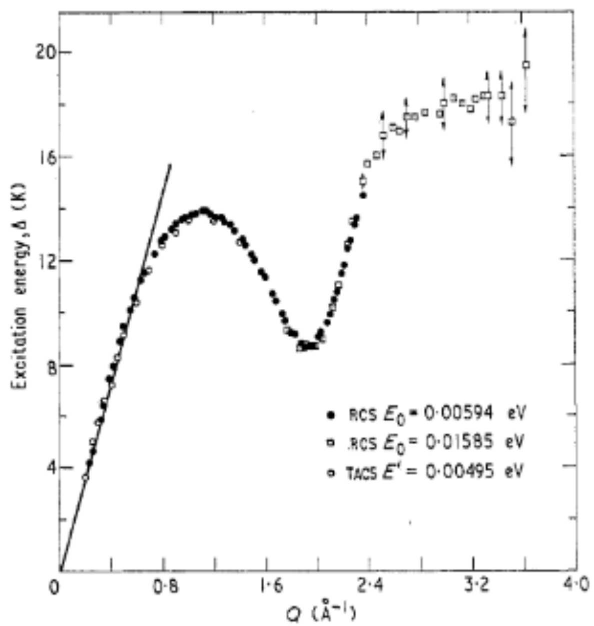
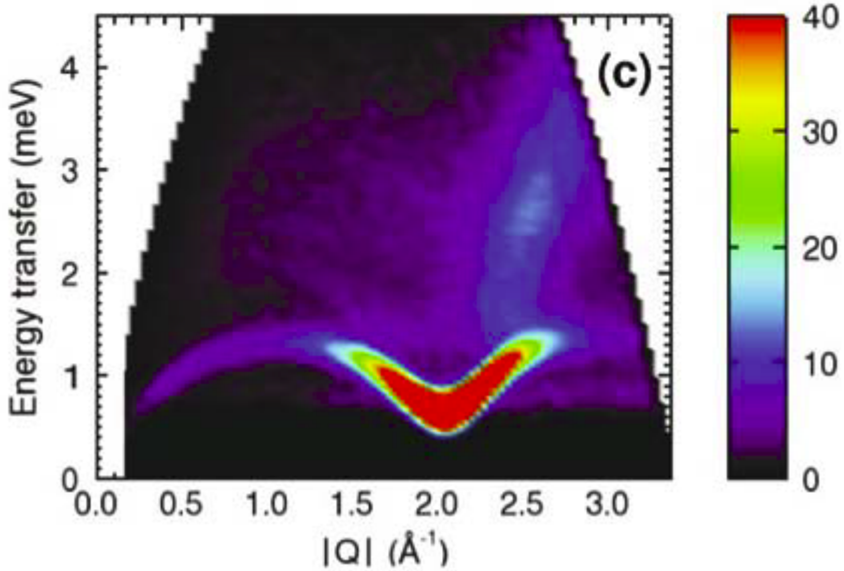
Excitations

Elizabeth Blackburn

University of Birmingham



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



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

14th 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?

14th Oxford School of Neutron Scattering

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

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

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

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

$S(\mathbf{Q},\omega)$ is determined by the physics of the material.
You have already seen expressions for this for phonons and for spin waves.

Recap from Intro to Neutron Scattering

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

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

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

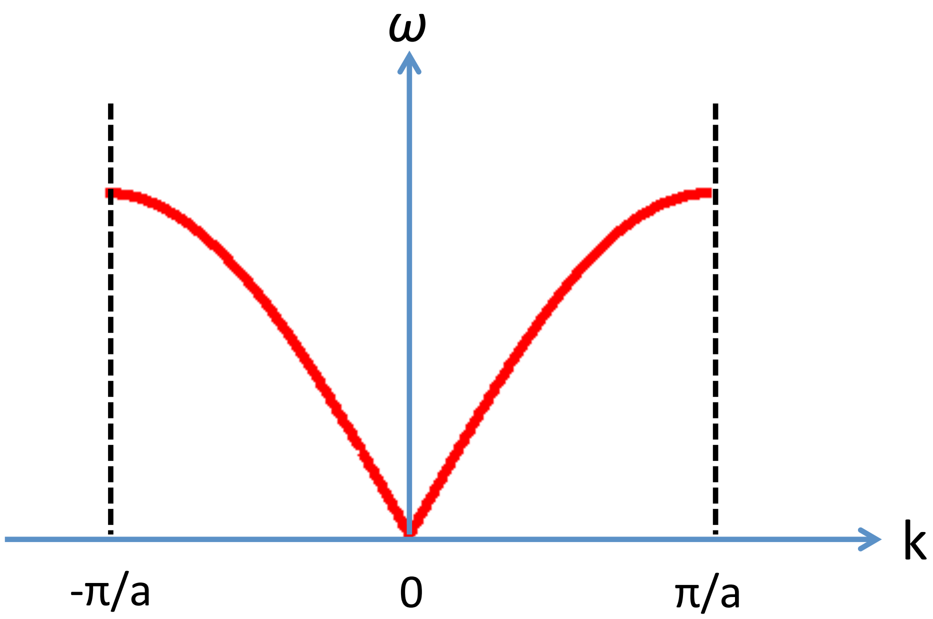
(i) Phonons

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

(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 chains



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

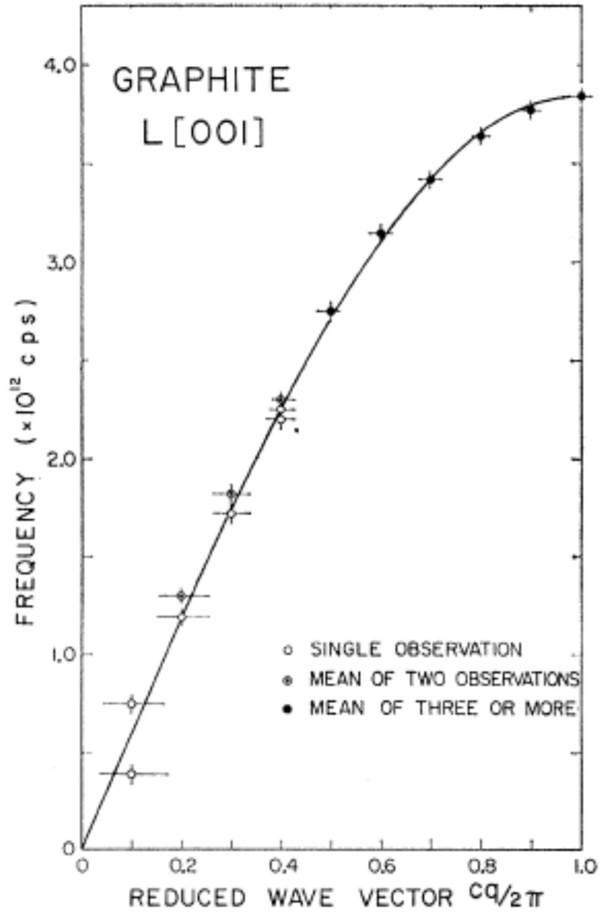
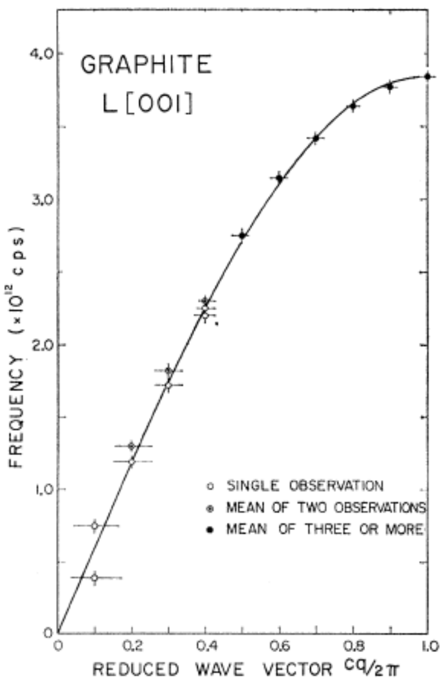


FIG. 3. Dispersion curve for the $L[001]$ modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

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

What can we learn from them?



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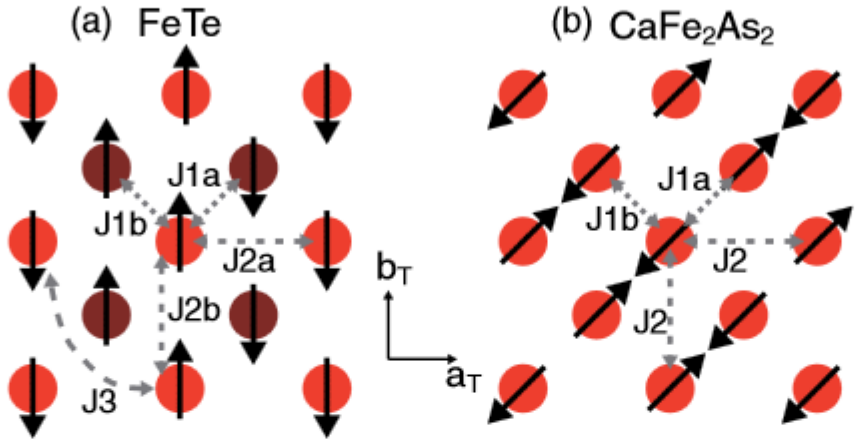
$$\omega_{ph} = 2\sqrt{\frac{K}{M}}\left|\sin\left(\frac{k_{ph}\,a}{2}\right)\right|$$

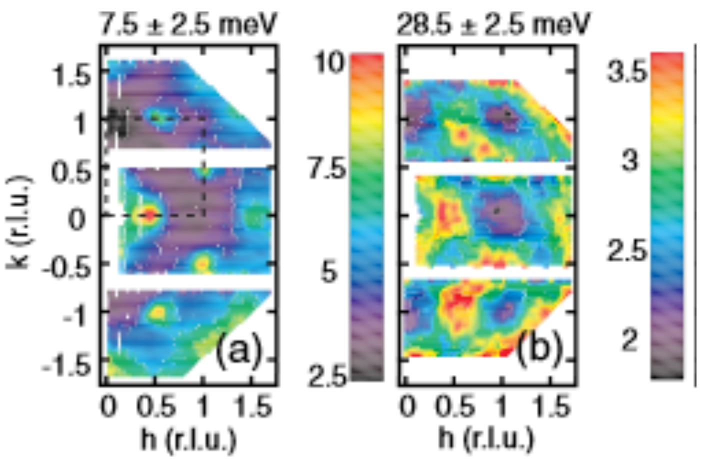
- Elastic constants

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

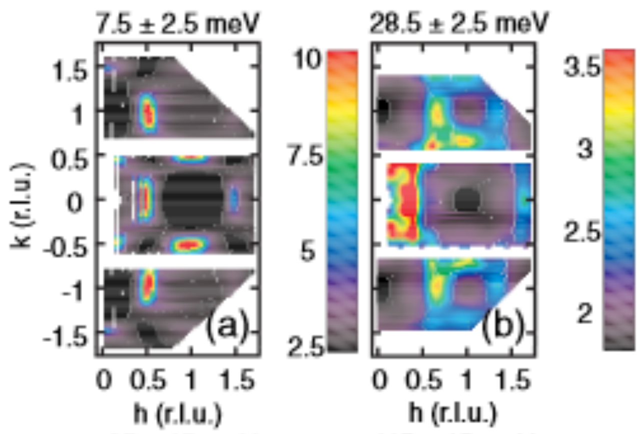
What can we learn from them?



- Elastic constants
- Spin Hamiltonians



Data



Simulation

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

What can we learn from them?

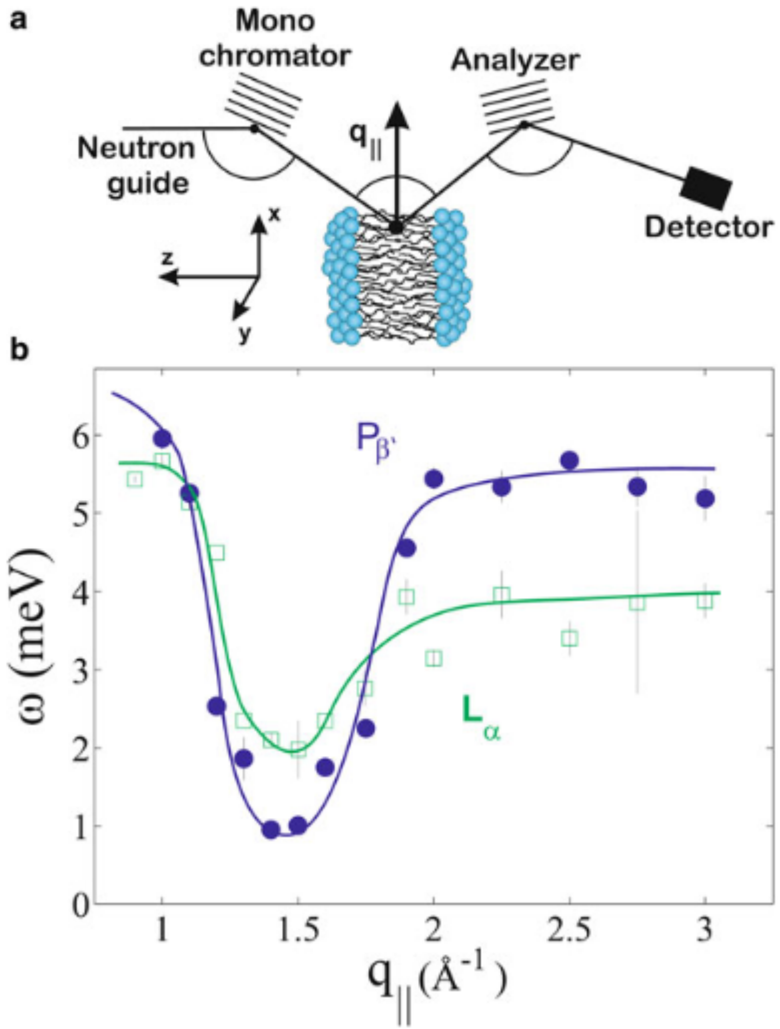


Fig. 10.5 (**a**) Schematic of a triple-axis spectrometer. (**b**) Short-wavelength dispersion relations in the gel ($P_{\beta'}$) and fluid phase L_α of the phospholipid (DMPC) bilayers [42]

- Elastic constants
- Spin Hamiltonians
- Membrane dynamics

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

What can we learn from them?

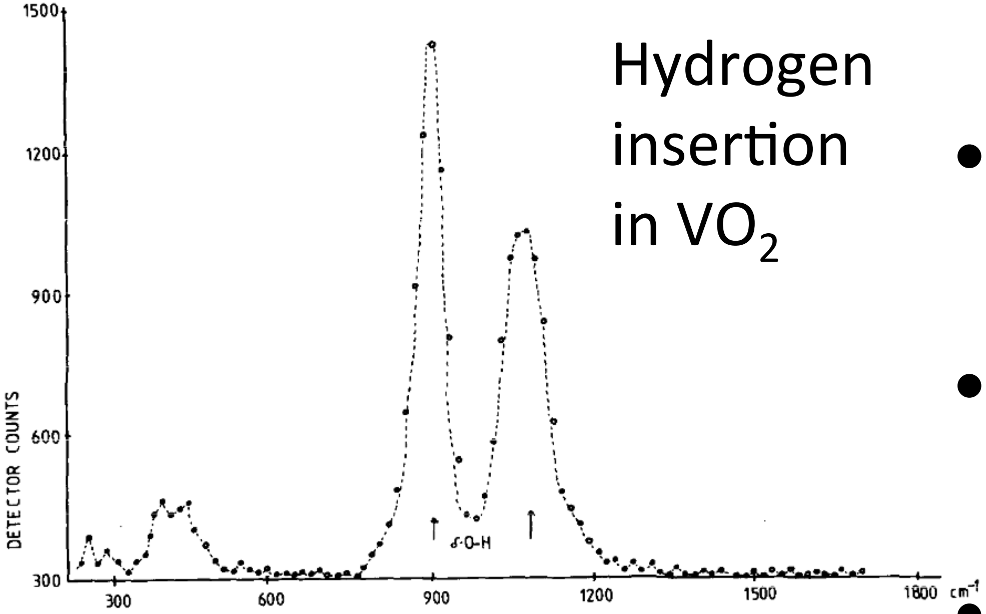


FIG. 2. Inelastic neutron scattering spectrum of $H_{0.3}VO_2$ at 80 K.

Hydrogen insertion in VO_2

TABLE V

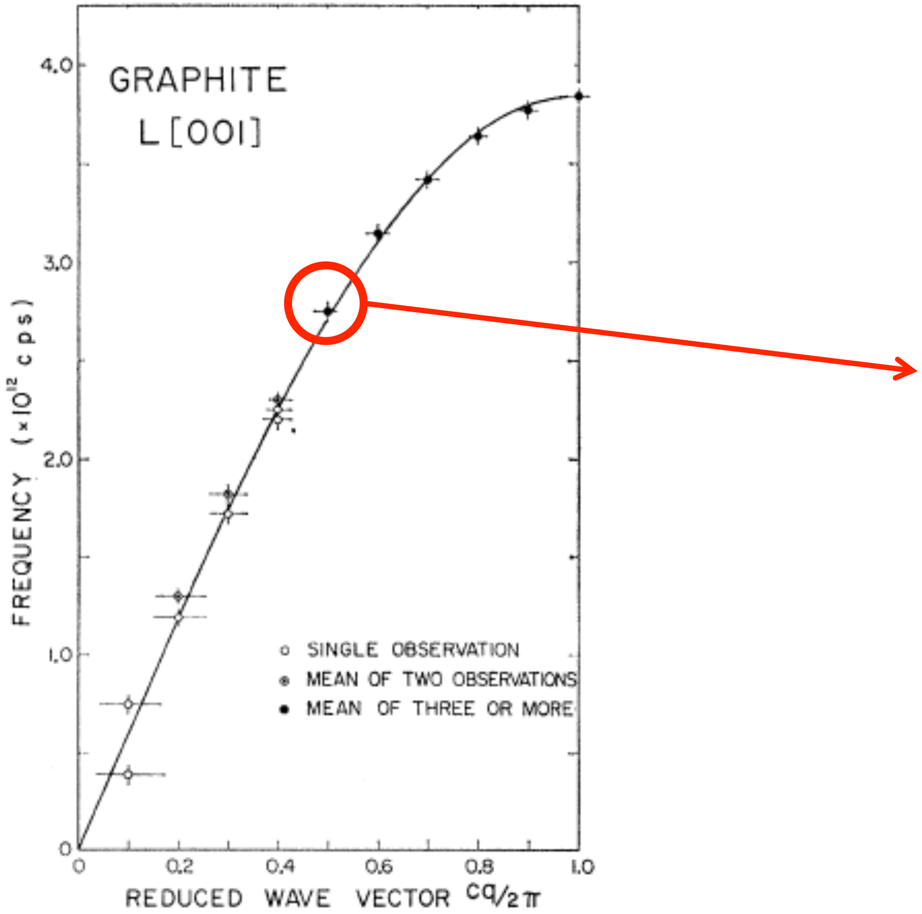
FREQUENCIES AND ASSIGNMENTS OF GENERATED VIBRATIONAL MODES OF H_xVO_2

Calculated frequency (cm^{-1})	Observed frequency (cm^{-1})	Assignment	Mode
3407 (×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			

- Elastic constants
- Spin Hamiltonians
- Membrane dynamics
- How to synthesise particular molecules

Chippindale *et al.*, J. Solid State Chem. 93, 526 (1991).

How do we measure excitations?



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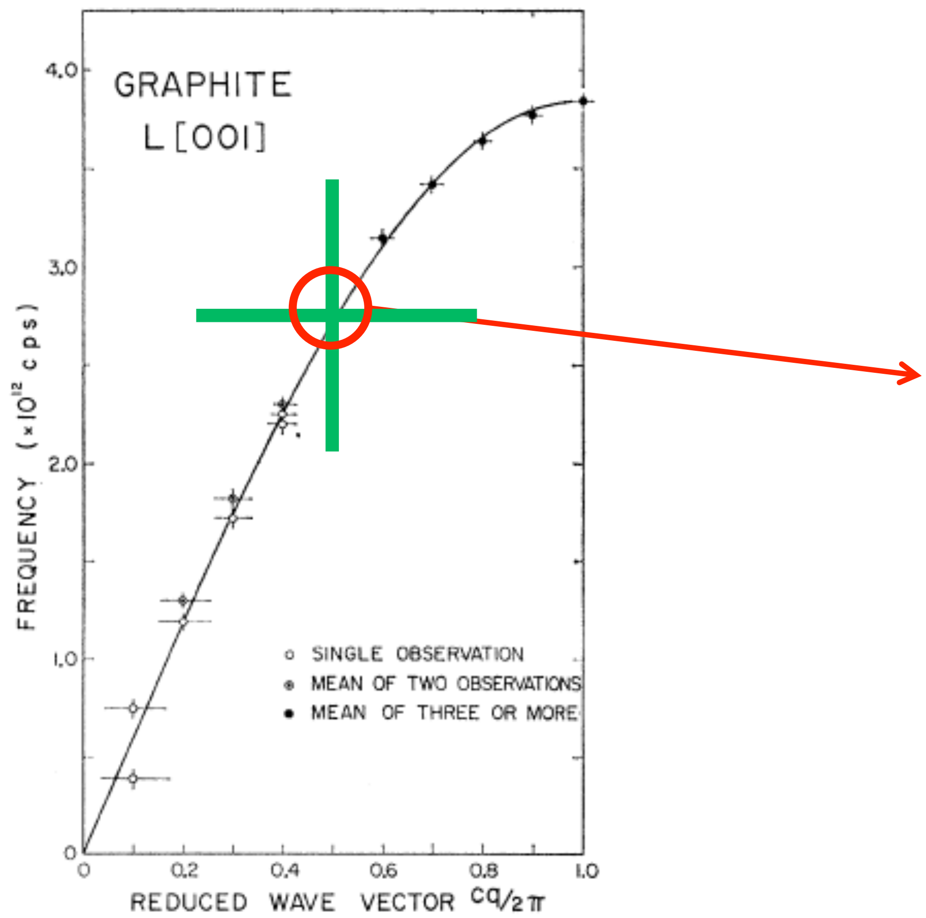
Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

Measuring a phonon spectrum



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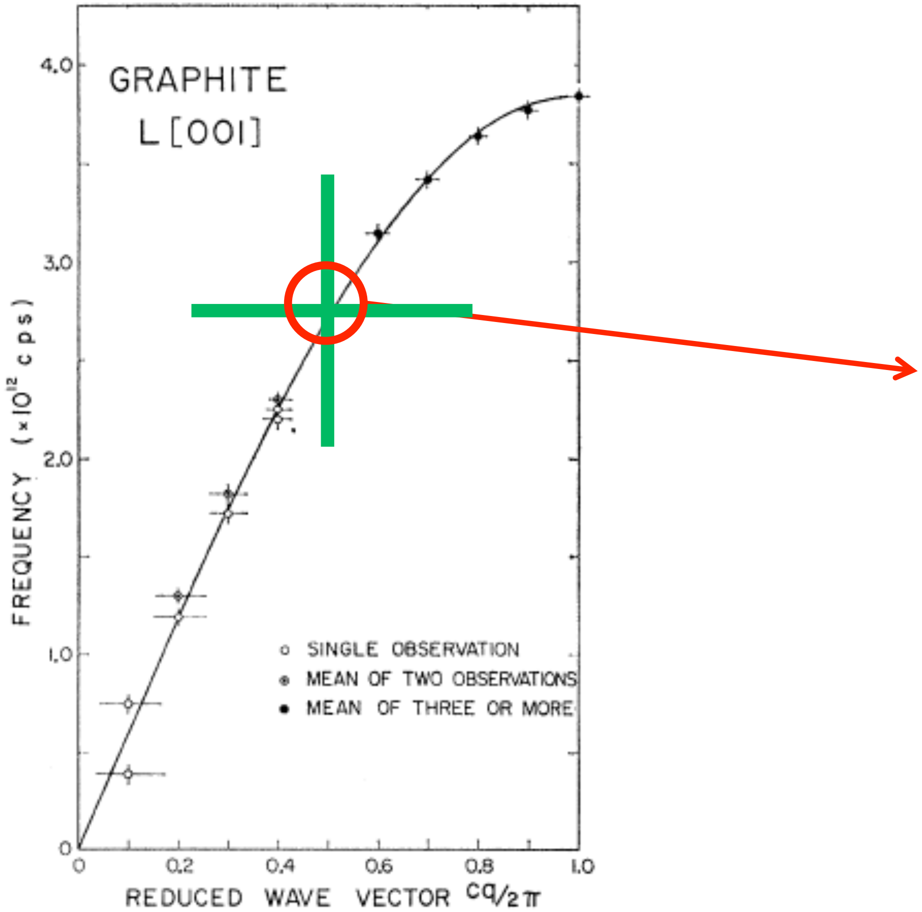

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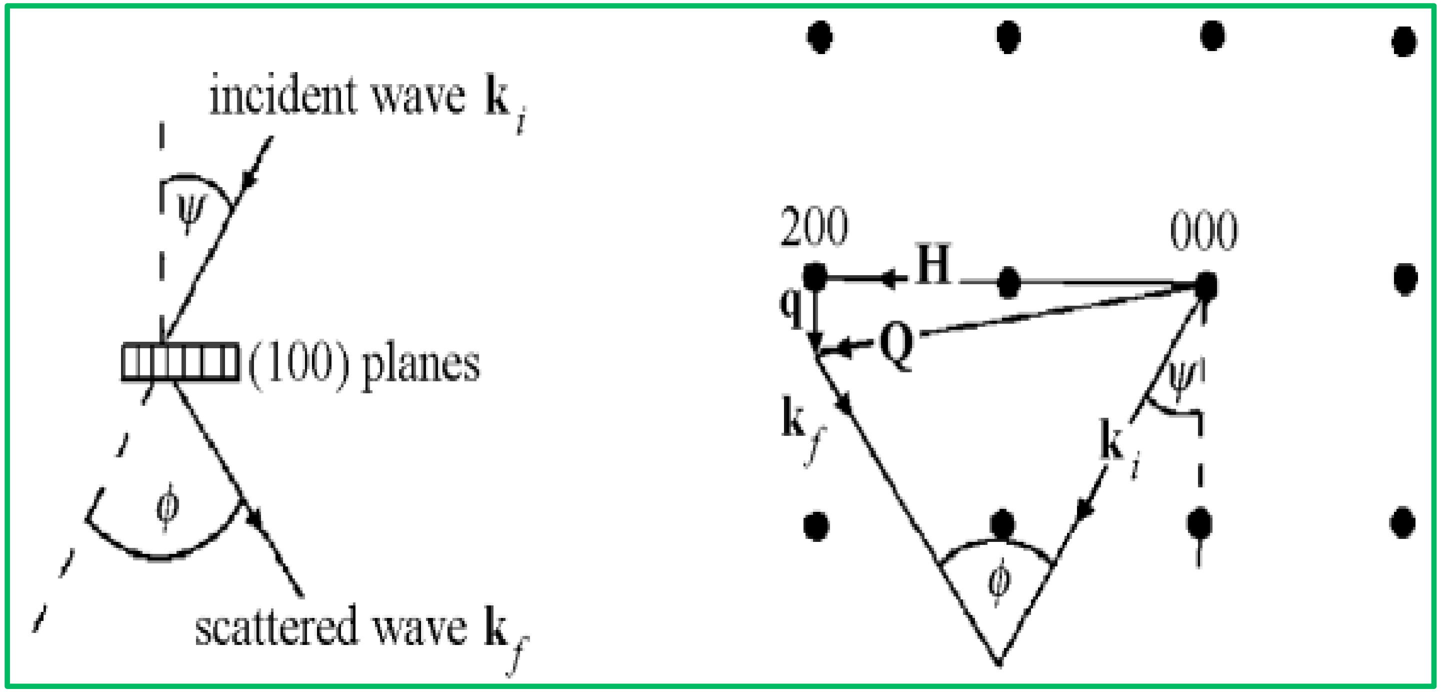
Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)

Things to consider:

- resolution
- spurions

How do we measure these points?

- Momentum transfer $\hbar\mathbf{Q} = \hbar(\mathbf{k}_i - \mathbf{k}_f)$
- Energy transfer $\hbar\omega = E_i - E_f = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)$

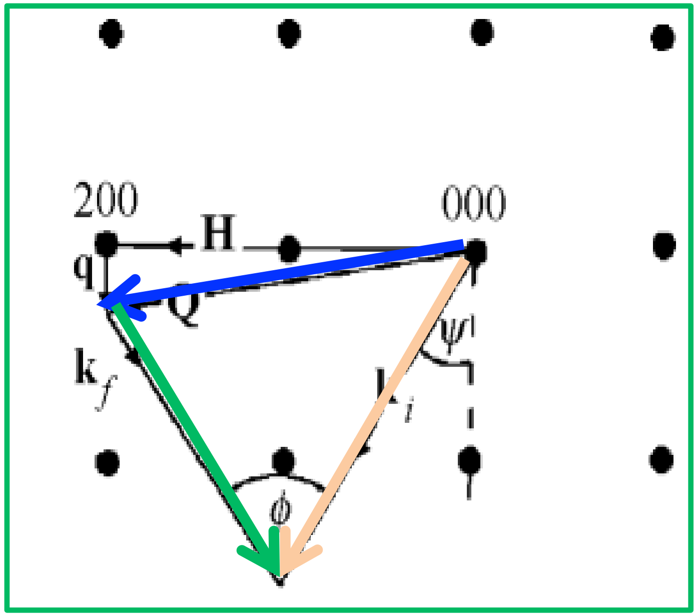


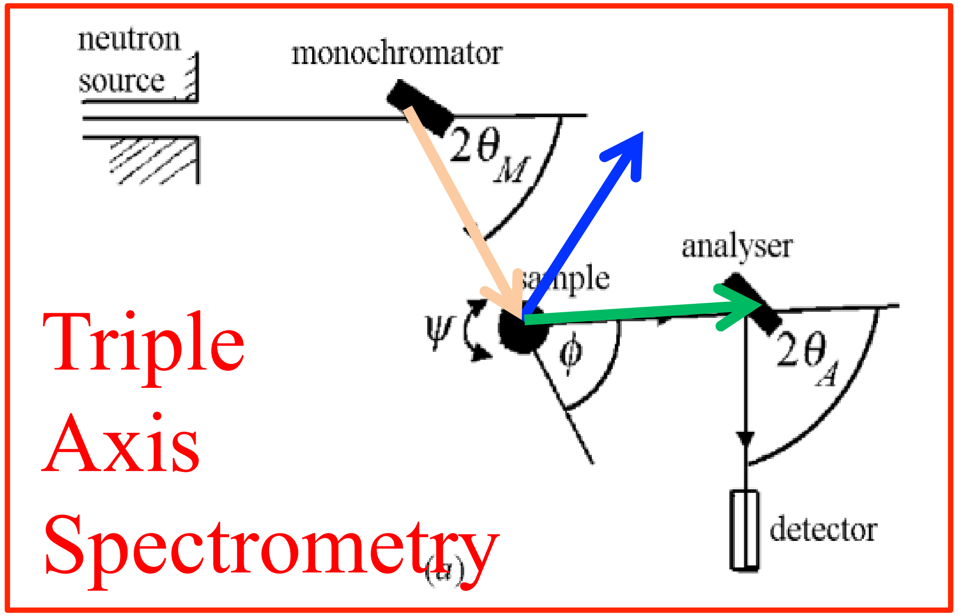
Real space **Reciprocal space**

How do we measure these points?

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

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

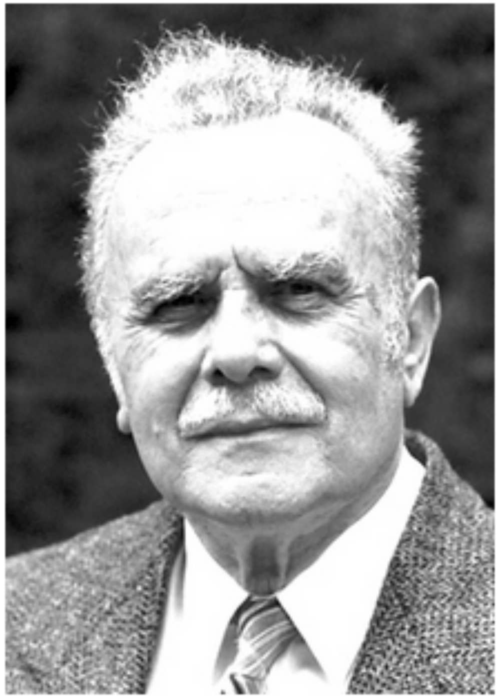




$$|\mathbf{Q}|^2 = |\boldsymbol{k}_i|^2 + |\boldsymbol{k}_f|^2 - 2\,|\boldsymbol{k}_i||\boldsymbol{k}_f|\cos(2\theta)$$

The Nobel Prize in Physics 1994
Bertram N. Brockhouse, Clifford G. Shull

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

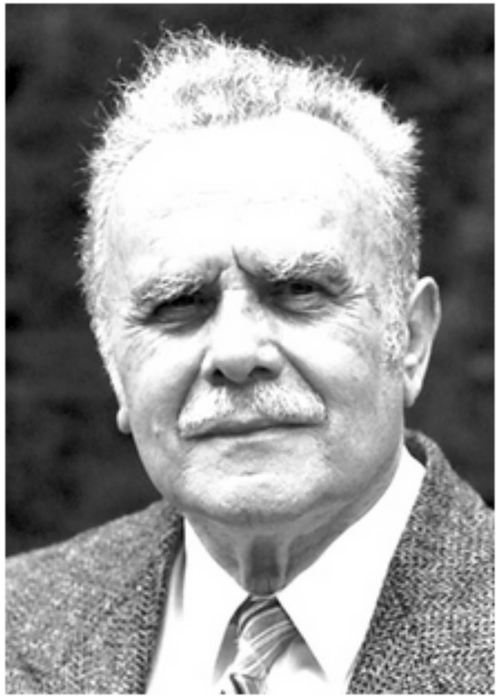
Prize motivation: "for the development of neutron spectroscopy"

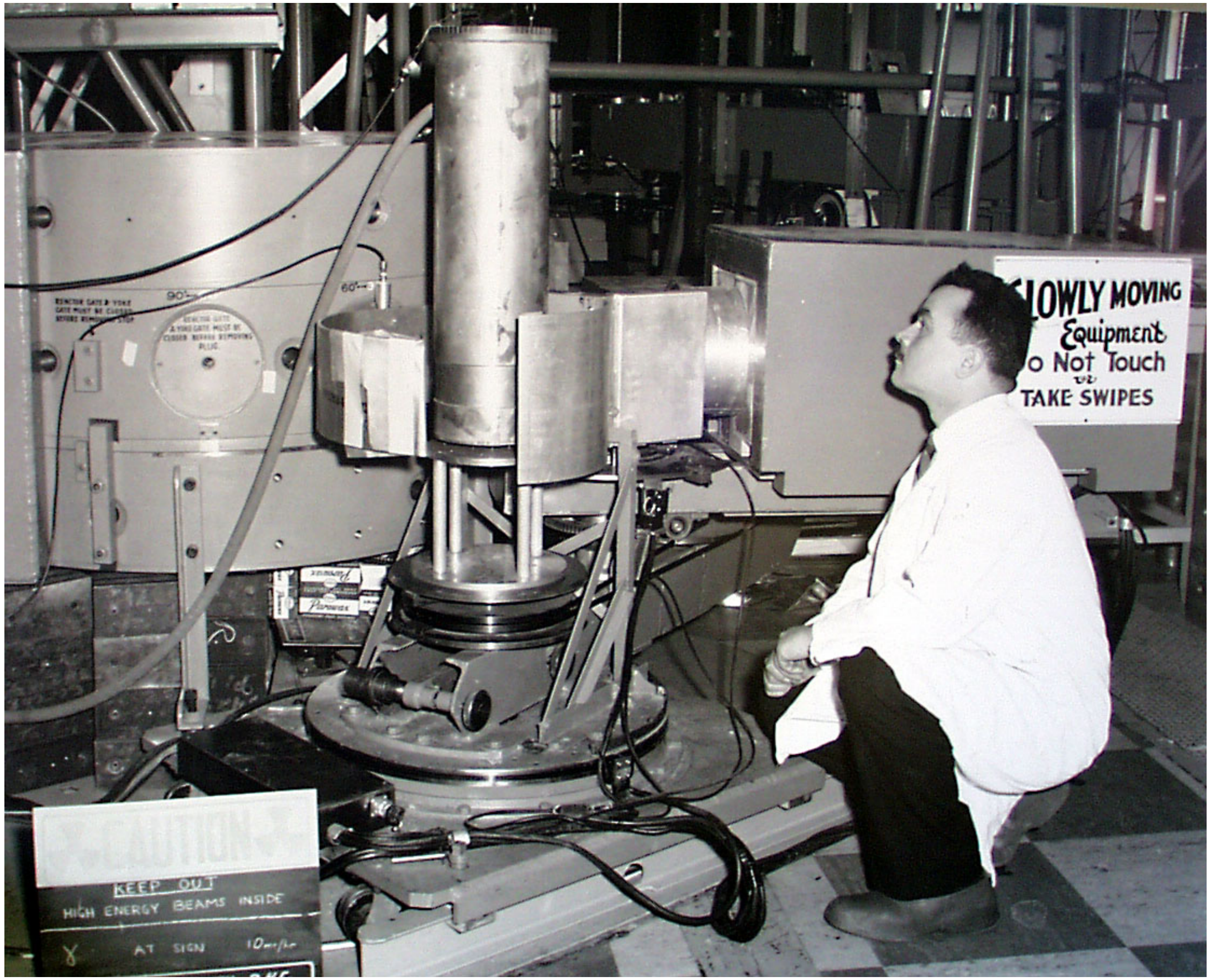
Field: Condensed matter physics, instrumentation

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

The Nobel Prize in Physics 1994
Bertram N. Brockhouse, Clifford G. Shull

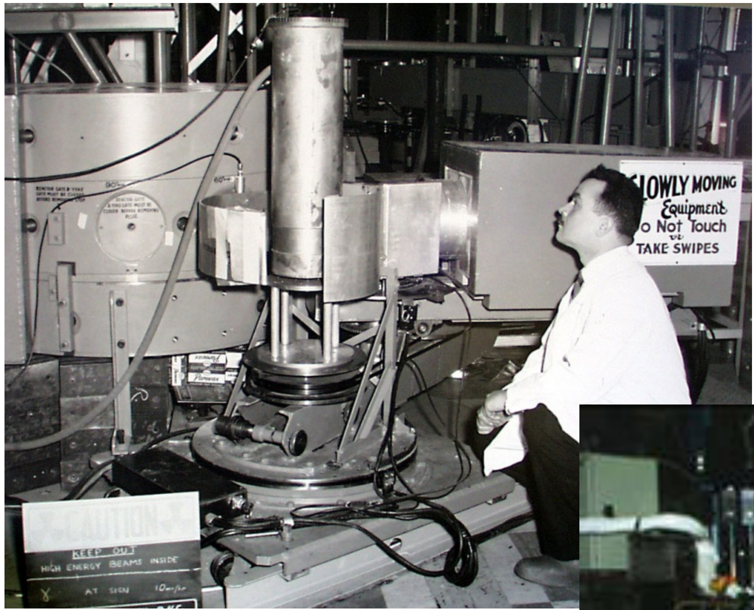
Bertram N. Brockhouse - Facts



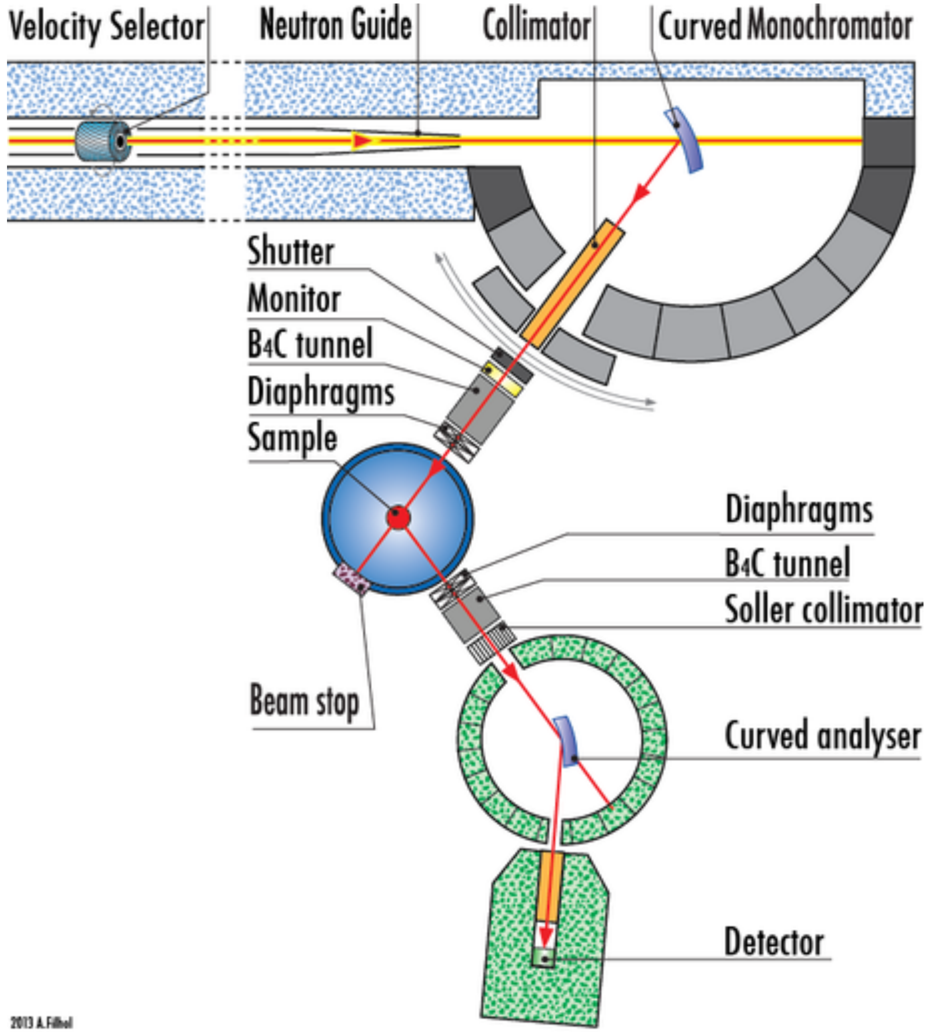


Source: Atomic Energy of Canada Limited, Chalk River, Ontario (CC BY-NC-ND 2.0)

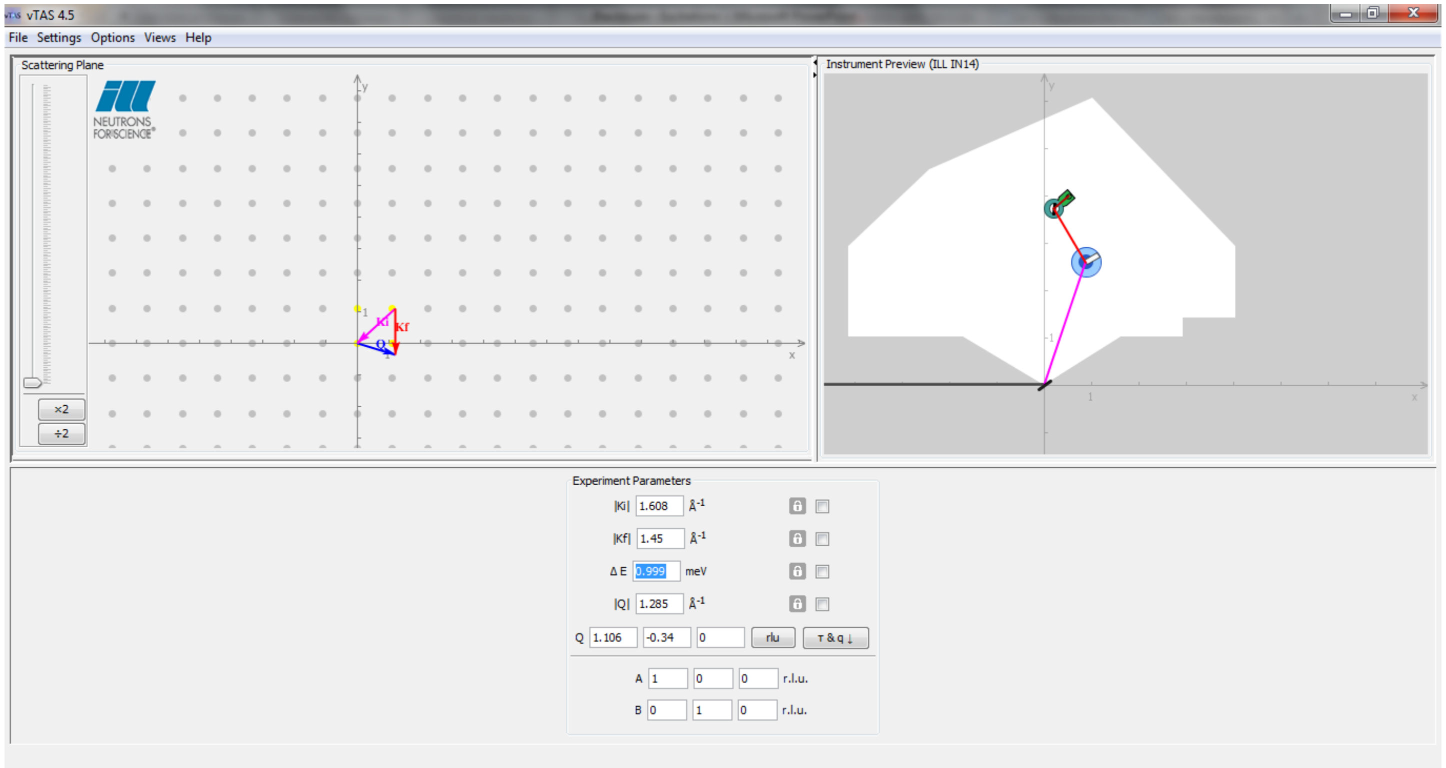
Bertram N. Brockhouse. *Nobelprize.org.* Nobel Media AB 2013.



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


Velocity Selector
Neutron Guide
Collimator
Curved Monochromator
Shutter
Monitor
B4C tunnel
Diaphragms
Sample
Diaphragms
B4C tunnel
Soller collimator
Beam stop
Curved analyser
Detector
2013 A.Filhol

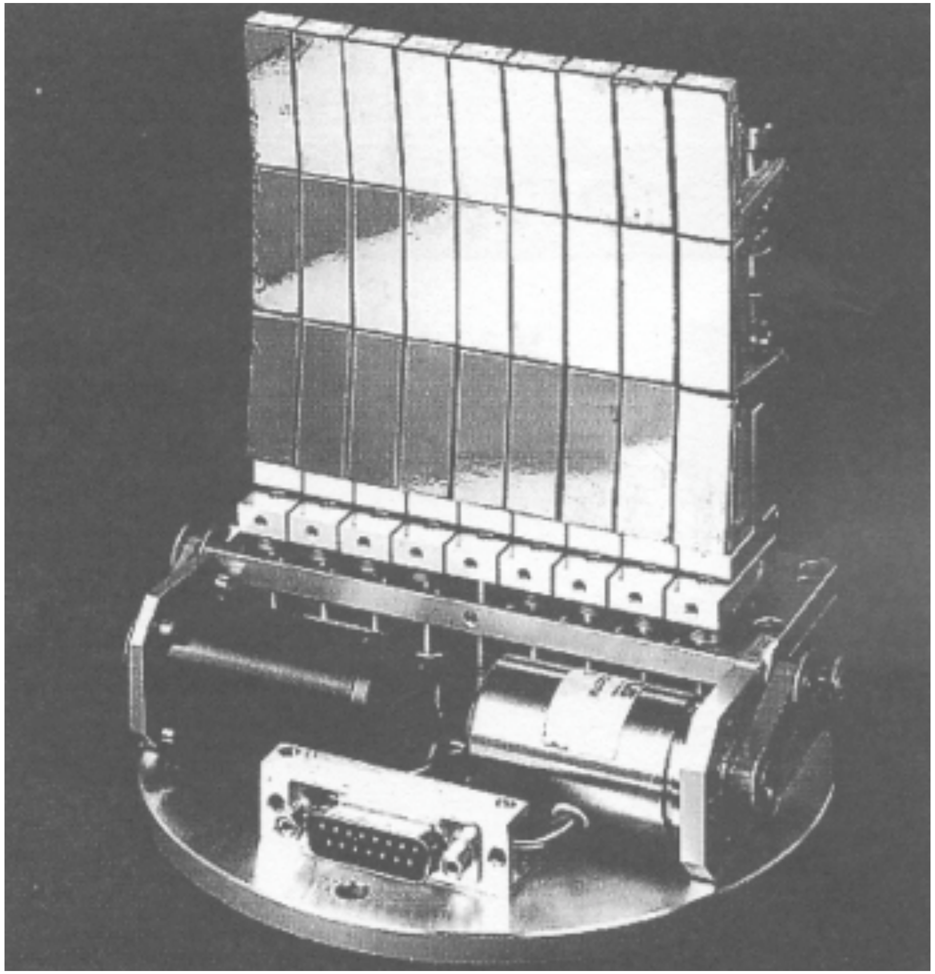
vTAS – a virtual TAS



http://www.ill.eu/instruments-support/computing-for-science/cs-software/all-software/vtas/

The vTAS suite: a simulator for classical and multiplexed three-axis neutron spectrometers
M. Boehm, A. Filhol, Y. Raoul, J. Kulda, W. Schmidt, K. Schmalzl,
Nuclear Inst. and Methods in Physics Research, A (2013) **697**, 40-44.

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

→ θ and λ are coupled.

k_i and k_f have their own resolution volumes, with distinct orientations in **Q**- ω space.

→ resolution ellipsoid.

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

BUT

It also leads to **lower** counts

Experimental Choices: Resolution

Q and ω only defined to a certain level of precision

Bragg's Law, $n\lambda = 2d \sin\theta$

→ θ and λ are coupled.

$\boldsymbol{k}_i$ and $\boldsymbol{k}_f$ have their own resolution volumes, with distinct orientations in **Q**- ω space.

→ resolution ellipsoid.

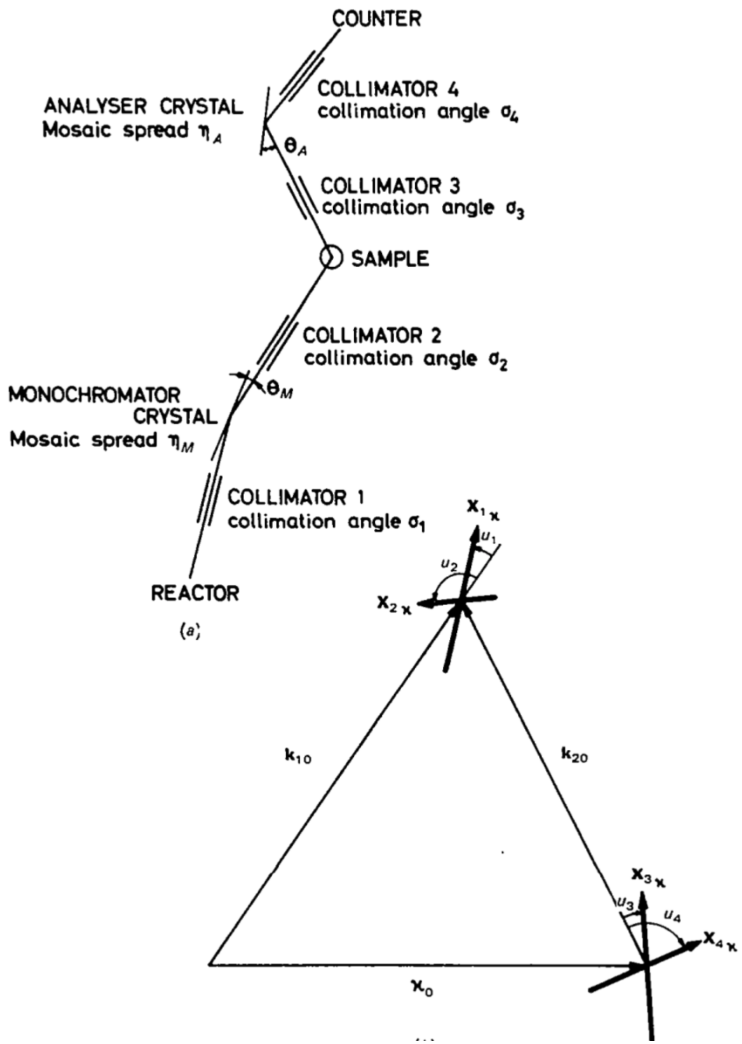
Experimental Choices: The resolution ellipsoid

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

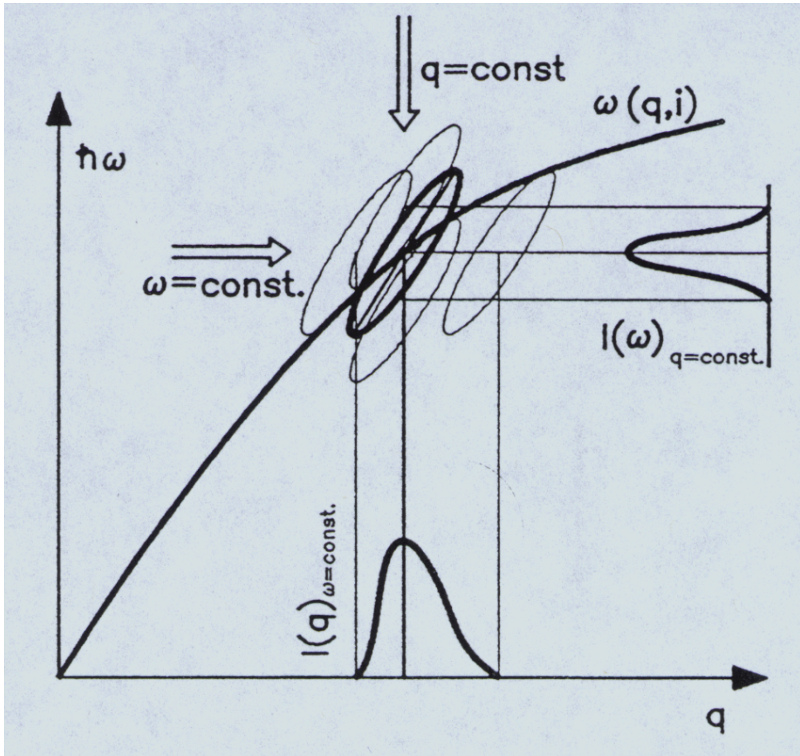
Focussing can also have an effect

Tune your experiment to give you what you want



Experimental Choices:
The resolution ellipsoid

Tune your experiment to give you what you want



References:

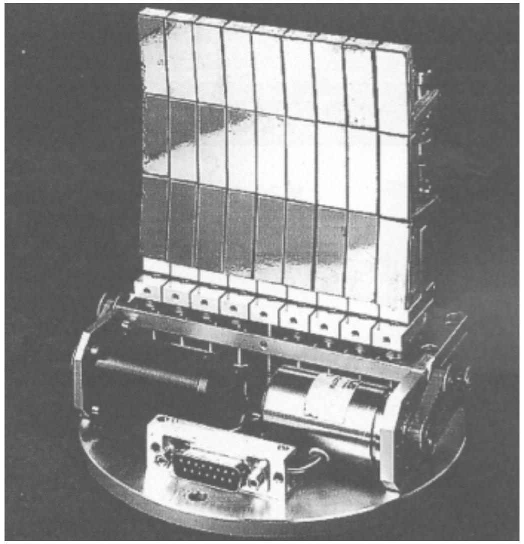
RESTRAX – Saroun and Kulda, http://neutron.ujf.cas.cz/restrax/doc/index.html

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)

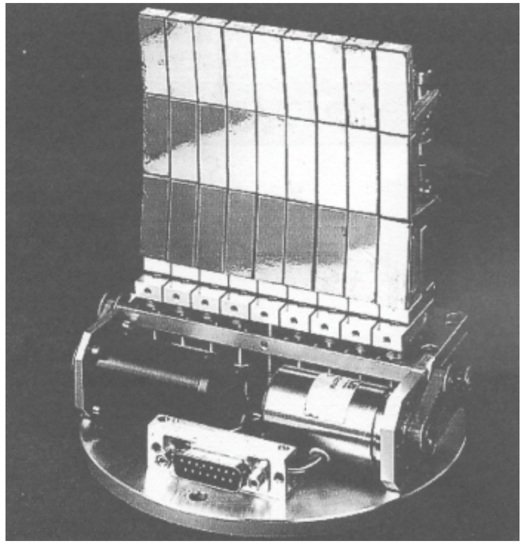
Focussing distance depends upon Bragg angle so the radius, *R*, of the mono/analyzer crystal needs to be variable.

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

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Physica B 350 (2004) 11–16

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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.

PACS: 25.40.Fq; 75.40.Gb; 78.70.Nx

Keywords: Inelastic neutron scattering; Copper germanate $CuGeO_3$; Neutron polarisation analysis

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

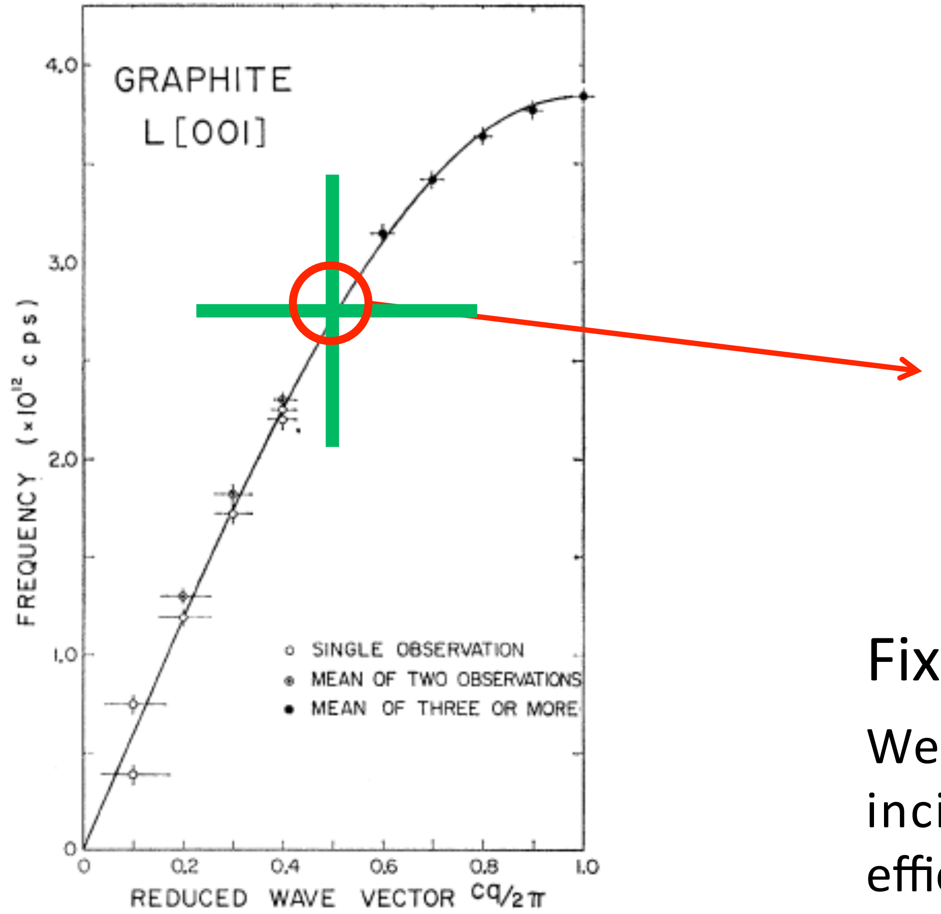


FIG. 3. Dispersion curve for the $L[001]$ modes in which the planes move as rigid units. The solid line is the best fitting (simple) sine wave to the experimental points. The assignment of errors is discussed in the text.

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

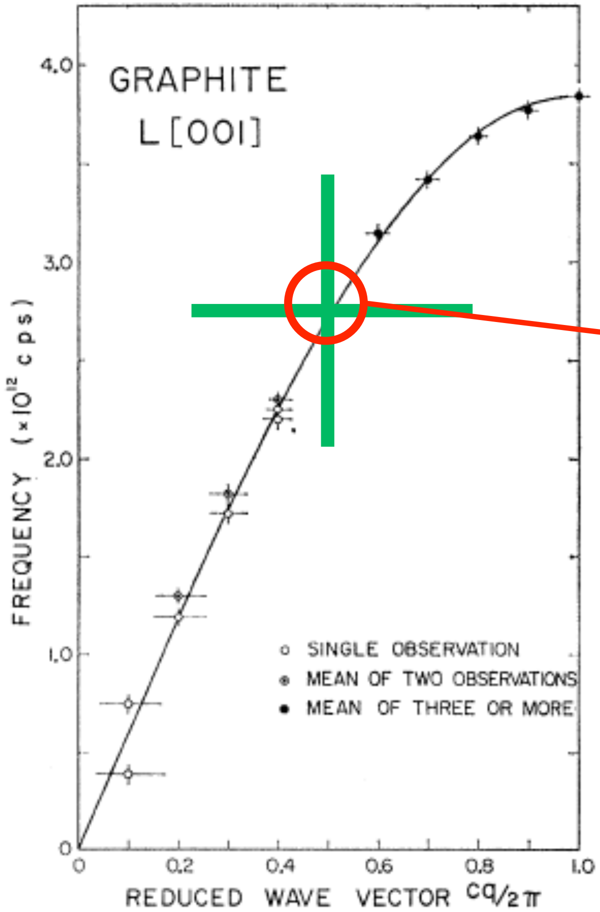
$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i} S(\mathbf{Q},\omega)$$

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

Fixed $\boldsymbol{k}_i$ or fixed $\boldsymbol{k}_f$?

We normalised to the number of incident neutrons using a low efficiency monitor whose efficiency depends on the neutron velocity. In fixed $\boldsymbol{k}_i$ mode, this efficiency is fixed and we therefore measure signal per monitor

Measuring a phonon spectrum



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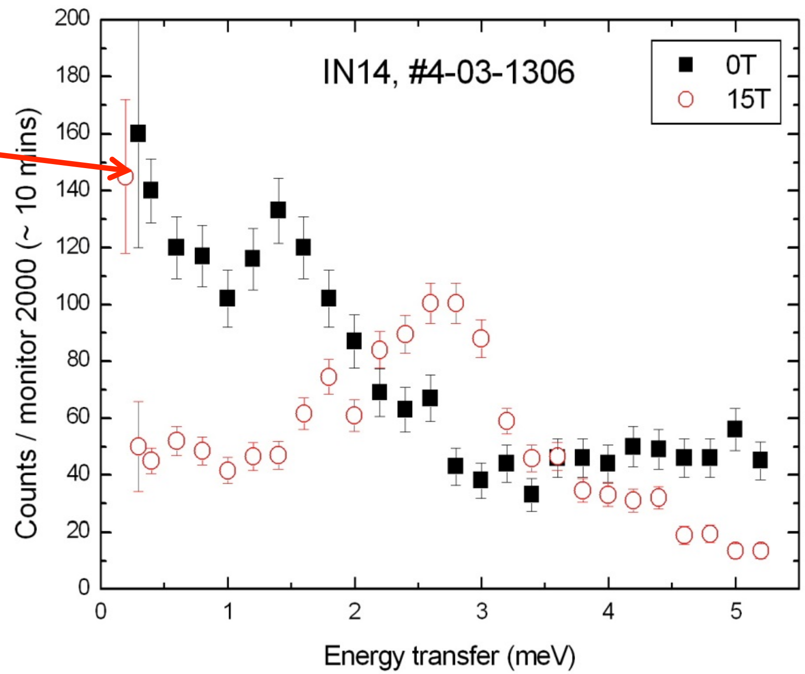
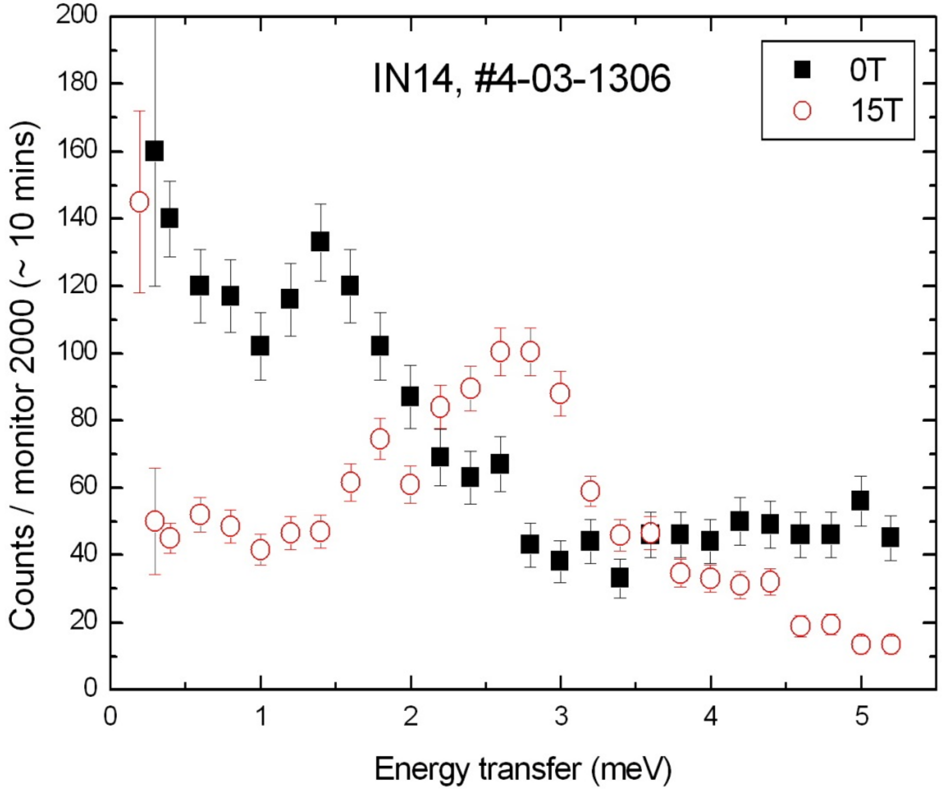


Dolling and Brockhouse., Phys. Rev. 128, 1120 (1962)


IN14, #4-03-1306
0T
15T
Counts / monitor 2000 (~ 10 mins)
Energy transfer (meV)
0
20
40
60
80
100
120
140
160
180
200
0
1
2
3
4
5

Advantages and disadvantages of the triple-axis method

Advantages

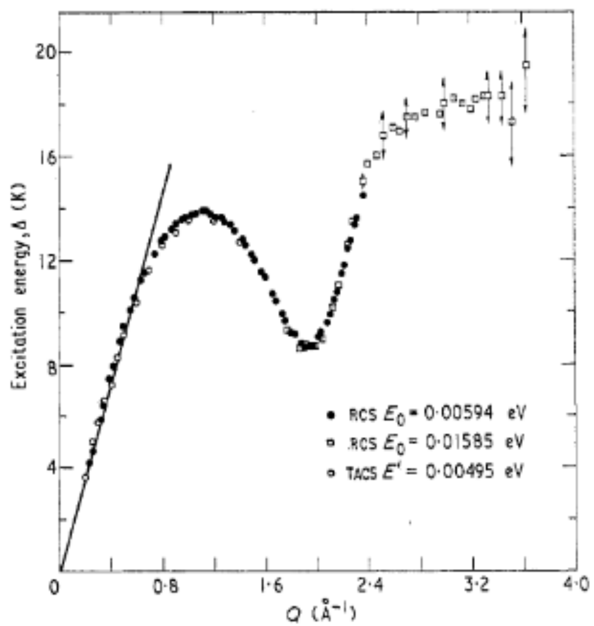
1. Can focus all intensity on point in reciprocal space that is important
2. Can make measurements along high-symmetry directions
3. Can use either constant-***Q*** or constant–E, depending on type of excitation being examined.
4. Can use focusing and other 'tricks' to improve the signal/noise
5. Can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

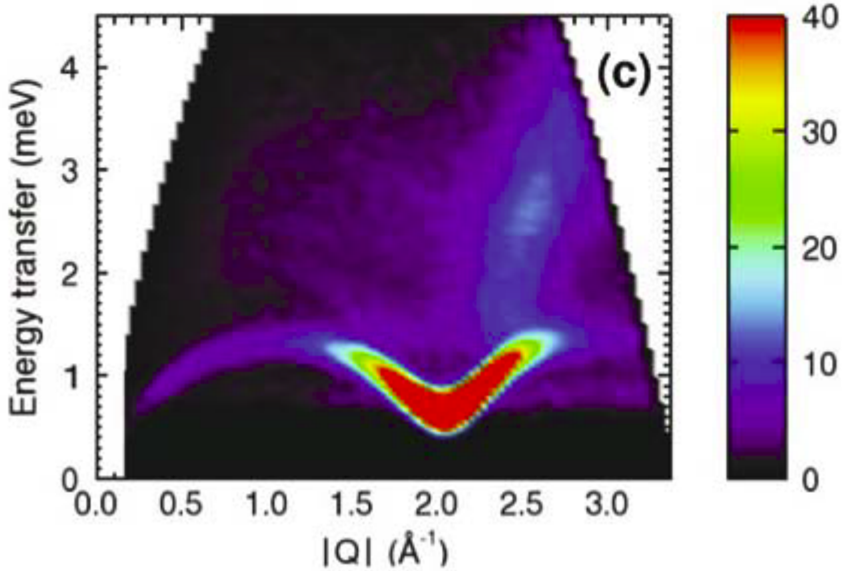
Superfluid helium

TAS



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

TOF



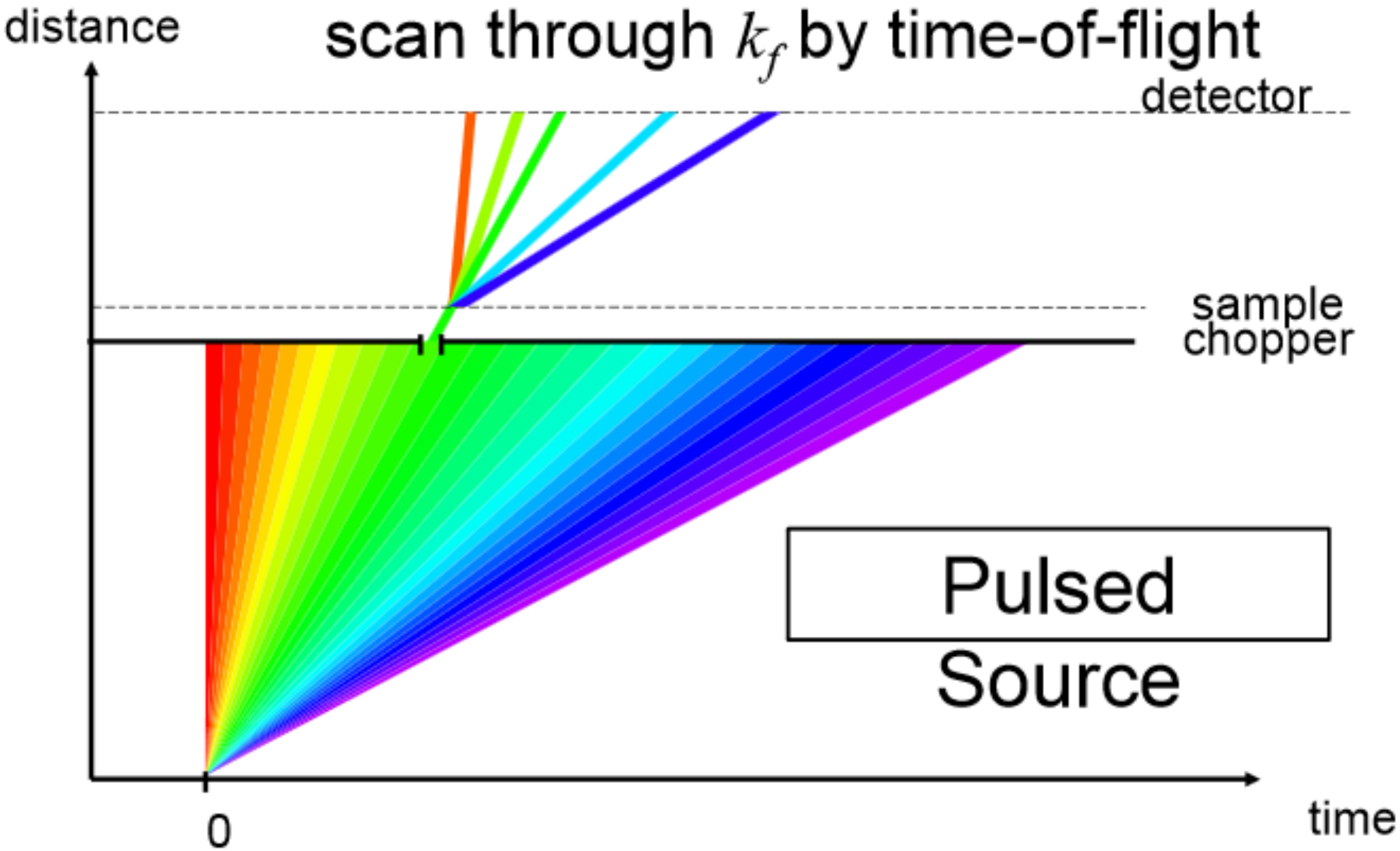
Blackburn *et al.*, Pramana 71, 673 (2008)

Time-of-flight spectroscopy

Direct geometry:

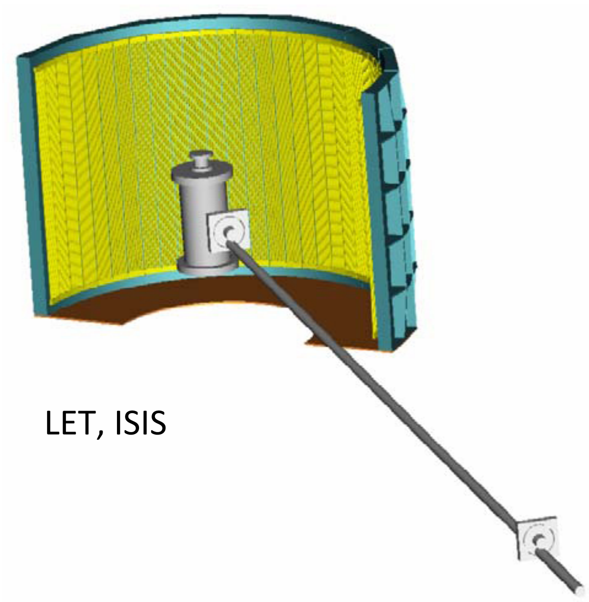
fix k_i by chopper phasing

scan through k_f by time-of-flight

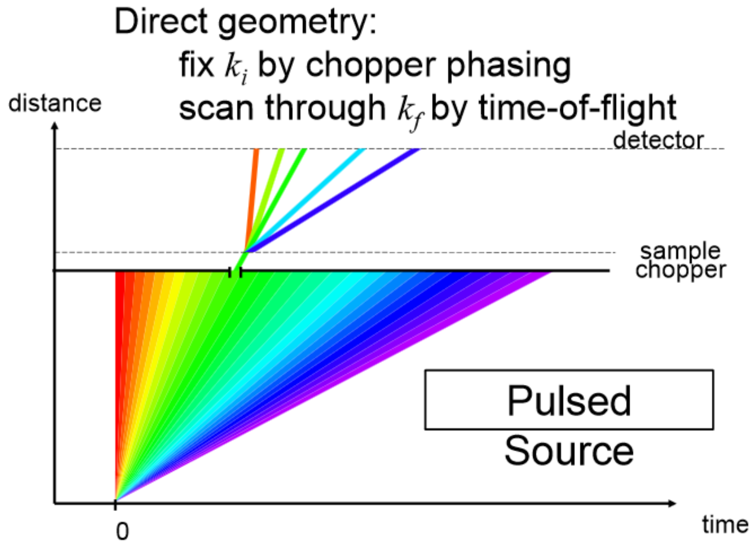


from Ken Andersen

Time-of-flight spectroscopy



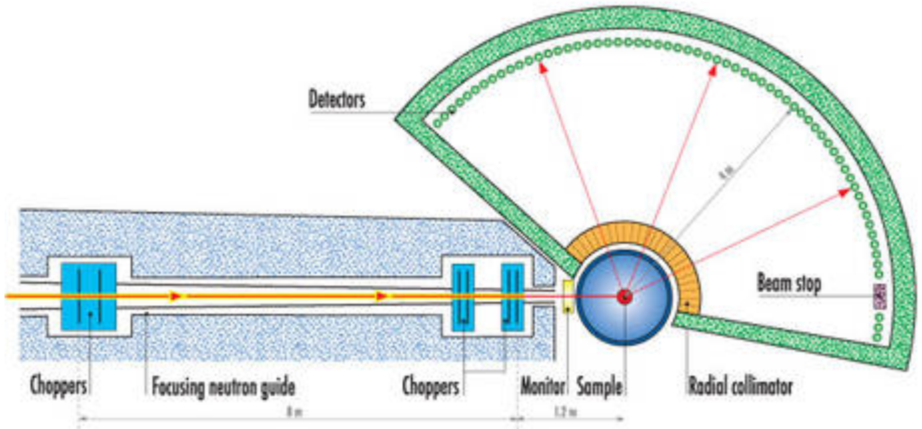
LET, ISIS



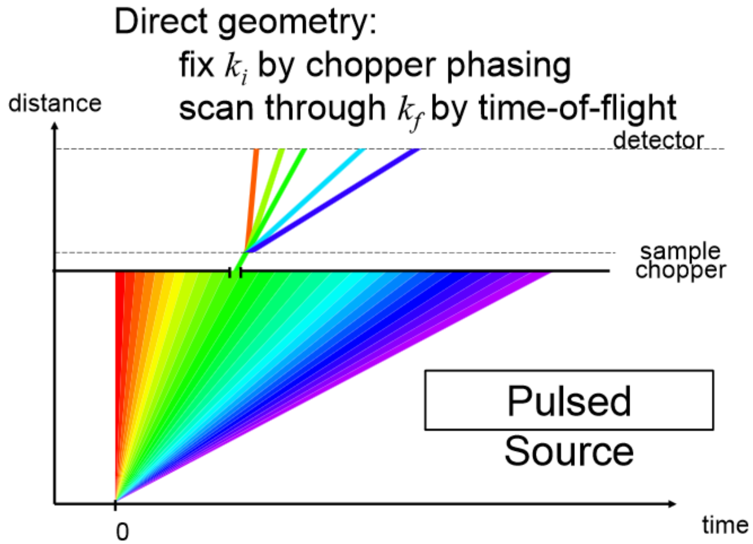
$$\text{final velocity} = \frac{\text{sample-detector distance}}{\text{time to detector} - \text{time to sample}}$$

http://www.ill.eu/instruments-support/instruments-groups/instruments/in5/how-it-works/3d-animation/

Time-of-flight spectroscopy



IN5, ILL



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

This gives us the neutron's final energy

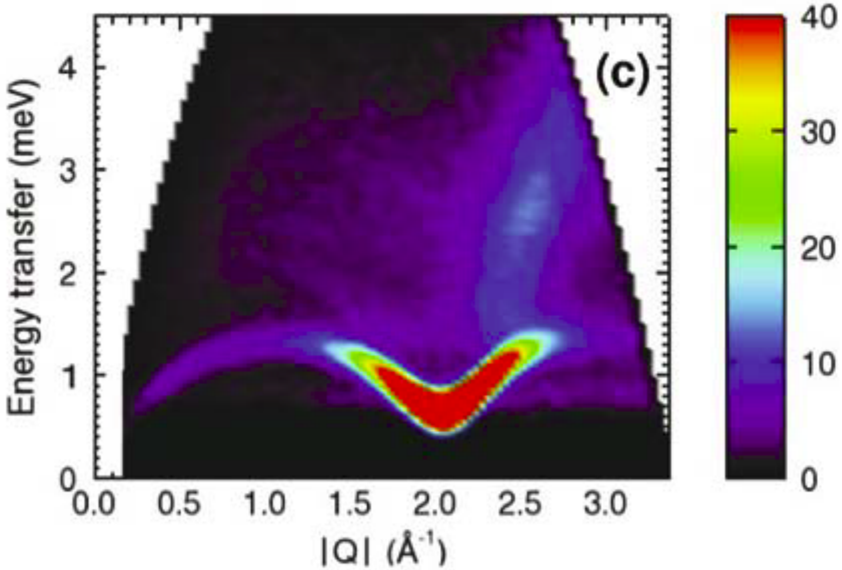
http://www.ill.eu/instruments-support/instruments-groups/instruments/in5/how-it-works/3d-animation/

Time-of-flight spectroscopy

Measured quantity:

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

Remember the bin sizes:



Time-of-flight spectroscopy

$$|\mathbf{Q}|^2 = |\boldsymbol{k}_i|^2 + |\boldsymbol{k}_f|^2 - 2\,|\boldsymbol{k}_i||\boldsymbol{k}_f|\cos(2\theta)$$

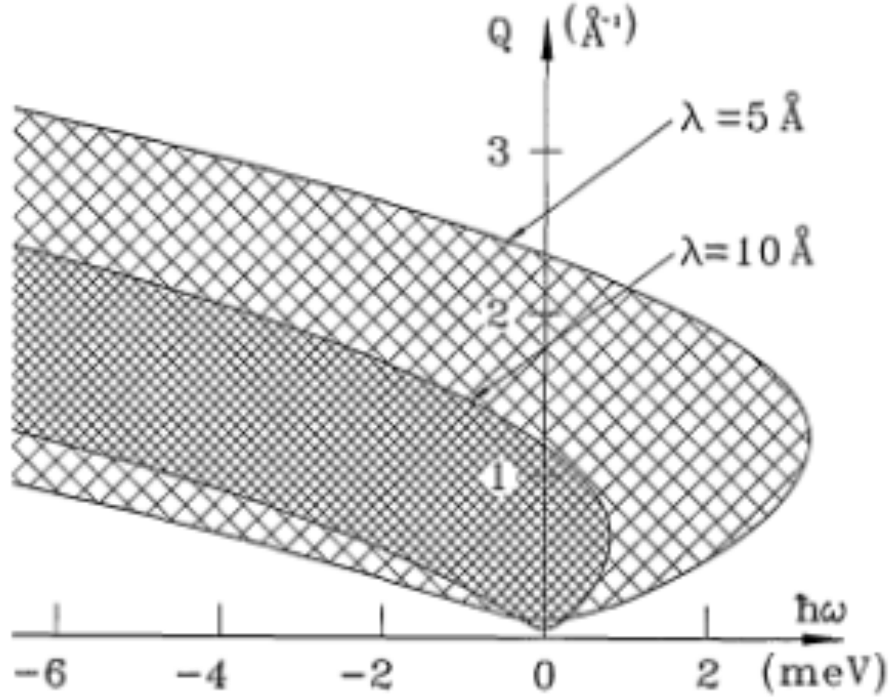
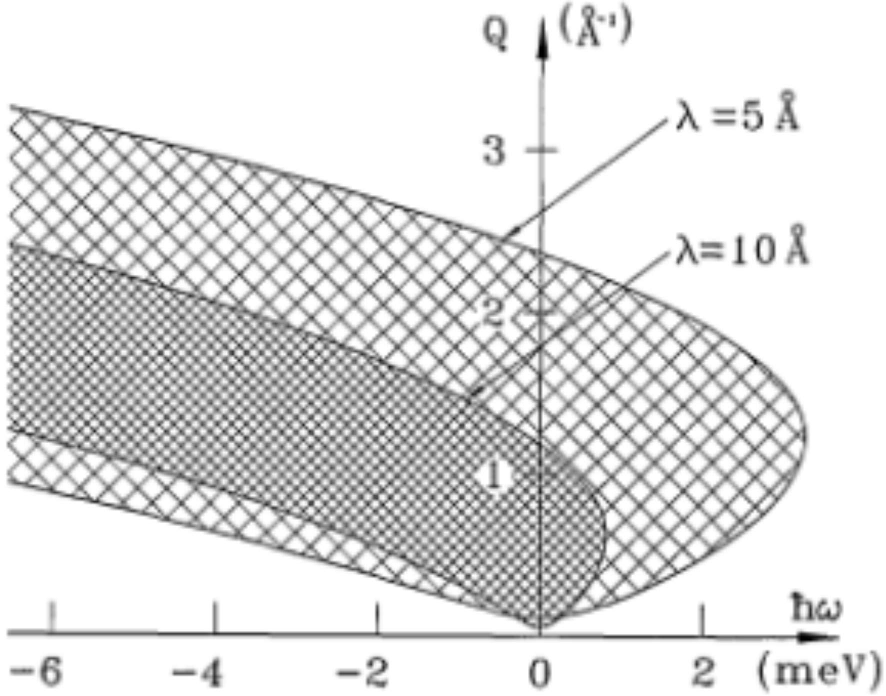


Fig. 17. Plots of the accessible region in (Q,ω) space for neutrons of wavelength 5 and 10 Å (energy 3.272 and 0.818 meV, respectively). The minimum and maximum scattering angles are 5° and 140°. There is no (theoretical) limit to the energy transfer in neutron energy gain.

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

Time-of-flight spectroscopy

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



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Experimental choices: Intensity vs Resolution

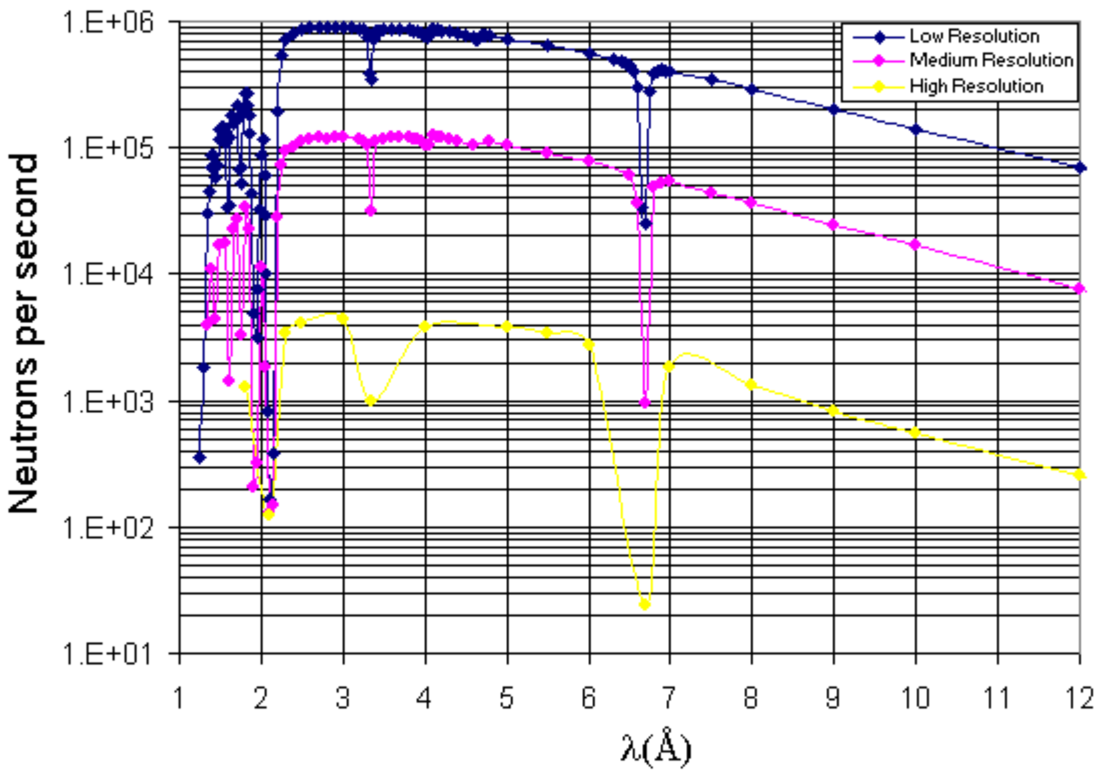
What can we change?

incident wavelength

pulse width at sample chopper

frame overlap ratio

DCS, NCNR, NIST



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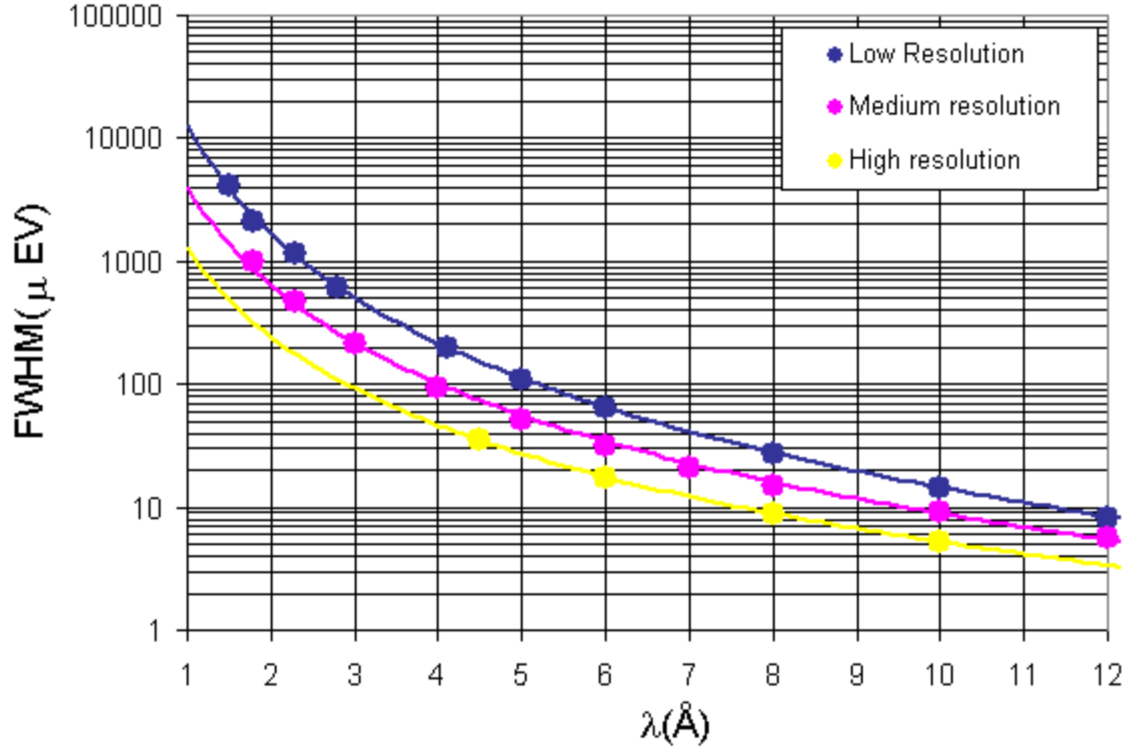

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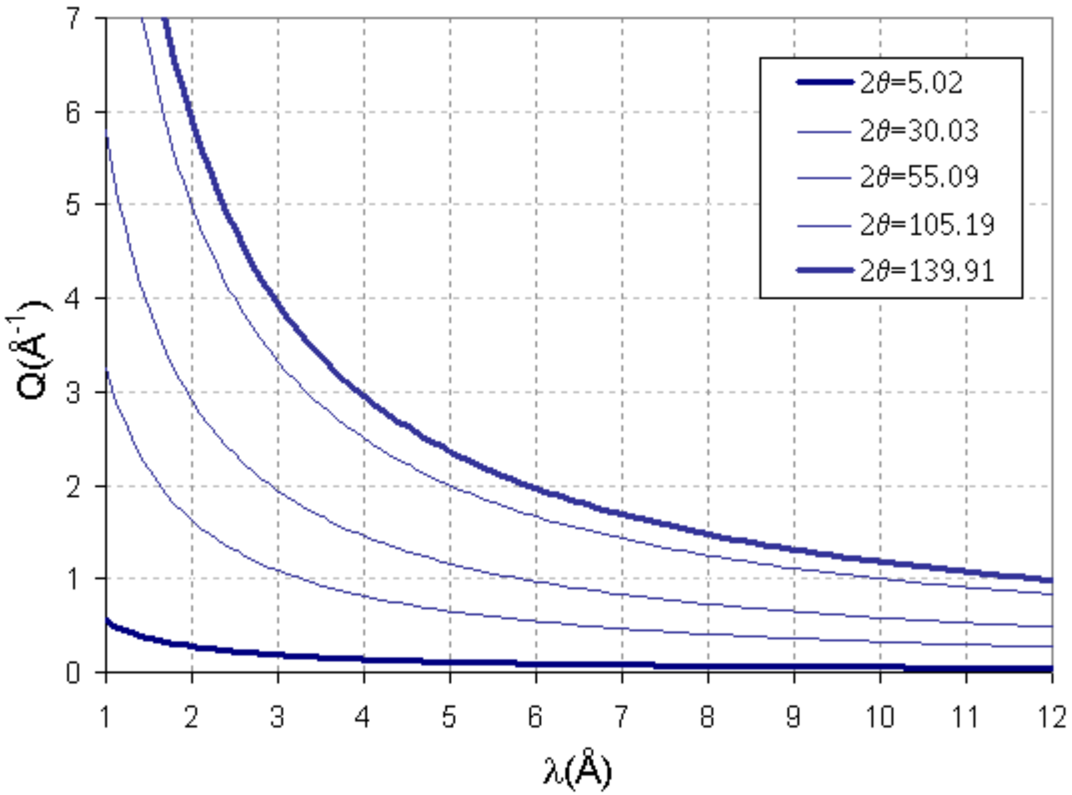
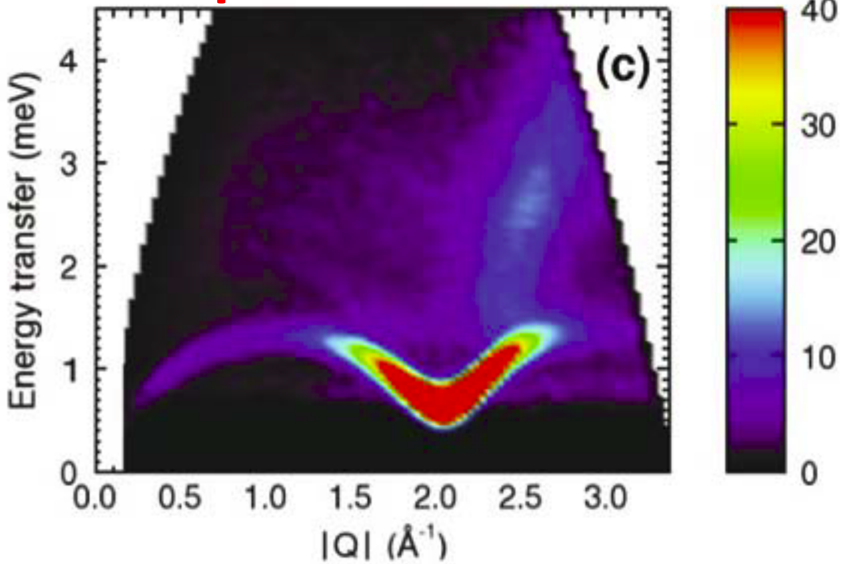
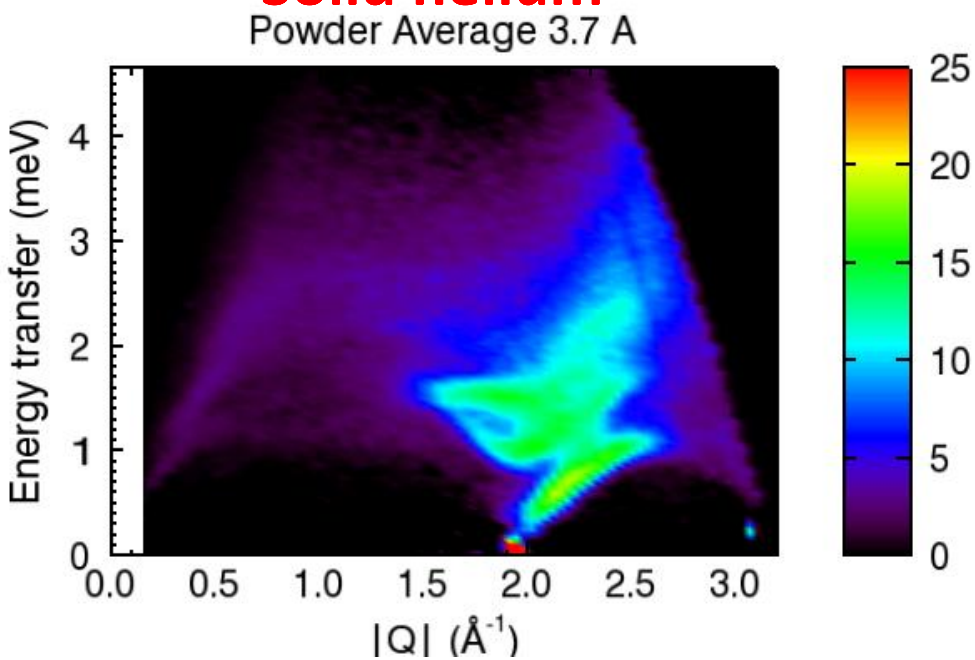


TOF on single crystals

Superfluid helium

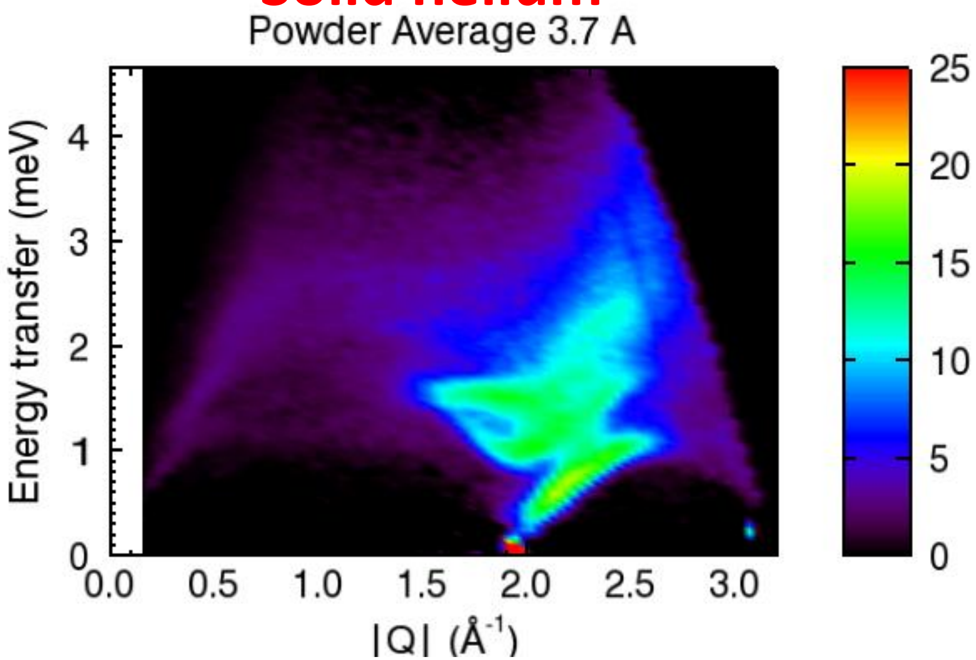


Solid helium

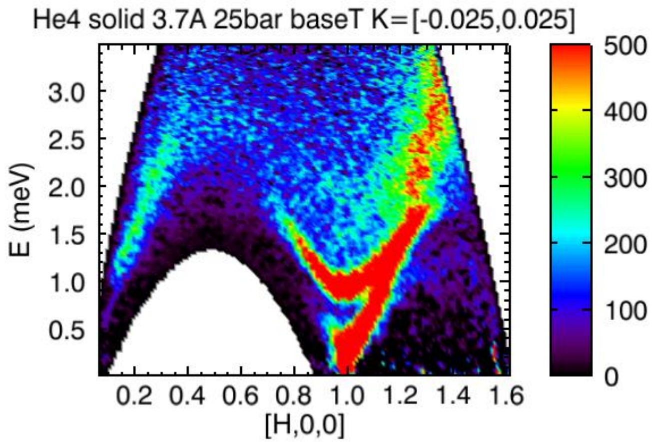


TOF on single crystals

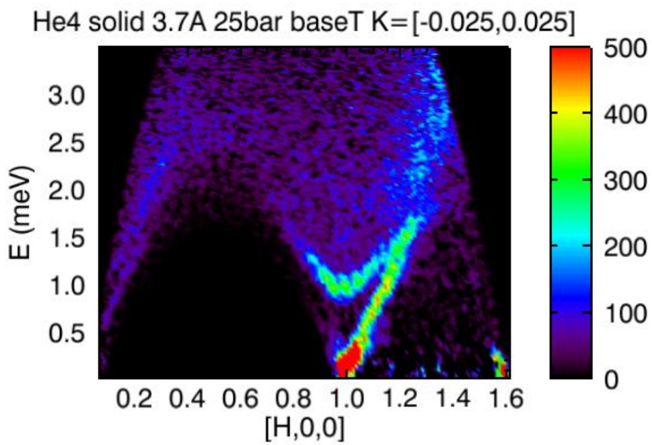
Solid helium



Angle 1

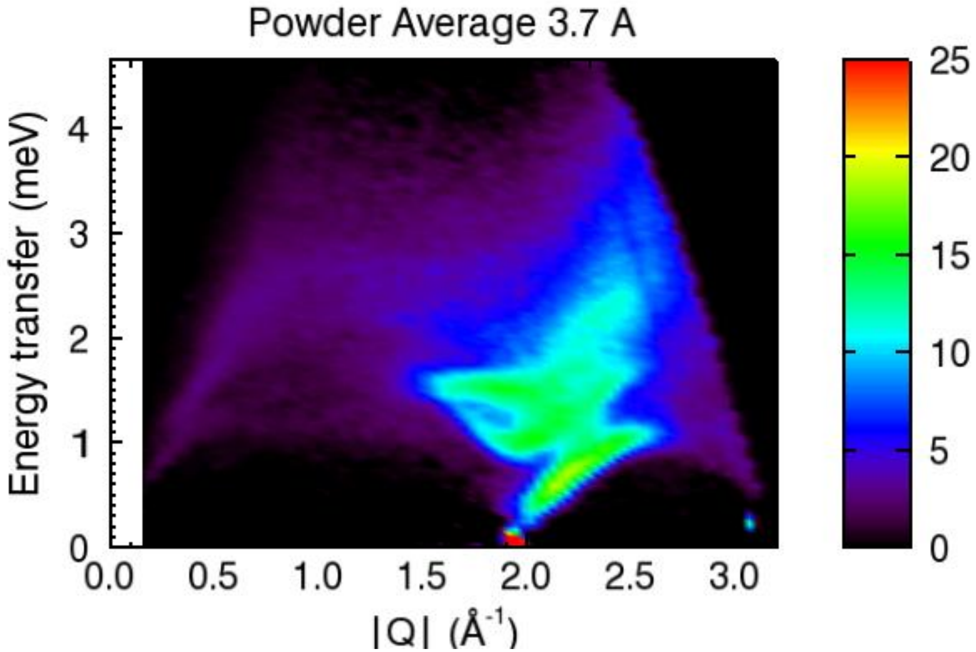


Angle 1 – 2 degrees

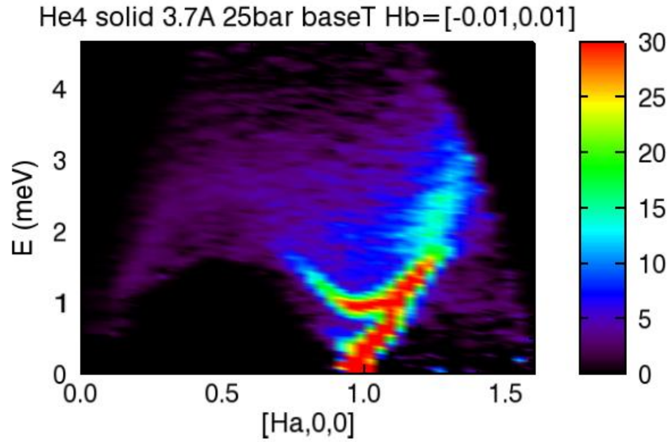


TOF on single crystals

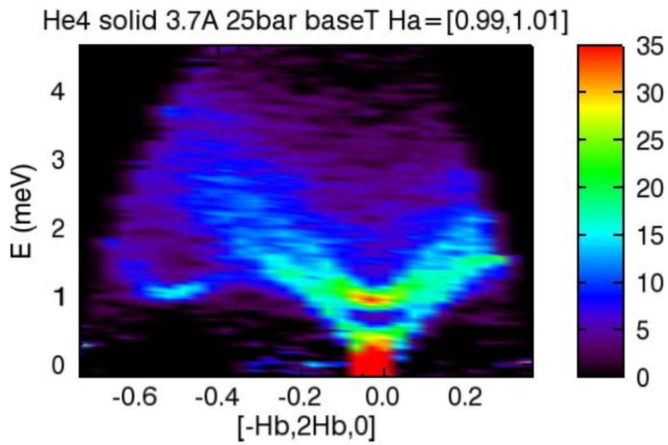
Solid helium



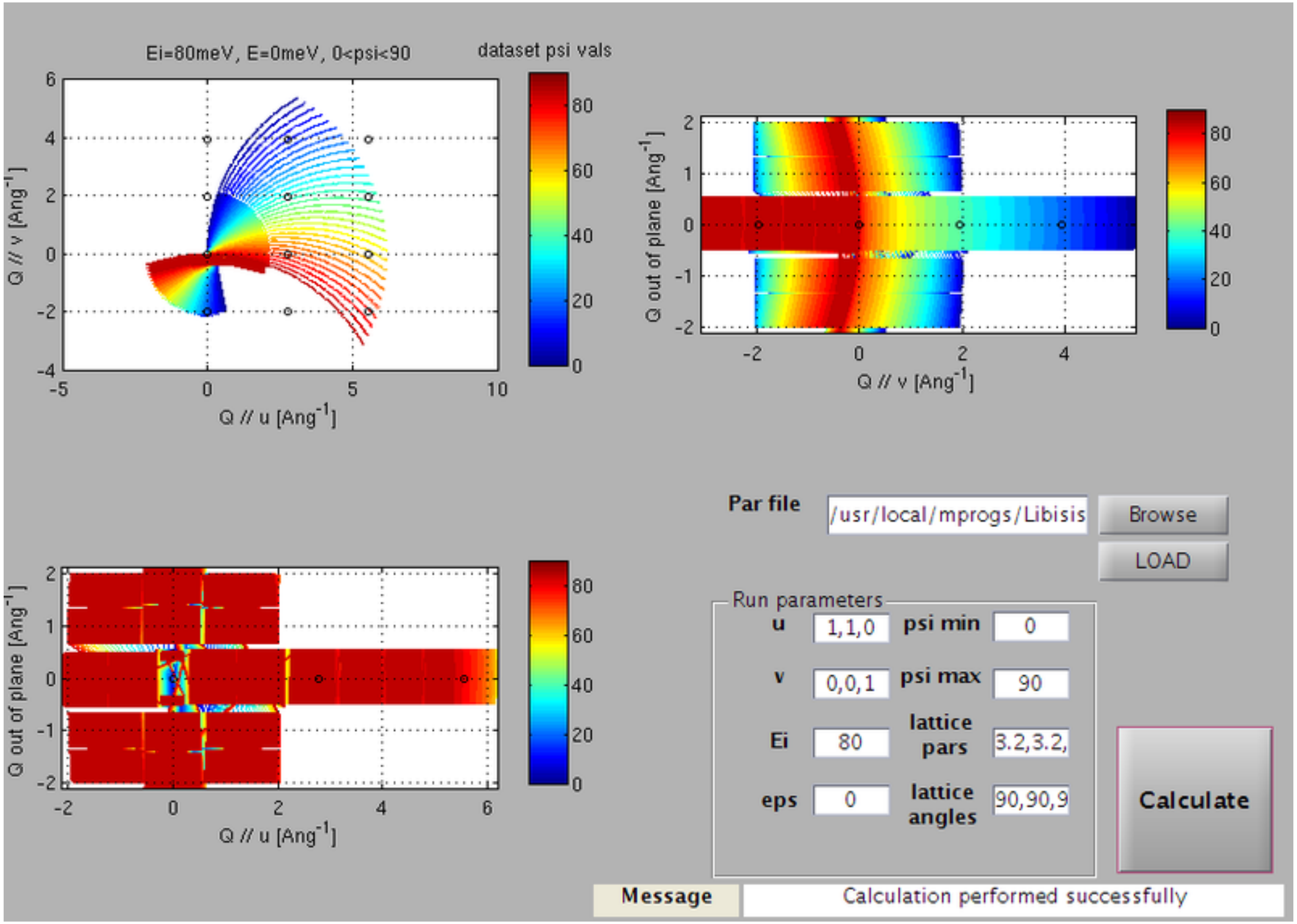
Longitudinal



Transverse



TOF on single crystals



HORACE screenshot – horace.isis.rl.ac.uk

Complete dataset in **Q** and *ω* space

Headings *et al.*,

PRL **105**, 247001 (2010) PHYSICAL REVIEW LETTERS week ending 10 DECEMBER 2010

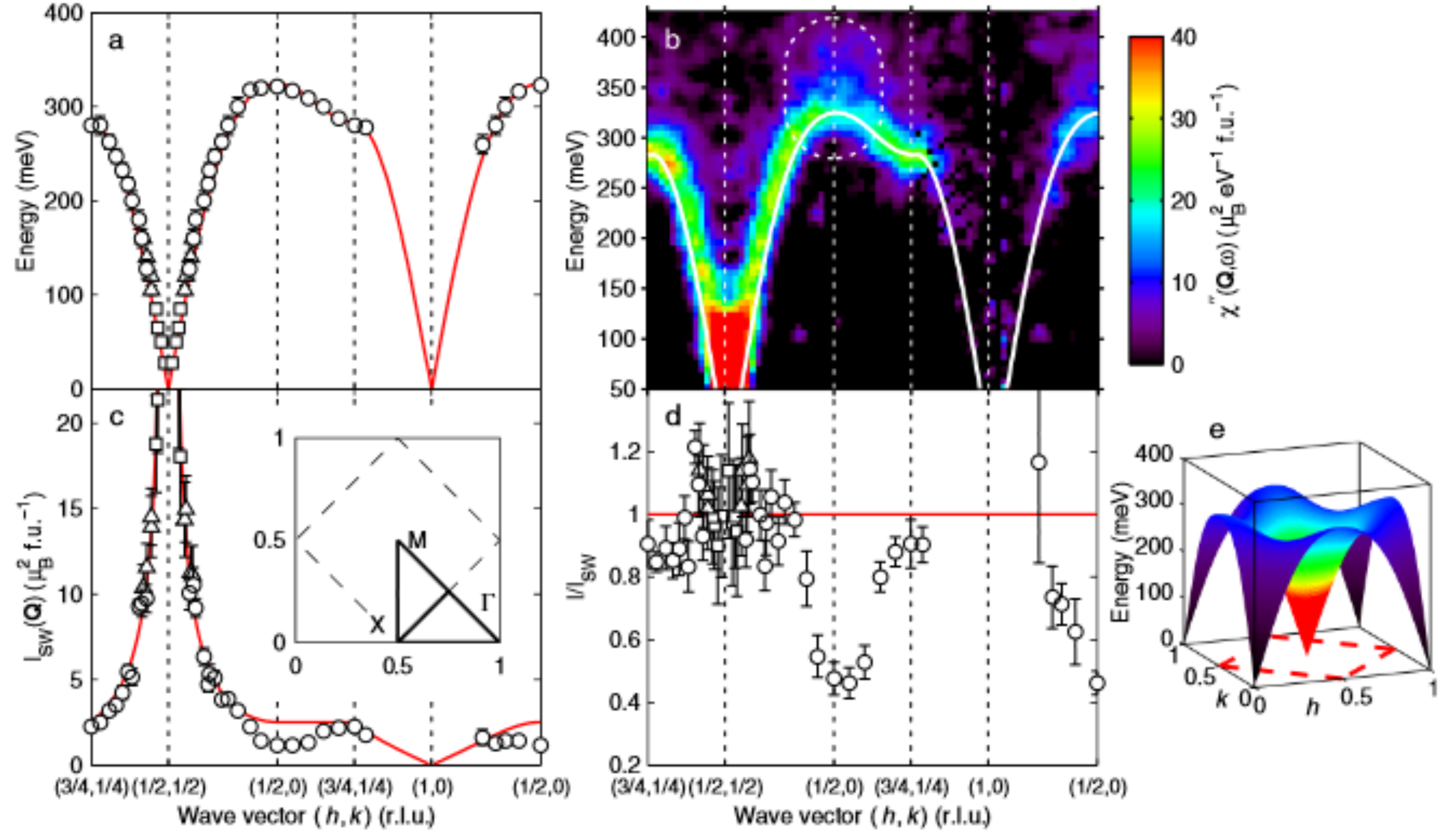
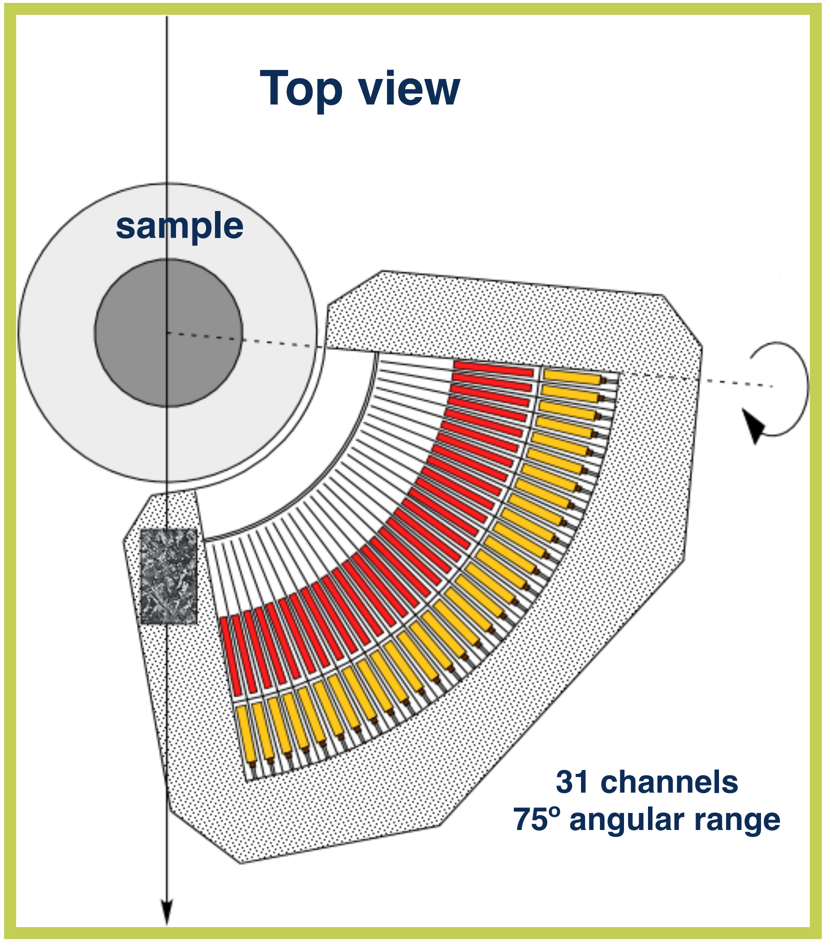


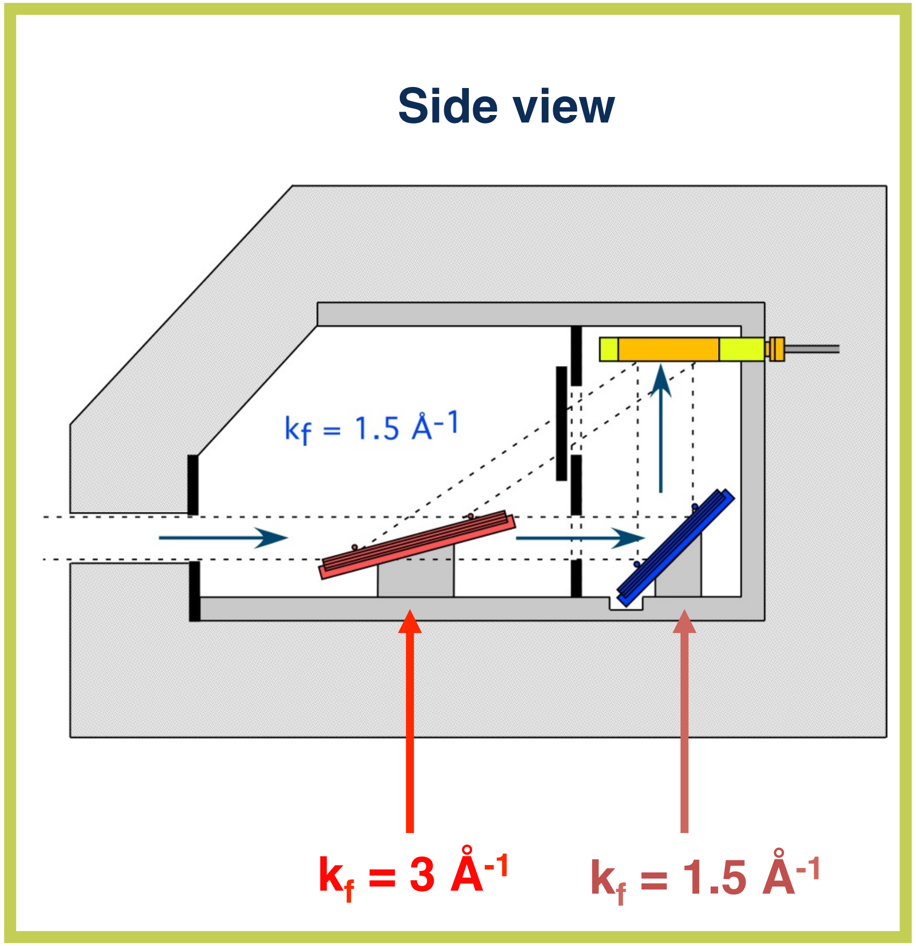
FIG. 2 (color online). **q** dependence of the magnetic excitations in La_2CuO_4. (a) One-magnon dispersion ($T = 10$ K) along lines in (c, inset). Symbols indicate E_i: 160 meV (□), 240 meV (△), and 450 meV (○). The solid line is a SWT fit based on Eq. (1). (b) Measured $\chi''(\mathbf{q}, \omega)$. 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).

Appendix

FLAT CONE

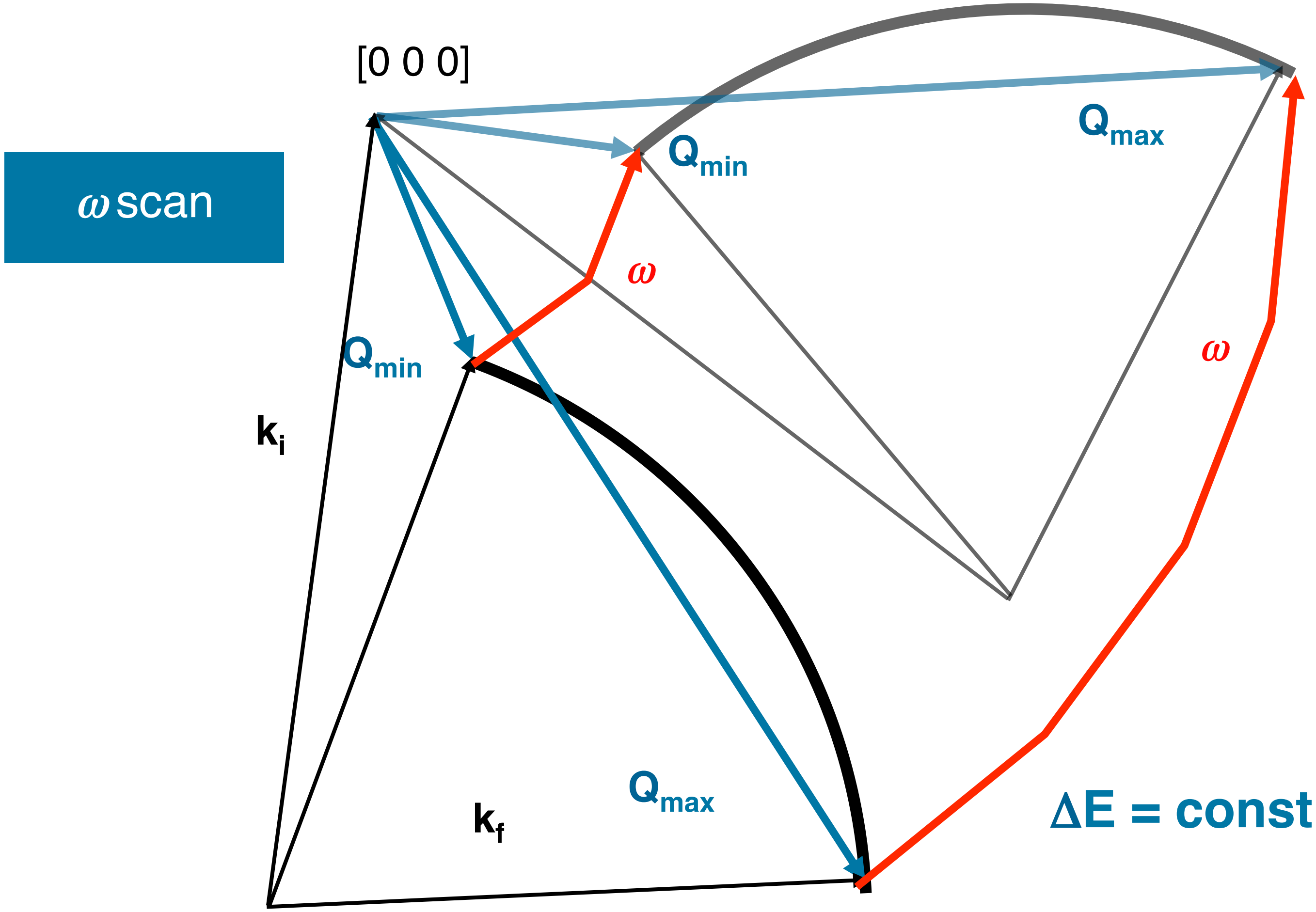


Top view
sample
31 channels
75° angular range



Side view
kf = 1.5 Å-1
kf = 3 Å-1
kf = 1.5 Å-1

FC scan modes



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