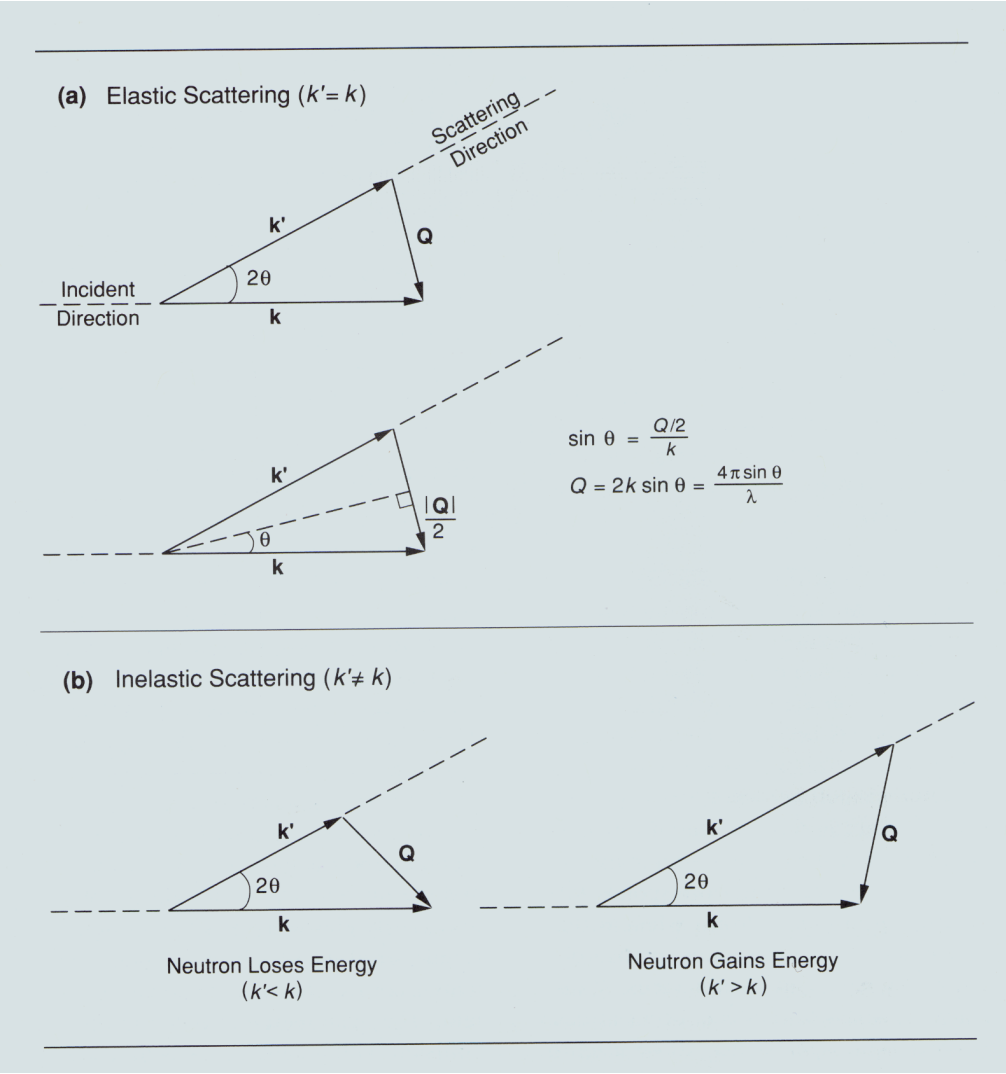
Three Axis Spectrometers

Roger Pynn

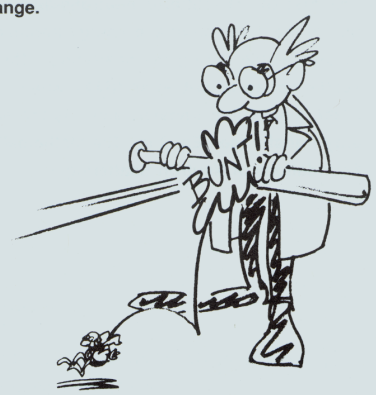
Indiana University & Oak Ridge National Laboratory

The Neutron Changes Both Energy & Momentum When Inelastically Scattered by Moving Nuclei



inelastic scattering

Scattering in which exchange of energy and momentum between the incident neutron and the sample causes both the direction and the magnitude of the neutron's wave vector to change.



PHYSICAL REVIEW VOLUME 128, NUMBER 3 NOVEMBER 1, 1962

Crystal Dynamics of Lead. I. Dispersion Curves at 100°K

B. N. Brockhouse,* T. Arase,† G. Caglioti,‡ K. R. Rao,§ and A. D. B. Woods
Neutron Physics Branch, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada
(Received June 4, 1962)

Brockhouse's first 3 axis spectrometer at NRU reactor in 1959

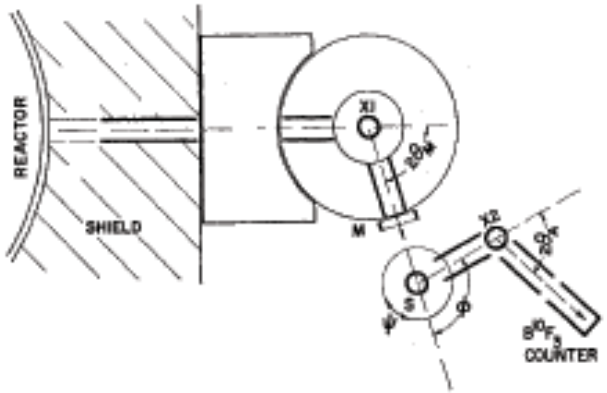
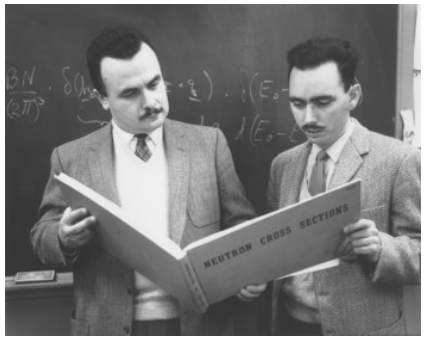
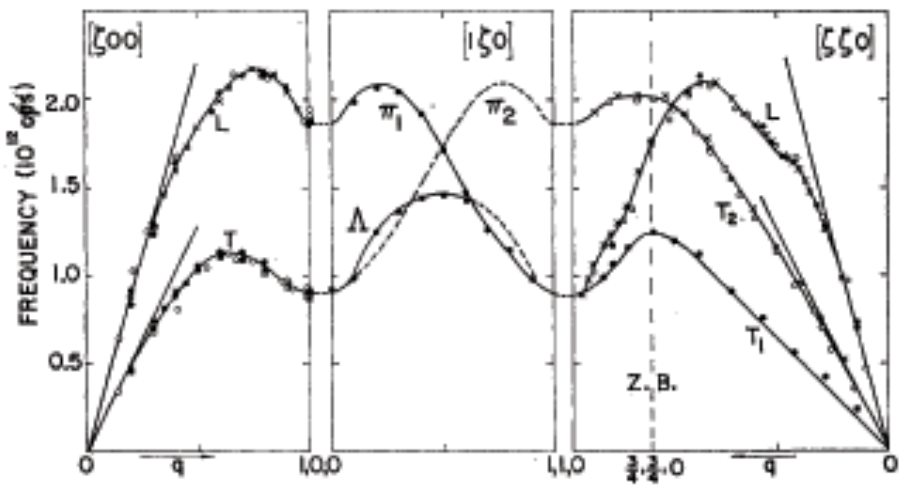
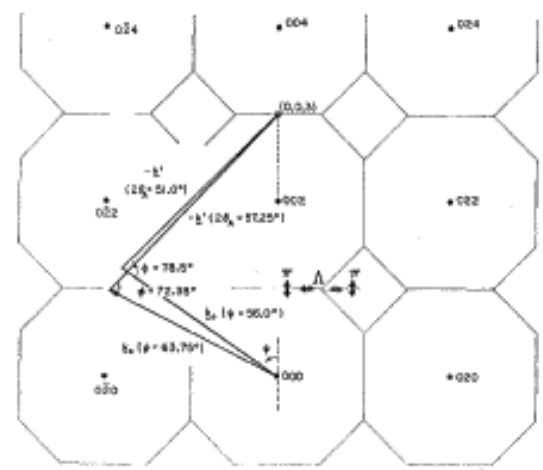


Fig. 1. Schematic drawing of the apparatus.



Brockhouse and Woods

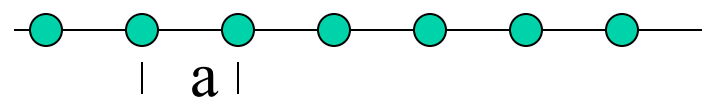




The constant-Q method

Diffraction from a Frozen Wave

- Recall that $S(\vec{Q}) = \frac{1}{N}\left|\sum_k e^{i\vec{Q}.\vec{r}_k}\right|^2$

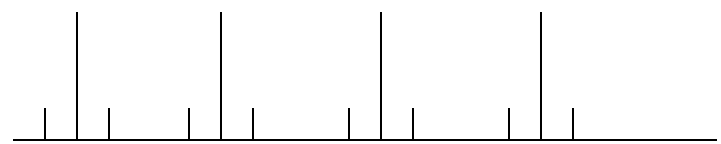


- We know that for a linear chain of "atoms" along the x axis $S(Q_x)$ is just a series of delta function reciprocal lattice planes at $Q_x = n2\pi/a$, where a is the separation of atoms

What happens if we put a "frozen" wave in the chain of atoms so that the atomic positions are $x_p = pa + u\cos kpa$ where p is an integer and u is small?

$$S(Q) = \left|\sum_p e^{iQpa} e^{iQu\cos kpa}\right|^2 \approx \left|\sum_p e^{iQpa}(1 + iQu[e^{ikpa} + e^{-ikpa}])\right|^2$$

$$\approx \left|\sum_p e^{iQpa} + iQu[e^{i(Q+k)pa} + e^{i(Q-k)pa}]\right|^2$$



so that in addition to the Bragg peaks we get weak satellites at $Q = G \pm k$

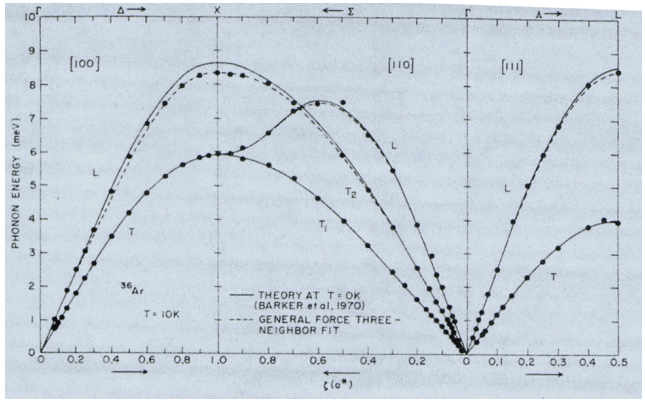
What Happens if the Wave Moves?

- If the wave moves through the chain, the scattering still occurs at wavevectors G + k and G – k but now the scattering is inelastic

- For quantized lattice vibrations, called phonons, the energy change of the neutron is $\hbar\omega$ where ω is the vibration frequency.

- In a crystal, the vibration frequency at a given value of $\vec{k}$ (called the phonon wavevector) is determined by interatomic forces. These frequencies map out the so-called phonon dispersion curves.

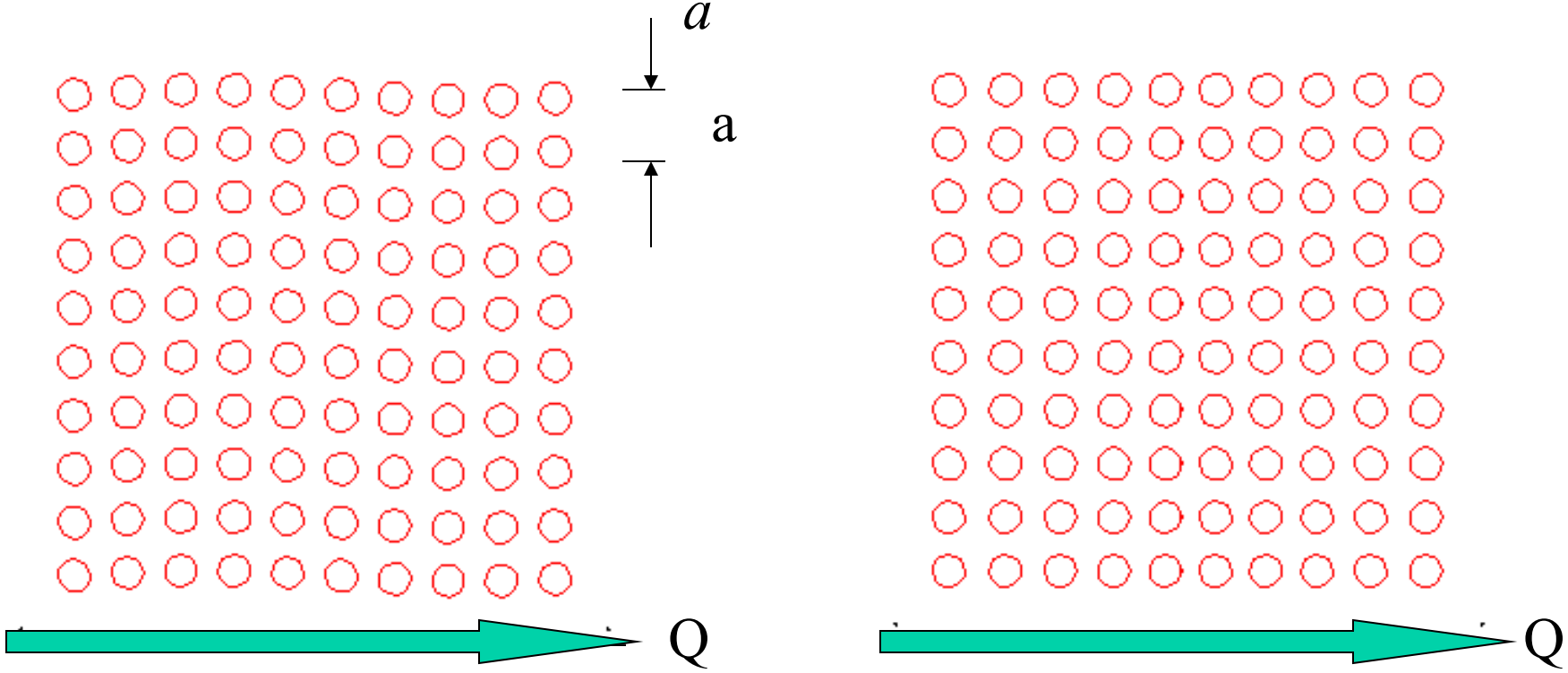
- Different branches of the dispersion curves correspond to different types of motion



phonon dispersion in ^{36}Ar

Atomic Motions for Longitudinal & Transverse Phonons

$$\vec{Q} = \frac{2\pi}{a}(0.1,0,0)$$



Transverse phonon

$$\vec{e}_T = (0,0.1,0)a$$

Longitudinal phonon

$$\vec{e}_L = (0.1,0,0)a$$

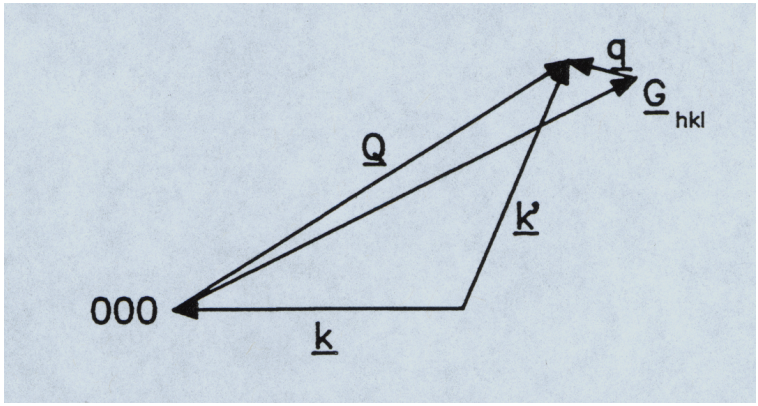
$$\vec{R}_l = \vec{R}_{l0} + \vec{e}_s e^{i(\vec{Q}.\vec{R}_l - \omega t)}$$

Phonons – the Classical Use for Inelastic Neutron Scattering

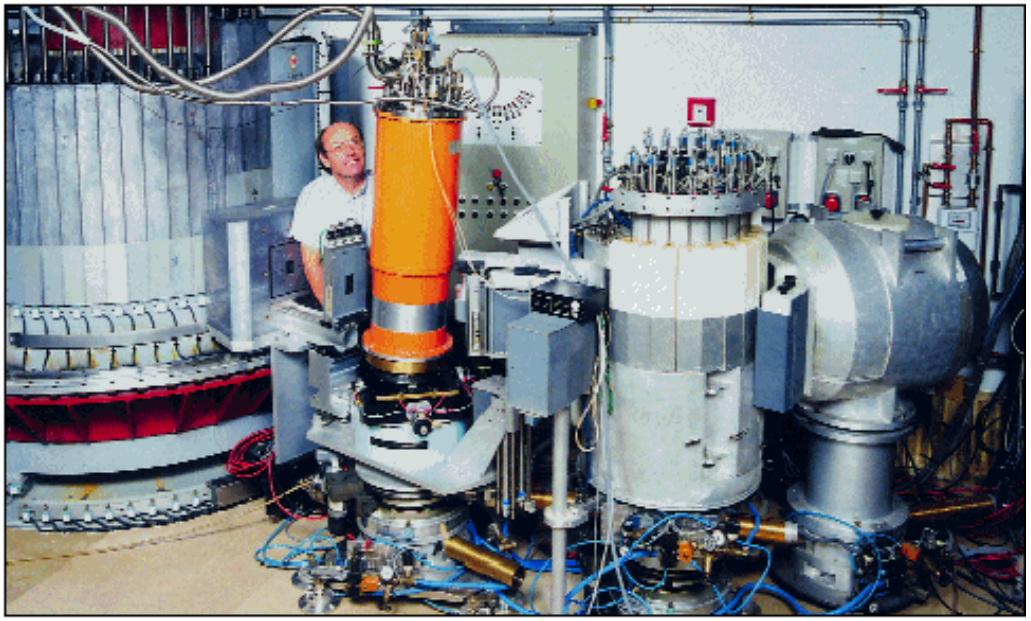
- Coherent scattering measures scattering from single phonons

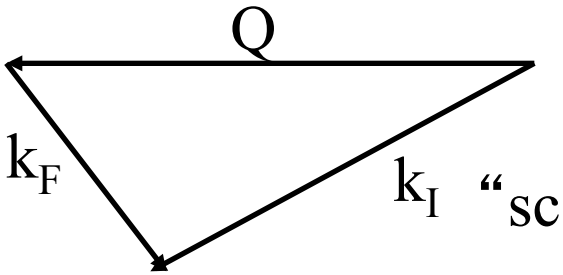
$$\left(\frac{d^2\sigma}{d\Omega dE}\right)_{coh\pm1} = \sigma_{coh}\frac{k'}{k}\frac{\pi^2}{MV_0}e^{-2W}\sum_s\sum_G\frac{(\vec{Q}.\vec{e}_s)^2}{\omega_s}(n_s+\frac{1\pm1}{2})\delta(\omega\mp\omega_s)\delta(\vec{Q}-\vec{q}-\vec{G})$$

- Note the following features:
  - Energy & momentum delta functions => see single phonons (labeled *s*)
  - Different thermal factors for phonon creation (n_s+1) & annihilation (n_s)
  - Can see phonons in different Brillouin zones (different recip. lattice vectors, ***G***)
  - Cross section depends on relative orientation of ***Q*** & atomic motions ($\boldsymbol{e}_s$)
  - Cross section depends on phonon frequency (ω_s) and atomic mass (M)
  - In general, scattering by multiple excitations is either insignificant or a small correction (the presence of other phonons appears in the Debye-Waller factor, W)



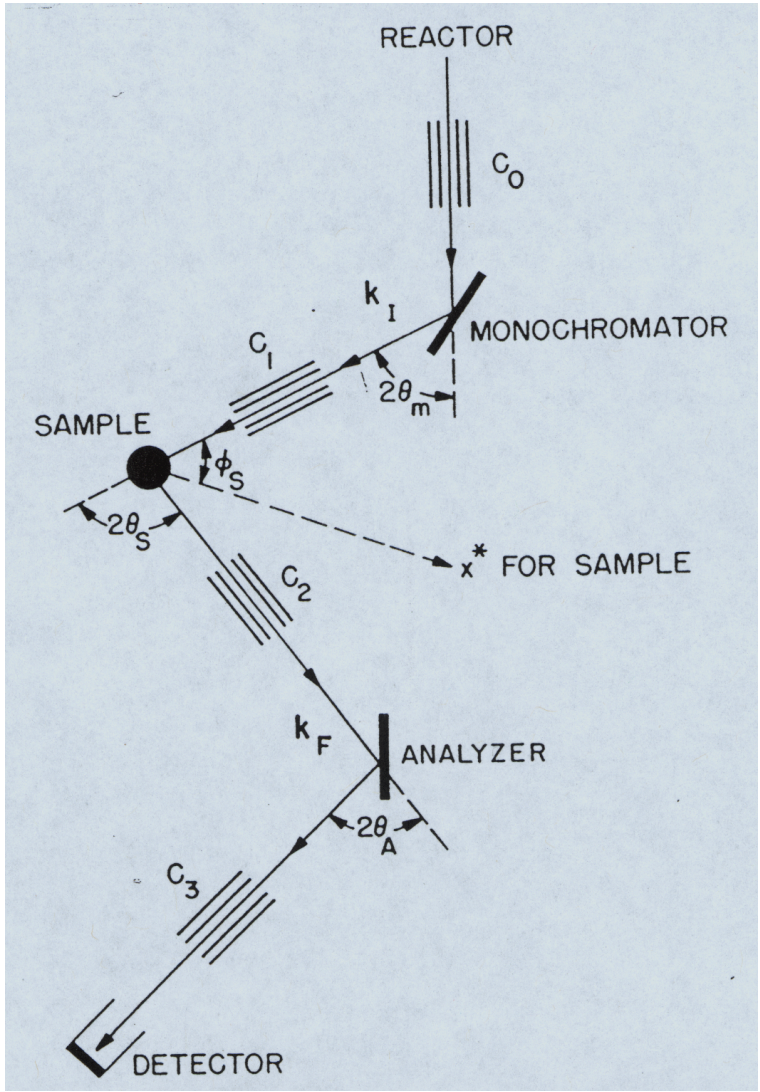
The Workhorse of Inelastic Scattering Instrumentation at Reactors Is the Three-axis Spectrometer





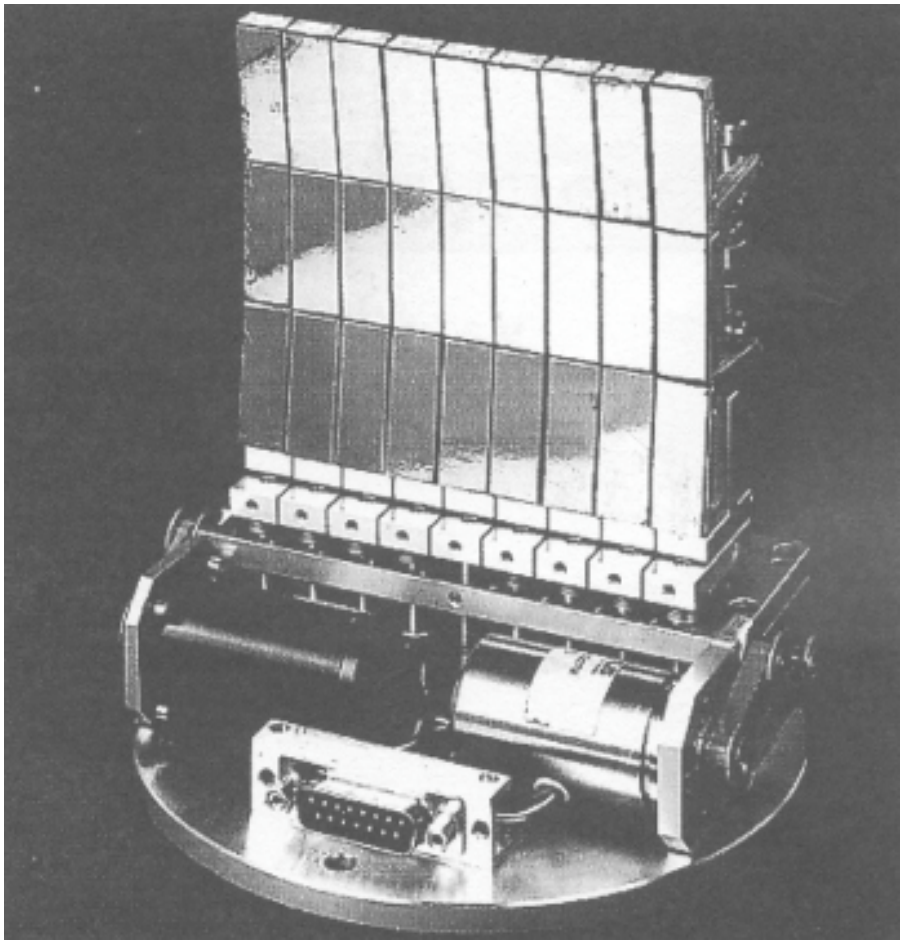
"scattering triangle"

Both k_I and k_F are defined by Bragg reflection from single crystals



TAS Monochromators and Analyzers

- Picture shows a modern monochromator made of 27 single crystals each of which can be oriented independently (Saclay)



- Different materials can be used: e.g. PG (002), Cu (220), Cu (002), Si (111), Heusler (111)
  - Different d-spacings give different energies at suitable scattering angles
  - Heusler alloy yields polarized neutrons

M & A make important contributions to TAS resolution function

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Monochromator/Analyzer Crystals

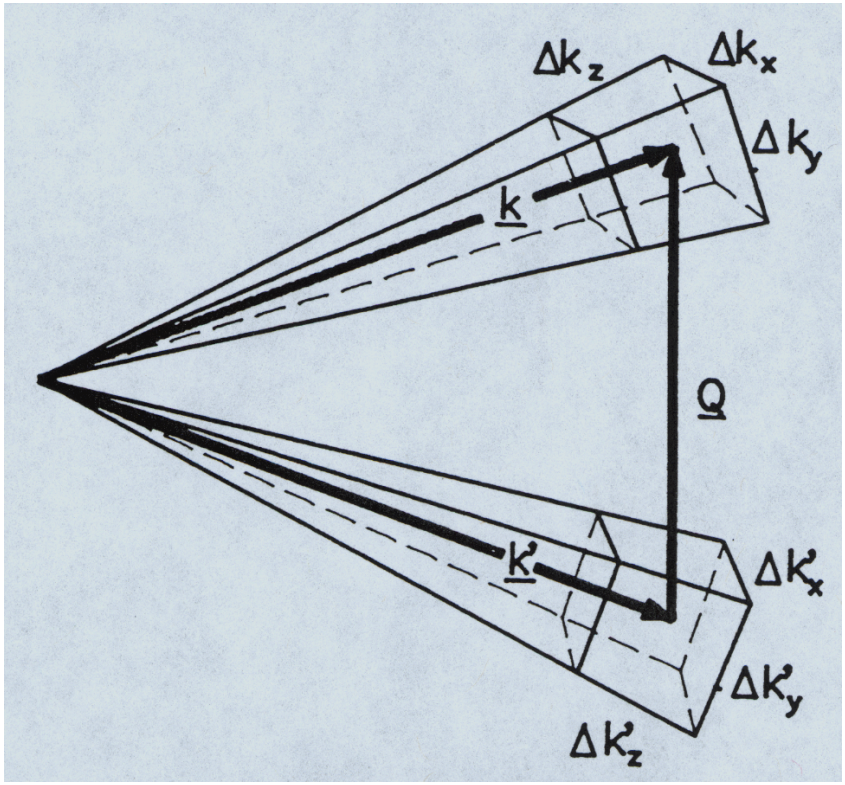
Table 9.1 Commonly used monochromator crystals and reflections. The energy ranges correspond to scattering angles Θ_m of 12° and 45°, respectively.

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	

Instrumental Resolution

- Uncertainties in the neutron wavelength & direction of travel imply that Q and E can only be defined with a certain precision

- When the box-like resolution volumes in the figure are convolved, the overall resolution width is the quadrature sum of the box sizes. Small “boxes” give good resolution.



- The total signal in a scattering experiment is proportional to the product of the “box” sizes

The better the resolution, the lower the count rate

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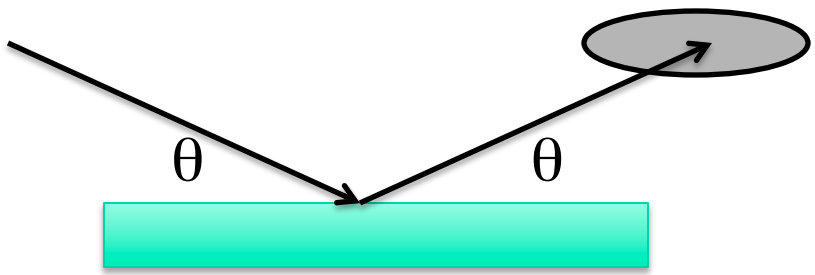
The Effect of Bragg's Law

- Bragg's Law $\lambda = 2d\sin\theta$ implies that the neutron wavelength and angle are correlated when neutrons are Bragg reflected
  - When θ increases so does λ

=>The resolution volume for each of k_i and k_f has a distinct orientation in Q-E space



- The result of convolving individual contributions is a resolution ellipse in Q-E
  - shape and size depend on the collimations and crystal mosaics of M and A
  - orientation depends on the sense of scattering at each of M, S, and A

⇒ focusing and defocusing arrangements

⇒ need for several M, A & C to tune intensity & resolution

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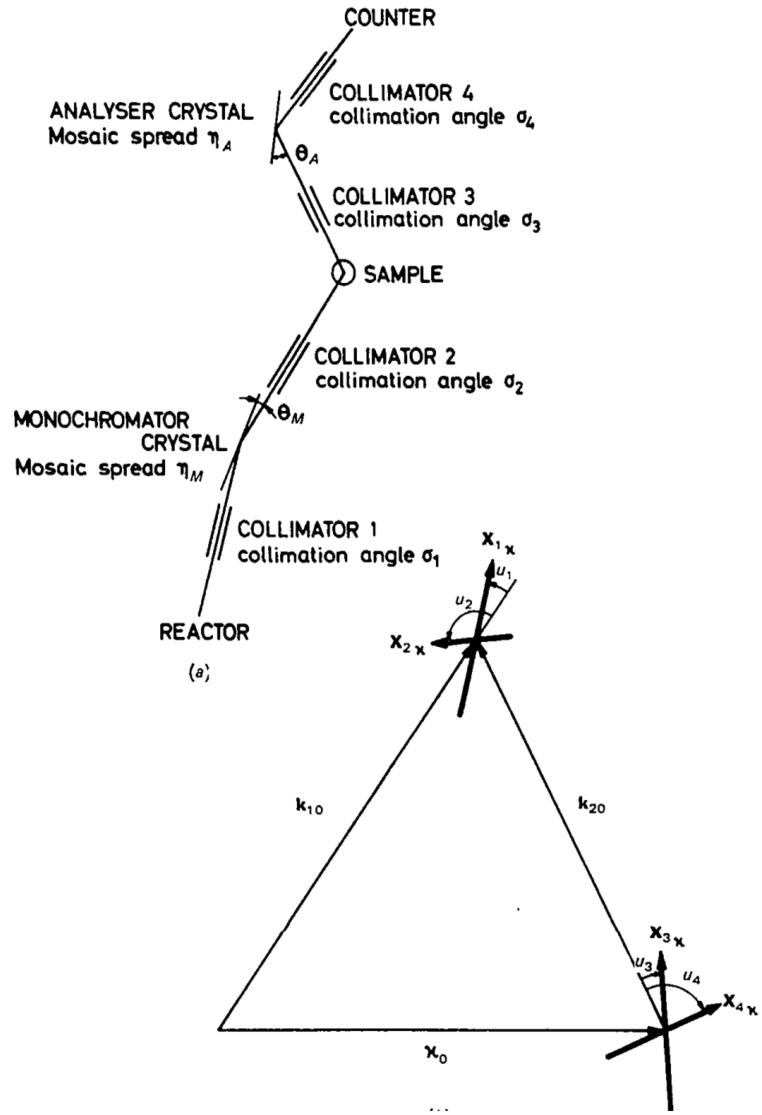
Working out the Resolution for a TAS

- It is possible to find a set of directions in Q-E space such that the neutron probability distributions along these directions are independent of each other
  - Usually each transmission function is considered to be a Gaussian function

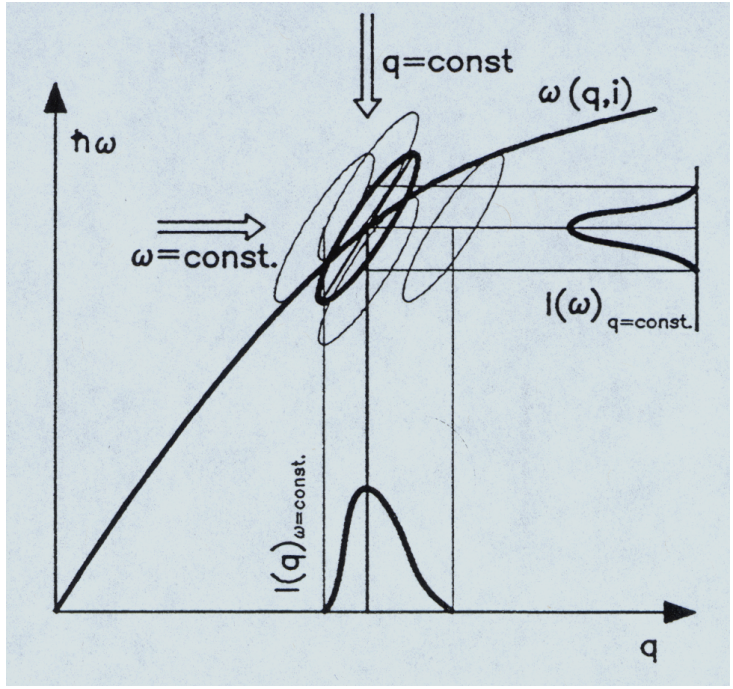
- Each distribution can then be convolved with the phonon cross section to calculate the measured intensity

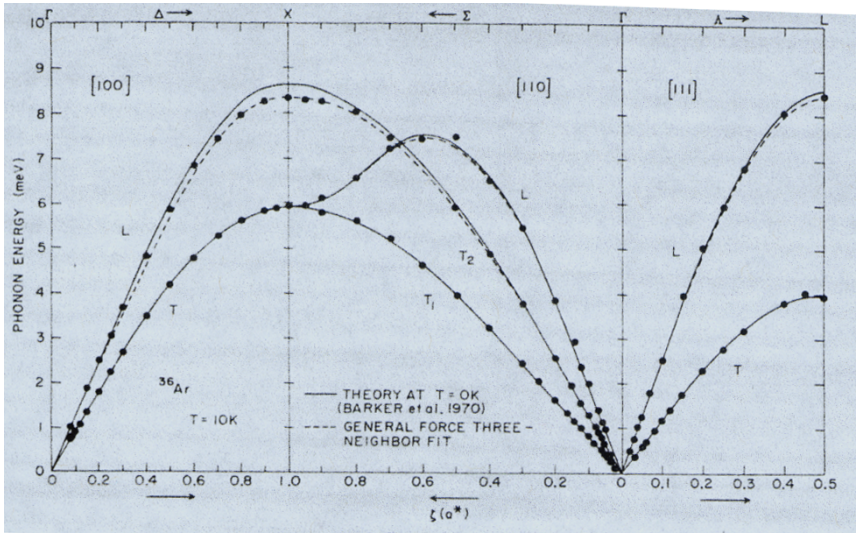
- See: Bjerrum Moller & Nielsen, Acta, Cryst., A25, 547 (1969); Werner and Pynn, J. Appl. Phys., 42, 4736 (1971); Pynn, Acta. Cryst., A39, 36 (1983)



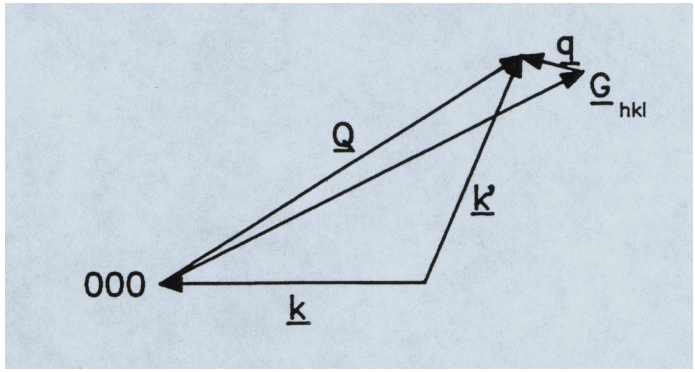
Focused Constant-Q and Constant-E Scans

- Point by point measurement in (Q,E) space
- Usually keep either k_I or k_F fixed
- Choose Brillouin zone (I.e. G) to maximize scattering cross section for phonons
- Scan usually either at constant-Q (Brockhouse invention) or constant-E





Phonon dispersion of ^{36}Ar



Vertical Focusing

- Phonon measurements are always made with single crystal samples, often with a plane of crystal symmetry coinciding with the scattering plane
  - Often phonon frequency achieves min or max value in this plane so Q-resolution can be opened up perpendicular to the plane to increase intensity without destroying measurement accuracy i.e. vertical resoln not important in this case
  - Exceptions exist: c.f. Werner and Pynn, J. Appl. Phys., 42, 4736 (1971)

- Vertical resolution is opened up by curving M &/or A vertically
  - Focusing distance depends on Bragg angle so radius, R, needs to be variable (L_0 = distance source to M; L_1 = distance M to S)
  - Vertical crystal mosaic of M simply blurs image at S and is undesirable
  - See Currat, Nucl. Inst. Meth., 107, 21 (1973)

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

Horizontal Focusing

- While vertical focusing does not (to lowest order) affect the Bragg condition at M or A, horizontal focusing does
  - With horizontal focusing, crystals at different horizontal positions on M or A have different Bragg angles and give different wavelengths
  - The effect is to increase intensity at the expense of resolution in both Q and E
  - Without horizontal focusing, increasing horizontal crystal mosaic of M increases intensity; same is true with horiz. focusing but image is also blurred by mosaic
  - Expressions for the resolution are given by Broholm, Nucl. Instr. Meth., A367, 169 (1996)

- Choice to be made
  - Can use real source as object to be focused => less control
  - Insert a variable aperture between source and M -> virtual source -> more control over resolution

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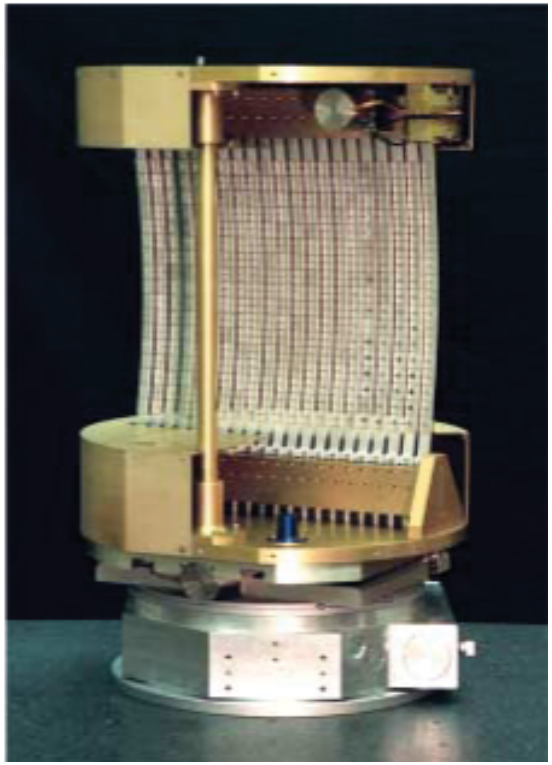
Neutron Monochromators

A simple, vertically focusing monochromator produced by Riso National Lab in Denmark comprised of 15 single crystals

Large single crystals of copper and Heusler alloy made at the ILL. The mosaic in different directions can be tailored by using controlled strain.



A vertical and horizontally focusing monochromator fabricated by a Johns Hopkins team for the NCNR. See: www.pha.jhu.edu/~broholm/homepage/talks/MACSmonochromatoracns.pdf

Design Process

1. Decide neutron energy-> choice of M and A crystals
2. Polarization? -> need non-magnetic construction
3. Reactor face or guide
4. Sample size (< 2 cm) and acceptable beam divergence (max 4° to give 10% resolution of BZ) ->
5. Guess L1 (want it as small as possible for focusing)->
6. L1 + divergence -> Monochromator size ->
7. Beam tube (or guide) size ->
8. Shielding requirements->
9. L1 -> step 5 and iterate
10. Secondary spectrometer is relatively simple
11. Use Monte Carlo to optimize distances, M & A sizes

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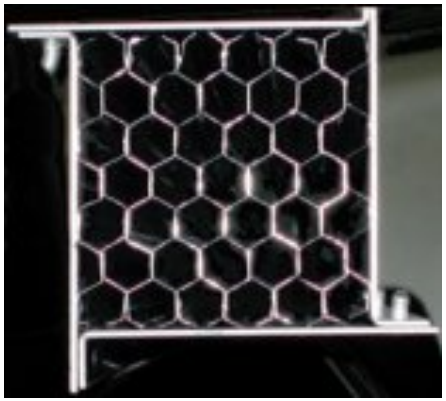
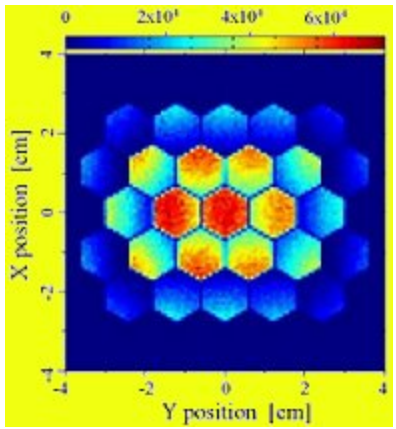
Direction of Travel of the Neutrons is Defined by Soller Collimators

- A Soller collimator is a set of parallel neutron-absorbing plates that define the direction of the neutron beam
  - Divergence distribution is triangular

Mylar collimator by Jens Linderholm, Denmark

- More sophisticated arrangements are possible but not generally used



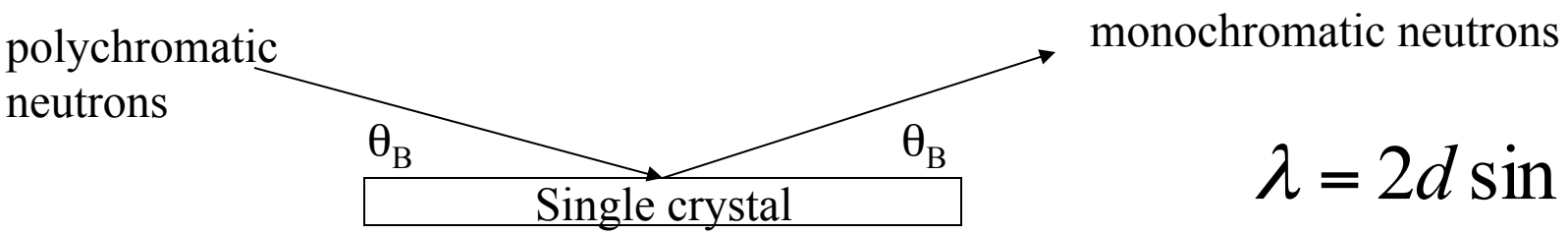


Honeycomb collimator for the Brillouin scattering instrument BRISP at ILL

- Intensity can be gained by:
  - Making absorbing plates very flat and straight (stretched mylar coated with Gd oxide paint)
  - Adding reflecting coatings
- Usual TAS collimators vary from 10’ to 80’ FWHM

Summary of Wavelength Selection

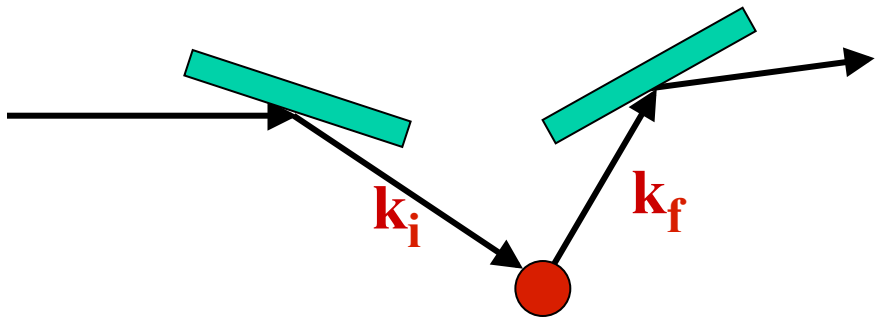
- Crystal monochromators are often used at CW neutron sources for diffractometers & three-axis machines
  - Bragg scattering from mosaic crystals defines the neutron wavelength
  - Crystal mosaic in the horizontal scattering plane controls intensity and $\delta\lambda/\lambda$
  - Crystal mosaic in vertical plane determines image beam size at the sample when focusing is used
  - Neutrons are often focused vertically – does not affect $\delta\lambda/\lambda$ but does worsen vertical Q resolution (often unimportant)
  - Neutrons are sometimes focused horizontally – degrades $\delta\lambda/\lambda$ and Q resolution in the scattering plane, but gains intensity



$$\lambda = 2d\sin\theta_B$$

Triple Axis Spectrometers

Triple axis spectrometers are typically used when either the *direction* of **Q** is important or the interesting region of **Q-ω** space is of limited extent.



Remember – Intensity ↓

Resolution ↑

Orientation of resolution function depends on the sense of scattering at M, S, and A. Leads to focusing and non-focusing configs.

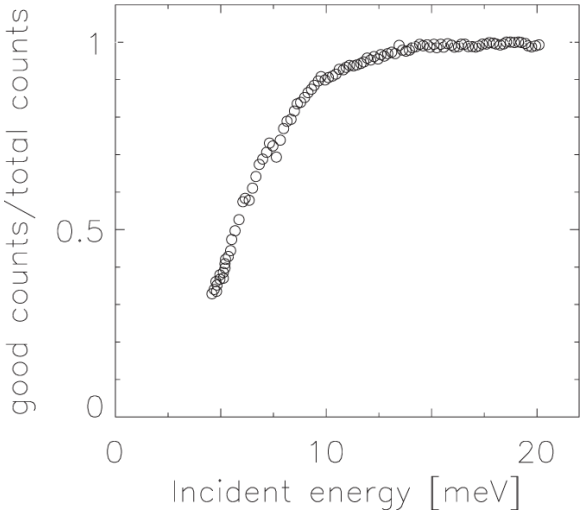
Collimation(')	λ	rel. signal	FWHM .
55-80-80-80	4 Å	1.00	0.28 meV
55-40-40-40	4 Å	0.24	0.17 meV
69-80-80-80	5 Å	0.26	0.13 meV
84-80-80-80	6.1 Å	0.03	0.05 meV

Viewgraph courtesy of Dan Neumann

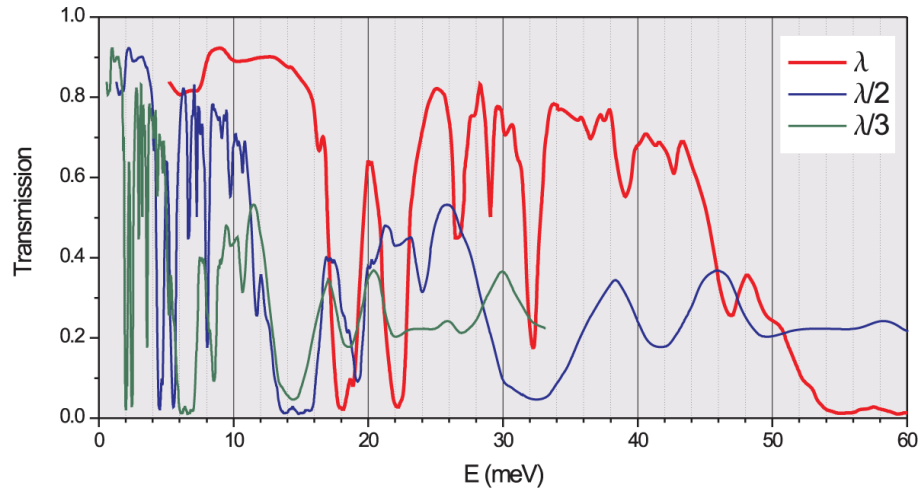
Scanning Modes

- Scans are usually either constant-Q or constant-E
- In both cases, either k_I or k_F can be fixed
- Notice that the phonon cross section has a factor k_F/k_I
- To measure the cross section we want to know the scattered intensity per incident neutron
- To measure the number of incident neutrons/sec we use a low efficiency monitor whose efficiency ~ 1/v
- If we fix k_I, this efficiency is fixed and we measure signal per N monitor counts
- An issue may arise if the monochromator has second order contamination e.g. if PG (002) reflects so does PG (004) because $n\lambda = 2d\sin\theta$

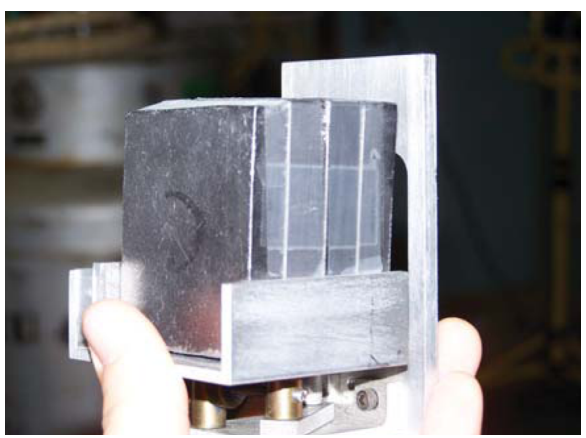
Second Order Filters



Example of monitor contamination due to higher order neutrons



Transmission of pyrolytic graphite as a function of neutron energy. Notice that 14 meV (k=2.6) transmission is good but 2nd and 3rd order are suppressed

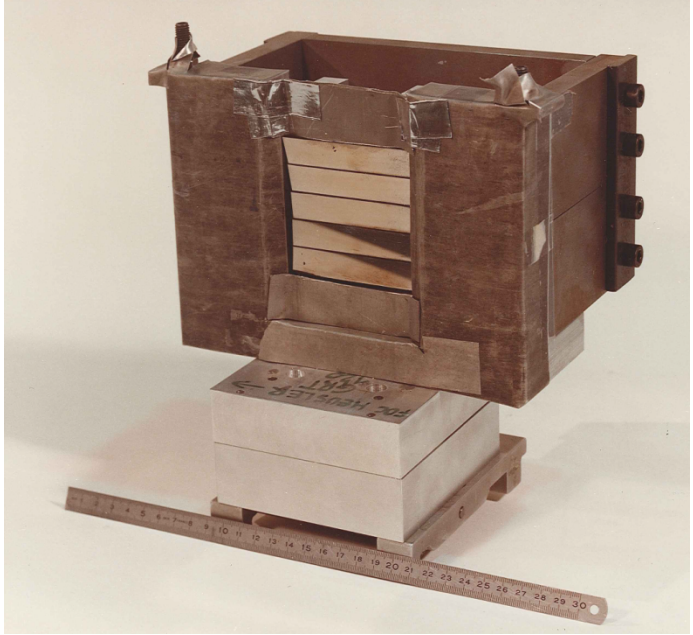


Typical pyrolytic graphite filter a few centimeters thick

Note: A filter may not be necessary on a cold neutron TAS on a guide because the flux of second order neutrons is much lower than on thermal TAS at a reactor face

For Polarized Neutrons Heusler Alloy Monochromators may be used

New IN20 polarized –neutron monochromator. The vertical curvature is fixed while the horizontal curvature is variable



Original IN12 polarized-neutron analyzer (circa 1980). The Heusler crystals are mounted on a spring-steel plate which can be curved in the vertical plane by tugging on the back

Spurions & Background

- TAS has low signal intensity => susceptible to background
- Careful shielding is needed
  - Monochromator drum must stop fast neutrons and gammas diffusely scattered by M and in line of sight; must also soak up unwanted thermal neutrons => mixture of heavy material (e.g. [non-magnetic] iron), moderator (e.g. hydrogen [in concrete or wax or polyethylene]) plus neutron absorber (e.g. boron)
  - Beam line shielding to define usable neutron beam
  - Tight shielding around analyzer and detector (last layer often Cd)

- Supposed to be M (strong) -> S (weak) -> A (strong) but any permutation is possible
  - This can lead to spurious peaks e.g. higher order from M &/or A plus Bragg in sample or sample can
  - Possible "spurions" discussed by Shirane et al "**Neutron Scattering With a Triple-Axis Spectrometer: Basic Techniques"** published by CUP

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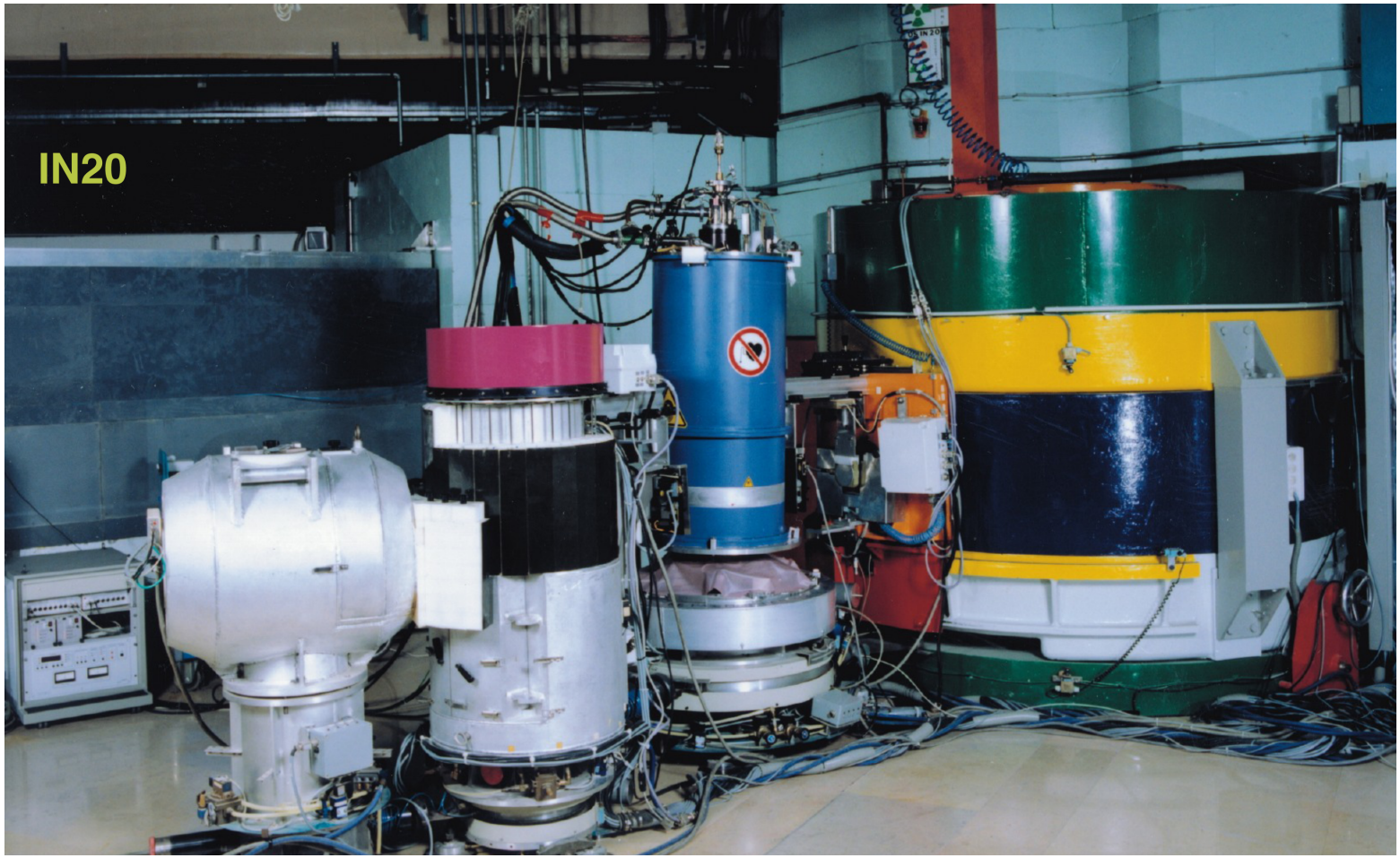
IN20 at ILL: A Thermal TAS with Polarization Analysis



Flexible distance between units allows additional equipment (eg magnets for spin echo phonon focusing). Non-magnetic construction

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Conclusion

Although the triple-axis crystal spectrometer has been in use at neutron scattering laboratories for some twenty-five years, it remains one of the most valuable tools for detailed studies of the dynamical response of a sample. In particular, where controlled (and understood) resolution in both energy and wavevector are required, this type of instrument is still unchallenged. Indeed in many neutron laboratories, triple-axis spectrometers are the principal instruments used in condensed matter research.

W.G. Stirling & R. Pynn,
IN14 project, AR ILL 1982

My Bottom Line

- I'm not smart enough to be able to tell you how to design a good TAS
- The modern machines like IN20, IN14 and IN8B are very sophisticated and have evolved incrementally over many years
- Each generation probably gains a factor of 3 – 5
- There is no reason to believe that this will stop as each new generation digests the lessons learned by their predecessors and adds their own refinements
- Whatever you do, study and understand all aspects of the existing designs before you try to design something new
- It may turn out that a cold TAS on an LPSS like ESS, using the pulse structure to reduce background and sophisticated optics (eg elliptical guides) is a real winner.

INSIS