

Diamond Light Source

Outline Proposal for a Phase III beamline

Mag-X: a dedicated beamline for diffraction and spectroscopy in high magnetic fields.

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1. Summary

We propose a medium-hard-X-ray beamline (undulator source, 3.5-30 KeV) for diffraction and spectroscopy in steady-state and pulsed magnetic fields up to 17 Tesla and 60 Tesla, respectively. The spectroscopy component will employ both a conventional monochromator (steady-state fields) and an energy dispersive bent crystal to focus and disperse a polychromatic beam onto the sample (pulsed fields). The diffractometer will employ a horizontal geometry, accommodating boosted magnets up to 17 Tesla. Bragg and anomalous scattering experiments will be performed in both single-shot and low-rep-rate pulsed mode. We will continue to develop ultra-stable high-rep-rate pulsed magnets (~1 Hz), which will enable magnetic scattering measurements on single-crystal to be performed. An *alternative plan* to achieve some of the same objectives by upgrading I16 and building a new bending magnet based beamline is also described. The main scientific areas to be addressed are that of *complex spin textures*, *quantum magnets* and *unconventional superconductors*. One of our main targets is to enable measurements on crystals that are too small for neutron scattering and, eventually, on high-quality grains of 100 μm on edge that can be extracted from powder batches. This, coupled with the availability at ISIS of the best neutron magnetic powder diffractometer in the world (WISH), will vastly expand the range of accessible model materials, and provide the strong UK community with a distinct competitive advantage.

2. Scientific Case

Following the first successful observation of X-ray magnetic scattering in 1971 [1], this field of research was successfully launched in the early eighties with a series of detailed studies by de Bergevin and Brunel and Gibbs and co-workers [2]. Less than two decades later, X-ray magnetic scattering was already considered a standard technique for the study of magnetism and magnetic materials, with a strong element of complementarity with neutron scattering [3]. Although limited to antiferromagnets with relatively large magnetic moments (due to the weak signal) and difficult to carry out at very low temperatures (due to beam heating effects), X-ray magnetic scattering, in both resonant and non-resonant variants, has many advantages:

- It can be performed on much smaller crystals than for neutron scattering. Millimetre-size crystals are already routine, as are measurements of non-magnetic resonant scattering from crystals as small as 50 μm [4]; magnetic signals from crystals of 100 μm or less can be often measured (our data), the main difficulty being the mechanical accuracy of the diffractometer.
- It can be thought as combining the advantages of neutron scattering and neutron polarimetry. Several projections of the complex magnetisation vector can be measured on the same Bragg reflections by rotating the incident or final polarization and/or the azimuth of the sample. Furthermore, by employing a circularly polarised incident beam and final polarisation analysis, it is possible to distinguish the signal inversion-related domains (“chirality” domains)[5].
- Uniquely, it provides information about multipolar ordering processes, which are essentially inaccessible to other techniques[6].
- It provides element-specific information, which is extremely helpful in solving complex multi-site magnetic structure, and to probe distinct order parameters.
- The extremely high resolution in reciprocal space yields an excellent accuracy on the determination of the magnetic propagation vector important to distinguish between commensurate and long-wavelength incommensurate structures.
- High real-space resolution, particularly with focussing, can be employed to image magnetic domains of different kinds, including chirality domains [7,8].

The level of maturity of this technique has been highlighted by the recent research in magnetic multiferroics (the literature is vast, but, for example [8,9]). This is an ideal subject for X-ray

magnetic scattering, since these materials are complex antiferromagnets with propagation vectors at the zone interior or zone border, and have chirality domain populations that can be manipulated with external electric fields. Further afield than the specific research area of multiferroics, there is evidence of a growing interest in **complex spin textures** developing at relatively high temperatures, and their potentially functional properties for both information storage and spin transport (the skyrmion lattices in metallic MnSi and related compounds is an example of the latter [10]). Another important area of strong growth is that of **quantum magnets**. These materials have been traditionally chosen among $S=1/2$ 3d transition metal compounds, which were until recently considered inaccessible to hard X-ray magnetic scattering. However, on I16 we have recently been able to perform azimuthal scans, even in *non-resonant* conditions, on Cu^{2+} $S=1/2$ materials with an ordered moment as low as $0.6 \mu_B$. Moreover, there is a recent trend towards 4d and 5d compounds with strong spin-orbit interactions; Sr_2IrO_4 [11] and Na_2IrO_3 [12] are two important examples in which the neutron signal is exceptionally small or absent, whereas the X-ray resonant signal is very strong. Crystals are at present too small for neutron scattering, and hard X-ray diffraction can exploit strong resonant enhancements to observe phenomena that are inaccessible to neutrons. Also in the field of frustrated magnets novel phases with complex order can be stabilized at high magnetic fields, for example a cascade of magnetic transitions [13] is observed (via torque measurements on small $100 \mu\text{m}$ diameter crystals) in the frustrated triangular magnet AgNiO_2 , potentially showing magnetic analogues of “supersolid” phases [14], again inaccessible to neutrons due to the small crystal size. A third, important class of materials is that of **unconventional superconductors**, many of which are generally bound by magnetic phases and strongly affected by magnetic fields.

In order to have a consistent impact in these research areas, it is imperative to be able to apply relatively large magnetic fields. In fact, the phase diagram of existing and, most likely, future materials in these classes are strongly affected by magnetic field, and their field behaviour is a keen indicator of the fundamental physical mechanisms at play. What is of interest here is not only the evolution of the magnetic scattering, but also of the lattice parameters (related to magneto-striction) and of the lattice modulations of magnetic origin giving rise to the so-called “2q” satellites, as well as the response of the local structure. Essentially all neutron diffractometers with a magnetism programme have a dedicated magnet, often with maximum fields $> 14 \text{ T}$. This is used for a large fraction of the beam time, sometimes in combination with electric fields or pressure, enabling multi-dimensional phase diagrams to be mapped.

In the case of X-ray scattering, the application of magnetic fields requiring a cryomagnet is still relatively uncommon. In Europe, 4 hard X-ray beamlines currently have this capability: XmaS (ESRF, maximum field 4T), MagS (Bessy, 5T), ID20 (ESRF, soon to be decommissioned, 10 T) and P09 (Petra III, 14 T, magnet still under commissioning). The main difficulty with these experiments is the need to position and orient crystals within a large cryogenic environment on a heavy-duty diffractometer. Although this can be overcome with a large and highly homogeneous crystal, the need for such samples removes perhaps the greatest advantage of X-ray scattering over neutrons. **The first main objective** of this proposal is to provide *facile* X-ray magnetic scattering and other techniques in applied steady-state fields at least of the order of 10-15 T with crystals that are too small for neutron scattering. An initial, very accessible target is 0.1 mm^3 (with a $1 \times 1 \text{ mm}^2$ crystal face), but we believe that crystals of $100 \times 100 \mu\text{m}^2$ can be measured, by appropriately engineering the diffractometer, sample environment and software. High-quality crystals of this size can often be obtained from a polycrystalline sample, and the ISIS facility on the RAL campus has the best magnetic powder diffractometer in the world, WISH, equipped with a 14 T magnet. Powder diffraction is also used to determine propagation vectors – an essential prerequisite for a successful X-ray experiment. WISH and Mag-X on the same campus will provide a unique set of capabilities, opening up new materials and new science that cannot be tackled anywhere else. **The second main objective** of this proposal is to provide access to higher fields in pulsed mode for both diffraction (conventional and magnetic) and energy-dispersive spectroscopy (determination of the electronic state and local structure at high fields). Superconducting magnets and resistive magnets can provide dc fields up to 25 T and 35 T respectively. Both types of system require large scale infrastructure if they are to be incorporated into X-ray beamlines, and resistive magnets in particular require significant and continuous

electrical input (~20 MW) to keep cool and to run. In contrast, capacitor-driven pulsed magnets provide an economical method of achieving 60 T fields of millisecond duration. Such magnets can be small enough to fit inside the cryostat with the sample, are readily configurable to different measurement geometries, and require minimal ancillary infrastructure. Obviously, the transient nature of the applied field precludes the possibility of some types of experiments; however the high intensities and collection rates at synchrotron facilities allows for the possibility of using pulsed fields to perturb quantum materials while investigating their response to X-ray radiation via a number of techniques. Pulsed fields have already been used to successfully measure single-crystal and powder diffraction, XAS and XMCD at SPring-8, ESRF and, most recently, the APS.

For the SAC's consideration, we indicate two possible ways to achieve these objectives. The first, as the title indicates, is a **dedicated beamline** for diffraction and spectroscopy in high magnetic fields. The main advantage of this approach is that it will increase capacity, compensating for reductions in the same area at ESRF. Furthermore, the scientific and engineering staff in charge of the beamline design and operation will be able to concentrate single-mindedly on the pursuit of a highly profitable area of science, without the distractions associated with a multi-purpose beamline. As explained above, the technique is very well established, so the obstacles to overcome are essentially of a technical and engineering nature. Furthermore, the UK community in this field is strong and faces at present a dramatic loss of capacity elsewhere (see below); a well-designed, well-run beamline focussed on delivery will be constantly busy and heavily oversubscribed, with a steady output of high-profile publications. Furthermore, the spectroscopy component of the programme can only be accommodated on a dedicated beamline, which would also house better infrastructures for the pulsed magnet (larger capacitor banks = longer pulses).

The second approach is a **substantial upgrade of the I16 beamline and the construction of a bending magnet beamline**. The I16 upgrade will require a second experimental hutch at the position of the current user cabin to house the new horizontal diffractometer. This approach will produce a net capacity increase in the domain of magnetic scattering as part of the studies performed on I16 could be performed on a bending magnet based beamline as demonstrated by the many successful experiments performed on XMAS. Since a significant number of proposals for the new end-station are likely to be successful, beamtime available for other science projects on I16 would be reduced but part of them could be run on the new bending magnet beamline.

3. Outline specification

New beamline design. This needs to be very flexible to accommodate both diffraction and spectroscopy. The low duty cycle of pulsed magnets (few cycles per hour) makes them hard to match with weak signals (e.g. XMCD for K-edges or small magnetic moments, X-ray magnetic scattering at K-edge resonances or non-resonant); therefore we propose to integrate both pulsed (<60 T) and steady (<20 T) magnetic fields. Ideally the energy range should cover the medium hard x-ray regime from 3.4 keV to 30 keV, with beam dimension at the sample position of about $5 \times 5 \mu\text{m}^2$. It is crucial to have a very fast acquisition and crystal rotation system synchronized with the pulses. The energy dispersive geometry is well matched to pulsed fields, as the whole energy range of the XMCD/XAS spectrum is recorded in parallel at each pulse using a PSD. Although dispersive spectroscopy beamlines are usually built on bending magnets, a canted undulator is also a good solution. The energy dispersive spectrometer would employ a bent crystal to focus and disperse the polychromatic X-ray beam onto the sample. The beam passing through the sample would diverge towards a pilatus PSD, where the beam arrival position will correspond to the energy. For the diffraction mode, the beamline will be equipped with an amagnetic horizontal diffractometer, optimized for heavy load sample environment. The incident polarisation will be linear (with continuous rotation) or circular, and analysis of the final polarization will be provided. Emphasis will be placed on the minimisation of the circle of confusion, high-end cryogenics to ensure good thermalisation and excellent software to enable fast screening of small crystals (crystal quality, at a level that cannot be checked in the lab, is of the essence).

I16 upgrade and new Material and magnetism bending magnet based beamline. We have verified that an adequately sized non magnetic horizontal scattering diffractometer could be accommodated in the existing I16 user cabin. This will have to be transformed into an experimental hutch and, importantly, fitted with a higher roof to enable sample changes with the

magnet on the amagnetic diffractometer. A new control cabin would have to be provided, together with space to house the capacitor banks for the pulsed field and an extension to the peripheral laboratory to accommodate new ancillary equipment and larger sample preparation area. There would need to be modifications to the existing hutch to allow a removable beam tube and a shutter. The optical layout of the Material and Magnetism bending magnet beamline will closely match the one of the existing B16 test beamline at Diamond. The energy range covered will go from 3 keV to 30 keV. The possibility of rotating the incident light polarization either going above or below the orbit plane or with phase plate devices is important and will need to be carefully explored. The beamline will be equipped with a 6 axis K-diffractometer and a non magnetic horizontal diffractometer and on both a vacuum linear polarization analysers and a small 2D pilatus detector should be installed. This would enable one to share ancillary equipment, controls and software with I16.

Pulsed fields. Pulsed magnets can be designed to have many different sizes, peak fields, pulse durations and repetition rates. As an example, the high-field system at the APS consists of two magnets, built and designed in Tohoku University, both achieving fields up to 30 T. One magnet (a 20x20mm mini-magnet) is in a split-coil arrangement allowing for magnetic fields to be applied perpendicular to the scattering plane. It has a 1 ms pulse duration, with a repetition rate of 20 minutes for peak fields, and sits on the cold-finger of a closed-cycle cryostat. The second magnet is larger, sits in a liquid-nitrogen cryostat to increase the repetition rate and has the field applied within the scattering plane. It has an open, 18 mm bore, a pulse duration of 4 ms and peak-field repetition rate of a few minutes. Both are powered by a 40 kJ capacitor bank the size of a standard instrument rack, which can be configured to give different sweep rates at the same field. Experiments in pulsed magnets can be performed either by chopping the radiation at a particular field or, if the count rate is sufficient, in a field-resolved mode. These parameters are sufficient for X-ray spectroscopy and conventional diffraction (which would require fast crystal rotation). Resonant magnetic scattering could be observed in favourable cases. Magnet developments, aimed at duty cycles of the order of 1/100, would open up a whole new range of scientific possibilities. The proximity of Diamond to the Clarendon, the location of the UK's only pulsed magnetic field laboratory and home to a number of experts in the design and use of pulsed magnets, means that magnets could be built to suit a particular experiment on a short time-scale.

4. Community

The UK has already an exceptionally strong, world-leading community of users of both X-rays and neutrons in this research area, with groups at UCL, Oxford, Royal Holloway, Durham, Warwick and several other Universities, as well as leading scientists at RAL, DL and Diamond. In addition to using I16 and XmaS, this community represented the backbone of the ID20 programme, with an average of 50% ID20 publications having a UK author (70% in 2010). The ESRF decision to close ID20 (which can be variously interpreted) will have a disproportionate effect on the UK, at a time of growing demand and output. Should EPSRC decide *not* to renew the XmaS CRG, the UK will lose all “institutional” access to X-ray scattering in high fields. Although well established, these techniques are still perceived as somewhat specialised, since they still requires complex crystal growth and surface preparation as well as non-standard data analysis. As we explained above, we believe that the field is ready for a revolution that will significantly increase the user base, similar to that experience 10 years ago by magnetic neutron scattering in the UK with the construction of GEM and WISH. Such revolution does not depend on any conceptual breakthrough, but rather on the determination of Diamond and its users to overcome technical and engineering difficulties and focus on science delivery.

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