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Measurement of *L* subshell fluorescence yields for high-*Z* elements excited by 22.6 keV photons

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ABSTRACT

The σ_{L1}, σ_{L2} and σ_{L3} subshell X-ray production cross sections for U, Th, Bi, Pb, Tl, Hg, Au, Pt, Os, W, Ta, Lu, Yb and Er have been measured at 22.6 keV incident photon energy. The measurements were performed using a ^{109}Cd radioisotope as the photon source and a Si(Li) detector. L_i subshell fluorescence yields ω_1, ω_2 and ω_3 have been determined from the measured σ_{L1}, σ_{L2} and σ_{L3} subshell X-ray production cross sections. The measured L_i subshell production cross sections and ω_i fluorescence yields were compared with theoretical, the semi-empirical and recommended values.

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1. Introduction

X-ray fluorescence (XRF) spectrometry is used worldwide. The most established technique is energy dispersive X-ray fluorescence (EDXRF) for quantitative analysis because EDXRF is relatively inexpensive and requires less technical effort to run the system. This method is very useful for the determination of fundamental parameters such as production cross sections, fluorescence yields, intensity ratios and vacancy transfer probabilities. Accurate values of these parameters are required in several fields such as atomic, molecular and radiation physics, material science, environmental science, agriculture, forensic science, dosimetric computations for health physics, cancer therapy, elemental analysis, basic studies of nuclear physics, etc.

The calculations of L_i subshell X-ray emission rates for single vacancy states based on two different independent particle approximation models are available in the literatures [1–3]. The first set is based on the Dirac–Hartree–Slater (DHS) model [1] and the second one is based on the Dirac–Fock (DF) model [2,3]. In the former model, the potential is assumed to be equal for the initial and final states of the atom undergoing transitions. In the latter model, the potential is assumed to be different for initial and final states and hence exchange and overlap effects were included.

The DHS model based emission rates were tabulated by Scofield [1] for all elements with $5 \leq Z \leq 104$ and included the most intense dipole transitions as well as less intense transitions of E2, E3 and M1 multi-polarities. The DF model based values were tabulated by Scofield [2] for 21 elements in the range $18 \leq Z \leq 94$ and included dipole transitions only. Later on, Campbell and Wang [3] reported a complete set of L_i ($i=1$–3) subshell X-ray emission rates for all the elements with $18 \leq Z \leq 94$, interpolated from the DF model based values tabulated by Scofield [2]. Recently, Kumar et al. [4] have reported the fitted values of these interpolated emission rates for different transitions as a function of atomic number. It has been

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shown experimentally [5,6] that, in case of heavy elements, the *L* X-ray emission rates based on the DF model [2,3] are more reliable than those based on the DHS model [1].

Three sets of the L_i (i=1, 2, 3) subshell fluorescence yields ω_i and Coster–Kronig yields f_{ij}(i ,j=1, 2, 3) are available in literature. In the first set, Puri et al. [7] presented theoretical values of the L_i subshell fluorescence ω_i and Coster–Kronig yields f_{ij} for all the elements with $25 \leq Z \leq 96$ using the radiative transition rates based on the relativistic Dirac–Hartree–Slater (DHS) model and the nonradiative transition rates interpolated from the DHS model based data given by [8,9]. The second set of these parameters included the semi-empirically fitted values of L_i (subshell X-ray fluorescence yields ω_i, Auger transition yields a_i and Coster–Kronig transition probabilities f_{ij} for all elements in the atomic number range $12 \leq Z \leq 110$), were tabulated by Krause [10]. Finally, Campbell [11] provided a set of recommended values of the ω_i and f_{ij} yields based on experimental data available until 2003 for the elements with $62 \leq Z \leq 96$ and recently reported [12] a revised set of recommended values for only the L_1 subshell fluorescence and Coster–Kronig yields, with uncertainties 15–30%, for all elements with $64 \leq Z \leq 92$ except for Z=75 and 76.

Experimental *L* X-ray fluorescence cross sections and yields of some elements have been measured by different researchers at various energies [13–30]. Garg et al. [13] measured *L* X-ray fluorescence cross sections and yields for elements in the atomic range $41 \leq Z \leq 52$ at 5.96 keV. L_i (i=l, α, β and γ). X-ray cross-sections have been measured for the elements Au and Pb at excitation energies 17.78, 25.78, 26.88 and 32.89 keV using an X-ray tube with a secondary exciter system as the excitation source [14]. Ozdemir et al. [15] measured L-subshell fluorescence yields (ω_1, ω_2 and ω_3) for elements in the atomic range $55 \leq Z \leq 68$ with a Si(Li) detector at 59.54 keV incident photon energy using a fluorescence excitation method. Ozdemir et al. [16] measured *L*-subshell fluorescence yield ratios (ω_1/ω_2, ω_1/ω_3 and ω_2/ω_3) in the elements from Cs to U following ionization for 59.54 keV γ-rays from an 241 A m radioactive source. Bastug et al. [17] measured the cross-section for the production of X-rays in each of *Ll, Lα, Lβ and Lγ* groups in four different elements Hg, Tl, Pb and Bi at eight energies of 16.90, 22.58, 25.77, 32.89, 38.18, 43.95, 50.21 and 59.54 keV photons. Total *L*-shell X-ray production cross sections induced by protons with energies between 400 and 700 keV were measured for elements with atomic number *Z* between 34 and 53 [18]. Singh et al. [19] have reported the L_i subshell X-ray production (XRP) cross-sections and fluorescence yields for $_{77}$Ir, $_{78}$Pt, $_{82}$Pb and $_{83}$Bi following direct ionization in the L_i subshells by the 59.54 keV γ-rays and the L_3 subshell by the Br/Rb/Sr/Y *K* X-rays. Puri and Singh [20] determined L_i subshell fluorescence yields (ω_i) and the L_1–L_3 Coster–Kronig transition probabilities (f_{13}) for elements with $50 \leq Z \leq 92$. Ertugrul [21] measured the L_{3l}, $L_{3\alpha}$, $L_{3\beta}$, $L_{2\beta}$, $L_{2\gamma}$, $L_{1\beta}$ and $L_{1\gamma}$ X-ray production cross-sections in Hg and Au. Han et al. [22] have measured *L* X-ray production cross-sections, *L* subshell fluorescence yields and *K* to *L* vacancy transfer probabilities for. Sm, Eu, Gd, Dy, Ho, Er, Pt, Au, Tl, Pb and Bi at 59.54 keV incident photon energy. Chauhan et al. [23] reported the L_1 and L_2 subshell fluorescence yields for $_{56}$Ba, $_{57}$La, $_{60}$Nd, $_{62}$Sm, $_{63}$Eu and $_{68}$Er elements deduced from the *L* X-ray spectra induced by 22.6 keV incident photons emitted from an annular radioisotope of ^{109}Cd. *L* subshell fluorescence yields ($\omega_{1,}$ ω_2 and ω_3) for the elements Re, Os, Ir, Pt, Au, Hg, Tl, Pb, Bi, Th and U at the 123.6 keV γ-ray emission excitation energy from a ^{57}Co annular radioactive source (925 MBq) using a Si(Li)detector have been measured by Sogut et al. [24]. Recently, Kumar and Puri [25] investigated the L_1 and L_2 subshell fluorescence yields for elements with $64 \leq Z \leq 70$ from the L_k (k=l,α,$\beta_{1,4}$,$\beta_{3,6}$,$\beta_{2,15,9,10,7}$,$\gamma_{1,5}$ and $\gamma_{2,3,4}$) X-ray production cross sections measured at 22.6 keV incident photon energy using a spectrometer involving a disc type radioisotope of ^{109}Cd as a photon source and a Peltier cooled X-ray detector. Han et al. [26] measured *L*α, *L*β and total *L* X-ray fluorescence (XRF) cross-sections for the nine elements (Zr, Nb, Mo, Ag, Cd, In, Sn, Sb and I) using photon energy 5.96 keV. Erdogan et al. [27] calculated the *L* shell X-ray production cross sections for the elements atomic number between $40 \leq Z \leq 92$ at the energy 1–1500 keV. Bonzi and Badiger [28] measured the *L* subshell fluorescence yields ω_1, ω_2 and ω_3 in the range $45 \leq Z \leq 50$ by exciting the elemental targets with a synchrotron radiation beam at 7 keV. Badiger and Bonzi [29] measured *L* subshell fluorescence yields ω_1, ω_2 and ω_3 for Ba, La and Pr elemental targets. The characteristic *L* X-ray photons, induced in the targets by synchrotron radiation, were measured with a Si(Li) detector coupled to multichannel analyzer. Durak and Ozdemir [30] derived *L*-subshell X-ray fluorescence yields ω_1,ω_2 and ω_3 for selected heavy elements with $70 \leq Z \leq 92$ at 59, 54 keV using the experimental *L* X-ray production cross-sections, theoretical *L* subshell photoionization cross-sections, Coster–Kronig transition probabilities and fractional X-ray emission rates.

In the present work, L_1, L_2 and L_3 subshell production cross sections (σ_{L1}, σ_{L2} and σ_{L3}) and fluorescence yields (ω_1, ω_2 and ω_3) have been investigated for the elements in the atomic region $68 \leq Z \leq 92$ at 22.6 keV. The results have been compared with theoretical values based DHS [7], the semi-empirical [10] and recommended [11,12]. To the best of our knowledge, the experimental values of σ_{L1}, σ_{L2} and σ_{L3} production cross sections and $\omega_{1,}$ ω_2 and ω_3 fluorescence yields for present elements at 22.6 keV (except Er and Yb) are reported firstly.

2. Experimental arrangement

The experimental arrangement is shown in Fig. 1. The experiments were carried out using a filtered point source of ^{109}Cd emitting monoenergetic (22.6 keV) γ-rays. Spectroscopically pure targets of thickness ranging from 18 mg/cm^2 to 36 mg/cm^2 were used. The direct beam from the source was directly incoming on the sample. The samples were placed at a 45° angle with respect to the beam from the source and fluorescent X-rays emitted in a direction perpendicular to (90°) the source were detected by a Si(Li) detector (full-width at half-maximum (FWHM)=160 eV at 5.9 keV, active diameter=3.91 mm, active area=12 mm^2,

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