Source characterization for x-ray proximity lithography

K. Gabel, M. Richardson, M. Kado, and A. Vassiliev

Laser Plasma Laboratory, Center for Research and Education in Optics and Lasers, University of Central Florida, 12424 Research Parkway, Suite 400, Orlando, Florida 32826

Received June 14, 1994

Calibrated x-ray spectra from laser-produced plasmas of materials with atomic numbers varying between 12 (Mg) and 83 (Bi) were recorded to optimize the conversion efficiency for proximity lithography in a 0.5-nm band centered at 1 nm. The highest efficiency (\sim 0.8%) was found for *L*-shell emitters such as Cu and *M*-shell emitters such as Ba. First-order debris measurements were carried out by measurement of the layer thickness deposited on witness plates 2 cm away from the target. Layers of 30-nm thickness were deposited in a single laser shot with Au and W targets.

Proximity x-ray lithography is one of the leading approaches to the fabrication of future high-density computer chips. Laser plasmas are increasingly being considered as an attractive x-ray source for this approach because of their compact size, flexibility, and cost.¹ The complex interplay between mask opacity and energy absorption in the mask and the resist currently sets the optimum x-ray wavelength at ~1 nm.

To our knowledge, a full examination of the optimum laser and target parameters for a productionline x-ray facility, in which x-ray conversion efficiency is evaluated together with the undesirable effects of target debris and the relative cost of long-term operation, has not yet been made. In this Letter we report concurrent absolute x-ray conversion efficiencies and first-order debris measurements for a range of target materials under laser conditions currently deemed to be achievable in a high-repetition-rate (>10-Hz) reliable source.²

The experimental setup is shown in Fig. 1. Plasmas were created by a Nd:glass laser delivering 15 J of energy in 17 ns and a laser intensity of 5×10^{12} W/cm² on target. The conversion efficiency to x rays was estimated with the use of a high-resolution crystal spectrograph. Targets were selected according to conversion factors obtained with a Be-filtered x-ray diode for an x-ray energy exceeding 1.5 keV (Ref. 3) to be representative for x-ray emission of different atomic shells.

To obtain absolutely calibrated x-ray spectra within a 0.5-nm bandwidth centered at 1 nm, we used both rubiduim acid phthalate (RAP) and potassium acid phthalate (KAP) crystals with known integrated reflectivity⁴ and absolutely calibrated Kodak DEF x-ray film.⁵ Thus we determined the absolute x-ray emission according to the following equation:

$$E_{\rm S} = \frac{2\pi r}{\Delta h_d R_{\rm int}} \Delta h_d \int_{\rm det} \mathrm{d}w_d \frac{e_d[\lambda(w_d)]}{t[\lambda(w_d)]}, \qquad (1)$$

where E_s is the x-ray energy emitted by the source within the 0.8–1.3-nm band, e_d is the energy density on the detector, r is the source-detector distance, dw_d is the size of the detector element in the dispersion plane of the crystal, Δh_d is the detector size perpendicular to the dispersion plane, $R_{\rm int}$ is the integrated crystal reflectivity, and $t(\lambda)$ is the filter transmission.

The integral was solved by numerical integration of densitometer traces. Tabulated data of absorption coefficients⁶ have been fitted with a third-degree polynomial to account for the wavelength-dependent spectrograph response arising from the x-ray transmission through light protection foils (Be, Al). To convert optical density into x-ray energy density, we used a two-dimensional polynomial fit (energy density on the detector as a function of wavelength and optical density) of Henke's calibration data for Kodak DEF film.⁵ The wavelength dependence of the integrated reflectivity $(R_{\rm int})$ in the band of interest is rather weak (7–9 \times 10⁻⁵ for KAP; 1–1.4 \times 10⁻⁴ RAP). Hence we assumed an average wavelengthindependent value for the reflectivity for the whole band.

Table 1 summarizes these elements and shows the emission of different atomic shells in the 0.8-1.3-nm band along with the elements/compounds used in our experiments.

Table 2 shows our experimental results for the conversion efficiency of laser energy into x-ray energy for different Z elements. The highest conversion efficiency was obtained for x-ray emission from a Cu plasma. The dominant emission features are the 3-2 transitions in Ne-like ions.⁷ The lower conversion efficiency observed for Fe results from the 4-2 transitions in Ne-, F-, and O-like ions with lower oscillator strengths.^{8,9} Figure 2 shows recorded spectra of x-ray emissions illustrating the strong 3-2 emission features of a Cu plasma and the 4-2 transitions of an Fe-plasma, respectively.



Fig. 1. Experimental setup used to characterize x-ray emission and debris from laser-produced plasmas.

	Atomic Shell					
	K	L	М		N	
Z of elements with emission partially covering 0.8–1.3-nm band ^a Z of elements with emission covering whole	7–14	23–37	48–68		>78	
0.8–1.3-nm band ^a Elements/compounds used	10, 11 ¹² Mg	28–31 ²⁶ Fe ³⁰ Cu ³¹ Zn	56—58 ⁵⁰ Sn ⁵⁶ Ba (BaCl)	^{74}W	⁷⁹ Au ⁸² Pb ⁸³ Bi	

Table 1. Elements That Emit in the 0.8–1.3-nm Band andthe Targets Used to Measure Conversion Efficiencies

^aRef. 3.

The lower conversion efficiency observed for Zn can be explained on the basis of its different ionization balance. Since the dominant emission of both elements in the 0.8-1.3-nm band is caused by Ne-like ions, a potential explanation for the lower conversion efficiency from Zn is that there is a smaller fraction of this ion in the plasma. For a collisionally dominated plasma the ratio of Ne- to Na-like ions is derived from the Saha equation. By using the ionization potential of the Na-like ion of each element (670 eV for Cu and 737 eV for Zn) and an electron temperature in the 150-250-eV range we obtain a 1.3-1.55 higher fraction in the Cu plasma, which indicates a larger Ne-like component in the Cu plasma and hence less emission from Ne-like Zn. To consider the influence of laser-plasma interaction dynamics on the electron energy distribution, we estimated thresholds for stimulated Raman scattering, two-plasmon decay, and nonlocal energy transport as possible sources of hot-electron production. The only process with a threshold $(5 \times 10^{12} \text{ W/cm}^2)$ close to our experimental conditions is two-plasmon decay. Since we could not detect any indication of $3/2\omega$ light, we believe that even two-plasmon decay is of no importance in our experiments. At an incidence angle of 45° resonance absorption is negligible for our intensity and laser wavelength.

Another promising candidate for having a high conversion efficiency in the 0.8–1.3-nm band is Ba. Our experimental value was obtained with a BaCl target. The simple assumption of two-times-higher emitter density in the case of a pure Ba plasma leads to a conversion efficiency of ~0.8%, which is as high as the conversion efficiency for Cu. Only poor efficiency was obtained from high-Z elements (the highest value was for a Bi target).

After the laser pulse has ceased the ions stream outward with energies acquired during the interaction process. The plasma cools, while further ablation of the material occurs; thus a number of neutral atoms are ablated. The ablation process drives a shock wave in the solid target, producing a crater along with a variety of particulate emission (molten target material and clusters).^{10,11} Hence high-velocity ions, atoms, and molten target material and clusters are a by-product of the laser-produced plasma.

Debris produced by the plasma from various targets was collected on witness plates located 2 cm from

the source. For every target material investigated we found particles several micrometers in size deposited on top of a thin evenly coated layer. We attached an aperture to the top of the witness plates in the experiments with W and Au targets. This aperture created an edge between exposed and unexposed areas, permitting a determination of the layer thickness with an atomic-force microscope. Figure 3 shows the topogram of the boundary region between the Si substrate and the deposited W layer. The layer thickness is \sim 30 nm, and it is approximately the same for Au deposition. Both layers were deposited in a single laser shot. W and Au were chosen because of their different melting points (3410 and 1064 °C, respectively), but they produced approximately the same coating thickness on the witness plate. Hence we estimate to first order that the same amount of debris is deposited for all targets selected. Based on the atomic-force microscope data for the laver thickness and assuming coatings of solid density, we estimate a deposition rate of 220 μ g/sr per pulse. This deposition rate exceeds by a factor of ~ 400 that reported in Refs. 12 and 13, which was obtained for a laser energy of 3 J. It is known that the amount of debris scales with laser energy,¹⁴ with the deposition rated varying from 15 to 500 ng/sr per pulse for energies of 300 mJ and 3 J, respectively.¹³ Hence we account for our high deposition rate by citing the five-times-higher laser energy. Moreover, we obtained the layer thickness from atomic-force microscope scans, which are not sensitive to the mass density of the deposited material, whereas the results reported in Refs. 11 and 12 were obtained from extinction of light in the coatings. The two different

Table 2. Measured Conversion Efficiency of Laser Energy into 0.8-1.3-nm X Rays

Element	Target Material	Atomic Shell	Conversion Efficiency (%)
¹² Mg	Mg	K	0.05
²⁶ Fe	Fe	L	0.25
³⁰ Cu	Cu	L	0.8
³¹ Zn	Zn	L	0.3
$^{50}\mathrm{Sn}$	Sn	M	0.35
⁵⁶ Ba	BaCl	M	0.4
⁸³ Bi	Bi	N	0.1



Fig. 2. L-shell emission from (a) Cu plasma and (b) Fe plasma.



Fig. 3. Atomic-force microscope topogram of the Si-W-on-Si interface of a witness plate obtained in a single laser shot (Si substrate on the left-hand side).

methods lead to discrepancies if the density of the coatings differs from solid density.

From our initial experiments with laser-plasma sources for proximity lithography we conclude that Lshell emitters such as Cu and M-shell emitters such as Ba are promising candidates to obtain high conversion into x-rays at 1 nm with a 0.5-nm bandwidth. Our Cu data agree with recently obtained Lawrence Livermore National Laboratory data for 1- μ m laser wavelengths although that group's results indicate higher conversion efficiency for 2ω light.¹⁵

From the 30-nm-thick layer coated on a witness plate at a 2-cm distance, a 0.075-nm buildup at 40 cm is estimated. This implies a deposition of 2.7 μ m/h for a source with a 10-Hz repetition rate, exceeding the tolerable buildup by approximately a factor of 1000. From the scaling of debris production with laser energy obtained by comparing our results with Refs. 11 and 12, we suggest that the necessary laser intensity on target be obtained by means of a tighter focus and reduced pulse duration rather than by means of increased energy. Source operation at lower energy can be achieved at higher repetition rates to provide the required throughput of a production line. In addition, considerable debris reduction is obtained with a He background gas to reduce the number of ejected ions and with thin tapes to direct cluster emission to the rear surface of the target.^{12,13}

We thank Ch. Brown and K. Abbott of the Naval Research Laboratory for their support in providing densitometer traces of our x-ray films. We acknowledge helpful discussions with H. Bender of the Center for Research and Education in Optics and Lasers regarding debris collection. We are grateful to R. Eby of Topometrix for providing us with atomic-force microscope data of our witness plates and thank J. Nilsen of Lawrence Livermore National Laboratory for helping us to find wavelength references. This study was supported in part by the State of Florida and the Advanced Research Projects agency through contracts from Lawrence Livermore National Laboratory and Sandia National Laboratories under contract B192600. K. Gabel is supported by the Deutsche Forschungsgemeinschaft of Germany, and M. Kado is supported by the Japanese Society for the Promotion of Science.

References

- 1. D. J. Nagel, R. R. Whitlock, J. R. Greig, R. E. Pechacek, and M. C. Peckerar, "Laser-plasma source for pulsed x-ray lithography," NRL Mem. Rep. 5731 (Naval Research Laboratory, Washington, D.C., 1987).
- C. B. Dane, L. E. Zapata, and L. A. Hackel, in *Conference on Lasers and Electro-Optics*, Vol. 11 of 1993 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1993), paper CWI1, p. 274.
- 3. T. Mochizuki and C. Yamanaka, Proc. Soc. Photo-Opt. Instrum. Eng. 733, 23 (1986).
- 4. A. Burek, Space Sci. Instrum. 2, 53 (1976).
- B. L. Henke, J. Y. Uejio, G. F. Stone, C. H. Ditttmore, and F. G. Fujiwara, J. Opt. Soc. Am. B 3, 1540 (1986).
- 6. B. L. Henke, E. M. Gullikson, and J. C. Davis, Preprint LBL-33908 (Center for X-ray Optics, Lawrence Berkley Laboratory, Berkeley, Calif., 1993).
- R. J. Hutcheon, I. Cooke, M. H. Key, C. L. S. Lewis, and G. E. Bromage, Phys. Scr. 21, 89 (1980).
- V. A. Boiko, A. Ya. Faenov, and S. A. Pikuz, J. Quant. Spectrosc. Radiat. Transfer 19, 11 (1978).
- H. Gordon, M. G. Hobby, and N. J. Peacock, J. Phys. B 13, 1985 (1980).
- 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).
- T. Trucano, D. Grady, R. Olson, and A. Farnsworth in Soft X-Ray Projection Lithography, Vol. 18 of OSA Proceedings Series (Optical Society of America, Washington, D.C., 1993), p. 146.
- F. Bijkerk, E. Louis, M. J. van der Wiel, E. C. I. Turcu, G. C. Tallents, and D. Batani, J. X-Ray Sci. Technol. 3, 133 (1992).
- E. Louis, F. Bijkerk, G. E. van Dorsen, and E. C. I. Turcu, in *Proceedings of 21st ECLIM* (Institute of Plasma Physics and Laser Microfusion, Warsaw, Poland, 1991), p. 261.
- 14. M. L. Ginter and T. J. McIlrath, Appl. Opt. 27, 885 (1988).
- 15. L. A. Hackel, B. Dane, M. Norton, L. Zapata, D. Matthews, L. de Silva, and J. Trebes, "Pointsource lithography at 0.25 μ m using solid-state laser-generated x-rays," presented at the Advanced Research Projects Agency Point Source/Stepper Workshop, Lyons, Colo., October 6–7, 1993.