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Measurement of Ll, $L\alpha$, $L\beta$, and $L\gamma$ x-ray-production cross sections in some high-Z elements by 60 keV photons

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Ll, $L\alpha$, $L\beta$, and $L\gamma$ x-ray-production cross sections have been measured in elements Ta, W, Au, Hg, Tl, Pb, Bi, Th, and U for 60-keV photons. The measured values have been interpreted in terms of photoelectric cross sections, fluorescence yields, Coster-Kronig transition probabilities, and radiative decay rates. A fairly good agreement is found between the experimental and calculated values.

INTRODUCTION

The L x-ray emission spectra of heavy elements taken with the currently available Si(Li) detectors, show four or five distinct peaks. Each peak covers a group of lines[1] of the L x-ray series which have very close energies and thus cannot be resolved due to limited resolution of the detectors. We have measured the cross sections for the production of x-rays in each of the Ll, $L\alpha$, $L\beta$, and $L\gamma$ groups in elements Ta, W, Au, Hg, Tl, Pb, Bi, Th, and U by 60-keV photons. These measurements are important for two reasons. First, the x-ray-production cross sections for various groups of lines are needed for the quantitative estimation of elements in various types of samples using photon-induced x-ray fluorescence technique. Second, these measurements serve to provide a check on the

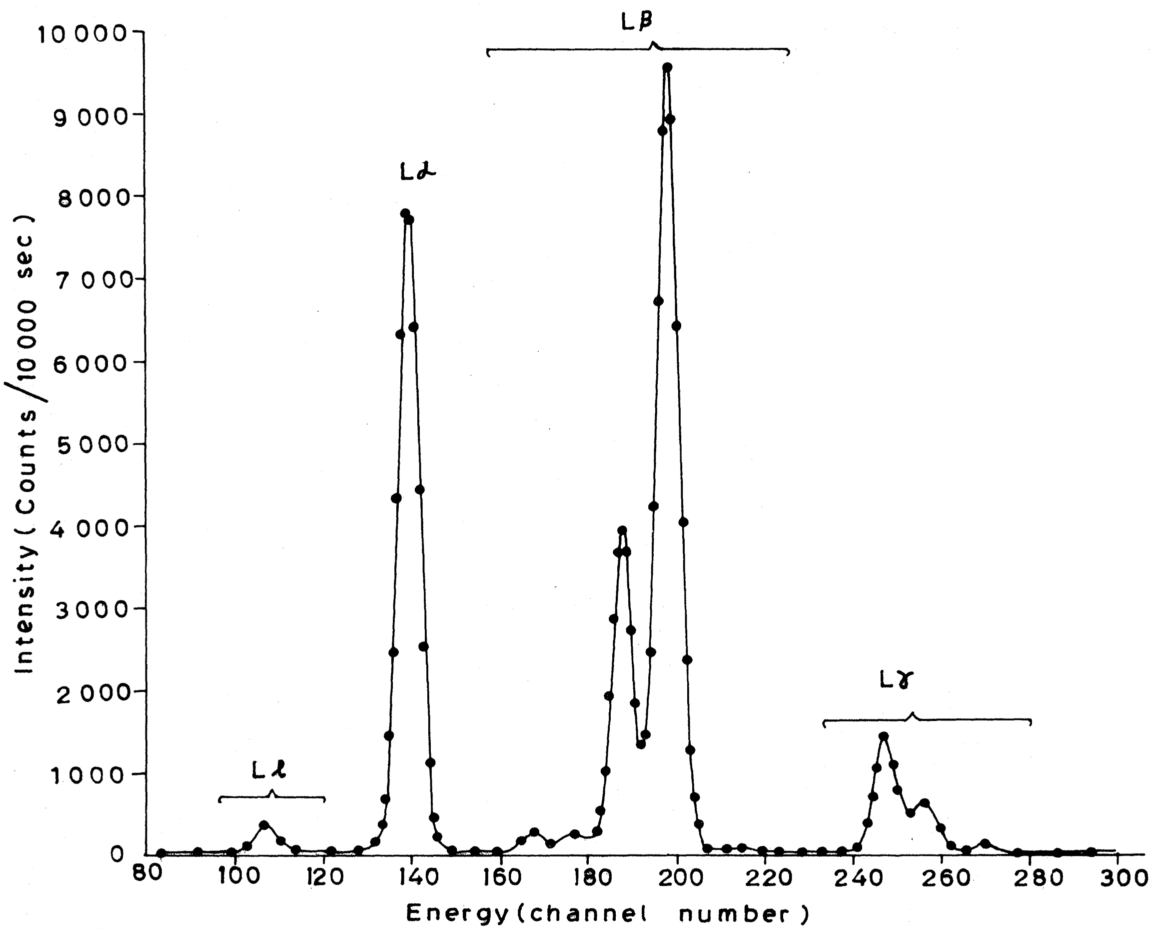

FIG. 1. Thorium L x-ray spectrum recorded with a Si(Li) x-ray detector when the target was irradiated with 59.57-keV γ rays from ^{241}Am (background subtracted).

theoretical calculations of some of the fundamental physical parameters such as L-subshell ionization cross sections, fluorescence yields, Coster-Kronig transition probabilities, and radiative decay rates, the direct determination of which render many difficulties. The methods of measurement and analysis of results are reported in this paper.

METHOD OF MEASUREMENT

The self-supporting targets of elements Ta, W, Au, Hg, Tl, Pb, Bi, Th, and U in the form of circular disks of 4 cm diameter were irradiated with 59.57-keV γ rays[2] from an $\sim$1-Ci ^{241}Am source in a 90° reflection geometry setup. The metallic targets of Ta, W, Au, Pb, Th, and U purchased from Reactor Experiments Inc., U.S.A., were used in the present measurements whereas the targets of Hg, Tl, and Bi were made from their stable compounds by the technique discussed earlier.[3] The L-shell fluorescent x rays emitted from the targets were analyzed by a Si(Li) detector (effective area 200 mm^2, thickness 5 mm, Be window thickness 0.05 mm) coupled to an ND 600 multichannel analyzer system. The resolution of the Si(Li) x-ray spectrometer was $\sim$240 eV at 5.9 keV. The other details of the experimental setup were similar to the setup discussed in detail in an earlier paper.[2] The choice of the incident energy in the present experiment was such that the K-shell electrons of the target elements were not ionized. The vacancies were therefore not transferred from K shell to L shell. The interpretation of the experimental results was simplified as all the initial vacancies were created only by the photoionization of the three L-subshell electrons. A typical L x-ray spectrum of Th is shown in Fig. 1. The peaks due to the Ll, $L\alpha$, $L\beta$, and $L\gamma$ group of lines are well resolved.

The x-ray-production cross section σ_{Li}^{x} for the production of x rays in the Li group is given by

$$\sigma_{Li}^{x} = N_{Li}(x)\frac{(4\pi)^2}{S\alpha\omega_1\omega_2\epsilon_{Li}}\frac{M_L}{Nt_L\beta_{Li}}, \quad i = l,\alpha,\beta,\gamma \tag{1}$$

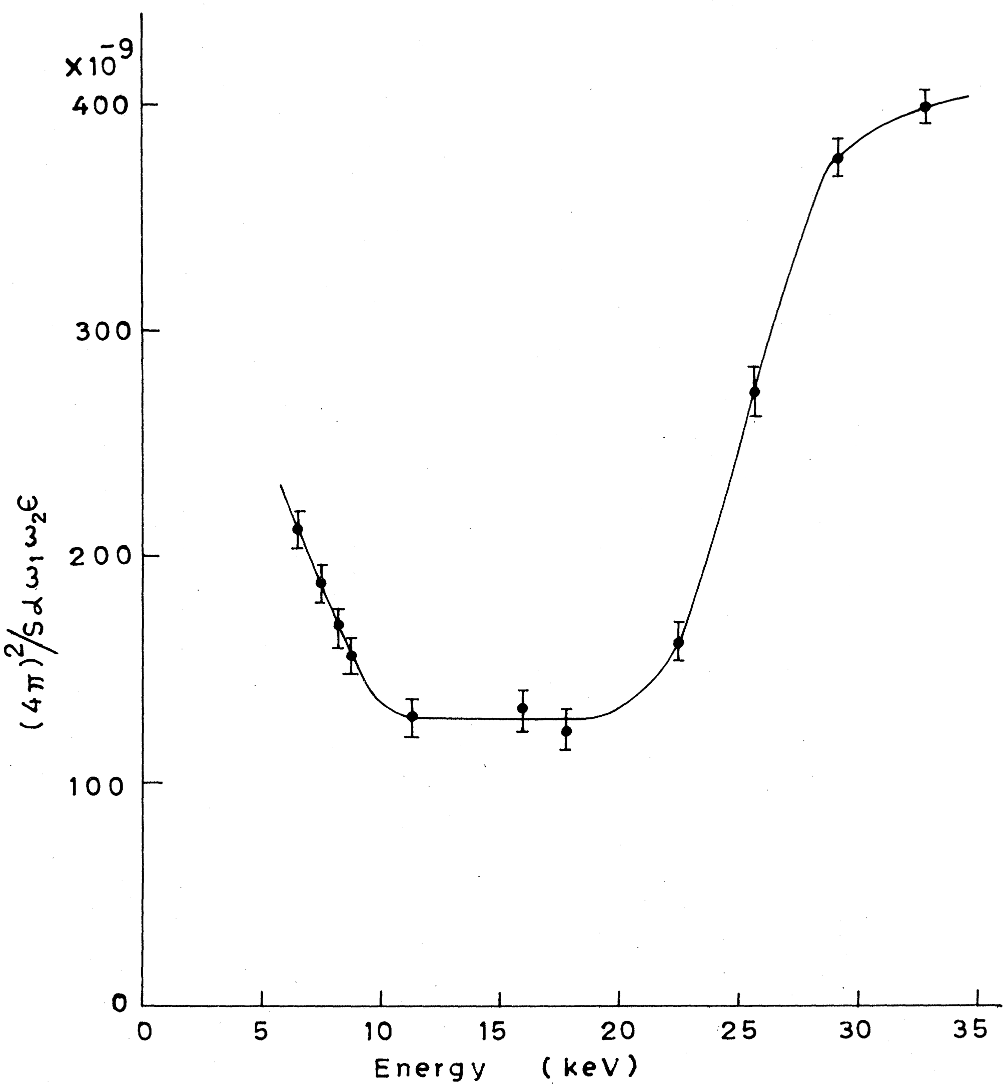

FIG. 2. Plot of $[(4\pi)^2/s\alpha\omega_1\omega_2\epsilon]$ vs photon energy.

where $N_{Li}(x)$ is the number of x rays falling under the peak due to the Li group as measured with the spectrometer. S is the number of γ rays emitted from the source, α is the correction factor for absorption of γ rays in source, air column, etc., N is Avogadro's number, M_L is the atomic weight of the target element, ω_1 and ω_2 are source-target and target-detector solid angles, respectively, ϵ_{Li} is the photopeak efficiency of the detector at Li x-ray energy, t is the thickness of target. β_{Li} is the self-absorption correction factor of the target for the incident γ-ray energy and the emitted Li x-ray energy and it can be expressed[4] in terms of the known values of fractional intensities p_j, absorption coefficients of the target element at the incident γ-ray energy μ_γ and emitted Li x-ray energy μ_{xi}, and the target thickness t as

$$\beta_{Li}=\sum_{j=1}^{n} p_j \frac{1-\exp\left[-\left(\frac{\mu_\gamma+\mu_{x_{ij}}}{\cos\theta}\right)t\right]}{\left[\frac{\mu_\gamma+\mu_{x_{ij}}}{\cos\theta}\right]t} . \quad (2)$$

The value of θ in the present experiment was 45°. The values of N_{Li} were determined from the areas under the Ll, $L\alpha$, $L\beta$, and $L\gamma$ x-ray peaks. Sufficient numbers of runs were taken to achieve a statistical accuracy of $\sim 1\%$ in the counting rates. The values of the factor $(4\pi)^2/S\alpha\omega_1\omega_2\epsilon_{Li}$, which contains the terms relating to the flux of 59.57-keV γ rays emitted from the source, geometrical factor, and absolute efficiency of the x-ray detector, were determined in a separate experiment. For this purpose the targets of elements Fe, Ni, Cu, Zn, Se, Zr, Mo, Ag, Sn, I, and Ba having the same diameter as in the main experiment were irradiated with γ rays from the source and in the same experimental setup, and the intensities of their K-shell fluorescent x rays emitted in each case were recorded with the same x-ray detector as used for L x rays. The number of K x rays $N_K(x)$ emitted from the target in this experiment and counted under the photopeak per unit time are given by a relation similar to (1) as above which can be rewritten as

$$\frac{(4\pi)^2}{S\alpha\omega_1\omega_2\epsilon}=\frac{\sigma_K^x N t_K \beta_K}{N_K(x) M_K} . \quad (3)$$

All the terms in Eq. (3) have the same meaning as discussed above in Eq. (1), but the subscript K indicates that these terms correspond to K shell. The K-shell x-ray-production cross section σ_K^x is related to the K-shell photoionization cross section σ_K^P through the K-shell fluorescence yield ω_K as

$$\sigma_K^x=\sigma_K^P\omega_K . \quad (4)$$

Using the measured values of N_K, and from the knowledge of K-shell photoionization cross sections,[5] K-shell fluorescence yields,[6] and absorption coefficients,[7] the values of the term $(4\pi)^2/S\alpha\omega_1\omega_2\epsilon$ were determined at weighted mean K x-ray energies[1] of elements $22\le Z\le 56$ and plotted against energy as shown in Fig. 2. The values of this term at Ll, $L\alpha$, $L\beta$, and $L\gamma$ x-ray energies of the target elements were read from the graph to calculate the values of the partial L-shell x-ray-production cross sections σ_{Ll}^x, $\sigma_{L\alpha}^x$, $\sigma_{L\beta}^x$, and $\sigma_{L\gamma}^x$.

RESULTS AND DISCUSSION

The measured values of the Ll, $L\alpha$, $L\beta$, and $L\gamma$, x-ray-production cross sections in elements Ta, W, Au, Hg, Tl, Pb, Bi, Th, and U are listed in Table I. To the best of our knowledge no other experimental data are available for comparison with the present results. The values of Ll, $L\alpha$, $L\beta$, and $L\gamma$ x-ray-production cross sections are calculated from the theoretical values of L-subshell photoionization cross sections[5] and radiative decay rates,[8] semiempirically fitted values of fluorescence yields,[6] and Coster-Kronig transition probabilities[6] using the following relations:[9]

$$\sigma_{Ll}^x=(\sigma_1 f_{13}+\sigma_1 f_{12} f_{23}+\sigma_2 f_{23}+\sigma_3)\omega_3 F_{3l} ,$$

$$\sigma_{L\alpha}^x=(\sigma_1 f_{13}+\sigma_1 f_{12} f_{23}+\sigma_2 f_{23}+\sigma_3)\omega_3 F_{3\alpha} ,$$

$$\sigma_{L\beta}^x=\sigma_1\omega_1 F_{1\beta}+(\sigma_2+\sigma_2 f_{12})\omega_2 F_{2\beta} + (\sigma_1 f_{13}+\sigma_1 f_{12} f_{23}+\sigma_2 f_{23}+\sigma_3)\omega_3 F_{3\beta} ,$$

$$\sigma_{L\gamma}^x=\sigma_1\omega_1 F_{1\gamma}+(\sigma_2+\sigma_2 f_{12})\omega_2 F_{2\gamma} ,$$

where σ_1, σ_2, and σ_3 are L-subshell photoionization cross sections of the element at 59.57 keV; ω_1, ω_2, and ω_3 are L-subshell fluorescence yields; f_{12}, f_{23}, and f_{13} are intra-

TABLE I. Comparison of the present measurements of Ll, $L\alpha$, $L\beta$, and $L\gamma$ x-ray-production cross sections in elements $73\le Z\le 92$ by 60-keV photons with calculated values.

		σ_{Ll}^x		$\sigma_{L\alpha}^x$		$\sigma_{L\beta}^x$		$\sigma_{L\gamma}^x$	
Z	Element	Present (b/at.)	Calc. (b/at.)	Present (b/at.)	Calc. (b/at.)	Present (b/at.)	Calc. (b/at.)	Present (b/at.)	Cal. (b/at.)
73	Ta	2.7±0.2	3.3	58±4	64	115±6	126	23±2	25.3
74	W	3.2±0.2	3.3	69±5	69	129±6	133	27±2	27.8
79	Au	7.7±0.6	7.3	146±9	142	154±7	149	34±2	26.5
80	Hg	9.1±0.5	8.3	155±9	156	152±7	158	30±2	28.8
81	Tl	10.3±0.6	9.3	166±9	175	168±9	182	37±3	35.0
82	Pb	9.6±0.5	10.4	187±12	192	225±13	216	44±3	40.9
83	Bi	13.4±0.7	11.3	217±14	205	233±15	217	50±4	41.4
90	Th	20.8±1.5	20.9	303±20	342	362±23	440	76±5	81.1
92	U	31.5±2.0	25.6	477±33	407	549±36	496	120±6	112.7

L-shell Coster-Kronig transition probabilities, F's are fractional radiative decay rates, and $F_{3\alpha}$ is the fraction of $L_{\rm III}$-subshell x rays which contributes to the $L\alpha$ peak of the x-ray spectrum of an element. All other F's are similarly defined. The present experimental values of Ll, $L\alpha$, $L\beta$, and $L\gamma$ x-ray-production cross sections show a good agreement with the calculated values. The errors in the present measurements are $\sim$6–8% and are due to the counting statistics and the uncertainties involved in the calculations and/or measurement of other parameters used for the determination of the production cross sections from Eq. (1).

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