Vacuum ultraviolet spectra in charge transfer collisions of multiply charged Sn ions

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Abstract. We measured the vacuum ultraviolet (VUV) emission spectra following charge transfer collisions of Snₙ⁺ (q = 3–6) with neutral rare gas targets at collision energies of 50–100 keV. We identified some of emission lines from reported energy levels. Several lines were observed in collisions of different charge states of ions. This finding means that the multiple-electron capture processes can take place effectively in our experiment. Significant target dependences were observed in the spectra with the projectile of Sn⁴⁺ and Sn⁶⁺.

1. Introduction
Nowadays the wavelength of the light for the semiconductor lithography is 193 nm, which is emitted from an ArF excimer laser. Since shorter wavelength is necessary for higher integration of semiconductor devices, the extreme ultraviolet (EUV) light, ≈10 nm, will be applied to the next-generation semiconductor lithography. The EUV light is easily absorbed by the air or optical lenses, so the EUV exposure system must be in vacuum and transmissive lenses are not available. The Mo/Si multilayer mirrors having peak reflectivity around 13.5 nm in the EUV region will be used for light focusing instead of optical lenses. Therefore light sources which emit the 13.5 nm light had been searched for actively [1-3]. Consequently, the laser-produced Sn plasma was selected as a light source [4-6]. However, spectroscopic data about multiply charged Sn ions were limited; therefore, it was hard to identify emission lines from Sn plasmas. To provide the EUV spectroscopic data of multiply charged Sn ions, we have measured the charge-selected EUV emission spectra by means of charge exchange spectroscopy [7].

The emission of 13.5 nm with 2% band width is called “in-band” emission. However, not only the “in-band” light around 13.5 nm, but also the “out-of-band” light with wavelengths longer than 130 nm can be reflected by the Mo/Si mirror efficiently [8]. Hence, VUV spectroscopic data of Sn ions were also required. In this work, we measured VUV emission spectra by charge exchange spectroscopy as we have done for the EUV spectra.

2. Experiment
Multiply charged Sn ions were produced as before [7]. We used a 14.25 GHz electron cyclotron resonance ion source (ECRIS) at Tokyo Metropolitan University. We put sintered tin oxide (SnO₂) pellets inside the plasma chamber at the ECRIS. Multiply charged Sn ions produced in the ECRIS were extracted with an electric potential of 20 kV for q = 4–6 or 17.5 kV for q = 3,
and were charge-separated with a magnetic field. We mention that collision energy dependences of emission cross sections are not significant in such energy region.

Charge-selected incident ions were made to collide with neutral gas targets in a collision chamber. The projectile beam current was measured with a Faraday cup at the end of beamline: it was typically 1 µA for \( q = 4 \)-6 or 0.5 µA for \( q = 3 \). The target gas jet was ejected from a multi-capillary plate to cross the ion beam. During the experiment, the gas pressure was low enough to avoid multiple collisions.

The emitted light following charge transfer collisions was observed by a Seya-Namioka spectrometer equipped with a Peltier-cooling CCD camera. The spectrometer was set up vertically to the ion beam. The measured wavelength range was from 40 nm to 225 nm. However, in the range of 145 nm to 225 nm, no emission line was observed. The resolution was determined by a slit which was located at the entrance of the spectrometer: FWHM was 0.4 and 1.0 nm for the slit width of 100 and 300 µm, respectively.

3. Result
3.1. Charge State Dependence
Figure 1 shows emission spectra in collisions of Sn\(^{q+}\) \((q = 3\text{–}6)\) with Xe. The emission lines labeled as 1–4 are listed in table 1.

![Figure 1](image.png)

**Figure 1.** Emission spectra in collisions of Sn\(^{q+}\) \((q = 3\text{–}6)\) with Xe. The emission lines labeled as 1–4 are listed in table 1.

In the Sn\(^{3+}\) - Xe collision, we observed only the 5s\(^2\) \((^1S_0)\)–5s5p \((^1P_1)\) emission line. At \( q = 4 \) we observed several lines, and identified almost all of them. We can see many emission lines at \( q = 5 \); most of them are the 4d\(^9\)5s–4d\(^9\)5p transitions, in which states have fine structures. In case of \( q = 6 \), we could not identify emission lines because of the absence of information about energy levels and wavelengths.

Generally single electron capture (SEC) is dominant in low energy collisions of highly charged...
ions with neutral targets \[11\]:

\[
\text{Sn}^{q^+} + \text{Xe} \rightarrow \text{Sn}^{(q-1)+} + \text{Xe}^+ \rightarrow \text{Sn}^{(q-1)+} + h\nu + \text{Xe}^+.
\]  

(1)

In addition, transfer ionization (TI) or autoionizing double capture (ADC) play an important role \[12\] :

\[
\text{Sn}^{q^+} + \text{Xe} \rightarrow \text{Sn}^{(q-2)+} + \text{Xe}^{2+} \rightarrow \text{Sn}^{(q-1)+} + e^- + \text{Xe}^+.
\]  

(2)

However, in cases of low charged ions, the cross section of the multiple electron capture might become comparable to that of the SEC because of the energy matching \[13\]. In figure 1, there exist several lines which were observed simultaneously in different spectra. The emission lines labeled as 1 and 2–4 were observed in the spectra of \( q = 3 \) and 4, and in those of \( q = 4 \) and 5, respectively. These lines observed in larger \( q \) should be caused by the true double electron capture (TDC), i.e.

\[
\text{Sn}^{q^+} + \text{Xe} \rightarrow \text{Sn}^{(q-2)+} + \text{Xe}^{2+} \rightarrow \text{Sn}^{(q-2)+} + h\nu + \text{Xe}^+.
\]  

(3)

3.2. Target Dependence

Figure 2 shows the target dependence of the emission spectra in collisions of \( \text{Sn}^{4+} \) with rare gas targets. The resolution is 1.0 nm for Ne target and 0.4 nm for others. The emission intensity is normalized with the incident ion current and the target gas pressure. The emission lines labeled as 1–6 are listed in table 1.

The electron configuration of \( \text{Sn}^{4+} \) is \( 4d^{10} \) for the ground state. After the SEC, electron configuration would be \( 4d^{10}nl \). On the other hand, it would be \( 4d^{10}nl'nl'' \) in the cases of TDC. The emission lines 1 and 5, which are identified as the \( 5s^2 - 5s5p \) and \( 5s5p - 5s5d \) of Sn III, are due to the TDC, whereas the others are due to the SEC.

We can see the strong target dependence in the intensity of the \( 5s^2 - 5s5p \) of Sn III and in the intensity-ratio of the \( 5s - 5p(2P_3/2) \) to the \( 5s - 5p(2P_1/2) \). The emission line of the \( 5s^2 - 5s5p \) appears for Ar, Kr and Xe targets. The emission intensity of this line is the strongest for Kr target. Although the intensity-ratio of the \( 5s - 5p(2P_3/2) \) to the \( 5s - 5p(2P_1/2) \) should be \( 2 : 1 \) if the population of the \( 5p \) states simply obeys the statistical weight, it shifts away especially in the case of Ar target.

Figure 3 shows a series of emission spectra in collisions of \( \text{Sn}^{6+} \) with rare gas targets. The resolution was 1.0 nm in these spectra. A significant target dependence can be seen. However, as we mentioned before, we could not identify emission lines. A precious calculation for transition energies considering electron correlations in detail is desired to identify emission lines.

| Line no. | Wavelength / nm | Charge state | Lower state | Upper state |
|----------|-----------------|--------------|-------------|-------------|
|          | this work       | NIST \(^a\) | Moore \(^b\) |             |
| 1        | 125.2           | 125.992      | 125.14      | III         |
| 2        | 112.0           | 112.068      | 111.93      | IV          |
| 3        | 131.5           | 131.455      | 131.45      | IV          |
| 4        | 143.8           | 143.752      | 143.75      | IV          |
| 5        | 134.8           | ——           | 134.76      | III         |
| 6        | 107.4           | 107.341      | ——          | IV          |

\(^a\) The wavelengths obtained from the NIST database \[10\]. \(^b\) The wavelengths deduced from the reported energy levels \[9\].

Table 1. A list of some identified lines. The line numbers correspond to labels in figure 1 and 2.
Figure 2. Emission spectra in collisions of Sn$^{4+}$ with rare gases. The emission lines labeled as 1–6 are listed in table 1.

Figure 3. Emission spectra in collisions of Sn$^{6+}$ with rare gases.

4. Conclusion
The vacuum ultraviolet (VUV) emission spectra after charge transfer collisions of Sn$^{q+}$ ($q = 3–6$) with rare gas targets were measured. Emission lines following both single and double capture were observed. Significant target dependences were observed in the spectra with the projectile of Sn$^{4+}$ and Sn$^{6+}$. A precious calculation for transition energies is desired to identify emission lines especially for the projectile of Sn$^{6+}$.

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