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ABSTRACT

The extension of the roadmap of shorter wavelength extreme ultraviolet and soft x-ray sources is a topic of considerable interest. We have studied the optimized emission from high power and/or high brightness sources based on unresolved transition array (UTA) emission. The peak UTA wavelengths follow a quasi-Moseley’s law as \( \lambda = 33.82 \times R^{-1/\infty} (Z - 20.86)^{-1.61} \) nm for the laser wavelength of 1064 nm (the critical density of \( 1 \times 10^{21} \) cm\(^{-3} \)) and \( \lambda = 165.8 \times R^{-1/\infty} (Z - 12.44)^{-1.94} \) nm for the laser wavelength of 532 nm (the critical density of \( 4 \times 10^{21} \) cm\(^{-3} \)), respectively. The photon flux decreased with increasing atomic number. We also mapped the optimum electron temperatures and corresponding charge states required to produce strong soft x-ray UTA emission with a photon flux of the order of \( 1 \times 10^{14} \) photons nm\(^{-1} \) sr\(^{-1} \). The present quasi-Moseley’s law is sufficient for identifying the optimum element for numerous applications, such as material ablation and ionization, nanolithography, and in vivo biological imaging.

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I. INTRODUCTION

Interest in the use of high power and/or high brightness extreme ultraviolet (EUV) and soft x-ray (SXR) sources has been motivated in various fundamental and/or engineering fields, such as the absorption spectroscopy,1,2 for material ablation and ionization,3,4 nano-lithography,5,6 and in vivo biological imaging.7,8 In one of the most important applications in this spectral region, namely, EUV lithography, an output power of a few 100 W at 13.5 nm has been achieved.9–11 On the other hand, we need to extend the
EUV/SXR roadmap to answer the “what’s next.” In the past, shorter wavelength EUV/SXR sources have been explored for their potential application in shorter wavelength lithography. However, the study of the atomic number scaling of the output flux under optimum illumination conditions remains an important issue to be explored. To produce high output power at 13.5 nm, the unresolved transition array (UTA) spectral emission from highly charged tin ($^{90}$Sn) ions resulting from thousands of resonant lines of Sn$^{74+}$–Sn$^{115+}$ due to the overlapping of 4$d$–4$f$ and 4$p$–4$d$ ($\Delta n = 0$) transitions is utilized, resulting in very high output intensity. The output power of the emission may be attributed essentially to the total effective emissivity of the UTA spectra. However, the potential photon flux should be also evaluated for these UTAs in other elements. It is important to clarify the atomic number scaling of not only the peak wavelengths, but also the output power (number of photons) of the UTA emission for use as high power EUV/SXR sources.

Understanding the atomic number dependence of the UTA wavelengths resulting from $n = 4–n = 4$ ($\Delta n = 0$), transitions has been the subject of a considerable amount of theoretical and experimental studies. According to atomic structure calculations, the UTA peak wavelength becomes shorter with increasing atomic number and the 4$d$–4$f$ and 4$p$–4$d$ ($\Delta n = 0$) transitions separate at $\gamma$Lu, due to increased spin orbit splitting of the 4$p$–4$d$ component arrays, as seen in Fig. 4 in Ref. 21. In other words, the two transition arrays are overlapped from $^{90}$Sn to $^{79}$Yb. As a result, the opacities of laser-produced plasmas from $^{90}$Sn to $^{79}$Yb are extremely large due to the overlapping of many resonant lines with large $A_g$-values in adjacent ion stages, where $g$ is the statistical weight of the upper level and $A$ is the spontaneous decay coefficient. The measured output flux, related to the opacity, includes the effects of both the spectral emissivity and the absorption coefficient. In addition, the absorption coefficient generally depends on the spectral functions of the emission and absorption processes. As these spectral functions are matched in lower ion stages, i.e., at lower electron temperatures in plasmas of the lower $Z$ elements, strong absorption will be resonantly induced in such plasmas. This means that longer wavelength UTA emission from these plasmas with lower optimum electron temperatures is expected to be optically thick. On the other hand, shorter wavelength emission from plasmas of the higher $Z$ elements associated with higher optimum electron temperatures is expected to be optically thinner due to the spectral function mismatch between the emissivity and the opacity. However, the photon flux and absorption spectral structure that can be observed in highly charged ions (HCIs) in high-$Z$ plasmas have not been studied in detail and remain unclear. Therefore, we explore here the effects of different optical thicknesses in order to optimize the strong UTA emission systematically with changing atomic number. In addition, no systematic investigation of the UTA spectral behavior at sub-nano-second laser pulse irradiation has been reported in the literature.

In this paper, we focus initially on the spectral comparison of the effects of different laser wavelength (different critical density), together with the observation of the emission spectra and absorption structure. After that, we discuss the optimum charge states that occur for different elements at different optimum electron temperatures.

II. EXPERIMENTAL SETUP

To compare the spectral structure, we observed the spectra from plasmas characterized by different optical thicknesses. In general, it is difficult to observe directly the absorption coefficient of HCIs, which is related to their opacity. Note that the absorption coefficient depends on the density and path-length product. There are some methods to observe opacity effects indirectly by changing the laser pulse duration (path-length), dual laser pulse irradiation (path-length), and changing the laser wavelength (plasma density). Because the critical densities are $1 \times 10^{21}$ and $4 \times 10^{21}$ cm$^{-3}$ for laser wavelengths of 1064 and 532 nm, we can infer the effects of optical thickness by comparing emission spectra of plasmas produced by pulses at these wavelengths. In the present experiments, we irradiated planar targets of the elements of interest...
with 150-ps laser pulses at wavelengths of 1064 and 532 nm, produced by Nd:yttrium-aluminum-garnet (Nd:YAG) lasers with focal spot diameters of 40 μm. The pulse energies were the same, 130 mJ at 1064 and 532 nm, respectively, corresponding to a laser intensity of $7 \times 10^{13}$ W/cm$^2$. Metal planar targets were placed at the center of a vacuum chamber. The laser beam was focused onto the targets at normal incidence. The calibrated spectrometer was a flat-field grazing incidence type with an unequally ruled 2400 grooves/mm variable line space grating with a spectral resolution of 0.01 nm. The spectrometer was positioned at 30° with respect to the incident laser axis.

### III. EXPERIMENTAL RESULTS AND DISCUSSION

We compare the spectra from plasmas characterized by different electron densities and consequently opacities, in Fig. 1. The peak wavelengths of the UTA spectra, attributable to the 4$d$–4$f$ and 4$p$–4$d$ transitions from $^{64}$Gd to $^{83}$Bi, become shorter with increasing atomic number, $Z$, as shown in Fig. 2(a). From, $Z = 64$ to 70, the peak wavelength and the photon flux were comparable for two color laser irradiation. Although the spectral structures for $Z = 72$–83 were similar, the spectral intensities for the 532-nm laser irradiation were higher than those for the 1064-nm laser irradiation, corresponding to the emergence of optically thin conditions in plasmas of the elements from $Z = 72$–83. We plotted the atomic number dependence of the wavelength and the photon flux of the UTA spectral peaks, as shown in Figs. 2(a) and 2(b), respectively. The dashed lines are an approximated curve with a power-law scaling of the peak wavelength given by $\lambda = a R_{\infty}^{s} (Z - s)^{-b}$ in nanometer, where $R_{\infty}$ is the Rydberg constant. The fitting curves were given by $\lambda = 33.52 R_{\infty}^{s} (Z - 20.86)^{-1.61}$ for the laser wavelength of 1064 nm (the critical density: $1 \times 10^{21}$ cm$^{-3}$) and $\lambda = 165.8 R_{\infty}^{s} (Z - 12.44)^{-1.94}$ for the laser wavelength of 532 nm (the critical density: $4 \times 10^{21}$ cm$^{-3}$), respectively. This result is close to the previous one. Specifically, $s = 20.86$ and 12.44 are the screening constants while Slater’s rule gives $s = 36$–39.15 for 4$d$ electrons. It is known that both the UTA width and its peak wavelength shift due to different abundances of ionic charge states in the plasma.

The photon flux was observed to decrease with increasing atomic number from $3 \times 10^{13}$ photons nm$^{-1}$ sr$^{-1}$ for $Z = 64$ to $4 \times 10^{13}$ photons nm$^{-1}$ sr$^{-1}$ for $Z = 83$. This photon flux was of the same order as in the previous report, where the driver laser pulse energy was 1 J.
class with a pulse duration of 10 ns. In the present results obtained by 100-mJ class lasers, the use of the sub-nanosecond pulse laser thus achieved efficient energy conversion from the laser pulse to the UTA emission, resulting from a change by one order of magnitude due to the higher electron temperature. Thus, the energy conversion was substantially enhanced by use of shorter duration pulses.

In order to calculate absorption effects, it is necessary to evaluate them with the radiative transfer equation. However, the lack of an adequate database of the equations of state for high-Z elements inhibits such a calculation. Thus, here we indirectly evaluate the spectral change of the emission by plasmas at different density. Because of their large transition probabilities, resonant lines that are strong in emission also strongly absorb in underdense, less than critical density or optically thick conditions. An optically thinner condition reduces opacity effects and increases the spectral efficiency of the ion distribution for efficient UTA emission. According to the present calculations of the atomic configuration-averaged atomic state, has also been developed.

According to the present calculations of the fundamental and second harmonics wavelengths of the induced laser pulse. This ratio of $I_{\lambda^{1064}} - I_{\lambda^{1064}}$ is shown in Fig. 3. We found that the relative decrease in the spectral intensities of emission produced by the fundamental and second harmonics wavelengths of the induced laser pulse. This ratio of $I_{\lambda^{1064}} - I_{\lambda^{1064}}$ is shown in Fig. 3. We found that the relative decrease in the spectral intensities was clearly evident along the UTA emission peaks, which became strong in emission also strongly absorb in underdense, less than critical density or optically thick conditions. An optically thinner condition reduces opacity effects and increases the spectral efficiency of the emitting region. Therefore, we can loosely optimize the electron temperature range to produce the optimum highly charged ion state distribution. In the present experiments, we fixed the laser intensity at $7 \times 10^{13}$ W/cm$^2$, corresponding to an electron temperature of the order of 500 eV calculated by the power balance model. The optimum electron temperatures and the ionic charge states of the $n = 4 - n = 4$ (Dn = 0) UTA transition emission are different for different elements. Therefore, the optimum laser intensity, corresponding to the electron temperature required to produce the optimum ion state distribution, is different for each UTA peak wavelength. We evaluated the electron temperatures and ionic charge states using the traditional CR model. Note that only ground state of each ion was taken into account. To compare the results of additional comparison to this one, a computer algorithm to generate a CR model with a sufficient large set of excited states of each ion, including the autoionizing states based on the nonrelativistic configuration-averaged atomic state, has also been developed.

Figure 4 shows charge-separated gA spectra calculated with the Flexible Atomic Code (FAC) and the electron temperature dependence of the atomic charge states for Gd in Figs. 4(a) and 4(c) and for Bi in Figs. 4(b) and 4(d), respectively. The charge states responsible for the UTA peak were Gd$^{4+}$–Gd$^{22+}$ ions for 6. x nm in Fig. 4(a) and Bi$^{28+}$–Bi$^{42+}$ ions for 3.9 nm in Fig. 4(b). The ranges of the electron temperature were evaluated to be 60–120 eV for Gd$^{4+}$–Gd$^{22+}$ ions for 6. x nm emission in Fig. 4(b) and 180–600 eV to Bi$^{28+}$–Bi$^{42+}$ ions for 3.9 nm emission in Fig. 4(d), respectively. According to the present calculations of the gA spectra by the FAC
and ionic populations by the CR model in Fig. 4, we mapped the optimum ionic temperatures and corresponding charge states required to produce strong UTA emission, and the results are shown in Figs. 5(a) and 5(b). The range of the electron temperatures necessary to produce the effective ionic charge states required is quite broad, which is a particular feature of the UTA emission. This result is also reproduced by the more detailed CR model computations.\

We found a relationship between the atomic number-electron temperature product dependence of the ionic charge states of the form \( q = 0.64 \times (ZT_e)^{0.38} \) for the UTA emission in the present results, close to the theoretical formula of \( q \approx (2/3) \times (ZT_e)^{1/3} \), where \( q, Z, \) and \( T_e \) are the charge state, the atomic number, and the electron temperature, as shown in Fig. 5(c). Moreover, the present results show that the charge states are different for HCI according to the regions of the optimum charge states consisting of the UTA spectra due to 4p–4d and 4d–4f array contributions, we have evaluated the atomic number dependences in \( Z = 50–83 \) using the FAC and CR model. We have also confirmed the relationship of \( q \approx 0.64 \times (ZT_e)^{0.38} \) between \( ZT_e \) and \( q \). The present quasi-Moseley’s law is sufficient for finding the optimum element for numerous applications, such as material, ablation and ionization, nano-lithography, and \textit{in vivo} biological imaging.

IV. SUMMARY

In summary, we have evaluated the formula of quasi-Moseley’s law as the \( Z \)-scaling at two different laser wavelength irradiation. The peak UTA wavelengths follow a quasi-Moseley’s law as \( \lambda = 33.82 \times R_m^{1.64}(Z - 20.86) \) nm for the laser wavelength of 1064 nm (the critical density of \( 1 \times 10^{22} \) cm\(^{-3}\)) and \( \lambda = 165.8 \times R_m^{1.94}(Z - 12.44) \) nm for the laser wavelength of 532 nm (the critical density of \( 4 \times 10^{21} \) cm\(^{-3}\)) respectively. The photon flux decreased with increasing atomic number. We have also mapped the optimum ionic temperatures and corresponding charge states required to produce strong soft x-ray UTA emission with a photon flux of the order of \( 10^{14} \) photons nm\(^{-1}\) sr\(^{-1}\) at the incident laser intensity of \( 7 \times 10^{13} \) W/cm\(^2\), which was the pulse energy of 130 mJ.

According to the regions of the optimum charge states consisting of the UTA spectra due to 4p–4d and 4d–4f array contributions, we have evaluated the atomic number dependences in \( Z = 50–83 \) using the FAC and CR model. We have also confirmed the relationship of \( q \approx 0.64 \times (ZT_e)^{0.38} \) between \( ZT_e \) and \( q \). The present quasi-Moseley’s law is sufficient for finding the optimum element for numerous applications, such as material, ablation and ionization, nano-lithography, and \textit{in vivo} biological imaging.

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