Neutron scattering, facilities and instrumentation

Robert Georgii

Literature

Neutron scattering: A Primer by Roger Pynn
Los Alamos Science (1990)
http://library.lanl.gov/cgi-bin/getfile?19-01.pdf

Elementary Scattering Theory: For X-ray and neutron users
D.S. Sivia (2011)
St John's College, Oxford
ISBN 978–0–19–922867–6

Properties of the neutron

- Mass: m_n = 1.675 x 10^{-27} kg
- Charge = 0
- Spin = 1/2
- Magnetic moment: m_n = - 1.913 μ_B

- Velocity v, kinetic energy E, wavelength λ, wavevector k, moderator temperature T

$$E = \frac{1}{2} mv^2 = k_B T = {\left(\frac{hk}{2\pi}\right)^2}\Big/_{2m} \text{, where } k = {}^{2\pi}\!/_{\lambda} = {}^{mv}\!\Big/_{\frac{h}{2\pi}}$$

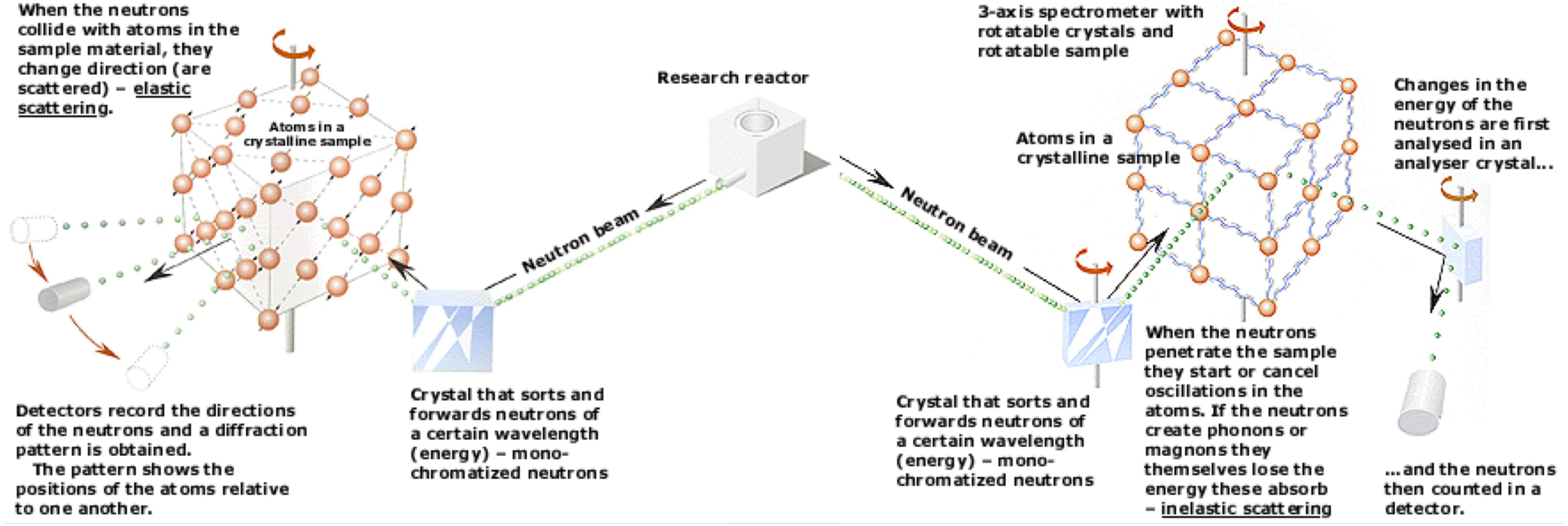
	Energy (meV)	Temperature (K)	Wavelength (Å)
cold	0.1 – 10	1 – 120	4 – 30
thermal	5 – 100	60 – 1000	1 – 4
hot	100 – 500	1000 – 6000	0.4 – 1

Nobel prize 1994 to Shull and Brockhouse

Neutrons see

Where atoms are

How atoms move



When the neutrons collide with atoms in the sample material, they change direction (are scattered) – elastic scattering.

Detectors record the directions of the neutrons and a diffraction pattern is obtained.
The pattern shows the positions of the atoms relative to one another.

Crystal that sorts and forwards neutrons of a certain wavelength (energy) – monochromatized neutrons

3-axis spectrometer with rotatable crystals and rotatable sample

Crystal that sorts and forwards neutrons of a certain wavelength (energy) – monochromatized neutrons

When the neutrons penetrate the sample they start or cancel oscillations in the atoms. If the neutrons create phonons or magnons they themselves lose the energy these absorb – inelastic scattering

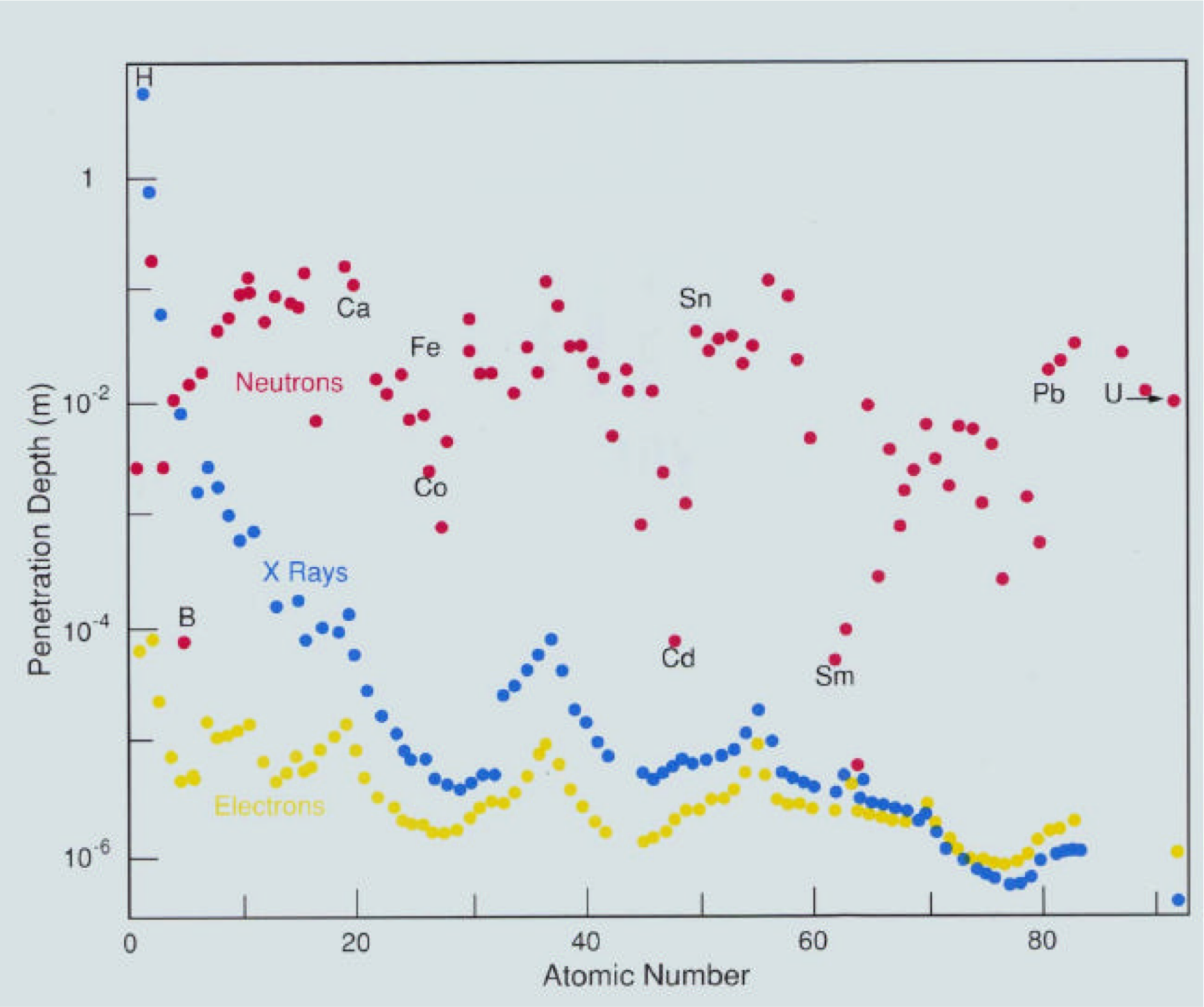
Changes in the energy of the neutrons are first analysed in an analyser crystal...

...and the neutrons then counted in a detector.

Why we use neutrons

- Advantages:
  - Wavelength is in the order of atomic distances (≈ 1Å = 10^{-10} m)
  - Energy is in the order of the kinetic energy of atoms (≈ meV) and much smaller as the binding energy (≈ eV)
  - Large penetration depth since uncharged particles
  - Scattering is dependent on the isotopic composition (Difference H,D)
  - Neutrons have a magnetic Moment, they "see" B-fields
- Disadvantages:
  - Neutron sources have a very low Brilliance
  - Neutrons are difficult to detect, guide and to shield

Comparison of different probes

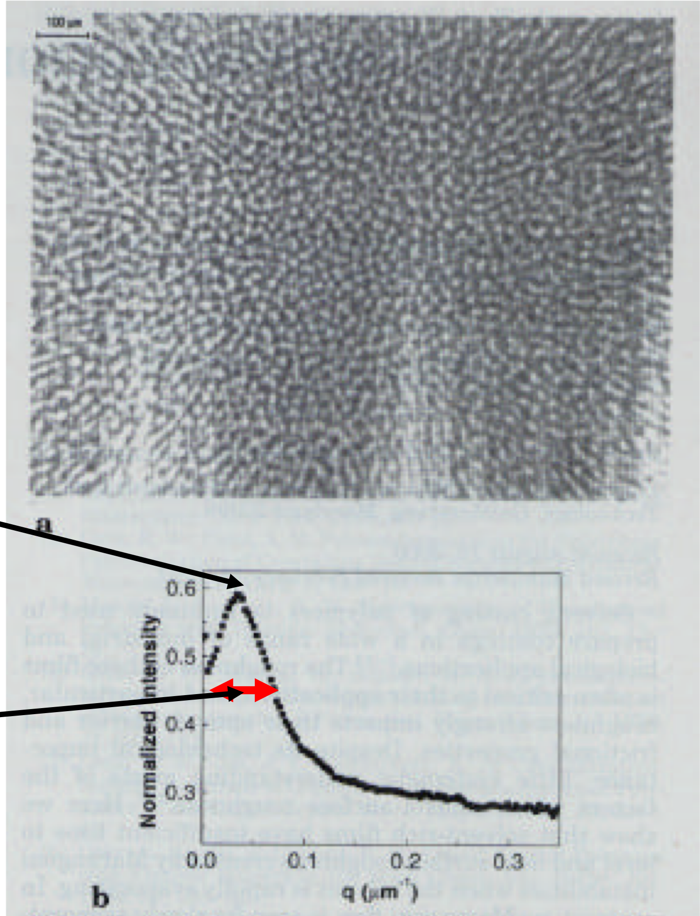


Neutrons:

- no systematic A-dependence
- Specific strongly absorbing isotopes: B, Cd, Sm, Gd
- Large difference for H/D

Scattering versus imaging measurements

- Imaging techniques are done in real space, like using a microscope
- Scattering techniques work on an ensemble of objects in reciprocal space
- Both methods are complementary

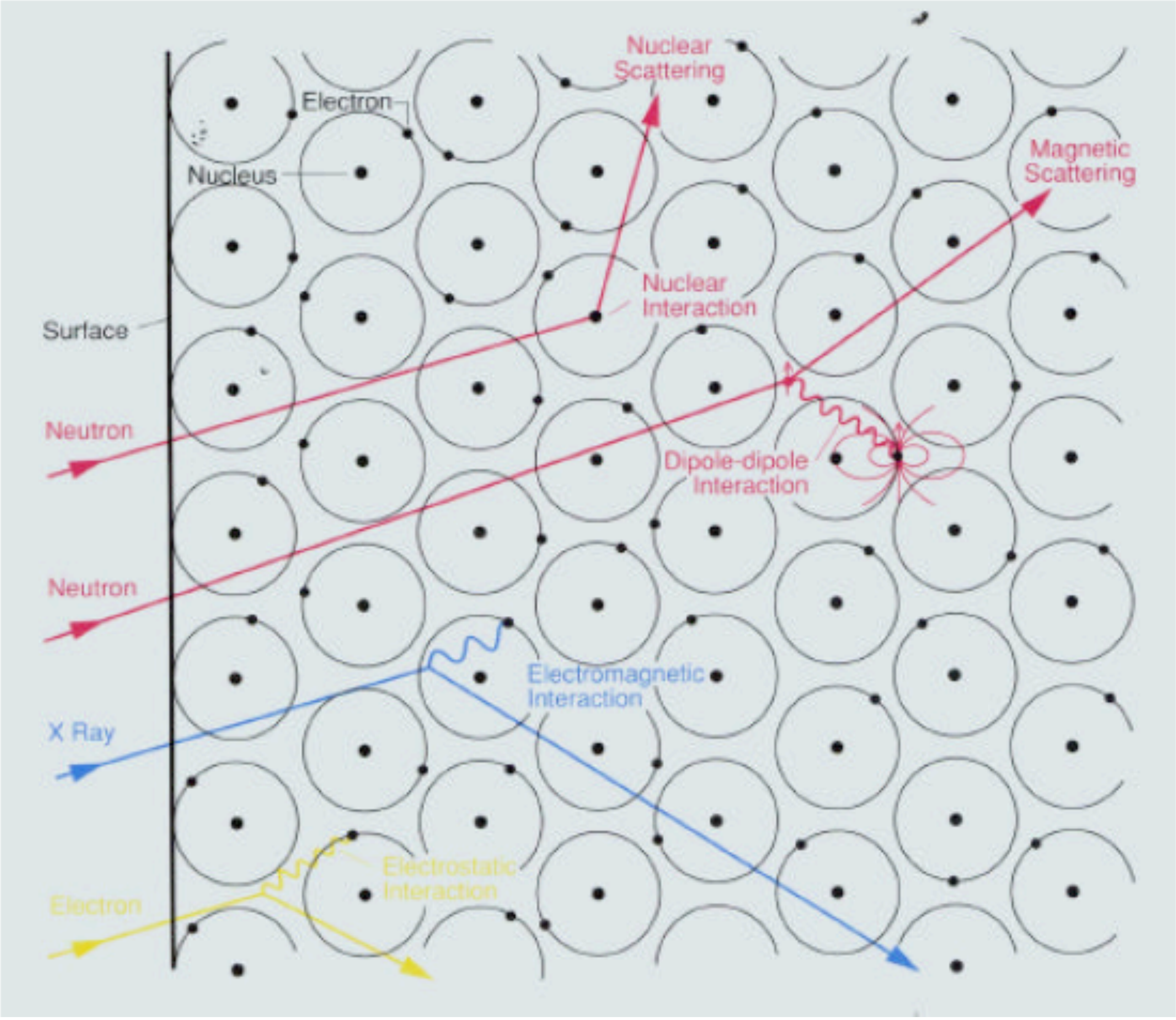


Average size

Standard variation

Foam in the sub micrometre range:
Picture from a microscope

Interaction mechanism

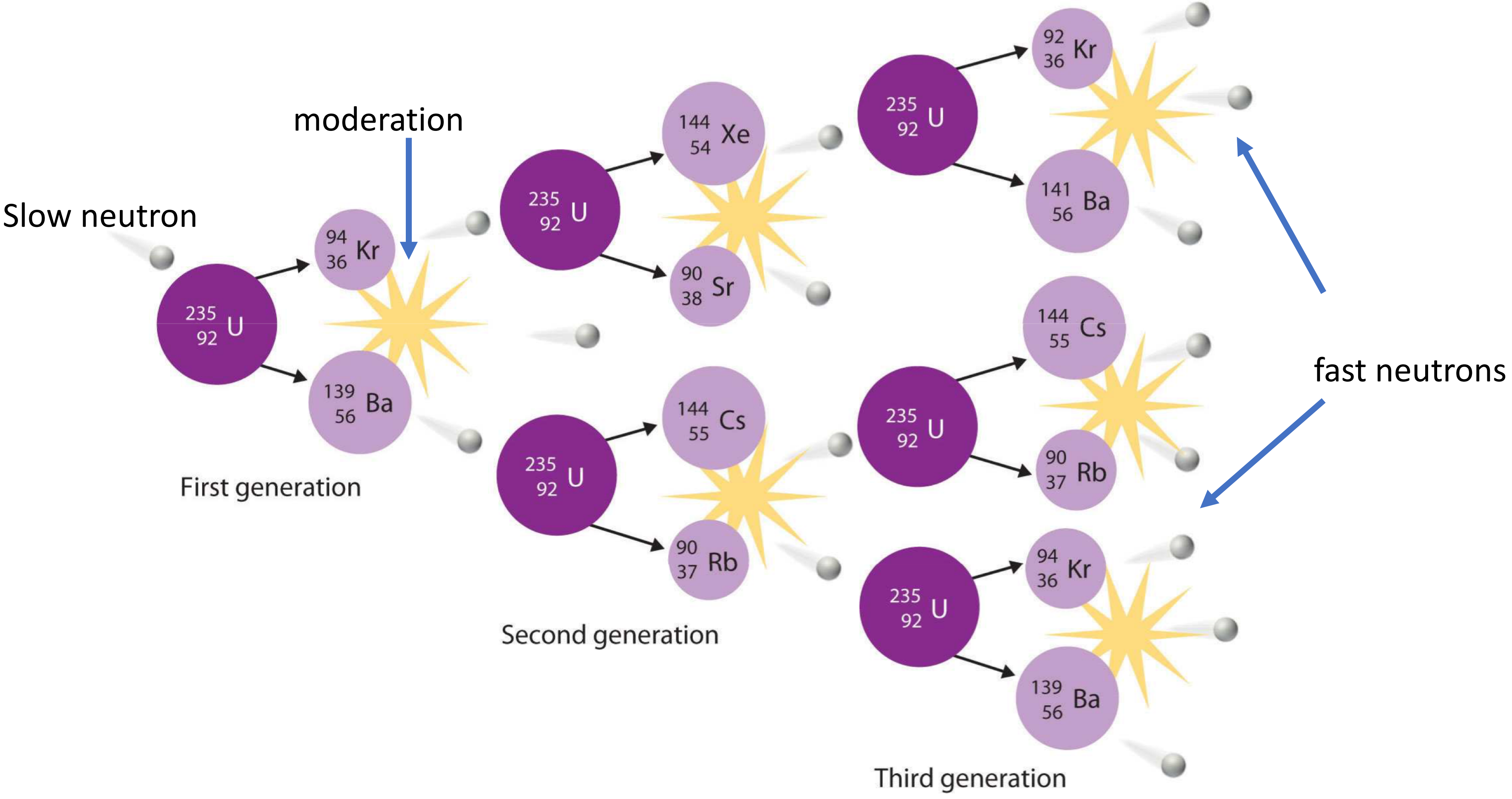


Interaction of neutrons

- only with the nucleus
  (point interaction ~ fm)

- with unpaired electrons
  (magnetic Dipol-Dipol interaction)

Fission: Chain reaction

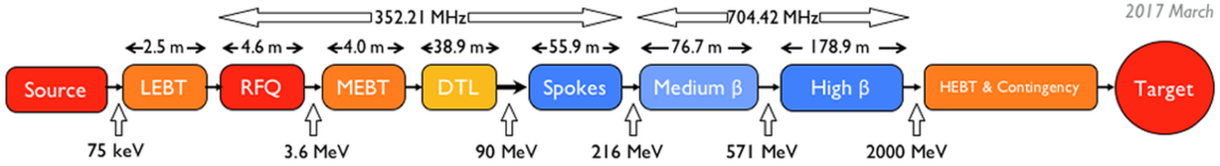


Stefan Doege – Annalisa Polidori, ILL Ph.D. Seminar, 2015

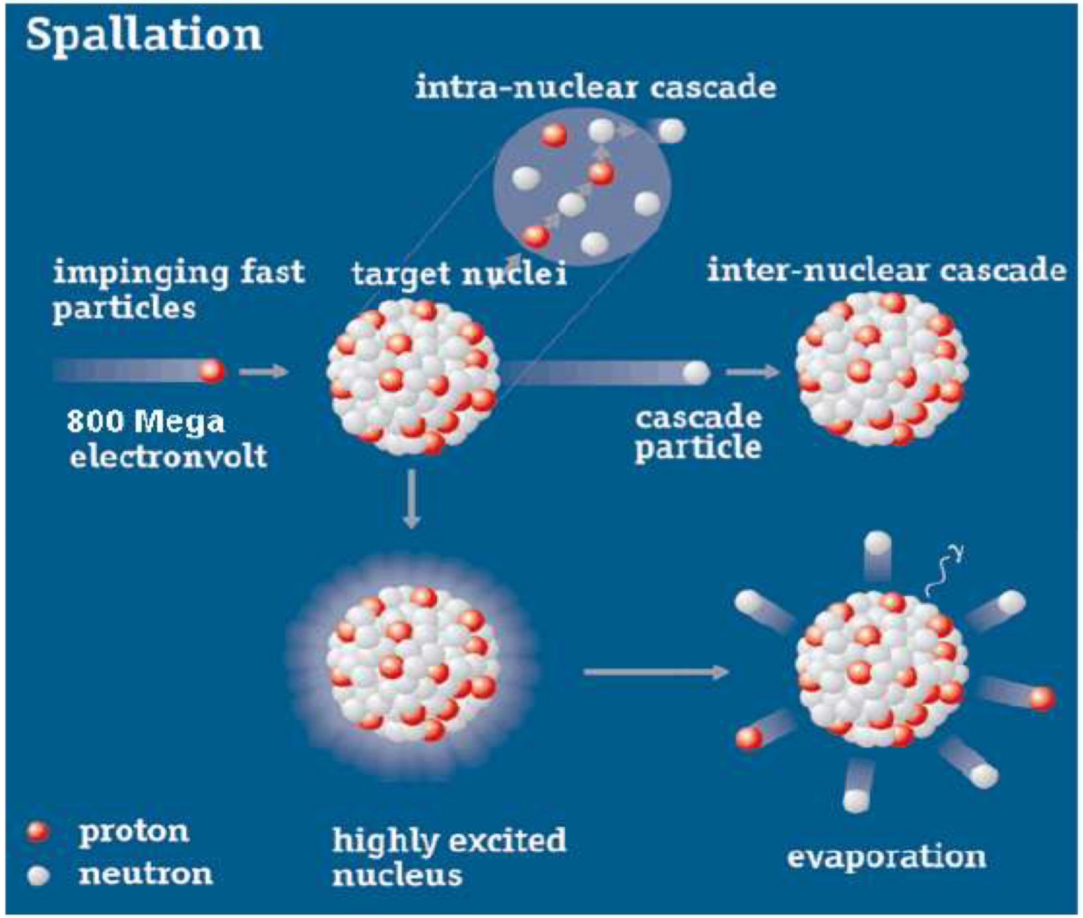
Spallation: Proton accelerator + heavy metal target

ESS: The world most powerful accelerator for the highest neutron flux

Accelerator for protons: 2 GeV and 5 mA



Tungsten target, He cooled

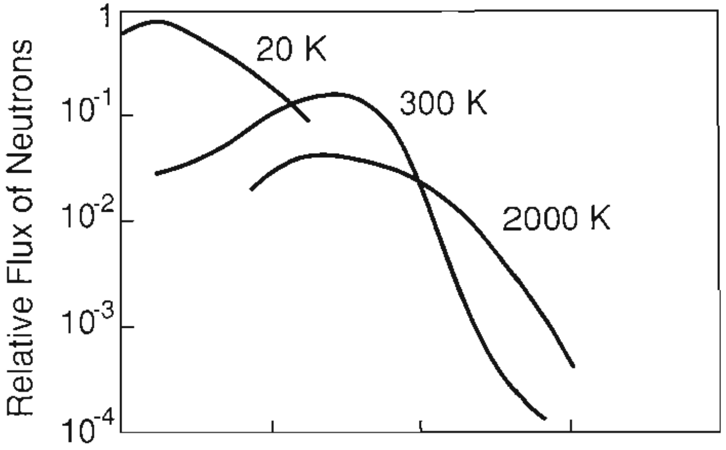


ESS Homepage

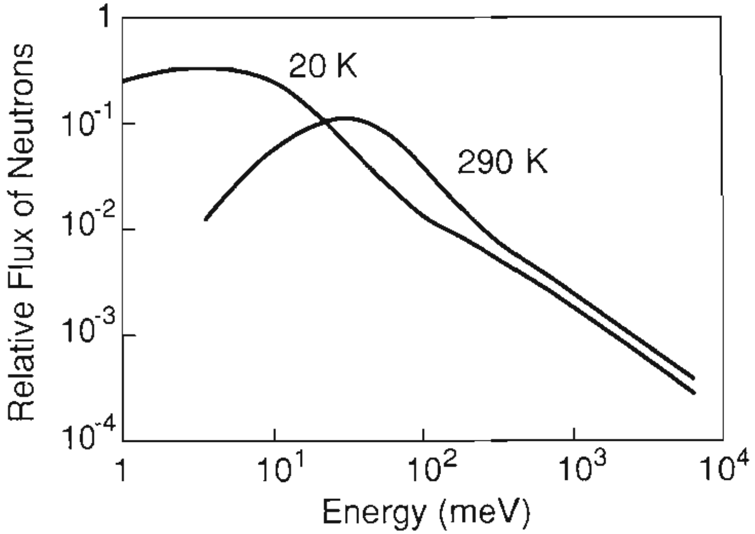
Fission versus Spallation

	Fission	Spallation
Energy per neutron	180 MeV	20 MeV
Neutron spectrum	Maxwellian	long tail of hot neutrons
Wavelength resolution	can be adopted to needs	constant
Time structure	continuous	pulsed
Stability	very stable	depended on accelerator
Problems	Nuclear reactor	much higher n energies
Building costs	About 0.5 Billion €	About 2 Billion €
Running costs	Current for pumps	Current for accelerator
Further improvements	Saturation reached	Higher accelerator energy

Reactor spectrum



Spallation spectrum



European Landscape of Neutron User Facilities

ESS
Sweden

ISIS
UK

ORPHÉE
France

ILL
France

PIK
Russia

JINR
Russia

BER II
Germany

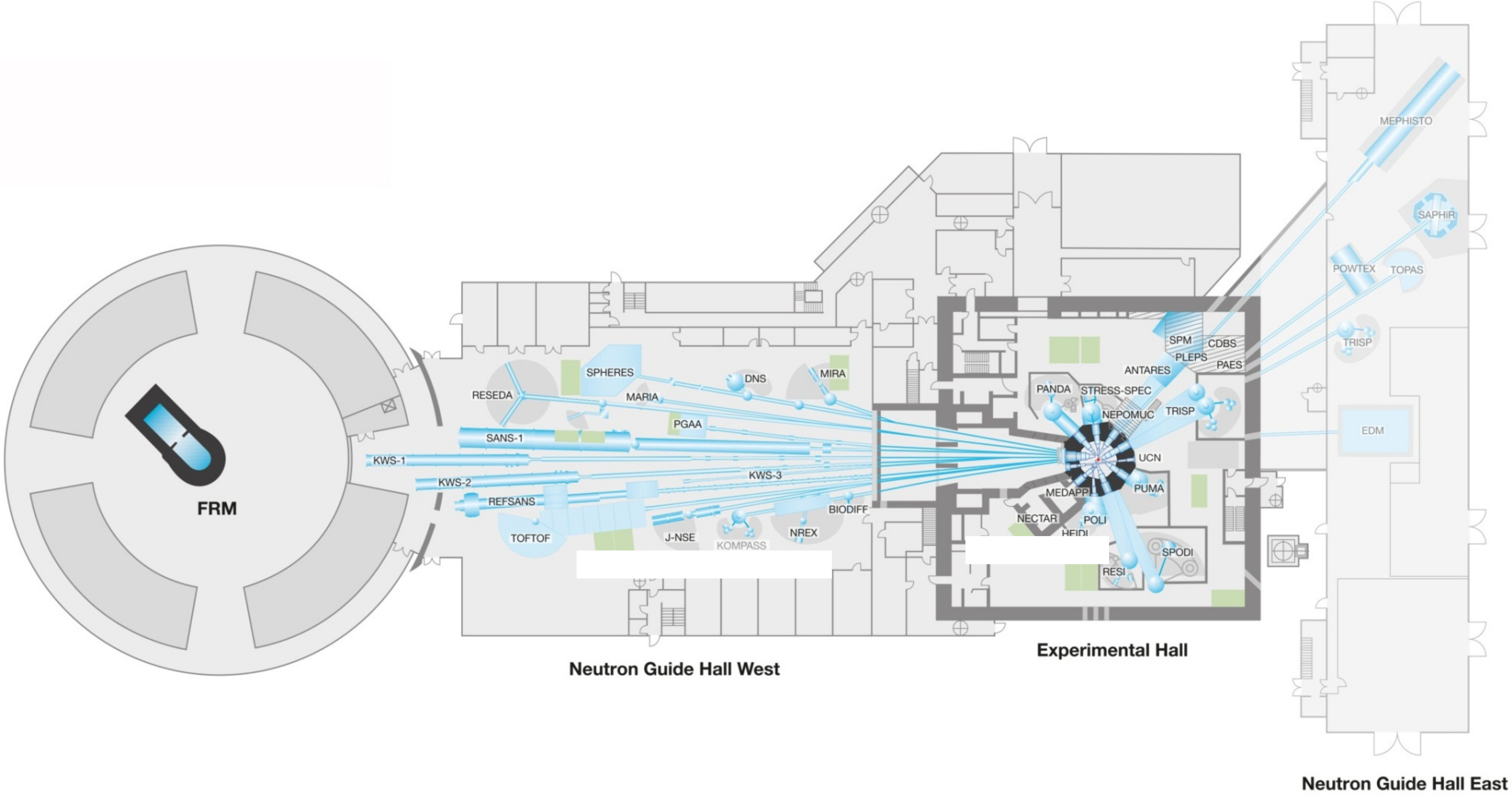
FRM II
Germany

SINQ
Switzerland

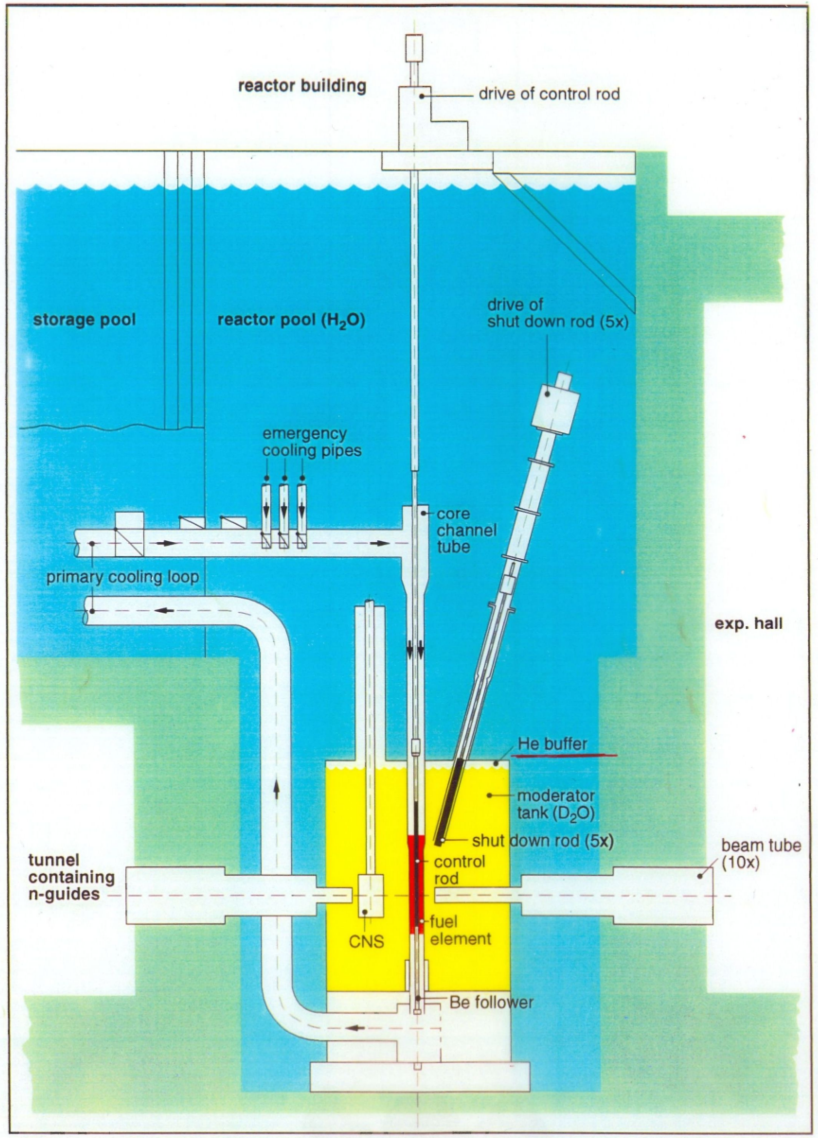
International Neutron Sources

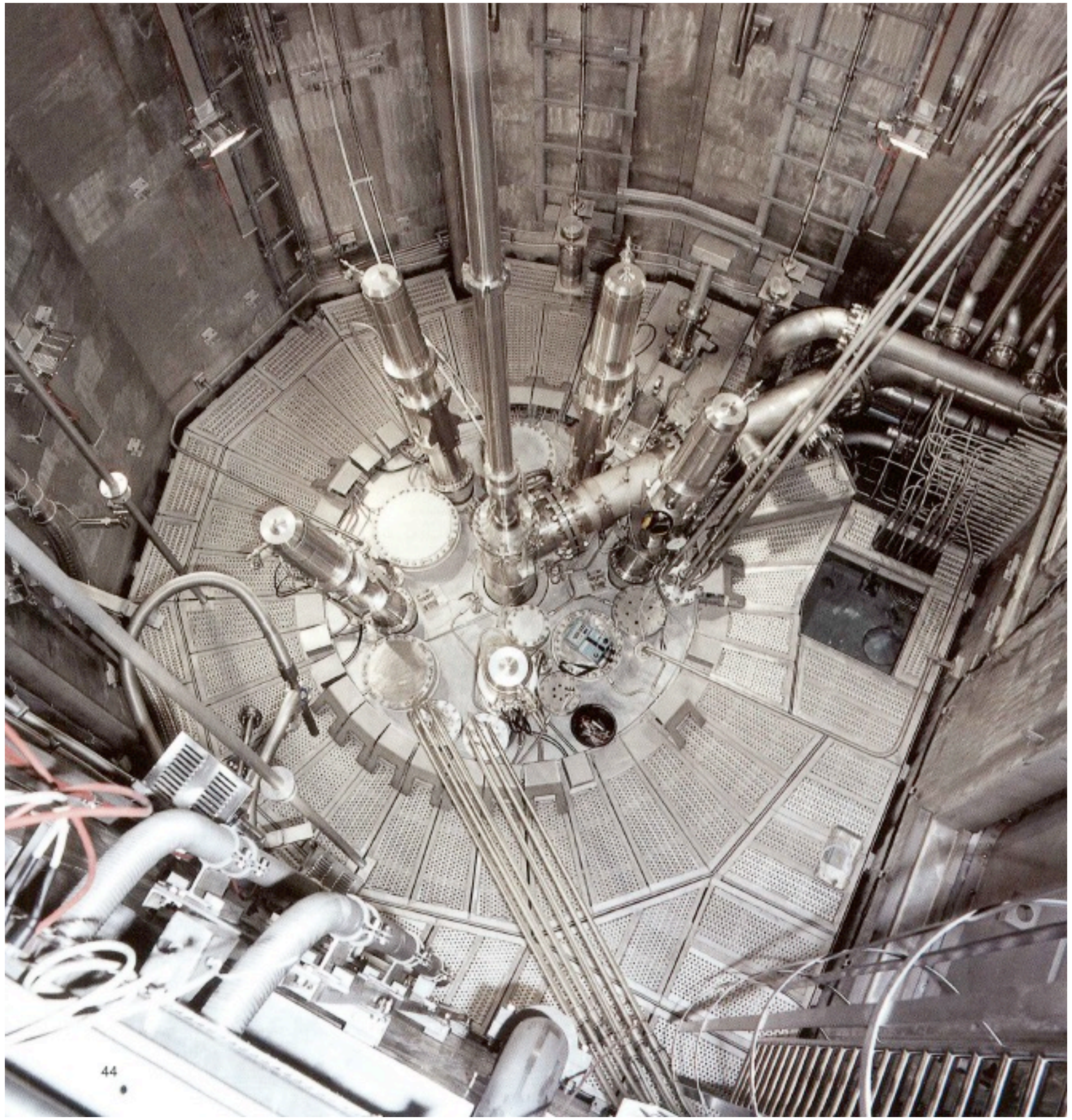
	ILL Grenoble (F)	IBR Dubna (Russia)	ISIS Chilton (GB)	FRM II	ESS Lund (S)	SNS Oak Ridge (USA)
$\Phi\ [cm^{-2}s^{-1}]$	10^{15}	$2\ x\ 10^{16}$	$4.5\ x\ 10^{15}$	$7\ x\ 10^{14}$	$1.5\ x\ 10^{17}$	$8\ x\ 10^{16}$
$\bar{\Phi}\ [cm^{-2}s^{-1}]$	10^{15}	$2\ x\ 10^{13}$	$7\ x\ 10^{12}$	$7\ x\ 10^{14}$	$0.6\ x\ 10^{15}$	$6\ x\ 10^{13}$
Pulse repetition rate [Hz]	-	5	50	-	14	60
Pulse duration [μs]	-	250	30	-	2860	20
P [MW]	57	2	0.2	20	5	2

The Neutron Source FRM II

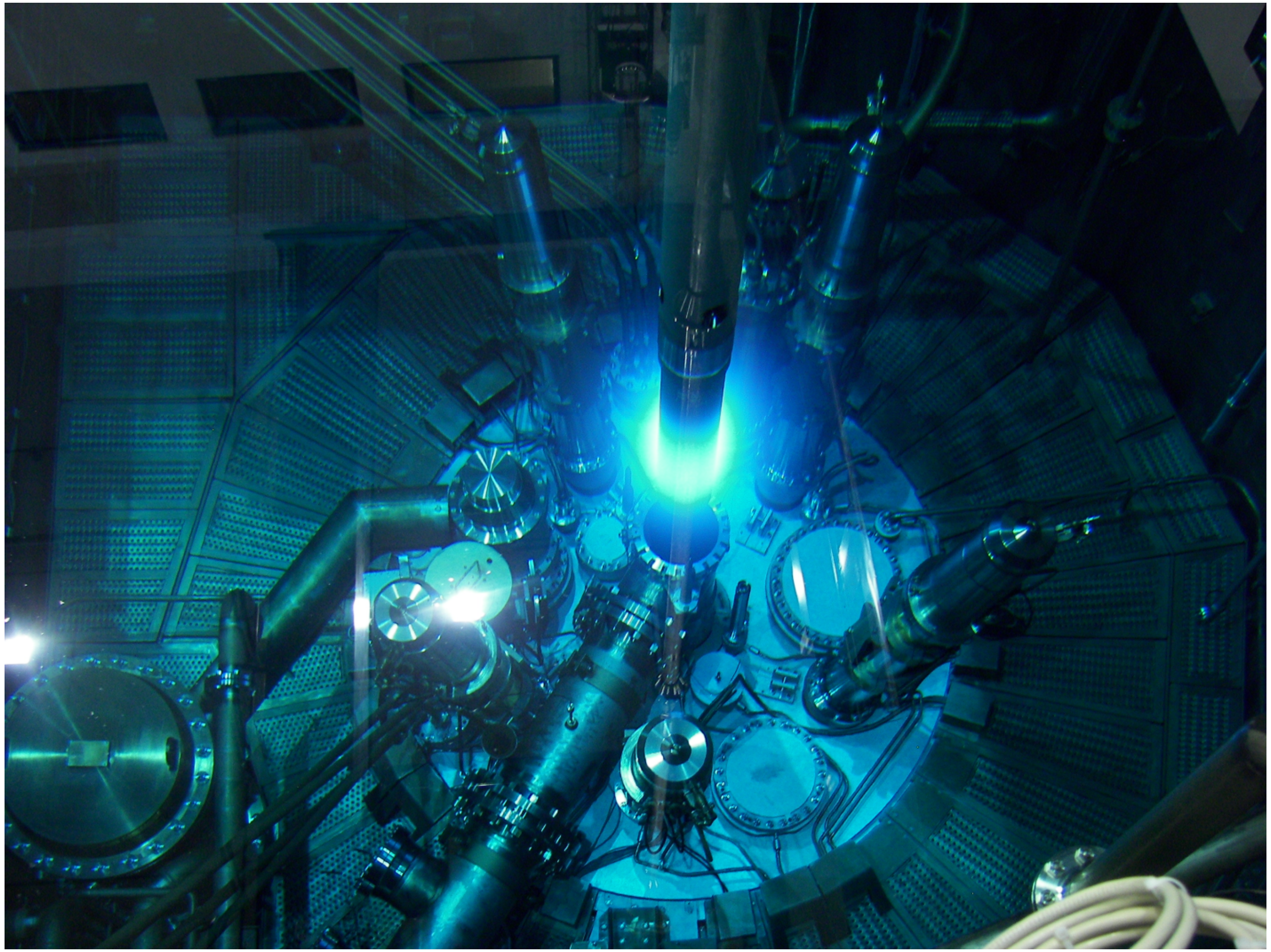


FRM II: A swimming pool reactor

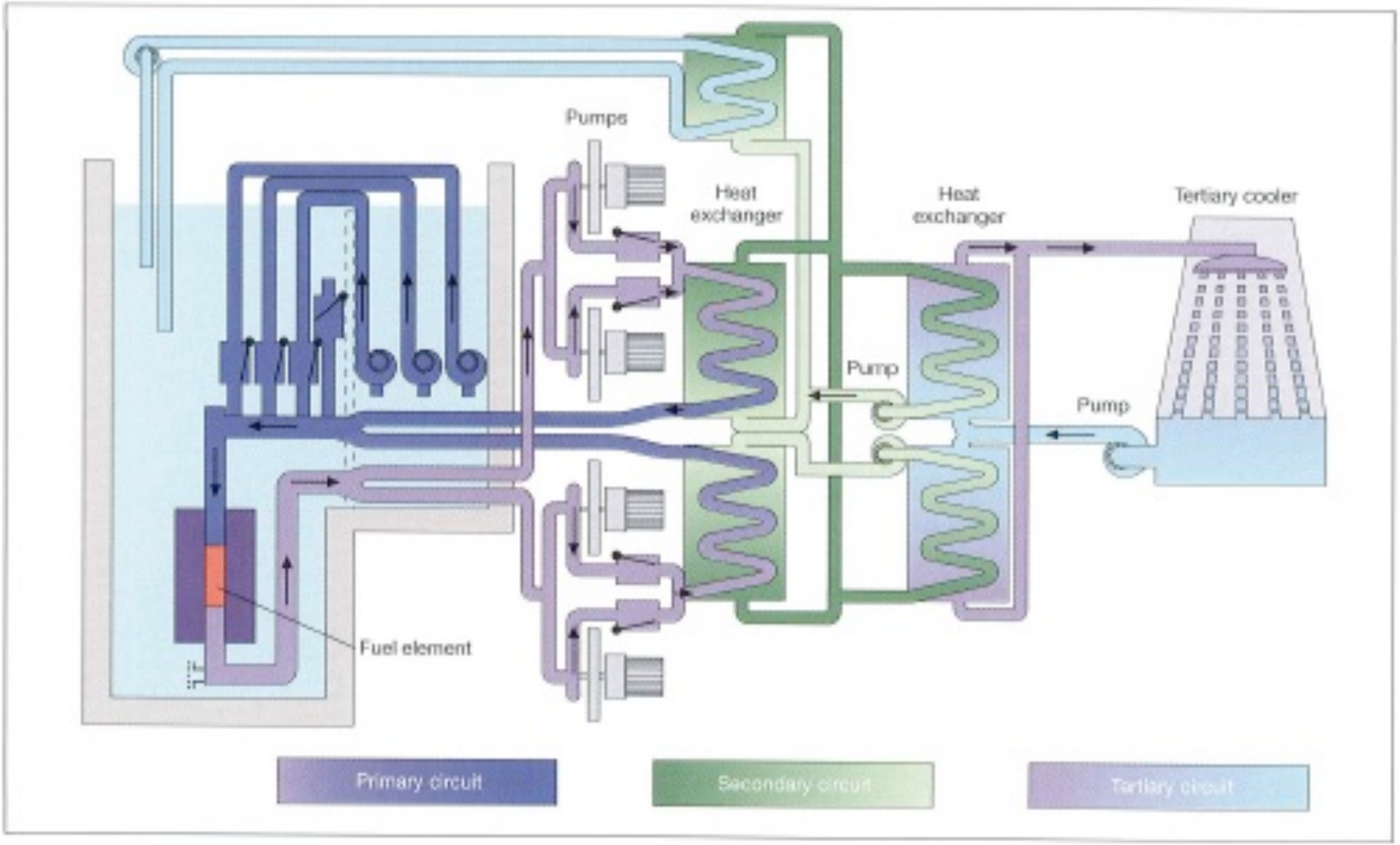




FRM II: Discharge of a spent fuel element

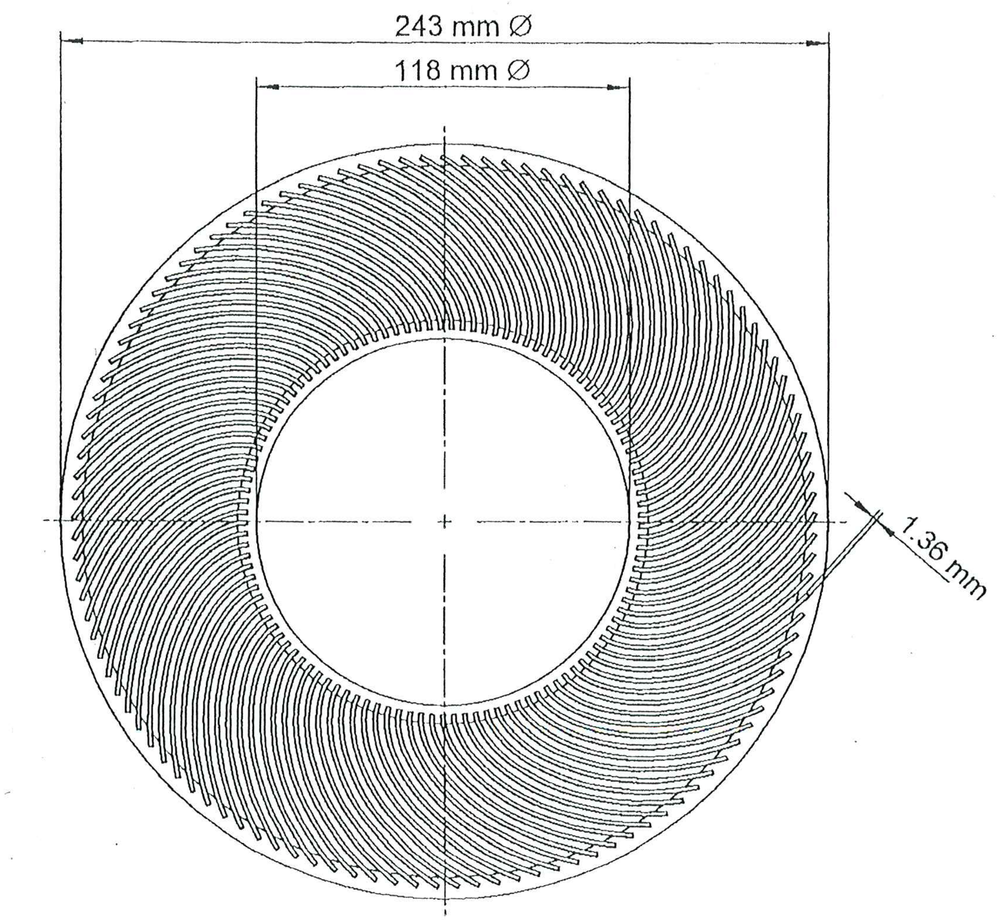


FRM II: Cooling system

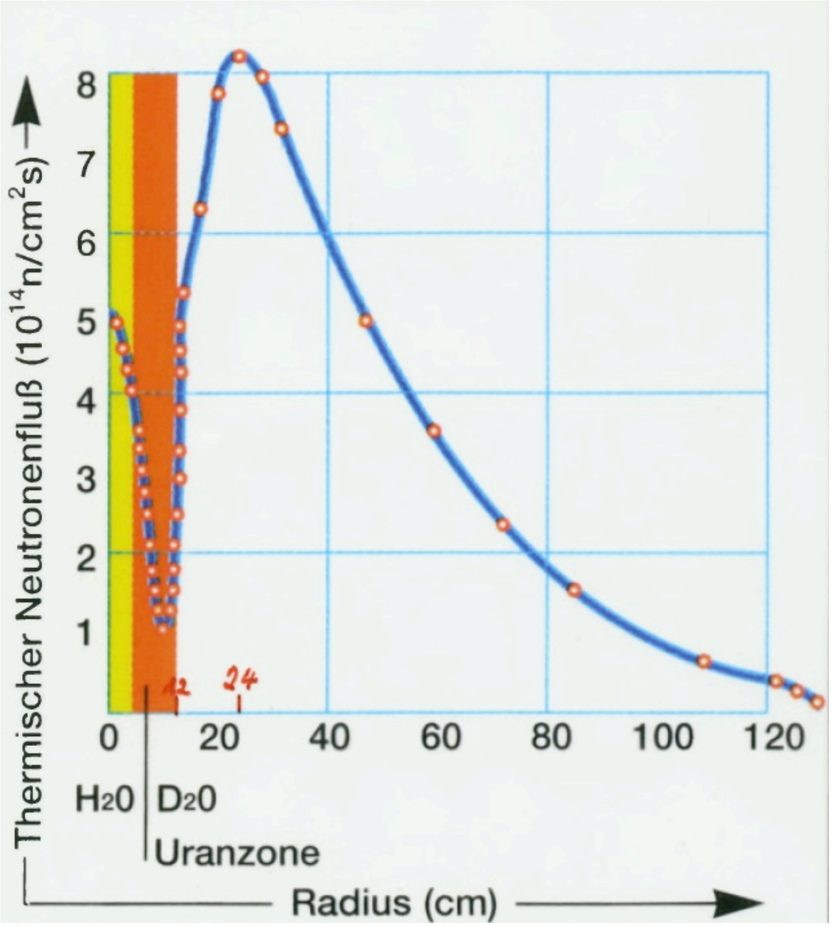


FRM II: The Fuel Element

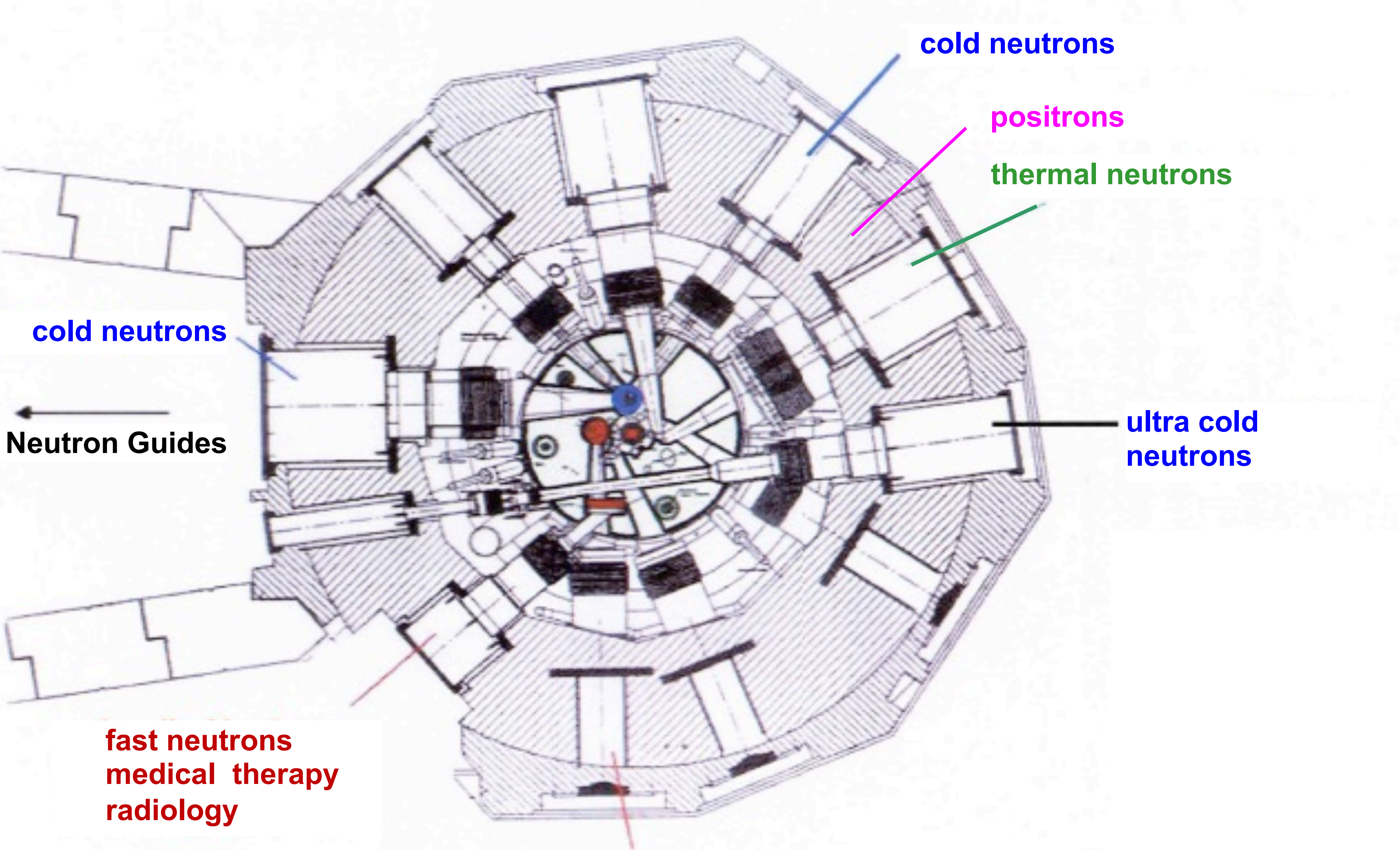
Cross section: 113 curved fuel plates



Flux maximum 12 cm above fuel element



FRM II: The reactor vessel



cold neutrons

positrons

thermal neutrons

cold neutrons

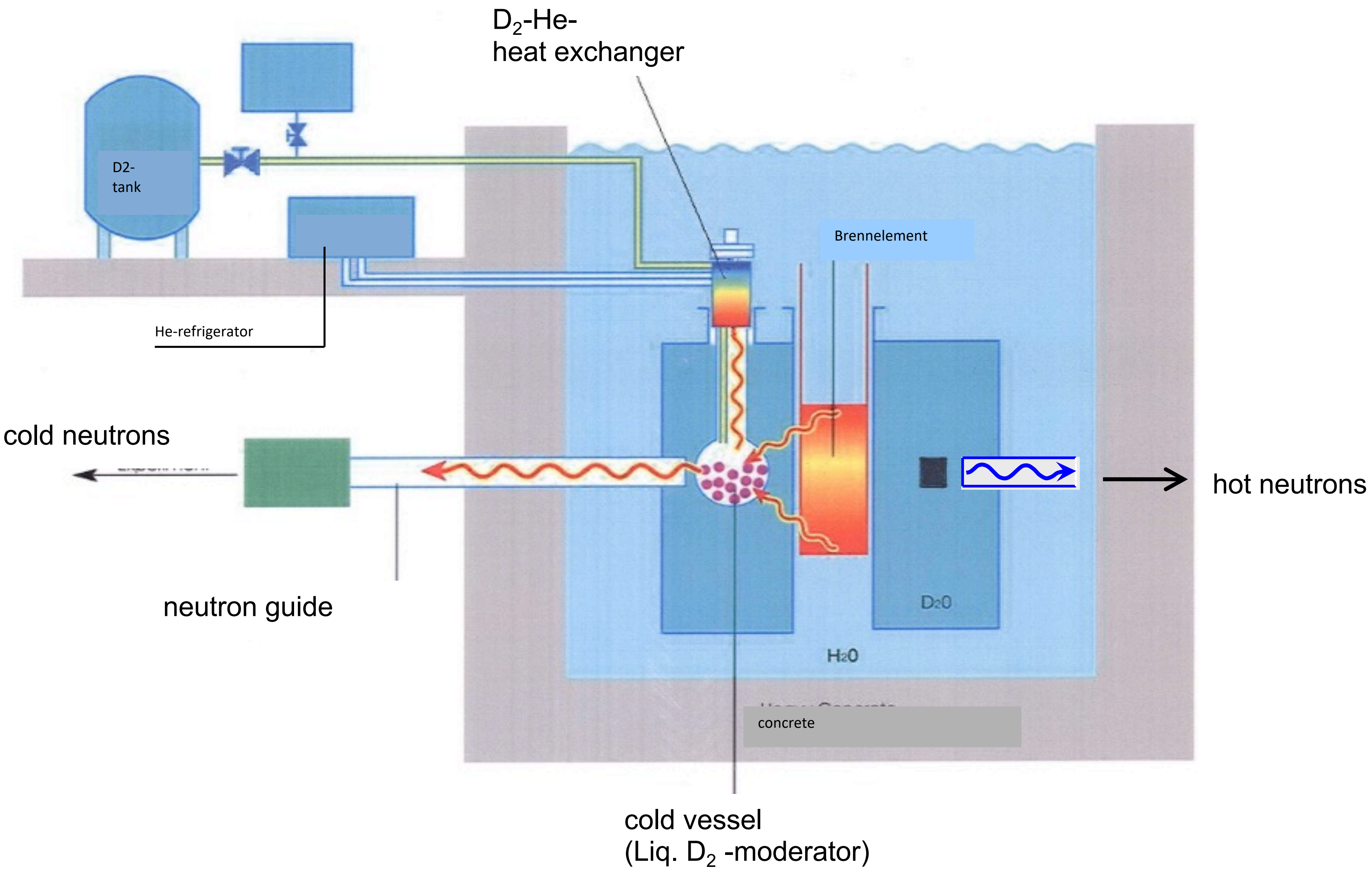
Neutron Guides

ultra cold neutrons

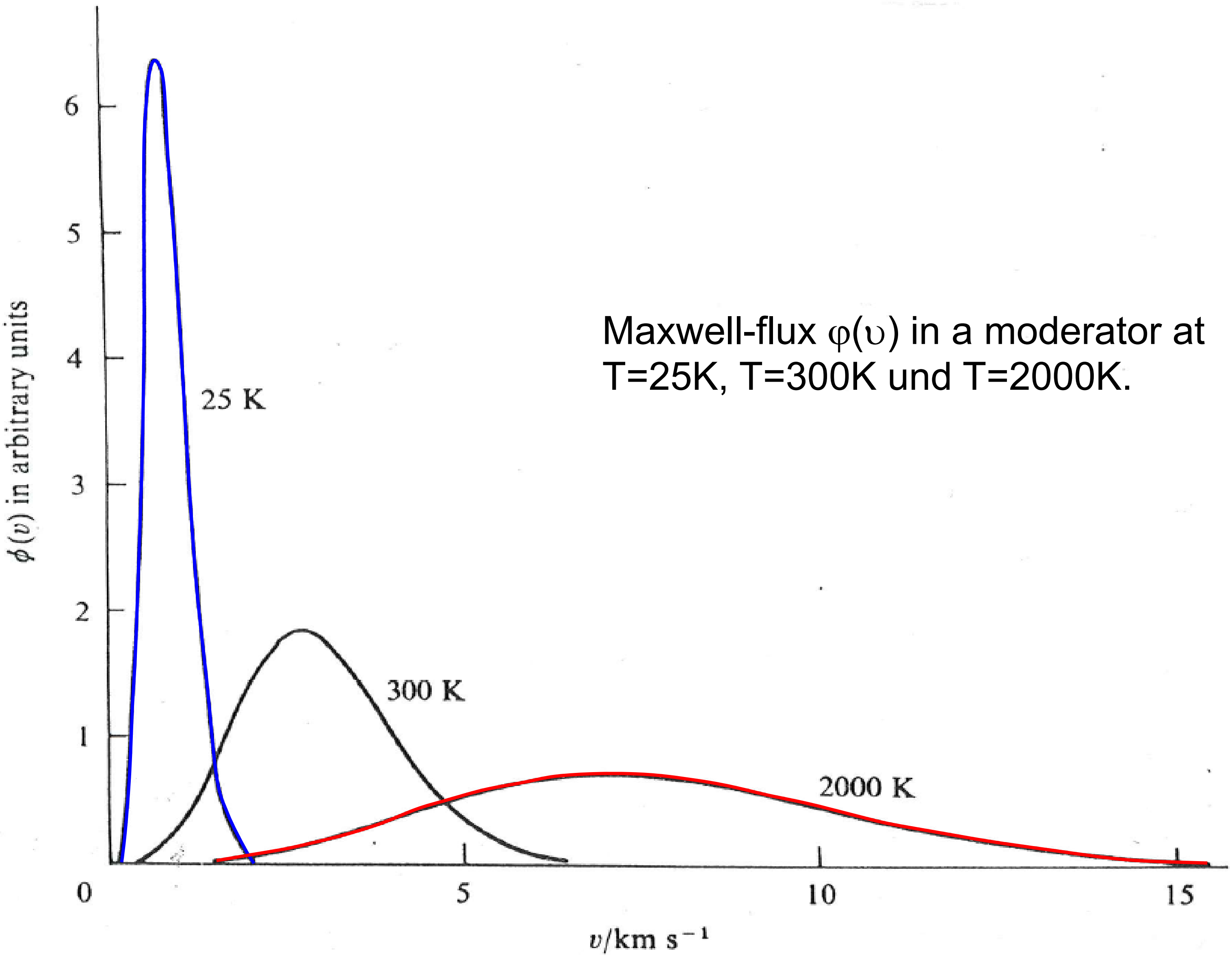
fast neutrons
medical therapy
radiology

hot neutrons

FRM II: Cold Source, Hot Source

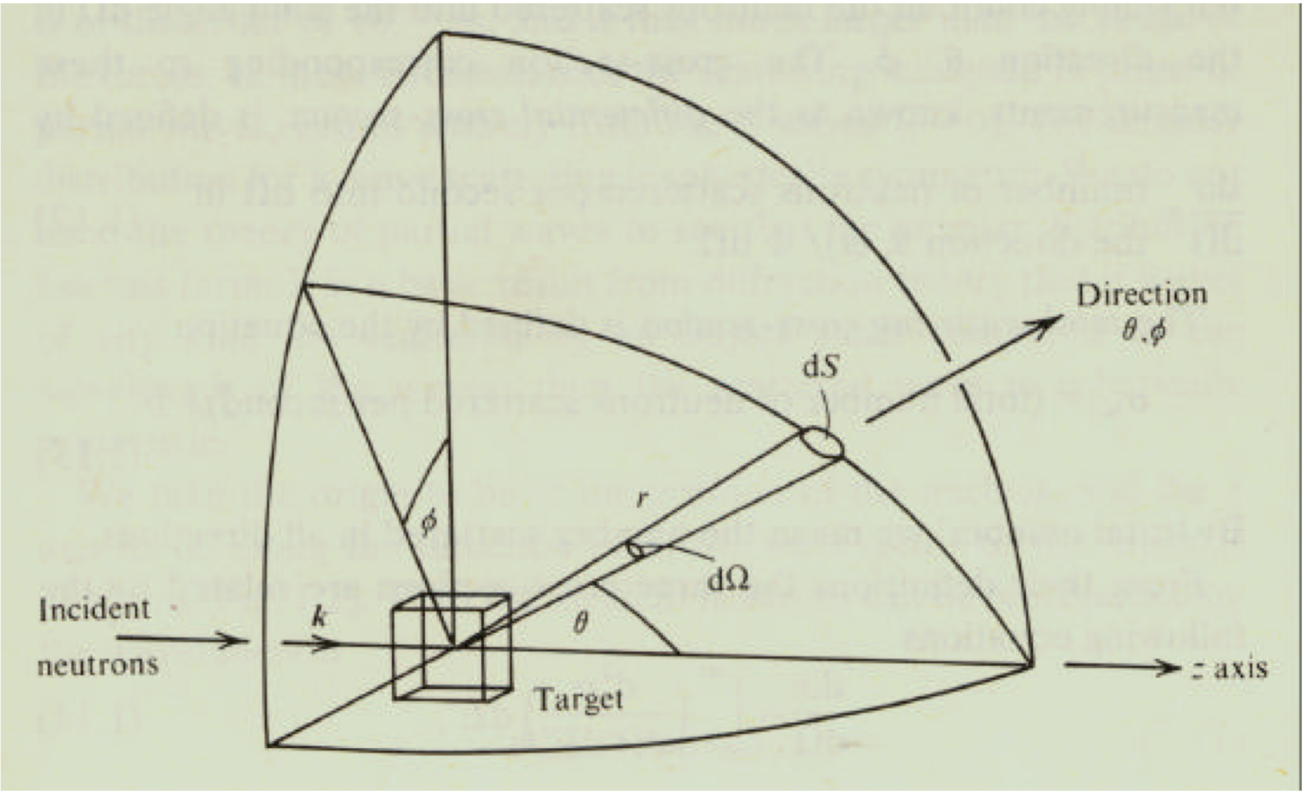


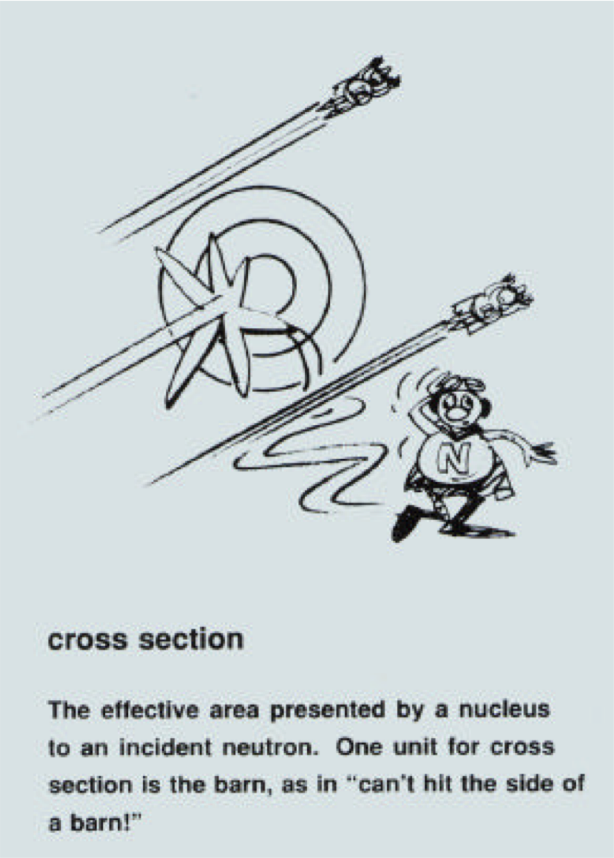
FRM II: Spectrum of the cold source and hot source



Maxwell-flux φ(υ) in a moderator at T=25K, T=300K und T=2000K.

Scattering theory: Cross section





cross section

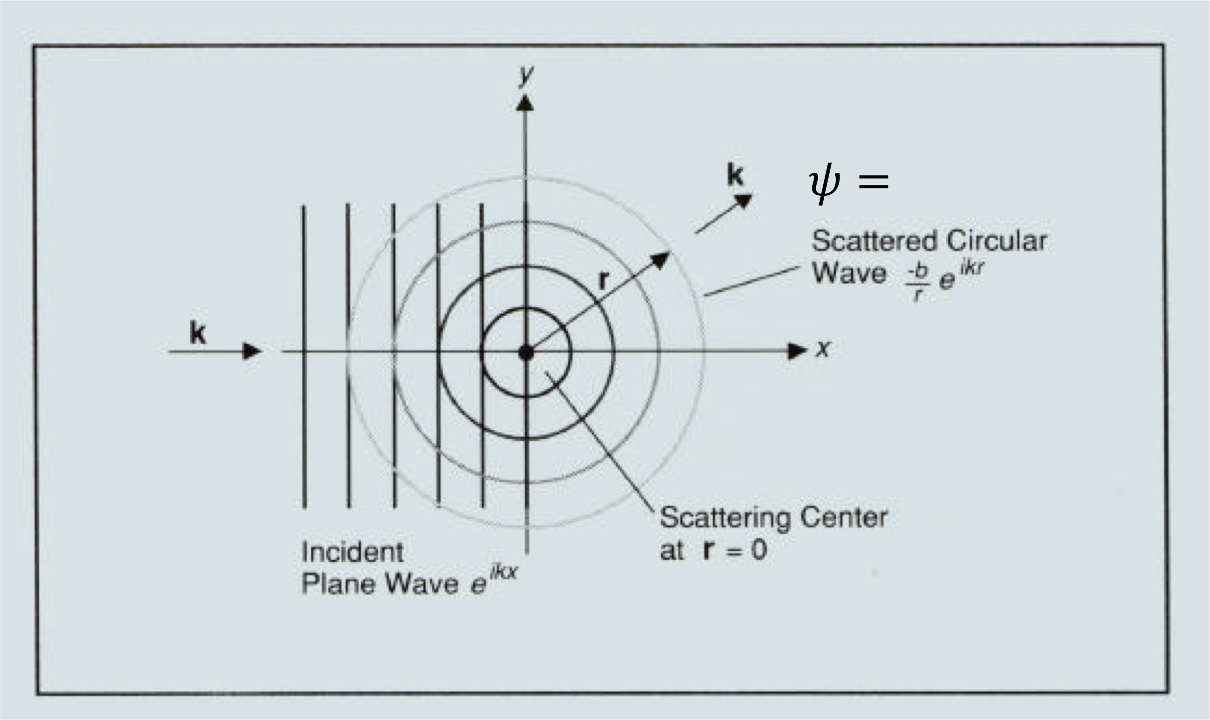
The effective area presented by a nucleus to an incident neutron. One unit for cross section is the barn, as in "can't hit the side of a barn!"

- Flux Φ = number of incident n/(s cm^2)
- Cross section σ = number of scattered n/s / Φ
- $d\sigma/d\Omega$ = number of scattered n/s / $\Phi d\Omega$

Attenuation = $e^{-N\sigma d}$

σ in barn:1 barn = 10^{-24} cm^2
N = Atoms/unit cell
d = thickness

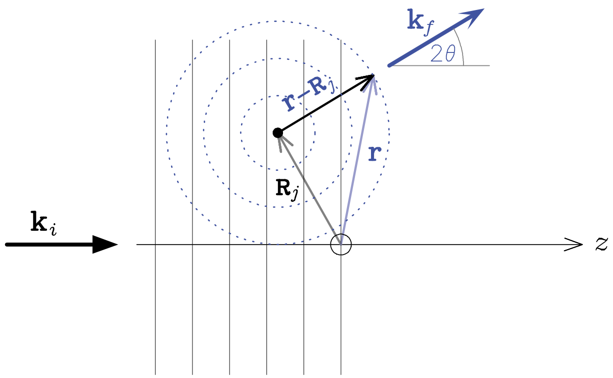
Scattering on a single nucleus



- Strong interaction short range (~1fm) << neutron wavelength
  $\Longrightarrow$ scattering is “point like”
- no absorption
- elastic (no energy transfer, no time dependency)
- b is the scattering length in cm, typical 10^{-12} cm (can not be calculated, needs to be measured !)

- Differential cross section: $$\frac{d\sigma}{d\Omega} = \frac{\textit{number of scatter n/s}}{d\Omega\,\Phi} = \frac{|\psi|^2}{d\Omega\,\Phi} = b^2$$
- Total cross section: $$\sigma = 4\pi\, b^2$$

Coherent and incoherent scattering



- Superpostition of all neutron waves: sum over all atoms N:

$$\psi_{scatter} = \sum_{i,j=0\dots N} e^{-i\overleftarrow{k}_0\overleftarrow{R}_i}\left[\frac{-b_j}{|\overleftarrow{r}-\overleftarrow{R}_j|}\, e^{-i\overleftarrow{k_f}(\overleftarrow{r}-\overleftarrow{R}_j)}\right]$$

- For neutrons the the scattering length depends on the isotope and the nuclear spin: $b_i = \langle b\rangle + \delta b_i$
- This gives in the end

$$\boxed{\frac{d\sigma}{d\Omega} = \langle b\rangle^2 \sum_{i,j} e^{-i\overleftarrow{Q}\,(\overleftarrow{R}_i-\overleftarrow{R}_j)} + (\langle b^2\rangle - \langle b\rangle^2)N}$$

coherent scattering

(Q dependent)

incoherent scattering

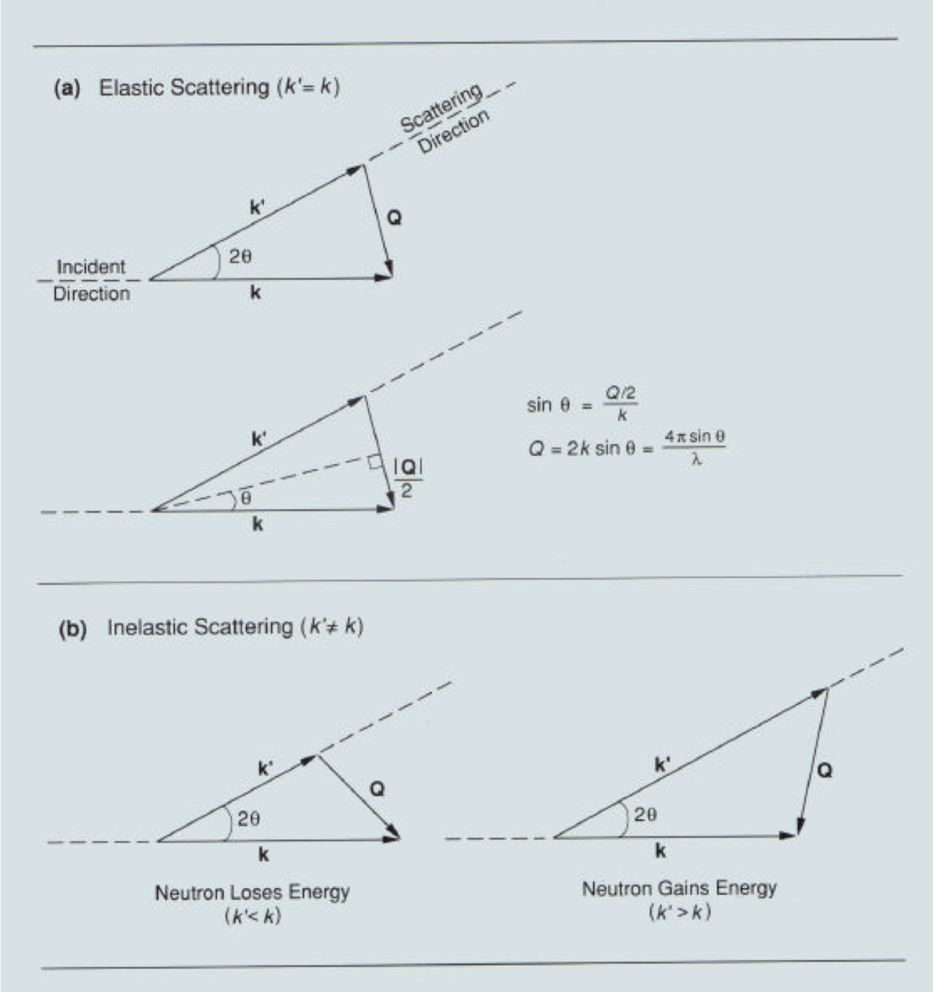
(in all directions)

Selected values for σ_{coh} and σ_{inc}

Nuclide	σ_{coh}	σ_{inc}	Nuclide	σ_{coh}	σ_{inc}
^{1}H	1.8	80.2	V	0.02	5.0
^{2}H	5.6	2.0	Fe	11.5	0.4
C	5.6	0.0	Co	1.0	5.2
O	4.2	0.0	Cu	7.5	0.5
Al	1.5	0.0	^{36}Ar	24.9	0.0

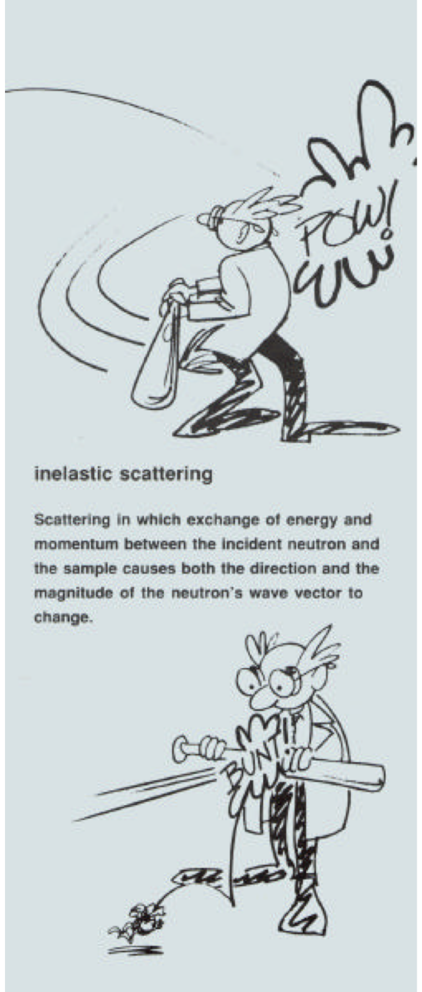
- Large difference for H/D which is used for contrast variation
- Al is used for sample environment and beam windows
- V is used as a standard scatter for inelastic scattering

Elastic versus inelastic scattering



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

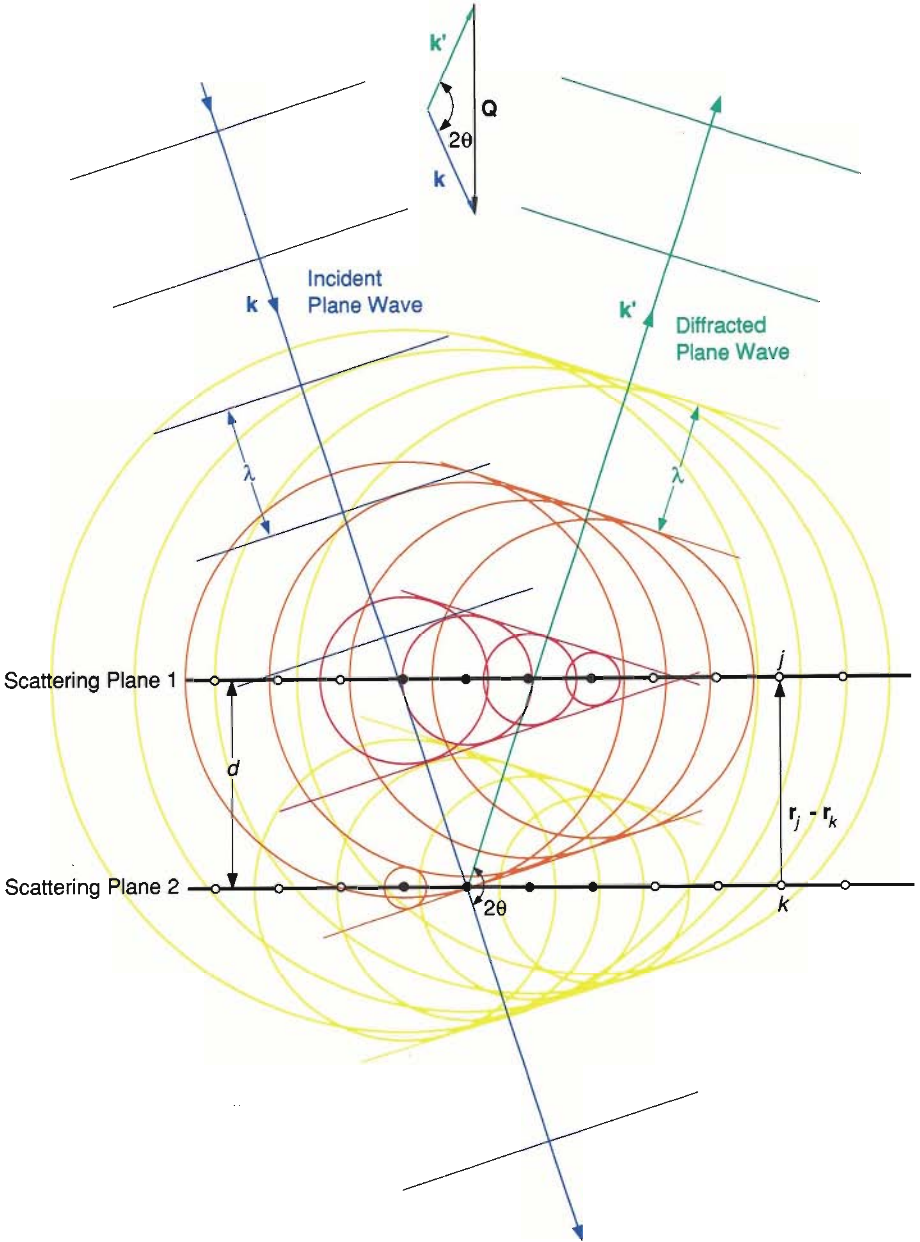
$$Q = 2k\sin\theta = \frac{4\pi\sin\theta}{\lambda}$$



inelastic scattering

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

Coherent elastic scattering at crystals



Condition for constructive interference:

- $\overleftarrow{Q}$ must be perpendicular to the diffracted wave front
- $\overleftarrow{Q} \cdot \left(\overleftarrow{r}_j - \overleftarrow{r}_k\right) = Qd = 2\pi n$, where d is the lattice spacing, and n is an integer number
- $Q = \frac{4\pi}{\lambda} \sin\theta$ is the condition for elastic scattering

$\Longrightarrow$

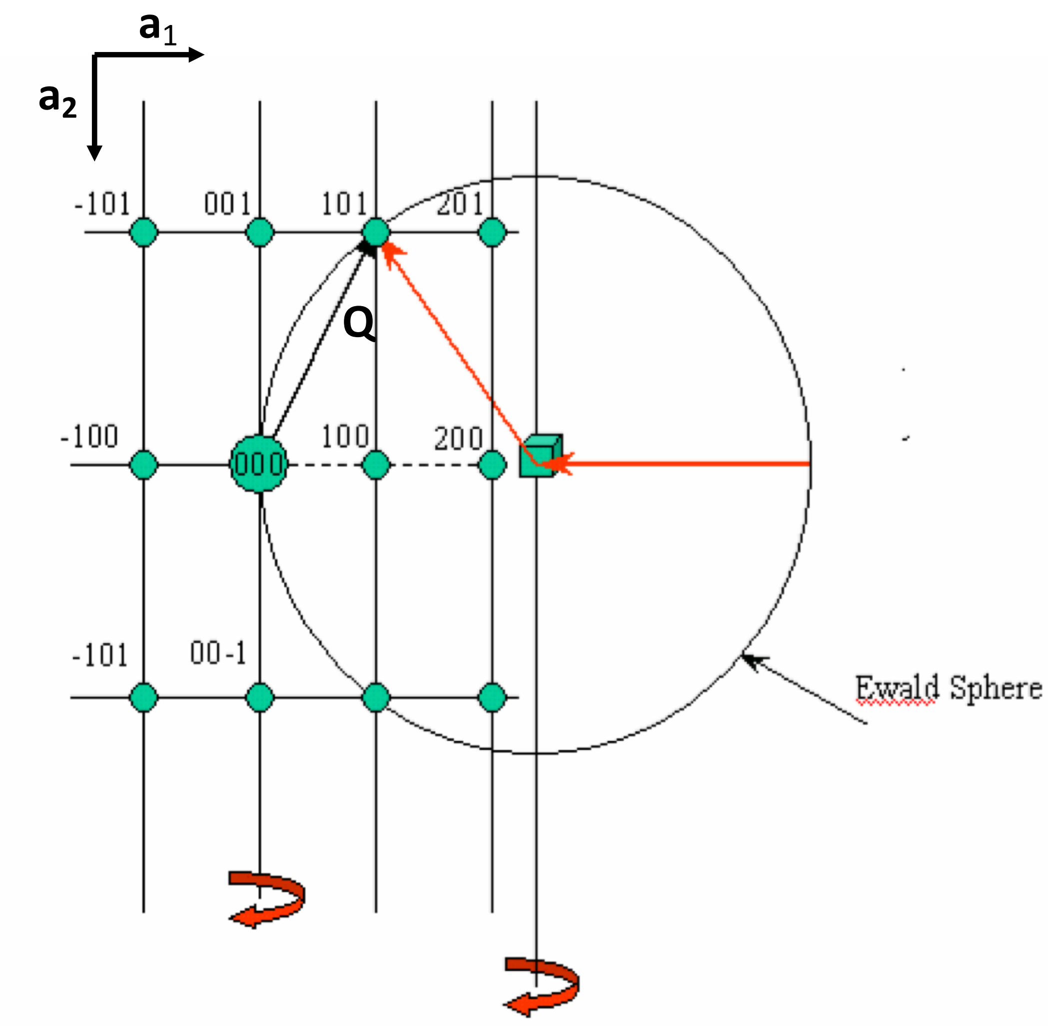
$n\lambda = 2d\sin\theta$ Bragg's law

Key Points about Diffraction

- A monochromatic (single λ) neutron beam is diffracted by a single crystal only if specific geometrical conditions are fulfilled

- These conditions can be expressed in several ways:
  - Laue's conditions: $\mathbf{Q}\,\mathbf{a}_1 = h$; $\mathbf{Q}\,\mathbf{a}_2 = k$; $\mathbf{Q}\,\mathbf{a}_3 = l$
    h, k, and l as integers; a_i the translations of the unit cell
  - Bragg'sLaw: $2d_{hkl}\sin\theta=\lambda$
  - Ewald's construction

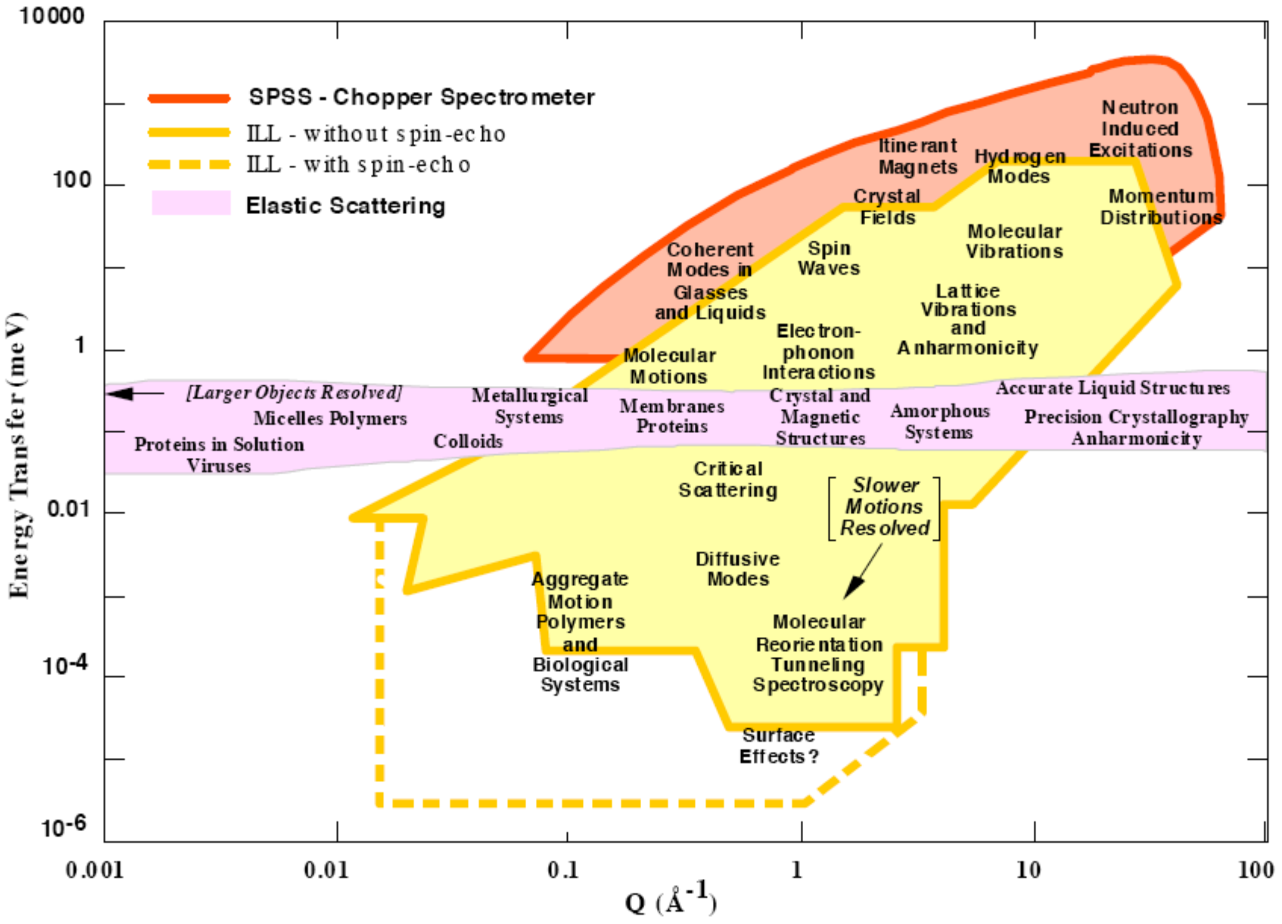
- Diffraction tells us about:
  - The dimensions of the unit cell
  - The symmetry of the crystal
  - The positions of atoms within the unit cell
  - The extent of thermal vibrations of atoms



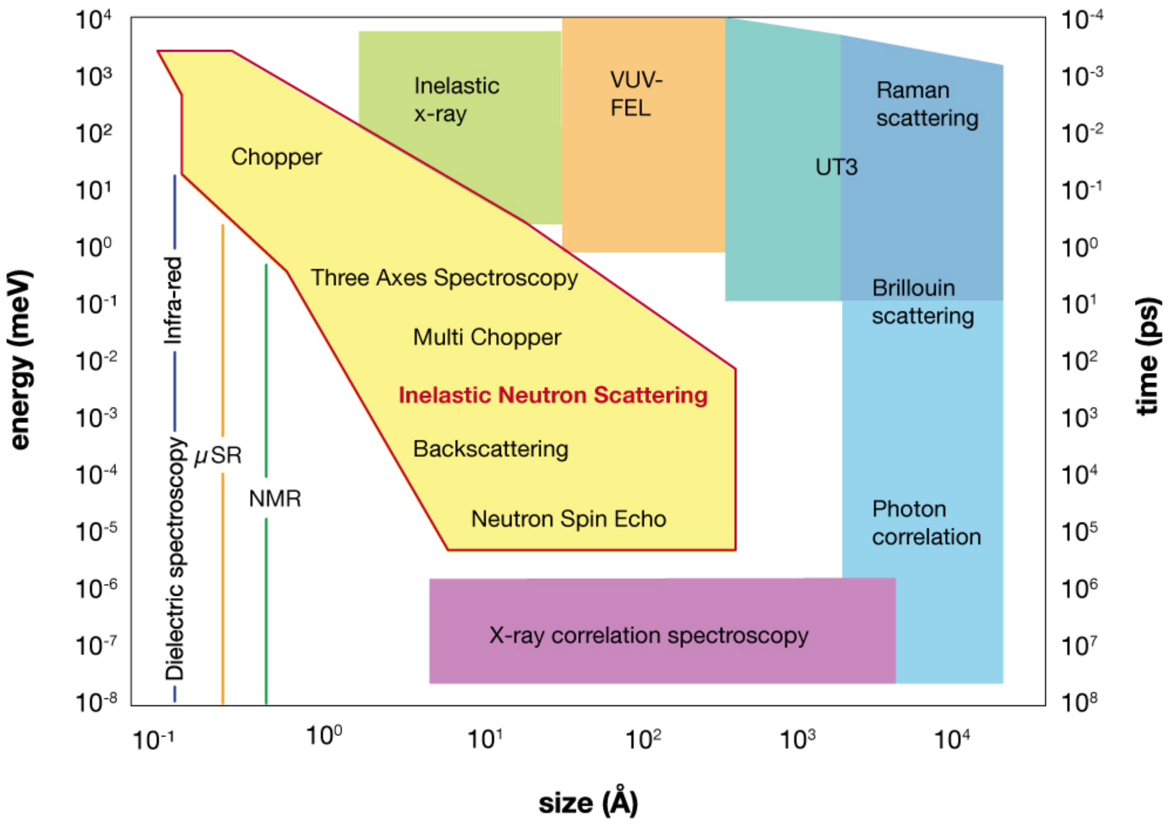
Take home message

- **Coherent, elastic scattering** shows where atoms are (**Bragg’s law**)
- **Incoherent, elastic** scattering contributes to the **background** independent of angle
- **Coherent, inelastic** scattering describes the **collective movement of atoms**
- Incoherent, inelastic scattering describes diffusion (the self-correlation function of atoms)

Physics explored with neutrons



ILL yellow book



MLZ blue book

Instruments

- Elastic scattering (Diffractometer)
  - Diffractometer (Powder and single crystal)
  - Small angle scattering (SANS)
  - Reflectometer

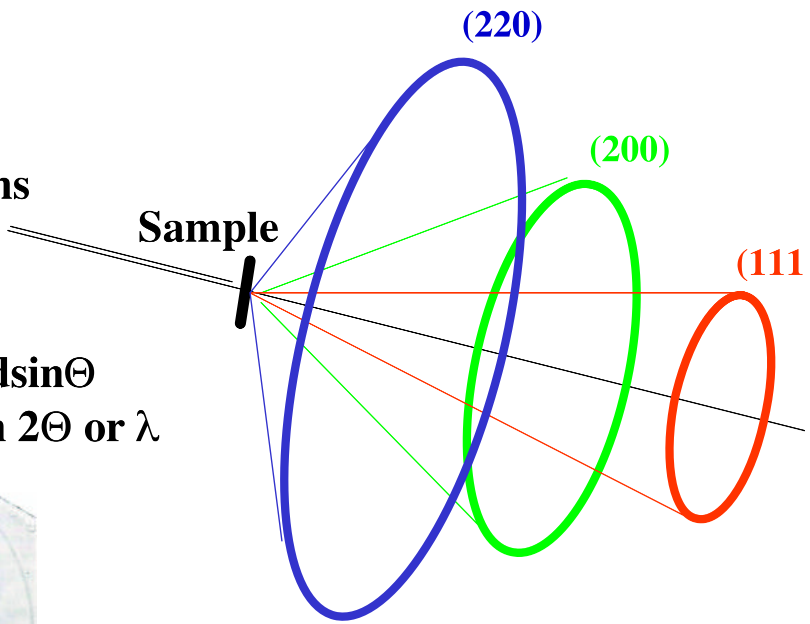
- Inelastic scattering (spectrometer, energy transfer meV region)
  - Three axis spectrometer
  - TOF spectrometer

- Quasielastic scattering (Energy transfer in the mev region)
  - Backscattering spectrometer
  - Spin-echo spectrometer

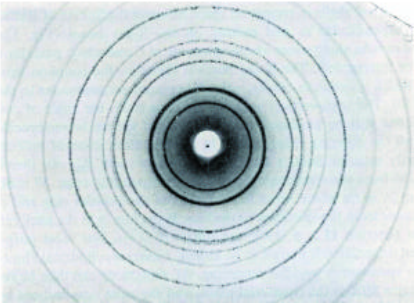
- Imaging instruments (direct observation), nuclear and fundamental physics, Positron source, medical applications and irradiation facility

Powder Diffraction

Incident beam
x-rays or neutrons

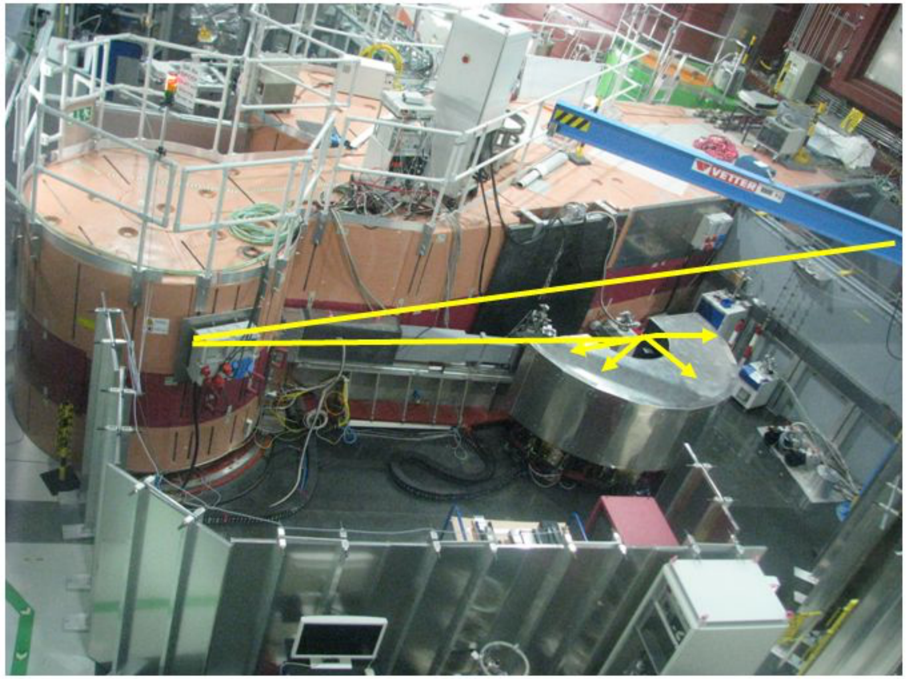


Bragg's Law $\lambda = 2d\sin\Theta$
Powder pattern – scan 2Θ or λ



A typical powder diffraction pattern

Pulverdiffraktometer SPODI am FRM II

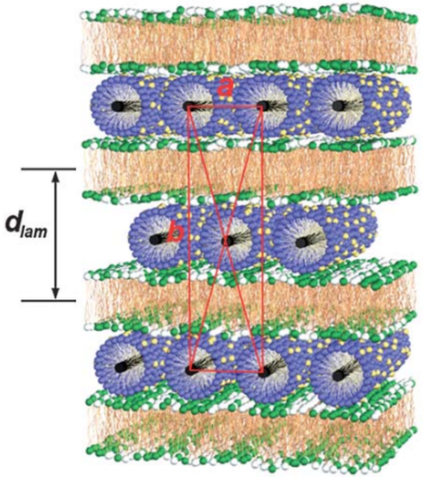


SANS - Resumé

SANS: Diffractometer specialized for small scattering angles

Large correlations in real space
20 to 40000 Å

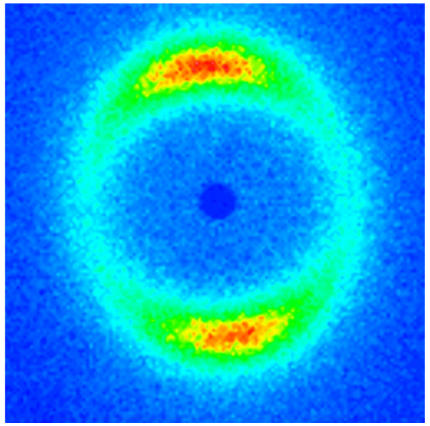
Low **Q** small scattering angles
~1 Å^{-1} to ~10^{-4} Å^{-1}



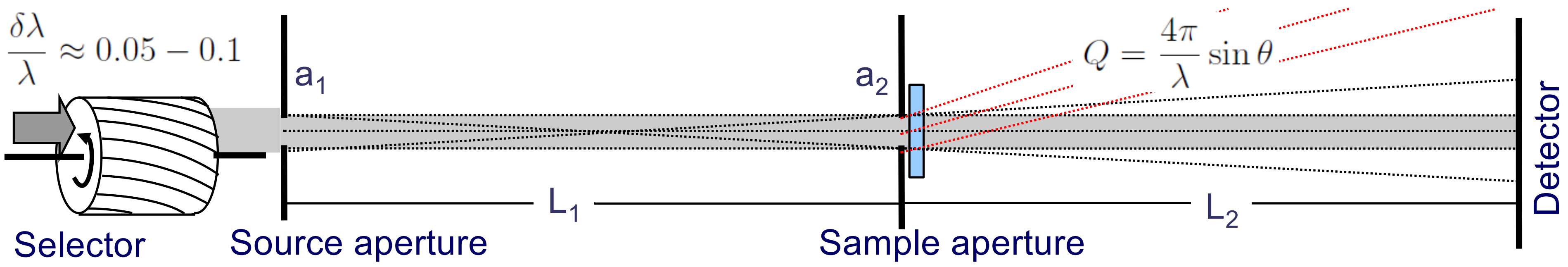
SANS tells you the statistical average of:

- **Shape** of scattering object
- **Size** (distribution) of scattering objects
- Surface of scattering objects
- Scattering length density (distribution)
- Arrangement (Superstructure?)

SANS 1 at FRM II



SANS – Resolution



- Angular resolution
- Monochromaicity
- Detector resolution
- Gravity

Treat as Gaussian distributions: $\left\langle \frac{\delta Q^2}{Q^2} \right\rangle = \left\langle \frac{\delta\lambda^2}{\lambda^2} \right\rangle + \left\langle \frac{\cos^2\theta\,\delta\theta^2}{\sin^2\theta} \right\rangle$

$\left\langle \frac{\delta Q^2}{Q^2} \right\rangle = 0.0025 + \left\langle \frac{\delta\theta^2}{\theta^2} \right\rangle$ ⇒ Angular resolution: $\delta\theta \approx \sqrt{\frac{5}{12}\frac{a}{L}}$

What is the largest object SANS can detect (limit small Q)?

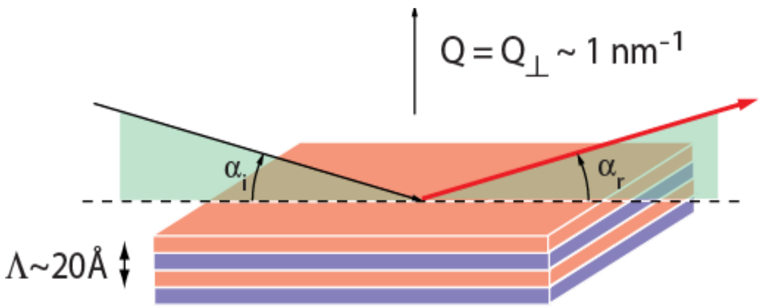
For $a_1 = a_2 = a$ and $L_1 = L_2 = L$

⇒ $\delta Q \approx \frac{\delta\theta}{\theta_{min}} Q_{min} \approx \delta\theta \frac{4\pi}{\lambda} \approx \frac{2\pi a}{\lambda L}$

Largest object: $\frac{2\pi}{\delta Q} = \frac{\lambda L}{a}$ On D11, ILL: L=40m, λ=15Å ⇒ D≈ 5μm

For large scattering angles (large Q) wavelength resolution dominates.

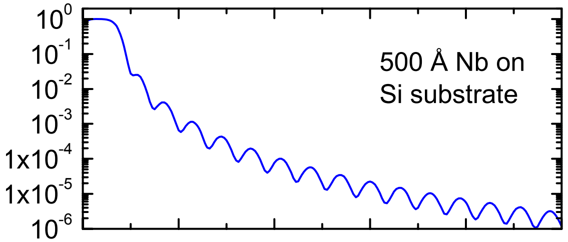
Reflectometer

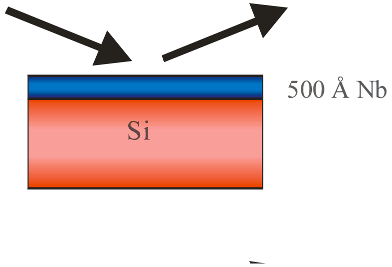


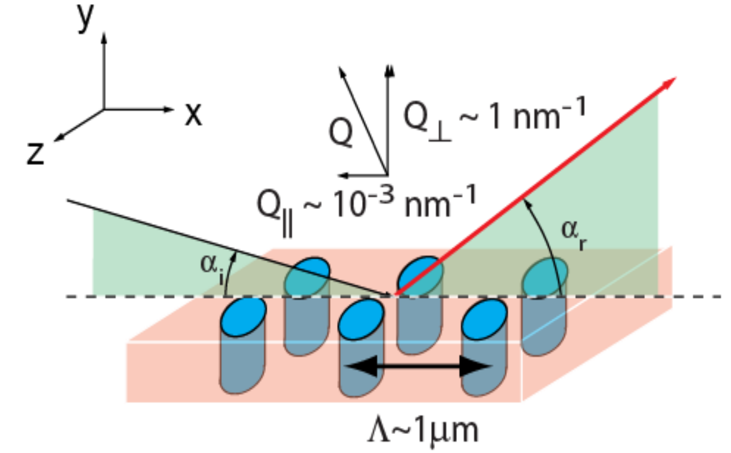
Specular reflectometry

Depth profiles

(nuclear and/or magnetic)





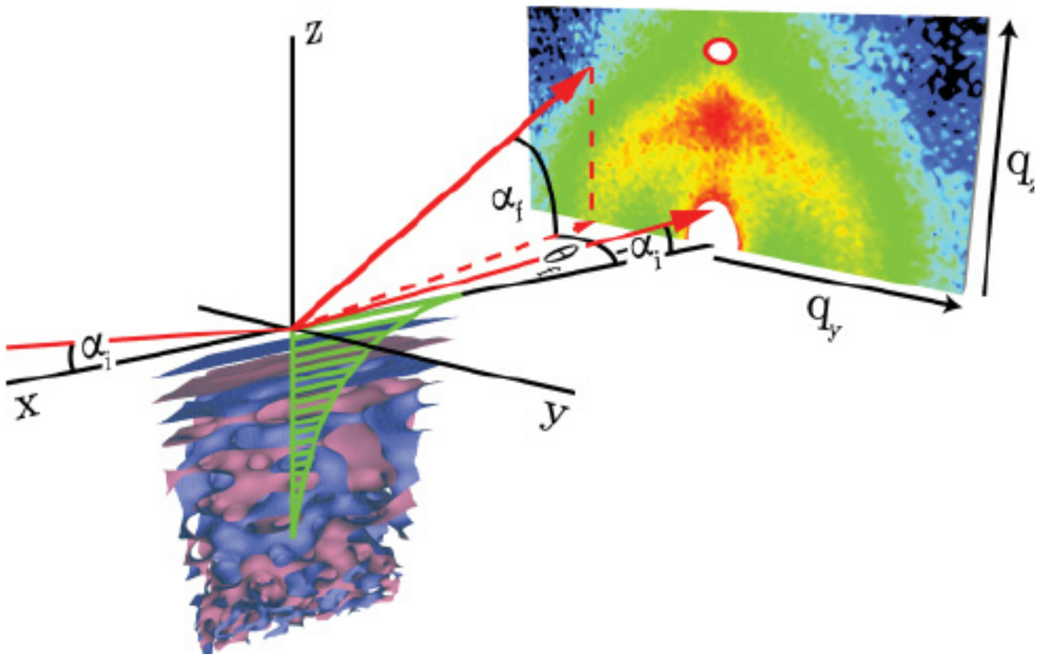


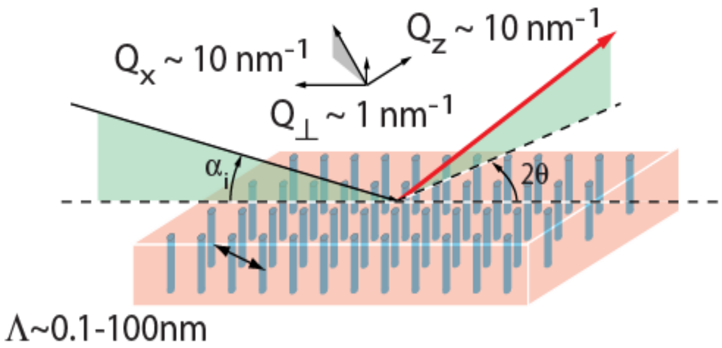
Off-specular (diffuse) scattering

In-plane correlated roughness

Magnetic stripes

Phase separation (polymers)





Glancing incidence diffraction

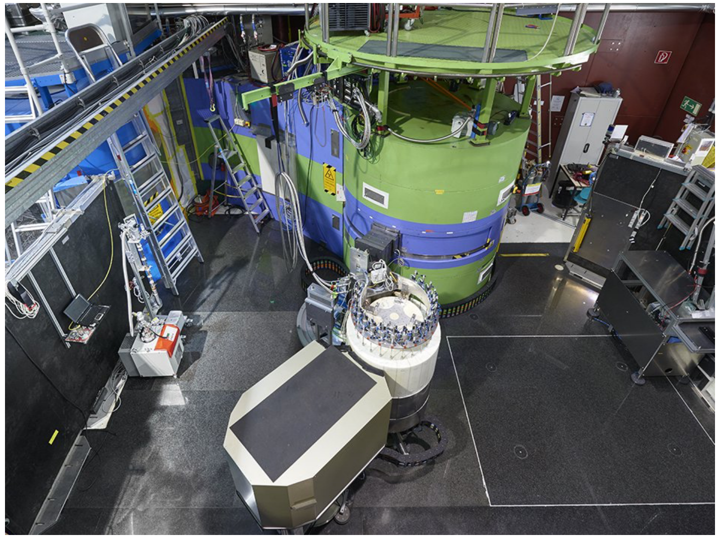
Ordering in liquid crystals

Atomic structures near surfaces

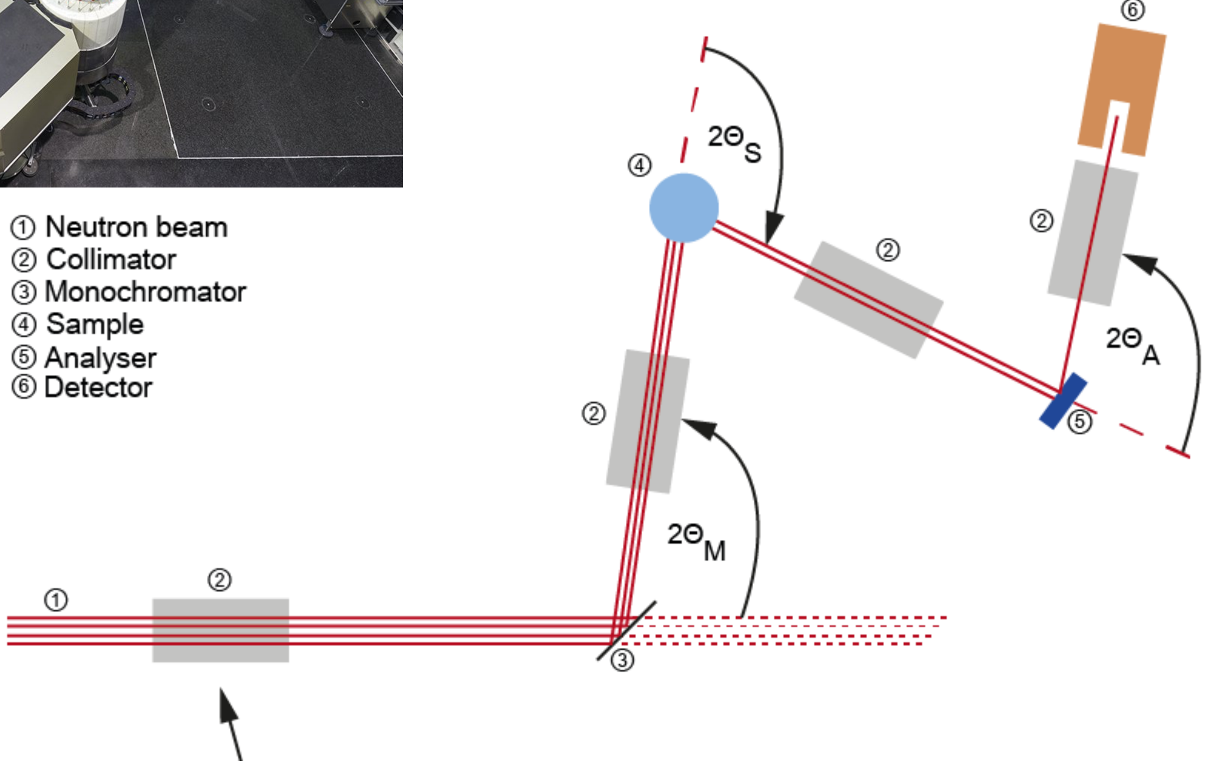
Interactions among nanodots

Viewgraph from M. R. Fitzsimmons

Three axis spectrometer (TAS)



PUMA@FRMII

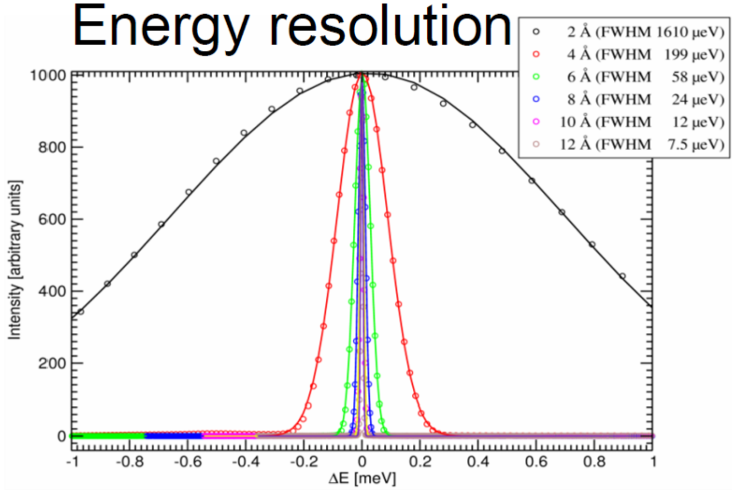


- cold source: $\lambda \approx 5$ Å
- monochromator: selects initial energy E_i
- sample environment: cryostat and magnets
- analyzer: selects final energy E_f
- scattering angle: selects momentum transfer $\mathbf{Q} = \mathbf{G} + \mathbf{q}$
- maps out the dispersion relation $\hbar\omega(\mathbf{q}) = E_i - E_f$

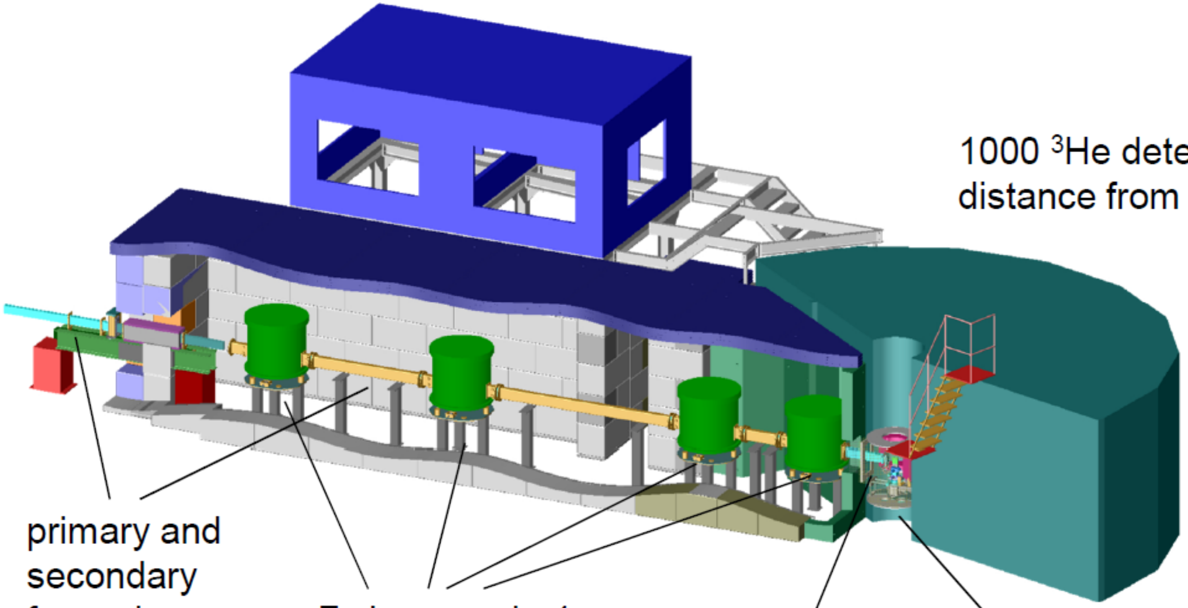
- here: keep E_i and $\mathbf{q}$ fix and scan $\hbar\omega$ for several $\mathbf{q}$'s and temperatures

Time of flight spectrometer

Energy resolution



Instrument Layout



1000 ^{3}He detector tubes at 4 m distance from the sample

primary and secondary focussing neutron guide

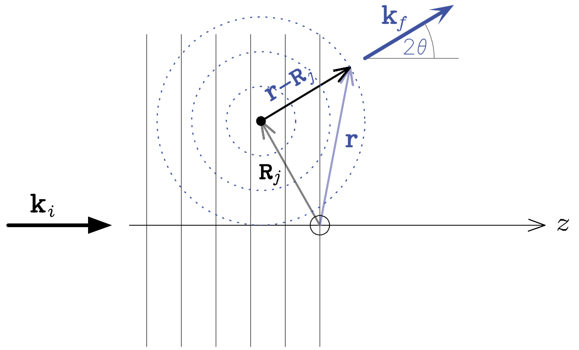
7 choppers in 4 vacuum vessels

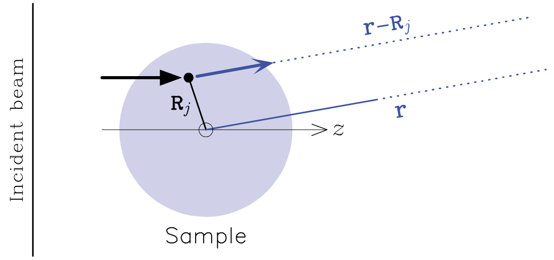
exchange guide:
extended 23 x 44 mm^2
focused, adaptive: 10 x 10 mm^2

sample chamber (argon filled)

T. Unruh, J. Neuhaus, W. Petry
The high-resolution time-of-flight spectrometer TOFTOF, Nucl. Instr. Methods A **580** (2007) 1414

Several atoms: Superposition of scattering waves





- sum over all atoms N: $\psi_{scatter} = \sum_{i,j=0\dots N} e^{-i\overleftarrow{k}_0\overleftarrow{R}_i}\left[\frac{-b_j}{|\overleftarrow{r}-\overleftarrow{R}_j|}\, e^{-i\overleftarrow{k_f}(\overleftarrow{r}-\overleftarrow{R}_j)}\right]$

- simplify using: $\overleftarrow{Q} = \overleftarrow{k}_f - \overleftarrow{k}_0$, the scattering vector and
- r >> R_j

- gives: $\frac{d\sigma}{d\Omega} = \sum_{i,j} b_i\, b_j e^{-i\overleftarrow{Q}\,(\overleftarrow{R}_i - \overleftarrow{R}_j)}$

- using