Optics and instrumentation upgrades for hard x-ray magnetic spectroscopy research at beamline 4-ID-D

This proposal seeks to upgrade the source, optics and infrastructure for beamline 4-ID-D, which is designed for hard x-ray spectroscopy experiments that utilize circularly polarized x-rays. These enhancements will produce a dedicated facility for cutting-edge hard x-ray magnetic spectroscopy research on novel magnetic systems and for materials under extreme conditions (high pressure and magnetic field). The primary components of this proposal include the installation of an arbitrary polarization insertion device, expansion of the experimental station, addition of a control area enclosure, and new instruments for high-field and high-pressure XMCD and magnetic reflectivity studies.

Scientific Justification

Discovery of novel electronic and magnetic materials together with a fundamental understanding of their properties is crucial in order to enable future technological advances. The tunable, circularly polarized hard x-ray source (5-12 keV) at beamline 4-ID-D, which couples a linear undulator with phase-retarding crystal optics, has provided invaluable atomic-level insight into the electronic interactions of a variety of complex functional magnetic materials such as super-strong permanent magnets and magnetocaloric materials. Novel magnetic behavior displayed by emerging materials, however, has raised new sets of fundamental questions in magnetism which cannot be properly addressed with the current beamline's capability: *Does ferromagnetism coexist with superconductivity in rutheno-cuprates? What is the origin of the novel magnetism found in 4d and 5d metal nanoparticles? How does the capping layer influence the magnetic anisotropy of 3d-4d metallic films? How does compressive strain affect the magnetism of half-metallic films? How does pressure-induced hybridization affect the indirect exchange coupling in ferromagnetic insulators?* Answering these questions requires (1) the ability to probe the itinerant magnetism of 4*d* (e.g. Ru, Rh, Pd, Ag, Cd) and 5*d* (e.g. Os, Ir, Pt, Au) systems, and (2) the ability to study magnetic materials under extreme pressure (1 Mbar) and magnetic field (10 T) conditions, using the element- and site-specific XMCD-based absorption and resonant-scattering probes already in place at 4-ID-D. This upgrade plan is designed to meet these future challenges.

Added Value of the Mid-term Upgrade

The primary goal of this proposal is to provide greater flux and expanded energy range for circularly polarized (CP) x-ray spectroscopy experiments in the energy range between 2.4 and 30 keV at the APS. Furthermore, it seeks to develop enhanced sample environment capabilities (magnetic field, electric field, and high pressure) for such studies.

- *Undulator*

The key feature of this proposal is the installation of a helical insertion device (Apple-II) for generating CP x-rays directly at the source. Currently phase retarding optics are used to provide CP capabilities on this beamline. While these optics work very well their use is limited for energies below ~ 4 keV and above ~10 keV due to the high attenuation and low Bragg angles, respectively. Expanding CP capabilities into these energy regimes will enable XMCD-based studies at the $L_{2,3}$ absorption edges of 4*d* and 5*d* elements. Furthermore, this ID upgrade will not only enable new science at lower and higher energies but will yield significant gains in flux (x 10-100) and reliability across all relevant energy ranges. Phase-retarder optics will remain at the beamline for cases where fast helicity-switching coupled with lock-in detection is desirable. Since magnetic resonance enhancements in the 2.4-4 keV range are very sizable (10-50 %) the fast switching of helicity required for lock-in detection is not as critical in this energy range. A longer term upgrade plan, however, which is tied to a similar upgrade of the intermediate energy branch 4-ID-C sharing the straight section with 4-ID-D, includes the installation of two Apple-II devices in a dedicated 8-m-long straight section to enable fast helicity switching through particle beam steering amongst the devices, each preset to generate either R- or L- CP x-rays throughout the whole energy range covered by the device.

- *Improved focusing and stability*

The upgraded focusing optics will allow XMCD-based experiments under extreme conditions of pressure (1 Mbar). This presents a fantastic opportunity in a rapidly expanding area of research, which is bound to have a large impact in our understanding of complex magnetic materials. The current focusing capability (150 x 250 μm^2) has permitted pressures of up to 0.2 Mbar to be reached before flux limitations due to beam slitting coupled with attenuation in phase-retarding optics render experiments unfeasible. Reaching 1 Mbar requires a ~10 μm^2 beam due to the related small sample volumes. Our plan is to upgrade the toroidal mirror in 4-ID-B to create a virtual source before the entrance to 4-ID-D and achieve the required demagnification with 20-cm-long, differentially-deposited K-B mirrors. Improved beam stability for these high-pressure studies will be achieved by adding an additional PZT stage to the second crystal of the monochromator to dynamically adjust mono-beam position also in the horizontal direction.

- *Instrumentation to enhance sample environments*

Currently XMCD studies on 4-ID-D are limited to applied fields of 4 Tesla at ambient pressure and 0.6 Tesla with applied pressure. We propose to replace the current magnets on 4-ID-D with a large-bore 10 T magnet for absorption-based XMCD spectroscopy. The large-bore will enable insertion of a membrane-driven diamond anvil cell into the magnet’s bore for high-pressure (1 Mbar) studies of magnetism at high fields and low temperatures. The large bore will also allow implementation of fiber-optic coupling to the DAC for *in-situ* pressure calibration using Ruby fluorescence. In addition, *in-situ* abilities to apply electric fields and measure electrical conductivity will be implemented to enable studies of novel magnetoelectric materials such as multiferroics and colossal magneto resistance manganites. Furthermore, the fiber optic feedtrough will enable studies of optically-induced magnetism; *e.g.*, in molecular magnets. An in-vacuum multi-element Si drift diode detector will also be installed to improve the fluorescence count rates of a factor of 5–10.

We also propose to install an additional two-axis 2 Tesla superconducting magnet to be mounted on a 4-circle diffractometer for magnetic-DAFS (site-specific magnetism in crystals and epitaxial films) and magnetic-reflectivity (interfacial magnetism in nanostructures) studies using techniques developed at Sector 4. The two-axis magnet allows application of the magnetic field in- and out- of the scattering plane permitting measurements of orthogonal components of magnetization density with element- and site/interfacial- specificity.

- *Experimental station expansion and control area enclosure*

The current size of the 4-ID-D experimental station was constrained by the original design of the 4-ID-C soft x-ray beamline. Since construction, however, the layout of the experimental chambers on 4-ID-C has been modified which has reduced the length that this beamline extends downstream. To take advantage of this, we propose to extend the 4-ID-D experimental station upstream, so that it can permanently accommodate instruments for magnetic reflectivity and XMCD studies at high-fields and high-pressures. At the same time we would construct an enclosure for an experimental control area, which would accommodate users of both the 4-ID-C and 4-ID-D beamlines. The current environment on the experimental hall floor has significant ambient noise from a number sources. A control station for the users of each of these beamlines would create a much more pleasant working space from which to conduct an experiment.

Expected user communities

In addition to the current user base of 4-ID-D, the expanded capabilities to study 4*d* and 5*d* systems is expected to nucleate two distinct groups of new users: those in the nanosciences working at the interface between chemistry and materials science where manipulation of optical/electrical/magnetic properties of materials at the nanoscale is taking place (e.g. novel magnetism in Pd, Cd, and Au-based nanomaterials); and those researchers of fundamental aspects of magnetism in 4*d* and 5*d* systems (e.g. ruthenate superconductivity and weak ferromagnetism in iridate and osmate materials). Preliminary tour-de-force experiments carried out at 4-ID-D in the 3.1-4.0 keV range (Pd, Cd, Ag, In) under far-from-optimal conditions showed great potential and a large user interest.

Studies of magnetism at high-pressure is a rapidly expanding area of research. High-brilliance, hard CP x-rays are key enablers of unique element- and site-specific XMCD-based probes. The current program at 4-ID-D (0.2 Mbar) has attracted 8 user groups in the very short 2 year span since the program's inception and we expect significant growth in this area. Beamline 4-ID-D already has sizable user communities in XMCD-based spectroscopies and the proposed magnets and detector upgrades will benefit the entire community.

Enabling technologies and infrastructure

- Apple-II type insertion device to replace linear undulator.
- New mirrors for enhanced focusing to enable high pressure experiments
- Expansion of the 4-ID-D end station.
- New monochromator to reduce depolarization effects in 2.5-3.5 keV range

- 10 T superconducting magnet for XMCD, 1-axis.
- 2T superconducting magnet for reflectivity, 2-axis.
- Multi-element Si-drift diode in-vacuum detector.

Partnerships and user interest

Dr. Valentin Iota, Lawrence Livermore National Laboratory.
Prof. Vince Harris, Northeastern University
Prof. Rosa Lukaszew, College of William and Mary
Prof. Diandra-Leslie Pelecky, University of Texas at Dallas.
Prof. Israel Felner, The Hebrew University, Israel
Prof. Jesus Blanco, Universidad de Oviedo, Spain
Prof. M. Luisa Fdez-Gubieda, Universidad del Pais vasco, Spain
Prof. Joaquin Garcia, Universidad de Zaragoza, Spain.
Prof. V. K. Pecharsky, Iowa State University.
Prof. K. A. Gschneidner, Iowa State University.
Dr. Ho-Kwang Mao, Carnegie Institution of Washington.
Dr. Yang Ding, Carnegie Institution of Washington
Dr. Anton Stampfl, ANSTO, Australia.
Dr. Flavio Garcia, LNLS Brazilian Synchrotron Laboratory, Brazil.
Prof. Gang Cao, University of Kentucky.
Dr. Vemuru Krishnamurthy, Oak Ridge National Laboratory.

Budgetary profile

Apple-II type insertion device		$1200k
Monochromator modifications		
Additional PZT stage for horizontal feedback	$ 50k	
Additional crystal for low E operation	$ 100k	
		$ 150k
Focusing mirror replacement		$ 300k
4-ID-D experimental station expansion		$ 300k
Control area enclosure		$ 150k
10 Tesla XMCD magnet		$ 400k
2 Tesla Reflectivity magnet		$ 250k
Multi-element energy dispersive detector		$ 250k
Total:		**$3000k**