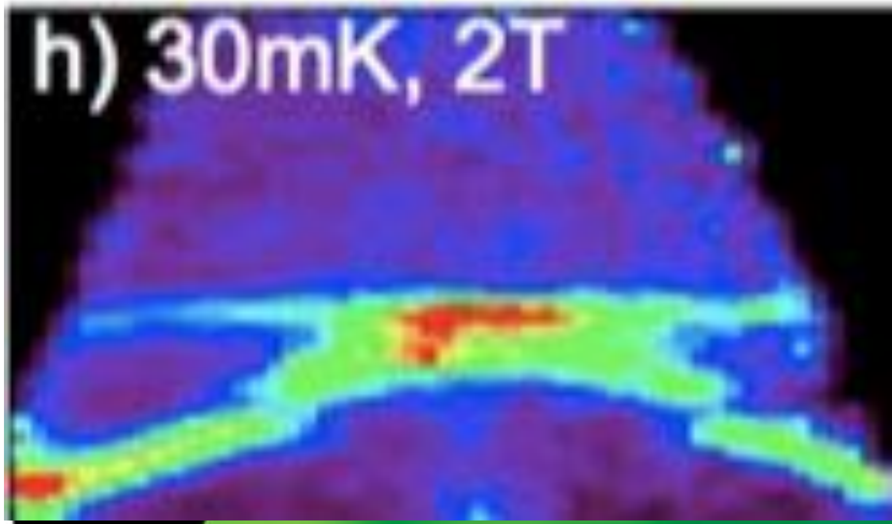
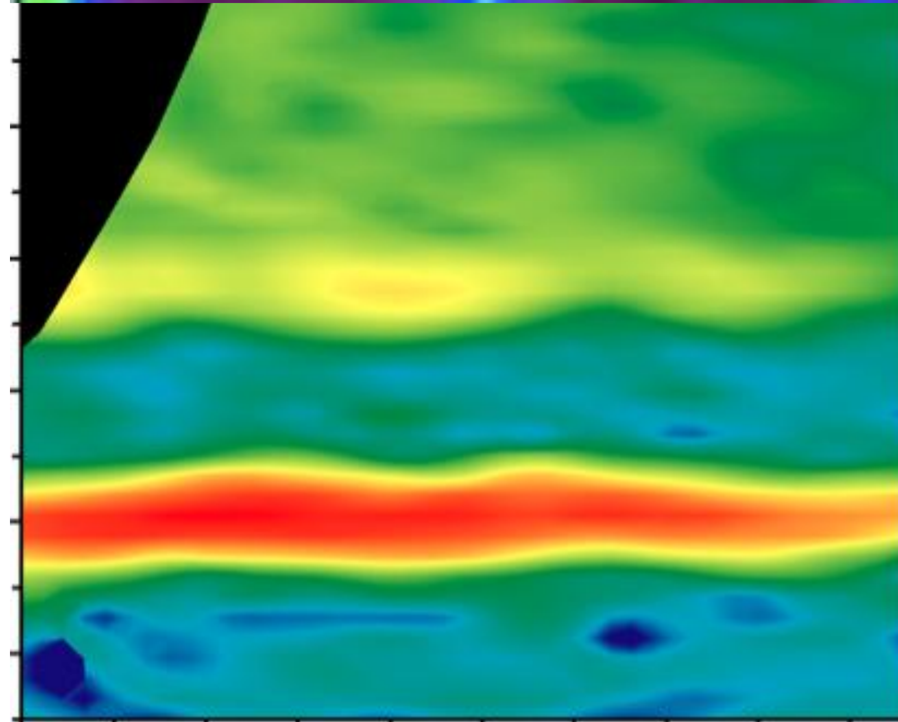
Introduction to Inelastic Neutron Scattering

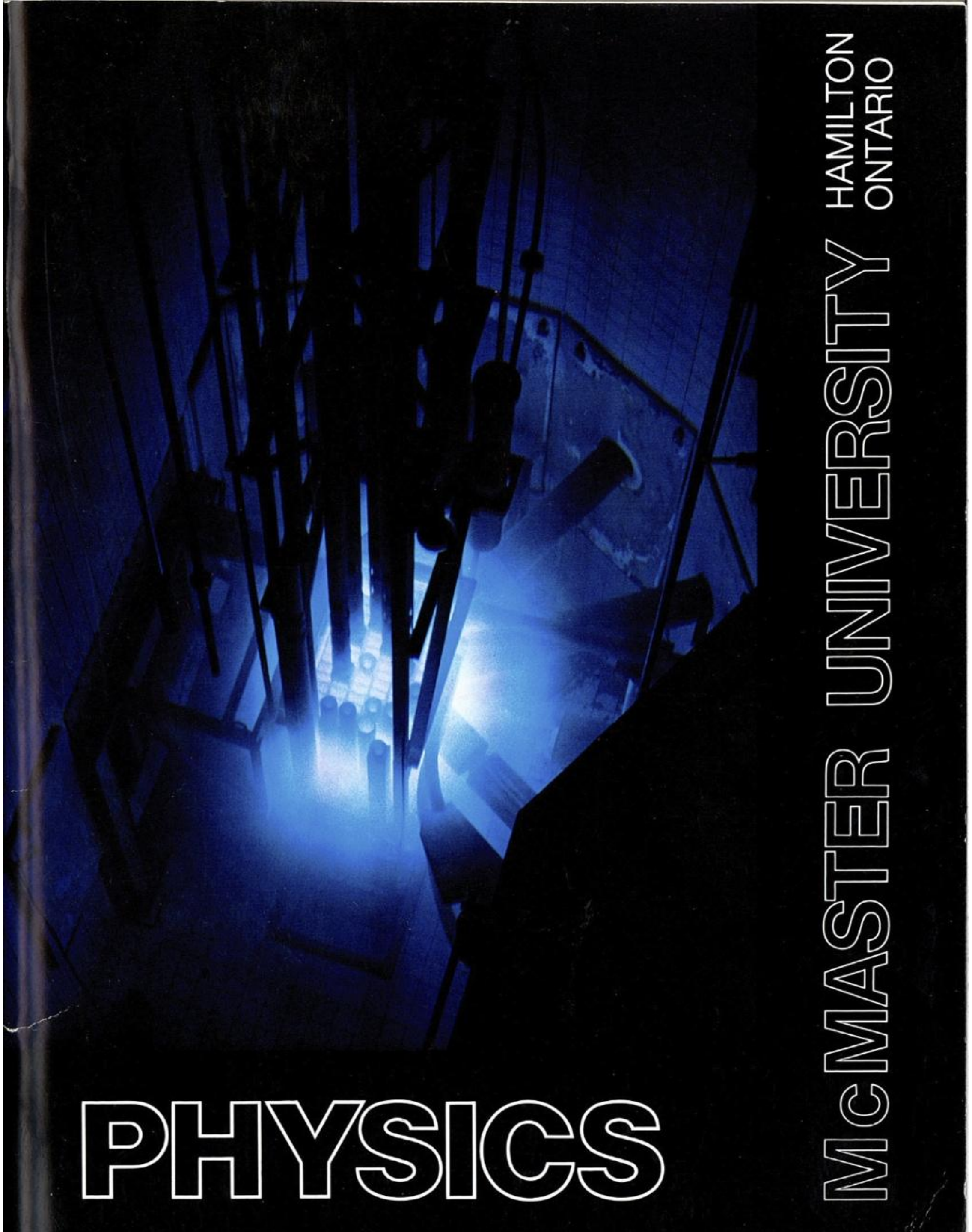
Bruce D Gaulin
McMaster University

Brockhouse Institute for Materials Research





- *Neutrons: Properties and Cross Sections*
- *Excitations in solids*
- *Triple Axis and Chopper Techniques*
- *Practical concerns*



^{235}U + n

→

daughter nuclei +

2-3 n + gammas

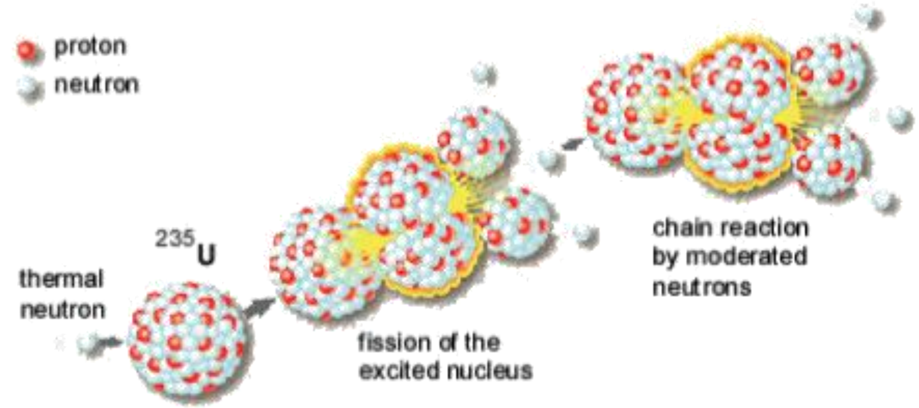
neutrons:

no charge

s=1/2

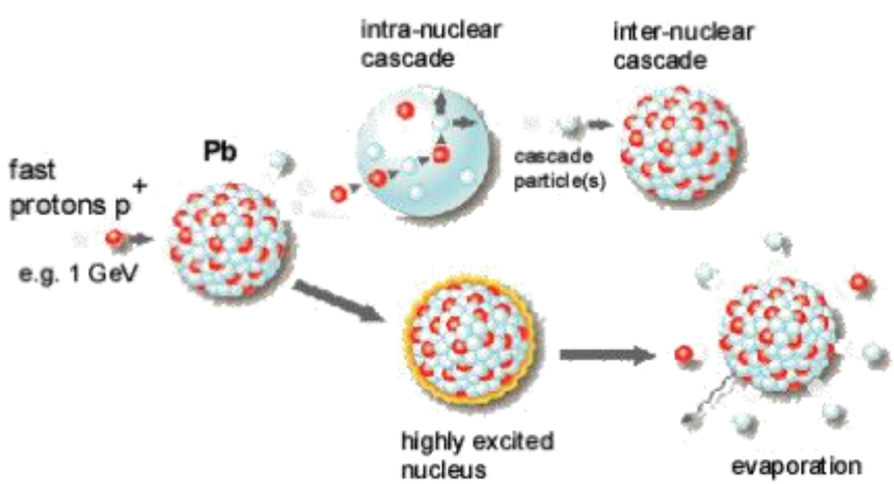
massive: mc^2~1 GeV

How do we produce neutrons



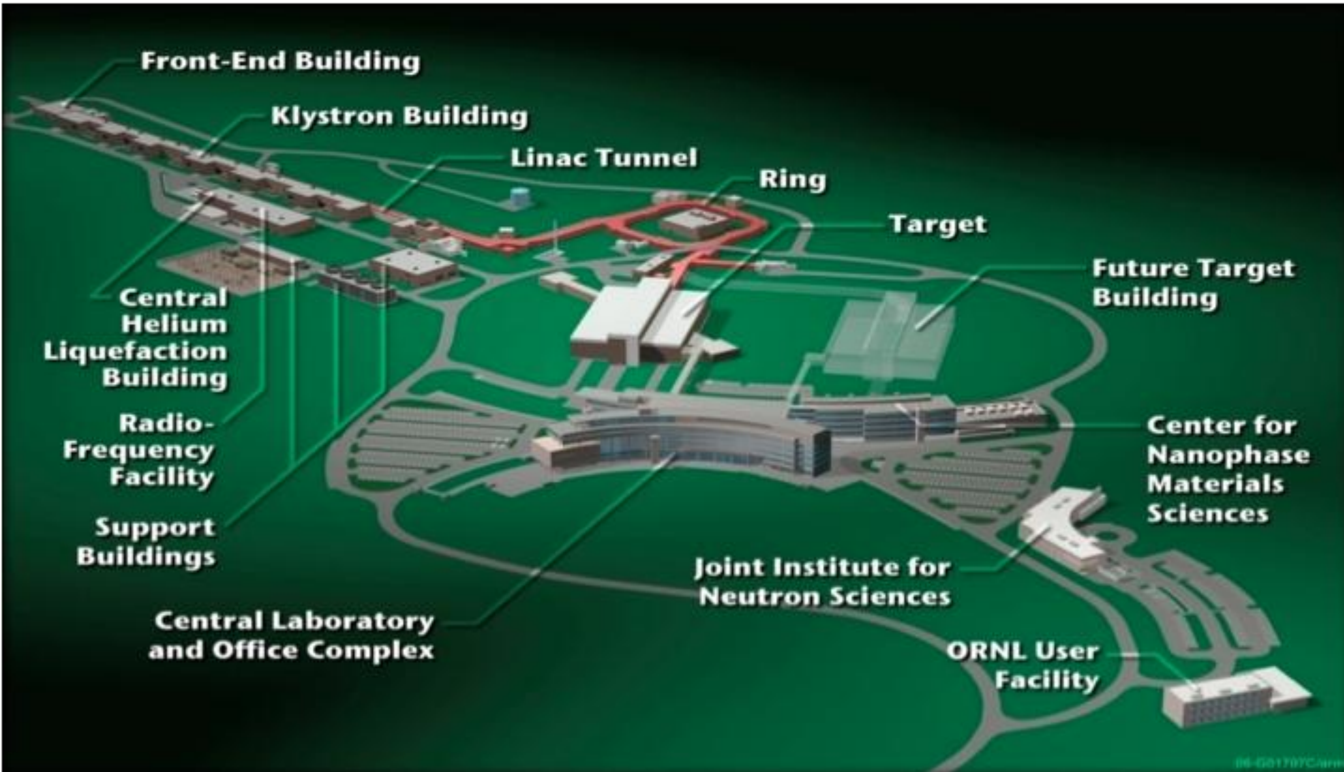
Fission

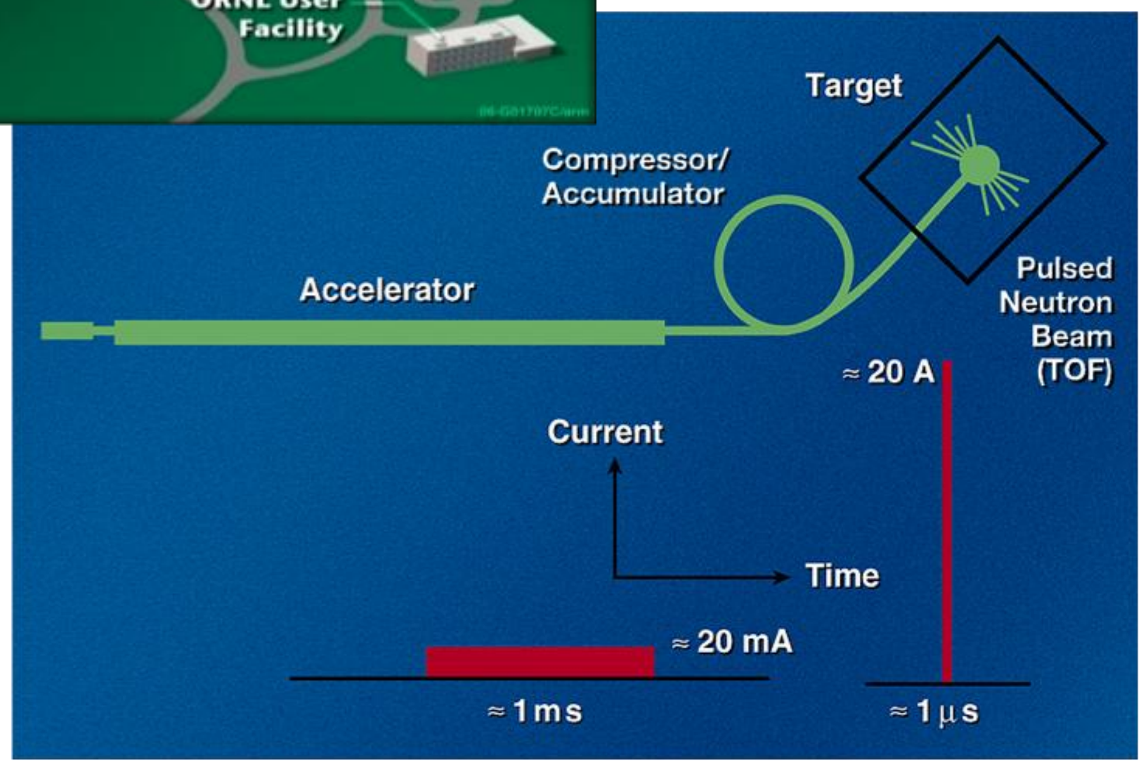
- chain reaction
- continuous flow
- 1 neutron/fission



Spallation

- no chain reaction
- pulsed operation
- 30 neutrons/proton

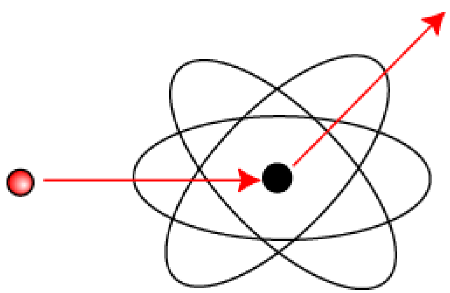

Front-End Building
Klystron Building
Linac Tunnel
Ring
Target
Future Target Building
Central Helium Liquefaction Building
Radio-Frequency Facility
Support Buildings
Center for Nanophase Materials Sciences
Joint Institute for Neutron Sciences
Central Laboratory and Office Complex
ORNL User Facility


Target
Compressor/ Accumulator
Accelerator
Pulsed Neutron Beam (TOF)
≈ 20 A
Current
Time
≈ 20 mA
≈ 1 ms
≈ 1 μs

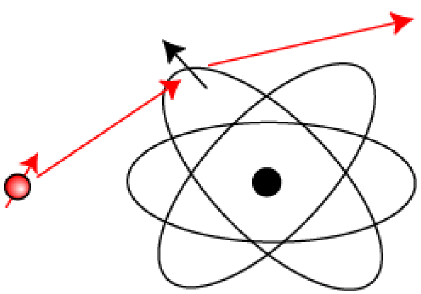
Neutron interactions with matter

- **Properties of the neutron**
  - Mass m_n =1.675 x 10^{-27} kg
  - Charge 0
  - Spin-1/2, magnetic moment μ_n = -1.913 μ_N

- **Neutrons interact with…**
  - Nucleus
  - Crystal structure/excitations (eg. Phonons)
  - Unpaired electrons via dipole scattering
  - Magnetic structure and excitations



Nuclear scattering



Magnetic dipole scattering

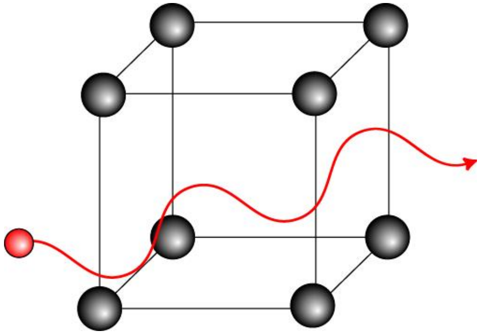
Wavelength-energy relations

- **Neutron as a wave …**
  - Energy (E), velocity (v), wavenumber (k), wavelength (λ)

$$k = \frac{m_n \mathrm{v}}{\hbar} = \frac{2\pi}{\lambda}$$

$$E = \frac{\hbar^2 k^2}{2m_n} = \frac{\hbar^2}{2m_n}\left(\frac{2\pi}{\lambda}\right)^2 = \frac{81.81\,\mathrm{meV}\cdot\text{Å}^2}{\lambda^2}$$

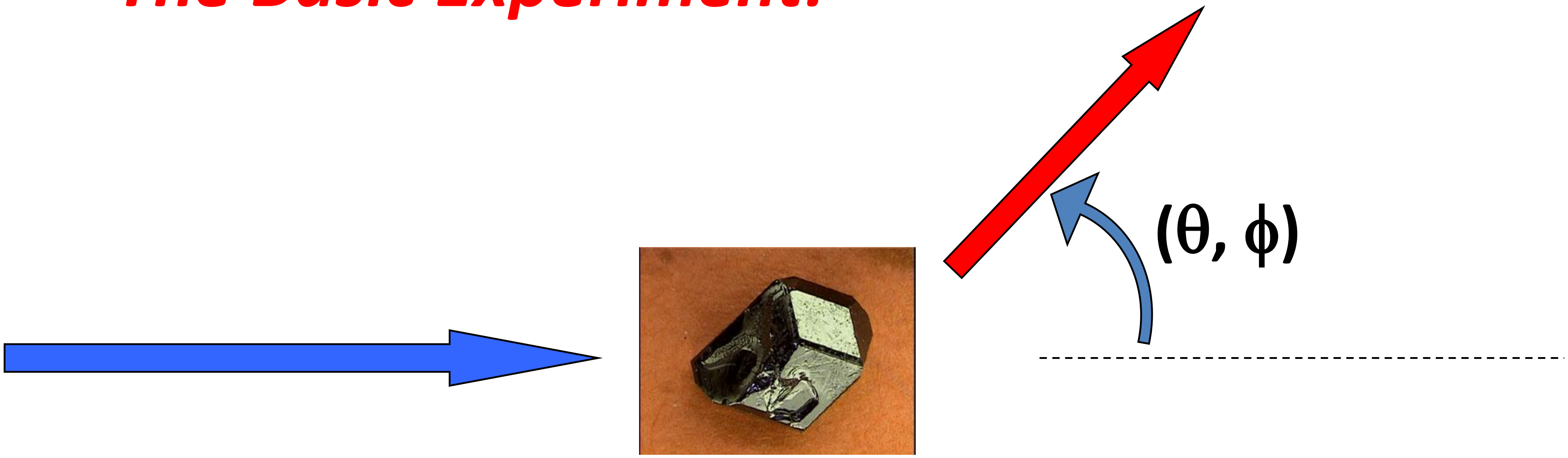
$$E = k_B T = \left(0.08617\,\mathrm{meV}\cdot\mathrm{K}^{-1}\right)T$$



λ ~ interatomic spacing → E ~ excitations in condensed matter

	Energy (meV)	Temperature (K)	Wavelength (Å)
Cold	0.1 – 10	1 – 120	4 – 30
Thermal	5 – 100	60 – 1000	1 – 4
Hot	100 – 500	1000 – 6000	0.4 – 1

The Basic Experiment:



Incident Beam:

- **monochromatic**
- **“white”**
- **“pink”**

Scattered Beam:

- **Resolve its energy**
- **Don’t resolve its energy**
- **Filter its energy**

Fermi's Golden Rule within the 1st Born Approximation

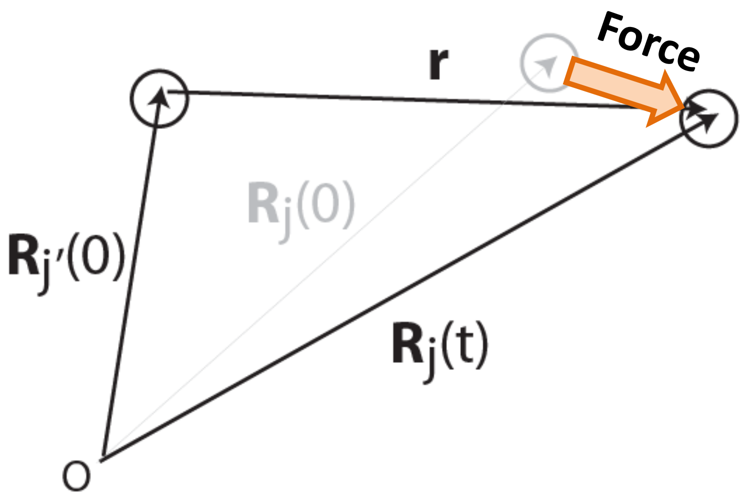
$$W = 2\pi / h \quad |< f \mid V \mid i>|^2 \quad \rho\,(E_f)$$

$$\delta\sigma = W / \Phi \;=\; (m/2\pi h^2)^2 \;\; k_f / k_i \quad |< f \mid V \mid i>|^2 \;\; \delta\Omega$$

$$\delta^2\sigma / \delta\Omega\, \delta E_f = \; k_f/k_i \;\; \sigma_{coh}/4\pi \;\; N \;\; S_{coh}(\mathbf{Q}, \omega)$$

$$+ \, k_f/k_i \; \sigma_{incoh}/4\pi \;\; N \;\; S_{incoh}(\mathbf{Q}, \omega)$$

Nuclear correlation functions



Pair correlation function

$$G(\mathbf{r},t)=\frac{1}{N}\int\sum_{jj'}\delta\big(\mathbf{r}'-\mathbf{R}_{j'}(0)\big)\delta\big(\mathbf{r}'+\mathbf{r}-\mathbf{R}_{j}(t)\big)d\mathbf{r}'$$

Intermediate function

$$I(\mathbf{Q},t)=\int G(\mathbf{r},t)e^{i\mathbf{Q}\cdot\mathbf{r}}d\mathbf{r}=\frac{1}{N}\sum_{jj'}\exp\big(-i\mathbf{Q}\cdot\mathbf{R}_{j'}(0)\big)\exp\big(i\mathbf{Q}\cdot\mathbf{R}_{j}(t)\big)$$

Scattering function

$$S(\mathbf{Q},\omega)=\frac{1}{2\pi\hbar}\int I(\mathbf{Q},t)e^{-i\omega t}dt$$

Differential scattering cross-section

$$\frac{d^2\sigma}{d\Omega dE_f}=\frac{\sigma_{scat}}{4\pi}\frac{k_f}{k_i}NS(\mathbf{Q},\omega)$$

Nuclear (lattice) excitations

McMaster University

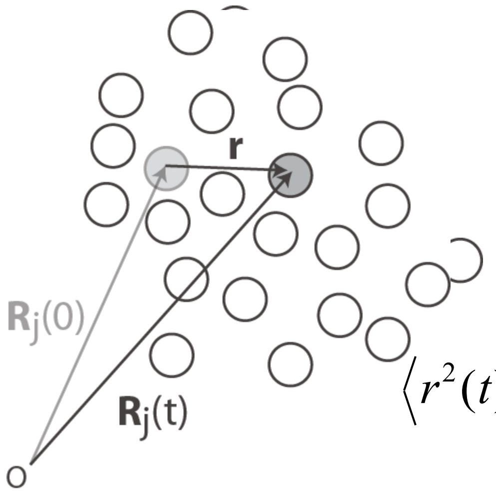
Neutron scattering measures simultaneously the wavevector and energy of **collective excitations** → dispersion relation, $\omega(\mathbf{q})$
In addition, **local excitations** can of course be observed

- **Commonly studied excitations**
  - Phonons
  - Librations and vibrations in molecules
  - Diffusion
  - Collective modes in glasses and liquids
- **Excitations can tell us about**
  - Interatomic potentials & bonding
  - Phase transitions & critical phenomena (soft modes)
  - Fluid dynamics
  - Momentum distributions & superfluids (eg. He)
  - Interactions (eg. electron-phonon coupling)

Atomic diffusion

McMaster University

For long times compared to the collision time, atom diffuses



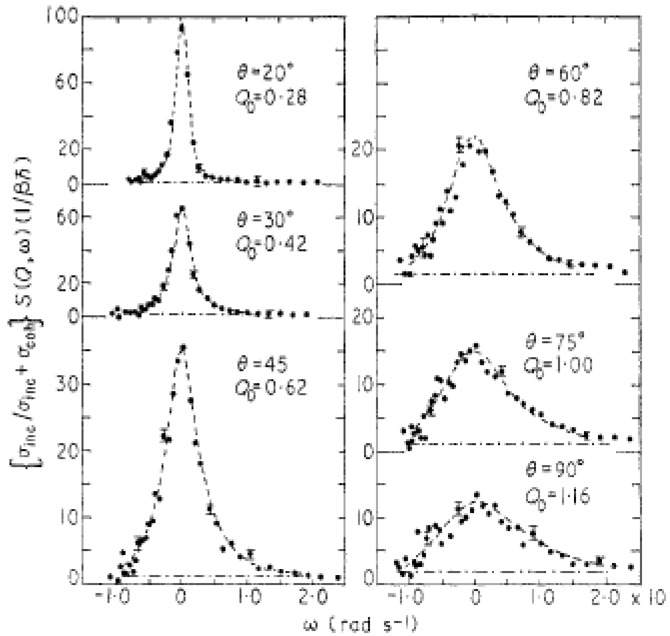
$$\langle r^2(t)\rangle \approx 6Dt$$

Auto-correlation function

$$G_s(r,t) = \left\{6\pi\langle r^2(t)\rangle\right\}^{-3/2}\exp\left(-\frac{r^2}{6\langle r^2(t)\rangle}\right)$$

$$S(Q,\omega) = \frac{1}{\pi\hbar}\exp\left(\frac{\hbar\omega}{2k_BT}\right)\frac{DQ^2}{\omega^2+(DQ^2)^2}$$

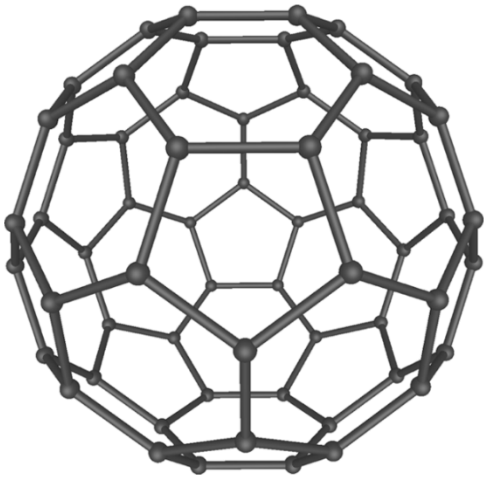
Liquid Na



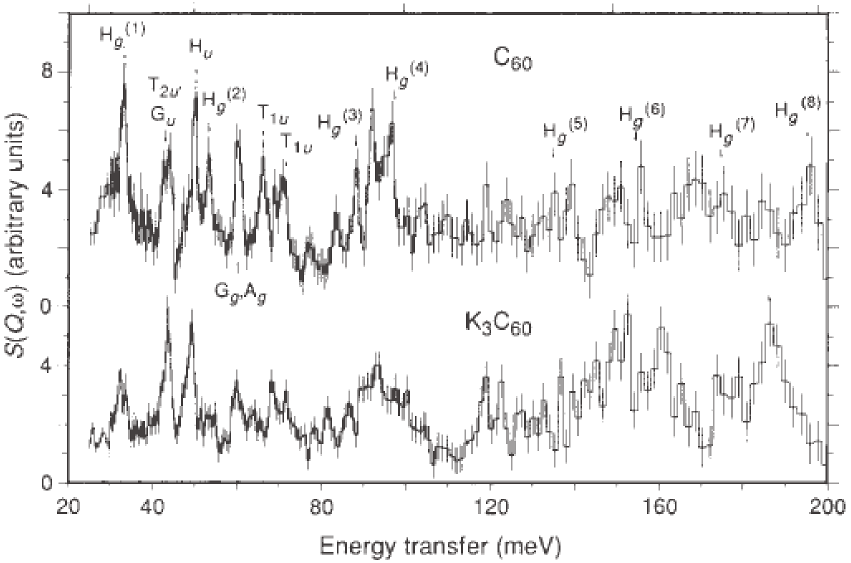
Cocking, *J. Phys. C* **2**, 2047 (1969)..

Molecular vibrations

- Large molecule, many normal modes
- Harmonic vibrations can determine interatomic potentials



C60 molecule

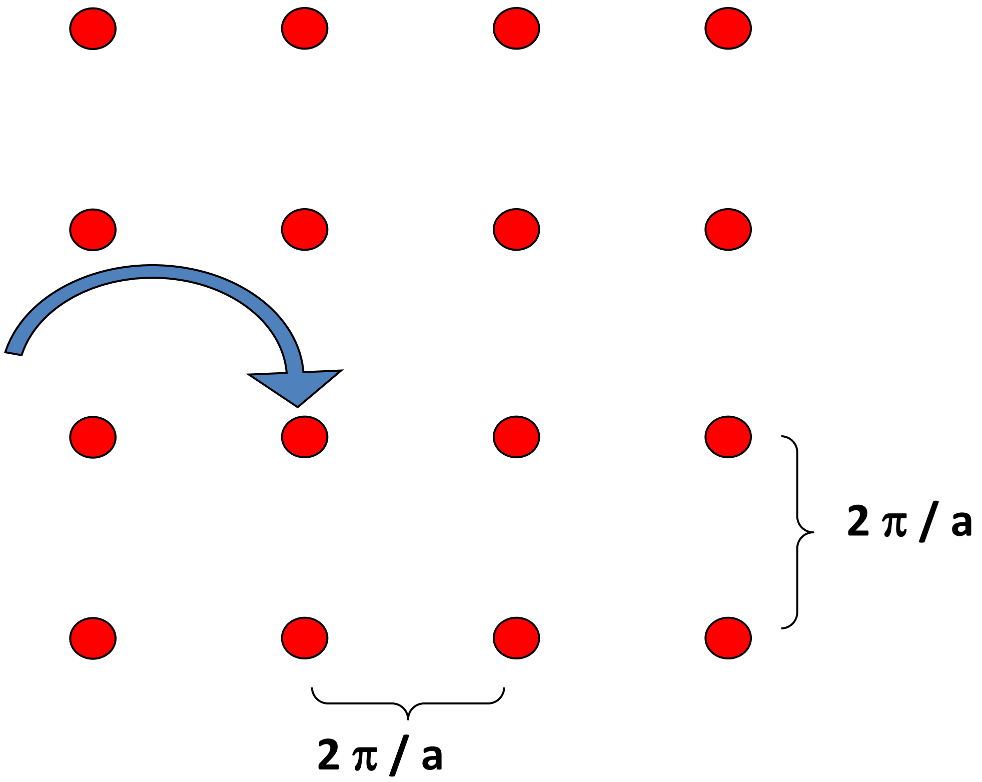


Prassides *et al., Nature* **354**, 462 (1991).

Mapping Momentum – Energy (Q-E) space

Origin of reciprocal space;

Remains fixed for any sample rotation

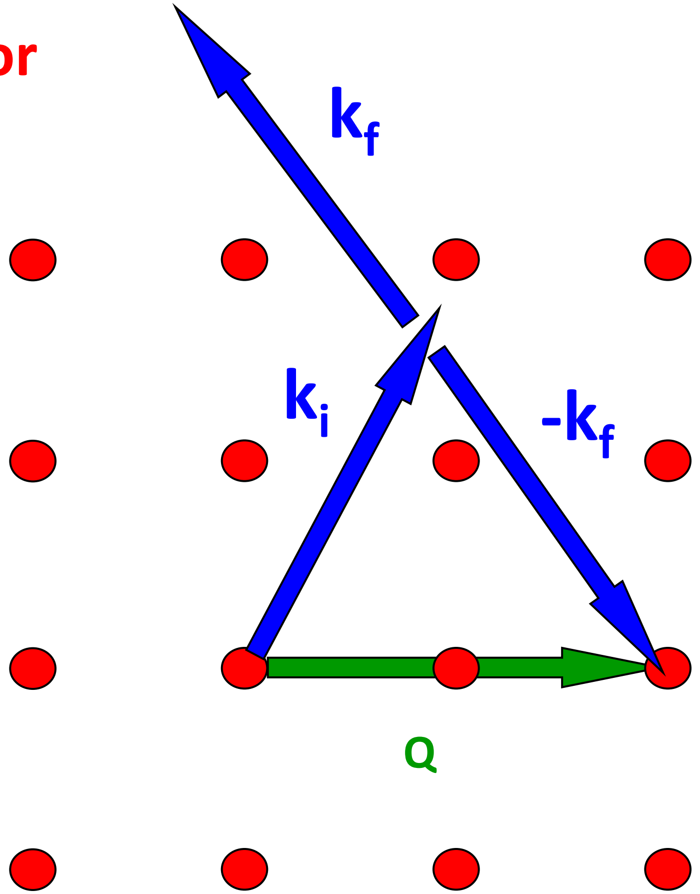


Bragg diffraction:

Constructive Interference

Q = Reciprocal Lattice Vector

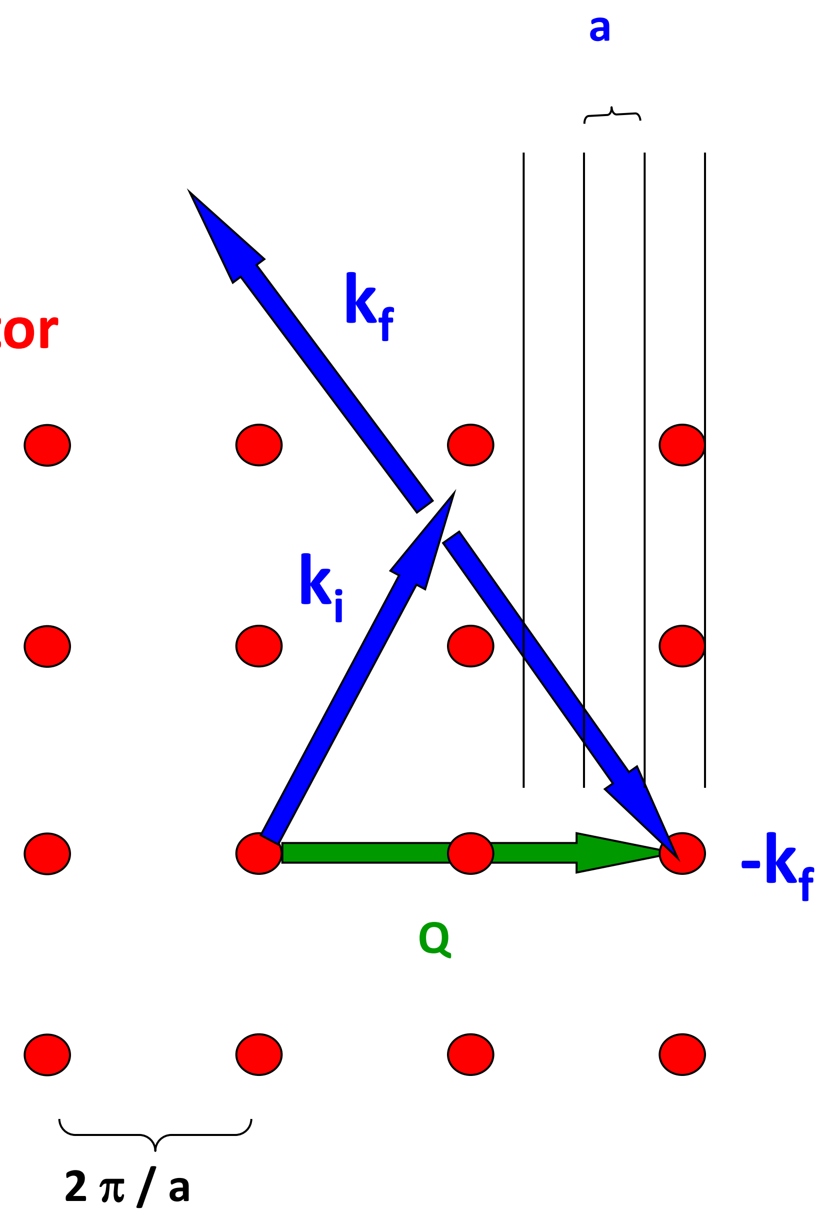
Elastic scattering : $| k_i | = | k_f |$



Bragg diffraction:

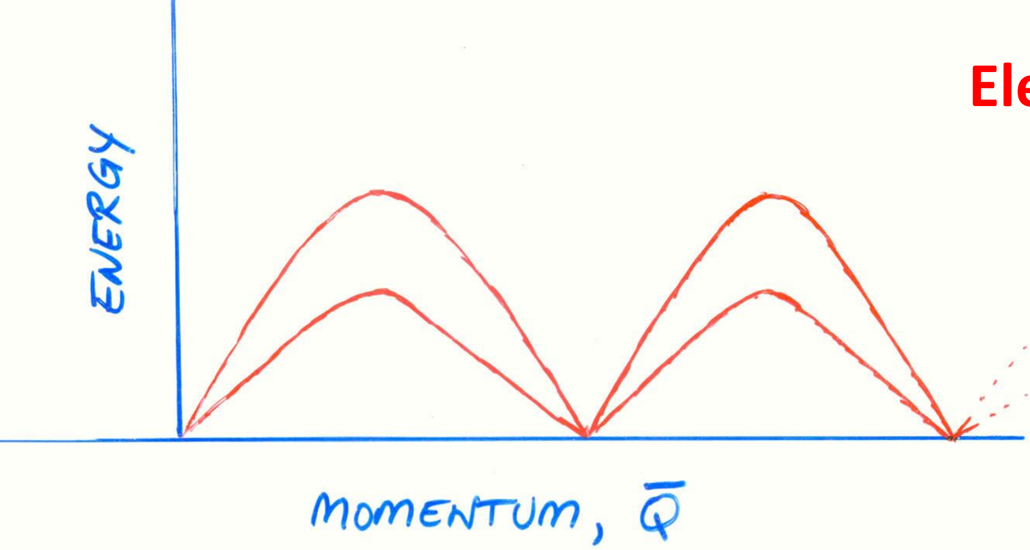
Constructive Interference

Q = Reciprocal Lattice Vector

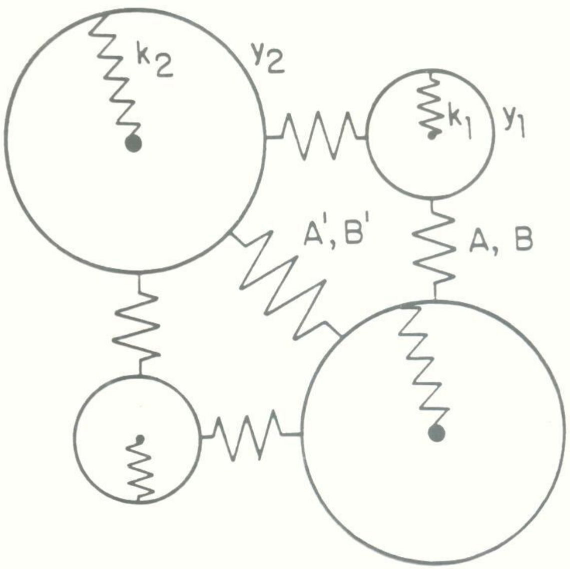


Elastic scattering : $|k_i| = |k_f|$

Elementary Excitations in Solids



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



Energy vs Momentum

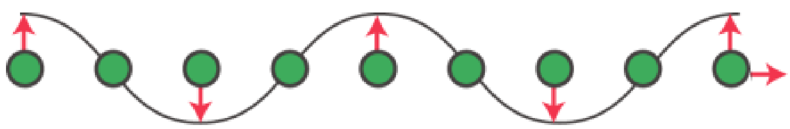
- Forces which bind atoms together in solids

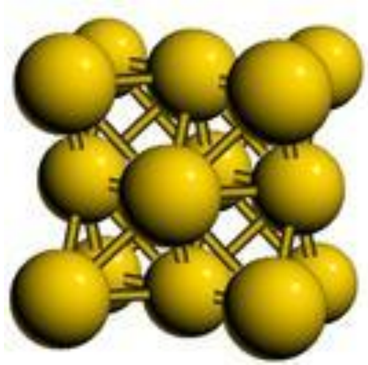
Phonons

- Normal modes in periodic crystal → wavevector

$$\mathbf{u}(l,t)=\frac{1}{\sqrt{NM}}\sum_{j\mathbf{q}}\boldsymbol{\varepsilon}_j(\mathbf{q})\exp(i\mathbf{q}\cdot\mathbf{l})\hat{B}(\mathbf{q}j,t)$$

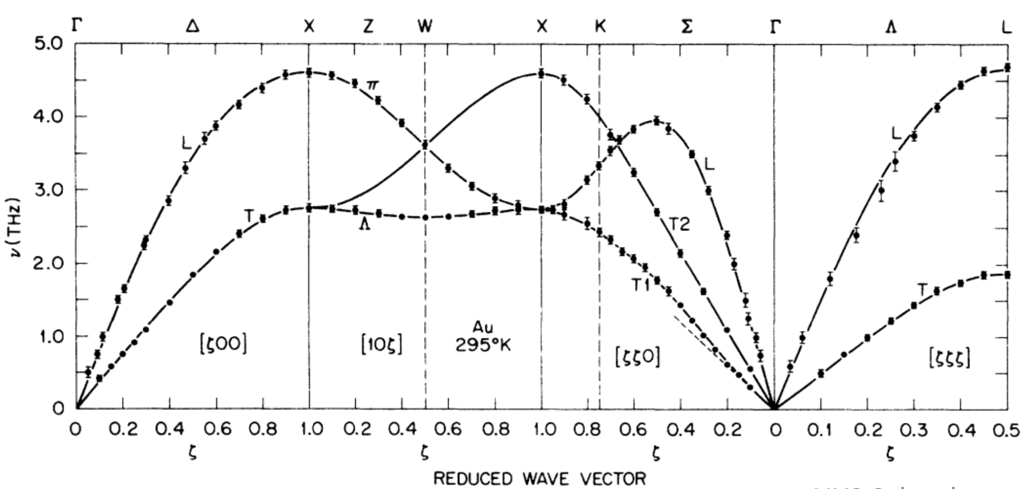
- Energy of phonon depends on **q** and polarization



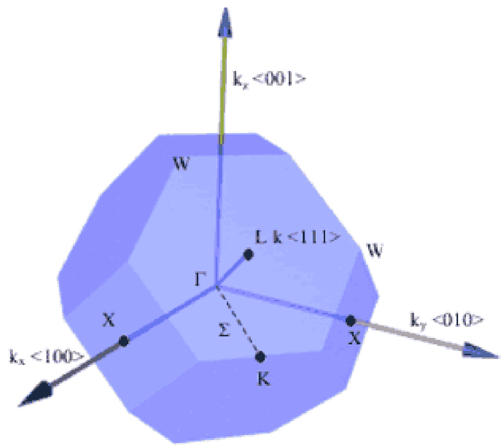


FCC structure

Longitudinal mode / Transverse mode



Lynn, *et al.*, *Phys. Rev. B* **8**, 3493 (1973).

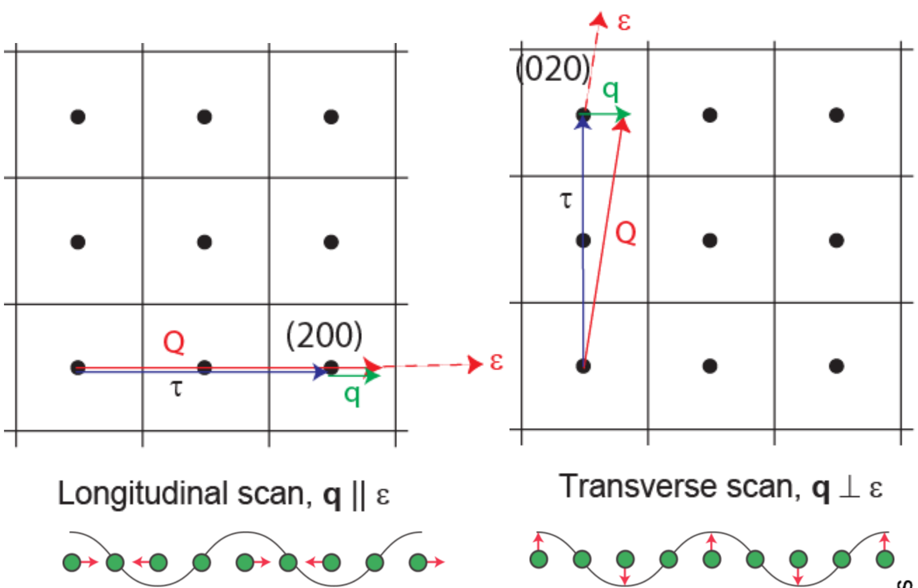


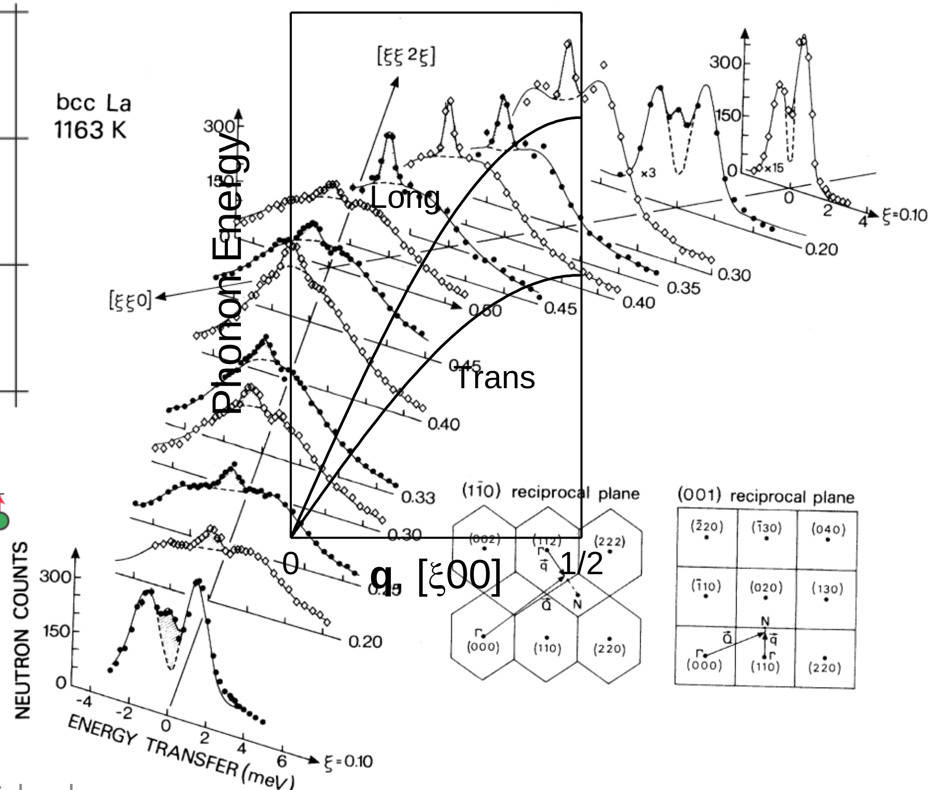
FCC Brillouin zone

Phonon intensities

Structure (polarization) factor

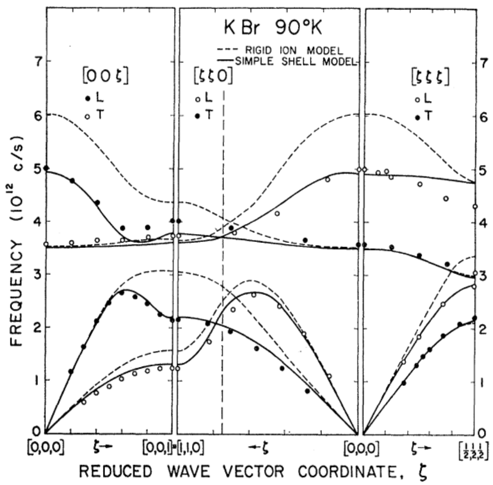
$$S_{1+}(\mathbf{Q},\omega)=\frac{1}{2NM}e^{-Q^2\langle u^2\rangle}\sum_{j\mathbf{q}}\frac{\left|\mathbf{Q}\cdot\boldsymbol{\varepsilon}_j(\mathbf{q})\right|^2}{\omega_j(\mathbf{q})}\left(1+n(\omega)\right)\delta(\mathbf{Q}-\mathbf{q}-\boldsymbol{\tau})\delta\left(\omega-\omega_j(\mathbf{q})\right)$$



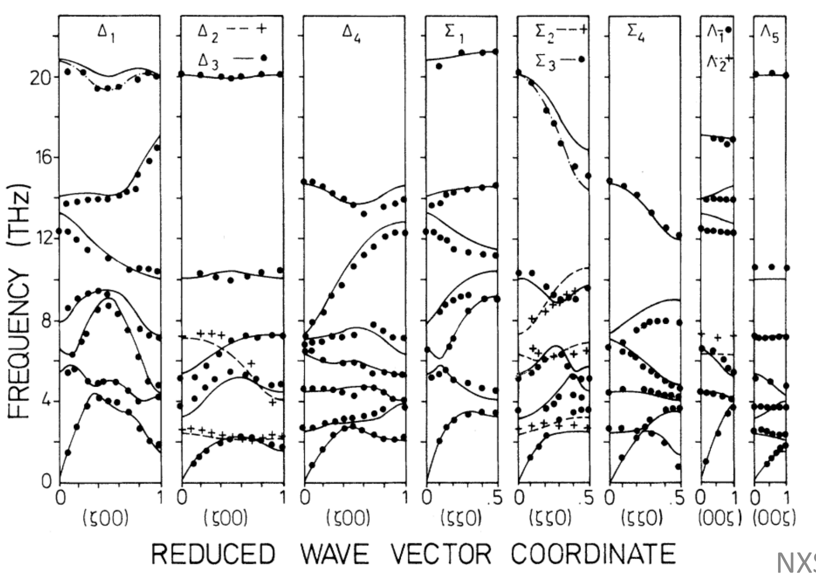


Guthoff *et al.*, *Phys. Rev. B* **47**, 2563 (1993).

More complicated structures

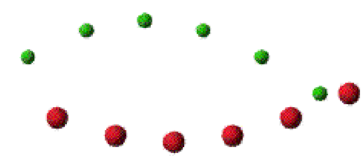


Woods, *et al., Phys. Rev*. **131**, 1025 (1963).

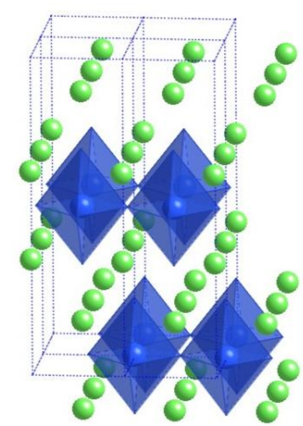


Chaplot, *et al., Phys. Rev*. B **52**, 7230(1995).

Acoustic phonon



Optical phonon



La_2CuO_4

Spin excitations

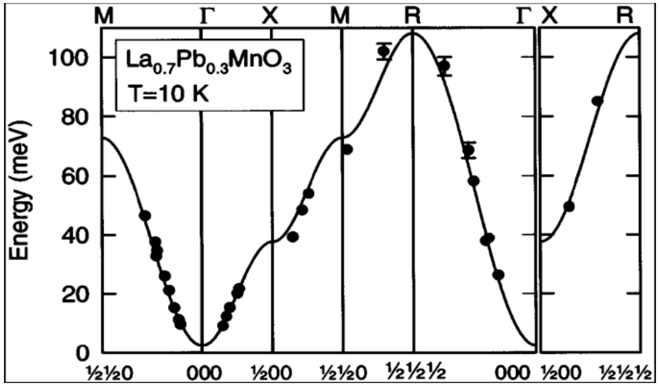
McMaster University

- **Spin excitations**
  - Spin waves in ordered magnets
  - Paramagnetic & quantum spin fluctuations
  - Crystal-field & spin-orbit excitations

- **Magnetic inelastic scattering can tell us about**
  - Exchange interactions
  - Single-ion and exchange anisotropy (determine Hamiltonian)
  - Phase transitions & critical phenomena
  - Quantum critical scaling of magnetic fluctuations
  - Other electronic energy scales (eg. CF & SO)
  - Interactions (eg. spin-phonon coupling)

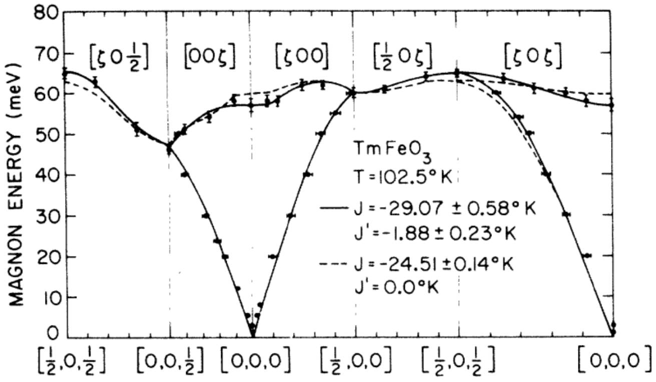
Spin waves

Ferromagnetic



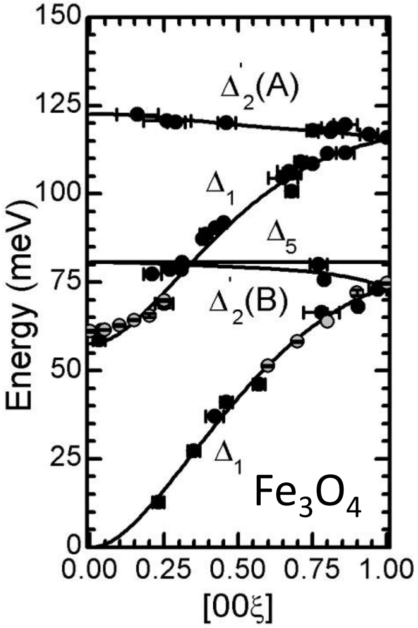
Perring *et al., Phys. Rev*. Lett. **77**, 711 (1996).

Antiferromagnetic



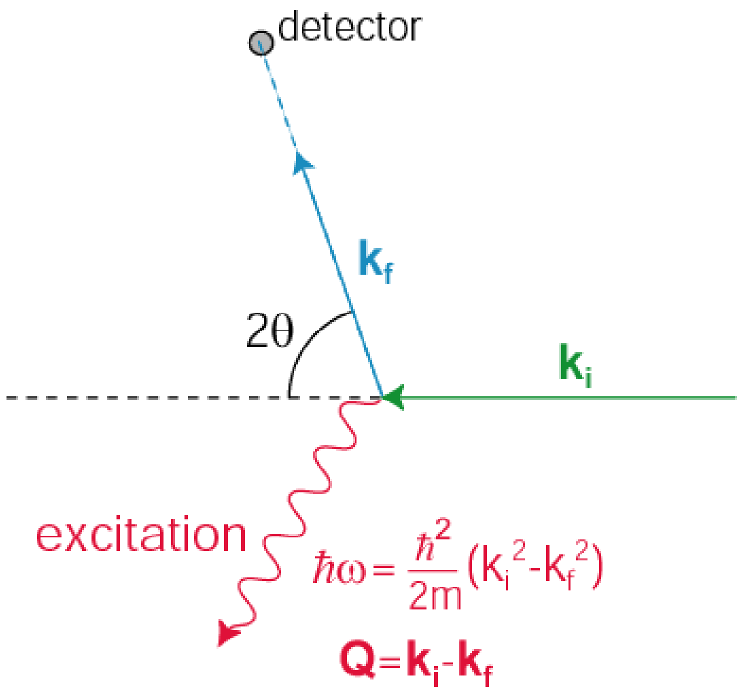
Shapiro *et al., Phys. Rev*. B **10**, 2014 (1974).

Ferrimagnetic

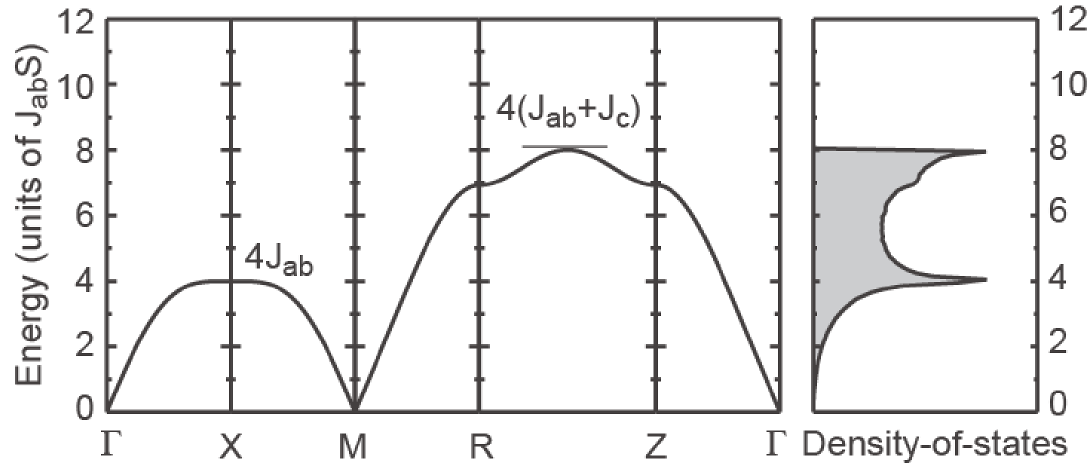


McQueeney *et al., Phys. Rev*. Lett. **99**, 246401 (2007).

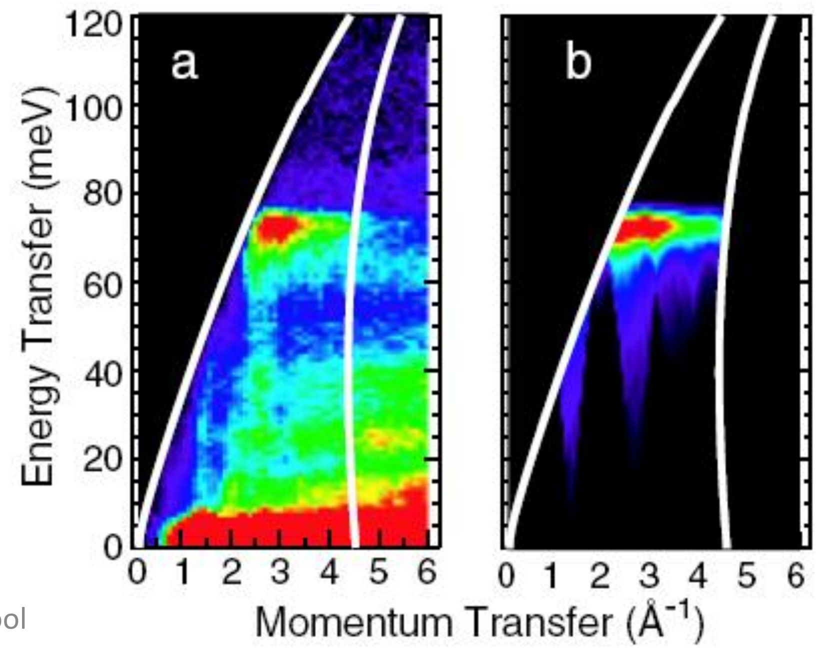
Scattering experiments



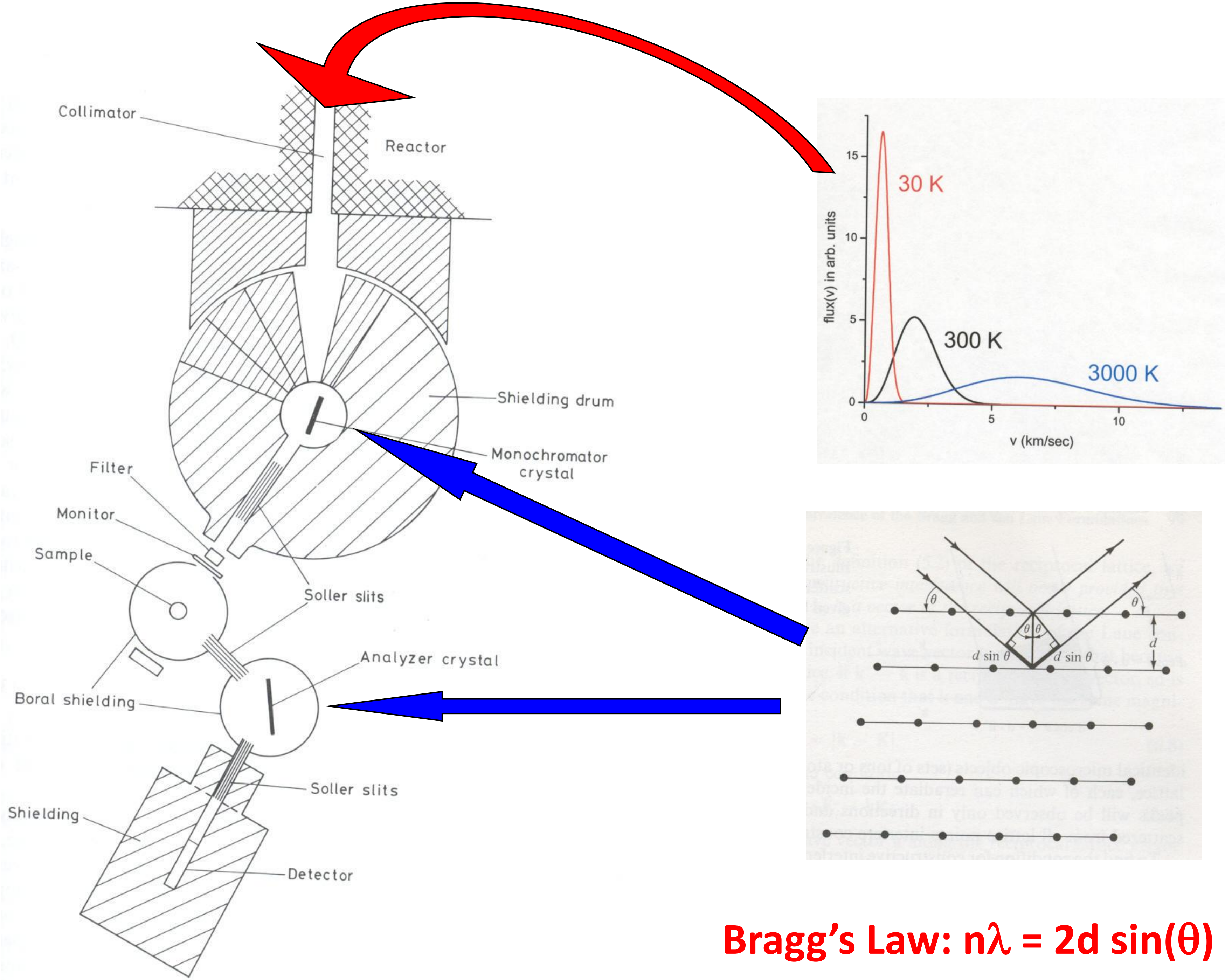
Single-crystal



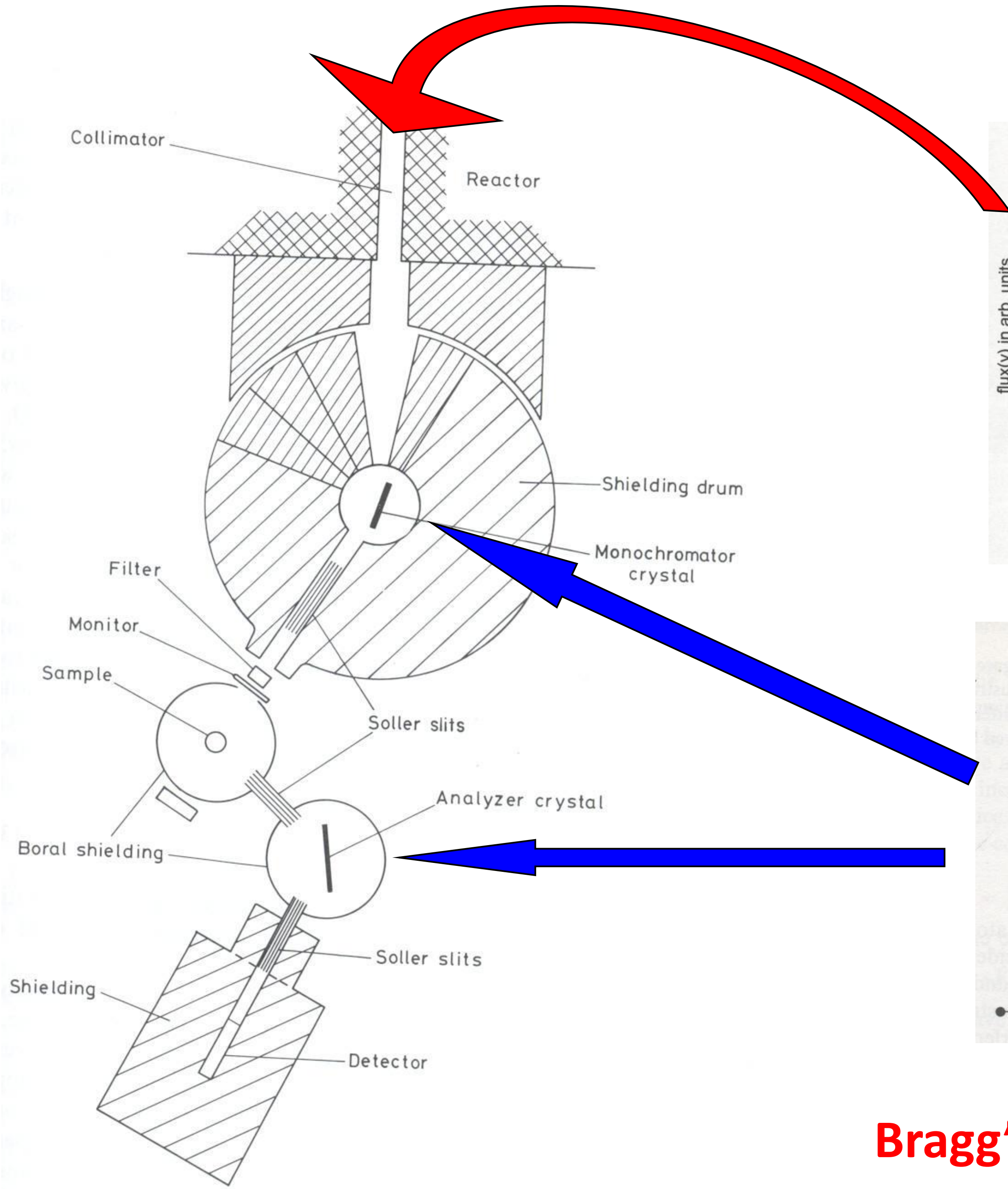
Powder $S(|\mathbf{Q}|,\omega)$

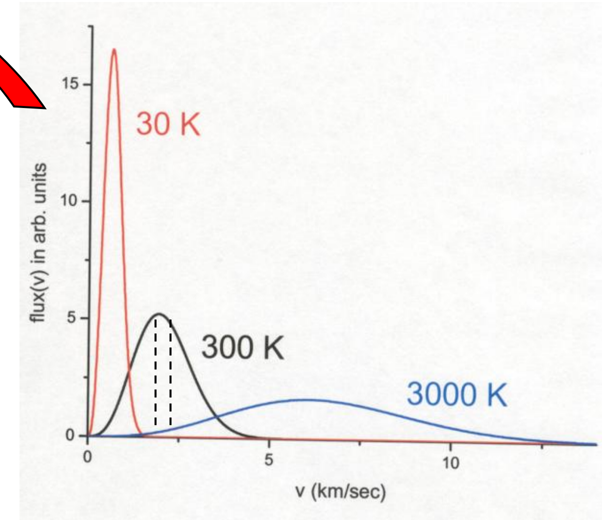


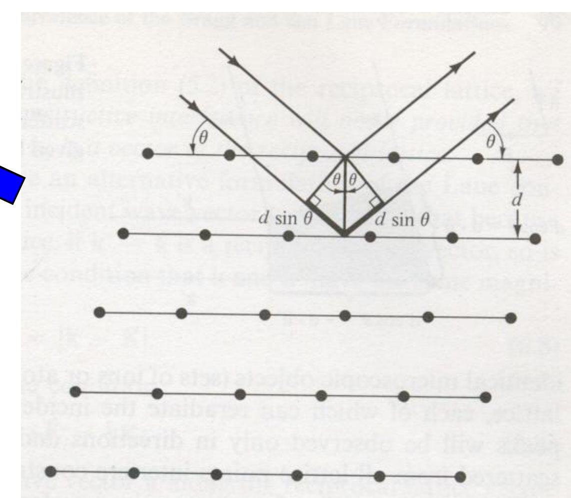
Instrument and sample (powder or single-crystal) determine how $(\mathbf{Q},\omega)$ space is sampled



Bragg's Law: nλ = 2d sin(θ)



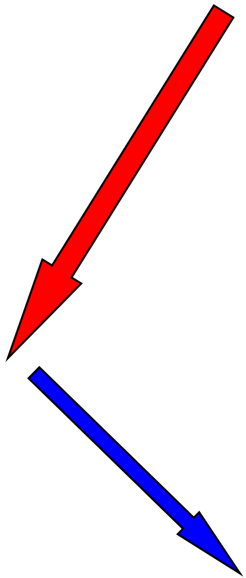




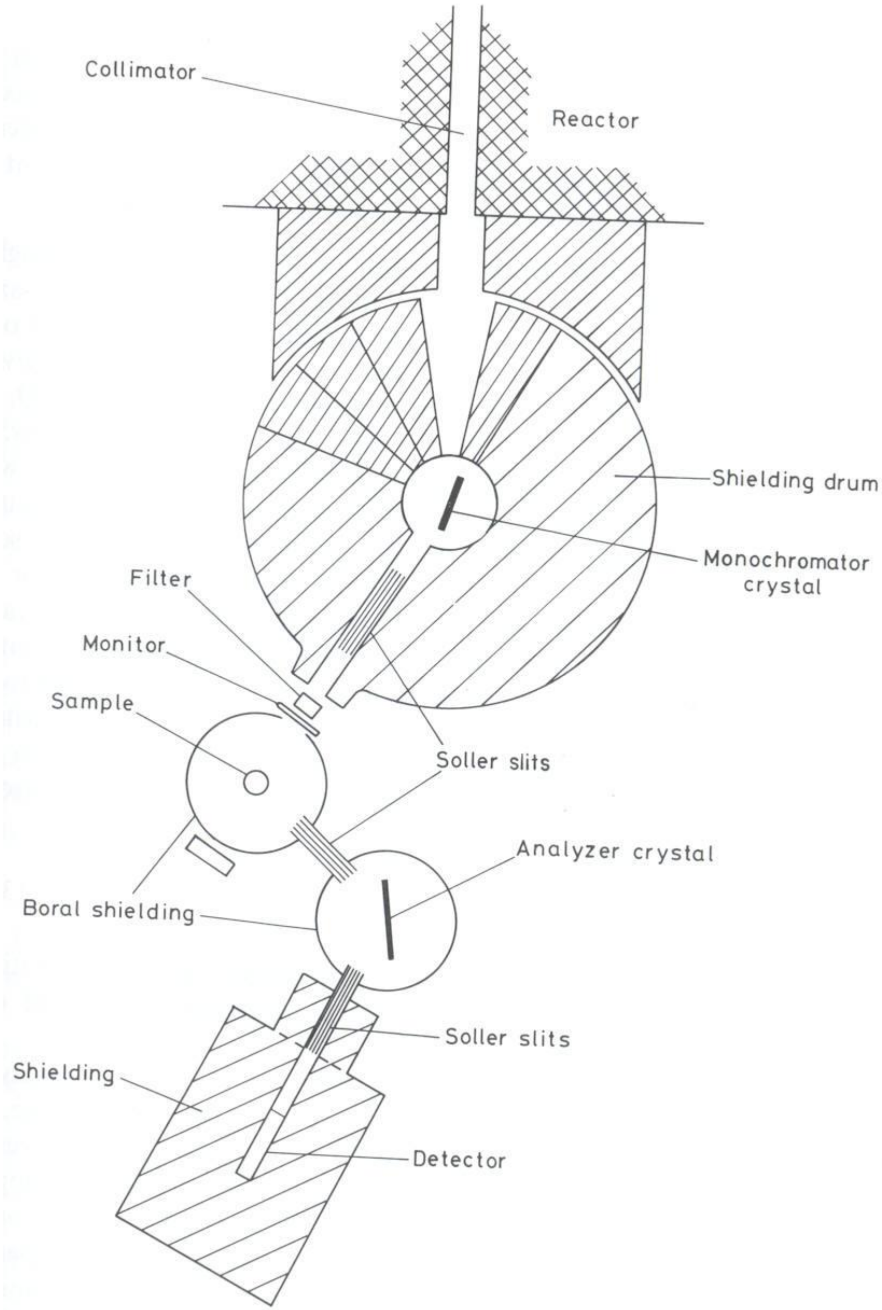
Bragg's Law: $n\lambda = 2d \sin(\theta)$

Brockhouse's Triple Axis Spectrometer

$| \mathbf{k_i} | = 2\pi / \lambda_i$

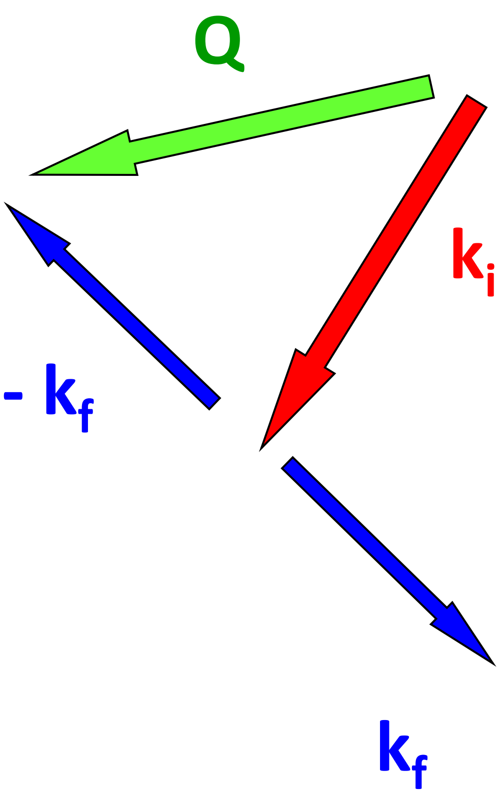


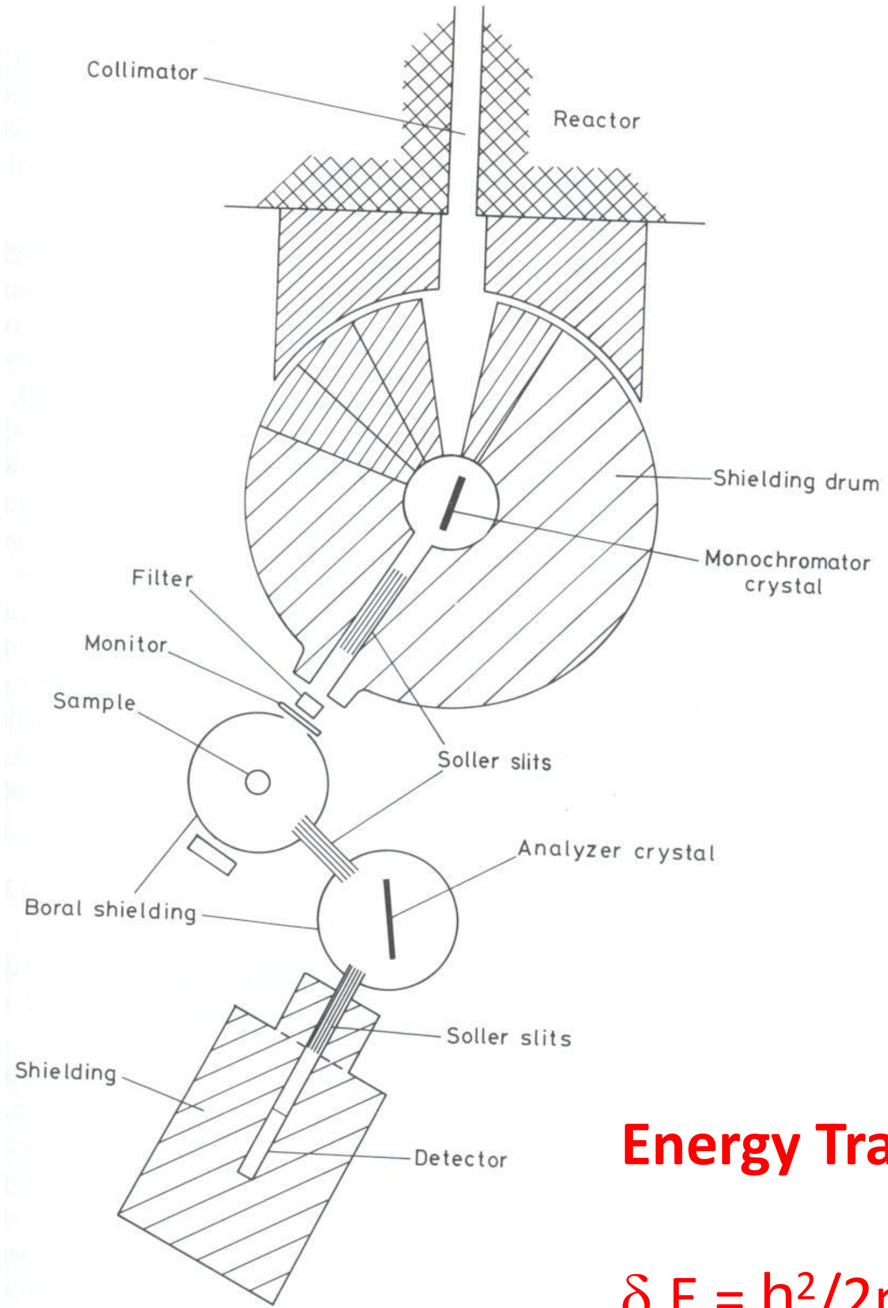
$| \mathbf{k_f} | = 2\pi / \lambda_f$



Momentum Transfer:

$Q = k_i - k_f$





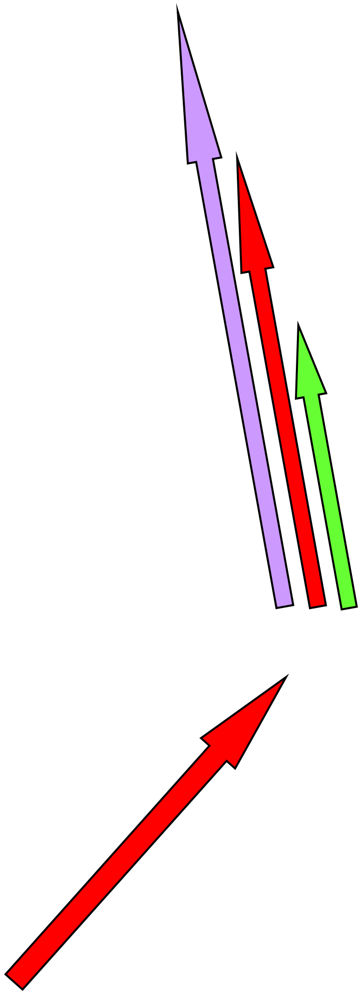
Energy Transfer:

$\delta E = h^2/2m\ (k_i^2 - k_f^2)$

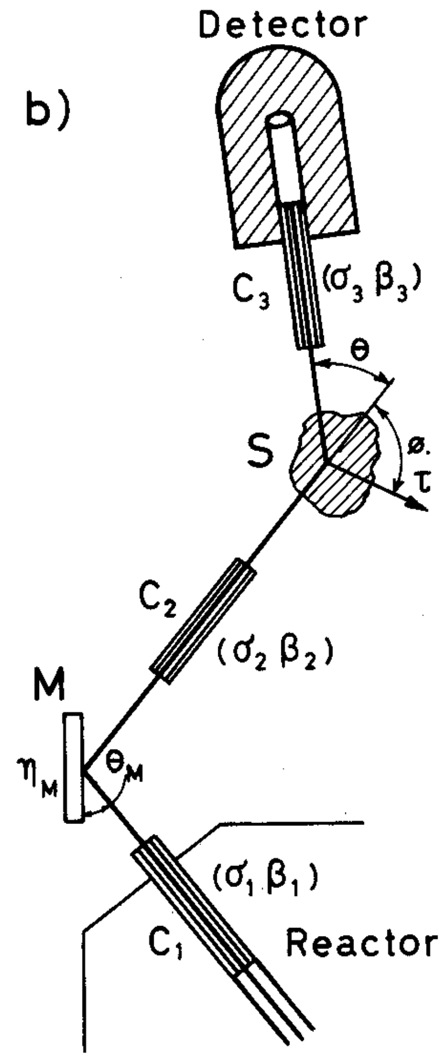
Two Axis Spectrometer:

- 3-axis with analyser removed

- Powder diffractometer
- Small angle diffractometer
- Reflectometers

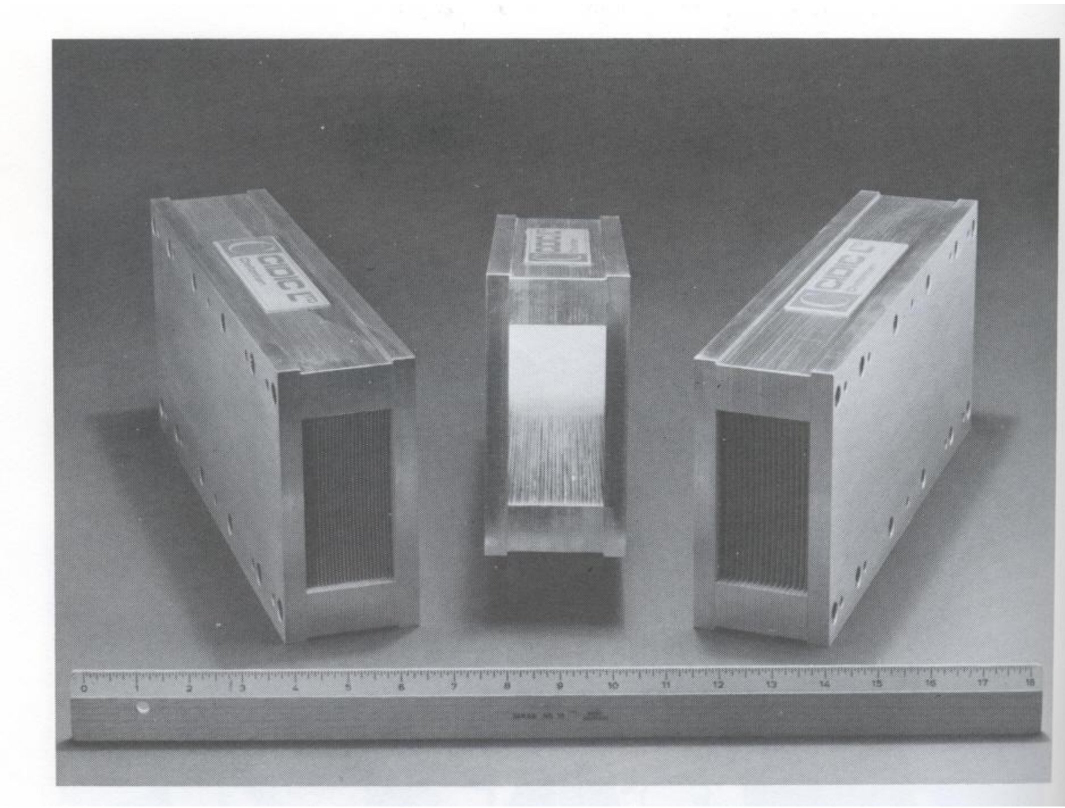


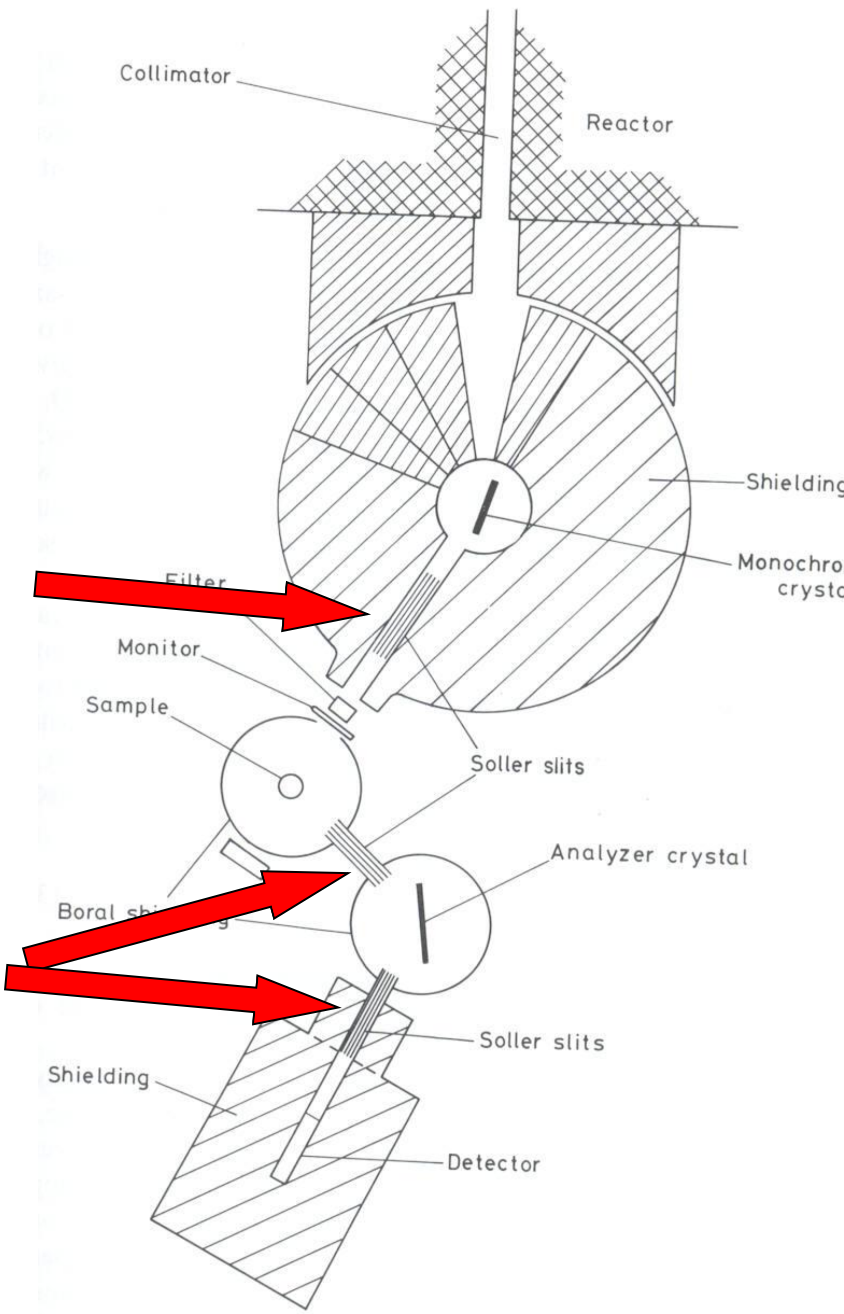
Diffractometers often employ working assumption that all scattering is *elastic.*



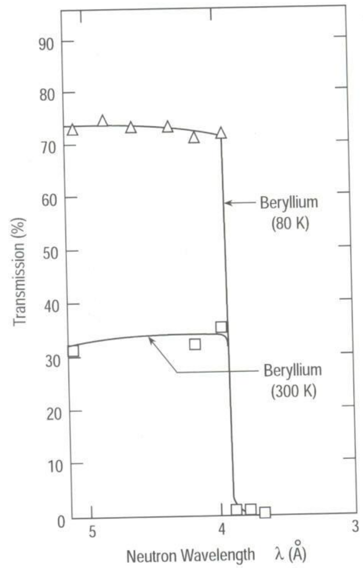
Soller Slits: Collimators

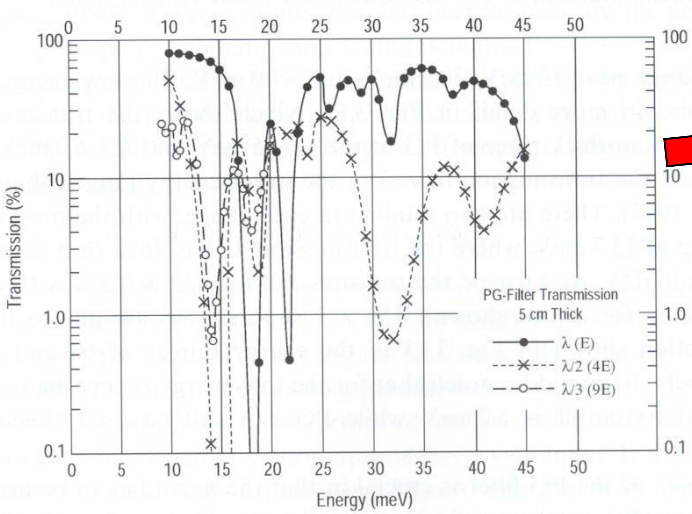
Define beam direction to +/- 0.5, 0.75 etc. degrees

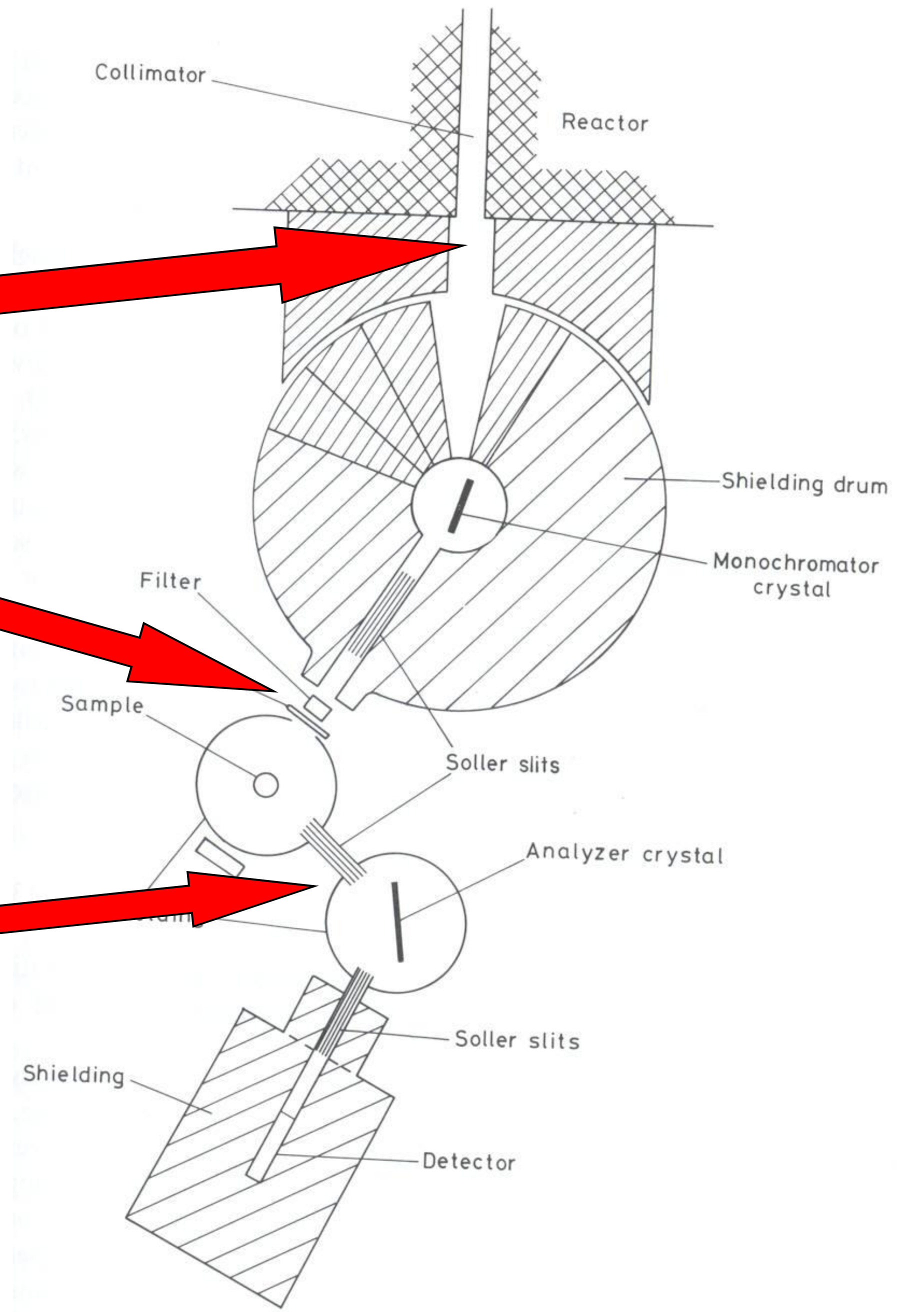




Filters:
Remove λ /n from incident or scattered beam, or both

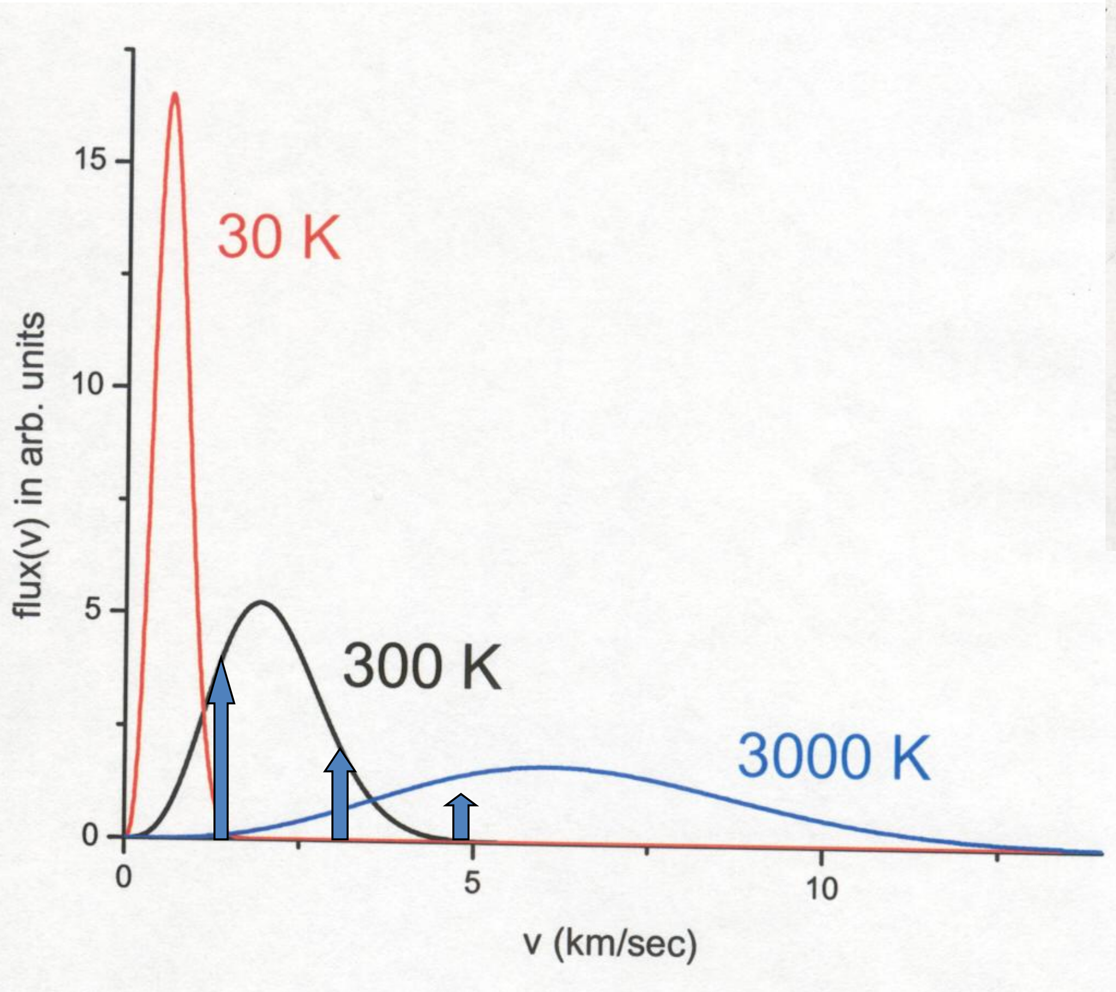


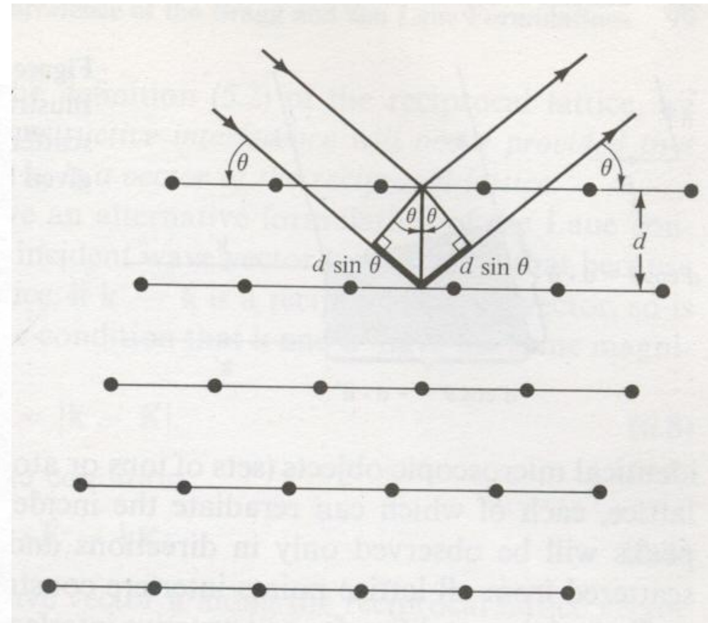




Single crystal monochromators:

Bragg reflection and harmonic contamination

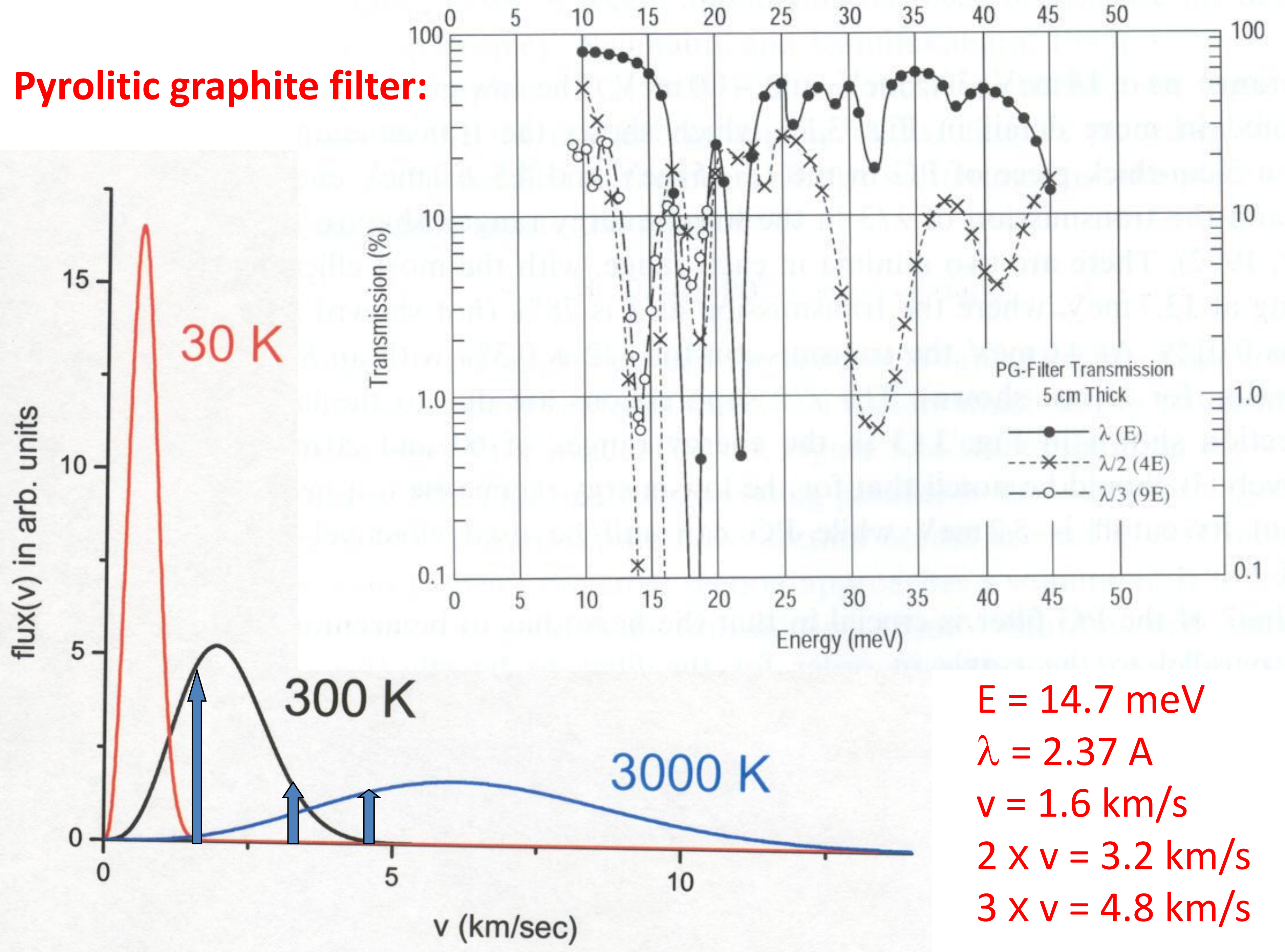




$n\lambda = 2d\sin(\theta)$

Get: λ , $\lambda/2$, $\lambda/3$, etc.

Pyrolitic graphite filter:



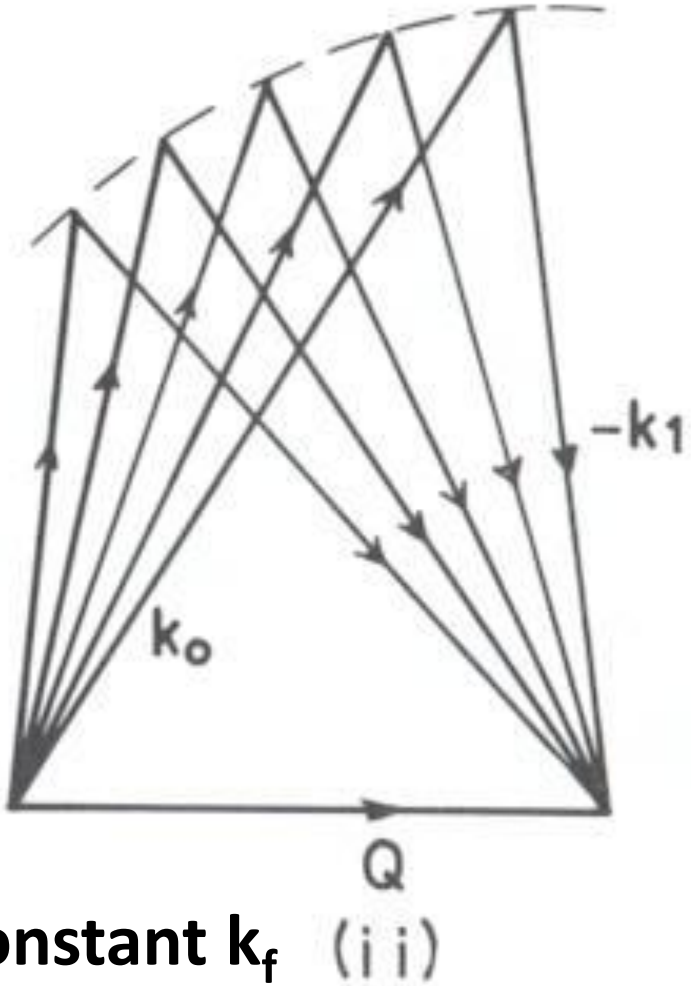
E = 14.7 meV

λ = 2.37 A

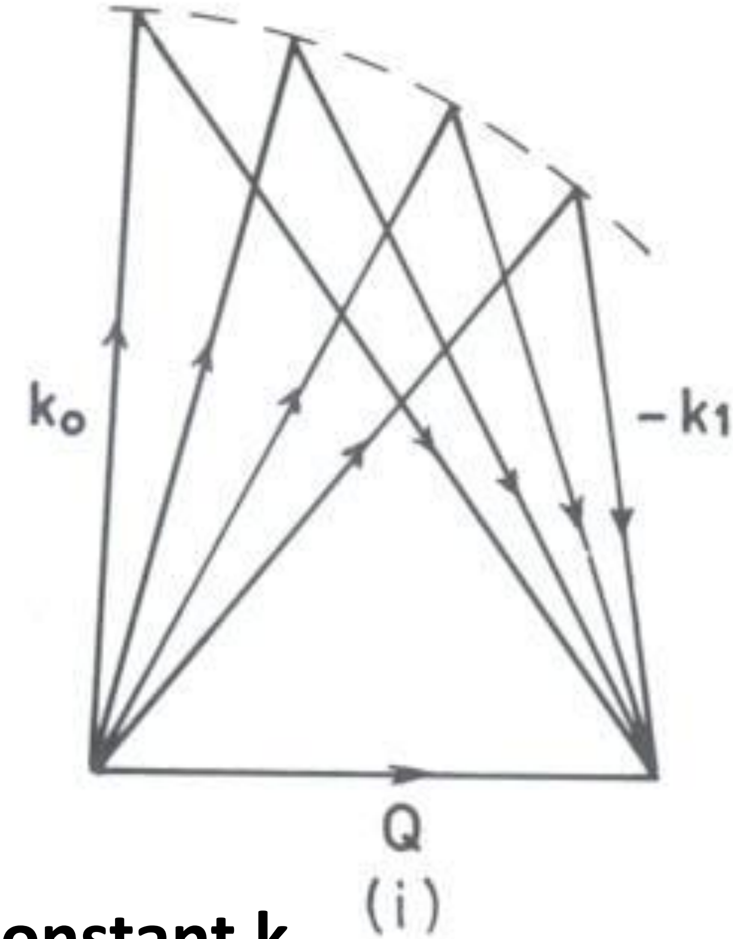
v = 1.6 km/s

2 x v = 3.2 km/s

3 x v = 4.8 km/s



Constant k_f



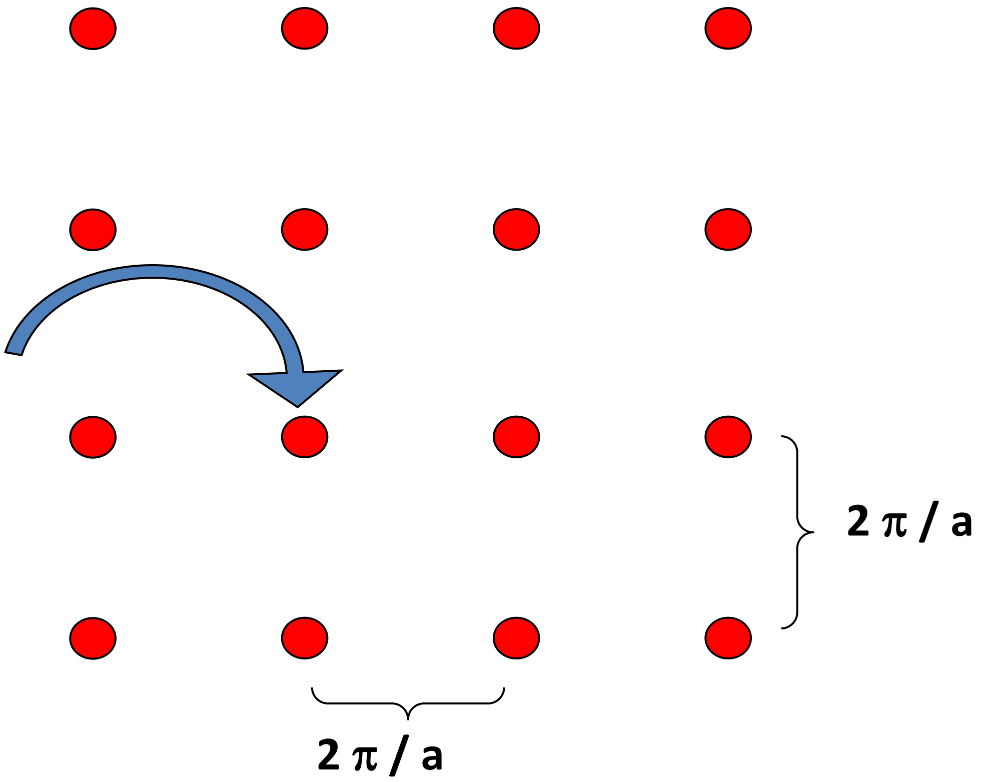
Constant k_i

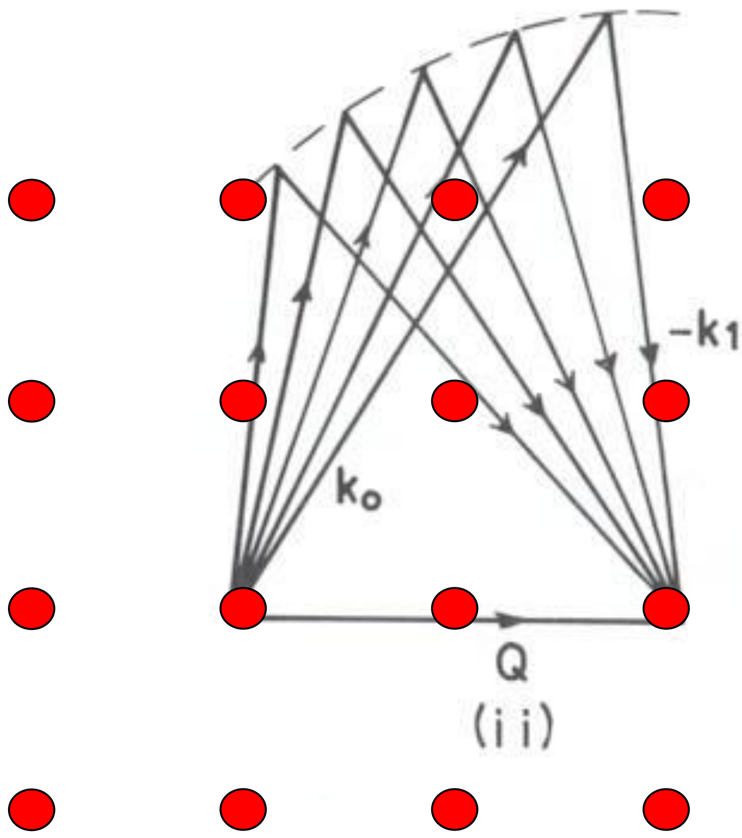
Two different ways of performing constant-Q scans

Mapping Momentum – Energy (Q-E) space

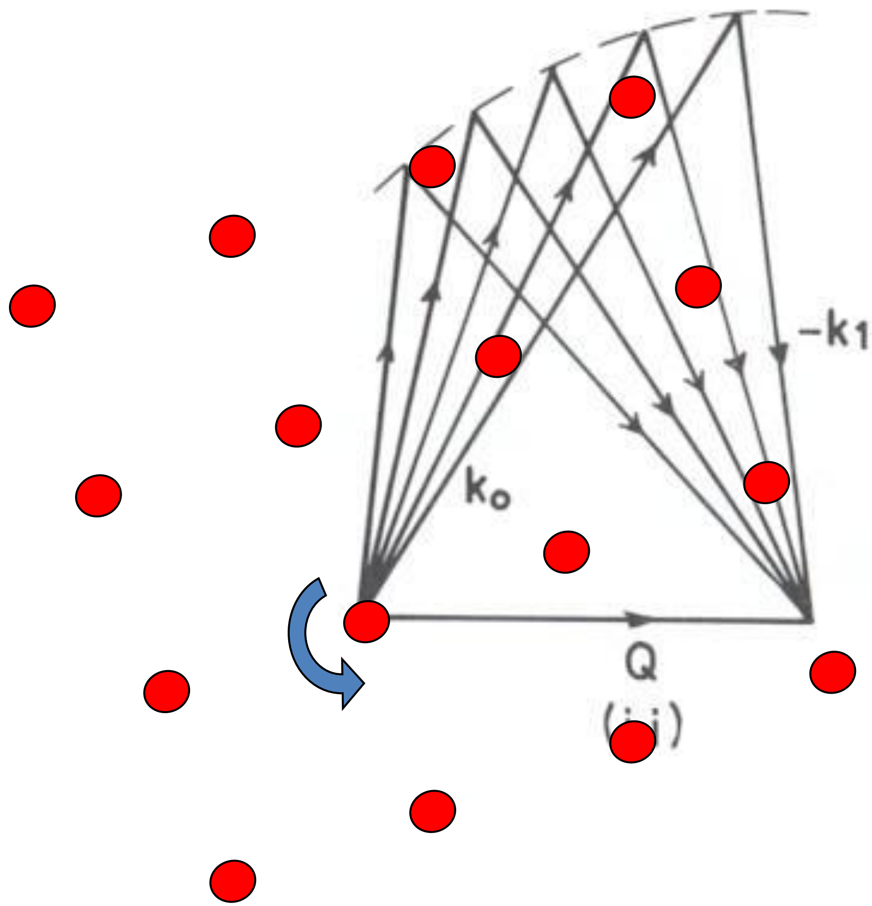
Origin of reciprocal space;

Remains fixed for any sample rotation

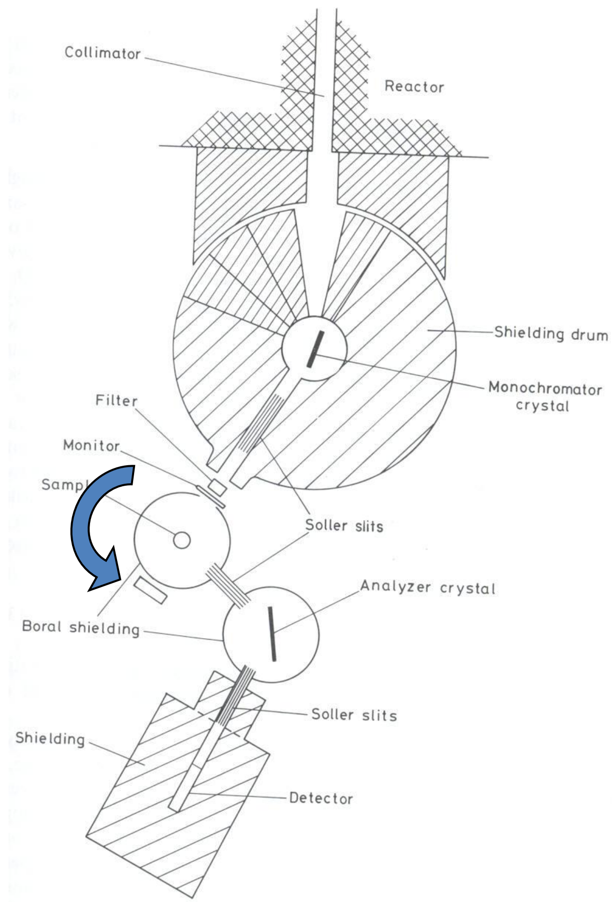




-k1
k0
Q
(ii)

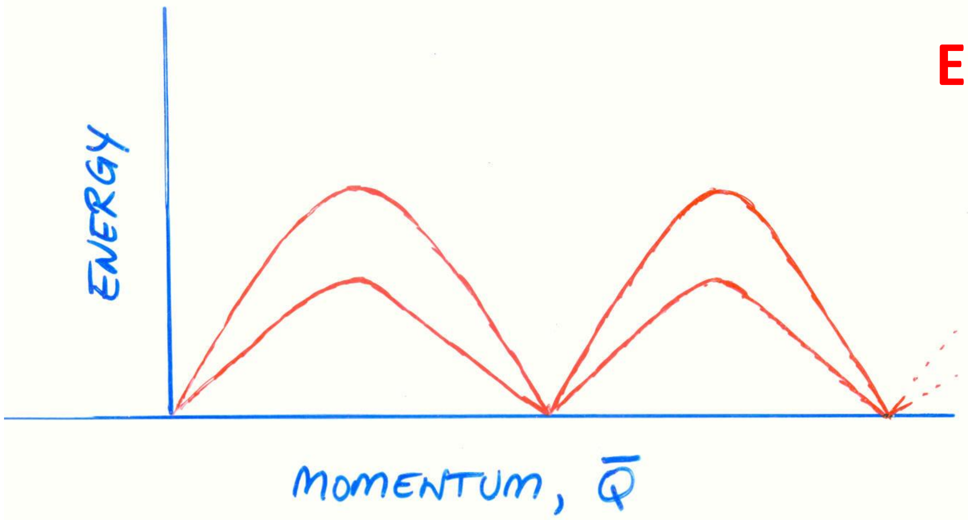


-k1
ko
Q

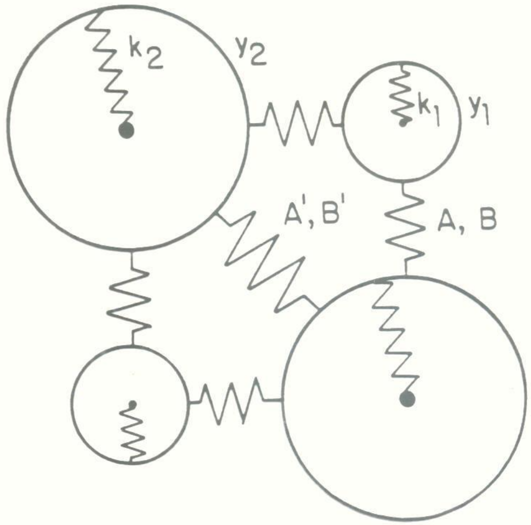


Collimator
Reactor
Shielding drum
Monochromator crystal
Filter
Monitor
Soller slits
Analyzer crystal
Boral shielding
Soller slits
Shielding
Detector

Elementary Excitations in Solids

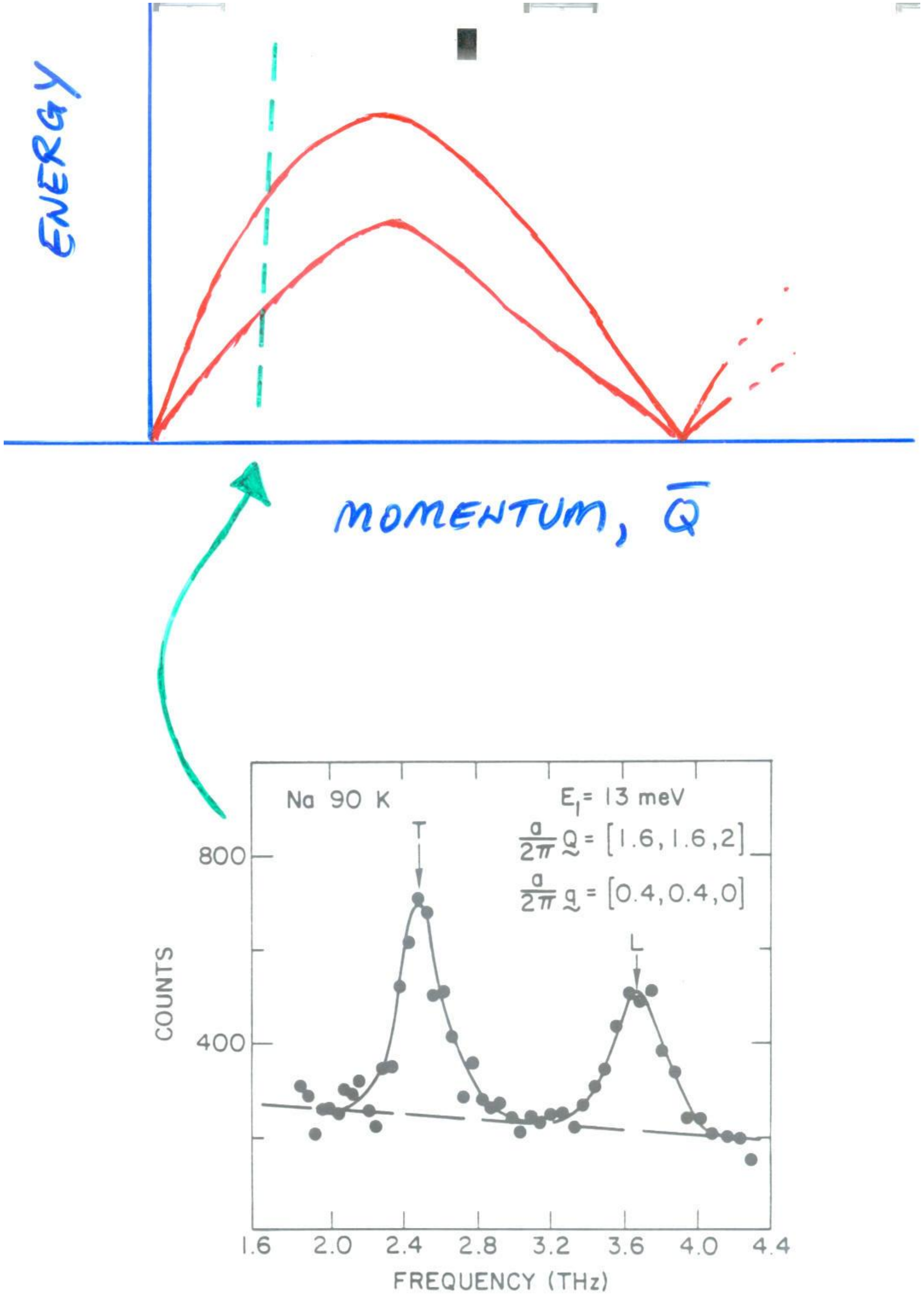


- Lattice Vibrations (Phonons)
- Spin Fluctuations (Magnons)



Energy vs Momentum

- Forces which bind atoms together in solids



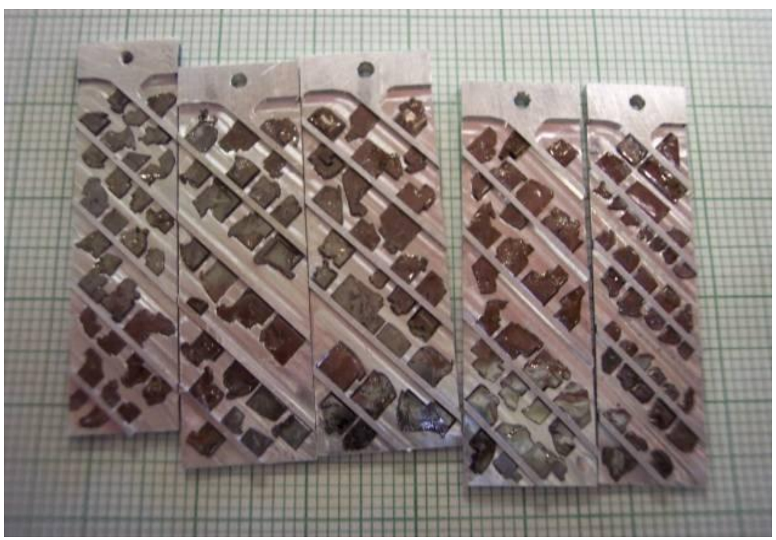
Constant Q, Constant E
3-axis technique allow us to
Put Q-Energy space on a grid,
And scan through as we wish

Map out elementary excitations
In Q-energy space (dispersion
Surface)

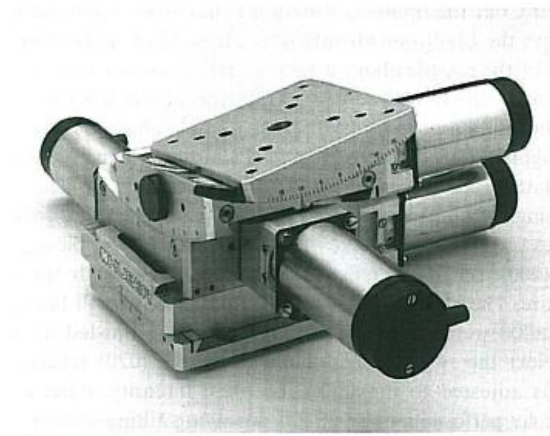
Samples

- **Samples need to be BIG**
  - ~ gram or cc
  - Counting times are long (mins/pt)

- **Sample rotation**
- **Sample tilt**

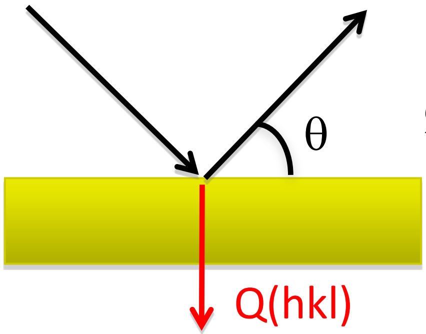


Co-aligned $CaFe_2As_2$ crystals

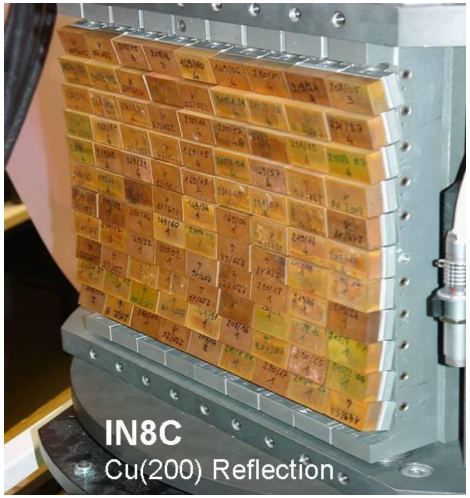


Monochromators

- **Selects the incident wavevector**



$$Q(hkl) = \frac{2\pi}{d(hkl)} = 2k_i \sin\theta$$

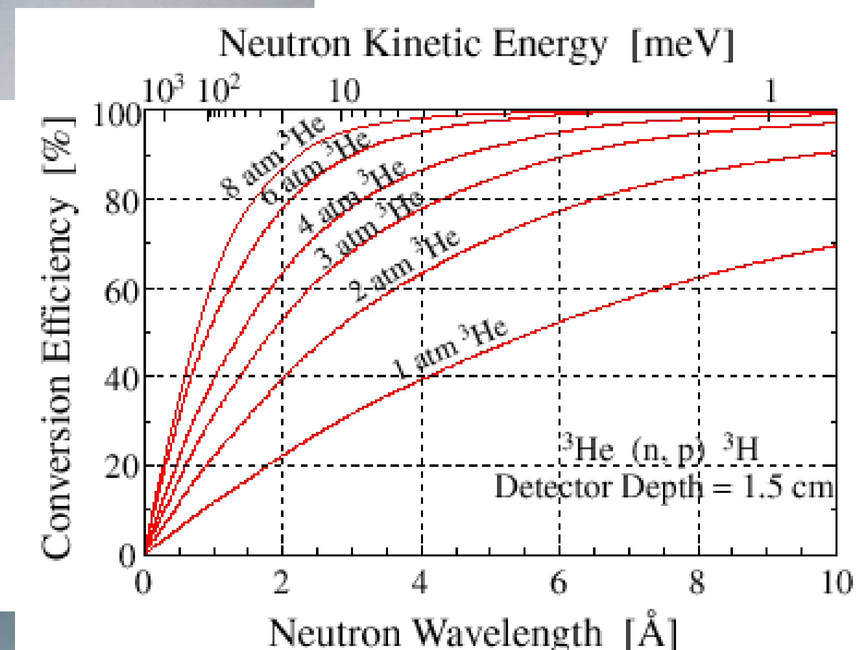


- Reflectivity
- focusing
- high-order contamination eg. $\lambda/2$ PG(004)

Mono	d(hkl)	uses
PG(002)	3.353	General
Be(002)	1.790	High k_i
Si(111)	3.135	No $\lambda/2$

Detectors

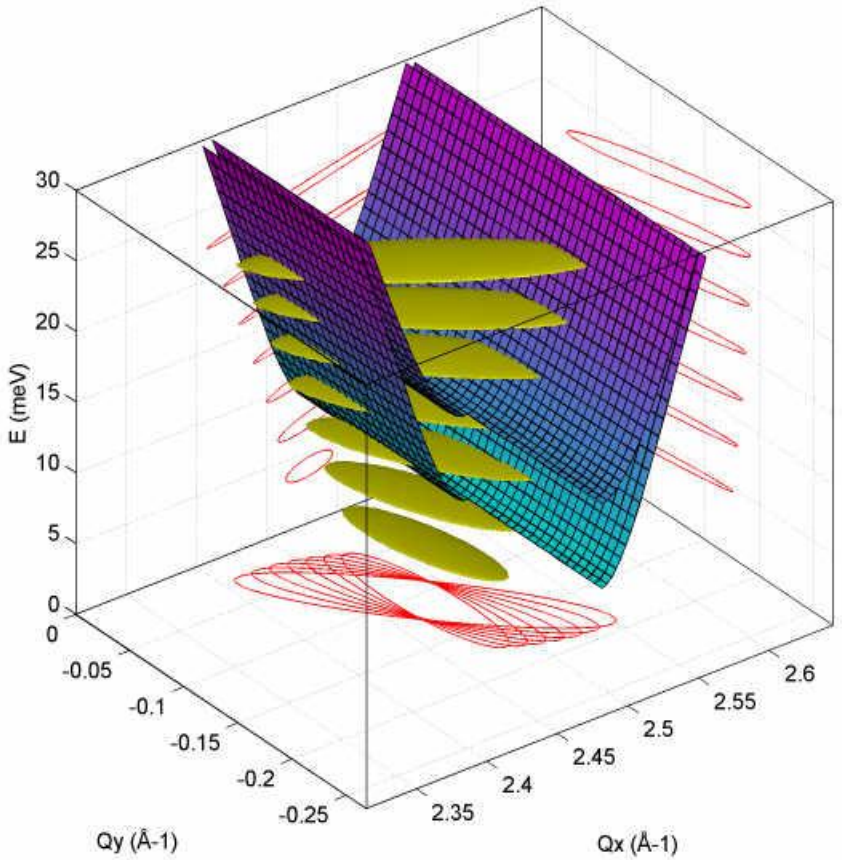
- **Gas Detectors**
- n + $^{3}He \rightarrow {}^{3}H$ + p + 0.764 MeV
- Ionization of gas
- e^- drift to high voltage anode
- High efficiency



- **Beam monitors**
- Low efficiency detectors for measuring beam flux

Resolution

- **Resolution ellipsoid**
  - Beam divergences
  - Collimations/distances
  - Crystal mosaics/sizes/angles

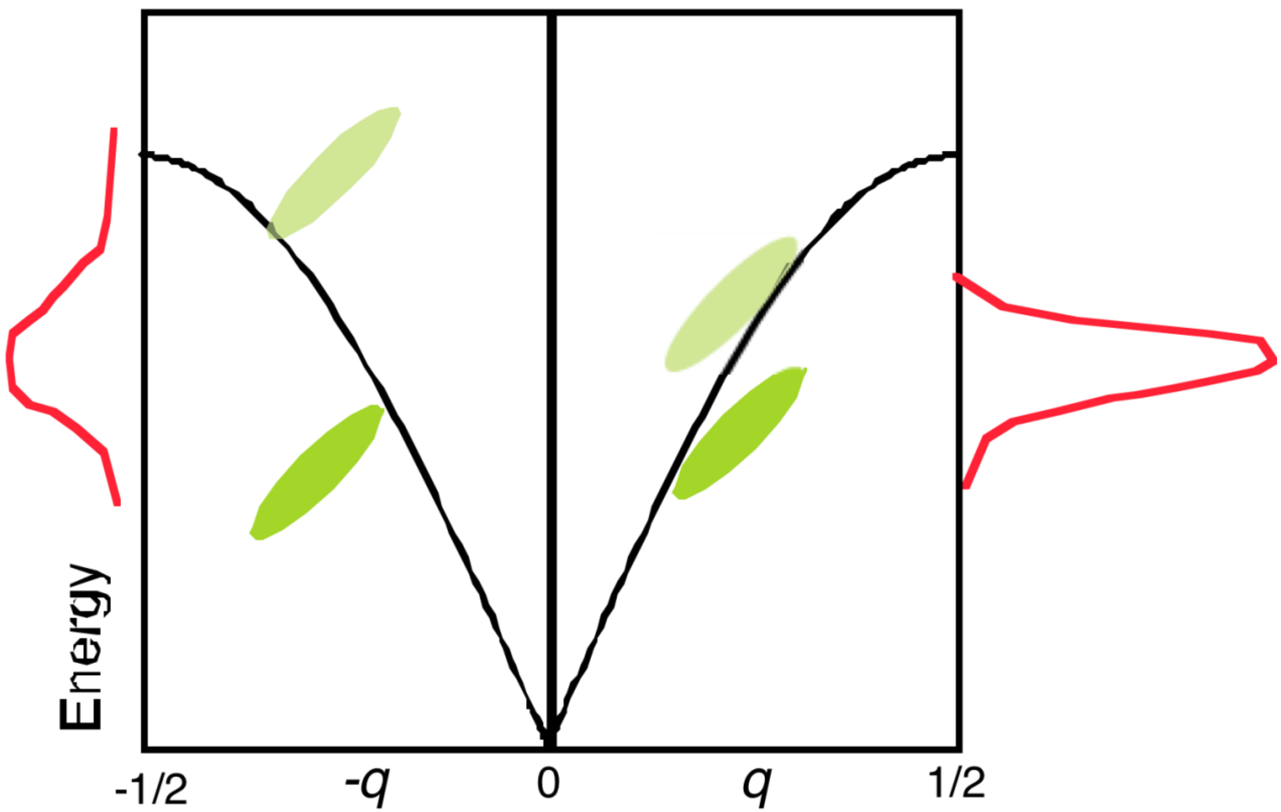


- **Resolution convolutions**

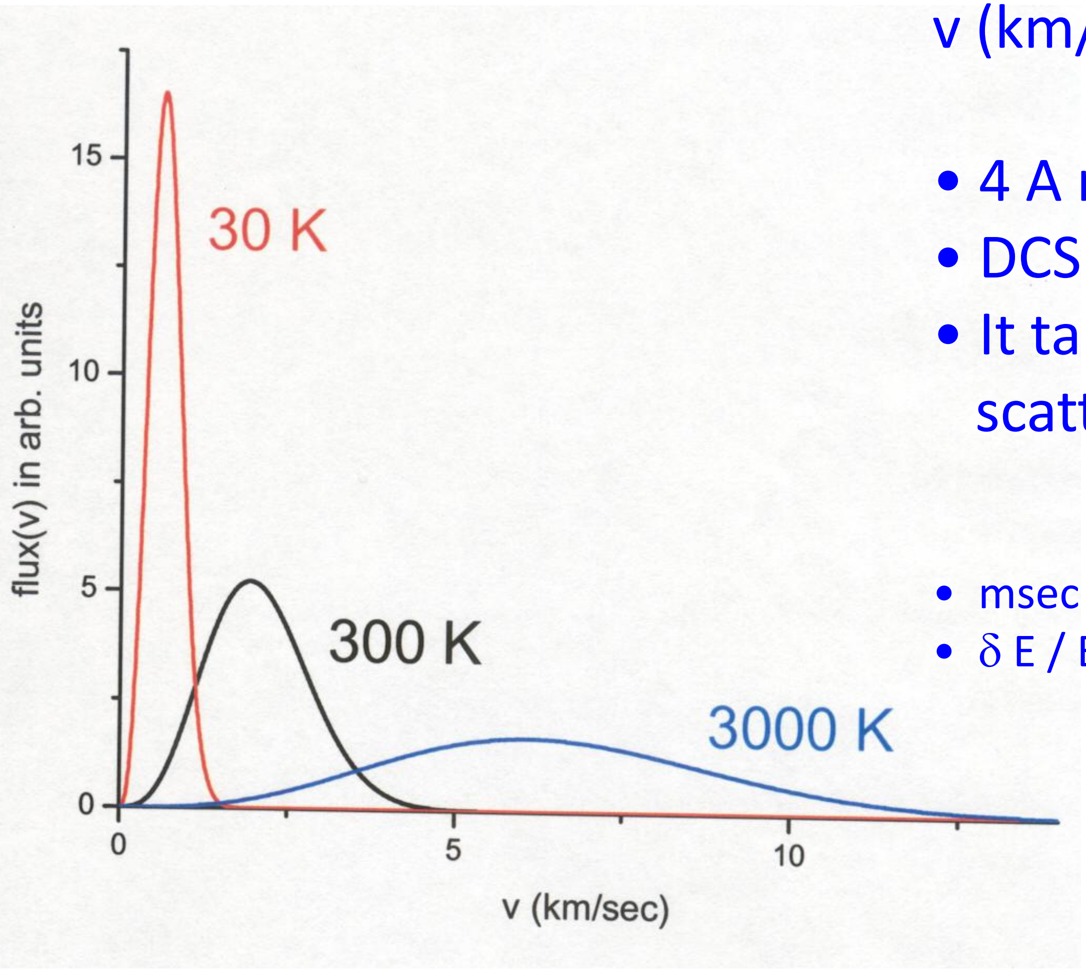
$$I(\mathbf{Q}_0,\omega_0) = \int S(\mathbf{Q}_0,\omega_0)R(\mathbf{Q}-\mathbf{Q}_0,\omega-\omega_0)d\mathbf{Q}d\omega$$

Resolution focusing

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



Neutrons have *mass*
so higher energy means faster – lower energy means slower



v (km/sec) = 3.96 / λ (A)

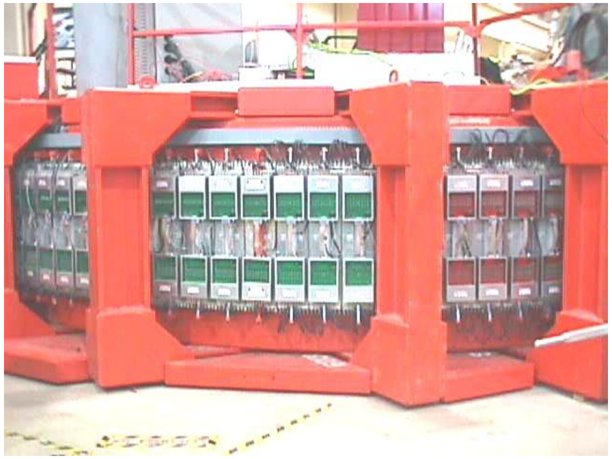
- 4 A neutrons move at ~ 1 km/sec
- DCS: 4 m from sample to detector
- It takes 4 msec for elastically scattered 4 A neutrons to travel 4 m

- msec timing of neutrons is easy
- δ E / E ~ 1-3 % - very good !

We can measure a neutron's energy, wavelength by measuring its *speed*

Time-of-flight methods

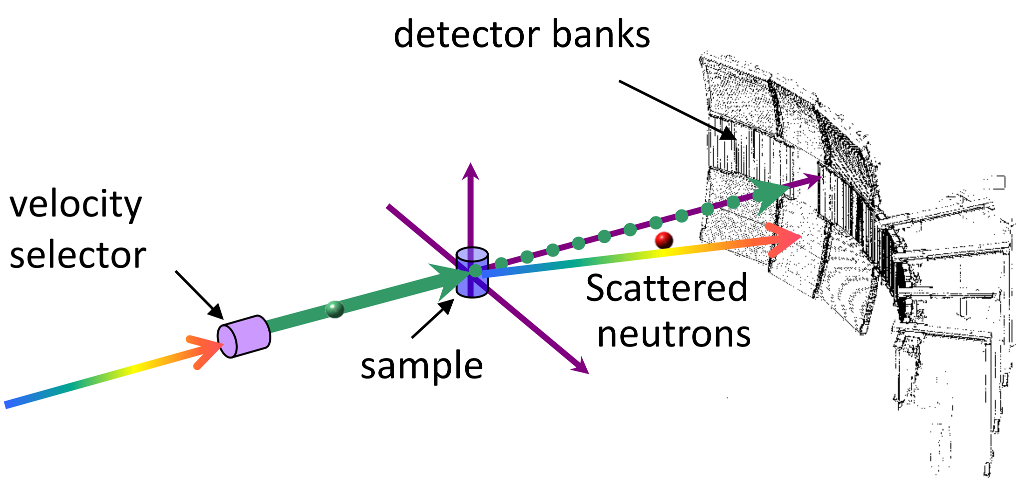
Spallation neutron source

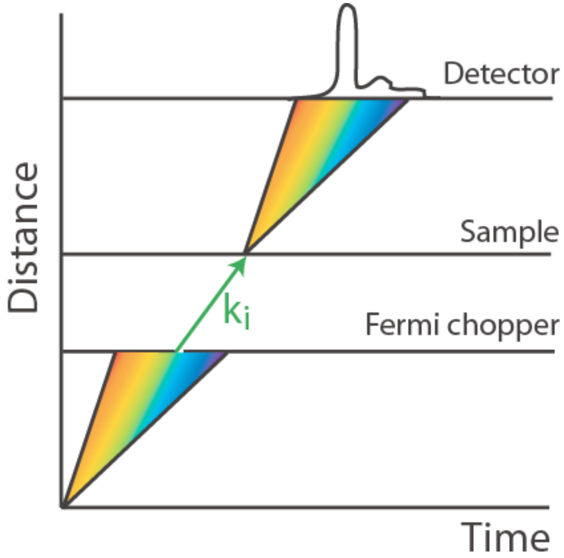


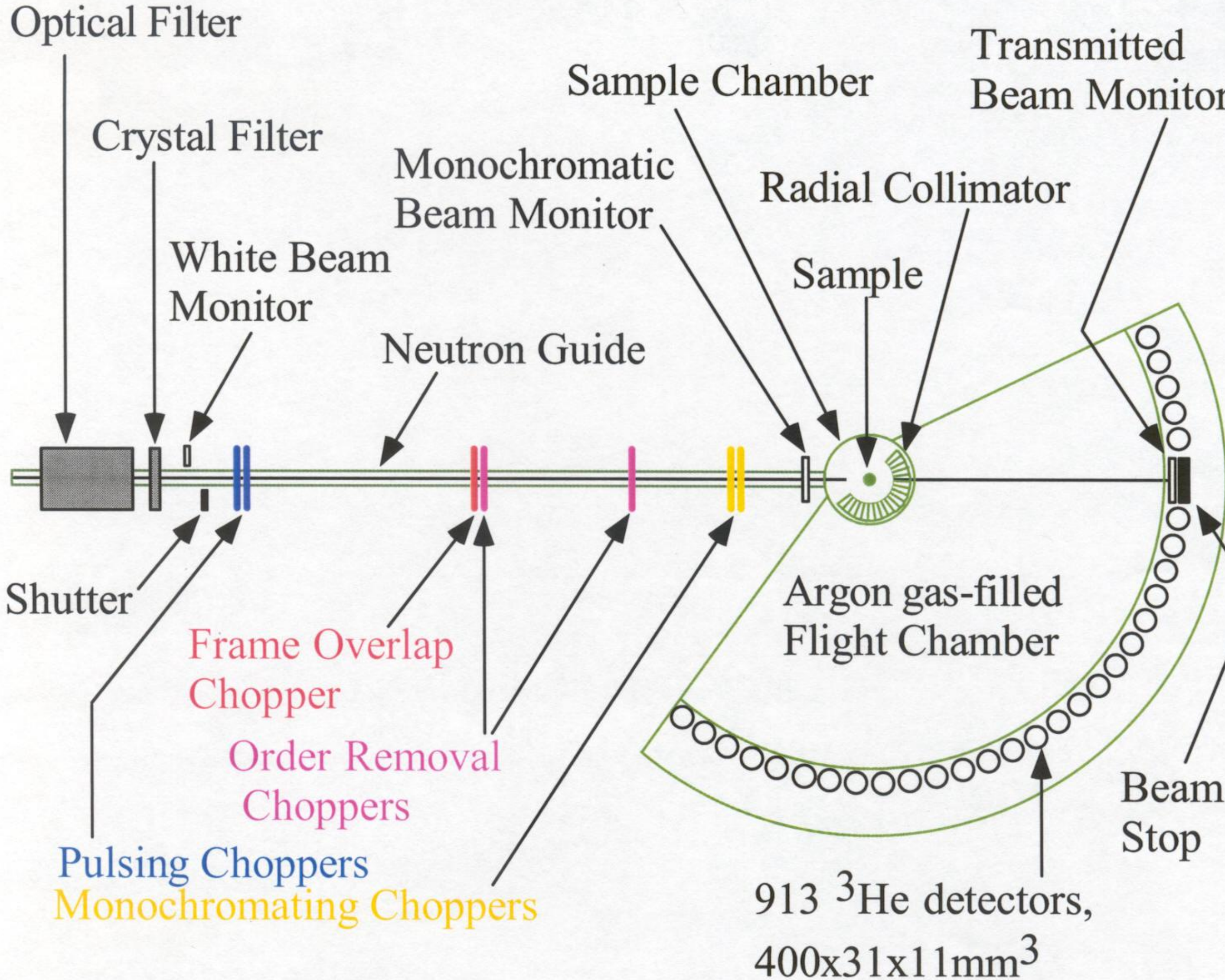
Pharos – Lujan Center

- Effectively utilizes time structure of pulsed neutron groups

$$t = \frac{d}{v} = \left(\frac{m}{h} d\right)\lambda$$



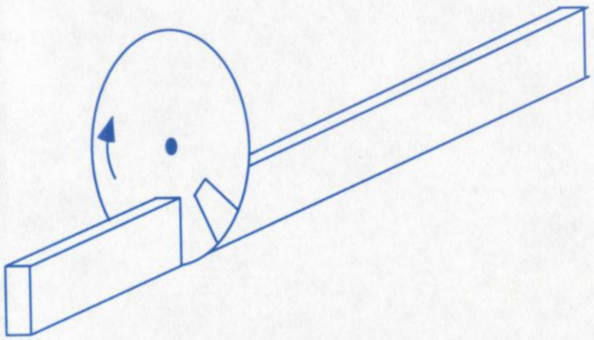


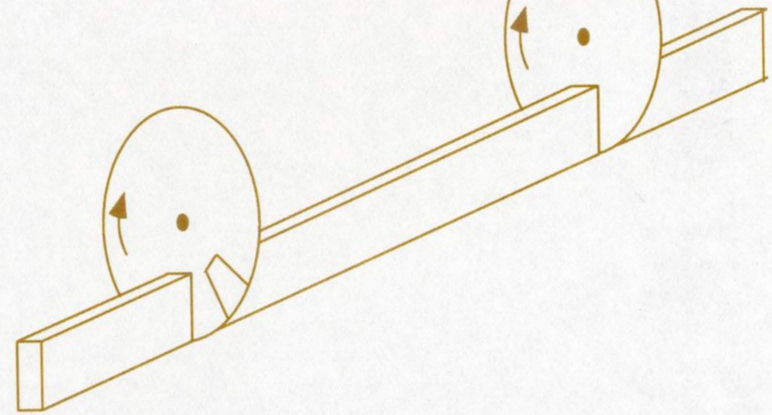


Optical Filter
Crystal Filter
White Beam Monitor
Neutron Guide
Monochromatic Beam Monitor
Sample Chamber
Transmitted Beam Monitor
Radial Collimator
Sample
Shutter
Frame Overlap Chopper
Order Removal Choppers
Pulsing Choppers
Monochromating Choppers
Argon gas-filled Flight Chamber
Beam Stop
913 ^{3}He detectors,
400x31x11mm^3

A single (disk) chopper pulses the neutron beam.

A second chopper selects neutrons within a narrow range of speeds.

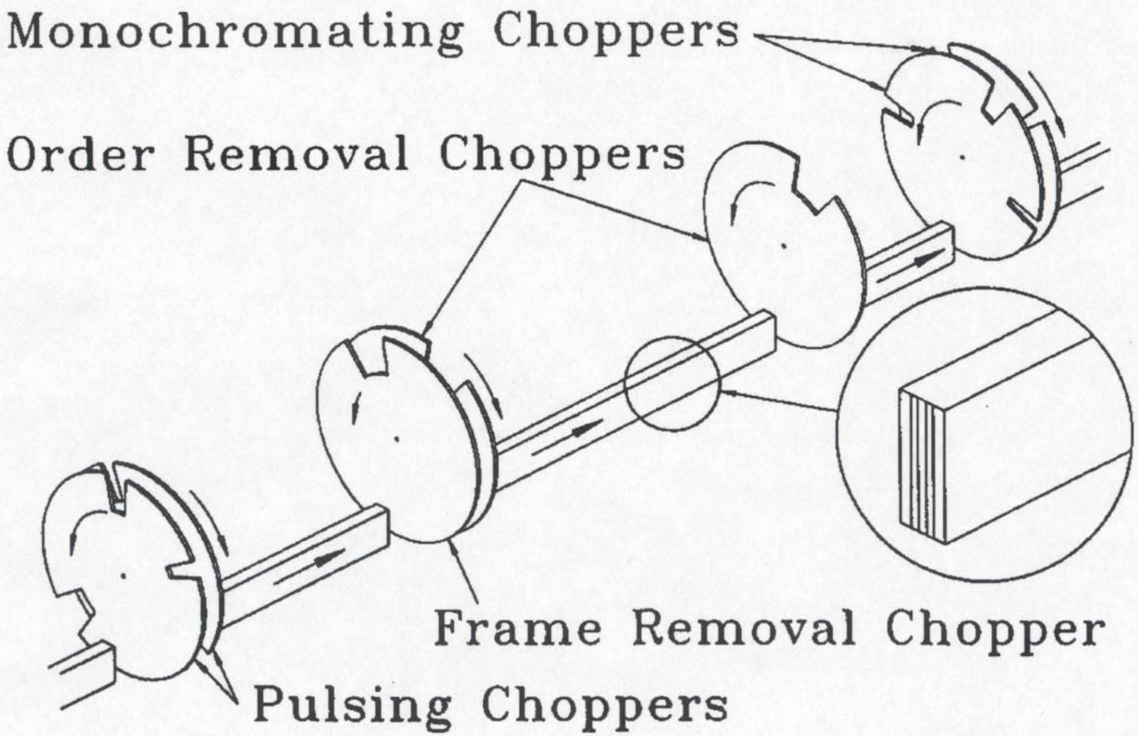




Counter-rotating choppers (close together), with speed ◆, behave like single choppers with speed 2◆. They can also permit a choice of pulse widths.

Additional choppers remove "contaminant" wavelengths and reduce the pulse frequency at the sample position.

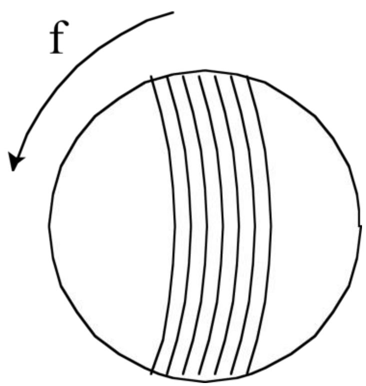
The DCS has seven choppers, 4 of which have 3 "slots"



Disk 4B

Fermi Choppers

- Body radius ~ 5 cm
- Curved absorbing slats
  - B or Gd coated
  - ~mm slit size
- f = 600 Hz (max)
- Acts like shutter, $\Delta t \sim \mu s$



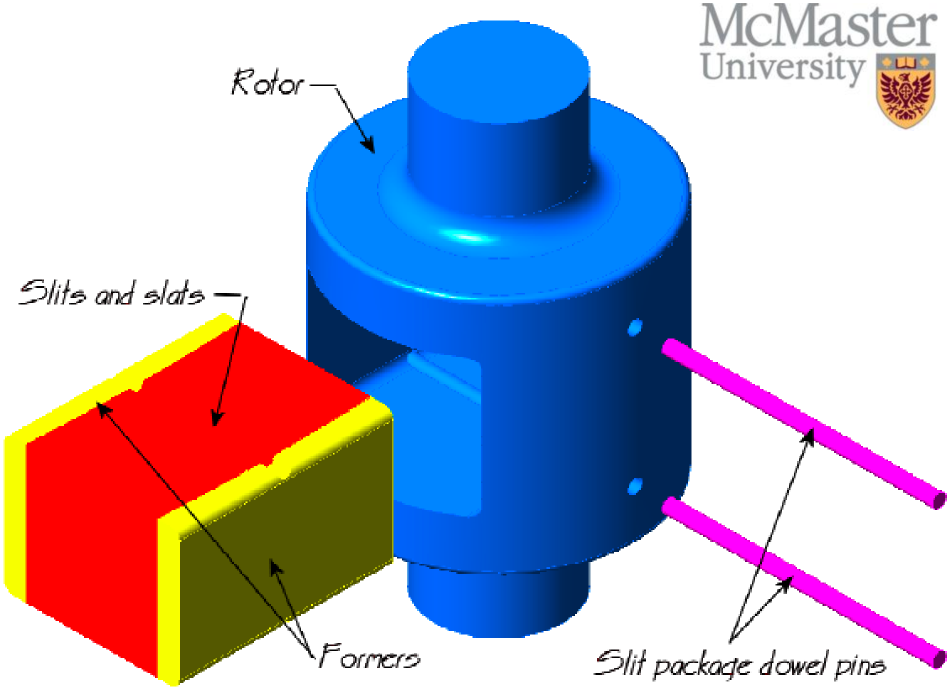
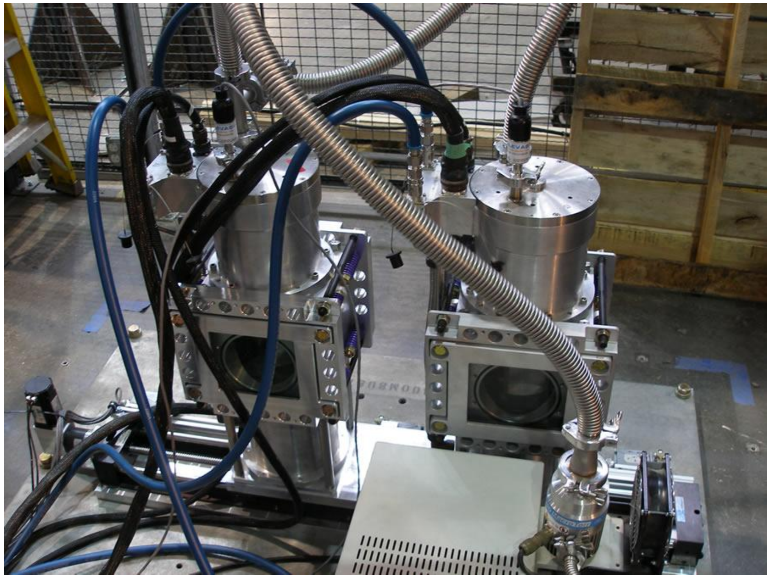
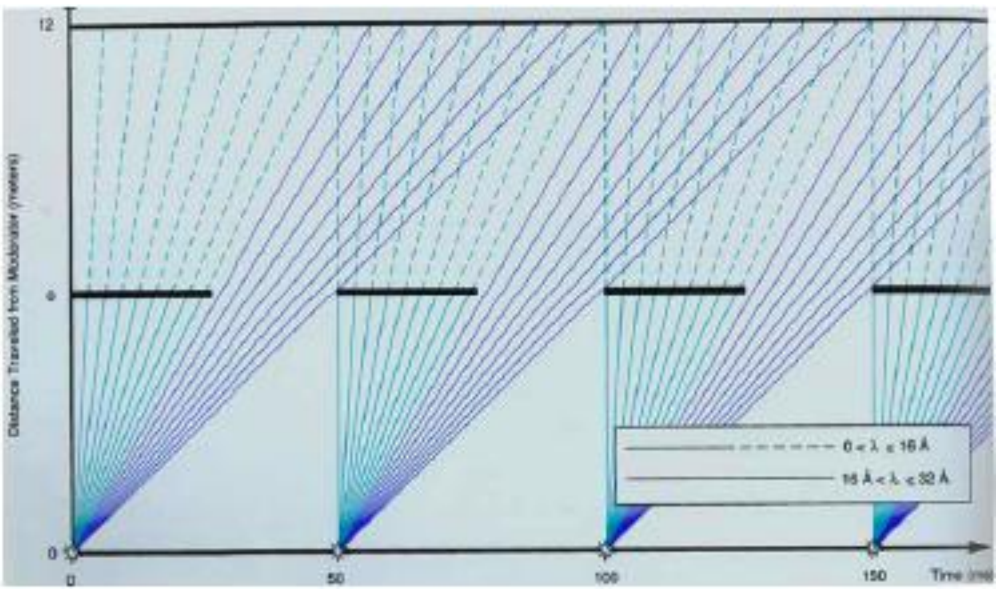


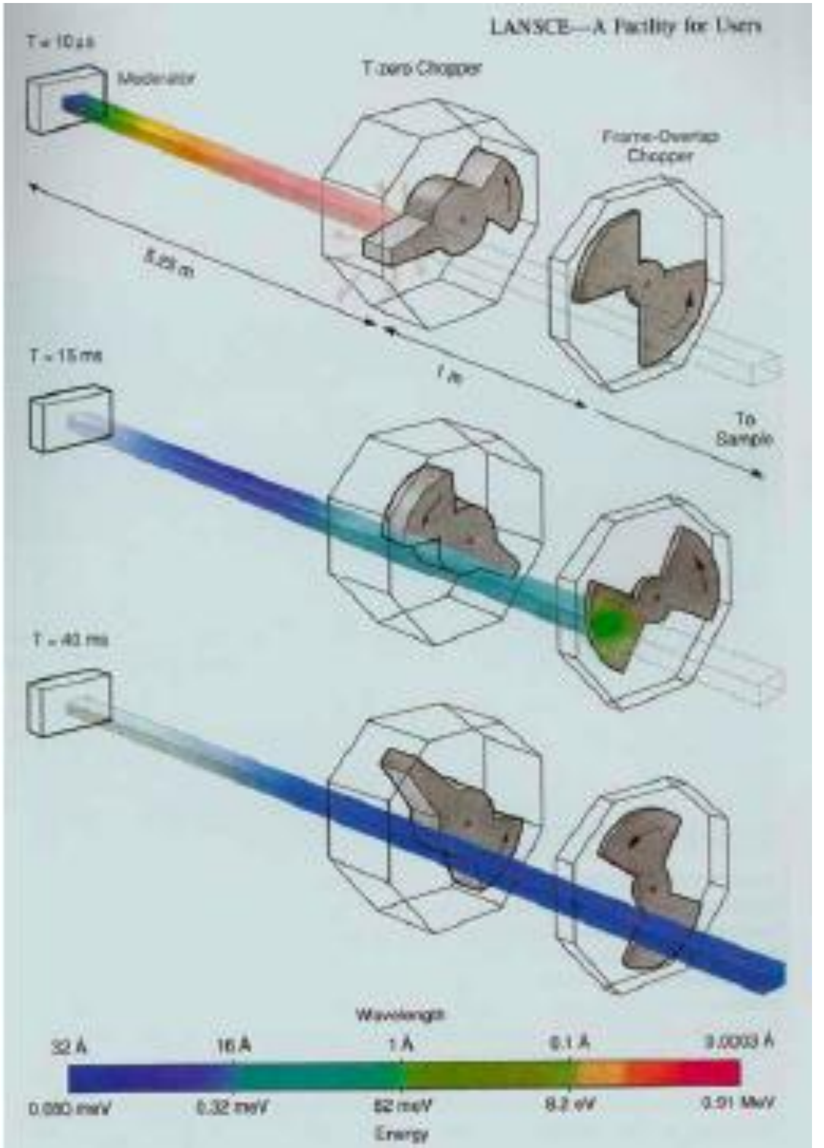
Figure 1. ISIS MAPS chopper and slit package assembly – exploded view



T-zero chopper

- **Background suppression**
- **Blocks fast neutron flash**



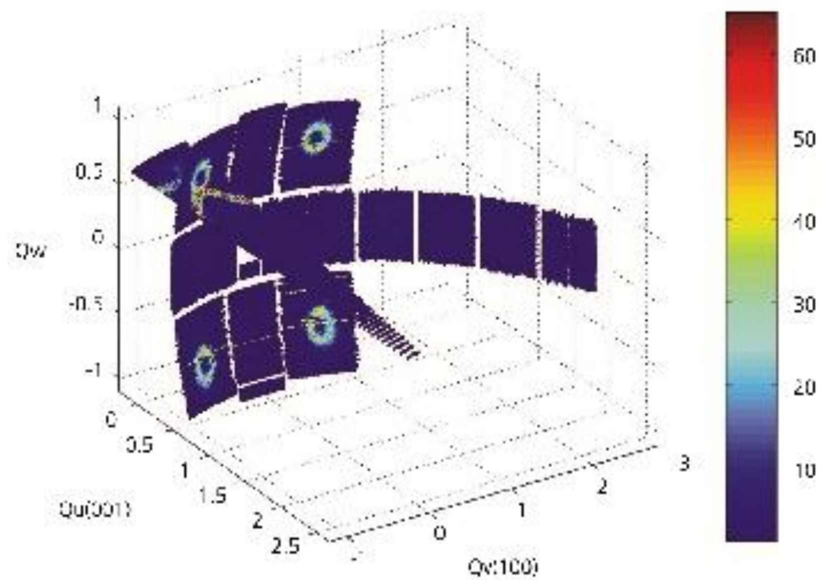


NXS School

Position sensitive detectors

- ^{3}He tubes (usu. 1 meter)
- Charge division
- Position resolution ~ cm
- Time resolution ~ 10 ns

MAPS detector bank



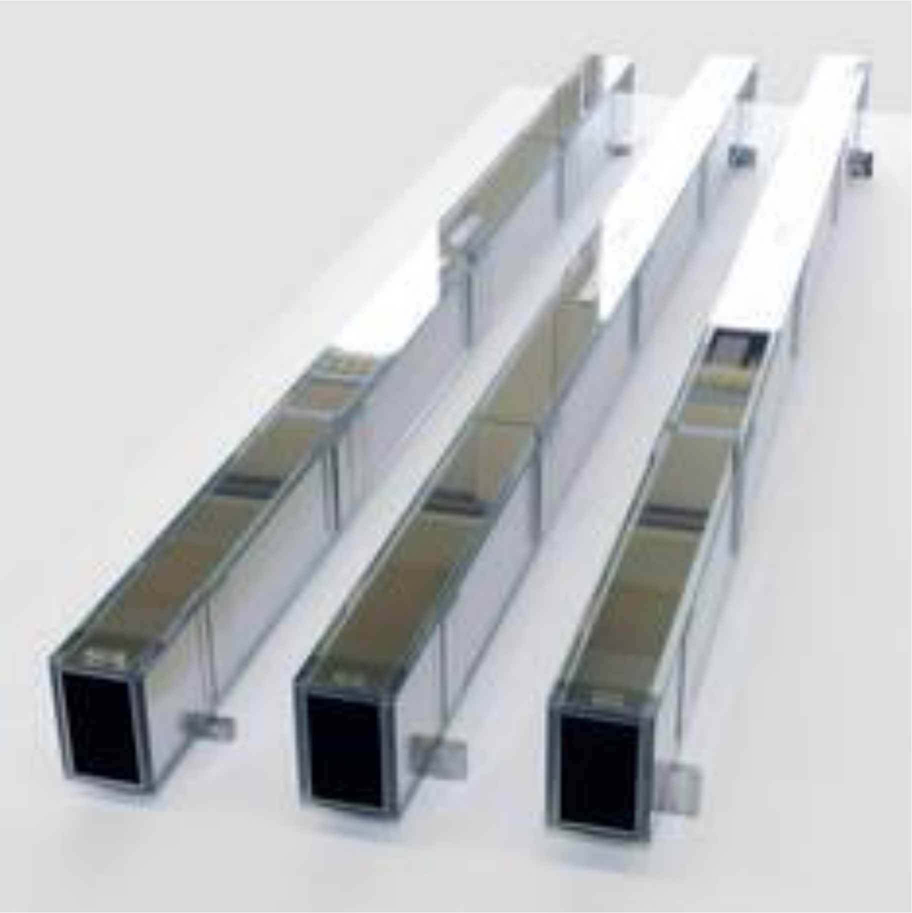
Sample environment

- **Temperature, field, pressure**
- **Heavy duty for large sample environment**
  - CCR
  - He cryostats
  - SC magnets
  - …

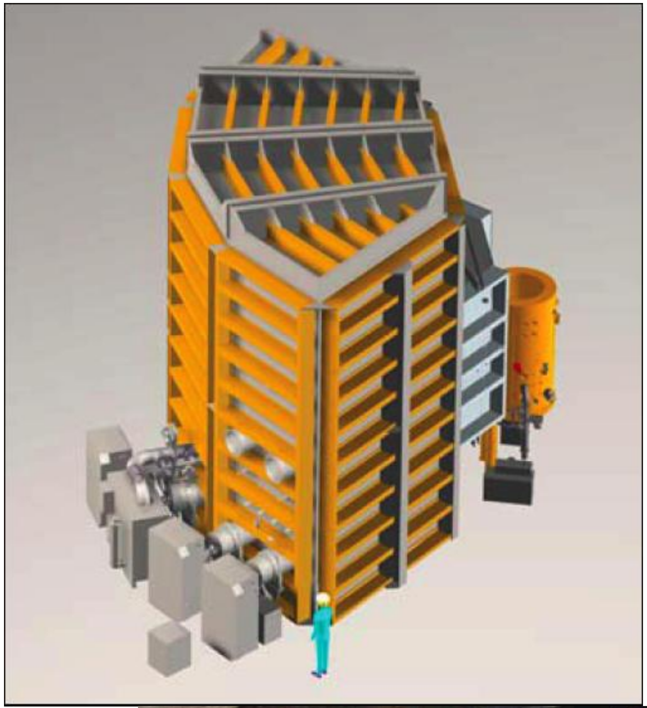
- Can be machined from Al
  ~ neutron transparent
  relatively easy to work with

Guides

- **Transport beam over long distances**
- **Background reduction**
- **Total external reflection**
  - Ni coated glass
  - Ni/Ti multilayers (supermirror)



Size matters



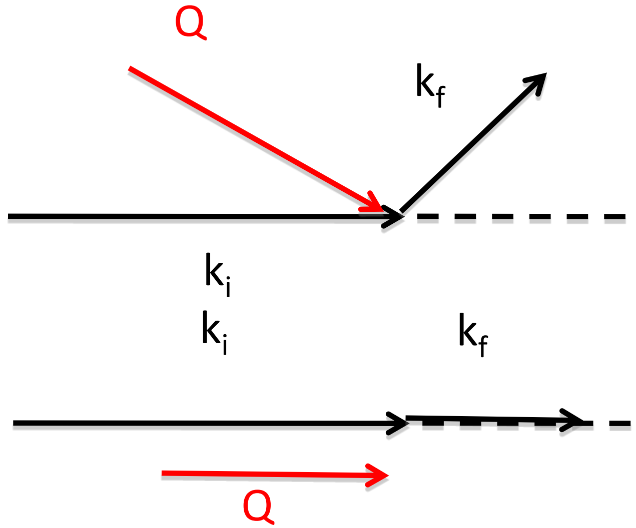
SEQUOIA detector vacuum vessel

- **Length = resolution**
  - Instruments ~ 20 – 40 m long
  - E-resolution ~ 2-4% E_i
- **More detectors**
  - SEQUOIA – 1600 tubes, 144000 pixels
  - Solid angle coverage 1.6 steradians
- **Huge data sets**
- 0.1 – 1 GB

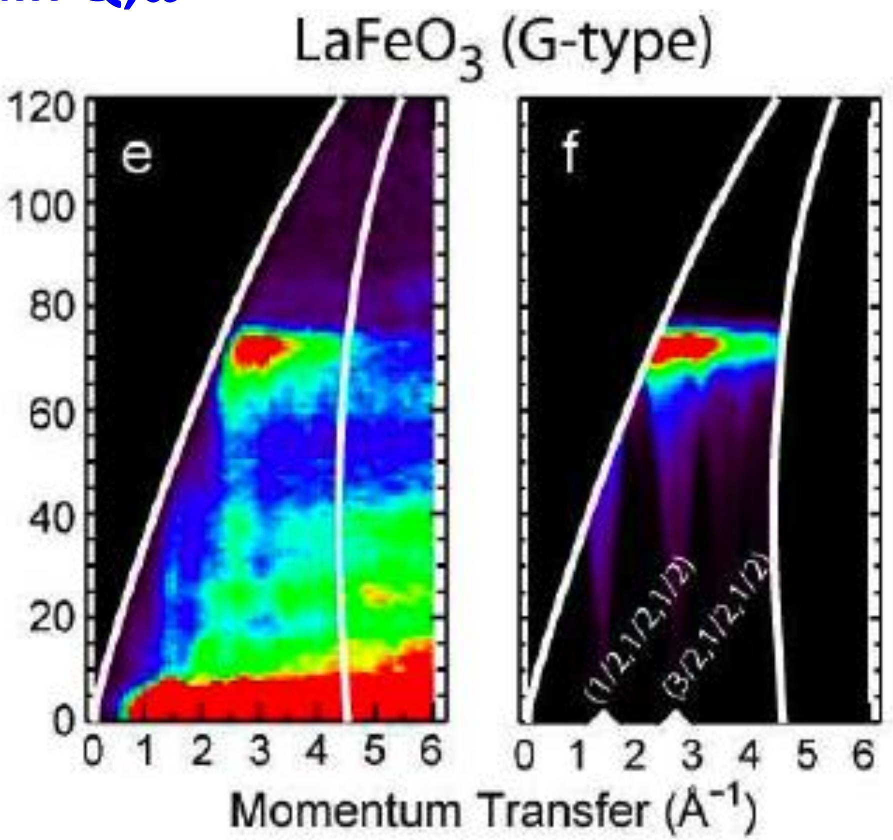
Kinematic limitations

- **Many combinations of k_i,k_f for same Q,ω**
  - Only certain configurations are used (eg. E_f-fixed)

- **Cannot "close triangle" for certain Q,ω due to kinematics**



Minimum accessible Q

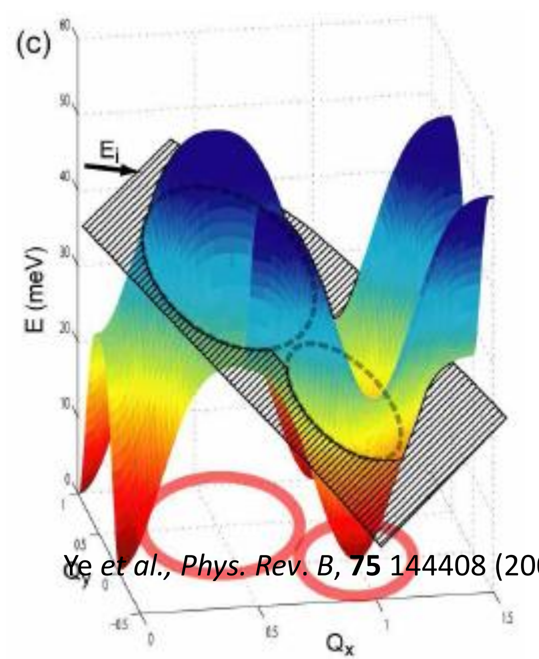


Kinematic limits, E_i=160 meV

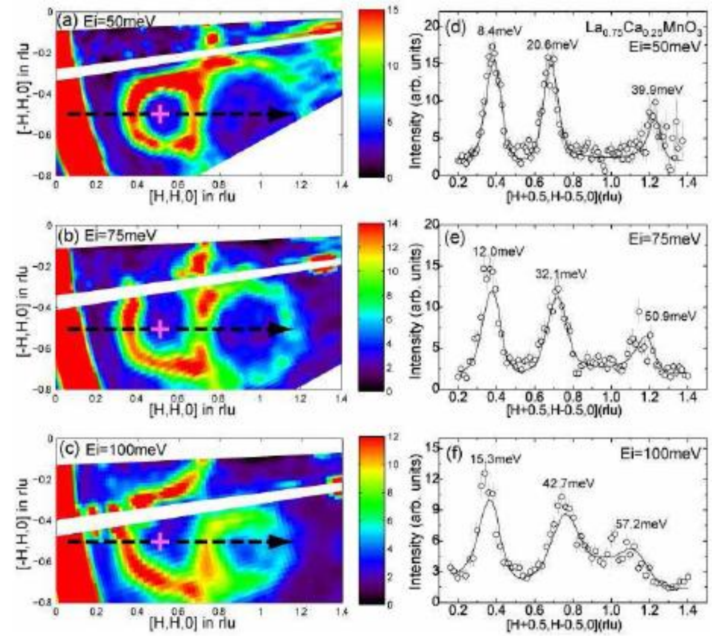
Data visualization

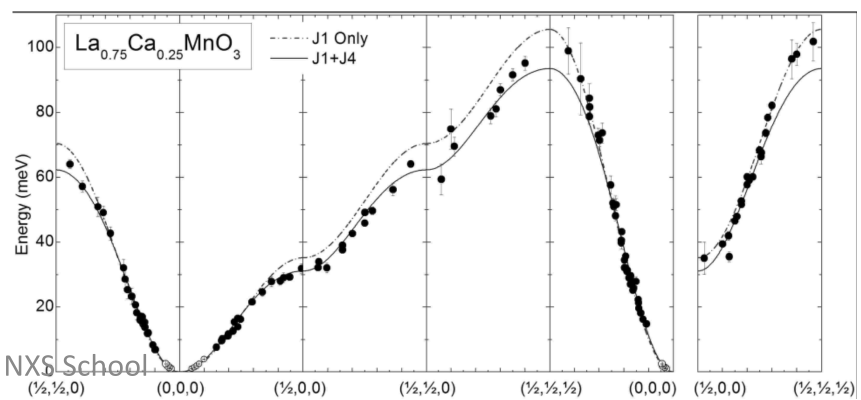
- Large, complex data from spallation sources
- Measure S(**Q**,ω) – 4D function

$La_{1-x}Ca_xMnO_3$



Ye et al., Phys. Rev. B, **75** 144408 (2007).

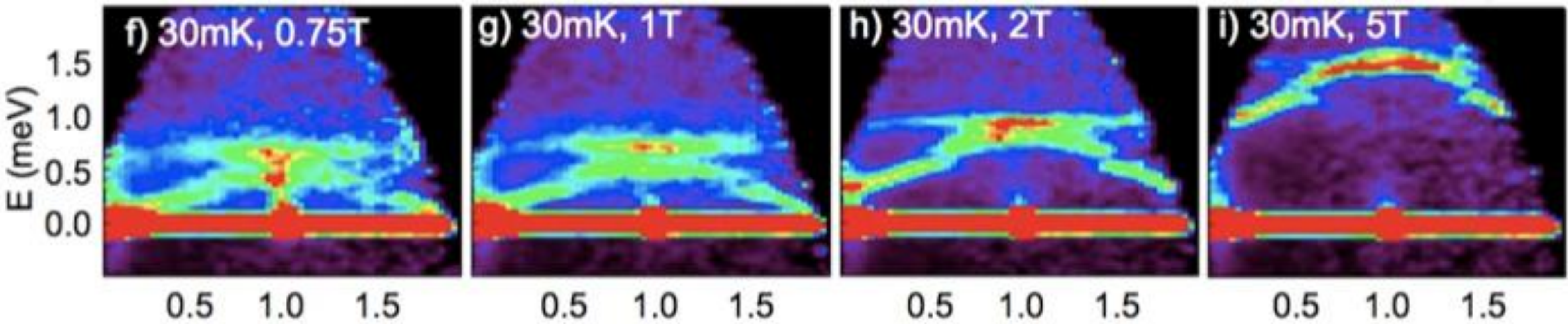




Field-induced order in the Pyrochlore Yb2Ti2O7:

Weak magnetic field // [110] induces LRO

appearance of long-lived spin waves
at low T and moderate H



References

General neutron scattering

G. Squires, "Intro to theory of thermal neutron scattering", Dover, 1978.

S. Lovesey, "Theory of neutron scattering from condensed matter", Oxford, 1984.

R. Pynn, http://www.mrl.ucsb.edu/~pynn/.

Polarized neutron scattering

Moon, Koehler, Riste, Phys. Rev **181**, 920 (1969).

Triple-axis techniques

Shirane, Shapiro, Tranquada, "Neutron scattering with a triple-axis spectrometer", Cambridge, 2002.

Time-of-flight techniques

B. Fultz, http://www.cacr.caltech.edu/projects/danse/ARCS_Book_16x.pdf