APPLICATION OF SPALLATION NEUTRON SOURCES

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Abstract

The first part is devoted to an introduction into the physical principles of spallation neutron sources. We emphasize the difference in the optimized lay-out of target-moderator assembly between continuous and pulsed sources. A short intermezzo on neutron optics is followed up by a presentation of the experimental principles and their scientific goals at these sources. Finally I would like to make a few remarks on the complementarity between neutron- and synchrotron light-scattering.

1 INTRODUCTION

The idea of accelerator driven neutron sources goes back to the fifties of last century. The project of a Materials Testing Accelerator in Livermore aimed at the production of fissionable materials. With the success of nuclear reactors for energy production and fuel breeding these projects lost significance. Beam tube reactors became the established neutron sources for scientific activities. Due to technical reasons this research reactors later on turned out to be limited to a flux of thermal neutrons somewhat above 10^{15} n/cm^{2}s, e.g. ILL (Grenoble). This led to a revival of accelerator driven neutron sources for research purposes in the eighties. A more economic use of the neutrons could be expected by producing only those, which can be finally used at the instruments by exploiting the time of flight technique at a pulsed source. Since pulsed reactors normally deliver pulses, which are not short enough, synchrotron driven spallation sources entered the scene. Since the peak flux plays a crucial role in time of flight (TOF) experiments, high data rate can be obtained at a relatively low thermal load of the target system.

In the nineties accelerator driven energy systems were reconsidered again, but now with emphasis of transmutation of actinides in the context of waste management from used nuclear fuels. While the pulse structure is a basic mean to economize the neutronics of a neutron source, it is quite obvious that for such highest power "energy" – applications a d.c. beam is preferable. In view of this development a continuous spallation neutron source has been built and attached to the PSI cyclotron system, which delivers a 600 MeV continuous proton beam with a power beyond 1 MW. During four years of operational experience of this source (SINQ) it has been demonstrated that

- SINQ is a research neutron source of the upper medium flux type. It is an alternative and equivalent to a beam tube reactor for thermal and cold neutrons.
- It can be used as a pilot station for target development and materials study for Accelerator Driven Systems (ADS). The project MEGAPIE is a joint initiative by six European research institutions and JAERI (Japan) to design, build, operate and explore a liquid lead-bismuth spallation target for 1 MW of beam power, taking advantage of SINQ.

In this presentation we emphasize the differences in the design and the applications of pulsed and continuous spallation sources. We do not address particularly applications of AD-energy systems. As reference for our discussion I have chosen the target stations ISIS (RAL) and SINQ (PSI). The accelerator driven sources nowadays in operation are listed in table 1.

Table 1:

	Accelerator	Energy [MeV]	Rep. Rate [Hz]	p-Current [μA]	Target Material
IPNS (Argonne)	Synchrotron	600	45	20	U
KENS (KEK)	Synchrotron	500	20	10	(U^{238}), W
ISIS (RAL)	Synchrotron	800	53	200	(U^{238}),, Ta
MLNSC (LANL)	Linac + PSR	800 800	120 20	100	W
SINQ (PSI)	Isochr. Cyclotron	600	-	1200	Pb

2 PRINCIPLES OF SPALLATION NEUTRON SOURCES

2.1 Proton Beam

The proton beam is extracted from an accelerator system and transported to the neutron production target.

For a pulsed source a synchrotron or a Linac followed by a synchrotron type of storage ring can give the suitable time structure, that is pulses with a width of less than 1 μs and a repetition rate of typically 20 – 50 Hz.

A cyclotron delivers a quasi-continuous beam. Its micro-time structure within nanoseconds becomes irrelevant for thermal neutrons. For both types of neutron

sources an energy of the primary proton beam between 500-1200 MeV (and possibly even somewhat higher) is adequate.

2.2 Neutron Production Target

Fast neutrons are produced from the nuclear interactions between the incident protons and the target nuclei. With this so-called spallation reaction about 10-20 neutrons are liberated by one proton depending on the incident energy and the heavy metal target materials. These are summarized in table 1 for a set of accelerator driven neutron sources nowadays in operation.

Note, that the very efficient depleted uranium targets have by now been replaced by targets made of non-fissionable materials, which are neutronically less efficient. This is due to the notorically short lifetime of an uranium assembly in this hostile environment.

2.3 Moderator / Reflector Systems

The low energy part of the neutron spectrum (E<15 MeV) consisting of about 90% of all the neutrons emitted from the target have an isotropic angular distribution and an average energy of $\overline{E} \cong 2\,MeV$ [1]. These fast neutrons are reflected into the moderators, where they are slowed down by collision processes to epithermal and thermal energies. This diffusive slowing down process determines the pulses shape and pulse length of a pulsed neutron source. The pulse properties depend therefore crucially on the size and the design of this assembly. While the pulse shape is a primary resolution element for a pulsed source it is not relevant for a continuous source. The moderator / reflector-system of either source looks completely different. Examples of the layout for the continuous source SINQ (PSI) [2] and the pulsed source ISIS (RAL) [3] are given in Figures 1 and 2.
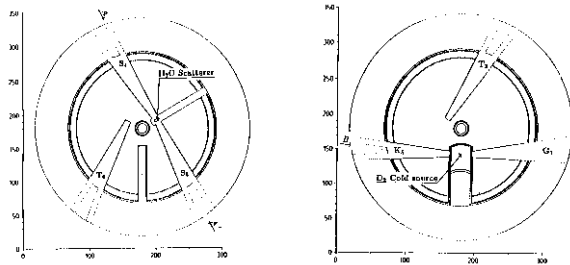

Figure 1: Cuts on two different vertical levels through target and moderator tank of SINQ viewing cold source insert and extraction channels [2].

In SINQ the target is by means of a double walled moderator tank concentrically surrounded by 6 m³ D₂O as moderator and by a 5 cm layer of H₂O. This assembly, including the tangential extraction channels indicated, is a welded aluminium structure. Note that this system looks rather similar to a beam tube reactor – the difference being, the spallation target replacing a reactor core. The beam injection onto the target is in this case from below. Obviously this system is optimised for maximal average flux of thermal neutrons [4] (Fig. 3). The assembly of ISIS as a pulsed source is more complex. Small hydrogen

rich moderator blocks are located adjacent to the target and surrounded by beryllium blocks as reflectors. In order to cut off long tails in the neutron pulses decoupler layers with relatively large absorption cross-section for thermal neutrons have been introduced between reflector- and moderator materials as well as at the inner surfaces of the extraction channels. Figures 4 and 5 show the neutron spectra and the spectral pulse shape from this assembly [5].
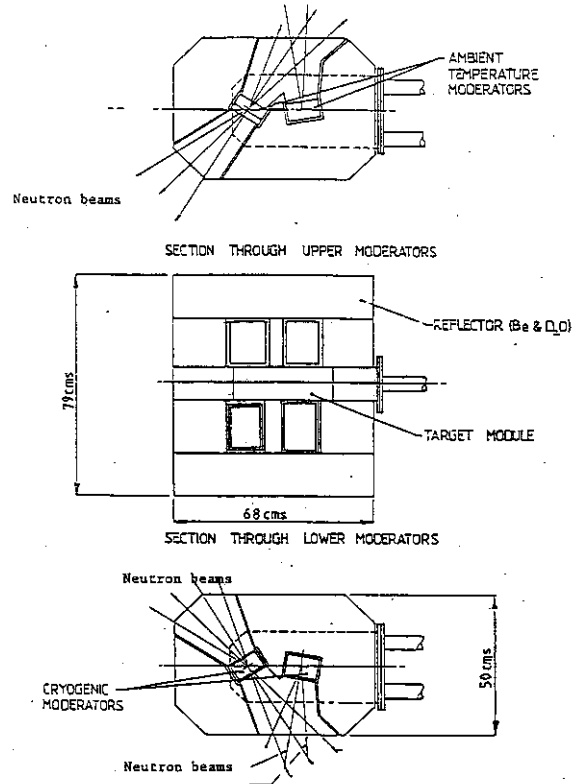

Figure 2: Geometric arrangement of the target moderator/reflector assembly of ISIS.

Note that for both types of sources part of the moderator is cryogenically cooled for the production of so called "cold neutrons", whereas the ambient temperature moderator delivers the thermal neutrons.
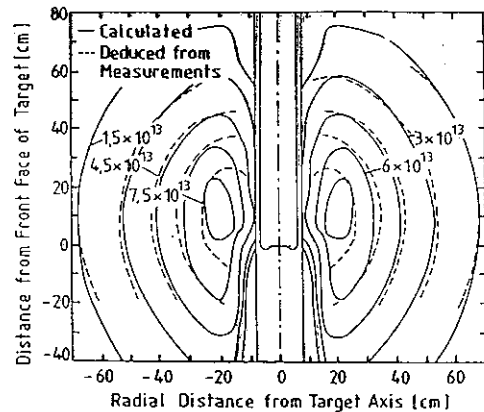

Figure 3: Vertical cut through target moderator assembly and thermal flux distribution measured and calculated (Mock up experiment).

2.4 Bulk Shield

A massive bulk shield around this internal assembly is needed in order to shield the vicinity of the target station from the high-energy neutrons which are emitted as direct knock-out at the very first instant of the spallation reaction. These neutrons – about one in ten – have a spectrum which in principle may reach up to the energy of the primary proton beam. Although not very abundant and emitted preferentially in forward direction, these neutrons determine the massiveness of the bulk shield. The weight of e.g. the SINQ bulk shield amounts to about 12000 t – mostly iron.
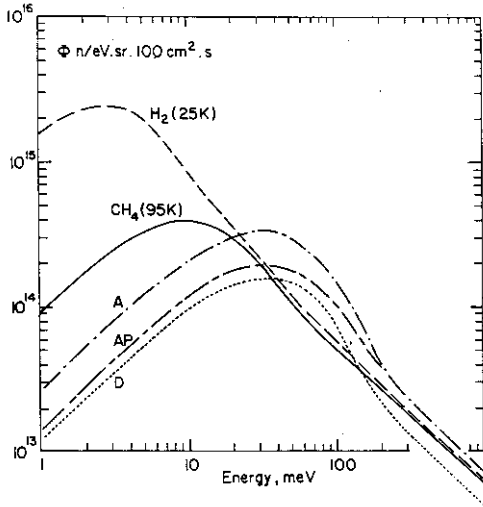
Figure 4: Spectral distribution of neutrons leaving the exit face of various moderators at ISIS (see also Fig. 2).
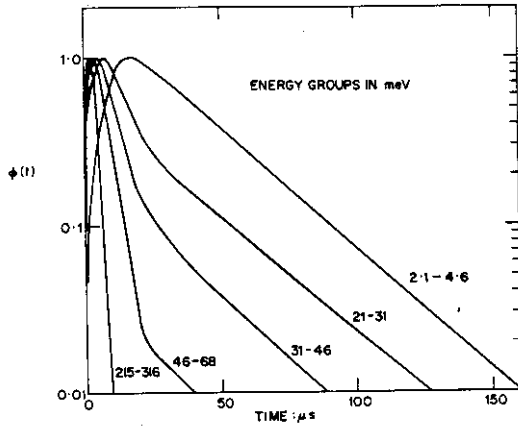
Figure 5: Spectral pulse shape for the liquid methane moderator a ISIS.

2.4 Neutron-Extraction and -Transport

As is indicated in Figures 1 and 2 the neutrons are extracted through evacuated beam tubes viewing the moderator materials. Given the tubes cross section F and its length L through the bulk shield, the neutron flux (e.g. at monochromators) is

$$\sim \frac{\phi}{4\pi} \cdot \frac{F}{L^2} \qquad (1)$$

where ϕ is the thermal flux at the entrance window. Typical numbers are
$\phi = 10^{14}$ n/cm^{2}s, F = 120x60 mm^2, L = 4-6 m
leading to an external flux of few times 10^{10} n/cm^{2}s within a divergence of 1-2°.

A more efficient way of extracting and transporting neutrons is provided by neutron guides – in particular for cold neutrons. Caused by the interaction of a neutron wave field with matter the neutrons show refractive and diffractive behaviour. With <b> as mean nuclear scattering length and N as number density of a surface material we obtain the refractive index:

$$n^2 = 1 - \lambda^2 \frac{N<b>}{\pi} \qquad (2)$$

where λ is the de Brogle wavelength of the neutron. Snell's and Fresnel's law then lead to a condition for total reflection

$$\varphi < \varphi_c = [2(1-n)]^{1/2} . \qquad (3)$$

The acceptance angle for total reflection in a guide is then

$$\delta = 2k \cdot \lambda \qquad (4)$$

where k is a material constant. For a sputtered Ni-surface k = 1.73 mrad/Å. Comparing (4) with the acceptance angle of a beam tube (1), we recognize that for neutrons with wave length $\lambda > 4$ Å the guide becomes superior to a beam tube.
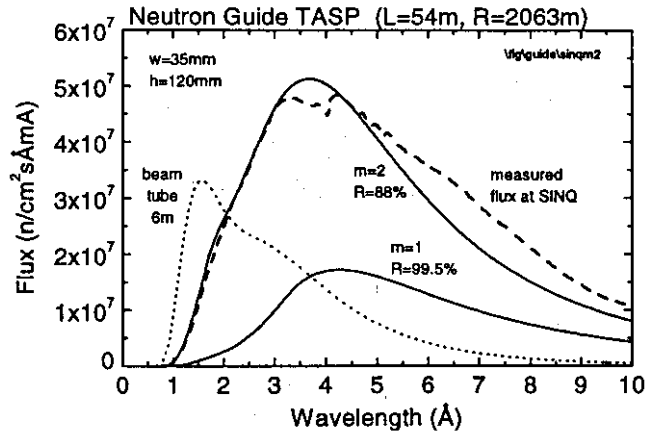
Figure 6: Spectral flux of neutrons at the exit of a super mirror guide viewing the cold source of SINQ.

The performance concerning the accepted divergence of a neutron beam can be improved by using diffractive effects on a mesoscopic 2d-layered coating on a substrate [6]. It consists of a few hundred alternate layers with varying thickness of two materials whose scattering lengths for neutrons differ as much as possible – e.g. Ni, Ti. In such a system – called super mirror – the angle of transport in a guide can be increased by a factor of two to four with respect to natural nickel. Fig. 6 demonstrates this performance [7]. Note that this super mirror becomes already superior to a beam tube for wave lengths $\lambda > 2$ Å. If we take into consideration, not only the nuclear – but also the magnetic interaction of the neutron by its

magnetic moment with the material the refractive index becomes

$$n = 1 - \lambda^2 \left[\frac{Nb}{2\pi} \pm \frac{m}{4\pi\hbar^2} \mu B \right] \qquad (5)$$

where μ is the magnetic moment of the neutron and B the magnetic field in the material. The two signs apply to the two spin directions of the neutron. This offers the possibility to choose the materials of the bilayers such that n becomes uniform through the whole stack for one spin but not for the other. By means of this construct the super mirror effect applies to one spin direction only.

3 SCIENCE WITH NEUTRONS

3.1 Why neutrons?

The properties of the neutrons, which contribute to their importance as probe for the investigation of condensed matter are listed in table 2. In view of the comparison with other probes some remarks are in order:

Table 2: The Neutron as a Probe in Condensed Matter Science

• No charge, small cross section	can investigate bulk material can use thick sample containers can treat scattering in first Born approximation
• Scattering by nuclei	can "see" hydrogen can distinguish isotopes ⇒ contrast variation
• Magnetic moment	can examine magnetic properties on microscopic scale (⇒ antiferromagnetism)
• Wavelength of thermal neutrons in the range of interatomic distances	can determine crystal structures and atomic positions
• Kinetic energy in the range of elementary excitations	can investigate dynamical properties and excitations energies
• Coherent and incoherent scattering (spin)	can investigate collective phenomena as well as single atom effects (e.g. diffusion)

- The first point is also valid for X-rays, at least in the regime of dominant Thomson scattering as elementary process.
- The second property is unique for neutrons and is responsible for their significance in e.g. biological applications
- Magnetic scattering is of approximately the same strength as the nuclear interaction. Hence neutrons are an excellent probe for magnetic structure and fluctuations. For X-rays this is a very small

relativistic effect. The interpretation of corresponding experiments are less direct.
- This can also be achieved by x-rays – with much more flux even.
- This is one of the most important unique properties of neutrons. To attain similar results with X-rays incredibly high energy resolution is needed.
- This is less important for X-rays. With neutrons it provides a chance for additional information, but for certain experiments it may also be a nuisance (background).

3.2 The principles of scattering experiments with neutrons [8,9]

Basically there are two types of experiments, namely
- **Diffraction**, where the momentum transfer only is determined

and
- **Inelastic scattering**, with simultaneous measurement of momentum- and energy-transfer. This experiment therefore measures the scattering function in both independent variables $S(\vec{\kappa}, \omega)$ [10].

Here is $\vec{\kappa}$ the momentum transfer

$$\vec{\kappa} = \vec{k}_0 - \vec{k}_1$$

and $\hbar\omega$ the energy transfer

$$\hbar\omega = E_0 - E_1 \qquad (6)$$

where indices zero and one denote the incident and exit probing particle.

In a diffraction experiment we measure, apart from the energy or wave length λ_0 of the incident neutron just the scattering angle ϑ. That is, in terms of the scattering function we observe

$$S(\vec{\kappa}) = \int d\omega S(\vec{\kappa}, \omega) \qquad (7)$$

Such an instrument is called a **diffractometer**. It provides information about crystalline structure of the sample. In an inelastic scattering experiment we measure the momentum transfer as well as the energy transfer that is the full scattering function. Knowing the wave length λ_0 (energy) of the incident neutron we have to determine the scattering angle and have to analyse the energy of the scattered neutron.

An instrument for these kind of jobs is called a **spectrometer** since it provides information on the dynamical properties of the sample.

Fig. 7 gives a compilation of the various combination of these options which may serve as diffractometers or spectrometers at either steady or pulsed neutron sources. The column to the left shows the versions using crystal analysers all over. These are the standard instruments at steady sources. Depending on the scientific purpose and the specific demand on energy resolution one or even both crystal analysers may be replaced by TOF-path by means of beam choppers even at a steady source. At pulsed sources however one of the analysers or choppers is superfluous – it is replaced by the pulse sequence of the

neutron source. This allows a rather economic use of the neutrons. Those which are produced can "all" be used, those one cannot use, are not produced.

Finally a remark concerning Fig. 7. At modern instruments, the neutron detectors are, where ever appropriate, position dependent "banana" shaped devices, which may cover an angular range up to 160 °.
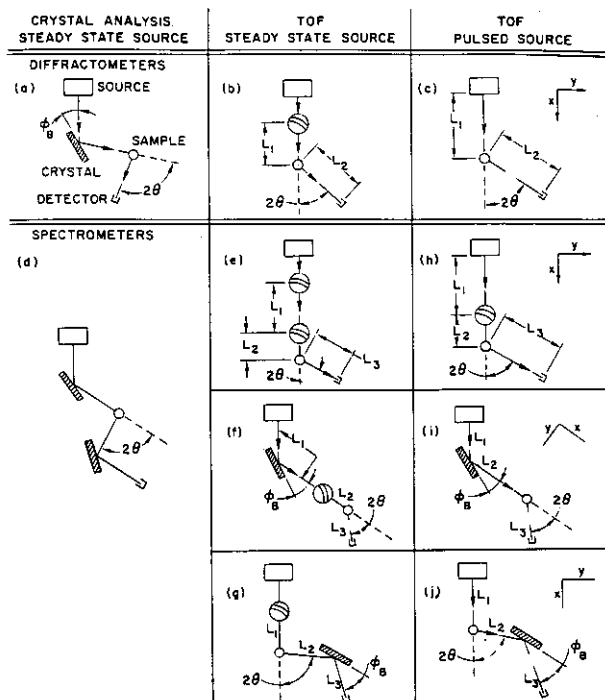

Figure 7: Comparison of diffractometer and spectrometer configurations at steady and pulsed neutron sources showing various combinations of crystal – (and TOF analysis).

Fig. 8 shows the $(\vec{\kappa}, \omega)$-domains, accessible by various experimental methods and probes. The regimes where particular phenomena are abundant are indicated outside the frame.

This representation takes for X-ray scattering only those techniques into consideration, which are kinematically equivalent to an inelastic neutron scattering experiment. Electron spectroscopic methods and resonance scattering are not taken into account. In this sense Fig. 8 does not do full justice to the potential of synchrotron radiation.
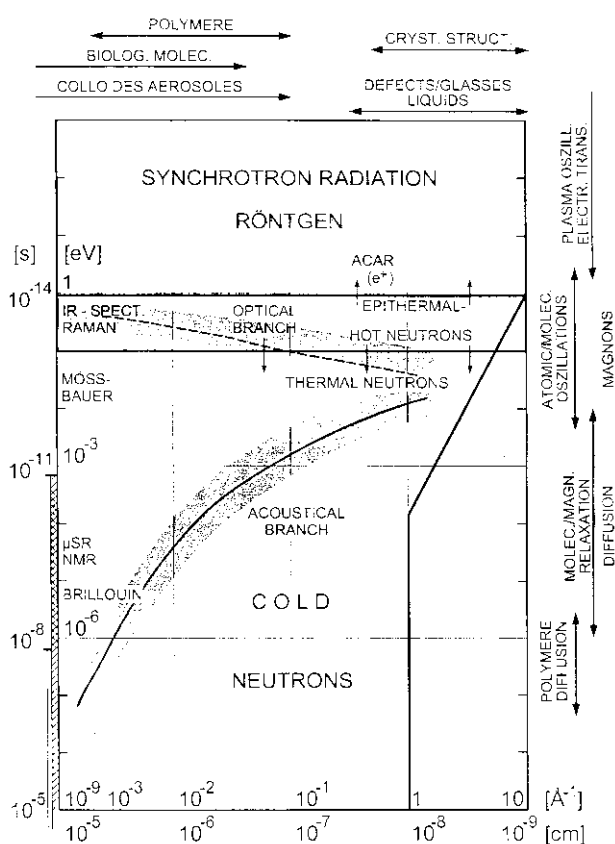

Figure 8: Range of access in energy and momentum by various experimental methods. The corresponding correlation-time and –length is also indicated on the scale. Outside the frame the particular phenomena abundant in that region are recorded. The shadowed strips correspond to the location of a typical phonon dispersion relation.

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