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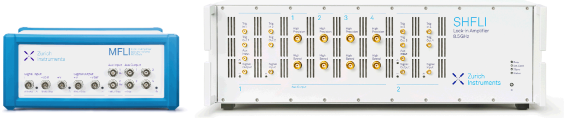

New Opportunities for the *XMaS* Beamline Arising from the ESRF Upgrade Program

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Abstract. The *XMaS* bending magnet beamline at the ESRF has been in regular user operation since the autumn of 1998 and has employed a very simple optical system consisting of a Si <111> monochromator and a toroidal mirror. The ESRF extremely brilliant source (EBS) upgrade program presents the bending magnet beamlines with a series of challenges and exciting new opportunities to extend the range of science performed, with emphasis on *in-operando* and *in-situ* studies. Geometrically, the new EBS lattice will move the source for bending magnet beamlines some 3 meters upstream and *XMaS* will use a newly designed 0.86 Tesla short bend, instead of the present 0.4 Tesla bending magnet. The higher field of the new source increases the available flux at high energies (>25 keV) by an order of magnitude and will result in a smaller brighter beam. To exploit the extended energy range, a dual toroidal mirror system, coated with chromium and platinum, will provide the focusing optics and enable continuous operations from 2.035 keV to 33 keV which will be coupled to a fast scanning LN_2 cooled, constant offset monochromator. We report here on the opportunities presented by the new machine lattice and the solutions chosen to deliver a state of the art beamline that utilizes a very wide range of x-ray techniques including scattering and spectroscopy from a broad spectrum of materials characterization.

INTRODUCTION

The *XMaS* facility is a synchrotron beamline funded by the UK EPSRC research council. It is embedded in the heart of the European Photon and Neutron (EPN) Science Campus in Grenoble, France and is managed by the Universities of Liverpool and Warwick. The beamline was originally conceived in the mid-1990s primarily to perform high resolution diffraction and magnetic x-ray scattering [1], hence the acronym X-ray Magnetic Scattering (*XMaS*). A major scientific interest at the time was studies of actinide magnetism [2] and so the beamline was optimized for operations down to energies as low as the U M_5 absorption edge (~3.55 keV) which motivated the choice of an ESRF 0.4 Tesla bending magnet, with a critical photon energy of approximately 9.8 keV as the source. The design concept employed a very simple optical configuration [3]. A directly water cooled Si<111> monochromator was chosen as the first optical element, followed by a single rhodium coated toroidal focusing mirror, with an incidence angle of 4.5 mrad to the incoming monochromatic beam. This rhodium mirror provides an upper energy cutoff of ~15 keV, whereas the lower energy limit of 2.4 keV is governed by the geometry of the monochromator. The primary instrument of the beamline is an 11-axis Huber diffractometer that can be configured to operate with a wide variety of detectors and analyzers.

The beamline has been supporting users since 1998, but now has a far broader scientific community than the original magnetic scattering remit and supports active research groups in over 50 UK universities (representing over 400 independent researchers). Indeed, during the last 20 years of operation of the beamline, the scientific portfolio has greatly expanded from the original core activity of resonant x-ray diffraction – hence its rebirth as the UK's X-ray Materials Science beamline at the ESRF. The facility is an enabling tool serving the materials science community, including academic researchers, national research laboratories and industry. It plays a major role in underpinning interdisciplinary projects and contributes directly to societal challenges such as energy storage and recovery, the digital economy and advances in healthcare technologies as well as contributing to the UK research infrastructure. The scientific areas tackled on the beamline cover a broader remit than is normally found on a single beamline and a wide range of materials are now studied, using a variety of x-ray techniques. Experiments are routinely performed, such as surface x-ray diffraction [4], small/wide angle scattering (SAXS/ WAXS) measurements of soft condensed

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matter systems [5,6] and x-ray absorption spectroscopy (XAS) within a wide range of *in-operando* sample environments [7] and under a range of external stimuli (applied electric and magnetic fields). Over the last ten years we have made considerable efforts to extend our operational energy range into the “tender” regime (2.4-5 keV). This opened up new spectroscopic and scattering opportunities at the S and Cl K-edges, as well as opportunities for resonant x-ray diffraction studies at the Ru and Pd L-edges and the Ir M-edges. Being open to such developments is at the heart of the operational ethos of the beamline and the range of sample environments is continually expanding.

However, after over 20 years of operations, core capabilities need to be updated. Serendipitously the European Synchrotron Radiation Facility (ESRF) undergoes the final phase of its upgrade program with the installation of an ultra-low emittance storage ring in 2019 [8]. The low-emittance ring will necessarily mean major changes for all the bending magnet beamlines at the ESRF. Within the lattice, a space has been identified in which a small source with 2 mrad acceptance could be located. The new source position will be slightly shifted with respect to the current source. With the smaller acceptance, the source size will be reduced but with significantly improved brightness. As well as exploiting the smaller beam size, a drive to higher operational energies whilst simultaneously continuing to deliver our current capabilities was identified as key by our user community.

Thus, a complete upgrade of the instrumentation is required to exploit the potential of a new source which will expand the scientific challenges that can currently be addressed. A high brightness source will allow the same sample volume across an extensive energy range and within the same sample environment to be studied. This will enable real time reactions to be followed on a site-by-site basis opening up new opportunities for catalysis and green chemistry. Higher flux at low energies permits studies at the K-edges of P, S and Cl in dilute systems and over shorter timescales. The facility will deliver new insights into quantum critical behavior as well as facilitating studies of confinement and proximity in magnetic and superconducting materials at low temperatures (1-10 K). Advances in simultaneous studies will give the direct correlation of functional properties with structure across the materials disciplines. Newly combined (GI)-SAXS/WAXS (grazing incidence) and x-ray reflectivity (XRR) metrologies enable structure to be measured across a wide range of length and time scales simultaneously. More systems will be studied *in-operando* and under technologically relevant conditions, for example, the study of ionic migration in battery systems and photovoltaics. The small beam facilitates future experiments on small samples or on localized regions of larger samples. Structural studies will become spatially resolved allowing studies of individual domains and their temporal evolution under external stimuli. An upper energy of ~33 keV will extend studies of buried interfaces in complex sample environments across the K-edges of the rare-earths as well as the L-edges of the transuranic elements. Solid-liquid interfaces, relevant to electrochemical processing, also become more accessible. Finally, the new lattice will greatly facilitate polarisation studies due to the better defined beam position and more efficient phase retarders leading to the development of new polarisation studies such as SAXS/WAXS from chiral systems. External stimuli including electrical and magnetic fields as well as humidity, gaseous atmospheres and temperature control (1 to 1200 K) will all be available.

In this paper, we detail the upgrade plans and the choices we have made to align the new facility to the new capabilities offered by the EBS upgrade.

CHOICE OF SOURCE

The most significant impact in the ESRF EBS program is the removal of the bending magnets as sources from the lattice. In the new ESRF lattice design, a new source located upstream of the current source position has been identified. Various options were considered: using a small insertion device or exploiting the steering magnets already part of the lattice design. In all of the six possible sources, a maximum horizontal fan of 2 mrad acceptance was possible. The first three options to be considered were a three pole wiggler (3PW), a two pole wiggler in a “plus-minus” configuration (2PA) and another two pole wiggler in a “minus – plus” geometry (2PB). The three pole wiggler looked initially attractive due to increased flux but unfortunately suffered from significant problems due to contamination from the upstream and downstream bending magnets (DQ2C and DQ1D) and a very non uniform horizontal flux distribution in the flux/energy spectra below 3 keV, an important energy regime that we wished to retain access to on the beamline. There was also a drop of intensity on the optical axis.

Both of the two pole wiggler options allowed the removal of *one* of the contaminant DQ bending magnet sources by using primary slits, however, only the “minus-plus” (2PB) allows the removal of both sources. Primary slits could have been used to remove the downstream (DQ1D) bending magnet radiation, but the upstream radiation (DQ2C) is only separated from the two pole wiggler radiation of interest by some 0.3 mm at the focal position, using a simple toroidal mirror. This contaminant radiation could in principle, be removed by a slit system very close to the sample position, but as the beamline regularly performs small angle scattering (SAXS) and x-ray reflectivity (XRR) measurements, this option was rejected due to the potential of a high x-ray background in this important small angle region.

The final three source choices were bending magnets, the DQ2C is a 0.39 Tesla dipole/quadrapole magnet, the DQ1D 0.57 Tesla dipole/quadrapole and finally a 1 pole "short bend", with a magnetic field of 0.86 Tesla. All of these bending magnet sources provide a clean uncontaminated x-ray source. However, the higher field 0.86 Tesla "short bend" source provides significantly more flux at higher x-ray energies and, given the increasing importance of this energy window this has been the source that has been chosen for the *XMaS* beamline for post EBS operations. The calculated flux produced by the source is shown in Fig. 1.

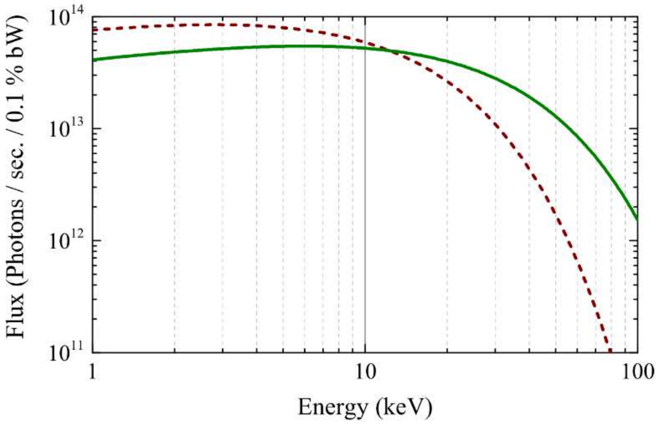

FIGURE 1. Calculated flux curves. The dashed curve shows the flux from the current 0.4 T, 3 mrad horizontal fan source, whereas the solid curve shows flux from the 2 mrad fan of the new 0.86 T short bend.

OPTICAL CONFIGURATION

From the very first design studies of the *XMaS* beamline, it was decided to use a very simple optical system consisting of a directly water cooled Si<111> monochromator and a toroidal mirror in a roughly 1:1 optical geometry. This simple, but robust system (Fig. 2) has ensured that a diverse and interdisciplinary science program can be maintained. It is intuitive for users and allows for rapid changes of x-ray energy by simply rotating the monochromator Bragg axis and adjusting the distance between the first and second monochromator crystals. Downstream of the monochromator the next optical element is the toroidal mirror which focuses the beam onto the sample. In addition, for further beam conditioning, two plane harmonic rejection mirrors and a phase retarder can be employed. The largest impact of the EBS upgrade will be the movement of the source some 3 m upstream in the lattice. Again, to ensure that the benefits from the new source are fully maximized, the experimental hutch will be extended by 4 m downstream and the diffractometer moved to maintain the current 1:1 focus. Overall, however, the upgrade will have a limited impact on the optical design but the main optical components will be refreshed to latest standards and re-designed to match the source characteristics. We detail these in the following sections.

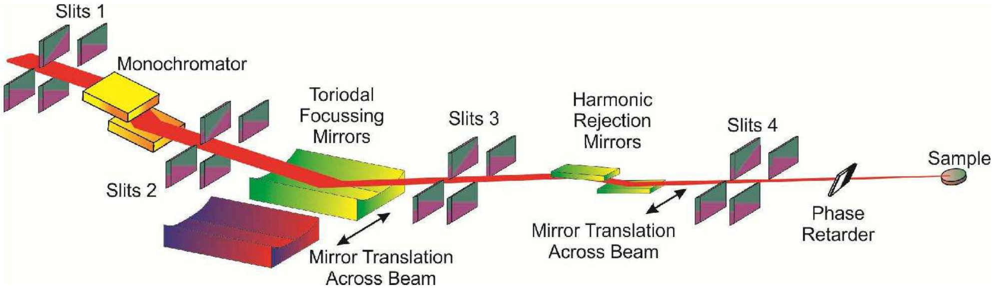

FIGURE 2. Schematic diagram of the optical configuration of the *XMaS* beamline. Post EBS upgrade an additional toroidal focusing mirror will be used to extend the upper energy range of the beamline and to take advantage of the new smaller source, 3 meters further upstream from the present source.

LN_2 Monochromator

It will be necessary to replace the current water cooled monochromator with a LN_2 cooled system to cope with the increased thermal load and reduce heat-bumps. In addition, to exploit the flux at higher energies, the energy range will be extended from 2.035 keV to around 33 keV. Directly cooled LN_2 copper blocks are to be mounted to the sides of both the first and second silicon monochromator crystals, clamped using a pre-defined stress with a thin layer of indium metal between the copper blocks and the silicon crystal, to ensure good thermal conductivity. Both cooled crystals are thermally isolated by intermediate invar plates mounted onto ceramic balls, creating point contacts, thus minimizing thermal contact between the crystals at cryogenic temperatures and the crystal cage at room temperature. Three high precision linear actuators control the distance between the first and second crystal, with the second crystal sitting on a three point kinematic mount. Considerable effort was made to design high side load linear actuators, with a non-rotating shaft that can maintain both the required precision and resolution when the monochromator is operating in the low energy range. For a Si<111> monochromator, the Bragg angle at the sulfur K-edge is 55° which increases to around 70° at the phosphor K-edge, placing a considerable side load onto the actuators, due to the mass of the second crystal assembly. To ensure increased accuracy and to enable fixed exit height operations, the actuator shafts are now guided by a sleeve bearing, ensuring both high side load capability and a 50 nm per step achieved by using a rotating nut to drive the threaded shaft, via a 200:1 gearbox.

Focusing Toroidal Mirrors

For focusing the beam, the same simple toroidal mirror geometry that is currently used has been retained. However, as the useable energy of the beamline is to be extended up to ~33 keV, new mirror coatings and angles of incidence would be required. One particularly demanding requirement, primarily for XAS studies, was the ability to rapidly change energy between 2.035 and ~33 keV whilst maintaining the focal spot at the same position. The current primary mirror is rhodium coated, with an incident angle of 4.5 mrad. Along with the mirror coating material, the angle of incidence effectively imposes an upper-energy cutoff for the beamline. To increase the energy cutoff, therefore we need, a metallic coating with a higher density, and/or a decrease in the angle of incidence of the mirror. Platinum is a good mirror coating for higher x-ray energies, however the Pt Ledges present problems at lower energies, especially as Pt is an important catalysis material. It was therefore necessary to incorporate a second toroidal mirror, that could be translated into the beam, to maintain a clean energy spectrum across the entire 2.035 to ~33 keV energy range. A chromium coating has been selected for the second mirror with the Pt mirror covering the energy around the Cr K-edge at 5.989 keV (Fig. 3). To ensure that the mirrors could be translated seamlessly and the focal spot to remain in the same position, the incidence angle for the two mirrors must be identical. The mirrors will use an angle of 2.5 mrad as this is the best compromise between the delivered flux for both high and low energies. One problem with operating x-ray mirrors at very low angles is that any ‘real’ mirror has a finite length and the incident beam from the monochromator will overspill the mirror at low angles resulting in a loss of transmitted flux, especially at lower x-ray beam energies where the vertical fan size increases. Considerable care has been taken when specifying the mechanics for these two mirrors to enable the focused beam to remain at the same point in space as the mirrors are translated across the beam. It is non-trivial task to align two toroidal mirrors on a mechanical translation slide, as any small misalignments are magnified by the 20 meter lever from the mirror to the focal point of the beam. The motorized step resolution for the yaw motion will be <5 μrad and <5 mrad for the less sensitive roll motion. Both mirrors will have a new fixed sagittal radius and variable meridional radius, the latter being controlled by independent mirror benders. This proposed optical solution will deliver a clean energy spectrum from 2.035 to approximately 33 keV.

SHADOW calculations [9] were performed to estimate the total flux transmitted assuming a 1.2 m long toroidal mirror, a 2 mrad horizontal fan of bending magnet radiation and an incident angle of 2.5 mrad. The mirror width was set to 120 mm, with the sagittal and tangential radii set to their theoretical values calculated by SHADOW. Incorporating realistic parameters for the optical elements has enabled us to model the new source characteristics as functions of both energy and focal length. We estimate the new focal spot size ~(60 m horizontally x 70 m vertically) will be significantly smaller than our current focused spot size of ~(300 m horizontally x 600 m vertically) but with a comparable flux (Fig. 4). As the source point has moved, aberrations in the beam shape are evident at the current diffractometer position but are minimised at the 1:1 focused position. Our experimental hutch will be extended by 4 m downstream which also allows additional beam defining and conditioning elements to be incorporated into the delivery design, increasing operational efficiency and allowing rapid switching between experimental configurations. As the majority of experiments performed on *XMaS* require a small footprint/small angular divergence, the more brilliant source increases usable flux density for most users by one order of magnitude or more. Further increases in the S/N ratio are derived from the reduced background as the beam is no longer being defined close to the sample.

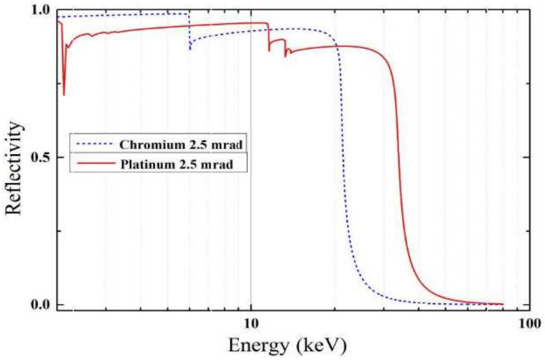

FIGURE 3. Reflectivity of the two different metallic mirror. The red and the blue curves show the reflectivity of the platinum and chromium mirrors at 2.5 mrads angle of incidence.

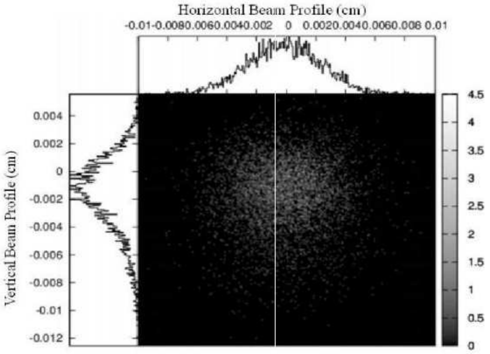

FIGURE 4. Shadow calculation of the focused beam profile at the new focal point, using a 1.2 meter toroidal mirror and 2 mrad fan of BM radiation. The focal spot size is approximately 60 μm horizontally by 70 μm vertically.

Ancillary Optical Components –Harmonic Mirrors

The beamline currently has two additional mirrors further downstream from the focusing toroid for the rejection of higher energy harmonics coming from the Si<111> monochromator. These mirrors are each 30 cm long and are fabricated from pyrex blocks, with part of the mirror coated with a rhodium stripe. For operation in the 6-15 keV region, the rhodium stripe is used and the pyrex section of the mirror below 6 keV. As the beamline will be operating above 15 keV post EBS, these mirrors will need to be upgraded to enable the transmission of higher energy x-rays. Also, as the beamline is now performing long energy scans (~800 eV) that are routinely used for XAS studies, the pyrex mirrors contain many elements, such as calcium, that can complicate any data analysis. Both of these harmonic rejection mirrors will be replaced by 30 cm long silicon mirrors, with rhodium and platinum stripes. All of the current mechanics that can perform a translation across the beam and adjust these mirrors up to 7 mrad angle of incidence will be maintained.

Ancillary Optical Components – Phase Retarders

The X-ray beam can be further conditioned by means of phase retarders (PR) to produce circularly polarized photons. The *XMaS* PR assembly was initially designed to perform polarization studies on rare-earth based magnetic materials in the range of 7-9 keV where most of the $_{L2,3}$ edges of the rare-earths are [10,11]. The scientific interests at *XMaS* have diversified over the years with a strong need for low energies such as the S Kedge (2.47 keV) for catalysts containing sulphur for example. Nowadays, four different PR crystals are used to cover the energy spectrum between 2.4 and 13.5 keV mostly for resonant magnetic reflectivity studies [12,13,14]. As more space will be available inside the extended experimental hutch after the EBS upgrade, the new PR assembly consisting of an Eulerian cradle with a high resolution circle for fine adjustment of the PR Bragg angle, will be mounted inside an in-vacuum chamber to allow windowless operations. The four PR crystals will be permanently mounted on a piezo flipper device, the later being used to reverse the handedness of the x-ray beam helicity between left and right at a few hertz [15]. A linear stage will allow the crystals to be driven in and out of the beam.

CONCLUSIONS AND FURTHER DEVELOPMENTS

A new optical configuration has been proposed for the *XMaS* beamline at the ESRF to maximize benefits from the EBS upgrade program, extending energy range of the beamline from 2.035 to ~33 keV. The water-cooled Si<111>monochromator will be upgraded to LN_2 cooling and two interchangeable toroidal focusing mirrors will replace the current toroidal mirror. These upgrades will increase the power density at the sample position by an order of magnitude due to the smaller focal spot size of some 60 μm × 70 μm. These upgrades will be supported by improvements in the control systems, upgraded to current standards. To facilitate rapid configurational changes, the spec control sessions will be consolidated and refined. The beamline diffractometer will run in a "psic" geometry [16] and upgrades will be in place to allow rapid energy scanning. Enhanced data pipeline to data visualization will ensure smoother user operations and more rapid impact and dissemination.

We have discussed an upgrade program that will deliver the exciting science that the EBS offers if the new source characteristics are to be fully exploited. After 20 years of operations we can make strategic use of the ESRF dark period to make the timely and necessary upgrades needed. The upgrade plan ensures competitive viability for the next 10 to 15 years and has been prioritized by our users. As well as the identified hutch extension, new mirrors and optimized monochromator we also plan to refurbish the diffractometer. To ensure sample environments are positioned rapidly, reproducibly and reliably, new beam positioning diagnostic tools and upgraded beam condition optics will be installed in the extended experimental hutch. New gas and liquid handling systems have been identified by users as critical for *in-operando* studies as well as a fast shutter and extension of the current SAXS system. We will invest in a new suite of detectors as the efficiency of Si-based systems degrades above 18 keV. Our detectors will include a Pilatus CdTe 100k for high energies and spectroscopic studies will be supported by a multi-element energy dispersive detector and fast MCA card. The current obsolete MAR CCD camera will be replaced with a Pilatus 1M-S. A small Eiger 500k detector with 2 s readout time will facilitate dynamic experiments.

Post EBS, the beamline will have more than one order of magnitude gain in usable flux density for most experiments with a smaller focal spot and access to higher x-ray energies and we believe this is the start of a new operational period supporting exciting and evolving developments in materials science on the *XMaS* beamline.

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