EUV spectra from highly charged terbium ions in optically thin and thick plasmas

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Abstract. We have observed extreme ultraviolet (EUV) spectra from terbium (Tb) ions in optically thin and thick plasmas for a comparative study. The experimental spectra are recorded in optically thin, magnetically confined torus plasmas and dense laser-produced plasmas (LPPs). The main feature of the spectra is quasicontinuum emission with a peak around 6.5–6.6 nm, the bandwidth of which is narrower in the torus plasmas than in the LPPs. A comparison between the two types of spectra also suggests strong opacity effects in the LPPs. A comparison with the calculated line strength distributions gives a qualitative interpretation of the observed spectra.

1. Introduction

Extreme ultraviolet (EUV) emission from highly charged heavy ions in laser-produced plasmas (LPPs) has widely been investigated in terms of their potential applications as EUV light sources. For example, quasicontinuum emission, referred to as unresolved transition array (UTA), from tin plasmas, the peak of which is around 13.5 nm, is now being industrialized for the EUV lithography of semiconductor devices [1]. The expected future trend in the semiconductor feature size has recently prompted the survey of deep EUV emission from highly charged lanthanide ions such as gadolinium (Gd) and terbium (Tb) at around 6.5–6.7 nm [2,3]. However, theoretical models of EUV spectra in dense LPPs are complicated because of opacity effects such as line broadening and self-absorption. Therefore, it would be worthwhile to make a comparative study of EUV spectra between LPPs and optically thin plasmas for benchmarking.

In this article, we present EUV spectra from highly charged Tb ions observed in optically thin and thick plasmas for a comparative study. Optically thin, high-temperature plasmas are produced in the Large Helical Device (LHD) at the National Institute for Fusion Science, while dense LPPs are generated using Nd:YAG lasers at University College Dublin. Though we have already performed a similar study for Gd [4], Tb should also be investigated because the bandwidth of a candidate multilayer mirror lies between the emission peaks of Gd and Tb [5]. In addition, signal-to-noise ratios of the data in the LPPs have been improved since the previous study by utilizing a new grazing incidence spectrometer with better throughput. Therefore, we have observed not only Tb but also Gd spectra with the new experimental setup. Also,
Table 1. Some of the major parameters of the experimental setup for the two lasers with ns and ps pulses.

|                      | ns-laser        | ps-laser        |
|----------------------|-----------------|-----------------|
| Model                | Continuum Surelite III | EKSPLA SL312P   |
| Maximum energy       | ≃ 670 mJ        | ≃ 250 mJ        |
| Pulse width          | ≃ 7 ns          | ≃ 180 ps        |
| Maximum power density on targets | ≃ 2×10^{13} W/cm^2 | ≃ 5×10^{14} W/cm^2 |

some qualitative interpretations are given based on the calculations of line strengths using a Hartree-Fock with configuration interaction (HFCI) Cowan suite of codes [6].

2. Experimental
The LPP experiment was performed using two Q-switched Nd:YAG lasers with 7 ns and 180 ps pulses at the wavelength of 1064 nm. The laser beam is guided into a chamber and focused onto pure Gd and Tb metal targets, placed on a movable stage, by a plano-convex lens (76 mm focal length at 1064 nm). Some of the major parameters of the experimental setup for these ns- and ps-lasers are summarized in table 1.

EUV spectra in LPPs were recorded with a 0.25 m grazing incidence spectrometer connected to the target chamber via a 75 μm entrance slit. The spectrometer is equipped with a flat field 2400 lines mm\(^{-1}\) grating (Shimadzu) and a CCD camera (Andor, DX436-BN) for direct detection of EUV photons. The wavelength range of this setup is 2–9 nm. The absolute wavelength was calibrated using known emission lines of silicon and nitrogen ions from LPPs using silicon nitride targets. Space- and time-integrated single spectrum can only be recorded for each laser shot. In order to obtain enough signal intensity, we usually accumulate 5–20 identical laser shots reproduced by moving the target stage.

Since the experimental setup used in the LHD has already been described elsewhere [7], only a brief explanation is given here. Small amounts of Gd and Tb powders are introduced into high-temperature hydrogen plasmas, the density of which are of the order of 10^{19} m\(^{-3}\), using tracer encapsulated solid pellets (TESPEL) [8]. Spatial profiles of electron density and temperature are measured precisely by a laser Thomson scattering diagnostic system [9]. EUV spectra are recorded by a 2 m Schwob-Fraenkel grazing incidence spectrometer [10] equipped with a 600 lines mm\(^{-1}\) grating. The temporal evolutions of EUV spectra in the 5.6–9 nm range were recorded with a frame rate of 0.1 or 0.2 s.

3. Results and discussion
EUV spectra from Tb ions observed in LPPs for the ns- and ps-lasers (referred hereafter as ns-LPP and ps-LPP) at their maximum energies are shown in figure 1 (a) in red and green, respectively. A sharp drop of the emissivity at 4.3 nm seems to arise from the K-edge absorption of carbon in the deposition on the surface of the grating or the CCD chip. Apart from this absorption feature, LPP spectra are characterized by broad quasicontinuum emission with a main peak around 6.6 nm and rugged structure with several noticeable peaks found on the shorter wavelength side of the main peak. As is well known, the main peak is caused by the UTA of the \(n=4\)–4 transitions of open N shell ions. The rugged structure is more pronounced in the ns-LPP than in the ps-LPP. The small dip found at 6.5 nm in the ns-LPP is not observed in the ps-LPP. Indeed, this dip is more pronounced in a Gd plasma (not shown in the figure).
Figure 1. Tb spectra observed in (a) LPPs and (b) LHD plasmas compared with (c) the weighted probabilities of spontaneous transitions for Tb XVII–XXIX calculated by the HFCI Cowan suite of codes.

For a comparison, two different types of EUV spectra from Tb ions observed in LHD are shown in figure 1 (b) in the range of 5.6–9 nm. The spectra in blue and orange are recorded when the peak electron temperature is around 400 eV (low) and 1100 eV (medium), respectively. It is apparent that the UTA bandwidth of the \( n=4 \rightarrow 4 \) transitions in the LHD plasmas is narrower than that in the LPPs. In particular, the spectrum is drastically narrowed with a sharp peak near 6.5 nm, and a clear double peak is observed near 6.9 nm under the low-temperature condition in the LHD. The positions of these peaks agree well with the previously identified lines of Ag-like Tb XIX and Pd-like Tb XX [11,12], which will be discussed elsewhere in detail. Interestingly, the position of the small dip in the ns-LPP spectrum completely agrees with that of the sharp peak in the low temperature LHD plasma, which is also the case in Gd plasmas. This indicates a strong effect of self-absorption by Ag-like and Pd-like ions in the ns-LPP.

The \( n=4 \rightarrow 4 \) and \( n=4 \rightarrow 5 \) transitions of Tb ions have recently been calculated [13,14] using the Cowan code and Flexible Atomic Code (FAC) [15]. In this study, we have compared the measured spectra with the line strengths calculated by the Cowan code to interpret them qualitatively. The weighted probabilities of spontaneous transitions for all of the open 4d and some of the open 4f subshell Tb ions with simple electronic configurations are shown in figure 1 (c). The minimum number of excited states and configuration interactions among them are included.
in the calculations. Following the general procedure in the preceding studies [2,13,16], Slater-Condor and configuration interaction parameters were empirically scaled to 90% of their \textit{ab initio} values, while the spin orbit integrals were unchanged. Ion stages lower than In-like Tb XVII have more complex ground-state configurations because of an interplay of 4f and 5s or 5p electrons [17]. It is obvious that the spectra in the medium temperature LHD plasma can be well explained by the spectral bands of \( n=4 \rightarrow 4 \) transitions for open 4d subshell ions. On the other hand, the LPP spectra have broad tail structure on the longer wavelength side, which seems to include the contributions from open 4f ions.

The present calculation also indicates the possibility of the contributions from \( n=4 \rightarrow 5 \) transitions to the rugged feature in the shorter wavelength side of the LPP spectra because the positions of \( n=4 \rightarrow 5 \) (\( \Delta n=1 \)) transitions move to shorter wavelength as the ion stage increases. Indeed, similar structures for tungsten and gold have been previously identified as \( n=4 \rightarrow 5 \) transitions [16,18]. The detailed analysis of these features is underway.

4. Summary
In terms of the application to semiconductor lithography, we have observed EUV spectra from highly charged Tb ions in dense LPPs and optically thin LHD plasmas by grazing incidence spectrometers. A comparative analysis of EUV spectra between optically thick and thin plasmas clearly shows the differences in the spectral feature and opacity effects. Though the rugged feature observed on the shorter wavelength side in the LPP spectra seems to arise from \( n=4 \rightarrow 5 \) transitions, further studies are necessary to make detailed identifications of these structures.

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