Inelastic Scattering I

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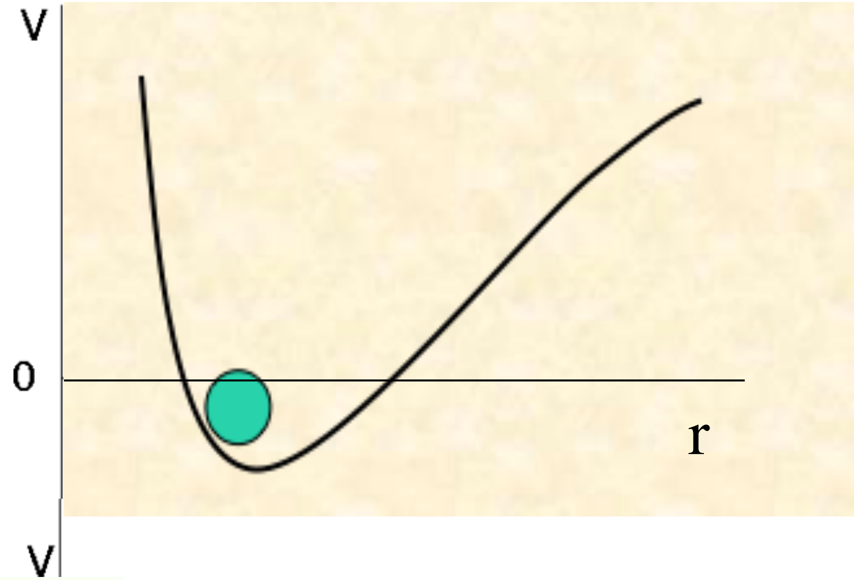
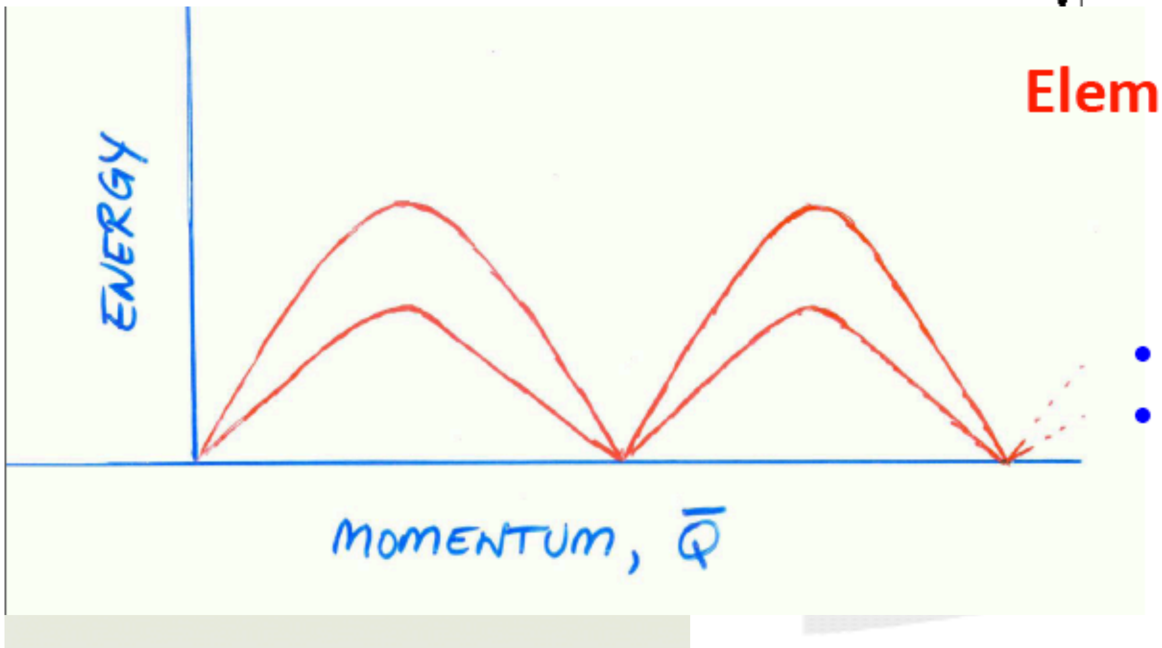
INELASTIC NEUTRON SCATTERING

Fundamental Information on Interactions in Materials

Structural probes yield indirect information on interactions in materials by locating the minimum of the potential. Dynamical probes, including neutron scattering, reveal information on the shape of the potential.

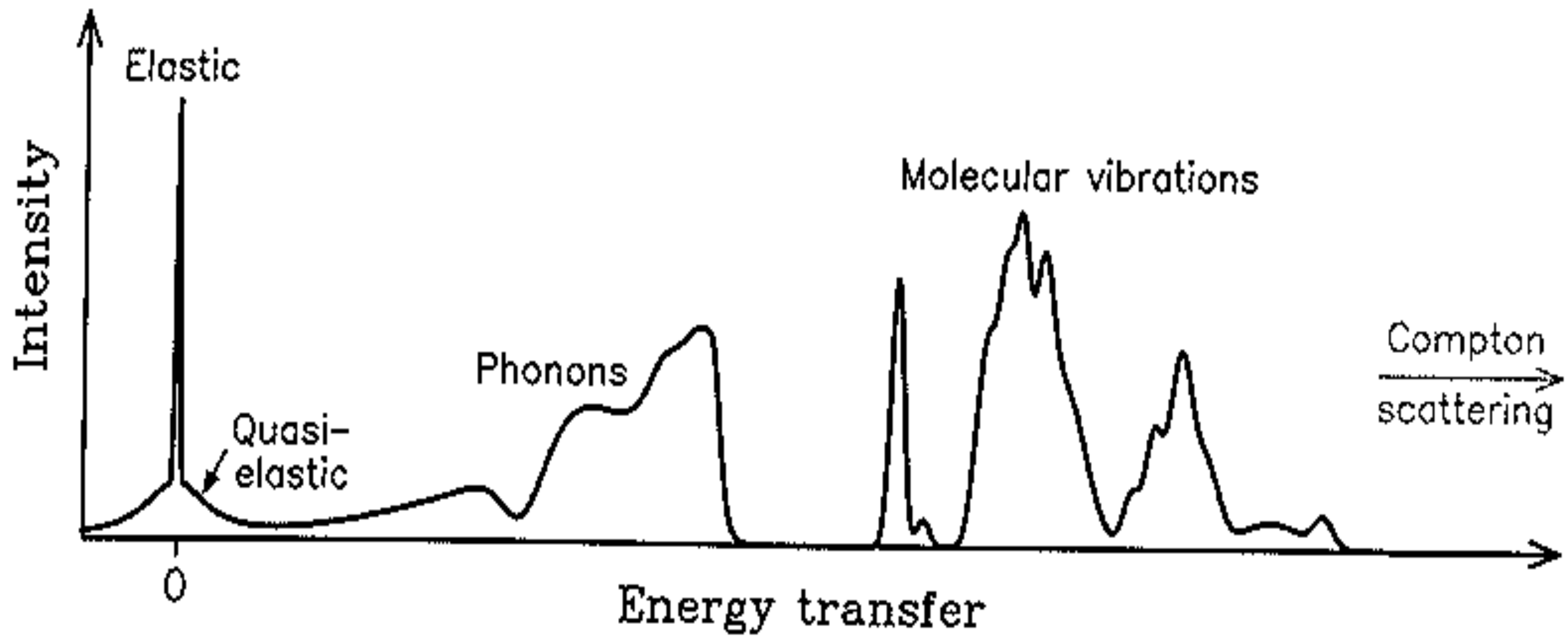




Elementary Excitations in Solids

- Lattice Vibrations (Phonons)
- Spin Fluctuations (Magnons)

A sketch of collective and single-particle excitations



From: "Elementary Scattering Theory For X-ray and Neutron Users" D.S. Sivia OUP (2011)

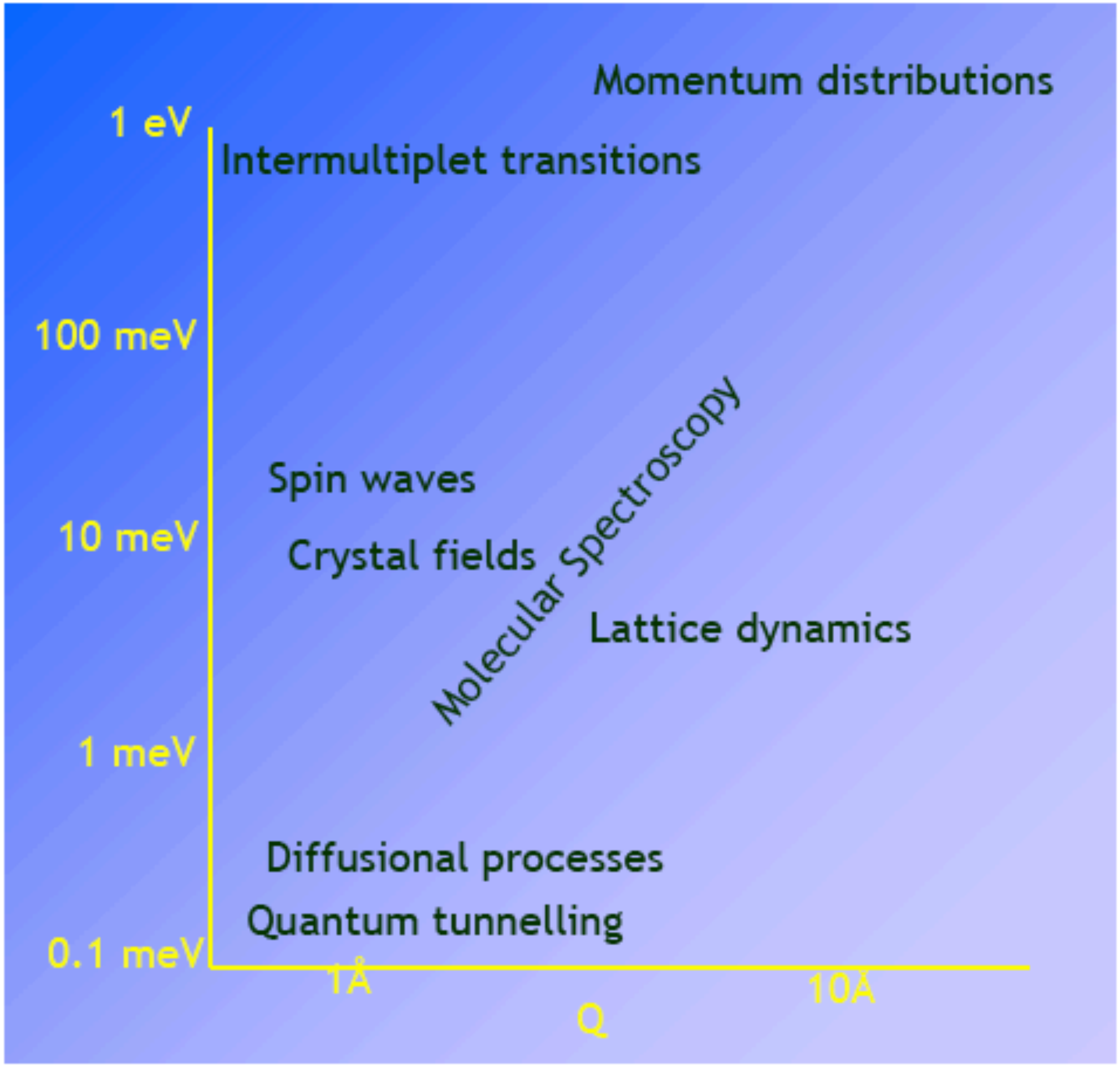
INELASTIC NEUTRON SCATTERING EXPERIMENT

- In the inelastic neutron scattering experiment, the quantity we measure is the double differential cross section

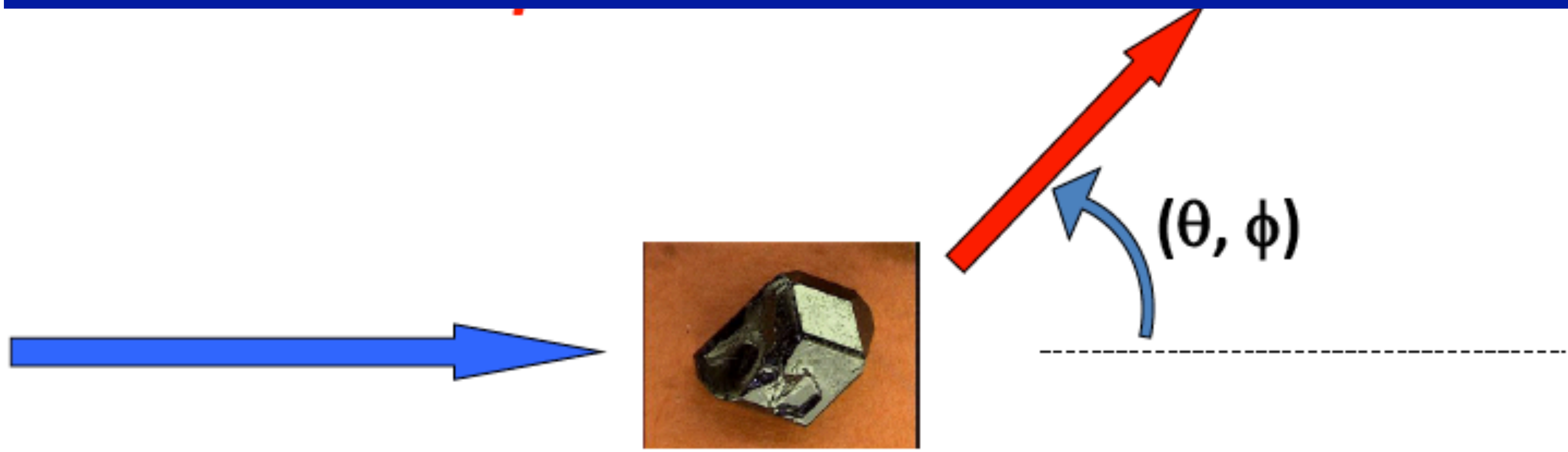
$$\frac{d^2\sigma}{d\Omega\, dE} = \frac{\mathbf{k}_f}{\mathbf{k}_i} b^2 S(\mathbf{Q}, \omega)$$

- It is the scattering function $S(Q,\omega)$ which provides the link between the scattering data and the physical system being studied.
- The type of experiment will dictate the portions of (Q,ω) space which are to be probed.

Regions of interest in (Q,ω) space



INELASTIC NEUTRON SCATTERING: THE BASIC EXPERIMENT



Incident Beam:

- monochromatic
- "white"

Scattered Beam:

- Resolve its energy
- Don't resolve its energy
- Filter its energy

ANALYSING THE SCATTERING TRIANGLE

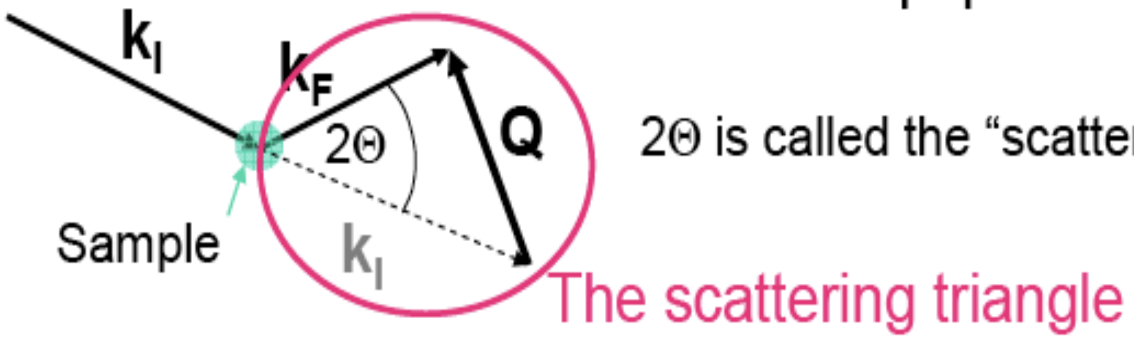
Determine the probability of finding a given change $\hbar\mathbf{Q}$ from the momentum $\hbar\mathbf{k}_I$ of a neutron incident on the specimen to the momentum $\hbar\mathbf{k}_F$ of the neutron scattered from the specimen.

In other words:

Measure the momentum transfer

$$\hbar\mathbf{Q}=\hbar\mathbf{k}_I-\hbar\mathbf{k}_F$$

or: $\mathbf{Q}=\mathbf{k}_I-\mathbf{k}_F$ (wave number notation)



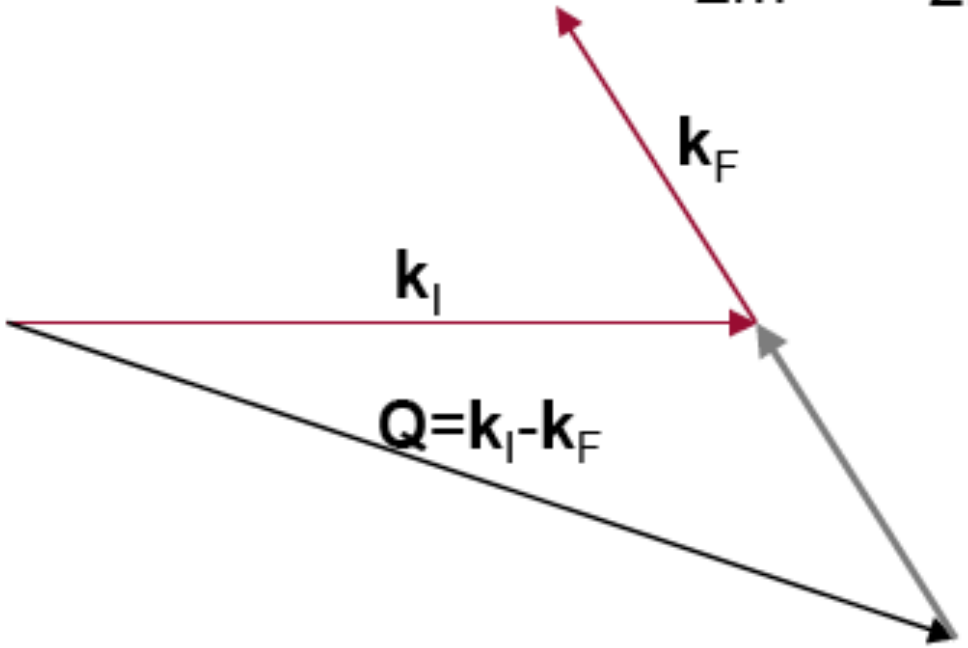
2Θ is called the "scattering angle"

ANALYSING THE SCATTERING TRIANGLE

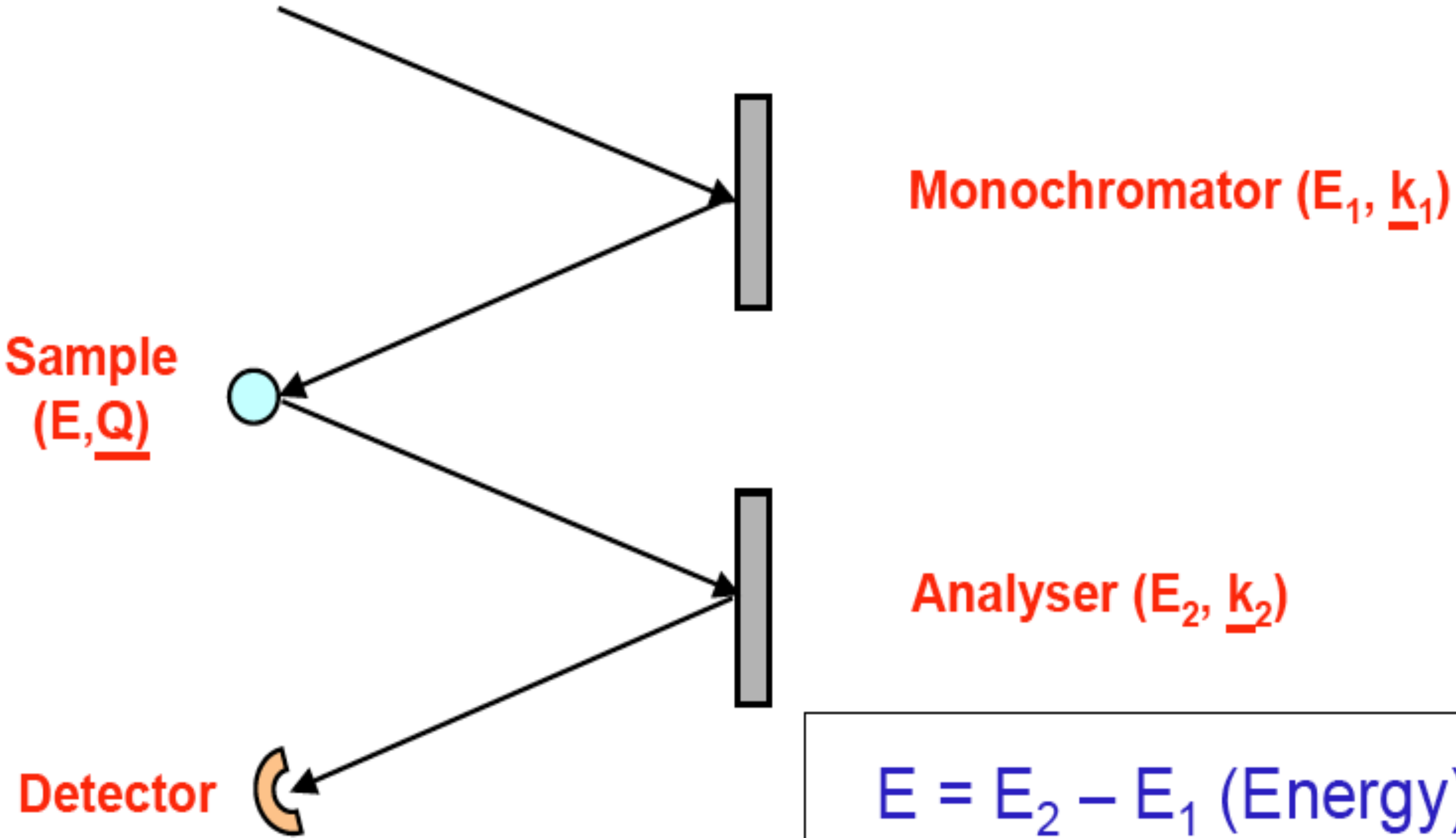
Basic relations:

momentum transfer: $\mathbf{Q}=\mathbf{k}_I-\mathbf{k}_F$

energy transfer: $\Delta E = \frac{\hbar^2 k_I^2}{2m} - \frac{\hbar^2 k_F^2}{2m} = \hbar\omega$



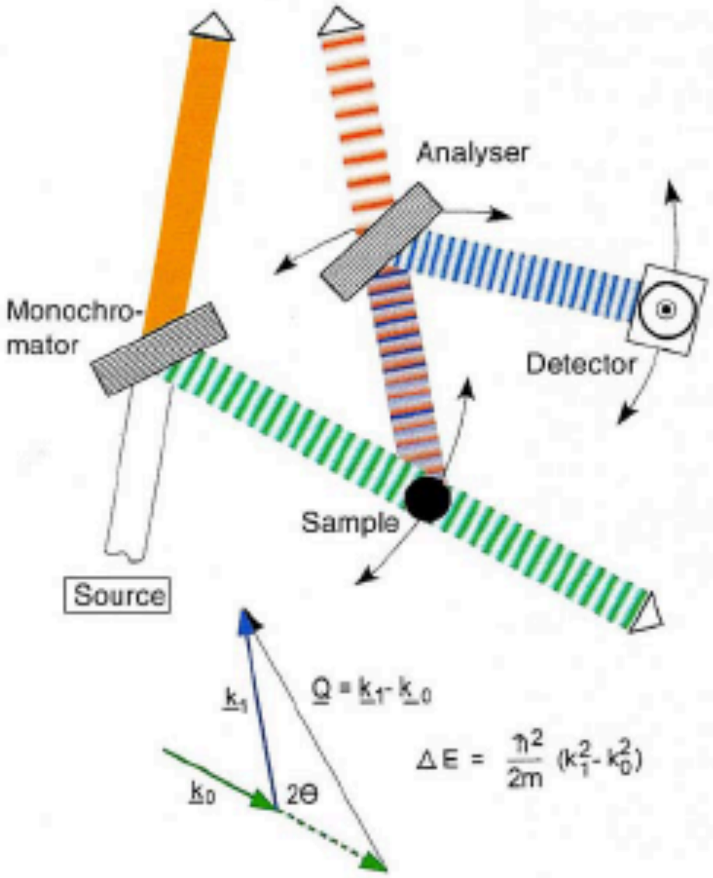
INELASTIC NEUTRON SCATTERING EXPERIMENT

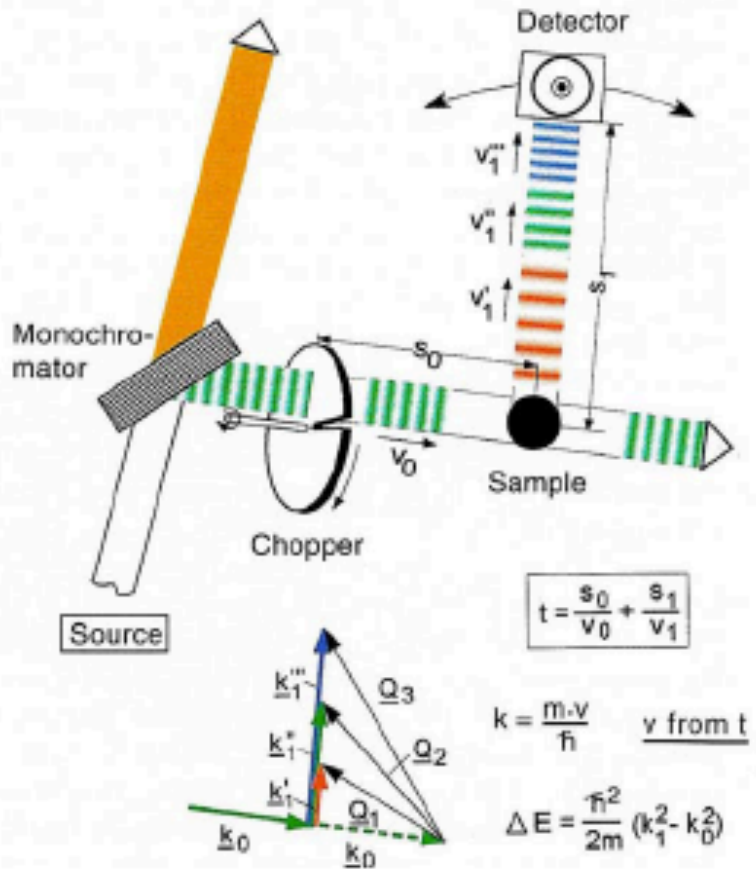


$$E = E_2 - E_1 \text{ (Energy)}$$

$$\underline{Q} = \underline{k}_2 - \underline{k}_1 \text{ (Momentum)}$$

NEUTRON SPECTROSCOPY ON A CONTINUOUS SOURCE

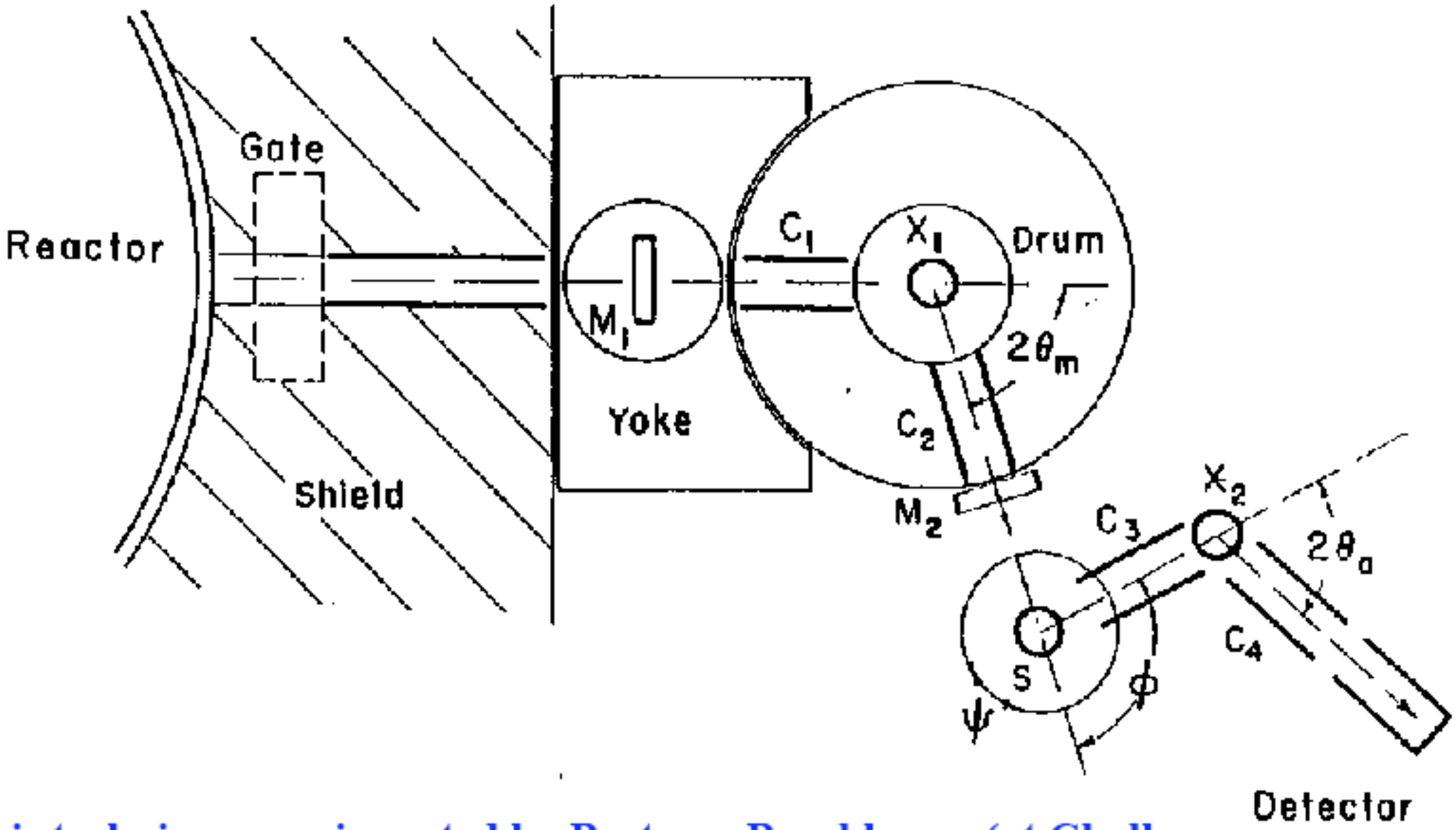




The triple axis spectrometer measures one Q-value at a time. For a scan two of the three axes must be activated

The time of flight spectrometer allows to measure several Q values at each detector setting simultaneously. Multi-detectors are generally used.

Triple Axis Spectrometer

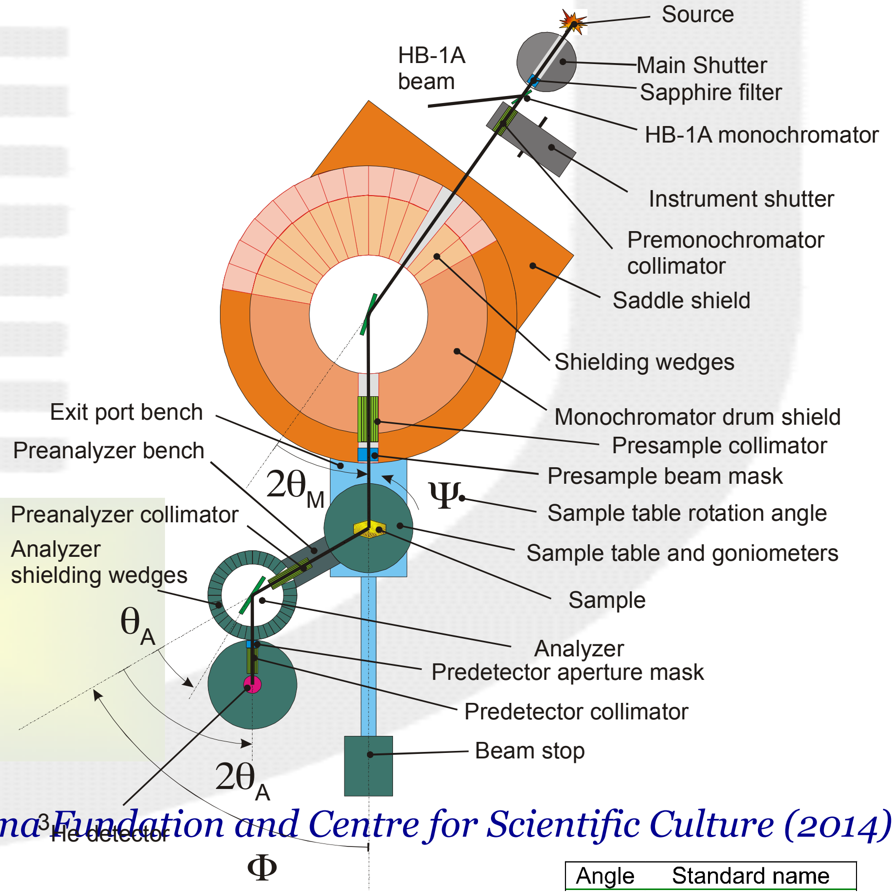


This technique was invented by Bertram Brockhouse (at Chalk River National Lab in Canada) [Nobel Laureate in 1994] in the 1950s and although the instruments have improved beyond recognition, the basic technique has not changed.

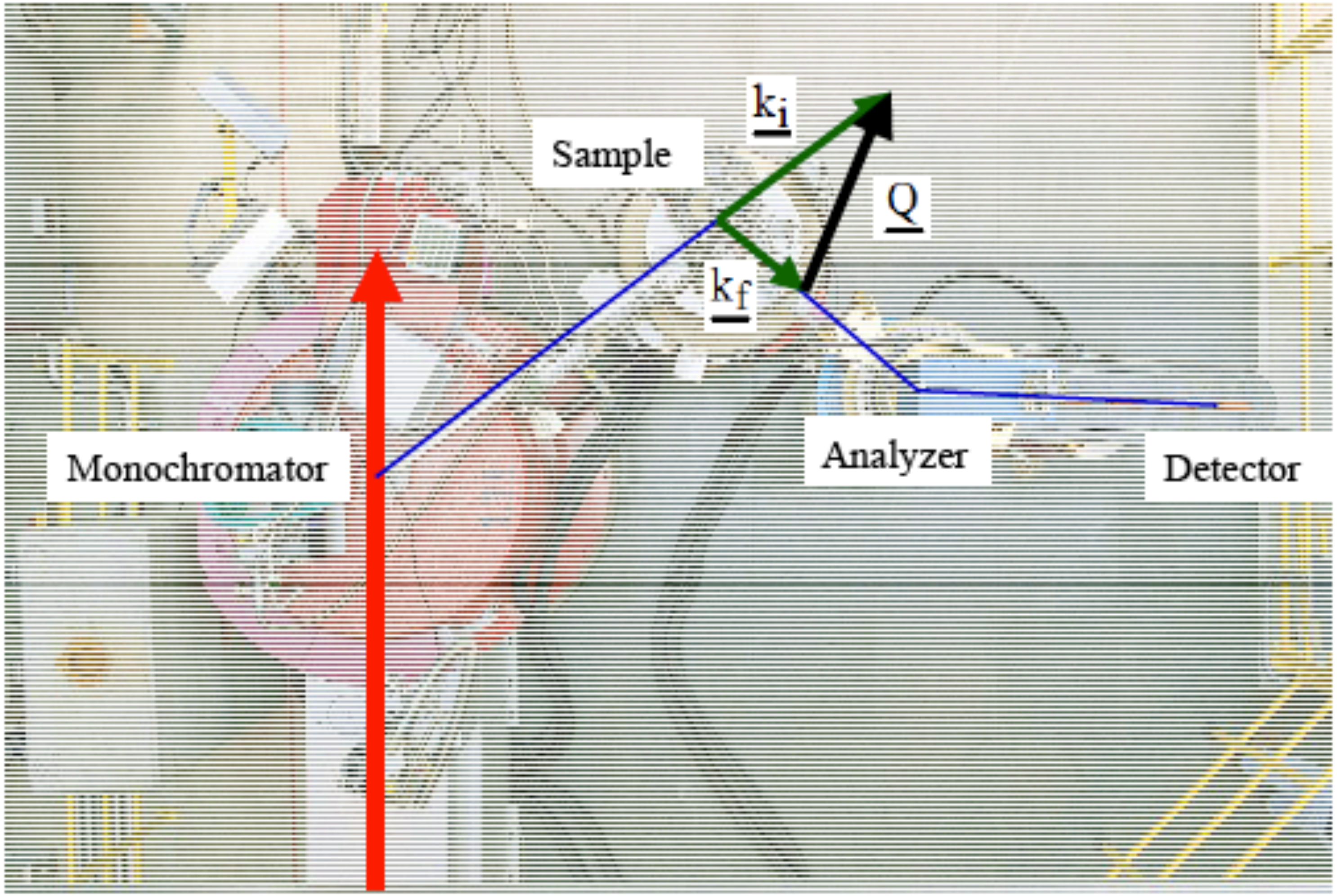
TRIPLE AXIS SPECTROMETER

"old" HB3, HFIR →

"new" HB1, HFIR ↓



TRIPLE AXIS SPECTROMETER



$$|\underline{k}| = \frac{2\cdot\pi}{\lambda}$$

$$E_i = \frac{\hbar^2\cdot \underline{k}_i^2}{2\cdot m}$$

$$E_f = \frac{\hbar^2\cdot \underline{k}_f^2}{2\cdot m}$$

$$E = \hbar\,\omega$$

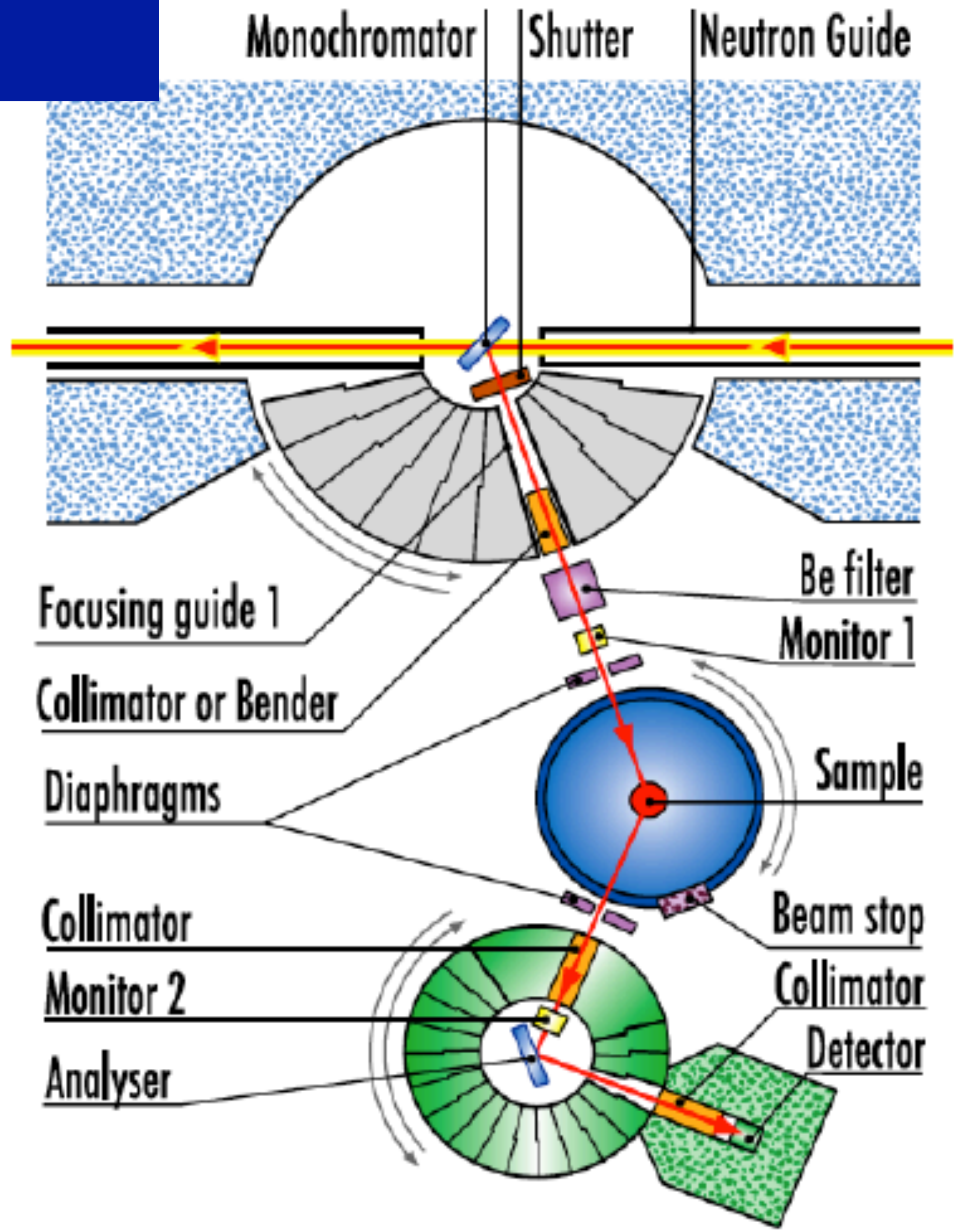
Energy transfer to sample $$E = \frac{\hbar^2}{2\cdot m}\left[\underline{k}_i^2 - \underline{k}_f^2\right]$$

Wave vector transfer to sample $$\underline{Q} = \underline{k}_i - \underline{k}_f$$

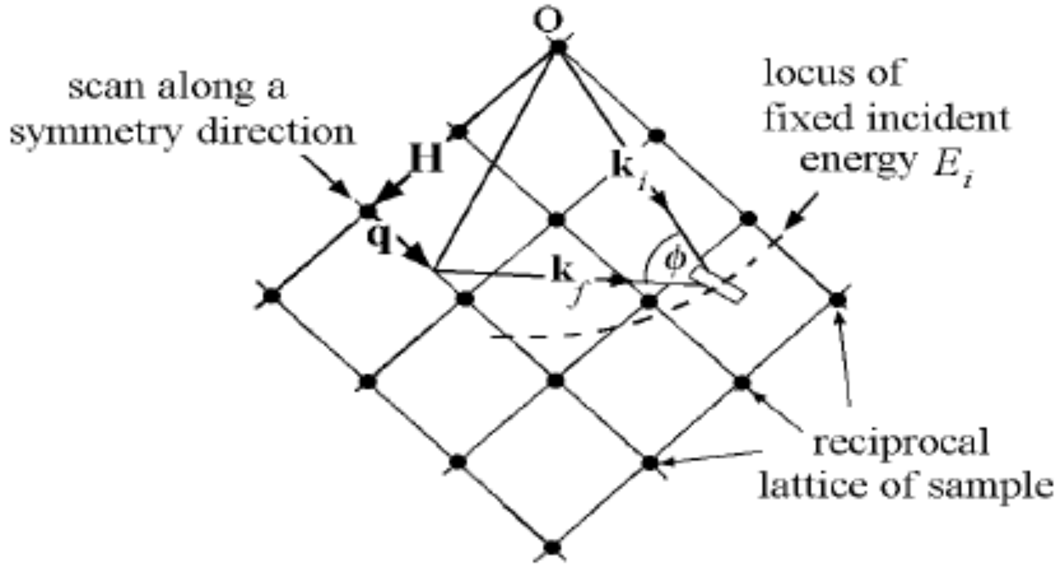
SCHEMATIC OF A TRIPLE AXIS SPECTROMETER

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

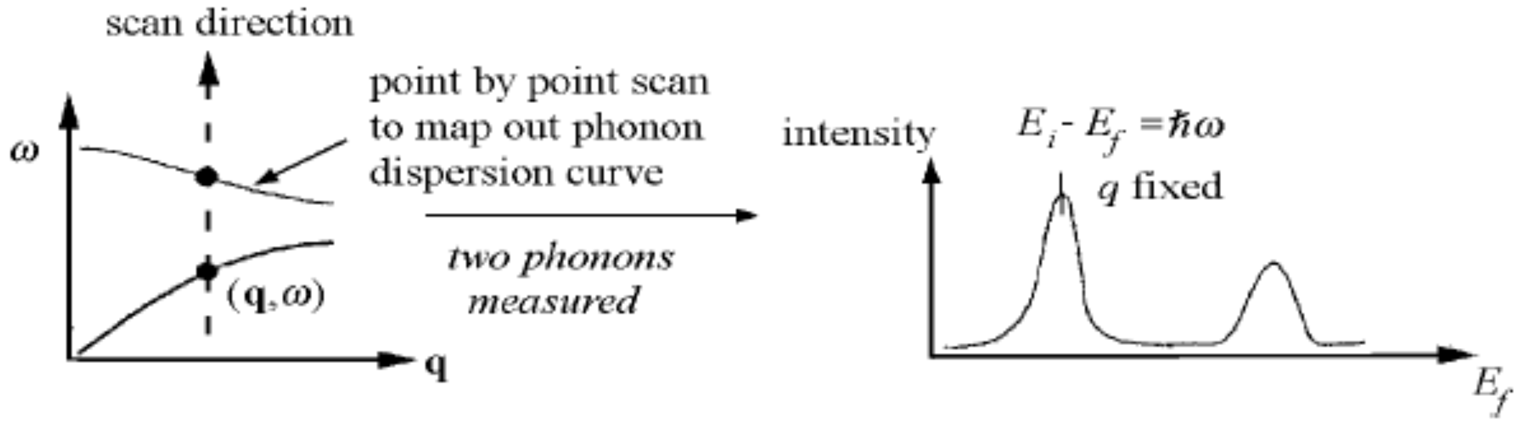
Note that the Be filter, which is used with cold neutrons, can either be before the sample (constant k_i) or after the sample (constant k_f)



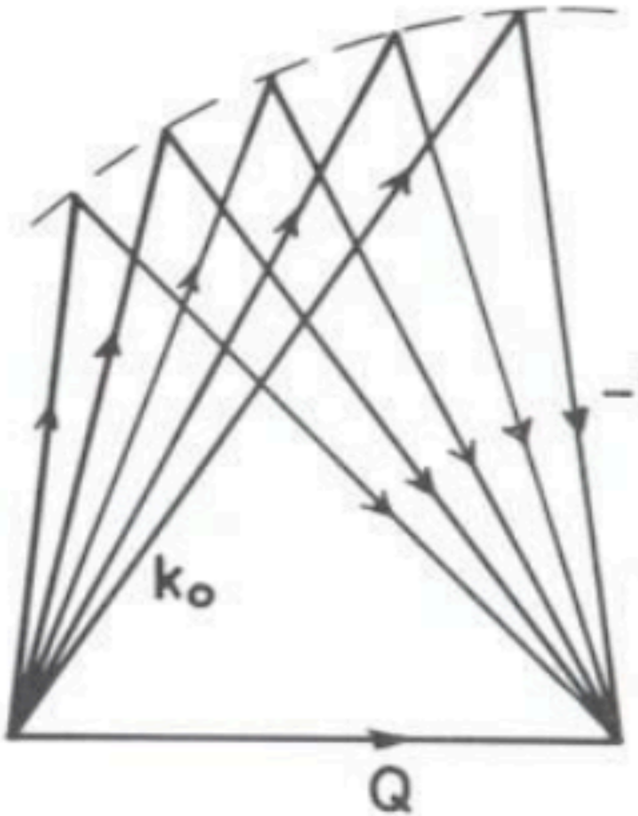
TRIPLE AXIS SPECTROMETER



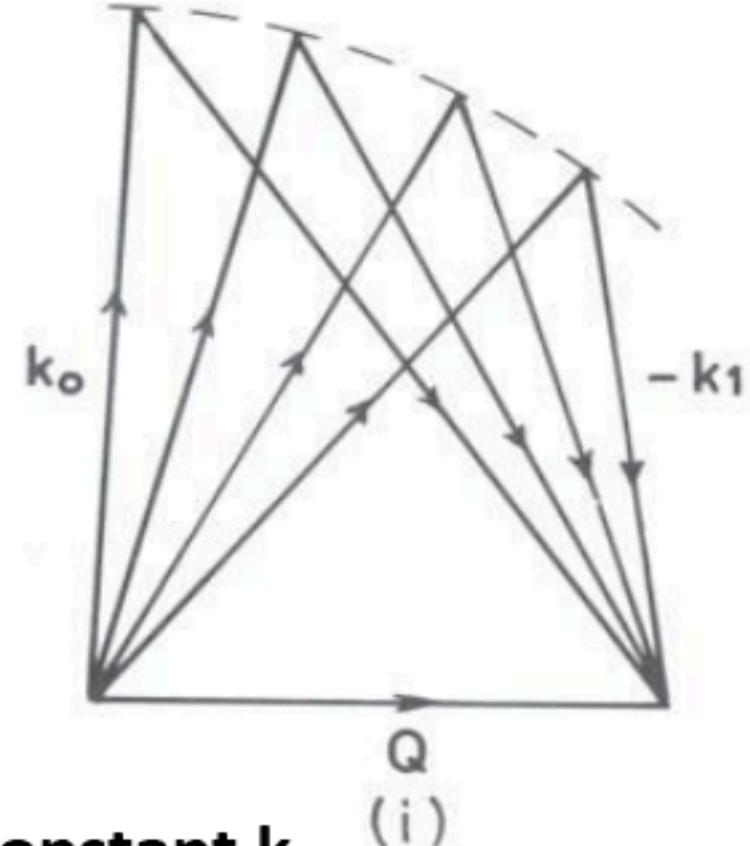
Focusing



TWO DIFFERENT WAYS OF PERFORMING CONSTANT-Q SCAN



Constant k_f



Constant k_i

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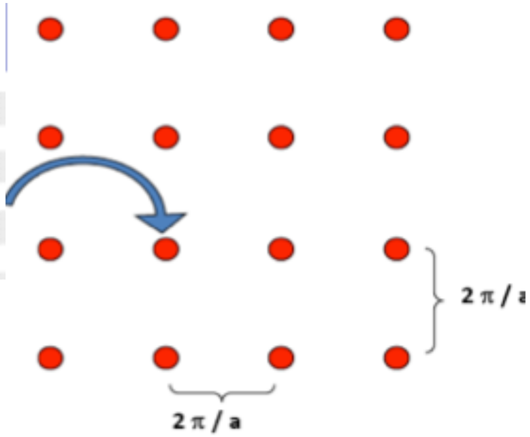
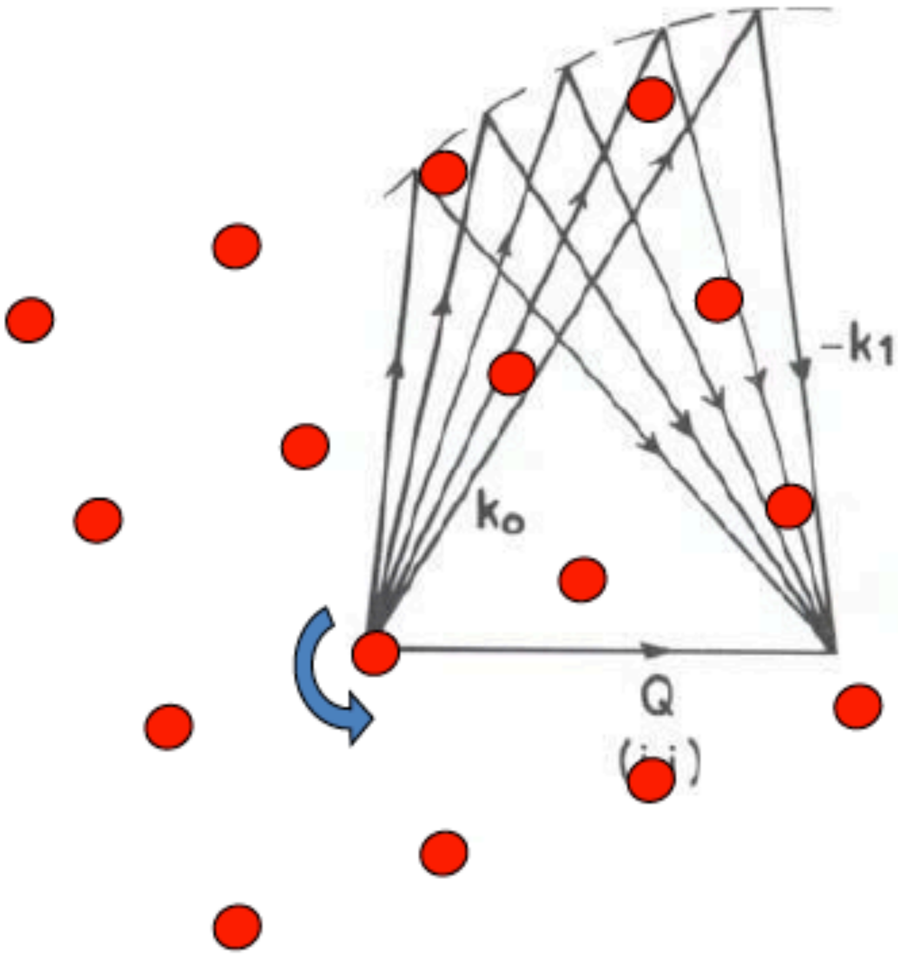
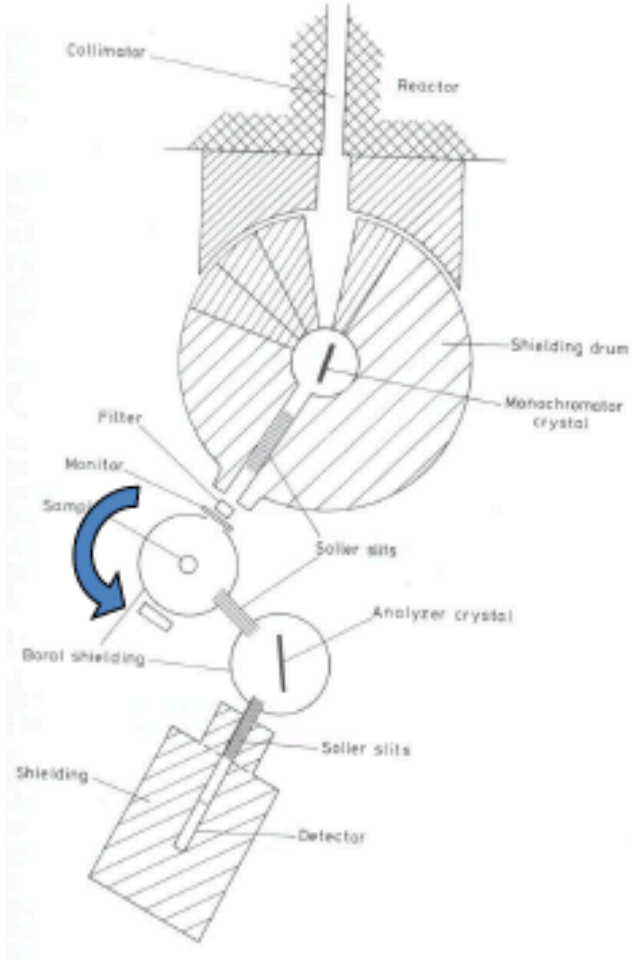
MAPPING MOMENTUM-ENERGY (Q-W) SPACE

Origin of reciprocal space;

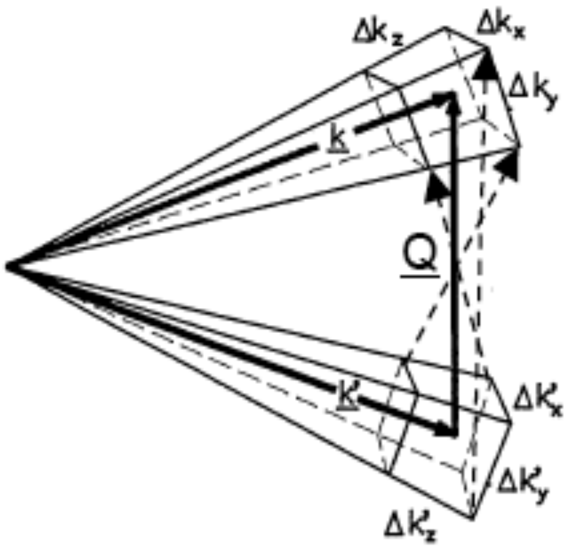
Remains fixed for any sample rotation

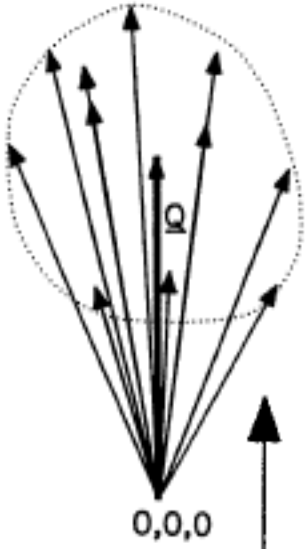






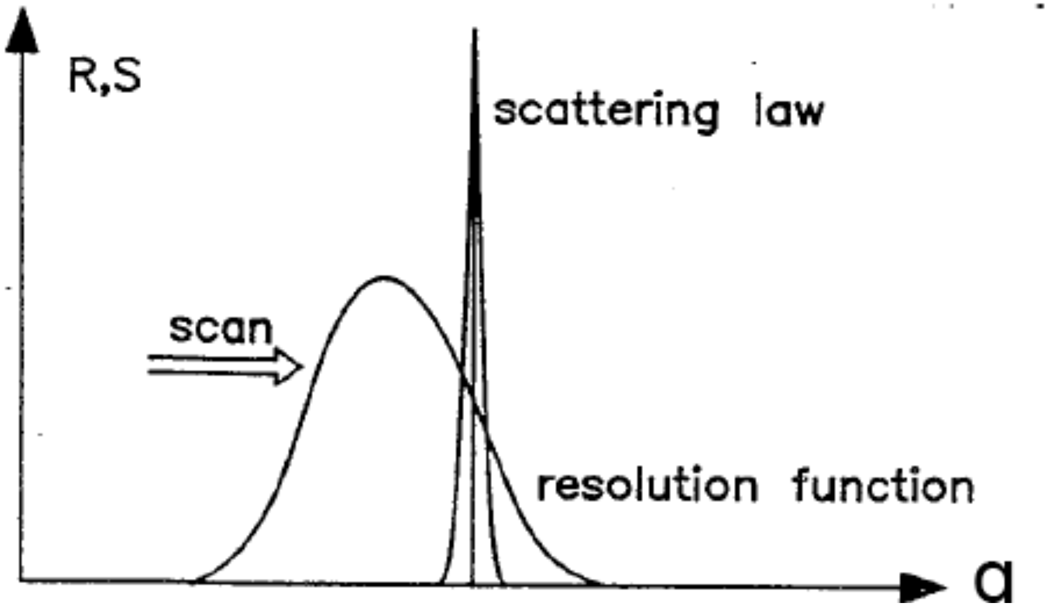
The next important question after what can be measured is, how precisely it can be measured.





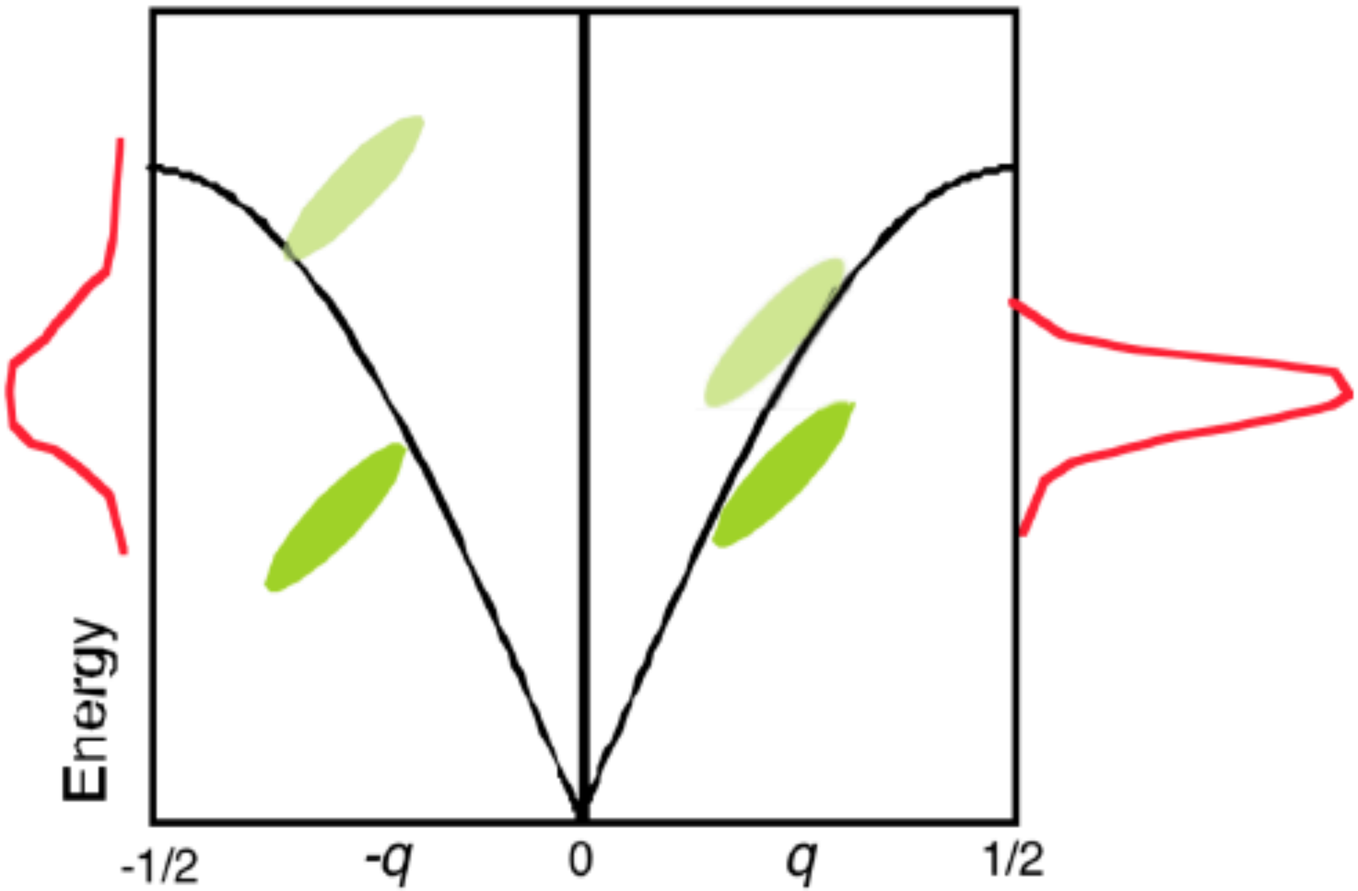
There is always a degree of uncertainty, as to how well K_i and K_f can be defined by experimental means.

Scanning a wide resolution function across a narrow scattering law essentially reproduces the resolution function



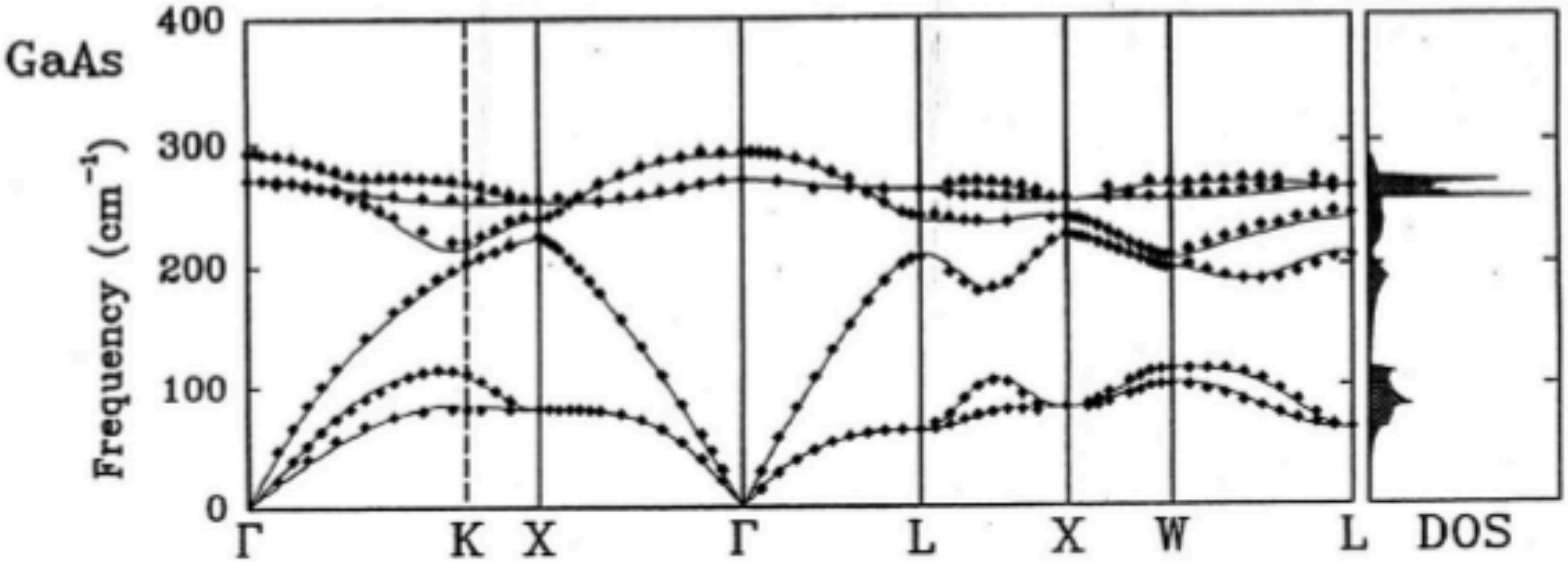
TRIPLE AXIS SPECTROMETER

- **Optimizing peak intensity**
- **Match slope of resolution to dispersion**



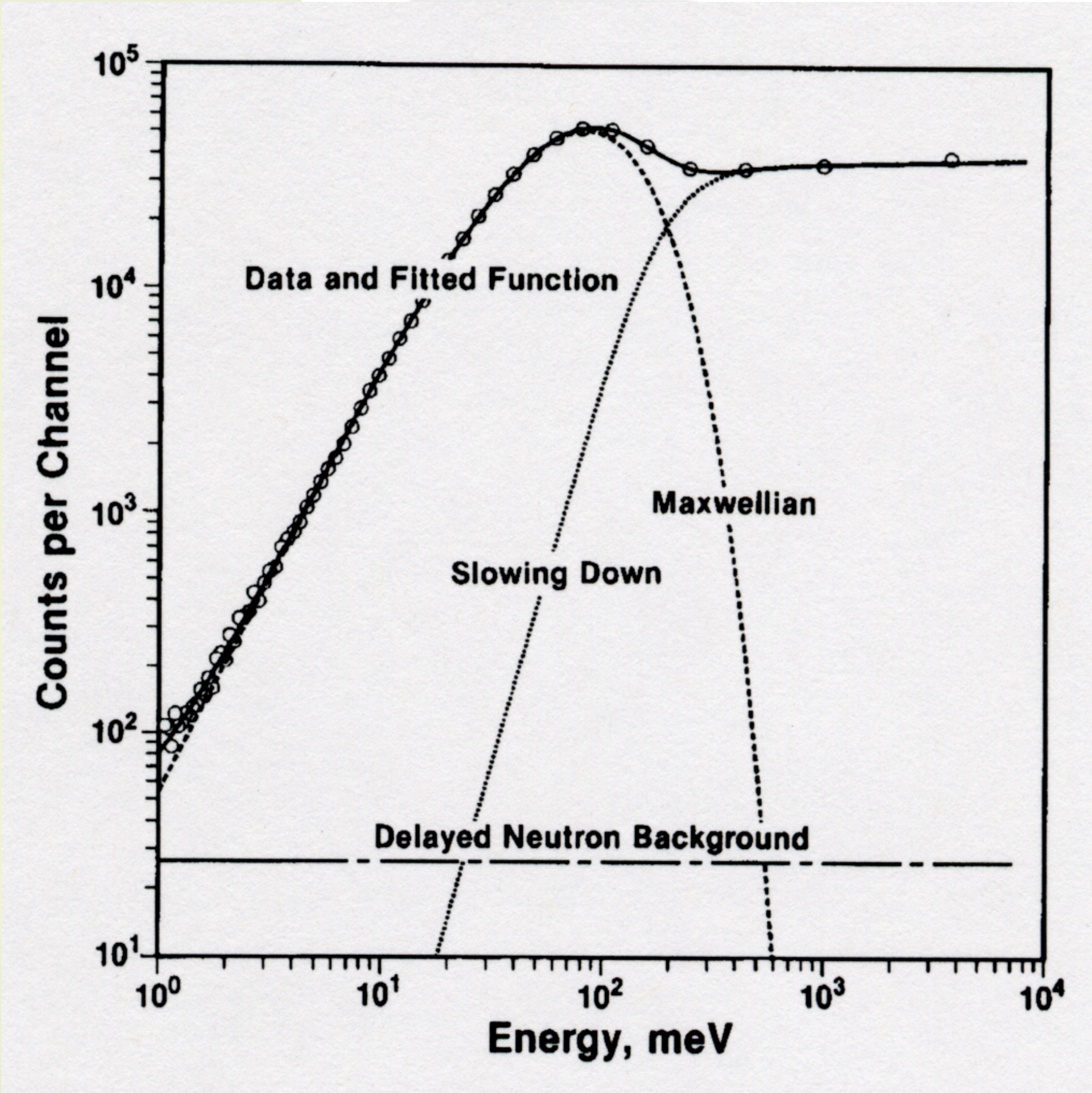
The IN14 triple-axis spectrometer at the ILL

Phonons in Gallium Arsenide, a III/V semiconductor, is used in the manufacture of devices such as microwave frequency integrated circuits



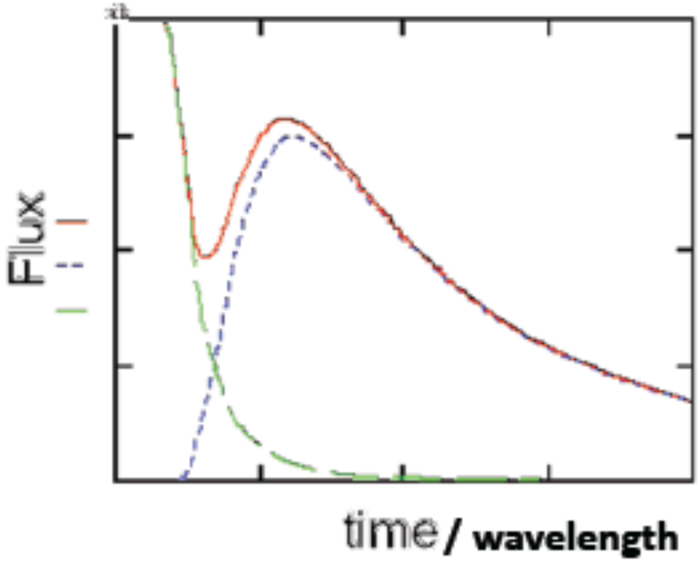
Phonon dispersion curves as measured for GaAs. Strauch & Dorner (1990). The lines give the result of *ab initio* calculations and show that the forces between the atoms are well understood. The letters below give the notation for the symmetry directions or points.

INELASTIC NEUTRON SCATTERING ON A PULSED SOURCE



From Colin Windsor, *Pulsed Neutron Sources* 1983

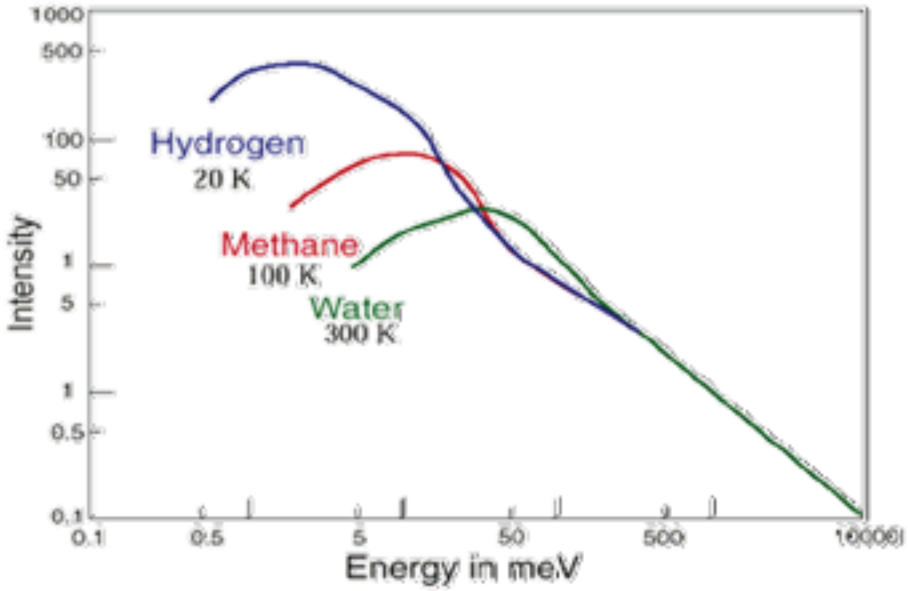
NEUTRON SPECTROSCOPY ON A PULSED SOURCE



Spallation source moderators under-moderate the neutrons to produce a high flux of epi-thermal neutrons

- - - - - - Moderated Neutrons

— — — — Under-moderated neutrons



The energy (or wavelength) range of the produced neutrons can be changed by controlling the temperature of the moderator

TIME OF FLIGTH (TOF) TECHNIQUES

- Velocity of thermal neutrons ~ km s^{-1}.
- Hence their energy can be determined by measuring time-of-flight over a distance of a few metres.
- All spectrometers at pulsed sources use time-of-flight. On steady state sources pulsing devices such as choppers are required.

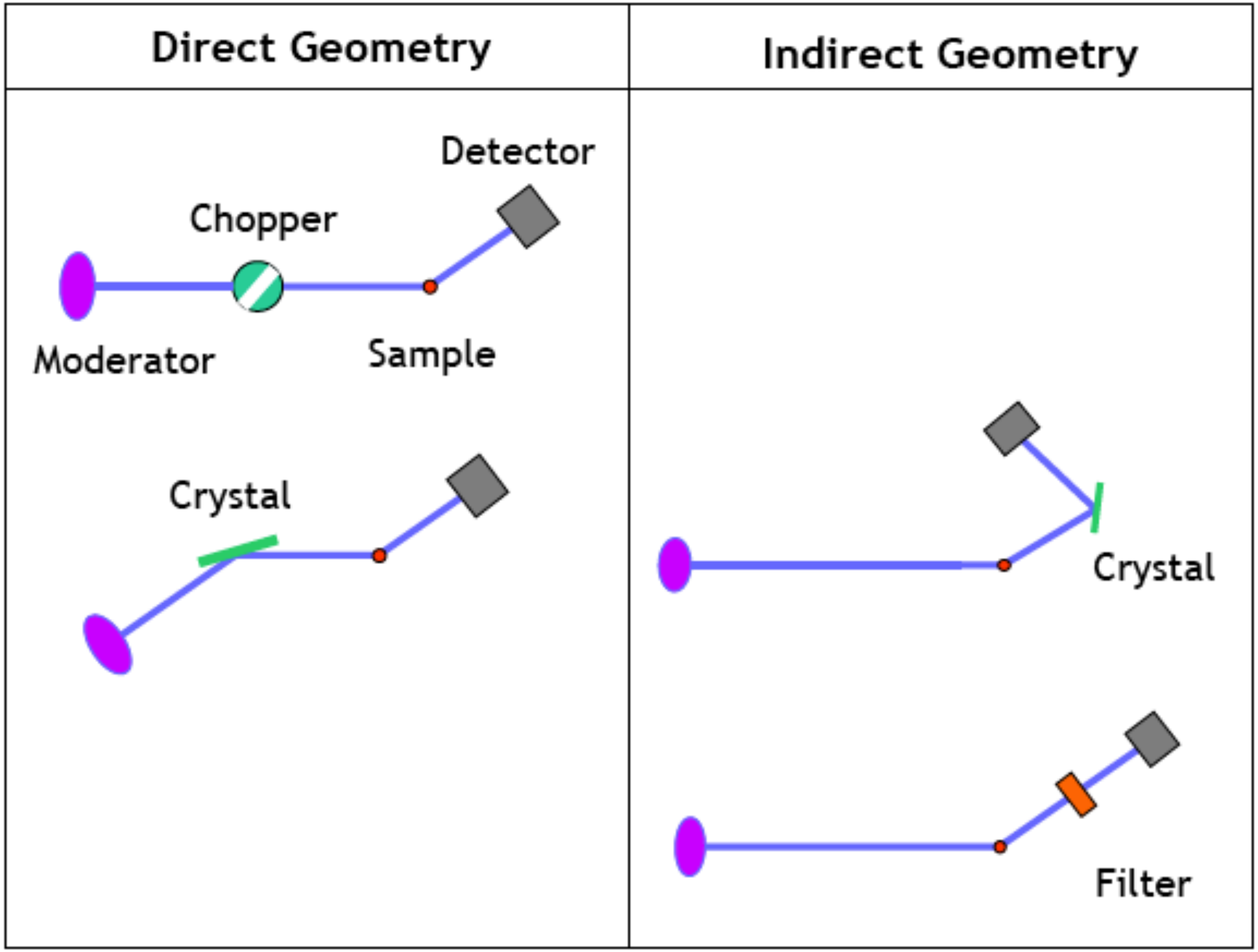
$$Q = k_i - k_f$$

$$\hbar\omega = E_i - E_f$$

TIME OF FLIGTH (TOF) SPECTROMETERS

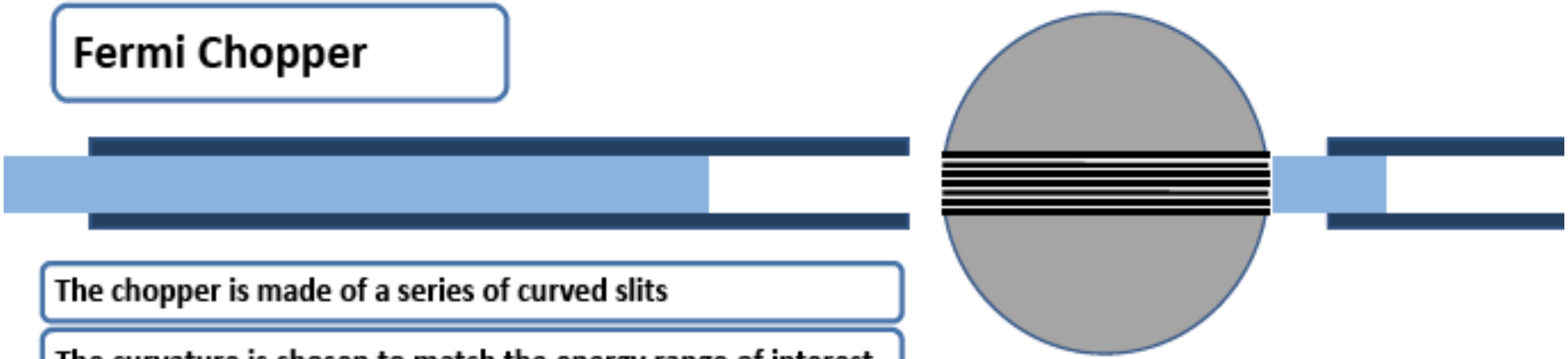
- Time-of-flight spectrometers may be divided into two classes:-
  - Direct geometry spectrometers: in which the incident energy, E_i, is defined by a device such as a crystal or a chopper and the final energy, E_f, is determined by time of flight.
  - Indirect (inverted) geometry spectrometers: In which the sample is illuminated by a white incident beam and E_f is defined by a crystal or a filter and E_i is determined by time of flight

TIME OF FLIGTH (TOF) SPECTROMETERS

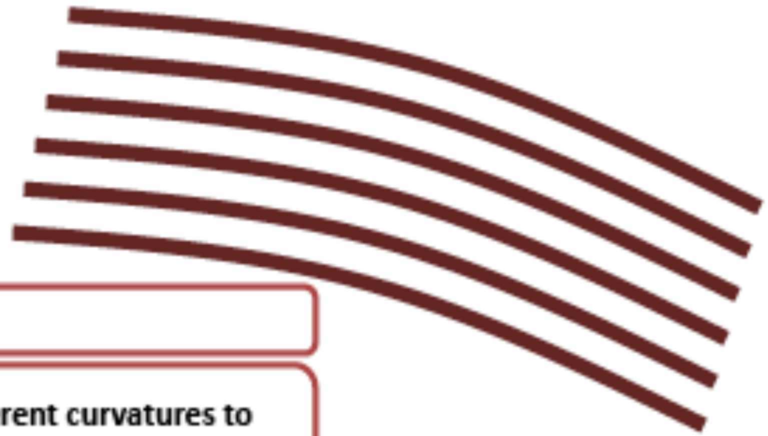


CHOOPERS

Fermi Chopper



- The chopper is made of a series of curved slits
- The curvature is chosen to match the energy range of interest
- Resolution is determined by slit width and rotation speed



Advantages

- By phasing the opening with the pulse from the target it is possible to select any neutron velocity
- Rotation speeds up to 600Hz are possible
- Usable over a wide energy range

Disadvantages

- Need several rotors with different curvatures to cover the full energy range
- Poor Transmission for low energy neutrons
- Small area – only suiable for primary flight path

Filters

- The Be filter
  - Scatters neutrons above the Bragg cut-off (5.2meV)
  - Absorbing slits absorb the scattered neutrons

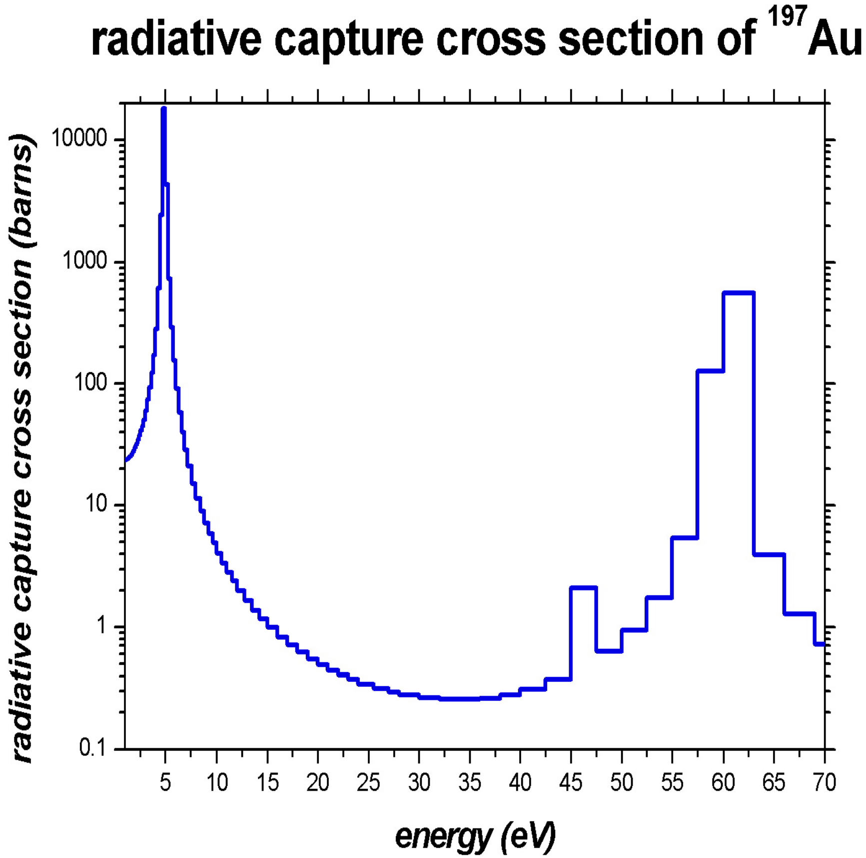
- Absorbs neutrons with energy above 5.2meV
- Cooling increases the filters effectiveness by reducing energy gain scattering from thermally excited phonons.

How to select neutrons in the range 1-100 eV ?

- ❖ **Mechanical analysers ("Choppers"):**
  - –Neutrons too fast
- ❖ **Crystal analysers**
  - -λ too short

Then…

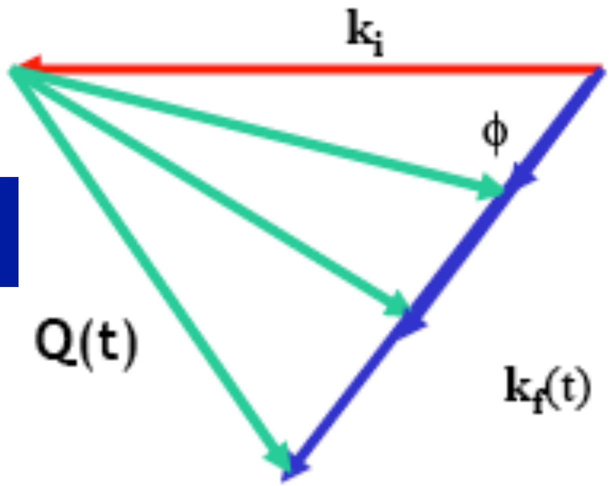
- ❖ **Nuclear resonances**
- ❖ **Two configurations possible:**
  - ❖ **RFS (Resonance Filter Spectrometer)**
  - ❖ **RDS (Resonance Detector Spectrometer)**

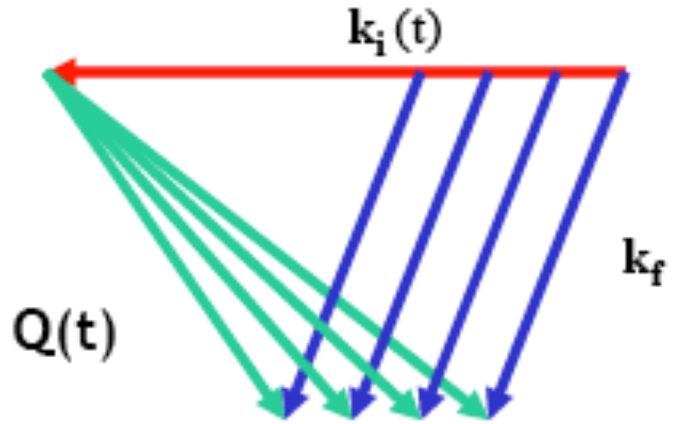


ACCESSIBLE REGION OF (Q, ω) SPACE

- To understand the trajectories traced out through (Q,ω) consider the scattering triangles.

DIRECT GEOMETRY





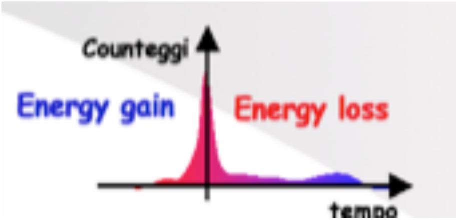
INDIRECT GEOMETRY

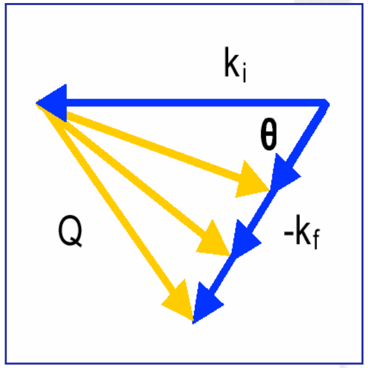
- Applying the cosine rule

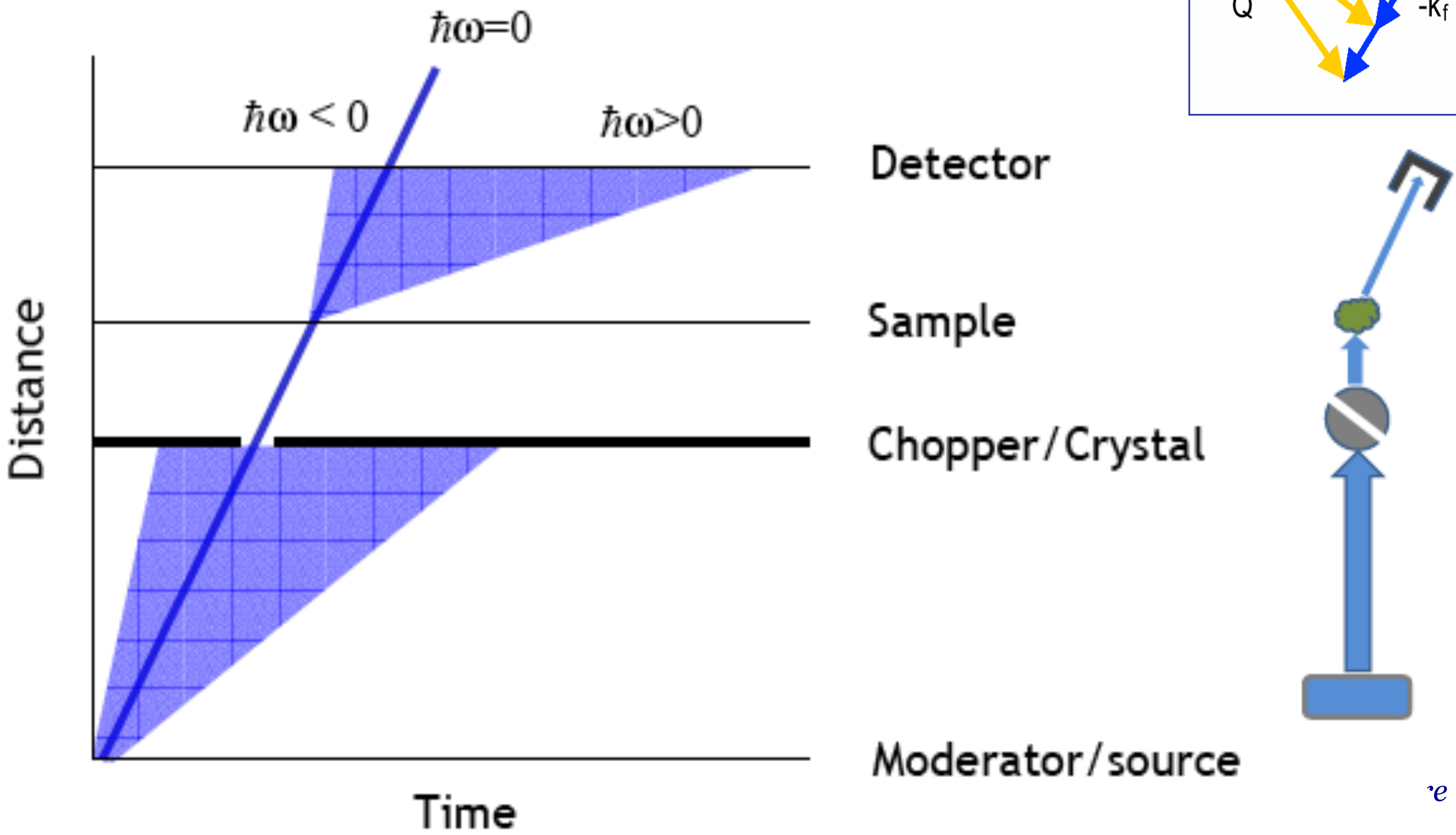
$$\mathbf{Q}^2 = \mathbf{k}_i^2 + \mathbf{k}_f^2 - 2\mathbf{k}_i\mathbf{k}_f \cos\phi$$

$$\frac{\hbar^2\mathbf{Q}^2}{2m} = E_i + E_f - 2\left(E_i E_f\right)^{1/2} \cos\phi$$

SPECTROMETERS OPERATING IN DIRECT GEOMETRY

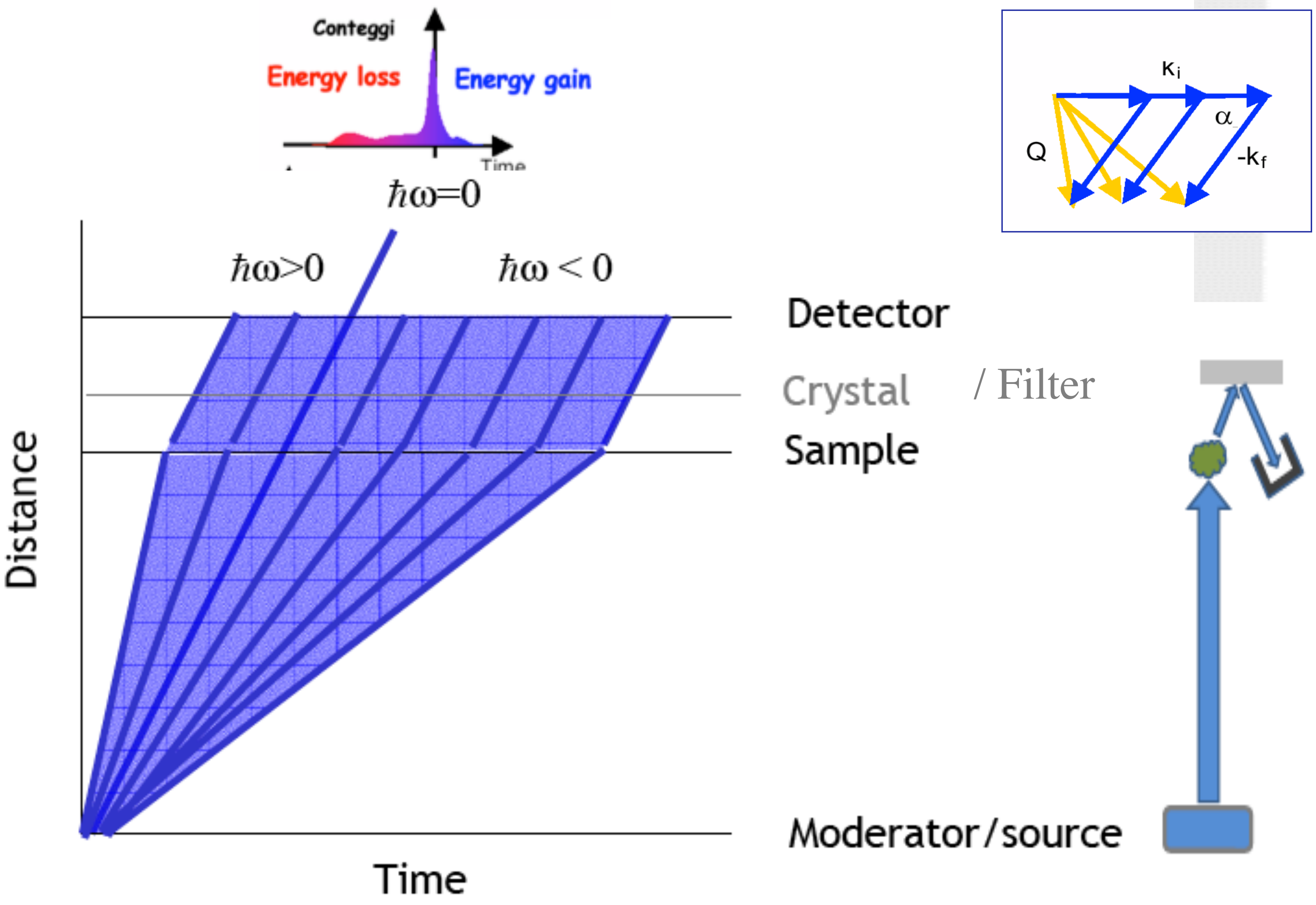






'e (2014)

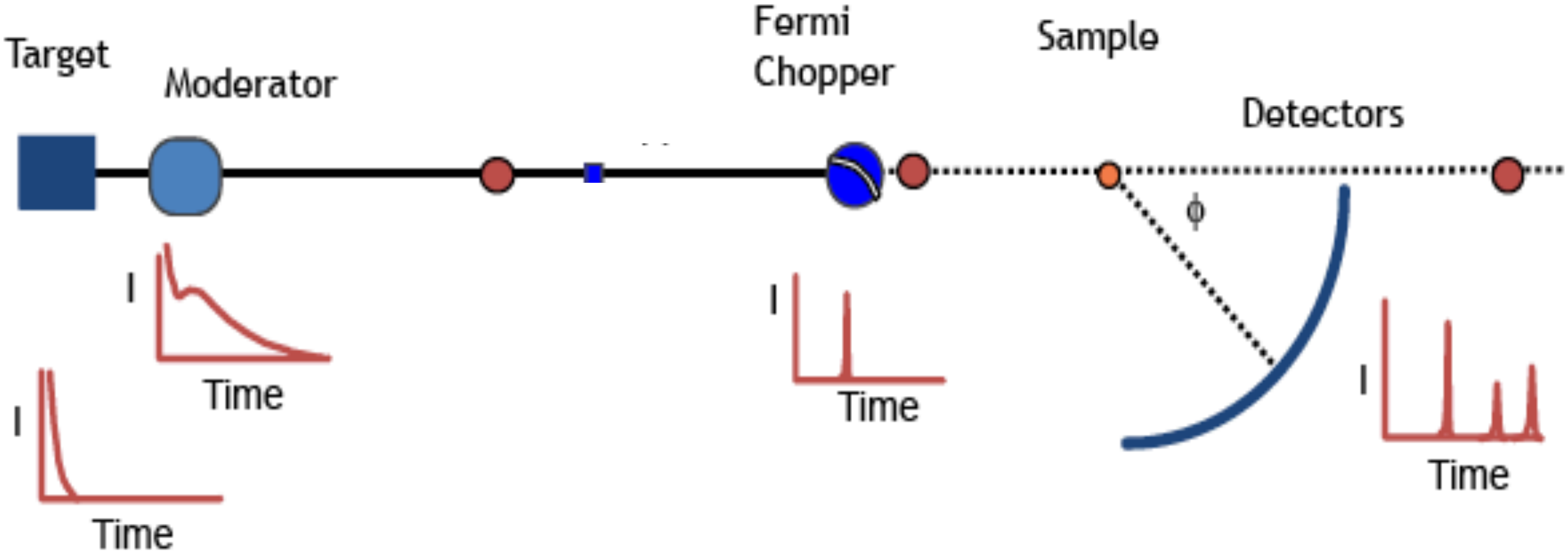
TIME OF FLIGTH (TOF) SPECTROMETER-→ INDIRECT GEOMETRY



SPECTROMETERS OPERATING IN DIRECT GEOMETRY

A chopper spectrometer on a pulsed source

- The source operates at 50Hz.
- The neutron beam is under-moderated to preserve a high flux of epithermal neutrons and a short pulse width.



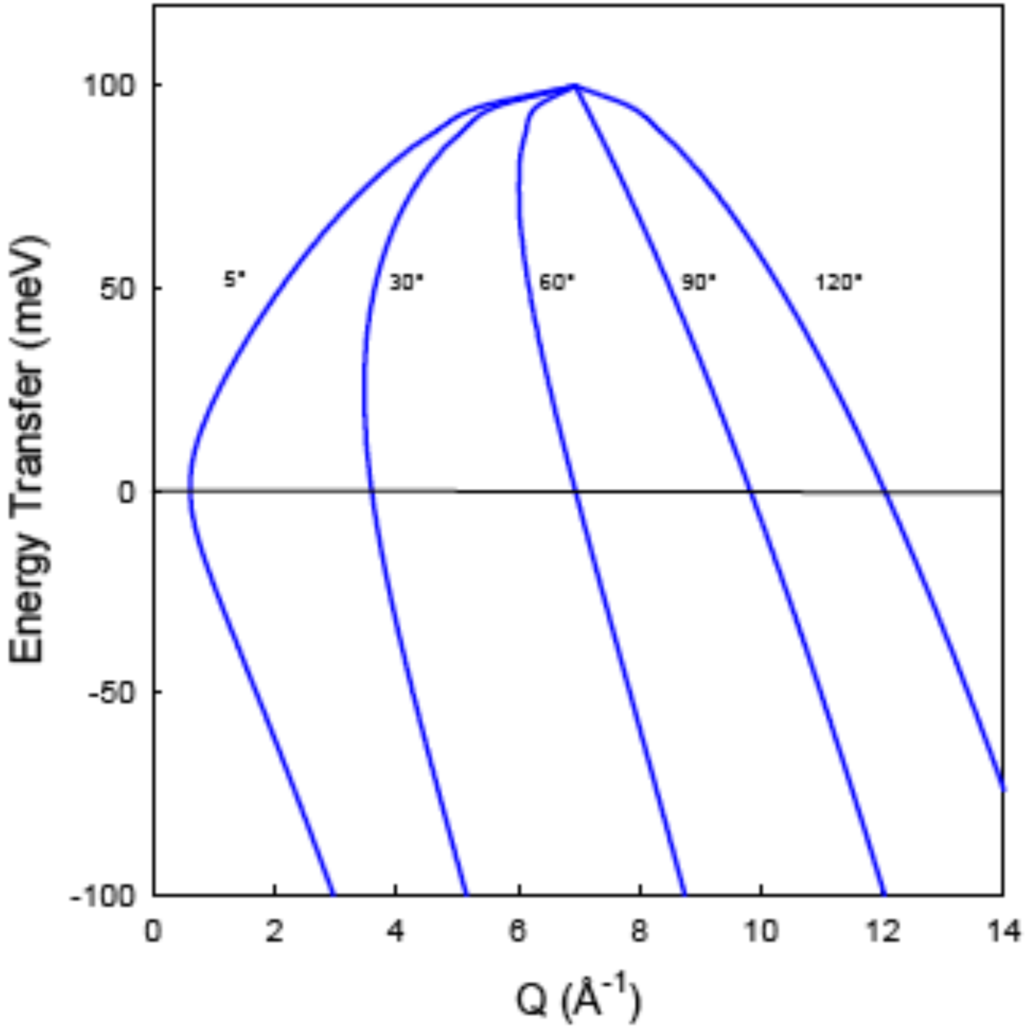
TRAJECTORIES IN (Q, ω) SPACE → DIRECT GEOMETRY

- For direct geometry eliminate E_f.

$$\frac{\hbar^2\mathbf{Q}^2}{2m} = 2E_i - \hbar\omega - 2\cos\phi\left[E_i\left(E_i - \hbar\omega\right)\right]^{1/2}$$

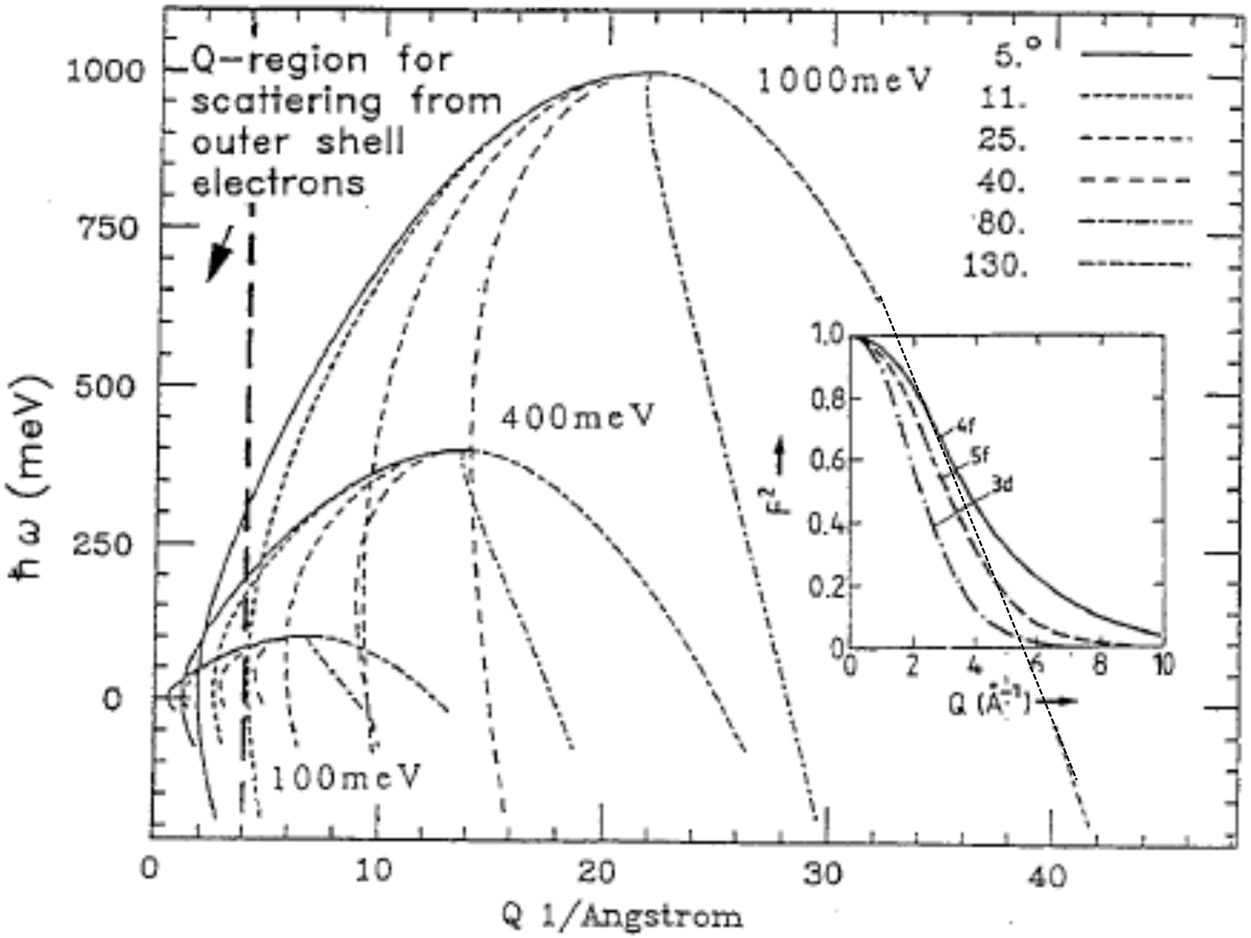
- Each detector has a parabolic trajectory through (Q,ω) space.

- To make optimum use of direct geometry spectrometers they feature large detector arrays, giving simultaneous access to a large area of (Q,ω) space.



KINEMATICS RANGE → DIRECT GEOMETRY

Kinematic range for different incident energies (from 100 to 1000 meV) and an angular detector range from 5° to 130 °.



The size of the kinematic surface grows as the incident energy increases.

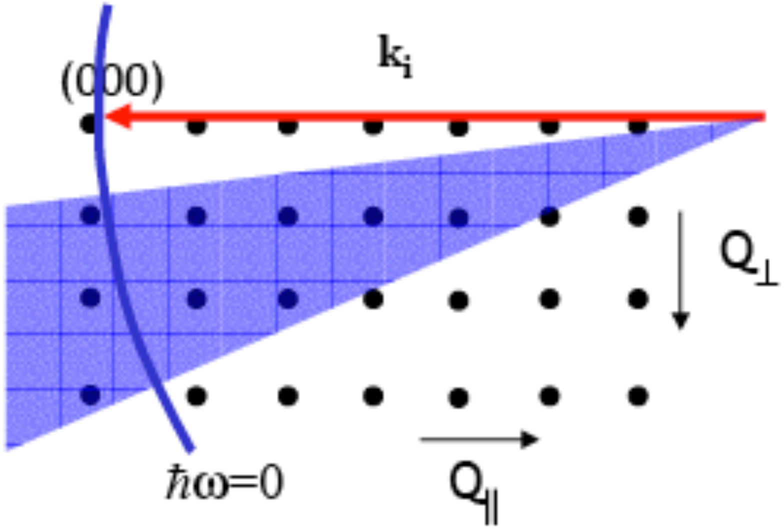
The insert shows the form factor of the outer shell electrons responsible for magnetic scattering

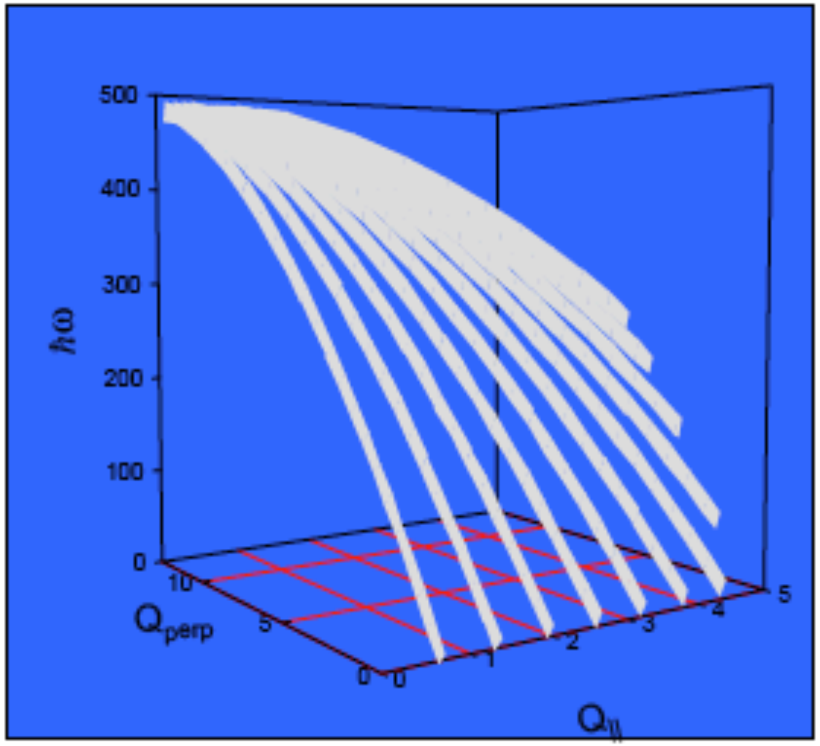
It is very difficult to measure high energy transfers at very small Q!

G. S. Bauer 2005

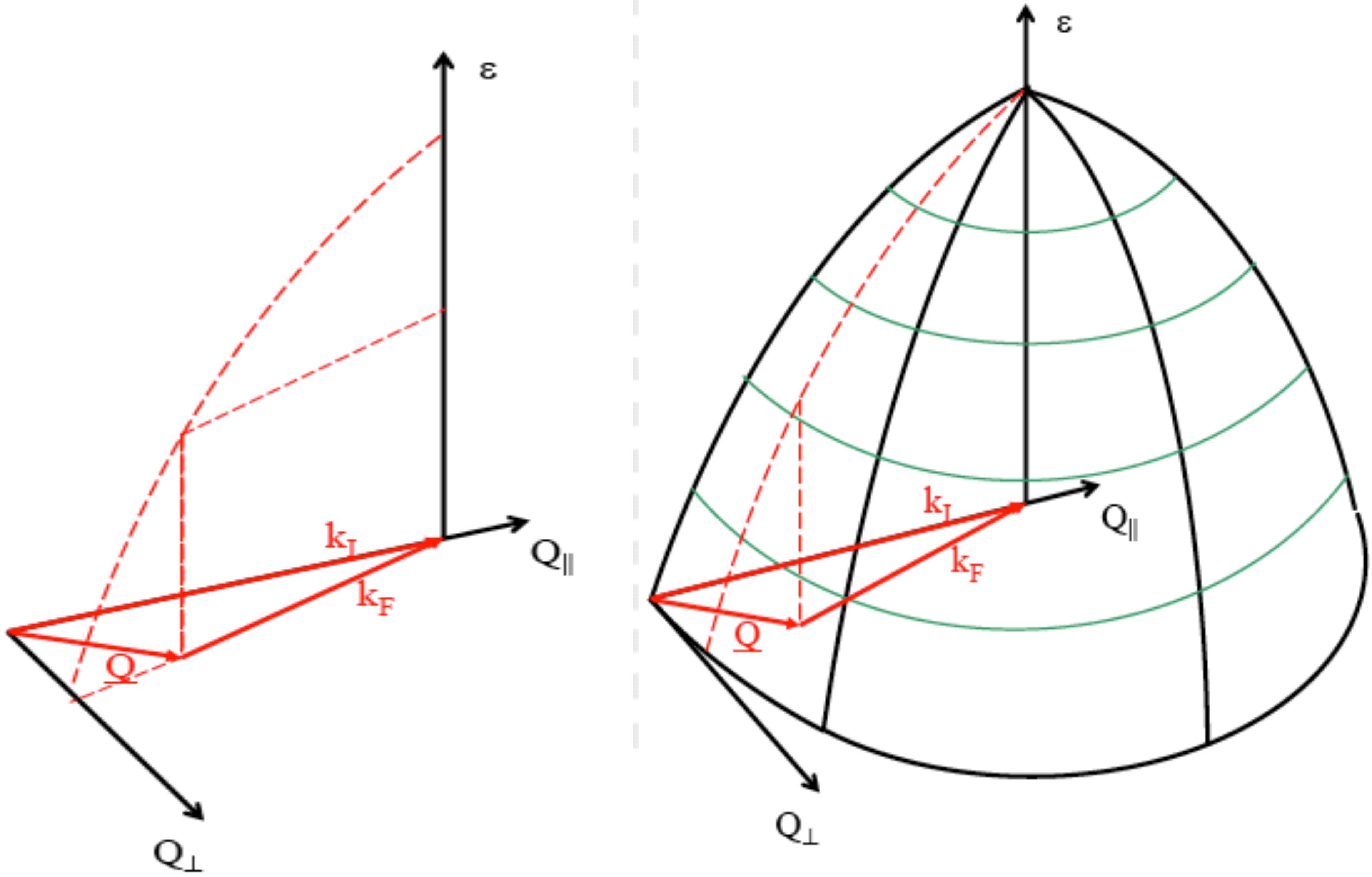
Single Crystal Experiments on a Chopper Spectrometer

- From the scattering triangle we can see that an array of detectors will trace out a sector in reciprocal space



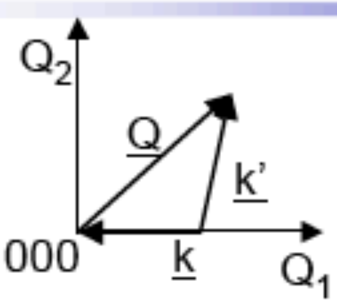


- Each detector has a parabolic trajectory through (Q,ω) space.
- The detector array produces a surface in $(Q_{||},Q_\perp,\omega)$ space



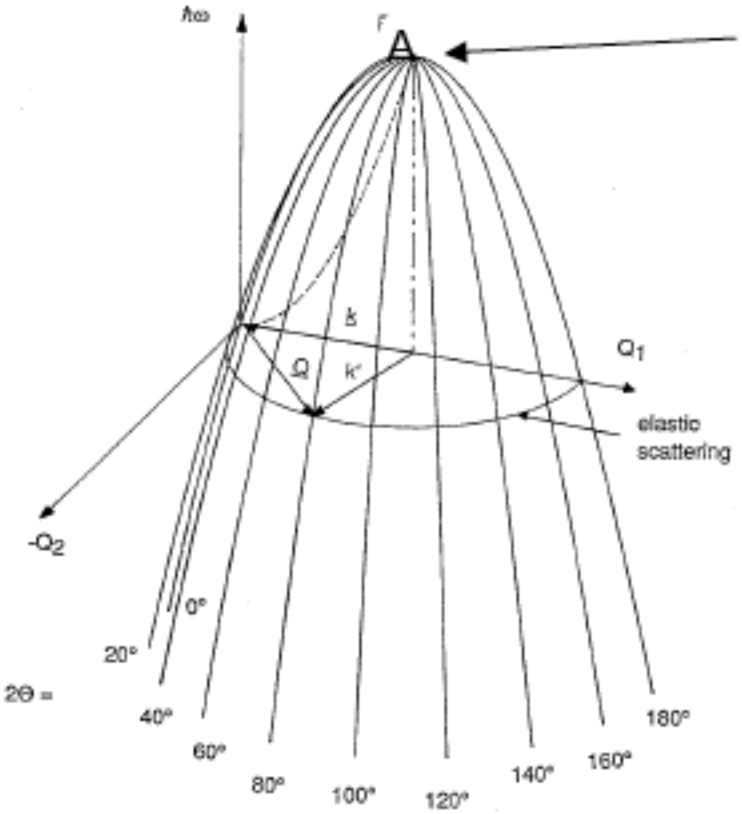
ε
k_I
k_F
Q
$Q_{\parallel}$
$Q_{\perp}$
ε
k_I
k_F
Q
$Q_{\parallel}$
$Q_{\perp}$

KINEMATICS RANGE → DIRECT GEOMETRY



$$Q_1 = k - k' \cdot \cos(2\Theta)$$

$$Q_2 = k' \cdot \sin(2\Theta) \qquad k' = \sqrt{k^2 - \frac{2m}{\hbar^2} \cdot \hbar\omega}$$



G. S. Bauer 2005

Energy of the incident neutron:
$E = \hbar\omega = \hbar^2 k^2/2m \Rightarrow$ Parabola in the ħω,k plane

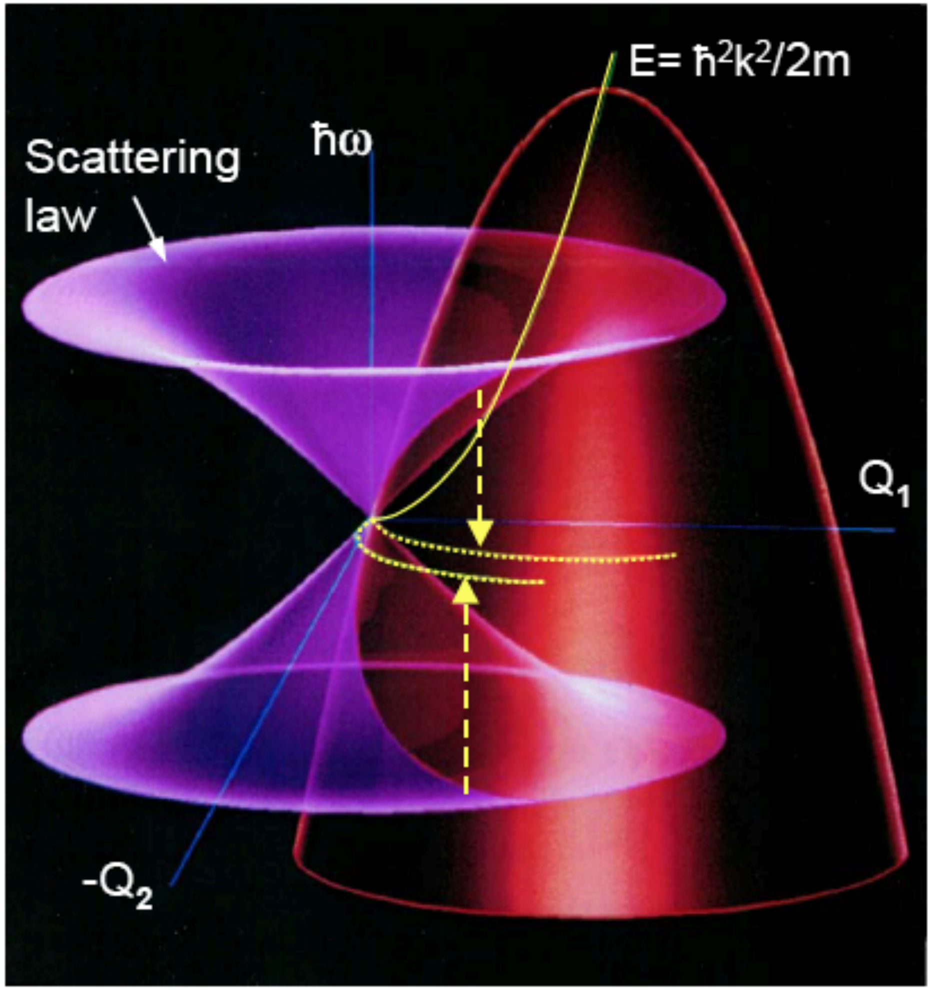
Similarly, the loci for all scattered neutrons are parabolae in the ħω,k' planes with apex point A.

The paraboloid spanned by all scattering angles is therefore the locus for all possible combinations of Q and ħω that can be measured with neutrons of incident wave vector k (kinematic scattering surface).

Elastic scattering occurs for $\hbar\omega = 0$; $\hbar\omega > 0$ means neutron energy loss, $\hbar\omega < 0$ means neutron energy gain.

KINEMATICS RANGE → DIRECT GEOMETRY

Back to 3 dimensions



G. S. Bauer 2005

In order for a neutron to be scattered its kinematic surface must intersect with the scattering law of the sample.

With a triple axis spectrometer, which does point wise scans, it is possible to follow the scattering law along symmetry directions in the reciprocal crystal lattice.

In a multidetector time of flight scan with fixed incident neutron energy the loci for the Q-vectors measured are curved. The scattering law along symmetry directions must be constructed from many scans at different orientations of the sample.

ENERGY RESOLUTION OF A CHOPPER SPECTROMETER

Energy Resolution

Moderator term

$$\Delta t_{md} = \frac{L_2 + L_3}{L_1} \Delta t_{mm}$$

Chopper term

$$\Delta t_{cd} = \frac{L_2 + L_1 + L_3}{L_1} \Delta t_{cc}$$

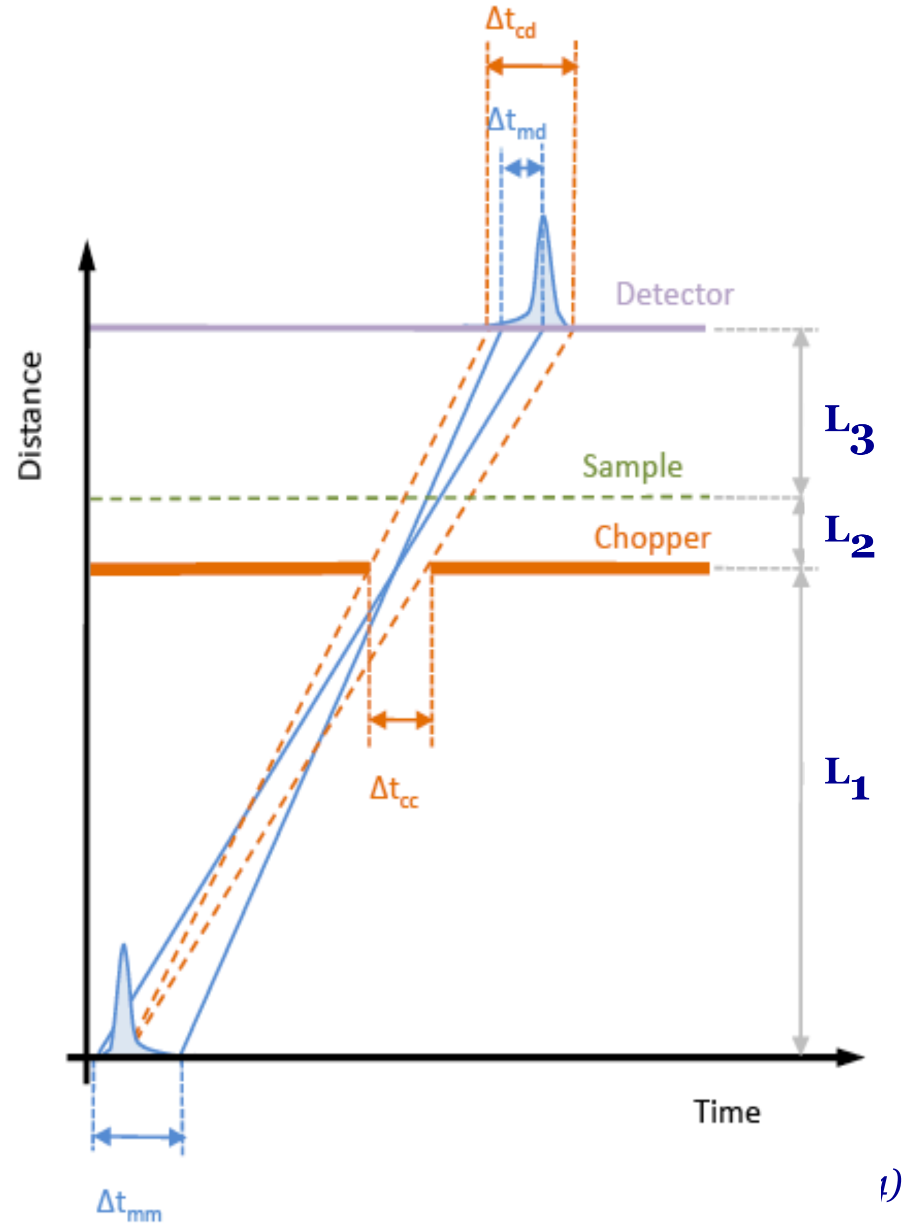
These are added in quadrature and converted to energy

$$\frac{\Delta \varepsilon}{E_i} = 8.7478 \times 10^{-4} \frac{\sqrt{E_i(meV)}}{L_2(m)} \Delta t(\mu s)$$

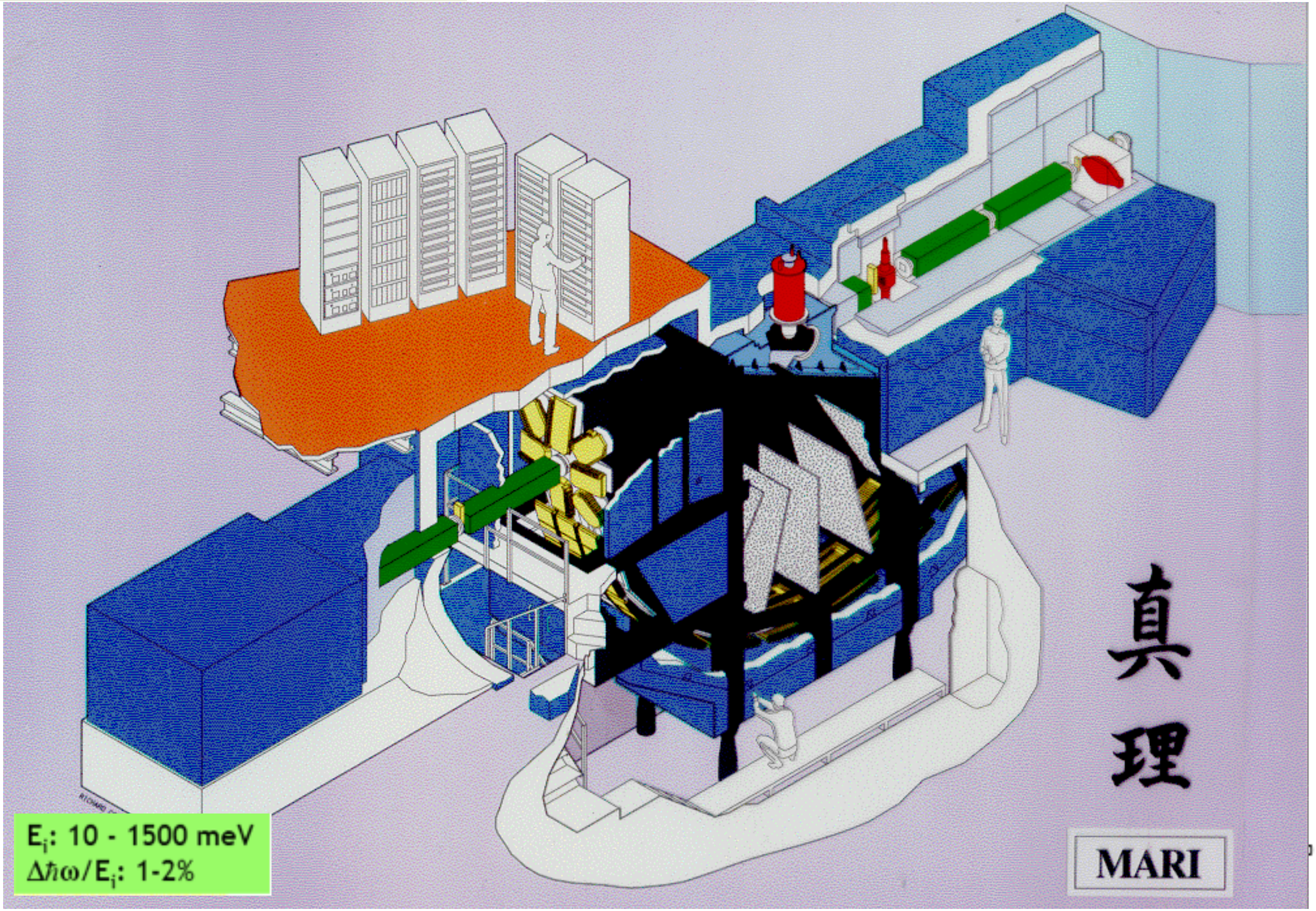
Resolution increases as L_2 lengthens

Flux

$$\Phi = \left(\frac{p}{d}\right)\left(\frac{WmHm}{(L_1 + L_3)^2}\right)\left(\frac{\Delta t_{cc}}{L_1}\right)$$

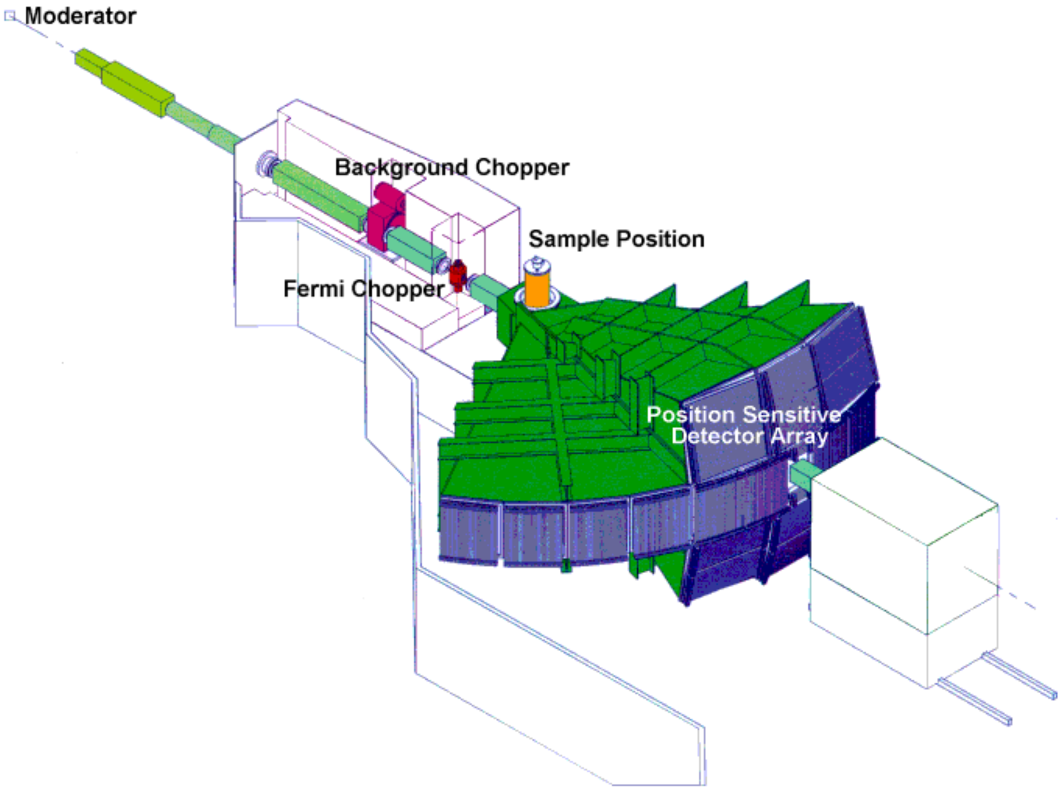


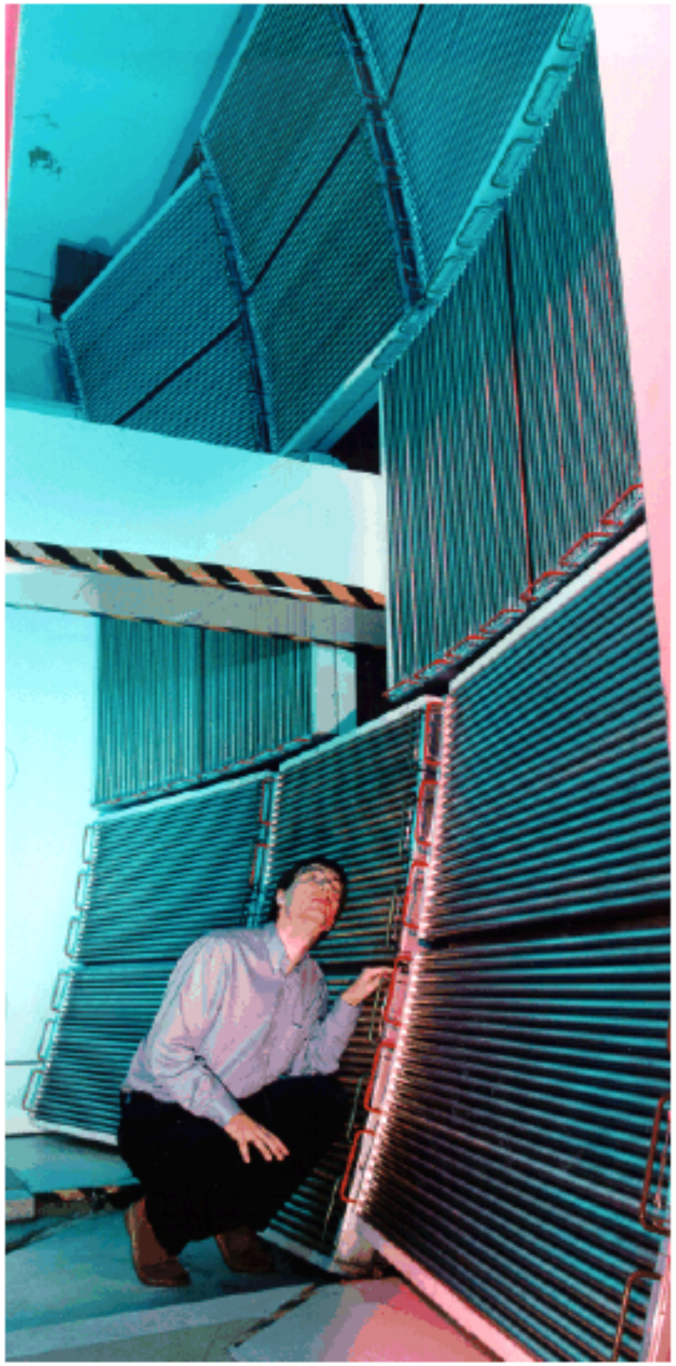
ENERGY RESOLUTION OF A CHOPPER SPECTROMETER



MAPS

- 16 m^2 of PSDs
- 600 1m detectors
- 70 000 pixels





2014)

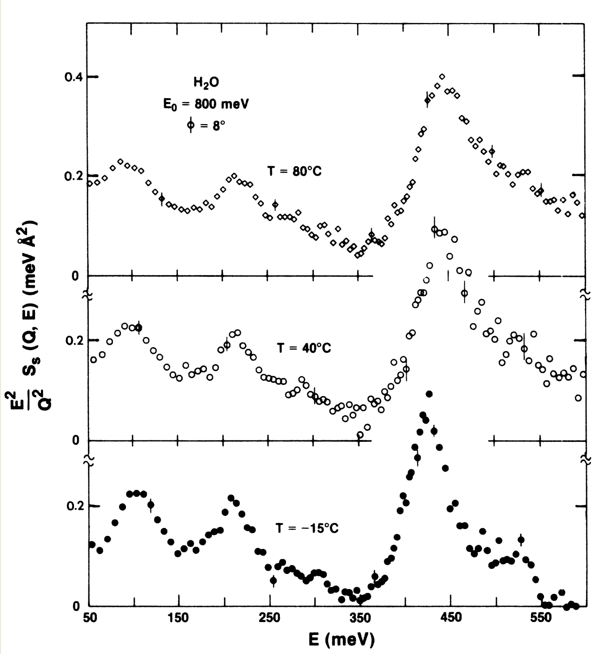
EXPERIMENTS ON DIRECT GEOMETRY SPECTROMETER

- Direct geometry spectrometers offer simultaneous coverage of a wide area of (Q,ω) space with a wide range in incident energies (IN6~ 2.3 meV, HET~ 2eV)

  - **Vibrational motions in molecular systems**
  - Magnetic excitations in polycrystalline samples
  - Amorphous materials
  - Quantum fluids
  - Excitations in single crystal
  - Momentum distributions

Inelastic Neutron Scattering: a probe for H vibrations

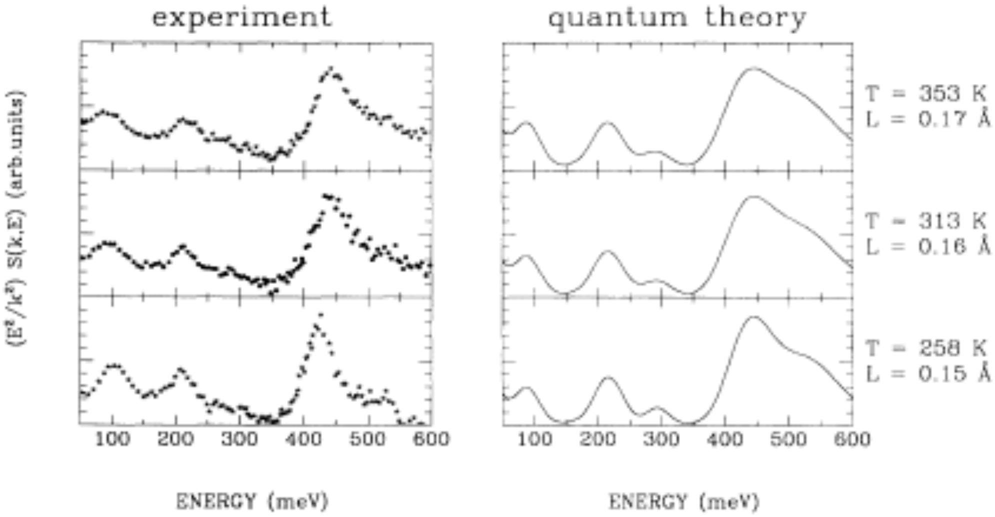
Inelastic Incoherent Neutron Scattering measurements of water at IPNS



Toukan et al., PRB (1988)

Incoherent inelastic neutron scattering from liquid water: A theoretical investigation

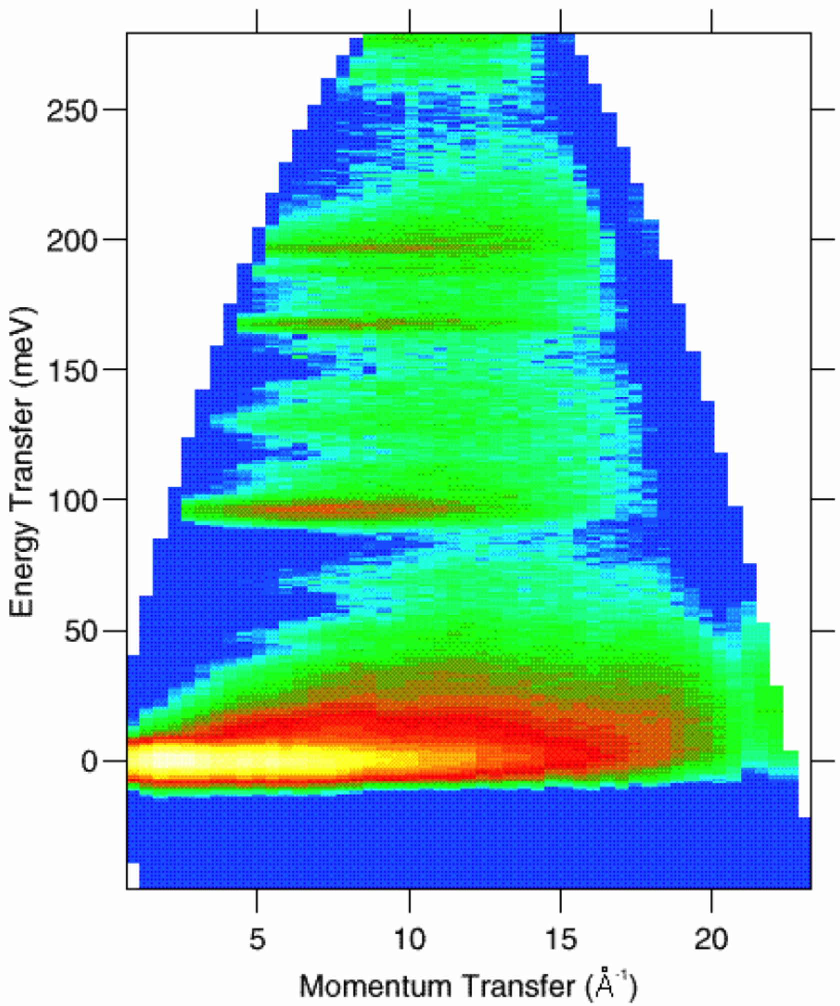
S. Bratos, M. Diraison, G. Tarjus, and J-Cl. Leicknam



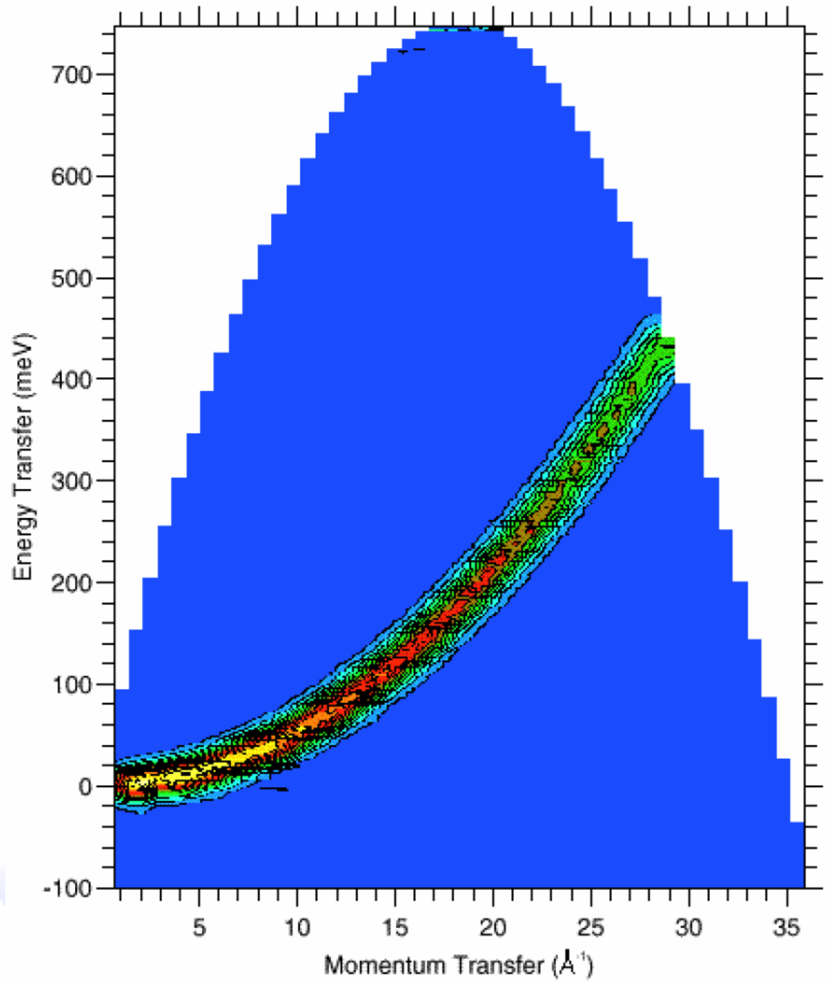
Bratos et al., PRA (1992)

@ HET → EXAMPLE OF EXPERIMENT

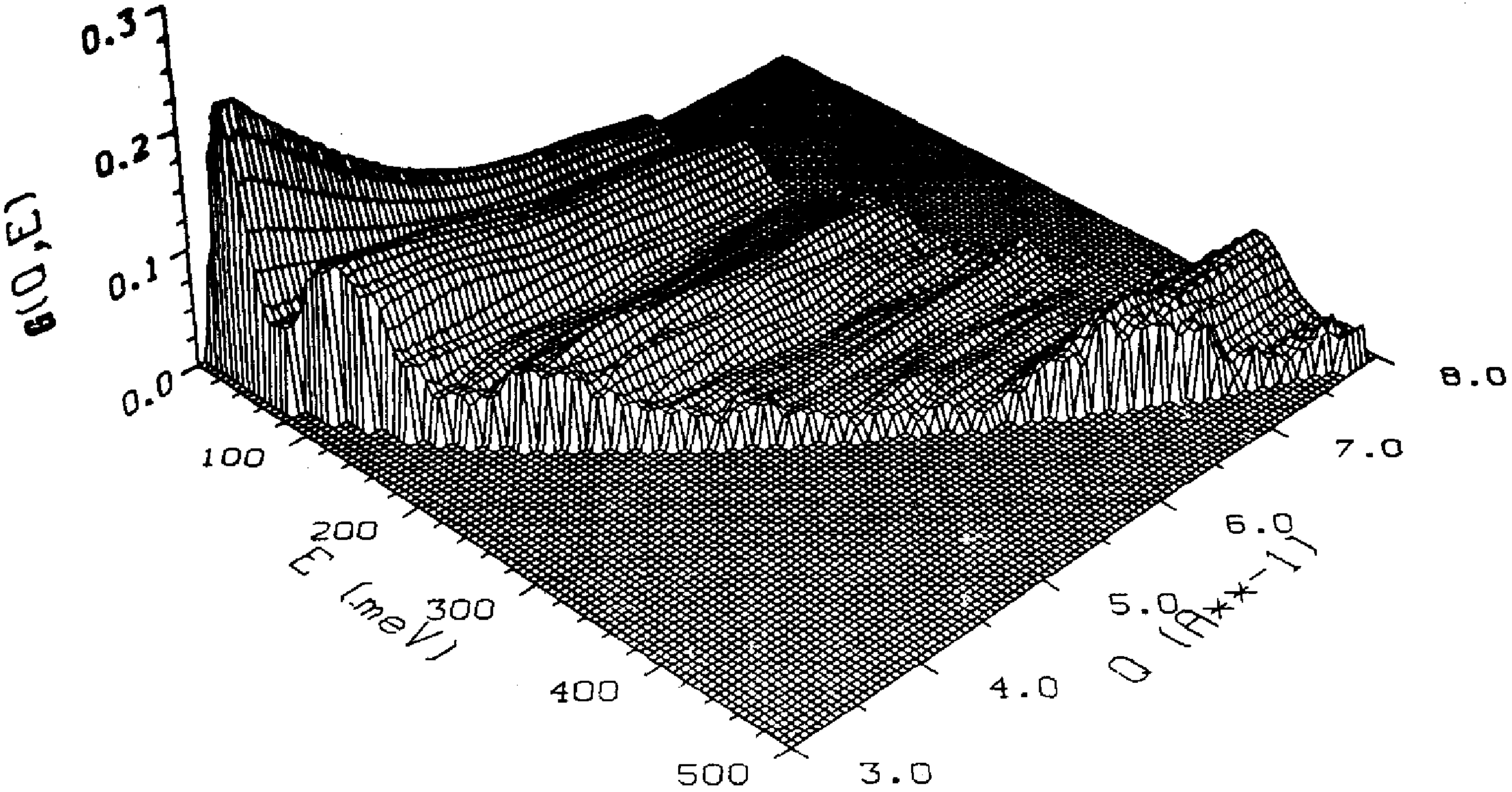
n-methyl acetamide (Fillaux)



^{4}He (Stirling et al)

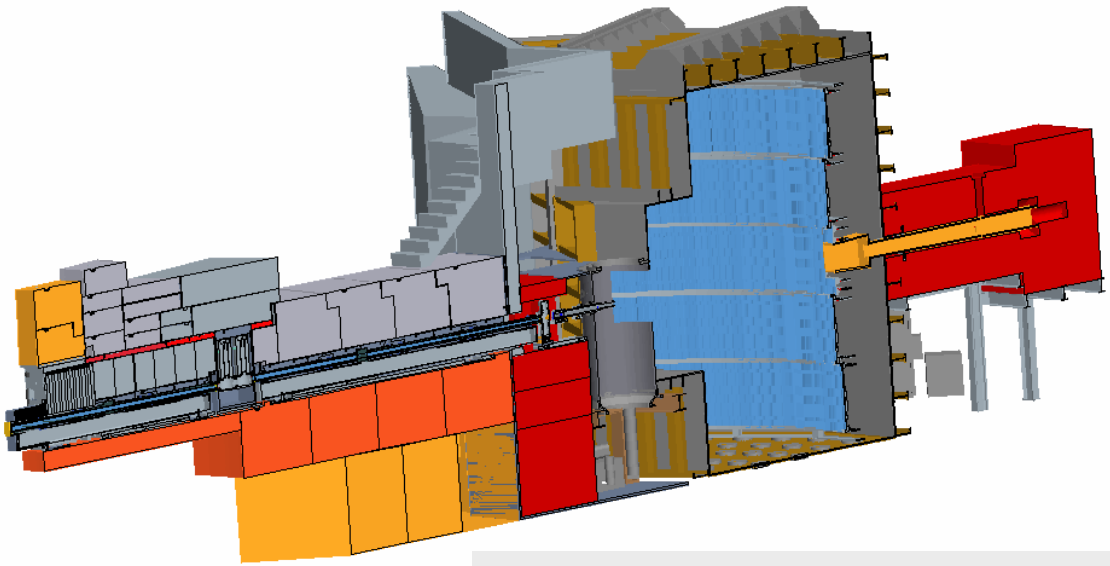


$S(q,\omega)$ for ice Ih, @ IPNS



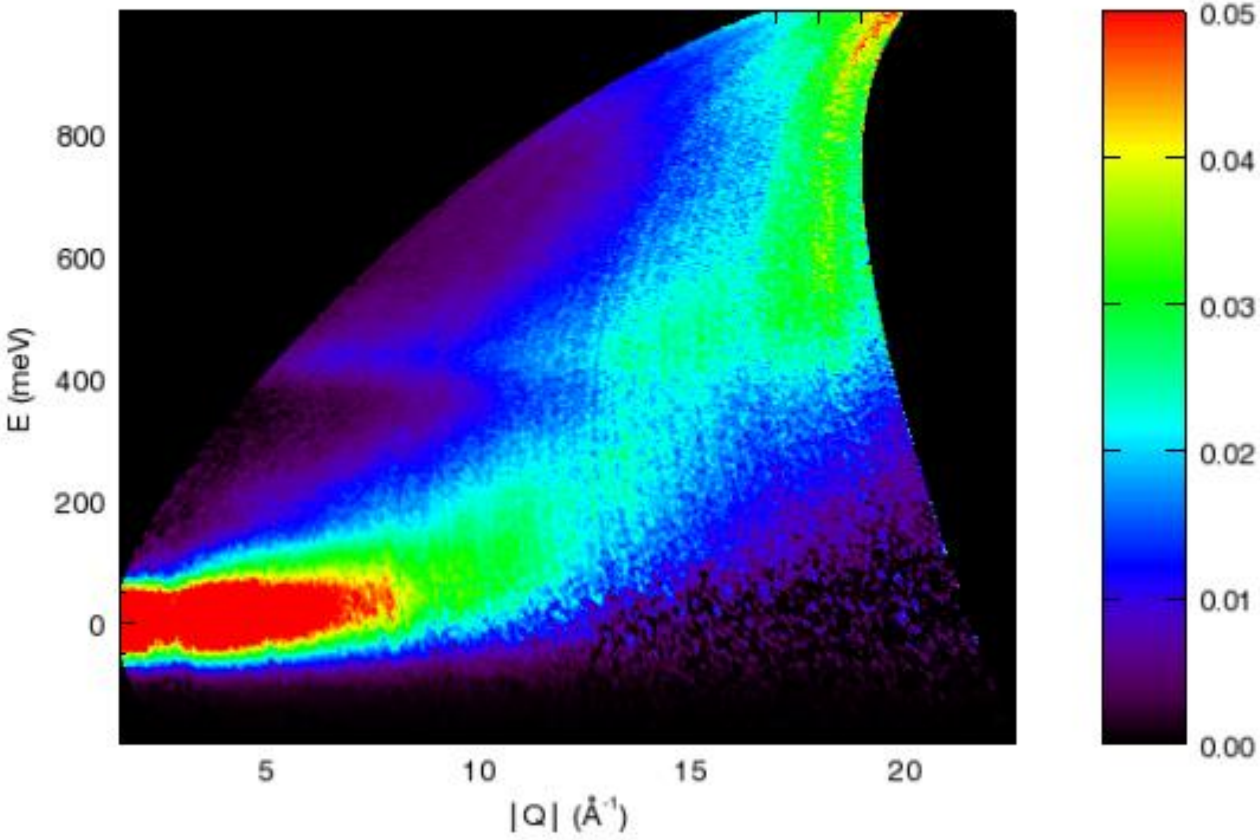
SEQUOIA @ SNS

http://neutrons.ornl.gov/sequoia/

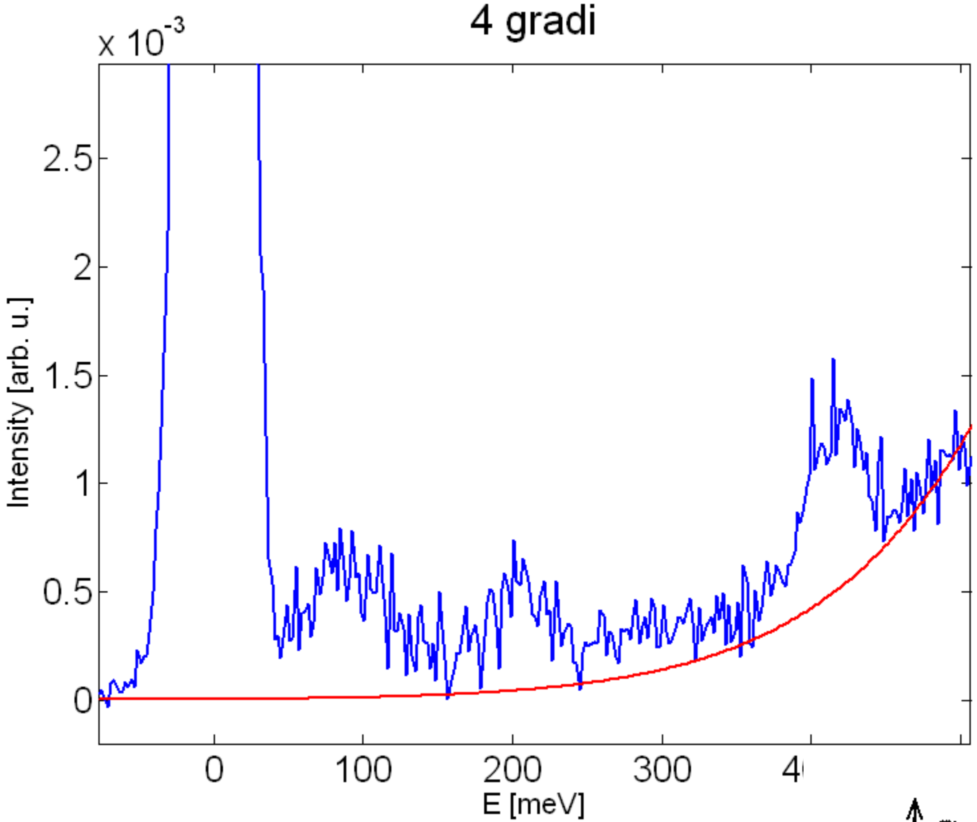


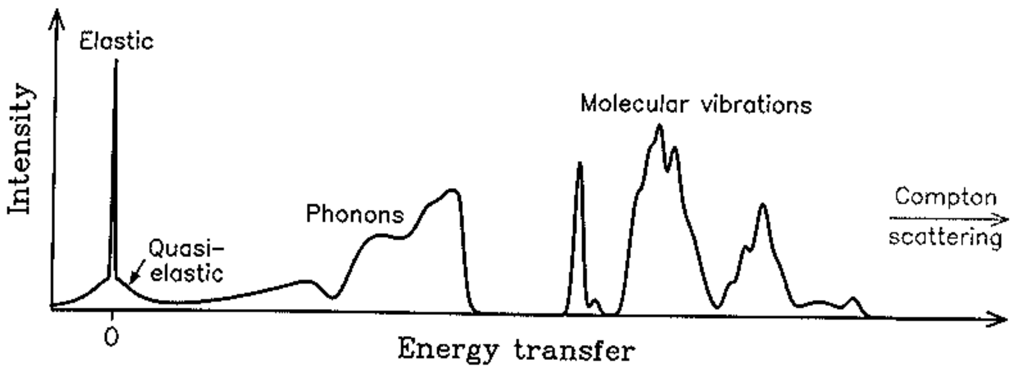
Incident energy	
Range	10–2000 meV
Resolution (elastic)	1–5% Ei
Vertical	
Detector coverage	-18–18°
Horizontal	
Detector coverage	-30–60°
Minimum detector Angle	3°

- Put a sample in the beam (ice and/or water)
- Record intensity maps as a function of energy-momentum
- Transfers using data reduction (softwares)

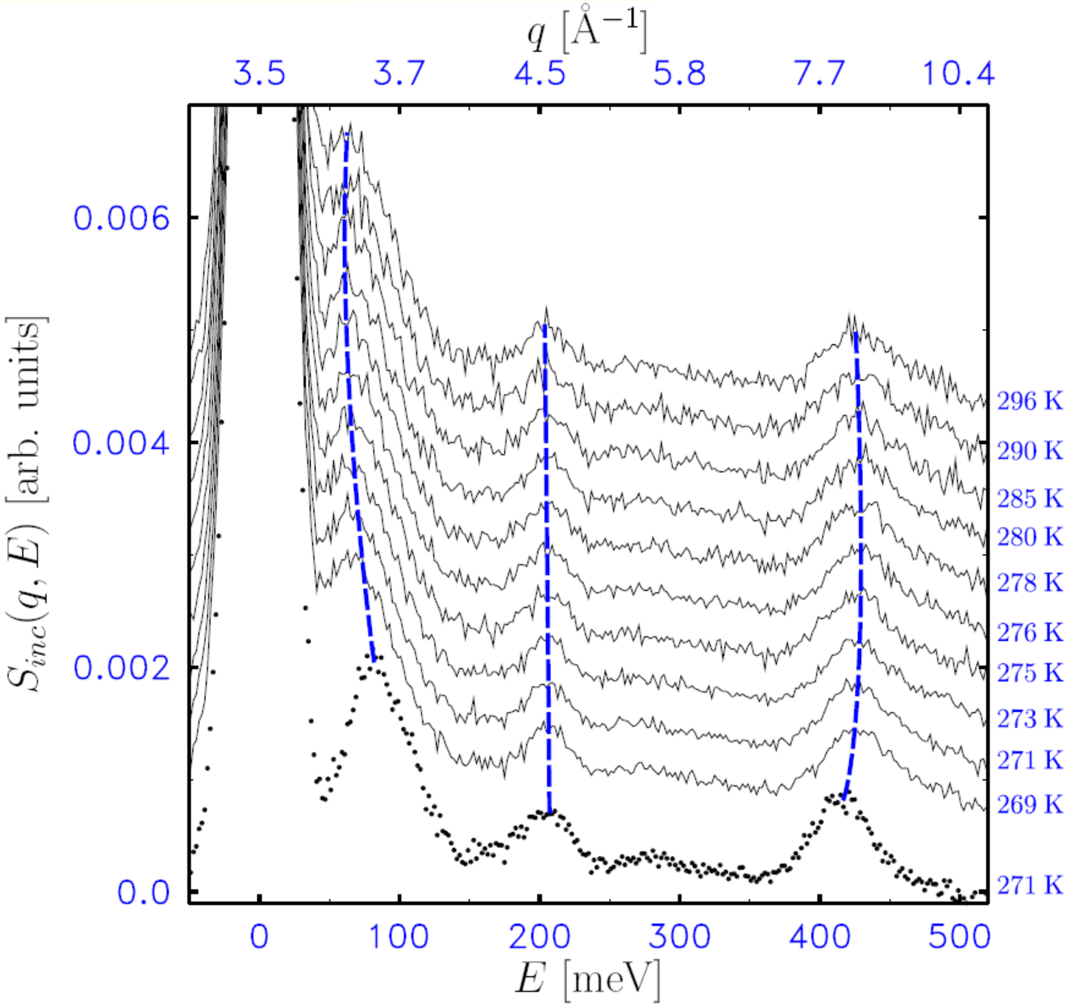


- data correction: multiple scattering, multiphonon, etc, to obtain S(Q,ω) [from S(Θ,ω)]





INS and *ab initio* electronic structure calculations on Results from Inelastic neutron scattering on water across the triple point



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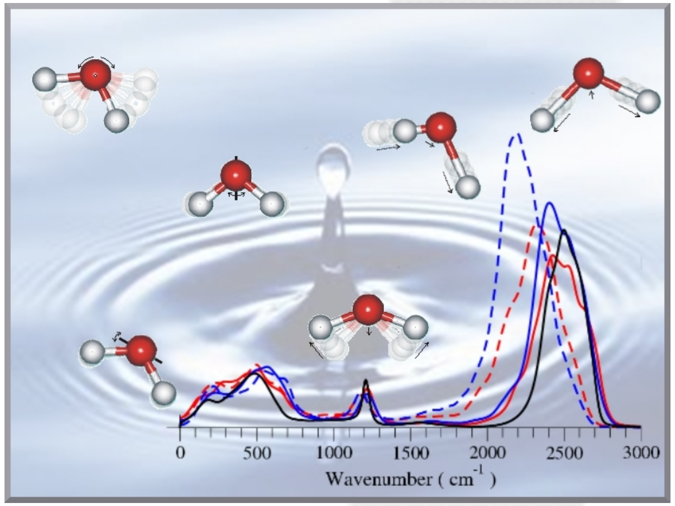


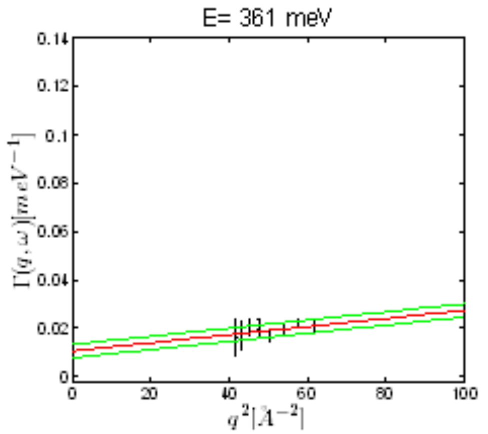
http://angstrom.ucdavis.edu/

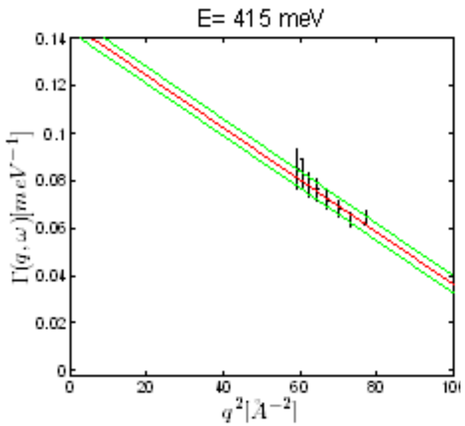
R. Senesi, *et al* J. Chem Phys. 2013

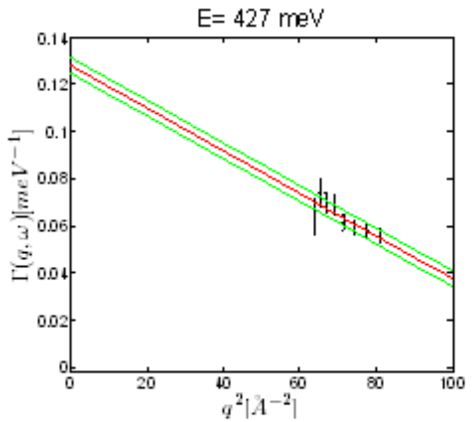
Make a constant ω cut towards Q=0!
i.e. for each ω within the range of the stretching band take the zero-Q limit by extrapolation.

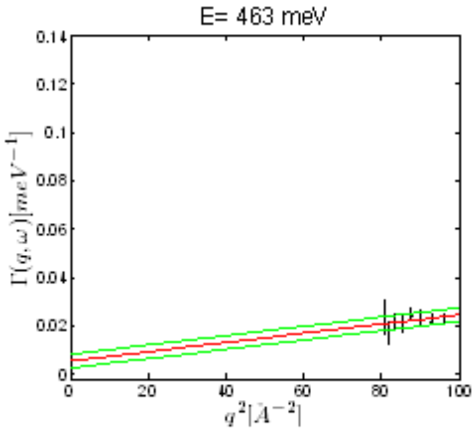
$$\lim_{Q\to 0}\left[\frac{S_{inc}(Q,\omega)}{Q^2}\,\frac{2m\,\hbar\omega}{e^{-2W}}\,\frac{4\pi}{\sigma_{inc}}\,\frac{1}{n(\omega)+1}\right] = \lim_{Q\to 0}\Gamma(Q,\omega)$$

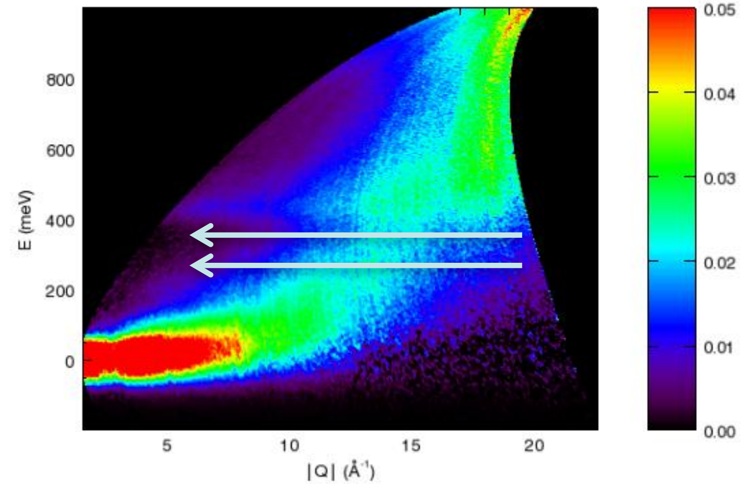
$$= g(\omega)\ or\ g(E)$$



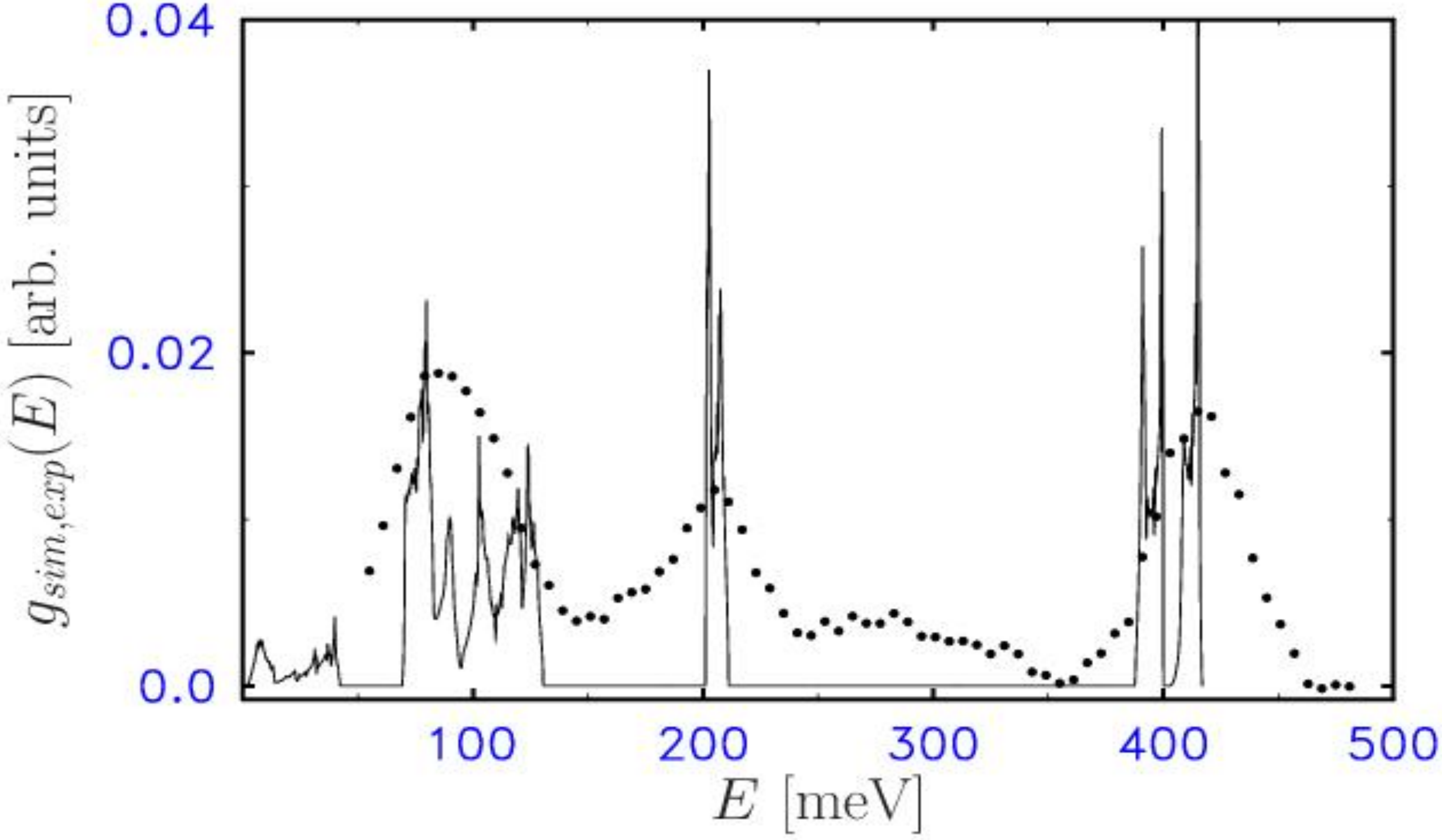




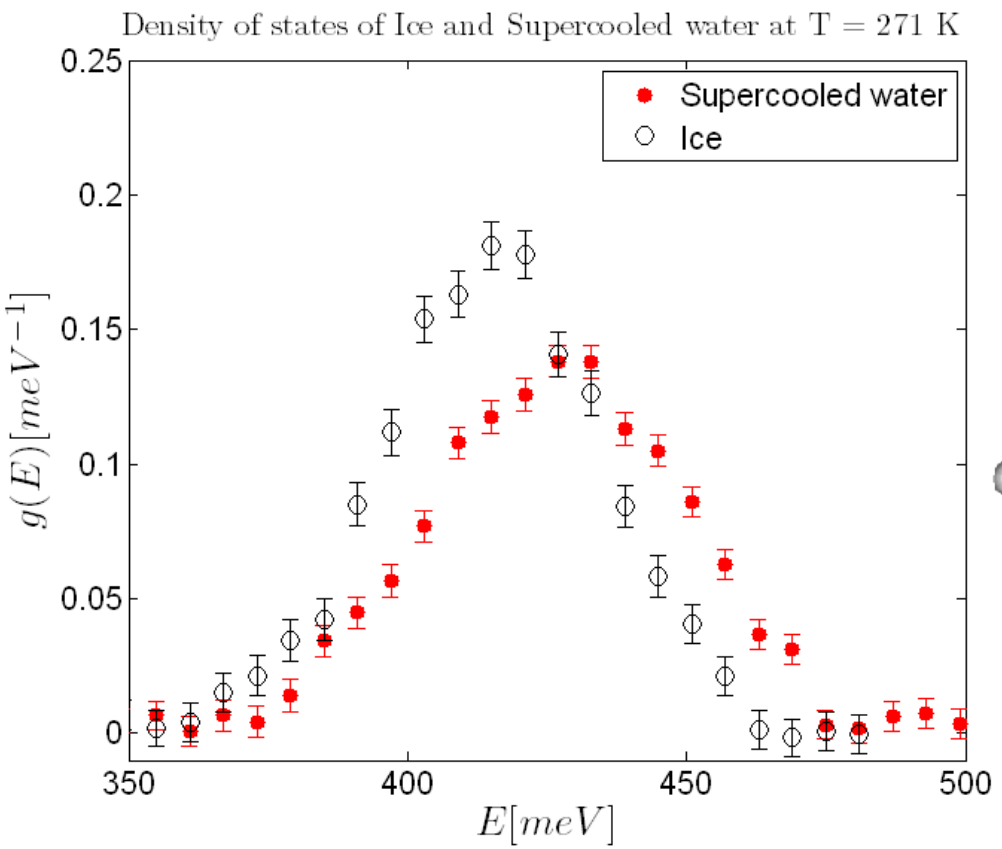


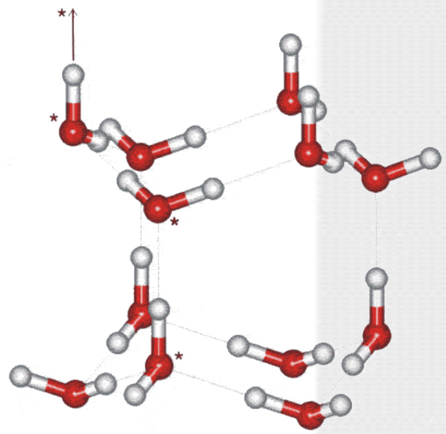


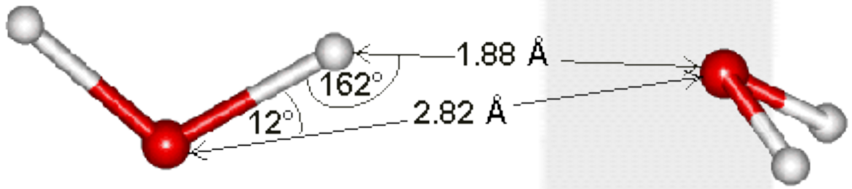
g(E): from *ab initio* electronic structure calculations and from INS



R. Senesi, et al, *J. Chem. Phys.* **139**, 074504 (2013)







Red-shifted in ice due greater association of the proton with the oxygen accepting the hydrogen bond, thereby resulting in the weakening of the covalent bond…

4)


THANK YOU
QUESTIONS?