Argonne NATIONAL LABORATORY | Advanced Photon Source

Strategic Renewal of the Advanced Photon Source

Proposal for Approval to Proceed with Conceptual Design (CD-0)

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ABSTRACT

This document proposes a coordinated upgrade of the accelerator, beamlines, and enabling technical infrastructure that will equip future users of the Advanced Photon Source (APS) to address key scientific challenges underpinning the need for sustainable energy, healthier living, and economic security. Today the APS is the preeminent hard x-ray source in the US and has made major contributions to these needs. The recent "New Era" report from the Department of Energy (DOE) (BESAC 2008) argues that transformational approaches to solving societal problems in energy and environment require sharpened scientific tools to observe, understand, and ultimately control materials function on the nanoscale. Our vision for APS would deliver on this through two themes that reflect the scientific power of high-energy x-rays: *mastering hierarchical structures through imaging* and understanding *real materials under real conditions in real time*. The proposal explains how these themes address key scientific problems and elucidates how they will be practically addressed in the technical upgrades. The renewed APS will greatly strengthen capabilities in high-energy x-rays and complement other light source developments in the US and worldwide. Not to follow through with this vision could damage the US arsenal of scientific tools, hampering progress and ceding leadership to other nations. The cost for the renewal is estimated at $350M, with a time scale of 6 years from start to finish.

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1. INTRODUCTION

This proposal requests a major renewal and upgrade for all elements of the Advanced Photon Source (APS), from accelerator through beamlines to data analysis tools, enabling an expanded user community to better address the scientific challenges of sustainable energy, healthier environment, improved human health, and economic competitiveness.

The APS is one of three high-energy (6-8 GeV) dedicated third-generation light sources operating around the world. The largest and most productive user community in the US, APS users publish well over 1000 refereed journal articles every year, with the percentage of "high impact" papers now comparable with the best worldwide. Amongst the most significant scientific highlights from the first 13 years of APS operation are:

- The most productive source of protein structures in the world, including many theraputically important G-protein receptors, begininng with rhodopsin (Palczewski et al. 2000) which has 2500 citations;
- A powerhouse of research on materials under high pressure, including the first evidence that inorganic components can form methane under geological conditions (Scott et al. 2004) [APS users produce more high impact papers in high-pressure science than any other facility worldwide];
- Pioneering development of techniques now widely used, especially x-ray photon correlation spectroscopy and nuclear resonant inelastic scattering;
- The first images of the internal workings of opaque living insects and fuel sprays;
- Development of the Multi-Layer Laue zone plate, which offers the best route to nanofocusing x-rays; and
- The first major user facility to develop and implement the top-up mode of operation for routine use, now emulated worldwide.

Since opening 13 years ago as the first dedicated high-energy synchrotron light source in the US, APS has continued improving and expanding operations. Around the world many new ~3 GeV third-generation machines are benefitting from the technology developed at APS, such as top-up operation and x-ray nanofocusing optics. National Synchrotron Light Source II (NSLS-II), under construction at Brookhaven National Laboratory, represents a pinnacle of performance for a 3 GeV machine.

One indication of the importance of APS to energy research is that almost 1/3 of the 46 Energy Frontier Research Centers across the nation identify work at APS as a major component of their proposed research.

In this changing landscape, APS remains unique as a 7 GeV source, and we expect increasing concentration on higher energy "hard" x-rays (E≥20 keV). These penetrating rays are a key weapon in the arsenal needed for the Grand Challenges facing the new "Control Age" (BESAC 2007), fulfilling a vision that mankind will develop materials and machines to satisfy our need for sustainable energy, healthy lives, and a thriving economy. Through targeted improvements to the machine, x-ray instrumentation, and detectors, the renewal will deliver more than an order-of-magnitude higher measured flux to the majority of experimenters in APS end stations and will increase the user capacity of the most demanded techniques. The APS renewal is a top priority for Argonne National Laboratory (ANL). ANL will continue to promote and grow materials and computation research which will accelerate the success of a renewed APS.

In extensive dialog, guided by our Scientific Advisory Committee, with our user community, staff, and scientific leaders, we have zeroed in on two compelling scientific themes that provide the focus for technical choices in the renewal. These themes were chosen because they encapsulate the unique strengths of high-energy x-ray radiation, and they point the way

to developing capabilities that will solve the real problems facing the world today. The renewal of APS will also strengthen the hard x-ray community in the US as it considers the potential for revolutionary new capabilities in high-energy x-ray sources to complement third-generation capabilities (USDOELightSources 2009, BESAC 2009).

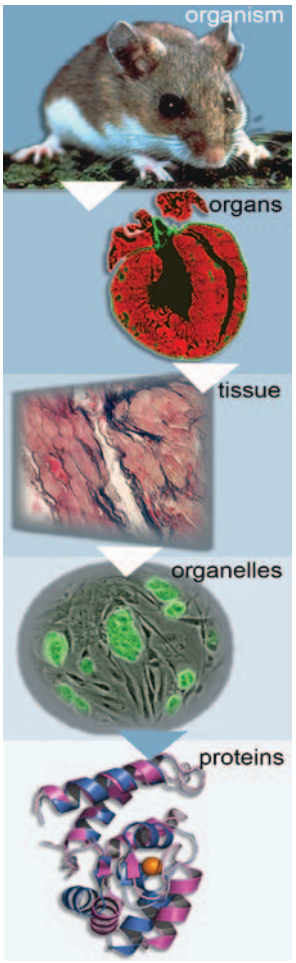


Fig. 1.1. *The hierarchy of structural scales from proteins to organisms, illustrated for a mouse.*

MASTERING HIERARCHICAL STRUCTURES THROUGH IMAGING

Scientifically, a hierarchy describes complex interrelated multi-level structures whose function relies on interactions between levels. The concept encapsulates the complexity of life, whose hierarchy includes organisms, cells, organelles, and proteins (fig. 1.1). The grand challenge of understanding life is identifying how proteins, whose structures are encoded in genes, determine organism function. Solving this challenge would unlock the secrets of health and disease. Protein structure is already the domain of synchrotron radiation, but the grand challenge demands observing structure on many hierarchical length scales in concert, a unique capability of high-energy x-rays. Images can reveal three-dimensional structure in macroscopic objects up to centimeters in size, yet probe in real space on length scales approaching 1 nm, and through scattering and spectroscopy, reveal structure, chemistry, and dynamics on the atomic scale (0.1 nm). In parallel with the hierarchy of structure is a hierarchy of time scales. Improvements in source properties, coherence, optics, and detectors offer dramatic improvements that can push temporal resolution towards the picosecond time scale.

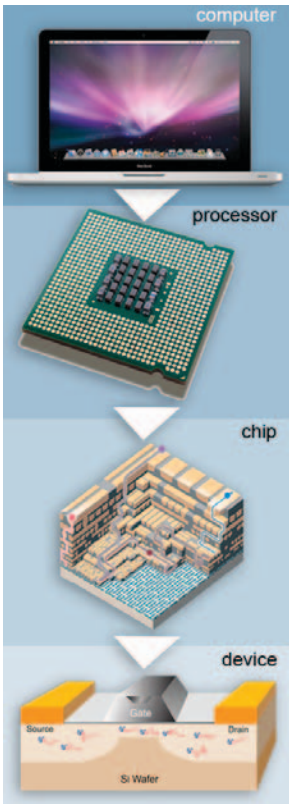


Fig. 1.2. *The hierarchy of length scales needed for a functioning computer chip ranges from the atomic scale of device interfaces (bottom) up to the centimeter scale of assembled packages.*

Wood and switchgrass are just two among thousands of examples of ligno-cellulose. This naturally nano-structured composite offers a unique route to renewable biofuels. Using genetic engineering we could optimize the structure of the composite and of enzymes or catalysts that would break cellulose into alcohol with a carbon footprint only 10% of commercial alcohol fuels today. Even the inorganic structures made by life, such as the abalone shell, are hierarchical, with astounding but ill-understood mechanical properties that combine low weight with high strength and toughness. We would like to imitate these materials, using nanoscale control of processing and synthesis, but to do so we need to understand how the materials work on all levels.

Stuff imitates life, and not surprisingly hierarchical structures are ubiquitous in machines and useful materials. The silicon integrated circuit (fig. 1.2), magnetic hard disk, and polymer composite car bumper are a few examples where improved x-ray imaging offers understanding and control.

The theme of control in hierarchical structures reflects another of the five Grand Challenges from the BESAC report, namely, “How can we master energy and information on the nanoscale to create new technologies with capabilities rivaling those of living things?” New capabilities planned for the APS renewal would transform imaging, allowing objects three times larger to be imaged, improving resolution several times over, increasing sensitivity more than an order of magnitude, and doubling available beamtime.

REAL MATERIALS UNDER REAL CONDITIONS IN REAL TIME

The conditions under which we manufacture most real materials are not at equilibrium. Another of the five grand challenges from the BESAC report is, "How do we characterize and control matter away – especially very far away – from equilibrium?" High-energy x-rays provide powerful tools to answer this question, with capabilities that will be dramatically enhanced through the APS renewal.

Catalysis in Chemical Production

Fig. 1.3. *Chemical production, such as in this picture of a benzene plant, relies on catalysts to control and accelerate reactions. These materials, often in the form of nano-dispersed particles, operate in a complex environment (inset) that can be studied with hard x-rays.*

Catalysts control over 70% of industrial chemical reactions, directly determine efficiency of energy and natural raw material needs, and determine the nature of unwanted effluent (fig. 1.3). A recently awarded DOE Energy Frontier Research Center (EFRC) at Argonne for Atom-efficient Chemical Transformations will use a multidisciplinary approach to address key catalytic conversions and improve the efficiency of producing fuels from coal and biomass. High-energy x-ray studies at APS allow the observation of *in-situ* processes, identifying structure, chemistry, and dynamics to complement basic studies on model systems, where more precise techniques may be used but behavior is often different. The ultimate goal is to develop computer simulations based on *in-situ* measurements that will permit predictive modeling and design of catalysts and other functional materials. Similar approaches are needed for battery (another Argonne led EFRC) and fuel cell development (fig. 1.4). Extreme conditions, such as occur inside planets, are important as a tool to probe new materials with properties beyond those available to us from known materials. In many cases, once these properties are known and understood, they can be expressed in functional materials. While APS is already amongst the most productive sources for this kind of research, the pool of important applications and users is large, and will be tapped by new experimental capabilities planned for the renewal. Through the APS renewal, users will be able to have access to significantly higher pressures, temperatures, and chemical activities and to shorter time scales in their studies of real materials under real conditions in real time.

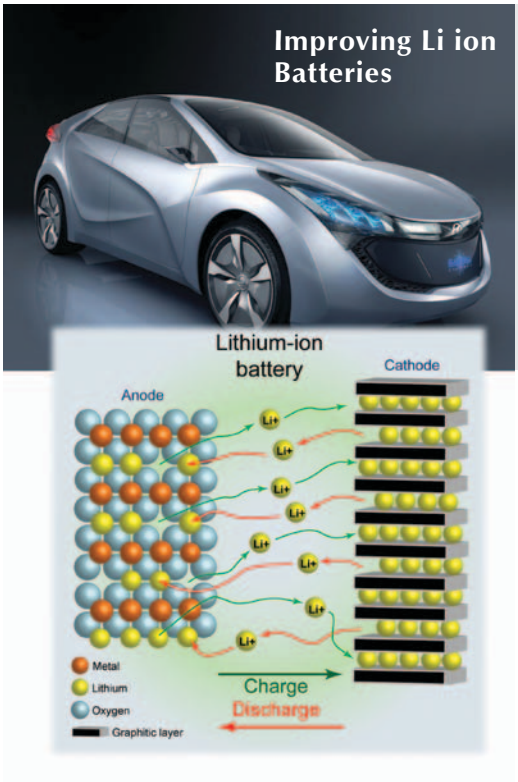


Fig. 1.4. *The complex process of charge transfer and storage in batteries limits performance. Improvements are expected by observing the electrochemistry in action with hard x-rays.*

These two compelling scientific themes for the renewal of APS hard x-ray science emerged from an extensive process of workshops and proposals from our user community. The process culminated in October 2008 with a strategic planning meeting in which we invited groups of experts representing 10 science areas to identify their scientific needs from a renewed APS (table 1.1).

Table 1.1: Science Areas from October 2008 Renewal Workshop

Chemical Science & Engineering
Condensed Matter and Materials Physics
Engineering Applications & Applied Research
Fundamental Interactions in Chemical, Atomic, & Molecular Physics
Geological, Environmental, & Planetary Sciences
Interfacial Science
Life Sciences (excluding MX)
Macromolecular Crystallography (MX)
Materials Science & Technology
Polymers and Soft Materials

The APS Scientific Advisory Committee helped identify the science themes. Using the science themes to select and optimize from the many dozens of technical proposals for possible machine and beamline upgrades, we developed the vision for the renewal described here (for more details on workshops and technical proposals, visit our website www.aps.anl.gov/Renewal/).

The renewal will incorporate many outstanding technical innovations into the APS, including

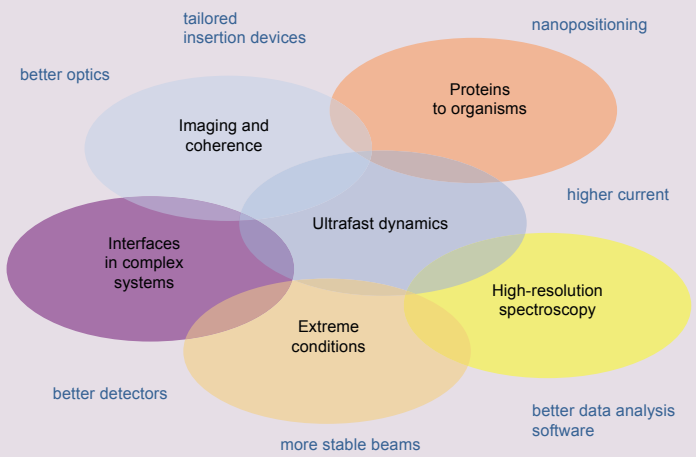
- Improvements to the accelerator and beamlines (e.g., canted undulators) that potentially double the number of simultaneous experiments.
- Incorporation of state-of-the-art hard x-ray focusing optics into beamline designs.
- Incorporation of greatly improved detectors for high resolution imaging, scattering, and spectroscopy in real time.
- Two orders of magnitude reduction in the source pulse width for experiments needing picosecond time resolution.
- Optimized high-brilliance hard x-ray undulator designs, which in many cases will lead to more than an order of magnitude improvement in flux on the sample.

The APS renewal is an opportunity to bring these transformational developments in hard x-ray technology to bear upon the scientific challenges of the nation. We have carefully planned to accomplish upgrades without machine shutdowns beyond our normal maintenance periods. A small number of beamlines must be moved, but we believe we can accomplish this in a minimally disruptive fashion by appropriately utilizing our open beamports.

The scientific and technical drivers for our choices are expanded in the next section on "Science Drivers for the Renewal." To present the scientific vision in this proposal, we have classified the most important beamline instrumentation developments into six groups, shown as ovals in fig. 1.5. These beamline capabilities are all greatly enhanced by accelerator (section 3) and enabling technical improvements (section 5). Necessary R&D is described in section 6. The pre-conceptual estimates for the project cost and schedule are contained in section 8.

Fig. 1.5. The ovals show beamline categories, reflected in sections 2 (scientific drivers) and 4 (beamline improvements in response). Those accelerator and enabling capabilities described in sections 3 and 5, supporting all of the beamline categories, are identified in the background of the figure.

Conceptual diagram of the components for the APS renewal



INSTITUTIONAL, NATIONAL, AND INTERNATIONAL CONTEXT

The APS currently has 61 operating beamlines, serving almost 3500 unique users per year (fig. 1.6). Of the 30 straight sections that can be instrumented, only four are not currently occupied. One of these vacant straights is committed to the Intermediate Energy X-ray (IEX) beamline, funded by the National Science Foundation (NSF) and DOE. The IEX beamline, specializing in high-energy photoemission and resonant scattering, was proposed by a Collaborative Development Team (CDT). CDT teams couple external scientific leaders with APS staff. Irrespective of the source of funding for construction, a CDT beamline is constructed and operated by APS. The CDT model has been successful in recent years – Inelastic X-ray Scattering (30-ID) and Powder Diffraction (11-BM) beamlines are examples – and effectively couples external scientists and their ideas to facility beamlines. Several of the proposed new beamlines in the renewal have been developed through this route.

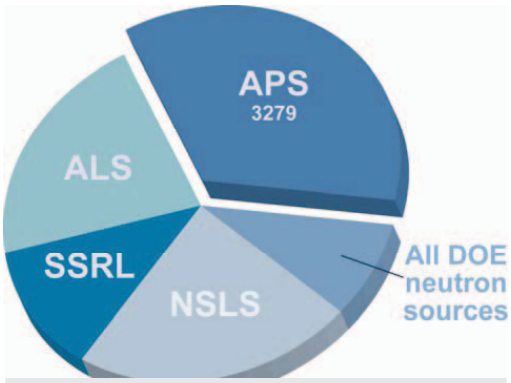


Fig. 1.6 The number of APS unique users in FY2008 compared with other US DOE light sources and neutron sources.

Today APS operates almost half of the 61 operating beamlines. Non-APS operated beamlines are Collaborative Access Teams (CATs), which construct and operate beamlines independently while making at least 25% beamtime available for general users. The majority of our CATs carry out macromolecular crystallography and are funded by the National Institutes of Health (NIH). In the renewal planning, we have worked hard with our Partner User Council that represents CATs to ensure that our vision encompasses both facility and CAT beamlines.

In the renewal we plan to build out the three remaining sectors and significantly renovate many other beamlines. We plan, for example, to install approximately 30 new insertion devices. All beamlines at APS will benefit from the accelerator and enabling technical capability developments, and we have included the cost for necessary x-ray delivery optics upgrades for all beamlines. Many of the facility-operated beamlines are former CATs, and the renewal provides an opportunity to significantly improve performance by optimizing beamlines that were originally designed as multi-purpose.

Argonne National Laboratory is home to several other user facilities that will leverage the renewal investment. The Center for Nanoscale Materials (CNM) is a close partner with APS, jointly operating the nanoprobe beamline (26-ID). Our success in nanoprobe developments has resulted from close collaboration with CNM. CNM is a stand-alone user facility with state-of-the-art tools for making, modeling, and characterizing nanostructures, and the shared user community is growing. The renewal will undoubtedly stimulate further growth of the nanoscience user community, benefitting both facilities. The Electron Microscopy Center (EMC) offers and develops state-of-the-art electron microscopy instrumentation that is of mounting interest to APS users. Our vision for *correlative microscopy* (Sect 2.1) involves expanding complementary microscopy capabilities, and we share with the EMC a vision for the growing importance of imaging at Argonne. Finally, the Argonne Leadership Computing Facility (ALCF) offers a national resource for high-end computation. This facility and the expertise of its staff are a tremendous asset for our efforts to develop next-generation data analysis and modeling software.

In our strategic planning we have been keenly aware of the role of APS within a regional, national, and international context. Many of our users have used other sources, and our staff has been deeply involved in the worldwide collaborative development of new sources. In addition, we have made an effort to include other synchrotron directors on our Scientific Advisory Committee (SAC). Today we have the current or former directors of the European Synchrotron Radiation Facility, the Photon Factory, and the Advanced Light Source as SAC members.

Detailed comparisons between source parameters of APS and other US and international sources can be found in the proceedings of the APS Renewal Workshop and APS Renewal White Paper [(APS 2009) and *www.aps.anl.gov/Renewal/*], and also in the appendices of two new reports on next-generation light source possibilities (USDOELightSources 2009, BESAC 2009). APS is expected to remain the only high-energy third-generation light source in the US at least for the next decade if not much longer, and as shown in fig. 3.2.1, APS will remain much brighter than NSLS-II above 20 keV. Thus we see NSLS-II, with its lower emittance and higher brightness at 10 keV, and a renewed APS as complementary state-of-the-art tools.

Free electron lasers are very exciting for future x-ray science, with the Linac Coherent Light Source (LCLS) at Stanford recently demonstrating lasing at 8 keV photon energy. We are proud to have built the LCLS undulator system. The x-ray science community understands that such fourth-generation sources address different experiments than do third-generation storage ring sources like APS. Our proposed Short Pulse X-ray (SPX) source is a complementary development for the ultrafast science community. In the future, APS would like to lead the US high-energy x-ray community as they explore next-generation possibilities beyond LCLS in the high-energy region (BESAC 2009). While beginning ANL-supported R&D on long-term future sources (Kim et al. 2008), we see the renewal of APS as the most important step for the nation in advancing the use of hard x-ray science and identifying and leading, as appropriate, needs for future sources.

The importance of a renewed APS to the worldwide effort on sustainable energy and improved health and wealth are recognized in the conclusions of the Grand Challenge report (BESAC 2007), which identifies the need for “instruments more precise and flexible than those used up to now for observational science” as one of three prongs of attack to solve the problems, and specifically notes that the DOE’s light sources “represent an extraordinary success story, enabling an enormous range of science across all the disciplines mentioned in this report. Maintaining and upgrading of this resource base is clearly critical to the broad success of Grand Challenge Science.” Similar observations, more specific to the APS, were made in the reports of the National Research Council decadal study on Condensed Matter and Materials Physics (NRC 2007) and a draft version of an upcoming report by the American Physical Society on utilization of US and international x-ray and neutron facilities.

2. SCIENCE DRIVERS FOR THE RENEWAL

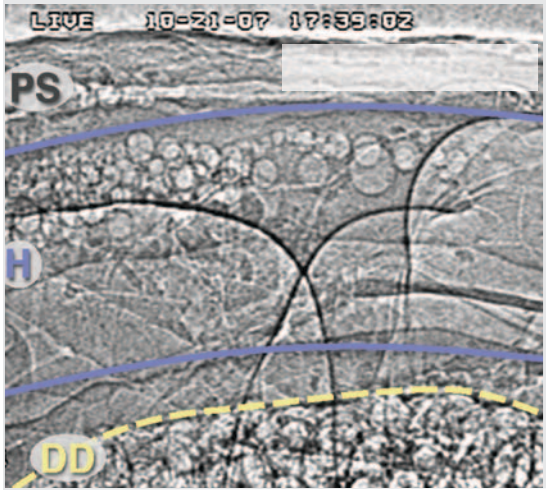
2.1 IMAGING AND COHERENCE

Roger Tsien, winner of the 2008 Nobel Prize in Chemistry for his discovery of green fluorescent protein labeling, stated in an opinion piece entitled "Imagining imaging's future" (Tsien 2003) that "Imaging specific molecules and their interactions in space and time will be essential to understand how genomes create cells, how cells constitute organisms and how errant cells cause disease. Molecular imaging must be extended and applied from nanometre to metre scales..." What Tsien believes is needed for biology is also true of the physical sciences. At the 2009 Spring meeting of the Materials Research Society there were several symposia specifically focused on complex hierarchical materials: "Three dimensional architectures for energy generation and storage," "Synthesis of bio-inspired hierarchical soft and hybrid materials," and "Architectured multifunctional materials."

X-ray imaging, especially with higher energy x-rays (20 kV and above), offers unique tools to address the critical issues of hierarchically structured systems. High-energy X-radiation penetrates through significant thicknesses of materials, for example, the entire active region of a semiconductor device, a small animal, or a sheet of structural alloy steel. Combined with high brightness at APS, this also enables the observation of fast and ultrafast processes as they are occurring, including fuel sprays, magnetic switching, and living organisms *in-vivo* (fig. 2.1.1). Images can be obtained using different contrast mechanisms, revealing light or dense elements, magnetic domains, or electric polarization for example. With improved -ray optics such as we are planning for the renewal, and new beamlines, we can image from the scale of nanometers to fractions of a meter. And through diffraction, scattering, and spectroscopy the lower end can be pushed down to the atomic level. Thus, x-ray imaging is the only tool that can encompass the many decades of length scales relevant to the control and understanding of hierarchical structures. Furthermore, hard x-rays are less damaging for thicker samples than soft x-rays and have greater elemental sensitivity because of increased fluorescence yield.

"Imaging specific molecules and their interactions in space and time will be essential to understand how genomes create cells, how cells constitute organisms and how errant cells cause disease. Molecular imaging must be extended and applied from nanometre to metre scales...", Roger Tsien

Phase contrast x-ray imaging, which requires excellent coherence in the delivered x-ray beam and favors a long beamline to increase spatial resolution and increase the coherent illuminated area, can visualize the internal structure of weakly-scattering objects, from insects (Socha

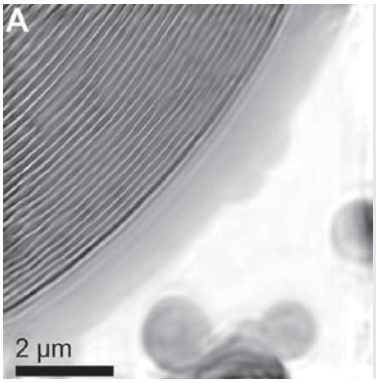


Pumping blood

Fig. 2.1.1. *From an x-ray video of the beating heart of a grasshopper. The round structures are air bubbles, which are revealed only by phase contrast, and allow visualization of the flow of hemolymph (courtesy W.-K. Lee). Insects are of interest not just because they include almost 80% of all living species, but because micromechanical systems inspired by insect function have promise for biomedical drug delivery. APS renewal will allow imaging of much larger animals.*

et al. 2007) to fuel sprays (Wang et al. 2008). Reduced radiation dose results from the improved sensitivity of phase contrast compared with conventional absorption contrast imaging. An exciting new development in the imaging field is coherent diffraction imaging (CDI) and related techniques such as ptychography (Thibault et al. 2008) (fig. 2.1.2). By these methods lensless imaging, or enhancement of resolution beyond the limitations of conventional optical resolution, is possible. CDI is also attractive for "real materials under real conditions in real time" because optics need not be placed close to the sample.

Phase contrast imaging and related techniques have wide application, for example, in understanding the drying of cement (Allen et al. 2007), materials fracture (Drummond et al. 2005), electromigration in semiconductor devices (Lee et al. 2007), and magnetic domain switching (Jaramillo et al. 2007). Hard x-ray imaging has typically had relatively poor resolution compared with soft x-rays due to the difficulties of making zone plates, but that limitation is no longer true, and there is real promise for a <5 nm resolution hard x-ray microscope with substantial depth of field using Multi-Layer Laue lenses (Kang et al. 2006) (see fig. 6.3.1).



Resolution extension

Fig. 2.1.2. *Using coherent illumination, the resolution of this x-ray image was extended [the original image did not resolve the outer zones of this zone plate (Thibault et al. 2008)]. A dedicated beamline in the renewal will allow resolution ~1 nm.*

Coherence is desired for imaging, but also for specialized scattering experiments. X-ray photon correlation spectroscopy (XPCS) exploits time correlations in the intensity of scattered coherent x-rays to characterize the dynamical behavior of samples (fig. 2.1.3). XPCS today addresses a unique combination of length and time scales for studying dynamics. Extending the spatio-temporal wndow will substantially expand the range of problems for study. In the renewal we will achieve an order of magnitude extension through a combination of improved source coherent flux, optics, and detectors.

A wide variety of complex fluids show structures with characteristic length scales that are in the few-tens-of-nanometers range and characteristic dynamics in the microsecond to millisecond range. Important examples of nanostructured complex fluids include mesophases of block copolymers and oil-water bicontinuous microemulsions that are stabilized by amphiphilic surfactants. In recent years, these sorts of phases have been the subject of intense interest, including detailed theoretical work focusing on their dynamics. Enhanced XPCS capabilities realized at a renewed APS to probe dynamics in this range of lengths and times will open new opportunities to investigate such materials. In polymer systems, despite the success of the concept of reptation in explaining many macroscopic properties, surprisingly few direct microscopic tests of reptation exist. Access to shorter times at a renewed APS will enable experiments that probe reptation in polymer melts on shorter length scales, thereby permitting critical examination of the reptation model for polymer dynamics.

X-ray imaging techniques are very powerful, but complementary to other methods. Transmission electron microscopy has extremely high resolution, albeit limited to thin specimens, whereas confocal optical microscopy can give rapid 3-D images of transparent objects. Users of the proposed x-ray biononoprobe will benefit from *correlative microscopy*. As an example, a user could image

Faster, smaller structural dynamics with XPCS

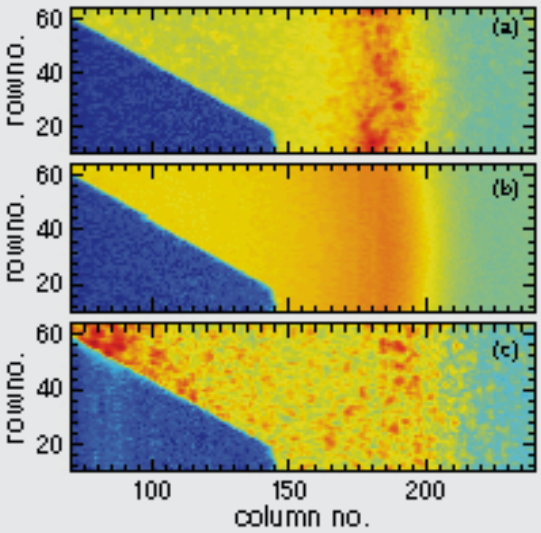


Fig. 2.1.3 *Access to dynamics at small length scales and short time scales makes XPCS valuable for studies of disordered materials, used for example in polymer processing of nanomaterials. The figure (Liu et al. 2008) illustrates averaged speckle patterns showing the relaxation in a model colloidal suspension as the strength of an attraction between the particles changes with temperature. (a) shows a repulsive glass at 33.00°C; (b) the fluid phase at 33.39°C; and (c) an attractive glass at 33.60°C. The fluid is dynamic so that the speckle evident for the glass phase is washed out. The repulsive and attractive glasses are unusual. Important faster processes like polymer reptation, which have eluded study so far, could be accessed with a renewed APS.*

the time-dependent distribution of specific proteins in live cells by using optical microscopy. At a suitable time the specimen could then be cryo-fixed, and its trace metal content measured in 3D and correlated with the protein distribution. This capability will allow users to interrogate much more precisely the interaction between necessary intracellular metals and proteins. Beyond the direct scope of the proposal, we hope that the State of Illinois will fund an Imaging Institute which could house long x-ray beamline end stations together with complementary tools and expertise for correlative microscopy.

Coherent diffraction imaging could unravel the structure of ligno-cellulose

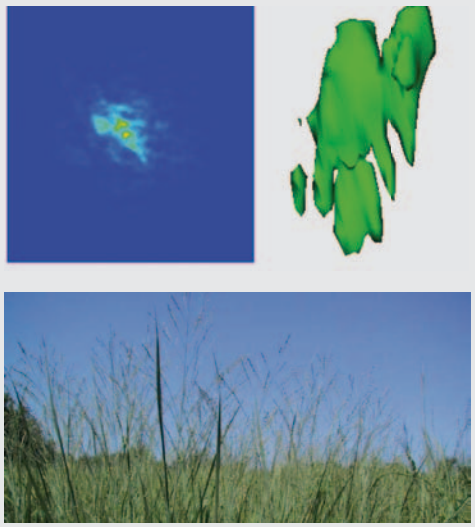


Fig. 2.1.4*. A coherent diffraction image (left) and 3D reconstruction (right) of a cellulose nanocrystal about 300-nm size, embedded in corn stalk. The image is reconstructed through the technique of Bragg CDI (Robinson et al. 2009). These preliminary results could be expanded with dedicated CDI capabilities in the renewal to unravel the structure of ligno-cellulosic material. The aim is to optimize the structure through genetic engineering towards a more efficient route to production of renewable biofuels (courtesy L. Makowski). Switchgrass is a possible source of biomass for such applications.*

Plant cell walls are complex polymeric matrices of cellulose, lignin, and other materials that represent the bulk of biomass available for conversion to biofuels. The vast range of mechanical properties of plants –from switchgrass to oak trees– is a reflection of the wide variety of ways in which these constituents can be organized. The cellulose in these materials is arranged in crystalline fibrils that aggregate laterally to form microcrystals which are highly resistant to degradation. Understanding the form of these crystals and the effect that various processing steps have on them is of critical importance for optimizing the production of biofuels from biomass. Bragg CDI (Robinson et al. 2009) is particularly well suited to 3-D imaging of small nanocrystals in a large complex matrix (fig. 2.1.4). X-ray fluorescence imaging can be used to establish true elemental concentrations for a large number

of chemical elements, at trace level sensitivity. Additionally, the chemical state of elements can be probed and elemental information can be combined with phase contrast imaging (or CDI) to form uniquely revealing pictures of hierarchical structures. Due to the large penetration depth of hard x-rays, this is not confined to thin sections. This capability is especially important in biology and environmental science (fig. 2.1.5) but also in materials energy research (Buonassisi et. al. 2005).

Viewing trace elements in 3D – a diatom that sequesters carbon

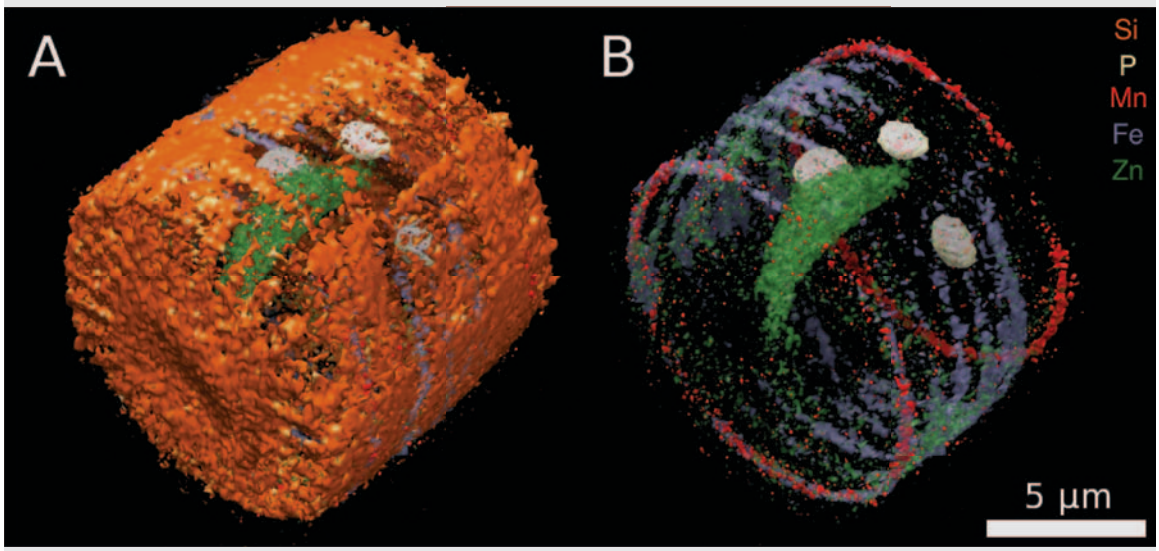


Fig. 2.1.5. *Shown above are 3D renderings of trace elements in a freshwater diatom (Cyclotella sp.), with (A) and without (B) its dense siliceous cell wall. Diatoms play a key role in global cycles of bioactive elements, including carbon. The growth of diatoms in the ocean is often limited by Fe, which has led some to propose or test fertilizing the ocean with iron to stimulate diatom growth and thereby sequester more atmospheric carbon within the deep ocean (Coale et al. 2004). The exact role of diatoms in these cycles, and thus efficacy of such concepts, are poorly understood, but x-ray fluorescence microscopy is an extremely well suited tool to study elemental distributions in these cells (Twining et al. 2003). Using tomography, we can now determine the 3D elemental distribution, and elucidate the biological basis that underlies differences in elemental composition and the functional consequences of those differences. Ultimately, such knowledge could improve models that link ocean productivity to atmospheric composition and climate. To acquire these datasets, several techniques had to be brought together, involving a widespread collaborative effort. The APS renewal will allow acquisition of these datasets with much improved spatial resolution, elemental sensitivity, and speed through dedicated, improved instrumentation and improved source brightness. For example, specialized phase contrast detectors (Honberger et al. 2008) will allow reconstruction of specimen mass (de Jonge et al. 2008), as well as alignment of individual projections that have poor statistics.*

2.2 EXTREME CONDITIONS

Substances studied under extremes of pressure, temperature, magnetic or electric field can point to new high performance materials (figs. 2.2.1 and 2.2.2), or explain seismic activity. Historically, extreme conditions have proved a significant tool in improving the properties of materials that have important applications to energy and economic vitality, such as high-temperature superconductors and high-mobility semiconductors. Recent high-pressure science has shown surprising complexity

Higher pressures

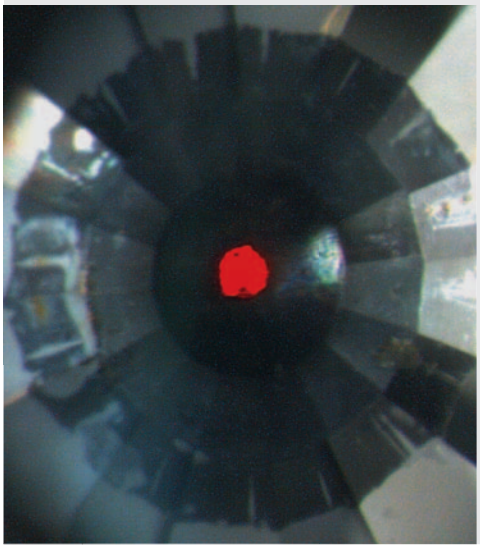


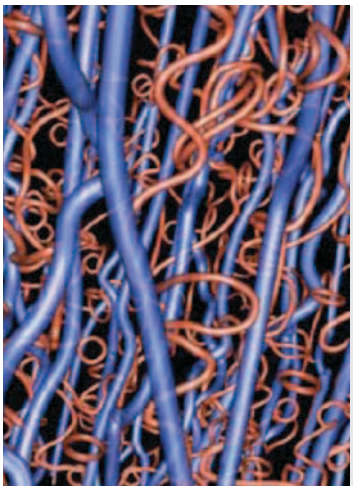
Fig. 2.2.1. *Even simple materials can reveal surprises at extremes of pressure. Solid oxygen at 38 GPa displays a red color characteristic of the epsilon phase (Meng et al. 2008). At higher pressures oxygen becomes a metal and a superconductor on cooling.*

Materials scientists desire higher pressures to explore new materials with new properties. Precise measurements under these conditions require that nanobeams be available at a renewed APS. High-flux, hard x-rays, and emerging techniques such as x-ray Raman inelastic scattering are essential to reveal the complex chemical bonding of light materials in pressure cells.

and richness in the structure and properties of even simple elements such as sodium (Gregoryanz 2008; Ma et al. 2009). The highest known transition temperature for a superconductor has been obtained under pressure (Gao et al. 1994). Under pressure a colossal increase in ferroelectricity has been observed in $PbTiO_3$ (Wu et al. 2005). "Extreme magnetic fields especially affect the outer-shell valence electrons that control insulating, magnetic, conducting and superconducting behavior through their strong correlations" states a recent report (BESAC 2007). It is imperative to note that extreme conditions are created in small volumes, in containments that lead to strong attenuation of x-rays with limited angular access. As such, the ability to obtain high-brilliance, high-energy x-ray beams of nanometer dimensions makes APS an ideal place to study extreme environments and reveal the structure, correlations, and dynamics of electrons, atoms, and spins.

Extreme conditions allow us to probe new properties of materials; test and strengthen our theoretical understanding and so address the grand challenge of materials by design; and understand the interior of our own and other planets. Pressure is the volume derivative of energy, so high pressure studies are important for revealing the equation of state. Synchrotron radiation studies have identified a post-perovksite phase of $MgSiO_3$, that is believed to explain the important D" seismic anomaly at the earth's core-mantle boundary (Hirose 2006). Among the topical problems in geophysics is the concept of a deep carbon cycle, coupling the atmosphere to the earth's interior, and impacting climate change and the options for carbon sequestration. Work at APS was the first to show, for example, that hydrocarbons could be formed at geological pressures from inorganic components (Scott et al. 2004).

High-energy synchrotron radiation has become an essential tool in the study of materials under extreme conditions. The APS has been amongst the most productive sources in these studies, leading to more than 55 papers in the high-impact journals *Nature, Science, PNAS,* and *Physical Review Letters* in the years 2006-2008, more than any other x-ray facility worldwide. But there is demand for new and improved techniques, and more access.



Ultradense hydrogen

Fig. 2.2.2. *Ultradense hydrogen has been predicted to exhibit a unique metallic superfluid phase (Babaev et al. 2005), which might be observable with the extended pressures and imaging resolution made possible by the renewal of APS.*

In the past decade, significant progress in high-pressure synchrotron research has been made in the study of charge and vibrational dynamics by spectroscopic techniques including inelastic x-ray scattering (section 2.5). These high-pressure techniques are particularly flux limited and will greatly benefit from the higher flux and more efficient detection made possible in the APS renewal.

Completely new regimes of phases of matter will become accessible through the use of nanobeams with higher flux, which are a major feature of the renewal. Such beams will probe more extreme conditions in smaller or less homogeneous samples. The impact of the proposed order-of-magnitude reduction in beam sizes is not limited to merely incremental improvements, but will greatly expand the range of questions that can be answered in static experiments. For example, precise single-crystal studies of high-pressure phases become feasible when beam sizes approach those of individual crystallites within a powdered sample. Even higher pressure conditions can be achieved for short periods of time in dynamic shock compression, also opening the important study of shock wave propagation in solids. Dynamic compression capabilities (utilizing high velocity impacts, high intensity lasers, and pulsed power generators), coupled with ultra-fast measurements facilitated by the renewal, represent the most versatile approach for achieving the widest range of thermo-mechanical conditions in a controlled manner. A beamline with dedicated dynamic compression capabilities would be the first of its kind at a third-generation high-energy synchrotron facility.

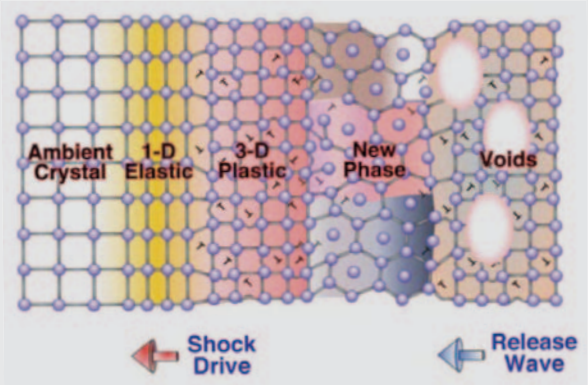
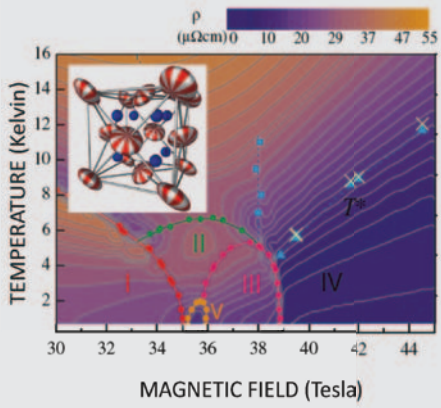


***Fig. 2.2.3**. Cartoon showing phenomena which can occur during shock deformation of a material, most of which have been predicted but not directly observed (courtesy H. Lorenzana).*

Shock deformation of materials occurs during materials failure and its understanding is important for stewardship of the nuclear stockpile. Novel, unexplored states of matter can occur at intense shock rates (fig. 2.2.3). Using intense focused beams from APS on small volumes permits more extreme conditions (e.g., pressures up to 500 GPa and temperatures up to 0.5 eV) to be attained with relatively simple equipment. At such pressures, localized electrons gain enough kinetic energy to mix with nearby valence and core electrons and create new phases of matter. Because of the interest in this capability from the DOE's National Nuclear Security Administration (NNSA), it has been separately proposed that a major part of the funding to build a dedicated shock compression beamline be provided by NNSA.

The magnetic field is a key fundamental parameter that enters directly into the energy expression (i.e., the so-called Hamiltonian) that governs all properties of materials. It is a contact-free experimental "knob" for tuning in novel phases that are of basic and applied interest. A dedicated beamline for application of high magnetic fields at APS has been recommended by a recent National Academy Study (COHMAG 2005) and is included in the renewal proposal. With the availability of high-magnetic fields (20-60 T) on a dedicated beamline, we may understand structure-property relationships in energy-related functional materials (multi-ferroics, shape-memory alloys, etc.), acquire insights into the mechanisms of high-temperature superconductivity, discover novel ordering phenomena and magneto-resistance pathways in complex magnetic systems for device applications, study the fundamental physics of of Bose-Einstein condensation in spin-gap compounds, and directly



Multipolar order and rich-phase diagrams

Fig. 2.2.4. *Given competing degrees of freedom and constraints imposed by a magnetic field, nature exhibits novel multipolar order and rich-phase diagrams in correlated f-electron systems. X-ray resonant and high-energy scattering studies of structural, magnetic, and electronic effects in high-field instruments can unravel the nature of such phenomena (Paixao et al. 2002, Kim et al. 2003).*

manipulate synthesis of real materials *in-situ*. Both pulsed and continuous high-field magnets would be included in the beamline. A collaboration with the NSF's National High-Magnetic Field Laboratory could extend the capabilities to include a split-gap continuous field magnet reaching well beyond 30 T (fig. 2.2.4).

An exciting frontier is the combination of high pressures, temperatures, and electromagnetic fields. Compact high-pressure cells, for example, can be accommodated in ~20 T commercially available cryogen-free magnets. We would plan to provide support for experiments utilizing these instruments through an extension of the HPSynC (High Pressure Synergistic Center) capabilities. HPSynC is a jointly funded activity today at APS which supports high-pressure experiments beyond HP-CAT and GSECARS, which have traditionally been the leading beamlines for high-pressure science. HPSynC and similar groups could provide a mechanism to coordinate the development of new "extreme environments" around the entire APS ring, while supporting their implementation in the user program.

Extreme chemical environments that are also of great importance are covered in section 2.4, "Interfaces in complex systems."

2.3 ULTRAFAST DYNAMICS

A fundamental and enduring scientific challenge has been the quest to understand the relationship between structure and function in complex systems. Most of the chemical, electronic, and spin interactions that are important occur, or at least begin, on ultrafast time scales ranging from below 1 ns down to below 1 fs. Ultrafast science aspires to visualize the structure-function relationship at time scales commensurate with fundamental molecular motions. Ultrafast x-ray science goes a step farther – permitting visualization at both atomic time and length scales.

The natural time scale of third-generation synchrotron sources is that of the stored electron bunch (~100 ps). There are many examples of important insights into ultrafast phenomena on this time scale, such as the dynamics during photosynthesis. The aim of photosynthesis research is to help us imitate this efficient and almost unlimited source of sustainable energy. For this problem and many others, there is a great desire to observe shorter time scales (fig. 2.3.1). Ultrashort laser-based x-ray sources have offered glimpses, but lack brilliance for many experiments. Free-electron lasers (FELs) offer a unique approach to ultrafast time scales; in fact, this has been a primary motivator for their development. While there will be no better source than FELs (such as the LCLS) on the horizon for single-shot pump-probe experiments, extensions to third-generation sources offer a complementary tool that avoids some of the limitations of FELs. Various approaches have been taken to "slicing" pulses in storage rings, with promising results. We propose to develop an rf-based slicing capability

inverted by Zholents et al. (1999) that offers a tunable, high-average flux source of pulses down to 1-2 ps in length. The SPX source can be accommodated seamlessly in the existing ring, assuming that R&D plans for its development are successful. The source would be able to take advantage of the very high-repetition-rate, high-power lasers being developed. Fig. 2.3.2 shows a comparison with the new LCLS FEL source and other laser slicing approaches.

The challenge of modern high-performance materials is that extreme properties or combinations of properties (e.g., colossal magnetoresistance) are often based on strong correlations between electrons and with the lattice, so that more sophisticated fundamental measurements are needed to understand and ultimately control strong charge-spin-orbit-lattice coupling. The SPX source will play an important complementary role in studying non-perturbative processes such as a photo-driven metal-insulator transition (Rini et al. 2007). These processes have been studied by soft x-ray spectroscopy, but key structural changes can be seen only with hard x-rays. The high-repetition rate and low charge per pulse afforded by the SPX source is also well suited for photoemission experiments due to low space-charge effects.

Understanding photosynthesis

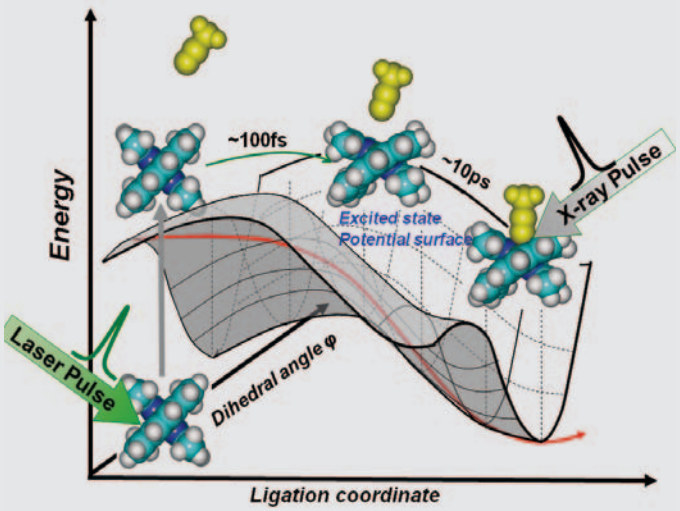
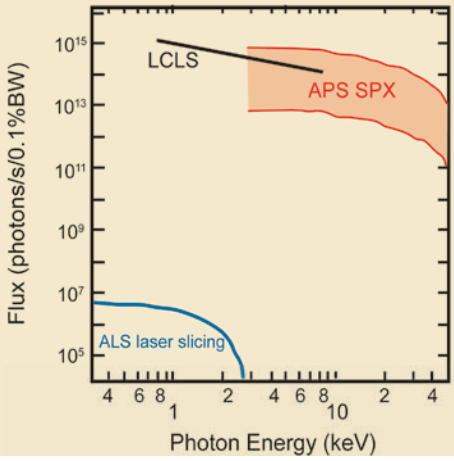
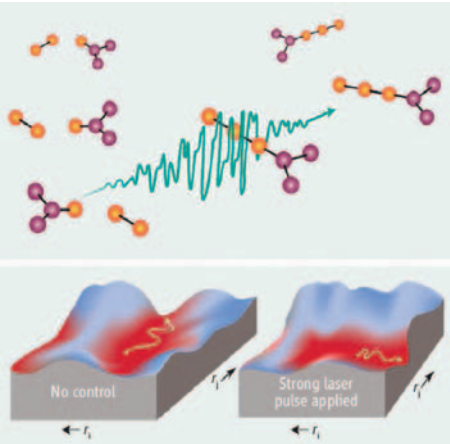


Fig. 2.3.1. *Structural dynamics of a photoactive copper(I) complex excited state created by green laser pulses could be followed by x-ray pulses to capture both charge transfer from Cu(I) to the ligand and dihedral angle movements on <100-fs time scale, intramolecular vibrational relaxation and vibrational cooling on the 10-ps time scale, and ligation coordinate changes on ps to ns time scales (Shaw et al. 2007).The current X-ray pulses from the APS can only capture the structure of the last species with solvent-dependent excited state lifetime of 1-100 ns, whereas the proposed short X-ray pulses will allow the ligation processes on the 10-ps time scale to be followed. The structural detail along the reaction coordinates captured by both ultrafast laser and x-rays will establish the correlation of the structure and excited state properties crucial to photochemical processes, such as photocatalysis. The structural dynamics insight may guide the synthesis in finding low-cost first-row transition metal complexes to replace precious metals in solar energy conversion processes, such as replacing ruthenium complexes in dye-sensitized solar cells.*



Comparison of ultrafast sources

Fig. 2.3.2. *Projected photon flux for various accelerator-based SPX sources. The LCLS has a projected pulse duration of ~230 fs, the Advanced Light Source (ALS) laser slicing source has a pulse duration of ~200 fs, and the APS SPX has a projected pulse duration of ~1 ps. The LCLS photon energy is discretely specified under accelerator operator control, whereas both synchrotron-based sources (APS-SPX and ALS laser slicing) are continuously tunable under local user control, thus enabling time-resolved spectroscopy. The high average flux of the APS SPX enables all x-ray techniques currently in use: spectroscopy, scattering, diffraction, microscopy, and imaging. Shorter wavelengths available from SPX favor atomic structural analysis.*



Photon control of chemical reactions

Fig 2.3.3. *The outcome of a photochemical reaction may be dictated by the application of strong non-resonant electromagnetic fields to modify interaction potentials (Rabitz 2006). These reactions can be studied by x-rays in the non-perturbative limit at the proposed APS SPX beamlines.*

By combining ultrafast x-ray probes with the quantum control enabled by recent advances in laser technology that tailor the phase and amplitude of electromagnetic pulses over a wide spectrum, we can uniquely tackle a 21st century challenge: photonic control of electron and atomic motion. One can address questions such as: Can one steer molecular processes to a desired outcome? Can one achieve ultrafast control of electron flow in semiconductors? Can one control non-radiative processes in semiconductors to enhance solar conversion efficiency? As a general tool, strong non-resonant electromagnetic fields can be used to modify potential energy surfaces to achieve a desired molecular transformation, as shown in fig. 2.3.3.

Using such fields, there have been remarkable advances in the photonic control of molecular degrees of freedom (Stapelfeldt et al. 2003) and of photo-induced reactions for simple (Sussman et al. 2006) and even complex (Laarman et al. 2007) molecules in the gas phase. The principal axes of isolated molecules can be aligned in the laboratory frame in the presence of a strong laser field and probed with the currently available 100-ps x-ray pulses at the APS (Peterson et al. 2008). With 1-ps x-rays it will be possible to go a step farther, i.e., to image laser-controlled rotational dynamics in the gas phase and thus define the spatial orientation of isolated molecules in a *field-free* environment (Ghafur et al. 2009). In the liquid phase, photonic control is at a more elementary stage due to complex solute/solvent interactions, dissipative processes, and the lack of incisive feedback tools as available in the gas phase (Brixner et al. 2001). The power of ultrafast x-ray spectroscopic or diffractive imaging to provide *in situ* information on molecular orientation and local structure as feedback for optimal control algorithms can be the key to unlocking the potential of coherent control in the solution phase.

Unfolding phonons

Another opportunity for SPX study is represented by phonon dynamics in solids, key to tailoring better-performance thermoelectrics that promise to mop -up wasted heat energy.

While ultrafast is usually taken to mean ps and below, many important materials-related phenomena, such as nucleation, occur on much longer time scales (ns to μs or above), either because they are improbable events limited by statistical mechanics (nucleation) or they involve massive cooperative phenomena (e.g., fracture)

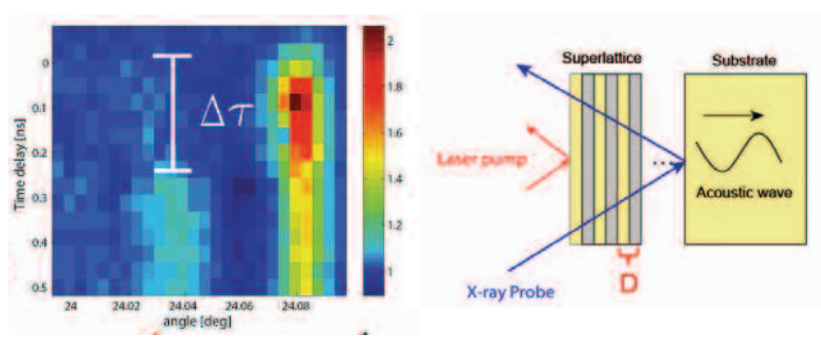


Fig. 2.3.4. *Phonons and their behavior in nanostructured materials influence important properties like thermoelectricity. In this study of InP superlattices, hard-to-detect acoustic phonons could be directly observed (Trigo et al. 2008). Shorter time scales from the proposed SPX source will allow higher frequency modes to be studied in the time domain.*

or diffusion over long length scales (μm to mm) (e.g., phase transformations). It is therefore of great interest to study phenomena over decades of times scales.

Fluid motion in sprays represents an example where sub ns-resolution is needed; in other cases phase transformations in heated ceramics may occur in seconds. Those can be studied by time-resolved x-ray imaging. In the areas of materials research, materials processing and fabrication, damage and failure can be visualized. Recently, ultrafast x-ray imaging found unique applications in studying transient fluid dynamics in high-pressure sprays (fig. 2.3.5). Many fluid dynamics problems need extensive theoretical and numerical simulation to understand the ultrafast process. Experiments are highly limited by current capabilities: flux, brilliance, and detector/optics efficiency. Ultrafast imaging will benefit from the imaging upgrades planned in the renewal (section 4.1), especially the long imaging beamlines that provide a wide imaging field while retaining high beam flux for time-resolved measurement.

A shot of fuel

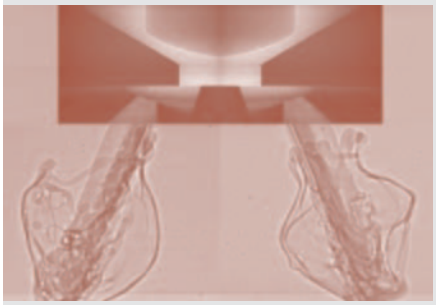


Fig. 2.3.5. *The image shows a single spurt of fuel from a gasoline injector captured with sub-microsecond pulses from APS (Wang et al. 2008). This image, using phase contrast to reveal the detailed hydrodynamics of the opaque fuel spray, also penetrates through the metal injector and plunger and yields understanding of the complex fluid flow that controls injection and combustion in modern internal combustion engines. Proposed long imaging beamlines will give better signal-to-noise ratios and allow imaging of much larger fields of view.*

2.4 INTERFACES IN COMPLEX MATERIALS

Modern surface science embraces the need to understand and study surfaces in complex environments. Interfaces between media (e.g., solids, liquids, gases, and soft matter) with distinct physical and chemical properties offer unique opportunity for discovery in diverse scientific areas such as catalysis, biomembranes, oxide film growth, semiconductors, geochemistry, surface physics, corrosion, nano-science, environmental science, tribology, and electrochemistry. The presence of an interface typically leads to gradients in the chemical potential, perturbs the properties of the two media to a depth controlled by the properties of the individual media, and introduces interfacial states (structural and electronic) that differ from those of either media—for example, in the formation of Schottky barriers.

Because of favorable cross sections, x-rays offer a unique opportunity to penetrate many complex environments (gas, liquid, or solid thin-film overlayers) to probe the structure and chemistry of surfaces and buried interfaces from macroscopic lengths down to the atomic level, in environments where traditional electron and other scanning probes are not applicable. High-brilliance hard x-ray sources such as the APS enable these *in-situ* studies, permit real-time investigations to elucidate mechanisms behind thin film growth, and allow for molecular-scale studies of important chemical interactions at internal boundaries using scattering, diffraction, resonance and absorption, fluorescence, standing wave, grazing-incidence small-angle X-ray scattering (GISAXS), and imaging techniques. Nanobeams made possible in the renewal will permit the observation of inhomogeneous surfaces, complemented by averaging approaches, such as extended x-ray-absorption fine structure (EXAFS) and small-angle and wide-angle x-ray scattering (SAXS/WAXS).

IN-SITU STUDIES OF MATERIALS SYNTHESIS

The design of new materials with emergent properties is a current and challenging problem in materials physics. Controlling the interplay between diverse microscopic degrees of freedom to create targeted macroscopic phenomena can result in the discovery of new materials with novel and technologically important properties. For example, the interface between $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) has been shown to exhibit metallic conduction. These are normally insulating materials that when grown epitaxially experience distortion and strains at the interface. In this case, the conducting 2D electron gas that forms at the LAO-STO interface is due to a subtle lattice dilation that lowers the conduction band minimum below the Fermi level (Wilmott et al. 2007). In general, heteroepitaxy is an important growth strategy in many technologies such as semiconductor devices, photovoltaics, ferroelectric memories, and superconductors; and controlling microstructural features such as strain and misfit defects plays a central role in thin-film science today.

Development of advanced materials is a theme that cuts across many of the grand challenges and priority directions for energy research. There are two types of challenges in developing new materials – the need not only to explore structure-property relationships to understand and ultimately predict structures of desired materials, but also to develop methods to synthesize these novel materials and process them into the optimal microstructure.

A traditional approach in film growth studies is to identify the most important growth parameters and investigate their effect on the structure and properties of the film. Despite the vast amount of experimental and theoretical knowledge that has been accumulated on epitaxial film growth, the growth of heteroepitaxial thin films is still largely dominated by empirical search and discovery methods that are slow, costly, and unpredictable. Even though it is known that the microstructure is affected by energetic and kinetic factors (such as temperature, background atmosphere, growth modes, lattice mismatch, and surface miscut), current approaches lack a fundamental understanding of the mechanisms needed to control the microstructure.

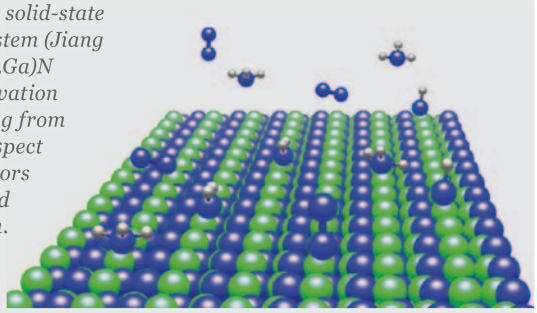
The penetrating nature of hard x-rays makes them a unique probe for *in-situ*, atomic-scale structural and chemical studies during materials synthesis. Current state-of-the-art *in-situ* x-ray studies of synthesis typically use diffraction or spectroscopy from millimeter-scale regions to observe the average properties of, for example, the growing surface in an epitaxial film. The nanoscale imaging capabilities provided by the APS renewal will allow observation of the atomic structure and composition of microstructural features underlying such complex synthesis processes, such as heteroepitaxial islands, domains, and phases. By observing these features in real time on the relevant time scales, e.g., from seconds to nanoseconds, researchers will determine the mechanisms of materials processes underlying the discovery of new materials for energy applications.

Semiconductors and related materials are ubiquitous in modern technology, yet improved growth and processing control reaching down to the nanoscale are demanded for a variety of new and extended applications. Efficient and precise techniques for semiconductor growth often involve complex conditions of high temperature and flowing gas, specifically in the case of metal-organic chemical vapor deposition (MOCVD). One topical example is shown in fig. 2.4.1. Alloys of InN and GaN are of critical importance for energy applications such as solid-state lighting and photovoltaics because the electronic band gap can be tuned across the complete visible and solar spectrum by varying the composition in this materials system (BESAC 2006). Synthesis of (In,Ga)N alloys with InN fractions higher than about 20% is difficult, however, because InN tends to decompose into elemental In and N. (In, Ga) N alloys are typically synthesized by MOCVD, where ammonia

can be used as a chemically reactive nitrogen precursor to grow this metastable material. It has been difficult to probe the surface mechanisms occurring in the high-temperature, reactive environment of MOCVD because of the lack of *in-situ* probes with atomic-scale sensitivity.

In-situ surface x-ray scattering and fluorescence measurements during MOCVD have recently revealed (Jiang et al. 2008) some surprising processes that are key to growth of (In,Ga)N(fig 2.4.1). Hard x-rays penetrate the reactive vapor phase and chamber walls for *in-situ*, atomic-scale structural and chemical studies during materials synthesis. In these experiments, the unusual phenomenon of a chemical reaction that oscillates in time has been observed during InN growth. Previously, oscillating chemistry had been found only in reactions of certain molecules in solution or on surfaces and in complex living systems such as microbe colonies and heart muscle. This new oscillatory system is the first one discovered that involves reactions and transformations between bulk condensed materials (solids and liquids). When a GaN surface is exposed to a steady vapor flow of ammonia and an indium compound at high temperature, a film of particles forms and then repeatedly transforms back and forth between crystalline InN and liquid elemental indium, alternately absorbing and releasing nitrogen to and from the vapor. Figure 2.4.1 (bottom) shows the oscillating fluorescence and diffraction signals that are proportional to the total (In liquid + InN) and crystalline InN. Modeling of the oscillatory behavior using continuum-level reaction-diffusion equations indicates that the production of active nitrogen by catalytic decomposition of ammonia at the surface is key to forming InN.

Chemical instability during growth



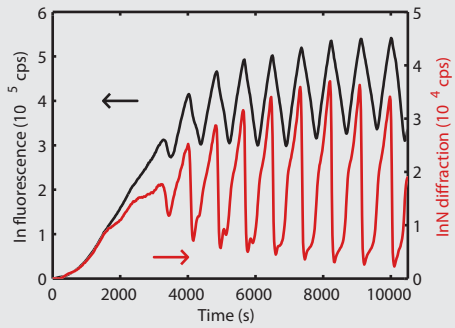


Fig. 2.4.1. *Oscillating surface chemistry, recognized in Gerhard Ertl's 2007 Nobel Prize in Chemistry, has been found for the first time in a solid-state vapor growth system (Jiang et al. 2008) – (In,Ga)N alloys. The observation is both fascinating from a fundamental aspect yet opens new doors to nanostructured control of growth. The improved facilities made possible by the APS renewal will be key to real-time imaging of the surface-vapor reactions on the sub-micron length scales needed to understand the fundamentals of materials synthesis. Determination and control of the surface chemistry responsible for high nitrogen activity during MOCVD, illustrated above (top), will allow creation of synthesis processes for the full range of (In,Ga)N alloys needed in lighting and related applications.*

Note that solid state lighting could obviate the need for 250 coal-fired power plants in the US.

By means of proposed nanoscale focused beams, the spatial variations in this fascinating chemistry could be directly resolved. Ultimately, such non-equilibrium phenomena provide a rich method to stimulate the growth of nanostructured materials. Coherent imaging also offers help in determining the dynamics of island formation, surface diffusion, and step attachment mechanisms leading to a more fundamental understanding of growth. This knowledge could result in the development of new synthesis strategies and an enhanced ability to use self-assembly techniques to produce novel structures and devices.

CATALYSIS

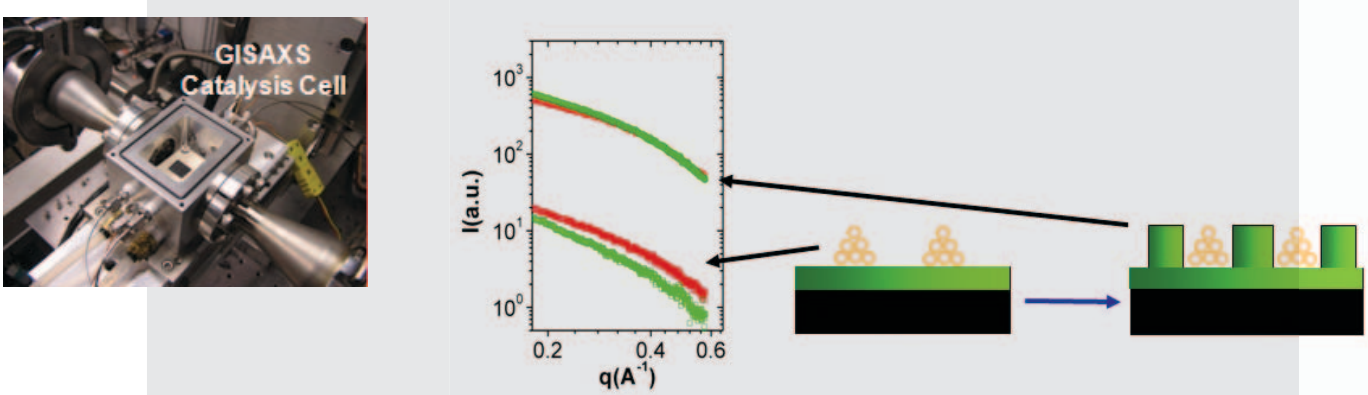
There is a mandate to develop catalysts with the specificity of enzymes, and control of the nanostructure is seen as the best hope. Since catalysts are used in 90% of chemical manufacturing, and affect over 30% of manufactured goods in the US (according to estimates by the Department of Commerce), reduction in the number of process steps and separations that would ensue from more selective catalysts offers greatly improved energy efficiency, reduced environmental damage, and better ecomomic competitiveness. Hard x-rays from a renewed APS will offer unprecedented capabilities for *in-situ* study of the structure and chemical dynamics of catalytic processes (fig. 2.4.2). Catalytic activity is predominantly an interfacial property, often at nanoparticles sitting on the surface of a support.

The two most significant barriers to the further use of efficient fuel cells are the high cost and limited durability of the platinum and platinum alloy nanoparticle cathode electrocatalyst. To solve these problems, we need to understand the atomic stucture and chemistry of electroctalysts under *operando* conditions (Wieckowski et al. 2003). In the renewal we will strengthen our catalyst research capabilities through a dedicated, primarily spectroscopic, beamline and a network of expertise and specialized equipment to bring catalyst research to other appropriate APS beamlines offering specialized techniques. In addition, APS, with support from Argonne, has jointly hired an expert in x-ray studies for catalysis (Jeff Miller). Jeff came from industry (BP), and his pioneering research is expected to attract many new uses and users from the chemical industry.

Making a more active catalyst, and keeping it so

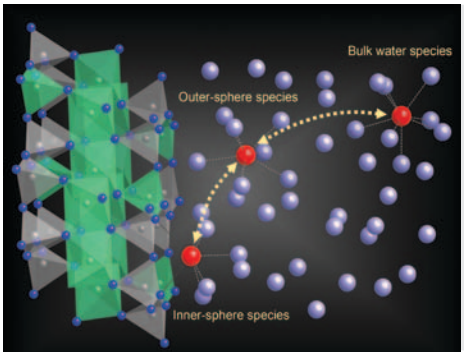
Fig. 2.4.2. *Catalytic conversion of propylene to propylene oxide (PO) is an important goal in the chemical industry, but selectivity of the silver catalyst is a problem. Nano-gold particles on titania have shown promise as a catalyst for the PO process but are poorly understood or controlled. GISAXS studies at APS point the way to improved performance. By a combination of theory, x-ray scattering, and mass spectrometry, the effects of size and shape of size-selected, soft landed sub-nanometer gold cluster catalysts and the nature of the support surface have been revealed. In particular, sintering of ultra-small particles reduces catalytic activity. Au_6 clusters shown below could be stabilized by selective atomic-layer-deposited (ALD) oxide overcoating (Lee et al. 2009). This Au_6 structure is known to exist in the gas phase and was calculated to be stable on a surface, but this is the first experimental evidence in the solid state.*

Understanding and controlling such phenomena invariably require observation of the entire system of catalyst and support under an active, realistic, and well-understood environment. We will make more extensive and sensitive capabilities available through the APS renewal.



GEOSCIENCES

Knowing how to meet global energy demand sustainably, without negative impacts on the environment, is a central challenge in the geosciences. In particular, a robust understanding of the processes that control the mobilities of the elements under environmental conditions is necessary to design safe and reliable geological repositories for sequestering energy-related by-products, including CO_2 and nuclear waste. The requisite knowledge spans large ranges in time and length scales, from those of fundamental molecular-scale chemical reactions to those of long-term large-scale transport processes involving groundwater flow and circulation of the atmosphere and ocean.



Rock meets water

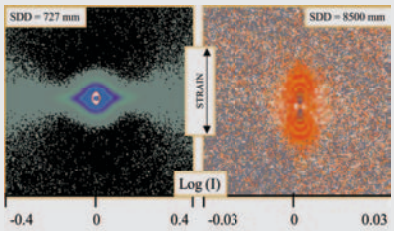
Fig. 2.4.3. *The process of cation adsorption at the muscovite-water interface, including a balance between adsorption of a fully hydrated (outer-sphere) species and partial dehydration of the adsorbing ion to form an inner-sphere species. Studies of this system could be extended to understand the role of surface inhomogeneities, such as steps, through the APS renewal.*

Many geochemical transport processes are effectively controlled by reactions occurring at mineral-water interfaces (e.g., adsorption, dissolution, precipitation, oxidation-reduction). These interfacial processes are best investigated experimentally through direct *in-situ* observations under the relevant physical and chemical conditions. Ongoing studies at APS have shown that the interaction of solute ions with even relatively simple mineral surfaces (e.g., muscovite) is more complex than previously realized. Ions are thought to adsorb solely as "inner-sphere" or "outer-sphere" species (i.e., as partially dehydrated or fully hydrated ions; fig. 2.4.3), but methods to test this conceptual model have been unavailable until recently. The application of *in-situ* resonant anomalous X-ray reflectivity has led to new clarity in this area by showing that cations can adsorb simultaneously as both types of species (Park et al. 2006). The influence of this new understanding on transport can now be investigated. This exemplifies the level of understanding required to refine molecular-scale models and to predict the large-scale processes of element transport in the Earth's near-surface environment.

HIGH-STRENGTH MATERIALS

The US Congress has mandated in the Energy Independence and Security Act (2007) that DOE carry out research "to determine ways in which the weight of motor vehicles could be reduced to improve fuel efficiency." This is just one example of the importance of lightweight high-strength materials. Materials that combine high-strength with toughness are always inhomogeneous–their properties are determined by intimately mixed nanoscale phases. Interfaces between components, whether it be glass/polymer (fig. 2.4.4), or grain-boundaries in steels, play a dominant role. A major limitation to the implementation of lighter-weight alloys is the lack of fundamental understanding of the mechanical properties under deformation. Because of a lack of predictive capability, the "spring-back" that occurs during metal stamping to produce car bodies cannot be reliably controlled. High-energy x-rays that are focused down to small volumes below 1 micron provide the only technique for spatially resolved determination of the Nye tensor, which describes elastic and plastic deformation in materials. Control of stress distribution during processing and materials service is critical to 3-D integration of semiconductor circuits, prosthesis, stress-corrosion cracking failure of infrastructure,

High-strength materials



Above, walking on glass over the Grand Canyon demands unique materials.

Fig. 2.4.4. *DuPont researchers have used the APS to watch the changes in a butacite polymer strengthening film as it is stressed (Londono 2009).*

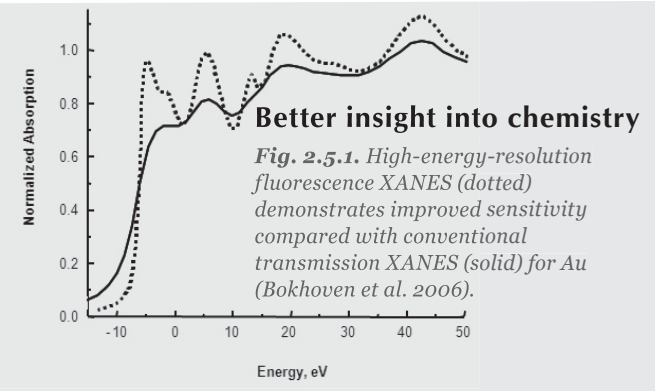
and extension of the lifetime of nuclear plants. Processing and control of soft materials down to the nanoscale is a major frontier in nanotechnology, where it promises a new paradigm for the manufacturing of materials through self-assembly. Figure 2.4.4 shows an application of *in-situ* testing of polymer films used to strengthen glass of the kind employed to form the "air bridge" over the Grand Canyon. These materials also have major security and safety applications across the world.

ELECTROCHEMISTRY

Lithium-ion batteries have cornered the lion's share of present day rechargeable batteries for consumer electronics and power tool applications. They are also poised for use in hybrid and plug-in hybrid vehicles. *In-situ* x-ray scattering and x-ray absorption spectroscopy have contributed significantly to the advancement of rechargeable battery technology. However, to minimize our dependence on fossil fuels and to help mitigate climate change, it is imperative that newer battery technology, which can be used in all-electric vehicles with useful range, be developed in the near future. Also, it is likely that storage of electricity generated from other renewable energy sources (such as solar or wind) will be an important component of the nation's future energy portfolio. Any game-changing energy storage technology should provide energy and power densities comparable to gasoline engines, yet be inexpensive, inherently safe, and green for large-scale deployment. Development of such transformational battery technologies requires tailoring novel materials and incorporating new chemical processes. Critical to such advances are breakthroughs in fundamental research involving a thorough understanding of bulk and interface structures, phase behavior, diffusion mechanisms, redox behavior, and electrocatalytic behavior. The development and application of emerging hard X-ray techniques will provide key new information that meets or exceeds the basic characterization needs articulated in the recent *BES Report* on *Electrical Energy Storage*. Fielding these batteries in a practical automobile fleet and other industrial applications requires a thorough study of reliability and failure mechanisms. These breakthroughs will be facilitated by our proposed renewal, specifically though better imaging beamlines, higher resolution spectroscopy capabilities with greater sensitivity, and better access to experiments in realistic environments.

2.5 HIGH-RESOLUTION SPECTROSCOPY

X-ray spectroscopy reveals details of the composition, chemistry, local structure, and excitations in materials, complementing data from scattering and imaging. Revolutions are occurring in many x-ray spectroscopic techniques, and we plan to develop these capabilities further for our users through the APS renewal.

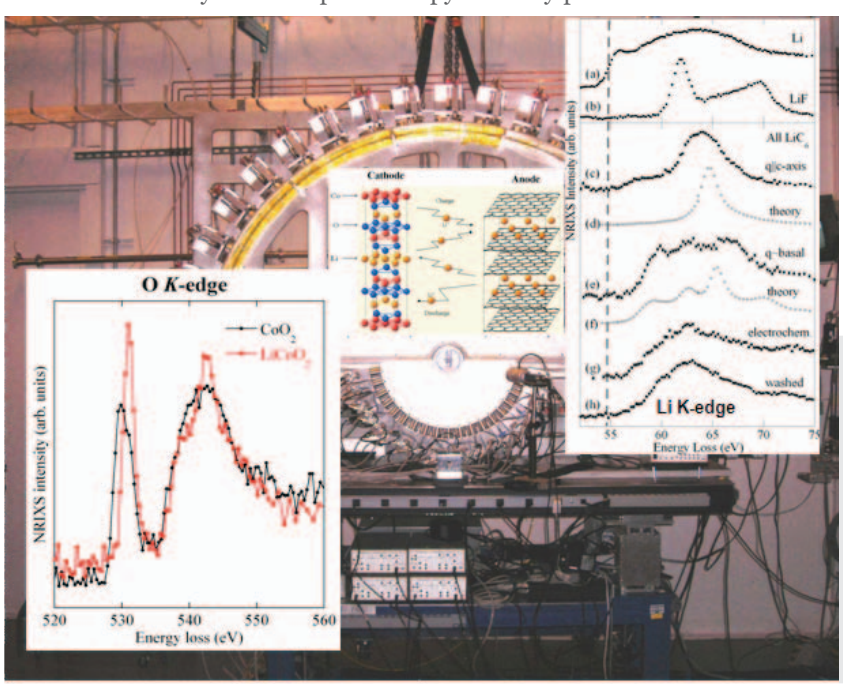


Better insight into chemistry

Fig. 2.5.1. *High-energy-resolution fluorescence XANES (dotted) demonstrates improved sensitivity compared with conventional transmission XANES (solid) for Au (Bokhoven et al. 2006).*

For example, applied to catalysis, x-ray absorption near-edge structure (XANES) and EXAFS are the most widely used techniques to characterize the structure and bonding of active species in real chemical environments. Increase in the brilliance and focusing efficiency of the source, together with improved detectors, will allow smaller volumes to be probed that are important since many industrial catalysts are heterogeneous. For determination of subtle yet important structures, better signal-to-noise ratio is demanded. XANES can reveal the chemical state of elements under complex conditions in real samples. While this analytical method may lack the chemical specificity of, for example, infrared spectroscopy, it is probably the only way to reveal the spatially and elementally resolved chemical state of a component in a multi-constituent reaction under realistic conditions. Improvements in sensitivity and nanofocusing planned in the APS renewal will allow wider exploitation of this sensitivity, building for example on the ability to distinguish amino acids (Kaznacheyev et al. 2002). A recent variant on XANES known as high energy-resolution fluorescent XANES (HERF) has demonstrated far greater sensitivity to subtle chemical and structural features (fig. 2.5.1).

X-ray Raman spectroscopy is a very powerful tool for examining the chemical structure and environment in light elements embedded in thick or absorbing objects (fig. 2.5.2). This technique has been widely used in high-pressure research to examine light elements, including hydrogen. Recently x-ray Raman has been demonstrated to be applicable to working Li-ion batteries, providing



Lithium ion batteries

Fig. 2.5.2. *Non-resonant hard x-ray scattering methods developed at APS provide definitive characterization of the redox behavior of Li-ion battery materials (Balasubramanian et al. 2007). Truly bulk-sensitive soft x-ray absorption information can be obtained by the X-ray Raman technique. The low cross section of the processes of interest necessitates an optimized third-generation source coupled with a specialized instrument such as the LERIX scattering spectrometer (shown in the background).*

Collective excitations in superconductors

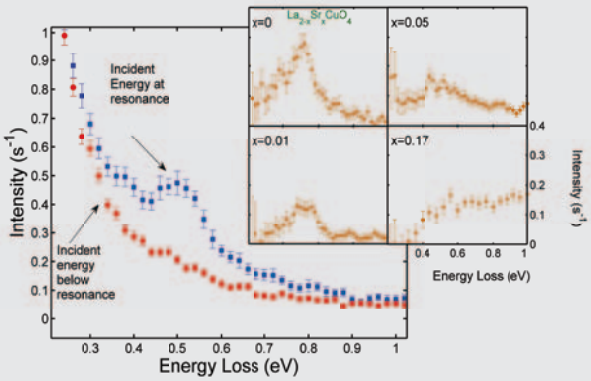


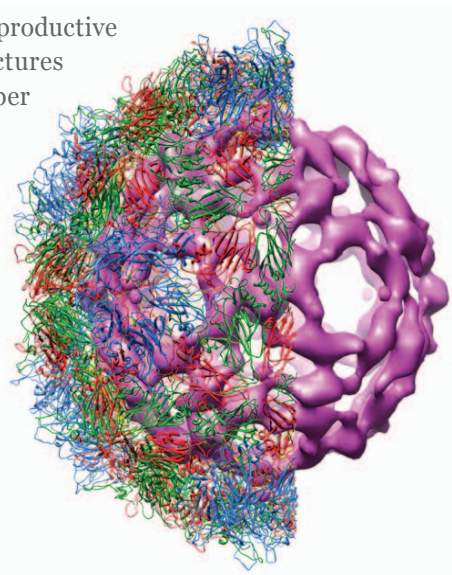
Fig. 2.5.3. *A RIXS study of collective excitations in the superconductor $La_{2-x}Sr_xCuO_4$ shows an excitation that has not been explained by conventional theories of this mysterious high-temperature superconductor (Trigo et al. 2008). (The inset boxes show the effect of doping on the excitation.) The renewal will offer ~10x better signal-to-noise ratio.*

important information about the lithium chemistry, as well as measurement of the oxygen K-edge and transition metal L-edges in the host metal oxides (fig. 2.5.2). The technique combines the power of soft x-ray spectroscopy with the *in-situ* capabilities of hard x-rays. With appropriate detectors it is possible to explore the Q dependence of the scattering and to access a unique region of the dynamic structure factor S(Q,ω), which cannot be observed with shorter wavelength electromagnetic radiation. The technique is sometimes known as low-energy-resolution-inelastic x-ray scattering (LERIX). We plan to develop a dedicated capability for LERIX in the renewal.

Medium-energy-resolution resonant inelastic x-ray scattering (RIXS) provides information about the excitations of materials that is important, for example, in the study of correlated electron materials. This is an area where theory is needed to interpret the complex spectra. It is nonetheless one of the only ways to access the intriguing momentum-resolved collective electron excitation spectrum, as demonstrated in fig. 2.5.3. High-energy-resolution and nuclear-resonant inelastic x-ray scattering techniques offer from 10^{-3} eV to 10^{-6} eV (in special cases) energy resolution and are able to reveal the phonon dynamics from very small volumes or with element-specificity. These techniques are well suited to the study of extreme conditions, where more conventional methods to measure thermodynamics or acoustic dispersion are thwarted. We plan to increase the capacity and improve the resolution, signal-to-noise ratio, and efficiency of these beamlines through the APS renewal. Magnetic circular dichroism is a powerful source of information about element-specific magnetism. A tenfold increase in sensitivity to magnetic moments is expected from new polarizing undulators.

2.6 CONNECTING PROTEINS TO ORGANISMS

Protein crystallography has been one of the most productive fields for APS users. More than 1200 protein structures per year are solved by APS users, the largest number produced by any source in the world. An example of a recent high impact structure is shown in fig. 2.6.1. We expect the demand for protein crystallography to be strong in the future, but the field is changing. Users are demanding more rapid structural analysis and screening, which is facilitated by advanced automation and improved detectors that are included in our renewal planning. However, the future is not determined only by throughput. It has often been



A virus that attacks lung cancer

Fig. 2.6.1. *Image from just one of the 50 high-impact scientific papers (Venkataram et al. 2008) published by structural biology users of APS in 2008. The 3D structure shows the Seneca-valley virus-001, that is of great interest because it infects lung cancer cells. An enemy of cancer cells is a friend from which we hope to learn new cancer therapies.*

assumed that the principle bottleneck in protein crystallography is that crystals sometimes cannot be obtained. But for many problems microcrystals that have been too small to study in the past are often produced. Alternatively, inhomogeneous crystals that do not diffract well when viewed over large regions have to be discarded today. All this promises to be improved by microcrystallography. Diffraction by micron-sized crystals, or micron-sized regions of larger inhomogeneous crystals, promises to open up previously inaccessible protein structures (fig. 2.6.2). Prime amongst these are membrane proteins. There is also a suggestion that smaller crystals may be less prone to radiation damage due to photoelectron escape, especially at higher x-ray energies in the 20-30 keV range (R. Fischetti, private communication), although dose limits the ultimate minimum size of microcrystals for practical use.

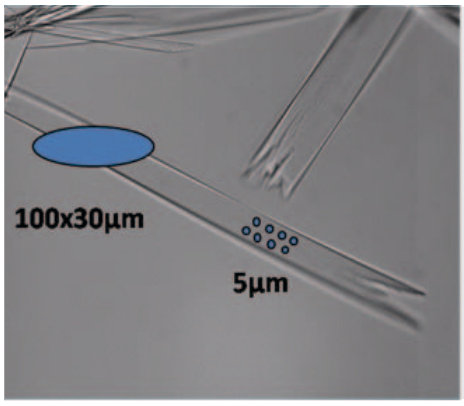
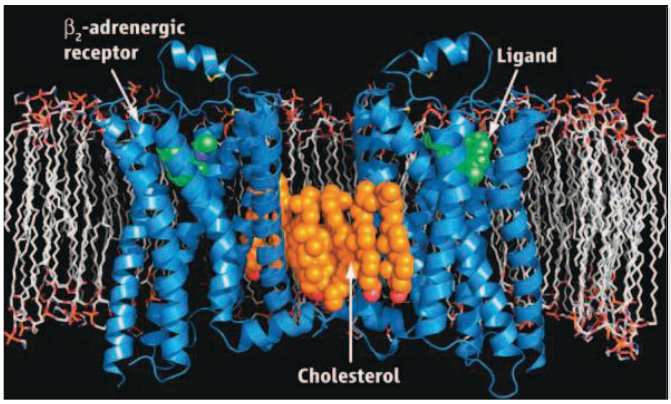


Fig. 2.6.2. *Optical imaging demonstrates the value of a micron-sized beam for protein crystallography on inhomogeneous or odd-shaped crystals (courtesy B. Koblika and W. Weis, Stanford University). Data collected within the 100x30 µm beam had a poor signal-to-noise ratio because of background from the surrounding mother liquor. The investigators were able to piece together a high-resolution full data set from merged 5 µm data sets at APS. The use of smaller or more imperfect crystals opens up the frontier to, for example, membrane proteins that have been hard to crystallize well. The challenge is to implement this with smaller beams and crystals.*

G-protein coupled receptors (GPCRs) represent the largest family of integral membrane proteins encoded by the human genome (Lefkowitz 2000). All GPCRs share a common structural motif, consisting of 7-transmembrane α-helices with N- and C-termini located inside and outside the cell, respectively (fig. 2.6.3). Together, these proteins are responsible for transmembrane signaling of most hormones and neurotransmitters, and constitute more than 30% of the targets of currently approved drugs. Rhodopsin, purified directly from bovine retina, yielded the first atomic resolution structure of a GPCR (Palczewski et al. 2000), providing critical insights into its role in the mammalian visual system and a long-awaited foundation to guide studies of other family members. During the past two years, our structural knowledge of GPCRs has increased 300%, following structure determinations of the β1 and β2 adrenergic receptors (Warne et al. 2008, Cherezov 2007, Rasmussen et al. 2007, Rosenbaum et al. 2007), which mediate the effects of adrenaline and various cardiovascular drugs, and the adenosine A2A receptor (Jaakola et al. 2008), which mediates the effects of caffeine. These landmark achievements relied on recombinant protein expression in bacteria and insect cells coupled with painstaking efforts to understand solubilization and purification behavior in the presence of lipids/detergents, stabilization via deletion of unstructured N- and/or C-terminal segments, introduction of point mutations, insertion of whole proteins into inter-helical segments, binding of monoclonal antibody Fab fragments, complex formation with small-molecule ligands, and the myriad ways that lipids/detergents can influence crystallization.

The appearance of crystals by no means signaled the end of the struggle. Typical GPCR crystals are minute (frequently no more than a few tens of µm in the longest dimension) and exquisitely sensitive to radiation damage. They also have the exasperating property of exhibiting high-resolution X-ray diffraction only from within certain µm-sized sub-domains and not others. Success with



Microfocus x-ray crystallography is essential for G-Protein coupled receptor structure determination

Fig. 2.6.3. *Structure of the β1 adrenergic receptor (blue) embedded in a lipid membrane and bound to a diffusible ligand (green) with cholesterol and palmitic acid (orange) between the two receptor molecules. [from (Cherezov 2007)]. More than 30% of human therapeutics act on G-Protein coupled receptors-membrane proteins that have been very hard to study without microcrystallography.*

rhodopsin and the β adrenergic and adenosine receptors ultimately relied on repeated access to microfocus beamline facilities at the APS and the European Synchrotron Radiation Facility, which offer high signal-to-noise X-ray diffraction cameras capable of illuminating crystal sub-domains as small as ~5 μm in diameter. Having broken the log jam, microfocus X-ray beamlines promise to reveal many novel GPCR structures over the coming years, thereby enriching our understanding of hormone/neurotransmitter signal transduction in both normal tissues and disease. We can also expect the pharmaceutical industry to follow the course charted by academic structural biologists and third-generation synchrotron radiation facilities as they, in turn, exploit structure-guided approaches to discover more-selective therapeutic agents targeting GPCRs. Improved beam stability, nanopositioning ,and detectors in the renewal plans will facilitate the success of this important avenue for structural biology.

Beyond static structure of proteins, it is important to understand protein dynamics. This is possible through inelastic scattering and through direct ultrafast scattering and spectroscopy. A particularly exciting direction is the activation of protein activity that in itself is not photosensitive, but uses a photo-activated modification that permits laser pump/x-ray probing of the dynamics of a more general class of biochemical reactions (see fig. 4.6.1).

Ultimately, the grand challenge is to connect proteins with organisms through their role in organelles, cells, organs, and beyond. The imaging and coherence beamline developments are motivated by this grand challenge of understanding the amazing hierarchical structure of life. The bionanoprobe beamline in the renewal will bring more sensitive nano-focused beams into the field of metals in biology and include cryogenic sample stages that are not available at our beamlines today. The sensitivity of x-ray fluorescence will illuminate the key role played by trace metal concentrations in both health and disease. Proteins in solution or in other environments typical of in-vivo conditions can be studied with greater and greater precision using SAXS/WAXS experiments. This field is experiencing rapid growth as more researchers learn that the techniques can provide information about protein folding, the conformation of multi-subunit macromolecular complexes, ligand/drug binding, and precise distances between wild-type or engineered anomalous scattering centers. Solution scattering is weak and anomalous effects in solution are even weaker. Current solution SAXS/WAXS capabilities at APS are not optimized. The renewal will concentrate these experiments on a dedicated beamline with state-of-the-art detectors, allowing for simultaneous collection of SAXS/WAXS data.

We also recognize the importance of providing ancillary and complementary equipment and biological expertise to our users and propose to do so.

3. ACCELERATOR SYSTEM UPGRADES

In this section we describe upgrades to the accelerator system, front ends, and initial x-ray delivery optics. The APS storage ring was designed with great flexibility, favored by the high electron energy and large ring circumference. In the planned upgrades to the accelerators and x-ray delivery optics we take full advantage of this flexibility to deliver optimized x-ray beams to users. Choices were driven by user demands for higher performance or increased capacity without loss of performance. These coupled improvements will deliver higher brilliance, more stable beams to all beamlines at APS, and in many cases, deliver one to two orders of magnitude improvement in delivered end-station x-ray flux because we will accommodate optimized insertion devices (e.g., for high-energy beams). In other cases, the capacity can be increased for very high demand techniques (such as SAXS) without loss of performance, through canting of undulators. The proposal also includes a comprehensive upgrading of outdated optical components in the x-ray delivery systems which will allow all beamlines to take full advantage of available undulators, higher current, and more stable beams.

3.1 LATTICE OF LONG STRAIGHT SECTIONS

The present APS lattice consists of 40 sectors that are largely identical in terms of the size, arrangement, and nominal settings of the main accelerator magnets. Each straight section allows a maximum of 4.8 m for insertion devices (IDs). However, based on workshops held to develop the science case for the APS renewal, it was clear that longer straight sections were required. We propose to provide eight 7.7-m long straight sections as part of the upgrade.

Increasing the length of an APS straight

Fig. 3.1.1. *Schematic of the lattice changes for a longer straight section. The proposed increase in length from 4.8 to 7.7 m is achieved as shown by removing Q1 and shortening Q2 (Key: dipoles -red; quadrupoles - blue; sextupoles - yellow).*

Perhaps the most obvious scientific driver for a long straight is the ability to use a single, longer device to deliver higher flux and brightness, e.g., for increasing the total flux for photon-hungry experiments such as inelastic x-ray scattering and increasing coherent flux for x-ray photon correlation spectroscopy. However, there are other uses. A long straight section (LSS) allows canted beamlines with longer devices compared to what is possible with the current 4.8-m straights, increasing capacity without sacrificing performance. Alternatively, a longer straight can accommodate multiple, in-line devices with different periods, which can optimize the spectral output for a one-of-a-kind beamline or end station (e.g., for high-pressure science). Yet another benefit is to better accommodate superconducting insertion devices that will require transitions from room temperature vacuum chambers to low temperature chambers. Finally, there is the ability to accommodate special equipment in the straight sections without excessively restricting the length of the insertion devices. An example of such equipment would be superconducting deflecting cavities for producing short x-ray pulses.

To ensure adequate dynamic and momentum apertures, LSSs must be arranged in a symmetric pattern, which requires that the number of LSSs should be a factor of 40, i.e., 2, 4, 5, 8, 10, 20, or 40. In addition to deciding how many LSSs to implement, we must choose the method of implementation. The most attractive option is to remove the Q1 quadrupoles on either side of a straight section and replace the 0.8-m-long Q2 quadrupoles with 0.5-m-long quadrupoles (see fig. 3.1.1). This allows 7.7 m for IDs. This scheme is the most economical and straightforward because no new magnet designs or power supplies are needed, and the LSSs can be installed in the normal shutdown periods of the storage ring.

We have developed a series of accelerator lattices that implement 2, 4, and 8 long straights using the approach described above. To satisfy user demands, we propose the 8 LSS option. Although these lattices perform well, it is not possible to control the lattice functions in the modified sectors as flexibly as in the normal sectors due to the reduced number of quadrupoles and sextupoles. The reduction of magnets also results in the dispersion in the LSS being somewhat larger than in the normal straights, giving an increased effective emittance for the LSS of 3.60 nm, compared to 3.36 nm for the normal straights. The present effective emittance is 3.15 nm. We are exploring ways to reduce the emittance of the LSS lattice with more aggressive lattice optimization schemes.

In addition to modeling the 8 LSS lattice to optimize its dynamic and momentum apertures, we have experimentally tested an 8 LSS "mock-up" lattice by removing power from the magnets we are proposing to remove, an experiment easily performed since all the magnets at the APS operate on independent power supplies. We have found that this lattice has excellent injection efficiency and lifetime, giving us great confidence in our ability to implement 8 LSS without major operational issues. Rather than evenly distributing the 8 LSS lattice, we are exploring a preferred option to group the LSS in pairs, making a four-fold symmetric configuration. This option would have the advantage of allowing all LSSs to be placed outside of the injection and rf areas and be available to users. In addition, it would better accommodate the SPX system, which may benefit from two nearby LSSs.

3.2 HIGH-BRILLIANCE INSERTION DEVICES

PLANAR UNDULATORS

There are presently 45 IDs installed at APS. Thirty remain standard 33-mm-period undulators A, that provide a continuous tuning range above 3.2 keV. Over the years, to meet users' needs, a variety of other period lengths, including 18, 23, 27, 30, 35, 55, and 128 mm, have been produced and installed. The undulator A, still predominant, has served APS users well as it is a flexible and broadly tunable device. However, as we move away from multi-purpose beamlines, there are opportunities to provide many experiments with an order-of-magnitude higher brilliance source, without increased heat load, by using undulators with lower k value optimized for a particular energy range. In the renewal we expect to install ~30 new optimized undulators. Many will be longer than the current 2.4-m standard to accommodate the long straight sections or fully utilize the 4.8-m straight.

Some of the more recently installed IDs are shorter than the standard 2.4-m length to permit "canted" installation, where two separate beamlines are fed from separate undulators in a single straight section. While the necessarily shorter IDs reduce the brightness, they allow for optimized performance of each beamline and/or a higher sample throughput from a single straight section. Increased beam current (section 3.3) will more than compensate for this brightness loss.

Tuning curves for several devices calculated for the renewed lattice are included in fig. 3.2.1. While longer period lengths, like 33 and 30 mm, are required for a continuous tuning curve with no gaps between the first and third harmonics, shorter period lengths can give higher brilliance within a limited tuning range at higher energies. For instance, the 23- and 27-mm devices developed to date were requested by users for their increased spectral brightness at high energies using the third and fifth harmonics. Even more dramatic enhancements in brightness at high x-ray energies can be obtained with superconducting undulators. We are presently prototyping a superconducting undulator with a period of 16 mm that would have a first harmonic tunable from 20 to 25 keV (see section 6 .2 on R&D activities).

Longer insertion devices, coupled with increases in stored current, will require upgrades to the front end and certain high-power beamline components (slits, windows, beamstops, etc.). This too will be a component of the APS renewal (section 3.4).

APS will remain the most brilliant source of US high-energy x-rays

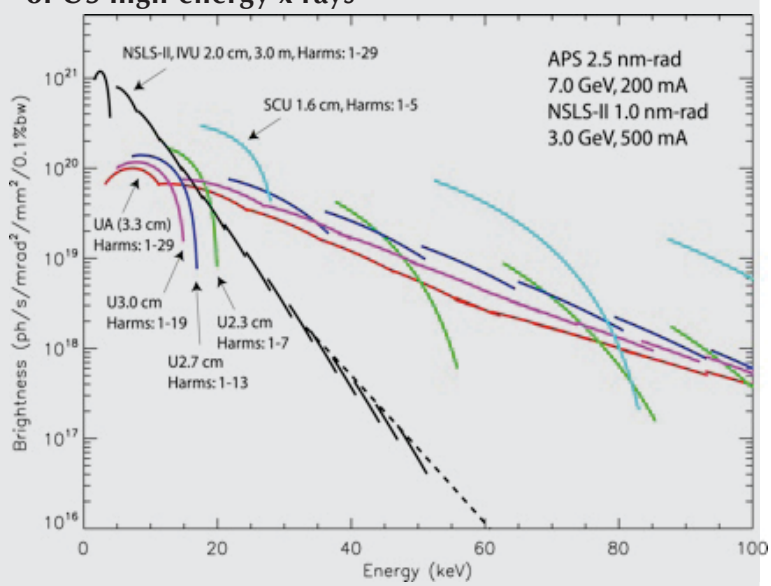


Fig. 3.2.1. *Tuning curves (odd harmonics) for existing APS undulators in the renewal (2.4-m length, 200-mA current), a future APS superconducting undulator (SCU 1.6 cm period), and the planned 2.0-cm-period in-vacuum undulator at the NSLS-II.*

POLARIZING UNDULATORS

The renewal plans for additional types of undulators – for example, an APPLE-style undulator. APPLE undulators have attractive polarization characteristics that include control of the direction of the polarization vector, the helicity of the polarization, and other features. In the circular-polarized mode, this device also produces mostly first harmonic on axis. By masking off-axis radiation that carries the higher harmonics, one can protect heat-sensitive optical components downstream.

For the intermediate energy x-ray beamline under construction we have developed a new type of undulator to produce lower energy photons, from 200 eV to 2.5 keV. It will be an all-electromagnetic, variably polarizing, quasiperiodic device. This device could be used in circularly polarized mode. Even with the finite e-beam emittance and finite aperture, the first harmonic noticeably dominates in the on-axis radiation. Additionally, the quasiperiodic magnetic structure generates high harmonics shifted in energy from those that are produced by a periodic device. As a result, the x-ray monochromator will transmit exclusively the first harmonic radiation that leads to substantial improvement in the signal-to-noise ratio for experiments.

3.3 HIGHER ELECTRON BEAM CURRENT

Higher stored beam currents increase the brightness and flux of all sources and can compensate for losses in shorter undulators used in canted configurations. A configuration of multiple, differently purposed, in-line undulators would also benefit from such compensation. The APS storage ring currently operates with 100 mA of stored beam current. Although the accelerator was originally designed for up to 300 mA operation, operations at higher currents with our existing fill patterns (required for dynamic or time-resolved studies) will require some accelerator upgrades. Through machine studies (Harkay et al. 2007), we have identified storage ring components that limit the operational current for present bunch patterns and have developed R&D plans to achieve a current of 200 mA in all operating modes. The principal limitations are coupled-bunch instabilities (CBI) and excessive beam-driven heating, both of which are to be alleviated with system improvements described below. The beam lifetime at 200 mA is expected to be reduced for the 24 equally spaced bunch pattern that we presently operate with a majority of the time. This limitation is due to the high chromaticity required to stabilize the bunch against transverse wakefields. These wakefields are mostly produced by the aperture transition of the ID vacuum chambers. New, optimized transitions will be installed to reduce the required chromaticity and increase the lifetime.

RADIO-FREQUENCY COMPONENTS

Coupled-bunch instabilities are caused by higher order modes (HOM) in rf cavities. HOM dampers are installed to suppress HOMs . Beam-driven heating is bunch-pattern dependent and occurs in rf-cavity components and in cavity-like storage ring components, such as beam scrapers. We presently have HOM dampers in 4 of the 16 rf cavities, which permit stability for all bunch patterns with total currents up to 164 mA, and for 324 bunches up to 245 mA. New rf cavity HOM dampers will be replacing the exiting HOM dampers and will be installed on four additional rf cavities (fig. 3.3.1). For reliable 200-mA operation, the existing input power couplers will be replaced with a modified design that is being fabricated and will be tested and evaluated in the rf test stand. It is believed that the CBI threshold will be raised through these improvements, and stable operation in all modes at 200 mA will be achieved.

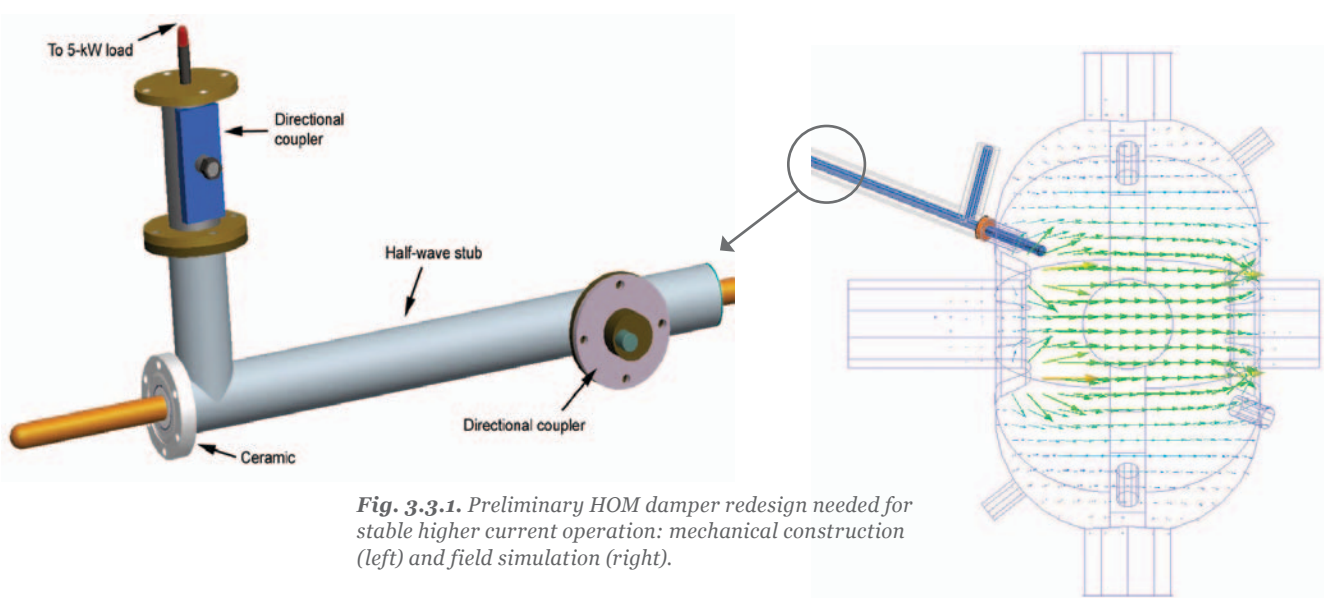


Fig. 3.3.1. *Preliminary HOM damper redesign needed for stable higher current operation: mechanical construction (left) and field simulation (right).*

OTHER NEEDED COMPONENT UPGRADES

Vertical scrapers are installed in the storage ring and are used for beam diagnostics and beam collimation, as needed. The beam-induced heating of these has been analyzed, and the cause found to be longitudinal HOMs . A new design has been developed to eliminate the resonant cavity in the existing chamber, thereby reducing the longitudinal loss factor (and power dissipation) by a factor of six. The present ID transition is a linearly tapering transition between two ellipses. A special non-linear transition between the aperture limits was designed (Podobedov et al. 2007) and could be implemented on all IDs. This will allow either an increase in single bunch current or a decrease in chromaticity and, therefore, will result in improved lifetime.

3.4 STABLE X-RAY BEAM DELIVERY

Cutting edge experiments at the APS place high demands on x-ray beam delivery systems. The wide range of experiments requires beam stability across multiple spatial (nanometer to micron) as well as time (microsecond to day) domains. Increasing the stability of delivered x-ray beams improves the quality of all APS science as it reduces jitter and beam-induced artifacts in experimental data and makes more challenging experiments possible. Due to the long distances inherent in x-ray beamlines, consistent improvements in beam stability are achieved by mitigating the largest contributions of instability from the storage ring down through the x-ray optical train.

LONG-TERM THERMAL STABILITY

At the APS, photon-beam instabilities at the hour to day level are primarily governed by thermal expansion of the supports that carry the storage-ring beam position monitors. Small temperature drifts in the 105 support stands for the insertion device vacuum chambers, which carry the rf storage ring beam position monitors as well as the 140 x-ray beam position monitors in the front end, lead to systematic beam position errors on the experiment hall floor. Temperature stabilization of these support stands to a level of around ± 0.1°C should provide 1 micron peak-to-peak beam position stability with 0.5 microradian peak-to-peak pointing stability over a one-week period. Fluctuations in the tunnel air temperature are a major component of the long-term drift. A recent energy savings project had the intended side benefit of improving the stability of the air temperature in the storage ring. However, there is still significant drift in the overall temperature (fig. 3.4.1).

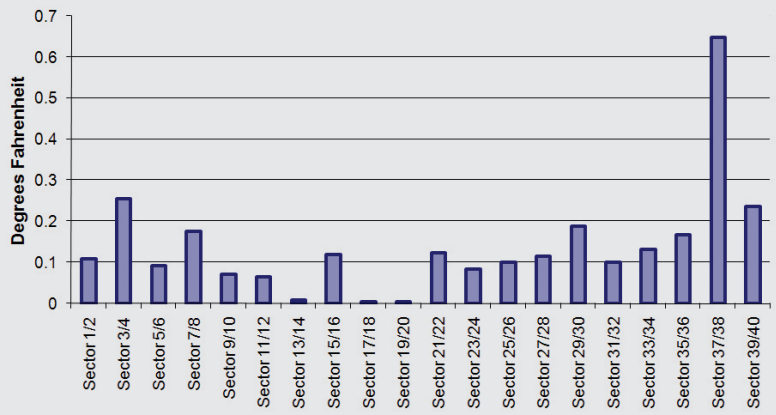
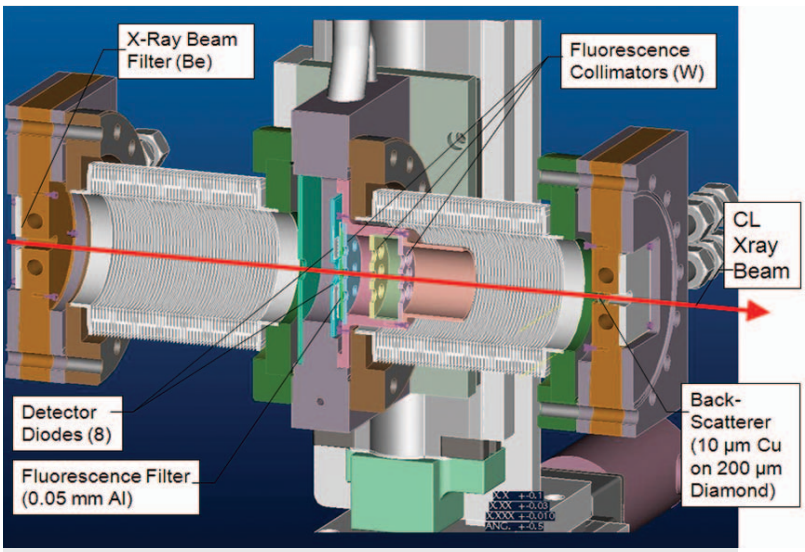


Fig. 3.4.1. *Fluctuations in storage-ring temperature over a one-week period, shown by sector. The target for improved stability is better than 0.1°C (0.2°F) over longer periods.*

Enhanced temperature stabilization techniques will be used to limit and control long-term drift in the most cost-effective manner. This effort may involve a hybrid solution of further controlling the air temperature along with local thermal stabilization techniques on the actual stands.



Feedback stabilization of the x-ray beam

Fig. 3.4.2. *Prototype fluorescence detector measures the beam intensity passing through the exit mask at the entrance to the beamline where the x-ray density and power are high. Eight photodiodes positioned around the beam measure the fluorescence. Since the hard x-ray core of the beam produces the copper fluorescence, the signal can be used to help steer the beam through the exit mask as well as determine the beam position and stability.*

BEAM DELIVERY DIAGNOSTICS

Currently deployed storage-ring beam position monitors do not intercept the x-ray but use some aspect of the "fringe" or spray around the beam. The development and global deployment of a hard x-ray beam position monitor on the insertion device beamlines will provide direct information on beam position and stability (fig. 3.4.2). Hard x-ray beam position monitors will be installed in the beamlines and provide a diagnostic tool to machine physicists as well as beamline scientists in improving short-term as well as long-term beam stability.

IMPROVED STABILITY THROUGH UPGRADED BEAMLINE OPTICS

The stability of the x-ray beams is strongly affected by the optics on the beamline. Increased power loading from higher current operations will exacerbate long-term drift effects as mechanical systems come to thermal equilibrium in accordance with the higher power levels. Significant refurbishment or replacement of optical components on many beamlines will be required to continue to deliver cutting-edge science. Recent engineering advances utilizing ideas developed for nanopositioning applications have been applied to optical components, resulting in extremely high stability as compared to older designs. For instance, the extremely high mechanical stability of a flexure-based crystal assembly combined with highly focused optics has been shown to significantly improve the results of x-ray photon correlation spectroscopy experiments (Narayanan et al. 2008). As part of the renewal plan, we propose to support appropriate improvements at all APS beamlines, whether facility operated or independent. The plan recognizes that some CATS will require expertise for design and maintenance of beamline components and other technical instrumentation.

3.5 SHORT-PULSE X-RAYS

To meet the scientific demand for better time resolution at the APS, we are proposing to implement a superconducting radio-frequency (SRF) deflection technique to generate x-ray radiation pulses on the order of 2 ps (FWHM) or less. Users performing state-of-the-art ultrafast dynamics studies have stated their need for high rf pulse repetition rates, including the continuous wave (CW) mode where every pulse from the storage ring is shortened. In the concept we are pursuing (Zholents et al. 1999), illustrated in fig. 3.5.1, transverse deflecting rf ("crab") cavities are used to impose a correlation (chirp) between the longitudinal position of an electron within the bunch and its vertical momentum. The x-rays emitted by each electron tend to travel along the path of the electron itself. Hence, by placing an undulator just downstream of the crab cavity (for example), we can produce a chirped x-ray beam. At a sufficient distance (e.g., 30 m) from the undulator, this angular chirp evolves into a spatial chirp, so that a strong correlation appears in the x-ray pulse between arrival time and vertical coordinate. Use of vertical slits then permits filtering the pulse in the time dimension, so producing an x-ray pulse that is shorter than the electron pulse.

As shown in fig. 3.5.1, two cavities are required, the purpose of the second cavity being to cancel the effects on the electron beam of the first cavity, so that the scheme does not degrade the source for other sectors in the storage ring. The second cavity must be placed at a vertical betatron phase advance N*180 degrees downstream of the first, where N is an integer. Given the normal properties of the APS lattice, this is readily accomplished by having the cavities separated by exactly n sectors and making slight adjustments to the electron beam optics. If N is greater than 1, the time-angle chirp of the electron beam will reappear at all intervening straight sections. This arrangement allows chirped pulses to be delivered to multiple beamlines.

Feedback stabilization of the x-ray beam

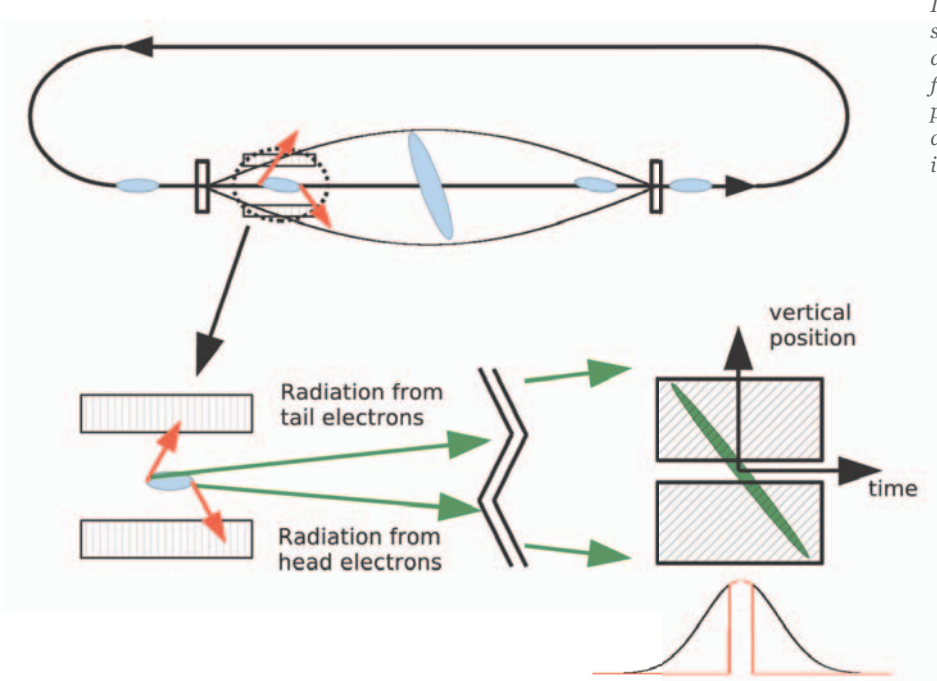


Fig. 3.5.1. *Illustration of the scheme (Zholents et al. 1999) proposed for short x-ray pulse production using deflecting rf cavities in a storage ring.*

Ideally, the second cavity exactly cancels the effect of the first. In real systems, nonlinearities, errors, and other details (Borland 2005) destroy the perfect cancellation of the kicks, resulting in vertical emittance growth. Vertical emittance together with the natural opening angle of the undulator radiation results in smearing of the chirp, which limits how short a pulse may be produced. As the deflecting voltage is increased, the emittance growth also increases and reduces the anticipated benefit. As a result, voltages above about 4 MV are of little value. Given this information, studies have shown that a drive frequency for the deflecting cavities of 2.8 GHz (corresponding to the 8th harmonic of the ring rf frequency, 352 MHz) will produce pulses less than 2 ps at 10 keV and above.

Pulse length versus photon energy

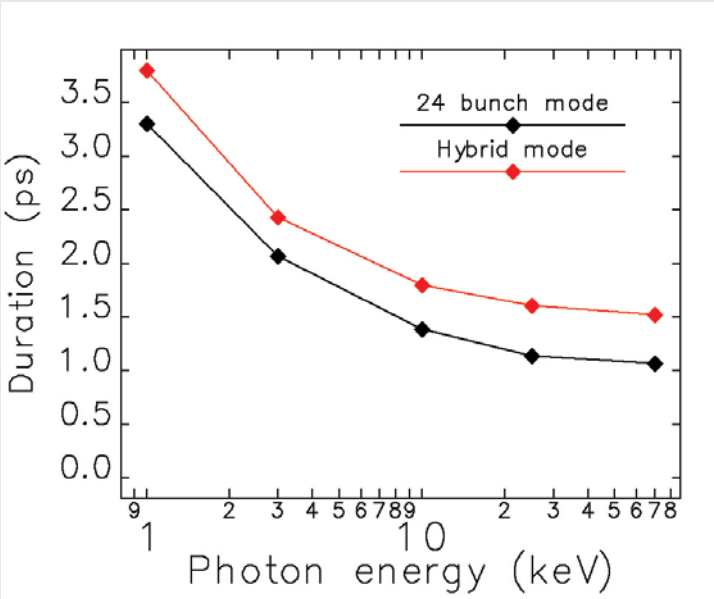


Fig. 3.5.2. *Predicted x-ray pulse duration (central 70%) as a function of the photon energy assuming 4 MV deflecting voltage and 1% transmission through the slits. "24B" is for 24-bunch operating mode, while "Hybrid" is for the intense bunch in hybrid mode. We see that the achievable pulse duration increases as the photon energy decreases, a result of the larger intrinsic opening angle of softer radiation.*

Figure 3.5.2 shows the results of a simulation of the expected SPX bunch length using a vertical slits 30 m from the undulator set to transmit 1% of the incident photons. The use of a slit will, of course, discard considerable intensity. Another option (Zholents et al. 1999) is to use asymmetric-cut crystals to perform pulse compression. We initially plan for the slit approach which is used for flux projections (fig 2.3.2), but will do further R&D on compression optics to deliver a higher flux beam to the experimenter.

A primary purpose of the R&D described in section 6.1 is to optimize the design and performance of the SPX deflecting cavity system and assure, through thorough testing, that the system will be transparent to other users around the ring.

4. NEW AND UPGRADED BEAMLINES

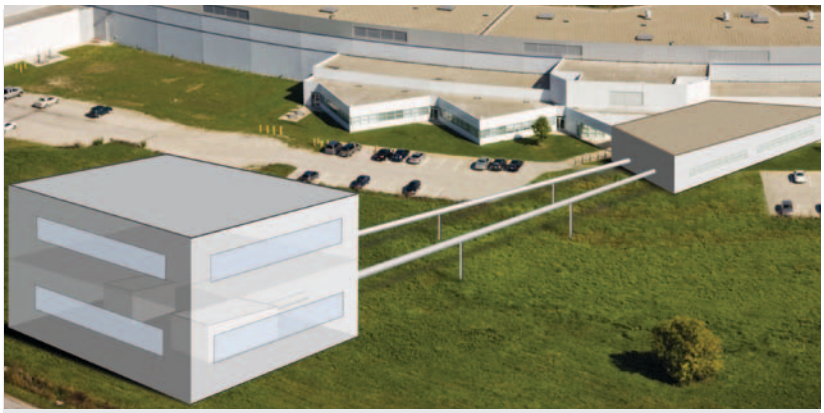
This section highlights some of the new and upgraded beamlines that support the scientific drivers in section 2, following the same six categories. Beamline proposals are in various stages of development. In some cases, full proposals that included preconceptual designs (e.g., X-ray Interfacial Science Facility, the Bionanoprobe, and Advanced X-ray Imaging beamlines) have already been submitted to the APS and discussed by the APS Scientific Advisory Committee (SAC). Other proposals are in a less mature state at this time and will require further refinement and optimization through discussions with APS users and staff before proposals and conceptual designs are finalized as discussed in section 8. In this section we do not provide extensive details. (More technical details on many proposals can be found at our website www.aps.anl.gov/Renewal/). Not all of the proposals found there will be included in the renewal. The initial cost estimate in the beamlines category of Table 8.3 comes from a portfolio of large and small developments which include the major new beamlines mentioned below, and many other appropriate upgrades from the technical proposals submitted last year in our renewal planning. The choices made reflect the science drivers for the renewal. Note that insertion device and front-end/delivery optics costs are included in the accelerator systems renewal budget line.

We will work with the SAC and technical advisory committees during the conceptual design phase of the project, as we develop with users a detailed set of the beamline upgrades that will be included in the renewal.

4.1 IMAGING AND COHERENCE BEAMLINES

LONG IMAGING BEAMLINES

To provide a large illuminated area with substantial coherence, needed for phase contrast imaging and related techniques, it is necessary to increase the length of a beamline beyond the space available within the experimental floor. The Advanced X-ray Imaging (AXI) beamlines at APS (fig. 4.1.1) will establish powerful new, dedicated x-ray imaging capabilities not currently available in the United States: a 200-m long undulator beamline for high-sensitivity wide-field-of-view x-ray imaging (WFI) and a long undulator beamline for coherent diffractive x-ray imaging (CDI). The WFI beamline will enable imaging at micrometer spatial and 100-ps temporal resolution, with a field size of a several centimeters. The large image field will greatly expand the



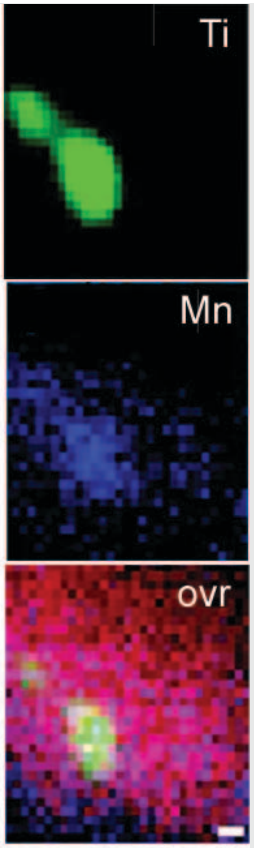
Long beamlines planned for APS

Fig. 4.1.1*. Shown at sectors 19 and 20, which are well positioned for beamline extensions of ~ 200 m in length, together with a separately proposed Imaging Institute, which is being discussed with the State of Illinois as a matching contribution to the renewal project. The Institute could house the long imaging beamline end stations and complementary imaging capabilities and expertise. The only remaining open space adjacent to the experiment hall is shown at left and would be used for beamlines needing an external end station (e.g. dynamic shock compression and very high magnetic fields).*

size and types of animals and specimens that can be examined, leading to breakthrough research in evolutionary biology, physiology, medicine and agriculture. The 100 ps time resolution of the WFI beamline will support critical advances in engineering of materials and industrial products, with applications ranging from materials failure, to fuel injection technology, and fluid dynamics. The CDI beamline will be the first dedicated facility of its kind for nanometer resolution x-ray imaging of the interior of thick samples, buried structures, and interfaces with elemental, chemical, and magnetic state sensitivity. In complement to the larger length scales addressed by WFI, the CDI beamline will enable ground-breaking research at the nanoscale in the structural biology, materials, and condensed matter sciences, including intact eukaryotic cells and collagen fibers, domains and domain transport in magnetic and multiferroic systems, catalytic properties of nanomaterials and nanoparticles, and strain and defects in nanocrystals. The AXI beamline concepts capitalize on the enhanced capabilities of the APS planned for the renewal, such as brighter undulator sources and high-speed computer networks.

BIONANOPROBE

The hard x-ray nanoprobe operated jointly with the Center for Nanoscale Materials has been highly oversubscribed with experiments from many scientific disciplines needing its 30nm resolution. Because of the importance of metals in cell biology, and the need for additional nanoprobe capacity and a specially-designed instrument for biological applications (including cryopreservation), a proposal for a "bionanoprobe" has been developed to address problems in the areas of bioremediation, climate change, biofuels, microbial interactions with contaminants, infectious and neuro-degenerative diseases, as well as novel nanovetor-based therapeutic drugs such as in fig. 4.1.2. Our understanding of diseases and of environmental contaminants is being significantly improved by our ability to image directly relevant metals in the context of tissues, single cells, and organelles. The possibility of resolving in yet more detail the specific interactions and motion of metals and contaminants at cell and vesicular membranes will lead to a yet greater understanding, with likely direct applications not only in areas such as bioremediation, microbial interactions with contaminants, and infectious and neuro-degenerative diseases, but also in climate-related studies (See also fig. 2.1.5.) .



Targeting nanoconjugates within a cell

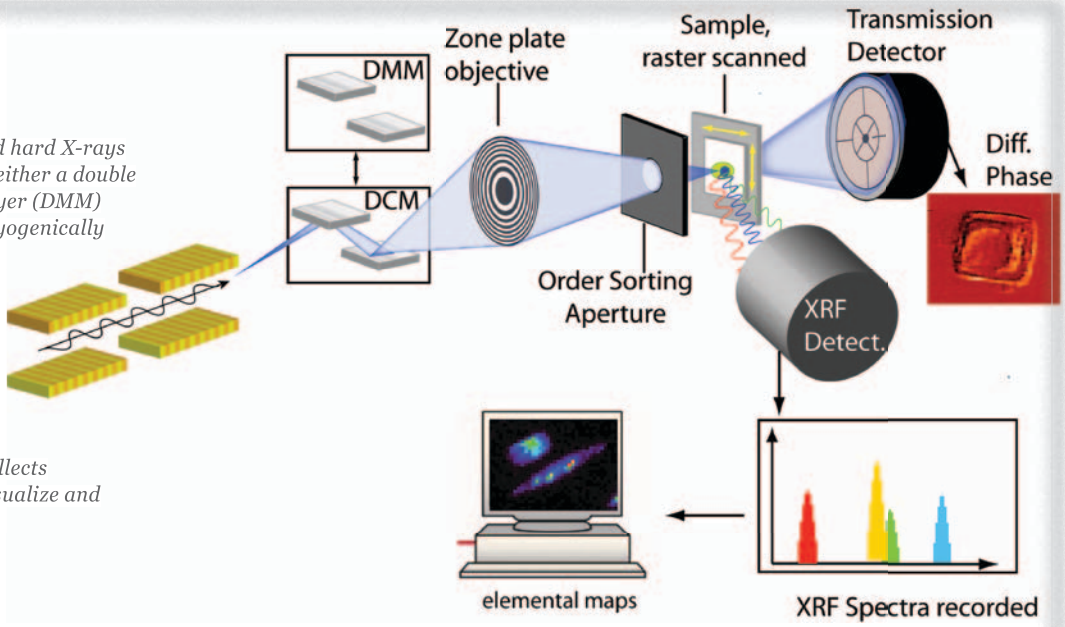
Nanoconjugate targeting has significant biomedical potential, permitting localization of nanoparticle-bearing therapeutic and imaging molecules to specific cellular and intracellular sites.

Fig. 4.1.2. *X-ray fluorescence microscopy maps of single mitochondria inside a PC12 cell, 48 hours after it was transfected with mitochondria-specific nanoconjugates (Paunesku et al. 2007). From top to bottom: Ti (green, 0.04-1.76 μg/cm^{-2}), which shows the nanoconjugates; Mn, revealing the mitochondria (blue, 0.0 – 0.04 μg/cm^{-2}); and an overlap (ovr) map of Ti, Mn, and S (red). The scale bar is 300 nm. The bionanoprobe will improve the sensitivity and resolution of this important technique by more than ten fold, enabling study of smaller organelles.*

The bionanoprobe (fig. 4.1.3) will improve the achievable spatial resolution on soft specimens to 20 nm, and, crucially, provide a cryogenic sample environment for optimum specimen preservation. We expect an improved absolute sensitivity by >2 orders of magnitude (through higher spatial resolution and large-area multi-element x-ray fluorescence detectors), so that in

Schematic view of the bionanoprobe

Fig. 4.1.3. *Undulator-generated hard X-rays will be monochromatized using either a double crystal (DCM) or double multilayer (DMM) monochromator. The sample (cryogenically preserved, in vacuum) is raster scanned through a 20-nm focus spot created by a Fresnel zone plate. A multielement silicon drift detector system records full x-ray fluorescence (XRF) spectra at each scan position; a dedicated segmented detector collects differential phase contrast to visualize and quantify specimen structure.*



ideal bio-specimens as little as 5-10 atoms will be detectable. Together with fast detector electronics, a dedicated differential phase contrast detector (for projection alignment), and the principle of dose fractionation, we will then be able to routinely image metal distributions in cryogenically preserved, whole, cells in 3D at ~20nm, within tens of minutes, as opposed to tens of hours.

XPCS

We will upgrade out XPCS capabilities at sector 8 to realize specific gains in coherent flux as follows: a 4.8 m long undulator with a 2.7cm period (gain by a factor of 4), increased current (gain by factor of 2) and efficient vertical focusing (gain by a factor of 10) for a total gain of 80X. Users will also benefit from improvements in rapid correlation function data reduction that is a bottleneck for XPCS experiments. Importantly, the upgraded APS XPCS facility will be unique in being able to extend these same gains to higher x-ray energies that are out-of-reach at lower-energy (i.e. 3 GeV) sources to facilitate, for example, measurements of biologically relevant lipid layers under water and to reduce sample beam damage. Also, with respect to XPCS at future free-electron lasers (FEL's), the upgraded XPCS beamline at the APS will permit sample dynamics to be studied at the nanoscale in the physically important time scale spanning μs to ms. Because of the low repetition rates of FEL's and limitations on pulse delay lines, XPCS time scales accessible at the APS after the upgrade will complement those accessible at FEL's. The proposed upgrades will enable studies of dynamics within complex mesoscopically structured fluids and in biological systems, relevant to how small particles self-assemble into potentially useful structures. The dynamics of solid supported membranes, diffusion and surface fluctuations in free floating vesicles, and Brownian motion within concentrated protein solutions are examples which utilize higher-energy. Such studies will provide crucial insight into the central role of thermal fluctuations in biological activity at the cellular scale.

COMPLEMENTARY IMAGING CAPABILITIES

We propose to expand capabilities in complementary microscopy tools (for correlative imaging) in collaboration with Argonne's Center for Nanoscale Materials (CNM) and the Electron Microscopy Center (EMC) and to use the Imaging Institute for additional laboratory space. Imaging is a major strategic direction for local universities, including the University of Chicago, Northwestern University, and the University of Illinois, and we have been planning joint activities with them. In particular, these institutions are strong in biomedicine, very relevant to a major application of our imaging tools in hierarchical systems. Specific instruments we would like to acquire include cryo-TEM and confocal optical microscopy, to supplement existing capabilities at CNM and EMC.

4.2 EXTREME-CONDITIONS BEAMLINES

FACILITY FOR MATERIALS STUDY IN HIGH-MAGNETIC FIELDS

High magnetic fields can influence the interactions of electron spin and charge with the lattice, giving a unique handle on important material properties such as superconductivity and quantum phase transitions. The National Academies study (Committee on Opportunities in High Magnetic Field Science 2005) concluded that "development of new high-field capabilities at x-ray and neutron scattering centers in the United States could have enormous scientific impact" (p. 5). We propose to develop a new dedicated high-field beamline offering both a cryogen-free ~20 T superconducting and a pulsed 60 T instrument (fig. 4.2.1). A set of split-pair and solenoid geometries are envisioned to fully utilize photon-atom interaction cross sections as required to probe various systems. Furthermore, we will explore with the National High Magnetic Field Laboratory (NHMFL) in Tallahassee and its sponsor, the NSF, the possible establishment of a high-field hybrid DC magnet reaching well beyond 30 T. We have been discussing a split-gap design for this high-field magnet with NHMFL (which has also helped us with our preliminary work on superconducting insertion devices). We note that such a dedicated beamline will provide unique opportunities in the western hemisphere. To take full advantage of these opportunities the beamline will have the flexiblility to accomodate the widest range of techniques. We envision the beamline will include a large energy range, comprehensive polarization manipulation, and adjustable focusing.

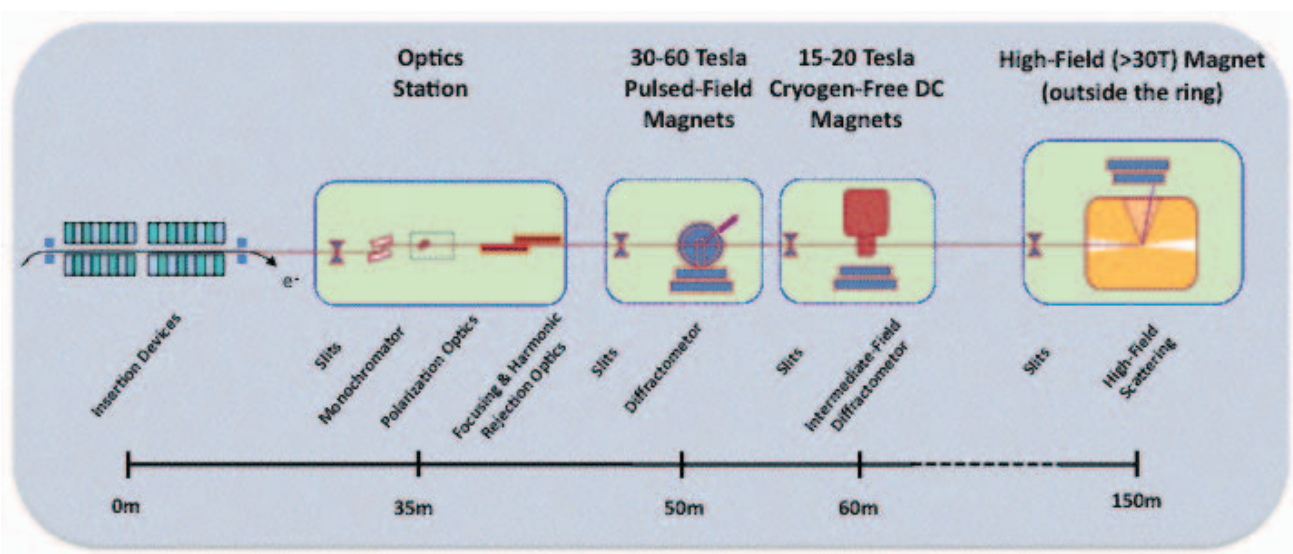


Fig. 4.2.1. Schematic layout of the high magnetic field beamline.

DYNAMIC SHOCK COMPRESSION BEAMLINE

Following a test experiment at the HP-CAT (fig. 4.2.2), a proposal has been made for a dedicated dynamic shock compression beamline. The DOE's NNSA is very interested and has indicated willingness to pay for construction and share in operation costs. We believe that the case for further capabilities in this area is strong, so that we are including it in our planning. If NNSA funding is unavailable, we plan to develop a more modest facility in an existing end station, utilizing high-power lasers as the trigger.

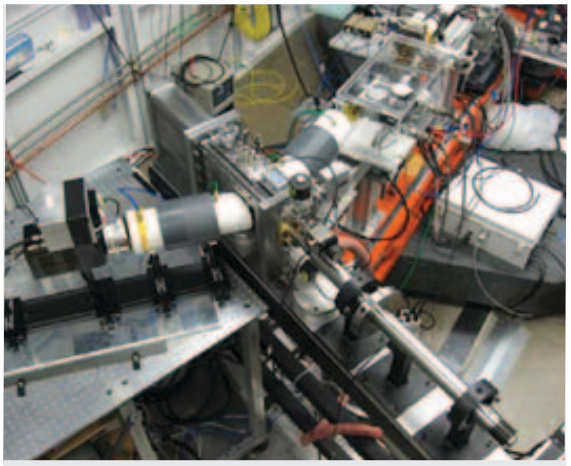


Fig. 4.2.2. *Setup for a shock-wave compression experiment at APS in 2007. The experiment was successful in observing shock compression approaching 5% in LiF, Cu, and Al (courtesy Y. Gupta, unpublished).*

IMPROVEMENTS TO DEDICATED HIGH-PRESSURE BEAMLINES

Sectors 13 (GSE-CARS) and 16 (HP-CAT) are dedicated facilities that are the most productive high-pressure (HP) beamlines on the APS floor today. Both ID beamlines have requested canted undulators, with optimized insertion devices and new focusing optics to improve their ability to access higher extreme regimes, add new techniques, and increase capacity. A factor of 10-30 increase in brilliance is anticipated for dedicated HP lines during the APS renewal. These improvements are prerequisite for developing other novel HP synchrotron techniques, including submicron HP diffraction, submicron HP imaging, Mbar single-crystal diffraction, Mbar HP spectroscopy, high-resolution HP x-ray diffraction, and time-resolved HP synchrotron instrumentation. These techniques will each open an entirely new branch of HP science that does not yet exist or is currently in its infancy. There are many high-pressure techniques that remain flux limited, not only spectroscopic techniques such as HERIX, MERIX, and x-ray Raman, but also diffraction from non-crystalline low-Z materials. Improvements at HP-CAT and GSECARS will greatly expand capability in these areas also.

The renewal improvements allow further reducing the x-ray beam for HP experiments into the sub-micron range. It took three decades to reduce the x-ray beam size from 50 μm to the current 5 μm in HP research. Yet, the impact of this one order-of-magnitude reduction is tremendously significant. While beam sizes of a few tens of nanometers have been reached in specialized beamlines, a beam size of 5 μm is still typical in all leading HP synchrotron beamlines in the world. The enabling of sub-micron beams, optimized and dedicated for HP research, will allow us to probe small sample sizes at ultrahigh pressures approaching the terapascal regime; to investigate grain-to-grain interactions of composite materials by three-dimensional submicron probes (fig. 4.2.3); to map out the grain boundary, phase boundary, chemical ordering, local stress/strain and structure evolution within the individual grain; and to study nanoscale single crystals. The combined orders-of-magnitude improvements from improved (1) x-ray brilliance, (2) spatial, angular, and temporal resolutions, and (3) detector speed and resolution will provide the superior tools for the HP community to lead the next level development of HP energy science in the foreseeable future.

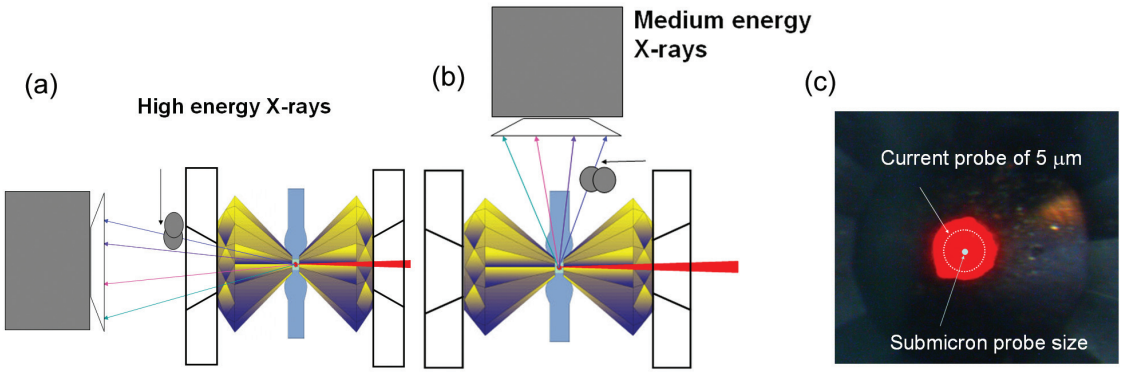


Fig. 4.2.3. *Illustrations of 3D diffraction at high pressures with sub-micron spatial resolution, (a) in the forward geometry with high energy x-rays, (b) in perpendicular geometry with medium energy x-rays; (c) shows an image of the HP sample chamber (red) relative to the size of the current probing beam size (5 μm) and future submicron beams.*

COORDINATED HP SCIENCE BEYOND THE HP BEAMLINES

HPSynC is currently a jointly funded entity that supports four staff members with a mandate to make the pressure variable available across many APS beamlines and to support users of these new capabilities. This approach recognizes that the growth of synchrotron science integrated around the entire APS ring progresses far faster than could be accommodated by a dedicated HP beamline. HPSynC provides a novel mechanism to couple HP science with cutting-edge synchrotron techniques and, thus, forms a valuable complement to "traditional" dedicated HP instruments. As one example, the expertise in 3D tomography on imaging beamlines at the APS is now being developed as a method of measuring equations of state in amorphous materials. This activity builds on the pioneering work of Liu et al. (2008), as shown in fig. 4.2.4) and with renewal improvements the precision of volumes measured in this way can be increased to 0.1% rivalling that achieved by diffraction in crystalline materials. It seems likely that the APS will be at the forefront of an entirely new field of HP imaging.

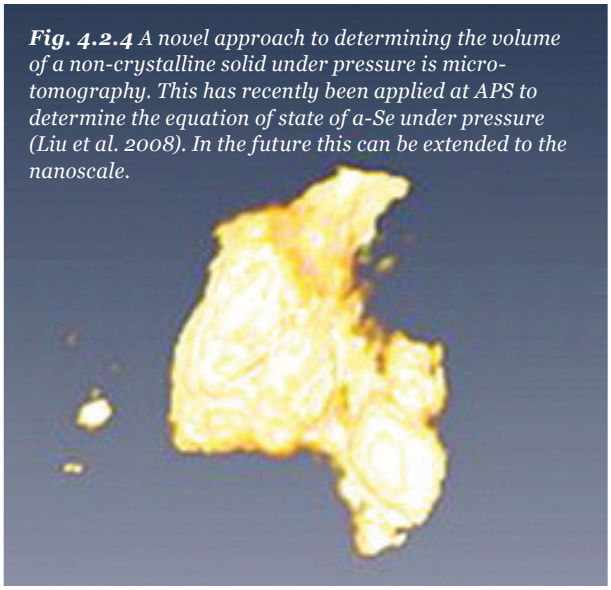


Fig. 4.2.4 *A novel approach to determining the volume of a non-crystalline solid under pressure is micro-tomography. This has recently been applied at APS to determine the equation of state of a-Se under pressure (Liu et al. 2008). In the future this can be extended to the nanoscale.*

In a revitalised APS, an HPSynC with expanded scope would continue to coordinate and extend the suite of capabilities, instruments, and user support available for experiments under extreme conditions. Ongoing innovative technique development will optimize data quality, extend capacity to higher pressures and temperatures, and embed support for combined multiple extreme environments across diverse beamlines.

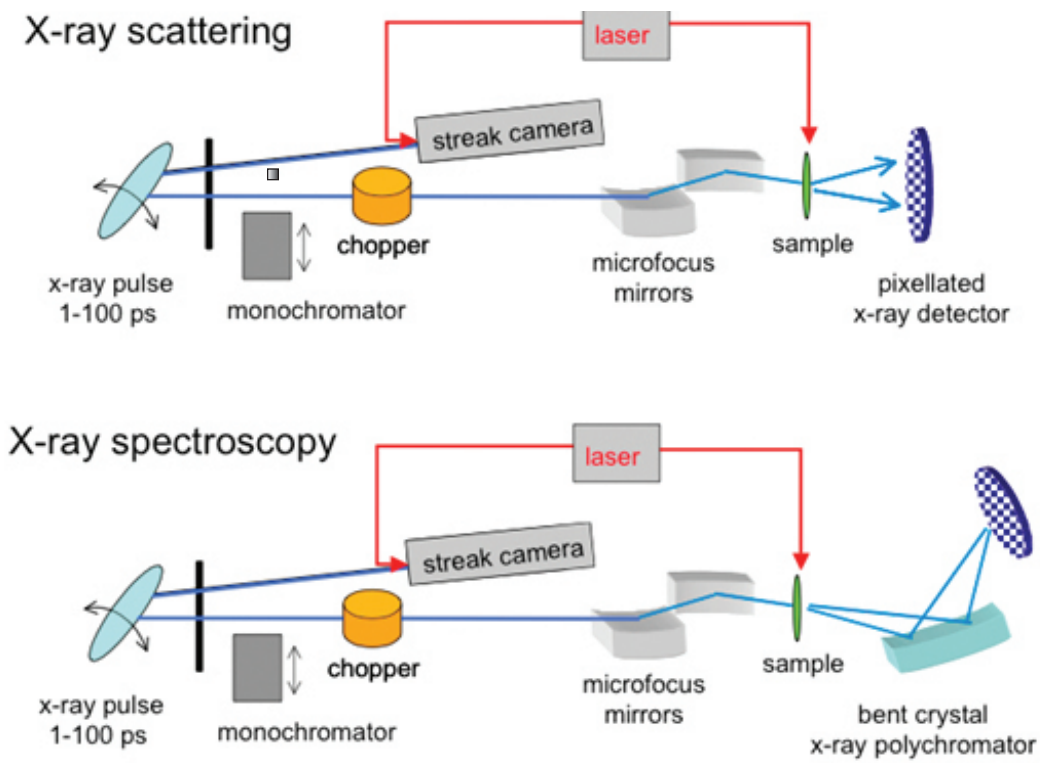
4.3 ULTRAFAST DYNAMICS BEAMLINES

SPX BEAMLINES

The crab cavity scheme (sections 3.5 and 6.1), offers a unique tunable high average flux source of ~1ps x-rays. We propose to develop two beamlines that would be fed by the SPX pulse slicing scheme. R&D on the SPX source (described in 6.1) is required before beginning its construction. Beamline designs have been considered. High-power lasers for pump-probe experiments using accurate sub-ps time synchronization are being developed as extensions of our existing ultrafast science programs and through collaborations with LCLS. The schematic in fig. 4.3.1 displays beamline configurations for scattering and spectroscopy.

Schematic of short-pulse x-ray beamlines

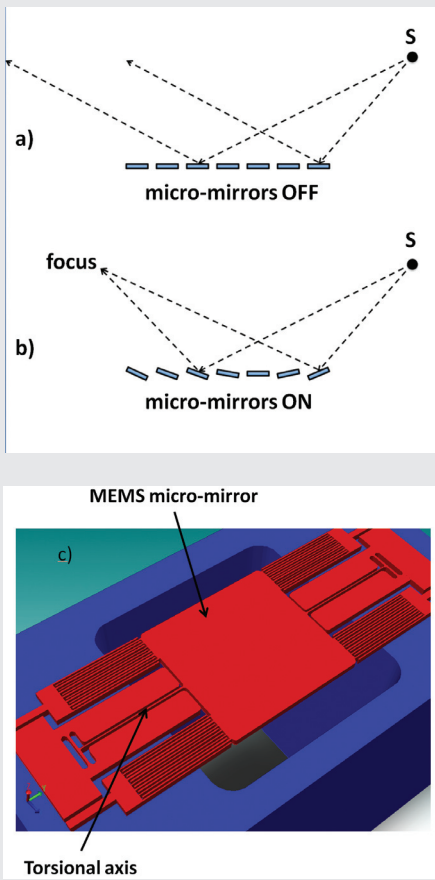
Fig. 4.3.1. *The source for these beamlines is the crab cavity slicing source described in section 3.5.*



We anticipate that we will have two insertion device beamlines and one bending magnet beamline that will have access to the chirped source. With two sectors the intervening bending magnet beamline can be instrumented for ultrafast experiments where brilliance is less important than flux. We anticipate that to satisfy the science needs described in Section 2.3, the undulators for the two ID beamlines will be chosen so that one beamline is optimized for higher-energy, and one for lower-energy experiments. During the R&D phase for the SPX source, we will develop detailed designs and work with users from the ultrafast community to accommodate high-impact experiments.

Micromirrors for adaptive optics and fast switching

Fig. 4.3.2. *The 1-D array of micromirrors shown (a and b) would be fabricated by MEMS technology. As an adaptive optic it could be used for dynamic optimization of the point-spread function. However, it also offers the potential of ultrafast switching of an x-ray beam. Image c) shows a finite element analysis of an individual micromirror.*



INSTRUMENTS FOR ULTRAFAST SCIENCE

High-performance lasers are key to pump probe experiments. We plan to take advantage of improvements in high rep-rate high power laser technology. In the detector program (section 5.1) we plan to develop a suite of ultrafast detectors for imaging and spectroscopy. Several beamlines at APS are already involved in ultrafast experiments or expect to move in that direction. Those experiments that do not require less than 100-ps time resolution are better suited to avoid the SPX sectors because the flux is attenuated by the pulse slicing. Streak cameras and other approaches can be used to reach below 100-ps time resolution. An innovative optics approach with potential for ultrafast studies is shown in fig. 4.3.2.

The APS is well-suited for ultrafast imaging studies on non-SPX beamlines, especially the planned new imaging beamlines (Section 2.1). Attractions include the flexible timing modes which allow for high-speed imaging ranging from 100 ps single exposures to MHz video rates, the wide energy range (10-100 keV) allowing for the study of soft (e.g. water) as well as hard (e.g. stainless steel) materials, and the large number of photons per pulse. There are many areas in science where ultrafast imaging by existing users has provided unique insights, both fundamental and applied, such as free surface flow singularities including droplets merging, pinch-off, and impact on liquids/solids, and the industrially-important process of liquid jet atomization.

Despite our state of the art detection schemes, we are limited to a detective signal to noise ratio of about 12 when using the 100 ps pulse without sample in the beam. The proposed upgrade to the accelerator systems, such as higher electron beam current, and specialized high-brilliance long insertion devices will provide 5 fold improvement in the S/N. Added to the wider field of view afforded by a long dedicated beamline, this will gain us access to more realistic sample sizes and materials of interest as mentioned above. Most importantly the proposed upgrade promises to deliver single-pulse, single-shot imaging using all the x-ray pulses in the standard 24-bunch mode, which translates into MHz frame rate for real-time dynamics studies of non-repeatable transient phenomena such as catastrophic transformations (materials failure, explosions, and rapid chemical reactions). Detector developments summarized in sections 5.1 and 6.4 will be very important. The resulting unprecedented capabilities will put the APS at the forefront of high-speed imaging.

4.4 BEAMLINES FOR INTERFACES IN COMPLEX SYSTEMS

X-RAY INTERFACIAL SCIENCE FACILITY

Interfacial science by its very nature brings together a diverse community with interests in catalysis, biomembranes, oxide film growth, semiconductors, geochemistry, surface physics, corrosion, nano-science, tribology, and electrochemistry. With close involvement of external users (through a Collaborative Development Team) we plan to build an innovative, dedicated X-ray Interfacial Science (XIS) facility (fig. 4.4.1) at a sector of the APS that will fully exploit the unique capabilities of the APS to advance our understanding of interfacial science. The XIS facility will provide state-of-the-art X-ray scattering, spectroscopy, and microscopy tools for multi-length-scale (from angstroms to microns), element-specific studies of vacuum/solid, gas/solid, liquid/solid, solid/solid interfaces and supported nanostructures. The proposed facility has two optimized, canted undulators producing four separate x-ray beams: one with variable-energy and three with selectable fixed energies. In addition, the bend magnet port will be developed to support those measurements that do not require the brightness of the ID beam lines. The tandem experimental hutch arrangement includes both general-purpose diffractometers and custom-designed spectrometers to provide an *in-situ* controlled environment accomodating growth chambers for various advanced materials synthesis activities involving molecular-beam epitaxy (MBE), metal-organic chemical vapor deposition (MOCVD), atomic-layer deposition (ALD), and pulsed-laser deposition (PLD), as well as a dedicated state-of-the-art interfacial X-ray microscopy spectrometer. Laboratory preparation and advanced off-line characterization facilities such as scanning tunneling microscopy (STM) and atomic force microscopy (AFM) are recognized as an important component of the XIS facility.

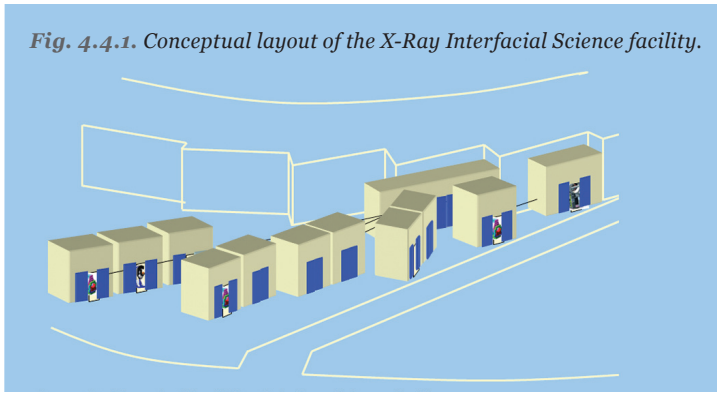


Fig. 4.4.1. *Conceptual layout of the X-Ray Interfacial Science facility.*

HIGH-ENERGY BEAMLINES

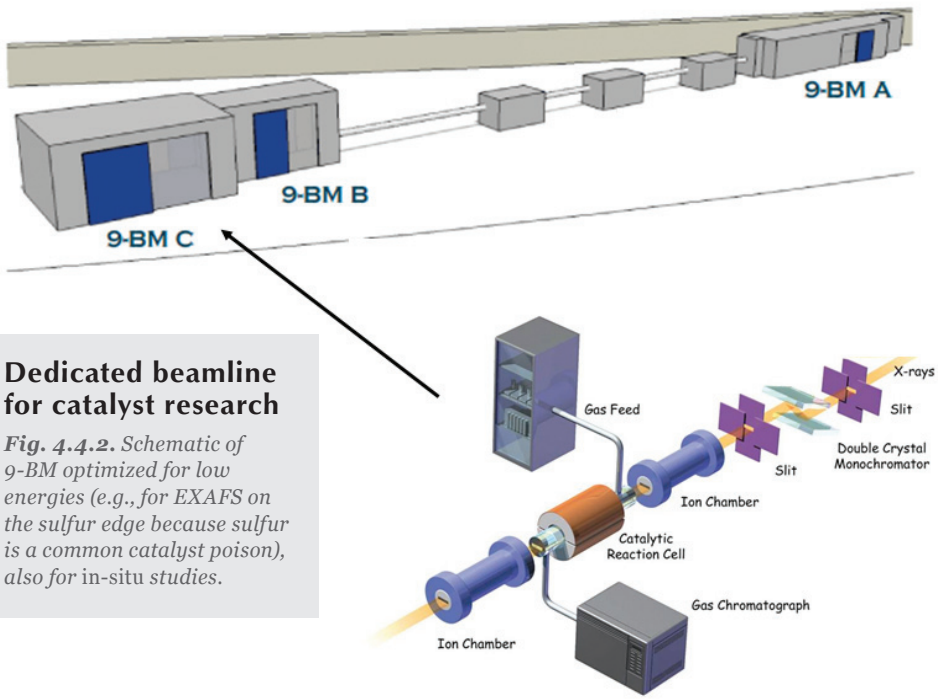
Our existing 1-ID beamline, the only beamline in the western hemisphere with energy tunability and high brilliance for photons in the range of 50-120 keV, is heavily oversubscribed for experiments in the areas of high-energy diffraction microscopy (HEDM), mechanical measurement of aggregate materials (i.e., stress, strain, texture), and microfocusing pair distribution function (PDF) measurements. We plan to replace the current monochromatic station with two larger white-beam capable stations and add a side branch beamline that can be used simultaneously with the main beamline. With this upgrade, the experimental instruments will be given dedicated space and, consequently, be optimized for significantly improved performance. We propose to develop a superconducting undulator (section 6.2) that will increase brilliance over one order of magnitude and to add improved optics and state-of-the-art area detectors. With the planned new configuration, both performance (100x) and capacity will be greatly improved because we obviate the need for moving and aligning key instruments. Upgrades are also planned for the high-resolution diffraction microscopy beamline at 34-ID-C, where a dedicated optimized undulator (at present one versatile undulator A serves two stations at 34-ID) and enhanced optics and detectors will improve the flux in a nano-focused spot and extend the mapping of strains to higher spatial resolution and greater

speed (allowing time-dependent measurements). 34-ID is complementary to 1-ID and has pioneered the technique of polychromatic micro-focusing (now nano-focusing) for mapping phases and the strain tensor at the nano-level. Software and computing infrastructure are recognized as an essential component of the 1-ID and 34ID-C beamline upgrades, because the analysis of complex HEDM data is a major barrier to expansion of the technique.

There is increasing demand for PDF data especially for nanocrystalline samples. PDF's must be taken at high-energies where the Q-range is suitably large. We plan to upgrade PDF capabilities at Sectors 11-ID and 1-ID.

CATALYSIS CAPABILITIES

The recommendations of a major user workshop on catalysis held at APS in 2005 were that we needed to establish a dedicated spectroscopy beamline on a bending magnet for *in-situ* catalysis studies and to make many other beamlines with other techniques available with expert assistance and a suite of mobile experimental equipment. Beamline 9-BM has been chosen as a focal point for catalyst research and will be upgraded for concurrent XAFS and x-ray diffraction on operating catalysts (fig. 4.4.2). XAFS and XANES techniques provide *in-situ* element-specific atomic-level chemical and geometric information on the structure of real working catalysts (both heterogeneous and homogeneous). *In-situ* synchrotron X-ray powder diffraction (SXRPD) is invaluable for structural analysis of catalytic materials. This central facility will complement other unique capabilities at the APS that will be applied to catalyst research, including techniques such as small angle scattering, PDF measurements, and inelastic x-ray scattering. A major component of this activity is to provide the nucleus of expertise and capabilities (such as sample cells) that can be transferred for use at these other beamlines and establish "CatSynch" as a gateway to facilitate this area of research.



Dedicated beamline for catalyst research

Fig. 4.4.2. *Schematic of 9-BM optimized for low energies (e.g., for EXAFS on the sulfur edge because sulfur is a common catalyst poison), also for* in-situ *studies.*

4.5 HIGH-RESOLUTION SPECTROSCOPY BEAMLINES

LERIX

Due to improved light source brilliance and novel detector development in only the last 5 years, non-resonant x-ray Raman spectroscopy (XRS) has become a powerful tool for examining low-energy loss features with hard x-rays. XRS gives a bulk-sensitive alternative to UV, soft x-ray, or electron spectroscopies, especially for situations where those methods are not applicable because of complex sample environmental chambers. This technique has been widely used in high-pressure research to examine light elements, and more recent work at APS has given the first demonstration that XRS is extremely valuable for *in-situ* studies of metal oxide battery electrodes (Balasubramanian et al. 2007) (see fig 2.5.2), as was projected in the 2007 DOE report on Basic Research Needs in Electrical Energy Storage (BESAC 2007). Other work at the APS has shown that the momentum-transfer (q) dependence of XRS can characterize dipole-forbidden final states in transition metal, rare earth and actinide materials. Building on the success of the existing LERIX spectrometer at the APS Sector 20 (see fig. 2.5.2 background), we plan to develop a dedicated capability for high-throughput XRS measurements. The new station will have more analyzers for enhanced detection efficiency, and better capabilities for *in-situ* sample environments such as catalytic reactors and high pressure cells. As demonstrated by the battery studies, the *in-situ* capabilities will be essential in further enabling basic energy science studies relevant for electrical energy storage, fuel cell operation, catalysis, and next-generation nuclear fission fuels.

MERIX

MERIX, the medium resolution (~100 meV) inelastic x-ray scattering spectrometer at APS 30-ID (fig. 4.5.1), is dedicated for the study of collective valence electron excitations in correlated electron systems. The incident x-ray photon energy range of MERIX is 5-12 keV. MERIX offers a unique set of x-ray analyzers optimized for 3*d* and 4*f* resonances. Understanding the polarization dependence of the scattering at resonance is crucial. This is addressed by the ability of the MERIX spectrometer to scatter both vertically and horizontally. The MERIX instrument is equipped with x-ray mirrors, providing a micro-focused beam on the sample (6 x 45 μm²), enabling studies of small samples and samples under high pressure. The instrument has been working very well (Wakimoko et al. 2009), but the scientific impact would grow substantially with planned upgrades to increase the energy resolution, the throughput of the spectrometer, and the flux on the sample; to decrease the beam size on the sample to sub-micron size; and to add new and improved analyzers.

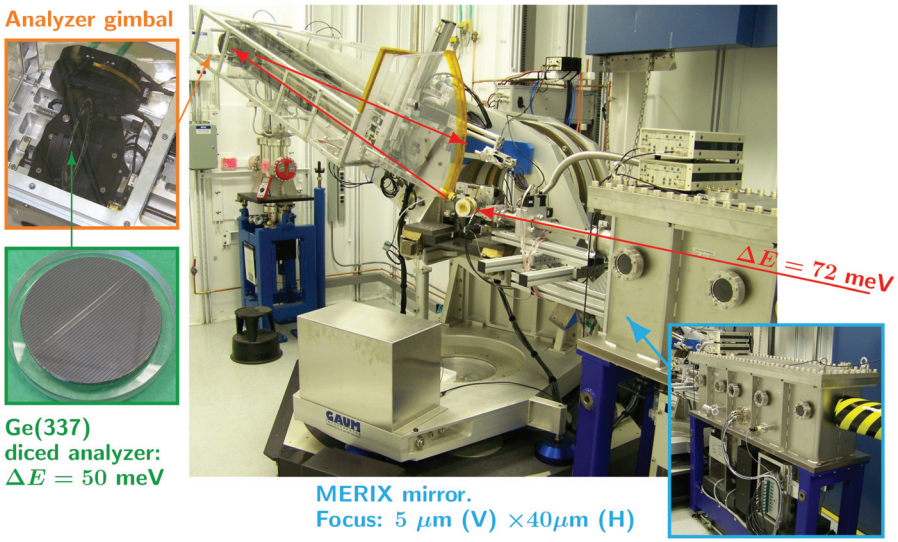


Fig. 4.5.1. *MERIX: Picture of medium-energy resolution (~ 100 meV) inelastic x-ray spectrometer at 30-ID with focusing mirror and analyzer crystals.*

HERIX

The HERIX spectrometer at 30ID is designed to study the phonon dispersion in crystals and the collective atomic excitations in disordered systems as a function of momentum and energy transfer. The HERIX spectrometer can from a micro-focused beam at the sample (15 x 34 μm^2), which allows the study of samples under extreme thermodynamic conditions. Also, the HERIX spectrometer has a high-energy resolution (1.5 meV), which enables the study of collective excitations in soft matter (fig. 4.5.2). Another advantage of the HERIX spectrometer is the ability to measure the dynamical structure factor at high momentum transfer (Q~75 nm^{-1}), which is crucial to determine the phonon density of state in polycrystalline materials as demonstrated recently on pure δ-Pu and δ-$Pu_{0.98}Ga_{0.02}$ at room temperature (Manley et al. 2009). A comparison of the phonon DOS of the two phases at room temperature allowed the conclusion that the heat capacity of α-Pu is accurately described in terms of the measured phonons and conventional electronic and thermal-expansion contributions. However, a comparison with the δ-phase demonstrates that most of the entropy stabilizing the δ-phase of plutonium at high temperature comes from unconventional sources, including electron correlations, intrinsic phonon softening, and possibly intrinsic localized modes. Options to increase the energy resolution of HERIX are being explored. One direction, ultra-high resolution resonant inelastic scattering (UHRIX) would open the realm of inelastic neutron scattering and Brillouin light scattering to x-rays (Shvyd'ko et al. 2006). A more modest approach, but a big step forward is the next generation IX spectrometer (ANGIX), which significantly increases the measured solid-angle and momentum transfer range.

At present the MERIX and HERIX instruments split beamtime on sector 30 and, while performing very well, would benefit from tailored insertion devices and higher flux and brightness. Our plans consider the separation of these instruments into two dedicated beamlines, each with tailored insertion devices. For example, the HERIX instrument would be ideally suited for a superconducting insertion device, providing an order of magnitude higher flux at 25-35 kV, which is desired for the highest energy resolution.

Shaking for action

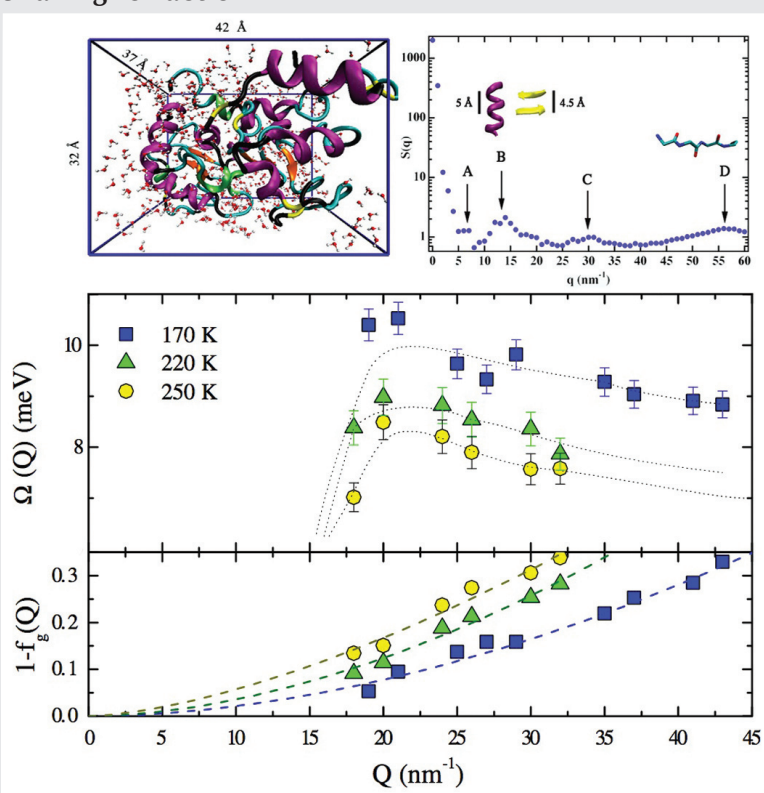


Fig. 4.5.2. HERIX spectra reveal a fascinating correlation between the temperature onset of biochemical activity of a protein and intramolecular vibrations (Liu et al. 2008).

X-RAY EMISSION SPECTROSCOPY

X-ray emission spectroscopy (XES) of the photons generated when valence electrons, or electrons in weakly bound orbitals, fill deep core holes is an experimentally versatile and reliable way to characterize the occupied local electronic states of a targeted atomic species. For example, the Kß and Kß" features for 3*d* transition metals often provide unique insight into the spin, valence, ligand species, and ligand bonding. Such information could be of critical importance for studies of bulk and nanophase transition metal oxides used in numerous energy science applications. In contrast, XANES is most directly sensitive to the unoccupied final density of states. Unfortunately, few XANES studies at hard x-ray energies are accompanied by XES measurements with resolution comparable to the core-hole lifetimes (e.g., 0.5-2 eV for 3*d* transition metals), even though many such studies would vastly benefit from the complementary information provided by XES. This neglect is due to the complexity, cost, and lack of portability of large-scale x-ray spectrometers based on spherically bent crystal analyzers. Recent work at the APS has successfully demonstrated a new type of "miniature" x-ray spectrometer (miniXS) that overcomes many of these issues, while also providing up to 50x improvements in measurements times compared to the existing APS apparatus (fig. 4.5.3). We plan to develop a portable x-ray emission spectrometer for operation at 3-10 keV. This energy range is of high scientific importance, as it spans the *K*-emission for 3*d* transition metals, the *L*-emission for lanthanides, and the *M*-emission for actinides. The spectrometer will be compatible with the beam characteristics of numerous APS beamlines, while also being easily integrated with important experimental apparatus, such as diamond anvil cells, laser systems for pump/probe studies, cryostats, furnaces, and x-ray compatible chambers for *in-situ* electrochemical or chemical studies.

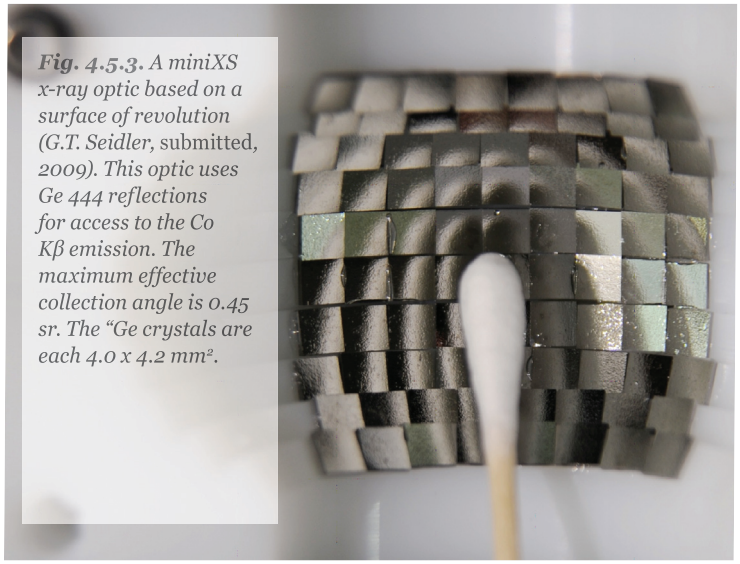


Fig. 4.5.3. *A miniXS x-ray optic based on a surface of revolution (G.T. Seidler,* submitted, *2009). This optic uses Ge 444 reflections for access to the Co Kβ emission. The maximum effective collection angle is 0.45 sr. The "Ge crystals are each 4.0 x 4.2 mm².*

4.6 INSTRUMENTATION TO UNDERSTAND THE CONNECTION OF PROTEINS AND ORGANISMS

Use of synchrotron radiation in life science research is expanding dramatically as researchers become aware of the capabilities of novel scattering and imaging modalities. These techniques range from solution scattering and fiber diffraction to phase-contrast and scanning imaging. In many cases, the APS is the only place in the western hemisphere with the capabilities to support this research. Future progress will be greatest at beamlines dedicated to a single experimental technique and equipped for the special needs of handling biological specimens. For example, high sensitivity of many specimens to radiation damage requires cryopreservation for measurement of relevant data. Studies of many variants under different conditions require high-throughput of specimens.

MICROCRYSTALLOGRAPHY

Microcrystallography was pioneered at the ID-13 of the ESRF. Several APS beamlines have already begun offering microcrystallography capabilities to users with beams sizes down to 5 microns. Other facilities around the world are also developing similar capabilities, but no beamline dedicated to macromolecular crystallography exists with a beam size smaller than 5 microns. (Smaller beams are available on a limited basis at the ID-13 of the ESRF.) The large unit cell parameters of crystals of proteins, viruses and macromolecular complexes constrains the optical design to be of low divergence. Typically, this implies long focal length optics resulting in a magnification of source motion due to imperfections in the optics. Maintaining the intersection of micron-sized beams and rotating, cryogenically cooled, micron-sized crystals is challenging. The APS renewal will allow the collection of data of high signal-to-noise ratio by improving the stability of the source, optical components, and endstation components by incorporating nano-positioning devices.

Efforts to validate the prediction of reduced radiation damage with micron sized beams mentioned in section 2.6 have been limited by the use of an energy-dependent, Fresnel Zone Plate focusing optic. Studies with a 1-micron, 15 keV beam show that the distribution of radiation damage about the 1-micron beam is anisotropic with more damage along the polarization vector. However, at 15 keV, the damage is observed to decay monotonically with distance from the beam center. No damage is observed beyond 4 microns (R. Fischetti, unpublished). The APS renewal will provide state-of-the-art, energy independent focusing optics allowing the high energy aspects of the APS to be exploited for further investigations of the possibility of reduced radiation damage with a 1-micron sized beam.

The planned improvements in photon beam stability outlined in section 3.4 will greatly enhance our ability to use small x-ray beams (of order 10 μm) to address common problems of crystal size and inhomogeneity. The improvements to stability and brilliance are essential for the development of microcrystallography using beams of ~ 1μm diameter.

Sample automounters which are critical to the efficient use of the APS beamlines allow screening of many crystals to find the “diamond in the rough”. However, the wide variety of sample automounters available on the beamlines reduces the ease of movement of users from one beamline to the next. The renewal will provide the possibility for standardization of relevant aspects of these systems.

The increased efficiency of pixel-array detectors and beamline automation will result in the acquisition of vast amounts of data in short time periods. The data will be accommodated by investments to improve our computer backbone, the addition of massive data storage arrays and through additional technical support. We will use the opportunity of the renewal, and the infrastructure investments, to assist older beamlines in improving their capabilities, and in providing leveraged support to many beamlines, e.g. night operators to assist MX users.

OTHER CAPABILITIES

Time resolved studies have great potential for the future. In the ultrafast section we discussed our plans to provide sliced ~1ps pulses to APS users and to upgrade existing ultrafast capabilities. Ultrafast studies of protein photochemistry have been very interesting, and their utility has been

increased by the development of optogenetics (Miller 2006), which permits the pumping of non-photo biochemical reactions using light. Figure 4.6.1 shows an example of light sensitizing a histidine kinase effector. Through this kind of approach, the time development of biochemical intermediate states might be studied with laser pump/x-ray probe techniques, as has been so effective for photoreactive proteins (Moffat 2001).

The imaging needs of the life sciences community will be addressed through the development of new beamlines that will form part of the imaging capabilities in support of our multi-scale characterization of materials (section 2.1) The three relevant capabilities will be:

- A 200-m-long beamline for high sensitivity, wide field x-ray phase imaging,
- A beamline with a 20-m-long experimental hutch for coherent diffraction imaging (CDI) accommodating next-generation focusing optics and high dynamic range area detectors, and
- Coherence-conditioning and focusing x-ray optics that provide well-defined wavefronts for CDI under both plane and curved-wave conditions.

Combined with upgrades to existing imaging capabilities, these new capabilities will provide an extraordinary set of tools for characterizing the hierarchical structures intrinsic to biological systems. The correlative microscopy capabilities will also be valuable.

Dedicated facilities for SAXS/WAXS with tunable x-ray energies (for anomalous diffraction) and time-resolved capabilities will provide a unique window into the dynamics of macromolecular systems. The APS renewal will enhance data quality and reduce the amount of protein needed for an experiment by providing improved beam stability and facilities optimized for solution SAXS/WAXS data collection. Large format detectors will allow the simultaneous collection of SAXS/WAXS data providing information on the secondary, tertiary and quaternary structure of proteins and macromolecular complexes. Automation of the sample delivery and cell cleaning process will allow the study of proteins under a much wider variety of conditions than is currently possible furthering our understanding of protein dynamics and drug/target binding.

Optogenetics – using light to probe biochemistry beyond photochemistry

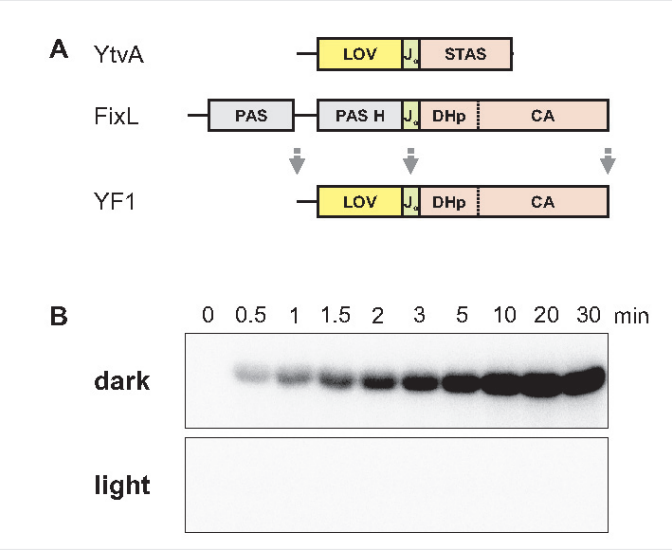


Fig. 4.6.1. *Design of mix-and-match proteins (Moeglich et al. 2009). A) The oxygen-sensitive PAS H sensor domain of one parent protein FixL is replaced by the blue-light-sensitive LOV sensor domain of the second parent protein YtvA, to produce the fusion protein YF1 whose histidine kinase effector domain DHp-CA is now light sensitive. B) Phosphorylation by the histidine kinase proceeds briskly in the dark but is completely abolished in the light.*

5. ENABLING TECHNICAL CAPABILITIES

The scientific performance of users depends on an appropriate source of x-rays, and, as importantly, on many enabling technical capabilities. Detectors, for example, are recognized as the "weak link" in many third-generation x-ray experiments. We propose to provide, either by acquisition or development, the best detectors available for the renewed beamlines. The other technical systems described in this section are equally important in leveraging the science of our users.

Strong technical support is the backbone of a vibrant, state-of-the-art science program at a user facility. This includes support for areas that are typically too large or complex for individual users and staff members to undertake by themselves. Items that fall under this category include detector development, precision positioning capabilities, beamline controls, scientific software, data analysis, and general information technology (IT support). In many cases these support areas are intimately coupled; for instance, framing-rate area detectors often drive the needs for high-performance computing, massive data storage, and higher speed connections among the experiment, storage, and high-performance computers. During the renewal, we intend to strengthen not only the hardware and software that enable high-impact experiments, but also work to strengthen the human capital in support of these systems (section 7).

5.1 DETECTORS

The science underlying the two overarching themes of the APS renewal will require advanced detectors that are efficient for energies up to 100 keV and are capable of high data throughput; large solid angle coverage for imaging, diffraction, and spectroscopy; high contrast; and improved spatial and energy resolution. No single detector can satisfy all of these requirements; the task at hand is then to define performance goals and to identify paths to achieve them, setting priorities in harmony with the science goals of the APS renewal.

The renewal is an outstanding opportunity, not to be missed, to provide state-of-the-art detectors to the users of APS. The detector strategy for the renewal is to acquire state-of-the-art detectors through commercial procurement, collaboration with various partners, and in-house development, and to deploy the detectors to the beamlines with appropriate software. We note that in-house development is crucial for nurturing a solid and long-lasting detector program at the APS and for establishing external collaborations. A reasonable portfolio for in-house effort includes developing sensors for very high energies (E > 40 keV), picosecond electronics and detectors, and novel superconducting sensors.

COMMERCIAL DETECTORS

As with most new light sources, a significant fraction of the detector needs at APS beamlines in the next 2-5 years can be satisfied by purchasing commercial off-the-shelf equipment or by contracting with companies for the fabrication of custom detectors. Most of the commercial detectors to be procured are fast, large-area detectors, either amorphous silicon (a-Si) flat panels for E > 40 keV or Pilatus silicon pixel array detectors (PAD) for lower energies. Additional commercial detectors that are planned include charge-coupled device (CCD) cameras with varied specifications, silicon strip detectors, and assorted single-element photon counting detectors. There is also a strong science case for custom-designed energy dispersive silicon drift detector (SDD) arrays; these custom

detectors, with a modest number of elements, can be purchased from commercial suppliers. For reference, table 5.1.1 shows the approximate number of commercial detectors requested by APS beamline advisory committees. (This does not reflect a detailed proposed list but is given as an indication of user needs).

Table 5.1.1. Detector types requested from beamlines

Detector type	Requests from beamlines
Si PAD (e.g. Pilatus, various sizes)	12-14
a-Si flat panel	12-14
CCD (varied specs)	8
Fluorescence SDD arrays	6
Si strip, single SDD, etc.	15

DETECTOR DEVELOPMENT

Expanded support for a strong detector development program must be an integral part of the APS renewal project. Some of the most challenging science being proposed requires novel detectors. A robust detector development program will include several avenues for collaboration, as well as in-house development.

The APS renewal vision has identified two overarching driving themes: *mastering hierarchical structures through x-ray imaging* and *real materials in real conditions and real time*. In the near future (3-7 years), detector technologies that can be implemented to support these themes include fast-framing area detectors (both photon counting and integrating), large solid angle spectroscopy detectors, and sensors efficient for "very hard x-rays" (E ~ 40-100 keV). Further in the future, improvements in energy and time resolution may be achieved with superconducting sensors and picosecond electronics.

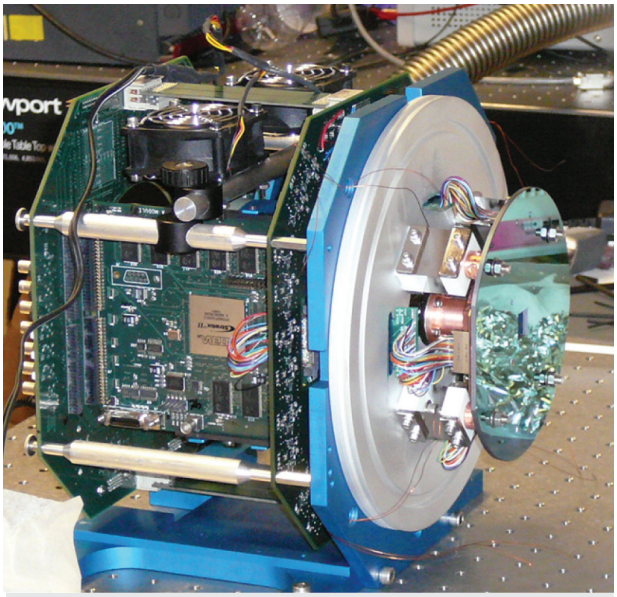


Fig. 5.1.1. *Prototype quasi-column parallel CCD detector for XPCS, developed by the APS Beamline Technical Support Group in collaboration with the Advanced Light Source (LBNL). This novel CCD detector can operate at 200 fps with full resolution.*

Area detectors capable of fast frames rates (100-1000 fps) with moderate pixel sizes (~ 50-100 µm) and large number of pixels (~ 2 k by 2 k) will be very effective for time-resolved diffraction, fast radiography, and time-resolved MX. For diffuse scattering applications (e.g., SAXS), single photon counting can be achieved, while radiography and MX need integrating pixel detectors due to the large instantaneous flux involved. Area detectors with smaller pixels (< 30 µm) and high frame rates, for example, parallel readout CCD cameras, will be appropriate for soft matter studies using XPCS and can also be used for time-resolved scattering. Avalanche photodiode (APD) arrays capable of nanosecond-time resolution are required for nuclear resonant scattering. These various types of area detectors can be developed through collaborating with other national laboratories, e.g., Lawrence Berkeley National Laboratory (LBNL) (fig. 5.1.1), Brookhaven National Laboratory (BNL), and Fermi National Accelerator Laboratory (FNAL), and universities such as Cornell (Vernon 2007); contracting industrial partners such as ADSC, Dectris, Voxtel, and RMD; or joining international collaborations with other facilities,

such as SPring8 and ESRF, and the DESY collaboration on APD arrays. We also expect to benefit from the Linac Coherent Light Source (LCLS) development projects (Haller et al. 2007).

Spectroscopy detectors with large solid angle coverage, i.e., hundreds of detector elements, will allow for fluorescence microscopy imaging with greatly increased elemental sensitivity. A modular detector that can be easily configured into custom geometries for different sample environments can have many applications, for example, in condensed matter physics and environmental science. A possible path to large solid-angle spectroscopy arrays is to pursue energy-resolving pixel array detectors, evolved from the existing BNL spectroscopic PAD (Siddons et al. 2006); another avenue is to build SDD arrays. In all cases, these detectors can be developed in collaborations or through contracts with industrial partners. Developing better sensors for efficient detection of high-energy x-rays ($E > 40$ keV) will enable studies of real materials in real environments. These sensors could be used in pixelated area detectors and would be beneficial for diffraction and imaging studies at high energies, with applications in engineering and applied research. Development in this area would be pursued with industry, for example, in the case of cadmium-zinc-telluride sensors, and with other national laboratories (LBNL, BNL) in the case of germanium. Collaborations on structured scintillators with Risø (Olsen et al. 2008), for example, can be a path to high energy detection with good spatial resolution.

Picosecond electronics and detectors provide a complementary approach to the scientific challenges addressed by picosecond x-ray sources. We plan to expand work at the APS on streak cameras, on fast photodiodes, and on x-ray photo cathodes coupled to fast-readout multichannel plates, some of which have been seeded by Argonne Laboratory Directed R&D (LDRD) funds.

Superconducting sensors for very high energy resolution can be of several types [superconducting tunnel junctions, transition edge sensors, and kinetic inductance detectors (Mazin et al. 2006)] and are capable of a resolution of ≤ 10 eV at 6 keV. This energy resolution allows for increased element sensitivity in fluorescence measurements and makes spin-dependent XAFS possible, with application to environmental, biological, and materials science. Because high-resolution superconducting detectors are inherently slow, they must be arrayed to achieve the count-rate capabilities required for use on synchrotron radiation sources. We will pursue such development in collaboration with the Argonne Materials Science Division and Center of Nanoscale Materials (CNM), and with collaborators at the National Institute of Standards and Technology (NIST) and University of California at Santa Barbara.

The APS renewal project also must include the implementation of detector development infrastructure (test facilities, simulation software, etc.) necessary to carry out the planned detector program. Access to beamlines to evaluate, debug, and calibrate detectors is difficult to obtain given the user demand for beamtime at the APS. We plan to mitigate this access problem by partnering with the optics group in their proposed development of a bend magnet beamline for at-wavelength metrology. (See section 6.3 for more details on the proposed metrology beamline.) Another fundamental component of our development plan is access to application specific integrated circuit (ASIC) technologies. Basic detector physics implies that faster, lower noise (from lower input capacitance) detectors must be realized at the integrated circuit level. We have recently strengthened our ASIC design capabilities by establishing a formal collaboration with the FNAL ASIC Design Group.

5.2 NANOPOSITIONING

Two areas of nanopositioning development are needed in support of the key APS renewal science drivers. In short, these areas are *more* and *better*. More positioning systems are needed to enable more users to employ optics with 100- to 30-nm spatial resolution. Better positioning systems need to be developed to keep the APS at the forefront of hard x-ray microscopy and imaging by enabling users to exploit the next generation of x-ray optics with 30- to sub-10-nm spatial resolution. A new paradigm in the design of x-ray nanopositioning systems is required. The domains of mechanics, sensing, actuation, controls, and environment must be considered as a coupled systems problem.

NEW POSITIONING SYSTEMS

Expanded engineering developments are necessary to underwrite the proposed APS renewal capabilities, especially to support the nanoprobe and nanoimaging applications. While the APS with its partner the Argonne CNM is currently a leader in hard x-ray nanofocusing and nanoimaging, additional engineering infrastructure is required to meet these needs, regardless of whether new instruments are designed and built in-house or conceptualized in-house and contractor built.

The demands on the positioning system are reduced when the operational environment is quiet. Passive isolation is insufficient to ensure sub-10-nm performance. New instruments will require active isolation to minimize the environmental disturbances. Active isolation systems create the quietest conditions for nanopositioning devices and have high stiffness and low drift. These characteristics are needed to maintain position relative to the x-ray beam.

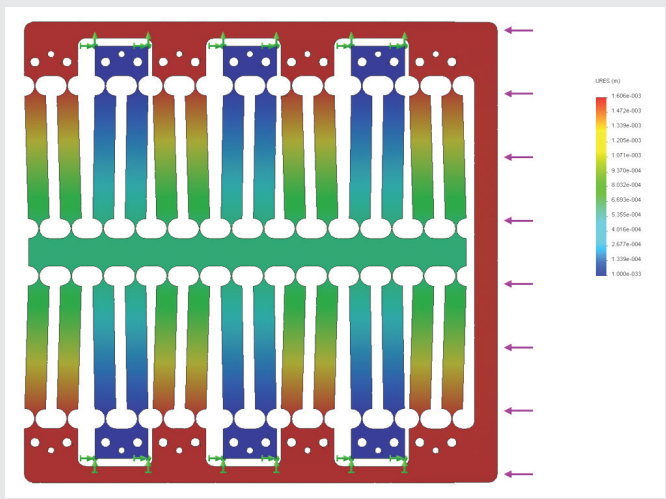


Fig. 5.2.1. *A finite element analysis result for displacement of the multiple parallelogram weak-link structure. Sub-nanometer positioning resolution could be achieved within a sub-centimeter level travel range.*

We will set up test facilities in the APS experimental hall associated with the proposed bending magnet optics metrology beamline to 1) evaluate dynamic performance of nanopositioning instruments to be used in the APS beamlines with the same floor vibration environment, 2) characterize instrument supporting structure vibration response to the APS floor environment, and 3) carry out R&D programs to develop new passive and active vibration isolation and damping mechanisms.

LINEAR WEAK-LINK STAGES WITH LARGE TRAVEL RANGE

X-ray scanning microscope scientists always dream of having a compact single stage to cover large travel range with very high positioning resolution and stability. Drawing upon a fishbone-shaped laminar weak-link structure design (Shu et al. 2009), we are exploring the feasibility of building a weak-link-based precision linear guiding system with sub-nanometer resolution and sub-centimeter travel range in a reasonably compact size. (See fig. 5.2.1.) Based on this novel design, a test bed

will be built to support the APS renewal project. The test bed includes a 3-D high-resolution, high-stiffness nanopositioning system with six laser interferometric encoders with 30-pm spatial resolution. A sub-nanometer closed-loop resolution is expected for this 3-D flexure nanopositioning system with a 3 mm x 3 mm x 3 mm travel range.

NANOPOSITIONING WITH CRYOGENIC SAMPLE CONDITIONS

Samples preserved at cryogenic temperatures are substantially more resistant to radiation damage than at room temperature. For the proposed bionanoprobe, and the CNM nanoprobe, a nanopositioning system must be compatible with cryogenic sample conditions and robotic sample exchange (Maser et al. 2006). To complete this challenging task, a nanopositioning test bed for cryogenic sample conditions will be employed.

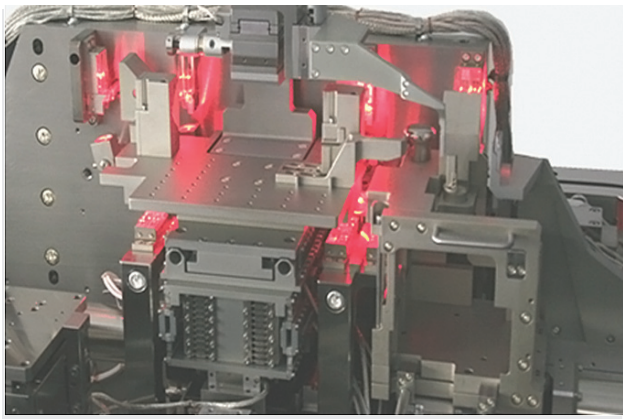


Fig. 5.2.2. *A photograph of the x-ray nanoprobe scanning stage system using seven laser interferometric encoders for nanometer-scale closed-loop resolution. Developed by Argonne, and manufactured by Xradia, Inc.*

Figure 5.2.2 shows a photograph of the CNM x-ray nanoprobe (APS designed and Xradia, Inc., manufactured) at APS/CNM sector 26. The ultimate performance of nanopositioning systems is limited by the ability of the control system to account for the system dynamics, environmental disturbances, sensor noise, and actuator authority. The bandwidth of the system is limited not only by control hardware, but also by the dynamics of the whole system. This holistic approach to x-ray nanopositioning designs will leverage advancements in fields such as scanning probe microscopy where CNM has excellent experience. An accurate model of the mechanism dynamics will be included in the control algorithm. Feed forward control will increase the operational bandwidth, while robust control techniques will balance resolution with environmental and sensor noise rejection. We propose applying these techniques first to a design based upon current positioning mechanics to meet the *more* need—that is, additional robust 30-nm resolution instruments. Then, the same techniques can be combined with the proposed new long-travel flexure systems to meet the *better* need—that is, systems with sub-10-nm resolution.

5.3 COMPUTING AND SOFTWARE

The summary report of the 2006 APS User Workshop on Scientific Software states, "New software analysis tools must be developed to take full advantage of these capabilities. It is critical that the APS take the lead in software development and the implementation of theory to software to ensure the continued success of this facility." Strong coupling is required between teams that implement scientific ideas in software and teams that establish and maintain the computing infrastructure.

DATA ANALYSIS SOFTWARE

Developments in software capabilities will play a key role in realizing the two overarching scientific themes: *mastering hierarchical structures through x-ray imaging* and understanding *real materials in real conditions in real time*. Clearly stated in one of the science cases (APS 2009), "Software is the key to widening the experimental community and is a ***deciding factor in facility productivity***."

Software development is a collaborative process, and our goal is to *enable partnerships*. Our partnerships across the APS user community and beyond will continue to evolve with our science objectives. The Argonne Mathematics and Computer Science Division is ready to help us with key technologies for exploiting IT-secure high-performance computing technologies and data visualization. Other facilities have been partnering with APS for years on the development and maintenance of the experimental physics and industrial control system (EPICS), especially through components that support beamline science. Our collaborations also engage similar groups at other x-ray and neutron sources. Further, we must remain aware of and involved with international standardization efforts such as NeXus (data storage) and DANSE (data analysis).

For the APS to take a more concerted and focused approach to the development of real-time analysis and data visualization software, we must enrich the suite of software tools. The goal is that users can input changes to their experiments based on real-time feedback from the data visualization, modeling, and visualization (fig. 5.3.1).

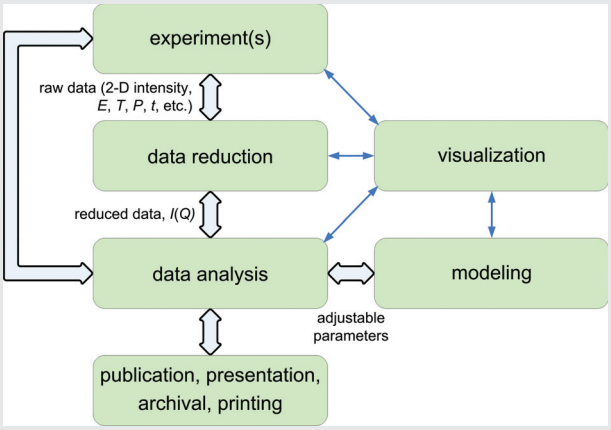


Fig. 5.3.1. *Scientific work flow diagram*

To illustrate, the experimental techniques of x-ray tomography and XPCS differ in their experimental protocols, yet both techniques share the need for high-performance computing (HPC) to reduce the experimental data in real time and provide indications about the outcome of the data just collected and feedback on how best to proceed. Once the experimental data are reduced, 3D x-ray diffraction (3DXRD) microscopy also requires HPC resources to visualize and render the 3D structures. Application of HPC resources to scientific investigations such as these requires specialized expertise that is beyond the scope and capability of an individual user or even group of users, yet common to many APS instruments. During the renewal, we intend to develop staff expertise that is ready to apply HPC and visualization to user's scientific investigations.

Increasingly, the APS will rely on remote (i.e., off-site) collaboration for routine data analyses and, in some cases, even experiments. Often automation is required to reach real-time scientific goals or for mastering one's understanding of the data. Strong, standardized information security protocols must underpin the software to enable remote access by our user community.

To realize the proposed science objectives of the renewal, we plan to expand in concert significantly the number of APS staff supporting the software and controls at each APS beamline. The software staff should work at the beamline *with* beamline staff as *application specialists*, where they will be able to identify and execute the specific software needs of the science program, whether that involves hardware-specific controls such as advanced detectors, application-specific user interfaces, or even implementation of high-performance computing technologies. Furthermore, we need to build and support the user interface software that joins controls together with data analysis and visualization.

During the renewal, APS will develop a central group, separate from the beamline support noted above, to coordinate the scientific software for data reduction, analysis, modeling, and simulation

used throughout the APS user community. Only by integrating that software with beamline data acquisition will the APS be able to fully support the theme of *real materials in real conditions in real time*. The Scientific Software Group will rely on high-speed data networks, abundant data storage, and a fast computation server, all of which must be able to provide service to both on-site and off-site users.

COMPUTING INFRASTRUCTURE

High-quality information technology resources and infrastructure, along with experienced, well-qualified personnel, are essential to the success of the APS. Information technology is our prime communication device internally and externally, our most widely used research tool, an essential resource for our programs, and an indispensable tool for administration. Our ability to accomplish our mission as a Laboratory and our ability to attract and retain the best employees are increasingly dependent on our information technology assets. Thus, it is critical that we provide well-coordinated, high-quality, cost-effective information technology services to all APS users.

The march of technology: Detector technology increases in resolution and acquisition rates every year. Most important, APS is committed to pursuing leading-edge science, and APS scientists are constantly pushing the envelope of available and upcoming technologies. This environment creates a huge challenge for APS IT Support to keep the APS technological infrastructure for data transmission, security, and storage up to date and able to meet the ever-changing needs of beamline users. We must respond to that challenge with innovative computer hardware, expertise in interfacing, and responsive support.

Future growth of APS beamlines: After the renewal is complete we will have ~10 to 12 additional operating beamlines (through build-out of the remaining sectors and canting of IDs in existing sectors) that will require additional support from APS IT. Cyber security is a growing challenge for all user facilities, and we will work to support this need while opening as efficiently as possible a gateway for legitimate data flow.

New servers will be needed to support new and upgraded beamlines. High-data-rate sectors will necessitate local storage systems for spooling real-time data. A data storage capacity of the order of petabytes will be required. Both high data-rate detectors and HPC demand large volumes of resident storage, but the additional needs of backup and archive of scientific data are significant. Remote collaboration and use of national HPC resources demand that such storage capacity is available to members of the APS user community whether they are on the APS campus or off-site.

5.4 LABORATORY FACILITIES

To make the best use of the new and refurbished beamlines that will result from completion of the renewal project, supporting facilities must be adequate. Perhaps most obvious is the need to provide ample laboratory and office space for scientific staff and visiting users. Not as visible to most, but just as vital is the need for sufficient space for other infrastructure components. We have identified the need for increased space for the data center and for more space for on-site early assembly and staging of components.

Fig. 5.4.1. Exterior of a lab-office module

LABORATORY AND OFFICE SPACE

The majority of beamline staff are housed in eight laboratory office modules (LOMs – see fig. 5.4.1) that are located on the outer side of the storage ring hall. As the name suggests, these modules provide laboratories and office space for scientific staff. The laboratories are essential for staff and users; they are used to prepare samples and beamline equipment for experiments.

The space currently available is barely sufficient for the current number of beamlines, staff, and users. The introduction of new beamlines and higher staffing levels at beamlines make it important to plan for provision of adequate accommodations. One of the existing LOMs (437) is currently only a building shell. We would like to build out the interior of LOM 437, providing 8,000 ft^2 of office and laboratory space, similar to that found in the other LOMs. Extending out at least one, preferably two (resources permitting), other existing LOM(s) will be needed to accommodate the anticipated increase in beamline staff population. Each extended LOM will provide an additional 8,000-14,000 ft^2 of space.

DATA CENTER

The current APS data center was designed over fifteen years ago. It has served well over the life of the facility, but the ever-increasing demands for computing capacity mean that it is ever more cramped. The vastly increased power density of today's servers and other computing equipment also makes it increasingly difficult to provide sufficient power and cooling for the data center. Today's data center design requirements are much different from those of 15 years ago. We include expansion of the data center to approximately double the size of the existing facility. It would be refurbished to modern standards and have sufficient power and cooling capability for anticipated future needs.

ASSEMBLY AND STAGING AREA

It is our plan to implement the accelerator, front-end, and insertion device enhancements within our present operations schedule. This means that tunnel installations will have to be all done in the three 3-week access periods each year. To accomplish this, an assembly and staging area must be available in close proximity to the facility to assemble, test, and partially commission the systems before installation in the tunnel. Up until now, the unoccupied sectors of the experiment hall (those sectors not yet containing beamlines) have served that purpose and, in fact, proved very useful to the everyday functioning of the APS. This area has been used for early assembly of large components and for storage of spare parts that need to be stored close to the accelerator for rapid access. Over the past few years, as we have built out the remaining sectors, we have been able to relocate spare parts stored in those sectors in the LOM 437 building shell. However, as the remaining unoccupied sectors and LOM 437 are being developed, this space will no longer be available for storage. Hence, we have a need for a general purpose facility that will be used for early assembly and staging during the renewal project and can later be repurposed as a "high bay" storage area to make up for the loss of space from the experiment hall.

IMAGING INSTITUTE

The APS is involved in negotiations with the State of Illinois for funding of what is known as the "Imaging Institute." In concept, this building would be two stories high and modeled on the footprint of the main APS office building (401). It would have a floor area in the region of 85,000 ft^2. The ground floor would house clean rooms dedicated to optics and detector research and would accommodate the end stations of the proposed long beamlines. The second floor would be outfitted with offices that could house up to 50 people. No DOE funding is sought for this building, but it may liberate pressure on the expansion needs for LOMs. This Institute is not in the scope of the proposed renewal project however ANL has approached the State of Illinois seeking it as matching support similar to the successful partnering in the CNM project.

UTILITIES

We expect that a modest increased capacity will be needed in the APS utility infrastructure system. Initial estimates are modest, but as part of the process in preparing conceptual design reports, we will evaluate in more detail to determine if expansions are required in systems for power demand, chilled water, compressed air, nitrogen storage, etc.

6. RESEARCH AND DEVELOPMENT

This section describes those R&D components that are necessary for successful completion of specific, well-defined renewal objectives, namely the short-pulse x-ray source (SPX) (section 6.1) and superconducting undulators (SCUs) (section 6.2). R&D activities in many other areas, such as beam stability, insertion devices, optics and metrology, and detectors will expand existing efforts at APS, and are included only in order to provide a comprehensive overview.

6.1 SHORT-PULSE X-RAY SOURCE DEVELOPMENT

To achieve some of the ultrafast science goals outlined earlier in this proposal, we are pursuing the development of a source of short x-ray pulses with deflecting cavities using superconducting rf (SRF) technology. SRF technology has several key advantages over the conventional normal-conducting room-temperature copper cavities; for example, it enables the cavities to operate at high-rf repetition rates, including continuous wave. This capability, in turn, allows "chirping" not only in the hybrid mode (the only possibility if room-temperature cavities are employed), but also in all operating modes of the storage ring. A second advantage of an SRF deflecting cavity is its ability to operate at higher deflection voltage without thermal or electrical breakdown, which is not possible with copper deflecting cavities due to pulse heating problems.

Our goal for the SPX R&D phase is to demonstrate the fabrication of suitable SRF deflecting cavities that (1) meet the technical specifications (as outlined below) , (2) are compatible with the APS storage ring operation, and (3) have acceptable operational reliability with minimal or no impact on the routine storage ring operation. The performance parameters for the SPX deflecting cavity are listed in table 6.1.1.

Table 6.1.1. SRF deflecting cavity parameters

Parameter	Value
Frequency	2815 MHz
Total deflecting voltage	4 ×2 MV
x-ray pulse length [full width at half maximum (FWHM)]	~2 ps
Nominal beam current	200 mA
No. of cavities/cryomodule	8 × 2 or 10 × 2

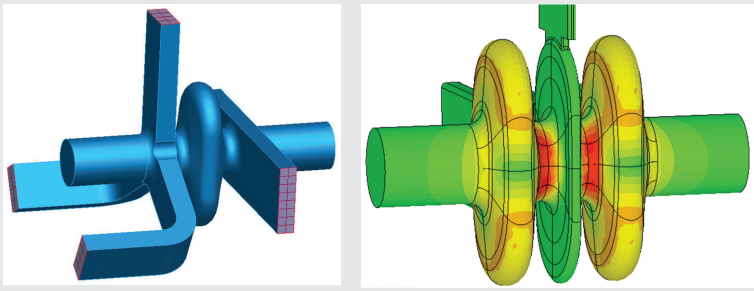
Since 2007, we have done feasibility studies, rf design simulation, and cavity prototyping to identify key technical challenges that need to be addressed in the R&D phase. We have achieved this goal through a partnership with Jefferson National Accelerator Facility (JLab).

Collaboration with JLab has resulted in significant and crucial progress toward a pre-R&D phase development of several prototype deflecting rf structures as potential candidates for the APS SPX cavities system. We were able to clearly identify key technical cavity design issues and fabrication challenges in this pre-R&D phase, which was supported by the Argonne Strategic Initiatives LDRD program. We are now in a prime position to launch a multi-year R&D program to demonstrate proof of principle and then to develop and fabricate a robust SRF deflecting cavities system to be installed in the APS storage ring.

The elements of the proposed 3-year R&D program are discussed below. When the program is complete, cavity design, construction, and installation would begin.

RF DEFLECTING CAVITY

We will continue our effort to further explore various cavity geometries, including single- and multi-cell structures, and decide on an optimized geometry that meets the SPX technical specifications. We will concentrate our effort on three potential candidates: a single-cell cavity with a beam-pipe waveguide damper (fig. 6.1.1), a single-cell cavity with on-cell waveguide damper (fig. 6.1.2), and a three-cell cavity (fig. 6.1.1). We will explore the pros and cons of each of the above geometries through detailed design, simulation. and prototyping.



SC cavity designs

Fig. 6.1.1. *Single cell with beam-pipe waveguide coupling (left) and a three-cell cavity in π/2 mode (right).*

Prototype SC cavities

Fig. 6.1.2. Prototypes of waveguide on-cell coupling.

HOM/LOM DAMPERS

The key to a successful deflecting cavity is managing higher order and lower order modes (HOM/LOM) excited within the cavity. To achieve the level of damping required at the APS, several monopole modes and vertical dipole modes in the cavity have to be sufficiently damped to meet the vertical stability impedance requirements for the stored beam. We will explore different waveguide damping geometries, including cold and warm damping sections, to efficiently extract HOM/LOM power without impacting the main deflecting mode operation.

CRYOMODULE

We will design and prototype an appropriately sized cryomodule that can house either a string of 8 to 10 single-cell cavities or 4 three-cell cavities. Because of the large number of penetrations required (due to multiple waveguide ports), it is important to develop a high performance cryomodule that meets the requirement for cavity rf power. We will implement improvements to the cryomodule thermal and magnetic shields during the prototyping phase.

RF CONTROL

A robust and reliable low-level rf (LLRF) control system is required to meet the stringent amplitude and phase errors requirements and to regulate and control the deflecting cavities fields while providing rf system diagnostics. We will explore and develop a suitable LLRF control system for the deflecting cavities system.

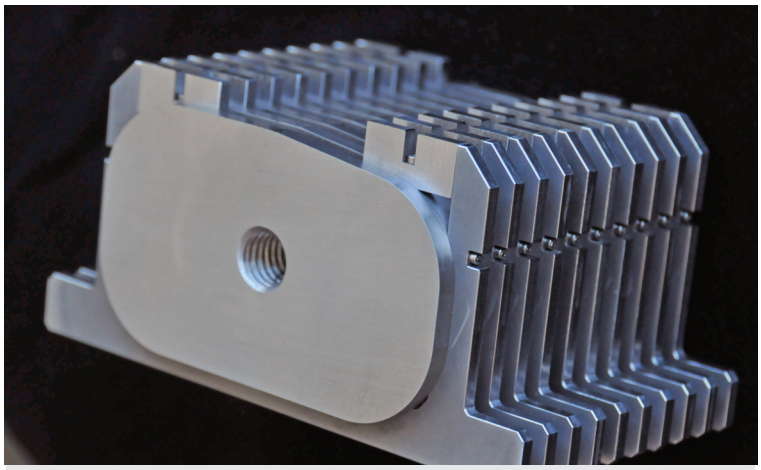
6.2 UNDULATOR DEVELOPMENT

R&D on undulators is part of normal business for APS, and several developments are ongoing, e.g., development of low-energy polarizing undulators and the examination of the possible use of in-vacuum undulators. This section describes the only substantial undulator R&D project that promises high payoff for the performance of high-energy sources needed by many of the science drivers in this proposal, namely, a superconducting, short-period undulator. After the R&D program, we would plan to install several superconducting undulators in the storage ring.

SUPERCONDUCTING UNDULATOR

For the highest energy applications at the APS, a superconducting undulator is very desirable, because of its very high brightness at higher x-ray energies. High energy x-rays are important for many of the planned science programs at the renewed APS, including stress/strain measurements in engineering materials, powder diffraction of compounds containing heavy elements, diffuse scattering by defects in complex oxides, PDF measurements of liquid and amorphous materials, and spatially resolved SAXS from gradient layered materials.

Short period undulators can provide a substantial increase in photon brightness at higher energies, as long as their on-axis magnetic field is strong enough to give a reasonably broad photon energy tuning range. Short-period superconducting undulators can produce a higher field than permanent-magnet undulators with the same period length, so a superconducting undulator opens access to shorter periods and higher brightness (up to an order of magnitude) at high photon energy while providing a tunability range from 20 to 25 keV in the first harmonic. Short-period optimized undulators can also be important for thermal management of the first optics for beamlines.



Superconducting undulator yoke
Fig. 6.2.1. *Superconducting wire will be wound in the grooves of this 10-pole prototype. This and a matching piece will form the top and bottom jaws of the undulator.*

The development effort is focused on a 16-mm undulator period based on a Nb-Ti superconductor. The design for the magnetic yoke on which the superconducting wire is wound has been completed. A number of short prototype yokes (see fig. 6.2.1 for one example) have been wound and potted in epoxy by using a potting technique that was developed in collaboration with FNAL. Those that have been cold tested all reached the expected critical current, giving confidence in the basic soundness of the magnetic design.

Most recently, a pair of 42-pole yokes has been produced. Magnetic measurements of one of the yokes that was cold tested showed a phase error at the desired maximum current of about 4 degrees, the same as a typical APS room-temperature undulator phase error. Test measurements of the assembled pair of 42-pole sections are now in preparation. Once successfully tested, 1.2-m-long yokes will be produced and wound.

To test the assembly, a cryostat to hold the magnetic structures horizontally, along with a cooling scheme to handle the beam heat load, must be developed. Computer-based thermal modeling is being used to study various design options. Experimental tests are also being developed, where a thermal load will be introduced to the assembly to emulate heating by the electron beam.

Another significant task is the improvement of the magnetic measurement system for the complete, horizontally oriented undulator. An existing vertical measurement system is being used for the vertical cryostat, but modifications will be required for it to operate in a horizontal orientation.

6.3 OPTICS R&D

Upgrading existing optics with state-of-the-art components and enhancing scientific capabilities with new optical components (compound refractive lenses, multilayer Laue lenses, etc.) that were not available when the APS beamlines were originally designed will be an important component of the APS renewal to bring the beamlines into the 21st century. For instance, over the last decade tremendous progress has been made in mirror fabrication and metrology technology. Mirror polishing capabilities have steadily improved; today, flat 1-m-long mirrors with surface slope error on the order of 1 μrad (0.5 μrad on best-effort basis) rms and microroughness on the order of 0.2 nm rms are readily available within a reasonable time and cost. Such a mirror was simply not available when many of the APS beamlines were built. Many of the required optical components associated with the renewal, such as mirrors, zone plates, compound refractive lenses, and single-crystal synthetic diamonds for monochromators, can now be procured through commercial vendors. However, some state-of-the-art x-ray optical components cannot be directly procured and, in fact, will require R&D to reach the projected goals. Below is a description of where we feel investment is needed in R&D to maintain the APS as a leader in x-ray optical developments.

Multilayer Laue lens

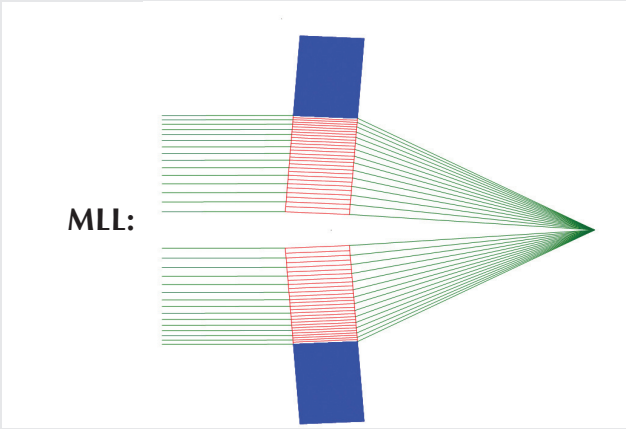


Fig. 6.3.1 *Each Fresnel zone corresponds to a thin film layer deposited by magnetron sputtering.*

NANOFOCUSING OPTICS R&D

Pushing the limits on hard x-ray focusing optics is, and will remain, a major area of x-ray optics R&D. Scientists are now just beginning to realize the power of focused x-ray beams for almost all fields. Indicative of the importance of sub-100-nm x-ray beams, focusing to below 30 nm is specifically mentioned in eight of the ten science case reports in the APS Renewal Workshop (APS 2009). APS has been a leader in developing nanofocusing using multilayer Laue lenses (MLLs, see fig. 6.3.1) that will allow hard x-ray studies of materials with 5-nm resolution. Now is the time to incorporate these new capabilities into the APS beamlines, and this renewal will allow us to do so. To quote from the science case on Engineering Applications & Applied Science (APS 2009); "Past progress at the APS has resulted in promising x-ray optics with demonstrations of a record smallest

one-dimensional hard x-ray focusing device – a Laue Zone plate lens with 16 nm FWHM – and demonstration of the world's smallest polychromatic probe, a Kirkpatrick-Baez total-external-reflection mirror system with 80 nm-diameter probe size. Yet these optics are not widely available at the APS and a major effort is needed to achieve these performance numbers routinely and for x-rays above 40 keV."

As part of the renewal, we are requesting new instrumentation for differential deposition of Kirkpatrick-Baez mirrors larger than 100 mm. This instrumentation would include a high-precision substrate transport system and a new ellipsometer.

HIGH HEAT LOAD R&D

One of the planned accelerator upgrades is an increase in stored current to 200 mA. Although this increase is only a factor of two over our present operating level, many of the first optical components (typically silicon or diamond double-crystal monochromators) that are presently installed may be at the "tipping point," will not be able to handle the modest increase in power loading, and will require substantial upgrades. Finite element modeling of the thermal distribution from new, and in many cases longer, insertion devices operating at 200 mA will need to be completed to develop the required cooling. The need for cooling of mirrors and multilayer optical components is anticipated in a renewed APS, and work in this area will also be required.

This renewal is also an excellent opportunity, during modification of the first optical components, to improve the overall mechanics of the optical systems and thereby improve the delivered x-ray beam stability.

COHERENCE-PRESERVING OPTICS R&D

As we move forward with an increased focus on the use of the lateral or transverse coherence of the x-ray beam, there will be a need for optical components that do not degrade the lateral coherence. R&D activities on the development of coherence-preserving windows, monochromators, mirrors, and other optical components, such as beam splitters, will be pursued.

X-RAY OPTICS METROLOGY R&D

Polishing techniques for x-ray mirrors have improved dramatically over the last decade. Driven by the semiconductor industry, sophisticated techniques have been developed to precisely manufacture aspheric optical mirrors for extreme ultraviolet projection lithography tools. Some of these, such as computer-controlled optical surfacing (Jones et al. 1991), zonal polishing, and magneto-rheological finishing (Jacobs et al. 1995), have greatly benefited x-ray optics. However, because of the finite size of the tool used, total conformance to the desired shape is hard to achieve. More important, mid-spatial surface features resulting from figuring imprints can degrade the beam coherence and are difficult or impossible to eliminate. Some of the promising techniques that are capable of removing minute surface defects include ion beam figuring (Wilson et al. 1988) and elastic emission machining (Yumuto et al. 2005). In fact, mirror technology has improved to the point that the existing mirror metrology at the APS can now barely measure state-of-the art mirrors because the quality of the mirrors is now approaching the noise level of the metrology instruments.

Conventional tools for standard laboratory optical metrology, such as slope measuring devices [e.g., the long trace profiler or LTP developed more than two decades ago at BNL (Takacs et al. 1987)], are no longer sufficient. In addition to being no longer commercially available, the LTP is a one-

dimensional profiler, while two-dimensional (2-D) metrology is necessary for characterization and fabrication processes. To meet future needs, an extensive R&D effort is needed to improve existing techniques and develop a new generation of metrology tools that are robust and capable of measuring 2-D mirror surfaces with a high degree of precision at all length scales. As part of the APS renewal, we will upgrade our laboratory-based x-ray mirror metrology capabilities with a new Fizeau interferometer with a sample stage capable of measuring large mirrors and a new atomic force microscope for surface roughness measurements. The Fizeau interferometer will incorporate a sub-aperture stitching capability. An upgrade of the environmental control (temperature and vibration) for the metrology lab is needed.

AN OPTICS METROLOGY BEAMLINE

In the future we will need to go beyond laboratory-based tools for x-ray mirror metrology. Hence, we are planning, during the renewal, to establish a metrology beamline on a bending magnet source that is suitable for at-wavelength metrology of x-ray mirrors. We plan that this beamline will be more than just a place to measure mirror quality. Multilayers and elliptical mirrors are synergistically related since they both aim at producing a nanofocused beam, and a nanofocusing test bed that can accommodate both types of optics is proposed as part of this beamline. We believe this new capability will significantly speed mirror and multilayer Laue lens development.

Phase retrieval techniques to invert the far-field diffraction pattern to calculate the wave-field intensity in the focal plane will become ever more important for nanofocusing optics and are key to our science plans. Implementing phase retrieval techniques at a metrology beamline makes efficient use of the synergy in the metrology tools needed to characterize both focusing mirrors and lenses. Such a beamline will provide for a stable setup for testing of nanofocusing optics and topography of Si and Ge monochromators and analyzers and diamond monochromators and beamsplitters.

The proposed beamline will also be shared with the engineering group to test new optics manipulators and with the detector group to test and calibrate new detectors.

6.4 DETECTOR R&D

A detailed plan of activities for detector R&D was described in section 5.1. That plan will be summarized here for completeness.

We plan to continue our very successful collaboration with LBNL on fast-frame rate area detectors, as advances in these types of detectors can impact many fields of science from XPCS to time-resolved imaging and scattering. Many of the scientific activities proposed for a renewed APS involve the use of hard (>20 keV) x-rays, and development of more efficient sensors (i.e., high Z materials) for high-energy detection will be an important component of the detector R&D portfolio.

We plan to continue our work at the APS on streak cameras, which has been seeded by Argonne LDRD funds.

ASIC technologies will be another fundamental component of our development plan. We need to grow our expertise at the integrated circuit level as that is often the basis for faster, lower noise detectors. We have already made progress in that area through the establishment of a collaboration with FNAL but will expand the work during the renewal.

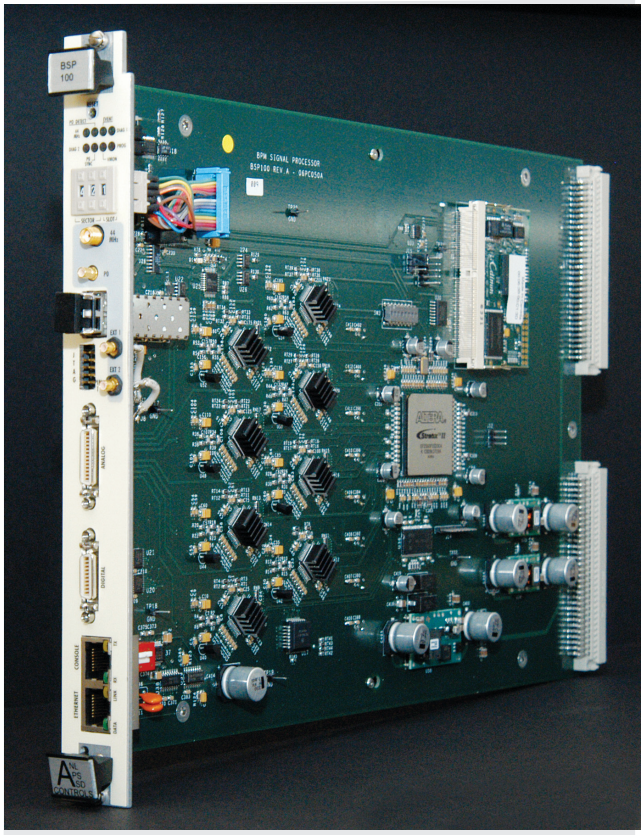
6.5 INCREASED BEAM STABILITY

Future beam stability goals for the APS storage ring positional and pointing stability are 1 micron and 0.5 microradians (peak-to-peak), respectively, over a week. These goals represent a level of improvement of between 5 to 10 times better than present performance. Similarly, AC beam stability goals have been tightened to require position and pointing stability at the level of 0.42 microns and 0.22 microradians rms in a frequency band from 0.017 to 200 Hz, roughly a factor of three improvement. The strategy for achieving these goals will involve research in several key areas: electron- and x-ray beam position monitoring, high-speed feedback system technology, mechanical engineering of detectors and detector support structures, and air- and water-temperature regulation technology.

BEAM POSITION MONITORING

Experience at new synchrotron light sources has resulted in a number of new types of rf beam position monitor (bpm) technologies for monitoring the electron beam. The rf bpms are key to improvements in AC beam stability and long-term source position stability. An important part of the R&D program supporting the APS renewal objectives will be a critical evaluation of the different technologies and how they perform for the different APS operating modes (top-up vs. beam decay mode, 24- 324- and hybrid fill patterns, and high-current operation) (Pinayev et al. 2009).

The APS has been a leader in the deployment of large-scale photon beam position monitoring and control systems for insertion device sources (Decker et al. 1999, Broadbent 2009). This work has made it possible to achieve long-term x-ray pointing stability at the level of a few microradians. The present technology, based on ultraviolet photons, required a major effort to compensate for systematic errors dependent on the insertion device gap. Even so, these detectors have residual errors at the level of 10 to 20 microns.



Fast feedback for stabilization

Fig. 6.5.1. APS broadband rf bpm data acquisition board employing field-programmable gate array technology.

Recent work at the APS diagnostic beamline 35-ID has resulted in a hard x-ray bpm design based on copper back-fluorescence, which measures the transverse position of the x-ray beam independent of the gap over two orders of magnitude in intensity with very high resolution. This work stemmed from earlier studies on copper fluorescence detection at beamline 19-ID (Decker et al. 2007). Further development is necessary to refine this design for operation with very high power density for installation in the beamline front ends.

FEEDBACK SYSTEM DEVELOPMENT

The primary goal for the feedback system is to increase the sample rate from 1.5 kHz to 15 kHz or more using modern data network technologies (fig. 6.5.1). This will require the construction of a test bed for the validation of interfaces to bpm and steering corrector systems to assess data latencies. A key feature of this system will be a fast real-time data network to relay data around the ring. Only after the test bed is done can a system design be completed.

MECHANICAL ENGINEERING

To achieve the very tight requirements for long-term stability, the support structures for critical beam position monitors must be engineered to be insensitive to small tunnel air-temperature variations while minimizing ground-motion-induced vibrations. Prototypes of such structures must be constructed and validated prior to deployment around the ring. In addition, the mechanical design of the high-power hard x-ray bpm will require prototyping and testing.

TEMPERATURE REGULATION

Long-term beam stability is dictated to a large extent by air- and water-temperature stability. However, since the storage-ring tunnel air handling systems are distributed over 1.1 km, significant improvements in the tunnel air-temperature stability may be prohibitively expensive. It is important to assess the trade-offs between air-temperature stability improvement and other improvements such as actively regulating bpm support structure temperatures with cooling water.

7. USER SUPPORT AND WORKFORCE DEVELOPMENT

To ensure maximum benefit from the exciting new scientific opportunities provided by the APS renewal, we plan to expand in parallel user support and outreach. User support, in the form of additional dedicated staff and expanded technical and software capabilities, will enable users to become productive far more quickly. User outreach is essential to acquaint both existing and new communities of users with the expanded capabilities of the APS, as well as help fulfill the stated DOE mission of providing a workforce of well-qualified individuals who meet the specific needs for future scientists and engineers in key technology areas. Funding for this activity, however, is not requested from this renewal project.

USER SUPPORT

A robust user support system is essential for full realization of the benefits from the strategic investment in APS beamlines. Technical support, in the form of enhanced software, beamline controls, and data analysis, as well as detector support and development, must be expanded. Beamline automation is expected to be a growing activity, but from experience, it does not lessen the need for support as the demand grows and throughput of users increases. Included in this scientific user support should be an increased number of expert staff scientists who understand the scientific needs of the appropriate community and can also help design experiments. An example of the success of such an approach is HPSynC, a jointly funded DOE effort to leverage high-pressure science. We hope to foster similar activities in catalysis, energy storage, transportation, and biomedical imaging, for example. Experts may be of particular value to potential industrial users, since in many cases there may not be the required expert knowledge in a small company. Because of the expected increase in the number of operating beamlines from 61 to more than 70, additional dedicated staff members will ultimately be needed to direct scientific programs, facilitate user research, collaborate with users, assist new users, provide safety assistance and oversight, and maintain or enhance beamlines. At least one additional beamline scientist (Ph.D. level) and one or two additional user support staff (B.S. or M.S. level) per beamline, as well as several shared support staff members, are required to provide the necessary capabilities.

USER OUTREACH AND WORKFORCE DEVELOPMENT

User outreach has two primary objectives. The first is to broaden the spectrum of the user community by educating current users about new capabilities and introducing APS capabilities to potential users in new disciplines. The second is to develop a workforce that ensures a steady supply of well-trained scientists and engineers to staff the beamlines and facility.

To address the first objective, schools, such as the annual National Neutron X-ray Scattering School (pioneered at Argonne and now run jointly by the APS and the Spallation Neutron Source) and the APS XAFS school, will provide a basic introduction to overall facility capabilities for new individuals in research fields where synchrotron radiation is traditionally used. Targeted smaller hands-on workshops, such as that illustrated in fig. 7.1, will enable current users to access the new capabilities most productively. These schools and workshops can be supplemented by teaching videos and/or webcasts and webinars.

Fig. 7.1. *BioCARS Workshop on Time-Resolved Macromolecular Crystallography, November 20-22, 2008*

Reaching potential users in new fields or disciplines is more difficult. A strong outreach program, directed by an APS User Outreach Coordinator and conducted in partnership with Argonne's Division of Educational Programs, should focus on trips to colleges and universities, as well as initiatives to historically black colleges and universities and minority education institutions, with the objective of stimulating interest in the APS and its capabilities. Follow-up can then take a number of directions: summer faculty research fellowships, student summer internships, and stipends to attend workshops/schools.

We also need to improve our outreach to the industrial community. Small companies may be unaware and/or lack in-house expertise to appreciate the capabilities of the APS. We are proposing to tackle this issue through the appointment of an industrial liaison whose primary mission is to provide outreach and assistance to potential industrial users. We would like to see this expand into a small group of staff well versed in those techniques that are likely to be of interest to companies who would assist industrial scientists in performing experiments at the APS.

The second objective, ensuring a steady supply of well-trained scientists and engineers to staff the beamlines and the facility, requires both a vibrant user community that includes undergraduate and graduate students and hands-on opportunities that could be provided via work/study arrangements with local community colleges and nearby four-year colleges/universities. These work/study or co-op programs can begin with initial beamline renovations, thus providing teaching opportunities for individuals interested in beamline development and maintenance. We are involved with such a co-op program with the engineering school at Northwestern University but need to expand the number of co-op students and universities that are involved.

Training of graduate students (both M.S. and Ph.D.) can also be improved through formal connections between staff of the APS and local universities. We are considering an arrangement where APS staff will develop (or co-develop with university faculty) and (co-)teach specialized courses dealing with various aspects of synchrotron radiation. These upper-undergraduate-level or graduate-level courses could be science-oriented (condensed matter, chemistry, etc.), technique-oriented (inelastic scattering, powder/single crystal diffraction, etc.), and/or instrumentation/engineering-oriented (optics, detectors, nanopositioning, etc.). Courses related to synchrotron radiation would be posted on the university website for use by APS staff and users who wish to enhance their knowledge in certain areas/topics. We believe a library of such courses would be a valuable training tool for new beamline scientists, novice users, and support staff.

8. MANAGEMENT, BUDGET, AND ES&H

8.1 MANAGEMENT AND BUDGET

USER INVOLVEMENT

The proposed renewal is the result of extensive strategic planning with the users, staff, and external scientists. Details of our planning can be found at the web site www.aps.anl.gov/Renewal/. Table 8.1 gives a list of workshops that have been held by APS in the last three years.

The APS Scientific Advisory Committee has been heavily engaged in strategic planning for the renewal. A list of 2008-2009 members is given in Table 8.2 on the following page.

A steering committee with representatives from the user community has been active in planning the renewal process. If and when we are granted the mission need decision (CD-0) by DOE, we will establish the project management leadership team and develop the conceptual design report (CDR). In doing so, we will invite existing and potential users to provide their critical input for the elements of the six beamline instrumentation themes. We will appoint a machine advisory committee and a beamline technical advisory committee to advise the project management on the development and eventual execution of the project. We will ask the existing Scientific Advisory Committee to continue to provide overall guidance during the project.

PROJECT MANAGEMENT

The APS and Argonne have much of the skills and expertise necessary to complete this project efficiently. Nevertheless, we estimate that we would need to secure additional effort in design and installation, computing, and key engineering R&D areas. The project will be led by a project director, who would report

Table 8.1. APS user workshops over last three years

Workshop Titie	Date
APS Renewal Planning Workshop	Oct 20-21, 2008
Small Angle Scattering: Beyond R_g	Jun 28-Jul 2, 2008
Understanding Condensed Matter Dynamics at the Microscopic Level	Jun 23-24, 2008
Workshop to Introduce High-Resolution Inelastic X-ray Scattering on Earth Materials using Synchrotron Radiation	May 31-Jun 1, 2008
Short Pulse X-rays at the APS	May 9, 2008
APS Upgrade/ERL Optics Workshop	Apr 23, 2007
New Applied Materials Research from Improved High Energy X-Ray Sources	Jul 28, 2006
Detectors	Jul 21, 2006
Sub-meV Energy Resolution	Jul 20, 2006
Microscopy	Jul 17, 2006
Biology and Life Sciences	Jul 14, 2006
New Structural Science from Improved High Energy X-Ray Sources	Jul 13, 2006
Picosecond Science	Jun 29, 2006
Novel Science with Polarized X-Rays	Jun 29, 2006
Interfacial and Surface Science	Jun 29, 2006
Coherence/Imaging	Jun 15 & Jul 11, 2006
Workshop on Intermediate Energy X-Ray	Jun 13, 2006
Science Opportunities using X-Rays and Split Gap High-Field Magnets	Jun 8-9, 2006
Ultrafast SAXS	Jun 6, 22, 2006
Fourth CW and High Average Power RF Workshop	May 1-4, 2006

Table 8.2. APS Scientific Advisory Committee 2008-2009

Name	Research Field
Prof. Jens Als-Nielsen *Neils Bohr Institute, Copenhagen*	X-ray physics
Dr. Michelle V. Buchanan *Associate Laboratory Director,* *Oak Ridge National Laboratory*	Molecular analysis, protein interactions
Prof. Philip Bucksbaum *Director, Ultrafast Science Center, SLAC*	Atomic molecular optical physics
Dr. Howard Einspahr *Research fellow, Bristol-Myers-Squibb (retired)*	Macromolecular crystallography, drug design
Prof. Britt Hedman Deputy Director, Stanford Synchrotron Research Laboratory	Biophysics, structural chemistry, x-ray absorbtion spectroscopy
Prof. Dame Louise N. Johnson University of Oxford	Macromolecular crystallography
Prof. Janos Kirz *Scientific Advisor, Advanced Light Source*	X-ray microscopy
Prof. Miles V. Klein *University of Illinois (retired)*	Experimental condensed-matter physics
Dr. Dan Neumann *National Institute of Standards and Technology,* *Center for Neutron Research*	Structure and dynamics in molecular material
Prof. Piero A. Pianetta *Deputy Director,* *Stanford Synchrotron Research Laboratory*	X-ray microanalysis, especially surfaces and interfaces
Dr. William Stirling *Director General,* *European Synchrotron Research Facility*	Magnetic scattering
Dr. Soichi Wakatsuki *Director, Photon Factory*	Macromolecular crystallography
Dr. Glenn Waychunas *Molecular Geochemistry Leader,* *Lawrence Berkeley National Laboratory*	Synchrotron radiation applied to geochemistry and environmental science
Prof. Donald J. Weidner *Department of Earth and Space Sciences,* *Stony Brook University*	Geophysics
Dr. Wei Yang *Chief, Structural Biology and Cell Signaling Section,* *National Institutes of Health*	Molecular biology and macromolecular crystallography
Prof. Larry Lurio (ex-officio) *Northern Illinois University;* *Chair, APS Users Organization*	Coherent x-ray scattering and physics of soft materials
Dr. Dennis Keane *(ex-officio)* *Director, DND CAT* *Chair, APS Partner User Council*	Surface x-ray scattering and reflectivity

directly to the APS Director. Within the project, we will prepare a work breakdown structure reflecting the accelerator, beamlines, and enabling technical capabilities.

PROJECT STAFFING

By the time of project completion we would need to enhance our staffing levels to support operation of the increased number of beamlines (from 61 to ~75), the increased support for theory and software, and the other kinds of user support discussed in section 7. Our current staffing level is ~450, and when the project is completed, we estimate the need for ~550 people, which would represent an operating budget increase of ~$17M per year (~14%) in today's dollars. During the construction period we would use temporary/contractor staffing and other Argonne personnel to cope with the effort associated with design and installation. Funding for increasing our permanent staffing gradually over the project period will be requested separately.

Table 8.3 Breakdown of preliminary total estimated cost and total project cost ($M)

Project Support	**$ 23.5**
Project Management	$ 3.8
Project Engineering	$ 19.2
ES&H Management	$ 0.5
Technical Equipment	**$246.5**
Accelerator Systems	$ 77.1
Beamlines	$115.4
Enabling Capabilities	$ 54.0
Contingency (25%)	**$ 67.5**
TEC	**$337.5**
R&D & CDR	**$ 12.5**
TPC	**$350.0**

TOTAL PROJECT COST AND SCHEDULE

Our preliminary total estimated cost (TEC) is $337.5M, and our preliminary total project cost (TPC) for the proposed renewal project is $350M, distributed as shown in Table 8.3.

SCHEDULE

After the conceptual design is complete, most elements of the project can be started aggressively and do not need further R&D or long lead-time procurements. Others, in particular the short-pulse x-ray project, require significant R&D and could not begin construction until the later years of the project. We will have high demand in the first two years for beamline engineering design. Fortunately, several of the major proposed activities have already resulted in substantial completion of preconceptual design as they worked through our SAC beamline review process. As a result, these could begin very soon after project start. Our estimate is that the project funds need not be heavily front-loaded and could be distributed relatively evenly over 6 years. We will develop more detailed scheduling plans prior to the CD-1 decision point.

8.2 ENVIRONMENT, SAFETY AND HEALTH

INTEGRATED SAFETY MANAGEMENT SYSTEM (ISMS)

Environment, safety and health (ES&H) requirements will be systematically integrated into management and work practices at all levels so that the APS renewal project is executed while protecting the public, the worker, and the environment. APS renewal project management will follow the Argonne Laboratory Management System (LMS) policies and procedures that embody the ISMS Guiding Principles and Core Functions. This effort includes making it clear that the responsibility for safety and environmental protection starts with the Argonne Laboratory Director and flows through the management chain to Associated Laboratory Directors, to Division Directors/ Department Heads, to the APS renewal project manager, to group leaders, to line supervisors, and finally to the workers. It is the responsibility of the APS renewal project management to ensure that staffs are trained and are responsible for ES&H in their assigned areas.

The APS renewal project work at Argonne will be executed in accordance with Argonne ES&H practices as required in the Argonne LMS documents. These require that hazards are identified and mitigated; work is authorized after ES&H analysis is completed; and technical oversight of work is conducted by APS renewal management and staff. The Argonne Environment, Safety, Health, and Quality Assurance Division will provide technical support to the project and will review project activities as required by the Argonne LMS procedures related to Work Planning and Control. Independent oversight will be conducted by the Compliance, Oversight and Assessments Division.

NATIONAL ENVIRONMENTAL POLICY ACT (NEPA)

The APS currently satisfies NEPA requirements under an existing environmental assessment (EA), which was last revised in June 2003. The APS renewal project will be reviewed to determine whether or not its activities will continue to fall under the revised EA. If not, project management will work with the DOE Argonne Site Office to determine whether or not the EA requires further revision, of if a site categorical exclusion is more appropriate for the specific activities.

FIRE HAZARD ANALYSES

A fire hazard analyses (FHA) was conducted of APS during its construction, and its conclusions were included in the APS Safety Assessment Document (SAD). The APS renewal project will be reviewed to determine whether or not its activities will remain within the FHA or require additional analyses. If additional analyses are required, the conclusions will be incorporated into the APS SAD and into the applicable specific designs involved with the project.

SAFETY ASSESSMENT DOCUMENT

Specific ES&H hazards and the means for their mitigation have been detailed in the existing APS SAD. The APS renewal project will be reviewed to determine whether or not the hazards associated with its design will remain within the existing SAD or require additional analyses. If additional analyses are required, the SAD will be revised to incorporate the results and any additional mitigation required. At a minimum the SAD facility descriptive material will be revised to reflect the changes from the project.

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