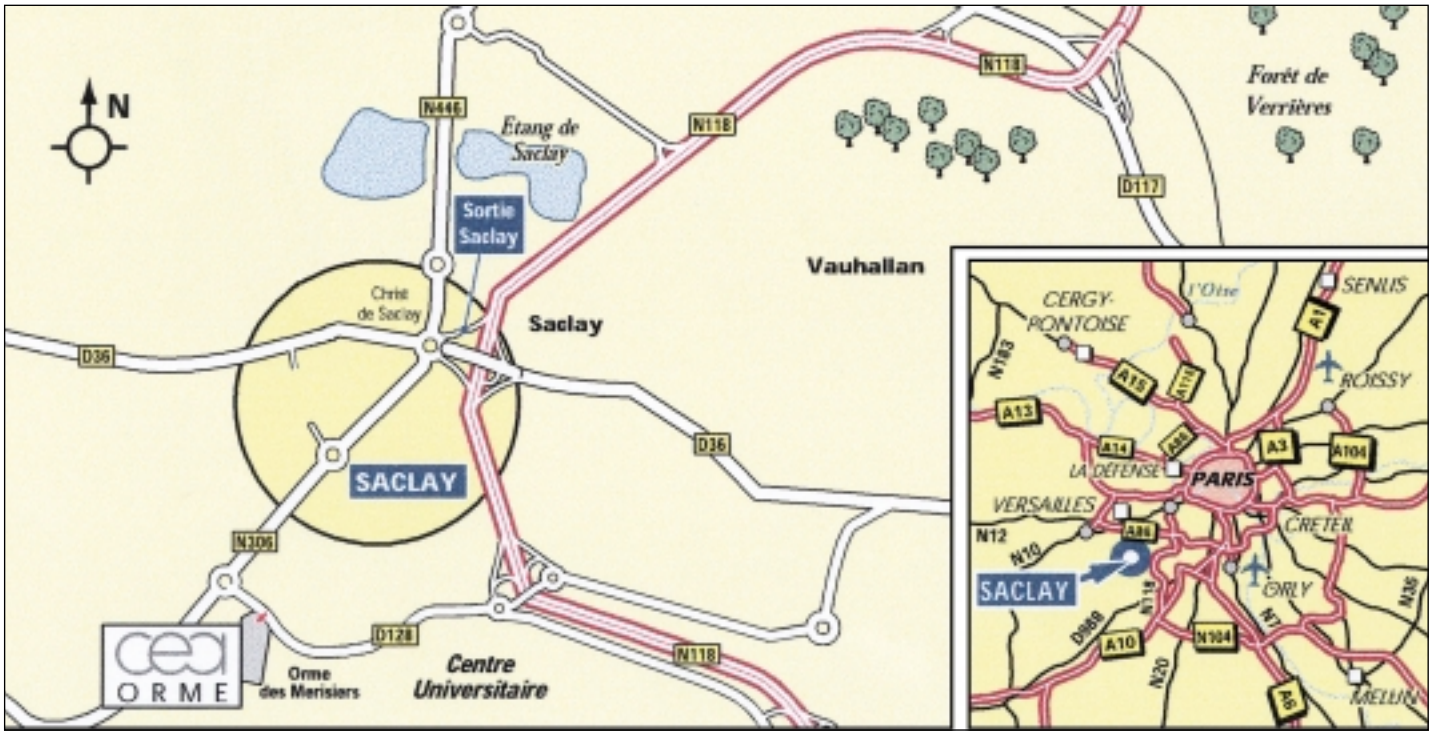
Laboratoire Léon Brillouin

Orphée à la Lyre, 1957

Laboratoire Léon Brillouin
CEA Saclay
bâtiment 563
91191 Gif-sur-Yvette cedex (France)
TÉL.: 33 (0) 1 69 08 52 41 - FAX: 33 (0) 1 69 08 82 61
Web site: http://www-llb.cea.fr



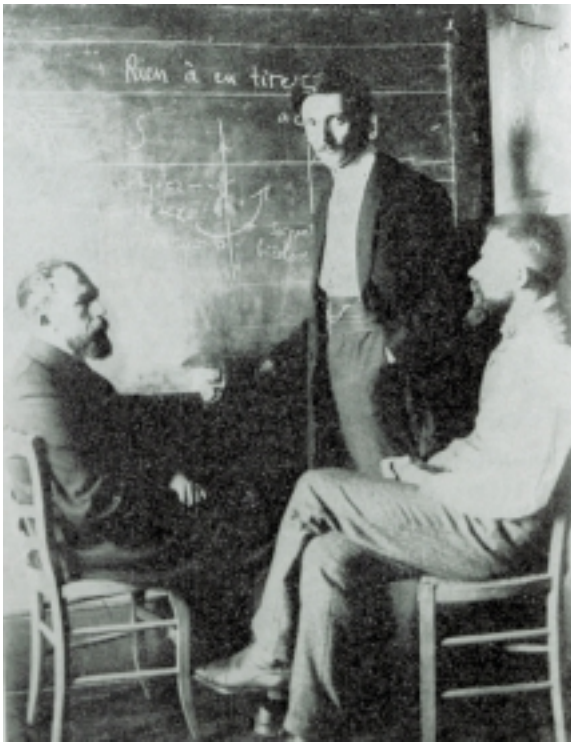
The Laboratoire Léon Brillouin

Neutron spectroscopy first appeared in the United States at the end of World War II, making use of the reactors built in conjunction with the Manhattan project.

The technique quickly proved to be a powerful means of investigating condensed matter and, in spite of its high cost, gradually saw expanded use throughout the world; in France, with the EL3 reactor at Saclay (1957) and with the Mélusine and Siloé reactors in Grenoble (1959).

In 1972, the start up of the High Flux Reactor at the Laue Langevin Institute (originally a Franco-German effort, later to be joined by the British) gave a powerful impetus to the development of this technique.

In order to preserve and develop the vitality of France in this field, the CEA and the CNRS decided in 1974 to create a joint laboratory charged with the construction and operation of several neutron spectrometers. The vocation of this laboratory was to make these tools available to the French scientific community as well as to develop its own research programs.



Léon Brillouin (standing) at the Ecole Normale Supérieure, with Beauvais and Perrot (1910).

The decision to construct a specialised reactor, optimised for the production of neutron beams, was made in 1976.

The reactor «Orphée» went critical in December 1980. Since that time, the «Laboratoire Léon Brillouin» has evenly balanced its two missions: to design and to ensure the evolution of high performance spectrometers; to develop contacts with a significant number of French laboratories, either in the form of scientific collaborations, or by welcoming and helping visiting teams of researchers to perform experiments. Moreover, for the last ten years, the Laboratory has actively promoted the participation of other European countries.

In 1997, the LLB welcomed around 750 visitors, and nearly 400 experiments were completed on its 25 operating spectrometers. Approximately 20% of the proposals for experiments originated from non-French laboratories within the European Union.

The neutron

The hypothesis of the existence of a neutral particle similar in mass to the proton, was formulated in 1920 by Rutherford as the result of four important earlier discoveries: the periodic table of elements (Mendeleev, 1869); the theory of natural radioactivity (Becquerel, 1896; P. and M. Curie, 1898); the discovery of the atomic nucleus and the planetary model (Bohr, 1913); the theory of artificial transmutation (Rutherford, 1919). This hypothesis explains why, when the atomic number is increased by one, the mass of the corresponding atom changes by about two times the mass of the proton. In 1930, Bothe and Becker observed that, if a beam of alpha particles bombards beryllium, the resulting radiation is more penetrating than any previously known radiation and can still be detected even after crossing 10 cm of lead. J. Chadwick, a student of Rutherford, showed in 1932 that this radiation consists of heavy particles, with no electrical charge: the neutron.

Nuclear reactors

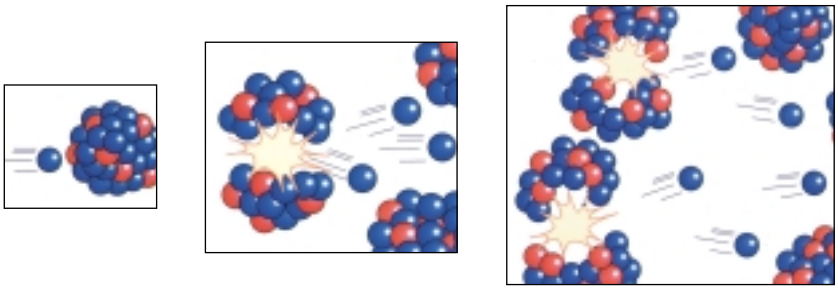
The fission of the uranium atom, that is to say its breaking up into two fragments after having absorbed a neutron, was demonstrated in 1938 (I. Joliot-Curie, O. Hahn). The emission of excess neutrons during this process was subsequently observed and, as a consequence, the fission of other uranium nuclei...and the possibility of a «chain reaction» (F. Joliot, Halban, Kowalski, 1939).

As sources for neutrons, non-military reactors may be categorised as follows:

- those intended to produce electricity. The goal is to maximise the amount of heat produced, in other words the total number of atoms that split per second. The core is not very concentrated (natural or slightly enriched uranium) but does occupy a substantial volume.
- those intended to produce a high flux of neutrons, either of high energy for the irradiation of materials (used in technological research), or «thermal» for outgoing beams (used in neutron scattering for basic or applied research). It is the specific power, in other words the number of atoms per cm^3 that split per second, which is to be maximised. The core is normally very compact and highly enriched with fissile uranium (^{235}U).

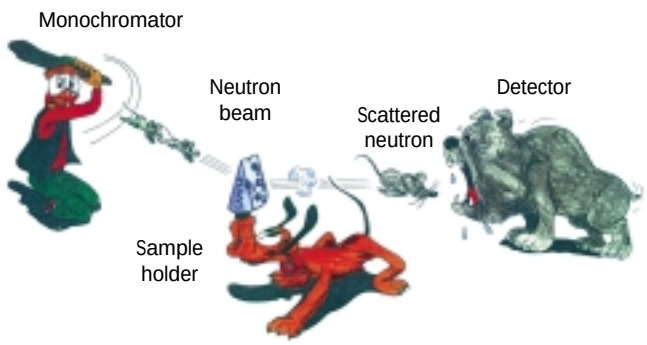
An other method exists for producing intense neutron beams : by knocking heavy atoms (Pb, Hg, ...) with high energy protons, the excitation state of the nucleus is such that they relax with emission of neutrons. It is the principle of the spallation sources.

Reactor



Fission and a chain reaction

Spectrometer



Scattering configuration in real space

Neutron scattering: direction for use

Neutron spectroscopy is typically done at large scale facilities where, as is the case of the « Laboratoire Léon Brillouin », scientists, engineers and technicians work jointly to perform experiments with thermal neutron scattering in a variety of fields.

In this laboratory, our sponsoring agencies (the CNRS and the CEA) have chosen to maintain basic and technological research activities as well as to provide a service to external users. Each year, numerous Ph.D. are defended at the LLB and countless results are obtained by visiting researchers, who come here from both French and foreign laboratories.

This document has been designed for the young (and not so young) scientist, so that he or she may discover the mysteries of thermal neutron scattering.

This brochure obviously cannot cover all aspects of neutron scattering. Subjects not included are, for example: the calculation of the scattering function and its links with the correlation functions of the position of the scatterers; the powerful tool represented by the analysis of polarisation ; ....

Books on the scattering of thermal neutrons include:

S.W. Lovesey-*Theory of neutron scattering from condensed matter*-Clarendon Press, Oxford (1984).

M. Bée-*Quasi-elastic neutron scattering*-Adam Hilger, Bristol and Philadelphia (1988).

P.A. Krupchitski-*Fundamental research with polarized slow neutrons*-(translated by V.I. Kisin)-Springer-Verlag, Berlin (1987).

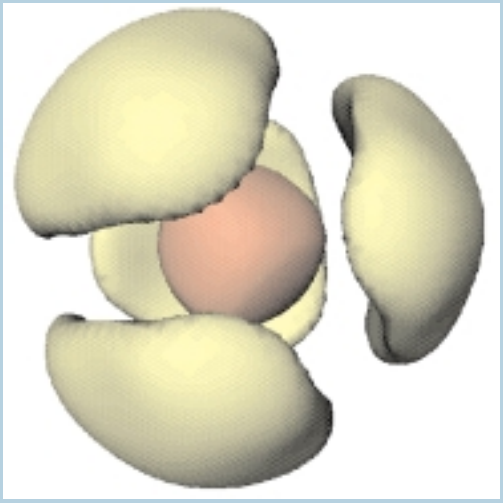
V.F. Sears-*Neutron Optics*-Oxford University Press, New York (1989).

«Hercules» - *Neutron and Synchrotron radiation for condensed matter studies*-Springer-Verlag, les éditions de physique (1993).

Thermal

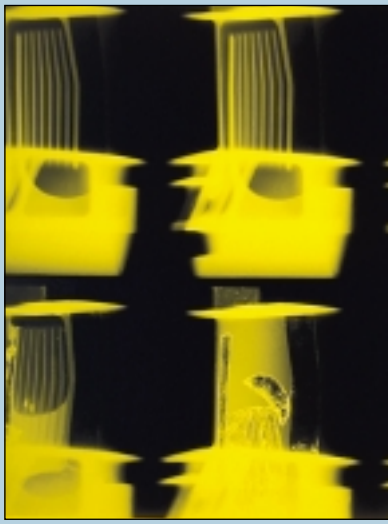
STRUCTURAL STUDIES PHASE TRANSITIONS

- Ceramics, zeolites, hydrides, alloys
- Intercalation compounds
- Molecular systems
- Quasi-periodic systems
- Lattice dynamics
- ....



Disorder of protons (in yellow) of an ammonium group in a molecular solid $(NH_4)_2SiF_6$ (3D reconstruction by entropy maximisation based on the diffraction spectrum).

NEUTRON



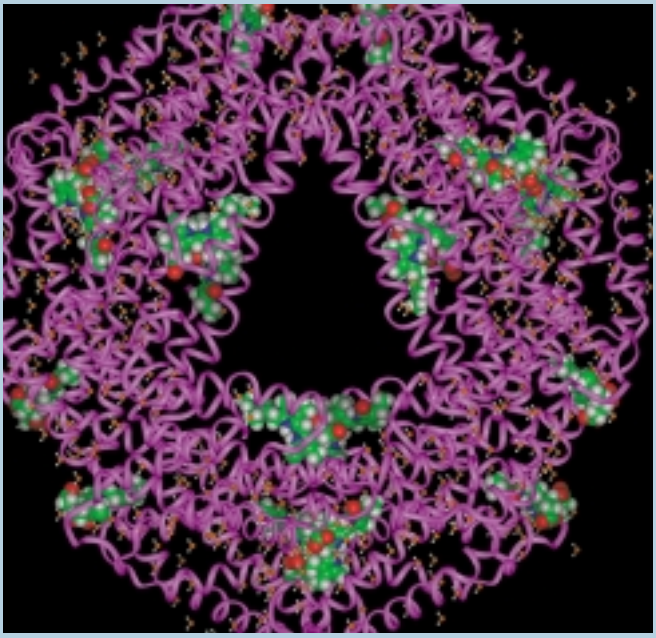
Neutron radiography check of a series of 8 paddle turbines (European Gas Turbine LTD).

BIOLOGY

- Protein folding
- Localisation of water molecules
- Membrane conformation
- ....

PHYSICAL CHEMISTRY

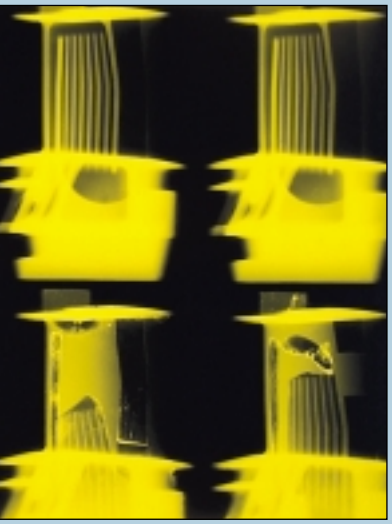
- Polymer conformation
- Vesicles, micelles,
- Microemulsions
- Electrolytes, gels
- ....



Helical structure of the hexamer part of an hydrated C-phycocyanin (simulation).

Neutron Scattering

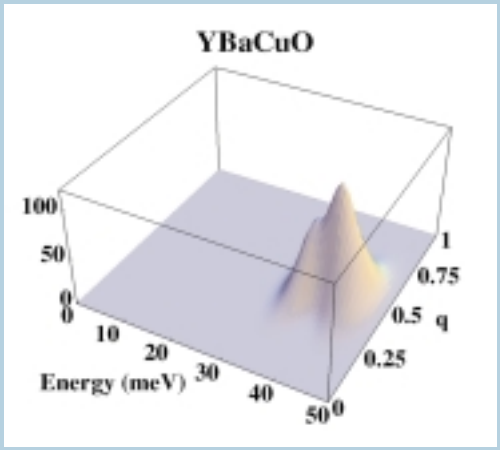
RADIOGRAPHY



The ribs of the 4 lower elements exhibit manufacturing defects.

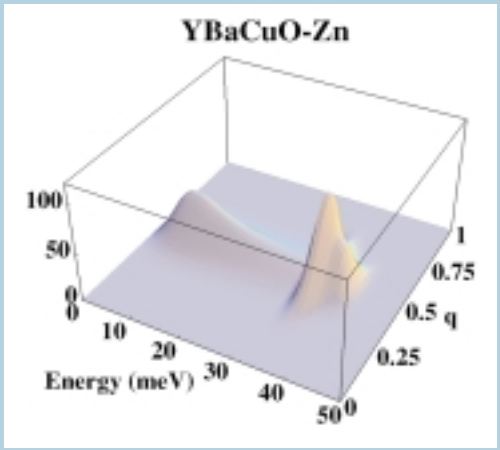
MAGNETISM

- Low dimension magnetism
- Molecular magnetism
- Multi-layers
- Nano particles
- Heavy fermions
- ....



SUPER-CONDUCTIVITY

- Structure
- Phase diagrams
- Excitations
- Electronic correlations
- ....



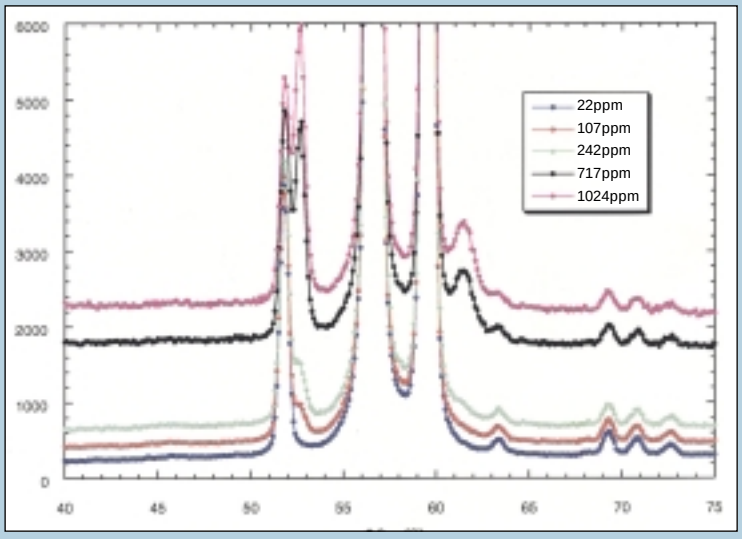
Spectrum of electron excitations obtained by inelastic neutron scattering:
1) YBaCuO pure (superconducting)
2)YBaCuO doped with zinc (non superconducting)

MATERIALS

- Textures
- Strains - Stresses
- Precipitates
- Voids
- Composites
- …

DISORDERED SYSTEMS

- Alloys
- Nanostructures
- Liquids, amorphous solids
- Dynamics
- Glass transitions
- …



Hydrogen content in a nuclear fuel cladding (made of zircaloy) determined by incoherent neutron scattering

1 - Radiation and matter

In order to explain the physical properties of a material or of a class of compounds, it is essential to understand the interactions between its elementary components. On a microscopic scale, these interactions determine:

- the ordering relations between atoms (or molecules) and between electronic magnetic moments;
- the dynamical characteristics of each atom (individual dynamics) and the phase correlations between the motions of two distinct atoms (collective dynamics). The same analogy exists for magnetic moments.

Over the past century numerous techniques have been developed, which investigate on an atomic scale and which enable the physicist to describe with increasing precision these fundamental interactions. Techniques where radiation interacts with matter occupy a special place in these investigations: beams of photons, electrons, protons, helium atoms...and slow neutrons.

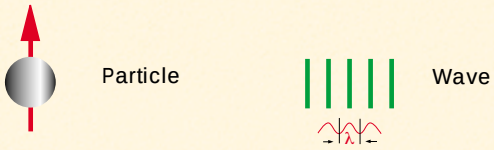
Wave-Particle duality

In 1924, Louis de Broglie wrote in his treaty on wave mechanics:

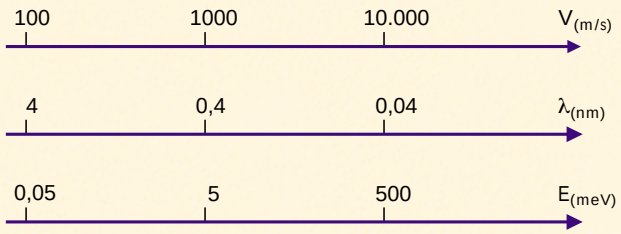
«A wave must be associated with every material particle. The particle motion can be deduced from the propagation laws for the corresponding wave.»

This statement, whose relevance has been proven by numerous experiments with electrons and photons, has divided the scientific world; since rational thinking would require that an object be classified as black or white, but not both at the same time : «It is obvious that the unconditional and simultaneous application of the undulatory and corpuscular representation leads to immediate contradictions. We must therefore conclude that the use of these representations must have certain natural limits».
(W. Heisenberg, Les principes physiques de la théorie des Quanta, 1932).

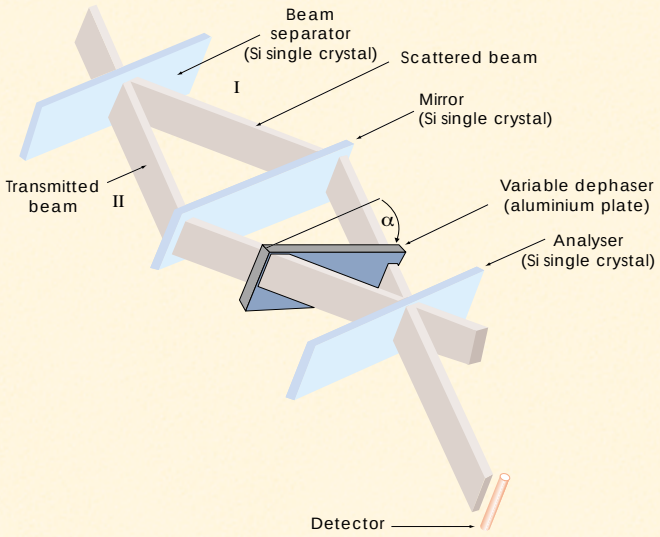
Characteristic neutron parameters and expressions for its moment and its energy in the two representations



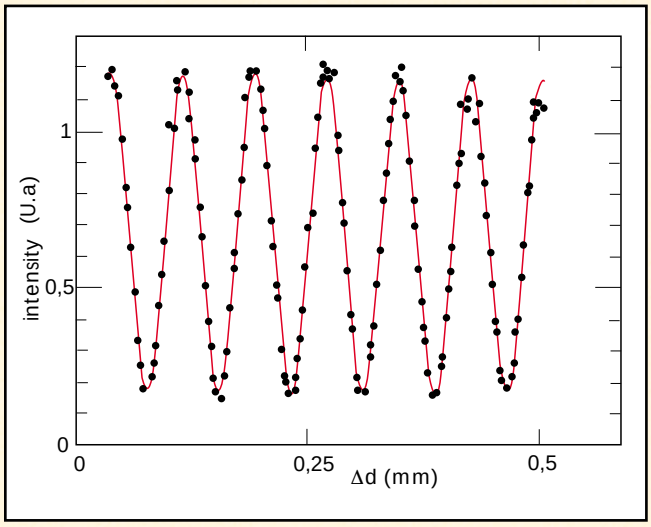
Charge:	0		
Mass:	$m = 1{,}67 \cdot 10^{-24}$ g		
Radius:	$r_o = 6 \cdot 10^{-16}$ m	Wave length:	$\lambda = \frac{h}{m\,v}$
Spin:	1/2	Wave number:	$k = \frac{2\pi}{\lambda}$
Magn. Moment:	$\mu = -1{,}9\mu_N$		
Momentum:	$\vec{p} = m\,\vec{v}$	Momentum:	$\vec{p} = \frac{h\,\vec{k}}{2\pi} = \hbar\,\vec{k}$
Energy:	$E = \frac{1}{2}\,mv^2$	Energy:	$E = \frac{h^2}{2m\lambda^2} = \frac{\hbar^2 k^2}{2m}$
(v = velocity)			(h = Planck's constant)



Neutron interferometry



(a) Diagram of a crystal interferometer



(b) Modulation of the beam caused by interferences in the analyser (from H. Rauch in «Neutron Interferometry», Oxford science publications, 1979).

A demonstration of the undulatory nature of the neutron is provided by the results of interferometry experiments done with neutron beams (which are similar to long proven results using light). The interferometer consists of 3 silicon single crystals whose lattice planes (containing the atoms) are all perfectly parallel. (Fig a).

Wave physics teaches us that the intensity of the outcoming beam is expressed as:

$$I = I_0 [\, a + b \cos(\Delta d)]$$

where Δd is the difference in the «optical paths» followed by the two beams. By introducing an aluminium plate (aluminium is very transparent for neutrons but its neutron index, and therefore its optical path per unit length, is different from that in air), we cause Δd to vary by changing the angle α (variable dephaser).

Figure (b) shows the number of neutrons counted per unit time in the detector as a function of Δd. The observed modulation indicates that, in the analyser crystal, the 2 beams have recombined coherently - in complete agreement with the undulatory hypothesis.

A beam with a well-defined and well-known energy and direction propagates through the material under study and interacts with its basic constituents. In the scattering process, which results in a change in the direction of propagation and/or in the energy of the radiation, the «radiation-scatterer» pair specifically determines the interaction. From theory, one can link the characteristics of the scattered beam to the properties (order and dynamics) of the scatterers. Thus, by using several «radiation probes», different and complementary informations can be obtained.

Other frequently used measuring methods include: magnetic resonance techniques (nuclear and electronic), different kinds of microscopies (electronic, near field...), infrared and Raman spectroscopies,...

2 - Interaction with matter

A: Scattering function

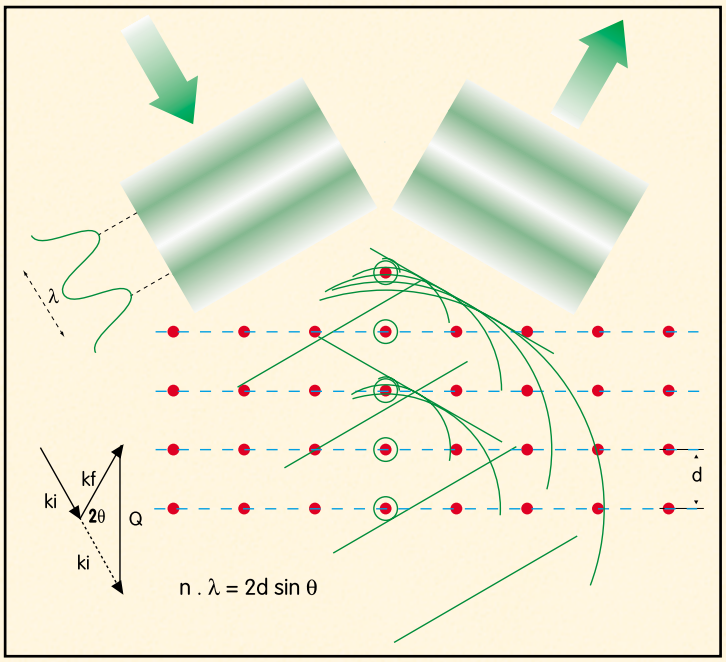
Let us return for a moment to the wave-particle duality. When the particles have a high energy (for neutrons, E = 40 keV corresponds to $\lambda = 10^{-4}$ nm, by far shorter than the distances between atoms in matter), the corpuscular approach illustrated by the image of a shock between 2 billiard balls is perfectly justified (cf. P.24 «Neutron thermalization process»). On the contrary, we are interested here in thermalized neutrons with wavelengths between 0.05 and 2 nm, therefore comparable to inter-atomic distances. Theory indicates that, in this case, it is the «diffraction» phenomena that are predominant; the latter can only be treated rigorously when using a wave point of view.

In the incident beam, a neutron is defined by its wave vector $\vec{k_i}$ and its energy E_i; after scattering, its wave vector becomes $\vec{k_f}$ and its energy E_f. Based on the conservation laws (moment and energy) resulting from the application of the basic principles of physics, we deduce that the neutron and the scattering system have exchanged:

• a momentum	$\vec{Q} = \vec{k_i} - \vec{k_f}$	$\vec{Q}$ = momentum transfer
• an energy	$\hbar\omega = E_i - E_f$	$\Delta E = \hbar\omega$ = energy transfer

Bragg's diffraction

If the scatterers (atoms) exhibit a periodic spatial order, the spherical wavelets produced by each scatterer add up coherently in certain directions (in other words, all have the same phase within 2π). Consequently, we will have in these directions a plane wave of great amplitude (Bragg diffraction). The angle 2θ by which the beam deviates from its incident direction depends on the periodicity of the lattice (d) and on the incident wavelength (λ).

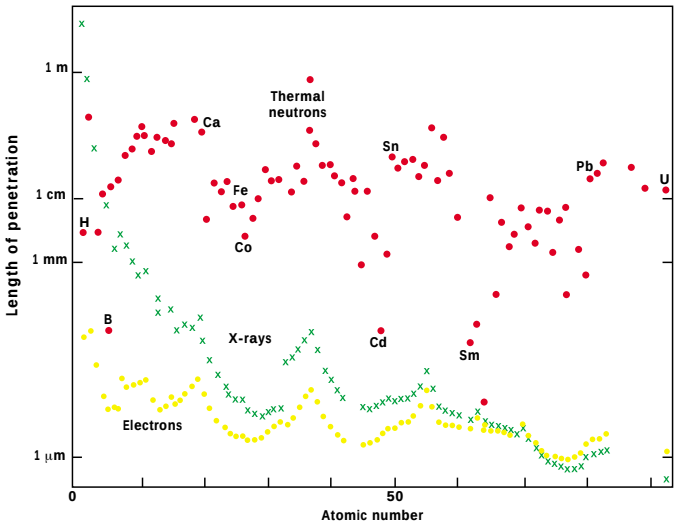


Wave scattering in an ordered plane of atoms.

The probability of this exchange (noted $S(\vec{Q},\omega)$, scattering function) is measured by the number of neutrons ($\vec{k_f}$, E_f); it is directly linked to the nature and to the force of the interactions between the wave and the scatterer.

It is the characteristics of this interaction, which will reveal the properties of the scatterers that are accessible for the experiment, making neutron scattering an indispensable tool in the study of numerous properties of condensed matter.

B: Neutron-atom interaction



Penetration depth of a beam of thermal neutrons (0.18 nm), of X-rays (0.1 nm) or of electrons (0.004 nm) as a function of the atomic number. Note the logarithmic scale along the vertical axis.

❑ Carrying no electrical charge, the neutron has no electrostatic interaction with the electron cloud of the atom.

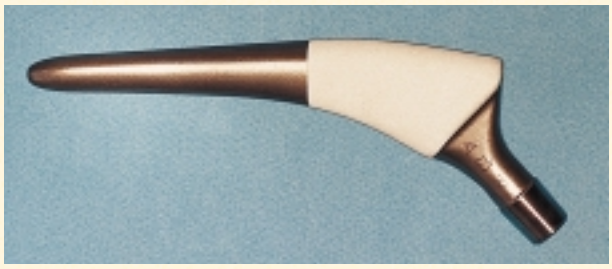
❑ The neutron is sensitive to the nucleons within the nucleus of the atom. It interacts with the latter via «nuclear» forces, that is to say at very short range (diameter of the nucleus ≈ 10^{-12} cm).

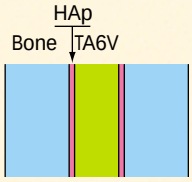
These characteristics entail 3 important differences between neutrons and X-rays:

- Due to its weak interaction with matter, the neutron has a high penetration power. Contrary to X-rays, which perceive atoms only within a thickness of a few µm (10^{-6} m) from the surface, neutrons can probe all the atoms in a large sample.

Measurement of residual stresses in a hip prosthesis

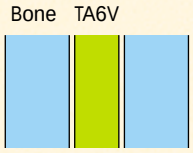
The hip prosthesis is made of solid titanium-based alloy (TA6V), covered with a layer of hydroxy-tapatite (HAp) a few tens of µm thick.



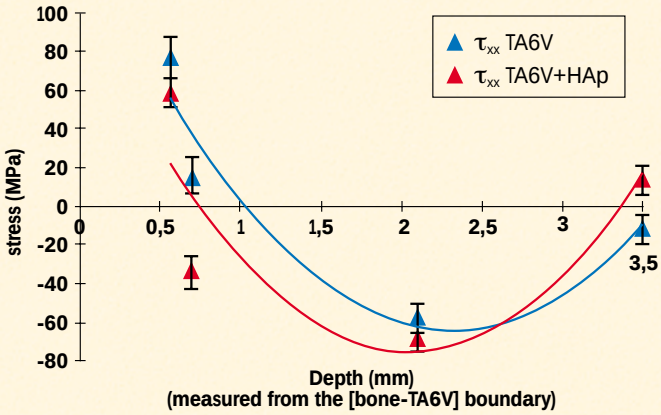


The prosthesis is implanted in the bone.

The bone «colonises» the layer of HAp and, over several months, transforms it, assuring adherence



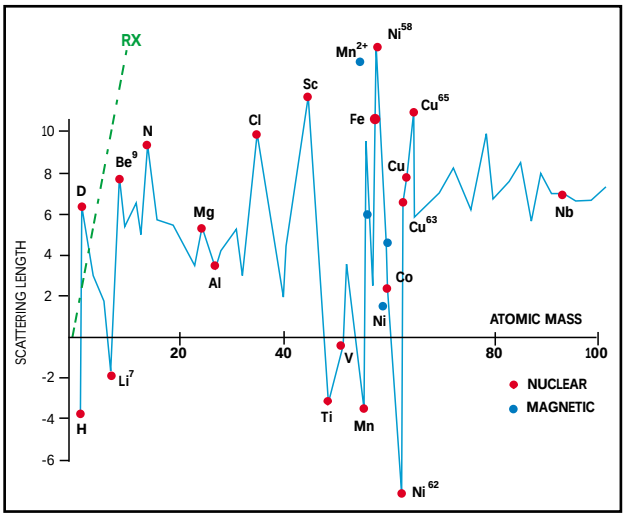
On the average, at the end of 20 years, there is a loss of adhesion. A new surgical operation is then necessary



Residual stress in titanium as a function of depth.

One of the possible causes of this limited lifetime could be the appearance of stress in the titanium during the transformation phase from HAp to bone. Thanks to their great penetration power, neutrons enable the scientist to measure these stresses in the prostheses. The results to the left show that, before implantation, the deposit of the HAp coating has little effect on the level of internal stresses, which is relatively weak on the whole.

- The «nuclear» interaction depends on the number of nucleons in the nucleus and on the energy levels that they occupy. It shows no correlation to the atomic number Z, whereas for X-rays it is proportional to the number of electrons present. The neutrons thus allow the scientist to «see» some light atoms that are barely visible with X-rays. An important application is the location of hydrogen in crystallised biological systems and in molecular crystals.

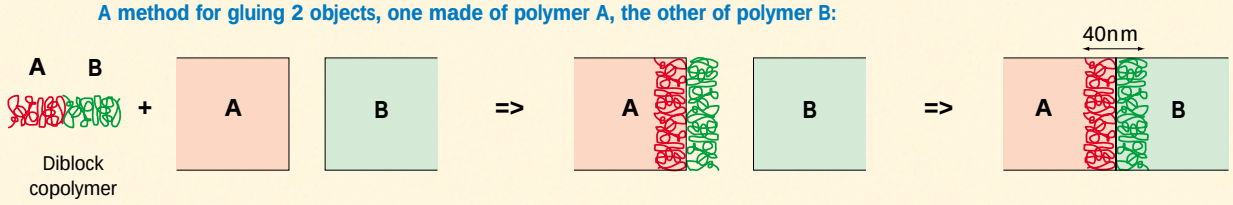


Thermal neutron scattering lengths of different elements.

Interaction of the neutron with the corresponding element, measured in unit of length (10^{-12} cm). In a material made of several atom species, the contribution of each atom to the total cross section is a function of its scattering length. The broken green line is the same quantity for X-rays.

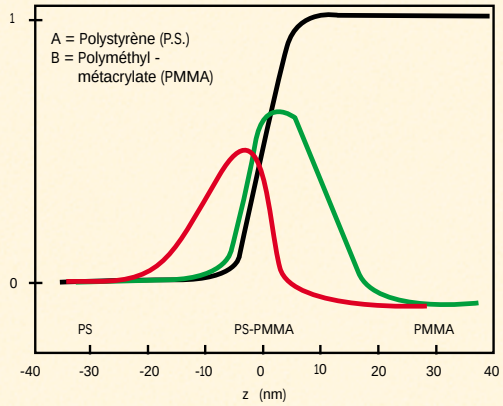
- Finally, 2 isotopes of the same element have different interactions, which is not the case for X-rays (2 isotopes have the same number of electrons). This property is the basis for «differential» measurements by isotopic substitution, such as the measurement of partial structure factors or of the conformation of objects (macromolecules, micelles, vesicles) by the contrast variation method.

A method for gluing 2 objects, one made of polymer A, the other of polymer B:



- prepare the copolymer A-B
- coat one of the sides to be glued
- wait for the penetration of the corresponding part of the copolymer (curing)
- bring together the two surfaces

Generally speaking, all polymers are made of carbon and hydrogen in similar proportions and thus possess the same scattering power (no contrast). Thanks to the selective deuteration of one or the other component of this system (A, B, A-B), the neutrons allow us to quantify the concentration of the different macromolecules when crossing through the glued interface.

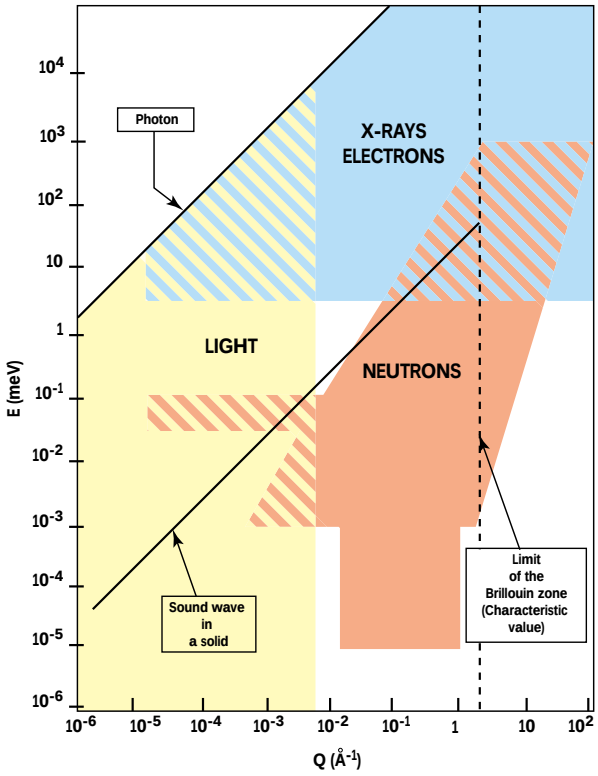


PS(h)/PS(h) - PMMA(d)/PMMA(d) : total concentration in PMMA
PS(h)/PS(d) - PMMA(h)/PMMA(h) : concentration in PS belonging to the copolymer
PS(h)/PS(h) - PMMA(d)/PMMA(h) : concentration in PMMA belonging to the copolymer

Concentration of the different components as a function of the position.

❑ Numerically the relation that links the energy (E) of the neutron to its associated wavelength (λ) is well adapted to the study of condensed matter. Indeed, thanks to its mass value, the neutron simultaneously satisfies 2 requirements:

- a wavelength comparable to the interatomic distances (a few 10^{-1} nm) and therefore the possibility of interference patterns (diffraction).
- energy of the same order of magnitude as the excitation energies of the scatterers (atoms or magnetic moments) which are thus easily measurable.



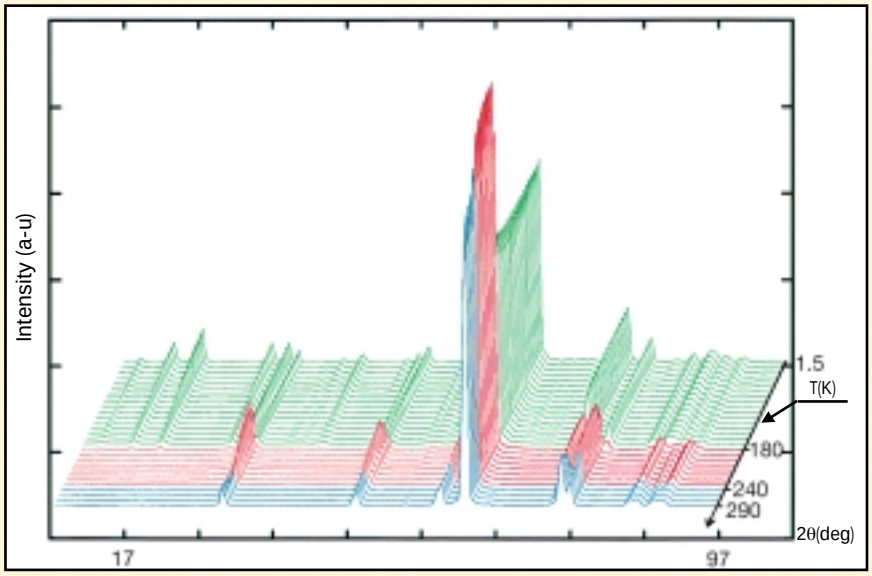
Regions of space [momentum transfer (Q), energy transfer (E)] accessible by different probing radiations.

❑ The neutron has a spin 1/2. It is equivalent to a magnetic moment $\vec{\mu}_N$ and, consequently, is sensitive to the magnetic fields created by unpaired electrons present in the material under study. In these magnetic materials, the [matter-neutron] interaction potential includes a coupling term [neutron spin-atomic magnetic moment] which, similar to the term for the [neutron-nucleus] interaction, allows us to study the order (magnetic structures) and the interaction of these moments.

Magnetic phase transitions

The manganese perovskite undergoes a succession of structural and magnetic phase transitions, as a function of temperature, which are seen in the modifications of the diffraction pattern:

T > 240 K,	paramagnetic phase (blue curves)
240 > T > 180 K,	ferromagnetic phase (red curves)
T < 180 K,	antiferromagnetic phase (green curves) with a charge order of the Mn^{3+}/Mn^{4+} ions

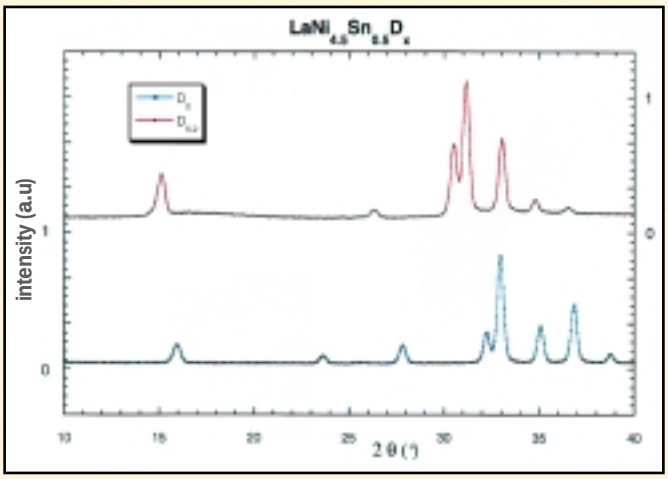


Neutron diffraction patterns at different temperatures.

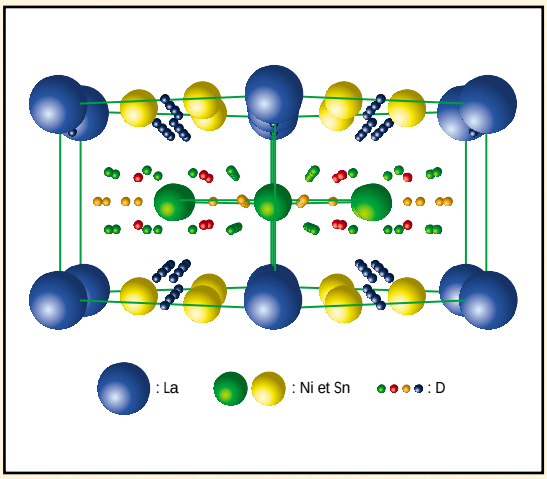
Singling out hydrogen atom

When a given structure associates elements of very different atomic numbers Z, the lightest element only makes a very weak contribution to the scattered X-ray beam (each species makes a contribution proportional to Z^2). On the contrary, thermal neutrons have a strong interaction with hydrogen.

The AB_5-type compounds have the property to reversibly store large quantities of hydrogen at room temperature and pressure. The replacement of a fraction of the atoms B by another metal often improves the storage performance. For instance, up to 6 hydrogen (or deuterium) atoms may be inserted in the hexagonal unit cell of La $Ni_{4.5}$ $Sn_{0.5}$. Neutron diffraction enables the scientist to pinpoint their positions.

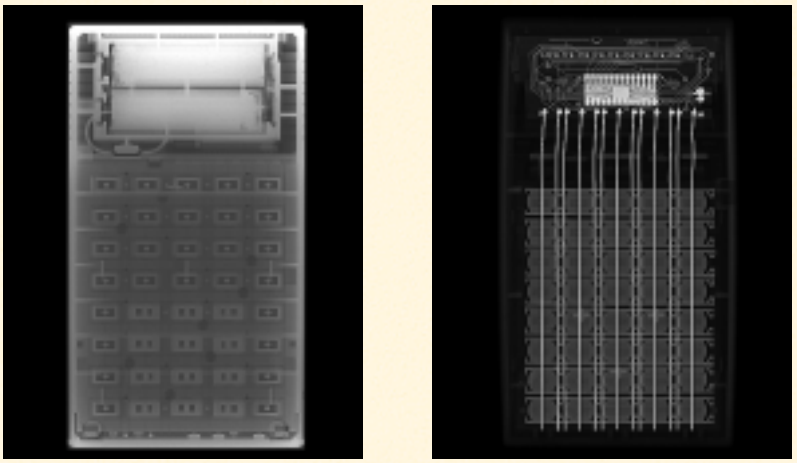


Powder diffraction pattern of $LaNi_{4.5}Sn_{0.5}$ and $LaNi_{4.5}Sn_{0.5}D_{5.2}$.



Position of the atoms in the unit cell.

The insertion of deuterium causes the unit cell to expand (the diffraction peaks shift) and changes the relative intensity of some peaks.



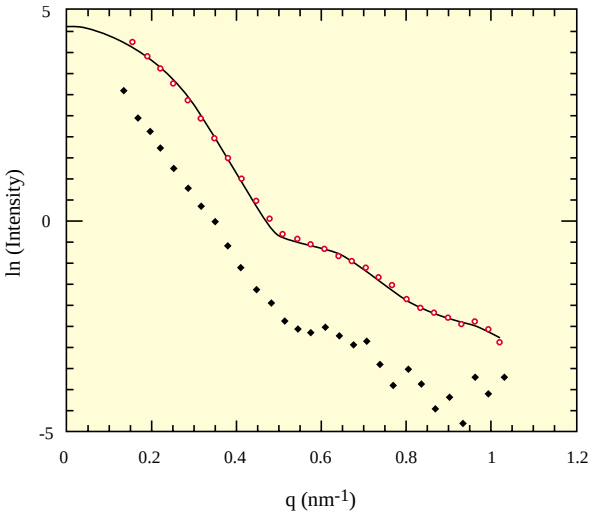
«Neutron» and «X-rays» pictures of a Texas Instrument pocket calculator.

A practical application of the great «visibility» of hydrogen is to allow observation, by neutron radiography of an organic material (containing mainly carbon and hydrogen), enclosed in a metallic host. For example, the pyrotechnic devices that ensure the in-flight separation of the different stages of the Ariane rocket are systematically «neutronographed» using the neutrons produced by the reactor Orphée.

3 - The specific contribution of neutron spectrometry

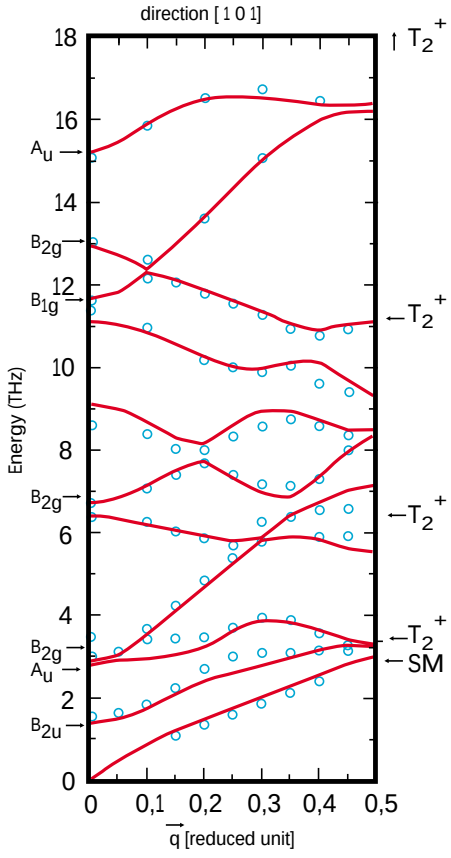
As a result of its characteristics (zero charge, mass, spin) the neutron possesses, on one hand, some unique properties (different interaction with 2 isotopes, penetration power, magnetic interaction) and, on the other hand, characteristic values (wave vector, energy) that are found together in no other probing radiation. To conclude this chapter, let us look at some examples in which these properties apply:

- X-rays beams are more intense, easier to produce and to operate; their use must be the basis of any structural study. But when trying to locate light atoms in the midst of heavy ones, to observe specific correlations between certain atoms in a liquid or to characterise clusters in an alloy, the advantage of neutron beams is easily proved. For example, the use of the magnetic interaction between the neutron and the iron atom provides information on the precipitation of copper aggregates in certain steels that have undergone significant thermal ageing or neutron irradiation.



Neutron scattering in a polycrystalline sample of Fe $Cu_{1,5\%}$ after thermal ageing (312h, 500°C).

Intensities measured by small angle scattering when applying a magnetic field of 1.4 T. The nuclear and magnetic contributions can be separated due to the anisotropy of the magnetic scattering: it vanishes in the direction $\vec{q} \parallel \vec{H}$ (◆) and has a maximum in the direction $\vec{q} \perp \vec{H}$ (o). The scattered intensity can be fitted (—) satisfactorily by taking into account spherical particles made of pure copper with a gaussian size distribution, and a mean radius of 8nm. This study is part of the research on the mechanisms of an eventual embrittlement of the pressure vessels used in some nuclear reactors after several years of use.



- Similarly, infrared and Raman spectroscopies provide access to the energy values of the elastic excitations in solids (phonons). But the wavelength of photons used, to first order at least, only allows coupling to modes near the zone centre (q~0). Measurement of the characteristics of the propagation (dispersion curves) in the entire Brillouin zone, which are indispensable when testing models of interaction potentials between atoms of a crystal, requires to use neutron spectrometry.

Spin-Peierls transition in $CuGeO_3$

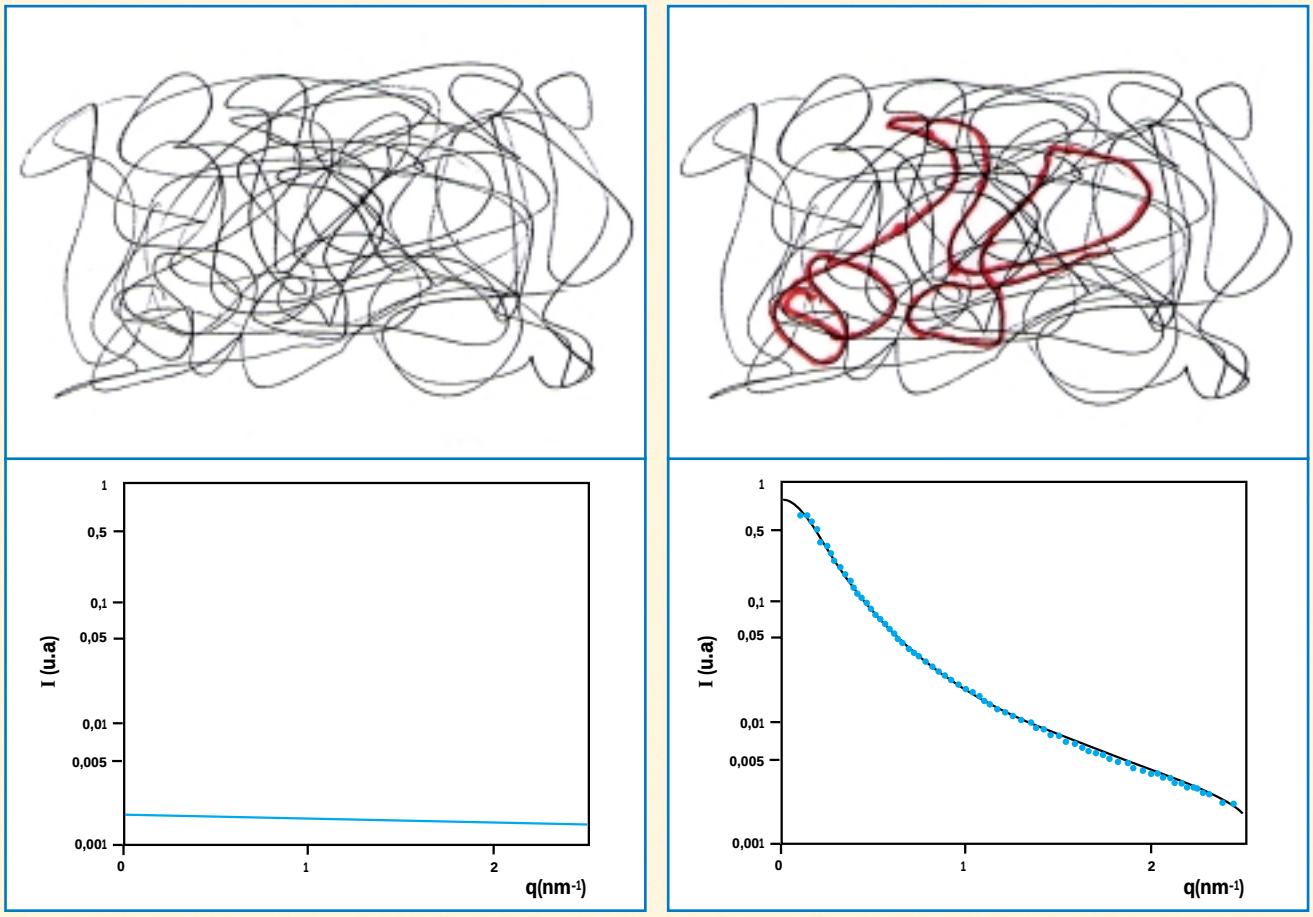
In $CuGeO_3$, the Cu^{++} ions form chains of spins 1/2 coupled antiferromagnetically. When the temperature is lowered, the tendency for an antiferromagnetic quasi-one-dimensional order competes with a dimerisation in which 2 neighbouring spins form an entity with zero net spin, resulting in a nonmagnetic ground state.

A coupling between the spins and the lattice vibrations (phonons) favours this dimerisation and leads to a structural transition characterised by the displacement of the Cu^{++} ions: this is the spin-Peierls transition. Measurements of the phonon dispersion curves by inelastic scattering of neutrons on a single crystal of $CuGeO_3$ provide indications as to the microscopic origin of the coupling. Part of the results obtained at room temperature and the calculated spectrum (full line) based on a lattice dynamics model may be found in the graph to the left. The modes having the symmetry involved in the transition are noted T_2^+ in the figure.

• In physical chemistry as well as in biology, it is important to be able to measure objects of nanometer size, to ascertain their form and, if they are heterogeneous, to know the distribution of each of their components. Again, if small angle scattering of X-rays is widely used, there are cases where only neutrons can provide an answer.

Melt of polymer as «seen» by a neutron beam

To ascertain the conformation of a macromolecule among similar molecules (solid or melted polymers), some macromolecules have been introduced in which hydrogen has been replaced by deuterium. These molecules become «different» for neutrons and are therefore observable. The isotopic substitution H→D introduces a «contrast» in the midst of a chemically homogeneous group.

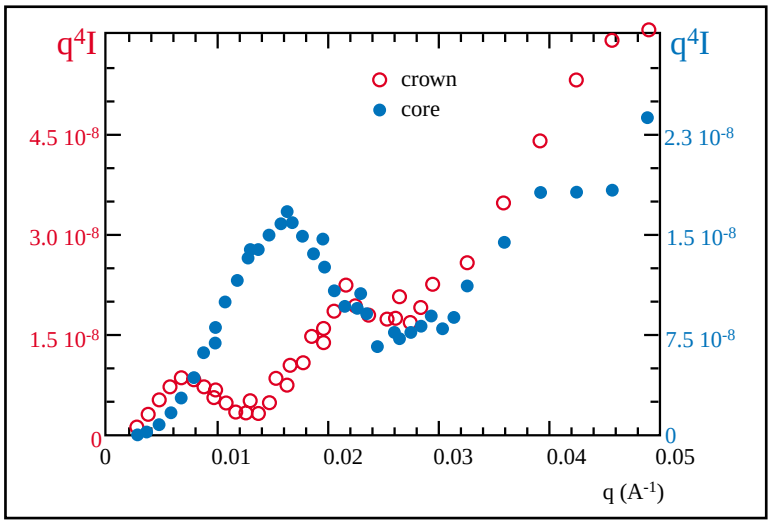


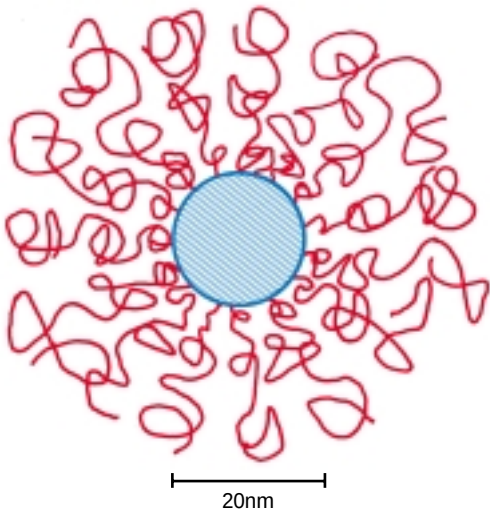
(a) All chains are hydrogenated: there is no contrast, thus little scattered intensity.

(b) 13% of the chains are deuterated: the H/D contrast allows for measurement of the characteristics of a chain.

**Chain conformation (above) and scattered intensity (below) by a polystyrene melt.
In black: hydrogen chains; in red: deuterated chains.**

Observation of the components of a heterogeneous object by the contrast variation method.





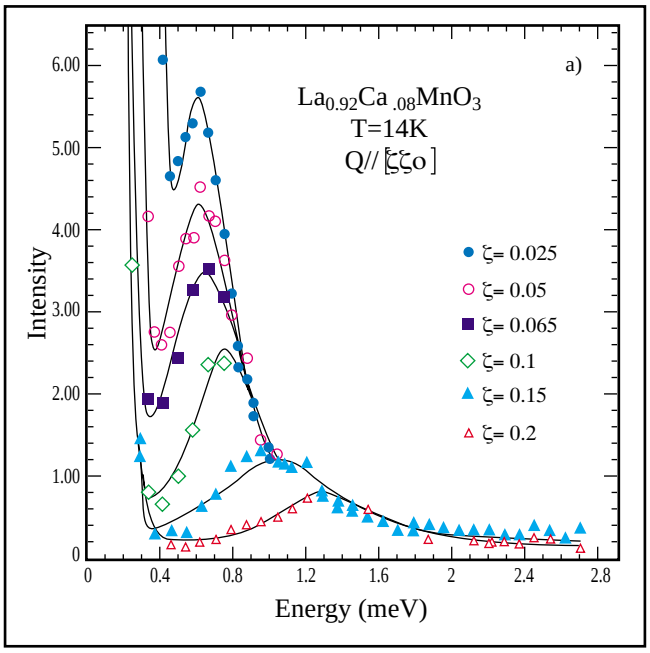
Scattered intensity from colloidal particles of silica surrounded with polymers grafted to their surface according to whether the silica core or the polymer crown is being observed. A judicious choice of the (hydrogenated/deuterated) solvent proportion allows adjustment of the neutron scattering length of the solvent to that of one or another component of the particle. The corresponding part of the particle thus becomes completely «transparent». By varying the «neutron contrast» in this way, different components appear, thus simplifying the problem that can then be resolved.

- The interaction between neutron and magnetic moments of electronic origin carried by the atom allows study of magnetism on a microscopic scale:

  - **order relationships** (ferro, antiferro, ferrimagnetism; canted and helicoidal magnetic structures,...) are measured by diffraction;
  - **magnetic excitations** are accessible by inelastic scattering.

Energy spectra for different wavevectors observed in the magnetic doped semiconductor $La_{1-x}Ca_xMnO_3$.

The compounds $RMnO_3$ (R=Rare Earth, perovskite structure) exhibit a strong variation in resistivity when placed in an external magnetic field (giant magneto-resistance).
The introduction of electron holes by La substitution causes an additional branch of excitations to appear. The very strong decrease in intensity when the wavevector increases reveals the existence of a small ferromagnetic region associated with these new excitations, indicating the existence of «charge droplets» in this compound.



The neutron-nucleus interaction: Fermi's pseudo-potential

A neutron and a nucleus interact through nuclear forces that, experience shows, are short range (of the order 10^{-13}cm).

This interaction distance is, on one hand small compared to the average radius of the nucleus (~ 10^{-12} cm), on the other hand insignificant compared to the wavelength of thermal neutrons (~ 10^{-8} cm).

It can be demonstrated that, in these conditions:

a) neutron scattering is isotropic, in other words equiprobable in all directions of space («s» character scattering);
b) outside of the nucleus, the wave function of the scattered neutron is barely different from the function of the initial wave (plane wave) and therefore, can be calculated by a perturbation treatment to first order (Born approximation);
c) a single parameter, independent of the energy of the incident neutron, is sufficient to describe the interaction. This parameter (b=scattering length) is a complex number which has the dimension of a length. Its real part can be positive or negative according to whether the neutron-nucleus interaction is attractive or repulsive. Its imaginary part represents the probability that the neutron will be absorbed by the nucleus.

Fermi proposed a phenomenological description of this interaction in the form of a «pseudo-potential» having the desired properties: relative to an arbitrary origin, if the neutron is at $\vec{r}$ and the nucleus at $\vec{R}$:

$$V(r) = b\,\delta(\vec{r} - \vec{R})$$

In the case where there is a group of N atoms, the nuclei are numbered from 1 to N

$$V(r) = \sum_{1}^{N} b_j \delta(\vec{r} - \vec{R_j})$$

b_j = scattering length of the atom labelled j

Neutron-atomic magnetic moment interaction

The neutron carries a spin $\vec{S_n}$ $\left(|S_n| = \frac{1}{2}\right)$. It is equivalent to a magnetic moment

$\vec{\mu}_N = -\gamma_N \frac{e\hbar}{m_n c}\vec{S}_n$; $\gamma_N = 1{,}91$ (gyromagnetic ratio) that creates a magnetic field at a distance r:

$$\vec{H}_N(r) = \mathrm{rot}\frac{\vec{\mu}_N \wedge \vec{r}}{|r|^3}$$

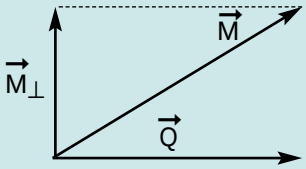
The interaction potential between the neutron and an unpaired electron of the scattering atom carrying a magnetic moment $\vec{\mu_e}$, is written:

$$V_e(\vec{r}) = -\vec{\mu}_e \cdot \vec{H}_N$$

and the total magnetic interaction is obtained by summing over all unpaired electrons:

$$V_j(\vec{r}) = \sum_e V_e(\vec{r})$$

It is much easier to work in the reciprocal space:



$$V_j(\vec{Q}) = \left(\frac{2\pi\hbar^2}{m_N}\right)\int d\vec{r}\; e^{i\vec{Q}\,\vec{r}}\, V_j(\vec{r})$$

$$V_j(\vec{Q}) = \left(\gamma_N \frac{2e}{\hbar c}\right)\vec{M}_\perp(Q) \cdot \vec{S}_n$$

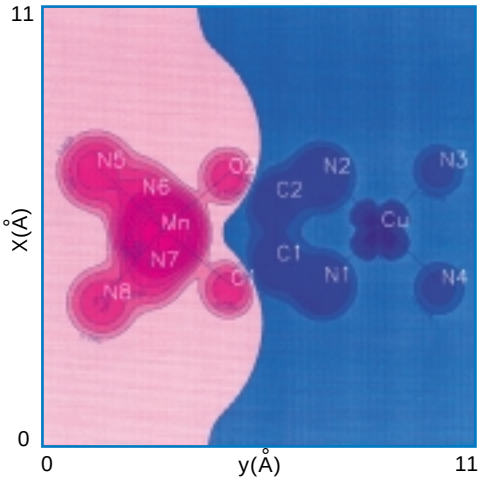
$$\vec{M}(Q) = \Sigma_e\, \vec{\mu}_e\, e^{-i\vec{Q}\,\vec{r}_e}$$

$\vec{r}_e$ = coordinate of the electron e

$\vec{M}_\perp(Q)$ = component of $\vec{M}$ (Q) in the plane perpendicular to $\vec{Q}$.

• Better still, with a beam in which all neutrons have the same spin orientation, for example $S_z = +\frac{1}{2}$ (a polarised beam that we know how to produce), the measurement of the number of neutrons whose spin has been reversed ($S_z = -\frac{1}{2}$) after scattering allows us to determine with great precision the ***local density*** of magnetic moment.

Map of the spin density induced by an external magnetic field in a paramagnetic molecular compound Mn(cth) Cu (opxn) $(CF_3SO_3)_2$ (diffraction of polarised neutrons).
(cth) = hexamethyl - tetraazacyclotetradecane
(opxn) = N, N' - bis - aminopropyl-oxamide



In this compound, the Mn^{2+} and Cu^{2+} ions are linked by an organic oxamide bridge (-O-C-N-). The spin density is represented in projection along a perpendicular to the bridge. The existence of a high positive density region (in red) centred on the manganese (Mn) and of a negative density region (in blue) centred on the copper (Cu), confirm the antiferromagnetic coupling between the metallic ions via the organic bridge, leading to a ground state with spin S = 2. The greater delocalization of the density of negative spin on the bridge reflects the stronger covalent character of copper as opposed to manganese.

The interest of this bimetallic compound resides in that it is similar to the basic links of the chains (Mn, Cu) that form one of the first molecular compounds to display a ferromagnetic order.

Polarisation of a neutron beam

The neutron carries a spin $S\left(|S| = \frac{1}{2}\right)$. Projected on an arbitrary direction in space (for example an external magnetic field), it can only take on the values $S_z^{(+)} = \frac{1}{2}$ et $S_z^{(-)} = -\frac{1}{2}$ (quantum mechanics).

The beam coming out of the reactor is non-polarised, that is to say it contains 50% of the neutrons with $S_z^{(+)}$ and 50% of the neutrons with $S_z^{(-)}$. The production of beams in which all the neutrons have the same spin direction is an excellent opportunity for magnetic studies. Let us look at two currently used methods and one method for the future.

1) Diffraction by a ferromagnetic crystal

When an atom possesses a magnetic moment $\vec{m}$, its interaction with the neutron is the sum of the neutron-nucleus (amplitude b) and the neutron-magnetic moment (amplitude f_{mg}) interactions.
If these atoms are arranged regularly (crystal), and if at the same time their magnetic moments are all parallel (ferromagnetic), the intensity of the Bragg peaks is proportional to $(b \pm |f_{mg}|)^2$. The sign to be chosen depends on the relative orientations of the neutron spin and of the magnetic moment of the atom. If the crystal and the Bragg peak are chosen such that $b \approx |f_{mg}|$, in other words if $(b - |f_{mg}|) \approx 0$, the diffracted beam will only contain the neutrons with $(\vec{S} // \vec{m})$ $(S_z^{(+)})$.

2) Reflection by a «magnetic mirror»

At the interface between the vacuum and a medium region with index n, a beam of neutrons undergoes total reflection if its incidence angle φ is inferior to a critical value $\varphi_c = \sqrt{2\,(1-n)}$ (cf. P.27, «Neutron Guide»). In the case of a magnetic medium subjected to an external field $\vec{H}$, the factor (1-n) contains 2 terms that are added up if the spin of the neutron $\vec{S}$ is parallel to the induction $\vec{B}$ (critical angle $\varphi_c^{(+)}$) and are cancelled out if spin and induction are antiparallel ($\varphi_c^{(-)}$). If these terms are approximately equal $\varphi_c^{(-)} \simeq 0$; for all incidences such that $0 < \varphi < \varphi_c^{(+)}$ only neutrons with $S_z^{(+)}$ will be reflected.
Please note: In this case, the reflected beam is not monochromatic.

3) Filtering of spins by polarised ^{3}He

This very promising method, presently under development at ILL (Grenoble), relies on the fact that the capture cross-section of ^{3}He, an isotope of the helium atom with 3 nucleons (2 protons and 1 neutron), depends on the spin of the incident neutron. If the latter can couple to the spin of a single nucleon to form a pair with a zero total spin (antiparallel spins), the capture cross-section is considerable; it is 1000 times weaker in the opposite case. When a neutron beam propagates through a volume of polarised ^{3}He (all nuclei have // spins), 1000 neutrons $S_z^{(-)}$ are captured whereas only one $S_z^{(+)}$ neutron disappears.

Coherent and incoherent scattering

Let's consider the interaction of a slow neutron ($\lambda_0 > 10^{-2}$ nm) with the atoms of a perfect crystal, in other words with a large number of scatterers placed at the nodes of a regular lattice. Since each incident particle has a wavelength that is comparable to the distance between 2 scatterers, the neutron is not going to interact with one specific atom but with all the scatterers in the sample.

In wave mechanics, the wave function of the incident neutron is a plane wave. This wave is scattered by each of the nuclei in the sample which, according to Fermi's hypotheses (cf. P.16), act as secondary sources: they reemit a spherical wave that is out of phase with respect to the incident wave by an amount that is proportional to b_l, the scattering length of nucleus l. This representation, originating in the optical theory of the propagation and scattering of waves developed since Huyghens' work («Treaty of Light», 1690), is applicable to any kind of wave; it allows us to understand an essential point concerning neutron scattering: the simultaneous presence in the detected signal of 2 components, one «coherent», the other «incoherent»:

- If all scatterers in our perfect crystal are strictly identical (same isotope of the same chemical species) and if also the nuclei have no spin, then $b_l = b$ whatever l is and the reemitted waves exhibit a definite phase relationship among themselves. At one point situated far from the sample all these waves interfere; the amplitude A of the resulting wave, obtained as the sum of all contributions, is non zero only in the directions in which the path difference between the two secondary waves is an integral multiple of 2π (Bragg direction); A is thus equal to the sum of the scattering lengths (or scattering amplitude):

$$A = \sum_{l=1}^{N} b_l = Nb$$

  All the scatterers contribute to the final result in a «coherent» manner. The scattering length of each nucleus is called coherent, $b_{coh}=b$. The number of scattered neutrons in the Bragg direction (coherent differential scattering cross-section) is given by the square of the amplitude of the neutron wave function. It is proportional to $(b_{coh})^2$.

- Let us now suppose that the scattering lengths fluctuate in a random way from site to site, around a mean value:

$$b_l = \bar{b} + \delta b_l \text{ with } \sum_{l=1}^{N} \delta b_l = 0$$

The system could thus be represented schematically by assuming that 2 scatterers at the same position occupy each site :

- one scatterer having a scattering length $\bar{b}$. This subset reemits waves in phase that interfere in an identical way to the preceding case. It produces a coherent component with $b_{coh} = \bar{b}$.
- a scatterer having a scattering length δb_l that varies randomly from site to site, thus with no correlation between the values of δb on 2 neighbouring sites. As a result, the phases of the waves re-emitted by these scatterers are also at random and, like light issued from 2 incoherent sources, it produces no interference. The resulting intensity is the sum of the intensities, in other words the sum of the squares of the amplitudes. Moreover, since the intensity emitted by each scatterer is isotropic, the same holds for the overall intensity. This random fluctuation of the scattering length produces an incoherent component. Its intensity $(d\sigma/d\Omega)_{inc}$ is isotropic and proportional to:

$$K^2 = \sum_{l=1}^{N} \delta b_l^{\,2} = N\left[(b_l - \bar{b})^2\right] = N\left[\overline{b_l^{\,2}} - (\bar{b})^2\right]$$

In comparison, we define an incoherent scattering length (per nucleus):

$$b_{inc} = \sqrt{\frac{K^2}{N}} = \sqrt{\overline{b^2} - \overline{(b)}^2}$$

We have thus the addition of a coherent and an incoherent component. This result is very general; it is so for the elastic as well as for the inelastic part of the scattered beam. As we have attempted to explain, the coherent component is the result of a collective process in which all the scatterers of the crystal contribute. The law of variation of its intensity with the momentum transfer Q and with energy exchanged $h\omega$ (coherent scattering function $S_{coh}(Q, \omega)$) reflects the spatial correlation and the collective dynamics of the scatterers. On the contrary, each scatterer contributes independently (simple addition of the scattered intensities) to the incoherent component. The corresponding scattering function $S_{inc}(Q, \omega)$ contains only their individual dynamics. The relative magnitude of each of the components is given by the respective values of the average scattering length b and of its fluctuation δb.

In conclusion, let us look at the two causes that bring about a fluctuation of b from site to site:

1) **isotopic disorder:** we have seen that 2 isotopes of the same element in the periodic table do not have the same scattering length. If no particular «care» is taken at the time of preparing the sample, each chemical species present in the sample is a mixture of its isotopes in such a way as they exist in the natural state, the spatial distribution being at random.
For example, if an element has 2 isotopes with concentration C_1 et C_2 ($C_1 + C_2 = 1$) whose respective scattering lengths are b_1 and b_2:

$$\left.\begin{aligned} \overline{b} &= C_1\ b_1 + C_2\ b_2 \\ \overline{b^2} &= C_1\ b_1^2 + C_2\ b_2^2 \end{aligned}\right\} \quad (\delta b)^2 = C_1\ C_2\ (b_1 - b_2)^2$$

2) **the nuclear spin:** the neutron has a spin 1/2. The nuclear forces responsible for neutron scattering by a nucleus depend, if the latter has spin **i** (nuclear spin), on the mutual orientation of their spins. In this case, there are 2 scattering lengths $b^{(+)}$ and $b^{(-)}$ according to whether these spins are «parallel» (total spin **i** + 1/2) or «antiparallel» (total spin **i** – 1/2). If neither the target nor the incident beam is polarised, the passage via one or the other of these total spin states is at random. Finally, everything happens as if scattering lengths $b^{(+)}$ et $b^{(-)}$ were randomly distributed on the scatterer lattice, in proportion to the number of corresponding states (there are (2s+1) states with total spin s). One can thus show that:

$$\overline{b} = \frac{\mathbf{i} + 1}{2\mathbf{i} + 1}\ b^{(+)} + \frac{\mathbf{i}}{2\mathbf{i} + 1}\ b^{(-)}$$

$$(\delta b)^2 = \frac{\mathbf{i}\,(\mathbf{i} + 1)}{(2\mathbf{i} + 1)^2}\ (b^{(+)} - b^{(-)})^2$$

Note: the distinction between «coherent» and «incoherent» in the expression of the effective scattering cross-section is not always obvious and is sometimes a source of confusion. For example: the chemical disorder in an alloy produces a fluctuation in the scattered length from site to site and, therefore, adds incoherent terms; on the other hand, a position disorder modifies the correlations and therefore the coherent terms. Since experimentally these two types of disorders both manifest themselves in a similar fashion by the appearance of a scattered intensity between the Bragg directions, one frequently speaks of «diffuse» scattering.

The

source and the output beams

The neutron is a difficult particle to produce. Its high production cost is due, on one hand to the harmful nature of the radiation generated: protection must be provided, fission products must be properly confined and stored; on the other hand to the many security elements and circuits that are required to insure the safety of the plant installation (prevention of accidents). The number, or more precisely the maximum flux of neutrons (number of particles crossing a unit area per second) that can be generated is limited by the amount of heat (by-product of fission) per unit volume of fuel that can be consumed. For example, the RHF in Grenoble, the highest performance reactor in the world, produces a flux of $1.5*10^{15}$ neutrons/cm^2.s, a figure to be compared to the flux delivered by a medium powered laser (10^{20} photons/cm^2.s) or to that supplied by a laboratory X-ray generator (10^{18} photons/cm^2.s).
The development of a neutron scattering experiment involves several steps and the use of several experimental systems; it is necessary:

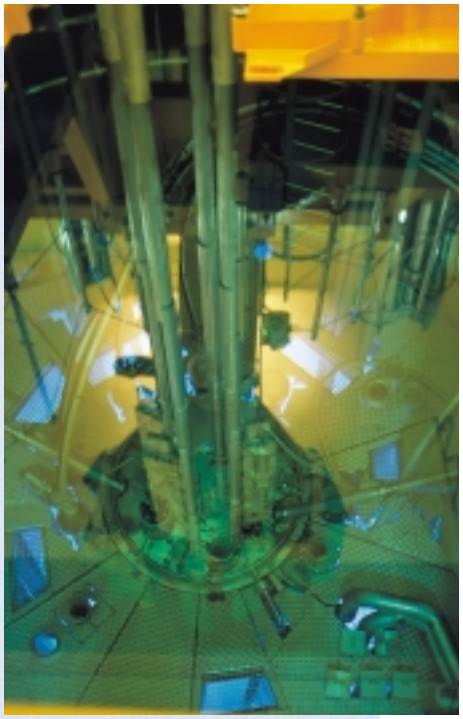
- to produce a neutron flux that is as intense as possible (fission of the uranium nuclei in the core),
- to adjust the energy of the produced particles so that they are compatible with the energy scale of the phenomena under study (thermalization),
- to select, with the least possible loss, all neutrons having the same propagation direction (collimation) and a chosen energy (monochromatization),
- to measure, after having interacted with the sample, the proportion of those neutrons that are deviated and whose trajectory makes an angle (2θ) with the initial direction (angular analysis) and/or those whose final energy has varied (energy analysis) by a specific amount ΔE.

1 - The Orphée reactor

Orphée is a fission reactor designed to furnish neutron beams needed for fundamental research.

Principal characteristics

Maximal thermal flux in the reflector (n cm^{-2}s^{-1})	-	3.10^{14}
Power of the core (MW)	-	14
Fluid thermal exchange	-	H_2O
Exchange surface (m^2)	-	20,68
Total volume of the core (dm^3)	-	56
Active height (cm)	-	90
Power delivered in the fuel elements (MW)	-	12,6
Heat flux (W cm^{-2}) :		
medium	-	61
maximum	-	172
Maximal heat flux in the hot water channel (W cm^{-2})	-	206
Maximum temperature of the lining (° C)	-	123,5
Power density in a fuel element (MW dm^{-3}) :		
medium	-	0,25
maximum	-	1,2
Core pressure (bar):		
entrance	-	4
exit	-	2
Water speed (m s^{-1})	-	7,5
Charge of ^{235}U (kg)	-	5,88
Duration of the cycle (days)	-	100
Rate of average burn-up (% ^{235}U burned)	-	30



Pool around the core

❑ The core

The core is very compact; it is housed in a parallelepipedic enclosure made of zircaloy in a squared section (25 x 25 cm^2); its active height is 90 cm.

It consists of 8 assemblies of parallel plates (fuel elements) that are made from a fissile material (an aluminium and uranium alloy, the latter enriched with ^{235}U) and that are arranged around a central beryllium reflector.

The fine division of the fuel elements into thin plates (1.27 mm) separated by narrow channels of water (2.1 mm) produces a very large surface for thermal exchange per unit volume (on the order of 0.6 m^2 per dm^3), therefore an elevated specific power. This is, in turn, a condition for the production of a significant neutron flux.

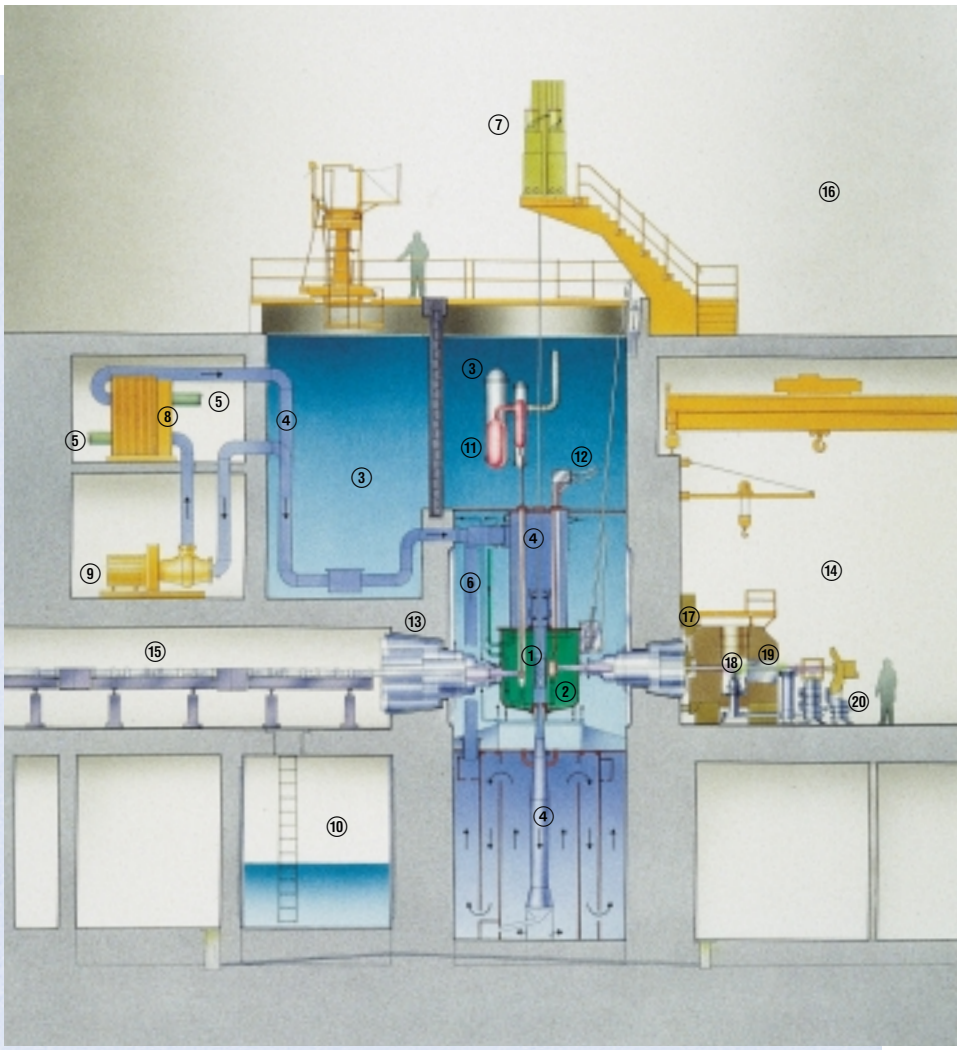
The total mass of uranium 235 of the core is less than 6 kg.

The core is renewed every 100 days.

The control of the reactivity is accomplished by means of neutron absorbing plates (Hafnium), moving vertically into the control rods.

The core is arranged in a reflector of heavy water circulating from bottom to top in a stainless steel vat. The biological protection is ensured by light water, contained in a pool measuring 15 m high and with a 4.5 m diameter; the pool is surrounded by a concrete wall 1.50 m thick. The total diameter of the reactor block is 7.50 m.

1. Core
2. Heavy water reflector
3. Pool and transfer channel
4. Primary circuit
5. Secondary circuit
6. Heavy water circuit
7. Command mechanism of the control rods
8. Exchanger
9. Pump
10. Drainage tank for the pool
11. Cold source
12. Hot source
13. Tangential tubes
14. Experimental hall
15. Neutron guide
16. Hall for access to reactor pool
17. Fixed primary protection
18. Monochromator
19. Protection of the monochromator
20. Spectrometer



Reactor block, side view

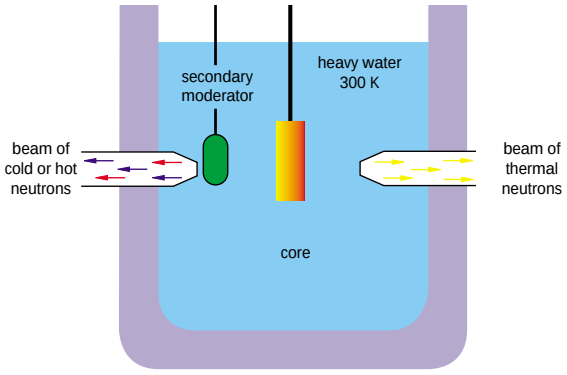
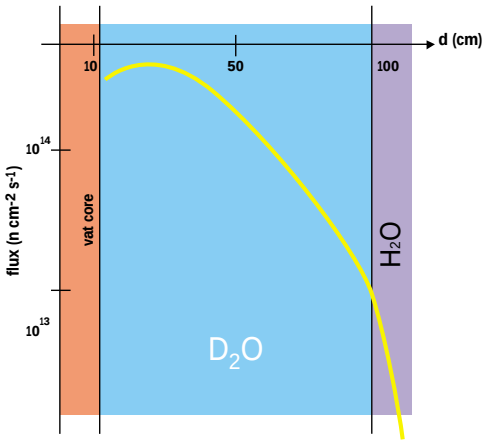


Diagram of the moderator



Flux of thermal neutrons as a function of the distance from the core

❑ The thermalization of neutrons

The moderator

The fission chain reaction in the core breaks the Uranium 235 nuclei into lighter elements and liberates, on average, 2.4 neutrons for every fissioned uranium atom. These neutrons have a kinetic energy of about 1 million electron-volt, much too high to be used for scatterring studies in condensed matter physics. To slow them down, they are put in contact with a special material (moderator) where, through successive collisions with the atoms, they give up most of their energy. The materials that are best adapted to this role are made of light atoms: water, graphite, beryllium,... Moreover, it is desirable to minimise the events where, during the course of a collision, the moderator atom captures the neutron; this condition results in the selection of heavy water to surround the core as the choice of a moderator for Orphée. The fission neutrons are slowed down and after several collisions, they have an average energy (speed) comparable to the kinetic energy of the slowing atoms (~0.025 eV for a moderator at 300 K). The tubes through which the «thermal» neutrons are taken extend deep into the heavy water moderator.

Cold and hot sources

For some experiments, it is desirable to have either a source of neutrons with lower energy (~0.001 eV), or with higher energy (~1 eV). Such neutrons may be obtained due to secondary moderators that, placed in heavy water, create local conditions which modify the average energy (speed) of the neutrons. A container filled with liquid hydrogen (temperature 20 K) constitutes a source of slow neutrons; a block of graphite heated to 1400 K provides neutrons of high energy. These are respectively called cold and hot sources.

Ring-like annular cold source

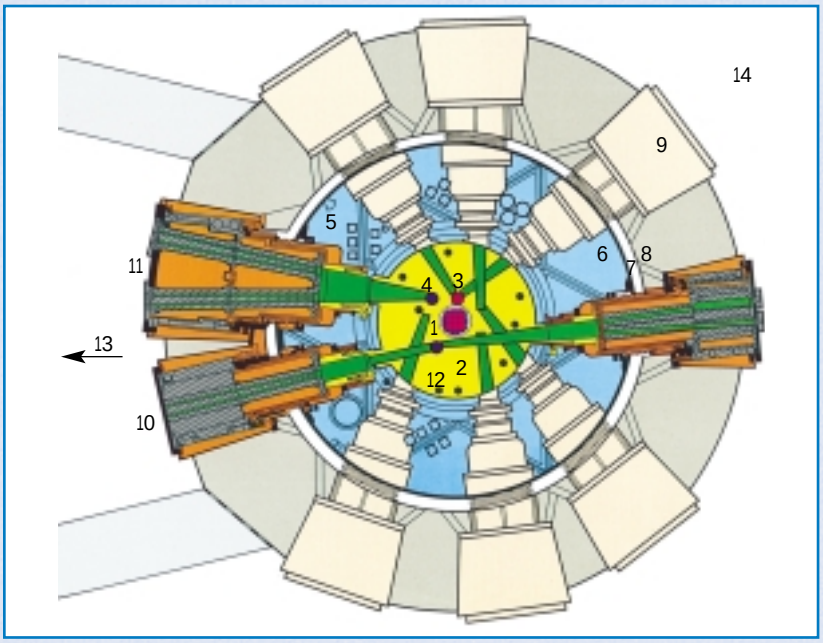
❑ The tubes

The reactor is equipped with 9 horizontal tubes, tangential to the core, allowing the use of 20 neutron beams. The «nose» of these tubes is situated in the moderator near the core, where the flux of thermalized neutrons is maximum; three tubes are aimed at two «cold sources», two other tubes at a «hot source».
It is thus possible to select the spectrum of neutrons that is best adapted to the desired use.

Six cold beams are extracted by «neutron guides» emerging from the reactor building so that they may be utilised in an adjoining hall (neutron guide hall).

Nine vertical tubes are used to irradiate different samples for analysis by means of activation. In the latter case, the samples are sent by a pneumatic connection to the Pierre Sue Laboratory, a joint laboratory of the CEA and the CNRS.

1. Core
2. Heavy water reflector
3. Hot source
4. Cold source
5. Pool
6. Pool inner wall
7. Annular space
8. Pool outer wall
9. Single tube
10. Single tube
11. Double tube
12. Vertical tube
13. Neutron guide hall
14. Experimental hall

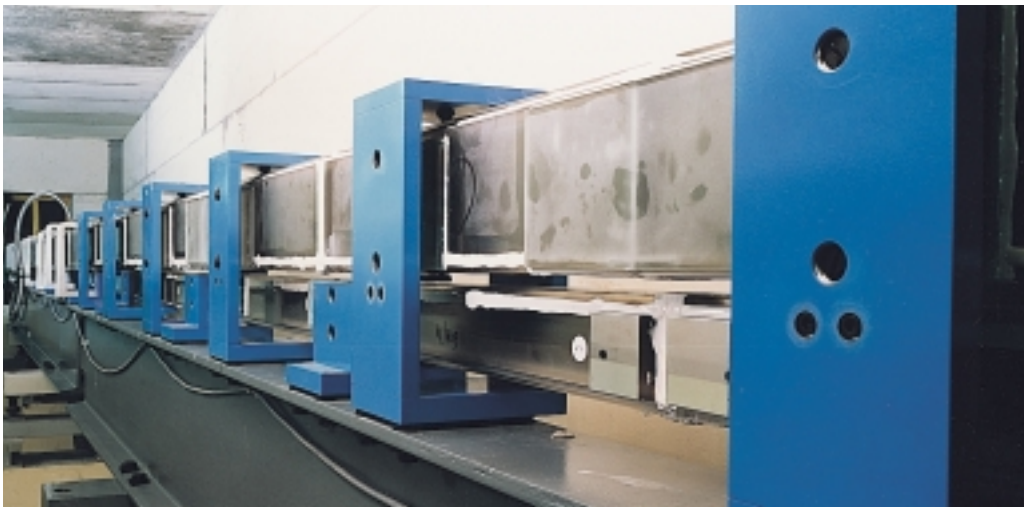


Pile block, horizontal cross-section

Control room of the Orphée reactor

2 - The output beams

❑ The neutron guide



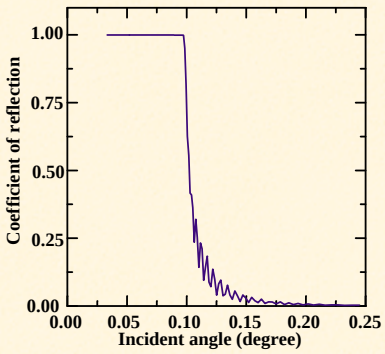
Guide G1 and bender G1bis (below)

The first component in the majority of neutron scattering instruments is a monochromator. Its function is to select, from the polychromatic beam extracted from the moderator, the neutrons that have a wavelength within a relatively narrow set band and to direct these neutrons onto the sample. The other neutrons of the beam, that is to say between 90% and 99% of the total neutrons, pass through the monochromator and are lost in the protective concrete! Clearly, an arrangement which would allow several spectrometers to extract different wavelengths from the same beam, would be a more rational utilisation of the neutrons produced. However spectrometers are huge instruments and this idea would only be feasible if the spectrometers were aligned one behind the other and thus, farther and farther away from the source. Thus arranged, successive instruments would be less and less luminous, remembering that neutrons are neutral particles that behave as a perfect gas: the flux of particles at distance d from the source is proportional to $1/d^2$ (solid angle under which the source subtends). Neutron guides, by «channelling» particles, permit distribution of beams with constant angular divergence far from the core, in other words without any loss of flux.

Guide coating by sputtering.
General view of the machine used for producing supermirrors coating (CILAS photography).

Guide with simple total reflection

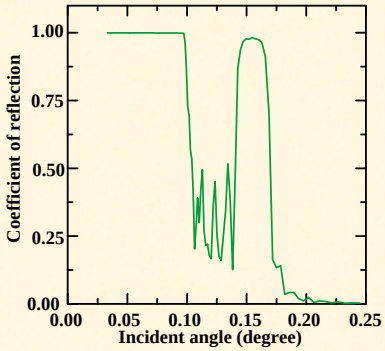
The propagation characteristics of the wave associated with a neutron involves the refractive index «n» of the medium, which itself depends on the nature of the atoms that make it up. At an interface, the passage from a medium with index «n_1» to a medium with index «n_2» will involve a change in the direction of propagation and, under certain conditions ($n_2<n_1$; incident angle < critical angle), the wave will not be able to pass through: it will undergo total reflection. The critical angle depends on the difference (n_2-n_1) and on the wavelength of the neutron. This phenomenon, well known for electromagnetic waves (optical fibers), is utilised to transport neutrons without loss over distances covering several tens of meters. The guide is a hollow tube made of thick glass whose internal walls are polished and covered with a layer of nickel. However, the index of this material, although one of the best, is only slightly different from the vacuum index and the critical angle of total reflection is small (0.5° for a wavelength of 0.5 nm).



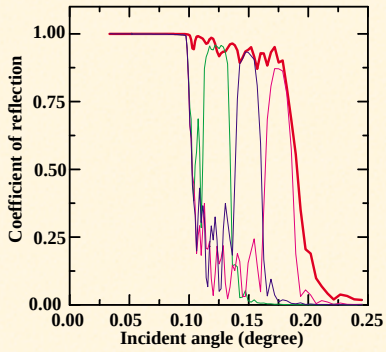
Reflection by a thick layer of nickel (300 nm).
All neutrons arriving at the surface with an angle less than the critical angle are reflected. This is the plateau of total reflection. For greater incident angles, the neutrons are partially transmitted, partially reflected.

Guide with supermirrors

In order to increase the performance of the guides, scientists use constructive interference between the waves reflected by alternating layers of controlled thickness, which creates a succession of diffraction peaks beyond the critical angle. Present day technology allows the deposition of multilayers of nickel-titanium giving guides with an effective critical angle 2 or 3 times that of a simple total reflection guide.



Reflection by a periodic multilayer made of layers of a reflective material like nickel (11 nm)) and a spacing material like titanium (11 nm) topped by a thick layer of nickel.
As in the preceding case, a plateau of total reflection is found. Superimposed on this plateau, the constructive interference between the waves reflected by the layers of nickel creates a reflection peak for a particular incident angle that is higher than the critical angle. This effect will be used to extend the plateau of total reflection.



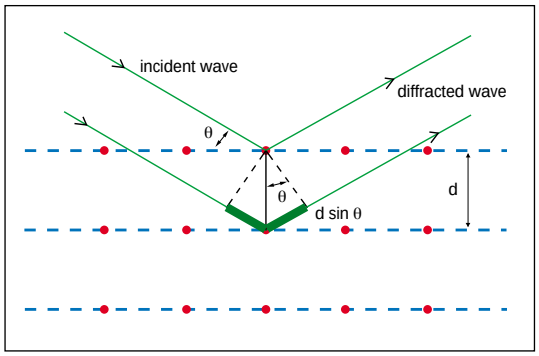
Reflection by a non-periodic multilayer of nickel and of titanium topped by a thick layer of nickel.
In this way, we are able to create a series of peaks beyond the plateau of total reflection. The sum of these peaks gives an extension to the total reflection plateau. Thus the apparent critical angle obtained is much larger.
The longer the desired extension, the greater the number of layers needed and the thinner each layer must be.

❑ Neutron selection

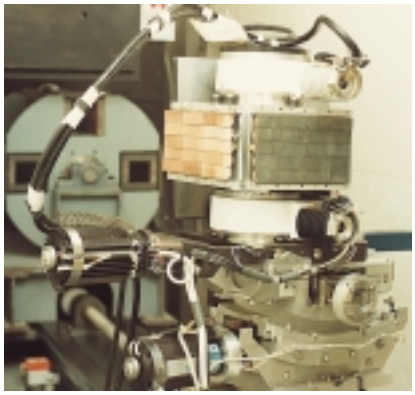
Monochromators

Wave-particle duality makes possible two families of monochromators:

- **Single crystals, which operate on the principle of wave diffraction by a periodic system.**



Bragg reflection from a periodic lattice of atoms.

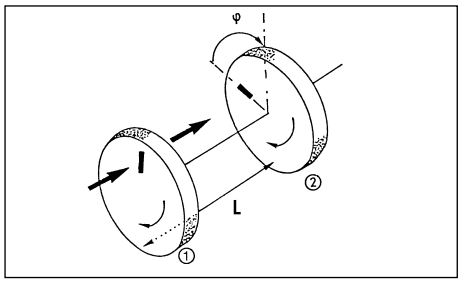


Setup allowing a choice between different crystal monochromators. (ILL photography).

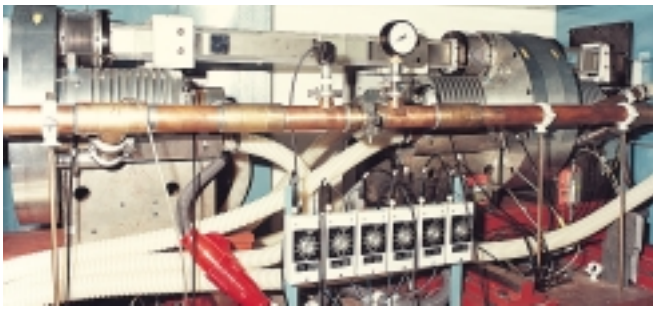
If d is the periodicity of the material, in the plane of incidence only neutrons having a wavelength (λ) obeying the Bragg relation $n\lambda = 2d\sin\theta$ will be reflected in phase. The most used crystals are copper, germanium, certain alloys and pyrolitic graphite.

If all the reflection planes are perfectly parallel to each other, only a few incident neutrons will verify the Bragg relation: the diffracted beam will be highly resolved but will have a low intensity. In order to increase the «reflectivity», crystals are used that have atom planes oriented with a certain amount of disorder (mosaicity). The most currently utilised crystals (copper, germanium, pyrolitic graphite) have mosaicities between 0.5 and 1°.

- **Mechanical systems, which select particles according to their velocity.**

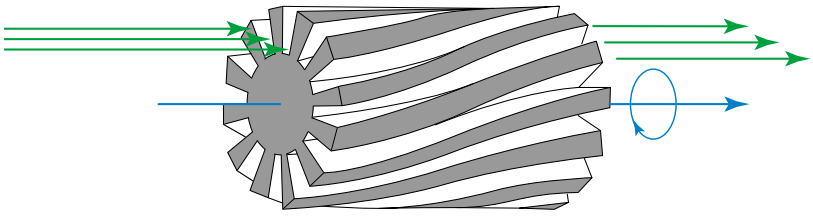


Chopper monochromator.



End section of the chopper monochromator «Mibemol».

Both disks are made of an absorbent material and have a transparent slit. The disks are separated by a distance L and turn at the same angular speed ω. If the slits form a constant angle φ among themselves, only neutrons that have a speed $V \simeq \omega L/\varphi$ are transmitted.



Mechanical selector.

On a cylinder of length L, one traces spiral grooves (pitch of spiral = p). If the cylinder turns on its axis at an angular speed ω, each groove transmits only neutrons having a speed $V \simeq p\,\omega / 2\pi$.

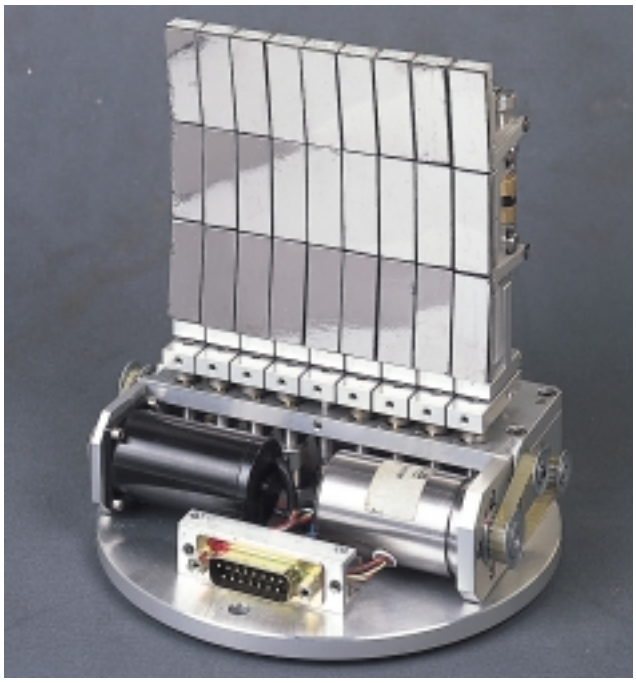
The mechanical selector is well adapted to the production of long wavelength beams ($\lambda_0 > 0{,}6$ nm). It also permits, when a good energy resolution is not necessary (for example for small angle scattering), an increase in the available flux by producing a barely monochromatic beam ($\Delta V/V \simeq 10$ to 20%).

Focusing monochromators

The source (the volume of moderator facing the nose of the tube) is extended, so one is able to extract large dimension beams: at Orphée approximately 24*90 mm^2.

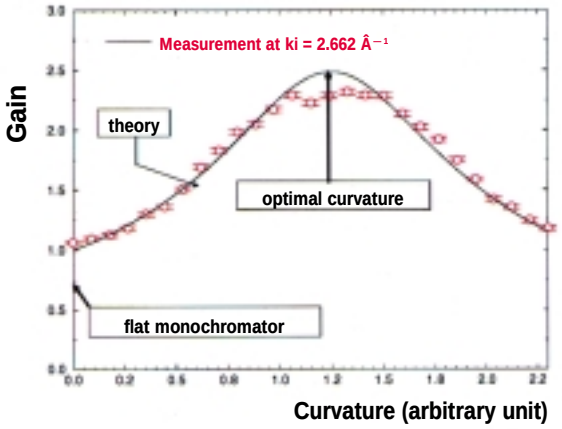
The focusing monochromator «concentrates» the monochromatic beam with a minimal loss of quality and thus increases the flux arriving on the sample.

It is based on the principle of wave reflection by a curved surface (in analogy with the focusing of a light beam).
But for neutrons, the Bragg condition in the incident plane should be taken into account. Because of this, the focusing conditions will depend on the wavelength.

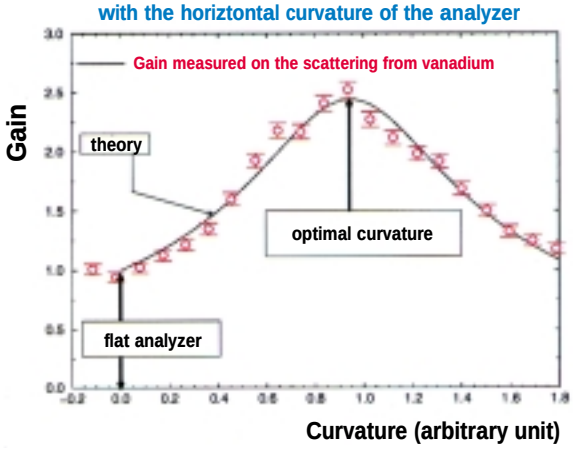


Variable focusing setup in two planes. (1 T analyzer).
Assembly of planar crystals with adjustable orientation allowing the variation of the curvature of the whole assembly.

Intensity gain
with the vertical curvature of the monochromator



Intensity gain
with the horiztontal curvature of the analyzer



Focusing conditions

- vertical

$$\frac{1}{R_V} = \frac{1}{2\sin\theta_0}\left(\frac{1}{Z_1} + \frac{1}{Z_2}\right)$$

- horizontal

$$\frac{1}{R_H} = \frac{\sin\theta_0}{2}\left(\frac{1}{Z_1} + \frac{1}{Z_2}\right)$$

with :
R_V radius of vertical curvature
R_H radius of horizontal curvature
Z_1 distance source-monochromator
Z_2 distance focal point-monochromator
θ_0 Bragg angle

❑ Neutron detection

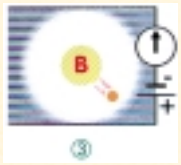
Detectors

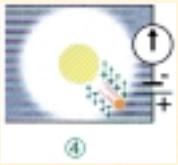
After having interacted with various components (nuclei and/or electronic spins) of the sample, the neutron has a different propagation direction and energy than it had initially. It is from the magnitude of these changes and from the proportion of neutrons that have undergone them that information can be obtained on atomic distances and movements present in the sample under study. The final component of the spectrometer is therefore a detector, which counts the number of neutrons that it receives (irrespective of their energies, this parameter being measured separately by a peripheral device).

Carrying no charge, the neutron creates no ions; therefore it can only be detected by activation of a nuclear reaction. The majority of neutron detectors work on the same principle: a chamber filled with a gas in which a constituent heavily absorbs thermal neutrons and emits charged particles; an electric field accelerates this charge which, by colliding with other gas atoms, ionises them, thus producing secondary electrons (amplification); the cathode collects these electrons and generates an electric impulse that can be detected.

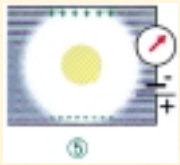
The six steps in the detection of thermal neutrons:

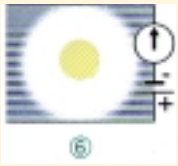
The absorption of a neutron by nucleus A





causes an ionising particle to be emitted which, accelerated by an electric field, creates secondary charges.





Collected by the electrodes, these charges generate a current impulse that is then detected.

Requirements for nucleus A:

- **must be a gas molecule at ambient temperature,**
- **must «like» to capture neutrons,**
- **must emit a very ionising particle.**

Two good candidates:

$^{10}B + n \Rightarrow {}^{7}Li + {}^{4}He$ (gaslike in the form of BF_3)

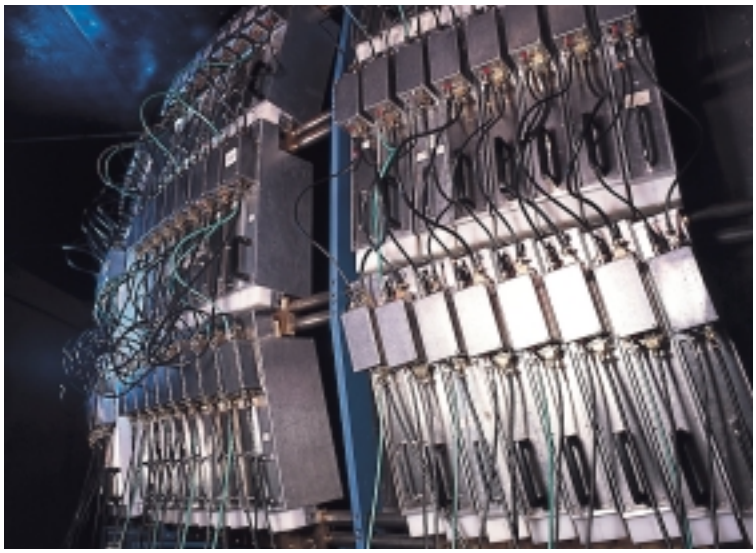
- **Advantages: highly efficient, inexpensive.**
- **Disadvantage: poisonous gas.**

$^{3}He + n \Rightarrow {}^{3}T + p$ (gaseous in standard conditions)

- **Advantages: highly efficient, non toxic.**
- **Disadvantage: very expensive, very sensitive to impurities.**

Multidetectors

After scattering by the sample, the directions of propagation of the neutrons are distributed over all directions. The measurement of their intensity distribution $I(2\theta)$ allows the scientist to obtain the particular order that characterises the scatterer under study (its structure), provided that a wide angular region is explored. The experiment can be performed even more quickly (or more precisely according to the choice of the researcher), if it is designed to measure this intensity simultaneously in several directions.



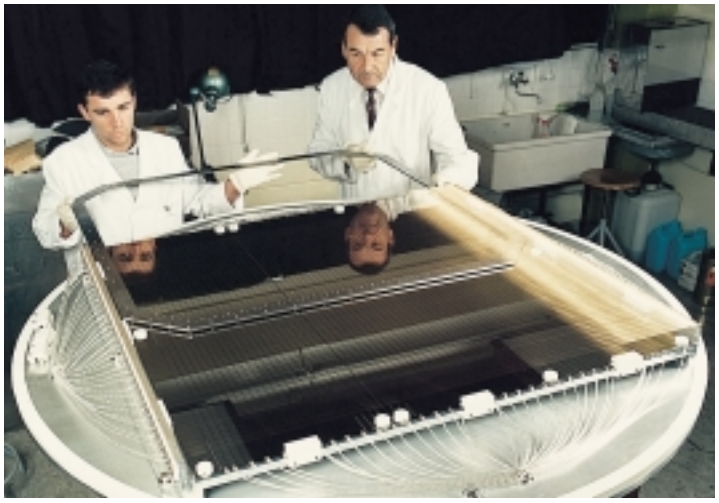
One method juxtaposes many individual detectors.

Bank of detectors of the Mibemol spectrometer

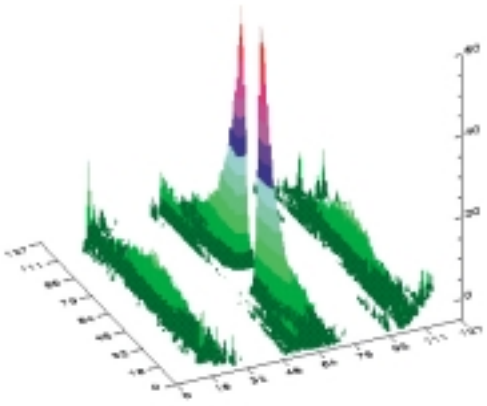
Another possibility is to devise an extended detector that, by its internal design, gives the position of the absorbing atom from which the detected ionising particle originated.
These are called multidetectors or PSD (Position Sensitive Detector).

There are several types:

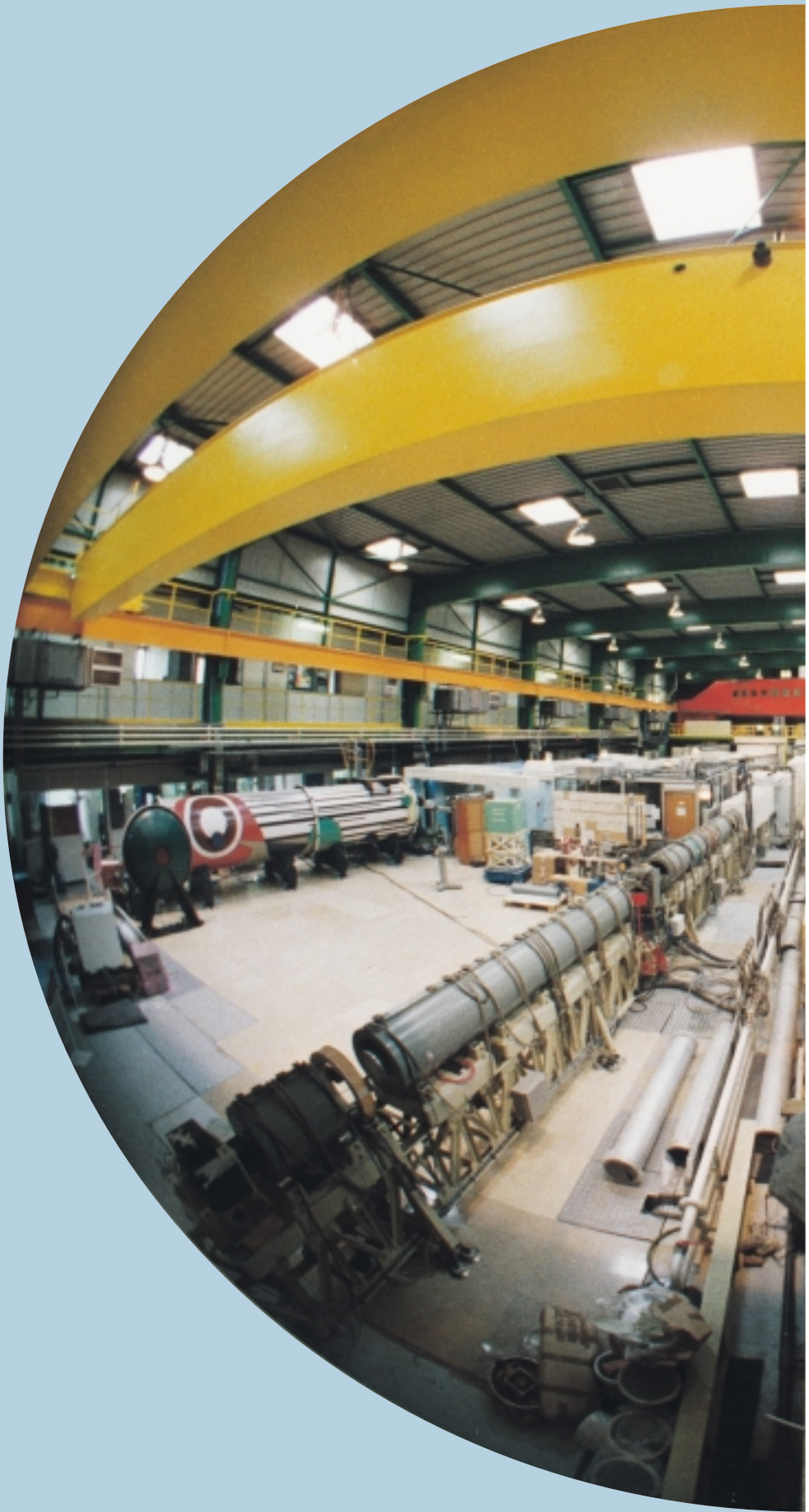
- one dimensional, pinpointing the position of the impact (linear) or the angular position (banana type),
- two dimensional in a plane where, thanks to a network of perpendicular wires, the abscissa and the ordinate of the absorption point of the neutron (detector XY) can be determined.



An XY multidetector of 64x64 cells, during the process of being assembled (ILL photography).



3D representation of the number of neutrons collected in each cell of an XY planar multidetector of 128 x 128 cells (scattering by a liquid cristal polymer).



Spectrometers

The scattering of thermal neutrons by condensed matter is governed by conservation laws (momentum, energy, magnetic moment,...) applied to the whole system (scatterers and neutrons). The purpose of the spectrometer is to measure the variation of these different quantities for each scattered neutron.

The majority of spectrometers that are set up around a continuous source, such as Orphée reactor, are based on the same principles:

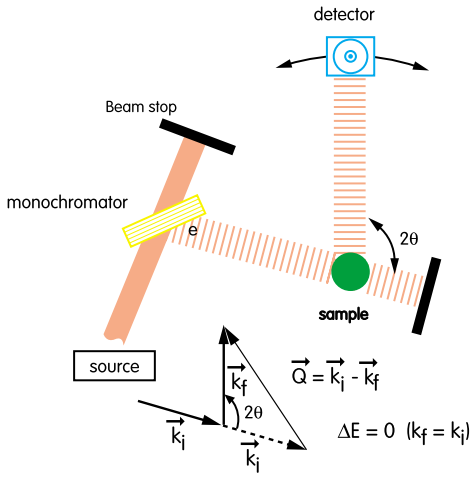
- to prepare, with the intention of «bombarding» the sample, a beam of neutrons that all have the same propagation direction $\vec{k_i}$, the same energy E_i and, in the case of a polarized beam, the same spin state $\vec{\sigma_i}$;
- to measure after scattering from the sample the proportion of neutrons that have the propagation direction $\vec{k_f}$, the energy E_f and, eventually, the spin state $\vec{\sigma_f}$.

The different types of spectrometers that we are now going to briefly describe differ in the nature and number of final parameters that are measured, in the techniques used, or in the range of values ($\vec{k}$,E) for which they have been designed.

1 - The diffractometers

This class of instruments measures the number of neutrons having undergone a change $2\theta = (\vec{k_f}, \vec{k_i})$ in their propagation direction due to the interaction with the scatterers in the sample. They allow to measure the average atomic and magnetic structure of a sample.

A parallel and monochromatic beam (wavelength λ_0) is sent onto the sample. The detector moves on a circle centred at the sample. Each position of the detector defines a propagation direction of the emerging neutrons (angle 2θ with the initial direction).
The recorded intensity measures the number of neutrons having undergone a momentum change $|Q| = |\vec{k_i} - \vec{k_f}| = \frac{4\pi}{\lambda_0} \sin\theta$ (assuming a purely elastic scattering).

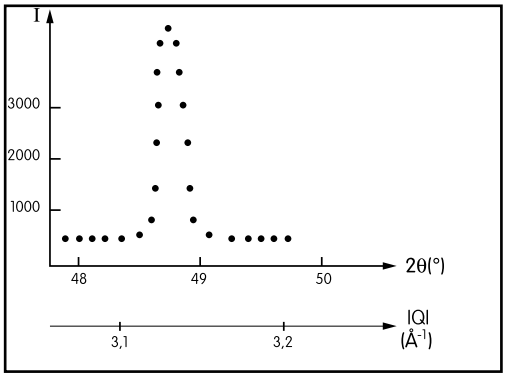


Schematic representation of a diffractometer

- The directions in which scattering is intense (Bragg peaks) correspond to the «long range order of the average structure»: a lattice of atomic planes separated by a distance (d) will induce a maximum of intensity in the direction $2\theta = 2 \text{ arc sin } (\frac{\lambda}{2d})$.

In the case of a crystal (periodic lattice in three dimensions $\vec{a}$, $\vec{b}$, $\vec{c}$), the Bragg directions are given by the vectors of the «reciprocal lattice»: $\vec{Q} \equiv \vec{G} \equiv h\vec{a}^* + k\vec{b}^* + l\vec{c}^*$

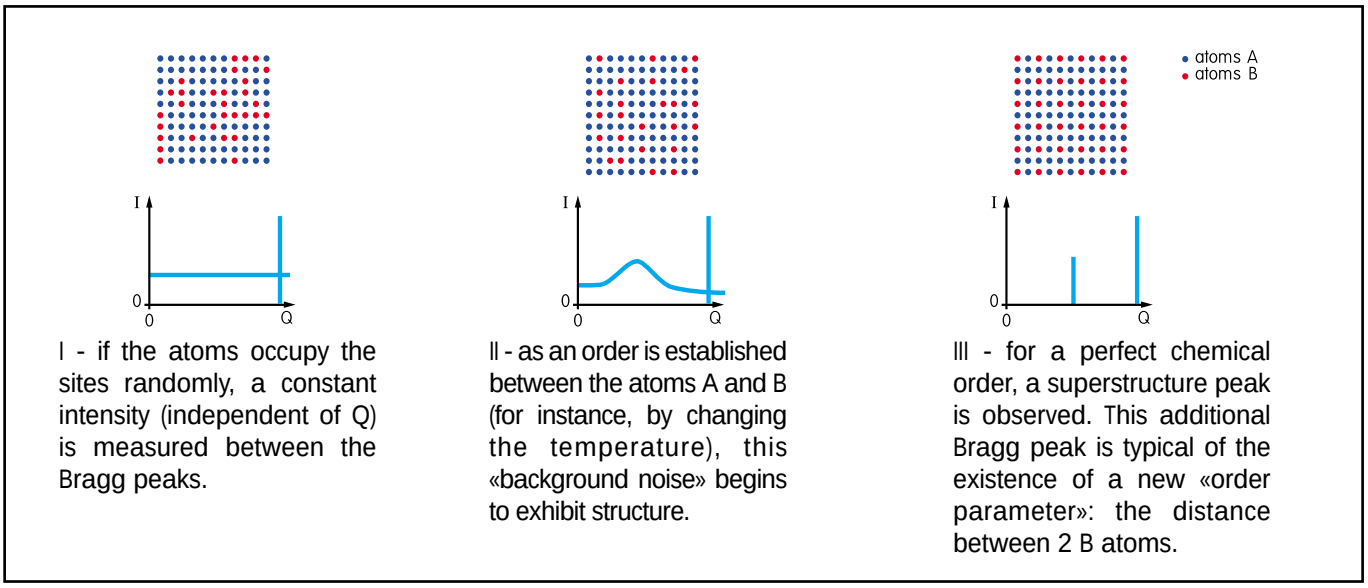
The relative intensity of the successive peaks (h,k,l) enables the scientist to determine the position of all the atoms in the unit cell.



Scattered intensity curve I= f(2θ), measured by the detector when passing through a Bragg reflection.

❑ Deviations from the perfect order are, according to their nature, revealed by different modifications in the angular distribution of the scattered neutrons.

In a crystalline alloy A_3 B the distribution at each site of the atoms A (•) and B (•) may be more or less random (chemical order or disorder):



I - if the atoms occupy the sites randomly, a constant intensity (independent of Q) is measured between the Bragg peaks.

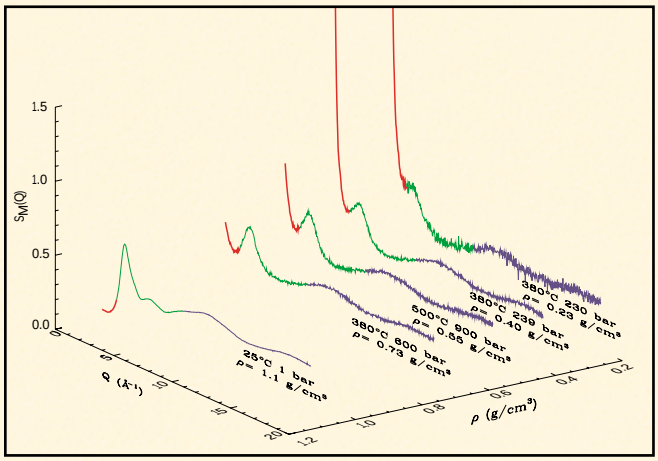
II - as an order is established between the atoms A and B (for instance, by changing the temperature), this «background noise» begins to exhibit structure.

III - for a perfect chemical order, a superstructure peak is observed. This additional Bragg peak is typical of the existence of a new «order parameter»: the distance between 2 B atoms.

Representation of the scattering diagrams of a crystalline alloy A_3 B: influence of a chemical order.

Supercritical Water

In the (Temperature-Pressure) plane, the liquid-gas transition occurs at precise (P, T) points. This transition only exists below the critical point (P_c , T_c); thus, starting in the liquid state, a path in the (P, T) diagram surrounding the critical point allows one to obtain a gas without crossing the boiling curve. The region of the plane ($P > P_c$; $T > T_c$) corresponds to the so-called supercritical fluid state. In their supercritical state, fluids have frequently new physico-chemical properties. For instance, supercritical water is chemically very active: it can degrade by oxidation most organic compounds with production of carbon dioxide and water. This property could be usefull in the treatment of household rubbish.

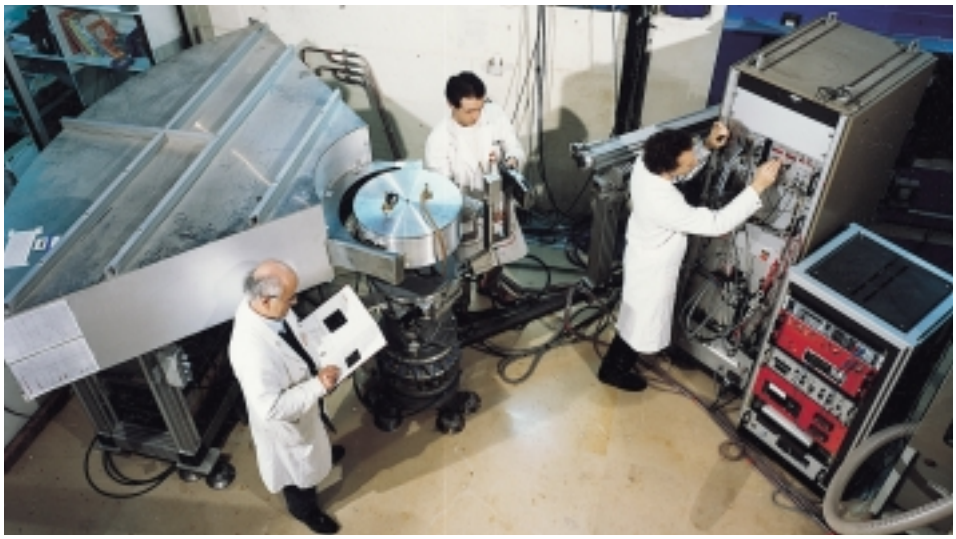


Structure factor of water (D_2O) for various temperatures and pressures. Critical point T_c = 371 °C, P_c = 218 bar, ϱ_c = 0,36 g/cm³.

The evolution with the pressure of the structure factor, measured over a large Q range, leads to 3 results:

- ***Q > 5 Å^{-1} (molecular dimension; in blue)***
  ⇨ ***the curves, which are very similar, show that the structure of the molecule is unchanged when crossing the critical point.***
- ***5 > Q > 1 Å^{-1} (intermolecular distances; in green)***
  ⇨ ***when the density decreases, the main peak becomes larger and shifts; the molecules are less localised and the distance between them increases.***
- ***Q < 1 Å^{-1} (local density; in red)***
  ⇨ ***in the vicinity of the critical point, the strong intensity observed at small q values is due to the onset of density fluctuations (critical scattering).***

❑ Two-axis diffractometer for powders and liquids



The «powder» diffractometer G41 and its 800 cells multidetector

❑ An isotropic powder is made of small crystals that are randomly oriented in every direction. The diffraction pattern does not depend on the direction of the incident beam since, by definition, the angular distribution of small crystals is the same relative to all axes. The scattering is distributed uniformly on a cone of axis parallel to the incident beam and open angle 2θ. The diffraction spectrum (Bragg peaks and/ or diffuse scattering between the peaks) is obtained by measuring the distribution of intensities reaching the detector when it moves in the horizontal plane on a circle centred at the sample (or more efficiently by using a multidetector). A second rotational axis, around the monochromator, allows the scientist to choose the incident wavelength λ_0 and, therefore, the range of momentum transfer $q = |\vec{Q}|$ that is analysed. In one direction, the intensity scattered is the sum of the scattering by each crystallite (powder averaging):

$$I(2\theta) = I(q) = \int_0^{2\pi} I(\vec{Q}) \sin\beta \, d\beta$$

❑ In a textured powder the distribution of the crystallite is non-isotropic. By measuring the scattered intensity versus the relative orientation of the sample with respect to the incident beam, the scientist can deduce the orientational distribution of the crystallite (texture).

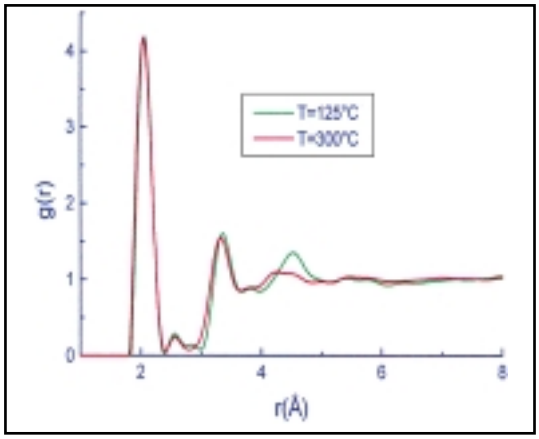
❑ In a liquid, the measurement of the angular distribution of the scattered intensity allows the scientist to obtain the pair distribution function g(r) that characterises the local order of atoms:

$$g(r) = 1 + \frac{1}{2\pi^2 \varrho_0} \int_0^{\infty} q^2 [I(q) - I(\infty)] \frac{\sin qr}{qr} \, dq$$

ϱ_0 = average density

$I(\infty)$ = scattered intensity for very large q

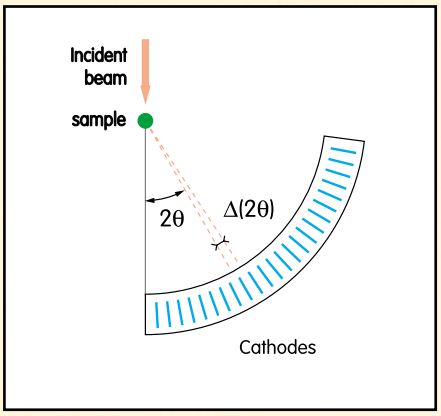
Around 160°C, liquid sulphur exhibits a discontinuity that affects numerous properties: viscosity, density, specific heat,... Measured at 125° and 300°C, g(r) shows the disappearance of a correlation peak around 4.5 Å. These results are well reproduced in a model in which liquid sulphur, made of S_8 rings at low temperatures, transforms into a liquid with long S_n chains at temperatures above 160° C.



Polymerization transition of liquid sulphur.

«Banana» type multidetector

This multidetector allows scientists to measure the distribution of the scattered intensity over a wide angular region in one single step.



In the volume between 2 sections of concentric cylinders (with a common axis perpendicular to the scattering plane and passing through the sample), a series of N cathodes, separated by an angle $\Delta(2\theta)$ have been set. The volume is filled with a detecting gas (BF_3 ou ^3He). Appropriate electronics indicate, for each detected neutron, the specific cathode from which the pulse comes (see P. 30). The content of the channel «n», which represents the number of scattered neutrons within the angle n x $\Delta(2\theta)$, is thus incremented by one unit.

3 types of «banana» multidetectors exist at LLB:

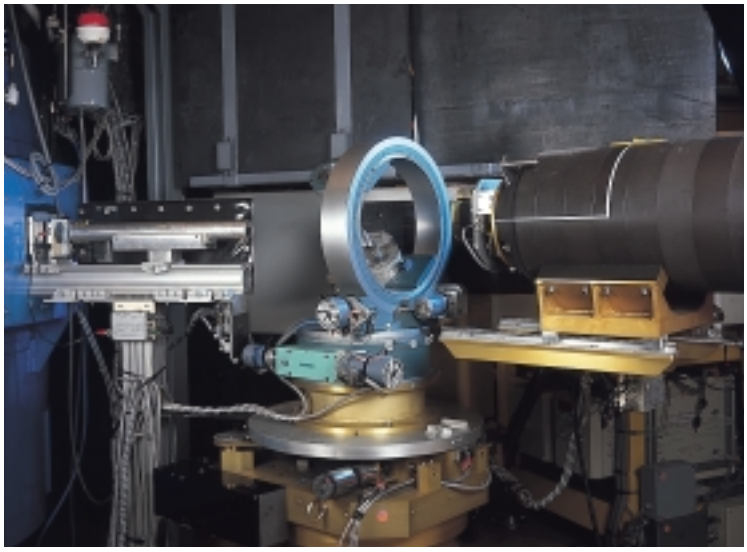
N = 400 cells on 80 degrees $\Rightarrow \Delta(2\theta)$ = 12 arc minutes

N = 800 cells on 80 degrees $\Rightarrow \Delta(2\theta)$ = 6 arc minutes

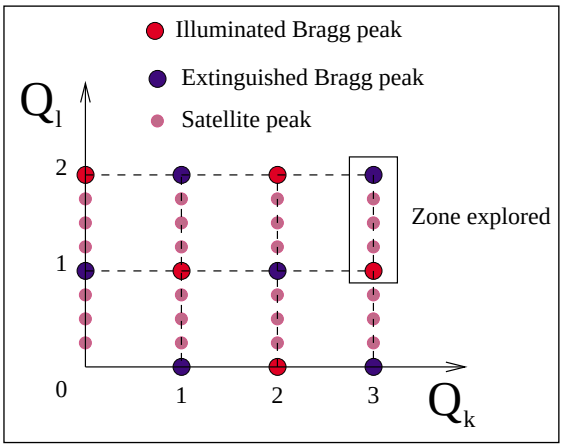
N = 640 cells on 128 degrees $\Rightarrow \Delta(2\theta)$ = 12 arc minutes

❑ Four circle diffractometer for single crystals

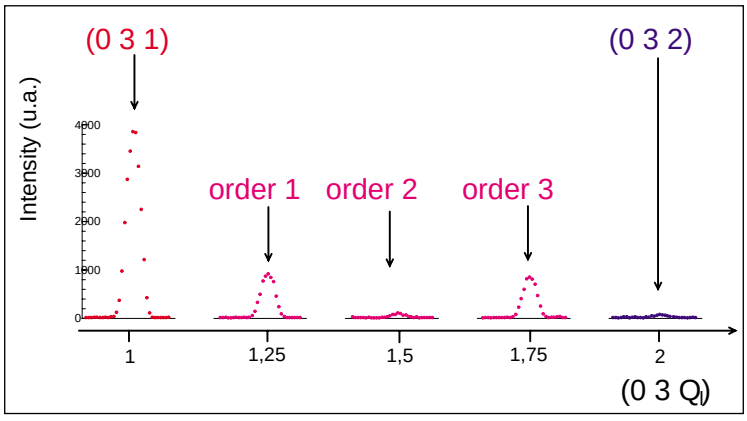
When the sample is a single crystal, the orientation of its crystalline axes relative to the directions $\vec{k_i}$ and $\vec{k_f}$ becomes an essential parameter of the measurement. This is why, besides the axis of rotation around the monochromator (choice of the incident wavelength λ_0), 4 rotation axes are needed that intersect at the sample position: 3 rotations (Euler angles) allow all possible orientations of the crystalline axes relative to the incident direction $\vec{k_i}$; the rotation of the detector defines the scattering angle 2θ.



Diffractometer 6T2



Modulated structure of calcium and dihydrated betaïne chloride (BCCD).



$\vec{Q}$ scan along the $\vec{c^*}$ axis between Bragg peaks (0 3 1) and (0 3 2) in the modulated phase $\vec{q} = \frac{1}{4}\vec{c^*}$ of BCCD at T = 100 K: one can see intense satellite peaks of order 3. The measurement of the intensity of the diffraction on a single crystal allows scientists to refine the structure in real space.

2 - Spectrometers for inelastic scattering

If, in addition to the average structure, the scientist wants to study the dynamics of the atoms or of the magnetic moments in the sample, the spectrometer must measure the number of neutrons that have undergone an energy change $\hbar\omega$ during the scattering process; that is to say the fraction of particles that, arriving on the sample with energy E_i, leaves in the direction $\vec{k}_f$ (forming angle ψ with direction $\vec{k}_i$ of the incident neutrons) with energy E_f (conservation law $\hbar\omega = E_i - E_f$). However, there are no physical phenomena that measure (or select) directly the energy of neutrons:

Neutron = wave ➡ energy ⇔ wavelength λ
Neutron = particle ➡ energy ⇔ speed V

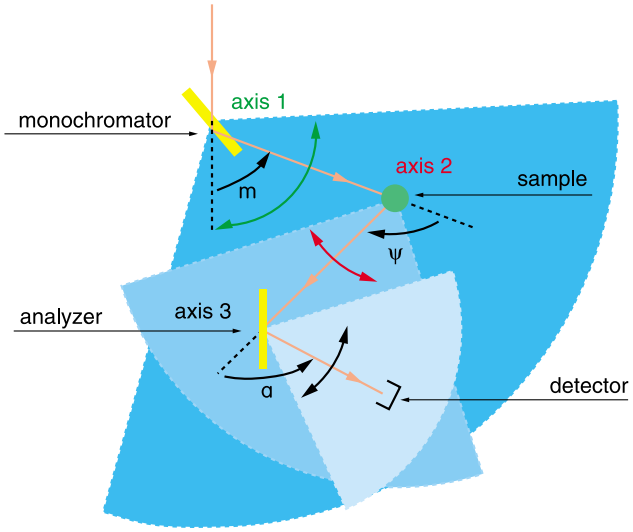
These two points of view are the basis for the 2 kinds of spectrometers that measure inelastic scattering (i.e. «triple axis» and «time of flight» types)

❑ Triple-axis spectrometer

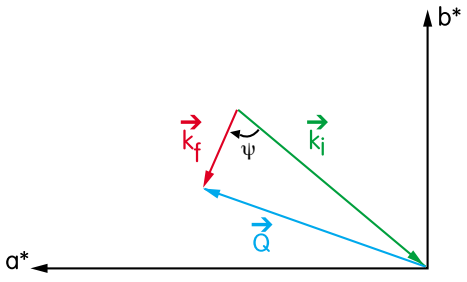
Before the sample, neutrons with a specific energy are selected; then, after scattering their energy is measured by their wavelength.

The parallel and monochromatic ($\lambda\omega$) incident beam is obtained by Bragg reflection on a single crystal (monochromator). It is possible to vary $\lambda\omega$ by rotating the entire spectrometer (angle m) around the vertical axis passing through the monochromator (1st axis).

The analysis of the scattered intensity is performed as a function of the angle ψ measured around the vertical axis passing through the sample (2nd axis). Finally, in the direction ψ the energy of the neutrons is analysed by a second single crystal and a mobile detector rotating with angle «a» around the corresponding vertical axis (3rd axis).



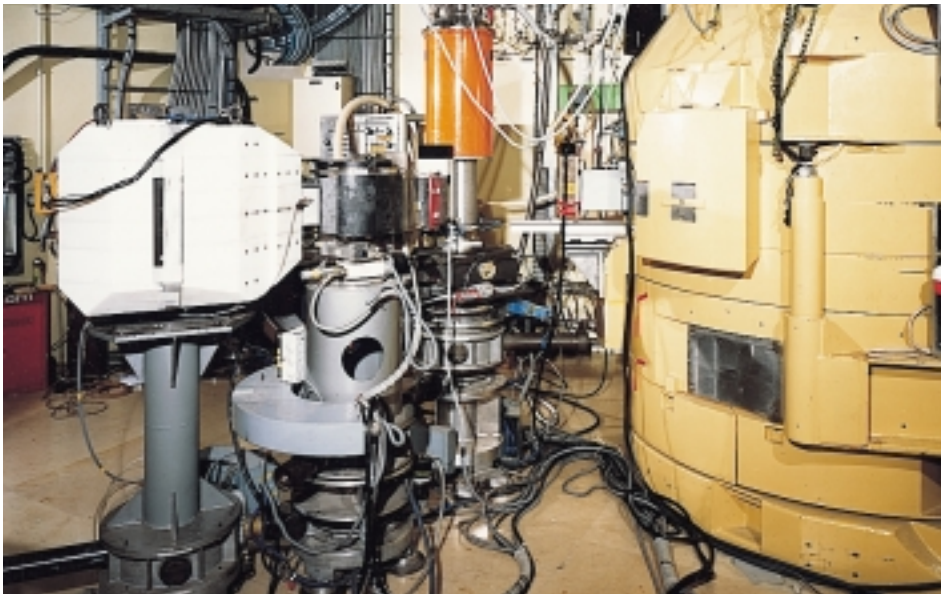
Outline of the principle of a 3-axis spectrometer.



Scattering configuration.

Plane a*, b* is the plane of the reciprocal lattice defined by the scattered and incident beams (scattering plane).

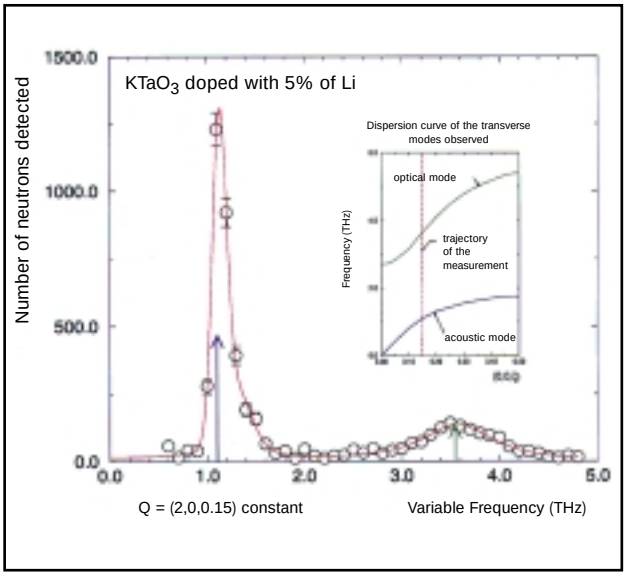
Provided that the sample is a single crystal, the analysis of the scattering diagram shows that the degrees of freedom available allow one to move at will in the chosen plan of the reciprocal lattice.



Triple-axis spectrometer 1T

Depending on the particular case, a path can be followed keeping $\vec{k}_i - \vec{k}_f = c^{te}$ [measure of $S(\vec{Q} = c^{te}, \omega)$], or $E_i - E_f = c^{te}$ [$S(\vec{Q}, \omega = c^{te})$]. These possibilities are extremely advantageous when using a single crystal of the material under study.

Constant $\vec{Q}$ method: observation of an acoustic mode and of an optical mode.

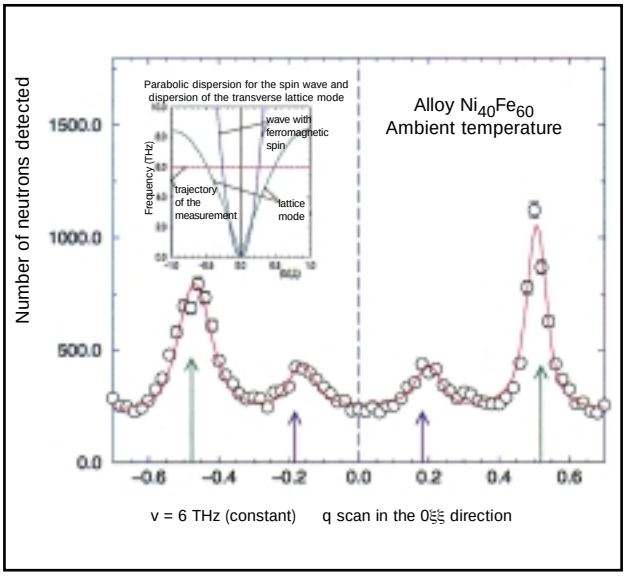


By probing the reciprocal space of the crystal lattice, the 3-axis spectrometer is able to measure the energy of the modes of vibration and their dispersion curves.

Neutrons are scattered when the measurement trajectory intersects the curves $\omega = f(\vec{Q})$ which are characteristics of the lattice dynamics of the crystalline system. The figure gives an example of a measurement at constant $\vec{Q}$.

It shows 2 modes (1 acoustic and 1 optic) with transverse polarization.

Constant energy method: observation of a lattice vibration mode and of a spin wave mode.



When the vibration modes exhibit a strong dispersion in $\vec{Q}$, the 3-axis spectrometer allows to follow a trajectory with constant energy (or frequency).

The figure gives an example of the simultaneous determination of a lattice vibration mode (green arrow) and of a spin wave mode (blue arrow) in a ferromagnetic alloy $Ni_{40}Fe_{60}$.

The $+\vec{Q}$ and $-\vec{Q}$ scans reveal the effect of the instrumental resolution.

❑ The time-of-flight spectrometer

The speed V, or more precisely the time t used to travel along a given distance $\left(t = \frac{L}{V}\right)$, measures the neutron energy before and after scattering by the sample.

The monoenergetic incident beam (V_i) is formed in bursts or pulses by a chopper monochromator. The moment when the burst passes through the sample is known thanks to the production of an electric impulse synchronised with the opening of the last chopper. By activating at this very moment a «chronometer», one can measure the time each neutron takes to «fly» across from the sample to the detector situated at a distance L in the direction ψ. Obviously, the measurement is repeated identically for each burst produced by the monochromator (for each turn of the chopper wheel).

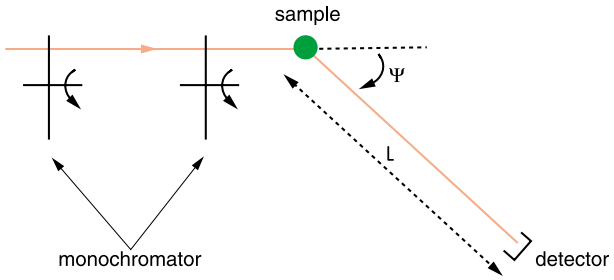
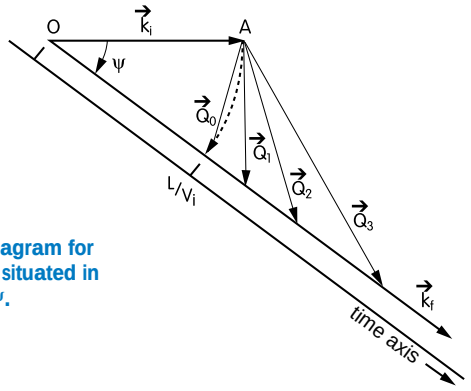
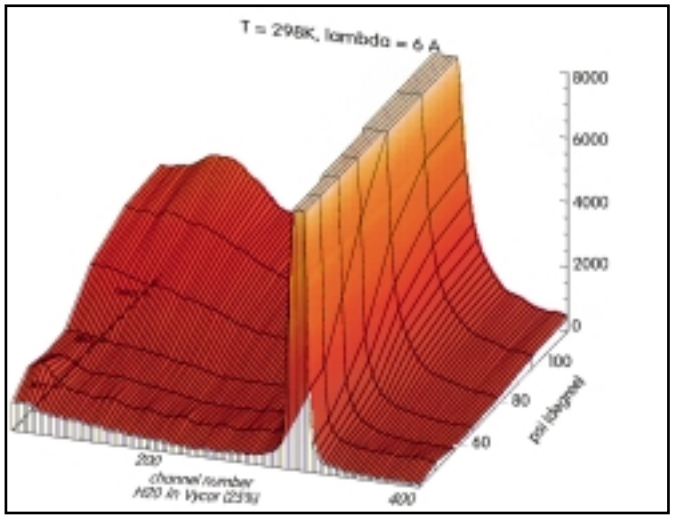


Diagram of a time-of-flight spectrometer.

The scattering diagram shows that each arrival time in the detector corresponds to a transfer of momentum Q which changes in module and direction. The path in the reciprocal space is complicated so that the Q dependence of the scattering function may be difficult to study accurately. If the sample is isotropic (powder or liquid) one shall measure the $S(|\vec{Q}_o|_\psi, \omega)$ function in the direction (ψ). Then by setting up numerous detectors, the inelastic transfers for several $|Q_o|$ may be simultaneously measured.



Scattering diagram for the detector situated in a direction Ψ.



Time-of-flight spectrum of confined water.

Vycor is a silicate glass with high porosity corresponding to a network of interconnected cylindrical pores (diameter 50 Å). In these internal volumes, numerous molecules can be adsorbed; there, they are confined in a very small space. This confinement influences their dynamics and, therefore their thermodynamic properties. The spectrum of neutrons scattered by water molecules adsorbed in vycor is measured by time-of-flight method as a function of the scattering angle and of the energy exchanged. The analysis of the quasi-elastic components indicates that the model of diffusion by jumps which is applicable to liquid water remains valid. By contrast, the long range diffusion coefficient at 25°C is about 10 times weaker than for non-confined water, but it remains measurable at -15 °C.

❑ Measuring very low energy transfers: the «spin-echo» spectrometer

The magnetic moment (spin) of a neutron provides the opportunity to precisely measure very weak energy changes that may take place during the scattering process. Immersed into a magnetic field H, the neutron spin rotates around the field (Larmor precession) with an angular speed $\Omega = \gamma H$ (γ = gyromagnetic ratio $\simeq 2.10^8$ rd/s.T). Used as a chronometer, this rotation is interesting for two reasons:

- By counting the number of turns, we know precisely the amount of time the neutron spends in the magnetic field. If the neutron travels along the axis of a solenoid with length L (precession coil) traversed by a current (that creates the field), one can deduce its speed V.
- Each neutron carries its own «clock» that is activated when it penetrates the coils and is deactivated when it comes out.

The "Spin-echo" spectrometer, MESS.

The long cylinders that are found before and after the sample are precession coils. They are identical but create opposite magnetic fields. If during scattering, the neutron does not change speed, its spin undergoes N_1 turns before the sample and $-N_1$ after; on exiting it will have the same orientation as it did on entering (echo). On the other hand, if the speed changes, the orientation of the spin will be different. By measuring this orientation we can detect a change of speed ($\Delta V / V_o \simeq 10^{-5}$).

3 - The reflectometers

In the preceding chapter we mentioned that every medium is characterised, from the point of view of wave propagation, by an index **n** (whose value depends on the nature of the wave). In the case of neutron propagation, we can show that this index depends only on **the scattering length density** of the material, which is the product of its density (ϱ) by the mean scattering length (b_{coh}) of the different atomic species present.

$$n = 1 - \frac{\lambda^2}{2\pi}\varrho\ b_{coh}$$

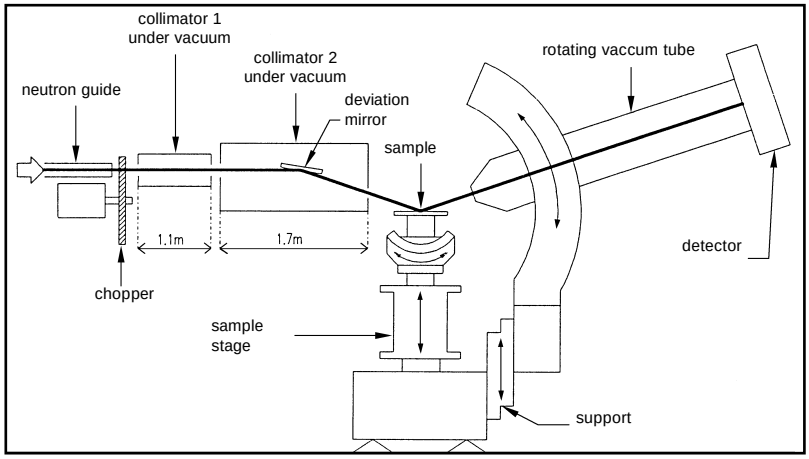


Diagram of the time-of-flight reflectometer DESIR (G5 bis).

Let us note immediately that, even in a material that is dense and made of atoms having a large scattering length, the index **n** always remains in the neighbourhood of 1 for the range of available wavelengths; in nickel for example:

$$\lambda = 10\text{Å} \Rightarrow \frac{\lambda^2}{2\pi}\varrho\ b_{coh} \approx 10^{-4}.$$

The laws describing the passage of the neutron wave from a medium with index $\mathbf{n}_1$ ($\mathbf{n}_1$=1 for air or vacuum) to the sample medium with index $\mathbf{n}_2$ are the same as for light. They are known since the 19th century (Fresnel, Maxwell). In particular, there is always the appearance of a **reflected beam**.

If $\mathbf{n}_2<1$, there exists a critical angle θ_c such that, for $\theta<\theta_c$ the beam undergoes a **total reflection**:

$$\theta_c \sim \frac{\lambda}{\sqrt{\pi}}\sqrt{\varrho_2 b_2}$$

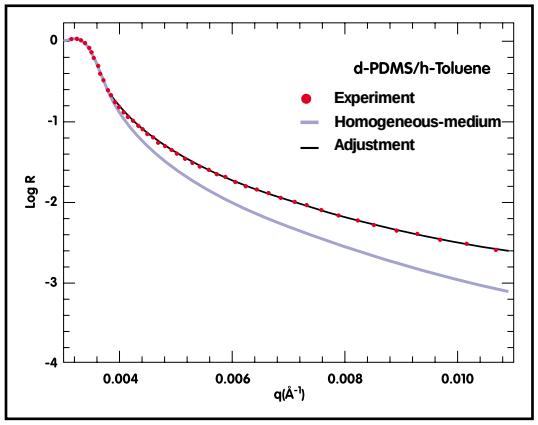
For $\theta>\theta_c$ the beam splits into a refracted and a reflected beam. The distribution of the intensity into these 2 components depends on the incident angle θ and on the variation of the index $\mathbf{n}_2$ (z) with the distance (z) to the interface.

- If the medium is homogeneous ($\mathbf{n}_2$ (z) = constant), the reflected intensity is given by Fresnel's formula:

$$I_r = I_o \left|\frac{1 - x}{1 + x}\right|^2 ; \; x = \sqrt{1 - \left(\frac{q_c}{q}\right)^2} \; ; \; q = \frac{4\pi}{\lambda} \sin\theta$$

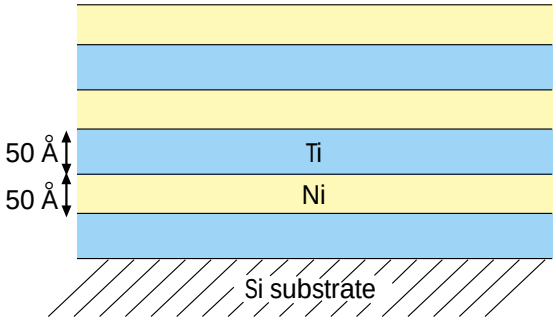
- If $\frac{d\,\mathbf{n}_2}{dz} \neq 0$ (index gradient), a deviation from this distribution will be measured. Choosing a particular form for $\mathbf{n}_2$(z), different theoretical curves can be calculated and compared with the experimental results.

Reflected intensity at the interface between air and a solution of PDMS in toluene.



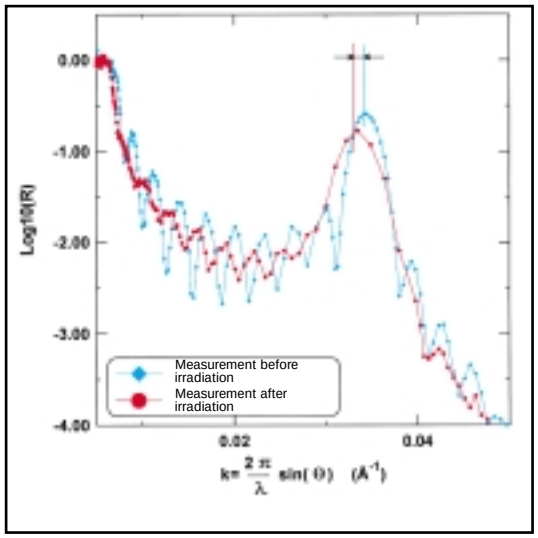
The deviation from the curve «homogeneous medium» is due to the concentration gradient of polymer molecules near the surface. The theoretical prediction Ø (z) = $z^{-4/3}$ is in good agreement with the experimental result (Ø (z) = polymer concentration).

- For a layered structure, the index undergoes abrupt changes (steps of a staircase) evidenced by a series of peaks in the decreasing part of the reflected intensity. These peaks come from interferences between the different rays reflected by successive interfaces. The number of peaks and their angular distances allow the scientist to measure the thickness of the layers and the quality of the interfaces (roughness, inter-diffusion,...).



Multilayer of 19 double layers Ni/Ti on silicon.

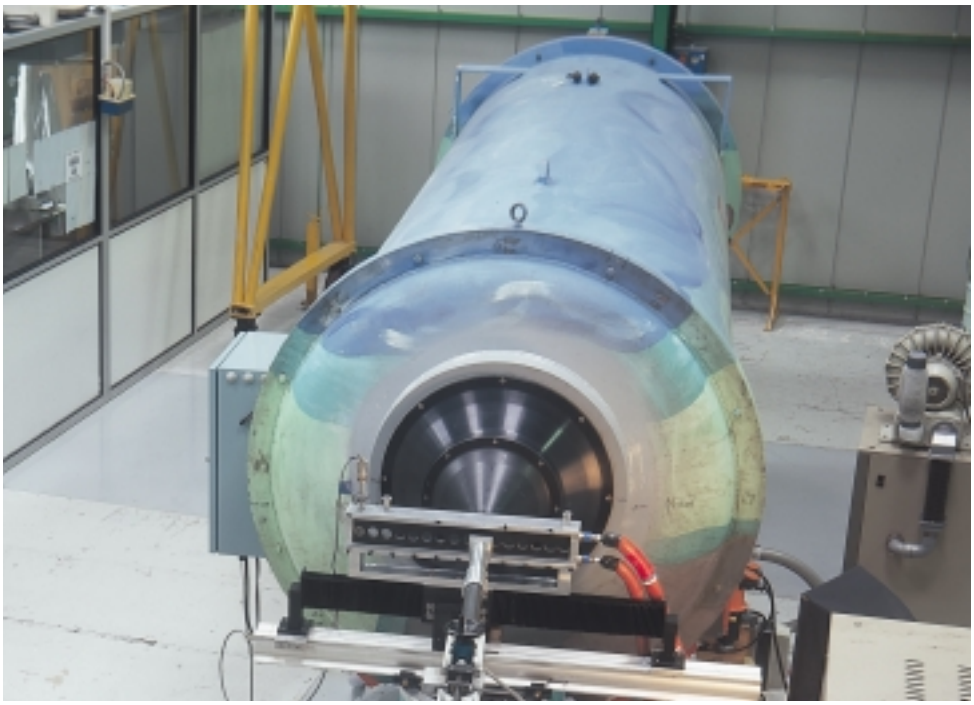
Treatment: irradiation with thermal neutrons under a dose of 6.10^{19}n.cm^{-2}.



Reflectivity curve of the multilayer measured before and after irradiation.

The shift of the peak is typical of a swelling of the layers. The lowering of its intensity is characteristic of the increase (from 5 to 30 Å) in the roughness between the layers.

4 - Spectrometers for «small angle scattering» (SANS)



The small-angle spectrometer PAXE.

Built to characterise large size objects (1 to 50 nm), these spectrometers measure the quantity of neutrons scattered «near the forward direction», that is to say resulting from a process characterised by very small transfers of momentum.

These spectrometers must satisfy 2 conditions: a very well collimated incident beam with a long wavelength λ_0; a wide planar detector located far from the sample (between 1 and 7 m).

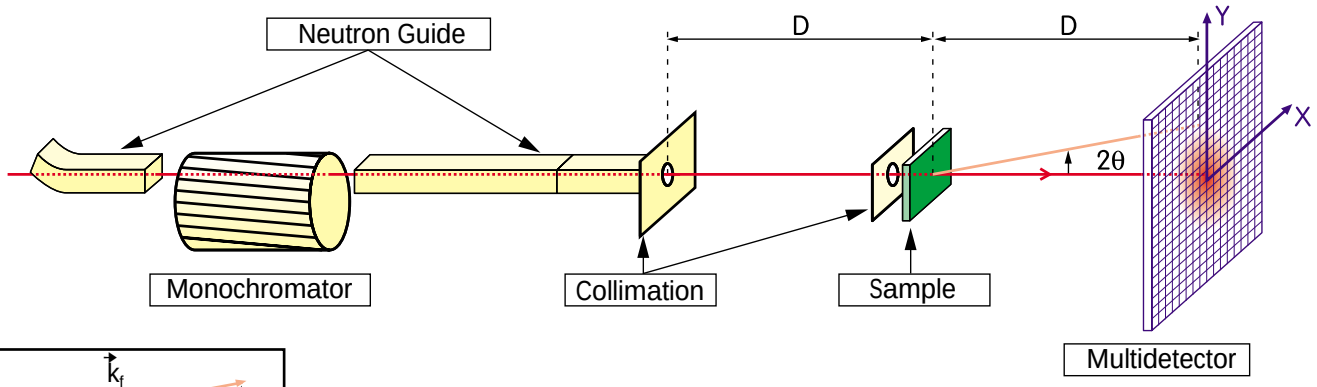
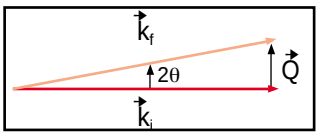


Diagram of a small angle scattering spectrometer.



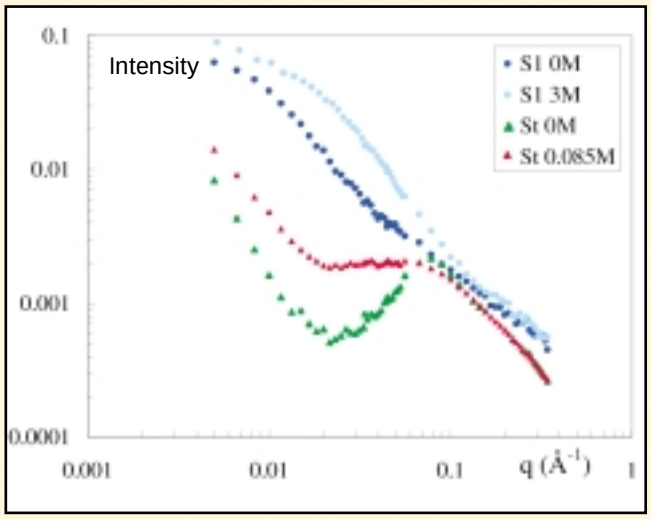
Scattering diagram.

The impact point of each detected neutron is located by its coordinates (X, Y) in a system of orthogonal axes linked to the detector. The origin of the axes is chosen at the impact point of the direct beam (not deviated by the sample). Therefore, a neutron detected at the point with coordinates (X, Y) has undergone a momentum transfer:

$$Q = \frac{4\pi}{\lambda_0} \sin\theta \sim \frac{2\pi}{\lambda_0} \frac{\sqrt{X^2 + Y^2}}{D} \quad (\theta \text{ small})$$

where D is the distance from the sample to the detector.

Inter and intra-chain electrostatic interaction in a poly-electrolyte solution



Intensity scattered by a solution of poly-electrolyte, with and without added salt.

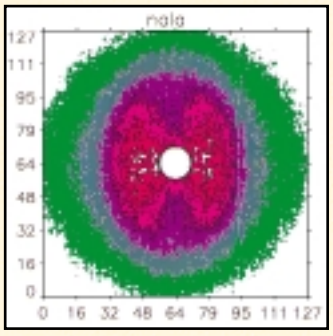
Intensity measured in the direction 2θ (abscissa: $q = \frac{4\pi}{\lambda}\sin\theta$) for poly-electrolyte solutions (charged polymers in water, 0.17 moles/liter):

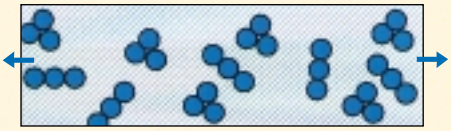
- If all the chains are deuterated, the scattered intensity (green triangles) shows a peak around $q = 0{,}1$ Å^{-1} characteristic of a repulsion between chains. This interaction is «screened» by the strong density of charges when salt is added (red triangles).
- If half the chains are deuterated and the other half hydrogenated, and if the index of the solvent is adjusted to an average value (mixture of heavy water and light water), one measures the signal from a single chain directly. In the absence of salt (dark blue circles), the repulsion stretches the chain and, as a result, its signal decreases more quickly with angle. In the presence of salt (light blue circles), this repulsion disappears and the chain has the same rigidity as in its neutral state.

The stretching effect in a composite material

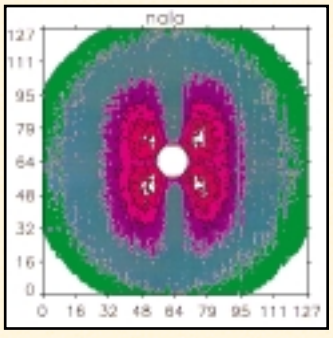
Isotropic

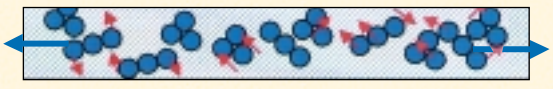
Experimental proof of the correlations between nanometric silica particles in a stretched polymer matrix: Intensity mapping on a bi-dimensional multidetector perpendicular to the beam. The progressive distortion of the originally isotropic signal (ring) evidences the displacement of the particles caused by the stretching; this enables scientists to understand the reinforcement of polymers by hard particles.





Moderate stretch:
displacement of the hard parts
(Opposite spectrum, 2 lobes)



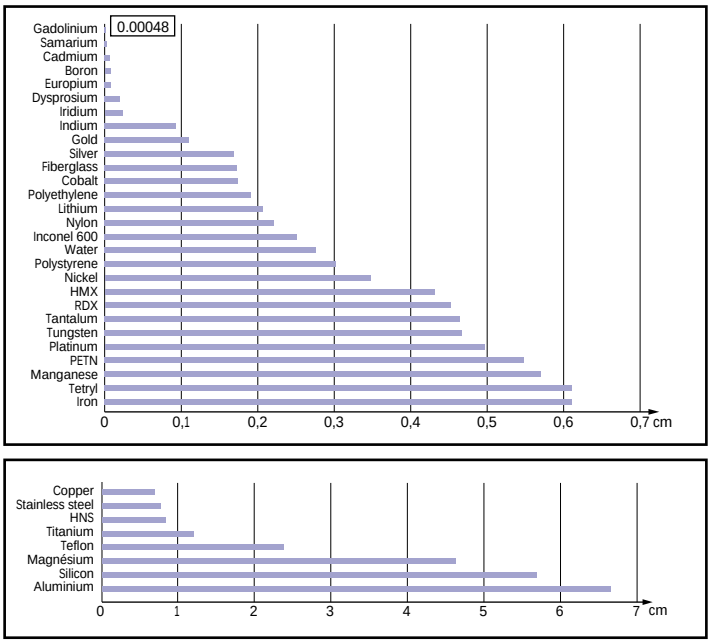


Avoidance by local shearing
(Opposite spectrum, 4 lobes)

Observation by SANS of the relative displacement of hard particles in a stretched polymer matrix.

5 - Neutron radiography

Totally different in its principle and in its goals, neutron radiography is a direct imaging technique.
A wide neutron beam is sent to the object to be observed. A photographic detector, which is sensitive to neutrons, is placed behind it. After exposure, an image of the «neutron transparency» averaged on the thickness of the object is obtained.

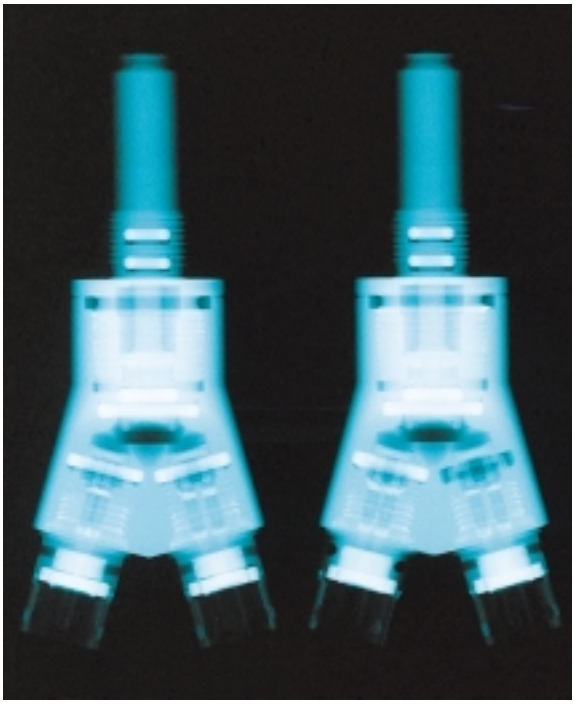


This transparency is a function of the probability that the neutron passes through the object without «disappearing» from the original beam, in other words without being either absorbed (in this case it would disappear completely) or scattered (then it would have been deviated from its initial trajectory and consequently it would not reach the photo plate). The result of a neutron radiography is related to the interaction of the neutron with the atoms (see P. 8-10) and thus is very complementary to the X-ray radiography.

Thickness of various materials which gives a beam attenuation of 50% (λ = 1.8 Å).

Space technology requires as much control as possible of a lot of critical equipment. This includes devices that make use of pyrotechnology (explosives) employed to ensure essential functions like the separation into stages and the release of satellites. The great sensitivity of neutron to hydrogen makes neutron radiography a trustworthy way to detect, across metallic envelopes, eventual defects in the placement of these explosives or in the final assembly.

In the photograph we see that a rubber joint (in black) is missing in the element to the right.



Neutron radiography of some pyrotechnic jacks used in the rocket «ARIANE» (DASSAULT-AVIATION).

Working at the LLB

Our goal

One important mission of the LLB is to encourage the use of neutron spectrometry in the various areas of fundamental and applied research. For this purpose, it is important to keep a high quality research program within the laboratory itself; this in turn requires the ability to receive numerous doctoral and post-doctoral students. Laboratory visits, either for individuals or for groups, are organised on request. Such visits allow one to discover the installation itself, and to develop contacts with the different research teams working on various fields.
The LLB gives, to external teams of scientists, access to its experimental facilities on the basis of a written proposal and scientific selection, in the following cases :
- scientists of french laboratories,
- scientists of european union or associated states, in the frame of E.C. funded access contracts,
- scientists of foreign laboratories preferably having a contractual agreement.

Access for industrials can be directly negociated with the Direction of LLB.

Submission of a proposal

Please contact the Scientific Secretariat of the LLB the first time you would like to submit a proposal; they will send you the appropriate application forms. Submission through the web is also possible. Each year (usually, at the end of november) we plan 4 thematics "Round Table" discussions where external users can meet and discuss with all LLB's members. Their announcement is made during summer.

A brochure giving the title and dates of each Round Table, and including a participation bulletin, can be sent on simple request to the scientific secretary.

A selection committee is associated with each Round Table; it is composed of scientist representatives from the French and European communities, as well as members of the LLB. The selection is made twice a year (deadline for proposal submission: 1st of april, 1st of october); each proposal is rated:

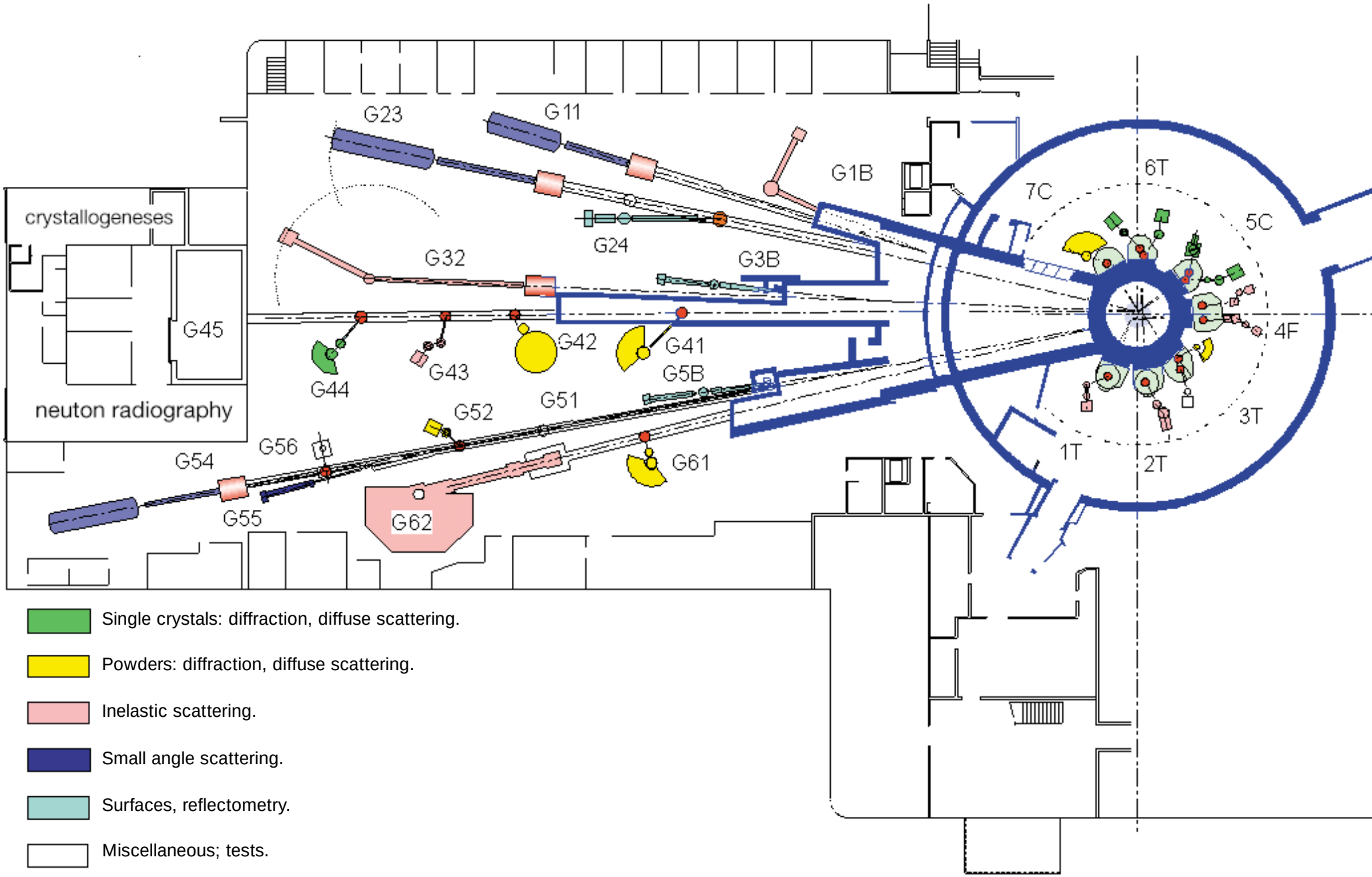
- A The experiment has been accepted on the basis of its scientific merits and will be scheduled.
- B The experiment has been accepted but may be completed only if sufficient beam time is available.
- C The experiment has not been accepted.

For additional information, please contact:

THE SCIENTIFIC SECRETARIAT
LEON BRILLOUIN LABORATORY
CEA SACLAY
Building 563
F – 91191 GIF-SUR-YVETTE CEDEX
FRANCE

✆ 33 (0) 1 69 08 60 38
FAX : 33 (0) 1 69 08 82 61
e-mail : experience@llb.saclay.cea.fr
web site: http://www-llb.cea.fr

GENERAL LAYOUT OF SPECTROMETERS



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Page 45	Neutron radiography of pyrotechnical jacks in Ariane	CEA/Dassault-Aviation
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Back cover	Portrait of Léon Brillouin	Academy of Science / Jean-Loup Charmet Paris

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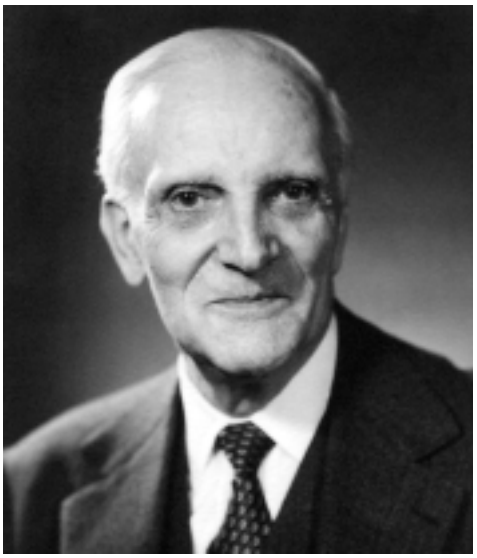
Léon Brillouin
1889-1969



Léon Brillouin, born in Sèvres in 1889, was admitted to the Ecole Normale Supérieure in 1908. In contrast with most young French physicists of the time, Brillouin continued his education (1912) at the Institute of Theoretical Physics in Munich, which was under the direction of A. Sommerfeld. The Von Laue experiment on «the diffraction of Roentgen rays» (X-rays) by a crystal took place there, several months before his arrival. On his return to France (1913), he began a thesis on the theory of solids; he proposed an equation of state based on the atomic vibrations (phonons) that propagate through a solid. He also studied the propagation of monochromatic light waves and their interaction with acoustic waves; he showed that the scattered wave is made up of the sum of three components (Brillouin effect): one at the frequency of the incident wave (ω_o), the two others at frequencies located relative to it ($\omega_o \pm \Delta\omega$) (Brillouin doublet); their separation is dependent on the scattering angle. It wasn't until ten years later that this theoretical prediction was observed experimentally. His work came to a halt during the First World War (1914-1918); in 1920 Brillouin defended his thesis (Jury: Marie Curie, Paul Langevin, Jean Perrin!).

Thus began for Léon Brillouin a period of great scientific production with major contributions in the «quantum revolution» in several areas of physics:

- he proposed an approximate resolution method of Schrödinger's equation (B.K.W. method: **B**rillouin, **K**ramers, **W**entzel), applied to electrons;
- he modified the theory of paramagnetism (Langevin had provided a «classical» model twenty years earlier) by introducing the quantification of the orbital moment (the Brillouin function, 1927);.
- during the course of his work on the propagation of electron waves in a crystal lattice, he decided to introduce a concept that would be found particularly useful in the theory of crystalline solids: the Brillouin zones (1930);
- he published a series of articles in which he discussed methods for the study of systems with several electrons (Brillouin-Wigner formula).

Along with his research activities, Brillouin also taught. His first position was at the Sorbonne where he was offered the Theoretical Physics Chair in 1928; he then taught at the College de France to which he was elected in 1932.

In August 1939, a month before the declaration of war against Germany, Léon Brillouin, as a specialist of wave propagation, was named director of the French National Radio-diffusion. In May 1940 came the collapse of France; the government and the high administration of which he was a member, retired to Vichy. He remained there for six months before resigning and leaving for the United States where he joined the « France libre » organization. There he participated in the war effort by working in the field of radar at Columbia University in New York. At the end of the war, he decided to stay in the United States where he taught at Harvard and at Columbia; Brillouin was elected to the U.S National Academy of Sciences in 1953. Far from abandoning research, he became involved in a new field: «The Theory of Information». He invented the concept of «Neguentropy» (negative entropy) to demonstrate the similarity between entropy and information, and accordingly, that «Maxwell's Demon» does not violate Carnot's principle. He died in New York in 1969.

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