Working Group I: Chemical Science and Technology Summary Report

J. Viccaro

1 Overview

As demonstrated by the Birgenau-Shen report, synchrotron x-ray facilities play an important role at the forefront of chemical research. Historically, each major development in laboratory and synchrotron sources has allowed new types of measurements, has expanded the realm of x-ray-based research, and has resulted in important and entirely new science. The most recent of these advances is the development of undulator-based, "third-generation" synchrotron sources, as exemplified by the ESRF, ALS, APS, and SPRing-8 facilities. In the hard x-ray region (~1 Å), the improved characteristics at third-generation undulator facilities include small beam size, low divergence, partial coherence, high flux per bunch, and enhanced brilliance. These important attributes of third generation facilities make it possible to perform many new types of measurements that were previously impossible, opening up entirely new areas of scientific study.

One expects no less scientific impact from the proposed 4th generation FEL-based hard x-ray source. The characteristics of these sources, which include high coherence, short-pulse duration, and extremely high peak intensity and brilliance, will bring x-ray science into entirely new regimes. The current challenge is to identify medium-term areas of research in which the characteristics of the fourth generation sources will result in important advances in the chemical sciences. While it is relatively straightforward to speculate on the impact of the increased peak source flux of the new sources, speculating on the scientific impact of the coherent, spatial, peak power, and dynamic properties of the new sources is much more challenging.

As will be discussed in this report of a preliminary meeting, some areas within the chemical sciences have been identified as areas in which these new sources could have significant scientific impact. These include areas currently using x-rays as probes or areas where molecular dynamics are studied using laboratory laser sources.

In addition to this cursory scientific impact study, the group spent some time identifying future steps necessary to flesh out a plausible scientific case in the chemical sciences. This includes planning for future meetings and participation of the extended scientific community involving researchers in related femtochemistry areas.

2 Representative New Scientific Thrust Areas

The majority of potential applications of the fourth generation sources discussed by the working group are based on pump-probe experiments based x-ray scattering or spectroscopy. These applications focus on the ps to sub microsecond time domain and involve several aspects of dynamics on a molecular scale. These include clusters, nanoscale materials, ionic systems, etc.

The discussion of pump-probe techniques was motivated by the expected large peak flux per bunch of the sources. The scientific impact of other aspects of the source involving the high degree of full spatial (temporal and transverse) coherence and the very high spatial forward spatial collimation are not considered in this report. These most certainly need to be topics at further meetings.

In this initial meeting, the candidate scientific areas were grouped under two major headings. In Group A, we included:

- Ultra fast photochemical processes < 1ps such as
    -Laser-induced chemical reactions
    -Bond-breaking (aftermath dynamics)
    -Charge / electron transfer
    -Photo catalysis
- Structure and dynamics of nanoscale clusters, free radicals etc.
    -Probing clusters, molecular free radicals, and ions produced by laser ablation or photodissociation of suitable precursors

One of the issues discussed which has potential bearing on the impact of the new sources is the question of x-ray induced damage. While x-ray induced damage is often viewed as a nuisance, the new sources can also be viewed as providing a means to elucidate the damage mechanisms and dynamics. Light scattering was mentioned as one potential way to study these effects in model systems. Such studies would involve:

- Laser probe of x-ray induced phenomenon.
    -Damage
    -Electronic rearrangements

In Group B were included:

- Atomic and molecular -scale dynamics
- Time-resolved holography (nuclear motions)
- Imaging using coherent radiation
- Spatially resolved probes on the sub-micron to nanometer scale

In the majority of theses areas, ultrafast IR, optical, or UV lasers are used as an excitation source, much the same as laboratory-based femtochemistry and the hard x-ray FEL is used as a probe. Some of the current scientific areas which could benefit from a hard x-ray probe are:

- Molecular dynamics and surface processes
- Photodissociation and photofragmentation
- Ion mobility and state-selected ion reactions
- Surface dynamics (growth) and related dynamics
- Product state analysis of multiphoton ionization and dissociation dynamics induced by short-pulse lasers

3 Source Spectral Properties

In discussing the possible impact of the FEL-based x-ray source, considerable time was spent identifying relevant source characteristics. As mentioned, this source offers an entirely new regime of beam characteristics that must be exploited if the full scientific potential of the FEL's is to be realized. The Working Group based its discussions on a set of assumed characteristics of the new sources, but better clarification of these new characteristics is necessary. The Working Group set forth a set of assumed characteristics on which to base discussions. It was clear that more clarification is necessary for the community. Based on experience at 3rd generation sources, the better educated the scientist concerning source properties, the more likely the dream can become a technical reality.

The general spectral properties assumed were:

- Full spatial coherence
- Size: 90-100 microns
- Divergence: ~ 1microrad x, y
- Natural energy bandwidth $\Delta E/E$: $\sim 10^{-5}$
- 120 Hz duty cycle. (This provides a much better match between x-ray, laser and detector for pump probe than for 3rd generation sources.)
- 100 fs pulse duration. (What is the time structure?)
- A large spontaneous background

The Working Group felt that certain aspects of the new source could have real bearing on potential applications and need to be addressed. These include:

- Energy tunability
- Capability for large energy bandwidth (~800-eV) which will be required for spectroscopy
- Polarization characteristics of the mode
- 'Complete' spectral output of the device
- Effects of 'convolution' of short pulse with optics. e.g. pulse stretching

4 Other Issues

Detectors, especially fast detectors (e. g. streak cameras) need to be developed. The role of x-ray optics and their effect on the intrinsic time structure of the sources needs to be clarified. For pump-probe experiments there are a number of issues regarding synchronization of the two pulses, whether they are generated by splitting the primary beam or by using two different sources. The latter will most likely require synchronization off of the FEL timing. This appears to be possible.

5 Concluding Remarks

The prospects for new science enabled by the FEL-based source appear very attractive indeed. However, much more discussion is required involving researchers already involved in forefront femtochemistry along with those at the forefront of current 3rd generation topics. Plans were made to hold a similar discussion of the Working Group with an extended community at the 18th International Conference on X-ray and Inner-Shell Processes (X99) to be held in Chicago on August 23 - 27, 1999. The time and date will be announced.

Participants of the Working Group:
Nora Berrah, LBL and Western Michigan University
Lin Chen, Argonne National Laboratory
Steve Pratt, Argonne National Laboratory
Ivan Tomov, University of California, Irvine
P. J. Viccaro, The University of Chicago