Introduction

to Neutron Scattering

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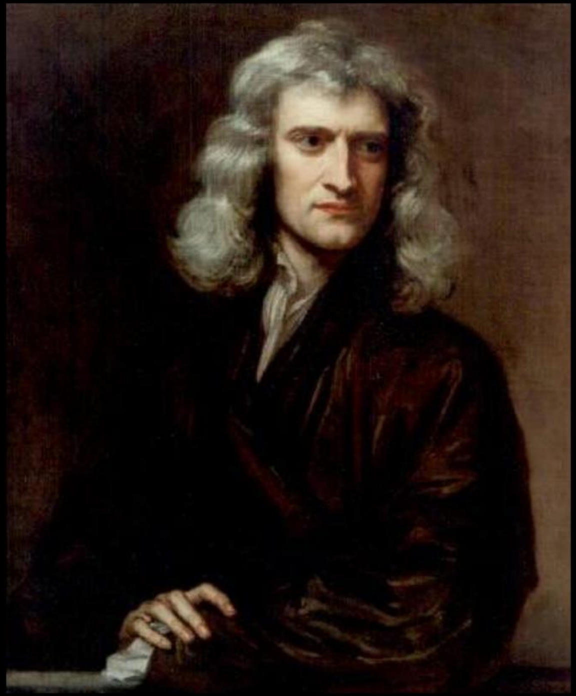
The task:

Isaac Newton

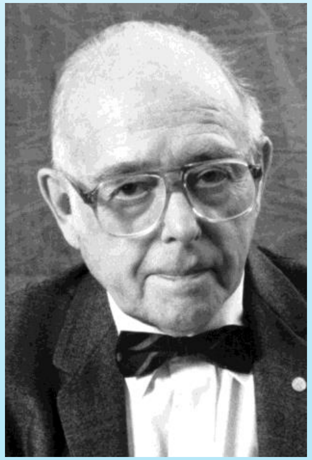


Now the smallest Particles of Matter may cohere by the strongest Attractions, and compose bigger Particles of weaker Virture... There are therefore Agents in Nature able to make the Particles of Bodies stick together by very strong Attraction. And it is the Business of experimental Philosophy to find them out.

Nobel Prize winners

James Chadwick — 1935 —

for the discovery of the neutron



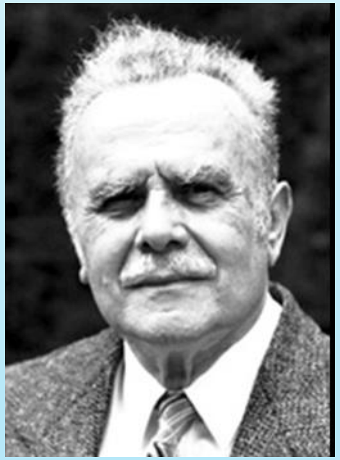
Clifford Shull — 1994 —

for the development of the neutron diffraction technique

has helped answer the question of **where the atoms „are”.**

Bertram N. Brockhouse — 1994 —

for the development of neutron spectroscopy

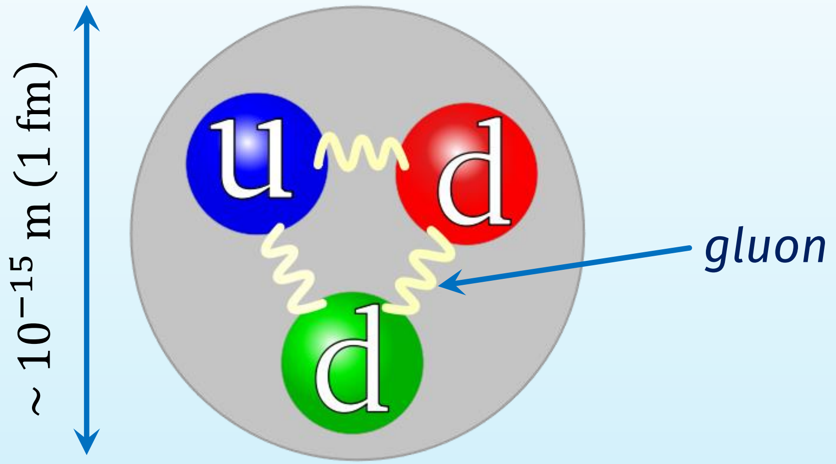


has helped answer the question of **what the atoms „do”.**

I. Introduction. The neutron.

The neutron:

- is a subatomic particle
- though not elementary:



Quantity	Symbol	Value
Rest mass	m_n	$1.674927211(84) \times 10^{-27}$ kg
		1.00866491597(43) at.u.
	m_nc^2	939.565346(23) MeV
Spin	I	½
Charge		0
Magnetic moment	μ_n	$-0.96623641 \cdot 10^{-27}$ J·T
		$-1.0418756272 \cdot 10^{-3}\ \mu_B$
Mean life time (free)	τ	879.6±0.9 s $877.75^{+0.50}_{-0.44}$ s (dependent on the method)

The neutron

- When bound within an atomic nucleus, the neutron is 'immortal'.
- If released free, however, it lives pretty short: 879.9±0.9 s (mean value, not to be confused with the *half-life*, which is by ln 2 less).

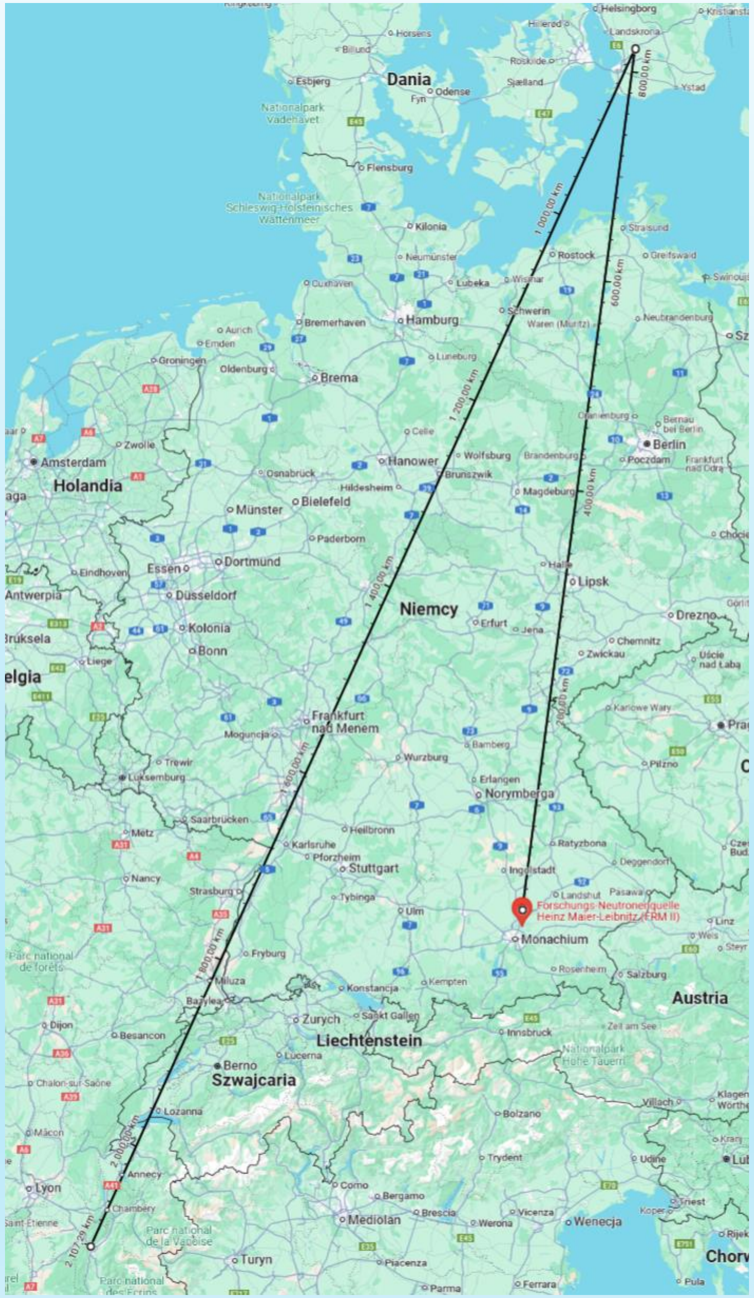
Example:

The neutron of parameters we would encounter
in a typical scattering experiment:

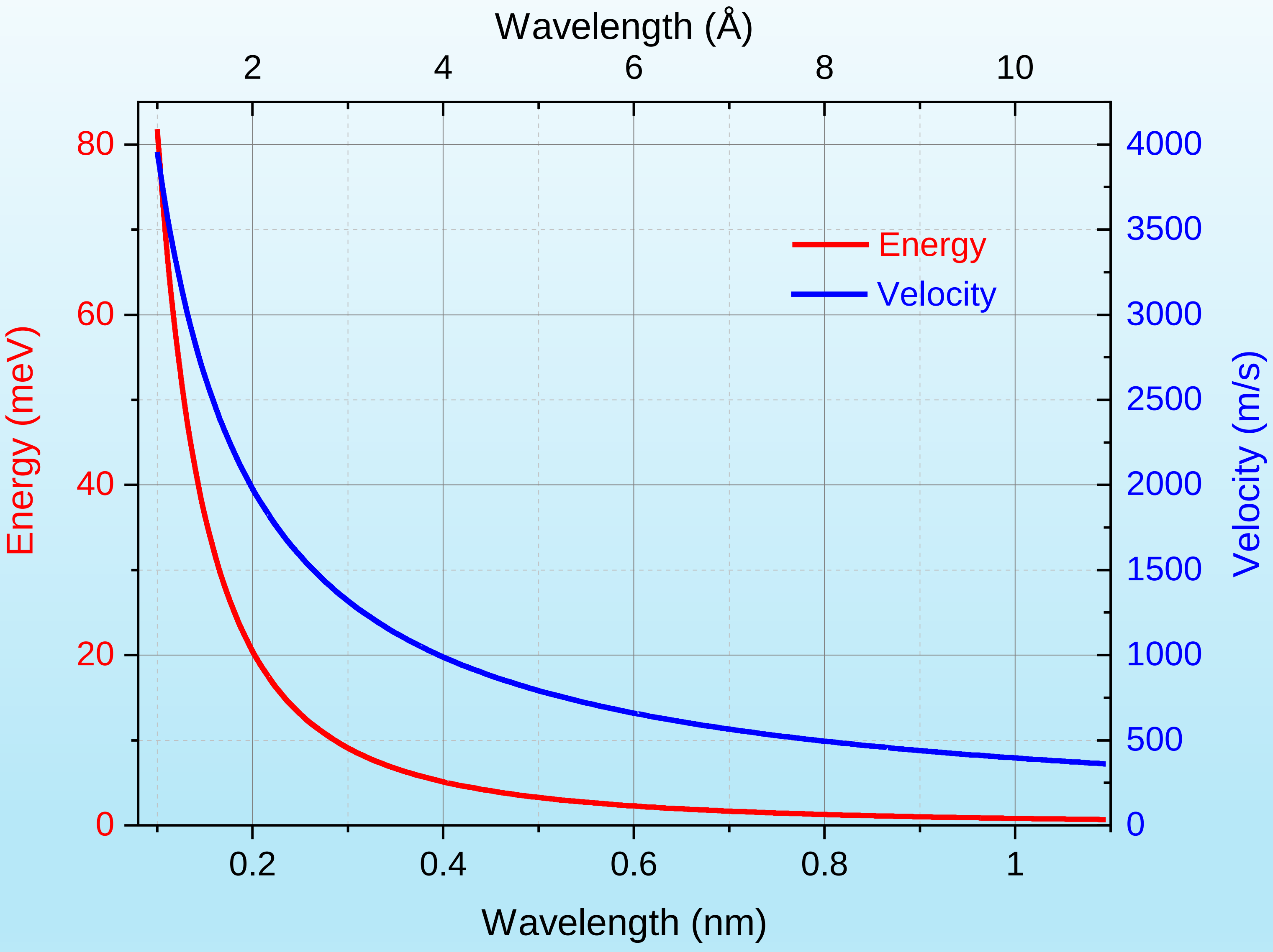
Quantity	Value
Energy	5.113 meV
Wavelength	0.4 nm (4 Å)
Wavevector	15.71 nm^{-1} (1.571 $Å^{-1}$)
Velocity	989.0 m×s^{-1}

Within its lifetime it will travel 870.32 km.

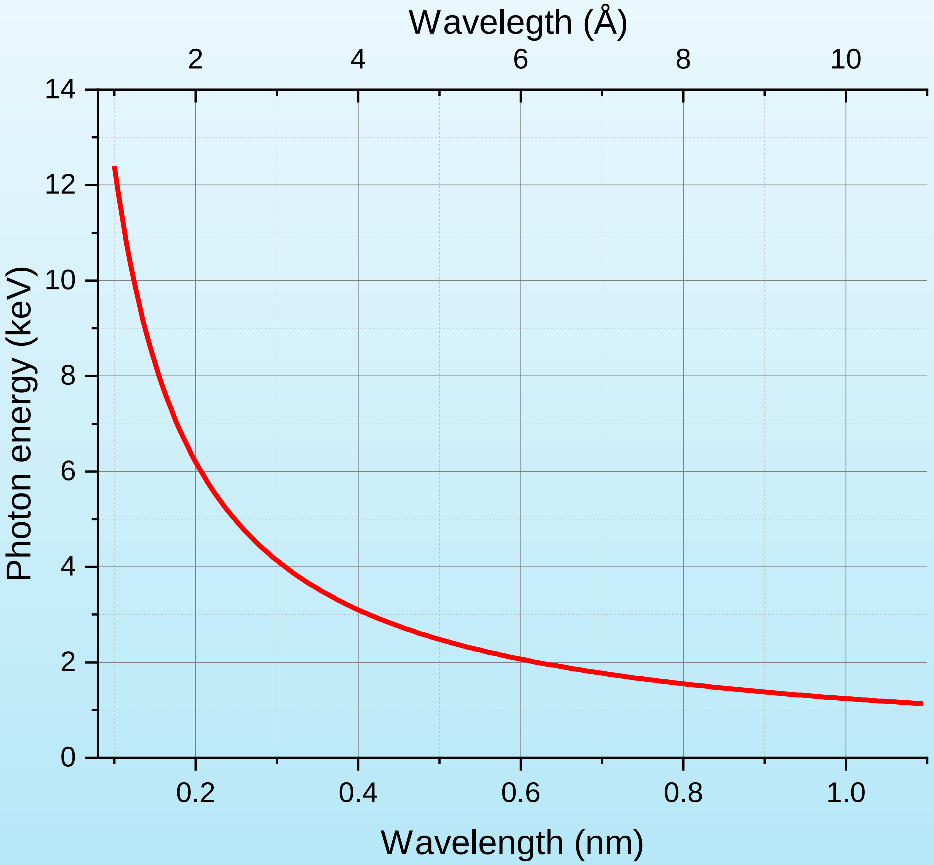
This means, it would make it from the ESS, Lund (822 km),
but not to the ILL, Grenoble (1286 km); «flat-earther's» convention.



The neutron



Neutron energy – wavelength – velocity

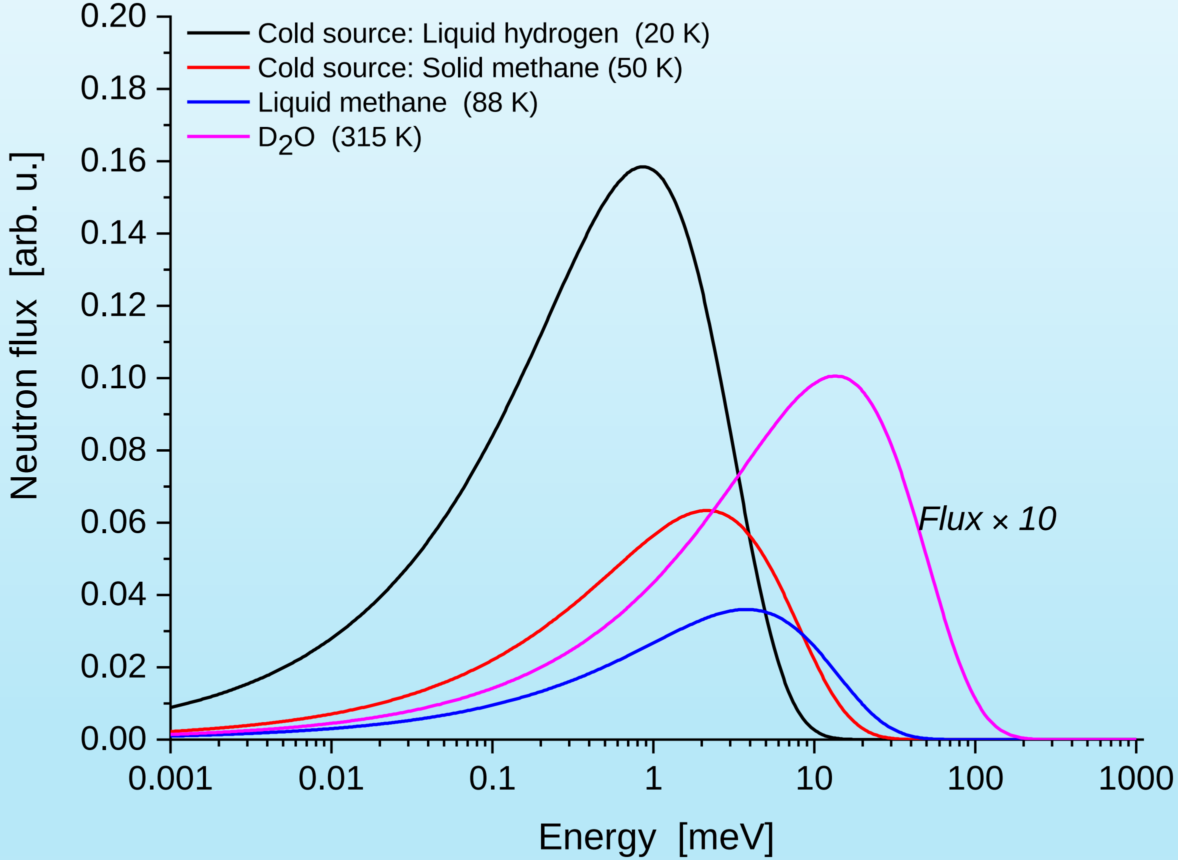


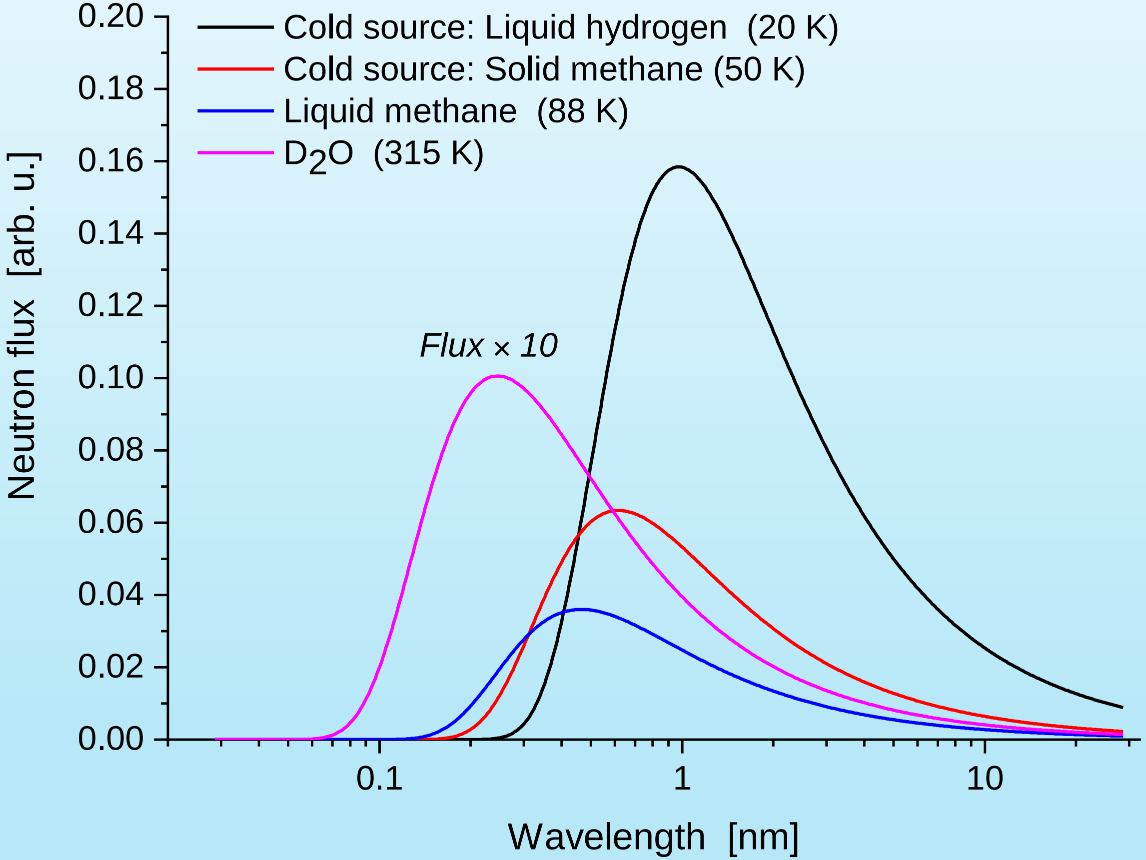
Photon energy vs. wavelength

– for comparison

Note 6 orders of magnitude difference.

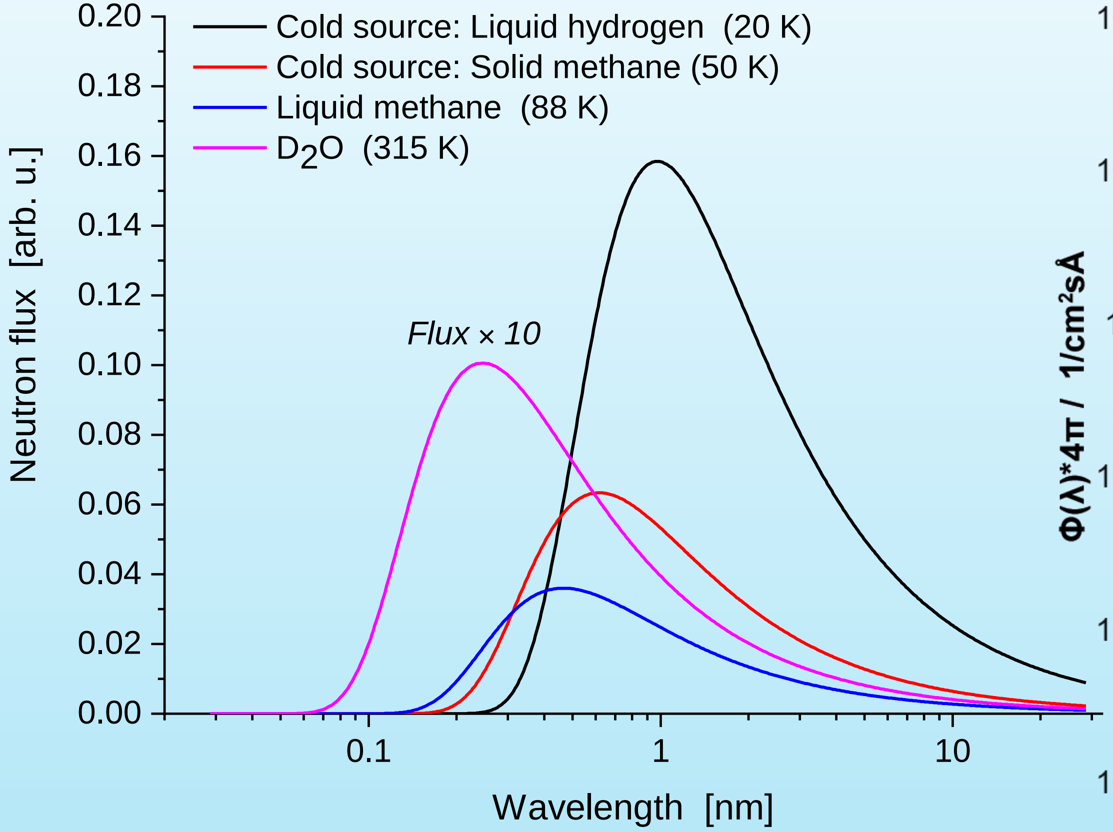
$$\Phi(E)dE = \Phi_0 \frac{2}{\pi} \frac{\sqrt{E}}{(k_\mathbf{B}T)^{3/2}} \exp\left(-\frac{E}{k_\mathbf{B}T}\right) dE$$

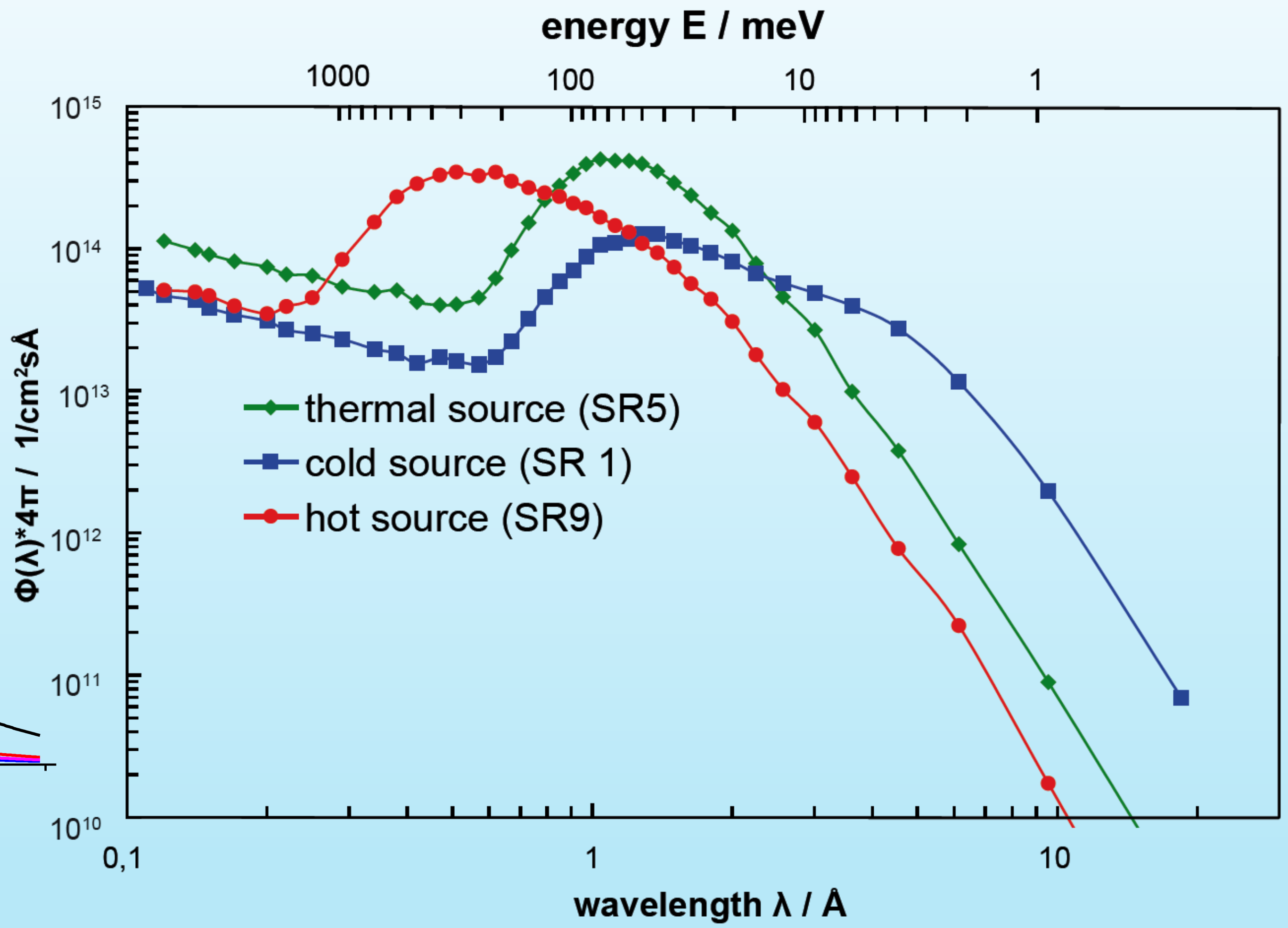




Neutron flux Maxwellian distribution

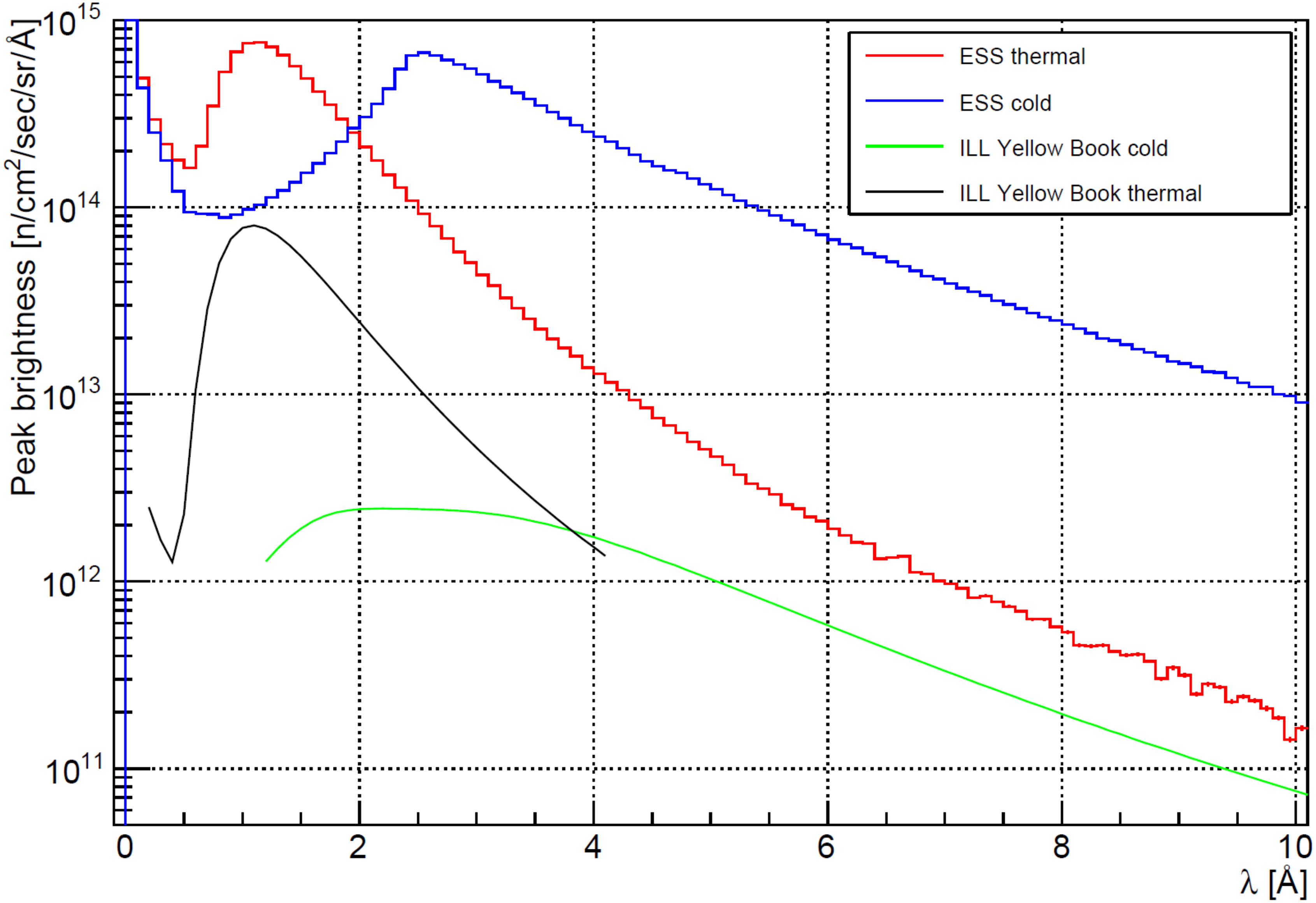
The neutron





Neutron flux Maxwellian distribution vs. Real neutron spectrum at the entrance to the beam tubes ar RFM II.

ESS vs. ILL moderated neutron spectra



ESS brightness spectra averaged over 42 beam ports for 3 cm high moderator, compared with ILL official curves.

L. Zanini, K. Batkov, E. Klinkby et al., IOP Conf. Series: Journal of Physics: Conf. Series 1021 (2018) 012066
doi :10.1088/1742-6596/1021/1/012066

A generally accepted classification with respect to neutron temperature, i.e. free neutron's kinetic energy (various sources may give slightly different classification borders).

Energy [meV]	**Wavelength [Å]**	**Wavelength [nm]**	**Energy classification**
0.05 ÷ 10	3 ÷ 40	0.3 ÷ 4	Cold
10 ÷ 100	0.9 ÷ 3	0.09 ÷ 0.3	Thermal
100 ÷ 500	0.4 ÷ 0.9	0.04 ÷ 0.09	Epithermal
500 ÷ 10^5	0.03 ÷ 0.4	$3 \cdot 10^{-3}$ ÷ 0.04	Hot
< 400	> 0.5	<0.05	Slow
> 1000	< 0.3	> 0.03	Fast
< 1000	> 0.3	< 0.3	low energy region
1000 ÷ 10^7	$3 \cdot 10^{-3}$ ÷ 0.03	$3 \cdot 10^{-4}$ ÷ $3 \cdot 10^{-3}$	resonance region
10^7 ÷ $2.5 \cdot 10^{10}$	$5 \cdot 10^{-5}$ ÷ $3 \cdot 10^{-3}$	$5 \cdot 10^{-6}$ ÷ $3 \cdot 10^{-4}$	continuum region

Useful relations

Particle properties Energy – momentum relations:	Wave properties Frequency – wavelength relations
$E=\dfrac{m_n v^2}{2};\quad E=\dfrac{p^2}{2m_n};\quad p=m_n v$	$E=\hbar\omega=\dfrac{\hbar^2k^2}{2m_n};\quad p=\hbar k;\quad k=\dfrac{2\pi}{\lambda}$

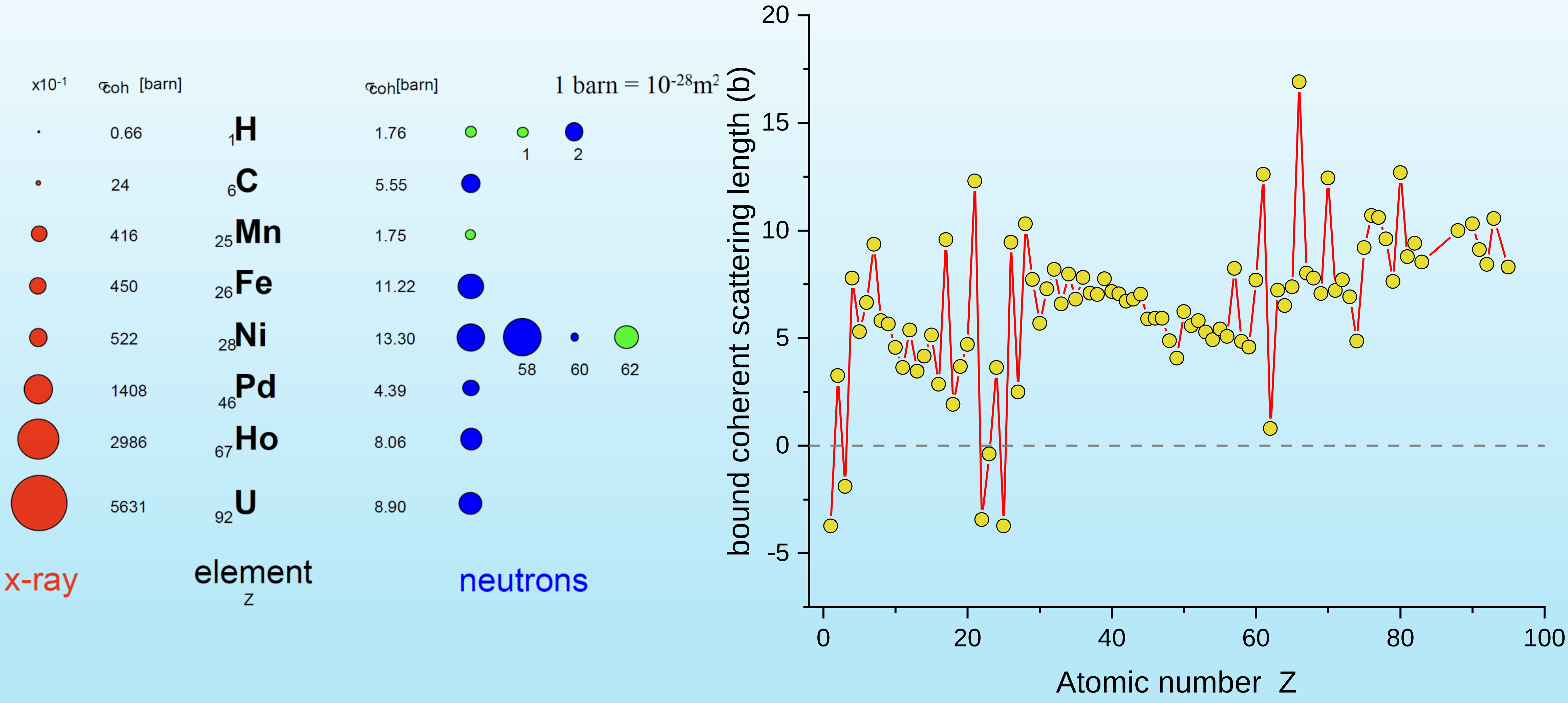
$$E\left[\mathrm{meV}\right] = 81.804\cdot\lambda^{-2}\left[\text{Å}^{-2}\right] = 0.81804\lambda^{-2}\left[\mathrm{nm}^{-2}\right]$$

$$E\left[\mathrm{meV}\right] = 0.0207212\cdot k^{2}\left[\mathrm{nm}^{-2}\right] = 2.07212\cdot k^{2}\left[\text{Å}^{-2}\right]$$

$$\lambda\left[\mathrm{nm}\right]=\frac{395.6}{v\left[\mathrm{m/s}\right]}\qquad \lambda\left[\text{Å}\right]=\frac{3956}{v\left[\mathrm{m/s}\right]}$$

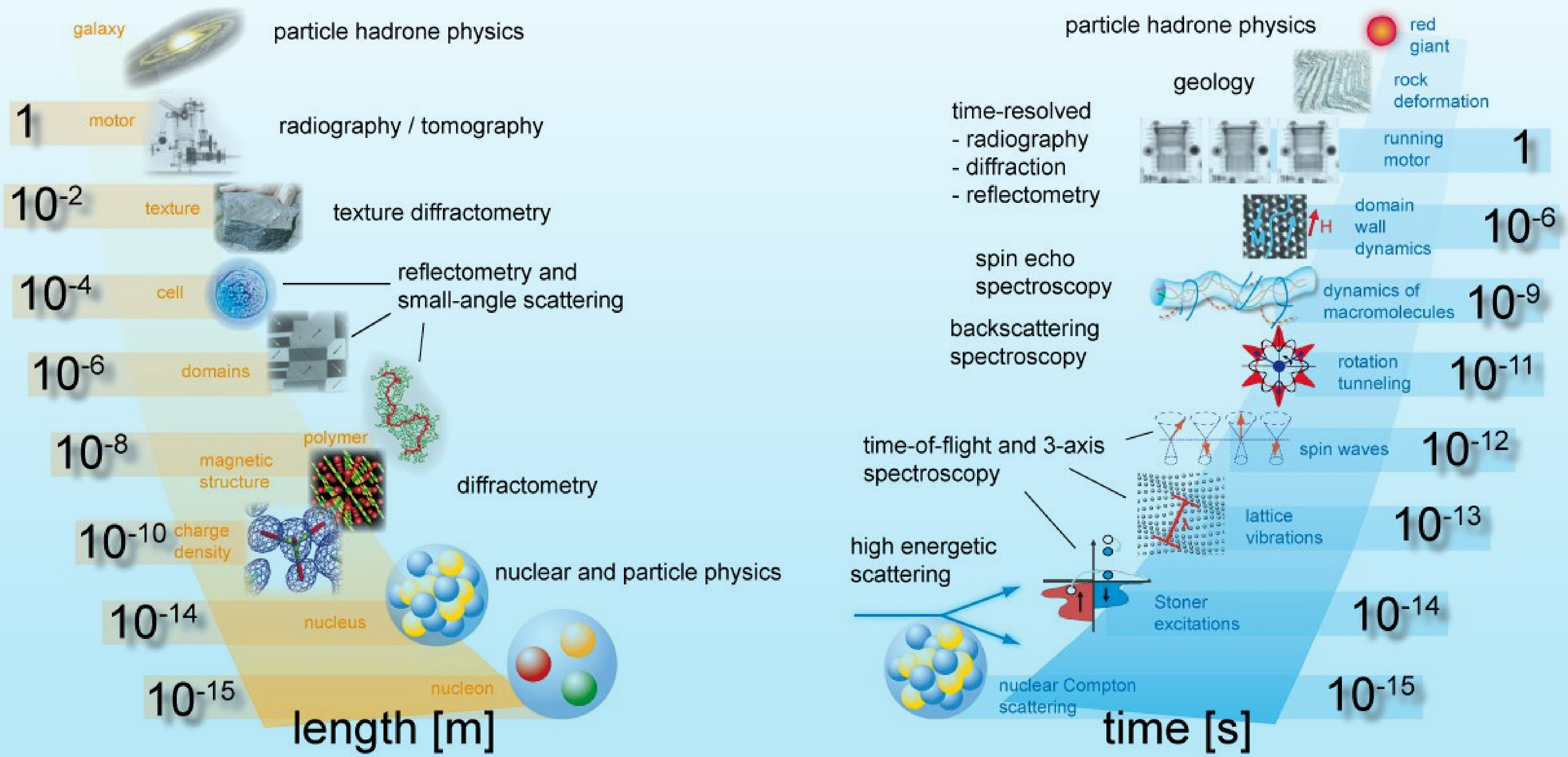
II. Why neutrons?

Why neutrons?



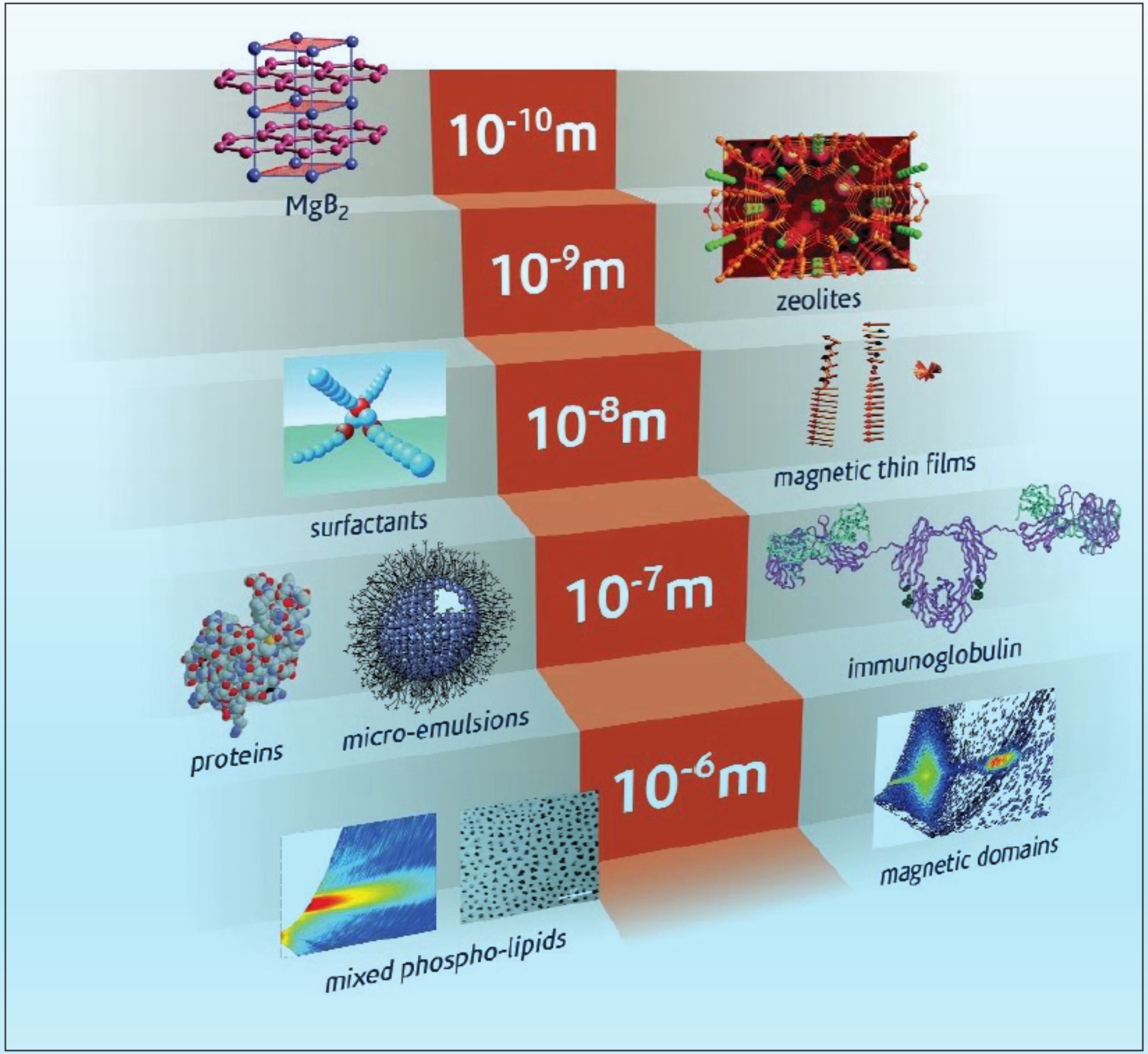
Comparison of x-ray and neutron scattering from single atoms. The filled circles represent a measure of the total cross section, i.e, of the probability for scattering.

Why neutrons?



Huge range of length (left side) and time (right side) scales covered by research with neutrons. Also indicated is the corresponding neutron technique.

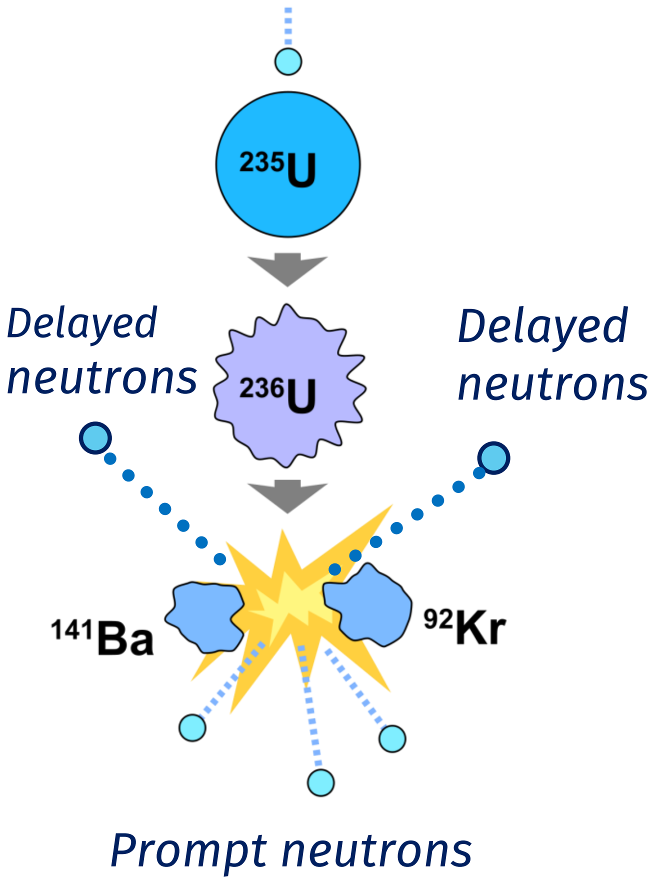
Why neutrons?

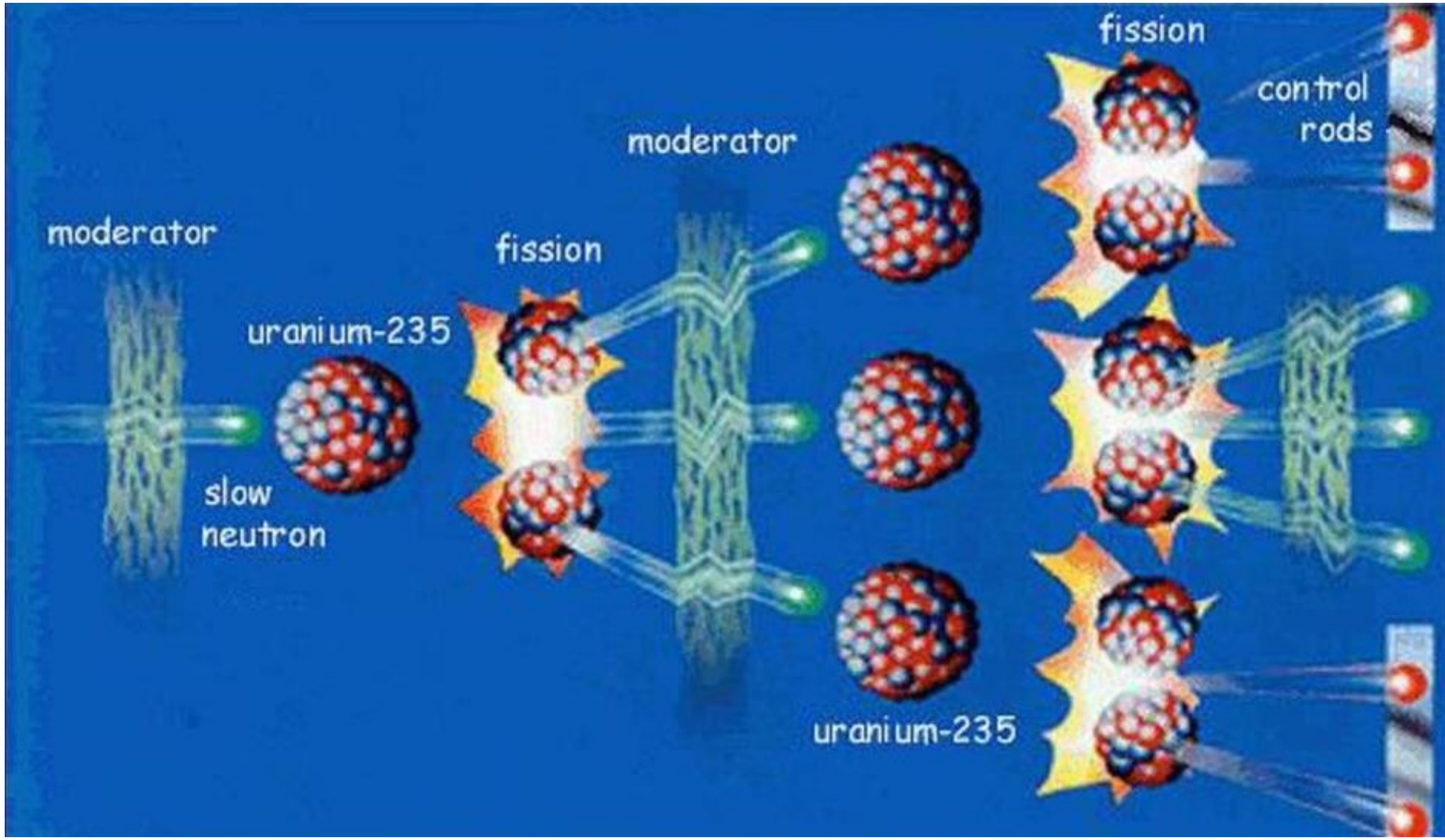


From Ångström to micrometer.

III. How neutrons are produced

(a) ^{235}U fission reaction

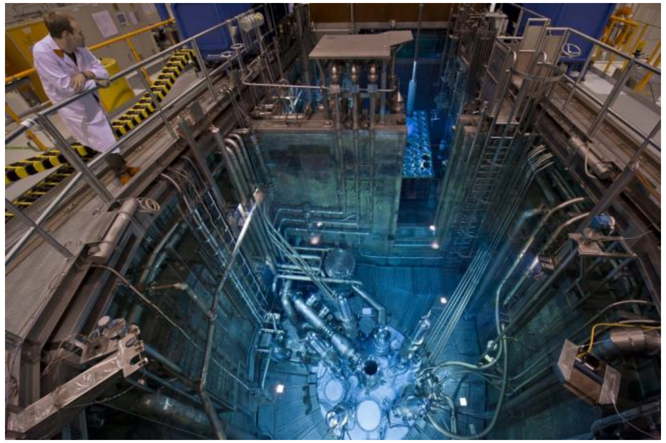




- *A prompt neutron is immediately emitted by a nuclear fission event.*
- *A delayed neutron appears within the same context, emitted after beta decay of one of the fission products anytime from a few milliseconds to a few minutes later.*

Some research reactors

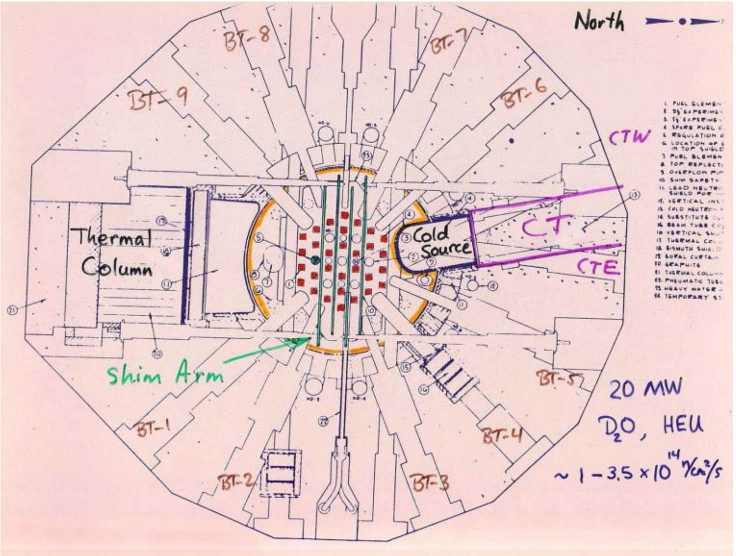
Facility	Thermal power [MW]	Neutron flux @ moderator [neutrons/cm^2/s]
HFR @ ILL, Grenoble	58.3	1.5×10^{15}
FRM II @ MLZ, Munich	20.0	8.0×10^{14}
HFIR @ ORNL (Oak Ridge)	85.0	2.6×10^{15}
NSBR, NIST, USA	20.0	3.5×10^{14}
BNC @ Budapest	10.0	1.0×10^{14}
EWA @ NCBJ, Warsaw	2.0	2.0×10^{13}



Research reactor vessels.

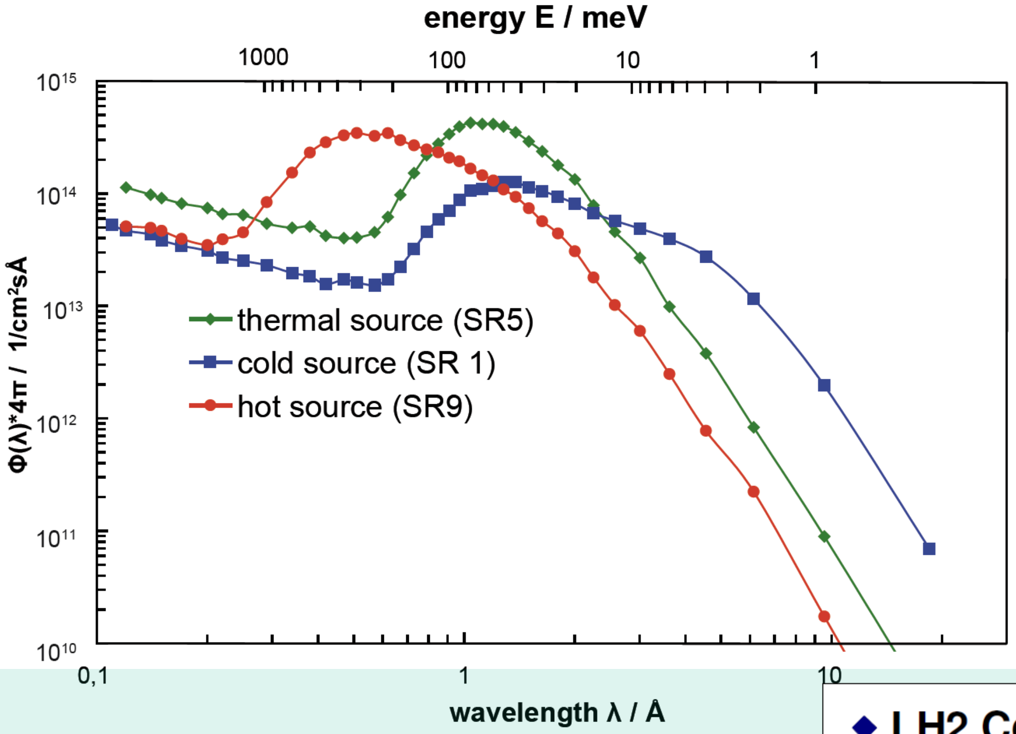
FRM II @ MLZ, Munich ↑

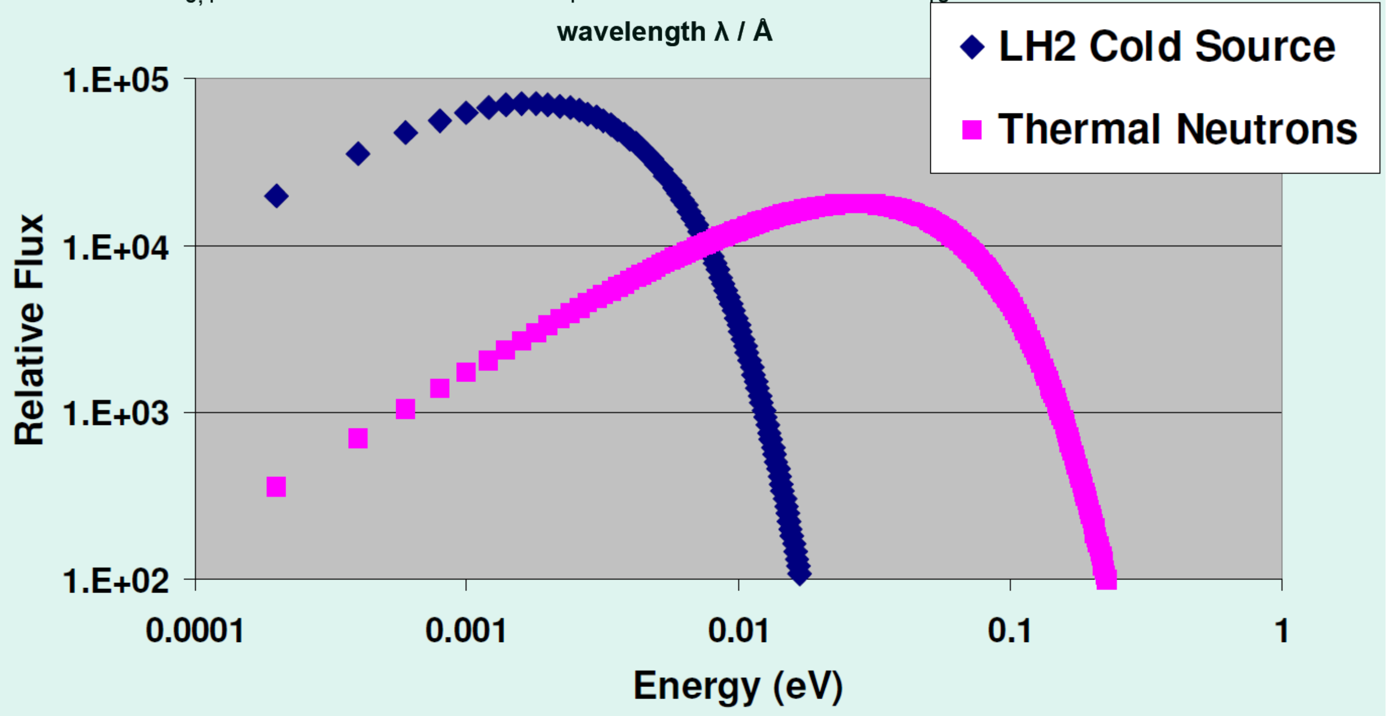
HFR @ ILL, Grenoble ↓

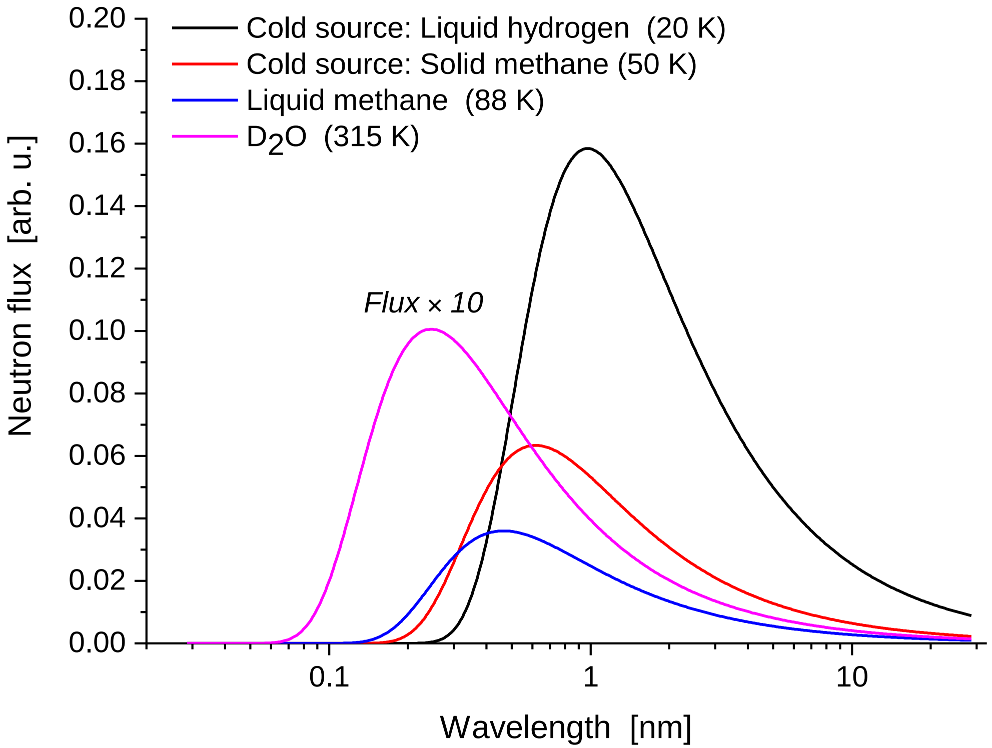


NSBR, NIST.

Neutrons after moderation

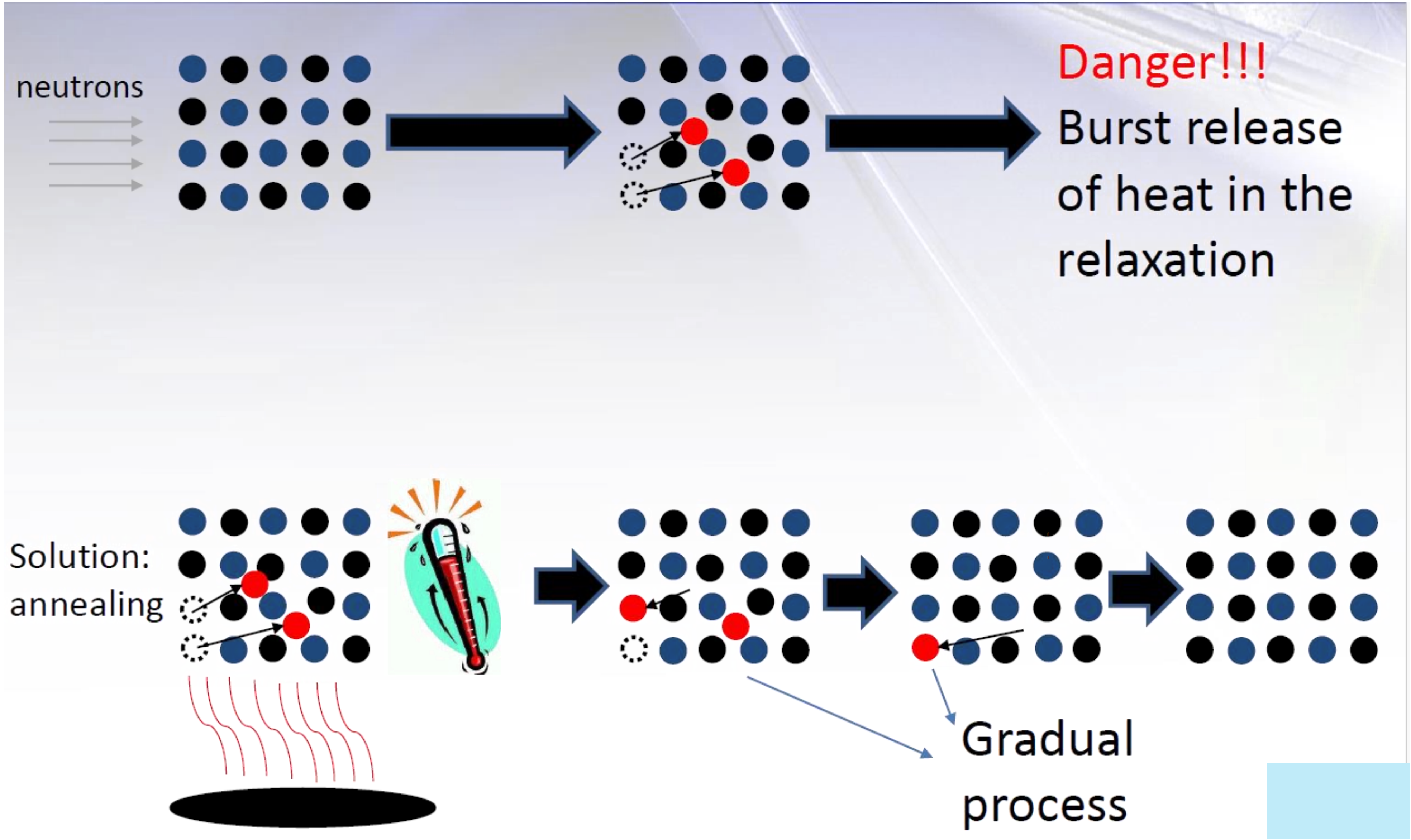






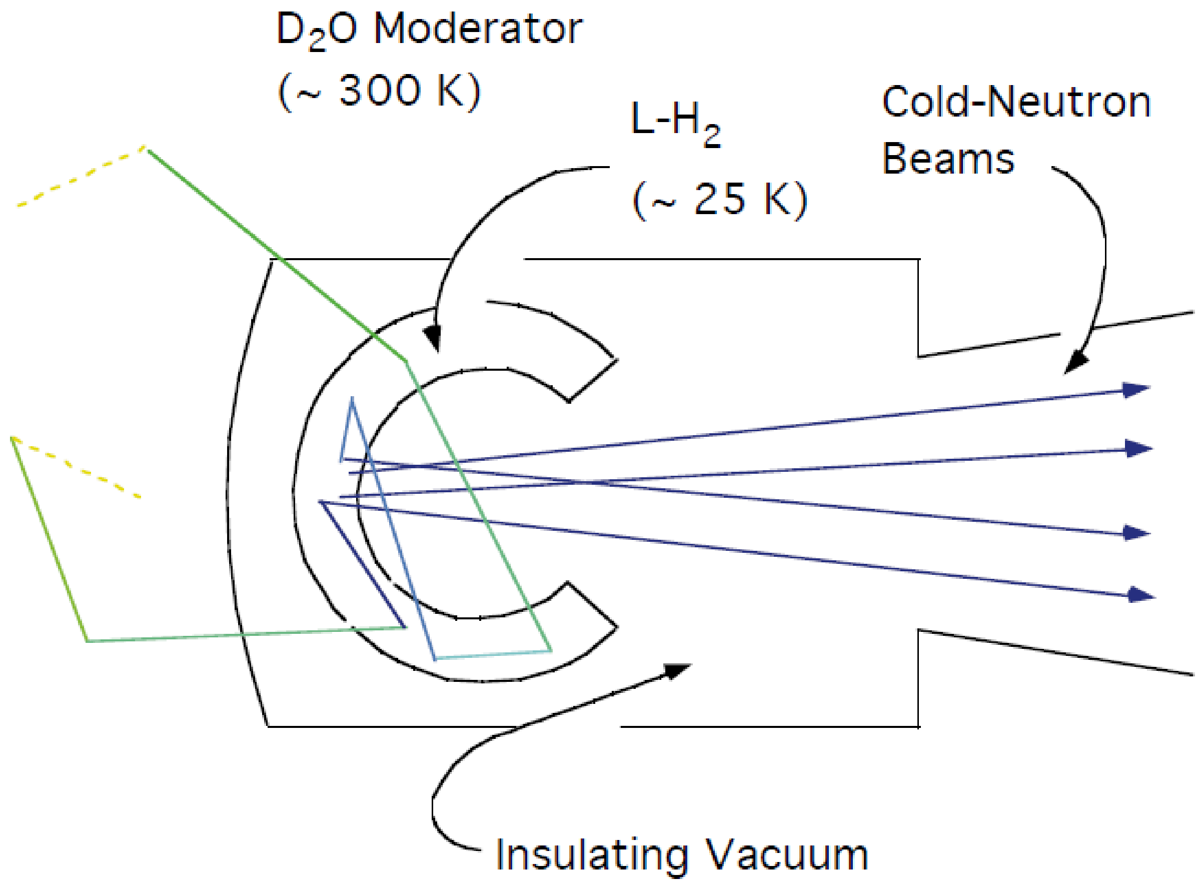
Neutron spectra at the entrance of the beam tubes at 20 MW reactor power at FRM II (top left) and NSBR, NIST (bottom right) compared to theoretical Maxwellian.

Neutron moderation



The Wigner effect.

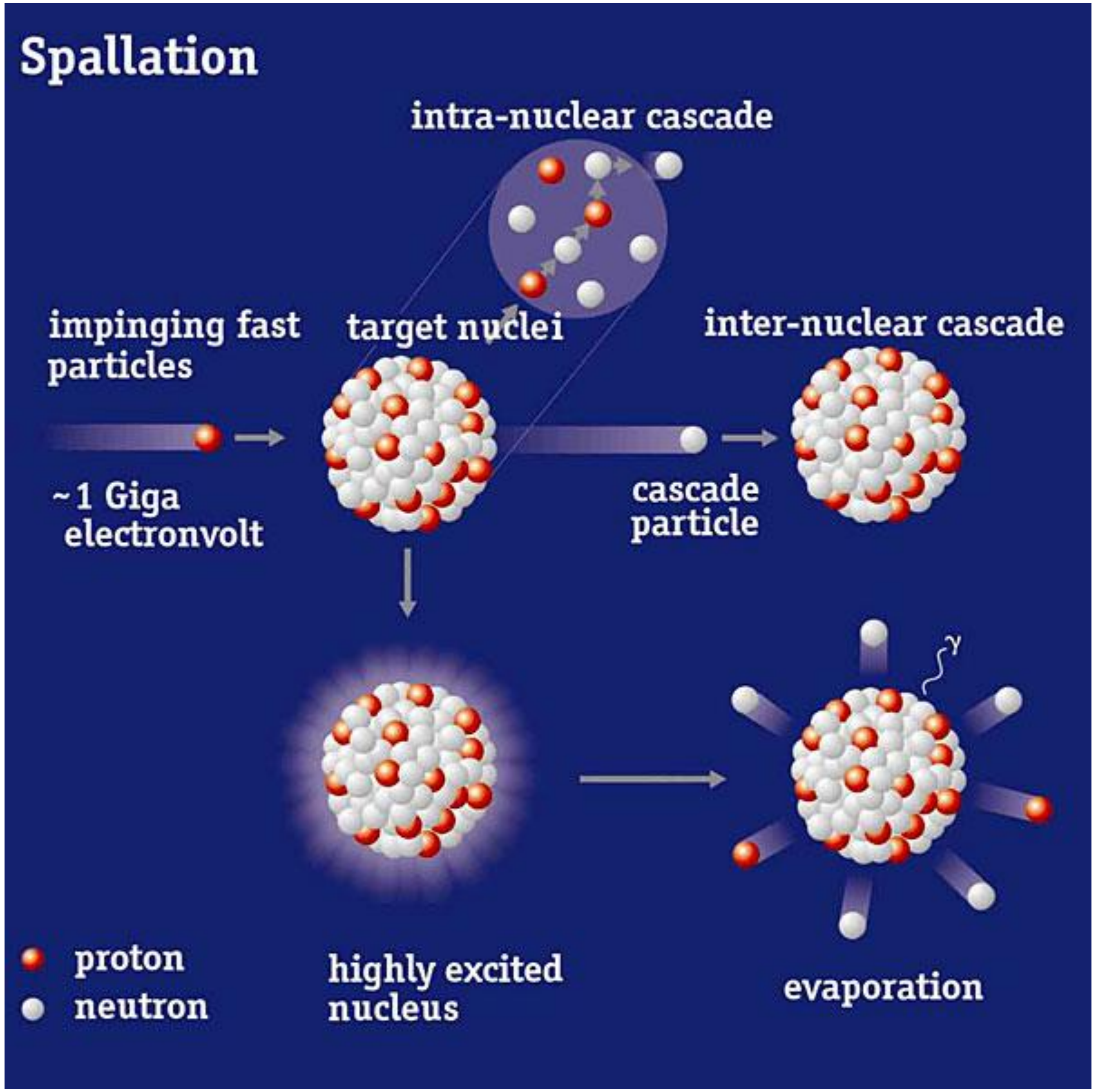
Neutron moderation



Commonj neutron moderators

	Neutron scatt. X-section (σ_s) [b]	Neutron abs. X-section (σ_a) [b]
Light water (H_2O)	49	0.66
Heavy water (D_2O)	10.6	0.0013
Graphite (C)	4.7	0.0035

- *Solids are dangerous as cold moderators due to Wigner energies.*
- *Complex liquids will dissociate as a result of the radiation.*
- *Usual choices are liquid H and D.\.*

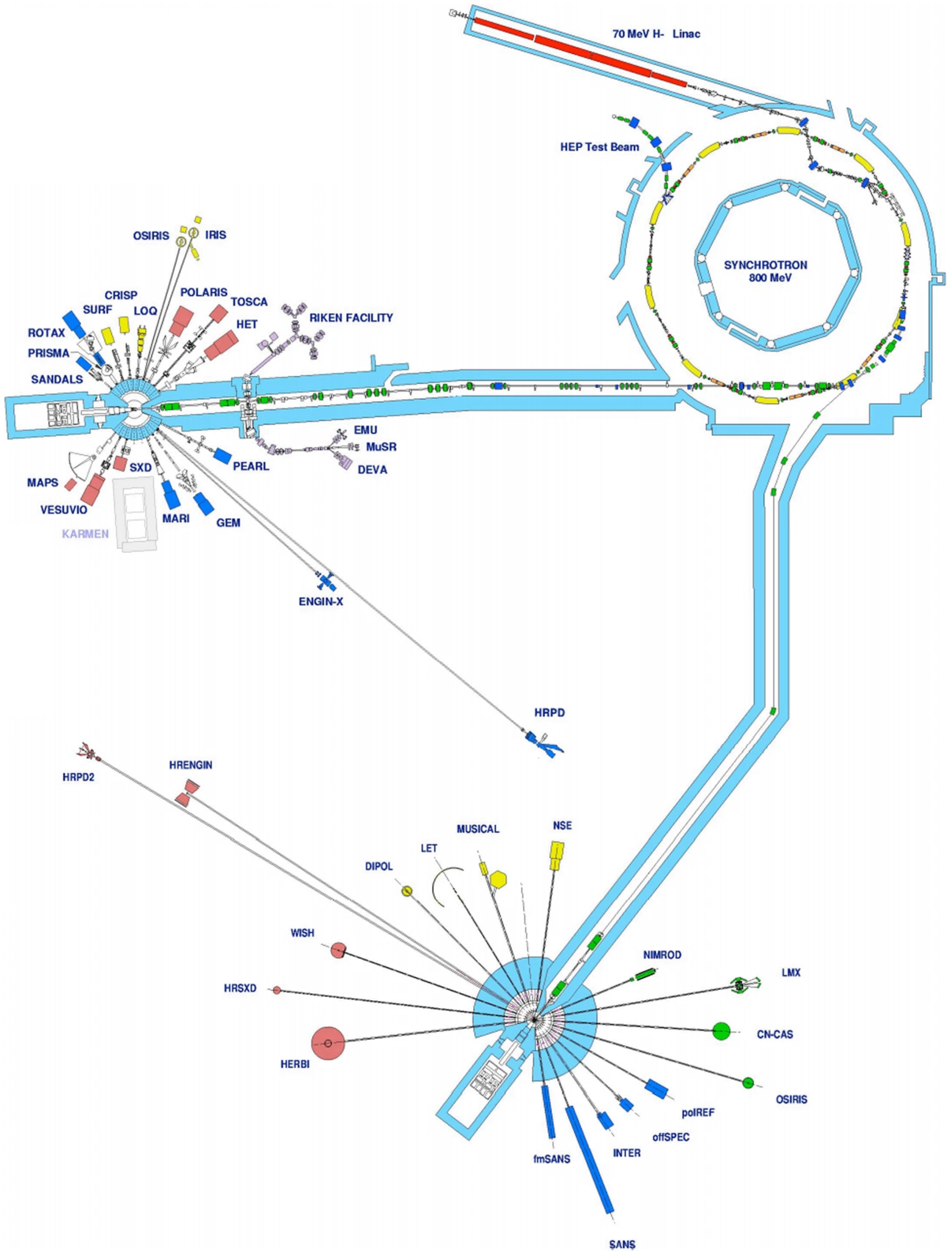


(b) Nuclear spallation

A spallation source produces pulsed or quasi-continuous neutron beams by the acceleration of protons hitting the target material of heavy nuclei, such as mercury, tantalum, or lead. In contrast to the fission of heavy nuclides, one spallation reaction, however, releases about 20 – 30 neutrons per incident particle.

- ISIS, RAL, U.K. (two target stations)
- SNS, USA
- J-PARC, Japan
- SINQ, Switzerland (quasi-continuous)
- ESS, Sweden. The strongest. Under construction.

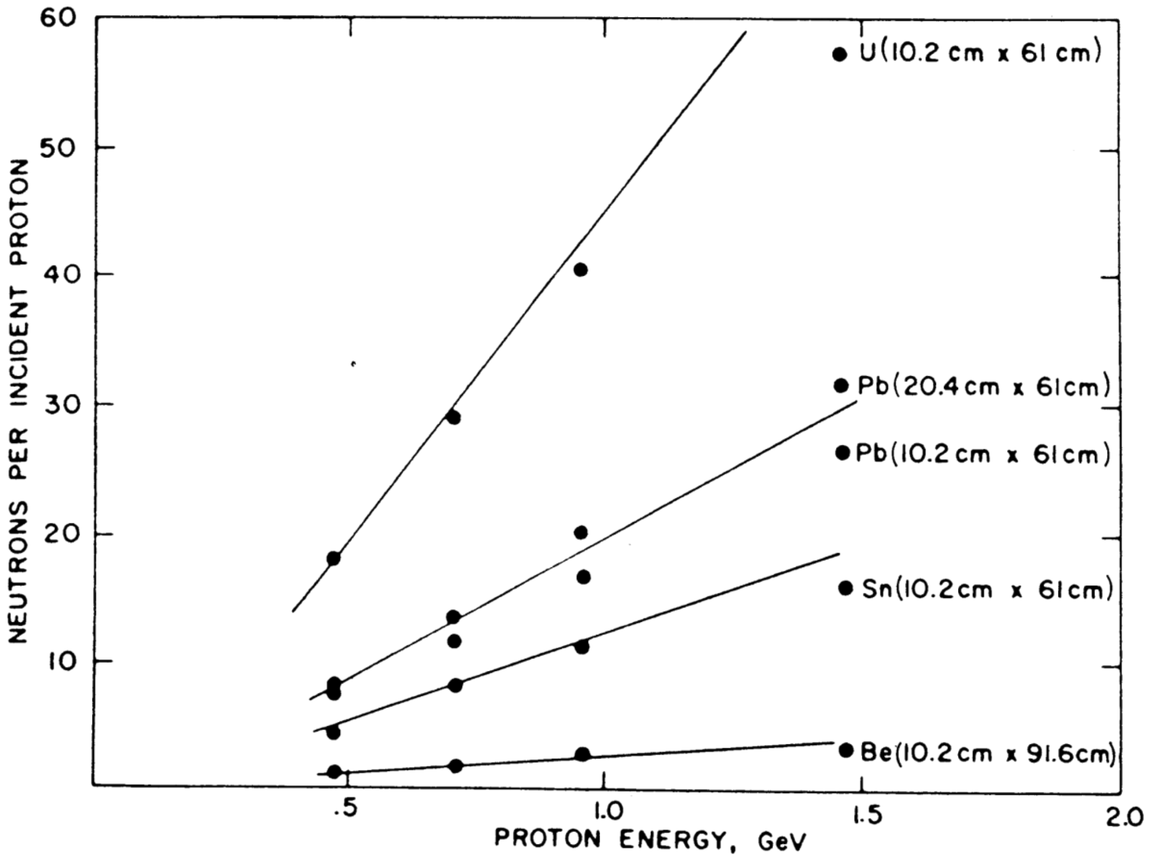
ISIS Neutron Source

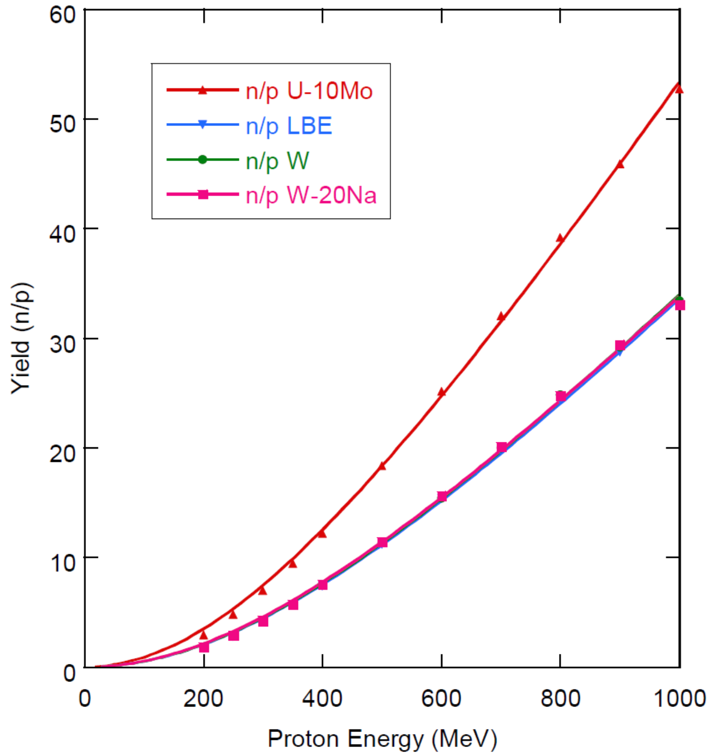


Source type	Examples	Power (MW)	Typical approximate neutron output (s^{-1})
Proton accelerator	CSNS	0.1	1×10^{16}
	ESS (~2023)	5	1×10^{18}
	ISIS	0.2	2×10^{16}
	J-PARC	1.0	1×10^{17}
	Los Alamos	0.1	1×10^{16}
	PSI (not pulsed)	1.3	1×10^{17}
	SNS	1.3	1×10^{17}
Reactor	ILL, Grenoble	58	1.5×10^{15} flux ($cm^{-2}\ s^{-1}$)
	HFIR, ORNL	85	2.5×10^{15} flux ($cm^{-2}\ s^{-1}$)

A beam of high-energy protons strikes a heavy metal target — tantalum, tungsten, mercury, lead, or even uranium. A high-Energy proton hitting a heavy-metal nucleus can knock a few particles or clusters of particles out of the nucleus and can also split the rest of the nucleus into two excited fragments. The fragments then de-excite by emitting neutrons with energies of the order of 1 MeV, and the knocked-out particles can go on to induce further spallation reactions. The overall result at ISIS is some ~10-15 neutrons produced per incident proton.

ISIS Neutron Source





Fraser's neutron yield calculation (J.S. Fraser, R.E. Green, et al., Physics in Canada 21 (2), 17, (1965))

- *W —38 n/p for 2 GeV protons*
- *Hg (Fraser's calc.) —19.4 neutrons per proton for 1 GeV p.*
- *Hg — 25 n/p for 1 GeV p (@SNS)*
- *^{126}Sn — 40 n/p*

A useful feature of spallation neutron production is that the energy deposited in the target per neutron produced is quite small.

European Spallation Source

1 Protons are generated in the ion source

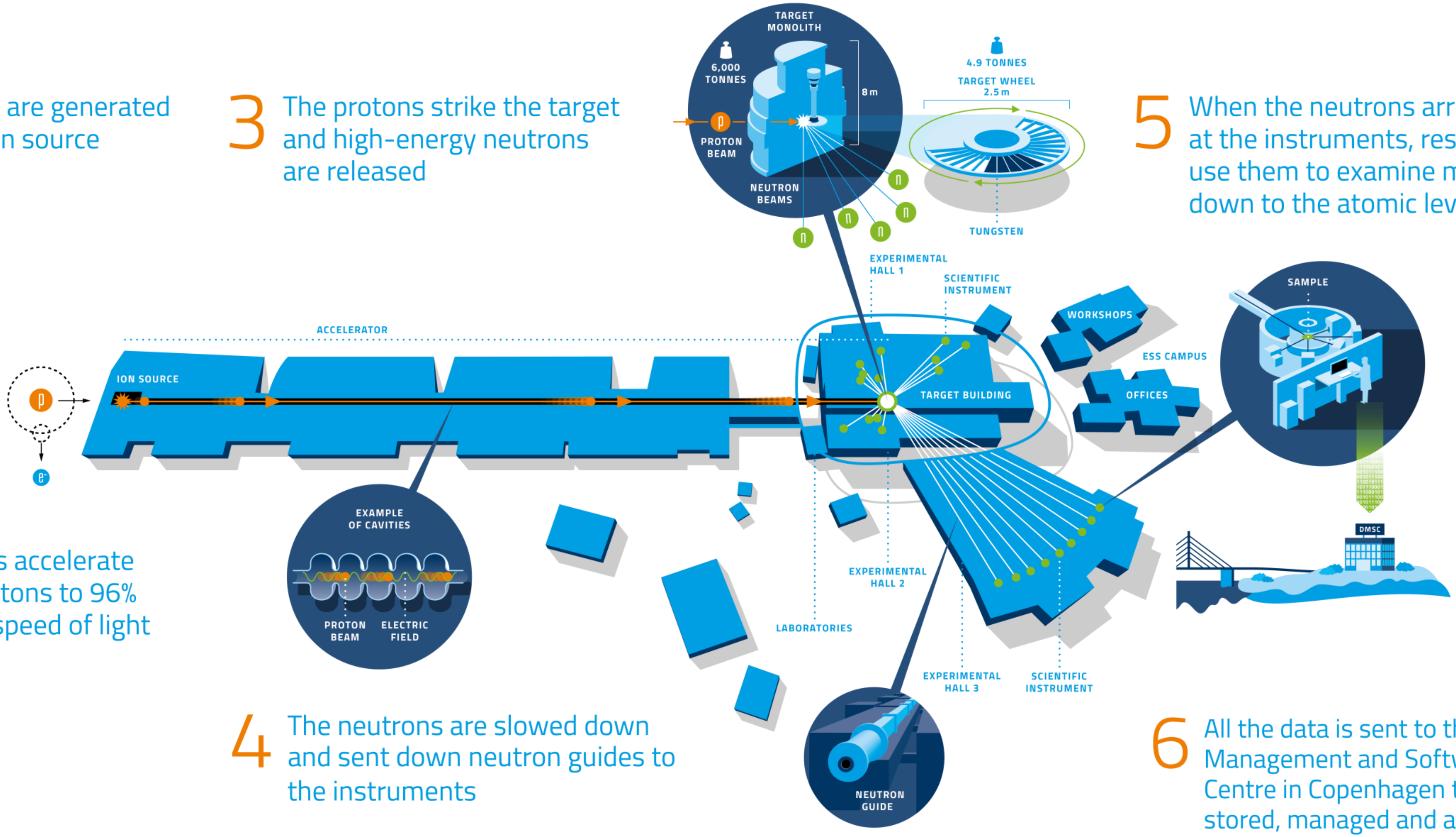
2 Cavities accelerate the protons to 96% of the speed of light

3 The protons strike the target and high-energy neutrons are released

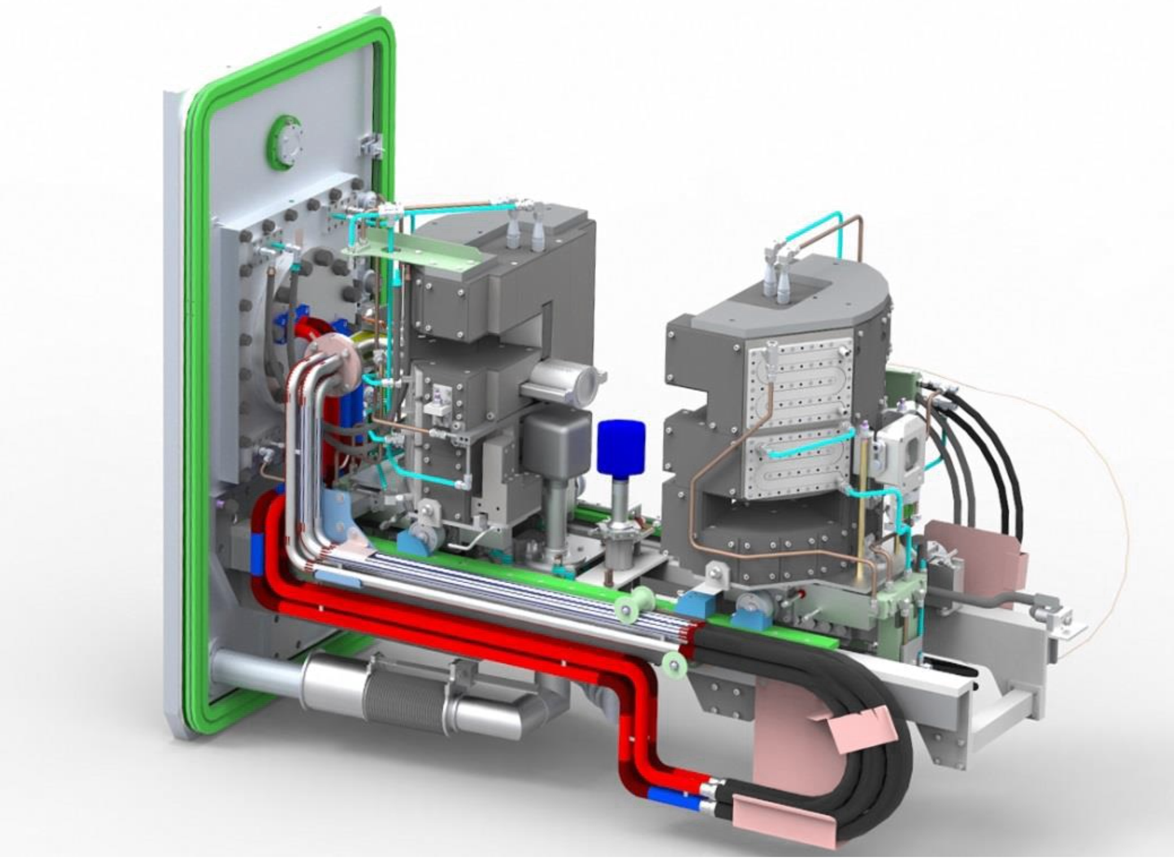
4 The neutrons are slowed down and sent down neutron guides to the instruments

5 When the neutrons arrive at the instruments, researchers use them to examine matter down to the atomic level

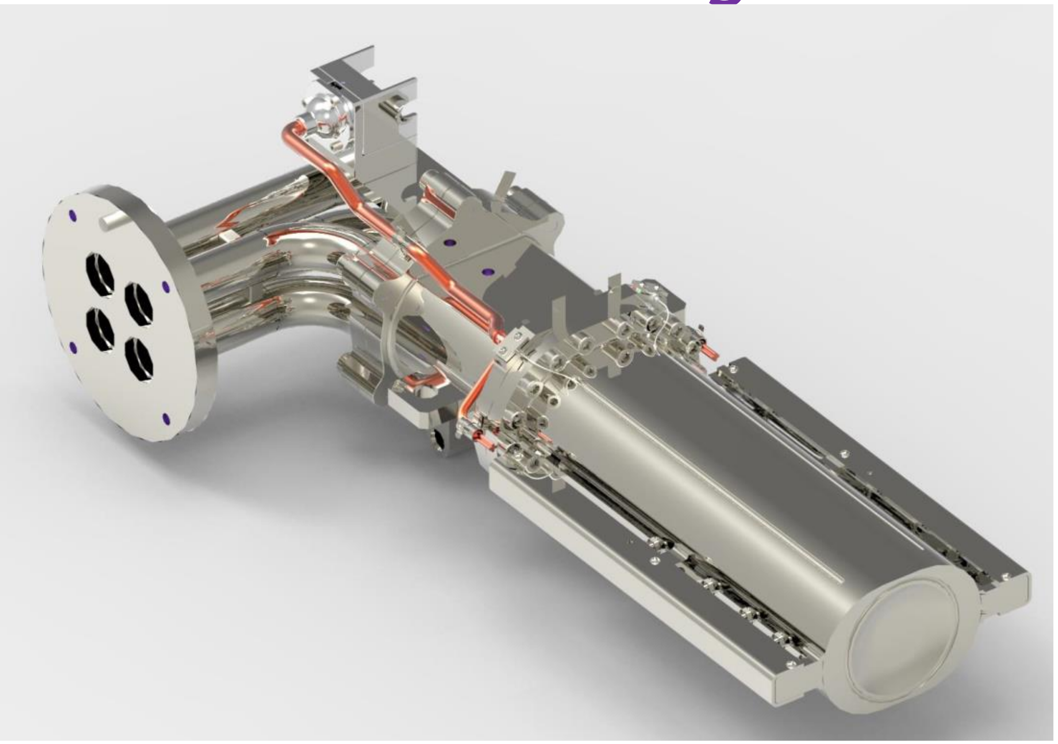
6 All the data is sent to the Data Management and Software Centre in Copenhagen to be stored, managed and analysed with the researchers

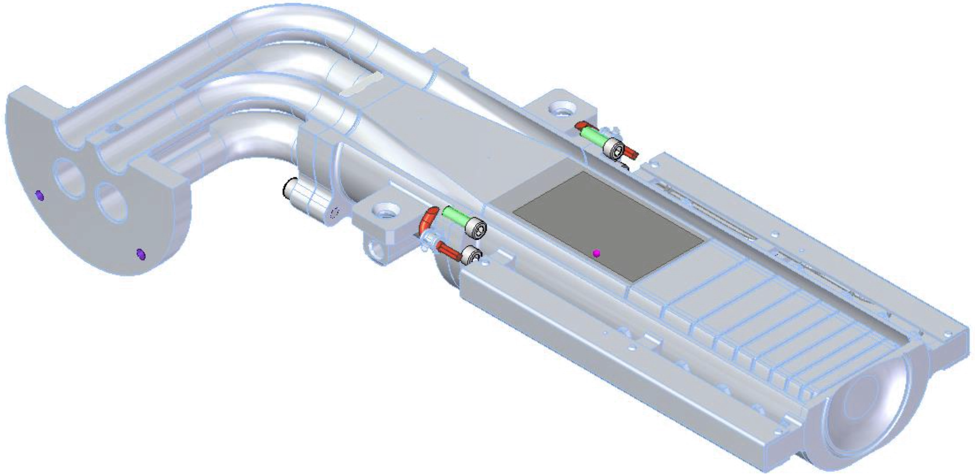


ESS target monolith



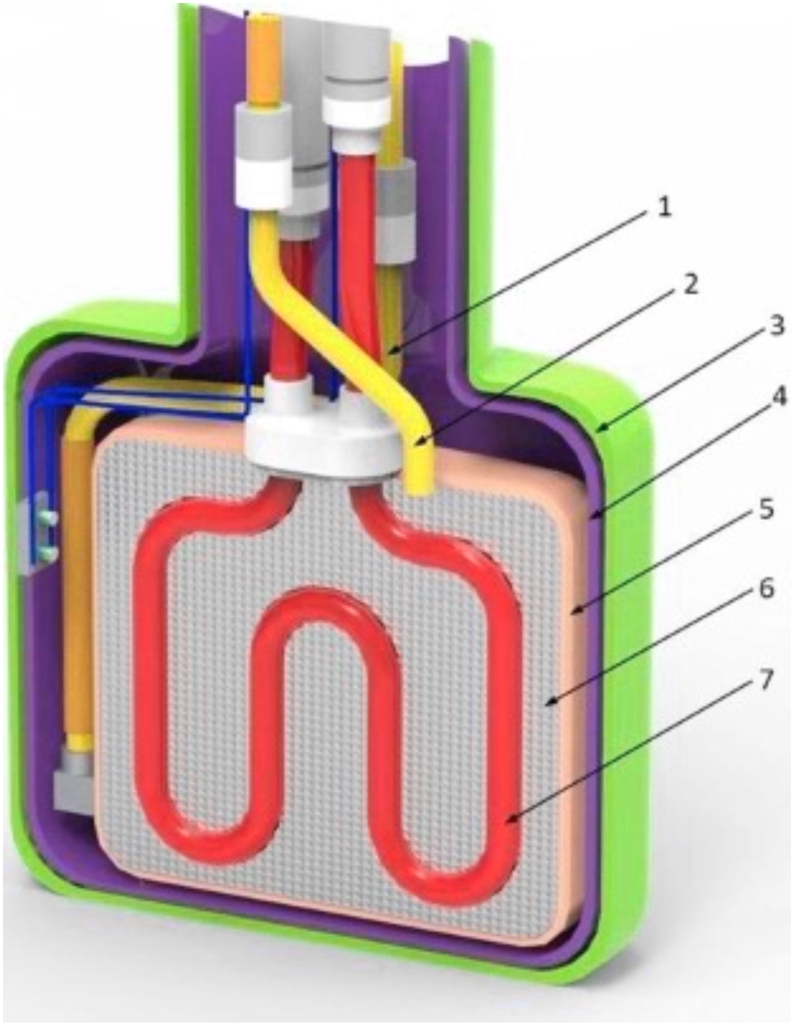
ISIS new Target Station S1 assembly.





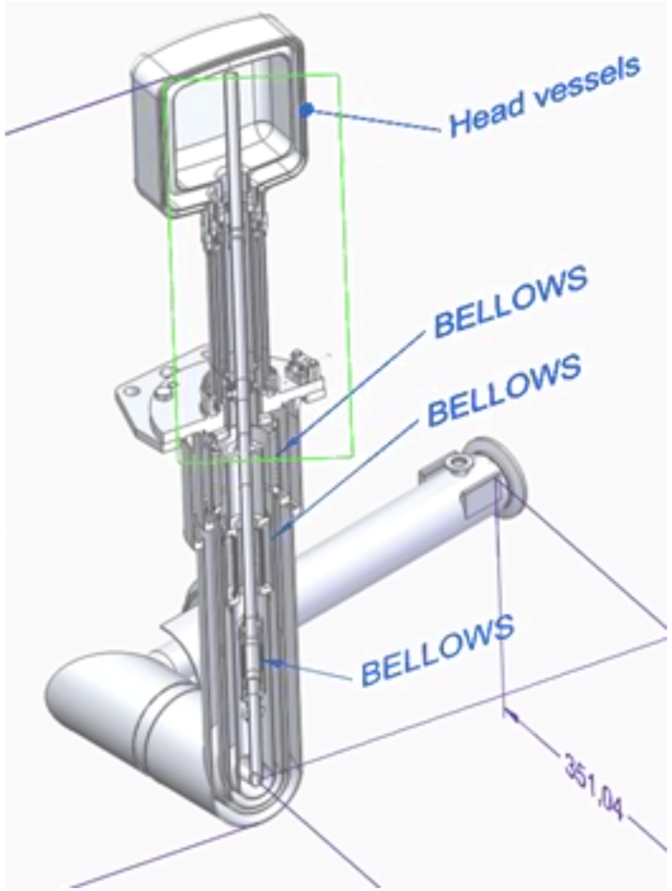
12 Tungsten plates of varying thicknesses clad in tantalum.

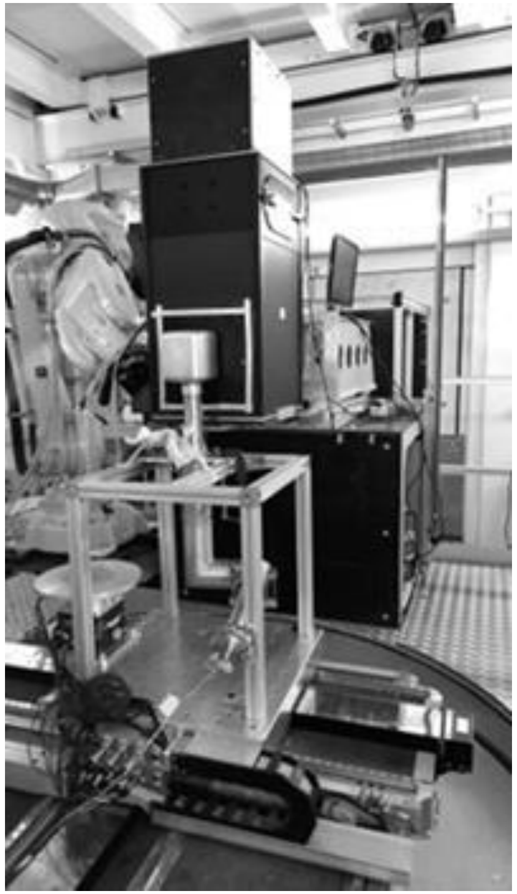
ESS target monolith



ISIS TS2 solid mehane moderator.

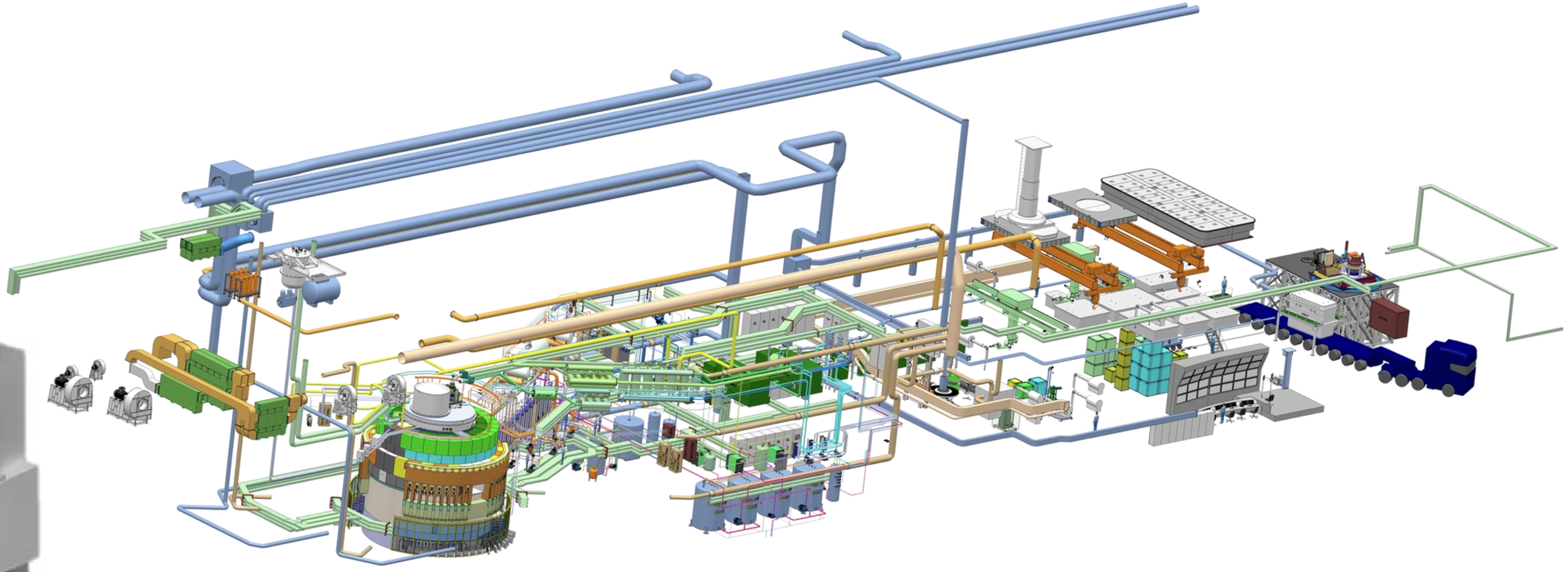
5 – moderator vessel
7 – heat exchanger



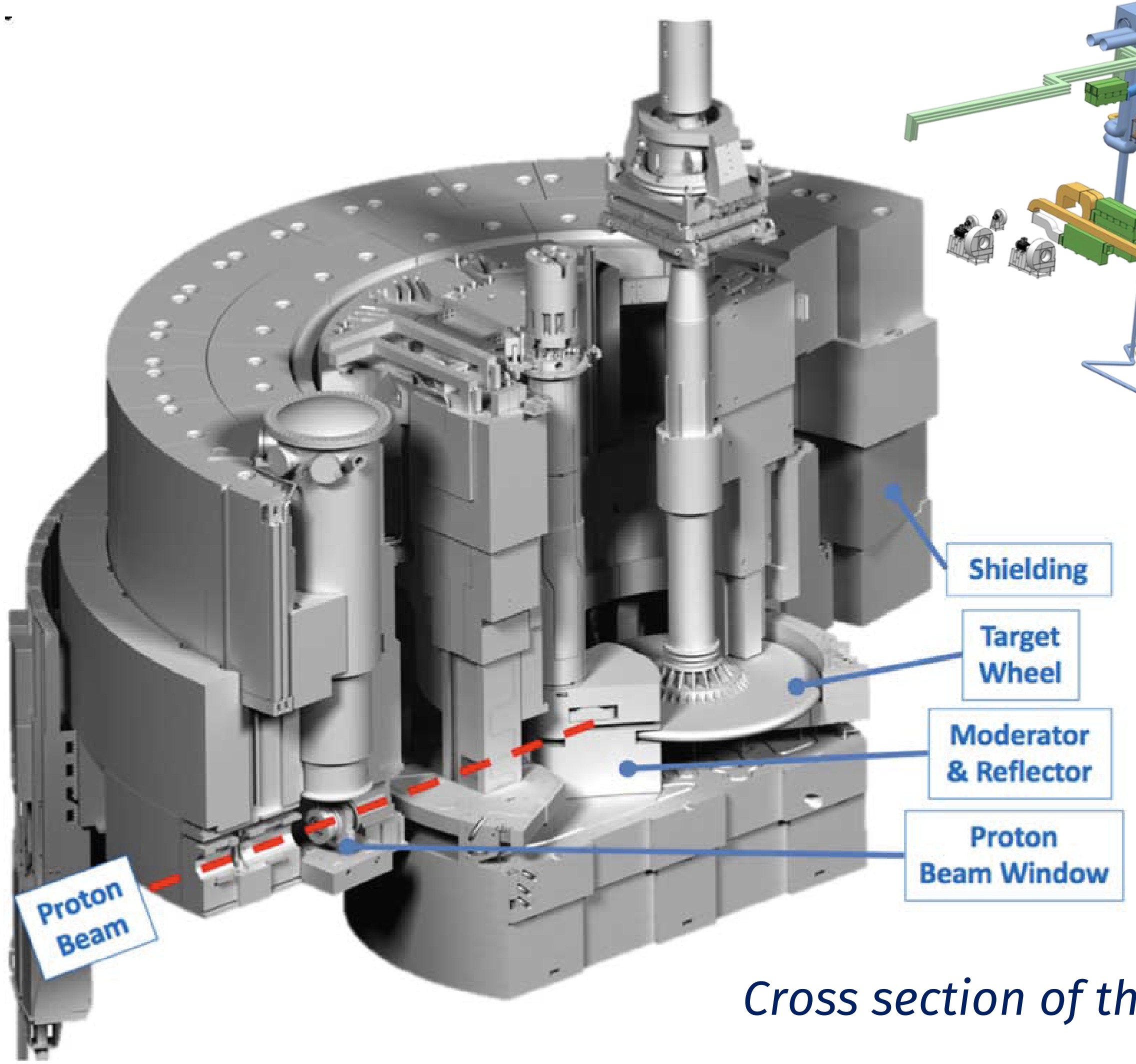


ISIS new TS1 hydrogen moderator.

ESS target monolith



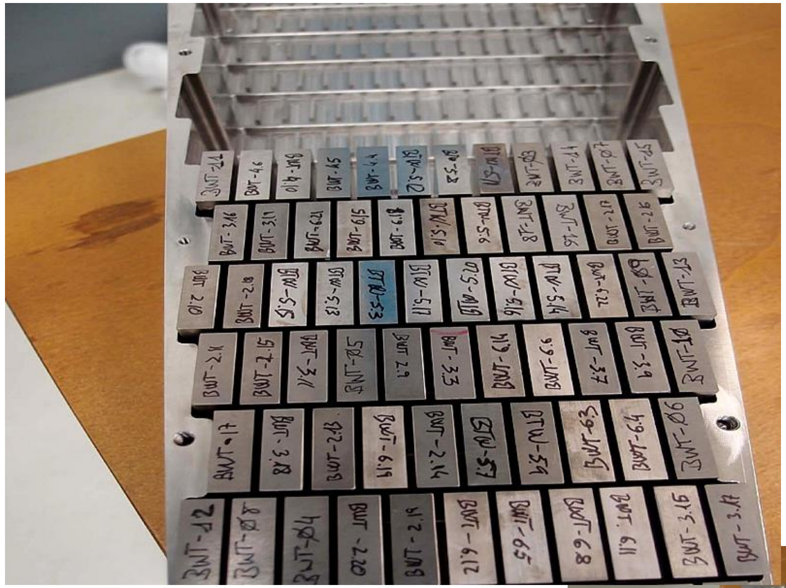
ESS target station assembly



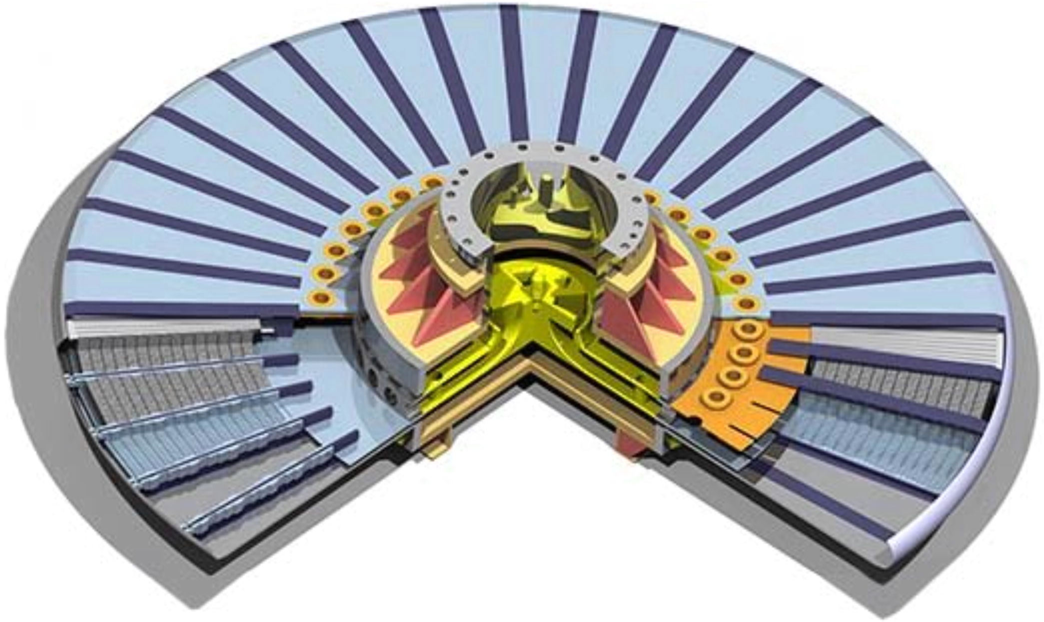
Cross section of the ESS target monolith

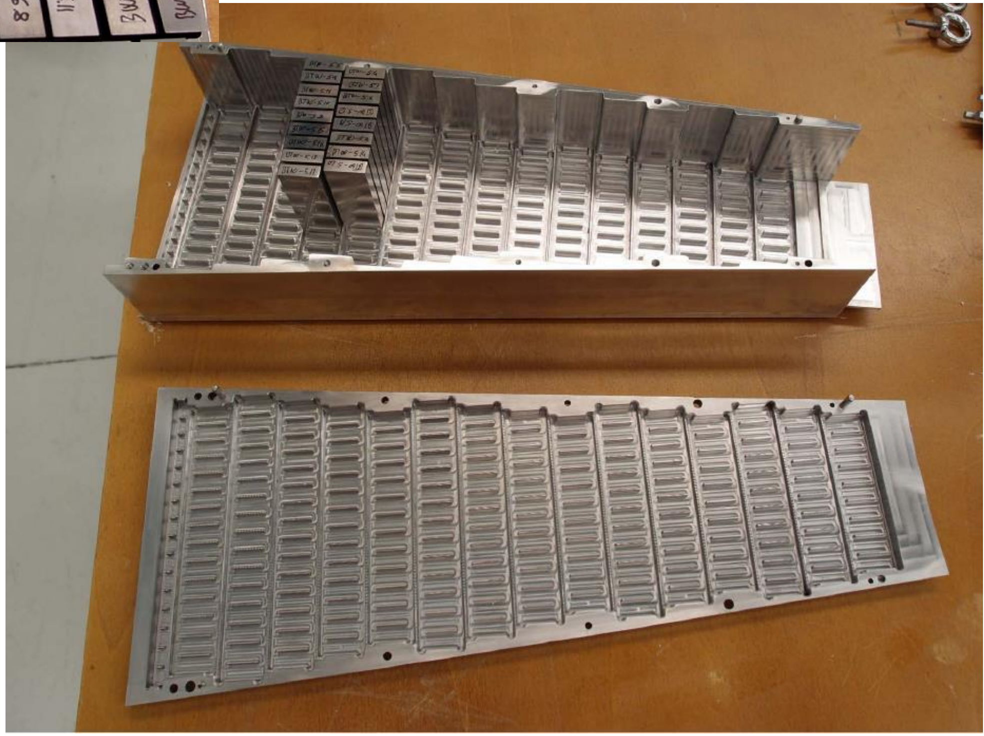
ESS target monolith


Target wheel prototype in the testbed

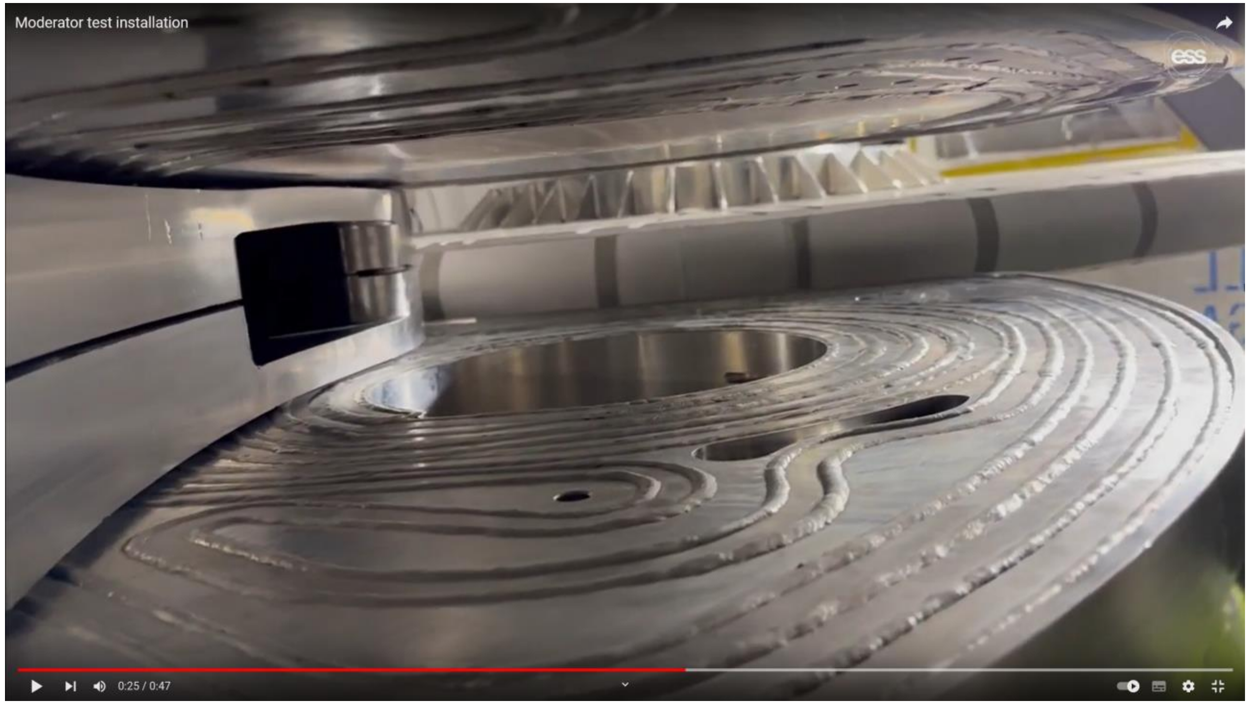


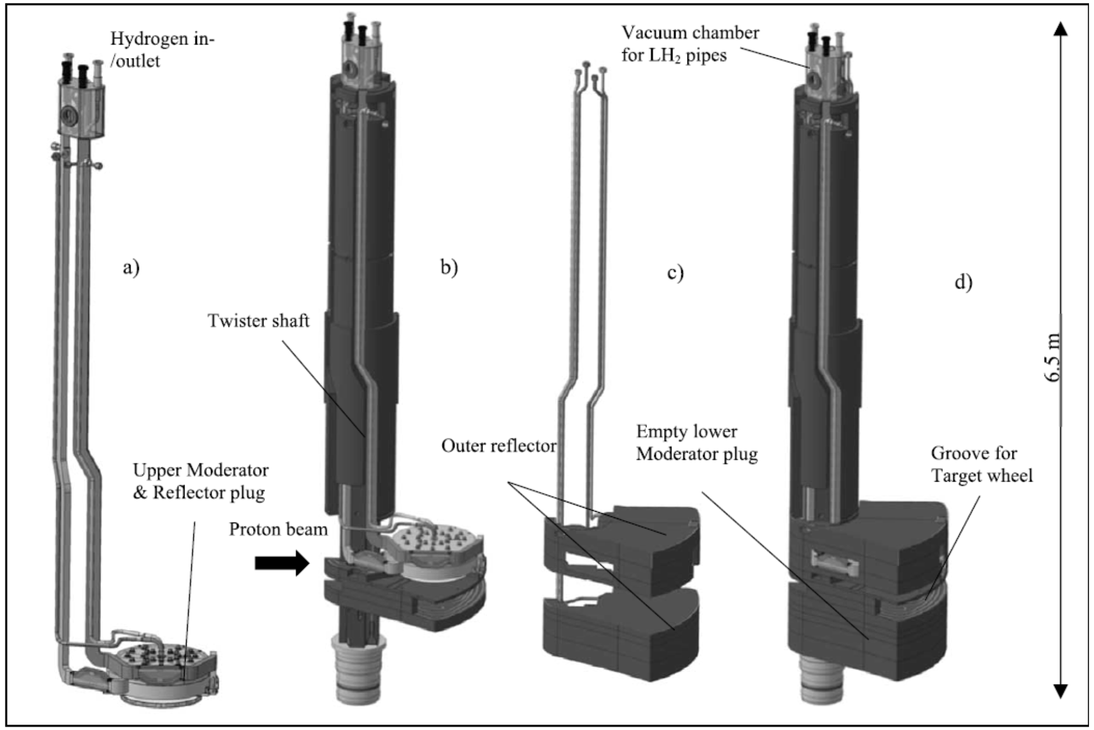
7000 tungsten bricks ready for mounting on the ESS target wheel prototype.



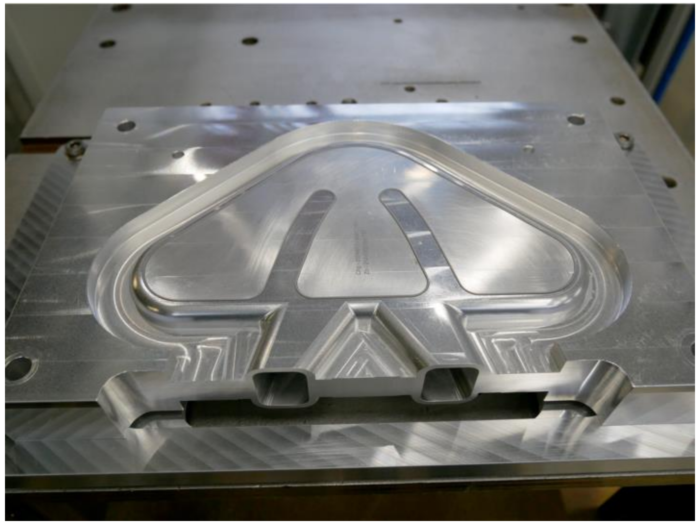


ESS moderators

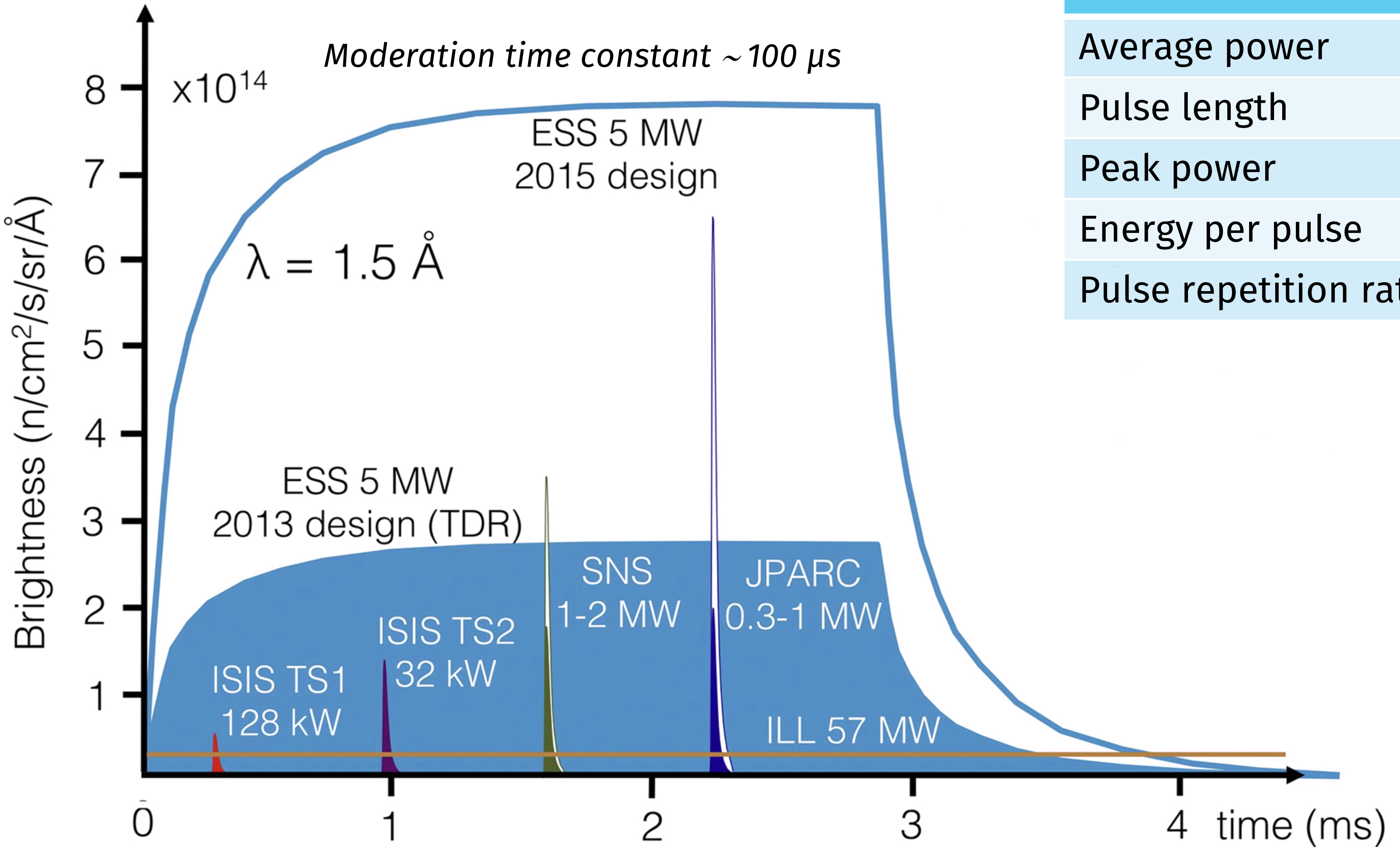




„Butterfly” moderator



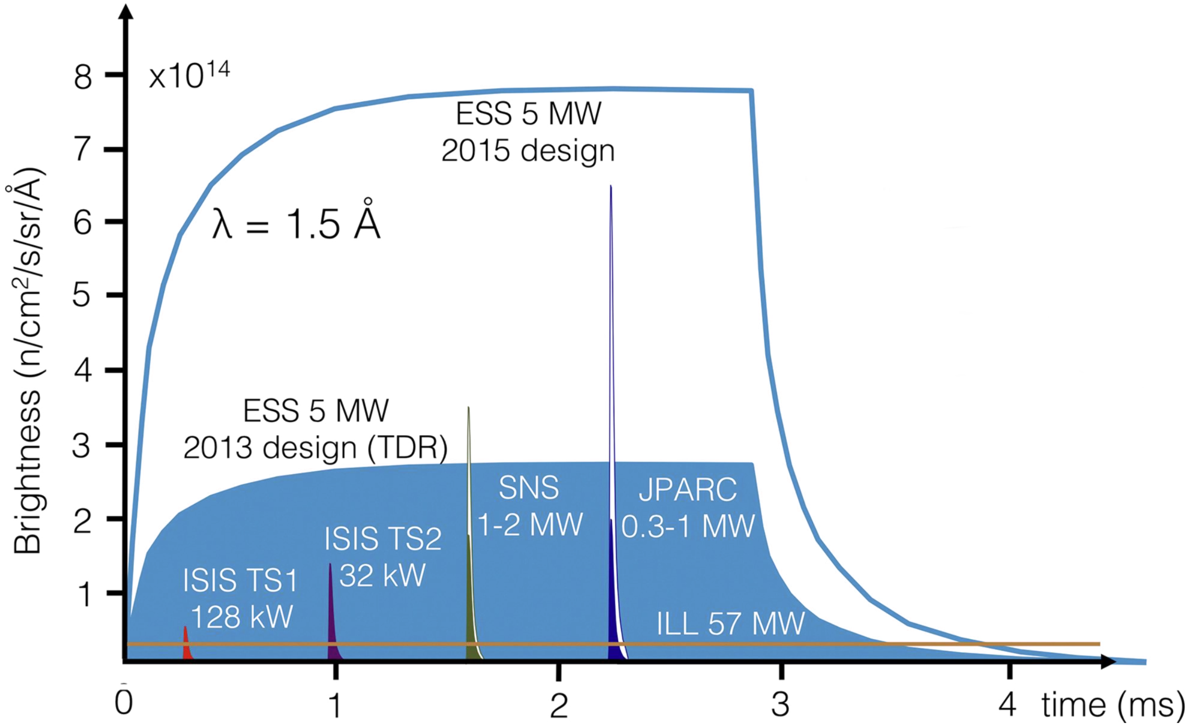
ESS – A long-pulse spallation source

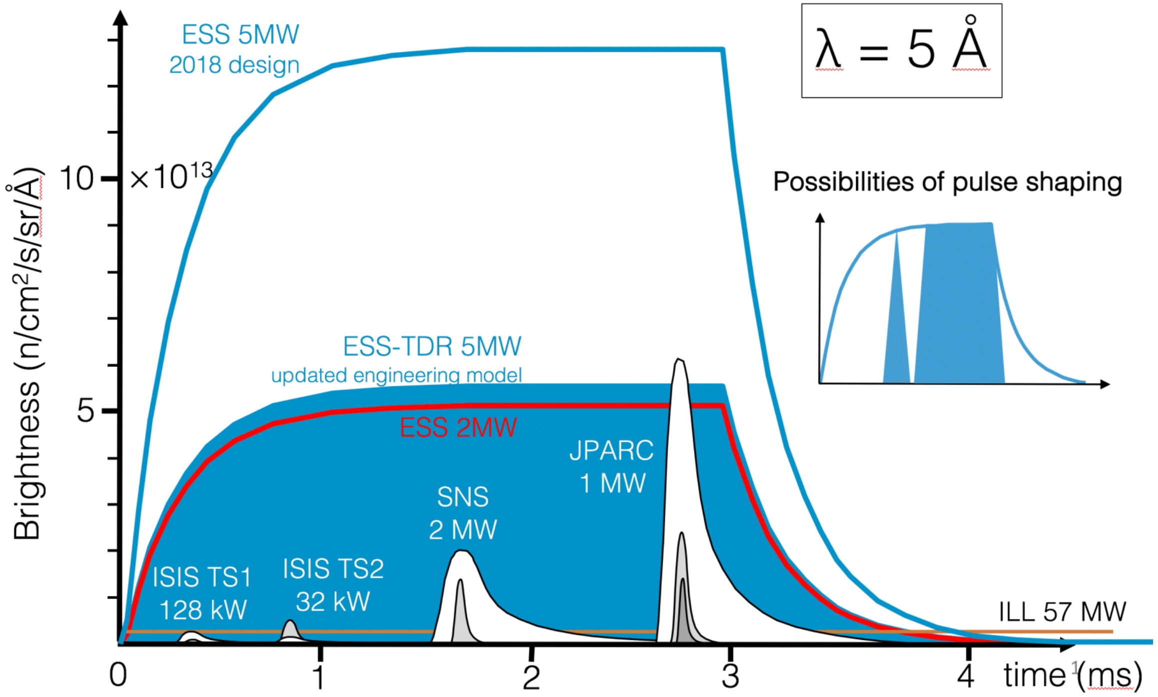


	SNS	ESS
Average power	1.4 MW	5 MW
Pulse length	695 ns	2.86 ms
Peak power	34 GW	125 MW
Energy per pulse	24 kJ	357 kJ
Pulse repetition rate	60 Hz	14 Hz

Time-averaged brightness:
ESS ~ ILL
Peak brightness:
ESS ~ 30 × ILL

Spallation sources pulse shape



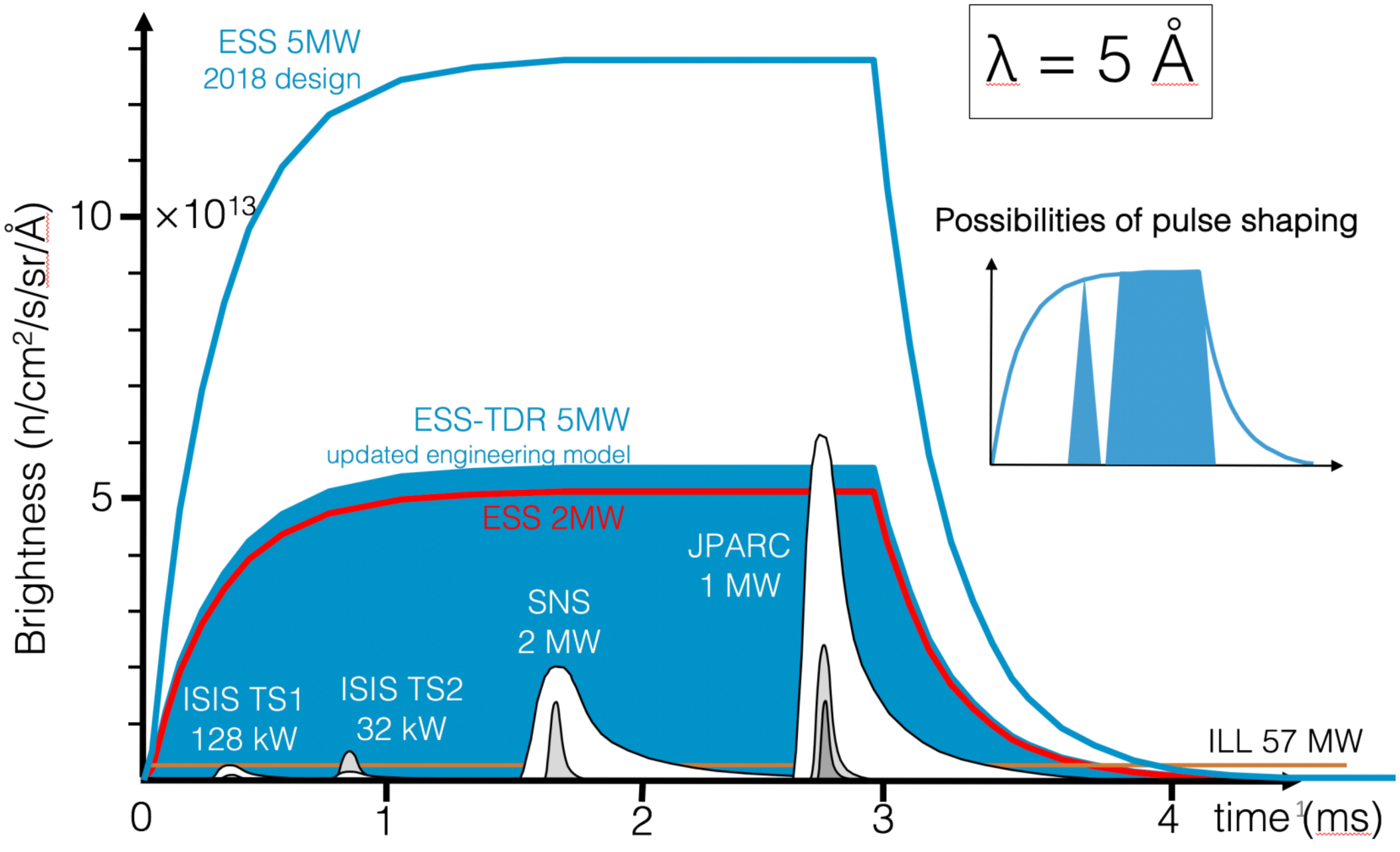


Mind the wavelength dependece!

The ISIS Second Target Station is a low-power, low-repetition-rate neutron source optimised to maximise the production of long wavelength neutrons. It is required to produce neutrons of two pulse shape varieties.

The first, a wide pulse shape with full width at half-maximum height (FWHM) no bigger than 300 μs and a modest tail, is generated by a coupled moderator; whilst the second is a simple pulse shape de-coupled moderator, with little or no tail, and a width comparable to those of the existing ISIS methane and hydrogen moderators (30–50 μs).

Spallation sources pulse shape



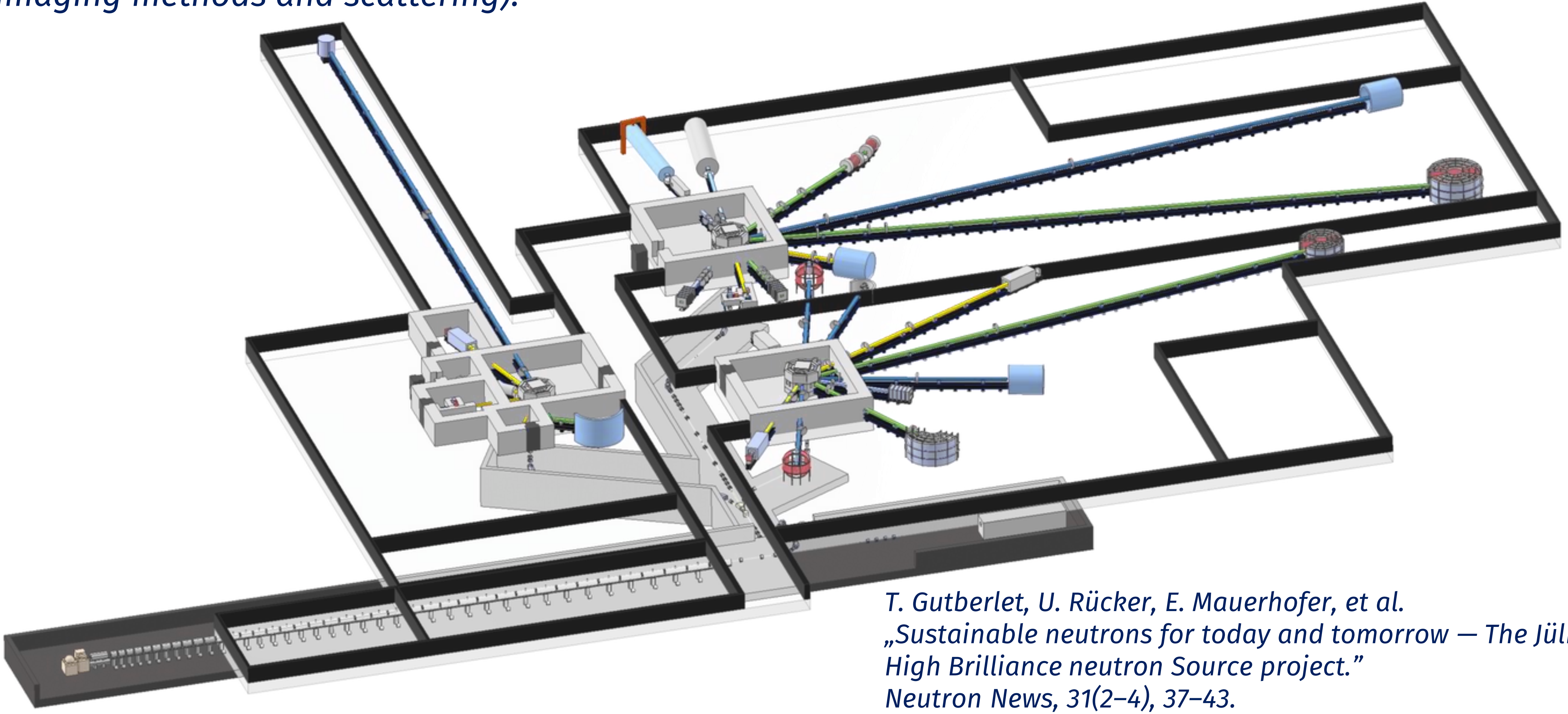
The ISIS Second Target Station is a low-power, low-repetition-rate neutron source optimised to maximise the production of long wavelength neutrons. It is required to produce neutrons of two pulse shape varieties.

The first, a wide pulse shape with full width at half-maximum height (FWHM) no bigger than 300 µs and a modest tail, is generated by a coupled moderator; whilst the second is a simple pulse shape de-coupled moderator, with little or no tail, and a width comparable to those of the existing ISIS methane and hydrogen moderators (30–50 µs).

(c) High Brilliance Neutron Source (HBS)

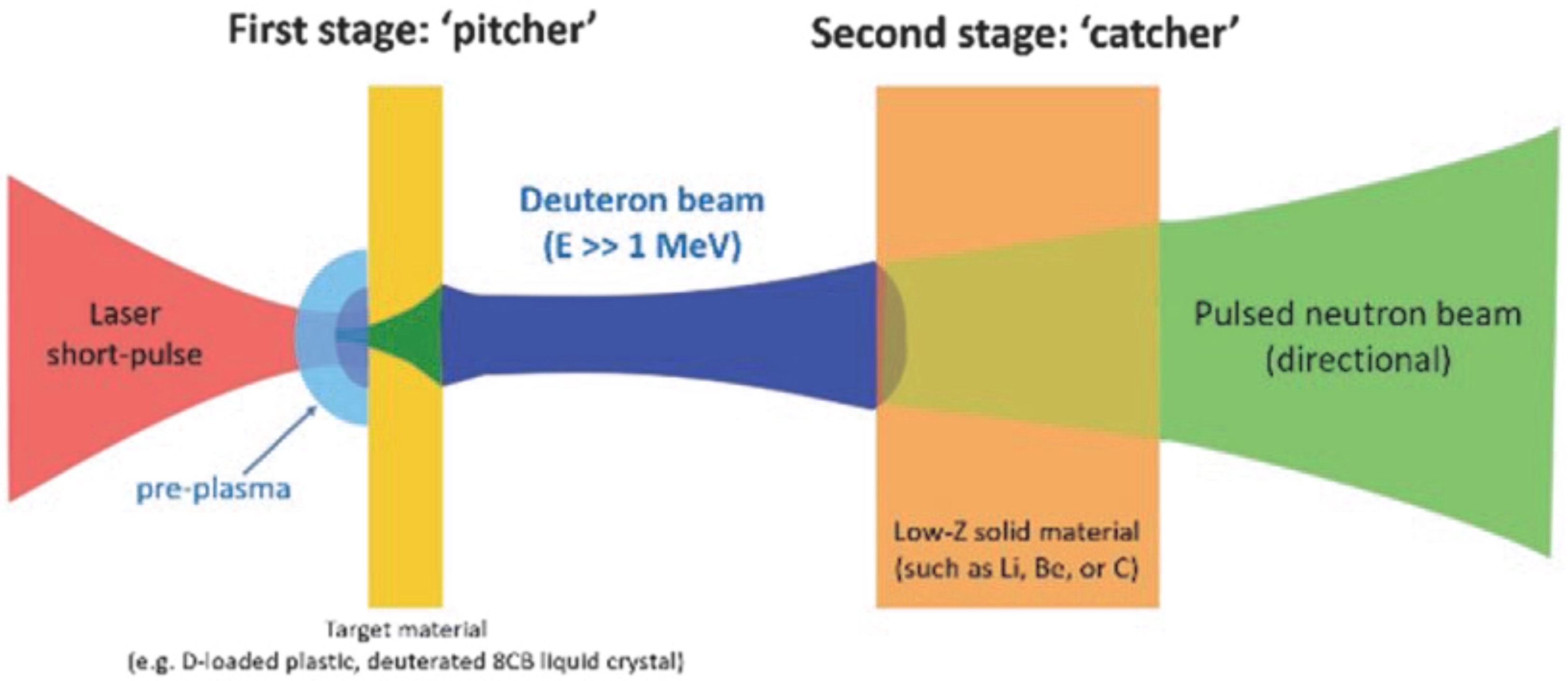
(Small and compact)

The project of a high brilliance accelerator-based neutron source HBS aims to develop and establish a unique infrastructure for neutron analysis (imaging methods and scattering).



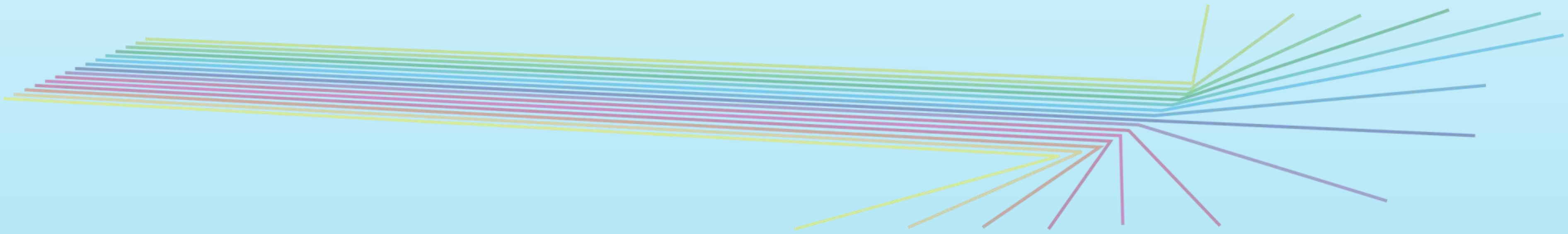
T. Gutberlet, U. Rücker, E. Mauerhofer, et al.
„Sustainable neutrons for today and tomorrow — The Jülich High Brilliance neutron Source project."
Neutron News, 31(2–4), 37–43.
DOI: 10.1080/10448632.2020.1819132

(d) Short-pulse laser-driven neutron source

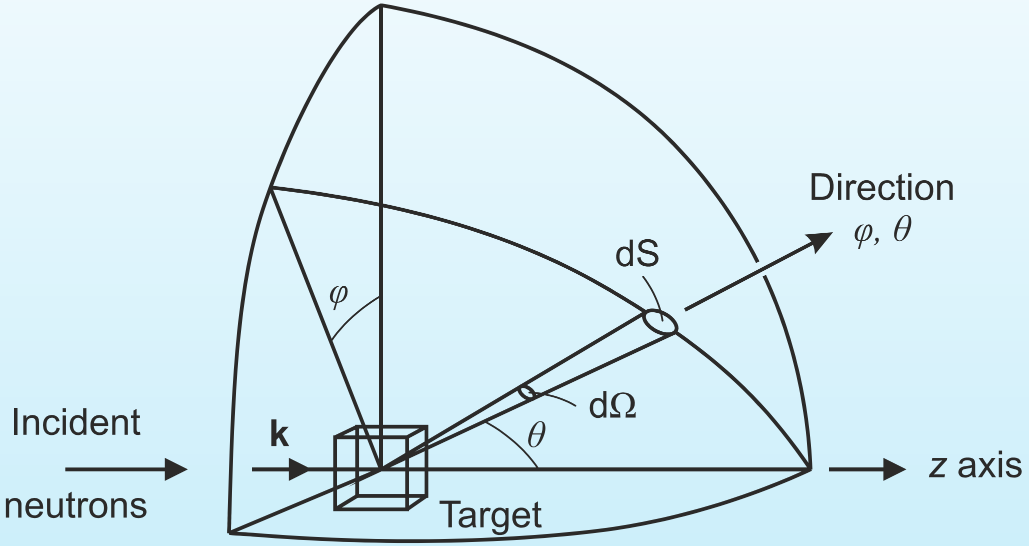


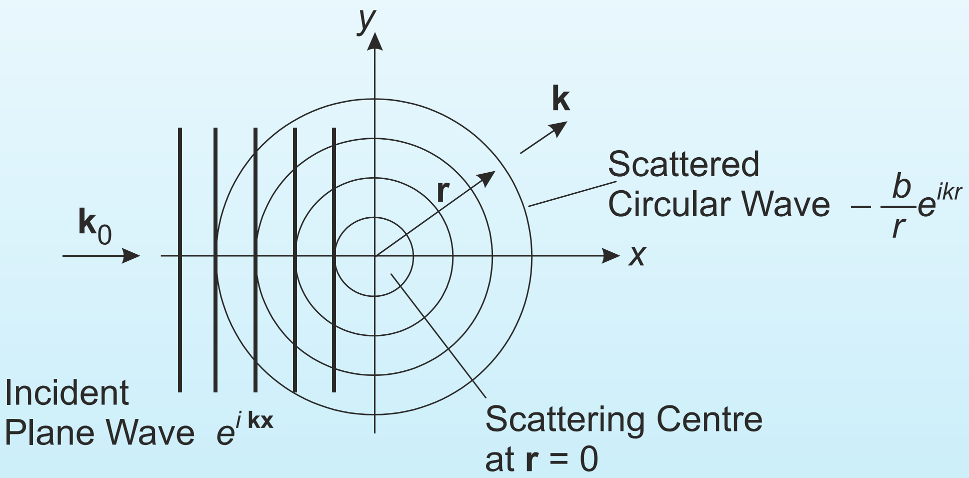
The laser beam is concentrated to a peak intensity up to 10^{21} W/cm^2. That intensity, when interacting with a sub-μm ultrathin CD (or CD_2) foil target, drives a high-energy (peaked around 15–30 MeV) deuteron beam. The latter then interacts with a beryllium target („catcher") to produce neutrons.

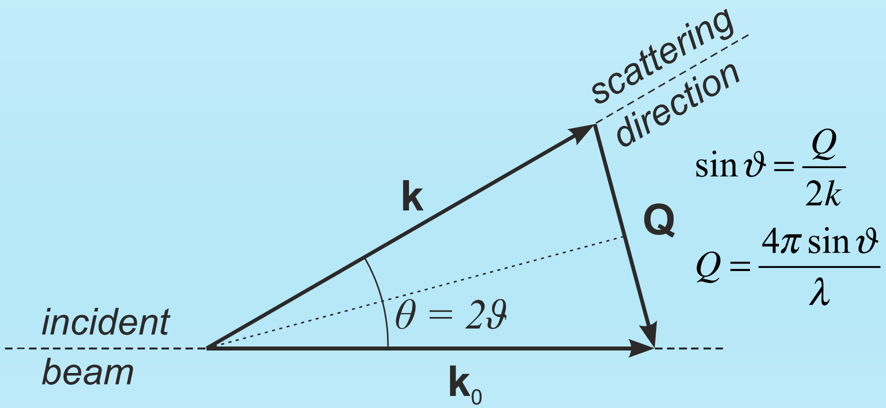
IV. Scattering on a nucleus



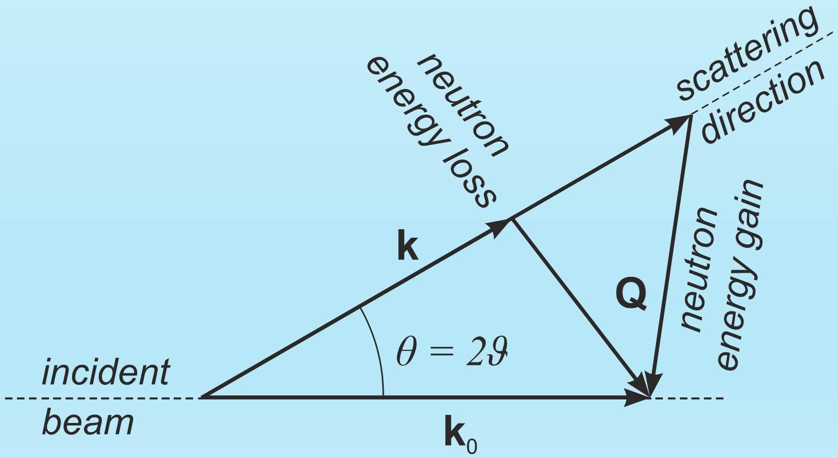
Scattering geometry







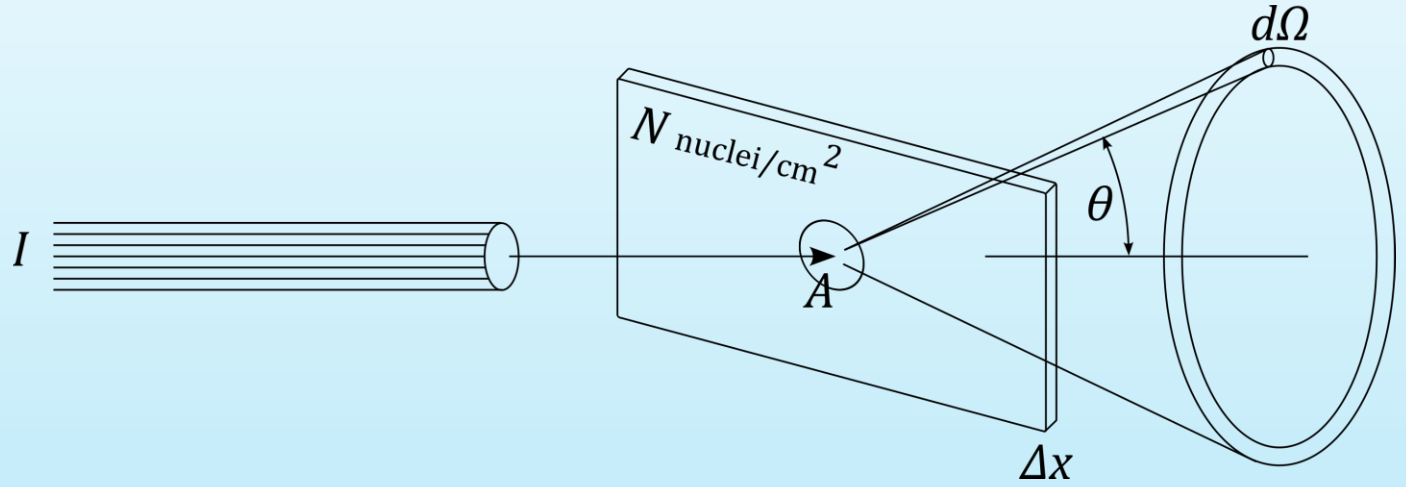
Elastic scattering



Inelastic scattering

The scattering cross section: a heuristic approach

The physical meaning of a cross section σ is a measure of the probability of a reaction.



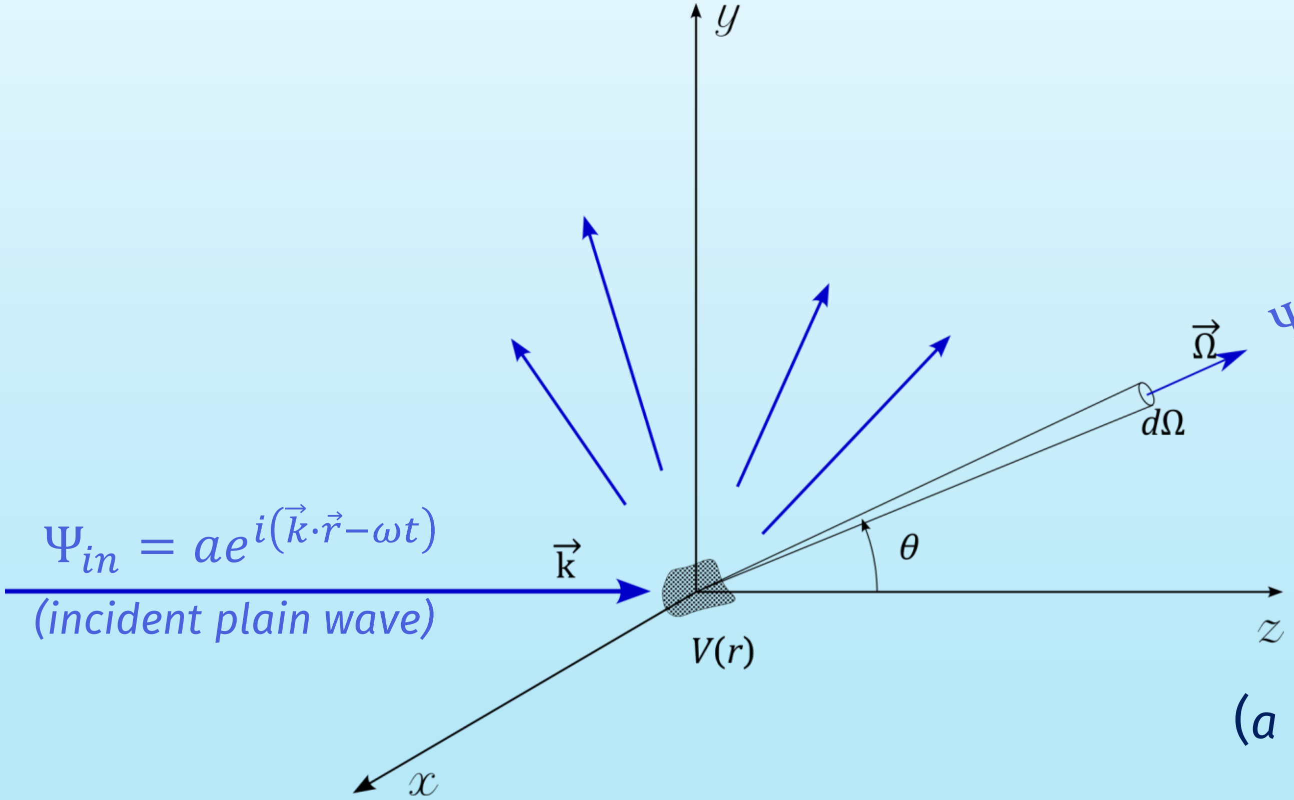
Schematic drawing of an incident beam striking a thin target with a particle emitted into a cone subtending an angle θ relative to the direction of incidence, the 'scattering' angle. The element of solid angle $d\Omega$ is a small piece of the cone.

The angular differential cross section $\frac{d\sigma}{d\Omega}$ can be seen as the number of reactions per second in an angular cone subtended at angle θ with respect to the direction of incidence.

cf. Sidney Yip, lecture 7 (10/2/06)

Elementary scattering theory

Theoretical treatment of neutron scattering usually starts with the quantum mechanical description of a two-body collision.



$$\Psi_{sc} = f(\theta)a\frac{e^{i(kr-\omega t)}}{r}$$

(scattered spherical wave)

(a is determined by normalization condition.)

$f(\theta)$ denotes the amplitude of scattering in the direction of angle θ relative to the direction of incidence.

Elementary scattering theory

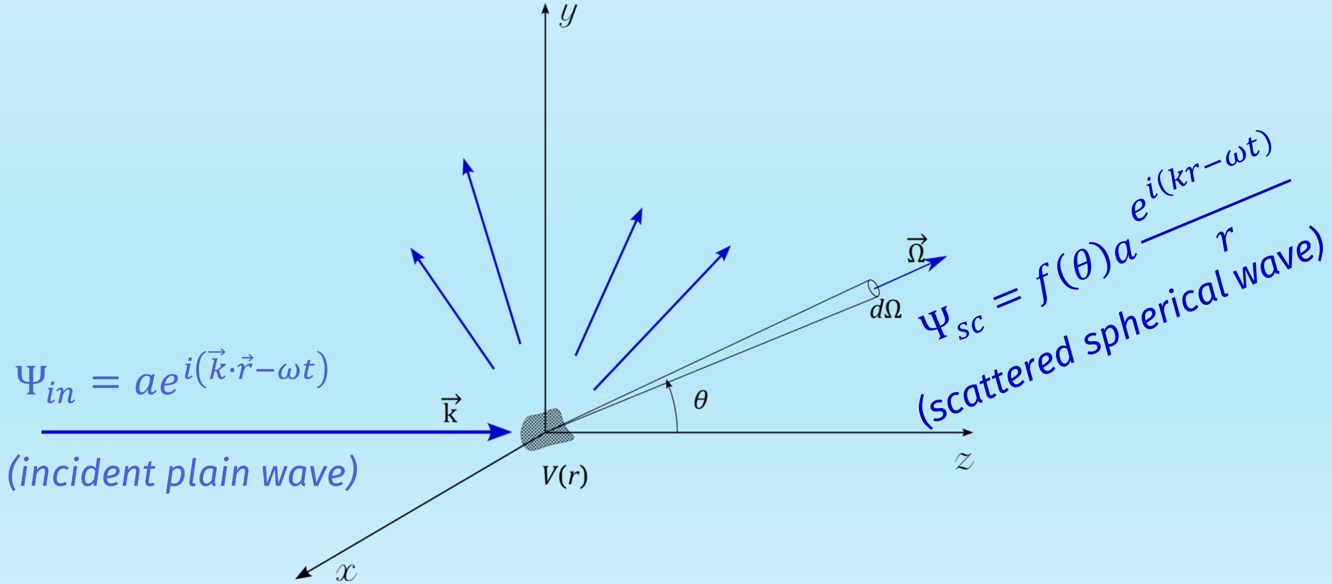
Quantum mechanical treatment of the potential scattering problem is usually approached in two steps:

1. to define the cross section $\sigma(\theta)$ in terms of the scattering amplitude $f(\theta)$
2. to calculate this quantity by solving the Schrödinger equation.

In the centre-of-mass coordinate system (CMS) the problem is to describe the motion of an effective particle with the reduced mass $\mu = \frac{m_1 m_2}{m_1+m_2}$, moving in a central potential $V(r)$, r being the separation between the two colliding particles. The incident beam is represented by a travelling plane wave, and the scattered wave, which results from the interaction in the region of the centrosymmetric potential, takes the form of an outgoing spherical wave:

$$\Psi_{in} = ae^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} \qquad \Psi_{sc} = af(\theta)\frac{e^{i(kr-\omega t)}}{r}$$

$f(\theta)$ *is* the scattering amplitude in the direction of angle θ relative to the direction of incidence.



ad 1. (to define the cross section $\sigma(\theta)$ in terms of the scattering amplitude $f(\theta)$)

We have heuristically defined the cross section as:

The angular differential cross section $\frac{d\sigma}{d\Omega}$ can be seen as the number of reactions per second in an angular cone subtended at angle θ with respect to the direction of incidence.

More precisely:

$$\frac{d\sigma}{d\Omega} = \frac{\{current\ density\ of\ scattered\ neutrons\ into\ (\boldsymbol{\Omega}, d\Omega)\}}{\{current\ density\ of\ incident\ neutrons\}}$$

It can be shown, that:

$$\boxed{\frac{d\sigma(\theta)}{d\Omega} = |f(\theta)|^2}$$

This is the fundamental expression relating the scattering amplitude to the cross section.

ad 2. *(to calculate $f(\theta)$ by solving the Schrödinger equation)*

By the Green's function approach, the Schrödinger equation can be written in the form of an integral equation and then solved to give:

$$f(\theta) = -\frac{2\mu}{4\pi\hbar^2}\int d^3r\, e^{i\mathbf{Q}\cdot\mathbf{r}}\, V(\mathbf{r}) \qquad \mathbf{Q} = \mathbf{k}_0 - \mathbf{k}$$

In the Born approximation the scattering amplitude is given by the Fourier transform of the interaction potential. The Fourier transform variable **Q** is a wave vector, the difference between the wave vectors of the scattered and incident waves **k** and $\mathbf{k}_0$, **Q** is referred to as the ***wave vector transfer*** or the ***scattering vector***, not to be confused with the momentum transfer. The two differ by the multiplication factor 2π.

However, strictly speaking, the condition for the validity of the Born approximation with the "true" interaction potential is not fulfilled in the case of thermal neutron scattering.

Elementary scattering theory

This was first noticed and solved by E. Fermi. The Fermi's idea involves the introduction of a pseudopotential in place of the actual neutron-nucleus interaction. Fermi suggested to distort the neutron-nucleus interaction by extending the range and decreasing the depth in such a way that the transformation preserves the scattering length.

The fictitious Fermi potential $V^*(r)$, or pseudopotential, has the well depth and range scaled so that $V^*(r)$ is a spherical well with depth V_0^* and range r_0^*.

$$V^*(r) = \frac{2\pi\hbar^2}{m} b\delta(r)$$

b, being in general a complex nuber $b = b' + ib''$, is known as the bound **scattering length**, "bound" because it refers to the fixed nucleus.

This formula has an important advantage:

the correct scattering cross section is already built into the potential.

In the low-energy limit the scattering (s-wave) cross section and the absorption cross section far form resonant capture are given by:

$$\sigma_s = 4\pi|b|^2 \qquad \sigma_a = \frac{4\pi}{k_0} b''$$

Coherent / incoherent b's and σ's

The contents of the columns are as follows:

Column	Unit	Quantity
1	---	Isotope
2	---	Natural abundance (For radioisotopes the half-life is given instead)
3	fm	bound coherent scattering length
4	fm	bound incoherent scattering length
5	barn	bound coherent scattering cross section
6	barn	bound incoherent scattering cross section
7	barn	total bound scattering cross section
8	barn	absorption cross section for 2200 m/s neutrons

Note: 1fm=1E-15 m, 1barn=1E-24 cm^2, scattering lengths and cross sections in parenthesis are uncertainties.

Neutron scattering lengths and cross sections							
Isotope	**conc**	**Coh b**	**Inc b**	**Coh xs**	**Inc xs**	**Scatt xs**	**Abs xs**
H	---	-3.7390	---	1.7568	80.26	82.02	0.3326
1H	99.985	-3.7406	25.274	1.7583	80.27	82.03	0.3326
2H	0.015	6.671	4.04	5.592	2.05	7.64	0.000519
3H	(12.32 a)	4.792	-1.04	2.89	0.14	3.03	0

When you look up the tables you see something like this.

Please note the coherent and incoherent scattering lengths and cross sections.

Coherent / incoherent b's and σ's

Still within the Born approximation: If the target nucleus has a non-zero spin then the neutron and nuclear spins could be either parallel or antiparallel during the scattering process. The neutron, being a fermion with spin ½, couples to the nuclear spin I to give :

Spin operators	**Eigenvalues**	**Number of degenerate states**	**Relative weights**
$\vec{\mathrm{I}} + \frac{\vec{1}}{2}$	$I + {}^{1}\!/_{2}$	$2\left(I + {}^{1}\!/_{2}\right) + 1 = 2I + 2$	$W_{+} = \frac{2I+2}{4I+2} = \frac{I+1}{2I+1}$
	$I - {}^{1}\!/_{2}$	$2\left(I - {}^{1}\!/_{2}\right) + 1 = 2I$	$W_{-} = \frac{2I}{4I+2} = \frac{I}{2I+1}$

- $2(I+1)$ degenerate states for the eigenvalue of $I + \frac{1}{2}$, corresponding to parallel arrangement of the neutron and nucleus spins, with the scattering length conventionally denoted by b_{+}.
- $2I$ degenerate states for the eigenvalue of $I - \frac{1}{2}$, corresponding to antiparallel arrangement of both spins. The related scattering length is denoted by b_{-}.

There are therefore altogether $2(2I+1)$ states with relative weights W_{+} and W_{-} (Breit-Wigner formalism).

Coherent / incoherent b's and σ's

Averaging over spins:

$$\bar{b} = W_+ b_+ + W_- b_- = \frac{(I+1)b_+ + Ib_-}{2I+1}$$

$$\overline{b^2} = W_+ b_+^2 + W_- b_-^2 = \frac{(I+1)b_+^2 + Ib_-^2}{2I+1}$$

Coherent scattering length: $b_{coh} = \bar{b} = W_+ b_+ + W_- b_-$ ← Average

Incoherent scattering length: $b_{inc} = \sqrt{\overline{b^2} - \bar{b}^2} = \sqrt{W_+ W_-}(b_- - b_+)$ ← SQRT(variance)

Coherent scattering length is the weighted average of b_+ and b_- , while incoherent scattering length is given by the variance of b_+ and b_-:

Zero-spin nuclei have $b_{inc} = 0$, hence in an experiment they show no incoherent scattering
(other sources of incoherence are still possible).

Coherent / incoherent b's and σ's

The corresponding cross sections are:

$$\sigma_{coh} = 4\pi b_{coh}^2 \qquad \sigma_{inc} = 4\pi b_{inc}^2$$

Coherent scattering is Q-dependent and therefore it contains structural information.

For hydrogen, the following numerical values have been measured:

	b_+	b_-	b_{coh}	b_{inc}	σ_{coh}	σ_{inc}
^1H	10.817(5)	-47.420(14)	-3.7406(11)	25.274(9)	1.7583(10)	80.27(6)
^2H	9.53(3)	0.975(60)	6.671(4)	4.04(3)	5.592(7)	2.05(3)
^3H	4.18(15)	6.56(37)	4.792(27)	-1.04(17)	2.89(3)	0.14(4)

V. Scattering on an ensemble of nuclei

Scattering on an ensemble of nuclei

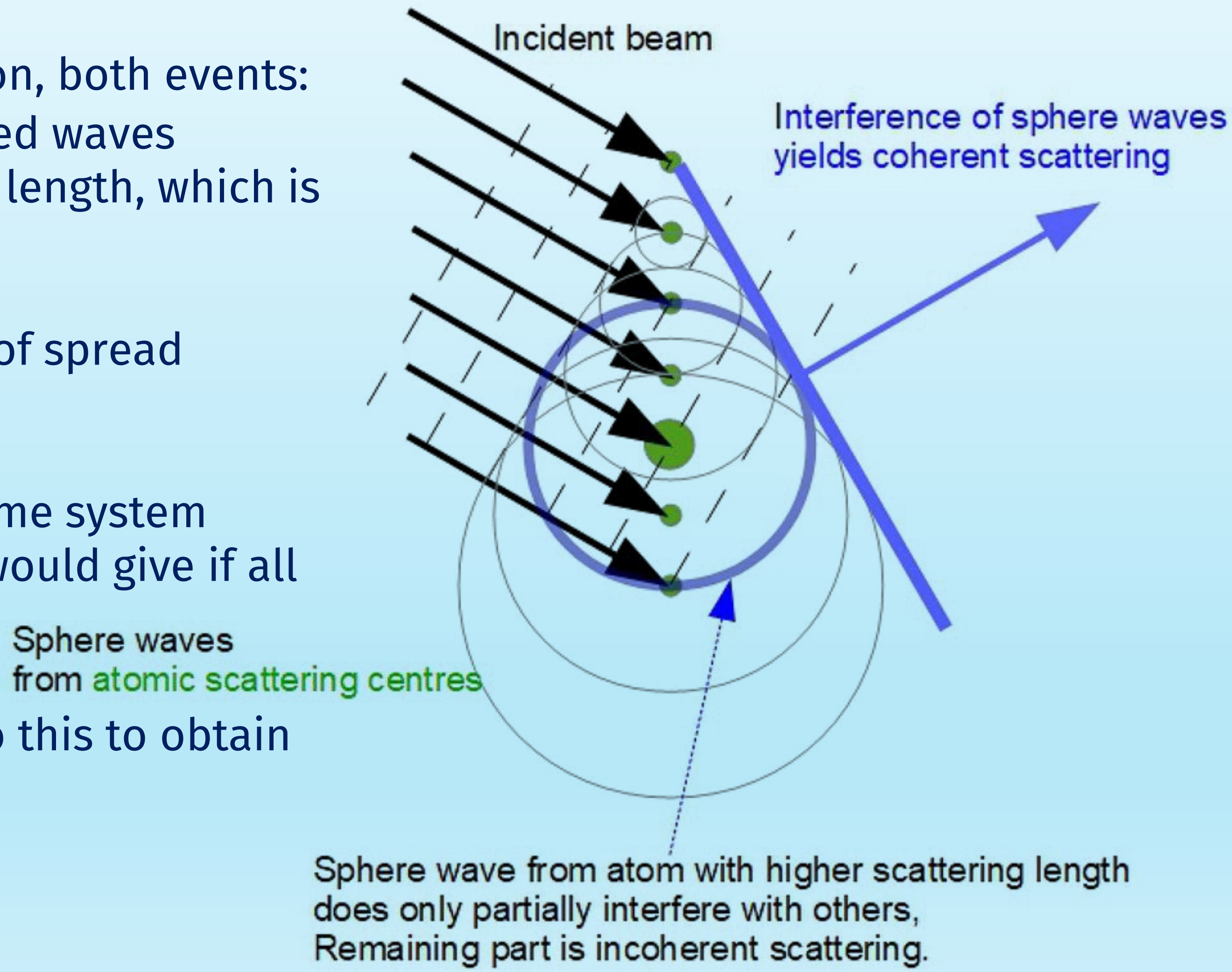
Imagine a sample cmposed of atoms only of one isotope. This was implicitly assumed in the previous considerations of b_+ and b_-, etc.

Since for unoriented, unploarized sample and neutron, both events: $\uparrow\uparrow$ and $\uparrow\downarrow$ are likely, any experiment with the scattered waves capable of interfering, will see an average scattering length, which is thus called coherent.

A variance, on the other hand, measures the degree of spread in the values of a variable.

The coherent scattering is the scattering such the same system (same nuclei with the same positions and motions) would give if all the scattering lengths were equal to $\bar{b}$.

The incoherent scattering is the term we must add to this to obtain the scattering due to the actual system.



Scattering on an ensemble of nuclei

Now, *for an* ensemble of nuclei, the Fermi pseudopotential becomes a sum:

$$V^*(r) = \sum_j b_j \delta\left(\mathbf{r} - \mathbf{r}_j\right)$$

Van Hove showed[1], that the so called **scattering law**, i.e. the number of neutrons scattered per incident neutron, can be written in terms of time-dependent correlation functions between the positions of pairs of atoms in the sample:

$$I(\mathbf{Q}, \omega) = \frac{1}{h}\frac{k}{k_0}\sum_{i,j} b_i b_j \int_{-\infty}^{\infty} \left\langle e^{-i\mathbf{Q}\cdot\mathbf{r}_i(0)} e^{i\mathbf{Q}\cdot\mathbf{r}_j(t)} \right\rangle e^{-i\omega t} dt$$

[1]Léon Van Hove, Phys. Rev 95, 1, (1954), 249–262 ; see also Squires book.

Scattering on an ensemble of nuclei

Let us define:

$$G(\mathbf{r},t) = \frac{1}{N}\sum_{i,j} \delta\left[\mathbf{r} - \left(\mathbf{r}_i(0) - \mathbf{r}_j(t)\right)\right]$$

N is the number of nuclei in the sample.

$G(\mathbf{r},t)$ is the probability that, within the sample, there will be a nucleus at the origin of our coordinate system at time zero *as well as* a nucleus at position $\mathbf{r}$ at time t. This is why $G(\mathbf{r},t)$ is called the **time-dependent pair correlation function.** It describes how the correlation between the positions of nuclei evolves with time.

Scattering on an ensemble of nuclei

Van Hove's scattering law can now be expressed in terms of $G(\mathbf{r}, t)$:

$$I(\mathbf{Q}, \omega) = \frac{Nb^2}{h} \frac{k}{k_0} \int_{-\infty}^{\infty} dt \int_{\substack{sam\\ple}} d^3r\, G(\mathbf{r}, t) e^{-i\mathbf{Q}\cdot\mathbf{r}} e^{-i\frac{\omega}{\hbar}t}$$

The scattering law is proportional to the space and time Fourier transform of the time-dependent correlation function.

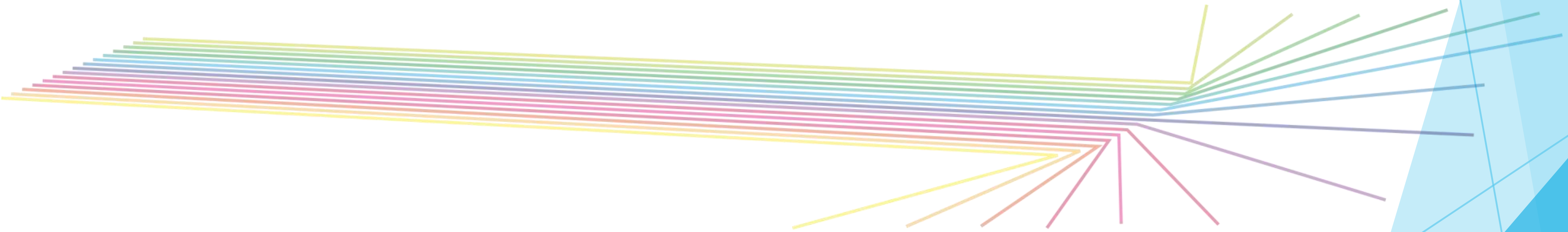
This general result provides a unified description of all neutron scattering experiments.

Neutron scattering can thus provide information about both the equilibrium structure of matter and the way in which this structure evolves with time.

Even if our sample is made up of a single isotope, all of the scattering lengths involved in the above considerations will not be equal, as neutron scattering depends on the spin state of the nucleus, and most isotopes have several nuclear spin states.

There is no correlation between the spin of a nucleus and its position in the sample. This is why our scattering lengths can be averaged without interfering with the thermodynamics.

VI. Coherent and incoherent scattering



Coherent and incoherent scattering

For simplicity, let us denote by $A_{i,j}$ the integral in the Van Hove formula:

$$I(\mathbf{Q},\omega) = \frac{1}{h}\frac{k}{k_0}\sum_{i,j} b_i b_j \int_{-\infty}^{\infty} \langle e^{-i\mathbf{Q}\cdot\mathbf{r}_i(0)} e^{i\mathbf{Q}\cdot\mathbf{r}_j(t)} \rangle e^{-i\omega t} dt \qquad A_{i,j} = \int_{-\infty}^{\infty} \langle e^{-i\mathbf{Q}\cdot\mathbf{r}_i(0)} e^{i\mathbf{Q}\cdot\mathbf{r}_j(t)} \rangle e^{-i\omega t} dt$$

Then, after appropriate averaging we get (for a rigorous derivation see e.g. S.W. Lovesey, Theory of Neutron Scattering from Condensed Matter, Oxford University Press (1984)):

$$\boxed{\sum_{i,j} \overline{b_i b_j} A_{i,j} = \sum_{i,j} \overline{b_i}\,\overline{b_j} A_{i,j} + \sum_i \left(\overline{b_i^2} - \left(\overline{b_i}\right)^2 \right) A_{i,i}}$$

The first term is coherent scattering in which neutron waves scattered from different nuclei interfere with each other. This type of scattering depends on the distances between atoms (through the integral represented by $A_{i,j}$) and on the scattering vector $\mathbf{Q}$.

In the second term, representing incoherent scattering, the intensities scattered from each nucleus just add up independently.

Coherent and incoherent scattering

Coherent scattering

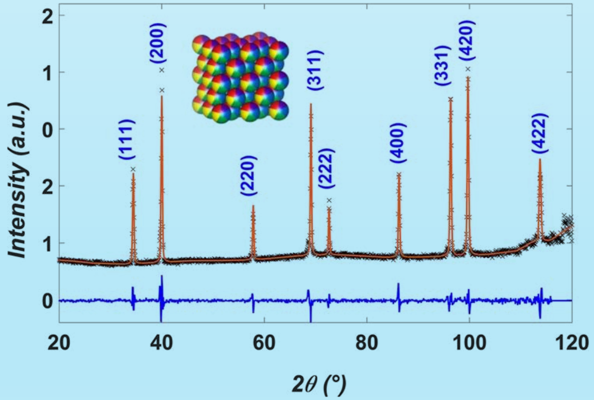
Incoherent scattering

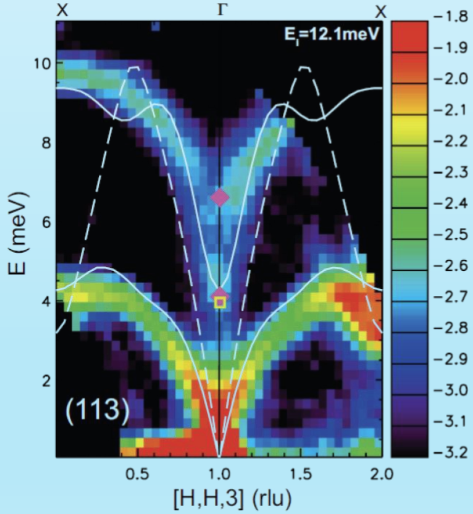
Elastic

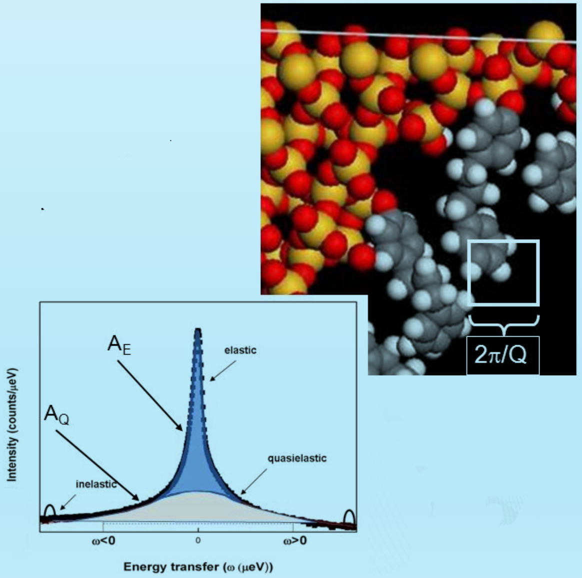
Inelastic

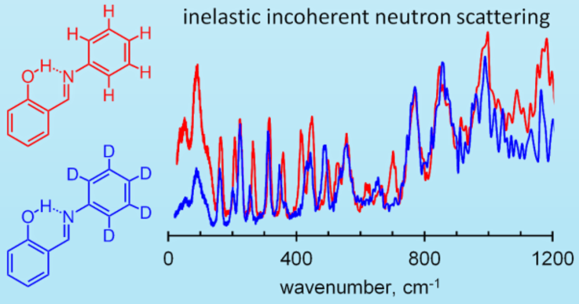
Elastic

Inelastic

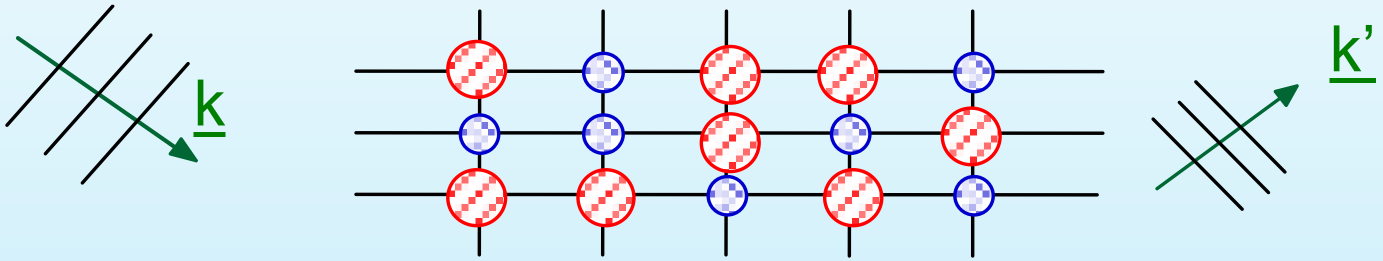




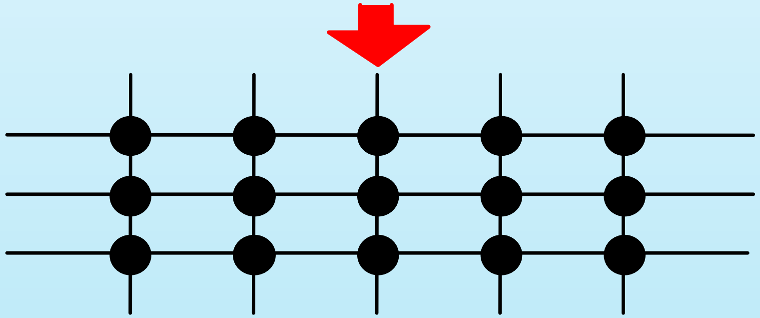




The isotope incoherence



Scattering from a regular lattice of chemically identical atoms but two isotopes with different scattering lengths.

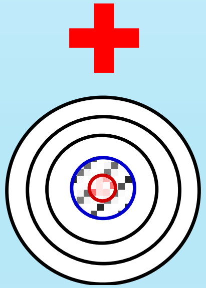


$$\langle b\rangle^2 \left|\sum_i e^{i\underline{Q}\cdot\underline{R}_i}\right|^2$$

Scattering from the mean regular lattice ⇨ <u>diffraction</u>

$$N\left\langle\left(b-\langle b\rangle\right)^2\right\rangle$$

N x



Incoherent scattering proportional to the number N of atoms and to the mean quadratic deviation from the average scattering length. ⇨ <u>incoherent isotropic background</u>

Spin & isotope incoherence

Isotope	Abundance	Nucl. spin	Scattering length [fm]
^{58}Ni	68.27 %	0	14.4(1)
^{60}Ni	26.10 %	0	2.8(1)
^{61}Ni	1.13 %	3/2	7.60(6)
^{62}Ni	3.59 %	0	−8.7(2)
^{64}Ni	0.91 %	0	−0.37(7)
Ni			10.3(1)

The most prominent example for isotope incoherence is **nickel**.

Total spin	Scattering length [fm]	Abundance
$J = 0$	$b_- = -47.50$	$\frac{1}{4}$
$J = 1$	$b_+ = 10.85$	$\frac{3}{4}$
	$\langle b \rangle = -3.7390(11)$	

The most prominent example for spin incoherence is **elementary hydrogen**.

$$\sigma_{coh} = 1.7568\ barn$$

$$\sigma_{\substack{nuclear \\ spin-incoherent}} = 4\pi\left[\frac{1}{4}(-47.5 + 3.7309)^2 + \frac{3}{4}(10.85 + 3.7309)^2\right] = 80.2\ barn$$

$$\sigma_{coh}^{D} = 5.592(7)\ barn \qquad \sigma_{inc}^{D} = 2.05(3)\ barn$$

Isotope incoherence

Another important element which shows strong nuclear incoherent scattering is vanadium.

Isotope	Abundance	Nucl. Spin	b_+ [fm]	b_- [fm]
^{50}V	0.25%	6	4.93 ±0.25	−7.58 ±0.28
^{51}V	99.75%	7/2	5.06 ±0.12 5.11 ±0.28	-7.60 ±0.12 −7.52 ±0.22

By chance, the characteristics of these isotopes (abundances, *b*'s, etc.) are such, that:

$$\sigma_{coh}^{V} = 0.01838(12)\ barn$$

$$\sigma_{inc}^{V} = 5.08(6)\ \ barn$$

For this reason, Bragg scattering of vanadium is difficult to observe above the large incoherent background.

On the other hand, this fact can be turned into an advantage:

By using vanadium metal, one can make sample containers which are practically invisible for coherent neutron scattering: They produce almost no reflections but rather a diffuse background (since incoherent scattering is isotropic) which usually doesn't cause severe problems in diffraction experiments.

Exercise:

Go to https://www.ncnr.nist.gov/resources/n-lengths/ and look up to find out about σ_{coh} and σ_{inc} for quartz (SiO_2)

VI. Separation of coherent and incoherent scattering

coh/incoh separation

Let I be the non-zero nuclear spin and let b_+ and b_- denote scattering lengths associated with the $I+\frac{1}{2}$ and $I-\frac{1}{2}$ compound states of the nucleus-neutron system. Let $\hat{B}$ denote the scattering length operator. Then let b_+ and b_- are its eigenvalues. It can be shown that:

$$\hat{B} = \bar{b} + \frac{1}{2} b_N \hat{\sigma} \cdot \hat{I}$$

Here $\hat{\sigma}$ is the Pauli spin operator and $\hat{I}$ is the nuclear spin operator. $\bar{b}$ and b_N are expressed in terms of b_+ and b_- through:

$$\bar{b} = \frac{(I+1)b_+ + Ib_-}{2I+1} \qquad b_N = \frac{2(b_+ - b_-)}{2I+1}$$

Further calculations are carried out by means of the density matrix formalism, and lead to the expression for the differential nuclear scattering cross-section:

coh/incoh separation

$$\frac{d\sigma}{d\Omega} = Tr\left[\rho\hat{B}^{\dagger}\left(\hat{\sigma}\cdot\hat{I}^{\dagger}\right)\hat{B}\left(\hat{\sigma}\cdot\hat{I}\right)\right]$$

Let $\mathbf{s}_n$ denote the spin of an individual ("*n*-th") neutron in the ensemble $\left(|\mathbf{s}_n| = \frac{1}{2}\right)$, and let's define the beam polarization **P** as the ensemble average over all the neutron spin vectors, normalised to their modulus:

$$\mathbf{P} = \frac{\langle \mathbf{s}_n \rangle}{\frac{1}{2}} = 2\langle \mathbf{s}_n \rangle$$

In the presence of an external field, providing a natural quantization axis, the beam polarization becomes a scalar value:

$$P = \frac{N_+ - N_-}{N_+ + N_-}$$

where N_+ and N_- are the number of neutrons with spin-up and spin-down with respect to the field direction. R is the so called flipping, a measurable quantity:

$$R = \frac{N_+}{N_-} \qquad \text{so} \qquad R = \frac{R-1}{R+1}$$

coh/incoh separation

Now the differential cross section (2.21) can also be expressed as a function of incoming beam polarization :

$$\frac{d\sigma}{d\Omega} = \hat{B}^\dagger \hat{B}\left[I(I+1) + i\mathbf{P}^{(in)} \cdot \left(\hat{I}^\dagger \times \hat{I}\right)\right]$$

If nuclear spins are unpolarized, which is normally the case, averaging over nuclear spin orientations must be performed. Then, after taking into account the properties of the nuclear spin operator and $\hat{I}$, one obtains for the differential cross section :

$$\frac{d\sigma}{d\Omega} = I(I+1)\hat{B}^\dagger \hat{B}$$

and for the scattered polarization:

$$P_\gamma^{(out)} \frac{d\sigma}{d\Omega} = Tr\left[\rho \hat{B}^\dagger \left(\hat{\sigma} \cdot \hat{I}^\dagger\right) \gamma \left(\hat{\sigma} \cdot \hat{I}\right) \hat{B}\right]$$

where γ denotes a particular direction in space. Again, evaluation of the right-hand followed by averaging over nuclear spin orientation, gives:

$$P_\gamma^{(out)} \frac{d\sigma}{d\Omega} = -\frac{1}{3} P^{(in)}\, I(I+1)\hat{B}^\dagger \hat{B}$$

coh/incoh separation

Let us introduce the conventional notation $I^{\uparrow\downarrow}$ and $I^{\uparrow\uparrow}$ for the scattered intensity with and without the spin flip. Then:

$$\frac{d\sigma}{d\Omega} = I^{\uparrow\uparrow} + I^{\uparrow\downarrow} = I_\gamma^{\uparrow\uparrow} + I_\gamma^{\uparrow\downarrow}$$

and:

$$P_\gamma^{(out)} \frac{d\sigma}{d\Omega} = \frac{I^{\uparrow\uparrow} - I^{\uparrow\downarrow}}{I^{\uparrow\uparrow} + I^{\uparrow\downarrow}} \left(I^{\uparrow\uparrow} + I^{\uparrow\downarrow}\right) = I^{\uparrow\uparrow} - I^{\uparrow\downarrow}$$

Rewritting the previous result:

$$\frac{d\sigma}{d\Omega} = I^{\uparrow\uparrow} + I^{\uparrow\downarrow} = I(I+1)\hat{B}^\dagger \hat{B}$$

and:

$$P_\gamma^{(out)} \frac{d\sigma}{d\Omega} = \frac{I^{\uparrow\uparrow} - I^{\uparrow\downarrow}}{I^{\uparrow\uparrow} + I^{\uparrow\downarrow}} \left(I^{\uparrow\uparrow} + I^{\uparrow\downarrow}\right) = I^{\uparrow\uparrow} - I^{\uparrow\downarrow} = -\frac{1}{3}\, I(I+1)\hat{B}^\dagger \hat{B}$$

coh/incoh separation

Finally:

$$\boxed{I^{\uparrow\uparrow} = \frac{1}{3}|B|^2 I(I+1)} \quad \text{and} \quad \boxed{I^{\uparrow\downarrow} = \frac{2}{3}|B|^2 I(I+1)}$$

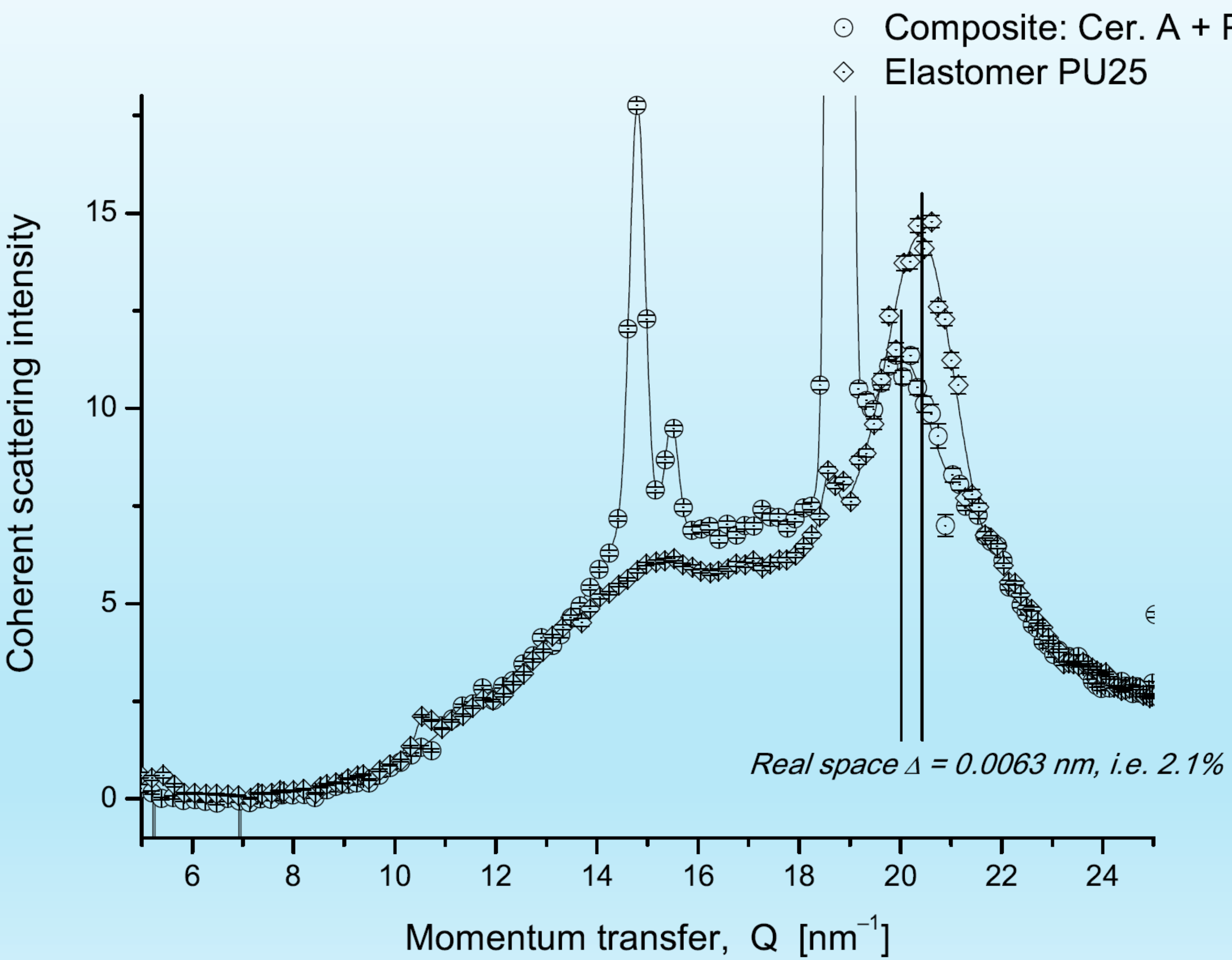
Thus the outgoing scattering intensity from unpolarized nuclear spins is always one third without, and two thirds with flip of the neutron spin from the polarized beam.

The coherent scattering Icoh and incoherent scattering Iinc are linearly related to the measured spin-flip and non-spin-flip scattering.

$$\boxed{I_{coh} = I^{\uparrow\uparrow} - \frac{1}{2} I^{\uparrow\downarrow}} \quad \text{and} \quad \boxed{I_{inc} = \frac{3}{2} I^{\uparrow\downarrow}}$$

In this way, shining a polarized neutron beam upon the sample and separately recording neutrons scattered with their spin conserved and with spin flipped, the experimenter can separate coherent from incoherent scattering right at the instrument level.

Example: residual strain in PU elastomer



W. Zając, A. Boczkowska, K. Babski, K. Kurzydłowski, P. Deen, *Measurements of residual strains in ceramic–elastomer composites with diffuse scattering of polarized neutrons*, Acta Materialia. 56 (2008) 5964-5971