UPGRADE PROGRAMME – PHASE I

CONCEPTUAL DESIGN REPORT

X-ray speckle dynamics and imaging

Current designated sector:	Facility goes to:
ID10A (Troika I + III)	ID22

1.1 ID CARD

The beamline specialises in time-correlation spectroscopy (XPCS) and diffraction imaging with coherent X-rays (CXDI) to perform measurements of static and dynamic properties of condensed matter through analysis of speckle patterns. The main focus in on soft-matter samples (gels, glasses, complex fluids and colloidal suspensions) but also hard-condensed matter and biological materials are routinely investigated. Scientific questions that can be targeted include dynamical scaling, anomalous diffusion, jamming and non-equilibrium dynamics as well as high-resolution 3D diffraction imaging of cells and bio-matter. With the move to a dedicated straight section there is a unique opportunity to capitalise on ESRF's leading role in XPCS and to build the ultimate facility for speckle imaging and dynamics.

1.2 SCIENTIFIC CASE

ID10A is specialised in applications of coherent X-ray scattering to study slow dynamics of condensed matter by X-ray photon correlation spectroscopy, also known as XPCS (Grübel *et al*, 2008). The current emphasis is on soft-matter dynamics (e.g. colloidal suspensions, glass formers, liquid crystals, gels, and polymers) but dynamical phenomena in hard-condensed matter are also investigated (e.g. critical dynamics, phase separation, and domain fluctuations). Recently, another application has emerged, namely coherent X-ray diffraction imaging (CXDI), also denoted XDM (X-ray diffraction microscopy). CXDI is a lensless imaging technique based on the possibility to phase and invert the diffraction pattern by a combination of experimental methods (over-sampling of the coherent diffraction pattern) and by an iterative phase retrieval algorithm (Miao *et al.* 1999). With the move to ID22 there is a unique opportunity to build a beamline optimised for coherent X-ray scattering, capitalising on the world-leading role of ESRF in XPCS and targeting the exciting new possibilities provided by CXDI.

X-ray Photon Correlation Spectroscopy

The dynamics of systems undergoing structural changes is of fundamental interest in both basic and applied science. Dynamical scaling is a prominent example among dynamic out-of-equilibrium phenomena and it describes the fluctuations of domains and phase boundaries as order slowly develops in disordered systems. Such behaviour can be observed in certain classes of metallic alloys and compounds, where a temperature quench can initiate the formation of medium- or long-range ordering. In this field XPCS is a unique method and many of the early experiments were indeed studies of growth and dynamics of ordered crystalline domains. XPCS can be tuned to the desired length scale sensitivity by selecting the right Q and, recently, nearest-neighbour hopping of atoms into vacancy sites was for the first time observed in an Au-Cu compound (Leitner *et al*. 2009). XPCS on atomic and molecular length scales is extremely brilliance hungry and can only be performed when the maximum intensity is available for the experiments, i.e. using three short-period undulators operating in series on the fundamental harmonic with 200 mA in the ring. Hence, it is mandatory if this field is to continue prospering that XPCS gets the maximum benefit from the ESRF upgrade.

In soft materials, non-equilibrium phenomena such as dynamical aging have been observed in a number of systems, sometimes accompanied by structural changes and dynamical heterogeneity. Many aspects relating to jamming and gelation in crowded media remain to be understood and XPCS constitutes already now an important source of experimental data to complement the on-going efforts in theory and simulations by numerous groups worldwide. Recently, is has appeared from XPCS studies that significant deviations from the Brownian motion explained by Einstein 100 years ago can happen in sluggish media of high viscosity (Caronna *et al*, 2008). This discovery will have fundamental influence on the way we think about diffusion and is a consequence of the length- and time scale-dependent visco-elastic properties of fluids. In super-cooled simple liquids these changes manifest themselves already tens of degrees above the glass transition temperature by a mean-square displacement which varies faster than linearly in time (super-diffusion). Time-resolved multi-speckle XPCS is one of the few techniques capable of measuring the dynamical susceptibility that is necessary for understanding complex molecular mechanisms e.g. leading to the glassy state of matter.

The dynamic properties of surfaces is another field where XPCS is steadily contributing with new insight. For example, it was observed that the dynamic behaviour of partially wetting thin liquid films deviates significantly from the capillary wave predictions due to interactions with the substrate (Gutt et al, 2007) and partial wetting can in fact result in a solid-like behaviour of the liquid thin film. Biological functionality is often linked to distinct surface/interface properties and model systems of cell membranes have been investigated by XPCS targeting e.g. the influence of Coulomb interactions between phospholipid head groups and components in the aqueous sub-phase. XPCS experiments at buried interfaces (liquid-liquid, solid-liquid) remain an open challenge, necessitating a higher coherent flux at higher energies which is within reach through the proposed upgrade.

With a reinforcement of ESRF's XPCS capabilities the community may also gain new knowledge about reptation dynamics of macro-molecules. Despite the development of elaborate theoretical models, only very little direct experimental evidence exists on the dynamics of macromolecular melts, e.g. on the transition from reptational to segmental chain dynamics. Slow molecular dynamic processes like folding-unfolding are also essential for protein activity and XPCS could provide new insight e.g. concerning enzymatic functionality by studying the Q-dependent diffusion coefficient in solution quantifying the differences with respect to rigid-body motion.

Other interesting subjects which could be addressed thanks to increased coherent flux and the combination of resonant scattering and XPCS, are dynamical phenomena in rare-earth magnetism (anti-ferromagnetic domain dynamics, spin ice, etc..) directly accessible through the L-edge resonances. Further, the dynamics of ordering in strongly correlated systems like transition metal oxides may be targeted with the possibility to separate the charge and spin contributions and characterise e.g. the dynamics of sliding charge and spin density waves in relation to the evolution of the underlying quenched disorder.

Coherent Diffraction Imaging

The scientific case for hard X-ray CXDI has important contributions from both material science and life sciences. CXDI provides nanoscopic information and in the case of high resolution (~10 nm) imaging of micron sized objects it has several advantages over competing methods such as visible light microscopy, electron microscopy, and scanning techniques with nano-beams. Important new CXDI schemes like ptychography combines the best of diffraction and scanning microscopy for the study of extended objects. However, CXDI remains very challenging and its feasibility depends critically on the data quality which again depends on the coherence properties of the beam. Obviously, it can be advantageous to perform CXDI with soft X-rays if possible given the constraints on the sample environment and the setup (UHV required). Another important limitation of soft X-ray CXDI is the increase of multiple scattering and absorption, rendering the phase retrieval and the image interpretation rather challenging. Thanks to their unprecedented coherent flux the forthcoming X-ray free electron lasers will certainly outperform storage ring sources when it comes to single projection (2D) diffraction imaging, thus the efforts at storage rings must necessarily aim for high-resolution 3D CXDI of samples that are not easily renewable or are even unique.

The above considerations lead to the identification of 3D Bragg coherent diffraction imaging (targeting e.g. strain mapping of small crystals) and 3D CXDI on soft matter and biological samples (e.g. bacteria cells and viruses) as the most obvious candidates for hard X-ray CXDI at storage rings. Magnetic imaging of rare-earths accessible through the L-resonances is also possible, at least in principle, but it remains to be demonstrated and will require a very high brilliance. Another field which deserves attention is grazing incidence CXDI where imaging of buried interface structures should become feasible by an increase of coherent flux in the 15-20 keV range.

The proposed setup will target 3D CXDI in SAXS geometry in the energy range 7-20 keV which is well in line with the XPCS requirements. Bragg-diffraction CXDI and CXDI with nano-beams is covered by the UPBL1 project and will not be discussed further here. Topics of high interest that can be addressed in SAXS geometry includes bio-imaging (Lima *et al*, 2009) e.g. of sub-cellular organelles and high-Z element storage in living organisms. 3D imaging of soft materials e.g. micro droplets containing self-assembling sub-units, vesicles, and pattern formation in block co-polymers is highly demanded but only rarely feasible with today's techniques. Holographic CXDI approaches are promising in order to improve the reconstructions, and novel cross-correlation imaging techniques have been developed that allow uncovering hidden local icosahedral symmetry in colloidal glasses (Wochner *et al.* 2009). Those are amongst the new exciting possibilities that a soft matter CXDI station at ESRF could build upon.

1.3 PROJECT HISTORY

This document builds upon the CDR "XPCS-CXS", first published in the Purple Book. In May 2008 both beamlines ID10A and ID10B were reviewed and the panels strongly recommended to split the beamlines in order to: **i)** meet the requirements from ID10A for a higher brilliance and optimum coherence properties of the beam and **ii)** to allow ID10B's severely underperforming semi-transparent diamond monochromator (which continues to cause problems for the beamline operation) to be replaced. During autumn 2008 the present scenario emerged with ID10A moving to ID22, and ID10B sharing a future canted ID10 section with the beamline for surface and interface science (ex-ID03). At present ID22 is used for micro- and nano imaging/spectroscopy (EH1 and 2) and nuclear resonance (EH3), see Figure 1. These activities need to migrate to other beamlines before the transfer of ID10A can begin.

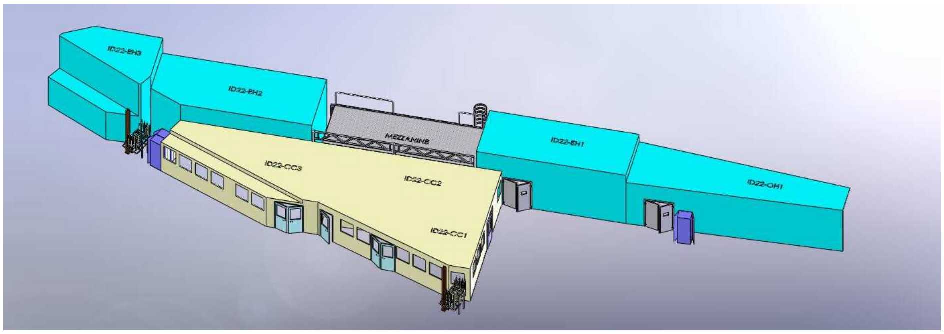

Figure 1. Outline of the existing hutches at ID22 (one optics hutch OH1 and three experimental hutches EH1, EH2, and EH3).

1.4 BASIC TECHNICAL CONSIDERATIONS

The decision to separate ID10A and ID10B, hence dedicating a full straight section to XPCS and CXDI is of vital importance for the further development of these activities. New, low-emittance sources in Europe and elsewhere are gaining on ESRF's leading role but as outlined above, exciting new science is within reach if we manage to gain about one order of magnitude in coherent flux on the sample. Today, ID10A is fully optimised for XPCS within the given constraints and is the benchmark in the world. Nevertheless, it is estimated that the desired gain is within reach (see below). Moreover, a stable operation with broad band pass (~1%) optics, e.g. a white beam mirror, would improve the current situation for "pink beam" CXDI and XPCS. Those are the goals of the new beamline.

Four technical key points can be identified where significant improvements are possible and required:

- *Source* – to achieve the maximum performance it is important that the beamline gets the highest possible brilliance. The usual working range will be 6-12 keV with the possibility to reach ~20 keV. In-vacuum undulators and a full 6 m (possibly 7 m in the future) straight section are mandatory.

- *White beam mirror* – This element is essential for the pink beam option and (optionally) to reduce the heat load on the components downstream. It should be specified to take the full beam in grazing incidence mode (cutting off at energies right above the undulator fundamental) with a minimum of aberrations and wave front distortions. A Si mirror body with partial Rh coating would suit for the working range ~6-20 keV. For pink beam experiments it should be possible to send the beam from the mirror directly into the experimental hutches without using a monochromator.

- *Monochromator* – High monochromaticity is required for XPCS and CXDI at large Q. A pseudo channel cut monochromator operating in vertical scattering geometry is proposed. It is ideal for the desired working range and excellent concerning stability and coherence preservation. A prototype of this mono is installed at ID10C but several points (e.g. the cooling and the vacuum vessel) must be improved. This work has already been initiated.

- *Detectors* – For XPCS the detector is almost as important as the beamline. We need fast, photon counting area detectors with as many pixels as possible and a maximum pixel size of 100 micron. Pixelated CMOS detectors (Medipix, Pilatus,..) and APD arrays belong to this category. It needs to be discussed whether ESRF should invest in a Maxipix-3 development and/or in the next generation Pilatus XFS detector which currently is being manufactured at PSI. It is expected that this detector will be commercialised by Dectris within the next 3 years. We should definitely be first in line when these detectors become available as they are fundamental for the scientific potential of the beamline.

The proposed new location at ID22 offers good possibilities for the development of the beamline. The existing lead hutch structure, the surrounding laboratories and the storage areas seem well adapted and can be used with only minor modifications. Parts of the instrumentation and optics from ID10A can be transferred without any problems and possibly integrated with equipment at ID22 that will remain in place. However, a serious commitment should be made towards the four main points/items listed above. This is described in more detail below:

The source

Initially, the existing U23 undulator can be kept and an additional 2 m long U20 in-vacuum device installed as soon as possible. When the ID22 section is ready to accommodate 6 m (or 7 m) of insertion device the U23 should be replaced by short period in-vacuum undulators (e.g. one long segment, two shorter) to fill the entire straight section. The details must be discussed and coordinated with the Accelerator and Source Division and the ID group. Altogether, this will increase the intensity by a factor of ~5 @8keV compared to the best situation today (when

ID10A and B can agree on common ID settings). With improved machine emittance, higher current, a vertically diffracting monochromator, and ID10B's mono out of the beam, one order of magnitude gain in brilliance is clearly within reach. This brings us into well into the 10^{21} (ph/s/mm^2/mrad^2/0.1%bw.) brilliance regime, with the possibility to match the NSLS-II and PETRA-III figures. For the fastest XPCS measurements it is important that the machine keeps the uniform filling pattern as an option. Top-up operation (like at APS or SLS) with "infinite" lifetime of the current would be very interesting for slow dynamics experiments.

White beam mirror

It is proposed to build a horizontally deflecting double mirror which can take the full load of the future source. Due to the source asymmetry, the coherence length is much larger in the vertical direction; this is the reason why a horizontal scattering geometry is preferred. The mirror should be located in a UHV vessel in the optics hutch about 30 m from the source and should accept the full vertical beam (~1 mm) and 0.4 mm in the horizontal direction (defined by slits due to coherence considerations and the acceptance of downstream optics). With a 2 m U20 undulator unit the unfiltered power is then ~55 W/100 mA i.e. ~500 W for 6 m U20 undulator at 300 mA (0.4 x 1 mm^2). The mirror should be fully illuminated along the beam direction to avoid a thermal bump. Silicon is proposed as material with a coated stripe of rhodium (Rh). Typical settings would be an incidence angle of 0.17 deg (3 mrad) which provides cut-offs at ~10 keV (Si) and ~21 keV (Rh). In this case the footprint of the beam is ~135 mm which should be equal to the length of the mirror. This gives a maximum power density of ~3.7 W/mm^2 at 300 mA. A vertical motion allows switching to be made between the Si and Rh stripes and the mirror must have state-of-the-art specifications. Both the slope error (goal: <0.3 μrad) and the roughness (goal: <0.1 nm) are critical to preserve the wave front. The detailed specifications and the cooling scheme need to be studied further. The pink beam power after filtering by the double mirror is ~10 W/100 mA for one U20 undulator, with the collimation given above.

Monochromator

A channel cut Si(111) geometry is proposed. With a 5 mm wide channel the desired energy range can be covered providing a quasi fixed-exit beam. The monochromator should operate in vertical scattering geometry and be non-monolithic, i.e. the two crystal surfaces must be polished separately before mounting on a common support. The support should feature the possibility to regulate the yaw, pitch and roll angles of the 2nd crystal. The cooling scheme of this device needs to be decided, based on a detailed study of the heat load; the optics hutch of ID22 is already equipped with liquid nitrogen supply pipes.

Detectors

It is mandatory to have a fast, photon counting 2D detector with sufficient resolution to map out the speckle patterns for CXDI and XPCS. Ideal specifications would be:

- Photon counting pixel detector (zero noise)
- 100 μm pixel size (or smaller)
- 1 MHz full frame readout

- Max. photon rate: 10^7 ph/pixel/sec
- 10^6 pixels

These numbers are strongly linked. For instance the requirements to dynamic range increase if the speed is reduced; the max photon rate is the decisive parameter and the counter depth must be adapted to the speed. For XPCS the detector should be as fast as possible. For CXDI the number of pixels is more important as it determines the ultimate resolution achievable. For example with 1000 x 1000 pixels a resolution of 100 (e.g. 15 nm features of a 1.5 μm object) can be obtained with a sampling frequency five times finer than the speckle size. If reduced sampling frequency can be tolerated (e.g. for 3D data) the achievable resolution increases proportionally. The pixel size required depends on the distance available from detector to sample, the size of the investigated objects, and the desired sampling. If 5 m is available, speckles from a 1.5 μm object may be over-sampled by a factor of five with 100 micron pixels @ 8 keV. For the moment no detector on the market fulfils the above specifications but several interesting R&D detector projects are in progress worldwide. The next generation Pilatus detector based on the XFS chip is the commercial product that best fits the above requirements and it will become available within a few years.

Instrumentation

The instrumentation and setups in the first and second experimental hutches must be adapted to our needs:

EH1: The beam is always delivered from the mono. This hutch should host a new four circle diffractometer with the possibility to carry heavy sample environments. Both horizontal and vertical scattering should be possible as well as a three-axis setup with an analyser. Grazing incidence scattering on liquids should be an option by using local mirrors. Fast point detectors (APD) as well as the above mentioned 2D detectors will be used. A He recovery line is already in place at EH1. An effort towards an extremely stable floor (for example through a thicker local concrete slab) as well as a noise-free electrical environment should be made.

EH2: Setup for coherent SAXS. The beam is delivered by the mono or by the mirror (pink beam). Local optics should allow for focusing as well as for a GISAXS option. The standard detector will be the 2D detector mentioned above. The sample-detector distance should be modular (as a minimum up to 5 m) and a long bench will host beam-pipes and detectors. A high precision goniometer (transfer from ID10A) will be located here for CXDI on small objects. EH2 is already equipped with advanced air conditioning and temperature control.

The existing EH3 will not be used; this lead hutch should be removed and the floor space used to host a sample preparation laboratory. The ID22 control cabins will be refurbished (floor, lighting, office furniture) but otherwise used as they are. At least two fast gigabit fibre connections to NICE should be foreseen. A local cluster (fast disk access, multi core environment) dedicated to on-line treatment of XPCS and CXDI data is highly desirable today and will become mandatory with the advent of ever-faster 2D cameras. Windowless operation of the beamline should be considered and space will be reserved for differential pumping. A sketch of the proposed beamline layout is given below:

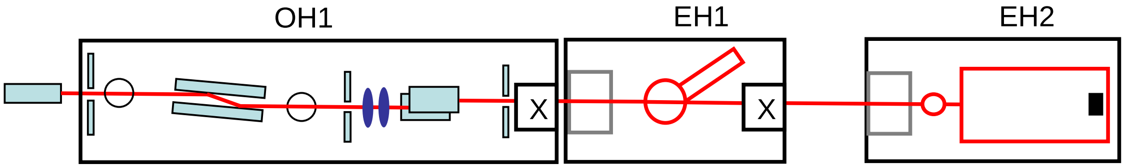

Figure 2. Sketch of the setup at the new ID22 beamline. The optics hutch OH1 hosts the mirror and the monochromator as described above. The new diffractometer is hosted in EH1 and the long detector table in EH2. The lens setup in OH1 will be transferred from ID10.

1.5 REFERENCES

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