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To cite this article: W Schweika et al 2016 J. Phys.: Conf. Ser. 746 012013

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DREAM – a versatile powder diffractometer at the ESS

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Abstract. The instrument DREAM, in construction at the long pulse European Spallation Source (ESS), is a new type of neutron time-of-flight powder diffractometer, which utilizes additional choppers to meet the typical high resolution requests. Pulses will be of symmetric shape and their width can be varied from 10\,\mu s to 1 ms, providing an unprecedented flexibility from highest to low resolution with optimized intensities at the superior brightness of the 5 MW source. The design is driven particularly by the needs and challenges for small and complex samples, large unit cell materials, thermoelectric cage structures or metal-organic framework structures, multiphase battery materials and complex magnetic structures. Therefore, the chosen wavelength bandwidth of 3.7\,\AA\,may cover well the peak intensities of the thermal and cold moderator used simultaneously and provides a sufficient $Q$ and $d$ range for obtaining diffraction patterns in a single setting. VITESS simulations show a performance that is about two orders of magnitude higher than current best instruments.

1. Introduction

Time-of-flight (TOF) neutron powder diffractometers benefit from the brightness of pulsed neutron sources and have gained increasing importance over the past decades with respect to monochromatic instruments [1] at continuous sources. Short-pulse spallation sources (SPSS) are well suited for powder diffraction offering the brilliance from the sharp neutron pulses extending to shortest wavelengths, with new applications in measuring the pair distribution functions from total scattering with high spatial resolution [2]. In contrast to monochromatic instruments at reactors, which measure angle dispersive like x-ray diffractometers, in neutron TOF diffraction another dimension is offered by measuring additionally energy-dispersive, that has immediately found new applications for special sample environments, by confining the gauge volume by diffraction angles near 90 degree [3], and for high resolution studies in backscattering essentially determined by the ratio $\tau/T$ of pulse width to neutron TOF to the detector. The time structure of SPSS moderators yields a wavelength dependent pulse length and keeps the relative time and wavelength resolution approximately constant over a broad wavelength range. By adapting further the detector geometry, it is possible to achieve a constant relative resolution $\Delta d/d$, as exemplified by the design of the POWGEN instrument [4] at the Spallation Neutron Source (SNS) in Oak Ridge.
In contrast, the new European Spallation Source (ESS) will have a long neutron pulse essentially determined by the proton pulse duration on the target. The pulse length of 2.86 ms is apparently not of practical use for typical powder diffraction tasks, since already medium time resolution $\tau/T \approx \Delta d/d = 0.005$ requires an extreme flight path $L = v(\lambda)\tau(\Delta d/d)^{-1}$, e. g. more than 2 km for $\lambda = 1$ Å neutrons. However, the neutron pulse of the 5 MW ESS will also be the most intense pulse with a superior peak brightness. Therefore, generating shorter pulses will still be gainful. It will create new opportunities and particularly a valuable new versatility in neutron powder diffraction.

The optimization strategies and instrument design at long pulse sources need to be novel and have led to the here presented new type of a neutron TOF powder diffractometer DREAM, which is highly flexible in resolution and versatile for applications. The obvious tools to produce shorter pulses of thermal and cold neutrons are fast, counter-rotating disc choppers producing a symmetric pulse shape, which is preferable in view of data refinement. Pulses can be shaped with high flexibility by varying the choppers speed, changing the time resolution over three orders of magnitude from 10 $\mu$s to ms-range, and trading resolution for intensity easily. This virtue of flexible resolution is illustrated in Figure 1. It shows a large accessible region, which will even extend current limitations of high-resolution neutron powder diffraction.

The pulse $\tau$ is simply chosen independent of the neutron wavelength and fixes the minimal absolute resolution $\Delta d$ in backscattering. With acceptable increase in chopper complexity [5] it would be possible to change to a mode of constant relative resolution $\Delta \lambda/\lambda$, which however implies a limitation on the achievable best resolution. Instead of aiming at a uniform relative $\Delta d/d$ resolution function for integrating the Debye-Scherrer lines to single peaks, the present concept rather intends to make best simultaneous use of high resolution backscattering and high intensity diffraction at lower angles. Therefore, we will imply an wavelength- and angle-dispersive 2D Rietveld refinement method [6], which has been developed for the TOF powder diffractometer POWTEX [8, 9] under construction at the research reactor FRM II in Munich. As soon as such constraints for a uniform resolution function can be overcome by an adapted 2D data refinement method, the instrument design and particularly, the detector and its geometry can be better optimized with respect to cost and efficiency and less complexity. Since this instrument is specifically designed to benefit from measuring and analyzing data versus wavelength and diffraction angle, we have given it the name DREAM, which stands for *Diffraction Resolved by Energy and Angle Measurements*. Beyond the instrument’s dedication for powder diffraction,
the name implies further capabilities in resolving the out-of-plane dependence of the scattering to study preferred orientation, texture and finally single crystal diffraction in neutron TOF Laue technique. Here, the instrument benefits from the covered large solid angle of detection including 2D position sensitivity, the flexibility in time-resolution, and the optimization for small samples.

Below, we present further design considerations of the instrument, the proposed instrument layout, and finally VITESS [11, 12] simulations of its performance, which is compared to existing world class neutron TOF powder diffraction instruments.

2. Design considerations and layout of the instrument
An overview on the scheme of the instrument and its components is given in Fig. 2.

2.1. Source parameters and bandwidth
The ESS parameters have been defined in the Technical Design Report [7] with a source frequency of 14 Hz, which in reverse gives a TOF frame of \( T_s = 1/14 \text{Hz} = 71.4 \text{ms} \). The duration of the proton pulse on the target is \( \tau_s = T_s/25 = 2.86 \text{ms} \) and large compared to moderation time. Target and moderator are surrounded by the monolith shielding and shutter extending 6 m from the center. A Pulse Shaping Chopper (PSC) to be placed outside the monolith at \( L_{PSC} > 6 \text{m} \) defines the instrument length \( L \) from the moderator to the detector position, \( L = L_{PSC}(T_s/n\tau_s + 1) \) and the wavelength frame \( \Delta \lambda \leq 1.88 \text{Å} \) by \( L_{PSC}/\tau \). Here \( n \) is the integer number of wavelength frames \( \Delta \lambda \) used in a single TOF frame of the source, \( T_s \), and \( n\Delta \lambda \) gives the total bandwidth.

Looking at choices for current instrumentation, a bandwidth of 1 Å as chosen for POWGEN is usually not sufficient to complete diffraction studies and most users request two or more settings for data refinements. The SHRPD [13] at JPARC operates most of its time at 5 Hz, id est with 80% pulse suppression to achieve a desirable larger bandwidth of 8 Å instead of 2 Å in normal 25 Hz source mode. The instrument WISH [14] at ISIS target station 2 with 10 Hz has a bandwidth of 9.4 Å. In principle, it is possible to cover a larger wavelength band in sequential steps with the same efficiency, while a narrow bandwidth might advantageously focus to special diffraction peaks of interest. On the other hand a sufficiently large bandwidth is important for real time experiments on irreversible processes, such as studies of chemical reactions.

DREAM has been designed as a bi-spectral instrument using two wavelength frames, \( 2\Delta \lambda \approx 3.7 \text{Å} \), to cover simultaneously the peak spectra of both thermal and cold fluxes with a dynamic range that perfectly suits for diffraction experiments in a single setting. While pulse suppression like for SHRPD gives similar opportunities for a longer instruments, the choice to set the instrument length to approximately 75 m is motivated by the reduced cost for a shorter instrument and better neutron transport by the neutron optics when taking gravity into account.

![Figure 2. Scheme of the instrument.](image-url)
2.2. Neutron optic elements and guide system
The original design was based on the assumption that two moderators, thermal and cold, each with 12x12 cm$^2$ surface, were placed side by side. Shielding of the moderator monolith and shutters extends to 6 m from the source. This geometry comfortably allows for extracting a suitable divergence through a pinhole at 6 m, without neutron guides in the monolith. The desirable phase space density at the pinhole can be efficiently transported to the sample position. The simplest geometry would be two consecutive ellipses with a common focal point and pinhole and sample placed in the outer foci. The reason for having two but one ellipse follows from the phase space transformation by an ellipse, which is restored by the second ellipse [9]. The optimization supported by simulations led to two modifications. First, the central part of this guide system could have simpler straight shape, with even small gain in transport efficiency. Second, including gravity effects a larger vertical opening at the pinhole accepting a larger phase space is essentially able to retrieve the phase space density for a sample smaller than the pinhole. This design with a straight view from sample to moderator is superior particularly for short wavelength neutrons. The pinhole concept combined with the defocussing and focusing optics accept essentially the useful neutrons, which largely reduces the background.

Recent developments to the current concept of a so-called "butterfly" moderator design [10] have increased the extractable brightness by reducing the surface area to 3 cm height. Our investigations have shown that this increased brightness can be fully exploited by shifting the vertical ellipse focus from 6 m to the source using guide inserts starting 2 m from the source. With respect to the horizontal guide layout, the pinhole near 6 m can be kept, only an additional mirror inside the monolith compensates for the slightly reduced source size. The instrument axis points straight to the center of the thermal moderator. Neutrons from the cold source placed next to the thermal moderator are mirrored into the beam by a bi-spectral device [15, 16] that is inclined about 0.5 degree with respect to the thermal beam axis. We considered various options to inflect the cold beam into the neutron guide system, (i) a straight mirror 0.5 mm thin glass coated with a NiTi supermirror (ii) the same materials assembled to a shorter 25 cm long mirror stack with 1 mm air gaps, (iii) a solid-state bender [17] made of a 60 mm short stack of 150 μm thin Si-wafers with total reflection at the Si/NiTi interface, (iv) the same assembled to a straight mirror stack of only 16 mm length. Our idea of using the compact Si-wafers has the advantage that it allows to place the device for better access outside the monolith near to the horizontal focal point at 6 m, and it can reduce actually the path through solid materials compared to the cases (i) und (ii). In VITESS simulations the bender (iii) provides best performance for cold neutrons, the transmission of the thermal neutrons however, is affected by Laue extinction. A straight Si-stack (iv) will be about 15% less efficient in inflecting neutrons from the cold moderator, however, it largely avoids Laue extinction. Since such a device is only of 16 mm length, it is of least impact to the transmission of the straight thermal beam.

The neutron guide is straight for optimal transport of short wavelengths neutrons. In combination with T0 choppers and a heavy shutter, we expect a reduction of background, of instrument shielding and of costs in comparison to a curved guide option only. The guide cross section opens up to 6 cm x 6 cm as it results from the optimization with a figure of merit of intensity for a small sample cross section of 1 cm$^2$ within a divergence of ±0.25° (FWHM). Near the sample the focusing section can be extended by an exchangeable part, thus approaching 50 cm distance to the sample focusing with ±0.8° (base to base) can be achieved, which will be valuable for small samples, e.g., under high pressure. A set of slits in final 4 m guide part allows further control of the divergence (see WISH [14]).

2.3. Chopper system
The chopper system and chopper positions are shown in Fig. 1. The system consists of two fast, counter-rotating disc choppers to shape the pulse (PSC), a 14 Hz overlap chopper (OC)
to eliminate all higher harmonics from the PSC, two band control choppers (BC), and 28 Hz T0-choppers to suppress the prompt pulse. The interplay of these choppers is depicted in the acceptance diagram, see Fig. 3, showing neutron time versus wavelength, \( t \propto s\lambda \). In the given coordinates the slope is related to the distance \( s \) from the source with \( t = 0 \) at \( s = 0 \). [18] The figure shows a typical setting for \( 0.6 \text{Å} < \lambda < 4.2 \text{Å} \) at a medium time resolution of 150 μs. The PSC have a 75 cm diameter disc and may operate in multiples of the 14 Hz source frequency up to 294 Hz for highest resolution. There are different pairs of openings for each frequency to achieve the pulse shaping for the two wavelength bands. The BC provide a better definition of the wavelength band and further suppress the exponentially decaying "afterglow" background from the source for 6 ms. PSC, BC, and OC may shift in phase to choose the wavelength range. The T0-chopper concept is still under development. Current simulations of high energy neutrons favor a solution using essentially several W-absorbers, each approximately 5 cm thick.

2.4. Diffractometer and detector

The diffractometer has a detector with 6 sr solid angle coverage, including backward, forward and a 2 sr section to one side, within a cylindrical geometry along the beam axis. Sample to detector distance is \( \geq 1.25 \text{m} \), the cylinder radius. The geometry is favorably adapted to the Debye-Scherrer cones. The spatial resolution of 5 mm (FWHM) at \( 2\theta = 90^\circ \) in two dimensions provides a better angular resolution definition, \( \Delta\theta = 0.13^\circ \) (FWHM), than given by the incoming divergence, and is similar to contribution from the sample size. The concept includes two further detectors at 2.5 m distance in order to access higher angles in backscattering and smaller angles in forward direction side to extend the Q-range to the scale of nano-particles covering in total three orders of magnitude in length scale with \( 0.01 \text{Å}^{-1} < Q < 25 \text{Å}^{-1} \).

The detector itself is novel and has been developed for the POWTEX instrument. It is a "Jalousie" type detector with \(^{10}\text{B}\) absorbing cathode layers in a multi-wire gas detection chamber [19]. In order to achieve sufficient absorption efficiency, several layers covered with 1 μm \(^{10}\text{B}\) are composed in an inclined geometry creating a volume detector with a predicted and measured detection efficiency of 54% at 1 Å wavelength. Noteworthy features of such a volume detector are the higher count rate capability due to a depth of approximately 20 anode layers, the particular insensitivity to gamma background by coincidence measurement, intrinsic collimation properties
by tracing diffraction signals back to the sample source, and finally to mention is the intrinsic energy sensitivity that can be obtained by analyzing the measured absorption depth profiles.

The diffractometer has top and side access to the sample position and will offer an open platform for various sample environments for experiments at low to high temperatures, applying magnetic fields and high pressure, for using chemical reaction cells, and with capabilities to scan larger objects.

### 3. Instrument performance

The instrument has been simulated by including all components in detail verifying a high brilliance transfer along the neutron guide system and a clean and flexible time structure achieved by the chopper system. Finally, we studied the performance of DREAM for different resolution options by VITESS simulating the diffraction pattern of a common reference sample of Na$_2$Ca$_3$Al$_2$F$_{14}$ cubic (123) $a=10.257(1)$ Å [20]. Here, we chose a small volume 0.4 cm$^3$, which will be a typical sample size for the instrument. We have not included many details of the detector but account properly for distance and efficiency. These simulations were based on the 2013 ESS thermal and cold moderators. The new "butterfly" moderator yields gains of 2.57 and 3.4 for the thermal and cold spectra respectively, which will increase the DREAM performance accordingly and which have been used to scale the results shown in Fig. 4. We have also modeled other diffractometers POWGEN, WISH, GEM, D20 in VITESS in order to benchmark the performance. Here, we chose a typical but comparatively larger sample size of 0.8 cm$^3$. The flux of WISH for the high resolution setting was smaller in our model than given in Ref. [14], probably due to actual changes in the moderator compared to the available model. This has been taken into account by scaling appropriately the data by a factor 2 to
Figure 5. Simulated diffraction pattern versus wavelength and diffraction angle. The resolution varies along the Bragg diffraction lines. The insets show clear separation of diffraction lines. Resolution is best for backscattering, where shortest pulses of 10 µs yield a resolution of \( \Delta d = 2.8 \times 10^{-4} \text{Å} \). Instead of integrating data to \( I(d_{hkl}) \), see Fig. 4, a two-dimensional data analysis will optimally exploit intensity and resolution. Relaxing time-resolution may yield very high intensities with a flux of \( 10^9 \text{n}/(\text{cm}^2\text{s}) \) at the sample and count rates larger than \( 10^6 \text{n/s} \) for strongest peaks with better than 1% resolution.

The experimental flux at the sample of \( 1.08 \times 10^7 \text{n}/(\text{cm}^2\text{s}) \) [14]. For better comparison, we tried to match the resolution of POWGEN and WISH approximately also in the simulation of DREAM, using 40 µs pulse width and \( \pm 0.167^\circ, \pm 0.33^\circ \) divergence horizontal and vertical. For better clarity two peaks are selected and presented in the upper panel of Fig. 4. The comparison reveals the strength of DREAM, symmetric peak shapes and about two orders of magnitude higher intensities at comparable resolution. The diffraction pattern also reveals that the 3.7 Å bandwidth is rather suitable choice. With respect to the POWGEN instrument we considered two settings of wavelength frame for better comparability.

The comparison made in Fig. 4 is based on the conventional integration along the Bragg lines for each \( d \)-spacing. This is particularly fine for the instrument POWGEN, which by design has essentially a constant resolution, however, it is less appropriate for DREAM and also for WISH. Fig. 5 shows the \( \lambda \) and \( \theta \) dependence of the diffraction pattern for the DREAM simulation. Note that the graphical resolution is misleading, the instrument resolution can be better seen by the zooming inserts to the right, showing that the peaks can in fact be nicely separated with a better resolution of \( \Delta d = 0.001 \). The line widths change with \( \theta \) by the typical divergence contribution proportional to \( \cot \theta \). While it is certainly possible to refine the diffraction pattern shown in Fig. 4, the new approach of a two dimensional Rietveld refinement [6] will make much better use of the data quality. Furthermore, the data inspection and analysis in its \( \lambda \) and \( \theta \) dependence will give better control on background and also better reveal systematic deviations, such as structured background from sample environment or problems with detector banks.

4. Final remarks

The DREAM powder diffractometer will offer truly new opportunities with its flexibility that builds on the ESS source parameters. The example shown in the simulation does not show the best resolution, which will be close to best synchrotron beam lines. The case of medium resolution with \( 10^6 \text{n/s} \) for the strongest peak will enable real-time studies on ms-time scale for irreversible processes. The intense peak flux of the cold coupled moderator combined with shortest wavelength neutrons available, yields a wavelength band that is most valuable for
meeting the challenges of the science case contributions to the DREAM proposal, which were
highly appreciated by the Scientific Advisory Committee. The cases discussed for magnetism,
functional materials, metal-organic frame work structures [22], thermo-electrics [23], batteries
[24] have in common complex structures with relative large unit cells. Cases particularly
appropriate for neutrons in view of the sensitivity to even small magnetic moments as well as for
light elements on heavier atom background, however, very demanding in diffraction quality with
respect to resolution and the ability to distinguish small near to strong peaks [21]. Challenges
from material and applied science point towards studies of devices in operandi, for example
super-capacitors, material assemblies at short-time scales.

There are many fascinating capabilities and only few can be spotted here, for example,
structure determination of magnetic nano-particles by total scattering over multiple length
scales, and by future possibilities to combine this with neutron polarization and relatively simple
polarization analysis in forward direction. Studies at highest pressure are limited by the flux on
small samples and limits can be pushed significantly with the DREAM instrument. However,
it further requires to improve signal to background and we expect that this can be largely
improved by the proposed two and three dimensional data analysis. Resolving diffraction data
with position- and, as we propose, even volume-sensitive detectors with intrinsic sensitivity to
direction and energy transfer is as appealing as challenging from the point of data analysis.

As a final point, scientific advisors addressed the need for single crystal studies for signals
too weak for powder diffraction. The instrument will fulfill such needs by neutron time-of-flight
Laue diffraction and even with best gains from the ESS long pulse structure.

Acknowledgment
We gratefully acknowledge the valuable support from the scientific community which defined
the science case of the DREAM proposal and the funding from the German Federal Ministry of
Education and Research (BMBF) in the frame of the ESS Design and Update Programme.

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