Time-Domain Workshop: Summary and Recommendations
August 29–September 1, 2004

A workshop on "Time Domain Science Using X-ray Techniques" was held from August 29 – September 1, 2004, as part of a series of workshops on the "Future Scientific Directions for the Advanced Photon Source." The goal of the workshop was to identify future directions in scientific research using time resolved x-ray techniques and to address possibilities to produce picosecond x-ray pulses at the Advanced Photon Source. The workshop brought together internationally renowned leaders in atomic molecular and optical physics, chemistry and biology, and condensed matter physics as well as accelerator physics. The participants presented the frontiers in their fields, not limited to x-ray techniques, so that in addition to the interdisciplinary format, there was also a balance of existing and potential synchrotron users.

Time-domain research has been a mainstay for the Advanced Photon Source. Researchers at the APS have contributed greatly to our understanding structural changes on the ~100 picosecond and longer time scale and on the atomic length scale. The workshop examined current research occurring on these and shorter timescales including examples in isolated atoms and molecules, chemical and biological systems and condensed matter. In order to ensure that APS remains a leader in these fields, specific recommendations to facilitate and enhance experiments on timescales ≥ 100 ps were made and will be detailed at the end of this summary. However, by far the most exciting element of the workshop was exploring the possibility of shorter timescales at the APS, i.e. the generation of 1 ps x-ray pulses whilst retaining high-flux. This important time domain from 1 ps to 100 ps will provide a unique bridge for hard x-ray science between capabilities at current storage rings and future x-ray FELs**.** This unique potential has generated substantial interest and technical activities both during and subsequent to the workshop, and we believe, is of primary strategic importance for future scientific directions for the Advanced Photon Source. Such a development has the potential to turn the Advanced Photon Source into a Mecca for time-resolved x-ray work!

Current time-domain work in atomic and molecular physics in the hard x-ray regime focuses on the understanding of strong-field effects on inner-shell processes and monitoring Coulomb explosion dynamics. Strong AC fields of (≥3V/Å) are generated with standard ultrafast lasers and will be present at t=0 of all laser-initiated dynamics. An understanding of accompanying perturbations to absorption and emission spectra is important for the interpretation of optical pump/x-ray probe experiments, particularly those planned for next generation light sources. Looking forward to the 1 ps era, many new classes of experiments emerge. Among the most exciting is the ability to monitor coherently-controlled molecular processes. An example is field-free molecular alignment, where an impulsive kick is used to transiently align molecular axes with respect to laser polarization axes. Since the alignment lasts only for ~3 ps, x-ray pulses of ~1 ps duration are required for probing. Aligned molecules under field-free conditions would be of substantial interest for the proposed single-molecule imaging with ultrafast x-rays, but the alignment process still requires study, as could be done with the 1 ps x-ray probes at the Advanced Photon Source.

Advances in chemical and biological sciences depend upon the development of correlated time-resolved structural, kinetic and functional analyses. X-ray techniques provide the most powerful means to resolve molecular structures at the atomic level. X-rays with high photon flux in each pulse enable the extension of molecular dynamics studies by sophisticated ultrafast laser spectroscopy to structural dynamics studies at similar time scales. Therefore, the community of chemists and biologists focused on time-resolved structural studies is growing fast. Increasing needs for time scales ranging from ≤ 1 ps to ≥100 ps were identified. On the time scale of ≥100 ps currently provided by synchrotron x-ray pulses, targeted scientific developments were: a) extension of time-resolved macromolecular crystallography, scattering and spectroscopy to a broader range of chemical and biological systems (such as light-sensitive or artificially engineered light sensitive systems, temperature or pressure triggered processes, fast chemical mixing initiated reactions); b) time-resolved structural studies of molecular electronic excited states and reaction intermediate structures; and c) structural intermediates in biological enzymatic/protein, chemical catalytical processes, nanomolecular machines and supermolecules. On the time scale of ≤ 1 ps, many more chemical and biological processes can be followed such as coherent structural movements from Franck-Condon state through vibrational relaxation to thermally-equilibrated excited states, atomic rearrangements in isomerization, bond breaking/making, electron transfer coupled atomic movements, transition structures in catalysis and enzymatic reactions. Moreover, the control of chemical dynamics through diverse structures of molecular surroundings can be investigated.

Dynamical studies in condensed matter physics span a wide range of timescales from subpicosecond to millisecond and longer, and, broad fields of science from phase transitions to photoluminescence to nonequilibrium processes. In the workshop we heard many examples of cutting edge research in all of these areas including nonequilibrium electron and phonon dynamics, shock compression, self-trapped excitons in molecular solids, domain reversals as well as nucleation, growth and phase separation. For long time dynamics, coherence studies using photon correlation spectroscopy access a unique area of spatial and temporal (wavevector and frequency) phase space. Faster detectors are required to close the gap between systems that can be studied by these techniques and those by inelastic neutron and x-ray scattering. Ultrasmall means ultrafast. Dynamics of nanoscale devices such as ferroelectric switching occur on subnanosecond timescales. The time domain aspects of nuclear coherent scattering and spin dynamics, both of which have world-leading programs at the APS, were covered in other workshops in this series. On the fastest time-scales, photoinduced phase transitions, and coherent excitations have been primarily studied by optical techniques. A few pioneering x-ray experiments show that there are interesting dynamics that remain unresolved due to the relatively long x-ray pulse duration. An example is the photoinduced insulator-to-metal transition in VO_2 in which it is known, from optical studies, that the band gap is lost in a few hundred femtoseconds. Heroic x-ray diffraction experiments using both low brilliance (but short pulse) laser produced-plasma and slicing sources (ALS) provide evidence that this transition is of an inverse Peierls-type distortion accompanied by a change in valence.

The workshop was an unqualified success: the participants were unanimous in their recommendations and excitement. The recommendations are seen as critical components for APS to maintain a leadership role in the rapidly growing field of time-resolved x-ray science:

1. The APS must maintain the successful environment that both enables and enhances experiments on the ~100 ps and longer timescale;

2. The APS should pursue the development of high-flux picosecond beamlines through the use of advanced accelerator techniques. Such beamlines could produce 1 ps x-ray pulses and would be complementary to future LCLS facilities.

The first goal can be best achieved by forming a time-domain advisory panel to interface with the APS on detailed issues as: 1) providing maximum beamtime for time-resolved research through a suitable choice of standard operating fill pattern with maximal single-bunch current, 2) optimizing insertion devices, x-ray optics and end stations (including a soft- x-ray beamline) on *dedicated* time-resolved beamlines, 3) performing necessary R&D to develop advanced chopper designs and time-resolved detectors (fast readout 2D detectors, streak cameras, avalanche photodiodes…). It was also recommended that time-domain science be well represented at the APS Users' Meetings. For the second item, it was realized that the development of a high rep-rate, high-flux, short-pulse beamline would be complementary (and in many cases preferable) to X-ray FEL and ERL sources. This can only be achieved through a high priority R&D program in both the accelerator and optics areas for short pulse generation. The participants of the workshop proposed to seize this unique opportunity to implement the short pulse scheme on presently unused sectors of the APS. Significantly shorter pulses will enable fundamentally new science, as discussed above, while providing a unique capability to the APS, *and* will minimally impact other sectors.

These recommendations were designed to effectively utilize unique intrinsic capabilities of the APS for frontier time-domain science; develop infrastructure/technology to make the tools for time-resolved science more broadly available; and expand the capacity of time-resolved experimentation at APS. Finally, the present and future scientific users of the time domain science who participated in the workshop were eager to be fully involved in the implementation and success of both of these recommendati