Working Group I
Imaging Techniques Summary Report

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The Imaging Techniques subgroup of Working Group 1 was attended by approximately ten people. Three short presentations were made, followed by detailed discussion. The presentations and some highlights drawn from them are given here.

M. Howells of LBNL presented an approach to single-shot three-dimensional (3D) x-ray holography using multiview methods. In this approach, a 2D Bragg structure is used to generate the multiple beams required to record several external-source x-ray holograms of a sample simultaneously over a range of incidence angles with a single FLS pulse. The holograms would then be reconstructed and assembled using the principles of diffraction tomography to obtain 3D images of the sample. The primary motivation for this approach is to acquire enough views for good 3D imaging in a time shorter than that in which radiation damage becomes manifest, i.e., a single FLS pulse. It may also provide an answer to the so-called "Spiller Paradox".

The main challenge of this approach is to develop a suitable diffracting beamsplitter structure with approximately uniform efficiency over all of the diffracted beams. Smaller diffracting features (approaching d functions) give a more evenly distributed field of diffraction peaks, but with less efficiency (more light goes into the zeroth-order). Howells estimates that it should be feasible to obtain 0.1% efficiency in the third diffraction order. On the other hand, an expendable structure is probably required due to the prodigious instantaneous power load posed by the FLS pulse. Lastly, a key problem still to solve is how to redirect these multiple beams back onto sample to record the holograms.

C. Jacobsen of the State University of New York at Stony Brook discussed the application of pump-probe techniques in x-ray microscopy and the key issues involved. Radiation damage to the sample is of concern, since specific chemical bonds break at absorbed doses of ~10^8 Gy and high resolution image fidelity suffers at doses beyond ~10^{10} Gy. However, it was pointed out that such damage can be mitigated more than 1000-fold by cryo-preparation of samples. Current flash freezing technique can achieve freeze rates of ~10^4 K/s in liquid ethane, approaching freeze times of ~1 ms.

Potentially applicable pump-probe methods include visible and x-ray luminescence labelling, time-coincidence fluorescence microscopy, and XANES resonance microscopy. Visible luminescence labels such as fluoroscein and rhodamine still have the drawback that they are not particularly friendly for live biological samples, but their use is not precluded in many cases. Due to the short lifetimes of many fluorescence states it may be feasible to perform time-coincidence fluorescence experiments with an FLS. Lastly, it may be possible to introduce a fluorophore into the sample, then illuminate it with a UV pre-pulse followed by two short x-ray FLS pulses to measure the XANES resonances (see Klems, et al., (1998???).

A short discussion by Howells and Jacobsen followed on multiphoton imaging that brought several points to light: (a) The n-photon absorption cross-section is proportional to the n-th power of the incident intensity, so the key advantages of multiphoton techniques still apply, i.e., optical sectioning and background rejection. (b) Wth the sub-ps time scales possible using an FLS, the probability is nonzero that $n > 1$ photon could be experienced within an atomic core lifetime. (c) The advantage of reduced bleaching using two red visible photons is lost with x-rays.

The last presentation, by C. Fadley of LBNL, dealt with issues in inner-source x-ray holography including polarization effects. Development of inner-source holographic imaging methods is driven by the extremely high (atomic scale) resolution of which it is capable, especially the study of atomic-scale structural problems that cannot be addressed by diffraction experiments such as:

- bond lengths in three dimensions
- epitaxial 8-layers
- atomic dopants and defects
- adsorbates (atomic, without long-range order)
- adsorbates (molecular)
- biological macromolecules
- self-assembled monolayers

Single-energy x-ray holography (SEXH), or the “direct” method, appears to be best suited to atomic imaging with an FLS. The key difficulty with multiple-energy x-ray holography (MEXH), or the "inverse holographic method", is incompatibility with single-pulse holography due to the requirement for sample rotation. A point in favor of MEXH is that the energy resolution required, 5-10%, needs only to be high enough to discriminate the fluorescence lines. With SEXH on the other hand, the twin-image problem and image cancellation ("winking") at certain energies must be handled. Other issues to resolve include deadling with sample anisotropies, nonidealities, and multiple strong-scattering effects, which result in efficiencies of 0.1-0.3%. The relative merits of the two methods are summarized as follows:

constraint	SEXH	MEXH
sample rotation	no	yes
energy resolution	high	low
twin image problem	yes	no
winking problem	yes	no

At present, many hours of data collection are required to record an inner-source x-ray hologram with a third-generation synchrotron source. For example, the CoO crystal sample used by Tegze and Faigel had a mosaic spread of 0.3°. The angular sampling increment used to record its hologram was also 0.3° and the sample rotation increment was 1.5° (continuous motion). Based on these constraints, a total of $\sim 5 \infty 10^4$ data points were collected with $\sim 5 \infty 10^6$ counts each ($2 \infty 10^6$ Hz count rate). Even so, the data acquisition required ~7 hr, primarily due to the limited speed of the scan motors. Specifications for an FLS-based SEXH experiment using a spherical detector to collec the entire hologram, are:

- single shot ~ 150 fs
- counts/shot ~ 10^{10}
- counts/element ~ 10^6
- ΔE ~ 150 eV

It may be possible to use lithographically fabricated arrays of dispersive x-ray optical elements ("micro-monochromators", e.g., composed of crystal segments) combined with a spherical area detector, perhaps with active elements ("PAD arrays"), to obtain both the required spatial and energy resolution.

Polarization effects will also have to be taken into account. For example, use of linearly polarized light can result in some information not being available in the recorded holoram (some scatterers may be invisible in certain geometries). A highly polarized x-ray beam may also prove to be useful. Magnetic holography may be feasible by recording holograms with first left then right-circularly polarized x-rays about specific absorption edges.

Fadley stressed that we should not forget photoelectron production, which will be extremely high with an FLS. This may make energy resolution possible at the ns time scale using time-of-flight methods (see P.M. Len, et al., *Phys. Rev*. **B55**, 3323 (1997)). Lastly, he asked whether we might be throwing away valuable information by low-pass filtering of the hologram before it is reconstructed. This data contains information on the long-range order, planarization, and other features of the sample.

In summary, imaging methods that appear to be the most suitable to use with an FLS are:

- atomic-scale x-ray holography (inner source)
- nano-scale x-ray holography (external source)
- multiphoton methods -- fluorescence, confocal
- direct imaging
- phase contrast, dark-field imaging
- coded-aperture imaging
- contact radiography
- polarized x-ray microscopy

The outstanding issues that must be addressed in order for these to become practical, are:

- sample damage
- precision optics (microfocusing, condensers)
- area detectors, energy resolving, ideally spherical
- background noise (spontaneous radiation)
- beam stability
- coherence (degree required)
- multiple pulses
- pulse gating
- cycling sample media, flow, etc.
- beamline requirements
- hutch (experimental station) requirements
- coherence requirements (temporal, spatial)
- pulse stretching, compression
- flux, brightness requirements
- “fallout” after prompt image is recorded
- better understanding of matter-radiation interactions