Charge exchange spectroscopy in Sn\(^{q+}\)(\(q = 6−15\))-He collisions

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Abstract. Extreme ultra-violet (EUV) spectra of multiply charged tin ions were measured in the wavelength range of 5–38 nm following the electron capture into excited states of slow Sn\(^{q+}\)(\(q = 6−15\)) ions passing through He gas target. The charge-state dependence of 4d-\(n\ell\)\((n\ell = 4f, 5p\) and 5f) transitions was obtained with assumption of the single-electron capture. Identification of the transitions has been carried out by comparison with the results of the theoretical calculation.

1. Introduction

Recently, the development of an extreme ultra-violet (EUV) light source for next generation semiconductor lithography is advancing a grate deal [1]. For the EUV lithography, the wavelength of 13.5 nm was chosen because Mo/Si multi-layer coated mirrors have a maximum of the reflectivity about 70% at this wavelength with 2% bandwidth. Tin plasmas have attracted attention as a candidate of a powerful EUV light source, because of their favorable emission around 13.5 nm [2]. The EUV emissions from Sn plasmas in the wavelength around 13.5 nm are considered to mainly correspond to optical transitions of multiply charged Sn ions with a wide range of charge states (from Sn\(^{7+}\) to Sn\(^{12+}\)) [3, 4]. Spectroscopic data of Sn ions for individual charge states is necessary to diagnose and to understand tin plasmas. Not only experimental investigations but also theoretical calculations of Sn plasmas have been performed to optimize them for the development of the EUV light source [5]. Many kinds of plasma, produced in such as capillary discharge and vacuum spark, were mainly used for the experimental spectroscopy of multiply charged Sn ions [3, 6]. These plasmas contain various charge states of Sn ions, hence data analysis must be complicated and difficult to be carried out. Therefore, the theoretical calculation with the relativistic atomic code is very useful to identify the observed emission lines.

Photon emission spectroscopy (PES) is mainly used for the cross section measurements of charge transfer processes. The line intensities of the photon emissions following the charge transfer reaction in collisions of multiply charged ions with neutral target gases are measured in the PES. However, it is also a very powerful experimental technique to investigate the transition energies and energy levels of multiply charged ions. In this study, we measured the EUV
emissions from charge-selected Sn ions with PES to provide the fundamental spectroscopic data of Sn ions which are necessary for the development of the EUV light source.

2. Experiment

Multiply charged Sn ions were produced in a 14.25 GHz ECR (electron cyclotron resonance) ion source at Tokyo Metropolitan University [7]. We inserted a rod of sintered tin oxide (SnO$_2$) in a plasma chamber and introduced O$_2$ gas of about 1×10$^{-5}$ Pa as a support gas for the plasma. The Sn$^{q+}$ ($q = 6$–15) ions were extracted with an electric potential of 20 kV and selected by a 110° double-focusing dipole magnet according to their mass-to-charge ratios. The ion-beam was directed into a collision chamber, where the ion-beam intersected an effusive beam of target gas ejected from a multi-capillary plate. The background pressure in the collision chamber was directed into a collision chamber, where the ion-beam intersected an effusive beam of target gas. The primary ion-beam, which had about 6 mm diameter, had typically electrical current of 0.01–1 μA as measured with a Faraday cup located behind the collision region.

Optical radiation in the EUV region from the collision volume was observed at 90° to the ion-beam direction with a compact flat-field grazing-incident spectrometer (SSK-260, Shin Seiki Co.) equipped with a toroidal-type converting mirror and a variable line space (ca. 1200 lines/mm) grating blazed at 100 nm. A liquid nitrogen cooled CCD (charge coupled device) camera (C4880, Hamamatsu) was installed in the EUV spectrometer. Emissions in a 18 nm range of wavelengths were observed and accumulated simultaneously. The slit width of 200 μm between the conversing mirror and the grating gave the wavelength resolution of about 0.03 nm. The calibration of wavelength was performed with observation of more than 20 lines of O VI and O VII in collisions of O$^{10+}$ and O$^{7+}$ with Xe gas. The uncertainty of observed wavelength was estimated at 0.02 nm.

3. Results and Discussion

Measured spectra in collisions of Sn$^{q+}$ ($q = 6$–15) with He at 20 keV/q are shown in Figure 1. In some spectra, the second-order diffraction was also observed with very strong intensity. Hence, we can see the emission peak corresponding to $2\lambda$ in the longer wavelength region when the peak is observed at $\lambda$ of the shorter wavelength. As can be seen at $\lambda \approx 10.5$ nm in the Sn$^{10+}$-He system. In low-energy collisions of Sn$^{q+}$ with He, the following processes take place:

\[
\text{Sn}^{q+} + \text{He} \rightarrow \text{Sn}^{(q-1)+}(n\ell) + \text{He}^+ \quad \text{single electron capture (SEC),}
\]

\[
\rightarrow \text{Sn}^{(q-2)+}(n\ell n'\ell') + \text{He}^{2+} \quad \text{true double electron capture (TDC),}
\]

\[
\rightarrow \text{Sn}^{(q-1)+}(n\ell) + \text{He}^{3+} + e^- \quad \text{transfer ionization (TI).}
\]

The emission from Sn$^{(q-1)+}$ and Sn$^{(q-2)+}$, therefore, must be considered for the incident charge of $q$. Generally speaking, in slow collisions of multiply charged ions with He, SEC is the dominant process and TI has a significant contribution, while TDC has very small cross section, as observed in Xe$q^+$ ($q = 10$–43), Ar$q^+$ ($q = 8$–16), and other multiply charged ions [8, 9, 10, 11]. Therefore, we can assume that the final charge states in the product Sn ions are $(q-1)+$ in collisions of Sn$^{q+}$ ions with He.

EUV emissions of multiply charged Sn ions are corresponding to the transitions of 4d-4f, 4d-5p, 4d-5f and 4p-4d, which stand for 4d$^k$-4d$^{k-1}$4f, 4d$^k$-4d$^{k-1}$5p, 4d$^k$-4d$^{k-1}$5f and 4p$^5$4d$^{k-1}$, 4p$^5$4d$^k$ transitions, respectively. Since the 4d$^{k-1}$n$\ell$ shell has quite a number of fine-structure levels, the 4d-n$\ell$ transitions make a band-like emission, which is called an UTA (unresolved transition array). In Figure 1, the arrows indicate the averaged wavelengths of the 4d-4f, 4d-5p and 4d-5f transitions for Sn$^{(q-1)+}$ ions calculated with the HULLAC (Hebrew University Lawrence Livermore Atomic Code) code.
Figure 1. EUV emission spectra in collisions of Sn$^{q+}$($q = 6−15$) with He at the energy of 20 keV/q. The arrows indicate the averaged wavelengths of the transitions calculated with the HULLAC code.

Figure 2. Charge-state dependence of the mean wavelengths of the 4d-4f, 4d-5p and 4d-5f transitions: solid symbols, peak positions of the observed prominent emissions; open symbols, averaged wavelengths calculated with the HULLAC code.

An averaged wavelength of the UTA is obtained by calculation as

$$\lambda_{\text{ave}} = \frac{\hbar c}{\sum_{i,j} (g_i A_{ij} E_{ij}) / \sum_{i,j} (g_i A_{ij})},$$

(1)

where $g_i$ is the multiplicity of the initial state $i$, $A_{ij}$ is the probability of transition between $i$ and $j$ states, and $E_{ij}$ is the transition energy. The 4p-4d transitions, which have very similar averaged wavelengths to those of the 4d-4f transitions in the theoretical calculation, are omitted in this figure.

These calculated wavelengths and the observed wavelength corresponding to these transitions are shown in Figure 2 for charge states of Sn ions between 5+ and 14+. Experimental peak positions corresponding to UTAs are determined by careful comparison with results of theoretical calculations. In the Sn$^{7+}$-He collision, we do not assign a very broad UTA around 19 nm to the 4d-4f UTA and regard it as the mixture of 4d-4f and 4p-4d transitions. And the prominent peak around 14 nm in the spectrum for Sn$^{8+}$-He can be considered as the 4d-6p transitions in agreement with the theoretical results.

The qualitatively good agreement in the comparison of observed and theoretical wavelengths, as seen in Figure 2, indicates that the prominent emission peaks are attributed to the single
electron capture and/or the transfer ionization which provides the charge states of \((q-1)^+\) in collisions of Sn\(^{q+}\) ions with He. In contrast to excellent agreements in the 4d-5p transitions, the 4d-4f transitions have approximately constant differences about 0.5 nm in higher charge states. This discrepancy in the 4d-4f transitions had also been observed in the cases of Xe ions [12]. As the reason of this finding, we consider the strong interactions between 4p\(^5\)4d\(^{k-1}\)4f\(^1\) and 4p\(^5\)4d\(^{k+1}\)4f\(^0\) configurations [13,14], which have been pointed out by O’Sullivan and Faukner [15]. The theoretical results with the HULLAC code in this work are similar to those with the Cowan code [4,5]. Both of the calculations are not perfectly coincident with the experimental observation in this work. Therefore, more accurate calculations should be performed for understanding of energy levels and transition wavelengths of multiply charged Sn ions.

In this study, we have confirmed that the emissions from Sn\(^{q+}\)(\(q=7\) and \(10 \leq q \leq 14\)) contribute to the EUV emission from Sn plasmas around 13.5 nm, which should be used for the EUV lithography.

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References
[1] Bakshi V 2006 EUV Source for Lithography (Washington: SPIE press)
[2] Svendsen W and O’Sullivan G 1994 Phys. Rev. A 50 3710
[3] Kieft R E, Graloff K, van der Mullen M A J J and Banine V 2005 Phys. Rev. E 71 036402
[4] White J, Hayden P, Dunne P, Cummins A, Murphy N, Sheridan P and O’Sullivan G 2005 J. Appl. Phys. 98 113301
[5] Kieft R E, van der Mullen M A J J, Kroesen W M G, Banine V and Koshelev N K 2005 Phys. Rev. E 71 026409
[6] Azarov V I and Joshi Y N 1993 J. Phys. B 26 3405
[7] Tanuma H, Matsumoto J, Nishide T, Shiromaru H and Kobayashi N 2001 J. Chin. Chem. Soc. 48 389
[8] Andersson H, Astner G and Cederquist H 1988 J. Phys. B 21 L187
[9] Selberg N, Biedermann C and Cederquist H 1996 Phys. Rev. A 54 4127
[10] Vancura J, Marchetti J V, Perotti J J and Kostroun O V 1993 Phys. Rev. A 47 3758
[11] Justiniano E, Cocke L C, Gray J T, Dubois R, Can C, Waggoner W, Schmidt-Boecking R and Ingwersen H 1984 Phys. Rev. A 29 1088
[12] Tanuma H, Ohashi H, Shibuya E, Kobayashi N, Okuno T, Fujikawa S, Nishimura H and Nishihara K 2005 Nucl. Instr. Meth. B 235 126
[13] Koike F, Azuma Y, Hasegawa S, Nagata T and Nishihara K 2005 J. Electron. Spec. Relat. Phenom. 144-147 1227
[14] Koike F and Fritzsche S 2006 Radiat. Phys. Chem. in press
[15] O’Sullivan G and Faukner R 1994 Opt. Eng. 33 3978