Physics Research Opportunities with XFEL's

Sunil K. Sinha

*Advanced Photon Source, Argonne National Laboratory
9700 S. Cass Avenue, Argonne, IL 60439*

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Abstract: The advent of x-ray Free Electron Lasers will present a number of unique new scientific opportunities. These arise from their special characteristics which include intensely brilliant pulses of x-rays delivered in very short times, complete transverse coherence, and high photon quantum degeneracy, amongst other things. While clearly much thought needs to be given to a detailed quantitative assessment of the feasibility of various experiments using such sources, including methods of dealing with heat loads on beamline optics and radiation damage to samples, there are a number of areas in which one can see new opportunities, and other exciting possibilities about which one might speculate. In this talk we shall briefly review some of these areas, such as x-ray holography, pump-probe type experiments, correlation spectroscopy and quantum optics experiments with x-rays.

Successful construction and operation of a hard x-ray free electron laser (XFEL) source will open up many exciting new areas for scientific research, some of which we will attempt to survey in this article. We are still at an early stage in our collective thinking and so many of the ideas put forth here are admittedly speculative. Much more detailed examination of the feasibility of some of these ideas is in order (including realistic assessments of the radiation damage to the samples) and will undoubtedly be undertaken over the next few years, based on data available from the current third-generation sources, detailed calculations of cross-sections, etc., and of course data from the Stanford LCLS when it comes on line. Many of these concepts have already been discussed in the proceedings of several workshops (1-5).

We start by listing the factors that will make the XFEL unique for doing new science. These are: (a) peak brilliance (a factor of 10^{10} - 10^{12} over that at current sources), (b) short time pulses (~ 100 fs), which combined with (a) results in (c) extremely high electromagnetic fields for very short times, (d) extremely high quantum degeneracy of photon states ($\sim 10^{12}$ times that obtainable at present at $1 \text{ \AA}$) (e) completely coherent, diffraction limited beams, and (f) even a time-averaged brilliance which is at least four orders of magnitude higher than that at current sources, particularly at higher photon energies. Details of these order of magnitude increases are given in the paper by Cornaccia in these proceedings (6).

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Perhaps the most obvious feature which can be exploited is the short time-pulse structure which will enable pump/probe studies on time scales hitherto accessible only to laser sources, but with the capability of studying phenomena at much shorter (mesoscopic to atomic) length scales, together with the penetrating power of hard x-rays. (It is perhaps relevant that the Nobel prize in chemistry was recently awarded to Ahmed Zewail for his pioneering studies of femtosecond phenomena with laser pulses.) An intense, short ($\sim$ fs) laser pulse can be used to induce a strain or shock-wave in a material, which can be probed by an x-ray pulse at a fixed time delay afterwards. (The technical issues of how stable and controllable such a time delay can be made are being actively worked on at several laboratories (7)). The x-ray pulse can be used to probe a specific or several Bragg reflections which suffer intensity changes with time as the system loses long-range order and then recovers (8). The higher intensities at XFEL's can also be used to study simultaneously the time dependence of the diffuse scattering between the Bragg reflections, which should answer very pertinent questions as to what sort of defects (strains, dislocations, etc.) develop as time proceeds. It should also be possible to do phase contrast imaging or even topography of the time-dependent development of defects such as cracks, dislocations and grain-boundaries as they develop, move and aggregate in response to the shock wave. Everything stated about shock waves above can also be adapted to the study of the structural and magnetic response of materials to extremely high pulsed magnetic fields, using magnetic charge x-ray scattering, x-ray magnetic circular dichroism, etc., as probes.

Of course, the ultimate dream of x-ray imaging has always been to produce a three-dimensional image of a molecule (such as a protein) with atomic resolution. Fig. 1 shows an empirical curve of imaging resolution obtained by x-ray holography as a function of

FIGURE 1. Plot of Imaging Resolution vs. Average Source Brilliance (from I. McNulty, unpublished)

average source brilliance (9) from which by extrapolation one may expect that at the XFEL levels of brilliance atomic resolution might indeed be possible. However, several problems need to be overcome, such as the fact that the sample might be destroyed by the beam (necessitating getting the full holographic information in one pulse before the sample was destroyed, which in turn necessitates bringing beams to bear on the sample from several directions at once). On the other hand, x-ray "flash imaging" may prove to be the best route to ultra high resolution imaging of, e.g., biological samples in their natural, physiological state. The potential and problems of such x-ray "flash holography" are discussed in more detail by I. McNulty, et al. (10).

Let us now consider dynamical studies (i.e. the study of equilibrium fluctuations in condensed matter rather than non-linear transient phenomena). The area of photon intensity correlation spectroscopy is currently regarded as one of the frontier areas of research at third generation SR sources (11-14), but is often intensity limited at these, owing to the requirement of having an incident beam size limited to the dimensions of the transverse coherence length (typically 5μ horizontally by 50μ vertically at current third generation sources). An XFEL source, with its beam fully coherent in the transverse direction and with even an average brilliance orders of magnitude greater than present sources, has the potential of making such experiments much easier to do with larger coherent illuminated areas on the sample, and of making accessible time-scales (down to picoseconds or less) and wave-vector transfers (out to $\sim 2\text{\AA}^{-1}$) which are currently inaccessible. (For large wavevector transfer experiments, the XFEL beams would have to have their longitudinal coherence considerably increased by suitable monochromation). Again, for soft condensed matter samples particularly, damage issues would have to be carefully assessed, but preliminary calculations (15) based on the full transverse beam rather than a focused beam seem to indicate that this may not be an insoluble problem. For such experiments, short pulses with an adjustable time delay τ between them would have to be delivered to the sample. Depending on the time-scale τ , this could be a train of pulses from the XFEL itself, or for shorter times, a delay-line scheme involving an extra path-length and beam-splitting or rapid beam-switching could be adopted. The dynamics of the samples is extracted from the usual type of expression used in dynamical light scattering, i.e.,

$$\langle I(\bar{Q}, t) I(\bar{Q}, t + \tau) \rangle / \langle I^2 \rangle = 1 + g_2(\bar{Q}, \tau) \quad (1)$$

where $I(\bar{Q}, t)$ is the intensity in a suitable pixel of the detector corresponding to a wave vector transfer $\bar{Q}$ at time t , and $g_2(\bar{Q}, t)$ in a gaussian approximation is often written as the square of the intermediate scattering function $F(\bar{Q}, t)$ or $\langle \rho(\bar{Q}, 0) \rho(\bar{Q}, t) \rangle$ where $\rho(\bar{Q}, 0)$ is the Fourier transform of the electron density at time t .

Another method of measuring $F(\bar{Q}, t)$ quite generally is the Mossbauer filter method (16), which has already been demonstrated at the ESRF, but is again quite intensity limited at current sources. In this method, a pulse is transmitted through a foil containing a Mossbauer isotope (e.g., Fe^{57}) before and after scattering from the sample. The x-rays

are tuned to excite a nuclear resonance in the foils, which then coherently de-excite with a decay probability $g_1(t)$ after the initial pulse, re-emitting the x-ray photons. After the instantaneously scattered pulse from the sample has gone, there is thus a combination of amplitudes reaching the detector consisting of the wave scattered from the incident pulse on the sample and then emitted by the nuclei of the second foil at time t , with amplitude $\propto g_1(t)p(\vec{Q},0)$ and the wave emitted by the nuclei of the first foil at time t and then directly scattered from the sample into the detector, with amplitude $\propto g_1(t)p(\vec{Q},t)$ ($t=0$ is the time the incident pulse was at the sample). The resultant intensity at time t in the detector

$$I(t) \propto |g_1(t)p(\vec{Q},t) + g_1(t)p(\vec{Q},0)|^2$$

and thus consists of a constant plus a term proportional to $\langle p(\vec{Q},0) p(\vec{Q},t) \rangle$ or the intermediate scattering function. Measurements made of the relaxational modes of glycerol in the vicinity of its structure factor maximum showed consistency with a stretched exponential decay with time scales of nanoseconds being measured. With the ~ 100 fs pulses at an XFEL, one may imagine using Mossbauer nuclei with shorter lifetimes and probing time-scales of 300 fs or less.

Yet another method of measuring dynamics is the recently developed technique of x-ray Fluorescence Correlation Spectroscopy (XFCS) (17) which does not explicitly rely on a coherent beam, but instead on a microfocused beam through which particles move in and out. If diffraction limited focusing is required, then the coherence of the XFEL beam would be necessary. By measuring the fluorescence intensity from particular atoms in these particles in a detector and the time autocorrelation function of this intensity, one may extract quantitative details of the dynamics, such as the nature of the motion (e.g., Fickian diffusion, sedimentation or flow, reptation, etc). and the constants involved. Interdiffusion of two different species at a boundary can be monitored by selectively looking at the fluorescence from each species. The method does not directly give $\vec{Q}$ -dependent information, unlike photon intensity correlation spectroscopy. A variation of this method which is able to yield such information and can be used to measure $F(\vec{Q},t)$ has been proposed (18) which relies on a standing wave being set up above an interface or surface (by reflection) which is coherent across the surface. It can be shown that for material inside this standing wave, the fluorescence autocorrelation function (measured in the usual manner) is proportional to $F(\vec{Q},t)$ for the fluorescent species, where $\vec{Q}$ is the wavevector of the standing wave (in general normal to the surface). At current third generation sources, the method is intensity limited so that only the motion of clusters of thousands of fluorescent atoms (e.g., colloids) can be studied by XFCS. With orders of magnitude greater intensity, even the motion of single atoms could conceivably be studied, as in the case of atomic or molecular diffusion across a surface.

There are also many possible applications of both the high photon degeneracies and the high field intensities available with such sources, following along the lines of studies currently carried out with optical lasers under the general heading of "quantum optics",

but extended to much higher frequencies and larger wave vectors (19-22). For instance, it is readily conceivable that the large degeneracies could induce stimulated Raman & Brillouin scattering (20,21). One might then think of a classical macroscopic electromagnetic wave propagating through the sample and exciting a classical or macroscopic wave of the excitation field in the sample, together with another classical frequency-shifted wave. The non-linear response functions of a medium are defined so as to relate the induced electron density response to vector potential fields $A_\alpha(k, \omega)$ given in wave-vector and frequency space by relations of the form

$$\Delta\rho(\bar{Q}, \omega_3) = \chi_{\rho ij}^{(2)ab}(\bar{Q}, \bar{k}_1, \bar{k}_2, \omega_1, \omega_2) A_\alpha(\bar{k}_1, \omega_1) A_\beta(\bar{k}_2, \omega_2) \quad (2)$$

where $\Delta\rho(\bar{Q}, \omega)$ is the Fourier component of the induced non-linear electron density response to the fields $\bar{A}(\bar{k}_1, \omega_1)$ and $\bar{A}(\bar{k}_2, \omega_2)$ and $\chi^{(2)}$ is the second order non-linear density/current response function (20). Here, $\bar{Q}, \omega_3$ must satisfy the conservation laws $\bar{Q} = \bar{k}_1 + \bar{k}_2$ modulo a reciprocal lattice vector and $\omega_3 = \omega_1 + \omega_2$

The above coefficient gives rise to three-wave mixing. 4-wave mixing, given by third-order non-linear coefficients, can also take place and has already been studied for x-rays (23,24). Non-linear processes can also give rise to 2-photon absorption/emission processes, second harmonic generation of x-rays and parametric down conversion. Multi-photon interference and preparation of specific photon number states could also be studied for the first time with x-rays. All such phenomena, barely measurable (if at all) at current sources, could be fully studied with the availability of the XFEL.

The extreme intensities in a focused beam may be utilized in other ways. Thus non-linear effects such as harmonic generation in the sample have been utilized by Piston et al. (25) in obtaining better depth-resolution in x-ray fluorescence microscopy. A beam of photons of frequency ω is focused inside the sample (using, e.g., a Fresnel zone-plate or other focusing device) and generates photons at frequency 2ω only in the immediate vicinity of the focus if conditions are arranged correctly, thus giving full point-resolution if the 2ω photons are used as the signal.

Finally, there are a number of applications in atomic and fundamental physics which can only be mentioned briefly here. They include tests of QED in the non-perturbative relativistic regime (26), achievement of the so-called Schwinger critical field to study pair production from the vacuum (26), study of non-linear stimulated Compton scattering, possible observation of Hawking-Unruh radiation (26), the study of entangled 2-photon states (27), the studies of clusters, multi-photoionization of atoms (important for astrophysics and plasma physics), above threshold ionization (ATI) studies in the x-ray regime (28), etc.

In the brief space available here, it is not possible to give more than a glimpse of the rich possibilities that will be opened up with XFELs. It is imperative therefore that we investigate these possibilities in more quantitative detail. If history repeats itself, it may also turn out that some of the most exciting applications will occur only once such source is available and have not even been conceived of at present.

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