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Measurement of L X-ray fluorescence cross-sections for elements with $45 \leqslant Z \leqslant 50$ using synchrotron radiation at 8 keV

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ABSTRACT

The L shell fluorescence cross-sections of the elements in range $45 \leqslant Z \leqslant 50$ have been determined at 8 keV using Synchrotron radiation. The individual L X-ray photons, Ll, $L\alpha$, $L\beta_1$, $L\beta_{II}$, $L\gamma_1$ and $L\gamma_{II}$ produced in the target were measured with high resolution $Si(Li)$ detector. The experimental set-up provided a low background by using linearly polarized monoenergetic photon beam, improving the signal-to-noise ratio. The experimental cross-sections obtained in this work were compared with available experimental data from Scofield [1,2] Krause [3,4] and Scofield and Puri et al. [5,6].

These experimental values closely agree with the theoretical values calculated using Scofield and Krause data, except for the case of $L\gamma$, where values measured of this work are slighter higher.

1. Introduction

Accurate values of L X-ray fluorescence cross-sections are needed not only in basic research such as atomic, molecular and radiation physics but also in applied research such as non-destructive elemental analysis and dose calculation. These cross-section values are also required to test the models used in predicting fluorescence data. In view of this, numerous authors [7–14] have reported experimental and theoretical L X-ray fluorescence cross-sections for an extensive range of elements and excitation energies, but there are still several elements and energy ranges that have not been investigated. Besides, from the literature we notice that there are some discrepancies among the measured values by different authors, especially when different experimental set-ups were used.

Most of the measurements were done using an X-ray tube with a secondary target that provides a quasi monoenergetic excitation beam. In these cases, the scattering radiation of the primary beam is a component of the excitation beam, producing an extra contribution to the X-ray spectra that has not been considered in the published data. Additionally, the results are only reported considering the L lines in groups referred to as Ll, $L\alpha$, $L\beta$ and $L\gamma$.

Discrepancies between the theoretical cross-sections calculated using different Coster-Kronig coefficients and fluorescence yields like Krause's [3,4], Chen's [15,16] and Puri et al.'s [5,6] tables, are found. Krause's tabulations are compilations of semi-empirical fitted values while Chen's and Puri et al.'s are theoretical values based on *ab initio* relativistic Dirac-Hartree-Slater calculations.

The aim of the present work was to measure L X-ray cross-sections with monoenergetic excitation beams at 8 keV for elements with atomic number in the range of $45 \leqslant Z \leqslant 50$.

The experimental determinations of L X-rays fluorescence cross-sections at 8 keV were carried out using a $Si(Li)$ solid-state detector which could resolve individual components of the L X-ray photons emitted from the elements used in the present experiment. The experimental cross-sections were grouped considering the transitions scheme, the energy of the emission lines and the detector's resolution. The cross-section is reported for each spectral line or group of lines, depending on the resolution of the detector used.

In the present experiment, the geometry factor and efficiency of the detector were determined measuring the emission of K-lines of Ca, Cl, Ti and Fe. The measured values were compared with the values obtained by Monte Carlo simulations using the package of subroutines named PENELOPE-2008 [17].

In general, the cross-section values obtained experimentally in this work are comparable with the calculated values using Scofield [1,2] and Krause [3,4], except for the $L\gamma$ lines. In particular, the Cd $L\gamma$I experimental value is about 50% smaller than the calculated values.

The $L\beta$ cross-sections show a regular trend in Z, the experimental results are similar compared with both Krause's [3,4] and Puri

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et al.'s [5,6] values. But in the case of the atomic number $Z = 50$ the experimental datum of $L\beta$I fluorescence cross-section is lower than the theoretical Puri et al.'s and the semi empirical Krause's data as well. The $L\beta$II cross-section experimental datum is much higher than the calculated values from Puri et al.'s and Krause's coefficients.

2. Experimental set-up

The experimental measurements were carried out at the X-ray Fluorescence beam line at the National Synchrotron Light Laboratory (LNLS), Campinas, Brazil [18]. The most important components of the experimental set-up were the following:

- Silicon (1 1 1) channel-cut double crystal monochromator, which can tune energies between 3 and 30 keV. The energy resolution is $3–4 \times 10^{-4}$ between 7 and 10 keV.
- A Si(Li) solid state detector, 5 mm thick and 5 mm in diameter, with a resolution of 170 eV at 5.9 keV and a 0.0127 cm thick beryllium window. The detector's efficiency was obtained using the model proposed by Jaklevic and Giauque [19].
- The whole set-up is mounted on a motorized lift table, which allows the vertical positioning of the instruments within the linearly polarized part of the beam.
- A motorized computer-controlled set of vertical and horizontal slits to limit the beam size, upstream and downstream the monochromator.

Foil samples of rhodium, palladium, silver, cadmium, indium and tin were used for the determination of the L fluorescence cross-sections. The foil samples were provided by Alfa products Inc., with a certified purity of over 99%. Foils thicknesses are shown in Table 1.

Chlorine, calcium, titanium and iron K emission lines were measured to determine the geometrical and detector efficiency factors. In order to check these measurements, these values were compared with the simulated efficiency obtained by Monte Carlo calculations using the PENLEOPE package subroutines [17].

The measurements of the fluorescent spectra were carried out by collecting 2×10^5 net counts for the $K\alpha$ or $L\alpha$ peaks for each element in order to have the same statistical error due to count rate in all the measured spectra. Vertical and horizontal opening of the slit at the exit of monochromator was adjusted to obtain a system dead time lower than 1%, by measuring the fluorescence emission of a Ti sample. All the samples were measured with the same slit aperture. This configuration allowed us to avoid unwanted effects such as piling up, and ensured that the geometric factors were the same for all the samples. In this way, it was not necessary to carry out corrections for count losses, spectra distortions or modification of the geometrical arrangement.

Table 1
Thicknesses of the sample foils used in the measurements.

Element	Thickness [cm]
Rh	0.0127
Pd	0.010
Ag	0.0127
Cd	0.0125
In	0.0127
Sn	0.0127
Cl (NaCl)	0.3
Ca ($CaHPO_4{\cdot}2H_2O$)	0.3
Ti (Ti foil)	0.01
Fe (Fe foil)	0.1

3. Spectra analysis

The L X-ray fluorescence lines were grouped considering the energy of the emission lines tabulated by Scofield [1,2] and the detector's resolution. This line arrangement was used for the fitting of the L spectrum, where the $L\beta$ and $L\gamma$ compound lines have been noted with a Roman subscript according to the most intense contribution line, with its corresponding atomic transition:

- $L_l = L_3 - M_{1,}$
- $L\alpha = L_3 - M_5 + L_3 - M_4$,
- $L\beta_I = L_2 - M_4 + L_1 - M_2 + L_1 - M_3 + L_3 - N_1$,
- $L\beta_{II} = L_3 - N_5 + L_3 - O_4 + L_3 - O_5 + L_3 - O_1 + L_1 - M_5 + L_1 - M_4 + L_3 - N_4$,
- $L\gamma_I = L_2 - N_4$
- $L\gamma_{II} = L_1 - N_2 + L_1 - N_3 + L_1 - O_2 + L_1 - O_{3.}$

A linear polynomial function of second order was used to fit the background radiation. The fluorescence peaks were fitted using four different approaches.

1. All peaks fitted using a Gauss shape function.
2. $L\alpha$ lines fitted using a modified Hypermet function type [20]. The rest of the peaks were fitted using Gaussian function.
3. $L\alpha$ and $L\beta_I$ lines fitted using a modified Hypermet function type. The rest of the peaks were fitted using the Gaussian function.
4. All peaks were fitted using a modified Hypermet function type.

The modified Hypermet function type [20] is the sum of a main Gaussian like function plus a low-energy tail term. The tail contribution comes from three individual components: an exponential term, a continuous step and a truncated step.

The escape peaks were fitted using a Gaussian function independently of the approach used for the other peaks.

Using these approaches four intensity values for each peak were obtained. The intensity average and the standard deviation were calculated from these four values and the standard deviation is used as intensity uncertainty.

The very low background contribution to the measured spectra is a consequence of the excitation with a linearly polarized beam of 8 keV photons. The linear polarization of the incident beam produces negligible scattered radiation at 90° with respect to the incident beam direction. The detector position is localized at the same height of the storage ring.

Measured spectrum of Cd L lines excited by 8 keV photons and corresponding fitted curve using the modified Hypermet function type (approach #4) is shown in Fig. 2.

4. Analysis of data

The L experimental fluorescence cross-sections were determined from the following expression [9]:

$$\sigma^e_{Li}(E_0) = \frac{I_{Li}}{I_0 \cdot G \cdot \varepsilon(E_{Li}) \cdot T(E_0, E_{Li})}$$

where $\sigma\ eLi(E_0)$ = experimental Li fluorescence cross-sections of the element observed at the energy E_0, with $Li = Ll, L\alpha, L\beta$I, $L\beta$II, $L\gamma_1$ or $L\gamma_{II}$, I_{Li} = measured intensity of the Li spectral line, $I_0 G\varepsilon(E_{Li})$ = factor comprising the intensity of the excitation beam I_0, the geometry of the experimental arrangement G and the detector's efficiency $\varepsilon(E_{Li})$, E_0 = energy of the incident beam, in this case 8 keV, E_{Li} = energy of the Li spectral line; the data was taken from Scofield [1,2], and $T(E_0, E_{Li})$ = correction factor for self-absorption in a infinitely thick sample.