Excitations
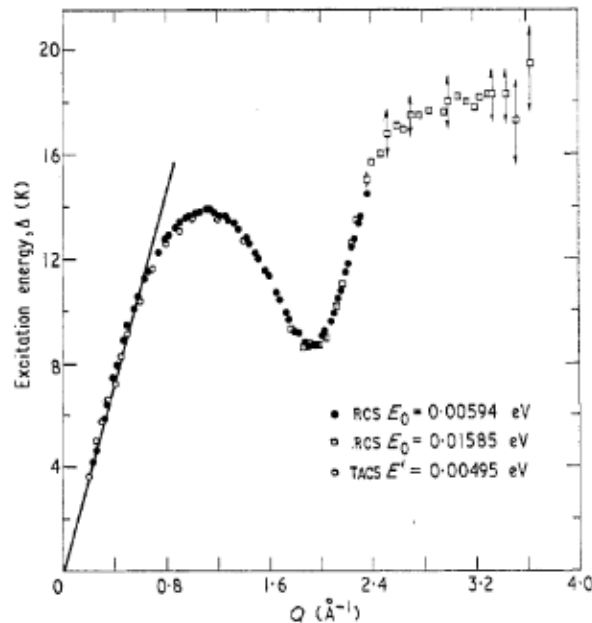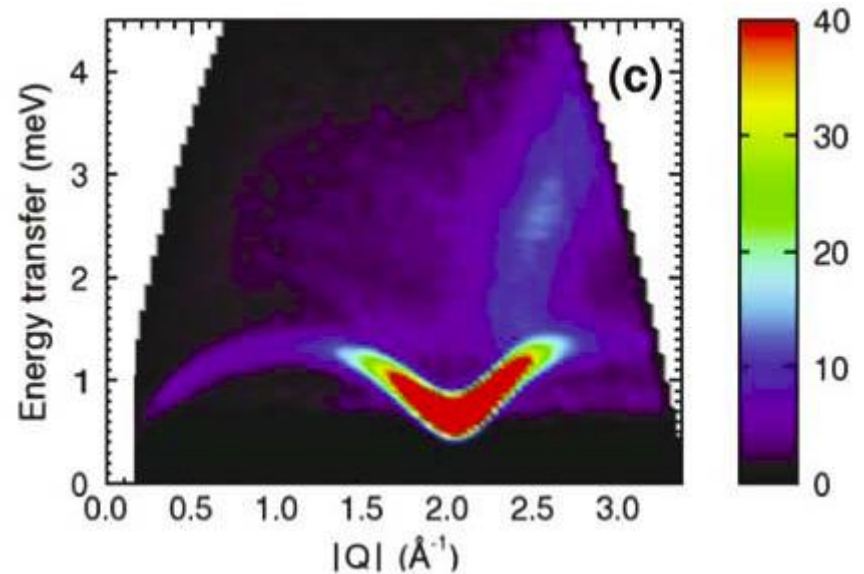
Elizabeth Blackburn

University of Birmingham

Cowley and Woods., Can. J. Phys. 49, 177 (1971)

Blackburn *et al.*, Pramana 71, 673 (2008)

15th Oxford School of Neutron Scattering

Excitations

Elizabeth Blackburn

University of Birmingham

- What are excitations?

- What can we learn from then?

- How do we measure them?

15th Oxford School of Neutron Scattering

What are excitations?

The inelastic (or dynamical) response of the material to stimulus.

What are excitations?

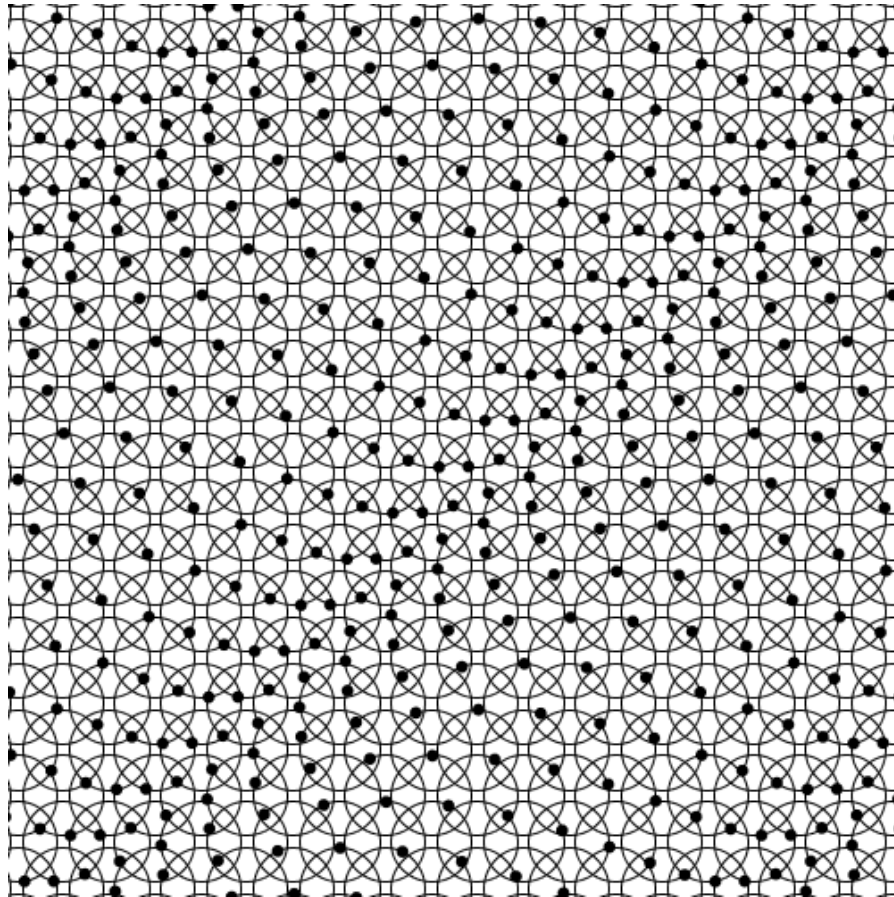The inelastic (or dynamical) response of the material to stimulus.

Dugald Duncan/Heriot-Watt University

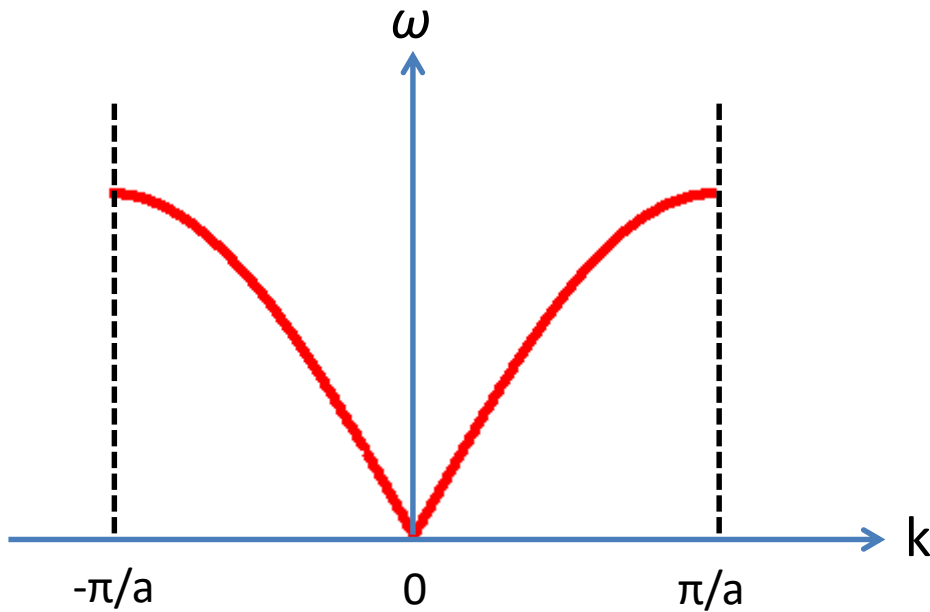Soliton on the Scott Russell Aqueduct, near Union Canal

What are excitations?

The inelastic (or dynamical) response of the material to stimulus.

FrozenMarshmallow, http://imgur.com/gallery/cSaBG0I

Phonons on a 1-D chain

$$\omega_{ph} = 2\sqrt{\frac{K}{M}} \left| \sin\left(\frac{k_{ph}\,a}{2}\right) \right|$$
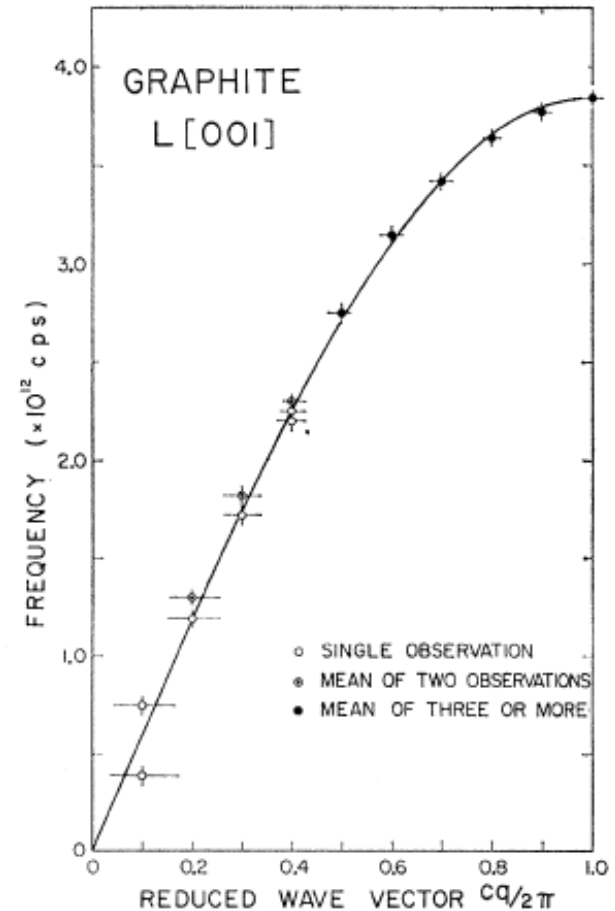

FIG. 3. Dispersion curve for the L[001] modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

What are excitations?

The inelastic (or dynamical) response of the material to stimulus.

Examples
Phonons (lattice vibrations)
Magnons (spin vibrations)
Crystal field excitations
Vibrations internal to a molecule

How can we measure them?
Direct observation where possible
Neutron scattering
Inelastic X-ray scattering
Raman spectroscopy
Ac susceptibility ….

It really depends on the energy associated with the excitation and any wavevector dependence it has.

What are excitations?

The inelastic (or dynamical) response of the material to stimulus.

Examples
Phonons (lattice vibrations)
Magnons (spin vibrations)
Crystal field excitations
Vibrations internal to a molecule

Magnon video
Martin Boehm and Alain Filhol

For neutron scattering
The excitation has to couple to the neutron, either through the strong nuclear interaction or via the magnetic moment.

Recap from Intro to Neutron Scattering

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E_f} = \frac{\text{No. particles scattered per sec. into solid angle } \mathrm{d}\Omega \text{ with final energies between } E_f \text{ and } E_f + \mathrm{d}E_f}{I_0 \times \mathrm{d}\Omega \times \mathrm{d}E_f}$$

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E_f} = \frac{k_f}{k_i} S(\mathbf{Q},\omega)$$

$S(\mathbf{Q},\omega)$ is the scattering function or response function

$S(\mathbf{Q},\omega)$ is determined by the physics of the material.
You have just heard about the expectations here for phonons and for spin waves.

Recap from Intro to Neutron Scattering

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E_f} = \frac{\text{No. particles scattered per sec. into solid angle } \mathrm{d}\Omega \text{ with final energies between } E_f \text{ and } E_f + \mathrm{d}E_f}{I_0 \times \mathrm{d}\Omega \times \mathrm{d}E_f}$$

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E_f} = \frac{k_f}{k_i}\, S(\mathbf{Q},\omega)$$

$S(\mathbf{Q},\omega)$ is the scattering function or response function

---

(i) Phonons

$$S(\mathbf{Q},\omega) \propto \exp\{-2W(Q,T)\} \times |G(\mathbf{Q})|^2 \times [n(\omega_{ph})+1] \times \frac{1}{\omega_{ph}} \times Q^2$$

(ii) Spin waves

$$S(\mathbf{Q},\omega) \propto \exp\{-2W(Q,T)\} \times [n(\omega_{mag})+1] \times \frac{1}{\omega_{mag}} \times f^2(Q)$$

Phonons on a 1-D chain
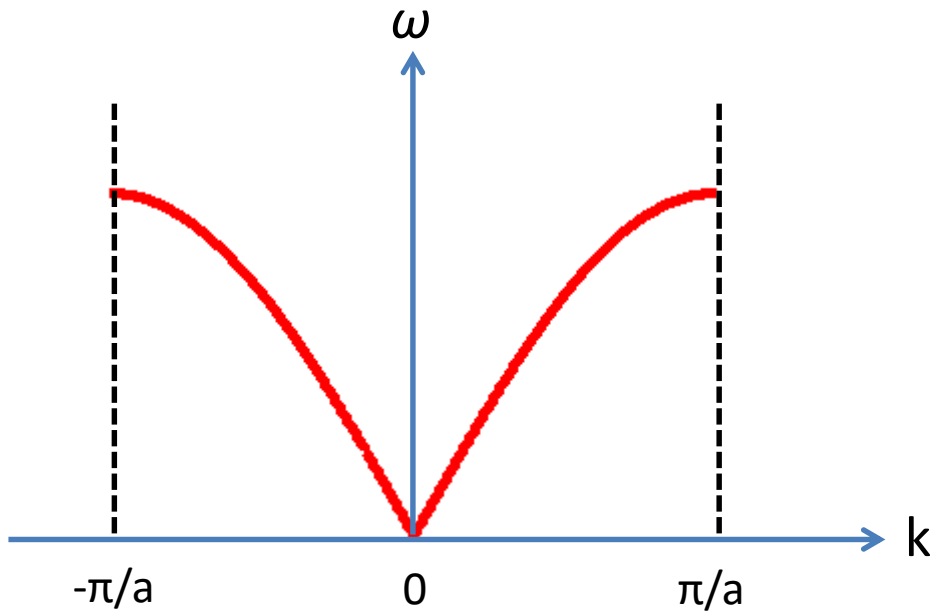
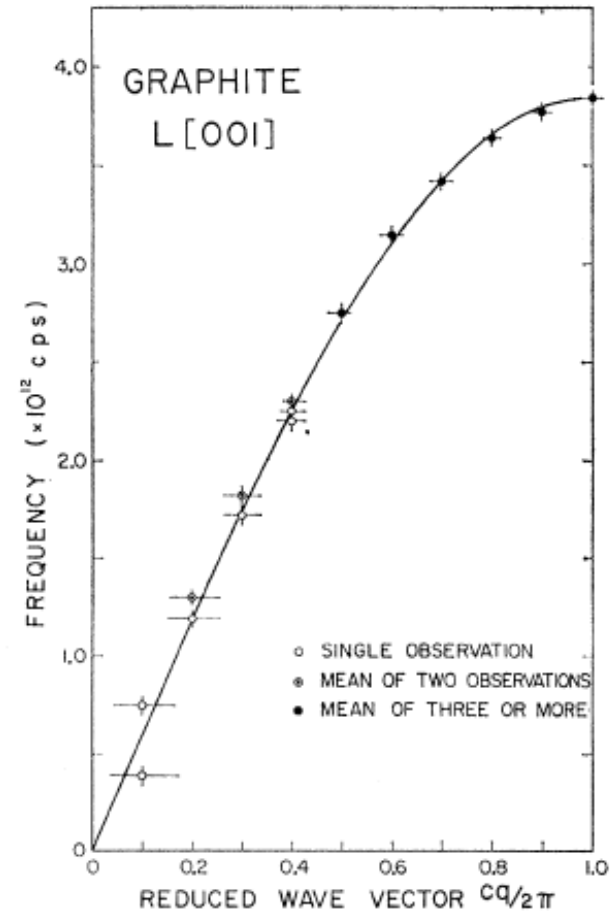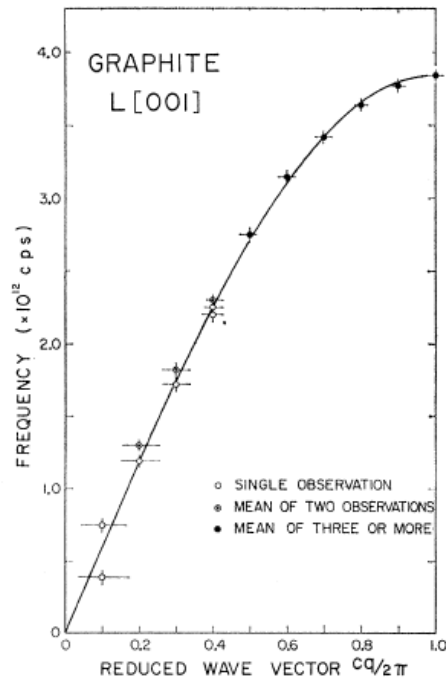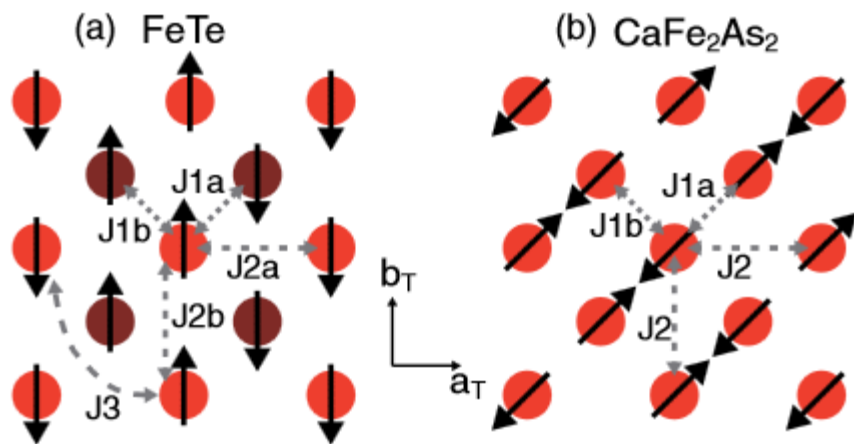$$\omega_{ph} = 2\sqrt{\frac{K}{M}}\left|\sin\left(\frac{k_{ph}\,a}{2}\right)\right|$$

FIG. 3. Dispersion curve for the L[001] modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.
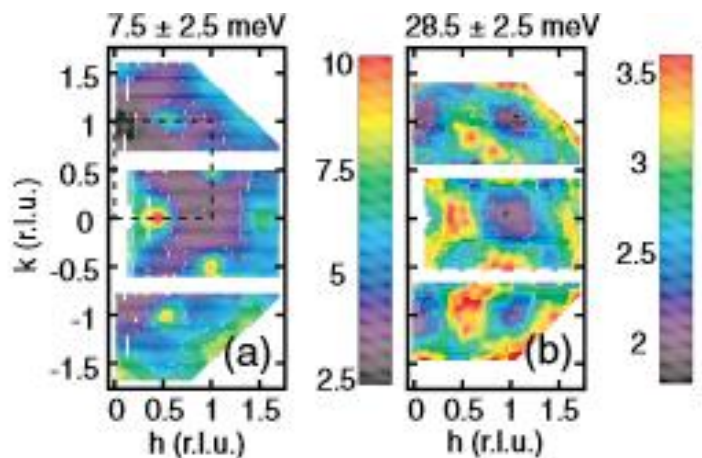
Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

What can we learn from them?

FIG. 3. Dispersion curve for the L[001] modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

- Elastic constants

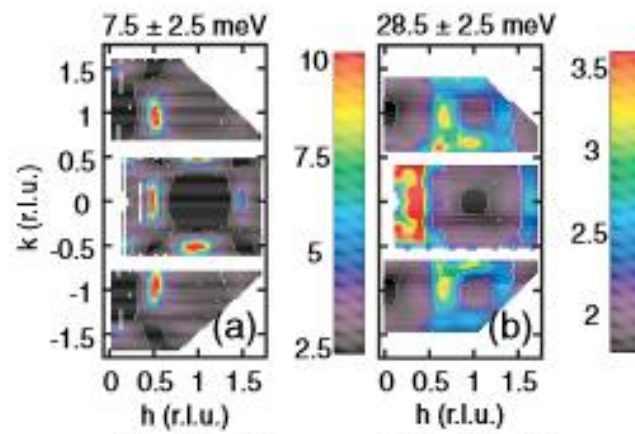$$\omega_{ph} = 2\sqrt{\frac{K}{M}} \left| \sin\left(\frac{k_{ph}\, a}{2}\right) \right|$$

Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

What can we learn from them?

(a) FeTe

(b) CaFe$_2$As$_2$

- Elastic constants

- Spin Hamiltonians

Data

Simulation
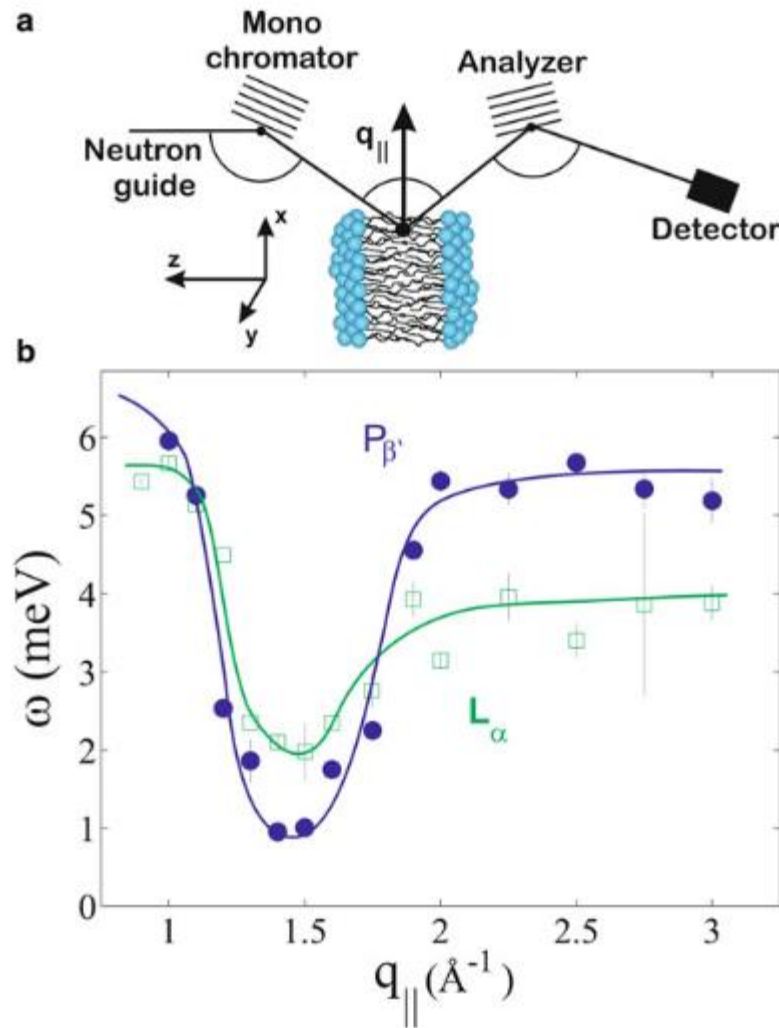
Lipscombe *et al.*, Phys. Rev. Lett. 106, 057004 (2011)

What can we learn from them?

- Elastic constants

- Spin Hamiltonians

- Membrane dynamics

Fig. 10.5 (a) Schematic of a triple-axis spectrometer. (b) Short-wavelength dispersion relations in the gel ($P_{\beta'}$) and fluid phase L_α of the phospholipid (DMPC) bilayers [42]

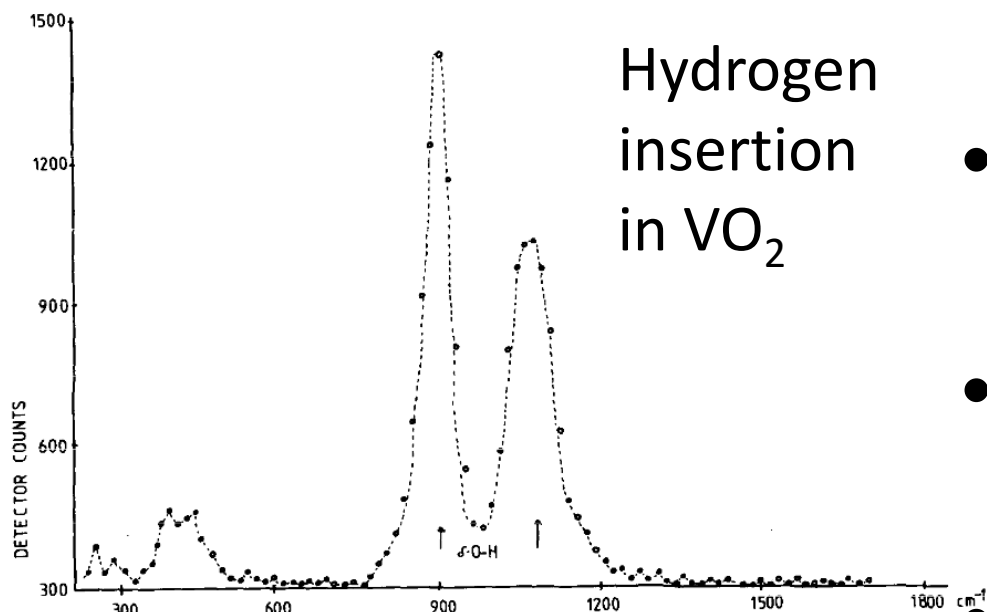Rheinstaedter, in *Dynamics of Soft* Matter, Springer (2012).

What can we learn from them?

Hydrogen insertion in VO_2

FIG. 2. Inelastic neutron scattering spectrum of $H_{0.3}VO_2$ at 80 K.

- Elastic constants

- Spin Hamiltonians

- Membrane dynamics

- How to synthesise particular molecules

TABLE V

FREQUENCIES AND ASSIGNMENTS OF GENERATED VIBRATIONAL MODES OF H_xVO_2

Calculated frequency (cm^{-1})	Observed frequency (cm^{-1})	Assignment	Mode
3407 ($\times 4$)		$B_{1g}\ B_{3u}\ B_{2u}\ A_g$	ν(O–H)
1098 1095	1083	$B_{2u}\ B_{3u}\ B_{1g}\ A_g$	δ(O–H) in xy plane
1093 1092			
918 916 914	909	$B_{1u}\ A_u\ B_{3g}\ B_{2g}$	δ(O–H) out of xy plane
914			
587 583 553	476 431	$B_{3u}\ B_{2u}\ B_{1u}\ A_g$	
517 515 493		$B_{1g}\ A_u\ B_{3g}\ B_{2g}$	Lattice modes
414 414 342		$B_{2u}\ B_{1g}\ B_{3u}$	
342 341 322		$B_{3u}\ B_{2u}\ A_u\ A_g$	
292 267 212			
177			

Chippindale *et al.*, J. Solid State Chem. 93, 526 (1991).
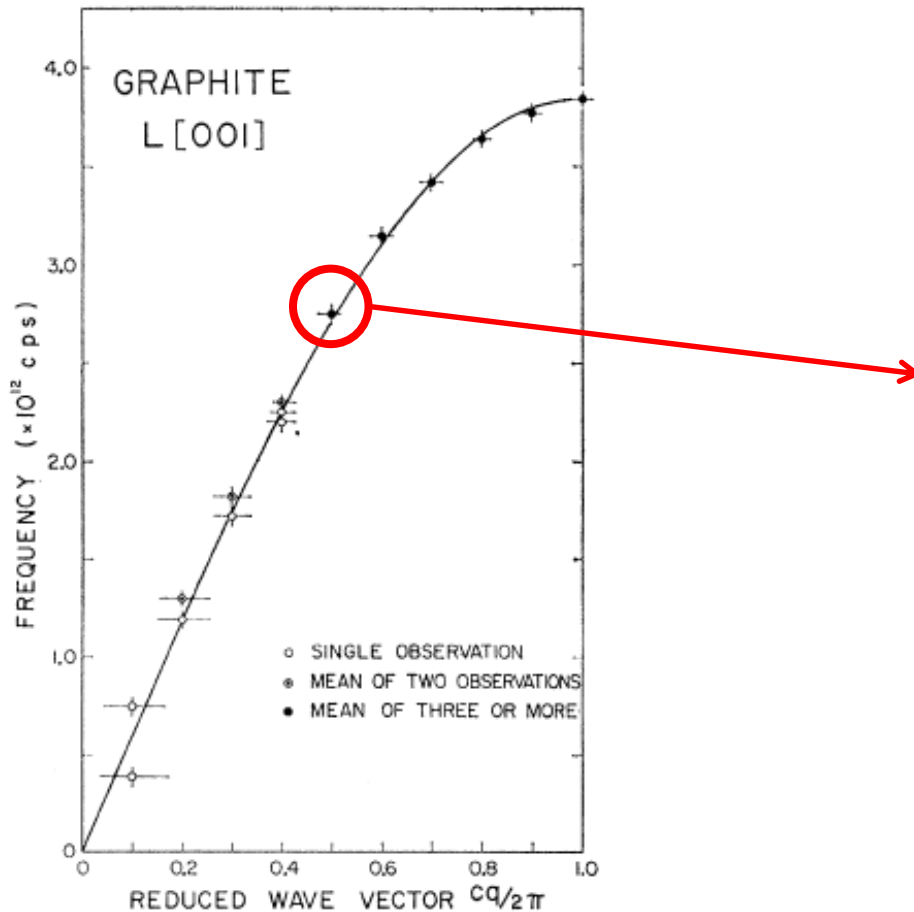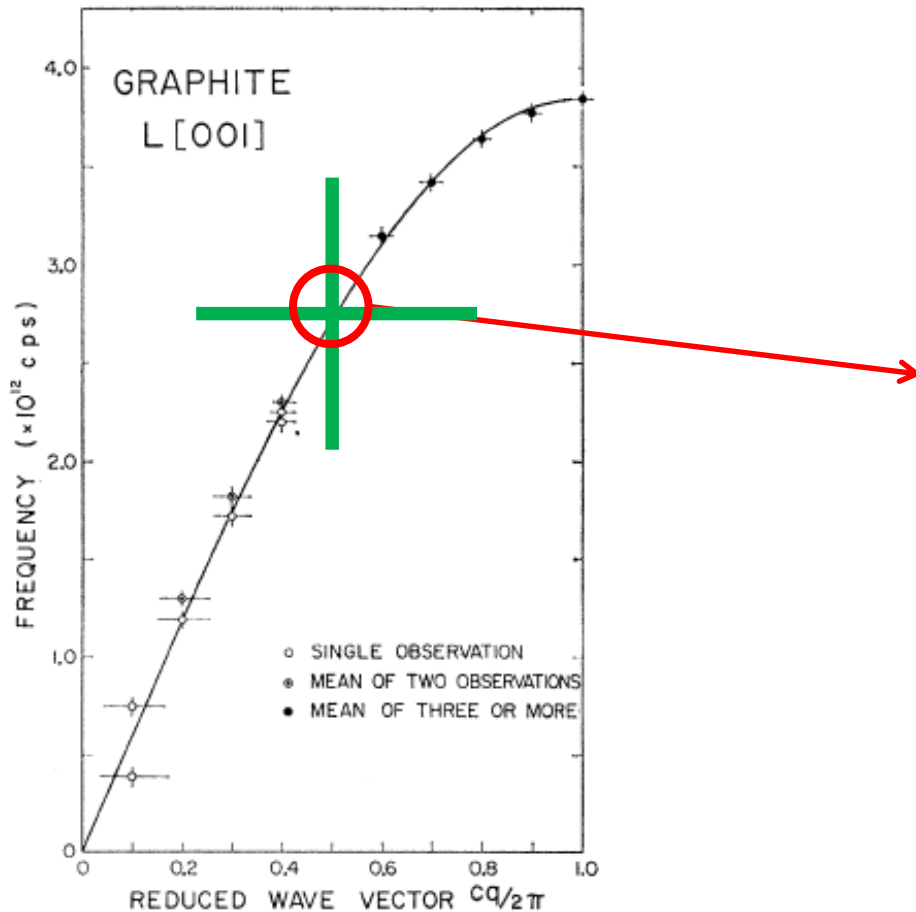
How do we measure excitations?

FIG. 3. Dispersion curve for the $L[001]$ modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

Measuring a phonon spectrum

FIG. 3. Dispersion curve for the $L[001]$ modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

Things to consider:
- resolution
- spurions

How do we measure these points?
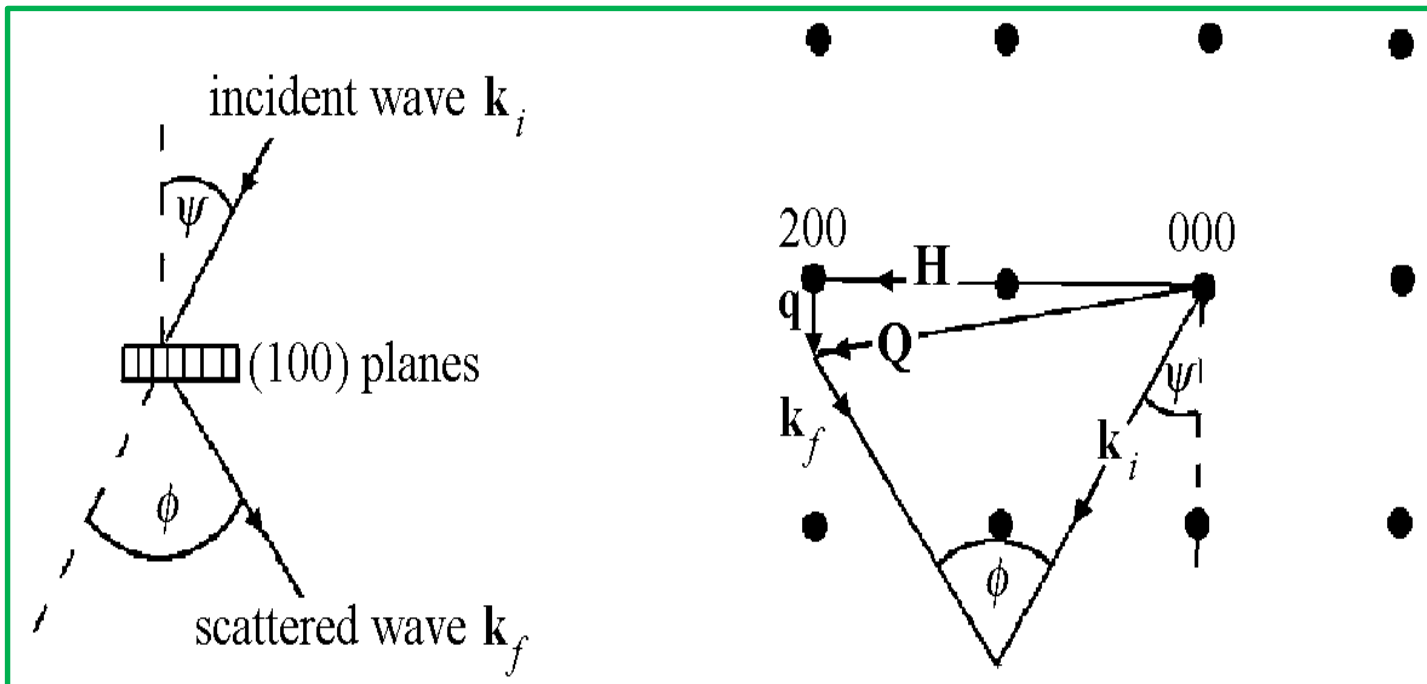
- Momentum transfer $\quad \hbar\mathbf{Q} = \hbar(\mathbf{k_i} - \mathbf{k_f})$

- Energy transfer $\quad \hbar\omega = E_i - E_f = \dfrac{\hbar^2}{2m}(k_i^2 - k_f^2)$

Real space   **Reciprocal space**

How do we measure these points?

- Momentum transfer $\quad \hbar\mathbf{Q} \;=\; \hbar(\mathbf{k_i} \;-\; \mathbf{k_f})$

- Energy transfer $\quad \hbar\omega \;=\; E_i \;-\; E_f \;=\; \dfrac{\hbar^2}{2m}\,(k_i^2 \;-\; k_f^2)$

There are basically two methods to do this:
- **Three (or Triple) Axis Spectrometry**
- **Time of Flight Spectrometry**

incident wave $\mathbf{k}_i$

ϕ

scattered wave $\mathbf{k}_f$

ϕ

Real space   **Reciprocal space**

How do we measure these points?

- Momentum transfer $\hbar\mathbf{Q} = \hbar(\mathbf{k}_i - \mathbf{k}_f)$

- Energy transfer $\hbar\omega = E_i - E_f = \dfrac{\hbar^2}{2m}(k_i^2 - k_f^2)$

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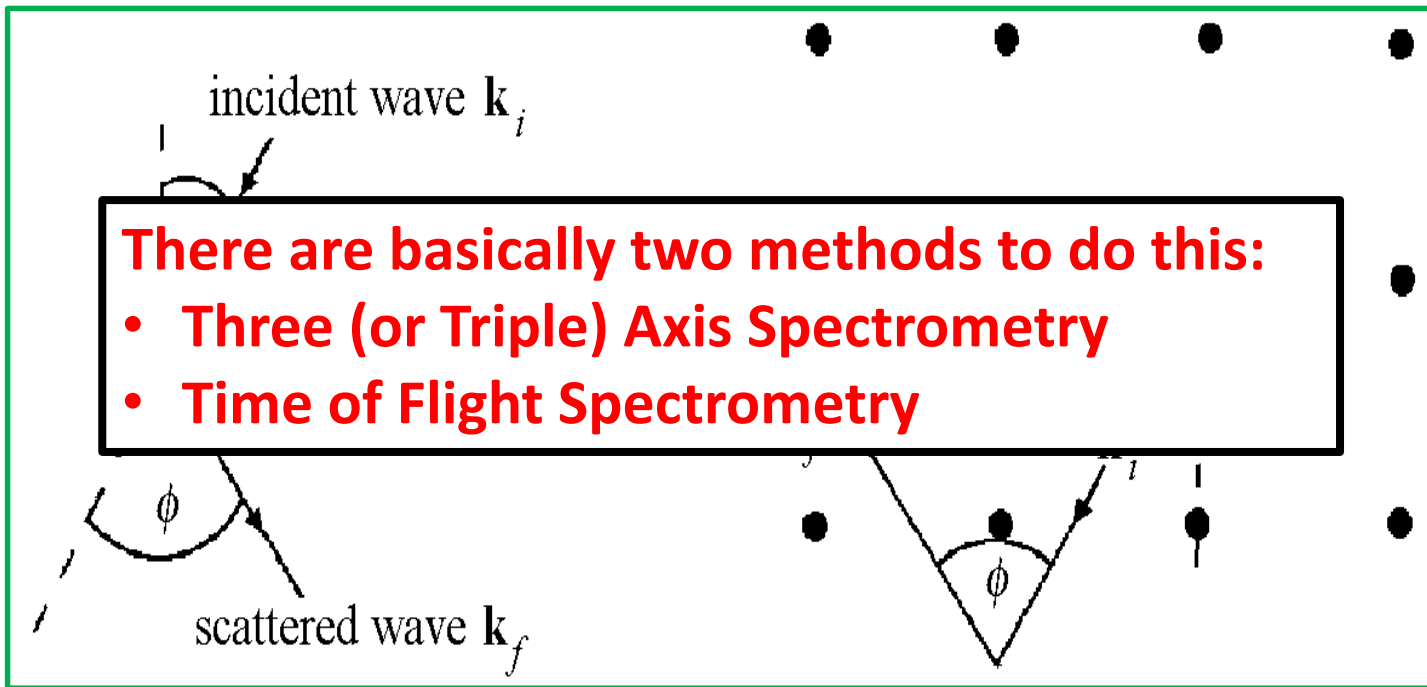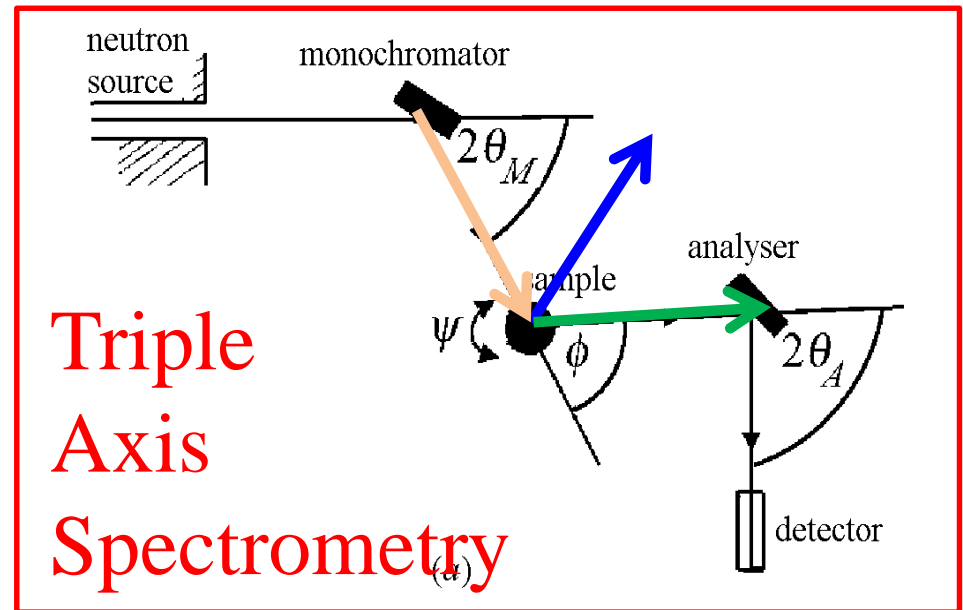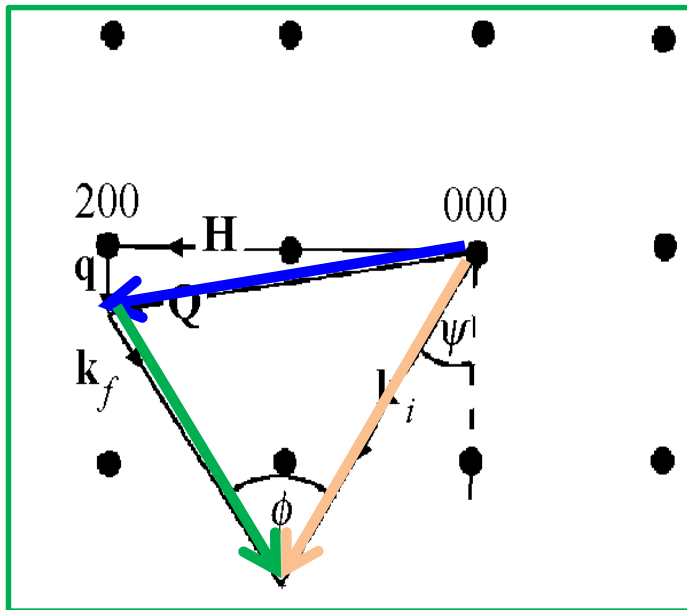
$$|\mathbf{Q}|^2 = |\boldsymbol{k_i}|^2 + |\boldsymbol{k_f}|^2 - 2\,|\boldsymbol{k_i}||\boldsymbol{k_f}|\cos(2\theta)$$

Bertram N. Brockhouse - Facts
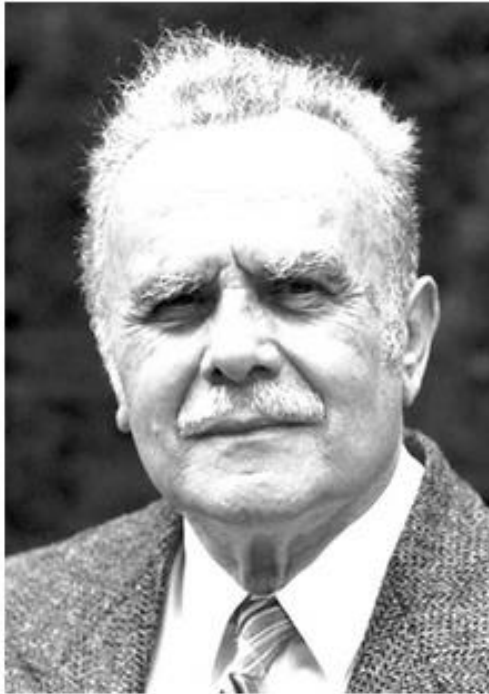
Bertram N. Brockhouse

Born: 15 July 1918, Lethbridge, Alberta, Canada

Died: 13 October 2003, Hamilton, Ontario, Canada

Affiliation at the time of the award: McMaster University, Hamilton, Ontario, Canada

Prize motivation: "for the development of neutron spectroscopy"
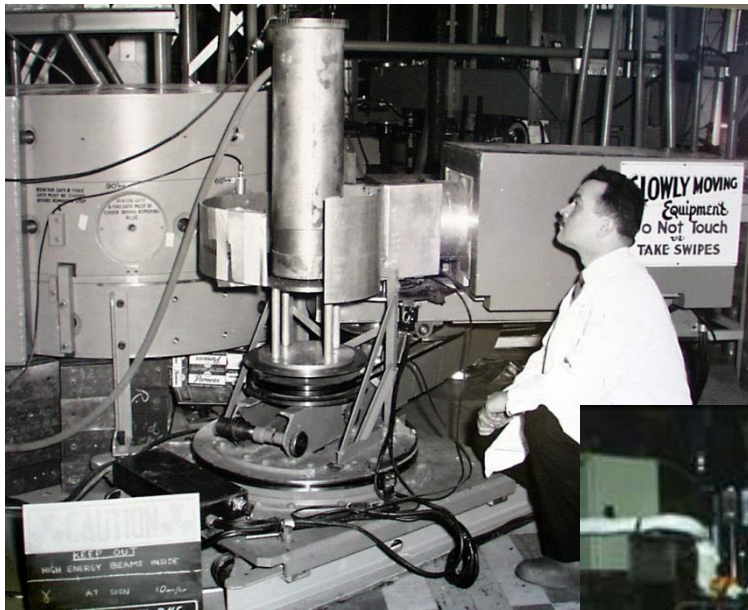
Field: Condensed matter physics, instrumentation

Bertram N. Brockhouse. *Nobelprize.org.* Nobel Media AB 2013.

Bertram N. Brockhouse - Facts
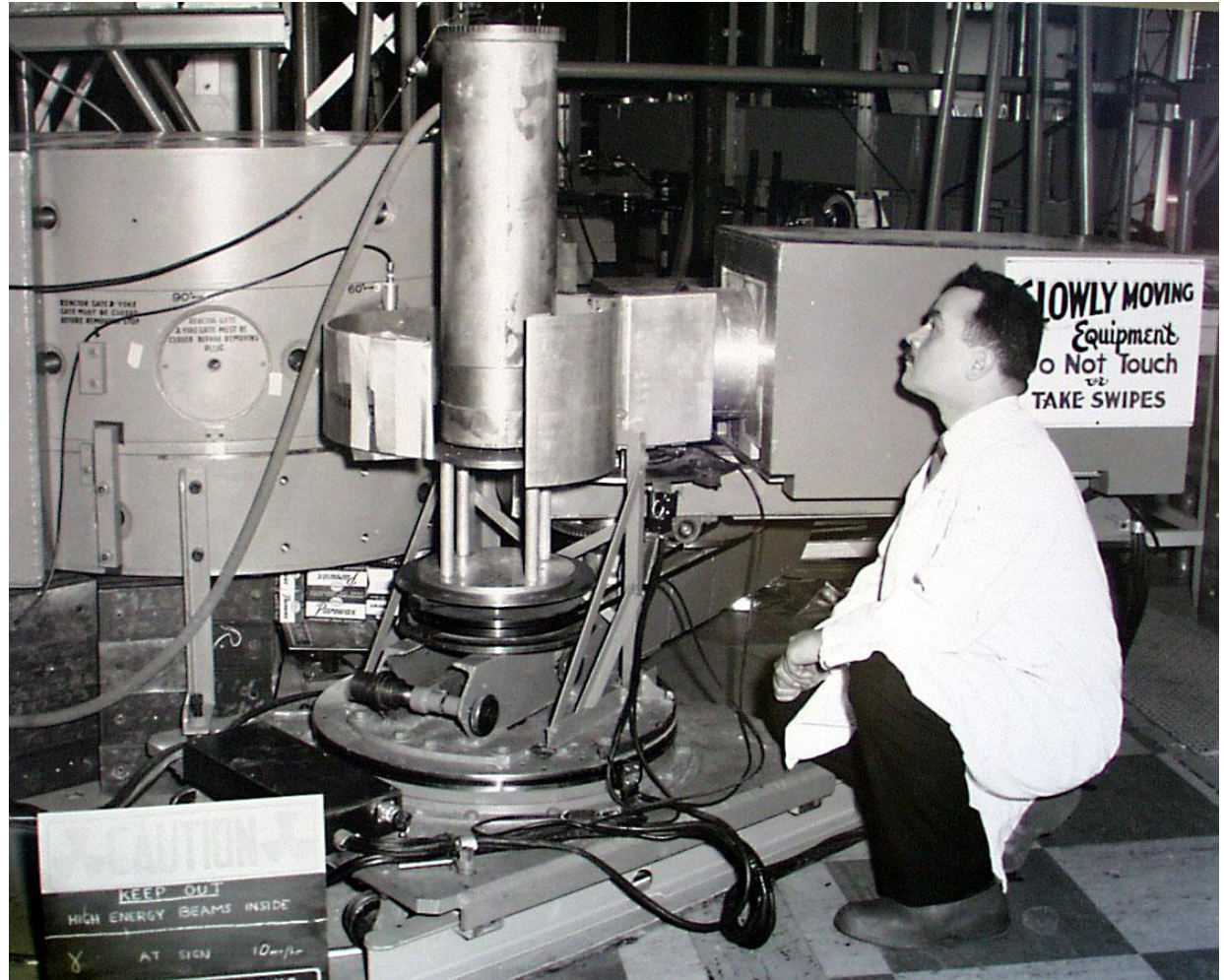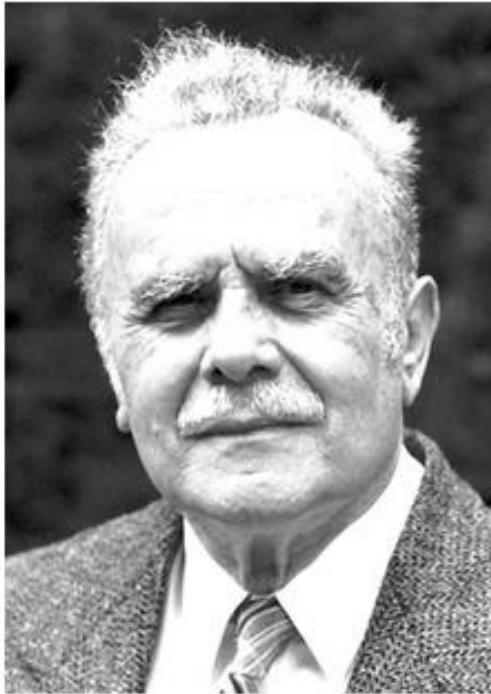
Source: Atomic Energy of Canada Limited, Chalk River, Ontario (CC BY-NC-ND 2.0)
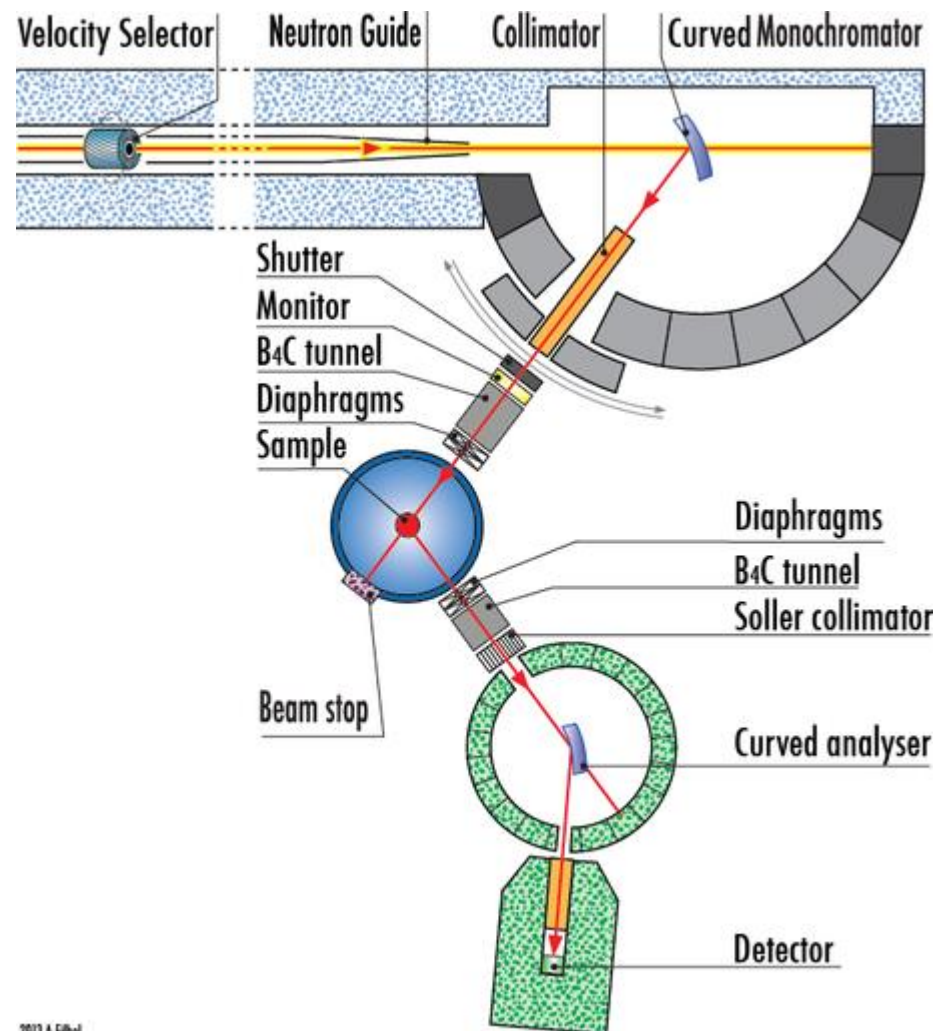
Bertram N. Brockhouse. *Nobelprize.org.* Nobel Media AB 2013.

Source: Atomic Energy of Canada Limited, Chalk River, Ontario

IN12, ILL

Velocity Selector   Neutron Guide   Collimator   Curved Monochromator

Shutter
Monitor
B_4C tunnel
Diaphragms
Sample

Diaphragms
B_4C tunnel
Soller collimator

Beam stop

Curved analyser

Detector

2013 A.Filhol

vTAS – a virtual TAS
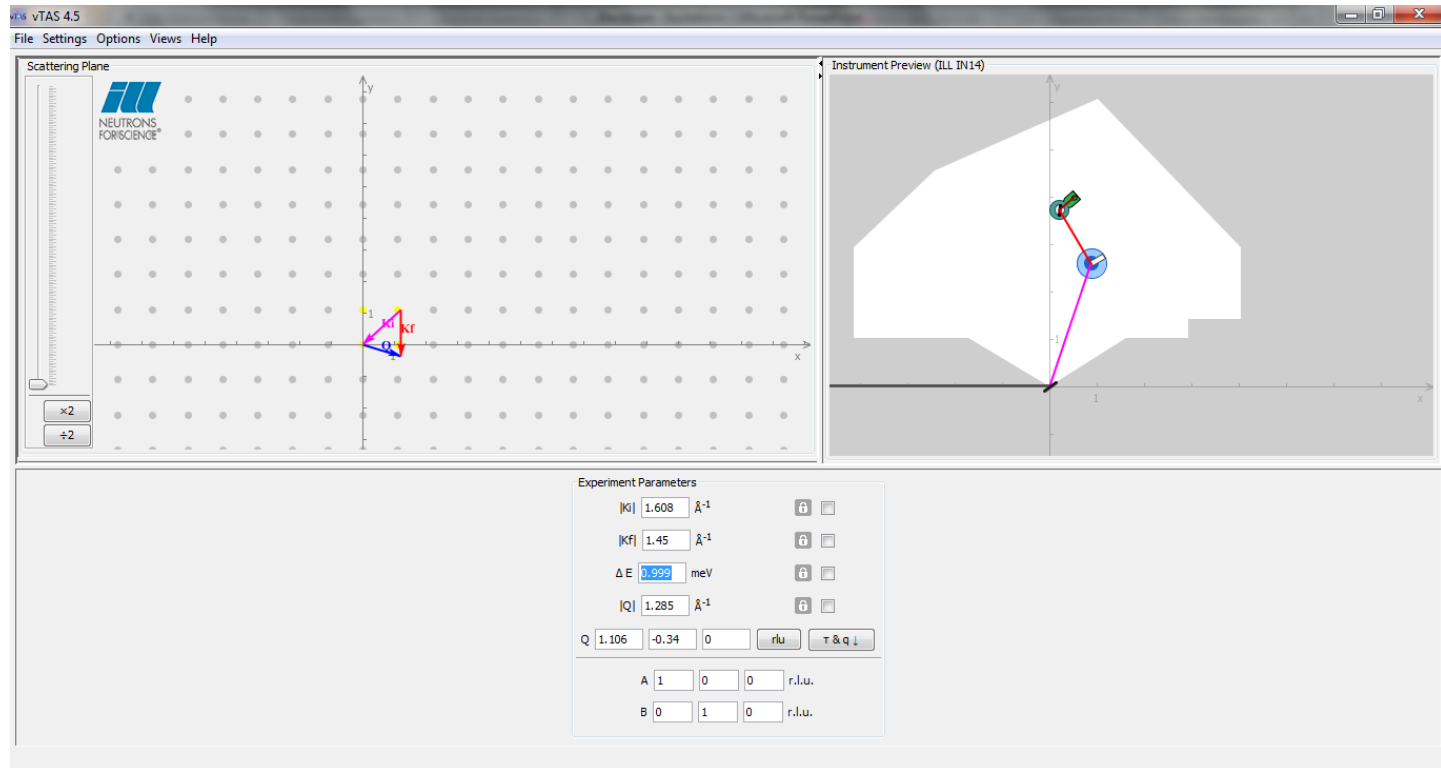
Experimental Choices: Monochromator and Analyser
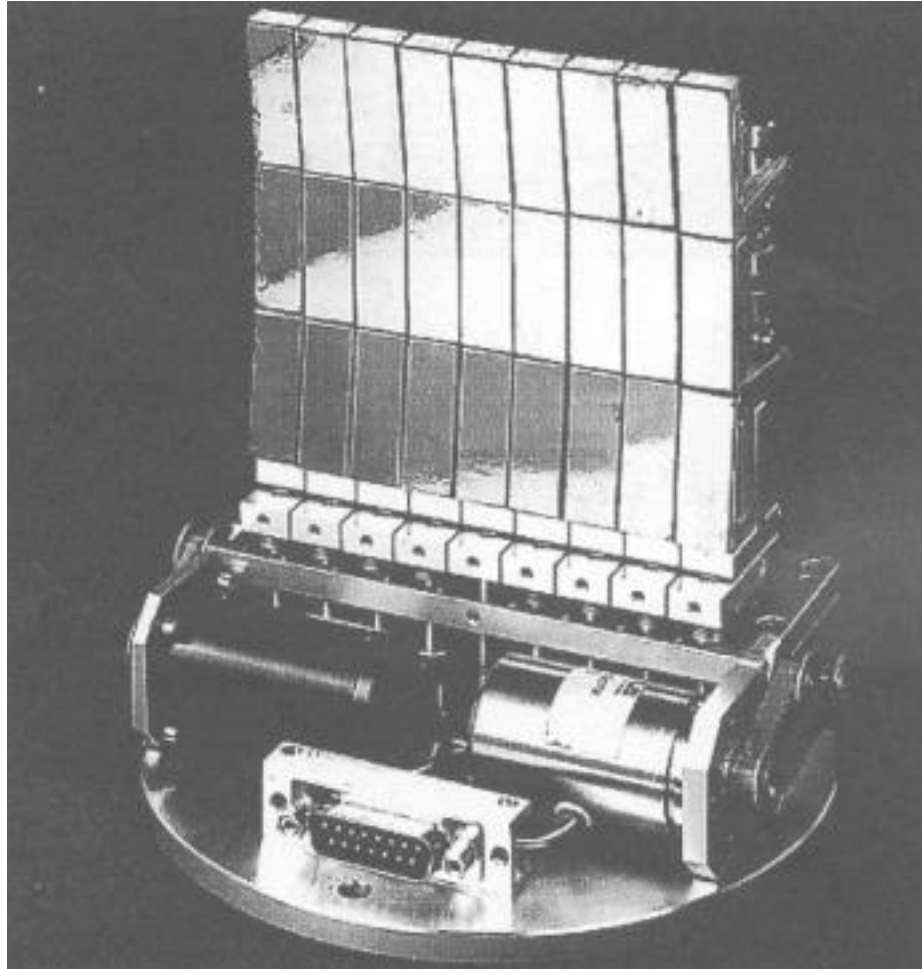
Experimental Choices: Monochromator and Analyser

Material	reflection	d-spacing [Å]	E-range [meV]	comments
PG	(002)	3.3539	42-3.6	high reflectivity
PG	(004)	1.6770	168-14.5	"
Cu	(200)	1.8075	145-12.5	used for high energies
Cu	(220)	1.27813	290-25	"
Si	(111)	3.13543	48-4.2	absence of second order
Si	(220)	1.92005	128-11.1	"
Si	(311)	1.63742	176-15.3	"
Si	(511)	1.04514	433-37.4	"
Ge	(111)	3.26651	44-3.8	absence of second order
Ge	(311)	1.70588	163-14.1	"
Be	(002)	1.79035	148-12.8	

Experimental Choices: Collimation

Why collimate?

> cut down beam divergence
> make sure you're looking at sample

Soller collimators

A set of parallel (neutron absorbing) plates (divergence distribution is triangular)

For TAS, about 10' to 80' FWHM

Experimental Choices: Filters

Why filter?

cut out higher order wavelengths

Which filter?

Be filters for cold neutrons (need to be kept at liquid nitrogen temperatures)

PG filters for thermal neutrons (might need rotating!)

Where?

Constant k_f

after the sample

Constant k_i

before the sample

Frikkee, Nucl. Inst. Meth. 125, 307 (1975) – studied wavelength range 1.12 – 4.25 Å.

Experimental Choices: Resolution

Q and ω only defined to a certain level of precision

Bragg's law: $n\lambda = 2d\sin\theta$

Reducing these uncertainties leads to a **better** resolution

$\lambda > 0$ and λ are coupled

BUT

k_i and k_f have their own

It also leads to **lower** counts resolution volumes, with distinct orientations in **Q**-ω space.

→ resolution ellipsoid.

Experimental Choices: The resolution ellipsoid

Shape and **size** depend upon:
Collimation and crystal mosaic of monochromator and analyzer

Orientation depends upon:
Sense of scattering at the monochromator, sample and analyzer

Focussing can also have an effect

Tune your experiment to give you what you want
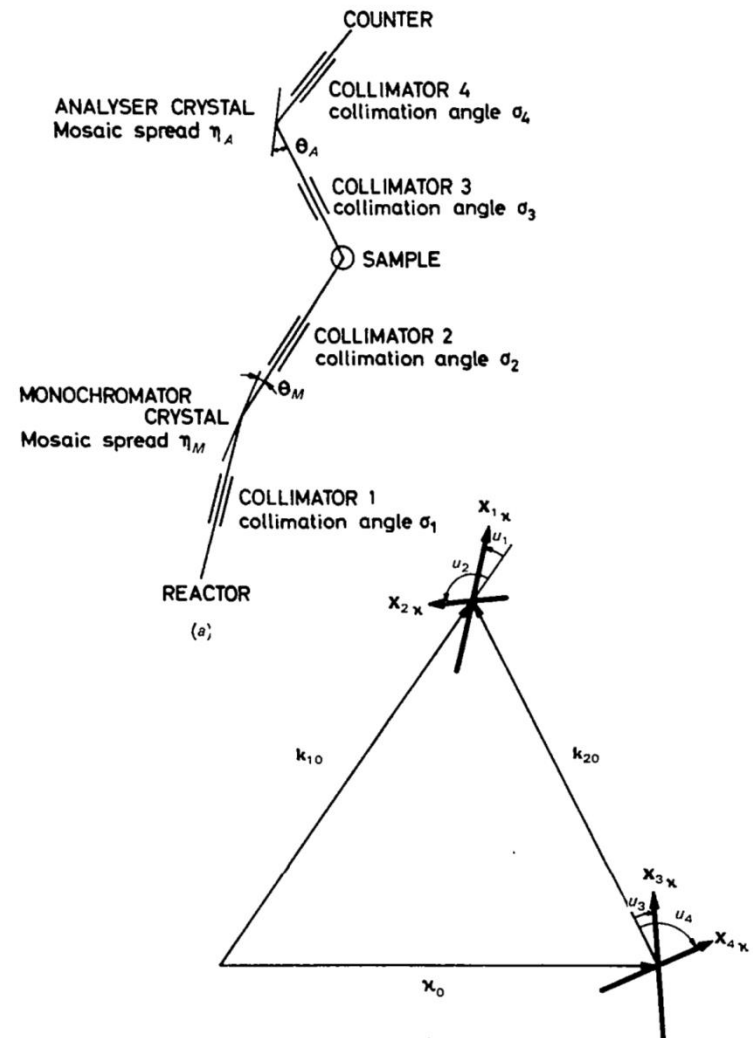
Experimental Choices:
The resolution ellipsoid

Tune your experiment to give you what you want
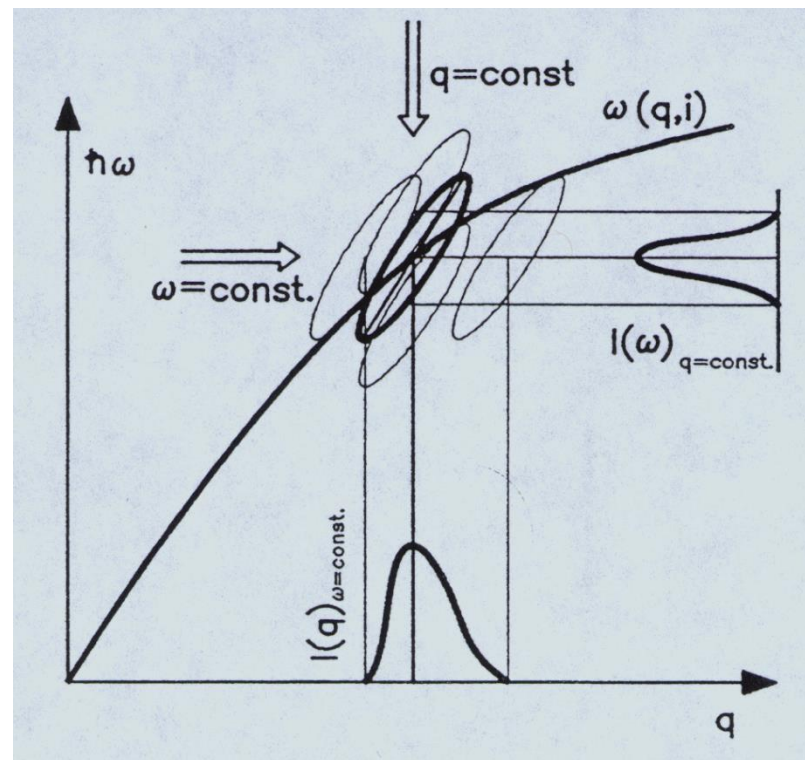
References:

RESTRAX – Saroun and Kulda, http://neutron.ujf.cas.cz/restrax/doc/index.html

ResLib – Zheludev, http://www.neutron.ethz.ch/research/resources/reslib

Cooper and Nathans, Acta Cryst 23, 357 (1967)

Popovici *et al.*, J. Appl. Cryst. 20, 90 (1987).

Experimental Choices: Focussing

Vertical focussing opens up Q resolution in the vertical direction (out of the scattering plane)
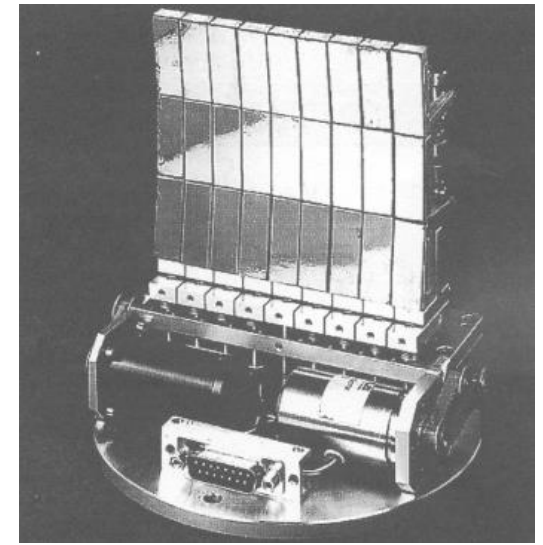
Focussing distance depends upon Bragg angle so the radius, R, of the mono/analyzer crystal needs to be variable.

L_0 = source to mono
L_1 = mono to sample

$$\frac{1}{L_0} + \frac{1}{L_1} = \frac{2\sin\theta_{Bragg}}{R} \quad ; \quad h_{image} = h_{source}\frac{L_1}{L_0}$$

Currat, Nucl. Inst. Meth. 107, 21 (1973)

Experimental Choices: Focussing

Horizontal focussing

messes up Bragg conditions at the monochromator and analyzer, but increases intensity

Affects **Q** resolution in the scattering plane, and the energy resolution
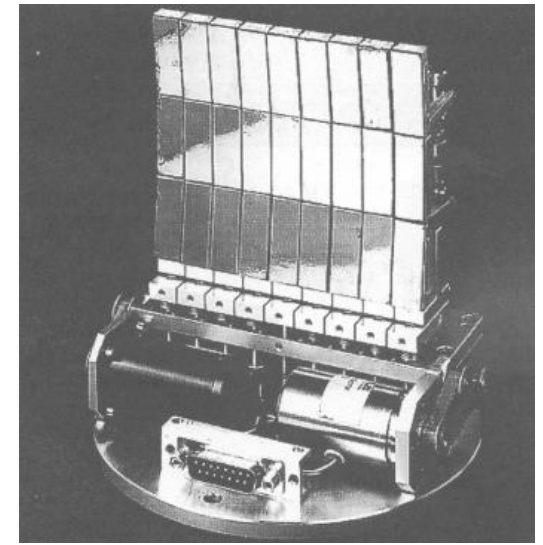
Broholm, Nucl. Inst. Meth. A367, 169 (1996)

Beware: Spurions

Chasing ghosts in reciprocal space—a novel inelastic neutron multiple scattering process

H.M. Rønnow[a,b,*], L.-P. Regnault[b], J.E. Lorenzo[c]

[a] NEC Laboratories, Princeton and University of Chicago, USA
[b] MDN/SPSMS/DRFMC, CEA-Grenoble, 38054 Grenoble, France
[c] Laboratoire de Cristallographie, CNRS, 38042 Grenoble, France

Abstract

We have discovered that a recently reported weak excitation branch in the spin-Peierls material $CuGeO_3$ is in fact a ghost image of the primary magnetic excitation shifted in reciprocal space by a novel multiple scattering process. A model is developed that predicts the occurrence of such multiple scattering and accounts for the observations in $CuGeO_3$. New 'ghostons' can occur when the magnetic unit cell is smaller than the structural, while mixing of intensities from different reciprocal space zones jeopardize accurate polarisation analysis and the study of weak modes in general.

Beware: Spurions

- Bragg peaks from the sample holder/cryostat

- Incoherent scattering from the mono/analyzer

- Beam on to detector

- Phonons from mono/analyzer

Check temperature dependence
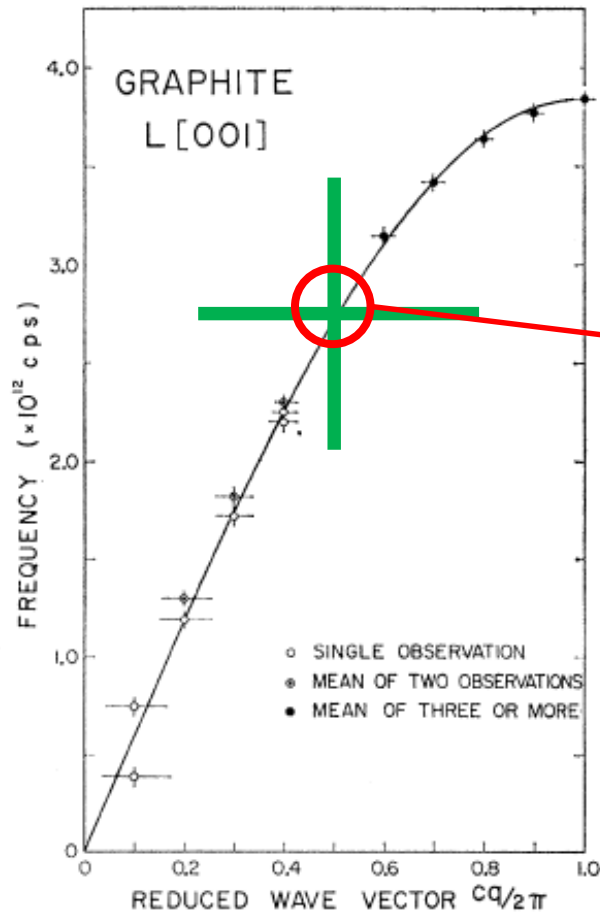Sample angle scans

Measuring a phonon spectrum
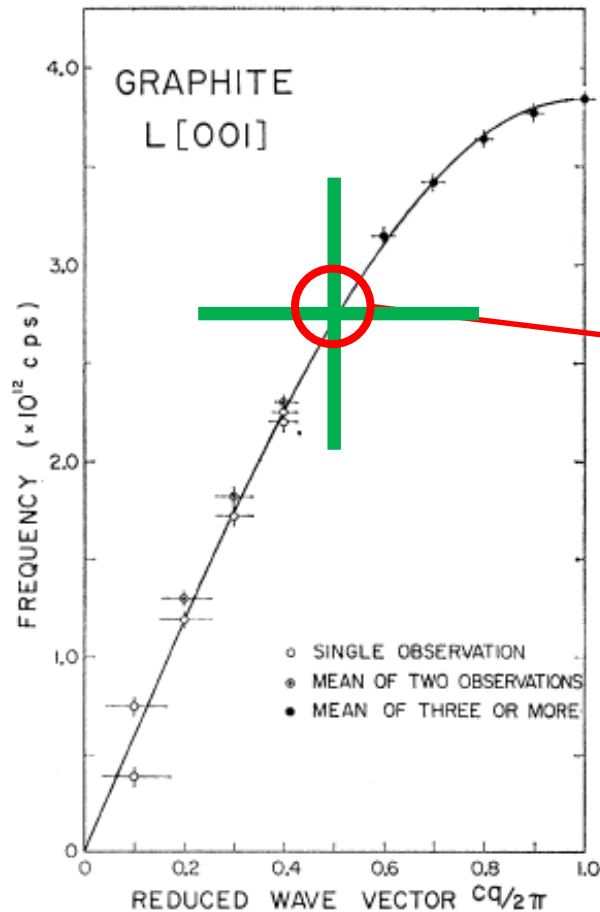
FIG. 3. Dispersion curve for the L[001] modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i} S(\mathbf{Q}, \omega)$$

Constant-**Q**
or
Constant-E ??

Fixed k_i or fixed k_f?

We normalised to the number of incident neutrons using a low efficiency monitor whose efficiency depends on the neutron velocity. In fixed k_i mode, this efficiency is fixed and we therefore measure signal per monitor

Measuring a phonon spectrum

FIG. 3. Dispersion curve for the L[001] modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

IN14, #4-03-1306
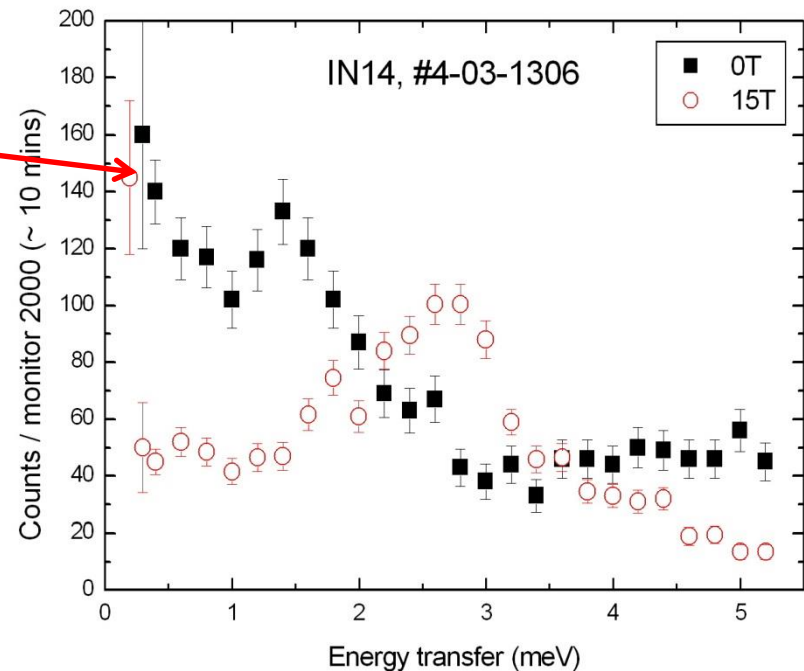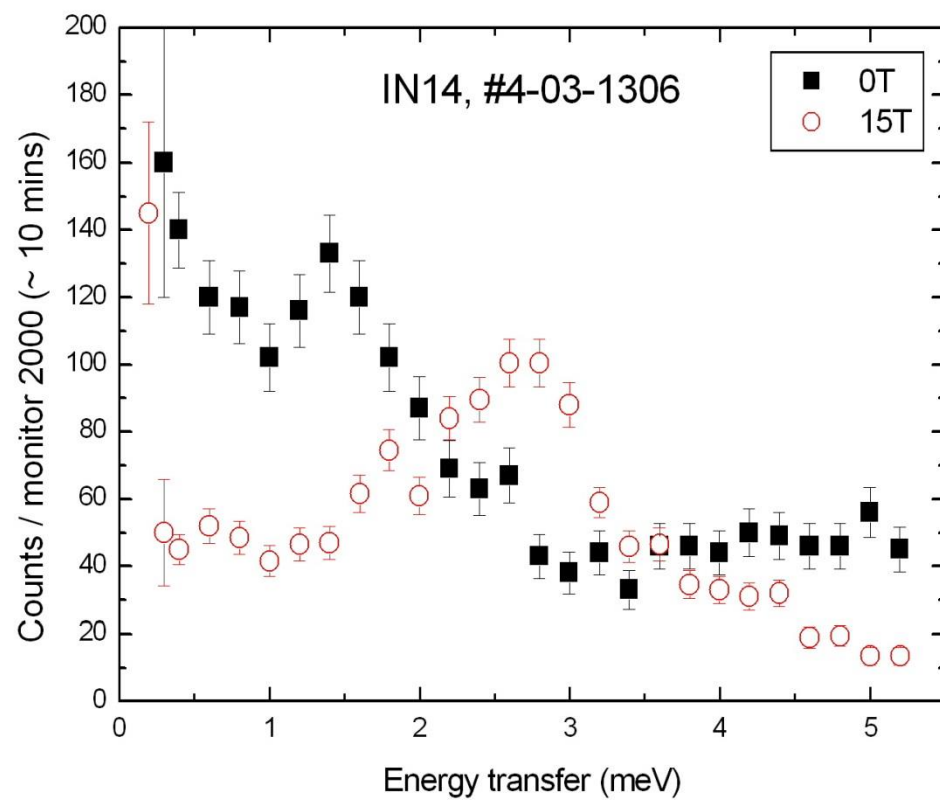
Advantages and disadvantages of the triple-axis method

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-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 eminently possible that something important might be missed

Superfluid helium

TAS

TOF
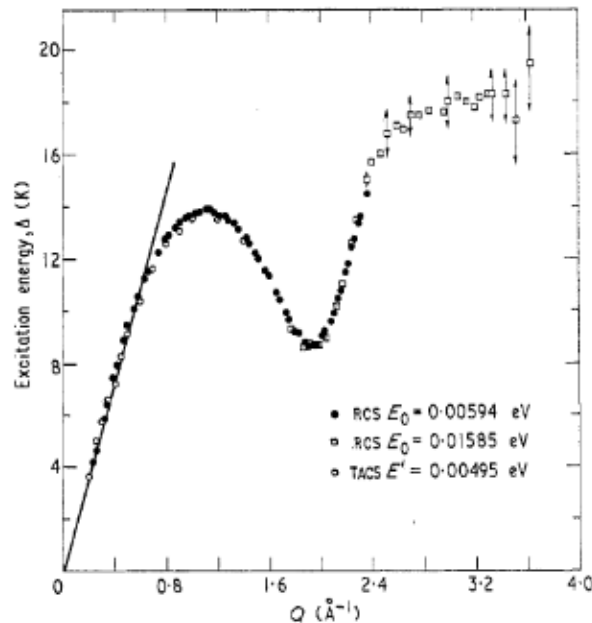
Cowley and Woods., Can. J. Phys. 49, 177 (1971)

Blackburn *et al.*, Pramana 71, 673 (2008)

Time-of-flight spectroscopy

Direct geometry:
fix k_i by chopper phasing
scan through k_f by time-of-flight
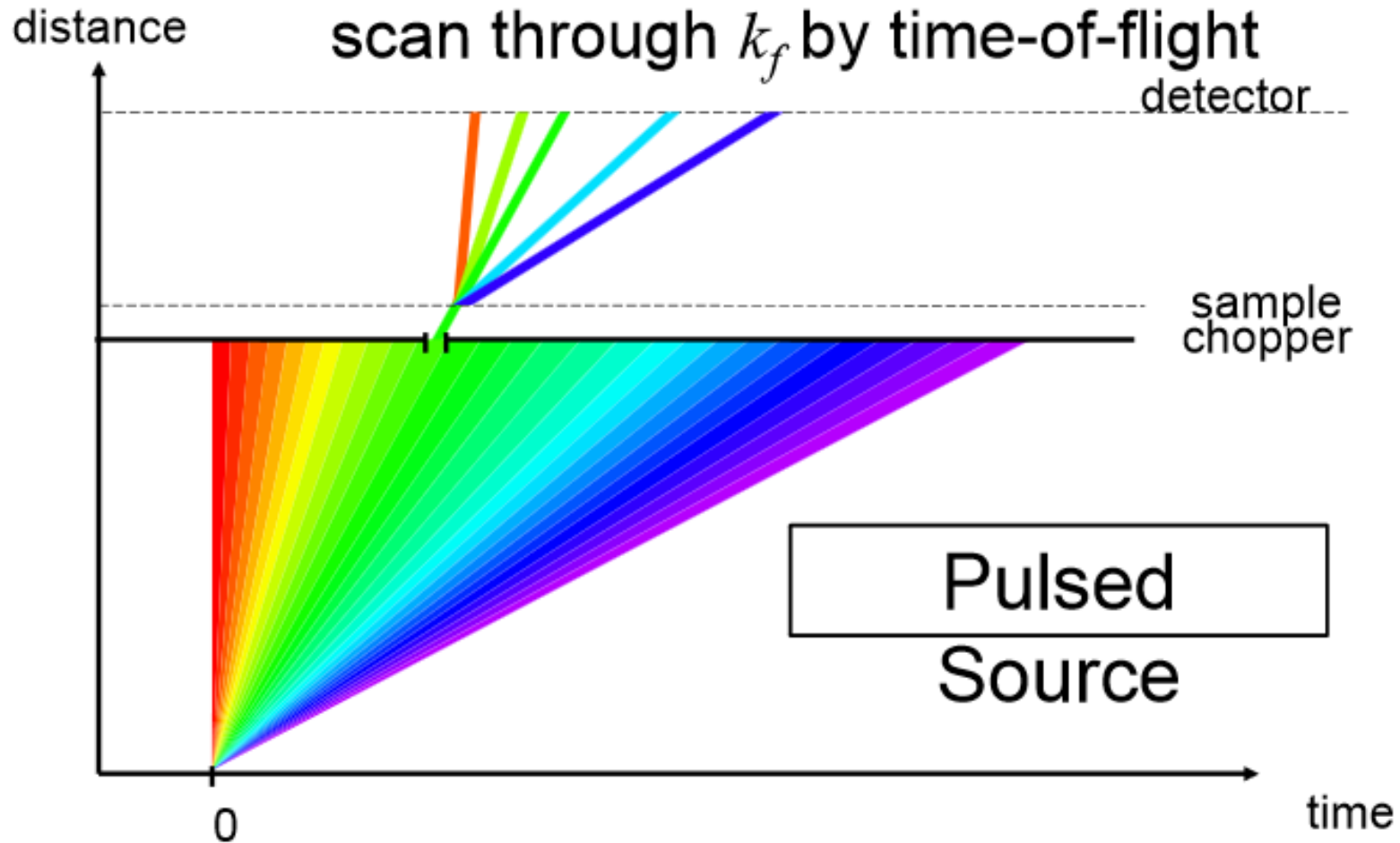
distance

detector

sample
chopper

Pulsed
Source

0

time

from Ken Andersen

Time-of-flight spectroscopy

LET, ISIS

Direct geometry:
fix k_i by chopper phasing
scan through k_f by time-of-flight

distance

detector

sample
chopper

Pulsed
Source

0
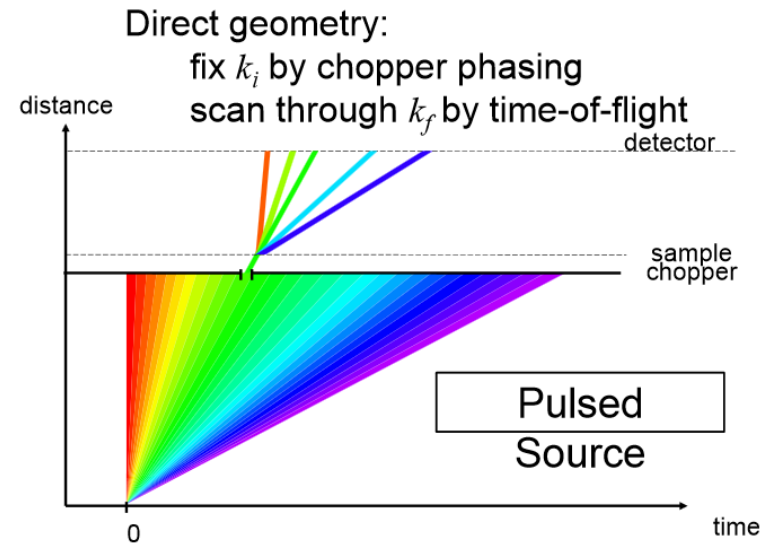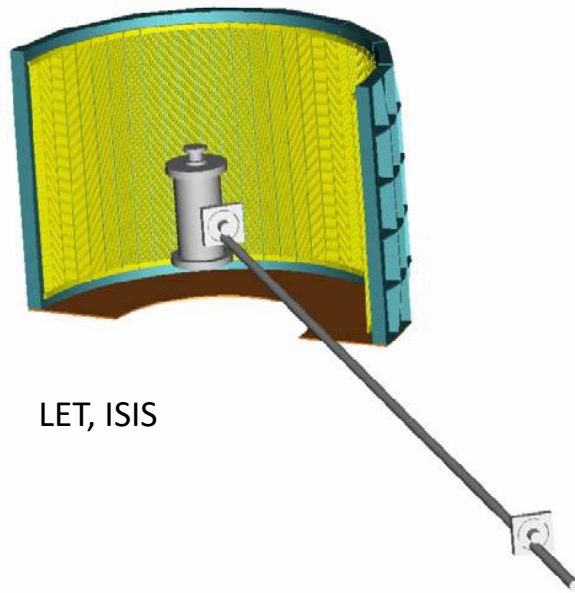
time

$$\text{final velocity} = \frac{\text{sample-detector distance}}{\text{time to detector} - \text{time to sample}}$$

Time-of-flight spectroscopy

IN5, ILL

Direct geometry:
fix k_i by chopper phasing
scan through k_f by time-of-flight
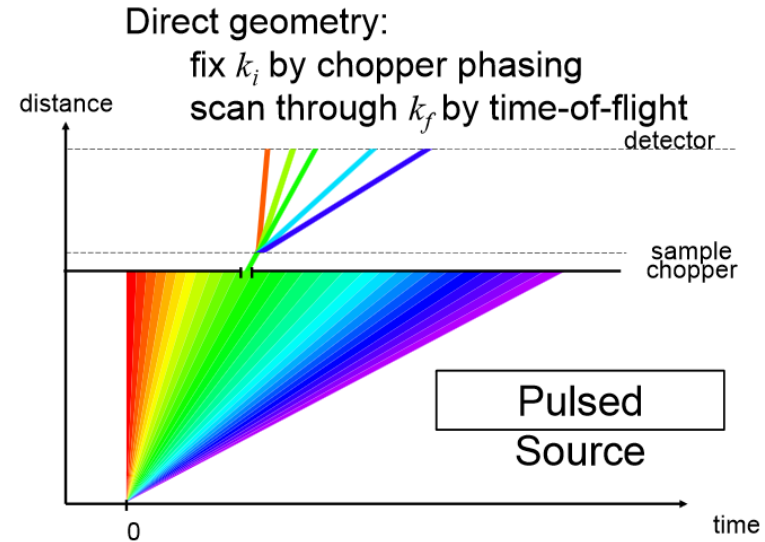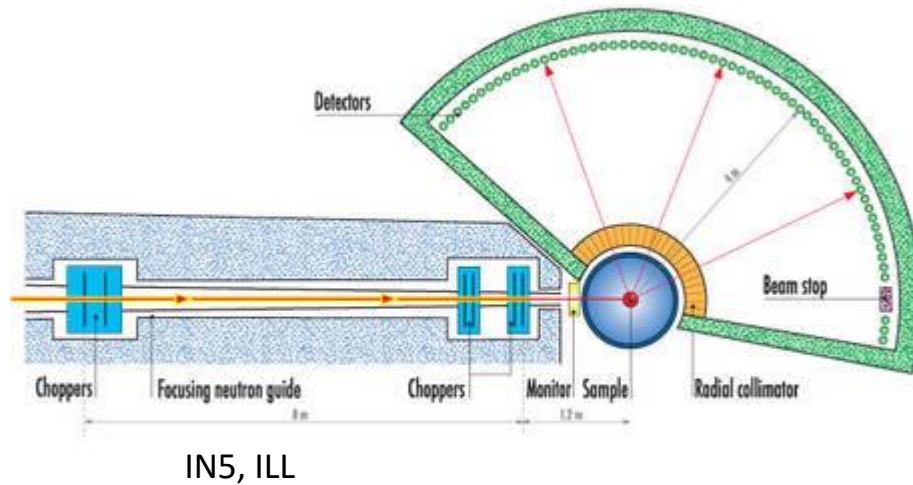
$$\text{final velocity} = \frac{\text{sample-detector distance}}{\text{time to detector} - \text{time to sample}}$$

This gives us the neutron's final energy

Time-of-flight spectroscopy

Measured quantity:

$$I(2\theta, t_D) \rightarrow S(\mathbf{Q}, \omega)$$

Remember the bin sizes:
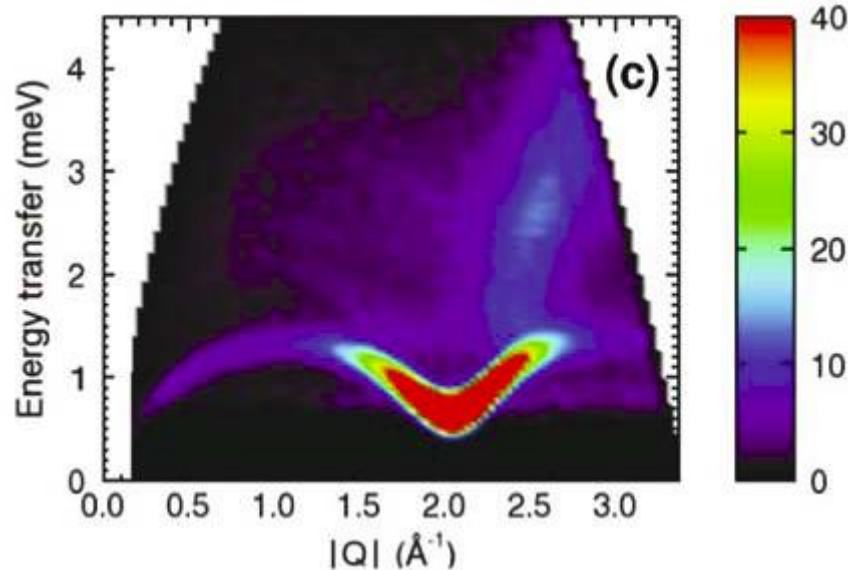
Time-of-flight spectroscopy

$$|\mathbf{Q}|^2 = |\mathbf{k}_i|^2 + |\mathbf{k}_f|^2 - 2\,|\mathbf{k}_i||\mathbf{k}_f|\cos(2\theta)$$
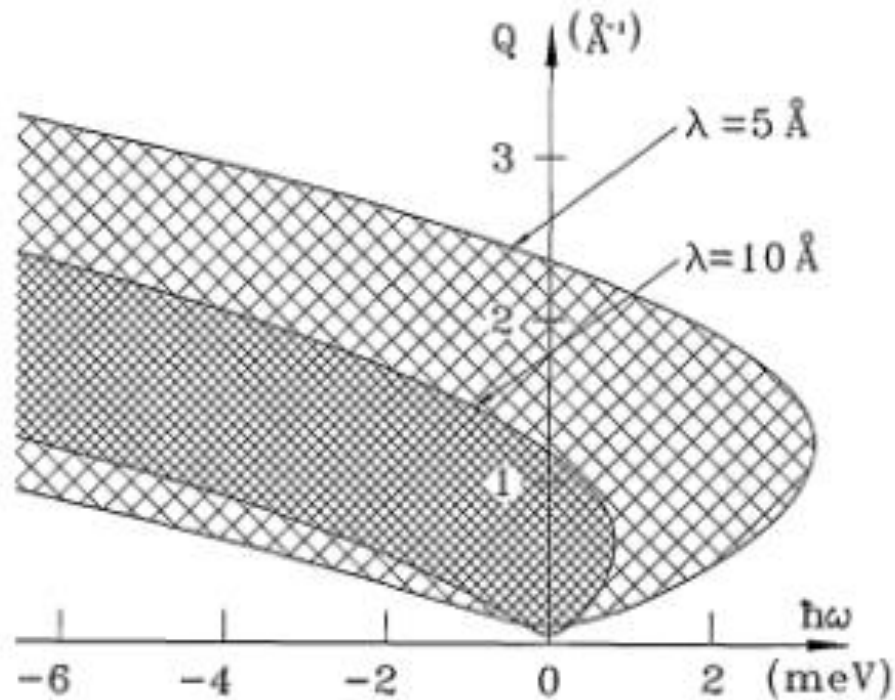
Fig. 17. Plots of the accessible region in (Q, ω) space for neutrons of wavelength 5 and 10 Å (energy 3.272 and 0.818 meV, respectively). The minimum and maximum scattering angles are 5° and 140°. There is no (theoretical) limit to the energy transfer in neutron energy gain.

Copley and Udovic., J. Res. NIST 98, 71 (1993)

Time-of-flight spectroscopy

$$\frac{\hbar^2 Q^2}{2m} = 2E_0 - \hbar\omega - 2\sqrt{E_0(E_0 - \hbar\omega)}\cos(2\theta)$$

Fig. 17. Plots of the accessible region in (Q,ω) space for neutrons of wavelength 5 and 10 Å (energy 3.272 and 0.818 meV, respectively). The minimum and maximum scattering angles are 5° and 140°. There is no (theoretical) limit to the energy transfer in neutron energy gain.

Copley and Udovic., J. Res. NIST 98, 71 (1993)

Experimental choices: Intensity vs Resolution

What can we change?

incident wavelength
pulse width at sample chopper
frame overlap ratio

DCS, NCNR, NIST

Experimental choices: Intensity vs Resolution

What can we change?

incident wavelength
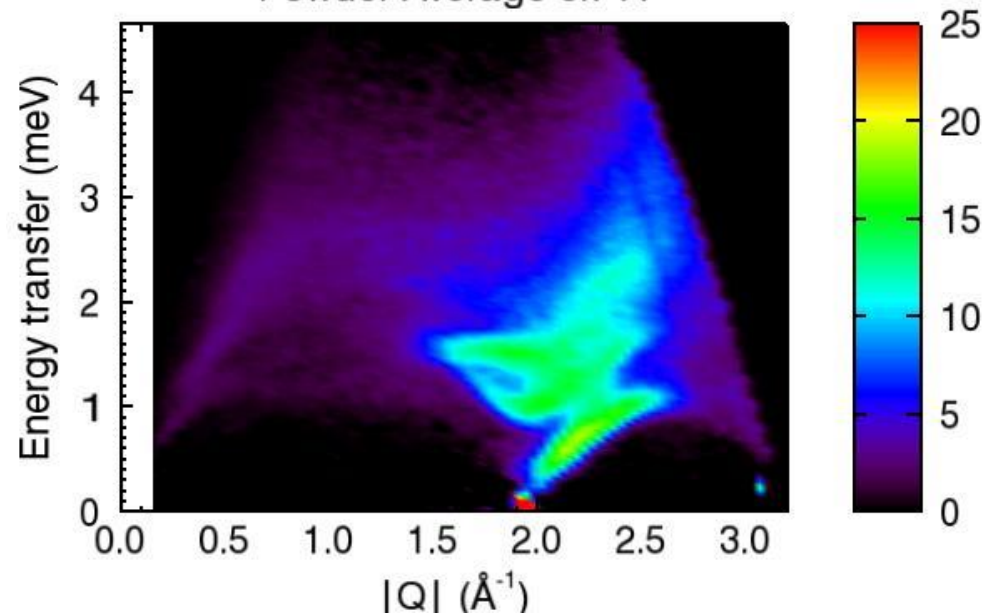pulse width at sample chopper
frame overlap ratio

DCS, NCNR, NIST

Experimental choices: Intensity vs Resolution

What can we change?

incident wavelength
pulse width at sample chopper
frame overlap ratio

DCS, NCNR, NIST

TOF on single crystals

Superfluid helium
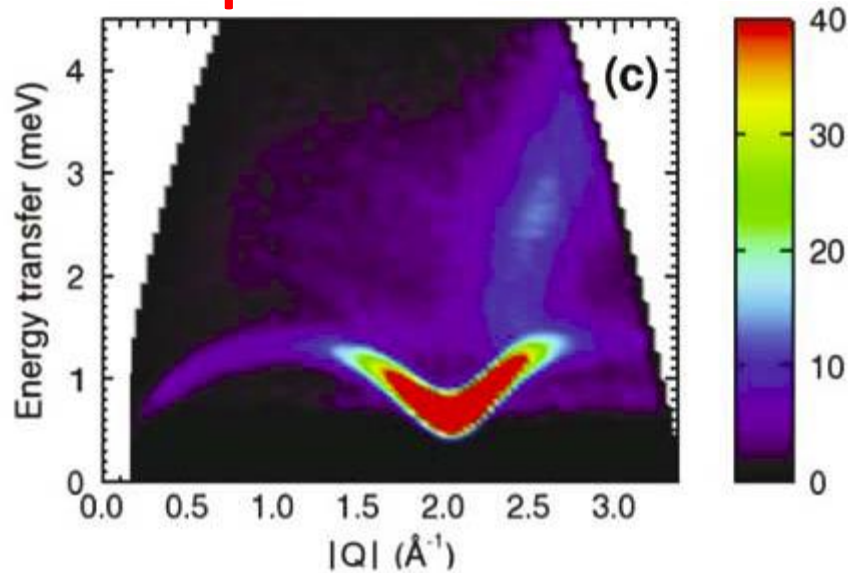
Solid helium

TOF on single crystals
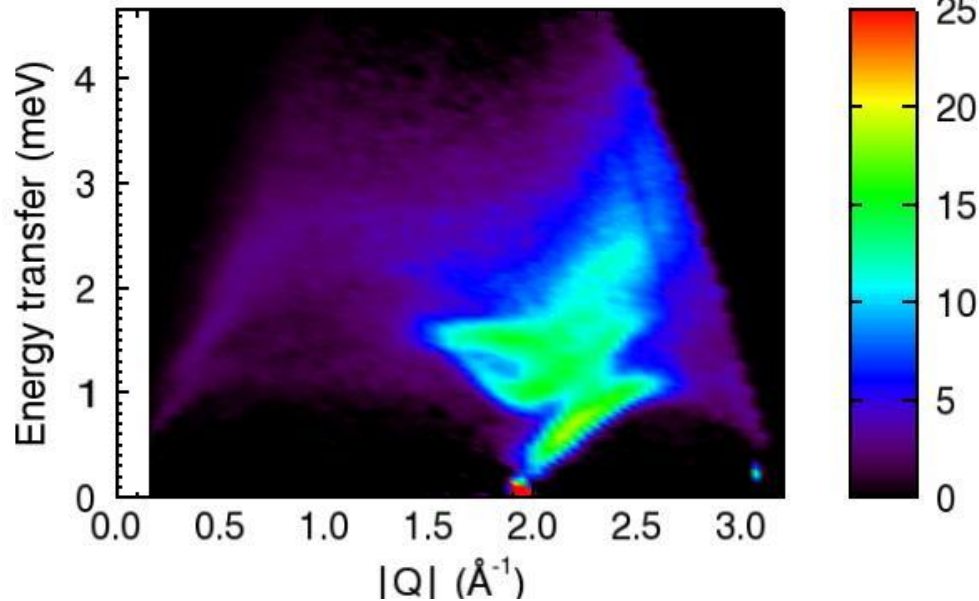
Solid helium
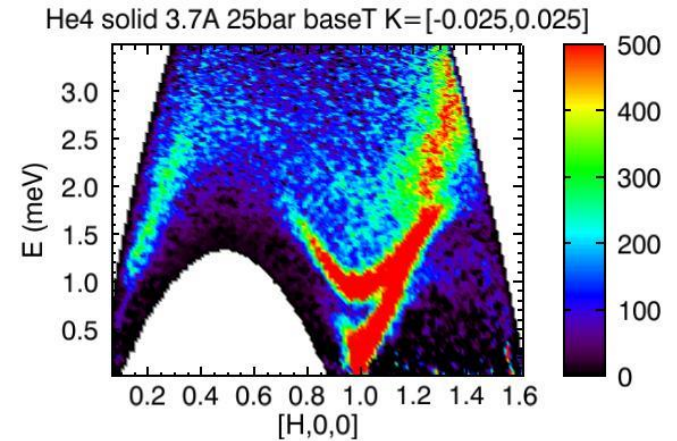
Angle 1

Angle 1 – 2 degrees

TOF on single crystals
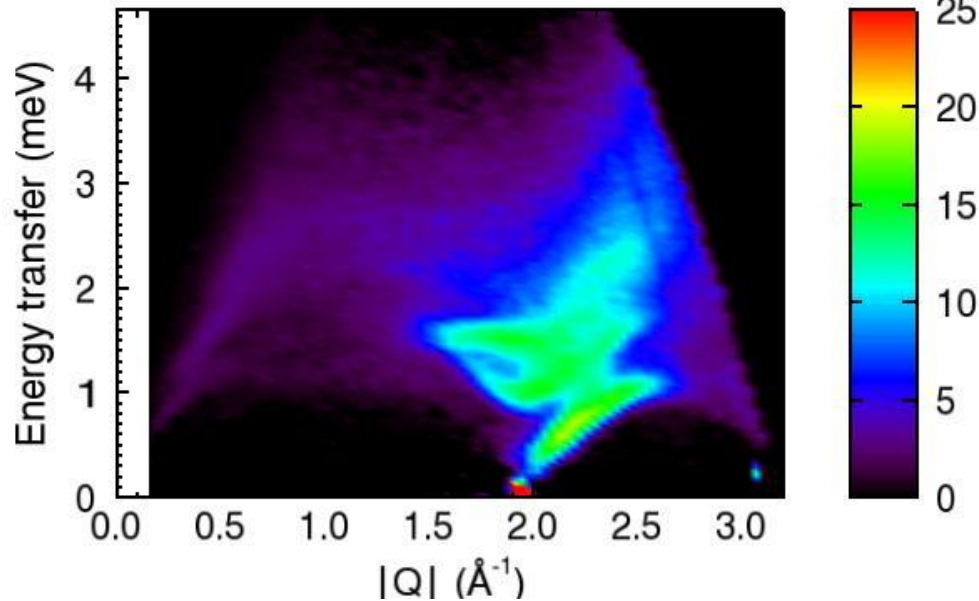
Solid helium

Longitudinal

Transverse

TOF on single crystals

HORACE screenshot – horace.isis.rl.ac.uk
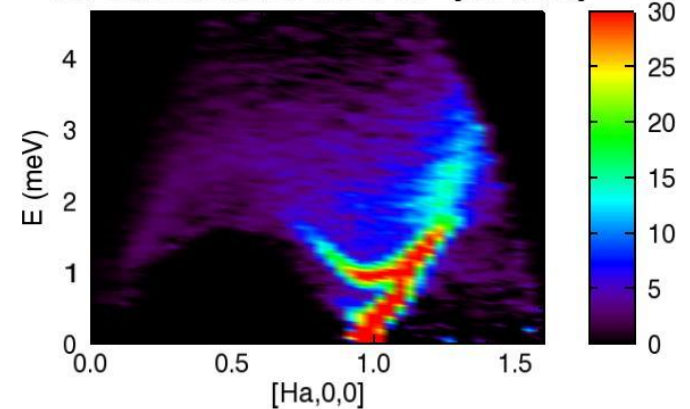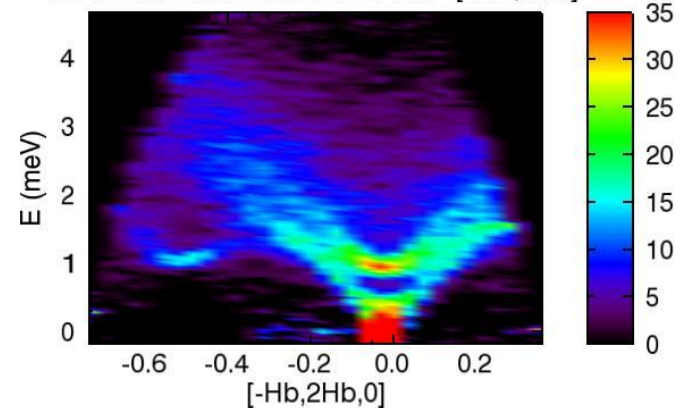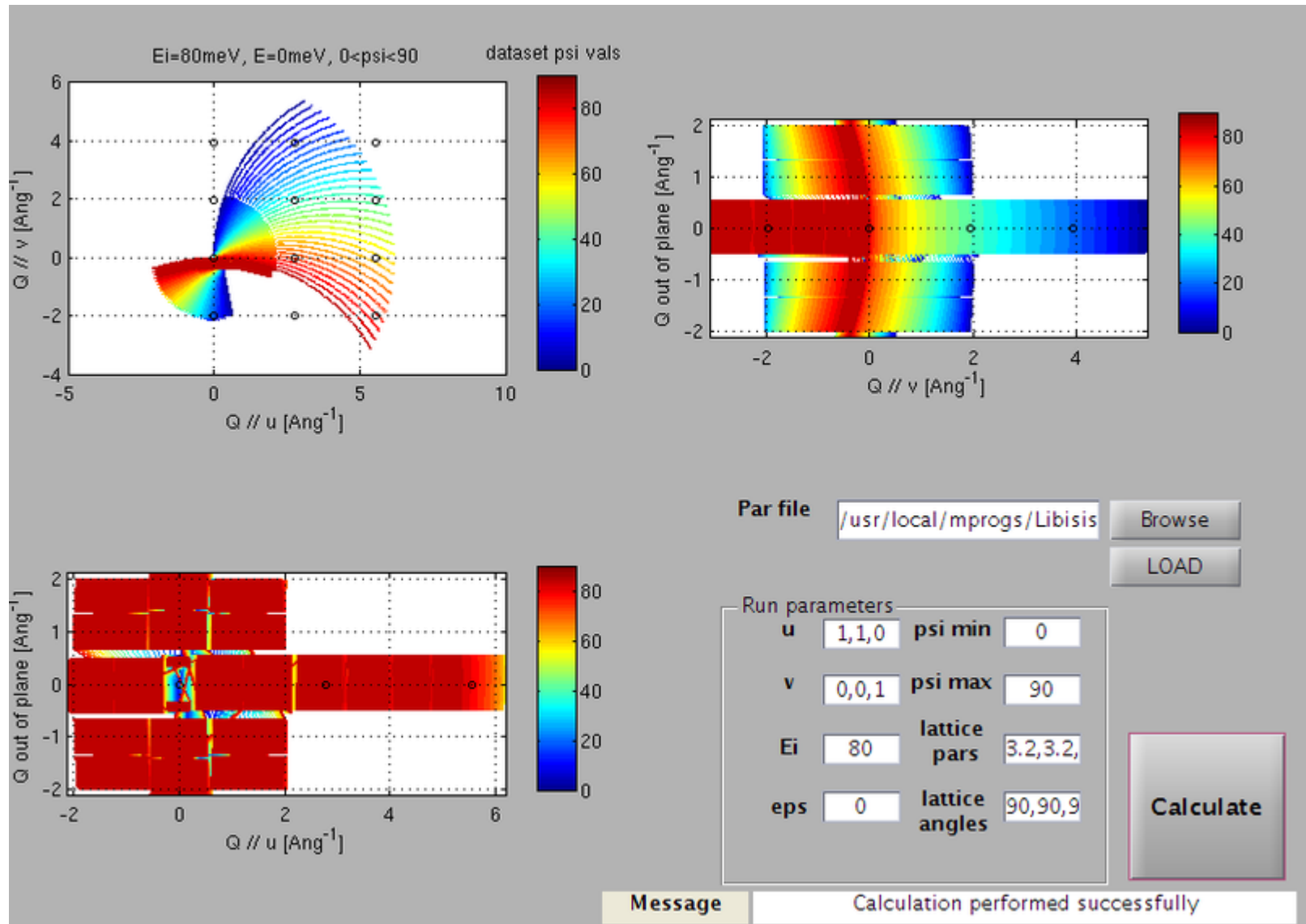
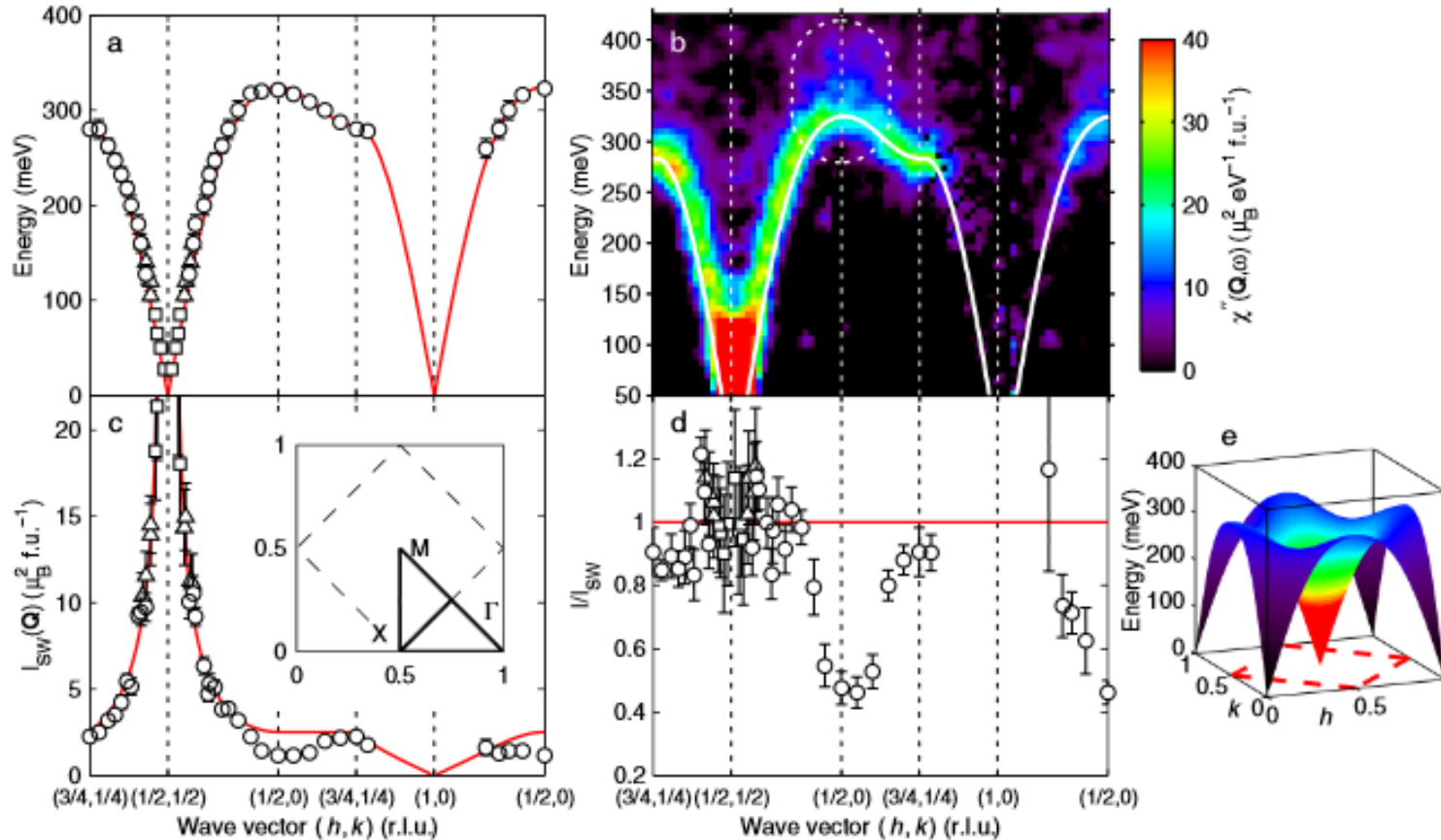# Complete dataset in **Q** and ω space

Headings *et al.,*

FIG. 2 (color online).   q dependence of the magnetic excitations in La_2CuO_4. (a) One-magnon dispersion ($T = 10$ K) along lines in (c, inset). Symbols indicate E_i: 160 meV (□), 240 meV (△), and 450 meV (○). The solid line is a SWT fit based on Eq. (1). (b) Measured $\chi''(q, \omega)$. Dashed circle highlights the anomalous scattering near (1/2, 0). An $\hbar\omega$-dependent background determined near (1, 0) has been subtracted. (c) One-magnon intensity. Line is a fit to SWT with renormalization factor $Z_d = 0.4 \pm 0.04$. (d) One-magnon intensity divided by SWT prediction. (e) SWT dispersion (color indicates SW intensity).
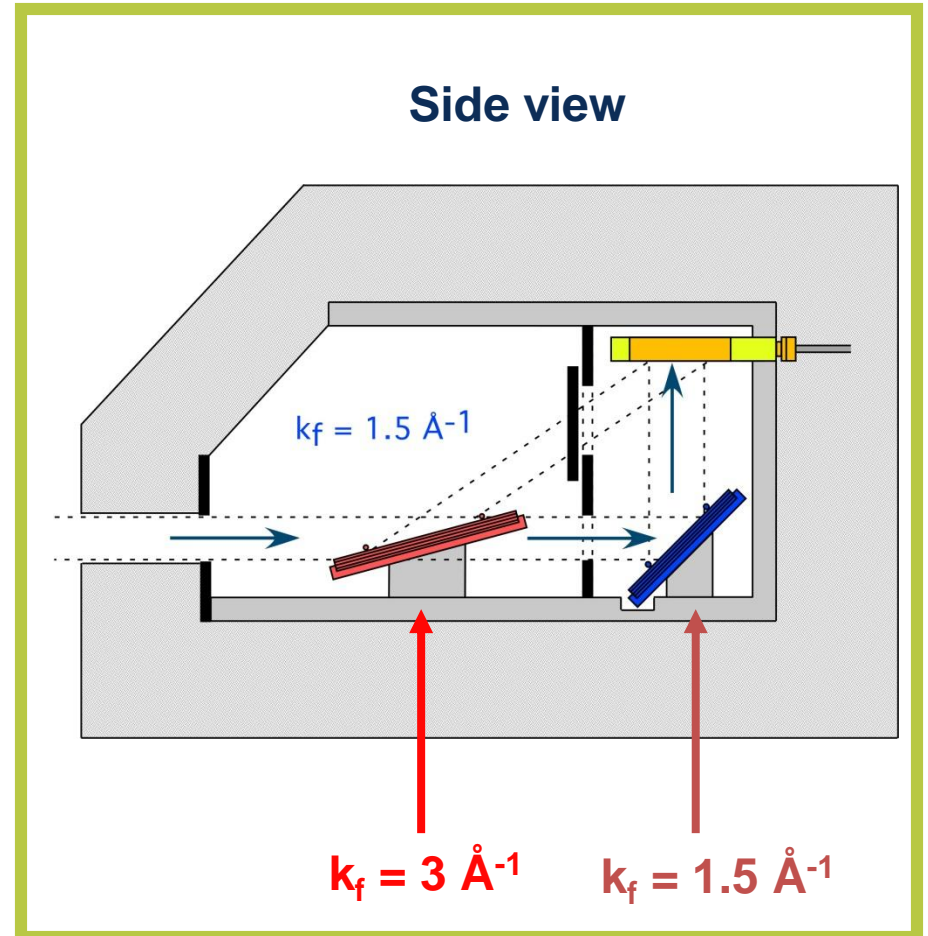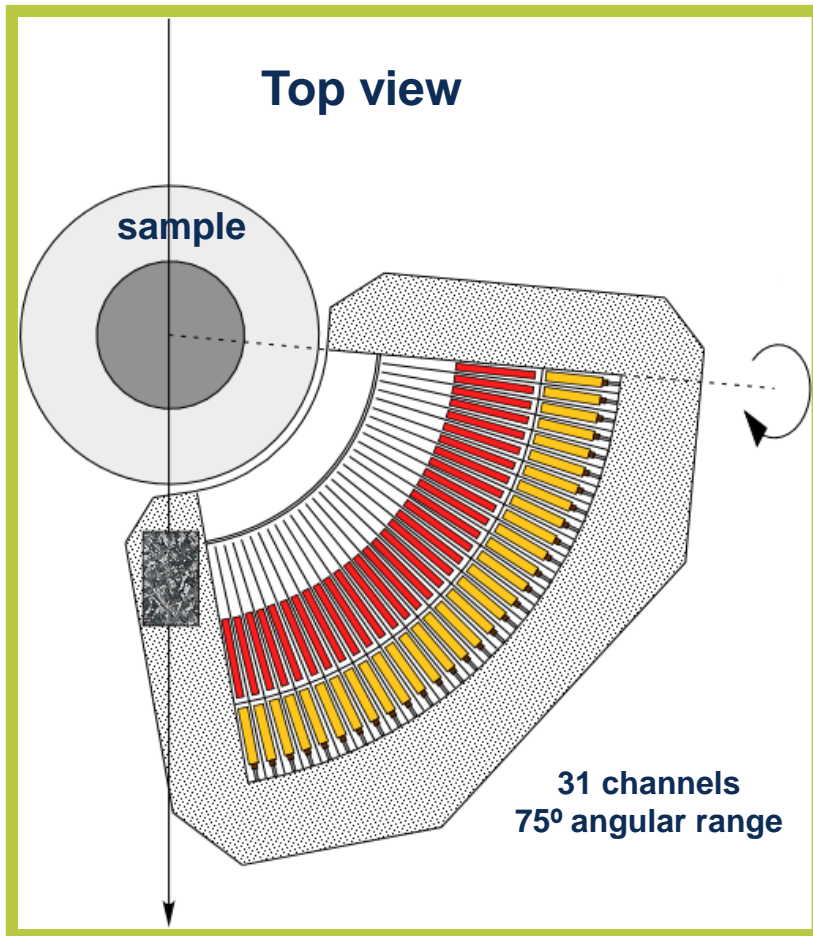
Excitations

Elizabeth Blackburn

University of Birmingham

- What are excitations?

- What can we learn from then?
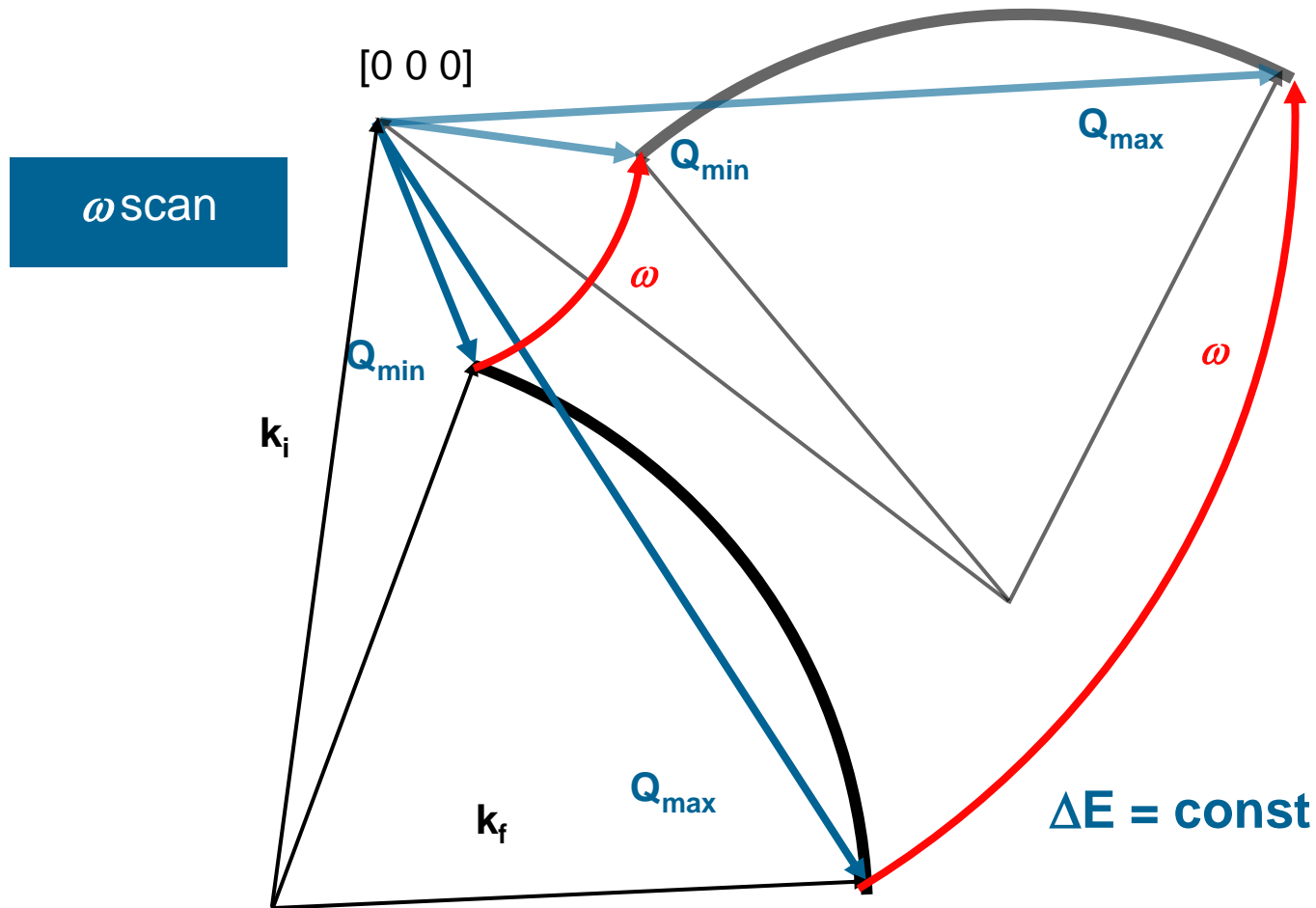
- How do we measure them?

15th Oxford School of Neutron Scattering

Appendix

FLAT CONE

Top view

sample

31 channels
75⁰ angular range

Side view

$k_f = 1.5$ Å^{-1}

$k_f = 3$ Å^{-1} $k_f = 1.5$ Å^{-1}

FC scan modes

ω scan

[0 0 0]

Q_{min}

Q_{max}

ω

ω

k_i

Q_{min}

k_f

Q_{max}

ΔE = const

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