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Published in:
Applied Physics Letters

DOI:
10.1063/1.116203

1996

Link to publication

Citation for published version (APA):
Malmqvist, L., Rymell, L., & Hertz, H. M. (1996). Droplet-target laser-plasma source for proximity x-ray lithography. *Applied Physics Letters*, *68*(19), 2627-2629. https://doi.org/10.1063/1.116203

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Droplet-target laser-plasma source for proximity x-ray lithography

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(Received 26 January 1996; accepted for publication 2 March 1996)

A compact, high-brightness and practically debris-free laser-plasma soft x-ray source for proximity x-ray lithography is described. The target of the source is small liquid fluorocarbon droplets injected into vacuum with a piezoelectrically vibrated nozzle. Emission from helium- and hydrogenlike fluorine in the 1.2–1.7 nm wavelength range was determined to $\sim 2\times10^{12}$ photons/(sr-pulse), which corresponds to a conversion efficiency of ~5% of the 70 mJ laser pulse. Exposure of a copolymer of PMMA–MAA confirms the measured photon flux. Debris production was approximately 70 pg/sr pulse. The applicability of the source for dedicated lithography systems is discussed. © *1996 American Institute of Physics.* [S0003-6951(96)02719-2]

Proximity x-ray lithography has shown significant promise for nanometer-scale lithography.[1–3] The synchrotron storage ring may be a suitable x-ray source for large-scale facilities but for many applications there is a need for compact granular sources. The laser-plasma x-ray source is a strong candidate for this due to its high brightness, high spatial stability, compact size, and relatively low cost. However, with conventional solid targets they suffer from disruptive interrupts due to changes of target and severe debris problems, which may damage, e.g., the lithographic mask. In this letter we demonstrate that the use of small liquid droplets as target permits extended high-brightness operation without interrupts at suitable x-ray wavelengths and practically eliminates the debris problem.

Granular x-ray lithography systems must rely on a compact and inexpensive x-ray source, such as an electron impact device, pinch plasma, or laser-produced plasma.[4] Electron impact sources are limited in output power. Pinch plasma sources produce a high single-pulse output power but the low repetition rate, spatial instabilities, and debris emission limit their applicability. The laser plasma is a spatially stable, high-brightness and potentially high-repetition-rate compact x-ray source, which has been shown to be suitable for x-ray lithography.[5–7] With conventional metal targets, conversion efficiencies of several tens of percent have been achieved.[8] However, with such targets, laser plasmas emit significant amounts of debris[6,9] which may coat or damage, e.g., sensitive x-ray lithography masks. The debris production may be somewhat lowered by the use of thin film tape targets[10] and the effect of debris may be partially reduced by employing a backing pressure[6] or a fast shutter system.[11] Still, the debris deposition remains intolerably high. However, with small liquid droplets as target, the debris production is lowered by 3–4 orders of magnitude compared to thin film tape targets of the same composition as the drops, while still maintaining high x-ray brightness.[12,13] The versatility of the droplet-target technique is illustrated by its applicability to projection lithography[14] and microscopy,[15] and that it also allows spectral tailoring of the emitted x-ray wavelengths.[15] To be suitable for proximity x-ray lithography the radiation from the plasma should be in the wavelength range of 0.7–1.7 nm.[6] The line radiation from hydrogenlike and heliumlike fluorine is in this range. In the present letter we show that the laser-plasma generation in fluorine-containing droplets is a suitable source for proximity x-ray lithography. Fluorine ion laser-plasma x-ray emission has been studied previously with a SF_6 gas puff target.[16]

The experimental arrangement is shown in Fig. 1. Liquid fluorocarbon (C_n,F_m, where n is typically 5–10 and m is typically 10–20) is forced through a piezoelectrically vibrated capillary glass nozzle under a pressure of ~40 bar, creating a stable train of droplets in the vacuum chamber. The approximately 10-μm-diam nozzle produces ~15-μm-diam droplets separated by ~50 μm and at a rate of $\sim10^6$ drops/s. Since only 10 drops/s are used for the plasma generation, a liquid-nitrogen trap collects the excess liquid and maintains the pressure at 10^{-4} mbar. Laser plasmas are produced by focusing the beam from a 10 Hz frequency-doubled active/active/passive mode-locked Nd:YAG laser (Continuum PY61C-10) onto the droplets. The laser is temporally synchronized with the nozzle piezoelectric vibration frequency in order to ensure that each laser pulse hits a single droplet. The $\lambda=532$ nm, 70–80 ps, 70 mJ pulses are focused with a 50 mm focal length lens to a full width at half maximum (FWHM) diameter of ~12 μm, resulting in an intensity of $\sim8\times10^{14}$ W/cm^2 at the target droplet. In order to increase the x-ray emission, the laser was operated with an

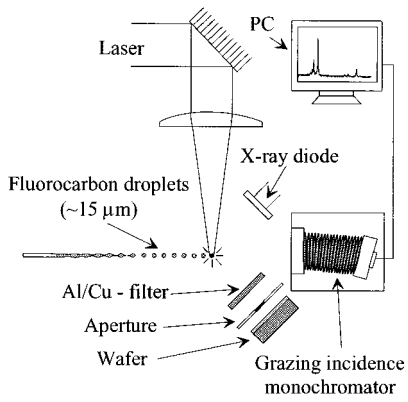

FIG. 1. Experimental arrangement for droplet-target laser-plasma source for proximity x-ray lithography.

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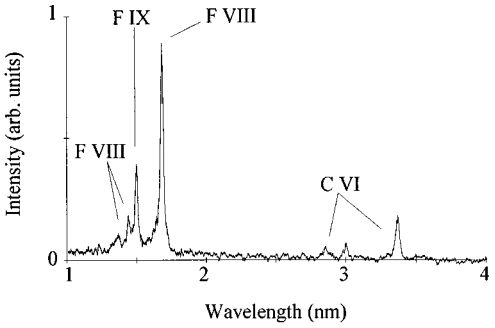

FIG. 2. Aluminum/copper filtered x-ray spectrum from fluorocarbon droplet-target laser plasma.

approximately 10% prepulse, 7 ns before the main pulse. The droplet is highly ionized, creating a plasma emitting x rays mainly as line radiation but also in a low continuum background. The spectrum of the emitted radiation was recorded with a 1 m grazing incidence monochromator (Minuteman 301-G), which was blazed at $\lambda=2$ nm, had a maximum resolution of 0.02 nm, and was equipped with a CsI photocathode electrode multiplier detector. The spectrum in the $\lambda=1-4$ nm range is shown in Fig. 2. Here a free-standing sandwiched metal filter (100 nm copper/100 aluminum) was placed in front of the monochromator to suppress radiation from lines above $\lambda\approx1.7$ nm. The spectrum has not been corrected for the wavelength dependence of the monochromator and the detector. The measured radiation is dominated by line radiation from F IX at $\lambda=1.495$ nm and F VIII at $\lambda=1.681$ nm and $\lambda=1.446$ nm. These are the lines we use for the lithography. The longer-wavelength emission from C V and C VI starting at $\lambda=2.603$ nm is observable but suppressed by the Cu/Al filter. Note that several of the C VI lines in Fig. 2 are superimposed on second-order lines from F IX and F VIII.

The emitted x-ray flux from the plasma was measured with an x-ray diode (Hamamatsu G-1127-02) through free-standing filters of 1.6 μm aluminum, 100 nm titanium, and 20 nm gold. This filter was chosen to suppress longer-wavelength lines and to provide sufficient attenuation to ensure a nonsaturated response of the diode. The source flux below $\lambda=1.7$ nm was determined to be $\sim2\times10^{12}$ photons/(sr-pulse). This corresponds to 0.06 mJ/(cm^2-pulse) at an exposure distance of 20 mm. Assuming that the $\lambda<1.5$ nm lines and the $\lambda=1.681$ nm line carry equal flux, the x-ray conversion efficiency to the spectral region below $\lambda=1.7$ nm is $\sim5\%$. The uncertainty in these measurements is about 50% due to the uncertainty in diode sensitivity, line strength, and filter thickness.[12]

To confirm our measurements of the x-ray intensity, test exposures of a copolymer of polymethyl methacrylate and methacrylic acid (PMMA–MAA) were performed. The required dose incident on the resist was determined from literature data on copolymer sensitivity[17] and x-ray absorption coefficients,[18] and found to be approximate 400 mJ/cm^2. The number has been increased to compensate for the x-ray absorption in the resist which results in a lower x-ray intensity at the bottom of our 0.5-μm-thick resist. The calculated dose was confirmed by exposing the copolymer at a distance of 20 mm from the droplet x-ray source through 1.6 μm aluminum and 100 nm copper filters. The filters had a total transmission of $\sim25\%$ in the $\lambda<1.7$ nm range while the transmission of the longer-wavelength carbon lines was less than 1%. After the exposure the resist was developed in methyl-iso-buthyl-ketone (MIBK) dissolved in isopropyl alcohol (1:3) for 90 s, resulting in a fully developed resist. The exposure time was 1 h, which corresponds to an incident dose on the resist of approximately 500 mJ/cm^2. Thus, the required dose was $\sim30\%$ higher than the calculated dose, which is within uncertainty in predicted data from the literature and the error in our x-ray flux measurements.

In order to estimate the exposure time for a realistic lithography system based on the droplet source, mask absorption and source–wafer distance should be considered. The 1 μm silicon nitride masks have several advantages[3] and are a suitable choice for the droplet source. The transmission through 1 μm silicon nitride is 20% at $\lambda=1.7$ nm and 32% at $\lambda=1.5$ nm. The transmission of the longer wavelength lines is low ($<1\%$ above $\lambda=2.6$ nm), thereby providing additional suppression of longer-wavelength carbon-ion emission from the source. The minimum source–wafer distance suitable for exposure may be limited by the overlay errors to ~200 mm.[4] With these numbers, the exposure time can be calculated to approximately 100 h using the present 10 Hz source. However, lasers with repetition rate of up to 1 kHz are currently developed,[11] thus shortening the exposure time by a factor of 100. The droplet-target method is well suited for such high-repetition-rate operation since it generates $\sim10^6$ fresh target droplets every second. Using conventional targets, such high repetition rates would result in frequent interrupts in order to change the target arrangement. Furthermore, the use of a more sensitive resist (e.g., SAL-601) may shorten the exposure time by an additional factor of 20.[18,19] Thus, the total exposure time would be on the order of 1 min. This exposure time is reasonable for small-scale fabrication of devices, especially considering that it might be further reduced employing collector optics.[20] Furthermore, the droplet-target laser plasma allows nearly 4π steradian geometric access and thus allows parallel processing of several wafers.

The achievable lithographic resolution is, among other factors, dependent on several source parameters. The wavelength range of the emitted fluorine lines (primarily $\lambda=1.5$ and 1.7 nm) is at the high end of the spectral range suitable for proximity x-ray lithography. This has the advantage that the photoelectron range in the resist is significantly smaller than for the shorter wavelengths.[3] The disadvantage is that the width due to Fresnel diffraction is approximately 40% larger than at $\lambda=0.8$ nm. However, recent detailed studies indicate that resolution is not limited by diffraction for linewidths above 50 nm.[3] Another factor determining linewidth is the penumbral blur[4] which is due to the size of the source and the spatial source stability. For the droplet-target laser plasma described here, these contributions are negligible since the spatial stability of the plasma is a few micrometers and the diameter is a few tens of micrometers.[14]

Debris is a major problem in laser-plasma x-ray lithography since it may damage or coat, e.g., the mask or collection optics. Quantitative debris measurements on the fluorocarbon droplet-target laser plasma were performed following

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Ref. 13. Gold-coated glass slides were positioned 20 mm from the plasma to collect debris for 15 min of 10 Hz operation. The debris formed a layer on top of the gold which was examined by x-ray photoelectron spectroscopy (XPS). The debris layer mainly consisted of fluorine and carbon (1:2 ratio). By measuring the attenuation of the $4f$ gold photoelectron signal in different angles (angle-resolved XPS), the thickness of the layer was determined to be 0.7 nm. Assuming a density of 2 g/cm^3, this corresponds to a debris deposition rate of approximately 70 pg/sr pulse. This determination is assumed to be correct within $\pm 50\%$. Thus, the droplet target debris production is 2–6 orders of magnitude less than those reported for tape or conventional targets.[9,13] It is interesting to note that it is approximately a factor of 15 higher than the debris emission from ethanol droplets.[13] This difference may be explained by the higher density, larger volume, and higher solid content of the fluorocarbon droplets compared to the ethanol droplets. The described debris-deposition measurements show that the combined emission of particulate and atomic/ionic debris is very low. However, the particulate debris is especially harmful since such projectiles may severely damage, e.g., the x-ray lithography mask.

Particulate debris was examined by exposing a free-standing 100-nm-thick aluminum filter positioned 20 mm from the plasma to debris during 1 h of 10 Hz operation. Within the 1 mm^2 observation area no new pinholes were produced. Thus, we conclude that the emission of harmful larger fragments is negligible. The low debris production of the droplet-target laser plasma is probably due to the fact that the full target material is ionized, thereby eliminating the production of larger debris particles. Thus, thin film filters may be used for long-term operation in order to eliminate the residual debris. Furthermore, additional shielding can be achieved using a localized gas shield.[13]

Finally, we will briefly discuss two practical aspects of granular x-ray lithography systems, namely noninterrupted operating time and the cost of the target. Conventional target systems, e.g., thin film tapes or rotating cylinders, are not suitable for high-repetition-rate operation due to their limited time of operation between interrupts, which are necessary to exchange target material. In contrast, the operating time for the droplet-target system is practically unlimited and only restricted by the volume of the liquid container. Also from a cost-of-target perspective, the droplet target is advantageous since there is, e.g., no cost of preparing (e.g., polishing or forming) the target surface, since this is intrinsic in the drop-formation process. For the fluorocarbon droplets used in this work, the major cost is due to evaporation of the liquid in the vacuum chamber. We have estimated that approximately 10% of the injected liquid evaporates resulting in an approximate target cost on the order of $\$10^{-7}$/shot for a 1000 Hz repetition rate system. Thus, this cost is below the challenging design goals calculated for other types of lithography.[21]

To conclude, we have developed a compact high-brightness laser-plasma source suitable for proximity x-ray lithography in the 1.2–1.7 nm wavelength range. The source practically eliminates the debris problem, allows high-repetition-rate operation without interrupts, provides 4π steradian geometric access and a high spatial stability. The conversion efficiency is approximately 5% and the projected exposure time for a dedicated system is on the order of 1 min.

The authors gratefully acknowledge valuable discussions with and assistance from Magnus Berglund, Derrick Mancini, Kristina Georgsson, and Sune Svanberg. This work was financed by the Swedish Natural Science Research Council, the Swedish Research Council for Engineering Sciences, the Swedish Board for Industrial and Technical Development, and the Wallenberg Foundation.

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