# Diamond-II
# Proposal for a flagship project:
# Investigations of Competing Interactions on Multiple Length-Scales (I16)

**Science Group: Magnetic Materials Group**

**Case prepared by:** **S P Collins**

**A Bombardi**

**G Nisbet**

**D Porter**

**S S Dhesi**

## 1. Summary/Impact statement

We propose to upgrade the current I16 beamline – a World-leading facility and one of the very first Diamond beamlines to be operational – to take full advantage of the new Diamond-II source and provide a globally-unique facility for the condensed matter physics community. The upgrade will complete an overhaul of the beamline, and provide a facility to support the entire experimental cycle, from high-throughput sample characterization to advanced, complex coherent and high-resolution diffraction with previously inaccessible sample environments such as high-field magnets.

## 2. Scientific Case

The significance of solid-state physics for the Digital Economy – a UKRI strategic priority area - hardly needs stating, with magnetism not only playing a central role in current information storage but (in the form of spintronics) expected to drive future high-speed, low energy, information processing. Looking even further forwards, a new paradigm in information processing - Quantum Computation – has the potential to solve complex (currently intractable) problems with manageable resource scaling. Common to both of these research areas is a renewed interest in topology in solid-state physics (*Diamond-II: Advancing Science* [1]), not only as a powerful concept that unifies seemingly disparate fields of science (e.g. cosmology and solid-state phase transitions[2]) but, specifically, as a route to discovering structures that are sufficiently stable to act as quantum computing substrates and advanced memory devices. Driven by these and other topical scientific challenges, I16 has seen a dramatic increase in research on strongly correlated electron system based on 4d[3] and 5d[4] elements (with the former now also accessible to complementary RIXS studies on I21) which, while analogous to well-studied 3d compounds, exhibit much stronger spin-orbit coupling. The latter is the driving force behind several topologically non-trivial phenomena such as magnetic skyrmions and topological insulators.

The strategic priorities for I16 are to address two important and related themes: competing electronic ordering interactions, and multiple length-scales.

Strongly-correlated electron systems, in which the essential physics of the material properties is driven by interactions between valence electrons, are characterized by multiple competing interactions and ordering phenomena, leading to highly complex ground-states that can be manipulated by external stimuli. Not surprisingly, these are of considerable importance both for their fundamental physics and their potential device applications. Complex patterns of atomic spins, formed by competing exchange interactions, can lead to symmetry breaking that allows electric polarization. Hard x-ray diffraction instruments, of which I16 is the only example in the Magnetic Materials group, have the unique capability to probe magnetic correlations at the fundamental atomic length scale, and simultaneously detect sub-atomic strain. Such magnetically-induced atomic displacements often couple strongly to external fields. For example, the electric polarization induced in $GdMn_2O_5$[5], driven largely by symmetric exchange interactions, produces a huge electric polarization, changing dramatically with applied magnetic fields of up to 6 T. The ability to study the induced changes in spin directions and atomic positions under such high field conditions, is crucial for understanding the microscopic origin of magnetoelectric coupling in a wide range of materials.

The celebrated room-temperature magnetoelectric $BiFeO_3$ exhibits complex cycloidal modulations forming large-scale domains in bulk crystals[6]. Entering the new paradigm of strain engineering in thin film materials leads not only to a striking change in the atomic spin modulation, as witnessed by x-ray and neutron diffraction, but a dramatic reduction in domain sizes down to sub-micron scales[7]. The structures, elucidated by photoelectron microscopy, are invisible to conventional diffraction techniques. However, the possibility of applying coherent diffraction (e.g. ptychography) opens opportunities for studying dynamics, while the application of external magnetic fields can change the balance of competing interactions and lead to novel ground-states. Applying magnetic fields to non-centrosymmetric bulk and thin-film materials can

lead to a rich phase diagram of topologically non-trivial ordering such as magnetic skyrmions and skyrmion lattices – systems that are the subject of intense topical scrutiny[8].
Most of the research in condensed matter is driven by the ability to change one or more thermodynamic variables to unveil new physics. Classic examples are superconductivity, until now only observed well below room temperature, and quantum criticality, where the Landau Fermi liquid theory is expected to break down and a new, yet to be fully described physics is expected to emerge, mostly approached in narrow regions by simultaneous variations of two thermodynamic variables such as temperature, and magnetic fields or pressure[9]. The discovery of skyrmions in extremely narrow pockets of the H-T phase diagram of helical magnets, and even more recently in a plethora of new materials as well as in thins films and multilayers[10], has sparked considerable interest in these phenomena, along with magnetic spin flips and flops[11], evolution and collapse of magnetic ordering, domain evolutions in multiferroics[12], and the rich physics predicted in systems described by the Kitaev model under pressure or magnetic field[9], all only accessible when external thermodynamic variables can be manipulated. Moreover, pump-probe techniques afford unique insight into short-lived and non-equilibrium dynamics[13]. A program to extend the accessible phase diagram must be seen not only as a priority for I16 but as imperative for Diamond-II to make the best possible use of the new enhanced capability of the source.

In micro- to nano-scale single crystals, coherent diffraction imaging offers a unique opportunity to study spatial variations in electronic ordering and atomic displacements. The latter has proved extremely efficacious for the study of topological defect dynamics in individual nano-grains of cathode material during battery charging and discharging cycles[14]. In ferroelectric $YMnO_3$ such defects appear to follow the same scaling laws on rapid cooling as string formation in the early universe[2]. Coherent Bragg diffraction imaging has the potential to provide a unique three-dimensional non-destructive probe of the topology of domain formation. The upgrade brings not only large gains in the coherent diffraction signal (sub-second count-times for dynamics, where tens of seconds were the norm) but also the necessary enhancements to stability of the diffractometer and cryostats, commensurate with the 10s-100s nm spatial resolution expected.

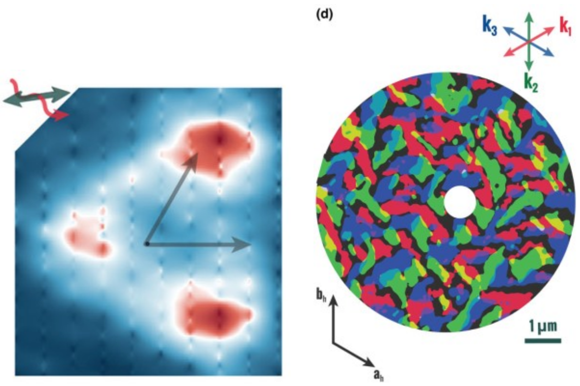


**Figure 1: Data from I16 and future possibilities. The left-figure is the diffraction pattern from helical domains in a $BiFeO_3$ thin film. The domains are averaged and their size can be estimated to be <1um, but only directly imaged using surface-sensitive PEEM on beamline I06 (right figure). The proposed upgrade should allow such domain patterns to be observed directly in the bulk, using ptychographic coherent diffraction imaging, detecting helicity, temperature and field dependence.**

The new concept for an I16 upgrade will allow the beamline to take full advantage of all of the improved beam properties of Diamond-II to provide a unique facility for coherent diffraction imaging with variable polarization and advanced sample environments. The beamline upgrade will complement plans for a small in-helium robotic diffractometer and remotely insertable microfocus optics. The combination of these developments should ensure that I16 remains at the forefront of research in complex quantum materials during the coming decades. The new suite of facilities at I16 will support the entire experimental life-cycle, from sample characterization and preliminary ordering measurements, through detailed analysis of ordered moment directions, and finally to changes induced by externally applied fields. An elegant example of a project that could be undertaken entirely on the upgraded beamline is the recent observation[15] of spontaneous cycloidal order associated with a spin reorientation in $Ca_3Ru_2O_7$, and its coupling to high (5T) magnetic fields.

## 3. Benefit to the Diamond research community

I16 is a World-class facility for resonant and magnetic hard x-ray diffraction, optimized for Diamond-I. Diamond-II provides an opportunity to develop the beamline to benefit the entire user-community, providing a low entry threshold for inexperienced users, and enhanced accommodation of state-of-the-art sample environments and techniques that are not compatible with the current instrument.

No other existing (or planned, as far as we know) beamline aims to offer a complete solution for research needs, from broad, high-throughput, exploratory surveys and characterization, to detailed investigations of the spatial and thermodynamic dependence of exotic ordered phases.

The benefits to the user community can be summarised in the context of three distinct *modi operandi* of the upgraded facility, where the first and last are particularly relevant to the industrial community:

- Robotic in-helium diffractometer (to be completed prior to the Flagship upgrade): sample characterization and preliminary magnetic diffraction measurements (T~20-350 K); fast reciprocal space mapping; training; preparation for XFEL experiments; preparation for I16 experiments. Support for remote operation.
- Main (horizontal) instrument: optimized for horizontal scattering (enhanced magnetic & resonant scattering, reduced charge scattering); full polarization control; high stability cryostat (low vibration, low sphere-of-error); larger x-ray area detector; remotely insertable microfocus optics. On-site operation by well-trained users.
- Advanced experiments (non-standard set-up of main instrument): support for large cryomagnets; support for coherent diffraction (long arm and high-resolution detectors); support for novel and user-supplied sample environments and experimental geometries. Operation by large, expert user groups.

These provisions are highly complementary to each other, with each stage building on the results of the previous, and requiring no compromise in the technical design.

## 4. Outline Specification

The proposed I6 upgrade, which retains the existing hutches, optics (2.5 – 16 keV energy range) is made possible by the reduction in horizontal divergence and source size of Diamond-II, and exploits directly the increased coherence.

Included: New large horizontal high-stability diffractometer with cryostats, magnetic fields, detectors, polarization analysis and an extendable arm for coherent Bragg diffraction imaging.

Excluded (expected to be covered by other projects): optics table upgrade with miniature diffractometer and KB mirror assembly; Stinger low-vibration cryostat.

### Design philosophy:

- Exploit horizontal scattering - better for resonant and magnetic diffraction but impractical with Diamond-I due to large horizontal source size and focus.
- Enhance stability in order to take advantage of improved coherence for coherent diffractive imaging (achieved by removing vertical degrees of freedom and using non-servo motion controls).

- Exploit new instrument design with horizontal scattering and non-magnetic components to facilitate use of large cryomagnet.

**Proposed baseline solution for a horizontally-diffracting instrument:**

- Split-chi diffractometer with +/- 10 degrees horizontal axis rotation
- Detector arm with PA and in-vac detector (Pilatus 1M)
- Extendable detector arm (2-4 m max, depending on two-theta)
- Sample xyz stage for cryostats
- Cryomagnet with vertical field (2K, 6T)
- ARS cooler with JT stage (2K) & ARS 800K cryofurnace

**Context:**

The upgrade would form part of an overall I16 provision that includes:

- Variable incident beam polarization
- Compact high-speed diffractometer with helium gas cooler and area detector
- Integrated/remotely insertable microfocus mirror system (for large diffractometer)
- Large diffractometer, optimized for horizontal scattering with high stability
- Eulerian cradle for ambient measurements, ARS and Stinger low-vibration coolers
- Large vertical theta axis mounting stage for a large cryomagnet
- Detector arm with integrated polarization analyser and area detector

## 5. State of the art benchmark

In all respects except coherence, the I16 upgraded instrument will have the almost identical properties (flux, focus, divergence, polarization) to other world-class facilities. Comparisons can then be made on the basis of beamline instrumentation. The most relevant variable polarization beamlines for resonant and magnetic hard x-ray scattering are:

- Petra-III P09 (six-axis Huber; separate horizontal instrument with 14 T magnet, 2.7-31 keV)
- APS 4-ID-D (8-circle Huber, separate 6.5T magnet not for diffraction. 2.7-40keV)
- APS 6-ID-B,C (6-circle Huber; separate horizontal instrument with 4.5 T magnet, 7-30 keV)

These world-class beamlines have adhered successfully to a fairly traditional design, with large magnets supported by separate horizontal instruments. They lack, for example, the integrated polarization analysis of the current and upgraded beamline. The I16 upgrade is a far more integrated 'three-scenario' suite that has no counterpart. The diffractometer concept is designed specifically for an ultra-low emission source, and is optimized for resonant and magnetic scattering.

For coherence applications the ESRF EBM provides a virtually identical 'coherent fraction' as Diamond-II. However, the beamlines (e.g. ID01) are not optimized for resonant and magnetic scattering, lacking sample environments and polarization control.

There is no obvious direct comparison between the future I16 and any other facility worldwide.

## 6. Community engagement

The user community will be engaged by: Sharing this document and inviting comments; direct discussion with key users; a workshop session.

## 7. References

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