

UPBL2: High energy beamline for buried interface structures and materials processing	
Current designated sector:	Facility goes to:
ID15	ID09

1.1 ID CARD

UPBL2 will provide the European scientific and industrial communities with an unprecedented tool for in-situ studies of deeply buried interfaces and novel materials processing at real working conditions. The relevant user communities are mainly from academic and industrial research on applied physics, materials science, interface science, chemical, mechanical and electrical engineering.

1.2 SUMMARY

Many countries around the world are currently proposing ambitious plans for reorganising the energy sector in order to meet major challenges such as reducing CO₂ emission and other greenhouse gases in accordance with the Kyoto protocol, reducing dependence on volatile states and securing a stable supply of renewable energy. These challenges require the development of new advanced materials and devices such as fuel cells, organic solar cells, rechargeable batteries, catalytic materials, etc. The complexity of these heterogeneous devices can only be studied adequately by a combination of experimental methods which can reveal the interplay between the microscopic material properties and the macroscopic device performance. As has the need for combining techniques been instrumental in the development of electron microscopy, it is seen as equally important for the evolution of in-situ hard X-ray synchrotron methods for studying both real devices under real operating conditions and idealised model systems under precisely controlled environments.

UPBL2 therefore enables a portfolio of hard X-ray characterisation techniques including reflectivity, wide angle diffraction both in transmission and grazing incidence geometry, small angle X-ray scattering, and imaging methods coupled with a great versatility in choosing beam sizes and detectors optimised for high energy X-rays.

1.3 PROJECT HISTORY

The existing high energy beamlines ID15A and B are unique since they are capable of delivering white and monochromatic X-ray radiation in the energy range 50-750 keV. The main research fields are surface science and the study of the buried interfaces and engineering materials research. The development of fast micro-tomography has added another dimension to the study of materials. Currently, the beamlines are highly over-subscribed and many strongly rated proposals can not be accepted due to an insufficient amount of available beam time. The proposed high energy upgrade project in the ESRF Purple Book was under the acronym HIENE which was an evolution of the existing high energy beamline ID15.

The last beamline review panel in May 2007 was convinced that there is insufficient provision of high energy X-rays at the ESRF and in Europe in general. One of the recommendations was to provide an additional high energy X-ray beamline with energies in excess of 100 keV and particularly up to 150 keV. Furthermore, the panel recommended that ID15 should focus and expand in the Upgrade highlight areas, interface scattering, ultra-fast tomography and materials engineering, and the beamlines should be rebuilt to take the full advantage of new generation optics. These recommendations, together with the exciting new scientific opportunities that will arise by further reducing the focal spot, led to this Upgrade project.

Now it is foreseen that the activities using very high energies above 150 keV, namely white beam radiation or circularly polarised radiation from the wiggler source, will stay at the existing end-station ID15A, whilst the imaging activities, the studies of buried interfaces and the angular dispersive diffraction studies using 50-150 keV radiation will be combined in a unique instrument in the Upgrade beamline UPBL2.

A brainstorming meeting was organised on the 9-10 December 2008 to assess and further develop the scientific case. Expert users covering areas of present and possible future research using high energy radiation for the studies of buried interfaces and for the in situ studies in chemical and materials processing gave input on what UPBL2 should offer to the user community.

1.4 SCIENTIFIC CASE

Technical premise

At high energies, the diffraction, even at large momentum transfer, is in the forward direction. This, together with high penetration, enables one to study samples in transmission geometry in harsh environments using reflectivity, imaging, small angle X-ray scattering (SAXS), and diffraction techniques. These techniques cover all length scales from atomic level up to millimetres as seen in Figure 1.

Even if the simultaneous use of these techniques is not usually possible, the switch from one technique to another will be fast. Imaging or SAXS techniques can be used truly simultaneously with diffraction, if one sacrifices the low momentum transfer information within the wide angle data. In addition, this limitation may also be overcome with future detector developments. Since there is space available around the sample other methods such as IR spectroscopy, mass spectroscopy,

fluorescence analysis etc. can be used truly simultaneously with the above mentioned X-ray methods. Furthermore, there is room for complicated auxiliaries.

The diffraction in the forward direction also simplifies the optics at high energies. Wide band-pass bent Laue monochromators or bent multilayers, both in fixed-exit geometry in the *horizontal* plane, are used together with refractive lenses to focus the beam to the sample. By having refractive lenses at different distances, users can select easily the desired beam size ranging from millimetres down to sub-micron length scales thanks to the long beamline. For a given focal spot size, the longer beamline also offers more space around the sample and beam divergences can be kept small ensuring good resolution in diffraction. The long focal depth is another important advantage offered by the long focal lengths and thick samples can be measured in transmission geometry without losing the resolution.

The design of beamline UPBL2 matches a large fraction of the requests from several areas of scientific interest and enables many different studies with tremendous potential. In the following we provide a few examples of scientific cases that will benefit greatly from the multiple techniques offered by UPBL2.

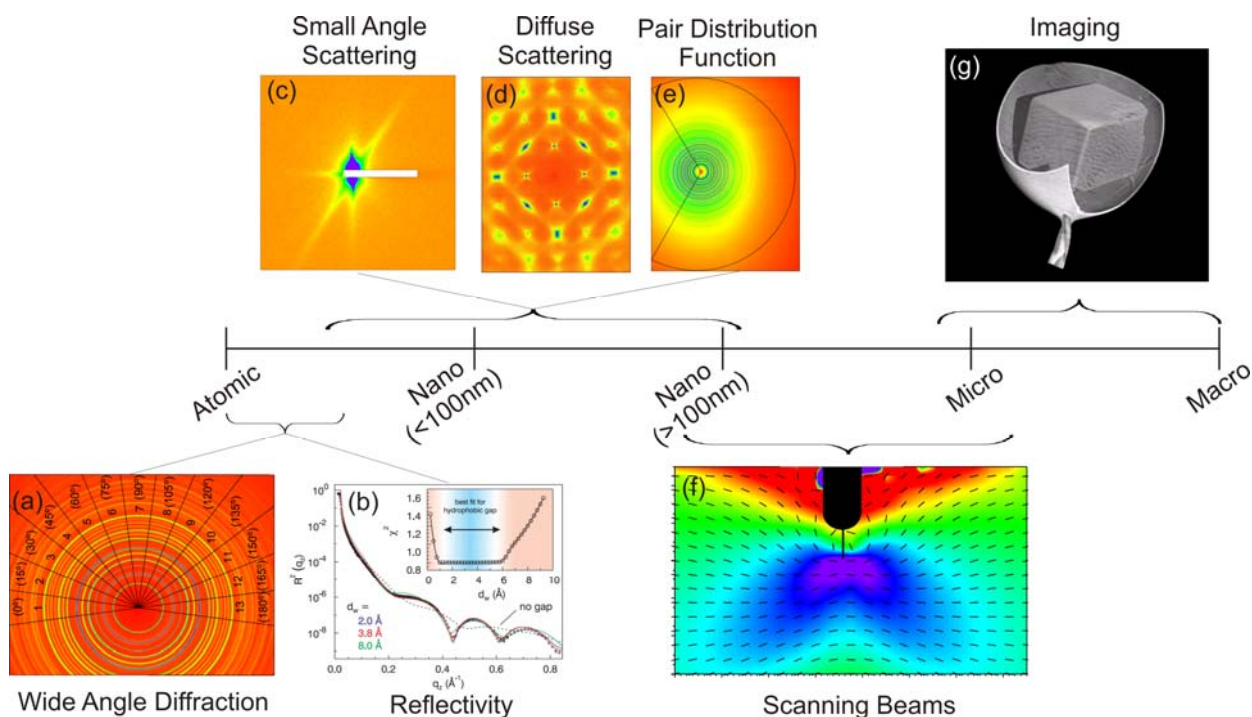


Figure 1. The multiple length scales covered by different techniques available at UPBL2: a) wide angle scattering from textured Lead Zirconate Titanate, b) reflectivity study of the hydrophobic gap at the water–OTS interface c) small angle scattering from nano precipitate hardened metallic composite, d) diffuse scattering from PZN-PT single crystal containing polar nano regions, e) high q ($>30 \text{ \AA}^{-1}$) scattering from Fe for PDF analysis, f) strain map around a loaded crack in Zr (150 \mu m resolution), g) phase contrast tomography image of protein crystal.

Buried interfaces

Room-temperature ionic liquids are promising candidates for a broad range of "green" applications, such as solvents in green chemistry, catalysis, batteries, and fuel and solar cells, for which their interaction with solid surfaces plays a crucial

role. They are interesting materials from a physical point of view as they allow us to modify the interactions between the molecular ions, as well the interaction with other materials by simply modifying the individual ionic molecules chemically. Up to now several thousand ionic liquids have been synthesised and many of them are available from commercial sources. Although crucial for the understanding of their properties in applications, little is known about the structural arrangement of the anions and cations at solid interfaces. A recent study on ionic liquid layering on charged sapphire interfaces (Mezger et al, 2008) is a very good example of what reflectivity studies at very high X-ray energies can offer. The high energy is, of course, a prerequisite for penetrating to the deeply-buried interfaces.

Liquid-liquid electrode interfaces are important in **electrochemistry** due to their ideal, structureless nature. X-ray reflectivity can provide detailed information on the potential dependent surface induced layering, electrocapillarity effects, and ion distribution. Due to the large path length (cm), a requirement to have sufficiently flat samples, high energy photons are essential. At the mercury-electrolyte interface, unpublished studies by Murphy B, Magnussen O, Deutsch M & Ocko B have shown the existence of mercury surface induced layering in mercury at the interface with non-adsorbing electrolytes. These are in reasonable agreement with previous observations at the vapour interface (Magnussen et al, 1995) and provide new information on the potential dependent roughness associated with electrocapillarity. This initial study provides the basis of a wide variety of future electrochemical studies. In a related study, the Schlossman Group (UIC) has investigated the liquid-liquid interfaces between an aqueous solution of hydrophilic ions and a polar organic solution of hydrophobic ions (Luo et al, 2006). They have used the potential of mean force in a generalised Poisson-Boltzmann equation to predict the full ion distributions. These distributions agree with the measurements without any adjustable parameters. These first studies have clearly shown the feasibility of this approach, yet to date this field is in its infancy.

Beyond reflectivity, grazing incidence diffraction (GID) and diffuse scattering allow the in-plane characterisation of liquid-liquid and solid-liquid interfaces. A combination of the three scattering techniques allows a very complete characterisation of interfacial phenomena, for example, in-plane and out-of-plane organisation and interfacial tension (Sanyal et al, 2008). UPBL2 also offers unique X-ray optics for liquid surface and interface studies (Honkimäki et al, 2006). This optics device is ideally suited for experiments where the sample has to be kept stationary during measurement. Another characteristic of the proposed beamline is sub-micron focussing. By reducing the beam size the bulk scattering volume can be reduced, and hence the signal/background ratio in reflectivity and GID measurements is improved. Furthermore, smaller beams allow a better definition of the geometry and help meniscus handling in liquid-liquid interfaces. These are major points, since at present GID studies of buried liquid-liquid interfaces are practically nonexistent.

In-situ experiments on in-plane ordering in adsorbed layers, particularly biomolecules are of great interest. Adsorption of various molecules on a variety of solids also has implications for catalysis, chemical reactions etc. Other interesting issues are structural transitions at solid/liquid interfaces in adsorbed layers. For example, a system of choice for studies of molecular recognition at the border of biology and materials are self-assembled monolayers of DNA brushes (Opdahl et al, 2007). When immersed in water, they can change from extended to collapsed

states under changes in pH. Their in-plane structure in vitro is not known. The in-plane order evolution in the process of self-assembly itself can also be studied, from its earlier stage, by GID.

The beam's small cross-section allows smaller substrates to be used. This is not just a convenience, but sometimes a necessary condition. For example, a fundamental question of great scientific interest is the reason why molecules in living systems employ **homochirality**. Answering this question requires the combined efforts of several fields of enquiry, including chemistry, biology, theoretical and experimental physics. Furthermore, approximately one third of all therapeutic drugs on sale today are based on chiral molecules. Understanding the interactions, self-assembly and crystallisation of chiral molecules is therefore of significant interest also for industry and technology. The study of chiral recognition and discrimination is much less complicated in 2D systems (Weissbuch et al, 2005). A great deal of experimentation has been performed on monolayers of chiral molecules at the water/air interface (Nandi et al, 2003) and crystallisation processes from bulk solutions (Weissbuch et al, 2005). More challenging is the determination of the molecular ordering effects taking place at the interface between a bulk solution of chiral molecules and a solid surface, particularly when the latter is a specific facet of a chiral crystal.

UPBL2 would allow detailed investigations of these previously mentioned aspects by combining:

- *The use of sub-micron beams to perform GID experiments at buried solid-liquid interfaces.* This would allow in-situ monitoring of the evolution of the in-plane structure of the interfacial monolayers of chiral molecules during their adsorption. The smaller beam size would decrease the background from the bulk liquid, thus improving the detection of weakly diffracting interfacial layers.
- *The use of 2D detectors optimised for high energy in reflectivity measurements.* This would improve in particular the delicate issues of background subtraction in the case of weakly scattering soft materials at buried interfaces.
- *The application of time-resolved reflectivity measurements to study interfacial reactions or adsorption at interfaces.* A convergent beam with a small focus can be used to measure a large part of the reflectivity curve simultaneously as well as the background. This has the advantage of having a fixed footprint, and allowing a fast measurement, so that the kinetics of interfacial adsorption processes could be measured with good time resolution.

Time-resolved measurements of **crystal truncation rods** (CTRs) using 2D detectors will also be one of the areas of further development at UPBL2. Since the Ewald sphere is relatively flat at high X-ray energies, it is possible to intersect a sizable region along one or more CTRs. Collecting data along a large section will enable real-time measurements of surface structural modifications from chemical processes, such as catalysis reactions. Pilot measurements were made on a sample with faceted, nano-particles of Pd supported on a MgO substrate (Nolte et al, 2008). Several CTRs from the nano-crystals were captured in one image. While the test measurements proved the concept can work, it will greatly benefit from increased X-ray flux and a higher efficiency 2D detector available at UPBL2.

Imaging

Since about half of raw materials come as a solid and are thus processed as granulates, questions on the mechanical behaviour of **dry and wet granulates** are not just of fundamental scientific interest, but also have a large impact on industrial processing. Amongst them, mixing, mechanical stability, and material transport are probably the most important and apply to the concrete industry, food industry, pharmaceutical industry, geology etc.

Characterisation techniques with high time resolution are essential to study cracks forming in dry or wet granulates under mechanical stress/strain and to study the mechanical behaviour – the flowing – of slurries and suspensions. The ultra fast imaging and micro-tomography techniques developed at ID15 are unique, exploiting pink beams from a powerful high energy undulator, a high speed and high accuracy rotation stage and the associated triggering capabilities, high luminosity optics, and high efficiency scintillators with short decay times. 3D particle imaging using a stereo beam technique recently developed at ID15 allows questions to be answered regarding the particle dynamics and thus to study the mixing behaviour of slurries and suspensions. Moreover, it allows flow velocities of liquid in confined geometries to be measured by tracking single X-ray absorbing particles suspended in the liquid. This technique can be used also to visualise fluid dynamics in porous materials with much better temporal and spatial resolution than any other technique to date.

Imaging **liquid fronts** with millisecond time resolution can answer a huge range of different fundamental questions such as studying the effects described by Washburn law or Darcy's law on a pore size level and not just on the level of an average porosity, as done to date. Applications of these questions can be found in e.g. impregnation, liquid infiltration causing frost damage of streets and houses, storage of nuclear reactive waste, agriculture, environmental geology (cleaning of soil from hydrocarbons), water reservoirs, oil mining, liquid transport in fuel cells, engineering of filter cakes, water separation (gas/chemical processing), water evaporation from reservoirs, spreading of liquids into (woven and non-woven) fabrics, printing techniques, polymer injection moulding, fibre-reinforced polymers, etc.

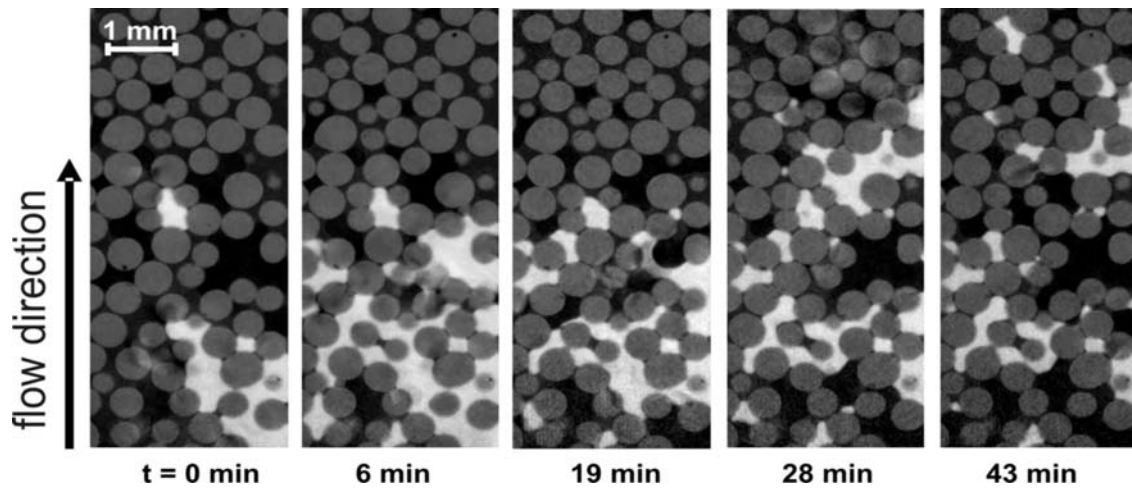


Figure 2. Plain section of a time series of X-ray micro-tomograms, where a water front (white) is driven into oil filled (black) pile of spherical glass beads (grey).

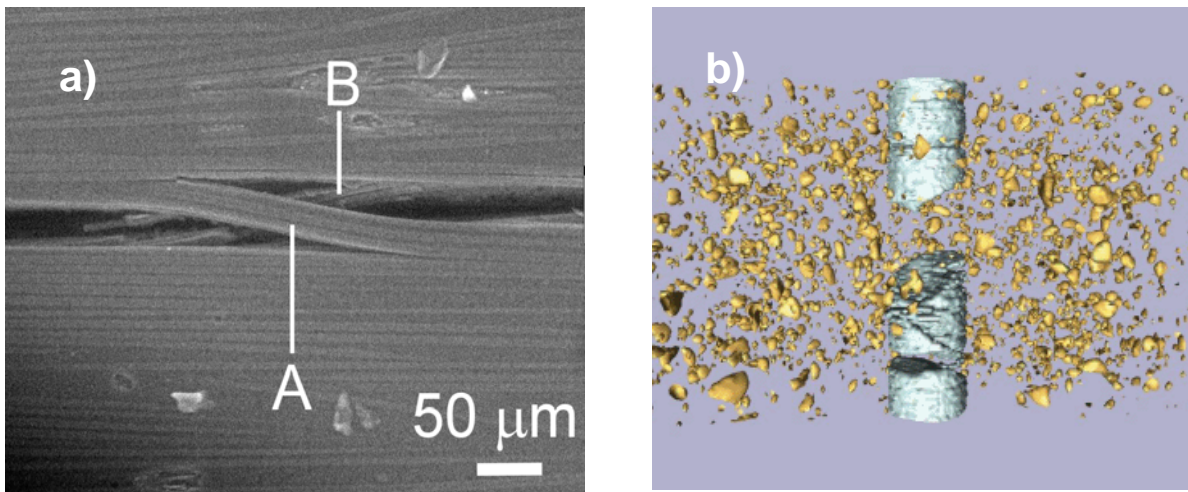


Figure 3. a) Interface cracking in an epoxy-glass fibre composite as observed on the surface by SEM. Courtesy of B. Sørensen. b) Reconstructed volume of a glass fibre (grey) – epoxy (transparent) composite with glass marker particles (yellow) from tomography. The fibre breaks after straining are evident.

To unravel these phenomena the topography of the matrix and the liquid front has to be imaged with excellent lateral and temporal resolution. This is needed to freeze the motion of the liquid and to follow the dynamic redistribution within a liquid front based on the local Laplace pressure and the flow resistance. An example of this kind of dynamic redistribution is shown in Figure 2, where a water front is driven into oil filled matrix consisting of glass beads. This time series reveals a major problem in oil mining, why only a fairly small percentage of the existing oil can be recovered: As the driven water front proceeds it drives the oil out, cf. image at 6 min. But as the water front proceeds further and roughens, the formerly water filled areas were filled again by oil, cf. image at 28 minutes. The oil that remains behind the water front is lost for the oil recovery. Since these effects are strongly dependent upon the driving speed of the water, in this example, 3D high speed imaging is mandatory to develop a detailed understanding of these effects.

The tomographic studies during *in situ* tests of **structural materials** are similar to a 3D extensometer, giving access to global and local (in the case of heterogeneous structures) geometric information. The global information can be exploited to evaluate the activation energy of steady-state creep on one single specimen having a constant temperature gradient along the sample tensile axis (Sket et al, 2008). Local information combined with powerful image correlation algorithms can be used to evaluate the volume growth-rates of single cavities, giving much more accurate experimental results than any previously reported (Isaac et al, 2009). This makes possible, for the first time, the test of existing creep damage theories. Since the tomographic resolution below micrometres cannot easily be obtained at high energies, the SAXS information can answer some questions in the nanometre regime.

Energy sector and hybrid methods

Modern wind turbine blades are made from advanced fibre composites of long oriented fibres in a polymer matrix. The components are inherently multi-scalar, with defects appearing at all length scales and effecting mechanical properties. An ultimate aim is therefore to establish a consistent hierarchical framework of 3D characterisation tools and 3D modelling tools that can predict the mechanical properties (strength and fracture toughness) of composites from the basic properties of fibre, matrix and interface, accounting for the presence of defects. An example of a typical crack is shown in Figure 3a. The experimental requirement is to perform *in situ* 3D mapping of the topology of the structure at the various length scales and its evolution, 3D mapping of the elastic and plastic strain fields and characterisation of the buried interfaces using sub-micron beams.

UPBL2 would allow comprehensive characterisation of these materials by combining:

- *X-ray tomography* for characterising fibres, crack openings and voids with a spatial resolution of several micrometres
- *Plastic strain tomography* for characterising the plastic strain field around the cracks and other defects (Nielsen et al, 2003; Haldrup et al, 2008). This technique is illustrated by Figure 3b, showing a detail from a 3D tomographic reconstruction of a glass fibre embedded in epoxy. A homogeneous distribution of small glass marker particles is also seen. The plastic strain can be derived by tracking the movements of the hundreds of thousands of particles
- *Maps of the elastic stress*. A new wide-angle X-ray scattering technique, capable of determining the local stresses in amorphous materials has been demonstrated at beamline ID15B (Poulsen et al, 2005). It was shown that the local elastic strain tensor in a bulk amorphous alloy can be determined with accuracy in the strain components of 2×10^{-4} . Characterisation of fields within fibres would be possible.
- *Interface characterisation*. By applying diffraction techniques in the vicinity of fibre interfaces using a 100 nm beam, it would become possible to elucidate alterations in the structure of the polymeric matrix.

The potential applications of the **solid oxide fuel cells** (SOFCs) range from auxiliary power supplies in trucks and ships to power plants in sizes from a few 100 kW to several MW. The first commercial penetration of niche markets is expected to occur within the next couple of years. A solid oxide fuel cell consists of

an anode and a cathode, separated by a ceramic electrolyte. The anode and cathode are both porous to allow fuel gas and air, respectively, to enter them. The relevant chemical processes producing the electricity occur at the embedded three-phase boundary where electrode, electrolyte and gas phase meet. The performance of the cell therefore depends crucially on the details of the microscopic structure at the boundary as well as the surrounding electrode (e.g. porosity and particle size) and the electrolyte.

UPBL2 would enable unique characterisation in several ways:

- *Redox chemistry of electrolyte/anode interface.* In situ studies of the phase and stress gradients near the interface are of major interest. Even more ambitiously, one may consider characterising the local electrochemistry as a function of temperature and partial pressures of various gases.
- *Grain resolved studies.* With grain and pore sizes of the order of micrometres, 3DXRD microscopy and tomography are ideal tools for structural characterisation of fuel cells or parts of fuel cells (Hagen et al, 2006). Notably, several key structural properties, such as the length of the three-phase boundary and the degree of percolation, can only be measured in 3D. Likewise, heterogeneities are a main concern, and it is of interest to follow – in situ or ex situ – the phase transformations during operation, which take place at ~800°C. Hence, use of these tools will not only provide qualitative information on phenomena that could not be visualised before, but is also seen as a prerequisite for boosting modelling efforts.
- Using high energy imaging techniques to observe the degradation of fuel cells –interfaces and their breakdown – in operation.
- Since the tomographic resolution better than micrometres cannot be obtained at high energies, the SAXS information can help to understand the breakdown of interfaces in the nanometres regime.

Another notable class of fuel cell is based on the use of a **polymer membrane** as the solid electrolyte. Since these cells work at moderate temperatures (100°C or less), they can be used in ordinary environments, for example as power sources of electric cars. The amount of water and its spatial distribution in the polymeric membrane of the device under working conditions is a fundamental point to address since the ionic conductivity and, consequently, the cell performance, depends on the hydration degree of the membrane. The few experimental techniques used so far to measure the water distribution in the membrane suffer from a rather low spatial resolution (if any). Moreover, they have other severe limitations, such as slow response to variations in the degree of hydration, a weak signal resulting in poor accuracy and an indirect dependence only on the quantity of interest.

The high energy X-rays available at ID15 enabled the first space/time resolved measurements of proton exchange membrane hydration to be made in a running fuel cell (Rossi Albertini et al, 2009). A vertical stratigraphy of the membrane was carried out, from one electrode to the other, corresponding to an imaginary “slicing” of the membrane into a stack of layers. Both the hydration degree in each layer and the overall amount of water in the membrane could be determined as a function of time, with the highest level of accuracy ever achieved. The further decrease of the beam cross section, down to 100 nm, expected in the new beamline, would represent a substantial improvement in the accuracy of the water dynamics studies,

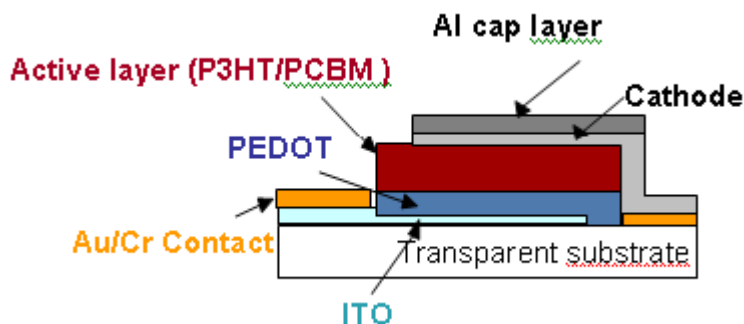


Figure 4. Plastic solar cell.

especially in the vicinity of the electrodes catalysts/polymer membrane interfaces, which represents a fundamental task for any future development of these devices.

New **electrochemical devices** of the uttermost importance are the **plastic solar cells**, which are based on a donor–acceptor bulk hetero-junction, obtained by blending two materials, one acting as electron donor, a conjugated polymer, and the other as electron acceptor (C60, or carbon nano-tubes). Potentially they can replace the doped Si based technology, which is relatively expensive and much less flexible. The components form a stratified medium (Figure 4), which can be studied using X-ray reflectivity (Paci et al, 2008) as a probe of the buried interfaces between adjacent layers. The time resolved reflectivity information obtained on the interfaces upon cell working can elucidate the relation between morphological degradations and fading of the cell electrical performances. Furthermore, with sub-micron focusing UPBL2 can provide an unprecedented tool for detailed in situ studies of the interactions between adjacent layers in the photovoltaic cell and to follow in real time small morphological changes.

Newton et al (2009) have just performed a successful experiment at ID15B on **catalytic** materials. They demonstrated a new experimental methodology combining time resolved hard X-ray diffraction, diffuse reflectance infrared spectroscopy, and mass spectrometry for the in situ time-resolved study of supported Pd nanoparticles during CO/NO cycling. The results obtained went beyond the original expectations and led directly to a number of new and potentially important lines of research in this field. The diffraction data up to very high momentum transfers revealed a variety of the structural reactive behaviour enhancing greatly the knowledge already obtained from the equivalent time resolved EXAFS studies made on ID24 (Newton et al, 2007). Moreover, novel aspects of the catalyst behaviour, which are either invisible or extremely difficult to quantify using EXAFS, were clearly observed in diffraction. Improved time resolution in diffraction at UPBL2 and SAXS information would reveal even more information of the oxygen storage and release functions, which lie at the very heart of their commercial utility as components in auto-exhaust and other types of catalyst.

1.5 TECHNICAL CONSIDERATIONS

The most important areas for the future technical developments in the high energy X-ray fields are i) cryogenic undulators, ii) focusing techniques and iii) more efficient detectors for high energies. Together these improvements constitute a revolutionary enhancement in performance enabling studies of the dynamics in materials as well as the studies of the kinetics of phase changes on new time scales.

Source

The Upgrade Programme will provide new sources of hard X-rays by implementing cryogenic undulators, with the added benefit of higher ring currents resulting in unprecedented X-ray flux in the 50 to 150 keV range. Two such devices should be installed, if the source size is not compromised. The in-vacuum undulator U22 from ID15 can be used in the starting phase. The front-end horizontal aperture should be ± 1 mm at 14 m from the source and the vertical aperture ± 2 mm at 23 m from the source. This will result in an aperture of $3.86H \times 4.70V$ mm² at 27 m, primary slit position, from the source. This large beam is needed for the imaging experiments. Since most of the experiments are done with focused beams, any improvement in vertical emittance is important.

Hutches

It is foreseen to build UPBL2 on the ID09 straight section. The layout of UPBL2 is shown in Figure 5. The second optics hutch (OH2) and the experimental hutch (EH) are shown in Figure 5. The second optics hutch (OH2) and the experimental hutch (EH)

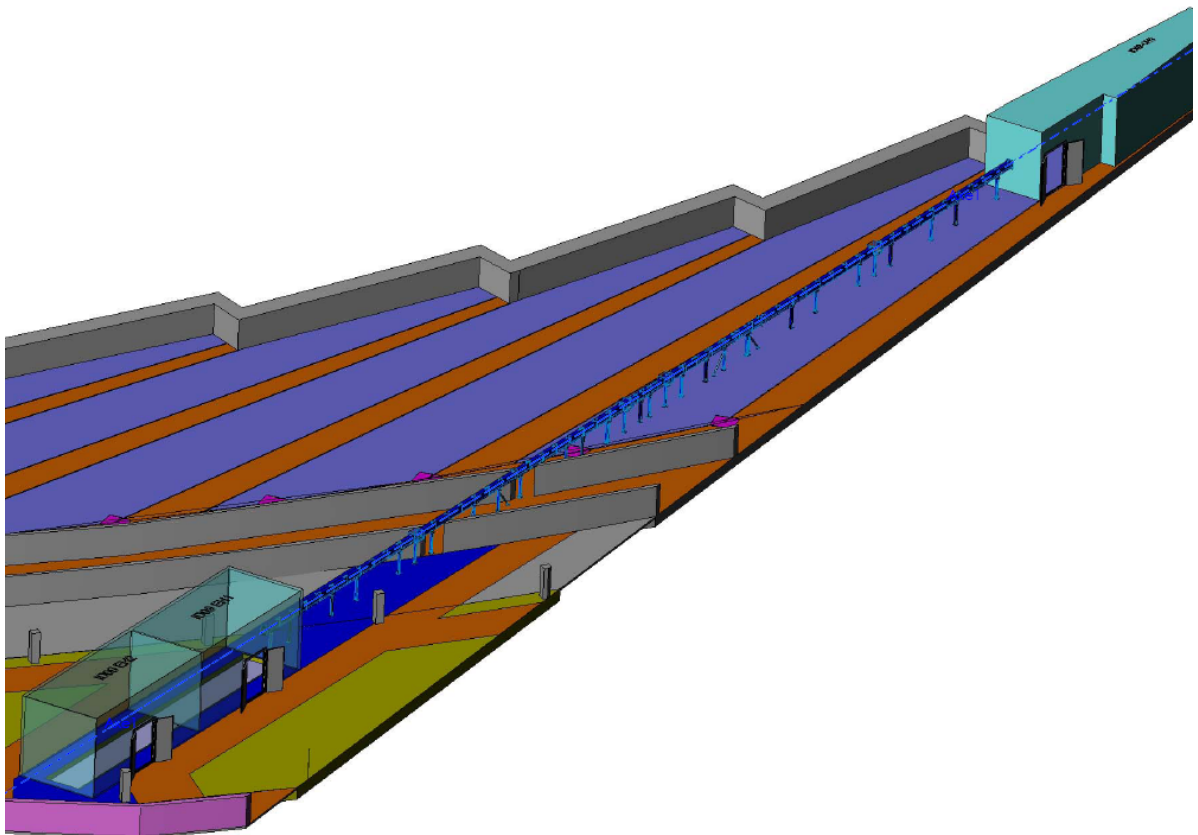


Figure 5. Layout of UPBL2. The first optics hutch is shown on the right and the second optics hutch and experimental hutch are on the left extended into the new experimental hall.

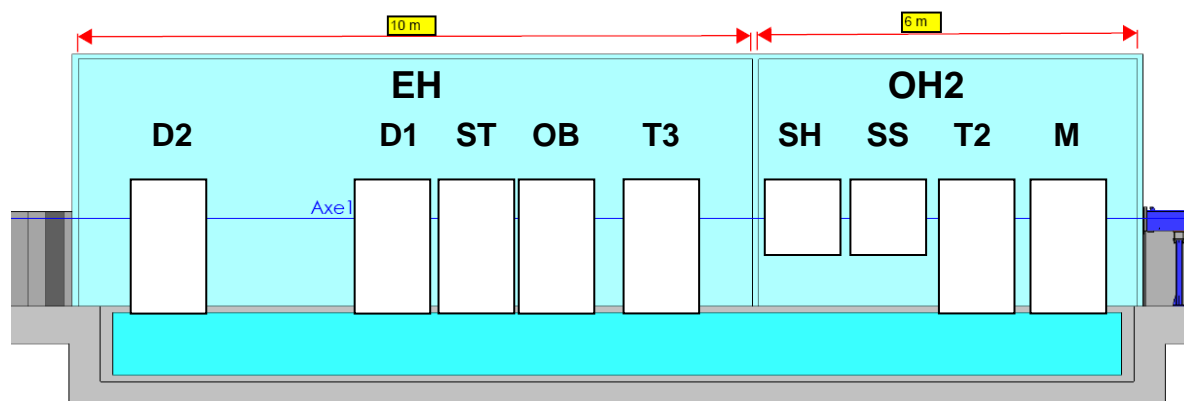


Figure 6. Optics hutch OH2 and experimental hutch EH seen from the left hand side: M is the monochromator tank for the fix-exit Laue and multilayer monochromators. T2 and T3 (T1 is in the OH1) are transfocators for micron and sub-micron focusing. SS and SH are secondary slits and shutter, respectively. The first element in the experimental hutch after T3 is the optics bench (OB) for liquid surface and interface studies and time-resolved reflectometry. ST is the sample table and D1 is the detector tables for reflectivity, diffraction and imaging detectors and D2 is the small angle scattering detector table. All the components could be mounted on the proposed long granite except the secondary slits and the shutter.

are in the new Vercors extension, where the total length of the beamline can be 125 m. Since the nano-focusing beamlines require excellent mechanical, acoustical and thermal stability, the temperature of OH2 and EH have to be controlled with 0.1 K accuracy. Furthermore, since the focal and detector distances at high energies are longer than other nano-focusing beamlines working at lower energies, it is proposed that all the optical elements in OH2 and EH, including sample stages and detectors, are on a 16 m granite slab embedded in the concrete floor.

This proposed granite slab has two purposes: i) to make the foundation as rigid as possible and ii) to isolate optical elements, sample tower and detectors from the sources of the vibration. These vibration sources are often outside of the experimental hutch and even outside of the beamline. A very careful analysis is needed to optimise the granite slab dimensions and to choose the passive isolating mounts and, especially, to find out if this type of isolation is advantageous at all. Due to big size of this slab (65 tons for a size of 16 x 1 x 1.5 m³) and since it is embedded in the floor, the analyses have to be done before the final design/construction of the floor. The cost of the above mentioned granite slab is about 105 k€.

The first optics hutch (OH1) is situated in the current experimental hall and it holds a fixed attenuator, the primary slits and first transfocator. The attenuator cuts all the energies below 50 keV to reduce the heat load on the following optical elements. It can be a radiation cooled rotating 3.5 mm thick SiC disc (~1000°C) like the one used at ID15 or an approximately 2 m long gas absorber which is under development at the ESRF. Before these attenuators a 0.5 mm thick diamond window is needed. The primary slits are special high energy slits developed at ID15 where the vacuum tubes themselves are made of tungsten carbide. The transfocator is similar to the one now installed at ID11 and is able to take white beam radiation.

The layout of the components in OH2 and EH is shown in Figure 6. The optics hutch holds monochromators, two additional transfocators for the adjustable focal

spot size, shutter and secondary slits. The experimental hutch holds the secondary optics for grazing incident studies and for time resolved reflectivity, the sample tower table and the detector tables.

Optics

The monochromators are based on the current design of the fixed-exit bent Laue crystals in the horizontal geometry used at ID15. The desired bandwidth can be easily adapted by the choice of asymmetric cut. Furthermore, in non-dispersive geometry the energy-angle coupling cancels out and the geometric and polychromatic virtual source positions are not differed. This is essential to achieve good focusing in the plane of reflection, i.e. in the horizontal plane. It is not clear if the water cooling of the first crystal is sufficient or if liquid nitrogen cooling is needed. A careful study has to be done, since water cooling with gravitational flow causes much less vibrations than liquid nitrogen cooling. Furthermore, it is not evident how to cool a bent Laue crystal with liquid nitrogen.

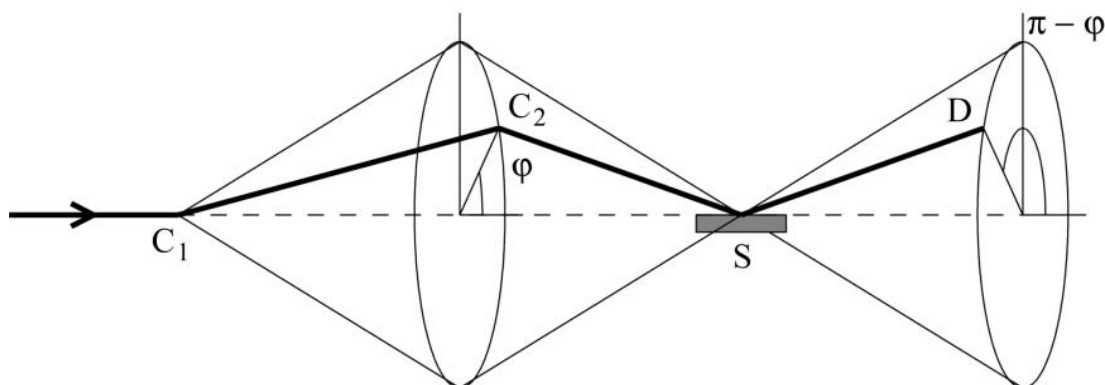


Figure 7. The incident radiation (thick line) is reflected by the first crystal C_1 and subsequently with a larger Bragg angle by the second crystal C_2 with higher order reflection. By rotating the two crystals in a coupled motion around the incident beam (dashed line), the vertical angle of the incident beam on the sample (S) changes. The detector (D) follows the reflected beam from the sample.

An alternative monochromator system in the same vacuum vessel is based on the use of multilayers with 0.3% bandwidth. These two multilayers are also in fixed-exit geometry in the horizontal plane to keep the monochromatic beam parallel to the incident beam with less than 10 mm offset. They are dynamically bent to a parabolic shape so that the beam is parallel in the horizontal plane between them. In the near future it might be possible to accurately machine the parabolic shape such that no dynamical bending will be needed.

The secondary optics for liquid surface and interface studies is based on the design at ID15 (Honkimäki et al, 2006). This optics device is ideally suited for experiments where the sample has to be kept stationary during measurement (Figure 7). The same optical device without the second crystal can be used for the time resolved reflectivity studies. Matsushita et al (2008) proposed time resolved reflectometry in multiwavelength-dispersive mode. They use a bent Laue crystal to focus the incident beam onto the sample and the reflected rays are detected using a one-dimensional detector. For UPBL2 we propose a similar kind of setup, where a second Laue crystal is added as an analyser (mounted on the detector table after

the sample) to reduce the background signal (see Figure 8). Furthermore, instead of using a very wide energy band by strong bending, the variation of the momentum transfer is mostly based on the grazing angle changes due to the fan of the rays. The mean grazing angle is selected by rotating the crystals around the incident beam. In this way the reflectometry can be also used for the studies of the liquid surfaces and liquid-liquid interfaces.

Sample tower

The heart of the setup is the sample tower for manipulation of the sample position and angles. The design of this equipment needs careful study since it has to handle relatively heavy and bulky sample environment systems with the accuracy of 10 nm. Furthermore, it has to be suited for grazing incident studies and it must offer sufficient room around the sample for auxiliary equipments.

Detectors

The first detector table holds:

- A scanning point detector with associated translations and rotations for reflectivity measurements with and without the secondary optics show in Figure 7.
- A 1D-detector which replaces the point detector in time-resolved reflectivity operation. At the moment there are no efficient 1D-detectors for high energies. Ge and CdZnTe strip detectors are most promising candidates. This detector can be replaced by imaging detector or high resolution area detector if the efficiency is good enough.
- A 2D-detector for diffraction studies with longitudinal translation to adjust sample detector distance and thus choose the maximum momentum transfer. The best “high energy” 2D detector currently available is probably the Mar555. For high time resolution (milliseconds or better) an old fashioned image intensifier coupled with a low noise CMOS is probably still the best solution.

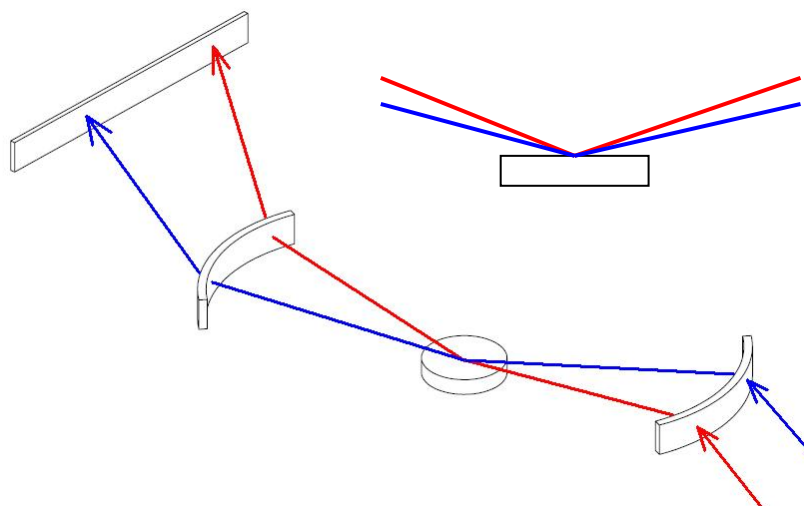


Figure 8. Time resolved reflectometry, where an X-ray beam is equatorially focused onto the sample by a bent Laue crystal. The axial focusing can be made using 1D refractive lenses. The second identically bent Laue crystal after the sample is used as an analyzer to reduce the background signal. The rays corresponding to different vertical momentum transfers are detected by a position sensitive detector. The effect of the energy gradient, in the equatorial plane, to the momentum transfer can be amplified by tilting the crystals. By this tilting the grazing angle increases as a function of energy (inset).

- A 2D-imaging detector and light optics with adjustable magnification. Longitudinal translation is needed to choose between absorption and phase contrast modes. The best time resolution can currently be achieved with CMOS detectors (up to 10^6 frames/s). In the future, low noise CMOS detectors will replace CCD cameras even at moderate frame rates.

The switch from one configuration to another is done by fast sideways translation. The second detector table holds the SAXS detector. This detector can be a CCD or a CMOS camera coupled with taper optics to CsI (structured) scintillator.

Developments of high efficiency and high speed high energy X-ray detectors are essential for ultra-fast imaging and time resolved diffraction. The efficiency of 2D diffraction detectors is very low at 100 keV, <10%, and the situation with imaging detectors is even worse, often <1%.

Hard X-ray imaging detectors

Pushing the spatial resolution of imaging methods towards the nano-scale is a prime objective for the ESRF Upgrade Programme. Direct imaging using a parallel beam provide very comprehensive 3D maps sufficiently fast to study dynamics by tomography, but is currently hampered by the fact that the type of high resolution detectors currently used (Koch et al, 1998) become extremely inefficient in the 30 to 100 keV range when one pushes the spatial resolution below a few micrometres. To overcome this obstacle, detector research and development is needed.

ESRF, Risø/Denmark and IMIT-KTH/Sweden are currently collaborating on a project to replace the homogeneous scintillators by structured scintillators (Olsen et al, 2008; Olsen, 2009) for the resolution in the range of 0.5 to 2 micrometres. The fabrication is initiated by etching a regular 2D array of deep pores into Si. After the pores are oxidised, generating a thin layer of SiO₂, the pores are filled by liquid deposition with the dense fluorescent material CsI:Tl. Finally, the free surface is coated by a thin polymer to make the surface optically flat, to isolate the Tl, and to protect the mildly hygroscopic CsI from water.

The structured scintillator works as a waveguide for the optical photons produced by the X-ray absorption event. The spatial resolution is therefore given simply by the distance between pore centres. Provided pores with large aspect ratios can be made, uniquely this enables one to have high efficiency of the scintillator (deep pores) and, at the same time, a high spatial resolution. For resolutions in the range of 1 to 5 micrometres, the efficiency gain factor can be ~10 with respect to the performance of traditional homogeneous scintillators. At the time of writing, promising results have been obtained with test-specimens having a pore distance of 1.4 micrometres. The first experimental use at ESRF is planned for 2009.

To improve the imaging resolution beyond 500 nm seems extremely difficult due to the range of the high energy electrons in the scintillator. Therefore, to reach higher resolution the images have to be magnified by the X-ray beam itself. As a result of the normal approach using bent crystals, KB-optics etc., the spatial resolution is achieved by a price of temporal resolution (by improving factor two the spatial resolution in tomography, the temporal resolution goes down by an order of magnitude). An interesting possibility is to focus the white beam with refractive compound lenses and then put a pin-hole at the distance corresponding to the

desired energy. This pin hole will now serve as a secondary source and also as a wide band monochromator. The width of the energy band depends on the size of this pinhole and the demagnification in the focusing.

Risø National Laboratory has proposed the construction of a so-called 3D nano-detector, possibly to be financed partly by the ESRF Upgrade Programme. The basic concept of the proposed detector is a 2D strip electrode design. Simulations show that, in the absence of noise, the spatial resolution will be in the range of 30 to 100 nm for 36 keV photons. With advanced fast electronics readout the aim is to be able to record up to 10^9 cps. A 3D detector with ten individual sensors placed close to each other opens up possibilities beyond that of the presently-used compound detectors. First of all it becomes possible to combine high spatial resolution and high efficiency. Secondly, new opportunities emerge for subsequent reconstruction, both for super-resolution algorithms and for ray-tracing in the case of a divergent beam.

Network and computing

The data flows at UPBL2 are amongst the highest at the ESRF. The fast imaging can produce in near future (CMOS is available at ID15 at the end of 2009) 12-bit 1k x 1k images with the unprecedented speed of 10^4 frames/s. With this speed, 15 GB/s, the data is saved on the internal memory of the CMOS camera. After filling the internal memory the data have to be written on the data storage as fast as possible to keep dead time as low as possible. The diffraction and SAXS detectors produce data with much slower flow, but data storage capacity remains very demanding: UPBL2 can produce several TB per day. To reduce the amount of the data, intelligent triggering mechanisms have to be used such that only successful data sequences are stored. Furthermore, the online analysis (at least partial) of diffraction and imaging data is essential to visualise in real time when the data is useful or useless. For example the Large Hadron Collider in CERN can produce 300 GB/s (only 20 times more than peak rate of UPBL2) but only 700 MB/s, 5% of the peak rate of UPBL2, is saved due to intelligent screening of successful data sequences. Even this results in 15 PB/year (about 100,000 DVDs).

The data processing speed is another bottleneck. While the collection of the full low resolution tomographic data set takes less than a second, the 3D image reconstruction takes about 3 minutes. To collect high resolution data set takes few seconds and the reconstruction time is already about 20 minutes.

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