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Measurement of vacancy transfer probability from K to L shell using K-shell fluorescence yields

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Abstract. The vacancy transfer probabilities from K to L shell through radiative decay, η_{KL}, have been deduced for the elements in the range $19 \leq Z \leq 58$ using K-shell fluorescence yields. The targets were irradiated with γ photons at 59.5 keV from a 75mCi ^{241}Am annular source. The K X-rays from different targets were detected with a high resolution Si(Li) detector. The measurement of vacancy transfer probabilities are least-squared fitted to second-order polynomials to obtain analytical relations that represent these probabilities as a function of atomic number. The obtained results agree with theoretical and fitted values.

Keywords. Emission probability; Auger effect; photon interaction with atoms; vacancy; Coster–Kronig.

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1. Introduction

When a single vacancy is created in an inner shell (e.g. the K-shell), the vacancy will be rapidly filled up (10^{-8} s) by an electron coming from some higher (sub)shell, so that in a radiative decay a photon and in a radiationless (Auger) decay an electron is emitted. Reliable accurate values of the decay probabilities are required in order to derive the vacancy creation from the observed photons or electrons. To estimate this contribution, knowledge of K to L shell vacancy transfer probabilities is required. However, knowledge of average vacancy distribution is also important for the study of processes such as nuclear electron capture, interval conversion of γ-rays, photoelectric effect and generally, whenever primary vacancies produced in the shell must be distinguished from multiple ionization due to the decay of inner vacancy [1]. The K X-ray production cross-sections and fluorescence yields were measured experimentally [2–4]. Puri *et al* [5,6] have measured K to L shell and L to M shell vacancy transfer probabilities for elements in the atomic range $37 \leq Z \leq 42$ and $18 \leq Z \leq 96$, respectively. Puri *et al* [6] have calculated 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 and L_3 to M shells. K-shell fluorescence yields, mean L-shell fluorescence yields, ratios of X-ray emission probabilities and ratios of emission probabilities of Auger electrons were collected from the literature and evaluated by Schönfeld and JanBen [7]. The variation of the chemical effect of K-shell fluorescence yield and K_β/K_α X-ray intensity ratios were studied in refs [8–13]. The K-shell fluorescence yield is also defined as the probability that one K hole is filled through by radiative electron transition and is equal to the ratio of the X-ray emission rate to the total decay:

$$\omega_K = \frac{S_K^{Rad}}{(S_K^{Rad} + S_K^{Auger})}, \tag{1}$$

where S_K^{Rad} and S_K^{Auger} are the total radiative and Auger transition rates, respectively. The fluorescence yields lie between 0 and 1. For low atomic number elements, Auger decay rate is larger than X-ray emission. For high atomic number, X-ray emission becomes more probable. Auger transition probability increases with decreasing binding energy of the outer shell electrons. Ertuğrul [13] has measured total radiative and radiationless vacancy transfer probabilities from K to L_i subshell for some elements in the medium atomic number region. The vacancy transfer probabilities in the atomic shell have been studied by a few authors [14–17]. Some authors have measured K to L shell vacancy transfer probabilities using different processes [18–20].

In this study, the vacancy transfer probabilities from K to L shell were experimentally determined using K-shell fluorescence yields for some elements in the atomic number range $19 \leq Z \leq 58$.

2. Experimental

The experimental arrangement is shown in figure 1. The typical K X-ray spectrum obtained from Mo is given in figure 2. The studied elements are K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Mo, Ag, Cd, Ba, La and Ce. High purity (99%) samples of thickness $2–4\times10^{-2}$ g cm^{-2} were used for the preparation of the targets. In the experimental set-up, 59.5 keV γ-rays emitted from a 75 mCi ^{241}Am radioisotope source were detected by a calibrated Si(Li) detector (FWHM = 155 eV at 5.96 keV, active area = 12.5 mm^2, sensitive depth = 3.5 cm, Be window thickness = 12.5 μm) and a system 100 card with pulse height analyzer was used to count K_α and K_β photons emitted from samples. But, 59.5 keV photons emitted by a 100 mCi ^{241}Am annular radioactive source were used to stimulate K and Ca samples. The fluorescence K X-rays from the sample were detected by using a collimated Ultra-LEGe detector having a thickness of 5 mm and an energy resolution of 0.150 keV at 5.96 keV. The output from the pre-amplifier, with a pulse pile-up rejection capability, was fed to a multi-channel analyser interfaced with a personal computer provided with suitable software for data acquisition and peak analysis. The net peak area of the K X-rays of each target was determined after background subtraction, tailing and escape peak corrections [21].

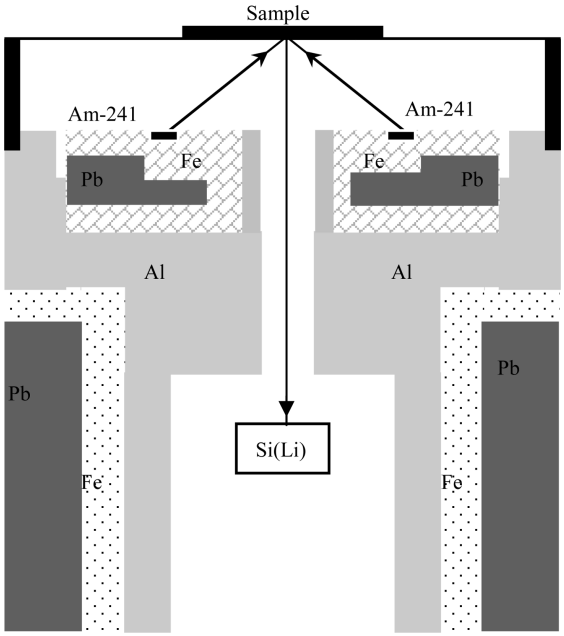

Figure 1. Experimental set-up.

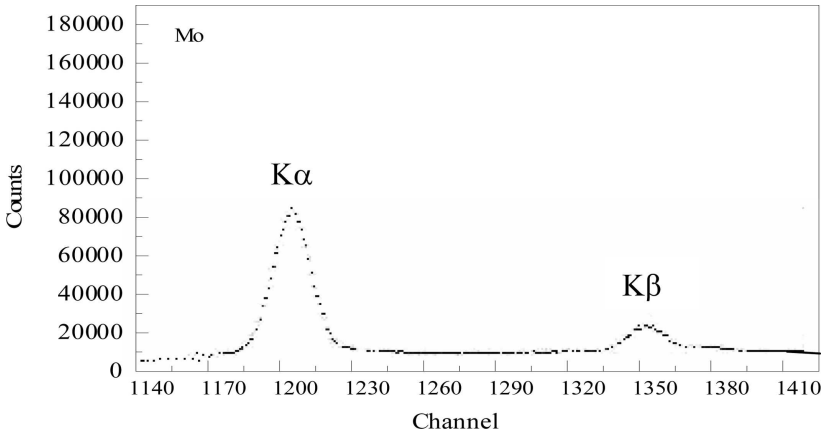

Figure 2. Characteristic K X-ray emission spectrum of Mo.

2.1 *Measurement of K-shell fluorescence yield*

K-shell fluorescence yield ω_{K} is determined using the following equation semi-empirically [5]:

$$\omega_{\mathrm{K}} = \frac{\sigma_{\mathrm{K}_\alpha,\mathrm{K}_\beta}}{\sigma_{\mathrm{K}}^{\mathrm{Photo}}}, \tag{2}$$

where $\sigma_{K_\alpha,K_\beta}$ is the total K X-ray fluorescence cross-section and σ_K^{photo} is the K-shell photoionization cross-section [22]. Experimental X-ray fluorescence cross-sections σ_{K_α} and σ_{K_β} were determined using the equation [5],

$$\sigma_{K_i} = \frac{N_{K_i}}{I_0 G \varepsilon_{K_i} \beta t}, \tag{3}$$

where $N_{K_i}(i = \alpha, \beta)$ is the intensity observed for K_i $(i = \alpha, \beta)$ X-ray line of element, ε_{K_i} is the detector efficiency for K_i X-rays, I_0 is the intensity of exciting radiation, G is the geometrical factor, t is the thickness of the sample in g cm^{-2} and β is the self-absorption correction factor of the target material. The self-absorption correction factor has been calculated using the following expression obtained by assuming the incidence angle of the fluorescent X-rays subtended at the detector to be approximately 90° [13].

$$\beta = \frac{1 - \exp\lfloor(-1)(\mu_{inc}/\cos\varphi + \mu_{emt})t\rfloor}{(\mu_{inc}/\cos\varphi + \mu_{emt})t}, \tag{4}$$

where μ_{inc} (cm^2 g^{-1}) and μ_{emt} (cm^2 g^{-1}) are the mass absorption coefficients [23] at the incident photon energy and fluorescent X-ray energy of the sample respectively and t (g cm^{-2}) is the measured thickness of the sample. $I_0 G \varepsilon_{K_i}$ values in the present experimental set-up were determined in a separate experiment. Targets of pure elements, having areas of cross-section similar to those used in the main experiment, with atomic number $19 \leq Z \leq 58$, emitting fluorescent radiation in the energy range 3.313–35 keV were irradiated in the same geometry and fluorescent radiation were counted. $I_0 G \varepsilon_{K_i}$ values for the present set-up were determined by the following equation [13]:

$$I_0 G \varepsilon_{K_i} = \frac{N_{K_i}}{\sigma_{K_i} \beta t}, \tag{5}$$

where N_{K_i} is the number of counts under the K_α or K_β peaks and σ_{K_i} is the photoionization cross-section [22]. The experimental probabilities of total vacancy transfer (η_{KL}) from K to L shell were evaluated from [7]

$$\eta_{KL} = \frac{2 - \omega_K}{1 + \left(\frac{IK_\beta}{IK_\alpha}\right)}, \tag{6}$$

where ω_K is the experimental K-shell fluorescence yield and IK_β/IK_α is the intensity ratios of K X-rays [24].

3. Results and discussion

The experimental results along with the theoretically calculated values are presented graphically as vacancy transfer probabilities vs. atomic numbers in figure 3 from which we observe that the values of η_{KL} decrease as the atomic numbers increase. The experimental and theoretical vacancy transfer probabilities are listed

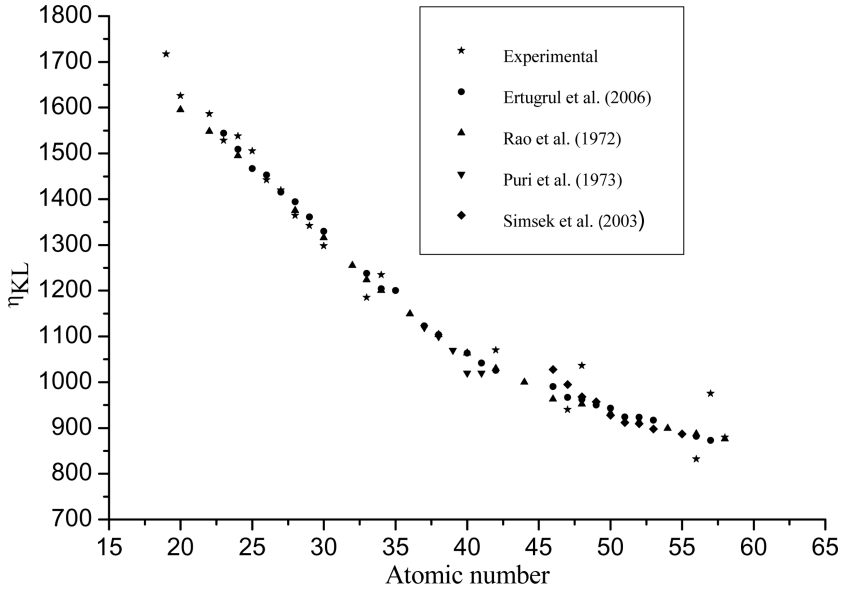

Figure 3. Total vacancy transfer probabilities of K to L shell as a function of atomic number.

in table 1. The overall error of these measurements is estimated to be 6–10%. The error in the evaluation of peak areas is 3%, the target thickness is ≤1–2%, I_0G product is 1–2% and the error in the absorption factor is ≤1–3%. It can be seen from table 1 and figure 3 that our measurement values are in good agreement, within experimental uncertainties, with the calculated theoretical values for the elements in the atomic range $19 \leq Z \leq 58$.

Our experimental values were fitted to a second-order polynomial as a function of atomic number $Z(\Sigma A_n Z^n)$ and fitted values of the K to L shell vacancy transfer probability are listed in table 1. These values have been plotted as a function of the atomic number and are shown in figure 3. The fitted equation is given as $Y(Z) = 2616.32434 - 57.7968Z + 0.48698Z^2$, $R^2 = 0.98054$ and SD = 40.11375.

As seen from table 1, the present values of η_{KL} are in good agreement (1–4%) with fitted values of Schönfeld and JanBen [7] (except for Sr and Fe) and the values of Ertuğrul *et al* and Şimşek and JanBen [14,25]. But, our fitted values are 1–3% different from Schönfeld's values [7]. Schönfeld and JanBen have evaluated the emission probabilities of X-rays and Auger electrons emitted in radioactive disintegration processes [26]. Furthermore, the probabilities of radiative vacancy transfer from L_i $(i = 1, 2, 3)$ subshells to the M, N and higher shells for elements with $77 \leq Z \leq 92$ have been measured by Sharma *et al* [20]. In addition, Çalışkan *et al* have measured the radiative vacancy distributions for the L_2, L_3 subshell and M shell of some elements in the atomic range $41 \leq Z \leq 68$ [27]. In early measurements [6,14,18] the authors used two radioisotope sources for the excitation of samples to determine the K to L shell vacancy transfers experimentally. This method was 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

Table 1. Comparison of experimental and theoretical results for K to L shell vacancy transfer probabilities.

	η_{KL}						
Element	Experimental	Ertuğrul *et al* [25]	Rao *et al* [1]	Puri *et al* [6]	Şimşek [14]	Schönfeld and JanBen [7]	Fit values
19	1.721±0.153	–	–	–	–	1.654	1.694
20	1.689±0.143	–	1.595	–	–	1.621	1.656
21	–	–	–	–	–	1.594	1.617
22	1.586±0.127	–	1.548	–	–	1.566	1.581
23	1.528±0.122	1.544	–	–	–	1.539	1.545
24	1.538±0.123	1.509	1.495	–	–	1.508	1.510
25	1.505±0.120	1.467	–	–	–	1.478	1.476
26	1.442±0.144	1.453	1.139	–	–	1.447	1.443
27	1.420±0.142	1.415	–	–	–	1.418	1.411
28	1.364±0.123	1.394	1.375	–	–	1.388	1.380
29	1.342±0.121	1.361	–	–	–	1.357	1.350
30	1.298±0.104	1.330	1.316	–	–	1.326	1.321
31	–	–	–	–	–	1.294	1.293
32	–	–	1.255	–	–	1.263	1.266
33	1.185±0.083	1.238	1.224	–	–	1.232	1.239
34	1.235±0.099	1.204	1.200	–	–	1.202	1.214
35	–	1.200	–	–	–	1.174	1.190
36	–	–	1.149	–	–	1.149	1.167
37	–	1.123	–	1.120	–	1.125	1.145
38	1.341±0.107	1.104	1.104	1.100	–	1.102	1.123
39	–	–	–	1.070	–	1.081	1.103
40	–	1.064	1.064	1.030	–	1.062	1.084
41	–	1.042	–	1.020	–	1.045	1.065
42	1.070±0.097	1.026	1.030	1.020		1.029	1.048
43	–	–	–	–	–	1.014	1.032
44	–	–	1.000	–	–	1.000	1.016
45	–	–	–	–	–	0.987	1.001
46	–	0.990	0.963	–	1.028	0.975	0.988
47	0.940±0.080	0.967	–	–	0.995	0.964	0.976
48	1.036±0.104	0.962	0.952	–	0.968	0.953	0.964
49	–	0.950	–	–	0.957	0.944	0.954
50	–	0.943	0.932	–	0.928	0.934	0.944
51	–	0.924	–	–	0.912	0.925	0.935
52	–	0.923	0.914	–	0.909	0.917	0.928
53	–	0.917	–	–	0.898	0.909	0.921
54	–	–	0.899	–	–	0.902	0.915
55	–	–	–	–	0.887	0.895	0.911
56	0.832±0.042	0.882	0.887		–	0.888	0.907
57	0.975±0.060	0.873	–	–	–	0.882	0904
58	0.879±0.079	–	0.876	–	–	0.876	0.902

to L shell vacancies comes from the decay of K shell vacancies. Consequently, their methods strongly depend on the L X-ray cross-sections. Moreover, the vacancy transitions from K to L shell enhance the L X-ray cross-sections. In addition, as seen from table 1 and figure 3, radiative vacancy transition probabilities decrease with the increasing atomic number. The reason of this may be that the linewidth increases with the increase in the number of electrons in the external shells.

Consequently, the probabilities of radiative and non-radiative vacancy transfers have been studied extensively but not in the range $19 \leq Z \leq 58$. Since the probabilities of radiative and non-radiative vacancy transfers directly depend on the electronic structure of the atom, it is necessary to reinvestigate these probabilities in accordance with the new developments in detection system technology.

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