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Centre for
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CENTRE FOR ENERGY RESEARCH

INTRODUCTION TO NEUTRON SCATTERING

Márton Markó

Budapest Neutron Centre // Centre for Energy Research

Neutron

NEUTRON IS AN ELEMENTARY PARTICLE

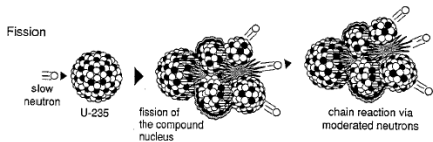
- Originated from the nuclei
- Freed by nuclear interaction
- Not stable in free form
- Interacts with nuclei / magnetic field

DATA

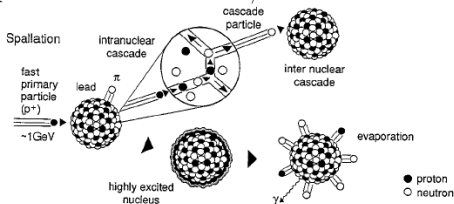
- Mass: $1.67476 \cdot 10^{-24}$ g
- Charge: 0 C
- Spin: 1/2
- Magnetic dipole moment: $-1,91315 \mu_n$
- Mean life time: 882.9 s

Sources of neutrons

Fission: 3 neutron / reaction

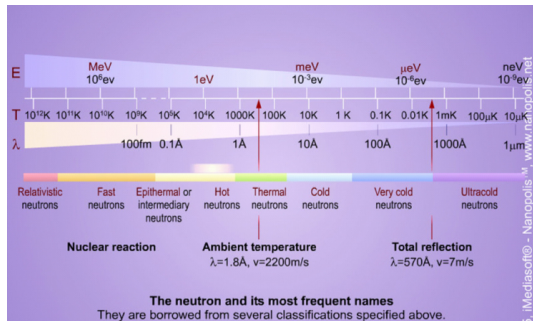


Spallation: 30 neutron / reaction



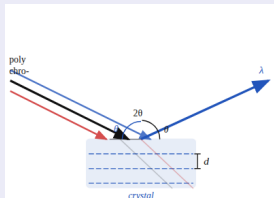
Moderators

- Hot: hot graphite
- Thermal: water, heavy water
- Cold: liquid H (orto or. para), deuterium, mesytelene..
- Ultracold: Solid deuterium



Neutron Monochromators

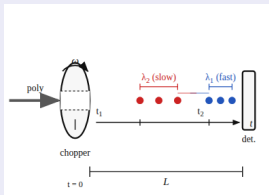
Crystal



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

- + Focusing; high resolution
- λ fixed by crystal & angle

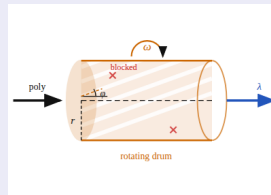
Time-of-Flight



$$\lambda = \frac{h}{m_n} \frac{t}{L}$$

- + Full spectrum, any λ
- Long flight path needed

Velocity Selector



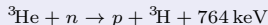
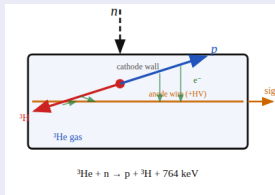
$$v = \frac{\omega r}{\tan \varphi}, \quad \lambda = \frac{h}{m_n v}$$

- + Compact; beam not deflected
- $\Delta\lambda/\lambda \approx 5-15\%$

Detecting Neutrons

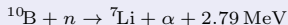
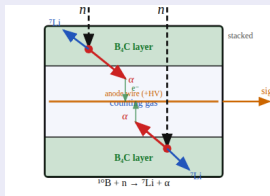
Thermal neutron cannot ionize directly
converter is needed

^3He Counter



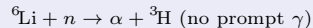
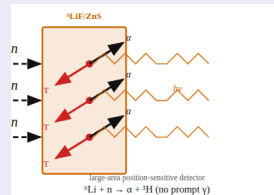
- + High efficiency; entire gas volume active
- ^3He scarce and expensive

^{10}B / B_4C Converter



- + Cheap; scalable to large areas
- Many layers needed; lower efficiency

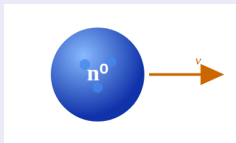
$^6\text{LiF}/\text{ZnS}(\text{Ag})$ Scintillator



- + High spatial res.; imaging capable
- High γ sensitivity (ZnS scintillates)

Properties of neutron

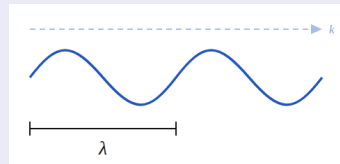
Particle



$$m_n = 939.6 \text{ MeV}/c^2$$

$$E_k = \frac{m_n v^2}{2} \quad p = m_n v$$

Wave



$$\lambda = \frac{h}{p} = \frac{h}{m_n v}$$

$$E = \hbar \omega = h \nu, \quad p = \hbar k$$

$$\Psi = e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$$

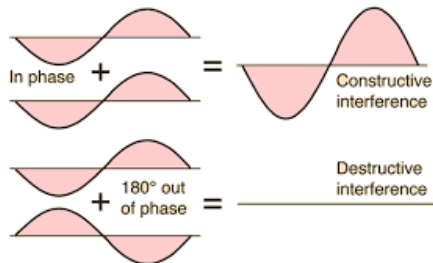
thermal: $\lambda \approx 1.8 \text{ \AA}$ $E \approx 25 \text{ meV}$ $v \approx 2.2 \text{ km/s}$

cold: $\lambda \approx 4 \text{ \AA}$ $E \approx 5 \text{ meV}$ $v \approx 1 \text{ km/s}$

Interference

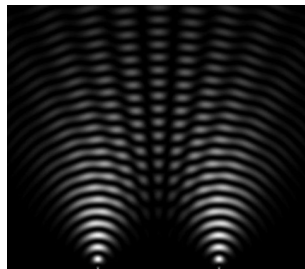
Wave

- Time period: T
- Frequency: $f = 1/T$
- Angular frequency: $\omega = 2\pi f$
- Wavelength: λ
- Wavenumber: k



Wave

- $\Psi = e^{i(kr - \omega t)}$
- Phase: $kr - \omega t$
- Phase difference: π : destructive interference, 2π : constructive interference



Neutron - X-ray differences

	Neutron	X-ray
Base of scattering	potential	charge oscillation
Scattering on	nucleus / magnetic field	electron density
Scattered intensity	Isotope - dependent	Z^2
Penetration depth	some cm	« mm
Energy @ 1 Å	81 meV	12.4 keV
Source Brillance	Unit	10^{10} Unit
Atomic scattering factor	isotropic	decreasing in backscattering

Neutron - matter interactions

- Absorption:
 - ▶ Activation analysis (NAA, PGAA)
 - ▶ Nuclear physics investigations
- Scattering
 - ▶ Elastic (**Energy of neutron does not change**): Investigation of static structure
 - ▶ Inelastic (**Energy of neutron changes**): Investigation of dynamic structure, excitation in condensed matter

The interactions are measured with cross section

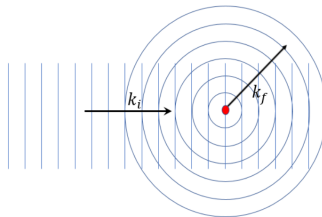
- Microscopic cross section (nucleus): σ [barn= 10^{-24} cm²]:
Number of interactions within a secundum pro unit intensity
- Macroscopic cross section (matter): $\Sigma = n\bar{\sigma}$ [1/cm] (sample)
where n [1/cm³] is the nuclear density
Number of interactions within a secundum pro unit intensity and unit volume of the sample

Scattering

The nuclei scatter the thermal neutrons in spherical wave

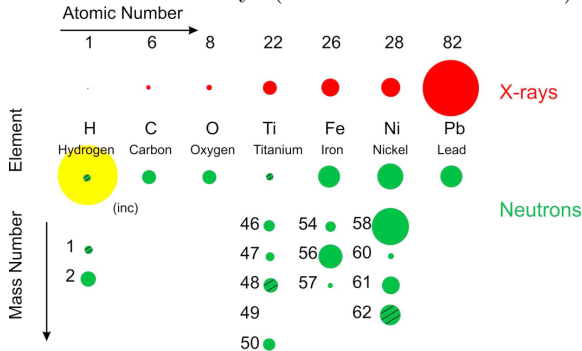
- Short range interaction (fm)
- The scattering is described by b (in fm units):
 b : The scattered amplitude over the amplitude of the incident wave (In the case of X-rays the scattering is anisotropic due to the size of the electronic shell)
- The scattered intensity is given by the scattering cross section: $\sigma_s = 4\pi b^2$

Note that b can be negative, and it can be complex if the nucleus is a high absorber



Scattering amplitudes (X-ray and Neutron)

- Neutron: Isotope sensitivity, non monotonic function of the elemental number. Isotropic
- X-ray: scattered by the electronic density: ($b \approx c \cdot \text{elemental number}$). Anisotropic



The diameters of the circles shown scale with the scattering amplitude $f_c(\sin\theta=0)$ for x rays, and $b_{\text{coh}} \cdot 10$ for neutrons. Hatching indicates negative scattering amplitudes.

Scattering on the magnetic field

Effect of the magnetic field

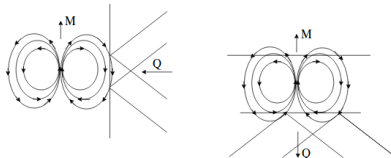
- The component of the magnetic field parallel with the neutron spin gives an extra potential field
- The component of the magnetic field perpendicular to the neutron spin causes a spin-flip
- Magnetic scattering factor:

$$f_m(\mathbf{q}) = F(q)s_n(\mathbf{e}(\mathbf{eh}) - \mathbf{h}), \quad e = |q|/q, \quad \mathbf{h}: \text{spin of the atom}$$

scattering on magnetic atom is not isotropic (like X-ray scattering):

Polarization factor: $\mathbf{e}((\mathbf{eh}) - \mathbf{h})$

magnetic form factor: $F(q)$



Kinematic Scattering Theory

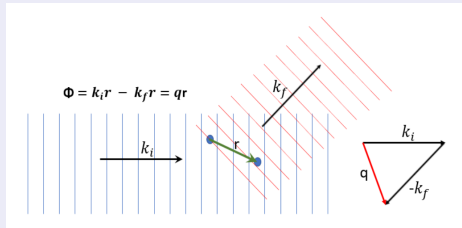
Gives a simplified model for the scattering on condensed matter

The wave is scattered only once and leaves the sample

Not applicable to diffraction on perfect crystals and reflectometry

Scattering on atomic structures

The spherical wave is close to plane wave at large distances



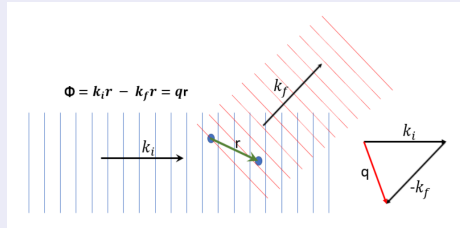
- Only the phase differences are important at the detector:

$$\Psi = \frac{1}{R} \sum_m b_m A e^{i(\vec{k}_i - \vec{k}_f) \cdot \vec{r}_m} = \frac{1}{R} \sum_m b_m A e^{i \vec{q} \cdot \vec{r}_m}$$

- The scattered intensity is measured as a function of q
 $\vec{q}$ spans the reciprocal space
- The scattered intensity: $|\Psi(\vec{q})|^2$

Scattering on atomic structures

Changing to scattering length density: $\rho(\vec{r}) = \frac{\Sigma b}{V}$



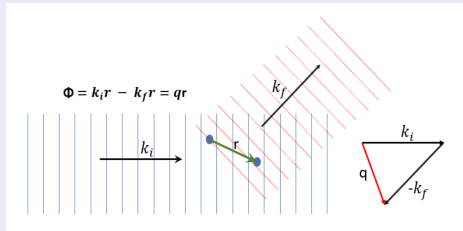
- The scattered wavefunction is:

$$\Psi = \frac{1}{R} \int \rho(\vec{r}) e^{i\vec{q} \cdot \vec{r}} d^3 \vec{r}$$

- The wave function is the Fourier-transform of the sample

Scattering on atomic structures

Changing to scattering length density: $\rho(\vec{r}) = \frac{\Sigma b}{V}$



If we step with π/q steps, the intensity shows how many times we have to step up and down

- large- q : small steps, we see the grass (atomic resolution)
- small q : large steps, we see the haystack (nano objects)

Elastic scattering

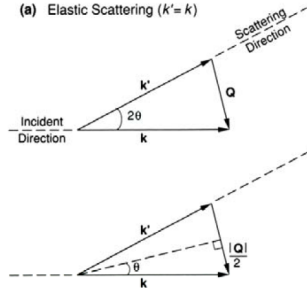
No energy change
more than 99% of the scattering events in solid samples

Incident and scattered wavenumbers are the same

Static structure

- Diffraction
- Small angle scattering
- Reflectometry

(a) Elastic Scattering ($k' = k$)



$$\sin \theta = \frac{Q/2}{k}$$

$$Q = 2k \sin \theta = \frac{4\pi \sin \theta}{\lambda}$$

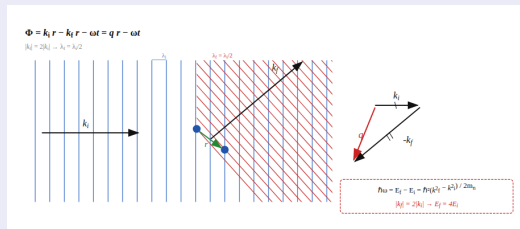
The scattered intensity depends only on q :

$$\vec{q} = \vec{k}_f - \vec{k}_i$$

$$q = 2k \sin(\theta)$$

Scattering on moving atomic structures

The spherical wave is close to plane wave at large distances



- Only the phase differences are important at the detector:

$$\Psi = \int_t \frac{1}{R} \sum_m b_m A e^{i(\vec{k}_i - \vec{k}_f) \cdot \vec{r}_m - (\omega_i - \omega_f)t} = \int_t \frac{1}{R} \sum_m b_m A e^{i\vec{q} \cdot \vec{r}_m - \omega t}$$

- The scattered intensity is measured as a function of $\vec{q}$ and ω
 $\vec{q}$ and ω spans the reciprocal space
- The scattered intensity: $|\Psi(\vec{q}, \omega)|^2$

Scattering on moving atomic structures

Scattering on non-stationary sample

Scattering on moving atoms cause a some kind of Doppler effect Incident frequency: ω_i
 Final frequency: ω_f

BUT

Energy change of the neutron is: $\hbar(\omega_f - \omega_i) = \hbar\omega$

Conservation of the energy:

Excitation or deexcitation of the sample

The total energy change is 0:

$$E = -\hbar\omega$$

Similar calculation gives:

- $\Psi = \frac{1}{R} \int \iiint \rho(\vec{r}, t) e^{i(\vec{q} \cdot \vec{r}(t) - \omega t)} \delta(E - \hbar\omega) d^3\vec{r} dt$
- The same Fourier transformation but in 4D

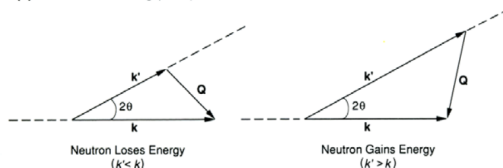
Inelastic scattering

The energy of neutrons change during scattering

Dynamic structure
(molecular vibration, lattice vibration, diffusion etc..)

- Three axis spectrometer
- Indirect - direct TOF
- Backscattering
- Neutron spin echo

(b) Inelastic Scattering ($k' \neq k$)



The scattered intensity depends on the momentum and energy transfer:

$$\vec{q} = \vec{k}_f - \vec{k}_i$$

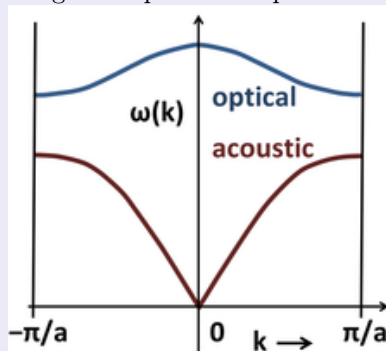
$$E = E_f - E_i$$

Weak intensity in solid samples

Scattering on moving atomic structures

Example: oscillations (phonons)

Oscillation has momentum and energy (quasy - particle)
Signal is q - and ω dependent



Coherent - incoherent scattering

Coherent scattering

Neutron is able to make interference

- Described by the average scattering length ($\bar{b}$)
- Scattering length density follows the structure of the atom

Incoherent scattering

Neutron is not able to make interference

- Random change of scattering length (density) in the sample:
 $\sigma_i = 4\pi \langle (b - \bar{b})^2 \rangle$

Investigation of structure

Neutron is able to make interference

- Relative positions of **different atoms**
- **Collective** excitation (eg. phonon, magnon)

Investigation of one atom

Neutron is not able to make interference

- "Smearing" of position of **one atom**
- Energy levels of **one atom**

Van Hove theorem

Convolution theorem

$$\mathcal{F}(f * g) = FG \text{ and } \mathcal{F}(fg) = F * G$$

Using of convolution theorem:

$$|\mathcal{F}(g)|^2 = \mathcal{F}(g)\mathcal{F}(g)^* = \mathcal{F}(<g, g>)$$

$$\text{Intensity in kinematic scattering: } |\mathcal{F}(\rho)|^2 = \mathcal{F}(<\rho, \rho>)$$

Van Hove rule:

Scattering function is the Fourier transform of the autocorrelation function

Note that the Fourier transformation is linear:

$$I_{f+g} = I_f + I_g + I_{fg}: \text{cross correlation appears}$$

Basic consequences

"Inverse" effect: resolution

Sharp changes in the sample gives signal in high q

Long range correlation in the sample gives sharp change in the scattering image (needs high q -resolution)

"Inverse" effect II: Particle sizes

Large objects with low resolution are visible in small q

Small objects with high (atomic) resolution needs the high q

Basic consequences II

Many particles

Scattering signal of two (many) particles is NOT the sum of scattering signals of each particle:

$$\text{Wave function: } \Psi = \Psi_1 + \Psi_2$$

$$\text{Scattered intensity: } |\Psi|^2 = |\Psi_1|^2 + |\Psi_2|^2 + |\Psi_1 \Psi_2|^2$$

The correlation of the particles appear

"Multiplication" effect

Scattering function of repeating parts of the sample is the product of:

- scattering function of repeating parts
- scattering function of the centres of the repeating parts

Incoherent scattering and contrast

Incoherent scattering

- Coherent scattering lengths: ρ_0
- Random change of scattering lengths: $\Delta\rho$
- Real distribution: $\rho = \rho_0 + \Delta\rho$

$\Delta\rho$ random:

- Spin dependent scattering: spin incoherence
- Different isotopes: Isotope incoherence
- Different elements: Chemical incoherence

Cause flat (incoherent) background

Incoherent scattering and contrast

Incoherent scattering

- Coherent scattering lengths: ρ_0
- Random change of scattering lengths: $\Delta\rho$
- Real distribution: $\rho = \rho_0 + \Delta\rho$

$\Delta\rho$ random:

- Spin dependent scattering: spin incoherence
- Different isotopes: Isotope incoherence
- Different elements: Chemical incoherence

Cause flat (incoherent) background

Contrast

Homogeneous sample scatters only forward

Scattering intensities depend on the square of the **not random** differences of the scattering length density: the contrast

Scattering on isotropic system

The systems are generally not isotropic, but in many case the particles can have any orientation giving an isotropic sample

Radial averaging: $\rho(\vec{r}) \rightarrow \rho(r)$

$$I(\vec{q}) \rightarrow I(q) = \left| \int \rho(r) e^{i\vec{k}\vec{r}} d\phi r^2 d\cos\theta dr \right|^2$$

$$I(q) = \left| \int r^2 \frac{p(r) \sin(qr)}{qr} dr \right|^2$$

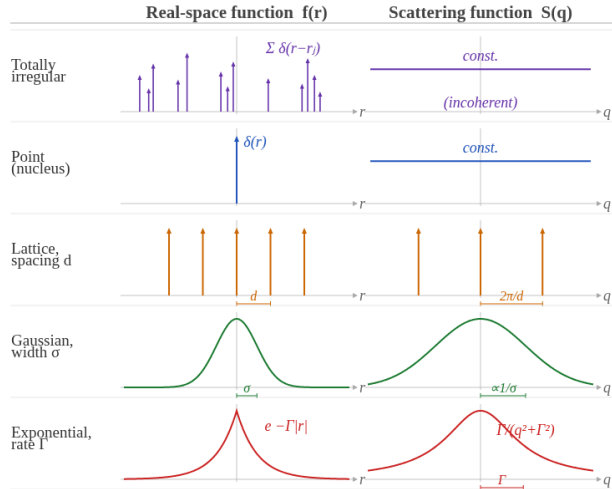
$$I(q) = \left| \int \frac{p(r) \sin(qr)}{qr} dr \right|^2$$

Mostly used in SANS and liquid diffraction

Scattering of different systems

- Atomic structure $\rightarrow$ small wavelength, large angle $\rightarrow$ diffraction
- Bulk nanostructure $\rightarrow$ large wavelength, small angle $\rightarrow$ small angle neutron scattering (SANS)
- Thin layers, flat interfaces $\rightarrow$ large wavelength small angle in one direction $\rightarrow$ reflectometry
- Lateral changes in thin layers / flat interfaces $\rightarrow$ large wavelength small angle in two direction $\rightarrow$ grazing incidence neutron scattering (GISANS)
- molecular vibrations $\rightarrow$ small wavelength (hot source) $\rightarrow$ molecular vibration spectrometer
- phonons - magnons $\rightarrow$ small wavelength $\rightarrow$ molecular vibration spectrometer

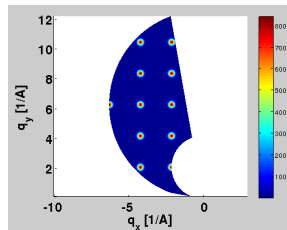
Scattering of basic structures



Scattering on atomic structures II

Example: Diffraction on a single crystal with d lattice spacing

- At $q = \frac{2\pi n}{d} \vec{e}$ there is a strong Bragg peak if d is the plane distance in the lattice for the planes with the normal of $\vec{e}$
- In every other case we see nothing

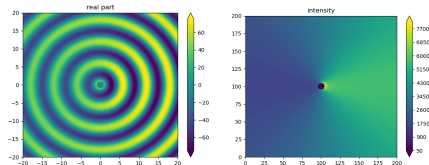


Simulated diffraction image of a cubic crystal using TOF diffraction

Scattering on atomic structures II

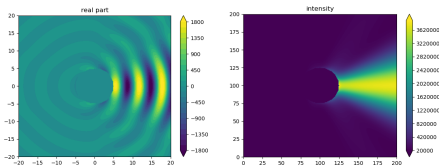
Example: Larger particles

If particle size is **comparable** to the wavelength, the intensity decreases in backward scattering (e.g. X-ray scattering factor)



Example: Large particles

If the particle larger than the wavelength, one cannot see the structure of the particle only its shape (Small Angle Scattering)



Properties of scattering function: scattering from monodisperse systems

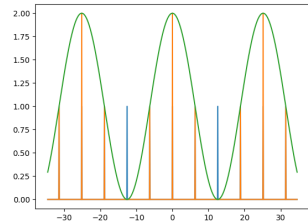
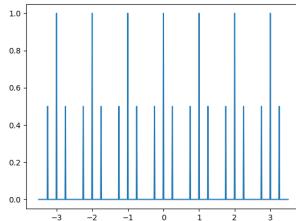
If the sample consists of structure of the same units (convolution):

- c : centers of units , $C = \mathcal{F}(\langle c, c \rangle)$ Intensity from correlation of the units
- u : unit: $U = \mathcal{F}(\langle u, u \rangle)$ scattering function of one unit
- full system is: $s = c * u$

$$s = l * u: \quad \langle s, s \rangle = \langle c, c \rangle * \langle u, u \rangle$$

$$I = \mathcal{F}(\langle s, s \rangle) = LU:$$

for crystals: L is a lattice U shows the heights of peaks of L :
 $|F(hkl)|^2$: Structure factor

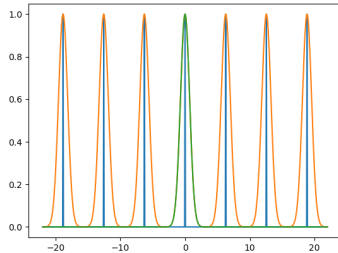


Properties of scattering function: non-ideal systems

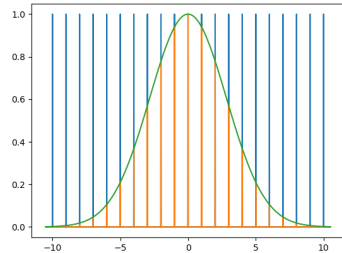
Use of convolution theorem

- Thermal motion: atomic positions are convoluted with a gaussian: peaks at large q are decreased: **Debye Waller effect**
- The finite size of a periodic system (e.g. nanocrystallites) infinite lattice is multiplied with the size distribution function:

Scherrer equation: The width of the peak is: $w = \frac{1}{d}$, d : size of cristallites, $k \approx 1$ is the shape factor

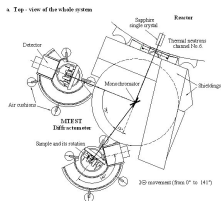


Debye - Waller
 $\rightarrow$
 $\leftarrow$
 Scherrer



Some examples: Elastic instruments

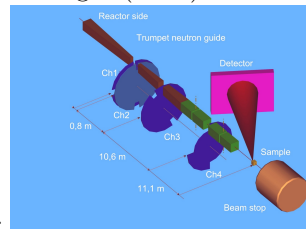
Monochromatic crystal diffractometer



Mtest diffractometer

- Monochromator
- Moving sample-table to change wavelength
- Moving detector to change angular range

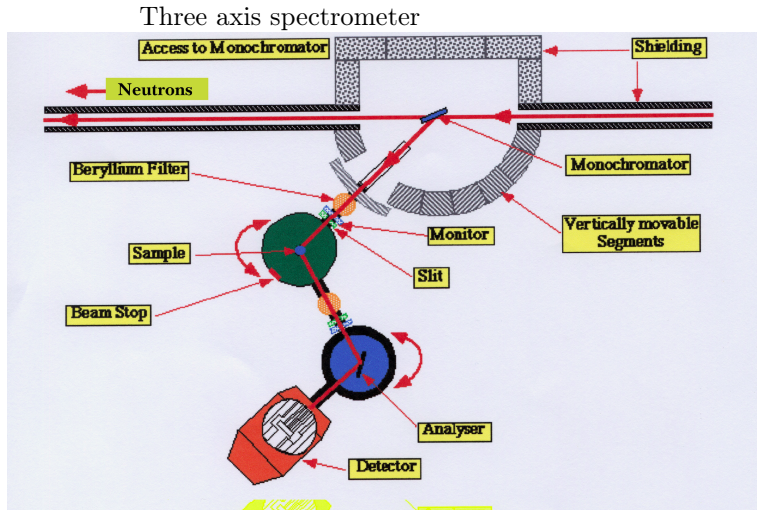
Time of flight (TOF)



Diffractometer

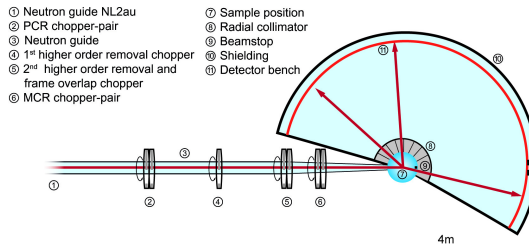
- Chopper produces pulses
- Large wavelength band
- Measurement mostly in one setup

Some examples: Three Axis Spectrometer

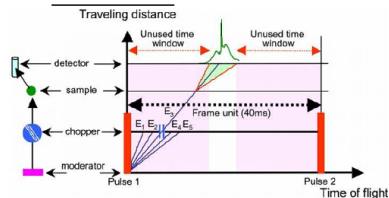


(crystal - crystal)

Some examples: Direct TOF spectrometer



- Monochromatic beam
 - ▶ TOF
 - ▶ Monochromator
- Chopper at sample
- Time of flight from the sample to the detector



Thank you for attention

Márton Markó

