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ESS Instrument Construction Proposal
DREAM (POWHOW)

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ENCLOSURES

EXECUTIVE SUMMARY

What are the dreams we have in neutron powder diffraction? The answers will be as diverse as the communities themselves. With respect to currently existing capabilities, there is as much a quest for high intensity and resolution for measuring weak spin half magnetic moments beside intense structural peaks in unconventional superconductors as there is for rapid measurements to follow fast reactions on *ms* and sub-*ms* scale. There is a need for a large Q-range for the most accurate structure determinations and also to analyse the deviations from crystalline order by measuring what is underneath the Bragg peaks. There are challenges to improve signal to background for pressure studies of tiny samples or to focus on small gauge volumes of battery samples studied *in operandi*. We wish to clearly distinguish imperfections of powders, analyse texture, grains and small single crystals. There is growing interest in studying materials simultaneously on various length scales, driving dedicated diffraction instrumentation. For nanostructured magnetic materials, there is still a dream of polarization analysis to reveal the structural and magnetic properties as well as the morphology in self-organized or man-made assemblies. The instrument proposed here will not be able to fulfil all of the dreams of the large and diverse user community; however, it addresses many of them and also some that we have not dreamed of before.

We have chosen the name DREAM for the proposed powder diffractometer, which during the design phase was called POWHOW. The name DREAM stands for **D**iffraction **R**esolved by **E**nergy and **A**ngle **M**easurements. The energy or wavelength dependence is crucial for high-resolution neutron time-of-flight powder diffraction, where dedicated backscattering detectors are used at today's pulsed spallation sources. A large 2D detector yields information on both the diffraction angle and on the texture angle along the Debye-Scherrer cones. The "E" also has a twofold meaning, as the new detector is a volume detector, which promises to retrieve information about the energy transfer upon scattering, from the wavelength-energy characteristics of the absorption profiles, using the depth of the detector volume. The "R" and "M" can alternatively stand for refinement and modeling, in which the variables "E" and "A" are fitted in 2D refinements to the varying resolution and exploit optimally the combination of high resolution and high intensity. Consequently, detectors are no longer "grouped" into complex bank arrangements. Form follows function: the diffractometer purposely has a simple cylindrical geometry adapted only to the Debye-Scherrer cones.

Flexibility is the most important and a new property of DREAM, which results naturally from the optimization at the ESS. The key design parameter of the new ESS source is the long pulse, which at first glance is not particularly desirable for powder diffraction, while full moderation and 5 MW yield an unprecedented peak brightness of the cold coupled H_2 moderator. Hence, different to instruments at short pulse spallation sources that are fixed in their properties by pulse width and geometry, here we need to tailor and shape the source pulse. Apart from achieving an ideal symmetric time profile, it is also easy to vary considerably the pulse width and, hence, resolution with the chopper frequency. Therefore, it is possible to offer unparalleled flexibility across a wide range of instrument set-up to users, from low resolution and high intensities to unprecedented high resolution – with the same instrument and without undesirable compromises. The highest resolution, Δd is limited

in backscattering by $\Delta d=(h/m_n)\Delta t/2s$ =2.8×10^{-4}Å with an instrument length of 75 m and a shortest pulse of 10 μs. In high intensity mode, by changing to a larger pulse width, the neutron flux on a standard sample will be sufficient to study changes on *ms* timescale, while investigations of irreversible process are only limited by the 10 *μs* time resolution. The ability and need to tailor the pulse assures a symmetric, proper shape profile. A new development is the data analysis in 2D refinements versus wavelength and diffraction angle, which will exploit even more efficiently intensities and resolution properties and with better control and quality on instrument and sample parameters.

The outstanding performance is gained by exploiting the ESS source strength through a variety of novel concepts and important new technologies. However, this also comes at low risk because of the experience gained from the instrument POWTEX, that is currently under construction the research reactor of the Maier-Leibnitz Zentrum in Munich. Based on this expertise, the current proposal is already quite advanced in details of new technologies involved. A key component is the large position sensitive detector based on solid ^{10}B thin film absorbers, which exhibits an excellent performance that is even superior to current ^{3}He-detectors. This multi-wire chamber detector is a volume, rather than an area, detector with additional time resolution and prototypes have been successfully tested. The detector is conceptually new and will provide new opportunities in neutron diffraction to distinguish signal from background. The pulse-shaping chopper is new, however in technical respect, the POWTEX developments can be taken with only little modifications. The new 2D-Rietveld analysis has already been successfully tested and will open new efficient ways to exploit resolution and intensities.

The qualitative features of DREAM create a broad functionality that can be adapted to the varying needs of the user. In quantitative comparisons using VITESS simulations we benchmarked the performance of DREAM with several neutron diffractometers, in particular WISH, POWGEN and D20. Among these, where the relatively modern WISH is the best performing and most competitive, we obtain a gain of about one order of magnitude in flux with better resolution and, with matching resolution, a gain of two orders of magnitude can be achieved. The 7 times higher peak brightness and the 10 times larger detector can easily rationalize this surprising performance. A further gain (x2.5) is expected with on-going development of the moderator design.

The proposer, the Jülich Centre for Neutron Science, has a high level of expertise in construction and operation of neutron instruments. The development of the instrument will further benefit from the involvement of the user community in accompanying the construction of the instrument, particularly in view of dedicated sample environment and software developments. We highly welcome the interest and possible future support of a strong partner, the Chemistry department of the Stockholm University, and we highly appreciate all the great contributions to the science case.

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1. INSTRUMENT PROPOSAL

1.1 Scientific Case

Piezo- and ferroelectric compounds

Gunnar Svensson, Mats Johnsson, Mari-Ann Einarsrud, Jekabs Grins

The perovskite structure is one of the most structural and compositional flexible structures and it is natural that numerous compounds with interesting properties have this structure. It is well known that chemical tuning resulting in subtle structural changes very often is the key to optimise an aimed property. Detailed structural investigations using neutron diffraction data of highest quality in resolution, q-range and intensity are necessary. We have a long experience in such fundamental studies of transition metal oxide compounds.

Recently, we have started a new collaboration with Professor Mari-Ann Einarsrud NTNU-Trondheim, Norway concerning piezo/ferroelectric compounds with perovskite related structures. These materials have a significant number of industrial and commercial applications. Among these applications are high-dielectric-constant capacitors, piezoelectric sonar and ultrasonic transducers, gas ignitors, sensors and switches as well as ferroelectric thin-film memories. Today lead zirconia titanate is one of the most used compounds in this field. There is a strong wish to replace this to more environmental friendly materials without lead and other harmful metals. Several candidates are considered such as bismuth potassium titanate (BKT), bismuth sodium titanate (BNT), bismuth ferrite (BF) and alkali niobates like potassium niobate ($KNbO_3$, KN) and sodium potassium niobate ($K_{0.5}Na_{0.5}NbO_3$, KNN). The main challenge with these materials is to obtain fine-grained and dense ceramics with high degree of preferential orientation in order to obtain improved physical properties. So far poor sinterability has been the main obstacle for the development of these materials but the use of alternative sintering methods like SPS (spark-plasma-sintering) and the use of reducing atmosphere have been shown to overcome these problems.

Ferro- and piezoelectricity depend on the direction of measurement with respect to the crystal axes of the material. For a polycrystalline material composed of grains with random orientation, those physical properties, being the average over each grain, are much lower than for a single crystal of the same composition. However, textured polycrystalline ceramics have a single crystal-like nature with respect to the orientation of the crystal axis and the physical properties are enhanced. High quality texturing of materials can also be achieved by the preparation of thin films on a selected substrate.

In this project we are combining the high scientific competence in SPS-sintering at the new Swedish national SPS-facility and the strong tradition in structural studies using powder diffraction and electron diffraction at the department of Materials and Environmental Chemistry at Stockholm University, Sweden, with the strong competence in high performance piezo/ferroelectric materials at NTNU-Trondheim, Norway. Neutrons have a high sensitivity for oxygen and a deep penetration length and the extraordinary performance of the planned POWHOW with high intensity high resolution diffraction data and ideal peak shapes will open up for new quality studies of the structure and texture of these materials.

Metal-organic frameworks

Xiaodong Zou

Metal–organic frameworks, or MOFs are porous crystalline materials constructed from metal ions/clusters linked by organic ligands (Figure 1). The metal building blocks can vary from rather simple polyhedron (e.g. $[AlO_6]^{-9}$ octahedron) to complicated clusters containing 2-8 metal polyhedra (e.g. $[Zr_6O_4(OH)_4]^{12+}$ cuboctahedron) or chains or layers. Typical pore sizes of MOFs range from 4 – 30 Å. The unit cell ranges from 10-100 Å.

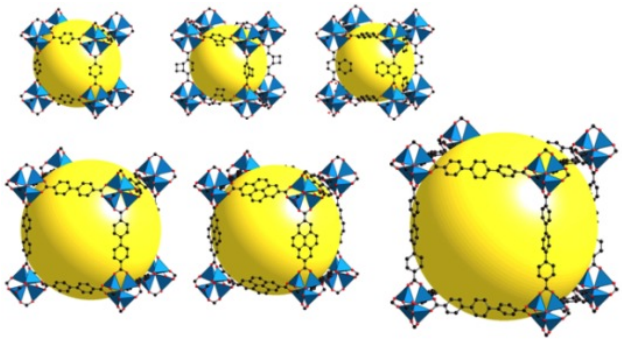

Figure 1. The pore-size and functionality can be altered in MOFs in a controlled way – reticular chemistry.[1]

MOFs can have large surfaces areas (>5000 m^2/g) and pore volumes, tuneable pore size and flexible frameworks.[1] In addition, MOFs can be chemically functionalised. The linkers may also contain functional groups to enhance chemisorption, selectivity and catalytic activity. The field of MOFs has tremendous potential to deliver applications in a wide range of areas, for example molecular storage, sensing, separation, controlled release and catalysis. With an almost endless choice of metals and organic linkers, these materials seemingly know no bounds.[1]

X-Ray powder diffraction (XRPD) has been widely applied to characterize MOFs. However, this technique is dominated by heavy scattering atoms, making the organic components (linkers) "invisible" to this technique. This has represented a big limitation so far, since the linkers are the most sensitive MOF components to be modified or functionalized by synthesis methods. Interestingly, the use of neutron diffraction would provide a unique opportunity to study the organic components. Recently, Zhou and co-workers have reported on the use of neutron diffraction to study MOF defects such as linker vacancies,[2] which have enormous implications in porosity. Another possible application of neutron diffraction is to study the location of guest species in the pores and their interaction with the MOF framework. Neutron diffraction is certainly an underexplored technique so far to study MOFs. However, its use would provide insights into how and to what extent defects in MOFs can modify properties.

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Microporous polymers

Niklas Hedin

Microporous polymers (MPs) are getting increasingly more studied and because of their organic composition neutron diffraction is an important tool to clarify their structural properties. These polymers have pores < 2 nm and can be synthesized in various ways. One

method is to use monomers that induce kinks and another is to condense robust monomers such as aromatic dialdehydes with aromatic triamines. They are typically X-ray amorphous and form under kinetic control, and our thesis is that they contain a fair amount of molecular order. Bennet et al. (PRL 104, 115503 (2010)) studied related amorphous metal organic frameworks (MOFs) with neutron total scattering at low-Q (Q<5.5 Å-1) showing that the molecular order of these amorphous MOFs are quite similar to those of their crystalline equivalents.

Elastic neutron scattering will provide answers to a range of questions about the local structure of these polymersand is particularly suited to study the short range molecular ordering within these polymers, chain-to-chain ordering, and various ordering of the polymerized units within the MPs. In addition, MPs may exhibit also ordering of pores on nanometer length scale. These polymers allow various labeling schematics including deuteration of the solvent or the various monomers.

In-situ neutron total scattering studies offer unique opportunities to study the molecular ordering during synthesis, polymerization of the MPs, and to refine the synthesis itself, which is typically conducted under reflux conditions with an organic solvent; however, also hydrothermal or other synthetics conditions are quite commonly used.

Magnetic structures

Mats Johnsson

During the last few years we have synthesized and described a number of transition metal oxohalide compounds containing also p-elements with stereochemically active lone pairs. The lone pair cations help to open up the crystal structures and act as structural scissors together with halide ions that have low coordination numbers. Such compounds are often geometrically low dimensional and the transition metal cations are arranged in layers or chains in the crystal structure. Those arrangements often lead to interesting physical properties such as magnetic frustration and even multiferroicity as indicated in measurements of magnetic susceptibility and heat capacity. For this kind of studies it is indispensible to solve the ordered magnetic structure at low temperatures from neutron powder diffraction data. Example of compounds we have studied are (i) $Cu_2Te_2O_5X_2$ (X=Cl, Br) that have weakly coupled frustrated Cu^{2+} spin tetrahedra, (ii) $FeTe_2O_5X$ that is an intrinsic multiferroic, (iii) The S = 1 quantum spin system $Ni_5Te_4O_{12}X_2$, (iv) recently we discovered the solid solution system $(Co_{1-x}Ni_x)_3Sb_3O_6F_6$ where the magnetic structure of the Co and the Ni analogues are different according to magnetic susceptibility measurements.

Organic-inorganic hybrids and nanoparticles

German Salazar-Alvarez

The research carried out in the Division of Materials Chemistry at the Department of Materials and Environmental Chemistry, Stockholm University consists of activities that focus on materials for fuels cells, ceramics, biomaterials for dental implants, and on porous adsorbents and organic-inorganic hybrids. The experimental techniques for characterization

are focused mainly on structural techniques imaging and scattering methods (electrons, X-rays, and to some extent neutrons); although thermodynamic and spectroscopic tools are also employed. Case studies of great interest to us are:

Organic-inorganic hybrids (OIH), with inherent lightweight and even flexible properties, are currently being explored for a number of applications in optics and photonics, health and life science, catalysis and sensing, energy, and transport and aeronautics. At MMK, we are particularly interested in the use of nanocellulose as the organic component and the formation of multifunctional materials in combination with inorganic nanomaterials.(Ref. 1,2) The use of neutrons is two fold: on one hand, a long-standing problem is structural characterization of OIHs as the interaction of the hybrids with X-rays and electrons is strongly influenced by the heavier inorganic components whereas the organic component is nearly impossible to distinguish. Neutrons can provide and improve dramatically the study of the formation and internal structures and morphologies of such organic-inorganic hybrids both in bulk and as coatings. On the other hand, although the crystal structure of cellulose Iα and Iβ has been solved relatively recently using a combination of X-ray and neutron diffraction, the structure of nanocellulose (and also nanochitin), composed of seemingly alternating crystalline and less-ordered domains is still under debate. Analysis of the neutron total scattering diffractograms would shed light into this issue.

Synthesis of heterogeneous nanoparticles and self-assembly. Particularly, we are interested in the study of the structural and magnetic order of heterogeneous nanoparticle systems (Ref. 3) and also of their self-assembly into large ordered arrays, both in the static and kinetic regime. In the case of heterogeneous systems, the magnetic interactions between the different components are of interest (Ref.4), whereas in the case of self-assembled particles, the magnetic interactions between them are very interesting. For this purpose we need an instrument with polarized neutrons and polarization analysis, which is as well of interest to separate the incoherent scattering of hydrogen in these organic-inorganic hybrids.
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Research topics from the Zernike Institute Groningen for Advanced Materials

Graeme Blake, Thomas T.M. Palstra

Multiferroics The magnetic ordering in multiferroic transition metal oxides is often complex, involving incommensurate structures, often with long periods. The high intensity cold neutron flux offered by DREAM would be advantageous in solving complex magnetic structures and particularly for mapping detailed phase diagrams as a function of temperature, pressure, magnetic and electric fields. The associated structural phase transitions can be difficult to probe. Here the single crystal Laue mode of DREAM might provide invaluable insight.

Magnetic alkali oxides In these materials, magnetic ordering involves the p-electrons of the superoxide anion. The superoxide anion is also Jahn-Teller active, thus orbital ordering often occurs with temperature. However, the non-spherical nature of the dioxygen anion gives an extra degree of freedom for the lifting of orbital degeneracy – the tilt angle – which results in the formation of (incommensurate) superstructures formed by small correlated tilts and shifts. These are extremely difficult to detect and probe accurately using X-ray scattering, especially in the presence of heavy Rb and Cs cations. Furthermore, samples are generally very small and until now it has not been possible to solve the magnetic structures of these materials. The greatly enhanced cold neutron flux and low background at DREAM (previously named POWHOW) would provide new insight into magnetic ordering and superstructures, whereas the high resolution mode would allow details of anion tilt/shift patterns to be elucidated.

Thermoelectrics We study a variety of novel thermoelectric materials. Here structural studies in the high resolution mode of DREAM would be most beneficial. Good thermoelectrics are often nanostructured and thus exhibit strain and complex peak shapes. Our interest here is to probe the evolution in (nano)structure of bulk samples with temperature and thermal cycling. The combined high resolution and intensity of DREAM would allow this to be probed quickly and with more accuracy than is possible by X-ray scattering.

Novel transition metal chalcogenides These materials have been explored to a much lesser extent than the corresponding oxides. However, coupled spin, charge and orbital degrees of freedom are often still at play, giving rise to a rich array of physical properties. As with the other systems discussed above, DREAM would provide new opportunities for probing magnetic structures and structural phase transitions and quickly mapping out entire phase diagrams.

Chasing Incommensurate Magnetic Ordering and Magnetoelastic Anomalies in Multiferroic Quantum Spin Chains

Reinhard K. Kremer

The quest for new high performance multiferroic materials, i.e. materials with simultaneous ordering of more than one ferroic order parameter, is stimulated by the expectation to control, for example, magnetic ordering properties by electric fields and, vice versa, ferroelectric polarization by magnetic fields. Such multiferroics systems are expected to open

new routes to efficiently tune magneto-optical/magneto-electric multifunctional memory devices. Understanding the underlying physics and especially the intricate coupling of the magnetic and the lattice degrees of freedom is a continuing challenge for high-resolution neutron powder diffraction.

In an interesting family of systems, multiferroicity is related to incommensurate spiral magnetic ordering in S=1/2 quantum spin chains which, for example, can favorable spawned in frustrated magnetic chain systems featuring a competition of nearest-neighbor and next-nearest-neighbor spin exchange interactions along the chains. Recently, we have demonstrated that anhydrous $CuCl_2$ and CuBr2 constitute such multiferroic materials, the latter with a remarkably high critical temperature of ~75 K.

Several attempts to grow single crystals of $CuBr_2$ were unsuccessful and the magnetic structure and the temperature dependence of the magnetic order parameter and structural anomalies had to fully rely on neutron powder diffraction experiments.

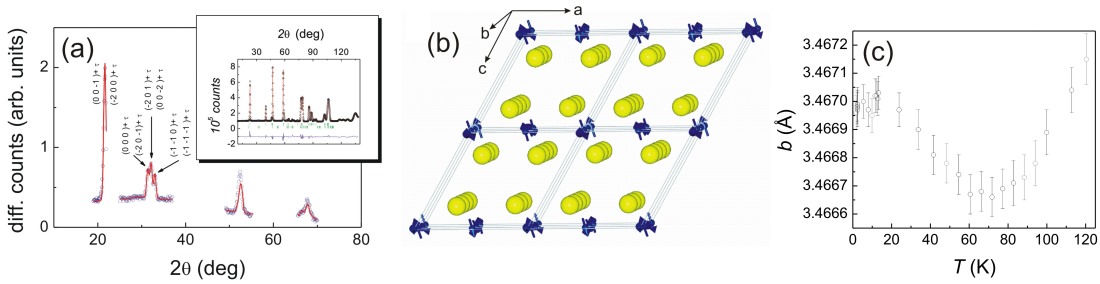

Fig. 2. *Neutron powder diffraction patterns of $CuBr_2$*. (a) Main frame: difference between the 2 K and 80 K patterns. The inset displays the powder diffraction pattern l = 2.4 Å at 2 K together with a FULLPROF profile refinement (solid line). Note that the intensity of the strongest magnetic Bragg reflection near 20° amounts to ~1% of the strongest nuclear Bragg reflection. (b) Magnetic structure of $CuBr_2$ at 2 K. The Cu and Br atoms are represented by blue and yellow circles, respectively. (c) Magnetoelastic anomaly of the Cu - Cu distances in the Cu^{2+} chains near the phase transition (l =1.9 Å).

Currently, neutron diffraction studies on low-dimensional quantum are still challenging due to the small magnitude of the S = 1/2 moments (typically 0.5 μ_B or less) and the very small magnetostructural anomalies associated to the quantum phase transitions. Studying simultaneously magnetic and lattice degrees of freedom of such systems by neutron powder diffraction requires incisive compromises and careful - and at current often mutually exclusive - optimization with respect to intensity and resolution of the neutron powder diffractometer.

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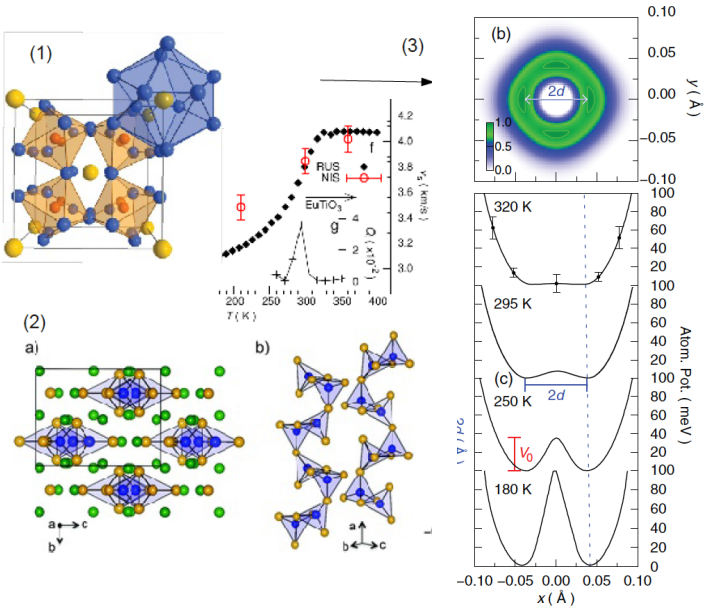
Thermoelectric materials, *Raphael Hermann*

Thermoelectric materials provide direct capability for transformation of thermal to electrical energy, or vice-versa. The figure of merit for this transformation contains properties that are contradicting in typical materials, namely both good electrical conductivity and poor thermal conductivity.

Recipes for reaching an effective compromise nowadays include band structure optimization with enhanced band multiplicity as well as the use of materials with very large unit-cells or paraelectric materials in close proximity to a ferroelectric phase transition.



Among the materials classes that are highly investigated, cage compounds such as skutterudites, Ref. 1, Fig. (1) and clathrates, or complex structured valence precise Zintl phases, such as *e.g.* in Ref. 2, which feature antimony multi-anions, Fig (2), play a prominent role. The unit cell volume of these materials typically exceeds 1000 $Å^3$, and detailed diffraction data that covers a large range in d-spacing is required in order to obtain a correct structural model, including atomic displacement parameters, which -prior to lattice dynamics investigation- provide first insights in potential mechanisms that lead to low thermal conductivity.

A second class of material that received significant interest are the rather simple perovskite or rocksalt structured materials such as $EuTiO_3$, PbTe or $AgSbTe_2$. In these materials entirely different mechanism lead to low thermal conductivity, namely proximity to lattice instability, Fig (3), and anomalously large or anisotropic displacement of the cations (Ref. 3). Here access to a large range of d-spacing with excellent resolution is highly interesting in order to refined the potential landscape that the cations explore.

A combination or x-ray and neutron diffraction studies has been driving research as the primary tool for reaching microscopic insight in the relation of structure and functionality in thermoelectric materials. The combination of the varied contrast for both methods and the power of neutron diffraction in obtaining reliable displacement parameters lies at the heart of past success. The future availability of an extended Q-range instrument with high throughput for structural analytics will enable extremely detailed insights of phase diagrams, notably through parametric studies as function of temperature and pressure, with the added strength of providing at the same time limited microstructural information.

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Potential user base

We performed a survey of the scientific publications on neutron powder diffraction experiments in the last 2 years, i.e. 2012 and 2013. Data were taken from the web pages of the existing operating instruments: POWGEN at SNS, D20 at ILL and WISH at ISIS. The scientific field of main interest for the single experiment was identified and the number of publications was classified according to their affinity. The result is shown in Table, where it can be seen that the majority of the experiments were performed on magnetism-related materials (50.7%) [Typically the investigations lead to determination of magnetic structure or crystal structure following change of magnetic behavior], followed by oxides compounds (14.5%), composites (typically Li- based) used for batteries (7.9%), identification of perovskite structures (7.2%), liquids (6.6%), hydrogen absorbing materials (5.3%).

	POWGEN		**D20**		**WISH**		**TOTAL**	
	publications	%	publications	%	publications	%	publications	%
magnetism	20	38,5	33	45,8	24	85,7	77	50,7
oxides	13	25,0	9	12,5			22	14,5
batteries	10	19,2	1	1,4			11	7,2
perovskites	7	13,5	3	4,2	1	3,6	11	7,2
liquids			10	13,9			10	6,6
hydrogen			7	9,7	1	3,6	8	5,3
others	2	3,8	10	13,9	2	7,1	14	8,6

Moreover we asked user offices to provide us with data of requested and scheduled beam time on their instruments. According to the data from POWGEN: since 2010, 316 experiments were requested by means of the regular calls for proposals and 169 experiments were performed, therefore leading to an overbooking ratio of nearly 2.

Searching on Web of Science, there are about 600 publications per year in neutron powder diffraction. The most highly cited papers from the last 10 years are related to research fields of metal-organic frameworks (MOF) and hydrogen storage in these micro-porous materials, followed by Li-battery research and, magnetism and superconductivity.

These science topics and the resulting specific needs for instrumentation were addressed in a workshop, which we organized in 2011 at an early state of the design phase. Large unit cell materials (MOF), multi-phase samples (study of batteries in operandi), weak magnetic signals request for high resolution, flexibility for high intensity, and sufficient Q-range covered with thermal and cold neutrons.

With the subsequent optimization of the DREAM powder diffractometer and its unprecedented potential, there are prospects to reach new users and communities as concluded from discussions with our partners at the chemistry department Stockholm. Magnetic nanostructures, texture and, MOF studied by smallest single crystals are such themes, where with the proposed instrument we may dream of new horizons.
There is a high potential for applications to a variety of challenging cases. The instrument can offer a superior high resolution in back scattering, exceeding the records of SuperHRPD. Tiny peak splitting upon symmetry lowering, incommensurate long period structures, weak magnetic peaks near the intense peaks of the crystal structure, details of the peak profile due to strain and crystal size, all this will be possible to explore with unprecedented quality. A most important difference to short-pulse spallation sources is the flexibility given without making undesirable compromises. It is easy to change the pulse width turning to high intensity mode, where a single ESS pulse contributes approximately 2×10^5 neutrons to a single strong diffraction. Such high intensity will enable to measure extreme cases of small samples or fast irreversible kinetics even at ms resolution. For periodic external fields, pulsed ultrahigh magnetic fields, laser heating, recorded in event mode, the ultimate time resolution limit of 10 μs is given by the shortest neutron pulses available and the backscattering option.

The new possibilities offered by the proposed neutron powder diffractometer DREAM can be illustrated by a calculated diffraction pattern of a typical sample, which is based on a (VITESS) simulation of the instrument in its very details. The result is presented in a diffraction intensity distribution as a function of wavelength and diffraction angle rather than the usual 1D diagrams of intensity vs 2θ or crystal lattice d-spacing. Intensity and resolution varies along the diffraction lines. Diffraction near backscattering with the best time-resolution chosen, see Fig. 3, warrants utmost high resolution and with sufficient intensity. The choppers can be easily tuned to larger wavelength and with fixed opening times the relative resolution Δd/d further improves.

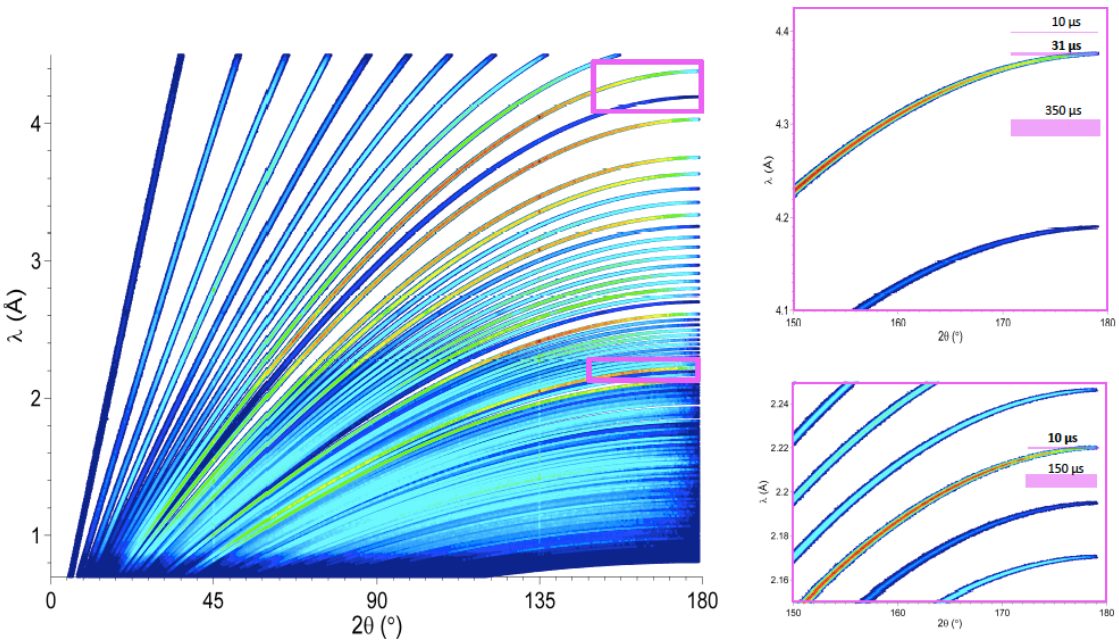


Fig. 3. Diffraction diagram of a reference sample (0.4 cm^3 $Na_2Ca_3Al_2F_{14}$) in high resolution mode (left). In backscattering, the asymptotic limit is essentially determined by the time resolution, see enlarged regions (right).

Comparison and complementarity to ESS instruments

The *Heimdal* instrument makes a new approach to measure at multiple length scales using an additional cold guide for small angle neutron scattering at $Q \sim 10^{-3} Å^{-1}$. The instrument includes a thermal powder diffractometer combined with a large detector including backscattering. The instrument will be highly performing for crystal structure determination, the instrument will be less well suited for magnetism and large unit cells, an important science case of DREAM. DREAM will not provide a dedicated SANS option, the upgrade option for forward scattering will include however $Q \sim 10^{-2} Å^{-1}$ for nanostructured materials. There is more complementarity rather than a conflict of interests.

Currently still under development is a D20-like instrument (HOD) using TOF mode and multiple wavelengths. This instrument may also address a similar science case as the original D20. However, time modulation offers efficiently means for separating inelastic background, and hence, the instrument will be dedicated for studies of hydrogenous materials. One may note that DREAM has an unexplored and interesting potential to discover inelastic scattering from the energy dependence of the absorption profiles measured in its volume detector. The approach of HOD is, however, based on a clear and direct discrimination of inelastic scattering by TOF measurements. The smaller detector array (of HOD) is more restricted in vertical acceptance, which is favorable for vertical magnetic fields and also fits to possible He-3 polarization analysis. The instrument is complementary to DREAM.

The Engineering Diffractometer provides a relatively small coverage of the solid angle of detection and uses a time-modulated intense beam. There is emphasis on 90 degree diffraction dedicated to stress strain measurements in complex samples with simple structures. There is no significant overlap or conflict of interests.

Comparison to existing instruments

A quantitative comparison to the instruments D20, WISH, POWGEN is given at the end of 1.2. Here we discuss the qualitative differences of existing instruments and the origin and motivations leading us to the new conceptual developments for DREAM.

The instrument POWTEX at the MLZ in Munich can be regarded as a conceptual forerunner. POWTEX is a Powder and Texture diffractometer, which operates wavelength and angle dispersive in TOF mode at a continuous reactor source. *Pulse shaping choppers*, *detector configuration, B-10 detector technology*, and *2D data Rietveld refinements* are all important new developments, of which we make use also in the current design. Compared to POWTEX, however, the DREAM at the ESS will be much more advanced, a very flexible instrument with optional highest resolution, it is not limited by the thermal moderator spectrum and benefits from the particular high brightness of the cold moderator at the ESS.

Compared to the traditional monochromatic diffractometers based at continuous sources and reactors, the wavelength and angle dispersive TOF instruments, without the need of crystal monochromators cover typically a larger solid angle of detection. The variation of wavelength provides an increase in d-spacing that can be covered in a single measurement. In addition, the modern TOF-diffractometers have a higher potential for high-resolution experiments using backscattering, see HRPD and SuperHRPD, and because of the large detector, better sensitivity and efficiency for texture, see POWTEX, and single crystal capabilities, see WISH.

To highlight the strategy and uniqueness of DREAM we discuss interesting differences to the instrument concepts of existing powder diffractometers at short-pulse spallation sources.

Flexible time resolution For these instruments typically, the pulses are sufficiently short that one may choose the primary flight path to set a fixed time resolution. The pulse width is approximately proportional to lambda resulting in a constant contribution to the resolution in $\Delta d/d$. A variation in resolution can made by changing the incoming divergence. Typically, the secondary instrument is designed to focus sets of detector banks to similar resolution properties, which makes the analysis similar to the monochromatic instruments. For a long-pulse spallation source, there is an evident need to tailor the pulse, and if this is required one can do this with flexibility and best adapted to the resolution needed. Tailoring the pulse also assures a better and symmetric peak profile. Pulse shaping has a price of intensity in favour for resolution. The question which source is more appropriate for powder diffraction is answered by the source brightness in the phase space of interest. The ESS will be superior for the cold neutrons and competitive or better for wavelength larger approximately 0.8 Å.

Data analysis Because of this property, it makes less sense to gather detector elements focusing to similar resolution properties. Rather we pursue a new development to refine the diffraction data varying on a 2D grid of wavelength and diffraction angle. This will also warrant to benefit best from the varying resolution function and intensity.

Detector A benefit from the new data analysis is that we can arrange the detectors in a much simpler, cost effective way and take into regards other aspects as sample accessibility. One may note that WISH also uses a relatively simple detector configuration with position sensitive He-3 tubes vertically aligned on a circle in the horizontal scattering plane. The POWHOW (POWTEX) detector marks a new milestone in detector development. It uses solid B-10 in thin film absorbers assembled to a 3D volume detector with high detection efficiency (>50% at 1 Å neutron wave length) and high spatial resolution of 3-4 mm. This new type of volume detector has further interesting potential for *reduction of background* from sample environment and intrinsic inelastic incoherent scattering within the data processing analysis.

Bispectral switch The proposal will include a relatively new concept, tested first at the HZB reactor, of a so-called bispectral switch, which selects and combines neutrons from cold and thermal moderators to the envelope of their respective higher brilliance. Taking the best of each moderator is rather favourable for the science case of POWHOW.

Background The need for suppressing the background from the prompt fast neutron pulse is crucial, avoiding direct line of sight will sacrifice the guide transport of neutrons with short wavelengths. To reduce sufficiently this background, to cause less shielding issues and disturbances of neighbouring beam lines, we foresee to include a massive T0-chopper spinning at the low source frequency.

Infrastructure and support facilities needed for the proposed experiments

The instrument will be able to work with standard sample environment of the ESS. For kinetic experiment, event mode and data organization will be a typical need. A promising task for DMSC is seen in developing data analyses, which utilizes the 4D information in the data with ray tracing to discriminate to non-sample-intrinsic background and with absorption profile analysis to discriminate inelastic background. We have already worked out and tested the 2D Rietveld refinement. Furthermore, the software needs to be developed to provide such refinements already during the experiment based on existing programs (GSAS, TOPAS, MAUD, Ref. 1-3). Users will need "on the fly" data inspection.

1.2 Description of Instrument Concept and Performance

1.2.1 Concept and general considerations

The key design parameters for the **bispectral powder diffractometer** are found by addressing needs for standard structural characterization, magnetic structures and larger unit cells. Therefore, **a combination of thermal and cold neutrons** is most wishful. The beam ports of the ESS will offer a view to both thermal and cold moderators being placed next to each other. As will be shown below, a bispectral extraction system may offer simultaneously a combined thermal and cold peak flux with high efficiency.

The available high peak brightness of the ESS, as compared in Fig. 4, is essential for TOF diffractometers with their typical need for high resolution, while large intensity gains can be obtained for relaxed time resolution from the average ESS brightness. The ESS moderators are still under optimization, a reduced moderator height of 3 cm for example yields a 2.5 times higher luminosity for the cold moderator with a full gain for the instrument performance.

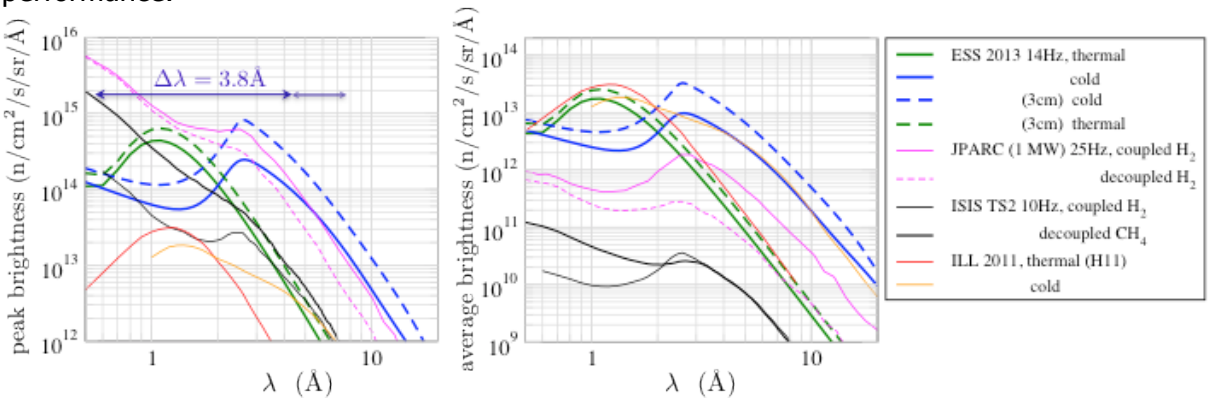


Fig. 4. Peak and average brightness of cold and thermal moderators at the ESS in comparison to the ILL, JPARC and ISIS target station 2. The wavelength band Δλ of DREAM is shown for the standard low λ setting and can be shifted to larger wavelengths up to 20 Å. (Ref.4,5).

At the ESS, the pulse duration of ~3 ms is not really compatible with the demands in resolution for powder diffraction, and the full flux is only useful in more exceptional cases. More important is the superior brightness of fully moderated ESS source, which will already guarantee the

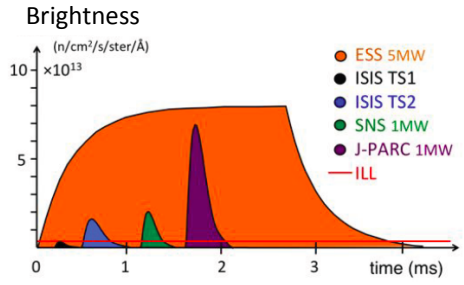


Fig. 5. Pulse shape and brightness.

possibility of *world leading performance for thermal and cold neutrons*.
The need for pulse shaping is mandatory, it is technically feasible and it provides a new *flexibility in trading intensity versus resolution with top-level brightness* on a single instrument superior to any instrument with highest flux and to highest resolution (SHRPD). In Fig. 5, one may note the asymmetric profile at short-pulse spallation sources. Here, pulse shaping will produce **ideal symmetric peak shapes**. Another difference will be that the pulses will be wavelength independent.

The accessible range in wavelength resolution due to pulse shaping is illustrated in Fig. 6.
For the shortest pulses of 10 μs and cold neutron, DREAM can achieve a world wide leading resolution utilizing in particular the backscattering option. More importantly, the instrument will have a large flexibility choosing and trading resolution for intensity without comprising its performance. Such properties cannot be realized at existing sources by a single instrument. The instrument is limited for short wave lengths to 0.5 Å. The standard setting covers a frame 0.5Å<λ<4.3Å offering a **Q-range of 0.01Å^{-1}<Q<25Å^{-1}** when including the whole detector. The chopper systems allows for shifting to cold neutrons up to 20 Å wavelength.
Different to the proposed instrument DREAM, the Δλ/λ resolutions of POWGEN (Ref.6) and SHRPD (Ref.7) are constant and results from the natural moderator characteristics at these short-pulse spallation sources. One should note that SHRPD operates 80% in 5Hz mode using only 1 out of 5 pulses to use a larger bandwidth of 8 Å, which is particularly interesting for high resolution backscattering using the cold neutron spectrum. DREAM will operate with Δλ=3.8 Å bandwidth using all pulses. Pulse shaping to 10 μs yields **ultimate high resolution** for neutron powder diffraction at λ>1.5Å. This high resolution is often comparable to an intrinsic broadening width of diffraction peaks due to surface effects of powder samples, which sets the ultimate resolution required for diffraction experiments.

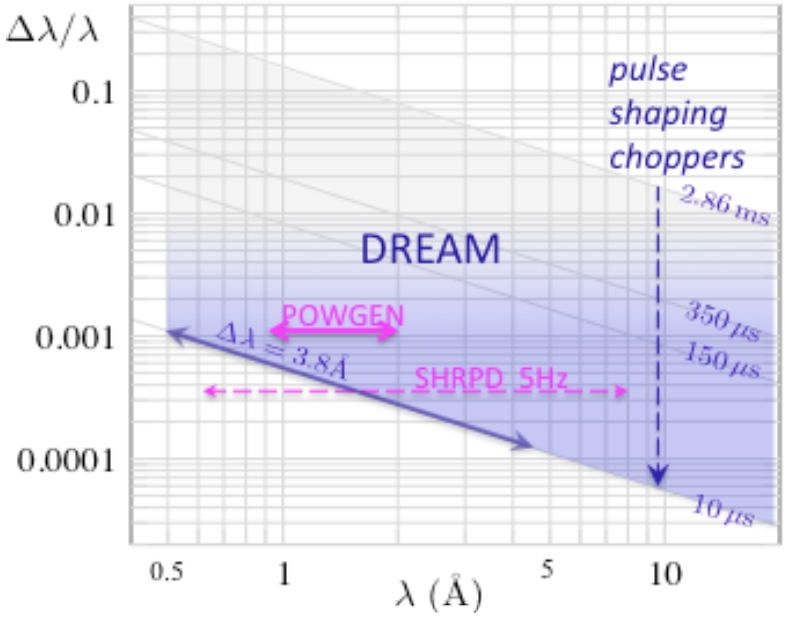


Fig. 6. Flexibility and range of wavelength resolution Δλ/λ versus λ due to pulse shaping from full pulse width 2.86 ms to high resolution 10 μs.

1.2.2 The DREAM instrument and its components

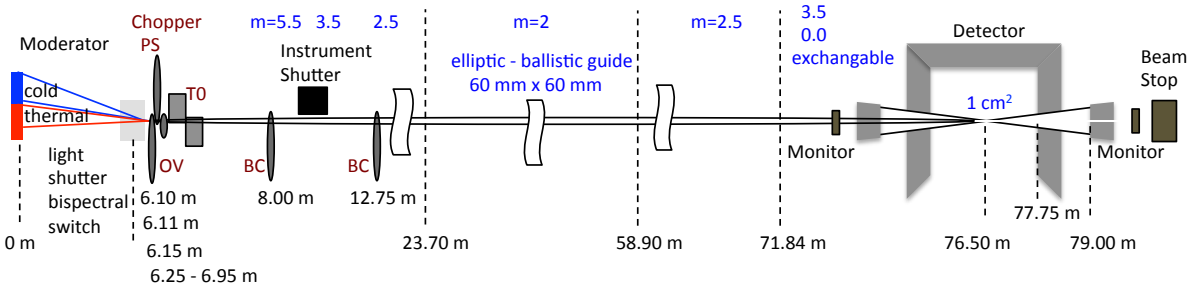


Fig. 7. Schematic layout of the DREAM diffractometer.

A schematic layout of the instrument is shown in Fig. 7. The instrument will have a common view to thermal and cold moderator. The elliptic guide system has the focal points at the pulse shaping chopper (PSC) and the sample. The focal point at the PSC is ahe needle eye in space and time and is illuminated from the source by the required divergence. PSC, a 14Hz overlap chopper and a 14 Hz T0 chopper for prompt pulse suppressing fit all near to the monolith wall; note, the 1m long chopper system can be placed favourably in the defocusing initial part of the elliptic neutron guide without losses for the neutron transport. Two additional choppers for and a heavy instrument shutter are placed in this initial part that is still with the copper shelf shielding as can be seen from the floor plan Fig. 8. The final guide section will have exchangable neutron guide parts either to focus the intensity to a smaller sample spot or, with absorbing end pieces to improve the divergence of the incoming beam. The sample position is at 76.50 m from the moderator and surrounded by a large detector. The detector geometry is cylindrical with its axis in common with the beam axis, rigorously adapted to the Debye-Scherrer cones.

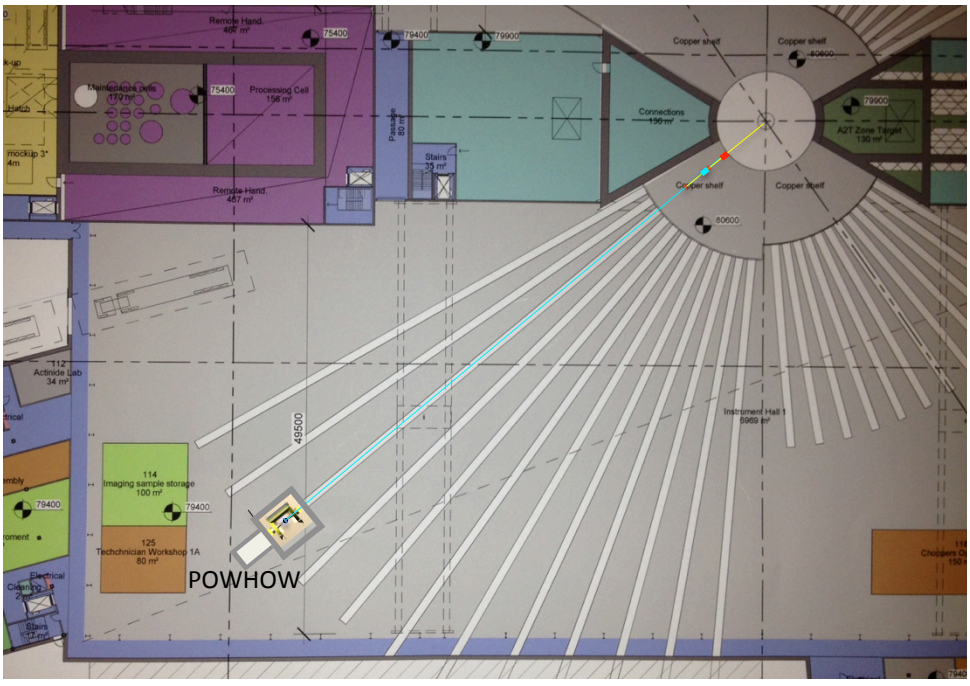


Fig. 8. DREAM in the floor plan of the ESS experimental hall. Choppers (red) and instrument shutter (cyan) are in the copper shelf shielding next to the monolith. The sample position and surrounding secondary instrument is at 76.5 m from the moderator surface.

Bispectral extraction system

We have investigated the performance of three possible reflection systems to combine both thermal and cold spectrum, a single mirror in the monolith, a stacked mirror system (Ref.8) and a very short solid state Si-bender placed moveable and easier accessible in the light shutter, see Fig. 9. The 60.5 mm long bender shows the best performance in the comparisons by VITESS simulations and can be placed even outside the monolith in front of the PSC.

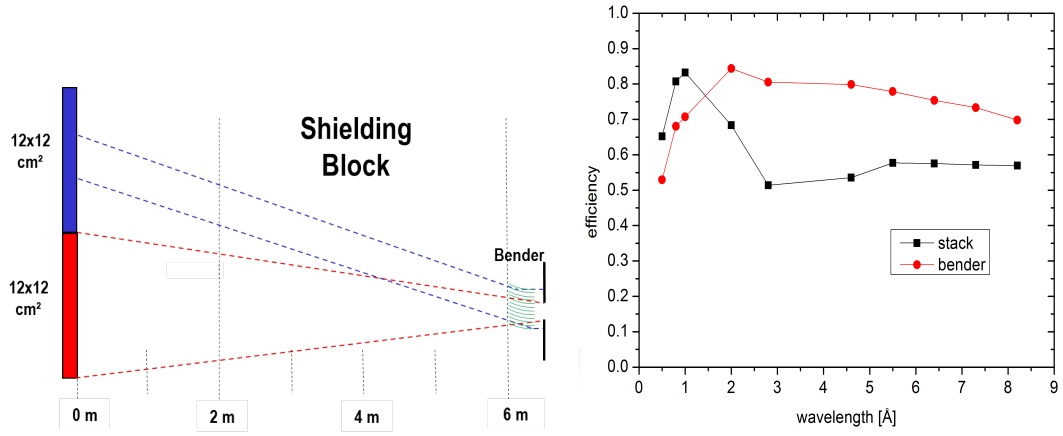
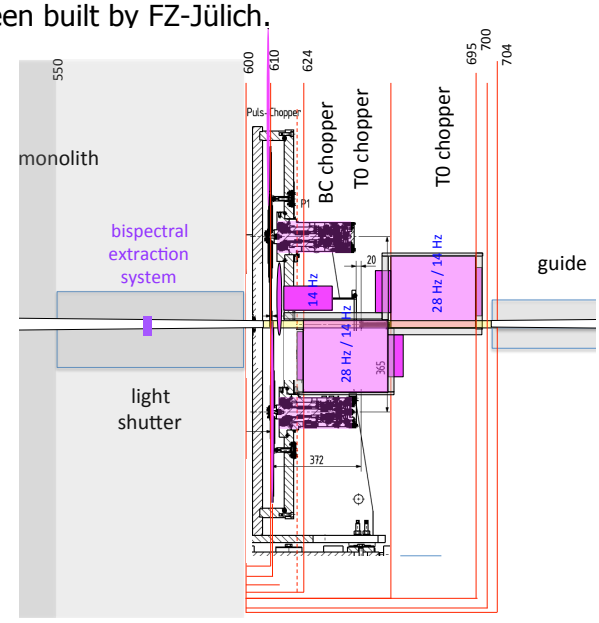


Fig. 9. Scheme of the bispectral bender-type extraction system and performance in comparison to a stack of neutron mirrors. Bender: coating m=2.87, L=60.5mm, R=3.051m.

The chopper system: Flexible pulse shaping, overlap & background control

The chopper system has been described in a detailed report (2012). The PSC consists of two counter-rotating disks able to rotate up to 210 Hz. Here we can use the identical chopper system of POWTEX with magnetic bearings and the same disk diameter of 75 cm that has been built by FZ-Jülich.



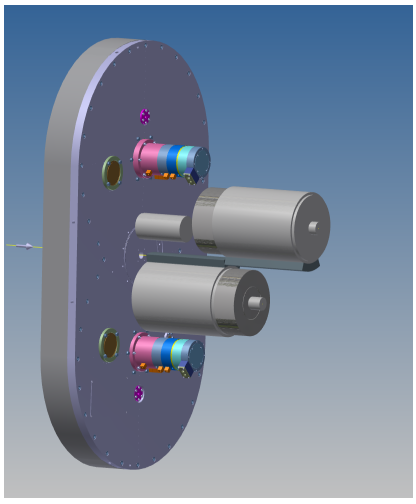
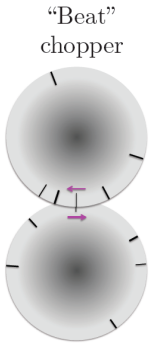


Fig.10. (left) The chopper arrangement near the monolith;(right) perspective view.

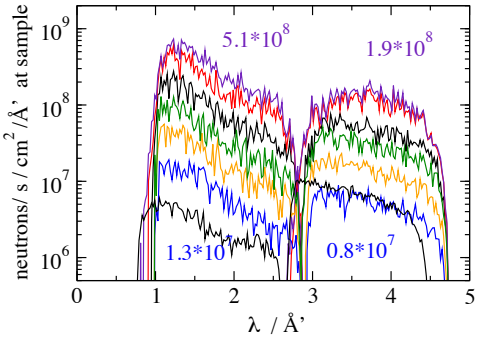
Fig. 10 shows the drawings for the counter-rotating PSCs complemented with small 14Hz BC (band control chopper) followed by two T0 choppers each with a hammer of 35 cm attenuation length. The composition of the hammer material is still to be optimized by neutronic simulations. A major part of it could be W. We have a design concept and have proposed to build a prototype of this T0 chopper in collaboration with the ESS chopper group. The whole assembly will be built on a common platform, which could be installed and access by lifting from the top.

For pulse shaping we realize a new idea of a "beat" chopper, which consists of two counter rotating disc choppers that spin in different multiples of the 14 Hz source frequency. Different slit pairs matched by different frequency ratios provide flexible resolutions, and different opening widths are chosen for the two sub-pulses.



Time resolution options

frame 1 $\delta t(\lambda_1)$	frame 2 $\delta t(\lambda_1 + 1.9\text{Å})$	$\omega_1 : \omega_2$ (Hz) : (Hz)
10.5 μs	31.3 μs	210 : 196
27.5 μs	82.5 μs	84 : 70
60.6 μs	181.8 μs	45 : 28
151.5 μs	353.5 μs	28 : 14
227.4 μs	606.3 μs	14 : 14
378.8 μs	681.8 μs	0 : 14



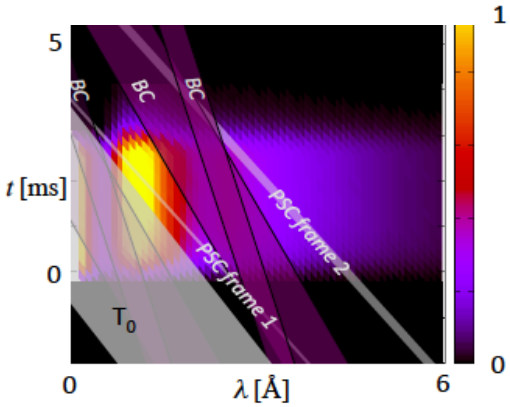


Fig. 11. The counter-rotating disks of the pulse shaping chopper, so-called "beat chopper", spinning in different multiples of the source frequency (left). The table shows the various pulse widths that can be by achieved, and (right) VITESS simulations of the wavelength distributions at the sample position varying with chopper opening times.

Fig.12 Acceptance diagram (see Ref.9)

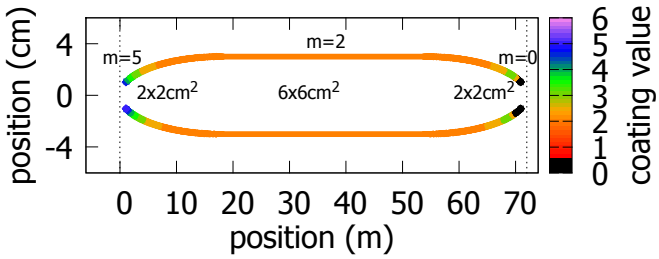
VITESS simulations have shown that the parameters needed further optimization to suppress background from the exponential decay of the source pulse. With appropriate positions at 8 m and 12.75 m for the 'bandcontrol' (BC) choppers, the afterglow of the ESS pulse can be suppressed for 6 ms. The principle of the interplay of the various choppers on the ESS pulse wavelength distribution is seen in the acceptance diagram (cf. TDR, not the actual flux distribution).

The neutron transport and guide optics

We have investigated several neutron guide concepts and optimized with respect to a best brilliance transfer for the desired neutron phase space at the sample, +/- 0.25 divergence for 1cm^2 beam cross-section and wavelengths larger than 0.75 Å. The relevant results for the current design are included in a detailed report on wavelength frame multiplication. In order to extend the transport for shorter wavelength, namely 0.5 Å, we decided recently to scale up accordingly the m-coating up to a maximum of m=7. This is possible at reasonable cost

and yields an important gain in Q-range up to 25 $Å^{-1}$. Note however, the figures shown below refer to the original definition.

The proposed neutron guide is of double elliptic shape (horizontal and vertical) at the beginning and at the end with a straight section in the middl optimised and determined by a "backtracing" method (Ref.10) In order to refine the initial divergence, the last parts of the guide can be exchanged by absorbing elements (not shown in the figure).



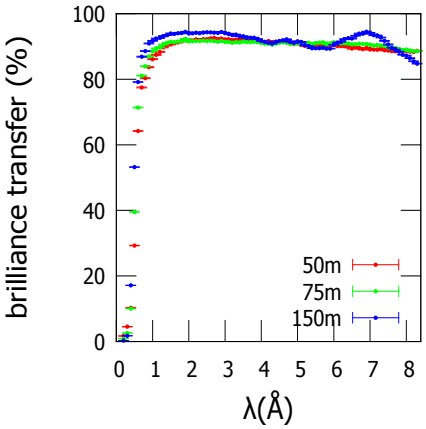


Fig. 13. The brilliance transfer (see right figure) for the desired wavelength band is high and smooth for the choice of the 75 m instrument length.

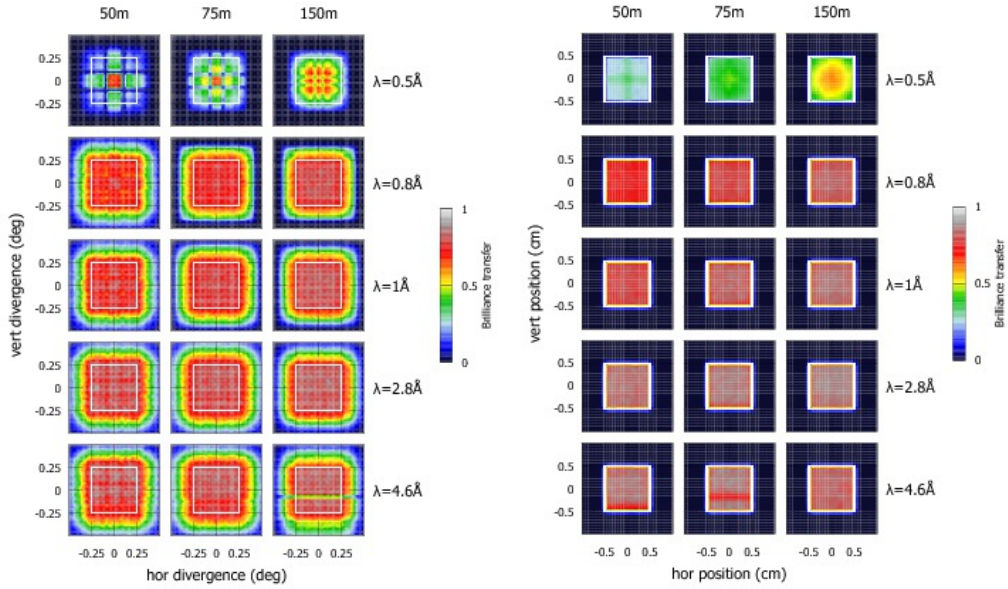


Fig. 14. The divergence profile at the sample, which is homogeneously illuminated, displays a smooth distribution, which is important for powder diffraction and data refinement.

The neutron transport and guide system for a reduced moderator height

Currently the ESS-Target group is investigation how to optimize the useful flux of the moderator essentially by increasing the luminosity by reducing the source height of the moderator. In order to benefit from such a luminosity source gain, the neutron optics and guide system need to be optimized in this respect. Therefore, it is important to realize a point-to-point focusing vertically from source to sample. For the proposed instrument it is straightforward to modify the guide system such that the vertical focus is on the moderator instead of the pulse shaping chopper (PSC) position. Since the horizontal focusing is still kept at the PSC position, this modification has essentially no impact on the time resolution. We

have performed VITESS simulation to investigate performance and benefit from such a new moderator. Our results show that the full luminosity gain can be exploited by the instrument DREAM for moderator heights of 3 cm and larger, with an average gain of 2.7 for the standard wavelength band. For a smaller moderator height, inhomogeneity develops in the vertical divergence pattern. A report has been given to the ESS.

The diffractometer

The part of the instrument in front of the diffractometer provides a beam definition in divergence that is variable up to **0.5°**=0.00873 rad, which sets the minimal requests for ratios sample size to sample-detector distance and spatial detector resolution to sample-detector distance. Choosing a **sample-detector distance of 1250 mm**, the incoming divergence is matched for samples and spatial detector resolution up to 1 cm. For backscattering the divergence contribution becomes negligible as it scales with cot(θ), and vice versa for forward scattering divergence will be the dominant resolution term. Considering typical samples of 5mm thickness and scattering at 90°, there is an option to improve the incoming divergence to +/- 0.125°.

For powder diffraction, a cylindrical detector geometry around the beam axis is best adapted to the Debye-Scherrer cones and facilitates an easy integretation along the angle φ at constant θ. The detector will be based on the development by the company CDT for the POWTEX instrument, which has achieved a rather high and desirable performance (Ref.11). Based on B-10 sputtered Al absorbing cathodes with inclined geometry, the efficiency is 53% at 1 Å as calculated and verified by the first detector moduls build. This will be also the choice for the PowHow detector. It will have a spatial resolution of 3-4mm horizontally and verticallly, important for backscattering and single crystal requirements and it is matched to the symmetric incoming divergence. The solid angle will be covered by 6.2 sr practically without blind parts. This detector has actually a spatial resolution in 3D, which is needed to related detection events in the volume properly to time and wavelength. According to the information requested from CDT, the detector developed for POWTEX can be scaled in straightforward manner to the larger dimensions needed for DREAM.

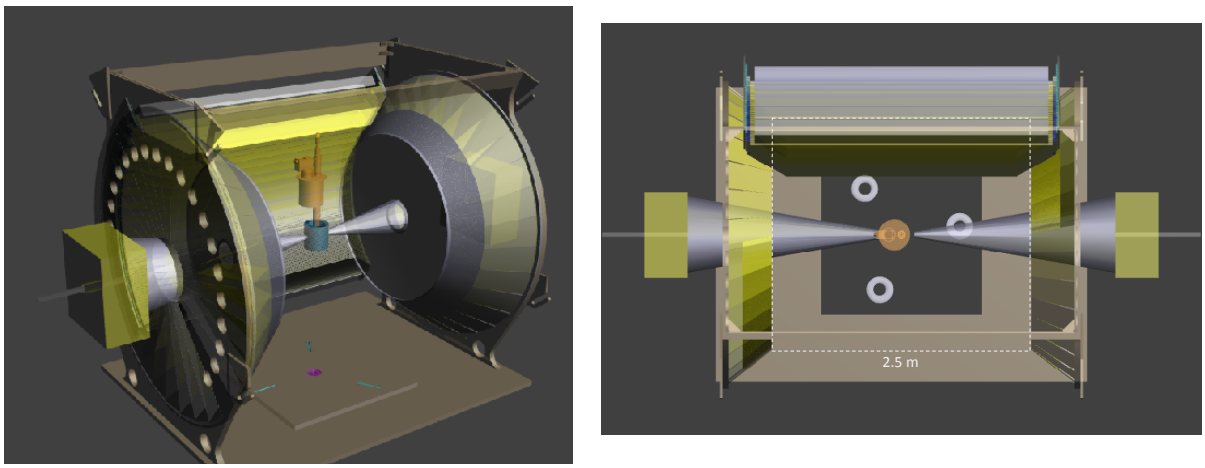


Fig. 15. The diffractometer. (side and top view). The cylindrical detector configuration with 6.2 sr coverage is adapted to the Debye-Scherrer cones around beam axis. The detector is a 3D wire chamber with B-10 film absorbers of high efficiency, homogeneous 2D response with high spatial resolution. Background is reduced by evacuating the primary flight path, a

vacuum vessel around the sample and Argon-atmosphere in the scattered flight path. There is convenient sample access from top (standard), side and bottom.

In backscattering within +/- 12 degrees from the beam axis, there is a dedicated 1 m^2 backscattering detector behind the larger circular detector covering the 135 to 168 degree section. Primary and scattered flight path share a common vacuum within this cone covered by B and/or Gd, Gd_2O_3, see Fig.15. The figure shows in forward direction the same detector arrangement, which is foreseen as a possible upgrade path to include the small Q region within the natural properties of the diffractometer. The incoming divergence will define the accessible Q-range and absolute Q resolution to 0.01 $Å^{-1}$. This option may serve for powder diffraction studies of nanostructured materials, which simultaneously want to monitor microstructural changes within the available range. The detector configuration gives convenient access for sample environment from the top, bottom and one side.

[We like to note that based on the same structure, there is an attractive alternative (patent filed) using BF_3 at normal pressure as a detection gas. This can raise the detection efficiency near to 100% with similar high spatial resolution and it will save the costly B-10 sputtering process. These are promising prospects for future instrumentation, however, licensing, prototyping and testing is needed, which makes it an unlikely technology for the first ESS instruments.]

Two monitors in the evacuated flight path before and behind the sample for wavelength dependent calibration. (Cascade-Gem-type: gamma insensitive, position sensitive 1 to 2 mm, absorption ~ 10^{-4}, high dynamic range).

Background.

Note, the new volume detector itself offers completely *new opportunities* to improve the signal to background ratio. Consider two signals detected nearby in a detection volume, see scheme (left) below, originating for a shield 5 cm from the center and from the sample itself. The projection and integration along the detector depth of all detected counts yields a sharp signal only from the sample, while the spurious signal from the shield will be smeared out. In addition, the 3D data can be cleaned up by analysing the characteristic signals with ray tracing to discriminate to non-sample-intrinsic background. We call this the ***intrinsic collimation*** property of the volume detector. The second new opportunity is to use the 4D information with time and wavelength correlation to analyze and to compare the absorption profile of a group of signals with the expected absorption profile, which will exhibit possible deviations from elastic diffraction due to for example parasitic inelastic background from incoherent hydrogen scattering. We call this the ***intrinsic energy sensitivity*** of the volume detector. The development of appropriate software tools for such an advanced data processing will be an important task of the DMSC.

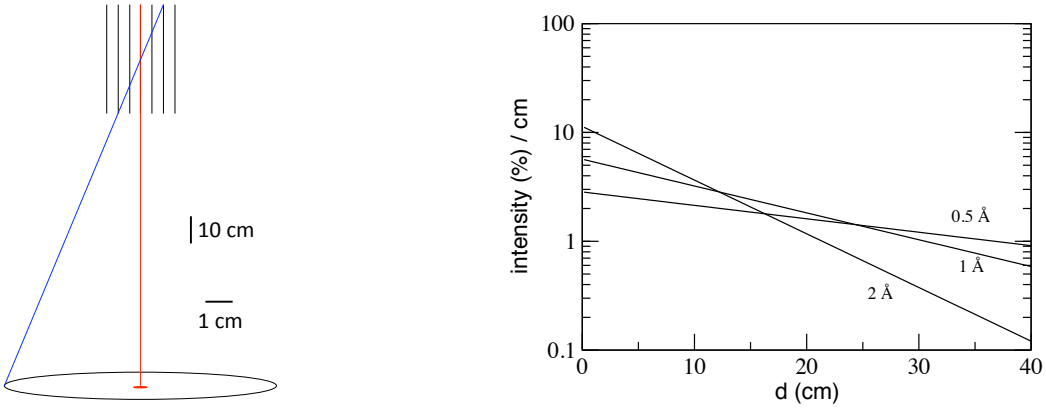


Fig. 16. (left) intrinsic collimation and (right) intrinsic energy sensitivity via wavelength absorption depth profiles.

Within the conventional measures taken to minimize background there will be an optional vacuum vessel to house standard sample environments by top-loading. The sensitive detectors are housed in protective shields, whose volumes can be flooded with Ar to reduce scattering on the secondary flight path. The volumes can be extended to the vacuum vessel, and their side walls will be covered by neutron absorbers providing additional coarse collimation between the detector elements. Alternatively, a radial oscillating collimator can be set around the sample. Instead of using a beam stop in the experimental area, the transmitted beam is kept in an evacuated flight path. It will be captured by a "get lost pipe", a neutron guide behind the sample, and can be guided to a possible second experiment station. This could be, for example, a neutron Laue instrument like FALCON from HZ Berlin, as proposed (IKON-4 G. Iles).

Sample geometry

The focal spot size homogeneously illuminated by the neutron guide system is 1 cm^2. For the simulations we choose a sample shape of an cylinder of 1 cm diameter, with its axis common with the beam axis and a thickness (height) of 5 mm in beam direction. This shape is optimal for reducing absorption and multiple scattering effects, to adopt for low divergence near 90 degree scattering angles, and to relax divergence requirements for backscattering.
In practice, a typical sample will be top-loaded either with a cylinder shape vertically (d=0.5mm, h=10mm) or more favorable with d=10 mm perpendicular to the beam and only squeezed in beam direction to 5 mm, which as V-cans of 0.15 mm wall thickness are available (Metal Technology Inc.). There is also access from the bottom and one side for any specialized sample environment from users.
It can be expected for a number of important cases that the sample geometry will be optimized only for the backscattering and forward detector. Theses cases are (i) using chemical reaction cells with shadowing wall materials perpendicular to the beam, (ii) use of optional polarization and polarization analysis with He-3 filter cells in and near the beam axis for separation of coherent and incoherent scattering.

Sample environment, Michael Meissner

The instrument will be prepared to accommodate standard sample environment (SE), which allow to remotely operate a central axial sample stick where the mounted sample(s) can be precisely rotated and vertically positioned accordingly to the neutron beam cross section.

Dedicated for the instrument we envisage a PTR cryostat (see Fig 17. a,b). Such a prototype of is under commissioning phase at ISIS. Commercially produced at Oxford Instruments and designed in cooperation with the ISIS sample environment team (see Ref. Kirichek et al., JLTP 30-01-2013), the Orange-type cryostat provides cryogen-free operation to the user in a temperature range T = 1.4 – 300 K. Inserted into the vacuum chamber (see Fig 17. c), the sample is surrounded by a d ≃ 1 mm thin walled sample tube (dia. 75 mm) and the thermal shield (dia. 150 mm) of even smaller thickness, both cylinders can be made from 'neutron friendly' Al-alloy.

Furthermore, we provide for work up to ~2000°C an instrument specific ILL-type high temperature furnace.

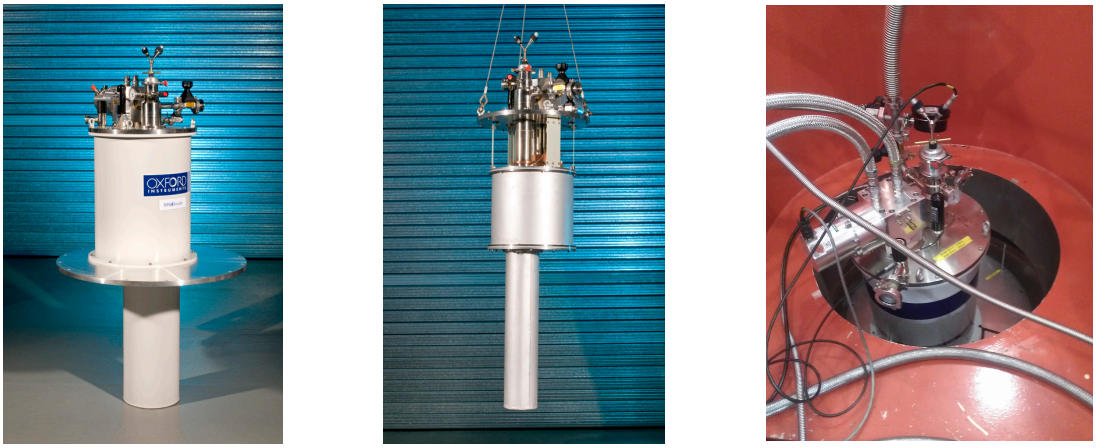


Fig. 17. (a) FPTR 'Orange' cryostat (b) w/o vacuum housing (c) inserted to detector chamber.

On top of the DREAM detector housing, a vacuum flange will allow mountable access for the PTR cryostat or for the high temperature furnace.

ESS provided sample environments will also be used. For experiments with the PTR environment below 1.5 K, low temperature inserts (so-called ^{3}He sorption or ^{3}He/^{4}He dilution 'stick', with T_{min} ≃ 0.3 K or 0.03 K, respectively) will provide well established performance. The central axial sample stick access will be standard with superconducting magnets cryostats, as well. For **magnetic fields** up to ~10 T, again PTR cooling technology provides cryogen-free operation; whereas for magnetic fields strength up to ~15 T a liquid helium bath today still is standard technique (zero boil-off accomplished by PTR re-condensing).

Given the sample environment geometry sketched above, powder samples contained in thin-walled vanadium cans or glass capillaries (wall thickness ~200 … ~20 µm) can be performed

on a wide range in temperature and magnetic field. Using 'neutron friendly' gas pressure cells, powder material under variable pressure (p_{max} ≃10 kbar), can be studied in the same {T, B} range.

For automated measurements with canned samples a robot arm technique will be designed (on top of the detector housing platform) capable to change sample cans at the bottom end of the sample stick followed by an axial transfer into the SE device via an automated double-valve air-lock systems. Given a stick length L ≃ 2 m (top-to-sample) a coarse-to-fine vertical positioning of a powder sample should be possible. For a temperature range T ≃ -100°C to 300°C, rapid sample changes based on this outside operating robotic system could allow transfer, positioning and thermalization of the sample can within < 5 minutes. For work at low temperatures (T < 200 K), aiming for comparable sample change time below 5 min., a robot sample storage & changing technique operating inside the cryostat / cryomagnet should be applied.

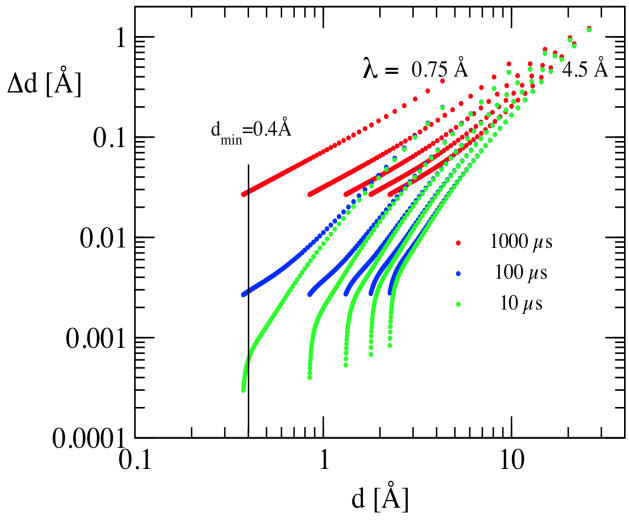
References: O. Kirichek et al. Top Loading Cryogen Free Cryostat for Low Temperature Sample Environment, J. Low Temp. Phys. 170, DOI 10.1007/s10909-013-0858-x

Calculated resolution

The resolution of the instrument is determined by time resolution, incident divergence, sample size and geometry, sample to detector distance and spatial resolution of the detector varying with wavelength and diffraction angle, see Fig. 18.

Fig. 18. Resolution *Δd vs d,* as a function of pulse width, wavelength and diffraction angle.

low *small unit cells*
diffuse scattering
medium *rapid measurements*
small samples
high *Δd /d < ~ 0.001*
in backscattering



Data structure, analysis and software.

An important part of the instrument is the data definition and analysis. Data will be stored necessarily in event mode as a function of 3D space and time and further parameters related to external parameters (sample environment) and variable instrument configurations. As mentioned previously, the aim is to analyse the diffraction pattern in a 2D dependence on wavelength and scattering angle to benefit from the largely varying resolution function. This approach is still new and under development for the *POWTEX* instrument at FRM-II. We expect that this desirable and advanced data analysis will be almost a routine standard in 2019, the earliest possible time of operation for **DREAM** at the ESS.

A rapid integration is possible along the azimuth angle φ and projecting through the finite depth of the detector, which yields intensities (λ,2θ), in live display and for data analysis "on the fly".

In order to demonstrate the method and recent progress, we show a figure from P. Jacobs et al., showing simulated data for POWTEX and a 2D Rietfeld refinement. The refinement provides a visualization in all variables 2θ, λ, d, t, Q. For comparisons, see Fig. 5 and 19: The data from DREAM will cover a substantially larger wavelength band of 3.8 Å, and this with better resolution and significant higher intensity. In addition, time resolution will be largely flexible at DREAM.

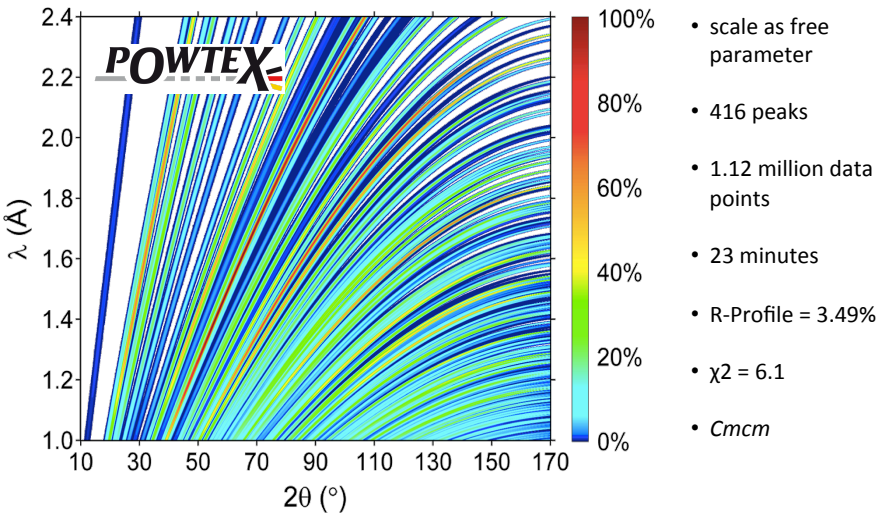


Fig. 19. Wavelength and angle dispersive 2D Rietveld refinement. The example is based on data of CuNCN as simulated by VITESS for the new instrument POWTEX at FRM-2, MLZ Munich.

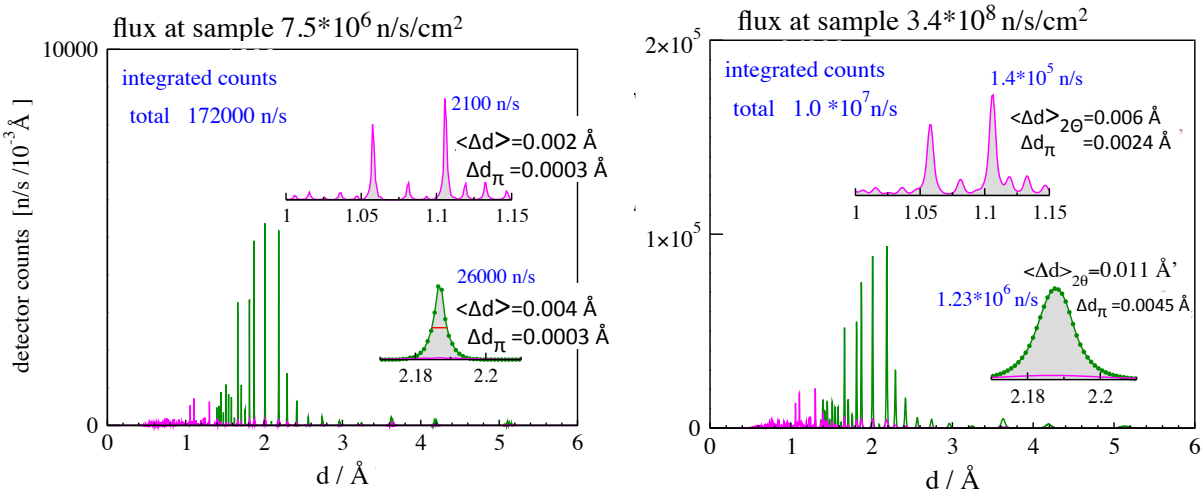
*Benchmarking **DREAM** (POWHOW) with D20, POWGEN and WISH*

As recommended by the STAP, we have made comparison to world leading neutron powder diffractometers build to address similar research areas, the instruments D20, POWGEN, WISH.
We made full VITESS simulations of these instruments and calculated the diffraction from a suitable reference sample $Na_2Ca_3Al_2F_{14}$ cubic ($I2_13$) a=10.257(1)Å (J. Solid State Chem.76 (1988) 426.) Overall the simulations are reproducing well with the known instrumental parameters. The largest discrepancy appears for the high resolution mode of the instrument WISH, where the intensity is underestimated. Here, the flux at the sample is 7.5 instead of $5.8*10^6$ n/s/cm^2 as given in the figure, and therefore we expect that all peak intensities need to be scaled up by 30%. We have to note further, that for sake of easy comparison with the following diffractometers here we use a conventional integration for detector banks. The peak widths are mostly defined by the lower 2θ values for the considered detector bank, which are not set in resolution focusing for DREAM. Therefore, this comparison does not yet exploit the full resolution properties for DREAM. Furthermore, the simulations do not yet include the possibility of shifting towards larger wavelength. Note, there is also a difference in sample volume, which is two times smaller in the case for DREAM. We made these choices, because they reflect the typical sample geometries at the instruments.
The results are displayed on the following two pages. For more detailed comparisons, we give the flux at the sample per cm^2, the total integrated count rate as detected, and integrated counts and resolution width of two diffraction lines, one at d=2.186Å and one at 1.10 Å to probe the performance at for varying wavelength.

Two typical modes of interest have been chosen a high intensity and medium resolution configuration and a high-resolution option. All cases reveal a very strong performance of DREAM compared to these leading existing powder diffractometers with significantly higher count rates at the detector at equal or better resolution. We gratefully acknowledge Pascal Manuel (ISIS), Thomas Hansen (ILL) and Asfia Huq (SNS) for supplying the information on the instruments, which we used for the VITESS Simulations.

DREAM (POWHOW**) ESS** *VITESS simulations, Klaus Lieutenant*

Sample 0.4 cm^3 , efficiency 0.53 (1Å), solid angle 6.2 sr

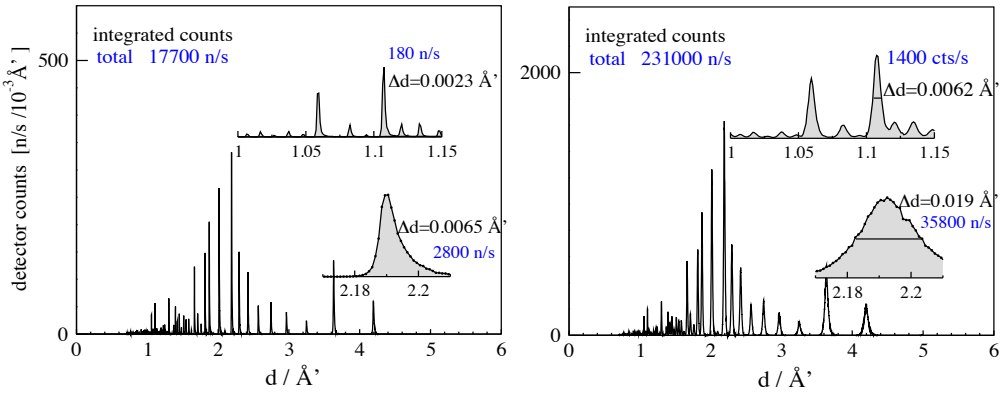


Left: High resolution (10.5µs,31.3µs) 1st λ-frame [0.75Å,2.6Å]

Right: Medium resolution (150µs,350µs) 2nd λ-frame [2.6Å,4.5Å]

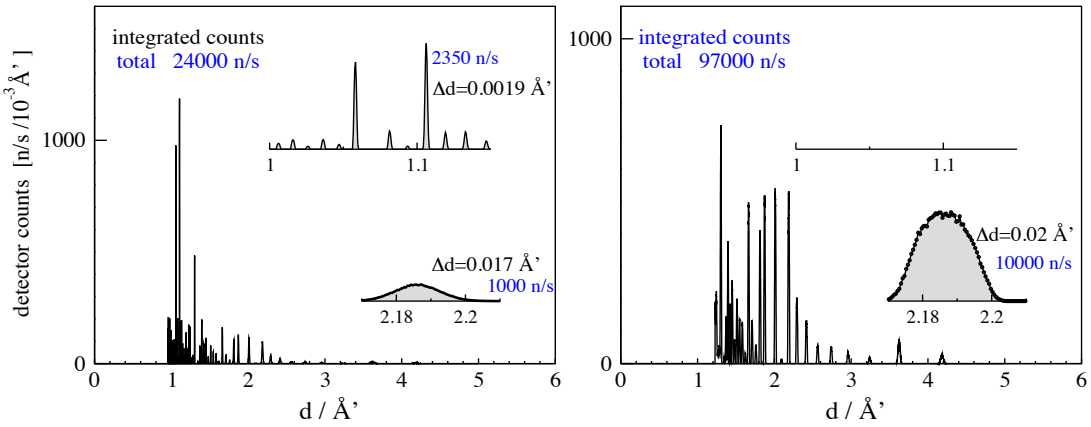
WISH – ISIS *VITESS simulations, Daniil Nekrassov*

Sample 0.8 cm^3 , efficiency 0.64 (1Å), solid angle 0.62 sr
flux at sample $5.8*10^6$ n/s/cm^2 flux at sample $7.0*10^7$n/s/cm^2



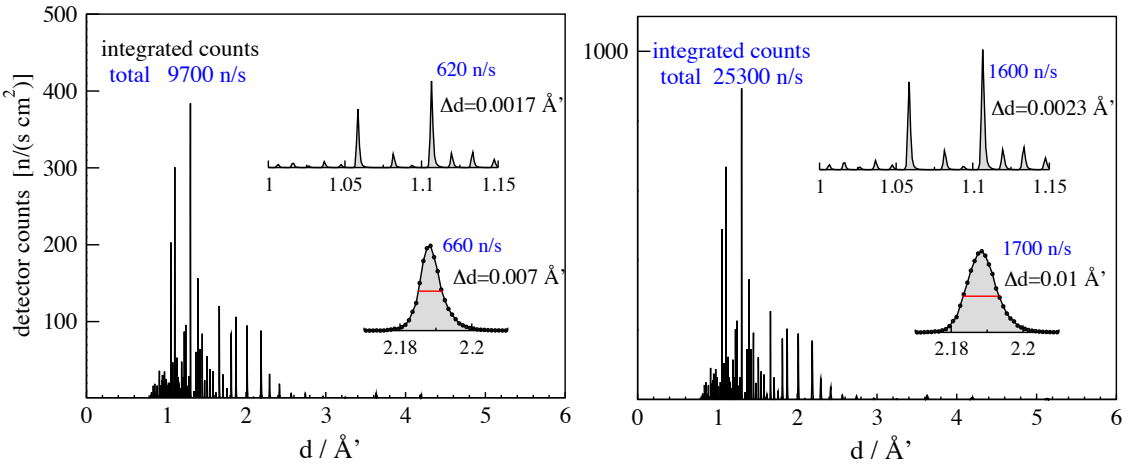
D20 – ILL *VITESS simulations, Daniil Nekrassov*

Sample 0.8 cm^3 , efficiency 0.64 (1Å), solid angle 0.28 sr
flux at sample $1.0*10^7$ n/s/cm^2 flux at sample $4.5*10^7$n/s/cm



POWGEN - SNS *VITESS simulations, Caroline Zendler*

Sample 0.8 cm^3 , efficiency 0.36 (1Å), solid angle 1.41 sr
flux at sample $1.0*10^7$ n/s/cm^2 flux at sample $4.5*10^7$n/s/cm



Complementarity to other ESS instruments

See page 9-10 for discussion of other powder diffractometers. With respect to the cold and thermal magnetism single crystal diffractometers using dedicated polarised neutron beams, DREAM with its large position sensitive detector will serve the needs for unpolarized experiments on single crystals in a complementary way, a need that has been specifically addressed by the STAP for magnetic single crystal diffraction.

Upgrade options from the basic design

Extended detector coverage.

We aim for a large covering of the solid detection angle of 6.2 sr, actually not more to give access to the sample station from bottom, top and one side, as well as to avoid background from detector counterparts. The detector has a modular structure, which allows for a partial implementation of the detector area. In order to mitigate financial risk, we consider for the basic version a possible reduced coverage of only 50% for the forward scattering detector, which assures full functionality and yields 5 sr. In view of the following upgrade options for magnetic studies, the forward detector is important and a complete assembly for optimal efficiency should be given high priority.

Polarization, *Earl Babcock*

Polarization could be another unique add-on to DREAM. The ability to measure magnetic scattering (and subtract incoherent background in possible hydrogen containing samples) would greatly enhance the capabilities. (We asume that the ESS will have ^{3}He polarization resources, which could be utilized for achieving polarization analysis on DREAM.)
Incident beam polarization would be achieved via a ^{3}He spin filter cell which gives a desirable high neutron energy performance and high polarizing efficiencies. A short gap, on the order of 50-80 cm must be left in the neutron guide, via a guide changer, about 3 meters upstream from the sample position. Then after this point the instrument must be made compatible with neutron polarization and should be constructed largely with non-magnetic components and have neutron guide fields plus installable field coils to provide field at the sample position. Since with ^{3}He flipping can be achieved with adiabatic fast passage, no additional components are needed. The ^{3}He neutron spin filter polarizer section will contain a magic-box style magnetic cavity with an integrated RF coil for the ^{3}He flipping. Later, online polarization could be adapted to this place using the initial components.

New developments in wide-angle cell manufacturing have progressed such that all blown C or banana-shaped cells and doughnut-shaped cells can be made of blown GE180 glass[2]. The banana-shaped cells are well suited to analyze the wide angle scattering and placing a doughnut-shaped cell around the incident beam, before the sample, could also enable polarization analysis for back scattering.

If institutional 3He polarization resources are available, they can be used for both parts of the (POWHOW) DREAM polarization and analysis upgrade. Alternatively, since both the polarization and wide-angle analysis 3He cells can be made as SEOP cells, a SEOP polarization system to polarize 3He gas for (POWHOW) DREAM could be envisioned so that DREAM would operate independently when using polarization and/or analysis. In any case, an online SEOP system is envisioned as an upgrade to a continuously polarized (i.e. constant

polarization) incident beam polarizer.

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2. Z Salhi, E Babcock, P Pistel, A Ioffe 2014 3He Neutron Spin Filter cell development program at JCNS, NOPD&D 2012 J. of Phys. Conf. Series, in process

Forward/total scattering

A more dedicated forward scattering detector will expand the Q-range towards small Q, down to 10^{-2}Å^{-1}, which provides a desirable overlap from wide angle to small angle diffraction and fits nicely to the Q-range of interest for *nanoparticles.* If there were only interest in the small Q-region, dedicated SANS instruments will be certainly the only approriate choice, however, the option will ideally serve interests in studying simultaneously structures on atomic- and meso-scale, *id est* the crystal structure, shape and morphology. For *magnetic nanoparticles*, the above mentioned option of an incoming polarised beam with applied magnetic field is straightforward to realize. Furthermore, it is similarly straightforward to realize polarization analysis in forward direction with a ^{3}He polarsation filter.

Sample environment: 10 T solenoid, chemical reaction cells

Dedicated sample environment adds important value to a powder diffractometer and will be needed to address and to extend properly the science case. A solenoid magnet along the beam axis will make best use of magnetic diffraction on the forward detector with Q<9.5Å^{-1}. With its high efficiency the instrument is ideal for parametric in-situ studies with high resolution in real-time. Chemical reaction cells are only one example of valuable sample environment, which would be ideally obtained from user community driven developments and supports.

2nd sample station

Instead of using a beam stop in the experimental area, the transmitted beam is kept in an evacuated flight path. It will be captured by a "get lost pipe", a neutron guide behind the sample, and can be guided to a possible second experiment station. This could be, for example, a neutron Laue instrument like FALCON from HZ Berlin, as proposed (IKON-4 meeting 2013, G. Iles).

1.3 Technical Maturity

(1) **Bispectral extraction switch**

Up today, there is one only device in operation, the one at the reactor in Berlin. There are different and new options to realize this device. Simulations show high efficiency and performance. One choice could be to place the device build as a multistack supermirror in the light shutter, which enables to move it out of beam in case of failure and to work with view to the thermal moderator only. The device will be tested prior to insertion into the monolith. Risk: A failure may occur due to misalignment, degrading of material properties. This risks need to be minimized in prior studies and appropriate technical solutions. A replacement of in-monolith components requires a lot of efforts and time, which we aim to avoid by our concepts. Furthermore, we studied and included in the proposal the option to realize the bispectral switch as a very compact solid-state bender. It shows the best overall performance and could be placed even outside the monolith on a translation in front of the chopper system. The latter option is currently favored in view of performance and has lowest risks.

(2) **Elliptic neutron guide sections**

- The technology exists and is similar to POWTEX (SN) and TOPAS (SDH)

(3) **Chopper**

- The pulse-shaping chopper, band control choppers are all using magnetic bearings, the 75 cm CFC-disc are coated with B-10 in epoxy. The technology exists and is identical to one used for POWTEX (FZ Jülich/MLZ Garching)

- T0-chopper

The proposed design is new, the engineering group at JCNS considers the proposed chopper as feasible with low risk. We are interested in building a prototype in collaboration with the ESS.

(4) **Detector**

The technologies of the B-10 Jalousie detector have been developed by CDT within the POWTEX project and can be used for the DREAM detector.

The proposal to use a BF-3 gas volume detector at ambient pressure is new. A possible application however requires prototyping, testing and (from the ESS side) to be included in the safety considerations of the facility. It promises at lowest costs the best performance.

1.4 Costing

The overall cost is majorly determined by detector costs, and the detector costs are the highest within the project and have the largest uncertainty too. Therefore, we use the conservative estimate and provide two costings for a basic and a complete version of the detector, see tables. The basic version provides the full functionality, though lower efficiency. A preference is given for the backscattering detector.

Integrated design: The effort from the Lead Scientist and Engineer, as well as other scientist and engineers involved in the overall instrument design

Systems Integration: Systems engineering to ensure compatibility between components and compliance with ESS standards

Detector and data acquisition: The detector technology is based on the POWTEX detector, which is currently under construction. Based on this experience it is straightforward to achieve a cost estimate on the same grounds for complete system including electronics and installation. However, the discussions with the company CDT and with ESS detector group identified clearly potentials for cost saving based on new developments. Approximately 30% cost savings are likely only with improved large scale B-10 sputtering technology and further one may expect decreasing electronic costs.

In order to mitigate the financing risk the detector can be build with giving lower priority to the forward scattering detector. Covering only 50% this detector part reduces costs by 1 M€. The dedicated forward scattering detector will cost additionally 420 k€ and is an upgrade option for the nanostructured materials.

Sum: 5.6 M€ + 1.0 M€ +0.4 M€ = 7 M€

Detector housing: This is needed for detector installation and to keep Ar-atmosphere, need not be designed for vacuum.

Neutron guide system: The costing estimate is based on the recent information given by a leading company for neutron optical components. The neutron guide is a double elliptic, straight guide with high m-coating ($3.2<m<7$) optimized to transport neutrons of 0.5 Å wavelength. The costs for the 70m long guide, vacuum housing, support system, installation of the guide system amount to 1.270.000 Euro. Costs for the vacuum pumps and vacuum control systems are estimated to 80.000 Euro. The optical component for the bispectral extraction in the solid-state bender version is estimated to 150.000 Euro.

Sum: 1.500.000 Euro

Choppers: The estimate for the pulse shaping chopper and band control choppers are based on estimates for POWTEX. For the T0 chopper we have quotes for the parts. We expect a participation in development cost by the ESS chopper group in a collaborative work with an uncertainty of 100 k€.

Sample environment: Instrument dedicated sample environment will include an ISISstat Cryo (80 k€), robotic sample changer (200 k€), high-T furnace (60 k€), system integration and controller (80 k€). Sum: 420.000 €.

Shielding: The price is scaled according to the instrument length to the NMx estimate.

Instrument infrastructure: Buildings and facilities not provided by the ESS, such as instrument cabin, floor height adjustment.

Summarized costing for DREAM

in k€	Phase 1 (Design and Planning)			Phase 2 (Final Design)			Phase 3 (Procurement and Installation)			Phase 4 (Beam Testing and Cold Commissioning)			To	
	Hardware	Staff (k €)	Staff (months)	Hardware	Staff (k€)	Staff (months)	Hardware	Staff (k€)	Staff (months)	Hardware	Staff (k€)	Staff (months)	Hardware	Staff (k€)
Integrated Design	0	300	30	0	600	60	0	500	50	0	360	36	0	1760
Systems Integration	0	0	0	0	30	3	0	120	12	0	60	6	0	210
Detectors and Data Acquisition	0	30	3	0	30	3	6600	60	6	20	120	12	6620	240
Detector Vessel	0	0	0	0	90	9	500	60	6	20	20	2	520	170
Optical Components	0	30	3	0	30	3	1480	30	3	20	30	3	1500	120
Choppers	0	60	6	0	60	6	1100	30	3	20	30	3	1120	180
Sample Environment	0	0	0	0	30	3	420	30	3	20	30	3	440	90
Shielding	0	30	3	0	60	6	2100	60	6	20	60	6	2120	210
Instrument Specific Support Equipment	0	0	0	0	30	3	300	120	12	20	30	3	320	180
Instrument Infrastructure	0	30	3	0	30	3	300	60	6	20	30	3	320	150
Total	0	480	48	0	990	99	12800	1070	107	160	770	77	**12960**	**3310**
Grand total (no VAT)								**16270**						
Percentage of total cost		2.95021511985249			6.08481868469576			85.2489244007376			5.7160417947142			
k€/person-month	10													

Costing for upgrade options

I. Polarized neutrons and ^{3}He polarization analysis: total 155 k€.

(i) Initial beam polarization includes magnetic environment, guide fields(10 k€), Control/monitoring electronics and AFP amplifier (10 k€), ^{3}He incident beam cell/with or without valve (10 k€) : 30 k€.

(ii) online polarization upgrade (utilizing above components) +lasers+optics, chiler, heating source : 25 k€.

(iii) PASTIS coil set (40 k€), PASTIS cells (30 k€), SEOP option for polarization (30 k€): PASTIS upgrade 100 k€.

II. Dedicated forward scattering detector for nanostructures: total 420 k€.

III. Solenoid 10T magnet instrument specific: total 350 k€.

For all these three upgrade options, a complete filling of the forward detector is recommended first.

List of Abbreviations

Abbreviation Explanation of abbreviation

PROPOSAL HISTORY

New proposal:	(yes)
Resubmission:	(no)

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