Neutron spectroscopy: the triple-axis spectrometer

G. H. Lander

Oxford Summer School 2011

Neutron inelastic scattering

Two equations are: : $\vec{Q}=\vec{k}_i-\vec{k}_f$ $\hbar\omega=\Delta E=\hbar/(2m_n)\left(k_i^{\,2}-k_f^{\,2}\right)$

Elastic scattering has $k_i = k_f$

In terms of space-time correlation functions *elastic* scattering measures at t = 0 the position of particles j and j′

$$S(\vec{Q},0)\propto\sum_{jj'}\left\langle\exp[-i\vec{Q}\bullet\vec{R}_j(0)]\,x\,\exp[+i\vec{Q}\bullet\vec{R}_{j'}(0)]\right\rangle$$

We will now extend this to measure the correlation of particle j at t = 0 with particle j′ at t = t

$$S(\vec{Q},\omega)\propto\int_{-\infty}^{+\infty}\exp(-i\omega t)dt\sum_{jj'}\left\langle\exp[-i\vec{Q}\bullet\vec{R}_j(0)]\,x\,\exp[+i\vec{Q}\bullet\vec{R}_{j'}(t)]\right\rangle$$

In a crystal there is a periodic arrangement of atoms; we write this as $\vec{R}_l = \vec{l} + \vec{u}_l$ where $\boldsymbol{u_l}$ is the displacement of the atoms from their mean position $\boldsymbol{l}$

The correlations between particles l and l' depends only on their vector difference $|\boldsymbol{l - l'}|$ so the sum over j and j' is the same

$$\sum_{jj'} \left\langle \exp[-i\vec{Q} \bullet \vec{R}_j(0)] \, x \, \exp[+i\vec{Q} \bullet \vec{R}_{j'}(t)] \right\rangle$$

$$= N \sum_j \exp(i\vec{Q} \bullet \vec{l}_j) \left\langle \exp[-\vec{Q} \bullet \vec{u}_o(0)] \, x \, \exp[+\vec{Q} \bullet \vec{u}_j(t)] \right\rangle$$

leading to:

$$S(\vec{Q}, \omega) \propto \sum_j \exp(i\vec{Q} \bullet \vec{l}_j) \int_{-\infty}^{+\infty} \exp(-i\omega t) dt \; x \left\langle e^U e^V \right\rangle$$

where $\sum_j \exp(i\vec{Q} \bullet \vec{l}_j)$ is the ***elastic structure factor.***

$\left\langle e^U e^V \right\rangle$ where $U = -i\vec{Q} \bullet \vec{u}_o(0)\ and\ V = +i\vec{Q} \bullet \vec{u}_j(t)$

We assume the displacements are *small* and can be described in terms of an harmonic oscillator. Then we can use the identity

$\left\langle e^U e^V \right\rangle = e^{\left\langle U^2 \right\rangle} e^{\langle UV \rangle}$ and note that $e^{\left\langle U^2 \right\rangle}$ is independent of **t**

This term is the Debye-Waller factor representing uncorrelated motions of the nuclei from their mean positions $e^{\left\langle U^2 \right\rangle} = \left\langle \left[\vec{Q} \bullet \vec{u}_o(0) \right]^2 \right\rangle = e^{-2W}$

Finally $e^{\langle UV \rangle} = 1 + \langle UV \rangle + \frac{1}{2}\langle UV \rangle^2 + ...$

One phonon **two phonon**

Final phonon cross section

$$\left(\frac{d^2\sigma}{d\Omega d\omega}\right) = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{(2\pi)^3}{v_0}\frac{1}{2M}\exp(-2W)$$

$$x\sum_s\sum_\tau \frac{\left(\vec{Q}\bullet\hat{u}_s\right)^2}{\omega_s}\left(n_s+\frac{1}{2}\pm\frac{1}{2}\right)\delta(\omega\mp\omega_s)\,\delta(\vec{Q}\mp\vec{q}-\tau)$$

Intensity $\propto |Q|^2$ $\left(\vec{Q}\bullet\hat{u}_s\right)^2$ **Selects phonons** $\|\vec{Q}$

$\propto 1/\omega$

$\propto \langle n_s + 1/2 \pm 1/2\rangle$ gives the population of phonon states.

There are more phonons at high T as phonons and magnons are bosons, no limit to population

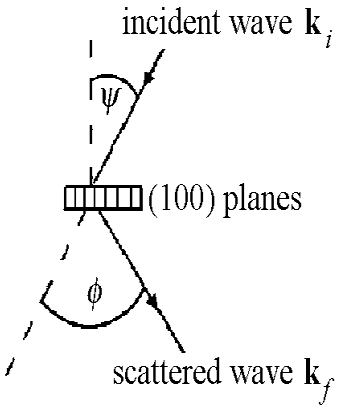
$\propto \delta(\omega \mp \omega_s)$ gives Stokes and anti-Stokes lines

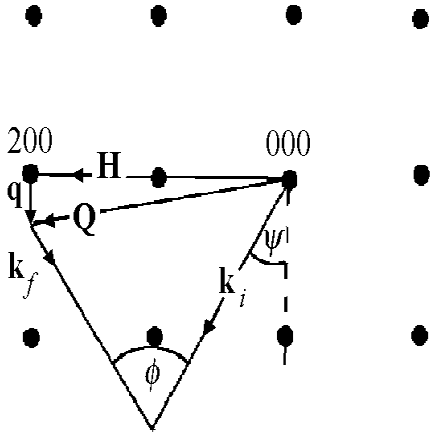
$\propto \delta(\vec{Q}\mp\vec{q}-\tau)$ gives conservation of momentum for phonon of wavevector ***q***; note

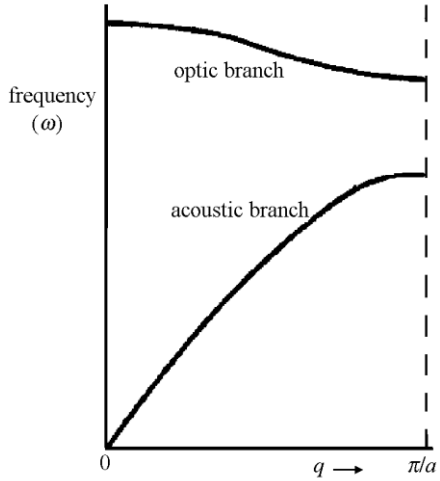
$$|\vec{q}| = 2\pi/\lambda_{phonon}$$

Schematic of measurement of inelastic features

Note **H** ≡ τ in last equations







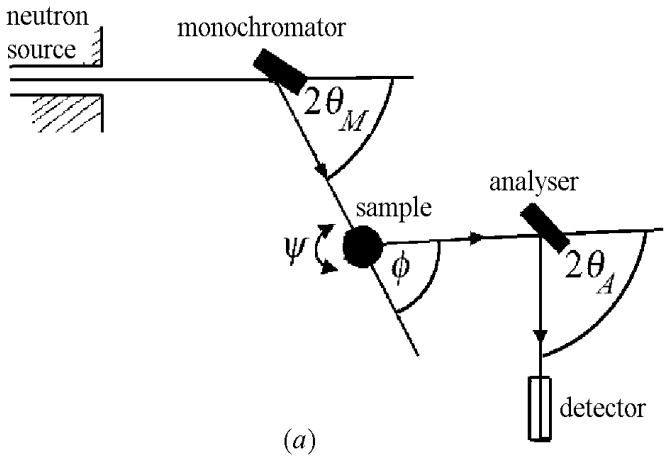
Real space **reciprocal space** **E vs. *q***

$$|\mathbf{Q}|^2 = |\boldsymbol{k_i}|^2 + |\boldsymbol{k_f}|^2 - 2\,|\boldsymbol{k_i}||\boldsymbol{k_f}|\cos(2\theta)$$

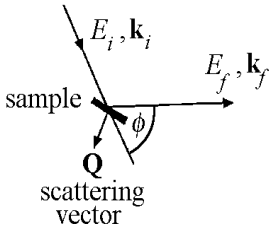
$$\boldsymbol{Q = k_i - k_f}$$

$$\hbar\omega = E_i - E_f = \frac{\hbar^2}{2m_n}\left(k_i^2 - k_f^2\right)$$

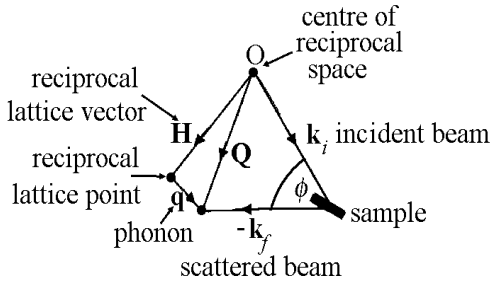
Schematic of triple-axis spectrometer



(*a*)



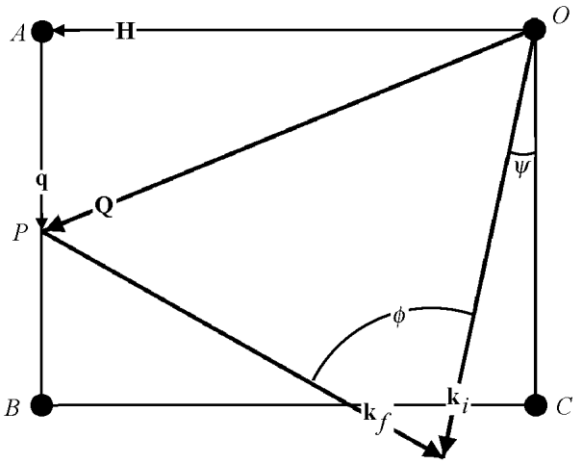
real space



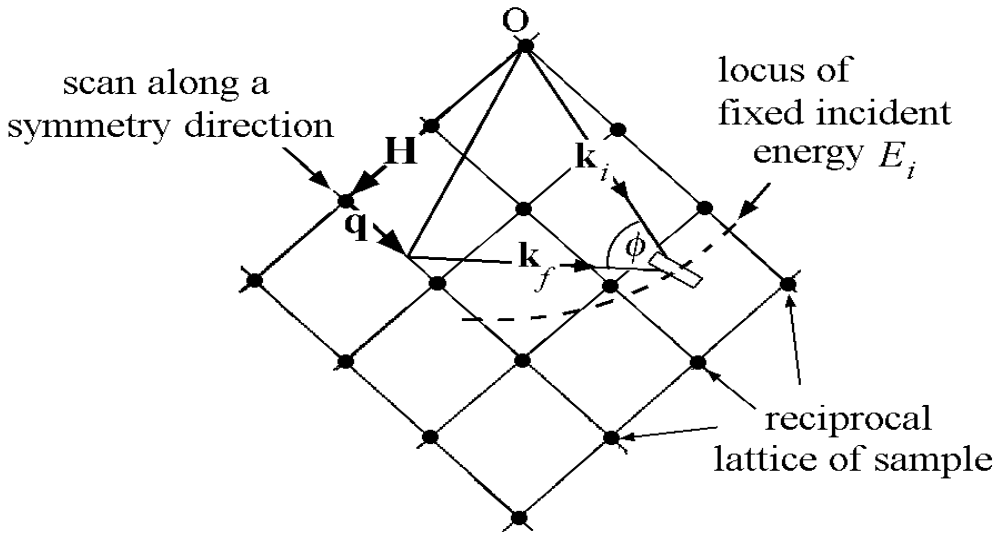
reciprocal space

(*b*)

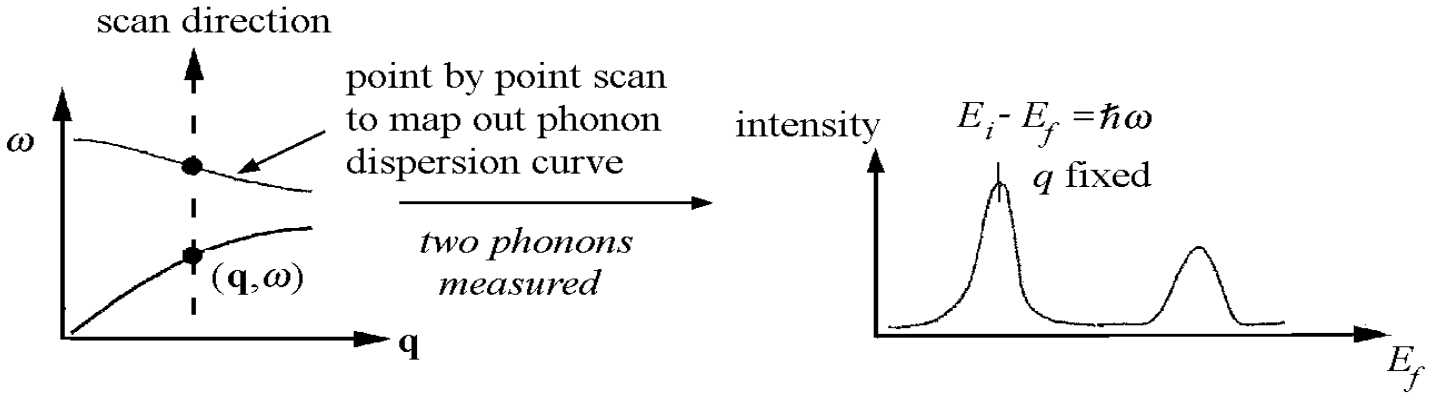
Can be operated in constant-***Q*** or constant E mode.



Techniques of the triple-axis spectroscopy

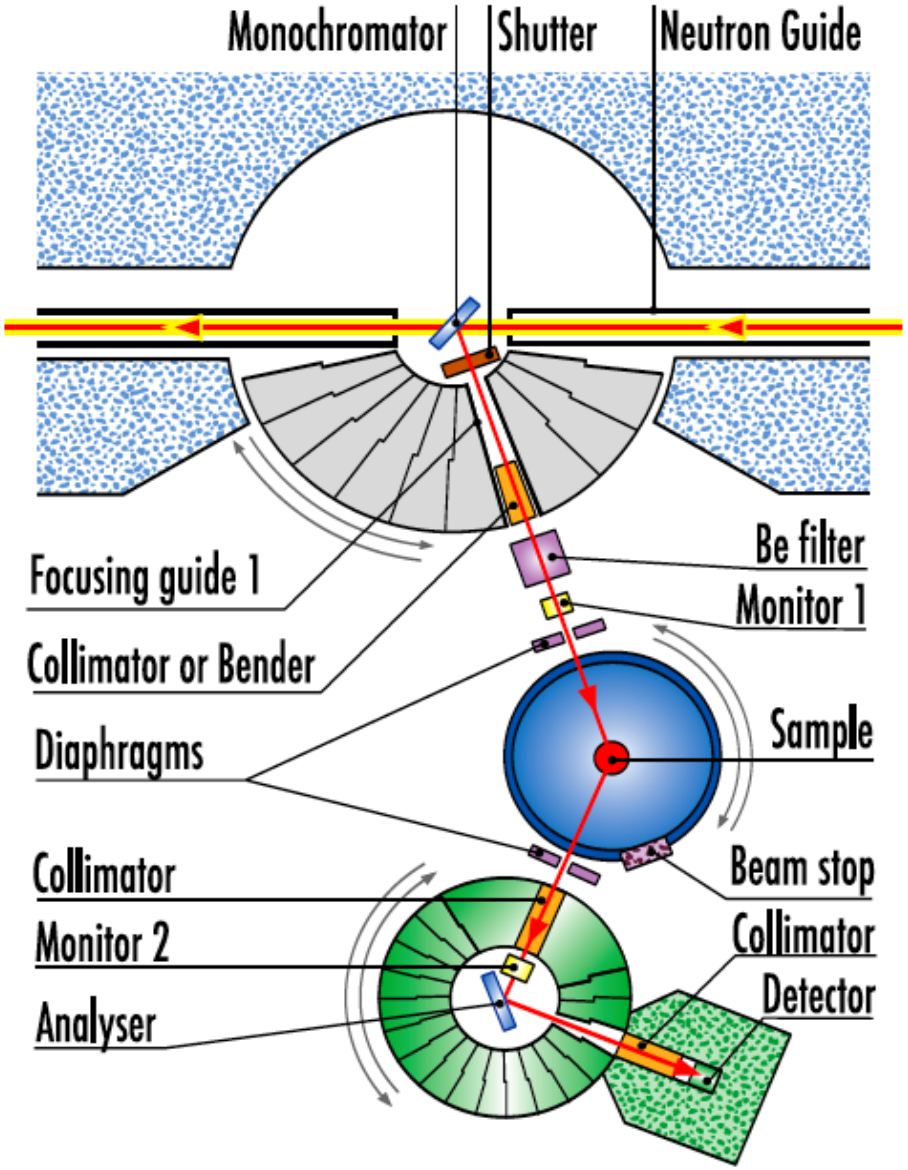


Focusing



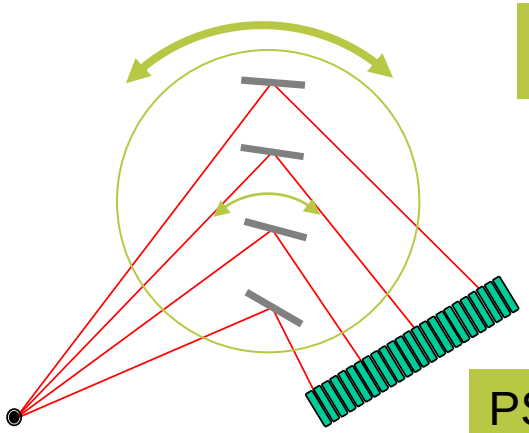
Schematic of triple-axis spectrometer

Note that the Be filter, which is used with cold neutrons, can either be before the sample (constant k_i) or after the sample (constant k_f)



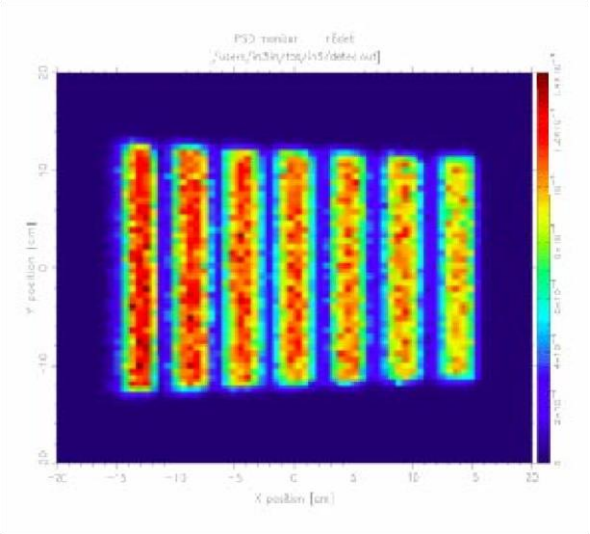
The IN14 triple-axis spectrometer at the ILL

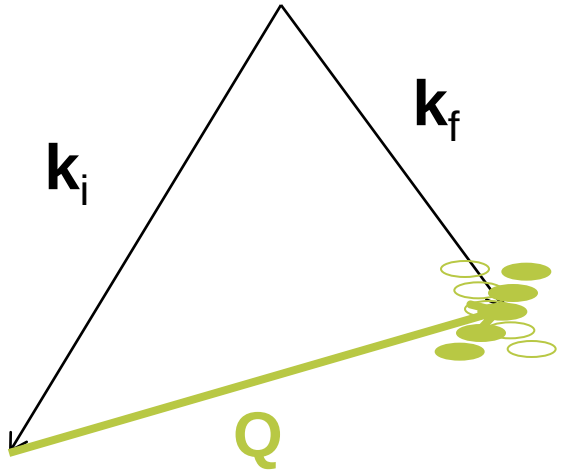
Multiplexed analyzer II



***RITA* - type**

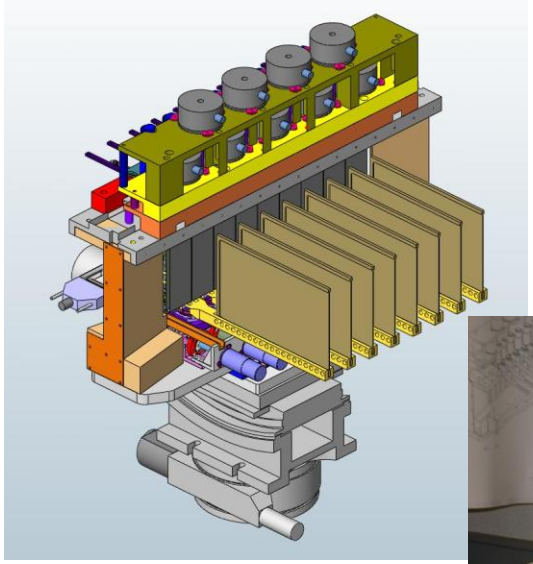
PSD





- freedom in defining foot-print in ***Q***,ω
- "imaging" details of S(***Q***,ω)
- needs expertise

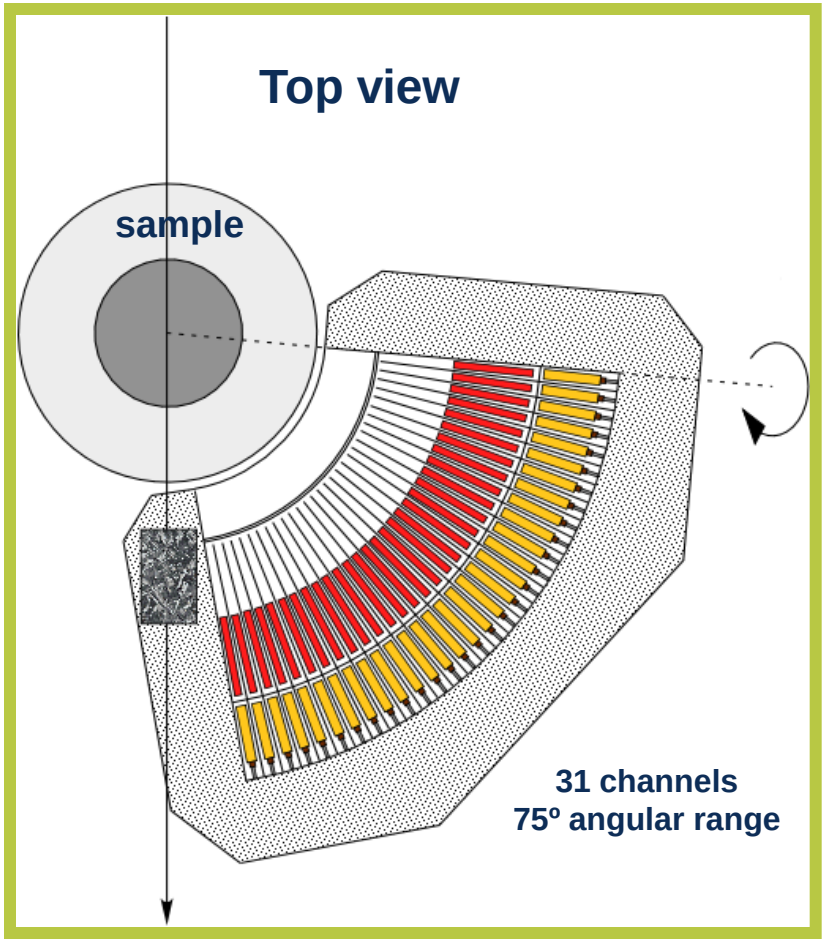
IMPS hardware

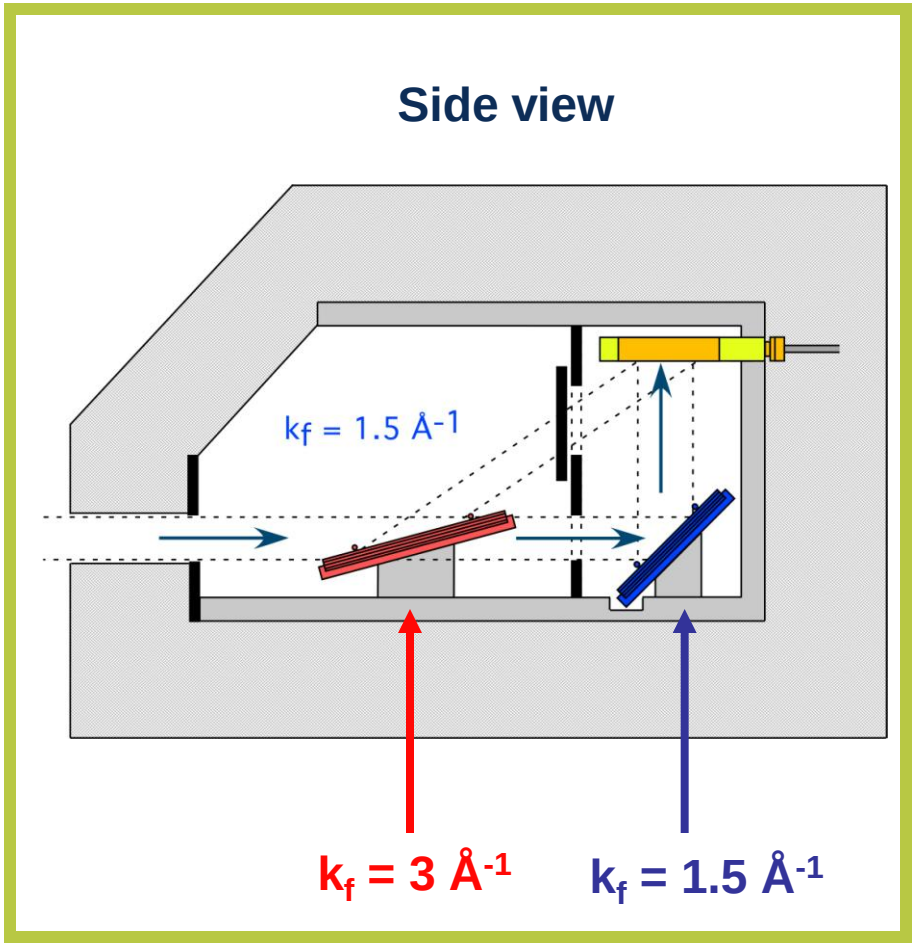


Detector
- resolution: hor. 5 mm, vert. 5-10 mm
- linear 3He filled PSDs
- 54 tubes of Φ = 9 mm
- total size = 509 x 200 mm^2

Multianalyser:
- 9 blades (40 mm x 120 mm each)
- Acceptance angle ~ 5°
- Variable collimator system
- Ge111 plastically bent (R = 5 m)
- no 2nd order
- Gain in counting rate: factor 3

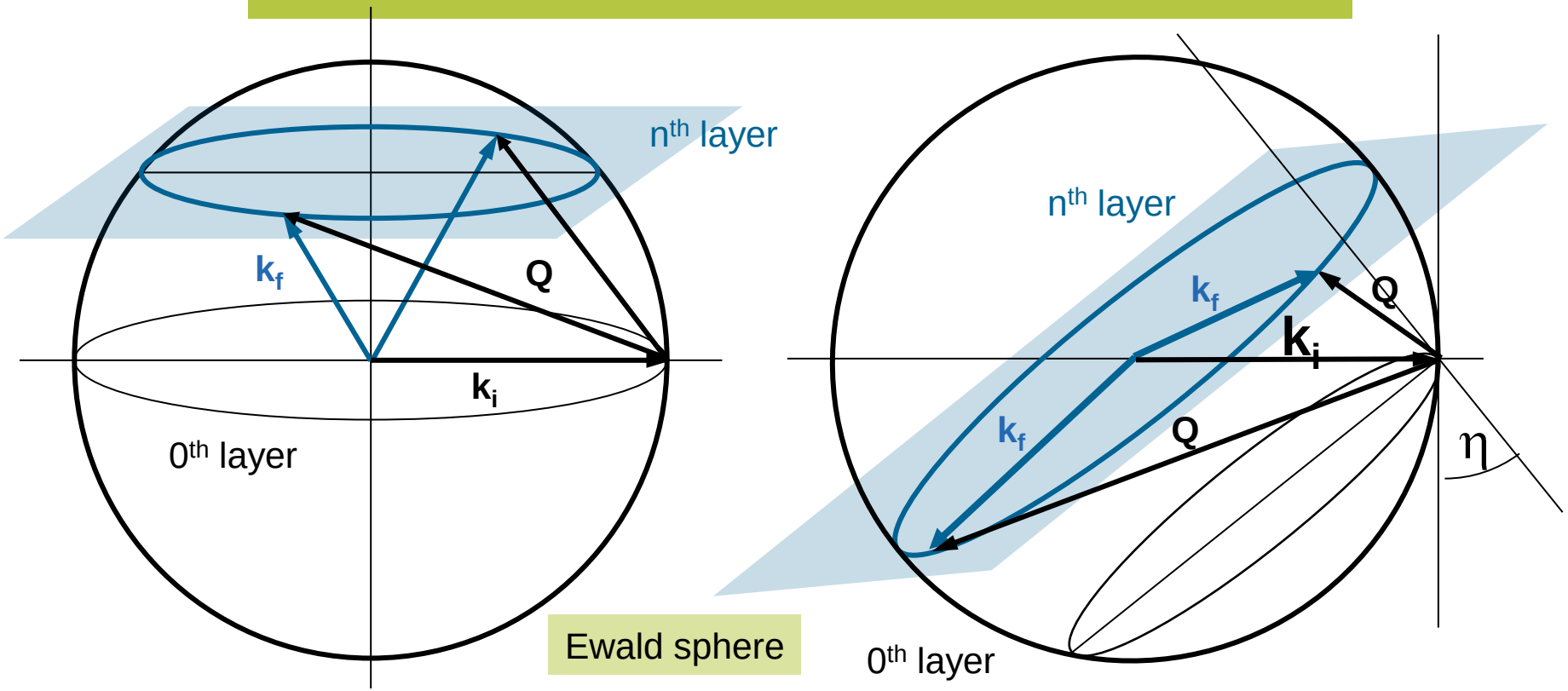
FLAT CONE II


Top view
sample
31 channels
75º angular range


Side view
k_f = 1.5 Å^{-1}
k_f = 3 Å^{-1}
k_f = 1.5 Å^{-1}

FC geometry

- **access to nth layer scattering with a linear multidetector**

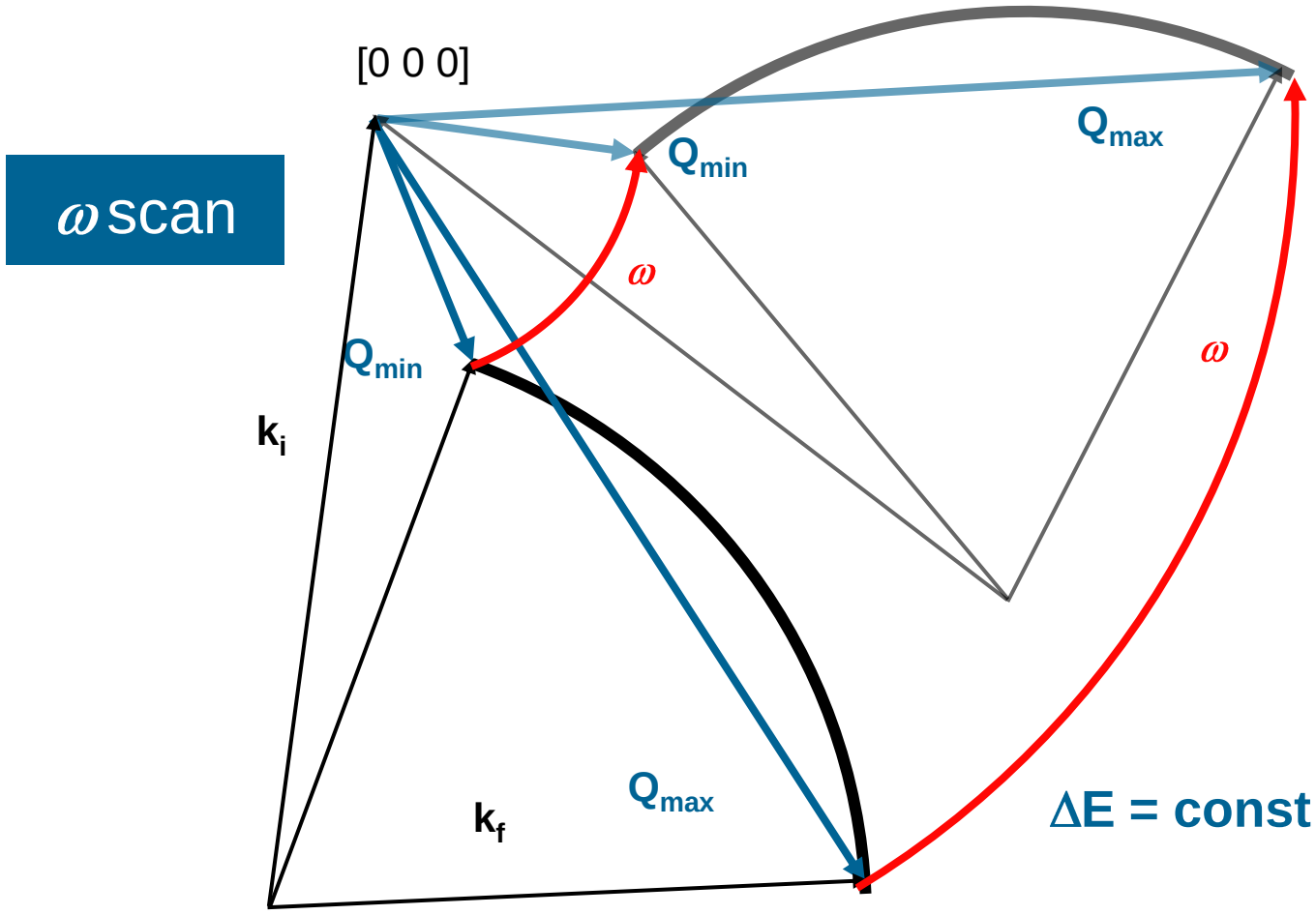


Ewald sphere

- **conventional rotating crystal method:**
  k_f's form a cone
  out of equatorial plane

- **tilt crystal to bring k_i into nth layer:**
  k_f's become coplanar
  forming a "flat cone"

FC scan modes (I)



- **essential for mapping Q,E space:**
  sweeps a plane in reciprocal space at ΔE = const

Relaxor ferroelectrics

$PbTiO_3$

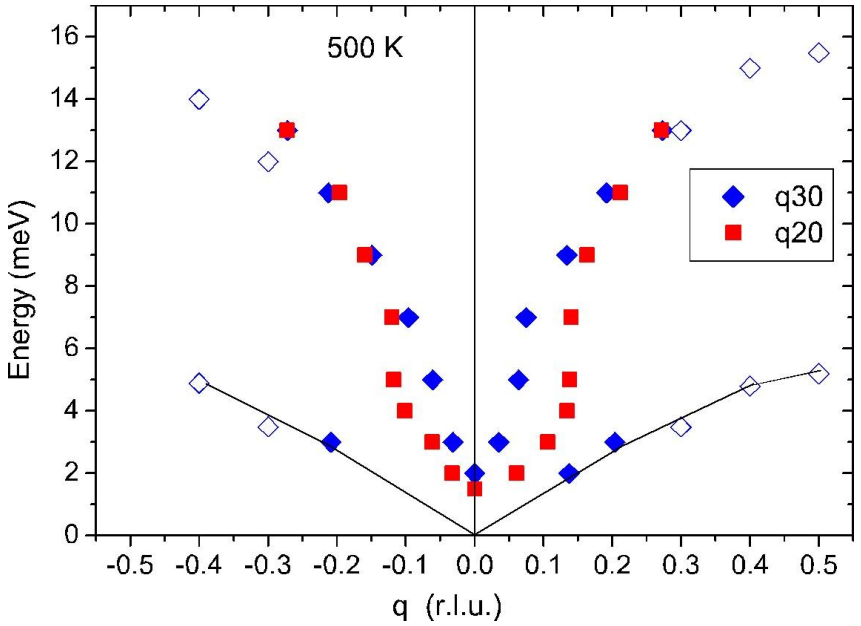
Ti

Ba O

perovskite lattice
(high T: cubic)

"waterfall" anomaly in relaxor ferroelectric
PZN-8%Pt

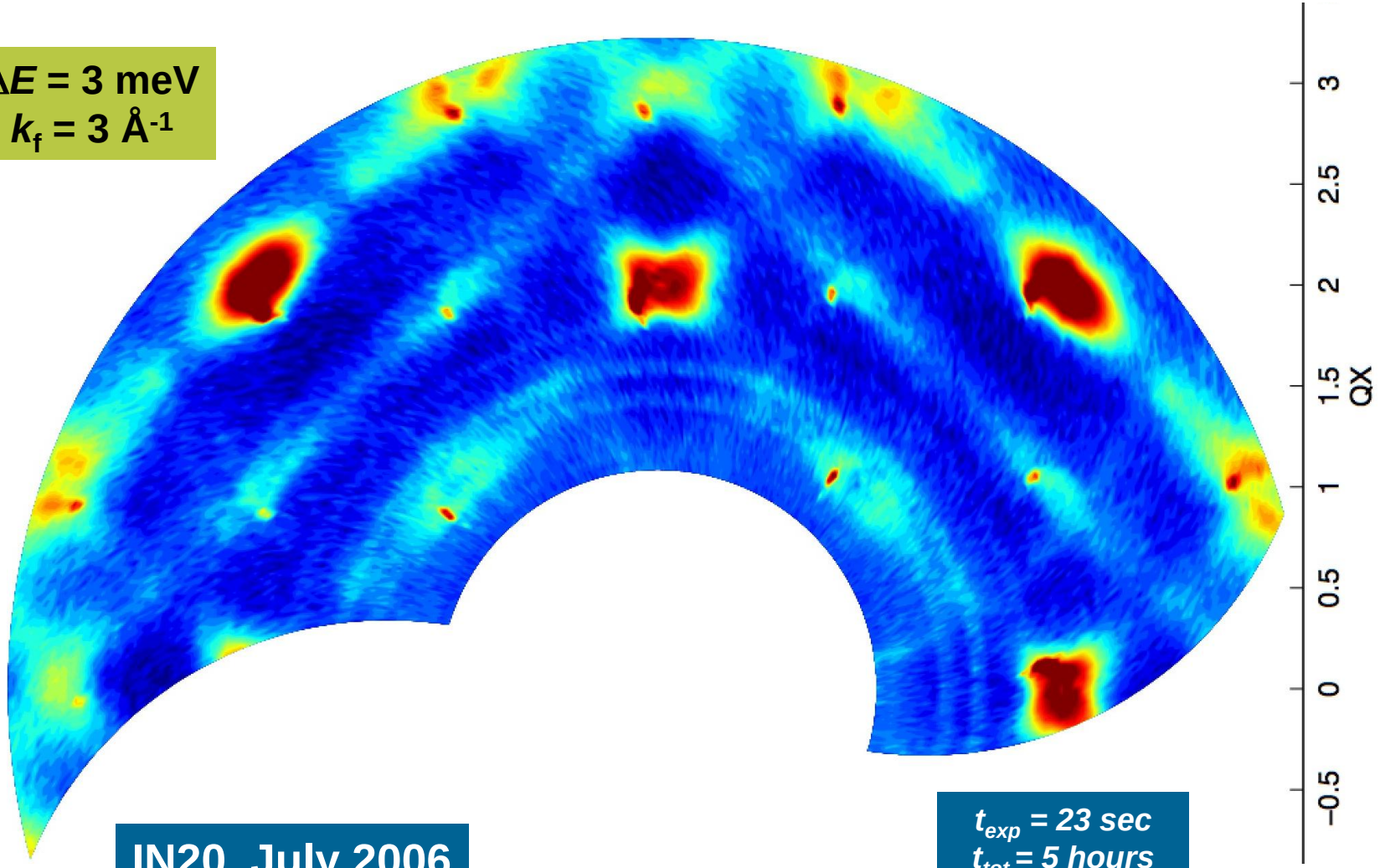
$(Pb(Zn_{1/3}Nb_{2/3})O_3$ (PZN) with 8% PbTiO3)



J. Hlinka et al., Phys. Rev. Lett. 91 (2003) 107602

PZN-8%Pt relaxor

ΔE = 3 meV
k_f = 3 Å^{-1}



IN20, July 2006

t_{exp} = 23 sec
t_{tot} = 5 hours
62 x 360 pixels

Advantages and disadvantages of the triple-axis method

Advantages

1. Can focus all intensity on point in reciprocal space that is important
2. Can make measurements along high-symmetry directions
3. Can use either constant-$\boldsymbol{Q}$ or constant–E, depending on type of excitation being examined.
4. Can use focusing and other 'tricks' to improve the signal/noise
5. Can use polarisation analysis to separate electronic and phonon signals

Disadvantages

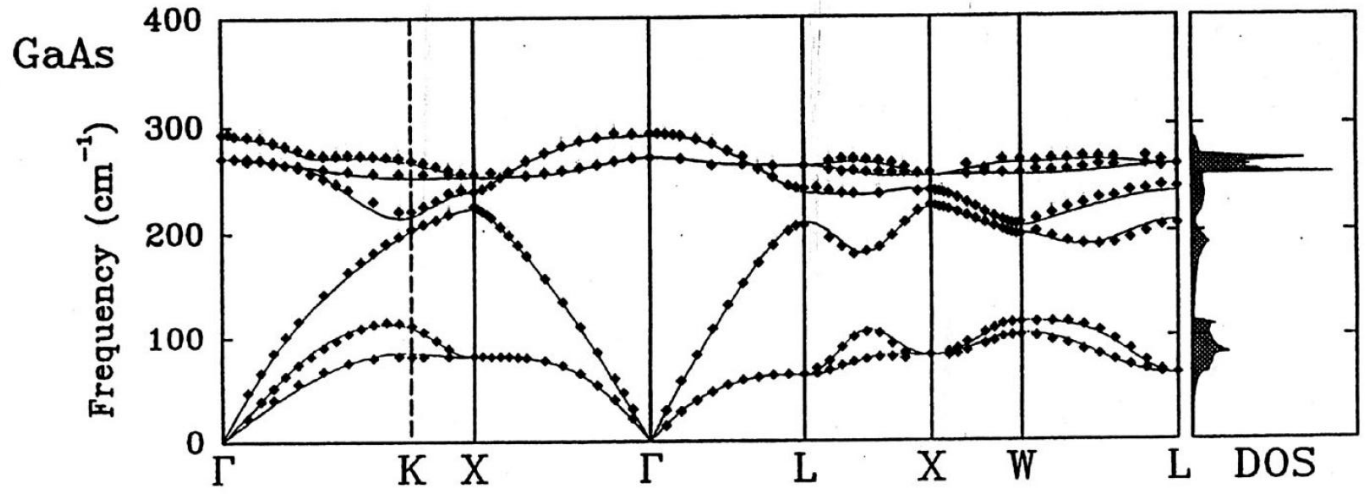
1. Technique is slow and requires some expert attention
2. Use of monochromators and analysers gives rise to possible higher-order effects that give rise to "spurions"
3. With measurements restricted to high-symmetry directions it is possible that something important might be missed

Examples of triple-axis spectroscopy

This technique was invented by Bertram Brockhouse (at Chalk River National Lab in Canada) [Nobel Laureate in 1994] in the 1950s and although the instruments have improved beyond recognition, the basic technique has not changed.

There are thus 100's of examples to choose from and we have time for only a few.

Phonons: agreement with experiment & theory

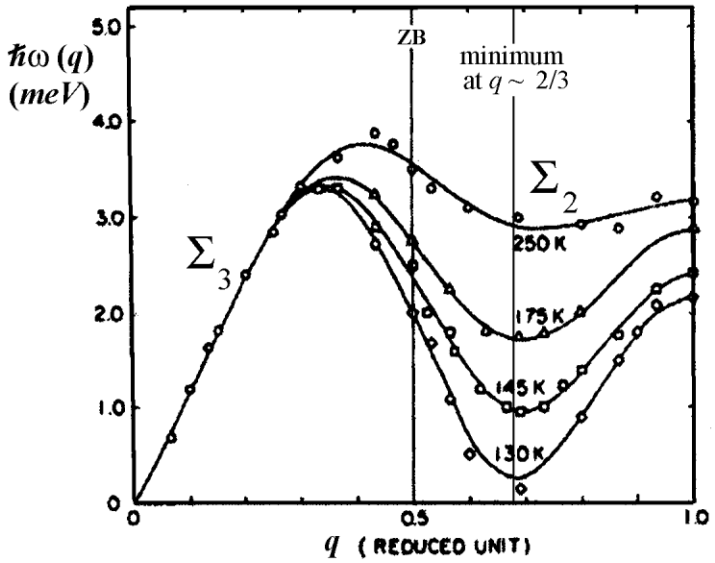


Phonon dispersion curves as measured for GaAs. Strauch & Dorner (1990). The lines give the result of *ab initio* calculations and show that the forces between the atoms are well understood. The letters below give the notation for the symmetry directions or points.

Note the calculated density-of-states on the extreme right.

1 meV = 8.065 cm^{-1}

Phase transitions in solids



Dispersion relations of the Σ_2 soft mode and the Σ_3 acoustic mode in K_2SeO_4 plotted in an extended zone. Z.B. indicates the original zone boundary. The solid lines show the results of fitting force constant models to the data.

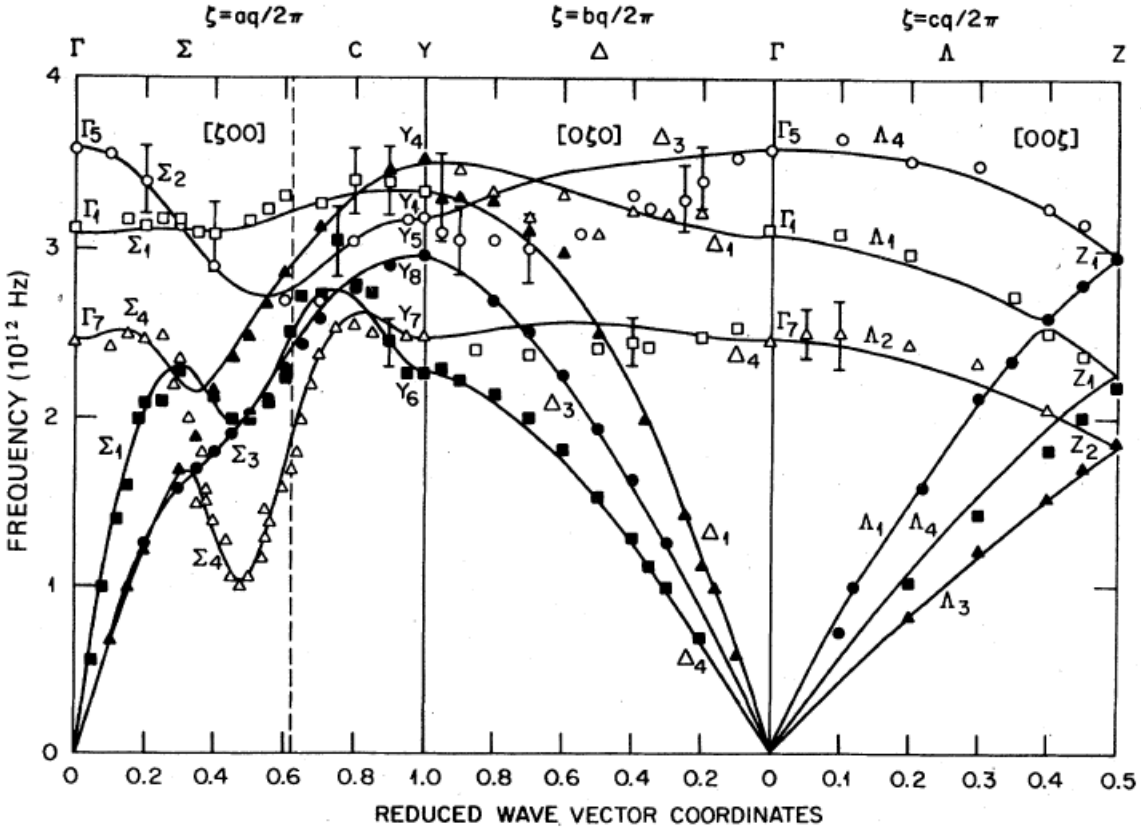
If one looks carefully at these data one can see that the initial "soft" point is not at $q = 2/3$, but rather at $q = 2/3 + \delta$

This is an example of an incommensurate phase transition, stabilised by higher-order terms in the Landau expansion.

Phonons in alpha-Uranium at T=300 K

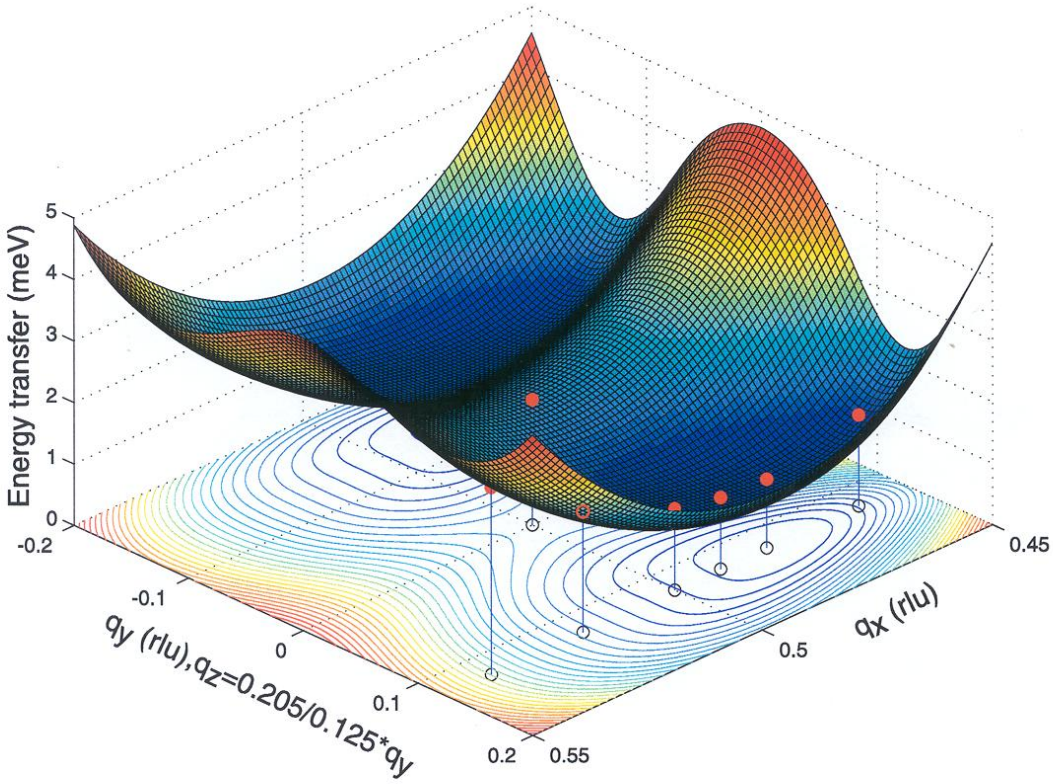
Note the anomaly in the ***a*** direction [100] and the incipient minimum of the Σ_4 mode. The other two directions are 'normal'

At low temperature the material transforms in a complex way, developing a charge-density wave.



Crummett *et al.* Phys. Rev. B (1979)

Characterization of the soft mode in alpha-U



The transition does ***not*** occur at a high symmetry direction. One needs components along all three crystallographic directions, and none of them are commensurate at T_{CDW}

T = 50 K

T_{CDW} + 7 K

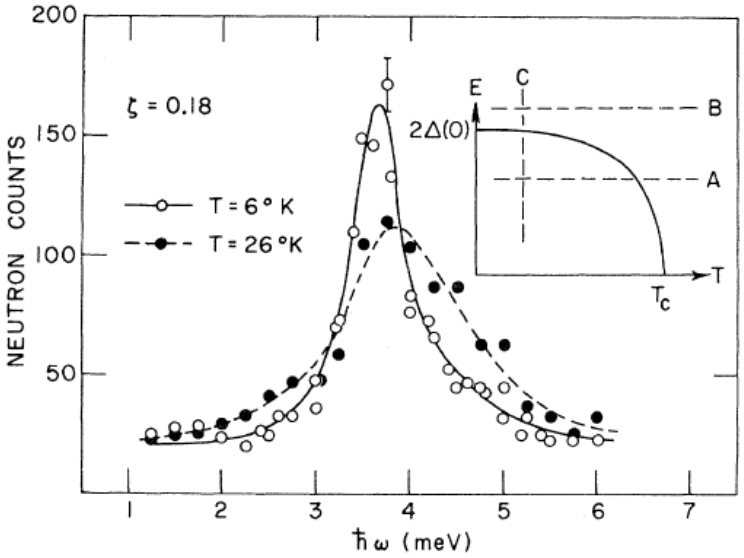
Experiments at ILL, J. C. Marmeggi *et al.*

Phonons and superconductivity

In the BCS theory (1957) of *s*-wave superconductivity the lattice vibrations (phonons) mediate the attraction between electrons and form the Cooper pairs. Thus the measurements of phonons, in particular, the total phonon-density of states, is important to understand the overall mechanism of superconductivity.

Many phonon studies using neutrons have been done with this motivation.

An example of an electron-phonon interaction as measured by neutrons



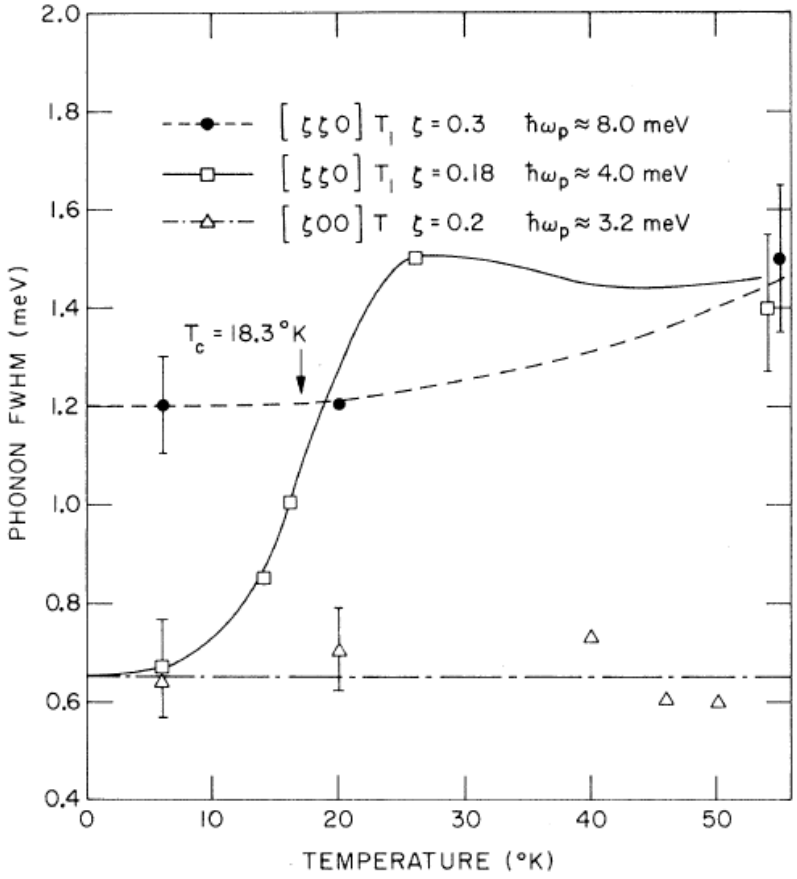
Inelastic scattering from Nb_3Sn, $T_c = 18.3$ K.

For phonons with $E < 2\,\Delta(T)$, where $\Delta(T)$ is the s/c energy gap, there is a loss of a damping mechanism as the low-energy electron states form Cooper pairs.

Notice how this phonon is *much* broader for $T > T_c$.

Axe & Shirane, PRL 30, 214 (1973)

Electron-phonon interaction in Nb_3Sn



Summary of line-widths as f(T)
Note that there is *anisotropy* in the damping mechanism. No measurable effect is observed along [100], but a large effect along [110]

No effect is observed for a phonon of E = 8 meV. This is above 2 Δ(0).

The gap is well determined as 2 Δ(0) = 7 ± 1 meV = (4.4 ± 0.6) $k_B T_c$ in excellent agreement with the specific heat value of 4.8 $k_B T_c$.

Correlated electrons and superconductivity

This is a subject of great current activity so a final "answer" cannot be given. The start of this subject came with the paper by Steglich *et al.*, in 1979 on $CeCu_2Si_2$, with $T_c = 0.6$ K. Following this, a number of Ce & U compounds were found s/c up to ~ 2 K. All these materials have a large enhanced electronic specific heat, i.e. a γ value of up to 1000 mJ/mol/K^2 as compared to Cu with $\gamma = 1$. Many, if not most, are near magnetic instabilities, suggesting that magnetic fluctuations play *some* role in the s/c.

What followed was the discovery of s/c in Sr doped La_2CuO_4 by Bednorz & Müller (1986) and then $YBa_2Cu_3O_{6+x}$ and a whole zoo of new materials so that now maximum $T_c \sim 120$ K.

Correlated electrons and superconductivity

It was quickly discovered that the parent compounds, e.g. $LaCuO_4$ and $YBa_2Cu_3O_6$ are *antiferromagnetic*. The order disappears on doping, but do the magnetic fluctuations remain and are they important for the superconductivity?

Just to complete the "story" other compounds, such as UGe_2 and URhGe have been found *ferromagnetic* and s/c.

Perhaps most amazing of all is the 2002 discovery of s/c in a $PuCoGa_5$ compound at 18 K. This is the first such Pu-based superconductor!

Correlated electrons and superconductivity

A great problem with superconductivity is that we do not have a microscopic probe such as the neutron, that couples to the s/c order parameter. That would allow the high-T_c problem to be solved and the 2nd Nobel Prize to be given to theory!

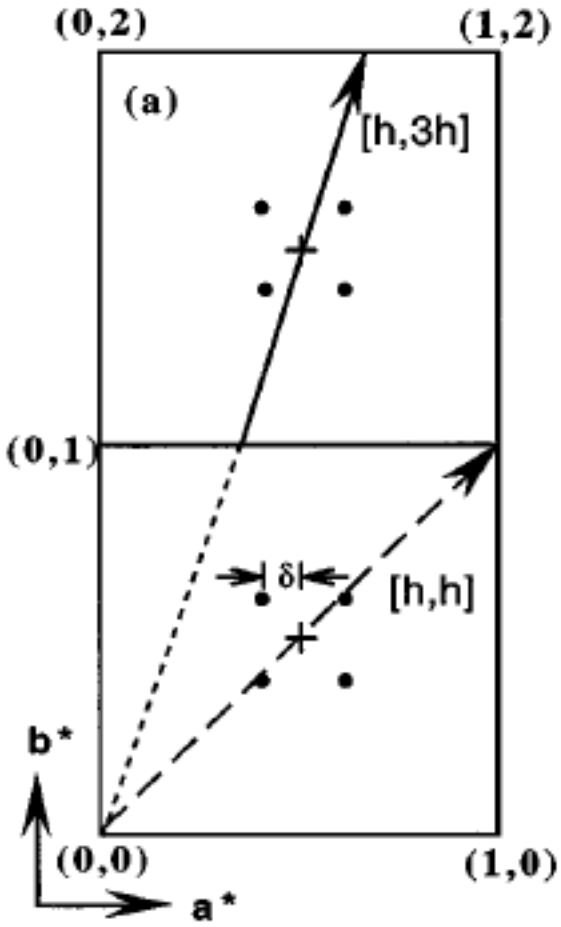
More modestly, I shall discuss some aspects of the magnetic fluctuations in doped $YBa_2Cu_3O_{6+x}$.

Magnetic fluctuations in superconducting $YBa_2Cu_3O_{6+x}$

Work on doped $LaCu_2O_4$ showed that inelastic scattering, corresponding to magnetic fluctuations, appeared incommensurate. The situation in Y123 remained unclear for some time. Difficult because many optic phonons at high energies. PA loses too much intensity.

First a spin "gap" was found in Y123 at about 41 meV for x =0.93
{If T_c = 94 K; 2 $\Delta(0) = 5.1\ k_B T_c$}
In this example x=0.6, T_c = 62.7 K and spin gap found at 34 meV; 2 $\Delta(0) = 6.3\ k_B T_c$

What is the nature of the excitations?



P. Dai *et al.* PRL 80, 1738 (1998)

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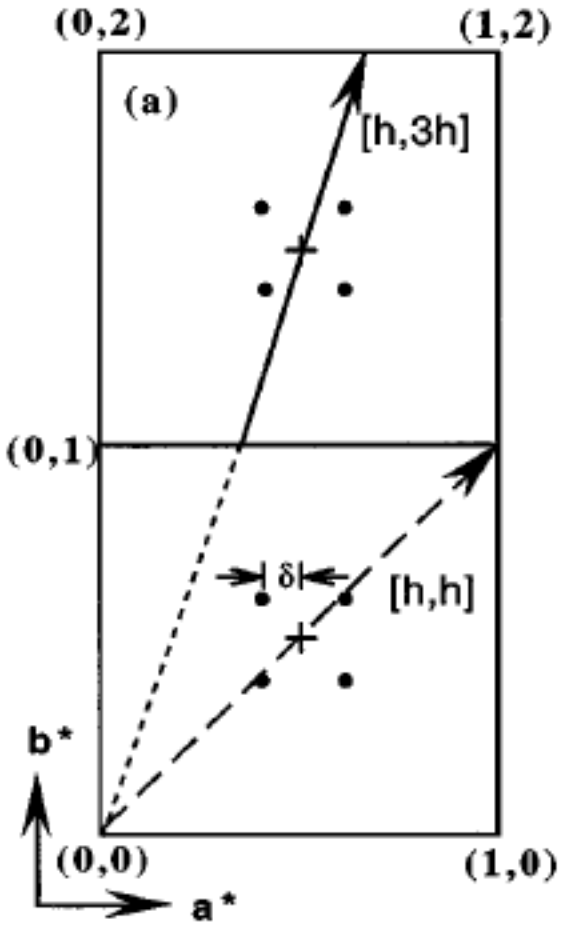

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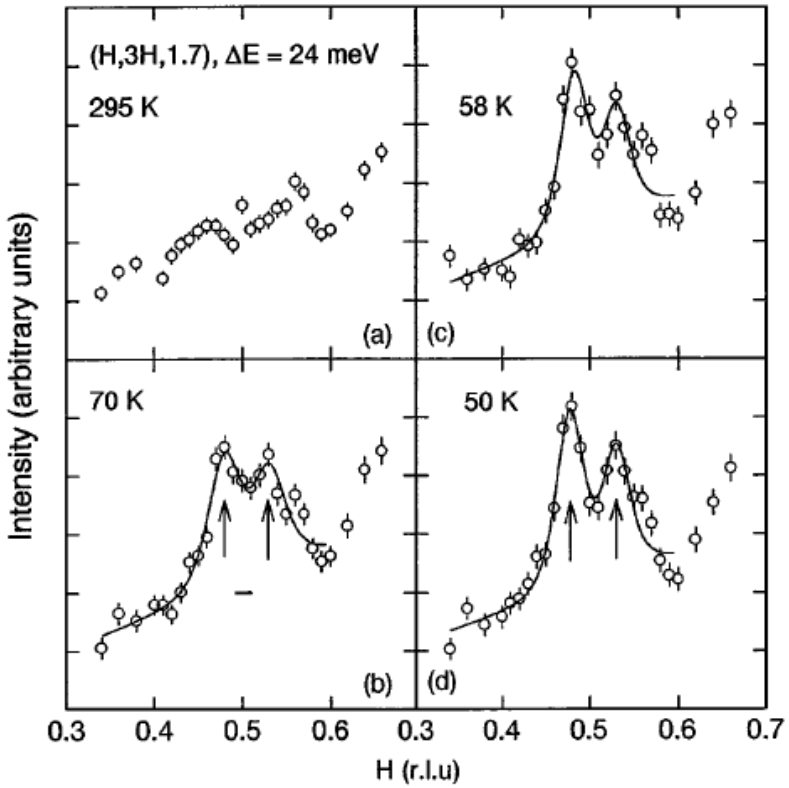
Magnetic fluctuations in superconducting $YBa_2Cu_3O_{6+x}$

Scans at E=24 meV show an incommensurate signal in the same way as found in the 214 materials.

Notice that these energies are below the "resonance" energy of 34 meV for this material. At the resonant energy there is a large change in the intensity at T_c.

What happens for higher energies?



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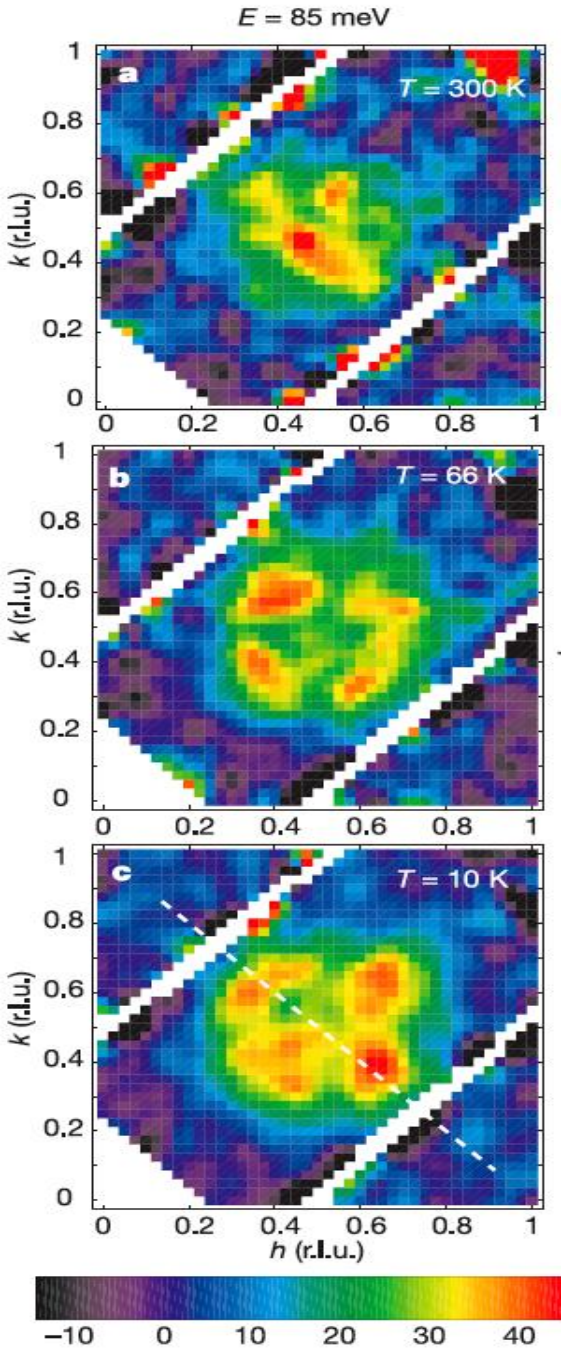


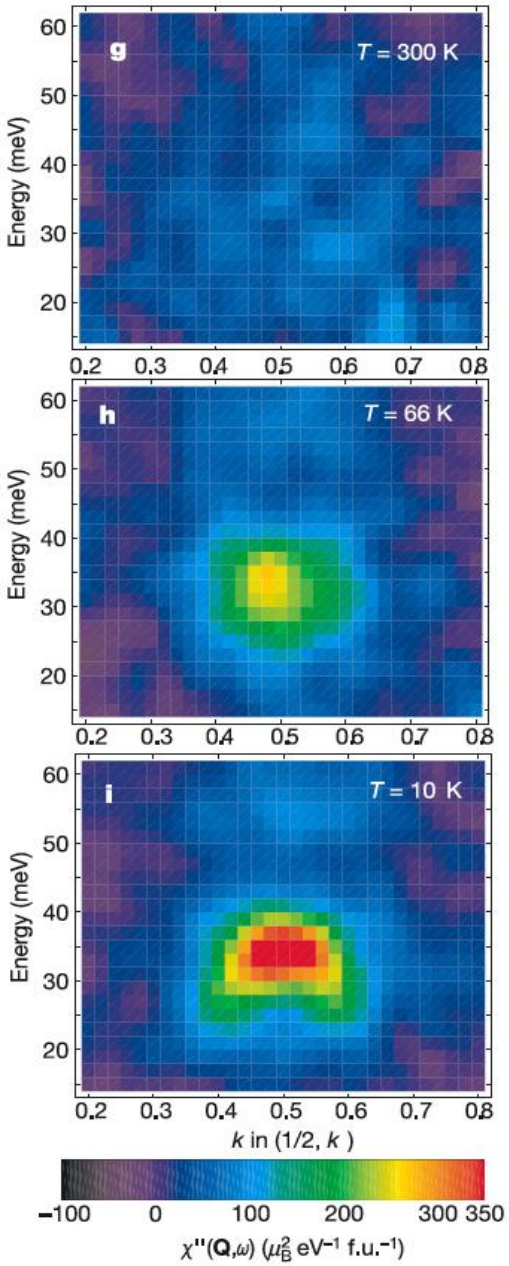
Magnetic fluctuations in superconducting $YBa_2Cu_3O_{6+x}$

The excitations are incommensurate below T_c even at high energies, in fact to 100 meV.

These data have be taken on the MAPS spectrometer at ISIS and show the great advantage of the multidetector system coupled to a pulsed source.

E_i = 170 meV; crystal 25 g. T_c = 63 K

S. M. Hayden *et al.*, *Nature* 429, 531 (2004)



Magnetic fluctuations in superconducting $YBa_2Cu_3O_{6+x}$

Although there are magnetic excitations over a wide range of energies near (1/2 1/2), they are by far the strongest at the resonant energy, in this case 34 meV, and below T_c = 63 K

This is an integration so cannot show the incommensurate nature.

S. M. Hayden *et al.*, *Nature* 429, 531 (2004)

Magnetic fluctuations in superconducting $YBa_2Cu_3O_{6+x}$

What causes these excitations and are they related to the special superconductivity in these materials?

(1) Incipient spin-charge separation leading to "stripes"

(2) Electron-hole pair excitations governed by the underlying Fermi surface.

Many theories are presently trying to reproduce these effects. Together with photoemission (ARPES), these neutron scattering experiments are probably the most important set of data in the quest to understand high T_c mechanism.

Conclusions on neutrons and superconductivity

Unlike, for example, the determination of magnetic structures or the measurement of phonon frequencies, neutrons do not give unique answers to the question of what causes the formation of Cooper pairs.

The results need to be compared to theory and in this way both can advance.

Summary: Triple-axis spectroscopy

From its beginning in the 1960s 3-axis spectroscopy has been greatly influential in problems of physics.

Now, pulsed sources are able to compete with reactors.

It is *not* the first experiment to be done: it demands *large* single crystals (0.2 → 20 g) and long experiments.

However, in terms of the detailed physics of a material it brings unmatched detail.