X-ray generation by femtosecond laser pulses and its application to soft X-ray imaging microscope

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Abstract. We have developed laser-produced plasma X-ray sources using femtosecond laser pulses at 10Hz repetition rate in a table-top size in order to investigate basic mechanism of X-ray emission from laser-matter interactions and its application to a X-ray microscope. In a soft X-ray region over 5 nm wavelength, laser-plasma X-ray emission from a solid target achieved an intense flux of photons of the order of $10^{11}$ photons/rad per pulse with duration of a few 100 ps, which is intense enough to make a clear imaging in a short time exposure. As an application of laser-produced plasma X-ray source, we have developed a soft X-ray imaging microscope operating in the wavelength range around 14 nm. The microscope consists of a cylindrically ellipsoidal condenser mirror and a Schwarzschild objective mirror with highly-reflective multilayers. We report preliminary results of performance tests of the soft X-ray imaging microscope with a compact laser-produced plasma X-ray source.

INTRODUCTION

The present high-brightness X-ray sources have been developed as third generation synchrotron light sources based on a large-scale high energy electron storage ring and magnetic undulators. A compact, tunable, high-brightness X-ray source has basic and industrial applications in a number of fields, such as solid-state physics, material, chemical, biological and medical sciences. Particularly for biological and medical applications, such as X-ray imaging, radiography and therapy, it is essential to downscale the present synchrotron light sources into the environment of a laboratory or a hospital, keeping properties of X-ray radiations. Recently availability of compact terawatt lasers arouse a great interest in the use of lasers as a compact bright X-ray source generated by intense laser-plasma interactions, replacing conventional X-ray tubes.

We have developed laser-produced plasma X-ray sources using femtoseconds laser pulses at 10Hz repetition rate in a table-top size in order to investigate basic mechanism of X-ray emission from laser-matter interactions and its application to a X-ray microscope. In a soft X-ray region over 5 nm wavelength, X-ray emission from a solid metal target, such as Al, Cu, and W irradiated by a 130 mJ laser pulse with duration of 100 fs, achieved an intense flux of photons of the order of $10^{11}$ photons/rad
per pulse with duration of a few 100 ps, which is intense enough to make a clear imaging in a short time exposure.

As an application of laser-produced plasma X-ray source, we have developed a soft X-ray imaging microscope operating in the wavelength range around 14 nm. The microscope consists of a grazing incidence cylindrically ellipsoidal condenser mirror and a Schwarzschild objective mirror with highly-reflective multilayers. We report preliminary results of performance tests of the soft X-ray imaging microscope with a compact laser-produced plasma X-ray source.

**LASER-PRODUCED PLASMA X-RAY EMISSION FROM SOLID TARGETS**

Intense ultra-short laser pulses can produce a high density plasma associating a strong electron heating generated by strong field interactions with solid state matter. As the electric field of the laser radiation exceeds the atomic electric fields above the intensities of the order of $10^{16}$ W/cm$^2$, a solid matter is ionized in a fraction of the wave oscillation through processes of the multiphoton ionization and the tunneling ionization to produce plasmas with a solid electron density of the order of $10^{23}$ cm$^{-3}$.

Hence intense femtosecond laser pulses generate interactions with plasmas characterized by solid density and very steep gradients with the scale length of a few hundreds of angstroms in the plasma expansion region. In the plasma region over a length of the skin depth, typically of the order of a few hundreds of angstroms, a strong electron heating occurs in a very short time to produce the plasma heated up to temperatures of several keV at the intensity of $10^{17}$ W/cm$^2$ away from thermal equilibrium. The high density and the electron kinetic energy of these plasmas generate bright X-ray pulses with photon energy extending up to several keV through fundamental emission processes, known as bremsstrahlung, recombination, and line emissions[1].

**Measurements of X-ray intensity and pulse duration**

A laser-plasma X-ray source has been simply made by focusing a high peak power laser onto the surface of a target placed in a vacuum chamber. Our laser system is a table-top size Ti:sapphire laser based on the chirped pulse amplification at the wavelength of 790 nm. This system can produce the output pulses with the pulse duration of 100 fs and the maximum pulse energy of 200 mJ at the repetition rate of 10 Hz. In order to investigate characterization of laser-produced plasma X-ray emission, the output pulses were focused by the off-axis parabolic mirror with a focal length of 150 mm. The measured focal spot profile was an elliptical shape with a horizontal diameter of about 10 μm and a vertical diameter of about 30 μm at the focal point. The peak focused laser intensity exceeds $4 \times 10^{17}$ W/cm$^2$ for a 100 mJ pulse energy focused on the target. The solid target mounted on the translation stage was scanned by a step of 100 μm so that a fresh surface of the target was exposed at each laser shot. We measured intensity and pulse duration of X-ray radiation emitted by laser-produced plasmas for various targets with different atomic numbers ($Z$) using the X-ray streak.
A typical temporal pulse profile of X-ray emission from plasma produced by the irradiation of a 130 mJ, 100 fs laser pulse on a Al target.

The number of detected photons was calculated from the pulse height counts of X-ray streak divided by the photon-electron conversion efficiency of the photocathode made of a 50 μm wide and 8 mm long Au thin film. The photon intensity per shot was obtained from integrating the number of photons over the X-ray pulse duration divided by a solid angle of the photocathode mounted on the X-ray streak camera, assuming an uniform angular distribution of X-ray emission from laser-produced plasmas. The total photon intensity and the pulse duration for various solid targets irradiated by a 130 mJ laser pulse energy are summarized as a function of the atomic number Z as shown in Figure 2. It is found that the laser-irradiation on a high-Z target can produce a strong photon flux of the order of $10^{11}$ photons/radian per pulse.

**FIGURE 1.** A typical temporal pulse profile of X-ray emission from plasma produced by the irradiation of a 130 mJ, 100 fs laser pulse on a Al target.

**FIGURE 2.** The photon intensity (a) and the pulse duration of X-ray emission from plasmas by laser irradiation on various targets as a function of the atomic number Z.
Measurements of X-ray spectrum

The radiation spectrum of a laser-produced plasma was measured with a flat-field normal-incident spectrograph with a platinum-coated concave grating of 1200 lines/mm blazed at 100 nm. The spectrograph equipped with a microchannel plate (MCP) detector covers the spectral range from 5 nm to 50 nm by changing the position of the 1-in. diameter MCP viewed with a charge coupled device (CCD) through an image reduction optics. The X-ray spectra of the radiation emitted by plasmas produced by Al and Cu targets are shown in Figure 3.

![X-ray spectra](image)

**FIGURE 3.** The measured X-ray spectra from plasmas produced by laser irradiation on (a) Al and (b) Cu targets.

APPLICATION TO A SOFT X-RAY IMAGING MICROSCOPE

We have developed a soft X-ray imaging microscope in a table-top size using a laser-produced plasma X-ray source as shown in Figure 4. The X-ray microscope is capable of observation of thick specimens in their natural environments, such as a living cell or biological samples in water, providing high-contrast X-ray imaging of micron-sized hydrated specimens with high penetration depth. Although the spatial resolution of an X-ray microscope is lower than that of an electron microscope, the use of X-rays as a probe can provide valuable complementary information[2, 3].

**Femtosecond laser-produced plasma X-ray source**

In applications to X-ray microscopy, laser-plasma X-ray sources evidently make it possible to reduce its system size and capital costs compared to the accelerator-based synchrotron radiation sources. In particular, a high brightness and a short duration of laser-plasma X-ray sources by the use of femtosecond laser irradiation can improve image quality with reduced exposure for the X-ray microscopy of micro-biological systems.
We constructed the target system consisting of a rotating solid target and its driving mechanisms. We used the Cu target of 3-cm diameter to generate bright soft X-rays ranging from 13 nm to 15 nm where the X-ray optics of the microscope is designed with a high efficiency. The target was irradiated by laser pulses of a 100 fs pulse duration through a 160 mm focal length lens at a 10 Hz repetition rate. The laser pulse energy of less than 50 mJ was used for a typical measurement.

**Microscope optics**

A schematic optics of the imaging X-ray microscope is shown in Figure 5. As an ordinary optical microscope, the imaging X-ray microscope consists of a condenser and an objective. The condenser is an ellipsoidal mirror of revolution that focuses the grazing incident X-ray radiation emitted at its one focal point almost over the 4π solid angle onto the sample placed at the other focal point. The distance between two focal points is about 50 cm. The objective is a Schwarzschild optics[4] comprising two concentric spherical mirrors, obtained from NTT-AT. These objective mirrors are coated with Mo/Si multilayer coatings consisting of 40 layers with each thickness of 7.14 nm to enhance the reflectivity at a wavelength of 13.9 nm to 73 % for normal incident X-rays[5]. Figure 6 shows a design of the Schwarzschild objective. This design can produce the magnification of 25. The numerical aperture (NA) of 0.0085 was determined by taking into account the best compromise between resolution and aberrations. With this NA, the resolution R of the microscope is expected to be R=1...
μm for the wavelength $\lambda = 13.9$ nm as obtained from the Rayleigh criterion $R = 0.61\lambda/NA$.

We use a back-illuminated CCD camera as a soft X-ray imaging detector. The CCD arrays consist of $1340 \times 1300$ square pixels of $20 \times 20$ μm$^2$ with a $26 \times 26$ mm$^2$ sensitive area. A microscopic image is viewed through a 0.3 μm Be filter with the CCD arrays placed at the imaging plane 1000 mm distant from the object plane.

FIGURE 5. A schematic optics of the imaging soft X-ray microscope.

Radius of curvature of the inner convex mirror = 55.845 mm
Radius of curvature of the outer concave mirror = 125.11 mm
Aperture radius of the inner convex mirror = 4.9 mm
Aperture radius of the outer concave mirror = 25.4 mm

FIGURE 6. A design of the Schwarzschild objective for the imaging soft X-ray microscope.
Experiments for characterization of microscope performance

The optics alignment of the microscope was made using a visible He-Ne laser light. First, a spot size of X-ray beam at the focal point of the condenser mirror was evaluated by a knife-edge scan as shown in Figure 7. The spot size was estimated to be about 300 μm rms radius by differentiating an edge-scan profile.

FIGURE 7. Knife-edge scan measurements of (a) a horizontal spot size and (b) a vertical spot size of the soft X-ray beam at the focal point of the condenser mirror.

FIGURE 8. A soft X-ray microscopic image of a 150 lpi mesh (169.3 μm period).
In order to investigate a performance of the microscope, we used a mesh of 150 lpi (169.3 μm period) as a test object. A CCD image of the mesh is shown in Figure 8. The image was taken for the condition of laser irradiation of 50 mJ and about 3 second exposure time. The adjustment of imaging optics was made by changing position of the object so that a high contrast image should be obtained. From the intensity profile, spatial resolution of about 1 μm was defined by distance required to reduce the intensity from 25 % to 75 %.

CONCLUSIONS

We have investigate characteristics of laser-produced plasma X-ray emission by irradiating femtosecond laser pulses on various solid targets. It has been found that the brightness of a soft X-ray emission is strong enough to make imaging in a short exposure time.

We have developed the soft X-ray imaging microscope on a table-top for the wavelengths ranging from 13 nm to 15 nm using a bright X-ray source generated by femtosecond laser-produced plasma interactions on a Cu target. We successfully demonstrated imaging of a test mesh with spatial resolution of about 1 μm. Next we are exploring microscopy of micro-biological objects to verify capabilities of our laboratory-scale imaging soft X-ray microscope.

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