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Experimental Determination of Relative Radiative Decay Rates of Vacancies in the K Shell*

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A high-resolution Ge(Li) x-ray spectrometer was used for measuring relative radiative decay rates of vacancies in the K shell of the following atoms: Au, Hg, Tl, Pb, Bi, Rn, Ra, Th, and U. In the most favorable cases, the x rays following the filling of a K vacancy by L_{II}, L_{III}, M_{II}, M_{III}, M_{IV}, M_V, N_{II}, N_{III}, $N_{IV\text{-}V}$, and $O_{II\text{-}III}$ electrons were observed and measured. The results were compared with recent relativistic calculations carried out by Scofield.

I. INTRODUCTION

The use of silicon- and/or germanium-lithium-drifted detectors is giving a new impulse to x-ray spectrometry. The very good resolutions that can be attained, the high peak-to-Compton ratios, and the possibility of obtaining very accurate efficiency-vs-energy curves render this instrument comparable or even superior to the traditional x-ray spectrometers, viz., bent-crystal systems and ioniza-

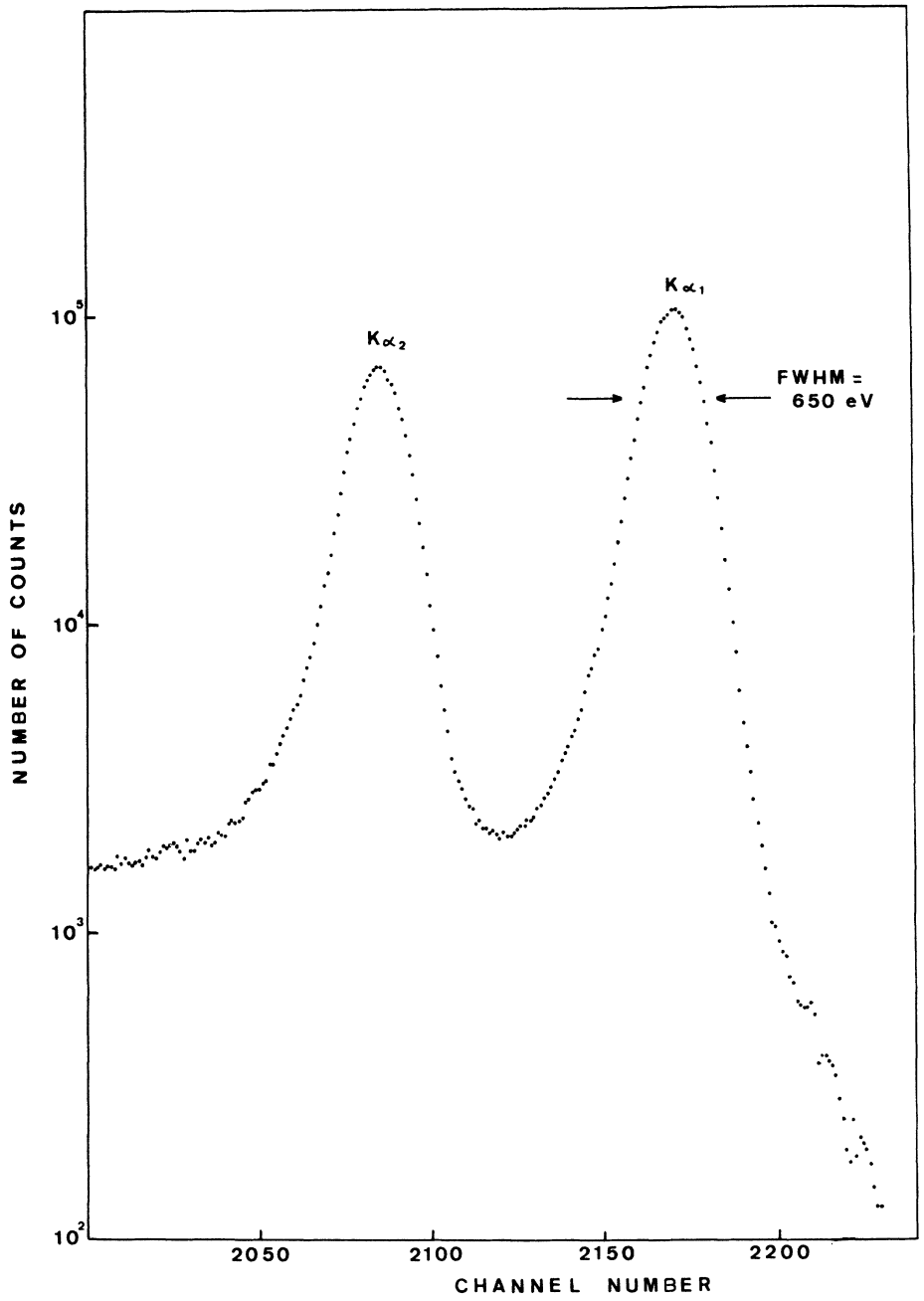

FIG. 1. KL_{II} and KL_{III} lines of thorium. The Compton continuum due to higher-energy photons was previously subtracted. The measured FWHM is 650 eV. The peak-to-valley ratio is about 70.

tion chambers. This technique can be used to study the relative radiative decay rates of vacancies in the K shell of heavy elements where there is a considerable lack of experimental information. Even theoretical calculations do not exist for elements heavier than lead.

Recently some new results appeared, both theoretical and experimental. Scofield[1] published a calculation of the rate of decay of vacancies in the K shell accompanied by the emission of x rays. In this calculation the electrons were treated relativistically and the effect of retardation was included. The electrons were considered as moving independently in a central Hartree-Slater potential. Results for some atoms of the actinide group were calculated. On the other hand, Hansen *et al.*[2] published a comprehensive experimental systematic study covering atoms up to lead. The agreement between measured values and theoretical predictions is quite satisfactory, and it is the purpose of this work to extend this study up to uranium, as well as to test more detailed predictions concerning transitions from the subshells of the M, N, and O major shells.

TABLE I. Experimental methods used for observing K x rays in the investigated atoms.

Atom	Experimental method
Au	External conversion
Hg	Internal conversion of the 412-keV γ ray from ^{198}Au
Tl	Internal conversion of the 279-keV γ ray from ^{203}Hg and of the 351-keV γ ray from ^{211}Bi
Pb	External conversion and electron capture in ^{207}Bi
Bi	Internal conversion of the 405- and 427-keV γ rays from ^{211}Pb taken in coincidence with each other
Rn	Internal conversion of γ rays following the decay of ^{223}Ra and ^{226}Ra
Ra	Internal conversion of the 236-keV γ ray from the decay of ^{227}Th taken in coincidence with the 50-keV γ ray
Th	External conversion
U	External conversion

In a previous work[3] we were faced with the problem of determinating accurately the x-ray intensities in actinium. This was done by interpolation using data obtained in neighboring atoms. These data were then restudied and completed in a systematic way and are now being presented.

II. EXPERIMENTAL PROCEDURES

Our detector was the ORTEC model 8213-08 x-ray spectrometer, consisting of a nearly cylindrical volume of lithium-compensated germanium (~0.4 cm^3) with a surface-barrier contact (~150 Å of gold) as the entrance window. The detector is kept in a cryostat with a beryllium window 0.25 mm thick. A cooled FET preamplifier, a shaping amplifier, and a base-line restorer complete the system. The detector was operated at 940 V, and the over-all system resolution measured with an Intertechnique 4096-channel pulse-height analyzer was of 700-eV full width at half-maximum (FWHM) for the 122-keV γ rays of ^{57}Co (see Fig. 1).

The gold contact was responsible for the presence of gold x rays whenever we had γ rays with enough energy to produce external conversion in gold. In many cases it was important to discount this background.

The x rays were obtained by external conversion of the 662-keV photons from a ^{137}Cs source or by nuclear decay (K-electron capture or internal conversion). The analyzing methods were those used in nuclear spectroscopy and are described elsewhere.[3]

The atoms we investigated are presented in Table I with the method of observation employed.

III. RESULTS AND DISCUSSION

The measured relative intensities of the K x-ray groups are presented in Table II and compared with the results of Scofield. The agreement is, in general, quite good but a few remarks are worthwhile. The KL_{II}/KL_{III} ratio is systematically slightly lower than the calculated[1] value.

As a rule, our measured KM_{II}/KM_{III} ratios were found to be slightly lower than the very recent values reported by Salem *et al.*[4] and they are not in contradiction with the earlier values of Beckman.[5] It was

TABLE II. Experimental and theoretical (Ref. 1) relative K x-ray emission rates.

Element	Z		L_{II}	L_{III}	M_{II}	M_{III}	$M_{IV,V}$	N_{II}	$N_{III,IV,V}$	$O_{II,III}$
Au	79	Expt.	57 ±3	100	11 ±1	21 ±2			10 ±1	
		Theor.	58.8	100	11.0	21.9			9.1	
Hg	80	Expt.	58 ±3	100	11 ±1	22 ±2	0.6 ±0.1		10 ±1	
		Theor.	59.1	100	11.1	21.4	0.63		9.3	
Tl	81	Expt.	58.5±2.5	100	11.3±0.8	22.0±1.5	0.63±0.05		9.8±0.8	
		Theor.	59.3	100	11.1	21.5	0.65		9.4	
Pb	82	Expt.	59 ±3	100	11 ±1	22.0±1.8	0.70±0.07		10.0±1.0	
		Theor.	59.6	100	11.2	21.6	0.67		9.6	
Bi	83	Expt.	59.3±2.5	100	11.6±0.7	22.0±1.4	0.70±0.05		10.2±0.6	
		Theor.[a]	59.9	100	11.2	21.7	0.65		9.8	
Rn	86	Expt.	59.8±2.5	100	11.6±0.7	22.0±1.4	0.70±0.05	8.4±0.7		1.8 ±0.2
		Theor.[a]	60.6	100	11.3	22.0	0.74	8.5		1.80
Ra	88	Expt.	59.3±2.5	100	11.6±0.7	22.5±1.4	0.70±0.05	8.6±0.7		1.8 ±0.2
		Theor.[a]	61.2	100	11.4	22.2	0.78	8.6		1.89
Th	90	Expt.	59.8±2.5	100	11.4±0.7	22.5±1.4	0.75±0.05	2.8 ±0.2	5.8 ±0.5	1.88±0.16
		Theor.	61.9	100	11.5	22.4	0.82	2.85	5.96	1.98
U	92	Expt.	60.8±2.2	100	11.5±0.6	22.0±1.2	0.75±0.05	2.9 ±0.2	6.1 ±0.4	2.00±0.15
		Theor.	62.6	100	11.6	22.6	0.86	2.89	6.10	2.07

[a]Interpolated values.

not possible to note any tendency towards an increase or a decrease of this ratio with the atomic number.

A decrease in the slope of the K_β/K_α-vs-Z curve was observed by Hansen *et al.*[2] in the lanthanide region. Tentatively this was attributed to the influence of filling the 4*f* shell. The same effect would be expected in the actinide region where the 5*f* shell is being filled. In this region, however, we have only two points, for Th and U, and here the K_β/K_α ratios are equal within experimental errors. In fact, this ratio was carefully measured in mercury, giving 0.275 ± 0.004. It rises gradually up to 0.284 ± 0.005 in thorium and 0.283 ± 0.005 in uranium. This feature of the x-ray spectrum is very interesting and should be studied in greater detail.

In these measurements the most important source of experimental error is due to the efficiency curve of the detector. The spread in energy between the KL_{II} and the KO_{III} x rays is about 14 keV in mercury and 21 keV in uranium. It is hard to reduce the experimental error due to efficiency differences below 1% for the first interval and 1.5% for the second.

An attempt was made to resolve the transitions from the M_{IV}-M_V and N_{III}-$N_{IV\text{-}V}$ subshells in the heavier elements. In Ra and U we found $KM_{IV}/KM_V = 1.1 \pm 0.3$ and $KN_{IV\text{-}V}/KN_{III} = 0.08 \pm 0.03$. The first value is in good agreement with the theory.

Note added in proof. In a recently published work Hansen *et al.* [Nucl. Phys. A153, 465 (1970)] measured relative K x-ray transition probabilities at $Z = 96$ from ^{249}Cf decay. The reported values $KL_{II}/KL_{III} = 0.626 \pm 0.006$ and $K_\beta/K_\alpha = 0.327 \pm 0.010$ are slightly below and above the calculated[1] values, respectively. This agrees with the general tendency shown by our measurements.

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Theory of Relativistic Magnetic Dipole Transitions: Lifetime of the Metastable $2\,^3S$ State of the Heliumlike Ions

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It has recently been established that the radiative lifetime of the metastable $2\,^3S$ state of helium and the heliumlike ions is determined by single-photon magnetic dipole ($M1$) transitions to the ground state, rather than the two-photon process proposed by Breit and Teller. The theory of $nl - n'l$ $M1$ transitions with $n \neq n'$ is developed in the Pauli approximation and extended to two-electron systems. Terms arising from relativistic energy corrections and finite-wavelength effects are included. The results for hydrogenic systems are shown to be identical to those obtained in the relativistic four-component Dirac formulation. The coefficients in the Z^{-1} perturbation expansion of the $1s2s\,^3S$–$1s^2\,^1S$ $M1$ transition integral are evaluated through ninth order and used to calculate the $M1$ emission probabilities from the $2\,^3S$ state of the two-electron ions up to Fe XXV. The emission probability for neutral helium is 1.27×10^{-4} sec^{-1}. The results are compared with recent solar coronal observations by Gabriel and Jordan, and with a measurement of the $2\,^3S$ state lifetime in Ar XVII by Schmieder and Marrus.

I. INTRODUCTION

The radiative lifetime of the metastable $1s2s\,^3S$ state of helium and the heliumlike ions has been a subject of controversy for some time, despite its importance in a variety of astrophysical problems.[1] Breit and Teller[2] suggested that the state decays primarily by two-photon electric dipole ($2E1$) emission, incorrectly estimating the single-photon magnetic dipole ($M1$) process to be much slower. A calculation by Mathis[3] yielded 2.2×10^{-5} sec^{-1} for the $2E1$ decay rate, a value used in the astrophysical literature for many years. However, Drake and Dalgarno[4] showed Mathis's calculation to be