X-ray flash microprobing of structural dynamics in exploding crystals

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ABSTRACT

We describe a new technique for investigating transient structural changes in solid materials involving short pulses of hard x-rays emitted by laser produced plasmas. Using a subpicosecond terawatt scale laser system, this technique promises to be simple and broadly applicable to both single-crystal and polycrystalline samples of thickness up to millimeters and mean atomic number up to approximately 30. Spatial resolutions of order 10 μm should be possible. Of particular interest are the so-called energetic materials, chemical high explosives, whose initiation and reaction dynamics in relation to physical microstructure are poorly known.

Keywords: laser plasma, x-rays, backlighting, point projection imaging, Laue diffraction, shock waves, explosives.

1. INTRODUCTION

Recently, development of ultrashort pulse optical lasers and high bandwidth solid-state amplifiers has resulted in relatively compact systems capable of multiterawatt power levels[1]. Plasmas formed by focussing these laser pulses onto solid targets were discovered to radiate prompt hard x-rays at extremely high brightness[2,3]. We believe that a number of new techniques can be generated quite naturally from this discovery, for probing transient density and microstructural variations occurring in solid density materials of low to medium atomic number. In particular, the propagation of shock waves and the initiation of detonation wave fronts in organic explosive materials promises to be an ideal candidate for study. The recent episodes of American terrorism using explosives made from common materials emphasizes the need to develop an understanding of the small-scale morphology of exploding reaction fronts in the hope of finding inexpensive and nonperturbative reaction inhibitors.

The significance of these new hard x-ray laser-plasma sources is that they combine three unique characteristics. They are small in size (5 μm is reasonable), they are very bright (of order 10^{19} photons/mm^2•s•mrad^2 into 0.01% bandwidth at 5 keV, see Fig. 1), and the x-rays are hard (out to MeV, with reasonable fluences for single-shot imagery at 10 keV). Each of these attributes is substantially different from the radiation emitted by plasmas generated using longer-pulse high energy lasers such as YAG and Nd:glass lasers, because the laser-matter interaction physics is very different for very short pulses. Long pulses produce plasmas with outflowing electron density profiles having very shallow gradients; the dominant absorption mechanism is inverse bremsstrahlung, and the energy is thermalized by electron collisions. Laser focal spot sizes are generally large (> 50 μm) with these systems, so the thermal energy is distributed among many ions. The result is, except for the very largest lasers, plasma electron temperatures of only hundreds of eV, and x-ray spectra falling off exponentially in intensity above 1 keV (following the blackbody curve). In contrast, short-pulse lasers (that emit clean single pulses without prepulse) interact with plasmas having very steep electron density fronts, whose 1/e scale lengths are comparable to an optical wavelength. This changes the

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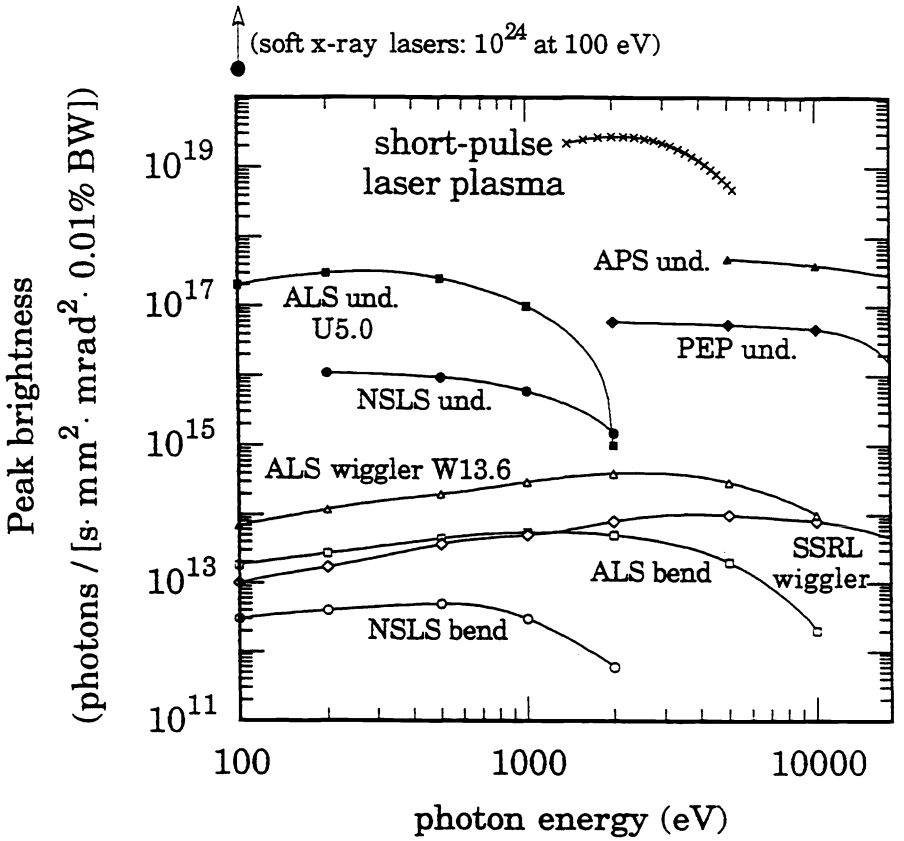

Figure 1. Some of the world's brightest x-ray sources. A comparison of the brightness figures between short-pulse laser plasma sources (data from Ref. 2, assuming blackbody) and large synchrotron facilities (ALS: Advanced Light Source, APS: Advanced Photon Source, PEP and SSRL: Stanford Synchrotron Radiation Laboratory, NSLS: National Synchrotron Light Source).

dominant absorption mechanism to resonance absorption, generating much higher energy electrons. As well, the optical pulse has an electric field gradient which is very steep, and this gives rise to a "ponderomotive potential" which accelerates the electrons into the target surface. The effect is like a high-current (10^{16} A/cm^2) electron beam at 10 keV or higher energy bombarding a tiny spot on the target. The bulk of the x-ray emission consists of high-energy continuum bremsstrahlung. So we have in these sources a unique microscopic "flashbulb" of penetrating radiation. An obvious use of these sources is therefore to take "snapshots" of the interiors of objects.

X-rays are sensitive to mass density through absorption and to atomic order through diffraction. We outline in this paper two techniques derived directly from these interaction properties, respectively, point projection imaging and micro-Laue crystallography.

2. POINT PROJECTION IMAGING

Imaging of experimental objects by the point projection backlighting technique has been a longstanding tool in studies of dense plasmas, in particular for investigations of plasma opacity[4] and hydrodynamic flow interactions[5] associated with inertial confinement fusion and weapons physics. Our extensions of these ideas center on imaging larger, higher density objects, with better spatial and temporal resolution, because of the unique character of the short pulse plasma x-ray sources as

discussed above. The basic concept of the technique, however, is quite straightforward and is schematically pictured in Figure 2. A very small bright source of x-rays is created by tightly focussing a short pulse of laser light onto a solid target. In a line with this source spot is the sample which is desired to be imaged, and a large format spatially-resolving detector (which could be, in the simplest case, x-ray sensitive photographic film). What is seen by the detector is a magnified shadow image of the sample, the magnification given by the ratio of the source-detector distance to the source-sample distance. Typical magnifications are of order a few tens, so that a 50 μm resolution element size at the detector scales to a few μm at the sample, spatial resolution at that point being determined by the backlighter spot size. The image is much the same as a conventional x-ray radiograph, the small source size allowing for the preservation of image detail in the long projection of the shadow. Like a radiograph, each point on the image results from a ray passing through the bulk material of the sample. The ray intensity is determined by the integrated absorption experienced along its trajectory. Under proper conditions, small changes in density of a particular part of the sample such as those caused by the passage of a shock wave are seen as direct (proportional) changes to the transmitted ray intensity. The short duration of the x-ray backlighter pulse effectively freezes motion, even in rapidly changing samples.

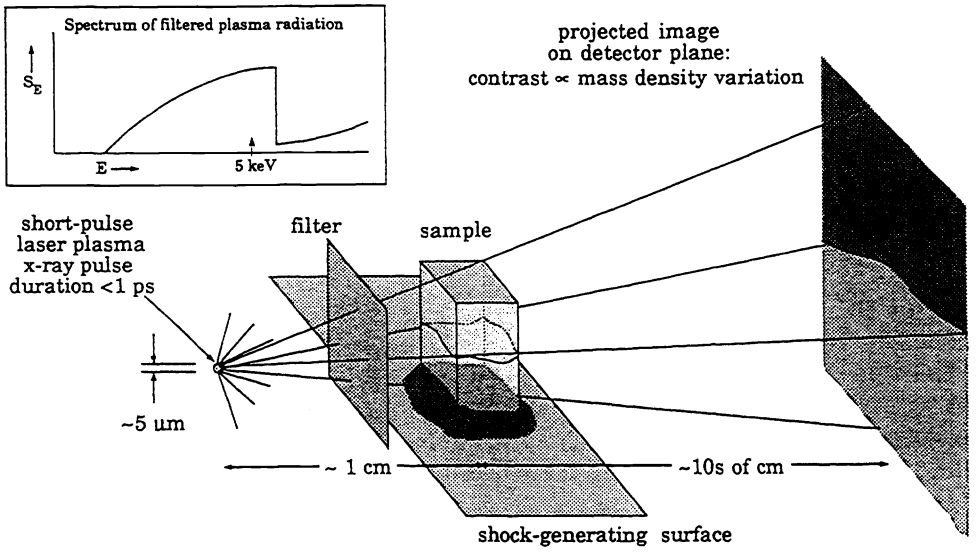

Figure 2. Schematic of the point projection imaging technique using short-pulse laser produced plasma backlighters.

Previous generations of backlighters, those involved in the high energy density plasma experiments, involved large-scale laser facilities whose beams were limited in concentration at focus to of order 10^{16} W/cm^2, with large (50-100 μm) focal spot diameters and tens of picoseconds duration. Two serious limitations are consequences of these conditions. First, to achieve good spatial resolution, one needed to use a localized backlighter target such as a fine wire tip, overfilling with laser light and effectively throwing substantial amounts of the laser energy away. Secondly, the irradiance and pulse duration resulted in a thermalized electron energy distribution at kilovolt or subkilovolt energy, hence yielding an x-ray output dominated by low energy emission. However,with enough laser power to overcome the poor conversion efficiency, plus sophisticated x-ray imaging technology, backlighters have been successfully used to image shock waves in submillimeter solid density plastic samples[6]. We believe that the engineering scale of such experiments can be downsized from the Nova laser by at least three orders of magnitude in size and

cost to systems such as our CREOL Cr:LiSAF laser (see Section 4 below), which fit into a 1-by-5 meter footprint, simply by virtue of their natural proclivity to create pointlike hard x-ray sources.

With short-pulse laser plasmas, the broadband high-energy x-ray emission spectrum must be tailored to optimize signal contrast in order to be useful in detecting small density changes in the sample. In Figure 2, a Cr or V edge filter maximizes the 5-6 keV band for passage through thin organic samples. The choice of x-ray energy is made by requiring around 50% transparency through the thickness of material being studied; higher transparency decreases contrast to small density variations, while higher absorption robs projection signal photons. For millimeter thick organic samples, around 10 keV would be optimum, for example. If sufficient source intensity can be obtained at 20 keV, samples such as glasses, silicon, or even steel are possible.

Also, the self-luminosity of exploding materials at these high photon energies should be negligible. Temperature increases in the sample in the course of an explosion are far smaller than required to emit hard x-rays. Hence multiple scattering of the probe photons, which is a resolution-limiting characteristic of optical techniques, is rather naturally mitigated against. Effectively, the x-ray source is spectrally much "hotter" (i.e. bluer) than the object's own emissions. Even an object which is luminous and blindingly bright in the visible spectrum should be able to be probed at ~10 μm resolution using flash x-rays. The mean x-ray opacity of the sample should remain about that of the cold, pre-exploded material, around 50%.

A single backlit image basically gives a two-dimensional map of areal densities across the sample. By taking a separate, temporally simultaneous and spatially orthogonal view using a second backlighter beam and target, fully three-dimensional mass density data can be extracted for the whole sample.

Measurements of the shock propagation velocity can be made by firing a close-by pair of backlighters sequentially in time, with a sufficient delay imposed (hundreds of ns) for the shock front to move. Gated framing cameras can easily shutter open and closed on such time scales so as to record each image separately. A "strobe" of sequential x-ray pulses can be fired and images recorded on an x-ray streak camera. Since the x-ray flash duration is so brief, blurring effects of the electrons sweeping in the streak tube are minimized with this approach.

We envision the first experiments involving shocked material to incorporate shock initiators such as direct laser-irradiated impulses as well as laser-driven flyer plates. We have at the Laser Plasma Lab a 20J Nd:glass laser capable of launching multigigapascal compression waves into solids, which can be synchronized to the TW laser for precise relative timing. Additionally, wire detonators are a likely future initiator because of the practical similarity to field hardware, with timing being the important technical hurdle to be addressed. The work of Wark and collaborators[7] on laser-shocked crystal surfaces suggests that 6-10% planar compressions can be routinely launched into silicon; using this number as a benchmark, signal-to-noise figures of 50-100 will be required to visualize the shock wave. This performance level for backlighters is challenging but certainly feasible. The Livermore brute force backlighter data in Reference 6 clearly displays the advancing shock front in solid density plastic samples.

3. MICRO-LAUE CRYSTALLOGRAPHY

We intend to pursue, in addition to the various imaging techniques outlined above, a new time-resolved, spatially selective diffraction technique to examine bulk crystallographic structure during the passage of a shock front. A collimated beam of filtered, but essentially continuum x-rays is directed to the sample. Under ideal circumstances, this x-ray beam strikes the sample preferentially favoring a bright Bragg reflection plane parallel to the shock front, though this is not a necessary condition. In the example shown schematically in Figure 3, the setup is similar in several ways to that of point

projection imaging, except that irradiation is limited to a localized region of ~10 μm umbral diameter, determined by the source size (~5 μm) and a pinhole adjacent to the sample, here shown as a single crystal. The x-rays are filtered to a broad band around 5 keV. The source-sample distance has been reduced to ~1 mm and the sample-detector distance to ~1 cm. The crystal is assumed to be an organic material of thickness ~100 μm. Most of the pencil beam of x-rays passes straight through the crystal, making a saturated spot on the detector. However, a number of atomic planes match the Bragg condition and reflect pencils of narrow energy slices out to diffracted angles away from the straight through. Because the source has a broadband spectral character, even modestly precise alignment is not required. And, in principle, single crystal samples are not required because Laue crystallography works with polycrystalline samples (though the polycrystallites in micro-Laue work must be made very small).

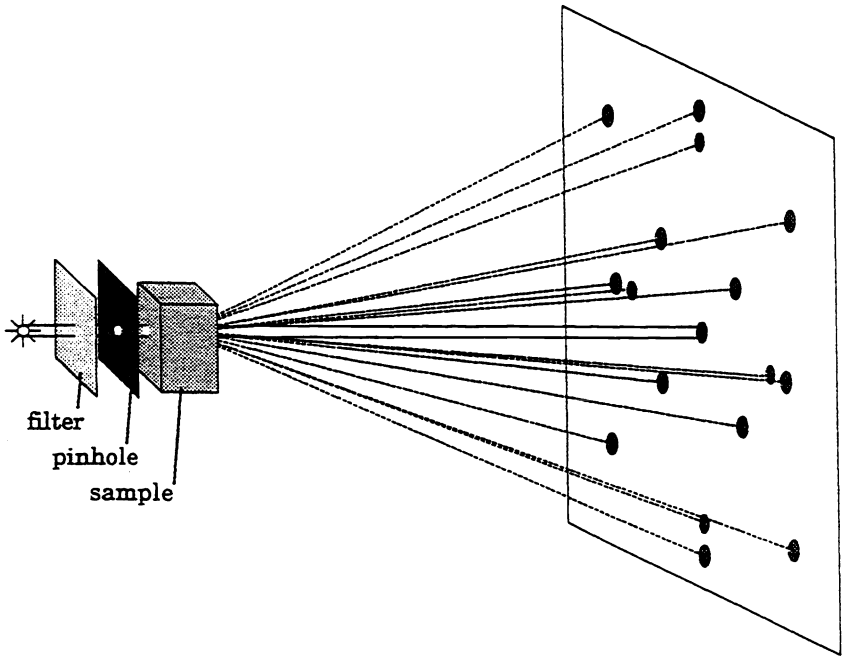

Figure 3. Schematic of the Micro-Laue Crystallography technique. Like miniaturized traditional Laue crystallography, but with a highly non-traditional x-ray source, this new technique utilizes an x-ray pencil of diameter ~10 μm of picosecond to subpicosecond duration.

A double pinhole can probe simultaneously two localized regions of the crystal, such as above and below a shock front. Owing to the magnification inherent in projecting the Bragg reflection, a pencil separation of 15 μm in the crystal should be resolved at the detector plane, and a 30 μm separation in the crystal maps out to 330 μm in the film plane. Each diffracted spot is expected to have an umbral diameter of 55 μm and penumbral diameter of 165 μm at the detector.

Micro-Laue crystallography measures the time-resolved and spatially localized 2d interplanar spacing by virtue of the diffraction angle through which the x-ray beam is reflected. It also measures the long-range order of the atoms in the crystal lattice through the shape and contrast of the diffracted spot. Effects of compression or rarefaction of the crystal planes will deflect the beam by a different angle than for the unperturbed crystal, while structural or chemical decomposition will deform and wash out the reflection. On the other hand, it has been suggested that recrystallization might take place behind the shock front. Micro-Laue Crystallography holds a unique position as a technique which might unequivocally determine if such effects exist.

The concept of narrow spatial and temporal resolution for a diffraction process through a bulk sample has heretofore been out of the question because of the signal levels which could be reasonably expected. As it turns out, a quite conservative calculation, based on actual x-ray measurements by other groups using lower laser powers, results in reasonable signal levels. Taking the x-ray spectrum from the data of Gordon et al.[2], its form at 5 keV is approximately

$$S_E = C_1 E^3 \exp(-E/kT) \quad (1)$$

where $C_1 = 1\ \text{mJ}/(4\pi \times 4.3 \times 10^{11}\ \text{eV}^4)$ and kT = 700 eV, determined by a few calibrated total-energy measurements with various filtrations. The x-ray energy seen in the diffracted spot by the detector is

$$E_{det} = S_E (\Delta\Omega) \mathcal{R} \quad (2)$$

where $\Delta\Omega$ is the solid angle determined by the pinhole, and $\mathcal{R}$ is the integrated reflectivity (expressed in eV) of the crystal. Now, the explosive PETN (pentaerythritol tetranitride, $C(CH_2ONO_2)_4$, tetragonal crystal) is very closely related to the well-characterized x-ray diffraction crystal PET (pentaerythritol, $C(CH_2OH)_4$, tetragonal crystal). At 5 keV, the PET rocking curve gives an integrated reflectivity of 5.4×10^{-5} radians, or 0.9 eV, for the dominant [002] reflection. This, by the way, is a fairly narrow rocking curve; many low-Z crystalline materials have larger values for $\mathcal{R}$ than PET. Assuming this to be a reasonable estimate for the explosive PETN, and using the geometry assumptions mentioned above results in a diffracted fluence of 1.3 pJ into a 50 μm spot, or 1600 photons into 1900 μm^2, giving a specular density greater than 1 in DEF x-ray film. Further, this is a conservative calculation because we are using x-ray fluence data taken from early reports with laser drivers which can credibly be exceeded in performance today by as much as 2 orders of magnitude, so the potential to do pulsed Laue diffraction in these tiny samples is reasonably in hand.

4. CAPABILITIES OF THE CREOL LASER PLASMA LABORATORY CR:LISAF LASER

The solid-state Cr^{3+}:$LiSrAlF_6$ (Cr:LiSAF) laser at CREOL represents a substantial new tool for the production of short-duration pulses of high-energy x-rays. The amplifier material combines a long list of desireable qualities: a long exited state lifetime (67 μs) which simplifies pumping requirements, a large gain bandwidth (220 nm) which allows ultrashort optical pulse generation, a large emission cross section, low thermal lensing, low nonlinear refractive index, high damage threshold, high uniformity of doping, and low toxicity. Development of this material won a Research and Development Magazine R&D-100 Award in 1991.[8] The CREOL laser has a maximum amplifier diameter of 25 mm, and has demonstrated 90 fs single-pulse operation at the 8 TW level.[1]

Because of the wide bandwidth, it is reasonable to expect that pulses of a few tens of femtoseconds can be generated with comparable total energy (of order 1 J); this would increase the intensity on target into the 10^{19} W/cm^2 domain. Wilks's calculations[9] suggest tens to hundreds of keV per particle would be imparted to the hot electron distribution at these intensities, and the resulting x-ray spectrum would harden substantially. Practical fluences of 10-20 keV x-rays for imaging cm-size objects would likely become routine under this development.

5. ACKNOWLEDGEMENTS

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