the scattered neutrons is known.

The Time of Flight Spectrometer - Choppers

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

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



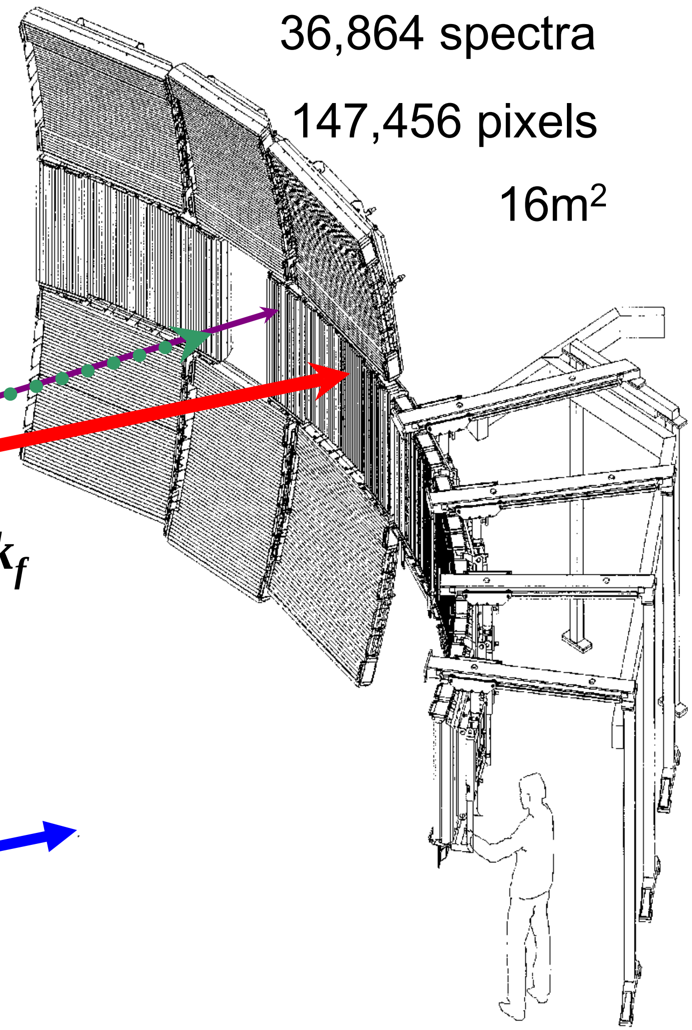


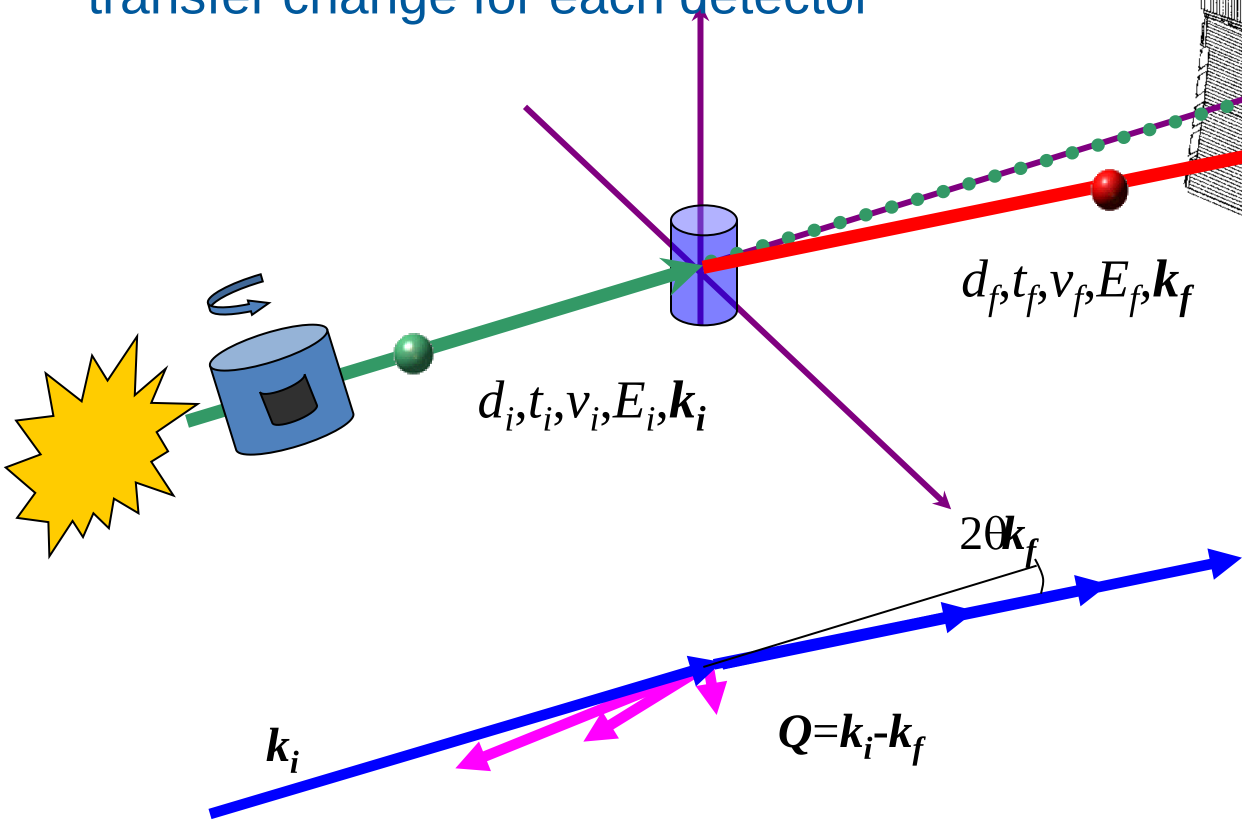
- First chopper sets the initial time.
- Second chopper sets the initial energy
- Detectors measure final time and energy.

Time of Flight Spectrometer – Detectors

MAPS time-of-flight spectrometer

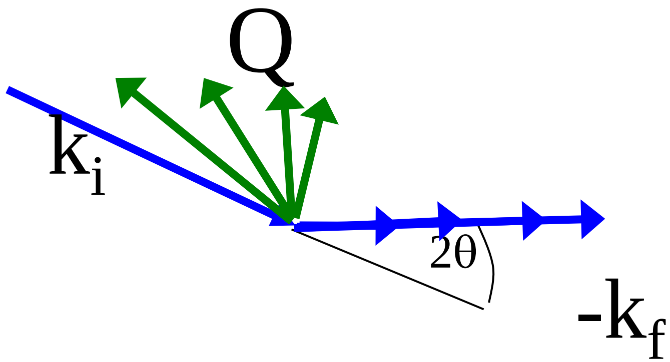
As time of flight is changed both energy transfer and wavevector transfer change for each detector

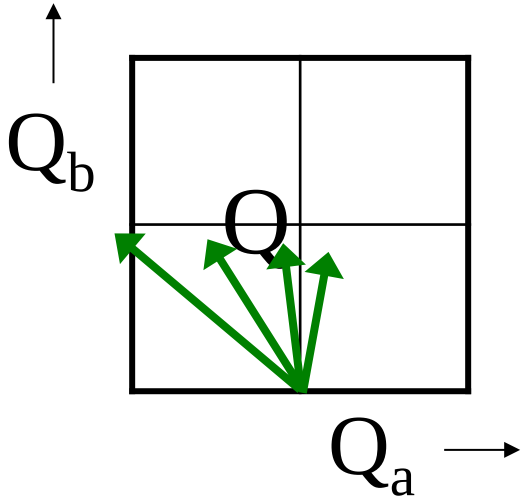




$$E = E_i - E_f = \frac{\hbar^2}{2m_n}\left(k_i - k_f^2\right) = \frac{1}{2}m_n\left(v_i^2 - v_f^2\right) = \frac{1}{2}m_n\left[\left(\frac{d_i}{t_i}\right)^2 - \left(\frac{d_f}{t_f}\right)^2\right]$$

Time of Flight Spectrometer – Measuring

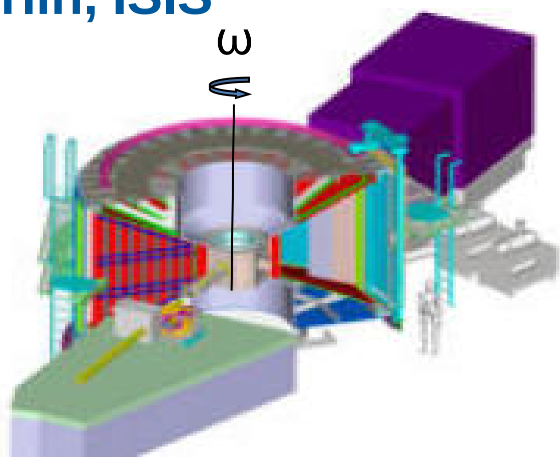


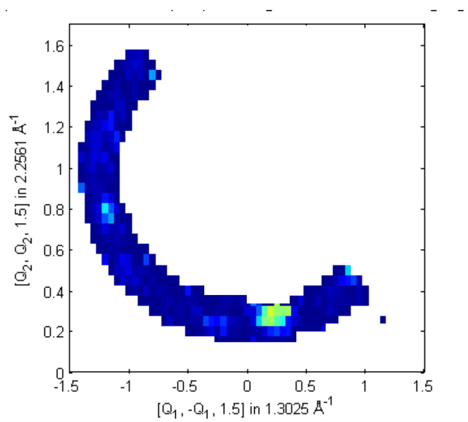


- Every detector trances a different path in E and Q transfer
- A large dataset is obtained from all detectors containing intensity as a function of three dimensional wavevector and energy

Single Crystal Inelastic Neutron Scattering

Merlin, ISIS



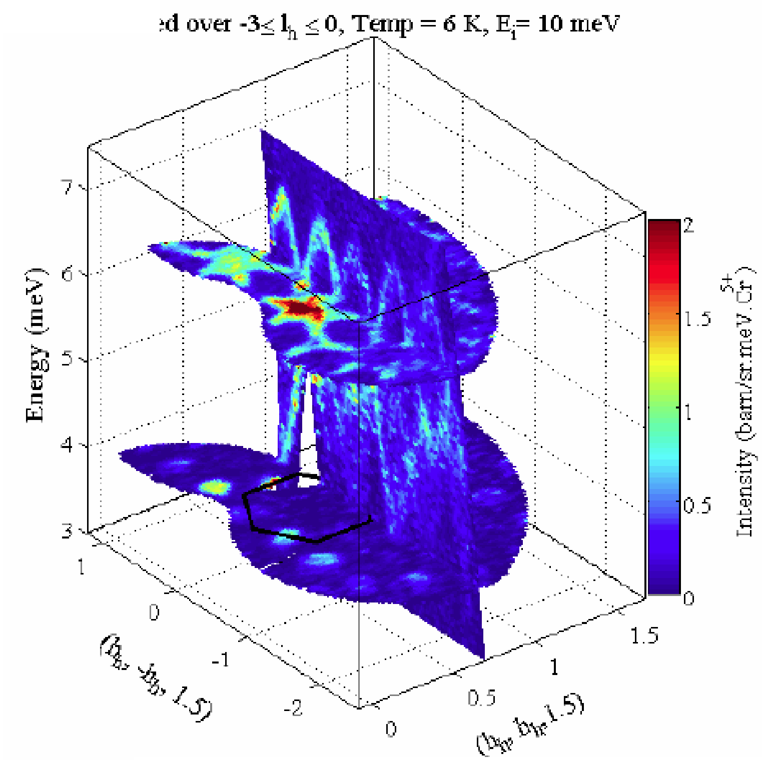


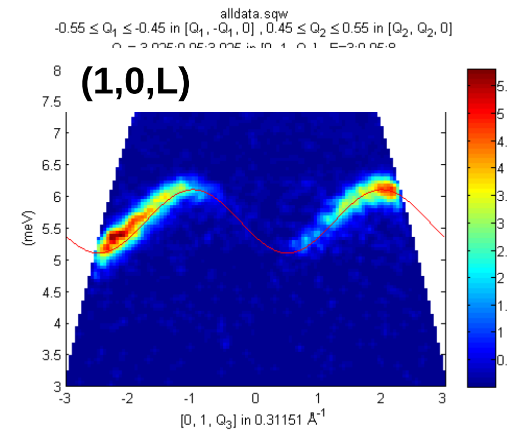
Individual scans combined to create a single file $S(Q_h, Q_k, Q_l, E)$.

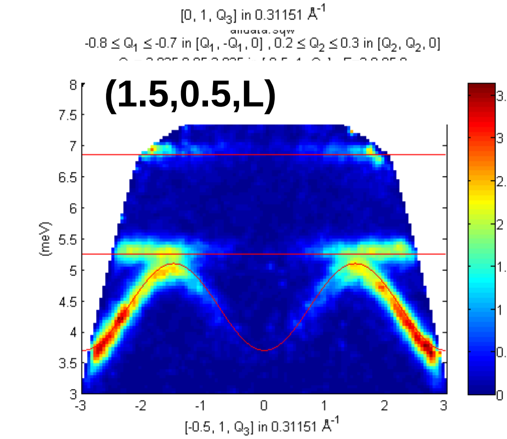
large region of the energy and reciprocal space.

detectors:
180° horizontal
±30° vertical

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







B. Lake; Oxford, Sept 2019

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

Time of Flight Spectrometer – Pros and Cons

Advantages

- It is possible to simultaneously measure a large region of energy and wavevector space and get an overview of the excitations
- This allows unexpected phenomena to be observed
- It does not have the same problem of second order scattering as the triple axis spectrometer

Disadvantages

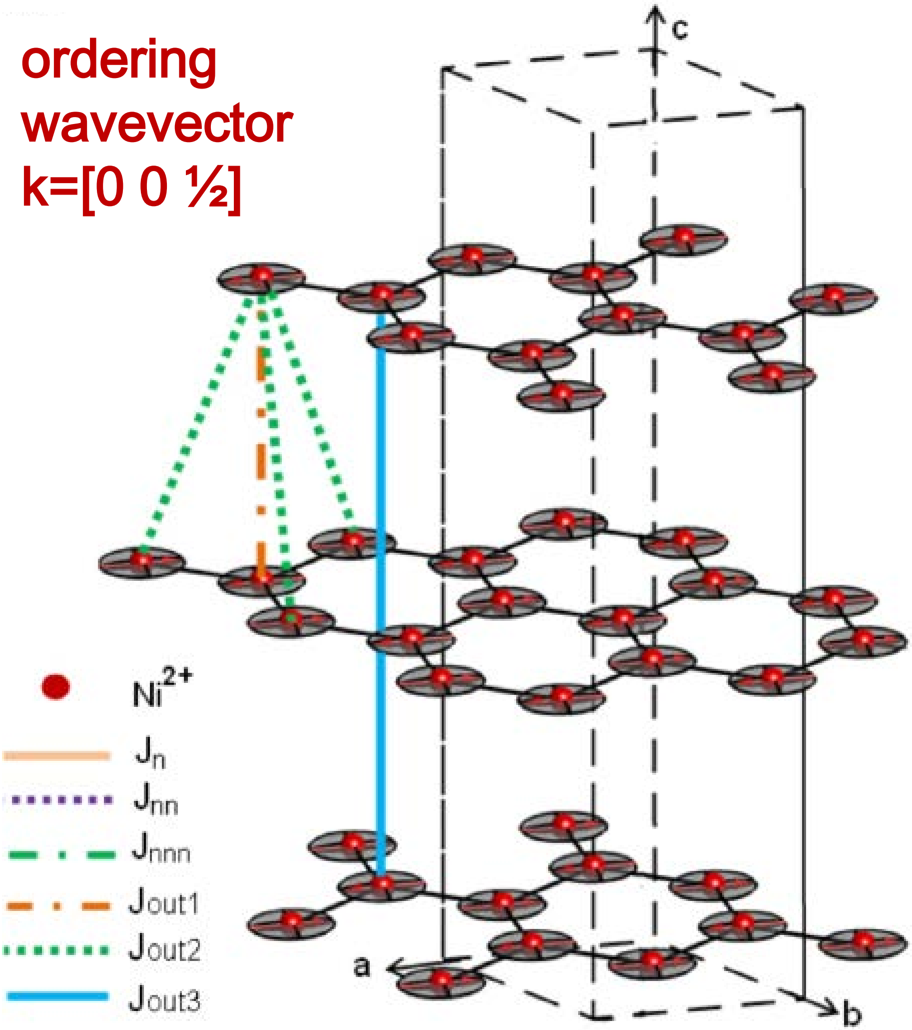
- Time-of-flight instrument have low neutron flux for an specific wavevector and energy but the ESS will be different
- It is difficult to do polarised neutron scattering

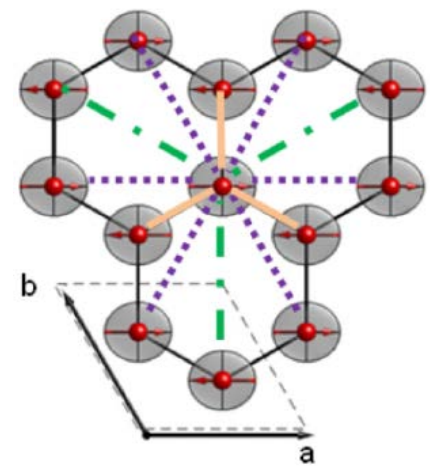
Example

Spin-Waves in $BaNi_2V_2O_8$

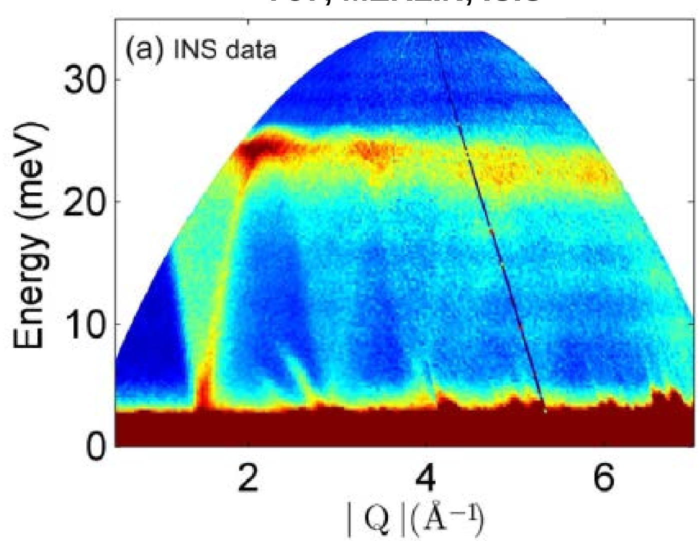
$$H = \sum_{i>j} J_n \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} J_{nn} \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} J_{nnn} \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} J_{out} \cdot \boldsymbol{S_i} \cdot \boldsymbol{S_j} + \sum_{i>j} D_{EP} \cdot S_i^{c^2}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

ordering wavevector k=[0 0 ½]





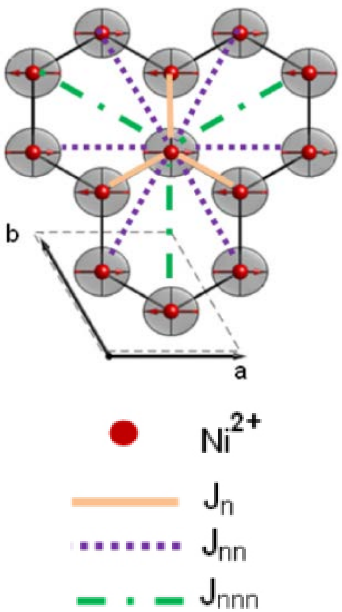
TOF, MERLIN, ISIS

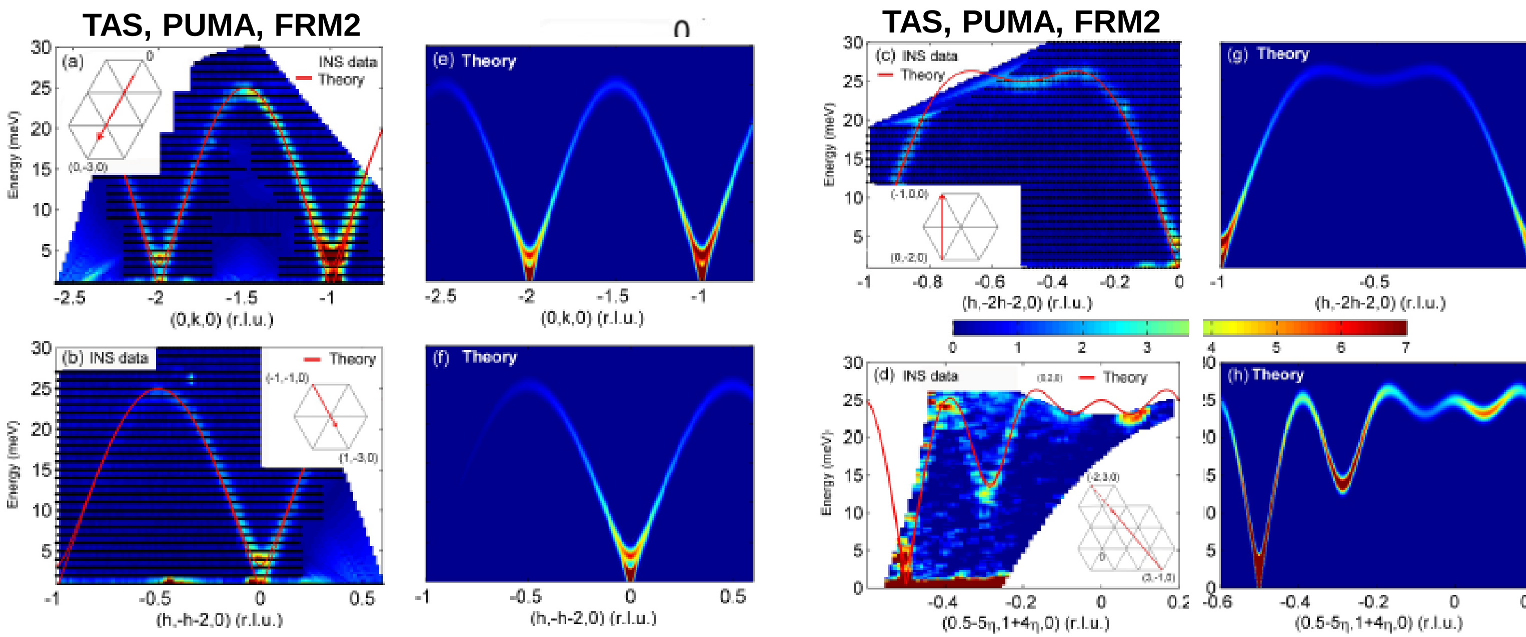


B. Lake; Oxford, Sept 2019

Spin-Waves in $BaNi_2V_2O_8$

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





k=[0 0 ½]

1st neighbor interaction

10.9 meV< J_n <11.8meV

2nd neighbor interaction

1.1meV< J_{nn} <0.65meV

3rd neighbors interaction

-0.1meV< J_{nnn} <0.4meV

Interplane coupling

J_{out} <0.0001meV

Easy-plane anisotropy

0.8< D_{EP} <0.73

Easy–axis anisotropy

-0.00105< D_{EA} <-0.0009

Summary

Conventional Magnets

- long-range magnetic order, spin-wave excitations

Inelastic Magnetic Neutron Scattering Cross-Section

Measuring spin-waves

- Triple-axis and time-of-flight spectrometers