PHYSICAL REVIEW A VOLUME 57, NUMBER 4 APRIL 1998

Measurement of *K*-shell fluorescence yields of selected elements from Cs to Pb using radioisotope x-ray fluorescence

Rıdvan Durak and Yusuf Şahin
Department of Physics, Faculty of Sciences and Arts, Atatürk University, 25240 Erzurum, Turkey

(Received 19 February 1997)

K-shell fluorescence yields were measured for Cs, Sm, Eu, Ho, Ta, W, Hg, and Pb using a Ge(Li) detector employing the reflection geometry. The target atoms were excited by using γ rays from ^{57}Co radioactive sources of strength 100 mCi. Recently determined values of w_K for Ba, Ce, Nd, Gd, Dy, Er, and Yb are also tabulated. The experimental results are compared with the literature experimental values, theoretical predictions, and the semiempirical fits. [S1050-2947(98)07703-8]

PACS number(s): 32.30.Rj, 34.50.Fa, 32.80.Fb, 32.80.Hd

I. INTRODUCTION

The deexcitation of an atom with an inner-shell *K* vacancy can proceed either by the emission of an x-ray photon or by the ejection of Auger electrons. These decays are assumed to be energy independent. The deexcitation of an atomic shell is characterized by the fluorescence yield and it is defined as the probability that a vacancy in the *K* shell is filled through a radiative transition. An accurate knowledge of fluorescence yield from the *K* shell is required in various applications such as atomic, molecular, and radiation physics studies, elemental x-ray fluorescence analysis, medical research, cancer therapy, and irradiational processes [1–4].

K-shell fluorescence yields w_K for different elements have been investigated for many years. Bambynek *et al.* [2] in a review article have fitted their collection of selected *most reliable* experimental values in the $13 \leqslant Z \leqslant 92$ range. Krause [5] presented a table of w_K *adopted values* for elements $5 \leqslant Z \leqslant 110$ by using all theoretical and experimental data on the parameters contributing to the *K*-shell fluorescence yield. Bambynek [6] reevaluated the *K*-shell fluorescence yields incorporating about 100 new measurements and produced an improved fit. In a recent review Hubbell *et al.* [1] calculated up-to-date fitted *K*-shell fluorescence yield values in the $1 \leqslant Z \leqslant 100$ interval. Theoretical values of w_K were obtained in the region $4 \leqslant Z \leqslant 54$ by McCuire [7,8] and Walters and Bhalla [9] using the Hartree-Fock-Slater model. Kostroun *et al.* [10] presented computations for elements in the range $10 \leqslant Z \leqslant 70$ by combining Scofield's radiative widths [11] with radiationless transition probabilities calculated from nonrelativistic hydrogenic wave functions. Chen *et al.* [12] used a Dirac-Hartree-Slater approach to list the w_K values of elements in the $18 \leqslant Z \leqslant 96$ range. However, measured [3,13,14] and theoretical [12] *K*-shell fluorescence yields data for rare-earth and heavy-elements are scarce.

K-shell fluorescence yields for different elements, in the atomic range $56 \leqslant Z \leqslant 70$, has been undertaken previously [15]. In a continuation of this work, the *K*-shell fluorescence yields for Cs, Sm, Eu, Ho, Ta, W, Hg, and Pb have been measured using a fluorescence excitation method to excite target atoms and a reflection geometry setup. The results of the present work are compared with earlier experimental results obtained by other methods, theoretical predictions, and semiempirical fits reported in the literature. To the best of our knowledge the *K*-shell fluorescence yields using a fluorescence excitation method are measured for the first time for Cs, Eu, and Hg.

II. EXPERIMENT

The experimental arrangement employed for the measurements has been described elsewhere [15]. In the arrangement low-energy photon sources of ^{57}Co with strength 100 mCi is used. The *K* x-ray spectra from different samples were recorded with a Ge(Li) detector coupled to a 4096 channel Nd-66B multichannel analyzer. The measured energy resolution of the detector system was 190 eV full width at half maximum at 5.9 keV (^{55}Fe). Spectroscopically pure (purity better than 99.9%) circular disk samples of 31 mm diam and thickness from 15 to 65 mg cm^{-2} have been used for the measurements. The contribution to the production of target x rays due to the 136.48-keV photons emitted from the ^{57}Co source is insignificant because of its low intensity and low

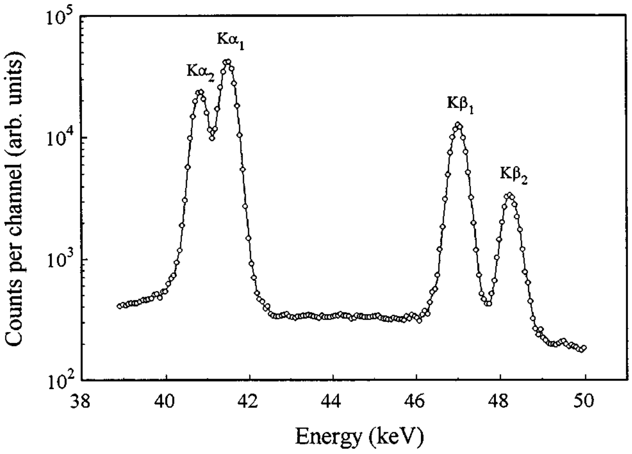

FIG. 1. Typical *K* x-ray spectrum of Eu excited by 122-keV γ rays from ^{57}Co.

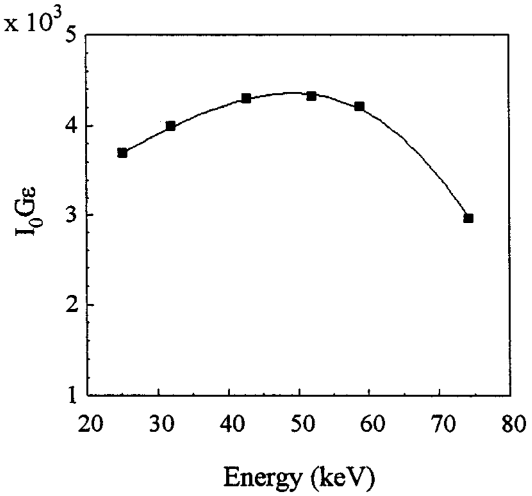

FIG. 2. Factor $I_0G\varepsilon$ as a function of mean K x-ray energy.

photoionization cross section. So photons of 122-keV energy from this source were considered primary photons. An Al shield of thickness 0.25 mm was used with the ^{57}Co source to suppress the low-energy photons emitted from the radioisotope. Targets were measured for a time interval ranging from 500 to 6000 s depending on the counting statistics. Due to incomplete charge collection in the detector, characteristic peaks contain a low-energy tail, which makes it difficult to obtain the net peak areas. A tail stripping procedure was therefore applied for the spectrum to obtain net peak areas [16]. In addition, escape corrections were also made. A typical K x-ray spectrum of Eu is shown Fig. 1. The effective incident photon flux $I_0G\varepsilon_{K\alpha}$ was determined by measuring the K x-ray yields from Sn, Ba, Gd, Yb, W, and Pb in the same geometry. The measured $I_0G\varepsilon_{K\alpha}$ factor was plotted as a function of the mean K x-ray energy as shown in Fig. 2.

III. DATA ANALYSIS

The experimental K-shell fluorescence yield was measured according to the equation

$$\sigma_{K\alpha}^{x}=\sigma_{K}^{p}(E)w_K f_{K\alpha}, \tag{1}$$

where $\sigma_K^p(E)$ is the K-shell photoionization cross section for the given element at excitation energy E, w_K is the K-shell fluorescence yield, $f_{K\alpha}$ is fractional rate of the $K\alpha$ line intensity relative to that of the K shell and is given by

$$f_{K\alpha}=[1+I_{K\beta}/I_{K\alpha}]^{-1}, \tag{2}$$

and $\sigma_{K\alpha}^x$ is K x rays production cross section [17] and is defined as

$$\sigma_{K\alpha}^{x}=\frac{I_{K\alpha}}{I_0GT\varepsilon_{K\alpha}t}, \tag{3}$$

where $I_{K\alpha}$ is the net counts under the corresponding photopeak, I_0G is the intensity of exciting radiation falling on the sample, $\varepsilon_{K\alpha}$ is the detector efficiency for the $K\alpha$ x rays, t is the mass of the sample in g cm^{-2}, and T is the self-absorption correction factor of the target material, which accounts for the absorption in the target of the incident photons and emitted characteristic x rays. T is evaluated by the relation

$$T=\frac{1-\exp[-\beta(E_i)t]}{\beta(E_i)t}, \tag{4}$$

with

$$\beta(E_i)=\frac{\mu_{\text{inc}}}{\cos\theta_1}+\frac{\mu_{\text{emt}}}{\cos\theta_2}, \tag{5}$$

TABLE I. Comparison of present experimental and literature values of the K-shell fluorescence yields.

Element	Present work	Durak (1997)	Balakrishna (1994)	Sidhu[a] (1988)	Al-Nasr (1987)
^{55}Cs	0.9137± 0.028			0.899± 0.015	
^{56}Ba		0.9242±0.068			0.920± 0.051
^{58}Ce		0.9308±0.067			
^{60}Nd		0.9416±0.069			
^{62}Sm	0.9421± 0.053		0.933± 0.046		
^{63}Eu	0.9437± 0.049			0.957± 0.030	
^{64}Gd		0.9458±0.061	0.922± 0.045		
^{66}Dy		0.9560±0.063	0.954± 0.048	0.975± 0.027	
^{67}Ho	0.9534± 0.057		0.939± 0.049		
^{68}Er		0.9394±0.061			
^{70}Yb		0.9661±0.049	0.925± 0.051		
^{73}Ta	0.9641± 0.047		0.962± 0.054	0.955± 0.011	
^{74}W	0.9683± 0.054		0.956± 0.054		
^{80}Hg	0.9707± 0.036			0.980± 0.009	
^{82}Pb	0.9732± 0.058		0.961± 0.055		

[a] This value has been measured from a knowledge of nuclear decay parameters.

TABLE II. Present experimental results and theoretical predictions of w_K.

		Theoretical prediction[a]		
Element	Present work	Durak (1997)	Kostroun (1971)	Chen (1980)
^{55}Cs	0.9137±0.028			
^{56}Ba		0.9242±0.068	0.916	0.899
^{58}Ce		0.9308±0.067	0.926	
^{60}Nd		0.9416±0.069	0.935	0.918
^{62}Sm	0.9421±0.053			
^{63}Eu	0.9437±0.049			0.929
^{64}Gd		0.9458±0.061		
^{66}Dy		0.9560±0.063		
^{67}Ho	0.9534±0.057			0.940
^{68}Er		0.9394±0.061		
^{70}Yb		0.9661±0.049	0.963	0.947
^{73}Ta	0.9641±0.047			
^{74}W	0.9683±0.054			0.954
^{80}Hg	0.9707±0.036			0.962
^{82}Pb	0.9732±0.058			

[a]The theoretical values reported by Chen *et al.* are available for four of the elements studied in the present work.

where μ_{inc} and μ_{emt} are the total mass absorption coefficients ($\mathrm{cm}^2\,\mathrm{g}^{-1}$) of incident photon and emitted characteristic x rays, respectively [18]. The incidence and emission angles with respect to the target normal, θ_1 and θ_2 were set to 45° and 0°, respectively. In the present calculations, the values of $\sigma_K^p(E)$ were taken from Scofield [19] based on Hartree-Slater potential theory.

IV. RESULTS AND DISCUSSION

The measured K-shell fluorescence yields are presented in Table I and compared with the available literature experimental results. The errors in the experimental K-shell fluorescence yields are estimated to be 4–6%. This error arises from uncertainties in the various parameters used to calculate the K fluorescence yields, including errors due to peak area evaluation ($<2\%$ for $K\alpha$ and $<4\%$ for $K\beta$ peaks), $I_0G\varepsilon$ factor (3%), target thickness measurements ($\sim 4\%$), and absorption correction factor ($\sim 2\%$). All the errors were compounded according to the classical rules of the propagation of errors and the resultant error is quoted on the measured fluorescence yields [20]. From Table I, it can be seen that the present data are in agreement within the experimental uncertainties with the literature experimental results.

In Table II the present experimental results of K fluorescence yields are compared with only one theoretical prediction reported in the literature [12]. Chen *et al.* [12] have used a relativistic Dirac-Hartree-Slater model to derive theoretical K-shell fluorescence yields. The theoretical values reported by Chen *et al.* are available for four of the elements studied in the present work. The agreement between the present results and theoretical predictions of Chen *et al.* is within the range 1–1.5%.

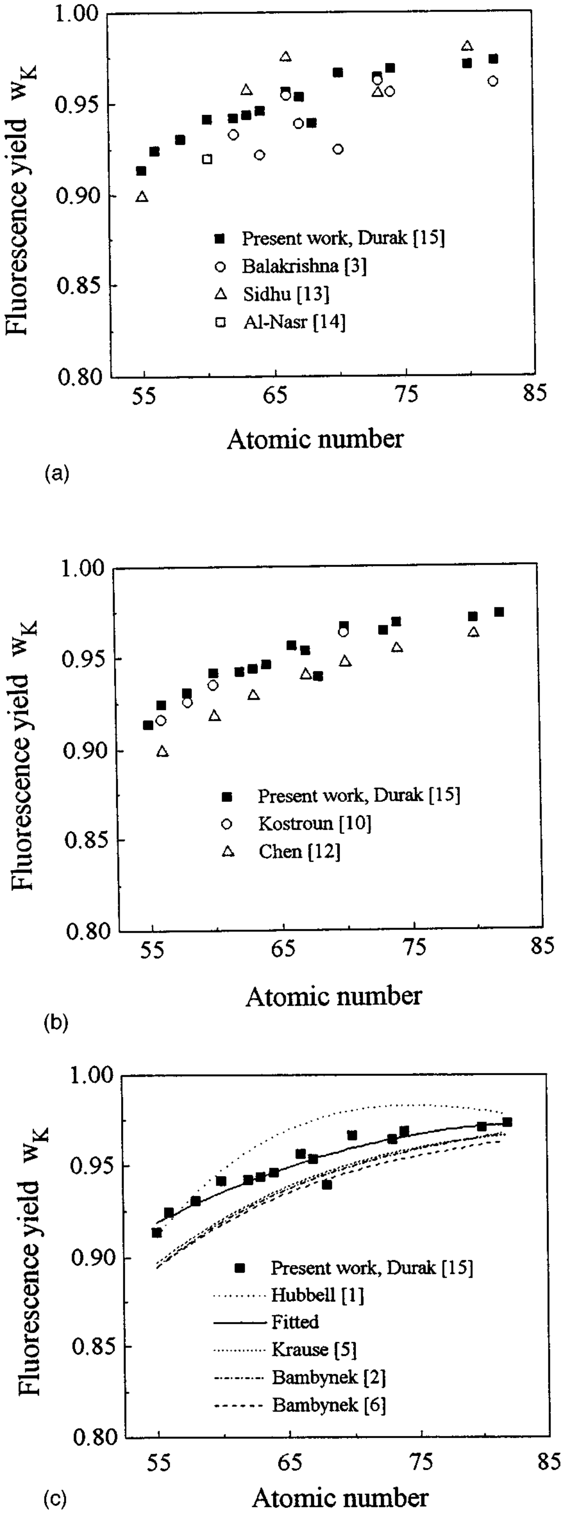

FIG. 3. Comparison of measured w_K values with literature experimental results (a), theoretical prediction (b), and semiempirical fits (c).

The present measured values of w_K are compared in Table III with the semiempirical fits [1,2,5,6]. Our experimental data were fitted to a third-order polynomial as a function of atomic number and fitted values of K-shell fluorescence yields w_K for all elements in the range $55 \leqslant Z \leqslant 82$ listed in the same table. The experimental results agree within 0.2–2.3% with the K fluorescence yields calculated using a semi-

TABLE III. Present experimental results and semiempirical fits values of w_K.

				Semiempirical values			
Element	Present work	Durak (1997)	Fitted values	Bambynek (1972)	Krause (1979)	Bambynek (1984)	Hubbell (1994)
^{55}Cs	0.9137±0.028		0.917	0.895	0.897	0.8942	0.912
^{56}Ba		0.9242±0.068	0.922	0.900	0.902	0.8997	0.920
^{57}La			0.926	0.906	0.907	0.9047	0.928
^{58}Ce		0.9308±0.067	0.929	0.911	0.912	0.9096	0.935
^{59}Pr			0.933	0.915	0.917	0.9140	0.941
^{60}Nd		0.9416±0.069	0.936	0.920	0.921	0.9181	0.947
^{61}Pm			0.939	0.924	0.925	0.9220	0.953
^{62}Sm	0.9421±0.053		0.942	0.927	0.929	0.9255	0.958
^{63}Eu	0.9437±0.049		0.945	0.931	0.932	0.9289	0.962
^{64}Gd		0.9458±0.061	0.947	0.934	0.935	0.9320	0.966
^{65}Tb			0.949	0.937	0.938	0.9349	0.969
^{66}Dy		0.9560±0.063	0.952	0.940	0.941	0.9376	0.972
^{67}Ho	0.9534±0.057		0.954	0.943	0.944	0.9401	0.975
^{68}Er		0.9394±0.061	0.956	0.945	0.947	0.9425	0.977
^{69}Tm			0.957	0.947	0.949	0.9447	0.979
^{70}Yb		0.9661±0.049	0.959	0.950	0.951	0.9467	0.980
^{71}Lu			0.960	0.952	0.953	0.9487	0.981
^{72}Hf			0.962	0.954	0.955	0.9505	0.982
^{73}Ta	0.9641±0.047		0.963	0.956	0.957	0.9522	0.983
^{74}W	0.9683±0.054		0.965	0.957	0.958	0.9538	0.983
^{75}Re			0.966	0.959	0.959	0.9553	0.983
^{76}Os			0.967	0.960	0.961	0.9567	0.983
^{77}Ir			0.968	0.962	0.962	0.9580	0.982
^{78}Pt			0.969	0.963	0.963	0.9592	0.981
^{79}Au			0.970	0.964	0.964	0.9604	0.980
^{80}Hg	0.9707±0.036		0.971	0.966	0.965	0.9615	0.980
^{81}Tl			0.972	0.967	0.966	0.9625	0.979
^{82}Pb	0.9732±0.058		0.974	0.968	0.967	0.9634	0.978

empirical expression. The present experimental values agree better with the semiempirical values deduced by Krause [5]; the agreement is within 1.5% for all elements except for Cs. The fitted w_K values are seen to be in general agreement within the uncertainties indicated in the measured values columns, which range from 0.5% to 2.5%. The comparison with theoretical predictions and semiempirical fits are also shown graphically in Fig. 3. Consequently, more experimental and theoretical data for the elements in the region of $Z > 54$ are needed for full knowledge of w_K.

---

[1] J. H. Hubbell, P. N. Trehan, N. Singh, B. Chand, D. Mehta, M. L. Garg, R. R. Garg, S. Singh, and S. Puri, J. Phys. Chem. Ref. Data **23**, 339 (1994).

[2] W. Bambynek, B. Crasemann, R. W. Fink, H. U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. V. Rao, Rev. Mod. Phys. **44**, 716 (1972).

[3] K. M. Balakrishna, N. G. Nayak, N. Lingappa, and K. Siddappa, J. Phys. B **27**, 715 (1994).

[4] J. H. Hubbell, National Institute of Standards and Technology Center for Radiation Research, Document No. NISTIR 89-4144, 1989 (unpublished).

[5] M. O. Krause, J. Phys. Chem. Ref. Data **8**, 307 (1979).

[6] W. Bambynek (unpublished).

[7] E. J. McGuire, Phys. Rev. **185**, 1 (1969).

[8] E. J. McGuire, Phys. Rev. A **24**, 273 (1970).

[9] D. L. Walters and C. P. Bhalla, Phys. Rev. A **3**, 1919 (1971).

[10] V. O. Kostroun, M. H. Chen, and B. Crasemann, Phys. Rev. A **3**, 533 (1971).

[11] J. H. Scofield, Phys. Rev. **179**, 9 (1969).

[12] M. H. Chen, B. Crasemann, and H. Mark, Phys. Rev. A **21**, 436 (1980).

[13] N. P. S. Sidhu, B. S. Grewal, and H. S. Sahota, X-Ray Spectrom. **17**, 29 (1988).

[14] I. A. Al-Nasr, I. J. Jabr, K. A. Al-Saleh, and N. S. Saleh, Appl.

Phys. A: Solids Surf. **43**, 71 (1987).

[15] R. Durak and Y. Şahin, Nucl. Instrum. Methods Phys. Res. B **124**, 1 (1997).

[16] Y. Şahin, R. Durak, Y. Kurucu, and S. Erzeneoğlu, J. Radioanal. Nucl. Chem. Articles **177**, 403 (1994).

[17] R. Durak, S. Erzeneoğlu, Y. Kurucu, and Y. Şahin, Radiat. Phys. Chem. (to be published).

[18] J. H. Hubbell and S. M. Seltzer, National Institute of Standards and Technology Center for Radiation Research, Document No. NISTIR 5632, 1995 (unpublished).

[19] J. H. Scofield, Lawrence Livermore Laboratory, Report No. UCRL 51326, 1973 (unpublished).

[20] R. Tertian and F. Claisse, *Principle of Quantitative X-Ray Fluorescence Analysis* (Heyden-Son Ltd., London, 1982).