2. Ultrafast, Laser-Based, X-ray Science: The Dawn of Atomic-scale Cinematography

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The characteristics of ultrafast chirped pulse amplification systems are reviewed. Application of ultrafast chirped pulse amplification to the generation of femtosecond, incoherent, 8-keV line radiation is outlined and the use of femtosecond laser-based, x-rays for novel time-resolved diffraction studies of crystalline dynamics with sub-picosecond temporal resolution and sub-picometer spatial resolution is reviewed in detail. Possible extensions of laser-based, x-ray technology and evaluation of alternative x-ray approaches for time-resolved studies of the atomic scale dynamics are given.

Keywords: Chirped pulse amplification, femtosecond pulses, femtosecond x-ray generation, time-resolved x-ray diffraction, relativistic laser/matter interactions, x-ray movies.

1. Introduction

The continued development of new ultrafast amplification technologies has led to the production of a class of laser systems which are simultaneously capable of producing ultrafast (sub-20-fs), ultra-high-peak-power (100-Terawatt) pulses at repetition rates which are 4 to 5 orders of magnitude greater than previously possible.[1-3] The combination of high peak power, high repetition rate and ultrashort pulse duration makes these systems ideal for investigations of novel relativistic laser-matter interactions, and for the production of high fluxes of energetic particles and ultrashort duration x-rays.[4]

Laser-based, 10-keV, sub-picosecond x-ray sources have been used at the University of California, San Diego for investigations of sub-picosecond,

laser-initiated atomic dynamics via time-resolved x-ray diffraction and time-resolved absorption spectroscopy.[5-7] Time-resolved x-ray diffraction has allowed the production for the first time of atomic-scale, ultrafast movies of crystal lattice motion. These movies reveal clearly the propagation of coherent acoustic phonons, non-thermal melting of semiconductor thin films and interfacial energy transfer in layered crystal structures. Such investigations are not presently possible with any other x-ray technology including state-of-the-art, 3rd-generation synchrotron sources.

In this paper, an overview of ultrafast laser technology and x-ray applications will be presented. A brief discussion of possible extensions to much higher x-ray fluxes and more demanding applications will be given.

2.Ultrafast Chirped Pulse Amplification

Modern solid state laser materials have many favorable properties which make them ideal for high peak power pulse generation.[8] In particular materials such at Titanium doped sapphire and Neodymium doped glass have both high energy storage and ultra-wide amplification bandwidths capable of supporting pulses of 10 fs to a few 100 fs in duration, thus allowing in principle one to generate pulses of many Terawatts to Petawatts peak power with relatively low energy.

There is, however, one fundamental problem with respect to generating ultrashort duration, high-peak-power pulses directly from solid state media. In any single pass amplifier the stored energy can only be extracted efficiently if the input pulse fluence (i.e. the energy per unit area) is on order of the saturation fluence of the amplifier material.[9] Materials such as Ti:sapphire and Nd:glass have relatively large saturation fluences (1 J/cm^2 and 8 J/cm^2 respectively) and relatively short potential pulse durations. The Fourier transform of the gain bandwidth of Ti:sapphire and Nd:glass would imply that pulses as short as 3 fs and ~50 fs could be generated with these materials, respectively. If the final amplifier is operated at the minimum pulse duration and at or above the saturation fluence of the material, then the peak intensity of the amplified pulse in the final amplifier would be on the order of 10^{14} W/cm^2. Such intensities are considerably above those necessary to induce optical breakdown of dielectric materials, e.g. ~5×10^9 W/cm^2, and

therefore the final amplification stage would not survive undamaged. The common method of circumventing this fundamental problem is chirped pulse amplification (CPA)[10] in which an ultrashort duration, low energy seed pulse is passed through a dispersive delay line and broadened in time by a factor of between 1000 and 100,000. This low energy pulse is then amplified to an energy which is commensurate with the saturation fluence of the gain media and the damage threshold of the optical materials and coatings in the amplification chain. After amplification the pulse is passed through an inverse optical delay line thus recompressing it to an ultrashort pulse duration.

In practice the longer the input pulse duration is lengthened before amplification, the more efficient the final energy extraction will be. The minimum pulse duration for saturated amplification is determined by dividing the maximum damage intensity of the amplification chain by the saturation fluence of the amplifier material. For Ti:sapphire and a damage limit of 5 GW/cm^2, one must stretch the input pulse to 200 ps in order to run the final amplifier at one times the saturation fluence. At three times the saturation fluence, it is possible to extract more than 90% of the theoretical maximum stored energy of a single pass amplifier. Therefore, larger initial stretching is better. The practical limit to which the duration of the input pulse can be stretched is set by the size of the dispersive delay elements in the pulse compressor. 10-fs pulses compressors are composed of parallel grating pairs. The net delay of such pairs of gratings is set to first order by the linear dimension of the grating in the dispersive plane, e.g. a 30-cm wide grating can be used to compress roughly a 1-ns chirped pulse. Commercially it is hard to obtain gratings with dimensions much above 40 cm, therefore stretched pulse durations are normally much below 1.25 ns. In this respect, the intermediate saturation fluence of Ti:sapphire makes it nearly an ideal CPA material. Higher saturation fluence materials require larger stretched pulse durations for the same percentage of energy extraction from the final amplifier and lower saturation fluence materials will not compactly store as much energy in the final amplifier. Ti:sapphire is also the largest bandwidth solid state amplifier material presently available. It can be produced with excellent optical quality, has very favorable thermal properties and a high damage

resistance. Therefore it is possible to produce 10-fs range, multiple Terawatt to Petawatt pulses at high repetition rate with Ti:sapphire.

The properties and problems associated with 10-fs range CPA are significant and different from picosecond CPA. So much so that it is useful to classify 10-fs range CPA as Ultrafast CPA. Pulses produced by ultrafast CPA have many interesting and unique properties. First of all pulses durations of the order of 10-fs are extremely short. Light travels only 3 microns in 10-fs. This timescale is much shorter than that of atomic vibrational motion in molecules and therefore such pulses have potential for imaging molecular or crystalline dynamics. 10 fs is also of the order of inner shell lifetimes in intermediate atomic weight atoms and therefore such pulses have potential for pumping inner shell x-ray lasers. Because Ti:sapphire is an excellent optical material, it is also possible to produce near diffraction limited spatial quality beams. Diffraction limited beams can be focused to mircon diameter spot sizes and therefore in principle it is possible with ultrafast CPA systems to produce focussed intensities on the order of 10^{21} W/cm^2 or greater. Intensities above $2x10^{18}$ W/cm^2 are considered to be "relativistic". At this level, a free electron will experience a pondermotive energy equal to its rest mass and therefore will be relativistically accelerated on each half cycle of the laser pulse . At 10^{21} W/cm^2, the interaction is highly relativistic and new phenomena such as coherent, Larmor x-ray radiation and astrophysically relevant matter acceleration are expected to be observed.[11] The electric of the laser pulse at this level is many TeV/m or more than 4 orders of magnitude higher than the highest electric field used in state of the art particle accelerators. Also unique to ultrafast CPA systems is the peak energy density which can be achieved at the focus. Because the longitudinal length of a 15 fs pulse is only about 5 microns the volume that the pulse occupies as it is focused to 1 micron spot is of the order of 10^{-11} cm^3. With only one joule of pulse energy, the energy density of at the instant the pulse traverses the focus will be 10^{11} J/cm^3 which is an energy density equivalent to 20 Tons of TNT/cm^3 or a mass equivalent of 1 mg/cm^3.

Perhaps the most important property of ultrafast CPA systems however is that because low energy is required to reach high peak power and because the optical properties of the amplifier crystals are conducive to high

average power operation, it is possible to generate terawatt level pulses at unprecedented repetition rates. In order to generate a 100 TW's of optical power a decade ago required the use of the world's largest laser facility, the NOVA laser at the Lawrence Livermore National Laboratory. This laser consisted of 10 beams lines each capable producing a 10 TW, 1000 J, 1 ns pulse at one shot every hour. When fired simultaneously this laser could produce 100 TWs of optical power. The entire laser system occupied a facility with dimensions on the order of 100m x 30m x 30m. On the other hand, a modern ultrafast CPA system can produce 2-J pulses of 20-fs duration at a repetition rate of 10 Hz or 36000 times greater than technology of only a decade ago. Such a system can occupy an area of less than 10 m^2.

Let us define the product of laser system peak power times laser system repetition rate as the laser system "Utility". It is this product that will ultimately determine the flux of particles and x-rays generated from laser/matter interactions and it is this product that will determine what level of signal averaging may be employed in the investigations of relativistic laser/matter interactions. The "Utility" of ultrafast CPA systems is tremendous and more than 4 orders of magnitude greater than any other laser technology. Because of this high utility many new applications are now possible. There are many important technologies which have gone into the development of ultrafast CPA systems, including higher order phase control, regenerative pulse shaping, thermal lens management. [1, 3, 12-23] We will not treat these in this paper but will instead concentrate on the emerging applications of ultrafast CPA technology, in particular ultrafast, laser-based, x-ray science.

3.Ultrafast X-ray Diffraction

One particularly exciting application of ultrafast CPA systems is the generation of high fluxes of ultrafast, incoherent x-rays via relativistic laser/matter interactions.[24] Because of their potential sub-ps pulse duration, such x-rays are potentially ideal for time-resolved studies of molecular and crystalline dynamics. To understand fundamental dynamic processes in physics, chemistry and biology it is necessary to observe atomic motion on atomic temporal and atomic spatial scales. To capture one frame of atomic

motion via x-ray diffraction, we will therefore need ~100-fs, ~10-keV x-rays. With such a source it will be possible to obtain a sequence of stop action pictures of atomic positions which can then be integrated into a movie of the atomic process of interest. At the University of California, San Diego we have investigated a number of laser initiated processes in crystalline materials with ultrafast, time-resolved diffraction.[5-7, 25-27] In each case we use a laser-driven, copper K-alpha radiation source.

The generic interaction which we use is similar to that first described by Kmetec et al. We focus a 20-fs, multiterawatt pulses at a 20 Hz repetition rate onto a moving copper wire target. The 20-fs pulses are accompanied by a low intensity, nanosecond duration, pulse pedestal which is due to amplified spontaneous emission from the amplification chain. Even though the intensity in this pulse pedestal is 5 orders of magnitude smaller than the peak intensity at the focus, it is still sufficient to pre-ionize the target before the arrival of the main pulse and thus to produce a low density plasma in front of the target. The main pulse is focused to an intensity $>10^{18}$ W/cm^2 by an inexpensive, diamond turned off axis parabola. At these intensities, free electrons in the pre-plasma are accelerated to energies up and beyond 1 MeV. These energetic electrons are then incident upon the target and where they produce x-ray inner-shell vacancies and subsequent x-ray line radiation. The electrons also produce Bremsstrahlung radiation. Bremsstrahlung x-ray radiation of >1 MeV has been reported previously from interactions with laser pulses of as little as 0.3 TW peak power.[24] A typical x-ray spectrum of the radiation produced in the 4 to 20 keV spectral region with a 25 fs, <100 mJ laser pulse is shown in Figure 1. The spectrum is obtained using an x-ray ccd camera in "single-photon per pixel" mode. The spectrum is dominated by the copper K-alpha and K-beta radiation. At 20-Hz repetition rate, 5×10^{10} photons per second per 4π steradians are produced in the K-alpha line and 75% of the radiation between 4 and 20 keV is contained in this line. The spectrum is therefore non-thermal in origin. To be "thermal" would require an unrealistically high black-body temperature, i.e. a radiation temperature for which the total radiated energy would exceed the input laser energy. Conversion efficiency from laser to k-alpha x-rays is on order of 10^{-5}. It is

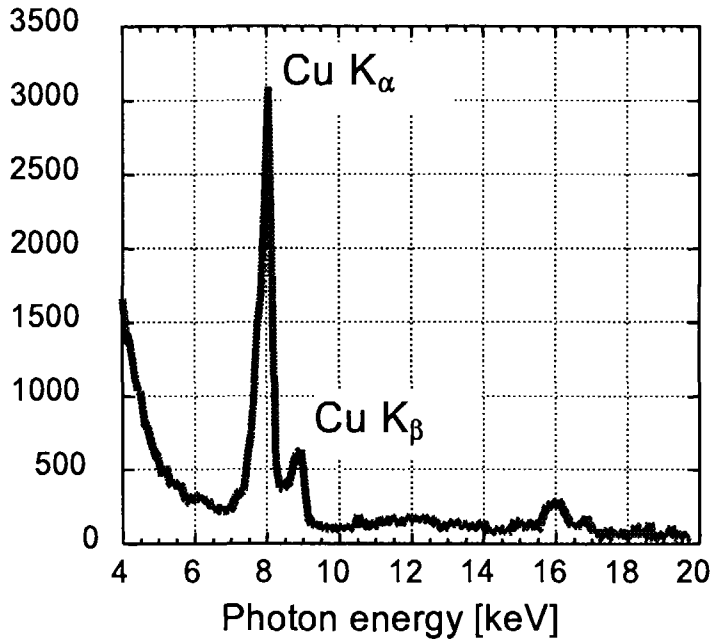

Figure 1. X-ray spectrum from copper wire target illuminated with 25 fs, ~100 mJ pulses.

believed that the x-ray pulse duration is simply the convolution of the laser-driven electron pulse duration, the electron stopping distance in the material and the lifetime of the k-shell vacancies which produce the x-rays. Since the 1/e escape depth of 8 keV photons from copper is of the order of 20 microns and since the multiple keV to MeV electrons are only accelerated to relativistic velocities during the peak of the laser pulse, it is believed that the x-ray pulse duration is of the order of 100-fs. Direct measurement of the duration of 8 keV x-ray pulses with 100-fs resolution is not presently possible and therefore we must rely on indirect measurements and observations.

One scenario under which the pulse duration might be longer than the convolution mentioned above would be if there were energetic, e.g. >10 keV, electrons which streamed away from the target and latter re-collided with the target. If such electrons were present, then the x-ray spot size would be larger than the laser spot size and the pulse duration of the x-rays would be considerably longer due to the long transit time away from and back to the target. In order to evaluate this possibility a simple imaging test was performed, in which the laser-generated k-alpha x-rays were used to pin-hole image a fixed target. The results of this test are shown in Figure 2. The fixed target is a 16 micron spaced copper mesh. From the resolution present in this

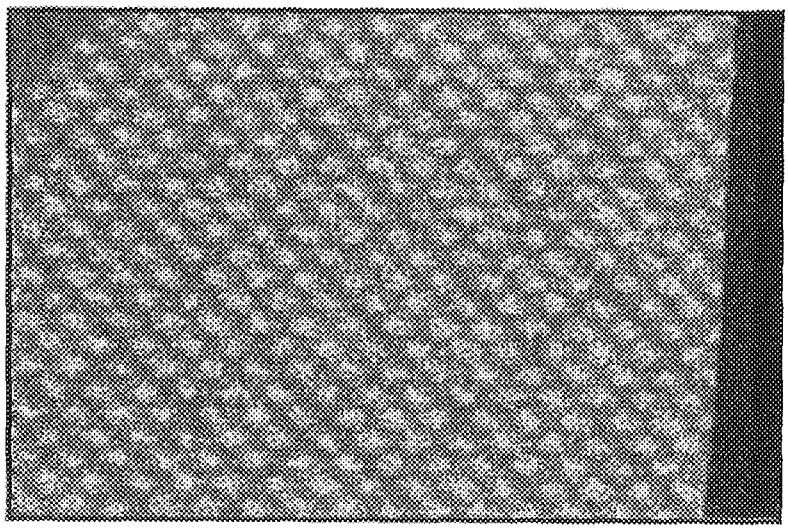

Figure 2. 8 keV X-ray shadowgraphy of 16 micron copper mesh

image it is possible to conclude that the x-ray source size is nearly identical to the laser spot size and therefore little, if any, back streaming of electrons is present. As we will see later, our x-ray source is easily able to resolve physics occurring on a picosecond timescale. If one were able to construct a target which was as thin or thinner than one k-alpha escape depth, i.e. 20 microns, then in principle the duration of the k-alpha x-rays in the forward direction of the laser pulse would simply be the convolution of the drive laser pulse duration and the lifetime of k-shell vacancies or on the order of 20 fs.

A schematic of a simple time-resolved, x-ray diffractometer arrangement is given in Figure 3. In this arrangement a portion of the x-ray generating laser pulse is split-off, sent through an adjustable optical delay line and used to rapidly heat a crystalline target. K-alpha x-rays are then incident upon the target in a symmetric Bragg configuration. The diffracted x-rays are collected with an x-ray CCD camera and stored on a personal computer for later analysis. Good quality crystals, e.g. GaAs, Si, Ge, were used in order to increase the diffracted signal.

When the pump beam is turned off, a static diffraction image is obtained. In Figure 4 the static diffraction image from 111 GaAs is illustrated. The exposure for this image is 2 minutes. There are approximately 500,000

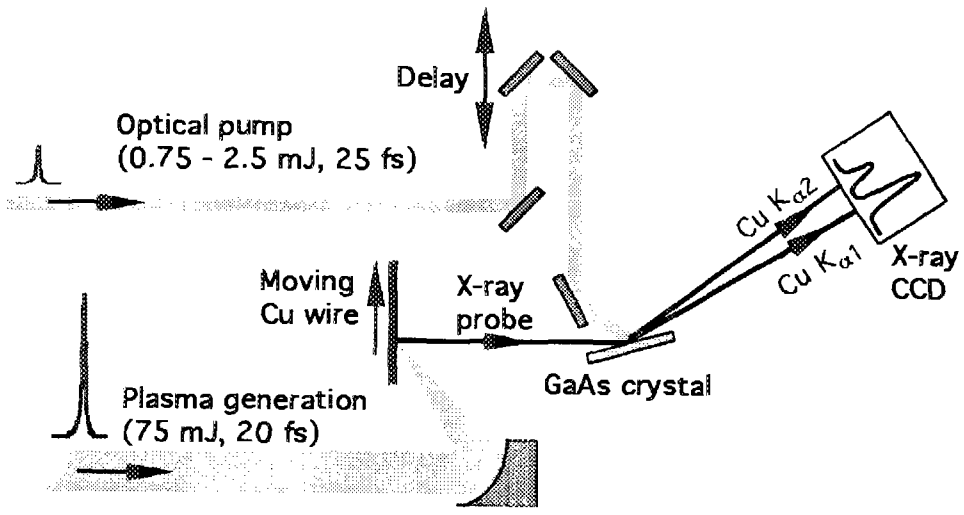

Figure 3. Schematic of time-resolved x-ray diffractometer

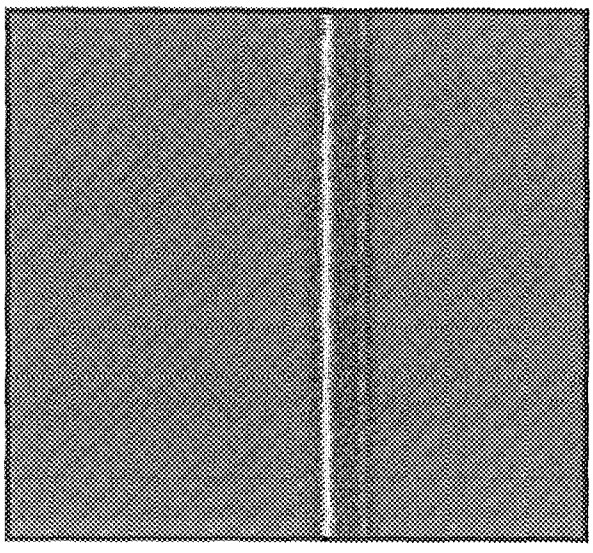

Figure 4 Static diffraction image from 111 GaAs. 2 minute exposure

diffracted photons used to produce this lineout and the resolution is high enough to clearly resolve the fine structure split k-alpha1 and k-alpha2 lines. The exact line shape of the diffracted signal is a function of the crystal quality and the quality of the x-ray source. In particular, if one includes the measured x-ray spot size, the known x-ray linewidth, then one expects to see the diffracted signal indicated by in Figure 5. The measured and expected lineshapes show excellent agreement. The important point to take from this figure is that the linewidith and source size of the x-ray source matter greatly.

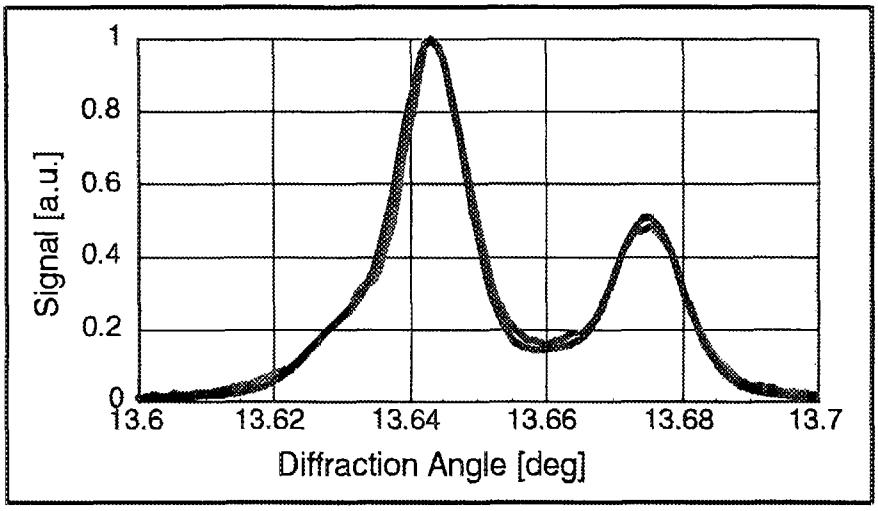

Figure 5. Measured and calculated lineshapes using the measured spot size.

It will be small shifts of these lines which determine the dynamic lattice spacing information. In the case of laser heated GaAs, one expects to generate coherent acoustic phonons which propagate into the crystal lattice.[28] These phonons produce slight changes in crystal lattice spacing . For a point source of x-rays, as the lattice spacing changes, the position upon the crystal at which the Bragg condition is satisfied changes. Larger spacing will shift the lines to smaller relative angle. The story is complicated by the fact that the pump laser penetration depth is less than the depth to which the probing x-rays penetrate. Nonetheless it is possible to obtain quantitative temporal and spatial information using this technique.

Figure 6 illustrates three separate time steps during which diffraction signals were accumulated. Note that the region of the crystal which is illuminated by the heating laser is small. Diffracted signal from regions outside the illuminated region can be conveniently used to normalize the "pumped" diffracted signal at each time step. At early times we see that the diffracted signal is split from 2 lines into 4 lines. To first order the peak lattice spacing change can be determined by the maximum shift of the diffracted signal. In this case the maximum shift corresponds to a lattice spacing change of only 0.3% or 10 mAngstroms! It is possible with well established models for coherent acoustic pulse generation and propagation to predict the exact

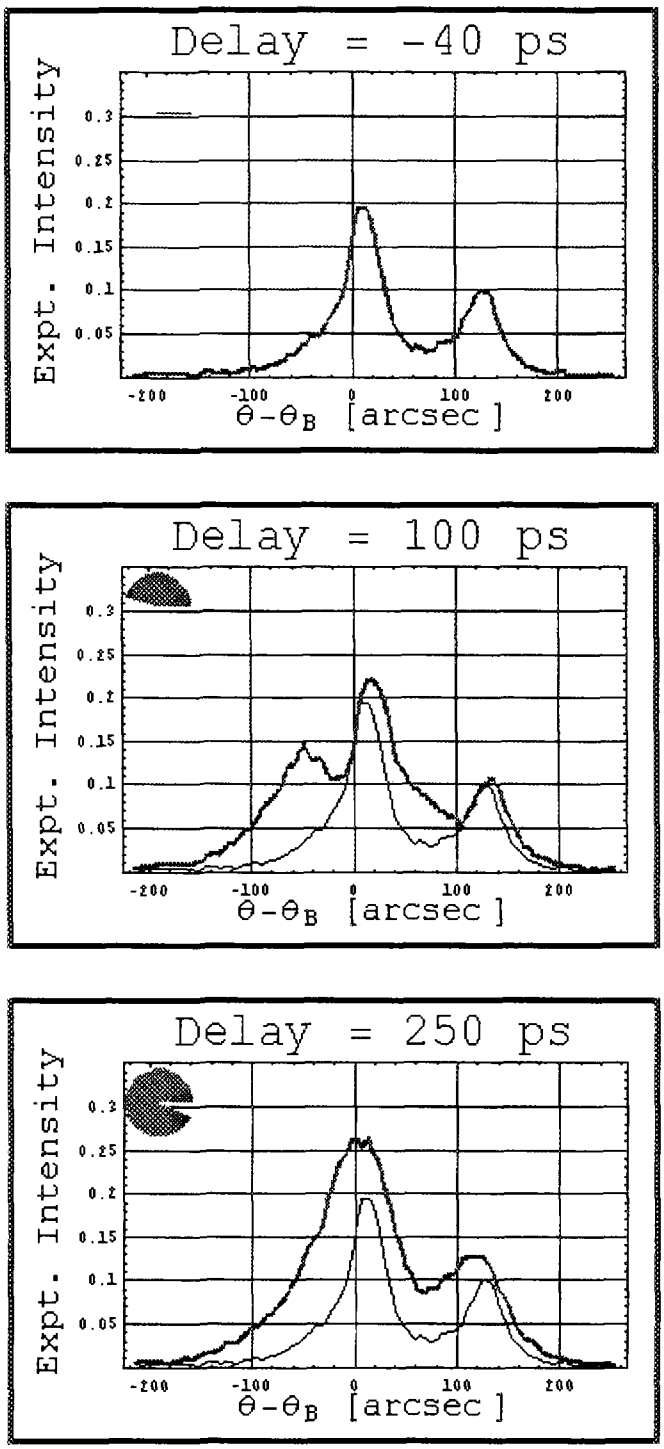

Figure 6. Lineouts from three separate time delays.

lattice spacing distribution as a function of depth and time using only the measured initial peak shift and the known values for the pump light absorption coefficient and known values for the speed of sound in GaAs.[5] From these predictions it is straightforward to then calculated the expected

diffraction signal and compare it with the measured signal. A comparison of the expected and measured signals is given in Figure 7. As can be seen, there is remarkable agreement. Clearly laser-based, time-resolved diffraction can determine ultrafast crystalline dynamics with picosecond accuracy and milliAngstrom spatial resolution.

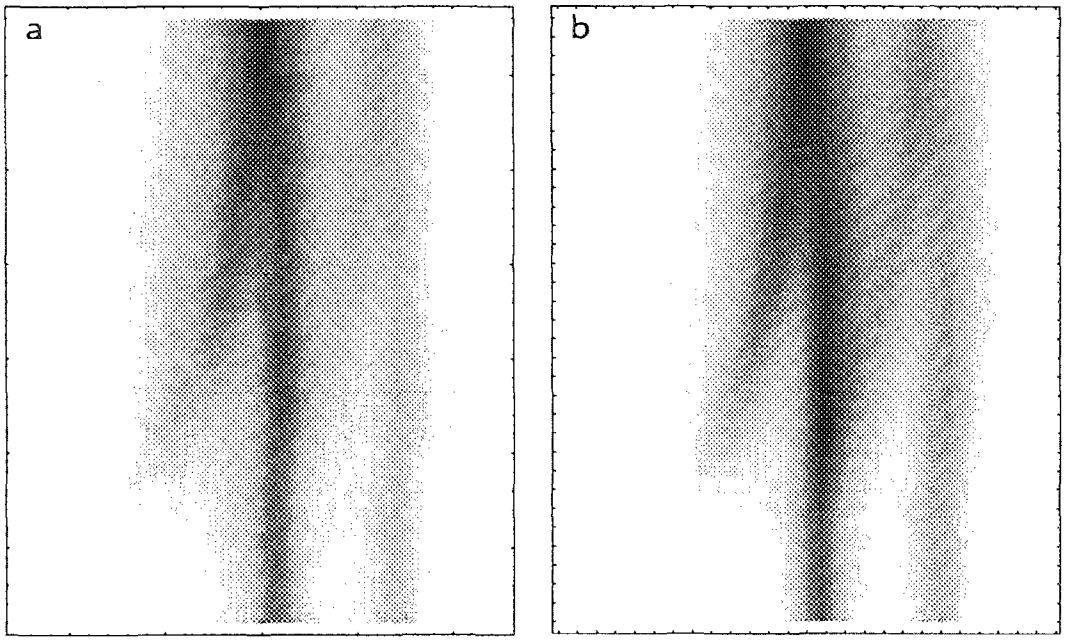

Figure 7. Grayscale measured (a) and calculated (b) time-resolved diffraction from 111 GaAs. Note the horizontal axis is diffraction angle and the vertical axis is time from minus 40 ps to plus 250 ps.

As another illustration of the power of this technique, time-resolved diffraction was used to determine how crystalline samples melt when illuminated by an intense laser pulse. There are two possibilities. The sample can absorb energy at the surface creating a thin super heated liquid region whose front then propagates into the material OR intense incident radiation may promote enough carriers into the conduction band that the entire region into which the laser light is deposited becomes unbound and thus instantaneously dissociates. The former is considered to be thermal melting and is an inhomogeneous spatial effect the latter is considered to be non-thermal melting and occurs homogeneously. There is evidence[29] using time-resolved, visible light reflection that non-thermal melting occurs for

sufficiently fast and intense laser illumination, however, such measurements can only probe a small region (one skin depth of the reflecting light) near the surface of the material and therefore cannot say anything about the bulk crystal state. Time-resolved x-ray diffraction on the other hand can be used to see directly the bulk lattice condition.

In order to provide a uniformly heated region, a thin semiconductor sample was prepared which consisted of 150 nm of 111 Ge on a 111 bulk Si substrate. Since the illuminated region is damaged by the laser on each shot, it was necessary to raster the sample as the x-ray data is collected. 100's of shots are required for each time step, therefore the size of the sample determines how many time frames can be acquired. In our case approximately 10 time steps can be obtained from one wafer. Figure 8 shows the raw diffraction data at 4 different time steps. Note that the laser illuminated region is small. Because the film is thin it is no longer possible to resolve the k-alpha1 and k-alpha2 split lines. Figure 9 is a plot of the diffracted signal from the center of the pumped region and the edge of the

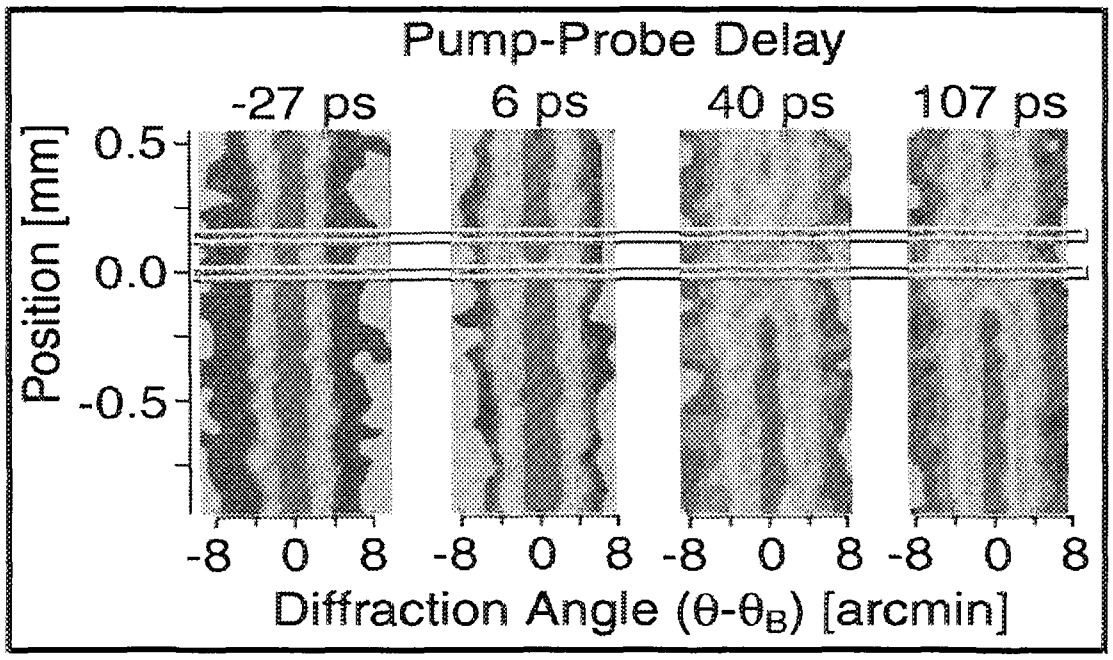

Figure 8. Diffraction images at different time delays. Note the lower rectangle corresponds to the center of the pumped region and the upper to the edge of the pump region

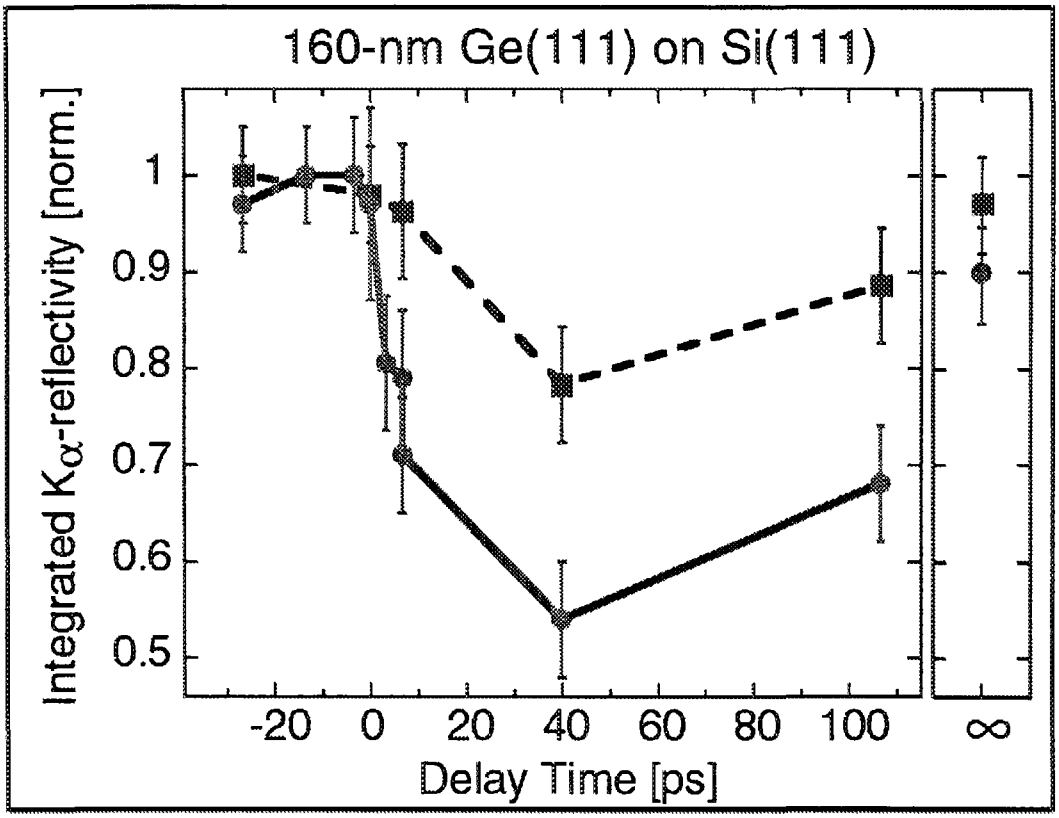

Figure 9. Magnitude of diffraction vs. time for the center of the pumped region (solid curve) and edge of the pump region (dotted curve)

pumped region immediately adjacent to it. From this plot it is possible to conclude that the sample has undergone homogeneous, non-thermal melting. The argument is as follows. The center region shows an instantaneous drop (to within our time step accuracy) in diffraction efficiency consistent with a homogenous or non-thermal melting process while the diffraction signal from the "edge" region drops only monotonically as the melting front propagates outward. Both regions decrease monotonically after the initial heating, as the melting front propagates in all three dimensions. Later in time both regions begin to re-crystallize and diffraction increases. At infinite time after the initial heating pulse, the non-laser-illuminated region returns to its initial diffraction value while the laser-illuminated region returns to a value slightly below its initial value. This slight decrease is due to the evaporative loss of material from the pumped region.

As a final illustration of the potential power of the laser-based time resolved diffraction, the thin film samples were also illuminated at an intensity which was below that which would induce melting but high enough for coherent phonon generation. Since the film was thin, diffraction from both the Ge and the Si could be obtained simultaneously. In this way it is possible to watch the expansive acoustic wave hit and reflect from the Ge/Si interface. The position of the centroid of the normalized diffracted signal from the pumped regions is illustrated in Figure 10. In this Figure it is clear that the diffracted signal from the expansive strain produced in the outer thin film undergoes a marked change in magnitude at exactly the time predicted by the speed of sound in Ge and thickness of the thin film. At the time of this reflection, the expansive wave in the Ge layer produces a compression of the underlying Si substrate as illustrated in Figure 10. It is interesting to note the difference in vertical scales of in this Figure. The magnitude of the compressive strain that is observed in Si corresponds to a lattice change of only 20 femtometers! Such measurements of buried ultrafast dynamics are not possible presently with any other method.

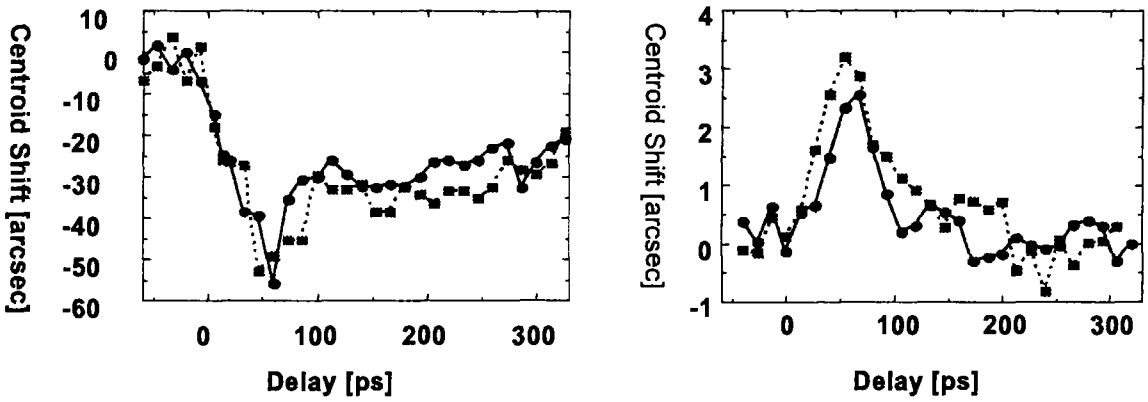

Figure 10. Shift of the centroid of the diffraction lines for Ge film (left plot) and Si substrate (right plot). Note the difference in vertical scales. Doted curves are k-alpha1 and solid curves are k-alpha2 data

Future laser-based x-ray diffraction investigations may allow direct observation of optically driven phonons, single shot observation of laser driven shocks, and time resolved studies of amorphous solids and liquids.

Optically driven phonons should produce lattice vibrations which are sub-picosecond in duration and therefore may yield information about the exact duration of the laser-generated x-rays. These experiments also can be arranged to be zero background, i.e. there will be no diffracted signal unless the optically driven phonon is present.

Single shot investigations of laser driven shocks should be possible if the x-ray yield is improved. Empirical evidence suggests that the x-ray yield scales as the 3/2 power of the input energy.[24] Using 100-TW class lasers it may be possible to obtain enough x-ray line radiation to produce a high resolution diffraction signal on a single shot. A generic experiment would place a thin layer of material which is to be compressed on top of a good quality crystal such as Si or GaAs. The change in diffraction from the underlying material would be used to monitor both the magnitude and speed of the shock propagation. In this way it may also be possible to directly measure the photon pressure of the incident, ultrahigh intensity pulse.

Investigations of amorphous solids and liquids will require both higher fluxes of x-rays and detectors which are able to subtend a larger solid angle. The diffraction signal from these samples will be qualitatively similar to that from a randomly oriented crystalline powder. High energy Bremsstrahlung radiation which is produced concurrently with the production of line radiation must be controlled in these low signal to noise experiments. 100 keV and higher energy radiation is easily generated during relativistic interactions and can be sufficiently energetic to cause traditional Pb shielding to fluoresce. Such fluorescence background can significantly decrease the signal to noise of the collected diffraction signal. Figure 11 shows the static diffraction signal obtained with an amorphous Ni target, illuminated with x-rays which are refocused by an grazing incidence ellipsoidal reflector. By refocusing the x-rays it is possible to move the sample sufficiently far from the source that high energy background radiation may be reduced with a low-Z/high-Z filter arrangement. The final diffraction signal is collected with an 80 mm diameter, spatially resolved photon counting detector.

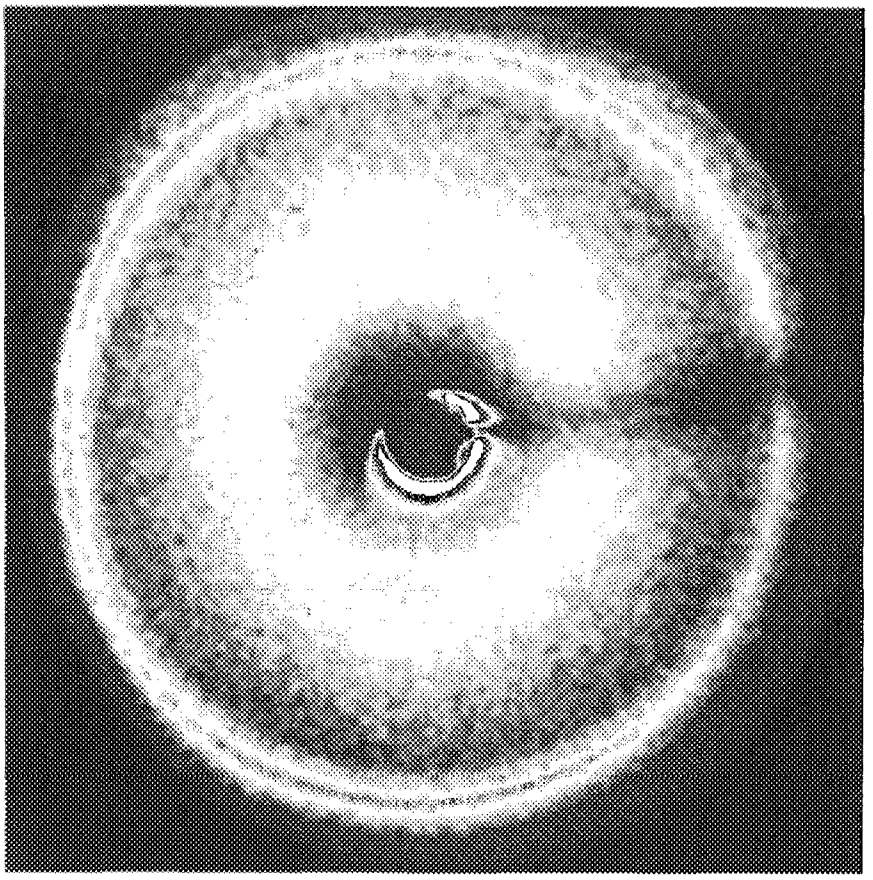

Figure 11. Static diffraction from an amorphous Ni foil using 8 keV x-rays and an ellipsoidal reflector and spatially resolved photon counting detector. The intense signal at the center is unblocked, direct radiation from the target. The full diameter of the image is 80 mm and the exposure is 5 minutes using a 4 TW, 20 Hz repetition rate laser.

4.Other X-ray Sources and Improvements in Laser Based Sources

The laser-based, x-ray source used in the UCSD time-resolved diffraction experiments had a number of very useful properties. The linewidth was very narrow, of the order of 10^{-4} $\Delta\lambda/\lambda$. The source size was extremely small, approximately 10 microns. The combination of small source and narrow linewidth allowed high resolution diffraction. The wavelength was commensurate with atomic spacings, i.e. in the 10 keV energy range. The potential pulse duration was on the order of 100 fs. Finally it produced x-rays at a repetition rate which was commensurate with the optical pumping of the sample, i.e. at 20 Hz. Nominally it takes mJ's of energy to excite all the atoms in a sample probed by 10 keV x-rays, i.e. a sample of several tens of

cubic microns in volume. Providing sufficient excitation energy at rates above a kHz will be difficult. Considering all of these parameters, the laser-based, k-alpha source appears to be an ideal choice for performing time-resolved diffraction studies.

In principle there are a number of alternative short pulse x-ray technologies which could be employed for time-resolved diffraction experimentation. Of these laser-Thomson scattering, 3rd generation synchrotron radiation and radiation from x-ray free electron lasers are the principle alternatives.

In laser-Thomson scattering, an intense, femtosecond laser pulse is scattered off of an energetic electron pulse.[30] The scattered laser photons are upshifted into the x-ray regime. Advantages of this technique are that the pulse duration can be a few 100 femtoseconds, the exact duration being set by the crossing time of the laser pulse and the electron pulse. The x-rays are reasonably collimated and the energy of the x-rays is tunable by varying the energy of the electrons. These sources, however, also have some rather severe drawbacks with respect to their use in x-ray diffraction studies. First of all the scattered radiation is broadband, set by the energy spread of the electron bunch. The bandwidth of the Thomson x-rays is at least 100 times that of the laser-based, Cu k-alpha source. In the diffraction studies discussed above, it would not be possible to observe milliAngstrom and smaller motion with this linewidth. Secondly, the efficiency of the scattering process is small, thus much longer accumulation times are needed for each time step. The typically electron bunch frequency at most electron storage ring facilities is much in excess of 1 kHz. Lasers of sufficient intensity have not yet been constructed with high enough repetition to utilize the full electron bunch frequency. Finally, synchronization of laser which pumps the sample which is probed with the Thomson scattered x-rays will require careful locking of its repetition rate to some harmonic of the RF which drives the storage ring. Achieving ps accuracy is difficult and will require locking to better than one part in 1000 of the RF frequency.

3rd generation synchrotrons are purpose built machines for producing x-ray radiation. These machines presently produce the highest average brilliance x-rays of any source and do so by several orders of magnitude.

With respect to laser-based, Cu k-alpha sources, the average brilliance of the ESRF synchrotron in Europe is at least 20,000,000,000 times greater at 8keV. Average brilliance, however, is only one consideration when evaluating the usefulness of the source for time-resolved diffraction studies. 3rd generation synchrotrons are broad band sources. In order, to produce the same linewidth as a laser-based, Cu-k-alpha source would require the use of a additional x-ray monochrometer. Even if the monochrometer were 100% efficient the relative advantage of the synchrotron would be reduced by a factor of 10. Next the pulse duration of the synchrotron radiation is typically on the order of 100 ps and not 100 fs. Assuming that one could arrange to slice a 100 fs pulse from the 100 ps initial pulse with an efficiency of up to 1%, the relative advantage of the synchrotron would be further reduced by another factor of 10^5. Finally the pulse repetition rate of the synchrotron radiation at ESRF is very high, of order 350 MHz, and not 20 Hz. Pumping the sample with a few mJ's of optical energy at 350 MHz is not possible. If one considers doing experiments at 20 Hz with the ESRF synchrotron, the relative advantage will be reduced by another factor of $2x10^7$. Combining all factors, a 20-Hz, time-resolved, x-ray diffraction experiment can be performed on the order of 1000 times faster with a laser-based, Cu-k-alpha source than with present 3rd generation synchrotrons. Furthermore, the use of synchrotron radiation for ps time-resolved, laser-pump/x-ray-probe experiments will once again require synchronization of the pump laser pulse with RF of a storage ring to better than one part in 1000. It is interesting to note also that this comparison is made between a laser-based source requiring 2 watts of optical power to generate x-rays while the ESRF synchrotron requires 2 MW of RF power to maintain the electrons in the storage ring.

At least two proposals currently exist for producing single pass x-ray amplification from an x-ray FEL, one using DESY in Germany and one using SLAC at Stanford University. Of these only the Stanford University Linac Coherent Light Source (LCLS) proposes to produce 10 keV light in the near future. If funded and built on schedule and to specification, the LCLS will produce in 2004, sub-100-fs, 10-keV x-rays with 10 orders of magnitude higher peak brightness than presently possible. This source holds great promise for a number of time resolved studies as well as for macromolecular

crystallographic investigations. It, however, will be a unique and expensive ($100,000,000 not including the 15 GeV accelerator) device and will, like all accelerator based setups, require synchronization to better than one part in 1000 with respect to the linac RF if visible-pump/x-ray-probe studies are to be conducted with better than 1 ps relative accuracy.

For the near future, laser-based, x-ray sources seem to have clear advantages for pump/probe x-ray diffraction experiments. In addition to incoherent line radiation, 10-fs range, high-peak-power lasers can be used to generate ultrafast bremsstrahlung x-rays from solid targets, ultrafast line radiation from interactions with cold atomic-clusters[31], ultrafast semi-coherent radiation via relativistic interactions, e.g. larmor radiation, and possibly even coherent keV radiation from inner-shell x-ray laser schemes. In all cases, the ultimate useful flux of x-rays will increase as the "utility" of ultrafast CPA systems increases.

Possible routes to improved CPA utility include, shorter pulse duration, higher pulse energy and higher system repetition rate. Reductions in pulse duration are currently limited by the bandwidth of high damage threshold optics. Presently the widest bandwidth commercial optics are only capable of supporting 12 fs pulses. The shortest duration CPA pulses produced to date are 16 fs. Increases in pulse peak power will be limited by the size and damage threshold of gratings used in the final pulse compressor of the ultrafast CPA system. Meter diameter gratings should allow the production of between 10 and 100 Petawatt, 15 fs pulses without damage. Increases in pulse repetition rate are limited by the availability of high energy, high repetition rate pump lasers. This factor has probably the most room for improvement. Present pump lasers used in ultrafast CPA systems have average powers of order 10 Watts, while commercial industrial lasers can operate at 10's of kW's. Improvements in pump laser technology will come from both diode pumping of the pump laser gain media and the use of phase conjugation or active beam shaping to control thermal distortions. Taken together, it is not unreasonable to assume that it may be possible to increase ultrafast CPA "utility" by 6 or 7 orders of magnitude over present levels. This combined with the fact that present laser-based x-ray sources and experimental arrangements have only been minimally optimized to date,

would suggest that laser-base x-ray science and atomic-scale cinematography has a very exciting future ahead of it.

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