Neutron spectroscopy 2: Chopper Spectrometers

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Science and Technology Facilities Council

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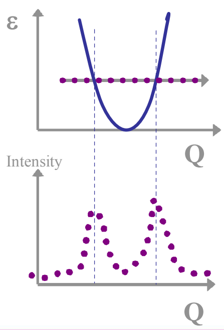
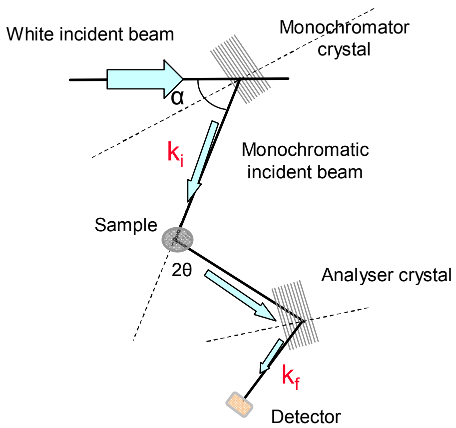
Outline

- Overview of chopper spectrometers
  - Introduction and comparison with triple axis
  - Detailed description of operation and characteristics
  - Breadth of scientific areas
- Examples of magnetism in single crystals
  - 1D magnetic chains
  - 2D magnetic planes
  - 3D ferromagnet

Triple Axis Spectrometer

Triple Axis Spectrometer

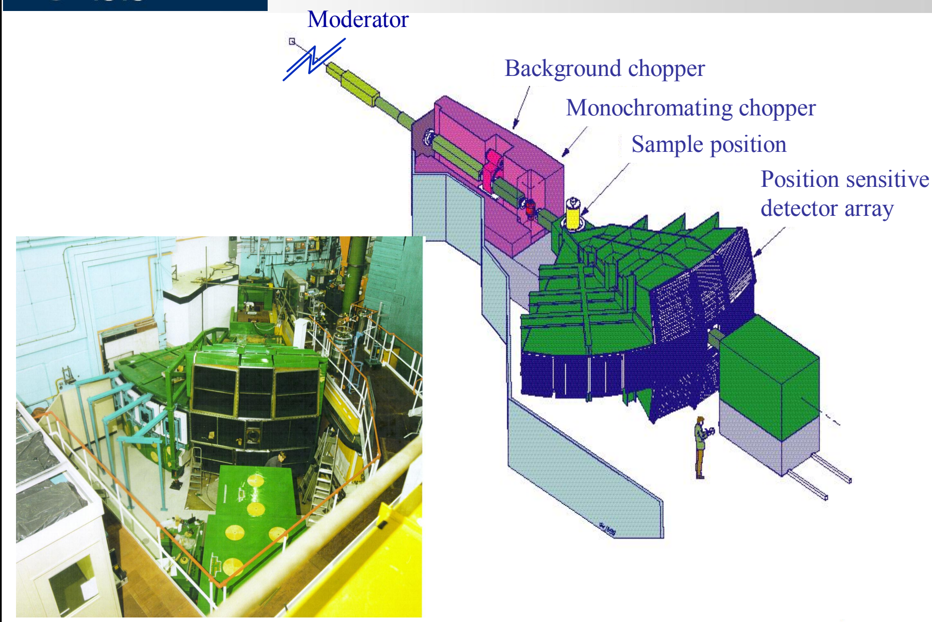




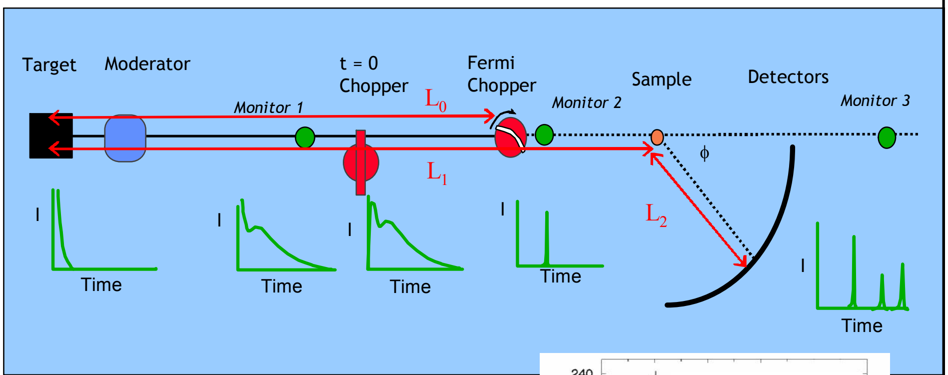
$$\mathbf{Q} = \mathbf{k_i} - \mathbf{k_f}$$

$$\varepsilon = \left(\hbar^2/2m_n\right)\left(k_i^2 - k_f^2\right)$$

Chopper spectrometer

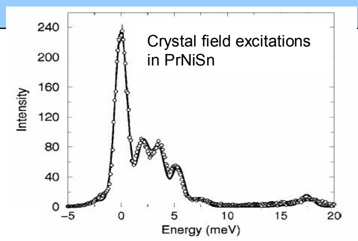


Chopper spectrometers



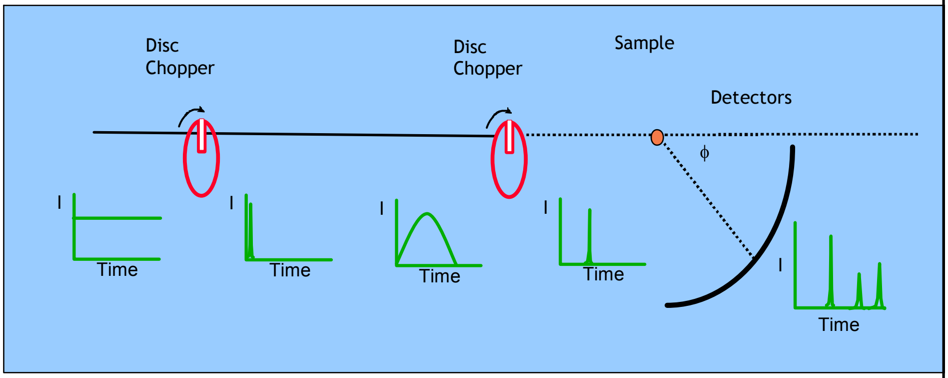
$$v_i = L_0 / T_{chopper}$$
$$T_{sample} = L_1 / v_i$$
$$v_f = L_2 / (T_{\det} - T_{sample})$$

$$\varepsilon = E_i - E_f$$
$$\mathbf{Q} = \mathbf{k_i} - \mathbf{k_f}$$

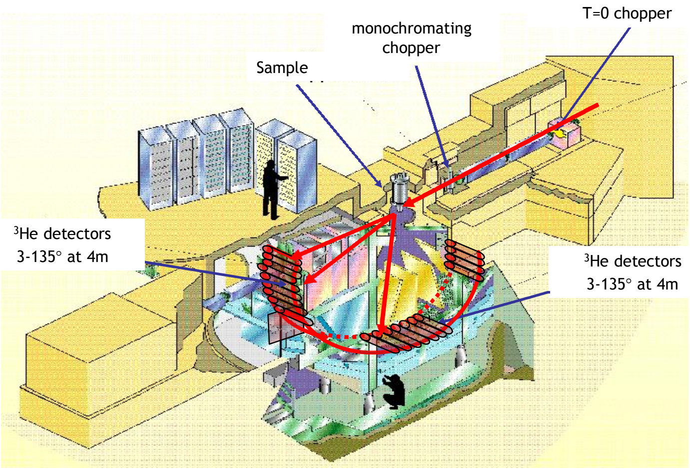


K. McEwen et al., Phys Rev B **73** 014402 (2006)

Chopper spectrometers – reactor



MARI spectrometer (ISIS)

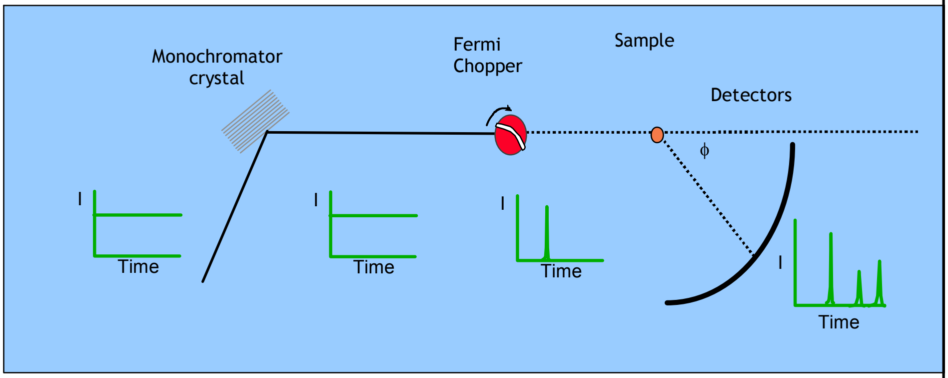


Specification:

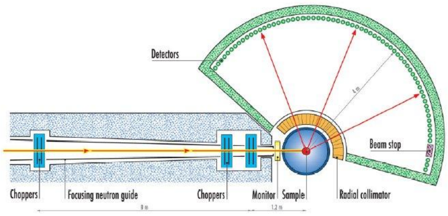
- $10meV < E_i < 1500meV$
- $l_{mod\text{-}chop} = 10m$
- $l_{chop\text{-}sam} = 10m$
- $l_{sam\text{-}det} = 4m$
- Detectors: 3°-135°
  922 detectors
- Fermi Chopper:
  50-600Hz
- Resolution:
  $\Delta\varepsilon/E_i$ = 2 - 5% (FWHH)
  (depends on Fermi chopper frequency)

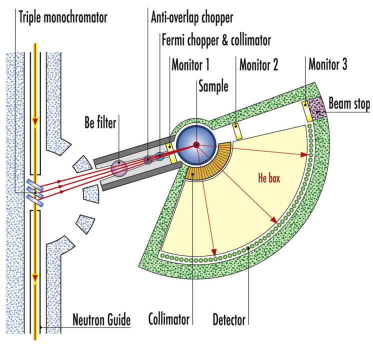
E_i= 10meV $\Delta\varepsilon$=200μeV
500meV $\Delta\varepsilon$= 10-25meV

Chopper spectrometers – reactor



IN5, IN6 (ILL)





IN5:

- 0.2 meV< E_i< 25 meV
- Detectors: 14.5°-132.5°
  (552 detectors)
- Resolution:
  $\Delta\varepsilon/E_i$ = ≈ 3% (FWHH)

e.g. E_i=3.27meV $\Delta\varepsilon$ = 100μeV

IN5:

- E_i = 2.35, 3.15, 3.87, 4.87 meV
- Detectors: 10°-115°
  (337 detectors)
- Resolution:
  $\Delta\varepsilon/Ei$ = ≈ 2-3% (FWHH)

e.g. Ei=2.35meV $\Delta\varepsilon$ = 50μeV

Accessible energy and momentum

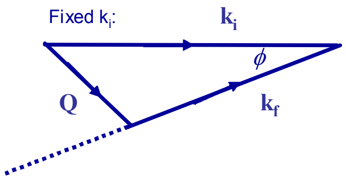
Neutron kinematics:

$$\mathbf{Q} = \mathbf{k_i} - \mathbf{k_f}$$

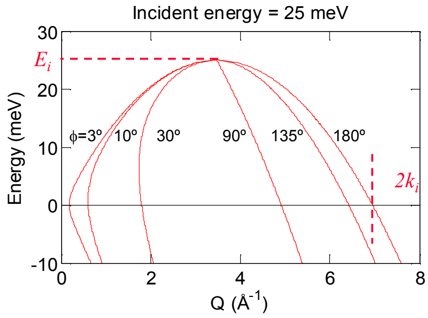
$$\varepsilon = \left(\hbar^2/2m_n\right)\left(k_i^2 - k_f^2\right)$$

$$\varepsilon_{meV} = 2.072\, k^2_{Ang^{-1}}$$

Scattering triangle:



$$Q^2 = (\mathbf{k_i} - \mathbf{k_f}).(\mathbf{k_i} - \mathbf{k_f})$$
$$= k_i^2 + k_f^2 - 2k_i k_f \cos\phi$$



Accessible energy and momentum

Choose E_i according to the problem at hand:

- What is the energy range of the excitations you expect ?
- What is the momentum dependency ?

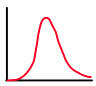
  Magnetic form factor:

  $S(\boldsymbol{Q}) \sim \exp(-\alpha Q^2)$ FWHH ~ 4Å^{-1}

  Phonons, molecular vibrations:

  $S(\boldsymbol{Q}) \sim Q^2\exp(-\alpha Q^2)$

- What energy resolution is required?

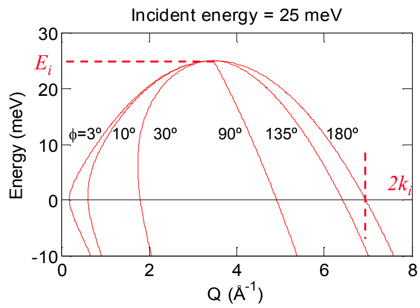
Example:

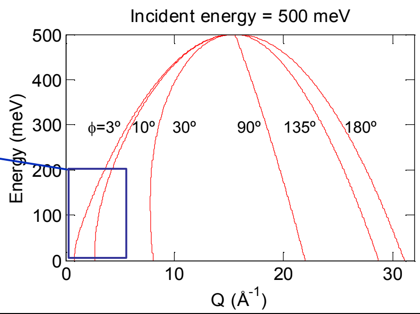
Spin waves with energy 0 - ~200meV: E_i >200meV

Magnetic form factor: |Q| < ~4Å^{-1} ⇒ E_i ~500meV

Want to discriminate between theories with effects with magnitude ~10% of bandwidth at the maximum energy: Δε ~ 20meV = 4% Ei ⇒ $\Delta\varepsilon/E_i$ ~ 2%

Change conditions according to what you learn during the experiment





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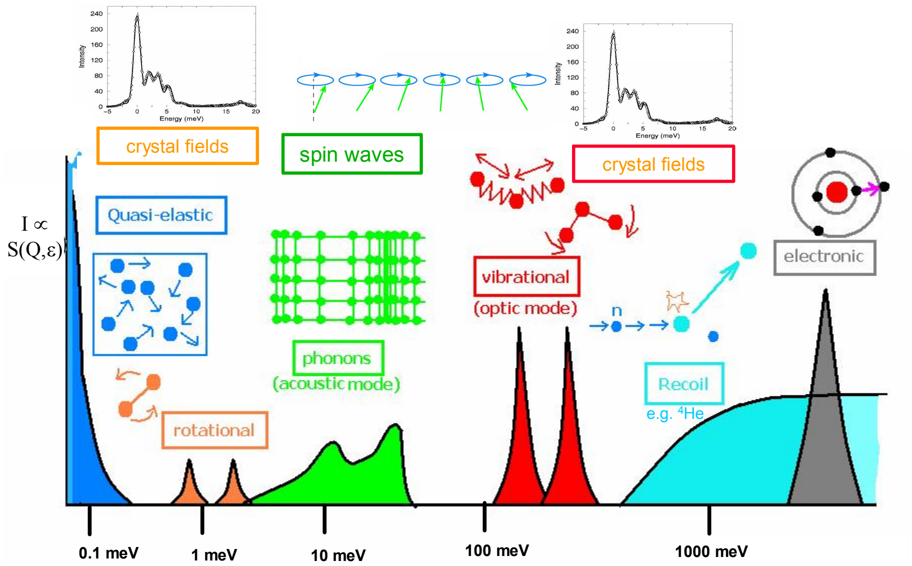
Science on chopper spectrometers



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Comparison of triple-axis and time-of-flight

Triple axis spectrometer

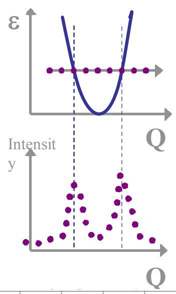
•Very flexible: focus on a single given (**Q**,ε) point
•Build up scans **serially**

Advantages:

•Optical components can be designed to create huge flux at sample position (e.g. curved monochromators and analysers): 10^7-10^9 n/cm^2/s

•Flexible choice of monochromators and collimators to independently tune spectrometer before and after the sample

•Polarisation analysis on some instruments



Time-of-flight instruments

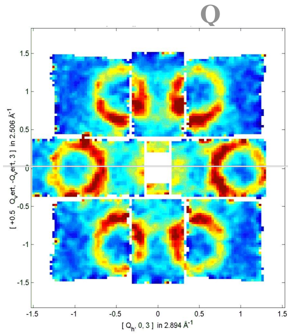
•Comprehensive measurement of S(**Q**,ε)
•Intrinsically **parallel**

Advantages:

•Reduced flux 10^4-10^6 n/cm^2/s st sample but more than recover using time-of-flight and large detector solid angle (up to 3 Steradian)

•(Spallation sources) Measure energy transfers up to 0.5eV

•(Spallation sources) negligible background noise as source is 'off' during measurement.



Excitations in single crystals

One-dimensional example – spin ½ chain

Two dimensional example – spin 1/2/ square plane

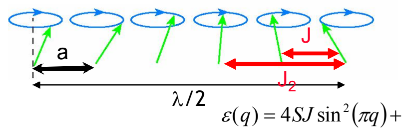
Three dimensional example – ferromagnetic metal

Spin waves

Dispersion relation $\varepsilon(\mathbf{q})$ reveals strength, range and symmetry of magnetic interactions

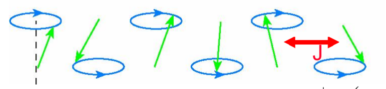
e.g. Heisenberg Hamiltonian $H = -\sum_{<ij>} J_{ij}\mathbf{S}_i.\mathbf{S}_j$

Ferromagnet

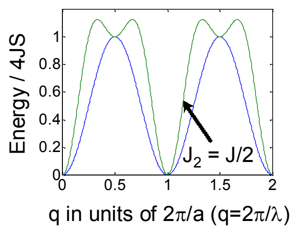


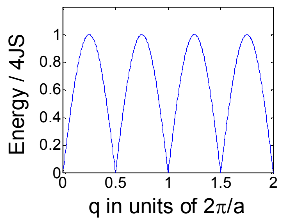
$$\varepsilon(q) = 4SJ\sin^2(\pi q) + 4SJ_2\sin^2(2\pi q)$$

Antiferromagnet

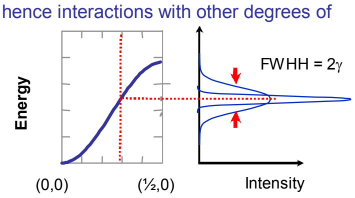


$$\varepsilon(q) = 2SJ\left|\sin(2\pi q)\right|$$

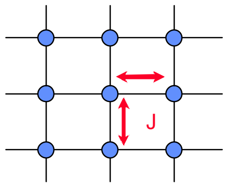


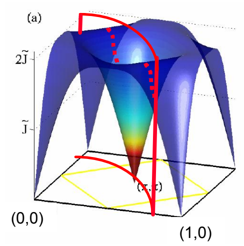


Lifetime $\tau(q)=1/\gamma(q)$ reveals information about damping mechanisms and hence interactions with other degrees of freedom



Why single crystals ?





Single crystal: full vector information retained

-(In principle) full mapping of dispersion relations, intensity and lifetimes

-General map of $S(\mathbf{Q},\varepsilon)$ (we may not have much idea of form of scattering)

Powder: Spherical averaging of cross-section

- Directional detail lost (anisotropy)
- Loss of sensitivity to range of interactions
- Weak signals localised in $(\mathbf{Q},\varepsilon)$ are smeared over a sphere: they may be lost in background

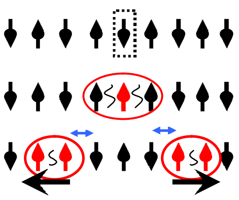
One dimensional systems

Spin ½ HAFM with nearest neighbour interaction only: $H = J\sum_i \mathbf{S}_i.\mathbf{S}_{i+1}$

Ground state wavefunction : Bethe Z. Phys. 71 205 (1931)

Lowest energy excitations: des Cloizeaux and Pearson PR 128 2131 (1962)

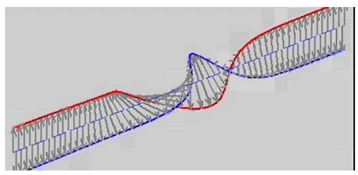
Actually fermions forming continuum of scattering (Fadeev & Takhtajan Phys. Lett. 85A 375 (1981), G. Müller et al PRB 24 1439 (1981)



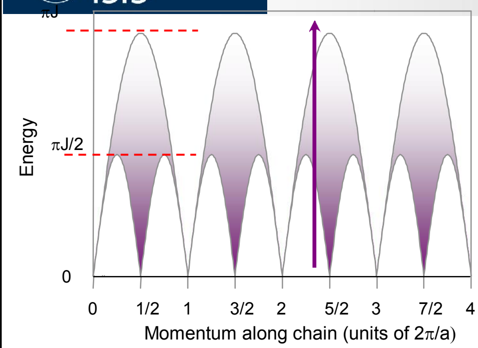
Flip one spin

The bound pairs of spins propagate away

– these are *Spinons*



One dimensional systems



Spinons are created in pairs

The total energy and mometum is given by sum of values for two independently propagating spinons

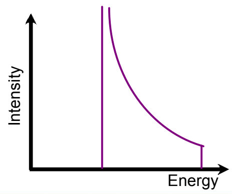
$$q = q_1 + q_2$$

$$\varepsilon(q) = \varepsilon_1(q_1) + \varepsilon_2(q_2)$$

The spinon dispersion therefore leads to a continuum of excitations

$$\varepsilon_U = (\pi J)\left|\sin(\pi q)\right|$$

$$\varepsilon_L = (\pi J/2)\left|\sin(2\pi q)\right|$$

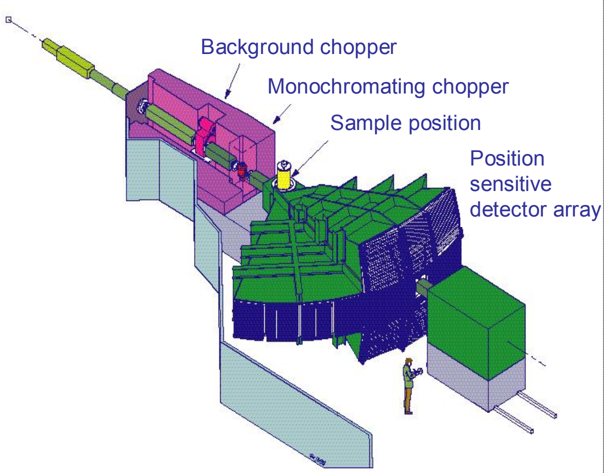


Numerical calculations:

$$S^{xx}(q,\varepsilon) = \frac{A}{2\pi}\frac{\vartheta(\varepsilon - \varepsilon_L(q))\vartheta(\varepsilon_U(q) - \varepsilon)}{\sqrt{\varepsilon^2 - \varepsilon_L(q)^2}}$$

$A \approx 1.2$

MAPS – position sensitive detectors



Specification:

- $20\text{meV} < E_I < 2000\text{meV}$
- $l_{mod\text{-}chop} = 10\text{m}$
- $l_{sam\text{-}det} = 6\text{m}$
- low angle bank: 3°-20°
  high angle bank: → 60°
- $\Delta\varepsilon/E_I$ = 1- 5% (FWHH)

40,000 detector elements
2500 time channels
→10^8 pixels ≡ 0.4GB datasets

Measuring Excitations

Momentum:

$\underline{Q} = \underline{k}_i - \underline{k}_f$

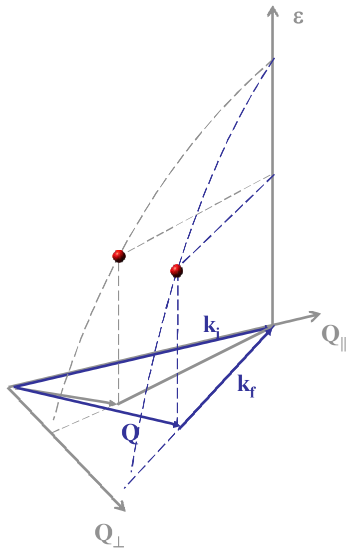
Energy:

$\varepsilon = (\hbar^2/2m)\,(k_i^2 - k_f^2)$

Detector Bank:

Many Trajectories through

$(Q_\perp, Q_\parallel, \varepsilon)$



Measuring Excitations

Momentum:

$\underline{Q} = \underline{k}_i - \underline{k}_f$

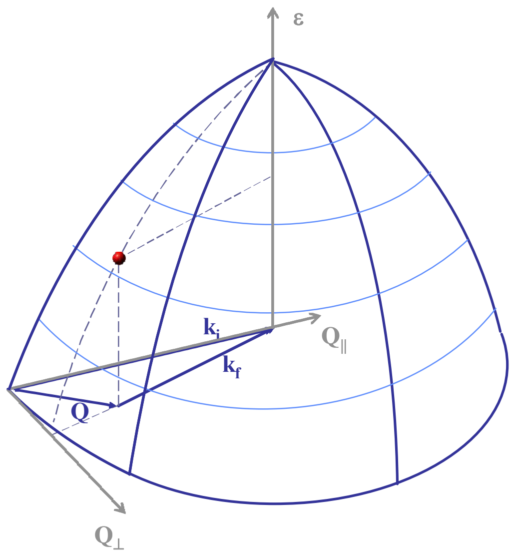
Energy:

$\varepsilon = (\hbar^2/2m)\,(k_i^2 - k_f^2)$

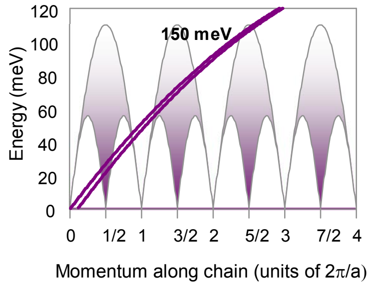
Detector Bank:

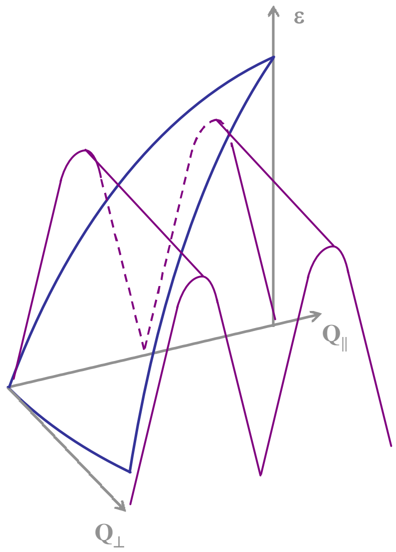
Phase space trajectory becomes a surface in

$(Q_\perp, Q_\parallel, \varepsilon)$

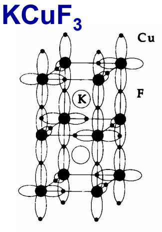


$KCuF_3$ - Spin ½ chain



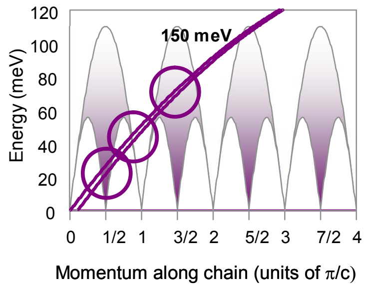


$KCuF_3$



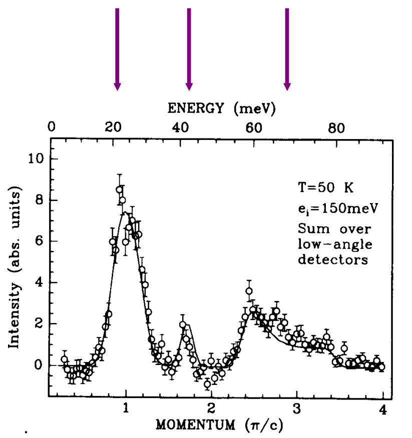
chains

$KCuF_3$ - Spin ½ chain



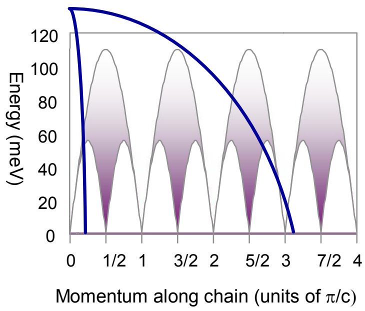
Broad peak between 60-80mev can only be explained by continuum

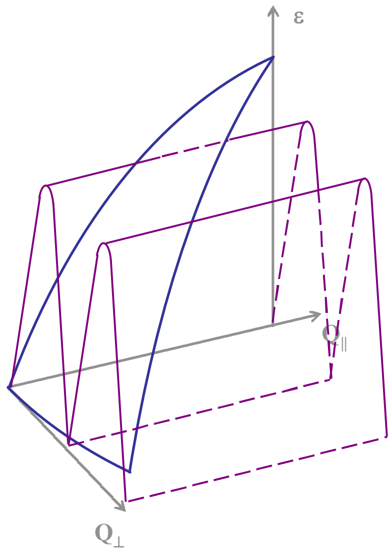
Not two-spin-wave spectrum as that spectrum inconsistent with data



D.A.Tennant, T.G. Perring, R.A. Cowley & S.E. Nagler, Phys. Rev. Lett. **70** 4003 (1993)

One dimensional systems





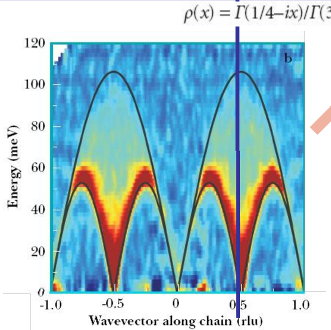
$KCuF_3$ - Spin ½ chain

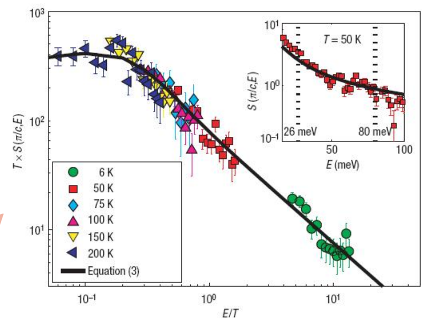
S = ½ chain has a quantum critical point:

Quantum critical case: expect E/T scaling at q=0.5 :

$$S(\tfrac{1}{2}E) = \frac{e^{E/kT}}{e^{E/kT}-1} \; \frac{A}{T} \mathrm{Im}\left[\rho\left(\frac{E}{4\pi T}\right)^2\right]$$

$$\rho(x) = \Gamma(1/4-ix)/\Gamma(3/4-ix)$$





Field theory predicts form of function - experiment agrees

B. Lake, D.A. Tennant, C.D. Frost & S.E. Nagler *Nature Mater.* **4,** 329–334 (2005).

$KCuF_3$ - Spin ½ chain

S = ½ chain has a quantum critical point:

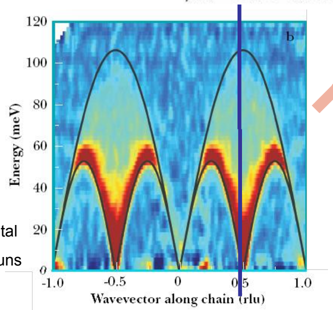
Negligible interchain J_{inter} interactions lead to long-range ordering at very low temperature, $T_C \to 0K$ as $J_{inter} \to 0$

KCuF3 has interchain coupling: T_N=39K: 3D spin waves better description in ordered state at low energy.

Quantum critical case: expect E/T scaling at q=0.5 :

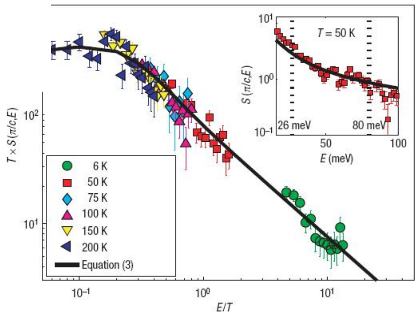
$$\boxed{S(\tfrac{1}{2},E) = \frac{e^{E/kT}}{e^{E/kT}-1}\;\frac{A}{T}\,\mathrm{Im}\left[\rho\left(\frac{E}{4\pi T}\right)^2\right]}$$

$$\rho(x) = \Gamma(1/4-ix)/\Gamma(3/4-ix)$$



7g crystal

~15hr runs



Field theory predicts form of function -

Experiment agrees in regime defined by:

(i) 2 x zone boundary energy of interchain spin waves from ordered state (T_N=39K) (i.e. minimum energy at which each spinon exceeds energy of interchain spin waves

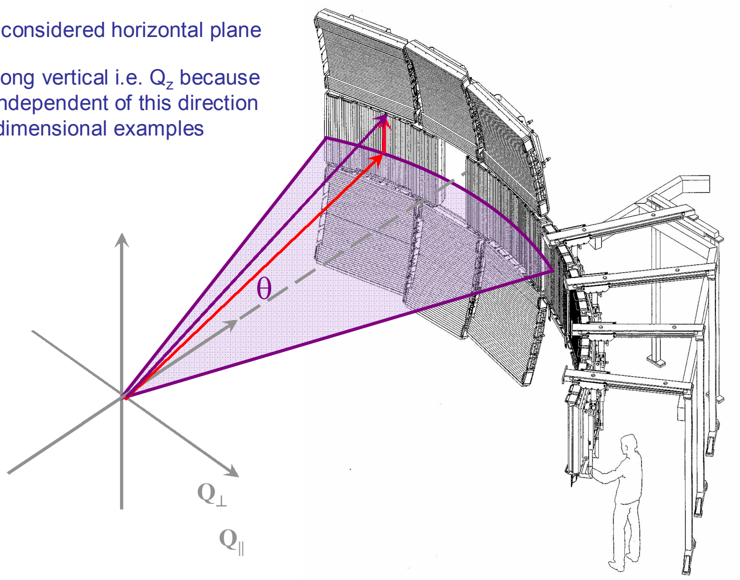
(ii) 2 x energy where linear approximation to lower bound fails

B. Lake, D.A. Tennant, C.D. Frost & S.E. Nagler *Nature Mater.* **4,** 329–334 (2005).

Measuring Excitations

So far only considered horizontal plane

Integrate along vertical i.e. Q_z because scattering independent of this direction in the one-dimensional examples considered



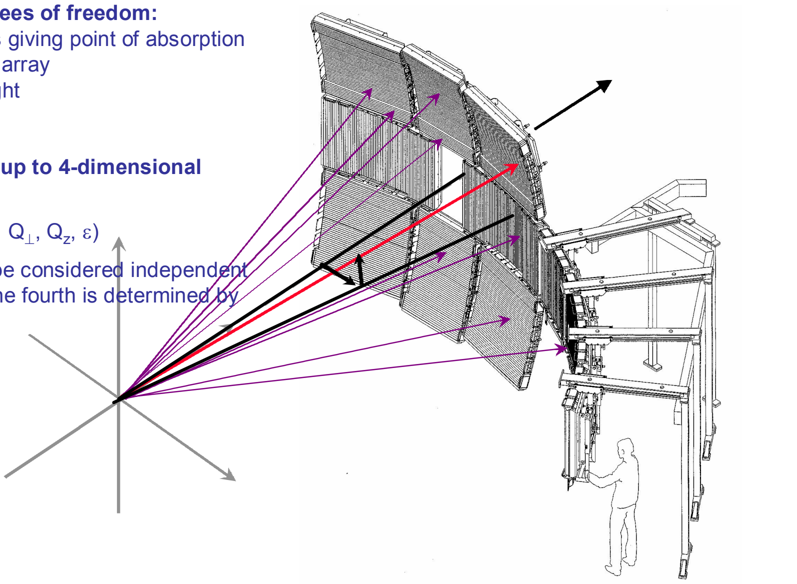
Measuring Excitations in 2D, 3D systems

Three degrees of freedom:
- two angles giving point of absorption on detector array
- time-of-flight

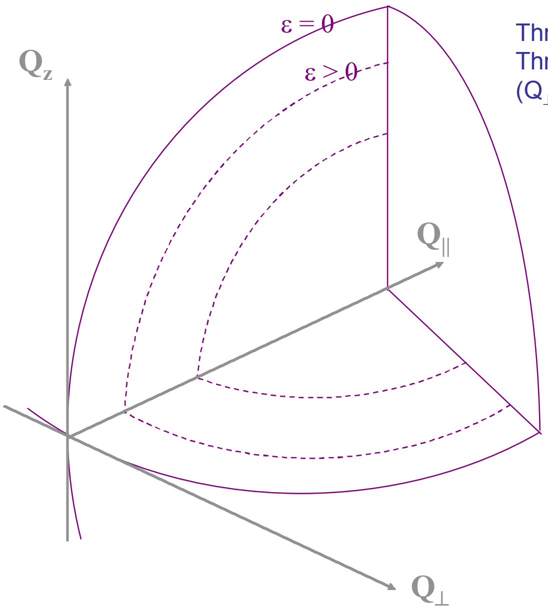
Physics in up to 4-dimensional space:

$(\mathbf{Q}, \varepsilon) \equiv (Q_{||}, Q_{\perp}, Q_z, \varepsilon)$

Three can be considered independent variables; the fourth is determined by those three.

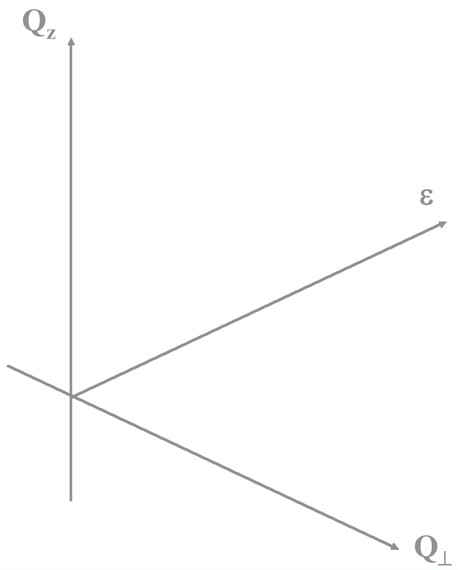


Measuring Excitations



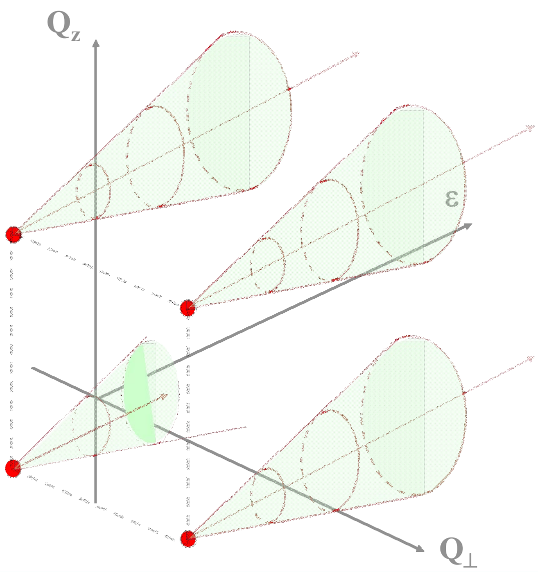
Three degrees of freedom
Three independent coordinates
$(Q_{\perp}, Q_z, Q_{||})$

Measuring Excitations



Three degrees of freedom
Three independent coordinates
$(Q_\perp, Q_z, \varepsilon)$

Measuring Excitations – 2D

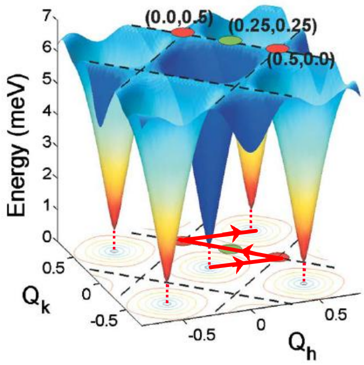


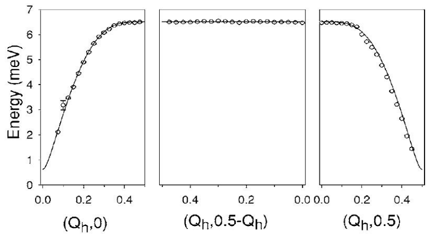
Rb_2MnF_4 - 2D S=5/2 antiferromagnet

- S=5/2 (so expect classical approximation to be OK);
- Two dimensional (so Néel state reasonable starting point)

Spin wave dispersion in linear spin wave theory:

$$\varepsilon(q_h, q_k) = 4JS\sqrt{(1+\delta_z)^2 - (\cos(2\pi q_h) + \cos(2\pi q_k))^2/4}$$





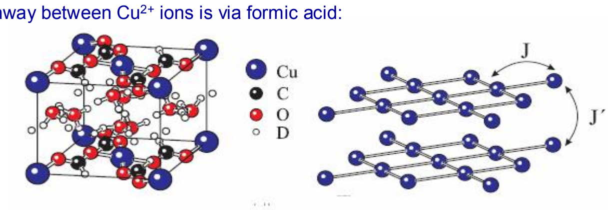
Full understood by conventional spin wave theory

T Huberman et al. *Phys Rev* B **72**, 014413 (2005)

CFTD - Copper Deuteroformate Tetradeuterate

CFTD: S=1/2 HAFM on a square lattice

Exchange pathway between Cu^{2+} ions is via formic acid:



$J' \approx 10^{-4} J$

With S=1/2, spin wave dispersion in linear spin wave theory:

$$\varepsilon(q_h, q_k) = 2J\sqrt{1 - (\cos(2\pi q_h) + \cos(2\pi q_k))^2/4}$$

Expansions in 1/S and series expansions from the Ising limit (i.e. $H = J\sum\left[\delta_{xy}(S_i^x S_j^x + S_i^y S_j^y) + S_i^z S_j^z\right]$ and expand in δ_{xy}) yield the same form, but with renormalisation of energy scale (c.f. 1D HAFM chain) and intensity:

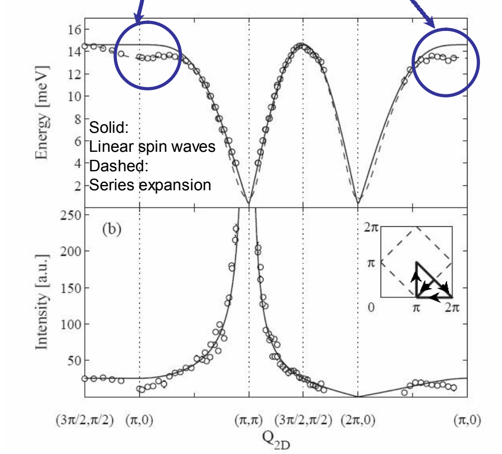
energy scale $Z_c = 1.18$

intensity scale $Z_\chi = 0.51$

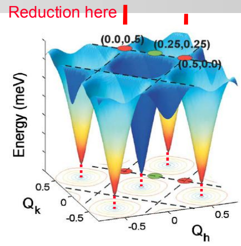
CFTD - Copper Deuteroformate Tetradeuterate

Deviation from linear spin wave dispersion of 7%

$$\varepsilon(q_h, q_k) = 2J\sqrt{1-(\cos(2\pi q_h)+\cos(2\pi q_k))^2/4}$$



Reduction here



Quantum Monte Carlo and exact diagonalisation on small clusters both reproduce this effect for nearest neighbour interactions only *(Syljuåsen et al., J.Phys. C 12 L1 (2000); Rønnow et al PRL 87 037202 (2001);*

true quantum many body effect

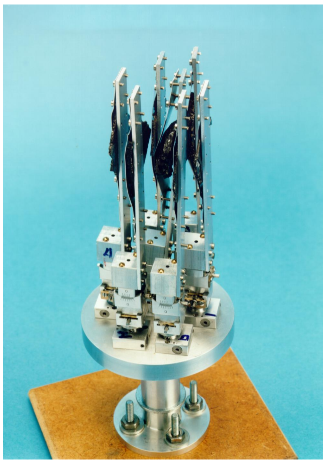
Intensity renormalised by $Z\chi$ = 0.51±0.04 (cf 0.51)

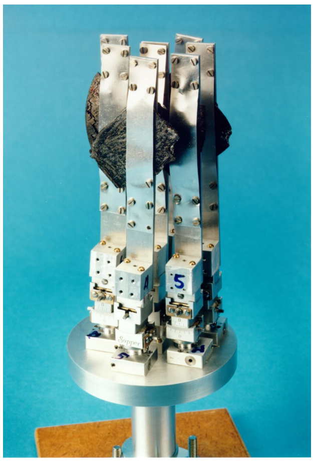
Also deviation at same point in Q: reduced to 0.45 ±0.12 of (renormalised) value. *QMC predicts reduction of 0.7.*

H.M.Rønnow et al., *Phys. Rev. Lett* **87** 037202 (2001); N B Christensen et al., *J Magn Magn Mater* **272-76** 896-897 (2004)

La_2CuO_4

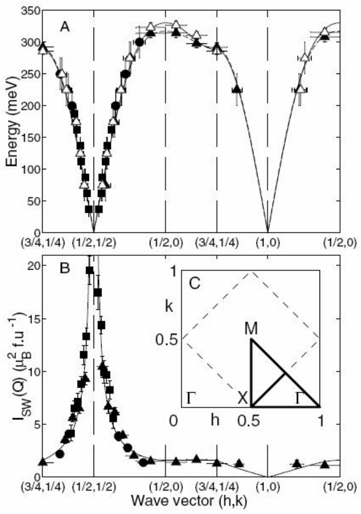
La_2CuO_4: also S=1/2 HAFM on a square lattice

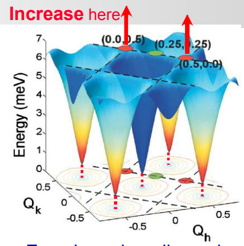


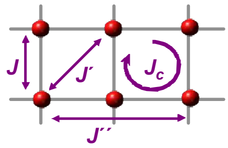


One of the big samples: ~40g

La_2CuO_4







Zone boundary dispersion opposite to CFTD – not explained by nearest neighbour exchange only

Characterise by next nearest neighbour interactions:

$$H = J\sum_{<ij>nn}\mathbf{S}_i.\mathbf{S}_j + J'\sum_{<ij>nnn}\mathbf{S}_i.\mathbf{S}_j + J''\sum_{<ij>nnnn}\mathbf{S}_i.\mathbf{S}_j + J_c\sum_{<ij>}\left\{(\mathbf{S}_i.\mathbf{S}_j)(\mathbf{S}_k.\mathbf{S}_l) + (\mathbf{S}_i.\mathbf{S}_l)(\mathbf{S}_k.\mathbf{S}_j) - (\mathbf{S}_i.\mathbf{S}_k)(\mathbf{S}_j.\mathbf{S}_l)\right\}$$

J = 112 ± 4meV

J' = -11 ± 3 meV (ferromagnetic)

or

J = 139 ± 4meV

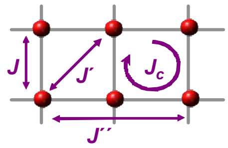
J_c = 38 ± 8 meV

R Coldea, S M Hayden, G Aeppli, T G Perring, C D Frost, T E Mason, et al *Phys Rev Lett* 86 5377 (2001)

La_2CuO_4

Cyclic dispersion ***Jc*** naturally arises from finite *U/t* in analysis of Hubbard model:

$$H = -t\sum_{<ij>,\sigma=\uparrow\downarrow}\left(c_{i\sigma}^{+}c_{j\sigma} + H.c.\right) + U\sum_i n_{i\uparrow}n_{i\downarrow}$$



Leads to the effective spin Hamiltonian with coupling constants:

$$J = 4t^2/U - 24t^4/U^3$$
$$J' = J'' = 4t^4/U^3$$
$$J_c = 80t^4/U^3$$

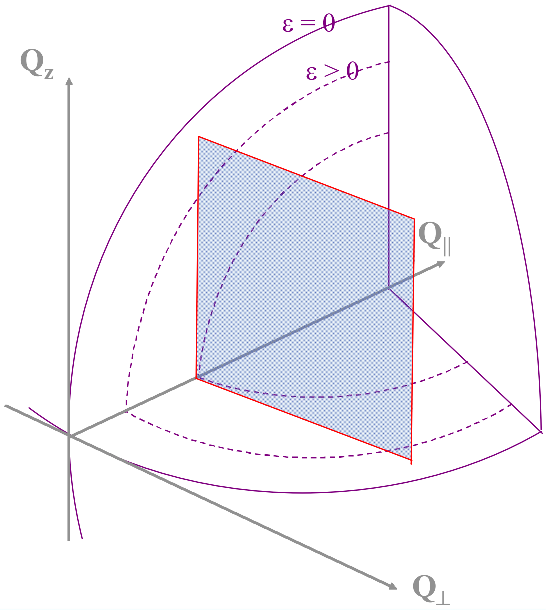
Cyclic dispersion is 20x the size of the next and next-next nearest neighbour terms; leads to

t = 0.33 ±0.02 eV

U = 2.9 ±0.4 eV

In agreement with values determined from photoemission and optical spectroscopy

Measuring Excitations

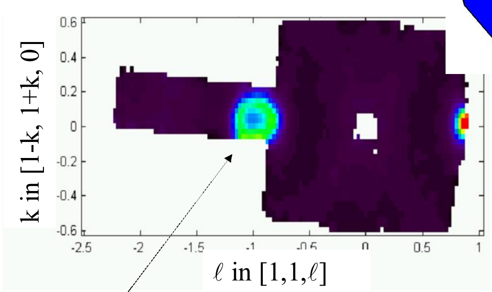


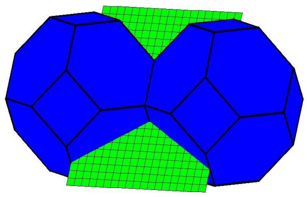
Align symmetry plane if crystal perpendicular to incident wavevector.

Plane is tangential to constant energy surface so is ***approximately*** constant energy surface

Nickel - canonical itinerant ferromagnet in 3D

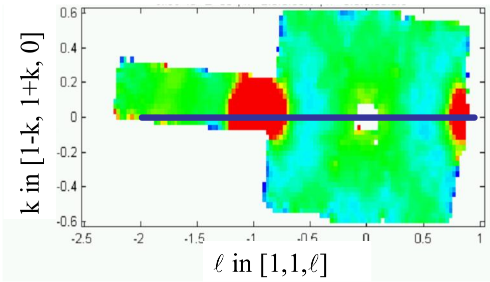
Excitation energy = 50 meV (45-55 meV)

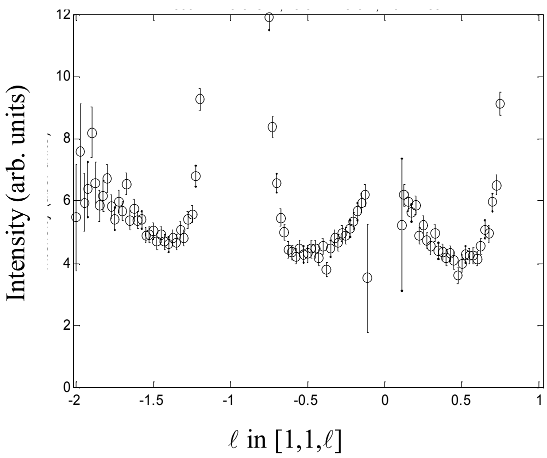




Spin waves around (1,1,-1)

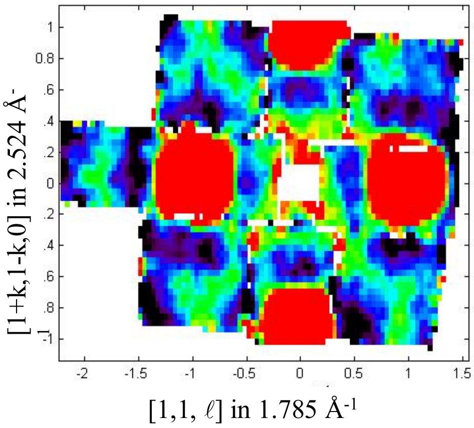
S.M.Hayden, T.G.Perring, O.Stockert, H.A. Mook


k in [1-k, 1+k, 0]
ℓ in [1,1,ℓ]

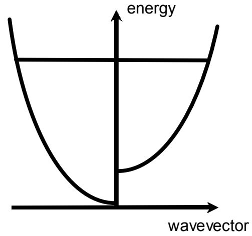

Intensity (arb. units)
ℓ in [1,1,ℓ]

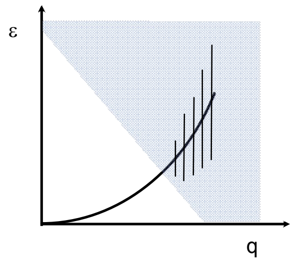
Nickel

Excitation energy = 90 meV

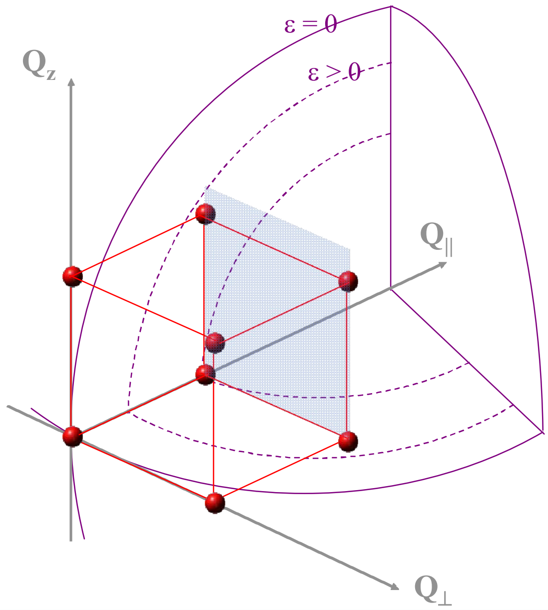


Zone boundary modes observed well above ε = 300 meV





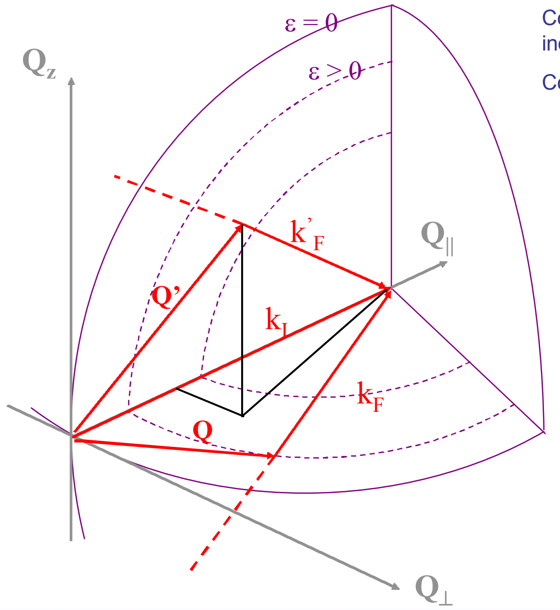
Measuring Excitations - 3D



Three degrees of freedom

Four independent coordinates: $(Q_{||}, Q_{\perp}, Q_z, \varepsilon)$

Measuring Excitations

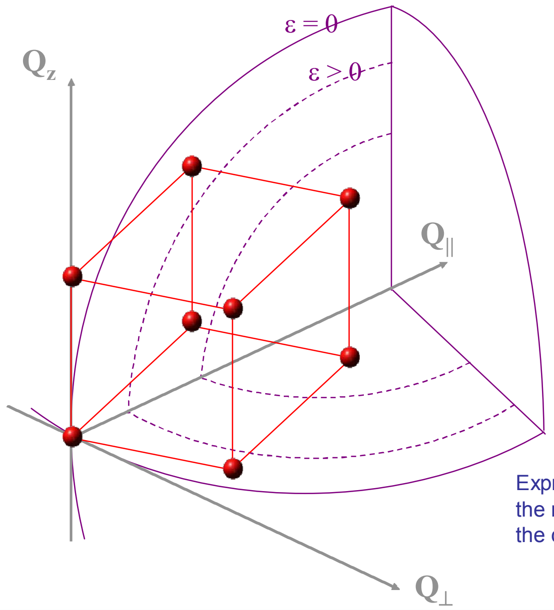


Consider $Q_{||}$, $Q_{\perp}$, Q_z as independent variables

Constant ε surfaces are spheres:

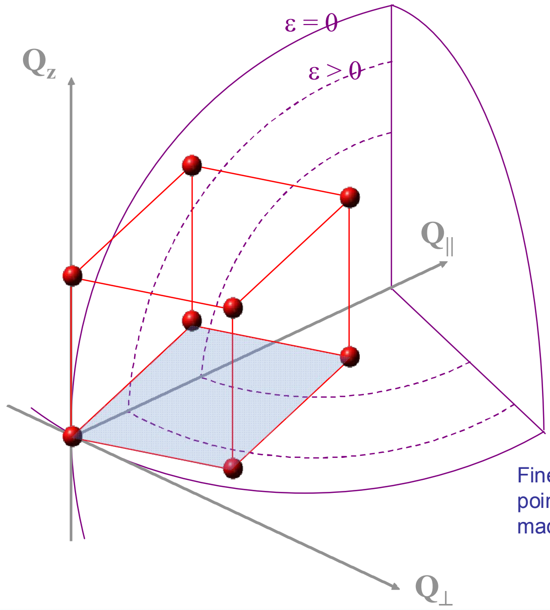
$\varepsilon = (\hbar^2/2m)\,(k_i^2 - k_f^2)$

Measuring Excitations



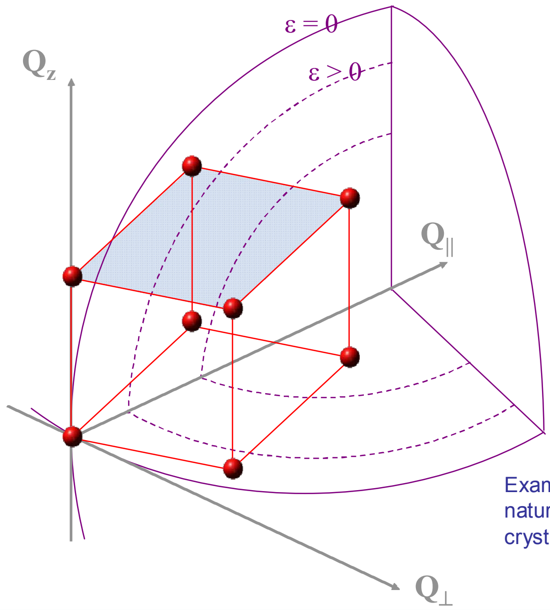
Express **Q** in components along the reciprocal lattice vectors of the crystal

Measuring Excitations



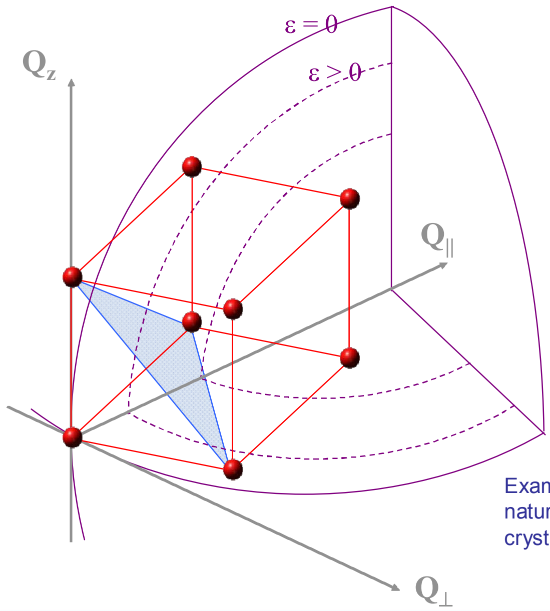
Fine pixellation of the volume (~10^7 points) allows arbitrary slices to be made through this space

Measuring Excitations



Examine the excitations in the natural physical space of the crystal's reciprocal lattice

Measuring Excitations



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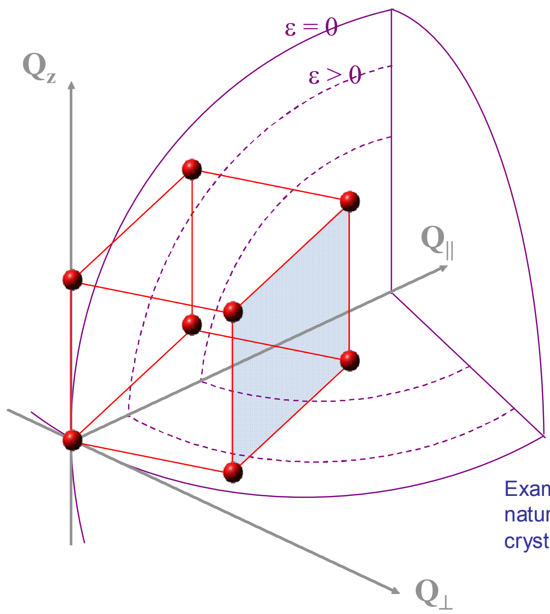
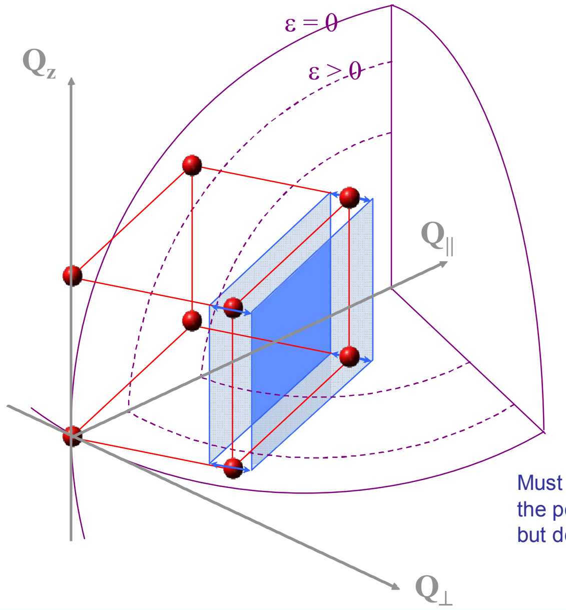
Measuring Excitations



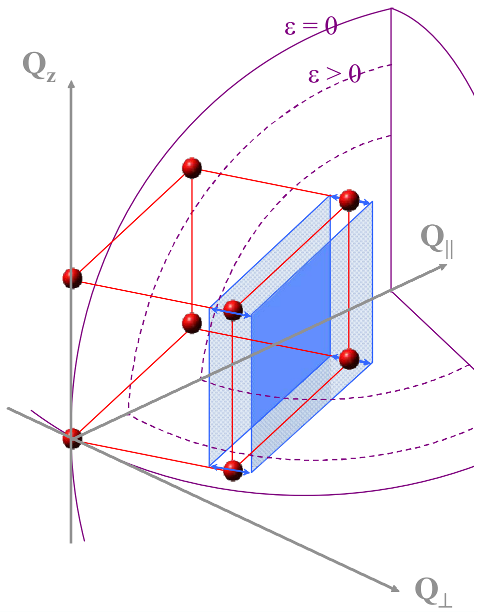
Examine the excitations in the natural physical space of the crystal's reciprocal lattice

Measuring Excitations

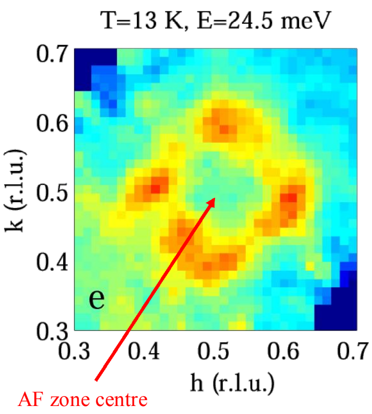


Must add a thickness to the slice – the position sensitivity gives control, but does not increase the count rate

Measuring Excitations



Incommensurate spin fluctuations in $YBa_2Cu_3O_{6.6}$



Why does the technique work ?

Large solid angle of detectors and close to continuous coverage:

- 1-D and 2-D systems: appropriate configuration provides large single effective detector
- Comprehensive views of S(**Q**,ω) gathered in parallel
- Non-magnetic "backgrounds" gathered simultaneously
- Broad features clearly visible
- Copious cross-checks in different zones

Low intrinsic backgrounds:

- Fixed detectors + evacuated flight-path
  - ϕ_{min} ~ 3°
- Lack of spurions with Fermi chopper
  - Chopper is open every $2\pi/\omega$
  - e.g. At E_i=100 meV, 400Hz: spurion at ε/E_i = 90%
- Source is 'off' between pulses
  - Low intrinsic electronic noise on detectors ~ 3 cnt/hr/30cm

Review: why does the technique work ?

Computing power + analysis software:

Visualisation + analysis

Integral part of the spectrometer

“Tertiary Spectrometer”

Comparison of triple-axis and time-of-flight technique

Time-of-flight chopper instruments

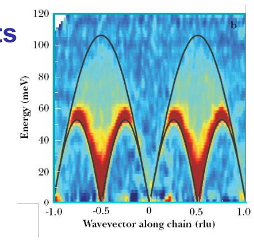
- Equivalent workhorse spectrometer
- 3 at each of ISIS, SNS, J-PARC:
  - -high resolution, high flux, cold
  - MAPS MERLIN LET
- Established for single crystal measurements

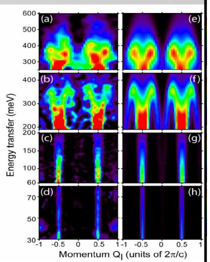
Why successful?

- Comprehensive measurement of S($\mathbf{Q}$,ε)
  - Intrinsically **parallel**
  - Large solid angle and bandwidth
  - Full tests of models for S($\mathbf{Q}$,ε)
- Complementary to triple axis
- Negligible background

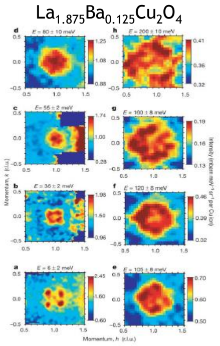
But…

- Highly successful in 1D and 2D systems
  - S=1/2 chain, square lattice, high-Tc, …
- Not so many studies in 3D
  - Co, $La_{0.7}Pb_{0.3}MnO_3$, Cr(V), $La_{1-x}Ca_xMnO_3$





$La_{1.875}Ba_{0.125}Cu_2O_4$



Why do we need to measure in 3D systems ?

- **Real materials are 3D**
  - Quantum magnetism:
    - -Model systems are chosen to be 1D or 2D
    - -Real systems often have non-negligible interchain/interplanar interactions
    - -Want to follow cross-over as these interactions appear…
  - Complex materials of current interest intrinsically 3D:
    - -multiferroics, frustrated magnetic systems, CMR, quantum critical materials, classic ferromagnets (MnSi, Cr, Fe/Co/Ni…)
    - -Coupled degrees of freedom $\Rightarrow$ hybrid modes (spin-phonons), (orbital/magnetic)
- **Need survey capability**
  - Do not know a priori where to look in ($\mathbf{Q}$,ε)
    - e.g. bilayer manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$:
      - AF fluctuations (0,0.5,ℓ) Polaron correlations (0.3,0,1) CE correlations (0.25,0.25,0)
  - -Phonons, hybrid modes need many zones to disentangle
    - \- Phonons: $S(\mathbf{Q},\varepsilon) \sim (\mathbf{u}.\mathbf{Q})^2$
  - -Zone boundary modes need constant–Q scans to measure
  - -Extract lifetimes in model independent fashion
  - -Full $S(\mathbf{Q},\varepsilon)$ gives most exacting test of theory

Make surveys of $S(Q,\varepsilon)$ in 3D systems just as routine as in 1D, 2Dsystems

Summary

Shown that full dispersion in 3D materials can be measured with time-of-flight

- Used large scattering samples – but not excessively so
  - 10g of $RbMnF_3$ data (3days) $\Rightarrow$ 50g of S=1/2 crystal
- Instruments at ISIS: MERLIN (commissioning) and LET (construction)
  - 10x flux $\Rightarrow$ 5g of CMR manganite in 36hrs
  - Multiple Ei scans – LET will have several Ei simultaneously
- New facilities SNS J-PARC:
  - 1.4 MW c.f. 200KW $\Rightarrow$ 5g of CMR manganite in 5hrs …

Effective, useable, software essential to project

- Software integral part of the spectrometer
- Early stages of development:
  - Not yet as fast as can be achieved on desktop machine
  - Solution: parallel processing and parallel visualisation
    - Already resolution calculation - 'Tobyfit' - is on Grid)
    - Exploring use of 34 processor visualisation server

Believe will become the routine technique for single crystal spectroscopy at pulsed sources