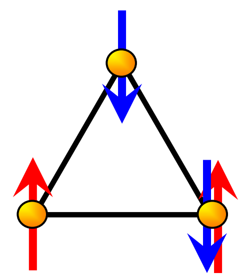
LQM

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

Neutron Scattering in Magnetism
- focus on dynamics



Winter School on Magnetism, Stuttgart 2008

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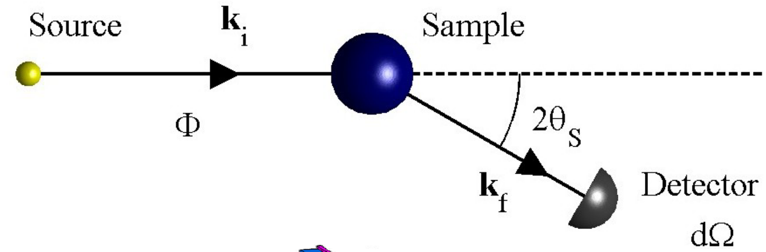
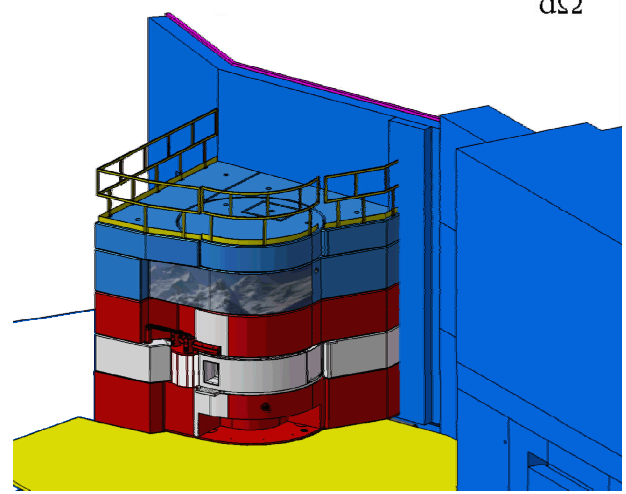
Outline

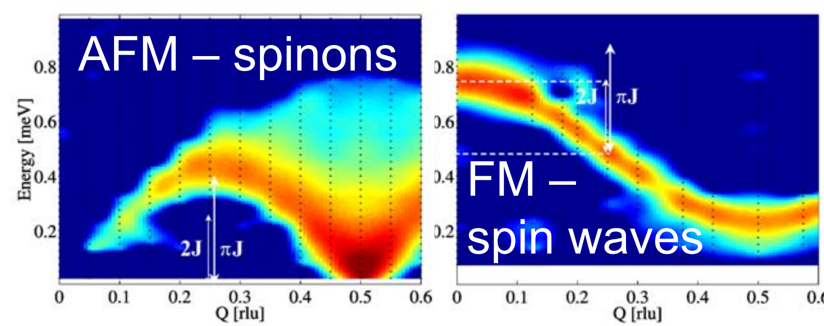
1) Theory
   – what can we measure
   the cross-section





2) Experiment
   – how do we measure
   instruments and methods

3) Physics
   – what do we measure
   examples



“Theory” – what do we measure

– Plane waves: diffraction and magnetic structures

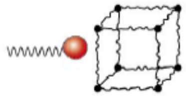
– Conservation: momentum, energy, spin

– Cross-section

– Magnetic scattering

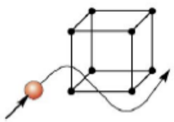
– Correlation function

Neutrons



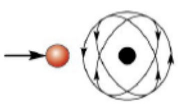
The ENERGIES of thermal neutrons are similar to the energies of elementary excitations in solids. Both have similar

- molecular vibrations,
- lattice modes, and
- dynamics of atomic motion.



The WAVELENGTHS of neutrons are similar to atomic spacings. They can determine

- structural sensitivity,
- structural information from 10^{-13} to 10^{-4} cm, and
- crystal structures and atomic spacings.



Neutrons "see" NUCLEI. They

- are sensitive to light atoms,
- can exploit isotopic substitution, and
- can use contrast variation to differentiate complex molecular structures.

Neutrons are NEUTRAL particles. They

- are highly penetrating,
- can be used as nondestructive probes, and
- can be used to study samples in severe environments.

Neutrons have a MAGNETIC moment. They can be used to

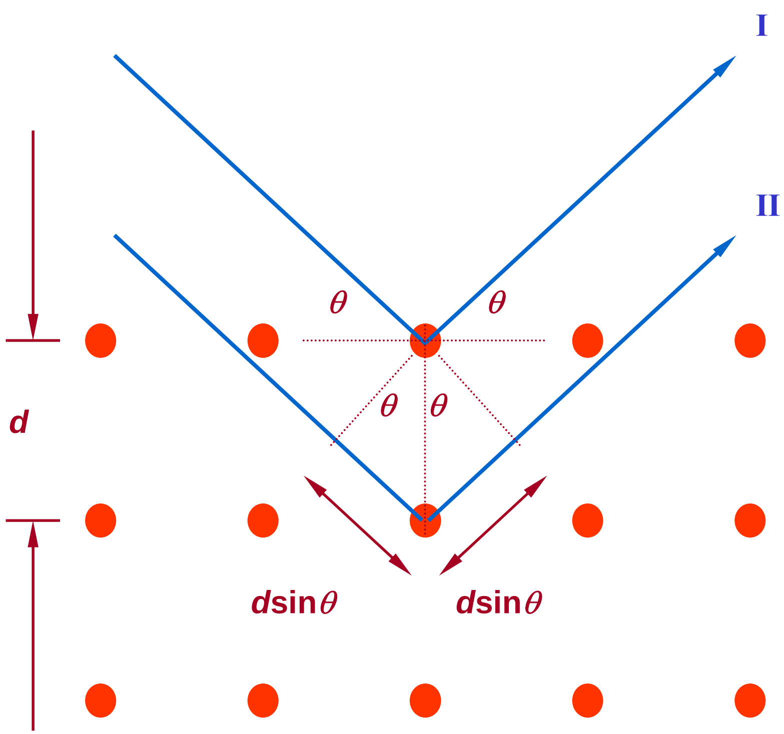
- study microscopic magnetic structure,
- study magnetic fluctuations, and
- develop magnetic materials.

Neutrons have SPIN. They can be

- formed into polarized neutron beams,
- used to study nuclear (atomic) orientation, and
- used for coherent and incoherent scattering.

Neutron Plane Waves

- Plane waves and Bragg's law



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

Diffraction – magnetic structures

- Bragg's law:

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

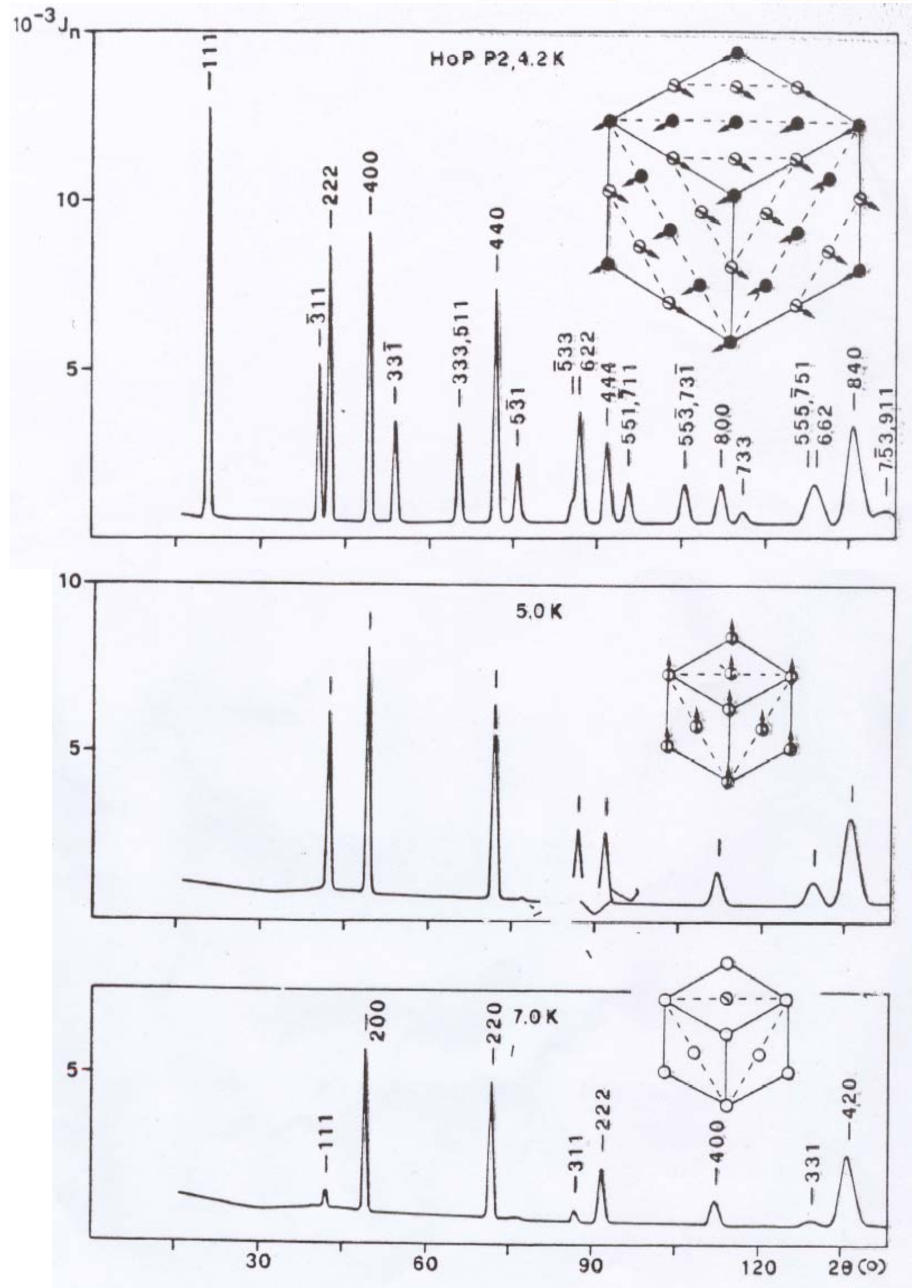
- Reciprocal lattice

  $\tau = ha^* + kb^* + lc^*$

  $\tau = 2k \sin\Theta$

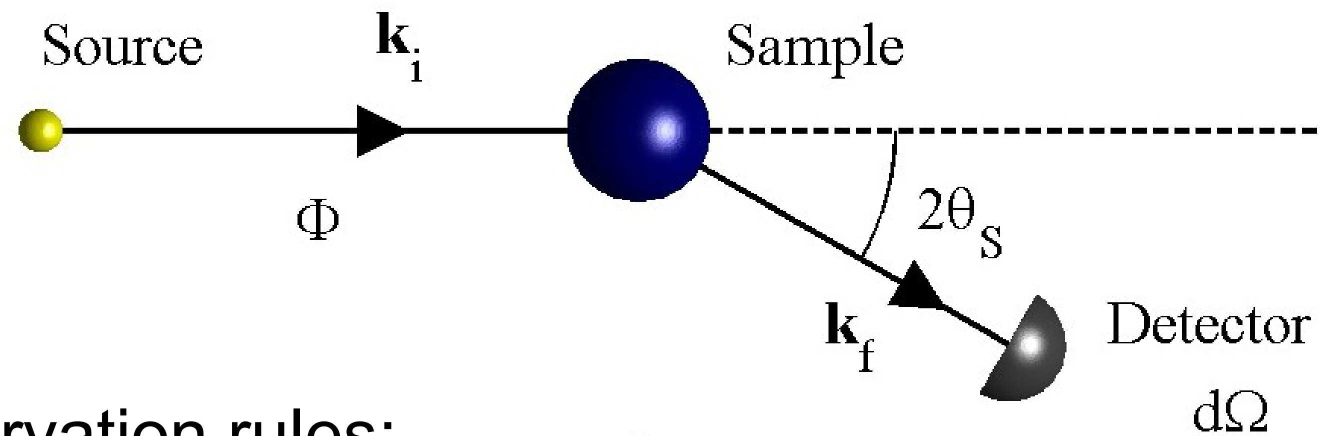
- Powder diffraction – example: HoP
  - Structural peaks at 7K
  - Ferromagnet at 5K
  - Non-collinear at 4.2K

- Structure refinement:
  - Standard 'black box' methods
  - Absorption, mutimple scattering, texture etc.
  - Positions, displacements, moments, directions



Scattering & conservation rules

Consider scattering neutrons on a sample into a detector.



Conservation rules:

	Sample		Neutron	
Momentum	$\hbar\mathbf{Q}$	=	$\hbar\mathbf{k}_i - \hbar\mathbf{k}_f$	
Energy	$\hbar\omega$	=	$E_i - E_f$	$= \hbar(k_i^2 - k_f^2)/2m_n$
Spin	ΔS	=	$\sigma_i - \sigma_f$	

$\Rightarrow$ We can control and measure these quantities !

Cross-section

Intensity in angular $d\Omega$ and energy dE_f elements:

$$I_{d\Omega\ dE_f} = \left(\frac{d^2\sigma}{d\Omega\ dE_f}\right) \Phi\ d\Omega\ dE_f$$

From initial state ***i*** to final state ***f*** of neutron $\mathbf{k}$ and sample λ

$$\left(\frac{d^2\sigma}{d\Omega\ dE_f}\right)_{\lambda_i\to\lambda_f} = \frac{k_f}{k_i}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 |\langle \mathbf{k}_f\lambda_f\,|V|\,\mathbf{k}_i\lambda_i\rangle|^2\ \delta(E_{\lambda_i} - E_{\lambda_f} + \hbar\omega)$$

Neutrons treated as plane waves:

$$|\mathbf{k}\mathbf{s}_n> = V^{-1/2}\exp(i\mathbf{k}\cdot\mathbf{r}_n)\,|\mathbf{s}_n>$$

Energy conservation ⇒ integral rep.:

$$\delta(\hbar\omega + E_i - E_f) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} e^{i(\hbar\omega+E_i-E_f)t/\hbar}dt$$

Fourier transform in

- space/momentum
- time/energy

Magnetic scattering

$$|\langle \mathbf{k}_f \lambda_f \, | V | \, \mathbf{k}_i \lambda_i \rangle|^2$$

Dipole interaction – electron spin and orbit

$$V_{\mathrm{mag}}(\mathbf{r}) = -\frac{\mu_0}{4\pi} 2\gamma \mu_N \mu_B \, \boldsymbol{\sigma}_n \cdot \left(\nabla \times \left(\frac{\mathbf{s} \times \hat{\mathbf{R}}}{|\mathbf{R}|^2} \right) + \frac{1}{\hbar} \frac{\mathbf{p} \times \hat{\mathbf{R}}}{|\mathbf{R}|^2} \right)$$

Non-polarised neutron average over spin states:

$$\left(\frac{d^2\sigma}{d\Omega \, dE_f} \right)_{\mathrm{mag}} = \frac{(\gamma r_0)^2}{2\pi\hbar} \frac{k_f}{k_i} \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{\mathrm{Q}}_\alpha \hat{\mathrm{Q}}_\beta) \sum_{\boldsymbol{l}'\boldsymbol{d}'\boldsymbol{l}\boldsymbol{d}} \frac{1}{4} g_{\boldsymbol{d}'} g_{\boldsymbol{d}} F^*_{\boldsymbol{d}'}(\mathbf{Q}) F_{\boldsymbol{d}}(\mathbf{Q})$$

$$\times \int_{-\infty}^{\infty} dt \exp(-i\omega t) \langle \exp\{-i\mathbf{Q}\cdot\mathbf{R}_{\boldsymbol{l}'\boldsymbol{d}'}(0)\} \exp\{i\mathbf{Q}\cdot\mathbf{R}_{\boldsymbol{l}\boldsymbol{d}}(t)\} \rangle \langle S^\alpha_{\boldsymbol{l}'\boldsymbol{d}'}(0) S^\beta_{\boldsymbol{l}\boldsymbol{d}}(t) \rangle$$

- fudge factor
- dipole factor
- magnetic form factor
- spin-spin correlation function
- Fourier transform

Polarised neutrons:

$$\left(\frac{d^2\sigma}{d\Omega \, dE_f} \right)_{s_i \to s_f} = \frac{k_f}{k_i} \sum_{if} P_{\lambda_i} \, | \, \langle \lambda_f \, | \sum_{\boldsymbol{l}} \exp(i\mathbf{Q}\cdot\mathbf{r}_{\boldsymbol{l}}) U^{s_i s_f}_{\boldsymbol{l}} \, | \, \lambda_i \rangle \, |^2 \, \delta(E_{\lambda_i} - E_{\lambda_f} + \hbar\omega)$$

$$U^{s_i s_f}_{\boldsymbol{l}} = \langle s_f \, | \, b_{\boldsymbol{l}} - p_{\boldsymbol{l}} \, \boldsymbol{\sigma}_n \cdot \mathbf{S}_{\boldsymbol{l}\perp} + B_{\boldsymbol{l}} \, \boldsymbol{\sigma}_n \cdot \mathbf{I}_{\boldsymbol{l}} \, | \, s_i \rangle$$

Dynamic structure factor

Spin-spin correlation function

$$S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi}\sum_{\boldsymbol{l}'\boldsymbol{d}'\boldsymbol{l}\boldsymbol{d}} \exp(-i\mathbf{Q}\cdot\{\boldsymbol{l}'+\boldsymbol{d}'-\boldsymbol{l}-\boldsymbol{d}\})\int_{-\infty}^{\infty} dt\,\exp(-i\omega t)\langle S^{\alpha}_{\boldsymbol{l}'\boldsymbol{d}'}(0)S^{\beta}_{\boldsymbol{l}\boldsymbol{d}}(t)\rangle$$

Dynamic structure factor

Theory !

$$\left(\frac{d^2\sigma}{d\Omega\, dE_f}\right)_{\mathrm{mag}} = \frac{1}{\hbar}\frac{k_f}{k_i}p^2\exp(-2W)\sum_{\alpha\beta}(\delta_{\alpha\beta}-\hat{\mathbf{Q}}_\alpha\hat{\mathbf{Q}}_\beta)S^{\alpha\beta}(\mathbf{Q},\omega)$$

Fluctuation dissipation theorem $\Rightarrow$ gen. susceptibility

$$S(\mathbf{Q},\omega) = [\,n(\omega)+1\,]\,\chi''(\mathbf{Q},\omega) = \frac{\chi''(\mathbf{Q},\omega)}{1-\exp(-\hbar\omega/k_BT)}$$

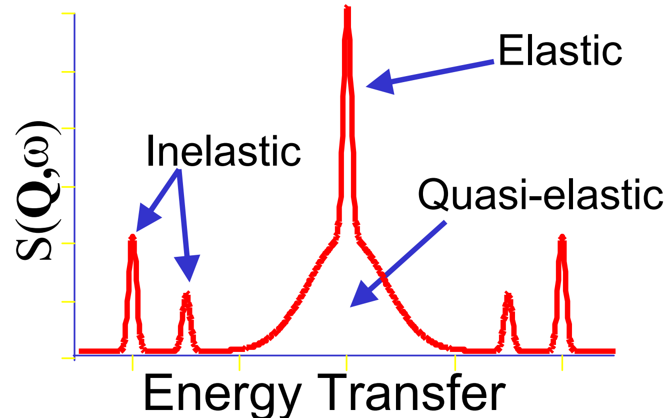
intrinsic dynamics $\Leftrightarrow$ response to perturbation

Structure factors – time and energy

- Dynamic structure factor: inelastic

$$S(\mathbf{Q},\omega) \propto \int_{-\infty}^{\infty} dt e^{-i\omega(t-t')} \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t')\rangle$$

  – periodic: $\sin(\omega_0 t) \Rightarrow$ peak: $\delta(\omega_0-\omega)$



- Static structure factor: elastic

$$S(\mathbf{Q},\omega=0) \propto \int_{-\infty}^{\infty} dt \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t')\rangle \simeq \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(\infty)\rangle$$

  – Bragg peaks at $\omega = 0$

- Instantaneous structure factor

$$S(\mathbf{Q}) = \int d\omega S(\mathbf{Q},\omega) \propto \int_{-\infty}^{\infty} dt \delta(t-t') \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t')\rangle = \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t)\rangle$$

  – Exponential decay: $\exp(-t/\tau) \Rightarrow$ Lorentzian: $1/(1+\omega^2\tau^2)$

Recap: what do we measure

Conservation rules $\Rightarrow$ momentum, energy, spin dependence

Intensity $\Rightarrow$ Cross-section, magnetic dipole interaction

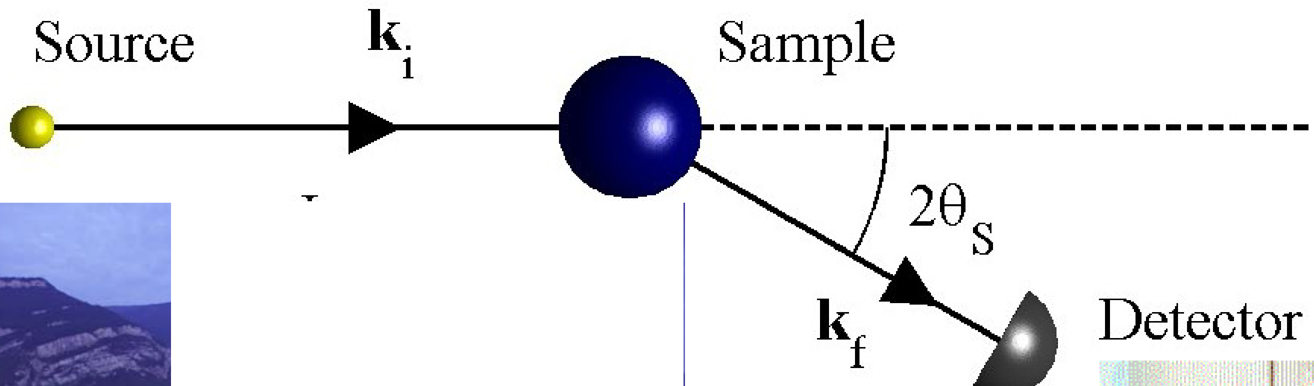
Correlation function $\Rightarrow$ dynamic structure factor:

$$\text{Intensity} \propto \left(\frac{d^2\sigma}{d\Omega\, dE_f}\right)_{\text{mag}} = \frac{1}{\hbar}\frac{k_f}{k_i} p^2 \exp(-2W) \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{\mathbf{Q}}_\alpha \hat{\mathbf{Q}}_\beta) S^{\alpha\beta}(\mathbf{Q}, \omega)$$

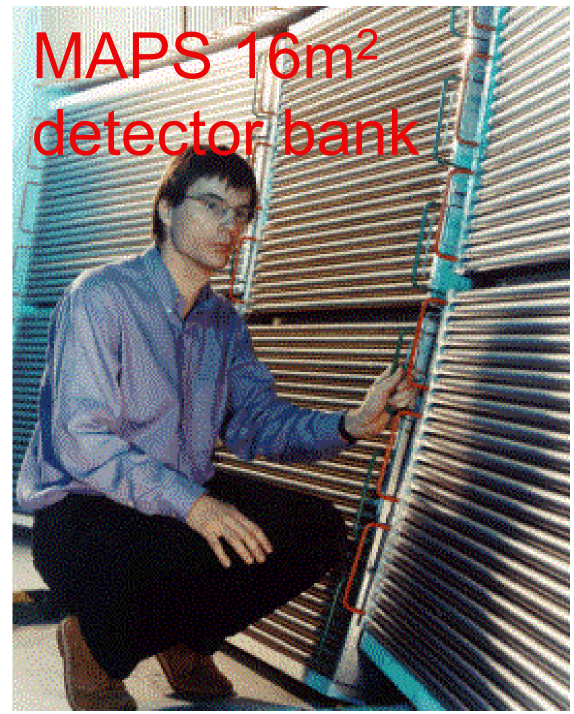
$\Uparrow$ Experiment

$\Uparrow$ Theory

Experiment – how do we measure *instruments and methods*

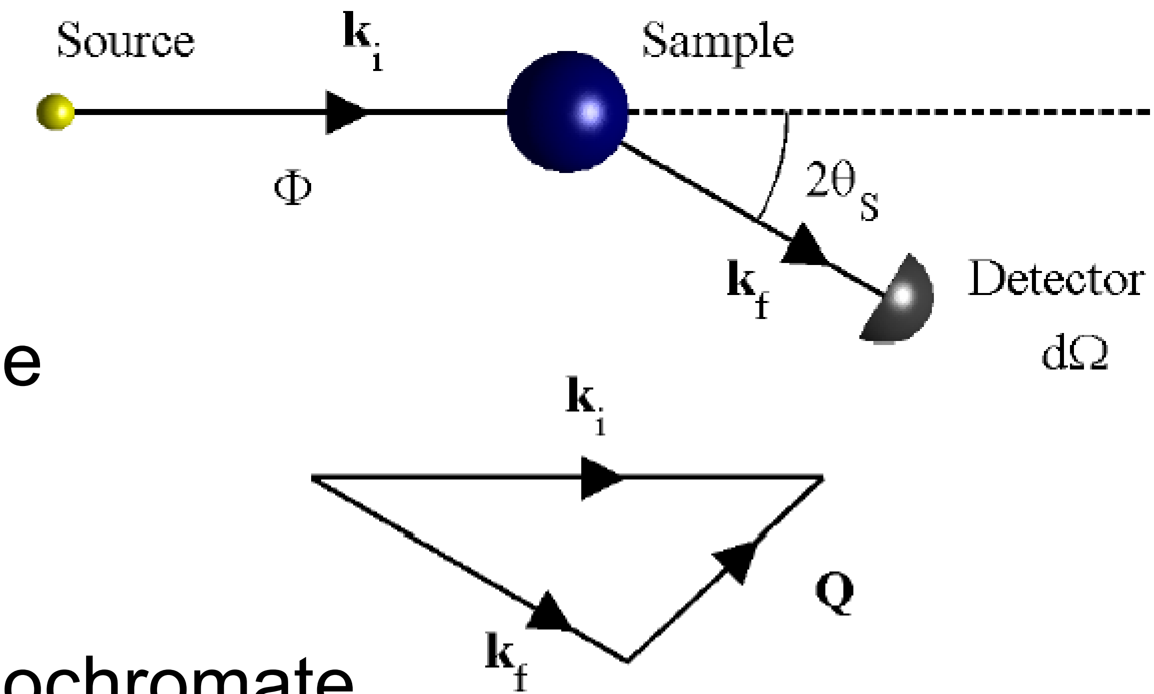


Christian Rüegg



Chris Frost

Energies and angles

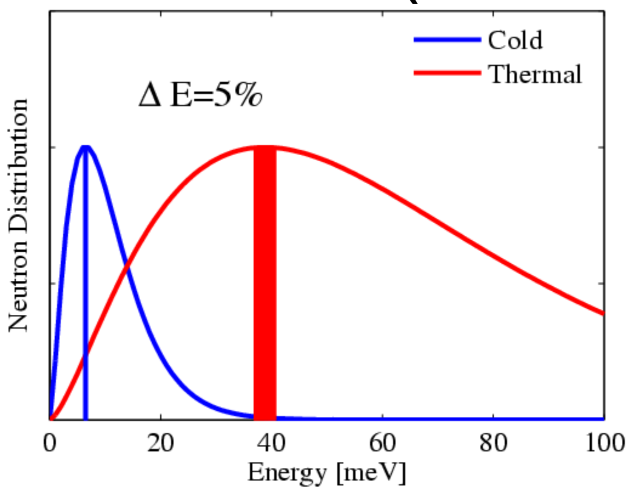


- Need to define $\mathbf{k}_i$, $\mathbf{k}_f$ and angle between them relative to sample

- Define $E_i \propto |\mathbf{k}_i|^2 \Rightarrow$ monochromate
- Know angle 2θ between ki and kf relative to sample orientation
- Determine $E_f \propto |\mathbf{k}_f|^2$
  $\Rightarrow$ analyse

The neutron energy

- We cannot (controlled) change the neutron energy



Must select neutrons with wanted energy from a (Maxwellian) distribution

(Liouville & the phase space transformer)

- We have no energy sensitive neutron detector… yet!

  n + ^{3}He $\rightarrow$ ^{3}H + ^{1}H + 0.764 MeV – want meV need 10^{-9} precision!

- Make another selection (analyse) to know energy
- Or, if we know starting time we can use time-of-flight

Useful numbers

- Energy units:

eU 1 meV =

$k_B T$ 11.6 K =

$h\nu$ 0.24 THz =

hc/λ 8.06 cm-1 =

$\mu_B H$ 17.3 T =

E 1.6x10-22 J

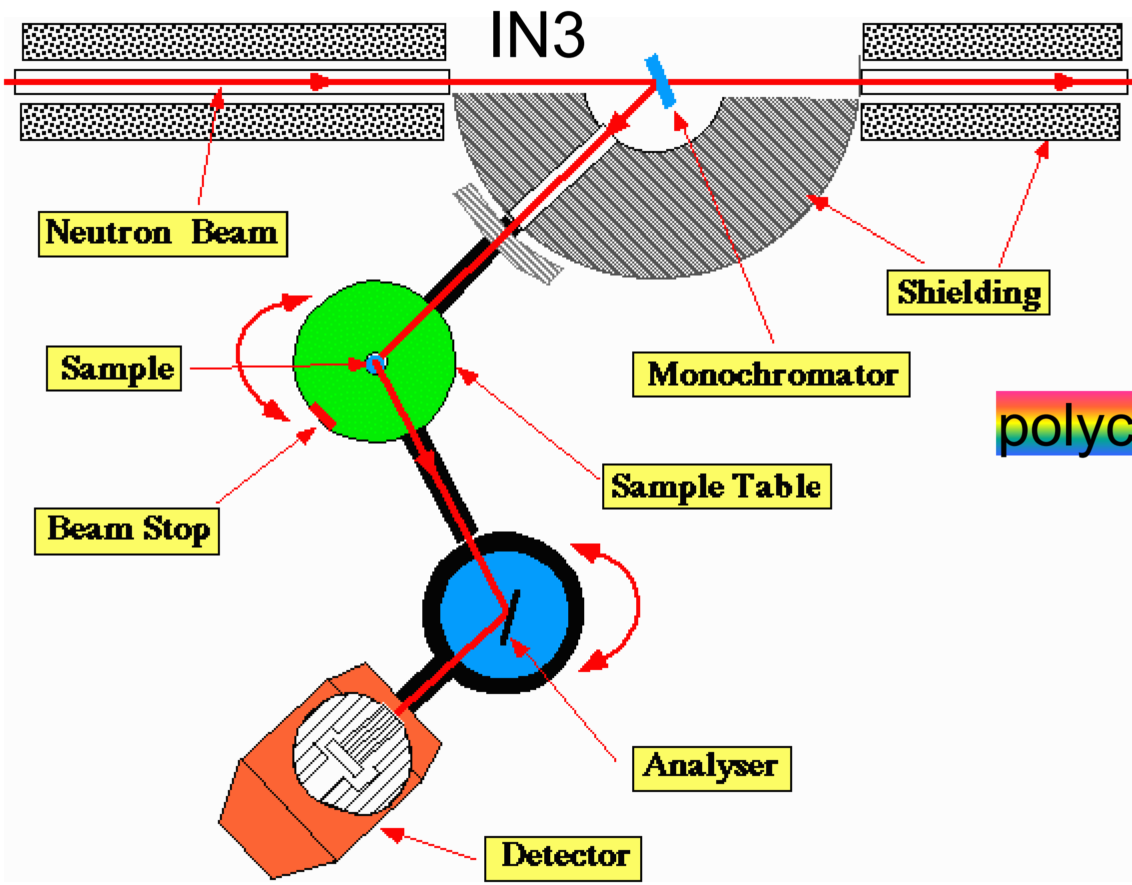
- Neutron Energies:

E [meV] =

$2.072\ k^2$ [meV Å^2] =

$81.8\ /\ \lambda^2$ [meV Å^2]

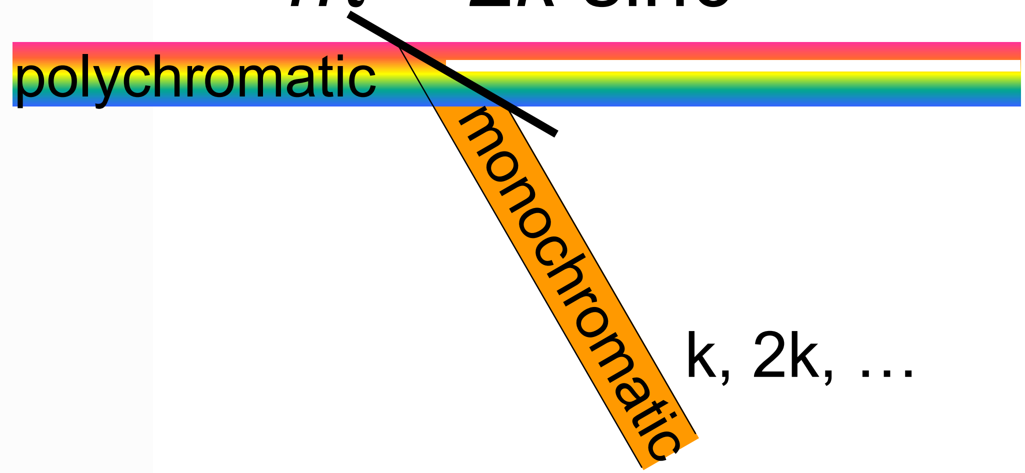
TAS: three axis spectrometer



Bragg's law

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

$n\tau = 2k \sin\theta$



Higher order (2k) filters:

Be: <5 meV

BeO: <3.8 meV

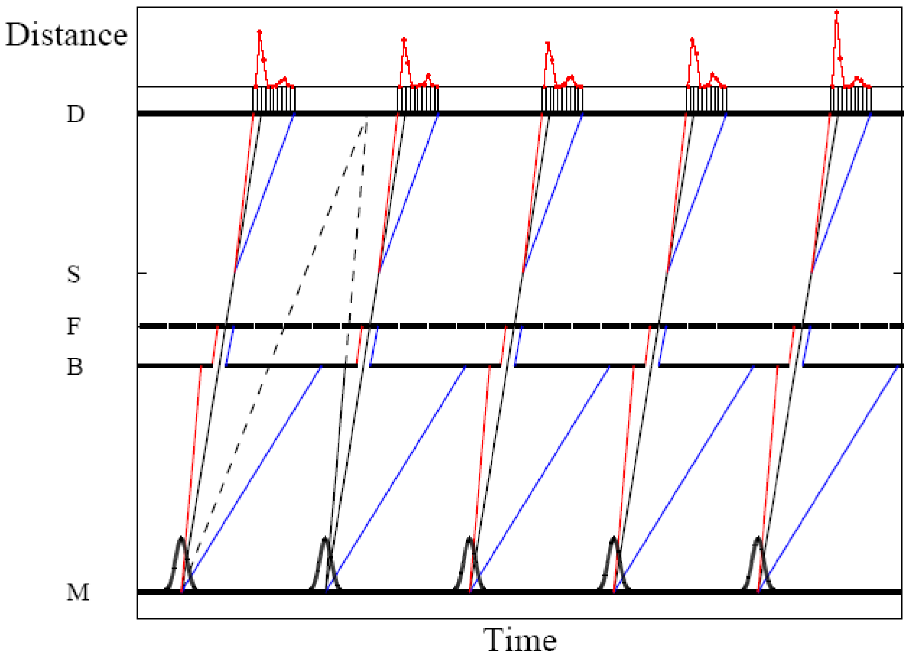
PG: 14.7 & 35 meV

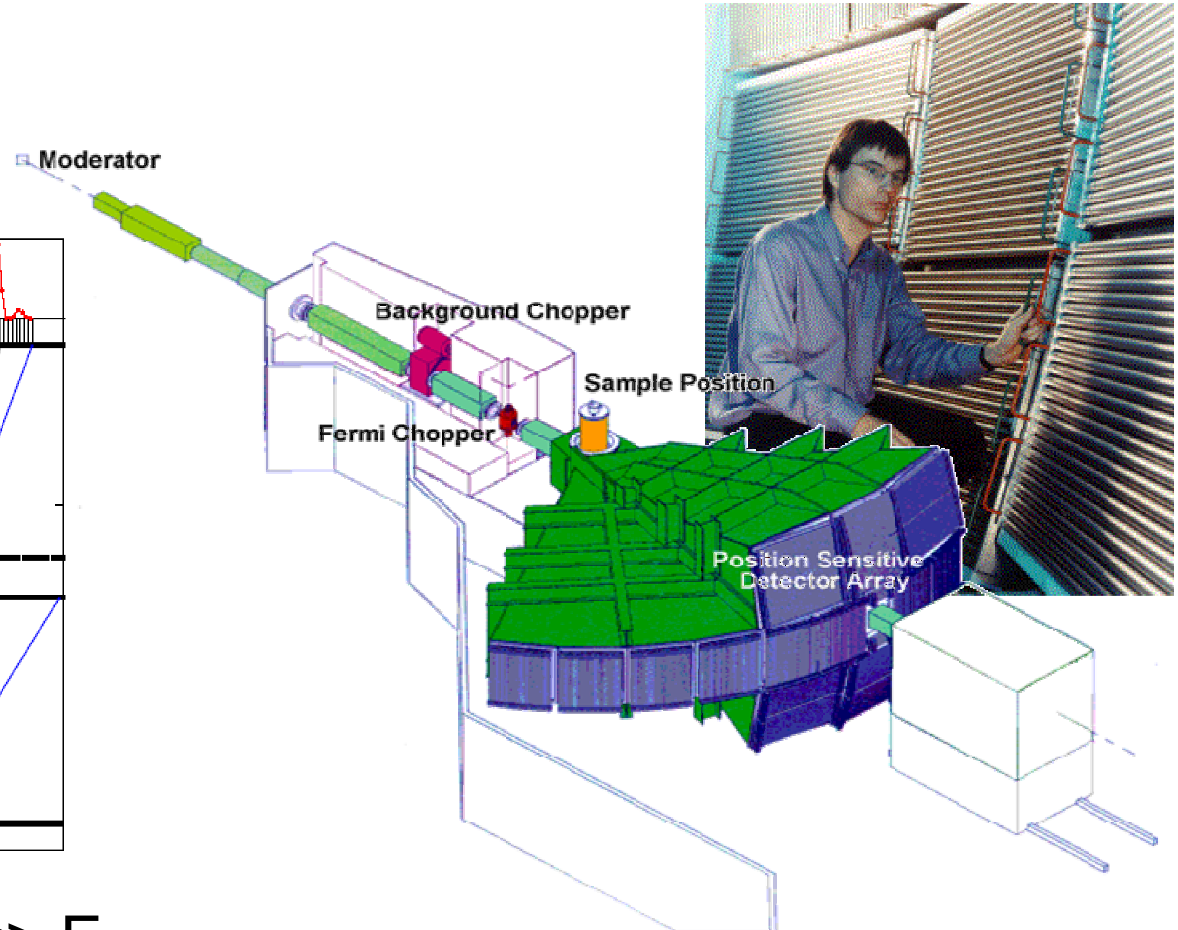
velocity selector: 'any'

TOF: time-of-flight

Pulsed beam

known start time





Monochromatising chopper => E_i

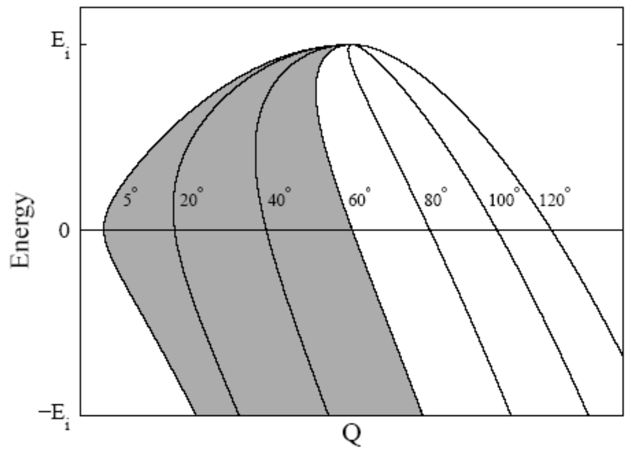
Time-of-flight => E_f

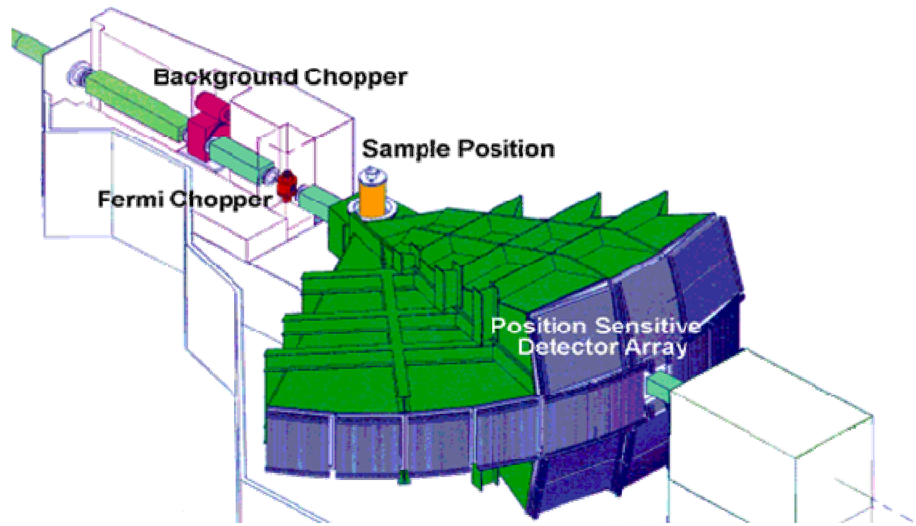
Advantage: nothing between sample and detector => easy multiplexing

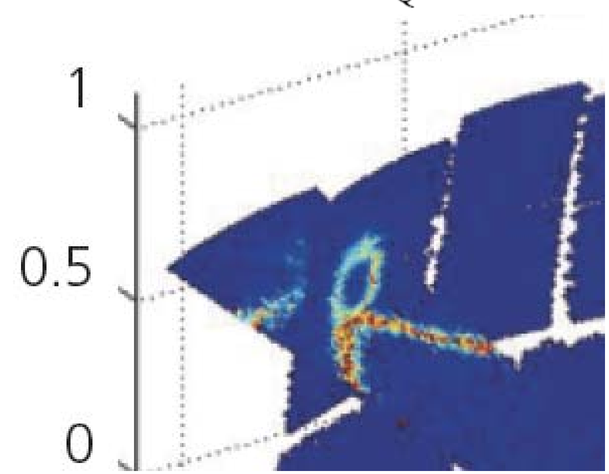
TOF: Q-E parabolas and maps

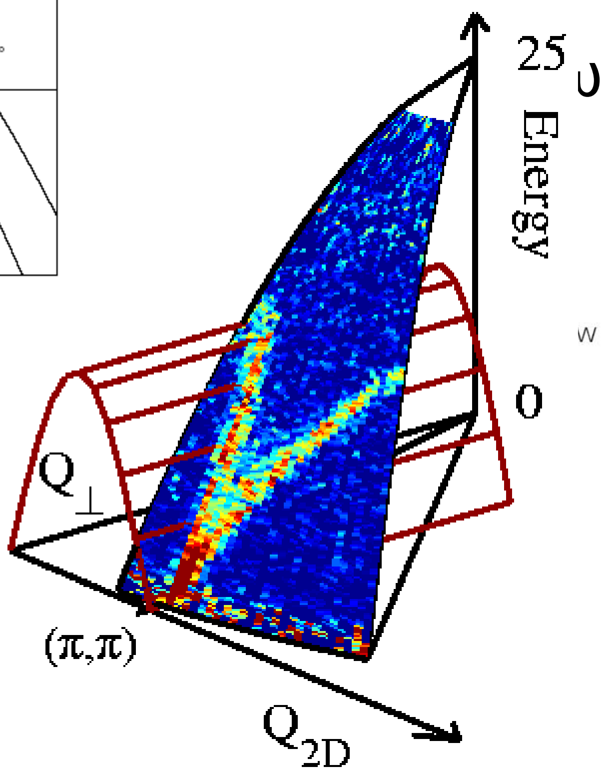
- Q for fixed E_i and θ:

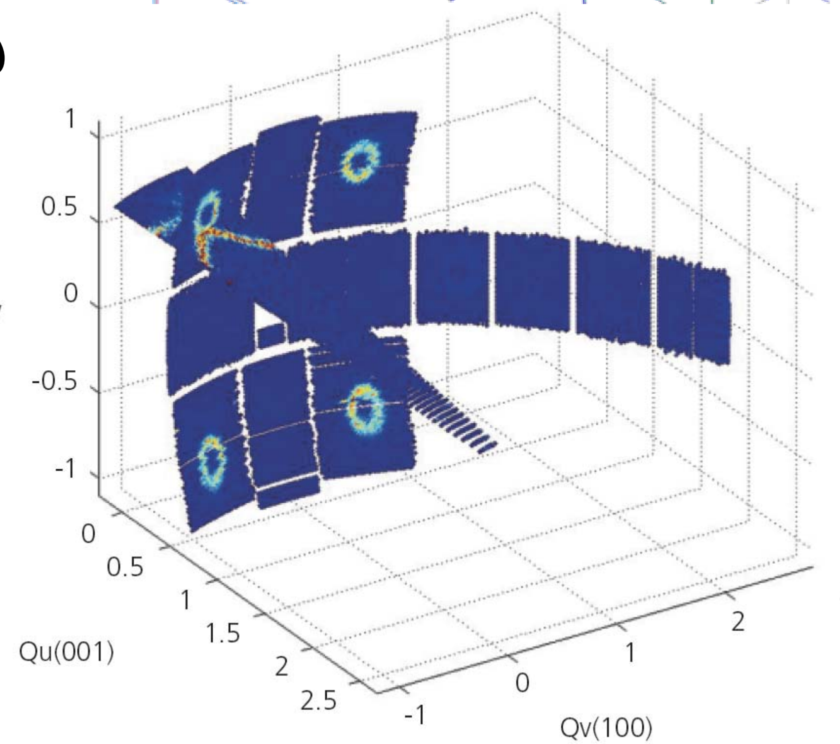
$$Q^2 = k_i^2 + k_f^2 - 2|k_i||k_f|cos2\theta$$
$$= 2E_i - \hbar\omega - 2(E_i^2 - E_i\hbar\omega)^{1/2}\cos 2\theta$$







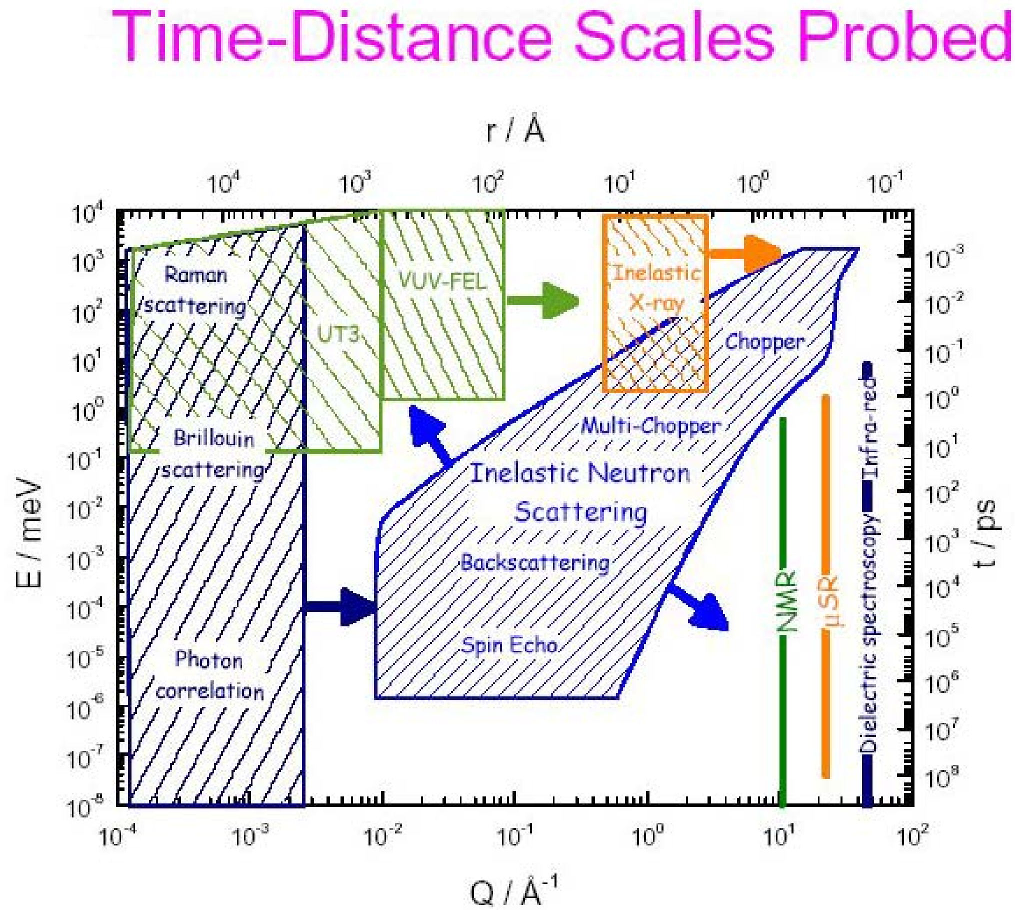




Direct TOF in practice

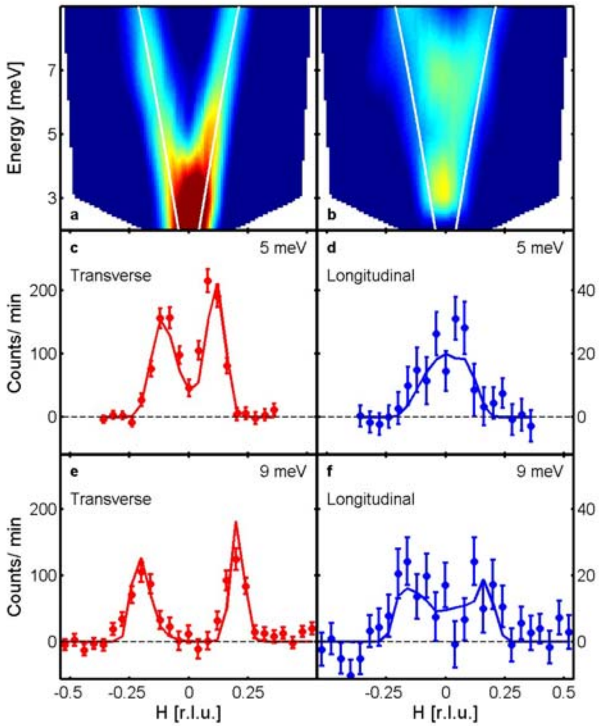
- Pulsed sources:
  MAPS, Merlin, LET…
  E_i=20meV to 2eV
- Continuous sources:
  chopper define pulse
  ΔE~μeV to meV
- Sample: 5-50g

- Counting: few days!
  - Need x10-100 for T-dependence
  - Need x100-1000 for P-dep.

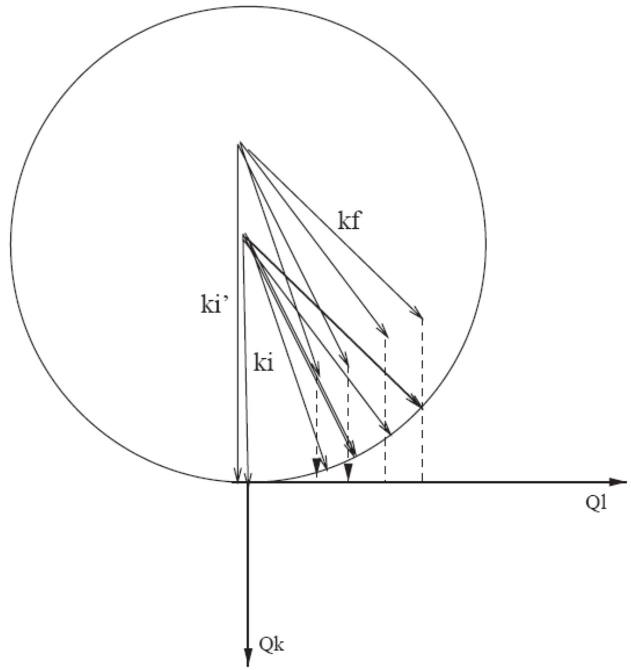


TAS v.s. TOF ⇒ Multi-TAS

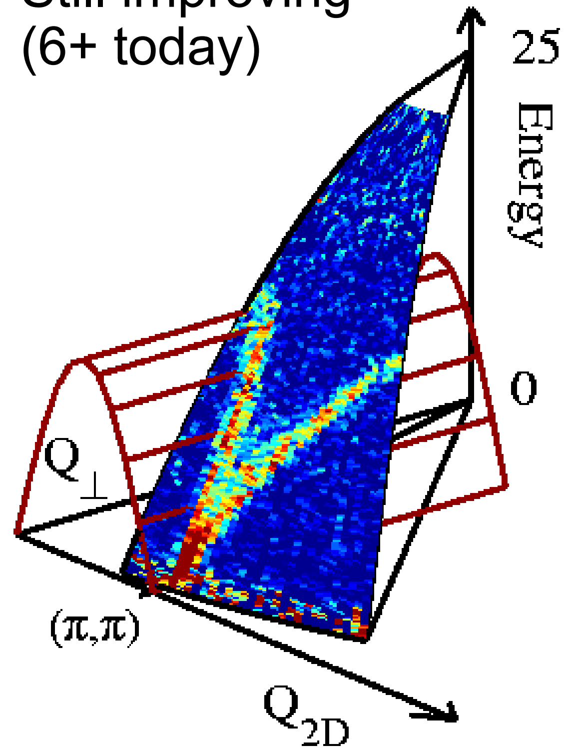
TAS
- Focus on one Point
- Flexible
- Polaristion analysis
- Already “optimal”



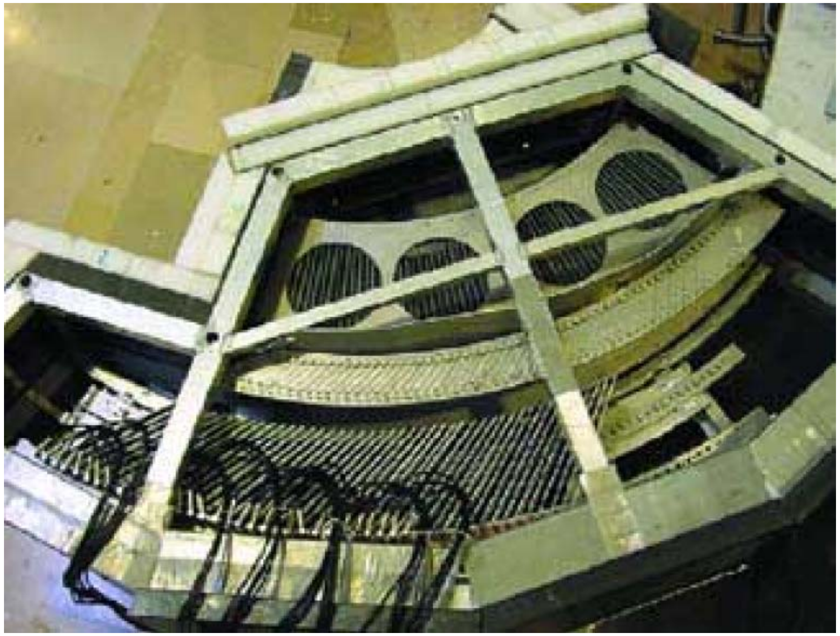
Multi-TAS
- a line in momentum-energy-space
- More Neutrons recorded than TAS
- More flexible than TOF

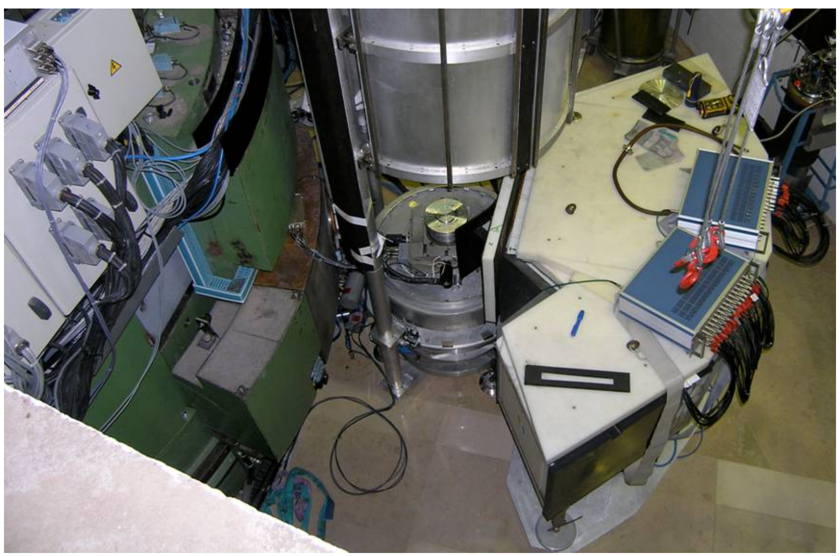


TOF
- 2-3D manifold
- Overwiew – sees “everything”
- Less flexible
- Still improving (6+ today)



The MAD box

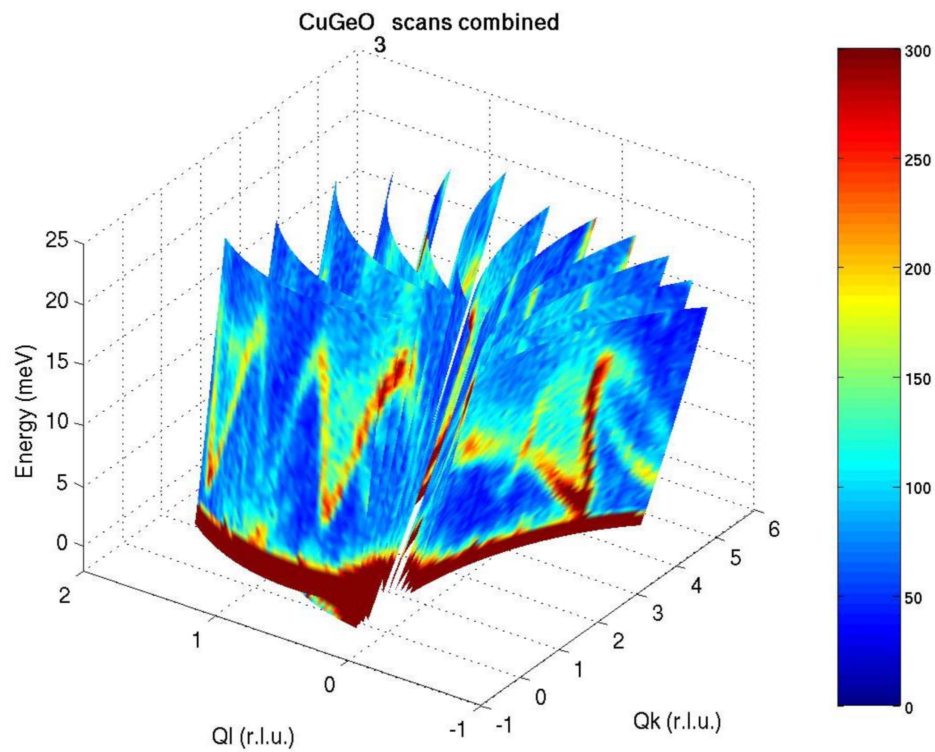




Multi (47) analyser-detector

- Analysers Cu(200) E_f=31 meV
- Detectors ^{3}He, 0.3° each 1°
- IN8, 7 samples in 1 week

(Jimenez-Ruiz, Demmel & 7 students)

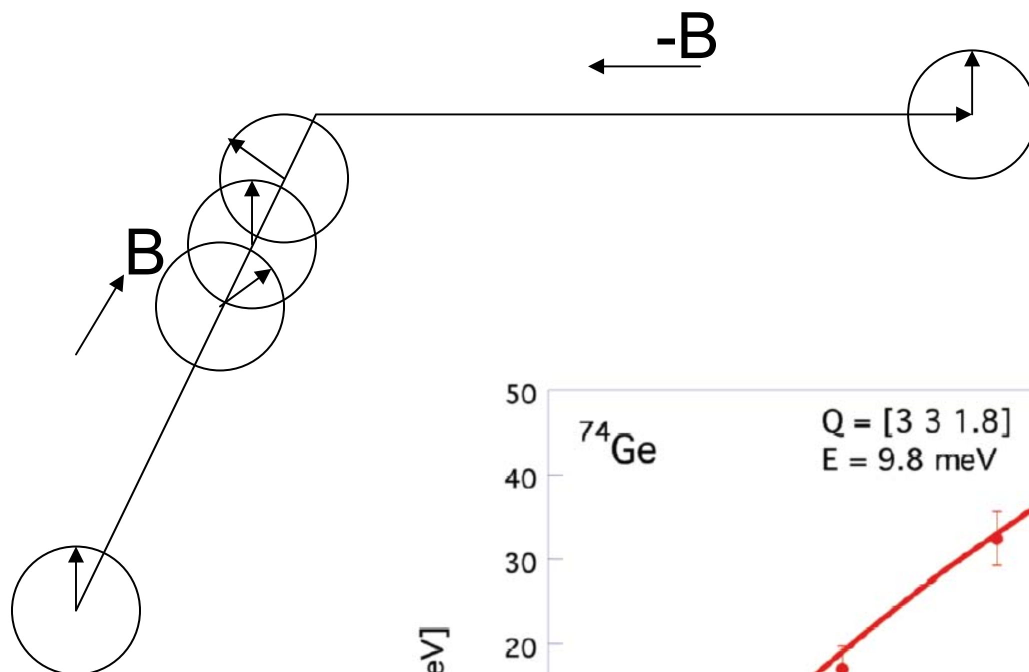


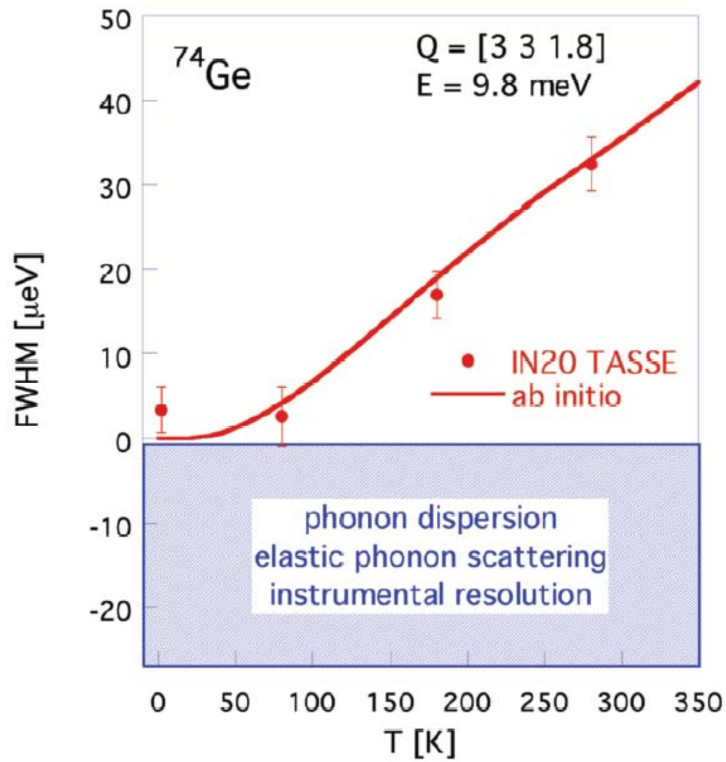
Recap.: How do we measure

- Control k_i and k_f
- Three axis spectrometers (TAS)
- Time-of-flight spectrometers (TOF)
- Energies from μeV to eV
- New sources in recent years!
- New spectrometers emerging!
- Many other neutron techniques in magnetism

Other techniques: Dynamics

- Indirect TOF
- Backscattering:
  – better resolution
- Spin-Echo:
  – ‘count’ precessions
  – combined with TAS
  – μeV resolution





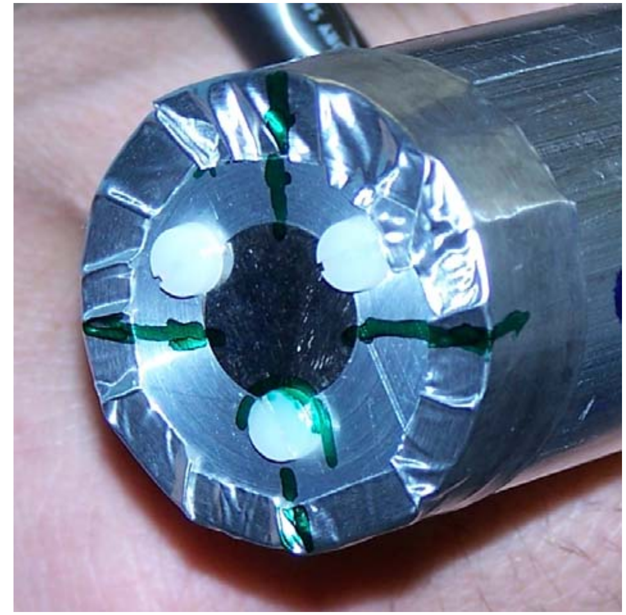
Other techniques: Structure

- Powder & single crystal diffraction
- Reflectivity (surfaces, films and multilayers)
- Small angle scattering (large objects: domains, magnetic vortices in superconductors etc.)
- …

Other techniques: Imaging

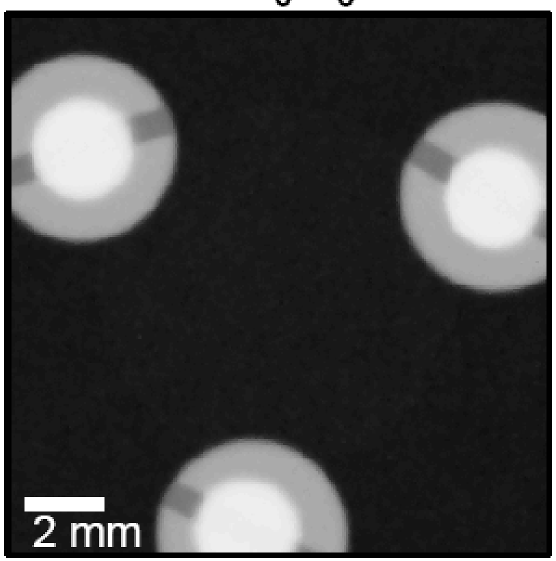
Coherent neutron phase imaging

- FM domains in $FeSi_{3\%}$ disk
- Promise of 3D domain imaging!



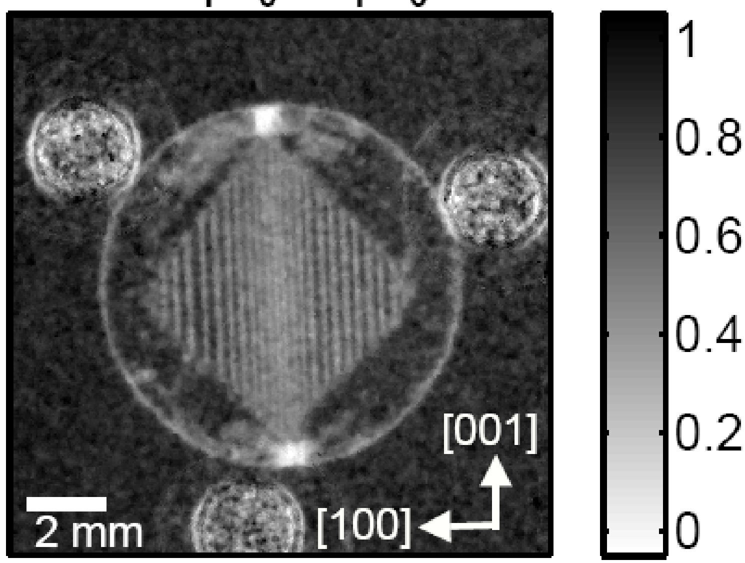
Absorption

a_0^s/a_0^r

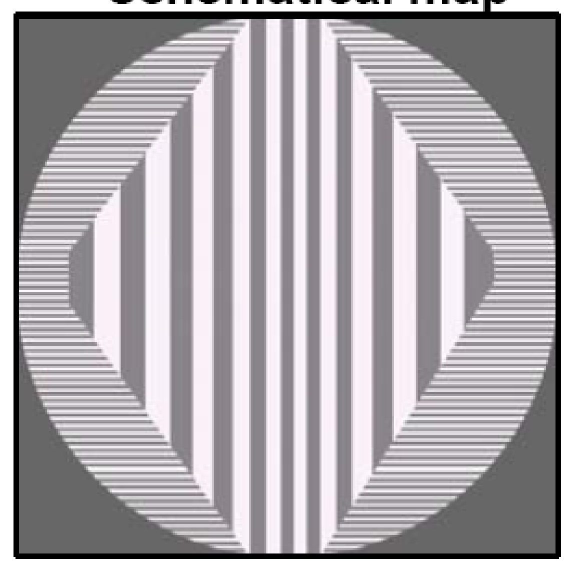


Phase contrast

$a_1^s \cdot a_0^r \,/\, a_1^r \cdot a_0^s$



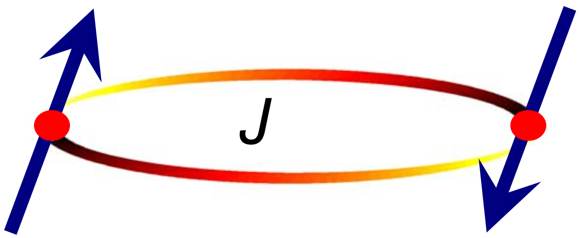
schematical map



Physics – what do we measure *examples*

- ~~Spin-waves, Holstein-Primakof transformation ...~~
- A pair of spins:

☞ $= J \sum \boldsymbol{S}_i \cdot \boldsymbol{S}_j$



Ferromagnet: $J < 0$

|GS⇧=|↑↑⇧ or |↓↓⇧

"Classical"

Antiferromagnetic: $J > 0$

triplets

E = 3/4J S_{tot}=1 |↑↑⇧, |↓↓⇧, |↑↓⇧ + |↓↑⇧

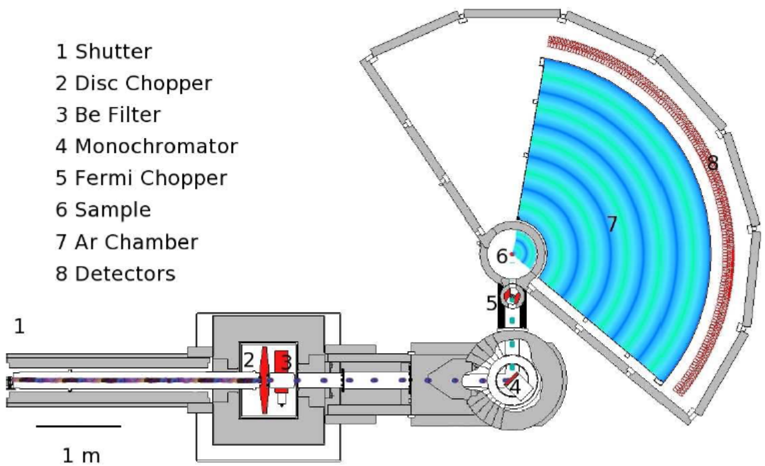
J

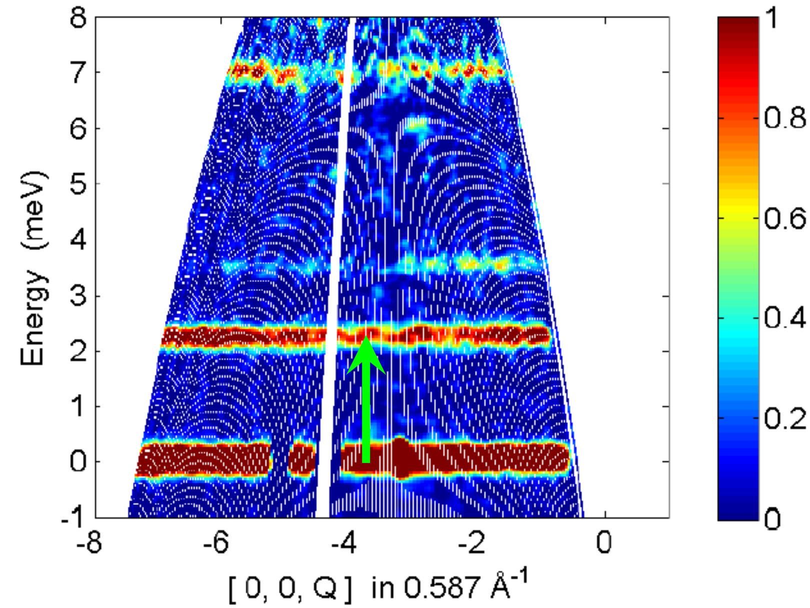
singlet

-1/4J S_{tot}=0 |↑↓⇧ - |↓↑⇧

Excitations

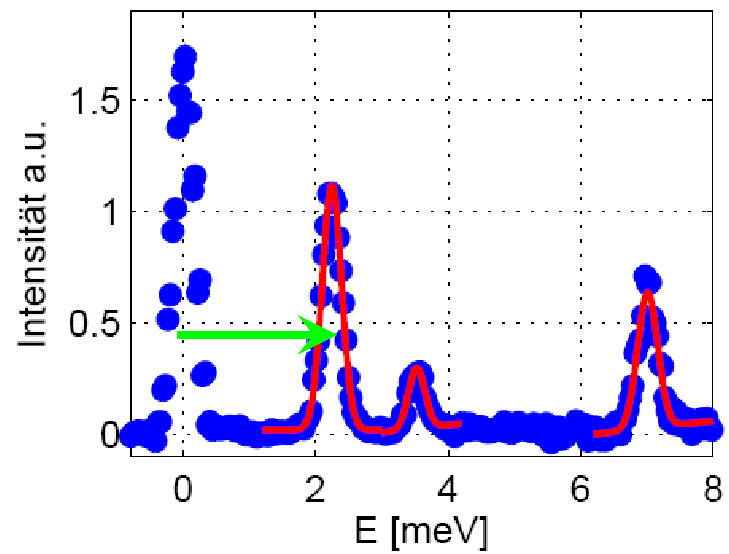
- Transition from ground to excited state





- TOF powder spectrum

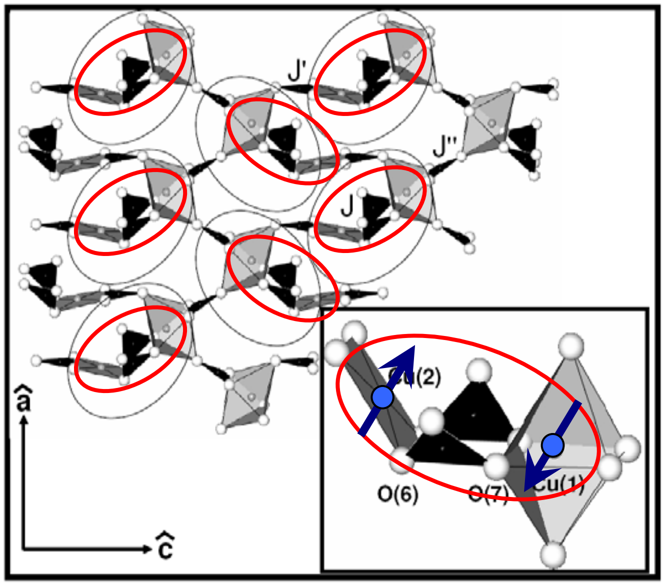
  (Crystal fields in $LiErF_4$)



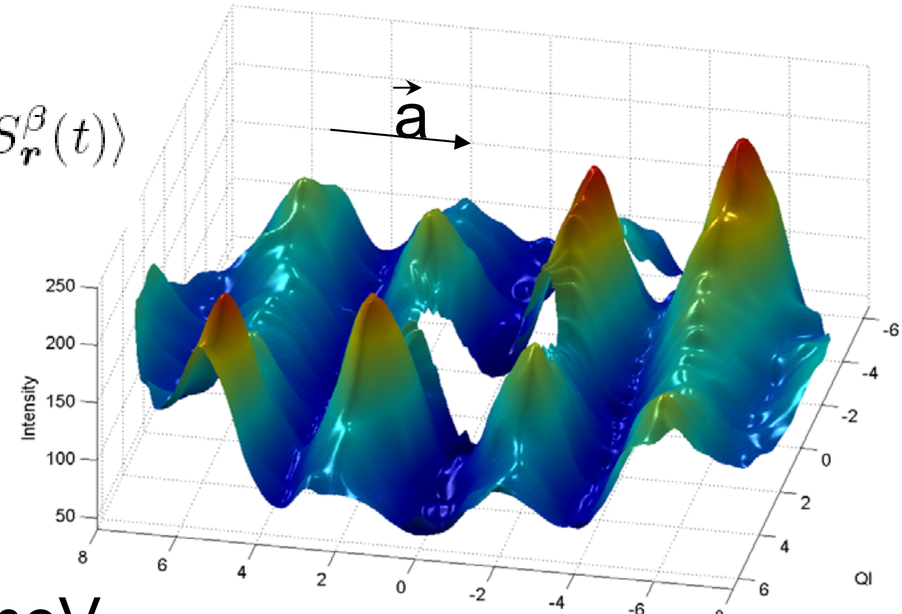
Q-dependence – Structure factor

$$S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi}\sum_{\boldsymbol{r}'\boldsymbol{r}} e^{-i\mathbf{Q}\cdot(\boldsymbol{r}'-\boldsymbol{r})}\int_{-\infty}^{\infty} dt e^{-i\omega t}\langle S^{\alpha}_{\boldsymbol{r}'} S^{\beta}_{\boldsymbol{r}}(t)\rangle$$

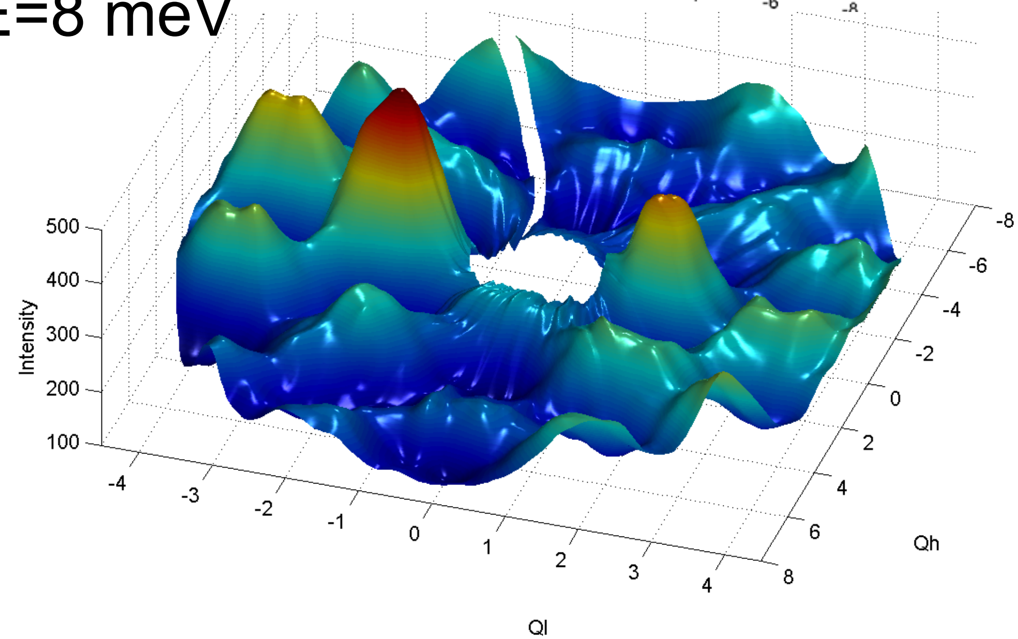
$$S(\vec{q}) = \tfrac{1}{2}(1-\cos(\vec{q}\cdot\vec{a}))$$



$Ba_2Cu(BO_3)_2$
Spectrometer IN8+MAD



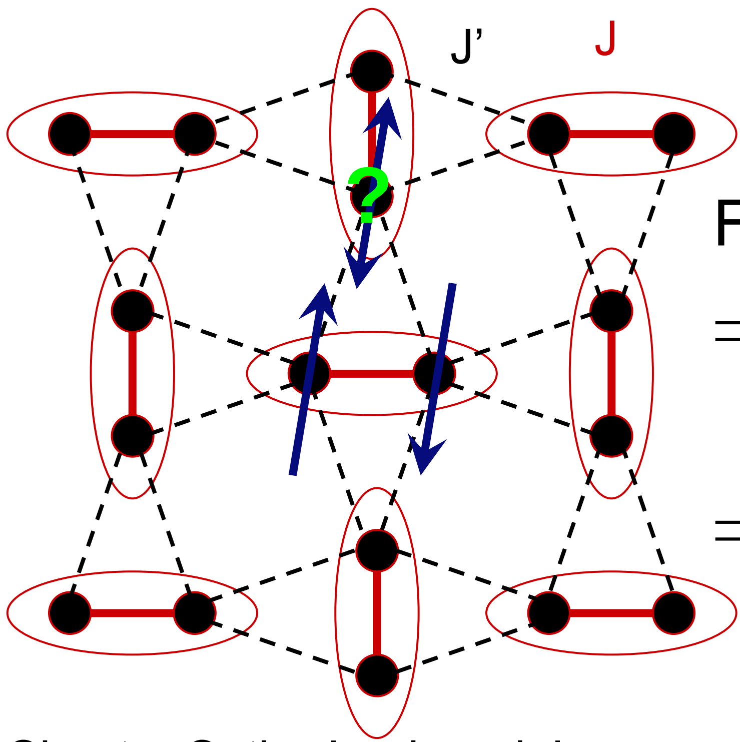
E=8 meV



Isolated localised excitations

Localised excitation ⇒ Q-indep. Energy spectrum

$SrCu_2(BO_3)_2$

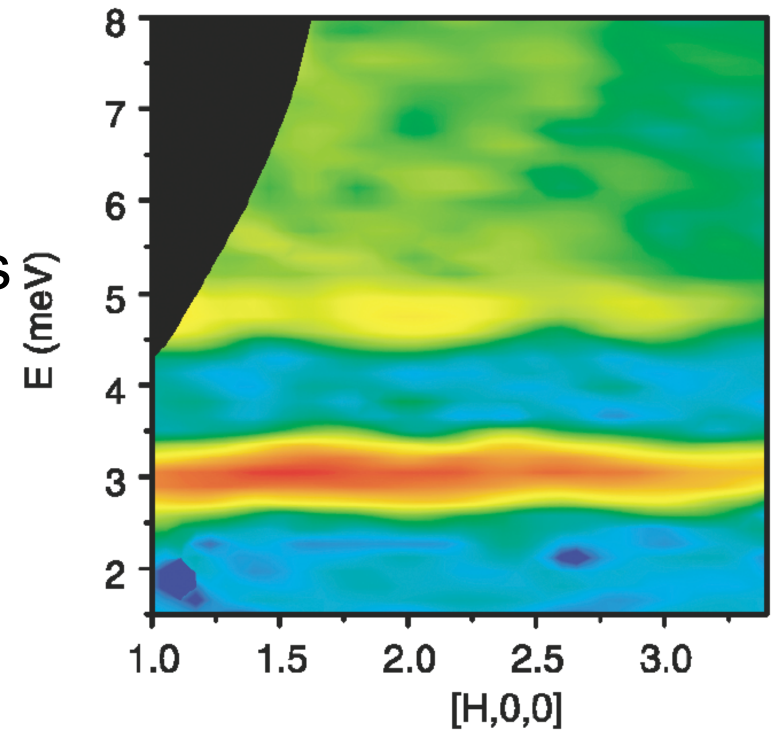


Shastry-Sutherland model

Frustration

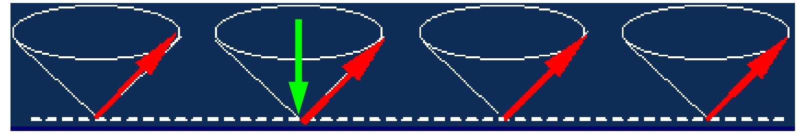
⇒ excited triplets don't move

⇒ No dispersion

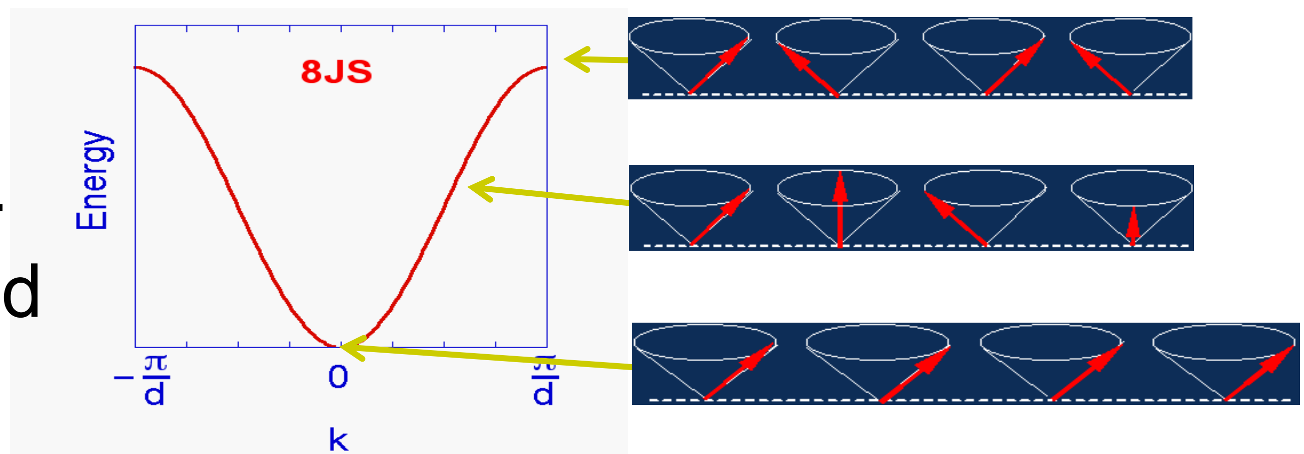


Dispersive excitations

Spin waves (ferromagnet)



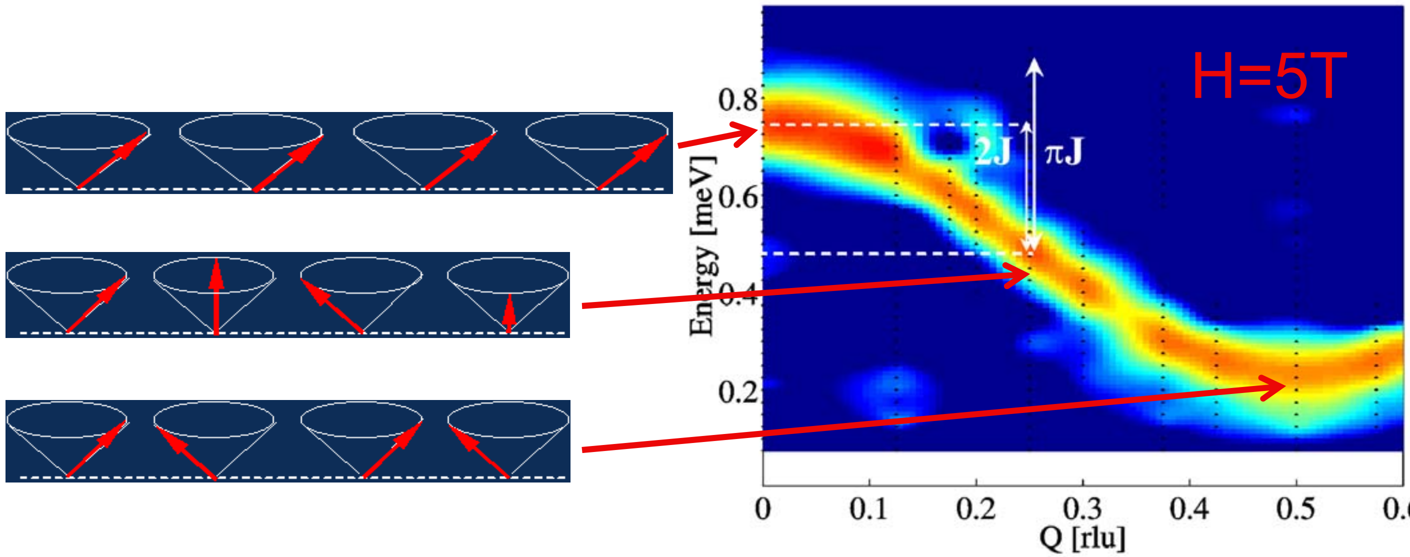
- Ordered ground state $H|g\rangle = E_g|g\rangle$
- Single spin flip not eigenstate: $S^{\pm}_r|g\rangle$
- Periodic linear combination: $|k\rangle = \Sigma_r e^{ikr} S^{\pm}_r|g\rangle$ (wave)
- Is (approximately) eigenstate: $H|k\rangle = E_k|k\rangle$
- Time evolution: $|k(t)\rangle = e^{iHt}|k\rangle = e^{iE_k t}|k\rangle$ (sliding wave)
- Dispersion:
  relation between
  time- and space-
  modulation period



Spin wave example

Cold TAS – point-by-point …

$CuSO_4 \cdot 5D_2O$



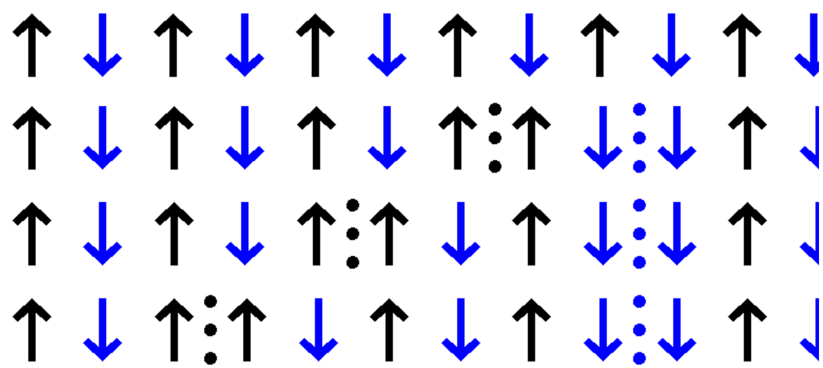
Antiferromagnet! – forced ferro by magnetic field

1-dimensional, Spin ½, antiferromagnet

⇒ quantum fluctuations and behaviour !

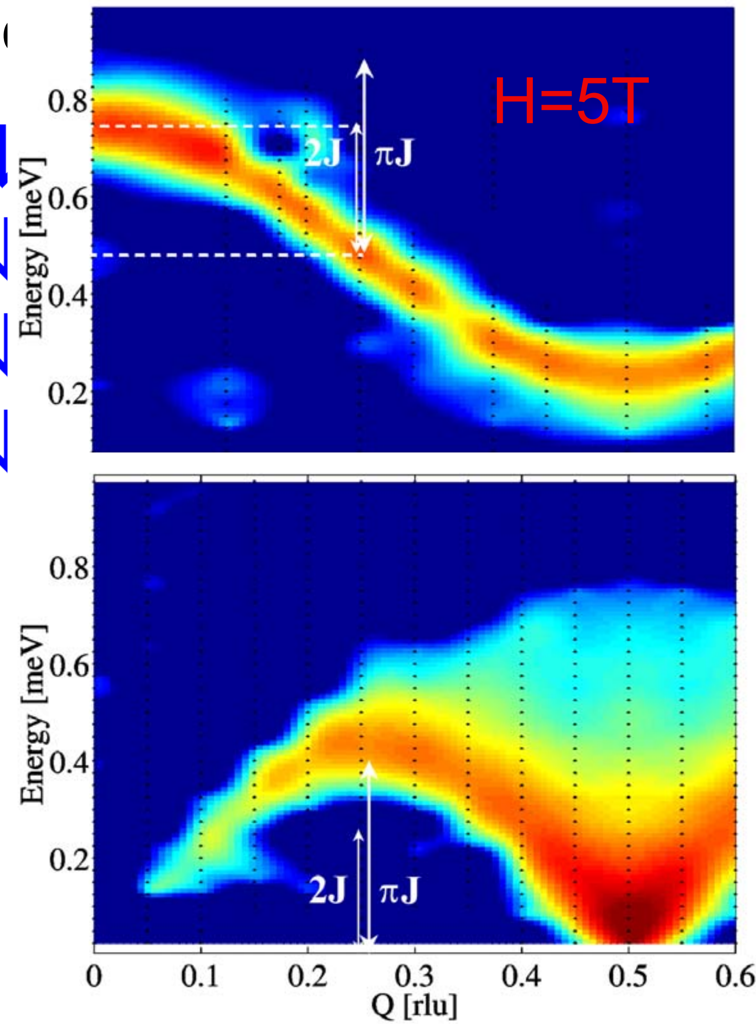
Spinon excitations

– In 1D a flipped spin ‘unbind’ to two domain walls
– “Spinons”: spin S = ½ domain walls with respect to local AF ‘order’
– Need 2 spinons to form S=1 excitation we



Energy:	$E(q)$ =	$E(k_1)$	+	$E(k_2)$
Momentum:	q =	k_1	+	k_2
Spin:	S =	½	±	½

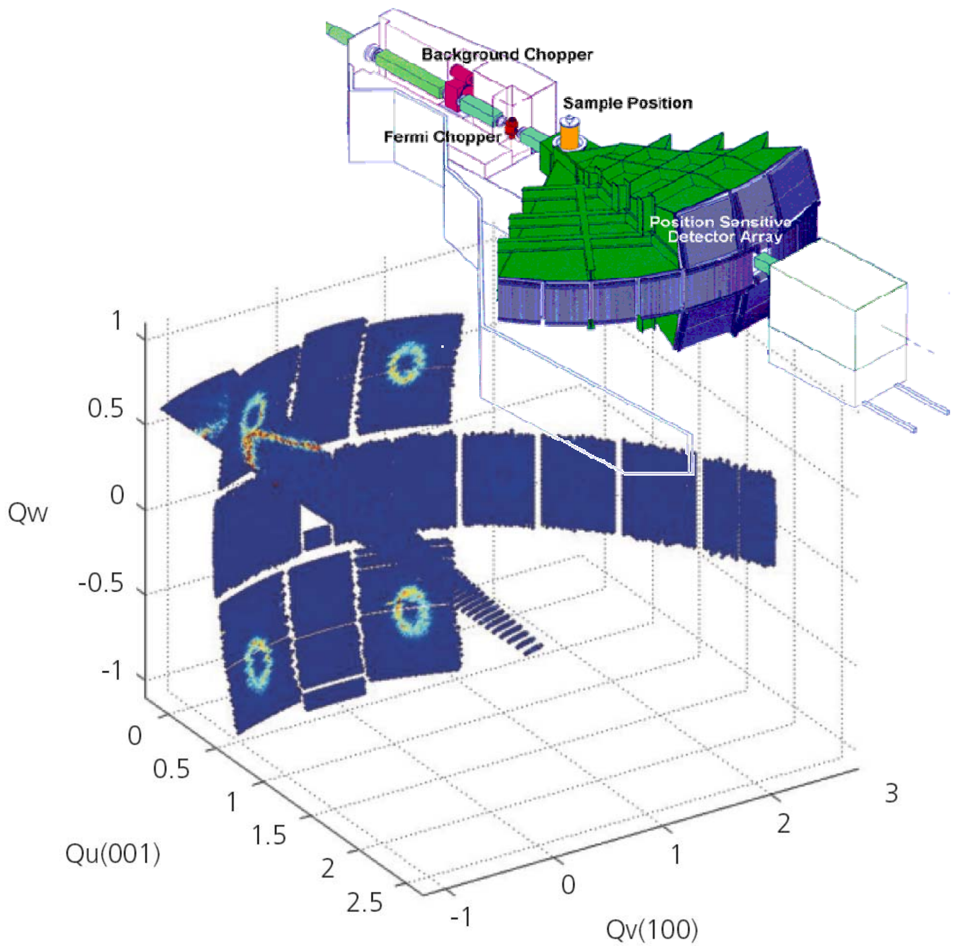
Many possibilities for each q
Continuum of scattering ⇒

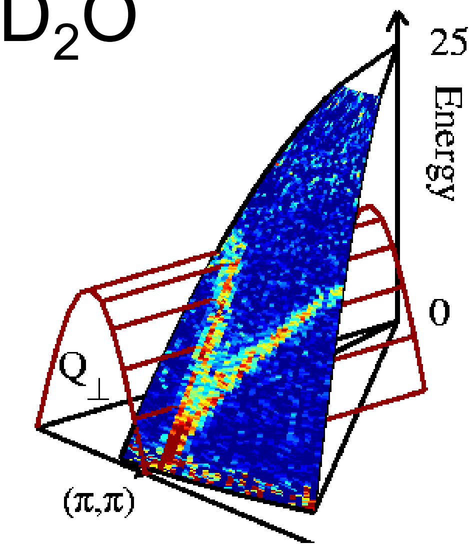


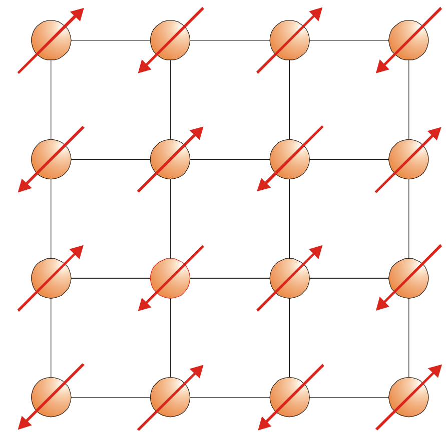
TOF example – 2D spin waves

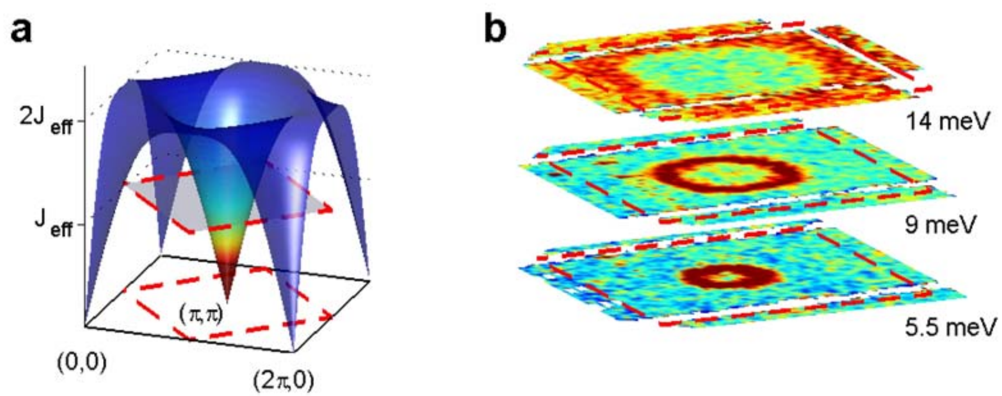
2D Heisenberg antiferromagnet – long range ordered

Spin waves in $Cu(DCOO)_2 \cdot 4D_2O$









2-spin wave scattering

Ordered moment < S > = 60%,

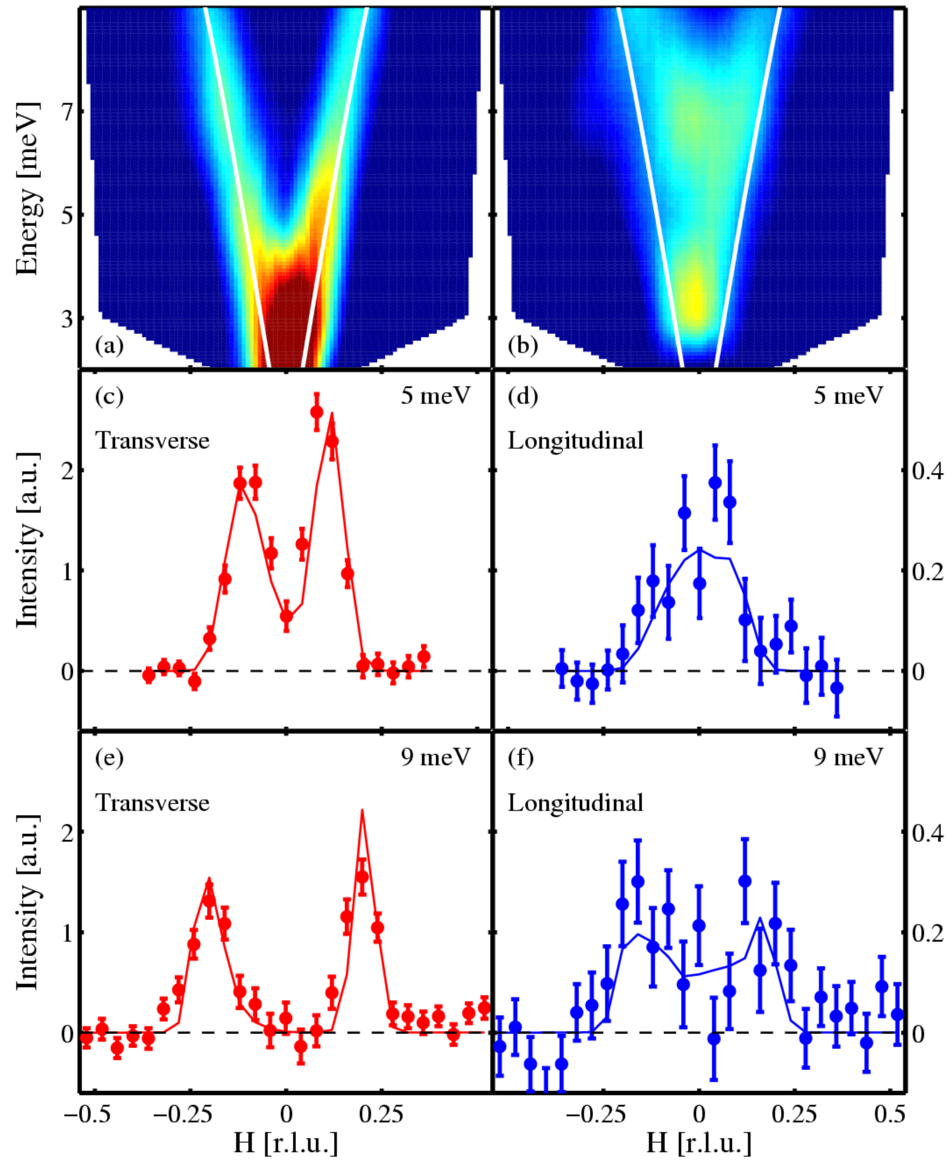
Spin wave amplitude $Z\chi$ = 51%,

Where is the remaining weight?

Polarised neutrons:

Spin-flip / non-spin-flip channels

- Separate transverse and longitudinal



$|G\rangle = |Neel\rangle + \Sigma_k a_k|1\text{-spin waves}\rangle + \ldots$

spin-wave

2-spin-waves:

$k = k_1+k_2$

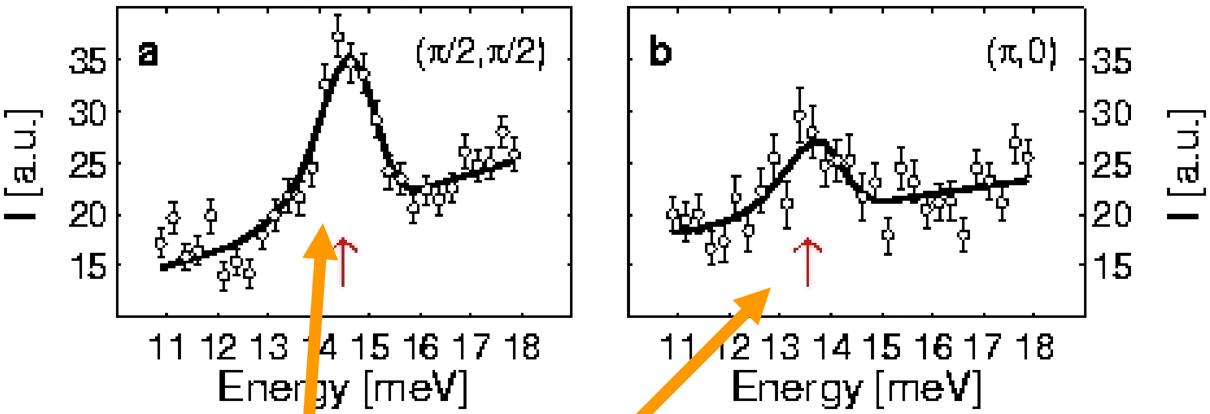
$E = E_1+E_2$

transverse

longitudinal

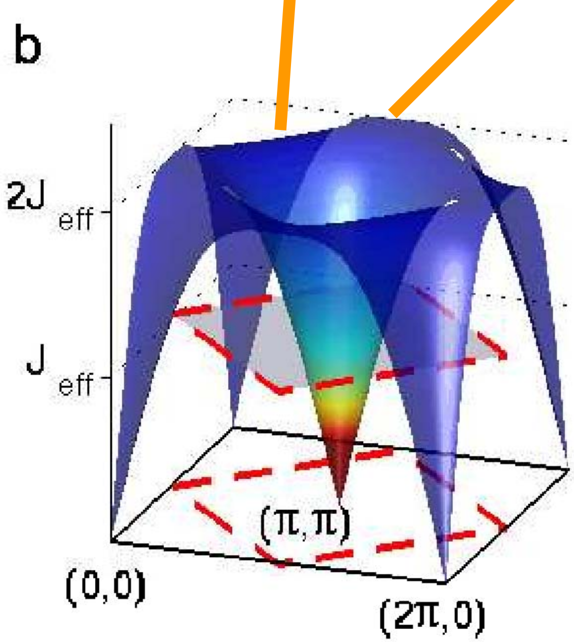
(spin-waves ok for long wavelengths)

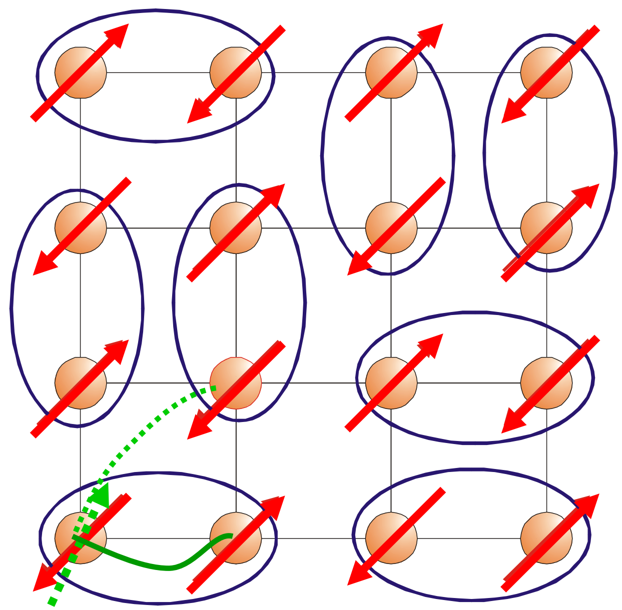
Surprise: zone boundary anomaly!



Should be same:
7% energy effect
50% intensity effect

What is nature of
✓π0n [pi0n] excitation





Hidden singlet correlations
doping: afm order dies
singlets survive ⇒ SC?

S(Q) instantaneous correlations

S(Q) is Fourier transform of 'snap-shot'

$$S(\mathbf{Q}) = \sum_{\boldsymbol{r}'\boldsymbol{r}} e^{-i\mathbf{Q}\cdot(\boldsymbol{r}'-\boldsymbol{r})} \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t) \rangle$$

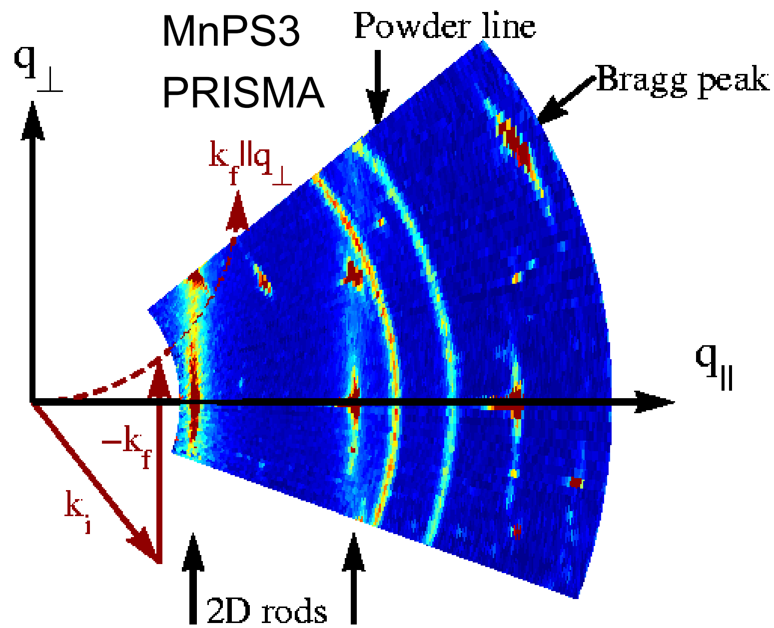
Real space exponential decay:

$$\langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t) \rangle \propto e^{-|r-r'|/\xi}$$

Reciprocal space: Lorentzian

$$S(\mathbf{Q}) \propto \frac{1}{1 + Q^2\xi^2}$$

Correlation length ξ



Wildes et al. ⇑

In 2D, S(Q) gives rods along Q ⊥

Integrate by having kf || Q⊥

S(Q) instantaneous correlations

$$S(\mathbf{Q}) \propto \frac{1}{1 + Q^2 \xi^2}$$

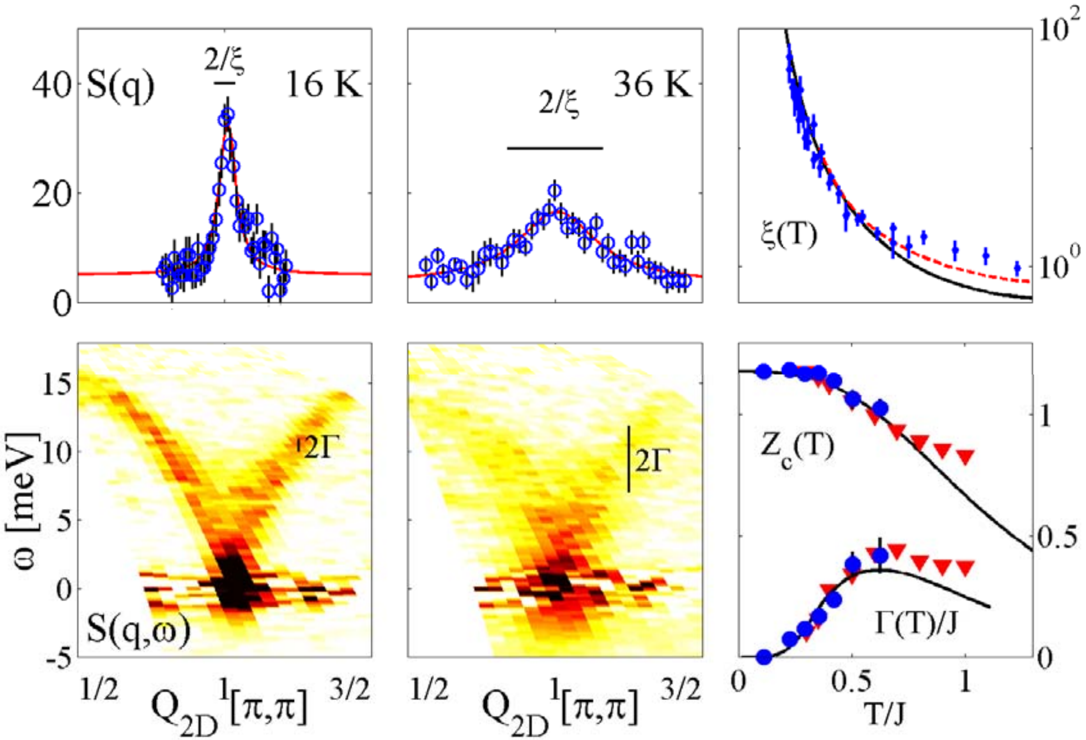
Width ⇒ Correlation length ξ

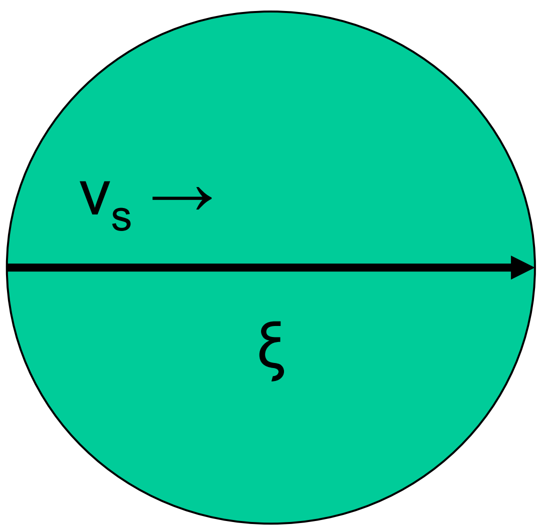
Softening

v_s ✪ Z_c

Damping

$\Gamma = v_s / \xi$



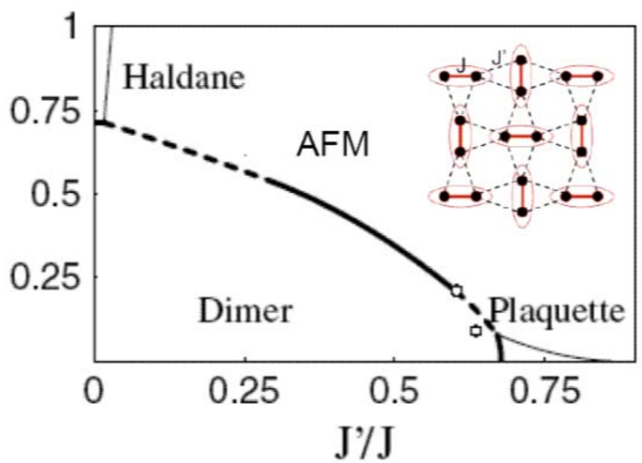


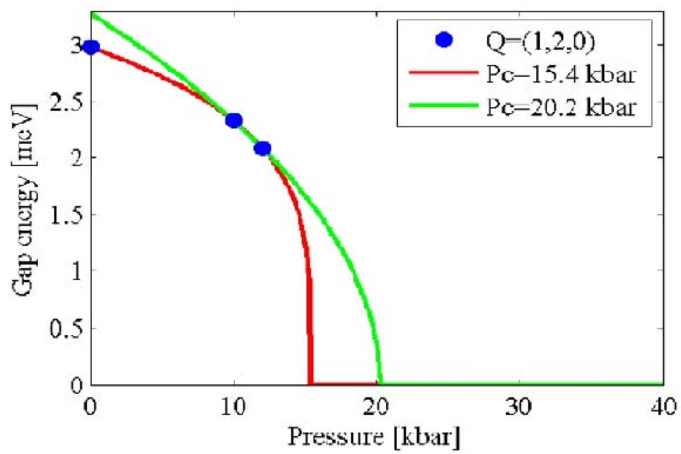
Life-time

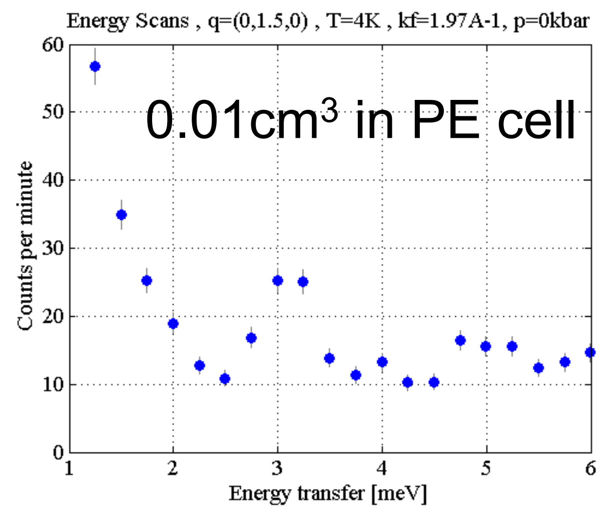
$\tau = \xi / v_s = 1/\Gamma$

Parameters

- E from micro eV to eV
- T from few mK to 1000K
- H to 15T
- P to 17kbar / 50kbar
  example: $SrCu_2(BO_3)_2$







Recap.: What do we measure

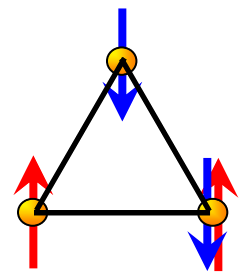
- Localised excitations

- Disperive excitations

- Dynamic correlations

- Polarisation analysis
- Dependencies:
  Temperature, Magnetic field, Pressure field

- It’s a battle: samples, statistics, resolution

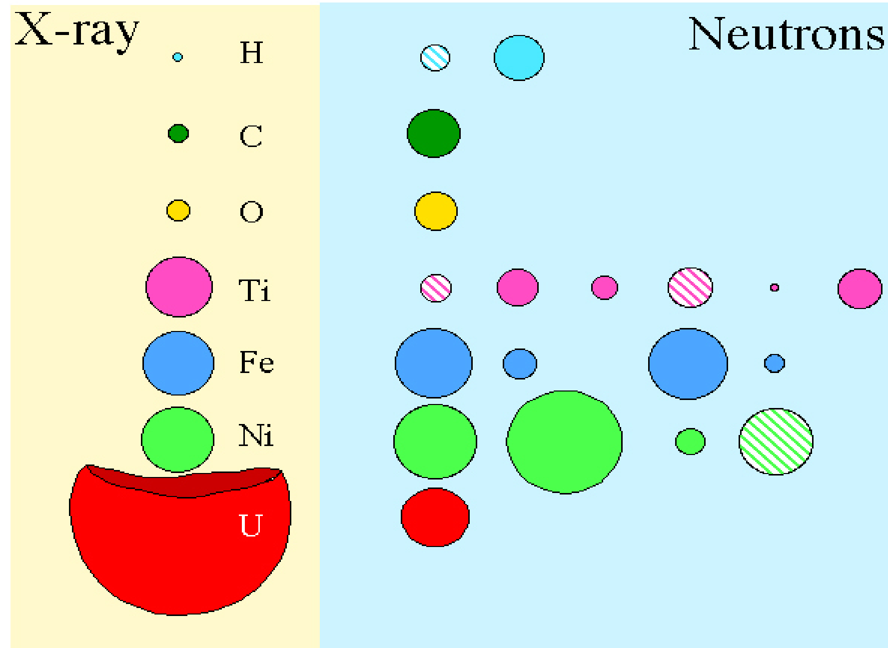


- But often *‘the final kill’* for problems in magnetism

Thank you

Neutron and X-ray scattering

- Similar in technique, but
  - X-rays much more intense
  - Different scattering lengths:



  - Neutrons simpler for magnetism
  - Neutrons better for energy resolution

- Mostly, only one will work for a given problem

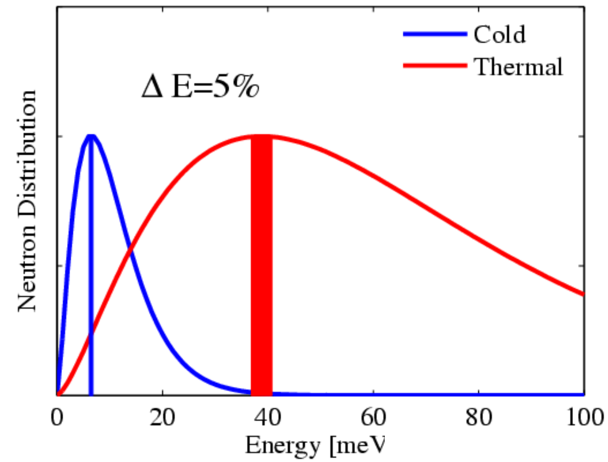
TAS resolution

- Source spectrum:
  - Cold: E_i=2-15 meV
  - Thermal E_i=15-100 meV
  - Epithermal E_i=> 1-2 eV
- Intensity / resolution:

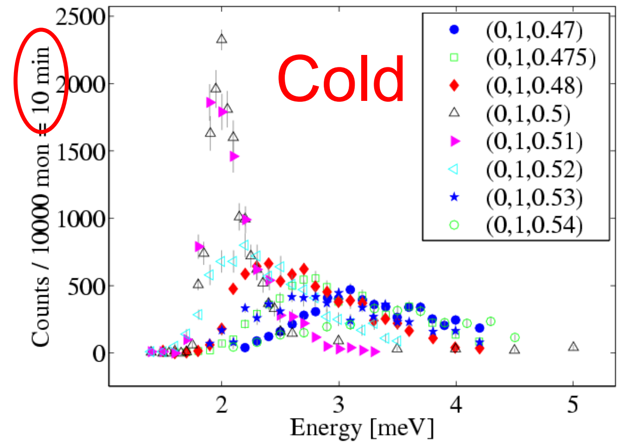
  Some typical numbers (PG002):

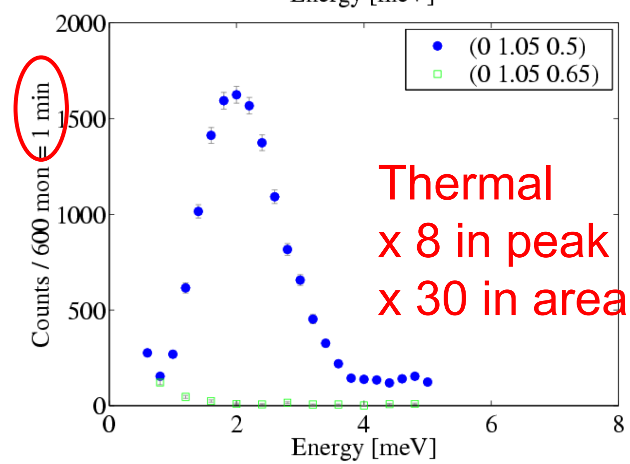
E_i,E_f	resolution (fwhm)
2 meV	30 μeV
5 meV	0.15 meV
14.7 meV	1.2 meV

- Typical counting ~1-10 min/pt



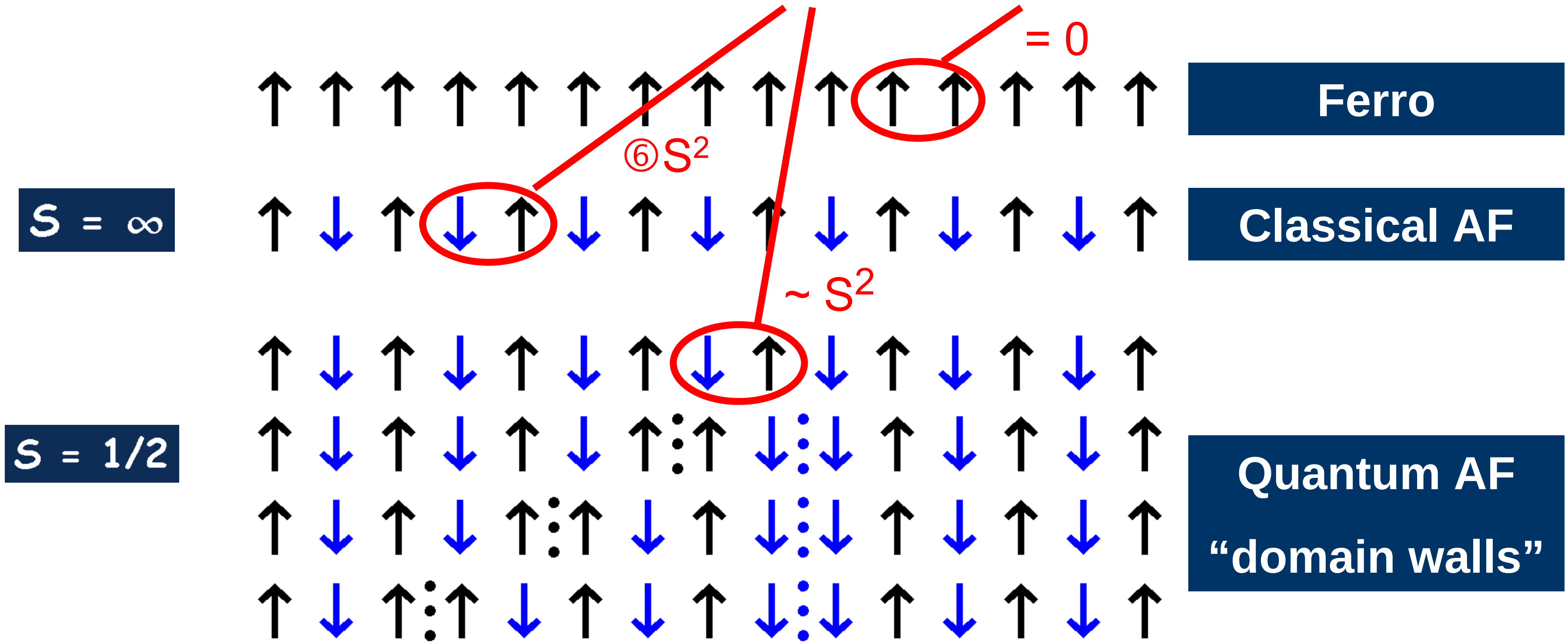
CuGeO3 IN12 2001/7





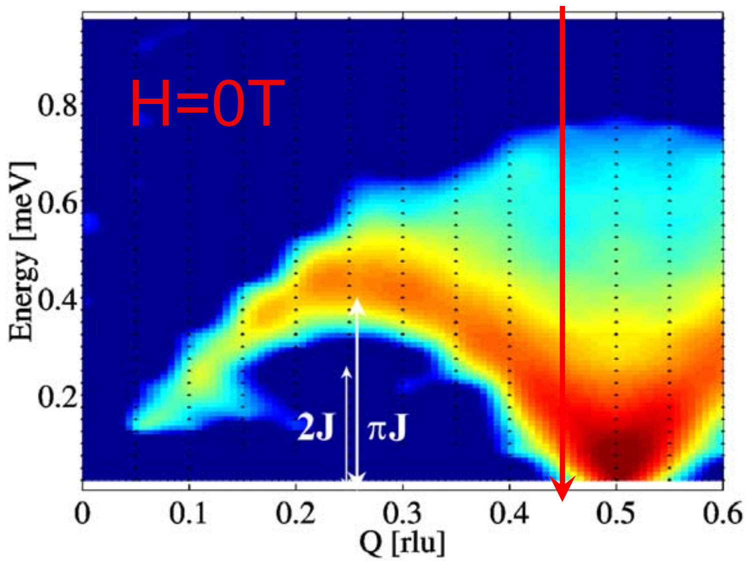
The spin ½ chain

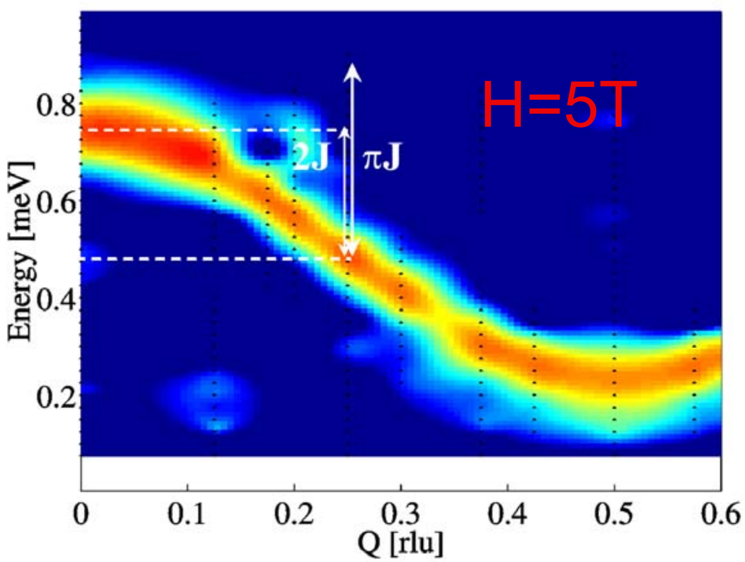
$$\mathcal{H} = J \sum S^z_n S^z_{n+1} + \frac{1}{2}\left(S^+_n S^-_{n+1} + S^-_n S^+_{n+1}\right)$$

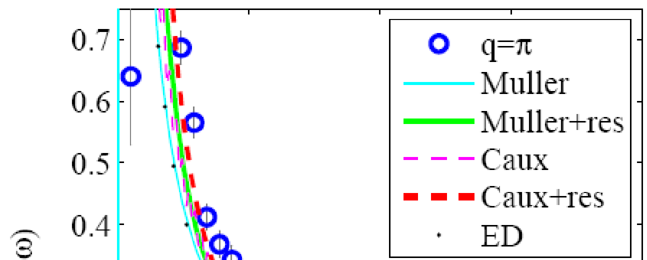


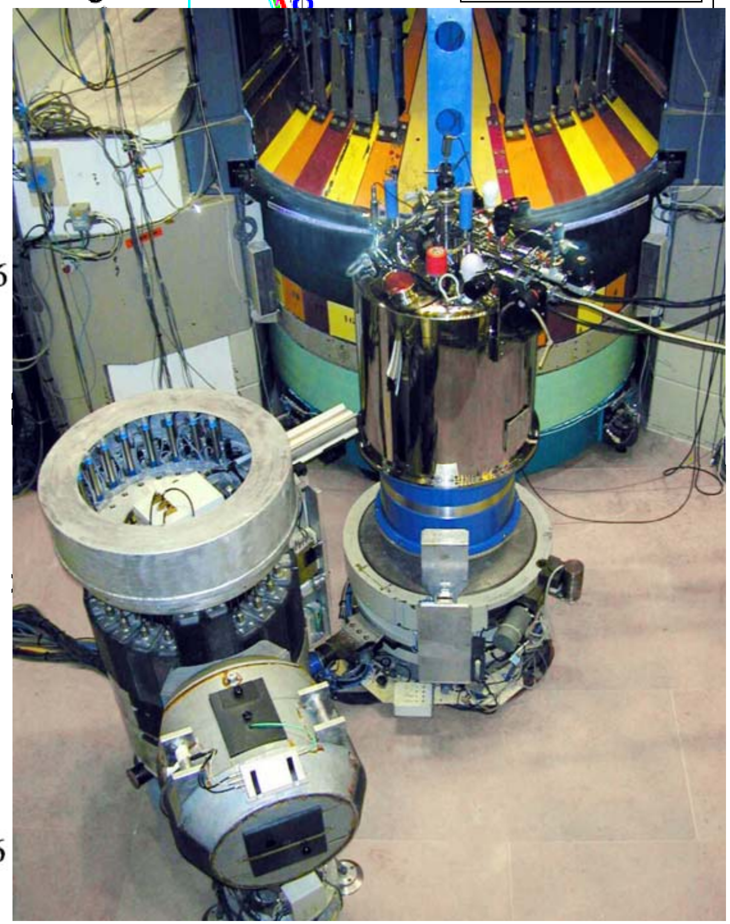
Ground state (Bethe 1931) disordered by quantum fluctuations

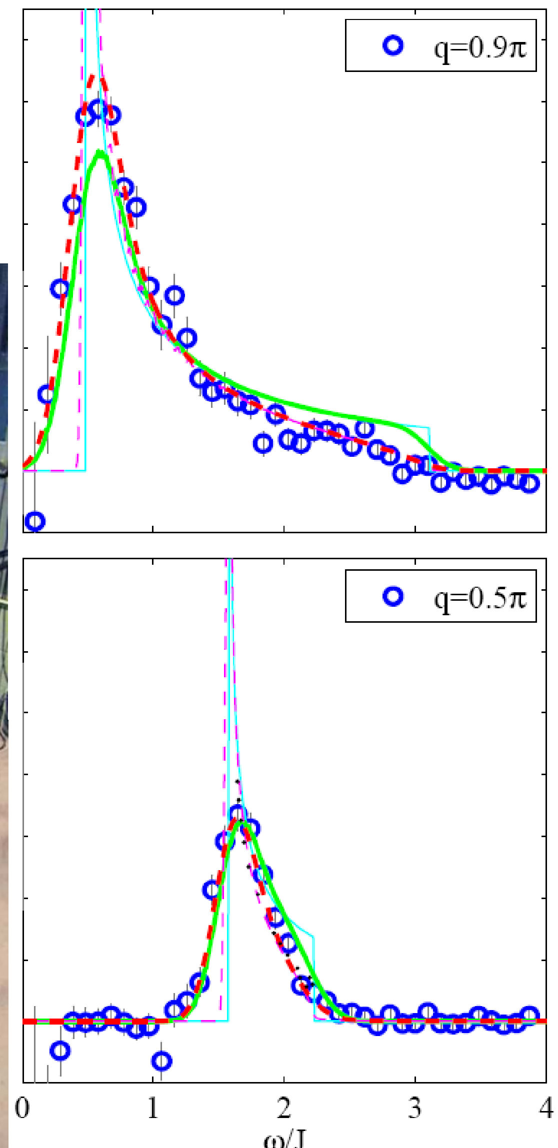
Continuum in 1D











Cold TAS: IN14

2-spin wave scattering

Ordered moment < S > = 60%,
Spin wave amplitude $Z\chi$ = 51%,
Extra 25% dip at $(\pi,0)$
Where is the remaining weight?

Ground state:

$|0\rangle = |\text{Neel}\rangle + \Sigma_k a_k |\text{spin wave with momentum } k\rangle + \ldots$

spin-wave
transverse

2 spin-waves: $k_1+k_2=k$ and $E_1+E_2=E$
longitudinal