Dynamics and Extreme Conditions Group – Plans and Visions for Upgrade Phase II

Executive Summary

The scientific programs on the group's beamlines address all five "highlight" areas with the strongest emphasis on science at extreme conditions. Within phase I of the Upgrade, UPBL6@ID20 will offer unique opportunities in the study of electronic and magnetic structure and dynamics. ID26 pursues a closely related and complementary research program with a broad range of applications in chemistry, biology, materials science, geology/mineralogy, and environmental sciences. ID26 has started an important refurbishment initiative which shall remain the core activity over the next years. The development of a dedicated station for time-resolved XAS-XES studies in the picosecond- to nanosecond regime, as initially proposed in the ID26 CDR, is currently not considered.

Structural studies at very high-pressures (and low/high temperatures) form an important part of the research performed within the group. Within the scope of Phase I ID09A shall move to ID15B and ID06LVP to ID15A. Together with ID27 this corresponds to a potential capacity increase from 1.5 to 2.8 beamlines in a field where the ESRF is extremely successful, but severely suffers from a lack of available beamtime. The average oversubscription rate for ID09A and ID27 over the last five years was 2.8, with a large number of very good (A) proposals rejected. The Large Volume Press (LVP) project has gone in partial user operation during the second half of 2012, a prerequisite in order not to jeopardise its medium- and long-term future. To further strengthen the research programs and offer further opportunities it is proposed to extend ID27 to 110 m into the Chartreuse building to host an experimental station for micro/nano-imaging and XRD at extremely high pressures as well as a set-up to study laser-shocked matter.

Following the closure of ID22N in March 2011, the capacity for Nuclear Resonance studies at the ESRF was significantly reduced. In order to bring back the offer in beamtime back to at least 70% of a full beamline equivalent, two experimental stations on a canted straight section at ID18 are proposed, in line with the updated CDR Nuclear Resonance, which has been very well received at the last beamline review in November 2009.

The upgrade of ID28 foresees an experimental set-up in EH1 for (thermal) diffuse scattering to be operated in parallel and in close complement to the existing IXS spectrometer, and a small angle IXS spectrometer with sub-meV resolution for the study of liquid and disordered materials with an overall energy resolution of 0.6-0.9 meV.

Beyond the ambitious projects on the individual beamlines it is proposed to develop a platform/partnership for extreme conditions science. The concept is based on the Partnership for Extreme Conditions Science (PECS), but with a wider scope not only focusing on high-pressure, but including as well very low temperatures, static and pulsed high magnetic fields, and laser shock techniques. Instrumentation for transport measurements and other complementary techniques shall be considered as well.

A PECS with such an enlarged scope will provide the fertile ground for cutting-edge science on all beamlines of the DEC group and beyond, most notably for the ESM group, but as well for other ESRF Beamlines and CRGs. Furthermore, the role of the ILL as partner is strengthened, and obvious synergies with institutes on the GIANT campus should be exploited.

In the following the proposed beamline upgrades for ID18, ID26, ID27 and ID28 are presented.

ID18

The Nuclear Resonance project aims at strengthening the capabilities for the study of materials at the micro-scale, at extreme conditions, and of dynamics of disordered and biological systems and glasses. It fully exploits the strengths of the ESRF and the Mössbauer effect and adds new elements: implementation of a Synchrotron Mössbauer Source, access to Mössbauer isotopes with high transition energies, and fast pixel detectors for (nuclear) quasi-elastic and small-angle scattering. The ultimate goal of the proposal is to offer the entire suite of nuclear resonance techniques to study simultaneously and possibly *in-situ* structure, dynamics, magnetic, and electronic properties urgently needed to tackle the new emerging questions out of reach with the existing instruments.

The main pillars of the proposed upgrade are:

- A canted beamline with two independent branches including a permanent installation of a new high temperature – high pressure environment for geoscience and novel materials and of an UHV facility for applications in nano-technology/-science.

- Access to Mössbauer isotopes with high transition energies (~ 100 keV).

- Implementation of a nuclear monochromator for a Synchrotron Mössbauer Source (available in all filling modes).

- Launching a fast detector development programme in order to efficiently cope with the available photon flux and to open new scientific fields, which would become available with fast pixel detectors like (nuclear) quasi-elastic scattering (neV-µeV) and (nuclear) small angle scattering.

Fascinating new opportunities will open up, addressing open questions both in fundamental and applied science:

<u>Disordered systems, glasses, biology</u>

- What is the origin of the soft modes (quasi-elastic scattering) emerging in glasses close to the glass-liquid transition? What is the reason of the documented relation of atomic vibrations in glasses with many-orders-of-magnitude slower atomic relaxation?

<u>Nanotechnology</u>

- For exchange bias systems the understanding of layer-by-layer magnetization is crucial. How does the interaction at the interface depend on growth properties? Is there domain formation?

- How does the phonon DOS in a superconductor correlate with the magnetism in the adjacent ferro-magnet? How does the interface behave? An example would be the system Sn/Fe.

• How do quantum dot arrays order magnetically? Do they favour a domain structure? How do they interact with each other and with an external field? With such systems fundamental questions regarding the dynamics of finite spin systems can be addressed. Particularly interesting is the study of non-equilibrium magnetic states if such systems are subjected to external perturbations like oscillatory or pulsed fields.

Extreme conditions

• Determination of the oxidation state of metals in planetary materials, core-mantle interactions, chemistry (chemical reaction) of materials in the Earth's mantle, and dynamical properties such as sound velocities from which conclusions about the compositional profile within the Earth's interior are drawn.

• Investigation and discovery of novel properties of iron-containing nano-materials at high pressure. Cooperative phenomena in strongly correlated 4f and 5f electron systems that emerge under conditions of ultra-high pressure and very low temperatures.

• Low-temperature and high-pressure properties of novel iron-based superconducting materials.

ID26

Non-resonant and resonant X-ray emission spectroscopy (XES) are a powerful complement to the more traditional X-ray absorption spectroscopy (XAS) with applications in chemistry, biology, materials science, geology/mineralogy, and environmental sciences. Though ID26 is the very successful (average oversubscription rate of the last five years: 3.3; accumulated impact factor in 2010: 142), the beamline needs to undergo a substantial refurbishment program in order to stay competitive in the long-term. This comprises:

- Development of crystal analysers for the X-ray emission spectrometer in order to maximise the solid angle (number of analysers/reflection, bending radius) and the analyser quality. A medium- to long term strategy is currently being developed in collaboration with ID20 and the ISDD with the aim to (i) improve/automate the analyser production procedure and (ii) start the “mass” production of crystal analysers.

- Upgrade of the focusing mirror optics allowing a variable spot size from 300 x 50 μm^2 (horizontal x vertical) at the sample down to 50 x 50 μm^2 for medium-pressure applications.

- Upgrade of the double crystal monochromator in order to improve beam stability, increase the angular range and give the possibility to mount 3 crystal reflections and thus to optimise flux and energy resolution. It may become necessary to make the entire optics hutch compatible with the white beam.

- Upgrade of the X-ray source: installation of three revolving undulators with periods u35/u27.

Furthermore, there are plans to develop and install an in-vacuum low-energy XES spectrometer to reach the K edge of S and Cl, the L-edges of 4d elements, and the M edges of 5d transition elements and radionuclides. The instrument will be installed in the unoccupied hutch (EH1) located between the optical hutch and the actual experimental hutch of ID26. Funds for this project are available within the EcoX-Equipex project (Alain Manceau et al).

ID27

ID27 undertakes a very strong and diversified research program. Remarkable progress has been achieved in the determination of melting curves, x-ray fluorescence at high pressure and temperature, tomographic and imaging applications. In order to keep its world-leading role and to accommodate the various different experimental set-ups in a user-friendly fashion, a major upgrade of ID27 is required. This necessitates:

- The installation of cryogenic undulators to replace the U_{23} in vacuum undulators presently installed. This will result in a gain of a factor 4 in flux in the energy domain of interest.

- The purchase of a high Z, large area, high resolution (e.g. 50 x 50 cm with 5000 x 5000 pixels), high dynamic range (>18 bit), fast readout (100 ms) pixel detector. A Perkin-Elmer flat panel detector was recently purchased; this investment represents a viable solution for the next few years until the required detector technology has matured.

- An end-station in the EX2 extension building at ~110 meters.

This end-station will be devoted to combined high energy two and three-dimensional micro/nano-imaging and XRD at high pressure. The possibility to perform X-ray computed micro and nano-tomography combined with micro and nano-XRD is a very promising emerging domain and will clearly have a major impact in the future capabilities of beamline ID27. These methods are well-known for reconstructing three-dimensional images of a structure from a finite number of radiographic images, but due to experimental difficulties, such techniques have never been applied to samples subjected to extreme conditions. These experimental difficulties have been recently overcome at the APS by a group of researchers from the Geophysical Laboratory. Indeed, they were able to collect micro and nano-tomographic images of compressed micro-samples at high pressure in a large volume press (Drickamer type) and a diamond anvil cell. This new x-ray micro/nano-tomographic technique will find widespread use in accurate determinations of the equation of state of glasses and melts up to megabar pressures, which is crucial to many problems in Earth, planetary, and materials sciences. The combination of micro/nano-imaging and micro/nano-XRD will also provide a unique tool to the high pressure community to get better insight into the local structure/texture relations of highly compressed materials. In addition, the possibility to perform nano-diffraction experiments is certainly a way to extend the accessible pressure domain to the intrinsic limit of the diamond anvil cell (P>4 Mbar).

The micro/nano-tomographic end-station will also allow the collection of time resolved high quality two-dimensional images which can be used for the direct determination of fluids viscosity at high pressure. The determination of viscosity variation at high pressure and high temperature is of particular importance in various domains of research ranging from materials science (structure/viscosity relation) to geophysics where this parameter is the driving factor of mass and energy transfer of planetary interiors. This technique is based on the falling sphere method in the Paris-Edinburgh press. The displacement of the centre of gravity of a heavy metal sphere is followed in situ and real time (typically 10 ms resolution) using a high speed CCD camera. The viscosity is then directly obtained through the Stokes relation.

Finally, this new end-station will provide enough space to install the first generation of laser driven shock experiments coupled to XRD. This will shed new light on the elastic properties of highly compressed materials.

ID28

Throughout the last decade many applications of IXS with meV energy resolution have matured and in several aspects the optimum configuration has been implemented. This relates to, for example, the focal spot size and the number of crystal analysers. On the other hand, there are two projects, for which a substantial investment would offer new scientific possibilities and stimulate the user community, namely (i) a set-up for diffuse scattering studies to be operated in intimate conjunction with the existing IXS spectrometer, and (i) and a new IXS spectrometer with 0.6 – 0.9 meV energy resolution for the study of disordered materials.

For crystalline systems one of the main current limitations is the relatively long data acquisition time. On the other hand, it has been recognised since a long time that the intensity distribution of x-ray scattering by thermally populated phonons also carries information on the phonon dispersion. Though quantitative information from thermal diffuse scattering (TDS) can only be extracted for the most simple (mono-atomic, one atom per unit cell) systems, the strength of diffuse scattering studies relies in the data acquisition speed (complete 3-dimensional data set in typically 30 minutes), thus allowing the exploitation of a large parameter space (temperature, pressure, …) in a reasonable time. The combination of (T)DS with IXS studies and *ab initio* calculations offers fascinating new possibilities: (T)DS allows to rapidly identifying the interesting region in reciprocal space, and, in a subsequent focused IXS study, to disentangle static and dynamic (phonon) contributions. Along the same lines, the combination of IXS (from single and/or polycrystalline materials) and TDS provides very stringent constraints for the validation of *ab initio* calculations. This methodology leads to a significant reduction of data collection times. An experimental facility combining both techniques on the same beamline will offer a wealth of new opportunities. Representative examples comprise:

- Determination of elastic properties of elements and minerals in conditions of high pressure (and high temperature) using diamond anvil cell and laser heating techniques.
- Study of the coupling of electronic and/or magnetic fluctuations with phonons in strongly correlated electron systems.
- Vibrational properties of nanomodulated and low-dimensional systems.
- Time-resolved (ms -> s) TDS experiments in the study of phase transformations, *i.e.* the microscopic decomposition kinetics in ionic solids, and non-equilibrium phenomena.
- Determination of the shape of the Fermi surface in metallic systems by a directional tracing of Kohn anomalies.

Another important limitation is the achievable energy resolution and shape. This is of central importance for the investigation of disordered systems. A resolution of 0.9 meV has been reached already a few years ago, but the count rate was too low to perform routine experiments. Another difficulty often faced when dealing with disordered systems is the dominant quasi-elastic contribution, which obscures the inelastic spectral features. An improvement of the resolution function and its shape will represent a decisive improvement and pave the way for a new class of investigations, addressing a series of very fundamental issues:

- Viscoelastic crossover behavior of liquids and supercritical fluids, and new low-energy modes in complex fluids.
- Dynamics of confined liquids of relevance for materials science, biology, and geology.
- Sound bifurcation in binary mixtures.
- Acoustic properties in glasses: investigation of the cross-over regime (between low frequency (Brillouin Light Scattering) and high frequency (IXS) behavior of the damping coefficient in the $0.1 < Q < 1$ nm-1 region.
- Collective dynamics of lipid membranes and other biological systems, where correlated molecular motions and density fluctuations on the meV energy scale play a significant role in determining their physical and biological properties.

In order to enable the new scientific opportunities outlined above and to further strengthen the existing research programmes the following upgrades are proposed:

- An experimental set-up in EH1 for (thermal) diffuse scattering equipped with a state-of-the-art area pixel detector to be operated in parallel and in close complement to the existing IXS spectrometer. The set-up will offer the unique possibility to combine energy-integrated diffuse scattering with energy-resolved IXS studies.

- A small angle IXS spectrometer with sub-meV resolution for the study of liquid and disordered materials, which provides an overall energy resolution of 0.6-0.9 meV, and is equipped with at least five analysers at the same momentum transfer Q. In parallel, and starting in 2012, a R&D program will be launched to investigate and implement alternative monochromatisation schemes aiming at sub-meV energy resolution with steep tails.

The outlined upgrade plans not only signify a substantial increase in the performance and capabilities of the existing beamline, but offer a world-wide unique facility combining IXS and TDS in the study of lattice dynamics. Another key ingredient is the further development and availability of software packages and *ab initio* codes for the efficient preparation of the experiment and a rapid, on-line analysis of the collected data. This is in particular important for complex materials and (textured) polycrystalline materials.