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L X-ray energy shifts and intensity ratios in tantalum with C and N ions – multiple vacancies in M, N and O shells

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Abstract. The energy shifts and intensity ratios of different L X-ray components in tantalum element due to 10 MeV carbon and 12 MeV nitrogen ions are estimated. From the observed energy shifts, the possible number of simultaneous vacancies in M shell are estimated. A comparison of $L_\alpha/L_{\beta 2,15}$, $L_{\beta 1}/L_{\gamma 1}$ and $L_{\gamma 2,3}/L_{\gamma 4,4}$ with the ratios due to Scofield theoretical transition rates indicate that the number of multiple vacancies in N shell are higher than the vacancies in M and O shell. Employing Larkin's statistical scaling procedure, the number of possible multiple vacancies in N and O shells are estimated quantitatively.

Keywords. L X-ray energy shifts and intensity ratios; heavy ions; estimation of multiple vacancies in M, N, O shells.

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1. Introduction

Study of atomic inner shell ionization process is important to understand ion–atom interaction process. Vacancies in the atomic inner shells are produced by photons, X-rays and charged particles. Study of atomic inner shell ionization by heavy ion bombardment is of current interest because of the production of multiple vacancies. These spectator vacancies in the higher shells may not be filled prior to radiative filling of the vacancies in the lower shells, thus causing different screening potential to the transition electrons which result in an increase in the binding energies. The presence of multiple vacancies causes a change in (a) X-ray energies, (b) X-ray intensity ratios and (c) fluorescence yield values. Several authors [1–10] have observed that the X-ray energy shifts in heavy ion collision process are relative to the X-ray energies of a single hole atom. Li and Watson [11], and Ramachandra Rao *et al* [12] have observed different K X-ray intensity ratios in heavy ion collision process compared with the values due to photon excitation.

Not much work is carried out in the study of L X-ray energy shifts and intensity ratios with heavy ion projectiles. This is because of the complex nature of L X-ray spectrum.

Olsen *et al* [13] studied the L X-ray spectrum of Sn with proton, α-particles and oxygen ion bombardment and observed the presence of simultaneous multiple vacancies in M shell in addition to a vacancy in L shell. Bissinger *et al* [14] studied the L X-ray spectrum of Au bombarded with 12–15 MeV oxygen ions and observed L X-ray energy shifts. Burkhalter *et al* [15] have observed satellite peaks of L_α and L_β lines by bombarding Ge with α particle and interpreted these energy shifts due to 3d shell vacancies. Saha *et al* [16] have observed a variation of $L_{\beta 1}/L_\alpha$ intensity ratio using gas targets. Similar L X-ray energy shifts have been observed by Sarkadi and Mukoyama [17], Venugopala Rao *et al* [18], and Schonfelds [19] during heavy ion collision process. Uchai *et al* [20] used Ag ions and observed the L X-ray energy shifts of different L X-ray components in some high Z elements. They have calculated the possible number of multiple vacancies in M, N, O shells using the intensity ratios of suitable L X-ray components. In the present work the L X-ray energy shifts and intensity ratios in tantalum element using carbon and nitrogen projectiles are measured. From the measured energy shifts and intensity ratios, and applying Larkins scaling procedure, the possible number of simultaneous vacancies in M, N, O shells are estimated.

2. Experimental details

The present experiments are carried out using 3MV pelletron accelerator facility at the Institute of Physics, Bhubaneswar. Carbon (4^+) ions with 10 MeV energy and nitrogen (5^+) ions with 12 MeV energy are employed to excite the sample. Thin foils of tantalum prepared by vacuum deposition technique are employed in the present work. Targets are kept at an angle of 45° to the beam direction. The resulting L X-ray spectrum is recorded with Si(Li) detector, which is kept at an angle of 90° to the beam direction. The resolution of the detector is 160 eV (FWHM) at 5.9 keV energy. The target is first exposed to a proton beam of energy 2 MeV and the resulting L X-ray spectrum of Ta is recorded. In the same gain conditions, the L X-ray spectra due to carbon and nitrogen ion bombardment are also recorded. The gain stability of the system is continuously monitored using a pulsar. Spectra are collected for sufficiently long time to ensure good statistics. The overlapped L X-ray spectrum of Ta with proton and carbon ion bombardment is shown in figure 1.

3. Data analysis

3.1 *L X-ray energy shifts*

It is observed from figure 1 that different L X-ray components due to carbon ion bombardment are broadened and their centroids are shifted towards high-energy side relative to proton bombardment. The shift in centroid of the L X-ray line due to carbon ion bombardment is referred to as ‘energy shift’. The number of channels corresponding to the shifts in certroids of different L X-ray components due to carbon and nitrogen ion bombardment are estimated relative to the centroids of L X-ray components due to proton bombardment. The corresponding energy shifts are obtained by multiplying the number of shifted channels with eV per channel. The resulting energy shifts of different L X-ray components due to carbon and nitrogen ion bombardment are shown in table 1.

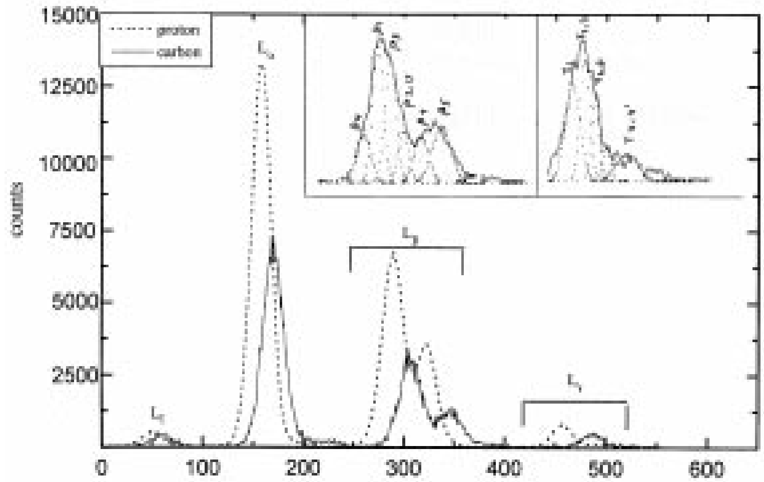

Figure 1. Overlapped L X-ray spectrum of tantalum with proton and carbon.

Table 1. Energy shifts of different L X-ray components in Ta due to carbon and nitrogen ions.

No.	X-ray components	Energy shifts (eV) Carbon	Nitrogen
1	L_1 ($L_3 \to M_1$)	61	62
2	L_α ($L_3 \to M_{4,5}$)	88	114
3	$L_{\beta 1}$ ($L_2 \to M_4$)	105	132
4	$L_{\beta 2,15}$ ($L_3 \to N_{4,5}$)	229	255
5	$L_{\gamma 1}$ ($L_2 \to N_4$)	255	317

3.2 *L X-ray intensity ratios*

The L X-ray spectrum is very complex. The L_β line consists of several components in which $L_{\beta 1}$, $L_{\beta 2,15}$, $L_{\beta 3}$ and $L_{\beta 4}$ components are of considerable intensity. Similarly the L_γ line is also complex having several components in which $L_{\gamma 1}$, $L_{\gamma 2,3}$, $L_{\gamma 4,4}$ components are of considerable intensity. The complex L_β and L_γ spectral lines are decomposed into different components and their areas are estimated using FIT software program [21]. The intensities of different L X-ray components are estimated from the respective areas by applying efficiency correction. The efficiency values are taken from the efficiency graph described in our earlier paper [22]. These intensities are corrected for absorption in the target material. The attenuation coefficients used in self-absorption correction for different L X-ray components are taken from the tables of Storm and Israel [23]. Using the intensities of different L X-ray components thus obtained, the intensity ratios L_α/L_1, $L_\alpha/L_{\beta 2,15}$, $L_{\beta 1}/L_{\gamma 1}$ and $L_{\gamma 2,3}/L_{\gamma 4,4}$ are estimated. The intensity ratios thus obtained for tantalum element due

Table 2. Intensity ratios of different L X-ray components of Ta with 10 MeV carbon and 12 MeV nitrogen ions. The number of possible multiple vacancies in M, N and O shells in Ta due to carbon and nitrogen ions are also included.

No.	Intensity ratios	Experimental		Theoretical Scofield	Multiple vacancies	
		Carbon	Nitrogen		Carbon V_M, V_N, V_O	Nitrogen V_M, V_N, V_O
1	$\frac{L_\alpha(L_3 \to M_{4,5})}{L_1(L_3 \to M_1)}$	18.2(9)	18.1(9)	21.08	4,18,6	5,18,5
2	$\frac{L_\alpha(L_3 \to M_{4,5})}{L_{\beta 2,15}(L_3 \to N_{4,5})}$	7.13(35)	7.14(35)	5.7		
3	$\frac{L_{\beta 1}(L_2 \to M_4)}{L_{\gamma 1}(L_2 \to N_4)}$	9.32(45)	9.21(45)	5.4		
4	$\frac{L_{\gamma 2,3}(L_1 \to N_{2,3})}{L_{\gamma 4,4}(L_1 \to O_{2,3})}$	4.33(21)	4.20 (20)	6.8		

to carbon and nitrogen ions are given in table 2. In the same table, the theoretical intensity ratios due to Scofield [24] theoretical transition rates are also given.

4. Results and discussions

4.1 *Estimation of multiple M vacancies*

From the observed energy shifts, the number of possible M vacancies is estimated as follows: Uchal *et al* [25] have calculated energy shifts of different L X-ray components for different M shell spectator vacancies using Dirac–Fock computer program. The present experimental energy shifts are compared with the theoretical energy shifts due to Uchal *et al* [25] and the possible number of spectator vacancies in M shell are estimated. The number of possible M vacancies, thus obtained in tantalum element with 10 MeV carbon and 12 MeV nitrogen ions are $V_M = 4, 5$ respectively. The number of M vacancies due to nitrogen ions (5^+) is higher than those due to carbon (4^+) ions. This may be due to higher charge state of nitrogen ions. Uchai *et al* [20] have observed the number of M vacancies in Ta as 8 which is higher than the present value. This may be due to the use of different projectiles.

4.2 *Estimation of multiple vacancies in N and O shells*

From table 2, it may be seen that the intensity ratio L_α/L_1 is less than the ratio due to Scofield theoretical transition rates. This indicates that more numbers of simultaneous vacancies are produced in $M_{4,5}$ sub-shells than M_1 sub-shell. The intensity ratios $L_{\beta 1}/L_{\gamma 1}$ and $L_\alpha/L_{\beta 2,15}$ are higher than the same ratios due to Scofield transition rates. The deviation between the experimental and theoretical intensity ratios may be due to the presence of multiple vacancies. The higher $L_{\beta 1}(L_2 \rightarrow M_4)/L_{\gamma 1}(L_2 \rightarrow N_4)$ and $L_\alpha(L_3 \rightarrow M_{4,5})/L_{\beta 2,15}(L_3 \rightarrow N_{4,5})$ intensity ratios over Scofield theoretical values indicate that more number of vacancies are produced in N shell than in M shell. The intensity ratio $L_{\gamma 2,3}(L_1 \rightarrow N_{2,3})/L_{\gamma 4,4}(L_1 \rightarrow O_{2,3})$ are lower than the intensity ratio due to Scofield transition rates which suggest that less number of simultaneous vacancies are produced in O shell than in N shell.

The number of simultaneous vacancies in N and O shells are quantitatively estimated using Larkin's statistical scaling procedure [26]. According to this scaling procedure, the Scofield transition rates are multiplied by a scaling factor to reproduce the experimental values. The number of multiple vacancies in N shell are estimated using the relation

$$\left(\frac{L_{\beta 1}(L_2 \rightarrow M_4)}{L_{\gamma 1}(L_2 \rightarrow N_4)}\right)_{\text{exp}} = \left(\frac{L_{\beta 1}}{L_{\gamma 1}}\right)_{\text{Sco}} * \left(\frac{n_M - V_M}{n_M}\right) * \left(\frac{n_N}{n_N - V_N}\right)$$

where $(L_{\beta 1}/L_{\gamma 1})_{\text{exp}}$ represents the experimental ratio, $(L_{\beta 1}/L_{\gamma 1})_{\text{Sco}}$ represent the ratio of Scofield transition rates, n_M, n_N, represent the total number of electrons in M and N shells and V_M, V_N, represent the number of multiple vacancies in M and N shells respectively. Substituting the value of V_M (obtained from energy shifts) and $n_M = 18$, $n_N = 32$, the number of N vacancies are calculated. The number of N vacancies thus obtained in tantalum element due to 10 MeV carbon and 12 MeV nitrogen ions respectively are $V_N = 18, 18$.

The number of simultaneous vacancies in O shell are calculated with the experimental intensity ratio $L_{\gamma 2,3}/L_{\gamma 4,4}$ and using relation

$$\left(\frac{L_{\gamma 2,3}(L_1 \rightarrow N_{2,3})}{L_{\gamma 4,4}(L_1 \rightarrow O_{2,3})}\right)_{\text{exp}} = \left(\frac{L_{\gamma 2,3}}{L_{\gamma 4,4}}\right)_{\text{Sco}} * \left(\frac{n_N - V_N}{n_N}\right) * \left(\frac{n_O}{n_O - V_O}\right).$$

Substituting the values $n_N = 32$, $n_O = 18$ and $V_N = 18$, the number of simultaneous vacancies in O shell for tantalum due to C and N ions respectively are obtained as $V_O = 6, 5$.

From these results, it is concluded that the number of simultaneous vacancies in N shell are higher than vacancies in M shell and the number of simultaneous vacancies in O shell are less than the vacancies in N shell.

5. Conclusions

From the present experiment, the following conclusions are arrived:

1. The energies of different L X-ray components due to carbon and nitrogen ion bombardment are shifted towards high-energy side relative to the energies due to proton bombardment. The energy shifts may be due to the production of simultaneous vacancies in M, N and O shells. Comparing these energy shifts with Uchai theoretical values, the number of possible simultaneous vacancies in M shell is estimated.
2. The intensity ratios $L_{\beta 1}(L_2 \rightarrow M_4)/L_{\gamma 1}(L_2 \rightarrow N_4)$ and $L_\alpha(L_3 \rightarrow M_{4,5})/L_{\beta 2,15}(L_3 \rightarrow N_{4,5})$ are higher than the intensity ratios due to Scofield theoretical values. This indicates that more number of vacancies are produced in N shell than in M shell. Similarly, the intensity ratio $L_{\gamma 2,3}(L_1 \rightarrow N_{2,3})/L_{\gamma 4,4}(L_1 \rightarrow O_{2,3})$ is lower than the

same ratio due to Scofield theoretical transition rates which indicate that less number of vacancies are produced in O shell than in N shell. Using Larkin's statistical scaling procedure, the number of simultaneous vacancies in N and O shell are estimated. The number of M, N and O vacancies in tantalum element due to C and N ions are respectively obtained as 4,18,6 and 5,18,5.

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