School on Pulsed Neutrons - October 2005 - ICTP Trieste

"Methods and Techniques"

Experimental Techniques (I) Crystal & Time-of-Flight Spectrometers

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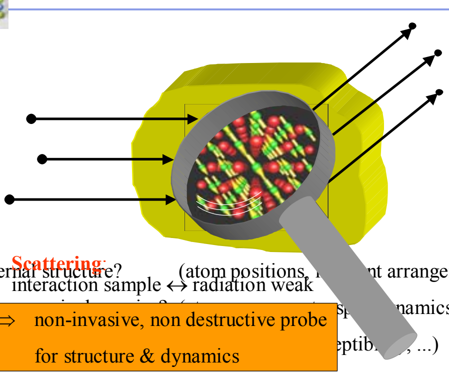
Institute for Solid State Research

Outline

- Scattering experiments
- Crystal spectrometers
- Time-of-flight spectrometers

Scattering



→ Internal structure? (atom positions, ... arrangement)

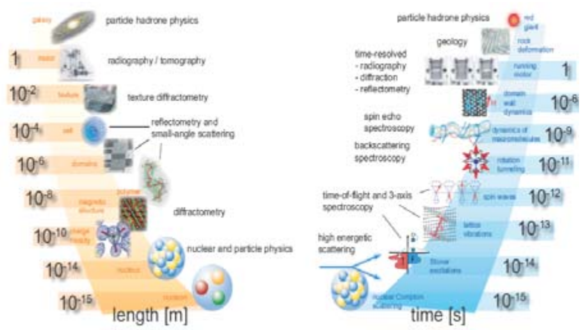
→ Mi...

⇒ M... ...eptib..., ...)

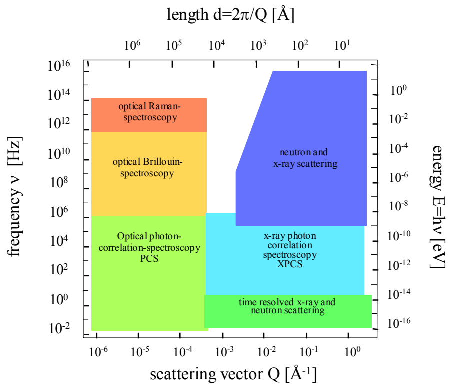
Scattering:
interaction sample ↔ radiation weak

⇒ non-invasive, non destructive probe for structure & dynamics

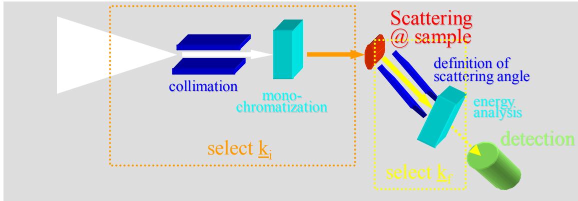
Neutrons: Length and Time Scales



Length- and Time Scales



Principle of Scattering Experiment



- define $\underline{k}_i$ ($k_i = 2\pi/\lambda_i$) and $\underline{k}_f$ with collimators and "monochromatizers"
- inelastic scattering (spectroscopy):
  determine change of neutron energy $E = \frac{\hbar^2 k^2}{2m}$ during scattering process
- two possibilities to define neutron energy E:
  - diffraction from single crystal (Neutron as wave)
  - time-of-flight (Neutron as particle)

Neutron

- matter wave:

  momentum $\underline{p} = m \cdot \underline{v} = \hbar \underline{k}$; $p = h/\lambda$

  energy $E = \frac{1}{2} m v^2 = \frac{\hbar^2 k^2}{2m} = \frac{h^2}{2m\lambda^2} \equiv k_B T_{eq}$

  in practical units:

$$\lambda[nm] \approx \frac{396}{v[m/s]} = \frac{6.28}{k[nm^{-1}]} \; ; \; \lambda[\mathring{A}] \approx \frac{3956}{v[m/s]}$$

$$E[meV] \approx \frac{0.818}{\lambda^2[nm]} = \frac{81.8}{\lambda^2[\mathring{A}]} \approx 0.0207 k^2 [nm^{-2}] = 2.07 k^2 [\mathring{A}^{-2}]$$

Example: "thermal neutrons"

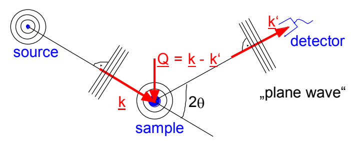
$T_{eq} = 300$ K　　E = 25 meV　　≈ ΔE elementary excitations.

λ = 0.18 nm　　≈ d atoms

v = 2 200 m/s

Fraunhofer Approximation

- Fraunhofer approximation: Ø sample << {distance source, distance detector}
  (far field approximation)



- scattering vector $\underline{Q} = \underline{k} - \underline{k}'$

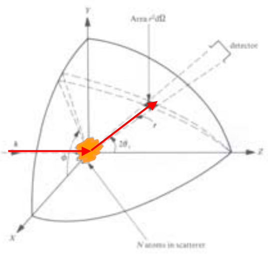
  $Q = |\underline{Q}| = \sqrt{k^2 + k'^2 - 2kk'\cos 2\theta}$

- elastic scattering (diffraction): $k = |\underline{k}| = |\underline{k}'| = k' = \frac{2\pi}{\lambda}$

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

Cross Section

→ **scattered intensity**



n': particles scattered per second into

dΩ: solid angle seen by detector under angle 2θ and into

dE': energy interval between E' and E' + dE'

j: incident beam flux (particles / (area · time))

double differential cross section:

$$\boxed{\frac{d^2\sigma}{d\Omega dE'} = \frac{n'}{j d\Omega dE'}} \qquad (I.4)$$

differential cross section:

$$\boxed{\frac{d\sigma}{d\Omega} = \int_{-\infty}^{\infty} \frac{d^2\sigma}{d\Omega dE'} dE'} \qquad (I.5)$$

total scattering cross section:

$$\boxed{\sigma = \int_{0}^{4\pi} \frac{d\sigma}{d\Omega} d\Omega} \qquad (I.6)$$

Inelastic Scattering

particle neutron … m — E_i → E_f

many body system M — E_0 → E_1

energy transfer

$$\Delta E \equiv \hbar\omega = E_1 - E_0 = E_f - E_i$$

momentum transfer

$$\hbar \underline{Q} = \hbar \underline{k}_f - \hbar \underline{k}_i$$

van Hove 1954 Phys. Rev. 95, 249

cross section ➡ scattering properties of the probe

× spatial dynamic correlations of the unperturbed system: $S(\underline{Q},\omega), (G(\underline{r},t))$

$$\frac{d\sigma^2}{d\Omega d\omega} = \left(\frac{d\sigma}{d\Omega}\right)_{\text{probe}} \frac{k_1}{k_0} S(\underline{Q},\omega)$$

nuclear scattering of neutrons: $\left(\frac{d\sigma}{d\Omega}\right)_{probe} = b^2 = \frac{\sigma}{4\pi}$

Scattering Law S(Q,ω)

Dynamic structure factor $S(\underline{Q},\omega) = \int dt S(\underline{Q},t) e^{i\omega t}$

integral: $\int S(\underline{Q},\omega) d\omega = \int dt S(\underline{Q},t) \underbrace{\int d\omega e^{i\omega t}}_{\delta(t)} = S(\underline{Q},0)$

⇒ snapshot

elastic: $S(\underline{Q},0) = \int dt S(\underline{Q},t)$

⇒ time average

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Reciprocal Lattice

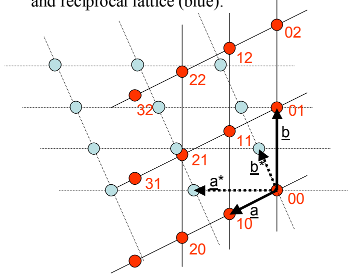
basis vectors of the reciprocal lattice:

$$\underline{a}^* = 2\pi \frac{\underline{b}\times\underline{c}}{\underline{a}\cdot(\underline{b}\times\underline{c})}$$

$$\underline{b}^* = 2\pi \frac{\underline{c}\times\underline{a}}{\underline{a}\cdot(\underline{b}\times\underline{c})}$$

$$\underline{c}^* = 2\pi \frac{\underline{a}\times\underline{b}}{\underline{a}\cdot(\underline{b}\times\underline{c})}$$

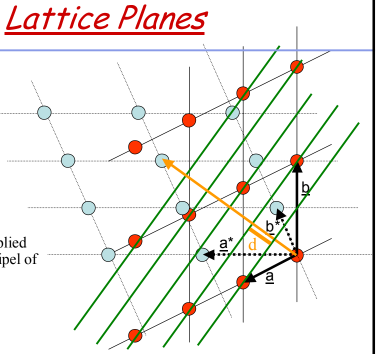
Example for 2d lattice (red) and reciprocal lattice (blue):



Lattice Planes

- Atoms lie on lattice planes
- parallel lattice planes form sets of lattice planes
- They are labeled by Miller indices h,k,l

the integer intercepts of the direct lattice are inverted and multiplied by an integer number to obtain a tripel of integer numbers.
In 2d example: Intercepts 2,1 →
Miller indices: (h,k) = (1,2)

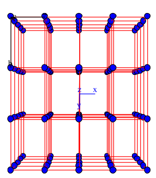


- The reciprocal lattice vector $\underline{G} = h\underline{a}^* + k\underline{b}^* + l\underline{c}^*$ is perpendicular to the lattice planes (h, k, l)
- $|\underline{G}| = 2\pi / d\,(h, k, l)$, where d (h, k, l) = distance of lattice planes along $\underline{G}$.

Laue Function

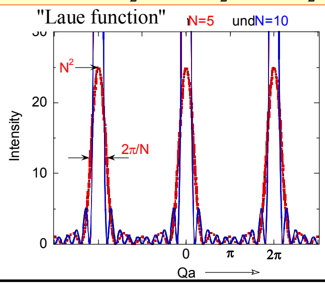
describes scattering from 3d lattice

$$|\Psi(\underline{Q})|^2 \sim |f(\underline{Q})|^2 \cdot \frac{\sin^2 \frac{1}{2} N \underline{Q}\cdot\underline{a}}{\sin^2 \frac{1}{2}\underline{Q}\cdot\underline{a}} \cdot \frac{\sin^2 \frac{1}{2} M \underline{Q}\cdot\underline{b}}{\sin^2 \frac{1}{2}\underline{Q}\cdot\underline{b}} \cdot \frac{\sin^2 \frac{1}{2} P \underline{Q}\cdot\underline{c}}{\sin^2 \frac{1}{2}\underline{Q}\cdot\underline{c}}$$



$$\underline{R}_{n,m,p} = n\cdot\underline{a} + m\cdot\underline{b} + p\cdot\underline{c}$$

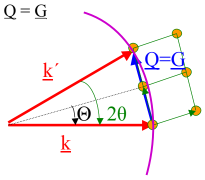
$$\Rightarrow \Psi(\underline{Q}) \sim \sum_{nmp} f_{nmp}(\underline{Q})\, e^{i\underline{Q}\cdot\underline{R}_{n,m,p}}$$



Bragg Condition

Laue function → main maxima for $\underline{Q} = \underline{G} = h\underline{a}^* + k\underline{b}^* + l\underline{c}^*$

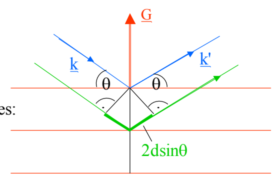
Ewald-construction:
- elastic scattering k = k' = 2π/λ
- $\underline{Q} = \underline{G}$



$$\sin\Theta = \frac{Q/2}{2\pi/\lambda} = \frac{G/2}{2\pi/\lambda} = \frac{2\pi/2d}{2\pi/\lambda}$$

⇒ Bragg condition:

$$\boxed{2d\sin\Theta = \lambda} \qquad \boxed{G = 2k\sin\theta}$$

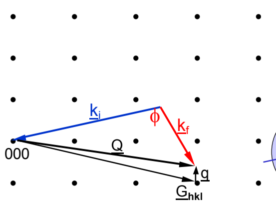


Interpretation:
scattering from set of lattice planes: constructive interference, if

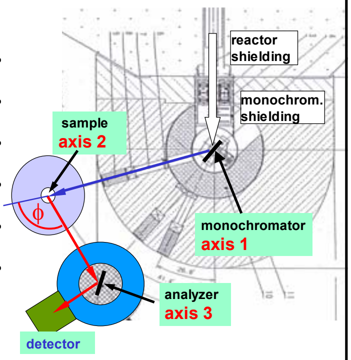
$$2d\sin\Theta = n\cdot\lambda$$

Triple-Axis Spectroscopy

. . . and in reciprocal space



in real space . . .



$$\underline{Q} = \underline{k}_f - \underline{k}_i = \underline{G}_{hkl} + \underline{q}$$

inelastic scattering !

Why Triple Axis Spectroscopy ?

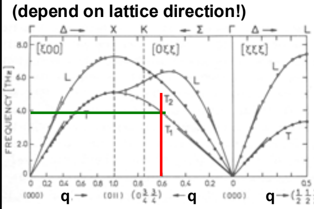
one can follow any desired path in reciprocal space

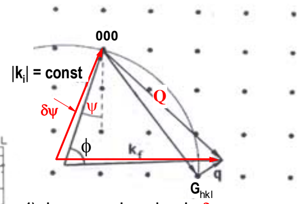
two typical scan types:

constant energy scan

constant Q scan

phonon dispersions in copper
(depend on lattice direction!)





1) change sample angle ψ by $\delta\psi$

2) calculate new $k_f(\delta\psi)$ so as to close the scattering triangle

3) change scattering angle ϕ by $\delta\phi(k_f, \delta\psi)$

4) set analyzer to new k_f, i.e. change θ_A and θ_D

5) go to 1)

Kinematic Conditions

a) Energy and momentum conservation

$\underline{Q} = \underline{k}_f - \underline{k}_i$

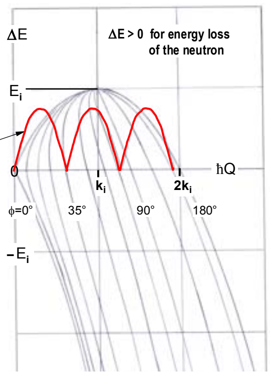
$Q^2 = k_i^2 + k_f^2 - 2\, k_i\, k_f \cos\phi$

$\Delta E = E_i - E_f = (\hbar^2/2m)(k_i^2 - k_f^2)$

$\Delta E = \Delta E(Q,\phi)$

dispersion relation for phonons
$\omega = \omega(\mathbf{q})$

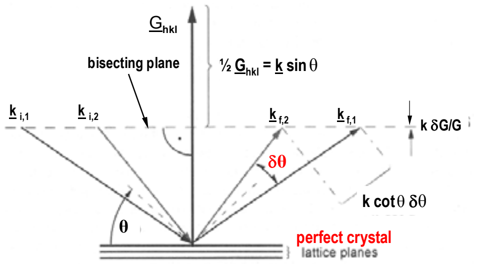
$\Delta E = \hbar\omega(\mathbf{q})$



3. Resolution and Intensity of a Crystal Spectrometer

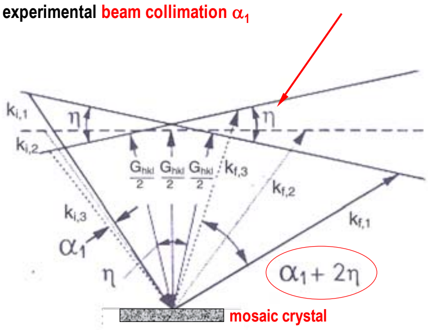
wave vector resolution δk_i of monochromator and analyzer for a perfect crystal and experimental beam collimation $\delta\theta$

$$G_{hkl} = 2\, k_i \sin\theta \quad\Rightarrow\quad \delta k_i = k_i \cdot (\,\delta G_{hkl}/G_{hkl} + \cot\theta \cdot \delta\theta\,)$$



Wave Vector Resolution δk_i

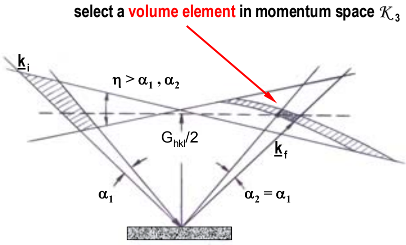
for a mosaic crystal with orientational variance η and experimental beam collimation α_1



Intensity $\propto$ Sizes of Resolution Volume Elements

by choosing mosaic spread η and collimations α_1 and α_2 before and behind monochromator / analyzer

select a volume element in momentum space $\mathcal{K}_3$



in fact the 3-dimensional shape of the volume elements is ellipsoidal

Intensity (Quantitative)

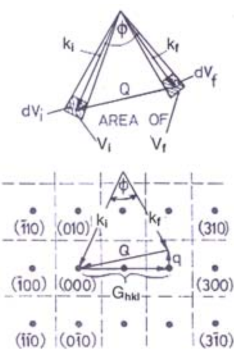
source →	primary spectrometer →	sample →	secondary spectrometer
	1. collimator monochromator 2. collimator		3. collimator analyzer 4. collimator detector
$(\Phi/k_T)f(k)d^3rd^3k$	$dj(k_i)$	$n_S\, t\, dxdy\, [d^2\sigma/d\Omega dE_f]$	$\Delta\Omega\Delta\omega$
	$(\Phi/k_T)\, f(k_i)\, k_i\, d^3k_i$	$n_S\, t\, dxdy\, [(k_f/k_i)(\sigma/4\pi)S(\underline{Q},\omega)]$	$\Delta\Omega = \Delta(k_f)_x\Delta(k_f)_y / k_f^2$ $\Delta\omega = (\hbar/m)\, k_f\Delta k_f$

Intensity at the detector:

$$\Delta I_D = dj(k_i)\, n_S t\, dxdy\, d^2\sigma/d\Omega dE_f\, \Delta\Omega\Delta\omega = (\Phi/k_T) f(k_i)\, n_S t\, dxdy\, (\sigma/4\pi)\, S(\underline{Q},\omega)\, (\hbar/m)\, \Delta(k_f)_x\Delta(k_f)_y\, \Delta k_f\, d^3k_i$$

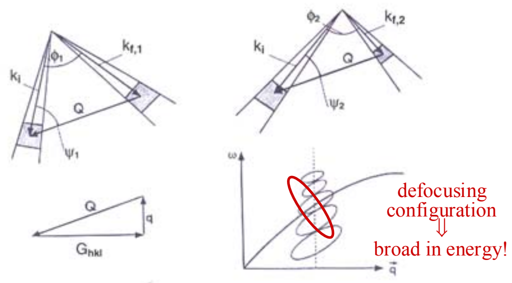
$$\Delta I_D = (\Phi/k_T)\, f(k_i)\, dxdy\, n_S\, t\, (\sigma/4\pi)\, S(\underline{Q},\omega)\, (\hbar/m)\, d^3k_i\, \Delta^3k_f$$

4. Resolution and Focusing in Inelastic Scattering

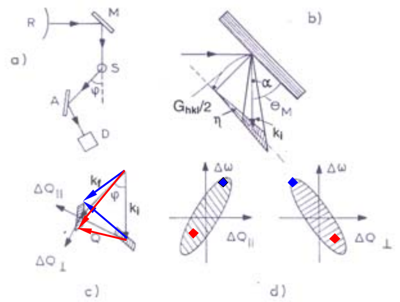


Focusing

adapt the inclination of the resolution ellipsoid to the slope of the dispersion curve

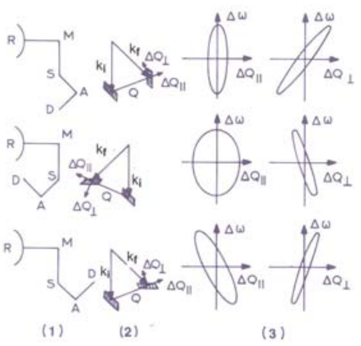


Visualization of the Inclination of the Resolution Ellipsoid



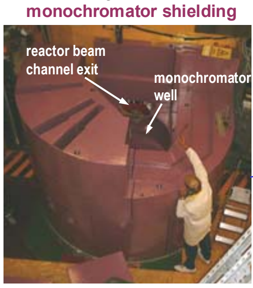
Visualization of the Inclination of the Resolution Ellipsoid

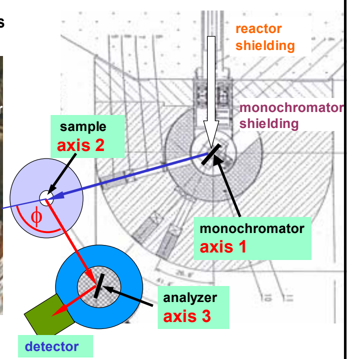
How to select size and inclination of the resolution ellipsoid



5. The Jülich Triple Axis Spectrometer SV-30

SV-30 during set-up of the 70 tons monochromator shielding



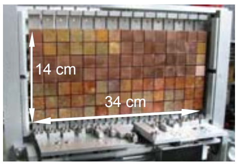


The SV-30 Double Focusing Monochromators

available incident wavelengths λ_i and energies E_i at SV30

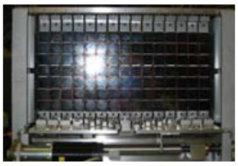
Cu[200]

λ [Å]	E [meV]
0.84	**117**
1.28	**50**
2.18	**17**

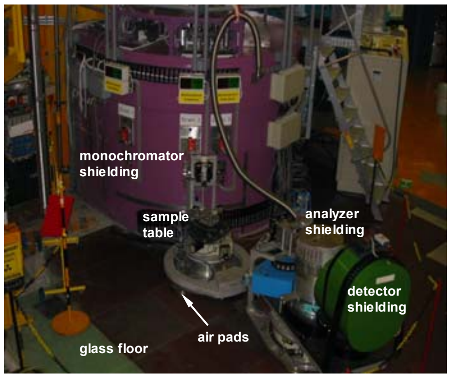


PG[002]

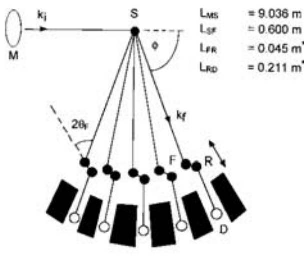
λ [Å]	E [meV]
1.55	**34**
2.37	**14**
4.05	**5**



SV-30 Completed



PRISMA @ ISIS



Schematic drawing of the PRISMA-3 detector

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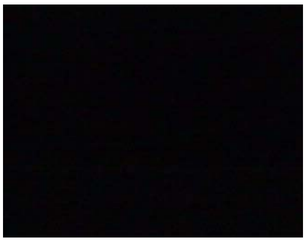
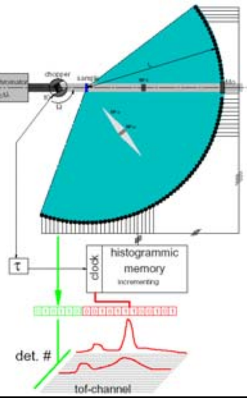
Generic TOF spectrometer





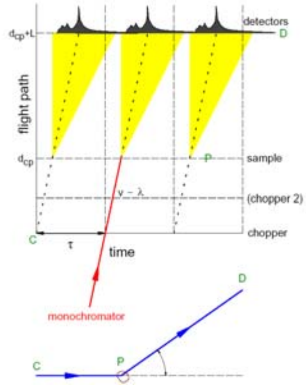
Path-time diagram

$$p = \frac{h}{\lambda} = m \cdot v$$

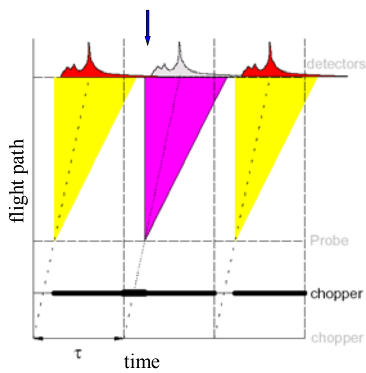
$$\& \;\; v = \frac{s}{t}$$

$$t = \frac{m}{h} \cdot s \cdot \lambda$$

(typically 1 ms/m)

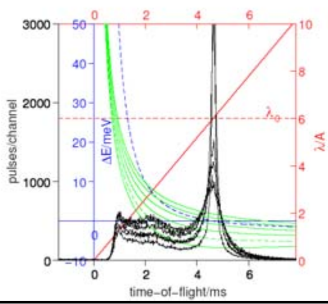


Frame Overlap



Example

- Liquid water (H_20) at T=300 K
- $\Theta = 30^0 \cdots 140^0$
- $H_20 \rightarrow$ incoherent scattering



Time to Energy Spectrum

$$I(\Delta t,\theta) \rightarrow S(Q,\omega)...$$

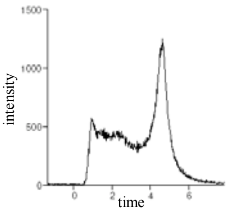
$t = L/v'$ and $t_0 = L/v$ yields

$$\omega(t) = \frac{m_n}{2\hbar} L^2 \frac{t^2 - t_0^2}{t^2 t_0^2}$$

and

$$Q = \frac{m_n}{\hbar} L \sqrt{\frac{t^2 + t_0^2 - 2cos(2\Theta) t_0 t}{t_0^2 t^2}}$$

Strongly varying energy-width of TOF-channels, K !



Energy Spectrum

$$I(2\Theta, K) \propto \int_{(K-1)\Delta\tau}^{K\Delta\tau} \frac{k'}{k} 4\pi b^2 S(\underline{Q}, \omega(t)) \frac{d\omega}{dt} dt$$

or somewhat simpler

$$I(2\Theta, K) \propto \frac{k'}{k} 4\pi b^2 S(\underline{Q}, \omega(K\Delta\tau)) \frac{d\omega}{dt} \Delta\tau$$

with

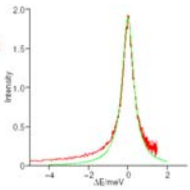
$$\frac{k'}{k} = \frac{t_0}{t}$$

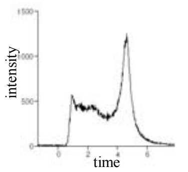
and

$$\frac{d\omega(t)}{dt} = \frac{m_n}{\hbar} L^2 \frac{1}{t^3}$$

it follows:

$$I(2\Theta, K) \propto S(\underline{Q}, \omega(K\Delta\tau)) \; \frac{1}{t^4}$$

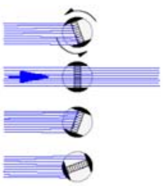


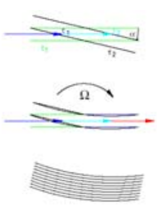


Fermi Chopper

$$\frac{\text{time open}}{\text{time closed}} \approx 1\%$$

Fermi-Chopper
rotating collimator
$\Delta\alpha \simeq 1^0$
$\Delta\tau \simeq \Delta\alpha/\Omega$
$\Delta\tau \simeq 10\mu s$
$\Omega \simeq 2\pi 200\text{Hz}$

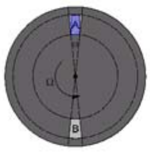




Curved slit collimator
wavelength selectivity
higher λ harmonics suppression

Disk Chopper

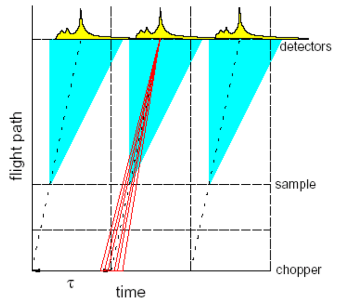
TOFTOF @ FRM-II



$$\frac{\text{time open}}{\text{time closed}} \approx 10\ \%$$

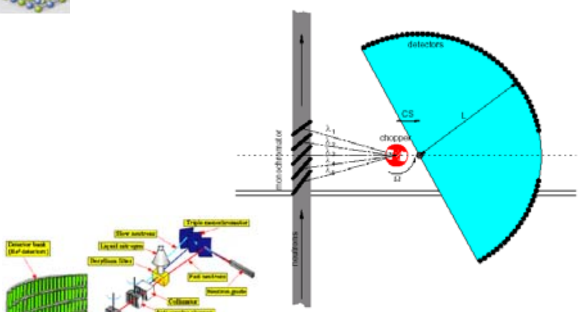
⇒ use sequence of disk choppers and/or counter rotating choppers

Time Focusing



– time sequence of wavelength $\lambda_0(t)$ at start
– slow neutrons first, fast last
– convergence in arrival time at detector plane

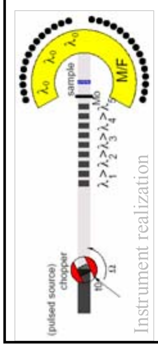
Time Focusing Instrument

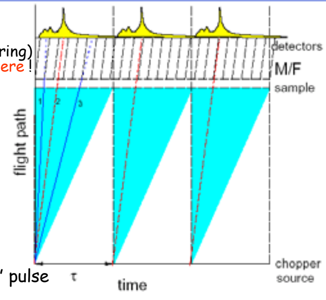


– slow neutrons first, fast last
– convergence in arrival time at detector plane
– energy transfer of focus depends on Ω

Inverted Geometry

Monochromatization (λ-filtering)
Done here !





"white" pulse

– "white" pulse at start (wide λ-band)
– only λ_0 neutrons pass the analyzer/filter (M/F)
– at sample $\lambda = \lambda(t)$
– at detector $\Delta\lambda(t) = \lambda_0 - \lambda(t)$

Resolution vs. Intensity

$\Delta\omega \sim 1/\lambda^3$

- $I \sim \Delta t$ -
- $\sim \Delta\lambda$ -
- $\sim \Delta\Omega$ +
- $\sim \Phi(\lambda) \sim \lambda^{-3..5}$ -

Inverted geometry:
Exchange λ and λ'

$$\Delta\omega = \sqrt{(\frac{\partial\omega}{\partial t}\Delta t)^2 + (\frac{\partial\omega}{\partial\lambda}\Delta\lambda)^2}$$

With

$$\hbar\omega = (\hbar^2/2m_n)(k^2 - k'^2)$$

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

$$\omega(t) = \frac{m_n}{2\hbar}L^2\frac{t^2 - t_0^2}{t^2 t_0^2}$$

it follows

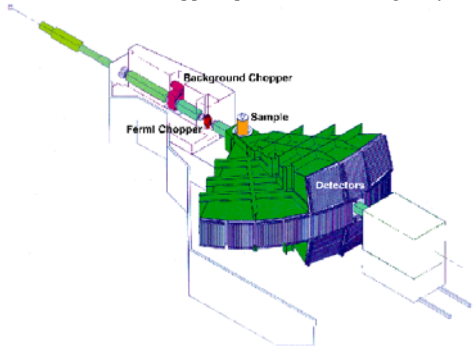
$$\Delta\omega = \sqrt{(\frac{m_n L}{\hbar t^3}\Delta t)^2 + (\frac{4\pi^2\hbar}{m_n\lambda^3}\Delta\lambda)^2}$$

$\lambda' \propto 1/v' \propto t$
$\lambda' \approx \lambda$ close to the elastic line
$\rightarrow \Delta\omega \propto 1/\lambda^3$
in some more detail....

$$\Delta\omega = \sqrt{(\frac{m_n L}{\hbar t^2}\Delta L)^2 + (\frac{m_n L}{\hbar t^3}\Delta t)^2 + (\frac{4\pi^2\hbar}{m_n\lambda^3}\Delta\lambda)^2}$$

MAPS @ ISIS

Chopper Spectrometer for single crystal studies



Acknowledgements

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