Transition assignments to L\(\beta_1\) satellites in X-ray emission spectra of first transition series

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Received 24 July 2001; revised 18 October 2001; accepted 29 October 2001

The X-ray satellite spectra arising due to \(2P_{1/2} 3x - 3x3d\) \((x = s,p,d)\) transition array, in elements with \(Z = 26\) to 30, have been calculated. The bar at the top denotes that these are the positions of ionization in the atom. While the energies of various transitions of the array have been determined by using available Hartree-Fock-Slater data on \(1s - 2p3x\) and \(2p_{1/2} - 3x3x'\) Auger transition energies, their relative intensities have been estimated by considering cross-sections of singly ionized \(2x\) \((x = s,p)\) states and then of subsequent Coster Kronig and shake off processes. The calculated spectra have been compared with the measured satellite energies in \(L\beta_1\) spectra. Their intense peaks have been identified as the observed satellite lines. The one-to-one correspondence between the peaks in calculated spectra and the satellites in measured spectra has been established on the basis of the agreement between the separations in the peak energies and those in the measured satellite energies. It has been established that two satellites observed in the \(L\beta_1\) region of the X-ray spectra of the elements with \(Z = 26\) to 30 and named \(\beta_{II}\) and \(\beta_{II}'\) in order of increasing energy are mainly emitted by \(2P_{1/2} 3d - 3d_{2}\) transitions. The satellite \(\beta_{I}'\) has been assigned to the superposition of the transitions \(3P_2 - 3P_2, 3P_2 - 3P_2\) and \(3P_2 - P_1\), contributing in order of decreasing intensity, and the line \(\beta_{II}'\), has been assigned to mainly the \(3P_2 - F_3\) transition. The possible contribution of other transitions of the \(2P_{1/2} 3x - 3x3d\) \((x = s,p,d)\) array having appreciable intensities have also been discussed.

1 Introduction

For past some years the identification of the transitions in doubly and triply inner shell ionized atoms, which can be assigned to various X-ray satellites\(^1-3\), has been in progress in this laboratory. The method adopted is more reliable than Wentzel's \(Z+1\) approximation one\(^4,5\). The transition energies have been calculated using Hartree-Fock-Slater (HFS) formulas\(^6\) in intermediate coupling scheme and their relative intensities have been estimated in L-S coupling scheme. In continuation, the authors present below the transition assignments to the satellites namely, \(\beta_{I}'\) and \(\beta_{II}'\) in the L-emission spectra of the elements of first transition series.

A survey of the literature reveals that in \(L\beta_1\) spectra, a maximum of four satellites have been reported in the elements with \(Z = 26\) to 69 only, with a few gaps\(^7\). These have been named, \(\beta_{I}'\), \(\beta_{II}'\), \(\beta_{III}'\) and \(\beta_{IV}'\), in order of increasing energy. The transition assignments to all these lines have been reported by one of the authors\(^8\), in which it has been shown that these lines are emitted by the superposition of all the intense \(2P_{1/2} - 3d^n\) transitions taking place in the atom when an additional ionization in M-shell is present. In this paper, the relative transition probabilities had been calculated by considering \(2s_{1/2} - 2P_{1/2} 3x\) Coster Kronig transitions and also by considering shake off probabilities associated with a \(2P_{1/2}\) ionization, such that, a \(2P_{1/2} 3x\) state may be generated. Thus, in both these processes, the probability of formation of singly-ionized state, \(2s_{1/2}\) or \(2P_{1/2}\), plays a very
important role. In the paper mentioned above, both these probabilities were taken as equal.

Furthermore, the calculated spectrum in each element was presented in the form of bar graphs, in which the energy of the transition was represented by its placement on X-axis and its relative intensity by the height of the bar, taken parallel to Y-axis. Such calculated spectra were compared with the measured positions of Lβ₁ satellites.

The transition assignments to these satellites have, therefore, been re-investigated by adding two features in the calculations over those used earlier. The first one is calculation of the \( \frac{2s_{1/2}}{2p_{1/2}} \) and \( \frac{2p_{3/2}}{2p_{3/2}} \) ionization probabilities, needed as base states for Coster Kronig and shake off processes, respectively. Secondly, each transition has been assumed to give rise to a Gaussian line and the theoretical spectrum has been calculated as the sum of such Gaussian lines. The results for the elements \( Z = 26 \) to \( 30 \) are presented below.

2 Calculation

The authors had undertaken the studies of all those transitions of \( \frac{2p_{3/2}}{3x - 3x_2} \) array which were allowed according to selection rules \( \Delta S = 0, \pm 1, \Delta I = 0, \pm 1 \). They calculated their HFS energies in intermediate coupling and their relative probabilities in L-S coupling scheme.

2.1 Transition energies

The energies of the transitions, used in the present study, have been calculated by the combination formula:

\[
E(\frac{2p_{3/2}}{3x - 3x_2}) = E(K\alpha_t) - E(\frac{1s - 2p_{1/2}}{3x_2}) + E(\frac{2p_{3/2}}{3x})
\]

where \( E(K\alpha_t) \) is the energy of \( K\alpha_t \) line. Its values have been taken from the tables of Bearden and Burr. \( E(\frac{1s - 2p_{1/2}}{3x_2}) \) and \( E(\frac{2p_{3/2}}{3x}) \) are the Auger electron energies for the \( \frac{1s - 2p_{1/2}}{3x_2} \) and \( \frac{2p_{3/2}}{3x} \) transitions respectively. These energies have been taken from the tables of Larkins, who has calculated various two-hole state energies of atoms with \( Z = 10-100 \) in the intermediate coupling approximation and has, also, corrected them for adiabatic relaxation of the orbitals due to a sudden creation of an inner hole as well as for the solid state of the sample. His values are in good agreement with the available experimental Auger electron energy data in the region of \( Z \) values, presently under study, as claimed by him, and also as shown by one of the authors in case of \( \frac{1s - 3x_2}{2p_{3/2}} \) radiative Auger spectra of \( _{27}Co \) and \( _{28}Ni \), and \( \frac{1s - 2p_{3/2}}{3x} \) RA spectra of \( _{27}Co \) and \( _{28}Ni \).

2.2 Transition probabilities

For the emission of Lβ₁ satellites, those transitions are being considered in which the initial states are doubly ionized, one vacancy lying in \( 2p_{1/2} \) subshell and second one in any of M-subshells. Such states are formed by two processes.

(1) \( \frac{2s - 2p_{1/2}}{3x} \) Coster Kronig transitions, namely, conversion of one-hole state \( \frac{2s}{3x_2} \) to a two-hole state \( \frac{2p_{1/2}}{3x} \) through the Auger transition \( \frac{2s - 2p_{1/2}}{3x} \).

(2) Shake-off process, namely, an electron from M-subshell of the atom may escape out simultaneously to the formation of a \( 2p_{1/2} \) vacancy. This additional vacancy is created due to shaking of the atomic orbits caused by a sudden change in the potential field in the atom, taking place when a \( 2p_{1/2} \) electron leaves the atom with a fast speed.

The Coster Kronig transition probability can be written as \( \sigma \sigma' \), where \( \sigma \) denotes the probability of formation of a vacancy in \( 2s_{1/2} \) subshell of the atom and \( \sigma' \) is the probability of its decay through the CK transition \( 2s - 2p_{1/2} \). Factor \( \sigma \) has been calculated by the formulas given by Moores et al.:

\[
\sigma_{\theta} = (\pi n^2 a_0^2 Z_a/ Z_f) \sigma_a(R) \quad \text{(2)}
\]

or, say

\[
\sigma_{\theta} = (1.628 \times 10^{-14}) Z_a \sigma_a(R)/ E_a^2 \quad \text{(3)}
\]

where, \( n \) and \( / \) denote the subshell of the atom in which a hole is created, \( Z_a \) denotes the total number of electrons in this subshell and \( E_a \) denotes the binding energy of an electron in this subshell. \( \sigma_a(R) \) is known as reduced cross-section, and is calculated by the formula:

\[
\sigma_{\theta}(R) = (1/\mu)[A \ln (1-\mu)^2 + (C\mu + D\mu^2) (1-1/\mu)]
\]

... (4)
These formulas have been theoretically derived by Moores et al.\textsuperscript{14} and are applicable to single ionization of atoms in inner shell by electron bombardment. The $A, B, C$ and $D$ are constants, whose values for ionisation in $2s_{1/2}$ subshell are $A = 0.823$, $B = 3.69$, $C = 0.62$, $D = 1.79$ and in $2p_{1/2}$ subshell are $A = 0.530$, $B = 5.07$, $C = 1.20$ and $D = 2.50$ [Ref. 14]. The dimensionless parameter $u$ denotes the ratio:

$$u = \frac{E}{E_B} = \frac{\text{Incident energy of incoming electron}}{\text{B.E. of the nl electron}}.$$ 

Since different researchers have measured satellite spectra of various elements experimentally, they have also used different excitation energies. The authors have taken arbitrarily the value of $u$ as 2.5, a practical value found\textsuperscript{15} to give a measurable intensity of satellites. The same value has been used by them earlier also\textsuperscript{2,3}.

The value of $\sigma(\overline{2s})$, so calculated, has been multiplied with Coster Kronig transition probability $\sigma'$, taken from table of McGuire\textsuperscript{6}.

Coming to shake-off process, the authors have first calculated the cross-section $\sigma(2p_{1/2})$ by formulas given in Eqs (2) and (4) and have, then, multiplied it with the shake-off probability of a M-subshell electron. This probability has been calculated by interpolation from the percentage probabilities of shake-off processes occurring with a single photo-ionisation in inert gases\textsuperscript{17}.

Subsequently, the total probability of creation of an initial state $2p_{1/2}3x$ has been determined by adding both these cross-sections, calculated above. The cross-section for a set of $2p_{1/2}3x$ levels with $x$ denoting any one subshell of M-shell, so calculated, has been assumed as the total probability of all the transitions from this set. This has been distributed statistically among all the allowed transitions from this set of levels, considering first all the multiplets of supermultiplets from various $\overline{2s}^{(L)}(L)$ levels of the set and then using tables of White and Eliason for relative probabilities of the transitions of each multiplet, as presented in Ref. 6. The detailed method of this distribution has been presented in earlier papers\textsuperscript{2,3}.

2.3 Formation of the spectrum

The authors calculated energies and intensities of all the possible transitions of the $2p_{1/2}3x - 3x3d$ array as mentioned earlier. The transitions having intensity less than $1/10^9$ of the maximum intensity out of all of them have been ignored. A composite spectrum formed by spectral lines emitted by these transitions has been computed by taking each as a Gaussian line. The choice of a Gaussian shape has been favoured over the Lorentzian one as it is more suitable to satellite spectra, as discussed by Maskil and Deutsch\textsuperscript{18}. For this, the authors have taken energy on X-axis and intensity on Y-axis. The peak height of each line is taken equal to the transition probability and the peak position on X-axis is taken

![Fig. 1 — Computed $2p_{1/2}3x - 3x3d$ (x = s,p,d) spectrum of $\alpha$Fe. The measured L$\beta_1$ are shown in two rows at the top. The upper row of bars are marked at energies taken from Ref.9. In the lower row, these are marked after shifting the zero so that agreement between measured satellites and calculated peaks becomes obvious.](image-url)
at the energy of the transition. The widths of all the lines in one element have been assumed equal and its value has been decided by trial and error method in such a way that the number of peaks obtained from the calculated spectrum is at least equal to or greater than the number of satellites observed experimentally in the spectrum of the element. The calculated spectra of elements with $Z = 26$ to $30$, thus obtained, are shown in Figs 1-5. In these spectra, peaks of higher intensities have been recognised as the observed satellites. For one-to-one correspondence between peaks and measured satellites, the relative energy separations of peaks and those of measured satellites have been taken into consideration.

3 Results

Out of all the 28 transitions of $2p_{1/2}^1 3x - 3x3d$ ($x \equiv s,p,d$) array, 8 belonging to $x = d$ group, 4 to $x = p$ and 2 to $x = s$, have intensities so as to warrant attention. These are presented in Table 1, in order of

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**Fig. 2** — Computed $2p_{1/2}^1 3x - 3x3d$ spectrum of $^{57}$Co

**Fig. 3** — Computed $2p_{1/2}^1 3x - 3x3d$ spectrum of $^{59}$Ni
decreasing intensity and are named A through N. All these transitions in the elements $\text{Fe}$ to $\text{Zn}$ have energies and intensities such that, a super-position of corresponding Gaussian lines gives rise to a spectrum consisting of at least four peaks, marked 1 to 4 in Figs 1-5. A comparison of the computed spectrum of an element with the measured satellite spectrum reveals that, whereas, the measured satellite energies are close to those of intense peaks, their separations in the computed and measured spectra are in good mutual agreement. The measured spectra are also presented in Figs 1-5, in each of which the measured satellite data are shown by vertical bars at the top of the figure. This agreement in all the elements, studied presently, is also shown in Fig. 6. The transition assignments to the satellites based on the identification of the peaks is discussed below.

3.1 Transitions A, B and C

The main feature in each of the spectra, studied presently, is produced due to three most intense peaks...
Table 1 — Intense transitions of $2p_{1/2}^{3}3x - 3x3d$ (x = s,p,d) array, with their names, as used in the text. Their relative intensities are presented in col.(a). The relative intensities of strongest transition $3F_{3}-F_{3}$ in each case normalized to 100 are shown in col. (b). In col. (c) the percentage contribution of these transitions to the total intensity of this spectrum is shown.

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transitions of the array, namely, A, B and C. Amongst weaker transitions, E, N and L merge with A, while M and F overlap with B and C respectively. All these transitions jointly produce a nearly symmetric line in the spectrum of 26Fe, marked as peak No. 1 in Fig. 1. The separations in the transition energies increase with rise in Z such that, this symmetric line of the spectrum of 26Fe takes the shape of two notable structures in higher Z spectra, marked nos. 1(a) and 1(b), in Figs 2-5. The structure 1(a) is a complex shoulder to line 1(b) in case of 27Co (Fig. 2), while it is a clear resolved peak in other cases (Figs 3-5). However, in the spectrum of 29Cu, the intensity of peak 1(b) suddenly rises due to reason given below and so peak 1(a) appears like a highly pronounced shoulder to the peak 1(b). The feature 1(a) is produced due to the super-position of B, C, F and M transitions, while 1(b) arises due to that of A, E, N and L transitions. The separations in energies of 1(a) and 1(b) agree very well with the measured separations for the $\beta_{I}^{I}$ and $\beta_{I}^{II}$ satellite energies in the spectra of $^{29}$Ni, $^{28}$Cu and $^{30}$Zn, (Figs 3-5), which suggests that these two structures, namely 1(a) and 1(b) can be identified as the satellites $\beta_{I}^{I}$ and $\beta_{I}^{II}$ respectively. However, in the spectrum of 26Fe, the measured $\beta_{I}^{I}$ and $\beta_{I}^{II}$ separation is smaller than the width of the peak 1, and in case of 27Co, this separation is much larger than that between structures 1(a) and 1(b). In absence of any other measured energy data, except that reported by Cauchois & Senemaud, and also in absence of any data on intensity of these satellites, both these lines in the spectrum of 26Fe are assigned to structure No.1 (Fig. 1), while in that of 27Co, only the line $\beta_{I}^{I}$ is assigned to the peak No. 1(b) (Fig. 2).

3.2 Transition D

The transition $2p3d^{3}P_{1} - 3s3d^{3}D_{2}$ array has an intensity nearly equal to and higher than that of C in

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Table 2 — Calculated energy and height of peak No. 1(a) and of the transitions B, C and F which give rise to this peak. The corresponding values of measured $\beta_{I}^{I}$ energy are also shown.

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Table 3 — Calculated energy and height of transitions A, D, E and L and of corresponding peak no. 1(b). The satellites $\beta_{I}^{I}$ to which this peak is identified are also shown.

$\beta_{I}^{I}$ & $\beta_{I}^{II}$ energies (eV)
$^{56}$Fe, but gradually reduces with rise in $Z$, so that in $^{58}$Ni to $^{60}$Zn, it becomes weaker than $C$. It produces well resolved and intense peak marked No.2, on higher energy side of the main peak (Figs 1-5). This transition gradually shifts towards $A$, as $Z$ increases. It forms a shoulder on high energy tail of peak No.2 in $^{58}$Ni (Fig. 3), and in the spectrum of $^{69}$Cu it combines with $A$ so that height of peak No.1(b) increases prominently (Fig. 4). In $^{60}$Zn, it lies on lower energy side of $A$ and cannot produce a notable effect. However, no line has been observed corresponding to this transition in the spectra studied presently.

3.3 Transitions G and K

These weak transitions are mutually close in energy and produce a small but notable feature, marked No.3 in Figs 1-5. Except in the spectrum of $^{57}$Co, no line has been found to be associated with these transitions. In case of $^{57}$Co, the line $\beta_{11}^{n}$ can be identified with this peak. This assignment is only very tentative because the peak No.2 in this very spectrum (Fig. 2) is much intense than this peak.

3.4 Transition I

The second most intense transition of $2p^{3}p - 3p3d$ array is the $^{3}D_{2}-^{3}D_{1}$ one and has the highest energy of all fourteen transitions being considered in the present study. It gives rise to a well-separated but a small peak in each of the spectra, marked No. 4 (Figs 1-5). No line has been measured so far corresponding to this peak, in these spectra.

4 Conclusion

The present studies have revealed that, both the satellites $\beta_{1}^{1}$ and $\beta_{11}^{n}$, observed in L$\beta$, spectra of elements in the range $Z = 26-30$, with a few exceptions, arise mainly due to $2p_{1/2} - 3d$ transitions in presence of a M-shell spectator vacancy. On the basis of agreement between computed spectra and measured satellites, it is observed that the satellite $\beta_{1}^{1}$ in the spectra of $^{56}$Fe, $^{58}$Ni, $^{69}$Cu and $^{60}$Zn is emitted by the super-position of three intense transitions, $^{3}F_{2}-^{3}F_{2}$, $^{3}P_{2}-^{3}P_{2}$ and $^{3}P_{1}$, in order of decreasing contribution of intensity in each of these spectra. It has been well established that the transition $^{3}F_{2}-^{3}F_{2}$ is the main source of the emission of the satellite $\beta_{11}^{n}$ in the elements $^{56}$Fe and $^{58}$Ni to $^{60}$Zn. In the spectrum of $^{60}$Co, $\beta_{11}^{n}$ is assigned to this transition, while $\beta_{11}^{n}$ is tentatively assigned to the super-position of two, much weaker transitions $^{3}P_{2}-^{3}D_{1}$ and $^{3}D_{1}F_{2}$.

References