Diamond-II
Proposal for flagship project

Upgrade to beamline I13L

Science Group: **Imaging**

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1. Summary/Impact statement

Over the last few years, X-ray coherent diffraction imaging techniques in their different modalities have evolved rapidly from proof of concept to supporting discoveries in many research fields. Hard X-ray ptychography, in particular, is transforming observations made in nanoelectronics, complex domain structures in nanomagnetism, understanding nanoscale transformations and processes in single grains of metal ion-batteries, understanding pharmaceutical chemistry or probing bio-medical systems. The brightness increase of Diamond-II offers an unmissable opportunity to leverage the capital investment made to date in the infrastructure of I13L to provide world leading experimental capabilities. The project aims to deliver a state-of-the art facility for hard X-ray ptychography and multi-scale imaging. By exploiting the increased brightness of the source, using the latest undulator technology, and by fully rebuilding the endstations, including provision of cryo-capabilities, we will deliver higher-resolutions, shorter timescales and higher throughput with impact both pre and post Diamond-II. This step change will enable nanoscale in-operando coherent imaging to become a routine technique at Diamond-II and provides a platform for the development of hard X-ray 3D cryo-biological imaging. This will open the door to probing material properties and biological samples at unprecedented length scales or enable parametric studies at the current resolution.

2. Scientific Case

The increased brilliance of Diamond-II, combined with the proposed beamline upgrades, including a change of source and the use of broad bandwidth radiation, will boost scientific discovery by enabling dynamic experiments rather than static measurements, allowing statistically significant studies of large set of samples, and provide environments which best preserve the native state under investigation. Coherent imaging is a multidisciplinary tool with widespread applications and so we have selected just a few examples of where these upgrades will make a significant impact.

Science driver I: Imaging the brain and the study of neural circuits

Large volume imaging is essential to understanding hierarchical structures in biology such as the brain. Imaging of a 0.5 mm^3 region of the brain currently requires around 2.5 months using a destructive serial block electron microscopy approach with several days of pre-staining to improve contrast. Statistical studies are therefore limited, and larger volumes and image quality are impacted by staining protocols.

The question of how neural circuits receive, process and propagate information to drive behaviour is of central importance in neuroscience. Linking the physiological neuronal responses that distinct stimuli such as for example images, sounds, or odorants evoke, to its underlying anatomy is one of the big challenges in today's research. Currently, sub-cellular structures are studied with electron microscopy techniques that provide ~10nm resolution but are destructive and require weeks of dedicated instrument time for limited volumes of little more than ~100-500 μm^3. Synchrotron X-ray tomography at sub-micrometer resolution can provide insight into neuronal morphologies, where cells receive information from and where they relay it to. Even deeper understanding of the neural circuit function is given when synapses, the connections between neurons, can be revealed. This allows to describe the full wiring diagram and thus provides the structural underpinning for information processing in the brain.

The re-design of the ptychography endstation coupled with increased automation and cryogenic capabilities would enable fast throughput nanoscale imaging to build up statistically relevant information spanning nano- to micro- length scales. The increased flux on I13L from Diamond-II and ongoing method developments (multi-slice ptychography) should increase both the volumes and resolutions obtained to enable non-destructive imaging of ~1mm^3 volume of brain with a resolution of 100nm (morphologies) or 30nm (synapses) which would be transformative for bio-imaging workflows. Synchrotron 3D biological nano-imaging is still in its infancy and to date there are less than a handful of examples which show the potential impact and role of X-ray imaging for large volume imaging of biological cells and tissues. The development of a cryo-end station at I13 which can be made available to the wider UK user community is an essential part of Diamond's strategy to build the bio-imaging infrastructure.

Science driver II: Battery materials and electrodes

The challenges of battery research are straightforward to state but difficult to answer – why does the voltage drop on the first cycle of the battery and how do we design materials to control it? What are the mechanisms for long-term degradation of the voltage – what role do strain, chemical state, volume effects (fracture, expansions) at the micro- and nano-scale play?

Large volume changes, for example, are known to occur in the cathode and anode during the de-lithiation and lithiation process. First-cycle voltage drop is believed to be related to a collapse of the overall superstructure and long-term degradation is related to the evolution of cracks and strain with volume change at each charging cycle. Common battery materials such as NMC811 consist of hierarchy of microscale (10's-100's µm) particles made up of nanoscale secondary particles (50-300 nm). As a result, properties such as turtuosity which describes the pathlength of the charge carrying ion along the electrode material can be described across the micro- and nano-length scales[1]. Progress in battery materials relates to a deeper understanding between structure and properties of the constituents, the capability of observing dynamic processes and the study of the basic chemical exchange processes involved.

In-operando micro- to nano-scale imaging of volume, chemical, compositional and crystallographic changes is key to answering these questions. Many of the tools on I13L have been used for different aspects of battery research: In-line phase-contrast (PC) for in-operando imaging of the deterioration of battery electrodes[1], the nano-structure of the electrodes with full-field microscopy and ptycho-tomography and the lithiation process of graphite electrodes with Bragg-CDI (CDI: coherent diffraction imaging). However, imaging across multiple charging cycles with coherent imaging of nanoscale properties has been limited to static measurements or via removal of the sample to cycle externally. A transmission X-ray microscope would allow routine rapid nanoscale imaging of the cathode in-operando. Improvements to the coherence branch endstation (Bragg-CDI and ptychography) coupled with the flux increases from Diamond-II will enable rapid *in-operando* nanoscale imaging across various electrode types to track the chemical, structural, compositional and crystallographic changes essential to fully explore battery operation and inform future developments.

Science driver III: Understanding the crystallisation process for pharmaceutical products and industrial processes

The understanding of the nucleation process is one of the grand challenges for the chemical and pharmaceutical industry, but in-situ and in-operando monitoring of very small mass density differences is challenging requiring high sensitivity and short timescales.
The nucleation process has a critical impact on the properties of chemical and pharmaceutical products such as purity, morphology and crystal size distribution. The main research interest currently focuses on the nucleation stages and organic crystals growth from solution. Possible nucleation pathways include sequences of self-assembly events from individual solvated molecules in solution, followed by desolvation and self-assembly into pre-nucleation clusters, and subsequent nucleation and crystal growth[2].

Nucleation is a rare stochastic event in both time and space, and it is a multiscale process involving molecular, mesoscopic and macroscopic length scales. X-ray imaging can play a major role but needs to satisfy several criteria: (i) sufficiently time-resolving, (ii) sufficiently spatially resolving (imaging, microscopy), (iii) provide information across the various length scales, (iv) observe the process under practical conditions (i.e. in-situ and in-operando experiments at ambient pressure).

Pre- and crystallisation events have been imaged successfully for the first time using a trial grating interferometry setup on I13L using a concentric mixing flow crystalliser to compare and contrast crystallisation of glycine and lovastatin. Endstation improvements and increased flux will enable routine observation of faster dynamics with higher sensitivity, which is important, for example, to observe for the phase preceding crystallisation.

3. Benefit to the Diamond research community

The proposal builds on the existing facility and expertise at Diamond with upgrades that will benefit users now and immediately after Diamond-II. The proposed improvements evolve from the ongoing imaging developments at I13L and pushes new capabilities to meet emerging scientific challenges along with a need for greater simplicity and automation for an increasingly novice and varied community. In particular, with a large and vibrant life sciences community and the increasing reach of structural biology to image macromolecules in situ, the development of large volume biological X-ray imaging is a key part of Diamond's plan to integrate cellular and structural biology. This proposal is a crucial first step in building up expertise and increasing awareness and easy access to X-ray imaging capabilities that can then be fully exploited by the life sciences at Diamond II through the dedicated bio-imaging beamline proposed as part of the second tranche of new beamlines. The higher flux at higher energies will have broad impact. Industry process-replication with in-situ cells, for example, typically requires the use of denser materials, higher temperatures and pressures requiring higher energies to penetrate the rigs. The proposal allows immediate delivery of the benefits of Diamond-II compared to a new facility and leverages the unique co-location of the imaging and coherence branch to deliver multi-scale imaging approaches.

4. Outline Specification

The upgrade of the machine and the beamline enables significantly new experimental capabilities and delivering on this potential and exploiting Diamond-II requires:

- Dedicated endstation(s) for ptychography (fast scanning (≥9kHz) – improved stability, ptychography in transmission and Bragg geometry along with consideration of sample conditions such as cryo, air-sensitive, battery cells).
- Sample environments with a major focus on the delivery of an in vacuum cryogenic endstation optimised for ptychography of biological samples with the capability to correlate to optical fluorescence microscopy.
- Dedicated transmission X-ray microscope for fast nano-tomography.
- Increase throughput developments (automated sample changing, pink-beam ptychography upgrades, remote experiments).
- Automated workflows (from samples to science images).
- Higher flux at higher photon energies – new sources (radiation damage, penetration depth, heavier elements). For the imaging and coherence branch the source, coupled with new undulator sources will improve the photon flux across the energy range (Figure 1).

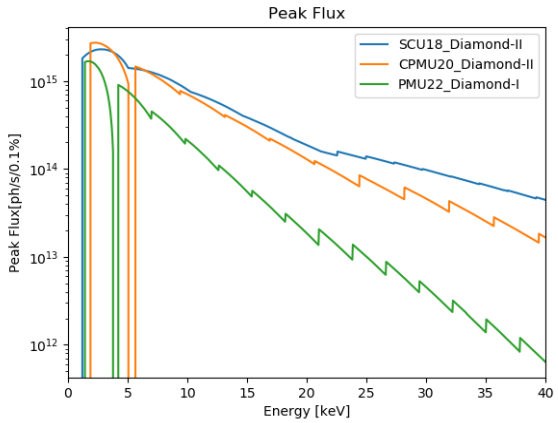

Figure 1a: Imaging Branch Peak flux for different insertion device options for Diamond-II compared to current Diamond I performance.

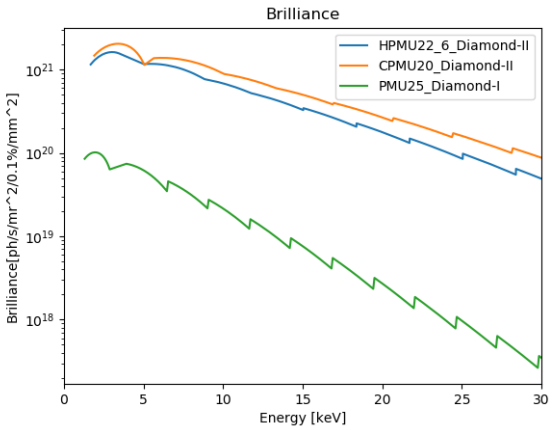

Figure 1b Coherence Branch Brilliance for different insertion device options for Diamond II compared to current Diamond I performance.

Table 1 summarises the imaging methods for I13L scaled for Diamond-II performance based on the expected peak flux and brilliance curves.

Table 1: Imaging methods at I13L

Method	Micro-tomography	Grating interferometry	Full-field microscopy	Ptychography	Bragg-CDI
Resolution	1-8 µm	>5 µm & SAXS	50-100nm	10-100nm	TBD
Field of view*	1-10 mm	1-10 mm	50-250 µm	<300 µm	<10µm
Energy range	8-38keV	14-25keV	8-14keV	6-25keV	6-20keV
t_{exp}	10µs-100ms	5-100ms	2-50ms	10µs-100ms	sec.-min.
$t_{Tomo\ scan}$	<sec.-min.	<sec.-min.	<sec.-min.	sec.-min	~5h
Pink/MLM	Pink	Pink/MLM	MLM	Mono/Pink	DCM
Element specif.	Yes[1)]	Not tested	Not tested	Yes[2)]	-
Spectroscopy	-	-	-	Yes[3)]	-

* baseline
1) with DCM and MLM
2) with QCM, detector or reconstruction method [3]
3) with fluorescence detector [4]

5. State of the art benchmark

I13-1 coherence branch currently aligns with the performance of other ptychography beamlines in the main synchrotron facilities for energy range (6-20 keV)[5], coherent flux and multimodal approach (Bragg-CDI, XRF)[3,4,6] as shown in Table 2. I13-1 excels in the ptychography acquisition speed, as shown in Figure 2. The outstanding performance (9kHz acquisition[7]) has been achieved with a combination of beamline design and acquisition control development. Diamond-II upgrade is expected to enhance the speed and performance, enabling a further increase of more than one order of magnitude.
Diamond II will also give access to higher resolution (from currently < 50nm to < 10nm) and wider energy spectrum (up to 30 keV) which will position I13-1 in a leading position in a worldwide picture of ptychography beamlines.
Sample environments play a critical role in the science possible and state of the art for imaging can be the ability to operate in-operando or at cryogenic conditions to push new science areas. cSAXS at SLS, is the current, and only, benchmark for state of the art cryo-ptycho-tomoography and in-operando studies are

still at very early stages worldwide. I13-1 is well placed to leverage the on-site expertise from the electron and X-ray facilities to build a leading platform for native or in-situ sample studies.

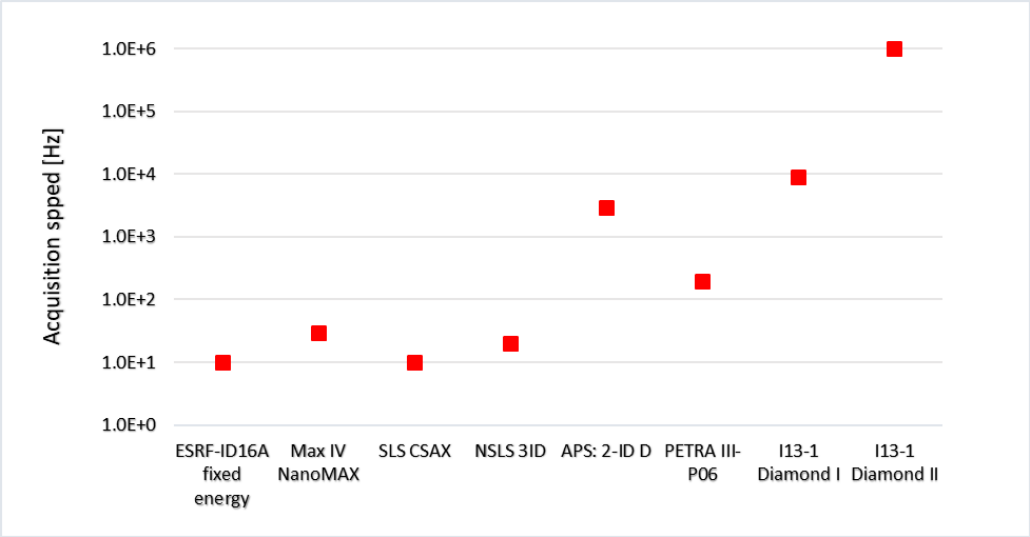

Figure 2: Acquisition speed and coherent flux comparison for hard X-ray ptychography beamlines.

Table 2: Comparison of ptychography beamlines

Instrument	Technique	Energy Range [keV]	Coherent Flux [photons/s]	Resolution [nm]
ESRF-ID16A[8,9]	Ptychography, XRF, tomography,	17; 33.6 Fixed energy	1.00E+10	<10
Max IV NanoMAX[10]	Ptychography, Bragg-CDI, XRF	5-28	2.00E+11	<50
SLS cSAXS[11]	Ptychography, SAXS, WAXS	4.4-17.9	7.00E+08	<10
NSLS 3ID[12,13]	Ptychography, fluorescence, Bragg-ptychography, XANES	6-25	5.00E+08	<30
APS: 2-ID D[14]	Ptychography , micro-fluorescence, micro-XANES	6-30	1.00E+10	<10
Petra III- P06[15]	Ptychography, micro-XRF, micro-XRD, micro-XAS	7-30	1.00E+08	<10
I13-1 Diamond I	Ptychography, Bragg-CDI, Bragg-ptychography, 3D XRF	7-20	1.00E+09	<50
I13-1 Diamond II	Ptychography, Bragg-CDI, Bragg-ptychography, 3D XRF	7-30	5.00E+10	<10

I13-2, the imaging branch, compares very favourably to other full-field imaging facilities for multi-scale, multi-modal capabilities (Table 3). I13-2 offers FOV from 10s μm to cm and resolution from 100nm to 1μm with a multi-scale and multi-modal approach with implemented techniques including TXM[16], grating interferometer[2] and micro-CT. The Diamond-II upgrade will enable to reach higher energies (from max 30 keV to max 38 keV) allowing to fill an existing energy gap within Diamond imaging instruments and, with increased flux, to enhance the throughput and acquisition speed.

Table 3: Comparison of full field imaging beamlines

Facility	Energy range	Technique	Sample environment	Speed	Resolution
ESRF-ID19[17,18]	10-250 keV	Laminography, Phase-contrast, micro-CT, ultra-fast radiography	Various in-situ furnaces and mechanical loading stages	1kHz[17] or 1MHz[18]	1μm-100μm

Max IV-DanMAX[19]	15–35 keV	Full-field Imaging, powder diffraction	Provided by users	ms-s exposure	50nm-5µm
SLS TOMCAT[20]	8-45 keV	micro-CT, TXM	A laser-based heating system and a cryo-jet and -chamber	20 Hz micro-CT	voxel size 0.16 to 11µm
NSLS-18ID[21]	5-11 keV	TXM, 2D, 3D nano-XANES		1 min nano-CT	30 nm
Soleil Anatomix[22]	10-50 keV	micro-CT, TXM	Provided by users	3D TXM ~ 3min, 20 Hz micro-CT	80nm - 1µm
APS: 2BM[23]	11-35 keV	micro-CT	Various in-situ environment cells	<1-25min 3D micro-CT	1-10µm
Petra III: P05[24]	5-50 keV	Full-field tomography, micro-CT, TXM, grating interferometry	Furnace, corrosion cell, load frame, hanging axis	<1 min 3D micro-CT, 10ms TXM exp.	100nm -1µm
I13-2 Diamond I[5]	7-30 keV	micro-CT, TXM, grating interferometry	Provided by users	100µs-100ms <min-min CT	100nm-1µm
I13-2 Diamond II	7-38 keV	micro-CT, TXM, grating interferometry	In-house & collaborative sample environments	10µs-100ms <s-min CT	50nm-1µm

6. Community engagement

For Diamond-II, the user working group meetings were restarted with a broad range of science represented (chair Alexander Korsunsky). The most recent user working group meeting (April 2020) discussed the requirements for cryo-capabilities with particular input from Andreas Schaefer (Crick), for a bio-imaging perspective, and the case for multi-scale and multi-modal imaging including the different options for the implementation of large volume/high resolution or imaging with zooming capabilities were discussed and supported. Further meetings are planned for the summer 2020 and joint workshops covering this proposal and the dedicated Cryo biological nano-imaging beamline will be held to engage with the nascent biology community. Andreas Schaefer will also be involved in the Cryo biological nano-imaging beamline user working group which forms the second phase of Diamond's X-ray imaging strategy.

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