Debris-free Single-line Laser-plasma X-ray Source For Microscopy

Rymell, L; Berglund, M; Hertz, H. M

Published in:
Applied Physics Letters

DOI:
10.1063/1.113105

1995

Debris-free single-line laser-plasma x-ray source for microscopy

L. Rymell, M. Berglund, and H. M. Hertz[a)]
Department of Physics, Lund Institute of Technology, P.O. Box 118, S-221 00 Lund, Sweden

(Received 19 January 1995; accepted for publication 3 March 1995)

A debris-free, narrow-bandwidth, single-line laser-plasma soft x-ray source has been developed suitable for water-window microscopy using zone-plate optics. The 10 Hz table-top source utilizes microscopic ammonium hydroxide droplets as target. The N VI emission at $\lambda=2.88$ nm is $\sim 1\times 10^{12}$ photons/(sr pulse). As an alternative to ammonium hydroxide, it is demonstrated that urea dissolved in water also produces strong emission from nitrogen ions, thereby extending the applicability of the droplet x-ray source to a new range of target materials. © *1995 American Institute of Physics.*

In soft x-ray microscopy, the natural contrast betwen carbon (e.g., proteins) and oxygen (water) in the water window ($\lambda=2.3-4.4$ nm) is utilized to allow high-resolution imaging of, e.g., unstained biological material in an aqueous environment. Imaging and scanning soft x-ray microscopes based on Fresnel zone plate optics require a narrow-bandwidth, high-brightness soft x-ray source.[1–3] Single-line emission and low continuum background is important to avoid image degradation. Emission in the lower wavelength range of the water window is advantageous since it minimizes water absorption. In this letter we show that laser-produced plasmas in ammonium hydroxide and urea-water droplets provide a table-top soft x-ray source with these properties.

To date, most work on x-ray microscopy is performed using synchrotron radiation sources.[1–3] Noncompact laser-plasma sources have been used for flash contact microscopy[4] and x-ray laser imaging microscopy.[5] However, table-top microscopes show promise for single-shot exposures and increased accessibility for applied researchers. Experiments with table-top sources include the use of a nitrogen-gas plasma focus[6] for water-window imaging microscopy,[7] a carbon-target laser plasma[8] for water-window scanning microscopy[9] and laser-plasma for higher-wavelength microscopy.[10] The high brightness, small size, and spatial stability make the laser-plasma sources attractive. Using conventional metal targets, conversion efficiencies of several tens of % may be reached with laser intensities of $\sim 10^{14}$ W/cm^2.[11] However, with solid state targets, the emission of debris may damage fragile x-ray optics positioned close to the plasma. A short distance between the plasma and the component is important to increase the useful x-ray flux since the plasma is an incoherent x-ray source. We have previously shown that debris is practically eliminated by using microscopic ethanol droplets as target.[12,13] With this target liquid, multiline emission from C V and C VI ions in the upper wavelength half of the water window is produced. Below we describe the use of ammonium hydroxide droplets as a target for 10 Hz debris-free generation of N VI ($\lambda=2.88$ nm) and N VII ($\lambda=2.48$ nm) radiation in the lower part of the water window. With proper filtering, a high-brightness, single-line, narrow-bandwidth source at $\lambda=2.88$ nm with low continuum background is produced. To our knowledge, there is only one previous investigation of x-ray sources for x-ray microscopy based on laser plasmas with a nitrogen target. In Ref. 6, a solid boron nitride target was illuminated by a 1 pulse/10 min J Nd laser resulting in, beside the debris issue, significant broadband emission superimposed on the N VI and V VII line emission. Boron nitride has also been used as a target for spectroscopic investigations.[14] Other solid target materials, such as nitrogen-rich organic salts (e.g., urea), have to our knowledge, not been investigated. In this letter, we show that practically debris-free droplet target operation can be extended to this class of materials by dissolving them in a suitable liquid.

The experimental arrangement for the ammonium hydroxide droplet-target soft x-ray source is shown in Fig. 1. Concentrated ammonium hydroxide (32% NH_3 in water by volume) at a pressure of ~ 40 atmospheres is forced through a piezoelectrically vibrated glass capillary nozzle, resulting in a stable train of ~ 10 μm diam droplets in the 10^{-4} mbar pressure vacuum chamber. The nozzle produces $\sim 10^6$ drops/s with a droplet velocity of ~ 50 m/s. A 50 mm focal length lens focuses the beam from a 10 Hz frequency-doubled, active/active/passive mode locked Nd:YAG laser (Continuum PYC61-C) to a FWHM of ~ 12 μm. The 100–120 ps pulses have an energy of 70 mJ, resulting in a focal spot intensity of $\sim 5\times 10^{14}$ W/cm^2. The small size and high speed of the droplets in combination with the 12 μm focal diameter makes spatial stability of the droplets and accurate temporal triggering of the laser important for a pulse to hit a single target droplet. Stable drop formation depends on the

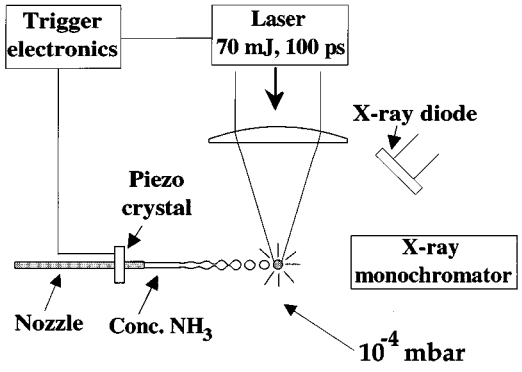

FIG. 1. Experimental arrangement for debris-free laser-plasma soft x-ray generation with ammonium hydroxide droplet target.

a)Electronic mail: hertz@lucas.lu.se

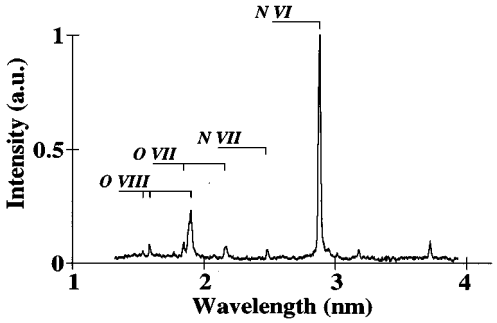

FIG. 2. Titanium filtered soft x-ray spectrum from ammonium hydroxide droplet-target laser plasma.

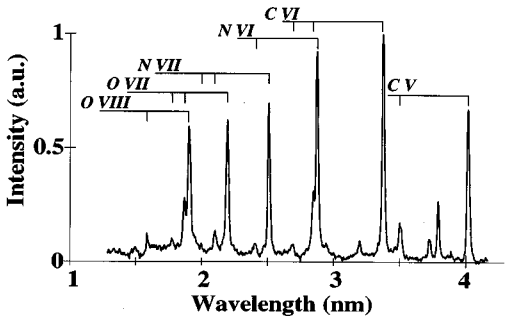

FIG. 3. Unfiltered spectrum from urea dissolved in water.

hydrodynamic properties of the liquid jet[15] and was achieved for several different piezoelectric vibration frequencies between 0.7 and 1.5 MHz. Correct triggering was obtained by synchronizing the *Q*-switch of the laser to the piezoelectric vibration signal. Droplets not used for plasma formation were collected in a liquid nitrogen trap.

The spectrum from the laser plasma was recorded with a 1 m grazing-incidence monochromator (Minuteman 301-G) blazed at $\lambda=2.9$ nm and a CsI-coated electron multiplier detector. The spectrum around the water window consists of characteristic line emission from highly ionized nitrogen (N VI and N VII) and oxygen (O VII and O VIII). In contrast to carbon laser-plasma targets, which emit C V and C VI radiation over the full water window,[8,12] the ammonium hydroxide target has its highest wavelength line at 2.88 nm, thus being suitable for thin-film metal filters having a sharp absorption-edge cutoff for higher-energy photons. Furthermore, carbon targets result in closely spaced lines (e.g., the $\lambda=3.50$ nm C V line is close to the dominant C VI line at $\lambda=3.37$ nm), which are very difficult to isolate spectrally.[8,12] As a demonstration, the droplet-target x-ray emission was filtered by a 300 nm titanium filter. The spectrum is shown in Fig. 2, where the $\lambda=2.88$ nm line dominates strongly. Due to the titanium L-absorption edge at $\lambda=2.72$ nm the transmission of the N VII line at $\lambda=2.48$ nm is negligible. The transmission of the $\lambda=2.88$ nm line was determined to be $\sim$60% while the photon flux from the O VII line at $\lambda=2.2$ nm was reduced to $\sim$7% of the unfiltered value. Should higher extinction of the low-wavelength oxygen emission be necessary, additional suppression of these lines may be obtained by combining the titanium with a thin layer of, e.g., chromium. The lines between $\lambda=3$ and 4.5 nm are second diffraction order lines of the oxygen ion emission. The experimentally recorded FWHM line width ($\Delta\lambda$) of the $\lambda=2.88$ nm line was determined by the monochromator's 0.009 nm spectral resolution. Thus, $\lambda/\Delta\lambda>300$, making the source suitable for zone-plate x-ray optics.[2,6] The broadband background in Fig. 2 is mainly due to interference from the monochromator's zeroth order, resulting in an estimated contrast between line intensity and continuum intensity of $>$100:1. The longer-wavelength x-ray flux is negligible.

The photon flux from the $\lambda=2.88$ nm line was determined by measuring the time integrated signal from x-ray diodes (Hamamatsu G-1127-02). Titanium filters covered the diodes and measurements were performed at 45° and 135° to the incident beam. In both directions the plasma emitted $\sim 1\times10^{12}$ photons/sr pulse. This number is expected to be correct within $\pm$50%.[12] The size of the soft x-ray source was determined with a pinhole camera. We estimate that $>$50% of the radiation was emitted from $\sim$15 μm kernel. Thus, the average integrated spectral brightness over the central kernel of the 10 Hz source is $\sim$0.1 μJ/(sr μm^2 pulse). This is approximately a factor 2 below what was achieved with a boron nitride solid target laser plasma source using a 1 pulse/10 min laser with 100 times higher pulse energy than ours.[6]

Debris production in droplet-target laser-plasma generation is several orders of magnitude lower than for comparable solid targets since no target material is presented in the low-intensity Gaussian tails of the focused laser beam. This has been shown for ethanol droplets, where solid carbon is the main debris substance.[13] It is anticipated that debris production from the ammonium hydroxide droplet target should be even lower since only gaseous compounds are present in the target. This was verified experimentally by collecting debris on carefully cleaned glass slides positioned 23 mm from the ammonium hydroxide plasma for $\sim$2 h of continuous 10 Hz laser-plasma operation. The glass slides were investigated with x-ray photoelectron spectroscopy (XPS)[13] by comparing photoelectron emission intensities from glass areas exposed and unexposed to the debris. The debris deposition was not measurable and determined to be $<$0.01 pg/(sr pulse), where the number is due to the noise in the measurements. This is two orders of magnitude less than for ethanol droplets, which, in turn, are $\sim$3 orders of magnitude cleaner than low-debris plastic tape targets.[13]

There are few alternative nitrogen-rich target liquids with suitable hydrodynamic properties allowing stable droplet formation[15] and, thus, debris-free laser-plasma operation. However, urea $[CO(NH_2)_2]$ is a harmless nitrogen compound that is soluble 1:1 by weight in water with little change in hydrodynamics compared to water. The spectrum from a urea droplet target is shown in Fig. 3. The higher background level is due to interference from long-wavelength emission scattered by the zeroth order of the monochromator since no filters were used. The photon flux from the N VI $\lambda=2.88$ nm line was $\sim 1\times10^{12}$ photons/(sr pulse). The debris deposition was mainly due to carbon and determined to $<$10 pg/(sr pulse) using XPS and optical absorption measurements.[13] Due to the higher-wavelength carbon lines, single-line N VI

or N VII operation is not equally easy to obtain as with the ammonium hydroxide target. Potentially, multilayer condenser optics could be used for spectral selection of, e.g., the $\lambda = 2.48$ nm line.[16] However, the important conclusion of this experiment is that it extends the applicability of laser-plasma droplet target operation to a new class of liquids, i.e., solutions. By dissolving solid substances in a suitable liquid, spectrally tailored x-ray emission is obtained.

The laser-plasma droplet target has many advantages compared to conventional laser-plasma targets, such as bulk solids or low-debris thin films. In summary, it has high brightness, produces narrow-bandwidth soft x-ray radiation suitable for, e.g., zone plates, provides fresh target drops for full-day operation without interrupts, allows excellent geometric access and reduces debris production several orders of magnitude compared to conventional targets. In this letter we have extended the applicability of the drop target to single-line emission in the lower range of the water window, making it useful for table-top soft x-ray microscopy. Although the brightness of the source does not allow single-shot x-ray imaging in its present state, the high rate of production of fresh target drops makes it suitable for high-repetition rate lasers (100 Hz[8] or higher[17]). With such lasers, the source would allow subsecond exposure times, and is ideally suited for samples which are not critically sensitive to radiation damage such as cryogenically cooled samples.[18] Furthermore, we work on improving the brightness by decreasing the plasma size and increasing the nitrogen content in the target liquid, aiming at single-pulse exposures. In addition, we have shown that laser-plasma droplet x-ray generation is possible with a new class of liquids, namely solutions. Thus, practically debris-free spectrally tailored emission from many new target materials is feasible.

The authors gratefully acknowledge discussions with and assistance from Johan Anderberg, Lars Malmqvist, Terje Rye (Siemens-Elema), Hans Stattin, and Sune Svanberg. This work was financed by the Swedish Natural Science Research Council, the Swedish Research Council for Engineering Sciences, and the Wallenberg Foundation.

[1] See, e.g., several papers, in *X-Ray Microscopy III*, edited by A. G. Michette, G. R. Morrison, and C. J. Buckley (Springer, Berlin, 1992).
[2] A. G Michett, Rep. Prog. Phys. **51**, 1525 (1988).
[3] J. Kirz, C. Jacobsen, and M. Howells, Quart. Rev. Biophys. (to be published).
[4] T. W. Ford, A. D. Stead, C. P. Hills, R. J. Rosser, and N. Rizvi, J. X-Ray Sci. Technol. **1**, 207 (1987).
[5] L. B. Da Silva *et al.*, Science **258**, 269 (1992).
[6] W. Neff, D. Rothweiler, K. Eidmann, R. Lebert, F. Richter, and G. Winhart, SPIE **2015**, 32 (1994).
[7] B. Niemann, D. Rudolph, G. Schmal, M. Diehl, J. Thieme, W. Meyer-Ilse, W. Neff, R. Holz, R. Lebert, F. Richter, and G. Herziger, Optik **84**, 35 (1990); M. Diehl, Ph.D. thesis, Georg-August Universität, Göttingen, 1993.
[8] H. Daido, I. C. E. Turcu, I. N. Ross, J. G. Ross, J. G. Watson, M. Steyer, R. Kaur, M. S. Schultz, and M. Amit, Appl. Phys. Lett. **60**, 1155 (1992).
[9] A. G. Michette, I. C. E. Turcu, M. S. Schultz, M. T. Browne, G. R. Morrison, P. Fluck, C. J. Buckley, and G. F. Foster, Rev. Sci. Instrum. **64**, 1476 (1993).
[10] J. A. Trail and R. L. Byer, Opt. Lett. **14**, 539 (1989).
[11] R. Kodama, K. Okada, N. Ikeda, M. Mineo, K. A. Tanaka, T. Mochizuki, and C. Yamanaka, J. Appl. Phys. **59**, 3050 (1986).
[12] L. Rymell and H. M. Hertz, Opt. Commun. **103**, 105 (1993).
[13] L. Rymell and H. M. Hertz (unpublished).
[14] A. M. Cantú, E. Jannitti, and G. Tondello, J. Opt. Soc. Am. **64**, 699 (1974).
[15] J. Heinzl and C. H. Hertz, Adv. Electron. Electron Phys. **65**, 91 (1985).
[16] G. Gutman, J. X-Ray Sci. Technol. **4**, 142 (1994).
[17] W. T. Silfvast, M. C. Richardson, H. Bender, A. Hanzo, V. Yanovsky, F. Jin, and J. Thorpe, J. Vac. Sci. Technol. B **10**, 3126 (1992).
[18] G. Schneider and B. Niemann, X-Ray Sci. **2**, 8 (1994).