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<tr>
<td>Publication date</td>
<td>2011</td>
</tr>
<tr>
<td>Publication information</td>
<td>Physica Scripta, 2011 (T144): 014026</td>
</tr>
<tr>
<td>Publisher</td>
<td>IOP Publishing</td>
</tr>
<tr>
<td>Item record/more information</td>
<td><a href="http://hdl.handle.net/10197/3749">http://hdl.handle.net/10197/3749</a></td>
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<tr>
<td>Publisher's version (DOI)</td>
<td>10.1088/0031-8949/2011/T144/014026</td>
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Configuration Interaction in Charge Exchange Spectra of Tin and Xenon

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Abstract. Charge state specific extreme ultraviolet spectra from both tin ions and xenon ions have been recorded at Tokyo Metropolitan University. The Electron Cyclotron Resonance Source spectra were produced from charge exchange collisions between the ions and rare gas target atoms. In order to identify unknown spectral lines of tin and xenon, atomic structure calculations were performed for Sn¹⁴⁺ - Sn¹⁷⁺ and for Xe¹⁶⁺ - Xe²⁰⁺ using the Hartree-Fock with Configuration Interaction code of Cowan. The energies of the capture states involved in the single electron process that occurs in these slow collisions are estimated using the classical over barrier model.

1. Introduction

Charge state specific spectra from a range of tin and xenon ions have been recorded in the extreme ultraviolet (EUV) wavelength region at Tokyo Metropolitan University. The study was prompted by the importance of these species in source development for EUV lithography and metrology at the operating wavelength set by industry of 13.5nm [1]. In Sn, an intense unresolved transition array (UTA) resulting from 4p⁷4dⁿ – 4p⁷4dⁿ⁺¹ + 4p⁷ 4dⁿ¹4f transitions in Sn IX through Sn XIII emits at the desired wavelength [2,3]. However as the spectra from individual ion stages overlap in energy and plasma sources always contain a range of ion stages, unambiguous line identification is almost impossible in such regions of high line density. Thus ion separation techniques are an essential prerequisite to any analysis. In this work, we report on the EUV emission spectra of multiply charged Sn and Xe ions measured following electron capture in collisions with He.
2. Experimental
The detailed description of the experimental setup has been given elsewhere and will only be briefly presented here [4]. Multiply charged ions were produced in a 14.25 GHz ECR (electron cyclotron resonance) ion source at Tokyo Metropolitan University. The $\text{Sn}^{q+}$ and $\text{Xe}^{q+}$ 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 ratio. The ion beam was directed into a collision chamber, where it interacted with a target gas jet. The background pressure in the collision chamber was $6 \times 10^{-6}$ Pa and the target gas pressure in the chamber was held at about $1 \times 10^{-3}$ Pa during the measurements and was low enough to guarantee single-collision conditions. The primary ion-beam, which was approximately 6 mm in diameter, had typically an electrical current of 0.1–2 $\mu$A as measured with a Faraday cup located behind the collision region. The EUV emission from the collision center was observed at 90° to the ion beam direction with a compact flat-field grazing-incidence spectrometer equipped with a toroidal collecting mirror and a 1200 lines/mm grating blazed at 100 nm. The detector was a liquid nitrogen cooled CCD (charge coupled device) camera (C4880, Hamamatsu) which enabled an emission spectrum in the wavelength range of 6–24 nm to be accumulated simultaneously. A slit of width 200 $\mu$m placed between the mirror and the grating gave an instrumental resolution of approximately 0.03 nm. The uncertainty in the observed wavelength was estimated at 0.02 nm.

3. Results and discussion
The emission spectra of Sn and Xe ions obtained at the ECR source are shown in figure 1 and figure 2, respectively.

![Figure 1. EUV emission spectra in collisions of $\text{Sn}^{q+}$ with He](image1)

![Figure 2. EUV emission spectra in collisions of $\text{Xe}^{q+}$ with He](image2)

In slow collisions of multiply charged ions with He, single electron capture may be regarded as the dominant process with transfer ionisation making a significant contribution [5-9]. Therefore the final charge states in the production of Sn and Xe ions due to collisions of $\text{Sn}^{q+}$ with He and $\text{Xe}^{q+}$ with He are $\text{Sn}^{(q-1)+}$ and $\text{Xe}^{(q-1)+}$ respectively.
For single electron processes the energies of the capture states can be estimated from the classical over barrier model [10, 11].

\[ E_{\text{capture}} = I^{q-1} - \left(2 \sqrt{q} + q\right)(2 \sqrt{q} + 1)^{-1} I^{T} \]  

(1)

where \( I^{q-1} \) is the ionisation potential of the projectile ion after capture and \( I^{T} \) the ionisation potential of the target gas. The model assumes that states exist in the vicinity of \( E_{\text{capture}} \). The validity of equation (1) improves if there is a high density of states at this energy, i.e. for capture into a high \( n \) state and for compact high-\( q \) projectile which is the case here. From equation (1) the states populated with maximum probability by electron capture are those with energies close to 290eV above the ground level for Sn\(^{14+}\) and those with energies close to 330eV above the ground level for Xe\(^{17+}\) corresponding to high \( l, n = 7 \) or lower \( l, n = 8 \) orbitals. The energy level scheme for Xe\(^{17+}\) is shown in figure 3. For the following ion stages the capture states are essentially the same. Thus the \( n = 4 \) are not the dominant electron capture levels in these collisions and the states must derive from a cascade process that initially favoured the population of higher levels. For Yrast decays, following capture into the high \( l \) and \( j \) states [12], population of excited 4f states is favoured and thus we would expect to observe 4f-4d transitions.

Calculations were performed using the Hartree Fock Configuration Interaction (HFCI) code of Cowan [13] in order to interpret the intense UTAs exhibited by the measured spectra of Xe and Sn ions. In spectra where the strongest transitions satisfy \( \Delta n = 0 \) configuration interaction (CI) effects have a dramatic effect on the spectra due to the proximity of excitation energies [14, 15]. From the calculations it was found that the spectra were dominated by excited to excited state transitions while the resonance lines appeared relatively weak. The calculations showed that the UTA arises from \( 4p^5 4d^{m+1} - 4p^4 4d^{m+2} + 4p^5 4d^m 4f \) transitions in the case of open 4d subshells and \( 4p^{m-1} 4d - 4p^{m-1} 4f + 4p^{m-2} 4d^2 \) transitions for stages with a 4p valence subshell. For Sn XV this calculation gave good agreement with the observed spectrum but for higher stages of Sn it was found necessary to allow for interaction with the lower energy core excited \( 4s4p^{m+1} \) configuration for optimum agreement. This interaction was also included in the Xe calculations although the effects were found to be less pronounced, presumably as the energy separation of the interacting configurations is greater in the case of Xe. In both the Sn and Xe spectra CI effects were found to have a very strong impact on the emission profiles and lead to a dramatic spectral narrowing in each ion stage. Figures 4 and 5 illustrate the effects of CI where a direct comparison is made between calculations with and without the inclusion of CI.
The EUV spectra produced from charge exchange collisions of Sn ions and Xe ions with helium are dominated by excited to excited state transitions. The 4-4 transitions seem to derive from a cascade process that favours selective population of the excited levels leading to relatively strong emission from a number of lower oscillator strength transitions resulting in a broadening of the array. The population of the upper doubly excited states in the transfer process may result from either a transfer excitation process in which electron capture is accompanied by the excitation of a 4p or 4d electron or a transfer ionization process in which the projectile ion autoionizes to a doubly excited state.

References

Figure 3. Comparison between CI (top) and non-CI (bottom) calculations for Sn XVII
Figure 4. Comparison between CI (top) and non-CI (bottom) calculations for Xe XX