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Measurement of the radiative vacancy transfer probabilities from the L_3 to M and to N shells for W, Re and Pb using synchrotron radiation

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Abstract

The radiative vacancy transfer probabilities from L₃ to M shell, $\eta_{L_3M}(R)$ and L₃ to N shell, $\eta_{L_3N}(R)$, have been determined for W, Re and Pb. The pure elements samples were excited by monochromatic synchrotron radiation. The X-rays were generated by excitation of L₃ edge and measured using a high resolution Si(Li) detector. The experimentally determined radiative vacancy transfer probabilities were compared with the theoretical values deduced using radiative X-ray emission rates based on the relativistic Dirac–Hartree–Slater (RDHS) model. In the case of Pb, the experimental data were compared as well with experimental values of Simsek. In both cases, a good agreement was found between the datasets.

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1. Introduction

The innershell vacancy produced in an atom is filled through radiative and non-radiative transitions. In the radiative transitions, the X-ray photons is emitted, but in the non-radiative transitions, the Auger electrons is emitted. In these processes, the vacancy moves from innershell to higher shells. The vacancy transfer can also take place within the subshell and such transitions is known as Coster–Kronig transitions.

Knowledge of vacancy transfer is important in understanding several atomic processes such as internal conversion of gamma rays, nuclear electrons capture, nuclear electron capture, characteristic X-ray productions and photoelectric effects among others.

Several investigators have already measured K-to-L and L-to-M vacancy transfer probabilities by exciting the target with radioactive sources. Ertugrul et al. [1] have measured

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the radiative vacancy transfer probabilities from the K to the L₂, L₃ subshell and to the M shell in the atomic regions $69 \le Z \le 92$. Recently, Simsek [2] has measured the radiative vacancy transfer probabilities from L₃ to M and N shells for Pb, Th and U, using radioactive sources.

Rao et al. [3] have obtained the theoretical values of the vacancy transfer probabilities from L_i to M, η_{L_iM} (i =1,2,3) for the elements in the atomic region $26 \le Z \le 92$. Mc Guire [4] has also reported the values of vacancy transfer probabilities from L_i to M for elements in the range $50 \le Z \le 90$. Puri et al. [5] have evaluated the probabilities η_{KL} , η_{L_iM} and η_{KM} for elements in the region $18 \le Z \le 96$.

With the best knowledge of the author, the experimental radiative vacancy transfer probabilities from L_3 to M and N shell are not available in literature for W and Re elements, in view of this, these parameters were measured for the elements W, Re and Pb. Measured values are compared with theoretical values deduced using radiative X-ray emission rates [6] based on the relativistic Dirac–Hartree–Slater (RDHS) model. The experimental values of probabilities for Pb were compared with theoretical and experimentally

determined values of Simsek [2] and remarkably agreement was found between these two datasets.

2. Experimental method

The experimental determinations were carried out at the X-ray fluorescence beam line at the National Synchrotron Light Laboratory (LNLS), Campinas, Brazil [7].

The main components of the experimental set-up were the following:

- Silicon (111) 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 Delgado et al. [8].
- The whole set-up is mounted on a motorized lift table, which allows the vertical positioning of instruments within the linearly polarized part of the beam.
- A motorized computer-controlled set of vertical and horizontal slits to limit the beam size, before and after the monochromator.

The sample was placed perpendicular to the plane generated by the directions of the primary beam and the fluorescence X-ray emission and it is normal at 45° in relation to the incident beam and to the detector axis. The experimental set-up is shown in the Fig. 1.

Tungsten, rhenium and lead foils were used for the measurement of the probabilities of radiative vacancy transfer. The foil samples were provided by Alfa Products Inc., with a certified purity better than 99%, their thickness are listed in Table 1. Cobalt, nickel, copper, zinc, germanium and arsenic K emission lines were evaluated to determine the

 Table 1

 Target thickness used in the present measurements

Element	Thickness (cm)
Rh	0.0127
Pd	0.010
Ag	0.0127
Cd	0.0125
In	0.0127
Sn	0.0127
Ti	0.01
W	0.05
Re	0.05
Pb	0.05

 $I_0G\varepsilon(E)$ factor, comprising the intensity of the exciting beam, the geometric factor and the detector's efficiency. The excitation incident energy were 11.0 keV to tungsten and rhenium and 13.2 keV to lead case. The $I_0G\varepsilon(E)$ factor was obtained separately for each case.

The measurements of the fluorescent spectra were carried out taking as a counting limit an intensity of 2×10^5 net counts for the $K\alpha(K-L_2 + K-L_3)$ or $L\alpha(L_3-M_5 + L_3-M_4)$ lines of the element observed in order to have the same statistical counting error in all of the spectra recorded. The aperture of the post monochromator vertical and horizontal slits was adjusted to obtain a system dead time lower than 1%, by measuring the fluorescence emission of a Cu sample. All the samples were measured with the same slit aperture. This configuration allowed 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 make corrections for count losses, spectrum distortion or modification of the geometrical arrangement.

2.1. Spectra analysis

The peaks were fitted with a Gaussian function using a cubic polynomial function to fit the background radiation.



Fig. 1. The experimental set-up.

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