ABBREVIATED VERSION

ALS-U: Solving Scientific Challenges with Coherent Soft X-Rays

Workshop report on early science enabled by the Advanced Light Source Upgrade

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Executive Summary

A revolution in x-ray science is underway that will take advantage of new technologies to enable the study of nanometer-scale features and interactions and the real-time observation of evolving chemical processes and functioning materials. The resulting information will pave the way to new scientific discoveries by enabling the understanding, prediction, rational design, and assembly of structures that exhibit emergent functionalities needed to address the world's most pressing technological problems.

So-called "third-generation" synchrotron light sources, like the Advanced Light Source (ALS), offer stable, nearly continuous x-ray beams that are ideal for studying homogeneous and simply organized systems. The vast majority of real-world systems, on the other hand, are heterogeneous and hierarchical. Today's soft x-ray storage-ring-based light sources lack the high transverse coherent flux that is necessary to effectively study these systems. A planned upgrade of the ALS, dubbed "ALS-U," will leverage recent advancements in accelerator technology to transform the ALS into a fourth-generation light source that can produce x-ray beams with 100–1000 times more coherent soft and tender x-ray flux than today's ALS—well beyond any storage-ring-based light source in operation, under construction, or planned.

ALS-U's capabilities will unlock the potential to image the locations of molecules, ions, and electrons and to measure how they migrate and interact. This information will lead to scientific advances in areas like ultralow-power information storage and processing, efficient chemical synthesis, highly selective ion transport and water purification, and artificial photosynthesis.

A workshop entitled "ALS-U: Solving Scientific Challenges with Coherent Soft X-Rays" was held at Lawrence Berkeley National Laboratory on January 18–20, 2017 to identify scientific challenges that ALS-U will address early on and to determine the instruments and tools essential for taking full advantage of ALS-U's capabilities. This report delineates the findings of the workshop.

ALS-U Techniques

The high coherent flux of ALS-U will offer the opportunity to perform ***3D nanoscale imaging*** with ***high spectral sensitivity*** over ***broad space and time scales***. Three interrelated classes of techniques, and the simultaneous application of combinations of them, are required to take full advantage of these capabilities:

1. **Nanoprobe spectroscopies** performed at ALS-U will allow many conventional soft and tender x-ray techniques to be performed with nanoscale spatial resolution.
2. **Coherent scattering and imaging** at ALS-U will encode information from inhomogeneous materials into scattered wave fronts, which can then be deciphered from speckle-diffraction patterns.
3. **Interferometry,** widely used in laser-based techniques, will become possible with ALS-U, allowing access to chemical and material properties with nanoscale resolution and very high spectral sensitivity.

ALS-U Early Science Opportunities

Equipped with the high coherent flux of ALS-U and the techniques above, new science opportunities will arise. Workshop participants discussed early science that will be made possible by ALS-U in three topical areas:

Spin, quantum, and topological materials: With applications in computing, information storage, transportation, and many more, numerous technologies will be revolutionized if we can learn to understand, predict, and control the properties of this class of materials at length scales from atomic to mesoscopic. Workshop participants identified key examples of early ALS-U science in this area, ranging from probing electronic states, imaging spin currents and magnetic moments, and characterizing response to external stimuli such as applied fields, strain, or changes in temperature.

Multiscale chemical processes: A wide range of processes, from catalysis and energy conversion to environmental transformation, occur over broad length and time scales. The ability to observe and model how these processes work together across scales is essential to determine system function. Workshop participants identified a number of applications of ALS-U capabilities towards this goal, including designing nanoparticle catalysts, imaging natural nanoparticle phenomena, and following biological and electrochemical transformations.

Soft and biological matter: Many soft and biological systems exhibit unusual properties and phenomena that we do not yet understand, including efficient thermally driven kinetics, self-assembly, and adaptation to environmental changes. We, therefore, are unable to predict these behaviors and design such systems from first principles. Workshop participants identified important early contributions that ALS-U's dynamic spatial and temporal range will make in this area, ranging from probing charge dynamics in organic materials and predicting rare events in soft condensed matter to examining the dynamics of soft-material interfaces and interactions between biological molecules.

Stemming from the early science opportunities, ***five crosscutting challenges*** were identified that ALS-U will address using its advanced capabilities:

1. **Spectral mapping of nanostructures and nano-objects:** ALS-U will offer a combination of few-nanometer resolution and soft x-ray electronic, magnetic, and chemical contrast that will complement scanning probe and electron microscopy to transform our understanding of and ability to control these systems.

2. **Designing functional interfaces:** ALS-U's photon-based microscopies, with few-nanometer resolution in 3D, will enable revolutionary probes of heterogeneous interfaces in diverse environments, taking advantage of a core expertise of ALS scientists and users who have led efforts to deploy these powerful chemical and material spectroscopies.

3. **Harnessing entropy as a material and chemical design tool:** ALS-U will enable the ability to measure, with chemical and material contrast, the thermally excited nanoscale fluctuations that store entropy.

4. **Manipulating nanoscale flows:** ALS-U will integrate highly sensitive interferometric detection with soft x-ray spectromicroscopy and dynamical measurements to detect nanoscale flows in diverse environments.

5. **Controlling materials chemistry and physics in confined spaces:** Powerful ALS-U spectromicroscopy tools will allow researchers to probe networks of confined chemical and material systems to optimize diverse functionalities.

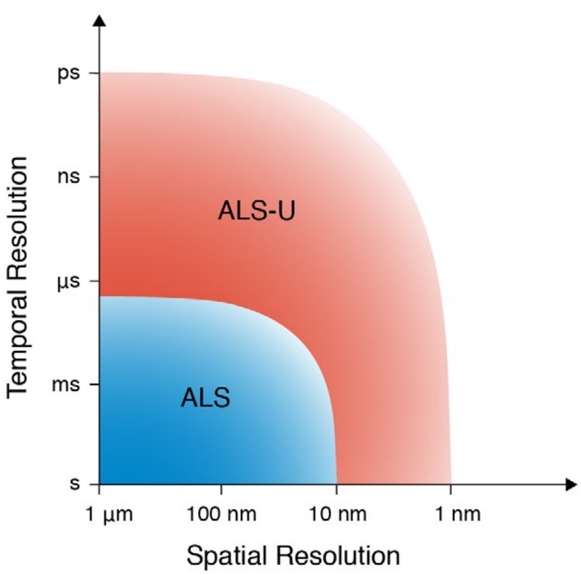
These overarching, crosscutting challenges align closely with recent Department of Energy strategic reports, further cementing the case for upgrading the ALS to establish the world's leading fourth-generation soft x-ray synchrotron light source.

1 Introduction

Chemical and material products are crucial to our nation's security, economy, and standard of living. Advances in these products, including the technologies that power machines and devices, drive industries, and protect our health and physical well-being, increasingly depend on our ability to understand and control underlying chemical and material processes, particularly at the nanoscale, where many macroscopic properties emerge.

The scientific case for improved capabilities in material and chemical analysis has been building for more than a decade within the Basic Energy Sciences program of the U.S. Department of Energy Office of Science (DOE-BES). Numerous Basic Energy Sciences Advisory Committee (BESAC) studies and basic research needs (BRN) and roundtable discussion (RTD) reports point to a future in which hierarchical chemical and material structures support interacting processes across broad length and time scales to provide the functionality needed to revolutionize a diverse array of energy systems.

Soft x-rays are critical to effecting this revolution since they can reveal chemical, electronic, and magnetic properties. Optimizing chemical and material processes—for instance, how various nanostructures are positioned and interconnected and how their collective operation is regulated—will require the ability to image the locations of molecules, ions, and electrons and to measure how they migrate and interact to support efficient function. Although the Advanced Light Source (ALS) has a strong track record of leadership in soft x-ray science over the last 24 years, it, like other storage-ring-based x-ray light sources, lacks the combination of nanometer spatial resolution, diverse spectroscopic contrast, and broad temporal sensitivity that is required to design and synthesize organized nanoscale structures and to optimize the nano- and larger-scale chemical and material processes that lead to useful functionality.



The Advanced Light Source Upgrade (ALS-U) project will leverage cutting-edge accelerator technologies to offer up to a 1000-fold increase in coherent flux compared to today's ALS across the soft x-ray energy regime. This will enable transformational improvements in spatial, temporal, and spectral resolution, as well as the required sensitivities for a broad range of experiments in material, chemical, biological, and earth systems—precisely the capabilities required to optimize chemical and material processes as described above (Figure 1.1).

Figure 1.1. Compared to the capabilities of the current ALS, the upgraded facility, ALS-U, will offer access to a much broader range of space and time scales, enabling imaging of the locations of molecules, ions, and electrons and measurement of how they migrate and interact to support efficient function.

In September 2016, DOE initiated the ALS-U project by approving its "mission need" and assigning it critical decision (CD)-0 status, the first milestone in making ALS-U a reality. To delineate scientific opportunities enabled by ALS-U and identify tools and techniques required to take full advantage of ALS-U capabilities, a workshop entitled "ALS-U: Solving Scientific Challenges with Coherent Soft X-Rays" was held at Lawrence Berkeley National Laboratory on January 18–20, 2017. The 170 workshop participants from 45 institutions engaged in vigorous, future-oriented discussions, the outcomes of which are detailed in this report.

The workshop participants' primary charge was to identify scientific challenges that ALS-U would help solve early on in three topical areas—(1) spin, quantum, and topological materials; (2) multiscale chemical processes; and (3) soft and biological matter—and determine the instruments and tools that are essential for taking full advantage of ALS-U's capabilities. Stemming from the early science opportunities they identified were five multidisciplinary challenges spanning the three topical areas, which ALS-U will address:

1. Spectral mapping of nanostructures and nano-objects
2. Designing functional interfaces
3. Harnessing entropy as a material and chemical design tool
4. Manipulating nanoscale flows
5. Controlling materials chemistry and physics in confined spaces

These overarching and crosscutting challenges, described in more detail below, align closely with critical needs identified in BES reports, further cementing the case for upgrading the ALS to establish the world's leading fourth-generation soft x-ray synchrotron light source.

1.1 REVOLUTIONARY CAPABILITIES PROVIDED BY THE ALS UPGRADE

A revolution in x-ray science and technology is underway in which the high phase coherence enabled by next-generation x-ray sources, coupled to powerful x-ray techniques developed over the last several decades, will be leveraged to probe heterogeneous chemical and material systems and processes. Third-generation storage-ring-based light sources, like the ALS, offer stable, nearly continuous x-ray beams that are ideal for studying the properties of homogenous and simply organized systems. The vast majority of functional materials and chemical synthesis platforms, on the other hand, are heterogeneous and hierarchical, exhibiting multiple phases and key structural features at the nanoscale. Today's soft x-ray storage-ring-based light sources lack the high transverse coherent flux that is necessary to effectively study these systems.

ALS-U will combine state-of-the-art multibend achromat (MBA) lattice accelerator technology, recently demonstrated successfully at MAX IV in Sweden, with a new injection system based on a concentric accumulator ring. The upgrade will also capitalize on modern undulator technology and other components of the existing ALS infrastructure (Figure 1.1.1). With these advanced tools, ALS-U will produce 100–1000 times more coherent soft and tender x-ray flux than today's ALS—well beyond any storage-ring-based light source in operation, under construction, or planned (Figure 1.1.2).

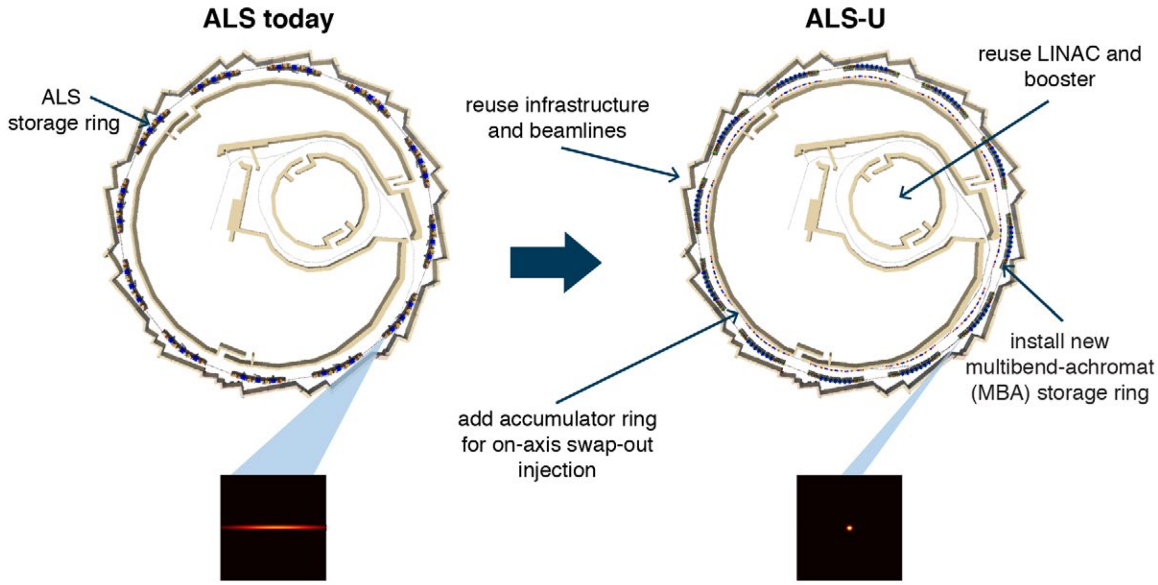
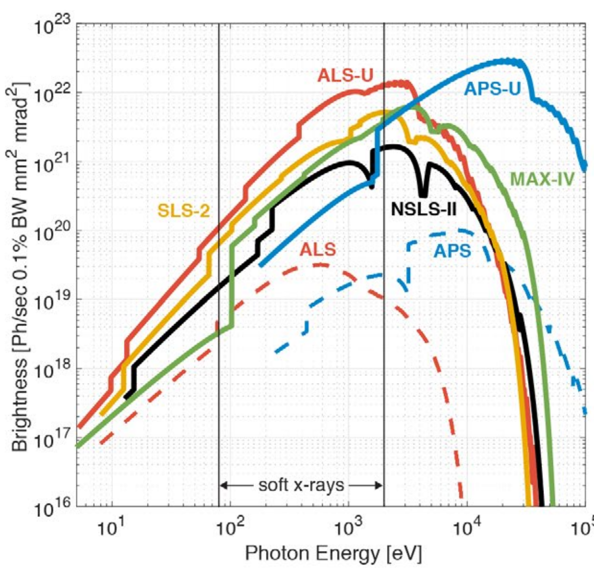


Figure 1.1.1. The planned ALS upgrade will involve removal of the existing accelerator lattice and installation of a multibend achromat lattice and an accumulator ring for swap-in, swap-out injection. The electron beam cross section will change from wide horizontally (left) to approximately circular (right) and small enough that the resulting x-ray beams will be transversely coherent (i.e., diffraction limited) through the entire soft x-ray regime.

An x-ray source that is small and sufficiently collimated (i.e., bright enough) will be diffraction limited, with a fixed phase relationship between any two locations on a wave front. The direct consequence is that all of the intensity of the beam can be focused into the smallest possible size, as defined by the wavelength. As a result, coherence-based experiments that are now done routinely with longer-wavelength lasers will become possible with x-rays with up to 1000-fold shorter wavelengths that are therefore sensitive to nanoscale phenomena.



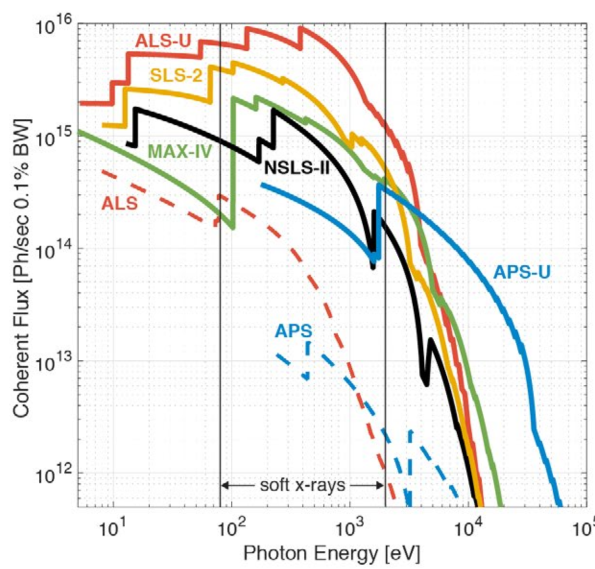


Figure 1.1.2. Envelopes of brightness (left) and coherent flux (right) for undulator sources at ALS, ALS-U, and several other operating and planned x-ray facilities. Coherent flux is the metric that determines the time required to accomplish an experiment with a given spatial, spectral, and temporal resolution, and is proportional to brightness divided by the square of the photon energy.

Previous generational advances in storage-ring-based x-ray facilities have provided similar degrees of increase in brightness and coherent flux, enabling new and powerful capabilities that have revolutionized our ability to understand chemical, biological, and material systems. The planned upgrade will transform the ALS into a world-leading fourth-generation facility characteristic of future synchrotron science. More specifically, the sensitivity of soft and tender x-rays to chemical and material processes will be dramatically enhanced when the x-ray wave fronts become smooth and transversely coherent.

Coherent soft x-ray beams can resolve nanometer-scale features and interactions while following real-time kinetics with high spectral sensitivity, revealing how chemical processes evolve and materials function. More specifically, the high coherent flux of ALS-U will enable a suite of tools that encompass ***3D nanoscale imaging*** with ***high spectral sensitivity*** over ***broad space and time scales,*** dramatically amplifying the already high impact of ALS soft x-ray spectroscopies. For example, chemical imaging experiments at the ALS have recently achieved few-nanometer spatial resolution in 2D on a model material, but on a more typical, realistic object can provide only 15-nanometer resolution with limited spectral coverage, resolution, and temporal sensitivity. ALS-U, on the other hand, would allow spectroscopies like angle-resolved photoemission spectroscopy to be applied to functional systems as nanoprobe techniques with natural or designed spatial or temporal heterogeneity. The suite of coherence-enabled tools is described in more detail in Chapter 2 and summarized by Table 2.1.

ALS-U's combination of capabilities, and their simultaneous application, will enable the understanding, rational design, and assembly of structures that exhibit emergent functionalities needed to address the world's most pressing technological challenges. These include materials and structures that can store and process classical and quantum information with ultralow power dissipation; microbial cells engineered to produce commodity and specialty chemicals from abundant starting materials; chemical microreactors designed to achieve efficient and selective multistep chemical syntheses; photoelectrochemical cells that enable artificial photosynthesis; and nanoporous membranes optimized for ion transport and water purification with high selectivity and efficiency.

1.2 FIVE CHALLENGES ADDRESSED WITH COHERENT SOFT X-RAYS

The science case for an improved source of coherent soft x-rays is supported by a host of BES documents. BRN reports elaborate the content of a broad-reaching series of BESAC-sponsored documents including *Directing Matter and Energy: Five Challenges for Science and the Imagination* (2008), *From Quanta to the Continuum: Opportunities for Mesoscale Science* (2012), and *Challenges at the Frontiers of Matter and Energy: Transformative Opportunities in Discovery Science* (2015). The output from several recent BES RTDs is also relevant, including *Controlling Subsurface Fractures and Fluid Flow* (2015), *Neuromorphic Computing* (2015), and *Sustainable Ammonia Systems* (2016), each of which is loosely related to the subject of one of the BRN reports. The recent BRN and RTD reports also provide updated advice on the tools needed to connect basic research needs to trends in translational energy research for many years to come.

Analysis of the multidisciplinary science opportunities delineated by this workshop revealed five overarching challenges spanning multiple disciplines that ALS-U will address. These crosscutting challenges align well with the research priorities discussed in BES reports, providing strong validation of the output of the workshop and illustrating the breadth and expected impact of the facility's next 20–30 years. In addition to being described in detail below, the challenges are presented in Table 1.1 along with several relevant BRN and RTD reports[1] and science opportunities identified by this workshop that link the two together.

Challenge 1: Spectral Mapping of Nanostructures and Nano-Objects

Nano-objects

Scanning probe and electron microscopies are currently some of our most powerful tools for studying nanomaterials, from synthetic carbon nanostructures to biological nano-objects like protein molecules and complexes. These tools can produce a dazzling array of micrographs, yet they often lack the contrast to map the data contained in those images onto the elemental, chemical, and magnetic information that would help us better understand how macroscopic function emerges from nanoscale properties. ALS-U will offer a combination of few-nanometer resolution and soft x-ray electronic, magnetic, and chemical contrast that will complement scanning probe and electron microscopy to transform our understanding of and ability to control these systems.

Challenge 2: Designing Functional Interfaces

Interfaces

Soft x-ray photoelectron spectroscopy has contributed significantly to our understanding of homogeneous surfaces and thin films. Yet our inability to adequately carry out nanoscale probes has limited our understanding of material and chemical systems with natural or designed heterogeneity. These systems—and the interfaces between them—are poised to enable many future energy-related technologies like artificial photosynthesis. ALS-U's photon-based microscopies, with few-nanometer resolution in 3D, will enable revolutionary probes of heterogeneous interfaces in diverse environments, taking advantage of a core expertise of ALS scientists who have led efforts to deploy these powerful chemical and material spectroscopies.

Challenge 3: Harnessing Entropy as a Material and Chemical Design Tool

Entropy

Thermal motion and the entropy it produces play crucial roles in our ability to synthesize novel material and chemical phases and understand new phenomena. Our knowledge of the entropic contributions to

[1] When this report was written, the BESAC and RTD reports noted above were all complete, and new BRN reports on Synthesis Science, Quantum Materials, Environmental Management, and Carbon Sequestration were also available. A BRN report on Water and Energy was in progress and is included here based on participation of workshop leads and attendees. An update of a BRN on Electrical Energy Storage is also in progress. Our discussion in these areas is based in part on RTD and BESAC documents and on previous BRN reports on similar subjects.

a system's free energy often relies on theoretical simulations that lack detailed experimental validation, and is not nearly as sophisticated as our understanding of the energetic contributions. This situation limits our capacity to design, create, and optimize targeted phases and structures. ALS-U will enable the ability, using quasielastic and inelastic soft x-ray scattering, to measure the thermally excited nanoscale fluctuations that store entropy. These measurements will be done with chemical and material contrast, so that, for example, the entropic contributions of spin, charge, and lattice degrees of freedom to the total entropy of a quantum material can be probed independently.

Challenge 4: Manipulating Nanoscale Flows

Flows

Nanoscale currents of molecules, charges, spins, and elementary excitations are poised to regulate the designed hierarchical structures of next-generation functional material and chemical systems. As in biological systems, the overall efficiency and selectivity of material and chemical systems will be determined largely by the degree to which we can localize and control these flows. Understanding currents in confined spaces, where chemical and material processes can be controlled and optimized, is particularly important. ALS-U will integrate highly sensitive interferometric detection with soft x-ray spectromicroscopy and dynamical measurements to detect nanoscale flows in diverse environments.

Challenge 5: Controlling Materials Chemistry and Physics in Confined Spaces

Confined Spaces

Biological systems are based on confined but connected spaces (e.g., subcellular components). This structure facilitates individual processes and allows overall cellular function to be regulated. The next generation of functioning chemical and material structures will be bio-inspired, with individual processes optimized in separate confined spaces. These processes will be connected by internally regulated nanoscale flows, and the overall network will be configured for specific applications like multistep catalysis and neuromorphic or quantum processing. Powerful ALS-U spectromicroscopy tools will allow researchers to achieve a precise measure of the internal workings of such systems, which will be crucial to their design and optimization.

Table 1.1. Close alignment of ALS-U crosscutting challenges and BES research priorities. Crosscutting challenges are listed in the lefthand column, and BES BRN and RTD reports are listed across the top row. Science opportunities identified by this workshop, and which link the challenges and reports together, are listed with the corresponding section of this report in which they are discussed in more detail.

ALS-U CROSSCUTTING CHALLENGES	BES REPORTS			
	Quantum Materials (2017)	**Synthesis Science (2017)**	**Electrical Energy Storage (2007, update in progress)**	**Energy and Water (2017), Environ. Mgmt. (2015), Carbon Capture (2010)**
Spectral Mapping of Nanostructures and Nano-Objects	Interferometric microscopy (3.1.5)	Multistep nanoparticle catalysts (3.2.3)	Granular electrodes (3.2.2)	Macromolecular machinery (3.3.6)
Designing Functional Interfaces	Antiferromagnetic spintronics (3.1.3)	Imaging electrocatalysis (3.2.2)	Soft material interfaces (3.3.4)	Environmental interfaces (3.2.6, 3.3.4)
Harnessing Entropy as a Material and Chemical Design Tool	Fluctuating spin phases (3.1.4)	Kinetic control of chemical processes (3.2.1)	Transport in soft matter (3.3.5)	Nucleating environmental and biological nanoparticles (3.2.4, 3.2.7)
Manipulating Nanoscale Flows	Imaging spin currents (3.1.1)	Imaging catalysis (3.2.2, 3.2.3)	Charge motion in photovoltaics (3.3.1)	Multiphase flows in nanoscale cracks (3.2.5)
Controlling Materials Chemistry and Physics in Confined Spaces	Imaging functionalized nanotubes (3.1.7)	Designing and controlling chemical kinetics (3.2.1)	Designing biosystems for biofuels (3.3.7)	Nanoscale environmental chemical kinetics (3.2.6)

1.3 REPORT OUTLINE

The remainder of this report describes in detail the opportunities and recommendations resulting from the charge to the workshop participants, which was to delineate the frontiers of soft- and tender-energy x-ray science enabled by ALS-U with the following goals:

- Identify and highlight some of the first transformative spectroscopy, scattering, and microscopy experiments that will be enabled by fully coherent and/or intense nanofocused soft x-ray beams provided by ALS-U;
- Determine essential ALS beamline and endstation developments and upgrades needed to execute these proposed experiments; and
- Explore complementary tools and techniques that are needed to take full advantage of the new capabilities of ALS-U, e.g., data processing, analysis, and interpretation tools; in situ sample preparation facilities; multimodal characterization techniques such as optical, electron, and scanned probe microscopies; and hard x-ray and infrared tools.

Chapter 2 discusses the research techniques of primary interest for coherent soft and tender x-rays at ALS-U, including: required upgrades to existing tools and the development of new tools as part of the ALS-U project to produce a suite of capabilities that will enable those opportunities, and ancillary tools that will be required to take full advantage of ALS-U capabilities.

Chapter 3 summarizes each of the three topical areas discussed in the workshop—spin, quantum, and topological materials; multiscale chemical processes; and soft and biological matter—and presents a number of specific early science opportunities that ALS-U will address. Each subsection describes the scientific importance, the essential role of ALS-U, and the broader impact of one of the opportunities. Labels in the header of each subsection identify the crosscutting challenges the opportunity addresses and the most applicable ALS-U techniques.

A related workshop on *Soft X-Ray Science Opportunities Using Diffraction-Limited Storage Rings*[2] was held in October 2014 at the ALS. The goal of that workshop was to evaluate how high phase coherence and stability could be leveraged more generally, whereas the current workshop and report concentrates on ALS-U science opportunities and the instruments needed to enable them. This workshop constituted an early step in ensuring that the upgrade will support a broad-based, vibrant, and world-leading scientific program. We look forward to continuing interaction with the ALS user and broader science community and to other, more focused workshops in the coming years.

[2] The corresponding workshop report is available at **https://als.lbl.gov/wp-content/uploads/2016/09/sxr_workshop_report.pdf**.