

# Cryogenic liquid-jet target for debris-free laser-plasma soft x-ray generation

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A new target system based on a continuous cryogenic liquid jet for debris-free laser-plasma soft x-ray generation is described. The system is experimentally evaluated with liquid nitrogen as target material. With this target the photon flux is  $4.5 \times 10^{11}$  photons/(sr×pulse) from the  $\lambda=2.88$  nm N VI line. Brightness and stability are also investigated for this table-top soft x-ray microscope source. The possibility to utilize other cryogenic liquids such as neon, argon, and xenon and, thus, making the system interesting for short-wavelength lithography applications, is also discussed. © 1998 American Institute of Physics. [S0034-6748(98)02506-4]

## I. INTRODUCTION

The rapid development of soft x-ray optics during the last decades has increased the interest in soft x-ray applications such as microscopy<sup>1</sup> and lithography.<sup>2,3</sup> The growth of these applications outside the synchrotron radiation community would further benefit from the development of laboratory-scale, high-brightness soft x-ray sources. The laser-produced plasma (LPP) is a promising candidate due to its relatively low cost. However, with conventional solid targets the applicability of the LPP is limited by the emission of debris,<sup>4</sup> which may damage sensitive components positioned close to the source. It has previously been shown that this debris emission can be lowered several orders of magnitude<sup>5</sup> or even eliminated<sup>6</sup> with a microscopic liquid-droplet target system. This x-ray source also features high flux and brightness, allows long-term operation without interrupts, emits narrow-bandwidth radiation suitable for zone-plate optics, and provides fresh target material at high rates, thus, allowing the use of high-repetition-rate lasers. Since our target liquids typically consist of light elements generating characteristic line emission, it is possible to tailor the soft x-ray spectrum by selecting the target material. For example, with ammonium-hydroxide<sup>6</sup> and fluorocarbon<sup>7</sup> droplets, intense x-ray emission suitable for microscopy and proximity lithography, respectively, is generated. For liquids which do not have the suitable hydrodynamic properties for droplet formation, a target system based on a microscopic liquid jet has been demonstrated.<sup>8</sup>

In the present article we extend the liquid-jet target concept to cryogenic liquids, i.e., substances that are in gas phase at normal temperature and pressure (NTP). The cryogenic liquid-jet target combines the debris-free nature of a gas target with the high density necessary for efficient x-ray generation. It also offers the possibility to generate single-

element emission and, thus effective conversion from laser energy to useful x-ray flux. The system described in this article was developed for soft x-ray microscopy and employs nitrogen as target liquid. Helium- and hydrogen-like nitrogen ions (N VI and N VII) emit in the lower part of the water-window region ( $\lambda=2.4$ – $4.4$  nm). This region provides a natural contrast between carbon (proteins) and oxygen (water) therefore making it suitable for imaging of biological material.

Other cryogenic-target liquids with similar hydrodynamic properties as liquid nitrogen are oxygen, neon, argon, and xenon, which are suitable laser-plasma targets for, e.g., proximity and EUV projection lithography. Previous laser-plasma x-ray source work on these elements includes frozen solid nitrogen,<sup>9</sup> xenon,<sup>10,11</sup> and neon,<sup>12</sup> which all exhibit significant debris emission and a high-pressure gas target system for xenon<sup>11</sup> having the disadvantage of low number density. Xenon especially has attracted interest due to its high atomic number which should theoretically result in a high conversion efficiency.<sup>13</sup> Liquid xenon drops were suggested already in 1989 by Trail<sup>14</sup> and a cryogenic xenon droplet generator based on the drop-on-demand method operating at atmospheric helium pressure has recently been presented.<sup>15</sup>

## II. THEORETICAL BACKGROUND

Cryogenic liquids generally have lower surface tension and viscosity than ordinary room-temperature liquids such as, e.g., water or ethanol. This will make them less suitable for continuous liquid jet operation. In order to understand the difficulties in cooling and forming a microscopic jet out of a liquified gas, the theory of continuous liquid jets and the drop formation of such jets needs to be discussed. This theory is well known and theoretically described in Ref. 16 and summarized in Ref. 17.

A stable liquid jet requires a laminar flow, resulting in that the Reynolds number  $Re$  should be lower than  $\sim 900$ .

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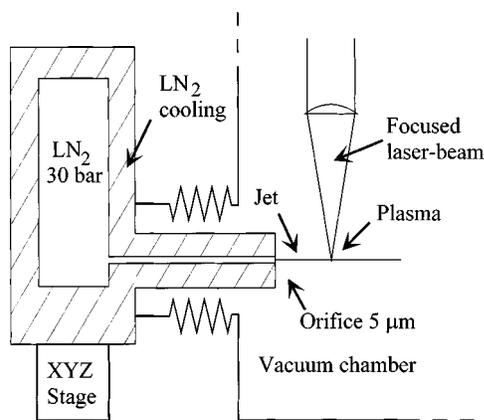


FIG. 1. Schematic nozzle design for stable liquid-nitrogen liquid-jet operation.

Here

$$\text{Re} = \frac{\rho v d}{\eta}, \quad (1)$$

where  $d$  is the diameter,  $v$  the velocity,  $\rho$  the density, and  $\eta$  the viscosity of the liquid jet. Clearly, a low-viscosity liquid such as liquid nitrogen is difficult to operate, especially in combination with large-diameter nozzles. The low viscosity will limit the jet diameter in order to keep the Reynolds number below  $\sim 900$ .

A cylindrical liquid jet is inherently unstable and tends to spontaneously break up into drops. This drop formation is due to minimization of surface energy and occurs at a break-up distance  $L$  (the spontaneous drop formation distance) from the nozzle orifice. Here

$$L = 12\nu \left( \sqrt{\frac{\rho d^3}{\sigma}} + \frac{3\eta d}{\sigma} \right), \quad (2)$$

where  $\sigma$  is the surface tension. In order to generate a stable train of equally sized and equally spaced drops, the nozzle must be stimulated at a certain frequency close to the spontaneous drop formation frequency. With a well-designed nozzle this can be achieved by, e.g., vibrating it with a piezocrystal. Due to growing jet instabilities far away from the nozzle orifice, a reasonably short drop formation distance is necessary. This will make liquid nitrogen and similar liquids, having low surface tension and, thus, long break-up lengths inappropriate for stable droplet production. Therefore the liquid-jet method<sup>8</sup> was chosen in the present article.

### III. EXPERIMENTS AND DISCUSSION

The experiments described in this article were performed with liquid nitrogen as target material. We have investigated x-ray flux, brightness, debris emission, and stability of this new laser-plasma x-ray source.

The liquid-nitrogen jet is generated in a vacuum chamber. Figure 1 shows the schematic nozzle design. The liquid jet is formed by forcing the cryogenic liquid with a pressure of  $\sim 35$  bar through an  $\sim 5 \mu\text{m}$  diameter orifice. We use a commercially available electron microscope aperture as the orifice. A metal filter is mounted inside the nozzle to obstruct

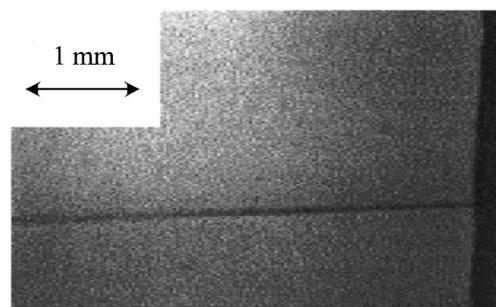


FIG. 2. Microscope image of a stable  $5 \mu\text{m}$  diameter liquid-nitrogen jet in vacuum.

dust particles, which could clog the orifice. In Fig. 2, a microscopic image of the stable liquid-nitrogen jet is presented. With our current arrangement the nozzle exit diameter is limited by vacuum considerations and probably also by Reynolds number. With a 500  $\ell/\text{s}$  turbomolecular pump the pressure in the chamber is  $1 \times 10^{-3}$  mbar. At this pressure the absorption in the water-window wavelength region is less than 1% per meter for nitrogen, thus, making it negligible. If it is necessary to work with better vacuum conditions, this may be arranged with a larger vacuum pump or with a differential pumping stage. The cryogenic target liquid requires a cooling system for the nozzle and construction materials that can endure low temperatures in combination with high pressures. In our current arrangement the cooling of the target liquid is maintained by a liquid-nitrogen system and the nozzle is made of metal, mainly stainless steel. The rigid construction complicates the stimulation necessary for stable drop generation and consequently the liquid-jet approach is applied.<sup>8</sup> XYZ translation of the nozzle from outside the vacuum chamber is available, making it easy to adjust the liquid jet to the laser focus inside the vacuum chamber.

The x-ray-generating laser plasma is 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<sup>18</sup>) onto the liquid jet. With a diameter of  $\sim 12 \mu\text{m}$  (FWHM) this pulse results in a peak intensity of  $4 \times 10^{14}$  W/cm<sup>2</sup>, which is suitable for water-window soft x-ray generation. The emitted x-ray photon flux was monitored at  $45^\circ$  angle to the incident laser beam with an x-ray diode (Hamamatsu G-1127-02) covered by a 1050 nm free-standing Ti filter. With this filter the diode signal is due to the N VI line at  $\lambda = 2.88$  nm (cf. Fig. 4).

The spectrum was recorded with a 10 000 lines/mm free-standing transmission grating<sup>19</sup> covered by a 250 nm thick Al filter to remove visible light. An x-ray sensitive CCD (Photometrics AT200L with Tektronix TK1024AB) was used as detector. Figure 3 shows the spectrum which has been corrected for the wavelength dependence of the grating, filter and CCD. The absolute efficiency of both the grating and the CCD were calibrated using synchrotron radiation from the Berlin electron storage ring (BESSY) thus allowing us to measure absolute photon numbers. With the geometry used, an exposure time of 60 s (utilizing the full CCD capacity) and a resolution of  $\lambda/\Delta\lambda \approx 100$  were achieved. In imaging applications, where zone plates are used, it is important

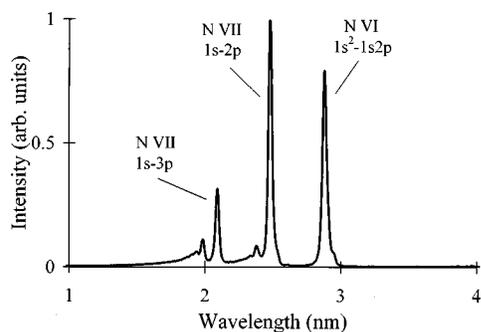


FIG. 3. Fully corrected spectrum from cryogenic liquid-jet nitrogen target.

to have a monochromatic light source in order to avoid chromatic aberrations. With our source this can be accomplished by using a titanium filter. Figure 4 shows the spectrum filtered by 700 nm Ti.

By integrating over the linewidth the photon flux for a line can be measured. This was done for the  $\lambda=2.88$  nm N VI line resulting in  $4.5 \times 10^{11}$  photons/(sr1.4 \times higher than what was achieved with ammonium hydroxide as target liquid<sup>20</sup> (32% NH<sub>3</sub> in water by volume) despite the fact that the jet diameter in the present work (5  $\mu$ m) is only half of what was used in Ref. 20. We will attempt to increase the photon flux by carefully designing the nozzle to allow laminar liquid nitrogen flow at larger diameters and consequently creating more target material in focus.

In many imaging applications, including microscopy, brightness of the x-ray source (photons/sr  $\times$   $\mu$ m<sup>2</sup>  $\times$  pulse) is more important than the total emitted photon flux. For this reason the size of the plasma was measured with an 8  $\mu$ m diameter pinhole camera, covered with a 700 nm Ti filter and with the same type of CCD detector as mentioned above. With full prepulse operation the source diameter (FWHM) at  $\lambda=2.88$  nm was measured to 35  $\mu$ m as shown in Fig. 5. This corresponds well to previously used room-temperature target liquids such as ethanol and ammonium hydroxide. In contrast to these liquids, however, the pinhole camera images from liquid nitrogen exhibit long radial tails of low-intensity x-ray emission (cf. Fig. 5). One probable reason for this is that the rapid evaporation of the liquid nitrogen results in a thin layer of N<sub>2</sub> gas surrounding the jet which contributes to the x-ray generation.

As mentioned in the introduction the liquid-jet target is a

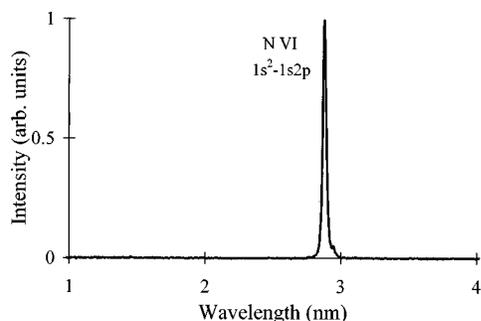


FIG. 4. Titanium-filtered single-line spectrum from cryogenic liquid-jet nitrogen target.

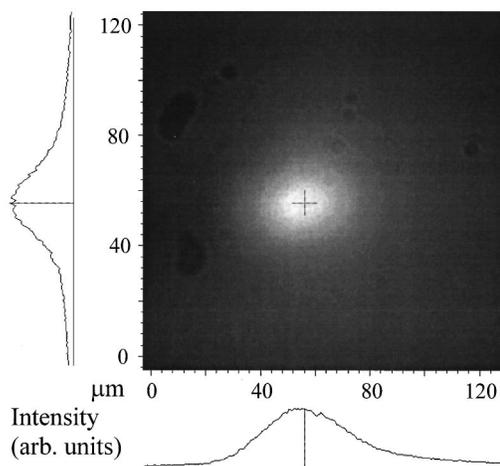


FIG. 5. Pinhole camera image at  $\lambda=2.88$  nm. Line scans indicate vertical and horizontal cross sections.

low-debris source. It is common to distinguish between atomic/ionic and particulate (here, frozen nitrogen or small liquid droplets) debris. The atomic/ionic debris results in a coating on, e.g., x-ray optical components positioned close to the source, thereby slowly degenerating the components. Particulate debris, due to its larger mass, may often directly damage optics and filters. We have previously shown that a size-optimized liquid target consisting of only gaseous components results in debris-free laser-plasma operation.<sup>6</sup> To verify the absence of particulate debris from the liquid-nitrogen source, thin metal filters (300 nm Ti) with a diameter of 5 mm were positioned  $\sim 60$  mm from the source. After several hours of 10 Hz plasma operation the filters showed no signs of pinholes or other damage.

Another important feature for an LPP is spatial stability. Therefore the jet stability was measured with an optical microscope 3 mm from the nozzle orifice (normal working distance for plasma production). At this distance the jet was found to be stable within the resolution of the microscope ( $\sim 6$   $\mu$ m). This is well within the size of the laser focus. Furthermore, the pulse-to-pulse fluctuations of the x-ray emission was estimated to  $\sim 25\%$ . This is the same value as we have previously achieved with ethanol as target liquid<sup>18</sup> and indicates that the liquid-nitrogen jet is spatially stable. The main reason for the fluctuations is pulse-to-pulse variations of the prepulse intensity. An unresolved problem is that the jet occasionally changes direction with several degrees. This only occurs when the laser is focused on the jet and the phenomenon is not fully understood yet. We have observed the same behavior when operating the metal nozzle with ethanol as target liquid. One possible explanation is a charging effect in combination with the metal nozzle.

#### IV. DISCUSSION

We have developed a soft x-ray source based on a cryogenic liquid-jet target. With nitrogen as target liquid this source is a promising x-ray source candidate for future tabletop x-ray microscopes. This source works for a full day of 10 Hz operation in a  $10^{-3}$  mbar vacuum chamber, i.e., no buffer

gas is necessary. A theoretical investigation of the hydrodynamic properties of liquid oxygen, xenon, and neon shows that these liquids may also be used as target material. Thus, the arrangement presented in this article is an interesting source also for proximity ( $\lambda \approx 1$  nm) and EUV ( $\lambda \approx 13$  nm) lithography. Compared to existing xenon target systems<sup>11,15</sup> our approach will allow for a potentially debris-free source with high brightness, long operating times with high-repetition-rate lasers and full vacuum compatibility.

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