Conversion Electron Spectra of Cm^{242} and Cm^{244}†

W. G. SMITH* AND J. M. HOLLANDER
Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California
(Received August 10, 1955)

The conversion electron spectra of Cm^{242} and Cm^{244} have been studied by means of two 180° photographic recording beta-ray spectrographs. Conversion coefficient ratios in the L and M subshells have been measured for several $E2$ transitions in the decay of these nuclides, and the values so obtained compared with theoretical ratios. Accurate energy determinations of the first three excited states in Pu^{238} are reported, and the validity of the Bohr-Mottelson rotational energy formula, including the vibration-rotation interaction term to describe these states, is discussed. A more accurate value is reported for the energy of the first excited state in Pu^{240}. The decay of I^{131} is discussed briefly.

I. INTRODUCTION

THE study of low-energy conversion electron spectra among elements of high atomic number is especially attractive because here the energy separation between atomic levels is large and one is able, at moderate resolution, to obtain information regarding internal conversion in the several L, M, and N subshells for a given gamma ray. Such information can be of value not only insofar as the precise measurement of gamma ray energies is always helpful in the solving of complex decay schemes, but also because the determination of the relative probabilities of internal conversion in the various subshells can lead to an elucidation of the multipolarities of the transitions; or, in those cases where theoretical calculations of the conversion coefficient ratios have not yet been made, one can hope that experimental measurements with gamma transitions of known multipolarities will provide some basis for further theoretical work.

The systematic behavior of the excited states of even-even nuclei in the heavy element region has been a subject of much interest and relevance to current nuclear models; thus it is also desirable to obtain a more detailed knowledge of the energies of these states than is presently enjoyed.

In this paper we report on the following: (1) precision measurements of the energies of the first excited states in Pu^{238} and Pu^{240}, (2) determination of the L, M, and N shell internal conversion ratios for $E2$ transitions in the above nuclei and comparison with theoretical predictions, and (3) measurement of the energies of the second and third excited states in Pu^{238} and a comparison with the energies expected for these states from the Bohr-Mottelson rotational model.

II. INSTRUMENT DESCRIPTION

Two flat 180° spectrographs of the permanent magnet type have been constructed, which employ photographic recording of the conversion lines. Mihelich[1] and Slätis[2] have pointed out the advantages of this type of instrument for conversion electron studies: (a) moderately high resolution is readily available, (b) very good stability of the field is obtained, provided that the magnet temperature is held constant, and (c) the photographic film records line positions accurately and integrates the entire spectrum simultaneously, providing a permanent record of primary data. Disadvantages of this method are principally that: (a) photographic film is an extremely nonlinear exposure recording device and can be used for obtaining relative intensities of lines only after extensive calibrations and (b) to approach optimum resolution with these instruments one needs thin sources deposited on fine wires, a condition which is often difficult to meet in practice.

The two spectrographs used in this work have field strengths of ~53 gauss and ~99 gauss, respectively. Their vacuum chambers and cameras are a modification of the Brookhaven design.[3]

The maximum radius of curvature is about 20 cm, which corresponds to maximum electron energies in the two instruments of ~80 and ~275 kev, respectively. No provision is made in the cameras for rapid source placement or film removal because only relatively long-lived isotopes are being studied at the present time.

Eastman No-Screen x-ray film on glass backing $\frac{3}{4}\times 15\times 0.04$ inch has been used to record the lines.

III. SOURCE PREPARATION

The spectrograph sources were prepared by electrodeposition of the active materials upon 10-mil or 14-mil platinum wires using a procedure suggested by B. G. Harvey, in which $M(OH)_3$ (where M is the actinide cation) is deposited on the wire cathode of an electrolysis cell using NH_4HSO_4 at a pH of 3.6 as the electrolyte. A drawing of the electrolysis cell is shown in Fig. 1. The active wires are mounted in a special source holder whose position in the camera could be adjusted and reproduced within several mils.

† This work was performed under the auspices of the U. S. Atomic Energy Commission.
* Present address: Physics Department, Indiana University.
[1] J. W. Mihelich, Phys. Rev. 87, 646 (1952).
[2] H. Slätis, Arkiv Fysik 6, 415 (1953).
[3] We are indebted to Dr. G. Friedlander and Dr. J. W. Mihelich for their kindness in furnishing us with drawings of their instruments. Dr. M. I. Kalkstein and Dr. I. Bergström were also of great assistance in the adaptation of these designs to the present use.

TABLE I. Calibration standards.[a]

Standard	Energy or $H\rho$	Reference
ThB	A line, $H\rho=534.11$	b
	F line, $H\rho=1388.5$	b,c,d
	I line, $H\rho=1754.0$	b,c,d
I^{131}	80.164 kev	e
	284.307 kev	e
Am^{241}	33.20 kev	f
	43.46 kev	f
	59.57 kev	f

[a] Electron binding energies used both in calibration and in subsequent experiments were taken from the table by Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).
[b] D. I. Meyer and F. H. Schmidt, Phys. Rev. **94**, 927 (1954). Internal consistency of the Cm^{242} and Cm^{244} conversion line energies (using the binding energies of Hill *et al.*) could not be obtained when the Meyer and Schmidt value for the $H\rho$ of the ThB A line was used. On the other hand, the Am^{241} gamma-ray energies given by Day did provide such consistency. It has been called to our attention that Lindström is currently examining the ThB A line at resolution greater than 0.1%, and that he finds it to be a doublet. From our work at ~0.1% resolution, the doublet is not resolved, and an average value of 534.5 gauss cm is suggested.
[c] G. Lindström, Phys. Rev. **83**, 465 (1951).
[d] H. Craig, Phys. Rev. **85**, 688 (1952).
[e] H. C. Hoyt and J. W. M. DuMond, Phys. Rev. **91**, 1027 (1953).
[f] P. P. Day, Phys. Rev. **97**, 689 (1955).

IV. INSTRUMENT CALIBRATION

Calibration of the spectrographs consisted in the determination of the effective magnetic field at various ρ values, using accurately known gamma-ray energies of I^{131} and Am^{241} and conversion line energies of ThB. The energies used for calibration are given in Table I. The effective field in this instrument is known to about 0.1% from $\rho=9$ cm to $\rho=19$ cm; the field in the 99-gauss spectrograph is known to a similar accuracy.

As part of the calibration procedure, a series of accurately machined notches was cut into the film holder so as to render a permanent set of light and dark reference edges exposed into each photographic plate. With this device, similar to that described by Slätis,[2] the notch distances and other dimensions pertinent to a determination of ρ are all constants of the spectrograph camera and need be measured only once. In subsequent experiments only the distance of each electron line to the nearest notch shadow need be measured; in no case will this distance be greater than 2 cm, the distance between notch edges. For the energy determinations we have used an ordinary x-ray diffraction film comparator, with which the distances could be measured reproducibly to ±0.01 cm.

The resolution (full width at half-maximum) $\Delta\rho/\rho$ obtained in the 53-gauss spectrograph for the I^{131} (80 kev) K line is 0.17% and for the L_I line 0.13%; in the 99-gauss spectrograph the best resolution has been 0.17%. The instruments are located in a room whose temperature variation is only a few degrees Fahrenheit; better thermostating is being planned, so that loss in resolution due to temperature variation will be minimized.[2]

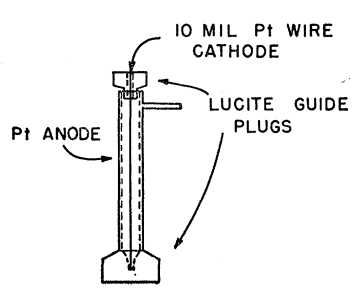

FIG. 1. Drawing of electrolysis cell used for preparing wire sources.

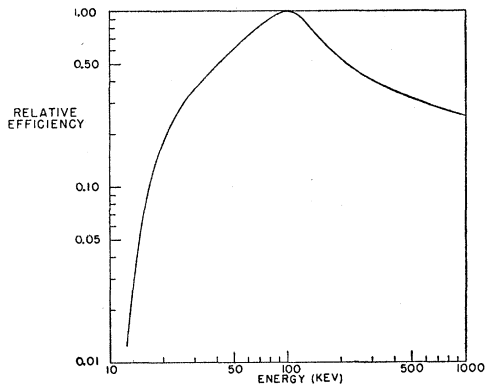

FIG. 2. Photographic efficiency *vs* electron energy (kev).

V. FILM CALIBRATION AND RELATIVE INTENSITY DETERMINATIONS

In order to determine relative intensities of lines one must study several features of the photographic action of electrons: (a) the density *vs* exposure relation, (b) the dependence of line intensity on ρ, and (c) the photographic efficiency as a function of electron energy.

Previous experimenters have shown that the D *vs* E curve is linear to at least $D=1$; we have verified this experimentally for the film used in this study.

We have assumed, with Slätis,[2] that the intensity is an inverse function of ρ. Rutledge *et al.*[4] have found that this approximation is not exact over a wide range of ρ; however, the present experiments are concerned with intensity ratios of lines which are very close together in ρ. Over such small changes in radius, a simple inverse relationship should be valid.

For the photographic efficiency as a function of electron energy, we have normalized and combined the curves of Cranberg and Halpern[5] and Dudley[6] to obtain the curve shown in Fig. 2. We have assumed

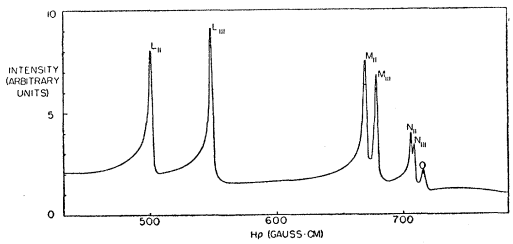

FIG. 3. Densitometer plot of Cm^{242} lines.

[4] Rutledge, Cork, and Burson, Phys. Rev. **86**, 775 (1952).
[5] L. Cranberg and J. Halpern, Rev. Sci. Instr. **20**, 641 (1949).
[6] R. A. Dudley, Nucleonics **12**, 24 (1954).

TABLE II. Energies of Cm^{242} conversion electrons, in kev.

Exp. No.	Standard on same wire	Conversion subshell and electron energies							
		L_{II}	L_{III}	M_{II}	M_{III}	N_{II}	N_{III}	O_{III}	M_I(?)
a	I^{131}	21.85	26.05	38.55	39.55	42.73	43.00	···	···
b	I^{131}	21.87	26.06	38.60	39.62	42.75	43.01	43.90	38.23
c	ThB	21.86	26.05	38.56	39.55	42.70	42.95	43.85	···
d	···	21.87	26.04	38.53	39.56	42.70	42.96	43.88	···
Average electron energy		21.86	26.05	38.56	39.57	42.72	42.98	43.88	···
Binding energy		22.25	18.06	5.56	4.56	1.38	1.13	0.23	5.93
Transition energy		44.11	44.11	44.12	44.13	44.10	44.11	44.11	44.16

that the glass-backed and celluloid-backed No-Screen x-ray films behave similarly.

The relative intensity of an electron line is given by:

$$I = A\rho/\eta,$$

where A = area under line, ρ = radius of curvature, and η = efficiency of film. In our experiments, the area A (as obtained from a densitometer plot of the blackening) was determined by the method of Mladjenovic and Slätis.[7]

It should be emphasized that many of the uncertainties usually associated with photographic relative intensitity determinations, for example, the fraction of total line area located in the (unscattered) peak, disappear when one studies relative intensities of lines whose energies and radii of curvature are very close. These effects can then be considered as constant.

VI. EXPERIMENTAL RESULTS

A. Iodine-131

I^{131} was used primarily as a calibration standard, but some observations made with very active sources of this isotope are perhaps worth reporting.

In addition to the K, L_I, M_I, and N lines of the well-known 80.16-kev transition, we have observed in low intensity the L_{II} line of this transition, previously unreported. Its intensity, relative to that of the L_I line, appears from the densitometer tracing to be roughly 0.10. This figure had previously been given as an upper limit by Mihelich. This transition is known to be a magnetic dipole, and the theoretical calculations of Rose *et al.*[8] predict an L_{II}/L_I ratio (for $Z=55$, $E=75$ kev) of 0.07.

Three other novel lines were also seen. These were the K and L_I lines of a 163.8±0.1-kev transition, and the K line of a 177.1±0.1-kev transition. The 164-kev level has been assigned to the 12-day Xe^{131m_2} isomeric level, and its energy was previously determined by Bergström[9] to be 163.9 kev. The 177-kev K line has been sporadically reported[10,11] but it does not fit into the presently accepted[12] decay scheme of I^{131}.

TABLE III. Relative intensities of conversion lines from 44.1-kev transition of Cm^{242}. Theoretical value: $L_{II}/L_{III}=1.2$.

Exp.	L_{II}/L_{III}	M_{II}/M_{III}	N_{II}/N_{III}	$L_\Sigma/M_\Sigma/N_\Sigma$
A	1.23	1.25	~1	2.5/1.0/0.3
B	1.27	1.22	~1	2.9/1.0/0.3
C	1.16	1.20	~1	2.4/1.0/0.3

B. Curium-242

The energy of the first excited state of Pu^{238}, populated by the alpha decay of Cm^{242}, has been measured in several ways. O'Kelley[13] and Passell[14] using a double-focusing beta spectrometer observed conversion electrons corresponding to a gamma ray of 44.9 kev. Asaro *et al.*,[15] using a magnetic alpha spectrograph, have studied the complex alpha spectrum of Cm^{242} and have found an energy difference between the most prominent alpha groups which corresponds to an energy of 44.3 kev for the first excited state of Pu^{238}. More recently, Rasmussen, Slätis, and Passell[16] have studied the beta decay of Np^{238} using a double-focusing spectrometer at 0.5% resolution and have found the energy of the first excited state of Pu^{238} to be 44.0 kev.

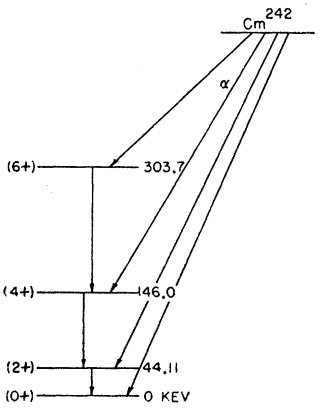

FIG. 4. Cm^{242} decay scheme.

Our present data, from four separate one-day

[7] M. Mladjenovic and H. Slätis, Arkiv Physik 8, 65 (1954).

[8] Rose, Goertzel, and Swift, privately circulated tables of conversion coefficients calculated with relativistic, screened electron wave functions.

[9] I. Bergström, Arkiv Fysik 5, 268 (1954).

[10] I. Bergström and R. D. Hill, Arkiv Fysik 8, 21 (1954).

[11] J. M. Cork, Nucleonics 7, 24 (1950).

[12] Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953).

[13] G. D. O'Kelley, PhD thesis, University of California Radiation Laboratory Report UCRL-1243, May, 1951 (unpublished).

[14] T. O. Passell, PhD thesis, University of California Radiation Laboratory Report UCRL-2528, March, 1954 (unpublished).

[15] Asaro, Thompson, and Perlman, Phys. Rev. 92, 694 (1953); and private communication.

[16] Rasmussen, Slätis, and Passell, Phys. Rev. 99, 42 (1955).

TABLE IV. Conversion electrons from decay of second and third excited states of Pu^{238}.

Electron energy (kev)	78.8	79.6	83.8	96.3	97.3	100.7	135.5	139.6
Relative intensity	~0.2	1.4	1.0	···	···	···	···	···
Subshell	L_I	L_{II}	L_{III}	M_{II}	M_{III}	$N_{II}+N_{III}$	L_{II}	L_{III}
Binding energy (kev)	23.1	22.3	18.1	5.6	4.6	1.2	22.3	18.1
Transition energy (kev)	101.9	101.9	101.9	101.9	101.9	101.9	157.8	157.7

exposures in the 53-gauss spectrograph, are given in Table II. From these experiments a best value for the energy of the first excited state of Pu^{238} is given as 44.11±0.05 kev. The absolute limit of error placed on this measurement is 0.02 kev smaller than the total maximum error possible assuming errors of ±0.01 cm and ±0.05 gauss in the radii of curvature and effective magnetic field measurements, respectively. If one considers each conversion line energy determination as a separate measurement, a total of 27 measurements were made of this gamma ray. In addition, on some exposures the internal standards ThB and/or I^{131} were placed on the same wire with the Cm^{242} providing lines of known energy very close to the unknowns. This error does not include absolute binding energy errors, but these might be expected to be very small for the M, N, and O shells.

This transition, like most transitions from the first excited state to ground state in even-even nuclei, is of multipole order $E2$. It has been so characterized by measurements of the total conversion coefficient[15] and from the L subshell ratios. Previously reported values for the L_{II}/L_{III} intensity ratio as studied from the beta decay of Np^{238} are Freedman *et al.*[17] 1.91, Passell[14] 1.44, and Rasmussen *et al.*[16] 1.37. From the decay of Cm^{242}, Passell[14] obtained a value of 1.26. We have measured the L and M subshell ratios for this transition and have estimated the N ratios. A linear tracing of one of the densitometer plots is shown in Fig. 3. The relative intensity determinations, which are summarized in Table III, indicate that the L_{II}/L_{III} and M_{II}/M_{III} ratios for this transition are identical, to within about 10%, and have the value 1.2±0.1. M_{IV} and M_V conversion is not observed, and M_I conversion is extremely weak. Rasmussen *et al.*[16] also observed identical L_{II}/L_{III} and M_{II}/M_{III} ratios.

Our M ratios should be somewhat more reliable than our L ratios for this transition because the parameters ρ and η remain practically constant over the small energy range of the M lines and hence any uncertainty in their absolute values will cancel out in the ratio; however, in the region of the L electron energies the slope of the efficiency-energy curve is steep and therefore the accuracy of the L_{II}/L_{III} ratio depends strongly on the accuracy of this calibration. The N_{II} and N_{III} lines were not completely resolved, but their intensities were estimated visually to be about equal.

The theoretical L_{II}/L_{III} ratio given by extrapolation or interpolation from Rose *et al.*[8] and Gellman *et al.*[18] for an $E2$ transition of 44 kev in $Z\sim94$ is 1.2. The L_{II}/L_{III} ratio obtained in these experiments is in good agreement with their theoretical value. There are at present no theoretical calculations of M subshell conversion coefficients with which to compare the experimental results other than the "threshold" calculations of Church and Monahan[19] which indicate that $E2$ radiation converts primarily in the M_{II} and M_{III} shells. It appears that such calculations would be of value especially in the study of higher multipoles where M_{IV} and M_V conversion becomes important.

It was desirable to study also the conversion lines from the higher excited states of Pu^{238}. Since the population of the second and third excited states from the alpha decay of Cm^{242} is only 0.035% and 0.009%, respectively,[15] an exposure of one month's duration was taken in the 99-gauss spectrograph in an attempt to record these weak lines. Although many "extra" lines appeared on this plate because of very small

TABLE V. Energies of Cm^{244} conversion electrons, in kev.

	Electron energies								
Exp. no.	L_I	L_{II}	L_{III}	M_I	M_{II}	M_{III}	N_{II}	N_{III}	O
a	19.76	20.60	24.80	36.93	37.31	38.28	41.49	41.75	42.63
b	···	20.64	24.84	···	37.36	38.35	41.57	41.79	···
c	19.77	20.60	24.80	···	37.32	38.29	41.49	41.74	42.65
Average electron energy	19.77	20.61	24.81	36.93	37.33	38.31	41.52	41.76	42.64
Subshell binding energy	23.10	22.25	18.06	5.93	5.56	4.56	1.38	1.13	~0.23
Transition energy	42.87	42.86	42.87	42.86	42.89	42.87	42.90	42.89	42.87

[17] Freedman, Jaffey, and Wagner, Phys. Rev. **79**, 410 (1950).
[18] Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).
[19] E. L. Church and J. E. Monahan, Phys. Rev. **98**, 718 (1955).

TABLE VI. Relative intensities of conversion lines from 42.9-kev transition of Cm^{244}. Theoretical value: $L_{II}/L_{III}=1.2$.

Exp.	L_{II}/L_{III}	M_{II}/M_{III}	N_{II}/N_{IV}	$L_\Sigma/M_\Sigma/N_\Sigma$
A	1.08	1.15	~1	2.5/1.0/0.3
B	1.13	1.06	~1	2.4/1.0/0.3

amounts of Cm^{243} and Am^{241} in the sample, a series of lines could be assigned to a 101.9±0.3-kev transition and a 157.7±0.5-kev transition in Pu^{238}. The electron energies and their assignments are given in Table IV, and the level scheme is shown in Fig. 4.

We have measured the relative intensities of the subshell electron groups from the 101.9-kev transition, with the result $L_I/L_{II}/L_{III}=\sim 0.2/1.4/1.0$. These ratios confirm the predominantly $E2$ nature of this transition and check with the theoretical values of Rose *et al.*[8] and Gellman *et al.*[18] for $E=100$ kev and $Z\sim 94$, which are $L_I/L_{II}/L_{III}=0.14/1.8/1.0$. The subshell ratios indicate an upper limit for possible $M1$ mixing of about 15%. As was the case with the 44-kev transition, the M subshell ratios are here similar to the L ratios; the M lines were weak, but a rough value is $M_{II}/M_{III}=1.5/1.0$. Also $L_\Sigma/M_\Sigma\approx 3$.

The excited states of Pu^{238} form a band which is characteristic of the heavy even-even nuclides.[20] These levels are described by the unified nuclear model of Bohr and Mottelson[21] as rotational states of a deformed nucleus. The configurations expected are $I=(0+)$, $(2+)$, $(4+)$, etc., and the energies are approximately equal to $(\hbar^2/2\Im)I(I+1)$, where $\Im$ is the effective moment of inertia of the deformed nucleus. Deviations from this strict $I(I+1)$ dependence are predicted[21] for the higher excited states where centrifugal distortions slightly increase the equilibrium nuclear deformation; the correction term is of the form $\Delta E=-2(1/\hbar\omega)^2(\hbar^2/\Im)^3I^2(I+1)^2$, where $\hbar\omega$ is the vibrational quantum.

[20] I. Perlman and F. Asaro, Ann. Revs. Nuclear Sci. 4, 157 (1954).

[21] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 27, No. 16 (1953).

The measurements reported here for the energies of the $(2+)$, $(4+)$, and $(6+)$ states in Pu^{238} allow two independent calculations to be made of the constants A and B in the Bohr-Mottelson equation $E=AI(I+1)-BI^2(I+1)^2$. From the energies of the $(+2)$ and $(+4)$ states one has $A=7.37$ kev and $B=0.0035$ kev; from the $(2+)$ and $(6+)$ states one finds $A=7.37$ kev and $B=0.0033$ kev. This close agreement, unless fortuitous, indicates that the Bohr-Mottelson formulation accurately describes the energies of the first four rotational states in Pu^{238}. With the above constants, the equation may be used to predict the energy of the fifth rotational state, the $(8+)$ state, to be at 513.1 kev.

C. Curium-244

A sample of Cm^{244} containing a few percent Cm^{242} was plated onto a 10-mil platinum wire, and several exposures were made in the 53-gauss spectrograph. The energies of the electron lines so obtained are summarized in Table V and indicate that the first excited state of Pu^{240} has an energy of 42.88±0.05 kev. The Cm^{242} in this sample served as an internal standard.

Relative intensities for L, M, and N subshell conversion were measured, with the results given in Table VI. These results are essentially the same as obtained with Cm^{242} and again support the theoretical L_{II}/L_{III} ratios of Rose *et al.*[8] and Gellman *et al.*[18] for an $E2$ transition. L_I and M_I conversion lines were observed but were so weak that their relative intensities could not be measured quantitatively.

VII. ACKNOWLEDGMENTS

It is a pleasure to express our appreciation to the many members of the Chemistry Group for the assistance which they have given us. In particular, I. Perlman, J. O. Rasmussen, H. P. Robinson, F. Asaro, M. I. Kalkstein, B. G. Harvey, S. Thulin, and J. G. Conway have been especially helpful. The Cm^{242} was kindly supplied by F. Asaro, and the Cm^{244} by A. Chetham-Strode. Finally, we wish to thank Professor G. T. Seaborg for his interest and encouragement.