

APS for an XMCD and XMLD beam line on SOLEIL in the 350 to 2000 eV range

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Summary of beamline specifications

- Photon energy : 350 to 2000eV
- High flux : $\sim 10^{11}$ ph/s on the sample
- Energy resolution $\Delta E/E \geq 6000$ over the whole energy range
- Very high purity and stability of the beam at source and experimental station
- Variable linear and circular polarization
- Monochromator with different gratings
- A two stages horizontally focusing optics
- A KB system for a beam spot size of $\sim 10 \times 20 \mu\text{m}^2$
- 2 XMCD end-stations (with introduction chambers and at least vertical and polar angle degrees of freedom):
 - High field (7T superconductor magnet) and very low temperature (lowest T ~ 50 mK) (pumped ^4He cryostat),
 - Fast flipping magnetic field (2T quadrupole electromagnet) and temperature range from 10 to 1000 K (^4He cryostat and oven).
- A reflectivity studies end-station (under development at the *Laboratoire de Cristallographie de Grenoble*)
- X-ray microscope
- A MBE chamber, for in-situ sample preparation and characterization (including: evaporators, AES, LEED, RHEED, STM), connectible to the different analysis chambers.

Average cost : 20 MF

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I – Introduction

The community of magnetic x-ray absorption is well identified in France. Several major French schools have been held in Mittelwihr under the supervision of the Institut de Physique et Chimie des Matériaux de Strasbourg (IPCMS) where the whole community gathered (Mittelwihr 1989, Mittelwihr 1996, Mittelwihr 2000). Every year a national dedicated workshop is also held in Strasbourg to outline the new trends in the field. Moreover two workshops have been organized by SOLEIL in Strasbourg, first during the Foundation Phase (1997) and at present (May 2001) in order to identify the objectives that should be fulfilled by the SOLEIL beam lines and the expectations of the French community.

The magnetism community using synchrotron radiation has also established connections with other scientific communities making use of circular polarization among which one can quote the community gathered under the action of José Goulon in the workshop on gyrotropy: "X-ray gyrotropy and synchrotron radiation based chiroptical spectroscopies" (ESRF-Grenoble, 21 - 23 September 2000).^{1,2,3}

From these various workshops, active teams in the different domains have been clearly identified, their instrumental needs are particularly well expressed in synthesis documents and their interactions with related techniques are underlined. It is important to note that the updated scientific priorities of the laboratories involved in the field have been exposed during the last SOLEIL meeting (Strasbourg 2001) whose conclusions have inspired section II of the present proposal.

The various themes in which the French community is involved concern the emergence of magnetism in exotic compounds (molecular magnets, hybrid organic-inorganic magnets) and in compounds with low dimensionality (magnetism at surfaces, in thin layers and nanostructures, superparamagnetic nanoclusters, high spin molecules, ...).

The application of synchrotron radiation to magnetism is related to the development of circularly polarized sources. The main advantage of X-ray Magnetic Circular Dichroism (XMCD) over other techniques addressing magnetic behavior lies in its chemical and orbital selectivity. One can probe the magnetic moment of one specific atom in a compound in which the absorbing atom is not a major component. Due to the electric dipole nature of the Hamiltonian interaction, one can also probe specific shells for which the magnetic orbital and magnetic spin moments can be "independently" measured.

In section II we enumerate some of the major trends in physics, chemistry and Earth sciences for which XMCD and X-ray Magnetic Linear Dichroism (XMLD) should provide new insight and in section III, the main features of the beamline proposal, dedicated to XMCD and XMLD in the soft X-ray range are presented. Section IV places the proposal in perspective within the national and international context and outlines the supporting community.

II – Scientific Case

Purely molecular magnets: *Laboratoire de Minéralogie et Cristallographie de Paris (LMCP), Laboratoire de Chimie Inorganique et Matériaux Moléculaires (LCIMM), Laboratoire d'Utilisation du Rayonnement Electromagnétique (LURE).*

In most magnets, magnetism results from the presence of transition elements (essentially 3d elements), of rare earths or of actinides. There is another class of magnets, called *molecular magnets* that do not contain any of these elements. The magnetism in such compounds does not come from the build-up of a magnetic 3d band, nor from localized 3d, 4f or 5f electrons but from molecular complexes, generally nitroxides, in which the highest occupied molecular orbital possesses only one single electron. These NO groups are in ferromagnetic interaction inside the molecules and the molecules are organized in a three-dimensional network where they are ferromagnetically coupled through magnetic dipole interaction.

The first molecular ferromagnetic complexes have been synthesized ten years ago, starting from p-nitrophenyl nitrony nitroxide radical.^{4,5,6} The Curie temperature of such compounds is very low (less than 2 K up to now). The physics of magnetism at stake is completely different from what one is used to deal with: the spin carriers are not atomic orbitals, neither delocalized bands. Consequently, the methods of localized magnetism and band magnetism are unable to describe the magnetic behavior of such compounds. For instance the compound synthesized by the team of Rassat can be fully polarized in 10 mT at 10 mK, is not saturated at 1K in 1T and does not present any detectable coercive field in hysteresis curves.

In a related class of magnetic compounds, the nitronyl nitroxide group is a mediator of the magnetic interaction between 3d transition elements or rare earths. In case of diamagnetic rare earth, one can observe antiferromagnetic coupling. When the rare earth is paramagnetic and the nitronyl nitroxide groups are bound to the rare earth in a linear chain, one observes complicated magnetic behavior due to the various antiferromagnetic coupling between neighbors.⁷ The site selectivity of XMCD is at this point essential to detect the small although essential magnetic polarization of nitrogen and oxygen.

Langmuir-Blodgett films: *Institut de Physique et Chimie de Strasbourg (IPCMS), LURE.*

The spreading of amphiphilic molecules on a ultra-pure water surface gives rise to the formation of a monomolecular film called "Langmuir film": the hydrophilic part of the molecules is in contact with the water, whereas the hydrophobic part goes away from it as much as possible. The film is therefore vertically structured and confined in two dimensions (2D), at the air-water interface. Most of the time, Langmuir films can be transferred onto solid substrates. They are then called Langmuir-Blodgett (LB) films. In addition to offering a greater ease for the study of 2D phenomena, LB films provide us with an outstanding facility for the processing of supramolecular assemblies, allowing for the deposition of compounds in sequences of layers. In the case of homogeneous deposition, one can for example build up non-centrosymmetric structures of optically active materials, whereas alternate deposition enables the elaboration of complex structures in which each layer plays a precise part, e.g. electron donor or acceptor, conductor or insulator.

M. Pomerantz at IBM has processed the first LB films with magnetic properties.⁸ Magnetism was the result of the presence of Mn atoms, co-deposited with stearic acid. Since then, many organo-metallic compounds have been processed as LB films. Magnetism is brought in by some metallic ions, and these studies correlate the

magnetic properties of the molecules in the bulk and in the LB films. In another study,⁹ the possibility of creating multilayered LB films in which the metallic planes can be magnetically coupled or decoupled according to the elaboration procedure is demonstrated. Last, the observation of a phase transition to a magnetically ordered phase has recently been reported for a LB film containing gadolinium.¹⁰

On purely organic magnetic films, the observation of ordered structures in a LB film of a nitronyl-nitroxide radical has been done with Magnetic Force Microscopy,¹¹ and the magnetic anisotropy of the film revealed by ESR.¹²

Hybrid magnetic materials: *LCIMM, LURE, IPCMS.*

A large interest is devoted to the study of compounds of low dimensionality intermediate between magnetic chains and magnetic planes. In lamellar compounds where the 3d transition elements are copper and vanadium, ferrimagnetism has been obtained as a consequence of some topological order, also related to magnetic frustration. At low temperature, one assists to the emergence of a three dimensional antiferromagnetic order that is governed by the interplanar interaction monitored by hydrogen bonds. The intercalation of various organic species between the magnetic planes controls the dimensionality of the magnetic order.

In this vein, synthesis is developed to produce organic/inorganic compounds in which inorganic planes are related by organic pillars. The organic pillar can be magnetically inactive or active. Nitroxide radicals have been tested as well as ligands with π conjugated electrons. One expects original magnetic behavior from the presence of localized magnetism inside the planes coupled to strongly delocalized π electrons.

High spin molecules: *LMCP, Dante Gatteschi (from the LCIMM permanent address in Firenze), Laboratoire Louis Néel de Grenoble, LURE.*

The synthesis of polynuclear molecules has recently produced a large variety of compounds in which transition elements are magnetically coupled to produce a huge molecular magnetic moment.^{13,14,15,16} The perspective concerning this work is inscribed in the conception of materials for the nanotechnologies where the target is to address specifically one nanoparticle or molecule, to store in it a piece of information, to transmit it and to collect it at a molecular level. The high spin molecules can have large magnetic moments: such as heptanuclear complexes based on chromicyanide that can carry a moment as large as $27 \mu_B$.¹⁷ These molecules are completely monodisperse, carry a well-defined magnetic moment and possess also a well-defined anisotropy. These specific magnetic properties made possible the observation of superparamagnetism and quantum spin tunneling.¹⁸ This is a consequence of their inherent properties: large magnetic moments, large uniaxial or rhombic anisotropy and large exchange coupling and also intermolecular interactions. All these parameters can be tailored by the tools of coordination chemistry so that these types of compounds would be candidates for q-bits in the molecular electronics of the future.

Superparamagnetic nanoparticles: *LMCP, Laboratoire de Physique des Matériaux de Lyon, IPCMS, LURE, Institut des Matériaux de Nantes (IMN), Institut de Physique du Globe (IPGP), Laboratoire de Chimie de la Matière Condensée (LCMC), Laboratoire de Chimie Inorganique et Electrochimie des Matériaux Moléculaires, Laboratoire de Chimie des Matériaux Divisés et Catalyse.*

Through various routes of syntheses, one can produce nanoparticles such as magnetite (Fe_3O_4), maghemite ($\gamma\text{-Fe}_2\text{O}_3$), hematite ($\alpha\text{-Fe}_2\text{O}_3$), pyrrhotite (Fe_{1-x}S) or greigite (Fe_3S_4). These nanoparticles are present at the surface of the Earth and are responsible for what is called the paleomagnetism: paleomagnetism is the study of the magnetic properties of magnetic minerals.^{19,20} From paleomagnetism one can understand the evolution of the tectonic migration of the various Earth plaques. Paleomagnetic signatures are also geological thermometers and barometers informing on the condition of formation of minerals. It is known that the paleomagnetic signature comes from single domain magnetic nanoparticles that have been oriented in the dipole Earth magnetic field when cooling through the blocking temperature. The joint point of view of the chemist and the paleomagnetician is essential to unravel the differences between the magnetic properties of natural nanoparticles and their bulk analogs: magnetic surface canting, chemical and magnetic disorder, vacancies ordering.^{21,22,23}

The recent findings concerning the Martian magnetic map has revealed that an exceptionally large paleomagnetic signature was present at the surface of Mars.^{23,24} The origin of such a signature is still completely unknown. In the perspective of the spatial mission Mars Sample Return in which France is deeply involved at the side of the NASA, it is essential to refurbish our instruments in order to be able to handle and study small quantities of paleomagnetic materials. The expected size of Martian samples that the laboratories will be able to handle will certainly be limited to a few micrometers. The investigation methods will have to be non destructive so that x-ray techniques will be in good position. Moreover the search in such samples for nanospinels and nanogreigites produced by magnetotactic bacteria could be one of the clues in the quest for life from outside the Earth.

The magnetic anisotropies, either magnetocrystalline or shape anisotropy, govern the macroscopic magnetization that can be observed in a collection of nanoparticles. The anisotropies are directly related to the size of the particles and result from local anisotropies in the core and the surface of the particles that can also be influenced by interparticles interactions.^{25,26} The local phenomena are almost unexplored and require the use of site selective and chemically selective magnetic spectroscopies such as XMCD.²²

Beyond the paleomagnetism, granular systems are of high interest for their technological application since they are a direct extension of the studies performed on multilayers. However up to now, the superparamagnetism character is a technological obstacle for future applications in high density magnetic storage at room temperature, it is interesting to study pure or alloyed nanoparticles prepared by different ways (chemical, thin films lithography, clusters deposition...) in view to repel this limit²⁷ but also, from a fundamental point of view,²⁸ to describe the magnetic behavior of single molecules.²⁹ An XMCD measurement performed on magnetic nanoparticles assemblies under UHV (to avoid contamination in such reactive systems) and at various temperatures (from a few Kelvin to room temperature) is a unique tool to reach their local magnetic moments.^{30,31}

The idea to adjust the magnetocrystalline anisotropy and to raise the blocking temperature up to room temperature, by preparing alloyed nanoparticles comes up against segregation effects which play an important role in such systems where the surface/volume atomic ratio is nearly one.³² So, it would be of particular interest to anneal under UHV mixed nanoparticles embedded in a non miscible matrix in order to re-crystallize the alloyed grains while following in-situ the selective XMCD signals and the hysteresis loops as a function of fields and temperatures. In some cases, a parallel study of an assembly of non correlated clusters A diluted in a matrix

B, and of a bilayer sample A/B is fundamental to fully understand the interface magnetic behavior in terms of stresses (magneto-elastic anisotropy), magnetic polarization, charge transfer.

Surface and interface magnetism - Materials for spin electronics: *IPCMS, LURE, UMR CNRS/Thalès, Centre d'Etudes Nucléaires de Grenoble (CENG), Laboratoire Louis Néel de Grenoble, Laboratoire de Cristallographie de Grenoble, Laboratoire de Physique des Matériaux de Nancy (LPMN), LMCP.*

The magnetic behavior of a material can be deeply modified by reducing its dimensions along one or more directions in space. Atomic engineering is today able to produce nanostructures that disclose the interplay between dimensionality and magnetism. Magnetic properties without counterpart in the bulk have been discovered in quasi two-dimensional systems, such as ultrathin films and superlattices. Similarly small magnetic clusters, supported on surfaces or embedded in a matrix (see § *Superparamagnetic nanoparticles*), as well as magnetic oxides and semiconductors, will play an important technological role in the near future either in the field of spin electronics³³ or for high density magnetic storage. In this field we can quote the Magnetic Tunnel Junction³⁴ and the Spin-valve Transistor³⁵. In these scientific cases magnetic dichroism will play an important role. Its high sensitivity, its chemical selectivity and its unique capability to separate the spin and orbital contributions to the magnetic moment (using the XMCD sum rules) are ideal to characterize the magnetic properties of low dimensionality systems (evolution of the spin, the orbital magnetic moments, the blocking temperature in the superparamagnetic regime, ... with the size of the nanostructures).

In this framework, recent studies have been performed on self-organized systems. Such systems are obtained through the specific nucleation-growth process of an adsorbate on a substrate with “controlled” surface features like steps or surface reconstruction. By monitoring the amount of material deposited on the surface, it is possible, for instance, to induce morphological transitions going from finite size cluster, to one-dimensional wires and then to two-dimensional ultrathin films.³⁶ The main results of such studies is the observation of a large increase of the magnetic orbital moment when reducing the size of the nanostructures, a feature which can be explained as a higher contribution from the edge atoms where the quenching of the orbital moment is less effective. *A contrario* the magnetic spin moment does not show such a clear relation with the size of the nanostructures. Going towards the atomic character a very recent work³⁷ has considered transition metal impurities deposited on K and Na thin films. The multiplet structure of the XAS spectra indicates that Fe, Co, and Ni have localized atomic ground states. XMCD shows that the localized impurity states possess large, atomic-like, magnetic orbital moments that are progressively quenched as the isolated atoms form clusters.

XMCD also allows a direct access to the magnetocrystalline anisotropy (through the measurement of the anisotropy of the orbital magnetic moment), and this for magnetic nanoparticles as small as few tens of atoms. This potentiality is very important, since perpendicular magnetocrystalline anisotropy is a key parameter for the high-density magnetic storage technology. Several studies³⁸ have addressed the effects of the reduced dimensionality as well as those of anisotropic defects on the surface (such as regular steps on vicinal surfaces or induced dislocations) on this magnetocrystalline anisotropy. The goal is to understand the role of different parameters such as broken bonds or strain, among others, over the magnetocrystalline anisotropy and to be able to use them to favor a particular magnetic anisotropy.

The chemical selectivity of this technique allows to individually follow the magnetic behavior in complex magnetic systems such as alloys, multilayers (with the problematic of the induced magnetism at the interfaces) or particles embedded in a matrix. This is particularly important in the field of spin electronics with the advent of new materials such as complex magnetic oxides (double perovskites with high Curie temperatures) and magnetic semiconductors (II-VI and III-V semiconductors doped with 3d elements). Considering magnetic oxides, even the induced magnetism on oxygen can be measured. In the case of an interface (key parameter in such systems³⁹) between a metal and its oxide (e.g. $\text{Fe}_3\text{O}_4/\text{Fe}$ or NiO/Ni), it is possible to take benefit of the different signatures of the XMCD signals between the metal and the oxide to separate those magnetic contributions. Using linear polarization of the X-ray beam, the magnetic moment of antiferromagnetic material is accessible. This makes it useful in the field of magnetoresistive sensors where sandwiches of ferromagnetic and antiferromagnetic materials are used; moreover it would be ideal to understand the interface between the antiferromagnetic and the ferromagnetic layers, for instance the origin of the exchange bias is still not well understood.

Additionally using the time-resolved mode of SOLEIL dynamic studies in the nanosecond range can be performed with XMCD as demonstrated by Bonfin *et al.* from the Laboratoire Louis Néel (Grenoble).⁴⁰ Indeed the study of magnetization dynamics in thin magnetic films has become a matter of high interest for the future of magnetic recording and non-volatile magnetic memories. Parallel to the evolution towards smaller magnetic bits and memory cells, writing and reading times approaching the GHz range will be required in a few years from now. The group of *Laboratoire Louis Néel* has developed a new original technique to study the magnetization reversal dynamics of thin films with element selectivity in the nanosecond timescale. XMCD measurements in pump-probe mode are carried out taking advantage of the time structure of synchrotron radiation. Magnetic pulses created by a copper microcoil are synchronized with the photon pulses (in single bunch mode at the ESRF). The dynamics of the magnetization reversal of the studied layer is obtained by carrying out XMCD measurements as a function of the time delay between pump and probe. The ultimate time-resolution of the technique is given by the width of the photon bunch (100psec at the ESRF). The dynamics of the magnetization reversal of each of the layers of complex heterostructures (spin valves or tunnel junctions) can be probed *independently* by tuning the photon energy to the relevant absorption edge.

Less "usual" XMCD experiments can be carried out on "Ferromagnetic metal/Oxides/Ferromagnetic metal" systems by applying bias voltages between the uppermost layer (oxide) and the ferromagnetic metal. This procedure implies to use some nanophysics facilities⁴¹ in order to obtain good electrical contacts. For a given thickness of the oxide barrier (1 to 2nm), it is possible in principle to obtain the polarization of the tunneling current⁴² (the crucial parameter in which we are interested) by looking precisely to the magnetic properties of an ultra thin ferromagnetic layer deposited on top of the oxide barrier. This ferromagnet will be made of another material than the underlying ferromagnetic metal in order to have no interference between the two systems. By polarizing the system (a few volts will be sufficient) the spin polarized tunnel current will be significantly increased (value up to $1000 \text{ A}/\text{cm}^2$ may be expected). The effect on the topmost ferromagnet layer will be measured by XMCD. Those studies could be done by varying the thickness of the barrier and/or by changing the nature of the interface.

Related techniques: *Laboratoire de Cristallographie de Grenoble, CENG.*

Other techniques based on "reflectivity" using the "magnetic sensitivity" of the polarized X-ray light are possible: XRMS (X-ray Resonant Magnetic Scattering)⁴³ and DXRMS (diffuse x-ray resonant magnetic scattering).⁴⁴

While it can be expected that magnetic properties of nanoscale magnetic layers are different at interfaces with dissimilar materials than in the center of the magnetic layer, characterizing such differences with adequate depth resolution remains a great experimental challenge. This q -space depending technique has been used to directly probe the extension of the induced magnetism from the interface across a spacer layer in Ce/Fe and CeLa/Fe systems.^{45,46} In particular, together with structural studies (chemical profile), this approach allows one to describe the magnetic properties of the interfaces and should bring new insights in the investigation of systems exhibiting perpendicular magnetic anisotropy. A direct determination of the magnetization properties on both sides of the interfaces in multilayers and a determination of the extension of the interface effects throughout the layers is of prime importance and will allow us to go beyond the determination of the perpendicular magnetocrystalline anisotropy (PMA) constants averaged over the whole layer.

That approach should also be of great interest in the investigation of interlayer exchange coupling and exchange bias coupling. Nanoparticles of magnetic transition metals embedded in either a metallic or insulating matrix are also of interest (see above). For potential applications, as high-density magnetic recording media, a fine control of the particles size and inter-particle distance is crucial. Different synthesis methods are dedicated to this task. Moreover, strong efforts are currently undertaken in order to achieve a lateral patterning of magnetic films on a nanometer scale, by electron beam lithography or self-organizing growth mechanisms with the aim of confining the magnetic properties in an array of dots. In both kinds of system, the magnetic sizes and/or transport properties of the particles are sensitive to factors such as their lateral ordering, as well as to the interaction between them. Therefore, it is essential to develop new methods allowing to investigate directly the structural and magnetic properties of patterned and hybrid magnetic materials. The DXRMS use the large enhancement of the scattering amplitude together with the circular dichroism at the absorption edges of magnetic material resulting in a huge diffuse scattering contribution. This technique is successfully used to study magnetic correlations in sample presenting an in-plane periodicity in the nanometer range.^{44,47}

A defined experimental set-up is needed for such experiments in particular for the specular and off-specular XRMS (which could be possible through the installation of the reflectometer of J.M. Tonnerre for instance). The proposed beam line is particularly relevant for the energy range covering the $M_{4,5}$ edges of rare-earths. Indeed, for the $L_{2,3}$ edges of $3d$ metals, the reflectometer can also be installed on another proposed beam line dedicated to inelastic and elastic scattering. However, for DXRMS it is possible to take advantage of the proposed XMCD setting by adding spatial resolution to the photon detection (camera or slits moving in front of a diode).⁴⁴

III – Beam line characteristics

The source.

The source main characteristics will be a high intensity enabling to investigate highly diluted absorbing atoms, tunable polarization in order to select a specific state of polarization in the energy range 350-2000 eV covering the $L_{2,3}$ edges of $3d$ transition elements as well as the $M_{4,5}$ edges of rare earths. The access to the K-edges of important ligands such as nitrogen and oxygen as well as other light elements (sodium, magnesium, aluminium and silicon) will equally be possible.

The definition of the source is not completely settled. This will be done through a joint work with the team in charge of designing insertion devices on SOLEIL.

The Uxx sources proposed for SOLEIL have common features and are labeled by their xx period. For energies between 350 eV and 2 keV, a U50 or U60 design might be appropriate. We consider in the following a 3 modules device with a total length of 5.4 m. The brilliance for both undulators is in the range of 10^{20} ph/s/%bw/mm²/mrad². This corresponds to a photon intensity of more than 10^{15} ph/s/%bw through a pinhole aperture of 2mm(hor)×1mm(vert). Since the monochromator will have between 5 and 8 different reflecting surfaces, the photon intensity will be reduced by at least a factor ≈ 1000 for the highest energies (close to 2 keV). Considering an energy resolution $\Delta E/E$ of about 10^{-4} this should provide around 10^{11} photons/s on the sample. If one considers that the spectral region where the flux is never less than one tenth of the maximum defines the effective energy width of emission for an undulator, the intensity on the sample will always be larger than 10^{10} photons per second on the sample.

The design of the undulator should provide purely circularly polarized x-rays with Stokes quadrivector as close as possible to the optimum (1,0,0, ± 1). In order to perform XMLD measurements, it is also essential to have access to variable linear polarized light with optimal Stokes parameters (1, ± 1 ,0,0). Having the other types of linear polarization (giving (1,0, ± 1 ,0)) is still beyond what can be obtained by the technology of today.

The inversion of the polarization should be as fast as possible. If it can be done at high rate (faster than a few Hz) inversion of the photon helicity can be performed at each energy point of a scan, in the other case the polarization has to be inverted for each scan. In this scheme, an APPLE II undulator with permanent magnets is proposed. Due to technical constrains, the polarization will be more likely flipped at a moderate rate (several seconds) and will not be inverted at each energy point. The idea of an electromagnetic wiggler for which inversion of circular polarization would be in the range of several Hz is still under study (compatibility with other beam lines and stability of the electron beam).

We are still at work concerning the polarization state for photons above 1 keV (third harmonics).

A special care will be given to the stability of the source with respect to perturbations induced by other elements in the ring (gap and phase changes of other undulators or wigglers). The stability of the source is a crucial point for XMCD measurements that deal with differences of spectra usually registered sequentially. The phase change of the undulator should fulfill the following requirements:

- ° to be as fast as possible,
- ° avoid any intense transient heat load during the phase change,
- ° deliver identical spectral signature for both helicities,
- ° ensure that the Stokes parameter being initially (s_0, s_1, s_2, s_3) , should be $(s_0, s_1, s_2, -s_3)$ after the phase change with no change on the two other polarization rates $(s_1/s_0, s_2/s_0)$, a conservation of the same

polarization degree ($\sqrt{s_1^2 + s_2^2 + s_3^2} / s_0$) and a mere inversion of the circular polarization rate (s_3/s_0).

The spectral line width of an undulator such as Apple II or U50 is not large. In order to perform scans on a wide energy range (more than 100 eV), it is essential to pilot the width of the gap in a so-called gapscan mode. The data collection software should include this requirement.

Optics - Monochromator.

Due to the large horizontal size of the source, in the 350 μm range, it is necessary to have a two stage horizontally focusing optics in order to achieve an almost circular image on the sample with sizes in the range of 10 to 20 μm . The first optical element will be a horizontally focusing mirror (HFM1). It will be manufactured in crystalline silicon with roughness better than 3 \AA rms and coated with platinum. The incidence angle on the HFM1 will be 1.5 $^\circ$. The slope error will depend on the actual shape of the mirror. A spherical shape will be preferred. This first optical element is necessary to satisfy three conditions:

- reduce the heat load on the dispersive optics
- deviate the photon beam from the bremsstrahlung spurious radiations
- reduce the horizontal size of the photon beam

For all the following elements, the optical surfaces (mirror and grating) will equally be made of crystalline silicon. The surfaces will be coated with Pt, Au or Rh.

We are still considering the presence of an entrance slit. For XMCD measurements, one of the essential requirements is the energy stability of the monochromatic flux. This is best achieved with an entrance slit coupled to a vertically focusing mirror (VFM1). However this may induce flux variation due to changes in the beam positioning in the ring that should be compensated by an appropriate linear I_0 monitor.

The optical elements of the monochromator will be:

- plane grating
- spherical mirror
- exit slit

The plane gratings grooves will be lamellar. This tends to give an effective reflectivity (shape reflectivity and material reflectivity) between 10 and 20 %. The main advantage of such a technique is a very low roughness that can be achieved on such surfaces (essentially the same as for ordinary mirrors) thus reducing scattered intensities. The purity of the monochromatic beam is an important factor for XMCD measurements. We are now considering three different gratings between 600 and 2000 lines per mm. The working resolution will be in the range 5000 to 10 000 for the whole energy range from 350 to 2000 eV. The stability of the energy is more important than the absolute energy resolution. The various gratings will be changed easily and the reproducibility of the movements will ensure the conservation of the zero order for the various gratings.

The spherical mirror will be a vertically focusing mirror (VFM2). It will be made of c-Si, with roughness less than 3 \AA rms and coated with a heavy element (Pt, Au, Rh).

The exit slit will be fixed in position, with variable opening by steps of 0.1 to 0.2 μm . Great care will be brought to the reproducibility of the position and opening of this slit.

In order to reduce the size of the image on the sample, a Kirkpatrick-Baez device will be inserted in the beam line. It will be made of one c-Si horizontally focusing mirror HFM3 and one c-Si vertically focusing mirror VFM3. HFM3 and VFM3 roughness and coating will be as for the other optics. The VFM3 mirror will be spherical and the HFM3 will be elliptical to achieve a smaller horizontal image.

We are following two strategies to reject spurious harmonics present in the “monochromatic” flux at low energies (at high energy the horizontally focusing mirror as well as the other optical surfaces should reject the unwanted harmonics). We could install a harmonic rejection device made of two highly reflecting (zerodur or c-Si uncoated mirrors) parallel plane mirrors for which the incidence angles can be varied. This has the disadvantage to deflect the vertical position of the beam when the harmonic rejecter is inserted. This also introduces two reflecting surfaces that reduce the flux. The second strategy is to use various coatings for the VFM2 associated to the monochromator gratings. With appropriate coatings, it is possible to reject unwanted photons. This has the disadvantage to impose one specific grating for a given energy range, though in practice, various gratings for a given energy can be used if the energy resolution is not critical.

One essential point concerns the *thermal stability* of the optical surface. The expected flux on the sample should be above 10^{10} photons per second for a spectral resolution equal $(E/\Delta E)$ to at least 6000 in the whole energy range.

Great care should be brought to the thermal stability at various frequencies: long term thermal drift of the heating load on optical surfaces, short term heat load variation due to phase changes. The temperature around the experiment should be regulated within 1 degree over long periods (several days). In our project, the surfaces are crystalline silicon. By cooling the surface to 150 K, it is possible to reduce to zero the thermal expansion of silicon. In such a case, all the surface deformations due to the photon flux are removed. This contributes to a better stability with time (at the scale of hours) of the mirrors output. The stability concerns the beam intensity as well as the outgoing angles, and then the energy stability for the dispersive optics. The cooling is obtained by cryocooling the surfaces with helium gas at high pressure.

Phonic insulation is also essential: due to the high degree of coherence of the beam, any vibration of the reflecting optical surfaces will transform into variations of intensity. Reducing at maximum the roughness of the optical surfaces can minimize this. The stability of the ground, on which the experiment will be built, should be bent within 1 μm over long periods and should also be independent of variation of weight load in the vicinity of the experiment.

The focused beam on the sample should be around 10 μm vertical and 20 μm horizontal. This requirement is not necessary for most experiments but it proved at ESRF that all experimental set-ups tend one day or the other to be concerned with micrometric samples. The expected Mars Samples are one example among others. We plan to develop a scanning x-ray microscope making use of Bragg-Fresnel zone plates. Such a device will be able to focus the x-rays on a spot smaller than 1 μm^2 with good efficiency. The samples will be mounted on x-y scanning table and images will be recorded by x-ray fluorescence.

The experimental set-ups.

In a first step we have designed different experimental set-ups that should fulfill the needs of the largest part of the community of magneticians using synchrotron radiation. The different types of experimental set-ups have to be versatile for the various types of XMCD envisaged and will be fully computer-controlled: temperature control, magnetic field, detectors and electrometers. The UHV compatibility is necessary for surface study and also for all types of low temperature measurements. An average pressure of 10^{-10} mbar is required. These experimental set-ups are in the vein of those developed by Jean-Paul Kappler and Philippe Saintavit and operating at LURE (The future of these existing equipments, that belong to a pool of several laboratories, remains to be decided).

Each set-up will have a small preparation chamber connected allowing an easy transfer of the sample (load-lock) from air to vacuum. Then, from this chamber, the sample will be transferred to the analysis stage where the sample is cooled and the high magnetic field applied. This preparation UHV chamber will have enough flanges to fix *in situ* sample preparation facilities (ion bombarding, heater stage, e-beam evaporator, Knudsen cells, scrapper, quartz micro-balance). In case of more sophisticated sample preparation and *in situ* sample characterization, an individual chamber with additional degrees of freedom will be connected to the small preparation chamber. Beyond the possibilities offered by this MBE chamber to the user to connect their own facilities, different spectroscopies and diffraction techniques for surface characterization are envisaged (LEED, RHEED, Auger, MOKE), along with a STM (Scanning Tunneling Microscope) apparatus necessary in the case of *in-situ* prepared nanoparticles study. The use of this chamber will be compatible with the synchrotron environment. This chamber should evolve with time and try to fulfill the changing needs of users. Parallel to these features an UHV portable chamber compatible with other deposition sources or analysis chambers is considered.

1-Dilution refrigerator (50 to 1000 mK) and superconducting coil (7 T)

The first set-up is a dilution $^3\text{He}/^4\text{He}$ refrigerator to cool down the sample at very low temperature between 50 and 1000 mK. The device will have a cryogenic power of 50 μW . This is enough to get rid of the heating produced by a powerful photon flux. The sample will be in a UHV environment (in the 10^{-10} mbar). The sample will be put in position from an introduction chamber without breaking the vacuum of the main experimental chamber. Since such a dilution refrigerator will work in UHV, there will be no exchange gas to cool to 4 K the various parts of the cryostat. A special attention will be paid to this by fast cooling produced by an extra circuitry. Cooling of the whole sample holder from 300 K to a working temperature below 1000 mK will take less than 6 hours. A superconducting split coil will produce the magnetic field. The maximum field will be 7 T and the inductance of the coil will be minimized to allow fast reversing of the field. The possible perturbations induced by the large magnetic field on the storage ring will have to be investigated in order to be minimized. Foucault currents heating the sample during field reversal will have to be minimized. The detection systems will depend on the type of samples:

- total electron yield photocurrent measured by high precision electrometer,
- silicon diodes working in current mode (or photovoltaic mode) coupled to low noise electrometers,

- ° energy resolved multi-element detector, such as Ge crystals working in a counting mode.

1bis-Pumped ^4He cryostat (1 to 300 K) and Superconducting coil (7 T)

By simply changing the insert inside the previous disposal, it will be possible to work at higher temperature between 1 and 300 K. Cooling will be produced by pumping on a ^4He bath for temperatures between 1 and 4.2 K and by heating He gas for temperature between 4.2 and 300 K. The same magnetic field and detection systems will be used.

2- ^4He cryostat (10 to 300 K), oven (300 to 1000 K) and electromagnet (2 T)

In many cases, it is essential to have a fast inverting magnetic field. This is best achieved with an electromagnet producing 2 T. For such devices the field can be inverted at the rate of 10 Hz. The space between the poles of an electromagnet is too small to accommodate a UHV vessel containing all the thermal screens necessary to reach the lowest temperatures. Then a good compromise is a regular compact ^4He cryostat, cooling the sample between 10 K and 300 K. Higher temperatures will be obtained by a oven to reach moderate temperature (up to 1000 K) that are usually enough to explore the magnetic behavior close to the Curie temperatures of most compounds.

With an electromagnet, it is possible to reverse the magnetic field at each energy point of a scan. This procedure is essential to measure very small XMCD signals.

3- ^4He cryostat (10 to 300 K), oven (300 to 1000 K) and quadrupole electromagnet (2 T)

The previous disposal can be upgraded into a quadrupole electromagnet for which the magnetic field in the horizontal plane will be allowed to point in any direction. This type of device is essential to measure XMLD on any type of magnetic compounds: ferromagnetic, ferrimagnetic or even antiferromagnetic samples. For single crystals, turning the sample is not equivalent to turning the magnetic field or turning the polarization.

4 - Specular and off-specular reflectivity.

The set up is under development at the *Laboratoire de Cristallographie de Grenoble*. It includes quadrupole electromagnetic setting where the field (0.2 T) can be rotated in the sample plane over 2π . One sample holder has been designed for low temperatures measurements and another, in collaboration with Michel Belakhovsky (CENG) and co-workers, for high temperatures measurements.

5 - X-ray microscope.

This is an x-ray microscope working with a scanning stage. The x-ray beam will be focused by a Fresnel zone plate. With today technology it is possible to manufacture zone plates giving spatial resolution of 0.1 μm . The efficiency at the energies of the beamline will not be larger than 20 %. Due to focal distance, signals for the two photon helicities will be recorded in a fluorescence detection mode. The mapping will be made by moving the sample mounted on an x-y scanning table. We expect that improvements in the technology of zone plates conception will allow in the future to increase the efficiency and also the spatial resolution of such devices. This assembly will be devoted for instance to the study of single domain nanoparticles.

IV – National Context

The project of this beam line answers an already large demand by French users. In LURE, two beam lines are presently serving the request by French, and European scientists in the field of XMCD. The x-ray source is an asymmetric wiggler designed by José Goulon. The two beam lines are SU22 for energies between 850 eV and 2 keV ($M_{4,5}$ edges of rare earths) and SU23 for energies between 200 eV and 1 keV ($L_{2,3}$ edges of transition elements). Moreover, an international collaboration with ELETTRA (5 years convention) has been settled to benefit from a 3rd generation source using the cryostat developed at LURE for very low temperatures in the hundreds of mK range. At the European Synchrotron Radiation Facility (ESRF) the ID12 and ID8 beam line (formerly ID12B) have promoted the development of XMCD measurements on 3rd synchrotron beam lines. The use of Helios and Apple II undulators coupled to highly linear detectors has allowed the French community to acquire XMCD data of an unprecedented quality. They have also promoted theoretical achievement in XMCD, XMLD and gyrotical spectroscopy. Nevertheless, the French beam time request is far from being fully satisfied: only one fourth of the projects deposited on SU23 are accepted. Such a strong pressure is also felt on the ESRF ID8 beam line dedicated to measurements at low energy: 26 projects were rejected out of 35 for the first semester of 2001. Consequently, the French demand is large and far from being fully satisfied by the today available beam time. The necessity for a beam line devoted to XMCD is then fully acknowledged. Moreover the French community using XMCD in the soft x-ray energy range is well distributed throughout France as illustrated by the second paragraph:

- **Centre d'Études Nucléaires de Grenoble (CENG)** through different collaborations (among others the Daresbury Laboratory) they are using magnetic scattering techniques to investigate patterned and hybrid magnetic materials.

- **Laboratoire de Cristallographie de Grenoble** is also concerned by XMCD measurements on rare earth-3d intermetallic compounds. In that laboratory, J.M. Tonnerre and co-workers are strongly involved in the development of related techniques based on reflectivity and diffraction.

- **Laboratoire Louis Néel (Grenoble)**. Several teams are using synchrotron light to study the magnetic properties of rare earth-3d intermetallic compounds (alloys, thin films, nanostructures). The group of Alain Fontaine has developed the technique of time resolved XMCD to measure the magnetization dynamics of thin films and complex structures at nanosecond timescale.

- **Laboratoire de Physique des Matériaux de Lyon**. People from the "clusters" group (part of an European Network) are current users at LURE and at the ESRF of Anomalous diffraction, EXAFS, XPS and XMCD beam lines. They work on an original synthesis technique of magnetic nanogranular films from Low Energy Clusters Beam Deposition, thin film protected or transferred under UHV conditions into the analysis chambers (as on the SA 32 beam line at LURE).

- **Laboratoire de Physique des Matériaux de Nancy**. several teams are current users at LURE as well as at ESRF of XMCD beam lines. They have developed a special knowledge of measurements done on samples synthesized in on-site MBE chambers and transferred under UHV conditions into the XMCD analysis chamber.

- **Laboratoire d'Utilisation du Rayonnement Electromagnétique** (Orsay): several teams are using the magnetic dichroism on a wide range of sample type. Different international collaborations have been settled, as for instance the already mentioned collaboration with ELETTRA.

- **Laboratoire de Minéralogie et Cristallographie de Paris**. Philippe Saintavrit and co-workers have developed experiments available on the two LURE XMCD beam lines. Theoretical achievements in the field of magneto-optic and chiral spectroscopy are fully used by the community.

- **Unité mixte de Physique CNRS/THALES** has developed an experimental program aimed at determining the magnetic properties of various interfaces designed for spin-electronics. This research program is strongly connected to XMCD measurements and coupled with other laboratories in charge of synthesis (Laboratoire de Minéralogie et Cristallographie de Paris for magnetic semi-conductors).

- **Laboratoire de Chimie Inorganique et Matériaux Moléculaires**. Michel Verdaguer and Dante Gatteschi (permanent address in Firenze) have explored several aspects of XMCD in molecular based magnets, high spin molecules, photosensitive magnets and chiral magnets. They are strongly engaged with LURE, LMCP and ESRF in various collaborations where chemists and physicists join to apply XMCD to chemical magnets.

- **Institut de Physique et Chimie de Strasbourg** (IPCMS): several laboratories are deeply involved in research program making a large part to XMCD and magneto-optical spectroscopies. Instrumental developments lead by Jean-Paul Kappler have also been essential for the performances of the two XMCD beam lines of LURE.

V – Conclusion

This APS evidences the *very large French scientific community* that is contributing to the development of X-ray dichroic spectroscopies and the *wide research themes involved* going from technological application to paleomagnetism. This is illustrated by the strong pressure over the XMCD beam lines already existing (ratio ~1:4). We would also point out that this pressure will increase with the cessation of beam delivery by LURE (fall 2003). All these elements strongly support the need for a *beam line fully dedicated to dichroism* in order to serve the scientific community of magnetism in the soft x-ray range. Moreover, the thematics approached will favor the insertion of the synchrotron community inside the scientific network of Orsay, in particular through the programs MINERVE (Microtechnologie, Nanoscience, Enseignement, Recherche, Valorisation, Entreprise) and ISARD (Interface et Structures Artificielles Réduites en Dimension) that are jointly pushed by several laboratories working on the physics of small particles.

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Annex B – Human capital

Two scientists are necessary from the start to manage the final conception, the realization and the assembling of such a beam line. A mechanical engineer and an electro-computing engineer should help them.

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