Accurate measurement of K x-ray intensities of elements with $Z=79-82$

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Using high-resolution and high-efficiency high-purity Ge Gamma-X detectors, relative intensities of K x rays of elements with $Z=79-82$ have been very accurately (uncertainty $\approx 1\%$) determined. Comparison of the data with other experimental works and theoretical calculations have been done. It is seen that a theory with the exchange effect taken into consideration is quite adequate to explain the radiative transition rates for the elements measured in the present work.

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I. INTRODUCTION

Since the initial state (vacancy in the K shell) is same for all the components of K x rays, the relative electromagnetic transition rates from different atomic states (branching ratios) can very easily be obtained from the measurement of the relative intensities of these K x rays following the excitation of the atoms through the vacancy produced in the K shell. Therefore, the accurate measurement of these relative intensities is of great importance in testing the theoretical calculations of the atomic transition rates.

Within the uncertainty of the experimental data it has been shown by Scofield that the systematic deviation of the experimental results from his earlier calculations [1] based on a single potential Hartree-Slater description of the atom could be removed by including an "exchange correction" in the calculation [2] of the transition rates. A closer look at the comparison with the experimental data, however, shows that although the agreement is quite good in general, there are discrepancies; and accuracy of the intensity measurement could prove to be very vital for further improvement in the theoretical calculations.

Accuracy in the measurement of the K x ray intensities, on the other hand, is limited by several factors, e.g., choice of excitation of the atom, self-absorption and absorption in the materials in between the source of K x rays and the active volume of the detector, resolution and relative efficiency of the detector, etc.

For elements with high Z using thin radioactive sources as the sources of K x rays, most of the above difficulties can largely be overcome, except for the last one (relative efficiency of the detector), which for high-resolution semiconductor detectors changes with energy and thus the uncertainty in the efficiency calibration of such detectors mainly limits the accuracy of the relative K x ray intensities [3,4].

It is thus apparent that if we can have a detector with a constant efficiency over the energy range desired, the accuracy in the relative intensity measurements could greatly be improved. In our recent work [5] on the measurement of K x-ray intensities of ^{197}Au we found that the large volume high-purity Ge (HPGe) detectors with Be window (Model Gamma-X detectors), which are now routinely used for nuclear γ-ray spectroscopy, have this feature; namely, they have a large plateau ($\approx 30-90$ keV) in the efficiency vs energy curve (Fig. 1). The above characteristic of these detectors makes them very suitable for a precision measurement of relative intensity of K x rays ($\approx 70-90$ keV) of elements with $Z \approx 80$.

The atoms with $Z=79-82$ could be readily excited

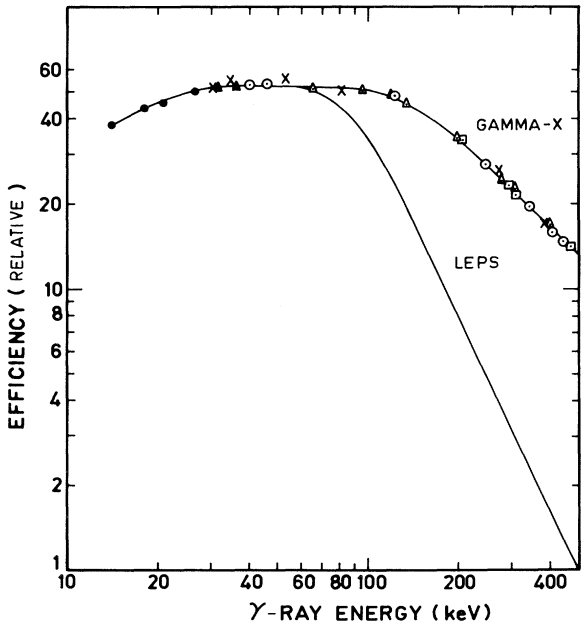

FIG. 1. Relative full energy peak efficiency curves of the Gamma-X detector (PGT) and LEPS (ORTEC). Strong lines of different sources used for calibration are shown by the following symbols: (●) ^{241}Am, (×) ^{133}Ba, (▲) ^{137}Cs, (⊙) ^{152}Eu, (△) ^{75}Se, and (□) ^{192}Ir. For the clarity of the diagram, data of only the Gamma-X detector are shown.

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TABLE I. Nomenclature of K x-ray transitions.

$1s_{1/2}$-$2p_{1/2}$	K-L_2	$K\alpha_2$	
$1s_{1/2}$-$2p_{3/2}$	K-L_3	$K\alpha_1$	
$1s_{1/2}$-$3p_{1/2}$	K-M_2	$K\beta_3$	
$1s_{1/2}$-$3p_{3/2}$	K-M_3	$K\beta_1$	
			$K\beta'_1$
$1s_{1/2}$-$3d_{3/2}$	K-M_4		
		$K\beta_5$	
$1s_{1/2}$-$3d_{5/2}$	K-M_5		
$1s_{1/2}$-$4p_{1/2}$	K-N_2		
		$K\beta_2$	
$1s_{1/2}$-$4p_{3/2}$	K-N_3		
			$K\beta'_2$
$1s_{1/2}$-$4d_{3/2}$	K-N_4		
		$K\beta_4$	
$1s_{1/2}$-$4d_{5/2}$	K-N_5		

through radioactive decays. Very thin radioactive sources can be prepared on thin mylars. Since the K x rays of these elements are in the energy range 67.0–87.3 keV, the resolution of the Gamma-X detectors enables complete separation of the K x rays: $K\alpha_1$, $K\alpha_2$, $K\beta'_1$, and $K\beta'_2$ (defined in Table I) and the uncertainty in the relative efficiency values, when measured with these detectors as mentioned above, will be less. Also at such energies self-absorption and absorption in the materials in between the source and the detector could practically be neglected.

II. EXPERIMENT AND RESULTS

A. Radioactive sources and the detectors

Radioactive sources of ^{197}Hg(64.14h), ^{204}Tl(3.78y), ^{203}Hg(46.61d), and ^{207}Bi(31.55y) were prepared by drying first a drop of insulin followed by dilute drops of the radioactive solution on thin mylar ($\approx$25 μm). After drying they were covered with very thin (3.6 μm) x-ray mylar films. The sources were placed at a relatively large distance (15–20 cm) and the strengths were such that the analog-to-digital converter dead time was practically zero. We used two large volume HPGe (Gamma-X) detectors and one small volume HPGe x-ray low-energy photon spectrometer (LEPS) detector. The particulars of these detectors are given in Table II.

B. Efficiency calibration of the detectors

The efficiency (relative) calibration of the detectors was done using relative intensity of the strong lines (with uncertainty $\sim$0.2–2%) of the following radioactive sources: ^{152}Eu(13.54y) [6], ^{192}Ir(73.83d) [7], ^{133}Ba(10.52y) [8], ^{75}Se(119.78d) [9], ^{241}Am(432.2y) [10,11], and ^{137}Cs(30.1y) [11,12]. A portion of this efficiency curve is shown in Fig. 1. The flat nature of the efficiency curve of the Gamma-X detectors in the energy region $\approx$30–90 keV was established from a repeated number of measurements. Because of this plateau, the uncertainty in the efficiency of the K x rays relative to $K\alpha_1$ (say) is practically zero and the uncertainty in the relative intensity of the K x rays is essentially due to the error in the area measurement (Sec. II C). This is contrary to the use of small volume HPGe or Ge(Li) x-ray detectors (used by previous authors) for which the efficiency falls rapidly with energy (Fig. 1). As a consequence, the uncertainty in the relative intensity measured with these detectors in this energy region is more (Table III) and thus the measured values are comparatively less reliable. For the energy region $\approx$30–60 keV, on the other hand, HPGe and Ge(Li) x-ray detectors should be preferred because in addition to the plateau in this energy region (Fig. 1) they have better energy resolution (Table II) and Si(Li) detectors having the plateau $\approx$8–15 keV [13] can be used for the accurate measurement of relative intensities in the lower-energy region.

C. Analysis of the spectra

Considering the shape of the spectrum obtained with Gamma-X detectors (Fig. 2), it is seen that the peaks: $K\alpha_2$, $K\alpha_1$, $K\beta'_1$, and $K\beta'_2$ are quite well separated so that the peak areas could be easily and accurately determined. The analysis of the spectra was done using the computer code VECSORT. In this code the peaks could either be taken as symmetric Gaussians or as asymmetric Gaussians with exponential tails on both sides and the underlying background could be taken as a polynomial of maximum order 3. There is also provision for incorporating a low-energy step simulating Compton background. The program enables one to fit multiplets of maximum 6 peaks. The fit is judged by the minimum χ^2 and the acceptable fitted background under the peaks. In analyzing the spectra we find that the main source of uncertainty is due to the position and shape of the background under the peaks. Fortunately, in the present case, as we are

Table II. Particulars of the HPGe detectors used in the present work. The measured values of the full width at half maximum (FWHM) were made for the characteristic γ rays of ^{241}Am (432.2y) and ^{109}Cd (462.6d), respectively, in columns five and six. The window material was Be for all three detectors.

Detector model	Manufacturer	Diameter-length (mm)	Active volume (cm^3)	FWHM (eV) at 59.54 keV	FWHM (eV) at 88.04 keV
Gamma-X	Princeton Gamma-Tech	50-49	90	818	852
Gamma-X	ORTEC	53.8-42.5	$\sim$90	653	698
LEPS	ORTEC	10-7	0.55	363	420

TABLE III. Intensity of K x rays of elements with $Z=79-82$ measured in the present work. Uncertainties in the last significant figures are given in parentheses. For example, 59.08(41)=59.08±0.41.

Element	Source of K x rays	Detector	Intensity $K\alpha_2(67.0)$ [a]	$K\alpha_1(68.8)$	$K\beta_1'(77.9)$[b]	$K\beta_2'(80.2)$
Au ($Z=79$)	^{197}Hg(64.14h) $\rightarrow{}^{197}$Au	Gamma-X (PGT)	59.08(41)	100	87.57(62)	9.54(11)
		Gamma-X (ORTEC)	59.34(41)	100	87.46(62)	9.96(11)
		LEPS	59.1(12)	100	86.9(28)	10.08(32)
Hg ($Z=80$)	^{204}Tl(3.78y) $\rightarrow{}^{204}$Tl		$K\alpha_2(68.9)$	$K\alpha_1(70.8)$	$K\beta_1'(80.2)$	$K\beta_2'(82.5)$
		Gamma-X (ORTEC)	59.39(41)	100	34.22(31)	10.10(11)
		LEPS	58.8(12)	100	34.5(10)	10.62(34)
Tl ($Z=81$)	^{203}Hg(46.61d) $\rightarrow{}^{203}$Tl		$K\alpha_2(70.8)$	$K\alpha_1(72.9)$	$K\beta_1'(82.4)$	$K\beta_2'(85.2)$
		Gamma-X (PGT)	59.39(41)	100	34.21(31)	10.32(11)
		Gamma-X (ORTEC)	59.15(41)	100	33.54(33)	10.23(15)
		LEPS	58.9(12)	100	35.4(11)	10.27(33)
Pb ($Z=82$)	^{207}Bi(31.55y) $\rightarrow{}^{207}$Pb		$K\alpha_2(72.8)$	$K\alpha_1(75.0)$	$K\beta_1'(84.9)$	$K\beta_2'(87.3)$
		Gamma-X (PGT)	59.84(42)	100	34.75(31)	10.56(12)
		Gamma-X (ORTEC)	59.98(43)	100	34.73(31)	10.43(12)
		LEPS	59.4(12)	100	35.6(11)	10.53(34)

[a]Energy in keV of the relevant K x ray is given within parentheses.
[b]Intensity given under this column for Au is the sum of the intensity of $K\beta_1'$ x ray and 77.35-keV γ ray. See text for explanations.

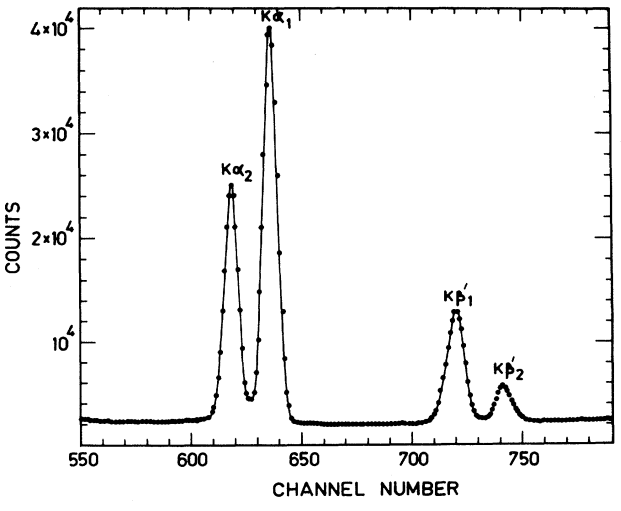

FIG. 2. Typical K x-ray spectrum of Tl in the decay of ^{203}Hg(46.61d) obtained with the PGT Gamma-X detector.

analyzing only K x-ray peaks in a limited energy region (67–87 keV) with almost a similar kind of background under the peaks, for the determination of relative intensities of these K x-rays, so long as we use the same procedure for the evaluation of peak areas we should get consistent results [14,15]. From a number of measurements the uncertainties in the peak area measurements were $\leq 0.5\%$ for $K\alpha_2$ and $K\alpha_1$, $\leq 0.7\%$ for $K\beta_1'$, and $\leq 1\%$ for $K\beta_2'$.

D. Relative intensity of K x rays

Table III summarizes the results of the present measurement. Unfortunately, the 77.35-keV γ ray in the decay of ^{197}Hg(64.14h)$\rightarrow{}^{197}$Au being very close to the energy of $K\beta_3$ (77.57 keV) of $K\beta_1'$(Au), it (77.35 keV), even with a high-resolution HPGe x-ray detector, could not be separated from $K\beta_1'$ and only the total intensity of the two could be measured. Nevertheless, we have measured the intensity of $K\beta_1'$ relative to $K\beta_2'$ from the fluorescent

TABLE IV. Comparison of the K x-ray intensities obtained in the present work with other experimental works and theoretical calculations.

Element	Reference	Intensity $K\alpha_2$	$K\alpha_1$	$K\beta_1'$	$K\beta_2'$
Au ($Z=79$)	Present work	59.21(41)	100	34.1(10)	9.77(11)
	Helmer and Gehrke [16]	62.8(5)	100	34.9(5)	9.82(13)
	Salem *et al.* [17]	58.6	100	35.1	9.35
	Scofield [1]	58.78	100	32.91	9.08
	Scofield [2]	58.74	100	34.14	9.87
Hg ($Z=80$)	Present work	59.33(41)	100	34.24(31)	10.15(11)
	de Pinho [4]	58(3)	100	33.6(22)	10(1)
	Salem *et al.* [17]	58.8	100	35.3	9.6
	Scofield [1]	59.04	100	33.09	9.25
	Scofield [2]	58.99	100	34.30	10.04
Tl ($Z=81$)	Present work	59.25(41)	100	33.96(33)	10.29(11)
	Hansen *et al.* [3]	58.0(30)	100	36.6(17)	10.1(5)
	Salem *et al.* [17]	59.1	100	35.3	9.9
	Scofield [1]	59.28	100	33.24	9.39
	Scofield [2]	59.24	100	34.44	10.23
Pb ($Z=82$)	Present work	59.84(42)	100	34.77(31)	10.50(12)
	Campbell *et al.* [18]	59.7(10)	100	34.41(47)	10.36(15)
	Salem *et al.* [17]	59.3	100	35.3	10.2
	Scofield [1]	59.58	100	33.42	9.56
	Scofield [2]	59.50	100	34.59	10.43

x rays produced by the 662-keV γ rays from an ^{137}Cs(30.1y) source bombarding a very pure gold foil (0.05 mm thick). This intensity is given in Table IV together with that of Helmer and Gehrke [16] measured by the same technique using a ^{147}Pm(2.62y) source. The results obtained with all three detectors are found to be consistent and their weighted average values are given in Table IV.

TABLE V. Ratio of experimental (present work) to theoretical (Scofield [1,2]) relative transition rates for the K x rays of the elements with $Z=79-82$. Theory (1) and theory (2) refer to the theoretical results of Scofield [1,2] without and with exchange corrections taken into account respectively.

Element	Ratio	$K\alpha_2$	$K\alpha_1$	$K\beta_1'$	$K\beta_2'$
Au ($Z=79$)	1	1.007(7)	1.0	1.036(30)	1.076(12)
	2	1.008(7)	1.0	0.999(29)	0.990(11)
Hg ($Z=80$)	1	1.005(7)	1.0	1.035(9)	1.097(12)
	2	1.006(7)	1.0	0.998(9)	1.011(11)
Tl ($Z=81$)	1	0.999(7)	1.0	1.022(10)	1.096(12)
	2	1.000(7)	1.0	0.986(10)	1.006(11)
Pb ($Z=82$)	1	1.005(7)	1.0	1.040(9)	1.098(12)
	2	1.006(7)	1.0	1.005(9)	1.007(11)

III. DISCUSSION

Table IV compares the results of the present work with other experimental works and the theoretical calculations of Scofield [1,2]. We show the earlier experimental results only with the least uncertainty reported so far. The data of Salem, Panossian, and Krause [17] are actually the values obtained from the adjustment of reported experimental values. We show these since they are very often quoted in the literature. The agreement of our results with other experimental works is in general very good. However, our intensity values have the least error and hence a detail comparison of experimental results with the theoretical calculations is possible using these intensity values.

To compare the results more clearly we have shown in Table V the ratio of experimental to theoretical relative transition rates. It is seen that considering $K\alpha_1$ and $K\alpha_2$ although there is little choice between the calculations with or without exchange effect, the intensity of $K\beta$ x rays can be explained with exchange effect taken into consideration.

IV. CONCLUSIONS

Using HPGe Gamma-X detectors accurate values of the relative intensity of the K x rays of elements with

$Z = 79-82$ have been obtained.

The experimental results achieved within an uncertainty $\approx 1\%$ are found to be explained nicely by the theoretical calculations with exchange effect into consideration.

It appears that with proper choice of the semiconductor detectors accurate values of relative K x-ray intensities of some more elements in selective energy regions can similarly be obtained.

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