EUROPEAN SPALLATION SOURCE

Neutron Sources

Christine Darve

African School of Fundamental Physics and its Applications

August 20, 2014

Outline

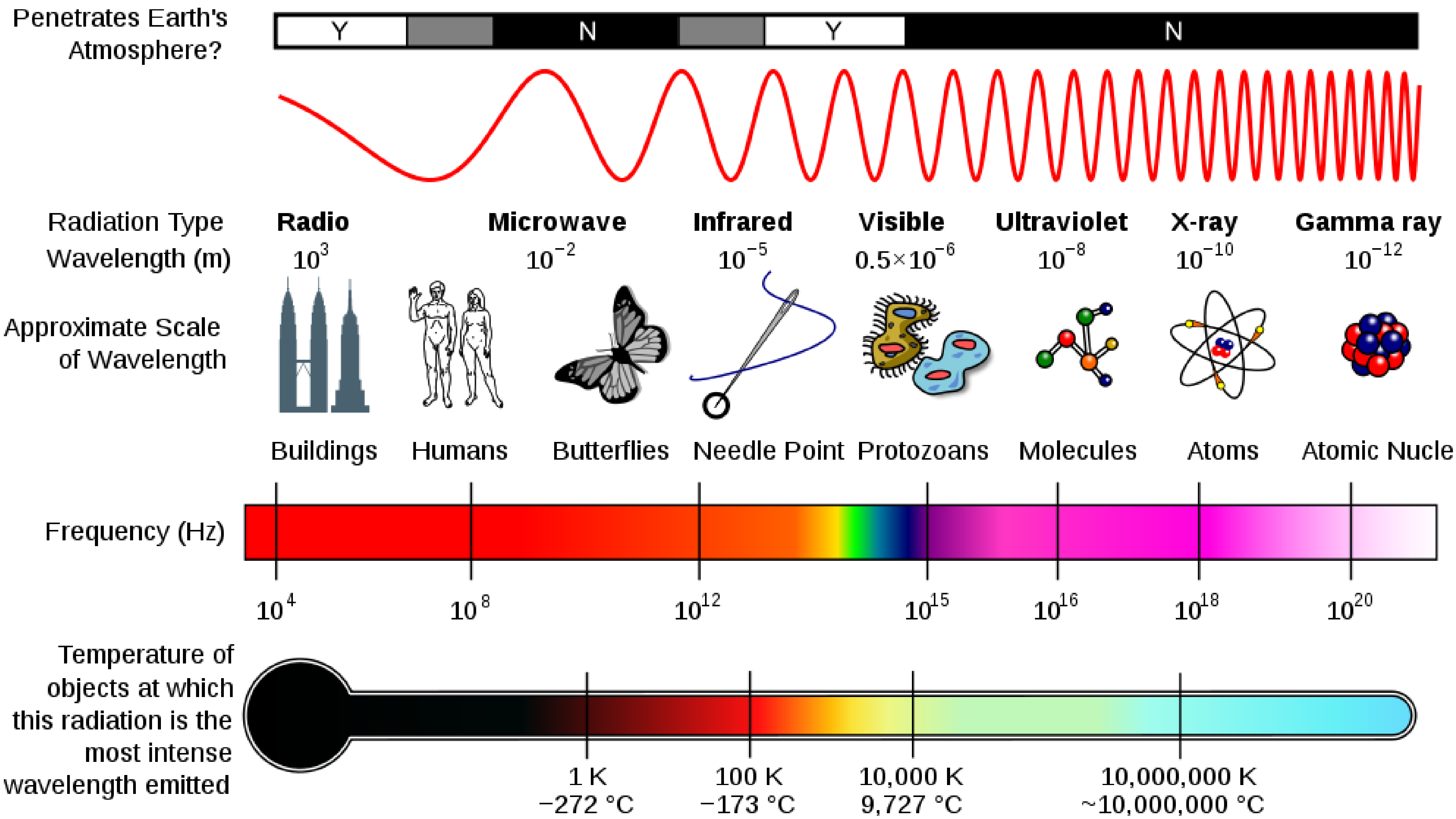
- Neutrons properties and their interactions

- How to generate intense neutron beams using high power proton linear accelerator: The example of the ESS

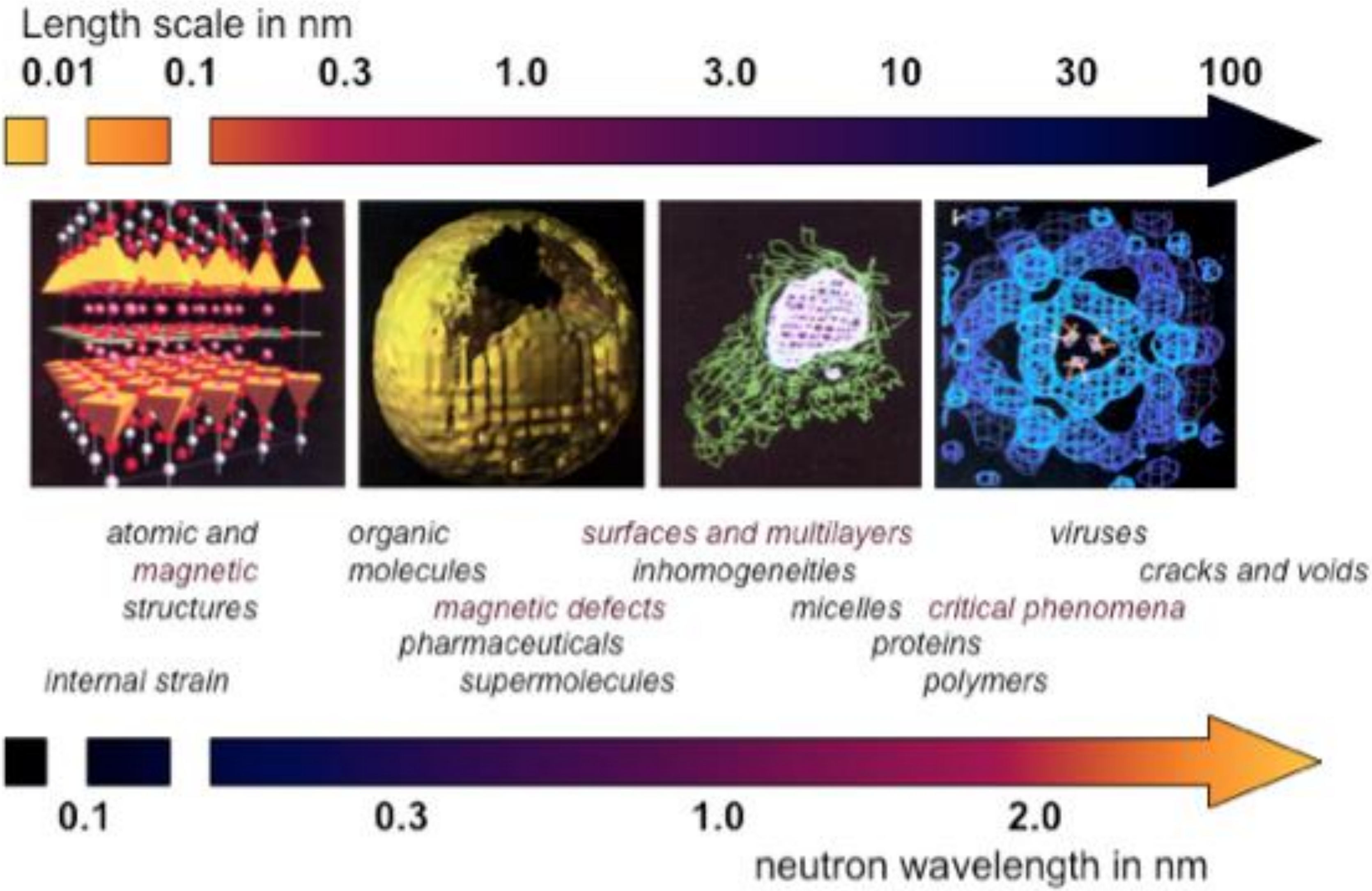
for further reading

- Applications using Neutrons

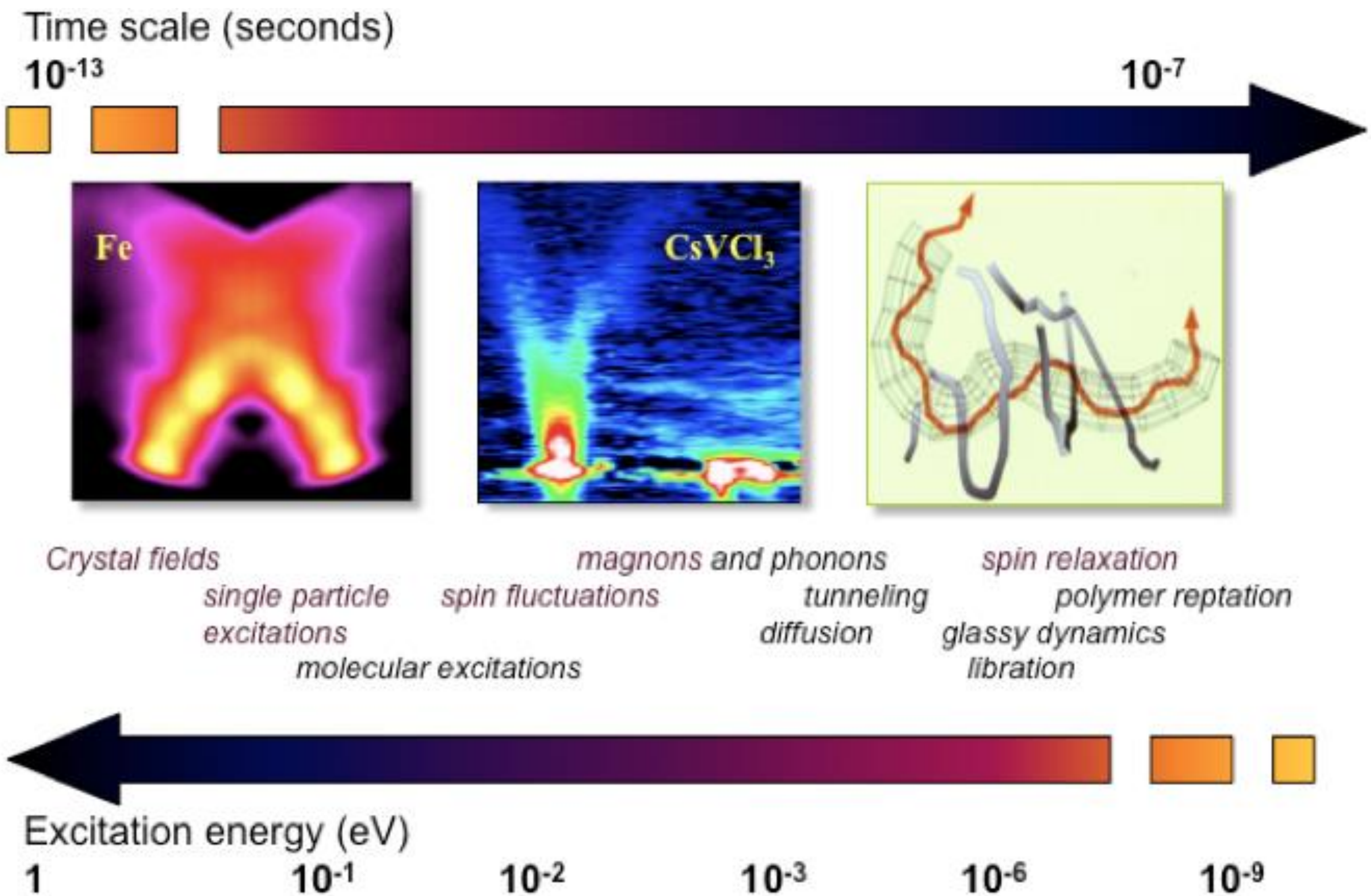
Electro-magnetic Spectrum



Neutron Microscope – Length scales

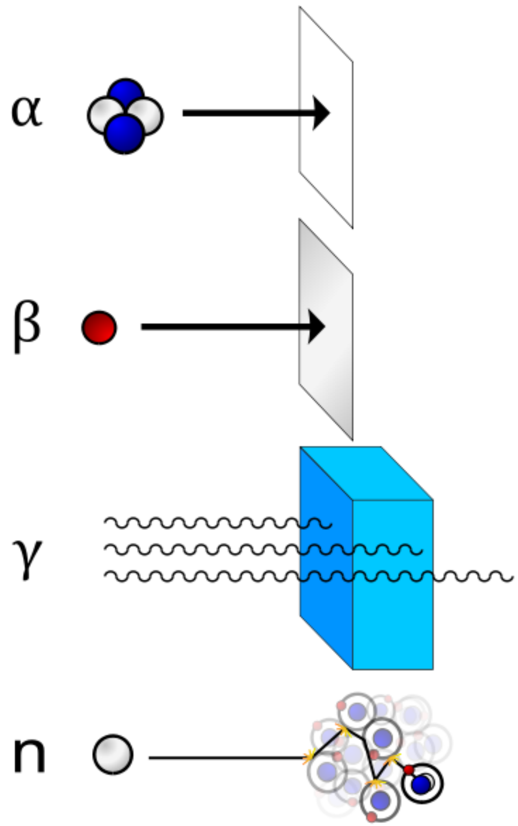


Neutron Microscope – Time & energy scales



Ionizing Radiation

Ionizing radiation is radiation composed of particles that individually carry enough energy to liberate an electron from an atom or molecule without raising the bulk material to ionization temperature.

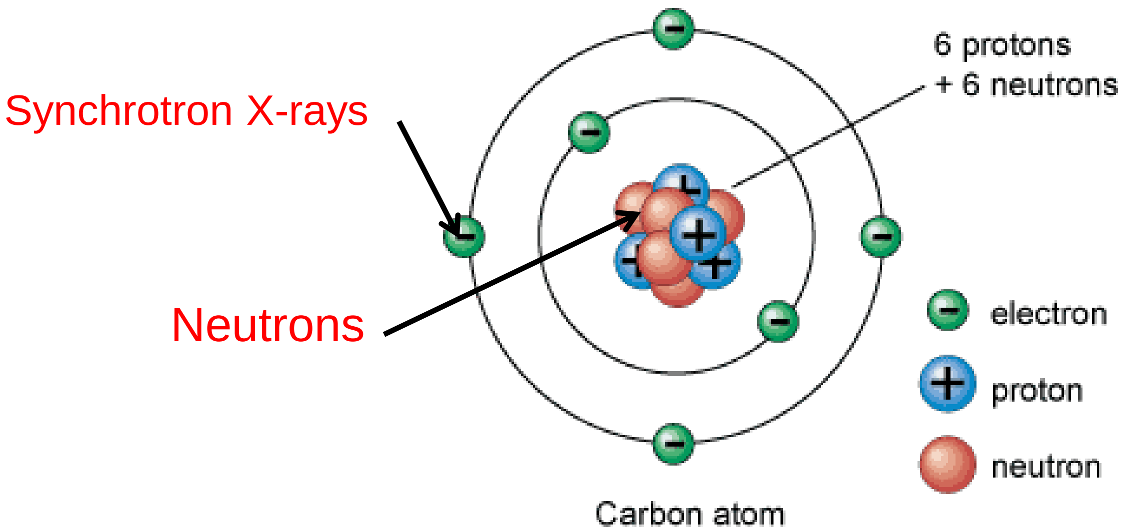


When ionizing radiation is emitted by or absorbed by an atom, it can liberate a particle. Such an event can alter chemical bonds and produce ions, usually in ion-pairs, that are especially chemically reactive.

Note: Neutrons, having zero electrical charge, do not interact electromagnetically with electrons, and so they cannot *directly* cause ionization by this mechanism.

➔ **High precision non-destructive probe … why ?**

Neutrons Properties



Note: X-rays are emitted by electrons outside the nucleus, while gamma rays are emitted by the nucleus.

1.675×10^{-27} kg Mass	939.57 MeV	$m_n \approx m_p + 2.5 m_e$
Mean lifetime	15 min	$n \longrightarrow p + e^- + \bar{\nu}_e$
Composition	udd	hadron
Electric charge	0	high penetration
Magnetic moment	$-1.04\,\mu_B$	feels the nucleus

Neutron Energy

$$E = k_B T$$

$$E = k_B T = \tfrac{1}{2} m v^2 = \frac{h}{2 m \lambda^2}$$

$$\lambda = \frac{h}{mv}$$

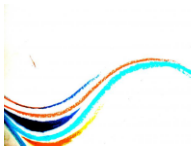
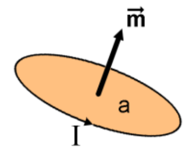
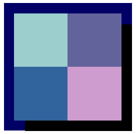
Boltzmann distribution

De Broglie

$$E\,[meV] = 0.0862\; T\,[K] = 5.22\; v^2\,[km/s] = 81.81 \frac{1}{\lambda^2}\,[A]$$

Source	Energy	Temperature	Wavelength
cold	0.1-10	1-120	30-3
thermal	5-100	60-1000	4-1
hot	100-500	1000-6000	1-0.4

Why neutrons?

Wave	Particle	Magnetic moment	Neutral
			

Neutron properties are used to understand the nature of the solid and liquid states of matter, as an analytical tool to aid the development of materials and as a tool to examine curiosity-driven research that spans from cosmology, superconductivity to the dynamics of the molecules of life.

1. Neutrons see the Nuclei
2. Neutrons see Elementary Magnets
3. Neutrons see light Atoms next to Heavy Ones
4. Neutrons measure the Velocity of Atoms
5. Neutrons penetrate deep into Matter
6. Neutrons are Elementary Particles

Why neutrons?

Electrically Neutral – neutrons are non-destructive and can penetrate deep into matter. This makes them an ideal probe for biological materials and samples under extreme conditions of pressure, temperature, magnetic field or within chemical reaction vessels.

Microscopically Magnetic – they possess a magnetic dipole moment which makes them sensitive to magnetic fields generated by unpaired electrons in materials. Precise information on the magnetic behavior of materials at atomic level can be collected. In addition, the scattering power of a neutron off an atomic nucleus depends on the orientation of the neutron and the spin of the atomic nuclei in a sample. This makes the neutron a powerful instrument for detecting the nuclear spin order.

Ångstrom wavelengths – neutron wavelengths range from 0.1 Å to 1000 Å, making them an ideal probe of atomic and molecular structures, be they single atomic species or complex biopolymers.

Why neutrons?

Energies of millielectronvolts – their energies are of the same magnitude as the diffusive motion in solids and liquids, the coherent waves in single crystals (phonons and magnons), and the vibrational modes in molecules. It is easy to detect any exchange of energy between a sample of between 1microeV (even 1 neV with spin-echo) and 1eV and an incoming neutron.

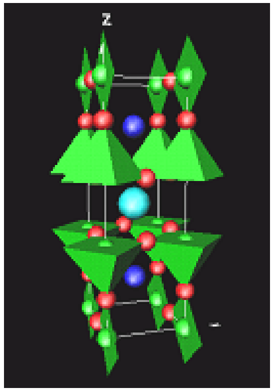
Randomly sensitive – with neutrons the variation in scattering power from one nucleus to another within a sample varies in a quasi-random manner. This means that lighter atoms are visible despite the presence of heavier atoms, and neighboring atoms may be distinguished from each other. In addition, contrast can be varied in certain samples using isotopic substitution (for example D for H, or one nickel isotope for another); specific structural features can thus be highlighted. The neutron is particularly sensitive to hydrogen atoms; it is therefore a powerful probe of hydrogen storage materials, organic molecular materials, and biomolecular samples or polymers.

Why neutrons?

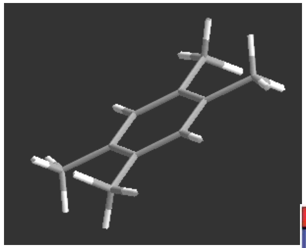
In half a century we have developed neutron scattering science enormously with an effective gain in source performance of only a factor of 4 !

- Diffractometers - Measure structures
  – Where atoms and molecules are

1 - 10 Ångström



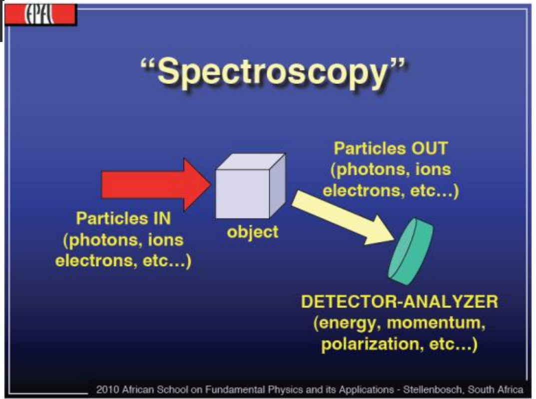
→ To analyze the structure of a material from the scattering pattern produced when a beam of radiation or particles (such as X-rays or neutrons) interacts with it



- Spectrometers - Measure dynamics
  – What atoms and molecules do

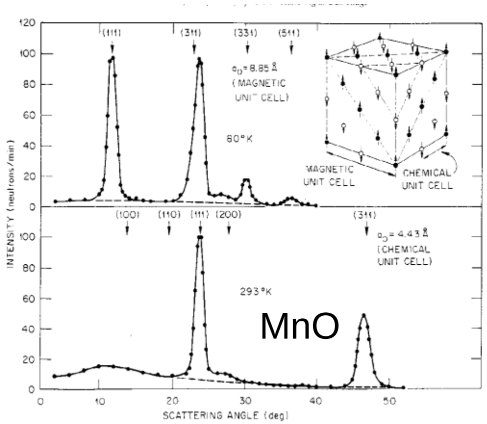
1 - 80 meV

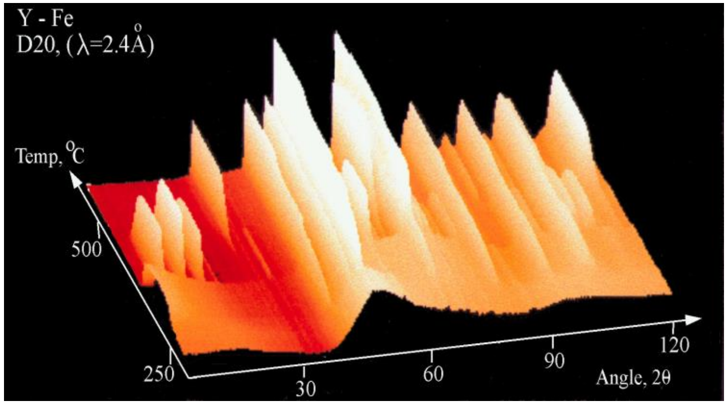
→ To measure properties of light over a specific portion of the electromagnetic spectrum



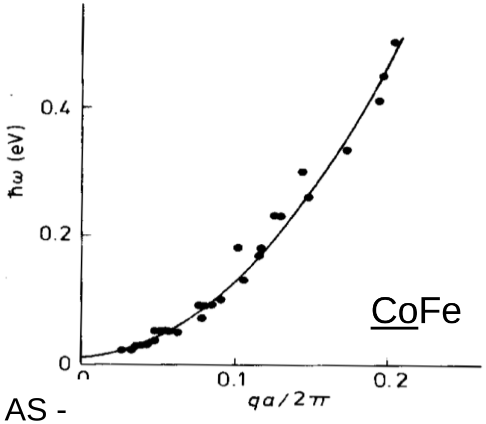
Why neutrons?

Cliff Shull – Neutron diffraction - showing where atoms are:

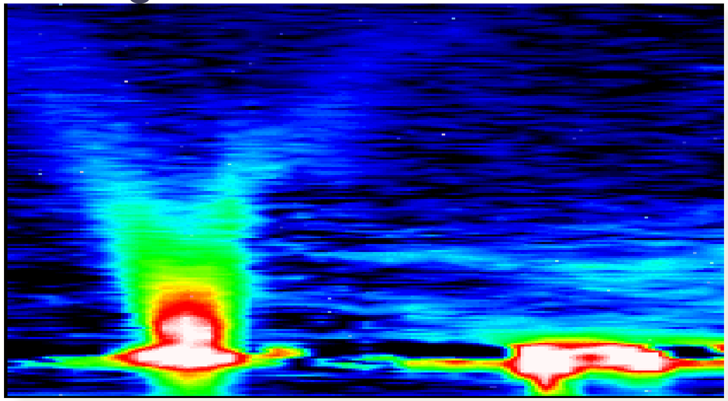




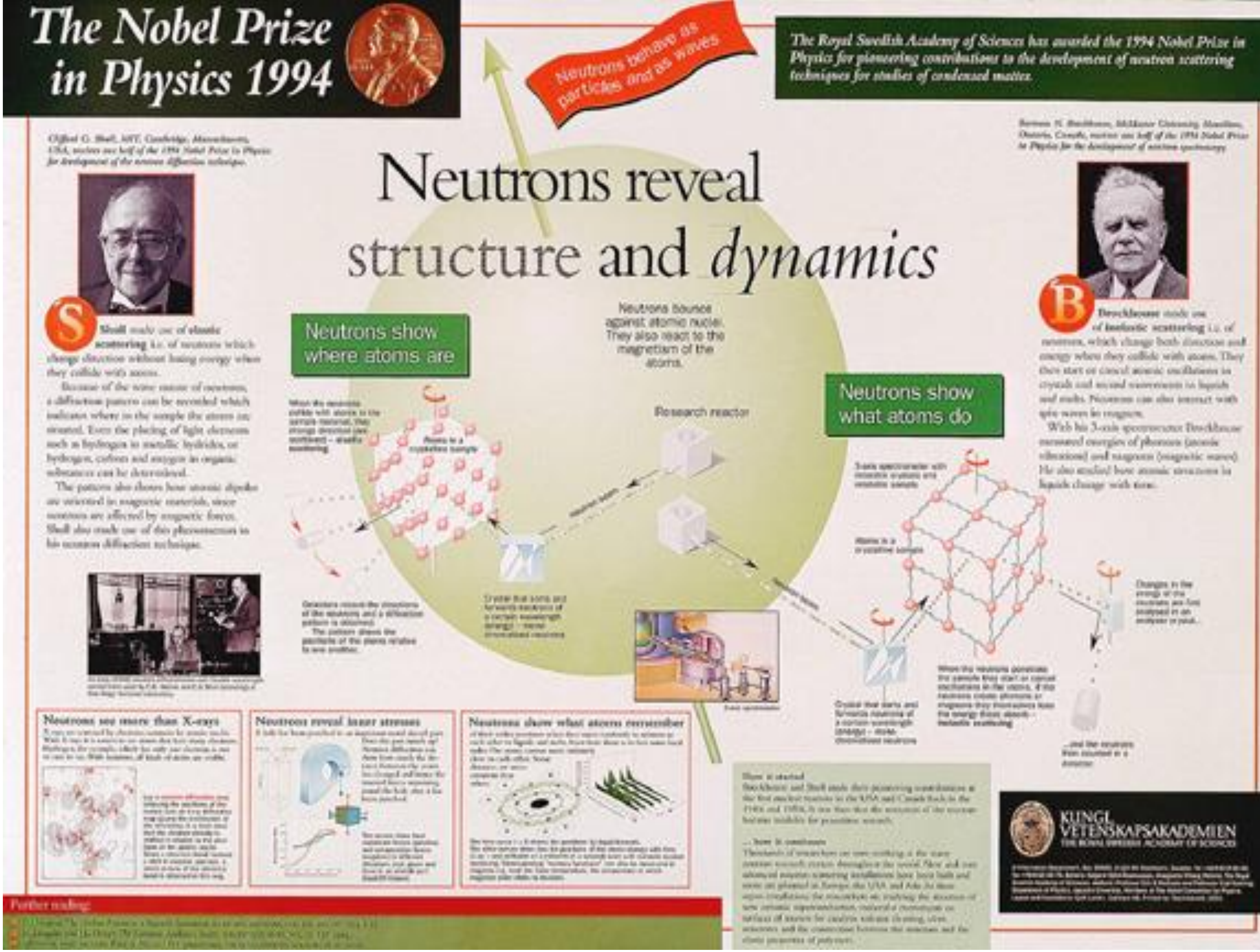
Bert Brockhouse – Spectroscopy - showing what atoms do:



AS -



Using nowadays technique..



Neutrons show us : where atoms are (left, Bragg scattering) and what they do (right, Spectroscopy).
Energies of neutrons are same scale as excitations. 1 Aneutron 6orders of magnitude less than 1 A Synch.

Scientific challenges

EUROPEAN SPALLATION SOURCE

Solid State Physics
Dynamics of superlattices, wires and dots, molecular magnets, quantum phase transitions

Liquids and Glasses
Solvent structures, influence of molecular structures on protein folding

Fundamental Physics
Left and right handedness of the universe, neutron decay, ultracold neutrons

Soft Condensed Matter
Time resolution, molecular rheology, structures and dynamics

Biology and Biotechnology
Hydrogen and water , membranes, biosensors, functions

Materials Science and Engineering
Real time investigations with realistic dimensions under real conditions

Chemical Structure, Kinetics and Dynamics
Thin films, pharmaceuticals, supramolecules - structures and functionality

Earth and Environmental Science, Cultural Heritage
Extreme temperatures and pressures simulating the mantle

Fields of interest

A wide range of length and timescales.

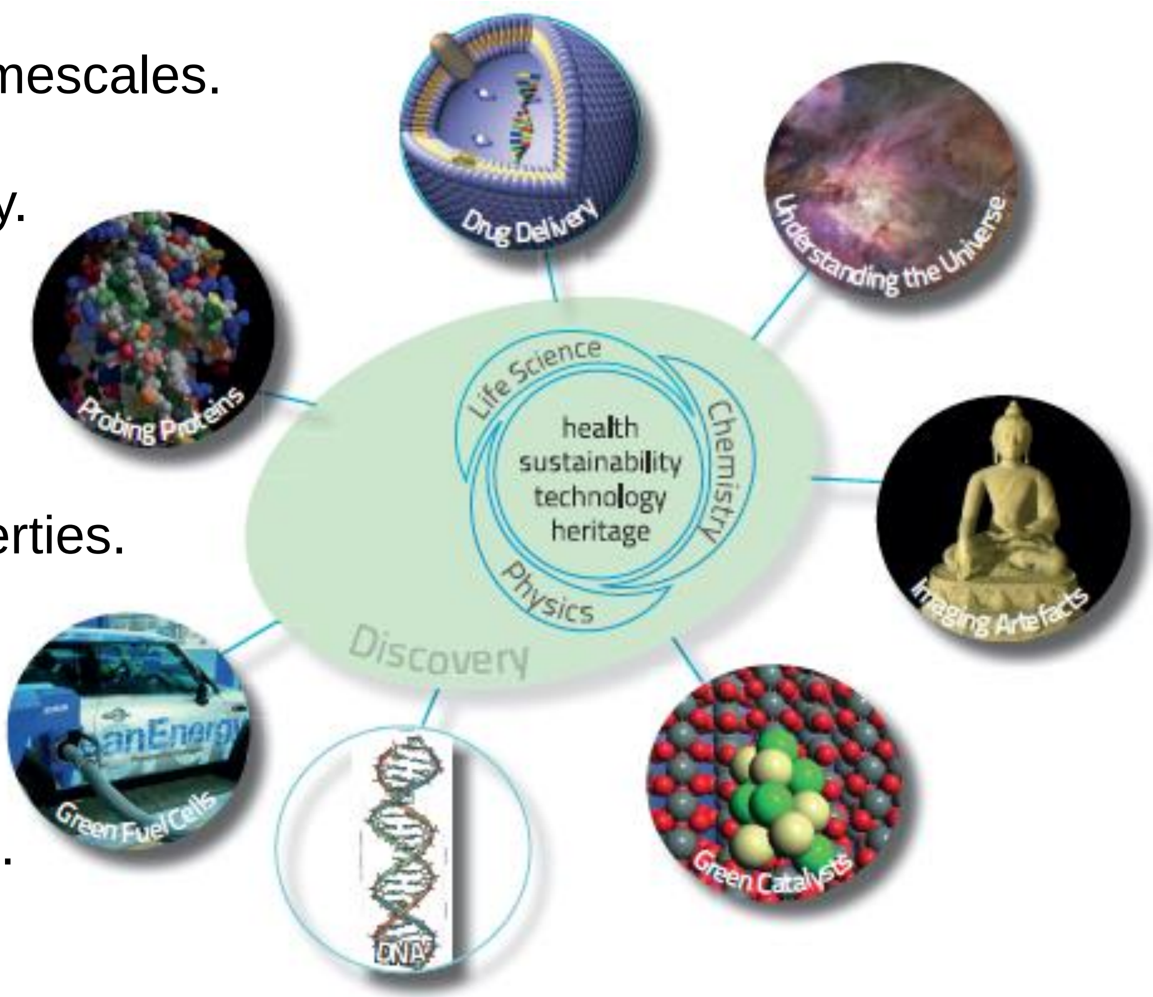
High sensitivity and selectivity.

Deep penetration.

A probe of fundamental properties.

A precise tool

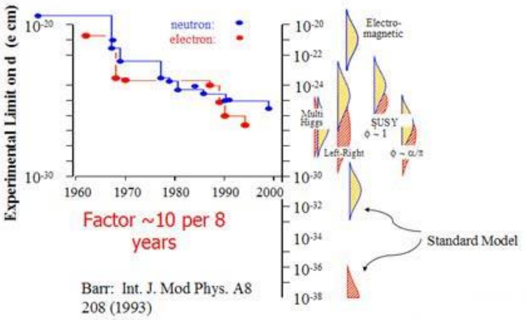
An ideal probe for magnetism.

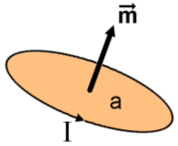


16

The visions

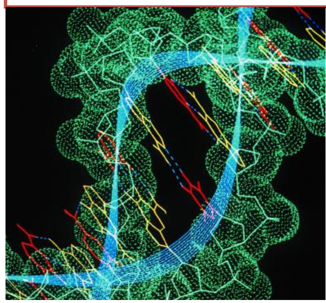
- Room Temperature Super Conductors
- Sterile neutrinos
- Hydrogen storage substrate
- Neutron electric dipole moment
- Efficient membrane for fuel cells
- Flexible and highly efficient solar cells
- Carbone nano-tubes for controlled drug release
- Self healing materials – smart materials
- Spin-state as a storage of data (10^{23} gain in capacity)
- CO_2 sequestration

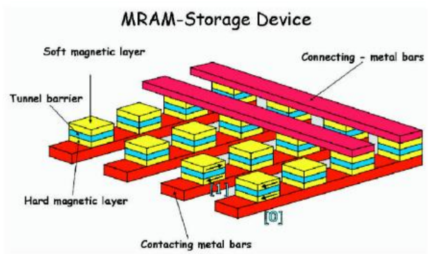


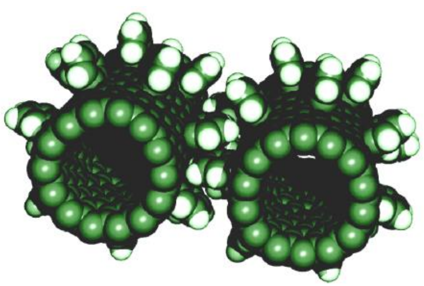


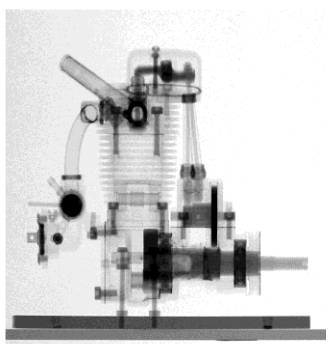
Multi-science with neutrons

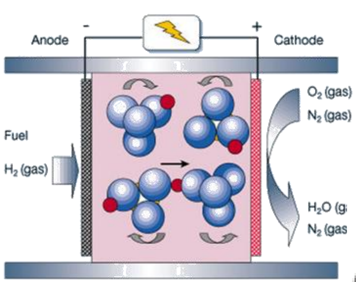
Materials science	Bio-technology	Nano science
Energy Technology	Hardware for IT	Engineering science











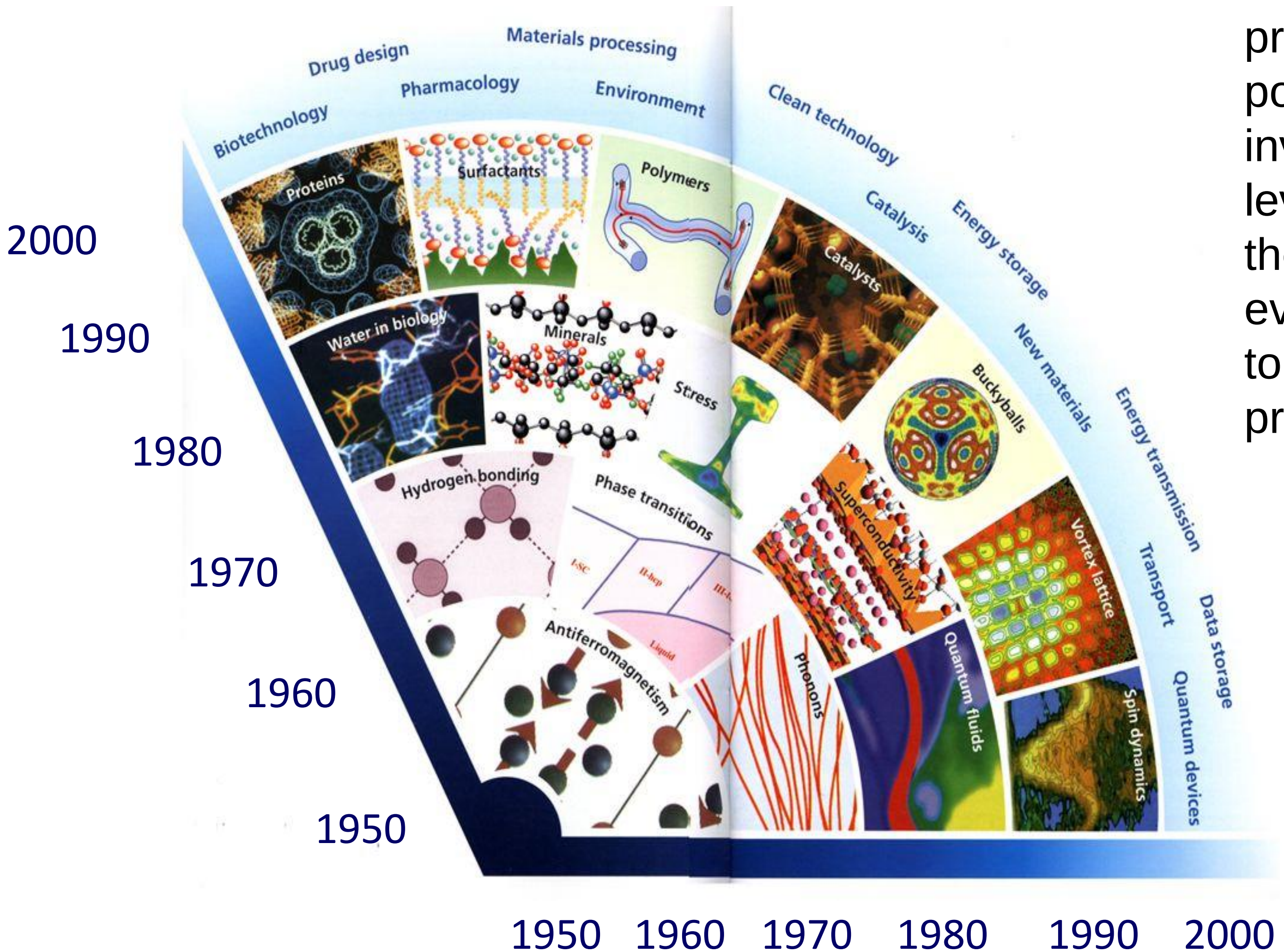
- Neutrons can provide unique and information on almost all materials.

- Information on both structure and dynamics simulaneously. ”Where are the atoms and what are they doing?”

- 6000 primary users in Europe today and 6000 secondary users. Access based on peer review.

- Science with neutrons is limited by the intensity of today’s sources

Neutron scattering - an expanding field

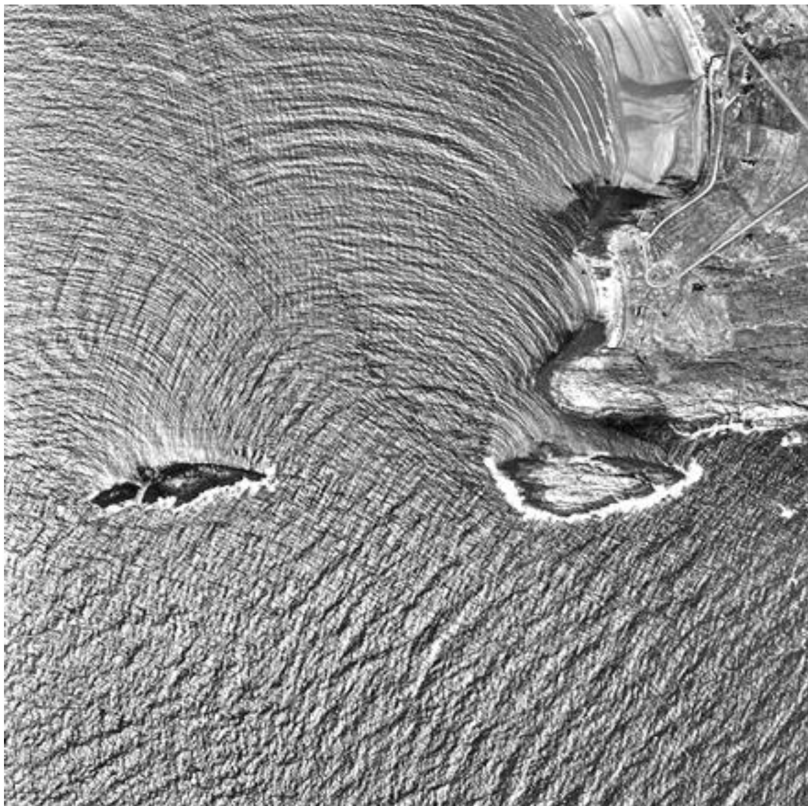


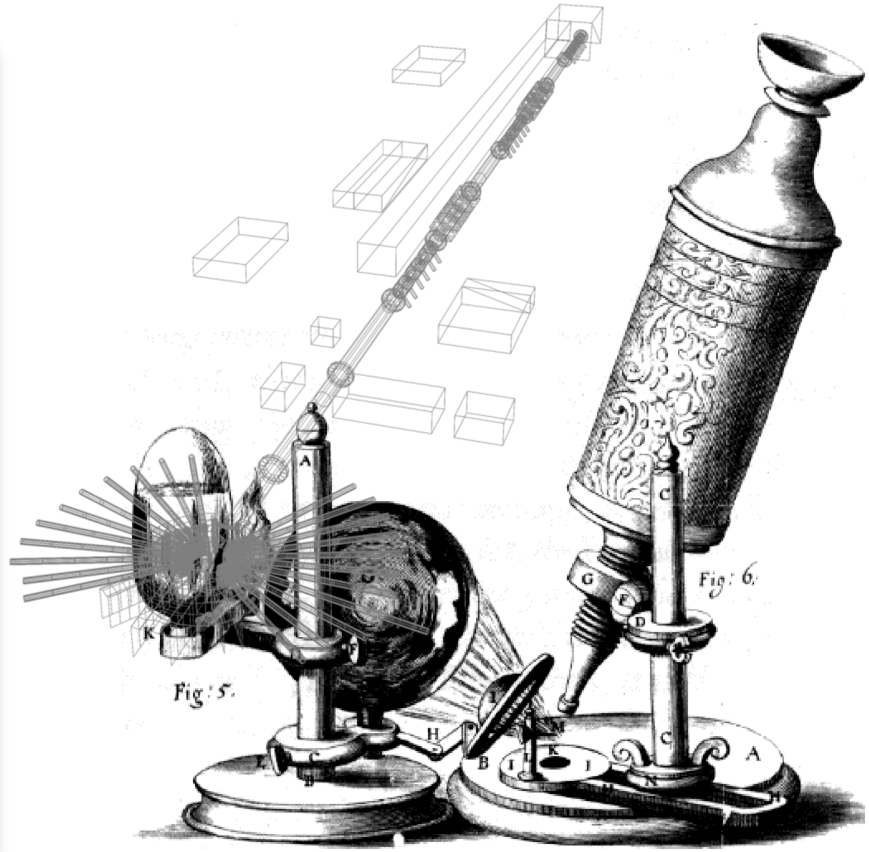
Because of their unique properties, neutrons are a powerful tool for investigating Nature at all levels, from testing theories about the evolution of the Universe to elucidating the complex processes of life.

X-Ray and Neutron beam

Complementarity between X-rays and Neutrons

EUROPEAN SPALLATION SOURCE





Consider ESS/MaxIV equal in terms of functionality

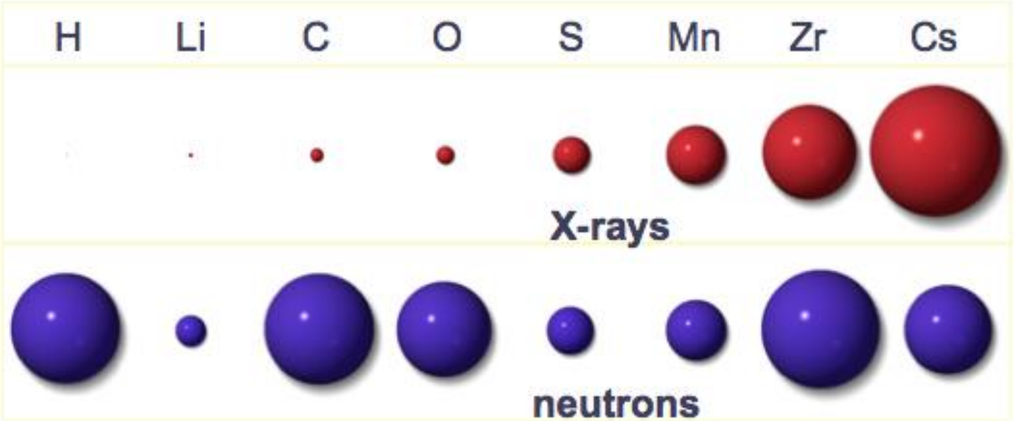
Complementarity between X-rays & Neutrons

Neutrons	Synchrotron radiation
• Particle beam (neutral subatomic particle) • Interactions with the nuclei and the magnetic moment of unpaired electrons (in the sample) • Scattered by all elements, also the light ones like the hydrogen isotopes • Deep penetration depth (bulk studies of samples) • Less intense beam measuring larger samples	• Light beam (electromagnetic wave) • Interactions with the electrons surrounding the nuclei (in the sample) • Mainly scattered by heavy elements • Small penetration depth (surface studies of samples) • Very intense beam measuring small or ultra-dilute samples

Complementarity between X-rays & Neutrons

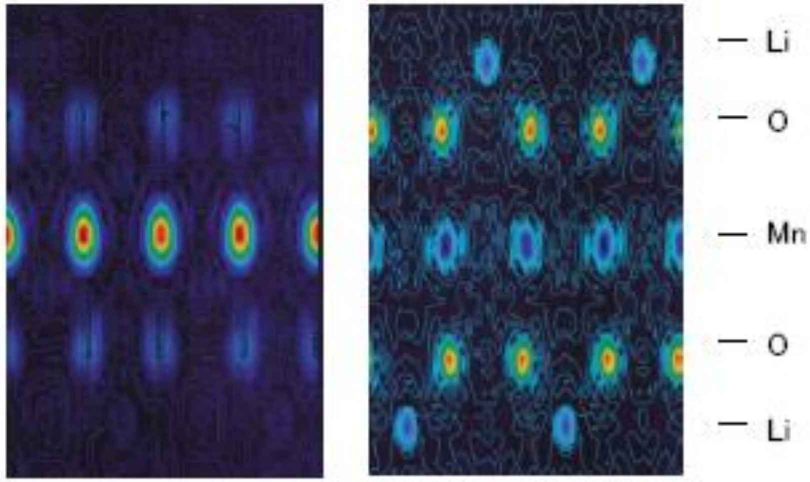
Neutrons Applications	SR Applications
• Magnetic structures & excitations • Organic structures using the H-D isotope effect • Bulk studies (strains, excitations) • Low-energy spectroscopy e.g. molecular vibrations	• Protein-crystal structures • Fast chemical reactions • Surface studies (defects, corrosion) • High-energy spectroscopy e.g. measurements of electron energy-levels

Complementarity between X-rays & Neutrons



X-ray interact with electrons
→ X-rays see high-Z atoms

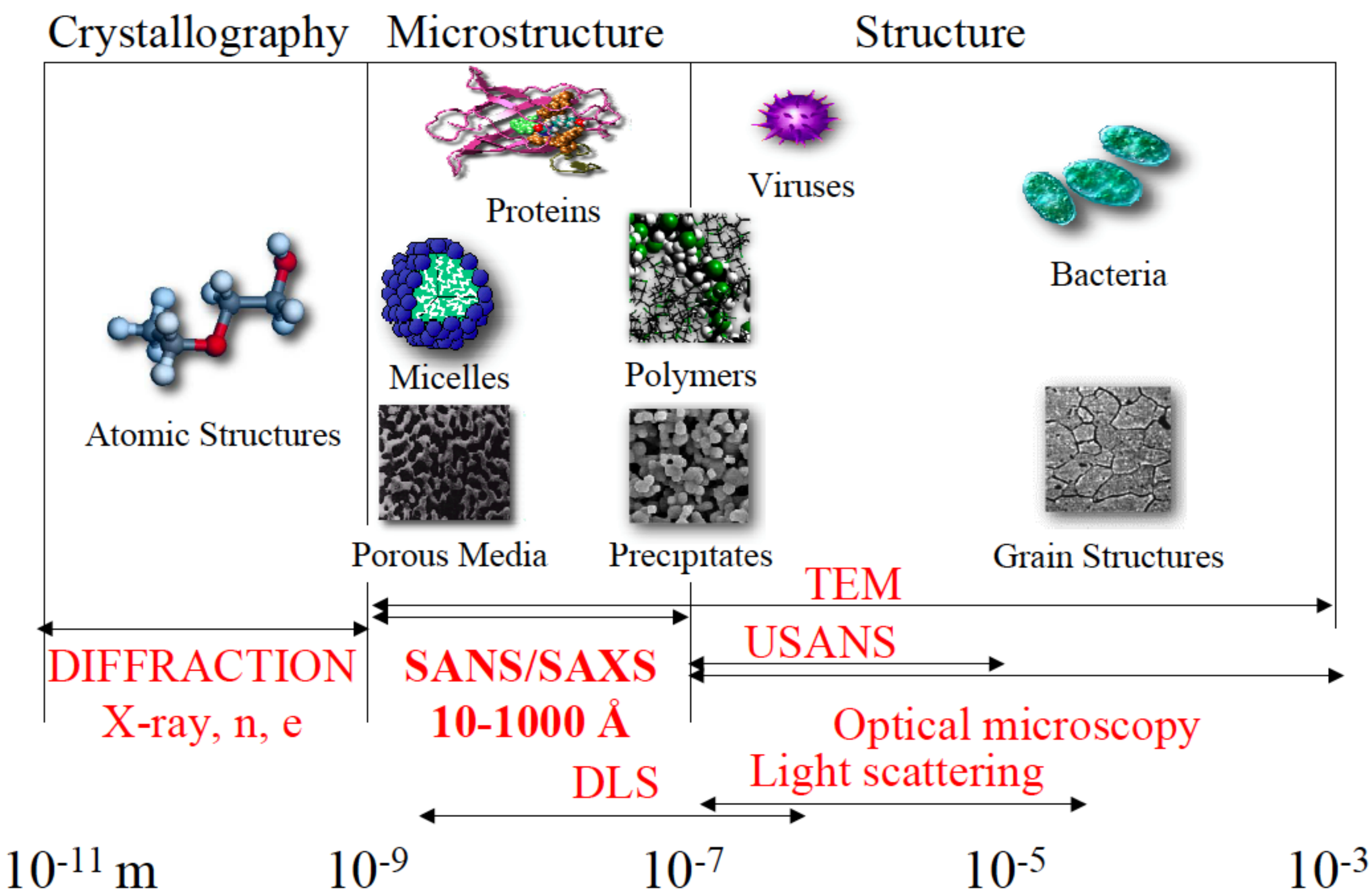
Neutron interact with nuclei
→ Neutrons see low-Z atoms



Material for Li-battery seen by
X rays (left) and
Neutrons (right)

T. Kamiyama, et al.

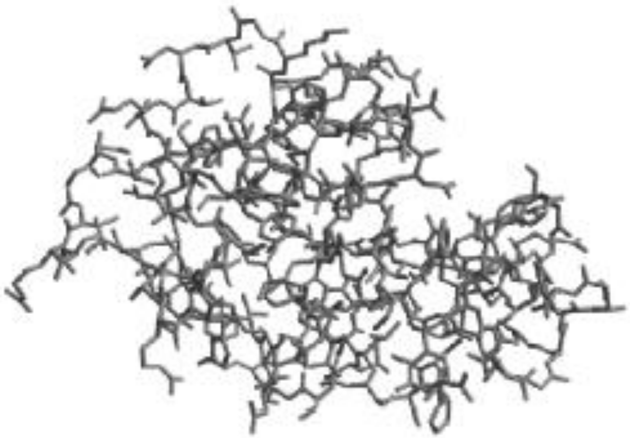
Small Angle Neutron Scattering



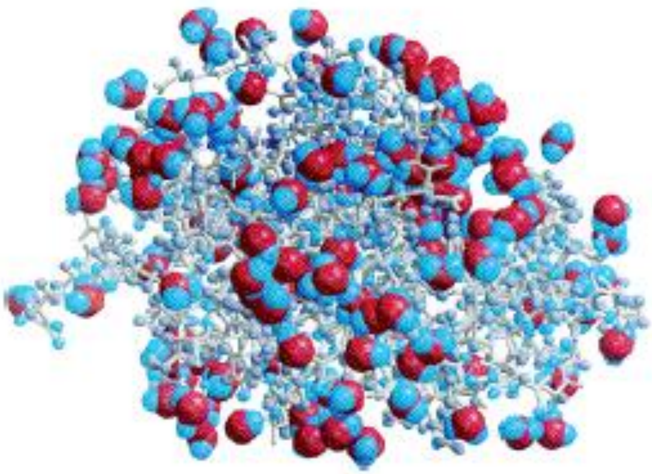
Advantages of SANS over SAXS are its sensitivity to light elements, the possibility of isotope labeling, and the strong scattering by magnetic moments.

Complementarity between X-rays & Neutrons

Hen Egg-White Lysozyme



X-rays

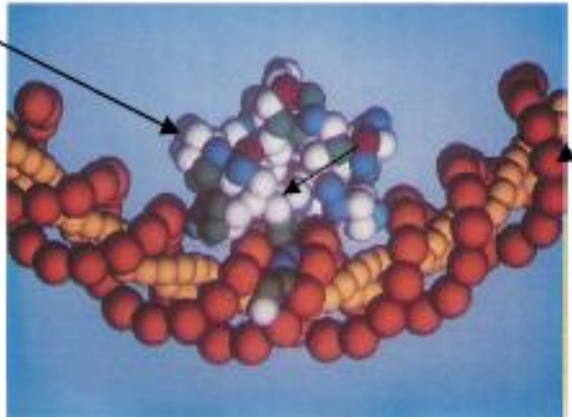


Neutrons

Water molecules Observed with neutrons

N. Niimura, et al.

Protein

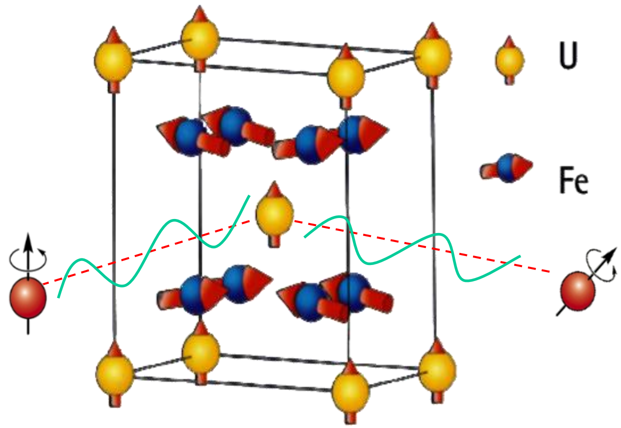


DNA

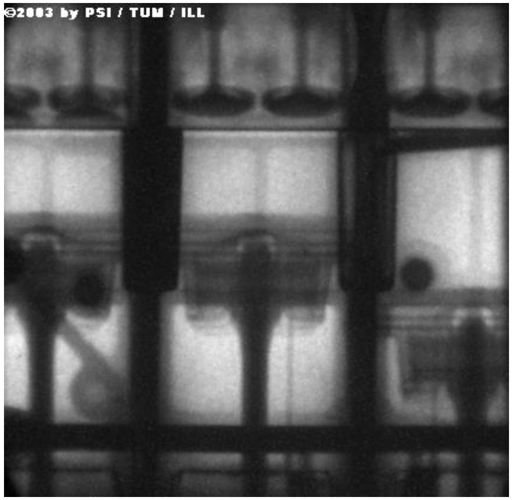
A proten molecule moving along the DNA chain

From structure to function

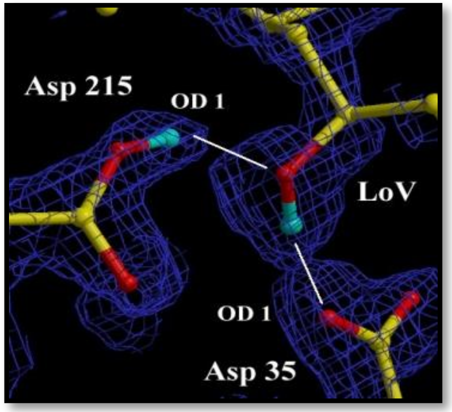
Complementarity between X-rays & Neutrons



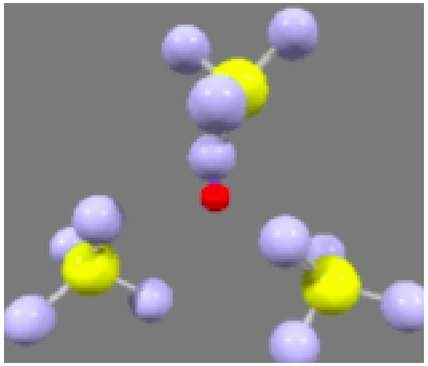
..see magnetic atoms



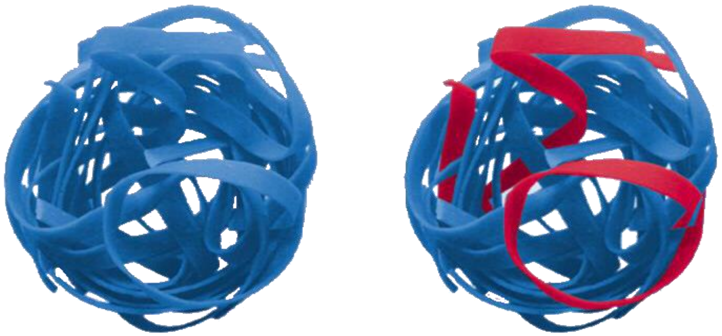
..see inside materials



..see light atoms

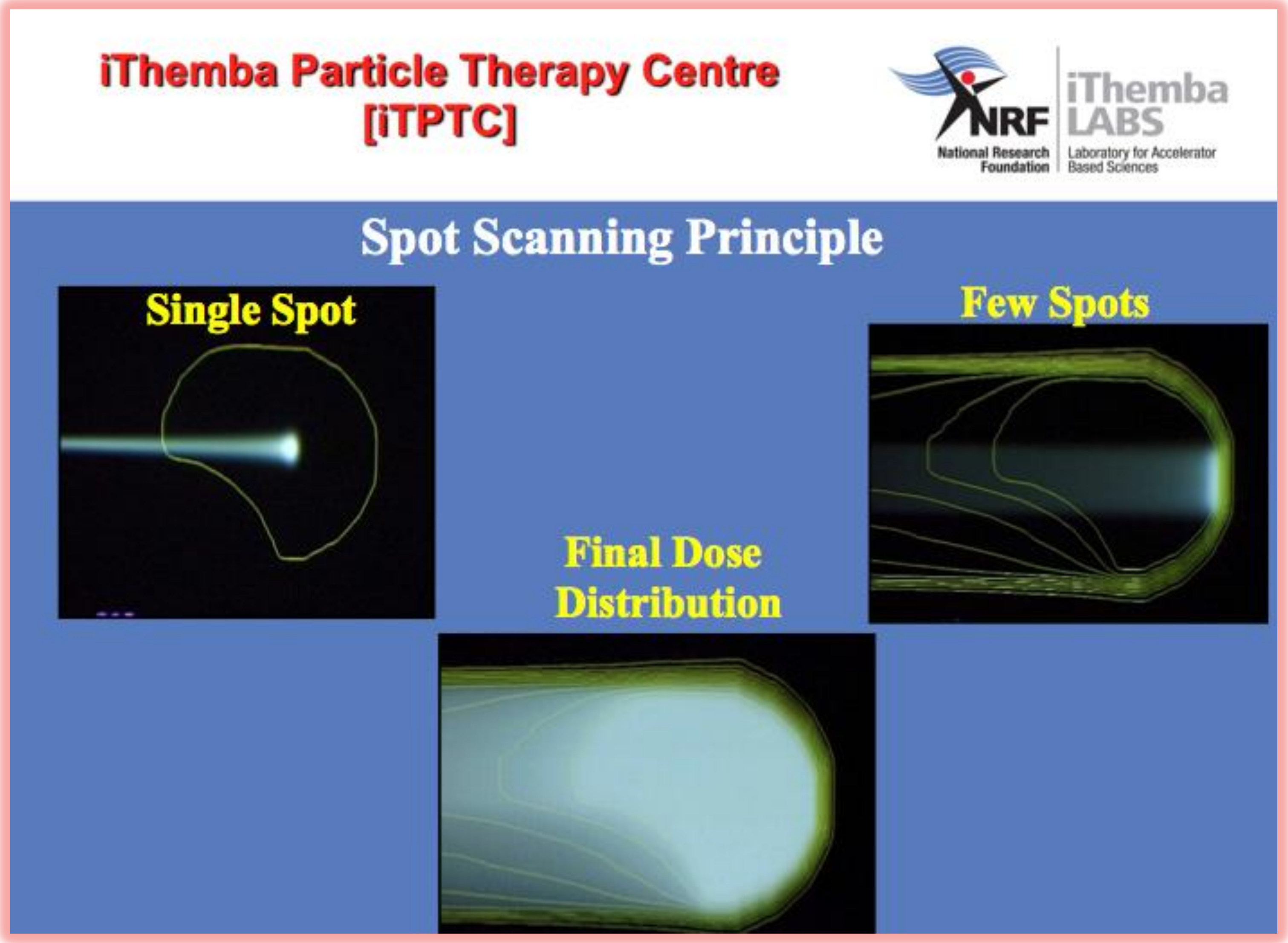


..see atoms move



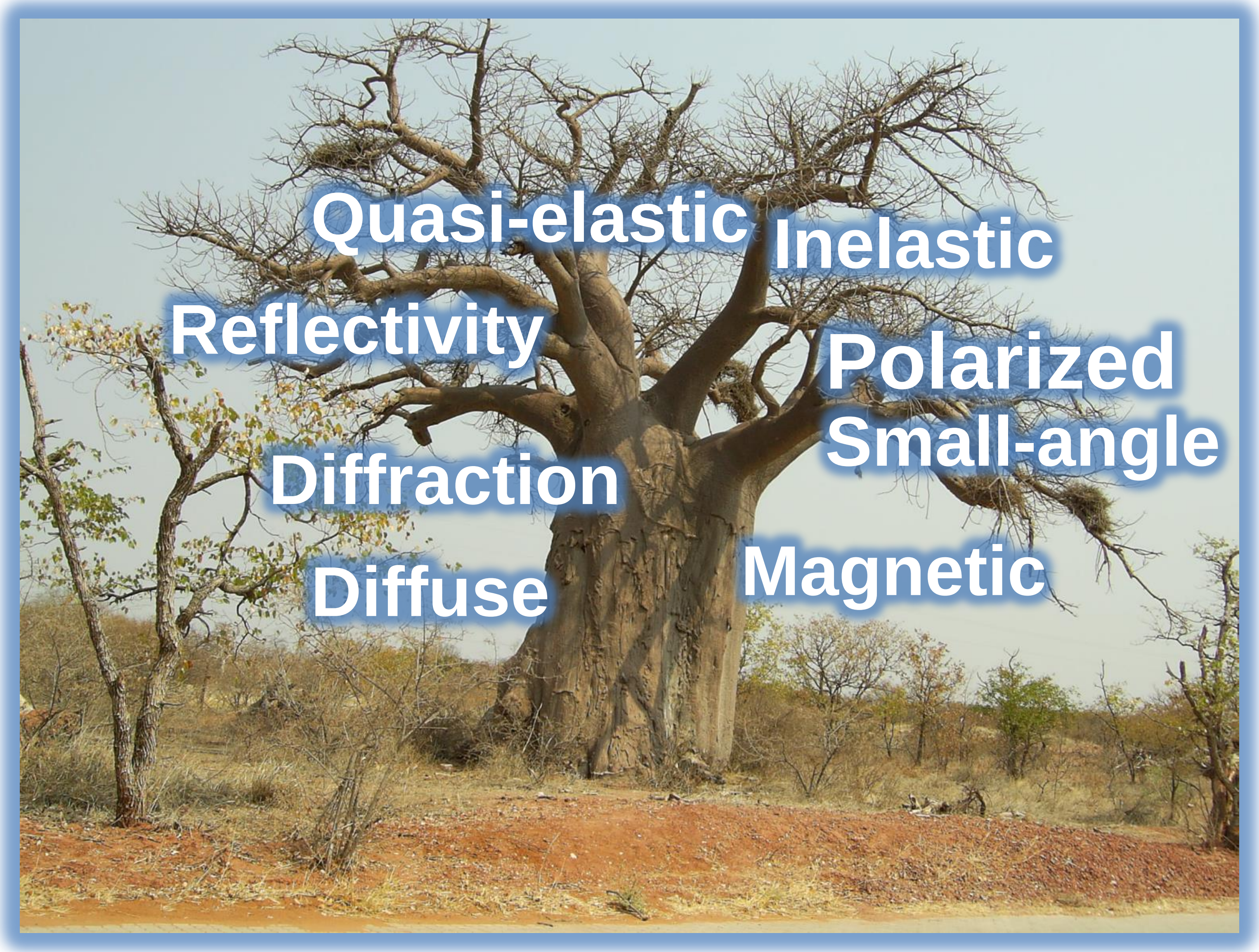
..see isotopes

Neutron beam and X-Ray for Medical Applications



Scattering and Diffraction

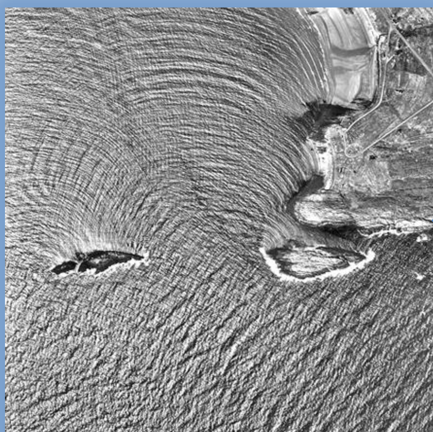
Neutron Scattering Techniques

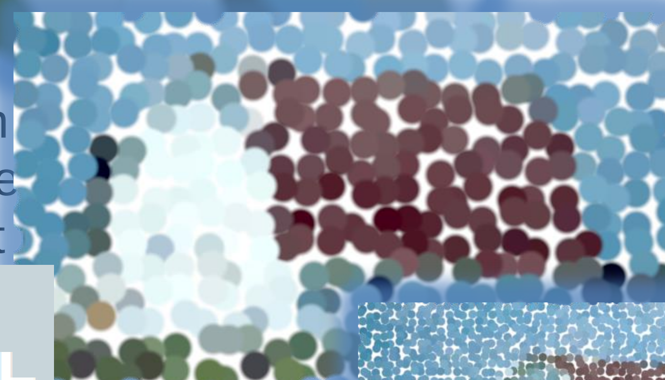


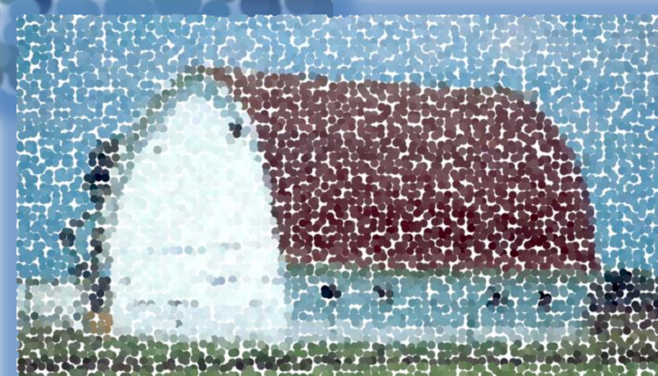
Scattering – coherence and incoherence

$$\sigma_{coh} = 4\pi\langle b\rangle^2$$

$$\sigma_{incoh} = 4\pi\left(\langle b^2\rangle - \langle b\rangle^2\right)$$







Definition ... ction units.
One depe... 't (where atoms
are, what ...

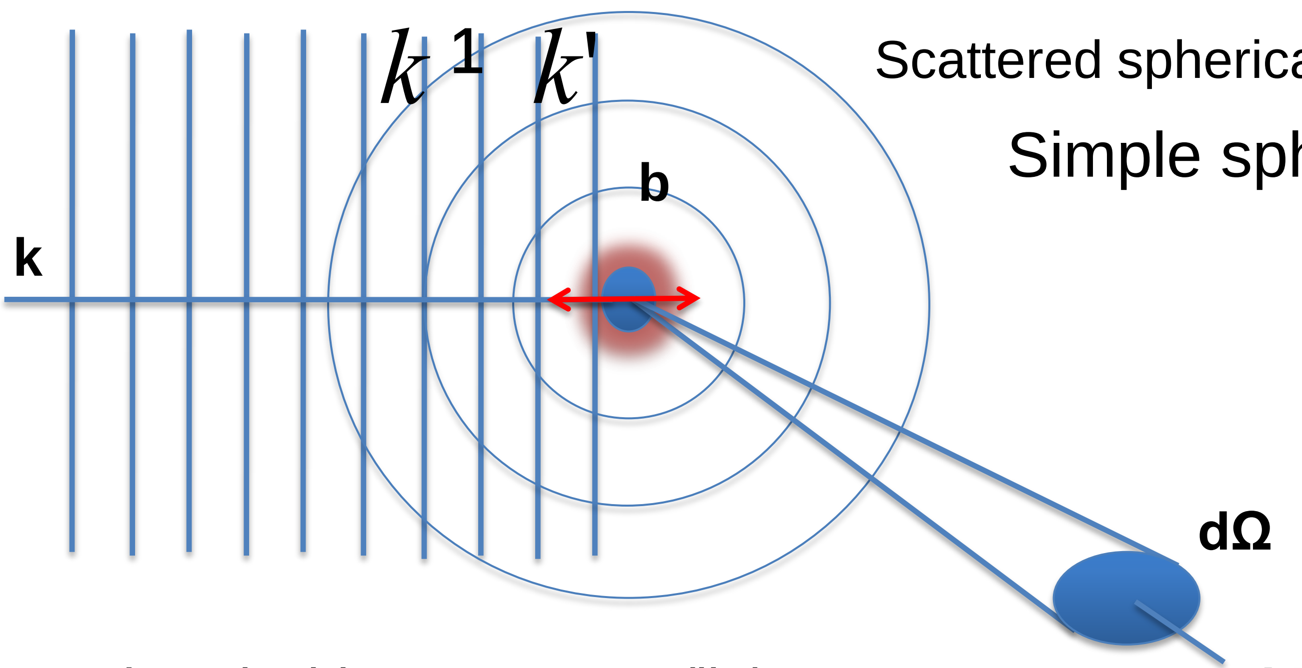
WHY NEUTRON SCATTERING IS USEFUL

When used as a probe for small samples of materials, neutron beams have the power to reveal what is invisible using other radiations. Neutrons can appear to behave either as particles or as waves or as microscopic magnetic dipoles, and it is these specific properties which enable them to uncover information which is often impossible to access using other techniques.

Isotopes and spin?

Inelastic Scattering of plane wave

σ_{tot}=number of neutrons scattered in all directions per sec/incident flux



Scattered spherical wave: –b exp(ikr)/b

Simple spherical object!!

Plane incident wave: exp(ikr)

$$\left(\frac{d^2\sigma}{d\Omega\, dE}\right)$$

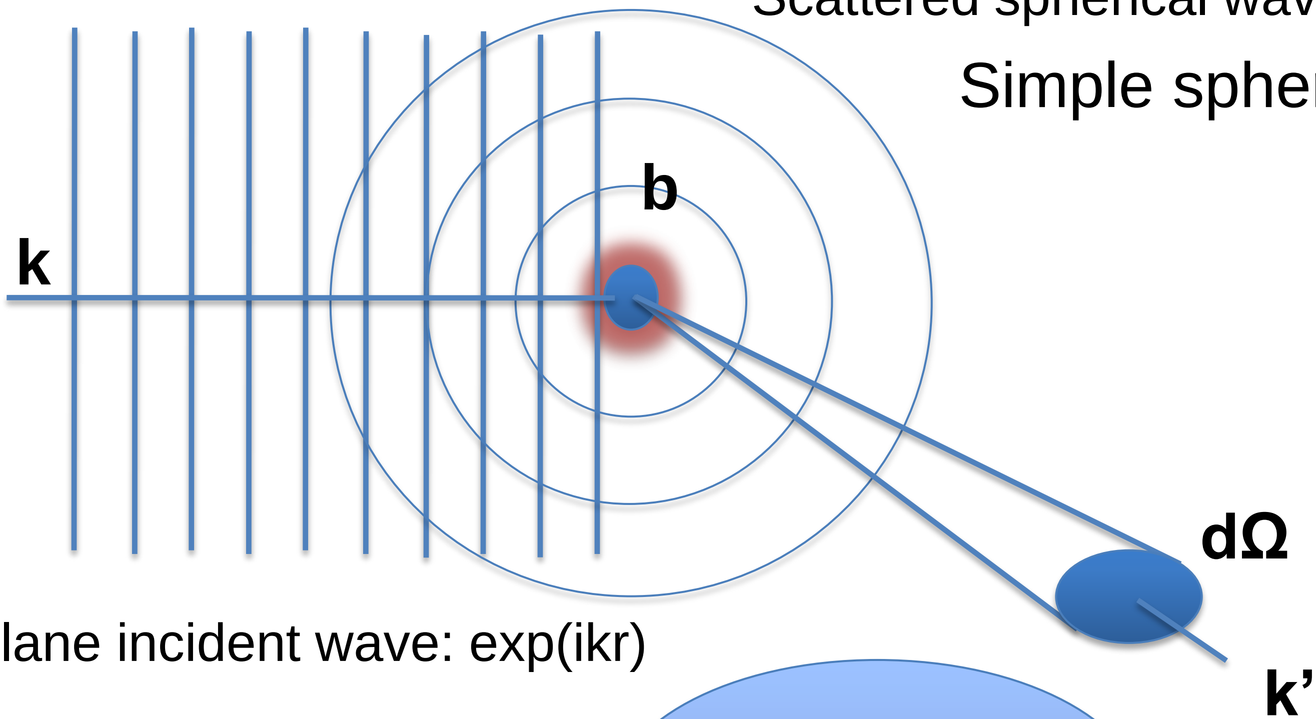
$$s_{tot} = 4pb^2$$

Diffraction = Coherent Elastic Scattering of plane wave

σ_{tot}=number of neutrons scattered in all directions per sec/incident flux

Scattered spherical wave: –b exp(ikr)/b

Simple spherical object!!



$$\left(\frac{d\sigma}{d\Omega}\right), \left(\frac{d^2\sigma}{d\Omega\, dE}\right)$$

$$s_{tot} = 4 p b^2$$

Scattering

- Cross section:

$$d\sigma = \frac{(\#\ \text{particles scattered into solid angle } \Delta\Omega/\text{s})}{(\#\ \text{particles incident/sec})(\#\ \text{scattering centers/area})}$$

$$\frac{d\sigma(Q)}{d\Omega} = N_p V_p \overline{\rho} P(Q) S(Q)$$

Volume fraction

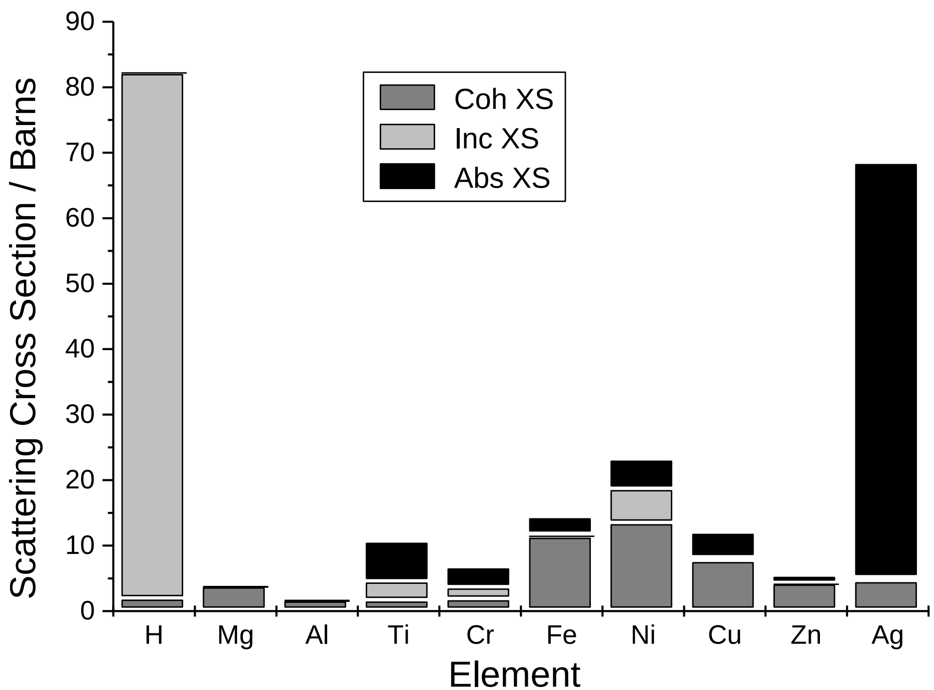
Contrast

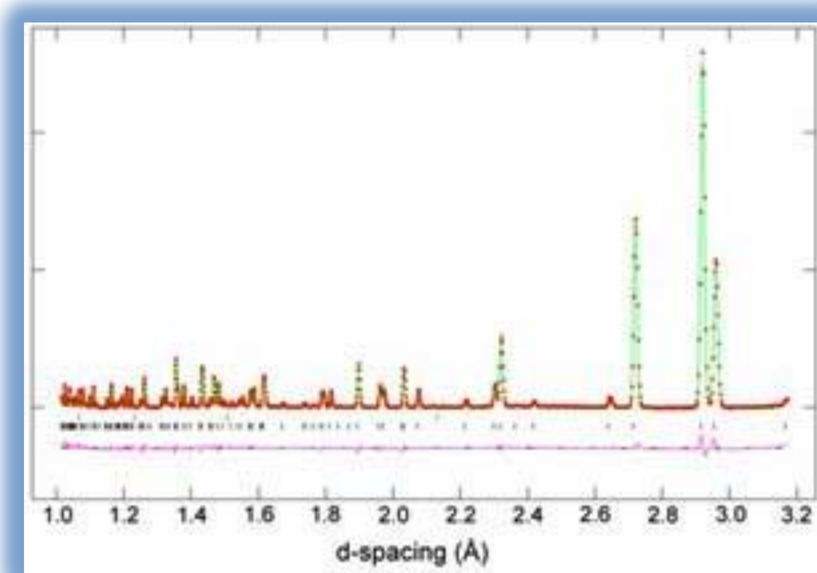
Shape

Interaction

Scattering

Coherent XS~ Signal
Incoherent XS~ Background
Absorption XS~1/Intensity





The neutron diffraction pattern of sulfuric acid tetrahydrate at 4.2 K as seen by the 90 degree detectors on HRPD, and fitted with the existing X-ray derived structural model for the deuterated species.

Neutron cross sections

EUROPEAN SPALLATION SOURCE

NIST Center for Neutron Research

NIST National Institute of Standards and Technology

Home ICP Experiments UserProposal Instruments SiteMap

Neutron scattering lengths and cross sections

Isotope	conc	Coh b	Inc b	Coh xs	Inc xs	Scatt xs	Abs xs
Fe	---	9.45	---	11.22	0.4	11.62	2.56
54Fe	5.8	4.2	0	2.2			
56Fe	91.7	9.94	0	12.4			
57Fe	2.2	2.3	---	0.66			
58Fe	0.3	15.(7.)	0	28			

He
C N O F Ne
Si P S Cl Ar

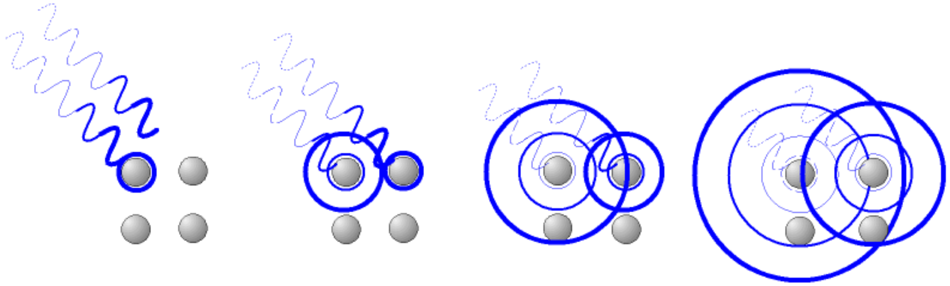
Isotope	conc	Coh b	Inc b	Coh xs	Inc xs	Scatt xs	Abs xs
Gd	---	6.5-13.82*i*	---	29.3	151.(2.)	180.(2.)	49700.(125.)
152Gd	0.2	10.(3.)	0	13.(8.)	0	13.(8.)	735.(20.)
154Gd	2.1	10.(3.)	0	13.(8.)	0	13.(8.)	85.(12.)
155Gd	14.8	6.0-17.0*i*	(+/-)5.(5.)-13.16*i*	40.8	25.(6.)	66.(6.)	61100.(400.)
156Gd	20.6	6.3	0	5	0	5	1.5(1.2)
157Gd	15.7	-1.14-71.9*i*	(+/-)5.(5.)-55.8*i*	650.(4.)	394.(7.)	1044.(8.)	259000.(700.)
158Gd	24.8	9.(2.)	0	10.(5.)	0	10.(5.)	2.2
160Gd	21.8	9.15	0	10.52	0	10.52	0.77

NOTE: The above are only thermal neu
dependent cross sections please go to t

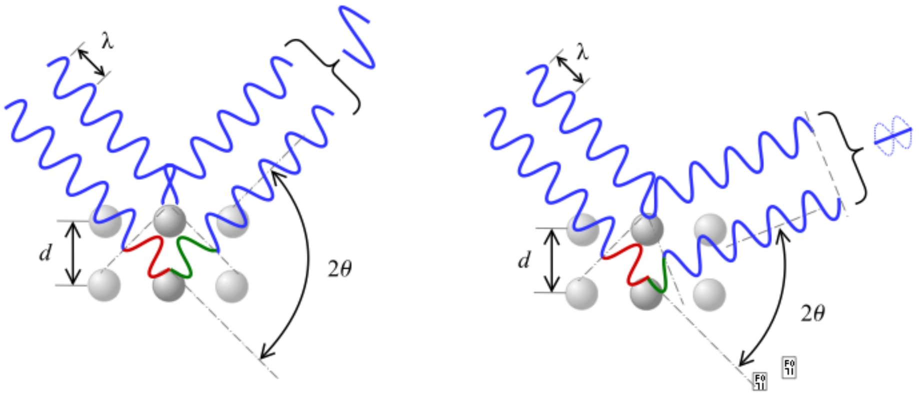
Select the element, and you will get a lis
Feature section of neutron scattering le
3, 1992, pp. 29-37.

Diffraction - Bragg

Diffraction of X-rays or neutrons by polycrystalline samples is one of the most important, powerful and widely used analytical techniques available to materials scientists. For most crystalline substances of technological importance, the bulk properties of a powder
or a polycrystalline solid, averaged throughout the sample, are required; in general a single-crystal data, even if they can be obtained, are usually of little interest except for determination of the crystal structure or for studying some other fundamental physical property. By *J Ian Langford and Daniel Louer*



X-rays interact with the atoms in a crystal.

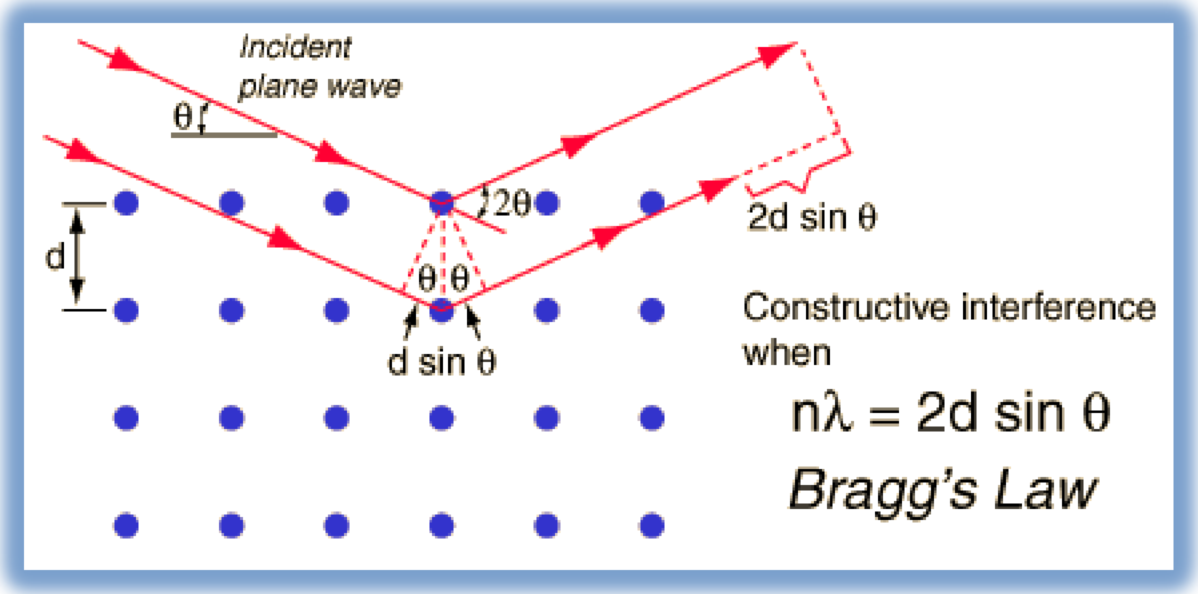


According to the 2θ deviation, the phase shift causes constructive (left figure) or destructive (right figure) interferences.

Diffraction - Bragg

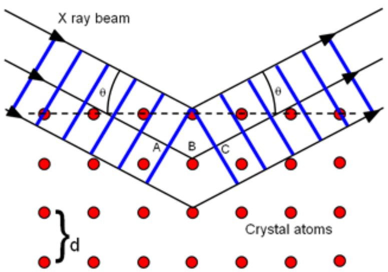
- Bragg / Laue scattering
- Coherent elastic scattering

Two beams with identical wavelength and phase approach a crystalline solid and are scattered off two different atoms within it. The lower beam traverses an extra length of 2dsinθ. Constructive interference occurs when this length is equal to an integer multiple of the wavelength of the radiation.



Diffraction - Bragg

Using the grains as internal strain gauges

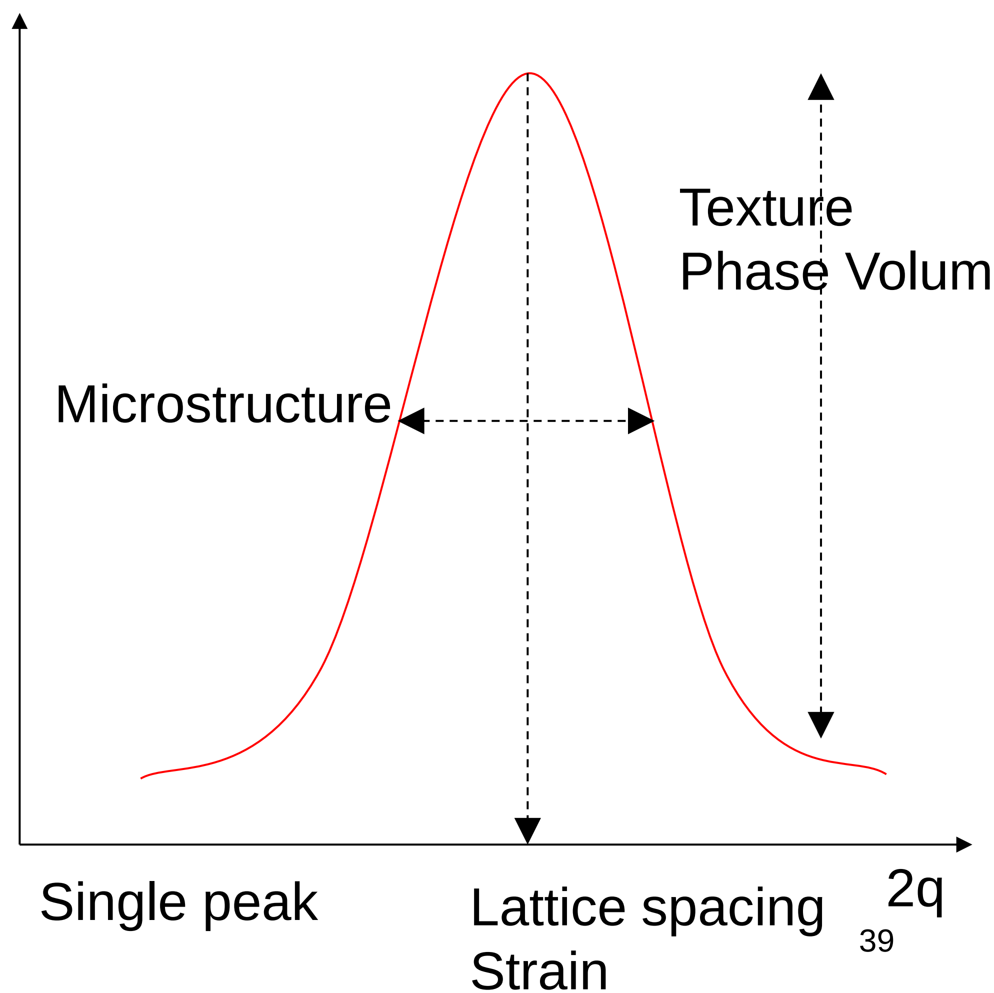


$$\lambda = 2d\sin\theta$$

$$\varepsilon = -\cot(\theta)\,(\theta - \theta_0)$$

Two ways to measure d:
-keep λ fixed and measure θ
constant wavelength
-keep θ fixed and measure λ
time-of-flight

Braggs law gives information about position of a diffraction peak from a type of lattice planes. Rietveld approach calculated height, position and width of diffraction peaks from first principles, I.e. Temperature, composition, vacancies, etc.

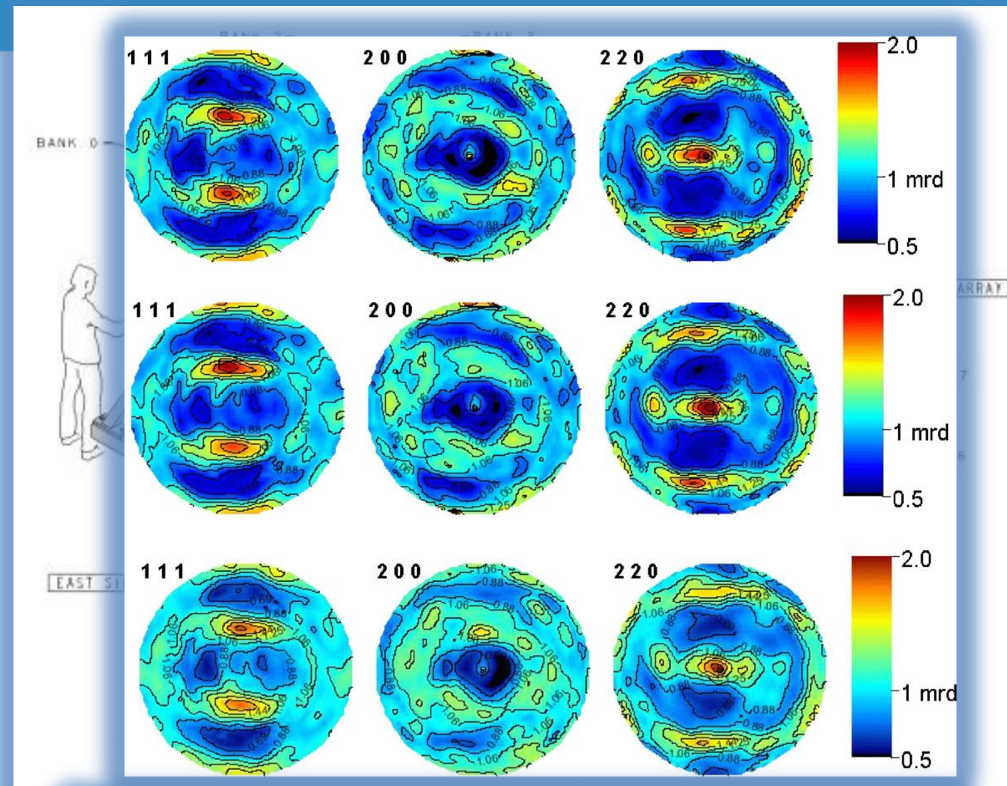


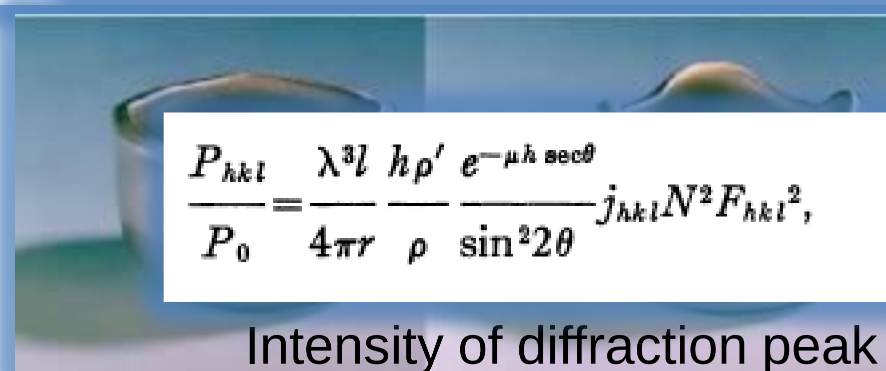
Diffraction: Texture

- ISIS: GEM instrument
- Near 4π coverage

Example:

- Cold rolled copper, simulating manufacturing process for archeometry
- 2mm thick disks
- $20 \times 20 mm^2$ beam
- 2 min counting times



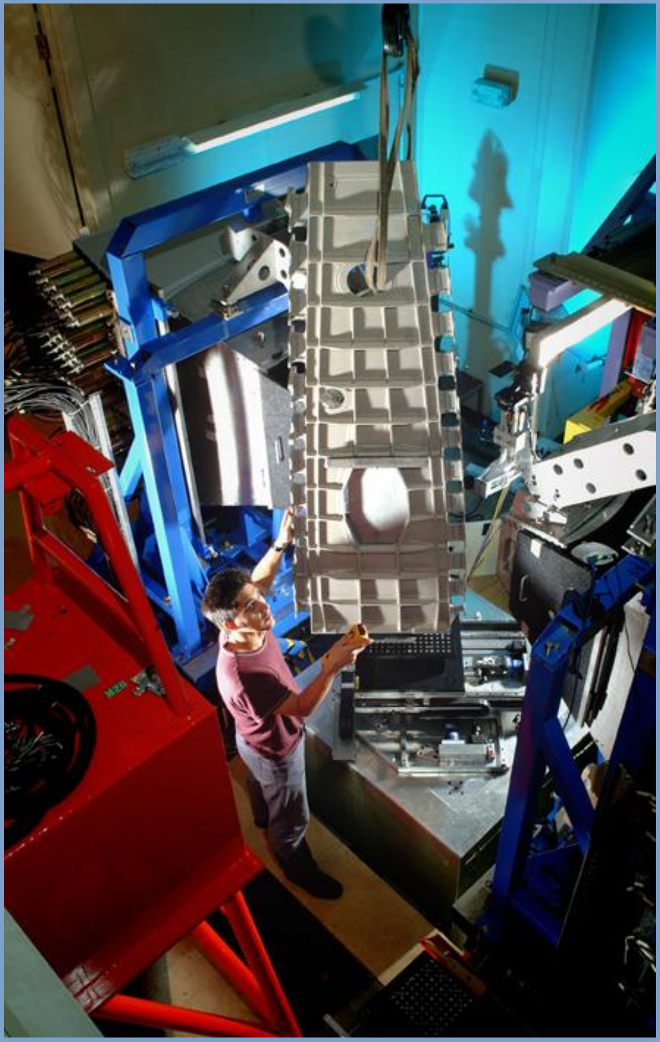


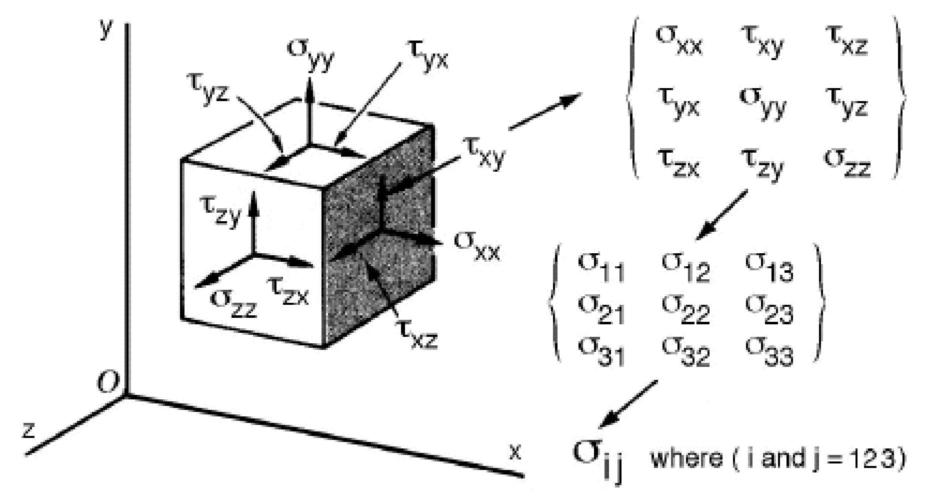
$$\frac{P_{hkl}}{P_0} = \frac{\lambda^3 l}{4\pi r} \frac{h\rho'}{\rho} \frac{e^{-\mu h \sec\theta}}{\sin^2 2\theta} j_{hkl} N^2 F_{hkl}{}^2, \qquad (15)$$

Intensity of diffraction peak

Neutron texture analysis on GEM at ISIS
W. Kockelmann, L.C. Chapon and P.G. Radaelli
Physica B, Vol. 385-386 (1), pp639-643 (2006)

Diffraction: Stress and Strain



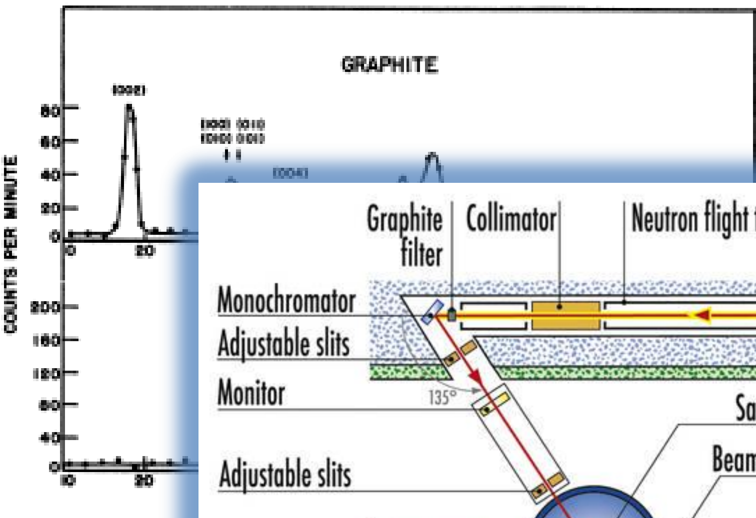


Applications:
Residual stresses
Fatigue/Structural Integrity
Welds
Alloy development
Microstructure/Texture
Phase transformation
In-situ experiments

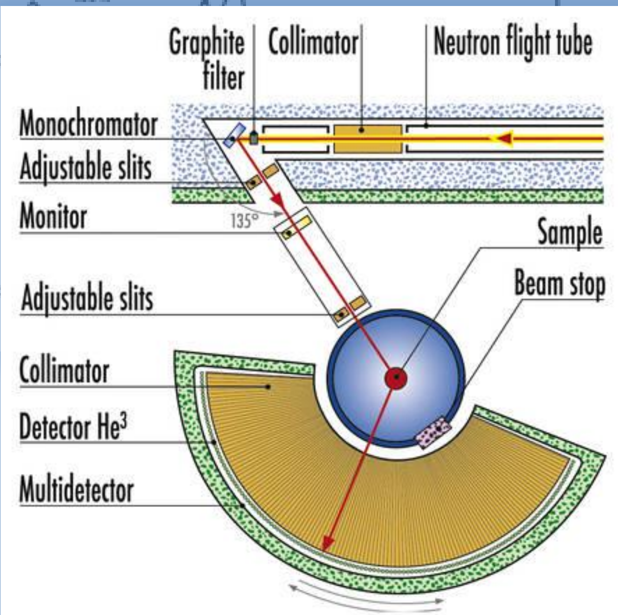
Example - Diffraction

PHYSICAL REVIEW VOLUME 73, NUMBER 8 APRIL 15, 1948

The Diffraction of Neutrons by Crystalline Powders



...ig. 3. Powder diffraction pat-...erns for diamond and graphite. ...he major part of the diffuse ...cattering in these patterns ...rises from multiple scattering ... the samples.



D2B at the ILL, Grenoble, 50 years later

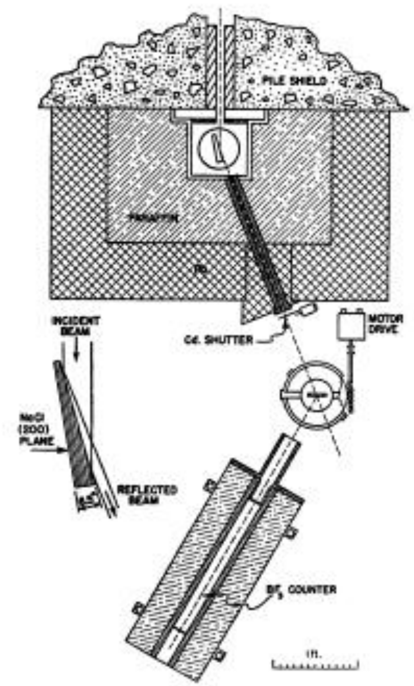
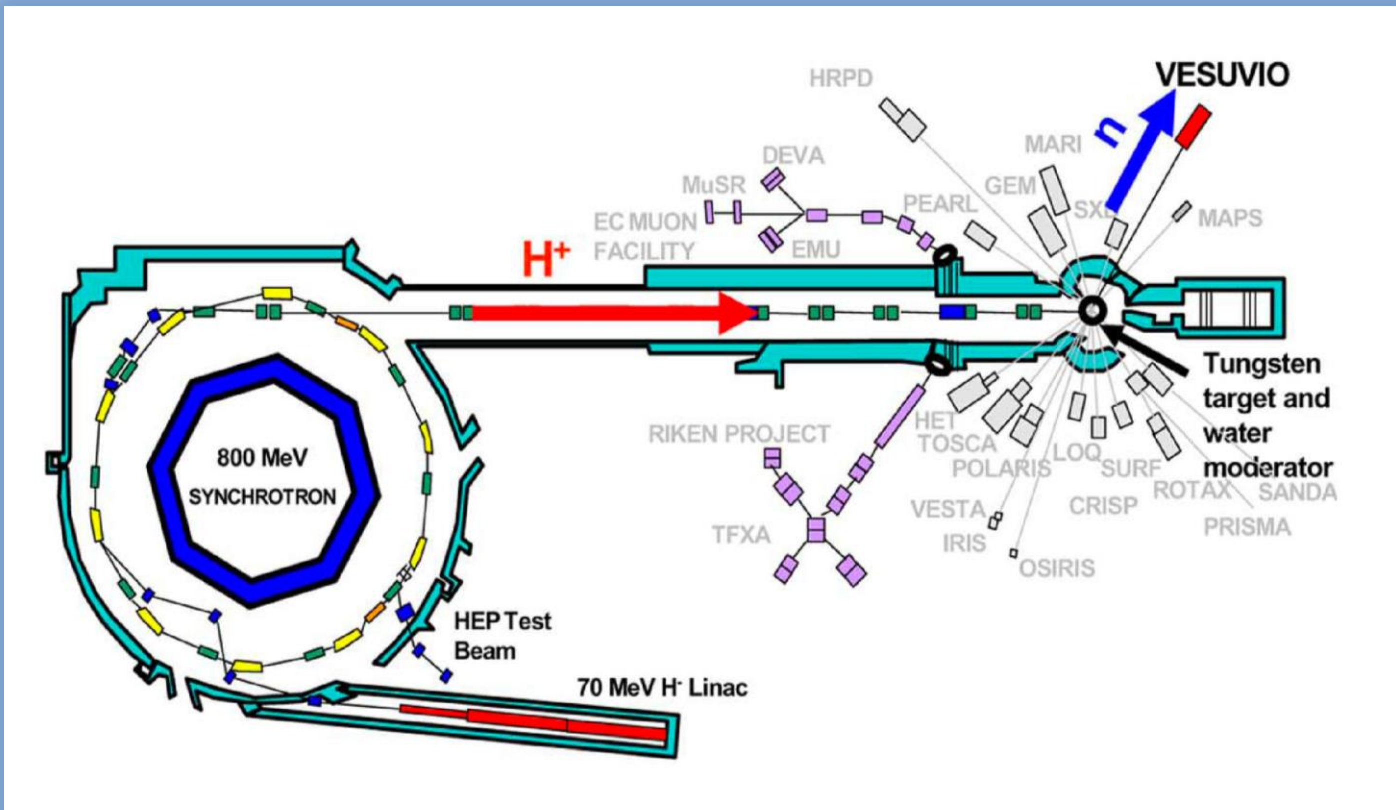


FIG. 1. Arrangement of apparatus, showing the monochromating crystal (detailed in left center) collimating slits, shielding, second spectrometer with location of powder specimen and counter.

$$\frac{P_{hkl}}{P_0}=\frac{\lambda^3 l}{4\pi r}\frac{h\rho'}{\rho}\frac{e^{-\mu h \sec\theta}}{\sin^2 2\theta} j_{hkl} N^2 F_{hkl}^2, \qquad (15)$$

Intensity of diffraction peak

Time of Flight



$$\lambda = \frac{h}{p} = \frac{h}{mv} = \frac{ht}{mL}$$

De Broglie

+

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

Bragg

=

Time-of-Flight:

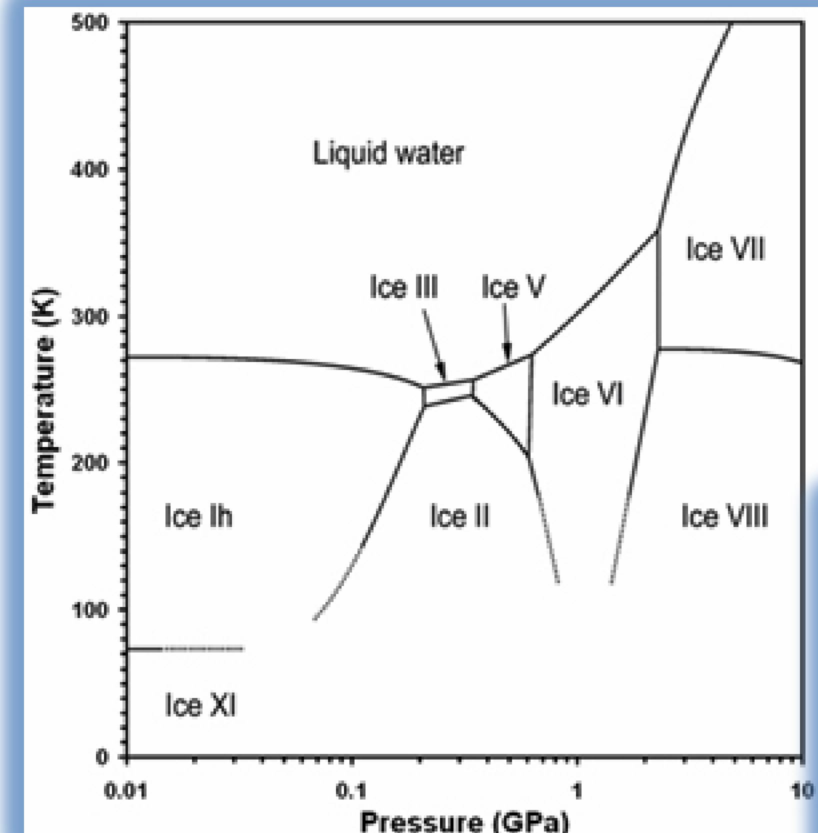
$$d = \frac{h}{2mL\sin\theta}t$$

Fast, short-wavelength neutrons arrive earlier at detector!

Diffraction in ToF mode at pulsed source. Flight time is propre to wavelength

Ice

EUROPEAN SPALLATION SOURCE



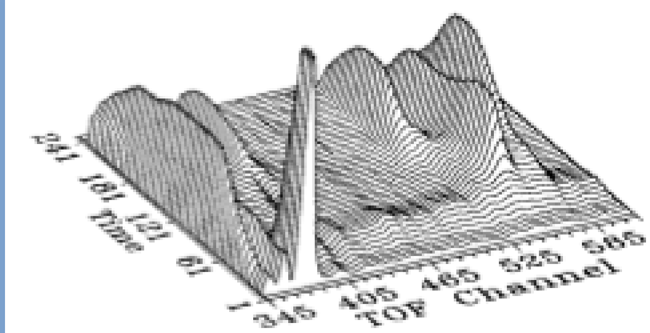


Figure 1. Neutron diffraction patterns measured in the course of heating of the recovered D_2O high-pressure phase ice VIII from 94 to 273 K.

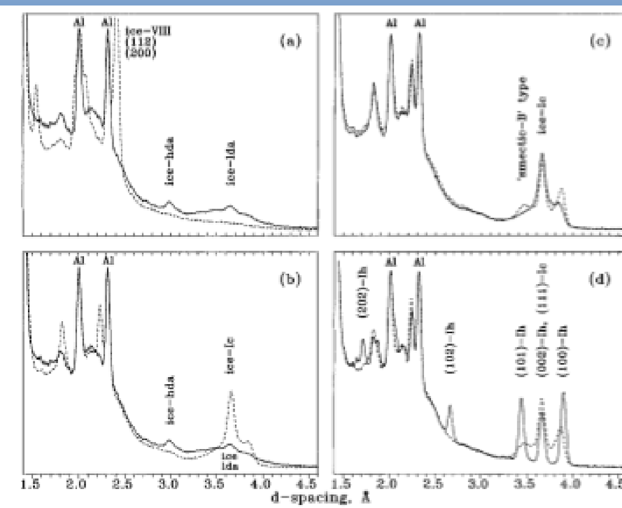


Figure 2. Neutron diffraction patterns of D_2O ices before and after phase transitions: (a) from ice VIII (dashed line) to a mixture of hda + lda ices (solid line) at 125 K; (b) from the mixture of hda + lda ices (solid line) to ice Ic (dashed line) at 160 K; (c) from ice Ic (solid line) to the hexagonal phase, "smectic B" type structure (dashed line), at 190 K; (d) from "smectic B" type ice (dashed line) to ice Ih (solid line) at 230 K. The diffraction patterns also contain peaks from the aluminum sample can and cryostat.

Batteries

EUROPEAN SPALLATION SOURCE

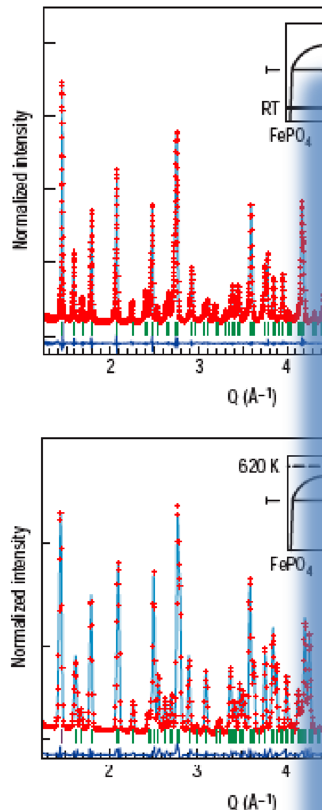


Figure 2 Neutron diffraction patterns measured at t… FePO$_4$–LiFePO$_4$ binary phase diagram. a,b, Rietveld r… time-of-flight neutron diffraction profile measured for L… (a) and the angle-dispersive neutron diffraction profile … 620 K (b). Two different neutron diffractometers were u… information for each measurement as explained in the … points are plotted using the common scale $Q = 4\pi\sin$… *Q* range for VEGA and HERMES for comparison. Specifi… composition and temperature are given in the inset pha… Delacourt *et al.*[6] and Dodd *et al.*[20]. Observed intensity … Y_{calc} are represented by red plus signs and the green so… blue curve at the bottom represents the residual differe… parameters are summarized in Supplementary Informa… impurity phase was identified, and the crystal structure… with the space group *Pnma*.

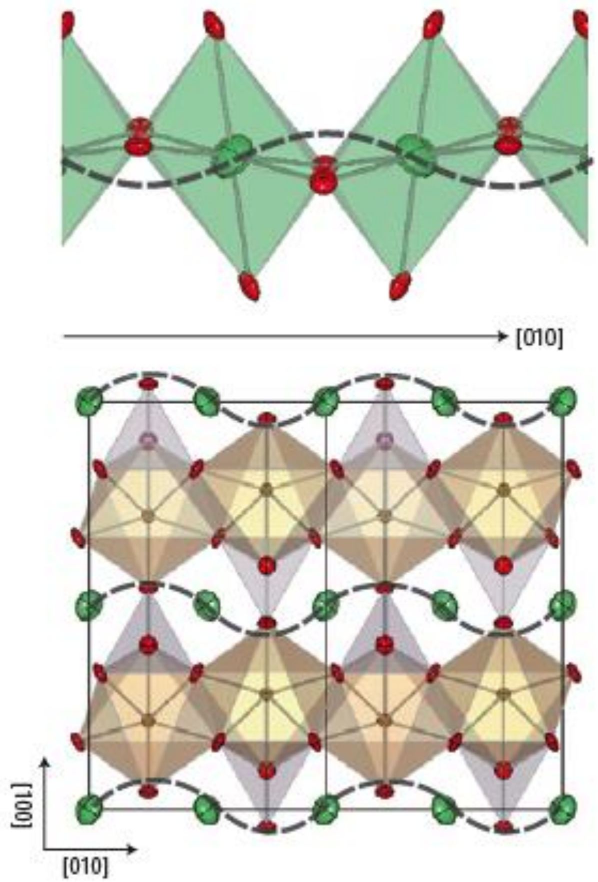
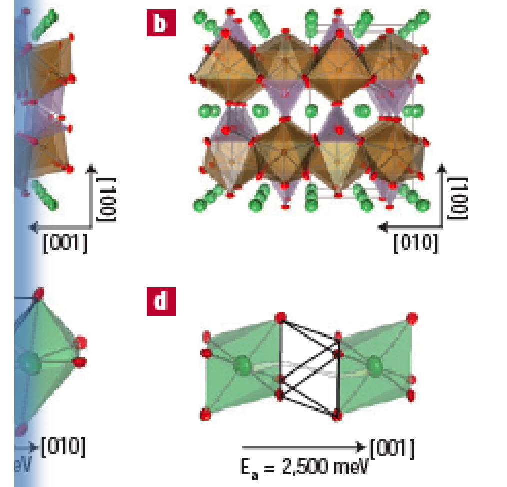


Figure 3 Anisotropic harmonic lithium vibration in LiFePO$_4$ shown as green thermal ellipsoids and the expected diffusion path. The ellipsoids were refined with 95% probability by Rietveld analysis for room-temperature neutron diffraction data. Expected curved one-dimensional continuous chains of lithium motion are drawn as dashed lines to show how the motions of Li atoms evolve from vibrations to diffusion.



…of LiFePO$_4$ and possible lithium pathways. a,b, The …projected along the [010] (a) and [001] (b) directions. …hways are parallel to these directions. The structures …l parameters obtained through this work and …ry Information, Table S1. The structure can be …agonal close-packed oxygen sub-array, in which Li, Fe …tial sites to form (1) corner-sharing FeO$_6$ octahedra that …a distorted two-dimensional square lattice perpendicular …ng LiO$_6$ octahedra aligned in parallel chains along the …$_4$ groups connecting neighbouring planes or arrays. The …d ellipsoids indicate Li, Fe, P and O atoms, respectively. …e lithium migration paths: c, along the [010] direction …tetrahedral sites; and d, along the [001] direction …dral sites. One-dimensional diffusion along the [010] …y the computational method[15,16].

N…

a…
p…
o…
o… …ng stresses…

Fuel Cells

EUROPEAN SPALLATION SOURCE

Electric Circuit
(40% - 60% efficiency)

Available online at www.sciencedirect.com
ScienceDirect
Solid State Ionics 177 (2006) 2357–2362
SOLID STATE IONICS
www.elsevier.com/locate/ssi

ELSEVIER

Proton c

Fig. 2. Low temperature (10K) neutron powder diffraction patterns of as-prepared and respective deuterated (marked with D) $BaZr_{1-x}In_xO_{3-\delta}$ (x = 0.00, 0.25 and 0.50) samples.

Table 1
Summary of results obtained from Rietveld analysis of neutron powder diffraction data for $BaZr_{1-x}In_xO_{3-0.5x}$, collected at 10K on the NPD diffractometer

	x=0.0 (as-prepared)	x=0.0 (deuterated)	x=0.25 (as-prepared)	x=0.25 (deuterated)	x=0.50 (as-prepared)	x=0.50 (deuterated)
Space group	Pm-3m	Pm-3m	Pm-3m	Pm-3m	Pm-3m	Pm-3m
a (Å)	4.1879(1)	4.1880(1)	4.1916(1)	4.1983(1)	4.1942(2)	4.2260(4)
Thermal parameters, B_{iso} (Å^2)						
Ba on 1(b) 1/2,1/2,1/2	0.09(3)	0.08(2)	0.10(3)	0.21(4)	0.48(6)	0.97(10)
Zr on 1(a) 0,0,0	0.14(2)	0.11(2)	0.19(3)	0.23(3)	0.43(5)	0.37(9)
In on 1(a) 0,0,0	–	–	0.19(3)	0.23(3)	0.43(5)	0.37(9)
O on 3(d) 1/2,0,0	0.24(2)	0.26(2)	0.63(2)	0.56(2)	1.16(4)	1.17(5)
Oxygen site occupancy	2.98(1)	2.99(1)	2.82(1)	2.90(2)	2.68(2)	2.98(3)
χ^2	1.96	1.99	1.85	1.64	1.72	3.16
Weighted R_{wp}	4.17%	5.16%	4.7%	4.74%	5.28%	6.62%
Bragg R_B	3.97%	3.86%	4.63%	4.40%	8.16%	6.62%
Fitted parameters	16	16	16	16	16	16

Abst

Th of lo group expe deute The conductivity of the h number of protons in proton conduction is

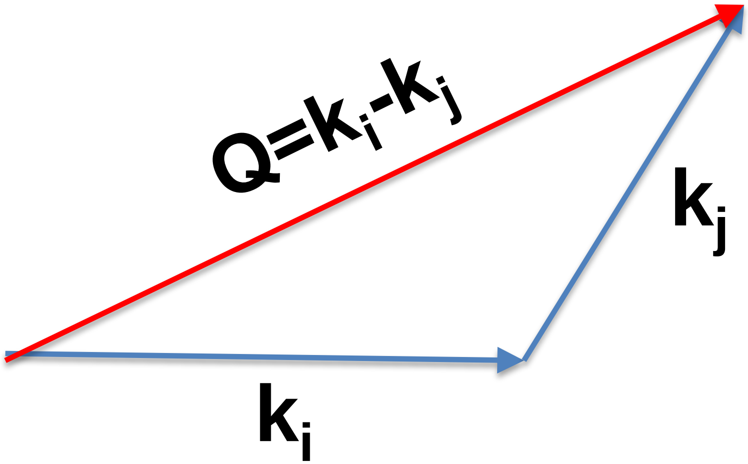
Ap
str
(water transport) on macro scale.

Small Angle Scattering

- Scattering Vector

$$\text{Sin } \theta = Q\lambda/4\pi = \lambda/2d$$

$$\rightarrow d = 2\pi/Q$$



Small angle → small Q → large distances
long λ (cold neutrons) = 6 Å, d = 200 Å,
$2\theta = 1.7\ °$

→ The larger the object, the smaller the angle

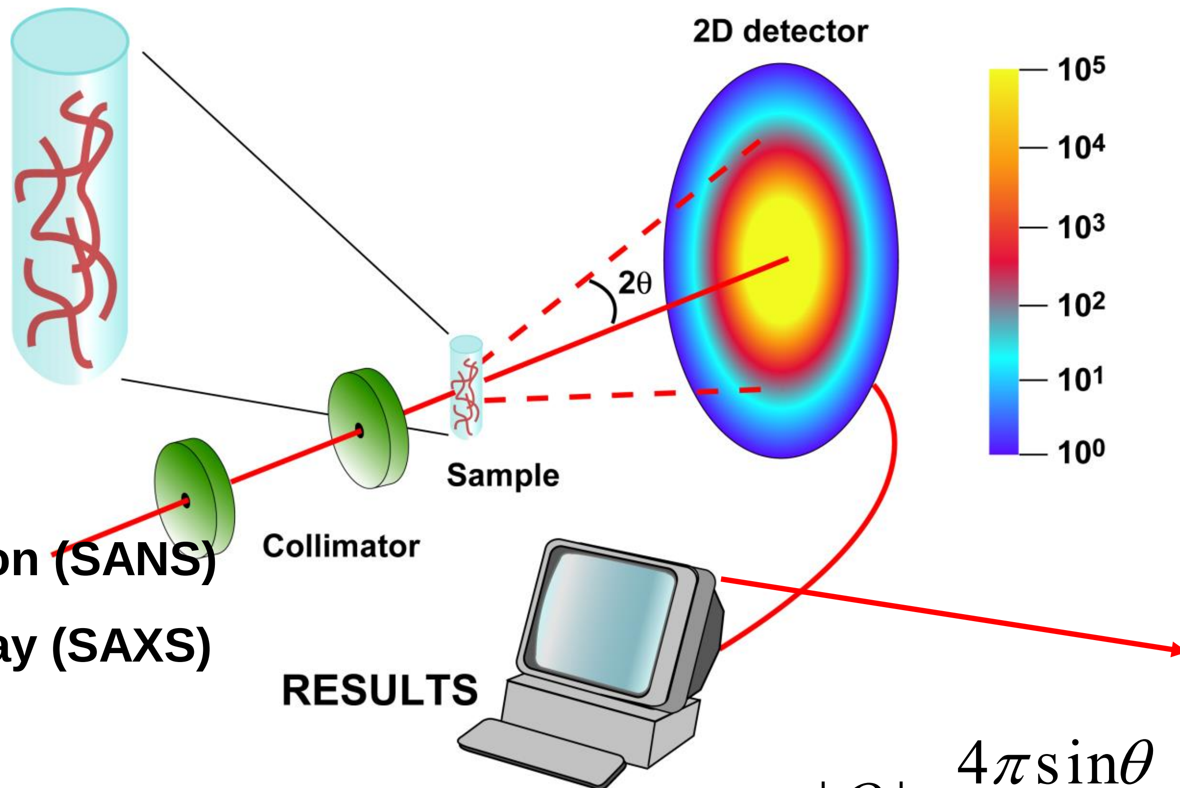
SANS: Experimental Setup

EUROPEAN SPALLATION SOURCE

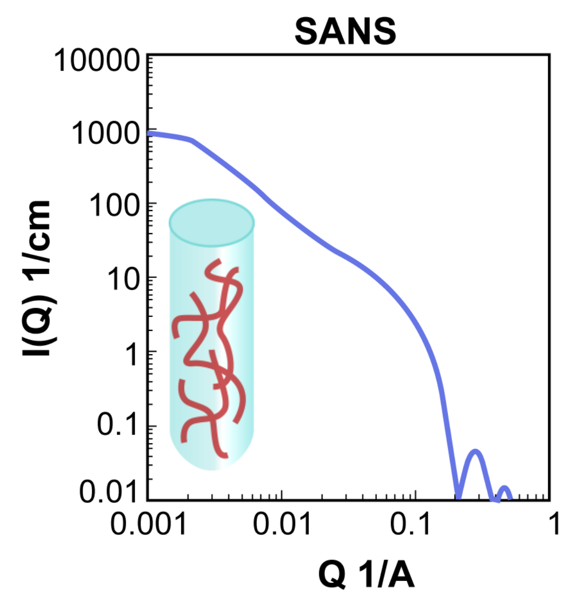
Beam:

Neutron (SANS)

or X-ray (SAXS)



Measures structures at length scales from 1 nm to 100 nm

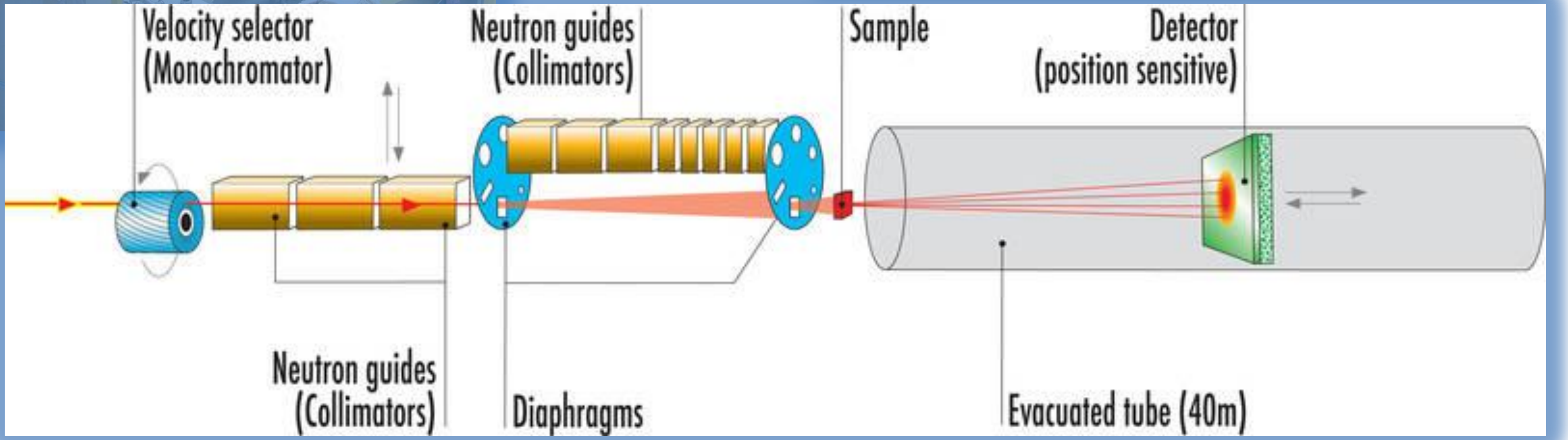


$$|Q| = \frac{4\pi \sin\theta}{\lambda}$$

$Q_{min} \approx 0.03°$, $Q_{max} \approx 3°$

In a standard crystallography experiment, theta_max is typically 45 degrees

Small Angle Instruments



Scattering

EUROPEAN SPALLATION SOURCE

- Cross section:

$$d\sigma = \frac{(\#\ \text{particles scattered into solid angle } \Delta\Omega/\text{s})}{(\#\ \text{particles incident/sec})(\#\ \text{scattering centers/area})}$$

$$\frac{d\sigma(Q)}{d\Omega} = N_p V_p \overline{\rho} P(Q) S(Q)$$

Volume fraction

Contrast

Shape

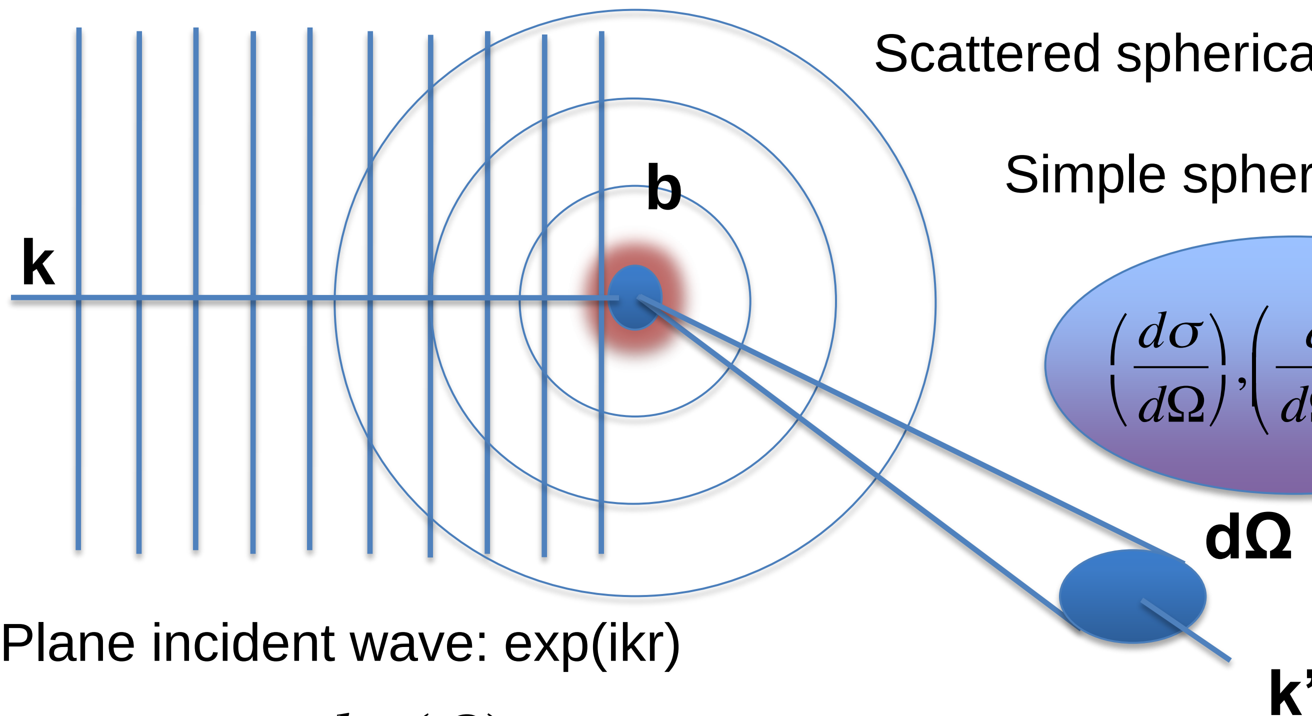
Interaction

SANS: Scattering of plane wave

σ_{tot}=number of neutrons scattered in all directions per sec/incident flux

Scattered spherical wave: –b exp(ikr)/b

Simple spherical object!!



$$\left(\frac{d\sigma}{d\Omega}\right), \left(\frac{d^2\sigma}{d\Omega\, dE}\right)$$

$$s_{tot} = 4pb^2$$

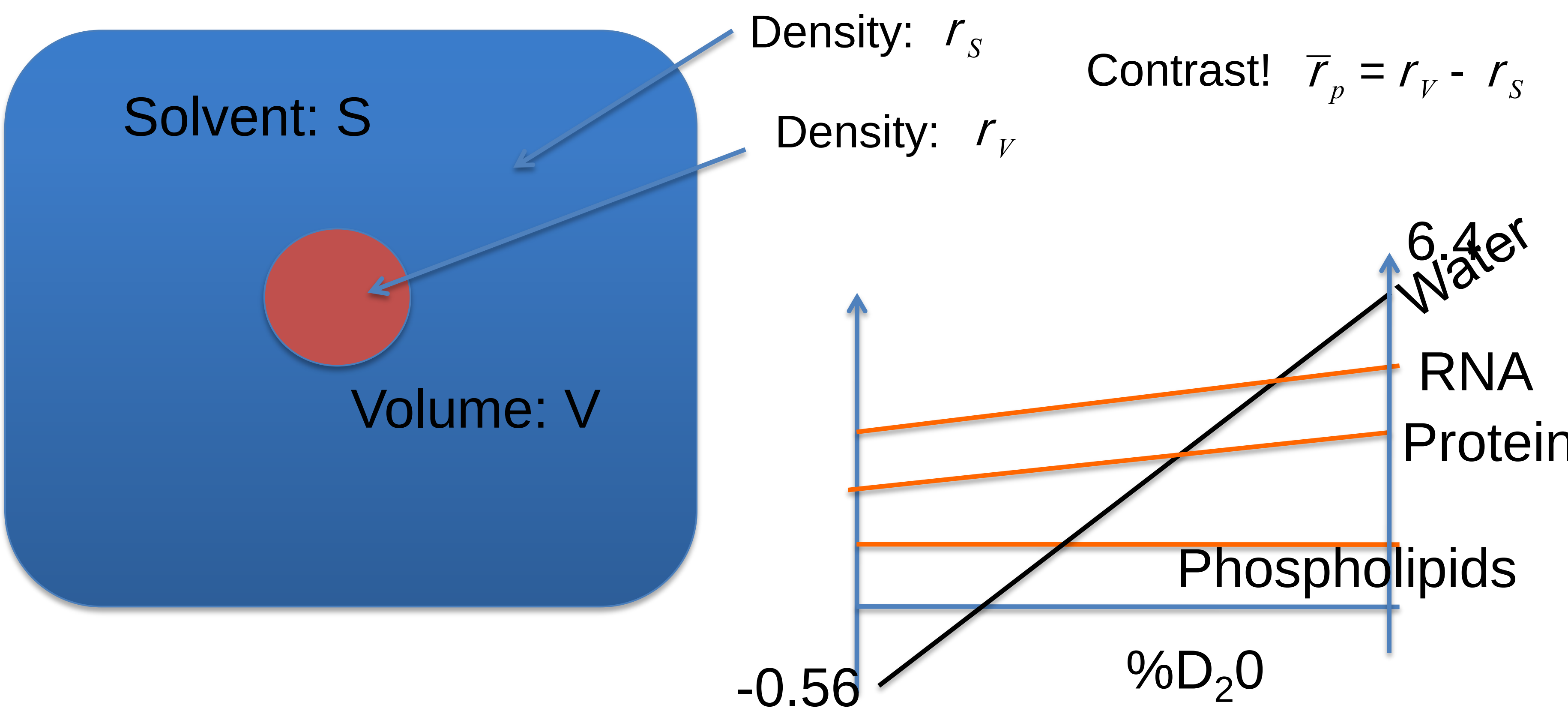
Plane incident wave: exp(ikr)

$$\frac{d\sigma(Q)}{d\Omega} = N_p V_p \overline{\rho} P(Q) S(Q)$$

Volume fraction

Contrast

SANS: Particles: contrast!

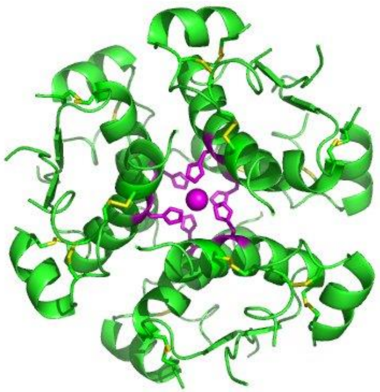


SANS: quick example

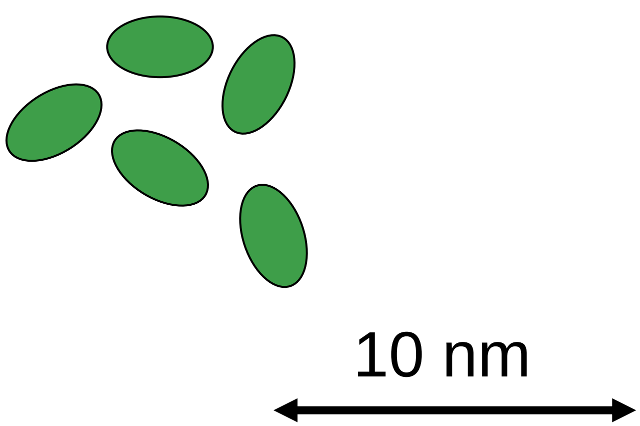
Protein based drugs:

- Typically proteins in solution to be injected
- Long shelf life (up to 2 years)
- Control of release profile is desirable

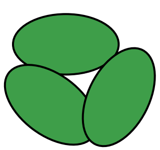
Insulin Hexamer



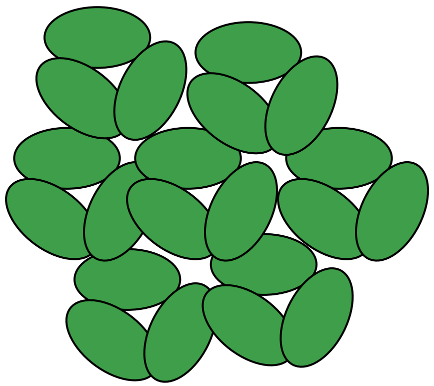
Fast action: Monomeric and dimeric insulin



Medium action: Hexameric insulin



Slow action: Large complexes of hexameric insulin

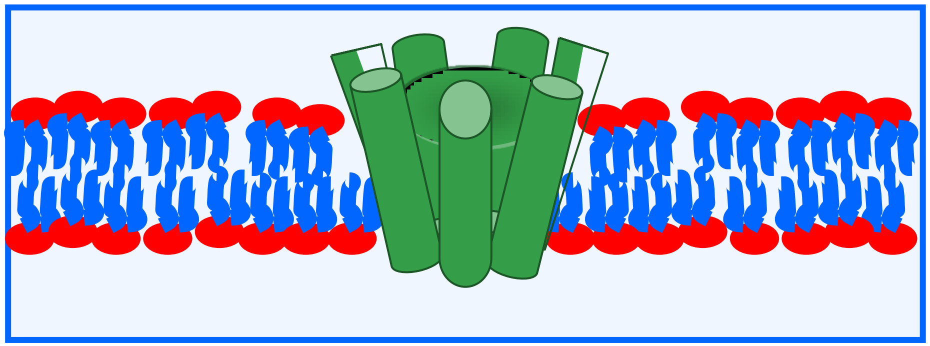


Knowledge and control of solution properties of the proteins are crucial

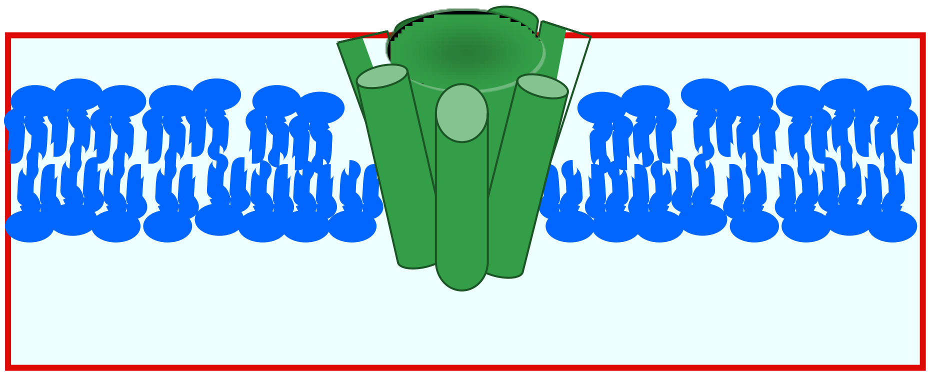
Source: L Arleth, Uni Copenhagen

SANS versus SAXS

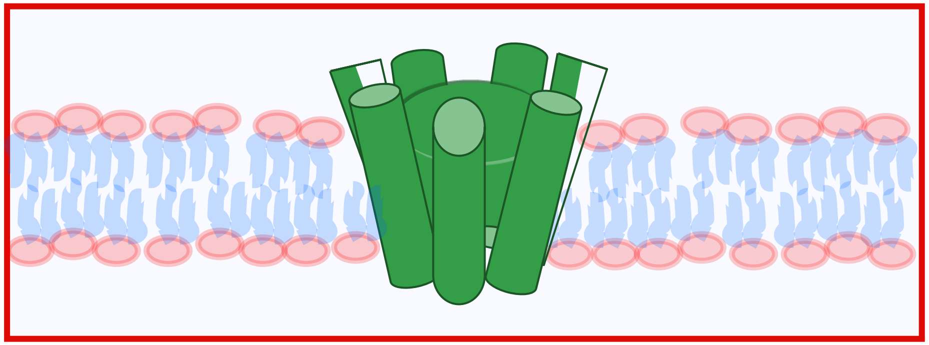
SAXS contrast



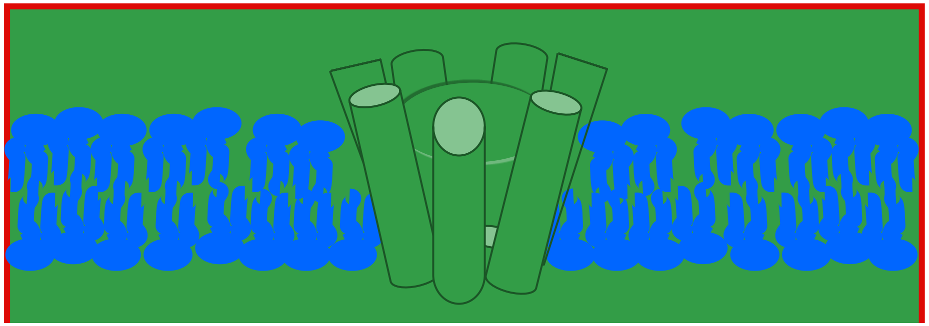
SANS contrast 1



SANS contrast 2



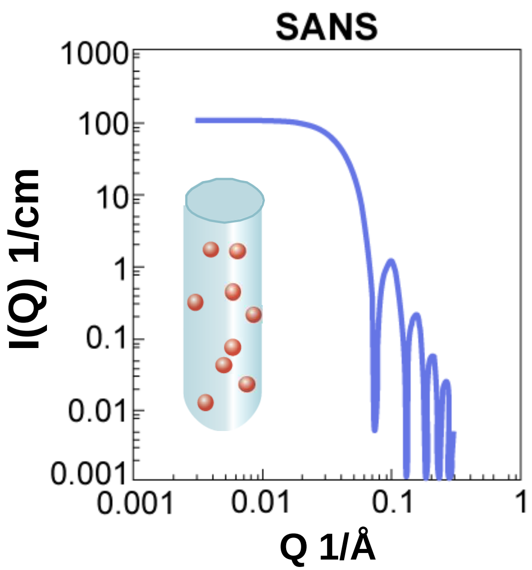
SANS contrast 3



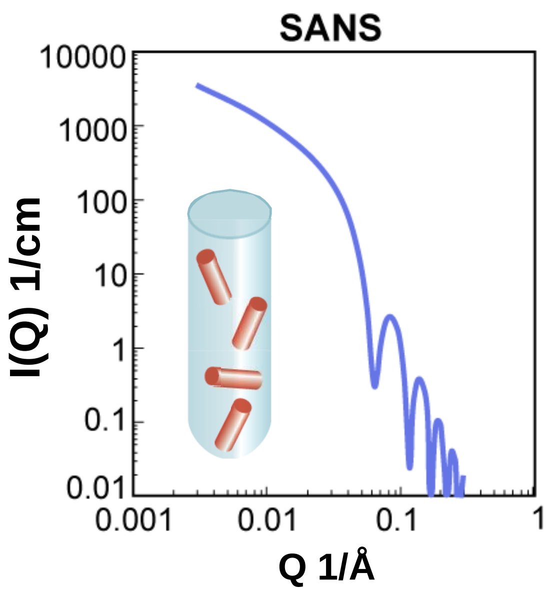
SANS gives the possibility of **not** seing everything at the same time….

SANS: Different Shapes

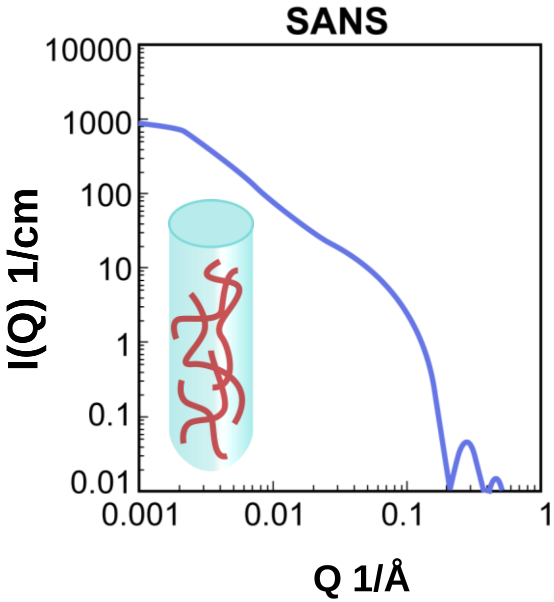
- Spheres:
  R = 60 Å



Rods: R = 60 Å
L = 1200 Å

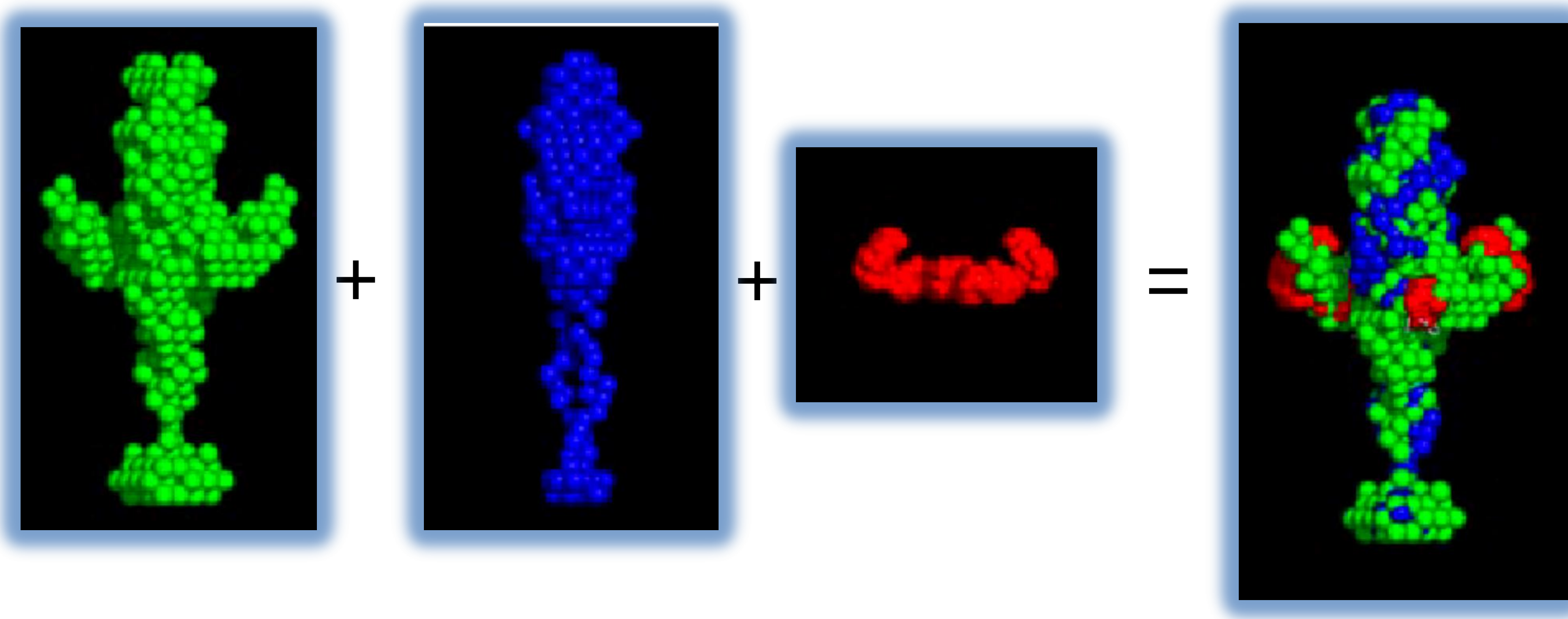


Worms: R = 18 Å
L = 5000 Å,
Kuhn Length = 300 Å



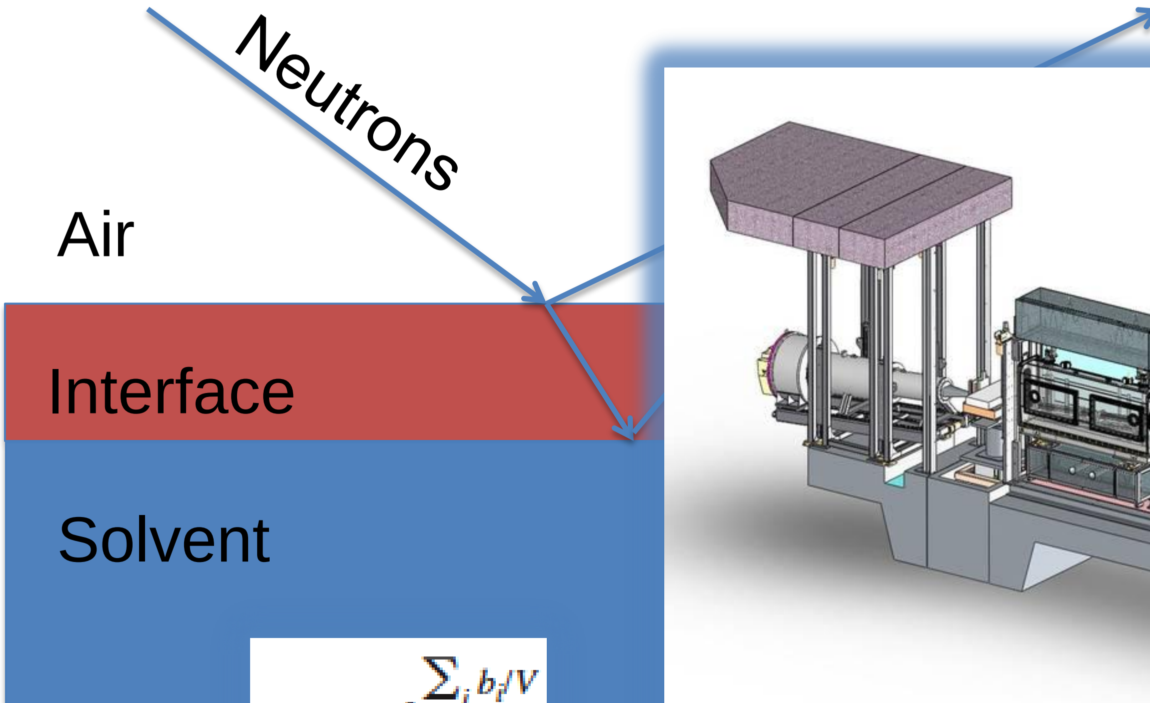
SANS: Selective Deuteration

sensitivity and selectivity
isotopic substitution/contrast variation

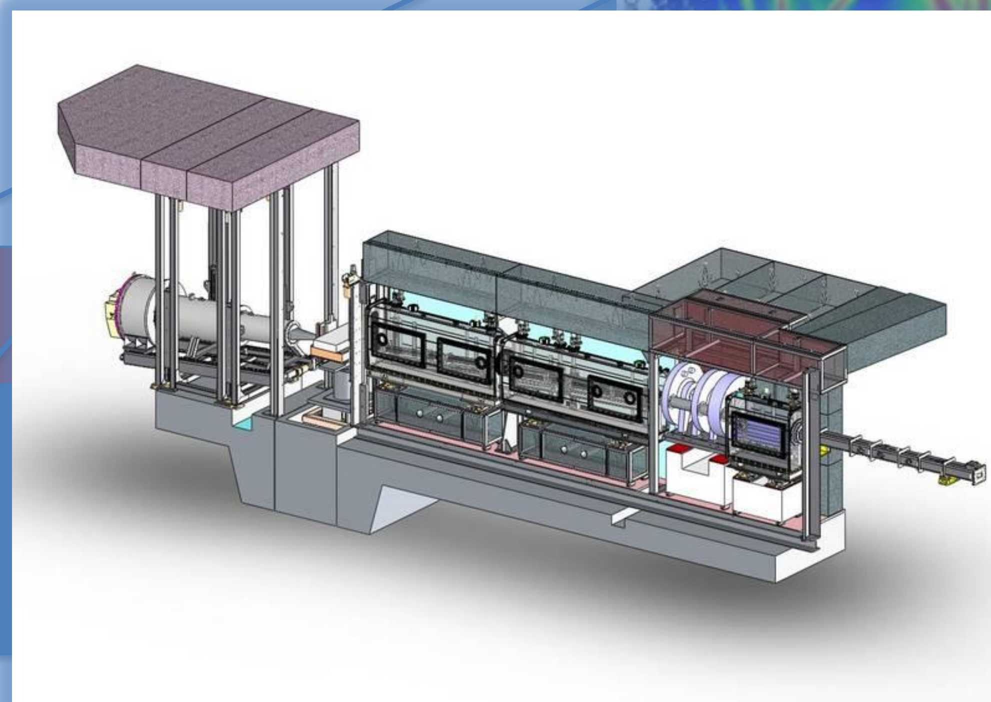


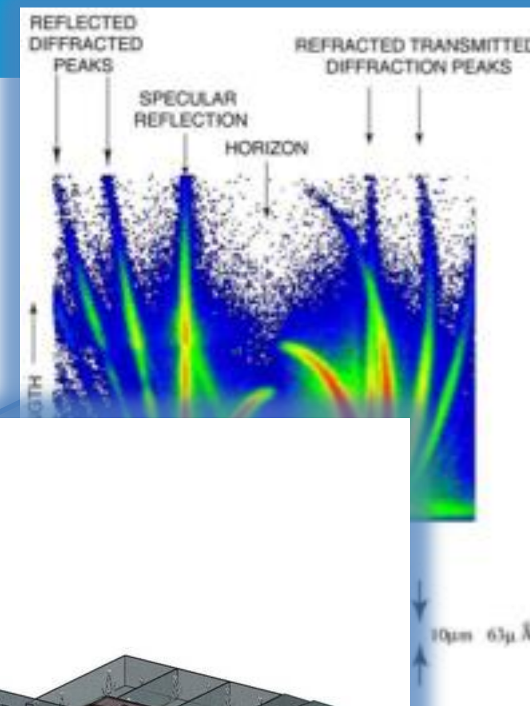
Neutron Reflectivity

- Basic principle



$$n = 1 - \lambda^2 \frac{\sum_i b_i/V}{2\pi}$$





Neutron Reflectivity

Neutron reflectometry to investigate the delivery of lipids and DNA to interfaces (Review)

Tommy Nylander
Physical Chemistry 1, Lund University, Box 124, SE-221 00 Lund, Sweden

Richard A. Campbell
Institut Laue-Langevin, BP 156, 38042 Grenoble Cedex 9, France

Pauline Vandoolaeghe
Physical Chemistry 1, Lund University, Box 124, SE-221 00 Lund, Sweden

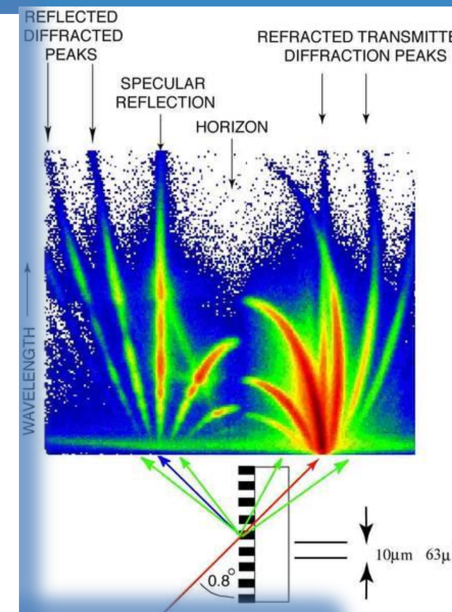
Marité Cárdenas
Biomedical Technology Laboratory, Faculty of Health and Society, Malmö University, SE-205 06 Malmö, Sweden

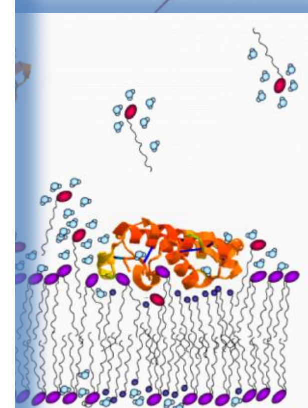
Per Linse
Physical Chemistry 1, Lund University, Box 124, SE-221 00 Lund, Sweden

Adrian R. Rennie
Department of Physics, Uppsala University, Box 530, SE-751 21 Uppsala, Sweden

(Received 6 May 2008; accepted 1 August 2008; published 19 December 2008)

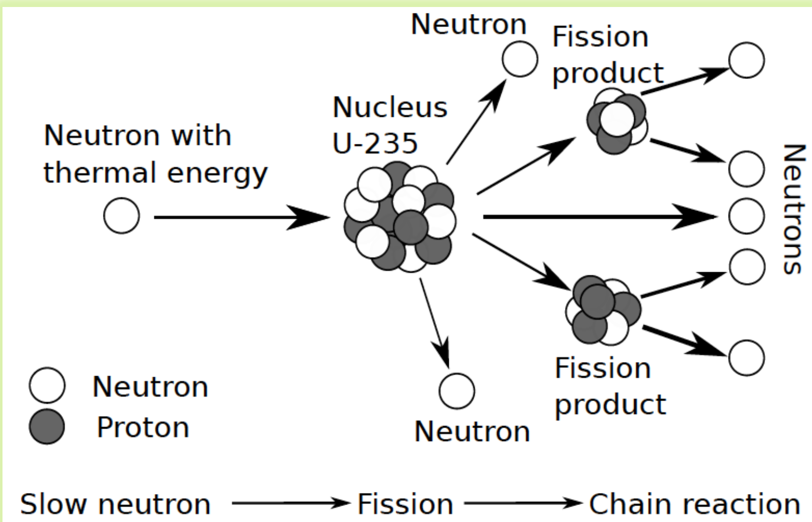
The application of scattering methods in the study of biological and biomedical problems is a field of research that is currently experiencing fast growth. In particular, neutron reflectometry (NR) is a technique that is becoming progressively more widespread, as indicated by the current commissioning of several new reflectometers worldwide. NR is valuable for the characterization of biomolecules at interfaces due to its capability to provide quantitative structural and compositional information on relevant molecular length scales. Recent years have seen an increasing number of applications of NR to problems related to drug and gene delivery. We start our review by summarizing the experimental methodology of the technique with reference to the description of biological liquid interfaces. Various methods for the interpretation of data are then discussed, including a new approach based on the lattice mean-field theory to help characterize stimulus-responsive surfaces relevant to drug delivery function. Recent progress in the subject area is reviewed in terms of NR studies relevant to the delivery of lipids and DNA to surfaces. Lastly, we discuss two case studies to exemplify practical features of NR that are exploited in combination with complementary techniques. The first case concerns the interactions of lipid-based cubic phase nanoparticles with model membranes (a drug delivery application), and the second case concerns DNA compaction at surfaces and in the bulk solution (a gene delivery application). © *2008 American Vacuum Society.* [DOI: 10.1116/1.2976448]





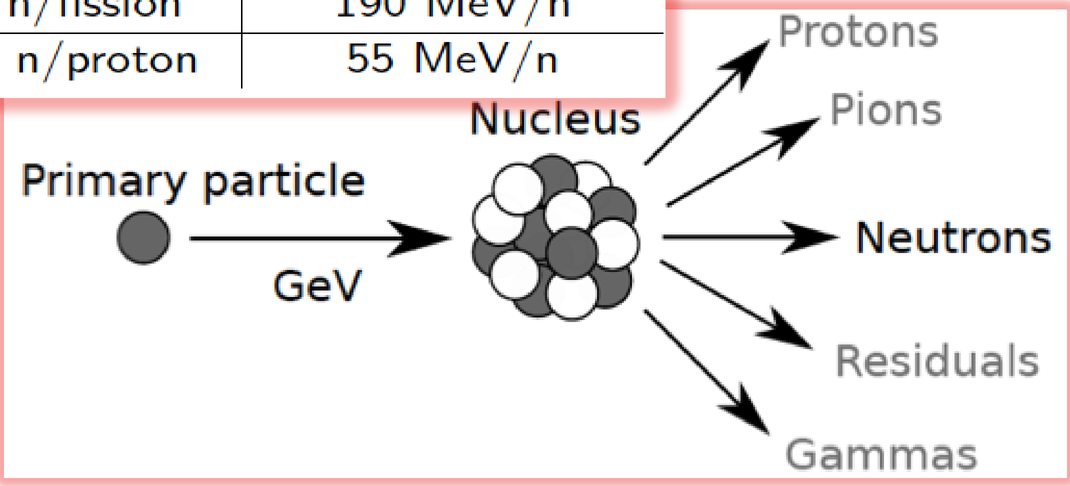
Fission and Spallation

Fission and Spallation

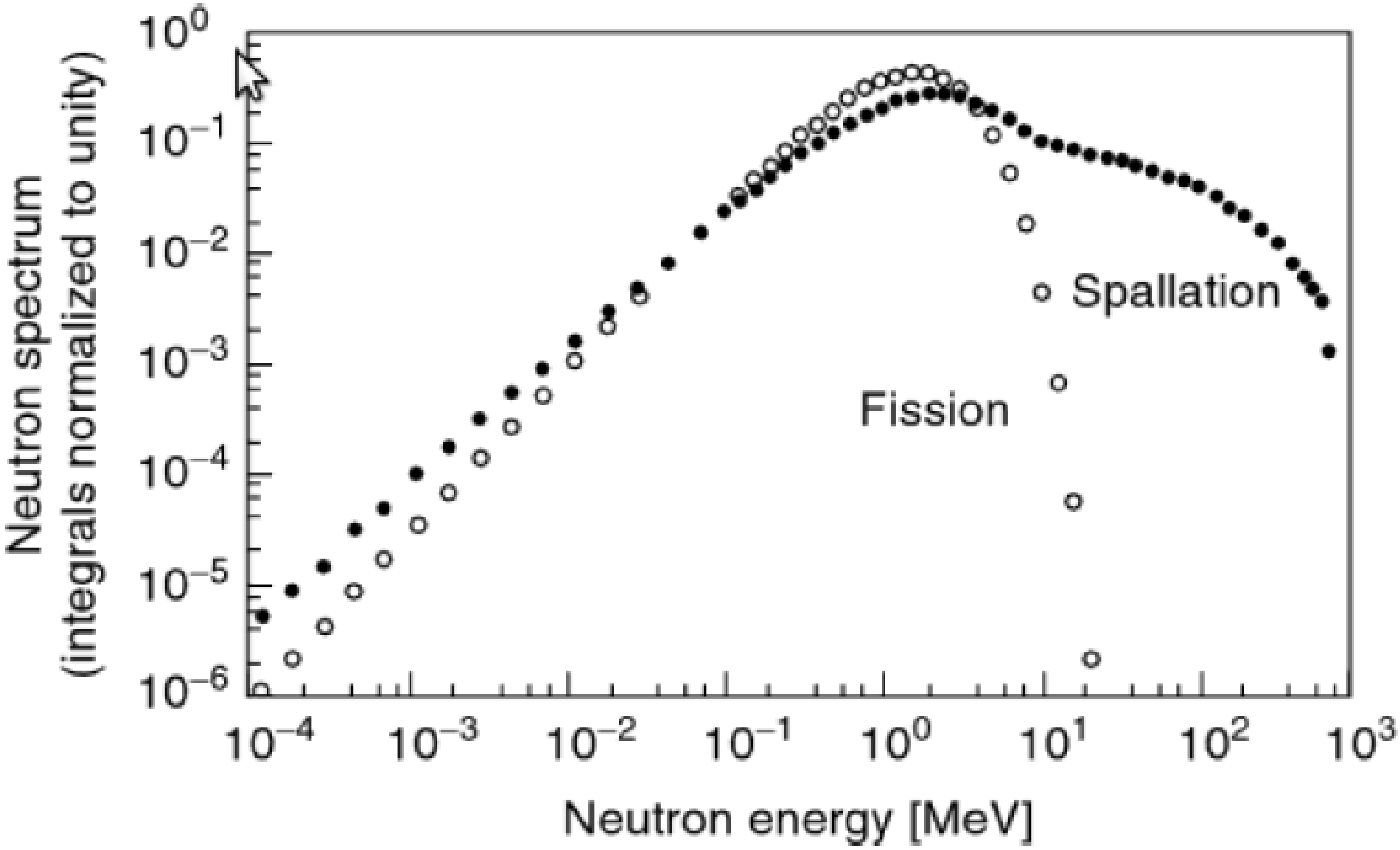


Spallation is a non-elastic nuclear interaction induced by a high-energy particle producing numerous secondary particles

Process	Reaction	Neutron yield	Energy deposition
Fission	$^{235}U(n,f)$	3 n/fission	190 MeV/n
Spallation	$p\ 1\,GeV \rightarrow Hg$	30 n/proton	55 MeV/n



Fission and Spallation



[D. Filges and F. Goldenbaum, Handbook of Spallation Research]

Fission and Spallation

Energy efficiency is key for high intensity neutron beam production

Fast neutrons produced / joule heat deposited in target station

Fission reactors:	~ 10^9	(in ~ 50 liter volume)
Spallation:	~ 10^{10}	(in ~ 2 liter volume)
Fusion:	~$1.5x10^{10}$	(in ~ 2 liter volume) (but neutron slowing down efficiency reduced by ~20 times)
Photo neutrons:	~ 10^9	(in ~ 0.01 liter volume)
Nuclear reaction (p, Be):	~ 10^8	(in ~ 0.001 liter volume)
Laser induced fusion:	~ 10^4	(in ~ 10^{-9} liter volume)

Spallation: most favourable for the foreseeable future

Spallation Process

Spallation Neutron Yield (i.e. multiplicity of emitted neutrons)

determines the requirement in terms of the accelerator power (current and energy of incident proton beam).

Spallation Neutron Spectrum (i.e. energy distribution of emitted neutrons)

determines the damage and activation of the structural materials (design of the beam window and spallation target)

Spallation Product Distributions

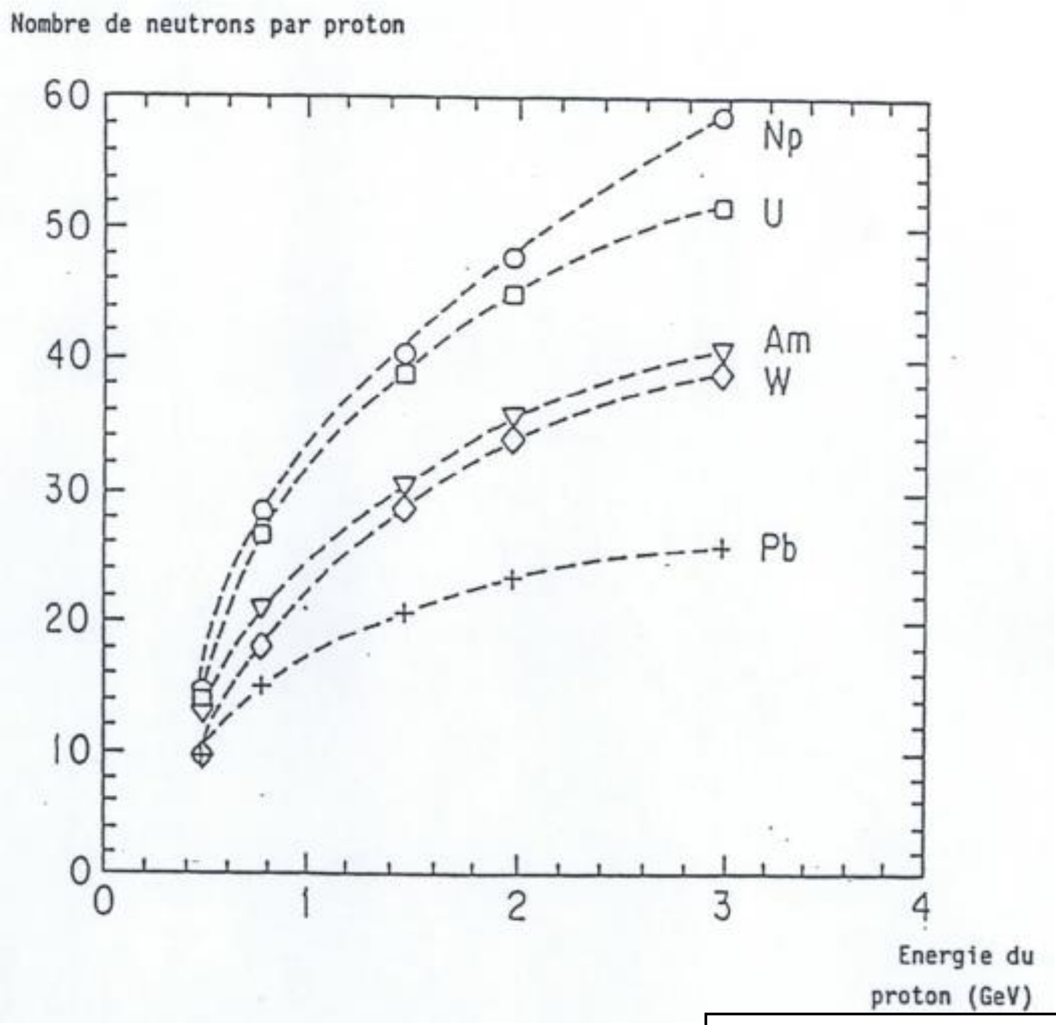
determines the radiotoxicity of the residues (radioprotection requirements).

Energy Deposition

determines the thermal-hydraulic requirements (cooling capabilities and nature of the spallation target).

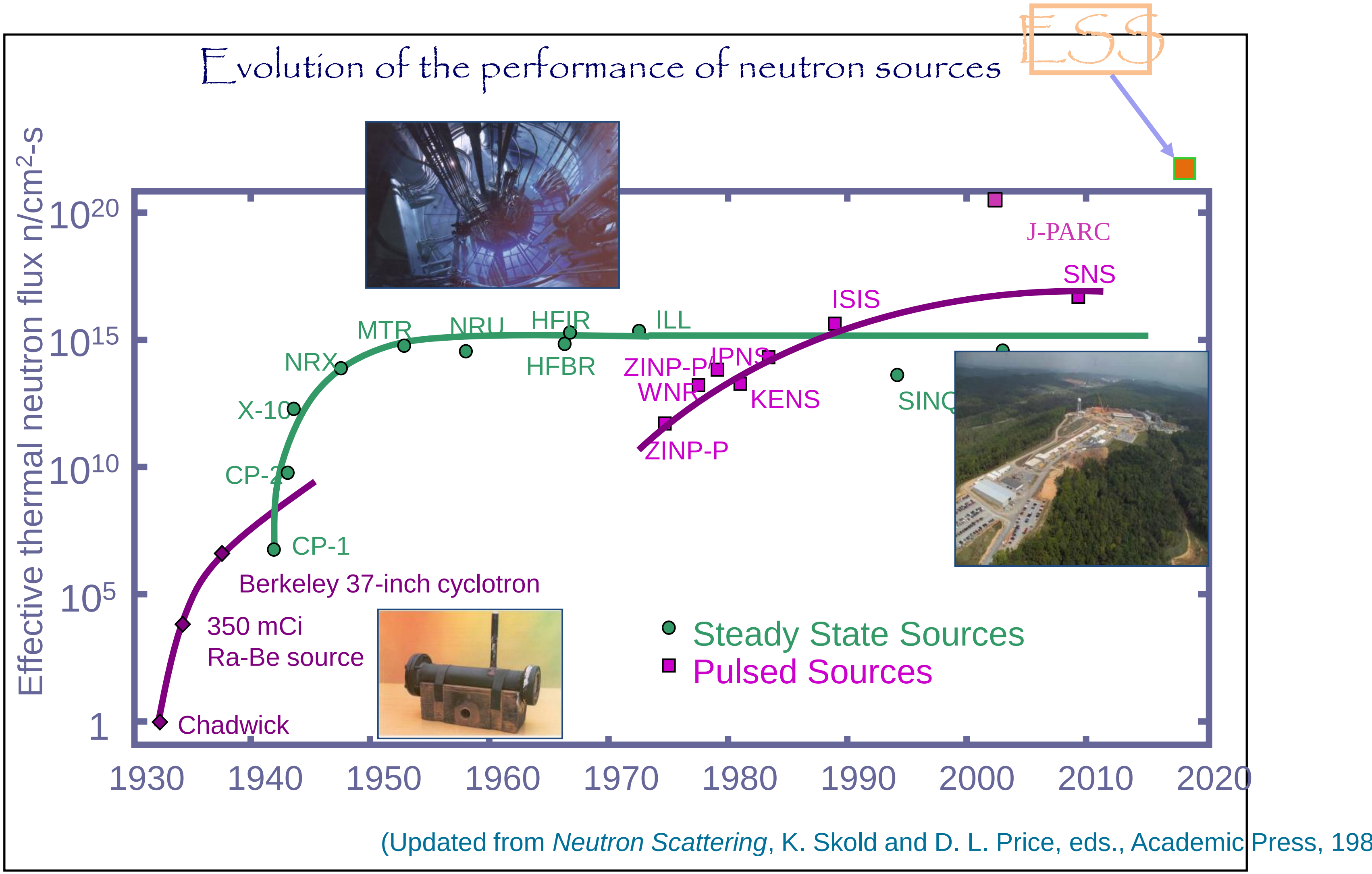
➔ **Sub-critical Reaction**

Spallation : choice of target materials



Y(E,A)=0.1 [A+20] [E (GeV) -0.12)] n/p

High time average and peak flux



Outline

- Neutrons properties and their interactions

- How to generate intense neutron beams using high power proton linear accelerator: The example of the ESS

for further reading

- Applications using Neutrons

ESS: Materials, Life Science and Society

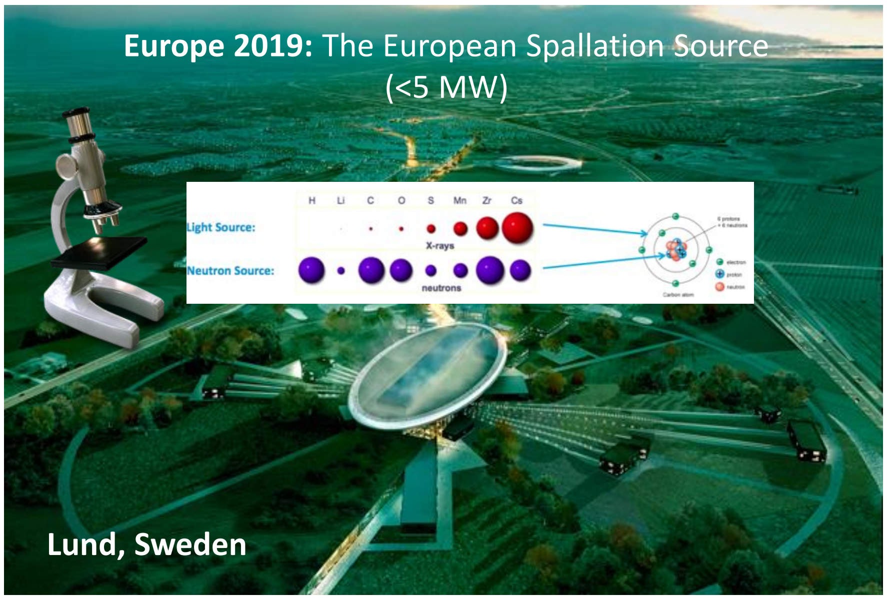
EU Horizon 2020 – strategy

The structure which the EC proposed consists of three basic priorities:

1. **Excellent Science**
2. **Industrial Leadership**
3. **Societal Challenges**

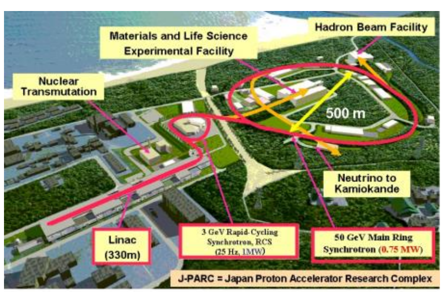
Spallation Sources

Philosophie de "Pré-vert": Greenfield



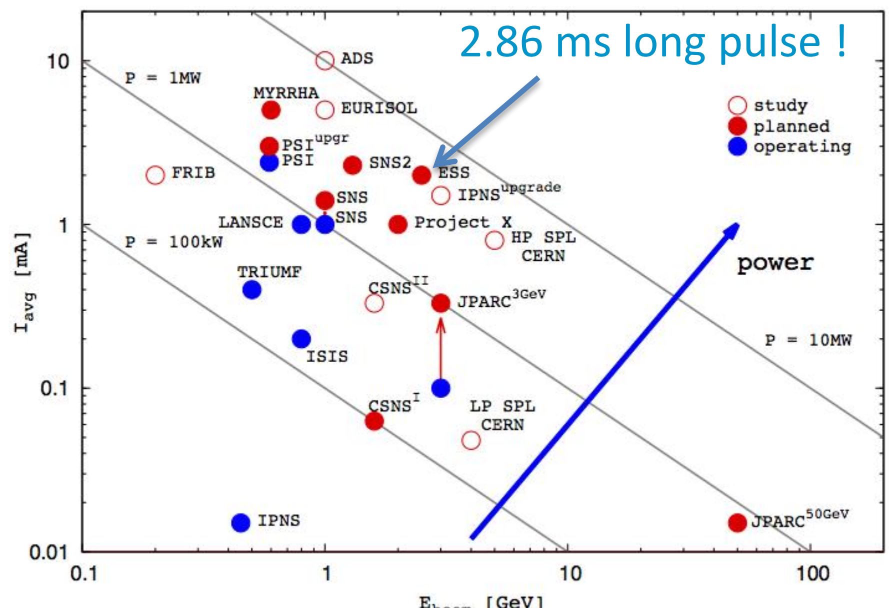
- Will bring new insights to the grand challenges of science and innovation
- Collaborative project: more than 17 countries
- 2014: Start of construction phase of the world's most powerful linear proton accelerator
- 2019: Provide the world's most advanced tools for studying materials with neutrons

(~ 450 employees; > 2500 users / year)



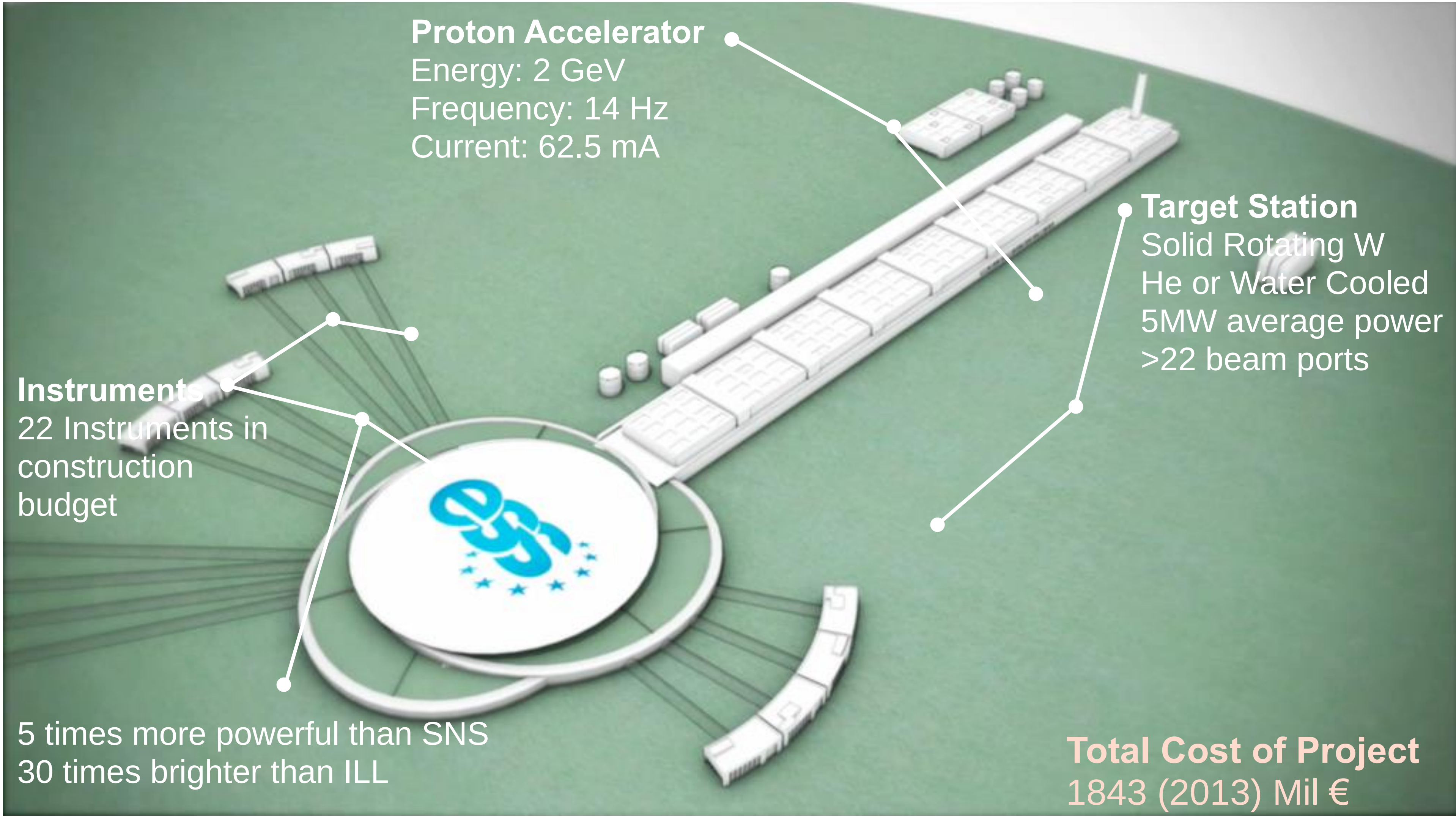
Japan 2008:
JPARC (<1MW)

USA 2006:
SNS (<1.4 MW)
1 GeV, 26 mA in linac, 627 ns long pulse, 60 Hz



Helicopter view of ESS

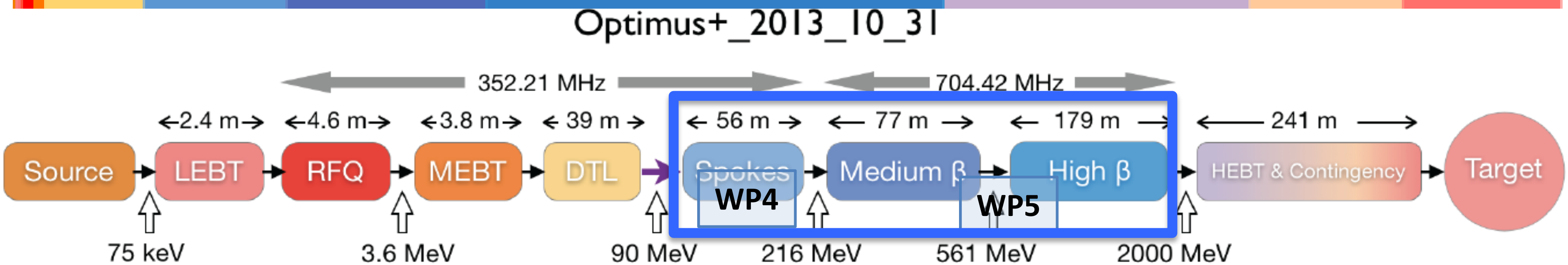
EUROPEAN SPALLATION SOURCE



Collaborative projects

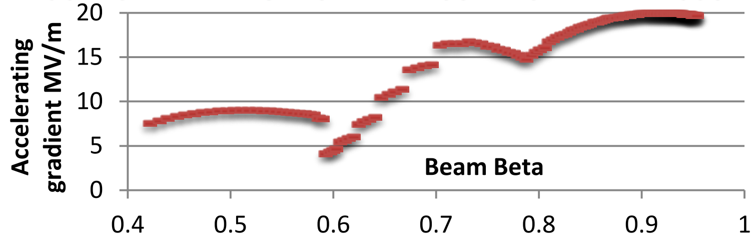
- ESS is an emerging research laboratory with (still) very limited capacity in-house
- Two possibilities:
  - Limit the scope of the project so that it can be done with in-house resources
  - Work in a collaboration where the scope of the project can be set by the total capacity (distributed) of the partners
- The accelerator part of the project well suited for this as this community has a strong tradition of open collaboration (e.g. E-XFEL, FAIR, LHC, European Commission Framework Programs and design studies)
- To keep cost down and to optimize schedule this requires that investments in required infrastructure is done at the partner with best capacity to deliver

Linac redesign to meet ESS cost objective



Beam power (MW)	5
Beam current (mA)	62.5
Linac energy (GeV)	2
Beam pulse length (ms)	2.86
Repetition rate (Hz)	14

	Num. of CMs	Num. of cavities
Spoke	13	26
Medium β (6-cell)	9	36
High β (5-cell)	21	84



Style	Spoke	Medium-β	High-β
Freq. (MHz)	352.21	704.42	704.42
Cavity #	26	36	84
Velocity range	0.42 to 0.58	0.58 to 0.78	0.78 to 0.95
Nom. Acc. Voltage (MV)	5.74	14.3	18.2
Loaded quality factor	2.85×10^5	8×10^5	7.6×10^5

[SRF2013 – "The ESS Superconducting Linear Accelerator", C. Darve, M. Eshraqi, M. Lindroos, D. McGinnis, S. Molloy, P. Bosland and S. Bousson]

Accelerator collaborations for ESS "start-up"

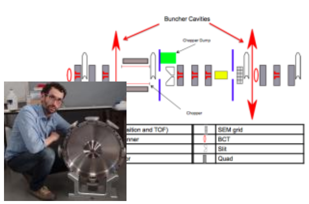
EUROPEAN SPALLATION SOURCE

WP4 : Sebastien Bousson

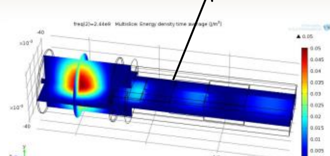
WP5 : Pierre Bosland

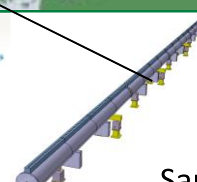
CERN

Roger Barlow

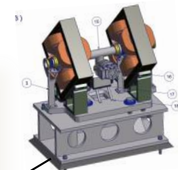


Ibon Bustinduy

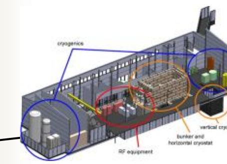




Santo Gammino

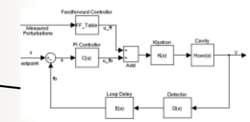


Søren Pape Møller



UPPSALA UNIVERSITET

Roger Ruber



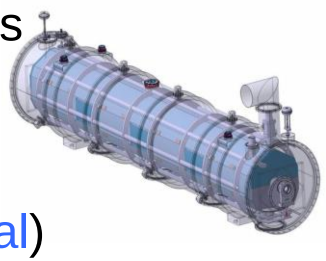
LUNDS UNIVERSITET

Anders J Johansson

The National Center for Nuclear Research, Swierk

Cavity Cryomodule Technology Demonstrators

One full scale 704 MHz medium and high-beta cavity cryomodules
A staged approach towards the series industrialization and the ESS Linac tunnel installation



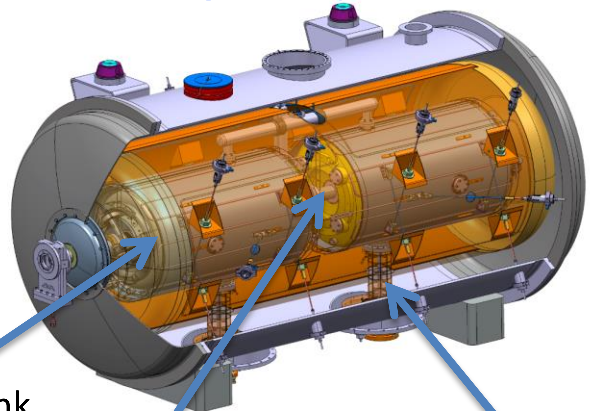
- Validate designs and construction capability of SRF components
- Prepare the industrialization process by validating component life-cycles (incl. assembling process, QA)
- Validate component performances (incl. RF, mechanical, thermal)
- Develop ESS 704 MHz SRF linac operating procedures
- Validate control command strategy (Control box, PLC, EPICS, LLRF)
- Validate ESS integration and interfaces with RF, cryogenics, vacuum and control systems
- Train people in ESS SRF Technology and build an ESS collaboration

→ **Similar process for the spoke cryomodules**

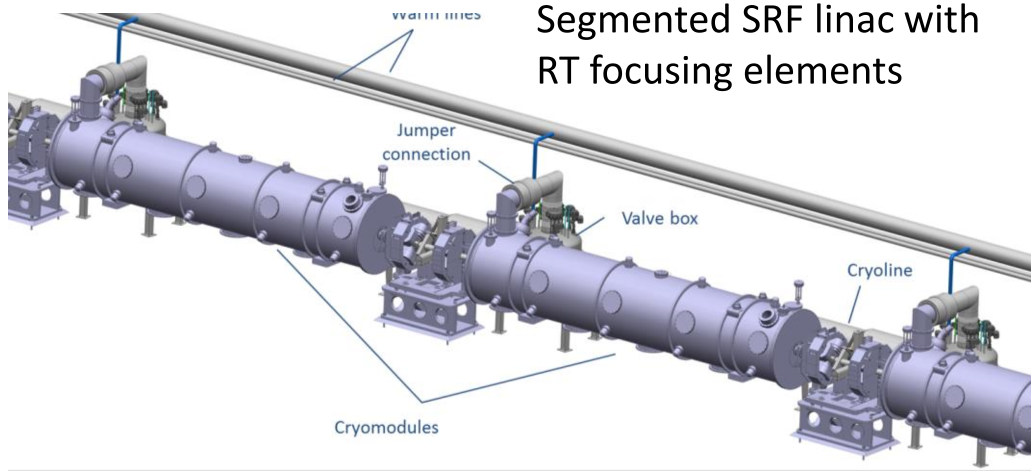
ESS SRF Cavity Cryomodules

EUROPEAN SPALLATION SOURCE

2 Spoke Cavities per Cryomodule



Segmented SRF linac with RT focusing elements

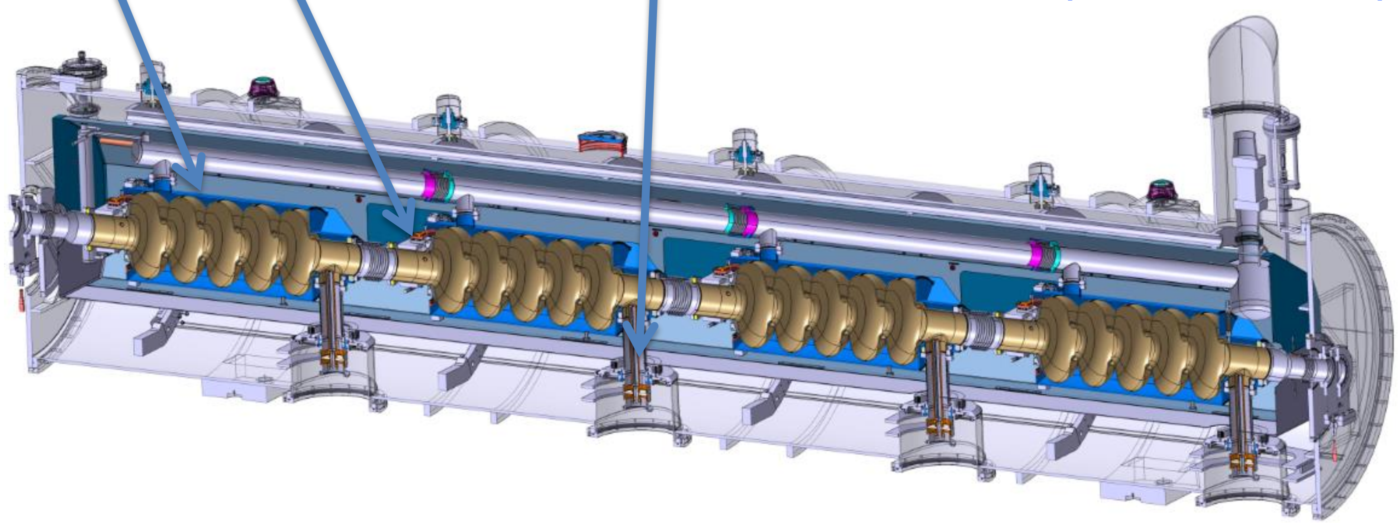


Ti. Helium tank

Cold tuning System

Power coupler

4 Elliptical Cavities per Cryomodule



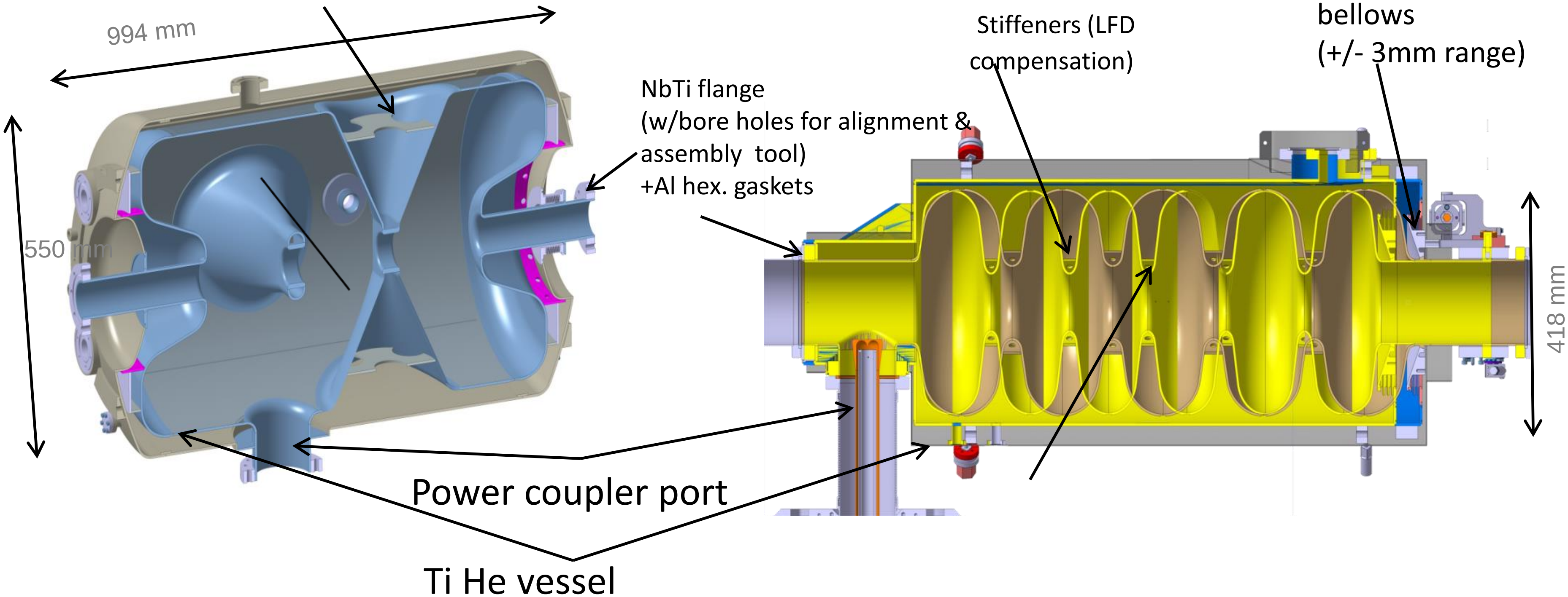
SRF Cavities Development

Spoke cavity

→ Stiffeners on the Spoke bars (vacuum pressure)

Elliptical cavities

→ No HOM power couplers



Medium beta:
- **6 cells – beta=0.67**
- **Length 1259,40mm**

High beta:
- **5 cells – beta=0.86**
- **Length 1316,91mm**

ESS Requirements and RF Parameters

EUROPEAN SPALLATION SOURCE

Spoke cavities

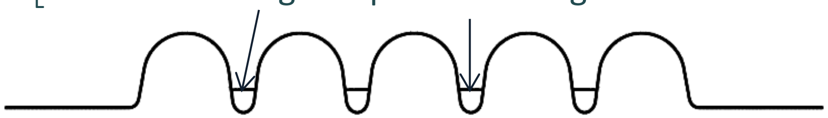
Frequency (MHz)	352,2
Optimum beta	0,50
Operating temperature (K)	2
Nominal Accelerating gradient (MV/m)	9
Lacc (βopt.x nb gaps x λ/2) (m)	0,639
Bpk (mT)	79 (max)
Epk (MV/m)	39 (max)
Bpk/Eacc (mT/MV/m)	<8,75
Epk/Eacc	<4,38
Beam tube diameter (mm)	50
RF peak power (kW)	335
G (Ω)	130
Max R/Q (W)	427
Qext	$2{,}85\ 10^5$
Q0 at nominal gardient	$1{,}5\ 10^9$

Elliptical cavities

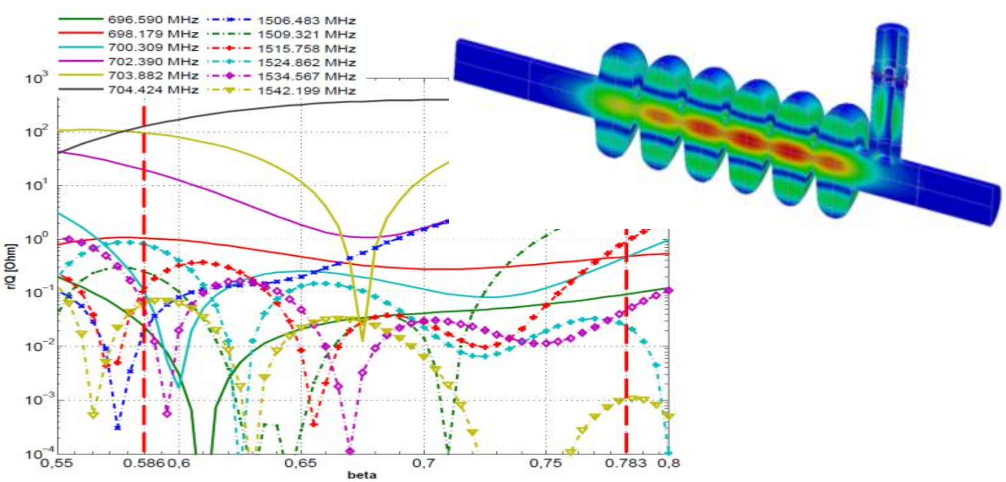
	Medium	High
Geometrical beta	0.67	0.86
Frequency (MHz)	704.42	
Number of cells	6	5
Operating temperature (K)	2	
Epk max (MV/m)	45	45
Nominal Accelerating gradient (MV/m)	16.7	19.9
Q_0 at nominal gradient	> 5e9	
Q_{ext}	$7.5\ 10^5$	$7.6\ 10^5$
Iris diameter (mm)	94	120
Cell to cell coupling k (%)	1.22	1.8
p,5p/6 (or 4p/5) mode sep. (MHz)	0.54	1.2
Epk/Eacc	2.36	2.2
Bpk/Eacc (mT/(MV/m))	4.79	4.3
Maximum. r/Q (W)	394	477
Optimum β	0.705	0.92
G (Ω)	196.63	241
RF peak power (kW)	1100	

Medium-β Elliptical Cavities

K_L reduction using compensation rings for medium and high-beta



Nominal wall thickness [mm]	3.6
Cavity stiffness Kcav [kN/mm]	2.59
Tuning sensitivity Df/Dz [kHz/mm]	197
K_L with fixed ends [Hz/(MV/m)²]	-0.36
K_L with free ends [Hz/(MV/m)²]	-8.9
Pressure sensitivity K_P [Hz/mbar] (fixed ends)	4.85

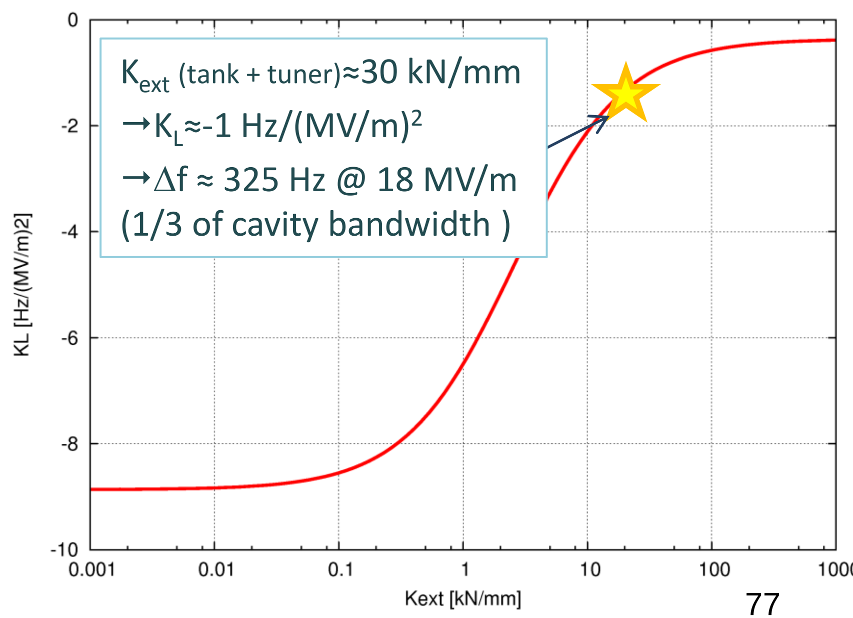


RF/mechanical design

Lorentz detuning

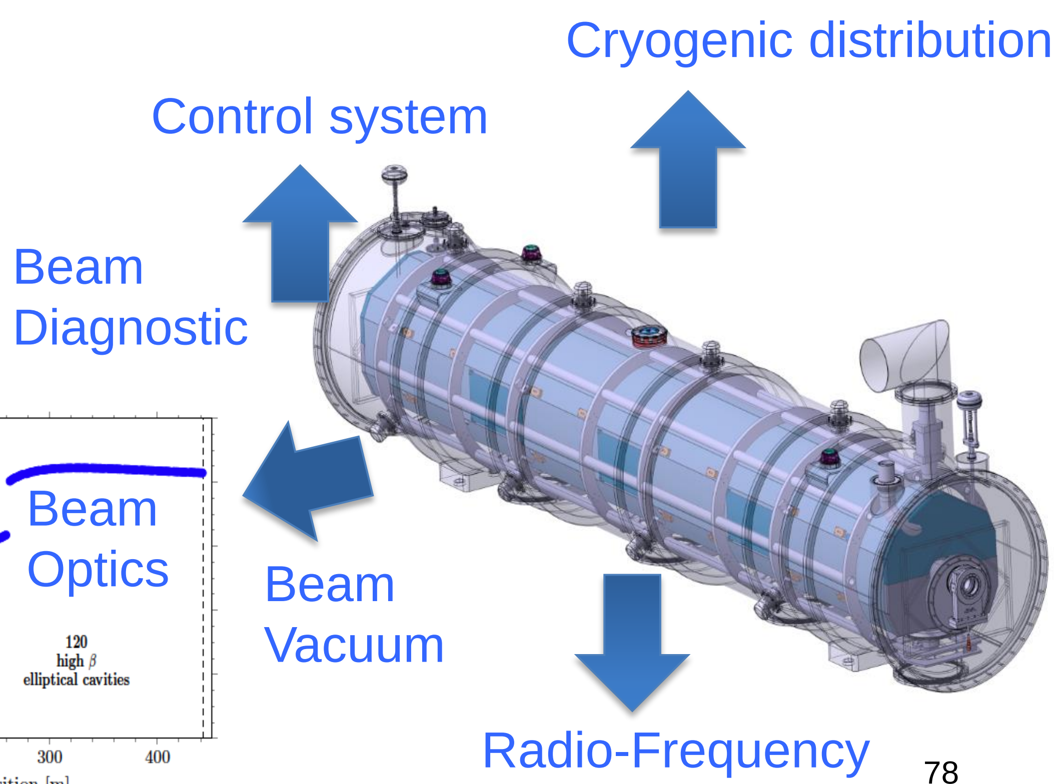
$$K_L = \Delta f / E_{acc}^2$$

$$K_L = K_{L\infty} + \frac{\Delta f}{\Delta z} \frac{\overrightarrow{F_\infty} \cdot \overrightarrow{u_z} / E_{acc}^2}{K_{ext} + K_{cav}}$$

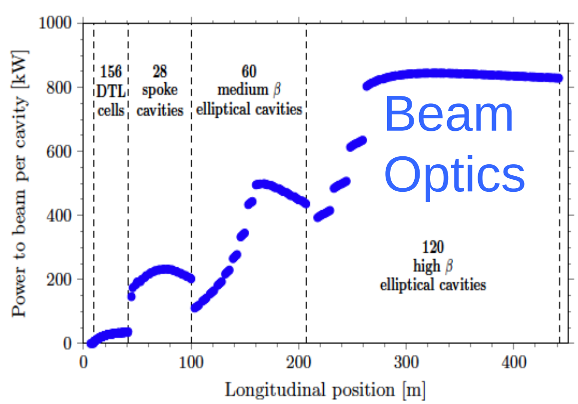


Cryomodule Interfaces

- Most AD internal Work Packages (beam optics, RF, cryo, vacuum, test stands, electrical, cooling, installation)
- External WPs cryomodule, cavity and designers and potential In-Kind collaborators
- Control command (Control Box, PLC, LLRF, MPS, EPICS)
- Data-logging ICS teams
- ESS ES&H
- Conventional Facility
- ESS system engineer, QA
- Survey experts
- Transport

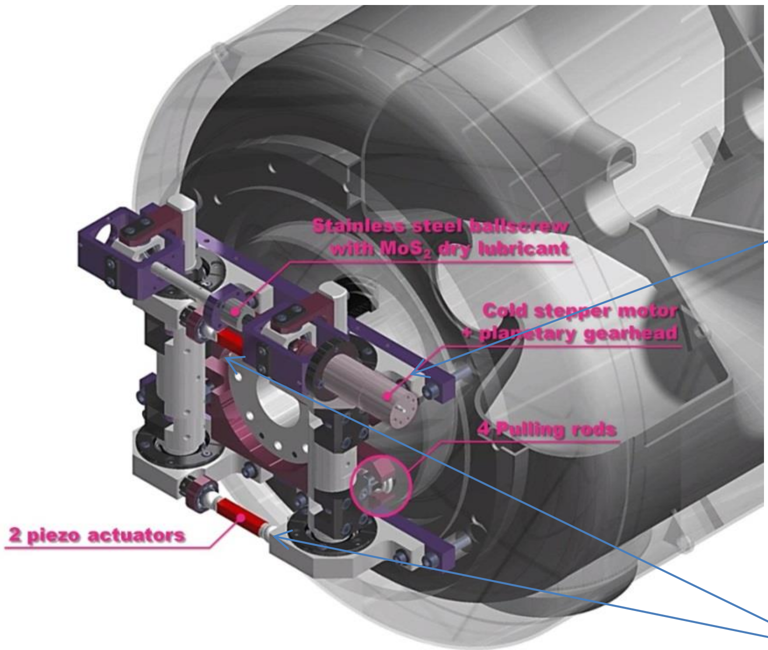


Previous Linac version for comparison ➔



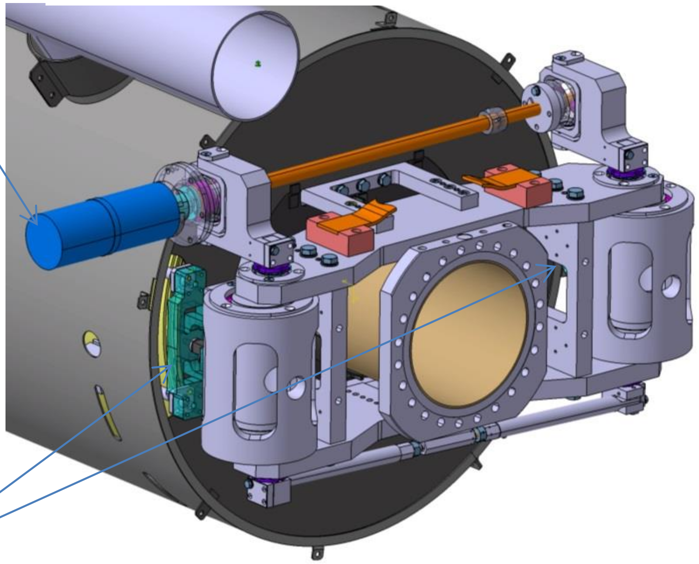
Cold Tuning System

Spoke CTS

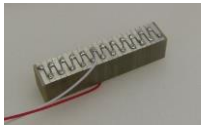


Elliptical CTS

Type V ; 5-cell prototype
+/- 3 mm range on cavity



Stepper motor and planetary gearbox (1/100e) at cold and in vacuum



2 piezo stacks

Slow tuner

Main purpose : Compensation of large frequency shifts with a low speed

Actuator used : Stepper motor

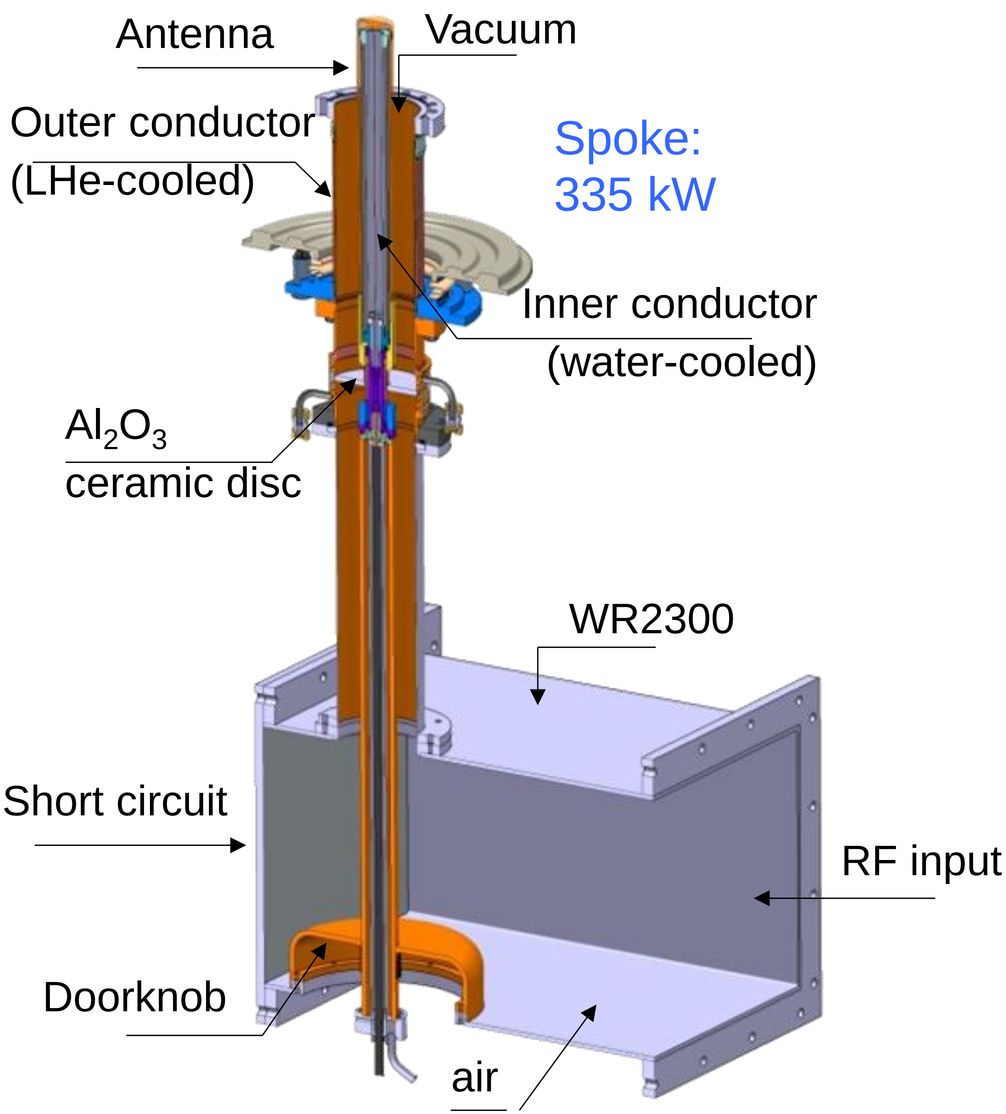
&

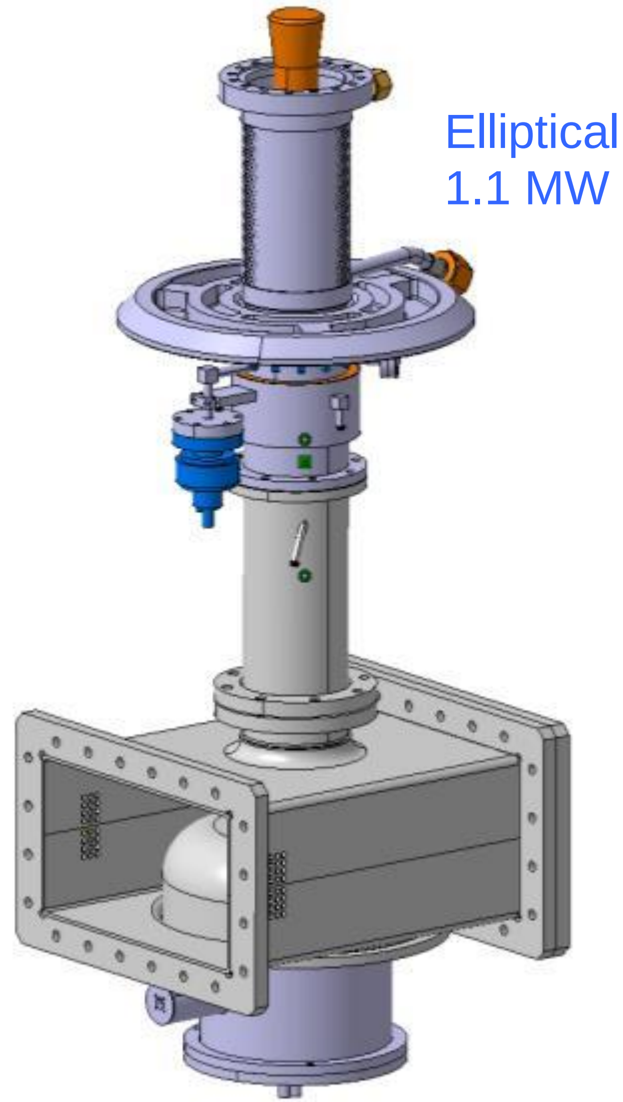
Fast tuner

Main purpose : Compensation of small frequency shifts with a high speed

Actuator used : Piezoelectric actuators

Fundamental Power Coupler

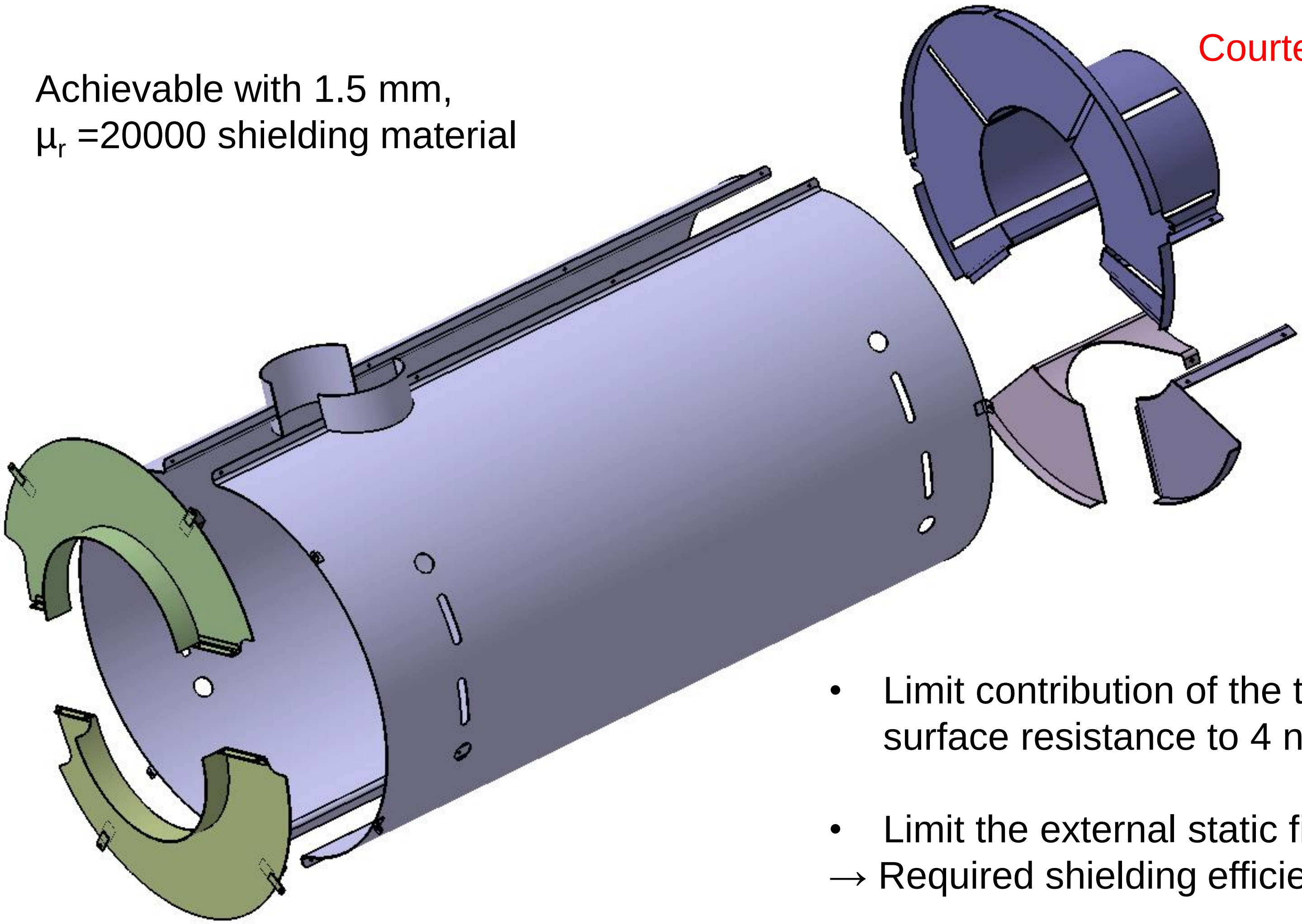




Magnetic shield, e.g. Elliptical cavity

Courtesy of J. Plouin/ CEA

Achievable with 1.5 mm,
μ_r =20000 shielding material

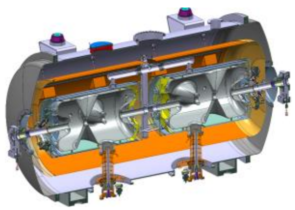


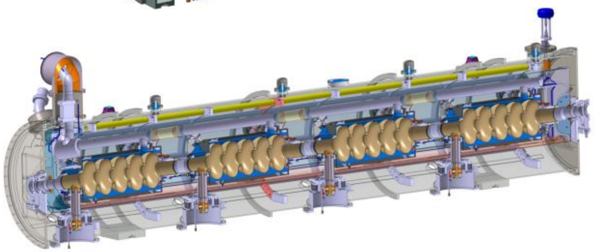
- Limit contribution of the trapped flux to the surface resistance to 4 nΩ

- Limit the external static field to Bext = 14 mG.

→ Required shielding efficiency equal to 35.

Cryomodule Heat Load Distribution





Per cryomodule

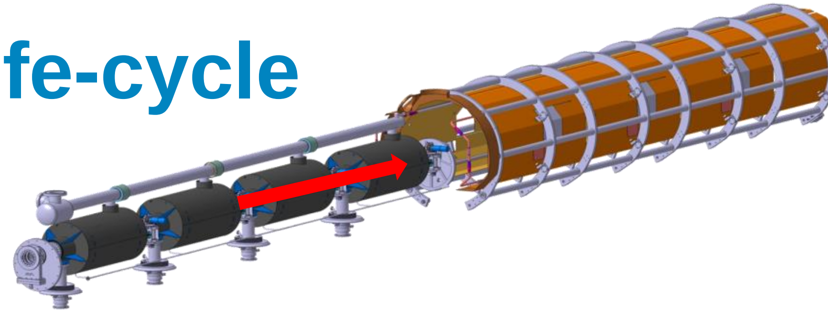
	Watts to 2 K							4.5 K Liquefaction (g/s)	Watts to ~50 K
	Static				Dynamic			Total	Total
	Others	Valves	Coupler	**Total**	Beam	Cavity	**Total**		
1 Spoke	3.3	0.2	3.5	**7**	1.5	5.0	**6.5**	0.092	30
1 MB	6.3	0.2	6.8	**13.3**	3.3	20	**23.3**	0.092	46.5
1 HB	6.3	0.2	6.8	**13.3**	3.3	24.4	**27.7**	0.092	46.5

Sum for the Linac cryoplant (incl. 14 extra HB for contingency space)

		Watts to 2K			4.5K Liquefaction (g/s)	Watts to ~ 50K
	Number of CMs	Static	Dynamic	Total	Total	Total
Spoke	13	91	84.5	175.5	1.196	390
Medium beta	9	119.7	209.7	329.4	0.828	418.5
High beta	35	472.5	627.9	1100.4	3.22	1627.5
Total	**57**	**683.2**	**922.1**	**1605.3**	**5.244**	**2436**

Heat load due to the beam losses deposit a maximum of 0.5 W/m to the Spoke, medium and high-beta cavities sections 2 K temperature levels.

Cryomodule life-cycle



Cavity fabrication

Chemical treatment

High pressure rinsing
In clean room (ISO5 or ISO4)

Validation test of the cavity in vertical cryostat

Qualified cavity storage

Power coupler fabrication

Assembling in clean room

Coupler RF processing

Processed couplers storage

Cryomodule components fabrication

Cavity string assembling in clean room

Tools fabrication

Cryomodule assembling

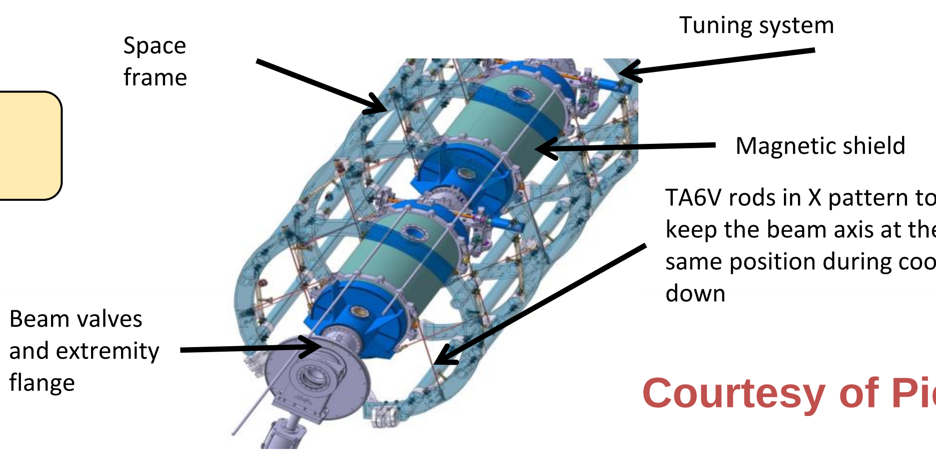
Transport

Cryomodule reception and storage

Validation test of the cryomodule

Cryomodule storage

Cryomodule on beam line



Courtesy of Pierre Bosland CEA/IRFU

Elliptical Cavity Preparation

High beta cavity fabrication (Zanon and RI)

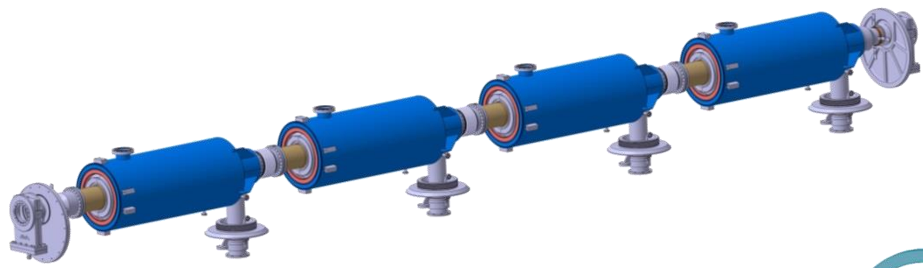
Vertical Electropolishing system@ CEA

Study of the tooling in progress @ CEA

Example of the tooling for the assembling of the coupler on the cavity in clean room

Elliptical Assembly Procedure

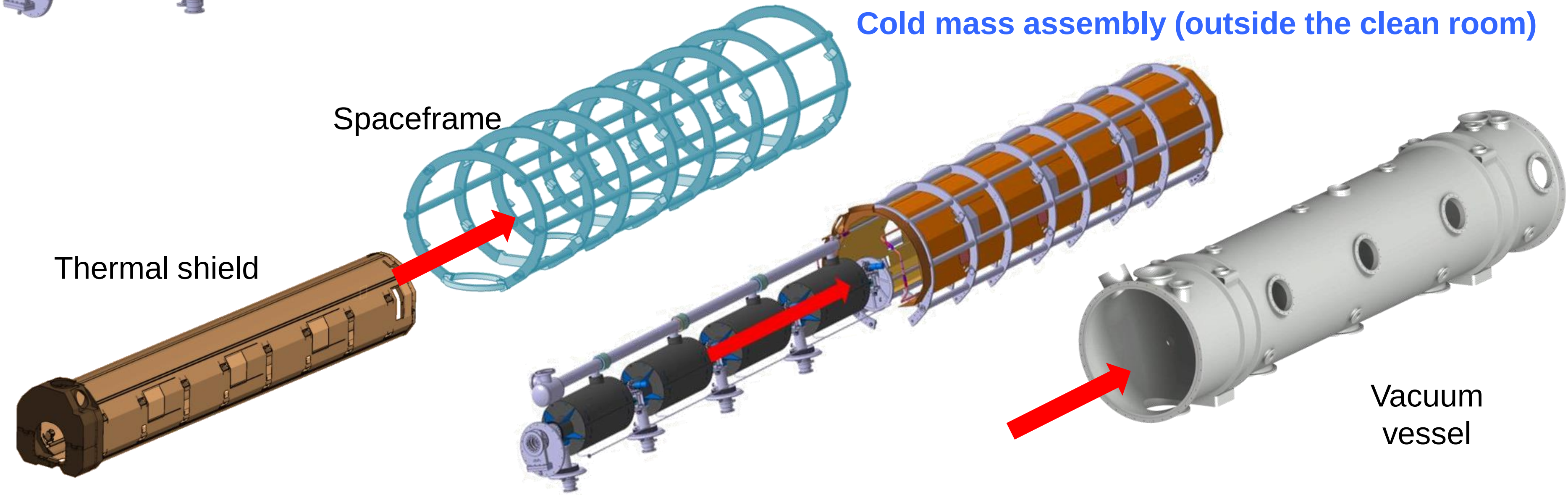
Cavity string assembly in clean room



Build on existing knowledge (SNS, XFEL)

- Develop Training and “Fabrication file”
- Pre-industrialization
- Industrialization

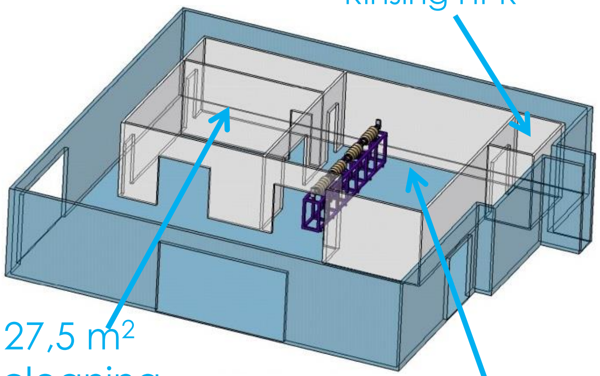
Cold mass assembly (outside the clean room)



Design concept of the tooling: most of parts will be used for both types of elliptical cryomodules

Infrastructure in Saclay

Clean room for the M-ECCTD
(and H-ECCTD)



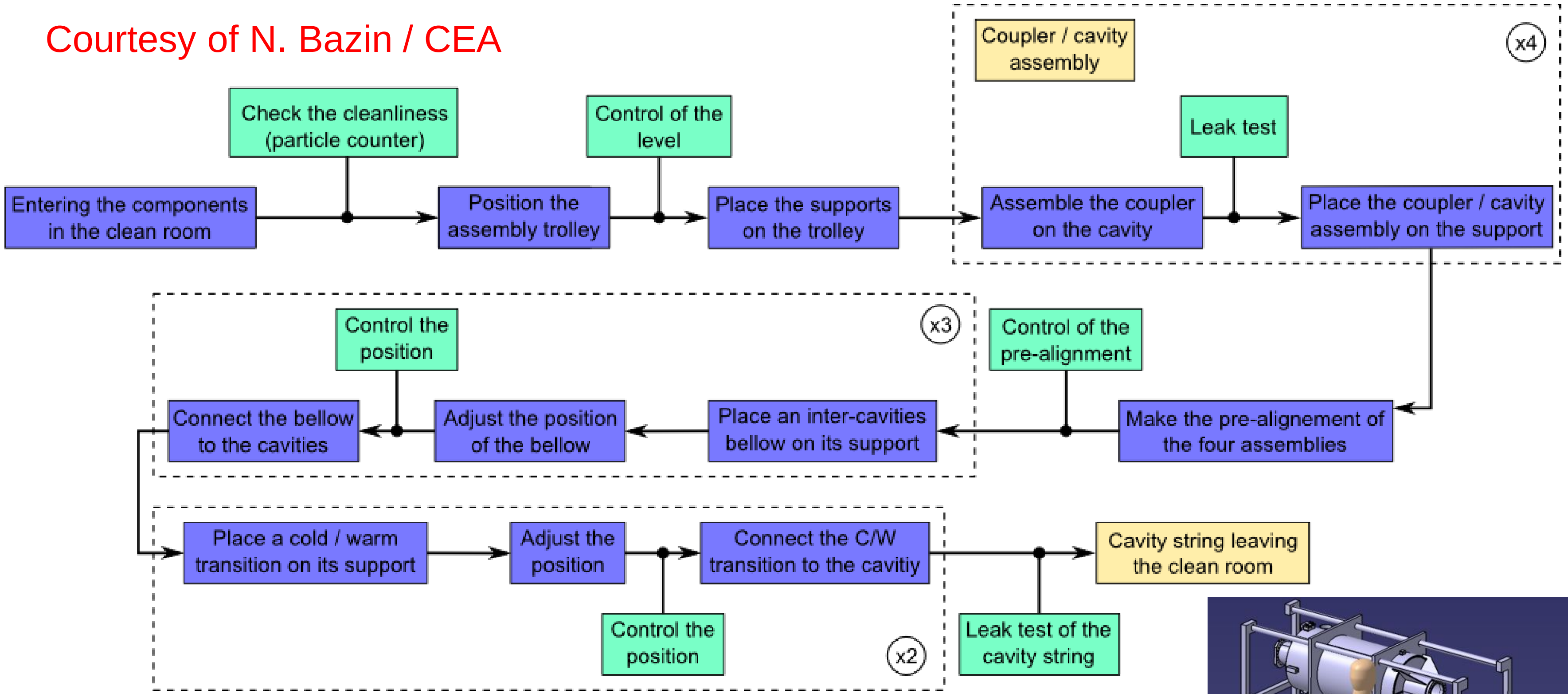
The clean room inauguration
→ May 13th 2014

Possible IKC for the assembly by industry at Saclay (XFEL cryomodules assembly)

- Uses the current infrastructure at Saclay
- Benefits from the experience of the XFEL cryomodule assembly (ALSYOM)

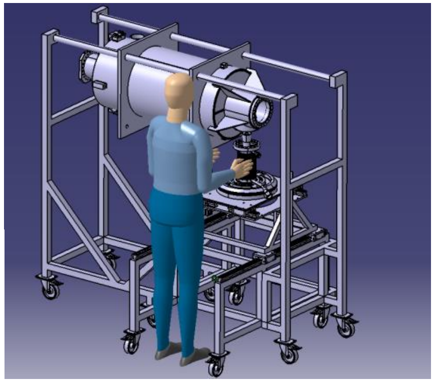
Assembly of elliptical cryomodules

Courtesy of N. Bazin / CEA



- Detailed procedures will be defined for every phases
  - Components and tools
  - Operations
  - Controls and tests

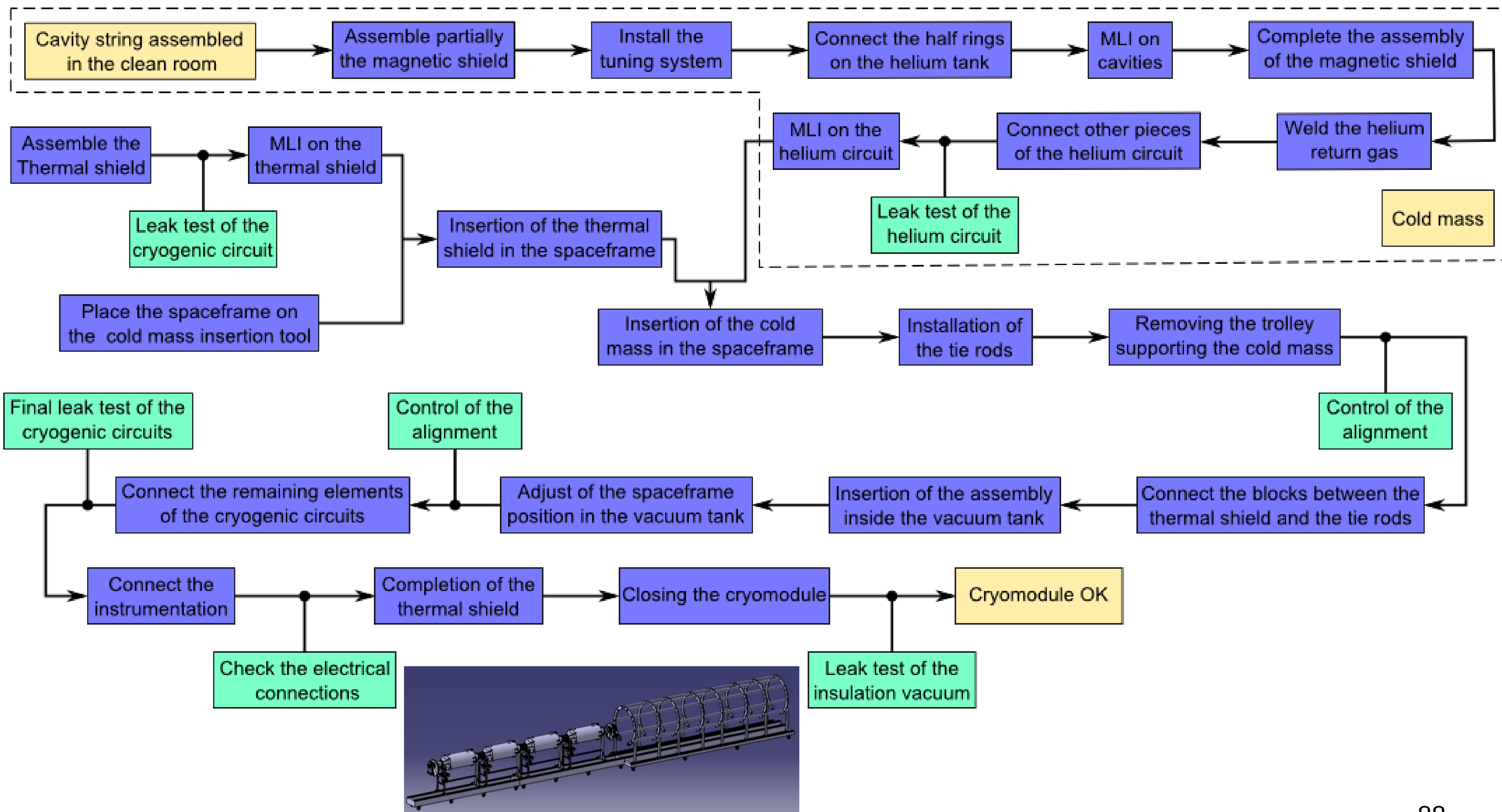
Tooling for elliptical cavities:



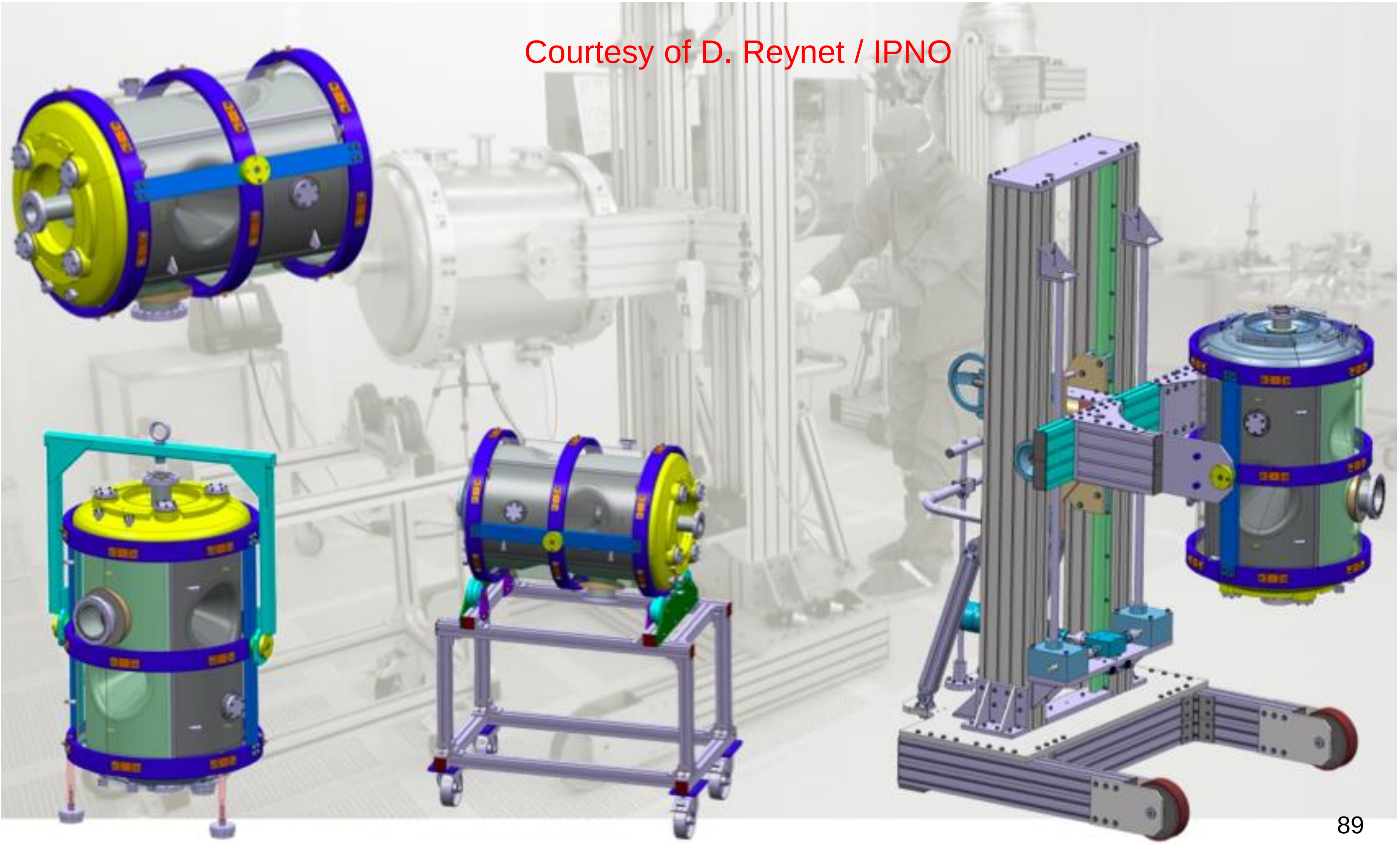
Assembly: outside clean room

EUROPEAN SPALLATION SOURCE

Courtesy of N. Bazin / CEA

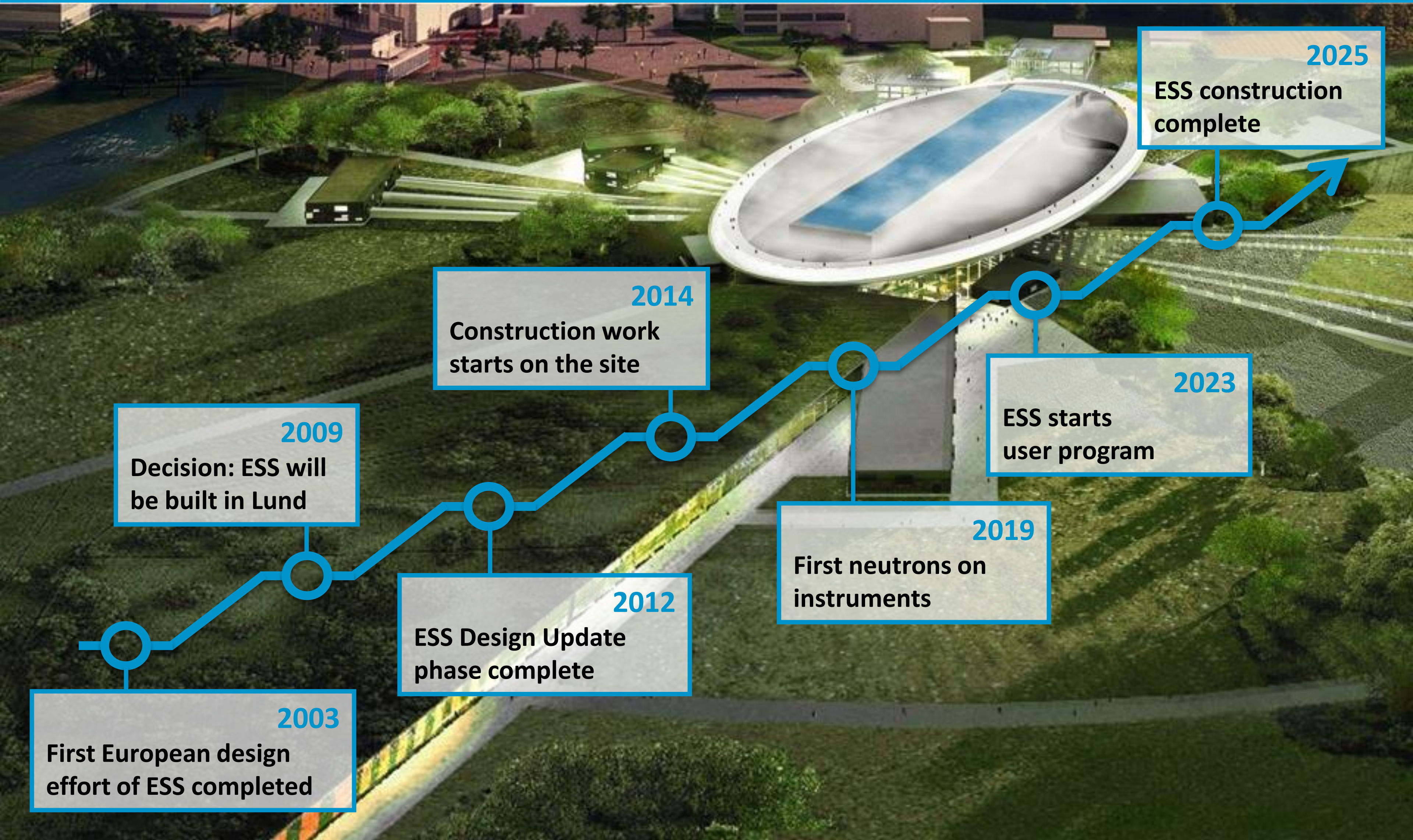


Spoke assembling in clean room/IPNO



Road to realizing the world's leading facility for research using neutrons

EUROPEAN SPALLATION SOURCE



EXTRA SLIDES

Secondary particle produced at J-PARC