Electronic Structure and Magnetism Group– Plans and Visions for Upgrade Phase II

Executive Summary

The beamline portfolio of the ESM group comprises two upgrade beamlines: UPBL 11 TEXAS (Time resolved and Extreme conditions XAS) , located at ID24 and BM23 and UPBL7, the soft x-ray beamline for magnetic and electronic spectroscopy. Within the scope of a possible Upgrade Phase II the following evolutions could be envisaged:

1. Upgrade project for replacing ID12– Advanced Spectroscopy Beamline
2. Upgrade project for advanced hard X-ray diffraction beamline under extreme conditions
3. A proposal for facilities to allow investigations of laser-induced extreme states of matter

The latter proposal is not for either upgrading a beamline nor a new beamline but rather to provide the required instrumentation across many beamlines (from soft to hard X-rays). This could easily fit with a major effort in extreme conditions. Here, extreme conditions are defined beyond the traditional high pressure and include very high magnetic fields, low temperatures (mK) high temperatures as reached by the laser heating, shock-waves, high electric fields as well as high pressure. The first two proposals also fit into this more general context. The idea of an extreme conditions partnership could also be thought about again in this more general frame.

1. Advanced X-ray Spectroscopy Beamline

The existing ID12 beamline which is dedicated to polarisation dependent X-ray spectroscopy in the medium to hard X-ray region shall be upgraded. This beamline has already contributed in establishing X-ray spectroscopy with polarised synchrotron radiation as very powerful tool to study fundamental properties of matter in an element selective manner *via* various order parameters, e.g., spin and orbital moments, electric dipole moment, orbital anapole *etc. X-ray magnetic linear and circular dichroism (XMLD, XMCD)* in ferro-, ferri- and paramagnetic systems, *X-ray natural circular dichroism (XNCD)* in noncentrosymmetric crystals as well as *non-reciprocal X-ray linear dichroism (XnrLD)* and *X-ray magneto-chiral dichroism (XMχD)* in multiferroics are now well established techniques to measure these moments.

This upgrade is aimed at developing the state of the art X-ray spectroscopy beamline dedicated to study behaviour of these order parameters under extreme conditions (high pressure, ultra low temperature, very high static magnetic and electric fields) and at the nanometers scale.

Sources: Helios type undulators
The spectral range from 2 keV up to 14 keV will be covered with the first harmonic of undulator spectra only. This provides highest flux and full control of polarisation of X-ray beam.

Optical scheme
X-ray spectrometer based on a double crystal monochromator with two independent pairs of crystals and focusing optics based on refractive lenses. Special arrangement is foreseen to compensate for chromatic aberrations of the lenses. This would be a unique beamline worldwide providing users with high flux of polarized X-rays with the beam size at the sample below 300 nm in the photon energy range from 2 to 14 keV .

Scientific areas

Science at Extreme Conditions

X-ray spectroscopy (including XMCD) under high pressure (up to 25 GPa), high magnetic field (up to 6Tesla) at low temperatures (down to 4K)
X-ray magnetic spectroscopy (XMCD and X-ray magneto-chiral dichroism) under very high static magnetic field going up to 35 Tesla using a HTSC solenoid.
X-ray spectroscopy studies of phase transitions at ultra low temperatures (down to 50 mK) and a magnetic field of up to 17 Tesla.

X-ray spectroscopy under applied electric field (up to 10^5 V/mm)

Nanoscience and Technology

Polarization dependent X-ray resonant reflectivity from ultrathin layers, trilayers and multilayers focused on either determination of the magnetization profile in magnetic systems or strains in semiconductor heterostructures

Dichroic imaging of polar domains in multiferroic thin films and crystals, ferromagnetic domains in thin films with strong magneto-crystalline anisotopy and twins in chiral crystals.

XMCD studies to fully characterize monodispersed magnetic nanoparticles, clusters, single molecular magnets or other nano-engineered magnetic nanomaterials.

The scientific scope of the beamline could go well beyond Magnetism and solid state physics to other fields like chemistry, environmental and geosciences, and even life sciences.

2. An advanced hard X-ray diffraction beamline under extreme conditions

Scientific motivations

This project is focused on an innovative instrumental development on high magnetic fields which will allow a new generation of synchrotron radiation experiments under combined extreme conditions to tackle several hot topics in the field of strongly correlated electron systems and advanced multifunctional materials. The aim is to attract a wide user community by providing an instrument able to exploite state-of-the-art methods of x-ray diffraction and spectroscopy in the hard x-ray energy range, combined with diversified and unique experimental environment facilities at the forefront of the synchrotron radiation instrumentation.

High magnetic fields interact with the magnetic moments, influencing the magnetic domain formation, inducing lattice displacements by magnetostriction, and promoting structural and magnetic phase transitions. In magnetic systems the ground state of a spin moment depends strongly on the relative sign and magnitude of exchange interactions and in turn on exchange pathways defined by interatomic distances and bond angles. By combining diffraction and spectroscopic experiments under simultaneous high pressure and high magnetic fields at low temperature new information could be obtained on the microscopic origin and symmetry of the ground state in unexplored region of the phase diagrams of these systems. The pressure, unlike doping, can tune the lattice constants without altering the topology of the Fermi surface, and such perturbations can result in changes that are manifested in the electronic behaviour, thus likely to lead to newly ordered phases. The measure of the variation of lattice and superlattice parameters by hard x-rays could provide detailed information on the symmetry of the ground state of several classes of materials like heavy fermion systems, low dimensional quantum magnets, geometrically and topologically frustrated magnetic systems, unconventional superconductors, multiferroic and multifunctional materials. The possibility to complement the standard resonant and non-resonant x-ray diffraction methods with x-ray absorption or emission spectroscopies (total fluorescence yield, resonant emission or Raman scattering) improve the versatility of the experimental setup and could attract the user community which already is active in this specific domain.

A new area of experimental development could be the introduction of a laser source to excite the low-high spin cross over transitions in the presence of high magnetic fields. For example the photo-induced magnetic transitions in charge transfer systems could be of decisive importance to understanding the role of spin-lattice coupling in the technological field of coupled magneto-optical devices.

Technical and experimental challenges

The beamline is designed for the energy range 3.5-50 keV with a standard design for diffraction studies (UHV, mirrors for harmonic rejection, fixed exit monochromator) with the possibility to implement a high resolution channel cut monochromator to perform spectroscopic studies. Polarization analysis and control in the resonant x-ray regime require a low divergent beam section and focusing could be provided downstream by KB focusing mirrors (or Be lenses at high energy experiments). A dedicated experimental hutch for high magnetic field will accommodate a dedicated diffractomenter for the following experimental environments:

- H: High magnetic fields
    split coil vert. superconducting magnets up to 17-20 T, with a dilution insert.
- T/VLT: Low Temperatures
    (T=1.5K) or Very Low T (dilution insert down to ~50 mK)
- P: High pressures
    membrane DAC and panoramic DAC up to 25 GPa
- E: High electric field
    Electric stick up to 3.5 kV/mm
- L: laser induced electronic transitions
    Optic window inserted into the 10T split coil magnet (already adapted for that)
- VH: very high magnetic field
    possibility to accommodate a future installation of resistive high field split-pair magnet up to 30 T.

Depending on the energy range, the following experiments could be envisaged:

H-E-T experiments (3.5-15 keV):
Polarization analysis of circularly and linearly polarized photons to investigate the multifunctional materials (multiferroics) under resonant and magnetic x-ray scattering regimes. This include the characterization of the magnetic structures in frustrated magnetic systems and the in -situ imaging of magnetic domains in compounds of technological interest.

H-P-T experiments (5-50 keV)
High photon energy experiments to observe the crystallographic symmetries and lattice displacements under high pressures (up to 25 GPa) and under a magnetic field (up to 10 T) by using the available 10T magnet. Intermediate pressure range could also be exploited in resonant x-ray scattering regime, as demonstrated in explorative experiments done in the past (resonant magnetic diffraction at Ce L_3-edge). Raman and fluorescence yield experiments could also be envisaged by adapting an appropriate analyser in the detection arm (3 m spherical analyser in horizontal configuration).

H-VLT experiments (10-50 keV)
Design of a new vertical split-coil superconducting magnet (up to 17-20 T) with a small bore and a dilution insert to reach very high magnetic fields at very low temperatures. This experiments could be done only at high energy because the thermal heating.

L-H-T experiments (3.5-10 keV)
Pump (laser) and probe (x-rays) experiments combining optical spectroscopy and x-rays diffraction under high magnetic field to investigate the induced magneto-optical transitions in spin-crossover systems. An optic window inserted into the 10T to combine laser illumination and magnetic diffraction.

3) Investigating laser-induced extreme states of matter at the ESRF

The scientific case

Under strong optical excitation conditions, it is possible to create highly nonequilibrium states of matter. Today, intense femtosecond laser excitation can produce transient states of matter that would otherwise be inaccessible to laboratory investigation. As visible and near-infrared laser light is absorbed by the electrons, intense short-pulsed optical excitation initially forms highly nonequilibrium conditions (that is, very hot electrons within a cool lattice). The nuclear response is determined by the rate of energy transfer from the excited electrons to the nuclei and the instantaneous effect of change in electron distribution on the interatomic potential energy landscape. At high excitation densities, the interatomic forces that bind solids and determine many of their properties can be substantially altered.

The time required to heat the phonon system depends on the particular material but lies typically in the range of picoseconds. Consequently by using short laser pulses (femto-to picosecond duration) of sufficient energy, very high heating rates of several tens or hundreds of Kelvin per picosecond can easily be reached. Thus, it is expected that within a few picoseconds a solid may be heated to very high temperatures which, transiently, exceed well the melting temperature and also the static stability limits.

If the laser pulse is sufficiently short, heating occurs at constant volume. However, if the high power laser pulse is long enough to allow energy transfer between the electrons and the ions (~ ns), heating occurs simultaneously to volume changes. Dynamic excitation in this case can be used to generate an equilibrium thermodynamical state along a Hugoniot equation, where temperature and pressure are related. By using high power ns long laser pulses, studies of transient states can be extended to very high pressures (> 100 GPa), and matter can be probed in a similar way as done in static high pressure experiments.

The internal energy associated with compression to the 100 GPa level is roughly $E \sim - P \quad V \sim 10^5$ joules per mole of atoms with volume changes V being ~ 20% of the 5 cm^3 typical molar volume of terrestrial planet matter. The work of compression thus corresponds to bonding energies (~ 1 eV = 97 kJ per mole, characteristic of the outer, bonding electrons of atoms) meaning that the chemical bond is profoundly changed at a pressure of 100 GPa. This expectation has been verified through numerous experiments showing that the chemical properties of matter are significantly altered under pressure: for instance, hydrogen, oxygen, and the “noble gas” xenon transform from insulating, transparent gas, fluids or crystals at low pressure to become metals by ~ 100 GPa.

Pre-compressed matter (e.g. using diamond anvil cells) under well known pressure conditions can be heated and further compressed using high power lasers, providing access to pressure-temperature conditions unobtainable by either technique alone. This combination of static and dynamic compression has been experimentally demonstrated and ultimately provides access to the 10-100 TPa pressure range, relevant to planetary science, testing first–principles theories of condensed matter, and experimentally studying a new regime of chemical bonding. In fact, compressional energy changes can reach the keV in the 100 TPa regime, comparable to energies of core-electron orbitals. Deep-electron levels within the atom can therefore participate in chemical bonding, and an entirely new type of chemistry becomes accessible in a (subnuclear) regime that is as yet unexplored.

There has been considerable experimental and theoretical interest in both laser-induced transient states of matter as well as shock wave compressed matter. In the former case, the scientific interest is to investigate phenomena for which the temperature of the electrons differs from that of the ions, and where the interaction between nuclei is altered. In the latter case, the interest is to study the formation and the properties of high density plasmas at local thermal equilibrium, also referred to as Warm Dense Matter (WDM). In density – temperature space, WDM is situated between solid (cold) matter and kinetic (hot) plasmas. This domain is broadly defined as the parameter space for materials with densities between standard density and 10 times this value, and temperatures ranging from 0.1 eV to 100 eV (10^3 to 10^6 K). Here we deal with plasmas that are i) weakly ionized ii) strongly coupled and iii) partially degenerate. Typically this thermodynamical regime can be found in the laboratory, during experiments that start with a solid and end in a plasma, as well as in astrophysics in the interior of giant planets and dense cold stars. One of the main purposes of studying WDM is to understand planetary interiors in terms of equations of state (EOS) and other material properties. Only through such understanding can we hope to predict planetary formation and evolution.

In the WDM domain, both types of material models (condensed matter and plasma) break down, such that at present there is no capability to predict accurately the behaviour of matter under such conditions. In WDM the plasma exhibits long- and short-range order due to the correlating effects of the atoms and ions. This intriguing regime, where the plasma environment can no longer be considered a thermal bath and the behaviour of its particles is no longer well described by characteristics of isolated ions provides a tremendous challenge to theory.

The current knowledge on this regime is limited because the standard theoretical approaches fail, but also because experiments are extremely difficult. The difficulties in the field of theory arise from the lack of obvious expansion parameters, as the usual perturbative approaches used in plasma phase theories are no longer valid. Furthermore, the importance of density effects (i.e. pressure ionization) increases, as the plasma environment starts to influence substantially the internal structure of ions or atoms. The experimental problems follow from a difficult isolation of well-defined plasma samples and their complicated diagnostics. So up to now there has been a serious lack of experimental data referring to matter in this regime.

Both theoretical and experimental difficulties arise also because before reaching this warm dense thermodynamical state, matter goes through all the complex and still unclear transient phenomena described above, that occur between the moment the laser hits the sample and the moment the sample exploses: the absorption of laser energy by free electrons, the transfer of part of this energy to ions, the dynamics of fusion of the crystalline lattice, the compression of the fluid (or solid – depending on the energy density deposited and on the confinement of the sample) due to the travelling shock wave, etc… From an experimentalist point of view, achieving insight into the electronic structure and local order in matter at these extreme conditions appears as very challenging and exciting.

Scope

The scope of this Phase II proposal is to make a a coherent action towards implementing the investigation of laser-induced extreme states of matter on several ESRF beamlines. This initiative would receive full support from many european research centers, such as Institute of Shock Physics (London, UK), CEA (Paris, France), XFEL and Petra III (Hamburg, Germany). In this context, the ESRF is still (maybe not for long) the only synchrotron worldwide that provides highly specialized beamlines for extreme conditions studies using diffraction, absorption, and emission spectroscopies. A coherent action involving these beamlines (and eventually more) would place the ESRF in a very favorable position for producing state of the art science with respect to other facilities.

The scientific focus shall be on the investigation of the dynamics and the electronic and structural properties of laser-induced extreme states of matter. Studies at P and T conditions of Earth core and other planetary bodies, or of dynamics of meteorites and asteroid impacts, and physics in the warm dense matter regime and beyond, etc…, are being routinely addressed through shockwave experiments at large high power laser facilities. But today these investigations can in principle be carried out at high brilliance synchrotron facilities coupled to relatively low power excitation lasers, thanks to the enormous reduction in size of the probed volume of the sample[1]. No synchrotron has yet invested into this very challenging domain with a coherent action, yet these studies would open the way to probing the structure and electronic states in matter at conditions so extreme that they are impossible to achieve by static methods.

Among the many physical states that can be produced in matter by laser excitation, those where the ESRF can make significant advances with respect to other facilities worldwide, because it has an evident advantage in terms of existing know-how and methods developed, are those at local thermal equilibrium. These can be probed in the ns timescale, with x-ray spots of dimensions of a few microns and equivalent sample thicknesses. This science is therefore fully compatible with the hard x-ray energy range and brilliance of ESRF beamlines.

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[1] To give a few numbers, it is well known that the laser power required to bring matter in a warm dense matter state depends on the value of the bulk modulus at ambient pressure and on the probed volume. The typical volumes of laser targets used today for these kinds of experiments are of the order of $\sim 10^{-3}$ cm^3 . These volumes require a laser power of the order of 100 J to bring Fe into the WDM regime. In view of future laser–shock experiments at the ESRF this value can be scaled down with the probed sample volume and can easily be reduced a factor 10^2 or 10^3 thanks to the small focal spot (< 500 m^2 total size) and sample thickness (~ 10 m). This is important because reducing the power (i.e. size) of the laser facility allows to make such experiments feasible at the synchrotron. Lasers of 10 J are table-top and transportable, and could be easily implemented at ESRF. This power would allow to investigate not only the very important 3d metals, but also access the whole 4d and 5d series.

Dynamic studies of transient states and of matter out-of equilibrium are on the other hand more challeging but not out of reach for the ESRF. Besides the need to push time resolution to within the ESRF bunch length (< 100 ps), the main difficulties lie in having to match the optical excitation and x-ray probed volumes. These processes, which occur within a few ps from excitation and concern at most the first few nm of exposed matter, are most efficiently studied using soft x-ray absorption spectroscopy, x-ray surface diffraction or reflectivity, techniques which are all implemented in highly specialized ESRF beamlines.