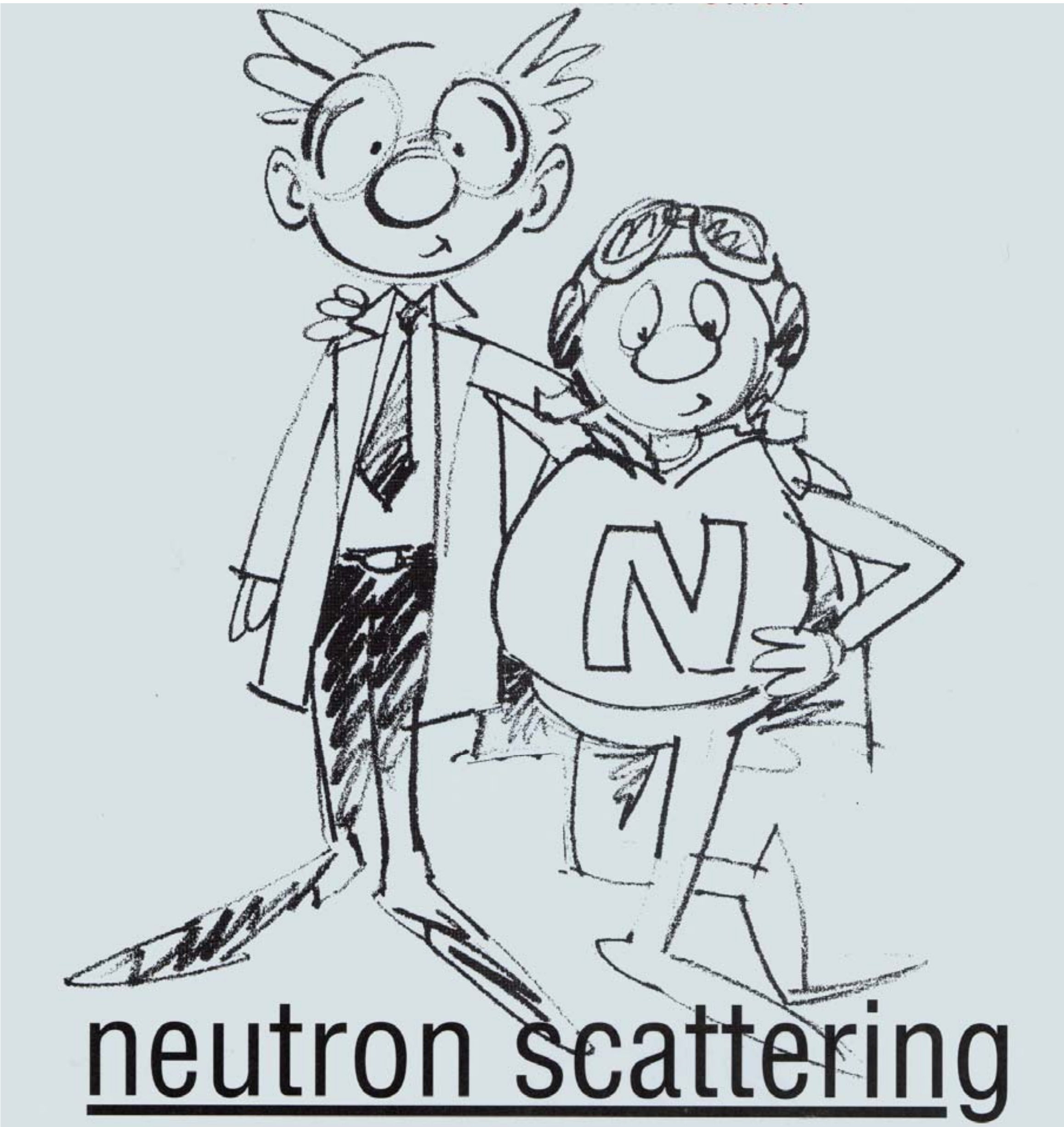

Magnetic Scattering

by

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Indiana University
&
Spallation Neutron Source

This Lecture

- Short revision:
  - Magnetic properties of the neutron
  - Magnetic scattering of neutrons
- Inelastic magnetic scattering of neutrons
- Scattering by unpaired electrons
- The effect of magnetic scattering on the neutron's spin state
- Examples of inelastic magnetic neutron scattering
- Spin waves in ferromagnets
- Kinematics of neutron scattering
- Triple axis spectrometers
  - Spin waves and Stoner modes in iron
- TOF spectrometers
  - Spin waves in cobalt
- Magnetic excitations in chromium
- Crystal field excitations
- Low dimensional magnets
  - $CsNiF_3$ (a 1d easy plane FM)
  - TMMC (a 1d easy plane AFM)
  - $KCuF_3$ (a singlet ground state chain)

Magnetic Properties of the Neutron

- The neutron has a magnetic moment of -9.649 x 10^{-27} JT^{-1}

$$\vec{\mu}_n = -\gamma \mu_N \vec{\sigma}$$

where $\mu_N = {e\hbar}/{2m_p}$ is the nuclear magneton,

$m_p =$ proton mass, $e =$ proton charge and $\gamma = 1.913$

$\vec{\sigma}$ is the Pauli spin operator for the neutron. Its eignevalues are ± 1

- Note that the neutron's spin and magnetic moment are antiparallel
- Because of its magnetic moment, the neutron feels a potential given by:

$$V_m(\vec{r}) = -\vec{\mu}_n.\vec{B}(\vec{r}) \quad \text{where} \quad \vec{B}(\vec{r}) = \mu_0 \mu \vec{H}(\vec{r}) = \mu_0[\vec{H}(\vec{r}) + \vec{M}(\vec{r})]$$

- Thus the neutron senses the distribution of magnetization in a material
- Homework problems: What is the Zeeman energy in meV of a neutron in a 1 Tesla field? At what temperature is the Boltzmann energy equal to this Zeeman energy? What is the effective scattering length of a "point" magnetic moment of one Bohr magneton?

Magnetic Scattering of the Neutron

- For nuclear scattering, the matrix element that appears in the expression for the scattering cross section is: $\sum_j b_j e^{i\vec{Q}.\vec{R}_j}$

- The equivalent matrix element for magnetic scattering is:

$$\gamma r_0 \frac{1}{2\mu_B} \vec{\sigma}.\vec{M}_\perp(\vec{Q}) \quad \text{where } \mu_B = \frac{e\hbar}{2m_e} \text{ is the Bohr magneton } (9.27 \text{ x } 10^{-24} \text{ JT}^{-1})$$

$$\text{and } r_0 = \frac{\mu_0}{4\pi} \frac{e^2}{m_e} \text{ is classical radius of the electron } (2.818 \text{ x } 10^{-6} \text{ nm})$$

- Here $\vec{M}_\perp(\vec{Q})$ is the component of the Fourier transform of the magnetization that is perpendicular to the scattering vector $\vec{Q}$. This form arises directly from the dipolar nature of the magnetic interaction.

- Unlike the neutron-nucleus interaction, the magnetic interaction of the neutron with a scattering system specifically depends on neutron spin

Derivation

The $\vec{\mathrm{B}}$ field at distance $\vec{\mathrm{R}}$ from a magnetic moment $\vec{\mathrm{M}}$ is

$$\frac{\mu_0}{4\pi}\vec{\nabla}\times\left(\frac{\vec{M}\times\vec{R}}{R^3}\right)=-\frac{\mu_0}{4\pi}\vec{\nabla}\times\left(\vec{M}\times\nabla(1/R)\right)$$

Since $\displaystyle\int\frac{1}{q^2}\exp(i\vec{q}.\vec{R})d\vec{q}=2\pi\int_0^\infty dq\int_{-1}^{1}\exp(iqR\cos\theta)d(\cos\theta)=4\pi\int_0^\infty\frac{\sin(qR)}{qR}dq=\frac{2\pi^2}{R}$

$$\vec{\nabla}\times\left(\frac{\vec{M}\times\vec{R}}{R^3}\right)=-\frac{1}{2\pi^2}\int\frac{1}{q^2}\vec{\nabla}\times\left(\vec{M}\times\nabla\{\exp i\vec{q}.\vec{R}\}\right)d\vec{q}$$

But $\vec{M}\times\nabla\{\exp i\vec{q}.\vec{R}\}=i\vec{M}\times\vec{q}\exp i\vec{q}.\vec{R}$ and $\vec{\nabla}\times\vec{M}\times\vec{q}\exp i\vec{q}.\vec{R}=i\vec{q}\times\vec{M}\times\vec{q}\exp i\vec{q}.\vec{R}$

so $\displaystyle\vec{\nabla}\times\left(\frac{\vec{M}\Lambda\vec{R}}{R^3}\right)=\frac{1}{2\pi^2}\int\frac{1}{q^2}\vec{q}\times\left(\vec{M}\times\vec{q}\right)\{\exp i\vec{q}.\vec{R}\}d\vec{q}=\frac{1}{2\pi^2}\int\vec{M}_\perp(\vec{q})\{\exp i\vec{q}.\vec{R}\}d\vec{q}$

The Magnetic Scattering Cross Section

- Development of the magnetic scattering cross section follows the same formalism as for the nuclear cross section, with nuclear matrix element replaced by the magnetic interaction matrix element given above

- Need to keep the explicit dependence on neutron spin (or average over neutron spin states for an unpolarized neutron beam).
  - Magnetic scattering may cause a change in the neutron's spin state

- General expressions tend to be complicated, so specific expressions are obtained for various contributions to sample magnetization e.g. unpaired electron spins

- Homework: compare typical coherent nuclear scattering lengths (a few fm) with the magnetic scattering length for an atom with a one Bohr magneton magnetic moment. What do you notice? What is the magnitude of magnetic scattering if all the sample moments are parallel to the scattering vector, Q?

What Happens to a Neutron's Spin When the Neutron is Scattered?

- The cross section for magnetic scattering that takes the neutron spin state from σ->σ' and the scattering system from λ->λ' is:

$$\left(\frac{d^2\sigma}{d\Omega.dE}\right)_{\sigma\lambda\to\sigma'\lambda'} = \left(\frac{\gamma r_0}{2\mu_B}\right)^2 \frac{k'}{k}\left|\left\langle\sigma'\lambda'\left|\vec{\sigma}.\vec{M}_\perp(\vec{Q})\right|\sigma\lambda\right\rangle\right|^2 \delta(E_\lambda - E\lambda' + \hbar\omega)$$

- One can show (see Squires) that if $\left|u\right\rangle, \left|v\right\rangle$ are the neutron spin eigenstates:

$$\left\langle u\left|\vec{\sigma}.\vec{M}_\perp\right|u\right\rangle = M_{\perp z};\quad \left\langle v\left|\vec{\sigma}.\vec{M}_\perp\right|v\right\rangle = -M_{\perp z};\quad \left\langle v\left|\vec{\sigma}.\vec{M}_\perp\right|u\right\rangle = M_{\perp x} + iM_{\perp y};\quad \left\langle u\left|\vec{\sigma}.\vec{M}_\perp\right|v\right\rangle = M_{\perp x} - iM_{\perp y}$$

so, components of M_{perp} parallel to the neutron's magnetic moment (z) does not change the neutron spin, whereas perpendicular components of M_{perp} 'flip' the neutron's spin

- Homework: show that for a paramagnet (where $\left\langle S_i^\alpha S_j^\beta\right\rangle = \frac{1}{3}\delta_{ij}\delta_{\alpha\beta}S(S+1)$ for spins i and j)
  - If z is parallel to **Q**, the scattering is entirely spin flip
  - If z is perpendicular to **Q**, half the scattering is spin flip

Inelastic Scattering by Ions with Unpaired Electrons

- Including only magnetization due to unpaired electron spins and assuming an unpolarized incident neutron beam:

$$\frac{d^2\sigma}{d\Omega . dE} = \frac{(\gamma r_0)^2}{2\pi\hbar}\frac{k'}{k}\sum_{\alpha,\beta}(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)\sum_{ldl'd'} F^*_{d'}(\vec{Q}) F_d(\vec{Q})$$

$$\text{x} \int_{-\infty}^{\infty} dt \left\langle \exp\{-i\vec{Q}.\vec{R}_{l'd'}(0)\} \exp\{i\vec{Q}.\vec{R}_{ld}(t)\} \right\rangle . \left\langle S^\alpha_{l'd'}(0) S^\beta_{ld}(t) \right\rangle e^{-i\omega t}$$

where $F_d(Q)$ is the Fourier transform of the electron spin density around atom *d,* often called the atomic form factor; S^α is the α component of the electron spin and *l,d* labels an atom *d* in unit cell *l*

- This expression can be manipulated to give the scattering cross sections for elastic magnetic scattering, inelastic magnetic scattering and magneto-vibrational scattering

“High” and “Low” Temperature

- At high T, electron spins often behave as if they are free -- paramagnet
- They can be progressively screened as T decreases by coherent reorganization of the conduction electrons leading to Kondo screening and mixed valency
- In metals, the spins often condense into ordered states – FM, AFM, SDW, CDW, SC
- A spin or orbital excitation of these states appears – the momentum dependence of excitation energy gives a measure of magnetic interaction (crystal field, spin-orbit, exchange interaction etc)
- Neutron can probe not only the linear excitations but have also discovered a host of exotic magnetic excitations
  - The underlying reason is that the neutron is a spin-1 probe – it links energy levels that differ by S = 1

Inelastic Magnetic Neutron Scattering

- Conventional spin waves (FM and AFM) in systems with itinerant & localized electrons
- Stoner modes
- Crystal field excitations
- Excitations in low dimensional magnets
  - Effects of Hamiltonian symmetry (Ising, XY, Heisenberg)
  - Effects of spin value (Haldane gap for S=1 and not S=1/2)
- Non-linear magnetic excitations (solitons, spinons, beathers etc)
- Diffusive versus propagating excitations
- Magnetic critical scattering
- Spin glasses
- Frustrated magnets
- Singlet ground state systems
- High temperature superconductors
- Correlated electron systems
- Quantum fluctuations and QCPs

Inelastic Magnetic Scattering of Neutrons

- In the simplest case, atomic spins in a ferromagnet precess about the direction of mean magnetization

$$H = \sum_{l,l'} J(\vec{l} - \vec{l}\,')\vec{S}_l.\vec{S}_{l'} = H_0 + \sum_q \hbar\omega_q b_q^+ b_q$$

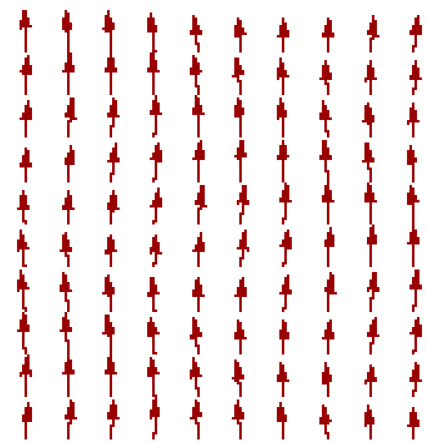
Heisenberg interaction

spin waves (magnons)

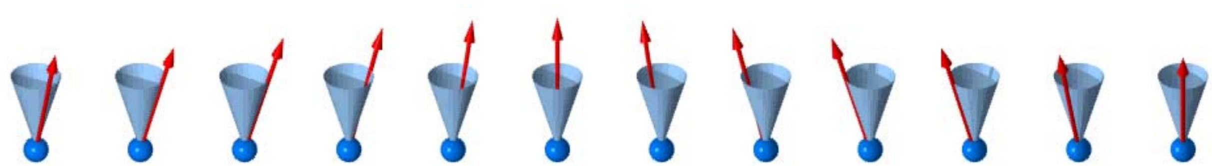
with

$$\hbar\omega_q = 2S(J_0 - J_q) \qquad \text{where} \quad J_q = \sum_l J(\vec{l}\,)e^{i\vec{q}.\vec{l}}$$

$\hbar\omega_q = Dq^2$ is the dispersion relation for a ferromagnet



Fluctuating spin is perpendicular to mean spin direction

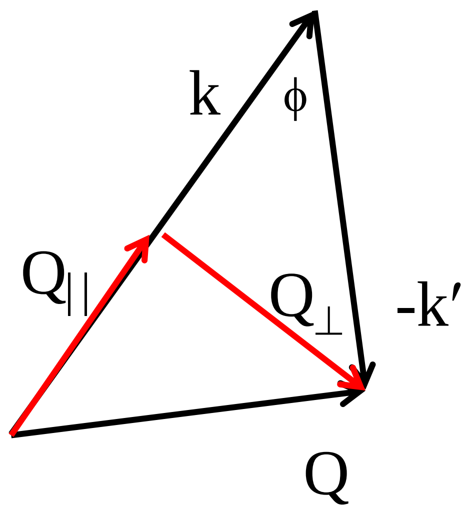


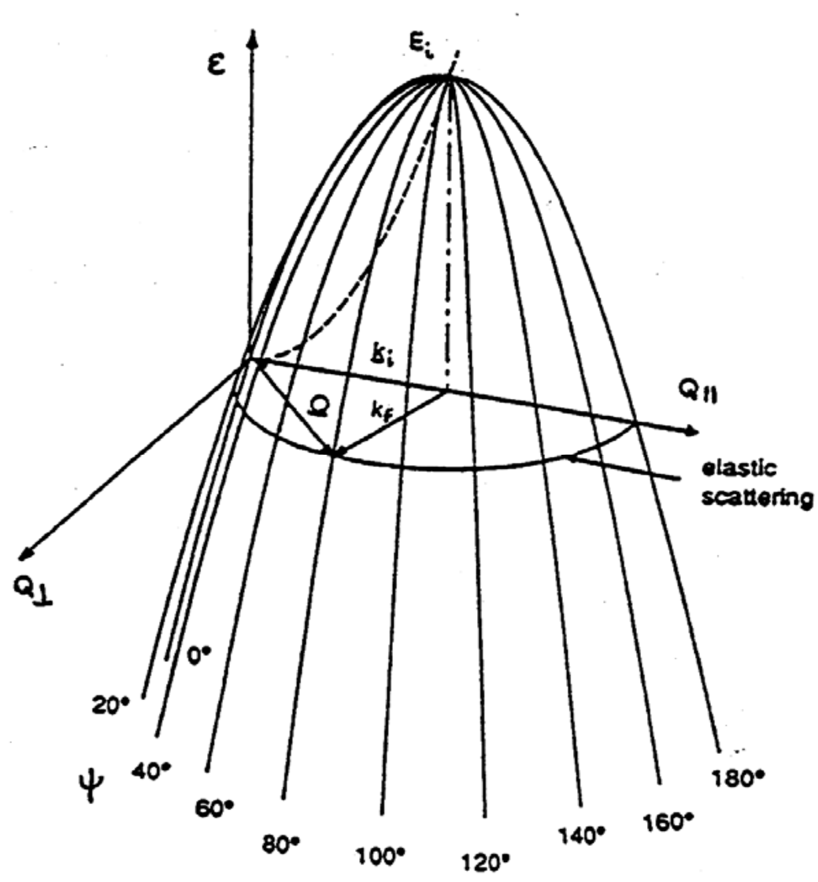
Spin wave animation courtesy of A. Zheludev (ORNL)

Kinematics of Neutron Scattering

- Conservation of energy and momentum applies to scattering and limits the accessible Q and ω values.

$$\vec{Q} = \vec{k} - \vec{k}' \quad \text{and} \quad \hbar\omega = E_i - E_f$$

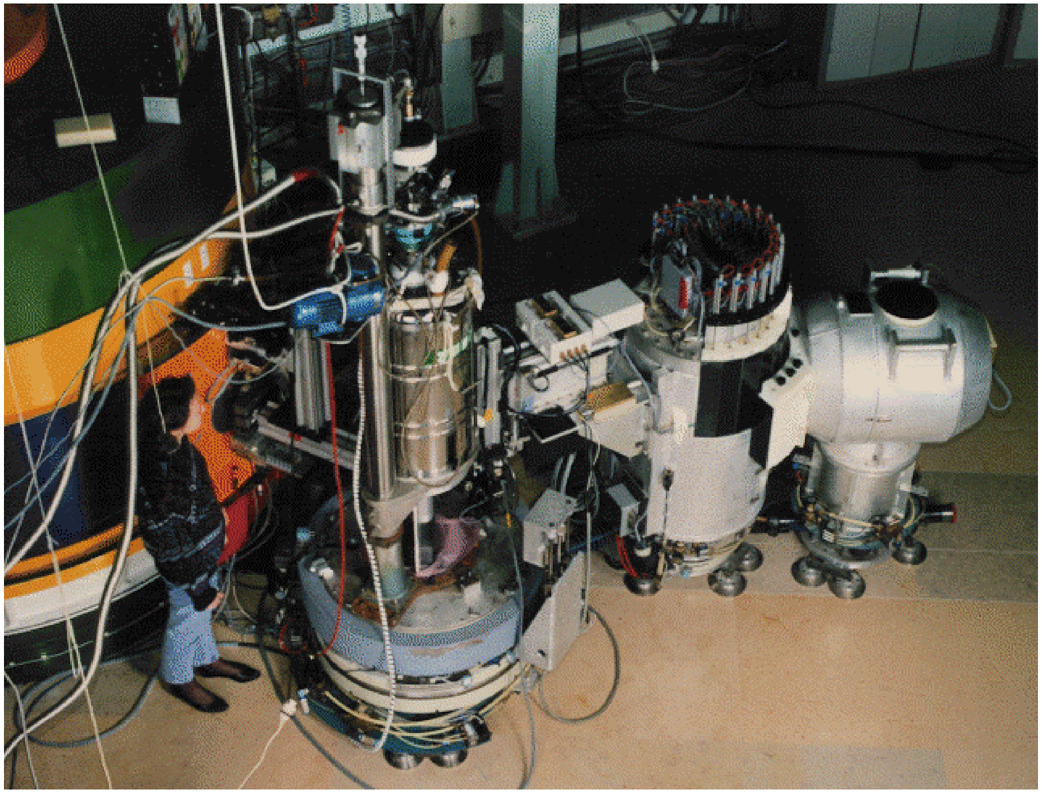




$$Q_{\perp} = \sqrt{\frac{2m(E_i - \omega)}{\hbar^2}}\sin(\phi)$$

$$Q_{//} = \sqrt{\frac{2m}{\hbar^2}}[\sqrt{E_i} - \sqrt{(E_i - \omega)}\cos(\phi)]$$

Triple-Axis Spectrometer



IN20 (ILL)

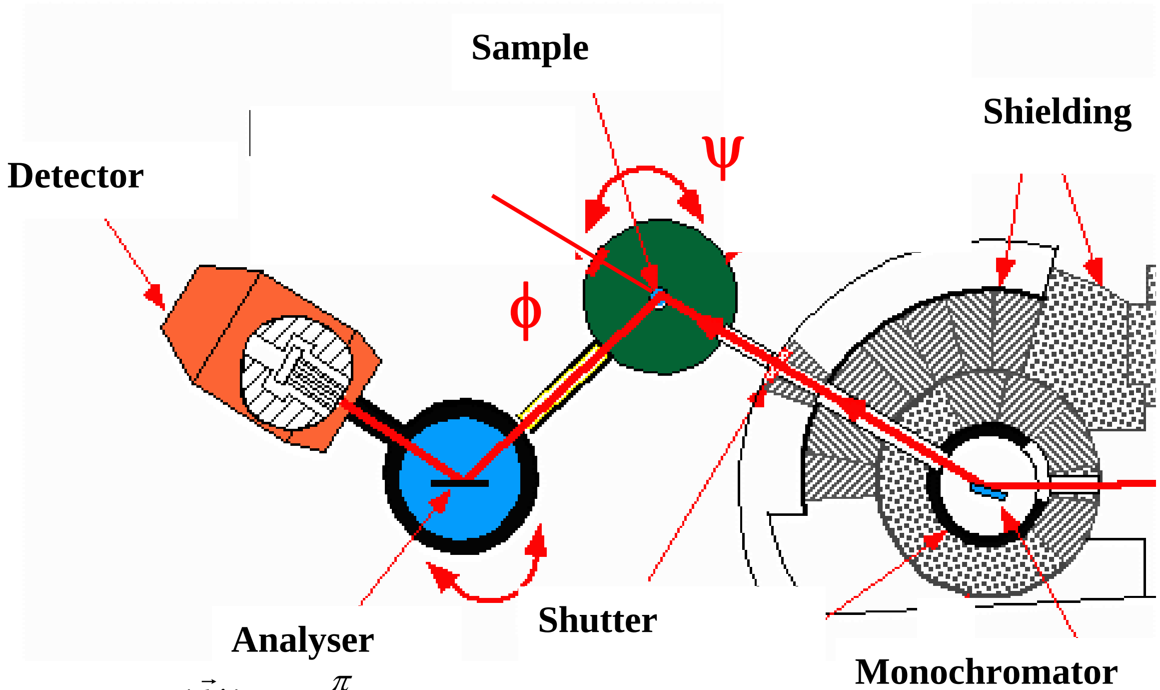
Used at reactors to measure excitations for 40 years

Every research reactor has one or a suite of TAS optimised for different energy ranges

Design principles essentially unchanged

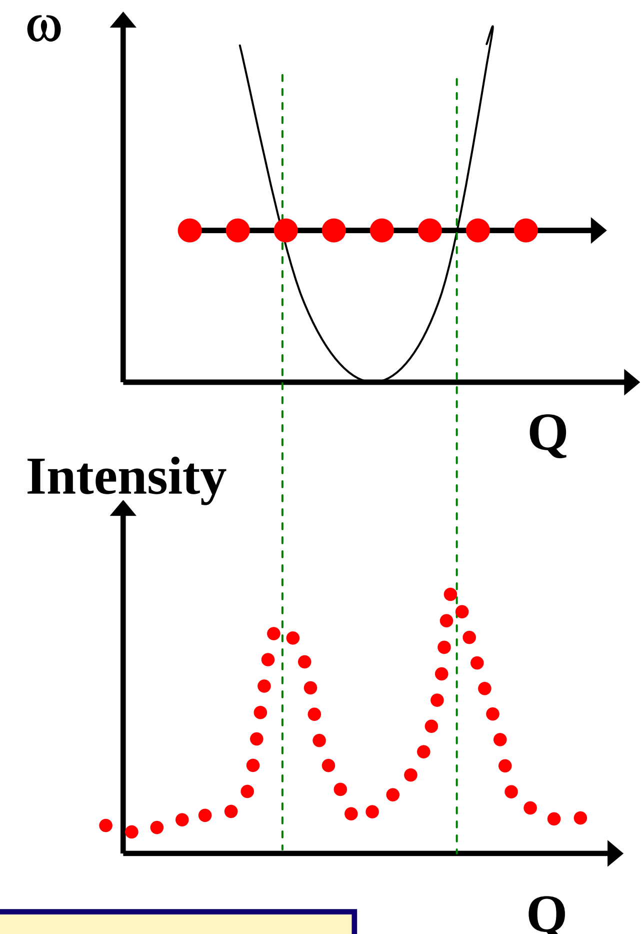
Constantly evolves as technology improves

Triple-Axis Spectrometer (schematic)



$$|\vec{k}'| = \frac{\pi}{d_A \sin(\theta_A)}$$

$$|\vec{k}| = \frac{\pi}{d_M \sin(\theta_M)}$$



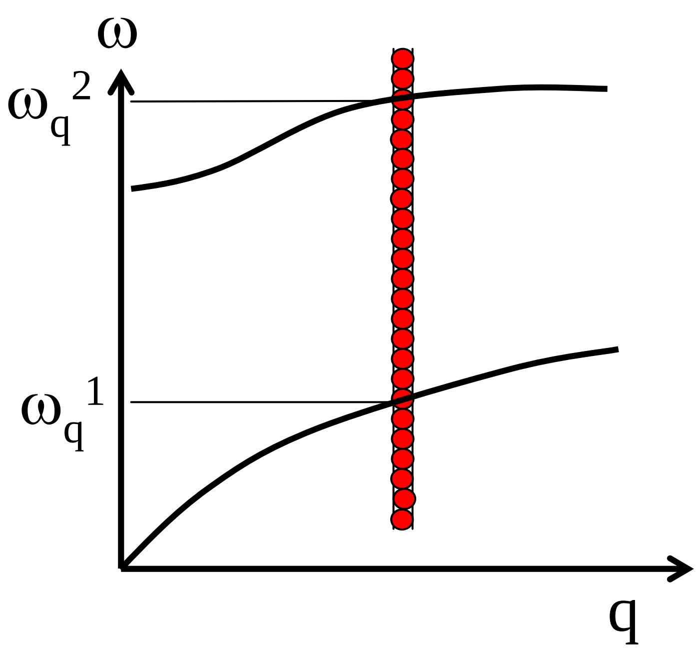
Monochromator $\Rightarrow$ **E**
Analyzer $\Rightarrow$ **E′**
Scattering angle, ϕ $\Rightarrow$ **|Q|**
Crystal orientation, ψ $\Rightarrow$ **Q**

- One point per setting -

Serial Operation

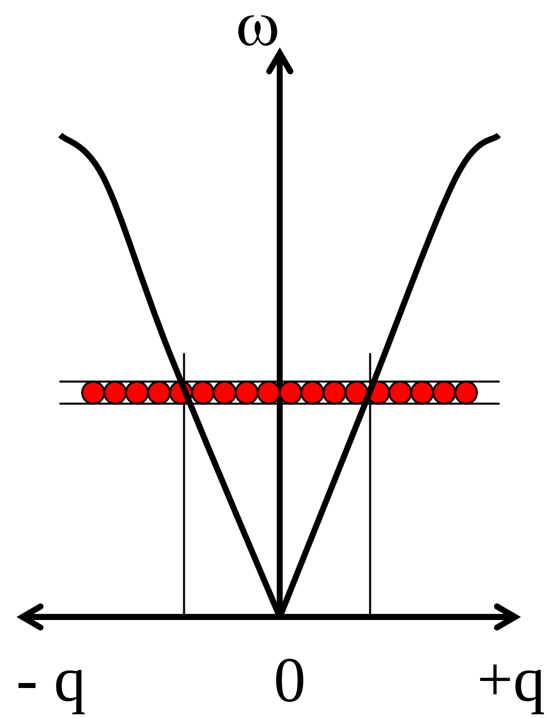
Viewgraph courtesy of R Osborn

Constant Q and Constant E Scans



Constant Q scan

Used to measure slowly varying dispersion relations.



Constant E scan

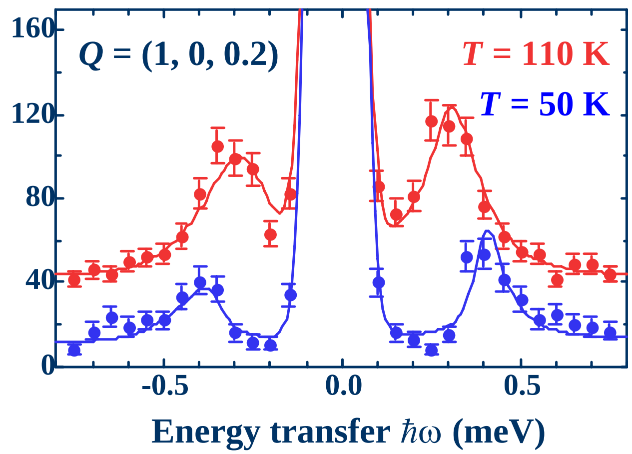
Used to measure steep dispersion relations.

Spin Wave Cross Section (Heisenberg Ferromagnet)

$$\frac{d^2\sigma}{d\Omega\, d\omega} = (\gamma r_0)^2 \frac{(2\pi)^3}{V}\frac{k'}{k}\left\{\frac{1}{2} gF(\vec{Q})\right\}^2 \exp\left\{-2W(\vec{Q})\right\}\left(1+\hat{Q}_z^2\right)\frac{1}{2}S$$

$$\sum_{\tau,q}\langle n_q+1\rangle\delta(\vec{Q}-\vec{q}-\vec{\tau})\delta(\omega-\omega_q)+\langle n_q\rangle\delta(\vec{Q}+\vec{q}-\vec{\tau})\delta(\omega+\omega_q)$$

Spin Wave Creation **Spin Wave Annihilation**



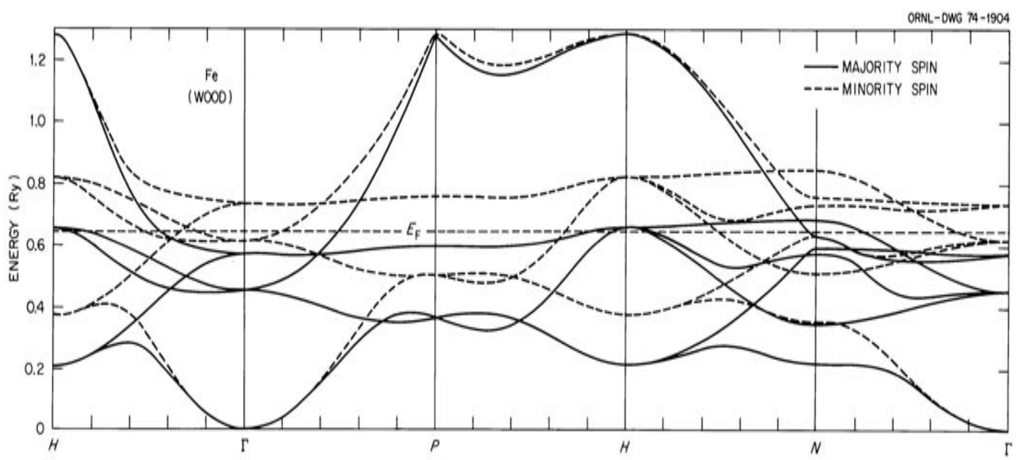
spin waves in $La_{1.2}Sr_{1.8}Mn_2O_7$

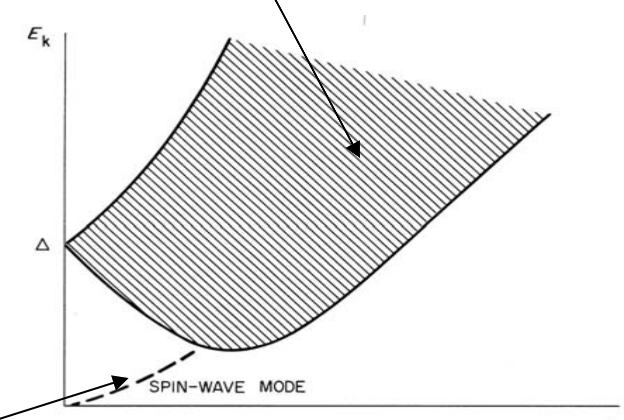
This is what we see in a constant-E scan.
Note the effect of detailed balance
$S(-\omega) = \exp(-\omega/kBT)\ S(\omega)$

Viewgraph courtesy of R. Osborn

Ferromagnets

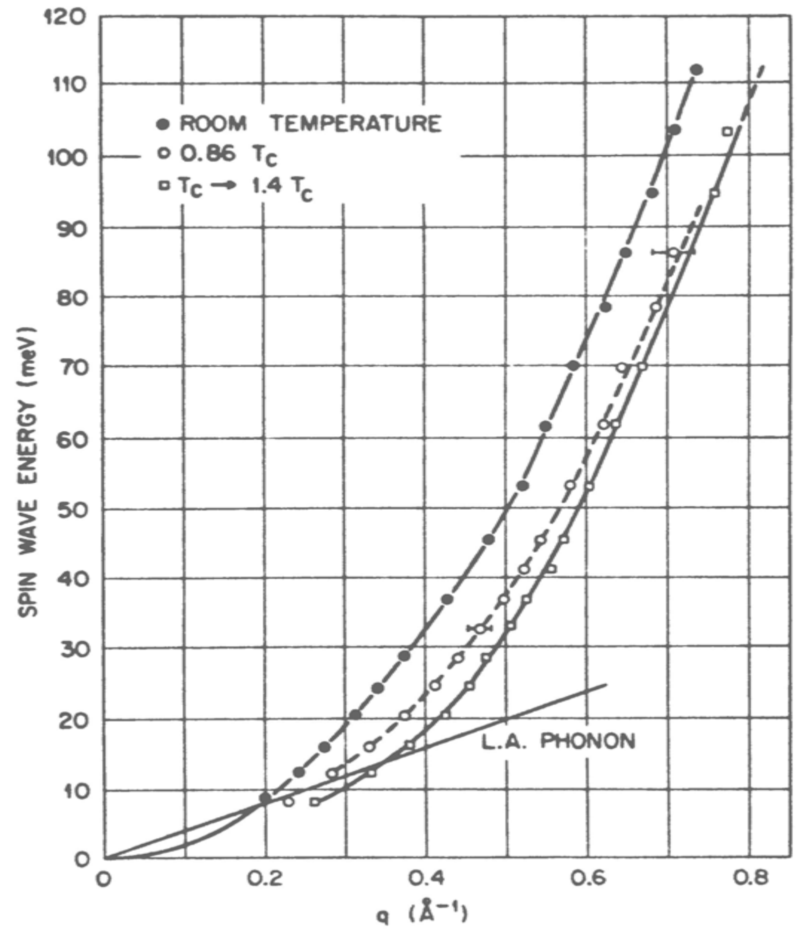
- Localized magnets (eg EuO and EuS) show spin waves (magnons) of the type pictured previously – i.e. precessing spins
- What about iron – an itinerant-electron magnet?
- Use the band structure to get the moment right -> band splitting parameter Δ = 2.2 meV
- Leads to single-particle excitations called Stoner modes





- Correlated electrons lead to spin waves
  - Izuyama, Kim, Kubo, J. Phys. Soc. Japan 18, 1025 (1963);
  - J. Hubbard, Proc. Roc. Soc. A276, 238 (1963)

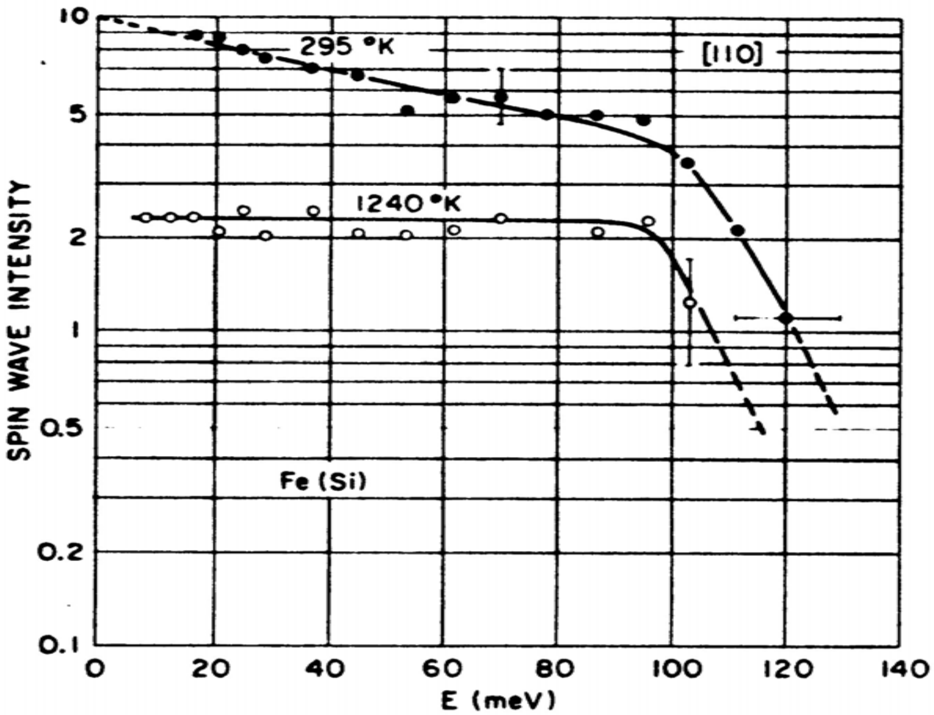
Fe Magnon Dispersion vs. T



Lynn, Phys. Rev. B**11**, 2624 (1975).

Viewgraph courtesy of J. Lynn

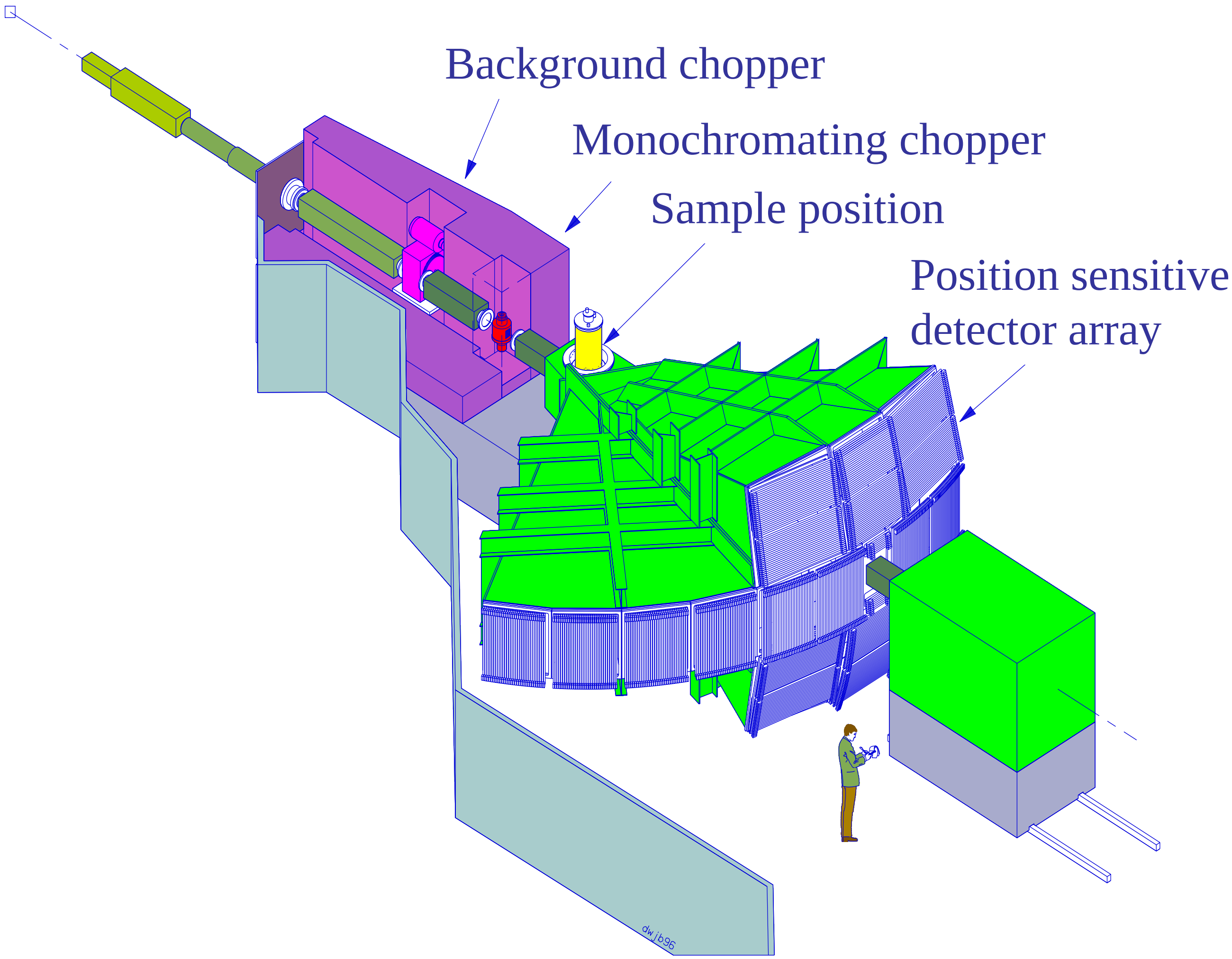
Iron: Magnon Intensities fall at the Boundary of the Stoner Continuum



Mook and Nicklow, PRB7, 336 (1973)
Lynn, Phys. Rev. B**11**, 2624 (1975)

Viewgraph courtesy of J. Lynn

Direct Geometry Chopper Spectrometer
e.g. MAPS at ISIS



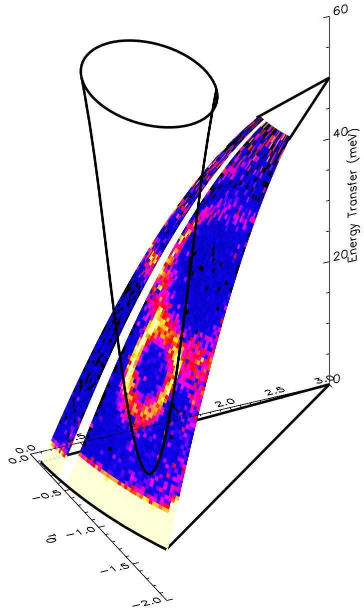
<u>Specification:</u>

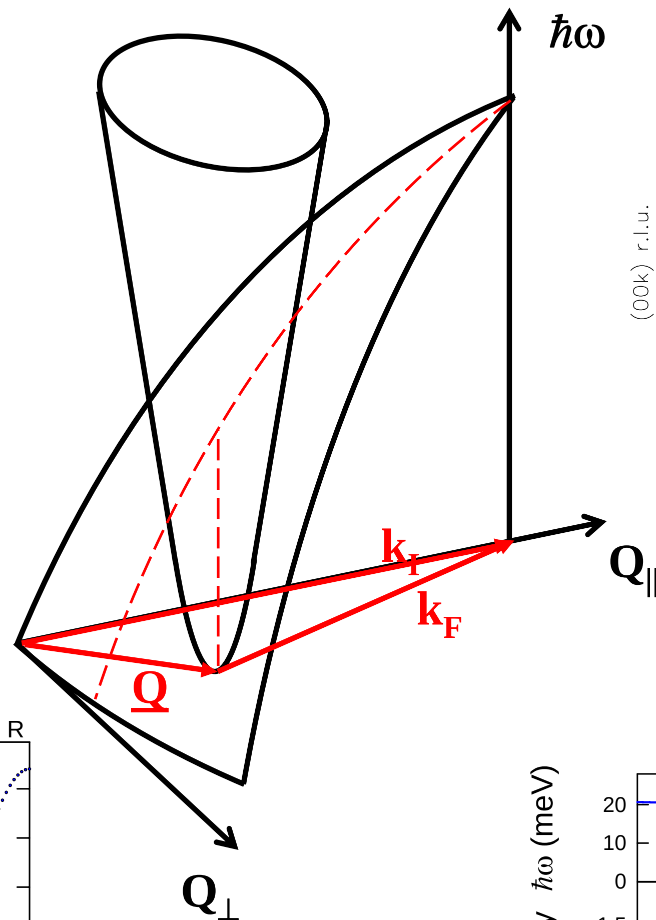
- 20meV< E_I< 2000meV
- $l_{mod\text{-}chop}$ = 10m
- $l_{sam\text{-}det}$ = 6m
- low angle bank: 3°-20°
  high angle bank: → 60°
- $\Delta\hbar\omega/E_I$ = 1- 5% (FWHH)
  ~ 50% more flux
  or ~ 25% better resln.
- 40,000 detector elements
  2500 time channels

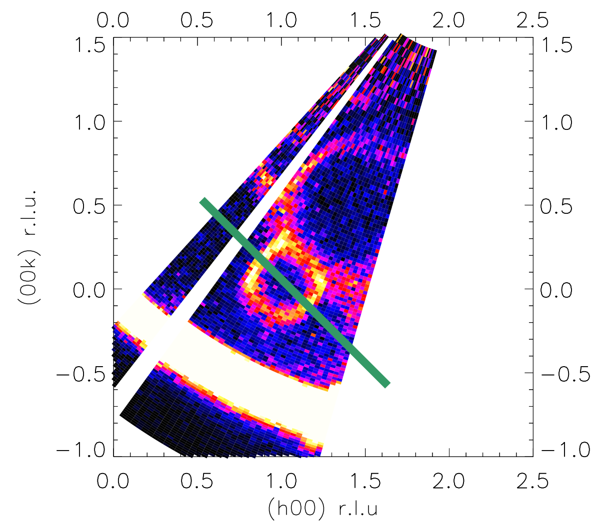
→10^8 pixels ≡ 0.4GB datasets

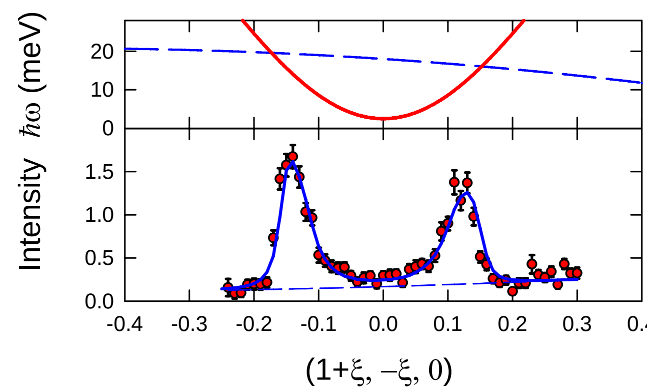
SNS has a similar instruments: ARCS and SEQUOIA

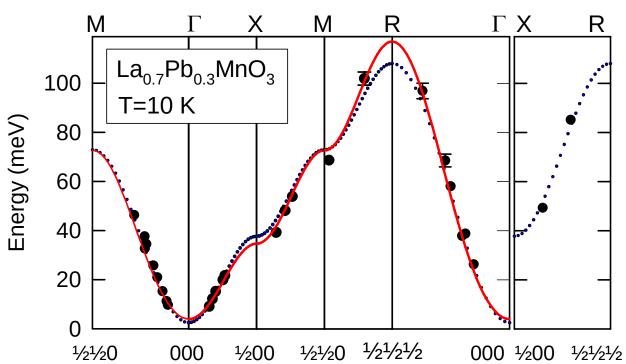
$La_{0.7}Pb_{0.3}MnO_3$ - CMR Ferromagnet





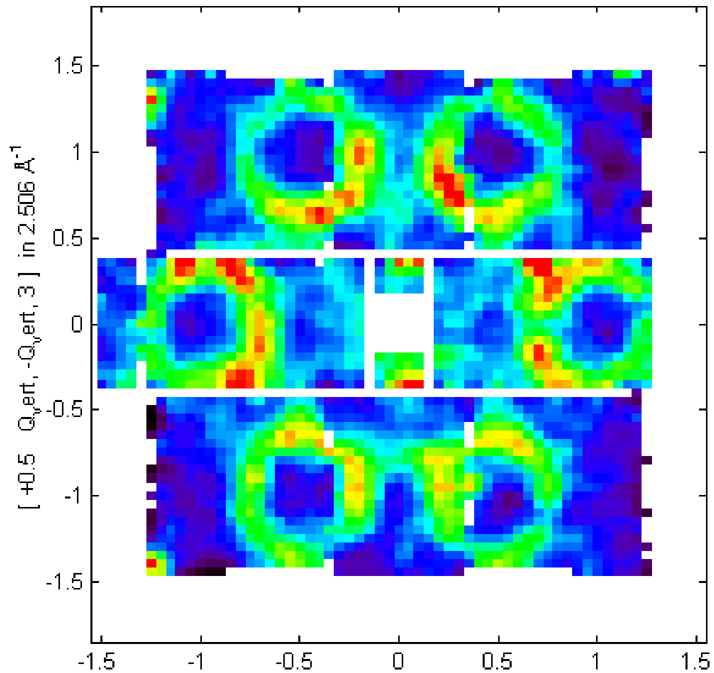


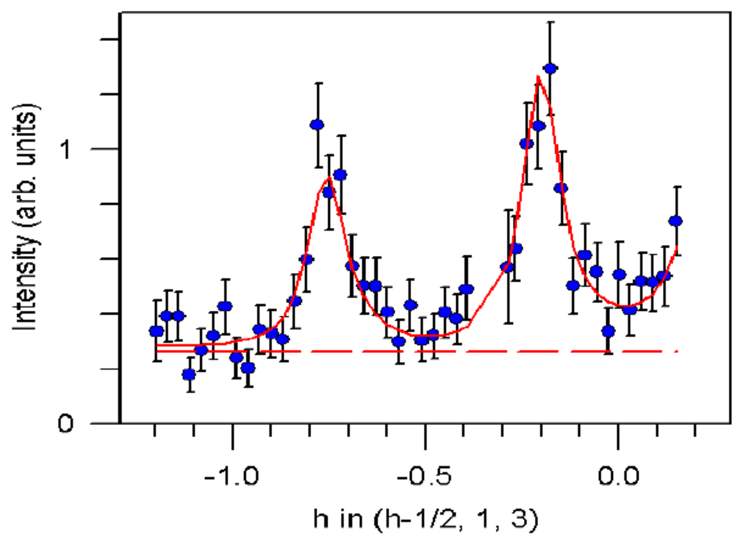


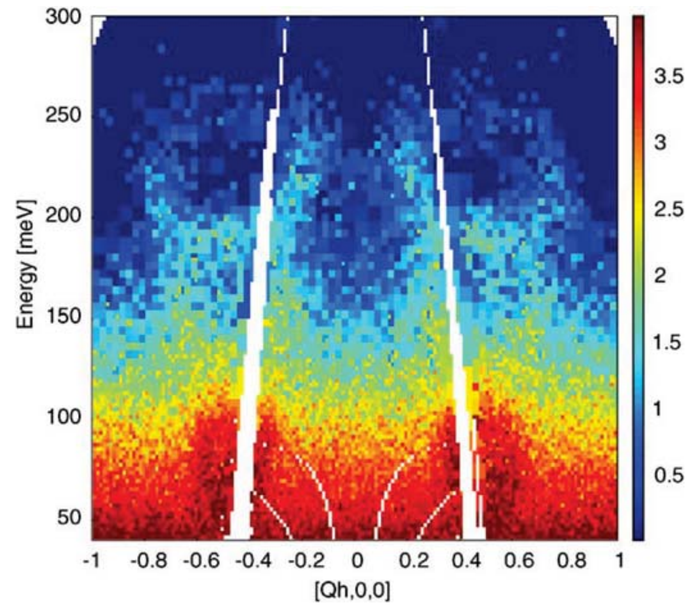


Perring et al., Phys Rev. Lett. **77,** 711 (1996)

Spin Waves in Cobalt Measured on MAPS at ISIS







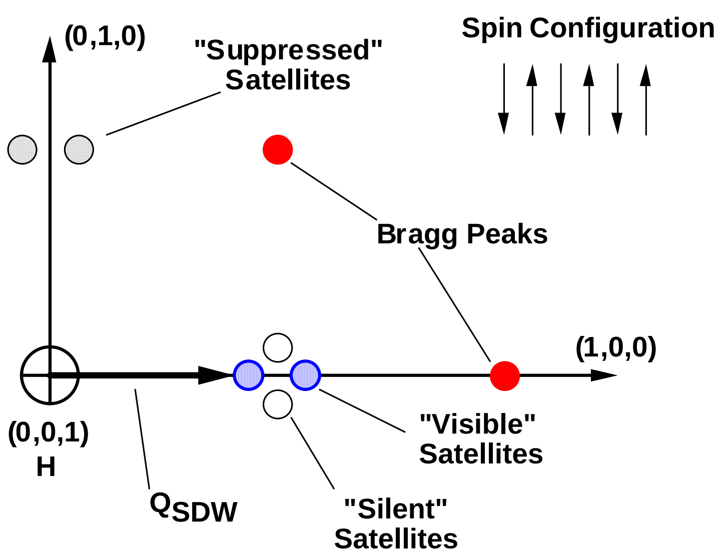
$$H = -J \Sigma \, \boldsymbol{S}_i . \boldsymbol{S}_j$$

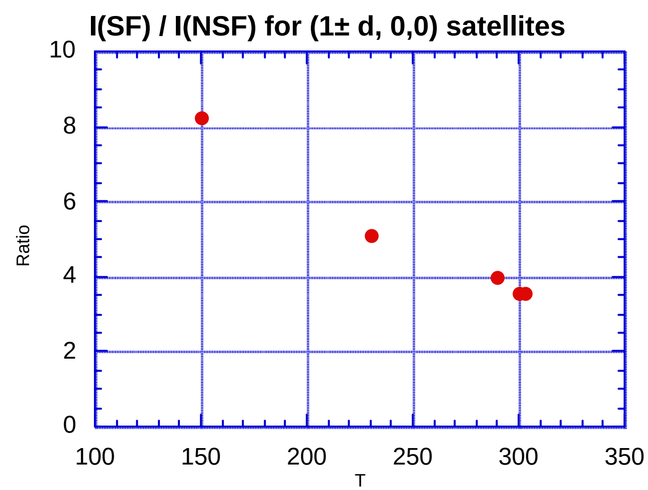
$12SJ$ = 199±7 meV

γ = 69 ±12 meV

Lest you think Magnetic Excitations in Metals are Simple….

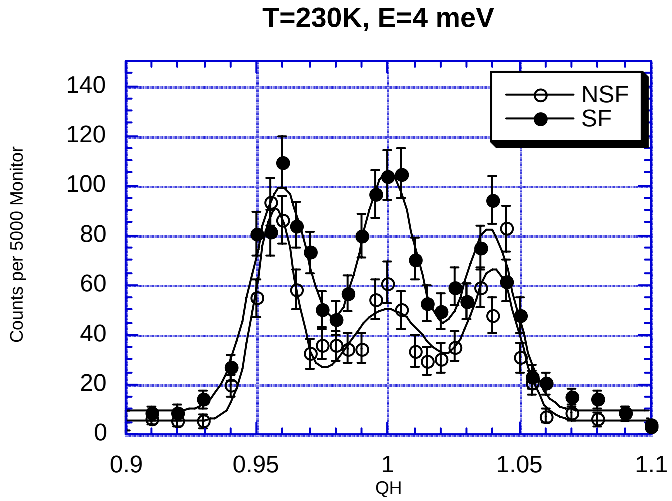
- Chromium has an SDW ground state at low T
- It can be placed in a single-Q state by cooling in a large field
  - Leads to “visible” and “silent” SDW peaks
- When a strong field is applied perpendicular to QSDW, a single spin direction is favored
  - Satellites corresponding to the other domain are “supressed”

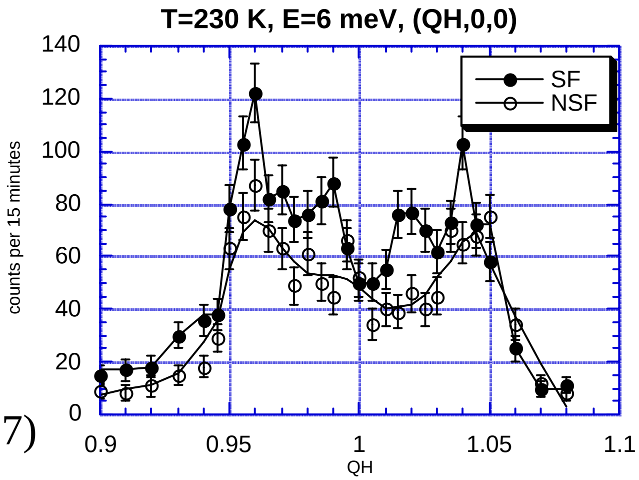




Magnetic Excitations in Chromium are Strange

- We would expect steep spin waves starting at the satellites
- A scan along (h,0,0) at E = 4 meV shows:
  - 3-peaked structure in nsf and sf channels
  - The side peaks are close to – but not at the SDW – wavevectors
  - If the side peaks were steep spin waves, they should be in the nsf channel *only*
  - We have no obvious explanation for the h = 1 peaks in either sf or nsf channels
  - At E = 6 meV, further "modes" appear
  - No existing theory can even qualitatively explain our results.

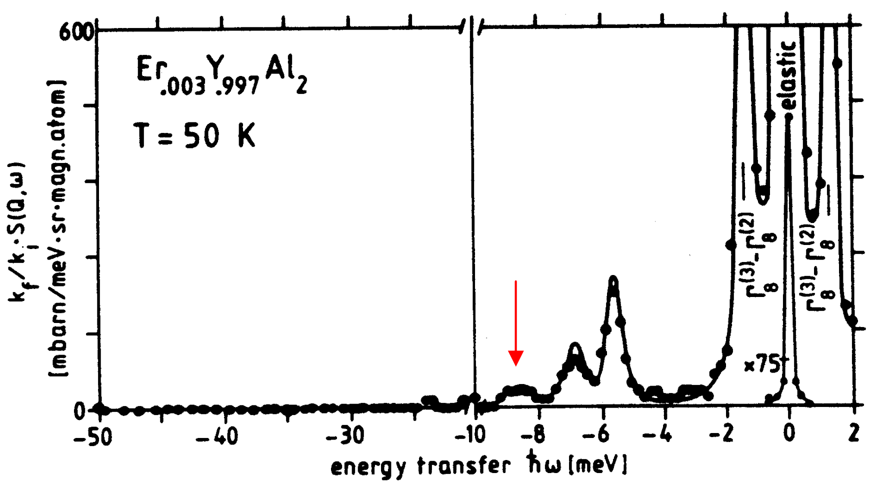


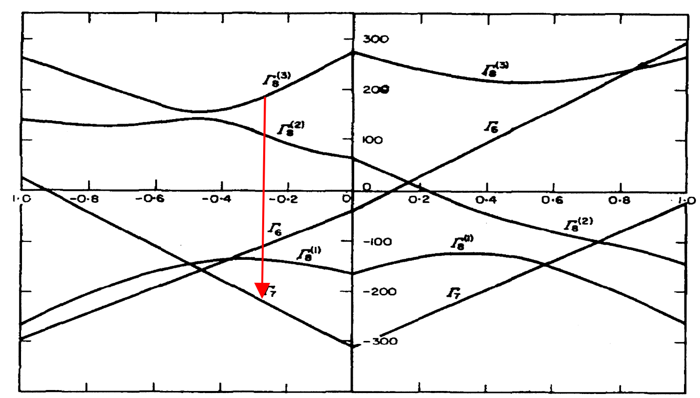


Pynn et al, in *Dynamical Properties of Unconventional Magnetic Systems* (ed Skjeltorp & Sherrington) NATO ASI E349, 267 (1997)

Crystal Electric Field Excitations

- Surrounding atoms generate an electric field (CEF) on atomic electrons -> a manifold of electron states whose energies depend on CEF.
- Excitations between these states provide info about CEF
- Example: $Er_{0.003}Y_{0.997}Al_2$ (cubic Laves-phase structure)





$$E_{\Gamma_7} - E_{\Gamma_8^{(3)}} = 8.5\,meV$$

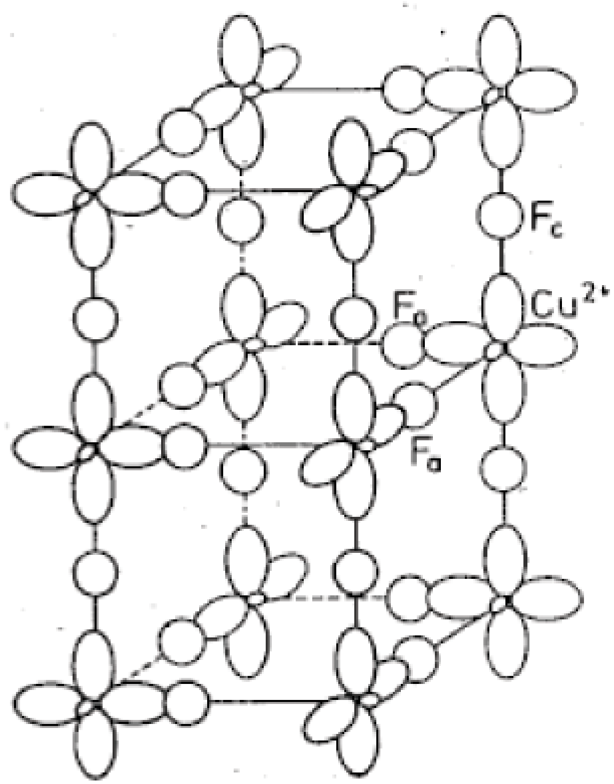
B. Frick and M. Loewenhaupt [Z. Phys. B, **63**, 213 (1986)]

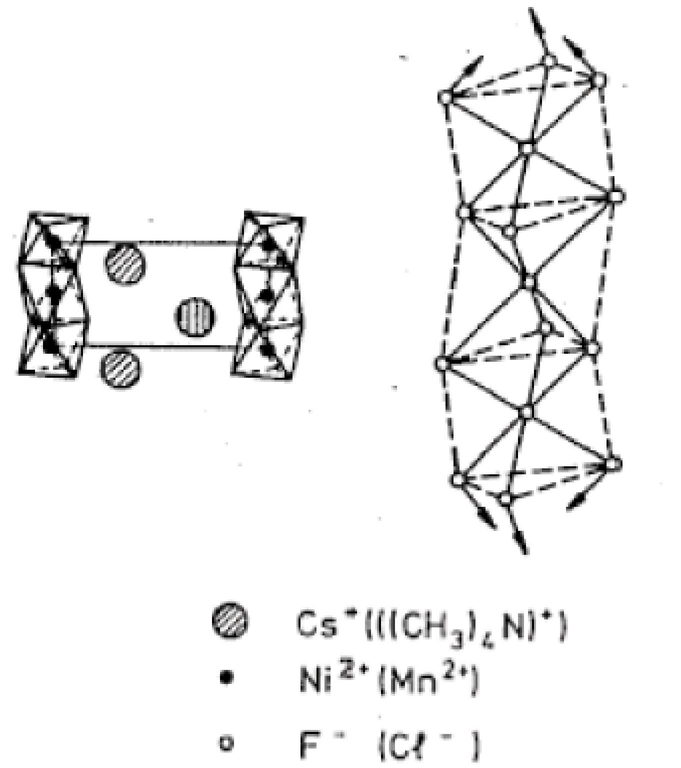
Low Dimensional Magnets

- Materials in which exchange coupling between magnetic ions varies with direction, usually due to spatial separation
- Magnetic anisotropy (usually single site) causes the number of coupled spin components to vary from material to material (n=1, Ising; n=2, X-Y; n=3, Heisenberg)
- Initial motivation was to understand long-range order in low-dimensional systems, phase transitions and the effects on excitations
- But then things got complicated:
  - Excitations can be non-linear/topological – solitons etc
  - The value of the magnetic spin matters – Haldane gap
  - Which spin components are really fluctuating?
  - Singlet ground states and quantum fluctuations
  - etc

Low-dimensional magnets

M. Steiner, J. Villain and C.G. Windsor, Advances in Physics 25(1976) 87





S=1/2 Heisenberg antiferromagnet
$KCuCl_3$

S=1 easy plane ferromagnet
$CsNiF_3$

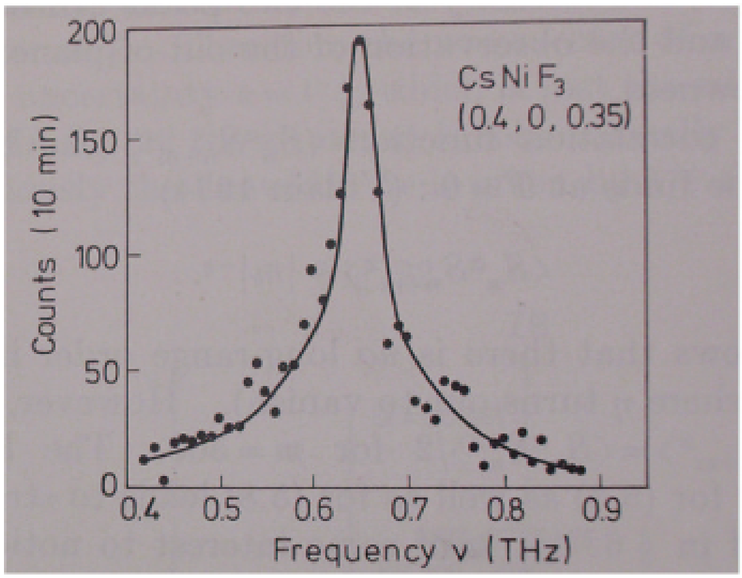
S=5/2 Heisenberg antiferromagnet
TMMC

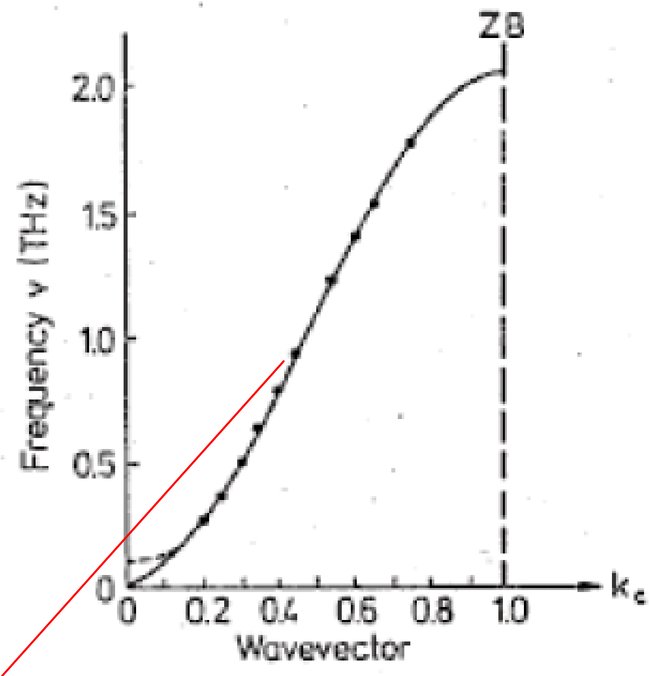
Viewgraph courtesy of K Kakurai

$CsNiF_3$ 1-D ferromagnet 1-D for T>4.2K

hexagonal crystal structure $P6\ 3/mmc$

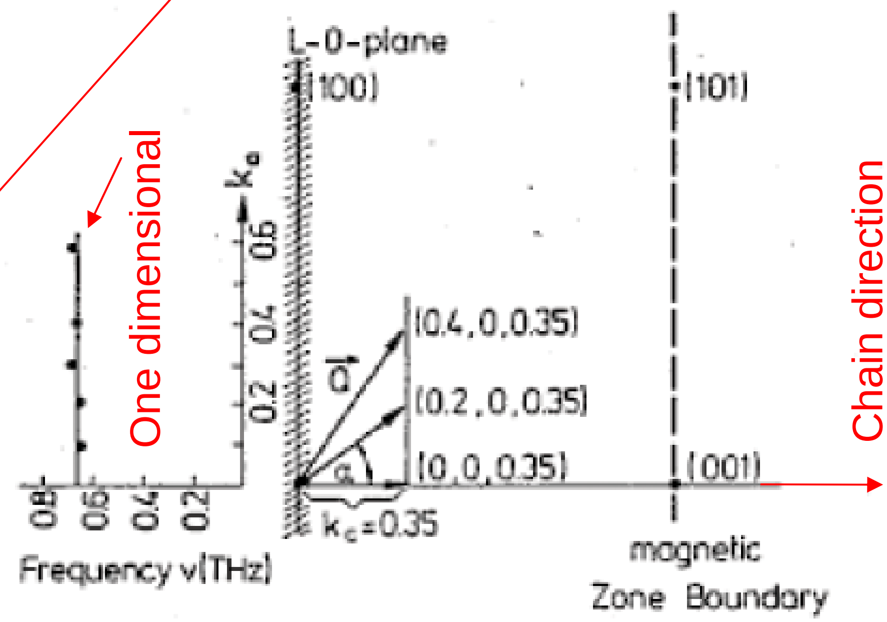
$a = b = 6.21$ Å and $c = 5.2$ Å





$$\hbar\omega = 2S[(2J - 2J\cos\pi k_c)(2J - 2J\cos k_c + A)]^{1/2}$$

$$|J|/k = 11.8\text{ K};\ |A|/k = 9.5\text{ K}$$

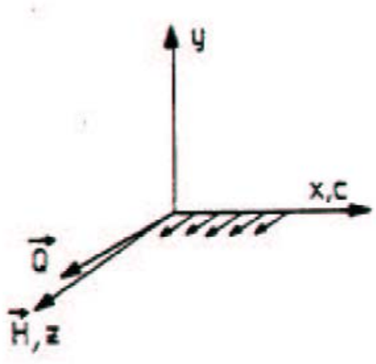


M. Steiner, B. Dorner: Spin Wave Measurements in the One Dimensional Ferromagnet $CsNiF_3$.
Solid State Communications 12, S. 537-540 (1973)

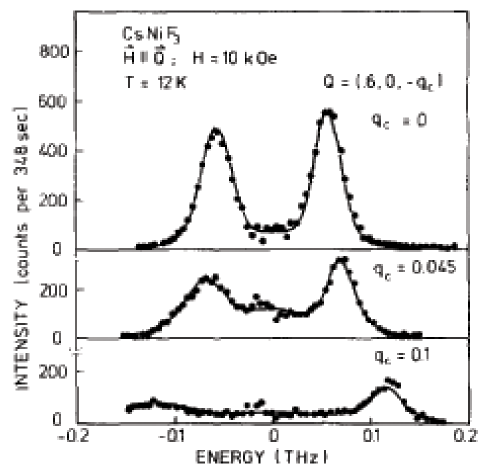
Viewgraph from K Kakurai

Solitons in $CsNiF_3$

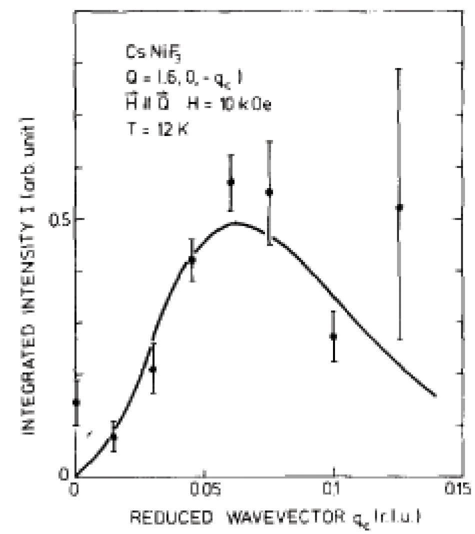
- In a strong magnetic field, Ni spins aligned perp to chain axis
- Possibility of solitons – a propagating "twist" of the spins about the chain axis
  - magnetic fluctuations predominantly perpendicular to the chain direction.
- Solitons give rise to peak at $\Delta E = 0$ in neutron scattering which has to be separated from:
  - incoherent scattering – estimate at low T where material is 3d ordered
  - double magnon – magnetization fluctuations parallel to chain so measure with Q close to parallel to the chain direction



Steiner et al, Solid St Commun, **41**, 329 (1982)



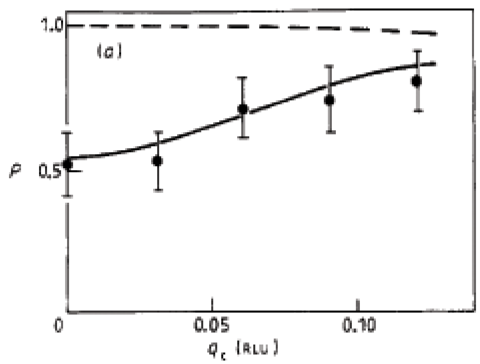
Soliton peak after subtracting Incoh



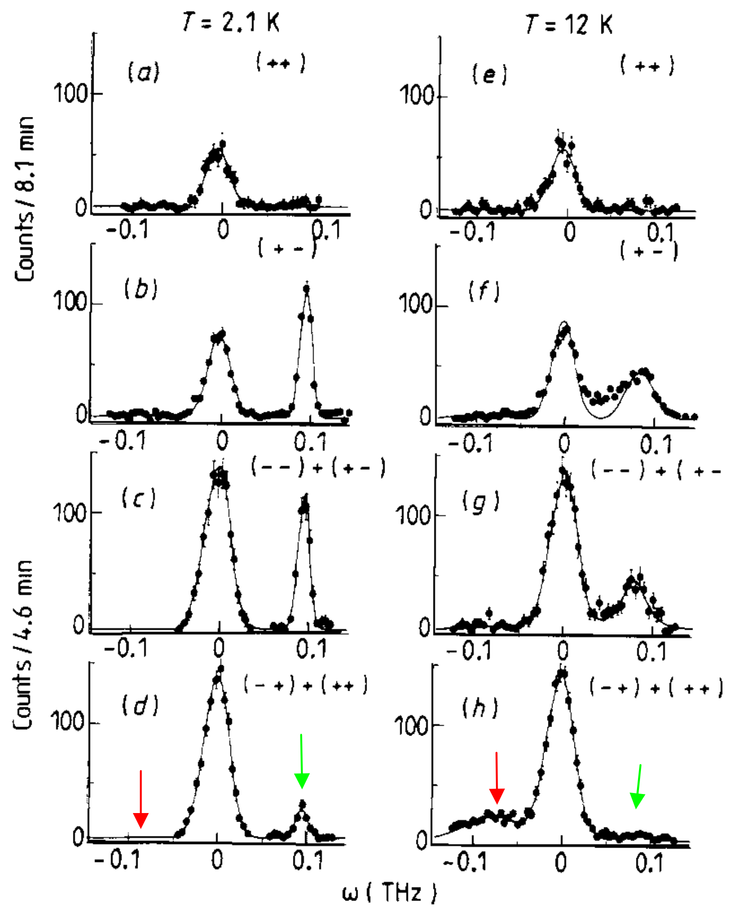
Exp & theoretical soliton intensity

$CsNiF_3$ with Neutron Polarization Analysis

- In a saturated FM with $\vec{H} // \vec{Q}$, spin wave scattering flips the neutron spin & the operators involved are the same as creation and annihilation operators for spin waves in an isotropic Heisenberg system, so SW creation should appear in the +- cross section.
  – Data are consistent with this at T = 12K but not at T = 2.1K (red & green arrows)
  – Hamiltonian for $CsNiF_3$ is NOT isotropic at low T (Kakurai et al, J. Phys C17, L123, 1984)

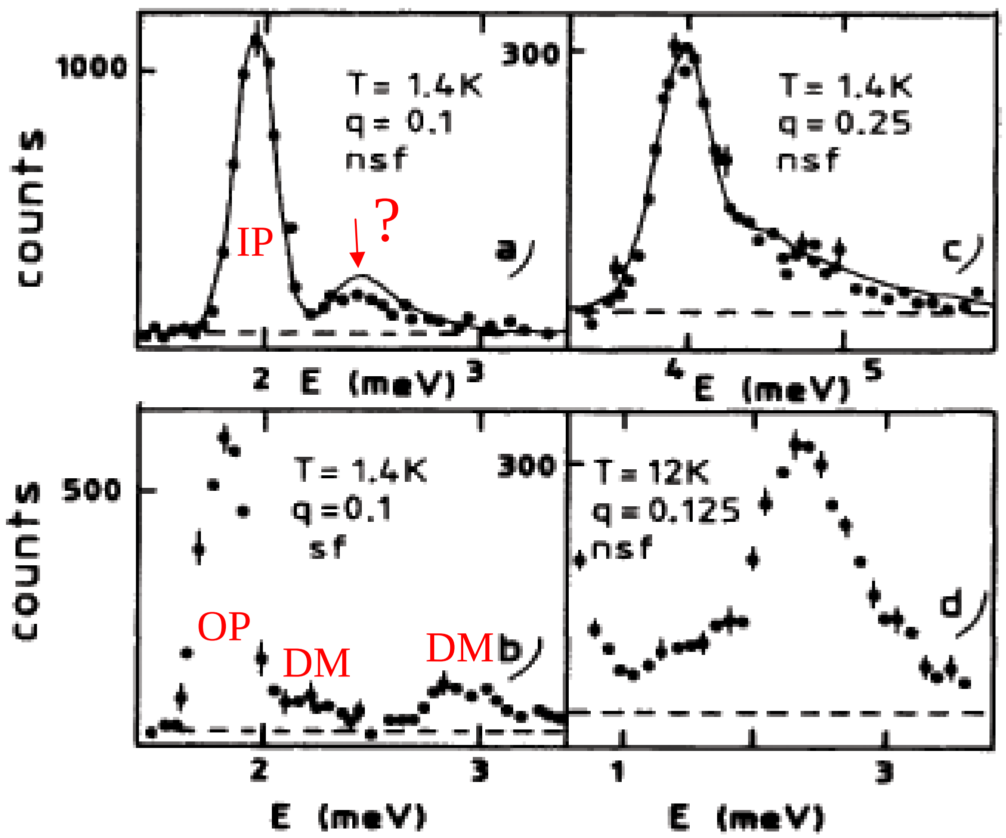


Neutron polarization for spin waves for an isotropic Heisenberg system (dashed) compared with observed and calculated for $CsNiF_3$



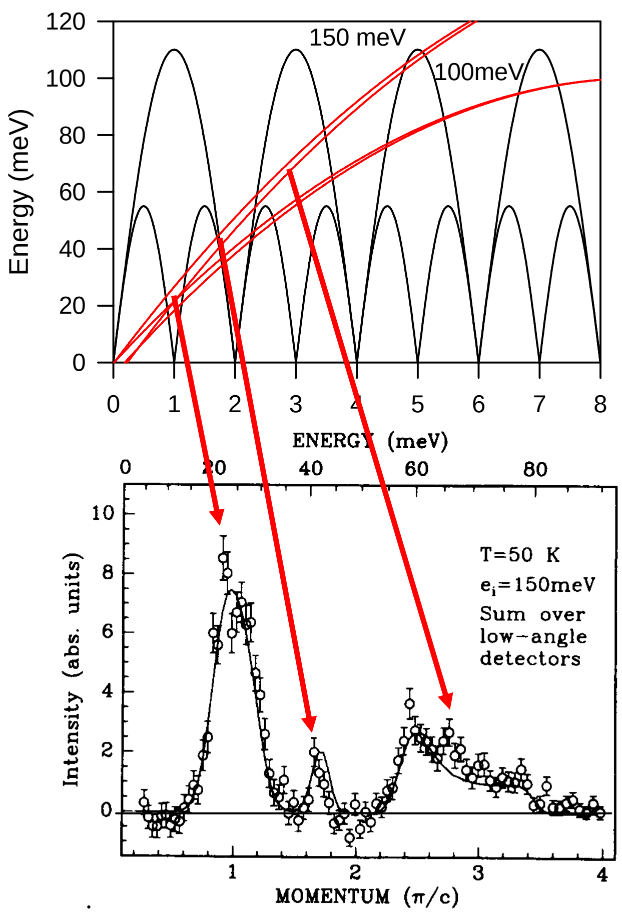
TMMC, a 1-d AFM, didn't even have the "right" number of modes!

- In spin flop phase, expect in-plane (IP; x) and out-of-plane (OP; z) spin wave fluctuations, plus double magnon modes (y) – 4 modes expected but 5 are immediately obvious.
- It turns out that the mag field causes a non-linear coupling of IP and OP excitations, giving new excitations with energies equal to the sum & difference of the single SW modes
- The new excitation is one of these – message non-linear effects are ubiquitous



Boucher et al; PRL 64, 1557 (1990)

$KCuF_3$ Excitations: s=1/2 Heisenberg AFM Chain



- **Singlet ground state with excitations originally interpreted as spin waves**
- **Broad peak can only be explained by continuum**

First clear evidence of continuum scattering in S=1/2 chain

- **Intensity scale:**

$A = 1.78 \pm 0.01 \quad \pm 0.5$

c.f. numerical work:

$A = 1.43$

- **Coupling constant:**

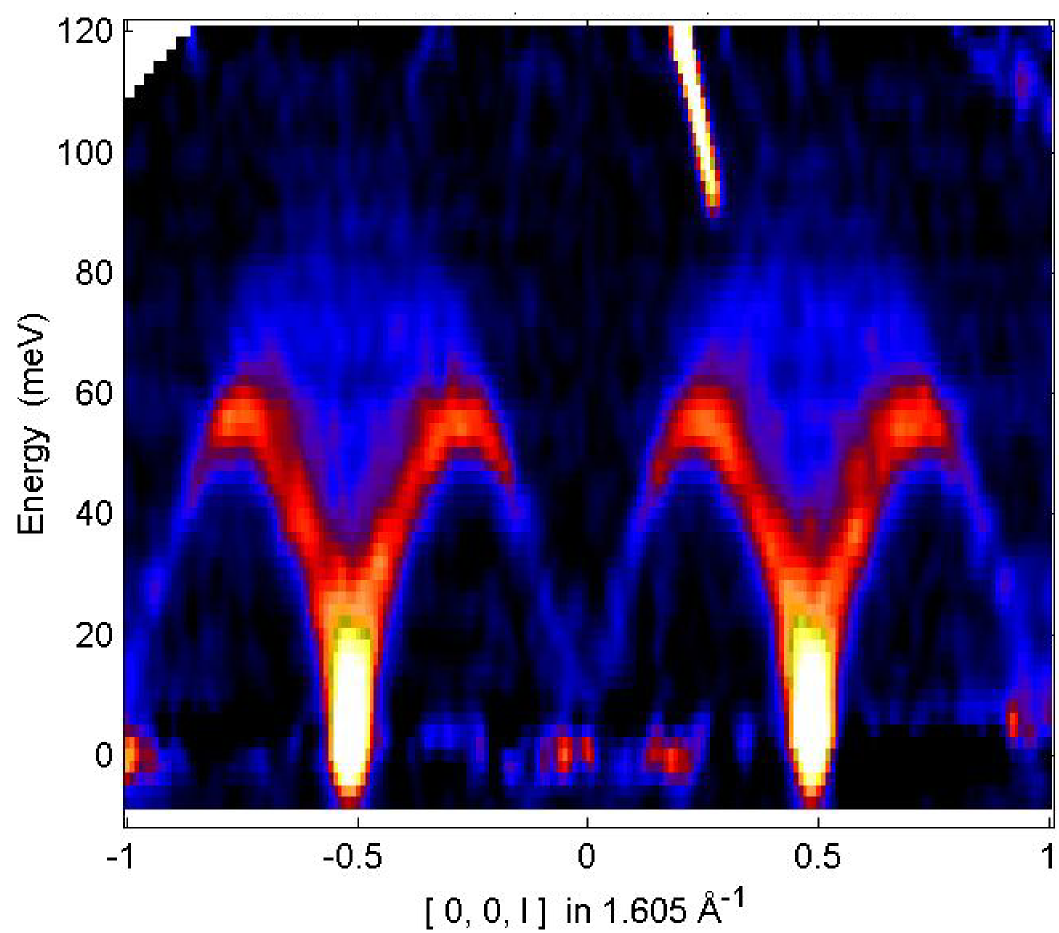
$J = 34.1 \pm 0.6$ meV

D.A.Tennant et al, Phys. Rev. Lett. **70** 4003 (1993)

Viewgraph from Ray Osborn

$KCuF_3$ Excitations (ISIS)

Direct observation of the continuum



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Viewgraph from Ray Osborn