Ultimate Efficiency of Extreme Ultraviolet Radiation from a Laser-Produced Plasma

Tatsuya Aota and Toshihisa Tomie*

National Institute of Advance Industrial Science and Technology 1-1-1 Umezono, Tsukuba, Ibaraki, 305-8568, Japan

(Received 9 May 2004; published 7 January 2005)

An analytical formula for maximizing radiation efficiency from a laser-produced plasma is derived. The maximum efficiency is achieved when the plasma expansion distance during laser heating is equal to the laser absorption length. The dependence of the radiation efficiency on the plasma density is confirmed by experiments using a particle-cluster target. By creating a relatively uniform density plasma with a 300 μm diameter by dispersing SnO_2 particles coated on a Si wafer, the conversion efficiency at 14 nm, as high as 4 times as that for a Sn plate target, is achieved.

DOI: 10.1103/PhysRevLett.94.015004 PACS numbers: 52.50.Jm, 33.70.Fd, 52.25.Os, 52.70.La

A laser-produced plasma (LPP) created on a solid target was found to generate strong x rays with photon energies from a few tens eV to several keV in the very early days of LPP study [1,2]. Numerous efforts to increase the conversion efficiency (CE) from laser energy to x-ray energy have been conducted for applications in x-ray radiography and x-ray lithography [3–6]. Presently, LPP is studied actively as an extreme ultraviolet (EUV) source at 13–14 nm for EUV lithography (EUVL) [7]. EUVL is the technology of printing 45 nm and even smaller features of semiconductor devices and is expected to be employed in full production by 2010. A detailed study on the CE of Sn, the best element for 13–14 nm emissions, was performed in the mid 1990s, and the maximum CE was reported to be around 1.5% [8]. Achieving the CE larger than 3% from the pumping laser energy to usable EUV energy at 13–14 nm within a 2% bandwidth into 2π sr solid angle is considered vital in order to make EUVL economically affordable [9]. Therefore, we need to clarify how large is the ultimate CE and how we can realize it. Through 30 years of efforts to improve the CE, almost all available experimental parameters, such as irradiation density, pulse duration, laser wavelength, target element, focused-laser spot diameter, and so on, have been scanned. For example, it is now very common to use a prepulse since the first discovery of its effectiveness [10]. However, efforts to increase the CE so far have been empirical only, in a sense, and there is no theory which leads us to design novel systems of achieving the ultimate CE.

In this Letter, we derive a simple analytical formula for maximizing the CE. We also report an experiment with a newly devised particle-cluster target to confirm the theory. Previous reports are also interpreted using the derived theory.

We can assume a laser light is absorbed in a LPP by inverse bremsstrahlung absorption. When resonance enhancement of the radiation field is neglected, the absorption coefficient is given by [11] $\alpha = aZn_e^2$, with $a = 6 \times 10^{-37} \lambda_{\rm laser}(\mu{\rm m})^2/T_e({\rm eV})^{3/2}({\rm cm}^5)$, where Z is the charge of ion, n_e is the electron density, $\lambda_{\rm laser}$ is the laser wavelength in μm, and T_e is the electron temperature in eV. We assume that radiation is emitted via deexcitation of levels excited by electron collision. Then, photon flux per ion $p_{\rm rad}$ is given by $p_{\rm rad} = bn_e$, where b is given by [12] $b = 3.2 \times 10^{-6} f/[\Delta E({\rm eV}) T_e({\rm eV})^{1/2} \times \exp(\Delta E/T_e)]({\rm cm}^3/{\rm sec})$. Here, f and ΔE are the ensemble averaged oscillator strength and the energy of excitation for radiation with the photon energy of $h\nu$. The ensemble averaged ionization stage is assumed to be determined solely by instant T_e by neglecting transient effect. When heat conduction is neglected, a laser absorbing layer having a thickness of $L_{\rm abs} = 1/\alpha$ can be considered as the emitting layer. Then, the total number of ions $N_{i,\rm emit}$ in the emitting volume is $N_{i,\rm emit} = (n_e/Z)SL_{\rm abs}$, where S is the area of the plasma, and the radiation energy $E_{\rm rad}$ of photon $h\nu$ for a pulse duration of $t_{\rm laser}$ is given by $E_{\rm rad} = h\nu t_{\rm laser} b n_e N_{i,\rm emit}$ when self-absorption is neglected. The energy $E_{\rm plasma}$ for creating a plasma is given by $E_{\rm plasma} = (\chi_{\rm sum} + ZT_e)N_{i,\rm total}$. Here, $\chi_{\rm sum}$ is the sum of ionization energies to create an ion of charge Z from a neutral atom, and $N_{i,\rm total}$ is the total number of ions including those outside the emitting volume. The radiation efficiency $\eta_{\rm rad}$ of photon with photon energy $h\nu$ is given by

$$\eta_{\rm rad} = E_{\rm rad} \Big/ \left(\sum E_{\rm rad} + E_{\rm plasma} \right) = h\nu b t_{\rm laser} n_e N_{i,\rm emit} \Big/ \left\{ t_{\rm laser} n_e N_{i,\rm emit} \Sigma(h\nu b) + (\chi_{\rm sum} + ZT_e) N_{i,\rm total} \right\}. \quad (1)$$

Here, $\sum E_{\rm rad}$ is the summation over the whole radiation.

The laser absorbing layer expands with time constant $t_{\rm exp} = L_{\rm abs}/V_{\rm exp}$. Here, lateral expansion is neglected. Plasma expansion velocity $V_{\rm exp}$ is given by [13] $V_{\rm exp} = (Z/M)^{1/2} T_e({\rm eV})^{1/2} 1.6 \times 10^6$ cm/sec, where M is the atomic mass number of the element. When $t_{\rm laser} < t_{\rm exp}$, plasma motion is neglected, and

$$N_{i,\rm total} = N_{i,\rm emit}. \quad (2)$$

Then, Eq. (1) becomes

$$\eta_{\rm rad} = h\nu b t_{\rm laser} n_e / \{t_{\rm laser} n_e \Sigma(h\nu b) + (\chi_{\rm sum} + ZT_e)\}. \quad (3)$$

If $\sum E_{\rm rad}$ is negligible compared to $E_{\rm plasma}$ or if $n_e \Sigma(h\nu b)$ is a weak function of n_e, $\eta_{\rm rad}$ increases linearly with n_e,

$$\eta_{\rm rad} \propto n_e, \quad (4)$$

because the photon flux per ion $p_{\rm rad}$ is proportional to the electron density. This applies to the case when n_e is low.

Because the absorption coefficient α is proportional to the square of the electron density, the layer becomes transparent to the laser after the expansion time. Then, the next layer in the target absorbs the laser light and expands shortly. When a target is thus ablated during laser heating, the total number of ions in the plasma $N_{i,\rm total}$ becomes very large. On the other hand, the emission power of a layer is lost after expansion because the radiation power per ion is proportional to the electron density, and the emitting volume remains nearly the same with that for a short pulse. Therefore, when $t_{\rm laser} > t_{\rm exp}$,

$$N_{i,\rm total} = (t_{\rm laser}/t_{\rm exp}) N_{i,\rm emit}, \quad (5)$$

and $\eta_{\rm rad} = h\nu b / \{\Sigma(h\nu b) + (\chi_{\rm sum} + ZT_e)/n_e t_{\rm exp}\}$. Because $1/t_{\rm exp} = V_{\rm exp}/L_{\rm abs} = V_{\rm exp}\alpha = V_{\rm exp} aZne^2$, $\eta_{\rm rad}$ is given by

$$\eta_{\rm rad} = h\nu b / \{\sum (h\nu b) + (\chi_{\rm sum} + ZT_e) V_{\rm exp} aZn_e\}. \quad (6)$$

Hence, $\eta_{\rm rad}$ is larger for lower n_e. If $\sum E_{\rm rad} \ll E_{\rm plasma}$,

$$\eta_{\rm rad} \propto 1/n_e. \quad (7)$$

This applies to the case when n_e is high.

In summarizing the discussion, when the expansion time $t_{\rm exp}$ of the emitting volume is longer than the laser pulse duration $t_{\rm laser}$, then the CE is higher for a higher density. On the other hand, when the density is high and $t_{\rm exp}$ is very short compared to $t_{\rm laser}$, which is the case for a conventional solid target, the CE is lower for a higher density. Thus the highest CE is achieved when

$$t_{\rm laser} = L_{\rm abs}/V_{\rm exp}, \quad (8)$$

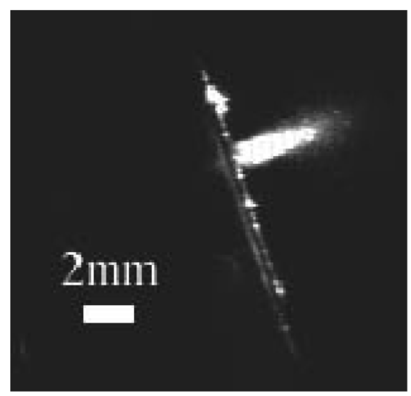
which is realized at the optimum electron density $n_{e,\rm opt}$ given by

$$1/n_{e,\rm opt} = (aZV_{\rm exp} t_{\rm laser})^{1/2}. \quad (9)$$

As clarified above, the ultimate CE is achieved for a plasma of thickness $V_{\rm exp} t_{\rm laser}$. The plasma expansion velocity $V_{\rm exp}$ is 5×10^6 cm/sec or larger in many case. If laser pulse duration is several ns, plasma thickness needs to be larger than a few hundreds μm. However, as discussed later, the density scale length of a plasma generated on a solid target is several tens μm even when a prepulse is irradiated. Therefore, in order to experimentally confirm the above derived dependence of CE on plasma density, we need a novel target by which we can distribute target material with controlled density into a large area of several hundred μm in diameter. For this purpose, we devised a technology of dispersing particles.

Tin oxide (SnO_2) powder dissolved in water was coated and dried on a Si wafer. The particle size of the powder is nominally 30 nm. However, we found a very wide distribution of size from 10 nm to a few μm under the electron microscope observation. Thus prepared particles were dispersed in a vacuum by a shock induced by irradiating a pulse laser, as schematically shown in Fig. 1(b). Previously, we successfully applied this laser-induced shock method to remove tens of nm Sn particles attached on a Si wafer without damaging the wafer. Spatial profile of dispersed particles was observed by imaging a scattered light of an illuminating 2nd harmonic Nd:YAG laser beam. The image taken at 150 μs after laser-induced shock is shown in Fig. 1(a). Observed images showed that the extension of dispersed particles was 0.2 mm at 15 μs, 1 mm at 30 μs, and 2 mm at 60 μs. From the images, we confirmed temporal progress of uniformly distributed particles with the expansion velocity of about 40 m/sec. The average density of target material n_i decreases with time t as given by $n_i = n_{i0}/(L_0 + V_{\rm disp} t)$, where n_{i0}, L_0, and $V_{\rm disp}$ are the initial density and thickness of particle powder coated on a Si wafer and dispersion velocity of particles, respectively. Therefore, by shooting the dis-

(a)



(b)

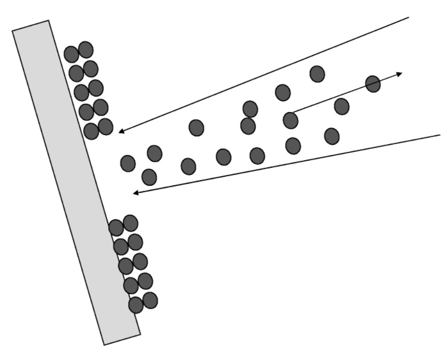


FIG. 1. (a) Expanding SnO_2 particles at 150 μs observed by laser scattering. (b) SnO_2 particles were dispersed by giving a laser-induced shock.

persed particles at various delay time after a laser-induced shock, we can generate a plasma of various density. A Nd:YAG laser of 1.06 μm wavelength and 8 ns duration was focused to a diameter of 300 μm onto the center of the column of scattered particles with 500 μm in diameter. Emission from the plasma was spectrally dispersed by a flat-field grating and a spectrum was recorded on a back-side illuminated CCD camera. The experiment was performed in a vacuum better than 1×10^{-3} Pa to avoid EUV absorption.

Figure 2 shows the observed EUV efficiency at 14 nm. For dispersed SnO_2 particles, the efficiency was nearly twice of that for a Sn plate when the laser pulse energy was larger than 250 mJ. The delay time of laser heating was 15 μs after the laser-induced shock for particle dispersion. By fixing the pulse energy to 300 mJ, i.e., at the irradiation power density of 5×10^{10} W/cm^2, the delay time from the laser-induced shock was scanned. As shown in Fig. 3, the EUV intensity increased with delay time up to 50 μs and reached nearly 4 times the intensity of that for a Sn plate target. When the delay time was increased further, the EUV intensity decreased slowly. The observed behavior of CE can be fitted with the curves for Eqs. (4) and (7). Thus, our theoretical derivation of the CE dependence on plasma density was experimentally confirmed.

In the configuration of the present experiment, it was very difficult to measure the absolute efficiency. In a different configuration, however, we measured the CE for a Sn plate [14], and the value was 0.82%. In this case, the focused diameter of a 1.06 μm laser of 8 ns duration with the pulse energy of 80 mJ was around 100 μm, smaller than the present experiment. There are many reports by other groups. The most extensive study was performed by Spitzer *et al.* [8]. Their value for the above condition was the same as ours. From their claim of a fair amount of robustness with respect to operating conditions for the temporal and physical dimensions of the emitted radiation, a tin plate can be a good standard target for the CE at 13.5 nm. According to them, the CE is larger for a larger diameter plasma and it was 1.3% when a 7.5 ns 1.06 μm laser irradiated the target at 5×10^{10} W/cm^2 with a spot size of 220 μm, similar to the present experiment. From these, the observed 4 times higher CE at 50 μs delay time than that for a solid target shown in Fig. 3 implies that 3% to 5% CE can be attained by a SnO_2 particle-cluster target.

When CE is high, the radiation energy $t_{\mathrm{laser}} n_e \Sigma(h\nu b)$ in Eqs. (3) and (6) can become non-negligible. Then, dependence of CE on plasma density is weaker. However, in the discussion above, we neglected self-absorption of radiation for simplicity. At both low and high density regions, the total number of ions in the plasma is very large, and then self-absorption becomes significant. The self-absorption effect reduces η_{rad} at both low and high density regions and dependence on plasma density is enhanced. Good agreement of experimental data with theoretical curves neglecting the radiation energy in Eqs. (3) and (6) might have been brought about by the combined effects of both $\Sigma(h\nu b)$ and self-absorption.

In the following we discuss two previous reports by employing the present theory.

The prepulse effect on CE was reported firstly by Kodama *et al.* [10], in which a Cu target was irradiated by a 0.2 ns duration 0.53 μm laser at 1×10^{14} W/cm^2 with a focus spot of 250×300 μm at delay times of 1.1, 1.8, and 3.1 ns after the prepulse irradiation by a 0.2 ns duration 1.06 μm laser. They observed the efficiency of x-ray generation in the range of 1.5 to 5 keV changed with a delay time. This experiment was motivated by an earlier report [15] of the increase of radiation efficiency with a

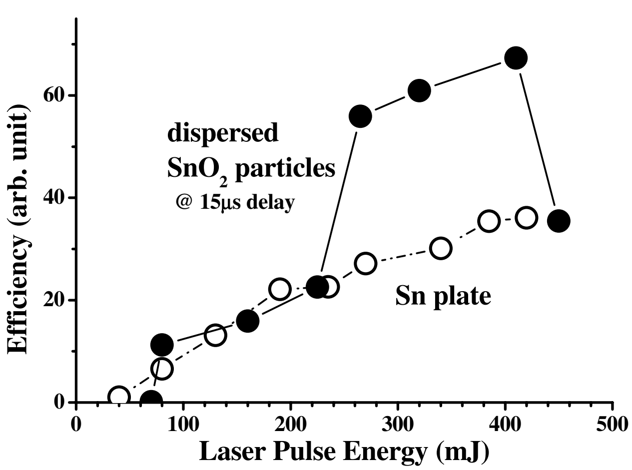


FIG. 2. Observed EUV efficiency at 14 nm for a Sn plate and for dispersed SnO_2 particles. The delay time of generating the plasma after the laser-induced shock was 15 μs.

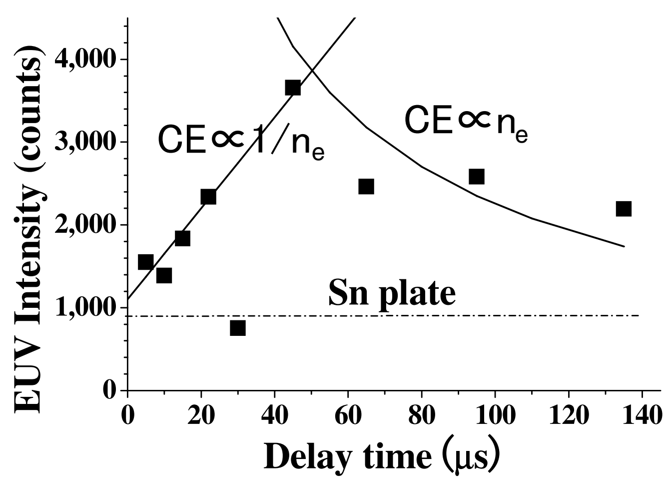


FIG. 3. The observed dependence of EUV intensity from a dispersed particle target on delay time of plasma generation after the shock for dispersion. EUV intensity at 50 μs delay was nearly 4 times higher of that for a Sn plate. The inserted curves show Eqs. (4) and (7).

plasma volume, and they plotted the CE as a function of a plasma scale length evaluated from a hydrodynamic simulation. The efficiency was found to increase linearly with a scale length from 5 μm and reached the peak value of 1%/sr at 30 to 40 μm and decreased quite slightly up to 60 μm. We can interpret their results with the present theory. Because the highest CE was observed for a scale length of around 40 μm, expansion velocity V_{exp} is estimated to be 2×10^7 cm/sec from Eq. (8). Then, V_{exp} gives T_e of 600 eV. From $L_{\mathrm{abs}} = 40$ μm for a 0.53 μm laser, we get n_e of $1 \times 10^{21}/\mathrm{cm}^3$, whose value is reasonable for a plasma preformed by a 1 μm laser. When the scale length is very short, a laser is absorbed near the critical density $4 \times 10^{21}/\mathrm{cm}^3$ of a 0.53 μm laser. Then, the laser absorption length is very short, near 1/16 of that for $1 \times 10^{21}/\mathrm{cm}^3$, the expansion time t_{exp} is very short, and the total number of ions is very large as described by Eq. (5). Therefore, the CE is lower for a smaller scale length as was observed. Slightly lower efficiency at a 60 μm scale length is interpreted as the start of efficiency decrease for a lower density expressed by Eq. (7).

Dusterer *et al.* [16] reported effects of a prepulse on EUV radiation efficiency. A water droplet of 20 μm diameter was irradiated by a 20 mJ, 1.8 ps, 800 nm wavelength laser after a prepulse of 2 mJ laser, and the EUV efficiency increased linearly with a delay time up to 3 ns and decayed slowly up to 12 ns. Very interestingly, the observed dependence on delay time shown in Fig. 3 in Ref. [16] is quite similar to the present observation shown in Fig. 3. They assumed T_e, V_{exp}, and Z were 30 eV, 1×10^7 cm/sec, and 5, respectively. Then, $L_{\mathrm{abs}} = V_{\mathrm{exp}} t_{\mathrm{laser}}$ was 0.2 μm. And then, according to Eq. (9), n_e at a delay time of 3 ns is estimated to be near critical density of the laser. Thus we understand that a prepulse was required to created the plateau profile at the critical density for efficient absorption of the laser.

In conclusion, an analytical formula for maximizing radiation efficiency from a laser-produced plasma is derived. The maximum CE is achieved when the plasma thickness, plasma expansion distance during laser heating, and laser absorption length are all the same. However, the density scale length of a plasma generated on a solid target remains several tens μm even when a prepulse is employed, and the ultimate efficiency will not be attained for a solid target when a pulse width is several ns. The derived theory guided us to devise a novel target scheme, a particle cluster by which we can distribute a solid element uniformly into a large area. The derived dependence of the efficiency on plasma density was confirmed in an experiment in which a uniform plasma with a 300 μm diameter was created by dispersing SnO_2 particles coated on a Si wafer. We observed the conversion efficiency as high as 4 times that for a bulk Sn target. A supply of target material at multi-kHz repitition rate, mandatory specification for an EUVL source, can be realized by delivering particle clusters in multi-kHz generated droplets [14]. Because previously demonstrated conversion efficiency by using a Xe plasma was 0.5% to 1% for kHz repitition rate [17] which is lower than that from a bulk Sn target, the present result makes bright the prospects for EUV lithography.

This work was financially supported by NEDO (New Energy and Industrial Technology Development Organization).

*Corresponding author
Electronic address: t-tomie@aist.go.jp

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