Measurement of probabilities for vacancy transfer from the K to L shell of the elements $73 \leqslant Z \leqslant 92$

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K- to L-shell vacancy transfer probabilities were measured for ten elements in the atomic range $73 \leqslant Z \leqslant 92$. Those measuring L x-ray yields from targets excited by 59.5 and 122 keV incident photons, i.e., below and above the K edge of elements, were detected with a high-resolution Si(Li) detector. For comparison with experimental results, theoretical calculations were made by using available data on radiative and radiationless transitions. The radiative transitions of these elements were observed from the relativistic Hartree-Slater model which was proposed by Scofield [At. Data Nucl. Data Tables **14**, 121 (1974)]. The radiationless transitions were observed from the Dirac-Hartree-Slater model which was proposed by Chen, Craseman, and Mark [At. Data Nucl. Data Tables **24**, 13 (1979)]. The measured results were found to be in good agreement with theoretically calculated values. The experimental and theoretical values were fitted versus atomic number Z. [S1050-2947(96)05712-5]

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INTRODUCTION

The accurate determination of characteristic x-ray fluorescence cross sections, relative intensities, K, L, M and other shell fluorescence yields for elements are important because of their widespread use in the fields of atomic, molecular, and radiation physics and in nondestructive elemental analysis of materials, radiational photon sources of synchrotron radiation. The L x-ray production cross sections were measured experimentally [3–6]. Ertuğrul, Doğan, and Şimşek [7] measured K- to L-shell radiative vacancy transfer probabilities for elements in the atomic range $72 \leqslant Z \leqslant 92$. The vacancy transfers of inner shells by Coster-Kronig were measured for L x rays in heavy elements by Ertuğrul [8]. Ertuğrul [9] measured $K\alpha$ to $L\alpha$ intensity ratios and enhancement factors of vacancy transfer for lanthanides. Puri *et al.* [10,11] have measured K- to L-shell and L- to M-shell vacancy transfer probabilities for elements in the atomic range $37 \leqslant Z \leqslant 42$ and $18 \leqslant Z \leqslant 96$, respectively. In addition, Puri *et al.* [11] have calculated values and fitted them versus atomic number Z values and fitted coefficients for vacancy transfer probabilities of K to L_1, K to L_2, K to L_3, K to L (average), L_1 to M, L_2 to M, L_3 to M shells. The values of the η_{KL_i} $(i=1,2,3)$ for elements in the atomic range $20 \leqslant Z \leqslant 94$ have been calculated by Rao, Chen, and Craseman [12]. In these calculations, the contributions due to Auger and radiative transitions were derived using the best-fitted experimental data on the fluorescence yields and the intensity ratios of different components of KLX $(X=L,M,N...)$ Auger electrons and K x rays currently available. The data regarding K x-rays used in these evaluations were reasonably reliable and in agreement with theoretically calculated values.

The method of Puri *et al.* [10] is based on the number of L x rays produced at the photon excitation energy below the K edge and at the excitation energy above the K edge, where the major contribution (80–85%) to L-shell vacancies comes from the decay of K-shell vacancies. In their experiment, ^{55}Fe and ^{109}Cd (photon energy 22.6 keV) annular photon sources were used for excitation below and above the K edge, respectively. In this study, we measured K- to L-shell vacancy transfer probabilities for the atomic range $73 \leqslant Z \leqslant 92$. The ^{241}Am (energy 59.5 keV) and ^{57}Co (energy 122 keV) low-energy annular radioisotope photon sources were used for excitation below the K edge and above the K edge, respectively. The measured values have been compared with the semiempirical values tabulated by Rao, Chen, and Craseman [12] and the theoretical values deduced using Auger transition rates [2] and radiative x-ray emission [1] based on the relativistic Dirac-Hartree-Slater (RDHS) model.

EXPERIMENTAL PROCEDURE

The experimental arrangement for the annular source geometry in the direct excitation mode used in this study is shown in Fig. 1. In this arrangement low-energy photon sources of ^{241}Am (100 mCi) and ^{57}Co (100 mCi) were used. The energies of the primary photons are 59.5 and 122 keV for ^{241}Am and ^{57}Co, respectively. Spectroscopically pure targets of Ta (99%), W (99%), Au (99%), $Hg(NO_3)_2$ (99%), Tl_2O_3 (99%), PbO (99.95%), Bi_2O_3 (99.98%), $Th(NO_3)_2$ (99%), and $(CH_3COO)_2$ $UO_2 2H_2O$ (99%) of thickness ranging from 25.84 to 51.68 mg/cm^2, have been used for measurements. The L x-ray spectra from various targets were recorded using a Si(Li) detector [full width at half maximum (FWHM)=160 eV at 5.96 keV, active area=12.5 mm^2, sensitive dept=3.5 cm] coupled to a Nuclear Data multichannel analyzer (MCA) system (ND 66 B). Each target was excited and recorded for the time intervals ranging from 3 to 12 h. A typical L x-ray spectrum of $Hg(NO_3)_2$ at 59.5 keV excitation energy is shown in Fig. 2.

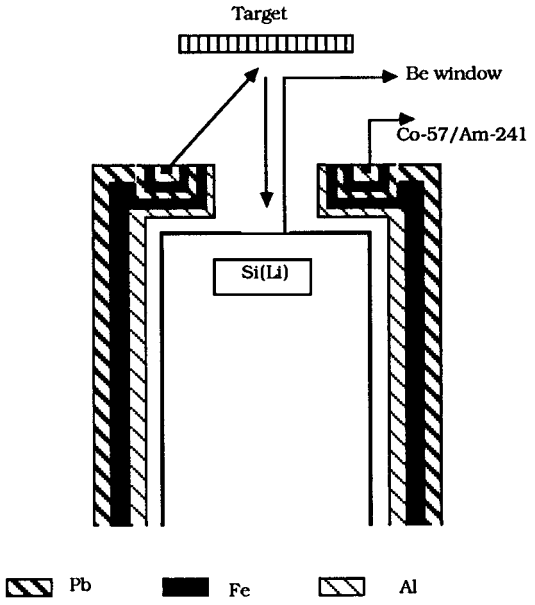

FIG. 1. Experimental arrangement.

DATA ANALYSIS

The L-shell x-ray production cross sections $\sigma_{Lx}^{(i)}$ $(i=1,2)$ at excitation energies below and above the K edge of elements are given by Eqs. (1) and (2), respectively,

$$\sigma_{Lx}^{(x)}=\sigma_L^{(1)}\overline{\omega_L}, \tag{1}$$

$$\sigma_{Lx}^{(2)}=(\sigma_L^{(2)}+\sigma_k^{(2)}\eta_{KL})\overline{\omega_L}, \tag{2}$$

where $\sigma_K^{(i)}$ and $\sigma_L^{(i)}$ are the total K- and L-shell ionization cross sections, respectively. The numbers (1) and (2) correspond to excitation energies 59.5 and 122 keV, respectively, i.e., below and above the K edge of the elements. $\overline{\omega_L}$ is the value of the L-shell fluorescence yield that does not differ significantly for moderately different distributions of initial L_i subshell vacancies produced by excitation energies below and above the K edge of the elements.

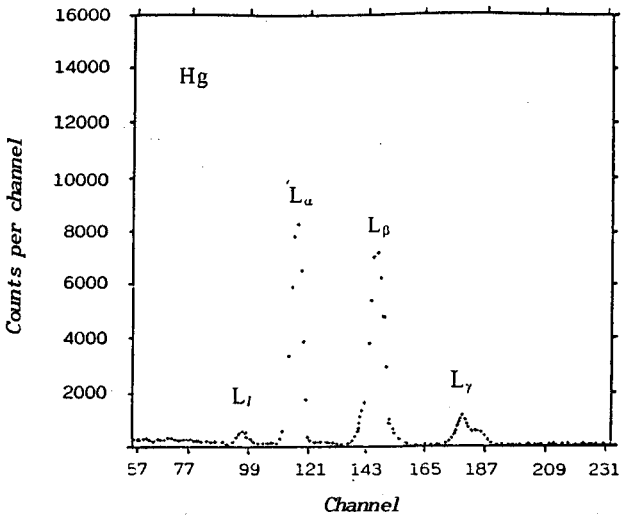

FIG. 2. L x-ray spectrum of Hg recorded by a Si(Li) detector.

The experimental L x-ray production cross sections, $\sigma_{Lx}^{(i)}$ were evaluated using the relation

$$\sigma_L^x=\frac{N_{Lx}^{(i)}}{I_0G\varepsilon_i\beta_i m}. \tag{3}$$

Using equalities (1)–(3), the experimental K- to L-shell vacancy transfer probability η_{KL} can be expressed as

$$\eta_{KL}=\frac{1}{\sigma_K^{(2)}}\left[\frac{(I_0G)^1}{(I_0G)^2}\frac{\beta_L^{(1)}}{\beta_L^{(2)}}\sigma_L^{(1)}-\sigma_L^{(2)}\right], \tag{4}$$

where N_i is the number of counts per unit time under the photopeak corresponding to the L x ray of the element, $(I_0G)_i$ is the intensity of the exciting radiation falling on the area of the target sample visible to the detector, ε is the efficiency of the detector at the average L x-ray energy of the element, and m is the mass per unit area of the element in the target. The β_L is the self-absorption correction factor for the incident photons and emitted L x-ray photons. The values of β_L have been calculated by using the following expression obtained by assuming that the fluorescence x rays are incident normally on the detector:

$$\beta_L=\frac{1-\exp[(-1)(\mu_{\text{inc}}\sec\theta_1+\mu_{\text{emt}}\sec\theta_2)]t}{(\mu_{\text{inc}}\sec\theta_1+\mu_{\text{emt}}\sec\theta_2)t}, \tag{5}$$

where μ_{inc} and μ_{emt} are the absorption coefficients at the incident and emitted x-ray photon energy. The values are taken from the tables of Hubbell and Seltzer [13]. In their tables, the μ/ρ values are taken from the current photon interaction database at the National Institute of Standards and Technology. t is the thickness of the target in g/cm^2, and θ_1 and θ_2 are the angles of incident photons and emitted x rays with respect to the normal at the surface of the sample.

However, in the present work, the value of the factor $I_0G\varepsilon_i$, which contains terms related to the incident photon flux, geometrical factor, and the absolute efficiency of the x-ray detector, was determined by collecting the K x-ray spectra of thin samples of Fe (99.9%), ZnO_2 (99.9%), As_2O_3 (99.9%), SeO_2 (99%), $SrCl_2\cdot 6H_2O$ (99%), ZrO_2 (99.9%), Mo (99.9%), and Pd_2Cl (99%) in the same geometry in which the L XRF cross sections were measured and using the equation

$$I_0G\varepsilon_{k\alpha}=\frac{N_{K\alpha}}{\beta_{K\alpha}m\sigma_{K\alpha}}, \tag{6}$$

where $N_{K\alpha}$, $\beta_{K\alpha}$, and $\varepsilon_{K\alpha}$ have the same meaning as in Eq. (3) except that they correspond to K x rays instead of the ith group of L x rays. In these calculations, the theoretical values of the K x-ray fluorescence cross sections $(\sigma_{K\alpha})$ were calculated using the equation

$$\sigma_{K\alpha}=\sigma_K(E)\omega_K f_{K\alpha}, \tag{7}$$

where $\sigma_K(E)$ is the K-shell photoionization cross sections [14] for the ten elements at the excitation energy E, ω_K is the K-shell fluorescence yield from the tables of Hubbell *et al.* [15], and $f_{K\alpha}$ [16] is the fractional x-ray emission rate for $K\alpha$ x rays and is defined as

$$f_{K\alpha}=\left(1+\frac{I_{K\beta}}{I_{K\alpha}}\right)^{-1}, \tag{8}$$

where $I_{K\beta}/I_{K\alpha}$ is the $K\beta$ to $K\alpha$ x-ray intensity ratio. In this present work, experimental η_{KL} have been evaluated using Eq. (4). The values of the K- and L-shell photoionization cross sections (σ_K^i and σ_L^i) have been taken from the tables of Scofield [14]. The ratio $(I_0G)_1/(I_0G)_2$ was determined by taking the x-ray spectra of thin sample Fe, ZnO_2, As_2O_3, SeO_2, $SrCl_2 \cdot 6H_2O$, ZrO_2, Mo, and Pd_2Cl excited by 59.5- and 122-keV photons and using the equation

$$\frac{(I_0G)_1}{(I_0G)_2} = \frac{N_1 \sigma_{K\alpha}^{(2)} \beta_{K\alpha}^{(2)}}{N_2 \sigma_{K\alpha}^{(1)} \beta_{K\alpha}^{(1)}}, \tag{9}$$

where all the symbols have a similar meaning, as explained above, except that these correspond to the K shell instead of the L shell. The $(I_0G)_1/(I_0G)_2$ ratios are found as 85 ± 7.

THEORETICAL K- TO L-SHELL VACANCY TRANSFER PROBABILITIES

The probabilities for vacancy transfer from K- to L-shell η_{KL_i} $(i=1,2,3)$, is defined as the number of primary L_i-subshell vacancies produced in the decay of one K-shell vacancy through radiative transition and through Auger transitions of the types $K-L_iL_j$ and $K-L_iX$ $(X=M,N,O,\ldots)$. This definition excluded the L_i subshell vacancies produced through a Coster-Kroning transition of the type L_i-L_jX. The Coster-Kronig (CK) transitions do not change the total K- to L-shell vacancy transfer probability. The η_{KL_i} have been evaluated using the following equations given by Rao and co-workers [12]:

$$\eta_{KL_1} = \frac{1}{\Gamma_{(K)}} [\Gamma_R(K_{L_1}) + 2\Gamma_A(K_{L_1}L_1) + \Gamma_A(K_{L_1}L_2) + \Gamma_A(K_{L_1}L_3) + \Gamma_A(K_{L_1}X)], \tag{10}$$

$$\eta_{KL_2} = \frac{1}{\Gamma_{(K)}} [\Gamma_R(K_{L_2}) + 2\Gamma_A(K_{L_2}L_2) + \Gamma_A(K_{L_2}L_2) + \Gamma_A(K_{L_1}L_2) + \Gamma_A(K_{L_2}X)], \tag{11}$$

$$\eta_{KL_3} = \frac{1}{\Gamma_{(K)}} [\Gamma_R(K_{L_3}) + 2\Gamma_A(K_{L_3}L_3) + \Gamma_A(K_{L_2}L_3) + \Gamma_A(K_{L_1}L_3) + \Gamma_A(K_{L_3}X)], \tag{12}$$

$$X = M,N,O$$

and

$$\eta_{KL} = \Sigma \eta_{KL_i}, \tag{13}$$

where $\Gamma_R(KL_i)$ is the radiative K-shell partial widths, $\Gamma_A(K-L_iL_j)$ and $\Gamma_A(K-L_iX)$ are the radiationless partial widths, and $\Gamma(K)$ is the total K-level widths.

The K- to L-shell vacancy probabilities based on the RDHD model, η_{KL} (RDHD), were evaluated using Eqs. (10)–(13). In these evaluations, we used the RDHS model-based K-shell radiative transition rates tabulated by Scofield [1] and the Auger transition rates based on the RDHS model calculated by Chen, Craseman, and Mark [2].

TABLE I. The comparison of experimental and theoretical results for K- to L-shell vacancy transfer probability.

Elements	$\eta_{KL}(E)$	$\eta_{KL}(T)$ Ref. [12]	$\eta_{KL}(T)$ Refs. [1,2]
Ta	0.829 ± 0.002		0.829
W	0.855 ± 0.004	0.821	0.827
Re	0.904 ± 0.005		0.825
Au	0.815 ± 0.008		0.816
Hg	0.815 ± 0.008	0.809	0.813
Tl	0.795 ± 0.009		0.812
Pb	0.805 ± 0.012	0.806	0.809
Bi	0.637 ± 0.013		0.807
Th	0.636 ± 0.013	0.795	0.794
U	0.682 ± 0.021	0.792	0.792

RESULTS AND DISCUSSION

The measured values of the K- to L-shell vacancy transfer probability η_{KL} for ten elements, namely, Ta, W, Re, Au, Hg, Tl, Pb, Bi, Th, and U are listed in Table I. The overall error in the measured values is estimated to be 8%. This error is attributed to the uncertainties in the different parameters used to deduce η_{KL} values, Eq. (4); namely, the error in the area evaluation under the L x-ray peak ($\leqslant 5\%$), in the absorption correction factor ($\leqslant 3\%$), in the $(I_0G)_1/(I_0G)_2$ factor ($\leqslant 4\%$), and other systematic errors ($\leqslant 4\%$). The experimental values η_{KL} are compared with the calculated values, η_{KL} (RDHS) and those tabulated by Rao, Chen, and Craseman η_{KL}(R) [12]. These values are found to be in good agreement with both the η_{KL}(R) values for the elements $73 \leqslant Z \leqslant 92$. The measured results show that the η_{KL}(R) values are quite reliable in the case of these elements. In this table, the calculated η_{KL} values using Eqs. (10)–(13) are also given. In these calculations, the radiative transition rates are tabulated by Scofield [1] and the radiationless transition rates are calculated by Chen, Craseman, and Mark [2]. These values are also in agreement with experimental results.

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