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ABSTRACT

Measurements have been made of the K- and L-Auger electron spectrum of uranium, which results from the beta decay of Pa^{233}. These data are presented and compared with previously known information on the Auger transitions in heavy elements. Low energy lines, including Coster-Krönig transitions in the energy range 0.9- to 4-kev, were observed by means of a permanent-field electron spectrograph with source pre-acceleration.

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I. INTRODUCTION

In a preceding paper[1] (referred to hereafter as I) an investigation of the beta decay of Pa^{233} is described. In the course of part of this work, information was obtained about the K- and L-Auger electron transitions in uranium. Because little information has been available concerning the Auger spectra of elements with atomic number greater than 80, it was thought worthwhile to report these data. Tables I and II summarize the results of this and of previous investigations.

II. EXPERIMENTAL METHODS

Details of the experimental techniques are given in I. In brief, Pa^{233} sources were prepared by neutron irradiations of thorium metal followed by chemical purification and electrodeposition of the activity on to 0.25 mm. platinum wires. The K-Auger spectra were recorded in 180° permanent-field electron spectrographs with field strengths 50 and 100 gauss, and a 50-gauss spectrograph which employs 10 KV pre-acceleration between source and aperture[2] was used to record the L-Auger spectrum. The Pa^{233} source used in the pre-accelerator spectrograph, made by dipping a 0.25 mm platinum wire into a solution of very high specific activity, was essentially mass-free and showed little line broadening even for electrons whose initial energies were less than 1 kev.

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III. K-AUGER SPECTRUM

Table I summarizes our measurements on the K-Auger spectrum. The energies listed are the mean averages of several determinations, and the error limits are standard deviations from the mean values. Where only a single energy measurement was made the estimated error is given in brackets.

In the initial state of a KLL-Auger process there is a "hole" in the K shell, and the final state has two holes in the same or different L-subshells. The energy of the emitted electron is written as $E_{KL_pL_q} = E_K - E_{L_p} - E_{L_q}$ where the E_x's are the individual electron binding energies. Because during the Auger process the emitted electron feels the potential of an ionized atom, the electron binding energies are not expected to be equal to those obtained from critical absorption tables (e.g. Hill, Church, and Mihelich[3]) but should be somewhat higher. Following a careful study of the Auger spectrum of mercury, Bergström and Hill[4] analyzed the electron energies by introducing empirically an effective nuclear charge increment ΔZ with which to correct the tabulated[3] binding energy of the last-written shell in the formula (L_q). ΔZ is defined as

$$\frac{E^*_{L_q} - \left(E_{L_q}\right)_Z}{\left(E_{L_q}\right)_{Z+1} - \left(E_{L_q}\right)_Z}$$

where $E^*_{L_q}$ is the empirically adjusted value. They find, for mercury, that $\Delta Z = 0.6$ when $L_p = L_I$, L_{II}, or L_{III} and $L_q = L_I$ or L_{II}; and that $\Delta Z' = 0.8$ when $L_p = L_I$, L_{II}, or L_{III} and $L_q = L_{III}$.

ΔZ and $\Delta Z'$ are not entirely independent parameters, however, because it is equivalent to write KL_pL_q or KL_qL_p, and it is arbitrary whether L_p or L_q is adjusted. If $L_p = L_{II}$ and $L_q = L_{III}$, for example, $\frac{\Delta Z'}{\Delta Z} =$

$$\frac{\left(E_{L_{II}}\right)_{Z+1} - \left(E_{L_{II}}\right)_Z}{\left(E_{L_{III}}\right)_{Z+1} - \left(E_{L_{III}}\right)_Z} .$$

For Hg, $\frac{\Delta Z'}{\Delta Z}$ is computed from the binding energy tables to be 1.3; this is about equal to 0.8/0.6, which is the quotient of the individual values reported by Bergström and Hill. We find for uranium (column 3) that an average value of ΔZ for the L_I and L_{II} shells is 0.56 and for the L_{III} shell, 0.82. The ratio $\frac{0.82}{0.56}$ is 1.46, which is close to 1.5, the value calculated from the above equation for Z = 92.

A single ΔZ correction factor is sufficient to correlate the observed Auger energies of uranium to about 1 part in 10^3 if the correction is applied to the <u>average</u> of both L subshell binding energies. That is, the Auger electron energy may be defined as

$$E_{KL_pL_q} = E_K - E_{L_p} - E_{L_q} - \Delta Z_{av} \left[\frac{\left(E_{L_p}\right)_{Z+1} - \left(E_{L_p}\right)_Z + \left(E_{L_q}\right)_{Z+1} - \left(E_{L_q}\right)_Z}{2} \right] .$$

The measured K-Auger energies of uranium, plutonium[5], and protactinium[6], are given in Table I, together with values calculated with $\Delta Z_{av} = 0.6$.

The relative intensities were measured by a visual comparison method described in Table I. These intensities are equal, within our experimental error of about ± 25%, to the more accurate values for Z = 94 measured by Ewan et al.[5] The measured intensities are normalized to "percent beta decay" as discussed in I.

IV. THE L-AUGER SPECTRUM

The uranium L-Auger spectrum, recorded with the 50-gauss pre-accelerator spectrograph, is shown in Table II. The spectrum was recorded both with and without the 10 KV accelerating voltage, and in most cases the deviation between the two energy determinations was less than 0.04 kev. If a line was seen in both measurements, the average value is given in the table.

TABLE I

Uranium K-Auger Spectrum

Auger Transition	Expt. Energy (kev) This work Z=92	E calc. $\Delta Z_{av}=0.6$ / Z = 92	ΔZ Berg.[a]	Expt. Energy (kev) Ewan et al[b] Z=94	E calc. (kev) $\Delta Z_{av}=0.6$ / Z = 94	Expt. Energy (kev) Hollander et al[c] Z=91	E calc. (kev) $\Delta Z_{av}=0.6$ / Z= 91	Relative intensity: This work[d] Z=92	Relative intensity: Ewan et al Z=94	Relative intensity: Hollander et al[c] Z=91
KL_IL_I	71.69±0.10	71.68	0.58	75.18±0.015	75.13	70.05	69.98	0.10	0.13	0.13
KL_IL_{II}	72.54±0.10	72.50	0.54	76.05±0.015	75.98	70.87	70.79	0.25	0.23	0.27
$KL_{II}L_{II}$	---	73.32		76.78±0.04	76.84	---	71.60	---	0.01	---
KL_IL_{III}	76.29±0.16	76.34	0.84	80.24±0.015	80.24	74.45	74.43	0.09	0.08	0.11
$KL_{II}L_{III}$	77.13±0.13	77.16	0.80	81.06±0.015	81.09	75.21	75.24	[0.14][e]	[0.14][e]	[0.14][e]
$KL_{III}L_{III}$	---	81.00	---	85.30±0.015	85.35	---	78.88	---	0.07	---
KL_IM_I	87.96±[0.2]		1.7							
KL_IM_{II}	88.33±[0.2]		1.8							
$KL_IM_{IV,V}$	90.02±[0.2]									
$KL_{III}M_{I,II}$	92.95±[0.2]									
KM_IM_{III} ?	105.6±[0.2]									

(a) Reference 4.

(b) Reference 5.

(c) Reference 6.

(d) Error limits are estimated to be ± 25%.

(e) Normalized to 0.14.

TABLE II

Uranium L-Auger Spectrum

Auger Transition	Relative Intensity[a] Z=80 Nall et al[b]	Z=83 Sujkowski and Slätis[c]	Z=92 This work	Energy (kev) Z = 92 Observed (Albouy and Valadares)[e]	Observed (This work)	Calculated
$L_1L_3M_5$			vw(b)		0.96	0.94
$L_2L_3N_1$			w		2.24	2.28
$L_2L_3N_3$			wm		2.64	2.70
$L_2L_3O_1$						3.44
$L_1L_3N_3$			w		3.48	3.52
$L_2L_3O_2$						3.52
$L_2L_3O_4$			w(b)		3.71	3.68
$L_1L_3N_4$						3.78
$L_3M_1M_2$	1 (?)					6.25
$L_3M_2M_2$						6.62
$L_3M_1M_3$	1.5	vw	vw(b)		7.17	7.18
$L_3M_2M_3$	4.0	w	(?)		7.47	7.55
$L_3M_1M_4$		vw	(?)		7.71	7.76
$L_3M_1M_5$		vw				7.95
$L_3M_2M_5$	0.76	vw	(?)		8.22	8.32
$L_3M_3M_3$	6.45	w	w(b)		8.42	8.43
$L_3M_2M_4$			--		--	8.13
$L_3M_3M_4$	5.6	m	w(b)	8.86	8.98	9.01
$L_3M_3M_5$	5.7	m	w(b)	9.08	9.12	9.20
$L_3M_4M_4$	2.1	vw				9.59
$L_3M_4M_5$	11.0	vs	m	9.66	9.72	9.78
$L_3M_5M_5$	(8.6)	s	wm	9.85	9.93	9.95
$L_2M_1M_2$			(?)		10.02	10.03
$L_2M_2M_2$	(0.5)					10.40

TABLE II (continued)

$L_1M_1M_1$			vw(b)	10.47	10.47
$L_3M_2N_1$					10.48
$L_3M_2N_2$	0.33				10.66
$L_3M_1N_5$	--	vw			10.83
$L_1M_1M_2$	--	--	vw(b)	10.88	10.85
$L_3M_2N_3$	(0.66)	--			10.90
$L_2M_1M_3$	--	vw			10.96
$L_3M_2N_{4,5}$	(1.6)				11.16[d]
$L_2M_2M_3$	5.0	w	(masked by 28-L_{III})		11.33
$L_3M_1O_4$	--	--			11.51
$L_3M_3N_2$	0.59	--	vw	11.54	11.54
$L_2M_1M_4$	--	--			11.54
$L_3M_2N_{6,7}$	--	w			11.57
$L_3M_3N_3$	(1.7)				11.78
$L_1M_1M_3$	(0.3)				11.78
$L_2M_2M_4$	1.7	w			11.91
$L_2M_3M_3$		--			12.21
$L_3M_{4,5}N_{1,2}$	(0.4)				11.94[d]
$L_3M_3N_{4,5}$	(1.4)				12.04
$L_2M_2M_5$	(2.4)	w	(masked by 17-M_{II})		12.10
$L_1M_1M_4$	(0.4)	--			12.36
$L_3M_4N_3$	(1.5)	--	vvw	12.39	12.36
$L_3M_3N_6$	--	--			12.45
$L_1M_1M_5$	(0.4)				12.55
$L_3M_3O_2$			vvw	12.59	12.60
$L_1M_2M_4$	(0.2)	vw			12.73

TABLE II (continued)

$L_2M_3M_4$	(2.2)	m	wm(b)	12.64	12.80	12.79
$L_3M_5N_4$	(4.3)	--		--		12.70
$L_1M_2M_5$	(0.2)	vw	(masked by 17-M_{III})			12.92
$L_2M_3M_5$	(1.0)					12.98
$L_3M_4N_{6,7}$	0.26	vw				13.03
$L_3M_4O_2$	--	--	vw		13.16	13.18
$L_3M_5N_{6,7}$	1.3	m				13.20
$L_3M_4O_4$	--	--	vw		13.34	13.34
$L_2M_4M_4$	(4.4)	m				13.37
$L_2M_4M_5$	7.2	vs	wm	13.43	13.56	13.56
$L_1M_3M_{4,5}$	(0.5)					13.61[d]
$L_2M_5M_5$	(1.5)	vw				13.73
$L_1M_4M_4$	0.62					14.19
$L_2M_1N_3$	--	--	w		14.32	14.31
$L_1M_4M_5$	0.81	w				14.38
$L_2M_2N_2$	(0.3)					14.44
$L_1M_5M_5$	(0.2)					14.55
$L_2M_1N_5$	--	w	vvw		14.67	14.61
$L_2M_2N_3$	0.78	--				14.68
$L_2M_1N_{6,7}$	1.0	w	(?)		14.98	14.98
$L_2M_2N_5$		--				14.98
$L_2M_3N_1$						15.14
$L_3N_1N_{6,7}$	--	w	vw		15.34	15.31
$L_1M_1N_4$	--	vw				15.39
$L_2M_3N_2$	0.67	vw				15.32
$L_2M_3N_3$						15.56

TABLE II (continued)

$L_3N_4N_5$	(0.4)	--	vvvw	15.60	15.60
$L_2M_4N_1$	1.1				15.72
$L_2M_3N_4$		vw	(masked by 17-N_I		15.82
$L_2M_5N_1$	(0.5)	w			15.89
$L_3N_2N_5$	--				15.11
$L_3N_3N_3$	(0.2)				15.04
$L_3N_{4,5}N_{6,7}$	(0.2)				15.97[d]
$L_2M_{4,5}N_3$	0.95				16.14[d]
$L_3N_6N_6$	0.14		vvw	16.35	16.36
$L_1M_2O_4$			(??)	16.48	16.48
$L_2M_5N_4$	(3.4)	w			16.57
$L_2M_4N_{6,7}$	(1.2)	w			16.81
$L_3N_5N_{6,7}$	--				16.01
$L_1M_3N_{6,7}$		vw			17.05
$L_2N_4N_{4,5}$	0.27	w	(masked by 40-L_{II})		19.34
$L_2N_5N_5$	--				19.42
$L_2N_{4,5}N_{6,7}$	0.1				19.75[e]

(a) Symbols are: w = weak, m = medium, s = strong, v = very, (b) = broad, ? = questionable.

(b) Reference 9. Parentheses around intensity values indicate uncertainty due to incomplete resolution of the lines.

(c) Reference 8.

(d) Of the choices given by Nall et al, only the lowest energy is listed here.

(e) Reference 10.

Assignments of lines to particular L-Auger transitions were made solely on the basis of energy agreement with values calculated from electron binding energies. An effective charge increment (ΔZ) of unity was assumed for the binding energy of the last written subshell in the formula LXY; the validity of this assumption has been discussed by Burhop,[7] and it has been used with good results by Sujkowski and Slätis[8] for calculation of the bismuth L-Auger transition energies. Because of the large number of possible Auger transitions per energy interval, these assignments are of course not certain.

Visual estimates of the strengths of the lines (uncorrected for geometry or efficiency) are also given in Table II, and the intensities reported by Sujkowski and Slätis for bismuth and by Nall et al[9] for mercury are also given, as well as data on six L-Auger lines of uranium reported by Albouy and Valadares.[10] There is a definite correspondence between the intensities of the four sets of data listed; in only a few cases were lines of reasonable intensity not observed by all three groups (Albouy and Valadares observed only the 6 most intense lines). It should be noted that lines which are close lying or unresolved in the spectrum of one element may not be near one another in the spectrum of a different element; this, of course, depends on how the binding energies vary with Z. For this reason, in Table II some of the lines are displaced from a strict order of increasing energy in order to compare the uranium spectrum with those of mercury and bismuth.

Eight of the electron lines listed in Table II have been assigned as Coster-Krönig transitions. In these transitions, which are designated by L_pL_qY, an initial L-subshell vacancy is transferred to another L-subshell with the emission of an electron from a higher (Y) shell. As discussed by Burhop, only those transitions are possible for which $(E_{L_p} - E_{L_q}) > E_Y$. In uranium, transitions of the type $L_IL_{II}M$ and $L_{II}L_{III}M$ (except M_V) are energetically forbidden. $L_IL_{III}M_I$

and $L_I L_{III} M_{II}$ are forbidden also, but $L_I L_{III} M_{III,IV,V}$ are allowed, and a transition which corresponds in energy to $L_I L_{III} M_V$ is seen in the Pa^{233} Auger spectrum.

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