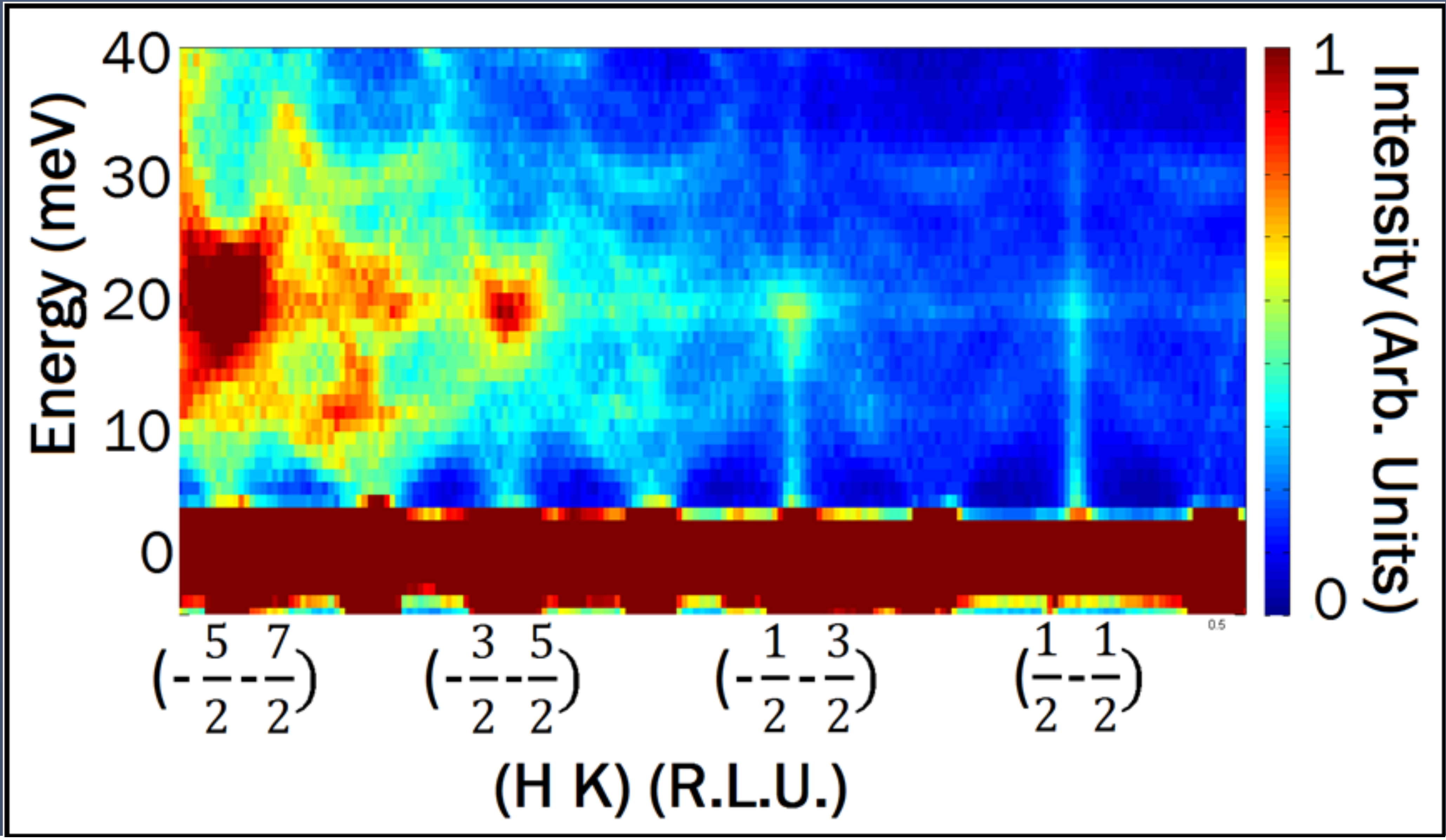
A Survey of Inelastic Neutron Scattering

- **Properties of the neutron**
- **The neutron scattering cross section**
- **The triple axis spectrometer**
- **Phonons**
- **Time-of-flight spectrometry**
- **Experimental details**



Bruce D. Gaulin
McMaster University

Brockhouse Institute for Materials Research

Neutrons:

no charge
spin = 1/2
massive:
$mc^2 \sim 1 GeV$

$^{235}U + n$

↓

daughter nuclei
+
2-3 n + γs

The Neutron as a Wave

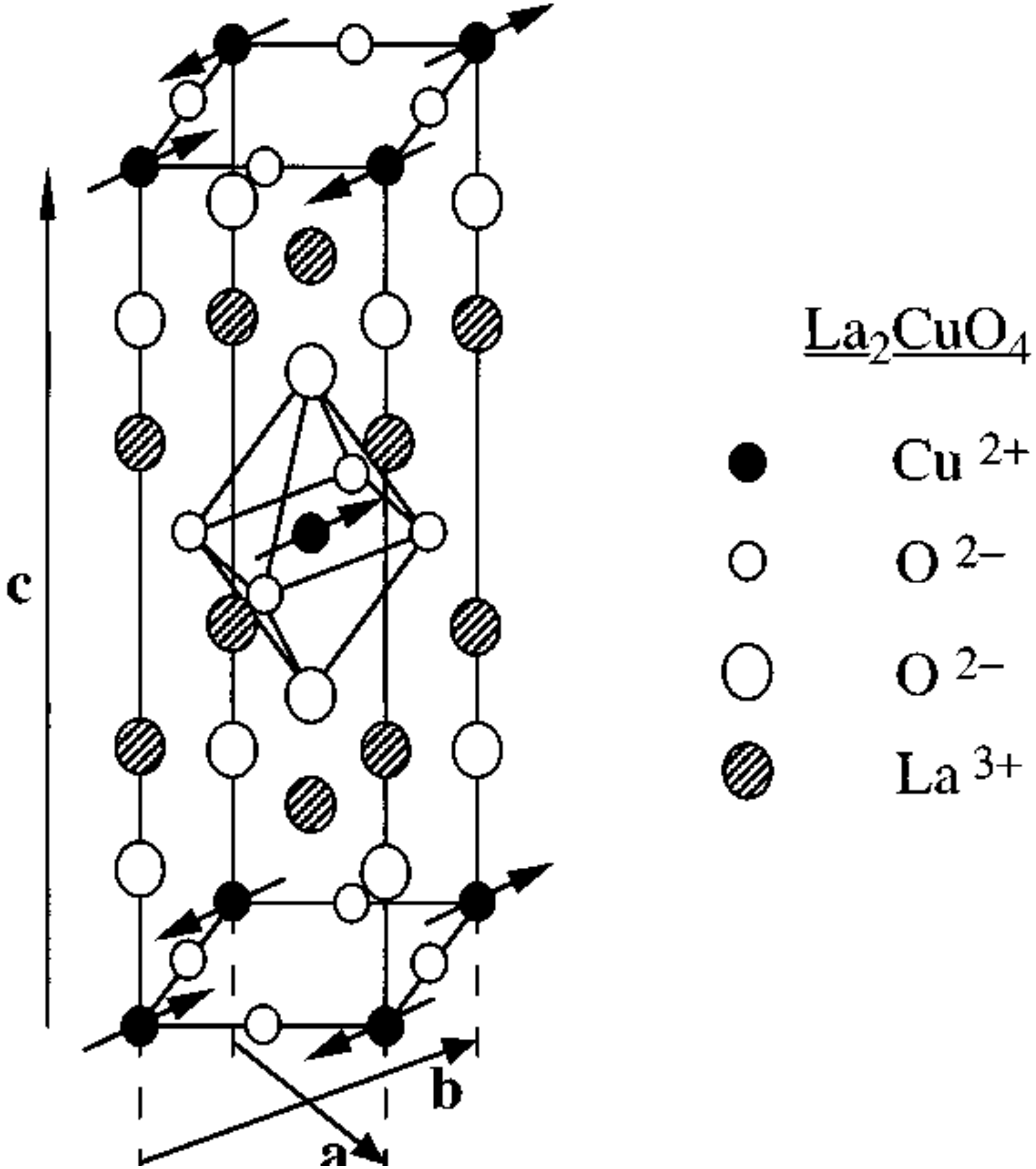
Energy, wave vector, wavelength, velocity :

$$k = \frac{m_n v}{\hbar} = \frac{2\pi}{\lambda} \qquad E = k_B T = 0.08617\, mev \bullet K^{-1} \times T$$

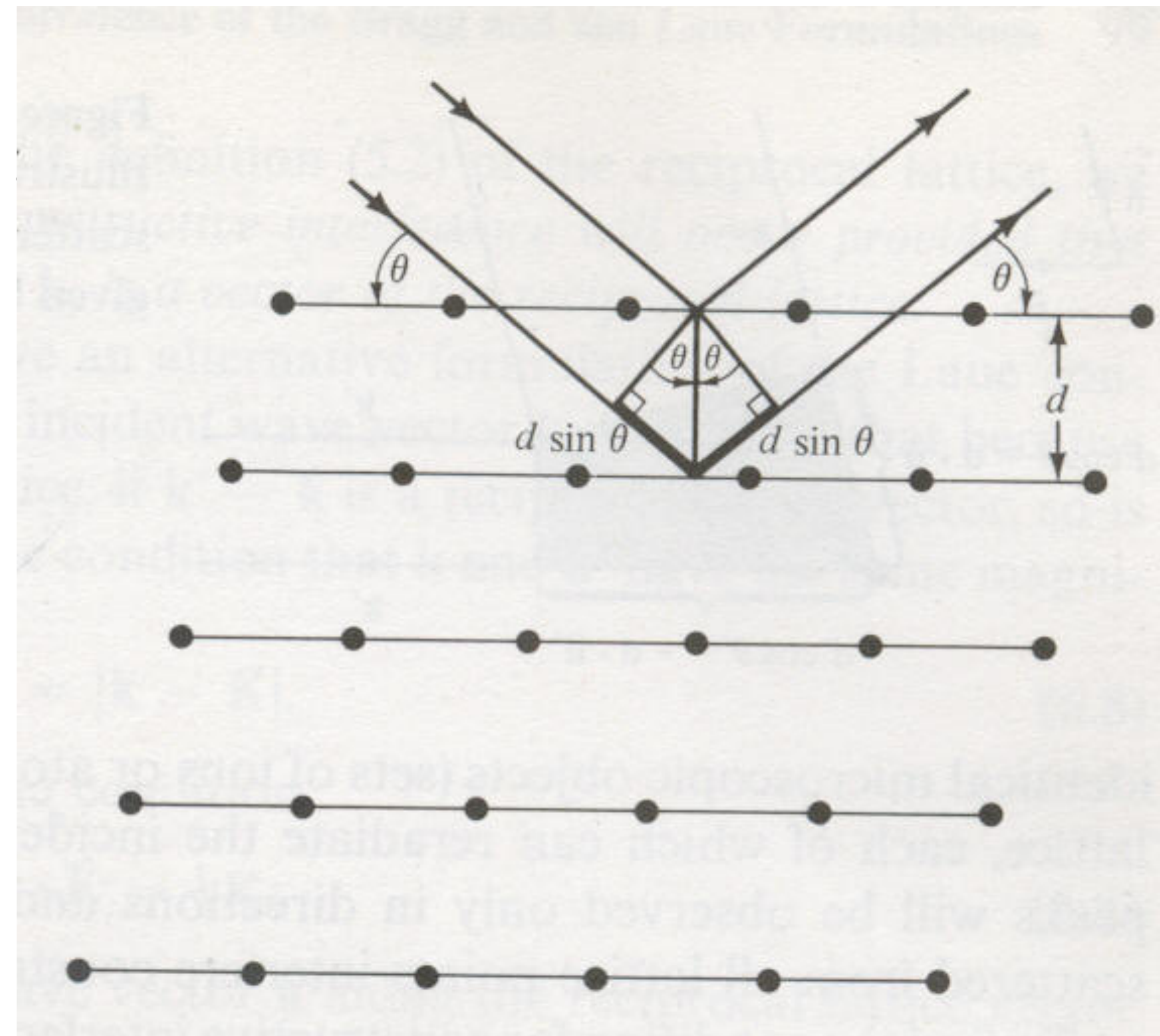
$$E = \frac{\hbar^2 k^2}{2m_n} = \frac{\hbar^2}{2m_n}\left(\frac{2\pi}{\lambda}\right)^2 = \frac{81.81\; mev \bullet \mathring{A}^2}{\lambda^2}$$

Neutrons with λ typical of interatomic spacings (~ 2 Å) have energies typical of elementary excitations in solids (~ 20 meV)

What are we typically trying to understand?

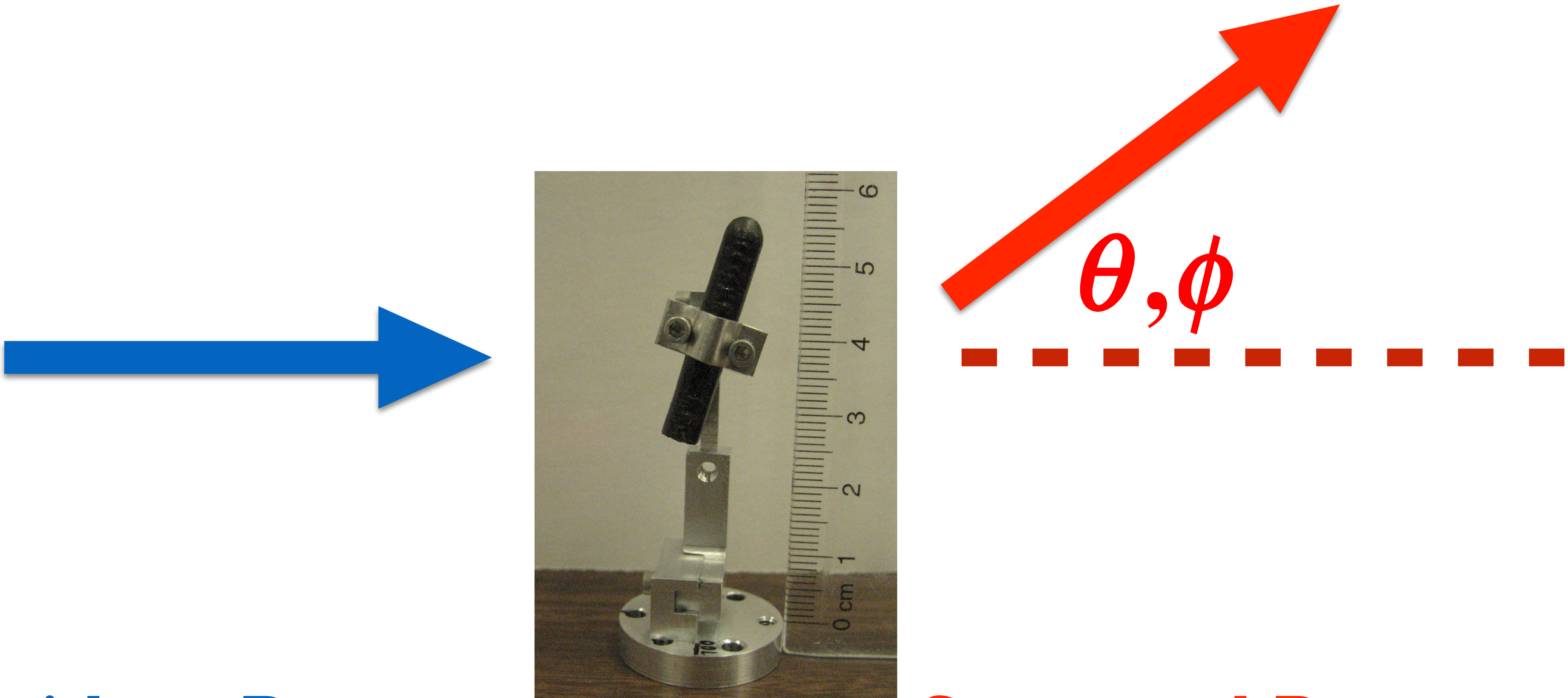


Bragg's law: $n\lambda = 2d \sin(\theta)$



- **What is the atomic and magnetic structure of new materials?**
- ***What are the dynamic properties of the atoms and the magnetic moments?***
- **How are structure and dynamics related to physical properties?**

The Basic Neutron Scattering Experiment



Incident Beam

- Monochomatic
- “White”
- “Pink”

Scattered Beam

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

Fermi's Golden Rule within the 1st Born approximation

$$W = \frac{2\pi}{\hbar} \, |\langle f | V | i \rangle|^2 \, \rho(E_f)$$

$$\partial\sigma = \frac{W}{\Phi} = \frac{m}{(2\pi\hbar^2)^2} \frac{k_f}{k_i} \, |\langle f | V | i \rangle|^2 \, \partial\Omega$$

$$\frac{\partial^2\sigma}{\partial\Omega\,\partial E_f} = \frac{k_f}{k_i} \frac{\sigma_{coherent}}{4\pi} \, N \, S_{coherent}(\vec{Q}, \hbar\omega)$$

Correlation Functions

Pair correlation function

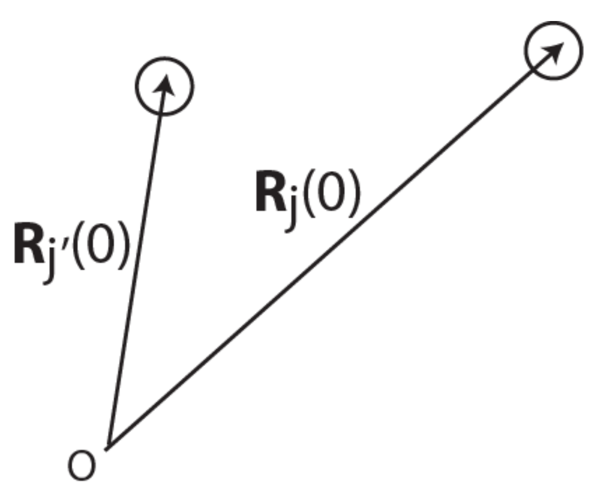
$$G(\vec{r},t) = \frac{1}{N}\int \sum_{j,j'} \delta(\vec{r}\,' - R_{j'}(0))\, \delta(\vec{r}\,' + \vec{r} - \vec{R}_j(t))\, dr'$$

Intermediate function

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

Scattering function

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



Correlation Functions

Pair correlation function

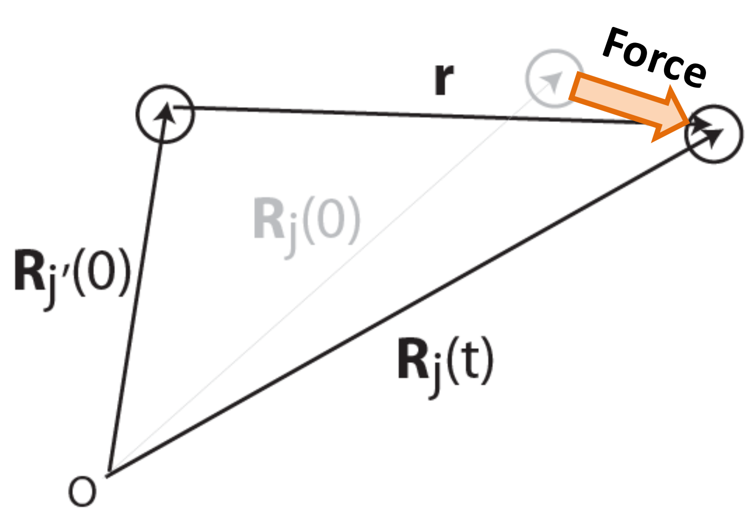
$$G(\vec{r},t)=\frac{1}{N}\int\sum_{j,j'}\delta(\vec{r}\,'-R_{j'}(0))\,\delta(\vec{r}\,'+\vec{r}-\vec{R}_j(t))\,dr'$$

Intermediate function

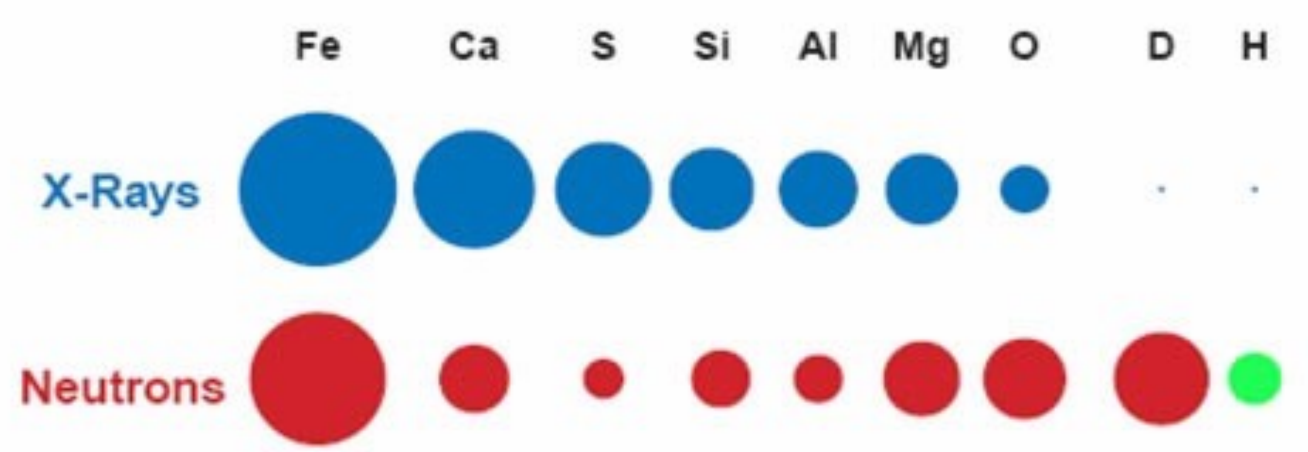
$$I(\vec{Q},t)=\int G(\vec{r},t)\,e^{i\vec{Q}\bullet\vec{r}}\,d\vec{r}=\frac{1}{N}\sum_{j,j'}e^{-i\vec{Q}\bullet\vec{R}_{j'}(0)}e^{i\vec{Q}\bullet\vec{R}_j(t)}$$

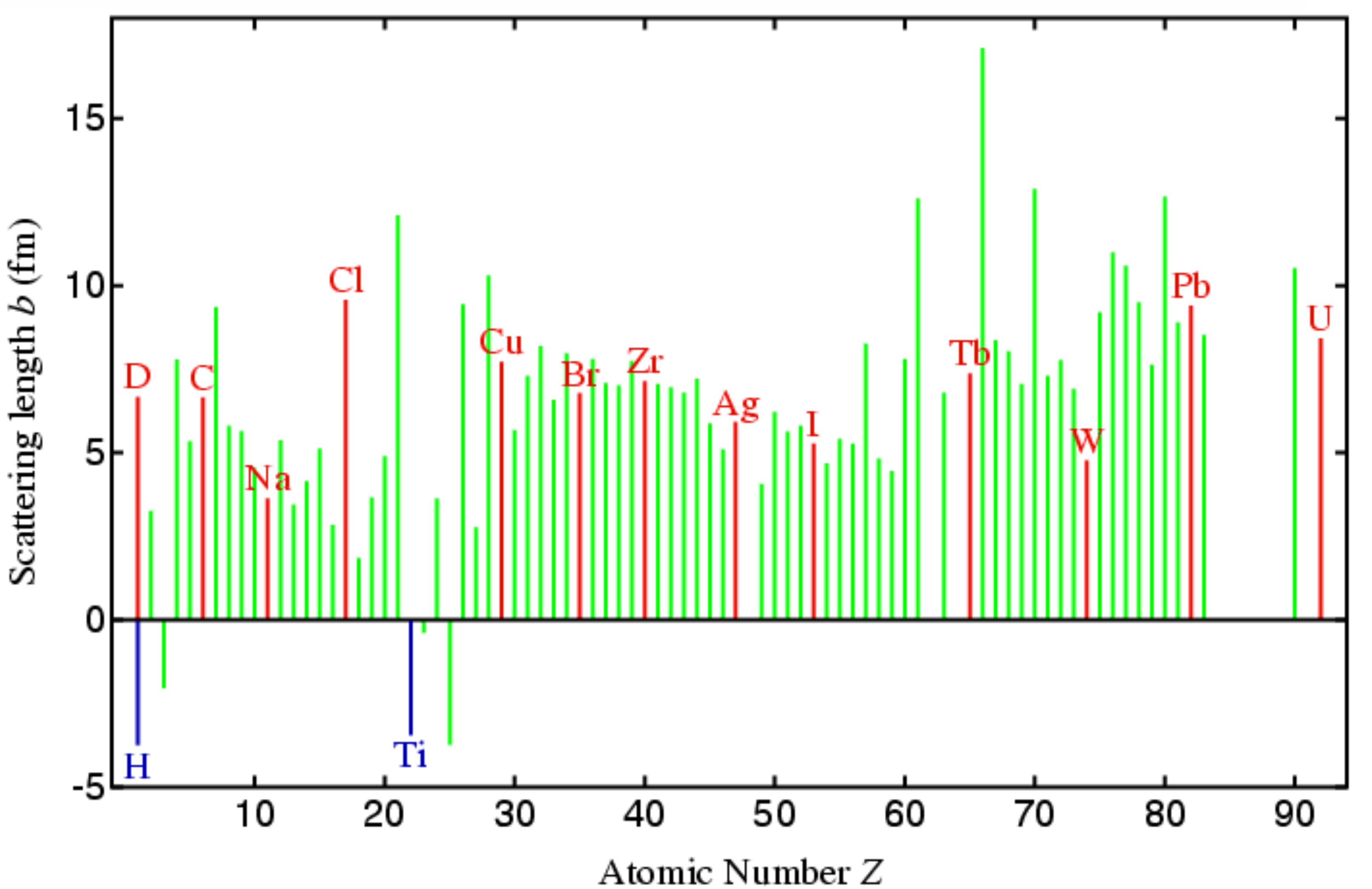
Scattering function

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



Neutrons scatter off *nuclei*





Neutrons "see" nuclei and magnetism

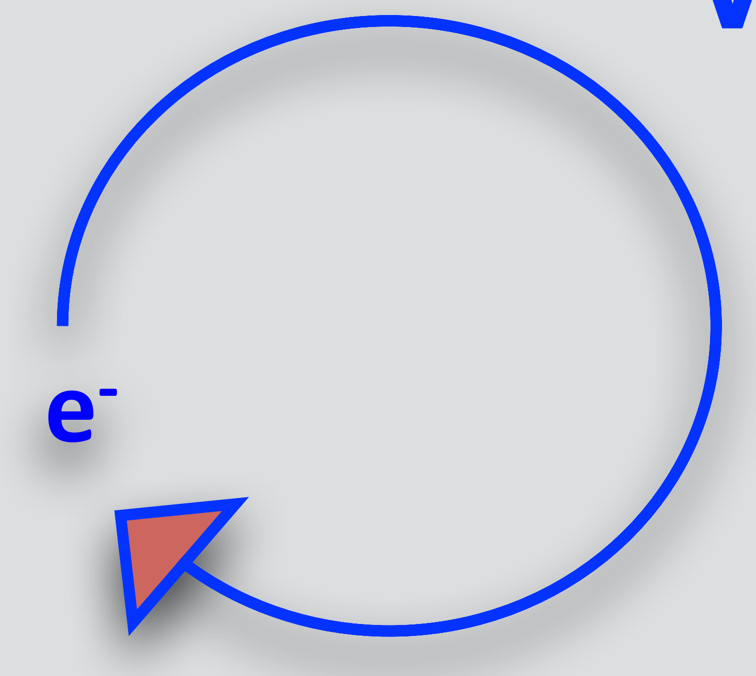
X-rays - electromagnetic radiation "see" electrons

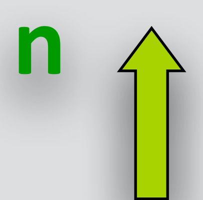
Dipole moment of the neutron interacts with the magnetic field generated by the electron

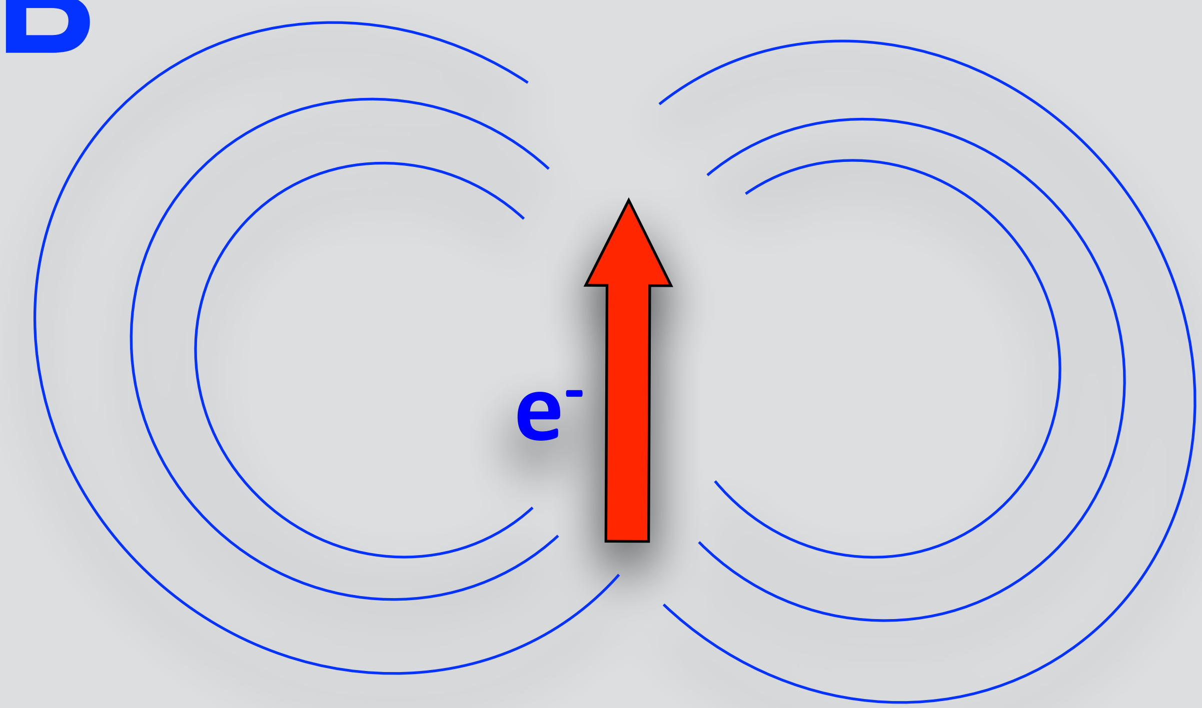
$$\mu_n = -\gamma\,\mu_N\,\sigma$$

$\gamma = 1.913$ nuclear magneton = $e\hbar/2m_n$ Pauli spin operator

$$V_M = -\mu_n \cdot B$$





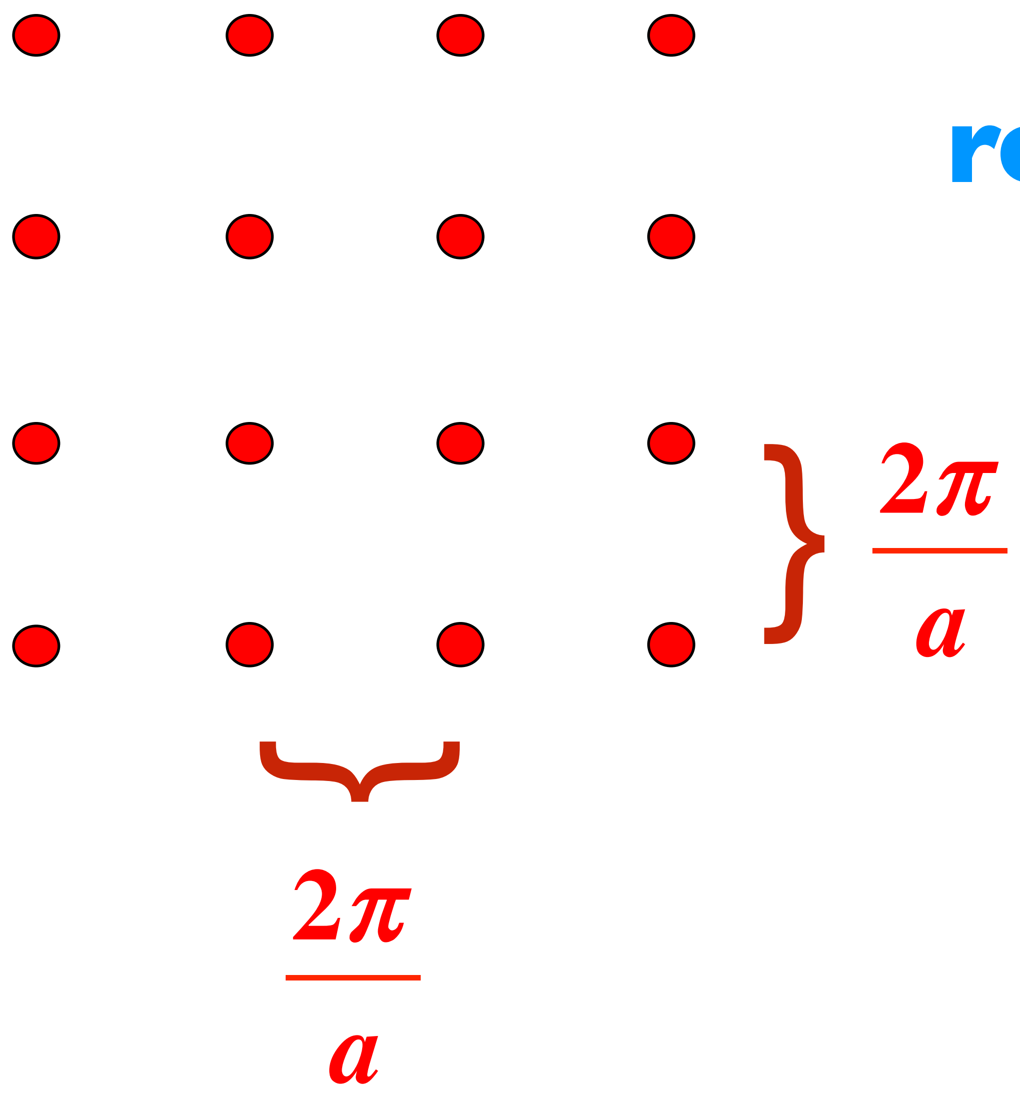


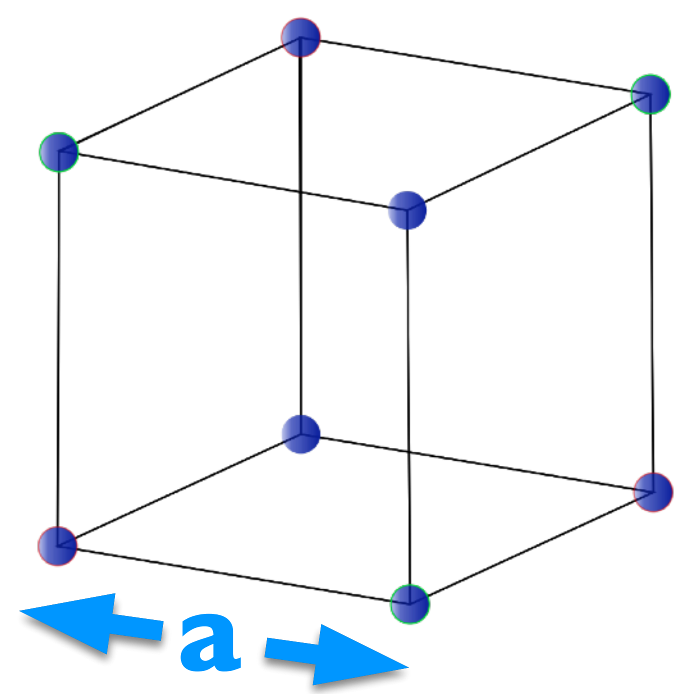
Dipole field due to orbital currents

Dipole field due to Spin of the electron(s)

Diffraction in Momentum (Q) space

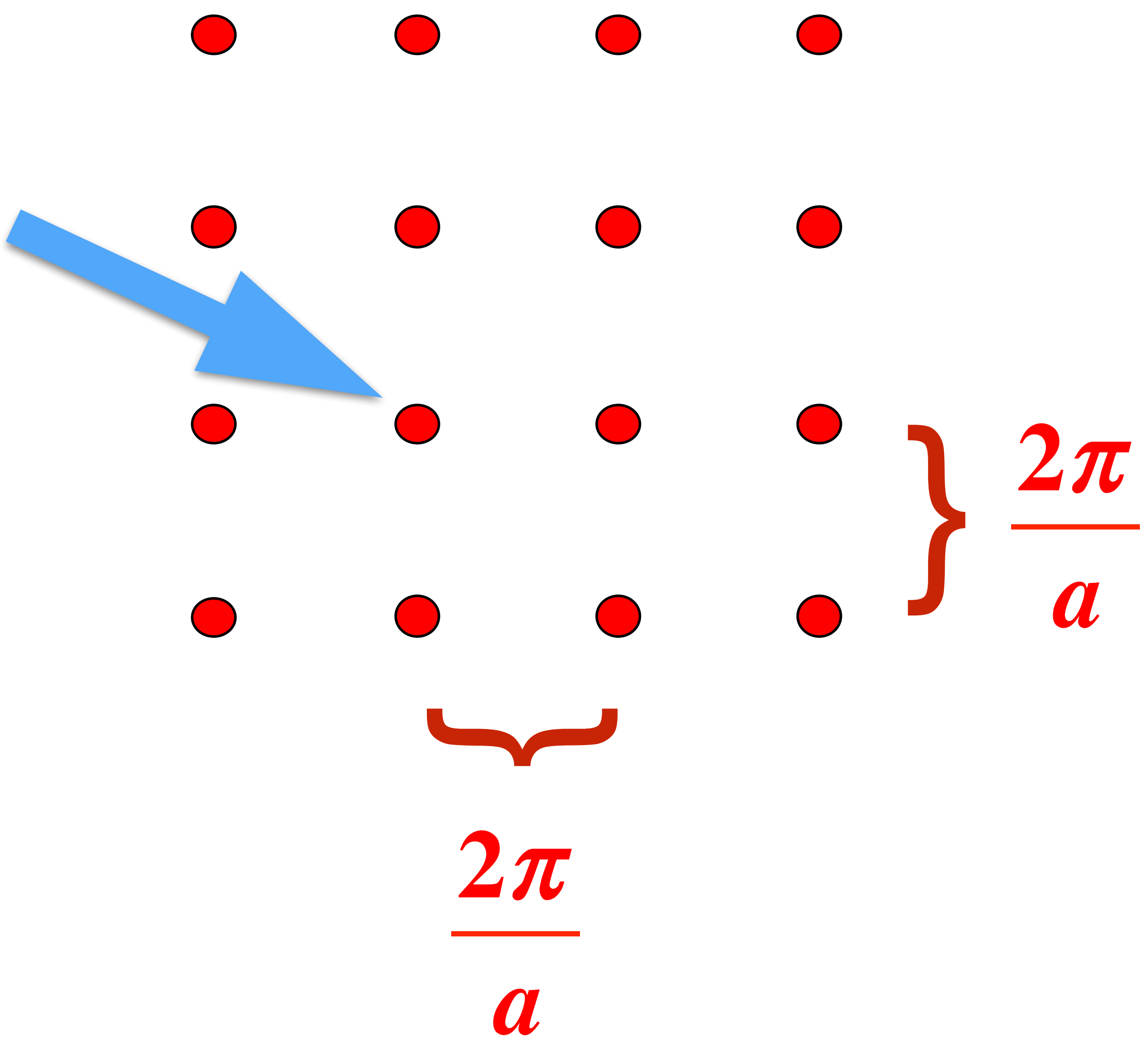
In momentum space, our sample is represented by its reciprocal lattice





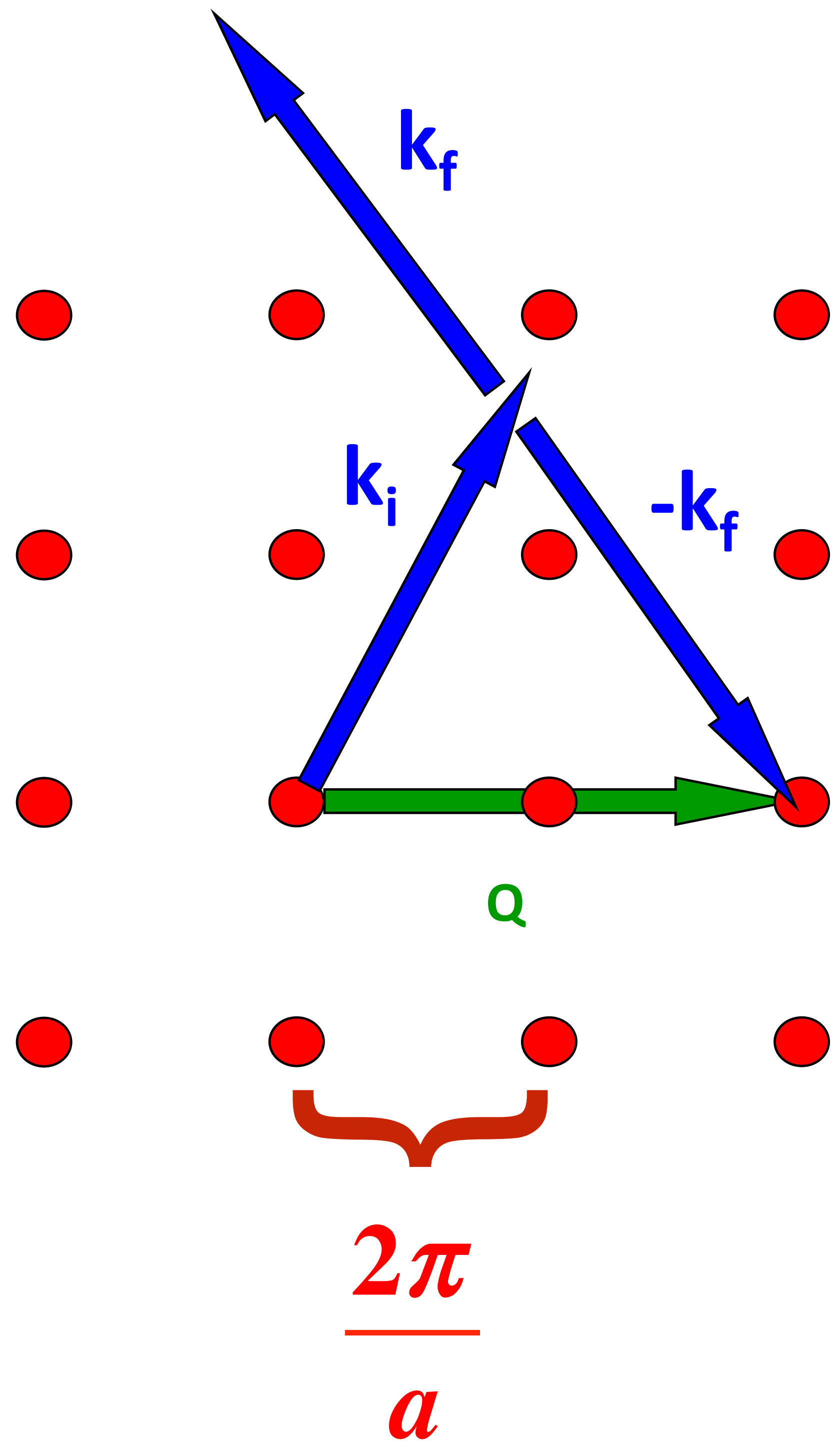
Diffraction in Momentum (Q) space

Origin of reciprocal space



Remains fixed for all sample orientations

Diffraction in Momentum (Q) space



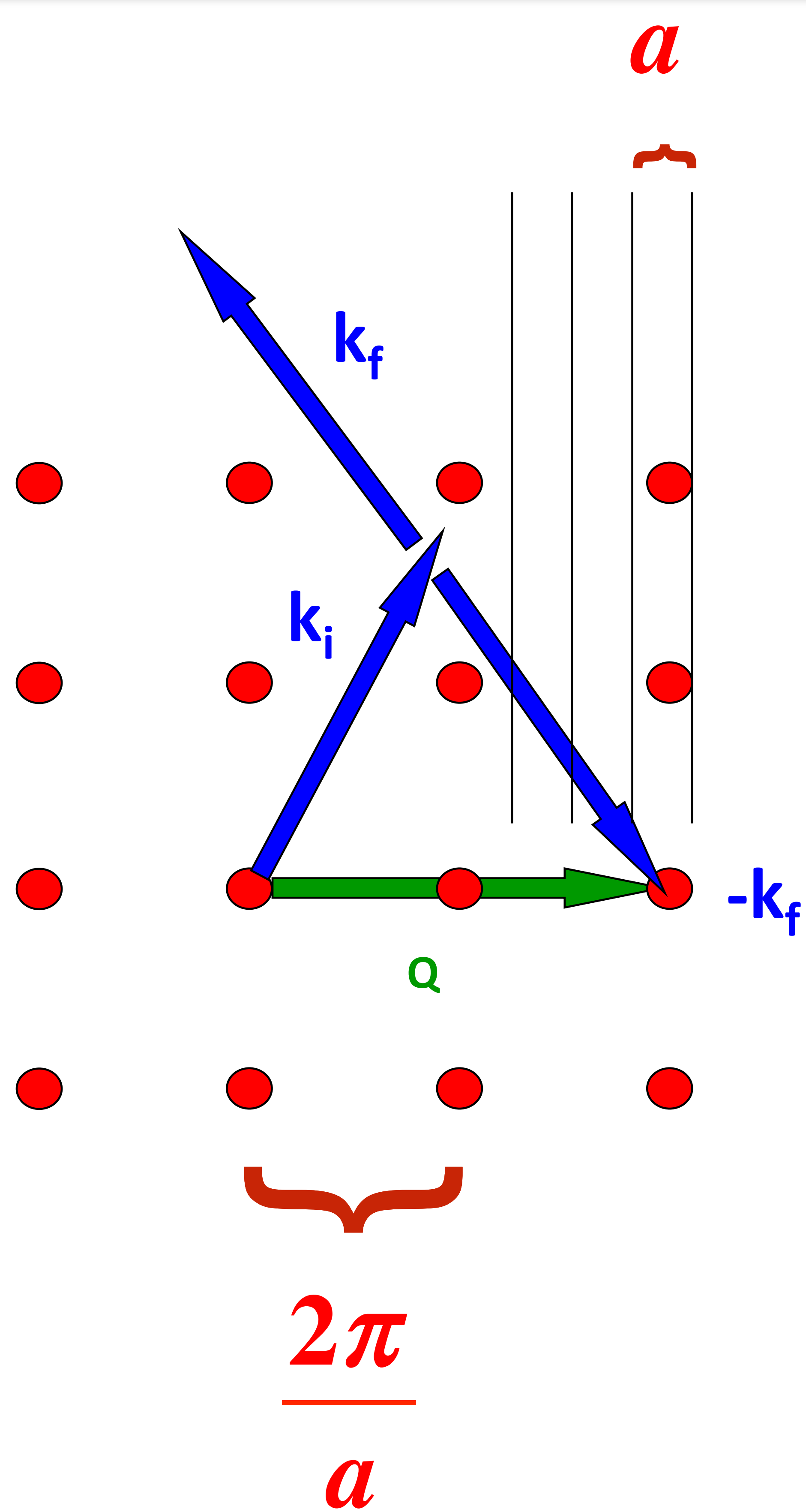
Bragg diffraction

constructive interference when

$$\vec{Q} = \vec{k}_i - \vec{k}_f = \vec{\tau}$$

= a reciprocal lattice vector

Diffraction in Momentum (Q) space



Bragg diffraction

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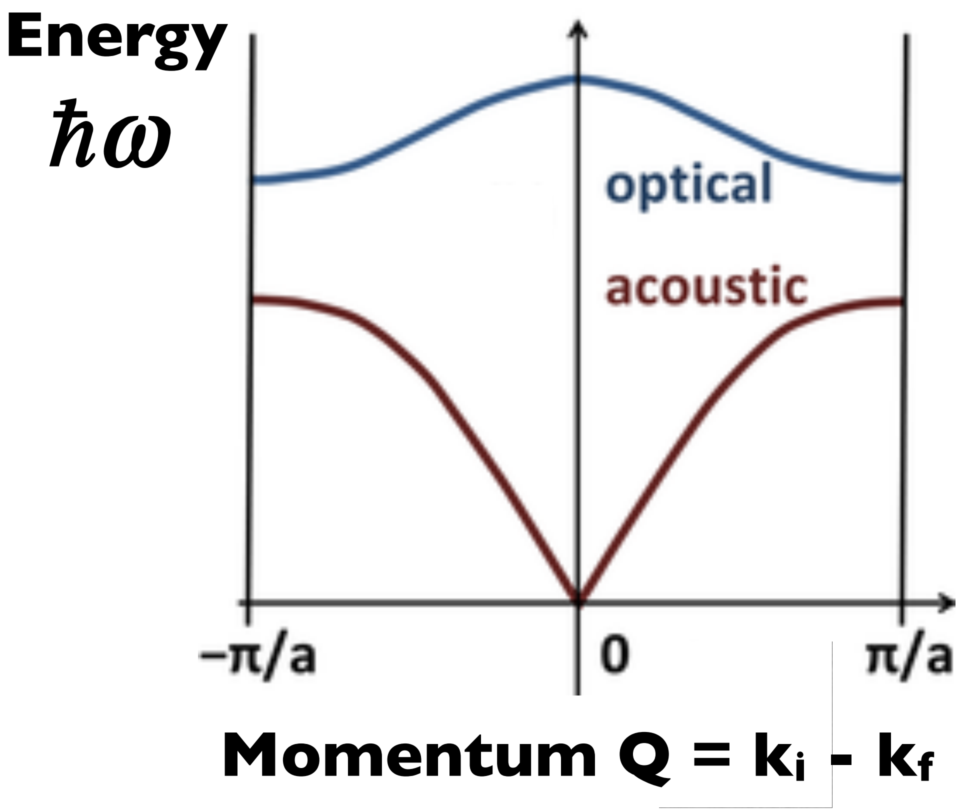
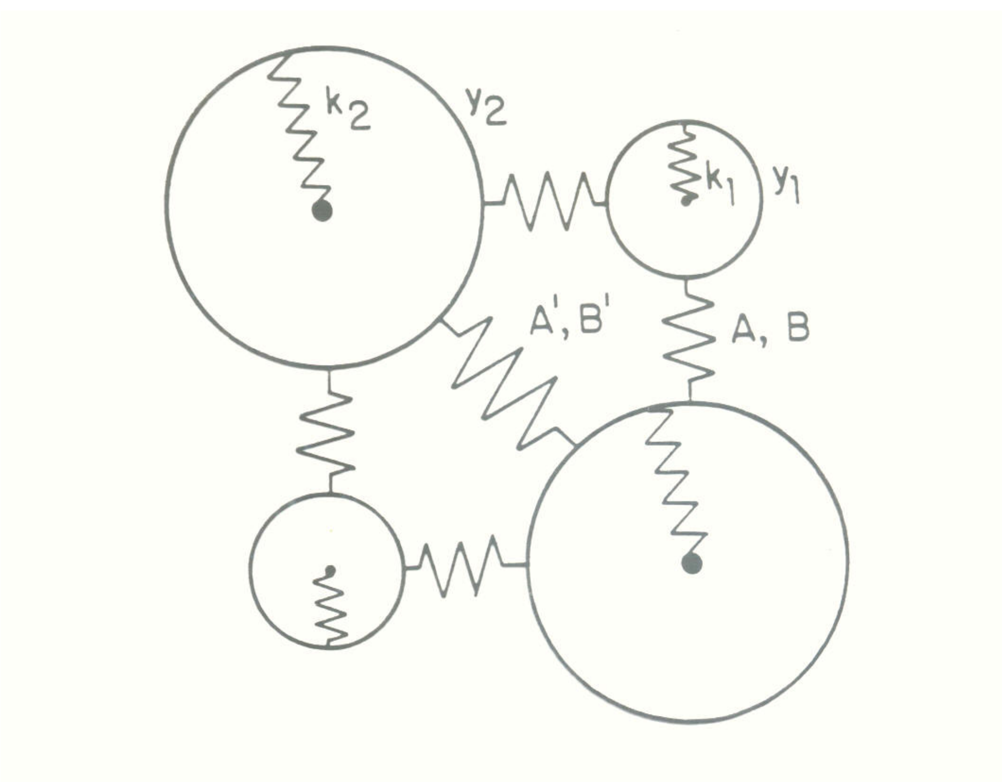
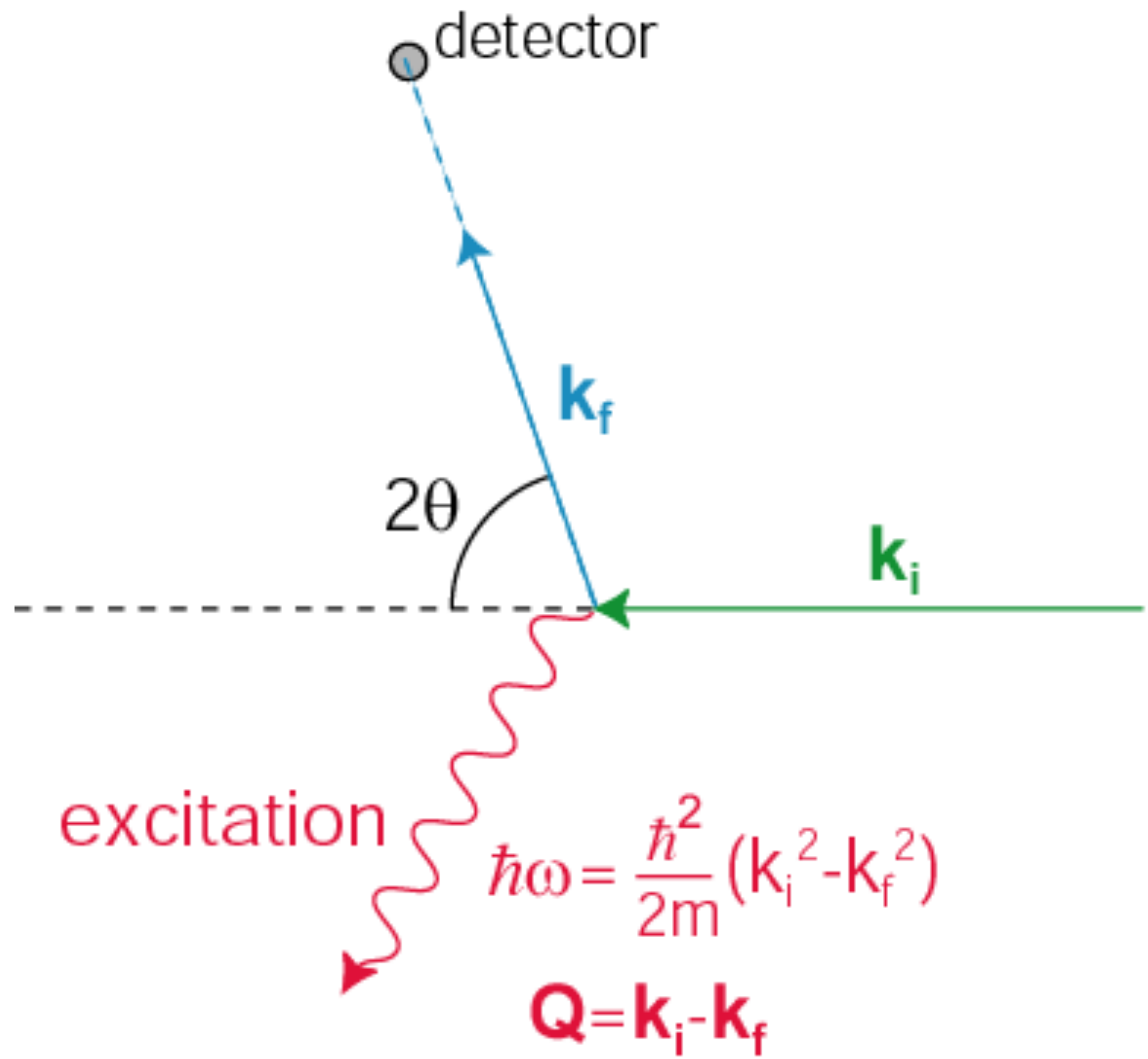
Elementary Excitations





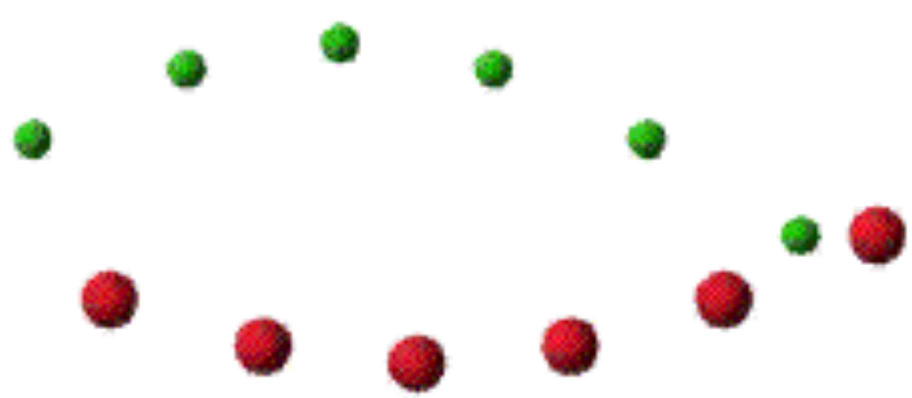


Phonon Polarizations

Longitudinal Acoustic

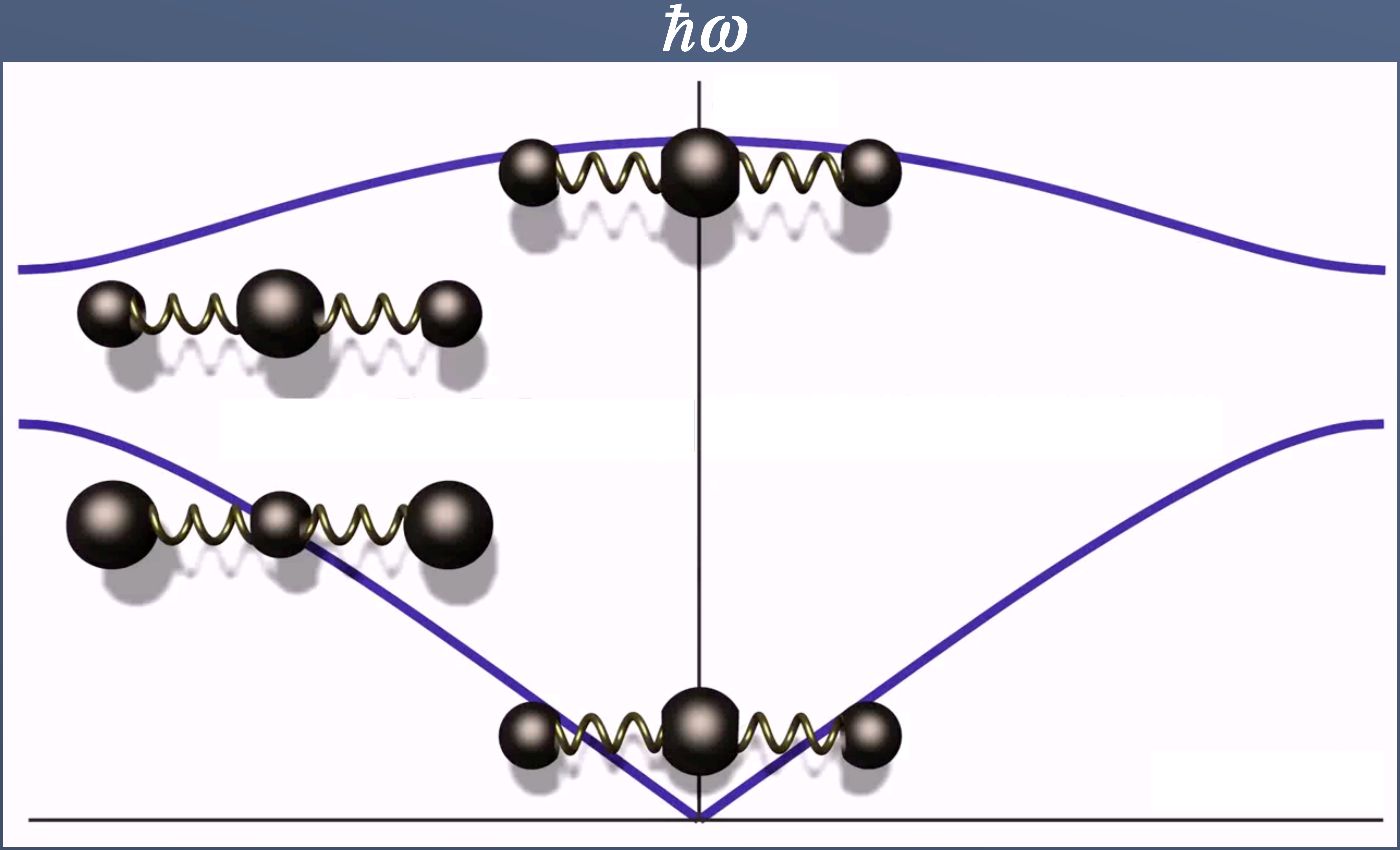
Transverse Acoustic

Transverse Acoustic



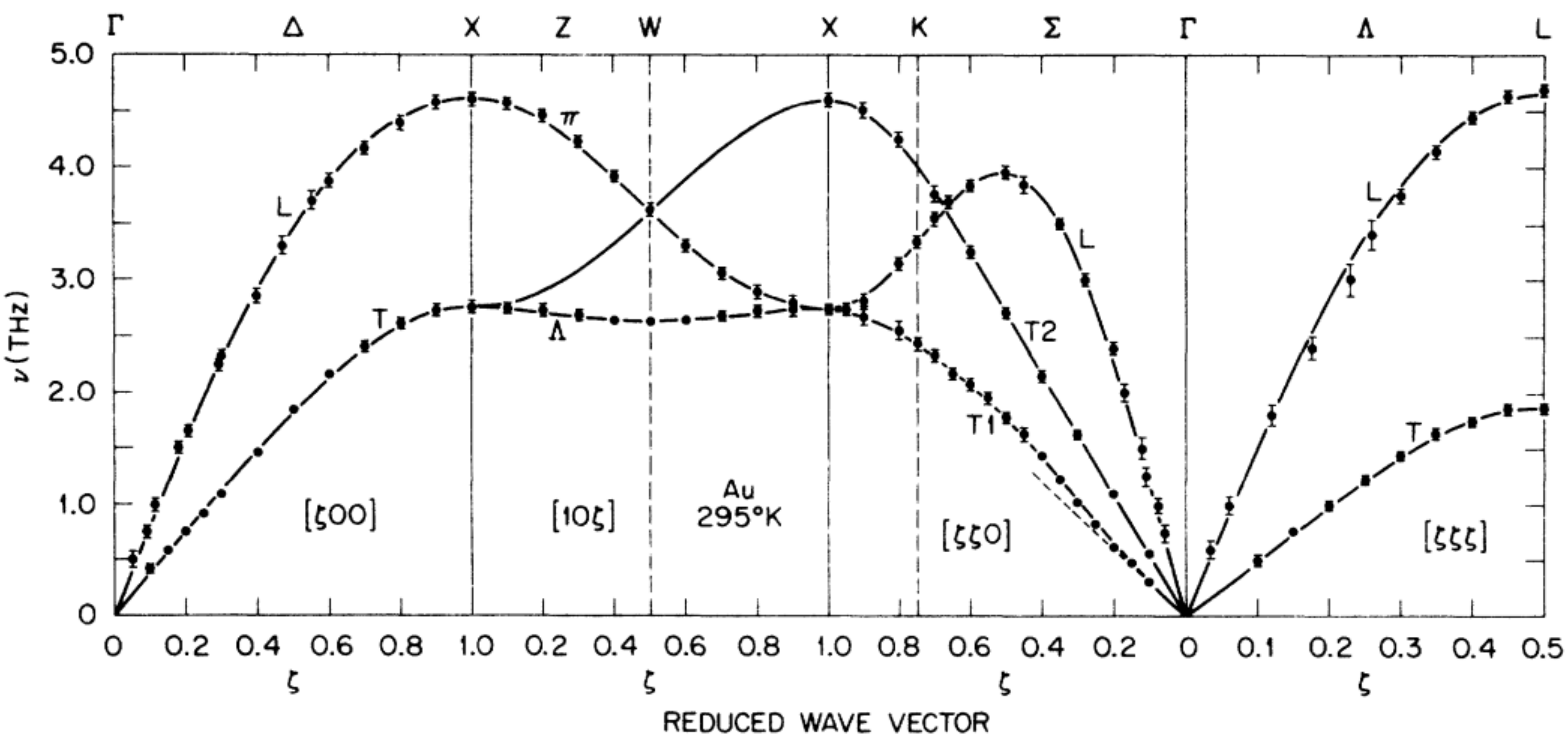
Transverse Optic

Phonon eigenvectors and eigenvalues

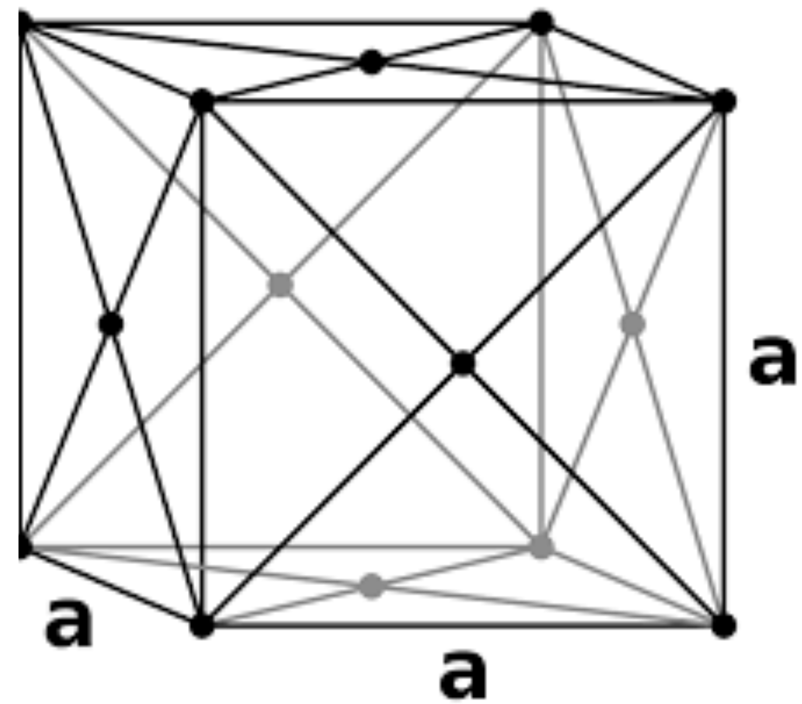


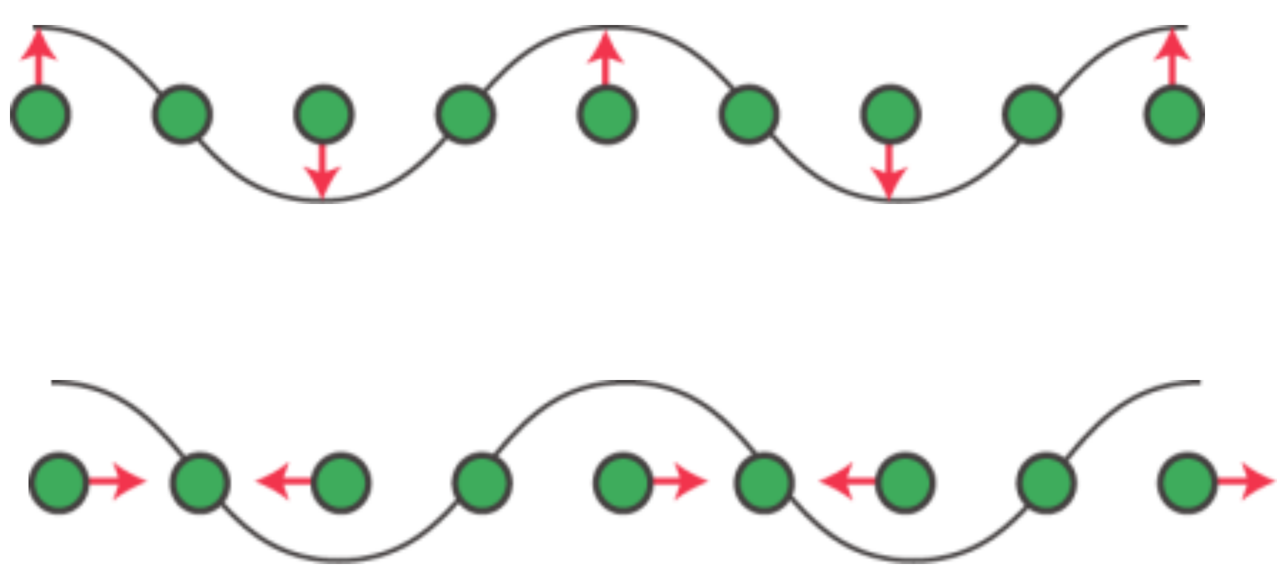
Momentum $Q = k_i - k_f$

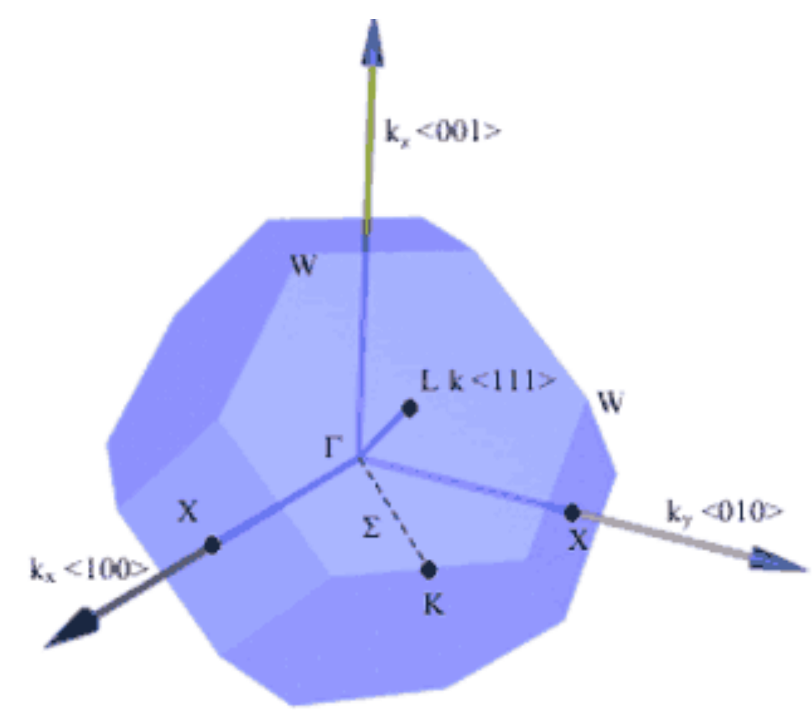
Phonons in 3D



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

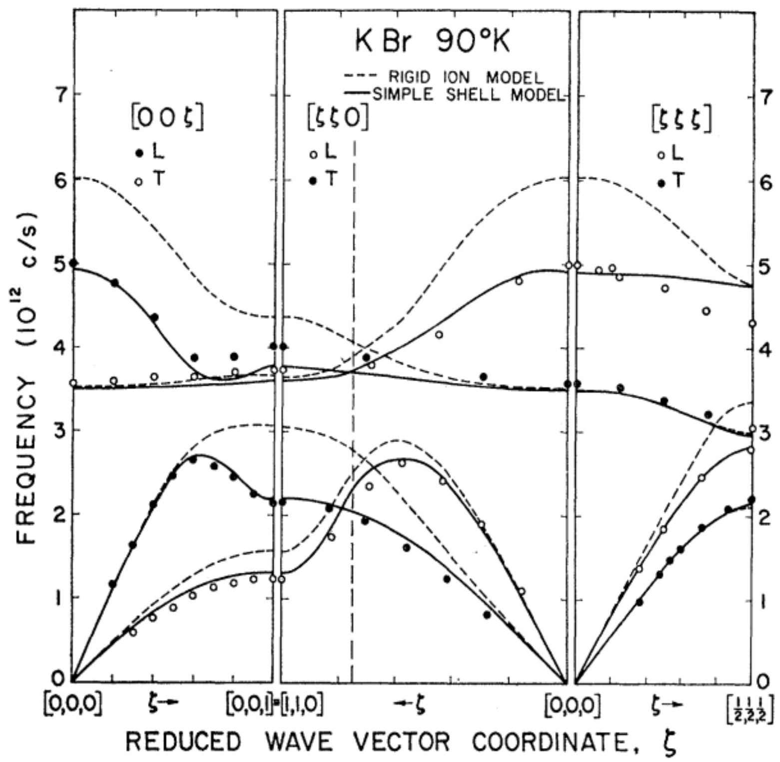




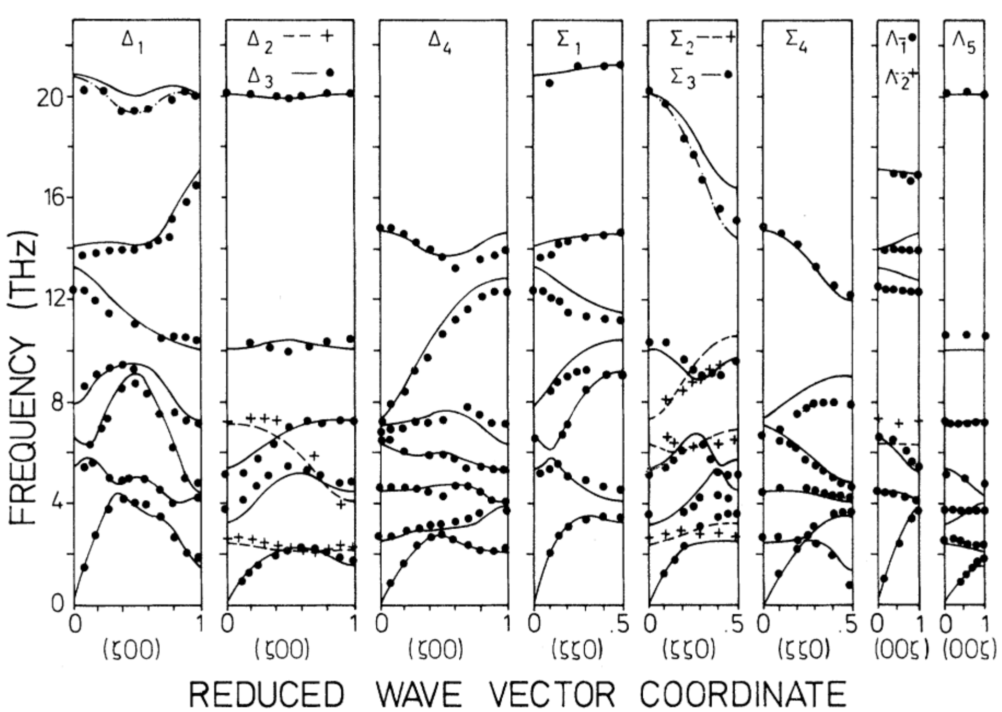


FCC Brillouin zone

Phonons in more complicated 3D structures



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



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

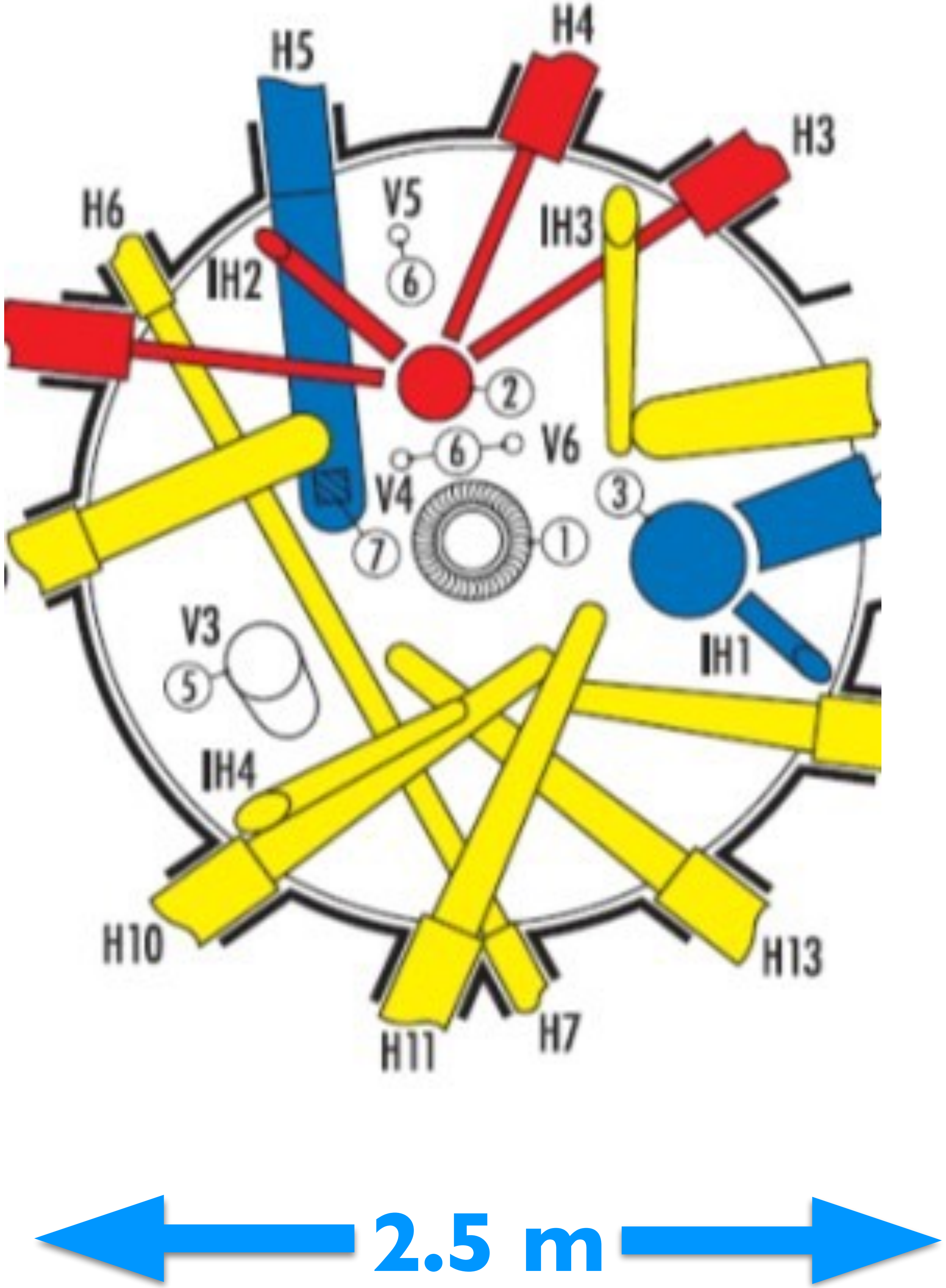
KBr - two atoms/unit cell

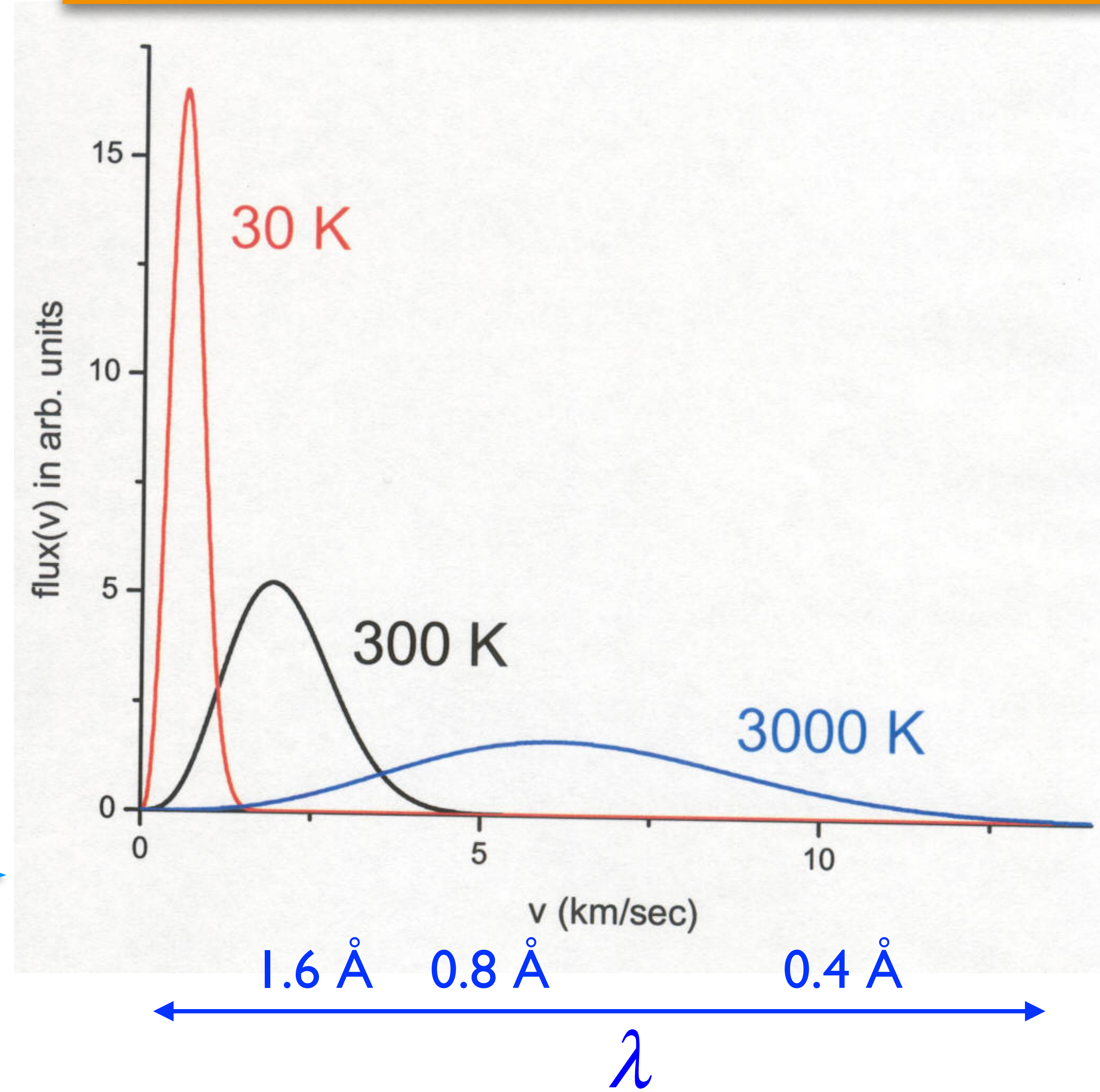
3 acoustic phonon branches
3 optic phonon branches

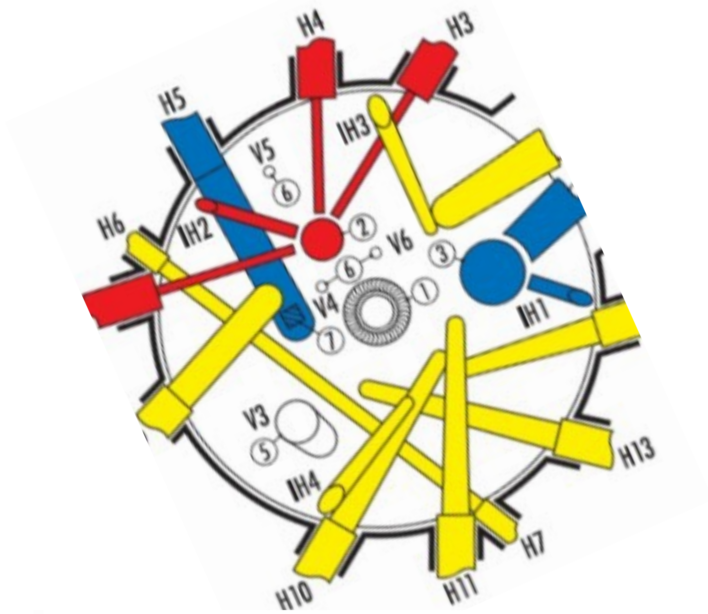
La_2CuO_4 many atoms/unit cell

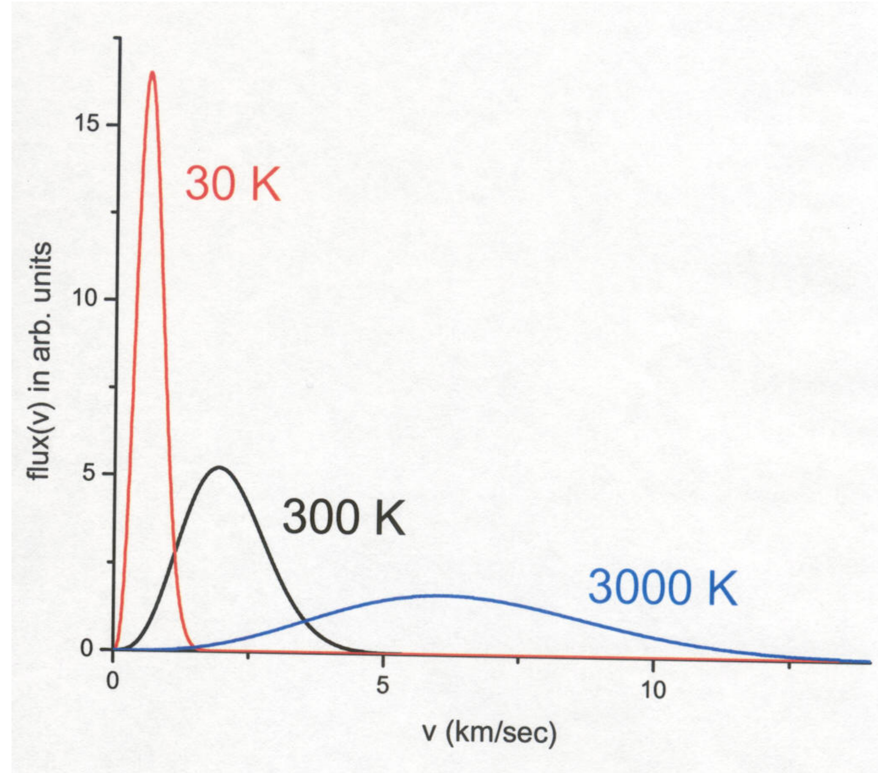
3 acoustic phonon branches
3n-3 = many optic phonon branches

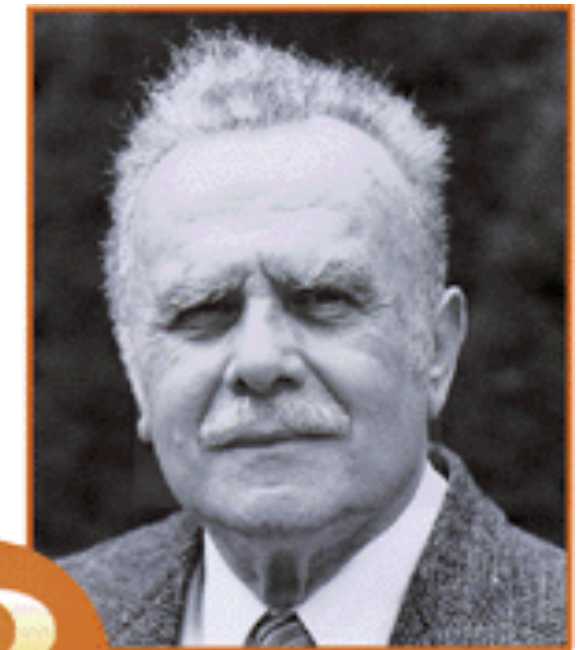
The High Flux Reactor at the ILL and its moderators and beam ports



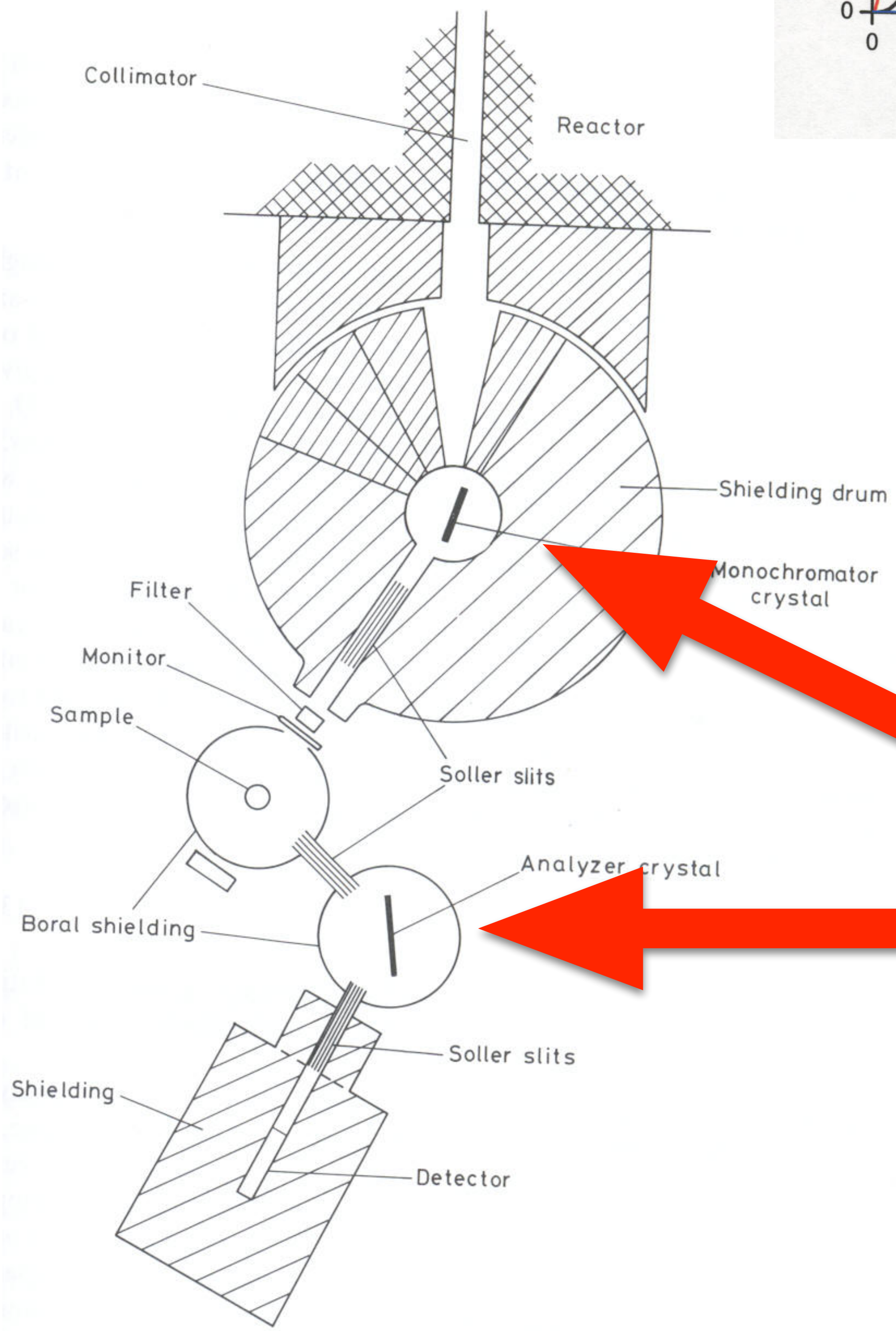


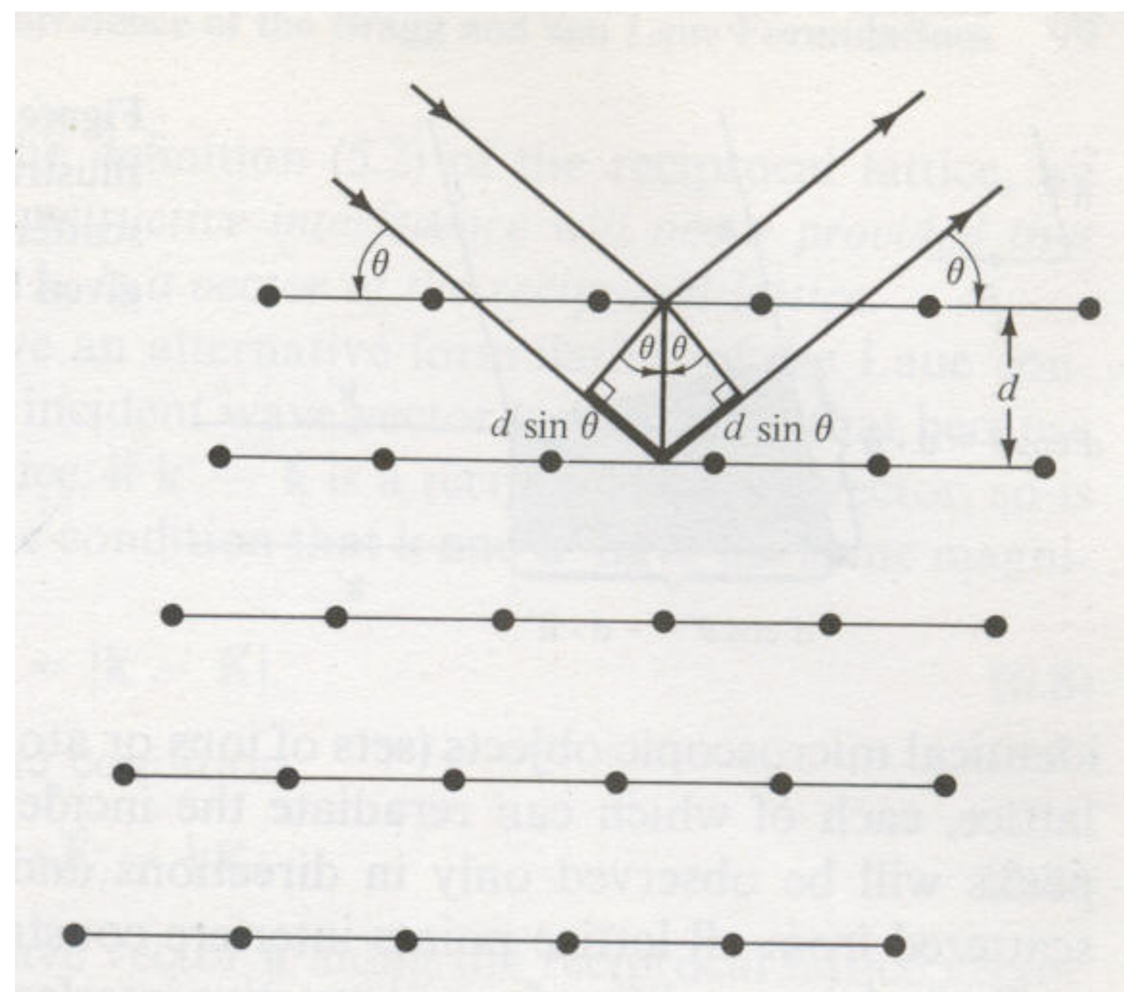




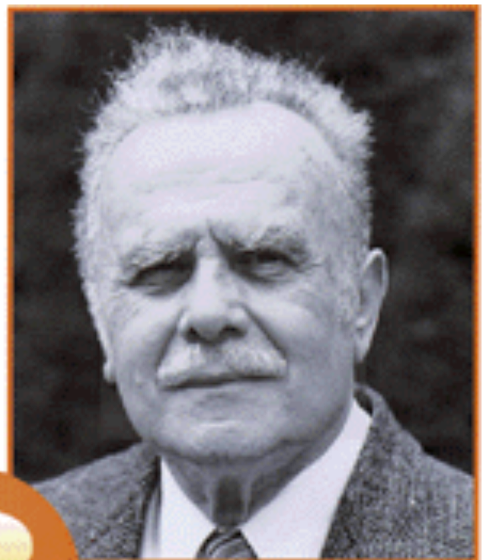


Betram N. Brockhouse,
McMater University, Hamilton, Ontario, Canada, receives one half of the 1994 Nobel Prize in Physics for the development of neutron spectroscopy.





Brockhouse's Triple Axis Spectrometer



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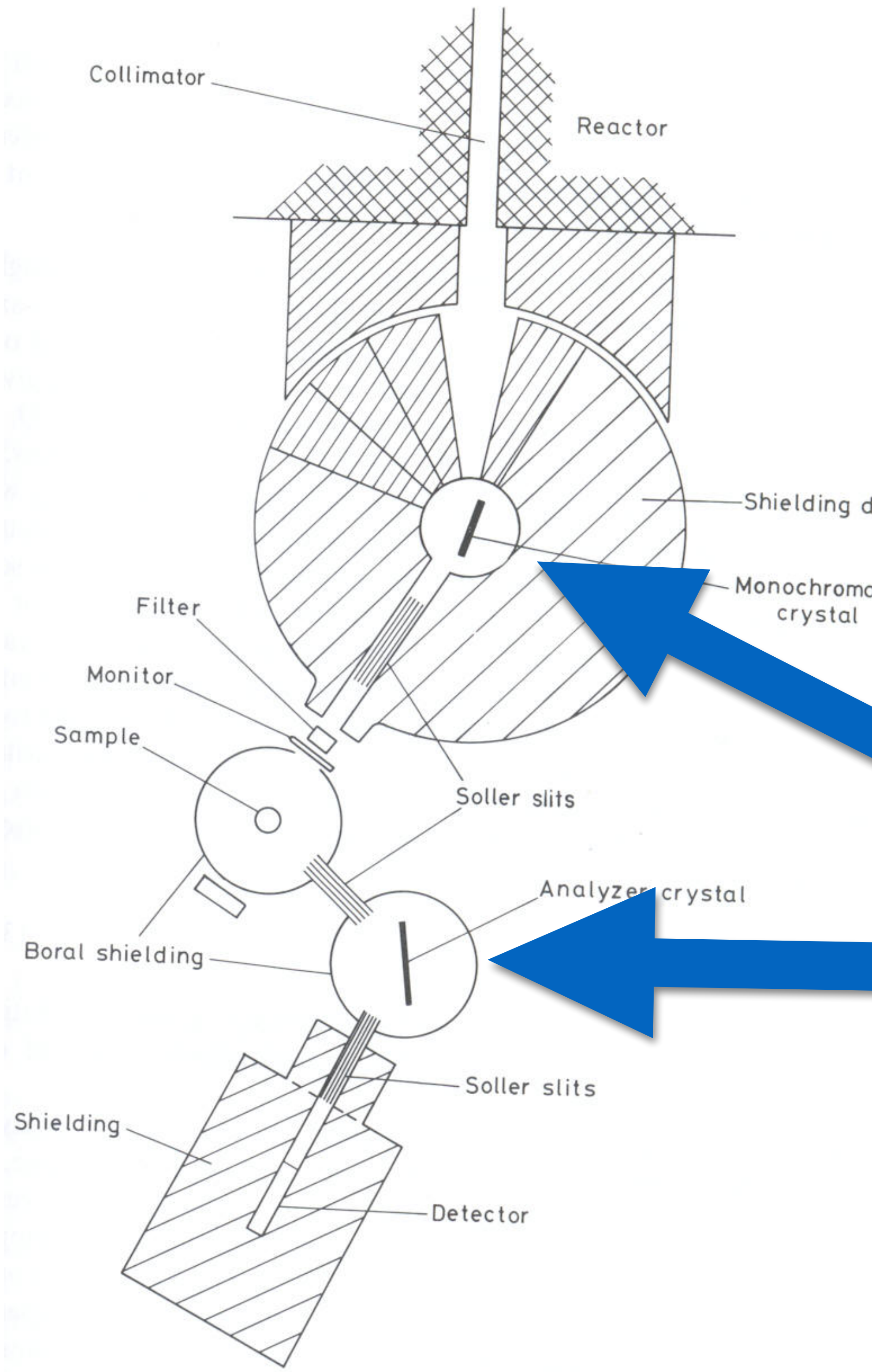
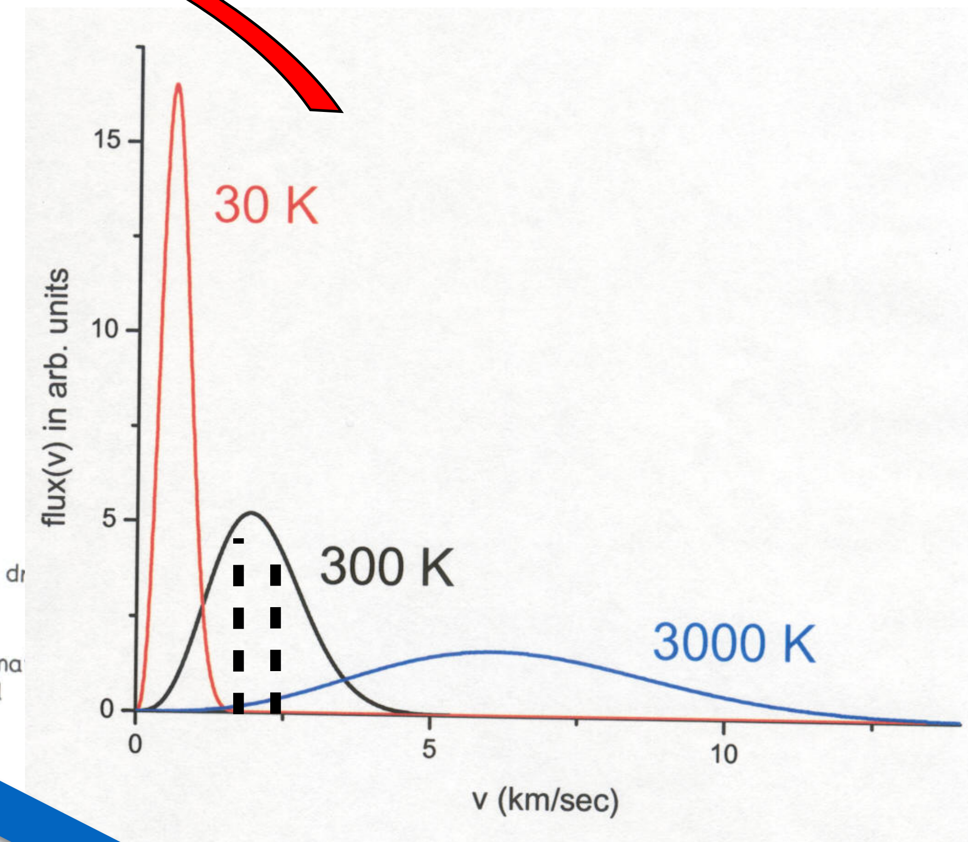
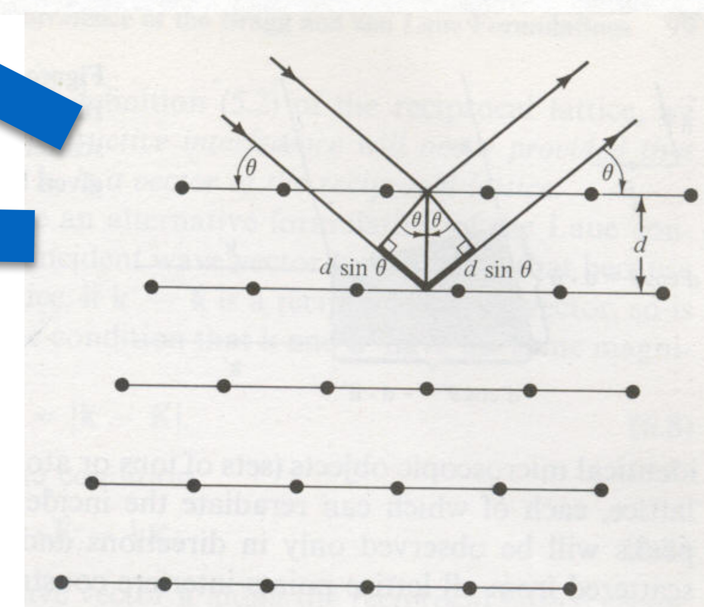
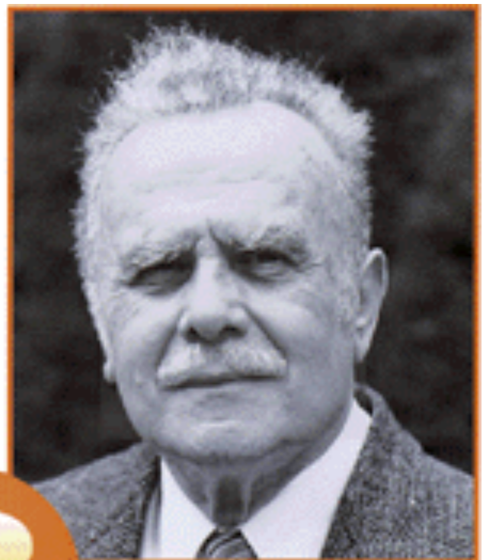






$$n\lambda = 2d\sin\theta$$

Brockhouse's Triple Axis Spectrometer



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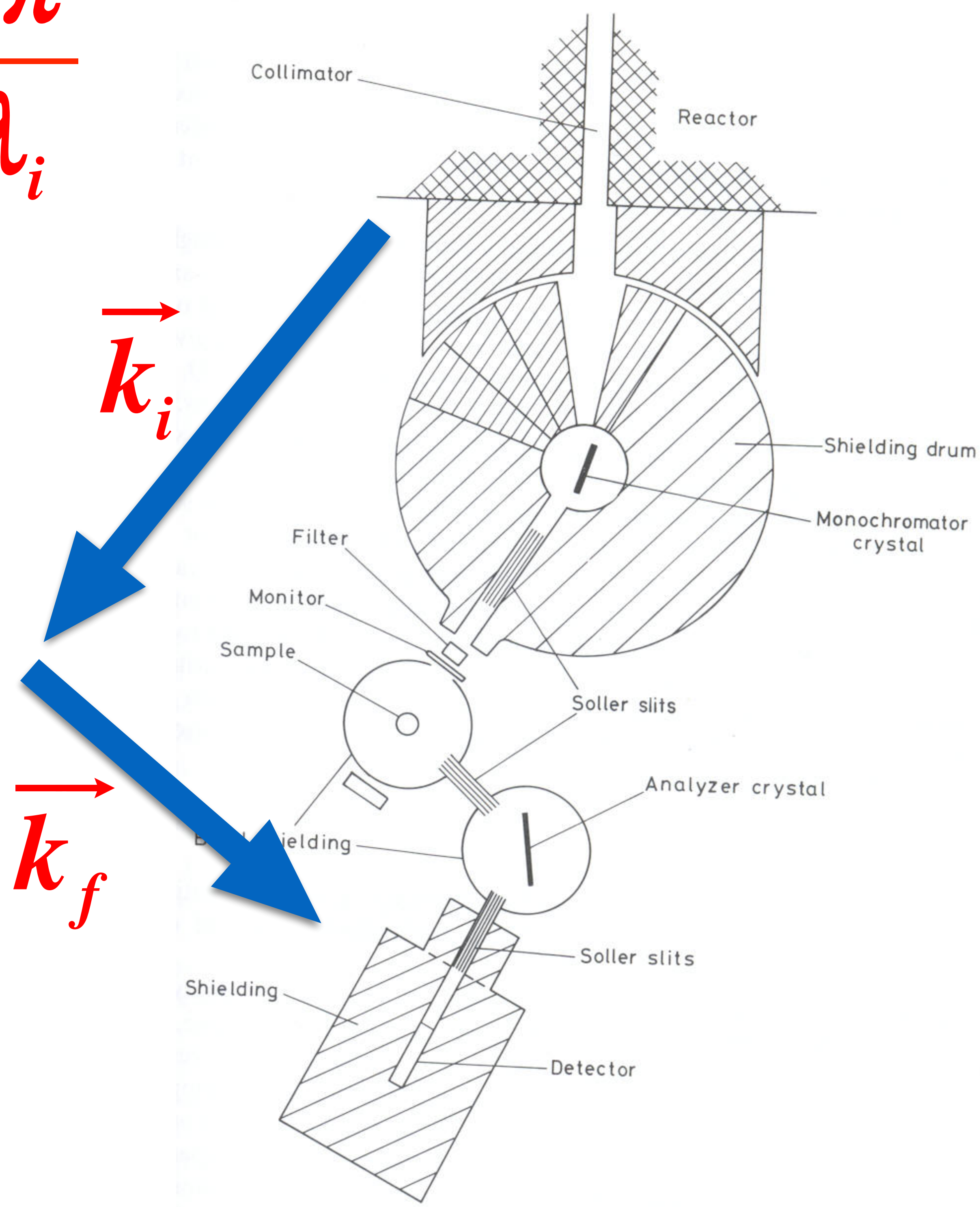
$$|k_i| = \frac{2\pi}{\lambda_i}$$



$$|k_f| = \frac{2\pi}{\lambda_f}$$

Brockhouse's Triple Axis Spectrometer

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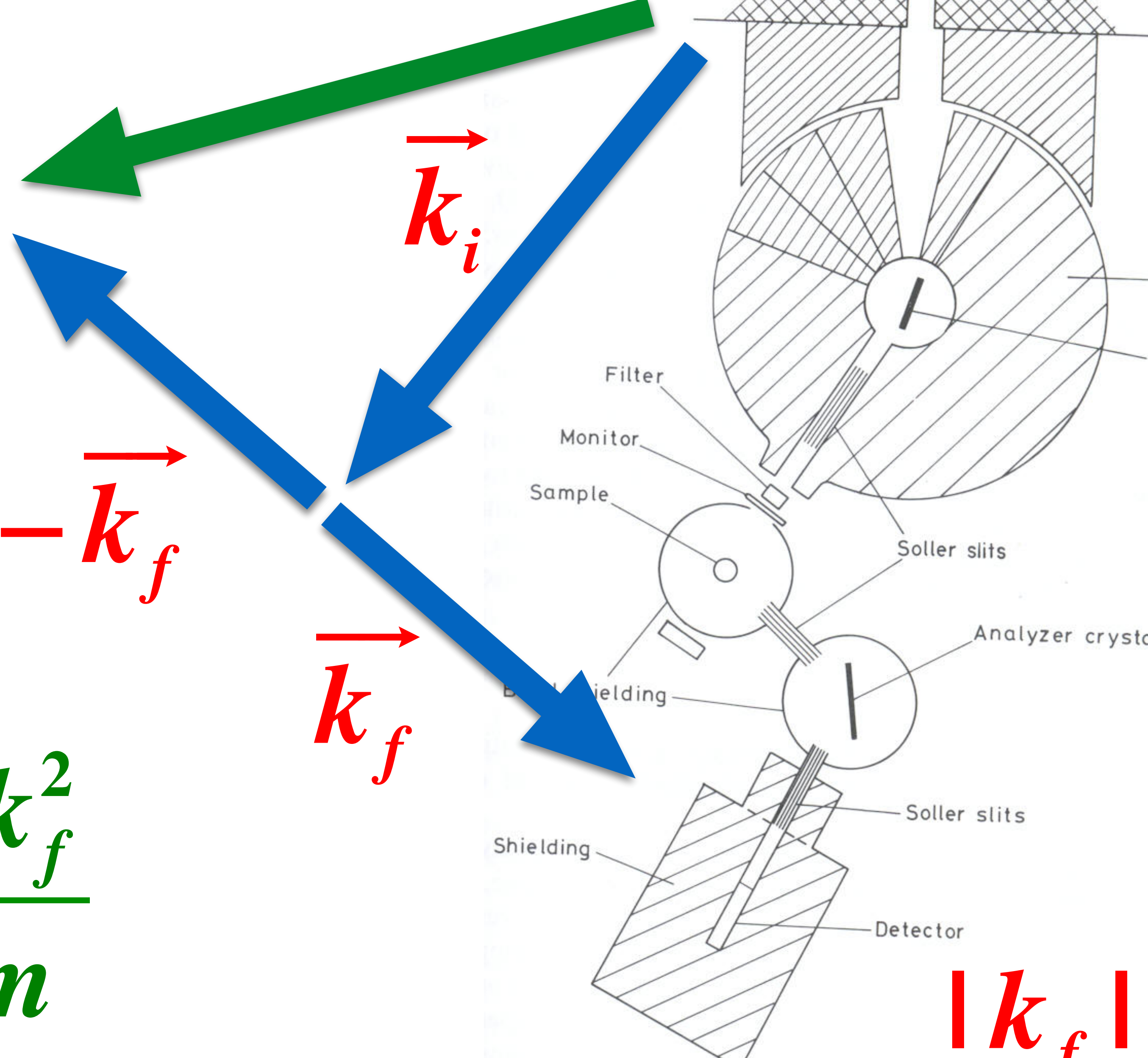
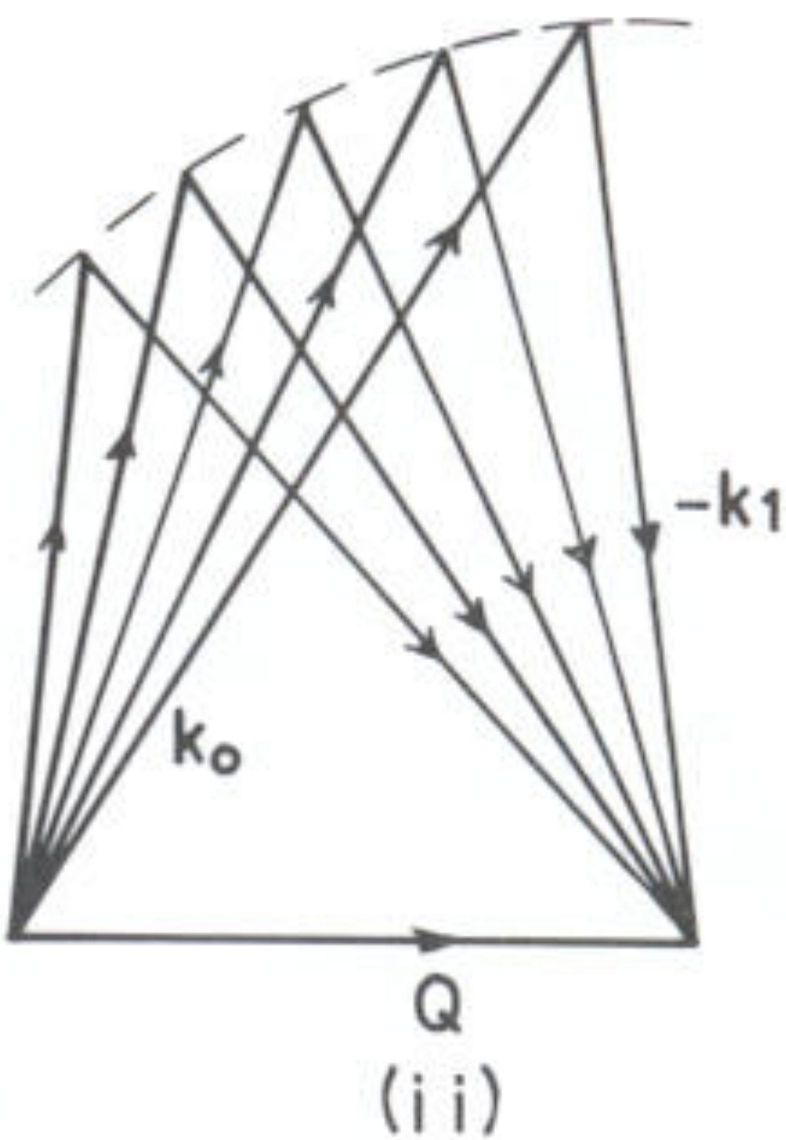


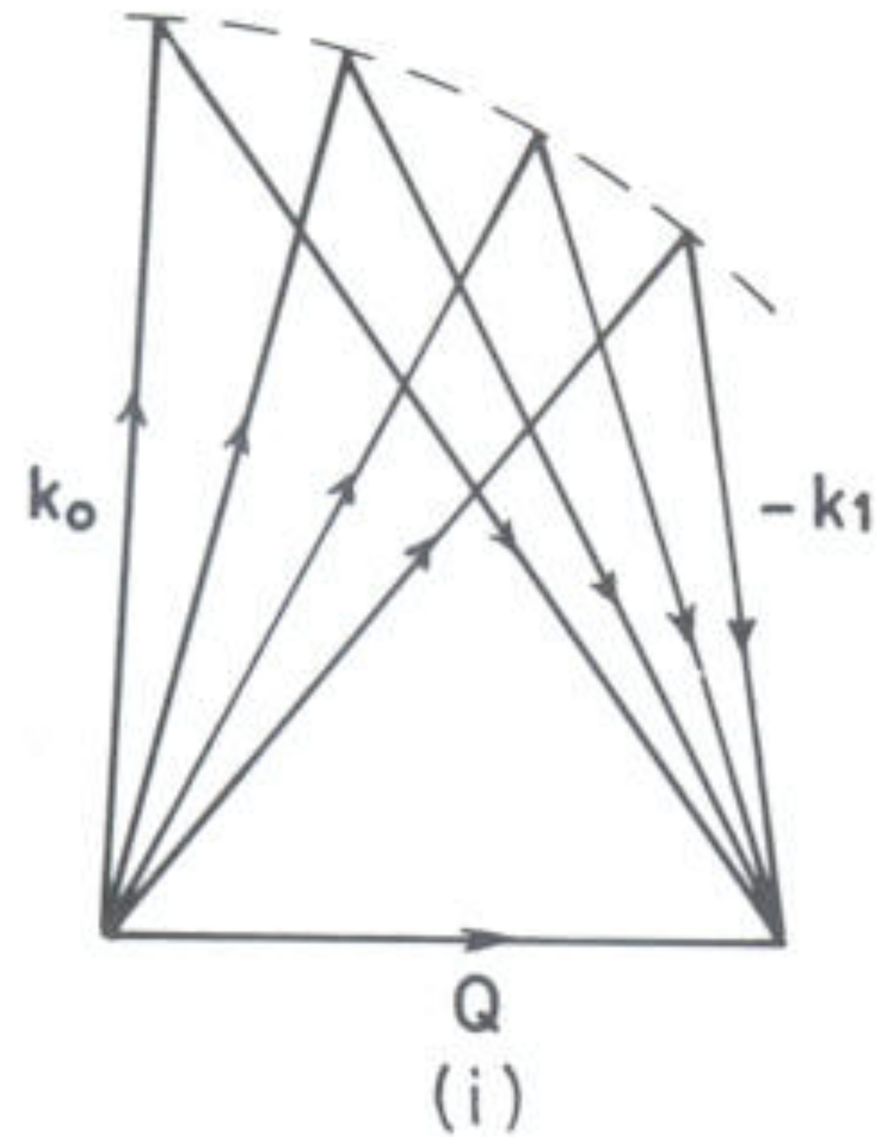
$$|k_f| = \frac{2\pi}{\lambda_f}$$

$$\hbar\omega = \frac{\hbar^2 k_i^2}{2m} - \frac{\hbar^2 k_f^2}{2m}$$

Two different ways of performing constant-Q scans

$Q = k_i - k_f$



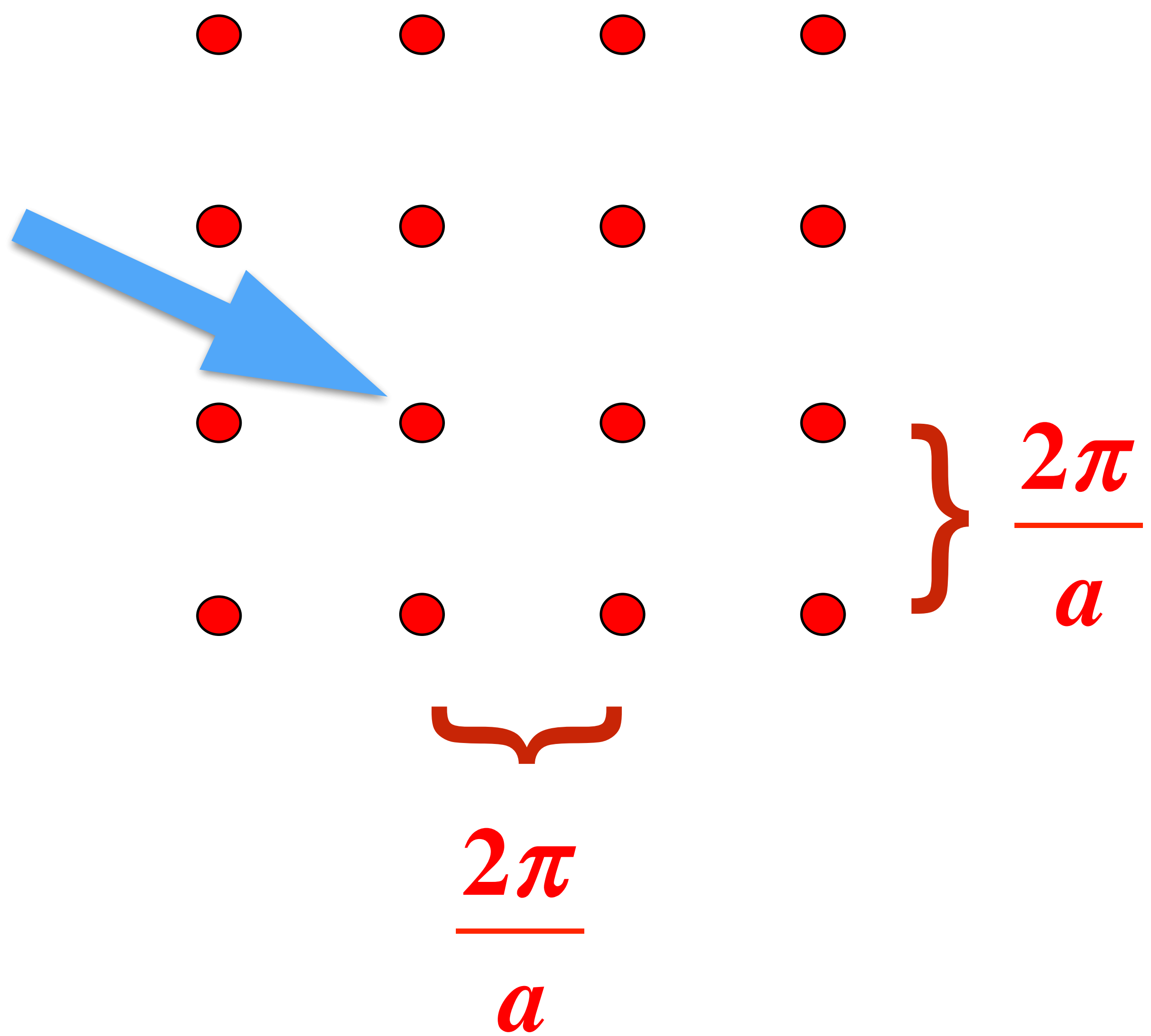


Q = Constant k_f

Q = Constant k_i

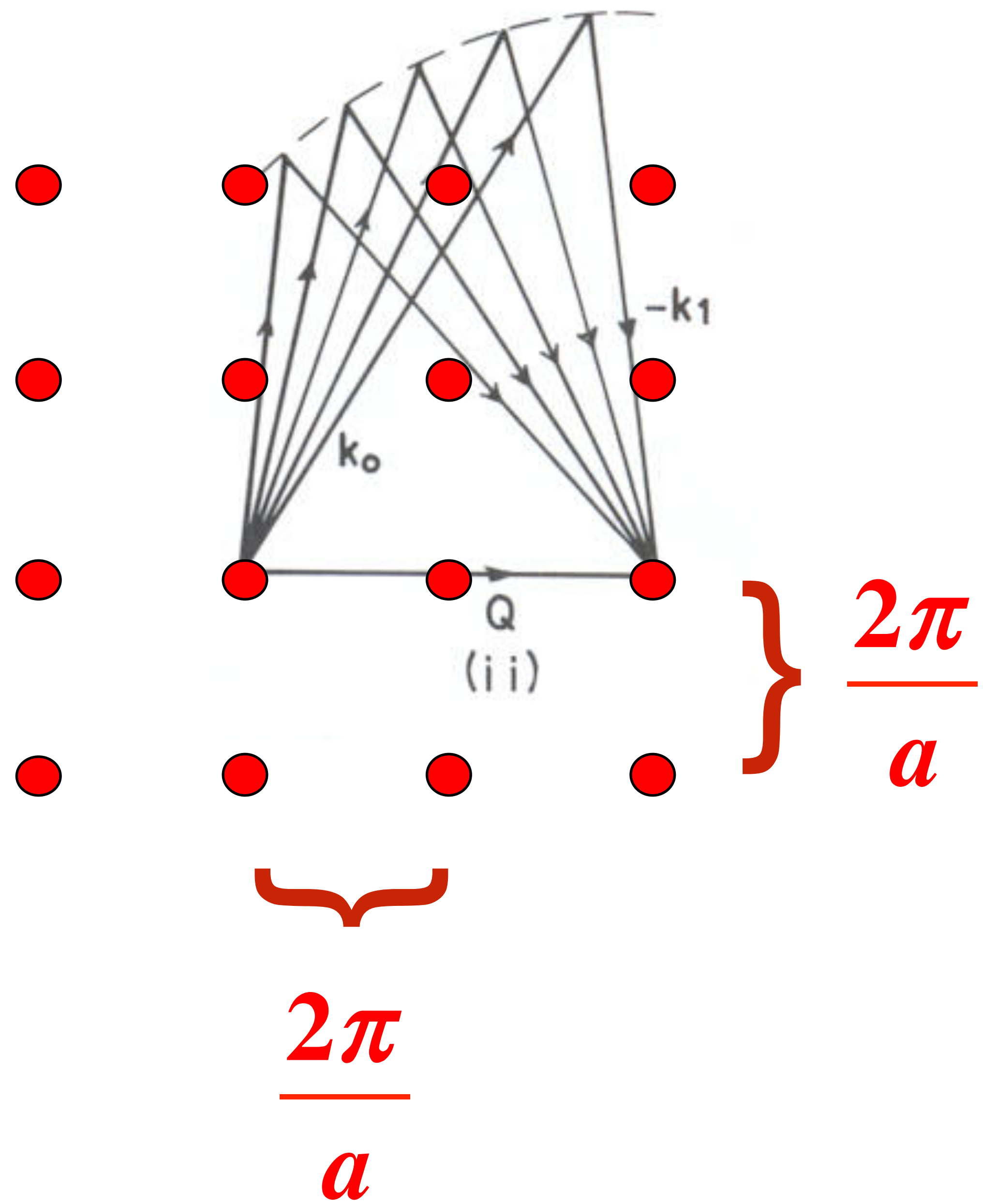
Mapping Momentum (Q) and Energy ($\hbar\omega$) space

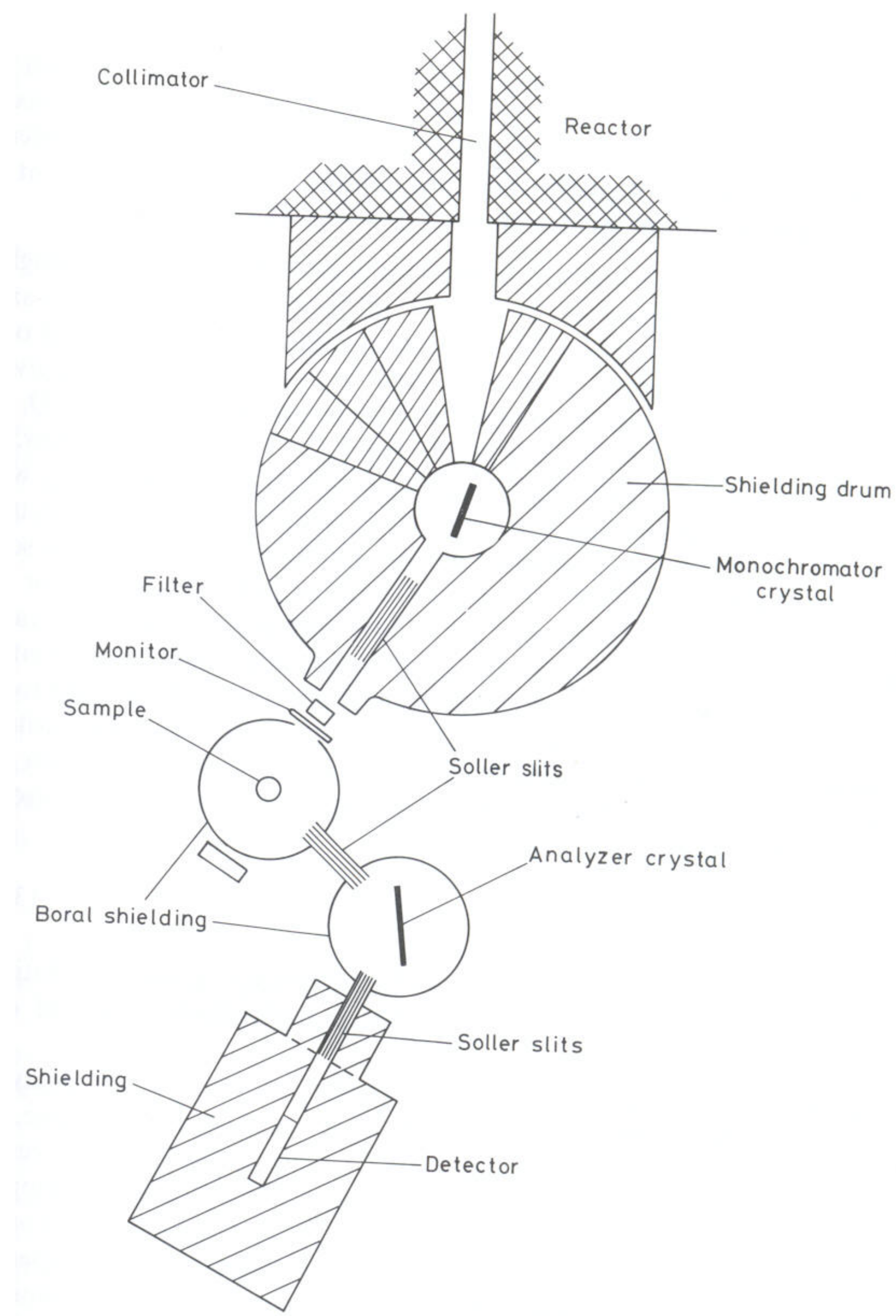
Origin of reciprocal space



Remains fixed for all sample orientations

Putting the Q-map of the scattering with the reciprocal lattice of the crystal





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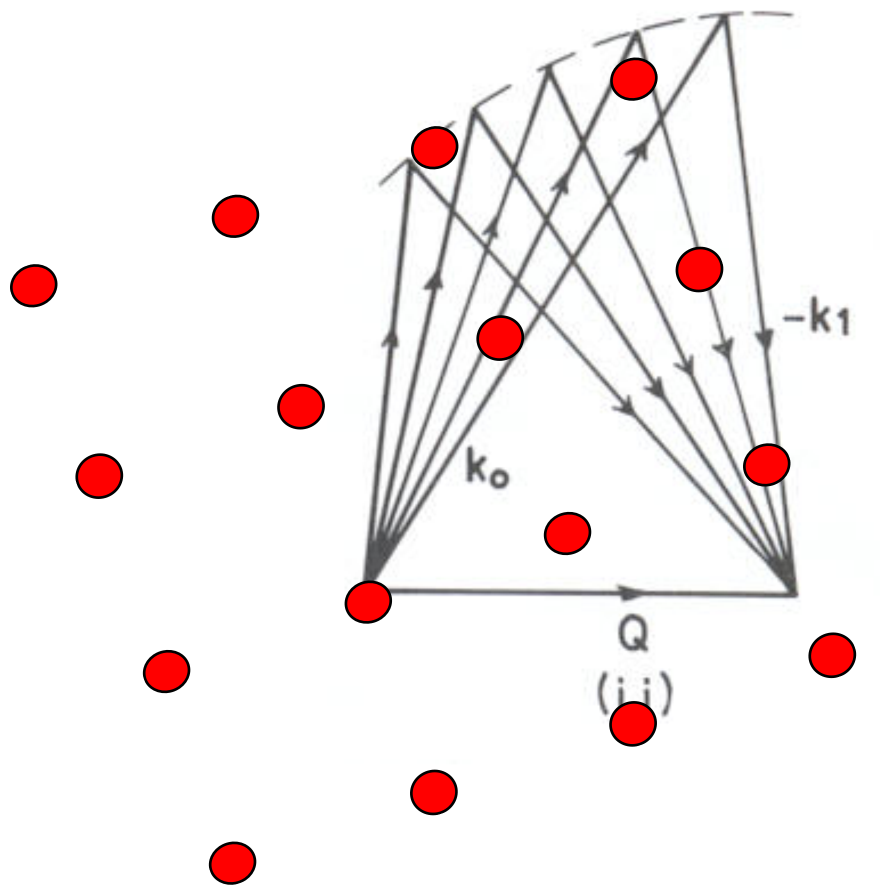
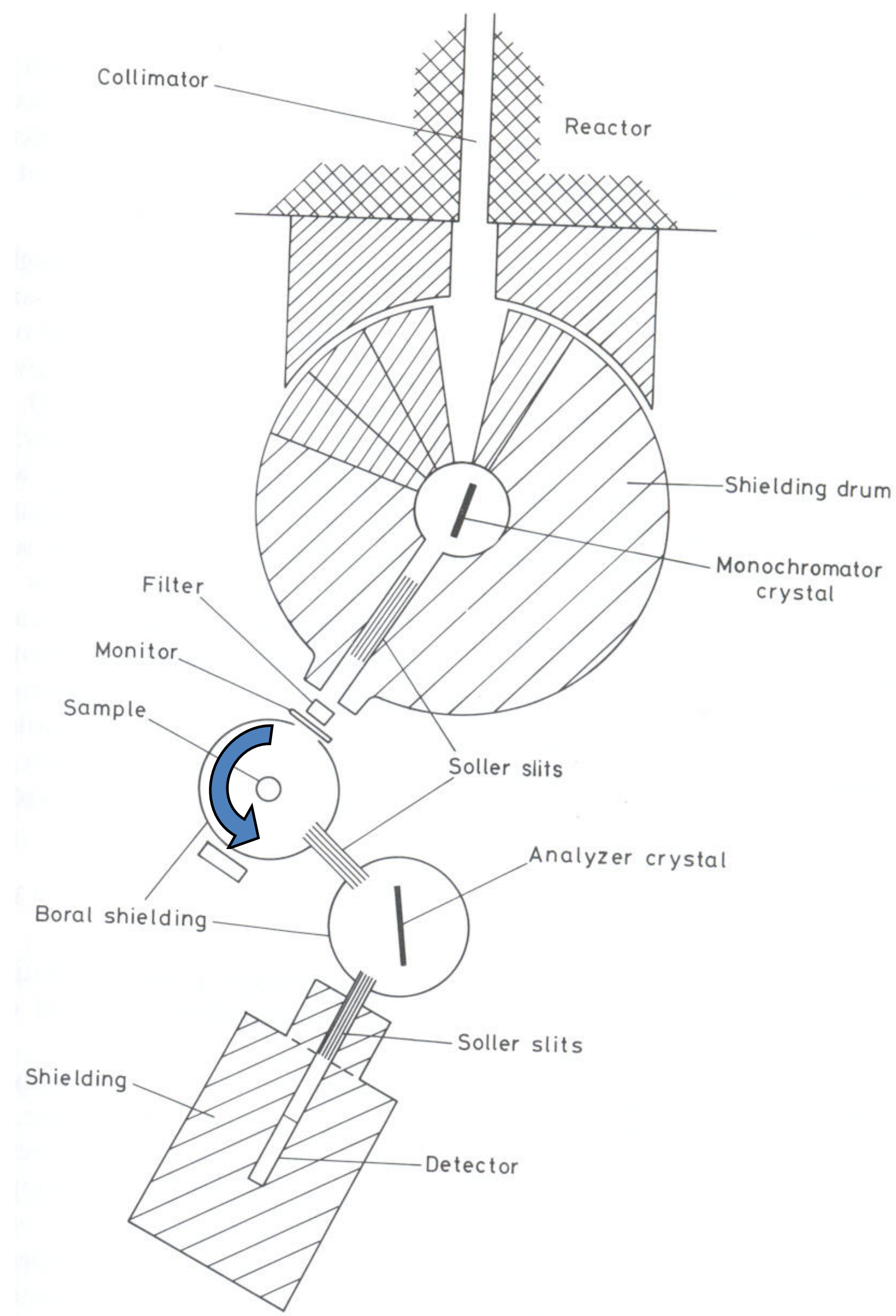
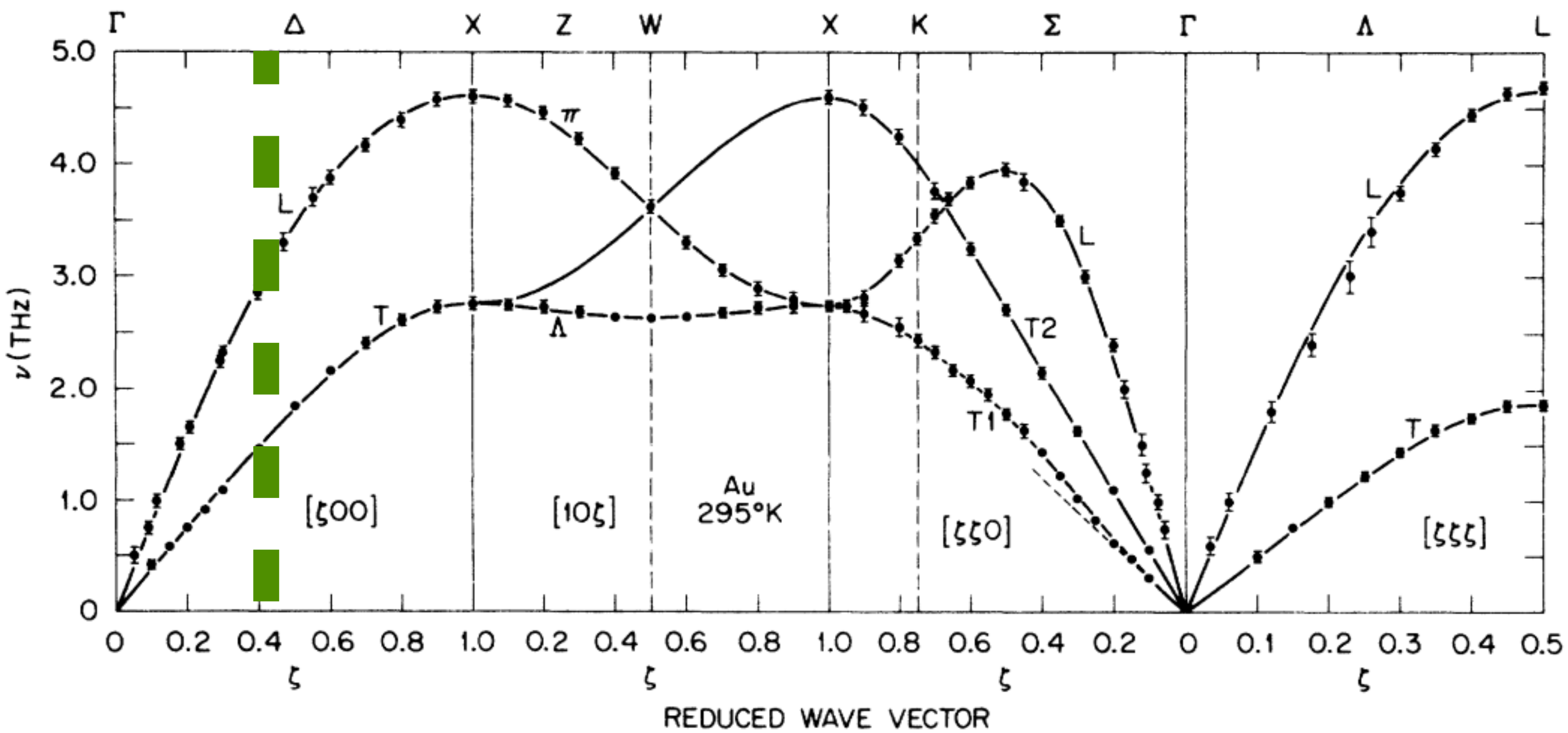
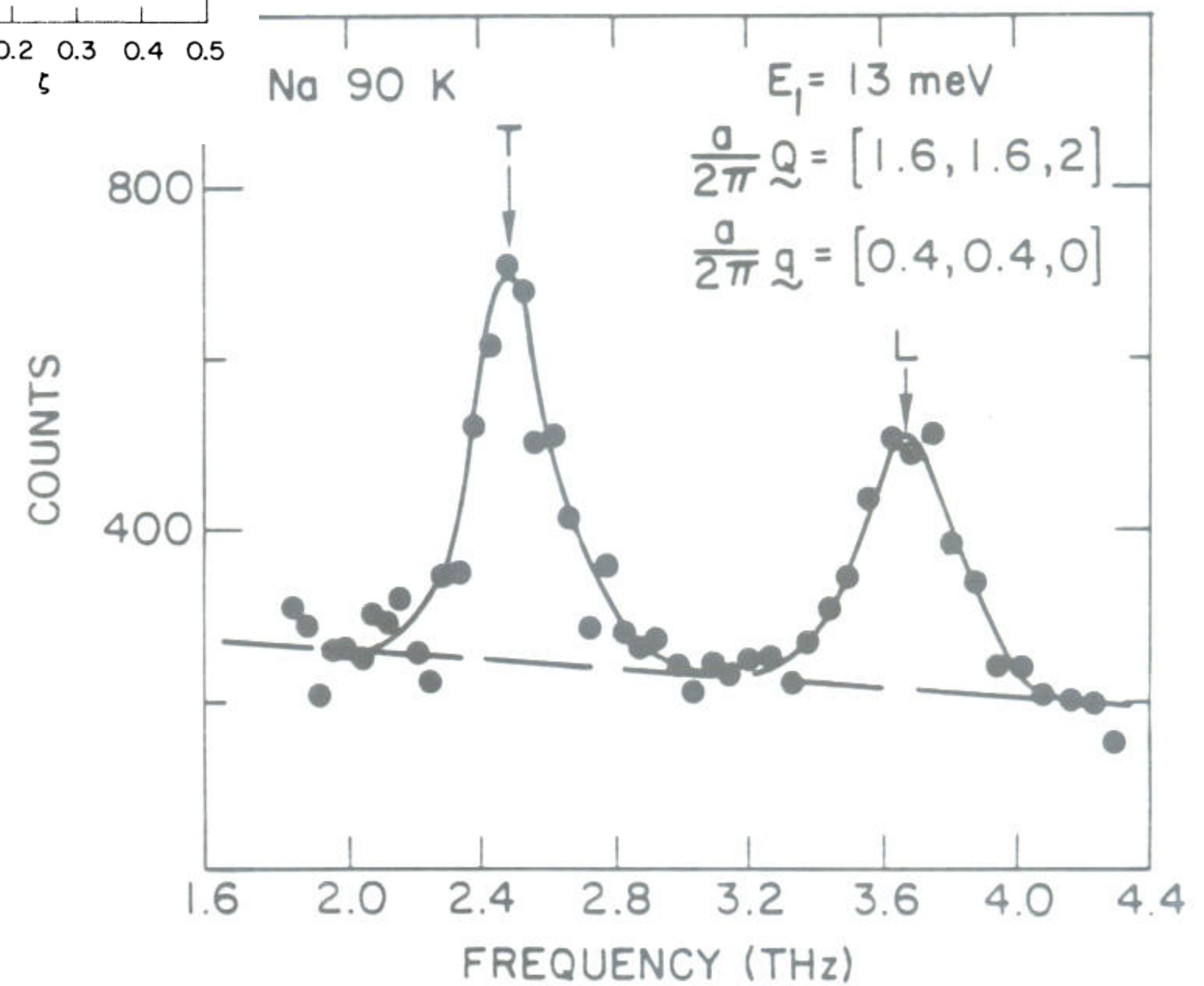




Constant-Q triple axis data

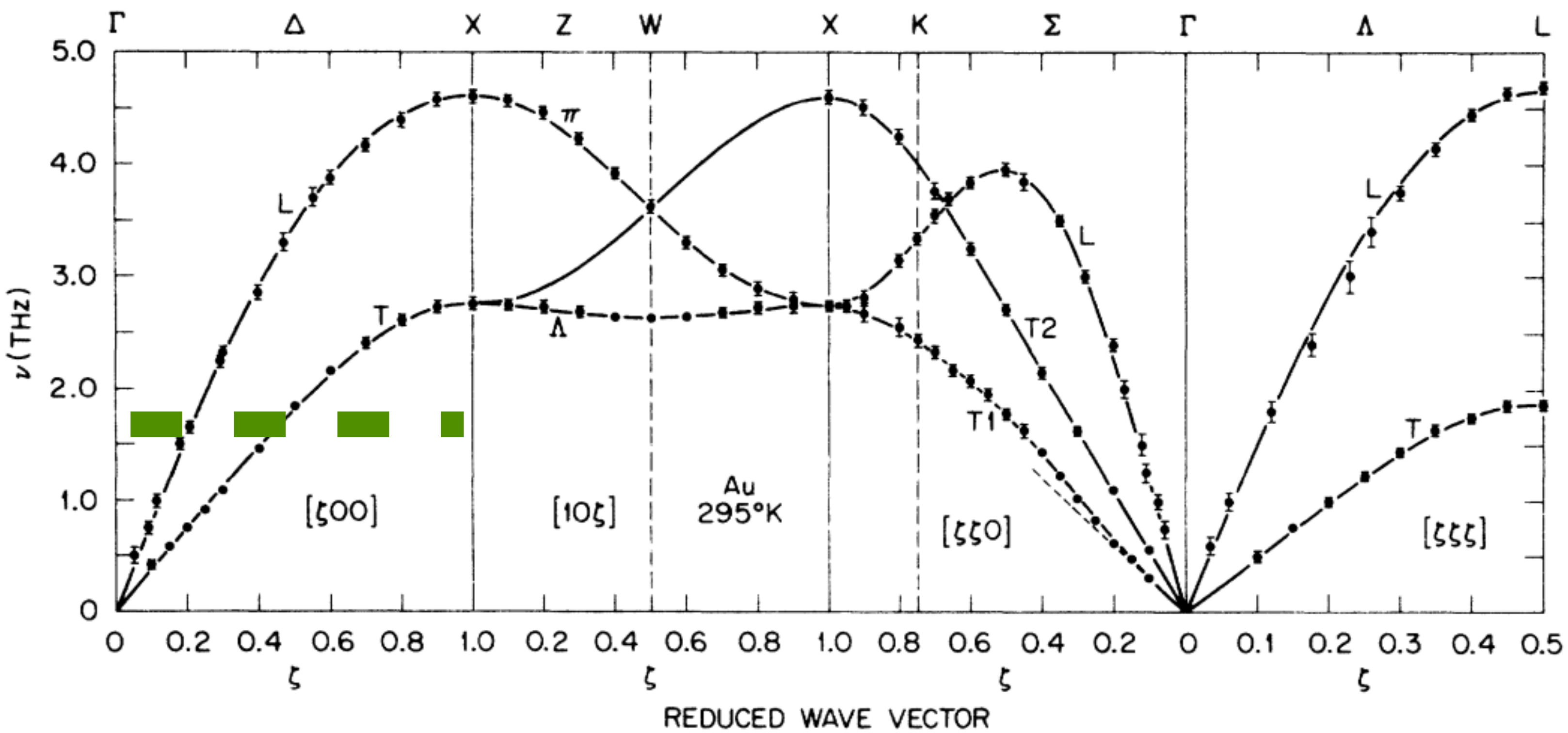


Lynn, *et al.*, *Phys. Rev. B* **8**, 3493 (1973).



Constant **Q**, constant E triple axis techniques allow us to put **Q** and E on a grid, and scan through as we choose

Constant-E triple axis data



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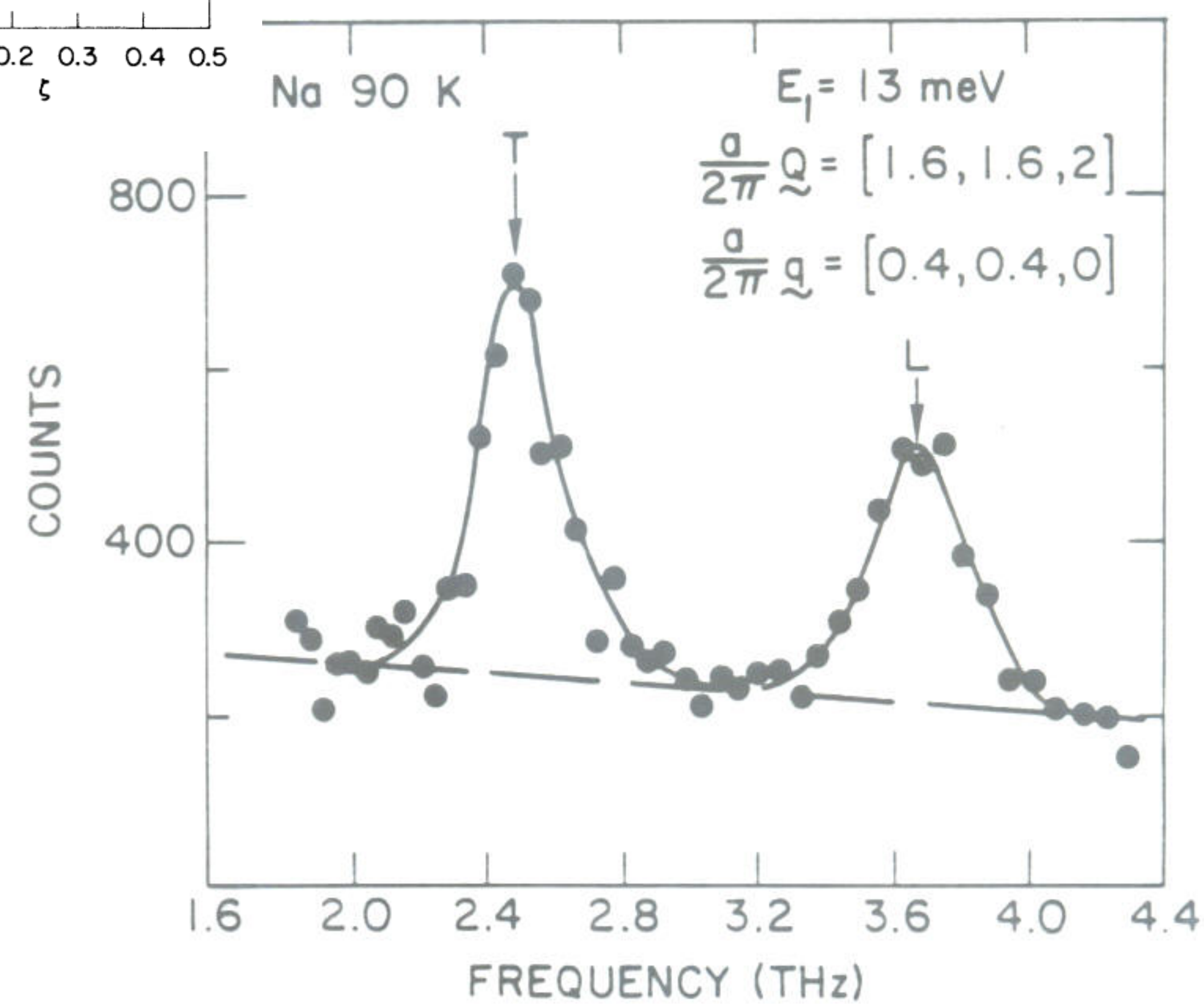
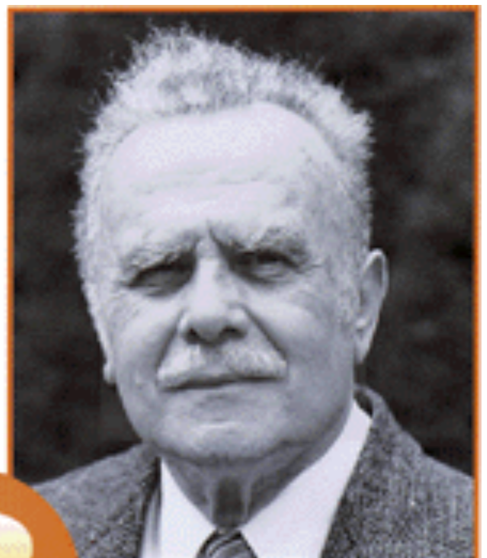


Constant **Q**, constant E triple axis techniques allow us to put **Q** and E on a grid, and scan through as we choose

Elastic scattering with a Triple Axis Spectrometer



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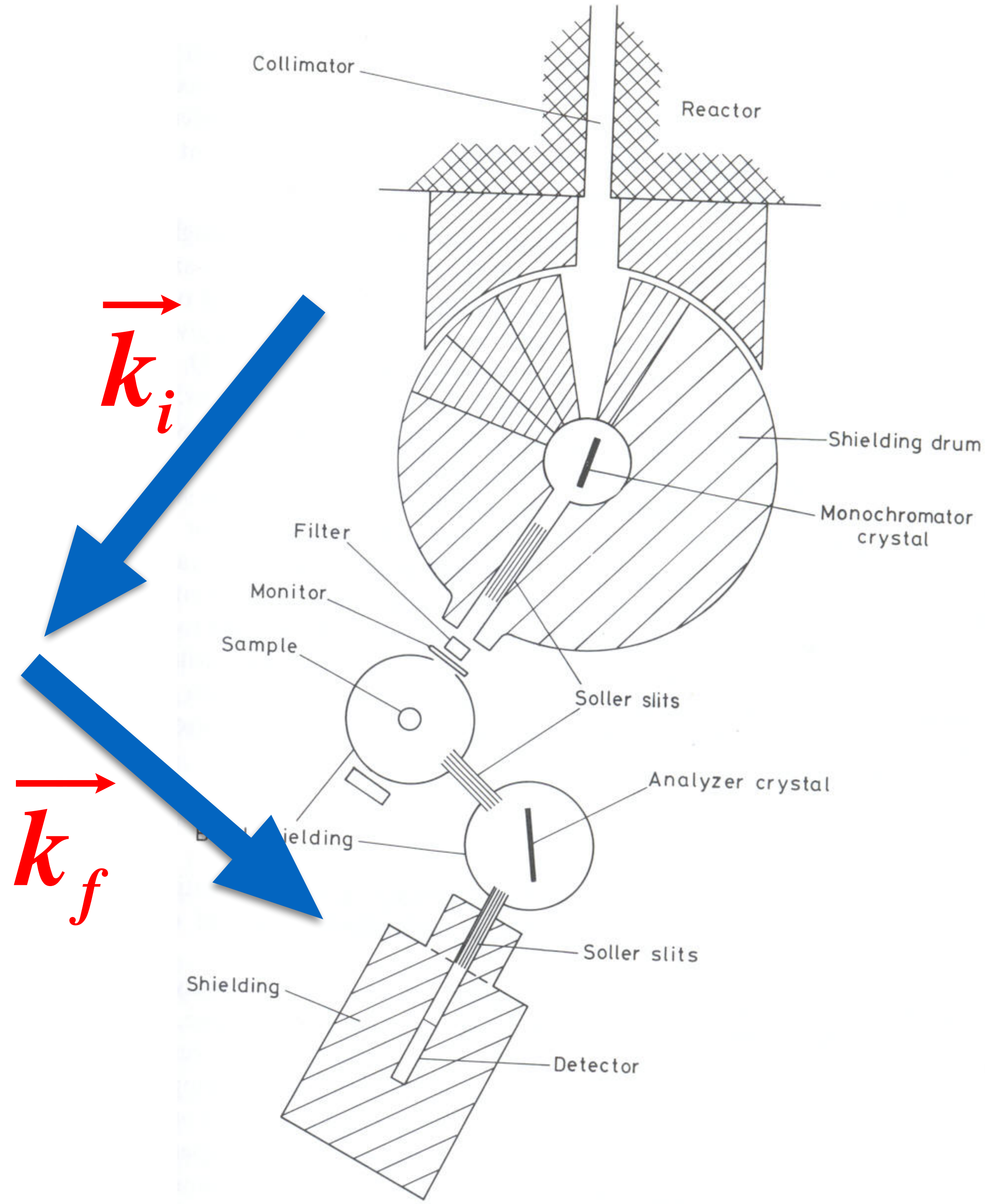
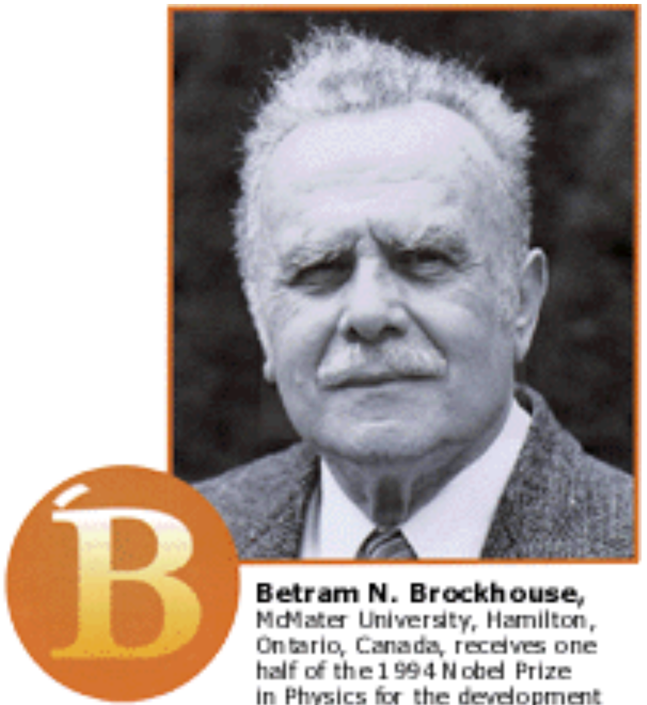


$$|k_f| = |k_i| = \frac{2\pi}{\lambda_i}$$

Two Axis "Spectrometer" integrates over k_f : diffraction



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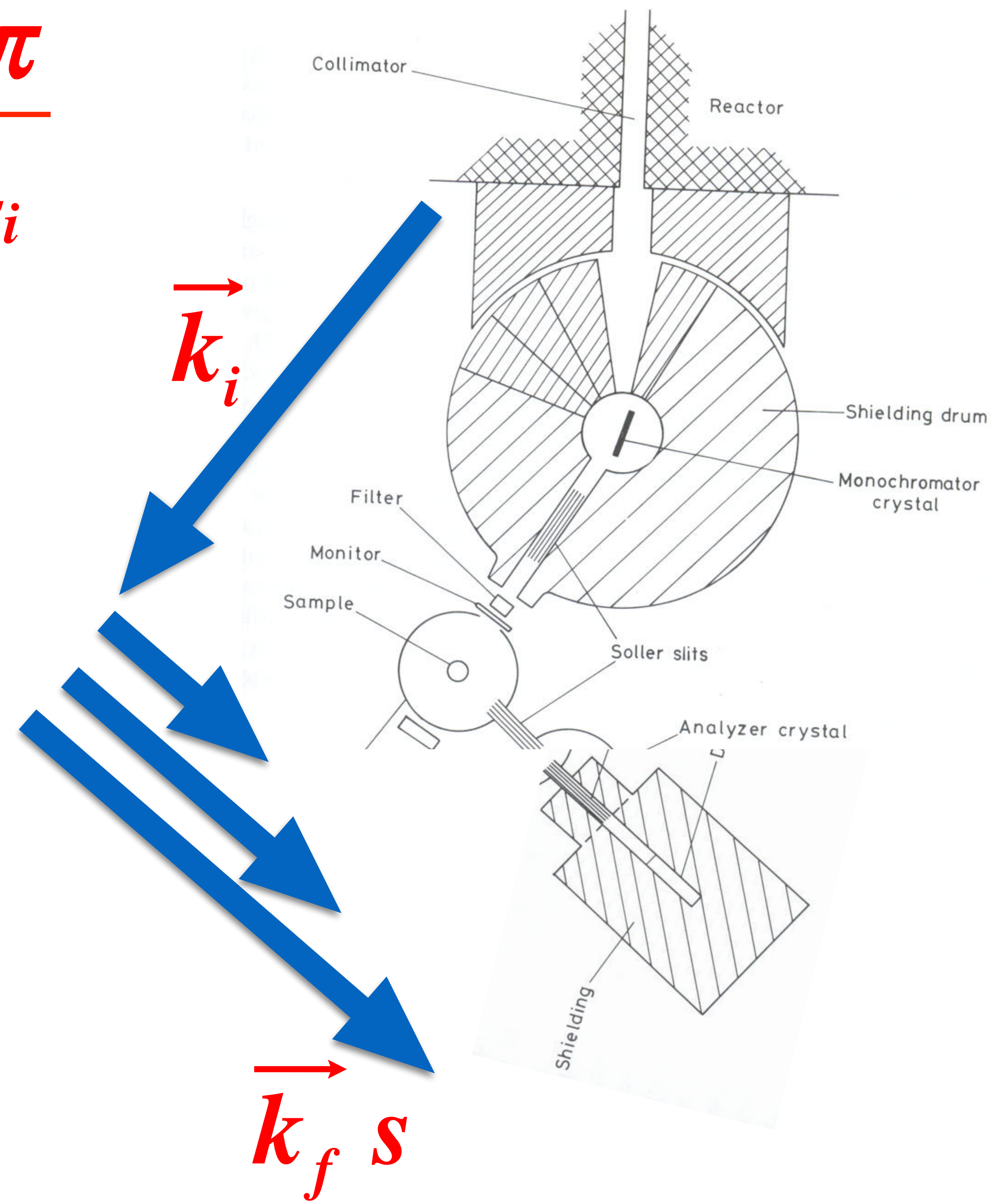


The assumption is often made that the scattering is elastic - but, *this is an assumption!*

The coherent neutron scattering cross section for phonons

$$S(\vec{Q},\hbar\omega) = \frac{1}{2NM} e^{-Q^2\langle u^2 \rangle} \sum_{j,\vec{q}} |\vec{Q}\bullet\vec{\varepsilon}_j(\vec{q})|^2 \frac{1}{\omega_j(\vec{q})}$$

The displacement (eigenvectors) of the atoms
must be // to the momentum transfer

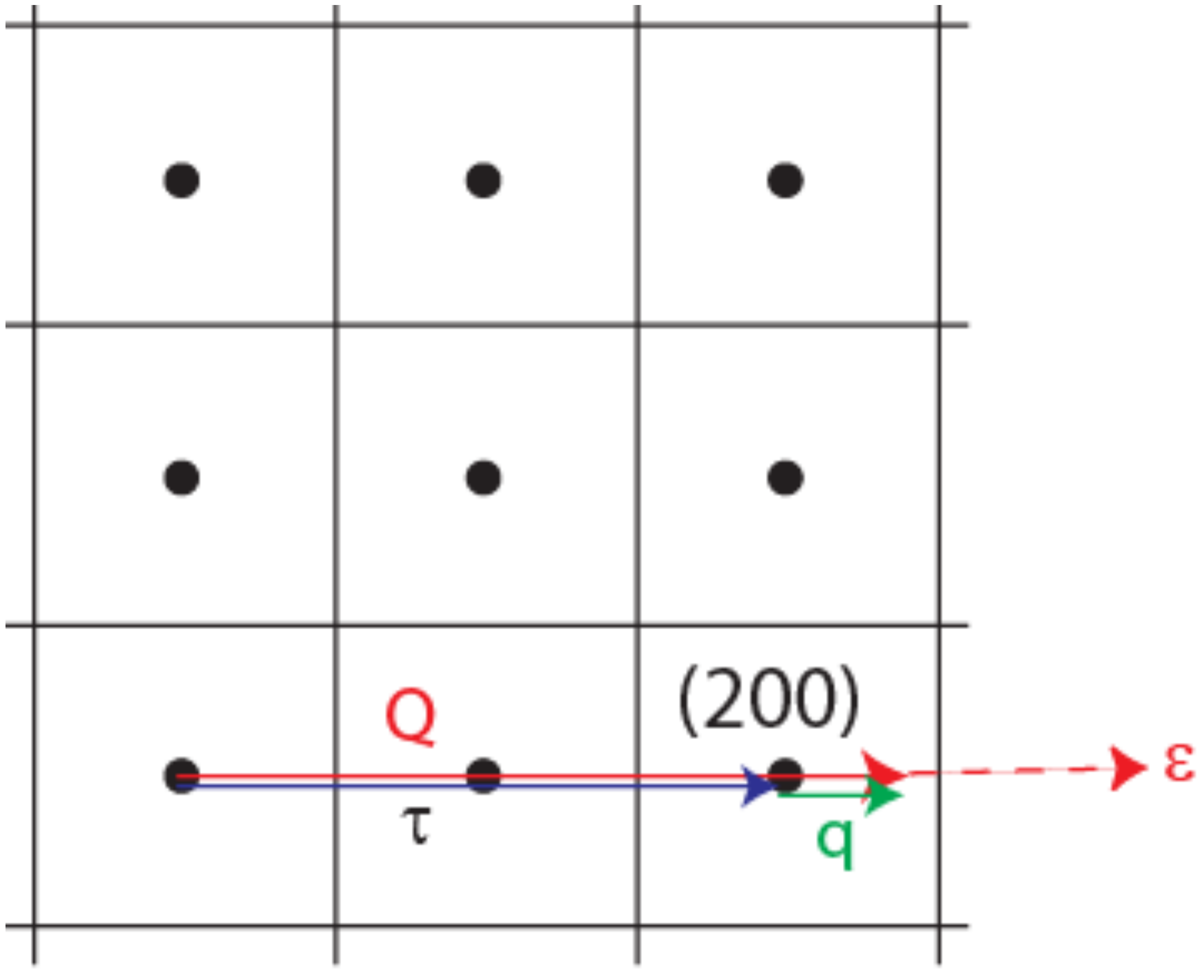
$$\times\ (1+n(\hbar\omega))\ \ \delta(\vec{Q}-\vec{q}-\vec{\tau})\ \ \delta(\hbar\omega-\hbar\omega_j(\vec{q}))$$

The neutron can always create a phonon, but it cannot destroy a phonon unless one is already present

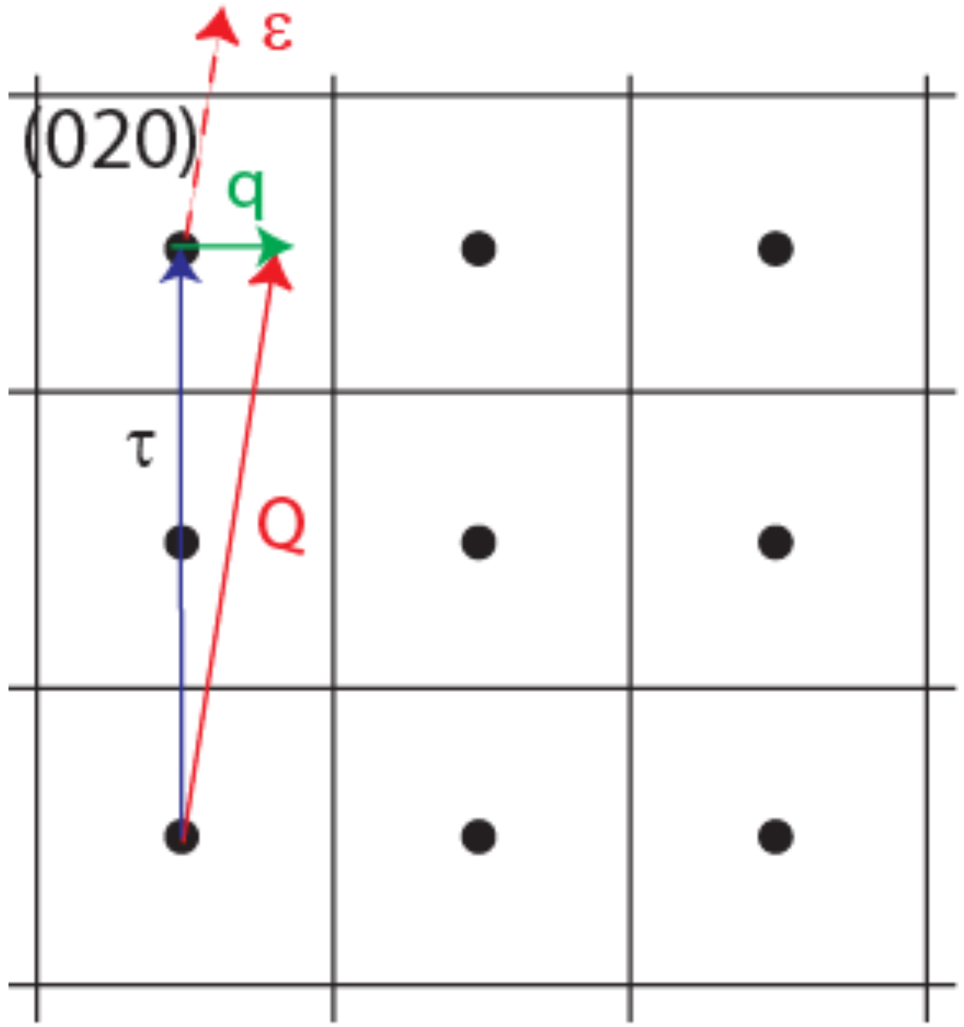
Momentum must be conserved

Energy must be conserved

The coherent neutron scattering cross section for phonons



Longitudinal scan, **q** || ε

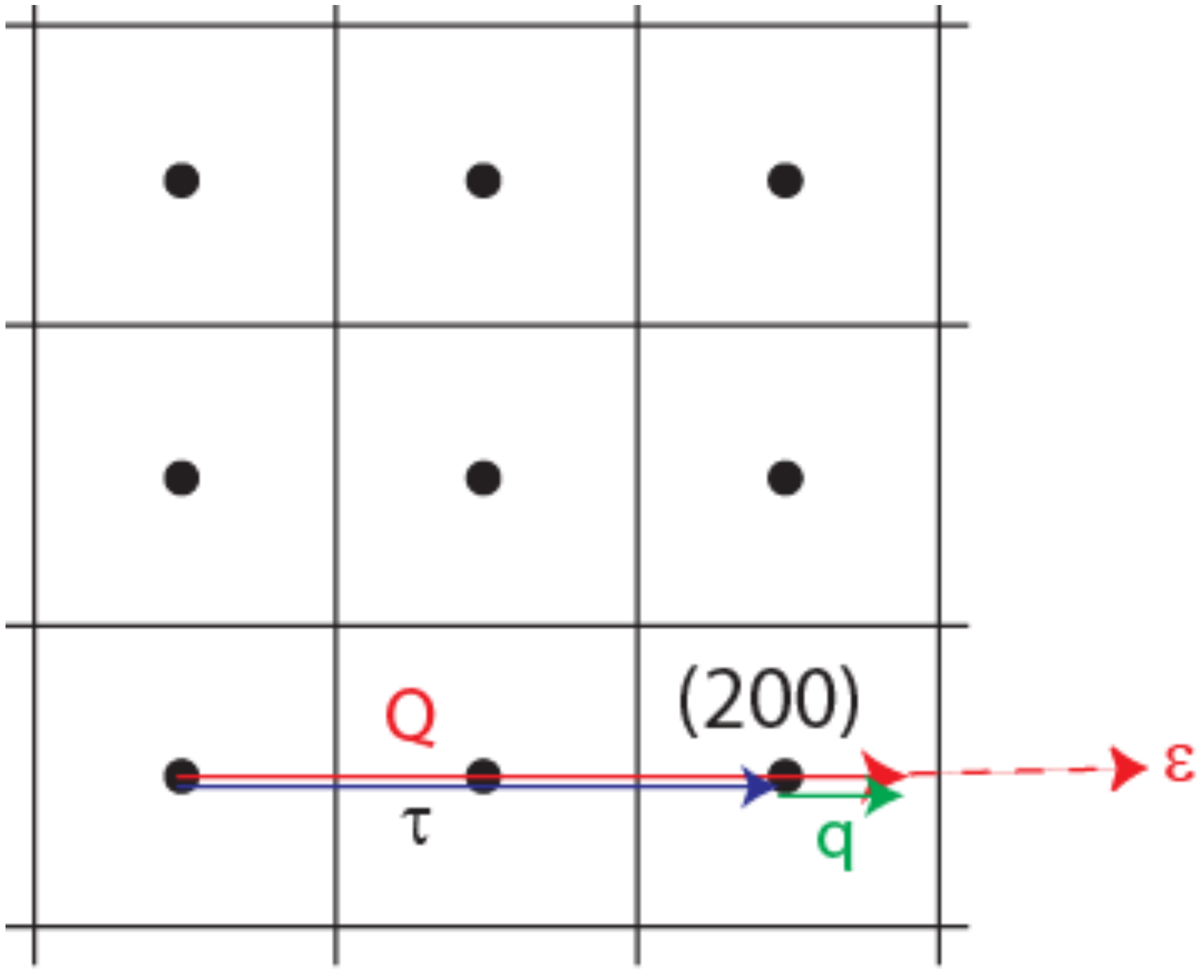


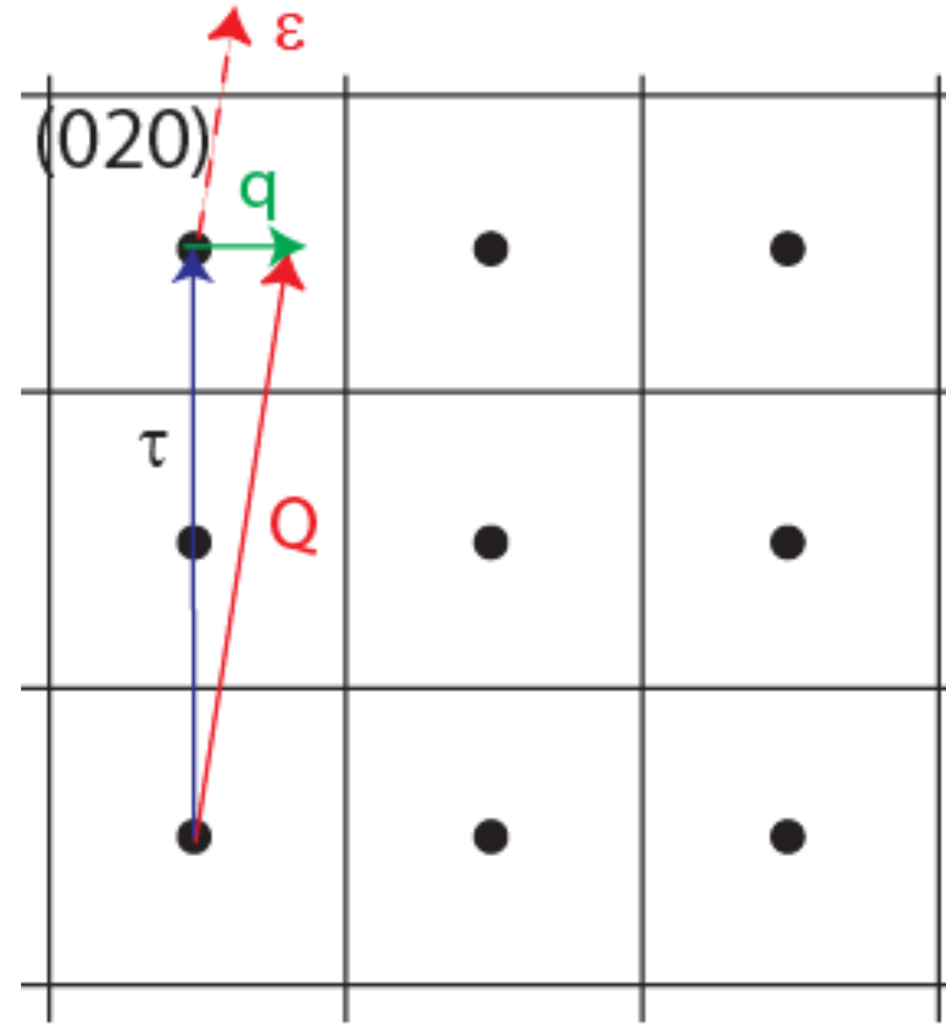
Transverse scan, **q** ⊥ ε

$$S(\vec{Q},\hbar\omega) = \frac{1}{2NM} e^{-Q^2\langle u^2\rangle} \sum_{j,\vec{q}} |\vec{Q}\bullet\vec{\varepsilon}_j(\vec{q})|^2 \frac{1}{\omega_j(\vec{q})}$$

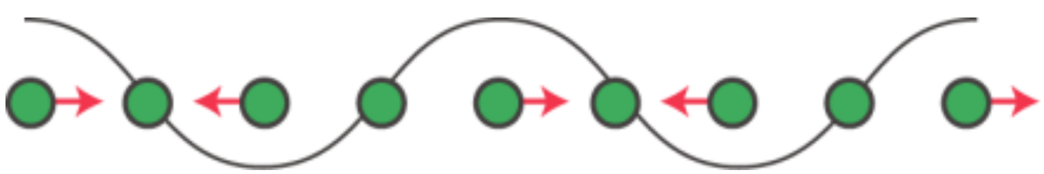
$$\times \ (1+n(\hbar\omega)) \ \ \delta(\vec{Q}-\vec{q}-\vec{\tau}) \ \ \delta(\hbar\omega-\hbar\omega_j(\vec{q}))$$

The coherent neutron scattering cross section for phonons





Longitudinal scan, **q** || ε



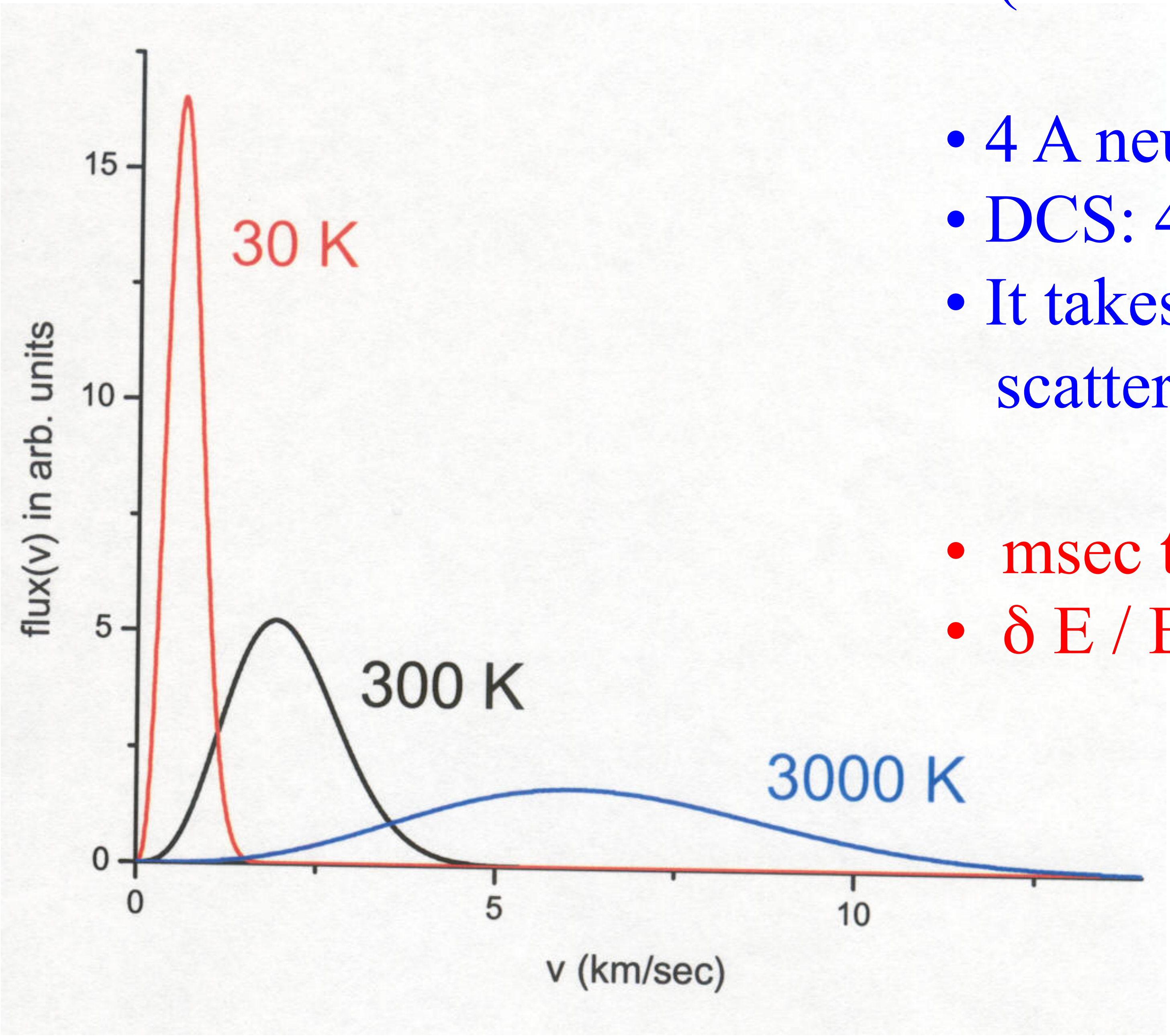
Transverse scan, **q** ⊥ ε

$$S(\vec{Q},\hbar\omega) = \frac{1}{2NM} e^{-Q^2\langle u^2\rangle} \sum_{j,\vec{q}} |\vec{Q}\bullet\vec{\varepsilon}_j(\vec{q})|^2 \frac{1}{\omega_j(\vec{q})}$$

Time-of-flight Neutron Scattering

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

v (km/sec) = 3.96 / λ (A)

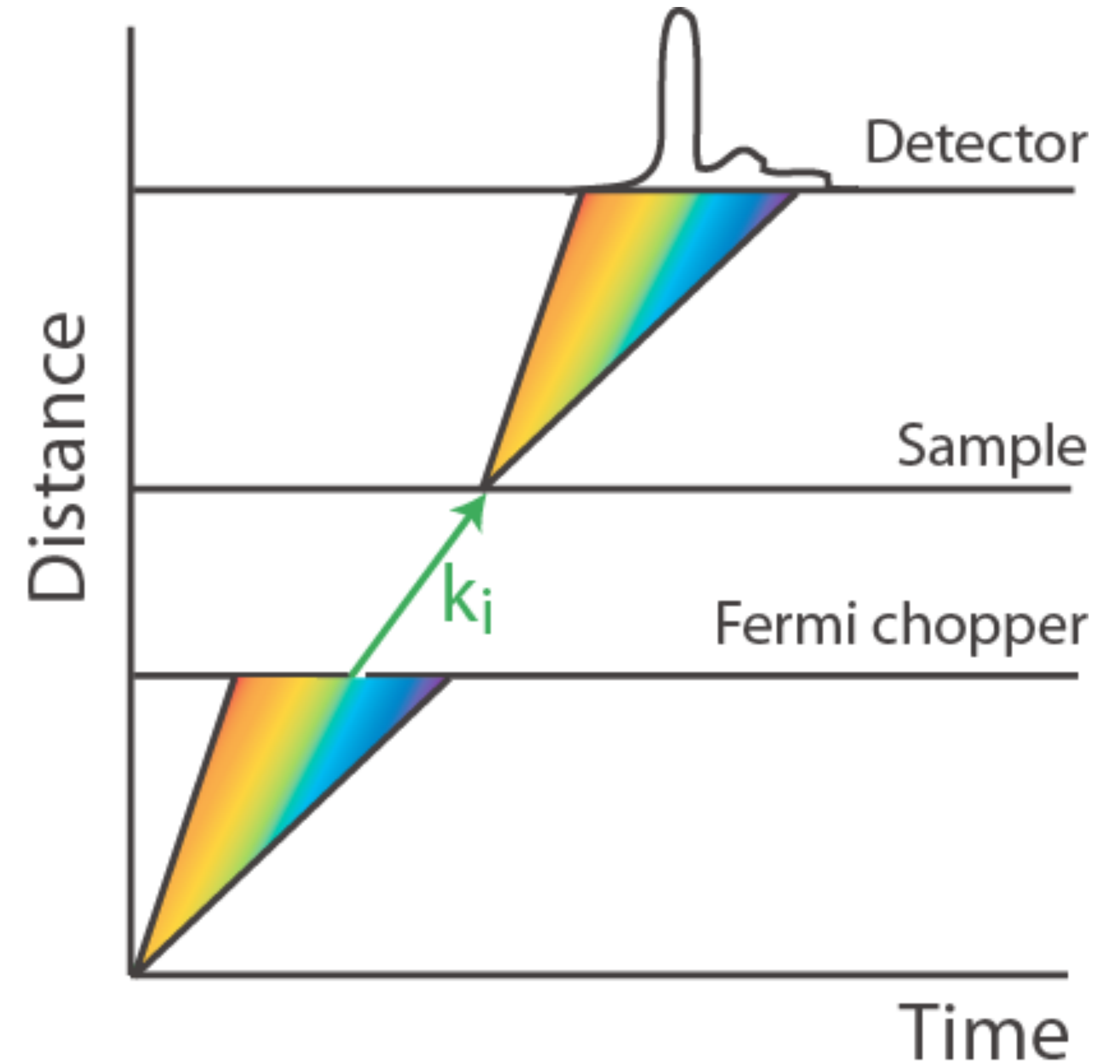


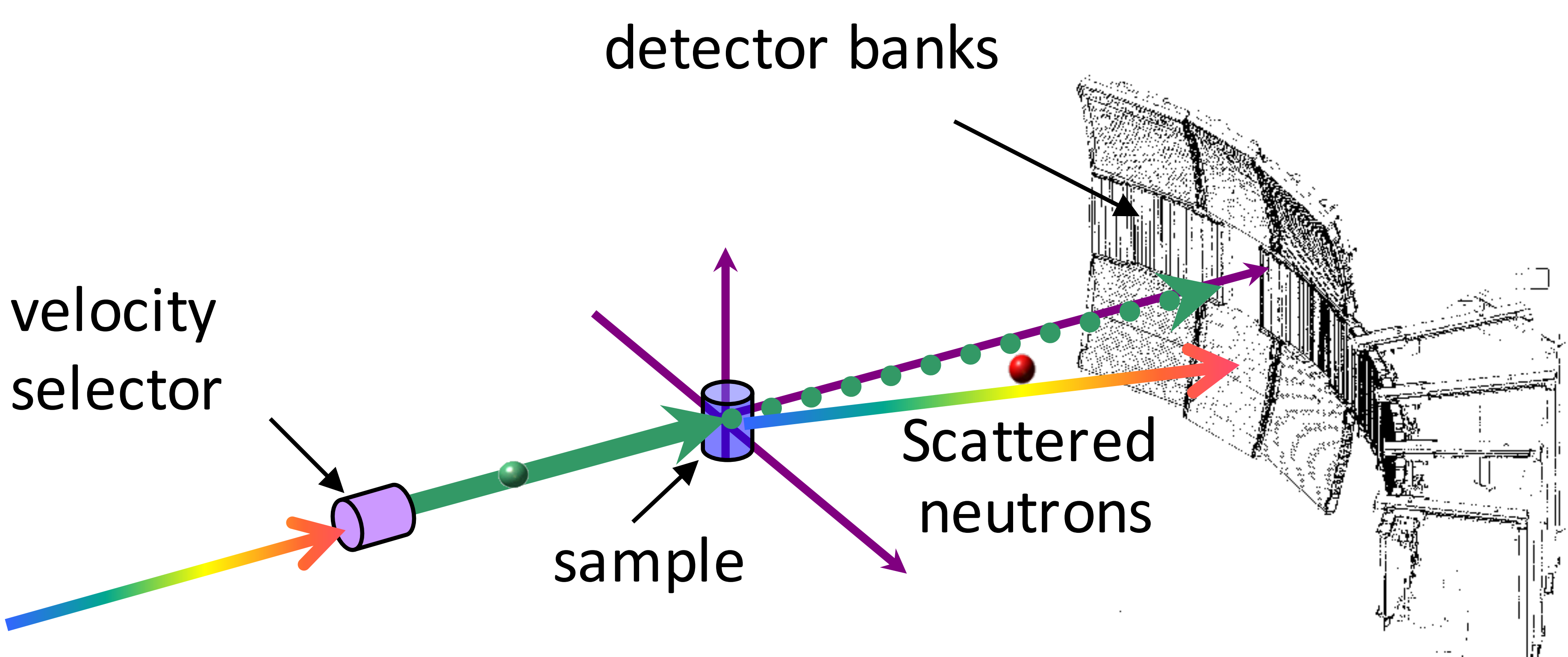
- 4 A neutrons move at ~ 1 km/sec
- DCS: 4 m from sample to detector
- It takes 4 msec for elastically scattered 4 A neutrons to travel 4 m

- 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*

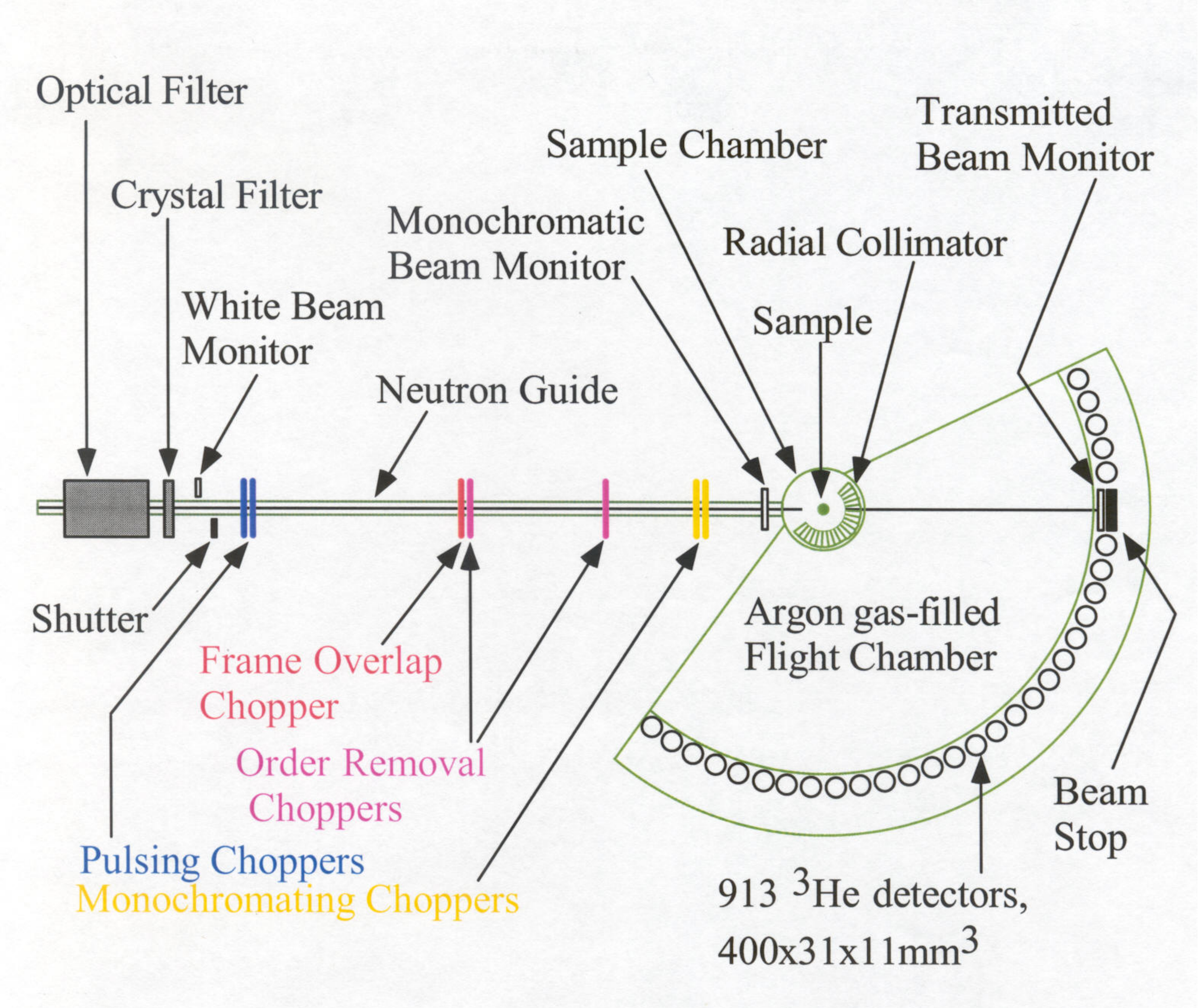
Time-of-flight Neutron Scattering



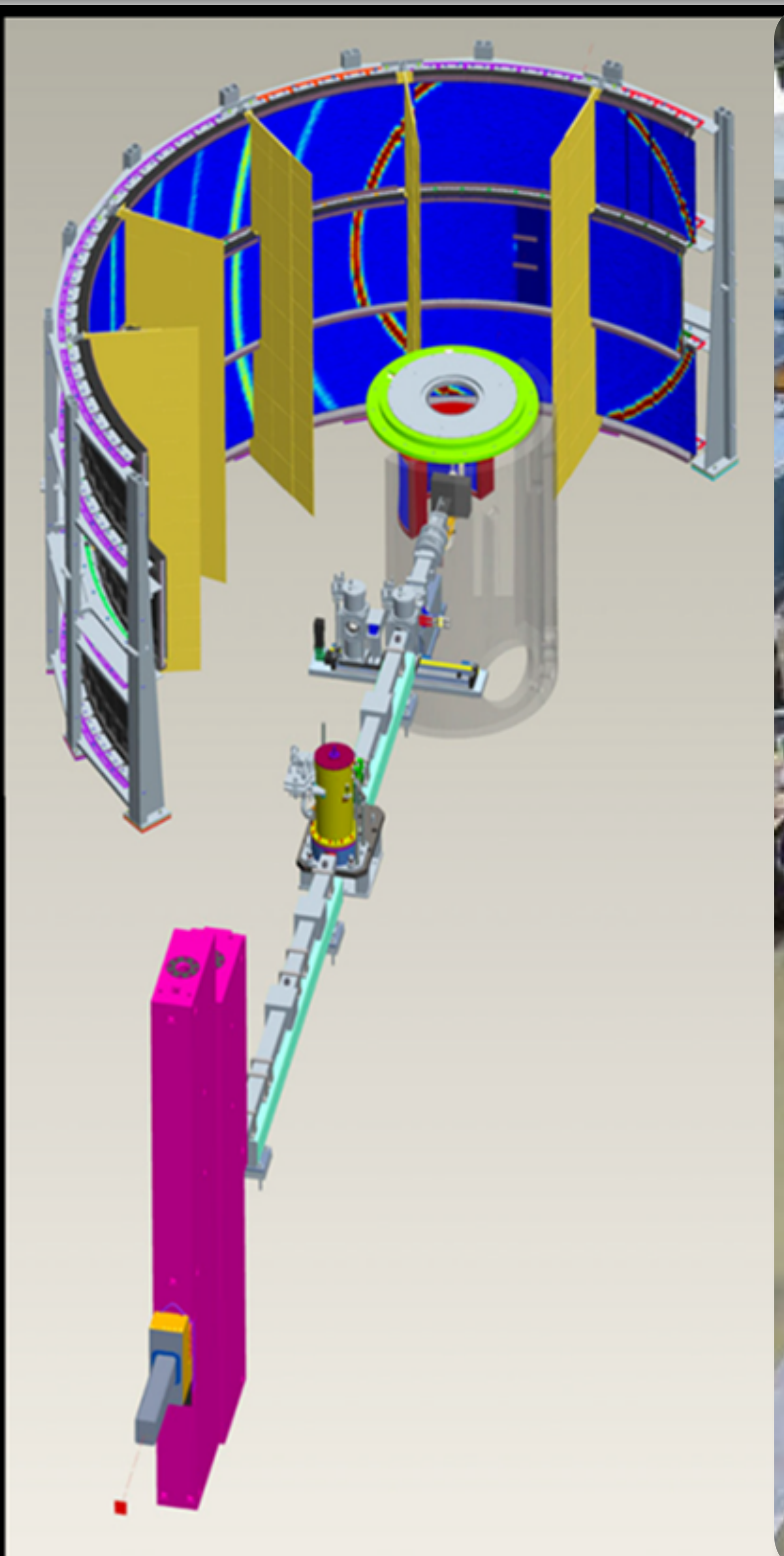


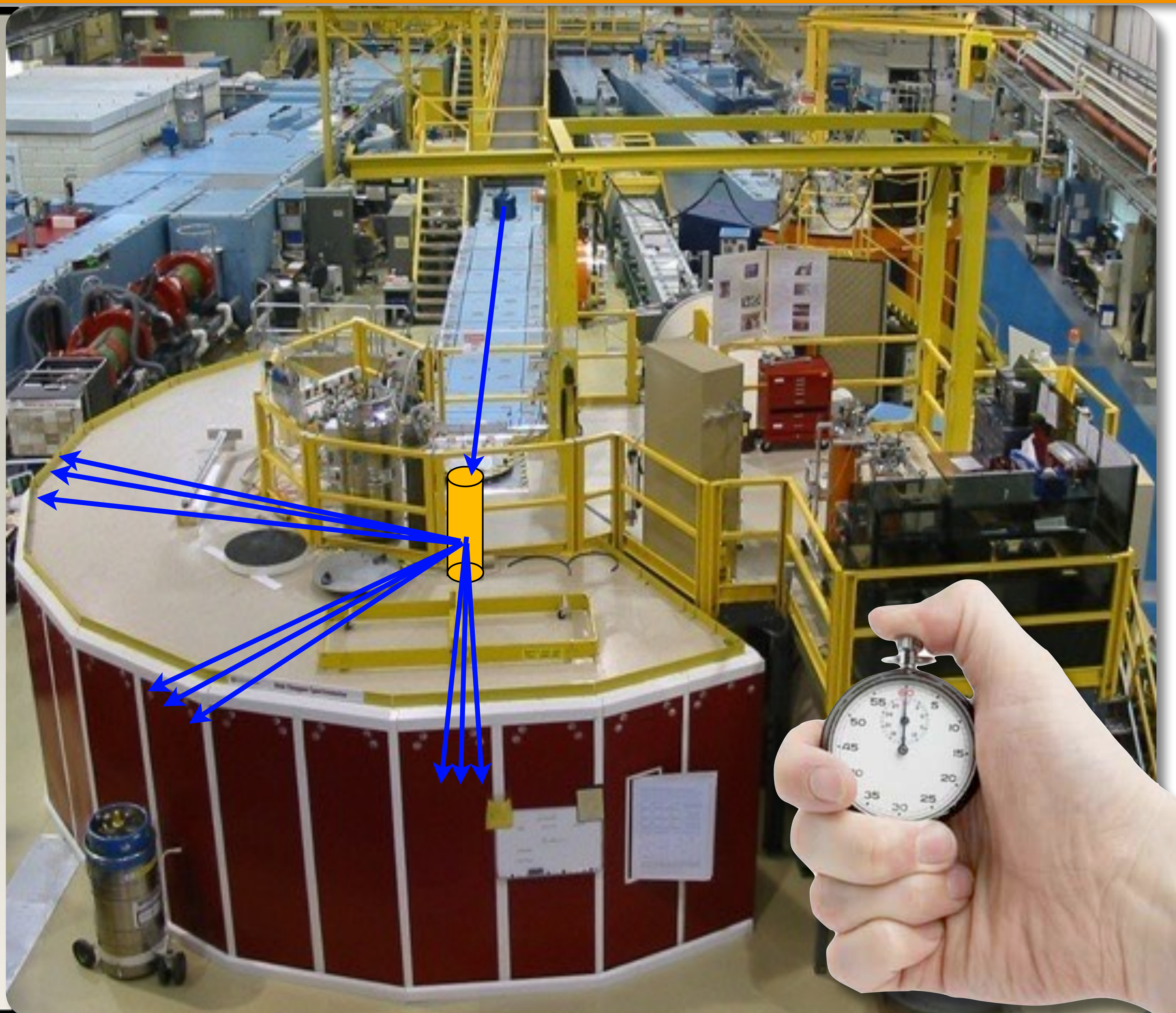
$$t = \frac{d}{v} = \left(\frac{md}{h}\right)\lambda$$

Time-of-flight Neutron Scattering



4D data sets for single crystals can be very large ~ 2 Tbyte

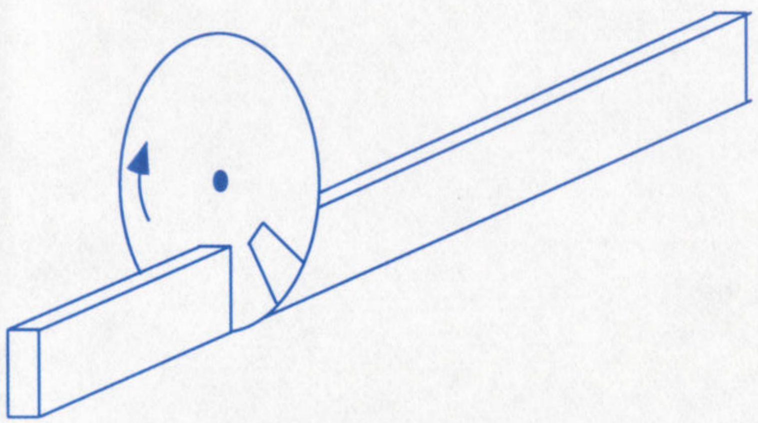


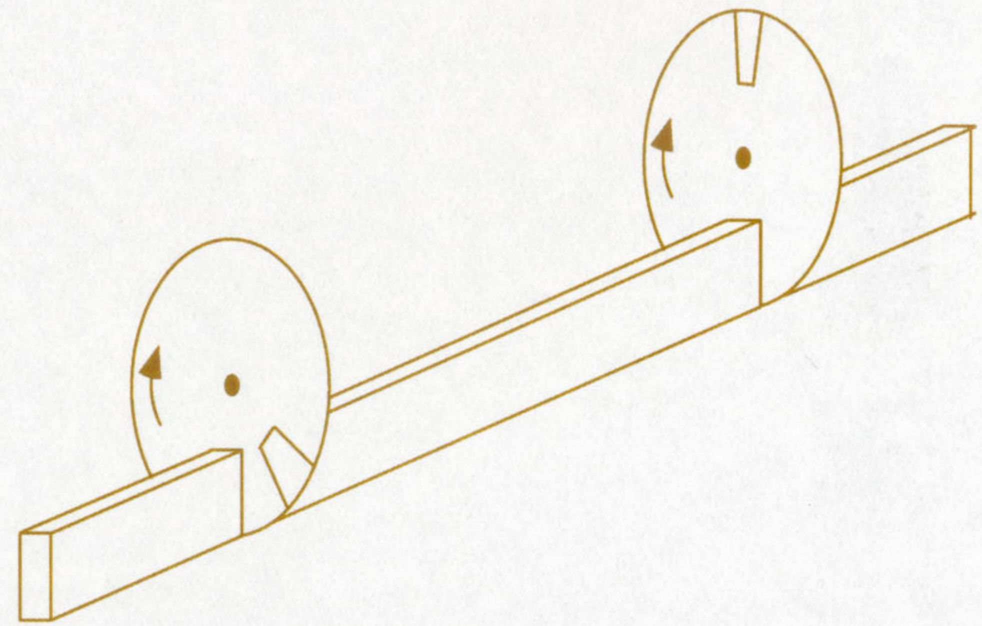


Time-of-flight Neutron Scattering: Disc Choppers

A single (disk) chopper pulses the neutron beam.

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



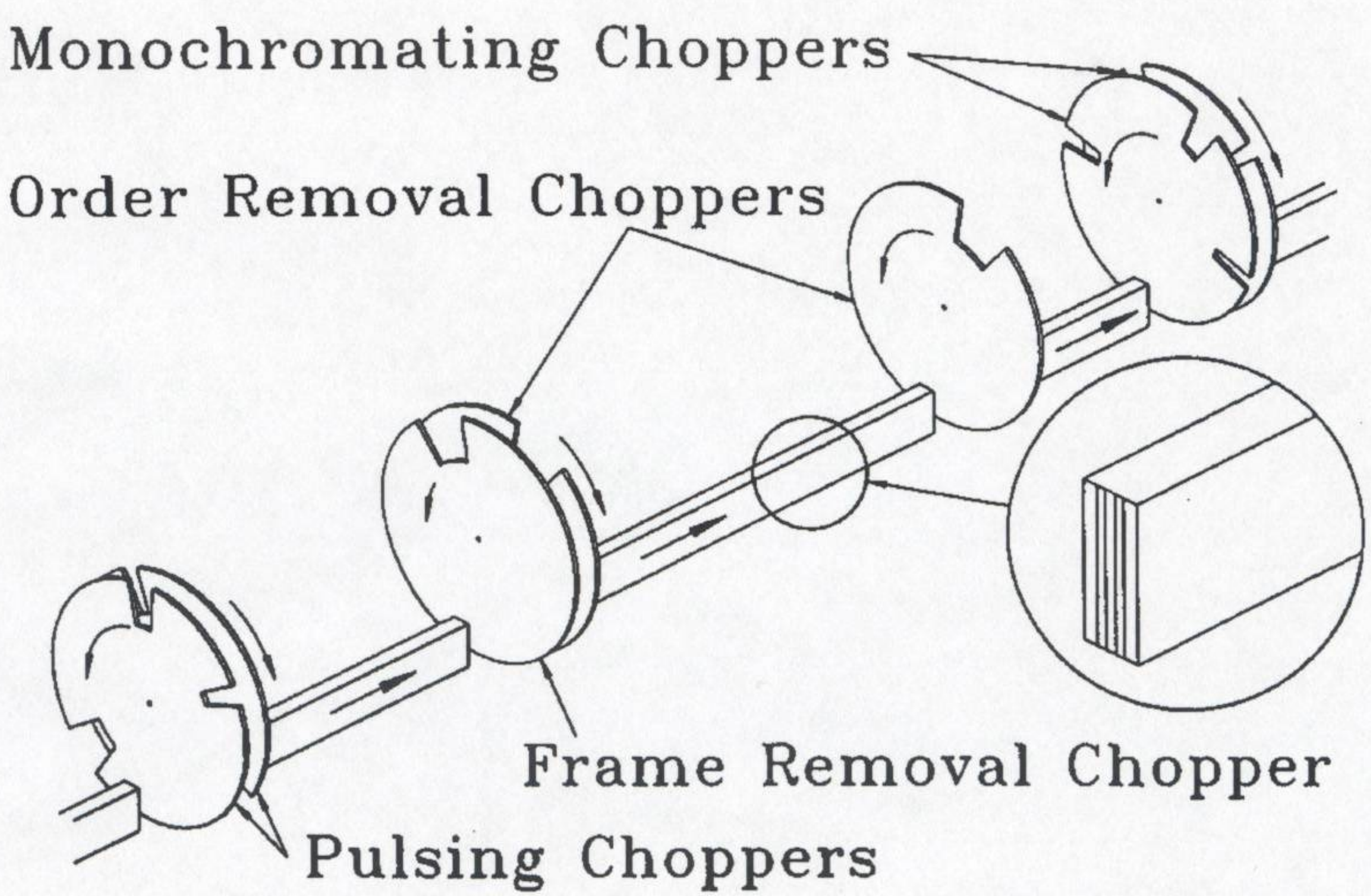


Counter-rotating choppers (close together), with speed ◆, behave like single choppers with speed 2◆. They can also permit a choice of pulse widths.

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

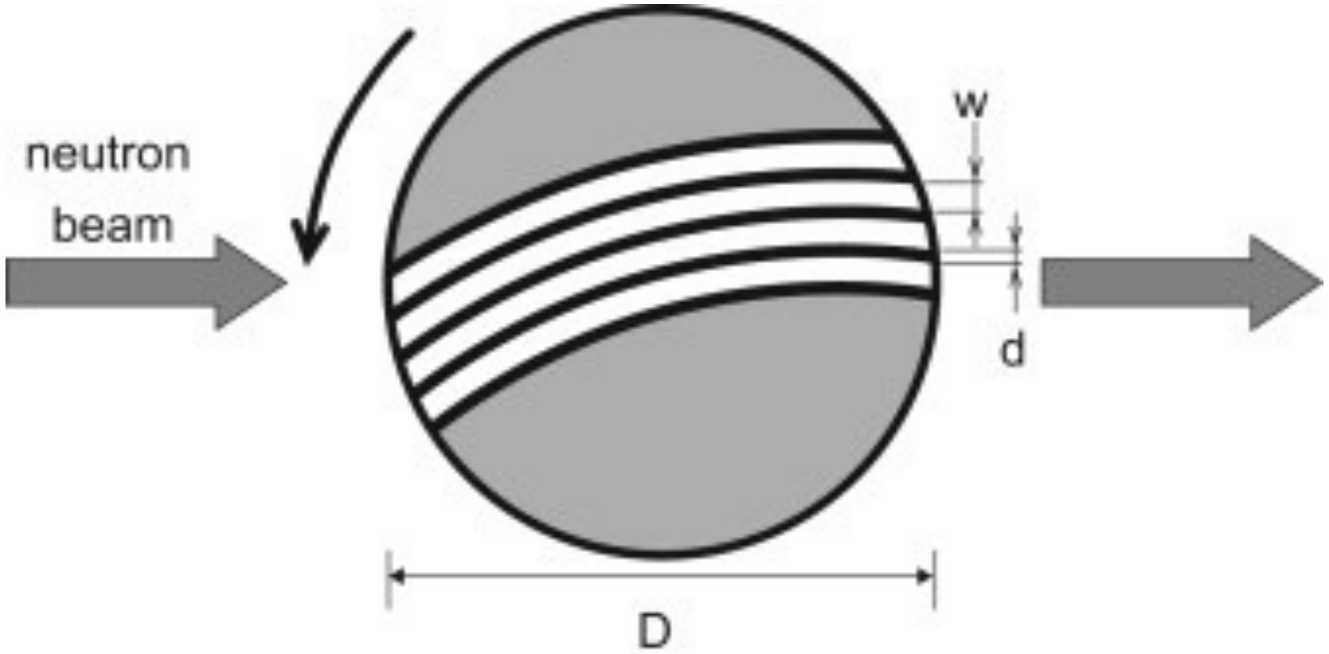
Time-of-flight Neutron Scattering: Disc Choppers

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



Disk 4B

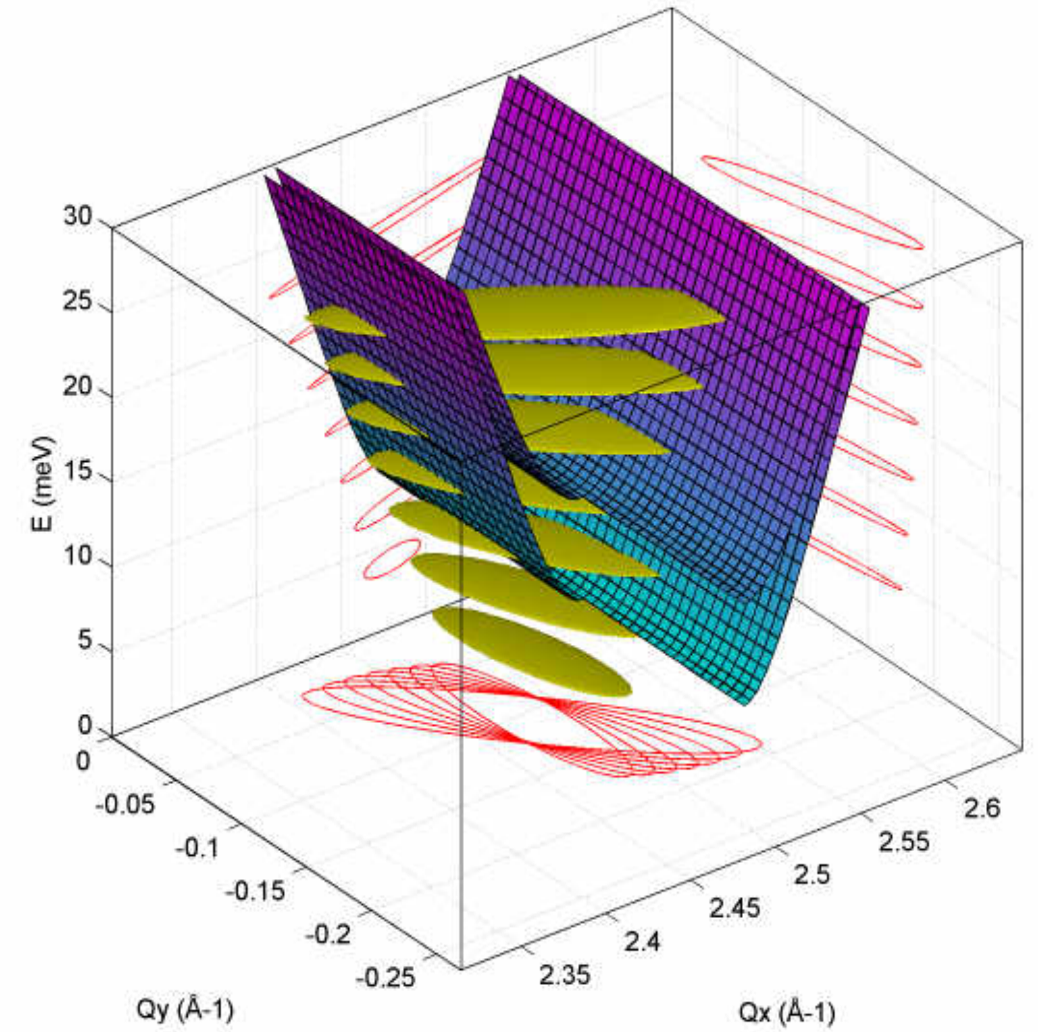
Time-of-flight Neutron Scattering: Fermi Choppers



Resolution Considerations

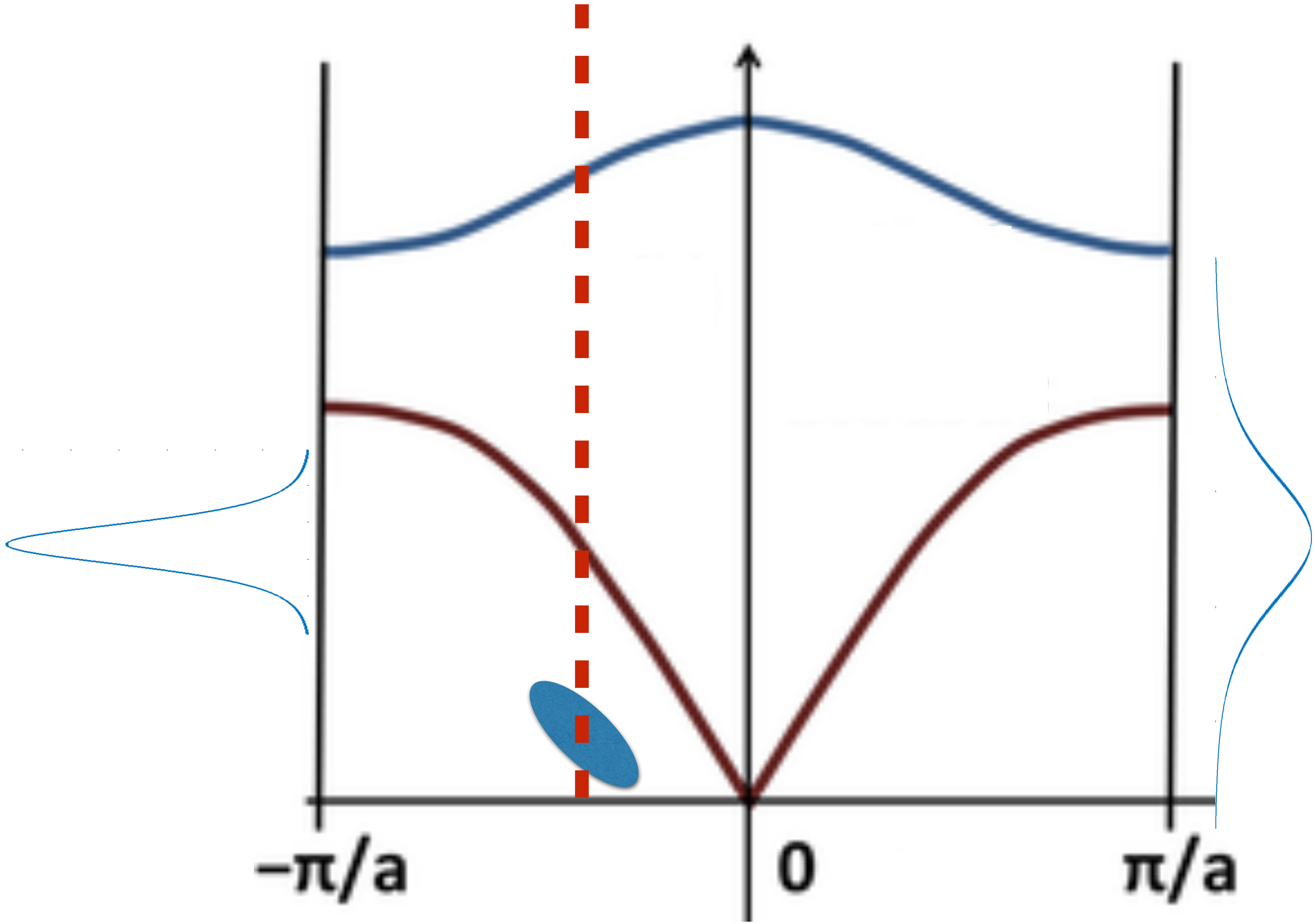
Resolution "ellipse" is defined by:

- **Beam divergences**
- **Collimation and distances**
- **Crystal mosaic, sizes**
- **Beam energy**

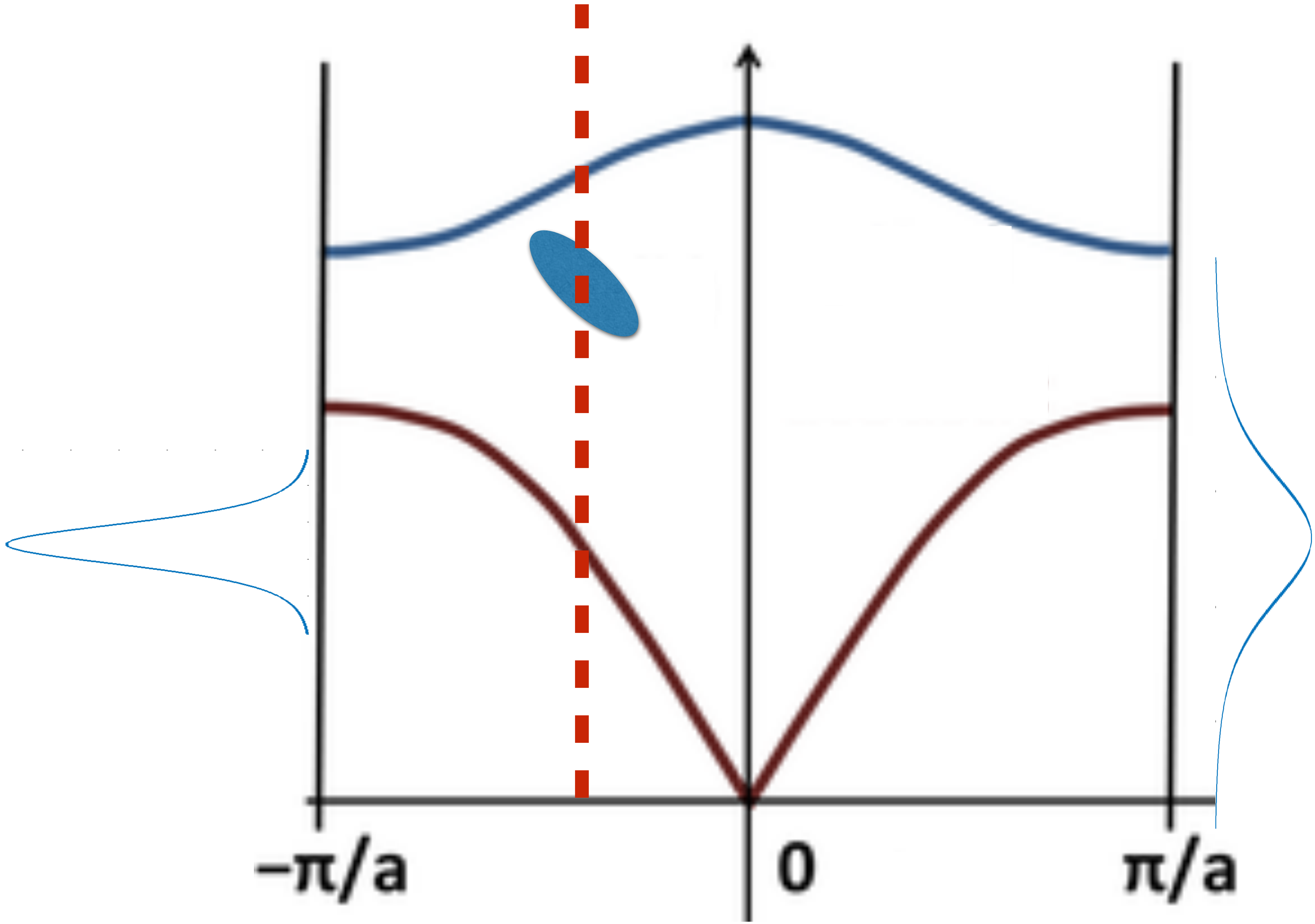


$$I(\vec{Q}_0,\hbar\omega_0)=\int S(\vec{Q}_0-\vec{Q},\hbar\omega_0-\hbar\omega)\,R(\vec{Q}_0,\hbar\omega_0)\,d\vec{Q}\,d\hbar\omega$$

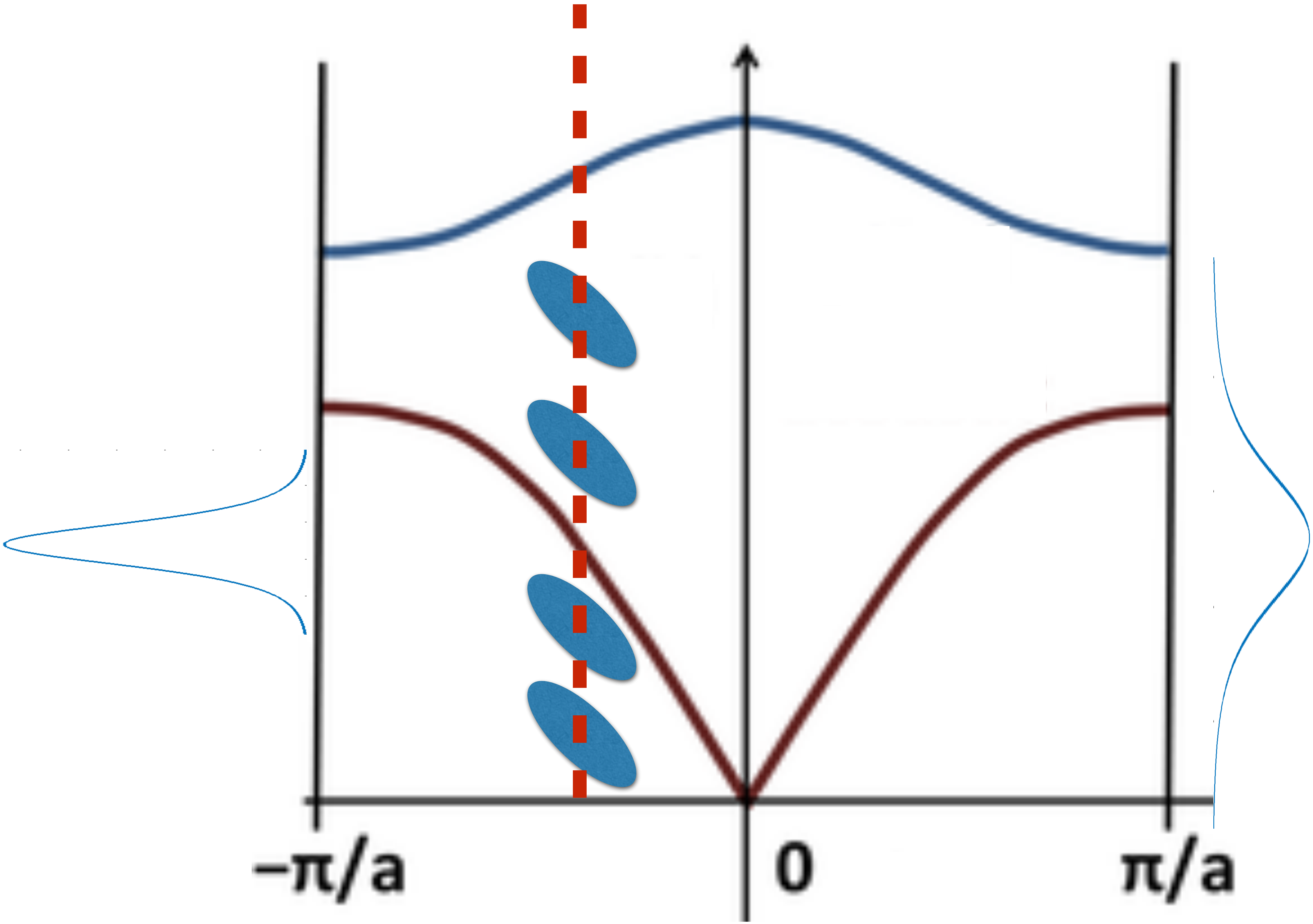
Resolution focussing



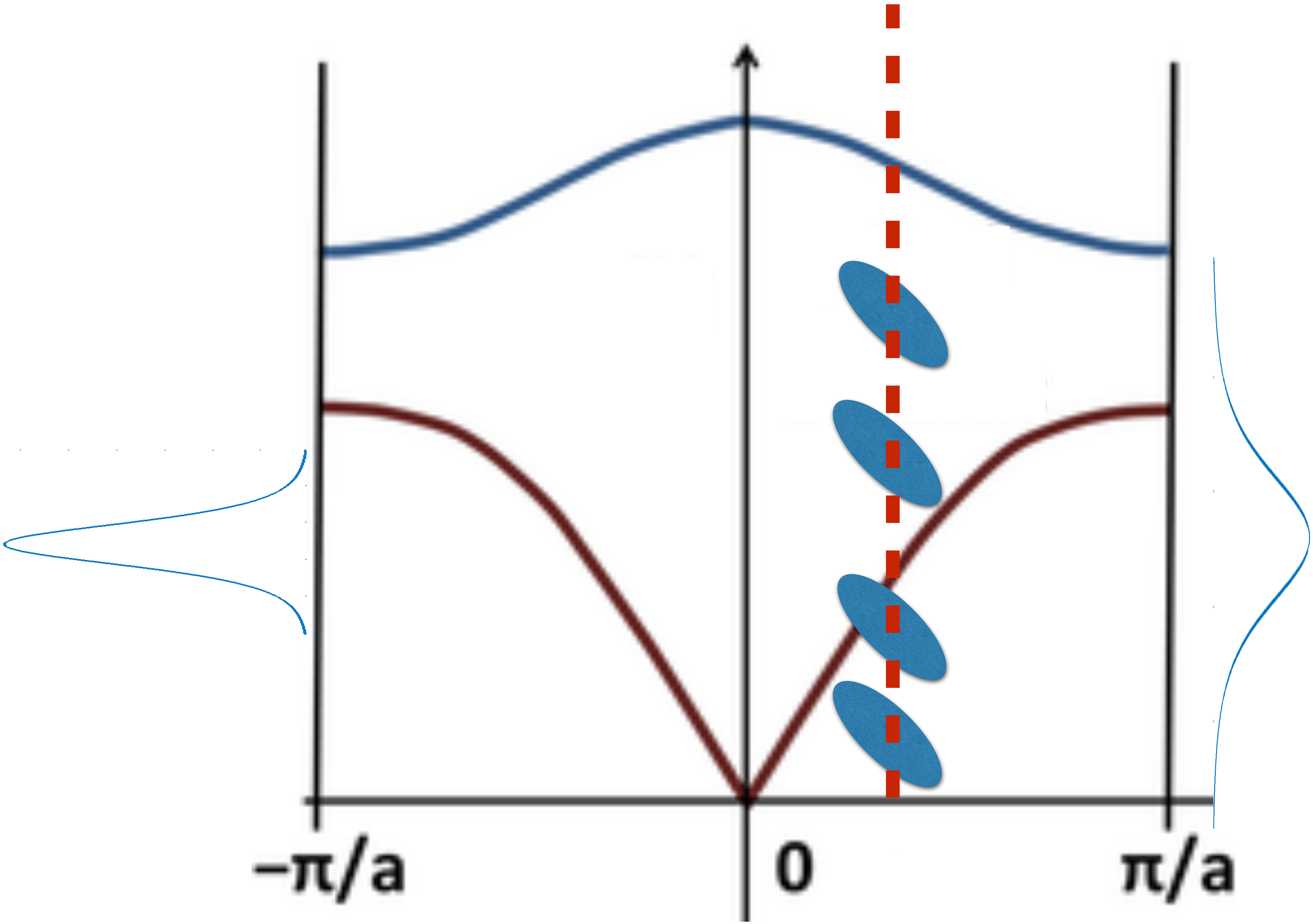
Resolution focussing



Resolution focussing



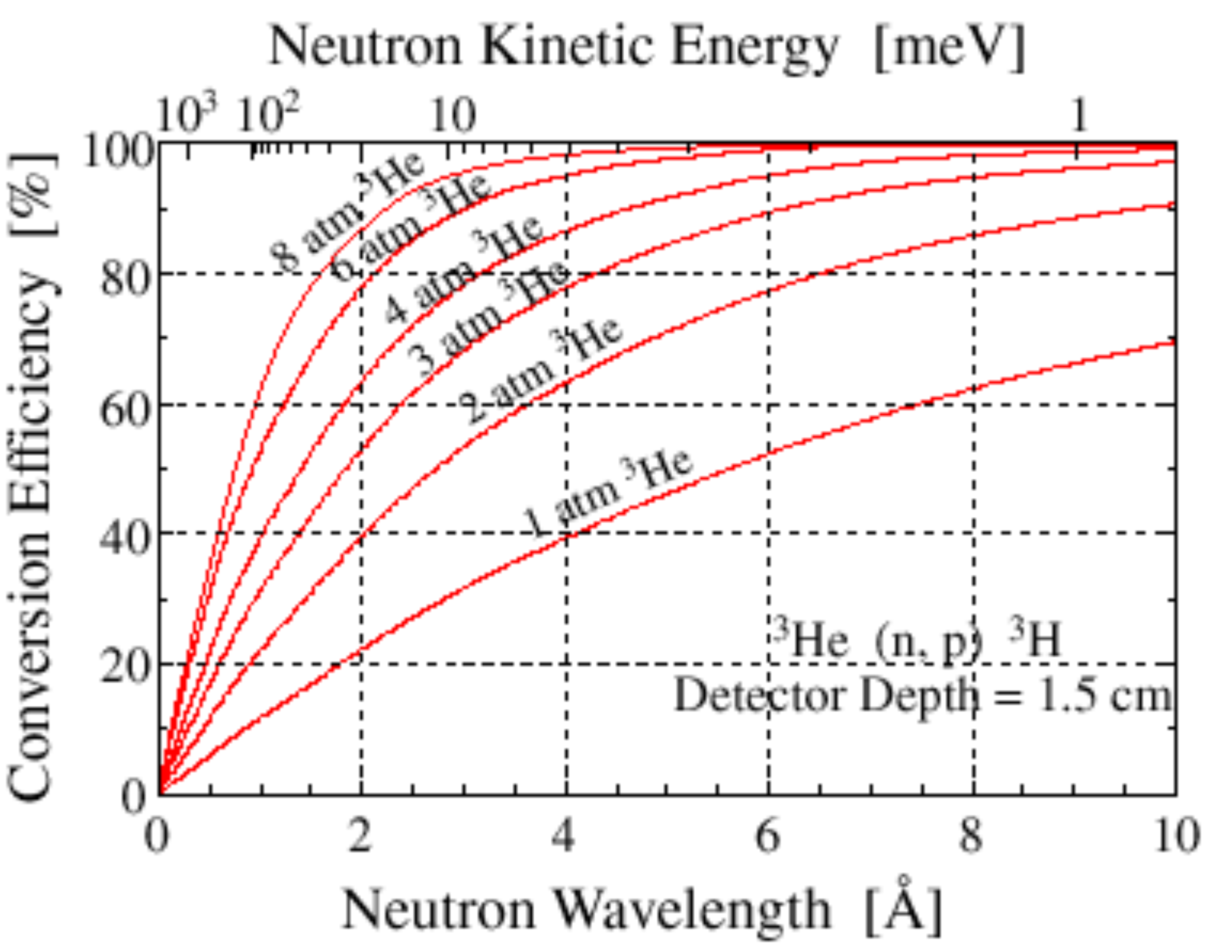
Resolution focussing



Neutron Detectors

Gas Detectors

- n + ^{3}He → ^{3}H + p + 0.764 MeV
- ionization of gas
- high efficiency

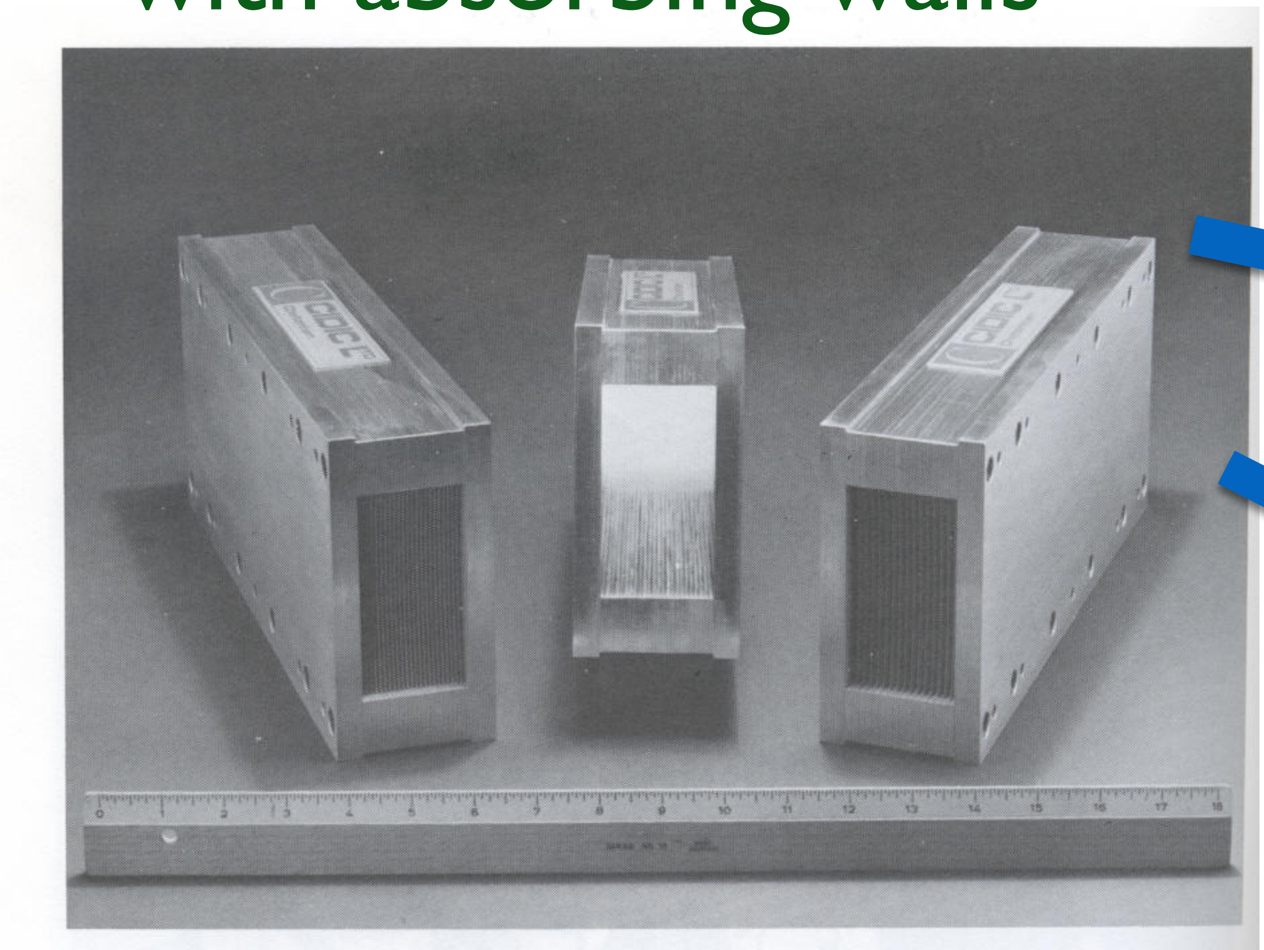


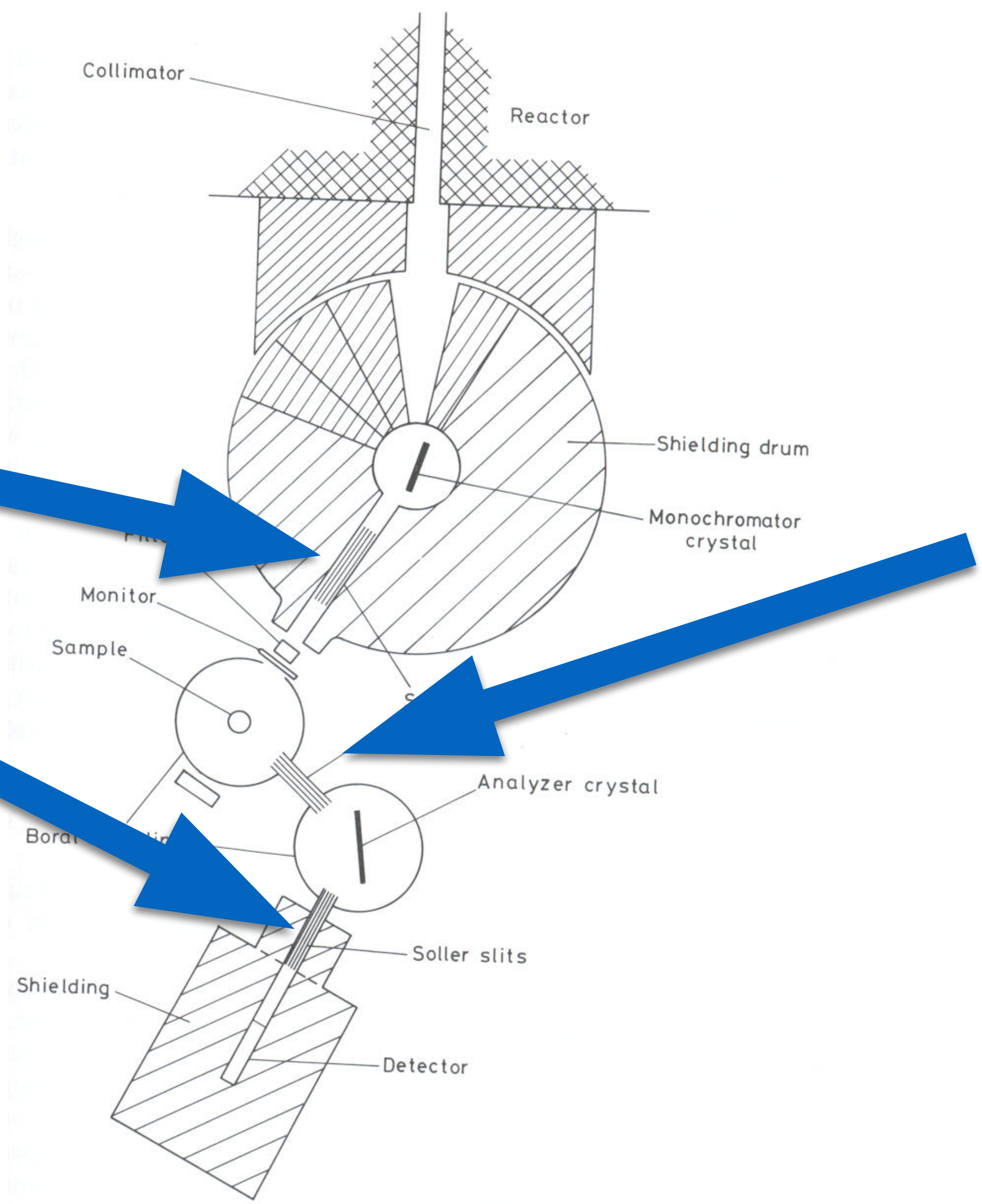
Beam monitors

- low efficiency detectors for monitoring beam flux

Q or angular resolution improved by using collimation (Soller slits)

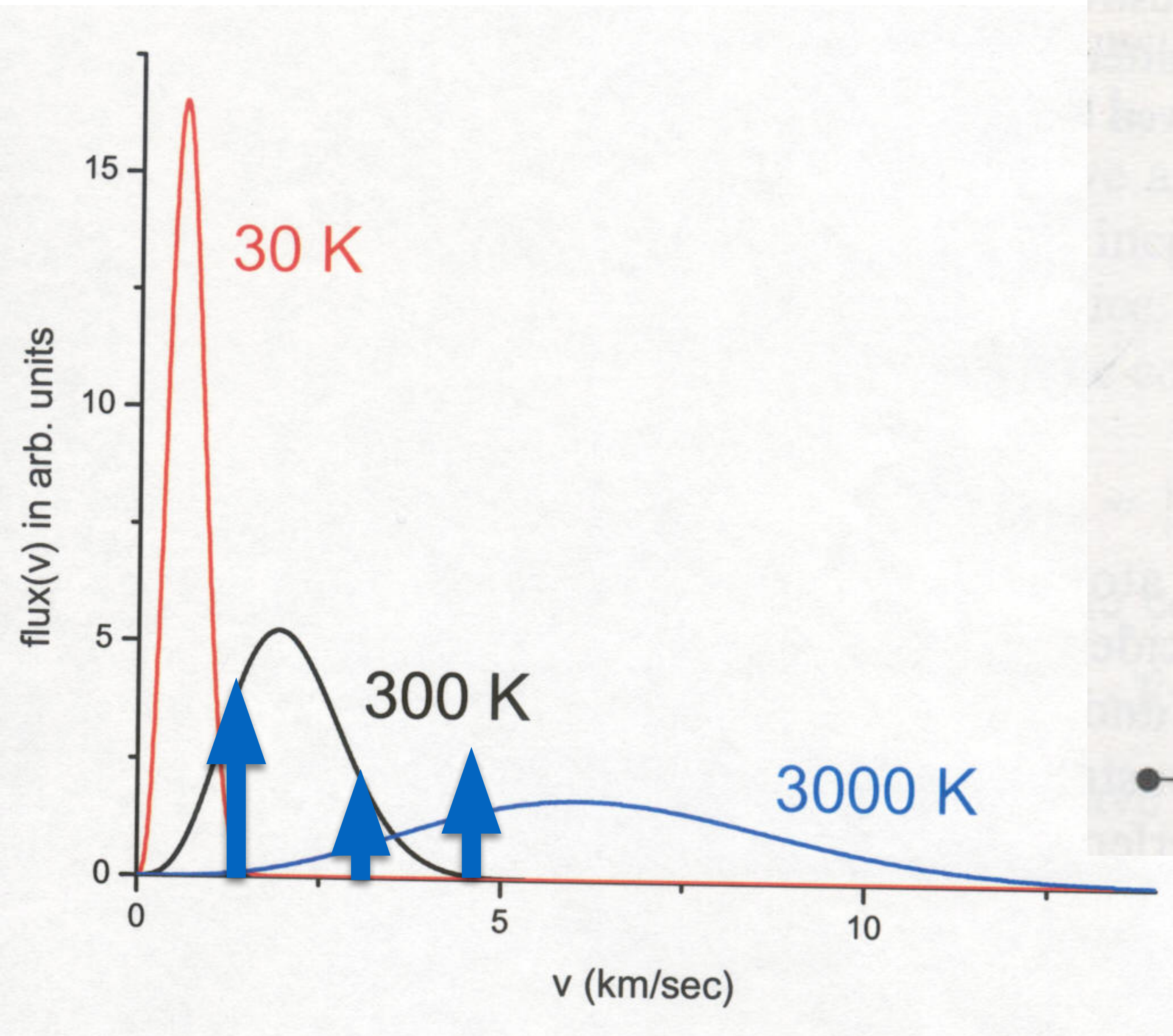
Soller slit collimators
neutron channels
with absorbing walls

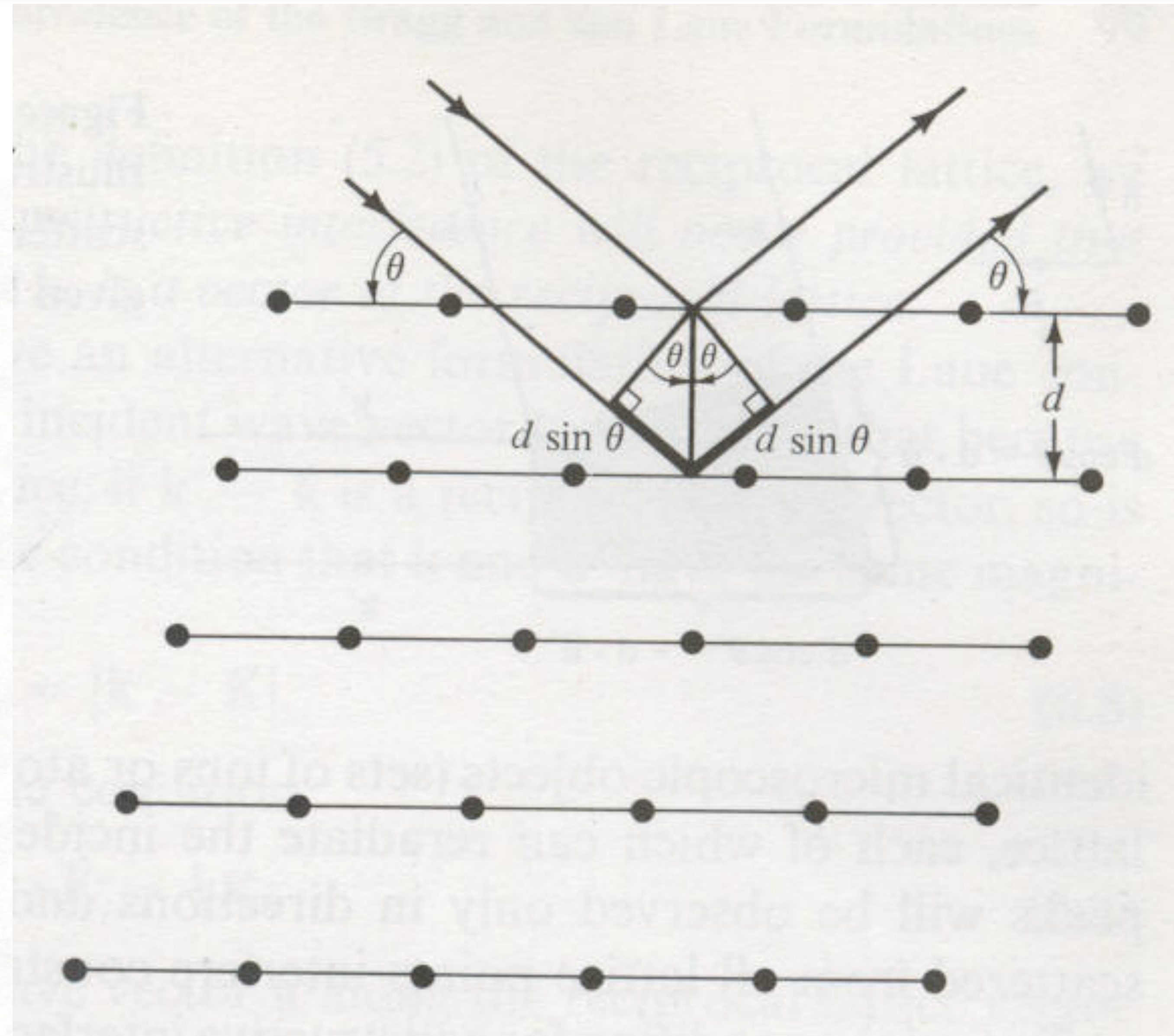




Allows the *angular* resolution of $\mathbf{k_i}$, $\mathbf{k_f}$ to be selected

Harmonic contamination from crystal monochromators

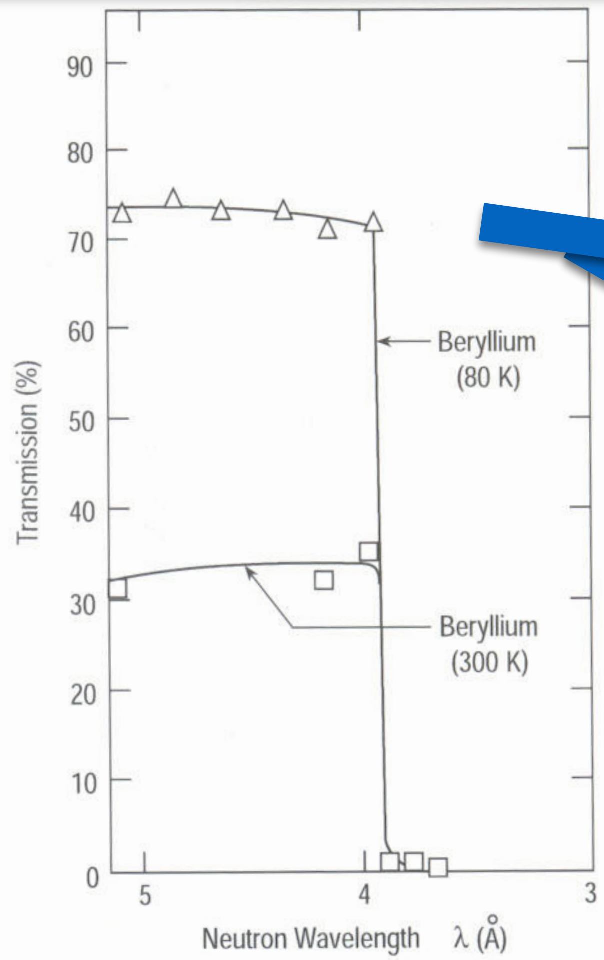


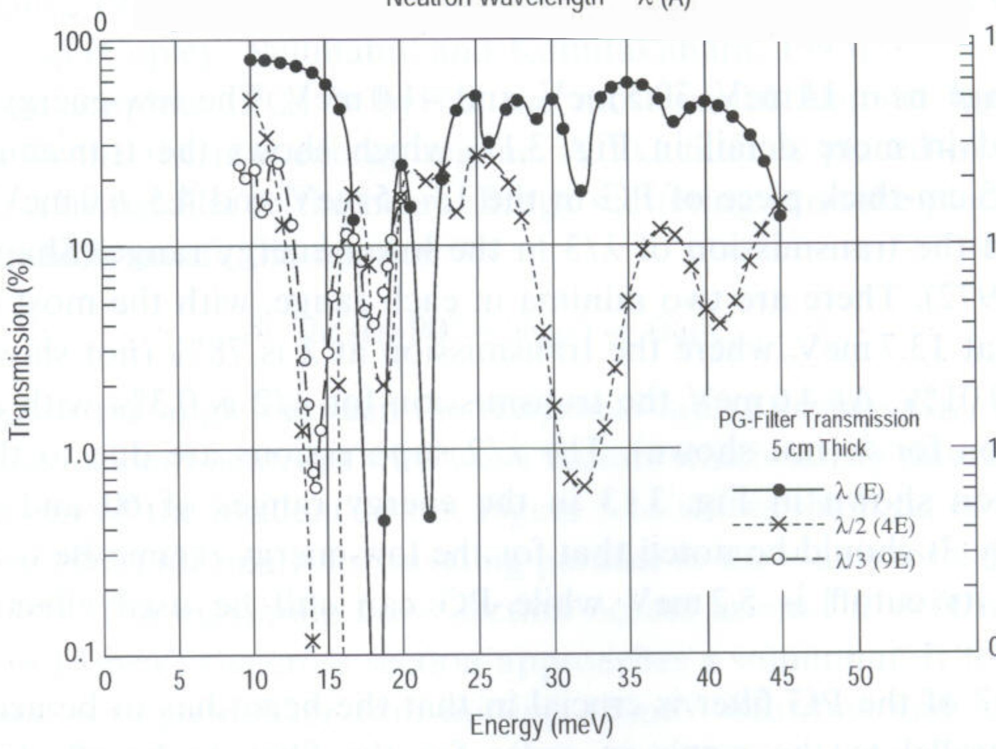


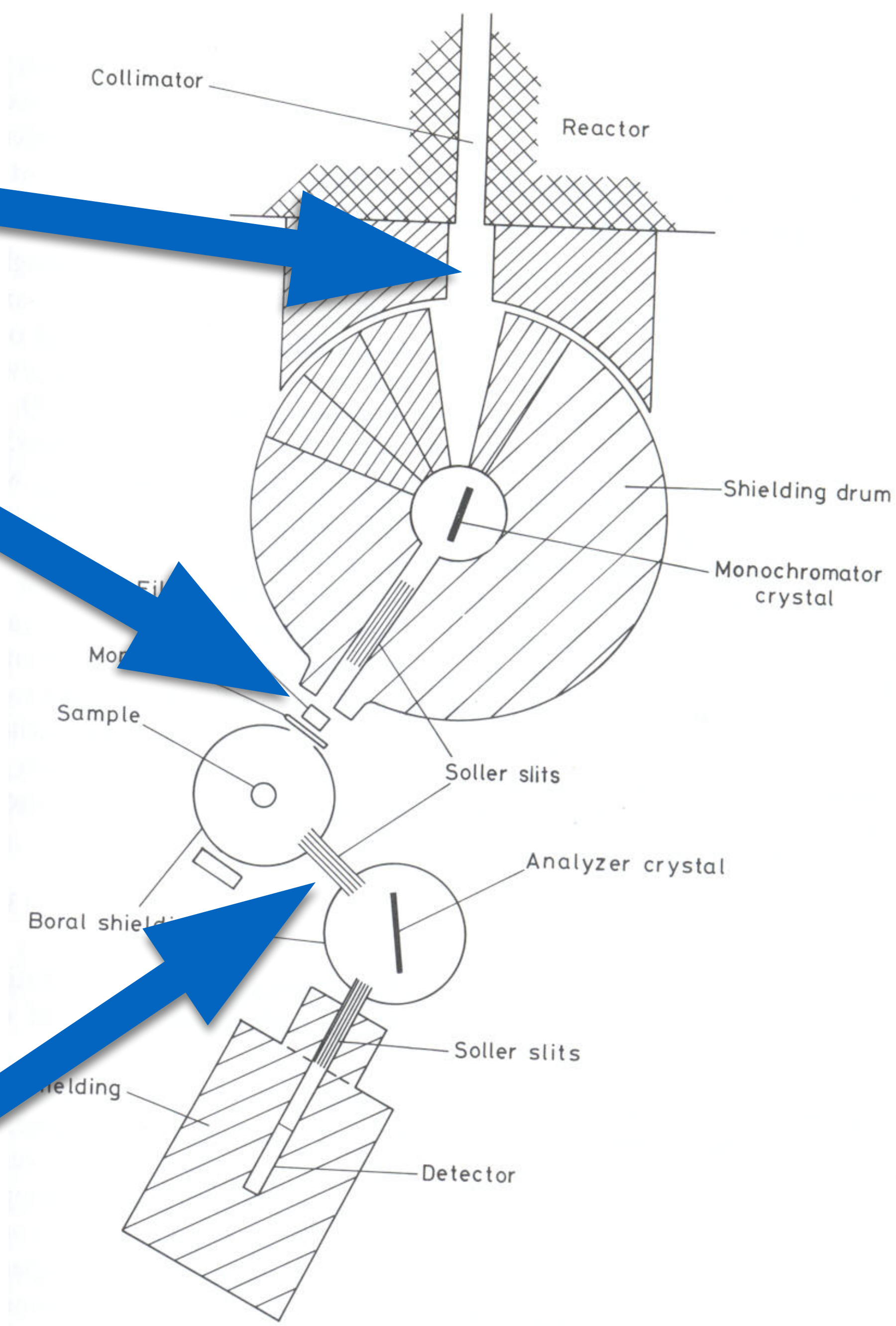
$$n\lambda = 2d\sin\theta$$

$\lambda, \frac{\lambda}{2}, and\ \frac{\lambda}{3}$ ***appear at the same*** θ ***with different n***

Neutron filters remove λ/n from incident or scattered beam, or both.

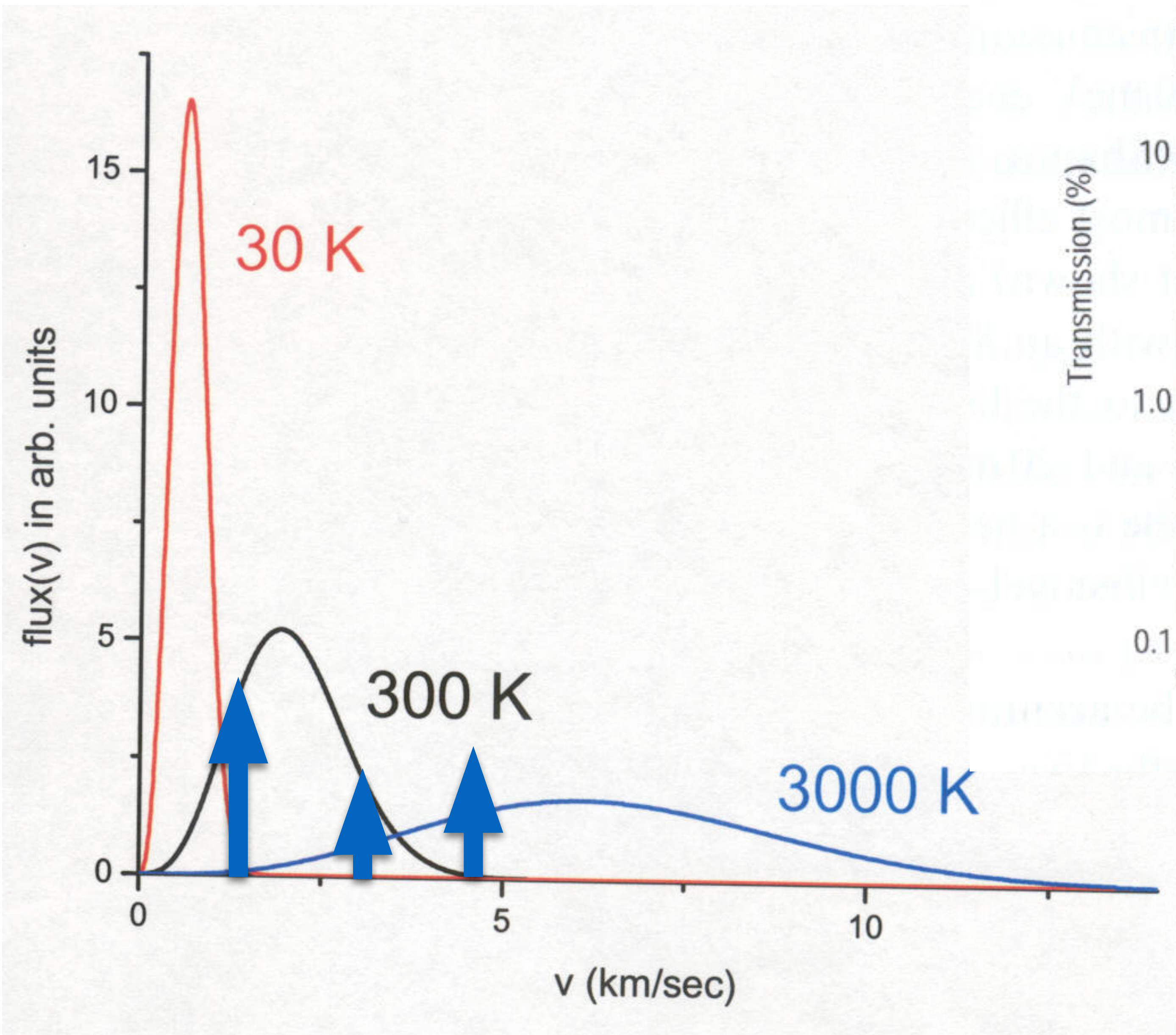


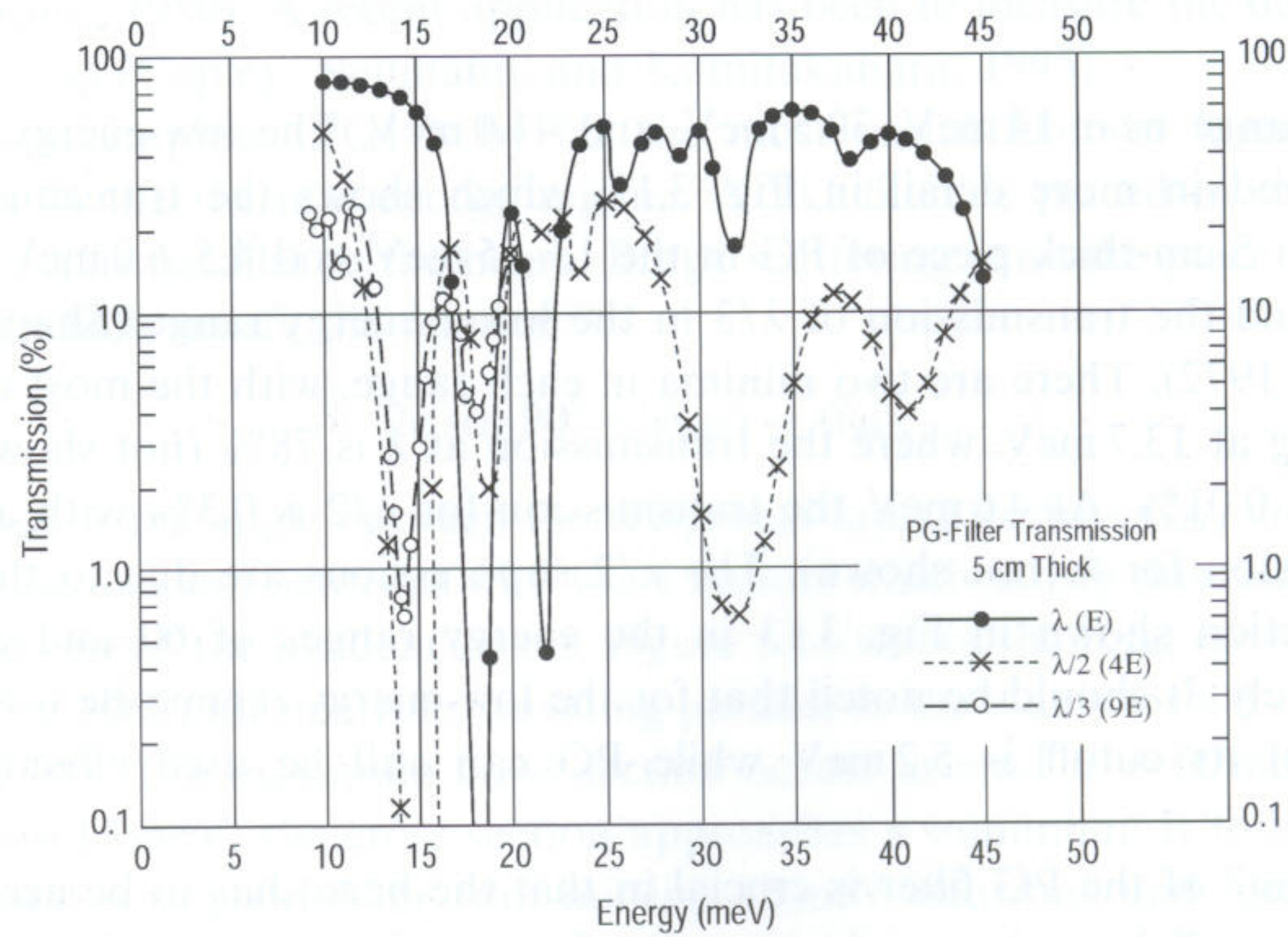




$$n\lambda = 2d\sin\theta$$

Harmonic contamination from crystal monochromators: Pyrolitic Graphite





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