

Stanford Synchrotron Radiation Lightsource

Strategic Plan:
2019-2023



Meeting the Scientific Challenges of the Future



November 2019

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Introduction

An abstract 3D rendering featuring a dark background filled with numerous glowing spheres in shades of teal, orange, and white. A prominent, bright orange beam of light streaks diagonally across the scene from the upper right towards the lower left. At the point where the beam meets a cluster of spheres in the lower-left quadrant, there is a bright, multi-pointed starburst effect. The spheres vary in size and are scattered throughout the space, creating a sense of depth and dynamic energy.

1 Executive Summary

The next decade will see a transformation in the capabilities of storage ring-based synchrotron radiation facilities. The construction of new synchrotrons, based on multi-bend achromat designs, will lead to significant increases in transverse X-ray coherence. The investment in high brightness facilities will enable advances in X-ray imaging, but the emphasis on X-ray coherence will not advance all X-ray techniques that are critical to discovery and use-inspired research or in demand by the user community. This plan set forth SSRL's role in this evolving landscape.

Many of the most critical scientific questions require moving beyond structure-function correlations to actually characterizing the structural evolution that dictates function. Progress in these directions places great emphasis on making the function of interest accessible to X-ray methods and tracking them in real time with the necessary temporal resolution. A focus on real time observation of function will enable Stanford Synchrotron Radiation Lightsource (SSRL) to contribute significantly to the diverse scientific challenges being tackled by our user community and prioritized by our scientific partners and sponsors.

The strategic emphasis on real-time characterization of materials, chemical, and biological function with *in-situ* and *operando* methods aligns strongly with SLAC's goal to be the premier X-ray and ultrafast science laboratory in the world. The strategy for achieving this objective is built on the bedrock of SLAC's two complementary, world-class X-ray facilities, the Stanford Synchrotron Radiation Lightsource (SSRL) and the Linac Coherent Light Source (LCLS). SSRL, as a Basic Energy Sciences (BES) Scientific User Facility, utilizes and amplifies SLAC's core capabilities and leverages the proximity to the exceptional intellectual environment at Stanford University to address critical scientific challenges in support of the national interest and the Department of Energy (DOE) missions in energy, environmental, biological, chemical, and physical sciences. Emphasis on 1-to-1,000 picosecond-resolution X-ray capabilities will critically advance the DOE mission and enable science distinct from the other BES supported Scientific User Facility light sources.

Throughout its history, SSRL has demonstrated a commitment to scientific discovery, innovation, exceptional user support, and renewal. Continuing SSRL's history of scientific excellence requires a sharp focus on the unique opportunities enabled by

the strengths of SSRL and the SLAC-Stanford research environment. The scientific foci of SSRL's Strategic Plan presented herein emphasize the following opportunities:

- Expand SSRL's unique connection to Stanford University. This connection will continue to provide very important research and educational components designed to develop future leaders in all fields impacted by storage ring-based synchrotron radiation, in addition to creating an atmosphere that enhances the user community's experience.
- Implement new SSRL beam lines that accentuate our strategic advantages and address grand challenge problems in energy, biosciences, the environment, and chemical and physical sciences and effectively couple to external partners from industry, national laboratories, and academic institutions.
- Emphasize the synergistic coupling between SSRL and LCLS in scientific research and as DOE BES national user facilities. This coupling will enhance research capabilities and the ability to offer users of SSRL and LCLS effective scientific complementarity, commonality in user services, and user support.
- Focus SSRL's accelerator development strategy on short-pulse capabilities with time-resolution between 1-100 ps that accentuate the synergy with LCLS and extend the current scientific strengths of SSRL.
- Maintain the outstanding support experienced by SSRL users, which is an overriding goal of SSRL. User satisfaction is consistently reflected in the "excellent" scores received in the end-of-run summaries and external reviews, and continues to be the hallmark and signature of the

SSRL experience. It is SSRL's goal to meet and go beyond the expectations of our user community in providing new capabilities and the highest level of integrated scientific, technical, safety, and administrative user support.

SSRL's Strategic Plan will enable scientific advancement in four major focal areas: accelerating functional materials discovery and design through incisive characterization, understanding catalytic function with atomic-scale precision, identifying how constituent interactions generate emergent behavior in quantum materials, and characterizing complex biomachinery and heterogeneous hierarchical natural systems. Each of these research areas has clearly defined strategic initiatives that ensure scientific impact and expansion of the SSRL research community:

- Enhance our scattering capabilities with a focus on energy transformations and storage. Expand our *in-situ* and *operando* scattering and imaging capabilities for characterizing energy transformation and storage materials and improving materials synthesis. Develop and refine high-throughput approaches and apply these to technologically important materials classes.
- Establish X-ray emission and high-resolution fluorescence detection mode X-ray absorption spectroscopies in the hard, tender, and soft X-ray regimes as standard spectroscopic methods for chemical, biological and environmental sciences at beam lines 15-2, 6-2, and 10-1. Develop time-resolved X-ray capabilities and multimodal methods to track chemical transformations in real time in the range of picosecond and longer.
- Integrate Molecular Beam Epitaxy (MBE), Pulsed Laser Deposition (PLD), and Scanning Transmission X-ray Microscopy (STXM) with state-of-the-art angle- and spin-resolved photoemission spectroscopy. Utilize resonant soft X-ray scattering to characterize the emergence of charge and spin order in correlated electron systems.
- Develop and expand micro-focus macromolecular crystallography in collaboration with LCLS. The new microfocus undulator beam line 12-1 at SSRL and the Macromolecular Femtosecond Crystallography (MFX) station at LCLS represent the complementary cornerstones of this effort, providing a user gateway that will maximize effective use of LCLS while providing next generation storage-ring based capabilities in its own right. Develop time-resolved SAXS methods to investigate the conformation and assembly of macromolecular complexes (microsecond time domain and longer). Develop time-resolved X-ray spectroscopy tools to study active-site intermediates in biological reaction cycles using both SSRL and LCLS (as short as femtoseconds).

The scientific and technical focus of the SSRL strategic plan will also enable the expansion of our user community:

- The expansion of high-throughput characterization and *in-situ* and *operando* studies of materials synthesis, growth, and assembly links directly to applied and translational research. This growth in technical capabilities is being achieved with support from the Office of Energy

Efficiency and Renewable Energy and provides a clear channel to increased support of industrial users.

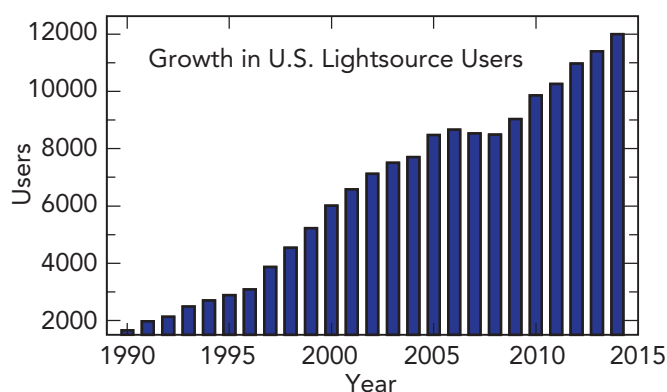
- The growth in *operando* and multimodal methods for catalyst characterization provides a foundation to increase our user base in academic, national laboratory, and industrial users.
- The development of 1-100 picosecond resolution capabilities and a macromolecular crystallography gateway between SSRL and LCLS will attract new users to SSRL from LCLS and other free electron laser facilities around the world.

The sections below provide background, overview, and a more detailed current and future perspective of SSRL's scientific programs in the main areas of materials energy science, chemistry, catalysis, interfacial and environmental science, structural molecular biology science, accelerator science, and instrumentation development.

2 The Role of Synchrotron Radiation Facilities in U.S. R&D

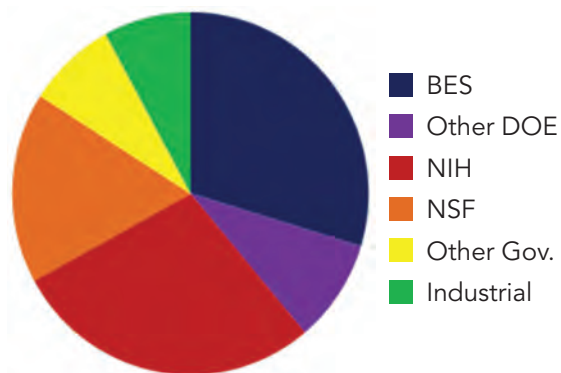
Synchrotron X-ray facilities provide a powerful toolbox for interrogating the properties of matter with atomic resolution and elemental specificity. This toolbox, like other innovative advances in experimental capabilities, has facilitated diverse scientific discoveries and technological breakthroughs. For example, five Nobel prizes have been awarded to scientists who used synchrotron light sources for their research.

Today, there are over 50 synchrotron light sources around the world and 6 operating in the U.S., with more either planned or under construction. These light sources together support the research of tens of thousands of researchers worldwide and have impacted most fields of science, engineering, and medicine. Within the U.S., the four DOE synchrotron light sources support over ten thousand users annually, and the demand for these light sources continues to increase. Synchrotron light sources support the full scope of research and development in the U.S., with over half the synchrotron users reporting funding

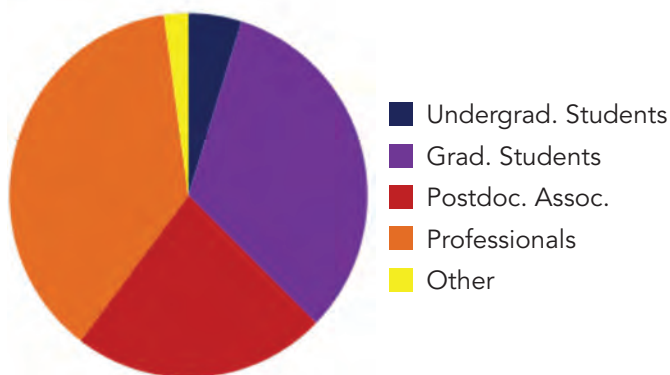


support from non-DOE federal agencies. Light sources also serve a critical role in training and education. Over half of the synchrotron users are undergraduate, graduate, or postdoctoral scholars, the next generation of scientific and technical leaders that will enable the technological advances that have driven economic development in the U.S.

Declared Funding Source of U.S. Light Source Users



Career Status of U.S. Light Source Users



3 Stanford Synchrotron Radiation Lightsource

The SPEAR3 storage ring is operated at 3 GeV, with 9.6 nm-rad emittance and top-off injection at a current of 500 mA with a reliability of over 97%. There is an active accelerator research and development program to continue improving the performance and reliability of the accelerator complex, including emittance improvements to 6 nm-rad and the development of short pulse operation in the few ps range. SSRL is increasing its number of undulator beam lines and continuously upgrading existing beam lines, including new optics and instrumentation to meet the needs of the user community.

SSRL, being the premier hard X-ray source serving the Western U.S., currently supports the research of more than

1700 users annually and operates 32 scattering, diffraction, spectroscopy, and imaging experimental stations, with expansion capacity and ability to serve more than 2200 users. It is a highly productive scientific user facility with high user satisfaction, generating more than 500 peer-reviewed publications annually. Research conducted at SSRL continues to have major impacts in condensed matter physics and materials sciences, structural biology, and chemical and environmental science.

As one of the DOE Office of Science’s major scientific user facilities, SSRL focuses on the scientific opportunities and research priorities identified by the DOE Office of Science and other agencies, in particular the National Institutes of Health (NIH) and the National Science Foundation (NSF). SSRL also works with a diverse community of researchers to develop new capabilities, as well as provide research infrastructure and support to enhance scientific productivity and attract and educate new user communities.

In looking to our future, a continued commitment to innovation, discovery, service, and training will guide SSRL to future successes. SSRL has a history of tackling important scientific challenges through targeted technical advances strongly supported by key scientific partnerships. Innovation and discovery drive research and development, but innovation and discovery languish without talented, ambitious, and well-trained scientists and engineers. Workshops, summer schools, and on-line and hands-on tutorials are central to SSRL’s strategy for user community outreach and growth – a commitment that (1) helps maintain strong partnerships with our academic user community, (2) ensures that the next generation of scientists has the know-how to integrate the unique capability of synchrotron radiation into their future research planning, and (3) supports the research of roughly 100 doctoral theses annually.

3.1 Building a Future for SSRL in a Changing Storage Ring Landscape

Widespread access to high-brightness synchrotron radiation presents an enormous opportunity for the scientific community to address large-scale and complex problems, working increasingly across facilities and even national boundaries, while expanding into new fields and applications. During this time, facilities need to both focus on their own strategic directions through interactions with key stakeholders – in particular their user community – as well as coordinate and collaborate with other facilities.

SSRL will focus on developing our real-time measurement capabilities and expanding our science and user base in this critical area of science. Many of the most critical scientific questions require moving beyond structure-function correlations to actually characterize the structural evolution that dictates function in real time. Progress in these directions places equal or greater emphasis on temporal resolution than on spatial resolution. A focus on real time observation of function will enable SSRL to contribute significantly to the diverse scientific challenges being tackled by our user community and prioritized by our scientific partners and sponsors.

Time resolved X-ray methods combine photo-, electrical-, or chemical-triggers with the incisive power of X-ray scattering, spectroscopy, and imaging to track in real time physical and chemical changes. A commitment to ultrafast accelerator based science provides a natural extension of SSRL's focus on *in-situ* and *operando* methods, enhances the synergy between, SSRL, LCLS, and the SLAC Accelerator Directorate, and leverages the strong commitment to ultrafast science in the SLAC Photon Science Directorate.

SSRL's strategy for expanding our footprint in ultrafast science has three key components:

- Building robust, low overhead optical pump-probe capabilities at our highest performing undulator beam lines. Combining ultrafast science capabilities with more traditional research capabilities will enable the organic development of an ultrafast scientific community at SSRL.
- Integrating the Ultrafast Electron Scattering program under development in the Accelerator Directorate into the core program at SSRL.
- Developing a longer-term SPEAR3 accelerator enhancement that will deliver more than an order of magnitude shorter X-ray pulse duration without adversely affecting normal operations.

Scientific opportunities

SSRL focuses on the scientific opportunities and research priorities identified by the DOE Office of Science and other funding agencies, and works with researchers from academia, industry, and national laboratories to develop new experimental and analytical techniques, design and construct state-of-the-art instruments, provide research infrastructure and support to enhance scientific productivity, and attract and educate new user communities.

Partnership with scientific community

For the scientific foci discussed in this document, SSRL will build partnerships with the leaders and the community to (1) identify the most important problems in the field, (2) guide the development of new experimental techniques, beam lines, instrumentation and research facilities optimized for pursuing those problems, and (3) provide support to the community for efficient access, successful experiments, and high-impact scientific results.

Discovery to deployment

Discoveries from basic research often lead to technology developments that have significant societal impact, and the needs of industry and national security often inspire new basic research directions. National user facilities are the ideal places to promote collaborations among researchers from academia, industry, and national laboratories. SSRL will facilitate and strengthen these interactions and the exchanges between basic and applied research. SSRL will specifically focus on developing strong relationship with industrial partners.

Dedication to users' needs

SSRL continues to identify the steps a new user or user community follows to take an idea through to the successful conclusion of an experiment, and improve each step of the user experimental cycle. This includes steps from proactively reaching out to new scientific communities to building the needed tools and support for experimental design, data collection and data analysis. These improvements help ensure that SSRL focuses on the most pressing scientific questions and that users have what they need to make their time at SSRL productive.

Materials at Work: Resolving Ultrafast Energy Transduction and Transport in Space and Time

The function of advanced energy materials involves a combination of energy storage, discharge, transformation, and transport. All of these functions involve transient changes in electronic and nuclear structure on a diverse range of time and length scales. Identifying and mitigating the processes' bottlenecks that limit the rate of energy storage in batteries or fuels is a critical objective in both discovery and use-inspired research.

With the inclusion of Ultrafast Electron Scattering and the development of high-flux, sub-5 ps resolution X-ray capabilities, SSRL will have a powerful set of tools for tracking the structural dynamics of atoms and molecules on their natural time and length scales. These research capabilities have particular relevance in energy conversion phenomena, where the non-equilibrium dynamics associated with light absorption, charge injection, as well as charge, spin, and thermal transport occur on time scales ranging from femtoseconds to milliseconds. X-ray laser sources, such as LCLS, have transformed ultrafast X-ray science, but complementary X-ray and electron tools are required to address the basic science challenges that underlie efficient and cost-effective energy conversion.

The efficiency of light conversion to fuels or electricity depends critically on metastable electronic excited states, making ultrafast methods exceptionally important to understanding, and ideally controlling, light conversion. The combination of ultrafast temporal resolution and atomic spatial resolution enabled by ultrafast X-ray and electron methods provides ideal methods for the detailed characterization of the catalyst-substrate electronic and nuclear structure during photocatalytic reactions and the site-specific characterization of carrier trapping in heterogeneous photovoltaic materials.

Light source initiatives

Short-term developments focus on the improvement of the SSRL beam brightness by nearly a factor of two. SSRL's longer term development strategy focuses on short-pulse duration capabilities that preserve the photon flux required of modern X-ray methods, accentuate the synergy with LCLS, and further develops the current scientific strengths of SSRL. A commitment to light source reliability and stability will remain a common goal to both short- and long-term planning.

3.2 Scientific Foci

The five main research areas at SSRL (**Use-inspired Materials Science, Quantum Materials, Chemistry and Catalysis, Structural Molecular Biology, and Environmental Science**) address three themes: Accelerating Materials Design, Understanding Catalytic Function and Interfacial Reactions with Atomic Precision, and Identifying How Collective Function Emerges from Constituent Interactions. SSRL's strategic plan pursues a range of scientific grand challenges related to these three themes and the five main research areas derived from the roadmaps of our major funding agencies, including the DOE Office of Basic Energy Sciences (BES). SSRL's scientific focus is further refined with input from the scientific community, guided by the advice of SSRL's Scientific Advisory Committee and Machine Advisory Committee, and through interaction with SSRL's Users' Organization. SSRL will continue to provide state-of-the-art capabilities directed at Use-inspired Materials Science, Quantum Materials, Chemistry and Catalysis, Structural Molecular Biology, and Environmental Science.

3.2.1 Accelerating Materials Design

Materials Function by Design is the ultimate goal of materials science. Recent progress in theory, computational power and methodologies, materials synthesis, and characterization tools has brought this goal within reach. Synchrotron light sources, with their exceptional properties, are playing an essential role in providing a wide range of powerful characterization tools that allow the materials research community to understand the relationship between structure and function as well as the relationship between synthesis conditions and structure formation, at an unprecedented level of detail. The challenges are to provide access to these tools in a timely fashion so that they can effectively guide the theory and synthesis of new materials and to develop sophisticated new tools that can address increasingly subtle scientific questions related to materials structure-function relationships and processing. This effort will be supported by the following strategic objectives:

- Building an undulator beam line optimized for interface scattering studies of energy storage and transformations in materials.
- Repurposing a wiggler side station for 30-40 keV diffraction, scattering, and pair distribution function determination.
- Expanding and enhancing *in-situ* and high-throughput methods for characterizing energy storage and transformation and for improving materials synthesis and processing methods.

- The development of picosecond resolution X-ray scattering methods for studying the structural dynamics of photo-voltaics, thermoelectrics, and phase change materials.

3.2.2 Understanding Catalytic Function and Interfacial Reactions with Atomic-Scale Precision

Energy efficient chemical transformations present a central objective of energy research. Research targets the cost-effective catalysis of CO₂ reduction, H₂O oxidation, N₂ fixation, and hydrocarbon functionalization with abundant and green materials. X-ray spectroscopy, scattering and diffraction provide critical approaches to determining the electronic and geometric structure of catalysts with atomic resolution and specificity. Incisive characterization necessitates tracking the properties of the catalyst, as well as the reactants, intermediates, and products. SSRL's strategy focuses on multimodal and *operando* measurements of catalysts with the full suite of X-ray spectroscopy, imaging, and scattering / diffraction techniques. This strategy is also critical for expanding our capabilities to determine the kinetics and mechanisms of interfacial reactions of importance in Molecular Environmental and Interface Science. SSRL also approaches this overarching theme inclusively, developing methods and supporting users working in thermal, electro-, and photo-catalysis, on solid state and molecular catalysts, as well as in bio- and biomimetic catalysis. This effort will be supported by the following strategic objectives:

- Develop X-ray emission spectroscopy (XES), high resolution fluorescence detection-mode X-ray absorption, and resonant inelastic X-ray scattering (RIXS) in the hard, tender, and soft X-ray regimes.
- Develop picosecond resolution hard X-ray XAS, XES, and RIXS for mechanistic studies of photocatalysis reaction mechanisms and charge transport in photovoltaic materials.
- Build a best-in-class wiggler hard X-ray spectroscopy beam line, with complementary diffraction capabilities, dedicated to multimodal catalyst characterization based on integration of X-ray, mass spectrometry, electrochemistry, and vibrational spectroscopy methods.

3.2.3 Quantum Materials: Identifying How Collective Function Emerges from Constituent Interactions

In many complex structures unique system-level behaviors and properties emerge from the interactions among the constituents of the system. The challenge is to understand the individual interactions and how these lead to the emergent system-level properties. Strong electron correlation has long been appreciated as a driving force behind high-T_c superconductivity; studies of topological insulators have more recently highlighted the importance of strong spin-orbit interactions in electron transport. Materials that possess both strong electron correlation and spin-orbit interaction present a key area for investigation for the discovery of novel properties and applications. Understanding how these strong interactions lead to emergent behavior requires integration of sample synthesis

with multiple X-ray characterization methods. SSRL will build on our current strength in angle resolved photoemission spectroscopy (ARPES) with the following strategic objectives:

- Integrate Molecular Beam Epitaxy (MBE) and Pulsed Laser Deposition (PLD) with state-of-the-art ARPES.
- Develop a high sensitivity spin-resolving area detector for spin-resolved ARPES.
- Utilize resonant soft X-ray scattering to characterize the emergence of charge, spin, and orbital ordering in correlated electron systems. This will involve the integration of a state-of-the-art superconducting Transition Edge Sensor detector to energy resolve the elastically scattered X-rays from the dominant inelastic scattering from charge transfer and d-d excitations.

3.2.4 Structural Molecular Biology: Identifying How Collective Function Emerges from Constituent Interactions

Understanding how biological structure influences biological function and malfunction is an essential component to solving grand challenge problems crucial to the health of the environment and humanity. SSRL's Structural Molecular Biology (SMB) program focuses on obtaining and utilizing biomolecular structural information on the nano-to-atomic scale to understand biological processes. The SMB program develops new and enhanced approaches to investigate biomolecular structure and function, making them widely available to the biomedical and bioenergy research communities and contributing to both basic scientific knowledge and translational research. Key developments in this research area will be supported by the following strategic objectives:

- Develop and expand micro-focus macromolecular crystallography in collaboration with LCLS. The new beam line 12-1 at SSRL and the Macromolecular Femtosecond Crystallography (MFX) station at LCLS represent the complementary cornerstones of this activity.
- Develop time-resolved SAXS methods to investigate the conformation and assembly of multi-component macromolecular complexes.
- Develop time-resolved X-ray spectroscopy tools to study active-site intermediates in biological reaction cycles.

3.2.5 Geochemistry and Biogeochemistry: Identifying How Collective Function Emerges from Constituent Interactions

Geochemical and biogeochemical systems are highly complex hierarchical structures where collective function depends critically on their physical structure, chemical constituents, and hydrological behavior. Examples include shales, soil aggregates, and floodplains. Deciphering chemical processes, kinetics, and their dependence upon 3D microstructural chemical / physical heterogeneities (nanometers to millimeters) is central to solving our Nation's most challenging energy security, water availability, and environmental challenges. SSRL is supporting the advancement of geochemistry and biogeochemistry research program priorities by

developing and supporting X-ray spectroscopy and X-ray imaging techniques. Strategically important areas to develop are:

- Develop partial fluorescence yield K-edge XAS measurement capability for routine measurement of C and N in soils, sediments, and environmental samples.
- Develop time-resolved XAS methods to investigate geochemical reaction mechanisms.
- Develop methodology for studying rocks under high-pressure, high-temperature, and flow-through conditions with tomography and XAS.

SSRL History

SSRL began in 1972 and helped establish synchrotron radiation as an essential research tool in the natural sciences, engineering, and biomedical research. The goal of SSRL has always been “to enable scientific research for a broad general user community and produce outstanding scientific accomplishments.” The facility is well known for pioneering contributions in new synchrotron methods and instrumentation and the new science these enable. Examples of important developments where SSRL has had major contributions include:

- Development of X-ray Absorption Spectroscopy Techniques: EXAFS, SEXAFS, NEXAFS
- Pioneering of MAD (multiple wavelength anomalous dispersion) phasing and synchrotron-enabled macromolecular crystallography
- Opening the Soft X-ray Region (200 - 3000 eV) (Grasshopper, Jumbo monochromators)
- Pioneering Synchrotron-Based Photoemission Techniques (core level photoemission, photoelectron diffraction, and ARPES)
- Development of Wigglers and Undulators
- Pioneering X-ray based Molecular Environmental & Interface Science (MEIS)

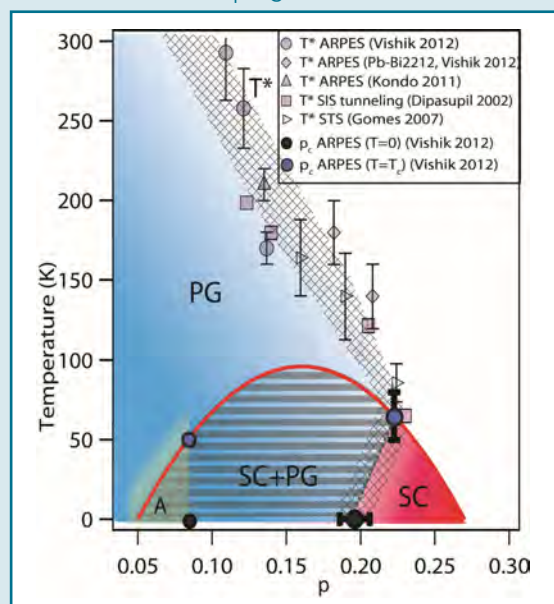
In the early 1990s, for example, SSRL identified understanding the function of RNA Polymerase II as a high-risk, high-reward scientific opportunity. Through strong collaboration with Roger Kornberg, Professor of Structural Biology at Stanford University, this scientific challenge led to many important technical advances in macromolecular crystallography, including the development of sample mounting robotics and high-throughput automated sample screening that greatly accelerated the rate with which high resolution structures could be determined. Kornberg was awarded the 2006 Nobel Prize in Chemistry for his research, for which the structural biology part was based in significant measure on data measured at SSRL.

In 2003, SSRL in collaboration with 9 outside institutions, began the Sub-Picosecond Pulse Source (SPPS) experiment to develop methodology for X-ray studies of ultrafast phenomena. SPPS produced pulses of <100 femtosecond duration of spontaneous X-ray radiation utilizing a compressed electron beam from the SLAC linear accelerator that was passed through an undulator and delivered into an X-ray hutch. SPPS both provided a powerful tool for research and served as a way to conduct important accelerator and X-ray optics R&D for the Linac Coherent Light Source (LCLS) – the world’s first X-ray free electron laser facility, that began operations at SLAC in 2009.

SSRL furthermore provided the platform, foundation and home for LCLS, from the early beginning to the creation of the LCLS construction project. This included the first demonstration of SASE at longer wavelengths, the gun development, and leading the workshops that developed the scientific case for the creation of LCLS, now SSRL’s sister X-ray facility at SLAC.

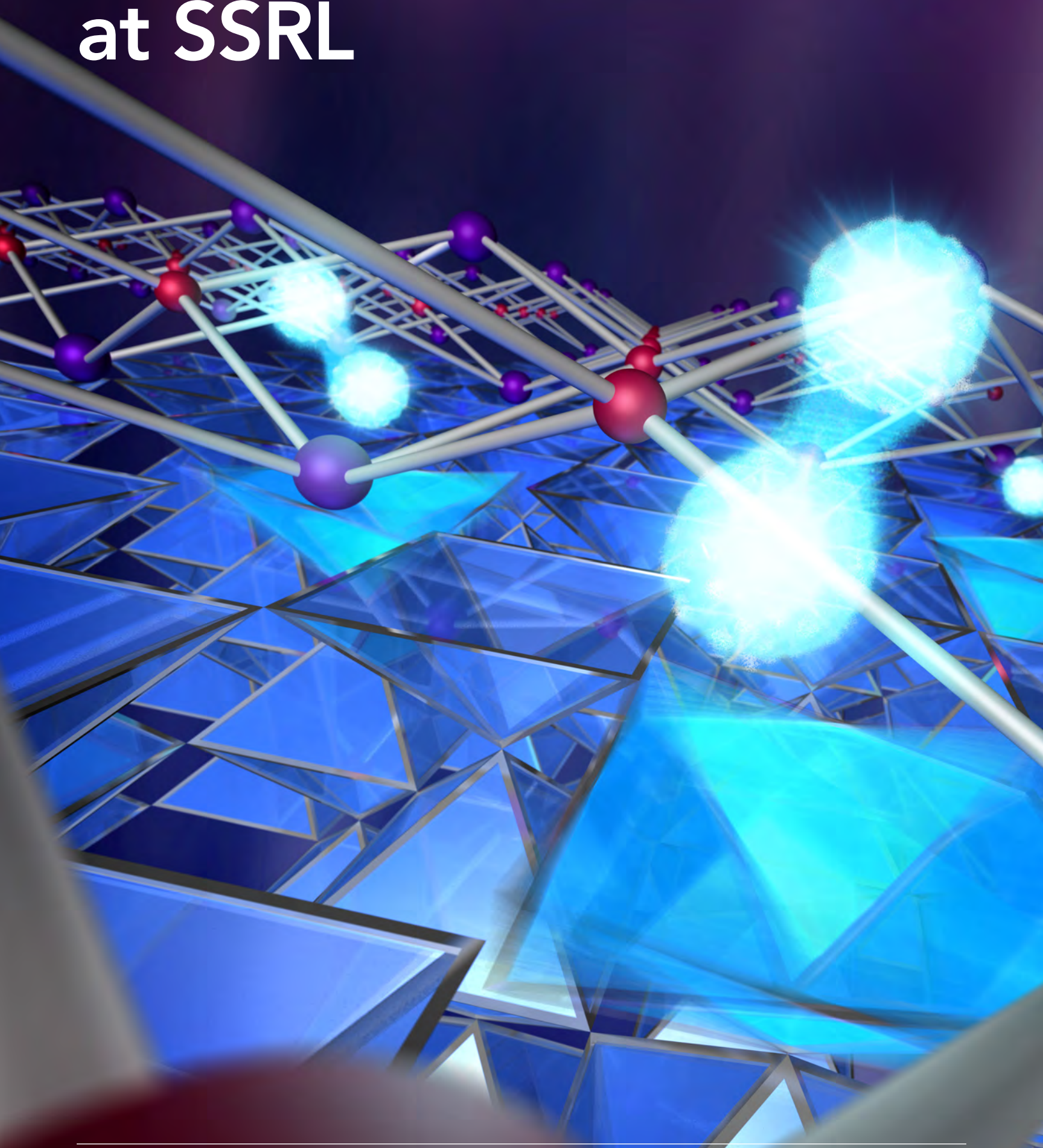
Core Scientific Program Development: Long-Term Sustained Local Collaborations

Addressing grand challenges, the extreme complexity of which rests on answers to critical scientific questions, requires systematic investigations over a long period of time, with clear scientific goals and advanced tools. SSRL is a unique facility that can synergistically accelerate the critical missions by locally collaborating with Stanford University and other units at SLAC. As illustrated below, a successful example is a Materials Science Program, ARPES on high- T_c cuprate superconductivity, driven by Prof. Z.-X. Shen. Having such a core scientific program, the ARPES beam line at SSRL has been highly productive for many years with its continuous technical improvements, keeping it as one of the leading synchrotron ARPES facility in the world. The core program has motivated the advancement of synchrotron ARPES, including the development of a new ARPES endstation, attracted more general users, and created numbers of successful quantum materials research projects. It will be highly important for SSRL to extend and expand this successful approach: development of core scientific programs leading the field, driven by long-term and sustained local collaborations, with deliberate strategic planning by SSRL to assess these high-risk, high-reward approaches in setting resources aside for such program developments (see separate sidebar above on the collaboration with Prof. Roger Kornberg).



High-transition temperature (T_c) cuprate superconductors, a prototypical strongly correlated electron system with T_c much higher than liquid nitrogen, have attracted great attention over the last three decades, due to its profound physics and broader possibilities for applications. Systematic investigation of the cuprate phase diagram has been an essential approach towards the understating of the high- T_c superconductivity mechanism, particularly because the mysterious “pseudogap” phase (“PG” in the Figure) with unknown nature has been a stumbling block. At the high-resolution ARPES beam line 5-4, an ideal spectroscopy setup to study fine electronic structures, a decade long-term systematic investigation of the phase diagram, with close collaboration between SIMES and SSRL, significantly advanced the understanding of the nature of the pseudogap phase: it essentially is a competing order that coexists with superconductivity under a portion of the superconducting dome (“SC+PG”), making the phase diagram even more complex and intriguing. These results have provided microscopic insights into the cuprate phase diagram and a possible pathway towards realizing higher T_c superconductors.

Science at SSRL



4 Science at SSRL

4.1 Materials Science

Advanced materials are at the heart of our technically advanced society. Materials science research at SSRL focuses on two broad themes: (1) accelerating materials discovery with advanced X-ray characterization methods and (2) identifying how collective function emerges from constituent interactions in strongly correlated electron systems.

The success of the Materials Genome Initiative has highlighted a key challenge that must be addressed to further accelerate the pace of fruitful materials discovery. A gap exists between theoretical and experimental approaches to materials discovery that suppresses the rate of materials discovery. Closing this gap requires a deeper understanding of how synthesis and processing conditions control the structure of new materials. We will focus on materials targeting sustainable energy independence and on advanced manufacturing that promises to initiate a digital renaissance in American manufacturing.

Strongly correlated quantum materials provide a compelling intersection between discovery-directed fundamental science and the potential to transform the way we transport energy and information. The transformative potential of strongly correlated electron materials originates from their emergent properties. Harnessing the emergent behavior of high T_c superconductors, topological insulators, and multiferroics requires a deeper understanding of how the remarkable properties of matter emerge from the complex correlations of the atomic and electronic constituents and, ideally, the ability to control these properties. Thin film and hetero-structured correlated materials provide one of the most promising approaches to understanding and controlling the emergent behavior of quantum materials. SSRL will focus on the application of angle resolved photoemission spectroscopy, resonant soft X-ray scattering, and interfacial scattering to characterize the properties of thin film and hetero-structured quantum materials.

SSRL's Materials Science programs will provide, and further develop, a comprehensive set of tools and methodologies to shrink the gap between computational and experimental materials discovery with a focus on the function of new materials. Special emphasis will be given to the following areas:

- Enhancing *in-situ*, real-time capabilities for the study of structure-function relationships of materials, materials responses under realistic operational conditions, and materials synthesis and processing.
- Developing tools and integrated approaches to probe materials over multiple length-scales and time-scales.
- Coupling experimental techniques closely to theory and modeling.

The Materials Science program at SSRL has a long and successful history of synergetic interactions with other programs at SLAC and Stanford. The collaboration with the Stanford Institute for Materials and Energy Sciences (SIMES) has been instrumental to the highly successful ARPES program on

quantum materials, which in turn strongly influenced the development of beam line 5, including the recent development of *in-situ* synthesis and analysis of novel quantum materials. The SSRL and SIMES collaboration on battery research has been an important stimulus for the development of *operando* X-ray tomography capabilities. Similarly, the oxide interface research effort serves as a driver for the interface scattering beam line development. This history of close collaboration between local scientific teams has been a hallmark of SSRL's approach to science and central to the development of technical capabilities beneficial to the general user community.

SSRL will broaden its impact through strategic partnerships with a number of institutions in sustainable energy materials research, including the Energy Innovation Hubs like the Joint Center for Energy Storage Research, Energy Frontier Research Centers, like the Center for Next Generation Materials by Design, the National Renewable Energy Laboratory, and Stanford's Institutes (like the Precourt Institute for Energy) and Schools. Often SSRL hosts EFRC and researchers. SSRL will closely coordinate with other DOE Office of Science user facilities to enhance industrial research, and support initiatives supported by DOE technology offices, for example, the Bay Area Photovoltaic Consortium (BAPVC), the EERE DuraMat and HydroGEN consortia in the Energy Materials Network (EMN), and the "*In-Situ* Data Acquisition and Tool Development for Additive Manufacturing Metal Powder Systems", supported by EERE. We will align our beamline, instrumentation and staff development strategies with new mission science priorities in Quantum Information Science, Microelectronics and Advanced Manufacturing. SSRL will also interact with the NSF Division of Materials Research.

Finally, in close collaboration with Stanford University and through introductory workshops for new synchrotron users, and one-on-one mentoring for undergraduate and graduate students, we will carry on SSRL's long tradition of educating and training the next generation of materials scientists and engineers.

The following sections focus on the scientific challenges in the major materials science research directions at SSRL, and new instrumentation being developed and planned.

4.1.1 Materials for Sustainable Energy

The development of sustainable energy solutions to power humanity is the most important scientific, technical and social challenges of our time. Dramatic improvements in energy conversion, transmission, storage and usage are critical to maintain the economy, environment and our quality of life. SSRL is playing a major role for detailed characterization and understanding of the structural and electronic properties of a wide variety of sustainable energy materials. This includes organic and inorganic photovoltaics, battery electrodes, electrolytes and systems, additive manufacturing processes, and catalytic particles.

This work will continue to be an important and growing effort at SSRL involving X-ray scattering, spectroscopy and imaging studies of materials' structure-function relationships; this necessitates time-resolved studies conducted *in-situ*, *operando*, and during synthesis and growth. Furthermore, these materials

are often hierarchically complex at the mesoscale, and it is essential to characterize them over length scales ranging from Å to μm . An example of this, for batteries, is shown below. The ability to rapidly collect an X-ray tomography image at a spatial resolution of μm to determine the interior structure of both batteries and metal components is an essential complement to the existing instrumentation at SSRL. This motivates a new bending magnet-based beam line for X-ray tomography.

Studies of interfaces will become increasingly important, since interfacial regions are crucially important to the function of the majority of materials used in sustainable energy applications. The atomic and molecular structural properties of interfaces affect the function of devices used for energy conversion, storage, and use. SSRL has – and will continue – to focus on *in-situ* and *operando* studies of materials growth and processing, and expand our abilities to characterize buried interfaces in real time. Although interfaces between two condensed phases prove challenging to characterize with atomic precision, X-ray scattering offers an unparalleled method of tackling this critical area of research. SSRL is developing a new in-vacuum hard X-ray undulator beam line for X-ray scattering (BL17-2), which will have excellent capabilities for interface and surface scattering, including bringing the experiments into the timing domain.

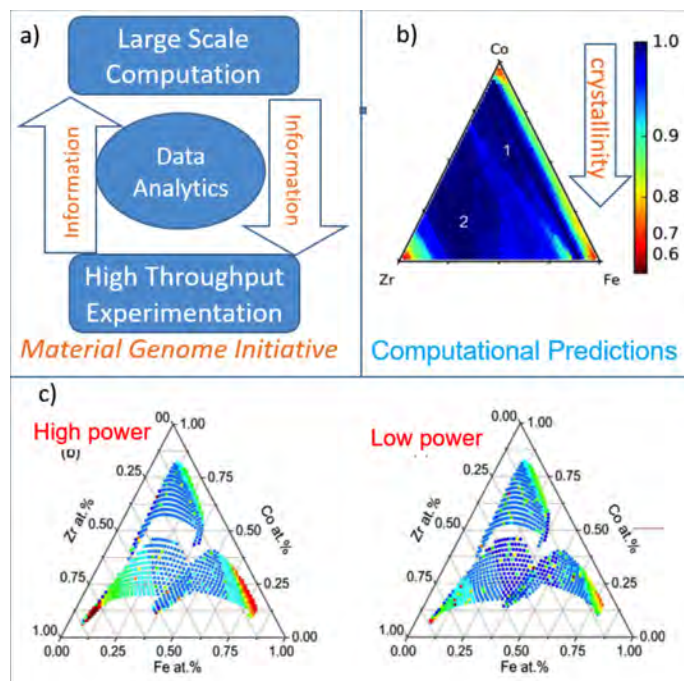
Cost-effective solutions to the materials challenges facing us today necessitate accelerating the discovery of new functional materials. The development of high throughput characterization methods will play a key role in achieving this objective. The present Edisonian paradigm based on serial experimentation takes decades from initiation of a new search for a material to marketplace deployment of a device. SSRL is pioneering high-throughput experiments with data analytics with the objective of developing the characterization methods needed to complement the large scale computational materials discovery tools already central to the Material Genome Initiative. The nearly real time extraction and fast dissemination of high level

information from each experiment will assist in cutting the time and cost of discovery and deployment of new functional materials by half.

SSRL has built a high-throughput X-ray diffraction facility and is developing on-the-fly data analytics that incorporates recent advances emerging from unsupervised pattern recognition, machine learning and artificial intelligence to produce between 2-5 ternary and quaternary phase diagrams a day. These phase diagrams, by concisely capturing the complex relationship between composition, process conditions, structures and properties, enable close collaboration among (computational and experimental) materials scientists and process engineers, facilitating fast materials discovery.

The increasing importance of energy materials research motivates enhancements to SSRL's capabilities in structure and morphology characterization with scattering, imaging, and spectroscopy:

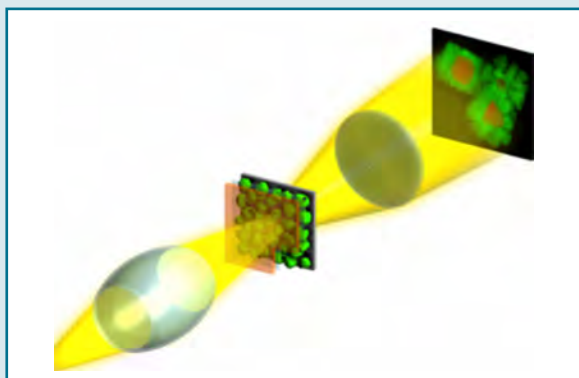
- **Interface Scattering:** The importance of interface structure in energy materials drives the requirement for an undulator source with a state-of-the-art diffractometer for accurate measurements of interface specific diffraction. This allows determination of the interface atomic and molecular structure, which affects the functioning of devices used for energy conversion, storage, and use. The use of this methodology for buried interfaces is a unique strength of hard X-ray diffraction. The new BL17 will further enhance picosecond time-resolved sciences efforts.
- **X-ray Tomography:** The wide range of important length scales in energy materials drives a need for a high-speed, high-energy X-ray tomography capability. A new bending magnet beam line is planned, that would be capable of both white light (for additive manufacturing) and monochromatic X-ray experiments. *Operando* studies of battery electrodes during cycling and of catalysts will lend insight into degradation mechanisms.



Metallic glasses are a class of functional material that are finding a wide range of applications, from biomedical implants to magnetic cores in high efficiency transformers. Over the last two decades, materials scientists have developed several empirical rules for finding these; but these rules are not universal, and it still takes extensive amount of blind experimentation and serendipity to locate a new metallic glass former. Ren et al. combined predictions from large-scale computation with high throughput experimentation to discover several new metallic glass forming systems. The computational approach was to build a machine learning tool that used several empirical factors, such as variance in atomic radii and mean atomic number with several known melt spun glass formers to predict new ternary glass forming systems. One prediction is shown in panel b). (Degree of crystallinity increases from blue to red.) Several versions of the predicted ternary composition spreads were synthesized by changing the degree of metastability via control of the sputter deposition power. High throughput X-ray diffraction measurements were used to establish the degree of crystallinity. (Result for two different processing conditions are shown in c)) (MRS Commun. 2017, 7, 613-620).

Cross-Cutting Theme: *In-situ* Scattering, Spectroscopy, and Imaging of Reactions and Materials Synthesis

The ability of hard X-rays to penetrate materials enables real time (*in-situ* and *operando*) studies of materials processing and reactions. While much has been done at SSRL and elsewhere in the past decades, with the emergence of Materials by Design pushing into synthesis space and the increasing importance of sustainable energy materials, hard X-ray scattering, spectroscopy and imaging will play a vital role in better understanding these reaction processes. This will facilitate the rational design and synthesis of novel functional materials. The structure and morphology of supported nanomaterials used as catalysts changes during the catalytic reaction and this can have an effect on the subsequent reactivity. Thus, to better understand these effects, it is important to probe these changes *in-situ* and in real time. This can be accomplished with *in-situ* imaging (using an upgraded transmission X-ray microscope (TXM)), spectroscopy (new advanced spectroscopy beam line) and *in-situ*, simultaneous SAXS/WAXS on the new undulator beam line (BL17-2). Similarly, for electrochemical energy storage, *operando* studies of how the anode and cathode morphology and physical and chemical structure change during discharge and charge are important for understanding capacity fading, an outstanding issue for electrical vehicle batteries.



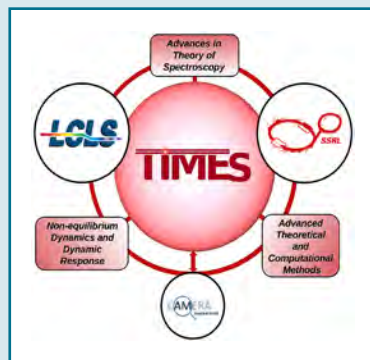
Lithium-ion battery technology has improved slowly. Research has been dominated by electrochemical measurements, which give only a general picture of a battery's health and ex-situ measurements of individual components after a cell is taken apart. By combining *in-situ* high resolution transmission X-ray microscopy with X-ray absorption spectroscopy, chemical and morphological changes occurring heterogeneously throughout a battery can be observed under realistic cycling conditions. This allows researchers to identify specific, localized hot spots that lead to battery failure. With this information in hand, researchers can devise ways to avoid battery failure and conduct more systematic searches for electrode materials that continue to perform at high capacity over the lifetime of an electric vehicle. By imaging across an absorption edge, Li-ion diffusion through electrode material can be tracked through a change in oxidation state at a length scale of a few tens of nanometer. For example, as illustrated in the graphics above, in the conversion of LiCoO₂ (red) micron sized particles to Co metal (green) nanoparticles during discharge, the lithiation front begins at the surface of the LiCoO₂ particles and travels inward, cracking and pulverizing the particles as the front moves through them (Electrochim. Acta **2017**, 247, 977-982).

X-ray scattering, spectroscopy and imaging will also enable better understanding of advanced materials processing, including additive manufacturing. Since the ultimate structure and morphology of a material depends on its synthetic conditions, studying this in real time provides a way to favorably control and tune the structure and morphology. For instance, understanding structure-activity relationships for nanocrystal catalysts requires synthesizing phase pure highly monodisperse nanocrystals. When such systems have been synthesized with control over the size, shape and composition, the nature of the catalytically active site can be inferred from the resulting morphologically dependent activity maps. However, synthesizing such systems has previously required an enormous investment of resources dedicated to optimizing each synthesis. Monitoring the synthesis *in-situ* using simultaneous SAXS/WAXS can expedite this synthetic development by giving researchers immediate feedback as to how and why differing synthetic conditions lead to differing morphologies and compositions, while simultaneously identifying the critical parameters in the synthesis and synthetic mechanisms.

An *in-situ* materials synthesis capability that combines Pulsed Laser Deposition (PLD) and Molecular Beam Epitaxy (MBE) is being developed for the new ARPES end station on beam line 5. A combination of the state-of-the-art ARPES system and a sophisticated thin film growth system will greatly enhance the applicability of ARPES. We also envisage both *in-situ* scattering and spectroscopy of atomic layer deposition (ALD) to be performed.

Theory Institute for Materials and Energy Spectroscopies (TIMES)

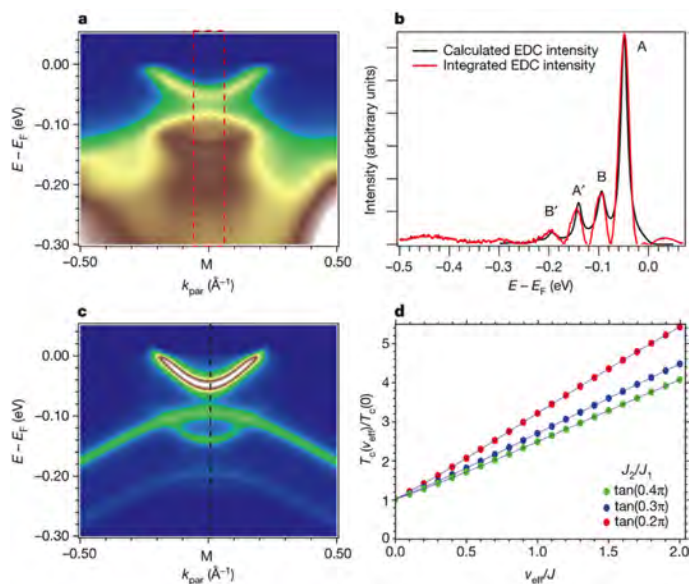
Advanced spectroscopic techniques play a pivotal role in detailed explorations of the electronic, geometric, and excited state properties of atoms, molecules, and their complex assemblies. *In-situ* and *operando* spectroscopic methods allow this exploration to include how the chemical and physical properties of matter respond to changes in temperature, pressure, electric and magnetic fields, or other external control parameters. This exploration is not only of intrinsic scientific interest, but also key for designing new materials with properties tailored for energy, catalysis, and other technological applications, and for the understanding of the electronic and geometric structural changes of inorganic and bio-inspired catalysts as they perform their function.



Complementary advances in the theory of X-ray spectroscopy must be achieved to expand the interpretive power of these advanced X-ray spectroscopies. The Theory Institute for Materials and Energy Spectroscopies (TIMES), a collaboration between the Stanford Institute for Materials and Energy Sciences (SIMES), SSRL, and LCLS, will develop advanced theories and numerical algorithms, and perform the associated simulations, to harness the potential of the advanced spectroscopic methods being developed and implemented at SSRL and LCLS (web.stanford.edu/group/TIMES). This will be accomplished through three interconnected efforts: (1) the creation of SLAC collaborations between theorists, staff and beam line scientists, experimentalists, and computational scientists; (2) the development of new theoretical paradigms and the extension of current models to provide a robust predictive basis for interpreting spectroscopic data; and (3) the development of user-friendly computational tools capable of simulating current and future experiments. This synergy between theory and experiment will enable the full exploitation of high-throughput, high-resolution modern instrumentation.

4.1.2 Strongly Correlated Electron Materials – Quantum Materials

The emergent phenomena of quantum materials present exciting opportunities to understanding and ideally control the transport of energy and information in strongly correlated electron systems. Creating and controlling the coherent transport of energy in condensed phase materials presents a compelling opportunity to greatly improve the energy efficiency of materials through the elimination of dissipative energy loss. For example, harnessing coherent transport would enable the dream of resistance-free conduction and points to the importance of Understanding How Collective Function Emerges from Constituent Interactions, particularly in strongly correlated electron systems. Important examples of emergent phenomena in quantum materials include high temperature superconductivity, colossal magnetoresistance, multiferroics, and topological insulators. These emergent phenomena result from the strong correlation between electrons present in these materials, correlations that are controlled by lattice, orbital, and spin excitations.



Single-unit-cell iron selenide grown on SrTiO₃ (1UC FeSe/STO) has recently demonstrated signatures of superconductivity close to the boiling point of liquid nitrogen, a record high T_c for iron-based superconductors that is also a dramatic enhancement compared with bulk FeSe. Understanding the causes for such an enhancement would lead to ways of engineering higher T_c 's and possibly help explain how high- T_c emerges in systems such as the cuprates. SIMES and SSRL scientists have used ARPES to study in-situ the 1UC FeSe/STO system grown by MBE. So-called shake-off bands (see Figure) are observed, which manifest as replicas of the original band structure shifted downwards in energy. These shake-off bands are the fingerprints of strong coupling between electrons in FeSe film and phonons in STO substrate, which is responsible for the T_c enhancement. By extracting the relative intensities of the shake-off bands the researchers were able to extract the magnitude of the electron phonon coupling and estimate the enhancement of T_c . The enhancement obtained agrees qualitatively with materials which have similar band structures in bulk but do not have the additional phonon coupling due to STO. *Nature* **2014** 515, 245

As better controlled model systems become available, a sophisticated understanding on the universality of these diverse materials will lead to great revelations influencing science beyond these specific properties. There is no doubt that surprising discoveries will be made that dramatically advance our understanding of these materials, such as the unexpected discovery of iron-based high temperature superconductors,

and the recent observation that the superconducting transition temperature in single layer chalcogenides can be significantly increased through coupling to substrate phonons.

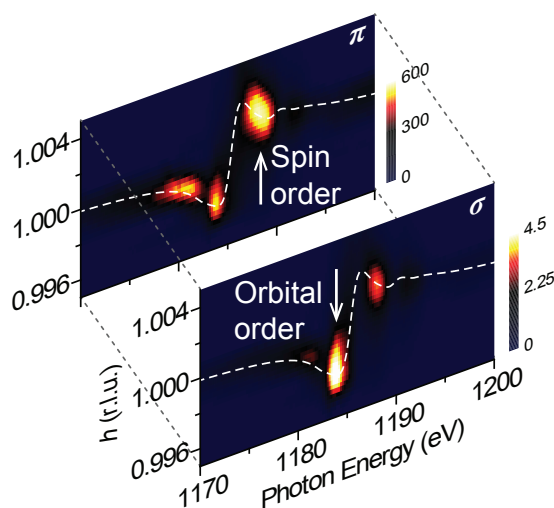
Just as these new materials have emerged, the lightsource tools for their study have also evolved substantially. In studying strongly correlated electron materials, a number of synchrotron-based experimental tools have gained prominence. Among the most important are:

- **Angle-resolved Photoemission Spectroscopy (ARPES):** As demonstrated by its strong impact on the understanding of high- T_c superconductors and topological insulators, high-resolution ARPES has proven to be the most direct and powerful experimental probe of the electronic structure of quantum materials. With the extremely high angular and energy resolution now achievable, this technique is capable of probing the electronic structure with unprecedented precision and sophistication – information that forms the foundation for a comprehensive understanding of complex quantum materials. SSRL will vigorously pursue ARPES studies of new materials, with the enhancements that the combination of the existing BL5-4 and the new BL5-2 will provide. SSRL will also pursue the development of a spin detector, which will play an important role in the characterization of topological insulators and other systems with strong spin orbit coupling.
- **Integrated Materials Growth Capabilities:** SSRL, in collaboration with SIMES, have integrated materials growth methods with the ARPES capabilities at beam line 5. This enables going beyond high energy and momentum resolution, through the integration of sophisticated material synthesis capabilities, including both Molecular Beam Epitaxy (MBE) and Pulsed Laser Deposition (PLD), enabling the scope of materials and structures to be investigated with the ARPES technique to broaden significantly. The past decade has witnessed revolutionary advances in the growth of novel materials and precise atomic control of thin films now happens on a routine basis. Layer-by-layer growth of exotic crystals and heterostructures opens up opportunities to fabricate materials with tailored properties. Such control enables researchers to better understand how structure, and the precise control of structure, affects physical properties. Therefore, the marriage of the sophisticated thin film growth techniques and the modern ARPES technique will enable new frontiers in quantum material research.

SSRL has strengthened the already strong foundation in photoemission spectroscopy with a new state-of-the-art undulator branch line for ARPES (BL5-2; see above). This addition will completely modernize and greatly expand the capabilities of the preceding branch-line with full polarization control and extended photon energy range, spin-resolved electron detection, and a significant improvement in flux and beam spot size. Complemented by the ultra-high resolution capability of the NIM branch line 5-4, we expect this beam line will become a leading photoemission facility in the world. In collaboration with SSRL, SIMES has developed dedicated MBE and PLD systems to capitalize on the full potential of this new ARPES end station. The recent success

in the *in-situ* ARPES study of single-unit-cell FeSe grown on SrTiO₃ by MBE (see Figure previous page) has demonstrated great potential in this synergetic investment by SSRL and SIMES.

Full characterization of the novel properties of reduced dimensional systems requires additional characterization tools. While the electrons transport the unique properties of strongly correlated systems, the nuclear arrangement dictates the potential within which these electrons move, creating a synergistic need for interfacial X-ray scattering in energy and quano of a strong research program in catalysis research, with the objective of forming an integrated program combining theoretical work with experiments in catalyst synthesis, characterization and testing. Catalyst characterization, particularly under working conditions, is central to the SSRL Strategic Plan, and the atomic resolution and specificity of X-ray methods naturally address many key questions in catalysis research.



Resonant soft X-ray scattering provides sensitive probe of charge and spin order in strongly correlated electron systems. Results shown for resonant scattering at the Gd M₅ edge of GdB₄ (Phys. Rev. Lett. **2016**, 117, 216404).

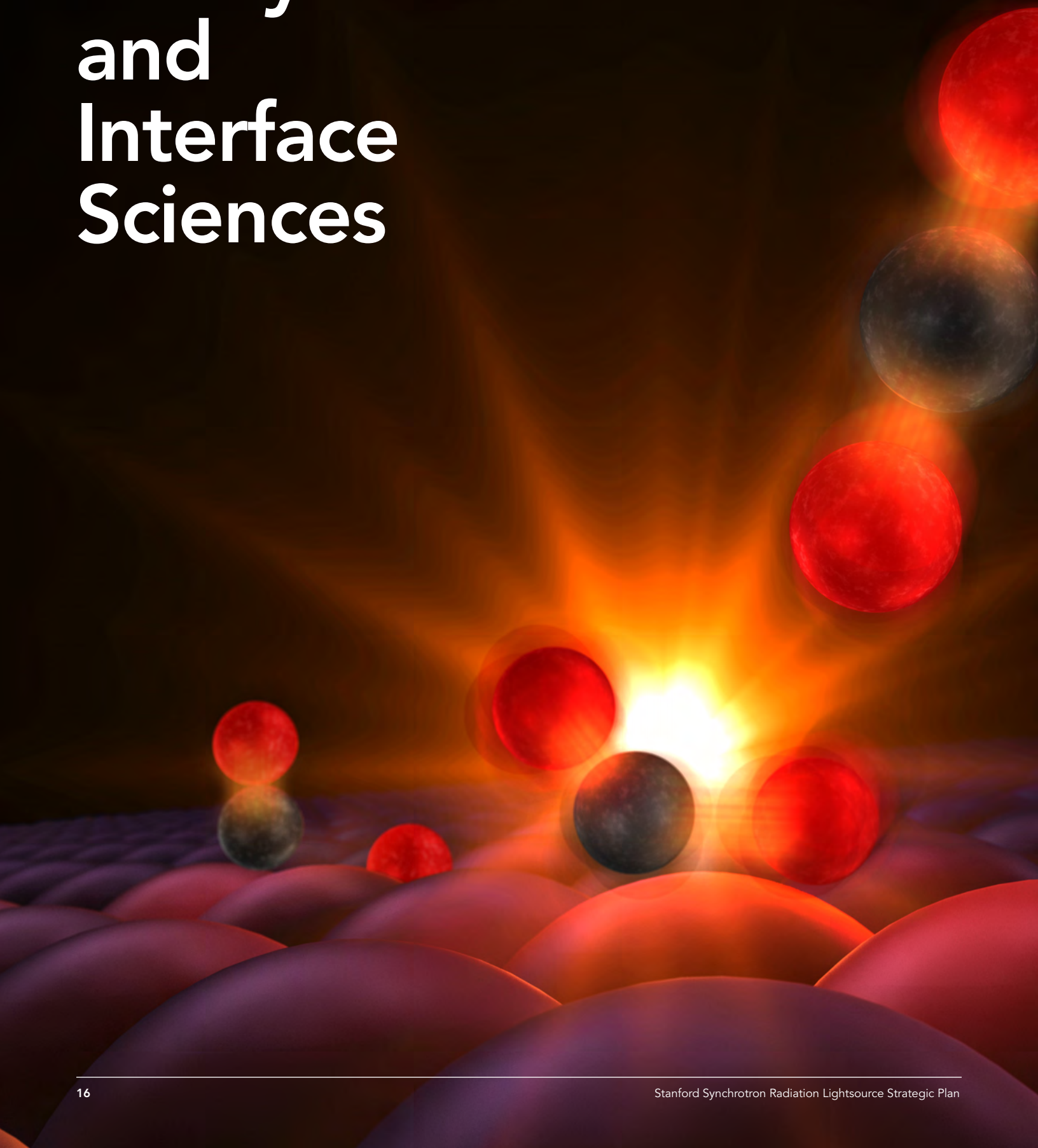
- **Interface Scattering:** In addition to charge, spin and orbital degree of freedom, the atomic structure of the strongly correlated electron materials is of considerable importance, as it dictates the emergent electronic properties. This is especially important for hetero-interfaces in metal oxides, such as LaAlO₃/SrTiO₃, where the precise atomic arrangements determine if the interface is doped n- or p-type. The quantum materials program will build on SSRL's strength in materials scattering to develop a program in hetero-interface structure determination on the new BL17-2, closely coupled to the SSRL spectroscopy program.
- **Resonant Soft X-ray Scattering (RSXS):** Recently, the effect of spin-orbit coupling in strongly correlated materials has spurred significant interest in the physics and materials science communities since the combination of strong spin-orbit-coupling and electron correlation lead to new emergent phenomena. Examples include exotic magnetism, spin liquids, and unconventional high-T_c superconductivity. To paint a complete picture of strongly correlated electron materials, it is important to complement spectroscopic approaches, such as ARPES, with X-ray scattering techniques. Resonant elastic soft X-ray scattering (RSXS) provides a powerful method for observing the spin, charge, orbit and lattice ordering that underlies many of the emergent properties of quantum materials. SSRL has recently completed the commissioning of an RSXS endstation at BL13-3 and opened the facility for general user operations. The RSXS endstation will be further advanced by the integration of a superconducting transition edge sensor detector with eV energy resolution. The energy resolving power of this detector will enable the suppression of inelastic X-ray scattering, and significantly enhance the signal-to-noise of the RSXS signal, opening up new science.

4.1.3 Summary of Planned Future Capabilities

The scientific strategy is driving the need for and planned around the following beam line and instrument developments:

Science themes	Experimental technique / beam line / instrument			
	Spectroscopy	Hard and soft x-ray scattering	Imaging	
Strongly correlated electrons	ARPES	Undulator beam line; diffractometer	STXM	
Energy: Photon conversion and batteries	Micro-focus spectroscopy	Advanced spectroscopy (x-ray inelastic scattering)	Undulator beam line; diffractometer	Upgraded TXM X-ray tomography
<i>In-situ</i> growth and synthesis		Advanced spectroscopy (x-ray inelastic scattering)	Undulator beam line; diffractometer	Upgraded TXM X-ray tomography
Picosecond time domain			Undulator beam line; diffractometer	

Chemistry, Catalysis and Interface Sciences



4.2 Chemistry, Catalysis and Interface Sciences

Catalysis plays a vital role in the world economy and human prosperity, underpinning for example fuels production and chemical and materials synthesis. Catalysis plays a significant role in fertilizer production to support the earth's burgeoning population, and in environmental remediation and control of pollutants – the 3-way catalytic converter in every automobile sold today being the most publically visible manifestation. Catalysis has also been the focus of six recent Nobel Prizes in Chemistry, demonstrating the centrality of catalysis to the chemical sciences.

SLAC's Strategic Plan has emphasized the development of a strong research program in catalysis research, with the objective of forming an integrated program combining theoretical work with experiments in catalyst synthesis, characterization and testing. Catalyst characterization, particularly under working conditions, is central to the SSRL Strategic Plan, and the atomic resolution and specificity of X-ray methods naturally address many key questions in catalysis research.

SSRL's chemistry, catalysis and interface science program will expand its existing specialized spectroscopy, imaging and scattering and diffraction capabilities, in close collaboration with the user community, to develop primarily molecular-level characterization techniques of catalysts under *in-situ* reaction conditions – whether the catalysis be molecular, thermal, electro- or bio-catalysis. The focus will be on enabling the understanding of the fundamental underlying electronic and structural properties, and course of chemical reactions, on relevant scales of space, time and energy. This includes the development of optical pump X-ray probe methods to investigate photocatalytic reactions. The Co-ACCESS program provides research capabilities to external collaborators including catalyst synthesis, off-line catalyst treatments, off-line experimental testing and prototyping/testing of experimental cells.

In the coming years, the emphasis will be on selected areas, such as photo- and electro-catalytic reactions, reactions related to producing hydrogen from solar water splitting, alcohols from the cellulose of plants, and hydrocarbons from recycled carbon dioxide, together with transformative breakthroughs in petroleum-based catalysis. Special emphasis will be given to the following developments:

- Bringing an integrated view of chemical reactivity and catalysis, where investigations of molecular, biological, nano-, environmental, and catalyst materials enable a comprehensive view of chemical reactivity in homogeneous, heterogeneous, biological, and environmental systems.
- *In-situ*, real-time characterization tools, based on hard, tender, and soft X-ray absorption (XAS) and emission (XES, PES) spectroscopies, inelastic scattering-based methods (RIXS), XAS imaging, X-ray microscopy and tomography with XANES and EXAFS capabilities, and X-ray scattering/diffraction, over multiple time and length scales.

- Integrating non-synchrotron based characterization and analytical tools, such as mass spectrometry, laser Raman spectroscopy, Fourier-transform infrared spectroscopy and UV-vis spectroscopy with the synchrotron techniques and facilities.
- Enabling picosecond resolution studies of photocatalytic and photochemical reactions and bridging scientifically to LCLS and LCLS-II for research in the ultrashort time domain.
- Coupling experimental techniques closely with theory, modeling, and new analysis methods.

We will expand on, and create new, partnerships with a number of institutions and industry in energy and catalysis research, and actively seek collaboration with leading groups in the Western U.S., as well as strategic partners throughout the world. We will also seek collaboration with other national laboratories, DOE Energy Innovation Hubs like the Joint Center for Artificial Photosynthesis (JCAP), Energy Frontier Research Centers and Bioenergy Research Centers, industry, and Stanford University Institutes, Centers and Departments. The continued strengthening of our scientific partnership with the SUNCAT Center for Interface Science and Catalysis at SLAC and Stanford (see Sidebar) is a central component of our strategy.

Finally, we will continue our long-standing tradition of educating and training the next generation scientific workforce through introductory and continuous learning workshops for both new and experienced users, summer schools for more advanced researchers, web-based experimental simulation and remote-access tools, and mentoring of undergraduate and graduate students, and postdoctoral scholars. Particular emphasis will be placed on catalysis-specific SSRL-related workshops and tutorials as these programs grow at SSRL.

The following sections focus on the scientific challenges in the chemistry and catalysis research directions at SSRL, new beam lines and instrumentation being developed and planned, and emerging plans for biogeochemistry.

4.2.1 Catalysts for Efficient Energy Production

Catalysis research is evolving to an integrated, continued cycle of theoretical modeling and prediction of optimized performance, synthesis of catalyst materials, and characterization in real time. Among the most important applications are catalysts for: (1) the synthesis of transportation fuels, (2) the conversion of absorbed solar energy into chemical fuels, and (3) sustainable and efficient synthesis of ammonia, methanol, and ethylene and propylene from non-fossil resources. Advances in petroleum-based catalysis also warrant attention due to their importance to the current global economy.

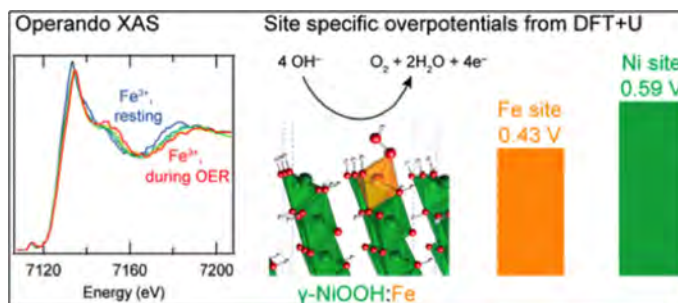
To support these thrusts SSRL will develop and provide a suite of complementary soft, tender and hard X-ray techniques, tailored to furthering our understanding of catalytic function and mechanism, from technical formulated catalysts to planar model catalysts. New directions include *in-situ* and *operando* approaches using X-ray emission spectroscopy, including

measuring data at reaction temperature, at reaction pressure, in a flow of reactant, and electrochemistry under potential control, with instrumentation implemented on existing and new insertion-device beam lines and existing bending magnet stations. The time domain will be addressed through continuous scanning techniques (minutes to seconds, with milliseconds possible), energy-dispersive X-ray emission spectroscopy, and optical pump X-ray probe measurements with picosecond resolution. Specific new directions include:

- In-situ Hard X-ray Spectroscopy:** SSRL will develop *in-situ* sample environments to apply X-ray absorption spectroscopy (XAS), partial fluorescence yield XAS, X-ray emission spectroscopy (XES), non-resonant X-ray Raman scattering, and resonant inelastic X-ray scattering (RIXS) techniques to characterize catalysts and catalyst function. The foci of the XAS developments will be on enhanced capabilities combined with improved efficiency and productivity. Simultaneous *in-situ* optical/XAS cells and instrumentation will be designed and implemented. A set of novel *in-situ* reaction cells, specific to the capabilities of each beam line and the science to be studied will be fabricated. Appropriate infrastructure necessary for *in-situ* catalysis work, such as gas handling systems, will either be implemented or improved at each catalysis-relevant XAS beam line. A repurposed wiggler end station will be converted to a dedicated catalysis XAS facility (Section 5.3.2). Finally, to improve efficiency and data quality, continuous energy scanning will be implemented, coupled with advanced algorithms to ensure that the appropriate signal/noise level is achieved for the desired information content.
- The high-energy resolution photon emission techniques, like partial fluorescence yield XAS, XES, and RIXS, are less commonly utilized within the catalysis research community and SSRL will lead their integration and application. A new undulator beam line, BL15-2, dedicated to these techniques, is central to the so-called “advanced spectroscopy” (XES, RIXS, X-ray Raman, etc.) platform for *in-situ* catalyst characterization and will become operational in 2020 .
- In-situ Tender X-ray Spectroscopy:** The tender X-ray energy region (2-5 keV) includes K-edges of the critical elements phosphorous, sulfur and chlorine, together with the L-edges of the catalytically-important 2nd row transition metals. Expanding on SSRL’s extensive expertise in tender X-ray absorption spectroscopy and advanced spectroscopy, a crystal analyzer-based X-ray spectrometer developed for tender XES studies will be fully implemented on BL6-2. The spectrometer includes a position-sensitive area detector for single-shot measurements, providing sub-eV energy resolution. Custom built reactor modules for *in-situ* catalysis studies will be integrated to meet science and experimental needs. Development of improved detection schemes underway will dramatically enhance the tender high-energy-resolution spectroscopy capability toward sub-monolayer detection sensitivity.
- 2D and 3D Imaging:** The relocation of the advanced

spectroscopy program to its own beam line (BL15-2), currently sharing time with SSRL’s hard X-ray TXM facility on BL6-2, will allow renewed emphasis and continued development of hard X-ray TXM methods at the 30-50 nm length scale, including tomography, and with XANES capabilities, for *in-situ* catalysis studies. Planned developments include implementation of complementary *in-situ* characterization techniques including laser Raman spectroscopy.

- Small-angle and Wide-angle X-ray Scattering:** The overall activity of a catalyst is often determined by the synthesis conditions. Therefore, an understanding of the



Highly active catalysts for the oxygen evolution reaction (OER) are required for the development of photoelectrochemical devices that generate hydrogen efficiently from water using solar energy. The 500-fold OER activity enhancement that can be achieved with mixed (Ni,Fe)oxyhydroxides ($Ni_{1-x}Fe_xOOH$) over that of the pure parent Ni and Fe compounds was identified using operando X-ray absorption spectroscopy with high energy resolution fluorescence detection (J. Am. Chem. Soc. 2015, 137, 1305-1313). The spectroscopy reveals that in this most active currently known OER catalyst in alkaline electrolyte, the Fe^{3+} in $Ni_{1-x}Fe_xOOH$ occupies octahedral sites with unusually short Fe-O bond distances, induced by edge-sharing with surrounding $[NiO_6]$ octahedra. Using computational methods, it was shown that this structural motif results in near optimal adsorption energies of OER intermediates and low overpotentials at Fe sites.

catalyst synthesis process is paramount to developing better catalytic materials. As such, the full power of *in-situ* SAXS/WAXS methods will be applied to relevant catalysts synthesis problems (see Section 4.1.1) in coordination with the Materials Science program.

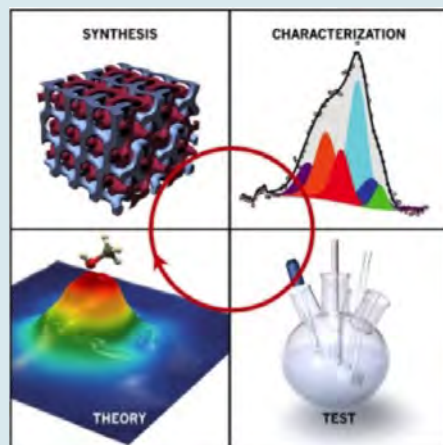
The chemistry and structure of a chemically active surface is complex, alters under reaction conditions and is often very different from the bulk of the catalyst. The search for a better (heterogeneous) catalyst is then a search for a surface that is chemically active, selective towards the desired products and resistant to corrosion and poisoning under long exposure to chemically harsh environments. Without a deep understanding of these chemical processes at atomic scale, the search is blind and depends on serendipity, and consequently slow and expensive. Atomic scale characterization and monitoring of the surface chemistry and structure of a catalytic surface under *operando* conditions can provide a rational pathway for discovery and optimization of much needed new catalysts. Moreover, the description of the chemical bond between a surface and a molecule is the fundamental basis for understanding surface chemical reactivity and catalysis. As such, SSRL will continue to develop techniques, instrumentation and methodology for the study of surface and interfaces for chemistry and catalysis, with emphasis being on *in-situ* and *operando* spectroscopy. Specific new directions will include:

- **Hard X-ray Grazing Incidence Methods:** SSRL will prioritize developments of critical instrumentation and scientific expertise in grazing incidence spectroscopy, scattering and diffraction measurements of chemically active surfaces, and solid-liquid interfaces under thermochemical, electrochemical and photochemical conditions.
- **Soft X-ray Surface Spectroscopy:** Ambient pressure photoelectron spectroscopy (APPEs) enables the investigation of the active surface of the catalyst and its chemical properties under reaction conditions. APPEs will be used to explore how temperature, pressure, electrochemical bias and composition of gasses influence the surface chemistry. Specific instrumentation will be developed for reaction temperature control using laser-based heating, and mass flow control using gas/liquid flow cells together with simultaneously detecting the products using mass spectrometry. This research represents an important stepping stone in exploring opportunities for APPEs at LCLS-II, which further enhances the capability of SLAC in the field of chemical transformations. The real-time observation of chemical transformation will address how these phases and species are involved in the catalytic reaction pathway. This combination of approaches will improve upon the current state-of-the-art.
- **High Efficiency Soft X-ray Spectroscopy:** SSRL will develop facile access to soft X-ray experiments for the catalysis community by providing novel, holistic approaches to perform electronic structure screening of a large number of catalysts, with high efficiency and productivity. This development will build on the recent successful staging of soft X-ray experiments at BL8-2 and BL10-1 that can handle large (>100) sample sets and a high degree of acquisition automation. The refinement of this capability with higher automation, pre-experiment simulation, and remote-access experiments will be aligned directly into the overall strategic direction of the SSRL catalysis strategy.
- **Ultrasensitive Soft X-ray Detection:** The soft X-ray regime remains relatively inaccessible to a large fraction of chemistry and catalysis science due, in large part, to poor spectroscopic selectivity and the high rate of X-ray induced damage. The use of soft X-ray spectroscopy for studying low-concentration atoms and molecules in a

larger matrix, such as those found in many catalysts, has been hampered by the lack of suitable detection systems that can provide sufficient sensitivity (energy resolution, solid angle, and detection efficiency). Transition-edge sensor (TES) based spectrometers, when designed as X-ray photon detectors, are quantum-noise limited devices that are capable of measuring X-ray photons in the soft X-ray regime down to a targeted 0.5-eV resolution with unprecedented solid angle. TES development promises exciting new avenues for exploring the electronic structure of reaction intermediates under operational conditions, with a target sensitivity of less than 1% of a monolayer and below 1 mM concentration in aqueous solutions. With tailored cell designs and appropriate gas and liquid flow control, this capability will encompass *in-situ* probing of supported metal nano-particles, dilute catalysts in zeolites and silica, homogeneous catalysis, and bio-catalysts in solution, as well as heterogeneous catalysis at more industrially relevant surface coverages. SSRL will work closely with the SLAC TID group led by Prof. K. Irwin on TES detector R&D and beam line tests at BL10-1, including development of integrated user interfaces, and versatile sample environments.

- **Theoretical Modeling:** A complete interpretation of the experimental spectra, and our understanding, can only be achieved through coupling with the appropriate theoretical modeling and advanced analysis tools. SSRL will work with the FEFF project, the TIMES program in collaboration with SIMES and LCLS (see previous sidebar), and other relevant groups, to develop and apply this understanding to XAS, XES, and RIXS spectroscopies as they are applied to catalysis science. In particular, interpretation of the X-ray emission data to elucidate bonding between the molecules and surface atoms, critical to the realizing the full power of the technique, will be addressed through specific collaborations with groups leading in the development of such codes.

The SUNCAT Center for Interface Science and Catalysis



The SUNCAT Center for Interface Science and Catalysis is a partnership between Stanford University School of Engineering and SLAC National Accelerator Laboratory. SUNCAT has as its primary purpose to develop the understanding of heterogeneous catalysis to the point that a set of catalyst design principles is revealed. The foundation of SUNCAT's approach is to combine computational and experimental investigations to develop a theory of heterogeneous catalysis.

The experimental aspects include new synthesis methods for atomic-scale control of surface composition and methodology for characterizing catalysts under realistic working conditions. SSRL is a strategic partner in this latter area – using the full power of the X-ray based methods to provide unprecedented understanding of the structure of the working catalyst.

4.2.2 Enzyme and Bio-Inspired Catalysts for Sustainable Energy

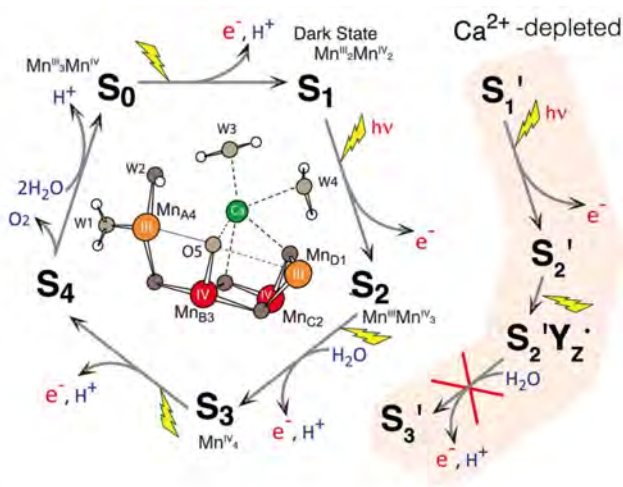
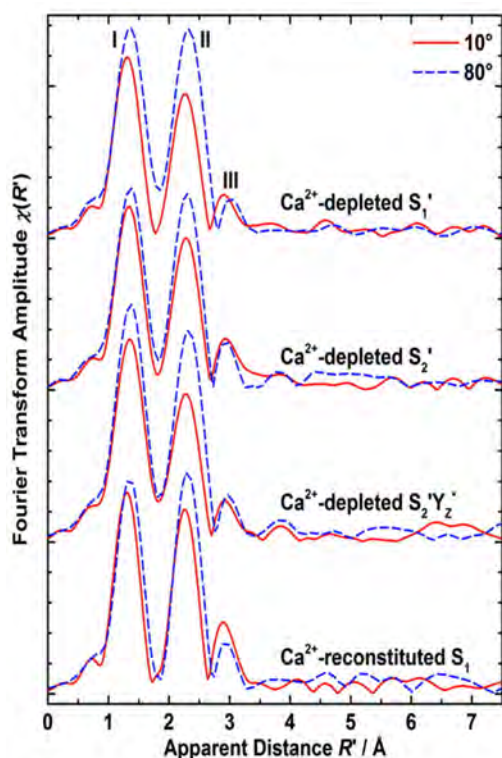
A key challenge in designing and developing alternative energy sources is the scarcity of suitable, inexpensive and sustainable catalytic systems based on relatively abundant, economically viable, and environmentally friendly first-row transition metals. Fortunately, Nature elegantly and efficiently utilizes these elements in complex metalloenzyme catalysis ranging from ammonia synthesis, production of potent fuels such as hydrogen and methane, functionalization of carbon dioxide, and the water-splitting reaction, with a high degree of precision under ambient, physiological conditions. To harness the principles of biological catalysis and develop biohybrid or bioinspired technologies for sustainable energy applications, a fundamental molecular understanding of metalloenzyme catalysis has to be developed, starting with atomic-resolution local geometric structure of the metal-based active site and extending to its detailed electronic structure in both its resting and transient intermediate states. The physico-chemical parameters emerging from such fundamental understanding of the metal active sites and their impressive catalytic activity can then be translated to industrial catalyst design to invigorate the principles of sustainable and environmentally friendly catalysis.

SSRL is playing a major national role in detailed characterization of the structural and electronic properties of metalloenzymes related to bioenergy and associated biomimetic model systems. A novel multi-edge XAS approach has been developed, in which XAS studies are performed at metal K- and L-edges, and at the complementary ligand (Cl, S, P, O, N, C) K-edges for a specific metal-ligand containing system, allowing for a holistic experimental definition of the electronic and geometric structure of the catalytic active site. This ability to conduct science-driven XAS experiments at a range of very soft

to hard X-ray energies is a unique strength of SSRL. Similarly, instrumentation and methodology has been developed for polarized single-crystal XAS, which provides unique capabilities for electronic and geometric structural determination of the active site by separately probing individual metal-ligand interactions by aligning the polarization vector along the bond of interest. In recent years, SSRL has added biological RIXS and $K\beta$ XES to expand the spectroscopic toolkit available for bioenergy focused science.

Specialized beam lines and instrumentation will continue to enable hard and tender X-ray absorption spectroscopy of the active sites in metalloenzymes, biomimetic model systems, and homogeneous catalysis systems with bioenergy applications, with additional developments described in Section 4.3.3. The X-ray measurements will be combined as appropriate with *in-situ* methods, such as electrochemistry and photo-excitation, as described in Section 4.2.1. Future plans will in particular be targeted on:

- Advanced Spectroscopy:** The new undulator beam line BL15-2 for “advanced spectroscopy” (e.g. non-resonant X-ray Raman scattering, hard X-ray emission spectroscopy, and resonant inelastic X-ray scattering (RIXS)) will add a new dimension to these studies, as it will enable the combination of static and time-resolved X-ray absorption and emission experiments on systems such as nitrogenases, hydrogenases and photosystem centers. This will be coupled to genetic manipulations and/or chemical and photo-stimulated creation of reaction intermediates. Such intermediates might display minor but very indicative changes in the electronic structure, which will be detectable through high-energy resolution X-ray emission spectroscopy, coupled to similar measurements at LCLS for the fastest time domains. Added features at BL15-2 will include *in-situ* monitoring, such as using UV-visible spectroscopy, or IR/Raman as appropriate.



Understanding and replicating biological water-splitting reaction is the holy grail of bio-inspired catalysis. Water is converted to H^+ and O_2 by plants at the uniquely designed Mn_4CaO_5 cluster of photosystem II, which is the first membrane bound protein system involved in the catalytic pathway of oxygenic photosynthesis. The unique cluster cycles through five S-states during light-assisted catalysis, losing an electron each subsequent state. Research at SSRL has aimed at trapping these S states at high-yields and characterizing their individual geometric and electronic structures using a combination of hard- and tender-XAS, polarized single-crystal XAS and static and time-resolved XES studies. (Science 2013, 340, 491-495; J. Phys. Chem. B 2015, 119, 13742–13754)

- **Tender X-ray Absorption and Emission Spectroscopy:** A combination of tender energy (2-5 keV) XAS (on existing beam lines), XAS imaging (at the micron level), and X-ray emission using the new tender X-ray instrument described above, will provide an unprecedented combination of information from complementary ligand and metal L-edge X-ray absorption and emission studies.
- **Theoretical Modeling:** Development of an integrated theoretical approach for spectral simulations of XAS/XES spectroscopic data, involving the specifics of biological environments, will be coupled to analysis and modeling of experimental data, through in-house research coupled with collaborations with groups that develop associated theory and codes, including involvement in the TIMES program described in the sidebar.

4.2.3 Geochemistry and Biogeochemistry for Subsurface and Ecosystem Science

Understanding and mitigating the environmental impacts of energy production are urgently needed in a world where natural systems are changing rapidly under the pressure of climate forces. Coupled biological-geochemical reactions occurring in soils, shallow aquifers, and geological reservoirs are profoundly important within this context. Biogeochemical processes govern the ability of soils to absorb and release greenhouse gasses, the movement of contaminants and nutrients in response to climate and land use change, the prudent extraction of lower-impact fuel resources, and the safe disposition of captured CO₂ and nuclear waste. Chemical processes in soils and the subsurface are driven by reactions occurring at the molecular scale at interfaces between water, minerals, and biological surfaces, and in complex natural mesoscale systems in which dimensions range from 100 nm to kilometers. Full understanding of these natural systems often requires consideration of both bio and geo components and how they interact. Several recent DOE reports have stressed the need to understand the molecular-scale basis of these processes, the most recent one being a joint BES, BER, and ASCR 2016 report on Basic Research Needs for Environmental Management.

SSRL will respond to these needs through scientific leadership and continued enhancement of experimental synchrotron techniques, to provide information about bonding environments and electronic structure under *in-situ* conditions over a continuum of length and temporal scales, and to serve the needs of the national and international geochemistry and biogeochemistry communities. X-ray absorption spectroscopy and XAS microprobe imaging continue to drive scientific frontiers. New and evolving research thrusts bring specialized analysis requirements. Strategic capability needs that will guide the development of beam line resources over the next few years include:

- **Soft X-ray Absorption Spectroscopy:** Biogeochemical cycling of C and N are high priority research subjects because of their roles in climate change. The pursuit of a deeper understanding of the C and N cycle drives a wide range of biogeochemical research activity funded by DOE,

USDA and NSF. XAS has, in this context, a high value as a research tool for evaluating the chemical state of carbon in environmental systems, however, there are no soft X-ray synchrotron X-ray beam lines in the U.S. with the required combination of experimental chambers, sample environments, detectors, and optics for environmental science. SSRL will vigorously pursue the creation of such a facility, potentially on BL8-2, to provide this capability to the scientific community.

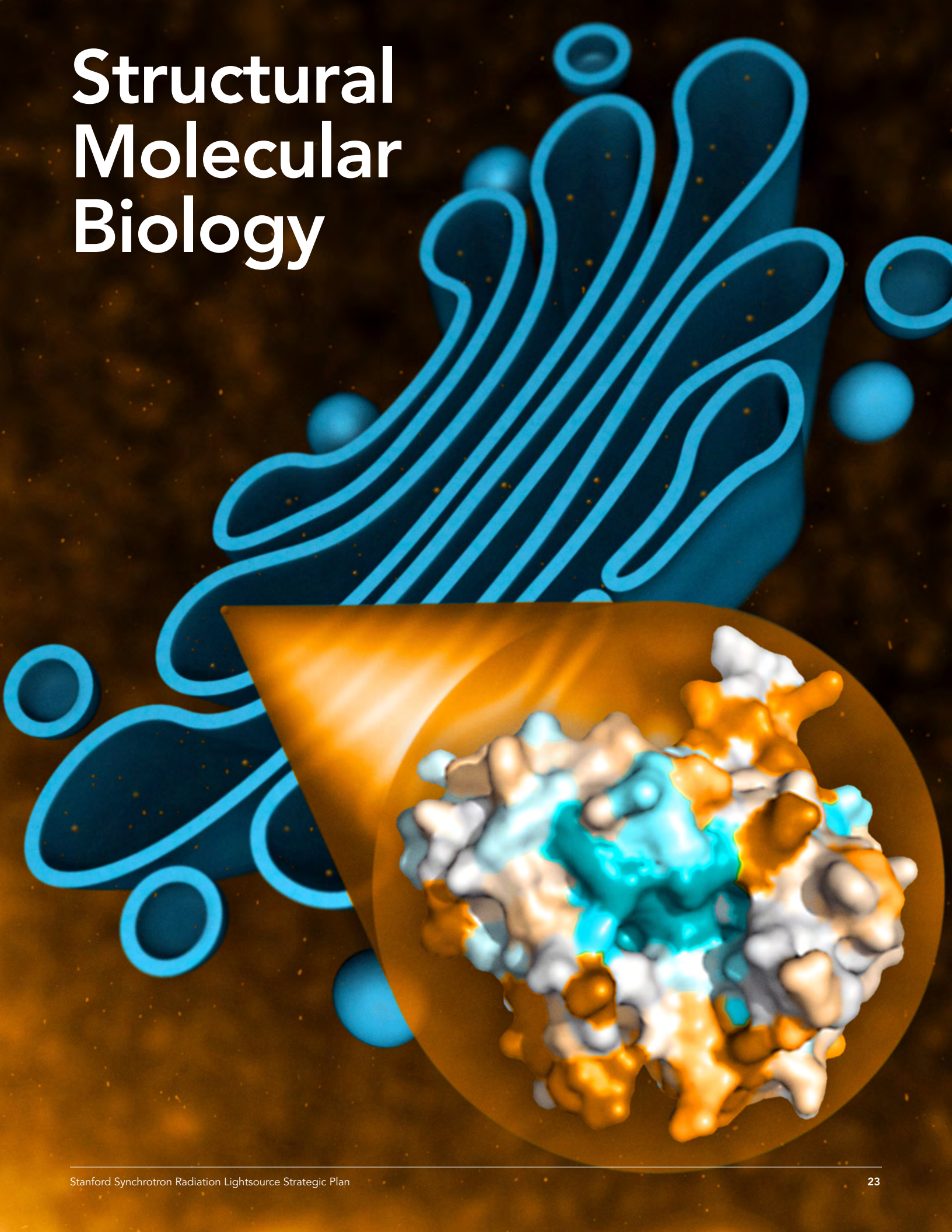
- **Time-resolved X-ray Absorption Spectroscopy:** Dynamic processes are of intense interest in geochemistry and biogeochemistry. Little is known in detail about specific structural transformations during multi-electron redox transitions of metal ions in natural systems. Mechanistic information derived from geometric conformations and electronic structure of intermediates is needed across a range of time scales, from femtoseconds to milliseconds. SSRL will develop methods and facilities to enable accessing the picosecond to millisecond time scale whereas LCLS-II and (if realized) LCLS-II-HE will extend these capabilities into the sub-picosecond time scales.
- **2D and 3D Imaging:** Earth materials are chemically and physically heterogeneous at length scales down to nanometers. Shales are an important example, as they are chemically and physically heterogeneous at nanometer length scales and provide a large fraction of the U.S. natural gas supply. Chemical reactions within shale control the release of environmental contaminants and hydrocarbon transport occur in pores ranging in size from nanometers to micrometers. X-ray-based computed tomography (CT) is the technique of choice for real-time imaging of chemical reactions at these length scales. SSRL will develop a CT microscope for real-time CT imaging at SSRL beam lines. Reaction cells and methodologies will be developed that will enable the measurement of micro- and nano-CT data and XAS spectra from rock samples at high pressure/temperature and under flow-through conditions.
- **Hard X-ray Absorption Spectroscopy:** Insertion-device beam lines and advanced detectors are required to provide rapid, high-throughput *in-situ* detection of dilute metals in complex natural samples (soils, sediments, biological materials) and extreme materials (tank wastes). Fluorescence signals from chemically heterogeneous matrices interfere strongly with desired fluorescence emission from elements of interest, particularly at dilute concentrations. High energy resolution partial fluorescence yield XAS at BL15-2 can address these needs.

4.2.4 Summary of Planned Future Capabilities

The scientific strategy is driving the need for and planned around the following beam line and instrument developments:

Science themes	Experimental technique / beam line / instrument				
	Hard x-ray spectroscopy		Tender/soft x-ray spectroscopy		Imaging
Catalysts for energy production	<i>In-situ</i> & high throughput XAS (wiggler and bending magnet)	Advanced spectroscopy undulator (XES, XRS, RIXS); LCLS	Tender x-ray XES and RIXS	TES detection, APPES, high-throughput XAS	<i>In-situ</i> hard and tender x-ray XAS imaging Enhanced TXM
Enzyme and bio-inspired catalysis		Advanced spectroscopy undulator (XES, XRS, RIXS); LCLS	Tender x-ray XES and RIXS		<i>In-situ</i> hard and tender x-ray XAS imaging Enhanced TXM
Molecular biogeo-, environmental and interfacial	<i>In-situ</i> , high throughput, and time-resolved XAS (wiggler)	Advanced spectroscopy undulator (XES, XRS, RIXS)	Tender x-ray XES	Soft x-ray XAS instrumentation for environmental samples	<i>In-situ</i> hard and tender x-ray XAS imaging; tomography
Picosecond time domain		Advanced spectroscopy undulator (XES, XRS, RIXS); LCLS			

Structural Molecular Biology



4.3 Structural Molecular Biology

The goal of understanding biological structure and function, and applying this knowledge to address a wide range of problems of broad societal importance has evolved into a large, worldwide multidisciplinary effort. It engages academic, national laboratory and corporate researchers whose goals range from innovative, discovery-based science through applied uses like more effective approaches to bioremediation and the acceleration of drug discovery. Knowledge in this field has relevance to solving grand challenge problems related to medicine, energy, and the environment. Within this context, the SSRL Structural Molecular Biology (SMB) program (funded by the Department of Energy, Office of Biological and Environmental Research, the National Institutes of Health, National Institute of General Medical Sciences, and non-federal partners) is focused on obtaining and utilizing biomolecular and biomimetic molecular structural information on the micron-to-atomic scale to understand function (and malfunction) of biological processes relevant to human health. The SMB program has pioneered and will continue to lead development of new and enhanced approaches for the investigation of biomolecular structure and function, making them widely and rapidly available to the biomedical, bioenergy, biogeochemistry, and environmental research communities.

The focus of the SMB program is on an integration of macromolecular X-ray crystallography (MC), biological small angle X-ray scattering/diffraction (SAXS), X-ray imaging, and X-ray absorption (XAS) and emission spectroscopy (XES) to study the most challenging and wide-ranging biological systems – leveraging on the powerful capabilities of the SSRL synchrotron (SPEAR3) and LCLS X-ray free electron laser. Collectively, these techniques provide a remarkably rich and broad window on structure and function across a range of biologically relevant length and time scales, providing the foundation to extend results at the atomic and molecular level to understanding complex macromolecular interactions, and to studies of organelle, cell and tissue organization and function.

The SMB program will work closely with the SLAC Biosciences Division in emerging scientific areas related to bioenergy and biogeochemistry, where X-ray techniques provide unique tools for studies in the length scale ranges from molecular to cellular and beyond. The current strategic emphasis at SLAC on evolving a cryo-electron microscopy (cryoEM) and cryo-electron tomography (cryoET) program, will provide a unique opportunity for joint scientific and technological developments, which will be vigorously pursued. The SMB program will form partnerships with Stanford University institutes, such as ChEM-H and Bio-X, expanding on existing joint programs for MC beam line development and science, with industry on drug discovery developments, and with private institutions on emerging scientific topics. The SMB program will engage with other user facilities, pursuing multi-user-facility arrangements in areas that provide user access to complementary techniques, such as with the Environmental Molecular Science Laboratory (EMSL) and the Joint Genome Institute (JGI), and in coordinated outreach programs within BER-funded facilities and research groups. We also plan to begin targeted outreach to potential new

SMB users at the BER Bioenergy Research Centers. The Joint Initiative for Metrology in Biology (JIMB) now resides administratively within SSRL, and we are pursuing synergistic lines of research connecting JIMB's synthetic biology and genomics expertise with SMB research and capabilities at SSRL.

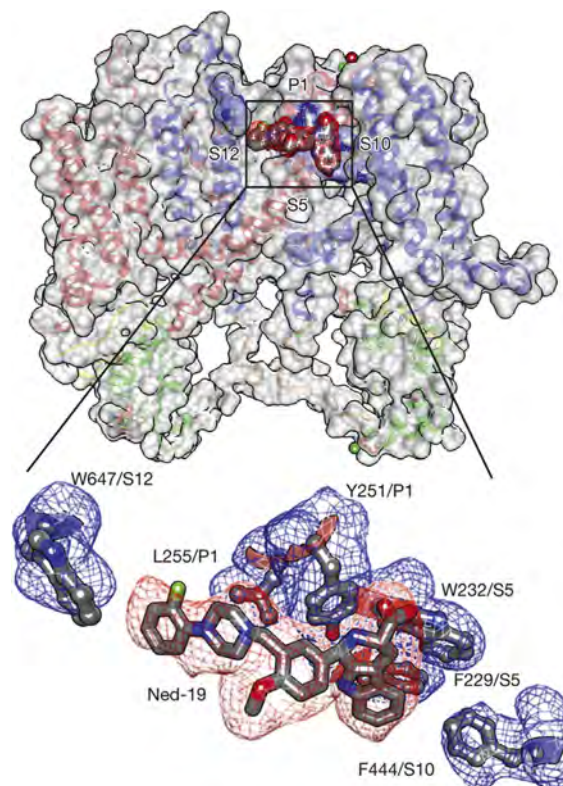
4.3.1 Macromolecular Crystallography

Macromolecular crystallography (MC) will increasingly focus on the understanding of the complex biological machinery that drives the biology in cells. This will be further enabled by the implementation of a world-wide competitive, microbeam undulator beam line (BL12-1) to be commissioned in 2020, and by a range of focused methodological and technical developments targeted towards addressing increasingly challenging biological systems. It will also be enabled through synergistic implementations of some of these developments at the Macromolecular Femtosecond Crystallography (MFX) station at LCLS, and in future coupled developments with cryoEM/cryoET.

Scientifically, a first focus will be on structural studies of increasingly large and challenging macromolecular assemblies and machinery, crucial to advancing the understanding of complex biochemical processes. A second focus will be on membrane proteins, which are of major biological importance, but exceptionally challenging to crystallize, and to study structurally at atomic resolution. While membrane proteins make up ~25% of an organism's genome, less than ~1% of their structures have been determined. With advances in crystallization, sample delivery, instrumentation, methodology and X-ray capabilities, a significant number of these structures can be determined, providing critical details for understanding a range of cellular functions regulated by these complex systems and leading to practical benefits such as the structure-based design of new and improved drugs. A third area will be in virology, gaining fundamental insights that will enable the development of vaccines or other disease prevention. A fourth area will focus on structure determination of specific chemical states of metalloproteins to understand enzymatic processes related to bioenergetics, the chemistry of small molecules like CO, CO₂ and N₂ involved in global carbon and nitrogen cycling, and in this context studying ways to mitigate radiation damage issues associated with these classes of metalloproteins. A fifth will be to pursue collaborative projects that require fully automated, sample preparation, data collection and high-throughput structure determination. A specific effort will be devoted towards structure-based drug design, emphasizing fragment-based screening and automated feedback of structural results to molecular dynamics and modelling approaches. This will be critical not only for finding new drugs for the most serious illnesses, but also in the fight against the growing problem of drug resistance.

The methodological and technological developments – summarized below – and that will enable the science outlined above, will build on the existing strong foundation of high automation with high reliability, full remote-access operation of all SSRL MC beam lines, specialized software for data collection and automation of data reduction and analysis, and high-capacity computing facilities. Specific emphasis going forward will be on the following:

- Development of advanced microfocus capabilities enabled by the new beam line BL12-1, including implementation of a 'next generation' pixel array detector, a high-speed microgoniometer, and a number of associated sample delivery systems such as injectors and multi-sample room-temperature devices.
- New automation approaches for the identification, centering and exposure of small radiation-sensitive crystals and development of new data collection methods (including at ambient temperature and using multi-crystal workflows) to advance and enable the challenging science projects.
- Development of fully integrated, remotely accessible, multi-mode spectroscopy systems for *in-situ* microcrystal identification and alignment, to determine ligand identity and oxidation states of poised intermediates, and to enable novel time-resolved experiments in crystallo.
- Synergistic developments on the new LCLS MFX station with emphasis on new and improved sample delivery and new software to address data reduction and analysis – with the goal of providing an effective Gateway between the SSRL BL12-1 (and others) and the LCLS MFX station for the global SMB research community
- Further approaches to fully automated high-throughput, multi-crystal data collection and structure determination pipelines, building on extensive developments at SSRL initially developed from within the NIH Protein Structure Initiative to accelerate fragment-based drug discovery, and/or other pipe-line approaches in collaboration with industry and research institutes

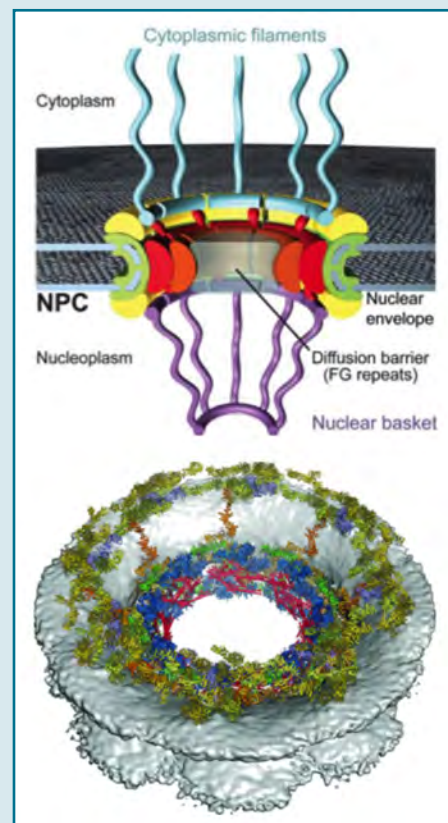


Two-pore membrane channels regulate the transfer of nutrients and other molecules in and out of our cells. Viruses such as Ebola gain entry through these membrane channels by fusing the channel open. Special characteristics of SSRL BL12-2 (high brightness and micron-sized beams) was used to solve the high resolution structure of a two-pore membrane channel in complex with the potent drug "Ned-19". The drug works by clamping the pores closed, blocking fusion, thus preventing Ebola virus and other filoviruses from entering cells (Nature 2016, 531, 258-264)

Macromolecular Crystallography and Cryo-Electron Tomography in Multi-Technique Synergy for Giant Complex Studies

The Nuclear Pore Complex (NPC), a very large macromolecular machine embedded in the nuclear envelope, i.e. the double membrane that surrounds the eukaryotic cell nucleus, is the sole gateway for the bi-directional transport of macromolecules between the nucleus and the cytoplasm. The NPCs are composed of 34 different kinds of proteins, called nucleoporins, which assemble into an eightfold-symmetric, ~1250 Å-diameter pore that fuses the inner and outer nuclear membranes. As the NPC is involved in critical processes, such as regulating the transport of RNA and ribosomal proteins, DNA polymerase, and signaling proteins, but also can be targeted by pathogens, obtaining atomic-level structural information is critical, but has been a challenge due to the size of the complex. Despite progress in visualizing the overall shape of the NPC by means of cryo-electron tomography (cryo-ET) and in determining atomic-resolution crystal structures of individual nucleoporins, the molecular architecture of the assembled NPC has remained elusive, hindering the design of mechanistic studies that could investigate its many roles in cell biology. Recently, a research group from Caltech, biochemically reconstituted the protomers of the symmetric core of the NPC, determined the interactions between them, and fitted them into a tomographic reconstruction of the intact human NPC. The interactions between nucleoporins was established via macromolecular crystallography, mainly at SSRL BL12-2, and identified flexible linker sequences that mediate the assembly of the inner ring complex and its attachment to the NPC coat. By docking these structures into the cryo-ET reconstruction, they built a near-atomic composite structure of the NPC symmetric core that contains ~320,000 residues and accounts for ~56 megadaltons of the NPC's structured mass.

With nucleoporins having been linked to a wide range of human diseases, including viral infection, cancer, and neurodegenerative disease, the structural and mechanistic details will permit a structure-function investigation of the NPC. The study also highlights the growing synergy between macromolecular crystallography and electron microscopy/tomography. (*Science* **2016**, 352, aaf 1015)



Schematic representation of the NPC (top) and overview of the composite structure of the NPC symmetric core (bottom).

4.3.2 Biological Small Angle X-ray Scattering

Biological small-angle X-ray scattering and diffraction (BioSAXS) is one of the primary tools to study the structure of non-crystalline biological macromolecular systems in solution or as partially ordered arrays of biomolecules. Such studies can be performed under near physiological conditions, require small amounts of material and are well-suited for time resolved measurements, e.g. measuring the kinetics of conformational changes, or the identification of folding intermediates under biologically relevant conditions. As a structural technique of moderate resolution (~7-10 Å or lower), but with the capability of studying very large protein assemblies, SAXS complements higher-resolution techniques such as MC, cryo-electron microscopy and NMR, since solution SAXS can model very large multi-component molecular complexes, whose overall structure is unknown. Where higher resolution structures of individual components are available they can be included in modeling to obtain a higher resolution perspective.

Scientific applications within the SSRL SMB program focus on a number of systems with specific relevance for understanding biomolecular structure and function, especially of complex systems, and will continue to drive developments at the BioSAXS beam line facility. Prime examples are the

maturation process of virus particles or protein folding (development of time-resolved SAXS), amyloid precursor proteins (*in-situ* size exclusion chromatography, SEC-SAXS), protein families related to the human and bacterial microbiomes (automation and full biophysical characterization pipeline), systems for drug delivery (lipid and fiber diffraction instrumentation), and mechanistic insights into biological process through understanding of conformational flexibility differences in solution and crystal forms (multi-method approaches). Future developments also include the further integration and simultaneous use of non-X-ray-based characterization tools, such as refractive index, UV/Vis absorption, and static and dynamic light scattering, and coupling to cryoEM developments and applications.

The BioSAXS beam line BL4-2 features state-of-the-art experimental facilities for solution scattering, lipid membrane and fiber diffraction at moderately high to very small scattering angles through a flexible camera approach. Specialized sample handling devices include an automatic fluid sample changer for high-throughput/small-volume measurements of large number of samples, *in-situ* size exclusion chromatography with the sample measured as it is eluted from the column. The facility features high-performing, interchangeable large area CCD and pixel array detectors and includes

advanced instrument control and on-line data processing software enabling data analysis during experiments to drive the scientific strategy. Non X-ray offline characterization tools include dynamic light scattering instrumentation, providing at times critical sample characterization information. In addition to static solution scattering, BL4-2 maintains a premier experimental setup for time-resolved studies providing access to reaction time scales in the milliseconds and longer.

Future developments will focus on:

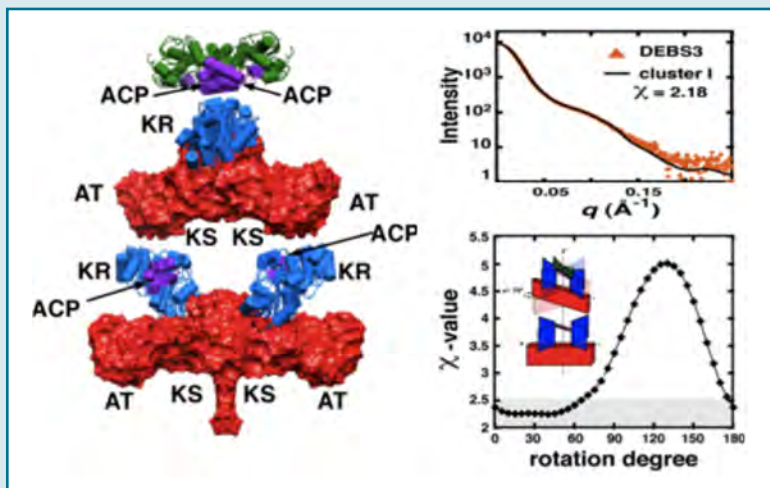
- Creating microfocus beam capabilities on BL4-2 for pushing the frontier of SAXS measurements, enabling measurements on very small sample volumes and with high time resolution
- Significantly advancing the instrumentation capability to enable measurement of high quality time-resolved SAXS data from biological samples, pushing the time resolution into the sub millisecond scale as well as reducing radiation damage issues for slower kinetics

- Increasing the data collection efficiency and data quality from problematic and aggregation prone samples by implementing new state-of-the-art size exclusion purification methods directly coupled to automated SAXS data collection
- Refining the automated data collection and reduction system for high-throughput solution scattering to minimize sample consumption, increase reliability and real-time data processing, and enable remote-access data collection for users.

Nature's Assembly Line of Metabolic Products

Secondary metabolites produced by microorganisms have a market value of over \$30 billion annually, and nearly half of these compounds are naturally produced by bacteria in the phylum, Actinobacteria. Although there are over a dozen classes of secondary metabolites, the polyketides are arguably the most versatile, with medically relevant activities including antibiotic, anticancer, immunosuppressive, anti-parasitic, and cholesterol-lowering properties. As an example the actinomycete *Saccharopolyspora erythraea* is one of many soil-dwelling bacteria that employ gigantic enzyme catalysts called polyketide synthases (PKSs) to construct complex polyketide products, such as the aglycone precursor of erythromycin, 6-deoxyerythronolide B, which is synthesized by a 2-MDa trimeric protein complex called the 6-deoxyerythronolide B synthase (DEBS). This megasynthase is comprised of three unique homodimers. Each homodimer contains two clusters of catalytically independent enzymatic domains, referred to as a module, which catalyzes one round of polyketide chain extension and modification. Since the discovery of the modular nature of PKS assembly lines, considerable research has focused on engineering PKS chimeras by swapping domains in and out of modules, as well as by mixing and matching phylogenetically distinct modules to produce new compounds. While this strategy is sometimes effective, the engineered systems are invariably inefficient, underscoring the importance of pursuing a deeper understanding of the relationship between PKS structure and function.

Using the known high-resolution crystal structures of the single components of the DEBS modules, size-exclusion chromatographic separation coupled with small angle X-ray scattering analysis (SEC-SAXS) was used to determine the relative position of these components within the catalytically active bimodular complex using ridged body refinement, and for the first time determine the molecular architecture of a functional DEBS bimodule (left Figure). The resulting structure of this 660 kDa large protein assembly suggests that the modules stack collinearly along the 2-fold axis of symmetry and that the ACP domain, which sequentially shuttles the growing polyketide chain to each active site in a module, can be positioned within 20 Å of each active site without dramatically changing the macromolecular architecture. The right hand top panel depicts the SEC-SAXS data together with the model fit of the complex, while the bottom panel illustrates possible relative rotation of the top and bottom modules, where the precise spatial positions and protein-protein interactions that the domain samples during catalysis will require higher-resolution insights. Further elucidation of the catalytic mechanism will unquestionably enhance our understanding of assembly line PKS function and our ability to engineer these remarkable megasynthases for the production of "unnatural" natural products for a wide variety of applications. (*Nat. Struct. Mol. Biol.* **2014**, *21*, 1068-1074)



4.3.3 Biological X-ray Absorption and Emission Spectroscopy

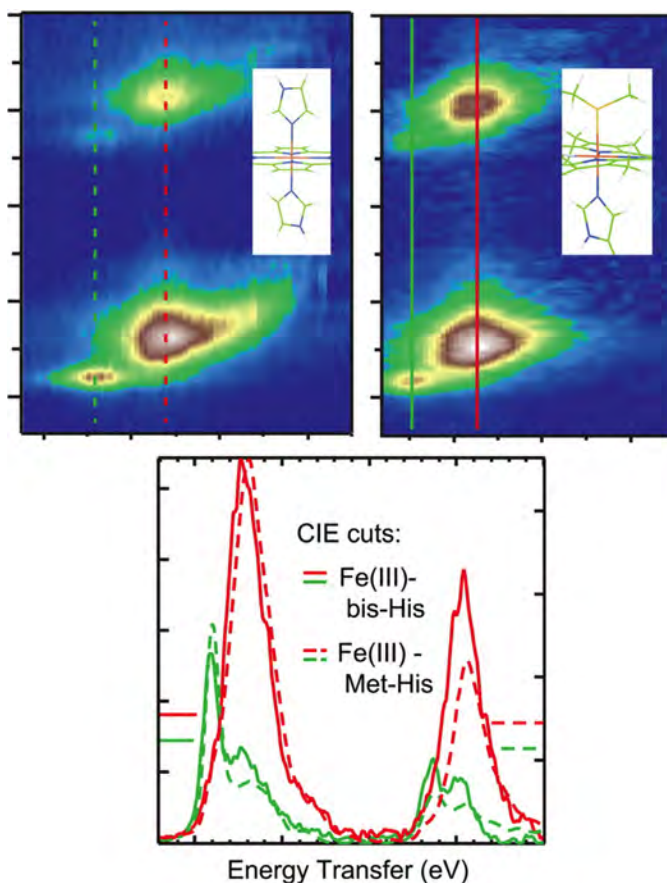
Metal ions play crucial roles in life processes - catalyzing central biochemical reactions, facilitating electron transfer in key metabolic processes, preventing cytotoxicity, modulating signal transduction across cellular systems, and participating in neurological disease control, prevention of a wide-range of diseases and participating in key processes of the microbiome with critical implication to bioenergy, the environment, and human health. X-ray absorption (edge and EXAFS) and emission (XES) spectroscopies provide comprehensive electronic and geometric local structure information about the metal active sites in biomolecules addressing structure-function relationships. These versatile techniques elucidate catalytic intermediates by studying biological processes in the fast time domain. XAS imaging (or spectromicroscopy) provides spatially resolved information about metal distribution and speciation in materials of biological and medical relevance, ranging in length-scale from the cellular to tissue and larger. These spectroscopy techniques provide detailed atomic-level local electronic and geometric information that is highly complementary to the global geometric information obtained by MC or SAXS by directly probing chemical properties of individual metal sites in the macromolecule, in any physical state.

The SSRL SMB BioXAS program has developed one of the largest dedicated and most impactful activities in the world with optimized beam lines and specialized instrumentation and analysis capabilities for enabling biological, biomedical, and bioenergy research. The future scientific emphasis will be to capture transient reaction intermediates using highly sophisticated measurement and detector systems, enabling understanding of catalytic mechanisms in systems that are relevant to core biological processes, such as oxygenase and oxidase catalysis, nitrogen reduction and oxidation, methane formation, CO₂ reduction, and electron transfer reactions. Future developments include:

- Building an advanced spectroscopy facility on the new microfocus undulator BL15-2, pushing the forefront of biological XES/RIXS/HERFD-XAS applications to increasingly dilute systems for static and dynamic measurements, including experiments in the ns-ms time domain at SSRL and with strong coupling to LCLS experiments into the fs time domain.
- New instrumentation development for parallel measurement of polarized single crystal XAS and protein crystallography data on increasingly smaller crystals, coupled with implementation of crystal analyzer detection approaches for enhanced S/N ratio and site-selective X-ray absorption spectroscopy capabilities.
- Implementation of remote access data collection and analysis capabilities for XAS measurements with increased automation, rapid scanning and high-throughput data measurement on selected biological X-ray spectroscopy beam lines, significantly extending the potential

user base and facilitating ease and flexibility of data acquisition.

- Development of an integrated approach for spectral simulations of XAS/XES spectroscopic data, coupling theoretical interpretation with experimental data, assisting in design of optimized data measurement protocols and providing a direct method for both geometric and electronic structure correlation to spectroscopy.
- Instrumentation and software developments for imaging of biological specimen, for integrated and seamless transition from mm to sub-microns in length scales, and over energy ranges from tender to hard X-rays, will be further developed, and extended to XAS tomography and XAS confocal imaging.



The Figure shows Fe 1s2p RIXS data for cytochrome c (histidine and methionine axial ligands; top right) and a related Fe(III) model complex (two axial histidine ligands; top left). The data indicate increased covalency for the Fe-S(Met) axial bond relative to Fe-N(His) as well as a higher degree of covalency for the ferric relative to the ferrous state in cytochrome c. (*Coord. Chem. Rev.* **2017**, 345, 182-208)

Fe 1s2p RIXS combines low-lifetime broadening, feature-rich 1s→2p L-edge XAS (d) with the measurement ease of hard X-ray 1s→3d XAS (c), yielding two-dimensional high resolution spectra that furnishes differential orbital covalency information, in addition to spin- and oxidation-states of the element of interest.

RIXS and Kβ XES are electronic-structure determination techniques, which utilize an analyzer spectrometer setup to provide high energy-resolution emission and RIXS spectra allowing measurement in static and ns-ms time domain. Together with soft-, tender- and hard-XAS, advanced spectroscopy is a powerful tool in the SSRL X-ray spectroscopy toolbox.

4.3.4 Training the Next-Generation Workforce

As a tightly integrated feature within the Structural Molecular Biology program activity, we will educate and train the next generation structural biology scientists through technique- or science-focused workshops, summer schools, web-based tools, mentoring of students and postdoctoral fellows, and by bringing the synchrotron to the home laboratory through advanced remote-access developments in tandem with rapid access beam time mechanisms. This will be coupled to developments of on-line learning tools, including simulation of experiments, in collaboration with Stanford University's



4.3.5 Summary of Planned Future Capabilities

The scientific strategy is driving the need for and planned around the following beam line and instrument developments:

Structural biology area	Experimental technique / beam line / instrument				
	Diffraction		Scattering	Spectroscopy and imaging	
Macromolecular crystallography	Micro-beam undulator for micro- to nano-crystallography	Micro- to nano-crystallography instrumentation at LCLS			
Biological SAXS			Microfocus optics; high-speed detector		
Biological XAS and XES				New detector approaches	Advanced spectroscopy undulator (XES, XRS, RIXS, HERFD/XAS)
Picosecond to femtosecond time domain		Micro- to nano-crystallography instrumentation at LCLS			Advanced spectroscopy undulator (XES, XRS, RIXS) and LCLS

Operational Excellence



5 Operational Excellence

5.1 Accelerator Improvement Plan

The SSRL accelerator, SPEAR3, provides a 3-GeV electron beam to deliver high brightness and high average power photons to multiple experimental stations over the soft to hard X-ray energy spectrum. SPEAR3 is operated with top-off injection at a current of 500 mA and with a reliability of typically above 97%. The SSRL strategy for SPEAR3 is to continually improve beam quality and innovate to keep SSRL competitive with synchrotron light sources around the world. An accelerator improvement plan has been developed with the following key elements:

- Performance improvements
- Accelerator reliability improvements
- Accelerator research and development
- Advancing the next generation light sources

5.1.1 Long-term Accelerator Improvements: Advancing Ultrafast Science Capabilities

The magnetic lattice design, electron beam energy, and ring circumference determines the X-ray beam brightness. SPEAR3, and many other existing storage rings, utilize a double bend achromat design resulting in transverse electron beam emittances between 1-10 nm-radians. More recently, accelerator scientists have developed multi-bend achromat (MBA) storage ring designs that promise improvements in the transverse emittance of up to two-orders of magnitude. The first such MBA storage ring, MAX-IV, has been built in Sweden, and numerous MBA upgrade plans at various stages of maturity exist at synchrotron facilities around the world.

The investment in high brightness facilities will enable advances in coherent X-ray imaging, imaging with nano-focused beams, and dynamic light scattering. The emphasis on X-ray coherence will not, however, advance all X-ray capabilities needed by the user community for discovery and use-inspired research and development. Many of the most critical scientific questions require moving beyond structure-function correlations to actually characterizing the structural evolution that dictates function in real time. Progress in these directions places equal or greater emphasis on temporal resolution than on spatial resolution. A focus on ultrafast time-resolution highlights the importance of electron bunch charge and bunch duration, properties that cannot be optimized simultaneously with the beam transverse emittance.

A key strategic driver for the SPEAR3 research and development program is a ten-fold or better reduction in X-ray pulse duration compatible with the high average flux capabilities central to core operations at SSRL. The need for simultaneous support of short pulse and high average flux operations reflects our expectation that a mature time-resolved experimental program will be 10-to-20% of our user community. This objective also has us moving away from low- α production of shorter duration X-ray pulses, because these operations negatively affected the majority of SSRL users.

SSRL, in collaboration with the Accelerator and Technology and Innovation Directorates at SLAC, is developing conceptual designs for achieving high flux, short pulse operations. The table below shows the results of an initial study. Balancing performance, cost, and technical risk, the transverse cavity design looks the most promising. Beam dynamics simulations demonstrate that transverse cavities can deliver compelling research capabilities. Current focus is on determining how to cost effectively build the RF cavities needed to achieve the accelerator performance achievable in a simulation.

Table1: Summary of performance parameters for various bunch-length reduction methods.

Short pulse mode	Pulse length (ps, fwhm)	Camshaft bunch charge (nC)	Repetition rate (MHz)	Camshaft 8 keV average flux ¹ (10 ¹¹ ph/s/0.1%BW)
Standard lattice, camshaft 13 mA	70	10	1.28	100
Low- α , $\alpha/20$	15	0.27	1.28	2.7
Standard lattice, w/ SRF	7.4	1.6	1.28	16
Ten-turn circulation ²	5.2	0.20	1.28	2
One-turn circulation	1	0.30	0.085	0.2
Crab cavity, large slice ³	6.0	2.0	1.28	20
Crab cavity, small slice	2.5	0.6	1.28	6

¹SPEAR3 BL12-2 is assumed for flux calculation ($\lambda=22\text{mm}$, 67 periods, $k_{\text{max}}=2.17$).

²Assuming beam power of 76.8 kW from LCLS-II.

³Charge given for slice from a 15.6 nC bunch with slitting. 1 MV deflecting voltage per cavity system is assumed.

5.1.2 Short-term Accelerator Improvements

Lower Emittance

A multi-year Accelerator Improvement Project is underway to increase the brightness of the SSRL photon beam lines by reducing the SPEAR3 horizontal emittance from ~ 9.6 nm-rad to 6 nm-rad. The emittance reduction is achieved by increasing the horizontal focusing of the storage ring quadrupole magnets. Delivering the 6 nm lattice in operations requires upgrades to two magnet systems – the injection magnets and the sextupole magnets. The sextupole magnet upgrade required additional power supplies and cables, which were installed in 2014. The injection magnet upgrade includes replacing the injection septum magnet. Work is progressing on the new septum magnet, with installation planned in the summer of 2019. The lower emittance lattice will then be commissioned during accelerator physics shifts in the FY2020 run, with delivery for operations expected that year.

Multi-bunch Feedback Kicker

SPEAR3 has had a transverse multi-bunch feedback system in operations since 2014. The feedback performance, however, is limited by the bandwidth and power capability of the presently installed transverse kicker. Upgrading to a high-bandwidth kicker will enhance storage ring stability and performance in several ways:

- Improve ion instability suppression, and enable running at lower chromaticity at the beginning of each run, when the vacuum pressure is recovering from the shutdown activity.
- Suppress resistive wall instabilities from future small gap insertion devices.
- Damp top-off transients faster, decreasing stored beam perturbations during top-off injection.
- Improve bunch purity with bunch cleaning to improve timing mode data quality.
- Damp multi-bunch instabilities driven by resonances in in-vacuum undulator chambers, such as those recently measured for the beam line 15 IVU.
- Possibly increase single bunch current limit.
- Enable novel timing operational modes, such as resonant crabbing and resonant pseudo single bunch.

In order to create a rapid approach, SSRL obtained a kicker on loan from the Advanced Light Source, which was installed and successfully commissioned in FY2017. The plan is to eventually replace this with an SSRL-owned device.

Pseudo Single Bunch

Implementing pseudo single bunch (PSB) operation at SSRL is a priority as part of the ongoing program to enhance timing mode operation. Pseudo single bunch is a method developed at the Advance Light Source in which the camshaft bunch is kicked on a different trajectory than the 500 mA stored in the storage ring bunch trains. PSB enables timing mode operation at one or more beam lines, while maintaining standard

operations at the rest of the beam lines. Implementing PSB at SPEAR3 will require building and installing a high repetition rate kicker with a short pulse length that can kick the camshaft bunch without affecting the adjacent bunch trains. Having camshaft timing bunch photons spatially isolated from the photons generated by the 500 mA in other bunches will benefit timing experiments by reducing sample damage and reducing background photons from the other bunches. The PSB kicker repetition rate can be varied to provide a variable repetition rate for timing experiments, from the 1.28 MHz SPEAR3 ring revolution frequency down to kHz repetition rates or lower. This provides the possibility selecting the repetition rate to match the time response of samples and detectors. A preliminary conceptual review was held in FY2017.

5.1.3 Accelerator Reliability Improvements

The projects in this category will improve the mean time between failure (MTBF) and the mean time to repair (MTTR). This is especially important for the injector, which is now over 20 years old and with some RF components over 30 years old.

SPEAR3 Low Level RF Upgrade

The SSRL booster RF upgrade, based on a modern solid state amplifier (SSA) and low level RF system, and which was completed in FY2017, will form the foundation for upgrading the SPEAR3 low level RF system to the same hardware and architecture. This will improve performance by allowing more powerful hardware and software to address more challenging problems of future SPEAR3 projects, such as improving longitudinal stability for timing mode operation. It will also improve reliability by addressing issues associated with limited personnel who are familiar with the present aging system. The new system will conform to the architecture and standards now used for RF systems elsewhere at SLAC.

Photocathode Gun Operation

SSRL intends to upgrade the injector gun cathode from a thermionic cathode to a photocathode. Photocathode operation will enable the production of a short train of several 2856 MHz bunches, rather than the couple-microsecond long train of bunches produced by the present thermionic cathode. A shorter bunch train will remove the need for the aging chopper currently in place to select bunches for injection into the booster.

Injection Interlock Upgrade

Work is ongoing to upgrade the injection interlock system required for the beam containment system for top-off injection. The new injection interlock system modernizes the existing system which pre-dates top off injection, offers expansion capacity needed for new beam lines, and introduces enhanced beam containment interlocks for beam line front end protection.

5.1.4 Accelerator Research and Development

SSRL supports a series of smaller scale research and development plans focused on both enhanced performance and improved reliability. The following projects will keep SPEAR3 at the cutting edge of accelerator research directly relevant to our mission and the accelerator research community.

RF Gun Development

The SPEAR3 accelerator group will continue to make contributions to RF gun development, including experimental work for the APEX project for the LCLS-II gun. SSRL has also recently installed a second operating one-and-a-half cell RF gun in the SSRL linac vault. This gun will serve both as a hot spare for the SPEAR3 injector, and as a research gun for medical applications. In addition, studies are ongoing with the SPEAR3 injector gun to test the viability of different laser light frequencies for photocathode operation.

Beam-based optimization algorithm development

The SPEAR3 group has a robust ongoing effort to develop beam-based algorithms for online optimization of storage rings and other accelerators. The work has included genetic optimization algorithms, particle swarm optimization, and an in-house developed algorithm named Robust Conjugate Direction Search (RCDS). This algorithm has been successfully used to optimize various parameters in the SSRL accelerator complex, including adjusting the sextupole strengths to maximize injection efficiency and dynamic aperture. It will play a critical role in commissioning the 6 nm-rad SPEAR3 optics. The RCDS algorithm has also been used at other facilities, including LCLS, IHEP, DIAMOND, ALS, and ESRF. At the ESRF, it has been used to improve lifetime and dynamic aperture for their timing mode operation.

Multi-pole injection kicker study

This study will determine if the SPEAR3 Lambertson injection method can be replaced with a multi-pole injection kicker design that does not disturb the stored beam and thus makes injection nearly transparent to users.

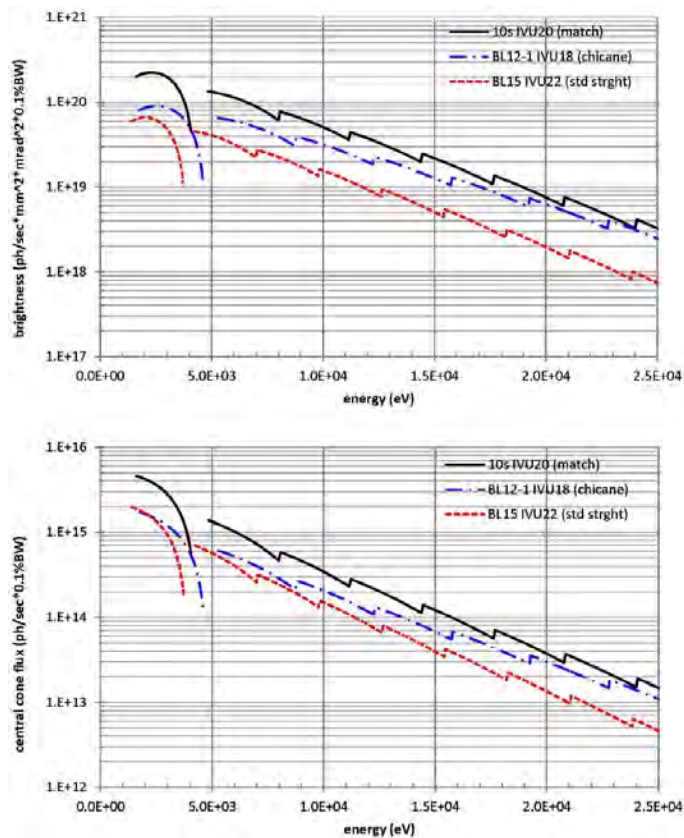
5.2 Beam Line Development and Technical Capabilities

SSRL has developed a plan that will keep our beam lines competitive and productive over the coming decade by using key scientific objectives to direct the integrated development of new and upgraded beam lines focused on innovative hutch equipment and state-of-the-art detectors. This beam line build-out plan will significantly increase both beam line capability and capacity. The light source currently features 24 operating beam lines (equipped with 32 stations); 8 independently operating bending magnet beam lines on 4 bending magnet sources and 16 independently operating insertion device beam lines on 9 insertion device sources.

SSRL has established an ambitious program of developing new undulator beam lines as the key goal of the development plan. The undulator-based BL15-2 (advanced hard X-ray

spectroscopy) has begun commissioning, and BL12-1 (micro-focus macromolecular crystallography) is under construction with anticipated completion in 2020. Another undulator beam line, BL17 (X-ray scattering) is in design with the undulator to be installed in 2018. Two new bending magnet beam lines, BL16-1/2, are in construction/commissioning (soft and hard X-ray metrology) sharing one port.

SPEAR3 can also accommodate two additional insertion device beam ports within the existing building footprints, offering impressive brightness and flux in the intermediate X-ray regime (see Figures). A further two bending magnet beam ports are also available for expansion into existing buildings. With more significant building modifications, further bending and insertion device beam ports could be instrumented.



Brightness of representative SSRL in-vacuum undulators (IVUs) assuming 6 nm-rad(H) by 10 pm-rad(V) emittance and 500 mA stored current. Only the odd harmonics are depicted. These IVUs are optimized for the 5-20 keV intermediate energy range. The energy gap at 4-5 keV can be eliminated by using a slightly longer period IVU or the undulator second harmonic.

Table 2: SSRL beam line sources and science disciplines served 2017. Six beam lines / stations serve multiple scientific disciplines so the count by discipline does not equal the total number of beam line stations (only major shared components included).

Circa 2017 (five BLs shared between disciplines)
24 BLs (13 wiggler, 1 IVU, 2 EPU, 8 bend)
12 Chemistry & Catalysis (8 ID & 4 bend)
11 Structural Molecular Biology (7 ID & 4 bend)
15 Material Science (10 ID & 5 bend)

5.2.1 New Undulator Beam Line Developments

The lower emittance of the SPEAR3 accelerator upgrade, coupled with incremental lattice improvements, will reduce the SPEAR3 emittance by almost three-fold relative to the SPEAR3 project design goal. These improvements in electron beam emittance make in-vacuum undulators (IVU) the optimal insertion device source for high brightness, intermediate energy X-ray beams at SSRL. The first IVU beam line on SPEAR3, BL12-2, was developed for macromolecular crystallography of small samples. This beam line has demonstrated the efficacy of teaming a high brightness, IVU source on SPEAR3 with stable, emittance-conserving optics and state-of-the-art experimental equipment and detectors. It has provided the foundation for the implementation of another three IVU beam lines (BL12-1, BL15-2, and BL17), which are in various stages of design, construction, and commissioning.

Taking advantage of the experience gained operating the BL12-2 small gap IVU, the new BL12-1 IVU (chicane IVU18, cf. Figure on previous page) has been designed for smaller magnet gap allowing the incorporation of 162 poles, compared to the 134 poles achieved with the same length BL12-2 IVU insertion device. This higher brightness source will be teamed with state-of-the-art Kirkpatrick-Baez (KB) mirrors, the well-established SSRL LN₂-cooled monochromator design and multilayer capabilities. The optical system will be designed to maximize source demagnification, yielding a focus optimally sized for crystallography of $\leq 5\mu\text{m}$ samples with beam stabilization servo systems to ensure stable illumination of small samples. The hutch equipment will include the evolving SSRL SMB macromolecular crystallography instrumentation for micro-crystal applications.

BL15-2 and BL17 each employ similar 22-mm period 174-pole IVUs located in standard length SPEAR3 straights. Spectroscopy BL15-2 features a LN₂-cooled double crystal monochromator with state-of-the-art KB focusing optics and beam stabilization servo systems. The XES, XRS, and RIXS instruments from BL6-2 will be relocated to BL15-2 in 2020. A Tangerine pulsed laser system for time domain studies has also been acquired for use at BL15-2.

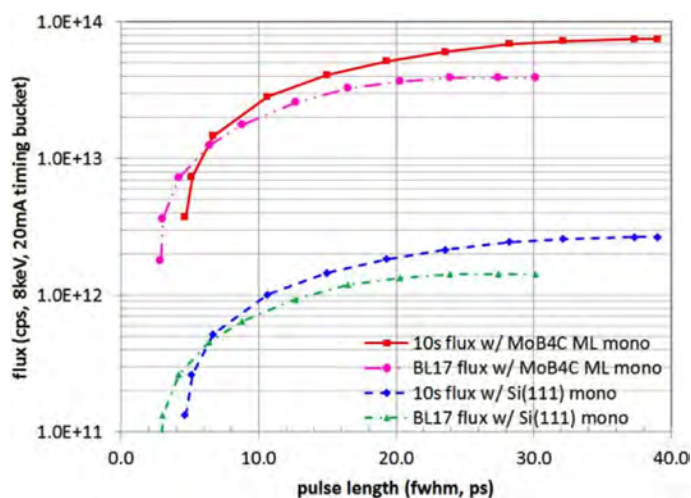
BL17, which will be optimized for materials scattering and diffraction applications, started development in late 2016 with the procurement of the IVU. This beam line will include optics similar to that of BL15-2 but augmented with a second vertical focusing

mirror. This mirror will create a demagnified vertical source image upstream of the KB optics. When the accelerator is operated with the differentiated orbits of pseudo single bunch mode per Section 5.1.2, a slit located at the focus of this mirror can be utilized to select photons exclusively from timing bunches for time domain studies. A similar vertical focusing mirror may be added to BL15-2 as a future upgrade.

All other existing SPEAR3 straight sections are IVU-capable and present additional opportunities for high brightness IVU beam lines similar to those described above.

5.2.2 Short Pulse Beam Line Studies

In support of the accelerator work on short pulsed sources discussed in 5.1.3 above, the performance of various beam line optics configurations are being studied to determine representative short pulse beam line performance and help inform design choices to optimally balance pulse duration, short pulse flux, and short pulse contrast. These studies involve differing combinations of crab cavity location, pseudo single bunch orbit perturbation, insertion device straight utilization, and photon optics configurations. Beam performance simulations have been performed for established beam line optical configurations, such as BL15-2, and unconstrained beam line layouts for new beam line locations.



Calculated 8 keV flux vs. photon pulse length for two different crab cavity and beam line locations: (a) crab cavity in 8s with the BL17 IVU22 in the 14s standard straight and (b) crab cavity in 1s with an IVU20 in the 10s matching straight. In each case both Si(111) and 2.5 nm MoB₄C multilayer mono-chromator simulation results are presented. The chirped timing bunch contains 20 mA in these studies.

Representative flux vs. pulse duration are depicted in the Figure below for two potential beam line locations: (a) BL17 on a standard straight and (b) an in vacuum undulator beam line on the unused 10s matching straight. Experiments that require 10⁻⁴ energy bandpass would utilize a Si(111) monochromator yielding almost 10¹² photons/sec into a 10 ps fwhm 8 keV pulse. Experiments that can tolerate the broader bandpass of a 2.5 nm MoB₄C multilayer monochromator would see $>2 \times 10^{13}$ photons/sec into a 10 ps fwhm 8 keV pulse. As evident in the Figure, the performance is a function of the phase advance from the crab cavity to the undulator source as well as the specifics of the undulator and beam line optics. Further assessment of various combinations of crab cavity location and target beam lines for time domain studies is needed to identify the optimal crab cavity location.

Calculated 8 keV flux vs. photon pulse length for two different crab cavity and beam line locations: (a) crab cavity in 8s with the BL17 IVU22 in the 14s standard straight and (b) crab cavity in 1s with an IVU20 in the 10s matching straight. In each case both Si(111) and 2.5 nm MoB₄C multilayer mono-chromator simulation results are presented. The chirped timing bunch contains 20 mA in these studies.

5.2.3 Revitalization of Existing Insertion Device and Bending Magnet Beam Lines

The goal of SSRL's long term beam line upgrade plan is to optimize each source and associated beam line optics for the intended application while leveling station demand. This plan is manifest in a continuing program of beam line optics upgrades, repurposing and addition of bending magnet stations for targeted applications, and the repurposing of wiggler stations as experimental programs move to the new IVU beam lines. Specific needs for APS users during the time when the facility will not be available during the APS-U shutdown are included in this SSRL planning process.

The materials scattering program will relocate from BL7-2 to BL17 following beam line commissioning. This end station vacancy permits migrating the current XAS imaging program from BL10-2 after modification of the BL7-2 beam line optics and end station. This move enables providing wiggler end station capability to the CoACCESS heterogeneous catalysis program, expanding the performance for these studies as compared to its current main location at bending magnet station BL2-2. Coupled with the addition of catalysis-required gas handling, novel *in-situ* reaction cells, and auxiliary sample characterization capabilities such as described in Section 4.2.1, will convert BL10-2 into a potent station for catalysis spectroscopy studies.

As noted in Section 5.2.1, the suite of advanced spectroscopy instruments from BL6-2b will be relocated to IVU BL15-2. The fraction of time on BL6-2 liberated by this change will be reappor- tioned to the TXM located in BL6-2c as well as tender XES and imaging programs in BL6-2a.

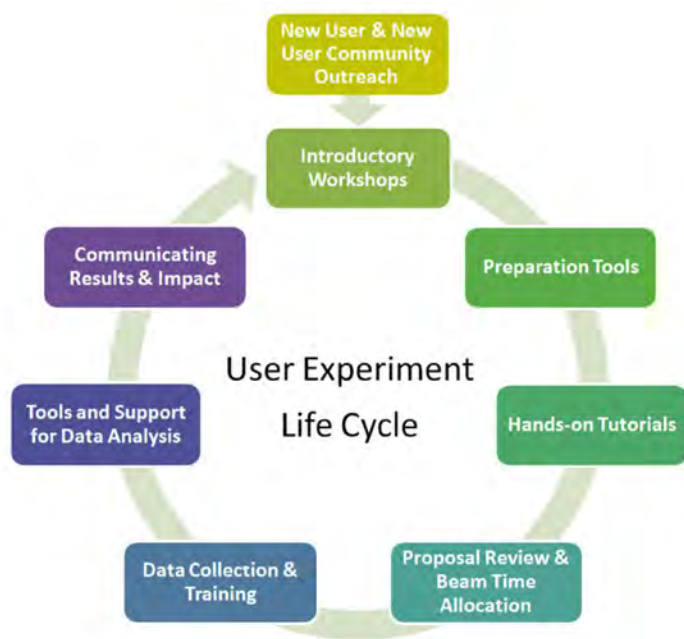
SSRL has several open bending magnet ports. One of these, designated BL16, is currently being developed into two stations for soft and hard X-ray metrology as part of a NNSA-funded effort to replace capabilities lost with the closure of NSLS. SSRL plans to develop a bending magnet station for X-ray tomography as discussed in Section 4.1.1. This could involve instrumenting a new bending magnet port or repurposing another bending magnet beam line after scientific review.

Outreach, User Support and Education



6 Outreach, User Support and Education

Building on SSRL's well-established roots within the synchrotron research community, a strong connection to Stanford University and close connections to technological developments in Silicon Valley, SSRL supports the research life cycle from beginning to end to ensure that users get the best science from their time at the facility. The SSRL approach to supporting the user community is illustrated in the User Experiment Life Cycle scheme to the right and described below.



Reaching Out to New and Diverse Scientific Communities

SSRL has remained at the forefront over the facility's ~40-year history by continually enhancing the synchrotron source, developing new methods, beam lines and instrumentation, and bringing in new ideas from users, staff and faculty. The facility has successfully fostered several new scientific communities in areas including structural molecular biology, hard X-ray scattering, photoemission spectroscopy, imaging, environmental science, and catalysis, while encouraging networking with established and emerging scientific research centers. Of the approximately 1,700 scientists who annually participate in experiments at SSRL, over 40% are first-time users. With our strategic plan that includes lowering the emittance, running at one of the highest-current levels on any mid-energy source world-wide, expanding in capability and capacity through developing new accelerator modes for timing studies and new or repurposed beam lines for emerging science, we have as goal to continue the growth and the support of existing and new communities.

SSRL staff scientists reach out to new users and communities through their participation in scientific conferences related to key scientific topics, organizations and educational programs. In the coming years, SSRL will continue to educate future generations of scientists and will increase the number of SSRL facility tours for local and visiting scientists who have an interest in conducting synchrotron research.

To reach industrial researchers, SSRL networks and collaborates with both local start-ups and large multi-national companies to pursue opportunities in energy research, biotechnology, and information technology (see Sidebar).

Finally, SSRL staff members are increasing their coordination with other light source facilities to create shared outreach materials, including Lightsources.org, a website that provides light source information to the academic, scientific, and industrial communities.

Industry Research – Engagement and Facilitation

The participation of researchers from industry is an essential part of a vibrant and innovative research program at SSRL. Researchers from industry bring a valuable practical perspective, with the potential for new ideas, to the SSRL research community. Researchers from industry typically have a problem that needs a solution, and as such they are typically interested in answers, and not necessarily the data per se. SSRL believes that there is a significant untapped potential for fostering innovation and economic competitiveness, and bringing new ideas and the resulting scientific impact to SSRL.

The strategy of SSRL's planned approach is based largely on the recommendations and findings of several recent reports and workshops, including the 2010 BESAC report, "Science and technology – Strengthening the link between Basic Research and Industry", and the 2014 "Workshop on Industry Research at NSLS-II". The operational structure of SSRL, based around collaboration among the scientific staff, ideally positions SSRL to ensure the scientific success of the industry engagement. SSRL will develop a plan for enhanced industry involvement focused on the concept of consortia and academic/lab/industry partnerships. It will, for example, engage faculty members, who will act as intermediaries between SSRL and the particular industry, being involved in the particular scientific problem that the industry is trying to solve. SSRL will also pursue the creation of consortia, selecting one relevant industry group based on a particular theme, e.g. catalysis, and invite appropriate industry representatives to join this consortium. Eventually, this might lead to an Industrial Affiliates program, analogous that of many academic institutions.

Providing Introductory Workshops to Potential New Facility Users

After reaching out to new user communities, SSRL staff members follow up with introductory workshops in selected areas of science and techniques. A large fraction of these workshops are held during the SSRL / LCLS Annual Users' Meeting and Workshops that attract a large number of participants. These workshops are organized by staff scientists in collaboration with the SSRL Users Organization and typically include national and international scientists in the targeted area of science and/or methodology.

Providing Tools for Experiment Design

SSRL provides multiple online resources to help users best design their experiments, and plans to increase these resources in the coming years. The Structural Molecular Biology Division at SSRL has received glowing feedback from users for its remote access systems that integrate an interactive interface with both real and simulated beam lines. SSRL is using lessons learned from the implementation of macromolecular remote user program to extend rapid through-put and other web-based visualization and simulation platforms across facility beam lines.

Reviewing Proposals and Allocating Beam Time

To ensure the facilities are leveraged for the most fruitful and important research, requests for beam time are peer reviewed on the basis of scientific merit and impact. Proposals are directed to the SSRL facility as a science proposal, and not for a specific beam line; accordingly a proposal can include several techniques, energy ranges and beam lines. While most proposals are valid for 1-2 years, in order to enable timely and current research, SSRL also provides several rapid-access proposal mechanisms. SSRL also has a mechanism through a letter of intent to provide a short amount of beam time for users to test the feasibility of new experiments.

Running Hands-on Tutorials

To ensure effective usage of beam time, meaningful data, and successful publications, new and returning researchers are invited to take part in SSRL's many tutorial sessions. These include hands-on training at summer schools, short courses and workshops that focus on synchrotron techniques, and one-on-one tutorials.

Assisting with Experimental Set Up and Data Collection

SSRL has nurtured a culture of pride among its staff in providing expert service and support. Facility staff members provide the following resources to help scientists make the most of their beam time:

- Specialized, state-of-the-art beam lines, instrumentation, and capabilities
- Technical support from experienced facility scientists, engineers, and support staff

- Ancillary laboratory equipment including wet laboratories, glove boxes, and anaerobic chambers
- Assistance with sample preparation
- Remote access where applicable, allowing users to collect and process data remotely from their home institution, using a remote desktop application and enabled by beam line automation
- User facilities including an on-site guest house, exercise facility, and a central check-in and orientation location for all SLAC users
- Multi-lingual support staff and safety training courses

In the next five years, SSRL will seek to increase staff and one-on-one training to help researchers optimize beam time and subsequent analysis.

Providing Tools and Support for Data Analysis

To optimize the productivity of users, the SSRL scientific staff has developed and/or imported from the international community several data analysis software programs that are made available for users during the experiments, and for download to home institutions in general. Training on how to use the software is provided during experiments and as topics during workshops and summer schools. The availability of analysis software at the beam line enables rapid analysis to drive the strategy of the experiment. A new tool will include the development and implementation of an XAS data base of known compounds (see Sidebar).

Assisting with Communicating Results

More than 500 papers are published annually as a result of research at SSRL, totaling over 14,000 publications since the facility began in 1974. SSRL staff members make a concerted effort to communicate these results with the general public, the media, the local community, and other scientists through public lectures, press releases, science articles, and brochures. SSRL also produces a monthly electronic newsletter that disseminates scientific results and new facility capabilities to the scientific community.

Training the Future Science Generation

Throughout the research lifecycle, SSRL actively participates in building a pipeline of future scientists and engineers. In addition to the workshops and tutorials described above, more than half of the experiments at SSRL are conducted by undergraduate students, graduate students, or postdoctoral scholars. The hands-on experience helps students learn to formulate new scientific ideas, prepare successful research proposals, plan and conduct experiments, and analyze and interpret data. It also clearly shows the next generation the potential of synchrotron research to enable faster, more novel, and more precise scientific discoveries.

XAS Database of Known Compounds

Efficiency of operations is important to both increasing the number of users and allowing sufficient time for more demanding experiments. One facet of this is ensuring that data are only collected on new samples and not on compounds for which data have previously been measured (such as basic model complexes). SSRL will work towards developing an internal database of XAS data for known compounds, contributed by the scientific user community. The data base will use the file format developed through the ad-hoc international working group, the XAS Data Interchange Group, formed under the auspices of the International XAFS Society and the XAFS Commission of the International Union of Crystallography.

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