THE ADVANCED LIGHT SOURCE STRATEGIC PLAN: 2009–2016

Addressing The Scientific Grand Challenges and Our Energy Future

February 2009

ALS

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EXECUTIVE SUMMARY

In 2008, the Advanced Light Source celebrated its 15th anniversary. Since it was first commissioned in 1993, the ALS has clearly emerged as the world leader in soft x-ray science. Invention and innovation in instrumentation, commitment to user support, and attention to safety have all contributed to its success, as measured by a growing and productive user community. The ALS presently supports about 2000 scientists per year whose ALS-based research appears in more than 500 refereed publications per year, with a large fraction of these articles appearing in high-impact journals. Over the last six years, in fact, the ALS has had the highest ratio of high-impact publications among all the U.S. synchrotron radiation facilities.

Continued success is not guaranteed, however. During these same six years, for example, an almost 150% increase in the number of submitted research proposals signaled a steadily increasing demand for beamtime by the ALS scientific community, but this growth is much faster than the pace at which new beamtime has become available. Moreover, the ALS leadership position is being challenged in almost every area of high-impact science, and some cases surpassed, by newer light sources, such as the Swiss Light Source, Soleil, and BESSY II, all of whom are aggressively capitalizing on our innovations. It cannot be over emphasized that to remain at the forefront, ALS must move rapidly to implement the novel ideas it generates before falling behind the other facilities.

Even more important, we are at the beginning of a new era of science with the promise of controlling energy and matter down to the most intimate level, a step up from the previous emphasis on observation and understanding. In the DOE BES research portfolio, energy research has long been a core mission, but the dual challenges posed by the world's increasing energy appetite in the face of our finite nonrenewable energy resources and the potentially devastating consequences of global climate change make it necessary to couple matter and energy research in a new and daring way. We need new discoveries that allow us to control both atomic and electronic structure, thereby yielding new materials and processes tailored to contribute to energy technology breakthroughs.

An incremental approach to beamline renewal and accelerator upgrades will not be up to the task. **It is the strategic vision of the ALS to meet these new challenges and maintain its leadership over the next two decades by renewing its infrastructure, upgrading its existing and developing new instrumentation, and strengthening its commitment to user science.**

There is every reason to believe that we can succeed. The ALS has led the world both in fundamental research and in inventing revolutionary new instrumentation. These accomplishments have had great significance and long-term impact. Five critical examples that establish the ALS track record of successful innovation include:

- *Nanoscale studies with pump-probe experiments.* The ALS is significantly advancing the real-time, pump-probe study (and possible application to new technologies) of patterned nanomaterials, self-organized nanodots, and nanowires with exotic material properties. One of the unique areas of ALS leadership in nanoscience involves magnetization dynamics of the vortex cores in magnetic materials carried out with the use of such advanced soft x-ray nanospectroscopy tools as aberration-corrected photoemission electron microscopy (PEEM3), scanning transmission x-ray microscopy (STXM), and imaging microscopy (XM1).

- *Ambient pressure photoemission spectroscopy (APPES).* The ALS pioneered the development of ambient pressure photoemission. Through the use of a unique set of transfer lenses and differential pumping, photoemission studies can by conducted at pressures many orders of magnitude higher than previously possible. For example, APPES was crucial in recent breakthrough research allowing the tailoring of properties of bimetallic platinum/nickel electrocatalysts with low platinum content. These alloys are more than 10 times as active by mass (and are more stable) than pure platinum for converting hydrogen to electricity in fuel cell applications.

- *Ultrafast science using electron beam slicing.* The revolutionary concept of slicing an electron beam in the storage ring with a laser modulation to obtain a photon pulse of less than 100 fs was pioneered at the ALS, opening the way for ultrafast x-ray science and time-resolved measurement. In addition to its intrinsic value, ultrafast x-ray science finds direct application in the experimental program at the new Linac Coherent Light Source (LCLS), and it can help identify the science drivers for future-generation photon sources.

- *Complex materials.* The ALS has emerged as the world leader in the study of the electronic structure and the resultant understanding of emergent phenomena in complex materials through the use of angle-resolved photoemission spectroscopy (ARPES) with state-of-the-art commercial Scienta electron energy analyzers, high-precision/low-temperature sample goniometers developed at the ALS, and in situ sample preparation capabilities. The ALS is presently developing the next generation of time-of-flight–based electron energy analyzers that have efficiencies one to two orders of magnitude greater than the commercially available systems. These developments will vastly expand the experimental "phase space" of ARPES to include spin, time and spatial resolution.

- *The Superconducting bending magnet (Superbends).* The ALS pioneered development of superconducting bending magnet technology, which has revolutionized the use of the low-energy storage ring for carrying out world-class hard x-ray science. In addition, protein crystallography experiments using superbends are producing high-impact publications on a par with many other third-generation high-energy storage rings.

To meet the needs of this new era of science that takes us beyond mere understanding and into control—an era in which materials functionality would be designed to specifications and chemical transformation would be manipulated at will—the ALS is ready to position itself for a new round of invention and innovation. **In particular, the ALS will continue to be the leader in soft x-ray microscopy, photoemission, and nano-magnetism, where the ALS has developed novel instrumentation and has established its international standing. In addition, the ALS accepts the challenge of establishing unique scientific programs capable of achieving the breakthroughs in fundamental research that will put new energy technologies within reach.**

To achieve these goals, the ALS has embarked on a program that covers four key areas of development (shown schematically in Figure 1) that together provide the foundation for the ALS core science and technology:

- Replacing four undulators and associated beamlines, which date from the start of operations over 15 years ago, with eight half-length insertion devices and a mix of updated and all-new beamlines. These will provide the most up-to-date facilities for whole new classes of experiments.

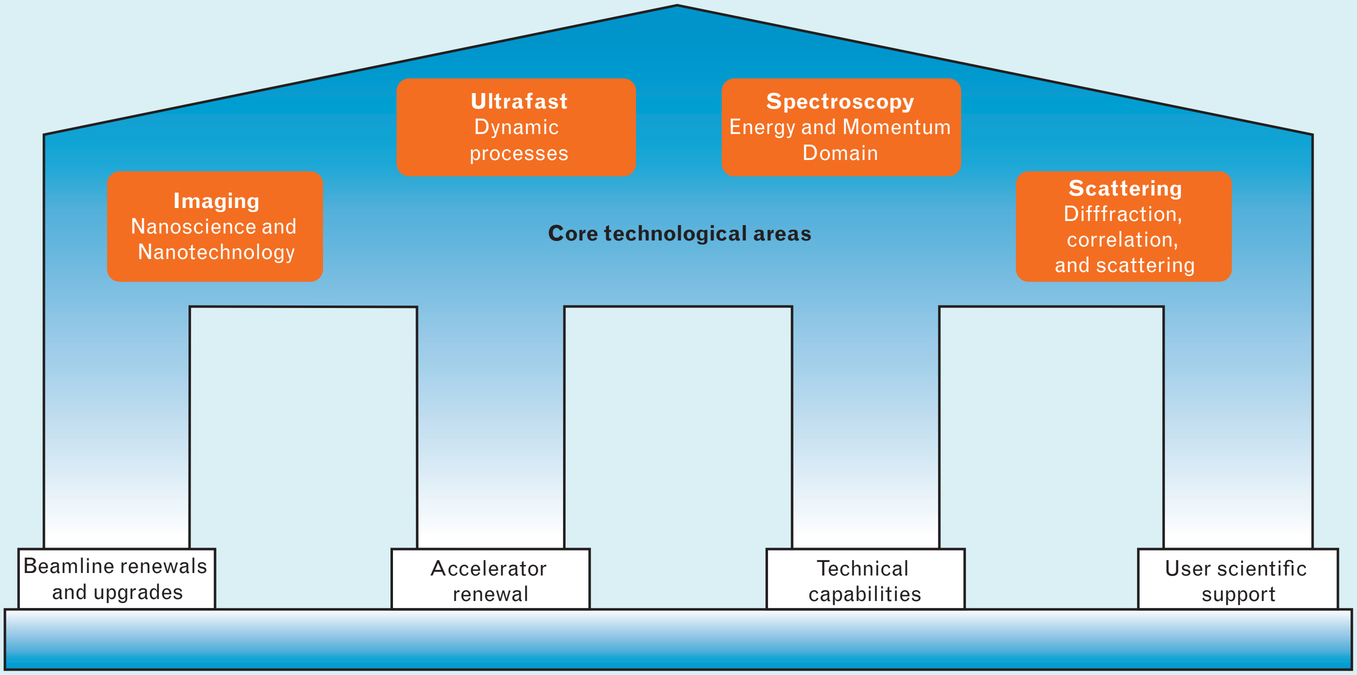


Figure 1. Four key focus areas of the ALS Strategic Plan provide the foundation that supports the ALS core science and technology needed to address the scientific grand challenges and our energy future: 2009–2016.

The new insertion devices will increase the number of undulators at the ALS by fifty percent, will take advantage of the source improvements and, for the first time, provide beams dedicated to coherence experiments, energy research, and nanoscience. The devices will feature optics to preserve the source brightness and end stations and detectors to make full use of the beams delivered. These facilities will assure a continued leadership role for the ALS.

- Renewing the accelerator complex in a cost-effective way so that the ALS can compete effectively with the newer sources.

The renewal of the source starts with the top-off upgrade, which was completed at the beginning of 2009. Top-off not only doubles average current and provides increased stability, it also makes it possible for the ALS to operate at a lower emittance mode and to use insertion devices with smaller gaps for dramatically increased brightness in the 0.5–2 keV region. As a second step in the source renewal process, the RF power system and the control system will be replaced to reduce vulnerability to a major failure of the infrastructure. The third step will be to add a set of sextupole magnets to further decrease horizontal emittance.

- Establishing enabling technical capabilities through an advanced detector program and the Theory Institute for Photon Sciences.

At present, synchrotron light sources far outstrip detectors in terms of capability. The development of novel detectors will provide notably enhanced experimental capability that cannot be over emphasized. The experimental requirements for addressing the grand challenges will require the development of not only sharper experimental tools but also new theoretical methods incorporating concepts beyond those we currently possess in order to foster an environment that is far better suited to tackling the grand challenges.

- Hiring more user support staff to improve productivity and assure that the current high level of support sustainable in the long term. In addition, the ALS will create a pipeline of future beamline scientists, which is necessary for staffing of BES user facilities, through doctoral and postdoctoral programs.

The ALS is recognized as the user facility that provides a very high level of scientific and technical support to all users. However, given the already severe underinvestment in staff for the existing facilities at the ALS, it is essential that we invest in more staff as we build for the future.

The completion of this Strategic Plan will propel the ALS to the next level with new sharper tools that extend our reach into higher energy and spatial resolution, capabilities that will not only maintain the ALS in its leadership role but also provide what the new era of science will require during the next 20 years: increasing our understanding of, and ultimately control of, matter at the level of atoms, electrons, and spins. It is important to emphasize that both advanced synchrotron radiation facilities, such as the ALS will be with the Strategic Plan completed, and the emerging 4th generation facilities based on free electron lasers are essential in addressing the grand challenges for directing matter and energy that when met will position our nation to develop advanced energy technologies.

The capital cost of this plan will be distributed over a seven-year period, during which our users will continue to have access to a large fraction of the beamlines. The new facilities will require additional manpower to run, so that there will be an ongoing need for increased operations support.

This document is a descriptive overview of the ALS Strategic Plan. A preliminary breakdown and time schedule for all the elements of the plan has been prepared; a detailed breakdown, including technical specifications, costs, and priorities, is being prepared for our funding sponsors.

I. INTRODUCTION

The 21st century requires us to take science beyond mere observation. Meeting the challenges posed by our nation's (and the world's) need for safe and secure sources of environmentally clean energy, as well as addressing many other needs of humanity, requires us to be able to design materials and processes that have the properties we specify. For this capability, we must first develop tools that allow us to probe more deeply in order to see functionality at the relevant length, time, and energy scales., and then we must go beyond probing what is there; we must learn to control matter and energy at the molecular, atomic, and subatomic levels.

Over the course of its first 15 years of operation, the ALS has matured and produced an enviable record of scientific output, owing to the world-class capabilities of its beamlines, strong scientific staff, and excellent international user community. In 2007, 1,748 users performed experiments at the ALS. As we look to the next 15–20 years, our strategic plan outlines an ambitious program deisgned to position the ALS at the cutting edge of synchrotron science where it can continue to contribute to society's needs. The result of an ongoing dialogue with our users about their needs, the plan focuses on the 2009–2016 time period and on the needs of 21st century science as discussed in such documents as the Basic Energy Science (BES) Grand Challenges Report ("Directing Matter and Energy: Five Challenges for Science and the Imagination") and the reports issuing out of the BES Basic Research Needs Workshops.[1]

— New and upgraded beamlines

— Upgraded accelerator complex

— Enabling technical capabilities

— User scientific support and future scientist pipeline

To achieve our goals, the ALS strategic plan focuses on four key areas of development: new and upgraded beamlines, an upgraded accelerator complex, enabling technical capabilities, and user scientific support and future scientist pipeline. We can summarize the needs they address as follows:

ALS Core Capabilities Will Address Fundamental Questions

- Where are the atoms and molecules, and how can we control them?
- Where are the electrons, and how can we manipulate them?
- Where are the spins, and how can we direct them?

Facility Upgrades Will Address New Areas of Science

- Tailoring of material properties for energy applications
- Emergent phenomena (quantum confinement and proximity effects) at the nanoscale
- Correlation and complexity in physical, biological, and environmental systems
- Ultrafast phenomena, dynamics, temporal evolution, and self-assembly

Sharper Tools Will Respond to the Needs of Our Users

- Upgrades and improvements to the accelerator
- New and upgraded beamlines with optimized and enhanced capabilities
- New high-performance detectors with higher efficiencies, parallel readout, and imaging capabilities
- Nanospectroscopy and nanodiffraction at a sub-10-nm spatial resolution using diffractive (lensless) and holographic imaging
- Capacity for carrying out two-photon pump and probe experiments
- Development of zone-plate focusing devices using advanced lithographic techniques
- Metrology for creation of the next generation of diffraction-limit-preserving optics to make use of the enhanced electron beam
- Enhanced theoretical support
- Training the next generation of synchrotron scientists

Each of the four key areas is discussed below. Bullets are used to point to specific elements of the plan for which funding will be needed in each of the areas, roughly in priority order.

[1] All BES reports cited in the ALS Strategic Plan can be found at http://www.sc.doe.gov/bes/reports/abstracts.html.

II. NEW AND UPGRADED BEAMLINES

The ALS leads the field in four core technology areas that will be critical in tackling the Scientific Grand Challenges: spectroscopy, imaging, scattering, and ultrafast experimental techniques.[2] Developing new and upgrading the key instruments in these areas will allow the ALS and our users to examine and characterize matter and energy at scales never before achieved. These upgrades and the subsequent discoveries that are made with these instruments will logically lead to the next phase, as reflected in the five Grand Challenges: control of material processes at the level of electrons, design of energy-efficient synthesis of revolutionary new forms of matter with tailored properties, understanding and control of emergent phenomena, mastery of energy and information on the nanoscale, and characterization and control of matter away from equilibrium. Breaking through these challenges provides the scientific foundation for addressing our nation's energy problem, as well as other needs of society.

The proposed suite of new and upgraded beamlines and major instruments are summarized below, with more detailed discussion appearing under the appropriate sections later in this document. The process of developing, installing, and testing the new and upgraded beamlines and instruments will not adversely affect the quality and continuity of the ALS's excellent user service. (Note that realization of the elements of this strategic plan will require additional personnel; e.g., added beamlines due to chicaning will require more beamline scientists.)

Develop MAESTRO nanoARPES and COSMIC coherent scattering and imaging beamlines at Sector 7.0. This will be achieved by chicaning the Sector 7.0 straight section and replacing the existing Beamline 7.0.1 undulator with two half-sized but higher performance devices. With the completion of the COSMIC beamline, the ALS will close down the diffraction microscopy branchline on Beamline 9.0.1 and the coherent scattering/science branchline on Beamline 12.0.2.

Install a second insertion device for the Ultrafast beamline at Sector 6.0. An elliptically polarizing undulator (EPU) will be installed to service the soft x-ray monochromator and thus effectively double the capacity

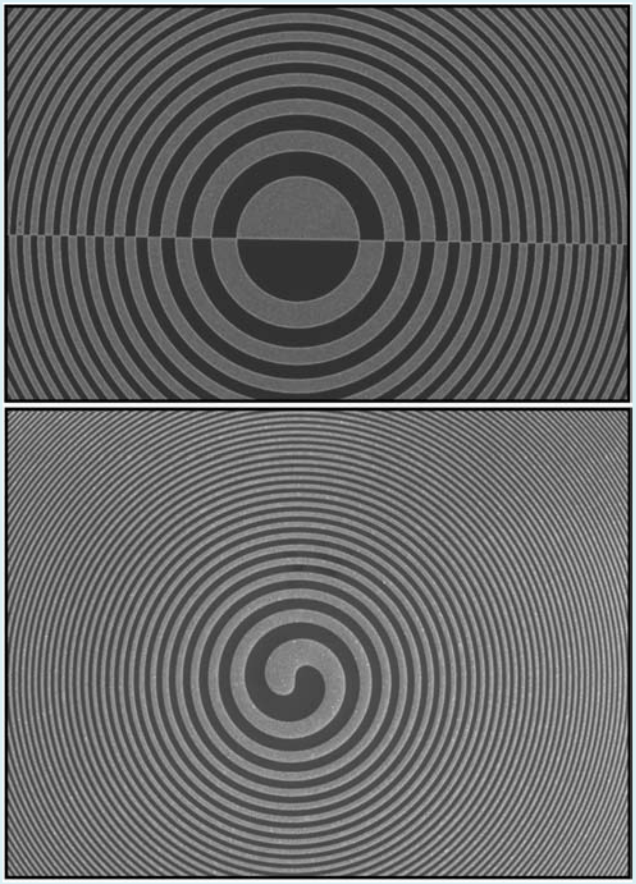


Figure 2. High-resolution zone plate lenses are presently achieving 12-nm spatial resolution for imaging and focusing of soft x-rays and are expected to reach 10 nm and beyond in the near future. Novel zone plates that include additional functionality are making rapid progress. Top: A diffractive structure combines a traditional zone plate and a grating for applications such as differential interference contrast (DIC) imaging and interferometry of materials properties. Bottom: A spiral zone plate combines a traditional zone plate with a spiral phase function to achieve isotropic phase contrast that enhances visibility at edges of any orientation. In addition, the spiral zone plate is compatible with spatially coherent light sources such as a filtered undulator beam and free electron lasers. A third type of structure, not shown, is a cubic zone plate which combines a traditional zone plate with a cubic phase function, permitting greater depth of field for tomographic applications, or use of broader bandwidth radiation to reduce exposure time. Figure courtesy of D. Attwood and A. Sakdinawat, Berkeley Lab.

[2] For a detailed discussion of these techniques, see "Experimental Techniques at Light-Source Beamlines," http://www.sc.doe.gov/bes/synchrotron_techniques/index.htm.

of the ultrafast facility by enabling both branchlines to operate simultaneously (with femtosecond x rays).

Develop Advanced Energy Research (AER) and Soft X-ray Scattering beamlines by chicaning the Beamline 8.0.1 undulator. This upgrade, in partnership with the Molecular Foundry, will allow us to retire two existing endstations (SGM spectrograph and display analyzer).

Chicane the Sector 10.0 straight section and give each branch—the condensed-matter-physics photoemission branch (known as "HERS") and the atomic/molecular physics branch—a separate EPU of its own. Beamline 10.0 is one of the ALS flagship beamlines, with a large number of high-impact publications.

Chicane the Sector 12.0 undulator, allowing the development of the next generation of imaging and scanning nanoscopes on an EPU beamline with sub-10-nm spatial resolution and complete spectroscopy capabilities. One beamline will be available for advanced nanospectroscopy and the second for high-demand angle-resolved photoemission spectroscopy (ARPES). The existing coherent EUV/soft x-ray and MET programs will be located at the second branchline shared with ARPES or moved to a bending magnet beamline.

Replace the existing bending magnet Beamline 9.3.2 with an optimized beamline. Beamline 9.3.2's primary focus is to carry out ambient pressure photoemission spectroscopy (a technique pioneered at this beamline), which requires an extended energy range from 50 to 2000 eV for achieving depth profile capability. With the addition of better refocusing optics along with the new analyzer system, the beamline performance will improve by an order of magnitude, which will provide the necessary next step for the study of kinetics of surface reactions.

Develop a Q-Resolved Inelastic Scattering beamline (QERLIN) in Sector 2.0 to extend the high-resolution resonant inelastic x-ray scattering (RIXS) capability up to 2.5 keV so as to be able to access the most important dipole excitations for complex materials. In doing so, one will also be able to access the momentum transfer q-vector range as far as the Brillouin zone boundary.

Increase the capability of the Vector Magnetometer in Beamline 4.0.2 (pioneered at the ALS) by development of superconducting magnets with a magnetic field of over 5 Tesla in any orientation relative to the sample and photon polarization. The high magnetic field will allow magnetic circular dichroism studies of novel magnetic oxides, which require a much higher magnetic field to reach saturation.

Move the remaining Hard X-Ray Bend Magnet beamlines to unused superbend locations. The recent successful move of the microdiffraction beamline from a bend magnet (7.3.3) to a superbend (12.3.2) has resulted in two orders of magnitude more flux, reducing data collection times and increasing throughput, and an extended x-ray energy range giving an order of magnitude increase in strain sensitivity. This move, which was partially supported by National Science Foundation funds, demonstrates the type of gains that we can expect when we move our existing chemical crystallography, SAXS/WAXS, and Micro-XAS beamlines to superbend sources. We have the user programs, the support staff, the beamlines, and the superbend sources; we just need to link these together.

A. Spectroscopy

Soft x rays are ideally suited for spectroscopic techniques that are used to study the energies of particles emitted or absorbed by samples exposed to monochromatized photons and to measure the electronic structure of materials. These

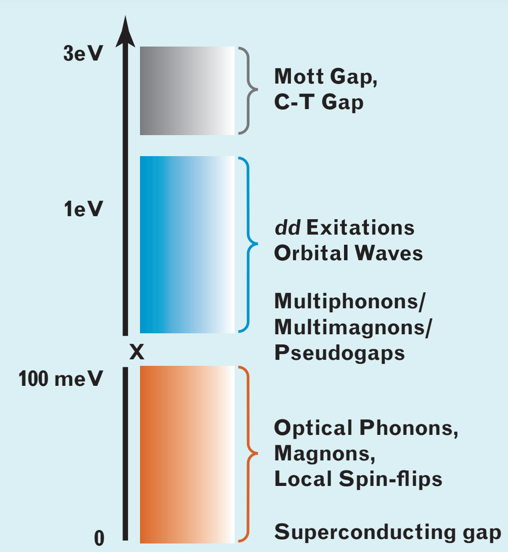


Figure 3. Energy scale of important excitations in complex correlated materials.

techniques can be separated into two classes: electron spectroscopy (photon-in/electron-out) and x-ray spectroscopy (photon-in/photon out).

Electron Spectroscopy

Unraveling Emergent Phenomena for Control of Material Processes at the Electron Level

Angle-Resolved Photoemission Spectroscopy (ARPES) for Measuring Complex Correlations of Electrons in Materials

The ALS is the world leader in the study of the electronic structure of complex materials through the use of ARPES. This is achieved with state-of-the-art commercial Scienta electron energy analyzers, high-precision/low-temperature sample goniometers developed at the ALS, and in situ sample preparation capabilities. The ALS is presently developing the next generation of time-of-flight–based electron energy analyzers that have efficiencies one to two orders of magnitude greater than the commercially available systems. As a result of this ongoing improvement in throughput, we will vastly expand the phase space of ARPES experiments. This will provide us with the capabilities to address the basic research needs for superconductivity and complex systems. (See the BES Workshop report, "Basic Research Needs for Superconductivity.")

During the next seven years, we plan to further strengthen our photoemission capabilities in the following areas:

- Development of a unique facility (Microscopy and Electronic Structure Observatory, or "MAESTRO") for characterization of electronic structure in complex nanostructured materials by replacement of the long undulator at Beamline 7.0 with two half-length EPUs. The MAESTRO beamline will be developed on one of these EPUs, which will feature a higher energy resolution monochromator and specialized endstation (nanoARPES) capable of determining the complete momentum-dependent energy bands of materials down to a size of 50 nm, which is impossible with current techniques. (The second EPU will be used at the other half of Beamline 7.0, the Coherent Scattering and Diffraction Microscopy beamline, or "COSMIC." COSMIC is discussed under "Scattering," below.) The nanoARPES endstation will utilize cutting-edge instrumentation for going beyond observational science to control of material parameters by the development of advanced in situ sample preparation capabilities integrated with high-spatial-resolution

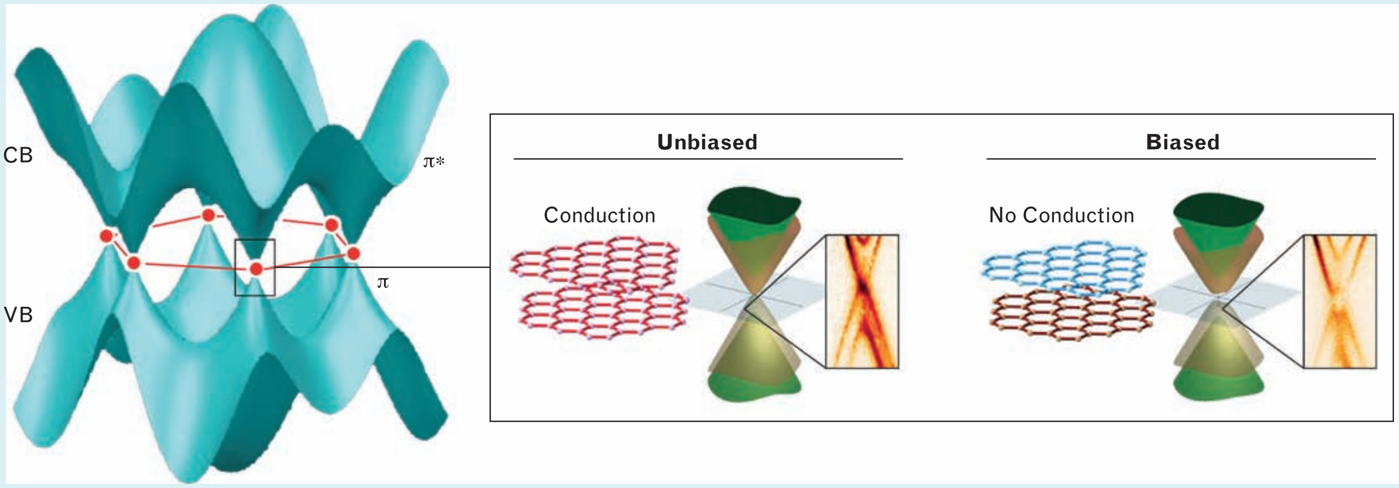


Figure 4. Graphene, a 2D Dirac fermion system. LEFT: Momentum space—dispersions resemble relativistic Dirac fermions, which are responsible for most of the intriguing properties in graphene. (CD = Conducting Band; VB = Valence Band.) RIGHT: "Unbiased"—when the graphene film has no net applied field, the symmetry of the film dictates, and ARPES data confirm, that two electronic bands cross without an energy gap. "Biased"—upon application of an electric field, the symmetry is broken and a gap is opened in the energy spectrum. S.Y. Zhou et al., *Nature Mater.*, **770**, 6 (2007); T. Ohta et al., *Science 313*, **951** (2006); A. Bostwick et al., *Nature Phys*. **3**, 36 (2007).

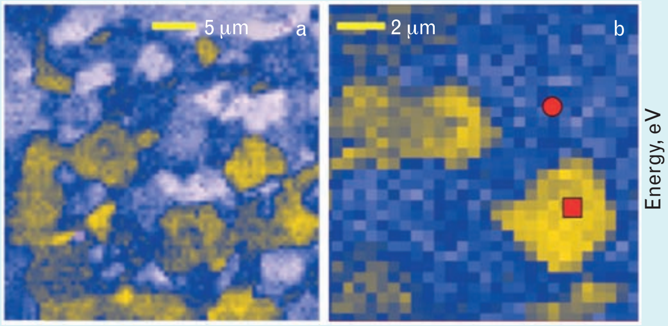


Figure 5. NanoARPES images of polycrystalline graphite, where the domains are individual single crystals of graphite. Figure courtesy of E. Rotenberg, ALS.

electronic structure measurements, data analysis, and theoretical support. It has a strong focus on combinatorial analysis of oxide and metal alloys to understand their phase transition and emergent properties.

- Chicaning the Sector 10.0 straight section and giving each branch—the condensed-matter-physics photoemission branch (known as "HERS") and the atomic/molecular physics branch—a separate EPU of its own for full polarization control and simultaneous operation. Beamline 10.0 is one of the ALS flagship beamlines. The beamline is highly oversubscribed, with research results appearing in a large number of high-impact publications.

  - Upgrading the condensed matter physics beamline will allow development of a monochromator with a broader energy range for control of depth sensitivity and higher energy resolution along with a higher purity of monochromotized radiation. In addition, the highly successful HERS experimental setup will be upgraded with a sample preparation and characterization facility on one branchline. The other branch on this EPU beamline will be optimized to develop new research programs in the areas of spin-resolved and time-resolved ARPES that will make use of both the two-bunch as well as the unique quasi-single-bunch-mode operation at the ALS. There will be two separate cutting-edge technology endstations (presently under commissioning at the ALS) positioned in tandem on this branch. Spin-resolved ARPES is based on time-of-flight (TOF) for energy analysis and exchange scattering for spin selection. This system is the first of its kind and has

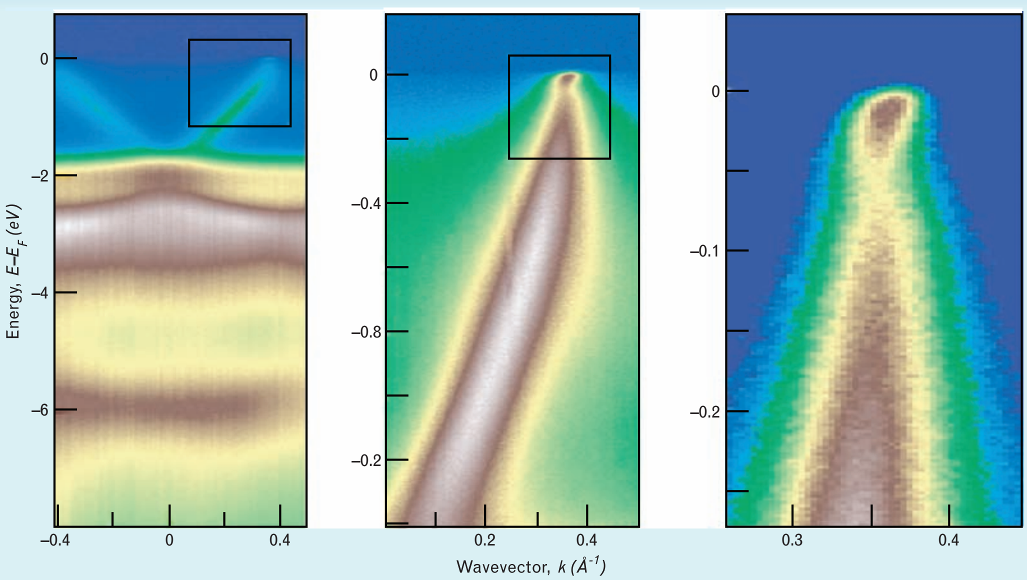


Figure 6. ARPES data for three magnifications plot photoelectron intensity variation with energy (relative to the Fermi level) and momentum in the nodal direction. Momentum is determined from the angle of emission. The need for high energy and angular resolution is apparent upon realizing that the data analyzed in this experiment are those contained in the right arm of the light blue "V" in the left low-magnification image. N. Mannella et al., *Nature* **438**, 474 (2005).

already been successfully tested and proven to have an efficiency three orders of magnitude higher than that of systems using a Mott detector with a dispersive analyzer. Time-resolved pump-probe science will be carried out with the use of a 3D momentum imaging TOF analyzer, which is also expected to perform with an efficiency two orders of magnitude higher than that of widely used dispersive analyzers.

- Developing a dedicated atomic and molecular physics beamline will allow significantly increased capability and the creation of new research areas that are better suited to tackling the Scientific Grand Challenges. The amount of highly successful positive and negative ion research at this beamline will be increased further with the addition of an electron spectrometer that will allow the measurement of differential partial cross sections and angular distributions of the Auger electrons and photoelectrons. A second branchline will be developed with a new experimental station optimized for the study of small quantum systems and clusters, paving the way to understanding the transition from atomic physics to solid-state physics. Installation of state-of-the-art detection systems, already developed by members of an approved program team, will allow the measurement of coincidences between electrons and ions subsequent to ionization of clusters. The complete study of metal clusters, produced by laser ablation, as a function of cluster size will allow investigation of quantum confinement and the evolution of various metal properties from the atom to the bulk.

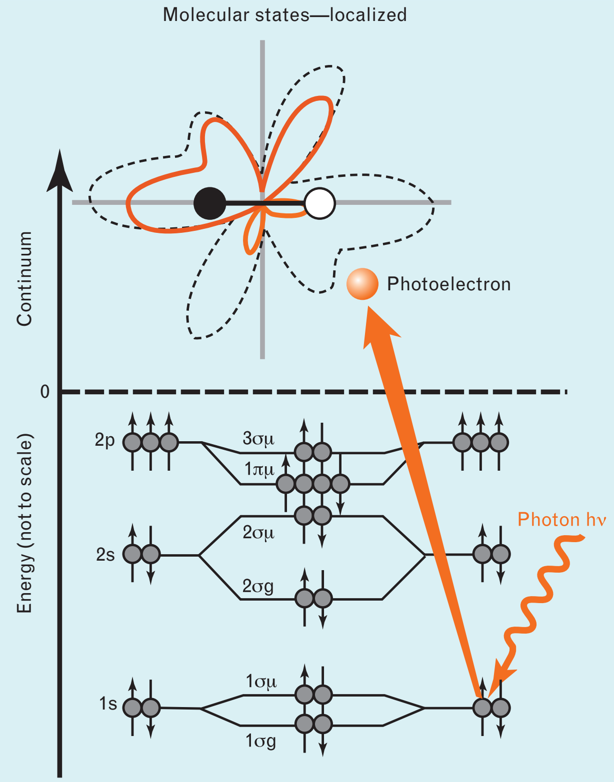


Figure 7. Ultrafast probing of core hole localization by coincident detection of a photoelectron and an Auger electron. Scenario for the case of a K hole localized at one atom: photoelectron is emitted from the atom on the right (the red line in the diagram shows the calculated angular distribution for this photoelectron at 9 eV); the light is circularly polarized, propagating into the plane of the figure. The orientation of the molecule is indicated by the barbell. M.S. Schöffler et al., *Science 320*, **920** (2008).

- The development of a facility for carrying out photoemission with hard x-ray excitation in the 2–10 keV range. Using hard x-ray excitation permits measurements that are much more bulk sensitive, both as to sample composition and valence electronic structure. With cryogenic cooling, ARPES should also be possible over the lower part of this energy range, and element-resolved studies of densities of states will be derivable over the higher part of the range. Using nanometer-scale standing wave excitation, another technique developed at the ALS, will also enhance the ability to study buried layers and interfaces. Photoelectron diffraction at higher energies also provides new potential for studying bulk atomic structure (e.g., of dopants) in complex materials. Hard x-ray photoemission is growing rapidly in several places outside the U.S. and has applications to a broad array of complex materials and layered nanostructures. Having such a facility at the ALS would thus beautifully complement the other photoemission activities at lower energies. We will explore the development of this facility either as a rebuild of bend-magnet Beamline 9.3.1 or as a new superbend beamline.

Ambient Pressure Photoemission Spectroscopy for Tailoring Catalytic Properties of Surfaces for Energy Applications

The ALS pioneered development of ambient pressure photoemission capabilities through the use of a unique set of transfer lenses that allow efficient differential pumping and transfer of the electrons to the analyzer without significant losses. Further improvement of the design, in collaboration with Gammadata Scienta and SPECS Scientific Instruments, is under way and will further enhance the overall efficiency and permit us to reach ambient pressures of up to 20 torr. This will provide the ALS and our users the opportunity to carry out a new class of dynamical studies of chemical reactions at the surface and address the basic research needs as detailed in the BES workshop reports "Opportunities for Catalysis in the 21st Century" and "Basic Research Needs: Catalysis for Energy." Our plan is to:

- Replace the aging bending magnet Beamline 9.3.2 with an optimized beamline having an extended energy range from 50 to 2000 eV, which is necessary for providing depth-profiling capability. In addition, installation of better refocusing optics along with a new analyzer system will further increase by one order of magnitude its detection efficiency and thus provide new capabilities for studying chemical reactions at the surfaces as a function of time. This facility is unique for in situ characterization of many surface properties such as electronic structure, kinetics of diffusion, and surface segregation. Measurements of in situ properties are essential for tailoring properties of novel materials, which are in turn necessary for developing potential applications in such important areas as catalysis and corrosion.

X-Ray Spectroscopy

Electronic Structure Determination of Bulk Materials with a Focus on Energy Applications and Magnetis

Soft X-Ray Absorption, Emission, and Resonant Inelastic Scattering for Measuring Electronic Structure in Complex Systems in an In Situ Environment

Development of in situ sample handling systems and high throughput emission spectrographs (two orders of magnitude higher efficiency than the previous generation) has put the ALS in a unique position to tackle scientific prob-

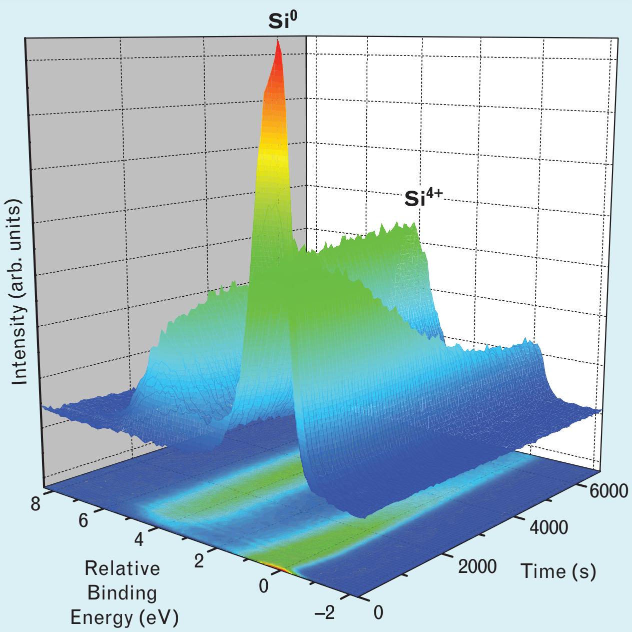


Figure 8. 3D plots of a series of Si 2p core-level spectra taken at an oxygen pressure of 1 torr and at an oxidation temperature of 450°C. Y. Enta et al., *App. Phys. Lett.* **92**, 012110 (2008); M. Rossi et al., *J. Appl. Phys.* **103**, 044104 (2008).

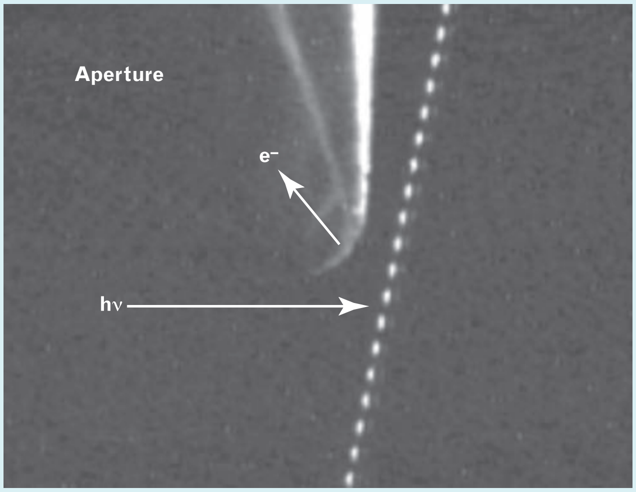


Figure 9. Stroboscopic photograph of a droplet train (dashed line) prepared from a 40% ethanol solution in front of the entrance aperture (diameter 0.3 mm) of the differentially pumped lens system. The illumination is represented by the hn arrow. The parameters are: background vapor pressure 4.3 torr, orifice diameter of the VOAG 50 µm, flow rate 2.2 mL/min, driving frequency of the piezo element 54 kHz, nominal droplet diameter Dd 110 µm, nominal droplet velocity 1870 cm/s. D.E. Starr et al., *Phys. Chem. Chem. Phys.* **10**, 3093 (2008).

lems with energy-related applications. In particular, we would like to understand and tailor properties of materials that are suitable for hydrogen storage as well as investigate band-gap (band-edge) engineering of water-splitting materials and novel multigap materials as photovoltaic suitable for harnessing the energy of the sun. This will help the ALS tackle the basic research needs as detailed in the BES workshop reports "Nanoscience Research for Energy Needs," "Basic Research Needs for the Hydrogen Economy," and "Basic Research Needs for Solar Energy Utilization." We plan to chicane the long undulator of Beamline 8.0 into two EPU beamlines: one (the Advanced Energy Research beamline) will be dedicated to x-ray spectroscopy, and the second will be dedicated to polymer/magnetic scattering (see Scattering section) in partnership with the Molecular Foundry.

- The Advanced Energy Research (AER) beamline will be developed as a high-intensity, wide-energy-range (100–2500 eV) beamline equipped with in situ sample preparation and control systems as well as a high throughput emission spectrograph and electron energy analyzer. The choice of detection system will allow the study of surface, interface, and bulk electronic structures of novel inorganic and biological systems. Combining characteristics of absorption spectroscopy that measures unoccupied conduction band and emission spectroscopy that gives information about the occupied valence band, the beamline will be uniquely equipped for a better understanding of the properties of matter and the tailoring of these properties for renewable energy applications. (Complementary information on bulk electronic structure will be provided by the hard x-ray photoemission facility described above.)

Chemical Dynamics for a Fundamental Understanding of Molecular Growth Mechanisms and New Directions in Chemical Imaging of Organic Aerosol Particles and Biological Cells

The Chemical Dynamics beamline (9.0.2) is a national user facility providing state-of-the-art experimental resources for the study of fundamental chemical processes using vacuum ultraviolet (VUV) radiation. Our motivating scientific goal is to expand the knowledge base required for our nation to provide clean, efficient-energy technologies and a cleaner and sustainable environment (see BES workshop report "Basic Research Needs for Clean and Efficient Combustion of 21st Century Transportation Fuels").

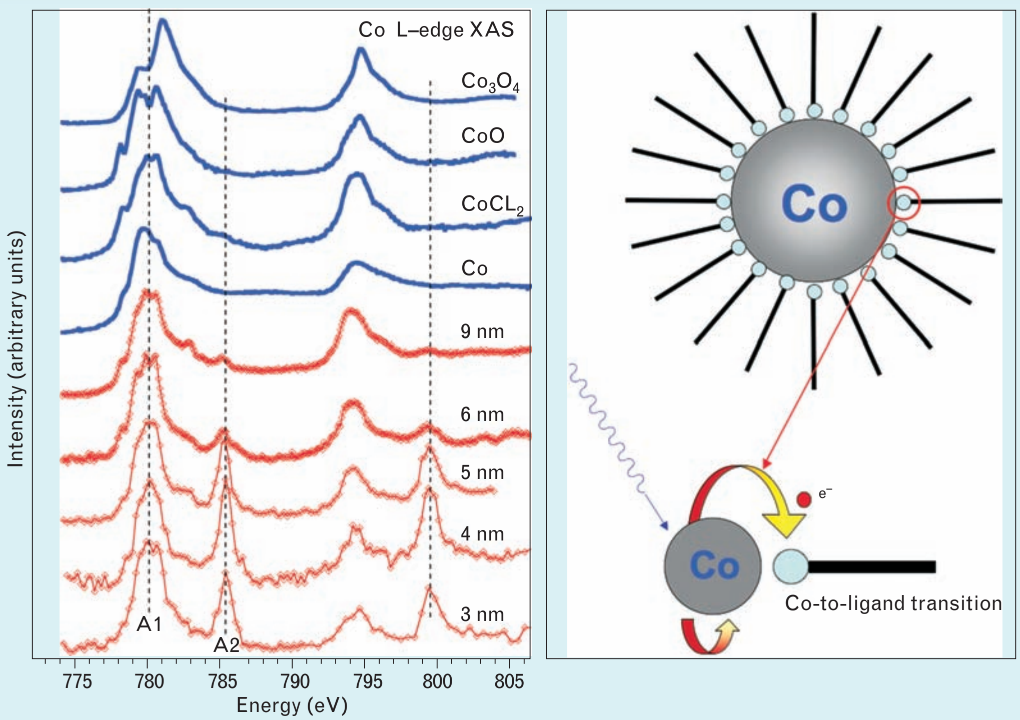


Figure 10. Left: The most notable feature in the spectra of the Co nanocrystals (red curves) is the absorption peak (A2) about 6 eV above the main absorption edge (A1) that is absent in the reference spectra for Co3O4, CoO, CoCl2, and Co metal (blue). Right: Illustration of MLCT transitions between cobalt and the oleic acid or 1,2-dichlorobezene. H. Liu et al., *Nano Lett.* **7**, 1919 (2007).

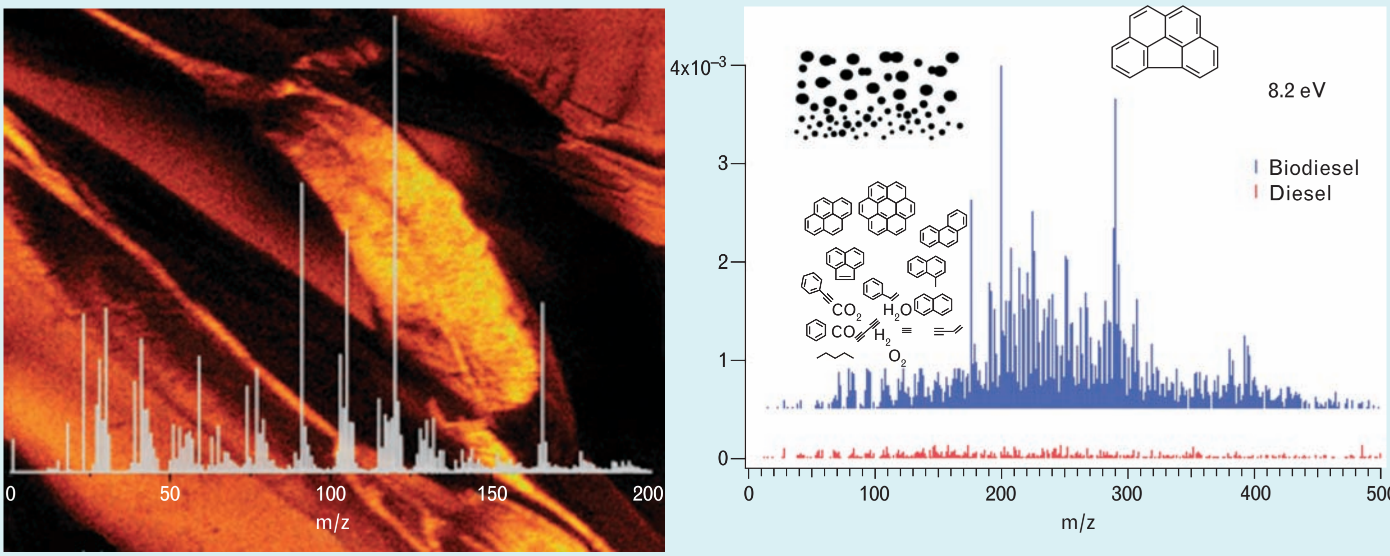


Figure 11. Left: Chemical imaging of cells and aerosols on the nanoscale. Right: Biomolecule aerosol/combustion chemistry. Figure courtesy of M. Ahmed, Berkeley Lab.

Soft X-Ray Vector Magnetometry for Measurement of Magnetic Properties of Novel Materials

EPU Beamline 4.0.2 and Beamline 6.3.1 are primarily used for measurement of x-ray magnetic circular dichroism (XMCD) and magnetic linear dichroism (XMLD) with the aim of tailoring magnetic properties of complex materials. The pioneering development of vector magnetometry at Beamline 4.0.2 has allowed a unique class of experiments, but presently the magnetic field is limited to less than one Tesla due to use of warm electromagnets that require the following upgrade:

- Development of a superconducting vector magnetometer with a magnetic field of over 5 Tesla in any orientation relative to the sample and photon polarization. The high magnetic field will allow the study of novel magnetic oxides that require a much higher magnetic field for reaching saturation. These studies will then lead to a significant increase in the use of the vector magnetometer for the investigation of a new class of novel materials for application in many areas, including spintronics and multiferroics.

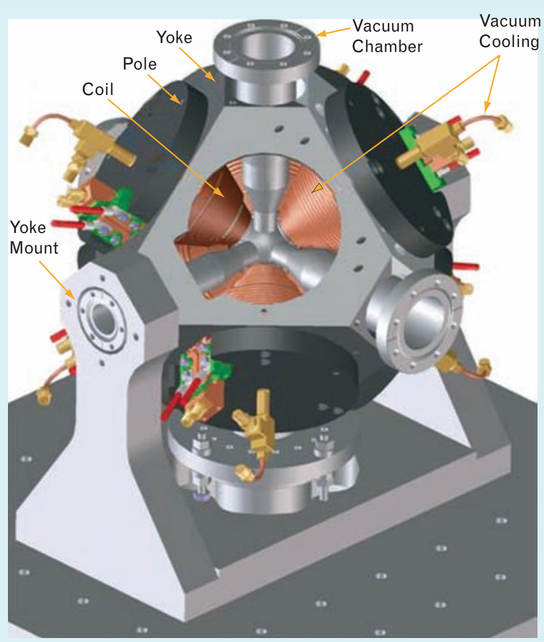


Figure 12. The eight-pole electromagnet developed at Beamline 4.0.2 allows the application of magnetic fields of up to 0.9 T in arbitrary directions. Combined with the elliptically polarizing undulator providing circular and variable linear polarization, this system allows exploration of magnetic dichroism effects in a vastly expanded range of geometries. Figure courtesy of E. Arenholz, ALS.

Infrared Spectromicroscopy

The ALS is at the forefront of environmental and biological science uses of synchrotron infrared (IR) spectromicroscopy and the development of coherent synchrotron radiation as a powerful broadband terahertz (THz) source. Beamline 5.4, which is presently under construction, will have a novel light-extraction port located just outside the bend-magnet source. This will allow for a greater collection angle and therefore improved performance for the IR and exploitation of high-power coherent synchrotron radiation pulses available from both femto-sliced bunches and from a new picosecond bunch mode of operations made possible by the improved ALS lattice. Opportunities also exist for replacing the ALS Sector 5 arc vacuum chamber with one that has been modified to allow a greater vertical acceptance angle (80 mrad) for IR Beamline 5.4. This will provide significantly higher collection efficiencies, particularly at longer wavelengths, at a location perfectly suited for the use of coherent terahertz radiation from the existing laser-sliced electron bunches.

In the longer term (not part of this renewal project), we are examining the scientific drivers and possibility of funding an IR ring, Coherent InfraRed CEnter (CIRCE). We want to go well beyond the diffraction limit into the nanoscale sample size regime. By pushing further into the far-infrared, specifically through the use of high-power coherent terahertz synchrotron radiation, we will be able to explore new effects in this exciting region of the EM spectrum (see Basic Research Needs workshop report "DOE-NSF-NIH Workshop on Opportunities in THz Science"). In the near future, we plan to develop our nanoimaging capabilities as follows:

- Implement the novel technique of scattering and resonantly enhanced scanning near-field optical microscopy for vibrational nanoimaging, which will take advantage of the high brilliance and broad spectral range of the IR beamlines at the ALS. This will provide ultrahigh spatial resolution down to the 10-nm scale, taking advantage of the optical antenna properties of nanoscopic and nanoengineered scanning probe metal tips. This full-spectrum nanoimaging capability will be a revolutionary leap forward.

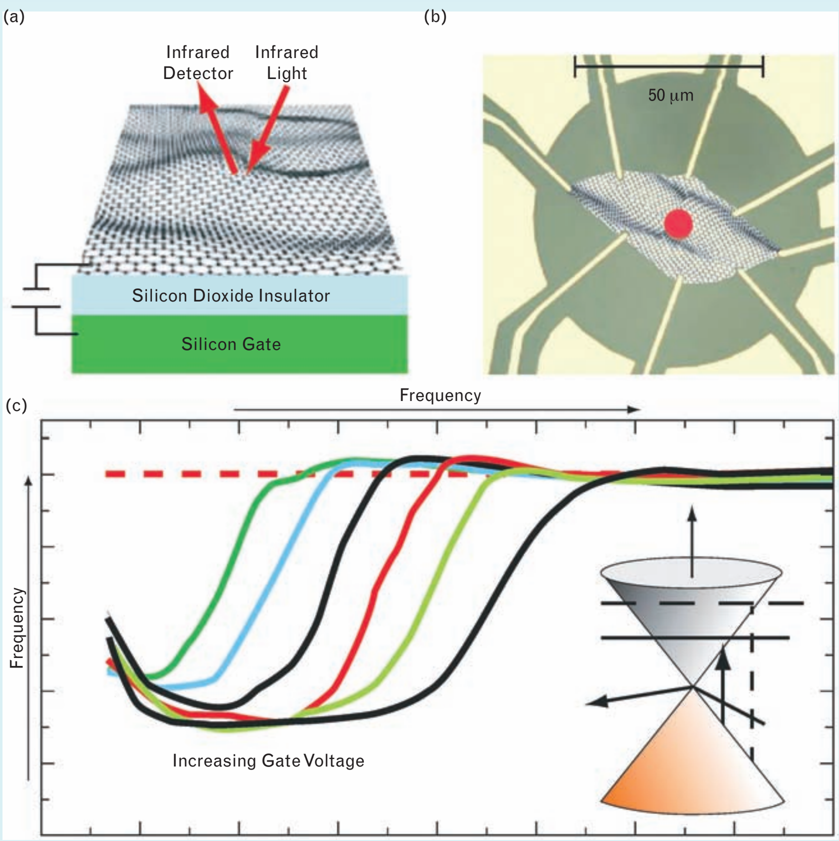


Figure 13. (a) A 50-mm square flake of exfoliated graphene on layers of silicon dioxide insulator and a silicon gate. (b) Gold contacts were attached to the graphene to apply gate voltage. A 10-mm beam of infrared synchrotron radiation (red spot) was focused onto the graphene to measure transmission and reflectance spectra. (c) The conductivity of graphene at different gate voltages, shown in graph as curves of different colors, was observed to change with frequency. At high energies (high frequencies), conductivity and thus absorption was the same for all voltages, but at energies below a threshold at twice the Fermi energy, the absorption of infrared light decreased. Inset: Fermi energy (horizontal lines) and absorption threshold at twice the Fermi energy (vertical arrows) on a graphene band-structure diagram. Z.Q. Li et al., "Dirac charge dynamics in graphene by infrared spectroscopy," *Nature Phys.* **4**, 532 (2008).

B. Imaging

Soft X-Ray Microscopy

Looking at the Nano World through Space and Time

The high brightness of the ALS is uniquely suited to carry out soft x-ray microscopy (and nanoscopy), and the ALS is presently recognized as the world leader in this area of science.

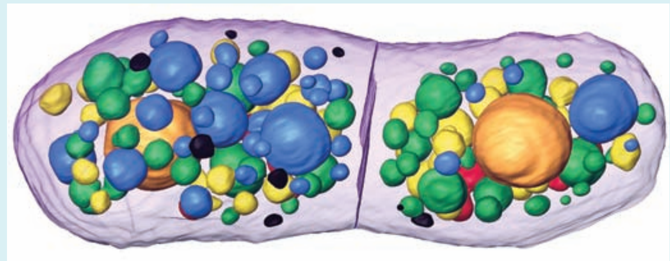


Figure 15. Yeast cell imaged by soft x-ray tomography. Parkinson, et al., *J. Struct. Biol.*, **162**, 380 (2008).

The Photo-Emission Electron Microscope (PEEM3) for Studying Magnetic and Polymer Nanostructures

Photoemission microscopy combines the spectral sensitivity of x rays with the high spatial resolution of electron imaging in a unique manner. With aberration correction, it becomes an almost ideal tool for spectrally resolved imaging, limited only in speed by the efficiency of the photocathode. PEEM3 is advancing the imaging capabilities of photoemission microscopy into the next generation with the use of aberration-corrected electrostatic lenses. The system is installed at undulator Beamline 11.0.1 and is primarily used for real-time and pump-probe study of elemental, chemical, magnetic, and topographical properties of materials.

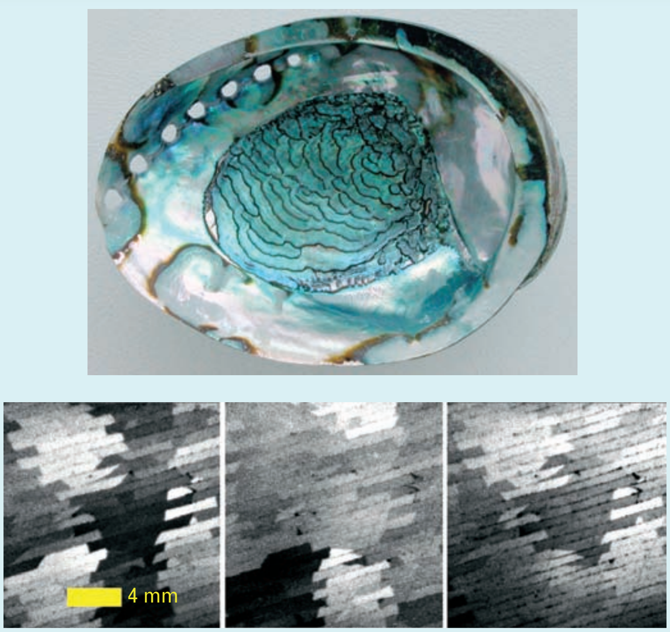


Figure 14. Linear dichroism imaging of nacre. Top: Light microscopy image of nacre. Bottom: X-ray linear dichroism (XLD) images at horizontal, vertical, and diagonal polarization show the columnar growth and the crystal orientation of nacre, mother-of-pearl. The contrast varies with angle between the linear x-ray polarization direction and the stacking direction of the tablets. R. Metzler et al., *Phys. Rev. B* **77**, 064110 (2008).

The upgrade plan currently in progress includes the following. Installation of an aberration-corrected mirror to improve spatial resolution from the current 30 nm to <5 nm and increase transmission by more than an order of magnitude. The availability of spatial resolution down to a few nanometers will allow study of nanoscale patterned structures, nanomaterials and self-organized nanodots, and nanowires with interesting material properties and possible application in new technologies. Other material classes, such as polymers and biomaterials, are sensitive to radiation. These require optimum spatial resolution at very low x-ray exposure, facilitated by the higher transmission and efficiency of the aberration corrected optics that will be available at this beamline.

The Full-Field Imaging Microscope for Magnetization Dynamics and Biological Tomography

The imaging microscope at Beamline 6.1.2 (XM1, operated by the Center for X-Ray Optics) uses a Fresnel zone plate (fabricated by the Center for X-Ray Optics, or "CXRO") for focusing and has demonstrated a spatial resolution of ≈15 nm. This beamline can now carry out magnetism research, and its capacity will be dramatically increased—from quasi-single-bunch operation of the ALS storage ring (see "Accelerator Renewal," above) to carrying out dynamical studies of materials with a long relaxation time during the standard multibunch mode of operation. The newly developed Beamline 2.1 (XM2, operated by Berkeley Lab's Physical Biosciences Division) images biological cells and their dynamics through the use of 3D tomography. To remain the leader in soft x-ray imaging microscopy, we plan the following:

- Development of an imaging microscope on an undulator beamline (by chicaning Beamline 12.0, see details in the following section) with wavefront-preserving optics and an optimized monochromator for nanospectroscopy. We also plan development of the next generation of zone plates for achieving sub-10-nm spatial resolution.

The Scanning Transmission X-Ray Microscope (STXM) for Environmental Science

STXM on Beamline 11.0.2 was developed for molecular environmental science research and is jointly operated by the Chemical Sciences Division and the ALS. This beamline has developed into a leading world facility for carrying out molecular environmental and magnetization dynamic studies at a spatial resolution of 25 nm with bulk sensitivity and unique multibunch operation for pump-probe experiments. This is our most oversubscribed beamline. The ALS upgrade plan therefore demands:

- Chicaning of Beamline 12.0 undulator into two EPU beamlines. One beamline will be dedicated to magnetization research with both a STXM and an imaging microscope (two separate sub-branchlines), and the second beamline will increase capability and provide more access to the existing ARPES program. Improved spatial resolution down to the sub-10 nm range will allow, for example, dynamic images of vortices in a magnetic system with a resolution necessary for pinning down the finer details of spin orientation. In addition, the development of ptychographic imaging facilities on the STXM branchline will enable a novel phase-contrast imaging mode, which will provide high-resolution scanning of samples at resolutions beyond the STXM probe size.

The Coherent Soft-X-Ray Diffraction (Lensless) Microscope for 3D Imaging of Noncrystalline Systems

The high brightness of the ALS provides a unique opportunity to develop imaging methods with the use of coherent radiation for the study of mesoscopic noncrystalline objects, including biological systems (from macromolecular assemblies to cells to entire organisms), with a depth sensitivity that cannot be achieved through diffractive lenses. Research and development efforts in this area of science will

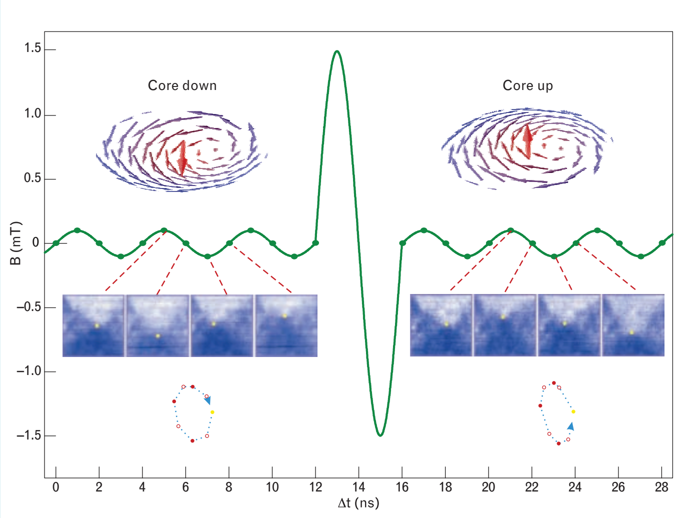


Figure 16. Points on the sinusoidal magnetic field (green curve) correspond to x-ray flashes that record individual frames of the vortex-core movie. When the frames are strung together, they reveal the sense of gyration of the vortex core. Before the burst, the gyration is clockwise, corresponding to a vortex core polarization pointing down. After the burst, the gyration is reversed, and the vortex core polarization points up. B. Van Waeyenberge et al., *Nature* **444**, 461 (2006).

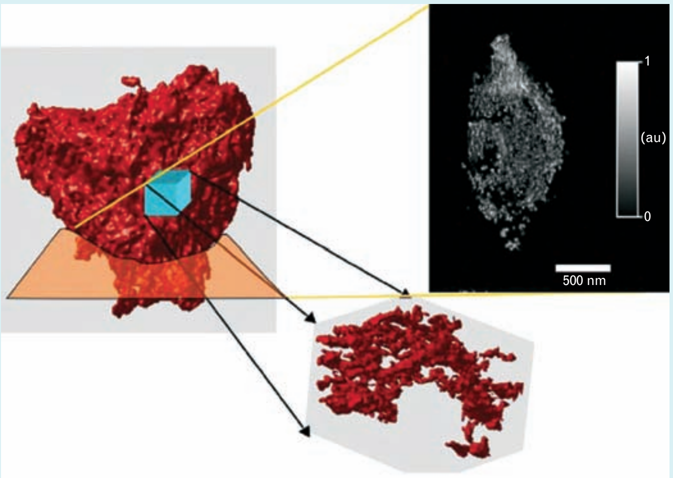


Figure 17. Section and isosurface rendering of a 500 nm cube from the interior of the 3D volume. The foam structure shows globular nodes that are interconnected by thin beamlike struts. Approximately 85% of the total mass is associated with the nodes, and there is no evidence of a significant fraction of dangling fragments. A. Barty et al., *Phys. Rev. Lett.* **101**, 055501 (2008).

also benefit future generations of free electron lasers that provide full transverse coherence and a unique opportunity for single molecule imaging. To support this cutting-edge science, we plan the following:

- Development of an EPU beamline in the second half of Sector 7.0 (coherent scattering and diffraction microscopy, or "COSMIC") that is fully optimized for high coherent power. This will allow us to transfer the existing diffraction microscopy research at Beamline 9.0.1 and the coherent scattering program at Beamline 12.0.2 to the COSMIC beamline. This upgrade and move will improve performance by three orders of magnitude and allow for a new class of experiments at beam energies up to 3 kV. Furthermore, this project can benefit from the R&D effort presently underway for the second branchline of Beamline 8.0.1, allowing for the incorporation of a similar type of magnet.

our core x-ray program or meet local user requirements. The tomography beamline provides three-dimensional imaging of objects with dimensions of tens of millimeters with resolutions of a few microns. Small molecule crystallography allows structure solution of novel materials from crystals too small for structure solution using laboratory systems. Microdiffraction allows characterization of the stress-strain relationship and grain orientation in millimeter-sized samples with submicron resolution. The high-pressure program allows the synthesis of novel materials under extreme conditions, as well as the application of strains of a few percent to materials, while measuring atomic structure and allowing a direct probe of interatomic potentials. Together with the soft x-ray microscopes, these beamlines allow us to study materials on length scales from angstroms to millimeters. Micro-XAS allows mapping of elemental, phase, and oxidation-state composition of samples, typically for environmental science

C. Scattering

The ALS has been a pioneer in the development of superconducting bending magnetic technology and has revolutionized the use of the low-energy storage ring for carrying out world-class hard x-ray science. We have a suite of beamlines for macromolecular and protein crystallography funded by the National Institutes of Health (NIH) and Howard Hughes Medical Institute (HHMI) and run by the Berkeley Center for Structural Biology (BCSB). We also have a suite of ALS-run hard x-ray beamlines located on bend magnet and superbend sources that underpin our core soft x-ray program. We have the opportunity to enhance our hard x-ray program by moving our bend magnet beamlines to vacant superbend slots, and we also have new opportunities to develop a different class of scattering techniques carried out with soft x rays.

X-Ray Diffraction and Scattering

For Measurement of Material Structures, Charge Ordering, and Low-Energy Excitations

Core Soft X-Ray Science Underpinned by Hard X-Ray Beamlines

The ALS hard x-ray beamlines consist of three superbend beamlines: high-pressure (12.2.2), tomography (8.3.2), and microdiffraction (12.3.2); and three bend magnet beamlines: small molecule crystallography (11.3.1), SAXS/WAXS (7.3.3), and micro-XAS (10.3.2). These beamlines either complement

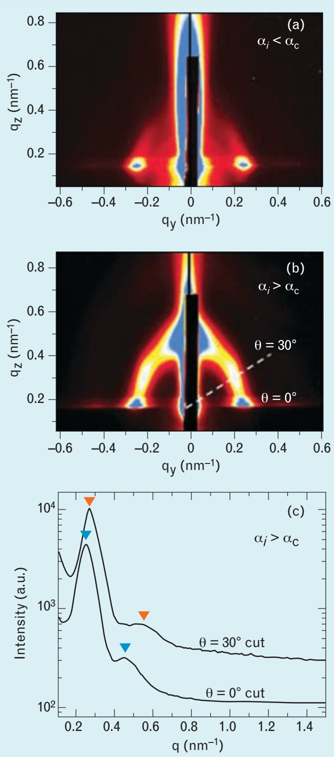
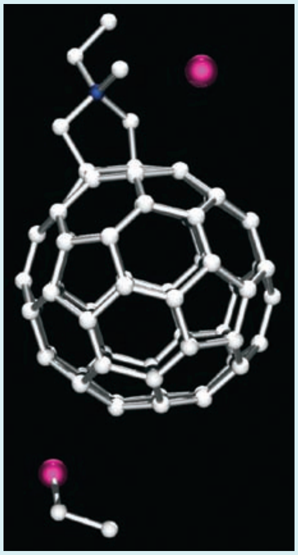


Figure 18. Left: Grazing incidence SAXS images from the surface (a) and bulk (b) of a block copolymer electrolyte membrane showing complete alignment between surface and bulk.



Right: View of half of the asymmetric unit of $[C60(CH_2)2N(CH_3)(CH_2CH_3)]I \cdot CH_3CH_2]$. This is the first example of a crystalline salt of a cationic fullerene that shows a bilayer. C.J. Chancellor et al., *Cryst. Growth & Des.* **8**, 976 (2008).

and materials studies, and as such provides complementary information to soft x-ray microscopy techniques.
SAXS/WAXS allows the study of materials on length scales from angstroms to microns and is extremely useful in simultaneously characterizing atomic- and nano-sized correlations during, for example, the solidification of a polymer or the self-assembly of nanoparticles. The hard x-ray SAXS/WAXS program probes bulks and near-surface structure and is complementary to the soft x-ray SAXS/WAXS program, which probes ultra-thin polymer films and surfaces with chemical specificity.

Significant gains in performance could be achieved by moving the three hard x-ray beamlines currently on bend magnets to superbends. For small molecule crystallography, the high energy provided by the superbends will improve structure determination precision and the flux increase will allow work on smaller crystals. Moving to a superbend would result in a hundredfold increase in flux, reducing data collection times from a few hours to tens of minutes. Moving the micro-XAS beamline to a superbend would also reduce scan times due to the higher flux, allowing larger areas to be scanned or smaller areas to be scanned more finely. Also, the higher energy x-rays would allow environmentally important elements such as uranium and the heavy metals to be detected. Moving the SAXS/WAXS beamline to a superbend would allow us to reduce our temporal resolution in SAXS from seconds to the millisecond range and open up the use of anomalous scattering to separate the effects of different elements.

Structural Biology for Macromolecular Crystallography and Imaging of Complex Systems under In Situ Conditions

The Berkeley Center for Structural Biology (BCSB) at the ALS (funded through the National Institutes of Health, Howard Hughes Medical Institute, and commercial and academic entities) and other beamlines (funded through DOE/BER, National Institutes of Health, and commercial/academic entities) plan to further increase their capabilities for high-resolution macromolecular crystallography and structure determination of complex large systems. In addition, a new array of diffractive imaging and small-angle x-ray scattering (SAXS) methods will be used in combination with advanced spectroscopies. To achieve these goals, the BCSB plans to install a short superconducting undulator in the straight section of Sector 5 alongside the wiggler (changing the source of Beamline 5.0.2 from a wiggler to an undulator). This configuration would permit all current operations to continue while also adding a highly parallel x-ray beam on Beamline 5.0.2.

The BCSB is also in an ongoing process to further improve automation and remote data collection, upgrade optics, and install high-performance detectors. Expansion of SAXS capabilities include implementation of a liquid pipetting robot at Beamline 12.3.1 (SIBYLS), enabling complete data collection on a project in less than 15 minutes. A more comprehensive automated system is being developed that will coordinate sample loading, data collection and data processing. The next step is to fully optimize the newly realized throughput potential of the beamline.

Optics improvements include upgrades to Beamline 8.2.1, which will allow the generation of an x-ray beam approximately five times brighter than at present. This will facilitate the analysis of smaller crystals with larger unit cells. Detector improvements include a prototype SAXS detector, which is planned for testing and evaluation on Beamline 12.3.1 in 2009. It will have a 64 ms time resolution and single-photon counting, up to 0.3 MHz count rate/pixel, and no "dead" spots. A new 40 cm x 20 cm detector prototype will be developed and installed on Beamline 4.2.2 in 2009, with a 0.25 second readout that is approximately five times more sensitive than current CCD systems. However, in the next

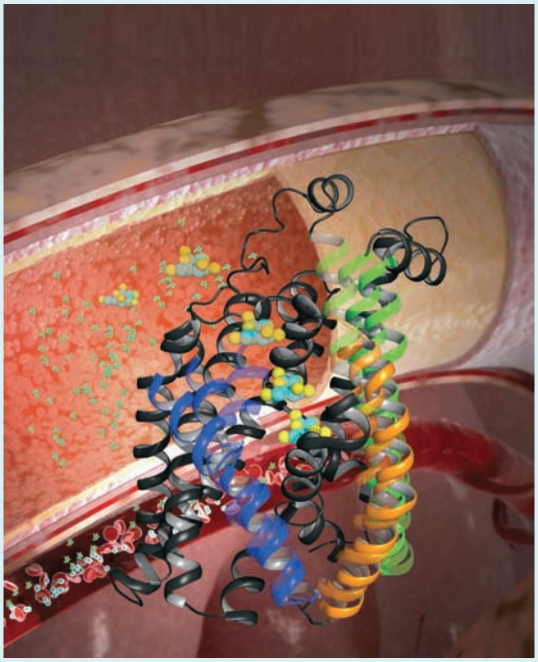


Figure 19. Colored corkscrews represent 3D models of a sodium glucose transporter protein, which propels glucose from the intestine back into the bloodstream. S. Faham et al., *Science 321*, **810** (2008).

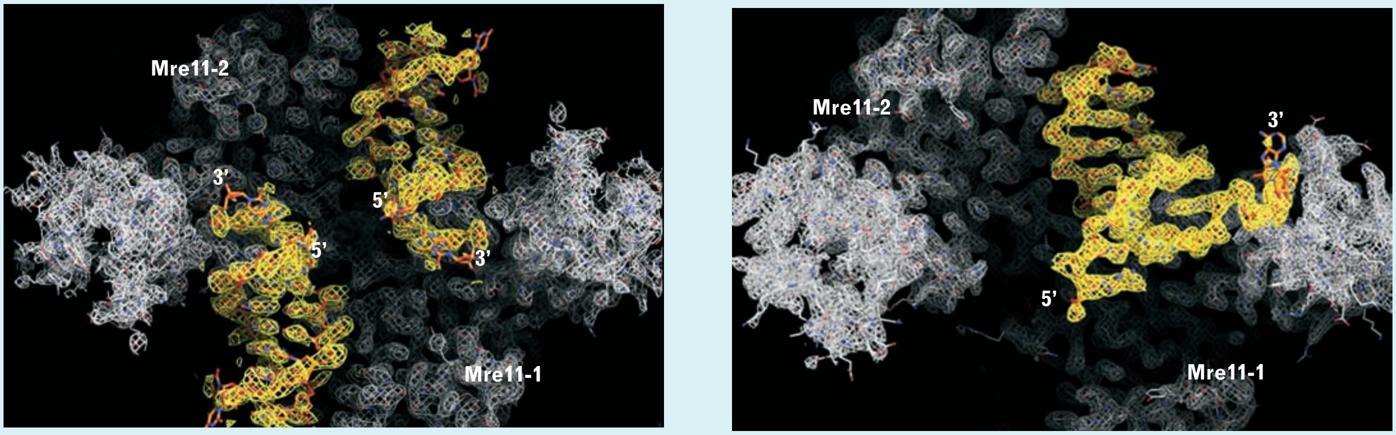


Figure 20. Mre11 proteins initiate repair of double-strand breaks in DNA, either on two ends of broken DNA brought together (synaptic complex at left) or at the site of a collapsed fork in replicating DNA (branching complex at right). J. Taner et al., *Cell* **135**, 97 (2008).

five years, resources will be needed to increase protein crystallography beamline throughput by installing faster, more sensitive detectors, which are currently in academic or commercial development. There can be a great synergy between the needs of the biological imaging community at the ALS and DOE-funded ALS/LBNL detector development efforts.[3]

Dynamic Coherent Scattering and Soft X-ray Correlation Spectroscopy

The goal of this research program is to probe spatiotemporal (space and time) hierarchies in materials systems that lead to complex mesoscopic behavior. The present focus is to investigate local exchange, hopping, and vibronic interactions in transition metal complex oxides (manganites and high Tc superconductors) in order to understand the spatial and temporal aspects of charge, spin, and orbital ordering with the use of dynamic coherent scattering and soft x-ray correlation spectroscopy. This research effort is presently being carried out at the coherent scattering branchline at Beamline 12.0. We plan to move this research effort to one of the branchlines on the proposed COSMIC beamline (shared with diffractive imaging, as described above). This move will provide three orders of magnitude more coherent power and thus tremendously improve our capabilities, which will make the ALS one of the world's leaders in the dynamical study of complex inhomogeneous materials.

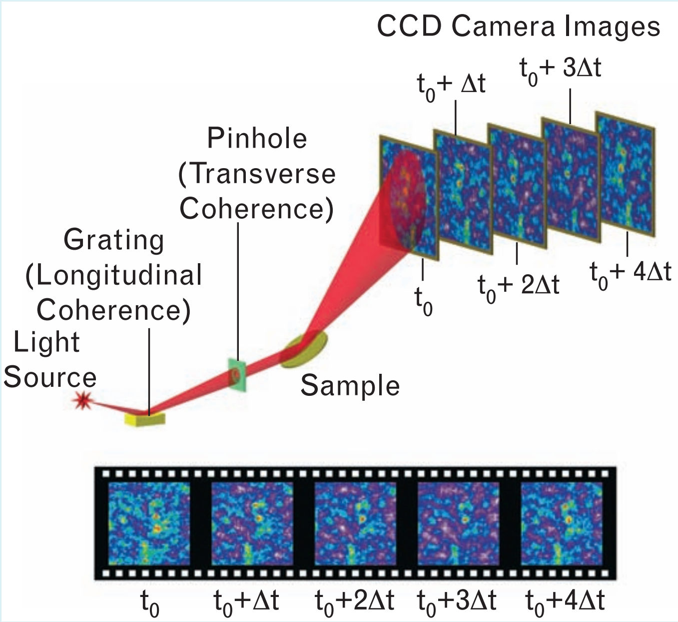


Figure 21. Movie of the "speckle" pattern (frames were taken 12 seconds apart, and each frame averaged 10 seconds) from the orbitally ordered domains in the cubic manganite $Pr_{0.5}Ca_{0.5}MnO_3$, recorded at the ALS. The slow dynamics of the image indicates that the domain walls are pinned by disorder, but the nature of the pinning is unknown. J.J. Turner et al., *New J. Phys.* **10**, 053023 (2008).

[3] To view the Center for Structural Biology's complete plan for renewals and upgrades, go to http://bcsb.als.lbl.gov/wiki/images/1/12/ALS_Structural_Biology_Strategic_Plan_9-8-08_rev.pdf.

High-Resolution Inelastic Scattering with Soft X-Rays: From MERLIN to QERLIN

High-resolution resonant inelastic x-ray scattering (RIXS) is the technique of choice in measuring low-energy coupled excitations that are crucial to understanding the behavior of strongly correlated electron systems. The MERLIN (meV Resolution Beamline) design employs a number of novel features to deliver ultra-high energy resolution ($R \approx 100{,}000$) with the use of an undulator beamline that has full polarization control from a quasiperiodic EPU in the VUV energy range below 150 eV. While MERLIN is designed to provide the ultimate energy resolution for inelastic x-ray scattering, there is a need to extend the capability to higher energies.

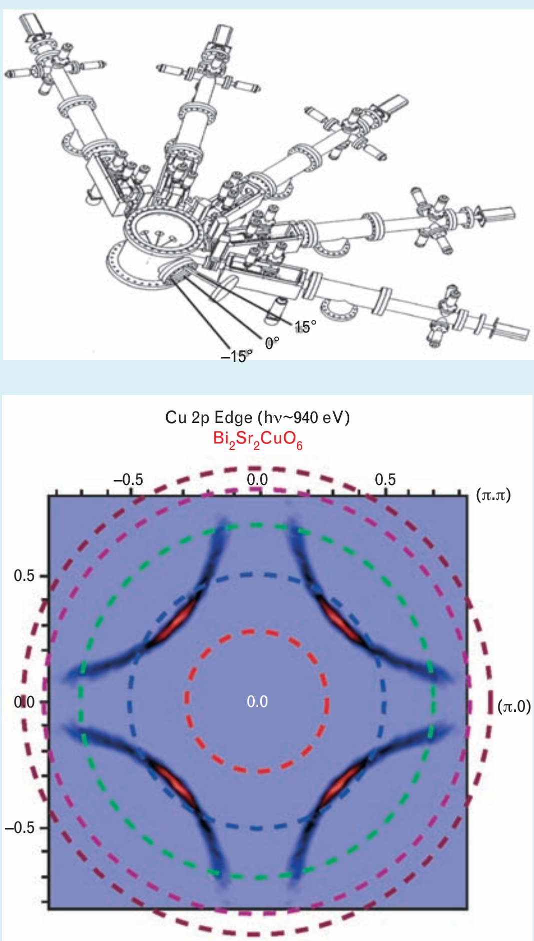


Figure 22. By combining rotation of the chamber and five mounting ports, one can perform momentum-resolved RIXS. Figure courtesy of Y.D. Chuang and Z. Hussain, ALS.

- Development of a Q-Resolved Inelastic Scattering beamline (QERLIN) to extend the capability of high-resolution RIXS investigation up to 2 keV, so as to be able to access the most important dipole excitations for complex materials, i.e., the 1s-2p of carbon and oxygen, the 2p-3d of transition metals, and the 3d-4f of the rare earths. In doing so, one also can naturally access the momentum transfer q-vector range out to the Brillouin zone boundary. The elemental, spin, and orbital sensitivity of resonant soft x-ray scattering offers a unique combination of capabilities that is applicable in the presence of external magnetic/electric fields and can also provide bulk sensitivity. We plan to build on the experience with MERLIN in designing the new facility at Sector 2.0 with a rearrangement of accelerator components. Incorporation of a second endstation on this beamline will allow for very high-resolution photoemission in the range up to 2 keV.

Soft X-Ray Resonant Scattering of Polymer and Complex Oxides in Partnership with the Molecular Foundry

Small-angle x-ray scattering (SAXS) combined with wide-angle x-ray scattering (WAXS) is a powerful tool for the study of the structure of matter on length scales from the atom to the micron. SAXS/WAXS is commonly carried out with hard x rays. However, the use of soft x rays allows scattering experiments to be carried out under resonant conditions that increase sensitivity by about two orders of magnitude for many systems. Therefore, combining spectroscopy with scattering makes this technique extremely powerful for the structure determination of surfaces and interfaces in soft matter; and spin charge, lattice, and orbital degrees of freedom in transition metals. We plan to develop the following soft x-ray scattering facilities on the chicaned second undulator at Beamline 8.0.1:

- Development of a soft x-ray scattering branchline for the study of soft matter, such as polymer and biological systems, in partnership with the Molecular Foundry. With this experimental setup, users will be able to carry out resonant scattering experiments in situ under a controlled environment.

- The second branchline on this EPU beamline will be dedicated to the resonant scattering study of complex transition metal oxides. The capability of applying magnetic field of up to 7 Tesla (presently under development with a unique configuration highly suitable for scattering) will allow the investigation of magnetic-field-stabilized phases and field-induced phase transitions in complex oxides.

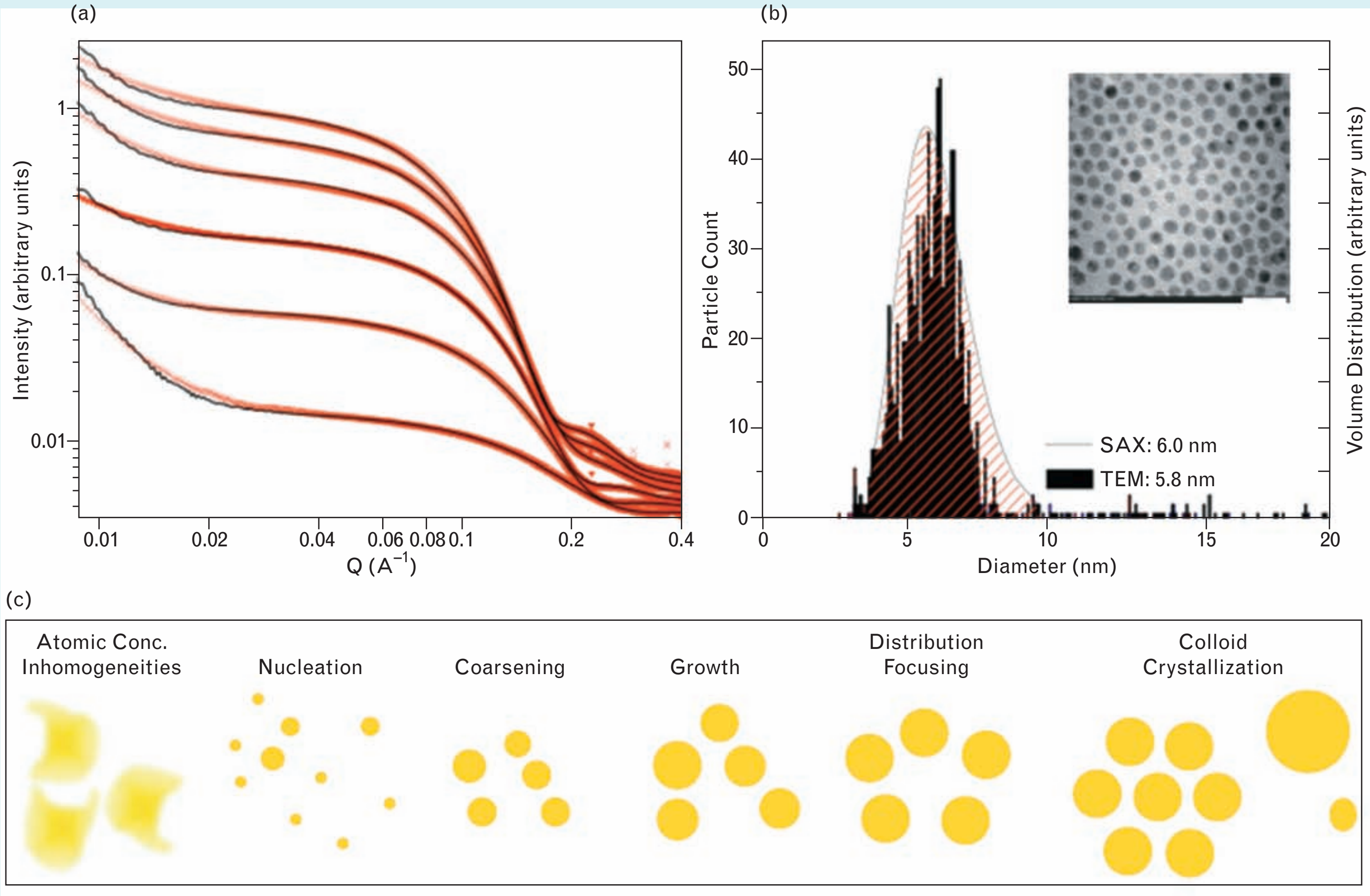


Figure 23. Thermodynamic and kinetic factors that determine morphology and Au particle growth and self assembly. Measurements taken at the SAXS/WAXS Beamline 7.3.3 by H. Koerner, R. Vaia, and R. MacCuspie (Air Force Research Laboratories, WPAFB, Ohio).

D. Ultrafast

Ultrafast X-Ray Facility Using Electron Beam Slicing

Time Evolution of the Formation/Breaking of Bonds and Phase Transitions

This facility is optimized for the generation of femtosecond x-ray pulses. At a millionth of a billionth of a second, this is the timescale at which chemical bonds are formed or broken, or materials transition from one phase to another. The Ultrafast X-Ray Facility is the first such facility at a synchrotron radiation source, and the present science program encompasses atomic and molecular (AMO) physics, condensed matter and correlated materials, solution-phase molecular dynamics, and warm dense matter. Much of the ultrafast x-ray science and time-resolved measurement techniques developed on the Ultrafast Facility beamlines will find direct applications in the experimental program at the new Linac Coherent Light Source (LCLS) under construction at the Stanford Linear Accelerator Center (SLAC). The tunability and high repetition rate of the ALS Ultrafast Facility will continue to be relevant for spectroscopy experiments. Presently, there is one insertion device feeding two branchlines—one optimized for the soft x-ray region and one for the hard x-ray region. We plan to develop the following:

- A second insertion device (an EPU) will be installed to service the soft x-ray monochromator. These will effectively double the capacity of the facility by enabling

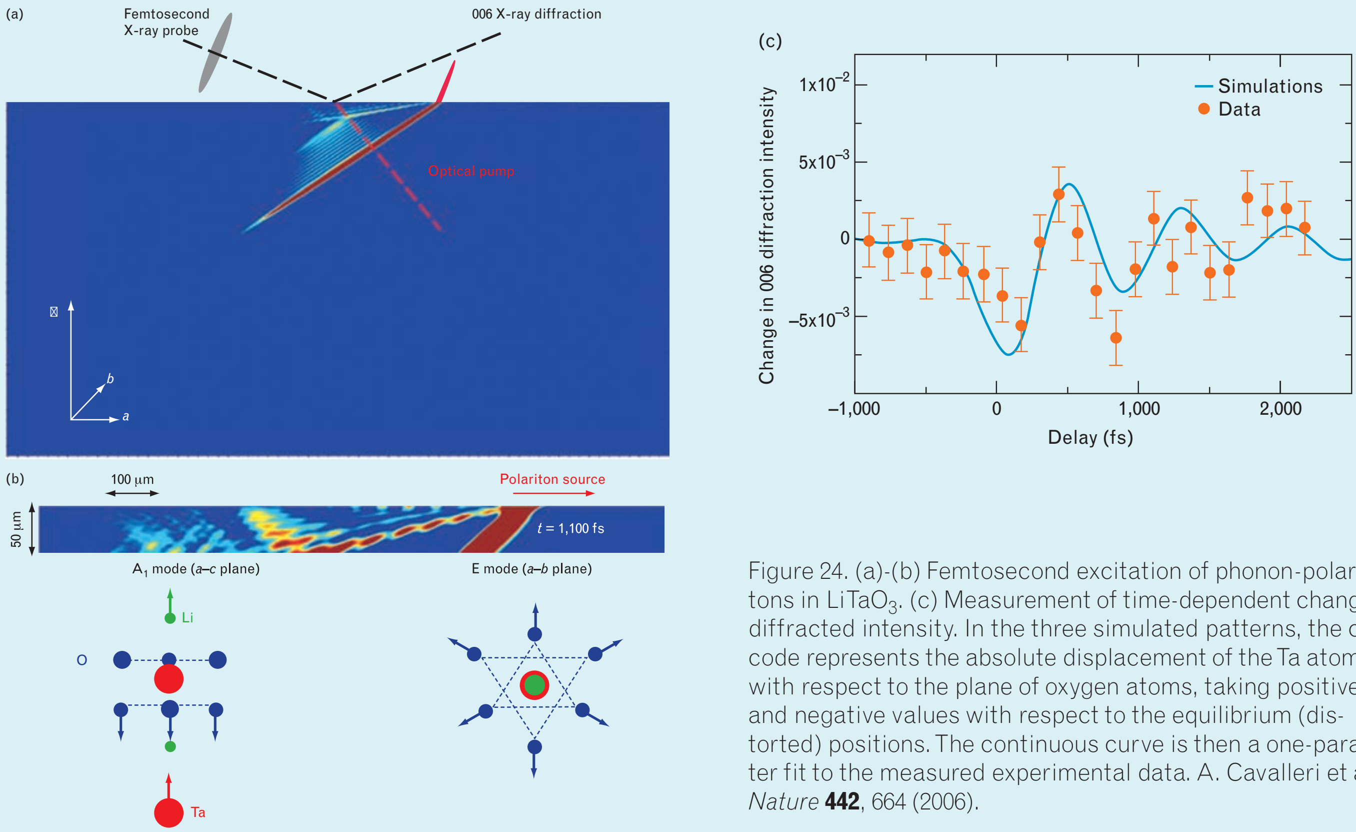


Figure 24. (a)-(b) Femtosecond excitation of phonon-polaritons in $LiTaO_3$. (c) Measurement of time-dependent change in diffracted intensity. In the three simulated patterns, the color code represents the absolute displacement of the Ta atom with respect to the plane of oxygen atoms, taking positive and negative values with respect to the equilibrium (distorted) positions. The continuous curve is then a one-parameter fit to the measured experimental data. A. Cavalleri et al., *Nature* **442**, 664 (2006).

both branchlines to operate simultaneously (with femtosecond x-rays). The straight section has already been modified to accommodate a second insertion device. The polarization control provided by an EPU will open up a new area of ultrafast research on magnetization and spin dynamics using linear and circular dichroism spectroscopy.

In addition, the Ultrafast Facility will benefit from key developments in two other program areas:

Two new operating modes (as part of the ALS "Accelerator Renewal," outlined in detail at the beginning of this document): (1) pseudo-single-bunch operation will enable ultrafast measurements at a high repetition rate with relaxed demands on gated detectors, thereby providing significant improvements in detection efficiency; and (2) the new ALS low-alpha lattice will enable time-resolved measurements with < 10 picosecond resolution, with a substantially higher flux and repetition rate than is now available.

High-speed gated 1D and 2D detectors (provided through the "Detector Development Program," see below).

III. ACCELERATOR RENEWAL

The ALS produces light over a wide spectral range for users from far infrared (IR) to hard x rays with the core spectral region in the ultraviolet (UV) and soft x-rays region. In this core region (relevant to life-science chemistry, catalysis, surface science, nanoscience, and complex materials), the ALS is the national leader and at the forefront of synchrotron radiations sources worldwide. The quality of the photons is intimately connected to the performance of the accelerator complex. The plan for accelerator renewal is to continually upgrade this complex with the goal of keeping the ALS at the forefront of synchrotron radiation sources. The proposed upgrades are summarized below followed by more detailed discussion under the appropriate section:

Higher horizontal brightness. Recent studies, using newly developed and powerful simulation techniques, have shown that with the modest upgrades of the ALS lattice, it is possible to double the horizontal brightness in the insertion device beamlines and superbend sources, as well as a tripling of the brightness in the central bend magnet beamlines. For experiments such as microscopy, the increase in brightness will result in the increase in throughput and or performance by a similar factor.

New Storage ring operational modes. The ALS has successful tested a novel mode of operation, known as quasi-single bunch operation, that will simultaneously satisfy the needs of high-flux/high-brightness users and dynamics/time-of-flight users. In this mode, one bunch out of 328 bunches in the ring is kicked onto a different vertical closed orbit. By spatially separating the light from this bunch from the main bunch train in the beamline, it is possible to have quasi-single bunch operation all year round.

Improved photon beam stability and control. Beam stability (orbit, beam size, and current) is one of the most important performance parameters that define a synchrotron light source. There are increasing demands from the user community to improve the stability in amplitude and in frequency as it increases the sensitivity at which one could carry out higher precision experiments with low signal strengths. Targeted upgrades in controls and diagnostics will provide state-of-the-art beam stability

Enhanced reliability of the accelerator complex. To ensure that the accelerator complex provides reliable operation for the next twenty years, we will need to upgrade or replace a number of critical systems including the storage ring klystron which has passed its age and its control system which relies on obsolete technology.

New insertion device (ID) R&D. The ALS has recently developed a quasi-periodic undulator that allows suppressing higher order components and thus increasing the purity of the synchrotron beam for high precision experiments. We plan to continue our innovative effort for carrying out R & D for non-mechanically-moving EPUs and/or improved field shapes as well as superconducting undulators for achieving ultimate brightness in the soft x-ray range.

A. Higher Horizontal Brightness

In a storage ring, the horizontal beam size and divergence is much larger than the vertical beam size and divergence. Many experiments at the ALS would benefit by increasing the horizontal beam brightness. Recent studies, using newly developed and powerful simulation techniques,

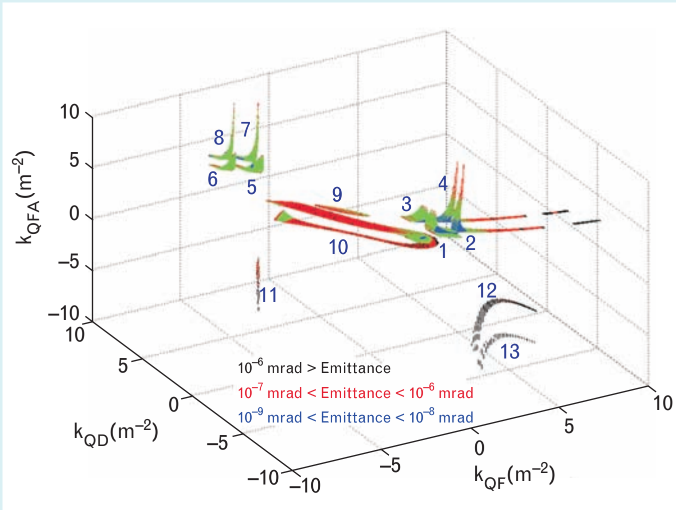


Figure 25. GLASS analysis showing all stable magnetic lattice settings and regions of low emittance. Figure courtesy of D. Robin et al., ALS.

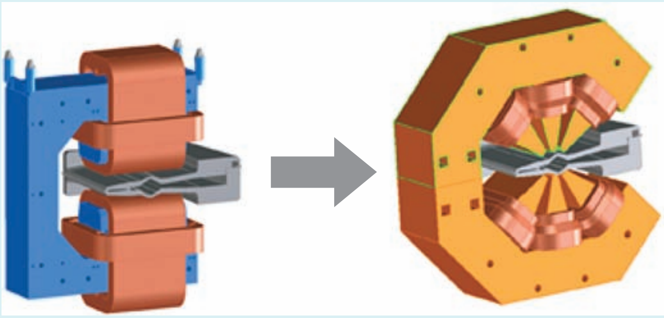


Figure 26. Corrector magnets replaced with sextupole/corrector hybrid magnets. Figure courtesy of D. Robin et al., ALS.

are examining the limits to horizontal brightness. These simulations have already shown that the present ALS settings are far from optimal. With modest lattice upgrades, it is possible to increase the horizontal brightness. For instance, the inclusion of additional sextupole field components in the storage ring opens the door to a doubling of the horizontal brightness in the insertion device beamlines, and superbend sources, as well as a tripling of the brightness in the central bend magnet beamlines. Also, the additional sextupoles would allow the ALS to operate in a shorter pulse (quasi-isochronous) operational mode, similar to BESSY-II. The lattice upgrade plan is as follows:

- Upgrade the storage ring lattice to include new sextupole components without using any additional straight section real estate. This can be done by replacing ~50 straight section corrector magnets with hybrid magnets having both corrector and sextupole components.

B. New Storage Ring Operational Mode

At present, it is not possible to simultaneously satisfy the needs of high-flux/high-brightness users and dynamics/time-of-flight users. Therefore, the two communities perform their experiments at different times using different bunch-fill patterns (90% of the time for high flux and 10% for dynamics). The inability of these users to operate simultaneously greatly limits the effectiveness of these fill patterns. This limitation can be overcome by tailoring the property of individual bunches. To that end, a new mode of operation, known as quasi-single bunch operation, has recently undergone testing. In this mode, one bunch in the ring is kicked onto a different vertical closed orbit. By spatially separating the light from this bunch from the main bunch train in the beamline, it is possible to have quasi-single bunch operation all year round. The initial results look very promising, and we may be seeing only the beginning of what is possible. Our plan is to upgrade and install equipment to further exploit these possibilities:

- A series of fast kicker magnets will be installed in various locations around the ring. This will allow for different modes of quasi-single bunch operation, including lower repetition rates and local bumps. In order to conserve space, a special kicker magnet needs to be developed that fits inside the vacuum chamber at the same location as (or near) the fast corrector magnets. Also, improvements will be made in the bunch cleaning system that will maintain low current gaps with minimal orbit disturbance.

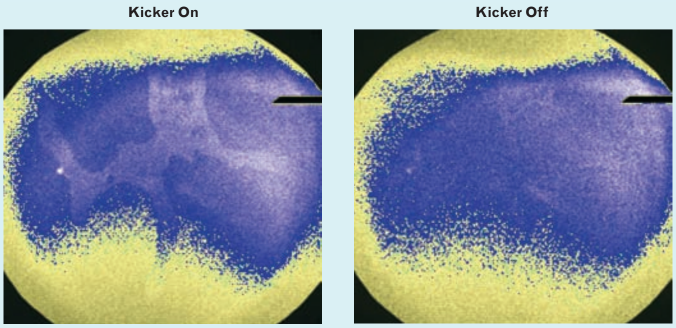


Figure 27. First results of bunch kicking mode:
- XMCD contrast at Fe L_3 edge (707eV)
- Sample: Ferromagnetic GdFe alloy film
- Off-orbit single bunch can be distinguished by offsetting the x-ray optic

Figure courtesy of P. Fischer, Berkeley Lab.

C. Improved Photon Beam Stability and Control

Beam stability (orbit, beam size, and current) is one of the most important performance parameters that define a synchrotron light source. There are increasing demands from the user community to improve the stability in amplitude and in frequency. These demands will increase as experiments become more sensitive and as the brightness increases and beam size become smaller. Stability improvements in orbit, beam size, and current need to be achieved to keep the ALS at the forefront of synchrotron radiation sources. Targeted upgrades in controls and diagnostics will provide state-of-the-art beam stability:

- Replace 15-year-old beam monitoring components such as our electron beam-position monitors. Add new high-bandwidth photon beam monitors and invar monitor stabilization. Increase the bandwidth of the fast orbit feedback system by installing new higher bandwidth chambers. Reduce the vibration of magnets with vibration damping pads and online vibration monitors. Add a bunch purity monitor and high-resolution synchrotron radiation monitors for smaller beam sizes. Upgrade injector diagnostics (mostly the BPM) and the fast longitudinal profile monitor.

D. Enhanced Reliability of the Accelerator Complex

To ensure that the accelerator complex provides reliable operation for the indefinite future, we will need to upgrade or replace a number of critical systems. The storage ring klystron has been in operation for more than 15 years and the model is no longer manufactured. The ALS control system relies on obsolete technology and is now presenting problems with maintenance and parts replacement. Several of the main power supplies, such as the main dipole and quadruple supplies, are failing more often. Upgrading these systems will not only help ensure high reliability but will also result in substantial performance improvements.

- To maintain the ALS RF system for the next 15 to 20 years, the ALS has chosen to replace the klystron with an inductive output tube (IOT) system. This is a commercially available power source. In addition to the IOTs, we plan to reconfigure the system to have the two RF cavities powered by separate RF sources and to upgrade the low-level RF system. This upgrade will ensure a RF system that is maintainable for the indefinite future. (The ALS has already received two thirds of the funds for this upgrade from BES.)

- To maintain and improve the ALS control system for the next 15 to 20 years, the ALS plans an evolutionary upgrade to replace obsolete components with modern control hardware. High-level software will be upgraded to accommodate the new hardware and improve performance. The control room also will be upgraded to provide improved functionality and ergonomics.

- The ALS storage ring employs several hundred magnets to guide, focus, and control the electron beam. A number of these supplies have been failing, such as the main bend and quadrupole supplies, which are also the biggest contributors to noise. In order to operate reliably for the next 15 to 20 years, these will need to be replaced.

E. New Insertion Device R&D

All except one of the insertion devices considered for the ALS renewal are soft x-ray elliptically polarizing ("APPLE-II") insertion devices. The other device would be a short period undulator for harder (12 keV) photons. Such instruments are very popular because of the great versatility in the polarization and energy range they can provide. However, they also present a number of challenges. They require, more maintenance than other insertion devices owing to their frequent mechanical motion; they perturb the orbit and beamsize stability and require very complex feedback systems; and they impact the nonlinear dynamics that can shorten the lifetime or reduce the injection efficiency. All of this impacts facility performance. However, along with other synchrotron facilities, we should explore possible avenues for improvement such as:

- Non-mechanically-moving EPUs and/or improved field shapes.

- Superconducting undulators for achieving ultimate brightness in the soft x-ray range and for significantly higher brightness than now is achievable with wiggler sources for operation at 12 keV. (Historically, upgrades in the higher energy range have been funded largely from non-BES funds.)

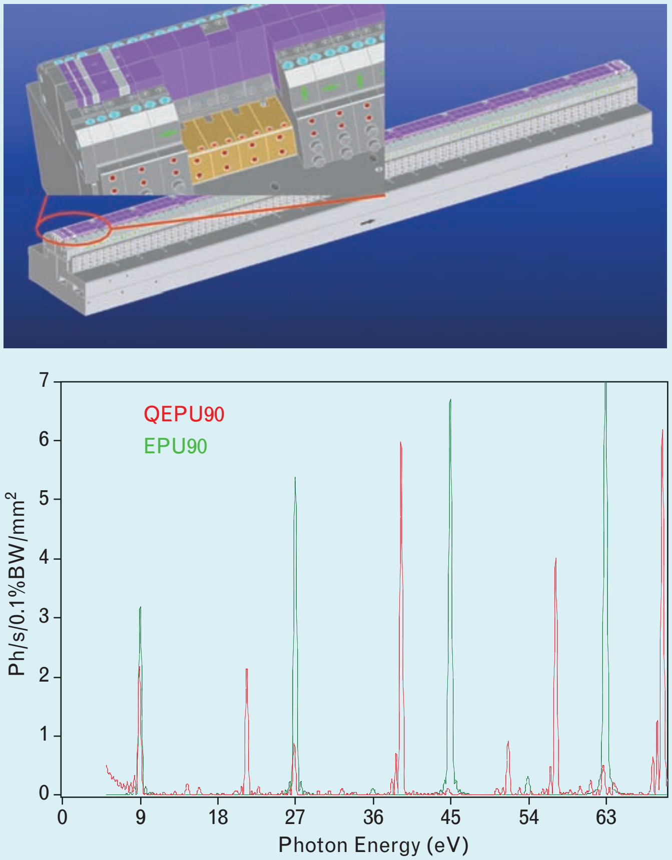


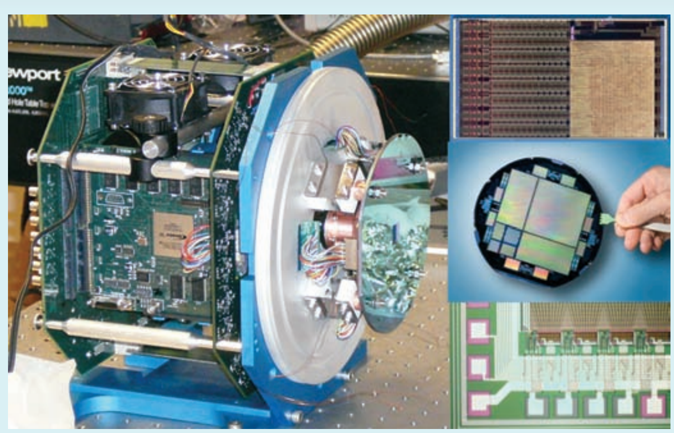
Figure 28. Quasi-periodic undulator. Figure courtesy of S. Prestemon, ALS.

IV. ENABLING TECHNICAL CAPABILITIES

A. Detector Development Program

Significant investment has been made over the last 20 years in the construction of ever-brighter synchrotron radiation sources but without a sufficient concurrent investment in detector technology. The result is that sources far outstrip detectors in terms of capability. The development of novel detectors will provide a notably enhanced experimental capability that cannot be over emphasized. At Berkeley Lab, an effort is now under way to focus our demonstrated expertise in semiconductor detectors for nuclear and high-energy physics on the detector needs at the ALS. This effort has been ongoing for several years at a low funding level and is presently focused on a small detector program based on Laboratory-Directed Research and Development (LDRD) funding, aimed specifically at very fast readout CCDs. This is now transitioning into an ALS-supported activity, with the goal of deploying variants of these detectors for soft x-ray speckle, soft x-ray coherent diffractive imaging, and several other areas. This is an efficient and low-cost way to vastly improve the performance of our experimental systems. Next-generation detectors will be needed to match the capabilities of the ALS and the increased demands of our user's experiments. To bridge the gulf between today's needs and the detection requirements addressed in the Grand Challenges, we will develop:

Figure 29. A 200-frame/second direct x-ray detection camera with custom CCD and custom integrated circuit readout developed at LBNL. Figure courtesy of P. Denes, ALS.



- A detector program that leverages the broad range of capabilities at Berkeley Lab and focuses them on solutions to ALS problems. Specific focus areas are: improving the fast readout CCDs for ultimate soft x-ray performance via delta-doped entrance windows (this will also benefit PEEM3 and soft x-ray scattering); developing sensitive, rapidly gated 1D and 2D detectors for an ultra-fast slicing source, thus enabling dispersive techniques that have orders of magnitude improved efficiency; developing ultrahigh-speed 1D and 2D detectors for photoemission experiments involving microchannel plate first stages.

B. Optics Development Program

As in the detector area, optics has been a neglected area of synchrotron radiation research, particularly at VUV–SXR beamlines, i.e., those using grating optics. Typically, one to two orders of magnitude in flux or brightness is wasted by nonoptimal optics. We will embark on an upgrade path that should yield enormous increases in performance for relatively modest costs. Indeed, this has already been applicably demonstrated in the area of protein crystallography, where on several beamlines gains of over an order of magnitude in useful flux were recently achieved. Beyond this, we are continuing to develop concepts for novel optics that may take some experimental techniques into new areas. In order to accomplish this, we must:

- Develop cutting-edge technology in soft x-ray optics, such as the use of very high-order multilayer blazed gratings based on anisotropic etched silicon substrates, which are capable of meV resolution at energies up to a keV and which have the potential to revolutionize ultrahigh-resolution soft x-ray spectroscopy. This development will also benefit the next generation of facilities based on free-electron generation.

C. Metrology Program

Continual investments are necessary to upgrade our metrology program to be commensurate with the increased brightness of the ALS and satisfy the increased demands of the experiments. To retain the high brightness of the source, beamline optical surfaces have to be manufactured to extremely high tolerances for the angular deviation from the perfect surface ($\sim$100 nanoradians) and for height deviations typically of nanometer precision. In particular, we need to:

- Develop the next generation of metrology tools and techniques, so that we will have the capability of measuring figure error down to the 100-nrad levels by making a transition to at-wavelength measurements using x rays.

- Make substantial investments in manufacturing R&D with a vendor outside of normal beamline projects and investigate the latest advances in manufacturing.

D. Theory Institute for Photon Sciences (TIPS)

The experimental requirements for addressing the grand challenges will require the development of not only sharper experimental tools but also new theoretical methods incor-

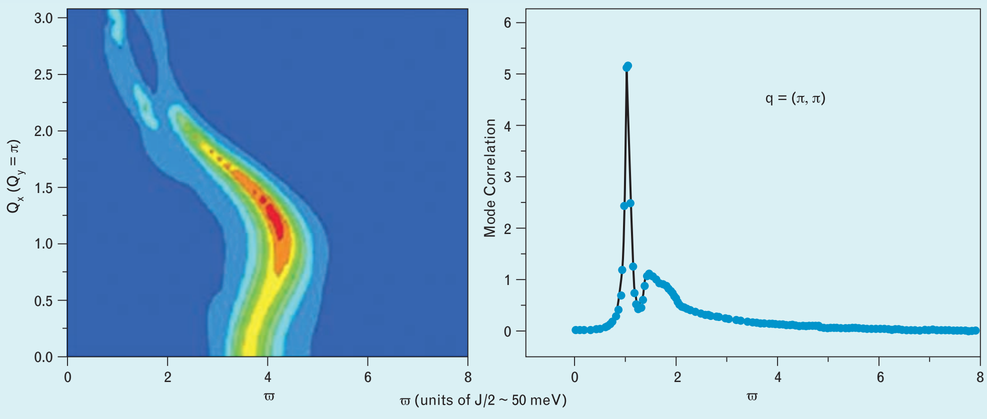


Figure 30. Existence of charge collective modes in cuprate superconductors. Figure courtesy of P.A. Lee, MIT.

porating concepts beyond those we currently possess In order to understand the resulting data (see the BES workshop report "Opportunities for Discovery: Theory and Computation in Basic Energy Sciences." We propose:

- Development of a Theory Institute for Photon Sciences (TIPS) with the primary mission of initiating a strategy of strong interaction between experimentalists and theoreticians. The goal of this institute is to significantly enhance not only the productivity of the users but also the process of scientific discovery, and to thus foster an environment that is far better suited to tackling the grand challenges. This will be a joint effort between Berkeley Lab and SLAC.

E. Facilities

Berkeley Lab Guest House

Groundbreaking for the 57-room Berkeley Lab Guest House occurred during a ceremony at the ALS Users' Meeting in October 2007. The facility will be completed in the fall of 2009 and will be a great benefit to our user community, offering well-designed on-site accommodations just a short walk from the ALS.

User Support Building

At the ALS, there is a critical shortage of high-quality user support space. In addition, current space is not large enough to accommodate the growth in our scientific programs. This shortage of suitable space for users creates significant impediments to the attainment of DOE objectives. However, a new ALS User Support Building (USB) has now received funding, and the project is underway. Our expectation is that the USB will be ready for occupancy in the summer of 2011. The USB will consist of approximately 30,000 gross square feet for precision component assembly of experimental equipment, beamline equipment staging with a high-bay, a potential future beamline extension from the ALS, chemical and biological preparation laboratories, chemical storage, and office space for about 85 persons, including 70 users.

V. USER SCIENTIFIC SUPPORT AND FUTURE SCIENTIST PIPELINE

A. Scientific Support

The ALS is recognized as the user facility that provides a very high level of scientific and technical support to all users. This has been highly appreciated by our users community as well as the BES review committee, which has clearly indicated the need and importance of the scientific staff that has helped to make the ALS an outstanding users facility. However, level of support is not sustainable over the long term, even for our existing beamlines, without increased staffing. New beamlines will only increase the need. Thus, it is essential to continue this effort by hiring necessary user support staff to improve productivity, keep the morale of the scientific staff high and assure that current practices can be maintained at our customary high level.

B. ALS Doctoral and Postdoctoral Fellowships

The ALS, through its doctoral and postdoctoral fellowship programs, leads the way in establishing a pipeline of future beamline scientists to BES user facilities. Through these programs, we welcome graduates and postgraduates from universities, industries, and government laboratories from around the world. The doctoral fellows are full-time Ph.D. students currently pursuing thesis research based on the use of synchrotron radiation. They are matched with an on-site mentor, have access to ALS resources, and are compensated with an annual stipend. (For 2007–2008, there are nine doctoral fellows.) The newly launched ALS Postdoctoral Fellowship program identifies outstanding individuals in new and emerging scientific and engineering research fields and provides them with advanced training. In addition, the fellows are paid a monthly salary. As integral members of ALS research teams, they exchange innovative scientific ideas and techniques and enhance their professional development. (For 2007–2008, there are between six and ten postdoctoral fellows working onsite.)