Beamline Name: EXEED

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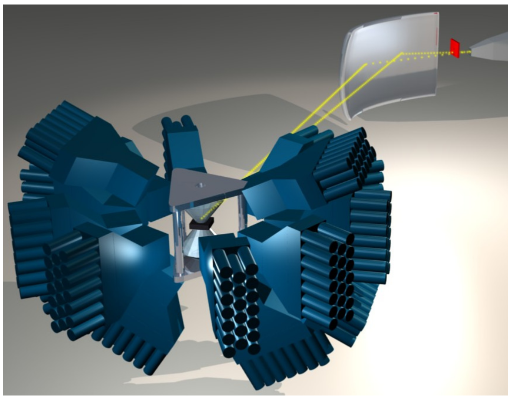
EXEED

An extreme sample environment diffractometer



Beamline	
Moderator	Coupled hydrogen moderator (or possibly methane once experience has been gained on the WISH diffractometer)
Choppers	1 10 Hz frame overlap chopper 1 background suppression chopper
Incident wavelengths	1 – 9 Å
Incident flight path	40 m parabolic m=3 guide, coupled with KB or ellipsoidal mirrors
Diffractometer	
Beam size	Typically 1 x 1 mm^2 and adjustable (3 mm to 100 μm)
d-spacing range	0.7 - 6 Å at 2θ=90°
Resolution	Δd/d ~0.7% to ~3% depending on optics settings
Detector type	Position sensitive detectors (see text)
Sample to detector	~ 1 m
Detector banks	Equatorial: ± 145° horizontal and ± 15° vertical 90° bank: + 30° to - 60°
Sample environment	
High Pressure	Gem anvil cells (>50 GPa, RT and >10 GPa < 5 K)
High Temperature	Container-less levitation (up to 3000 K) and laser heating for gem anvil cells
Magnetic fields	Dedicated 10 Tesla magnet for high pressure experiments

Introduction

eXeed will be a neutron time-of-flight diffractometer optimised for extreme environment studies of materials which will complement the capabilities of WISH on TS2 and PEARL/HiPr on TS1. eXeed will deliver an extremely bright, focused beam in the thermal-cold region, providing world-class access to regions of the phase diagram that have so far eluded neutron studies. This instrument will provide facility users with access to:

- High pressures (above 50 GPa using diamond anvil cells)
- Combinations of extreme environments: very low temperatures (mK) or very high temperatures (2000 K) with high pressure (including laser heating in cells with transparent gem anvils) and high pressures in high magnetic fields (up to 10 Tesla).
- High temperatures and liquid state studies (up to >3000 K) in levitation environments

Technical specifications

A summary of the main features of the instrument design is given in the table on the right. This diffractometer is designed to exploit the high flux of cold neutrons of the coupled hydrogen moderator of the ISIS second target station and uses developments in neutron optics to produce a bright, tightly focused beam of thermal-cold neutrons with tuneable flux and resolution at the sample position. Secondary collimation will be employed to reduce the background signal by means of a removable oscillating radial collimator to define a clean large gauge volume and optimised close collimation to define a small gauge volume at the centre of the sample environment.

A large array of position sensitive detectors will permit studies using powder or single crystal samples. The arrays will consist of two sections: an equatorial bank spanning ± 15° vertically and ±145° in 2θ; and two 90° banks extending vertically to +30° and extending underneath the instrument to -60°. All the detectors will be located approximately 1 m from the sample position and will consist of either pixelated ZnS scintillators or D19 style ^3He area detectors. These will need to be developed to provide the required 1 mm spatial resolution and high detection efficiency. The final technology choice will depend on which option gives the best performance, area coverage and resolution.

Sample environments combining high-P variable T cells and/or high field will need to be mounted on translation (xyz) and rotation stages to ensure the small samples are correctly aligned and centred with respect to the incident beam and detectors, respectively. The alignment process will also require the use of a pixelated transmission monitor with high spatial resolution, such as a PSD neutron camera. A transmission monitor will also be required to obtain information concerning Bragg edges, sample attenuation etc. When in use, both of these monitors will need to be positioned close to the sample to cope with beam divergence.

The ultimate pressure capabilities of the instrument will depend on the pressure cells developed for each type of experiment, with a small sample size determined by the neutron focusing optics. High pressure cells using the Paris-Edinburgh design can already reach 30 GPa at RT and 6-8 GPa at 1800 K. The tightly focused beam down to 100-200 micrometres for diffraction experiments will enable use of diamond anvil cells up to >50 GPa and will permit laser heating experiments. The small and large volume gem anvil cells will provide a wide panoramic view of the sample and lowered attenuation for through-anvil or through-gasket scattering geometries. The highest flux/low resolution configuration will be most suitable for single crystal measurements with the higher resolution being required for powder diffraction measurements of more complex materials.

Science Case

An instrument aimed at extreme sample environments will, by its very nature, have a wide range of scientific applications, ranging from studies of strongly correlated electron systems, through to geology and planetary science. Neutrons give complementary information to X-ray diffraction, Raman and IR spectroscopy, and an optimised instrument that is able to extend the accessible pressure and temperature range will give a greater overlap of sample conditions with the other techniques.

Earth and Planetary Sciences

The centre of the earth is at a pressure of 350 GPa and more than 90% of the matter in the solar system is at a pressure greater than 10 GPa. Pressure shapes the stars and planets, the continents and the oceans. Diffraction gives information on phase transitions, densities, lattice expansion and other vital microscopic and macroscopic quantities. Characterisation of the equation of state and the nature of phase transitions at higher pressures will make it possible to create and validate accurate computational models that can be extended to even higher pressures than can be measured experimentally.

The chemistry of the planetary gas giants (Jupiter, Saturn, Neptune, Uranus) and their larger satellites (Europa, Ganeymede, Titan, Triton) is dominated by the molecular 'ices' water, ammonia, methane. It is critically important to understand the structures and dynamics of these materials at high pressure in order to model the planetary formation, structure and evolution. Studies are now carried out to 30 GPa at existing neutron facilities, including ISIS, using the Paris-Edinburgh press. In future experiments, we must increase that pressure limit by using different or modified cell designs, by increasing the source flux and/or implementing neutron focusing strategies. This will allow us to expand the range of the studies further into the deep interiors of the gas giants, and make closer contact with experiments carried out in diamond anvil cells (DAC) using X-ray diffraction

Structural Physics at High Pressure

Fundamental ices like H_2O, NH_3, CH_4 and H_2S are also important model systems for understanding hydrogen bonding and molecular dissociation. There has been a large amount of recent high-profile work on H-bond symmetrisation and intermolecular bond dissociation in these systems, particularly so for the former case where the pressures are sufficiently high to induce the proton to migrate to the centre of the intermolecular hydrogen bond. This has been demonstrated in ice itself and in the hydrogen halides, where it is accompanied by complex excitation physics. A key problem in relation to centring in ice is the detailed nature of the ice VII phase, which centres to ice X above about 70 GPa. Evidence at lower pressures suggests the centring process may be complicated by oxygen site disorder and there is also some evidence for incommensurate modulation of the structure. In the case of ammonia, it has been predicted that the intramolecular N-H bonds will dissociate above 1500 K and 60 GPa to create a superionic solid. For methane, the full details of the complex orientationally ordered structures found at pressures above 5 GPa remain unknown, and it is an intriguing question as to what the ultimate, very high density methane structure will be. In general, the full H-bonding structures of NH_3, H_2S and the hydrogen halides – where strong H-bonding can be induced under pressure – need to be determined for above 10 GPa, and followed up into the realm characterised by bond centring and dissociation.

Strongly Correlated Electron Systems

Strongly correlated electron systems continue to surprise and delight in displaying novel and exotic ground states, some of which can be truly classified as new states of matter. Understanding the origin and nature of these states and in particular the phase transitions that occur between them, present significant challenges, thus explaining the central role occupied by the study of such systems in contemporary condensed matter and materials physics. Key to developing our understanding of strongly correlated electron systems is the ability to tune systems through phase transitions as a function of applied pressure, magnetic field, etc. The construction of eXeed will provide the large and diverse community of experimentalists working on strongly correlated electron systems with an unrivalled combination of instrument specifications and extreme sample environments, allowing ISIS to remain competitive in an area where other facilities are currently making large investments.

New Materials from High Pressure Synthesis

The search for new technological materials involves pioneering research at extreme high-P,T. Synthetic diamond and cubic-BN, prepared under high-P,T conditions are the hardest known materials that give rise to multi-million $/yr industries for cutting and grinding tools. Research to discover new superhard materials involves high-P,T studies of diamond-related phases in the C-B-N system and boron carbides and suboxides. These grow with an unusual icosahedral morphology in the 1-3 GPa range from B_2O_3-rich melts. There is intense interest in high-density sp^3-bonded C_3N_4, predicted to be harder than diamond. Research into synthesis approaches is being carried out at P > 30 GPa, where (C, N, H) molecular precursors react to form new high-density phases. In situ neutron diffraction under combined high-P,T conditions in the 1-50 GPa range will revolutionise these studies. At lower pressures (0.5-2 GPa range), new layered $(C,N)_x$ graphite-related materials are formed from heterocyclic aromatic precursors; the intercalation and ion-exchange chemistry of these materials must be established using a combination of techniques including neutron diffraction at high-P,T.

Amorphous diffraction under high-P,T conditions; liquid-liquid transitions and polyamorphism

Silicate magmas are the best known manifestation of volcanic activity. Today, it is likely that most magmatism occurring at depths down to 100 km below mid ocean spreading centres and at subduction zones. In the past however it is likely that a global magma ocean existed throughout what is now the mantle. Understanding melt density and rheology requires structure studies at pressures up to at least ~30GPa and T~1500℃. These conditions are already achievable with existing high-pressure cells adapted for neutron scattering. The eXeed instrument will increase the pressure range to P~50 GPa where large structure and coordination changes are observed among crystalline minerals. Technological glass-forming melts are similar to magmas: here the primary interest is in studying the refractory liquids in situ using levitation techniques. A levitator system will be developed for eXeed for studies up to T~3000K. Crystalline solids undergo large changes in their structure and properties as they transform between different polymorphs as a function of P and T. It now appears that analogous density- or entropy-driven phase transitions also occur within liquids at constant chemical composition. These represent a new family of thermodynamic phenomena within the liquid state. These "polyamorphic" transformations are also reflected within the solid amorphous state as dramatic structural changes as a function of P and T. The eXeed instrument will allow new studies of L-L transitions and polyamorphism in systems ranging from elements (Si, P, Se etc) to compounds (H2O, SiO2) and complex ceramics (Y2O3-Al2O3) as a function of T in levitation environments or at high P inside Paris-Edinburgh or diamond anvil pressure cells. The instrument will also permit new studies of pressure-induced amorphisation, where crystals collapse under conditions of increasing density, as well as similar collapse transitions in metastable low-density phases such as zeolites.

This text is based on the contributions of D McMorrow (UCL), RJ Nelmes (Edinburgh), PF McMillan (UCL), A. Harrison (Edinburgh), B Lake (Ames, USA), B Fak (CEA, France), M Wilding (Aberystwyth) and PG Radaelli (ISIS) in the original eXess case.

Business case

High pressure neutron diffraction experiments are already carried out at ISIS using PEARL/HiPr but these are limited to a maximum pressure of ~9 GPa. The new eXeed instrument will be developed around a focusing capability that will allow us to fully exploit the maximum pressure range available with Paris-Edinburgh type pressure cells (P~30 GPa at ambient T and low T; P~6 GPa at T=1800K) and will allow us to begin to use diamond anvil cells (up to P=50 GPa), with possibilities for laser heating to T~3000K. These new capabilities will greatly expand the user base.

A large amount of the extreme conditions science currently being carried out at ISIS would benefit from having the accessible pressure and temperature range increased. It is envisaged that the existing users of ISIS would also use eXeed to extend their studies. Due to the complementary nature of eXeed and existing TS1 instruments, studies that use the TS1 machines to explore lower pressures in greater detail (due to the higher resolution and larger d-spacing range) which could then use eXeed to obtain information at higher pressures albeit at a lower level of detail (due to the flux profile), are extremely likely.

Most high pressure diffraction studies are presently carried out at synchrotron X-ray facilities: a Web of Science search for ‘high pressure’ and ‘x-ray’ gave nearly 2000 hits for 2006-2007 whereas a comparable search for ‘high pressure’ and ‘neutron’ returned only 405 hits. This difference is primarily due to the available pressure range at neutron facilities. That will change with development of the eXeed instrument and allow user groups to take advantage of the unique capabilities of neutron diffraction for high-P structural studies.

The exploitation of the complementary between neutron and X-ray diffraction will become a distinct advantage as these studies proceed and we expect that outstanding structural and phase transition problems will be resolved. This is particularly relevant at RAL due to the due to the presence of the 3rd-generation synchrotron source Diamond, that is already being accessed by new families of UK and international users for high-P diffraction experiments.

For single crystal experiments it is intended that the flux gains on eXeed will be enough to make standard diamond anvil cell (DAC) experiments possible with neutron diffraction. Such a capability would make it extremely attractive for existing X-ray users to carry out experiments on eXeed at ISIS, perhaps even on preloaded samples in DAC's which they may have already studied at X-ray synchrotrons or their home laboratories. Furthermore, this would remove the barrier of special sample preparation for neutron experiments, which is one of the potentially limiting factors for attracting experienced X-ray source users.

Exploring the extremes of temperature and pressure simultaneously during neutron diffraction experiments is another under-exploited area of science at ISIS. The ability to use the physically smaller gem anvil cells would make experiments using pressures in excess of 10 GPa at temperatures down to the millikelvin range – using technologies such as pulsed-tubed coolers – a real possibility; also the combination of gem anvil cells with a laser heating system would make extreme temperatures and pressures attainable in a neutron diffraction experiment for the first time. This would make possible a whole new range of experiments and thus attract a new community of facility users.

The various user communities outlined above, both new and existing, come from within and outside the UK. The only other planned machine with similar capabilities to eXeed is the SNAP diffractometer at SNS. While the SNS source has a higher intrinsic neutron flux, for the type of sample geometry and neutron optics required for large volume gem anvil experiments, the TS2 coupled moderator will give a comparable flux at the sample on eXeed to that predicted for SNAP. We anticipate that the international user community is large enough to support two machines.

While the flux at the sample on eXeed will be much higher than the TS1 instruments (at the longer wavelengths), the small sample size and complex sample environment conditions will mean reasonably long experiments will still be required. If a typical experiment is 5-7 days, that would give ~19 – 26 experiments a year (based on ISIS delivering 130 user days per year). The instrument will therefore serve ~50 proposals a year from ~25 active groups (depending on proposal oversubscription and number of proposals/group/year). The anticipated user base will be at least as large as this, if not larger, and will ensure that eXeed is viable.

Instrument developments

Neutron Optics

The elliptical and parabolic guides used for the initial McStas simulations (see figure 1) can be manufactured, although they are only just being installed on neutron instruments. Indeed, WISH will have probably the most complicated and optimised guide of this type built so far, certainly at ISIS, if not in the world.

An attractive alternative to a full beamline guide is a shorter guide producing a virtual source, coupled with pinhole and neutron mirror combinations. There are two main types presently being considered for neutron applications. The first is based on the K-B mirrors used on X-ray synchrotron beamlines. This system has been prototyped by the SNS SNAP team at Chalk River and appears to be their preferred option for producing intense sub-millimetre beams. But as yet this system has not been tested in a real high pressure neutron diffraction experiment, so its true capabilities remain unknown; also this technique is intrinsically limited to sub-millimetre sized beams. The second option would be to use a mirror that is truly ellipsoidal in two dimensions. This could then be used to produce an image of a small pinhole source at the sample position with almost no loss of flux at the longer wavelengths. This option is only at the conceptual stage at present and the capability to physically build such a mirror is being explored. New Monte Carlo simulations are providing quantitative measures of the expected flux distribution in comparison to the other neutron optics options. However, with this technique the size of the beam should only be limited by the size of the pinhole source, which is tuneable by using a carrousel of different pinhole sizes, providing one clear advantage over the K-B mirror setup.

The components to simulate the neutron optics concepts described above have been developed by the McStas and ISIS groups; approximately a month or so of simulations would be required to determine the most flexible and optimised instrument for extreme environment applications. It would also be very useful to produce a physical prototype of the ellipsoidal mirror system to test its capabilities on an existing beamline

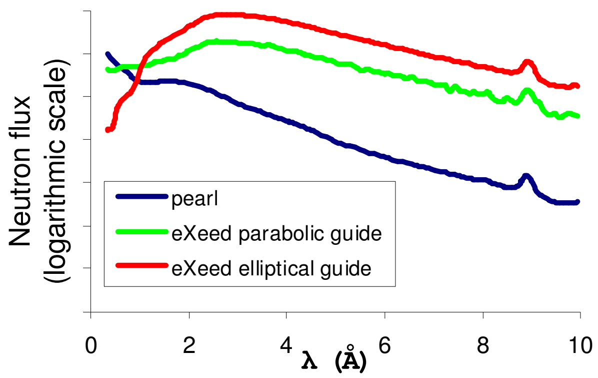


Figure 1: A comparison between the flux on PEARL and eXeed for the elliptical guide as employed on WISH and a simple parabolic guide.

Precise Sample Positioning

The neutron optics described above will produce a beam of varying size, flux distribution and possibly even position. To ensure that the maximum benefit is gained during a given experiment, the sample will need to be very carefully positioned and oriented with respect to the incident beam. The wide variation of sample environment equipment and sample size will make this a non-trivial task. It is envisaged that a system combining a precise xyz translation and rotation stages, a PSD neutron camera and a transmission monitor will be the most versatile and reliable solution. Similar systems using photodiode detectors are routinely employed for this purpose on X-ray synchrotron beamlines; however, at present no standard system exists for neutron beamlines.

Position Sensitive Detectors

As mentioned in the introduction, eXeed will require a large array of position sensitive detectors. At present the two viable detector technology options are an array of standard ZnS scintillators or ^{3}He area detectors similar to those employed on D19 at the ILL. Both of these technologies would need to be developed further to produce the pixel size, coverage and count rate required on eXeed for both single crystal and powder diffraction measurements, coupled with the required resolution. Ultimately this would also require prototypes to be constructed and tested, to provide 1 mm pixel resolution with high efficiency in the 1 – 9 Å wavelength range.

Sample environment developments

As the power of neutron sources increases and instrumentation improves, more and more experiments are being performed using extreme environments. A survey of Nature and Science papers shows that a large majority of the neutron scattering papers use some form of extreme or otherwise non-standard environment, indicating that facilities able to perform such measurements will attract high profile science.

The design of the proposed eXeed instrument is such that it will be suitable for studying a small gauge volume within a relatively much larger item of sample environment equipment. This could be a laser-heated diamond anvil cell, a hydrothermal ceramics processing furnace or even an operating fuel cell. Although it is impossible to predict exactly what such a versatile instrument will be used for, the following sections provide several examples of possible extreme sample environment scenarios that, if developed, will be most suited to experiments on eXeed.

High pressures

Figure 2: VX 5 Paris-Edinburgh press equipped with gem anvils

The development of the Paris-Edinburgh press at ISIS has made neutron diffraction at high pressures routine for pressures up to ~9 GPa. This is a high profile and still growing area of science, with many important structural studies requiring access to ever higher pressures, often in combination with a wide range of temperatures. Such experiments will inevitably require the use of sample volumes significantly smaller than the ~100 mm^3 typically employed in the Paris-Edinburgh cell. The use of a highly focussed incident beam on eXeed will give a significant gain in neutron flux over PEARL/HiPr at wavelengths above 1 Å (see Figure 1).

The existing Paris-Edinburgh cell is capable of reaching 30 GPa at room temperature and below (~100 K), or 7 GPa at 1800 K using the internal anvil furnace system. But with the smaller beam sizes available on eXeed it is likely that we will be able to use single crystal diamond anvil cells for diffraction studies, so pushing the ultimate pressures towards 80 GPa. These cells are currently being developed elsewhere (as illustrated in Figure 2) and the ultimate performance of eXeed in this respect will depend largely on the capabilities of these new pressure devices. Clearly, the ultimate success of this project will depend on ISIS forming a close relationship with the developers of these devices.

Extreme temperatures at high pressures

With the long wavelength flux available on eXeed this instrument will be particularly suited to studying magnetic materials at high pressures. The development of a system to cool a high pressure device in a cryostat has been funded by the Facility Development Advisory Board and this will mark the first step to performing high pressure experiments at milliKelvin temperatures. The design outlined will be suitable for use on eXeed, although it will need to be further developed to obtain the lowest temperatures required.

The ability to heat samples while under pressure is extremely important to both exploring p,T space and to help maintain hydrostatic pressure conditions at pressures above 15 GPa. The use of smaller gem anvil pressure cells on eXeed will allow existing laser heating systems to be used to extend the high temperature range available for high pressure experiments. While this technology already exists, combining it with the constraints of other sample environment and the instrument will require some development effort.

High magnetic fields at high pressures

Studies of materials exposed to high magnetic fields and relatively low pressure will be most suited to the WISH diffractometer. However, the ability to use small gem anvil cells on eXeed will mean experiments using these cells under magnetic fields at low temperatures and high pressures would be possible. For these studies a small high-field magnet could be developed which would obviate the need to compromise the secondary flight path of the diffractometer. Initial experiments could be carried out using the existing GEM 10 Tesla magnet if the pressure cell characteristics are compatible, though to extend the capabilities a dedicated system would need to be developed.

High temperatures

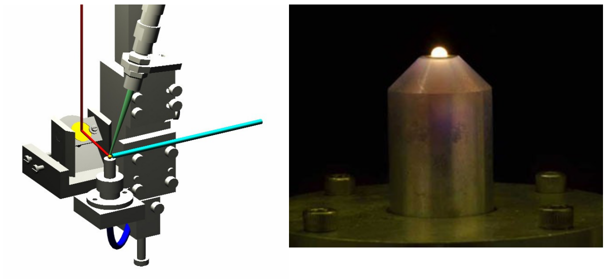


Figure 3: Container-less sample levitation system

Neutron diffraction using container-less sample levitation in an inert gas stream has been performed successfully several times (as illustrated in Figure 3). It is anticipated that the beam focusing on eXeed will make these experiments an order of magnitude faster. The samples used in these kinds of measurements are of the order of 2 – 4 mm across and are well matched to the beam size of the proposed instrument. Temperatures of the order of 3000 K will be possible and there will be provision for collecting diffraction data under different atmospheres. By using fast high-resolution optics it is also possible to extract complimentary viscosity and thermal expansion information.

A suitable system for use at ISIS would need to be acquired and probably developed further to be suitable for use on eXeed.

The ISIS team that has produced this outline case consists of:

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