

UPBL6: Inelastic hard X-ray scattering for electronic spectroscopy	
Current designated sector:	Facility goes to:
ID16	ID20

1.1 SUMMARY

The UPBL6 project aims at upgrading the inelastic X-ray scattering (IXS) programme currently carried out at beamline ID16. The scientific case of the beamline rests on the capability of providing a spectroscopic tool with the specific advantages of a hard X-ray probe: bulk information, high-penetrating power, elemental and spin sensitivity. The new beamline will feature two stations dedicated to electronic and vibrational excitations, respectively. The former station is new, whilst the latter one is largely based on the one today installed at ID16. The main idea of the present proposal is to strengthen the advantages of IXS over complementary techniques providing:

- an intense, stable beam focused down to few microns leaving at the same time enough free space around the sample for complex sample environments;
- two spectrometers for studies of electronic excitations optimised for high efficiency with medium energy resolution (~ 300 meV) and for high energy resolution applications (< 100 meV), respectively;
- an optical scheme to extend the energy range of applicability of the technique for studies of electronic excitations from the 6-12 keV interval used today up to ~ 20 keV; iv) an optimised setup for studies of vibrational excitations to reach sub-meV energy resolutions.

1.2 PROJECT HISTORY

This project originates from the INELX CDR presented in the Purple Book. The main modification to the previous CDR is the fact that stations 2 and 3 of the previous CDR are now merged in a single station dedicated to IXS experiments to study electronic excitations exploiting a beam few microns in size. The request for a long beamline (station 3 in the purple book) remains valid in the present version of the CDR.

The part of the UPBL6 scientific case concerning studies of electronic excitations has been further refined at a dedicated brainstorming meeting (12-13 January 2009).

1.3 SCIENTIFIC CASE

Inelastic X-ray scattering (IXS) is one of the techniques that most largely benefitted from the advent of third generation synchrotron sources and from the availability of very brilliant X-ray beams. Among other applications, it is nowadays used as a spectroscopic tool to investigate vibrational and electronic excitations in materials science, chemistry and physics on both fundamental and application-driven grounds; in this context, it is useful to separate non-resonant and resonant studies.

Non-resonant IXS is characterised by a simple cross-section that gives direct access to the dynamic structure factor (Schülke, 2007). It is a technique complementary to inelastic neutron scattering, electron energy loss spectroscopy and soft X-ray absorption. For simplicity reasons, the discussion can be divided into scattering from the cloud of core electrons bound to nuclei, weakly bound electrons and single core electrons (X-ray Raman spectroscopy).

Very-high energy resolution (meV) IXS is scattering from the cloud of core electrons bound to the nuclei: it probes vibrational excitations while the electronic degrees of freedom remain in the ground state (Schülke, 2007). Formally, the cross-section can be written as the space and time Fourier transform of the density-density correlation function, as in coherent inelastic neutron scattering (INS) experiments. The interest in this technique lies in the fact that it has some specific advantages over INS that make it a unique probe for some classes of problems. The most relevant ones are:

- kinematic limitations in the neutron-nucleus scattering process make impossible for INS to probe acoustic excitations in disordered systems when the sound velocity is high enough (~ 2000 m/s); these limitations are absent in the IXS process.
- X-ray beams, contrary to neutron ones, can easily be focused down to linear sizes of ~ 10 μm and even less. This allows experiments to be performed on samples available only in very small quantities.

This latter advantage has led to a large body of experiments aimed at studying phonon branches in single crystals difficult to grow to large sizes, e.g. novel superconductors (Shukla et al, 2003), or in samples subjected to extreme conditions of pressure and/or temperature (Fiquet et al, 2001). The former advantage has instead motivated experiments mainly on liquids and glasses aimed at studying acoustic excitations with wavelengths of few nanometres (Sette et al, 1998). In fact, the properties of these excitations - at odds with what happens in simple crystals - are very different from those of larger wavelength acoustic excitations studied e.g. with light scattering techniques (Rufflé et al, 2006; Monaco & Giordano, 2009). Their study is therefore fundamental to understand the universal trends found in the thermal properties of disordered systems at low temperature, as specific heat and thermal conductivity. Moreover, acoustic excitations with wavelengths comprised between few hundreds of nanometres, as those probed with light scattering, and few nanometres, probed using IXS, are for the time being not accessible at all. Current trends aim to push visible and ultraviolet light scattering techniques to access lower wavelength excitations and IXS techniques to access larger wavelength ones in order to bridge this gap.

The study of weakly bound electron excitations using IXS is mainly driven by questions of fundamental physics. In the past, these investigations have focused on valence electron excitations in metals and semiconductors, with the aim of obtaining a deeper understanding of band-structure and electron correlations effects (Schülke, 2007). Today, the IXS studies of valence excitations are more and more sophisticated following the continuous developments of the experimental setups, e.g. double-plasmon excitations have been discovered and characterised in simple metals (Sternemann et al, 2006), and non-trivial local-field effects have been found in MgB_2 (Cai et al, 2006) and in graphite (Hambach et al, 2008). One special class of these experiments, based on the use of a standing wave technique (coherent IXS), allows obtaining information on the off-diagonal dynamic structure factor (Schülke, 2007), a fundamental quantity not otherwise experimentally accessible that is of special interest for theoretical developments of *ab initio* codes. The knowledge of the off-diagonal dynamic structure factor is as well important to correctly invert the experimental spectra and to overcome the phase problem of this inversion. In this way the full two-particle electron density correlation function in space and time can be obtained, thus providing the most direct access to electron correlation in actual materials. First interesting applications of an inversion scheme, still without using the off-diagonal response, have recently been reported (Abbamonte et al, 2007). On a more general ground, the characterisation of neutral excitations provided by IXS is a key ingredient that, coupled with the information obtained by more standard spectroscopies like photoemission and X-ray absorption, can be exploited to obtain a detailed picture of the electronic structure. For example, soft X-ray absorption is a powerful technique to determine the ground-state structural properties, but is weak to determine the energetics of low lying excited states since i) it is limited by the lifetime of the core hole state and ii) the energetics are modified by the presence of the core hole. These low energy excitations can be measured in IXS experiments provided that enough intensity and resolution is available, as recently demonstrated in the case of *d-d* excitations in NiO and CoO (Larson et al, 2007; Haverkort et al, 2007).

X-ray Raman spectroscopy (XRS) is a rapidly developing, powerful technique in chemistry, physics, and materials science. It can be used as a complementary tool to soft-X-ray absorption to study the K-edges of low-Z materials including both the XANES (Tse et al, 2008) and the EXAFS regions (Bergmann et al, 2007); the K- and the L-edges of intermediate-Z materials (Sternemann et al, 2008); the M-edges of transition metals and the N-edges of rare earths (Gordon et al, 2008). XRS has been demonstrated to be particularly useful to provide structural information in disordered systems to complement that obtained from diffraction experiments. In fact it has already been successfully used on a number of systems, including pure liquids and solutions, ices/hydrates, silicon/clathrates, glasses, nanotubes/fullerenes (Schülke, 2007). More specifically, XRS provides two specific advantages.

- Being a hard X-ray technique, it can be used under circumstances where soft X-rays are unusable, e.g. high pressure studies (Mao et al, 2003). This is of particular interest for geophysical investigations, as for example recently demonstrated in the study of the local electronic structure of the MgSiO_3 glass under pressure (Lee et al, 2008).
- It is not limited to the dipole approximation, and the weight of non-dipolar scattering channels can be chosen by varying the momentum transfer. In particular, this has opened the possibility to measure angular-

momentum-projected DOS (Krisch et al, 1997). This opportunity is becoming more and more attractive since the widely spread FEFF code based on a real-space multiple-scattering approach with core-hole effects included has been extended to the q-dependent problem posed by XRS (Soininen et al, 2005). The combination of the experimental and calculated angular-momentum-projected DOS can indeed provide additional relevant information on the electronic structure of complex materials, as e.g. recently demonstrated for doped MgB₂ superconductors (Mattila et al, 2005).

Resonant IXS in the hard X-ray range is particularly attractive because it allows access to the K-edges of 3d transition metals and L-edges of rare-earths. It is characterised by a complex cross-section that has often made it difficult to perform quantitative studies. At the same time, however, the resonance enhancement of the cross-section makes the experiments easier than the corresponding non-resonant ones, and its richness offers unique possibilities that are attracting the interest of a large community worldwide (Schülke, 2007). In fact, RIXS is a powerful tool to study electronic excitations in solids giving access to site, element, and orbital selective information (Kotani & Shin, 2001), e.g. as a quantitative probe of the electronic structure of strongly correlated electron systems like heavy fermion compounds (Dallera et al, 2002). Especially, the coherence between the excitation and de-excitation process in resonant IXS leads to interference effects in the scattered radiation (fluorescence interferometry (Ma, 1994)) that can, for example, be used to assign excitations of certain symmetry to atoms on inequivalent lattice sites.

One scientific area in strong development is the study of low-energy electronic excitations in transition-metals based compounds (Hill et al, 2008). In this respect, resonant IXS in the hard X-ray range is complementary to resonant IXS at the corresponding L-edges and offers some interesting opportunities. Firstly, it has different selection rules than in the soft X-ray range – in practice, this allows probing different aspects of the physics of a given system, e.g. it is more sensitive to the charge transfer excitations (Collart et al, 2007) than to *d-d* transitions that dominate the spectra in the soft X-ray range (Ghiringhelli et al, 2002). Secondly, being a hard X-ray technique it is not surface sensitive, and in particular allows for experiments in complex sample environments, e.g. high pressure cells or interfaces in heterostructures.

The UPBL6 project aims to strengthen the main advantages of IXS with respect to complementary techniques.

One of the most important aspects of this project is to develop a focusing scheme to obtain a spot size of few microns, ideally 5 x 5 μm² V x H FWHM. The task is demanding since this beam size has to be reached without any relevant loss in intensity (IXS is a very photon-hungry technique), and the focused beam has to be extremely stable (typical IXS scans last several hours).

A beam size of a few microns opens scientific opportunities that are now out of reach. This is in particular the case for high-pressure studies, e.g. investigating pressure induced bonding modifications of carbon-based systems like intercalated fullerenes, or following the electronic structure modifications that are expected to accompany liquid-liquid transformations as in the case of Na at high-pressure

(Gregoryanz et al, 2005). The beam size is clearly even more an issue for high-pressure high-temperature studies using laser heating in DAC cells, like for instance the study of liquid iron at conditions of interest for geophysical investigations (Shen et al, 2004). It is as well interesting to note that coupling an X-ray beam of this size with a high-pressure cell designed to have a stress profile varying continuously in the sample, a complete study as a function of pressure could be realised using only one sample, as recently demonstrated in a visible Raman study of carbon nanotubes (Del Corro et al, 2008). Actually, the considered beam size would allow a simultaneous X-ray and visible-light Raman study, thus exploiting the strong complementarities between X-ray and visible-light Raman scattering for carbon-based systems. Moreover, an X-ray beam of the considered size would allow studies on small samples (tiny single crystals, wires, inclusions etc.), as well as spectroscopic mapping of samples inhomogeneous at the scale of a few microns.

An issue generated by extremely intense and focused X-ray beams is radiation damage. For this reason the option to use an unfocused beam should be available, e.g. for biological samples and soft matter (Galambosi et al, 2006) as well as to obtain proper averages in powder samples. However, it is interesting to underline that radiation damage can also be an opportunity: new forms of materials can be produced, as in the recently reported radiation/pressure induced dissociation in ices (Mao et al, 2006), or in metal-insulator transitions induced in strongly correlated electron systems (Kiryukhin et al, 1997). This opens the possibility of accessing unusual chemical processes, and calls for time resolved studies to follow them in situ.

Another opportunity opened by a small X-ray beam concerns tomography. It has already been demonstrated that tomographic imaging of samples in diamond anvil cells can be exploited as a route to obtain the sample density (Liu et al, 2008), that is often a critical parameter to obtain in high-pressure studies of disordered systems. More in general, and exploiting the 1:1 Rowland circle geometry for the IXS spectrometers foreseen in the UPBL6 project, tomographic imaging of samples can be routinely obtained simultaneously with the IXS measurements. With an incoming beam few microns in size, and improving the focusing capabilities of the spherical crystal analysers used in the IXS spectrometers, a combined 3D imaging and spectroscopic information can be then obtained on the scale of a few microns. This allows the unique possibility of imaging electronic excitations: in fact in addition to the three-dimensional spatial resolution, the use of IXS adds resolution in exchanged momentum and energy. There are no other imaging methods that could map, for example, collective valence-electron dynamics or soft X-ray absorption edges of samples buried in deep bulk.

A second very important aspect of the UPBL6 project is to push non-resonant IXS experiments towards energies higher than those where they are currently performed (6–12 keV). The present limitations are imposed by technical reasons (mainly energy resolution), and can be overcome by the development of crystal-analysers made out of Ge, thus allowing the technique to be extended to the ~20 keV energy range. These energies will open a number of very interesting opportunities:

- The access to higher-Z samples is facilitated (the absorption to scattering length ratio is more favourable), e.g. XANES studies at the Fe $M_{II,III}$ edge to study spin transition or bond hybridisation would become realistic.

- The access to buried samples/interfaces and to samples in demanding sample environments would become easier thus rendering possible experiments that are out of reach today, e.g. EXAFS-like experiments in a diamond anvil cell (DAC) to study for instance the modifications of the local structure in liquid water at high-pressure conditions, or samples pressurised using the Paris-Edinburgh press, or even samples in an environment prepared for investigations of chemical reactions in situ or catalysis. Of special interest for the high-pressure community, high-pressure experiments using DAC in standard transmission geometry through the diamonds would become feasible at these energies, and the limitations imposed by the use of panoramic DAC cells in transmission through a Be gasket would be overcome.
- Higher momentum transfers would get within reach, leading to an easier and more effective access to non-dipolar excitations at high- q , e.g. atomic-like $d-d$ transitions (Larsson et al, 2007), and to angular-momentum-projected density of states (Soininen et al, 2005).
- The Compton scattering spectrum is moved to higher energies, which can be in particular an advantage in terms of background for XRS experiments.
- The impulse approximation for the Compton scattering process becomes easier to reach and it is then possible to study the Fermi-surface properties of bulk samples via high-resolution Compton spectroscopy (Huotari et al, 2007).
- Radiation damage in sensitive samples is reduced.

A third very important aspect of the UPBL6 project concerns the increase of intensity provided by the increased performance of the ESRF source and the increased efficiency of collection of the scattered X-rays provided by the optimised designs of the spectrometers dedicated to both non-resonant and resonant IXS to study electronic excitations. In this respect, it has to be underlined that whilst only a limited increase in brilliance of the source can be expected in the near future (factor three to five, see Figure 1), most of the gain in efficiency for the IXS technique has to come from an improved collection of the scattered radiation over the solid angle, i.e. it has to come from the optimisation of the spectrometers. This increase in overall efficiency would allow better statistics to be obtained in experiments on high-Z materials where the count rates are very low and, more importantly, would open the way to experiments that today are not feasible or at the edge of being feasible, e.g. on gases (Minzer et al, 2008). Moreover, studies demanding higher energy resolution – today out of reach due to low intensity - will become feasible, and this is particularly interesting in connection to the study of low-energy collective excitations in layered and/or strongly correlated systems (Bozovic, 1990) and neutral excitations probed with both non-resonant and resonant IXS in correlated electron systems ($d-d$ excitations, $f-f$ excitations, 2-magnons) (Hill et al, 2008). Higher energy resolution could make it possible as well to measure the lifetimes of these excitations, possibly looking for electron-phonon coupling or more in general for coupling with other excitations. Clearly, higher energy resolutions will be very interesting also for studies in gases to resolve the sharp atomic lines.

It is clear as well that the sensible increase of performance of the source foreseen in the present project at energies of ~ 25 keV will have a direct impact also on the very-high energy resolution experiments to study the acoustic excitations in

disordered systems using the horizontal spectrometer arm installed at the moment at ID16 and to be moved to the new beamline. In fact, resolutions slightly below 1 meV are technically at hand already today, e.g. employing the Si(13,13,13) reflection for both main monochromator and diced analyser crystals. However, this setup is less luminous by a factor of eight with respect to that based on the Si(11,11,11) reflection routinely used today for experiments. This lower luminosity basically discourages for the time being its use for such delicate experiments. It is clear however that, with a brighter source, this setup can be successfully operated. Moreover, an incremental upgrade of the existing setup, mainly based on the construction of new, more performing diced crystal analysers, could open energy resolutions as low as 0.7 meV at the Si(13,13,13) reflection. It is however important to underline that every, even minor, gain in energy resolution with respect to the 1.4 meV limit reached today will pave the way to new physics being discovered since we know that the range of dynamical properties probed by IXS today in disordered systems lies exactly at the edge where major changes take place between the macroscopic limit and the mesoscopic regime (Monaco & Giordano, 2009).

1.4 TECHNICAL CONSIDERATIONS

The technical solutions presented here are aimed to design a beamline that provides the best possible beam characteristics in terms of spot size, stability, flux, and flexibility in choosing the incoming beam bandwidth in the energy range 6–26 keV. The beamline will feature two stations. One station is dedicated to studies of vibrational excitations and corresponds to that currently setup at ID16. The existing station will be basically moved to the new beamline location, with a minimum of changes in order to keep it compatible with the other, new station of the beamline that will be optimised for studies of electronic excitations. This new station will feature two independent spectrometers optimised for high efficiency with medium energy resolution (~300 meV) and for high energy resolution applications (<100 meV), respectively. The corresponding experimental hutch will be equipped as well with angle-resolved X-ray diffraction and energy resolved fluorescence detection, in situ laser heating and laser + spectrometer for pressure measurement, as well as a Raman spectrometer for combined X-ray and visible Raman measurements.

In the "Scenario of "All Beamlines" Portfolio at the end of the ESRF Upgrade Programme "Phase I Minimum" endorsed by the Autumn 2008 SAC meeting, UPBL6 was positioned at ID16. More recent discussions suggest a scenario where the UPBL6 project shall be realised at ID20. The following considerations are based on this second scenario.

The beamline should exploit an undulator source optimised to deliver the highest possible flux over the energy range 6-26 keV. We expect an average three- to five-fold increase in flux with respect to the present values, due to the combined effect of the undulator upgrade, the increase in ring current and an increase in straight section length. The expected photon flux provided by a 6 m long straight section equipped with 2 x 2.5 m U20.5 in-vacuum NdFeB cryogenic undulators with 200 mA in the storage ring is shown in Figure 1.

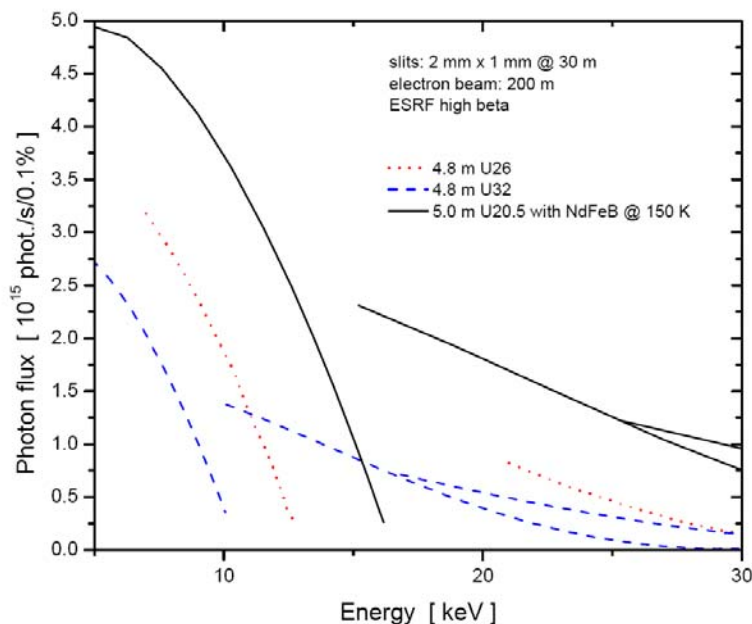


Figure 1. Comparison of current and new undulator performances for the upgraded beamline.

The conceptual layout for the UPBL6 project at the ID20 location is shown in Figure 2. The beamline includes five hutches, four of which correspond to those of current ID16 whilst the fifth will host the new station at 160 m from the source.

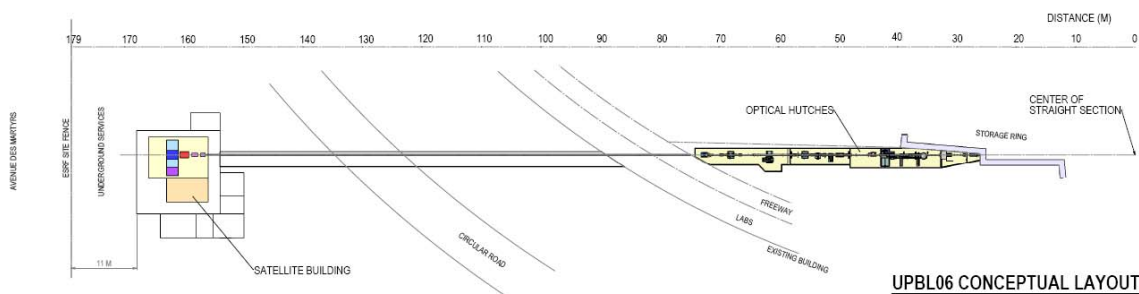


Figure 2. UPBL6 conceptual layout.

Figure 3 shows the optical layout of the beamline in the configuration for studies of vibrational excitations. This station is today installed at ID16 and will be moved to the new beamline location. The main components of this station are: backscattering monochromator, focusing mirrors, (horizontal) spectrometer arm. A few upgrades are foreseen on some vacuum chambers to make them compatible with the new beamline design, e.g. the main monochromator chamber; on the diced crystal analysers with the aim of pushing the energy resolution well below 1 meV; on some electronic components, e.g. temperature readout of the analysers; and on the motor control (migration from DPAP to ICEPAP).

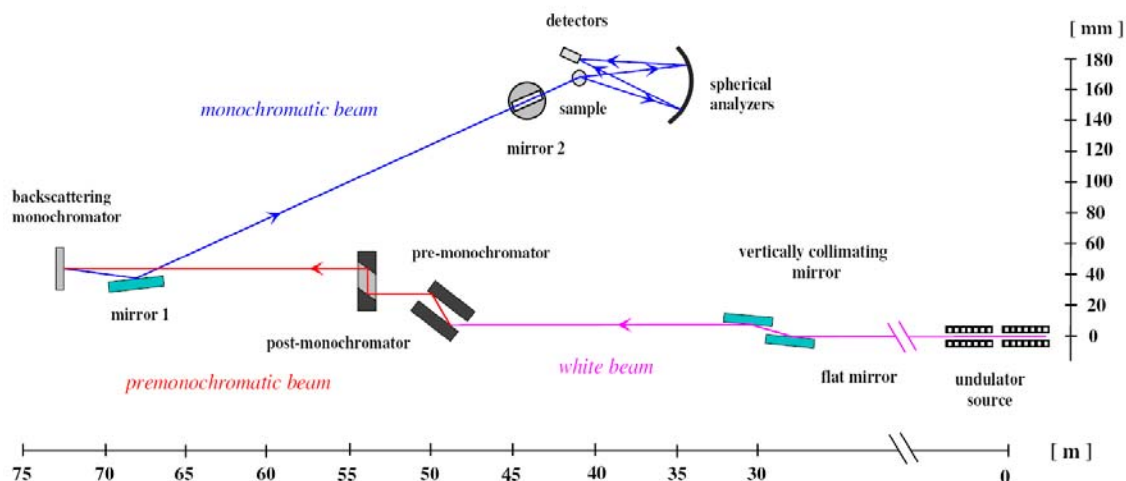


Figure 3. Optical layout of the upgraded beamline in the configuration for studies of vibrational excitations.

Figure 4 shows the beamline optical layout up to the sample position in the configuration for studies of electronic excitations. This is a new station. It is based on the idea of building a separate experimental hutch at ~160 m from the undulator source. The challenge here is to provide an intense, focused and stable beam whilst leaving enough space for complex sample environment and for the spectrometer around the sample position. The X-ray beam spot size should be not excessively larger than $\sim 5 \times 5 \mu\text{m}^2$ to provide a competitive setup for high pressure studies. This should of course be realised in minimising the intensity losses since IXS is a photon-hungry technique and many experiments are at the edge of being feasible. Moreover, the sample environment and the spectrometers require a minimum of ~ 1 m free space that sets an important constraint to the closest position where the focusing mirror(s) can be placed. Choosing, for overall efficiency reasons, a solution where the focusing takes place in one step (no intermediate source), then the need of a high-beta section and a long beamline becomes clear. In fact, given that the horizontal source size is $950 \mu\text{m}$ FWHM, a strong demagnification of more than 100:1 is required in order to reach the goal above. The requirements in the vertical direction are more relaxed.

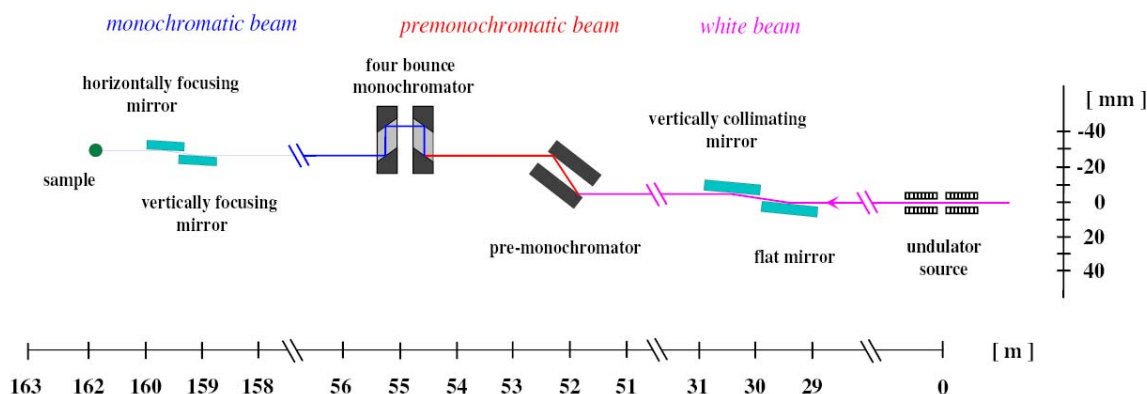


Figure 4. Optical layout of the upgraded beamline up to the sample position in the configuration for studies of electronic excitations.

More specifically, a ray-tracing study of the optics suggests the use of a vertically collimating mirror at about 30 m from the source in order i) to keep the focusing optics as short as possible and ii) to maximise the throughput of the high-resolution monochromator stages. As for the focusing optics, a standard KB scheme is proposed. The idea is to use a 500 mm long mirror in total reflection for the vertical focusing, with $p=160.25$, $q=1.75$ and a grazing angle of 2.7 mrad, aiming at a vertical spot size of $\sim 5 \mu\text{m}$ or better. For the horizontal focusing, the use of a depth-graded multilayer is proposed, with $p=160.75$, $q=1.25$ and a diffraction angle of $\sim 1.5^\circ$, aiming at a horizontal spot size of $\sim 7 \mu\text{m}$. These multilayers can be tailored to have a bandwidth of up to 20% (Morawe et al, 2002) with reflectivities up to 60%, and therefore are compatible with the scanning energy constraints of the UPBL6 project. Several multilayer stripes should be made available in order to cover the entire energy range of interest here. In addition to these focusing elements in KB geometry and in order to cope with samples that suffer radiation damage, the possibility of using a defocused X-ray beam should be provided as well. We propose to achieve that through an additional toroidal mirror mounted close to the vertically focusing one. The goal in this case will be to obtain a focal spot of $\sim 50 \times 500 \mu\text{m}^2$ (V x H).

It is clear that the proposed optical layout requires vibrations to be kept to a minimum in order not to spoil the focusing results.

The flexibility in the choice of the incoming beam bandwidth will be provided through a narrow-band-pass post-monochromator stage which follows the pre-monochromator. This could be based on a four-bounce design as a flexible but fixed-exit device. This post-monochromator should be equipped with different sets of crystals for the different energy ranges of interest and the different bandwidths required. For example, some reference numbers for a setup based on Si(4 4 0) reflections are reported in Table 1. The flux on the sample has been calculated using the source parameters given above and estimating an overall optics transmission of 10%.

Besides the focusing optics, the new hutch at ~ 160 m would host two spectrometers (optimised for high efficiency with medium energy resolution and for high energy resolution applications, respectively). The two spectrometers could be mounted on rails (or on air pads) and could be translated in and out of the beam, and would be operated alternately (see Figure 5).

Energy [keV]	10	20
Bandwidth [meV]	95	190
Flux on sample [10^{12} ph/s]	5.2	2.6

Table 1. Expected beamline properties at two characteristic energies using a Si(4 4 0) four-bounce monochromator.

The first spectrometer (Figure 5 left) is dedicated to non-resonant IXS experiments for studies of electronic excitations. The aim here is to collect the scattered radiation over a solid angle as large as possible with an energy resolution of ~ 300 meV, a requirement dictated in particular by X-ray Raman measurements of

XANES spectra of low-Z elements. The solution proposed is based on building two arrays of Si(1 1 0) spherical analyser crystals covering the full 0-180 degrees scattering angle range in vertical geometry. The analysers, 48 in total, will be mounted in Rowland geometry with a 1 m bending radius, and will focus the scattered radiation onto a 2D photon counting pixel detector system. This solution corresponds to a sizeable increase in the coverage of the solid angle for the scattered radiation with respect to the current situation and, in addition to the expected increase in flux from the source, leads to more than an order-of-magnitude increase in performance with respect to the situation today. This would clearly open the way to experiments that are currently at the limit of feasibility or not feasible at all, like for instance the study of *d-d* excitations in correlated electron systems, or X-ray Raman experiments (including EXAFS-like measurements) on systems under pressure.

The second spectrometer installed in the experimental hutch will be dedicated to non-resonant and resonant IXS experiments to study electronic excitations that require both high energy resolution and highly focused beam and will consist of a 2 m long arm capable of rotating both in the horizontal and in the vertical plane. This is particularly required since the resonant IXS spectra depend both on the incoming and the scattered beam polarisation. This setup will as well include a phase-plate in order to give the flexibility of choosing the polarisation of the incoming beam. The requirement of scanning over the scattered energy imposes to renounce to a full coverage of the solid angle, as for the first spectrometer. The spectrometer will be equipped with multiple (possibly seven) spherical crystal analysers in 1:1 Rowland geometry with a bending radius in the range 1-2 m, and a 2D photon counting detector. The arm should allow scans over analyser Bragg angles in the range 70-89 degrees. In addition, there should be the possibility to easily change the analyser crystals both to operate the spectrometer at several absorption edges for resonant IXS experiments, and to flexibly choose the spectrometer contribution to the energy resolution that depends on the kind of analysers that are used (elastically bent, patched, or diced), on the reflection order and on the material. For both non-resonant and resonant IXS measurements energy resolutions down to 50 meV will be possible.

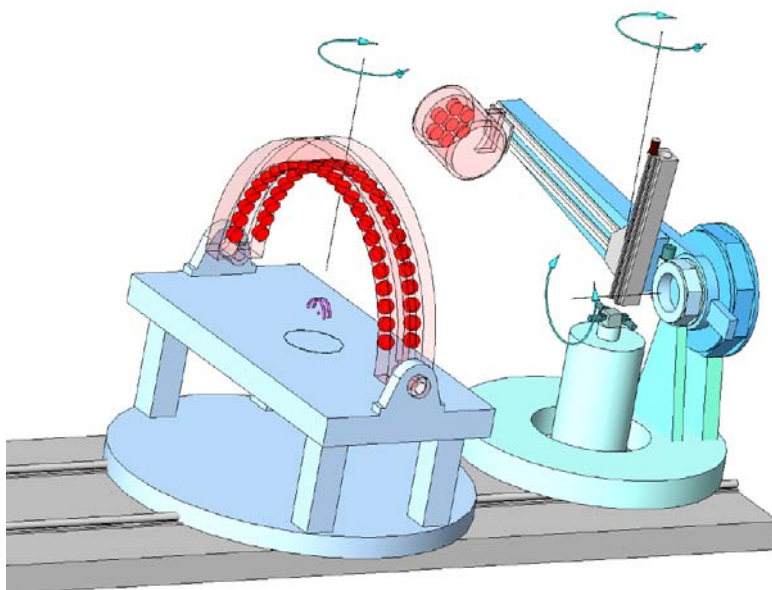


Figure 5. Sketch of the two spectrometers of the upgraded beamline.

As shown in the preceding section, these spectrometers should be capable to operate up to the ~ 20 keV energy range. This requires the development of the related and, up to now not yet existing, X-ray optics. In fact, the bent Si analysers that are in use today provide resolutions of few eV in this energy range. The technical solution proposed here is to use Ge instead than Si, exploiting the smaller absorption length that directly translates into smaller energy resolutions. Bent crystal analysers made out of Ge to provide energy resolutions in the 0.3-0.5 eV range for photon energies between 10-20 keV are being developed at the moment in-house, but this project remains challenging due to the difficulties in manufacturing Ge optics devices (fragility, material availability, purity, costs). Moreover, the 1:1 Rowland geometry that we plan to use for the design of the spectrometers allows a point-to-point focusing of the source onto a position-sensitive detector. This enables imaging capabilities, but requires the optimisation of not only the incident beam size on the sample but of the analyzer focusing capabilities to the same level as well. A second challenge in this respect is therefore to reduce the focal spot size of the bent crystal analysers from the ~ 100 μm achieved today to the ~ 10 μm range to match the X-ray beam size on the sample. Moreover, for resonant IXS applications the development of toroidal rather than spherical analysers would be highly beneficial when analyser Bragg angles smaller than 80 degrees are used. The achievement of this goal would be useful to obtain imaging capabilities for resonant IXS experiments as well thus allowing, for example, discrimination of the signal from a sample buried inside a matrix or imaging of an inhomogeneous sample.

It is important to underline that for these high-energy applications efficient 2D photon counting detectors do not yet exist, though the ESRF, together with collaborating partners, is currently strongly involved in the development of 2D GaAs position sensitive detectors.

Finally, the overall luminosity of these two spectrometers will likely allow non-reversible processes to be probed with the simultaneous acquisition of a full spectrum 0.2-2 eV wide (fixed analyser configuration with diced analysers). On selected scientific problems, time resolutions down to the ms range will be within reach.

1.5 REFERENCES

- Abbamonte P, Graber T, Reed JP, Smadici S, Yeh C-L, Shukla A, Rueff J-P & Ku W (2008) Proc. Nat. Acad. Sci. 105, 12159-12163.
- Bergmann U, Di Cicco A, Wernet P, Principi E, Glatzel P & Nilsson A (2007). J. Chem. Phys. 127, 174504.
- Bozovic I (1990). Phys. Rev. B 42, 1969.
- Cai YQ, Chow PC, Restrepo OD, Takano Y, Togano K, Kito H, Ishii H, Chen CC, Liang KS, Chen CT, Tsuda S, Shin S, Kao CC, Ku W & Eguiluz AG (2006). Phys. Rev. Lett. 97, 176402 (2006)
- Collart E, Shukla A, Rueff J-P, Leininger P, Ishii H, Jarrige I, Cai YQ, Cheong S-W & Dhalenne G (2006). Phys. Rev. Lett. 96, 157004.
- Dallera C, Gironi M, Shukla A, Vankó G, Sarrao JL, Rueff JP & Cox DL (2002). Phys. Rev. Lett. 88, 196403.
- Del Corro E, González J, Taravillo M, Flahaut E & Baonza VG (2008). Nano Lett. 8, 2215-2218.
- Fiquet G, Badro J, Guyot F, Requardt H & Krisch M (2001). Science 291, 468.
- Fister TT, Seidler GT, Wharton L, Battle AR, Ellis TB, Cross JO, Macrander AT, Elam WT, Tyson TA & Qian Q (2006). Rev. Sci. Instr. 77, 063901.

- Ghiringhelli G, Brookes NB, Annese E, Berger H, Dallera C, Gioni M, Perfetti L, Tagliaferri A & Braicovich L (2004). *Phys. Rev. Lett.* 92, 117406.
- Galambosi S, Knaapila M, Soininen JA, Nygard K, Huotari S, Galbrecht F, Scherf U, Monkman AP & Hamalainen K (2006). *Macromolecules* 39, 9261.
- Gordon RA, Seidler GT, Fister TT, Haverkort MW, Sawatzky GA, Tanaka A & Sham TK (2008). *Europhys. Lett.* 81, 26004.
- Gregoryanz E, Degtyareva O, Somayazulu M, Hemley RJ & Mao Hk (2005). *Phys. Rev. Lett.* 94, 185502.
- Hambach R, Giorgetti C, Hiraoka N, Cai YQ, Sottile F, Marinopoulos AG, Bechstedt F & Reining L (2008). *Phys. Rev. Lett.* 101, 266406.
- Haverkort MW, Tanaka A, Tjeng LH & Sawatzky GA (2007). *Phys. Rev. Lett.* 99, 257401.
- Hill JP, Blumberg G, Kim Y-J, Ellis DS, Wakimoto S, Birgeneau RJ, Komiya S, Ando Y, Liang B, Greene RL, Casa D & Gog T (2008). *Phys. Rev. Lett.* 100, 097001.
- Huotari S, Vankó G, Albergamo F, Ponchut C, Graafsma H, Henriquet C, Verbeni R & Monaco G (2005). *J. Synchrotron Rad.* 12, 467-472.
- Huotari S, Albergamo F, Vankó G, Verbeni R & Monaco G (2006). *Rev. Sci. Instr.* 77, 053102.
- Huotari S, Sternemann C, Volmer M, Soininen JA, Monaco G & W. Schülke (2007). *Phys. Rev. B* 76, 235106.
- Kiryukhin V, Casa D, Hill JP, Keimer B, Vigliante A, Tomioka Y & Tokura Y (1997). *Nature* 386, 813.
- Kotani K & Shin S (2001). *Rev. Mod. Phys.* 73, 203-246.
- Krisch MH, Sette F, Masciovecchio C & Verbeni R (1997). *Phys. Rev. Lett.* 78, 2843-2846.
- Larson BC, Ku W, Tischler JZ, Lee C-C, Restrepo OD, Eguiluz AG, Zschack P & Finkelstein KD (2007). *Phys. Rev. Lett.* 99, 026401.
- Lee SK, Lin J-F, Cai YQ, Hiraoka N, Eng PJ, Okuchi T, Mao H-k, Meng Y, Hui MY, Chow P, Shu J, Li B, Fukui H, Lee BH, Kim HN & Yoon C-S (2008). *Proc. Nat. Acad. Sci.* 105 7925-7929.
- Liu H, Wang L, Xiao X, De Carlo F, Feng J, Mao H, & Hemley RJ (2008). *Proc. Nat. Acad. Sci.* 105 13229-13234.
- Ma Y (1994). *Chem. Phys. Lett.* 230, 451-455.
- Mao WL, Mao H, Eng PJ, Trainor TP, Newville M, Kao C, Heinz D, Shu J, Meng Y & Hemley RJ (2003). *Science* 302, 425-427.
- Mao WL, Mao Hk, Meng Y, Eng PJ, Hu MY, Chow P, Cai YQ, Shu J & Hemley RJ (2006). *Science* 314, 636-638.
- Monaco G & Giordano V (2009). *Proc. Natl. Acad. Sci. USA* 106, 3659.
- Morawe C, Ziegler E, Peffen J-C & Kozhevnikov I.V. (2002). *Nucl. Instr. And Meth. A* 493, 189.
- Minzer M, Bradley JA, Musgrave R, Seidler GT & Skilton A (2008). *Rev. Sci. Instr.* 79, 086101.
- Rufflé B, Guimbrètiere G, Courtens E, Vacher R & Monaco G (2006). *Phys. Rev. Lett.* 96, 045502.
- Schülke W (2007). *Electron dynamics by inelastic X-ray scattering* (Oxford University Press, Oxford).
- Shen S, Prakapenka VB, Rivers ML & Sutton SR (2004). *Phys. Rev. Lett.* 92, 185701.
- Shukla A, Calandra M, d'Astuto M, Lazzeri M, Mauri F, Bellin C, Krisch M, Karpinski J, Kazakov SM, Jun J, Daghero D & Parlinski K (2003). *Phys. Rev. Lett.* 90, 095506.
- Soininen JA, Ankudinov AL & Rehr JJ (2005). *Phys. Rev. B* 72, 045136.
- Sternemann C, Huotari S, Vankó G, Volmer M, Monaco G, Gusarov A, Lustfeld H, Sturm K & Schülke W (2006). *Phys. Rev. Lett.* 95, 157401.
- Sternemann H, Sternemann C, Seidler GT, Fister TT, Sakko A & Tolan M (2008). *J. Synchrotron. Rad.* 15, 162-169.
- Tse JS, Shaw DM, Klug DD, Patchkovskii S, Vankó G, Monaco G & Krisch M (2008). *Phys. Rev. Lett.* 100, 095502.