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Liquid-jet target for laser-plasma soft x-ray generation

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We describe a new liquid-target system for low-debris laser-plasma soft x-ray sources. The system is based on a microscopic liquid jet and is experimentally evaluated for 0.7–1 keV proximity lithography and water-window x-ray microscopy applications. Compared to an existing liquid-droplet target, this target system has the same low debris emission, high x-ray photon flux, and narrow spectral bandwidth. The advantages of the liquid-jet target include improved x-ray flux stability, increased range of suitable target liquids, and elimination of the need for temporal synchronization, thereby allowing less complex laser systems to be used. © *1996 American Institute of Physics.* [S0034-6748(96)03412-0]

I. INTRODUCTION

The laser-produced plasma is an attractive compact x-ray source[1,2] suitable for applications in, e.g., microscopy[3,4] and lithography.[5,6] However, with conventional bulk targets, the applicability of laser plasmas is restricted due to the emission of debris, which may damage sensitive components positioned close to the plasma x-ray source. With liquid or frozen microscopic droplets as target,[7] the debris problem has been shown to be negligible[8] or eliminated.[9] These x-ray sources also feature high flux and brightness, allow long-term operation without interruption, provide excellent geometric access, emit narrow-bandwidth radiation appropriate for zone-plate optics, and provide fresh target drops at high rates to match high-repetition-rate lasers. Furthermore, spectrally tailored emission for a specific application can be produced by selecting a target liquid with proper elemental contents. For example, x-ray emission suitable for microscopy,[9] proximity lithography,[10] and projection lithography[11] have been generated with ammonium hydroxide, fluorocarbon, and water droplets, respectively.

Until now, stable generation of microscopic droplets with a continuous-liquid-jet method has been a prerequisite for efficient liquid-target laser-plasma operation.[7–11] In the present paper we extend the applicability of this method to liquids which do not have suitable hydrodynamic properties to form stable drops by utilizing a microscopic liquid jet as target. This also eliminates the need for laser-droplet temporal synchronization, resulting in significantly relaxed requirements on the laser and improved long-term stability while still maintaining the advantages of microscopic liquid targets discussed in the first paragraph.

II. THEORETICAL AND EXPERIMENTAL BACKGROUND

The key issue of the present paper is that the x-ray-emitting laser plasma is generated in a microscopic liquid jet instead of in a microscopic droplet. The experimental arrangement is schematically illustrated in Fig. 1. The liquid-jet target is produced by forcing the liquid at a pressure of approximately 50 bar through a $\sim$10-μm-diam glass capillary nozzle into a $\sim 10^{-4}$ mbar vacuum tank. The nozzle produces a $\sim$10-μm-diam liquid jet which spontaneously breaks up into droplets at a drop-formation point at a distance L from the nozzle orifice. Laser plasmas are produced by focusing $\lambda=532$ nm, 70 mJ, 100 ps pulses from a 10 Hz mode-locked Nd:YAG laser (with a 3 mJ, $\lambda=355$ nm prepulse for x-ray flux enhancement[12]) onto the liquid jet between the nozzle orifice and the drop-formation point. X-ray emission in the 1–5 nm wavelength range requires an intensity on the order of 10^{14} W/cm^2, corresponding to a focal-spot full width at half maximum (FWHM) diameter of 10–15 μm from these compact lasers.[7] Thus, the spatial stability of the jet is of great importance for efficient x-ray generation with low pulse-to-pulse fluctuations. The continuous liquid jet method used in the present paper is particularly suited for such high requirements of stability.

The arrangement for droplet target laser-plasma x-ray generation[7–12] is similar to the above method. However, then the glass nozzle is piezoelectrically vibrated at $\sim$1 MHz to produce a stable train of equally sized (10–15 μm) microscopic droplets having a speed of $\sim$50 m/s. In order to ensure that each laser pulse in the focused beam hits a single droplet, the piezoelectric vibration frequency, which controls the spatial position of the droplets, is electronically synchronized with the Q switch of the laser to $<\pm 30$ ns. Thus, in addition to the spatial stability requirement of the liquid-jet method above, the droplet method requires accurate temporal synchronization between the laser pulse and the droplet.

To understand the advantages and limitations of the liquid-jet target compared to the droplet target, the hydrodynamics of continuous liquid jets is briefly discussed. Forcing a viscous liquid through a nozzle results in a jet which spontaneously breaks up in a train of droplets according to Fig. 1. The drop formation is basically due to the liquid's tendency to minimize the surface energy and is theoretically described in Ref. 13 and summarized in Ref. 11. The distance L to the spontaneous drop formation point is

$$L = 12 \cdot \nu \left[\sqrt{\frac{\rho \cdot d^3}{\sigma}} + \frac{3\eta d}{\sigma} \right], \tag{1}$$

where d and ν are the diameter and velocity of the liquid jet, respectively. The liquid is characterized by its density ρ, surface tension σ, and viscosity η. For our $d \approx 10$ μm jets the

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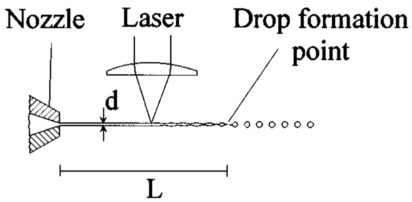

FIG. 1. Experimental arrangement for liquid-jet laser-plasma x-ray generation.

distance L is typically a few millimeters for common liquids such as water or ethanol. For these liquids a stable train of droplets is produced by applying a controlled piezoelectric vibration to the nozzle. Due to conservation of mass, the diameter of the droplets is typically 1.5–2 times larger than the diameter of the liquid jet. Thus, the target diameter of the liquid jet is significantly smaller than that of a droplet.

It is clear from Eq. (1) that liquids with very low surface tension are not suitable for droplet formation since then L grows to infinity. However, there are a significant number of liquids with reasonable hydrodynamic data which still do not provide stable drops. Many of these have somewhat low surface tension, resulting in spontaneous drop formation far away (typically centimeters) from the nozzle tip. However, further away from the tip, the stability of the drops decreases. This is not acceptable since the laser-plasma production requires very high accuracy in the drop position ($\pm$a few μm) to ensure stable x-ray generation. From the above it is clear that for liquids with unstable droplet formation, the liquid-jet target is better suited than the droplet target for laser-plasma operation. Still, it should be noted, that liquids with very low viscosity are not suitable for any of the two target methods since turbulence in the nozzle prohibits the formation of a liquid jet.[11]

III. EXPERIMENTS AND DISCUSSION

In the experiments described below we compare the liquid-jet and droplet target laser-plasma x-ray sources with respect to x-ray flux, brightness, debris, and stability. The experiments were performed using liquid fluorocarbon and ethanol as target liquids since these two liquids have been extensively characterized for droplet-target proximity x-ray lithography[10] and water-window microscopy[7,12] applications. While ethanol is perfectly suitable for stable droplet generation for a broad range of piezoelectric vibration frequencies, the fluorocarbon droplet generation is sensitive to small disturbances, making liquid-jet operation favorable.

The x-ray photon flux from the fluorocarbon plasma was measured at a 45° angle to the incident laser beam with an x-ray diode (Hamamatsu G-1127-02) covered by a free-standing sandwiched 2 μm/260 nm Al/Cu filter.[10] With this filter combination the x-ray signal is dominated by the $\lambda<1.7$ nm F VIII and F VIX emission, which is suitable for proximity lithography. For both the droplet and liquid-jet target, the x-ray photon flux was $\sim 2\times10^{12}$ ph/sr pulse. The C V and C VI water-window emission from ethanol was measured with a 160/100 nm Ag/Al filter.[7] Also for this liquid no difference in x-ray flux was observed between the droplets and the jet, resulting in $>3\times10^{12}$ ph/sr pulse line at the C VI $\lambda=3.37$ nm line.

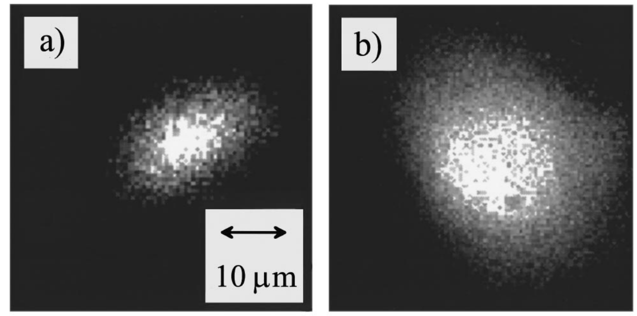

FIG. 2. X-ray pinhole camera images for liquid-jet target without prepulse (a) and with prepulse (b).

Spectra for both liquids were recorded with a 1 m grazing incidence monochromator (Minuteman 301-G).[7,10] The $\lambda<1.7$ nm fluorocarbon and water-window ethanol spectra were dominated by line emission from F VIII/F VII and C VI/C V, respectively. For ethanol, no difference in the spectrum was observed between the droplets and the jet while for the fluorocarbon a slightly higher degree of ionization in the liquid-jet measurements was detected. The determination of line width for ethanol resulted in $\lambda/\Delta\lambda>300$ for both the droplets and the liquid jet. In both cases this determination is limited by the resolution of the monochromator.

For many imaging applications the x-ray source brightness (ph/sr μm^2 pulse) is more important than the total photon flux. For this purpose the size of the x-ray-emitting plasma was measured with a pinhole camera. Here an 8 μm movable pinhole was positioned $\sim$10 mm from the fluorocarbon plasma and the image was recorded with a 24$\times$24 μm pixel thinned back-illuminated x-ray sensitive charge coupled device (CCD) detector.[12] Figure 2 shows the plasma emission at $\lambda<1.7$ nm for the fluorocarbon liquid jet with no prepulse (a) and with 3 mJ UV prepulse (b). The $\sim$10$\times$15 μm elongated structure in Fig. 2(a) has its major axis along the direction of the liquid jet, illustrating the influence of target dimension on plasma size. Similar images of droplet plasmas show a circular image with $\sim$10 μm FWHM diameter. Thus, for small plasmas, the emitting area of the liquid jet is somewhat larger than that of the droplets, resulting in a slightly smaller brightness. However, in a recent paper (Ref. 12) we have shown that the application of a UV prepulse enhances both the brightness and flux. Due to the plasma expansion between the prepulse and the main pulse, the difference in target type does not noticeably influence the plasma size in this case. This is shown in Fig. 2(b) where a close to circular plasma emission area is shown despite the elongated liquid-jet target. Recordings with the same prepulse condition and droplets as target show very similar x-ray emission images.

The debris emission from liquid jet and droplet targets were compared using the methods described in Ref. 8. To determine the total debris deposition (ions, atoms, and larger fragments), carefully cleaned glass plates were positioned 20 mm from the fluorocarbon plasma source. After 1 h of 10 Hz operation in either the droplet or liquid jet mode, the thickness of the deposited debris layer was determined by optical opacity measurements. From these measurements we conclude that the debris emission from droplet target and the liquid-jet target are equal, within the <20% relative error of the measurement method. The quantitative debris emission has been determined to typically 70 pg/sr pulse in Ref. 10. Measurements on ethanol show the same similarity between the two target types, although the quantitative numbers are approximately an order of magnitude less. Thus, compared to conventional targets both the liquid-jet and droplet target systems reduce debris by several orders of magnitude.[8]

The emission of larger debris fragments is particularly harmful since such projectiles may damage fragile x-ray optics or masks positioned close to the plasma source. The emission of such larger fragments was examined by positioning a 100-nm-thick freestanding Al foil 20 mm from the liquid-jet plasma and exposing it to debris for 60 min. The foil was then checked for pinholes in a CCD-camera equipped optical microscope. No new pinholes could be observed, indicating the absence of larger debris fragments.

Compared to conventional low-debris targets such as the thin film tape target, the liquid-jet target shows the same reduction of debris as the droplet target does, while still maintaining the same high x-ray flux. This may seem surprising since the negligible debris production of the droplet target has been attributed to the fact that all target material is positioned in the central high-intensity region of the focused beam and no target material is present in the cooler zones of the lower-intensity radial wings.[8] Thus, the production of large-fragment debris is assumed to be eliminated and the full target mass is efficiently heated and highly ionized, resulting in ionic and atomic debris. The similarity in the debris production of the two methods is probably due to the fact that approximately the same target mass is within the focal volume. This is because the droplet diameter is significantly larger than the jet diameter (cf. Sec. II). Furthermore, even when using the droplet method, all target material is not in the center of the focused beam due to uncertainty in the laser-droplet temporal synchronization. Thus, also for droplets, some target material may occasionally be present in the cooler wings of the beam, making the similarity of the two methods significant. Finally, it should be noted that the slightly higher degree of ionization observed for the fluorocarbon jet may be explained by the better stability of the jet in the laser-beam focus.

An important issue of practical concern is the long-term x-ray photon flux stability. With droplets as target, small changes in the target liquid pressure or temperature may result in a slow drift in the droplet position. Thus, the temporal synchronization of the laser pulse must be adjusted. This problem is distinct for the fluorocarbon but less pronounced for, e.g., ethanol. Naturally, a feedback loop automatically controlling the laser-droplet synchronization may be constructed. However, with the microscopic liquid jet as target, the stability is inherent in the system. Figure 3 shows the x-ray flux from a fluorocarbon target for 30 min of uninterrupted 10 Hz operation. Each data point represents an average of 30 pulses. The slow decrease in the detected x-ray flux is due to a long-term reduction in the laser intensity. The short-term pulse-to-pulse fluctuations without prepulse are typically <10%, which is lower than with droplets (<15%).[12] These fluctuations are assumed to be primarily due to laser pulse-to-pulse intensity fluctuations.

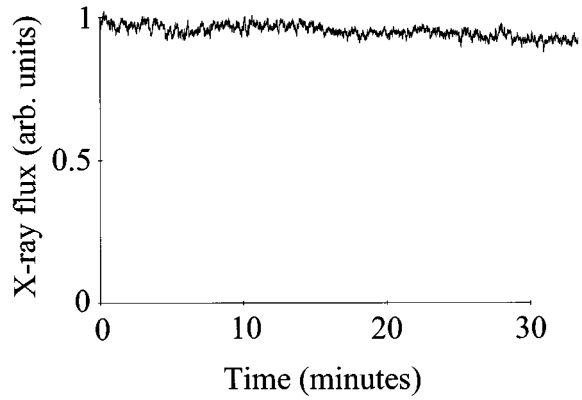

FIG. 3. Stability of x-ray emission from an unattended fluorocarbon liquid-jet-target laser plasma.

IV. CONCLUSIONS

It is clear from the discussion above that the liquid-jet target has several advantages compared to the droplet target. Primarily it allows stable liquid-target laser-plasma operation for liquids which are not suitable for stable droplet generation due to, e.g., low surface tension. We thereby extend the number of liquids suitable for laser-plasma x-ray generation significantly. Furthermore, the method requires no synchronization between droplets and laser. Thus, lasers with intrinsic high trig jitter, e.g., passive mode-locked systems, may be used and synchronization electronics are eliminated. In addition, the relaxed synchronization requirements result in increased stability by avoiding drifts in droplet position due to thermal and pressure changes, making uninterrupted stable operation possible over significant time periods. The x-ray flux, brightness, and debris emission are similar for the liquid-jet and droplet methods.

Naturally, the liquid-jet target cannot be operated with liquids having high surface tension since then the distance to the droplet formation point is short, requiring a short distance between the plasma and the nozzle orifice. Operating at a short distance may result in physical damage to the nozzle. Furthermore, at such distances we have occasionally observed plasma-induced instabilities of the jet that we attribute to the charging of the nozzle tip by the plasma. By grounding the nozzle, the problem is reduced but not always eliminated. Thus, for such liquids the droplet target is favorable.

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