University of Wisconsin FEL Workshop

Executive Summary

A Workshop took place on October 18-19, 2006, in Madison, Wisconsin to assess the scientific case for a next generation light source based on free-electron laser technology, and to specifically consider the potential for such a facility to be built at the University of Wisconsin. Over 100 scientists attended, including a significant international component, which was very important due to the lead that exists in Europe in this field. The workshop was organized in seven topical sessions, and the scientific highlights of each of these sessions are summarized below.

Femtochemistry. A femtosecond coherent soft x-ray/vuv laser provides a unique pump source that, when combined with 2-time correlation, pump-probe, and coherent multi-dimensional, and other nonlinear spectroscopic methods, would allow one to achieve temporally resolved mesoscale imaging (i.e. atomic level movies). Most of solution phase chemistry and all of biology involves atomic rearrangements on the nanometer length scale and 100 femtosecond time scale; whether it involves solvation dynamics or biologically relevant protein motions. By explicitly using the high time resolution and spatial coherence of the proposed soft x-ray FEL, it will be possible to project out the two-time correlations that lead to the key structural fluctuations driving the chemistry.

One can readily envisage observations of long range correlations in liquid water that are key to the special properties of water and direct observations of the structure-function relationship of biological systems on the pertinent length scale. Both of these example problems have been long standing issues; the resolution has the potential to lead to major advances in protein engineering as well as providing key fundamental insight into the key factors giving rise to living systems. Example research opportunities include: probing structure-function correlations of biological systems; directly imaging molecular scale motions, proteins, and molecular motors; defining the molecular origins of reptation in macromolecules; studying aligned molecules; achieving coherent control under both weak and strong field which could allow one to invert the temporal structure of control pulses to define the structural dynamics created by the control pulses; resolving structural fluctuations relevant to solvent re-polarization, reactive dynamics in solution, and functionally relevant motions of proteins; and, showing the participation of solvent dynamics in photo-dissociation and intermolecular charge transfer.

The proposed laser facility would also make it feasible to perform experiments on molecular Rydberg wavepackets. Rydberg wavepackets could image the molecular core using the amplitude and phase of high harmonics emitted during re-collision after ionization to reconstruct the highest occupied molecular orbitals. The sensitivity to the core arises because the quantum defect (i.e. changes in the Rydberg state energies) reflects the interactions with the atomic core. Further, it is known that photoionization spectra of Rydberg states are sensitive to large molecule conformation. This suggests that phase sensitive dynamics of Rydberg wavepackets could also allow reconstruction of
the shape of the core in very large molecules where Rydberg series and rotational band structure cannot be frequency resolved.

**Atmospheric and Intergalactic Gases.** An FEL in the vuv and soft x-ray region will enable new fundamental investigations of the interaction of intense and ultrafast photons with atoms, molecules, clusters and their ions. In particular investigations on ions, which suffer severely from low signal strength due to their low target density, will benefit enormously from an FEL. Also, the high flux and high spectral and spatial resolution of an FEL is needed to accurately model the whole atmosphere and to find the production mechanisms of pollution gases. Understanding this particle chemistry has a direct impact on societal concerns such as our health. Exciting new possibilities have emerged such as the study of multi-photon processes and of the electronic and structural rearrangement of molecules after photoionization on a femto-second scale.

**Pump-Probe Experiments.** The short pulse duration of the electron source and seed required to generate a fully coherent vuv/soft-x-ray FEL naturally lends itself to high time resolution studies of dynamics using pump-probe, stroboscopic techniques. For example, in atomic molecular and optical physics, the combination of high intensity optical lasers with x-rays enables researchers to study strong field effects in core-level photoionization. These effects are currently being studied in relatively high-Z atoms using hard x-rays, but there is also great interest in scaling to the vuv/soft x-ray regime. While in chemistry, such a source could be an important tool for studying reaction and solvent dynamics using ultrafast x-ray absorption spectroscopy (for example charge transfer to solvent reactions, spin-crossover, reactions etc.) And in condensed matter, magnetic x-ray circular dichroism experiments can view magnetization dynamics that can occur on a subpicosecond time-scale. In all of these fields, it is essential to have features described for the proposed machine including tunability (from at least a few hundred volts to a few kilovolts), high flux, a repetition rate commensurate with an external pump source (often an amplified laser), and polarization control, which is particularly important for magnetism. At the SPPS experiment at SLAC, which was the first hard x-ray FEL source, recent pump-probe experiments on structural dynamics in semiconductors and semimetals have shown that synchronization is a key issue in obtaining the highest possible temporal resolution. However, we also heard how it is possible to maximize the temporal resolution by performing single-shot experiments, or to utilize the residual timing jitter as a means of random sampling when the arrival time can be measured on a shot-by-shot basis. The proposed 10 fs synchronization capability of the proposed machine is very attractive.

**Biological Systems.** Nanometer scale direct imaging of biological structures in cells may be the key to understanding biological dynamics. Biology is deeply influenced by the structure of biomolecules, the dynamics of biomolecules, the way that biomolecules interact with each other, and how the cellular architecture is influenced by the structure and dynamics of the molecular components. At its most fundamental level, the length scale of these structures and dynamics of primary biological components is on the 1 to 100 nm length scale. Although there are ingenious optical techniques that reach into that length domain, they are indirect and of limited effectiveness. Ultimately, nanometer
length scale real time resolution in biology will require nanometer length scale optical probes: VUV and soft x-ray wavelength photons. Although energetic photons with this wavelength can potentially be damaging to cells, if the flux is large enough and the spot size small enough, then “flash” imaging of biological objects can be done in real time at the nanometer length scale. Such a high-brightness, high-fluence, short-duration source would revolutionize the imaging of biological objects.

Another area of biological opportunity includes the potential for two-color femtosecond or picosecond dynamics of biological dynamics. Biomolecules are nanomachines: they need to move to work, and these movements are frozen in crystals. Two color femto/picosecond pump probe experiments can track the energy and conformational flow of biomolecules in solution. By “color” we mean a huge range of photon frequencies, from the far IR of the thermally driven collective modes at 100 microns to the x-ray region at 10 nm where crystal field effects change the absorption edges of critical metal ligands in proteins. Only a Free Electron Laser can under one roof and with one facility provide ultrafast probes over this range.

Finally we mention the exciting possibility to deliver ionizing radiation to cells with specific ultrahigh spatial resolution. It is now clear that unraveling the response of cells to ionizing radiation is critical for understanding many fundamental problems in biology, from aging to apoptosis (programmed cell death) to the origins of cancer. Since a cell is spatially highly heterogeneous, simply irradiating cells results in the activation of many different biological pathways. The high brightness of a coherent x-ray/vuv light source would allow irradiating cell components with 100 nm or better spatial resolution, which would be a major step forward in unraveling the response of cells to genomic and component damage.

**Exotic Materials, Clusters and Nanostructures.** A free electron laser would open new frontiers in the development and exploitation of new materials and nanostructures. Unique opportunities in characterization include the potential to understand the influence of rare events on the properties of liquids and on epitaxial growth. Nucleation, for example, is a rare event that is universally important to producing epitaxial heterostructures that is understood only empirically. Similarly, structural fluctuations in complex liquids and amorphous solids can be addressed using emerging structural techniques based on coherent scattering. Spectroscopic probes of novel electronic and magnetic materials will be possible with unprecedented spatial resolution and will offer the possibility of directly probing many-electron wave-functions in nanostructures. These opportunities in characterization are complemented by opportunities to use these sources to point the way to new fabrication technologies based on lithography with coherent light and on the direct modification of materials using femtosecond-scale non-thermal processes.

**Resonant Inelastic X-ray Scattering (RIXS).** The basic purpose of RIXS is to study the elementary excitations in condensed matter, particularly those charge excitations that cannot be accessed by neutron scattering such as plasmons, excitons, orbitons, charge stripes, superconducting gap excitations, etc. Of particular interest is the dispersion of these excitations, i.e. how they propagate and transport energy about a material. Charge
excitations are a fundamental part of our understanding of condensed matter. However, RIXS methods, as currently implemented at storage-ring sources, cannot provide the high (milli-volt) resolution required for studies of such phenomena. Moreover even if such resolution were attainable it would require prohibitive compromises in intensity. Therefore, most of the dynamical charge processes that represent the foundation of our understanding of condensed matter today remain inaccessible to experiment.

There are two new and powerful approaches to RIXS that would be enabled by a soft x-ray FEL. The first is to exploit the huge increase of $10^6$ in the time-average flux and push existing technology to the limit, i.e. to use multi-stage grating spectrometers, as is used in optical spectrometers, to reject the elastic scattering allowing one to resolve features at much lower energy transfers. The second, which exploits the transverse coherence and time structure of an FEL, is to dispense with grating spectrometers and use photon cross-correlation techniques to map the phase space area of the pulse, from which the RIXS spectrum can be extracted. This has never been tried before but has the potential to provide sub-meV resolution at the MHz repetition rates of the proposed FEL facility. With these two methods one has a very high chance of success and which might finally permit access to the charge modes that are central to modern condensed matter physics questions.

**Time Resolved Imaging and Coherent Scattering.** The unprecedented coherence properties of soft x-ray FEL sources will enable unique and incisive probes of nanoscale complexity to be developed. The primary advantage of scattering transversely coherent wave fronts is that this process maps spatial and temporal correlations inside a soft, hard, or biological material into a far-field diffracted wave where they can be easily analyzed. For example, when coupled to well-known atomic, molecular, spin, and other more exotic sensitivities of soft x-rays, diffractive imaging will allow correlations to be mapped with few-nanometer resolution over a volume the size of a typical cell. Using photon correlation, pump-probe, and probe-probe techniques, fluctuations can be measured over a very broad time scale simultaneously with nanoscale spatial sensitivity. Most of these ideas are currently being developed using spatially-filtered undulator beams from third generation facilities, but all will be dramatically revolutionized by the availability of fully coherent FEL sources in the soft x-ray regime.