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Opacity effects on soft X-ray spectra from highly charged lanthanide ions in laser-produced plasmas

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Abstract

We have observed soft X-ray spectra from highly charged ions of seven different lanthanide elements with atomic numbers ranging from 60 to 70 in laser-produced plasmas (LPPs) using CO_2 and Nd:YAG laser systems, the wavelengths of which are 10.6 μm and 1.064 μm, respectively. The spectral feature drastically changes between the two types of LPPs due primarily to the difference in opacity. Narrowband quasicontinuum features arising from n=4–4 transitions, the centre wavelength of which systematically moves to shorter wavelength as the atomic number (Z) increases, are observed in the CO_2 LPPs, accompanied by sharp peaks coinciding with the strongest resonance lines of Pd-like ions for lower Z elements. In contrast, the quasicontinuum bands are broader and smoother in the Nd:YAG LPPs, appearing with bands of n=4–5 transitions on the shorter wavelength side. The results are also discussed based on comparisons with atomic structure calculations for ions with outermost 4d and 4f subshells.

Keywords: soft X-ray spectra, lanthanide ions, laser-produced plasmas, opacity

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1. Introduction

Soft X-ray emission spectra from highly charged lanthanide ions are of particular interest for basic atomic physics as well as applications to short wavelength light sources. In the last decade, laser-produced plasmas (LPPs) of gadolinium (Gd) and terbium (Tb) have been extensively investigated as possible candidates for light sources for semiconductor lithography in the wavelength range of 6–7 nm [1, 2, 3, 4]. Emission spectra of these plasmas typically form a quasicontinuum band, or the so-called unresolved transition array (UTA) [5], with contributions from overlapping n=4–4 transitions for a number of charge states having open 4d and 4f subshells. The UTA spectral features are also of interest in terms of physics of configuration interaction and N shell electron correlations [6].

Systematic studies of soft X-ray spectra from LPPs of lanthanide elements have been carried out so far in higher opacity conditions in which smooth broadband UTAs were recorded on photographic or microchannel plates [7, 8]. In general, the UTA spectral feature strongly depends on the opacity of the emitting plasma. In terms of the application to lithography, optically thin plasmas having lower density are more appropriate because of higher spectral efficiency for the required wavelength band. Therefore, several techniques have been explored to generate optically thin LPPs in the development of lithography at 13.5 nm using tin (Sn) plasmas [9, 10]. The solutions include the use of longer wavelength lasers (e.g., CO_2 laser) because critical plasma density is inversely proportional to the square of laser wavelength.

Recently, soft X-ray spectra from CO_2 LPPs of Gd have been newly reported and higher spectral purity has been obtained in comparison with Nd:YAG LPPs [11]. However, spectra from CO_2 LPPs of other lanthanide elements have not been reported yet. In this study, we have systematically observed soft X-ray spectra from highly charged ions of seven lanthanide elements with atomic numbers from Z=60 to 70 in CO_2 and Nd:YAG LPPs. The results are qualitatively interpreted in terms of opacity effects and comparisons with atomic structure calculations.

2. Experimental

All the experimental data in this article have been measured at Utsunomiya University where shortpulse laser systems are available. An ultra-shortpulse CO_2 laser and a Q-switched Nd:YAG laser (Continuum Inc.) were operated at wavelengths of 10.6 μm and 1.064 μm, respectively, to produce LPPs with different opacities. In order to produce a high-power CO_2 laser beam with a pulse duration of 3–20 ns, a semiconductor plasma shutter was used in combination with a master oscillator power amplifier (MOPA) system. The pulse duration of the Nd:YAG laser was fixed at 150 ps. The maximum beam energies are approximately 100 mJ for the CO_2 laser and 200 mJ for the Nd:YAG laser. Critical (cutoff) electron density n_{ec} in an LPP is inversely proportional to the square of the laser wavelength λ_{L}, $n_{\mathrm{ec}} \simeq 1.1\times10^{21}\lambda_{\mathrm{L}}^{-2}$, where n_{ec} and λ_{L} are in cm^{-3} and μm, respectively. Therefore, critical densities in CO_2 and Nd:YAG LPPs are estimated to be roughly 10^{19} and 10^{21} cm^{-3}, respectively.

The laser beams were introduced into a target chamber via a plano-convex lens to be focused onto a planar target made of pure lanthanide metals with Z=60, 62, 64, 65, 66, 68 and 70. The focal spot sizes are estimated to be 80 and 60 μm for the CO_2 and Nd:YAG lasers, respectively.

Time- and space-integrated soft X-ray emission spectra from the LPPs were recorded in the wavelength range of 2–10 nm with a flat-field grazing incidence spectrometer equipped with a 2400 grooves/mm grating and a back-illuminated soft X-ray CCD camera (Andor Technology). The spectral resolution was typically better than 0.005 nm. Single-shot spectra were recorded for the Nd:YAG LPPs, while 40 identical shots were accumulated to obtain one spectrum for the CO_2 LPPs due to weaker intensity.

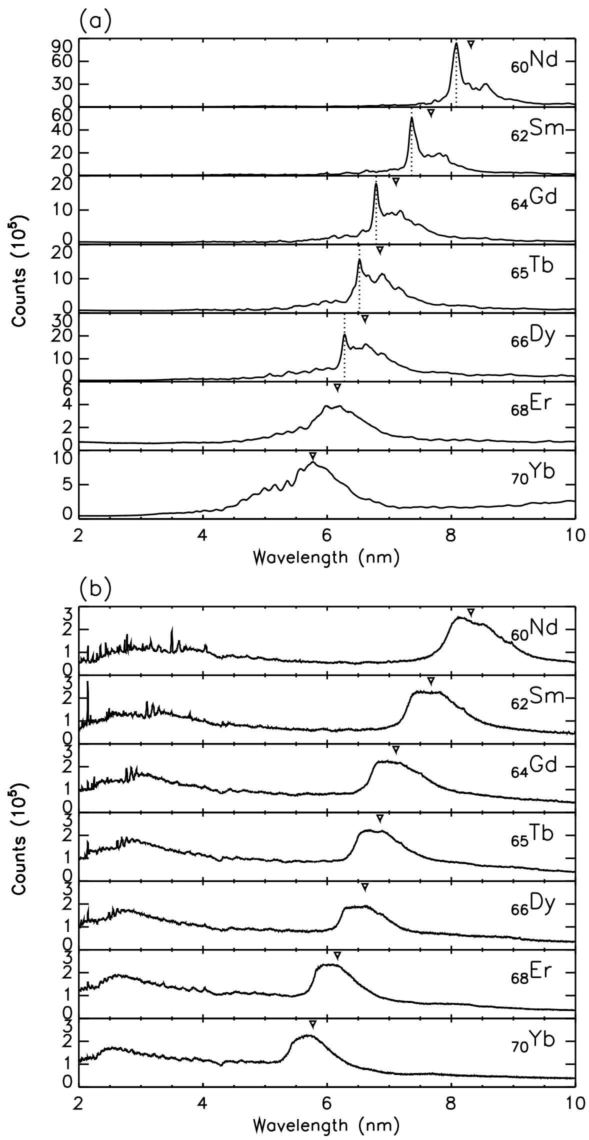

Figure 1: Time- and space-integrated soft X-ray emission spectra from highly charged ions of seven lanthanide elements with atomic numbers ranging from 60 (Nd) to 70 (Yb) observed in (a) CO_2 and (b) Nd:YAG LPPs. The laser power densities were (2–3)$\times 10^{10}$ and 4.8×10^{13} W/cm^2 in (a) and (b), respectively. Single-shot spectra are shown in (b), while 40 identical shots are accumulated to obtain one spectrum in (a). The vertical broken lines in (a) indicate the peaks coinciding with the strongest resonance lines of Pd-like ions. The n=4–4 UTA peak positions expected from the quasi-Moseley's law [12, 13] are marked by triangles.

3. Results

Figure 1 shows a series of soft X-ray spectra from highly charged ions of seven lanthanide elements in (a) CO_2 and (b) Nd:YAG LPPs. The laser power densities evaluated from the focal spot sizes for Fig. 1 (a) and 1 (b) are (2–3)$\times 10^{10}$ and 4.8×10^{13} W/cm^2, respectively, which are the maxima available in the present setup. As to the n=4–4 UTA positions, an empirical quasi-Moseley's law has recently been proposed [12, 13], given by $\lambda_{\mathrm{UTA}} = aR_\infty^{-1}(Z-s)^{-b}$, where λ_{UTA} is the peak wavelength of the UTA, a=21.86±12.09, b=1.52±0.12, s=23.23±2.87 and R_∞ is the Rydberg constant. The UTA peak positions expected from this law are also shown in Fig. 1 by triangles.

It is clearly seen in Fig. 1 that the spectral feature drastically changes between the two types of LPPs. The UTA features, the centre wavelength of which systematically moves from 8.1 to 5.7 nm as Z increases, have relatively narrower bandwidths and fine structures in the CO_2 LPPs, characterized by sharp peaks for the elements with Z=60–66 indicated by vertical broken lines in Fig. 1 (a). The positions of the sharp peaks systematically shift to shorter wavelength from the UTA peak positions expected from the quasi-Moseley's law. The emission intensities were much weaker and the UTA bandwidths were broader for erbium (Er) and ytterbium (Yb) in comparison with lower Z elements.

In contrast, the spectra measured in Nd:YAG LPPs show smoother broadband UTA features as well as rugged features with a number of small peaks on the shorter wavelength side of the main UTA as shown in Fig. 1 (b). The rugged features on the shorter wavelength side are negligibly weak against the main UTA features in the CO_2 LPPs. The Z dependence of the center wavelength of the main UTA in the Nd:YAG LPPs approximately follows the quasi-Moseley's law.

4. Sharp peaks in CO_2 LPPs

The earlier work on a CO_2 LPP of Gd suggests that the sharp peak originates from the strongest resonance transition $4d^{10}\ {}^1S_0$-$4d^9 4f\ {}^1P_1$ of Pd-like Gd^{18+}

overlapped with ^{2}F-^{2}D doublet lines of Ag-like Gd^{17+} [11]. In this work, we have compared the measured wavelengths of the sharp peaks in Fig. 1 (a) with those of the Pd-like resonance lines reported previously [14, 15] and calculated *ab initio* with the Flexible Atomic Code (FAC) code [16]. The results are summarized in Table 1 showing that the measured wavelengths for Z=60–66 are in very good agreement with the literature values. This indicates that charge states around Pd-like ions are dominant emitters in the CO_2 LPPs for lower Z lanthanide elements. As shown in Fig. 1 (a), this peak is unseen for Er and Yb with Z of 68 and 70, respectively. The absolute intensities of the overall soft X-ray emission were very weak for Er and Yb as mentioned in the previous section. These results imply that the CO_2 laser intensity in the present setup is too low to produce Pd-like ions for these higher Z elements having higher ionization energies.

The calculated wavelengths listed in Table 1 are systematically shifted to shorter wavelengths by 0.14–0.43 nm from the present or earlier measurements. This is probably because of the difficulty in the calculation of effective potential energy including complex correlation among N shell electrons for an inner 4d subshell excited configuration such as $4d^9 4f$.

In the Nd:YAG LPPs, the sharp peaks of the Pd-like resonance lines completely disappeared, and broader and smoother UTA features are observed as shown in Fig. 1 (b). When the laser energy is reduced to one order lower than the maximum, these features of the n=4–4 UTA are maintained though the emission intensity decreased over the entire spectral range. The doppler or other broadenings for each single spectral line are negligible because they are estimated to be much smaller than the instrumental width of the spectrometer. Therefore, these features are primarily attributable to higher opacities in the Nd:YAG LPPs in which strong self-absorption of the resonance lines occurs due to higher plasma density limited by n_{ec} in the emitting region.

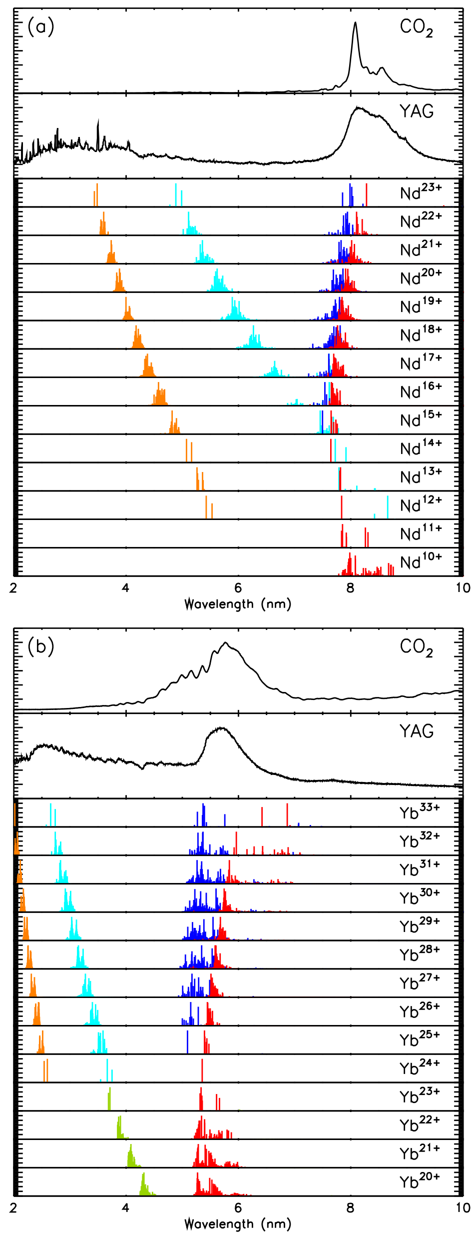

Figure 2: Comparisons of the normalized soft X-ray spectra measured in (a) Nd and (b) Yb LPPs with the line strength distributions of all ions with outermost 4d subshell electrons and some of lower charge states calculated with FAC code [16]. The calculated resonance transition types are 4d–4f (red), 4p–4d (blue), 4d–5p (light blue), 4d–5f (orange) and 4f–5g (green). The line strengths have been normalized for each type of transition to its maximum.

Table 1: Comparison of the wavelengths of the sharp peaks in CO_2 LPP with the reported and calculated wavelengths of Pd-like resonance line $4d^{10}\ ^1S_0$-$4d^9 4f\ ^1P_1$. The *ab initio* calculation has been performed with FAC code [16]. References: *a*–[14], *b*–[15]

		Wavelength (nm)		
Z	Ion	CO_2 LPP	Reported	Calculated
60	Nd^{14+}	8.08	8.0512^a	7.646
62	Sm^{16+}	7.36	7.3462^a	7.031
64	Gd^{18+}	6.79	6.7636^a	6.509
65	Tb^{19+}	6.52	6.5122^a	6.281
66	Dy^{20+}	6.28	6.2778^a	6.069
68	Er^{22+}	(unclear)	5.8609^b	5.689
70	Yb^{24+}	(unclear)	5.4996^b	5.355

5. Comparisons with calculations

In order to interpret the difference in the spectral feature, the measured spectra have been compared with the line strength distributions of resonance transitions calculated with the FAC code including the minimum numbers of configurations and their mixings. For example, the comparisons for neodymium (Nd) and Yb are shown in Fig. 2 (a) and 2 (b), respectively, where the normalized spectra in the two types of LPPs are plotted with the calculated line strength (gA: weighted transition probability) distributions of the resonance transitions of the types 4d–4f (red), 4p–4d (blue), 4d–5p (light blue), 4d–5f (orange) and 4f–5g (green). The line strengths in Fig. 2 have been normalized for each type of transition to its maximum. The atomic structure calculations in the FAC code is fully relativistic and *ab initio*. Note that the calculated wavelengths of n=4–4 transitions should systematically be shifted to shorter wavelength from the measurements as described in the previous section. Ground states for Nd^{10+}–Nd^{13+} include one or two 5s electrons because of 4f orbital collapse [17].

As shown in Fig. 2, the main UTA feature originates from n=4–4 transitions of a wide range of ion stages having 4d and 4f outermost subshells. This means that the n=4–4 emission of a particular charge state can be easily absorbed not only by the same charge state but also by other charge states in optically thick Nd:YAG LPPs. The 4d–5p and 4d–5f transitions contribute to the rugged feature with many individual peaks found on the shorter wavelength side in the Nd:YAG LPPs. The effect of absorption should be weaker for this feature because the wavelength of the n=4–5 UTA moves depending on ion charge.

A hydrodynamic simulation of the two types of Gd LPPs using MEDUSA code [18] indicates that the soft X-ray emitting region, where the electron temperature is roughly 100–200 eV, is located deep inside the plasma plume in Nd:YAG LPPs, while close to the expanding front in CO_2 LPPs [11]. Consequently, the intensity of n=4–5 emission is much more pronounced against the n=4–4 emission in the Nd:YAG LPPs than in the CO_2 LPPs because of the following two reasons:

- Larger absorption of n=4–4 feature by the 4d–4f resonances of lower ion stages (including open 4f ions) in lower temperature region surrounding the core plasma
- Larger population of n=5 levels as a result of collisional excitation due to higher electron density

It should be also noted that broader UTA features in the optically thick Nd:YAG LPPs contain contributions from satellite transitions relevant to doubly excited states such as $4d^{m-1}4f5s$, which appear at slightly longer wavelengths than the resonance transitions. The rugged structure observed on the shorter wavelength side of the main UTA in the CO_2 LPP of Yb may be contributed from n=4–5 transitions of ion stages lower than Yb^{20+}.

6. Summary

We have recorded a series of soft X-ray spectra from highly charged lanthanide ions with Z=60–70 in CO_2 and Nd:YAG LPPs having different opacities. As a result of the large difference in critical plasma density between the two types of LPPs, Pd-like resonance lines strongly dominate the spectra in the CO_2 LPPs, while smooth broadband UTA features are observed in the Nd:YAG LPPs. The intensity ratios of n=4–5 emissions to n=4–4 emissions are much larger in the Nd:YAG LPPs than in the CO_2 LPPs because of the effects of larger opacities and collisional excitations.

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