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Droplet target for low-debris laser-plasma soft X-ray generation

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A water window ($\lambda = 2.3$–4.4 nm) laser-plasma X-ray source which uses ethanol droplets as target is described. This target significantly reduces debris production compared to conventional targets. The < 25 μm X-ray source primarily produces narrow bandwidth carbon and oxygen ion line emission. Absolute emission at $\lambda = 3.4$ nm is 0.5×10^{12} photons/(ster line pulse).

1. Introduction

Laser-produced plasma (LPP) is an attractive table-top soft X-ray source for, e.g., microscopy or lithography, due to its small size, high brightness and high spatial stability [1]. However, the conventional LPP on solid targets produces debris which may destroy or coat fragile X-ray components, such as windows or zone plates, positioned close to the plasma. Unfortunately, increasing the distance or introducing filters in order to protect the components result in a reduced X-ray flux. In this paper we describe the use of small liquid droplets as target. This target reduces debris production by more than two orders of magnitude compared to a conventional target, allowing more efficient use of the X-ray source by smaller source–component distances.

X-ray emission from laser-produced plasmas has been extensively investigated both as a diagnostic parameter in inertial confinement fusion research and as a high-brightness soft X-ray source in itself [2]. Using intensities around 10^{13}–10^{14} W/cm^2 such LPP soft X-ray sources may reach conversion efficiencies of several tens of percents [3]. Compared to the synchrotron soft X-ray source the LPP has the advantage of high peak intensity, high spatial stability, small source size and that it does not require ultra-high vacuum. Furthermore, the LPP systems are compact and cheap. Other compact soft X-ray sources such as the plasma focus and the z-pinch both exhibit larger source size and less spatial stability than the LPP [4].

For convenience, solid targets have often been used for LPP soft X-ray sources. In general, light element targets yield a line radiation spectrum, and heavy element targets result in continuum radiation due to Brehmsstrahlung emission. Several studies of emission from different target materials have been performed [5]. Unfortunately, using conventional solid targets, laser plasmas eject significant amounts of debris, i.e. hot ions and larger particles, which may damage optical and X-ray components positioned close to the plasma. Several methods have been invented to reduce the effect of debris, e.g., using a small backing pressure of helium [6], a fast shutter system that eliminates part of the debris [7], or a toriodal relay mirror for the protection of sensitive components [8]. However, only few attempts have been made to eliminate the production of debris. Thin film tape targets reduce the amount of debris by avoiding chock wave ejection [7] or delayed evaporisation [9]. Still, significant amounts of debris particles are produced, presumably from cooler zones illuminated by the noncentral parts of the beam. Gas-phase targets would be another low-debris alternative. Unfortunately, the low density results in low X-ray intensity. However, frozen neon has been demonstrated [10] and the use of liquid xenon drops was suggested by Trail [9].

In the present paper we efficiently evaporate and ionize the whole target in order to reduce the production of debris particles. This is possible since the target is a small droplet comparative in size with the focused laser beam diameter. Thereby the production of debris particles in the cooler zones at the low-intensity tails of the beam is significantly reduced. We assume that the small residual production of debris consist primarily of ions. Thus, the LPP drop X-ray source may be positioned close to X-ray components without risking damage, thereby increasing the X-ray flux. Furthermore, the drop X-ray source has the advantage of providing fresh target drops for a full day of experiments, eliminating the need for the disruptive changes of target which are well known to users of conventional targets. Another advantage is that the influence on focal position due to surface nonuniformity or mechanical instability in conventional target systems is eliminated. Finally, since the X-ray source is generated from a droplet in free space and not from a conventional slab or cylinder of solid material, X-ray components may be positioned closer to the source and there is spatial access from all directions.

There are many appealing applications of a debris-free LPP X-ray source, e.g., soft X-ray microscopy [11] and lithography [12]. The two techniques show good promise for high resolution imaging of wet biological objects and narrow-linewidth semiconductor processing, respectively. To date most work has been performed using synchrotron radiation. Laser-plasma work for microscopy include, e.g., contact imaging in the water window [13] and a table-top LPP based scanning microscope at $\lambda = 14$ nm [9]. The attempts to use LPP for lithography are, e.g., contact lithography in the 1 nm region [14] and projection lithography at 14 nm [15]. In all cases debris is a problem and a debris-free source would significantly improve the performance of the systems.

2. Experiments

The experimental arrangement for the laser-plasma soft X-ray droplet source is shown in fig. 1. Liquid ethanol at a pressure of 30–50 atmospheres is injected into the vacuum chamber through an approximately 10 µm diameter vibrating capillary ink jet printing nozzle [16]. In order to achieve stable drop formation the nozzle is vibrated resonantly with a piezoelectric crystal at approximately 1 MHz. This results in $\sim 10^6$ drops/s with a diameter of ~ 10 µm and a speed of ~ 50 m/s being injected continuously into the vacuum chamber. The distance between the drops is ~ 50 µm. Since the laser operates at 10 Hz most drops remain unaffected by the laser beam. In order to maintain the vacuum, the train of drops is directed slightly downward into a liquid nitrogen trap which freezes the residual ethanol. The operating pressure in the system is $\sim 10^{-4}$ mbar. From pumping speeds and pressure considerations we estimate that 1/3–1/4 of each droplet evaporate before entering the liquid nitrogen trap.

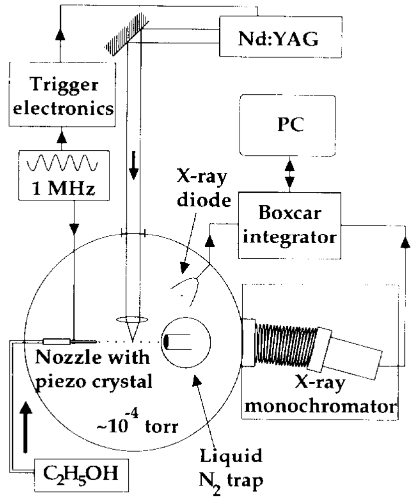

Fig. 1. Experimental arrangement for the laser-plasma droplet X-ray source

The beam from a frequency-doubled active/active/passive modelocked 10 Hz Nd:YAG laser (Continuum PY61 A/A/P) is focused on the droplets with a 50 mm lens. The laser produces 120–140 ps pulses with 70 mJ/pulse at $\lambda = 532$ nm. Using a knife edge method [17] the fwhm at the focal point of the beam was determined to ~ 12 µm, corresponding to an intensity of $\sim 4 \times 10^{14}$ W/cm^2. The small size of the droplets in combination with the strongly focused beam makes spatial stability important for stable operation, i.e., that each laser pulse hits the designated droplet centrally. Furthermore, due to the high speed of the droplets, the temporal jitter of the laser pulse emission has to be small. By using the 1 MHz generator, which controls the phase of the drop formation, in combination with frequency division and delay circuits as the trig source for the laser, the pulse emission is controlled to ±25 ns, which corresponds to approximately ±1.5 µm of droplet travel.

For comparison purposes, a thin film plastic tape target [7] was intermittently operated at the same position as that of the drop target. It consisted of a 12 µm thick cassette tape with its plastic side turned towards the laser. The laser angle of incidence was 45° to the tape normal and the tape was supported by a 13 mm diameter cylinder with a 3 mm diameter hole which allowed the free expansion of debris beyond the tape. The system reduces the backscattered debris significantly compared to solid targets [7].

The laser-plasma soft X-ray sources were spectrally characterized using a 1 m grazing-incidence monochromator (Minuteman 301-G) with an CsI photocathode electron multiplier detector. The monochromator detected X-rays at a 90 degrees angle to the incident laser beam. For the water-window measurements the monochromator was blazed at $\lambda = 2.9$ nm and had a resolution of 0.02 nm. Figure 2 displays the spectrum for the droplet source, showing a clean line emission of hydrogen- and helium-like carbon and oxygen ions. The spectrum is uncorrected for the spectral response of the monochromator and the detector. 30 shot averaging was used and the full spectrum was recorded in 20 minutes. Note that the background is very small compared to the line radiation, making this source suitable for diffractive optics such as zone plates. The tape target exhibits a similar spectrum due to the carbon and oxygen rich plastic tape. However, the tape target shows significantly lower CVI/CV and OVIII/OVII ratios, indicating a higher degree of ionization in the drop source. Using lower resolution gratings it was concluded that the drop-source emitted negligible continuous radiation at higher wavelengths. However, significant line emission was observed in the $\lambda = 10$–30 nm region.

The photon flux from the drop source was estimated by measuring the time integrated signal from an X-ray diode (Hamamatsu G-1127-02) [18] positioned at 45° angle to the incident beam. The diode was covered by a 170/100 nm Ag/Al sandwiched soft X-ray filter [19], which essentially transmitted the carbon lines at $\lambda = 3.37$, 3.50 and 4.03 nm while being nearly opaque to the rest of the emitted spectrum. From these measurements we deduce that the full plasma emits 1×10^{12} photons/(ster line pulse) from the stronger lines. Here the uncertainty in the efficiency and sensitivity of the diode is ±40% [18]. The largest other experimental source of error is approximately 10% due to a non-negligible transmission of the OVIII and OVII radiation around $\lambda = 2$ nm through the filter.

In order to determine the usefulness of the laser-plasma source for, e.g., microscopy, the size of the plasma source was measured. A simple knife edge method was used. Compared to a pinhole camera, the small knife edge may be positioned closer to the plasma without disturbing the exciting laser beam, potentially allowing higher resolution imaging. Furthermore, the alignment of a knife edge is easier than that of a pinhole camera when small distances to the plasma are necessary. As depicted in fig. 3, the knife edge was scanned close to the plasma by a DC motor and the X-ray flux at $\lambda = 3.37$ nm was detected through the 20 µm slit of the grazing incidence monochromator. In our case the plasma-knife edge

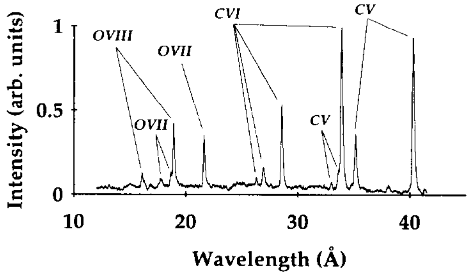

Fig. 2. Water window X-ray emission spectrum from droplet X-ray source.

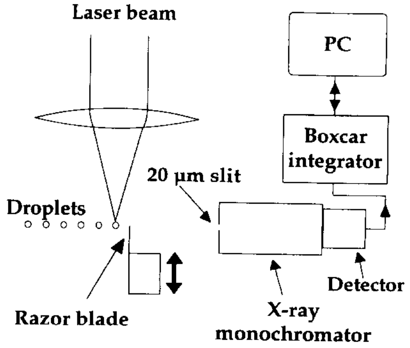

Fig. 3. Experimental arrangement for knife-edge determination of the plasma size.

distance was 4 mm and the plasma-monochromator distance was 180 mm. Diffraction calculations result in a resolution of <4 μm with the system, making this a potentially attractive method for measurements on small plasmas. The method can also be extended to two-dimensional imaging using tomographic techniques [20]. However, in the current measurements time constants in the detection system, geometrical effects and nonperfect razor edges all contribute to a total resolution limit of 10 μm. Figure 4 shows a typical recording using a laser beam focused for maximum soft X-ray flux. Note the sharp edge at a razor edge displacement of ~ 100 μm and the longer tails at distances further away from the center. The sharp edge is believed to be due to an intense central core while the long tails probably are due to a combination of the expanding plasma and plasma formation in the shell of residual evaporating ethanol that surrounds the droplet. Comparing the experimental measurements with theoretically calculated knife edge data of core-shell phantoms and assuming spherical symmetry, we conclude that $>50\%$ of the $\lambda = 3.37$ nm photons are emitted from a <25 μm diameter central core. The oscillations in the left part of fig. 4 are due to a slight temporal drift in the laser triggering, resulting in unstable plasma formation. By slightly defocusing the laser, the oscillation can be avoided albeit at the price of lower X-ray flux.

One of the major advantages of the droplet laser-plasma source is that it is nearly debris-free. Semi-quantitative debris measurements were performed by measuring deposition of debris on microscope slides as a function of time and distance from the source. For the droplet source the slides were positioned 15–50 mm from the source in the plane of the laser beam and the liquid jet, and "exposed" to debris up to 20 hours of continuous 10 Hz, laser-plasma operation. For comparison, similar measurements where performed with the low-debris plastic tape target, only differing in a much shorter exposure times. Both targets result in a deposition on the slides which appear similar when viewed in an optical microscope. X-ray photoelectron spectroscopy (XPS) analysis of the debris films show similar concentrations of the major constituents carbon and oxygen (2:1 ratio). The tape target debris film also included trace amounts of nitrogen and iron ($\leqslant 1\%$) while the droplet target film only showed carbon and oxygen. Due to the similarity of the films, the relative thickness of the deposited layers were measured by optical absorption at $\lambda = 632$ nm. From these measurements we deduce that the debris deposition rate of the droplet X-ray source is 200–300 times less than that of the tape target source. This relative determination of the debris deposition rate clearly shows the very low debris production of the drop laser-plasma source. It is interesting to note that under the plasma, i.e., perpendicular to the laser beam and the liquid jet, the debris deposition was another 1 to 2 orders of magnitude smaller. Due to geometrical reasons comparative measurements on the tape target are not possible. Measurements of debris behind the drop target should also be interesting but the heating and thus cleaning of the slides by the laser beam makes such measurements difficult.

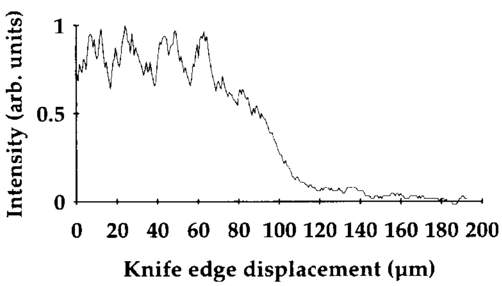

Fig. 4. Knife-edge scan of droplet laser-plasma X-ray emission at $\lambda = 3.37$ nm.

3. Discussion

The small size of the target is believed to be important for the nearly debris-free nature of the droplet laser-plasma source. The small dimension along the laser beam direction prevents debris production due to delayed evaporation or shockwave ejection and the small size perpendicular to the beam allows the full target to be efficiently vaporised and ionized. Thus the production of debris particles from the cooler zones in the radial tails of the beam should be small. Experimentally, the drop source debris deposition rate was found to be significantly lower than that of a low-debris plastic tape target. Quantitative determination of the deposition rate is difficult since very thin films have to be evaluated. However, we will attempt such measurements using angle-resolved XPS [21].

In addition to reducing the debris production significantly, the droplet X-ray source also exhibits high brightness. Using the knife edge method we determined the size of the central core plasma to $<25\ \mu m$ and the photon flux from the core to 0.5×10^{12} photons/(ster line pulse) at $\lambda=3.37$ nm. Assuming a plasma expansion velocity of $<2\times10^{7}$ cm/s, the knife edge size measurements suggest that the X-ray pulse temporally follows the laser pulse. With a Doppler broadened line shape, the brightness of the central core is then $>1\times10^{18}$ photons/(mrad^2 mm^2 0.01% BW s) at $\lambda=3.37$ nm. The occurrence of ionization stages indicates an electron temperature of one to a few 100 eV resulting in a Doppler broadening in accordance with the assumed plasma expansion velocity. The brightness compares well with other LPP soft X-ray sources.

At a first glance it may seem inconvenient to use a 1 MHz continuous liquid jet since the most drops that enter the vacuum system are not used for plasma formation and thus only contribute to gaseous evaporation and thus vacuum problems. Other drop generation techniques [16] may be operated at lower repetition rates resulting in less vacuum problems. However, we use a continuous jet for four reasons. (i) It produces sufficiently small droplets to allow complete evaporisation and ionization in a tightly focused beam of moderate laser energy. (ii) It allows very good spatial control of drop position which is necessary for stable operation with a tightly focused beam. The other drop generation techniques generally produce either larger droplets and/or unstable drop trajectories. Thus, larger lasers are required to reach the intensity and energy necessary for high-brightness and negligible-debris operation. (iii) The continuous flow prevents clogging of the capillary due to freezing. (iv) Continuous jets may be operated with a wide range of different liquids.

4. Summary and conclusions

We have developed a laser-plasma soft X-ray source for the water window which produces significantly less debris than conventional methods. The source utilizes ethanol droplets as target and emits predominantly helium- and hydrogen-like carbon and oxygen ion X-ray line radiation from $\lambda=2$–4 nm. The source has high brightness, allows full-day operation without interrupts, and significantly reduces damages on components positioned close to the source.

As observed in the introduction such a source is of great interest for X-ray microscopy and lithography. Calculations show that the brightness is sufficient for scanning X-ray microscopy of biological substances. For microscopy applications, however, it would be favourable to obtain the major part of the soft X-ray emission in the lower part of the water window, i.e. below $\lambda=3$ nm. This is due to the smaller water absorption at these wavelengths. We will develop such a drop source based on LPP on nitrogen rich liquids, and detecting the strong NVII and NVI lines at 2.5 and 2.9 nm, respectively.

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