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Dennis Slaughter, Paul Springer, Greg Le Sage, John. Crane, Todd Ditmire*, Tom Cowan+
Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94550

Scott G. Anderson, James B. Rosenzweig
UCLA Department of Physics and Astronomy, 405 Hilgard Ave. Los Angeles, CA 90095

Abstract:

The use of ultra fast laser pulses to generate very high brightness, ultra short (fs to ps) pulses of x-rays is a topic of great interest to the x-ray user community. In principle, femtosecond-scale pump-probe experiments can be used to temporally resolve structural dynamics of materials on the time scale of atomic motion. The development of sub–ps x-ray pulses will make possible a wide range of materials and plasma physics studies with unprecedented time resolution. A current project at LLNL will provide such a novel x-ray source based on Thomson scattering of high power, short laser pulses with a high peak brightness, relativistic electron bunch. The system is based on a 5 mm-mrad normalized emittance photoinjector, a 100 MeV electron RF linac, and a 300 mJ, 35 fs solid-state laser system. The Thomson x-ray source produces ultra fast pulses with x-ray energies capable of probing into high-Z metals, and a high flux per pulse enabling single shot experiments. The system will also operate at a high repetition rate (~ 10 Hz).

INTRODUCTION

The use of ultra fast laser pulses to generate very high brightness, ultra short (10^{-14} to 10^{-12} s) pulses of x-rays is a topic of great interest to the x-ray user community. In principle, femtosecond-scale pump-probe experiments can be used to temporally resolve structural dynamics of materials on the time scale of atomic motion. The development of sub-ps x-ray pulses will make possible a wide range of materials and plasma physics studies with unprecedented time resolution. The goal of this work is to develop such a novel x-ray source that will exhibit some very important features:

- Ultra fast pulses for probing dynamics on the time scale of atomic motion
- Hard x-rays capable of probing into high-Z metals
- High x-ray flux per pulse enabling single shot experiments
- Widely tunable x-ray wavelength

Bragg and Laue diffraction provide detailed information about the long-range order of a material. Short pulse length and high single-shot flux will allow these techniques to be applied on the time scale

* *Current address: University of Texas, Physics Department, Austin, TX 78712*
\+ *Current address: General Atomics, P.O. Box 85608, San Diego, CA 92186*

of phase transitions, or melt driven by fast heating or compression. In addition, information on the short-range order in a material (such as nearest neighbor distances and coordination numbers) may be obtained using extended x-ray absorption fine structure (EXAFS) spectroscopy[1]. The concept is illustrated in Figure 1.

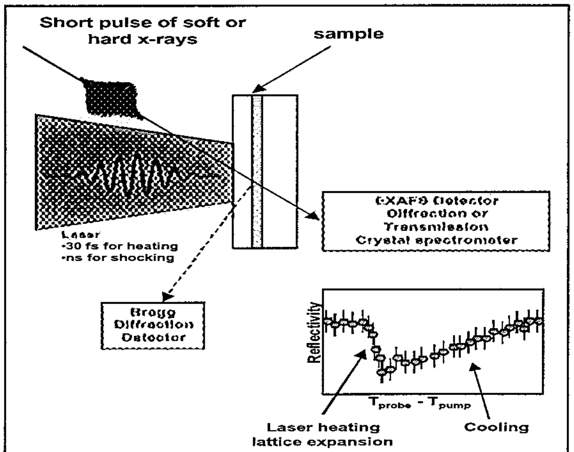

Figure 1: Schematic of a laser-x-ray pump-probe experiments.

Initial ultra fast dynamic studies of laser excited materials have been pursued in recent years using time-resolved x-ray detectors at synchrotron sources and with laser-plasma x-ray sources in pump-probe experiments. Third generation synchrotrons have been critical sources for x-ray studies in materials, but they have a major limitation: although they probe structure on the atomic length scale, their time resolution (~ 100 ps) is not well-matched to the natural dynamics of elemental processes in solids, such as the time scale for atomic motion (~10-50 fs). Laser melting in semiconductors may involve excitation, electron-phonon coupling, melt front motion and shock waves, all with < 30 ps relevant timescales. Notable recent successes have included the measurement of the internal strain profile in InSb following laser heating[2], using Bragg scattering to detect the 1–D compression of a crystal following laser irradiation. The experiments accumulated data averaged over a very large number of shots and could effectively utilize the x-ray flux available from synchrotron sources. However, the time resolution of these experiments was limited to greater than a few picoseconds.

Rose-Petruck, et. al.[3] recently characterized the propagation of femtosecond laser driven coherent phonons in a GaAs crystal using synchronized, laser plasma produced K-alpha radiation and later used a K_α source to time resolve dynamics in laser heated thin Ge layers[4] where they observed melting. Rischel et al.[5] used a similar technique to time resolve dynamics in an organic Langmuir-Blodgett film heated by a femtosecond laser. These studies were limited to time resolution of 1 to 100 ps and were, like the synchrotron experiments, limited to repetitively pulsed data collection.

The time resolved Bragg diffraction technique has been successfully applied on large-scale lasers with shock pressures of ≥ 1 Mbar. Wark et al. [6] performed some of the first, groundbreaking diffraction experiments on the Janus laser about ten years ago. More recently Remington and Kalantar et al.[7] have applied this technique to experiments on the Nova laser at LLNL.

We have undertaken the development of an x-ray source based on laser scattering off a relativistic electron bunch. This Thomson scattering approach offers the potential to produce x-rays in a unique regime. A temporally compressed laser pulse is focused onto a short relativistic electron pulse as shown in Figure 2 below.

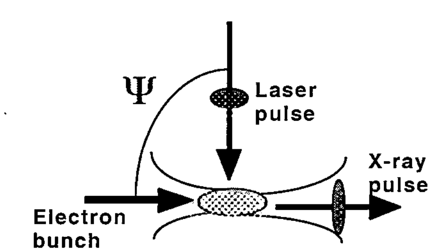

Figure 2 Scattering geometry

DESIGN AND PERFORMANCE

Ultra fast x-ray pulses are generated by scattering a high power, ultra fast, 800 nm laser pulse from a beam bunch of relativistic electrons at the LLNL 100 MeV electron Linac. Beam characteristics in several operational modes are summarized in Table 1 below. The scattered laser photons are relativistically up shifted in frequency into the hard x-ray range, and are emitted in a narrow cone about the electron beam direction. Leemans, Schoenlein and coworkers working at the LBNL Advanced Light Source injector Linac have previously demonstrated generation of-sub-ps pulses of hard x-rays by Thomson scattering in 1996[8, 9]. They were able to achieve an x-ray beam flux of ~10^5 photons in a ~300 fs pulse, at ~30 keV. However, the limited x-ray flux available in that first demonstration required the averaging of several thousand shots at each pump-delay time point. The LLNL Thomson source presently under development is expected to achieve an x-ray beam flux some four to five orders of magnitude larger, enabling the accumulation of sufficient data for a high quality Bragg diffraction spectrum on each shot.

As viewed in the frame of the moving electrons, the incident laser pulse train appears as an electromagnetic undulator of wavelength $\lambda_u=\lambda_L/\gamma(1-\cos\Psi)$, where $\gamma=E/m_oc^2$ and Ψ is the incident angle between electron and laser beams as shown in Figure 2. The electrons radiate photons, which are up shifted back into the laboratory frame by a second factor of 2γ. The x-ray wavelength is therefore related to the initial laser wavelength by

$$\lambda_x=\lambda_L/2\gamma^2(1-\cos\Psi)$$

where λ_L is the laser wavelength. In the laboratory frame, the up shifted x-rays are confined to a narrow cone with opening angle $\sim 1/\gamma$, and their energy varies with observation angle in the laboratory due to the kinematics of the Lorentz transformation. The laboratory x-ray energy (for scattering of the fundamental) is:

$$E_x=E_L\,2\gamma^2(1-\cos\Psi)/(1+\gamma^2\theta^2+a_o^2)$$

where E_L is the laser photon energy. Here, a_o is the usual normalized vector potential of the laser field, which is analogous to the K parameter of a static field undulator. For our parameters, the x-ray energy should be tunable over a range from 10 to 200 keV in the laboratory depending on the incident angle Ψ and initial electron beam energy. The number of x-ray photons produced is proportional to the strength of the effective optical undulator field (for $a_o<1$) and the number of optical cycles (N_o), by

$$N_x=(\pi/3)\alpha_f N_o N_e a_o^2$$

where α_f is the fine structure constant and N_e is the number of electrons in the beam bunch.

In comparison to the 10^5 x-rays per pulse observed by Leemans & Shchoenlien[8, 9], we expect to produce up to 10^8 x-ray photons in a ~100 fs pulse, and up to 10^{10} x-rays in a 1-10 ps pulse, by scattering the laser respectively either across the electron beam ($\Psi= 90^o$) which minimizes the temporal overlap, or in a head-on ($\Psi=180^o$) geometry, which maximizes the interaction of the electrons and photons. To achieve the predicted enhancements over the demonstrated LBNL Thomson yield, we are making two straightforward but critical improvements:

- Low emittance electron beam produced from a photoinjector
- High power 10 TW Ti:sapphire laser (with planned upgrade to 100 TW)

Predicted beam characteristics are shown in Table 1 below.

Table 1: Predicted Characteristics of the LLNL Thomson Scatter X-ray Source
(assuming 100 TW operation of Falcon)

	Short-pulse mode	*Long-pulse mode*
X-ray flux	10^9 photons per pulse	10^{11} photons per pulse
X-ray pulse duration	50 – 100 fs	<10 ps
X-ray energy	10 – 100 keV	20 - 200 keV (head-on)
Bandwidth	< 10%	< 10%
Divergence Angle	30 - 10 mrad	30 - 10 mrad
Peak Spectral Brightness	10^{21} (ph/s/mm^2/mrad^2/0.1% BW	10^{21} (ph/s/mm^2/mrad^2/0.1% BW
Electron bunch	1 nC in 1 ps	10 nC in 10 ps
Electron energy	30 – 100 MeV	30 – 100 MeV
Laser pulse[#]	4 J in 35 fs (currently 300 mJ)[#]	4 J in 1 ps (currently 300 mJ)
Laser Intensity	~10^{18} W/cm^2	~10^{18} W/cm^2

The LLNL Falcon laser[10, 11] currently produces 300 mJ pulses at repetition rate 10 Hz that are 5X more energetic than the 60 mJ pulses used by the LBNL group, and with their duration as short as 35 fs are nearly three decades brighter without further improvement. Anticipated laser upgrade to 4 J coupled with increases in electron bunch charge will extend this advantage to 4-5 decades.

The Falcon laser is a Ti:sapphire laser based on chirped pulse amplification. To date, we have integrated the Falcon laser with the Linac and made preparations to transport the laser pulses to the electron beam. Initial characterization of this source using 20 mJ laser pulses and 5 MeV electron bunches from the photoinjector at 10 Hz successfully demonstrated 0.6 keV x-ray production. This demonstrated the ability to focus the laser and electron beams to small spots (<10 μm), and to synchronize them (< 10 ps).

The estimated fluxes, summarized in Table 1, were predicted using the treatment of Esarey et al[12] assuming that the Falcon laser is upgraded to 100 TW operation. We have designed an interaction geometry in which roughly one x-ray photon is emitted per electron, and therefore predict 10^9 to 10^{11} x-ray photons per pulse, depending on the bunch charge and the interaction angle. In fact, the estimates in Table 1 are based on a somewhat idealized laser pulse that is assumed to be uniform (both spatially and temporally) over the laser-electron overlap region. With appropriate laser pulse shaping, maintaining $a_o < 1$ while optimizing the laser-electron overlap with, for example, a moving focus, we should be able to achieve this level of x-ray generation for different interaction geometries. For example, by injecting the laser pulse at $\Psi = 90^o$ to the electron beam direction, we will attempt to achieve x-ray pulses as short as ~100 fs with as many as 10^9 photons per pulse scattering off of a 1 nC bunch. These parameters are most relevant to experiments on ultra fast dynamics in solid-state systems, for example the electron-phonon relaxation process which governs the transfer of energy from x-ray or optical pulses to crystals, or for chemical reaction studies, in which the bond breaking and formation times are of order ~10 fs.

This is a significant improvement over existing 3rd generation light sources where the pulse widths attainable are usually long, typically greater than 100 ps. For fast dynamics, these pulses are of limited utility. On the other hand, laser produced x-rays via hot plasma production offer the potential for producing sub-picosecond x-rays. However, achieving very high peak brightness is difficult through this means. The Thomson scattering source we are developing at LLNL will yield peak brightness that exceeds those of 3rd generation synchrotrons while delivering x-ray pulses with pulse width below 1 ps. The comparison is illustrated in Figure 3 below.

[#] *Falcon laser currently generates 300 mJ pulses, is capable of 600 mJ, and a planned upgrade will increase the pulse energy to 4 J*

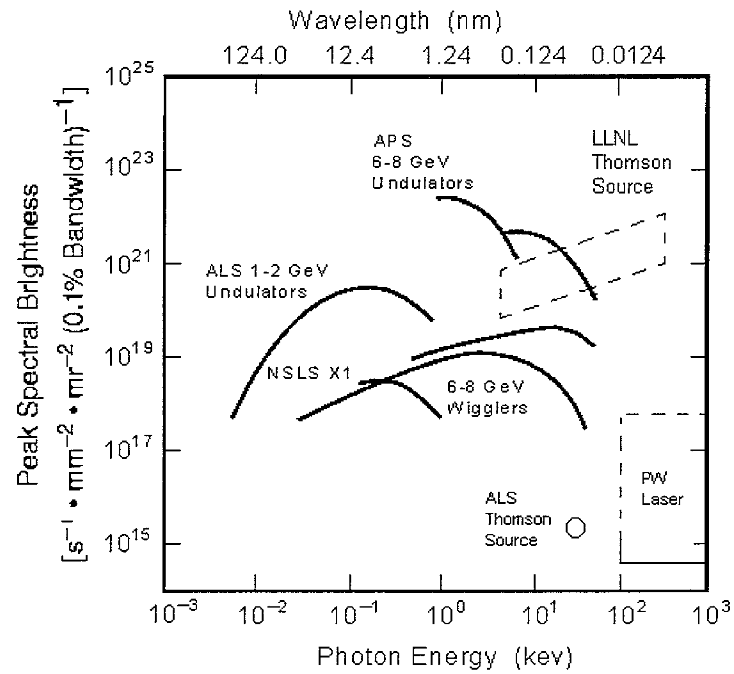

Figure 3: Comparison of the predicted peak brightness of the LLNL Thomson source with current 3rd generation synchrotrons.

LLNL LINAC ELECTRON INJECTOR

Our design is based on a RF photocathode electron injector as the key element for production of a high peak current, low emittance, short pulse, well synchronized electron beam. This photoinjector, shown schematically in Figure 4 below, has been designed, constructed, and characterized.

A pulse of S-band (2.86 GHz) RF input with 7 MW peak power and 3 μs pulse length produces a standing wave electric field with peak gradient greater than 100 MV/m. It accelerates electrons to 5 MeV over a distance of less than 10 cm. A laser pulse, split from the Falcon laser oscillator is amplified and frequency tripled before striking a copper photocathode near the peak RF field to produce the electron injector beam with 1-10 nC of charge per pulse in a <10 ps pulse length. A strong magnetic field matches the beam envelope into the Linac and preserves the low transverse emittance inherent in the high-gradient photoemission process.

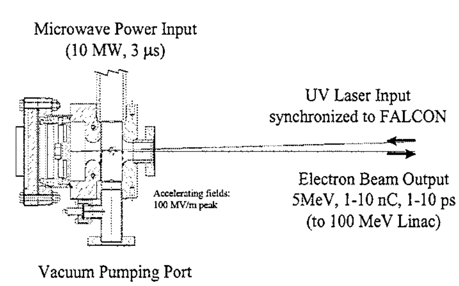

Figure 4: Schematic of the photoinjector electron beam source

Incorporation of the photoinjector into the Linac provides the needed low emittance, high charge electron bunches, and importantly, facilitates synchronization of these bunches with the compressed laser pulse. This essential to attaining the performance predicted in Table 1.

ELECTRON PULSE COMPRESSION

At high energy (100 MeV) the minimum focal spot obtainable for the electron beam is emittance dominated for bunch charge in the nC range. Simulations of the electron beam emittance have been carried out using the PARMELLA code and results have been used to predict beam performance for the Thomson scattering experiments. The results predict a low emittance beam that can be focused to a 10 μm spot with a convergence angle of 5 mrad. To accomplish this the required normalized transverse emittance is 10 mm–mrad. This requirement is well within the measured emittance of the photoinjector for a charge of a few nC. In a strong focusing field, low energy spread is also important to maintain a short longitudinal width at focus. The photoinjector can produce an energy spread of a few percent.

The pulse energy will be chirped by a dephased Linac section so that magnetic dispersion can be used to shorten the pulse. A complementary dephased Linac section removes the energy chirp after compression for maximum beam peak current. Achieving a very high peak current is most important for the transverse interaction of the Falcon laser pulse and electron beam. Ultimately, the goal is to place as many monoenergetic, collimated electrons as possible into the laser focus for the production of up–shifted x-ray photons.

We will install a magnetic chicane electron bunch compression system after TW section 4 of the Linac, as shown in Figure 5. This will enable shorter, more intense x-ray pulses. We expect to achieve an approximately ten–fold longitudinal compression of the electron bunches, with a corresponding ten-fold increase in peak current. This translates into a ten–fold increase in the x-ray flux for the short-pulse, 90° scattering geometry. For the 180° geometry, the x-ray flux will remain the same (determined by total bunch charge), but the x-ray pulse length will be reduced from 10 ps to ~1 ps.

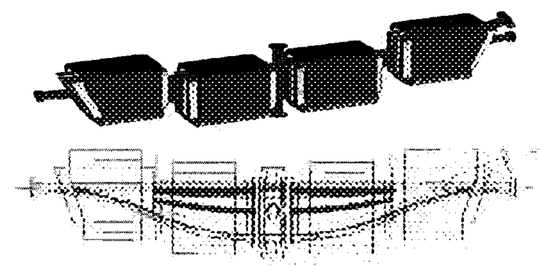

Figure 5. Schematic representation of an 80 MeV pulse compression chicane capable of > 1 kA operation with subpicosecond electron pulses.

Hartemann, et.al.[13] have developed a 3-D model for the beam, its interaction with the laser, and subsequent x-ray production. It has been used to predict the x–ray beam characteristics in this system and one result is shown in Figure 6 below. The predicted x-ray output is very intense and relatively narrow in wavelength (~ 10%).

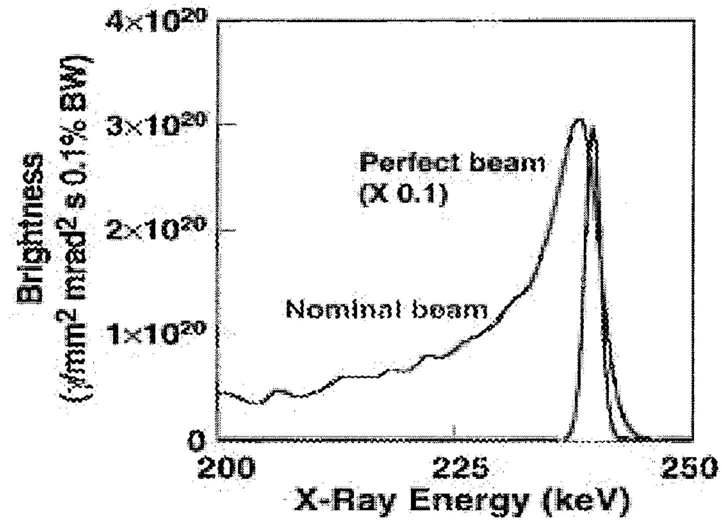

Figure 6. The spectral brightness of the LLNL Thomson source is modeled using the 3D electron/laser interaction model of Hartemann. Showing the expected beam pattern and the peak spectral brightness.

DISCUSSION

Integration of the laser and electron beams has been demonstrated and we will soon commission the photoinjector on the main Linac. Simulations predict a very bright source of x-rays, but that has not yet been demonstrated.

We propose to extend the experiments previously conducted with low time resolution on large–scale systems to the LLNL Thomson source. For the experiments proposed, we intend to use some of the energy of the Falcon laser pulse to drive a shock wave (blast wave). With the Falcon at the 4 J level, we will split off 1 J of energy and focus it to 1 mm on the sample to be shocked. Simulations using the Hyades hydrodynamics code of the compression and pressure as a function of time have been conducted for both Si and Al targets. These simulations indicate that we should be able to produce shocks of pressure >100 kBar. This is adequate to access the Hugoniot-Elastic Limit (HEL) in materials like Si (which has an HEL at 54 kBar). Furthermore, this shock-driving beam is a single spatial mode supergaussian pulse, which will aid in achieving good shock uniformity.

We will perform a simple EXAFS experiment in a mid-Z element such as Ag (K-edge at 26 keV) with a similar, follow-on experiment in a high Z material such as U (with the L-edge at 21 keV). We will start by conducting static EXAFS on unexcited materials. This will allow us to ascertain the brightness needed. These measurements will be followed by simple experiments in which the sample is mildly shocked; to look for shock induced shifts of the EXAFS features.

SUMMARY

A high power (10 TW) short pulse (35 fs) laser has been integrated with a 100 MeV electron Linac to provide a Thomson scattering source of x-rays tunable in wavelength over the range 20-200 keV. By using a photoinjector for the Linac driven by the same short pulse laser it is possible to provide well synchronized 1-10 nC electron bunches only 1-2 ps in duration with very low emittance so that very small focal spots may be obtained. When properly focused the scattered beams are predicted to provide an unprecedented high brightness (up to 10^{11} x–rays per pulse) in durations as short as 1-2 ps, and slightly lower intensity in durations as short as 100 fs.

The 10% bandwidth, tunable wavelength range, and high per–pulse flux of the Thomson source will make it well suited for performing absorption edge spectroscopy of metals at a variety of wavelengths. The 10%

bandwidth will be ideal to backlight an edge and the spectral features near it (spectra within a few hundred eV are necessary for EXAFS). Analysis of EXAFS spectra potentially achievable on the Thomson source indicate that, under the best conditions, EXAFS spectra will be possible on a single shot (if the Thomson source is operated in the high flux, longer pulse 180° mode). Even if the highest x-ray yields are not achieved, multi-shot experiments in which a few shots are averaged will be possible (with the added difficulty of maintaining a nearly uniform shock on every shot).

The goal of this work is to develop a short pulse high intensity x-ray source and demonstrate that EXAFS is indeed possible with the Thomson source.

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