4d-4f unresolved transition arrays of xenon and tin ions in charge exchange collisions

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Abstract. Extreme ultra-violet emission spectra of multiply charged Xe and Sn ions are measured following the electron capture in collisions with He. After the assumption of single-electron capture, we have found that the wide range of charge states, Sn¹⁰⁺ to Sn¹⁴⁺ for tin ions, and Xe¹¹⁺ to Xe¹⁷⁺ for xenon ions, have the prominent emissions at 13.5 and 11.0 nm, respectively. These emissions are attributed to the 4d-4f unresolved transition arrays in comparison with the results of theoretical calculations by using the HULLAC code.

1. Introduction
High temperature plasmas of heavy elements can be used as sources of extreme ultra-violet (EUV) light. Not only line emissions but also widely spread emissions called unresolved transition arrays (UTA) have been observed from the EUV light source plasma. It is known that the UTA around 11 nm of Xe plasma and that around 13.5 nm of Sn plasma have quite strong emissivity. In particular, the 13.5 nm of Sn plasma is applied to the promising light source for the next generation photo-lithography [1].

These UTAs have been found by O’Sullivan and his coworkers in laser-produced plasmas and known as the 4d-4f transitions of multiply charged ions with the half-filled N-shell configurations [2, 3]. Because of a large number of fine-structure transitions, the resonance emission lines compose a broad transition array. The details of the UTAs have been investigated with several kinds of multi-configuration relativistic atomic codes, e.g. Cowan code, HULLAC (Hebrew University Lawrence Livermore Atomic Code), GRASP (General purpose Relativistic Atomic Structure Program), RCI (Relativistic Configuration Interaction atomic structure code), and FAC (Flexible Atomic Code) [4, 5, 6, 7, 8, 9, 10, 11]. Also quite a number of experimental works on Xe and Sn plasmas have been reported in this decade [12, 13, 14, 15, 16]. However, the experimental study of optical emissions from Xe and Sn ions has been scarcely performed without the plasmas which contain several kinds of ionic species.

Intimate spectroscopic information on multiply charged Xe and Sn ions is necessary to understand the physics of EUV plasmas of Xe and Sn. The energy levels and observed spectral lines of Xe in all stages of ionization which were published before December 2002 has been compiled by Saloman [17]. The available data, however, is quite scarce for higher charge states,
in particular, for more than 11+. For multiply charged Sn ions, the situation is much worse than for Xe ions. Recently, we have measured the EUV emission spectra of charge-identified ions of Xe and Sn by means of charge-exchange spectroscopy to provide fundamental atomic spectroscopic data of these ions [18, 19]. In this work, we report the charge-state dependence of the 4d-4f UTAs of multiply charged Xe and Sn ions.

2. Experimental
Multiply charged ions were produced in a 14.25 GHz ECR (electron cyclotron resonance) ion source at Tokyo Metropolitan University [20]. Xe gas was introduced in a plasma chamber for making Xe ions, and O$_2$ gas was added as a mixing gas to improve the ion intensities of higher charge states. The ratio of O$_2$ to Xe in the chamber was between 0.1 and 1, and the total gas pressure was about 2×10$^{-5}$ Pa. On the other hand, to obtain Sn ions, we inserted a rod of sintered tin oxide (SnO$_2$) in the plasma chamber and introduced O$_2$ gas of about 1×10$^{-5}$ Pa as a support gas for the plasma. The multiply charged ions were extracted from the plasma 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 6×10$^{-6}$ Pa and the target gas pressure in the chamber was held at about 1×10$^{-3}$ Pa during the measurements. In this setup, we could not directly measure the target gas density in the collision volume. The target gas pressure, however, was low enough to maintain single-collision conditions. 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 converging 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 within a 18 nm range of wavelengths were observed and accumulated simultaneously. The slit width of 200 μm between the converging mirror and the grating gave a wavelength resolution of about 0.03 nm. The calibration of wavelength was performed by the observation of more than 20 lines of O VI and O VII in collisions of O$_6^+$ and O$_7^+$ with Xe gas. The uncertainty in the observed wavelength was estimated at 0.02 nm.

3. Results and discussion
Observed emission spectra in collisions of Sn$^{q+}$ (q = 10–15) and Xe$^{q+}$ (q = 11–18) with He target gas are shown in Figures 1 and 2, respectively. In the slow collisions of multiply charged ions with He gas, single-electron capture (SEC), true double-electron capture (TDC), and transfer ionization (TI) take place as inelastic reaction processes. Generally speaking, SEC can be regarded as the most dominant one as seen in collisions of Xe$^{q+}$ (q = 10–43) and Ar$^{q+}$ (q = 8–16) [21, 22, 23]. Therefore, we can assume that the observed emissions come from the ions in one less charge states than the incident ionic charge $q+$, i.e. (q-1)+.

In these figures, the averaged wavelengths of 4d-4f transitions of (q-1)+ ions calculated with the HULLAC code are shown as short bars. Obviously, the theoretical calculation always gives the wavelengths of about 0.5 nm shorter than the prominent peaks in measured spectra for both Sn and Xe ions. This systematic discrepancy between the experimental results and the theoretical calculations can be considered as the result of strong electronic correlation in the n = 4 shell, which is difficult to estimate accurately in the multi-configuration Dirac-Fock method. This effect have been discussed for Sn$^{12+}$ with careful analysis using the GRASP code by Koike et al. [9, 24]. They have found that the 4p$^6$4d$^14f^4$ and 4p$^5$4d$^3$4f$^0$ configurations...
mix strongly and the optical 4p-4d and 4d-4f transitions take place coherently in Sn$^{12+}$. In other charge states of Sn ions and also Xe ions, similar effects must be involved to provide the prominent UTA in the EUV emission spectra, as has been pointed out by O’Sullivan and Faukner [25].

The peak wavelengths of the 4d-4f UTA from Sn$^{10+}$ to Sn$^{14+}$, which are produced in the single-electron-capture collisions of Sn$^{q+} (q = 11–15)$ ions with He, are almost same as 13.5 nm. And every charge state between 11+ and 17+ of Xe ions has a peak at 11.0 nm. Such weak charge dependence in the wide range of charge states causes the concentrated strong emission from the plasmas as light sources.

The measured spectra have many important features other than the 4d-4f UTA. In lower charge states of both Sn and Xe ions, other UTAs have been observed with significant intensities and identified as 4d-5p and 4d-5f transitions after comparison with the theoretical calculations. The detailed discussion on other UTAs, however, is beyond the scope of this paper.

In collisions of Sn$^{14+}$, Sn$^{15}$, Xe$^{17+}$, and Xe$^{18+}$ with He, the EUV emission spectra show similar structures at longer wavelengths than the 4d-4f UTAs. These comb-like emissions, i.e. a number of discrete sharp lines, are tentatively assigned as the 4p-4d transitions [26]. Since both of Sn$^{14+}$ and Xe$^{18+}$ have Kr-like configurations, we expect some common reason why the specific comb-like structure appears in the iso-electronic ions. However, this structure is not so clearly observed in collisions of Sn$^{13+}$ despite the fact that it obviously appear in collisions of Xe$^{17+}$ which has the Rb-like electronic configuration, as Sn$^{13+}$. We hope that what’s behind...
these emissions will be revealed by the theoretical investigations. And furthermore we have a plan to measure EUV emission spectra for more highly charged Xe and Sn ions, at the very least Xe$^{19+}$, to compare the result on Sn$^{15+}$ both of which are Br-like ions.

In this paper, we have not describe the relative emission intensities of the 4d-4f UTA transition in charge-exchange collisions, which are related with both of the electron-capture cross-sections and the oscillator strengths of transitions. The experimental results on the emission intensities will be published anywhere else with the consideration based on the scaling law for the charge-transfer cross-sections.

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