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## ORIGIN OF THE OBSERVED ${\rm L\beta}_2$ satellite X-rays of tungsten not predicted by theory

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Résumé

Au cours de l'étude experimentale de la raie  $L\beta_2$  du tungstène, nous avons observé des radiations dont l'énergie est d'accord avec des transitions de Coster-Kronig  $L_1 \rightarrow L_3 M$ , non prévues par la théorie.

#### Abstract

We observed satellite X-rays in the LB<sub>2</sub> spectrum of tungsten with energies which show that<sup>2</sup> they originate from atoms with a second hole in the M level. We conclude that this extra hole was created by Coster-Kronig L<sub>1</sub>+L<sub>3</sub>M transitions which existing calculations do not predict.

#### 1. Introduction

The L X-ray satellite lines, characterized by slightly higher energies than their parent or diagram lines, are mainly due to the following processes in multihole configurations:

1. Satellites originated from LM and LN double hole states created by Coster--Kronig  $L_1 \rightarrow L_{2,3}$  and  $L_2 \rightarrow L_3$  transitions or by shake-off in M,N levels following the  $L_i$  (i=1,2,3) ionization. Satellite lines originated from double ionized states can be divided in several classes: LM  $\rightarrow$  MM and LM  $\rightarrow$  MN, which are usually separated from the parent lines (visible satellites), and LN  $\rightarrow$  MN and LN  $\rightarrow$  NN, which cannot be resolved from their parent lines (hidden satellites).

2. Satellites due to shake-off and Coster-Kronig transitions which yield states  $L_iMM$ ,  $L_iMN$  and  $L_iNN$ ; the last one leads to hidden satellites.

 $L_1 \rightarrow L_{2,3}M_i$  (i=1 to 5) Coster-Kronig transitions provide the dominant mechanism for the production of visible satellites and consequently, when they become energetically impossible, the intensities of the corresponding satellite lines will drop suddenly. The probability of shake-off in the M subshells when a single hole is created in the  $L_1$  subshell has been found theoretically to be very small [1].

Theoretical calculations of the energies of non-radiative transitions  $L_1 \rightarrow L_3 M_i$ ( i=1 to 5) for tungsten[2] predict that one cannot obtain a system with a second hole in the M level from an atom initially monoionized in the  $L_1$  level.

In this work we analyse experimental results obtained for the L $_2$  X-ray line (arising from the L $_3 \rightarrow N_5$  transition) and its satellites.

According to the Coster-Kronig theory, primary ionization of the  $L_1$  atomic shell is followed by the  $L_1 \rightarrow L_3$  radiationless transition with the simultaneous ejection C9-610

of a M,N electron, the atom being left in the double ionized state  $L_3X$  (X=M,N); a subsequent single electron transition gives origin to a satellite line. If the energy  $L_1 \rightarrow L_3$  is large enough to allow for a second hole in the M level, we may expect to observe  $L_{\beta_2}$  visible satellites.

Richtmeyer *et al.* [3] observed for tungsten two Lß satellite lines, denoted by Lß'<sub>2</sub> (shifted by 5.82 x.u. from the diagram line) and Lß''<sub>2</sub> (shifted by 8.50 x.u.); Raoult [4] observed also a line, denoted by Lß<sup>I</sup><sub>2</sub> (shifted by 6 x.u.). These authors considered these lines as Lß<sub>2</sub> visible satellites. However, according to theory they cannot be due to Coster-Kronig transitions.

To search for the origin of these lines, we observed the  $L\beta_2$  X-ray region of tungsten at different exciting electron energies.

#### 2. Experimental procedure

Lß X-ray spectra of tungsten were obtained using a Cauchois spectrograph (r=50 cm) fitted with a quartz crystal 2 mm thick. The spectra were recorded in photographic Kodirex film, single coated, and analysed by means of a Joyce-Loebl microdensitometer. The electron beam energies of the X-ray tube were measured by means of a vary accurate digital voltmeter. The electronic current was just below 20 mA.

#### 3. Results and conclusions

We observed the  $L\beta_2$  region of the X-ray spectrum of Tungsten, while varying the exciting electron energy ( $E_e$ ) between 10.5 keV and 50 keV. As the ionization energy of the  $L_3$  level of tungsten is  $WL_3 = 10.202$  keV, whereas for the  $L_1$  level that energy is  $WL_1 = 12$ . 0996 keV, we reached about four times the ionization energy of the  $L_1$  level.

The values obtained for the ratio  $I_s$  of the LB<sub>2</sub> satellite band intensity to the LB<sub>2</sub> diagram line intensity (in fact the LB<sub>2</sub> line is mixed with the LB<sub>15</sub> line, the latter being very weak) as function of the exciting electron energies are listed in Table I. The satellite band is seen only for electron energies above WL<sub>1</sub>. However, the ratio  $I_s$  is not constant as the electron energy varies. For  $E_e = 13$  keV to  $E_e = 20$  keV there is a small increase of  $I_s$  and above  $E_e = 20$  keV the value of  $I_s$  remains constant. For values of  $E_e$  below 13 keV it is not possible to measure the intensity of the satellite band. Due to uncertainties in the background determination, an error of 20% may be ascribed to the  $I_s$  values.

In Fig. 1 we show part of a microphotogram of the  $L\beta_2$  region of the W X-ray spectrum for E\_ = 50 keV.

The range of wavelengths of the satellite band goes from 4.5 x.u. to 8.1 x.u. relative to the diagram line. These values are in good agreement with the results of Richtmeyer *et al.* [3] and Raoult [4].

We conclude that the observed satellite band is not due to shake-off because it is not observed below the  $L_1$  ionization energy and its peculiar behaviour as the electron energy varies exclude also this possibility. However, the present results are in agreement with a Coster-Kronig origin of the satellite band. In fact, expe-

E <sub>e</sub>	I <sub>s</sub> (a)
(Ker)	
10.5	
11.0	
11.8	
12.0	
12.5	(b)
13.0	$0.57 \times 10^{-2}$
14.5	$0.70 \times 10^{-2}$
17.0	$0.90 \times 10^{-2}$
20.0	$1.50 \times 10^{-2}$
25.0	$1.60 \times 10^{-2}$
30.0	$1.70 \times 10^{-2}$
50.0	$1.96 \times 10^{-2}$

Table I. Ratio  ${\rm I}_{\rm S}$  of the intensity of the  ${\rm L\beta}_2$  satellite band to the diagram line intensity as a function of the exciting electron energy ( $E_{e}$ ).

(a) An error of 20% has been estimated for the values of I . (b) For this value of  $\rm E_e$  a very weak satellite band is seen.



Fig. 1. Microphotogram showing the  $L\beta_2$  line of tungsten and its satellite band, for 50 keV exciting electron energy.

rimental results of the ratio of the ionization cross sections of subshells L<sub>1</sub> and L<sub>3</sub>,  $\sigma_1/\sigma_3$  (M. L. Carvalho *et al.*, umpublished), show that this ratio increases abruptly at 13 keV incident electron energy and becomes constant for energies above 20 keV. As the increase of  $\sigma_1/\sigma_3$  corresponds to a relative increase of L<sub>1</sub> vacancies, this leads also to an increase of L<sub>1</sub>  $\neq$  L<sub>3</sub> Coster-Kronig transitions.

The work of Chen *et al.* [2] predicts that Coster-Kronig transitions  $L_1 \rightarrow L_3 M_5$  are energetically forbidden for  $50 \le Z \le 74$  and are allowed for  $Z \ge 75$ . In their tables the energy difference of levels  $L_1$  and  $L_3 M_5$  for W (Z = 74) is -2.70 eV, this element being in the borderline between the two regions of Z. Thus, we suggest that new accurate calculations of Coster-Kronig energies should be performed in this region of atomic numbers to remove this discrepancy between theory and experiment.

References

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