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Very cold neutrons in condensed matter research

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Abstract. In high resolution neutron scattering experimental work the use of significantly longer incoming neutron wavelengths compared to the currently widely used cold neutron range can be of significant advantage. Such advantages are to obtain higher data rates at equal resolution conditions, for example in small angle neutron scattering, and to obtain far better resolution, e.g., in neutron spin echo and time-of-flight spectroscopy or both.

Keywords: Neutron scattering, very cold neutrons, small angle neutron scattering, neutron time-of-flight spectroscopy, neutron spin echo

1. Introduction

In condensed matter research, neutron beam investigations primarily concern neutron scattering, where the energy and momentum change of the neutron in the scattering event is investigated. In general, small momentum and energy changes can be best resolved and analyzed with high resolution by using neutrons of lowest possible initial energy and momentum. Very Cold Neutrons (VCN) represent opportunities in high-sensitivity and/or high-resolution neutron scattering work compared to the today commonly used cold neutron energy range. These opportunities are analyzed in this note for three seminal cases, small angle neutron scattering (SANS) for elastic scattering, and time-of-flight spectroscopy (TOF) and neutron spin echo (NSE) for inelastic scattering.

2. Elastic scattering: Example of small angle neutron scattering

In SANS the goal of observing small neutron momentum changes in the neutron scattering process corresponds to recording small changes of neutron flight direction. For this purpose, the incoming neutron beam needs to be very well collimated in both directions perpendicular to the incoming beam, and the required beam divergence is inversely proportional to the incoming neutron wavelength λ, due to the definition of neutron momentum, $k = 2\pi/\lambda$. Thus, a longer-wavelength beam requires less stringent beam collimation. The angular resolution of the neutron flight direction change measured by an area detector requires limitation of the illuminated sample cross section perpendicular to the incoming neutron beam direction. Finally, a reasonable relative monochromaticity $\delta\lambda/\lambda$ of the incoming beam also allows for a larger used wavelength band $\delta\lambda$ for an incoming beam with larger λ. All these factors together imply that the fraction of the incoming beam that must be selected for a given δk resolution in a SANS experiment will be proportional to λ^5, e.g., for 2 times larger wavelength, a 32 times larger fraction of the generated neutron source spectrum will be utilized, via less stringent beam collimation, sample area and beam monochromaticity. In the long-wavelength tail of a Maxwellian neutron spectrum the beam intensity asymptotically drops as λ^{-5}. Thus, in the asymptotic long-wavelength spectrum, the neutron economy is independent of the choice of the mean wavelength λ selected for a SANS experiment.

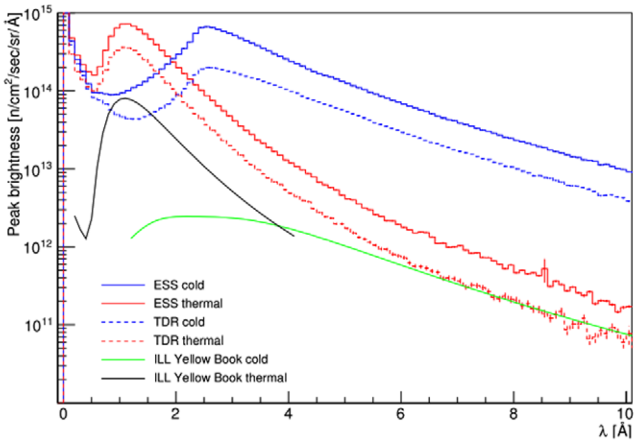

Fig. 1. Simulated neutron moderator spectra for ESS, after Ref. [8], vs measured spectra for ILL [4]. “ESS cold” and “ESS thermal” are the cold and thermal brightness spectra, respectively, from the latest ESS moderator design [8]. “TDR” spectra are from the earlier design [5]. The ESS and TDR thermal spectra correspond reasonably well to the Maxwellian λ^{-5} large-wavelength asymptotic tail above 4 Å, while the cold spectra show a weaker slope.

It is generally assumed based on practical experience that this asymptotic range of cold neutron moderator spectra is achieved for $\lambda > 6$–8 Å. VCN moderators, in order to be advantageous in SANS type of experiments, must therefore provide high intensity at wavelengths $\lambda > 10$ Å, that is above the presumed λ^{-5} dependence of the spectra of current cold moderators (which happens to be only well established in practice for neutron wavelengths below 10–20 Å).

More careful theoretical simulation studies, however, reveal that the large-wavelength asymptotic tails of cold neutron moderator spectra can drop less rapidly than the λ^{-5} approximation, as illustrated by the results of Zanini et al. [8] in Fig. 1. The ESS cold neutron spectrum corresponds to a configuration of parahydrogen moderators, as developed in Ref. [6]. In contrast to the thermal moderators, the ESS cold moderator spectra show indeed a slower falling long-wavelength tail than the canonically assumed λ^{-5}. Thus, by extrapolation, the incoming beam intensity gain in a SANS type experiment at equal resolution could even be an order of magnitude more in the VCN wavelength range around 40 Å than for cold neutrons in the common range around and below 10 Å. Different, innovative, more sophisticated moderator designs might eventually even offer larger favorable deviation from the λ^{-5} dependence. For practical use, one also has to consider issues due to large wavelengths, including higher absorption in samples and beam windows, and a larger effect of gravity on the beam trajectories.

In order to get a practical impression, let us consider the use of 40 Å neutrons from the ILL cold neutron sources under the hypothetical assumption, that the Maxwellian λ^{-5} dependence would apply for the VCN wavelength range. A workhorse conventional SANS machine could consist of a source diaphragm of $2 \times 2\ \mathrm{cm}^2$ at a distance of 20 m upstream from the $1 \times 1\ \mathrm{cm}^2$ sample area, followed by a 2D multidetector of $1\ \mathrm{m}^2$ area at 20 m distance downstream of the sample. The momentum transfer resolution of this configuration would be $\delta k \approx 0.001\ \text{Å}^{-1}$ at 6 Å neutron wavelength. $\delta\lambda/\lambda \approx 15\%$ beam monochromatization is reasonable to assume, typically produced by a velocity selector upstream from the instrument. In view of the ILL cold source brightness shown in Fig. 1, one would have a monochromatic, well collimated incoming beam flux on the sample of $\phi(\lambda)\delta\lambda\delta\Omega \approx 6 \times 10^5$ neutrons/cm^2/s, where $\phi(\lambda)$ is the moderator brightness at $\lambda \approx 6$ Å, c.f. Figure 1, and the impinging beam divergence $\delta\Omega$ at any point of the sample area is 10^{-6} sr. If we just move with the 15% monochromatic beam to the assumed moderator spectral tail at $\lambda \approx 40$ Å, the λ^{-5} dependence would lead to an expected beam intensity on the sample of $\approx$300 neutrons/cm^2/s only, but at a significantly superior momentum transfer resolution of $\delta k \approx 0.00015\ \text{Å}^{-1}$. Part of the price of this important resolution gain would also be significant gravity drop curvature of about 20 cm of the neutron trajectories over 20 m flightpath. Ways of handling such a situation have been analyzed in Ref. [2].

The other sample configuration worth to be considered at the very cold neutron wavelength range of 40 Å is the one with the same δk resolution and same observed momentum transfer range as we just had for 6 Å wavelength. This can be simply achieved by a feature common to many modern SANS machines: flexible variation of the distances between source diaphragm and sample, as well as between sample and detector. This corresponds here to changing both of these distances to 3 m, instead of 20 m at $\lambda \approx 6$ Å. In this case, monochromatic neutron flux on the sample would be $\approx 1.35 \times 10^4$ neutrons/cm^2/s and the data collection rate would be the same for samples with little neutron absorption as at 6 Å, since the same 1 m^2 multidetector will at 3 m distance cover $(20/3)^2 \approx 44$ times larger solid angle. Absorption of eventual beam windows (only the detector front window is not avoidable) are manageable by using, e.g., aluminum alloys with about 3% absorption per mm thickness at 40 Å wavelength. For such large wavelengths, the gravitational curvature of the neutron trajectories over distances of about 3 m is only a few mm, which is not a serious issue for SANS work with 1 cm^2 sample area. Development of a VCN moderator with brightness significantly above the theoretical λ^{-5} long-wavelength tail of the common cold moderators would indeed be an important step forward in the enhancement of practically precious beam intensities.

3. Inelastic scattering: Example of time-of-flight spectroscopy

In the investigation of very small neutron energy changes in the scattering process, the use of a larger incoming neutron wavelength is even more a significant advantage than in elastic scattering, as discussed above. In view of the relation of neutron energy to the neutron wavelength $E \propto 1/\lambda^2$, the change of neutron energy is related to the initial neutron wavelength as $\delta E = \delta\lambda \mathrm{d}E/\mathrm{d}\lambda \propto \delta\lambda\,(-2/\lambda^3)$. Using the measured neutron time-of-flight $\propto \lambda$ over a known distance for the determination of the neutron energy change, we conclude that for a given time resolution δt the corresponding neutron energy change will scale as

$$\delta E \propto \delta t/\lambda^3. \tag{1}$$

This is a much more significant variation than for the neutron momentum $\delta k \propto \delta t/\lambda$. Very similarly as we had above for the two aspects of incoming beam collimation and scattering angle measurement resolution in SANS, here we have the incoming beam monochromaticity (roughly determined by the pulse length of the first, beam monochromatizing chopper) and time-of-flight resolution for the scattered beam wavelength determination, which latter is roughly given by the neutron pulse length at the sample. Thus, these two together mean that the selected fraction from the neutron source spectrum at equal resolution for the neutron energy change in the scattering process scales as λ^6 for each neutron pulse to the sample. The acceptable frequency of incoming neutron pulses in equal spectrometer configuration is inversely proportional to the neutron velocity, since the time between pulses in the general case must be somewhat (about 50%) longer than the flight-time of the elastically scattered neutrons from the sample to the detector, in order to sufficiently reduce confusing mixing of neutrons at the detector, which come from subsequent neutron pulses on the sample. Thus, we end up with the same λ^5 intensity scaling factor as for SANS, i.e., wavelength independent flux on the sample for equal energy resolution in the asymptotic λ^{-5} long-wavelength tail of a Maxwellian source neutron spectrum.

However, in this case, there is a potential additional benefit of using larger wavelengths: in the same spectrometer configuration and energy transfer resolution at longer wavelengths, the momentum transfer resolution will be better as $\delta k \propto 1/\lambda$. As we have seen for SANS, if the improved momentum resolution is needed, this would result in an additional gain with neutron wavelength proportional to λ^4, corresponding to larger acceptable incoming beam collimation and beam cross section on the sample. Of course, at the same time the accessible momentum transfer k range decreases with increasing wavelength, so the required momentum transfer range sets an upper limit to the possible incoming neutron wavelength.

In sum, for TOF spectroscopy, the efficiency of making use of the available intensity of the source neutron spectrum depends on the incoming neutron wavelength:

a) as λ^{10} at equal energy and momentum transfer resolution,

b) as λ^5 at equal energy transfer resolution at a constant spectrometer geometry and results in a parasitic improvement of the momentum transfer resolution as: $\delta k \propto 1/\lambda$,
c) the choice of wavelength must stay below a maximum value depending on the required momentum transfer range to be studied in an experiment, which for quasi elastic scattering practically means incoming neutron wavelength $\lambda < 4\pi/k_{\max}$ by a margin of at least 5%.

4. Inelastic scattering: Example of neutron spin echo spectroscopy

Fundamentally, the case of NSE is quite different, but it also relates to what has been said above about SANS and TOF, with an additional twist. NSE is a unique TOF method, where the time-of-flight of each individual neutron is measured and kept record of by the Larmor precession of its spin, so one does not have to worry about finely monochromatizing and pulsing the neutron beams for good resolution. It is common practice in NSE to use a 10% monochromatic incoming beam for better than 0.001% resolution in wavelength change in scattering. Most NSE experiments track processes on larger than atomic length scale, i.e., at small momentum transfer, somewhat similarly to SANS, since we do not have to worry about pulses or fine monochromaticity.

However, in contrast to SANS, in practice – due to the fine tuning of Larmor precession field shapes and homogeneity across the beam – we cannot systematically relax the built-in incoming beam collimation angles and sample size when changing the wavelength. At shorter incoming wavelengths the required momentum transfer resolution might require stepping down divergence and beam cross section by diaphragms, but this will have moderate effect on the NSE energy resolution. Thus, this leaves us in NSE with the basic situation that the key parameter, the energy transfer resolution scales as $\delta E \propto \lambda^{-3}$, and the momentum transfer resolution as $\delta k \propto \lambda^{-1}$, but going to longer wavelengths in general does not improve the efficiency of use of the beam intensity provided by the neutron source. The reason to choose longer wavelengths is to improve the available energy resolution δE, which is about 0.75 neV (that corresponds to 900 ns NSE time) at the currently highest resolution NSE spectrometer worldwide, the upgraded IN15 at ILL. This is illustrated in Fig. 2 [3]. Thus, here the reason to use longer wavelengths is simply to achieve unprecedented energy resolution in neutron scattering research, under the assumption, that we cannot further improve the instrumental performance at a given wavelength. This assumption is pretty well justified, although rare miracles do happen from time to time.

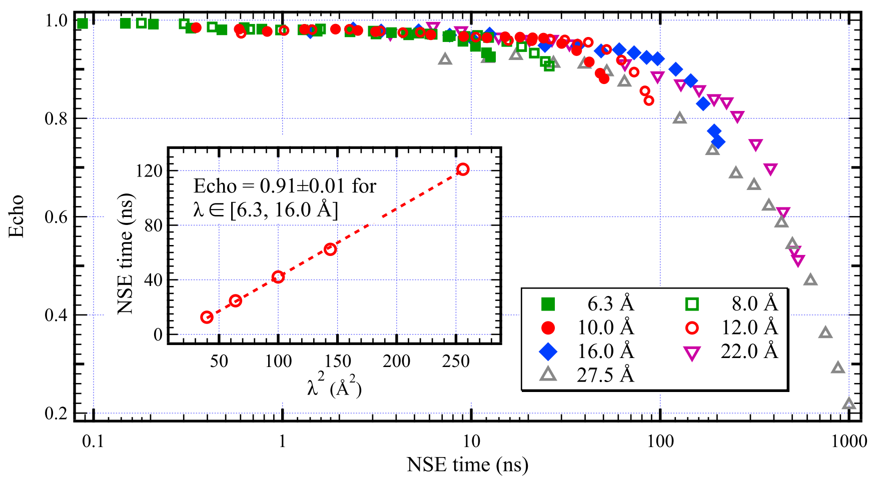

Fig. 2. Measured NSE signal at different incoming neutron wavelengths for a predominantly elastically scattering resolution calibration sample [3] at the IN15 spectrometer at ILL. From 6.3 to 16 Å, the NSE times where the NSE signal drops from its saturation value at short times to 0.9 is proportional to λ^2. At 22 and 27.5 Å, the neutron pathlength inhomogeneities strongly increased compared to shorter wavelengths.

As a matter of fact, the ultimate resolution of an NSE instrument depends on the value (i.e., the line integral along the axis of the precession field coils) and the homogeneity of the effective pathlength of the neutrons across all possible beam trajectories. Effective pathlength means the path integral along the neutron trajectory weighted by the varying magnetic field along the neutron path. The sensitivity to any pathlength differences increases with the time taken on average by the neutrons to travel across the spectrometer, i.e., proportionally to wavelength. Therefore, the final possible resolution of an NSE instrument is determined by these effective neutron pathlength inhomogeneities, and the ultimate resolution of a given instrumental configuration (independently of the strength of the precession fields) scales as $\delta t_{\mathrm{rms}} \propto \lambda$, where δt_{rms} is the scatter of the neutron transition time through the instrument due to differences in the effective neutron pathlengths. This way, the practical energy resolution limit δE_{res}, set by the effective neutron pathlength inhomogeneities across the various neutron trajectories, scales in view of Eq. (1) as

$$\delta E_{\mathrm{res}} \propto \delta t_{\mathrm{rms}}/\lambda^3 \propto 1/\lambda^2. \tag{2}$$

This scaling is perfectly borne out in Fig. 2. If we observe, e.g., the NSE times at the points where the NSE signal measured on a fully elastically scattering calibration sample (producing an inherently time independent NSE signal) drops from its saturation value at short times to 0.9, these convincingly bear out the expected proportionality to $1/\lambda^2$ from 6.3 to 16 Å wavelength, Eq. (2). The points at 22 and 27.5 Å deviate, which indicates that at these wavelengths the neutron pathlength inhomogeneities strongly increased compared to those at lower wavelengths.

The homogeneity of the effective neutron pathlengths is the key ultimate quality of a high resolution NSE machine. The data shown in Fig. 2 [3] suggest that the rms scatter in the effective neutron pathlength along the neutron trajectories is about 0.003 mm rms for all wavelength below 22 Å. This is a very impressive number, which is achieved by very careful mechanical design and a host of intricate correction coils to the magnetic field along the spectrometer. Figure 2 indicates a clear trend of increase of the inhomogeneity of the effective neutron pathlengths above 20 Å wavelengths.

The effect of gravity could be one reason. Indeed, over the length of a spectrometer arm the curvature of the neutron paths under gravity leads to about 2 mm drop at 22 Å and about 3 mm at 27.5 Å. Such drops are indeed negligible for the physical lengths of the quasi-horizontal neutron flightpaths, but they could be substantial for the length of the effective flightpaths by going with significant curvature across the magnetic field configuration carefully homogenized for much more straight flightpaths below 10 Å wavelengths. The gravitational drop is inversely proportional to the square of the initial, quasi horizontal neutron velocity.

5. Conclusions

We have considered a few examples where Very Cold Neutrons (VCN) with wavelength of 30–50 Å could provide for substantial gains in the range of an order of magnitude or more compared to the use of today's cold neutron beams in high-resolution neutron scattering studies.

In Small Angle Neutron Scattering, moderators with a long-wavelength tail in the emitted neutron spectrum could provide substantially increased beam intensities for high-resolution experiments, if their intensity at VCN wavelengths is significantly above the hypothetical extrapolation of existing cold neutron spectra to longer wavelengths by assuming λ^{-5} intensity scaling. As mentioned above, higher neutron absorption in the samples and beam window materials at longer neutron wavelength might void the advantage of higher numbers of neutrons being initially generated for the experiment. Working with hexapole magnetic field lenses for focusing the neutron beam has been experimentally shown to provide an additional important advantage in SANS work for long neutron wavelengths >40 Å [1,7]. Namely, effective optical focusing of neutrons without any material in the beam is impractical for much shorter neutron wavelengths.

Potential intensity gains using VCN can also apply for Time-of-Flight spectroscopy, with due caveats for higher absorption, if only the energy resolution is considered. However, the gains can be orders of magnitude higher, if combined high energy and high momentum transfer resolutions are of interest. In Neutron Spin Echo spectroscopy, in contrast, decisive and exclusive gains in energy resolution scaling with λ^{-3} could be obtained using VCN, as long as the increasing absorption losses in materials crossed by the beam (flippers and magnetic field correction coils) do not become prohibitively strong. Here we assume that the homogeneity of the effective neutron pathlengths can be maintained for longer wavelengths, where, e.g., gravity seriously influences the actual neutron trajectories across the instrument. Achieving unprecedentedly high energy resolution by using VCN also applies in similar manner to TOF spectroscopy, where caps of chopper speeds set limits to the energy resolution and leave taking advantage of the kinematical λ^{-3} scaling as the one feasible approach.

In sum, on the one hand, there are potentials of favorable cases of achieving higher neutron intensities using efficiently moderated VCN in both elastic and inelastic high-resolution neutron scattering experiments within a limited energy and momentum transfer range. In addition, on the other hand, in inelastic neutron scattering experiments the ultimate limits of highest achievable resolutions could be substantially improved using VCN, as long as practical neutron beam intensities become available, also for compensating the increased beam absorption losses. Both of these opportunities provide strong interest in future availability of improved intensity VCN moderators, either by cold moderator configurations with slower intensity decay with increasing wavelength, or by lower effective temperature of the moderating material.

References

[1] M. Bleuel et al., A small angle neutron scattering (SANS) experiment using very cold neutrons (VCN), *Physica B* **404** (2009), 2629–2632. doi:10.1016/j.physb.2009.06.048.

[2] C. Cubitt, R. Schweins and P. Lindner, Countering the effects of gravity on a small angle neutron scattering instrument, *Nuclear Instr. and Methods in Phys. Res. A* **665** (2011), 7–10. doi:10.1016/j.nima.2011.11.034.

[3] B. Faragó, private communication.

[4] Institut Laue-Langevin, *ILL Yellow Book 2008*, 2008.

[5] S. Peggs (ed.), ESS Technical Design Report, 2013. ISBN 978-91-980173-2-8.

[6] T. Schönfeldt, K. Batkov, E. Klinkby, B. Lauritzen, F. Mezei, A. Takibayev, P. Willendrup and L. Zanini, A model for non-thermalized neutron spectra emitted from para-hydrogen, in: *Proceedings of the 21st Meeting of the International Collaboration on Advanced Neutron Sources (ICANS XXI)*, 2016, p. 134.

[7] M. Yamada et al., A compact TOF-SANS using focusing lens and very cold neutrons, *Physica B* **406** (2011), 2453–2457. doi:10.1016/j.physb.2010.12.002.

[8] L. Zanini, K.H. Andersen, K. Batkov, E.B. Klinkby, F. Mezei, T. Schönfeldt and A. Takibayev, Design of the cold and thermal neutron moderators for the European Spallation Source, *Nuclear Instr. and Methods in Phys. Res. A* **925** (2019), 33–52.