

# Combined effects of pre-pulsing and target geometry on efficient EUV production from laser produced plasma experiments and modeling

A. Hassanein, T. Sizyuk, V. Sizyuk, and S. S. Harilal

Center for Materials under Extreme Environment, School of Nuclear Engineering  
Purdue University, West Lafayette, IN, USA

## ABSTRACT

Laser produced plasmas (LPP) is currently a promising source of an efficient extreme ultraviolet (EUV) photon source production for advanced lithography. Optimum laser pulse parameters with adjusted wavelength, energy, and duration for simple planar or spherical tin target can provide 2-3% conversion efficiency (CE) in laboratory experiments. These values are also in good agreement with modeling results. Additional effects such as targets with complex geometry and tin-doped targets using pre-pulsing of laser beams can significantly increase CE. Recent studies showed that such improvements in LPP system are due to reduction in laser energy losses by decreasing photons transmission (higher harmonic of Nd:YAG laser) or photons reflection (for CO<sub>2</sub> laser). Optimization of target heating using pre-pulses or ablating low-density and nanoporous tin oxide can further improve LLP sources by creating more efficient plasma plumes and as a result increasing CE, the most important parameter for EUV sources. The second important challenge in developing LPP devices is to decrease fast ions and target debris to protect the optical collection system and increase its lifetime.

We investigated the combined effects of pre-pulsing with various parameters and different target geometries on EUV conversion efficiency and on energetic ions production. The much higher reflectivity of CO<sub>2</sub> laser from a tin target leads to two possible ways for system improvement using pre-pulses with shorter laser wavelengths or using more complex targets geometries with special grooves as developed previously by the authors.

**Keywords:** Nanolithography, EUV, LPP, HEIGHTS, Debris mitigation, CO<sub>2</sub> lasers.

## 1. INTRODUCTION

There are still several challenges in the development and production of the currently progressing laser produced plasma (LPP) sources for extreme ultraviolet lithography (EUVL), and in particular using CO<sub>2</sub> lasers and tin targets. Plasma sources using CO<sub>2</sub> lasers have a number of advantages over other LPP sources using Nd:YAG lasers including higher laser efficiency, lower power needed to produce similar plasma conditions, less generated debris, etc. In addition to the extensive work on source optimization and collection of EUV photons, the lifetime of the collecting mirror system is also very important and is under comprehensive development and investigation. Damage of multilayer Mo/Si mirrors by the debris products of laser beam interaction with target materials can significantly reduce their lifetime and as result the efficiency and the economy of the entire EUVL system. For example, while debris production is more pronounced for Nd:YAG lasers, CO<sub>2</sub> laser beams produce more energetic ions at the optimum conditions for laser energy intensities needed for maximum EUV production [1], [2]. Additionally the impact of plasma fast ions and the deposition of atomic debris and the deposition of EUV and out of band radiation can further cause surface erosion and damage at the required higher fluences of the 13.5 nm radiation needed for high volume manufacturing [3].

The lower mass ablation rate by the CO<sub>2</sub> laser beams with the longer wavelength and comparatively low value of the critical density for absorption of these photons allow source operation using longer pulses (100 ns and more) without significant reduction in the efficiency of EUV output [4]. Increased pulse duration, however, leads to increasing debris producing and accumulation and therefore, decreasing mirror lifetime. Another disadvantage of longer pulses is the increase in EUV source size. While usually LPP devices provide small source sizes that satisfy the optical system requirements for the etendue, CO<sub>2</sub> laser systems with larger spot sizes (500  $\mu$ m and more) and long pulse duration can cause difficulties in EUV photons collection.

The above aspects and requirements will determine the preferences in target geometry and size. Currently tin droplets with sizes from 10 to 100  $\mu\text{m}$  are under investigation. We extensively modeled tin droplets ablation by  $\text{CO}_2$  laser and studied the dependence of laser energy losses on droplets and spots size, laser beam intensities, and time duration using our HEIGHTS full 3D simulation package. We predicted the optimum parameters in certain combinations of double-pulsed system to increase EUV production through the processes of the initial plasma creation by preheating of the tin droplets with 532 nm and 1.064  $\mu\text{m}$  wavelengths and then followed by the main  $\text{CO}_2$  laser pulse.

We utilized unique combination of our experimental facilities (CMUXE Laboratory) and our state-of-the art computer simulation (HEIGHTS package) for detail investigation, benchmarking, and optimization of LPP sources with various lasers, target parameters, geometries, and optical collection system. Comparisons of various experiments with modeling results were discussed and detailed plasma characteristics for more efficient LPP sources were analyzed.

## 2. MODEL DESCRIPTION

HEIGHTS simulation package is being developed as a comprehensive tool for the investigation and optimization of various radiation sources interacting with matter and that include both laser and discharge produced plasmas proposed for the next generation of nanolithography, i.e., the extreme ultraviolet lithography applications. We developed integrated multi-physics, multi-phase models to simulate the interaction of intense energy sources with target materials. We used HEIGHTS to model in detail LPP devices that consider and integrate all interaction phases and various processes, i.e., from the initial stage of solid/liquid target ablation by the laser photons up to final generation of the EUV radiation [5]-[8]. The developed models address and integrate several major research areas, i.e., physics of laser absorption in target materials, vapor/plasma evolution and magneto hydrodynamic (MHD) processes, thermal conduction in condensed material and plasma, atomic physics and resulting opacities, detailed photon radiation transport, and interaction between plasma/radiation and target material in full 3D geometry. HEIGHTS package utilizes various models and numerical solution methods to calculate details of energy deposition, MHD evolution, radiation transport, and heat conduction in solid/liquid/vapor/plasma target phases. Radiation transport methods using both direct methods as well as weighted Monte Carlo models are developed to specifically calculate in fine details both continuum and lines transport with fine spectral resolutions and profiles. In addition, another weighted Monte Carlo model is used to calculate laser energy absorption, reflection, and transmission of photons in various phases of the dynamically evolving target material.

Accurate photon radiation transport calculations are very critical in the assessment and evaluation of EUVL devices and their efficiency. HEIGHTS uses two different approaches to calculate radiation transport: Direct integration of the radiation transport equation and Monte Carlo techniques with weight factors hierarchy. Each method has its own advantages and disadvantages. These different approaches provide significant insight and self-benchmarking on the appropriate numerical technique for multidimensional solution of such complicated physics problems. The radiant energy flux is calculated by integrating the differential radiation transport equation (RTE), which represents the conservation of emitted and absorbed photons energy along the direction of photons movement. Direct Gauss integration methods digitize the MHD domain in space, energy, and angles. The major limitations of this approach are time-consuming and computer resources as well as numerical stability. Alternative to the direct resolution of the RTE, where we seek the continuous function of the radiation intensity, the probabilistic model of the energy redistribution operates with discrete portions of the radiative energies, as shown in Fig. 1(a). Each energy portion is individually analyzed. The trajectory and number of emitted and absorbed energy portions are evaluated in each point of the plasma domain (or in the points which represent major interest with fine details). The energy is then re-distributed as a result of photons motion and interaction in each cell domain. The energy and the trajectory of photon movement can be determined by Monte-Carlo techniques as well as by direct solution of photons transport equation as a way to check the accuracy of our calculations.

Laser absorption, reflection and resulting interaction/transport, and the reabsorption or transmission of laser photons energy are modeled similar to the plasma photon radiation transport with weighted Monte Carlo techniques. Figure 1(b) illustrates three possible events for laser photons: 1) absorption in solid matter or plasma; 2) reflection and reabsorption in plasma; 3) reflection and transmission out of the area of potential absorption. Our Monte Carlo method allows redistribution of laser photons energy in all directions that can be important in the domain of the plasma plume with nonuniform conditions in 2D or 3D configurations.

The absorption/reflection of laser photons in solid/liquid phases of the target in our models is based on experimental data and models of absorption/reflection in plasma utilize inverse bremsstrahlung mechanism since the electrons are the main interaction particles of the laser photon radiation energy [7].

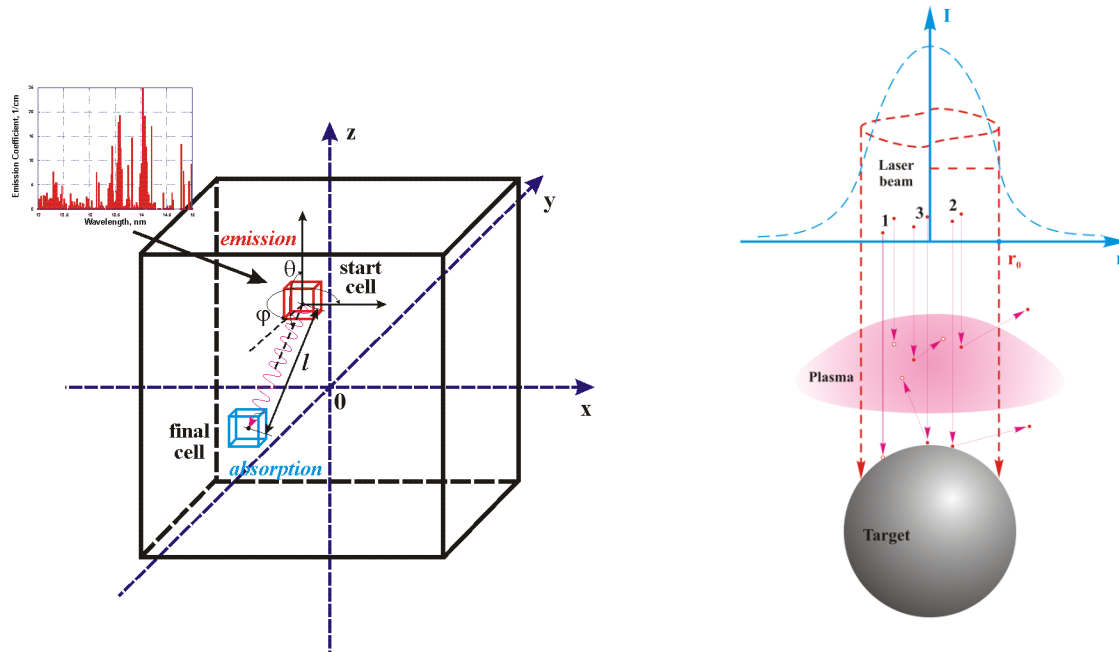


Fig. 1. a) Monte Carlo illustration of emission-absorption process for the radiation transport; b) interaction of the laser energy photons with target

For correct modeling and higher accuracy of the radiation transport processes, we considered two important features in our calculations. Radiation fluxes should first be determined with the correction of the plasma thermal energy and, as a result, the correction of plasma evolution and motion in the device domain and second, following the use of detail analysis of the final useful part of the very narrow EUV plasma radiation flux. These problems involve different requirements for determining the radiation flux and therefore require different numerical techniques for solving the photon transport problem. Correct calculation of the spatial energy redistribution in the full spectrum plays an important role in the accurate solution of the first problem. A model that adequately describes radiation transport is only correct if it takes into account the optical thickness of the plasma over a wider spectral range exists in plasma domain. The total spectral range must be optimized to sufficiently and accurately describe the radiation energy redistribution using reasonable computational capability. These opacities, however, will be inadequate for detailed investigations of the EUV photons in the required spectral band of  $13.5 \pm 2\%$  nm. Therefore, two sets of optical opacities are developed, i.e., a general one (for full energy redistribution calculations) and a specific detailed one (for a specific spectral band).

The equations-of-state (EOS) and opacities are calculated independently for a wide range of temperatures and densities. Tabulated opacities are used during the numerical simulation by interpolating of plasma density and temperature parameters.

The structure of atomic energy levels, wavefunctions, transition probabilities, ionization potentials, oscillator strengths, broadening constants, photoionization crosssections and other atomic characteristics are calculated using the self-consistent Hartree-Fock-Slater (HFS) method [9]. The collisional-radiative equilibrium (CRE) model [10] is used to calculate the populations of atomic levels and the ion and electron plasma concentrations. The ion and electron concentrations calculated from the CRE model are used in the EOS to calculate plasma pressure and internal energy.

First, we calculated the emission and absorption coefficient in the full energy spectrum with resolution of up to 100,000 spectral points. Then we analyzed the full spectrum, determined strong spectral lines, and integrated into spectral groups for accurate modeling and transport of these critical EUV photons.

### 3. DEPENDENCE OF LASER ENERGY ABSORPTION EFFICIENCY ON PLASMA CONDITIONS

We studied the effect of initial pre-pulses of laser energy and the dependence of laser wavelengths on the conversion efficiency for various target and laser beam conditions. First we evaluated the percentage of useful laser energy which is absorbed in matter for the optimum intensity of CO<sub>2</sub> laser beam with 100  $\mu\text{m}$  spot size (FWHM) and 30 ns pulse width on 30  $\mu\text{m}$  tin droplet. These values correspond to a mass-limited source that can reduce chamber contamination and having reasonable laser beam parameters for an efficient EUV source. Figure 2 shows HEIGHTS comparison of the input laser power and the lost power, i.e., escaped and reflected energy as a function of time. Since the spot size of the laser exceeded the droplet diameter in more than three times, major part of laser photons didn't interact with the target until a plasma plume was formed with sufficient density needed for the absorption of laser photons at this wavelength. The conditions at which the evolving target plasma started to interact with the laser beam are at about 20-25 ns from the start of irradiation. The corresponding mass density distribution of the initial plasma is shown in Fig. 3 at the time of 20 ns.

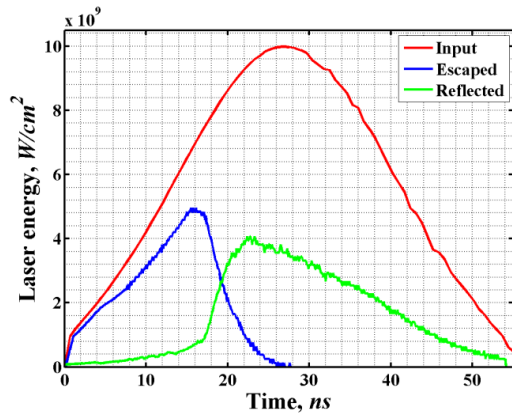


Fig. 2. Laser energy distribution for 100  $\mu\text{m}$  spot size (FWHM), 30 ns duration (FWHM) and  $1 \times 10^{10}$  W/cm<sup>2</sup> intensity; droplet diameter 30  $\mu\text{m}$

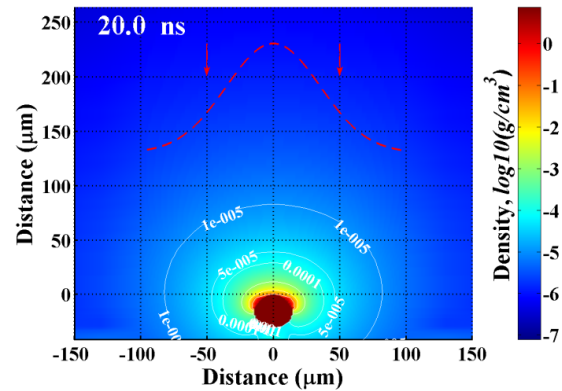


Fig. 3. Mass density distribution of plasma plume from tin droplet of 30  $\mu\text{m}$  diameter

Preheating the droplet by a laser with shorter wavelength and the resulting initial plasma plume formation can reduce energy losses of the main pulse. A second harmonic of Nd:YAG laser was used in our numerical simulations for the heating, evaporation, and initial ionization of the droplet target. Figure 4 shows comparison of laser energy absorption for Nd:YAG (second harmonic of 532 nm) at intensity of  $5 \times 10^{11}$  W/cm<sup>2</sup> during 10 ns and CO<sub>2</sub> at intensity of  $10^{10}$  W/cm<sup>2</sup> for two cases with and without pre-pulse interacting with 30  $\mu\text{m}$  initial droplet size. Figure 5 shows the mass density distribution as a result of the pre-pulse laser that expanded during 80 ns. In both cases CO<sub>2</sub> laser heated the plasma up to a maximum temperature of 50 eV; this temperature indicates that plasma plume with optimum conditions for EUV production was created [8]. The improved absorption of the laser energy increased CE value up to 50%.

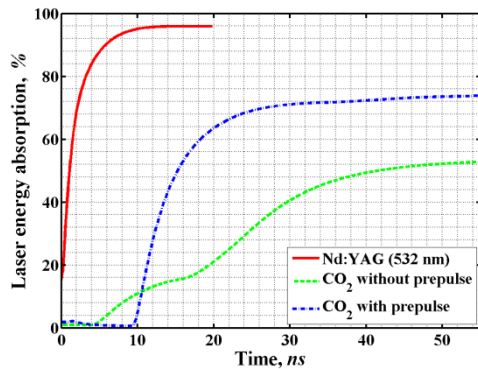


Fig. 4. Laser energy absorption at optimized intensities of: 1) 532 nm laser; 2) CO<sub>2</sub> without pre-pulse; 3) CO<sub>2</sub> after pre-pulse

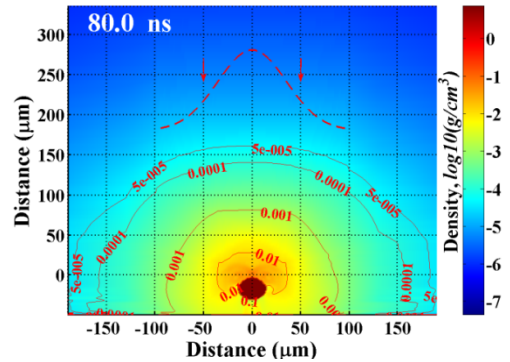


Fig. 5. Mass density of plasma plume from the droplet of 30  $\mu\text{m}$  created by 532 nm laser and expanded during 80 ns

In this numerical simulation the initial plasma was created by 532 nm laser with the intensity of  $10^{10}$  W/cm<sup>2</sup>, which did not create plasma with conditions suitable for EUV emission during pre-pulse.

#### 4. DEPENDENCE OF EUV EMISSION ON TARGET MASS AND SPOT SIZE

Increasing the delay time between the pre-pulse and the main pulse did not influence much on the EUV production by the main CO<sub>2</sub> laser. After the time moment when the mass density of about  $10^{-4}$  g/cm<sup>3</sup> reaches the borders of the spot radius (around 30 ns for this droplet size and the pre-pulse energy), additional plasma expansion does not significantly improve EUV production. The change in the CE for 30 ns and 80 ns delay times was only about 10%. A more important parameter in this case is the spot size of the main laser beam, which can be adjusted to the delay duration and as a result to the evolving plasma size. Double increase in spot size of CO<sub>2</sub> laser led to an increase in CE of more than two times with a conversion efficiency that is comparable to that received for planar targets, i.e., of about 1.8%.

There are several ways for optimizing LPP sources with mass-limited targets: 1) preparing an initial plasma by a pre-pulse; 2) adjustment of target size and spot size; and 3) increasing the time of laser pulse duration. We studied the above options in our optimization analysis. Since the time dependence of EUV output always follows the time profile of the laser beam, it is also reasonable to use pre-pulse laser intensity suitable for in-band emission at this stage. To achieve the maximum EUV output we used optimized parameters for pre-pulse beam, i.e.,  $10^{11}$  W/cm<sup>2</sup> of 1064 nm with 100  $\mu$ m spot on the same diameter of a tin droplet. We obtained a conversion efficiency of about 1.7% for this pulse. Next we allowed the plasma plume to expand during 40 ns and then applied a main CO<sub>2</sub> laser pulse with 30 ns duration and an intensity of  $10^{10}$  W/cm<sup>2</sup> with various spot sizes. Figure 6 shows the influence of the spot size on EUV production at the above stated conditions.

For mass-limited target systems two parameters can influence the efficiency of an EUV source: comparatively larger spot size and sufficiently prepared mass density to absorb the incident laser energy with such spot size. Mainly lasers with short wavelengths (harmonics of Nd:YAG) can be used for the first stage since they have better absorption in solid and liquid matter. The CO<sub>2</sub> laser can then be the best choice for the second stage, i.e., EUV production stage, since the evolving plasma plumes are more suitable for the absorption of such longer laser wavelength.

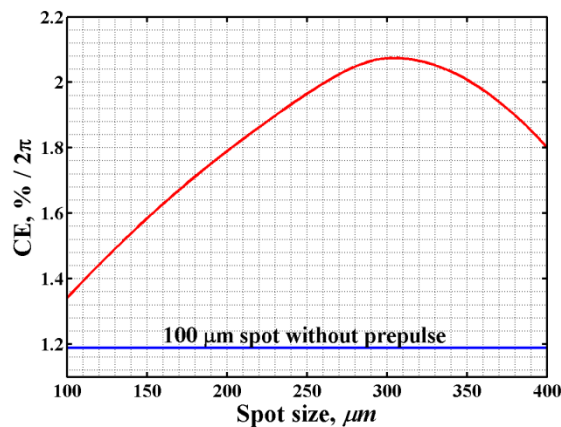


Fig. 6. The dependence of CE on CO<sub>2</sub> spot size applied after pre-pulse on tin target. For comparison CE of CO<sub>2</sub> laser with 100  $\mu$ m spot size and 100  $\mu$ m droplet without pre-pulse is given.

#### 5. HYDRODYNAMIC EFFECTS ON EUV PRODUCTION BY CO<sub>2</sub> LASER IN PLANAR AND SPHERICAL TARGETS

We analyzed the EUV source size and intensity from a tin plasma produced from both plate and droplet targets with the same parameters of a CO<sub>2</sub> laser. We used a 100  $\mu$ m spot size (FWHM) and long pulse duration of 90 ns. The difference in the hydrodynamics of plasma motion in these two systems determines the differences in the EUV source size and

shape. Figures 7 and 8 show the areas of emitting EUV photons that are collected in  $2\pi$  sr during 150 ns. The expanded source in the planar case is explained by the changes in the source location with time, and this is related to the movement of plasma plume with the optimal combination of temperature and density values. This is specifically related to the  $\text{CO}_2$  laser system, where EUV radiation is produced in low-density region that moves and expands more easily. Plasma motion around droplet in the spherical target case prevents plasma accumulation above target. Planar target provided more efficient source with increasing CE of more than 30% for these conditions. This is a consequence of the hydrodynamic evolution where the plasma geometrical confinement can play an important role [7].

Because the evolving plasma plume above the droplet surface, created by the pre-pulse laser beam, can change the EUV production area during the main pulse similar to the planar target, the spatial distribution of the EUV source will have the same tendency in the size expansion. Therefore, in the optimization processes, one need to further adjust the spot and droplet sizes as well as pre-pulse/delay/pulse times. The resulting source size should be evaluated to satisfy the optical extendue system requirements [11].

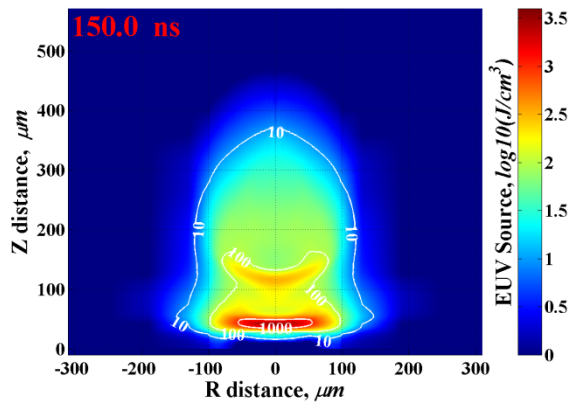


Fig. 7. Location and intensity of the EUV power collected during 150 ns in  $2\pi$  sr in planar target

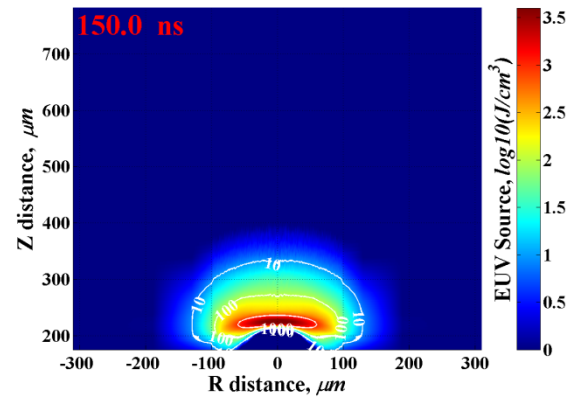


Fig. 8. Location and intensity of the EUV power collected during 150 ns in  $2\pi$  sr in spherical target

## 6. COMPARISON OF HEIGHTS RESULTS WITH EXPERIMENTS

We tested various models and components of HEIGHTS package to simulate EUV production at various experimental conditions directly without any adjustments in the package and benchmarking with different experimental results and conditions [12]. We considered several parameters that can influence the in-band photons production, i.e., laser beam wavelength, pulse duration, and spot size.

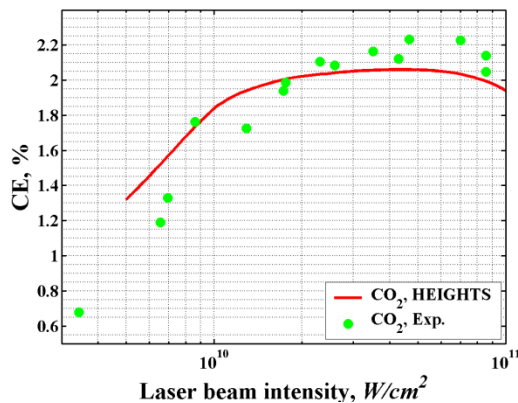


Fig. 9. Dependence of CE on laser beam intensity in experiments [12] and HEIGHTS modeling

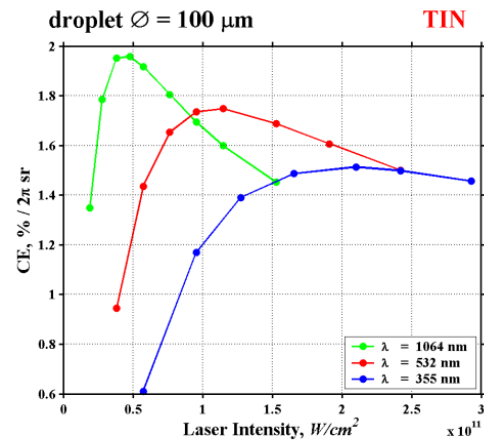


Fig. 10. Dependence of CE on laser beam wavelength for tin droplet target using HEIGHTS package [6]

Figure 9 compares the results of HEIGHTS modeling with the experimental results, where tin droplets with 100  $\mu\text{m}$  diameters were irradiated by a  $\text{CO}_2$  laser beam with 100  $\mu\text{m}$  spot size and with different intensities. HEIGHTS showed very good agreement with the data over a wide range of laser intensities. Since Nd:YAG laser is more suitable for the pre-pulse irradiation, and the pre-pulse stage can also be used for EUV production, we compared the efficiency of this laser with different wavelengths. The dependence of CE on laser beam wavelength from HEIGHTS simulation is shown in Fig. 10 for 100  $\mu\text{m}$  droplet. HEIGHTS showed that the CE exhibits similar peaking behavior with laser intensity for each of the laser wavelength studies with maximum CE predicted at the shorter wavelength and relatively lower intensity for the conditions studied in this case. Experimental data showed HEIGHTS similar behavior on wavelength intensities of Nd:YAG lasers [13].

## CONCLUSION

Further development of laser produced plasma (LPP) sources for EUV lithography should be based on mass-limited targets such as tin droplets with 10-100  $\mu\text{m}$  diameters using  $\text{CO}_2$  lasers. However, the  $\text{CO}_2$  lasers have high reflectivity from the solid and liquid tin that reduces efficiency of such small targets and result in low conversion efficiency (CE). To optimize and enhance the CE of LPP sources, several factors need to be taken into account, i.e., evaluation of Nd:YAG laser parameters needed for the pre-pulse to create sufficient mass density of plasma plume suitable for the main  $\text{CO}_2$  pulse, adjustment of the delay time between the two pulses and the laser spot size of main pulse, prediction of EUV source size at different combinations of laser beam characteristics and target evolution dynamics. We utilized our HEIGHTS package to simulate several of the above combinations, to determine the critical values that influence the CE of EUV protons and their collection and identified optimum spot size for the evolving plasma plume created at specific pre-pulse conditions. Computer simulation analysis can provide great guidance to predict the optimum conditions of LPP devices to enhance their performance and extend their lifetime.

## ACKNOWLEDGMENT

This work is partially supported by the College of Engineering, Purdue University. We gratefully acknowledge the computing resources provided by the Fusion cluster operated by the Laboratory Computing Resource Center at Argonne National Laboratory.

## REFERENCES

- [1] Takahashi, A., Nakamura, D., Tamaru, K., Akiyama, T., and Okada, T., "Comparative study on EUV and debris emission from  $\text{CO}_2$  and Nd: YAG laser-produced tin plasmas", *Journal of Physics: Conference Series* **112**, 042059 (2008).
- [2] Campos, D., Harilal, S.S., and Hassanein, A., "The effect of laser wavelength on emission and particle dynamics of Sn plasma", *Journal of Applied Physics* **108**, 113305 (2010).
- [3] Barkusky, F., Bayer, A., Döring, S., Grossmann, P., Mann, K., "Damage threshold measurements on EUV optics using focused radiation from a table-top laser produced plasma source", *Optics Express* Vol. 18, No. 5, 4346 (2010).
- [4] Tao, Y., Tillack, M. S., Sequoia, K. L., Burdt, R. A., Yuspeh, S., and Najmabadi, F., "Efficient 13.5 nm extreme ultraviolet emission from Sn plasma irradiated by a long  $\text{CO}_2$  laser pulse", *Applied Physics Letters* **92**, 251501 (2008).
- [5] Sizyuk, V., Hassanein, A., Morozov, V., Tolkach, V., Sizyuk, T., "Numerical simulation of laser-produced plasma devices for EUV lithography using the heights integrated model", *Numerical Heat Transfer Part A* **49**, 215-236 (2006).
- [6] Sizyuk, V., Hassanein, A., and Sizyuk, T., "Three-dimensional simulation of laser-produced plasma for extreme ultraviolet lithography applications", *J. Appl. Phys.* **100**, 103106 (2006).
- [7] Hassanein, A., Sizyuk, V., Sizyuk, T., and Harilal, S., "Effects of plasma spatial profile on conversion efficiency of laser-produced plasma sources for EUV lithography", *J. Micro/Nanolith MEMS MOEMS* **8**, 041503 (2009).



- [8] Hassanein, A., Sizyuk, V., Harilal, S.S., and Sizyuk, T., "Analysis, simulation, and experimental studies of YAG and CO<sub>2</sub> laser-produced plasma for EUV lithography sources", Proc. of SPIE 7636, 76360A (2010).
- [9] Herman, F., and Skillman, S., "Atomic Structure Calculations", Prentice Hall, Englewood Cliffs, (1963).
- [10] Zaltzmann, D., [Atomic Physics in Hot Plasmas], Chap.4. Oxford University Press., New York (1998).
- [11] Bakshi, V., "EUV Source Technology: Challenges and Status", [EUV Sources for Lithography], ed. V. Bakshi, SPIE Press, 7 (2005).
- [12] Soumagne, G., et al., "Combined Nd:YAG -CO<sub>2</sub> Laser Produced Plasma EUV Source Examination with the Code Z", EUV Source Workshop, Baltimore, USA (2007).
- [13] Nishimura, H., Fujioka, S., Shimomura, M., Sakaguchi, H., Nakai, Y., Aota, T., Kai, T., Nishihara, K., Miyanaga, N., Izawa, Y., Mima, K., Sunahara, A., Shimada Y., and Namba, S., "Development of Extreme-Ultraviolet Light Source by Laser-Produced Plasma", The Review of Laser Engineering, Vol. **36**, 1125 (2008).