Stanford Synchrotron Radiation Lightsource

Strategic Plan:
2021-2025

Meeting the Scientific Challenges of the Future

March 2021

SLAC NATIONAL ACCELERATOR LABORATORY

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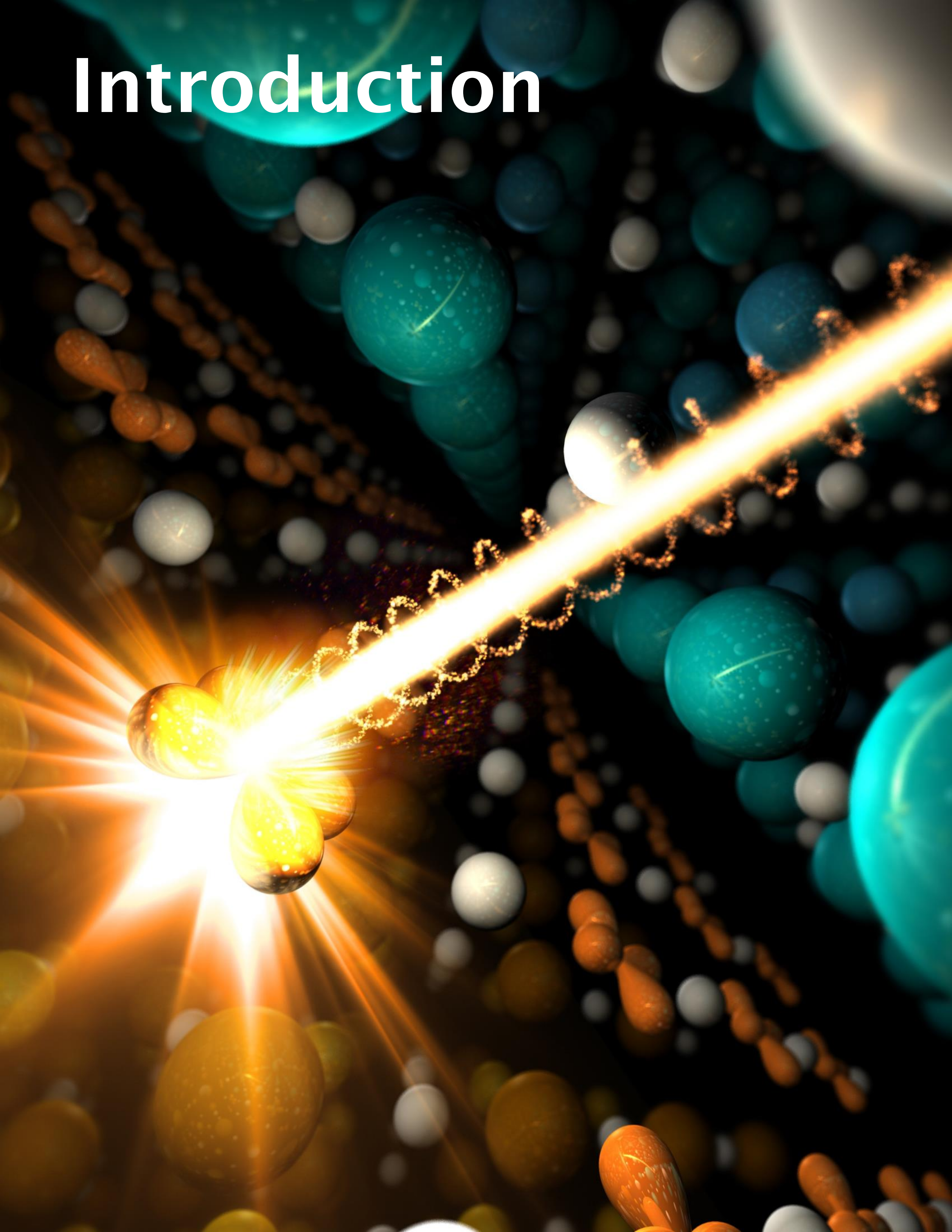
Introduction



1 Executive Summary

The next decade will see a transformation in the ways that storage ring-based synchrotron radiation facilities impact user science. The construction of new synchrotrons, based on multi-bend achromat designs, will lead to significant increases in transverse X-ray coherence, enabling significant advances in X-ray imaging. At the same time, breakthroughs in automation of instrumentation, data collection and analysis have the potential to boost the productivity of light sources and broaden their impact, as users increasingly access their capabilities remotely. Finally, light sources will increasingly transition from being “photon factories” to centers of innovation that are critical to discovery and use-inspired research performed by their users. This plan sets 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 connections 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 for time-resolved experiments 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) 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. 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 (micro-second 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).
- Continue to build and expand capabilities in cryo-electron microscopy and cryo-electron tomography for biological studies that range from proteins and nucleic acids to very large biological assemblies and complexes, at resolutions that range from nanometer to the atomic level.

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 areas.
- 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.

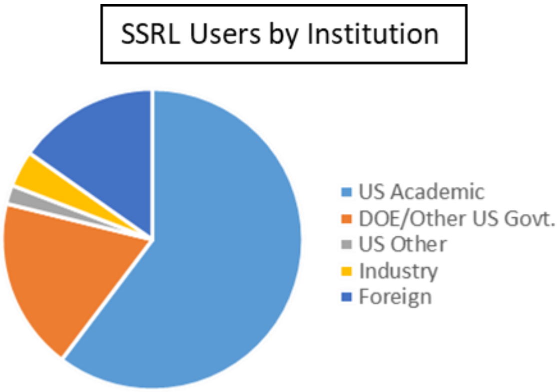
The sections below provide background, overview, and a more detailed current and future perspective of SSRL's scientific programs in the main areas of materials energy science, chemistry, catalysis, interfacial and environmental science, structural molecular biology science, accelerator science, and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

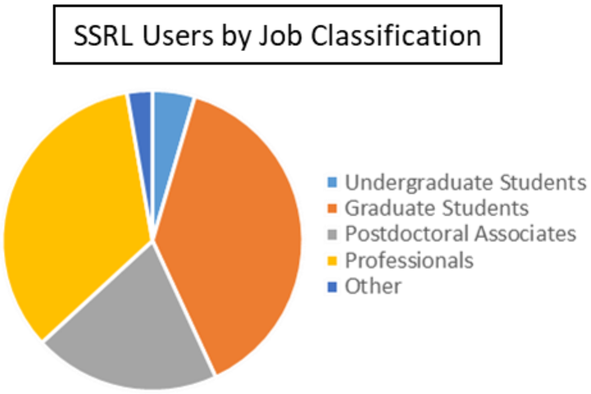
2 The Role of Synchrotron Radiation Facilities in U.S. R&D

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, six Nobel prizes have been awarded to scientists who used synchrotron light sources for their research.

Today, there are >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 scientists 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.





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 operates 33 scattering, diffraction, spectroscopy, and imaging experimental stations, with expansion capacity. It is a highly productive scientific user facility with high user satisfaction, generating more than 600 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 at SLAC.

SSRL's strategy for expanding our footprint in ultrafast science has two 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.
- Developing a longer-term SPEAR3 accelerator enhancement that will deliver 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. As the administrative home of the Stanford-SLAC CryoEM facilities, the SSRL Directorate is committed to providing world-class bioimaging capabilities using both X-ray and EM methods, while also broadening the range of applications of cryoEM to critical DOE-relevant priorities in materials science and chemistry.

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 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 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 and Devices, 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 X-ray absorption spectroscopy with focus on chemical catalysis
- 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 photovoltaics, 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 CO_2 reduction, H_2O oxidation, N_2 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 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 energy 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 photo-catalysis 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) 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.
- Investigate the structural origins of decoherence in quantum devices.

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:

- 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 hydro- logical 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 high-energy resolution fluorescence detection (HERFD) yield XAS imaging capabilities for actinide research related to nuclear materials, and geo-/ biogeochemical and environmental projects.
- Develop capabilities to image redox-sensitive element speciation at the micro-scale across large spatial domains in soil cores to detect and quantify the importance of microsites for modeling their contribution to the global carbon cycle.
- Develop multi-modal, multi-technique methods for integration of non-X-ray techniques to enhance the understanding of chemical processes.

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)

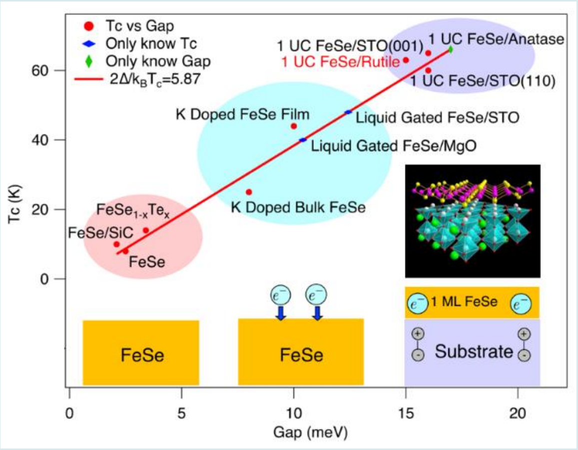
In the early 1990s, for example, SSRL identified understanding the function of RNA Polymerase II as a high-risk, high-reward scientific opportunity. Through strong collaboration with Roger Kornberg, Professor of Structural Biology at Stanford University, this scientific challenge led to many important 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. These facilities were again instrumental in enabling the structural work that formed part of the 2018 Nobel Prize in Chemistry to Professor Frances Arnold, Caltech, for her discoveries on the directed evolution of enzymes.

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.

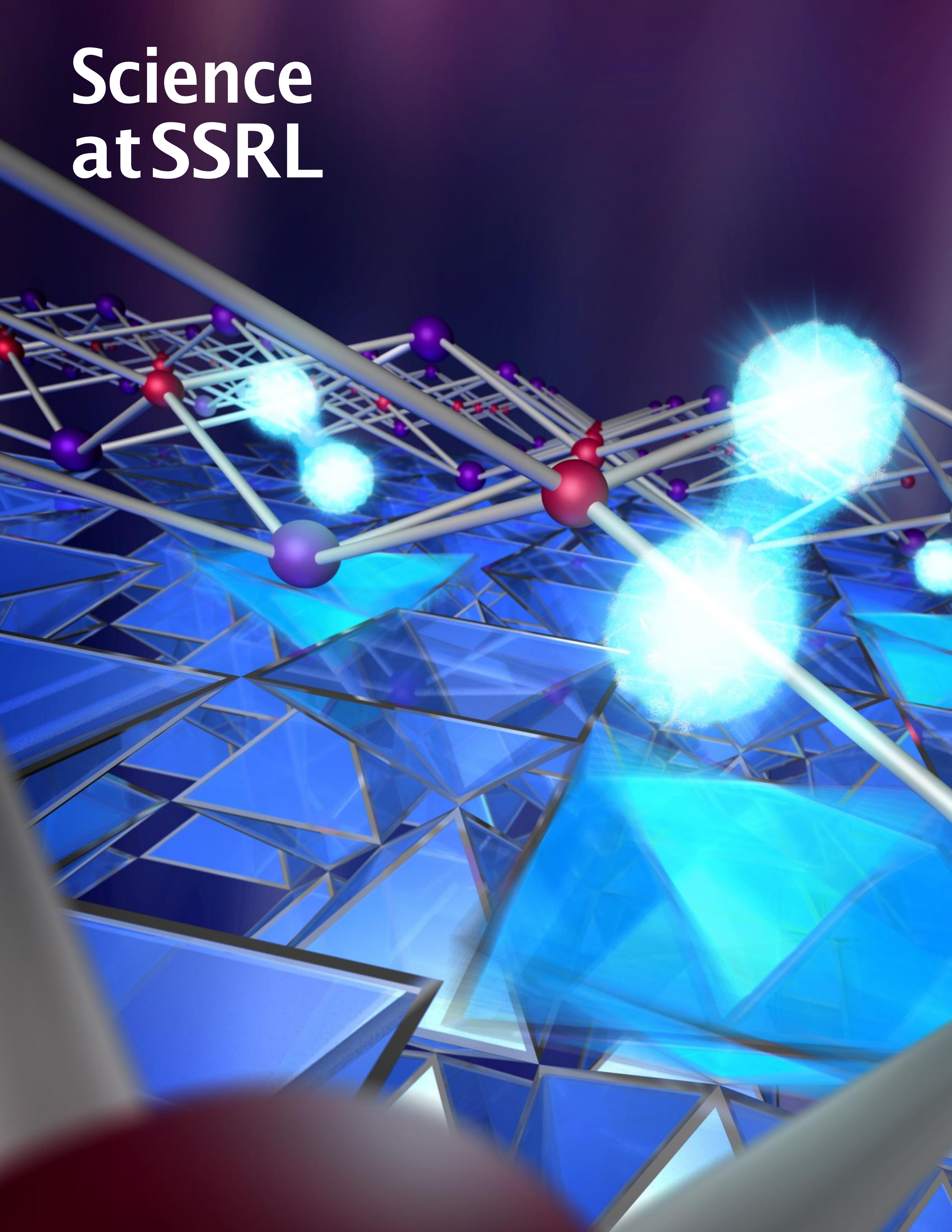
Core Scientific Program Development: Long-Term Sustained Local Collaborations

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-based ARPES facility in the world. The core program has motivated the advancement of ARPES program at SSRL, including the development of a new state-of-the-art ARPES end station equipped with sophisticated MBE thin film growth capabilities, attracted more general user teams, 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).



The discovery of iron-based superconductors in 2008 has revitalized research in the high-T_C community. More recently, the record high superconducting transition temperature discovered in monolayer FeSe thin film grown on $SrTiO_3$ has attracted great attention in the field. Understanding the mechanism of the significant T_C enhancement in the monolayer thin film compared to the bulk FeSe offers potentially a new pathway for future engineering of higher T_C superconductors. A pioneering ARPES study carried out on beam line 5-4 using ex-situ grown FeSe thin film unveiled the unique fingerprint of a strong coupling between the electrons in FeSe film and phonons in the underlying $SrTiO_3$ substrate, manifests as replica bands in the electronic structure. To better understand the cross-interface electron-phonon coupling and its relation to superconductivity, systematic in-situ ARPES measurements of monolayer/multilayer FeSe films grown on various titanate substrates have been performed on the new beam line 5-2, enabled by the integrated MBE thin film growth capabilities. The ubiquitous replica bands observed in all monolayer films clearly demonstrates that the interfacial electron-phonon coupling is a generic feature of the FeSe/titanate system and sets constraints on the ingredients necessary for the enhancement in superconductivity. This research project highlights importance of the strategic investment in the cutting edge instrumentations and long-term local collaborations.

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 Synthetic Control Across Length-Scales for Advancing Rechargeables (SCALAR), the National Renewable Energy Laboratory, and Stanford's Institutes (like the Precourt Institute for Energy) and Schools. 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 EERE Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) consortium, and the EERE HydroGEN consortia in the Energy Materials Network (EMN). We will align our beam line, 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 dedicated end station for X-ray tomography on a pre-existing wiggler source.

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 and imaging are unparalleled methods for tackling this critical area of research. SSRL is commissioning 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.

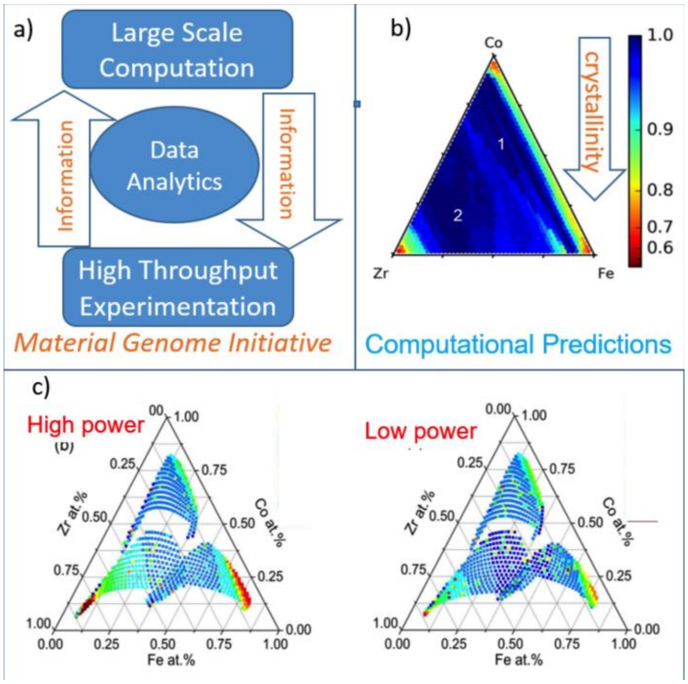
Cost-effective solutions to the materials challenges facing us today necessitate accelerating the discovery of new functional materials. The development of high throughput characterization methods will play a key role in achieving this objective. The present Edisonian paradigm based on serial experimentation takes decades from initiation of a new search for a material to marketplace deployment of a device. SSRL is coupling high-throughput experiments with data analytics, and active control algorithms with the objective of pioneering data driven discovery paradigms at user facilities. 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 an order of magnitude.

SSRL has built high-throughput X-ray scattering capabilities distributed across the suite of beam lines. These capabilities increase the number of experiments which can be performed by over an order of magnitude. To compliment this, SSRL is also developing on-the-fly data analytics that incorporates recent advances emerging from unsupervised pattern recognition, machine learning and artificial intelligence to interpret data in real time, and use this interpretation to guide experimental decision making by both humans and artificial intelligences. One example, highlighted below, uses machine learning to predict the structure of ternary alloys, and then uses high throughput X-ray diffraction to validate these predictions for up to 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 also enhance picosecond time-resolved sciences efforts.

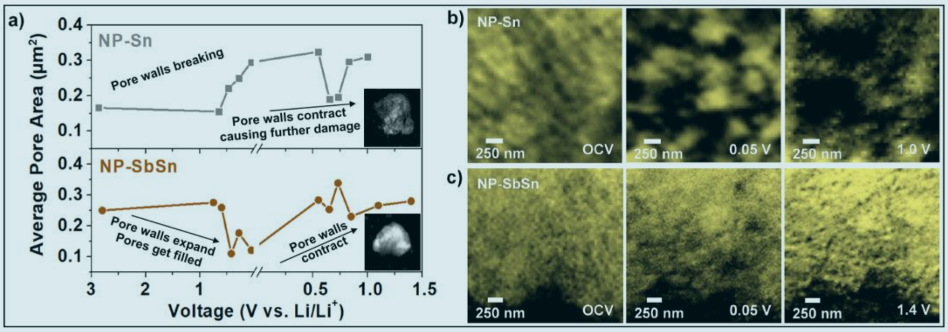
- **X-ray Tomography:** The wide range of important length scales in energy materials drives a need for a high-speed, high-energy X-ray tomography capability to compliment the higher resolution of the TXM. A new dedicated end station is planned. *Operando* studies of battery electrodes during cycling and of catalysts during deactivation will develop mechanistic understanding into degradation pathways.



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 machining 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.

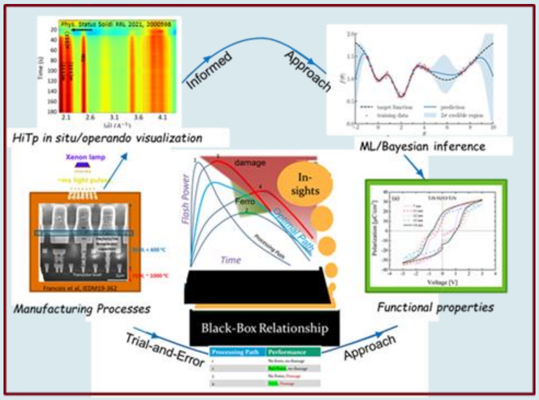


In situ X-ray characterization of battery materials during cycling under realistic conditions provides a window into the energy storage mechanisms and failure pathways. In-situ high resolution transmission X-ray microscopy is used to observe morphological changes occurring throughout a battery. This allows researchers to identify specific, irreversible morphology changes such as particle fracturing 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. For example, nanoporosity is often used to mitigate particle pulverization with Li insertion/removal in high capacity anode materials such as Si and Sn. As illustrated in the graphics above, nanoporosity alone does not prevent irreversible morphology changes to Sn nanoporous (NP) structure; however, a nanoporous SbSn alloy is stable under cycling since each metal alloys with Li at a different potential (ACS Nano ***2020****, 14, 11, 14820–14830).*

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.

HiTp, ML-guided, *In-situ* Visualization for Transformative Smart Manufacturing

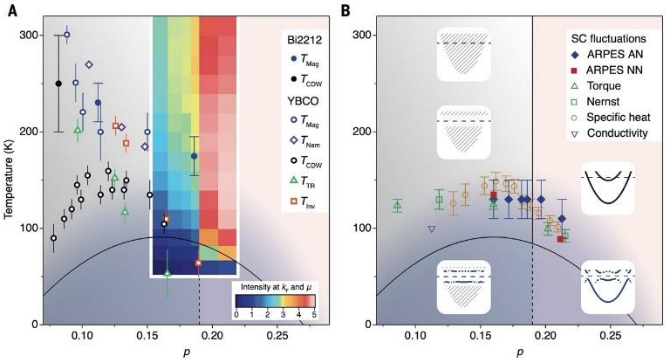
Continued advances in high performance computing, as embodied by Moore's law, will require power performance and cost scaling to be achieved through ultra-dense 3D integration of memory and logic, novel nanoscale device concepts, and the introduction of disruptive new materials into semiconductor manufacturing. This produces a cascade of manufacturing problems involving chemical compatibility, thermal budget constraints, and real-time detection and mitigation of manufacturing defects. Traditional approaches to manufacturing will be unable to meet the stringent specifications for manufacturing yield for future codesigned 3D heterogenous systems. High-throughput (HiTp) intelligent sensors and manufacturing tools, providing real-time information at the speed of manufacturing, will be keys to a transformative smart manufacturing strategy.



An example of this optimization paradigm being pursued at SSRL is HiTp, ML-guided, *in-situ* visualization of structural phase transitions using synchrotron radiation during highly non-equilibrium flash annealing to manufacture high performance ferroelectric HfO_2-ZrO_2 (HZO) alloys, integrated into field effect transistors or on-chip capacitors without thermally damaging underlying layers manufactured under typical back-end-of-line conditions. Expansion of this transformative manufacturing paradigm to include other areas in microelectronics will be accomplished through interconnected efforts: (1) the creation of collaborations between SLAC and microelectronics research at Stanford, (2) the development of HiTp *in-situ* visualization tools using synchrotron radiation for metrology of microelectronic materials and devices, including *operando* characterization, and (3) the development of ML/AI-guided formalisms capable of providing active learning and control of manufacturing processes. This synergy between HiTp experiment and ML/AI guided feedback will enable the full exploitation of this transformative smart manufacturing strategy that will enable tightly interwoven, codesigned assemblies of 3D heterogeneous circuitry comprised of trillions of nanoscale components.

4.1.2 Strongly Correlated Electron Materials – Quantum Materials

The emergent phenomena in quantum materials present exciting opportunities to understand 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 could 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. Correlation effect, spin-orbit interaction, and interplay between spin, charge and lattice give rise to a variety of extreme and remarkable emerging properties – high temperature superconductivity, topological phases of matter, interface driven superconductivity, to name a few examples.



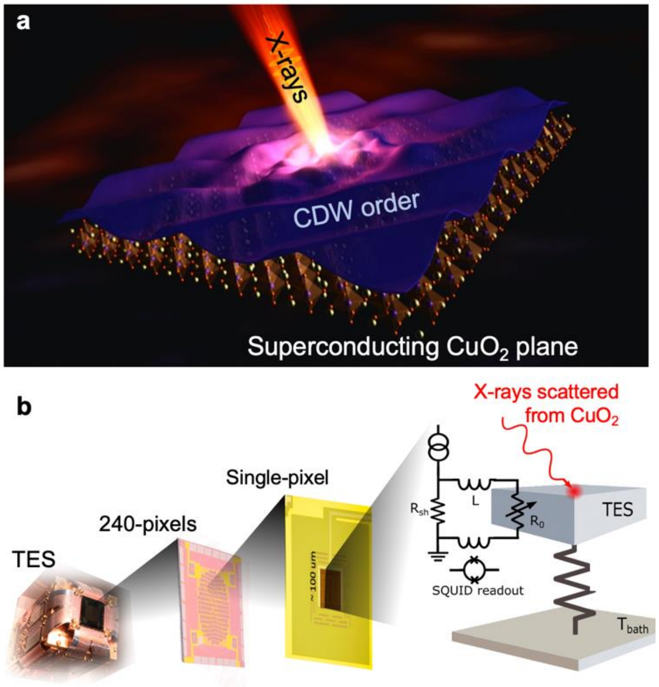
*Phase diagram of Bi2212. (A) The color plot outlined in white shows the zero-energy spectral intensity at the bonding band Fermi momentum along the Brillouin zone boundary. Also plotted are the transition temperatures of various broken symmetries in Bi2212 and $YBa_2Cu_3O_{6+\delta}$. (B) The temperature scale of superconducting fluctuations in Bi2212, from this work (blue diamonds) and several previous studies. Science **2019** 366, 1099.*

As better controlled model systems become available, a sophisticated understanding of 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 enhanced 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 quantum materials, with the enhancements that the combination of the existing BL5-4 and the new BL5-2 will provide.
- **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 using Molecular Beam Expitaxy (MBE), significantly broadening the scope of materials and structures to be investigated with the ARPES technique. 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 hetero-structures 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. The recent success in the in-situ ARPES study of single-unit-cell FeSe grown on $SrTiO_3$ by MBE has demonstrated great potential in this synergetic investment by SSRL and SIMES. 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 dedicated for high-resolution ARPES (BL5-2; see above). This addition completely modernizes and greatly expands the capabilities of the preceding branch-line with full polarization control, extended photon energy range, and a significant improvement in flux and beam spot size. In conjunction with the recent spectrometer upgrade that enables a complete mapping of the electronic structure without rotating the sample manipulator, the micro-focusing capability has transformed the new PGM branch line 5-2 into a top competitive micro-ARPES facility, which is further complemented by the sophisticated MBE thin film growth system and the ultra-high resolution capability of the NIM branch line 5-4.



Resonant soft X-ray scattering (RSXS) provides a sensitive probe of charge and spin order in strongly correlated electron systems. This top panel illustrates shows stripes of higher and lower electron density – "CDW stripes" – within a copper-based superconducting material. Experiments using X-ray directly observed how those stripes fluctuate when hit with a pulse of light, a critical step towards understanding how these CDW stripes interact with high-temperature superconductivity. (Nature Commun. ***2019*** *10, 3269). The bottom panel shows the recently developed transition-edge-sensor detector for the RSXS capability at BL13-3. Taking advantage of single photon sensitivity as well as the energy-discriminating detection, the intrinsic fluorescence background can be removed, enabling the detection of very weak signal such as the CDW order in high-T_C cuprates.*

- **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 $LaAlO_3/SrTiO_3$, 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.

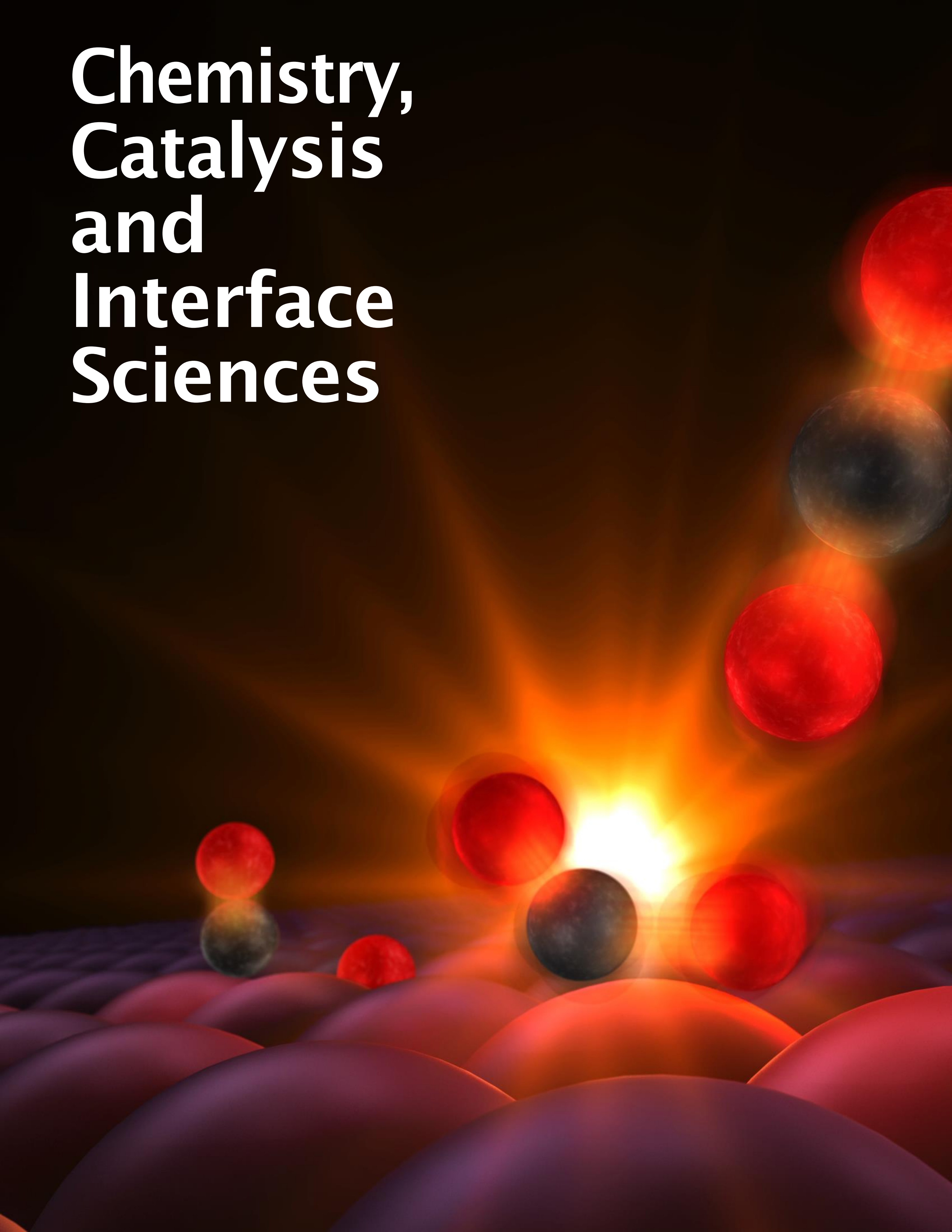
- **Resonant Soft X-ray Scattering (RSXS):** Recently, the effect of spin-orbit coupling in strongly correlated materials has spurred significant interest in the physics and materials science communities since the combination of strong spin-orbit-coupling and electron correlation lead to new emergent 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. A good example is the study of charge-density-wave (CDW) phenomena in high-T_C cuprates, for which RSXS has played an important role. SSRL has developed an RSXS endstation at BL13-3 and opened the facility for general user operations. More recently, the RSXS endstation has been further advanced by the integration of a superconducting transition edge sensor detector with eV energy resolution. The energy resolving power of this detector enables the suppression of the inelastic X-ray scattering signal as well as the fluorescence background, thereby significantly enhancing the signal-to-noise of the RSXS signal, opening up new science.

4.1.3 Summary of New 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, with diffractometer and simultaneous SAXS/WAXS, RSXS	
Energy: Photon conversion and batteries	Micro-focus spectroscopy	Advanced spectroscopy (x-ray inelastic scattering)	Undulator beam line, with diffractometer and simultaneous SAXS/WAXS	Upgraded TXM; X-ray tomography
***In-situ* growth and synthesis**		Advanced spectroscopy (x-ray inelastic scattering)	Undulator beam line, with diffractometer and simultaneous SAXS/WAXS; RSXS	Upgraded TXM; X-ray tomography
Picosecond time domain			Undulator beamline, with diffractometer and simultaneous SAXS/WAXS	

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 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 continue to 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 BES Chemical Sciences funded Co-ACCESS program provides research capabilities to external collaborators including a full suite of *operando* catalytic reactors with the necessary infrastructure at the beam line, off-line catalyst treatments, experimental testing and prototyping/testing of experimental cells, in addition to assistance with data collection, analysis and interpretation.

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, nano-, biological, 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, 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 Liquid Sunlight Alliance (LiSA), 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 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 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 based on ongoing R&D), 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 continue to 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 became operational in 2020.

- ***In-situ* Tender X-ray Spectroscopy:** The tender X-ray energy region (2-5 keV) includes K-edges of the 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 has been 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* catalysts 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.

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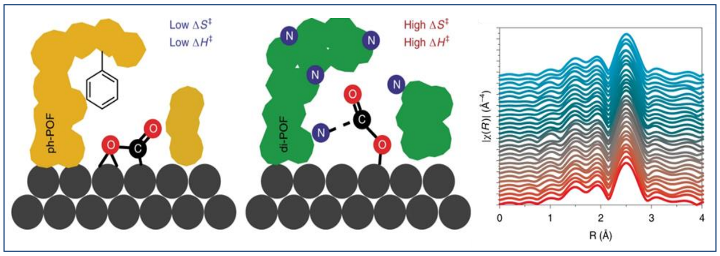
- **2D and 3D Imaging:** The relocation of the advanced spectroscopy program to its own beam line (BL15-2), previously sharing time with SSRL's hard X-ray TXM facility on BL6-2, will allow renewed emphasis and continued development of hard X-ray TXM methods at the 30-50 nm length scale, including tomography, and with XANES capabilities, for *in-situ* catalysis studies. Planned developments include implementation of complementary *in-situ* characterization techniques.



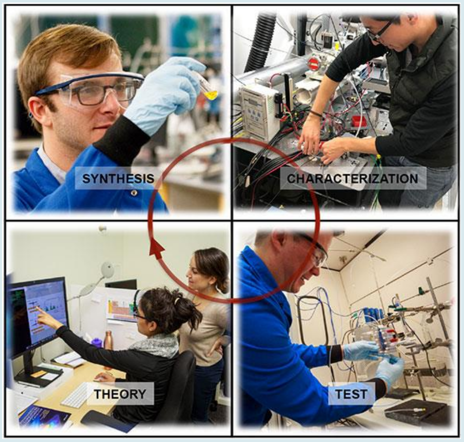
*A novel approach to synthesis of polymer-nanocrystal hybrid catalysts was shown to stabilize specific transition states and thus control the transport of species to and from the active sites. In this study, polymer layers provided a confined environment, impacting the catalytic performance, resulting in observed reaction oscillations specific to POF layer coverage and chemical composition. This was measured in situ with the operando EXAFS data at the Pt L3-edge displayed as a function of time (~2 min/scan). Nature Catalysis **2019**, 2, 852.*

- **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 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 under-standing 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 thermo-chemical, 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 successful staging of soft X-ray experiments at BL8-2 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 is working closely with the SLAC TID group led by Prof. K. Irwin on TES detector R&D and experimental use at BL10-1.
- **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, 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.

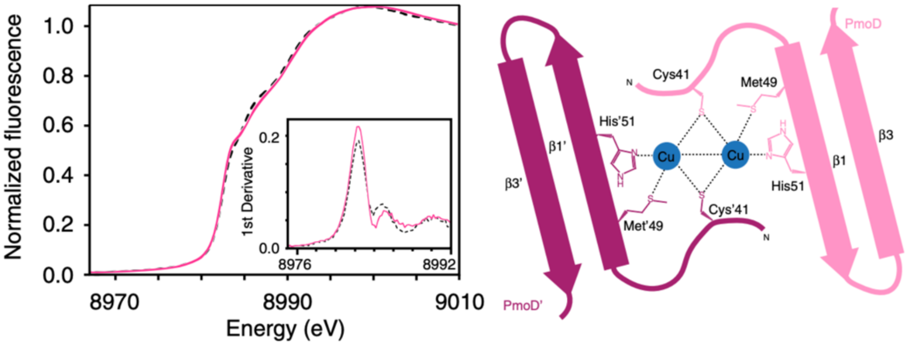
4.2.2 Enzyme and Bio-Inspired Catalysts for Sustainable Energy

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 have 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⍰ 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. In a new collaboration within the BOTTLE consortium, SSRL will help determine the role of bio-inspired and biological catalysts in plastics degradation and upcycling. 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.



Methane-oxidizing microbes catalyze the oxidation of the greenhouse gas methane using the copper-dependent enzyme particulate methane monooxygenase (pMMO). Isolated pMMO exhibits lower activity than whole cells, however, suggesting that additional components may be required. A pMMO homolog, ammonia monooxygenase, converts ammonia to hydroxylamine in ammonia-oxidizing bacteria which produce another potent greenhouse gas, nitrous oxide. Users at SSRL have applied Cu K-edge X-ray absorption spectroscopy to discover a copper dimer center with a previously unobserved ligand set. These findings identify a copper-binding protein that may represent a missing link in the function of enzymes critical to the global carbon and nitrogen cycles. (Nature Comm. ***2018****, 9, 4276.*

- **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.

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 CO_2 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 under- stand the molecular-scale basis of these processes, one of which 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:

- **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 Imaging:** Soils represent the largest dynamic pool of carbon on Earth and, hence, soil processes are major players in the global carbon cycle and the controls on climate change. Anaerobic processes in soils and sediments are essential to nutrient cycling and also play important roles in greenhouse gas emissions and contaminant mobilization. Anaerobic processes are conventionally assumed to be hosted within wetlands or deep/wet soil environments where oxygen availability is substantially limited, however nominally oxic subsurface sediments often exhibit aggregates, lenses, or layers that appear to have anaerobic properties, so called ‘hot spots’ of ‘anaerobic microsites’. SSRL will develop the ability to image redox-sensitive element speciation at the micro-scale across large spatial domains in soil cores to detect and quantify the importance of microsites across a wide variety of soil environments, enabling better modeling of the contribution of microsites to the global carbon cycle.

- **HERFD Spectroscopy Imaging:** Trafficked nuclear materials pose a world-wide threat, and it is of high importance to determine the origin of illicit materials. Subtle molecular clues can indicate specific nuclear fuel cycle chemical processes during fabrication and types of further chemical processing. SSRL will develop, high-energy resolution fluorescence detection (HERFD) XAS imaging spectrometer. This will enable quantification of actinide redox and chemical states at the L and M edges, at the micro-scale. The technique allows for the additional characterization of micro-scale variability. The instrument will also be used in geo/biogeochemistry user applications.

- **Multi-technique Approaches**. The X-ray imaging program at SSRL is developing the integration of simultaneous UV-visible spectroscopic measurements with XAS and imaging experiments, as well as the collection of spatially resolved FTIR microscopy data sets. The integration of this expanded photon range allows a holistic glimpse into the chemical nature of transition metals and actinides, biologically important elements (Ca, S, P) as well as the vibrational nature of organic materials.

4.2.4 Summary of New Capabilities

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

Science themes	**Experimental technique / beam line / instrument**				
	Hard x-ray spectroscopy		**Tender/soft x-ray spectroscopy**		**Imaging**
Catalysts for energy production	*In-situ* & 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	*In-situ* hard and tender x-ray XAS imaging
Enzyme and bio-inspired catalysis		Advanced spectroscopy undulator (XES, XRS, RIXS); LCLS	Tender x-ray XES and RIXS		*In-situ* hard and tender x-ray XAS imaging
Molecular biogeo-, environmental and interfacial	*In-situ*, high throughput, and time-resolved XAS (wiggler)	Advanced spectroscopy undulator (XES, XRS, RIXS)	Tender x-ray XES		*In-situ* 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, particularly in the face of emerging new and rapidly evolving viral pathogens and environmental challenges.

Knowledge in this field has relevance to solving grand challenge problems related to energy, environmental sustainability and medicine. 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 enabling scientists to rapidly obtain and utilize biomolecular and biomimetic molecular structural information on the micron-to-atomic scale to understand function (and malfunction) of biological processes. 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 bioenergy, chemical/biogeochemical, environmental and biomedical 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). There is further synergy with the LCLS X-ray free electron laser and the SLAC-Stanford cryo-electron microscopy (cryoEM) and cryo-electron tomography (cryoET) facilities which are co-located on the SLAC site. Collectively, these techniques provide a remarkably rich and broad window on structure and function across a wide 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 continue to pursue emerging scientific areas related to bioenergy, environmental sustainability, biogeochemistry, biosecurity and disease interventions, where X-ray techniques provide unique tools for studies in the length scale ranges from molecular-to-cellular and beyond. The recent developments in CryoEM and CryoET programs at SSRL/SLAC are providing unique opportunities for joint scientific and technological developments, which will be vigorously pursued. The SMB program will continue to strengthen partnerships with Stanford University Institutes, including ChEM-H, Bio-X, and IMA, expanding on existing joint programs for MC beam line development and science; with industry on high throughput drug discovery developments; and with private institutions on emerging scientific topics.

The SMB program will continue to engage with other user facilities and coordinate as appropriate, to pursue 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. This includes targeted outreach to potential new SMB users at the BER Bioenergy Research Centers and DOE-BER funded researchers at the national labs and universities.

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 particularly enabled by the new world-wide competitive, microbeam undulator beam line (BL12-1) commissioned in 2021, 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.

More specifically, aims are to enable: *i)* structural studies of increasingly large and challenging macromolecular assemblies and machinery, crucial to advancing the understanding of complex biochemical processes, *ii)* structural studies of membrane proteins and complexes, which are of major biological importance, but exceptionally challenging to crystallize, and *iii)* accessing biodynamics through room temperature and freeze-quench approaches.

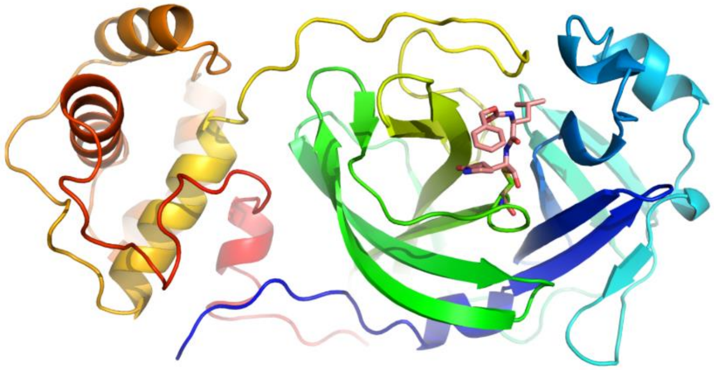
With advances in crystallization, sample delivery, instrumentation, methodology and X-ray capabilities, a significant number of new and challenging structures can be determined by SSRL's large and diverse user community, providing details essential for understanding a range of cellular functions regulated by these complex systems and leading to practical benefits such as the structure-based design of new and improved drugs. Another area will be in structural virology, gaining fundamental insights that will enable the development of vaccines and other therapeutics, including for SARS-CoV-2, and the mutant forms already appearing.

With continued R&D, structure determination of specific chemical states of metalloproteins will become feasible, enabling users to understand enzymatic processes related to bioenergetics, the chemistry of small molecules like CO, CO_2 and N_2 involved in global carbon and nitrogen cycling. In this context, new approaches will be developed to mitigate radiation damage issues associated with these classes of metalloproteins.

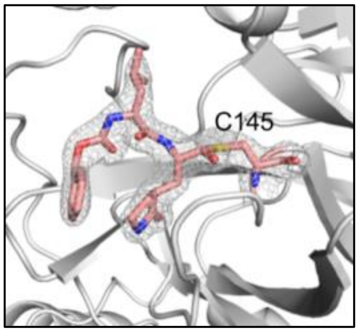
A specific effort will be devoted towards structure-based drug design, emphasizing fragment-based screening and automated feedback of structural results to molecular dynamics and modelling approaches. This will be important not only for finding new drugs for the most serious illnesses, but also in the fight against the growing problem of drug resistance.

The methodological and technological developments, summarized below, will enable the science outlined above, as well as a diverse array of other studies by SSRLs' user community, by building 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 areas:

- Development of advanced microfocus capabilities enabled by the new undulator micro-focus BL12-1, including implementation of a 'next generation' pixel array detector, a high-speed microgoniometer, and several associated sample delivery systems such as liquid injectors and multi-sample room-temperature devices.
- New approaches to enable challenging projects that probe the connection between biological function and structure, such as the study of intermediate states of metalloenzymes and large multi-component complexes, experiments that require data free of radiation induced aberrations and use of small sensitive crystals.
- 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 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 SMB research community at SSRL and LCLS.
- 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, Stanford and other universities and research institutes.



X-ray crystallography structure of the SARS-CoV-2 main protease with a potential COVID drug inhibitor bound in the active site (red sticks). The drug was originally developed to treat coronavirus in cats and it is a strong candidate for the treatment of coronavirus in humans.

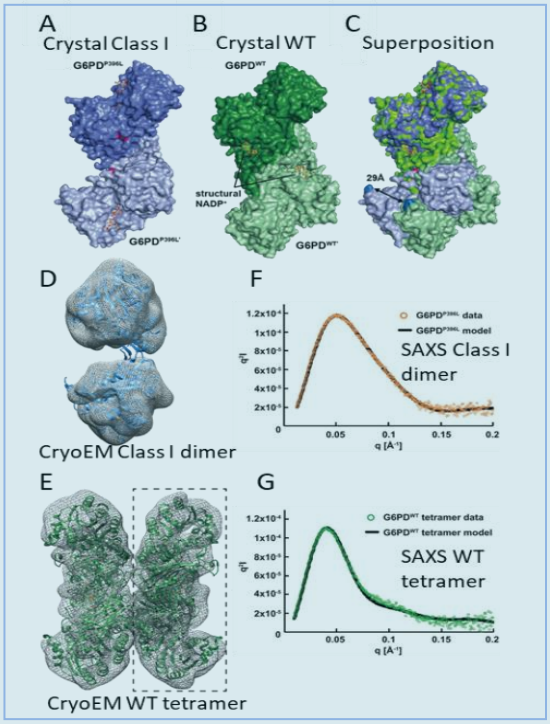


*A close-up view of the active site. Typical binding of molecules in the active site of drug targets is through relatively weak hydrogen bonding. Whereas the inhibitor drug shown here (red sticks), in addition to several hydrogen bonds, forms a strong covalent bond with a cysteine residue (Cys145) of the SARS-CoV-2 protease. This strong bond inhibits the protease's function in the life cycle of the SARS-CoV-2 virus, effectively preventing the virus from replicating. Electron density for the drug inhibitor is shown as a grey mesh. (Nat. Commun. **2020**, 11, 4282)*

Macromolecular Crystallography, SAXS, and Cryo-Electron Microscopy in Multi-Technique Synergy Solve Enigma of Prevalent Blood Disorder

About 400 million people worldwide live with potential of blood disorders due to deficiency in the enzyme glucose-6-phosphate dehydrogenase (G6PD). While some people are asymptomatic, others suffer from jaundice, ruptured red blood cells and, in the worst cases, kidney failure. The enzyme produces the electron carrier NADPH to help reduce reactive oxygen species, which accumulate and damage cells under stressful conditions. More than 160 mutations of G6PD are classified into 5 groups depending on the severity. The structure of the normal G6PD has been known for three decades but the real cause of the most severe Class I mutations have been unknown, hindering therapeutics developments. Structures of several Class I mutants have been determined for the first time using multimodal approaches including X-ray crystallography, SAXS, Cryo-EM and MD simulations. All Class I mutations show new significant and long distance structural changes in G6PD. The mutant dimers are significantly bent, preventing stable tetramer formation, leading to occlusion of the active site through long distance transmission of the structural defects from the dimer interface.

This discovery paves the way for developing novel therapeutics to address the structural and functional defects of the most severe class of G6PD mutations leading to blood disorder as well as other malfunctions in brain and liver. (*Proc. Natl. Acad. Sci. USA*, ***2021***, https://doi.org/10.1073/pnas.2022790118)



Crystal structures of Class I mutation and normal G6PD (A&B) show significant bending in the former, as shown in the superposition (C). CryoEM structures G6PD (D&E) and SAXS profiles (F&G) corroborate the crystal structures, showing the long range propagations of structural defects to the active sites. Together the multimodal structural studies explain, how G6PD enzymopathy is caused in the most severe Class I patient cases.

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.

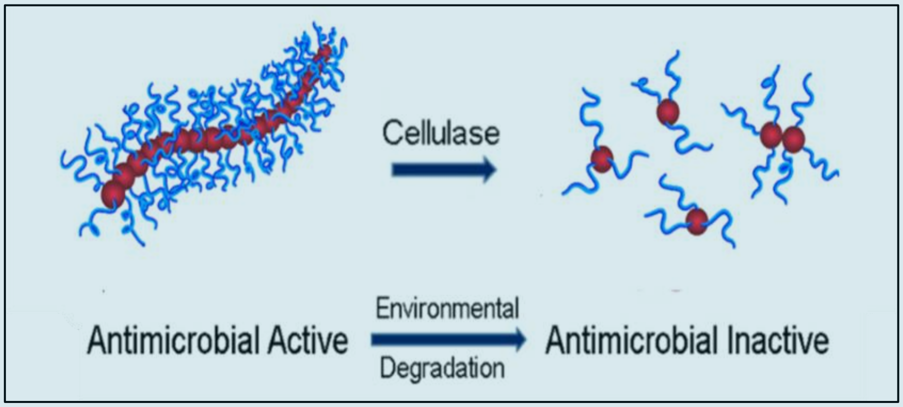
The BioSAXS beam line BL4-2 features state-of-the-art experimental facilities for solution scattering, lipid membrane and fiber diffraction at moderately high-to-very small scattering angles through a flexible camera approach. Specialized sample handling devices include an automatic fluid sample changer 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, state-of-the-art pixel array detectors and a gap-less, high-efficiency CCD detector that can be used interchangeably.

The BL4-2 facility further includes advanced instrument control and real-time 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 to advance BioSAXS will focus on:

- Expanding the microfocus beam capabilities at BL4-2 to push the frontier of SAXS measurements, enabling measurements on very small sample volumes, mixing experiments with high time resolution, as well as SAXS-based tensor-tomography measurements with high spatial resolution.
- Significantly advancing the instrumentation capability to enable automated and remotely accessible data collection 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, automation and data quality for state-of-the-art chromatographic purification methods directly coupled to the SAXS data collection to enable reliable high-quality SAXS measurements for problematic and aggregation-prone samples.
- Refining and further automating the data collection system for high-throughput solution scattering and diffraction measurements to minimize sample consumption, increase reliability and flexibility of the setup and fully enable remote-access data collection for users over extended periods of time.
- Expanding the real-time data processing and analysis pipelines to increase their flexibility and allow for experiment specific user defined workflows.

Antibiotics with a Built-in “OFF” Switch



Widespread use of antibiotics in healthcare and agriculture has led to their artificial accumulation in natural habitats. Emerging evidence has suggested the dire consequences caused by the increasing accumulation of antibiotic wastes in ecosystems, including short- and long-term adverse effects on the structure and function of microbial communities involved in biogeochemical cycling and organic matter degradation, contamination of water, plants, stockbreeding, and aquaculture products in the food chains, and promoting the development of resistome, *i.e.*, the environmental reservoir of resistance genes shared among pathogenic and nonpathogenic bacteria**.** Ideally, antibiotics should remain fully active in clinical services while becoming deactivated rapidly once released into the environment, however none of the currently used antibiotics meet this criterion.

In this study, antimicrobial compounds have a built-in “OFF” switch that is responsive to natural stimuli. The design is based on the self-assembly of nanoparticles from environmentally benign building blocks of polymer grafted cellulose backbones. Synchrotron based small-angle X-ray scattering and diffraction (SAXS/D) was used to probe the interaction of such nanostructures with membranes mimicking mammalian and bacterial cells and helped optimize the target compounds. The SAXS data were combined with biochemical assays to establish the antibiotic’s cytotoxicity and the enzymatic degradation. These nanostructured particles are potent agents against both Gram-positive and Gram-negative bacteria, including clinical multidrug-resistant strains, while they are harmless for mammalian cells. After being discharged into the environment, they are “shredded” into antimicrobially inactive pieces by cellulases that do not exist in the human body but are abundant in natural habitats. Thus, this study illuminates a new concept of mitigating the environmental footprints of antibiotics with rationally designed nanoantibiotics that can be dismantled and disabled by biorthogonal chemistry occurring exclusively in natural habitats, significantly reducing the environmental impact of such antibiotics (*Biomacromolecules,* ***2020****, 21, 2187-2198)*

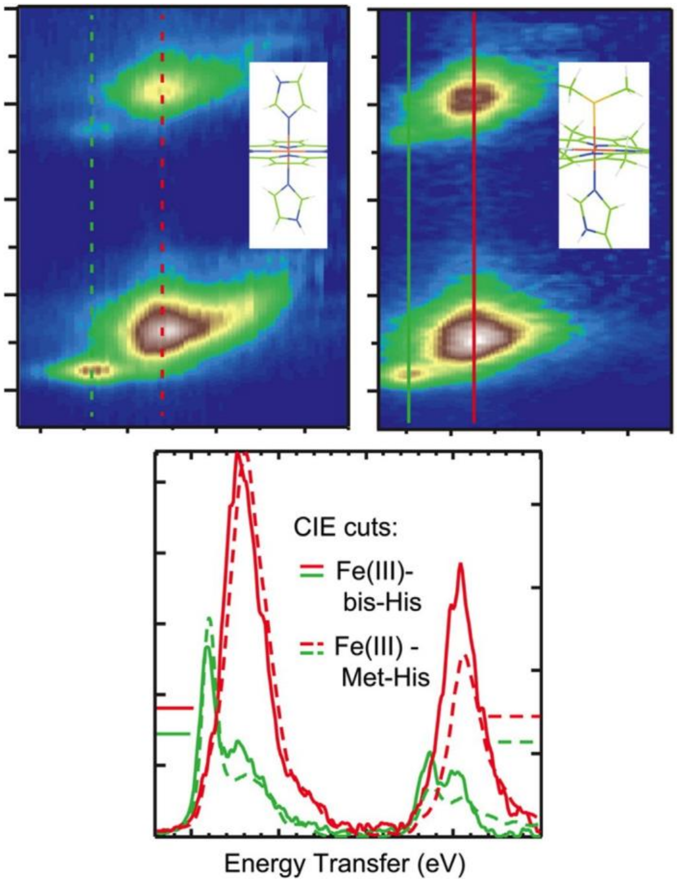
4.3.3 Biological X-ray Absorption and Emission Spectroscopy

Metal ions play crucial roles in many 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, when applied in the fast time domain, help probe transient catalytic intermediates formed in biological processes. 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 dedicated facilities 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 important biological processes and to biomimetic catalysis, such as oxygenase and oxidase catalysis, nitrogen reduction and oxidation, methane formation, CO_2 reduction, and electron transfer reactions.

Future R&D and methodological developments include:

- Further develop the 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 coupling to LCLS experiments into the fs time domain.
- New instrumentation development 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.
- Expansion 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, with enhanced focusing optics, and over energy ranges from tender to hard X-rays, extended to XAS tomography.



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 Cryo-Electron Microscopy and Tomography (Cryo-EM/ET)

Using electrons, Cryo-Electron Microscopy (Cryo-EM) and Cryo-Electron Tomography (Cryo-ET) techniques provide images of biological materials that are frozen in their native state, ranging from proteins and nucleic acids to very large biological assemblies and complexes, at resolutions that range from nanometer to the atomic level. Cryo-EM uniquely captures dynamic ensembles of macromolecular structures as they occur in solution. Image analysis can then separate these ensembles to produce high-resolution snapshots capturing their compositional and conformational variance and their dynamics. Cryo-ET is an emerging technique that can resolve subcellular structures inside the cell and tissue, with the capability to reach nanometer resolution for the entire sample and atomic resolution for abundant molecular components *in situ* with post-tomographic data processing of those structures. Cryo-Fluorescence Light Microscopy (Cryo-FLM) and subsequent Cryo-ET of frozen, hydrated cells can be used to label specific proteins and study cellular and molecular locations, functions and dynamics in the 3D context of cells and tissues at a higher resolution than any other imaging techniques. Cryo-ET can be preceded by Focused Ion Beam Scanning Electron Microscopy (CryoFIB-SEM), to produce lamellae from vitrified cells that are thin enough for Cryo-ET imaging.

Cryo-EM/ET facilities within the CryoEM & Bioimaging Division at SSRL are wide-ranging and currently include a suite of five 200-300 kV electron microscopes, a CryoFIB-SEM, and sample preparation and data analysis tools. Some of these are associated with two NIH funded centers, the Stanford-SLAC CryoEM Center, and the Stanford-SLAC CryoET Specimen Preparation Center. There is synergy between the Cryo-EM program and the SSRL's SMB X-ray facilities in R&D, science and user access programs.

The suite of Cryo-EM/ET instruments are used for a wide range of biological and biomedical research by scientific staff, faculty and external users. Scientific applications include high resolution structural study of viruses and their interactions with receptors and antibodies, membrane-bound proteins including transporters and pumps, molecular machines involved in metabolic pathways and in the carbon cycle, bacteria, fungi, free-living algae, plant tissues and cells under normal or environmentally stressed conditions.

4.3.5 Training the Next-Generation Workforce

As a tightly integrated feature within the Structural Molecular Biology and Cryo-EM/ET program activities, 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 and Cryo-EM/ET facilities to the home laboratory through advanced remote-access developments in tandem with rapid access scheduling mechanisms. This will be coupled to developments of on-line learning tools, including simulation of experiments, in collaboration with Stanford University's on-line teaching tool developers.

4.3.6 Summary of New Synchrotron Radiation Capabilities

The scientific strategy is driving the need for and planned around the following beam lines and instruments:

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 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 10 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 required 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 required a new injection septum magnet, which was installed in 2019. Running lower emittance in operations also requires a reduction of the aperture of the SPEAR3 beam dump to better contain electron losses at the dump. The reduced-aperture dump has been designed and built, and will be installed in the April, 2021, downtime. This will allow delivery of lower emittance in operations later in 2021.

Multi-bunch Feedback Kicker

SPEAR3 has had a transverse multi-bunch feedback system in operations since 2014. The feedback performance, however, was limited by the bandwidth and power capability of the transverse kicker. In FY2017, the kicker performance was found to be insufficient to control electron multi-bunch instabilities driven by the newly installed BL15 in-vacuum undulator. In order to implement a rapid solution, SSRL obtained a kicker on loan from the Advanced Light Source, which was installed in summer, 2017. This upgrade to a high-bandwidth kicker enhanced storage ring stability and performance in several ways:

- Improved 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.
- Suppressed resistive wall instabilities from future small gap insertion devices.
- Damped top-off transients faster, decreasing stored beam perturbations during top-off injection.
- Damped multi-bunch instabilities driven by resonances in in-vacuum undulator chambers, such as those recently measured for the beam line 15 IVU.

SSRL is still in a vulnerable position, however, because our operational kicker is on loan from ALS. To mitigate this vulnerability, two new kickers have been designed and are presently in fabrication. We plan to install these SSRL-owned kickers in the summer 2021 shutdown. They are yet higher performance than the ALS kicker, and will provide these additional capabilities:

- Improve bunch purity with bunch cleaning to improve timing mode data quality.
- Enable studies of novel timing operational modes, such as resonant crabbing and resonant pseudo single bunch.
- Possibly increase single bunch current limit.

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. Both simulations and experiments are presently ongoing to determine the ideal separation for the kicked camshaft beam. This effort will provide a desired kick amplitude for the PSB kicker design.

5.1.2 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 30 years old and with some RF components over 40 years old.

SPEAR3 Low Level RF Upgrade

The SPEAR3 low level RF (LLRF) system comprises analog RF signal processing hardware that drives the klystron, two programmable logic controllers (PLCs) for machine protection and controlling the high-voltage power supply, and other ancillary hardware primarily for machine protection. Most of these subsystems come from PEP, are antiquated and reaching their end of life, and support for these is very limited. Work is ongoing to upgrade the LLRF system to a modern architecture, addressing several serious maintenance vulnerabilities and improving reliability by addressing issues associated with limited personnel who are familiar with the present aging system.

We are in the process of evaluating a commercial system for replacing the RF signal processing hardware. Work is also underway on the machine protection PLC and on generally documenting the existing system in fully specifying the upgrade. The estimated installation timeframe of the LLRF Upgrade is during the summer downtime of 2022.

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.

Injector Modulator Controls Upgrade

Work is starting in FY2021 to upgrade our injector linac modulator controls to the system presently used for the LCLS linac. The benefits of the upgrade include:

- Standardization with the LCLS system makes our system easier to maintain
- More stable linac operation
- Quicker recovery when the linac systems trip off

5.1.3 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.

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 has played a critical role in commissioning the 6 nm-rad SPEAR3 optics. The RCDS algorithm has also been used at other 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.

The group has also developed a new optimization algorithm called RSimplex. As with RCDS, the R stands for robust, which means the Simplex algorithm was modified to make it robust to measurement noise. Many optimization algorithms have been developed that can converge to the best solution when the function to be optimized is calculated to numerical precision on a computer. When such algorithms are implemented to optimize a measured parameter on an accelerator, they tend to fail due to measurement noise. RSimplex converges despite significant measurement noise.

Yet another optimization algorithm has been developed – multi-generation Gaussian process optimizer (MG-GPO). This algorithm has been successful in improving the nonlinear performance of SPEAR3 and APS, and for tuning the APS linac.

Differing optimization algorithms tend to perform best on differing optimization problems. The tool chest of algorithms developed at SSRL can be used to tackle a wide variety of optimization of measured parameters both for accelerators and in general.

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.

MBA lattice Optimization Studies

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, was built in Sweden, followed by the MBA rings ESRF-EBS in France and SIRIUS in Brazil. Numerous MBA upgrade plans at various stages of maturity exist at other synchrotron facilities around the world.

The SSRL accelerator group has been using the optimization algorithms they have developed to optimize the linear optics of MBA rings. This work is in the process of expanding to include simultaneous linear and nonlinear optimization.

5.2 Beam Line Development and Technical Capabilities

SSRL has developed and substantially implemented a plan that will keep SSRL beam lines competitive and productive over the coming years by using key scientific objectives to direct the integrated development of new and upgraded beam lines with innovative hutch equipment and state-of-the-art detectors. This beam line build-out plan significantly increases both beam line capability and capacity. The light source currently features 27 operating beam lines equipped with 33 experimental stations: 8 independently operating bending magnet beam lines on 5 bending magnet sources and 19 independently operating insertion device beam lines on 12 insertion device sources.

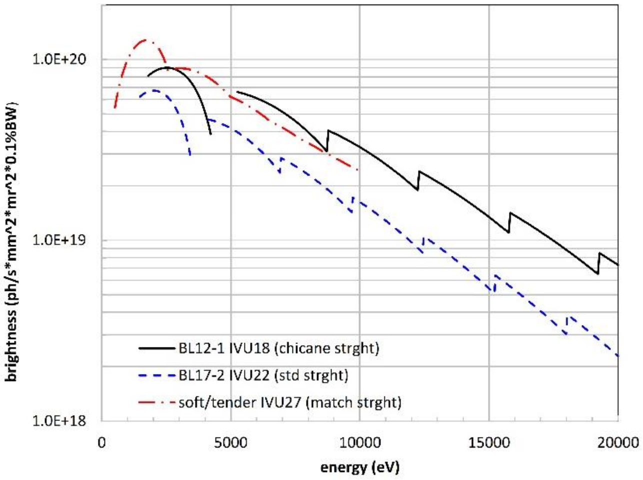
Table 1: SSRL beam line sources and science disciplines served circa 2021. Six beam lines / stations serve multiple scientific disciplines so the count by discipline exceeds the total number of beam lines (only major shared beam lines included).

SSRL Beam Lines by Discipline Circa 2021
27 BLs (13 wiggler, 4 IVU, 2 EPU, 8 bend)
13 Chemistry & Catalysis (10 ID & 3 bend)
9 Stuctural Molecular Biology (7 ID & 2 bend)
11 Material Science (7 ID & 4 bend)

SSRL has established and largely completed an ambitious program of developing new undulator beam lines as the key goal of the development plan. The first of these new in vacuum undulator (IVU) beam lines, BL15-2, was developed to support advanced spectroscopy applications including time domain measurements and has recently commenced regularly scheduled user operations. IVU BL12-1 was developed as a micro-focus macromolecular crystallography beam line, which expands upon the successes of the companion BL12-2 IVU crystallography beam line. BL12-1 completed an accelerated commissioning program in 2020 and was immediately employed in COVID-related research. IVU BL17-2 was developed to serve the materials scattering research community requiring a higher brightness source than existing wiggler beam lines at SSRL. This beam line, which features SAXS, WAXS, and time domain research capabilities, commenced user commissioning data collection in 2021.

In addition to these IVU beam lines, development of a bend magnet port for metrology applications is nearing completion. This port serves two beam lines with a 50-2300 eV soft X-ray beam line currently in operation and a hard X-ray beam line with both monochromatic and white beam capabilities expected to commence commissioning in 2021.

SPEAR3 can accommodate two additional insertion device beam ports within the existing building footprints, offering impressive brightness and flux in the soft and intermediate X-ray regime (see Figure). 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 IVU in chicane (IVU18), standard (IVU22), and matching (IVU27) straights assuming 6 nm- rad(H) by 10 pm-rad(V) emittance and 500 mA stored current. Only the odd harmonics are depicted. The IVU18 and IVU22 are optimized for approximately 5-20 keV while the IVU27 (not currently implemented) is configured to provide continuous tuning across the soft and tender X-ray energy range.

5.2.1 New Undulator Beam Line Developments

The lower emittance of the SPEAR3 accelerator upgrade, coupled with incremental lattice improvements, has reduced the SPEAR3 emittance to about 6 nm-rad or almost a three-fold reduction relative to the SPEAR3 project design goal. These improvements in electron beam emittance make in-vacuum undulators 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 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 a further three IVU beam lines (BL12-1, BL15-2, and BL17-2).

Taking advantage of the experience gained operating the BL12-2 IVU22 and the lower emittance ring, the new BL12-1 IVU (chicane IVU18, cf. Figure above) was designed for smaller magnet gap allowing the incorporation of 162 poles, compared to the 134 poles achieved with the same length BL12-2 IVU22. This higher brightness source has been teamed with state-of-the-art Kirkpatrick-Baez (KB) mirrors and the well-established SSRL LN_2-cooled monochromator design augmented with multilayer capabilities. The inclusion of the multilayers in the monochromator has been designed expressly to allow rapid, automated switching between multilayer and crystal to facilitate studies that benefit from the greater flux of a wider bandwidth beam such as crystal screening or serial crystallography. The focusing system yields a beam spot optimally sized for crystallography of ≤5 µm samples with beam stabilization servo systems to ensure stable illumination of small samples. The hutch equipment includes the evolving SSRL SMB macromolecular crystallography instrumentation for micro-crystal applications as well as emerging serial crystallography applications.

BL15-2 and BL17-2 each employ similar 22-mm period 174-pole IVUs located in standard length SPEAR3 straights.

Spectroscopy BL15-2 features a LN_2-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 have been relocated to BL15-2 with the spectroscopy instrumentation suite further augmented by a von Hamos spectrometer. A Tangerine pulsed laser system for time domain studies has been integrated into the BL15-2 experimental instrumentation as well.

BL17-2, which is optimized for materials scattering applications including both SAXS and WAXS, started optics commissioning in 2020. This beam line includes optics similar to that of BL15-2, but like BL12-1 includes a multilayer capability integrated into the LN_2-cooled double crystal monochromator . The multilayer offers enhanced flux for applications such as SAXS or selected timing studies, which are tolerant of a greater band width beam. The beam line end station has been designed to allow simultaneous SAXS and WAXS using Dectris Eiger X 4M (SAXS) and Eiger2 500K (WAXS) detectors. Once fully instrumented the BL17-2 instrumentation will include a Tangerine or similar laser system for pump-probe studies. Moreover, as developed in greater detail below, the optics design of BL17-2 allows addition of a second vertical focus mirror for enhanced isolation of pseudo single bunch flux when the accelerator is operated in this mode (see sections 5.1.1 and 5.2.2).

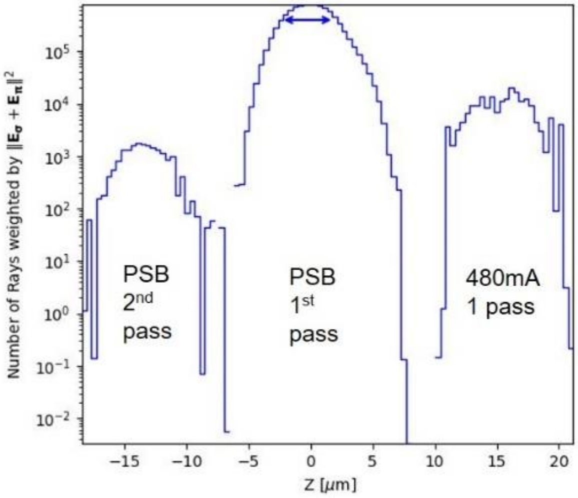
All other existing SPEAR3 straight sections are IVU-capable and present additional opportunities for high brightness IVU beam lines similar to those described above. Notably the matching straights on either side of the BL12 chicane in the East Pit are currently unoccupied and each offer a longer straight with lower βy for substantially improved undulator performance relative to a SPEAR3 standard straight.

5.2.2 Pseudo Single Bunch Timing Studies

As described in section 5.1.1, pseudo single bunch (PSB) involves deflecting a camshaft electron bunch into a different vertical orbit than the remainder of the filled bunches in the ring. Consequently the camshaft bunch photon emission cone occupies a different vertical phase space than the emission cone from the remaining filled bunches. Properly configured beam line optics can exploit these differences to isolate the X-rays emitted by the camshaft bunch while suppressing the X-rays from the remaining filled bunches. Using a combination of fixed vertical slits and the monochromator on BL17-2, for example, the simulated flux from a single turn of the 480-mA “normal” bunches is 0.02 that from a single turn of the 20-mA camshaft bunch.

Since the source position of the camshaft bunch is displaced in the vertical with respect to the remaining 480 mA, X-ray imaging optics will produce a focused beam spot for the camshaft photons that is displaced from the focus of the remaining 480 mA. This affords further isolation of the camshaft photons as a vertical slit at the focus can select only the photons emitted by the camshaft bunch.

For example the figure below depicts a SHADOW simulation of the vertical focus of BL17-2 at 12 keV. In this simulation the beam line optics are configured to accept photons from the first pass of the PSB and suppress most of the photons from the remaining 480 mA of stored current whose bunches are displaced 140 µm, 54 µrad, and 125-655 ns from the PSB. The source demagnifying KB focus optics result in approximately a 15 µm vertical displacement of the camshaft photon focus from the focus of photons remaining from the 480 mA stored current. (The PSB actually executes two revolutions around the ring on different vertical trajectories before it is kicked back into the same trajectory as the 480 mA beam. Thus the figure also shows a displaced X-ray focus from the second pass of the PSB which is displaced 122 µm, -83 µrad, and 780 ns with respect to the first pass of the PSB.) In this simulation introducing a 10 µm vertical slit centered on the first pass PSB beam spot essentially eliminates all photons other than those produced by the first pass of the PSB camshaft resulting in a well-defined X-ray beam pulse without significant contamination of X-rays outside the desired time window. Consideration of the figure reveals this means of isolating the PSB photons is very sensitive to the focus optics quality since extended tails of the undesired beam spots can contaminate the PSB photon focus.



SHADOW simulations of the BL17-2 KB mirror vertical focus at 12keV with realistic mirror shape errors assuming a 20 mA pseudo single bunch (PSB) vertically displaced 140 µm and 54 µrad with respect to the remaining 480 mA stored current. The PSB executes two revolutions on different orbits before it is kicked back into the same trajectory as the 480 mA beam. In addition to the obvious spatial displacement of the foci, the first PSB pulse is separated 125-655ns and 780ns from the 480 mA pulses and the second PSB pulse, respectively. In the simulation BL17-2 is configured to accept beam from the first PSB pass and suppress flux from the second PSB pass (displaced 122 µm and -83 µrad) and 480 mA stored current. A 10 µm slit centered on the first PSB peak effectively eliminates the second PSB and 480 mA flux. (Note the z axis is the vertical coordinate.)

While introducing a slit at the KB mirror system focus to isolate the photons from the camshaft PSB is effective, it requires the sample to be positioned downstream of the focus where the X-ray beam spot is both enlarged and beam structure is introduced owing to focusing optics residual shape errors. BL17-2 has been designed to allow retrofitting a second vertical focus mirror upstream of the KB focusing optics. This upstream mirror creates a demagnified source image on a slit configured to accept only the flux from first PSB in the same fashion as a slit at the KB focus depicted in the figure above.

The PSB X-rays transmitted through this selection slit propagate downstream and are refocused by the KB mirror system onto the sample thus avoiding focus spot degradation at the sample location. Current PSB plans envision operating BL17-2 with single stage vertical focusing while SSRL gains operational experience with the PSB operations mode starting in 2022. Both BL15-2 and BL17-2 can be retrofitted with two-stage vertical focusing for improved PSB performance if operational experience and user demand warrants the upgrade.

5.2.3 Revitalization of Existing 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.

For example, the materials scattering program relocation from BL7-2 to BL17-2 facilitated migrating the XAS imaging program from BL10-2 to BL7-2. In turn this enabled relocation of the Co-ACCESS heterogeneous catalysis program from bend magnet BL2-2 to the BL10-2 wiggler end station. As part of this BL10-2 repurposing the BL10-2 optics will be upgraded in 2022 for improved spectroscopy performance. Specifically the existing optical layout consisting of a toroid focusing mirror upstream of the monochromator will be replaced with the collimating mirror – monochromator – focusing mirror layout employed successfully on a number of SSRL spectroscopy beam lines. Moreover the standard LN_2-cooled double crystal monochromator will be exchanged with a LN_2-cooled fast scanning monochromator capable of collecting spectra at 20 Hz. In addition to the beam performance improvements owing to these source and optics upgrades, the BL10-2 end station affords the Co-ACCESS program the physical space to establish a full featured, catalysis research infrastructure including catalysis-required gas handling, novel *in-situ* reaction cells, and auxiliary sample characterization capabilities such as described in Section 4.2.1.

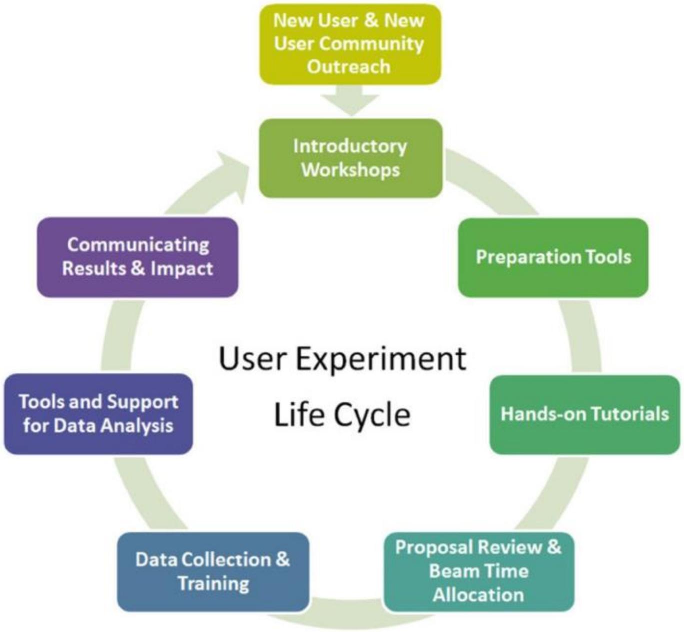
As noted in Section 5.2.1, the suite of advanced spectroscopy instruments from BL6-2 have been relocated to IVU BL15-2. The fraction of time on BL6-2 liberated by this change have been reapportioned to the TXM located in BL6-2 ES3 as well as tender XES and CT imaging programs in BL6-2 ES1 and ES2, respectively.

In addition to beam line repurposing and optics upgrades such as listed above, SSRL invests in experimental station instrumentation and detector upgrades to facilitate new research initiatives and greater productivity of existing programs as detailed in Chapter 4. With the broader use of remote data collection necessitated by the COVID pandemic, SSRL is leveraging SMB remote data collection experience by expanding implementation and utilization of sample handling robots and remote data collection software on chemistry/catalysis and material science beam lines. This investment anticipates that users will continue to exploit remote data collection capabilities even after the pandemic.

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 ~45-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. With our strategic plan that includes lowering the emittance, running at one of the highest- current levels on any mid-energy source worldwide, 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 topic-specific reports, and the 2010 BESAC report, "Science and technology – Strengthening the link between Basic Research and Industry". 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. SSRL 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. SSRL will continue to engage in workshops, such as the Workshop for Industry Researchers held at NSLS-II, December 2020.

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.

Assisting with Communicating Results

More than 600 papers are published annually as a result of research at SSRL, totaling over 17,000 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 60% 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.

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SLAC NATIONAL ACCELERATOR LABORATORY

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