Advanced spallation sources; scientific opportunities and technical feasibility

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1 The Scientific Case for an Advanced Neutron Source

Neutrons, among several other methods, are a scientific tool in the quest to understand the laws governing the behaviour and interaction of many particles in closed or open systems. While the properties of the individual particles in question (ions and electrons) are quite well understood and relatively simple in nature, the net effect of their interaction is the full complexity of matter and its macroscopic properties, which, to a large extent, still evade our understanding.

Traditionally, severals distinctive fields of research used to deal with this complexity and to try to unveil its secrets:

- physics
- chemistry
- molecular biology and
- microbiology.

In a certain sense, although dealing with different degrees of complexity, their common goal is to understand the laws that govern the interaction of many particles in a system and – ultimately – to exploit this knowledge for increasingly sophisticated technologies to produce subtances with new and predetermined properties. In most cases in the past such progress has been made in a heuristic fashion by trial and error but in recent years detailed understanding of the underlying laws has played an ever increasing role in this process. Nevertheless there remain wast numbers of phenomena and effects which are not yet fully understood and which require continuing efforts and scientific scrutinity together with the use of a variety of tools and techniques for their solution. Important examples are:

- the formation and transformation of structures and order in physical, chemical and biological systems
- the interplay of order and chaos
- the dynamics of states far from equilibrium
- and many more.

Understanding of such problems will come about in tiny steps and will require the use of whatever clue is available, an important one being the scattering of radiation from the constituents of the system in question. Since these constituents tend to respond differently to different types of radiation the possibility to apply the various types of radiation depends on the ability to couple to the phenomena in question and on the resolution that can be obtained to extract the desired information. X-rays, or light in general, and neutrons are two typical examples of such complementary tools which, in some cases many furnish equivalent information, in others reveil completely different aspects of a given problem. Despite a rapidly growing range of applications of light scattering as brought about by the advent of new and powerful light sources, there remain a number of cases in which neutrons, due to their very physical properties cannot be substituted (see also: D. Moncton, 1988). The most important ones of these properties are:

- the simplicity of the interaction of the neutron with matter, which leaves little ambiguity in the interpretation of measured data
- the isotope-specific scattering amplitude for neutrons which does not depend in a regular fashion on atomic mass or charge and hence allows distinction of nuclear species which are difficult to distinguish otherwise. This property is frequently used for contrast variation by isotopic substitution.
- the weak interaction of neutrons with matter, which not only allows investigation of bulk properties in thick samples but also makes extreme environmental conditions possible, often requiring massive sample containers.

- the fact that neutrons not only interact with nuclei but also with magnetic moments, which makes them the best available tool for the study of collective magnetic phenomena on a microscopic scale.
- the fact that not only the wavelength of thermal neutrons is in the range of atomic distances in condensed matter but also their kinetic energy is of the same order of magnitude as the energies of elementary excitations, which makes structural and dynamic studies possible at the same time.

For several decades it was a fact that neutrons are much more expensive than X-rays, but this situation is about to change with the advent of the synchrotron light sources, which are not much cheaper than neutron sources any more and which require instruments of a degree of sophistication which is at least comparable to, if not more demanding than, present day neutron spectrometers.

This fact, and the uniqueness of the neutrons as a scientific tool, together with the fact that neutrons are making their way also towards more routine technical and industrial applications not only justifies but makes it mandatory to give some thought to the question, how the full spectrum of neutron scattering techniques can be improved and made available to a growing community of users in the future.

2 Directions of Progress

Although generally hard to predict, the likely development of the scientific needs and the instrumental techniques is an important factor when contemplating the layout and design of a possible future neutron source. While a certain flexibility to adopt upcoming new ideas must always be retained, several trends are clearly observable at present:

- **Improved resolution**

  As the information extracted from scattering experiments becomes more and more detailed, the need for continuous improvement in resolution becomes more and more obvious. Since it is not possible to produce monoenergetic neutrons, the desired energies have to be selected from a wide band, with a corresponding loss in intensity and hence counting statistics as the resolution is improved. On cw-sources this tradeoff between intensity and resolution is unavoidable. The idea with pulsed sources has been to provide the higher resolution by time of flight techniques and compress as much as possible of the total intensity into those time bins actually used. This lead to the lengthy discussions on an optimum moderator design which have dominated much of the previous ICANS meetings.

- Smaller or more dilute samples and small cross sections

In many cases the amount of sample material available can determine the duration of a measurement because of the limited luminosity of the source. While in some instances focussing techniques can help to to a certain extent, the problem is not much diffrent from the resolution problem; we are just looking at different coordinates of phase space.

As for very dilute samples it is one of the beauties of neutron scattering that large volumes can be used if no strongly absorbing materials are present.

Small cross sections of the phenomena under investigation will require good enough statistics to separate off those events which are not of interest. This is sometimes a matter of resolution, some times of intensity but often also a question of the availability of the optimum measuring technique.

Parametric studies

Since transitions from one state to the other are among the most fascinating problems in many particle systems, it often becomes necessary to measure the scattering as a function of several external or internal parameters of the system, or, even more difficult, as a function of the rate of change of these parameters. This is certainly a field where substantial progress is still ahead, provided we can sharpen our tool enough to tackle these problems.

More clear cut information

Although neutron scattering in itself is comparatively easy to interpret and well understood, there remains one parameter which is usually not fully exploited and hence the interpretation of the scattering data often requires additional information. This parameter is the spin dependence of nuclear scattering which is the main cause for neutron scattering to be either coherent or incoherent. Separating these two types of scattering by analyzing the polarisation before and after scattering offers great potential but is on the expense of at least a factor 4 in intensity if done in the traditional way. However, recently some hope has been created that, on sources with a suitable time structure, this loss factor may be reduced by separating neutrons with the spin up and down in time and then flipping the spin of the ones coming in later (H. Rauch, 1985 / H. Rauch, 1986).

– Extended range of neutron energies

The fact that the neutron's kinetic energy when in equilibrium with an ambient temperature moderator is in the range of the most common exitation energies in solids originally triggered the success of inelastic neutron scattering. However, excitation energies in many particle systems range from very low up to several eV or more. As a consequence, and also to be able to work around existing dispersion relations and conservation laws, the range of neutorn energies used has been expanding continuously.

Mainly for practical reasons this expansion first went to the low energy side. Here it is now about to reach its limiting value with "ultra-cold" neutrons which can be stored in a vessel because they are totally reflected from the walls at all angles. Since the density which can be obtained with these neutrons depends on the phase space density and hence the flux level at which they are created, sources with time structure offer a potential for extra progress.

On the other end of the useful neutron spectrum, at epithermal energies, the expansion has been limited by experimental difficulties on cw-sources. These result from the difficulty of providing suitable phase space shaping devices such as high performance choppers and from the extremely small take-off angles from monochromating crystals. Here, too, pulsed sources brought about substantial progress because (a) epithermal neutrons are more abundant in their spectrum and (b) time-of-flight techniques are better suited in this energy range but require an intrinsically short pulse.

3 Ways of Improvement

The relatively short history of neutron scattering has seen a spectacular development both in the intensity available from the reactors, which are the traditional neutron sources, as well as in sophistication of the experimental techniques used. The new ANS project (C. West, 1988) represents the latest state of the development of fission neutron sources and, as far as one can see today, it is definitely exploiting the limits of what is considered as feasible with present day technologies.

On the other hand, the need to select only a small fraction of the neutrons produced for many of the experiments has triggered considerable thought, how the efficiency of neutron use could be improved. In many cases this is possible by moving away from an even time distribution of the neutron production and thus by exploiting an extra open parameter. Very significant advantage factors have been achieved in this way on the existing spallation neutron sources and we are presently in a situation where the pulsed source ISIS at the Rutherford-Appleton Laboratory can in several fields compete with or even outdo the thousand times more powerful high flux reactor in Grenoble. However, this spectacular success is limited to only a relatively narrow range of the spectrum of scientific activities in question. Also, in order to achieve this success, a specialisation on short pulse time-of-flight techniques was necessary. This generally means that only 5 % of the total possible flux is permitted to leave the moderators in order to obtain the desired short pulses. Still this figure compares favourably to the number around 0.01 % one would obtain if a similar TOF-resolution would be attempted on a cw-source.

Thus, on the source side, two ways of improvement can be seen at present:

- increase the neutron production rate and/or

- improve the utilisation factor for the neutrons produced.

For accelerator driven neutron sources, increasing the neutron production rate means either a more powerful accelerator or a higher yield target. While the first option is primarily a question of money, with a lot of progress possible both from a theoretical and a technical point of view, there is a physical limit to how high a target yield can be anticipated from the spallation process. Roughly speaking, this is about 20 n/p/GeV. If one wants to go beyond this number, one would have to ressort to fission as a way of neutron multiplication. Although a viable option on low power sources (J. Carpenter, 1988), this introduces a qualitative difference from the concept of a spallation target in various respects:

(a) **For a SPNS (Short Pulse Neutron Source), thermal fission in the booster has to be avoided since this would lead to a source pulse whose length is determined by the decay constant of the thermal neutron field in the moderators. As a consequence, the target (1) has to be heavily decoupled to allow fast fission only and (2) becomes very massive and large due to the smaller probability for fast fission (G. Bauer, 1986).**

(b) **A booster target will thus contain a substantial amount (up to more than 100 kg) of fissile material (U-235 or Pu) and hence will require a different operating license and safeguarding measures.**

(c) Most of the heat generated in a booster stems from fission and, depending on the desired multiplication, so do most of the neutrons, making the accelerator an expensive start-up source.

(d) As a consequence of more severe engineering constraints and increased requirements to safety barriers, as well as the larger extension of the primary source, the effective flux gain from the moderators will be at least a factor of 2 less than the multiplication in the target.

(e) Since the source multiplication is roughly given by

$$M = \frac{1}{1 - k_{eff}}$$

it is necessary to operate a near-critical arrangement to get a good increase in moderator leakage. This introduces a number of problems in avoiding criticality in all stages of manufacturing, installation, operation, removal and disposal.

(f) The target lifetime is not determined by fission burn-up, but by damage due to irradiation and cyclic stresses. It is, therefore, very costly to replace and reprocess the targets at the intervals needed.

(g) Since fission is the dominating process, delayed neutron precursors become abundant. Most of them are created during the pulse in proportion to the multiplication factor M. Since these neutrons will again be multiplied by a factor M as they decay between the pulses, the neutron background between pulses is likely to increase roughly as M^2 (not relative to a tungsten target, where it is very low, but relative to a depleted uranium target!)

(h) About 30 % of the neutrons escape from the moderator at high energies. These normally come during the t=0 pulse and can be suppressed by gating the detectors. This is not true for the delayed unmoderated neutrons. These therefore constitute an extra background problem.

(i) Reducing the problem of delayed neutron background between pulses either requires difficult to build background rotors at each beam line or a reactivity modulation for the target. The latter is technically very complicated although it does not have the same safety relevance as the reactivity modulator of a pulsed reactor.

In view of these facts the technical feasibility of a high-multiplication fissile target will not be discussed any further at this point. It seems more beneficial, at present, to review other possibilities, in which a time structure on the source flux can be exploited. (see also: G. Bauer and R. Scherm, 1986, R. Scherm and H. Stiller, 1984 and G. Bauer et al, 1981).

- The most obvious and widely used way is separation of neutron energies by direct or inverted TOF which either requires intrinsically short pulses or suitable pulse-shaping devices which operate in phase with the source; the first option being clearly preferrable.
- Another obvious option which works on all methods that do not utilize neutron flight time is to take advantage of the short periods in which the desired neutrons arrive at the detector

to reduce backgrounds and higher order contaminations in the beam by a suitable time gate. This option can probably compensate a flux disadvantage factor of 2 to 3.

- Correlation time of flight techniques which are designed to use up to 50 % of the available beam are normally plagued by the fact that the smaller peaks of interst are sitting on a high background of "non-successful" correlations caused by more intense peaks in the spectrum. With a suitable time structure on the source this can be avoided by a coarse conventional TOF selection and a correlation interval restricted to the region of interest (see Fig. 1a and 1b).
- Phase space transformations by moving crystal devices can provide substantial intensity gains in many cases. Such devices usually work in a periodic fashion. The times, when the desired conditions are fulfilled can be synchronized to the pulses from the source, thus using the full time average intensity.
- Another technique, which becomes mainly important when TOF separation is not used in very high resolution work, is to delay neighboring neutron energies by one pulse width in time through an appropriate "detour" in space. In this way, the average flux of the source can be used several times between pulses.
- Finally, using electromagnetic forces on the neutron spin (frequently in conjunction with perfect crystals, H. Rauch, 1985 / H. Rauch, 1986) makes it possible to successively transform different phase space elements or spin orientations into the desired one. These devices require ramping of fields and hence also work in a periodic way with the resulting option of handling the full time average flux on a source with appropriate time structure.

It is important to note that, out of all the options mentioned only the first one requires really short pulses and hence makes it necessary to compromise between pulse length and time average flux. The other methods, some of which still require further development, can mostly handle pulses of a few hundred microseconds duration and actually depend very little on the shape of the pulse.

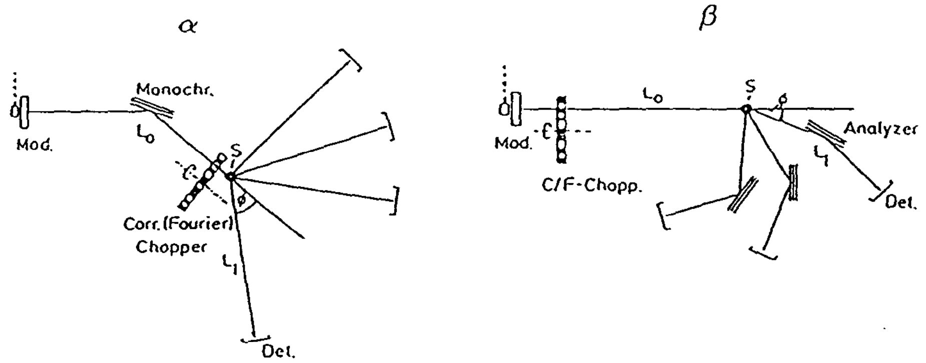

Fig. 1a Univariable correlation (or Fourier) spectrometer at a pulsed source with fixed incident (α) or fixed detected (β) neutron wavelength. The source acts as a conventional chopper of a host spectrometer with coarse time resolution.

Obviously, the gains that can be expected from the various methods of using the time structure relative to a cw-source of equal time average flux vary and also depend on the details of the time structure available. They range from the full peak-to-average flux ratio (or more, considering the transmission losses in choppers) to about an order of magnitude or even less in other cases (R. Scherm and H. Stiller, 1984). Nevertheless, considering what it means to increase the source flux by an order of magnitude, these options should be taken into account when planning a second generation spallation neutron source.

4 Outline of a Medium to High Power Target System

Cw-neutron sources are now aiming at a flux level of $10^{16}\text{cm}^{-2}\text{sec}^{-1}$ (C. West, 1988). Nevertheless it seems appropriate to use the presently available flux level of $10^{15}\text{cm}^{-2}\text{sec}^{-1}$ as a standard for comparison. This means that, in order to be competitive in the majority of neutron scattering techniques and significantly superior in all TOF-techniques one should aim at a time average flux of a coupled and unpoisoned moderator in the regime of a few times $10^{14}\text{cm}^{-2}\text{sec}^{-1}$. From various experimental and theoretical studies presented in detail in previous ICANS-meetings, it is clear that this requires about 1 MW of proton beam power on a pure spallation target.

Other demands to an advanced neutron source with the above characteristics and the resulting technical requirements are listed in Table 1.

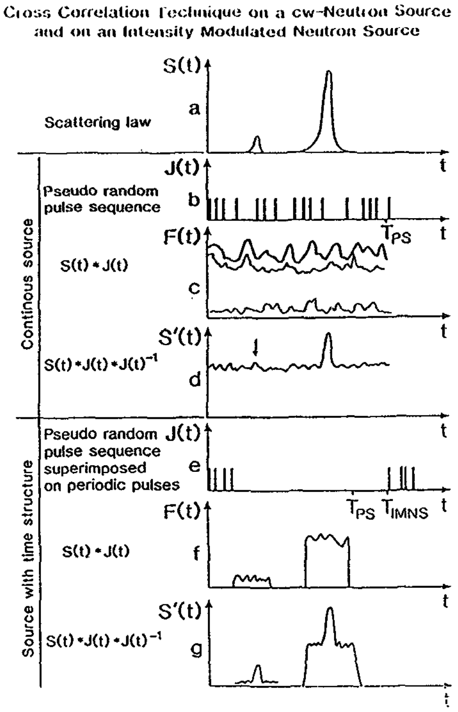

Fig. 1b Correlation spectroscopy on a spectrometer as shown in Fig. 1a

(a) simple TOF-spectrum of the sample showing a weak and a strong line.

(b) Pseudo-random pulse sequence of duration T_{PS} on a cw-source.

(c) Recorded spectrum at the detector composed from two components resulting from the two lines

(d) Spectrum obtained by cross correlating b and c. The weak line is wiped out by the statistical fluctuations of the high constant level caused by the total intensity.

(e) Random pulsing of an intensity modulated source with a period T_{PS} which should not coincide with the source repetition rate T_{IMNS}.

(f) TOF-spectrum consisting of two separated regions obtained by convential coarse time of flight analysis.

(g) Cross correlating e and f results in a very much lower constant level under the weak peak and hence the full gain of the cross correlation technique is obtained for weak as well as for strong lines.

It is obvious from looking at this list that there seem to be conflicting technical requirements especially in the moderator design. Fortunately, however, in contrast to the situation on a fission reactor, a spallation target does not depend on neutron return from the moderator. This gives substantial flexibility in arranging moderators of different characteristics around the target.

As a reference, let us assume that we have protons of 1.5 GeV, 750 μA time average current which are delivered at a repetition rate of 25 Hz in pulses of 5 μs duration. Such beam characteristics might result from acceleration in a linac or in an FFAG and injection into a post-accelerating system of rings like the CERN-booster with successive single-turn extraction from the rings.

Although, at the beam power chosen, an internally cooled tungsten target might still be feasible, it seems desireable to look for a system which has a perspective for further development and with no coolant present in the target region.

Since, as mentioned before, the precise needs which might arise in the future cannot be predicted, not even a possible shift of emphasis in known uses between now and the time when a new generation neutron source might go into operation, it is probably wise to retain as much configurational and operational flexibility as possible and fix those parameters only which require an immediate technical solution.

The configuration outlined in the following is by no means a technically mature proposal but it might indicate a route to follow when most of the above requirements are to be met.

The source shown schematically in Fig. 2 has a unified target-reflector and inner shield system of lead-bismuth or lead which is partly molten during operation with the liquid zore extending as fas as required by the heat transport mechanism.

Lead-bismuth is the choice material from the point of view of operating temperature and expansion upon melting, but, if more detailed studies showed that the temperatures are not a crucial problem and that excessive materials loads due to melting and solidification can be avoided, lead might be preferrable since virtually no α-active products would be created.

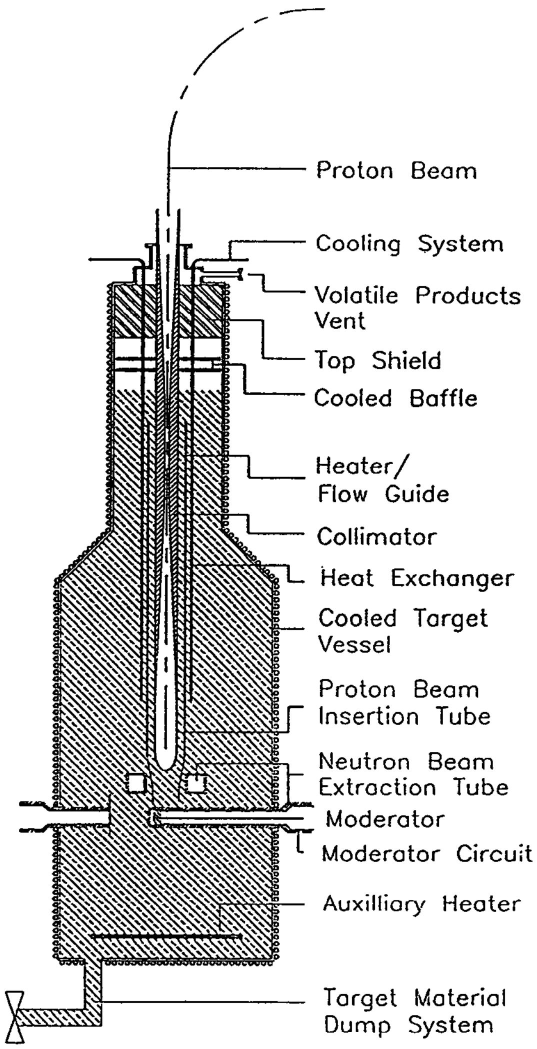

Fig. 2 Schematic representation of a spallation target with integrated heavy metal reflector and inner shield. The target material around the proton beam insertion tube can be melted by the concentric heater. Heat removal from the molten zone below the insertion tube can be either by natural convection (at relatively low power level) or by forced convection, if a pumping system is installed at the top. Cooling is effected by the vertical heat exchanger around the heater/flow guide. This concept allows efficient tailoring of the neutron spectrum by the position of the beam tubes relative to the tip of the beam insertion tube as well as by the choice of moderators inserted in the neutron beam extraction tubes. The extension of the molten zone depends on the proton beam power.

Into the vessel containing the heavy metal there intrude:

- the proton beam insertion tube from the top
- a heater system to melt the target before the proton beam is switched on
- a cooling system to remove the heat created in the interaction zone by natural convection or, if necessary by forced convection.
- several horizontal tubes which are designed to house the moderators and the neutron beam extraction tubes and which are held at low enough temperatures to be surrounded by a solid layer of target/reflector material.

The system also has a dump outlet at the bottom and an auxilliary heater to melt the whole contents of the vessel when it needs to be emptied.

With the proton beam inserted from the top, such a system has the following technical advantages:

- Only the beam insertion tube and the vertical heater are in contact with the liquid metal
- The only solid part with high temperature and high radiation load is the tip of the beam insertion tube. Its integrity can be monitored easily via the vacuum system. If it breaks, the only result is a rise of the target material in the tube.
- The volatile spallation products are collected in the space above the liquid target and hence do not constitute a hazard if the beam insertion tube breaks. They can also be removed continuously or in batches to minimize operational hazards even further.
- Exchange of the beam insertion tube can be accomplished relatively easily by first pulling it up when the target material is molten and then removing it when it is frozen again and all volatile spallation products have been trapped.
- There is no need to ever change the target material

- The water cooling of the horizontal tubes can be used as pre-moderator if designed suitably.
- The design of the moderators can be varied according to the needs arising. They can be exchanged without access to the top of the target area.
- Beam tubes can be designed to serve widely different purposes; from cold neutron beams all the way to keV-beams or irradiation ports, if desired even in the very hard forward or the much softer backward spectrum.

Although it is desireable to have a cooling system at the outside of the target vessel which can be subjected to an internal pressure to help melting the whole target material if necessary, the vessel might be surrounded by a water layer to moderate and absorb neutrons escaping from the reflector-shield system in order to reduce the activation of the surrounding outer shield.

Cold and ambient moderators which are inserted into horizontal beam extraction tubes have been in operation for examples at the 23 MW research reactor FRJ-2 at KFA Jülich for many years and have been performing satisfactorily. Recently such a cold source has also been installed at the HFR in Grenoble.

The decision, wether a given moderator should be viewed from two sides and hence be placed in a through tube or should rather be surrounded by as much reflector material as possible has to be made in the frame of a detailed study.

It is beyond the scope of this paper to persue the concept in more detail but it is likely that, if forced convection is used, such a system could work in the multi-megawatt range since the choice of the material for the beam insertion tube is neutronically not critical and can be entirely determined by radiation and temperature restistance as well as by resistance to corrosion. The tube will have to be exchanged as a measure to prevent damage after a certain operating period.

5 Conclusions

In order to meet with the growing demands both for neutron scattering facilities as such and for more sophisticated experiments to obtain more detailed and clear cut information, a new generation of neutron sources is necessary which allows optimum use of the neutrons produced to give enough intensity for very high resolution work as well as for the investigation of phenomena with very low signal rates. Spallation neutron sources with a short primary neutron pulse and flexibly designed "fast" as well as coupled moderators together with a variety of imminent new phase space handling techniques have a prospect of satisfying the scientific needs also in the future. They are technically feasible both on the part of accelerators, which have not been dealt with in this paper and on the part of target systems, where new design routes can be followed.

Table 1 Demands and Resulting Requirements to the Constituents of an Advanced Spallation Neutron Source

Demand	Technical requirement
Proton Beam	
Short neutron pulses possible	Proton pulses no longer than a few μs
Moderately extended primary neutron source	Proton energy between 1 and 1,5 GeV optimum
Possibility of long flight path spectrometers	Repetition rate around 25 Hz
Average power on target around 1 MeV	Time average proton current around 0,7 to 1 mA
Spallation target	
High neutron yield from spallation	High Z material (Pb, Bi, Ta, W)
Moderately extended primary source	High density target (minimum amount of cooling channels)
Long target life time	Little susceptibility to damage from radiation and stress (liquid optimum)
High safety standard	Target design that prevents escape of radioactivity in case of damage
High beam power	Good heat removal properties
Reflector	
Short neutron pulses	High scattering cross section for fast neutrons. Reflector close to moderator. High density. Little moderation.
High average neutron flux	Low absorption cross section for thermal and epithermal neutrons
High power dissipation	Good heat removal properties
Moderators	
Intrinsically short pulses	Good slowing-down properties, low escape depth; cryogenic moderator
High average leakage flux	Low absorption, good escape probability
Long wavelength neutrons	Thermal equilibrum at low temperature, good escape probability
Epithermal neutrons	High escape probability at epithermal energies; short slowing-down time

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