General Session: Science Highlights

Finding Gems in Low-quality Crystalline Proteins with an X-ray Mini-beam

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Protein crystals frequently suffer from problems such as high mosaicity, split or smeared diffraction maxima, small size, multiple lattices and radiation sensitivity. Many such crystals are useless in an "ordinary" beam, but can yield high-quality diffraction data in the 5-µm to 20-µm mini-beam at GM/CA CAT. The enzyme TE-DC is a good example. Twenty-five crystalline samples of selenomethionyl (SeMet) TE-DC were screened, but the only sample that diffracted beyond ~4 Å was a double crystal. A single-crystal region within the double crystal was selected by diffraction rastering with a 20-µm mini-beam; however, Friedel data from this region were incomplete due to radiation damage. A fresh region of the sample was selected by diffraction rastering with a 10-µm mini-beam, and additional Friedel data were collected. The merged, complete SeMet Friedel data set yielded a 2.1-Å electron density map of stunning quality 3 hrs after the diffraction experiment. The stable mini-beam combined with an intuitive user interface offering automated sample mounting and screening, beam-size selection and raster probing have allowed many users to find gems in their low-quality samples.

Research supported by NIH grant R01 DK042303 to JLS, and by NIH interagency agreements NIGMS Y1-GM-1104 and NCI Y1-CO-1020 to RFF.

APS Renewal Focus: Science

Dynamics of Life

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The organizers' title may be a bit over the top—but if it doesn't move, it's not "living". The "it" can refer to molecules small or large, organelles, whole cells or of course intact organisms. Synchrotron X-ray sources can in principle, and in many cases in practice, successfully probe biological, biochemical and chemical dynamics over a wide range of time and length scales, from femtoseconds to the generation time of an organism, and from ~10 pm to centimeters.

Illustrations will be drawn largely from studies at the molecular level in which, for example, a chemical process such as isomerization that lies at the heart of perception of light by a biological photoreceptor can be followed structurally in real time, as the atoms and groups of atoms move around.

The limitations of today's approaches to experimental dynamics—X-ray source, experimental design, detectors, radiation damage, data analysis—will be briefly discussed.

Materials for Energy Applications

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The world's demand for energy is projected to double by 2050, requiring tremendous growth in energy generation capacity. Clearly, a more diverse portfolio of energy sources must be developed, especially when one takes into account the limited supply of readily available fossil fuels and increasing environmental concerns. Energy systems of the future, whether they convert sunlight into electricity, produce fuel from splitting water, or transform carbon dioxide into fuel, are centered on materials. Next-generation energy technologies will place dramatically higher demands on materials—requiring enhanced functionality and performance under extreme environments. The ability to design materials at the atomic level, taking advantage of new synthesis approaches, characterization tools, and computational modeling, has the potential of yielding unprecedented advances in new materials that will be needed in future energy technologies.

Materials under Pressure

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Experiments based on diamond technology are now providing unprecedented insight into the nature of materials under extreme conditions. At pressures up to >300 GPa and temperatures from millikelvins to thousands of degrees, new phenomena are observed in hydrogen and other simple molecular systems; new superconducting, electronic, and magnetic materials are created; and unexpected findings in soft matter and biological systems are observed. These discoveries have been made possible by synergistic advances in synchrotron radiation methods, combined with allied developments in a variety of laser, electrical transport, magnetic, neutron scattering, and other analytical methods. Continued developments at dedicated high-pressure beamlines as well as beamlines optimized for particular techniques are essential for taking this research to the next level. New classes of experiments will require the use of nanobeams, higher energy photons, coherence, and beam time structure. There are important opportunities for dynamic compression and combined static/dynamic experiments at synchrotron and other advanced light sources in furthering our understanding of materials in extreme states.

APS Renewal Focus: User Visions, Part 1

A Path Forward to Advanced Nuclear Fuels, Founded upon a Unique and Crucial New Facility at the APS*

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In a break with past paradigms, the U.S. Department of Energy has proposed a novel approach to the development of advanced nuclear fuels: predictive numerical simulation. [1] The advent of massive parallel computing and other improvements in computation capabilities has opened the door to the possibility of simulating much of the work that would have necessarily been determined empirically in the past. Nevertheless, these simulations and projections require the input of fundamental physical parameters that are experimentally generated or at least benchmarked, as well as computational electronic structure models that accurately describe 5f electronic systems. In particular, there are two crucial problems that must be resolved: (1) a dearth of fundamental thermodynamic information and (2) an absence of an understanding of 5f electron correlation. To remedy the first malady, we propose a radical departure from past practices of calorimetry. Using the techniques first proposed by Martensson and Johansson [2] and then validated by Steiner et al. [3], we will use spectroscopically determined core level shifts to benchmark the computationally generated heats of solution. To resolve the second issue, we propose the pursuit of Fano spectroscopy [4] of the actinides, [5] including the minority transuranic daughter products so crucial to the reprocessing of nuclear fuels.

To truly resolve both of these sets of problems, a novel, dedicated Actinide Science Beamline is required, with full capabilities for handling highly radioactive transuranics, with exposed surfaces *in vacuo*, and high-brightness, high-resolution, circularly polarized soft x-rays in the hv = 100 to 500 eV range. The APS, where the possibility of a low-field device on a high-energy ring exists, is the best choice for such a synchrotron radiation source. Moreover, by utilizing the long beamline architecture of high-resolution beamlines, it will be possible to isolate the radioactive materials handling endstations in an adjacent building separated from the other APS beamlines.

**Based upon LLNL-PROP-404276.*

Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA27344. This work was supported by the DOE Office of Basic Energy Science

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APS Renewal Focus: Techniques

Coherent Diffraction and X-ray Imaging

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Coherent diffractive imaging is a rapidly developing technique with a number of emerging applications (e.g., [1]). In this talk I will review progress in the methodology, exploring how it can be applied to general extended objects using the ptychographical [2] and keyhole [3] approaches. I will also discuss methods that incorporate and use the less-than-perfect spatial coherence available from third-generation synchrotron sources.

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3. Abbey B, Nugent KA, Williams GJ, Clark JN, Peele AG, Pfeifer MA, De Jonge M, McNulty I: Keyhole coherent diffractive imaging. **Nature Physics** 2008, **4**(5):394-398.

APS Renewal Focus: User Visions, Part 2

Ultra-fast Polarization Phase Selective (PPS) Measurements at APS

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Tailoring electromagnetic field structure *ad breve tempus* by different physical objects has been identified as one of the most productive ways to image as well as to control physical processes. Although among the most difficult methods, combined PPS studies can reveal new selective and detailed information about electronic and nuclear processes from single molecules to large biomolecular systems and materials. Methods such as magneti circular dichroism can be applied to all systems and were shown to be very useful, but they are limited by instrumental difficulties. Our nuclear vibrational resonance spectroscopy studies at APS have not yet included PPS techniques. We have recently conducted PPS studies at National High Magnetic Field Laboratory ("PPS Studies," K. Rupnik, PI) of physico-chemical processes of significant interest to biomedicine and sub-nano material architecture. These studies indicate (1) that many of the obstacles to the PPS measurements can be removed and processes at much faster time scales can be observed, and (2) there are extremely promising new results in the areas where "no enzyme has gone before." We discuss here the work in the same area that can be done at APS, a higher generation light source.

Hard X-ray *in situ* Real-time Interfacial-structural-spectro-nanoscopy

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Heterogeneity is a key challenge for interfacial science that is a key element controlling the reactivity of realistic interfaces and offers a rich landscape of phenomena. I will describe some examples of novel opportunities that can be achieved through a fully optimized photon-in, photon-out interfacial x-ray "nanoscope" that could be realized through the APS renewal.

Rapid, Ultra-fast Time-resolved Laue Data Collection from Tiny Crystals after Homogenous Reaction Initiation

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Ultra-fast laser pump pulses to initiate reactions in protein crystals cannot be used to date, since the pulses do not penetrate deeply enough into the material, presumably due to two-photon absorption or other effects that make the crystals virtually black. Consequently, reaction initiation is very poor and time-resolved difference maps are usually empty when laser pulses a few pico-seconds long are employed. To circumvent this issue, small crystals a few micrometers in size must be used. Such crystals can be illuminated homogeneously even with ultra-fast laser pulses. Then, however, the scattering volume may become too small to collect a diffraction pattern with the existing synchrotron sources. At the moment, crystals with edge length of 100 μm are routinely used for time-resolved crystallographic experiments. The reduction of crystal edge lengths by one order of magnitude to 10μm also requires that the x-ray flux be increased by one order of magnitude to collect comparable time-resolved diffraction patterns, provided the beam can be focused to 10 x 10 μm^2. In this case however, potentially, only one diffraction pattern can be collected per crystal because of radiation damage. Multiple crystals must be brought successively and rapidly into the beam (see fig. 1). Then single-digit picosecond time-resolved experiments with homogenous reaction initiation will become feasible. We need to increase the x-ray flux in a single polychromatic pulse (in the hybrid mode) from 1011 to 1013 per pulse and decrease the duration of that pulse to single-digit picoseconds. By doing so, we will close the gap between the X-FELS and the synchrotrons available today.

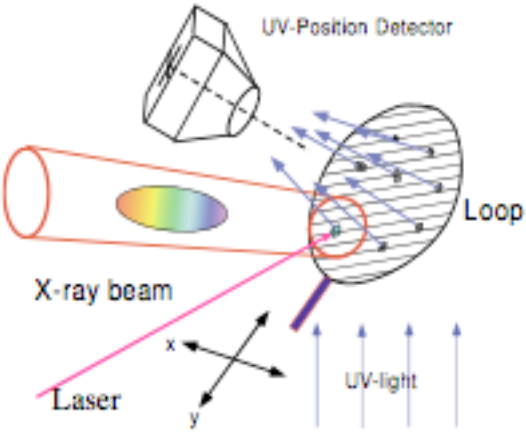

Fig. 1. Rapid scan mechanism to bring the tiny crystals in the X-ray beam. The reaction is initiated by a femtosecond or picosecond laser pulses. Either UV light or image processing can be used to determine the exact relative position of the tiny crystals.