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Neutron scattering/Diffusion de neutrons

New perspectives from new generations of neutron sources

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Abstract

Since the early 1950s the vital multidisciplinary progress in understanding condensed matter is, in a substantial fraction, based on results of neutron scattering experiments. Neutron scattering is an inherently intensity limited method and after 50 years of considerable advance—primarily achieved by improving the scattering instruments—the maturation of the technique of pulsed spallation sources now opens up the way to provide more neutrons with improved cost and energy efficiency. A quantitative analysis of the figure-of-merit of the specialized instruments for pulsed source operation shows that up to 2 orders of magnitude intensity gains can be achieved in the next decade, with the advent of high power spallation sources. The first stations on this road, the MW class short pulse spallation sources SNS in the USA (under commissioning), and J-PARC in Japan (under construction) will be followed by the 5 MW long pulse European Spallation Source (ESS). Further progress, that can be envisaged on the longer term, could amount to as much as another factor of 10 improvement. ***To cite this article: F. Mezei, C. R. Physique 8 (2007).***

Résumé

Nouvelles perspectives ouvertes par les nouvelles générations de sources neutroniques. Depuis le début des années cinquante, les progrès cruciaux réalisés dans tous les domaines pour mieux comprendre la matière condensée, reposent essentiellement sur les résultats d'expériences utilisant des faisceaux de neutrons. La diffusion de neutrons est, de manière inhérente, une méthode d'intensité limitée. Après cinquante années de progrès considérables, grâce, surtout, à une amélioration continue des spectromètres, la technique des sources pulsées à spallation arrive à maturité pour ouvrir la voie vers l'obtention de plus de neutrons, à moindre coût et avec une meilleure efficacité en énergie. Une analyse quantitative de la « figure de mérite » des appareils adaptés à une opération sur source pulsée, montre que l'on peut gagner plus de trois ordres de grandeur en intensité dans la prochaine décade, avec l'arrivée des sources à spallation de forte puissance. Les premières étapes sur cette voie, les sources de spallation à *pulses* courts, de niveau de puissance du MW, SNS aux USA (en cours de mise en service) et le J-Parc au Japon (en construction), seront suivies par la source à spallation européenne à longs *pulses*, de 5 MW. Les progrès envisageables à plus long terme pourraient permettre de gagner un facteur 10 supplémentaire. ***Pour citer cet article : F. Mezei, C. R. Physique 8 (2007).***

Keywords: Neutron scattering; Neutron sources; High power neutron sources; Pulsed sources; Spallation; Fusion

Mots-clés : Diffusion neutronique ; Sources de neutrons ; Sources de neutrons de haute puissance ; Sources pulsées ; Spallation ; Fusion

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1. Introduction

Neutron scattering as a crucial, powerful microscopic probe to explore condensed matter, effectively started in 1951 with the spectacular debut of the unambiguous experimental proof of antiferromagnetism, recognised by the Nobel prizes awarded first to L. Neel and later to C.G. Shull. For more than 50 years, neutron scattering has been one of the key elements of an ever-evolving suite of experimental probes at the disposal of an also ever-growing pluridisciplinary research community, from physics to cultural heritage, from life sciences to engineering, etc. An illustrative study aimed at giving an idea on the impact of neutron research found that in the period 2000–2004 about 5% of the articles in the field of condensed matter in Physical Review Letters were based on neutron scattering experiments. The current 330 M€ annual spending on the operation of 13 European neutron scattering facilities (including IBR2 in Dubna and the two stopped reactors in Risø and Jülich, which absorb for the moment substantial yearly budgets for decommissioning), and the about 270 M$ annual spending for 5 neutron sources in the US, are used to support many fields of research, with the share of condensed matter physics today being less than 50%. While it is difficult to evaluate the total yearly expenditure on condensed matter research, on the basis of an estimated 40 000 scientists, spending on average 50% of their time on research, one can guess that it should exceed 10 billion € per year on both continents combined. (This number looks reasonable in view of the total US basic research spending of about 70 billion $ per year.) Thus neutron scattering proves to be a cost effective way to produce significant, high impact scientific results, in contrast to the impression that the high individual costs of neutron facilities may superficially suggest. In this context it is also worth mentioning that, with on average 9 days of instrument beam time per refereed publication at the ILL, Grenoble, the current world leader in neutron scattering (including operational tests, instrument development and preparation) and 15–18 days at medium flux facilities such as LLB, Saclay, the purely neutron facility costs per publication (including all staff and material costs) amounts to 90–100 k€. This sum is rather less than the comparable full costs of the research behind a publication under common conditions, using in-house laboratory scale equipment e.g. at universities, but it does not contain the expenses and time spent on the users' side. Without users, the scientific productivity of neutron centres would be some 2–3 times less (judged from earlier examples), correspondingly driving the costs up to levels which would make the scientific case for funding rather unfavourable. The main innovation brought by ILL to condensed matter research at large scale facilities was the user operation concept, an extremely brilliant and far sighted idea of Rudolf Mössbauer, the first director of ILL after operation started. (This idea was first received by the community of neutron scientists with all the incomprehension and shock that characteristically meets true innovation). Thus, while the proven record of neutron scattering research shows that it is great value for the money, the costs involved are important (e.g. compared to synchrotron radiation facilities) and they represent one of the decisive issues to be considered for future neutron sources.

Although the brilliance of neutron sources has only very moderately advanced since the construction of the first steady state research reactor primarily designed for neutron beam research in the 1950s, neutron scattering made huge progress in the past half century by advances in neutron instrumentation techniques. Nevertheless, it remained well known as an intensity limited technique, and it will continue to remain so—although at a completely different level—even if we manage to gain a few orders of magnitude in neutron flux. The key question about future facilities is how to achieve substantial gains in beam intensity without exploding the costs. The only ways known to us to produce neutrons are nuclear physical processes, which produce fast neutrons, while requiring substantial energy input, or creating substantial amounts of energy (heat). So, ultimately, the cost of neutron production is basically tied to the costs of energy. In this respect, it makes no fundamental difference if the energy is produced in the process (as in fission reactors) or it is absorbed to operate the source (as in accelerator based neutron sources, e.g. spallation sources). Namely, in this example, the costs of operating a fission reactor, including fuel, are pretty comparable to the costs of buying the electricity which could have been produced by the fission reactor. One cannot optimize a facility at the same time for both purposes of producing neutrons with optimal parameters for beam research (i.e. high power density in a volume of few litres for high brilliance) and producing electricity (i.e. bringing large amounts of thermo-hydraulic medium to highest possible temperatures). A facility can only really do one task or the other.

Besides costs there is another fundamental aspect of life that cannot be circumvented by any kind of ingenuity: thermodynamics. Indeed, removing the heat deposited in a necessarily minimized volume, in order to maximize source brightness, is one of the toughest challenges in neutron source design, and in this respect it does not matter whether the heat was created in the process of neutron production, or deposited by the particles from an accelerator. A third, fundamental limitation to juggle with, is radiation damage and other material resistance issues in the high energy density environment of the neutron source.

All this leads to the inevitable conclusion that the energy creation and/or deposition per fast neutron produced and the efficiency of transforming the fast neutrons created at the first place into thermal ones on the sample are the key characteristics of the neutron sources, that determine their practical field of usefulness in the future. Therefore, for flux limited neutron scattering research in condensed matter, where the progress of neutron sources is measured by the useful neutron intensity, we need to minimize the energy involved per useful neutron delivered. For other purposes, for example irradiation, dosimetry, neutron technology developments, the optimization criteria might be completely different, for example minimizing the costs for a given modest neutron source intensity.

2. More neutrons by enhanced efficiency: basics

With the 58 MW thermal power of ILL we have practically achieved both the thermodynamic and cost limits for steady state high flux research reactors. Fission frees up about 190 MeV energy per extractable fast neutron (i.e. beyond those that are re-absorbed and feed the chain reaction) and the 1.5 MW/l heat produced within the fuel element at ILL is close to the upper limit of what can be removed. The slow neutron brilliance of the source is closely related to the fast neutron brilliance of the 'core' or 'active zone', but not in a simple fashion. Slowing down of the fast neutrons involves collisions in reflectors and in moderators. For example, for thermal neutrons at ILL, both the moderator and reflector is provided by an about 2.5 m diameter D_2O tank, while the hot and cold moderators (respectively, heated graphite and liquid D_2) occupy volumes of the order of 20–30 litres in this tank. Although with other choices of reflectors (Be, graphite, Pb) the reflector volume can be smaller, as determined by the mean free path of the fast neutrons, we would still have to work with a tank typically more than a meter in diameter. In order to maintain optimal efficiency of fast to slow neutron conversion in terms of moderator brilliance, the source of the fast neutrons must have a volume considerably smaller than that of the reflector. For this reason it helps little for the final source performance (a) to produce more fast neutrons by increasing the volume of the active zone at equal power per volume (the about $\sim$40 litres at ILL is a reasonable upper limit for good efficiency) or (b) to enhance the fast neutron brilliance by making the active zone much smaller than a few litres at equal total number of neutrons produced.

This latter point is particularly significant. By photo-neutron production processes (irradiation of light elements with γ-rays) we could achieve a higher fast neutron production density in a small volume than with other methods [1], due to the lower heat deposition per fast neutron. However, the energy needed to produce the γ radiation is about 300 MeV per neutron. Thus this thermodynamically favourable option is ruled out for future high flux neutron sources for condensed matter research by the unfavourable energy balance: it does not allow us to produce more neutrons at the same energy costs, and reducing the active zone volume way below that of ILL, does not much affect the slow neutron brilliance. On the other hand, it could be an interesting neutron source for nuclear physics purposes, with its high brightness of fast neutrons produced in small, pencil size volumes.

Steady state fission neutron source performance, not surprisingly, did not progress much over the past half century, since the first reactor primarily designed for neutron scattering research was completed at Chalk River in 1958. The thermal slow neutron flux (with neutron energies centred around some 30 meV) has only increased by a factor of 4 by now (at ILL), and the only progress exceeding an order of magnitude is due to the use of cold and hot moderators (neutron energies centred around 5 and 200 meV, respectively) that enhanced the brilliance in a specific neutron energy range. In contrast, the potentials of source performance improvements that have been recently opened up amount to 2–3 orders of magnitude, to be achieved by optimizing the energy efficiency of neutron production and, at the same time, approaching the source power levels comparable to steady state reactors. There are basically two contributions to this efficiency enhancement: (a) making actual use of a higher fraction of the produced neutrons by going from steady state to pulsed sources; and (b) reducing the energy involved in fast neutron production by going from fission to spallation.

Since neutron scattering work requires more or less monochromatic incoming beams and neutron moderators can only produce quasi-Maxwellian velocity distributions (leading to a distribution of wavelengths λ via the de Broglie relation $\lambda = h/mv$), the monochromatization process at a continuous source involves eliminating 90–99.99% of the beam spectrum, depending of the various experimental needs, essentially independently of the method used for velocity selection (crystal monochromators, time-of-flight choppers, velocity selectors, ...). This is a very fundamental point, and it can be illustrated by comparing two types of diffractometers for steady state sources, the standard crystal

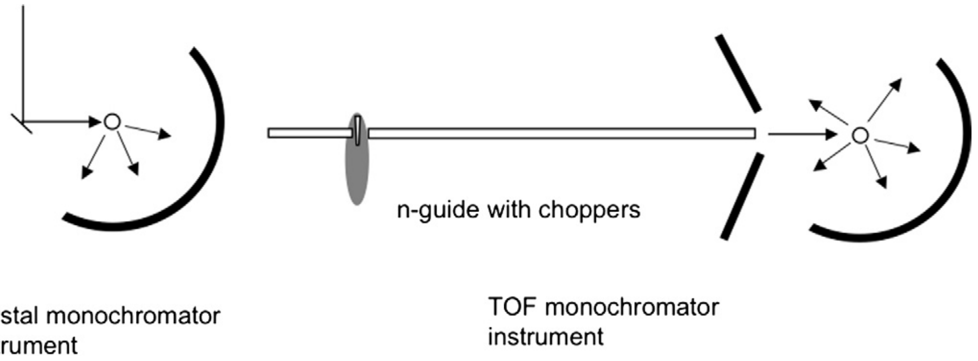

Fig. 1. Principles of crystal and time-of-flight (TOF) beam monochromatization options for neutron diffraction at a steady state neutron source. In the TOF alternative pulses are cut out of the continuous beam by a chopper. The thick black lines represent detector banks. For simplicity, auxiliary components, e.g. collimators in the crystal monochromator instrument or additional choppers to keep neutrons from different pulses separated in the TOF instrument, have been omitted [2].

monochromator instrument and the time-of-flight (TOF) diffractometer [2], cf. Fig. 1. The latter is a visionary proposal by Buras made in the 1960s, whose merits have been extensively analyzed a long time ago [3], but only recently experimentally demonstrated [4]. The crystal monochromator only reflects neutrons within a narrow selected wavelength band $\delta\lambda$ from the broad Maxwellian spectrum. In the TOF monochromator a mechanical chopper upstream in the incoming beam is only open for a time δt, but it lets through most of the neutron spectrum as determined by the auxiliary choppers downstream, i.e. to the right in the figure, not shown. (These auxiliary choppers also assure that at a given time only neutrons from a single primary chopper pulse can hit the sample [2,4].) Then the travel time to the detector will be the measure of the neutron wavelength λ. The time between subsequent neutron pulses needs to be comparable with the average travel time of the neutrons to the detector, t, so the fraction $c = \delta t/t$ of neutrons let through by the chopper (which is called the duty factor) is comparable to the wavelength resolution $\delta\lambda/\lambda$, which is just about the fraction of the neutron spectrum reflected by the crystal monochromator. Thus, indeed, at a steady state source, the beam intensity on the sample does not fundamentally depend on the method used for monochromatization (except for a few special secondary circumstances, whose impact will be discussed below).

The main difference is that for the TOF method the source does not need to be 'switched on' all the time; it can be shut off for most of the time between pulses. (Actually, making the source pulsed can assume the role of the primary chopper in Fig. 1.) This is a decisive opportunity to gain in efficiency of the use of the neutrons produced. This approach has been primarily pioneered by our Russian colleagues by building the pulsed reactor sources in Dubna. If the source is switched on with a duty factor c large enough for the TOF instrument to 'see' the source 'on' whenever it needs it, we realize an a priori gain in neutron production efficiency of $1/c$ compared to the source being 'on' all the time, as is necessary in order to run the crystal monochromator instrument. The fraction of the time the source needs to be 'on' can be roughly estimated as the relative beam monochromatization $\delta\lambda_{\text{required}}/\lambda$ one would aim for at the continuous source, to do the given experiment under optimal conditions (i.e. best compromise between intensity and resolution). Indeed, if we take the duration of the source pulse as the δt above and the time between source pulses t for the measure of the typical time left for the neutron to travel to the detector, the duty factor $c = \delta t/t$ will be the wavelength resolution $\delta\lambda/\lambda$ the pulsed source naturally provides. This implies that the source is not 'on' long enough to fully serve the TOF instrument if $c < \delta\lambda_{\text{required}}/\lambda$, which will reduce the efficiency gain f_P by pulsed operation correspondingly. This can be formulated by the following expression:

$$f_P = \min(1/c, \lambda/\delta\lambda_{\text{required}})$$

Note that for an actual pulsed source the duty factor c (defined in terms of the actual neutron pulse length) can be a strong function of the neutron wavelength λ, and we also need to keep in mind that the beam monochromatization needed in various cases can change from some 0.01% to about 10%. On average, across the board of the spectrum of the different kinds of neutron scattering experiments, the gain factor f_P can reach a value of about 50.

Another way of appreciating the source efficiency gain one can achieve by pulsed operation is to observe that neutrons with different velocities originating from the same source pulse arrive at the sample (detector) at different times. Thus at any given time we can work with monochromatic neutrons with known velocities (within the uncertainty determined by the source pulse length and the neutron travel time) while we essentially make use of the full neutron spectrum. This is in contrast with the need of eliminating a 90–99.99% fraction of the spectrum, as mentioned above for the monochromatization process at continuous sources, where neutrons with all the different velocities arrive all the time at the instrument.

This basic comparison of steady state and TOF experimental approaches is based on the assumption that incoming beam divergence and the scattered beam solid angle is similar in both cases and we can concentrate on the effects of beam monochromatization only. This assumption is somewhat oversimplified in cases where special circumstances allow us to use beam focussing and relax collimation or resolution in certain directions. This can work in both directions. For example in the case shown in Fig. 1, for the monochromator instrument, one can enhance the beam intensity by using an array of monochromator crystals which focus the beam perpendicularly to the horizontal plane of the drawing (scattering plane) and even some 10° vertical divergence of the focussed beam on the sample would not affect substantially the high precision angular scan in the horizontal plane. In the TOF case beam focussing can only be realized with much less efficiency since we will need to use instead of crystals neutron supermirrors, which reflect in a broad wavelength band but only under small angles. So the vertical beam collimation will only achieve 1–2° in the relevant wavelength range and this will affect the beam intensity in similar proportions. However, for a high resolution angular scan we will need horizontally better collimated beams and fine collimators in front of the detectors (which cut angular range of detection by more than an order of magnitude) for the crystal monochromator machine, while the wavelength scan on the TOF instruments makes it possible to obtain high resolution data with less fine beam collimation and without the collimators in front of the detectors, if we concentrate on high scattering angles. On average, these secondary circumstances largely compensate each other. As a matter of fact it incidentally turns out, in the case of diffraction shown in Fig. 1, that the collimation requirements make the TOF method clearly superior to the crystal monochromator approach in data collection rate at high resolutions, while the opposite is true at moderate resolutions.

There are different types of pulsed spallation sources in existence, under construction or proposed/planned. The pulsed fission reactor IBR 2 in Dubna produces neutron pulses of about 300 µs duration at a 5 Hz repetition rate. In the so-called short pulse (SP) spallation sources (such as ISIS near Oxford) an accelerator system delivers about 1 µs short proton pulses to a heavy metal target to produce fast neutrons, but after slowing down to the energies needed for neutron scattering work, the FWHM length of the neutron pulses will vary between a few µs and ∼300 µs, depending primarily on the neutron wavelength and also on the target assembly design details. The most advanced short pulse spallation sources under commissioning/construction, SNS at Oak Ridge, USA (cf. Fig. 2) and J-PARC at Tokai, Japan will be the first pulsed spallation sources to accede the 1 MW power. Long pulse (LP) spallation sources will represent the next leap in source power (such as the planned 5 MW European Spallation Source, ESS). Here the accelerator will send proton pulses in the range of 1–2 ms length to the target, and (in view of the above up to 300 µs response time in neutron slowing down) this will also be the FWHM pulse length of the neutron pulses essentially for all wavelengths.

The pulse repetition rates of pulsed sources in existence, or planned, range from 5 Hz to 120 Hz, as a result of complex practical compromises. The main boundary condition here comes from the fact that neutron instruments need to have sizable dimensions, typically from a few meters to a few times 10 m. The ultimate reason for this is the famous intensity limitation of neutron beams, their small brilliance compared e.g. to laser or X-ray beams. This forces us to use beam cross sections of several cm^2 illuminating samples of similar size (if we are lucky with sample preparation), as opposed to the minute beam cross sections in other types of scattering work. In consequence—in order to maintain good definition of beam direction—the size of the whole instrument needs to scale with the sample size. This is most significant for small angle neutron scattering (SANS) instruments, where angular resolutions for neutron trajectories in the 10^{-3} rad range are indispensable. There are several other practical conditions acting in the same direction, e.g. the ^{3}He gas detectors—which offer the lowest spurious background, a crucial requirement when working with limited intensity—need to be a few cm thick. Therefore long flight paths are needed if we want to determine neutron velocities with high precision. For these and additional practical reasons, the neutron detectors in the instruments will be placed typically between 20 and 100 m from the source, so we need to leave sufficient time for the Maxwellian neutron spectra emitted by the source moderators to arrive to the instruments with little overlap. This would suggest packing all the neutrons produced by the source into few pulses separated by long time intervals. On the other hand, the toughest technical challenge with pulsed sources is exactly to increase the energy deposited in a single pulse, and the same source power could be much easier delivered in mini-pulses at higher frequencies. The reasonable compromises for spallation sources between these two opposite boundary conditions are about 50 Hz pulse

Fig. 2. The SNS pulsed spallation source facility at Oak Ridge. H^- ions are injected at the front end into the 1 GeV linear accelerator (linac), in which most of the acceleration happens inside superconducting Nb cavities. The 1 ms long linac pulses are then injected as protons H^+ (after getting stripped of two electrons) into the accumulator ring, where the protons from the linac pulse go around with about 80% of the velocity of light many hundreds of times until the whole linac pulse has arrived. The protons thus accumulated from one linac pulse are then fired as a less then 1 μs burst from the ring to the target. Some of the neutron instruments surround the target moderator assembly within the target building, others are situated at the end of longer neutron guides outside the building. At a long pulse source the protons accelerated in the linac are led directly unto the target, without passing through an accumulator ring.

repetition rate for thermal neutrons and about 20 Hz for cold neutrons (with the typically exploited velocity range in the Maxwellian spectra starting at about 1500 m/s and 500 m/s, respectively).

In spallation, high energy protons are shot into a heavy metal target, and in a cascade of reactions, a spectrum of fast neutrons in the few MeV energy range is emitted by breaking up the nuclei. This approach of producing neutrons for condensed matter research was pioneered by Argonne National Laboratory since the early 1970s, although the idea came from Russia, and the US patent on spallation was awarded to a group from Leningrad. For best efficiency in terms of neutron emission per proton beam energy delivered, the protons need to have energies around 1.3 GeV, where about 40 MeV proton beam energy is needed to produce a fast neutron. The process is endothermic, so the heat deposited in the target is somewhat less (by about 30%) than the proton beam energy delivered. This is indeed much less than the about 190 MeV for fission. The neutron production efficiency is a rather flat function of the proton energy in the range of 0.5 to 10 GeV; the different spallation sources in operation or planned actually use proton energies from 0.45 to 3 GeV. The energy efficiency factor of neutron production f_E can be given as

$$f_E = \eta_{\mathrm{fs}}/E_n$$

where E_n is the energy associated with the production of one fast neutron and η_{fs} is the efficiency of converting fast neutrons (total production per unit time: [n/s]) to slow neutron moderator time average brilliance: [$\mathrm{n/cm^2/Å/sterad/s}$]. In absolute terms, this latter factor would be very small and hard to evaluate from first principles, so for our practical purposes we will measure this quantity relative to the efficiency achieved at ILL, i.e. per definition $\eta_{\mathrm{fs}}(\mathrm{ILL}) = 1$. For the generally adopted spallation source target moderator system design approach [5] ex-

Table 1
Empirical estimates for the relative efficiency η_{fs} of converting fast neutron production rates in state-of-the-art spallation target stations into slow neutron moderator brilliance

Type of spallation source moderator	η_{fs}
Thermal neutron coupled moderator	∼1
Cold neutron coupled moderator	∼2
Hot neutrons from thermal moderators vs. ILL hot source	∼0.1
Hot neutrons from thermal moderators vs. ILL thermal beam tubes	∼8

The definition of η_{fs} is given in the text.

perimental evidence and neutron transport calculations vary around the values in Table 1 below. Thus e.g. for cold neutrons at spallation sources the energy efficiency factor f_E will be about 10 times more favourable than for a fission reactor such as ILL.

The combined efficiency factor of spallation sources $f = f_P f_E$ show a gain compared to ILL type reactors, which amounts to 300 and 600 for thermal and cold neutrons, respectively. How much of this efficiency gain can be turned into beam intensity gain on the sample depends on the proton beam parameters we can envisage to use at a spallation source. One can safely assume that the technical limits of high power pulsed spallation sources lie above 10–15 MW proton beam power at the optimal 2% proton beam duty factor (in order to best match the average wavelength resolution requirement, cf. above) at about 20 Hz pulse repetition rates (in order to leave enough time between pulses to allow much of the cold neutron spectrum to arrive separately at the sample). Thus within the next few decades, spallation source technology will provide an unprecedented 50–150 fold quantum leap in neutron source performance across the board for all applications in condensed matter research. It is also to be expected that, on the longer run, the power limits, not yet known today, will be more closely approached, and we can gain another factor of 5–10 by increasing the spallation source power to the level of that of the most powerful research reactors, ILL and HIFR at Oak Ridge. Indeed, the proton linear accelerators (linacs) considered today are very far from the tough fundamental accelerator physical limits (such as space charge effects) and a solution to the target material damage issues could eventually be found, for example, by such old ideas like the liquid metal jet target, originally invoked in the Intense Neutron Generator (ING) project in Canada in the 1960s.

Note that in view of the relatively low extent of the use of hot and epithermal neutrons in scattering work (less than 10% of the experiments), the progress of neutron sources considered here primarily reflects the across the board average over the different kinds of thermal and cold neutron experiments. The hot neutron regime ($E > 100$ meV) is more complex for several technical reasons. Here, on the one hand, pulsed spallation sources without a hot moderator have a very hard time to compete in single crystal studies with steady state reactors equipped with a hot neutron source. On the other hand, the situation is fully the opposite for some other types of experiments, most notably eV inelastic spectroscopy. The main drivers of the evolution of neutron sources are, and must necessarily be, the cold and thermal neutron scattering capabilities, which provide the research opportunities most unique for neutrons e.g. compared to X-rays. Nevertheless a relatively small number of hot/epithermal beam lines distributed between existing reactors and spallation sources will continue to provide for impressive progress of research in this neutron energy range as well, for the foreseeable future.

We have stressed by now that, in the quest for highest neutron intensities, ultimately the energy efficiency will be the decisive factor. Indeed, in the higher power regime, most of the electricity consumed by an accelerator (especially for linacs) is used to accelerate the particles, and only a smaller fraction keeps the auxiliary systems (such as magnets, pumps, etc.) running. Current accelerators have not yet reached this limit, but this is what we are going to approach with sources such as ESS with 5 MW proton beam power. Looking to this from another side, aiming at low beam power will not reduce anything close to proportionally the costs of building and operating spallation sources. Just to achieve proton energies in the GeV range, especially if ring accelerators are considered in order to produce short pulses, requires a lot of hardware to start with, and lot of electric power to operate it, even at essentially zero particle beam power, which is the usual case in high energy physics. There are two points of prime interest to remark here.

A very innovative approach has recently been proposed and is, in the meantime, largely implemented at the University of Indiana in Bloomington, to produce neutron beams of moderate intensity practically on laboratory scale at unprecedented low costs [6]. This consists of using the p(Be)n reaction by irradiating a thin Be target with 7–13 MeV protons (which cannot traverse much material). The proton beam can be produced by a small linear accelerator of

10–15 m length. The construction costs of such a neutron source, dubbed LENS (Low Energy Neutron Source), are in the range of a few % of a modern spallation source. The energy efficiency of this method is rather poor (about 1500–2000 MeV per fast neutron produced), but at about maximum 50 kW power (the practical limit a thin Be target can probably take) a considerable pulsed neutron beam flux can be produced for many purposes, which earlier required a fission reactor, typically of a standard type called MTR ('material testing reactor').

On the other extreme of the power scale, a prospectively very important feature of high power spallation technology is that it produces large quantities of neutrons just from electricity—in contrast to fission or fusion. The energy efficiency of this neutron production is so high, that it can substantially change the macro-balance between energy production and neutron production in fission reactors: if all the electric energy produced by the fission process in a power plant is used in a spallation source, we could produce on average additionally 1.5 neutrons per fission reaction. This extra number of neutrons is more than sufficient to 'burn', i.e. fission, all actinides in the nuclear waste produced in the first place, such as Np, which requires to absorbe more than one neutron before coming to fission and liberating the usual about 190 MeV fission energy. Thus, using some extra neutrons from spallation in fission based nuclear energy systems, there is a potential to extract all fissile energy from the nuclear fuel, leaving behind a fully energy depleted, essentially actinide free nuclear waste, that would decay in a few hundred years to levels of radioactivity lower than that of the uranium ore originally collected.

The only known reaction that produces fast neutrons at the expense of lower energy/heat production than spallation is hydrogen fusion at about $E_n = 20$ MeV per neutron. The great disadvantage of this reaction is that the neutrons are generated with very high energy (14 MeV) and therefore the conversion efficiency η_{fs} from fast to slow neutrons will necessarily be much lower than either for spallation or for fission neutron sources, more than removing all the advantage from higher energy efficiency in fast neutron production. This is indeed confirmed in a recent publication, which estimates $\eta_{\text{fs}} \sim 0.05$ for thermal neutrons and even less for cold neutrons [7]. Therefore, to be competitive with a 10–15 MW long pulse spallation source one can envisage today on a medium term (cf. above), a fusion based pulsed neutron source should display about 100 MW power. Even if inertial fusion technology would become available within a few decades, the complications and costs due to lower energy efficiency would practically rule out the fusion source option. Considering much higher powers, say in the 100 MW range for spallation to be compared with a 1 GW inertial fusion source, on the one hand, the technology developments needed to realize spallation at this power level are far less far-fetched than those necessary in the case of fusion. On the other hand, the low energy efficiency of the fusion facility will remain a decisive drawback: the 1 GW fusion based neutron source cannot be at the same time a fully efficient 1 GW power plant, although some energy recovery is certainly envisageable. So the operating costs will add to the costs of lost electricity that the system failed to generate for the power grid from the amount of fusion fuel used. These costs alone might be sufficient to pay for the whole utility bill of the neutronically similarly powerful, 100 MW long pulse spallation source, that will in addition use less demanding technology. Some form of energy recovery may also become feasible for high power spallation sources since (unlike fission based research reactors) the cooling agents for spallation targets can, in principle, be allowed to reach the high temperatures common in the energy generation.

3. Source performance and Figures of Merit (FOM)

With the large variety of possible experimental conditions and requirements, the peak neutron brightness of the source pulse (which for a continuous neutron source is to be understood as the steady state brightness) can be shown to provide a good metrics for the figure of merit of the source for neutron scattering work, if and only if three main conditions are fulfilled:

(a) the neutron beam delivery from source to sample is accomplished without substantial beam losses, i.e. it is essentially governed by the conservation of the phase space density following Liouville's theorem;
(b) the actual monochromatization of the beam impinging on the sample is not better than the wavelength resolution required for the specific experiment considered;
(c) the source pulse parameters (pulse shape, repetition rate) are reasonably convenient.

In various neutron scattering experiments, different incoming neutron wavelengths are used, and the above three conditions are in general differently fulfilled for the different neutron wavelengths emitted by the source. Therefore we can reasonably define a wavelength dependent source figure-of-merit (FOM) as follows:

$$F(\lambda) = \Phi(\lambda).\eta(\lambda).\min\bigl[1, (\delta\lambda/\lambda)_{\text{delivered}}/(\delta\lambda/\lambda)_{\text{required}}\bigr].P(\lambda)$$

where $\Phi(\lambda)$ is the (instantaneous) brightness of the source moderator at the peak of the pulse; $\eta(\lambda)$ is the beam delivery efficiency with respect of absorption losses, data collection dead time etc.; the basic incoming wavelength resolution at the given instrument $(\delta\lambda/\lambda)_{\text{delivered}} = c(\lambda)T/t(\lambda)$ is determined by the source duty factor $c(\lambda)$, the source repetition time T (so that $c(\lambda)T$ is the actual neutron pulse lengths) and the arrival time $t(\lambda)$ of the neutron at the sample (detector); $(\lambda/\delta\lambda)_{\text{required}}$ is the ideal incoming wavelength resolution for the experiment considered and $P(\lambda)$ is the pulse shape factor to characterize the information content of a pulse compared to a standard Gaussian reference. This definition of neutron source FOM has been successfully benchmarked in a number of Monte Carlo virtual instrument simulation calculations within the framework of the European Spallation Source (ESS) project study [5].

For spallation sources $\eta(\lambda)$ can be assumed to be close to unity (within some 30%) for modern instrument design for all neutron wavelengths >0.9 Å. This is based on two recent developments, which have been adopted as design standards for pulsed spallation source instruments world wide: (a) supermirror based low loss 'ballistic' neutron guides [8]; and (b) 'repetition rate multiplication' (RRM) methods [9]. The first allows us to deliver neutron beams with moderate intensity losses, independently of the distance of the sample up to several 100 m. (With decreasing wavelength below 0.9 Å the flux on the sample will increasingly become a function of the distance). Repetition rate multiplication type chopper systems make possible to deliver to the sample sharp triangular or trapezoidal shaped pulses of adjustable length with arbitrary repetition frequency, within reasonable limits, independently of the source pulse repetition rate. In order to achieve this one needs to have a reasonable freedom in choosing the distance between source and sample (as provided by advanced, low loss neutron guides [8]) and in elastic (diffraction type) experiments the source pulses need to be 1 ms long or longer for high efficiency pulse shaping.

The $\min[1, (\delta\lambda/\lambda)_{\text{delivered}}/(\delta\lambda/\lambda)_{\text{required}}]$ 'resolution factor' implies that if the source pulses are too short, we will have to work with excessive resolution, which means a reduction of the beam intensity compared to the ideal case. Indeed, by the Liouville theorem, the incoming neutron flux on the sample (detector) scales as $\Phi(\lambda).\delta\lambda_{\text{delivered}}$ where the time dependence is implicit via λ. The significance of this factor can be best illustrated for small angle neutron scattering (SANS) instruments, for which about $\delta\lambda/\lambda = 15\%$ is required and used on continuous sources, where the beam monochromatization is a truly free parameter. At a short pulse spallation source with typically 250–300 μs neutron pulse width on the most intense (so-called coupled) cold moderators and 20–60 ms neutron travel time from source to detector, the delivered $\delta\lambda/\lambda$ will be some 0.4–1.2%, i.e. compared to a continuous source the delivered beam intensity will be a 12–35 times smaller fraction of the peak source brightness. For a 2 ms pulse length long pulse source this relative intensity loss compared to the peak brightness will be reduced to a factor 2–5. For other types of neutron scattering experiments, better wavelength resolution is required, so the 'resolution factor' will be closer to unity, its maximum value of 1 reached when the delivered resolution is fully needed.

The role of the pulse shape factor $P(\lambda)$ can be explained as follows. In the case of SANS just considered, when the inherent wavelength resolution provided by the source pulse duration is too good, it matters little for data collection efficiency what is the neutron pulse shape like. Instead, what only matters is the total number of neutrons per pulse and the pulse shape factor will be identically $P = 1$. However, if the 'resolution factor' is 1, i.e. the resolution provided by the source pulse length is a main factor in determining the global resolution in the experiment, it is much more advantageous to work with a sharp compact line shape, such as a Gaussian, than with an exponential or a Lorentzian displaying long tails. In these conditions the effective peak intensity characterizing the information content of the experimental data can be reasonably defined as the ratio of the integrated beam intensity per pulse to the root-mean-square (RMS) FWHM (as opposed to the actual FWHM). Stating this in other terms, we will obtain similar resolution quality of data for different incoming neutron pulse shapes with equal RMS FWHM. For sharper line shapes with lesser tails, equal RMS FWHM corresponds to larger actual FWHM, i.e. more neutrons per pulse for equal peak height. Numerical analysis gives the values shown in Table 2 for the so defined effective pulse peak brightness compared to the actual peak brightness.

When highest resolution is required, for the chopper shaped triangular LP pulses, or SP pulses in the case of time-of-flight spectroscopy (where pulse shaping choppers can be fully effectively used, cf. Fig. 3), the pulse shape factor P can be taken as unity with respect to the Gaussian reference, otherwise for SP sources we have a correction factor of $P \sim 0.6$. This lower P factor at SP sources is due for all neutron wavelengths to slowly (exponentially) decaying trailing edge of the source pulses after a much sharper rising edge [5].

Table 2
The ratio of the effective pulse peak values (viz. integrated pulse intensity/RMS FWHM) to the actual pulse peak height for different pulse shapes, including short pulse (SP) and long pulse (LP) spallation sources

SP source pulse (all moderators)	LP source pulse (duration $\geqslant$1 ms)	Triangular	Trapezoidal: 2:1 base to top ratio	Gaussian
0.7–0.75	$\geqslant$ 1.18	1.22	1.64	1.25

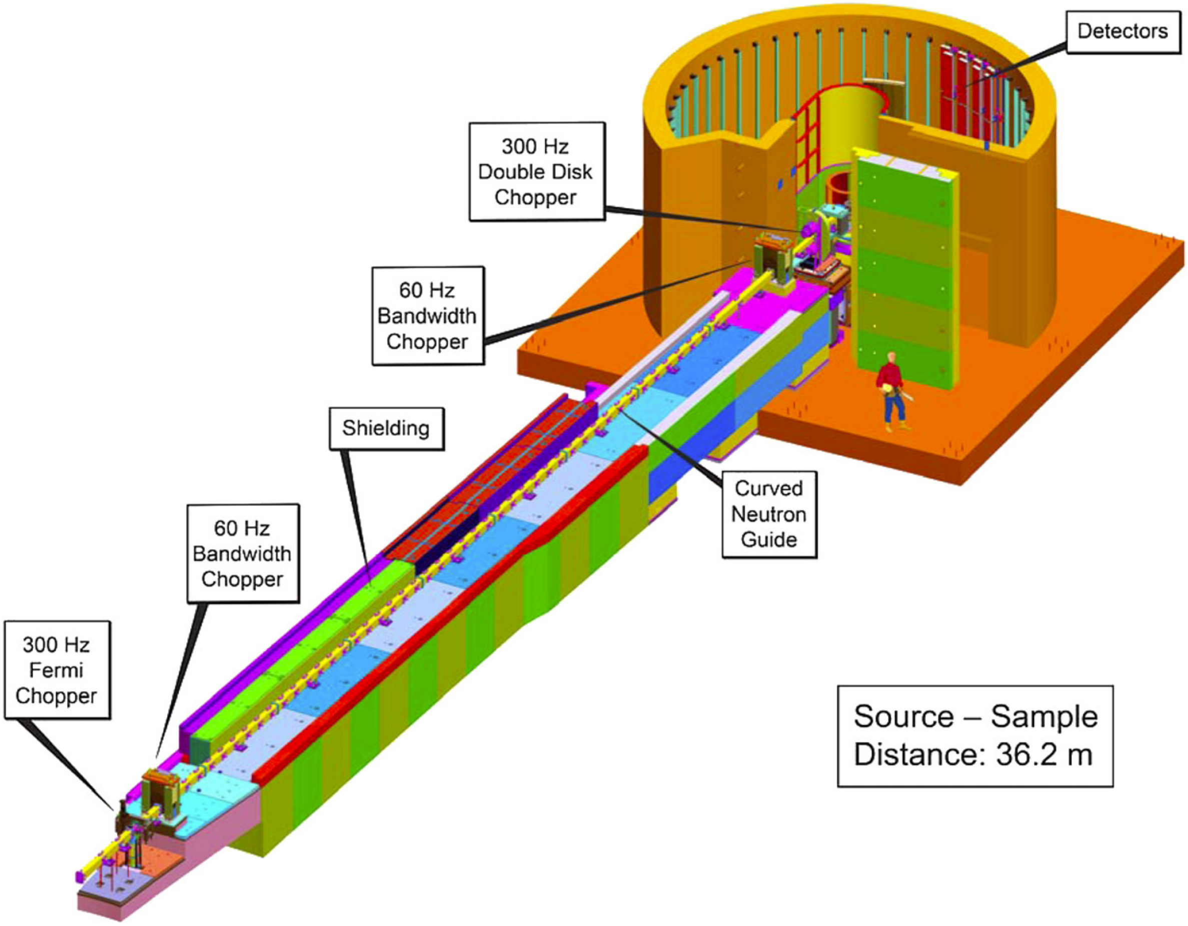

Fig. 3. The cold neutron chopper spectrometer CNCS under construction at SNS. The incoming beam monochromatization is generally determined by the source pulse length (typically 150–250 µs in the cold neutron range for the high intensity moderator used). The Fermi chopper on the left is optional; it serves to shape the source pulse by cutting it shorter if higher resolution is needed. The pulses on the sample are shaped by the 300 Hz high speed counter-rotating pair of choppers in front of the sample (typical pulse length 25 µs) and the scattered beam velocity is analyzed by measuring the time needed for scattered neutrons to travel from the sample to the position sensitive detectors at 3.5 m distance. The high chopper speeds are needed for resolution; the actual pulse repetition rate is determined by the slower bandwidth choppers, which cut out most pulses of the fast disc choppers. The total detector area will ultimately reach 40 m^2 (Courtesy of G. Ehlers, SNS).

With this definition of FOM the expected source performance of ESS (long pulse source of 2 ms pulses with 5 MW power at 16.67 Hz pulse repetition rate, 300 kJ energy per pulse) [5] is shown in Fig. 4, in comparison to the full, 1.4 MW power, 60 Hz short pulse SNS (23 kJ/pulse) [10] and the ILL 58 MW reactor source [11]. The J-PARC short pulse facility under construction in Japan will show similar characteristics to SNS, with its initial 20 kJ/pulse power (later to be enhanced to 40 kJ/pulse) at 25 Hz repetition rate [12]. The blue area of Fig. 4 corresponds to the range of FOM covered by SNS in different kinds of neutron scattering experiments, from the low resolution cases on the bottom of the area to high resolution time-of-flight inelastic spectroscopy at the top, while the upper limit in diffraction work is given by the dotted lines. The green area stands for the FOM range for ESS in the various types of experiments. The

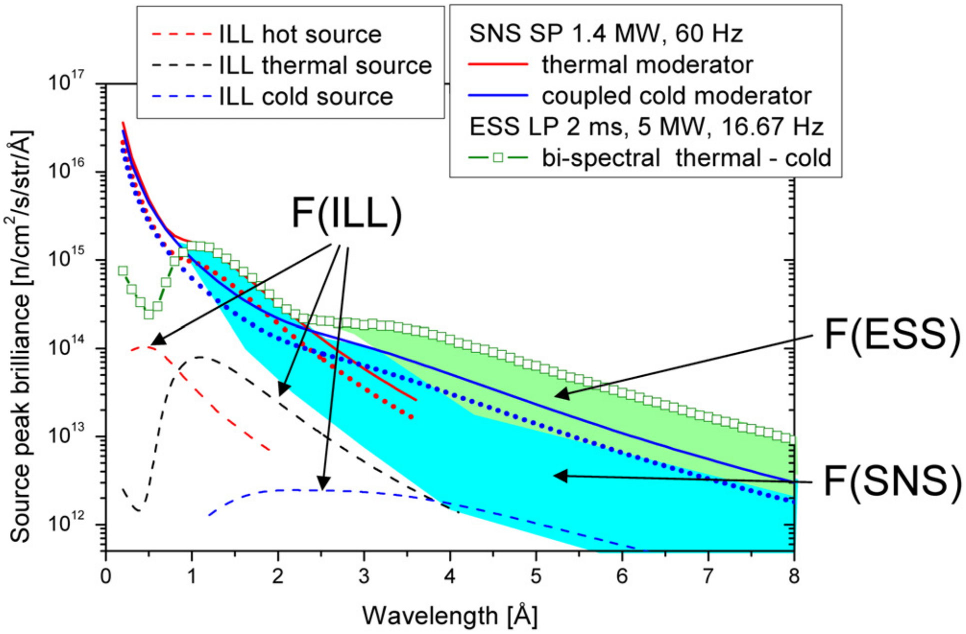

Fig. 4. Figure-of-merit F for various neutron sources for the typical variety of neutron scattering experiments using more or less monochromatic neutron beams of thermal and cold neutrons. For hot neutrons ($\lambda < 1$ Å) it was assumed that the wavelength resolution delivered by the source pulse length is never too good, which is an oversimplification. The dotted lines represent upper limits for F in diffraction type experiments on SNS moderators represented by the continuous lines of the same color. More details are given in the text.

two high power spallation sources SNS and ESS behave fairly complementarily, with ESS providing a much higher intensity for cold neutrons $\lambda > \sim 3$ Å, most relevant for the growing research fields of nanoscience and life sciences. For hot neutrons below 0.8 Å wavelengths the SNS SP source has a clearly superior FOM. In this range only high resolution applications are shown in the figure, while in many cases pulsed spallation sources will have a hard time to compete with the high average flux of the hot moderators at reactor sources (e.g. in single crystal diffraction work). Experiments with hot and epithermal neutrons (i.e. $\lambda < 0.8$ Å) represent an important, but in their share a less than 10% fraction of neutron scattering work. Both of these pulsed spallation sources are poised to provide substantial gains in source performance, compared to the most powerful fission reactor sources, with the exception of a few applications (e.g. in SANS at SNS, which is indicated by the bottom of the blue area at larger neutron wavelengths or in the hot neutron range in general for ESS).

One has to stress that in the spirit of the remarks on beam collimation above, in connection with Fig. 1, our basic analysis of the relative merits of top of the line continuous and pulsed sources of different types focuses on the general impact of source time structure and the related issues of beam monochromatization. More special secondary effects, although they can be quite substantial, have not been considered. As discussed above in connection with the more efficient beam focusing capabilities offered by arrays of monochromator crystals best adapted to continuous sources vs. the larger detector solid angles made accessible by the TOF technique best adapted to pulsed sources, many of these special, incidental conditions work in opposite directions. They undoubtedly complicate the details of the picture, but, on the whole, they do not change the general trends.

There remains, however, an important difference between monochromatic beam and inherently multiwavelength TOF techniques in the exploration of single crystal samples. The use of crystal monochromators and eventually crystal analyzers allows us to focus the whole data collection in the course of an experiment to a single point of interest in reciprocal space. In contrast, information in TOF experiments is inevitably spread over the reciprocal space, and only a small fraction of data contains relevant information if only a single point in this space is to be explored. Thus the most efficient way to perform such experiments will be the use of crystal monochromator instruments also on pulsed sources, i.e. similar ones to continuous source machines, which will then just make use of the time average brightness of the source. ESS with its time average flux comparable to ILL will still do pretty well and its pulsed nature will still provide fringe benefits e.g. for the background or sorting out higher order reflections on crystals. However, these benefits will not be comparable to the gains in FOM shown in Fig. 4 for the typical cases. A similar situation will occur in inelastic neutron spectroscopy when using RRM at short neutron wavelength. The different incoming neutron velocities in the different RRM pulses on the sample will probe quite different Q and E domains, and in many cases only a fraction of this information will be actually useful data for a particular experiment. At the same time the number of real cases in single crystal studies is found to be smaller and smaller, in which only a very small fraction of the reciprocal space is of any interest and a survey of a broader domain is of no added value. Thus, these special cases will not substantially modify the fundamental trends expressed in Fig. 4, due to their moderate weight in the bulk of neutron scattering research.

4. Conclusions

In the past 50 years progress in neutron scattering research was achieved by improving neutron instrumentation techniques, while the brilliance of neutron sources could be only moderately enhanced, basically limited in fission reactors by the heat production associated with the production of free neutrons. The spallation method, gradually established in the past 3 decades, offers two decisive advantages to improve the efficiency of the process: nearly an order of magnitude less heat deposition per fast neutron produced and a much higher fraction of these neutrons that can reach the sample in appropriately pulsed source operation. These features now allow us to envisage quantum leaps in neutron source performance, eventually approaching overall 3 orders of magnitude in many cases, which will make a huge difference in the inherently intensity limited neutron scattering work. Within the next decade the most powerful pulsed spallation sources under construction, or planned, will deliver up to 2 orders of magnitude gains in data collection rates compared to what exists today, and most of this progress will come from a leap in source performance of unprecedented proportions since the 1950s. The largest step forward will be provided by ESS in the cold neutron regime. In the longer term, the ultimate technical limits of the pulsed spallation approach may be higher by about another order magnitude. Other possible methods of extracting free neutrons from various nuclei (photoneutrons, p–n reactions, fusion) do not match the unparalleled energy efficiency of the pulsed spallation technique for producing the highest possible slow neutron beam intensities for neutron scattering research in condensed matter. However, they might provide valuable new alternatives for providing neutrons for other, less intensity dependent fields of neutron research.

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