# Diamond-II
# Outline for a flagship beamline project: Coherent Soft X-Ray Imaging and Dynamics (CSXID)


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## 1. Impact statement

Quantum materials are ubiquitous in our everyday lives from electricity generation to high-density data storage to medical imaging. Today, there is global effort to further understand and control the emergent properties of these materials, with the promise of next-generation low-cost, energy-efficient devices. Here, we propose to propel the understanding of quantum materials to new levels by constructing a state-of-the-art Coherent Soft X-ray Imaging and Dynamics (CSXID) beamline for high-resolution, element-selective 3D imaging and dynamic studies of new and novel materials. The new facility will revolutionise our ability to explore the static and dynamic 3D nanotexture of quantum materials with an array of leading-edge sample environments and detectors coupled to a high-intensity polarised soft X-ray beamline specifically designed to take full advantage of the large increase in coherent flux from the Diamond-II upgrade. The CSXID beamline will then offer our user community the cutting-edge tools with which to explore and develop new materials based on unprecedented nanoscale functional insights.

The CSXID beamline will facilitate state-of-the-art discovery research and innovation which directly relates to national grand challenges such as the Nanoscale Design of Functional Materials and New Quantum Technologies as well as research themes in the physical sciences such as Condensed Matter:Electronics Structure, Condensed Matter:Magnetism and Magnetic Materials, Spintronics, Materials for Energy Applications and Energy Storage [1]. These areas are addressed by enhancing nanoscale 3D imaging of the chemical and physical processes critical for the development of dial-up phenomena in complex oxides, low-energy consumption and secure data-storage materials as well as speeding up the dynamics of topologically protected spin textures.

## 2. Scientific Case

Quantum materials present immense challenges to our existing understanding of novel physical properties arising from correlated electronic structure and topological effects. In this respect, realising the full potential of quantum materials requires advanced probes essential for the prediction-production-characterisation (PPC) paradigm of developing new materials with optimised properties for next-generation devices. The transformative research directions outlined in Diamond-II | Advancing Science describe how the new capabilities offered by Diamond-II will unravel some of the fundamental nanoscale interactions governing the physical properties of quantum materials aiding methods to design, fabricate and control such materials for future applications. In this section, we present selected science topics highlighting how a new CSXID polarised soft x-ray beamline could have immediate impact in quantum material research. It is beyond the scope of this conceptual proposal to present an exhaustive review of the science disciplines that would benefit from the CSXID beamline, but we note that the beamline would host a wide user community working in diverse disciplines such as 3D electrochemical reaction mapping, 3D chemical speciation mapping and mesoscale diffusion in polymer films.

**A new dimension for magnetic imaging**

Magnetic material performance is optimised (*e.g.* in permanent magnets) and new functionalities are unlocked (*e.g.* giant magnetoresistance in the spin valve sensors) when we achieve good control of the influence of the material microstructure on the highly inhomogeneous 3D magnetisation vector field. Macroscopic probes give an average of all material properties and thus deliver, at best, indirect information about the key role of magnetic and microstructural inhomogeneities. In recent years magnetic microscopy, providing 2D magnetisation vector maps, has revolutionised our understanding of the performance and reliability of thin films devices. In particular, PhotoEmission Electron Microscopy (PEEM) has made significant contributions in this area, but it is a near surface technique which cannot be used with magnetic fields higher than a few mT. The historic lack of a 3D probe of the magnetisation vector field means that more than eighty years after the first magnetic imaging experiments, it was only very recently that a first glimpse into the inner workings of a magnetic system could be observed through the development of X-ray magnetic nanotomography [2] to map the 3D magnetisation dynamics with pump-probe magnetic laminography [3]. These recent breakthroughs in both hard [2,3] and soft [4] X-ray 3D magnetic imaging provide a route to a greater understanding of the processes that govern magnetic reversal or magnetisation dynamics, and therefore new developments of more energy-efficient materials. While first demonstrations of these techniques represent a breakthrough in terms of experimental capabilities, until now the achievable spatial resolution has been far from key material lengthscales, such as the magnetic exchange length, due to weak X-ray Magnetic Circular Dichroism (XMCD) signals, and the flux available. In this respect, the increase in coherent flux owing to the upgrade to Diamond-II will provide new capabilities to probe complete 3D magnetic profiles in *operando* conditions. In particular, it will give access to key material-specific lengthscales by employing state-of-the-art techniques such as ptychography [2] combined with the laminography geometry [3-5] (see Figure 1) using the large XMCD and X-Ray Magnetic Linear Dichroism (XMLD) signals available in the soft x-ray range. These probes will provide both high spatial resolution static imaging, as well as the mapping of magnetisation dynamics in 3D, allowing tracking of coherent magnetisation rotation, domain wall motion [3] and domain switching in the presence of defects giving unprecedented insights into the behaviour of nanoscale 3D magnetic textures controlling the macroscopic properties of 3*d*, 4*d* and 4*f* materials.

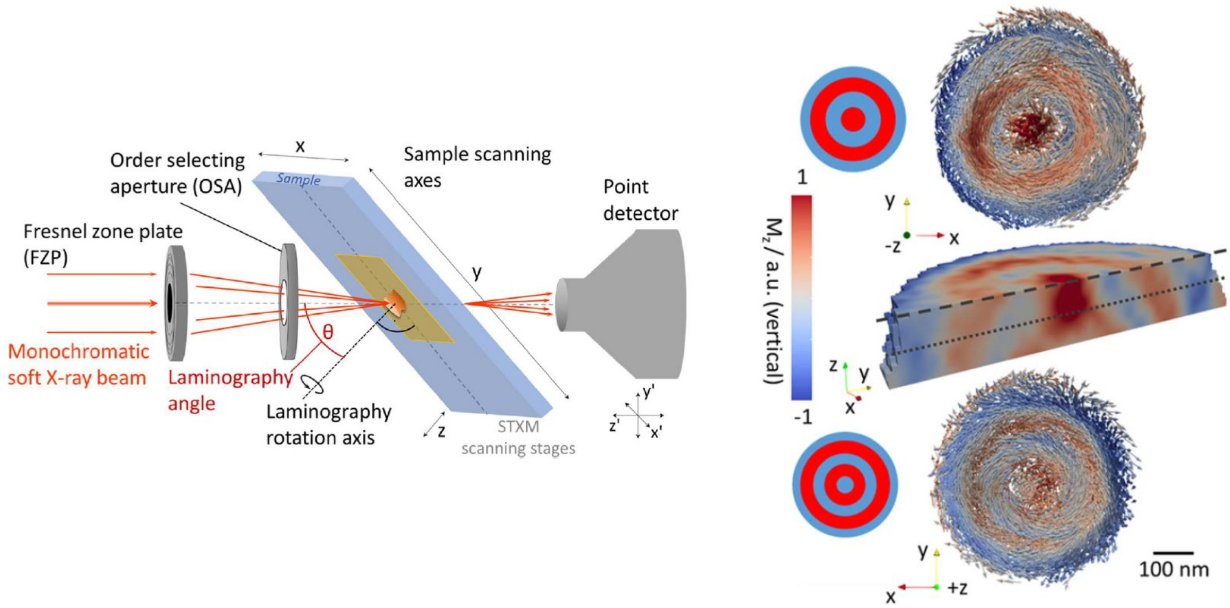


**Figure 1** (left) Soft X-ray Laminography using a scanning transmission x-ray microscope on a bending magnet beamline. (right – top) 3D magnetic laminography reconstructions from a 150nm permalloy disc showing the presence of 4 ring domains and a Néel closure cap at the top of the disc, (right-middle) cross-section through the reconstructed volume of the permalloy disc (right-bottom) magnetic structure showing the presence of Bloch domains walls and 5 ring domains at the bottom of the disc. See Ref. [4] for details.

The creation of materials with novel physical properties, predicted by advanced and intensive computational modelling, leads to an immense range of systems that require detailed characterisation to optimise performance. In this respect, one key area of activity is the development of composite materials with multiferroic properties. Naturally occurring multiferroic materials, in which electric fields can control magnetism and vice versa, have intriguing physical properties, but generally exhibit weak coupling even at cryogenic temperatures. On the other hand, thin film composites achieve electrical switching of a substantial net magnetization at room temperature using ferromagnetic thin films in which electrically driven magnetic changes arise due to strain or exchange bias from ferroelectric substrates [6]. By similar mechanisms, real-space topological features such as vortices and merons can also be transferred from functional oxides to ferromagnetic over-layers, creating stable hybrid structures offering new ways to store information at the nanoscale [7]. Here, the x-ray polarisation and element-selectivity of the CSXID beamline will reveal the details of the coupling mechanism at the oxide interfaces using, for instance the enhanced depth profiling capabilities of on resonance amplitude and off-resonance phase reconstructed magnetic images. These insights are essential to understand and control the complex interplay of strain, electronic and magnetic degrees of freedom at interfaces where new and unexpected topological features often emerge [7].

Electrical switching of antiferromagnetic ordering was achieved for the first time in 2016 with the aid of 2D magnetic imaging performed at Diamond [8]. The work represents the combined strength of predictive theoretical modelling, advanced sample synthesis methods and state-of-the-art X-ray imaging and represents a compelling case for the efficacy of the PPC paradigm. Understanding the effects of inhomogeneity, defect pinning, magnetostriction and switching using full 3D magnetic imaging will now be key to further develop antiferromagnetic materials for improved device performance. Recent studies have shown new routes for controlling antiferromagnetism, including magnetoelastic coupling, spin currents, and spin-orbit torques [9]. However, imaging antiferromagnetic domains in 3D with nanoscale resolution remains elusive, so that the increased coherence of the CSXID beamline will provide a huge boost to the field, stimulating the research and development of new probes combining tomographic methods with XMLD. Chiral spin textures, such as skyrmions and domain boundaries in thin films have also been the subject of intense studies in recent years with x-ray magnetic imaging providing key insights into the speed of these objects with current pulses. The discovery of skyrmion deflection [10] in nanowires has led to exploration of skyrmion motion in synthetic antiferromagnets which benefits hugely from 2D magnetic imaging [11]. The CSXID beamline will play a major role in the understanding of the distortions and acceleration of chiral spin structures by enabling 3D ultrafast magnetic mapping of the local spin structures with a resolution of a few nanometers.

The new capabilities of the CSXID beamline also have the potential to drive the development of entirely new fields, such as 3D nanomagnetism, where magnetic materials are sculpted into 3D configurations using advanced nanofabrication techniques [12], leading to exotic properties ranging from magnetochiral effects [13] to ultrafast domain wall motion [14]. This new field has the potential to revolutionise storage and information technologies based on the controlled movement of domain wall structures in engineered energy landscapes.

**Controlling emergence using phase separation**

To understand and manipulate the microscopic mechanisms leading to emergent phenomena such as superconductivity, magnetoelectric coupling and magnetism it is imperative to decode phase fluctuations with nanometer spatial and nanosecond temporal resolution. Here, X-ray Photon Correlation Spectroscopy (XPCS) will play an increasingly important role since it probes the dynamical structure factor in the time domain. The spatial correlations are then given by the scattering wavevector. Currently, up to millisecond dynamics, on length scales of hundreds of nanometres to several Ångstroms, are routinely probed using third-generation sources. The large increase in coherent flux on Diamond-II will then allow the possibility of nanosecond dynamics to follow spatial correlations probed on much smaller length scales for an array of materials. For instance, magnetic skyrmions are considered to be stabilised by spin fluctuations arising from the competition between the symmetric and antisymmetric exchange interactions giving rise to disordered phases with skyrmion-like short range order close to the ordering temperature in MnSi [15]. Element-resolved XPCS would give transformative insights into the dynamics of skyrmion formation and the relevant order parameters. XPCS would also make a huge impact on the understanding of nanoscale phase separation in quantum materials, which has the potential to create devices based on new functional properties.

## 3. Benefit to the Diamond research community

Lensless imaging, such as Coherent Diffraction Imaging (CDI) and ptychography, have developed considerably in recent years achieving a spatial resolution of ~15 nm in 3D under favourable circumstances [16]. A low-emittance machine would increase the coherent fraction by at least an order of magnitude from 500 eV to 3 keV, and together with improvements to the insertion devices and beamline optics, could lead to an increase in the coherent flux at the sample by several orders of magnitude. This then enables 3D nanotomography to become a routine tool with which to explore functional devices in *operando*. The time to generate a full 3D magnetic tomogram is expected to fall from 30 hours for the first results [2] to ~1 hour at a diffraction limited source.

There is a substantial cohort of users that will benefit directly from the new insights that can be gained from the CSXID beamline, from those researchers working on quantum materials to battery storage to those developing new and novel nanostructured devices. Coherent X-ray imaging will provide element-selective 3D scalar and vector mapping with nanometer scale spatial resolution transforming many fields by permitting, for instance, the position, velocity and chirality of novel spin textures to be controlled by spin polarised currents, magnetic and electric fields. Combining tomographic methods with circular and linear dichroism contrast mechanisms will further offer the possibility of obtaining 3D images of spin and electronic structures in ferroic materials, interfaces and nanostructures during growth or in *operando* conditions. The element-specific 3D tracking of electrochemical reactions in battery materials along with electrode degradation will become routinely available [17]. The ability of XPCS to cover timescales from ~1Hz to ~1GHz in an element- and site-selective manner (not available with neutron spin-echo) would allow access to a range of dynamics currently inaccessible in a single technique. The *ultrafast* 3D imaging capabilities, at low-temperatures and high magnetic and electric fields, will provide opportunities for step changes in our understanding of how nanoscale phenomena control macroscopic material properties.

## 4. Outline Specification

To realise nanoscale spatial resolution in 3D, and higher frequency dynamics in soft X-ray based XPCS, requires a beamline optimised to benefit from the significant increase in coherent flux on Diamond-II. The CSXID beamline would ideally be located on a straight-section providing the highest coherent flux possible which is I17. A mid-straight section is also a possibility with a shorter APPLE-II undulator which would require slightly longer times for data acquisition. Some of the key performance parameters of the beamline are outlined below.

- Source: APPLE-II
- Energy range: 250 - 3000 eV
- Energy resolution (E/ΔE): ~$10^4$ at 700 eV
- Coherent photon flux: ~$10^{12}$ phs/s into end station
- Beam-size at sample: 500nm (H) × 500nm (V)
- Sample environment: low temperatures (~10K), magnetic fields (~1T)
- Spatial resolution: ~5nm for 3D tomographic reconstructions

A variety of proven design options have been implemented on modern soft x-ray beamlines with VLS gratings providing focusing in the dispersive plane and therefore reducing the number of reflections. The beamline will most likely comprise two branches for imaging and dynamics and require a modest resolving power to maximize the coherent flux with low vibration and low thermal deformation of the optics essential to preserve the coherence. Detectors with a large dynamic range will be deployed for high spatial resolution, since the intensity falls rapidly as ~$q^{-4}$ with angle, and high-intensity low-$q$ scattering yielding information on large scale structures. High-frame rates (~1kfps) and high quantum efficiency

(~100%) gated detectors will be installed for the 3D tomography and dynamical studies. The dramatic increase in angular resolution and range in reciprocal space also means that 3D ptychographic imaging gives orders of magnitude more data volumes which will require high bandwidth data acquisition networks and high-performance real-time analysis on a dedicated cluster of computer nodes.

## 5. State of the art benchmarks

Coherent soft x-ray imaging has been flourishing worldwide since the first report appeared 16 years ago [18], but only very recently has soft x-ray ptychography, with its stringent requirements for ultrastable optics and sample environments, been considered for 3D imaging. There are a number of beamlines currently developing coherent x-ray magnetic imaging techniques as additions to their main activities including the SIM beamline at the SLS, SEXTANT beamline at Soleil and CSX beamline at the NSLS-II. The COSMIC beamline at the ALS is purpose built to exploit the coherent properties of the ALS upgrade and operates one branch for imaging and the other branch for resonant scattering experiments. Diamond is also building a branchline on I08 for ptychographic imaging which will share the available beamtime with the scanning transmission x-ray microscope. The I08 branchline is a multipurpose facility which will cater for a diverse user community working on organic and inorganic matter covering electrochemistry, geology, catalysis and related *operando* or *in situ* research. The CSXID beamline would be the first beamline worldwide equipped to discover the wealth of phenomena exhibited in emerging materials by housing state-of-the-art detectors, sample environments, low-temperatures and magnetic fields to image complete 3D structures in *operando* and access dynamics on timescales of hours to picoseconds.

## 6. Community engagement

The UK user community working in x-ray imaging and dynamics is strong with a diverse portfolio of activity benefitting from x-ray based imaging, spectroscopy, diffraction and time-resolved studies. The proposal will be developed by a champion together with a user working group representing the broadest range of stakeholders from the UK and international user communities as well as Diamond staff. A workshop will be held to engage with the user community to identify the key technical performance parameters.

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