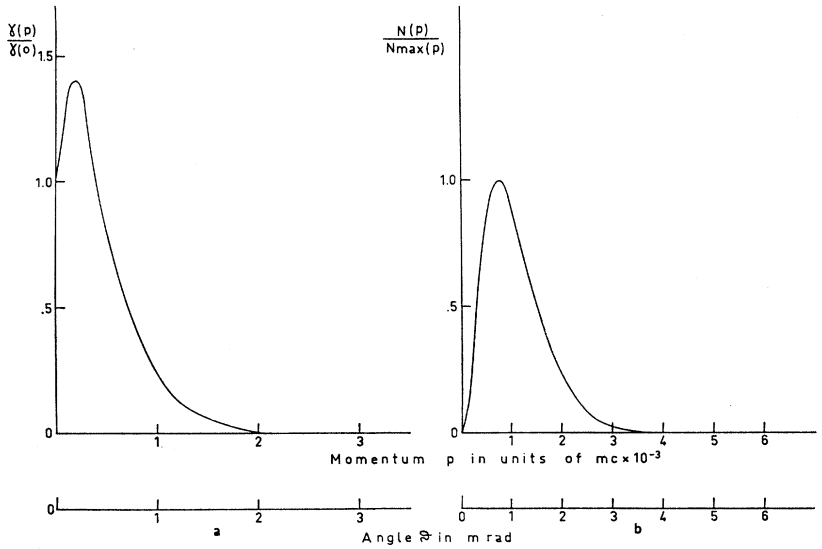

FIG. 2. (a) Curve for the angular correlation of the pair of the emitted photons. In the case of positronium we have a delta function $\delta(\mathbf{p})=\delta(\mathbf{k}_1+\mathbf{k}_2)$, corresponding to the fact that the angle between the directions of the emitted photons is π. The presence of a spectator electron in $e^-e^+e^-$, which may carry off part of the total momentum, causes the most probable angle between annihilation photons to be different from π by an angle ϑ of about 2×10^{-4} rad. Thus ϑ measures the departure from π of the angle between the directions of the two annihilation photons. (b) Momentum distribution curve.

positronium ion can exist as a separate, well-defined entity, available space comparable to the positronium-ion dimensions is needed. The gases, for example, provide such media. Among the real metals, probably, Cs, Rb, and K also have room enough to form the positronium ion.

ACKNOWLEDGMENTS

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L_2-Subshell Yield Measurements in Pu^{240}, U^{236}, and U^{234}

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The fluorescence, Auger, and Coster-Kronig yields of the L_2 subshells in Pu^{240}, U^{236}, and U^{234} have been determined from measurments on L x-rays emitted following α decay of Cm^{244}, Pu^{240}, and Pu^{238}. The techniques employed include α–L x-ray coincidence counting using a silicon detector and NaI(Tl) crystal, proportional-counter spectrometry and, in the case of Pu^{238}, the use of a curved-crystal spectrograph. The following results were obtained: Pu^{240}, $\omega_2=0.466\pm0.023$, $a_2=0.11\pm0.08$, $f_{23}=0.42\pm0.08$; U^{236}, $\omega_2=0.535\pm0.042$, $a_2=0.09\pm0.11$, $f_{23}=0.37\pm0.07$; U^{234}, $\omega_2=0.497\pm0.035$, $a_2=0.07\pm0.07$, $f_{23}=0.43\pm0.06$. The measured fluorescence yields show close agreement with those calculated on a semiempirical basis by Listengarten, but the Auger and Coster-Kronig yields are, respectively, much smaller and much larger than predicted.

1. INTRODUCTION

ALTHOUGH the Auger effect plays a fundamental role in the reorganization of the atomic electrons following inner-shell ionization of the atom, it was for many years neglected as a subject for quantitative study. Interest in the Auger effect has however been stimulated by the increasing study of processes, such as electron capture and internal conversion, which involve the interaction of the nucleus with the orbital electrons. Since these processes lead to inner-shell ionization, measurements of the subsequent x-ray or Auger electron emission may yield important information about the initial nuclear process.

One important measure of the mechanism of de-excitation of the ith atomic shell is the fluorescence yield ω_i, which is defined as the number of characteristic x rays emitted per primary vacancy in the shell. The corresponding Auger yield a_i is defined as the

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number of characteristic Auger electrons emitted per primary vacancy.

Many types of measurement in nuclear spectroscopy require for their interpretation an accurate knowledge of the atomic fluorescence and Auger yields. A complete discussion of these has been given by Burhop[1] and Bergstrom and Nordling.[2] In the case of the K shell, reliable information on fluorescence yields is available except in regions of high and low atomic number (see, e.g., Listengarten[3]). Present knowledge of the L-shell fluorescence yields is much less satisfactory and is inadequate for application to many problems in nuclear spectroscopy.

Most experimental studies of L-shell fluorescence have been concerned with measuring the mean L-shell fluorescence yield $\bar{\omega}_L$, which is defined as the weighted mean of the individual L-subshell yields. Since the subshell yields are in general different, the mean L-shell fluorescence yield has a precise meaning only when the distribution of vacancies in the subshells is specified. It is thus a quantity of limited significance which must be interpreted with care. In many instances, this limitation has been ignored and measured values of $\bar{\omega}_L$ have been applied in situations where they are not relevant.

The determination of L-subshell yields is further complicated by the occurrence of Coster-Kronig[4] transitions. These are Auger transitions of the type $L_i \rightarrow L_jX$, where an initial vacancy in the L_i subshell is filled by an electron from the L_j subshell with the simultaneous ejection of an electron from the X shell. Thus, by transferring vacancies from one L subshell to another, these transitions alter the initial distribution of vacancies among the L subshells. Coster-Kronig transitions are possible only if the binding energy difference of the L subshells participating in the transition is greater than the binding energy of the X shell in the atom ionized in a deeper shell. Because of the variation of these binding energy differences with atomic number Z, these transitions are energetically possible only for restricted ranges of Z. The possible Coster-Kronig transitions in the L, M, and N shells have been listed by Burhop,[1] together with the ranges of Z in which they can occur.

In a comprehensive review of the Auger effect, Listengarten[3] has presented curves showing the variation of all nine L-subshell yields with atomic number in a range of Z from 47–98. These curves were constructed first of all on a basis of pure theory and the results were then normalized in such a way as to accord with existing experimental data at $Z=47$, 54, 56, and 82. In this program, which in essential respects followed the method employed earlier by Kinsey,[5] the radiation, Auger, Coster-Kronig, and total widths, denoted by Γ_{xi}, Γ_{ai}, Γ_{ik} and Γ_i, respectively, were calculated for each L_i subshell, and the corresponding yields obtained by application of the relations

$$\omega_i=\Gamma_{xi}/\Gamma_i, \quad a_i=\Gamma_{ai}/\Gamma_i, \quad f_{ik}=\Gamma_{ik}/\Gamma_i, \qquad \Gamma_i=\Gamma_{xi}+\Gamma_{ai}+\sum_{i>k}\Gamma_{ik}. \tag{1}$$

Listengarten used the radiation widths calculated by Massey and Burhop[6] multiplied by enhancement factors to correct for an inadequate treatment of screening in the original calculations. The Auger widths were assumed to increase linearly with Z over the range from 47–82 and more rapidly above $Z=82$. Thresholds for emission of Coster-Kronig electrons occur at $Z=51$, 73, 91, and 92, and these were taken account of in the calculations. In relation to the present work, the most important threshold is that at $Z=91$, where L_2-L_3 $M_{4,5}$ Coster-Kronig transitions become energetically possible. In his treatment of these, Listengarten assumed that f_{23} increases above $Z=91$ in the same way as f_{13} increases above $Z=73$ following the onset of L_1-L_3 $M_{4,5}$ Coster-Kronig transitions.

The results of measurements of individual L-subshell yields have been summarized by Burhop,[1,7] Listengarten,[3] and more recently by Fink *et al.*[8] In general these measurements are limited in precision and do not agree well with each other. This lack of agreement is particularly evident in the L_2-subshell yields and it has been pointed out by Salgueiro *et al.*[9] that, even in the most intensively studied element (bismuth, $Z=83$) values of ω_2 have been reported which vary between 0.32 and 0.66. More precise measurements of L_2-subshell yields are clearly required, particularly for heavy elements. It has been the aim of the series of experiments reported here both to supplement existing data on L_2-subshell yields in the region of $Z\geq 91$ and to attempt an experimental check on the predictions of Listengarten's theory.

2. THEORETICAL BASIS OF THE PRESENT EXPERIMENTS

The relationship between the L-subshell yields and quantities which can be measured directly has been discussed in a number of papers.[1,3,8,10] The analysis of

[1] E. H. S. Burhop, *The Auger Effect and other Radiationless Transitions* (Cambridge University Press, Cambridge, England, 1952).

[2] I. Bergström and C. Nördling, in *Alpha, Beta, and Gamma Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Co., Amsterdam, 1965).

[3] M. A. Listengarten, Izv. Akad. Nauk. SSSR, Ser. Fiz. **24**, 1041 (1961).

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[5] B. B. Kinsey, Can. J. Research **26**, 404 (1948).

[6] H. S. Massey and E. H. S. Burhop, Proc. Cambridge Phil. Soc. **32**, 461 (1936).

[7] E. H. S. Burhop, J. Phys. Radium **16**, 625 (1955).

[8] R. W. Fink, R. C. Jopson, H. Mark, and C. D. Swift, Rev. Mod. Phys. **38**, 513 (1966).

[9] L. Salgueiro, J. G. Ferreira, J. J. H. Park, and M. A. S. Ross, Proc. Phys. Soc. (London) **77**, 657 (1961).

[10] J. L. Wolfson, W. J. King, and J. J. H. Park, Can J. Phys. **41**, 1489 (1963).

Ross *et al.*[11] takes as point of departure the three identities

$$\omega_1+a_1+f_{12}+f_{13}=1\,, \tag{2a}$$

$$\omega_2+a_2+f_{23}=1\,, \tag{2b}$$

$$\omega_3+a_3=1\,, \tag{2c}$$

which connect the nine L-subshell yields. These relations, which follow from the definitions of the yields, reduce to six the number of independent quantities to be determined. If the initial ionization is confined to the L_2 and L_3 subshells, the number of yields involved is reduced to five and the number of independent quantities to three. In addition the cumulative effect of experimental error is reduced.

The present study makes use of the simple properties of heavy even-even isotopes to examine the restricted situation where the primary ionization is effectively confined to the L_2 and L_3 subshells. This method was introduced by Booth *et al.*[12] in their study of L-shell fluorescence in radium.

The decay scheme of $Cm^{244} \rightarrow Pu^{240}$ illustrated in Fig. 1 is typical of a whole class of even-even heavy α-emitters. Approximately 75% and 25% of the α transitions populate the 0^+ ground state and 2^+ first excited state, respectively, in the daughter nucleus, less than 0.2% of transitions leading to excitation of higher states. The low-energy fast enhanced $E2$ transition to the ground state is almost entirely converted in the L and higher shells and the conversion electron intensities in the L subshells are approximately in the ratio 3:100:100. Ionization is thus virtually confined to the L_2 and L_3 subshells.

For this special case, the equations relating the L_2-subshell yields to experimental quantities have been given by Salgueiro *et al.*[9] The small error introduced by neglecting the L_1-subshell vacancies in the first place can be corrected using theoretical estimates of L_1-subshell fluorescence and Coster-Kronig yields.

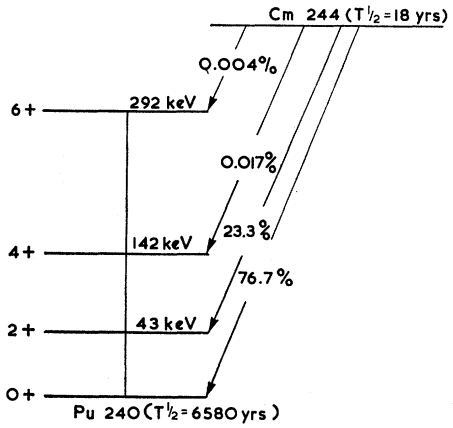

FIG. 1. Partial level scheme for Pu^{240} following Cm^{244} α decay.

The quantities n_i and I are defined as the number of L_i-subshell and total L-shell vacancies, respectively, per disintegration and the ratio of the number of L_3 vacancies to the number of L_2 vacancies is denoted by C_3'. F is the number of L x rays emitted per disintegration and F_3' is the ratio of the intensities of L x-ray emission from the L_3 and the L_2 subshells. In terms of these quantities the relations of Salgueiro *et al.*[9] may be written

$$\begin{aligned} &n_2+n_3=I\,, \quad n_3/n_2=C_3'\,,\\ &\omega_2 n_2+\omega_3(n_3+f_{23}n_2)=F\,,\\ &\omega_3(n_3+f_{23}n_2)/\omega_2 n_2=F_3'\,. \end{aligned} \tag{3}$$

Equations (3) may be combined to yield expressions for the L_2-subshell yields:

$$\omega_2=F(1+C_3')/I(1+F_3')\,, \tag{4}$$

$$f_{23}=(\omega_2/\omega_3)F_3'-C_3'\,. \tag{5}$$

From Eqs. (4), (5), and (2b) it follows that ω_2 may be determined from a knowledge of I, C_3', F, and F_3'. To determine f_{23} it is necessary to supplement these data with an estimate of ω_3.

In the experiments reported here values of F and F_3' were obtained for each of the daughter nuclei in the three decays $Cm^{244} \rightarrow Pu^{240}$, $Pu^{240} \rightarrow U^{236}$, and $Pu^{238} \rightarrow U^{234}$. I and C_3' were determined from published experimental data on the relative intensities of the α transitions in the decaying nucleus, the ratio of L-shell internal conversion to the total transition rate from the excited state, and the ratio of L_3- to L_2-subshell internal conversion. The small ($<1\%$) corrections for transitions from the higher excited states and for L_1-shell ionization required the use of some theoretical estimates. Where theoretical internal conversion coefficients were needed, the values compiled by Sliv and Band[13] were used since there is some evidence that Rose's[14] values do not agree with experiment for $2^+ \rightarrow 0^+$ transitions in this region of the periodic table.

The values of ω_3 used in the calculation of f_{23} were obtained by application of an empirical formula given by Burhop.[7] This formula was based on Kustner and Arend's[15] measurements of ω_3 for seven elements between $Z=73$ and $Z=83$ and on Stephenson's[16] measurements for $Z=82$, 90, and 92. These experimental data were fitted to a relation of the form

$$[\omega_3/(1+\omega_3)]^{1/4}=A+BZ\,, \tag{6}$$

[11] M. A. S. Ross, A. J. Cochran, J. Huges, and N. Feather, Proc. Phys. Soc. (London) **A68**, 612 (1955).
[12] E. Booth, L. Madansky, and F. Rasetti, Phys. Rev. **102**, 800 (1956).
[13] L. A. Sliv and I. M. Band, Leningrad Physico—Technical Institute Report, 1957 [English transl.: University of Illinois Physics Department Reports Nos. 57ICCKI and 58ICCLI, 1958 (unpublished)].
[14] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Co., Amsterdam, 1958).
[15] H. Küstner and E. Arends, Ann. Phys. (Leipzig) **22**, 443 (1937).
[16] R. J. Stephenson, Phys. Rev. **51**, 637 (1937).

TABLE I. (a) Intensities of α-particle groups emitted in decay of Cm^{244} and (b) intensities in conversion and emission of transitions in Pu^{240}.

(a)

Group	Final-state[a] energy (keV)	Intensity (%)	Reference
α_0	0	76.4	21
α_1	42.9	23.6	21
α_2	142.2	2×10^{-2}	21
α_3	292	3.4×10^{-3}	21

(b)

Transition energy (keV)	K	L_1	L_2	L_3	$(M+N+0)$	γ	Total	Reference
42.9		$L_1+L_2+L_3=0.710\pm0.007$					1	22
		3.87 ± 0.22	110 ± 4	100				23
100		0.3711	7.596	4.788	3.775	1.0	17.530	13
150	0.1892	0.0981	1.1420	0.6318	0.556	1.0	3.6171	13

where A and B were found from a least-squares fit to take the values $A=-0.221$ and $B=0.0126$. The results of Jopson[17] for twelve elements in the range from $Z=67$–83 were not taken into account since they have large experimental errors and do not show a systematic variation with atomic number.

Equation (6) gives $\omega_3=0.437\pm0.010$ for uranium and $\omega_3=0.462\pm0.010$ for plutonium. These results are in agreement with the less precise estimates of $\omega_3=0.458\pm0.1$ and $\omega_3=0.473\pm0.1$, respectively, taken from the work of Listengarten.[3]

3. EVALUATION OF I AND C_3'

A. $Cm^{244} \rightarrow Pu^{240}$

The α decay of Cm^{244} has been studied by Asaro *et al.*,[18] Hummel,[19] Dzhelepov *et al.*,[20] and most recently by Baranov *et al.*[21] The last named investigation was carried out using an α-particle magnetic spectrograph and a semi-conductor α spectrometer and eight α groups were identified in the decay. The relative intensities of the four strongest groups are summarized in Table I(a). From these results the absolute intensities of the 42.88-, 100-, and 150-keV transitions are found to be 0.23623, 0.000023, and 0.00003 per α-disintegration, respectively.

The spectrum of internal conversion electrons from the 43-keV transition in Pu^{240} has been studied by Halley and Engelkemeir[22] who have shown that (71.0 ±0.7)% of all conversions take place in the L shell and Hamilton *et al.*[23] find the intensities of conversion electrons from the L subshells to be in the ratio $3.87\pm0.22:110.0\pm4.0:100$. This information is sufficient to determine the number of vacancies in the L subshells arising from internal conversion of the 43-keV transition.

To estimate the contribution of the 100- and 150-keV transitions to the total number of vacancies, theoretical internal conversion coefficients were used. Table I(b) summarizes the relevant information on the three transitions. From these data we obtain $n_1=0.00307$, $n_2=0.08623$, and $n_3=0.07843$.

In calculating C_3' and I from n_1, n_2, and n_3 it is necessary to take account of the nonzero value of n_1. Since, according to the analysis of Listengarten,[3] the Coster-Kronig transfer yields f_{12} and f_{13} are approximately 0.1 and 0.4, respectively, in this region of Z, fully half of the L_1-subshell vacancies are transferred to the L_2 and L_3 subshells. Thus, the corrected values of C_3' and I are given by

$$C_3'=(n_3+f_{13}n_1)/(n_2+f_{12}n_2) \tag{7}$$

and

$$I=n_2+n_3+n_1(f_{12}+f_{13}). \tag{8}$$

On substituting for n_1, n_2, and n_3 in (7) and (8) we obtain the results

$$I=0.1666\pm0.0017, \quad C_3'=0.920\pm0.037.$$

B. $Pu^{240} \rightarrow U^{236}$

In this decay only one result for the relative intensities of the α-particle groups is available. These measurements were made by Goldin *et al.*[24] using a magnetic spectrograph and the results are summarized in Table II(a). From these data the absolute intensities of the

[17] R. C. Jopson, J. M. Khan, H. Mark, C. D. Swift, and M. A. Williamson Phys. Rev. 133, A381 (1964).
[18] F. Asaro, S. G. Thompson, and I. Perlman, Phys. Rev. 92, 694 (1953).
[19] J. P. Hummel, Ph.D. thesis, University of California Radiation Laboratory Report No. UCRL-3456, 1956 (unpublished).
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[23] J. H. Hamilton, B. Von Nooijen, A. V. Ramaya, and W. H. Brantley, in *Internal Conversion Processes*, edited by J. H. Hamilton (Academic Press Inc., New York, 1966), p. 541.
[24] L. L. Goldin, G. I. Novikova, and E. F. Tretyakov, Phys. Rev. 103, 1004 (1956).

TABLE II. (a) Intensities of α-particle groups emitted in decay of Pu^{240} and (b) intensities in conversion and emission of transitions in U^{236}.

(a)

Group	Final-state energy (keV)	Intensity (%)	Reference
α_0	0	75.5	24
α_1	45.28	24.5	24
α_2	151	8.5×10^{-2}	24
α_3	206	7×10^{-3}	24

(b)

Transition energy (keV)	K	L_1	L_2	L_3	$M+N+0$	γ	Total	Reference
45.28		$L_1+L_2+L_3=1$			0.337 ± 0.037			25
		$L_1+L_2=110\pm15$		100				25
105.7		0.236	4.640	3.011	0.233	1.0	11.217	13
162	0.1989	0.0662	0.6954	0.3745	0.336	1.0	2.5710	13

45.28-, 105-, and 162-keV transitions in U^{236} are deduced to be 0.24477, 0.00092, and 0.00003 per α disintegration, respectively.

In the 45-keV transition in U^{236}, the ratio of the intensity of conversion electrons in the M and higher shells to the intensity in the L shell has been determined[25] to be 0.337 ± 0.04, while the intensities in the L_1 and L_2 subshells combined has been measured as (1.10 ± 0.15) times the intensity in the L_3 subshell. In this case the small correction for L_1-shell ionization has been made by assuming that these occur with an intensity 3.7% that of L_3-subshell ionizations which is the result predicted by theory. The contributions from the weak transitions at 105 and 162 keV have also been estimated from theory and the relative intensities of all three transitions in emission and conversion are displayed in Table II(b). With these data, values of $n_1=0.00318$, $n_2=0.09326$, and $n_3=0.08739$ are obtained. The corresponding values for I and C_3' are

$$I=0.1826\pm0.006,\quad C_3'=0.945\pm0.128.$$

C. $Pu^{238}\rightarrow U^{234}$

Four sets[22,26–28] of measurements of comparable accuracy are available for the α disintegration of Pu^{238} and the relative intensities of the α-particle groups populating the excited states of U^{234} are shown in Table III(a). The four sets of results are in good agreement and accordingly the mean value of the measured intensities has been adopted. With these data the absolute intensities of the 43.5-, 100-, and 146-keV transitions are found to be 0.29335, 0.00135, and 0.00005 per α disintegration, respectively.

For the 43.5-keV transition in U^{234} two measurements have been reported for the fraction of transitions giving rise to L-shell conversion. Halley and Engel-

TABLE III. (a) Intensities of α-particle groups emitted in decay of Pu^{238} and (b) intensities in conversion and emission of transitions in U^{234}.

(a)

Group	Intensity (%) Reference[a]	Reference[a]	Reference[b]	Reference[c]	Adopted value
α_0	72.0	69.0	71.1	71.1	70.8
α_1	28.0	31.0	28.8	28.9	29.2
α_2	0.1	...	0.13	...	0.13
α_3	5×10^{-3}	...	5×10^{-3}	...	5×10^{-3}

(b)

Transition energy (keV)	K	L_1	L_2	L_3	$M+N+0$	γ	Total	Reference
43.5		$L_1+L_2+L_3=0.764\pm0.007$					1	22
		$L_1+L_2+L_3=0.752\pm0.022$					1	25
		$L_1+L_2=130\pm15$		100				25
100		0.288	6.087	3.968	3.072	1.0	14.415	13
146	0.2095	0.0776	0.8925	0.5020	0.438	1.0	3.1196	13

[a] Reference 26. [b] Reference 28. [c] Reference 22.

[25] D. H. Rester, M. S. Moore, F. E. Durham, and C. M. Class, Nucl. Phys. **22**, 104 (1961).
[26] F. Asaro and I. Perlman, Phys. Rev. **94**, 381 (1954).
[27] L. L. Goldin, G. I. Novikova, and E. F. Tretyakov, in Proceedings of the USSR Academy of Sciences Conference on Peaceful Uses of Atomic Energy, 1955 (unpublished).
[28] L. N. Kondratyev, G. I. Novikova, V. B. Dedov, and L. L. Goldin, Izv. Akad. Nauk. SSSR, Ser. Fiz. **21**, 907 (1957).

kemeir[22] find this fraction to be (76.4±0.7)% and Rester *et al.*[25] report a value of (75.4±2.2)%. The weighted mean of these results, (76.3±0.7)%, has been adopted in the present analysis. The ratio of (L_1+L_2)-subshell conversion to L_3-subshell conversion has been measured[25] to be 1.30±0.15 and this result has been corrected for L_1-subshell conversion exactly as for the equivalent situation in U^{236}. The higher-energy transitions have been treated theoretically just as before and the relevant information on all three transitions has been collected in Table III(b). These results lead to the values $n_1=0.00381$, $n_2=0.12291$, $n_3=0.09710$. The final values for I and C_3' are

$$I=0.2230\pm0.002, \quad C_3'=0.798\pm0.12.$$

The values for I and C_3' in all three decays are assembled in Table VIII. The experimental values of C_3' may be compared with the values of 0.920, 0.863, and 0.868 for Pu^{240}, U^{236}, and U^{234}, respectively, derived using the theoretical internal conversion coefficients of Sliv and Band.[13]

4. MEASUREMENT OF F

The angular correlation of α particles and L x rays from the decay of Th^{230} has been studied by Falk-Vairant *et al.*[29] Within the experimental errors (±4%), L x-ray emission was found to be isotropic with respect to the direction of α emission. Since Th^{230} is typical of many even-even α-active heavy nuclei, it may be assumed that isotropy of L x-ray emission is a property common to the whole class. Thus the number of L x rays emitted per α disintegration may be measured by counting the number of L x rays radiated into a known solid angle, which are in coincidence with α particles. The values of F for all three nuclei studied in the present experiments were obtained in this way.

A. Experimental Arrangement

Experiments were carried out on three carrier-free sources of Cm^{244}, Pu^{240}, and Pu^{238} which were obtained from AERE Harwell. These sources were deposited on aluminium foils of superficial density 0.68, 1.22, and 0.58 mg/cm², respectively. α particles were detected in a small silicon surface barrier detector placed a few millimeters from the open side of the source and L x rays were recorded in a 1-in.×1-in. NaI(Tl) crystal through a thin window composed of 5-mg/cm² aluminum, 6-mg/cm² MgO, and 7-mg/cm² araldite (composition by weight 66.7% C, 20.0% O, 11.2% H, and 2.1% N). The L x ray counter was placed facing the side of the foil opposite to that on which the source was deposited.

A lead collimator of accurately measured dimensions, placed in front of the scintillation counter, defined the geometrical solid angle for L x-ray collection. The solid angle, which was known to better than 1%, could be varied by altering the source-to-aperture distance using a series of accurately measured spacers of brass tubing interposed between the source holder and the photomultiplier housing.

The coincidence unit employed for measuring α–L x-ray coincidences was of conventional slow type and L x-ray spectra were recorded on a 120-channel Hutchinson-Scarrot pulse-height analyzer. Examples of single and coincidence L x-ray spectra from the decay of Pu^{238} are shown in Fig. 2.

B. Measurements

Before commencing coincidence experiments, the α and γ spectra of all three sources were investigated using the silicon detector, NaI crystal, and an argon-methane gas proportional counter. No trace of any contaminant was observed and repeated examination of the spectra over 18 months confirmed these results.

For each source the measurement of F was repeated at several different solid angles, and at each setting counting was continued until at least 6000 coincidences had been recorded. Coincidence counting rates observed

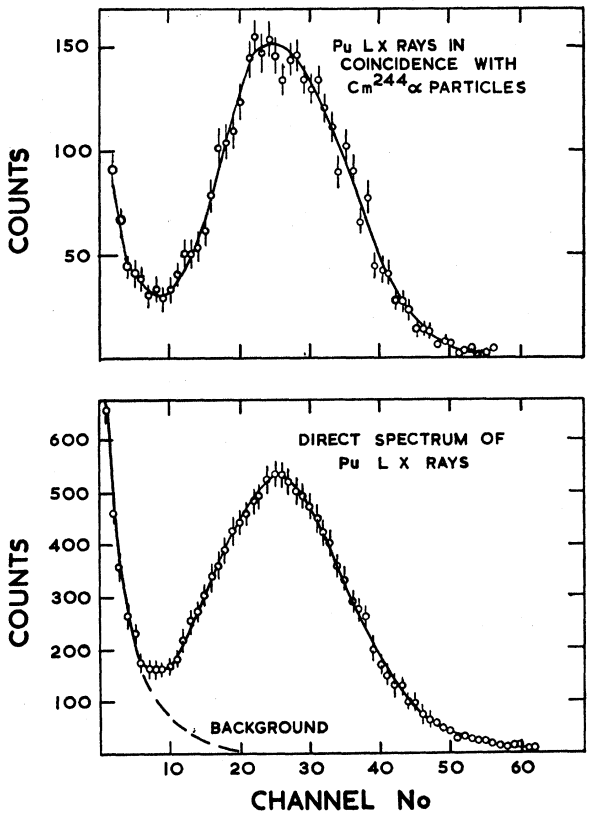

FIG. 2. In the upper half of this figure is shown a pulse-height spectrum of Pu L x rays in coincidence with Cm^{244} α particles. In the lower half of the figure is shown the direct spectrum of Pu L x rays.

[29] P. Falk-Vairant, J. Teillac, G. Valladas, and P. Benoist, Compt. Rend. 238, 1409 (1954).

varied between 0.8 and 10.5 counts/min depending on the source and solid angle used.

Since the detection efficiency of the NaI crystal is effectively unity in the energy range 10–20 keV which is characteristic of the L x rays from heavy elements, the observed number of L x rays per disintegration, denoted by F_0, is given by the relation

$$F_0=N_c/\Omega N_\alpha.$$

N_c is the coincidence counting rate corrected for random coincidences, N_α is the α-particle counting rate, and Ω is the solid angle expressed as a fraction of 4π sr.

The values of F were obtained from F_0 by correcting for absorption between source and counter using the known superficial densities of the intervening materials. The appropriate mass absorption coefficients were calculated from a quadratic fit to the logarithms of the coefficients for absorption and scattering (excluding coherent scattering) tabulated by McGinnies[30] at 10, 15, and 20 keV.

C. Results

The results of the measurements of F are summarized in Table IV. Since the most likely errors of instrumental origin would lead to a relation of the form

$$F_0=A+B/\Omega,$$

the correlation coefficient r for the variables F_0 and $1/\Omega$ was calculated for each source. The values of r may appear rather large, but application of Student's t test[31] shows that none of them is significant even at the 50% level. The variable t subjected to this test is defined by

$$t=r[(n-2)/(1-r^2)]^{1/2},$$

where n is the number of solid angles for which experiments were carried out and $n-2$ is the number of degrees of freedom. As the measured values of F_0 showed no significant correlation with solid angle, the value accepted was the weighted mean of results for all solid angles.

Very few measurements of F are available with which to compare the present results. Asaro and Perlman[26] found a value of $F=0.13$ for the decay $Pu^{238}\rightarrow U^{234}$, which agrees very well with the present result. This agreement may well be fortuitous however since their measurement was based on a comparison with the L x rays emitted in the decay of Am^{241}.

Halley and Engelkemeir[22] have measured the mean L-shell fluorescence yields in Pu^{240} and U^{234} using a number of techniques including that employed in the present work. Values of F are not quoted in their paper but using the data presented there, and making the small ($<1\%$) correction for L_1-subshell vacancies, values of $F=0.0876\pm0.0015$ for Pu^{240} and $F=0.1043\pm0.0017$ for U^{234} may be inferred from their work. In both cases the differences between these results and those reported in this study are significantly large and cannot be attributed to sampling error.

5. MEASUREMENTS OF F_3'

F_3' was determined for each of the three sources studied, from a proportional-counter measurement of the relative intensities of the $L\alpha$:$L\beta$:$L\gamma$ x-ray groups. In addition F_3' for Pu^{238} decay was determined from a measurement of the relative intensities of the individual L x-ray lines using a curved-crystal spectrograph.

A. Proportional-Counter Measurements

The proportional counter used to measure the relative intensities of $L\alpha$, $L\beta$, and $L\gamma$ x-ray groups was of simple design. It consisted of a cylindrical aluminium cathode, 10 in. long, with inner and outer diameters 1.875 and 2 in., respectively. The counter anode, a pure tungsten

TABLE IV. Experimental data for the determination of F.

α-decay	$10^3\Omega$[a]	$10^2\times F_0$	r (%)	$10^2\bar{F}_0$	(Transmission)$^{-1}$	$10^2\bar{F}$	$10^2\bar{F}$ corrected for L_1-subshell vacancies (0.6%)
$Cm^{244}\rightarrow Pu^{240}$	5.587	9.03±0.14	43.3	8.928±0.134	1.0623	9.484±0.142	9.42±0.14
	5.587	8.76±0.13					
	2.306	8.88±0.13					
	2.306	9.01±0.13					
	1.435	9.05±0.15					
$Pu^{240}\rightarrow{}^{236}$	5.872	9.68±0.11					
	3.428	9.77±0.12	58.0	9.735±0.050	1.0757	10.472±0.054	10.41±0.05
	2.242	9.73±0.13					
	1.715	9.83±0.20					
$Pu^{238}\rightarrow U^{234}$	5.100	12.05±0.22					
	2.338	11.83±0.25	55.4	12.019±0.131	1.0718	12.882±0.140	12.83±0.14
	1.658	12.10±0.21					
	1.465	12.06±0.27					

[a] The solid angle Ω is expressed in units of 4π sr.

[30] R. T. McGinnies, Natl. Bur. Std. (U. S.), Suppl. Circ. No. 583, 6 (1959).
[31] R. A. Fisher, *Statistical Methods for Research Workers* (Oliver and Boyd, Edinburgh, 1938).

wire of 0.004-in. diam was threaded through stainless-steel hypodermic needles in the center of two cylindrical ebonite blocks which fitted into the aluminium cathode. The complete assembly was housed in a brass cylinder of length 16 in., internal diameter 5.75 in., and external diameter 6 in. This apparatus was part of a closed system containing a gas mixture of 90% argon and 10% methane at a variable pressure. A purifier, which consisted of calcium turnings heated to 350–400°C by an electrically heated coil, was used to remove oxygen and other electronegative impurities from the gas. The flow of gas in the system was maintained by convection.

The sources were mounted outside the counter close to an aluminium window, 0.127 mm thick, set in the center of one side of the brass housing. The collimator between the window and the counter proper was lined with a layer of 0.002-in. molybdenum, and 0.001-in. aluminium to prevent the induced fluorescent radiation from the copper jacket from reaching the sensitive region of the counter.

For each of the three sources, the measurements of the relative intensities of the L x-ray groups was repeated five times, at gas pressures of 40, 60, and 80 cm Hg. The total counting time for each run was of the order of 5–6 h. The counting time was split into 30-min periods, and the L x-ray spectrum under examination and the background spectrum were recorded on the pulse-height analyzer in alternate periods. The two sets of measurements were then summed separately, and the total background spectrum was subtracted from the total L x-ray spectrum. The counting rates were low enough to eliminate any need for a counting rate correction.

Figure 3 shows an example of the Pu L x-ray spectrum following Cm^{244} decay as observed with the proportional counter at 80-cm Hg pressure. The natural background has been subtracted. The $L\alpha$, $L\beta$, and $L\gamma$ x-ray groups are clearly resolved in this spectrum. The peak on the low-energy side of the $L\alpha$ peak, which appeared consistently in all of the observed spectra, is the $L\alpha$ escape peak. This peak contains events where an $L\alpha$ quantum is absorbed by a photoelectric process in argon, the argon atom reorganizes by K x-ray emission and the Ar K x-ray quantum escapes from the counter. This escape peak had an intensity of the order of 5–6% of the intensity of the full-energy peak.

The spectrum was analyzed into its component lines on the assumption that the lines have the same shape as the 59.6-keV γ-ray line in Am^{241}, which was observed under the same conditions. The individual components are shown by the dashed lines in Fig. 3. At a given gas pressure the intensity of the escape peak relative to the full-energy peak is independent of energy, and depends on the counter geometry alone. Hence the intensity of the $L\alpha$ escape peak relative to the $L\alpha$ peak could be used to correct the observed intensities of the $L\beta$ and $L\gamma$ peaks. The observed intensities were also corrected

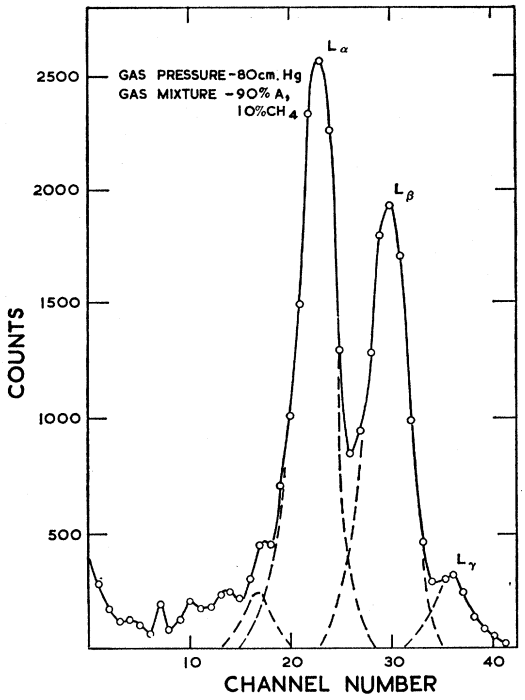

FIG. 3. An example of the Pu L x-ray spectrum as observed in the proportional counter. The spectrum was resolved into the $L\alpha$, $L\beta$, and $L\gamma$ x-ray groups as shown by the dashed lines. The small peak of the low-energy side of the $L\alpha$ peak is the $L\alpha$ escape peak. This peak appeared consistently in all the L x-ray spectra observed with this counter.

for the absorption of L x rays between the source and the sensitive region of the counter, and for the variation of the counter sensitivity with energy. The sensitivity correction was made using the known photoelectric absorption coefficients of methane and argon.[30]

The measured relative intensities of the L x-ray groups from Pu^{240}, U^{236}, U^{234} are listed in Table V. Since the L_1 subshell makes a negligible contribution to the total number of vacancies, it is possible to distinguish three independent methods of deriving F_3' from the relative intensities. The essential point is that, in these special circumstances, $L\alpha$ and $L\gamma$ x rays originate entirely from the L_3 and L_2 subshell, respectively, whereas $L\beta$ x rays originate in either subshell.

The first method uses the ratio $L\alpha/L_3$ which is defined as the ratio of the intensity of $L\alpha$ x rays to the intensity of all x rays originating in the L_3 subshell, to determine the relative intensity of L_3 x rays from the relative intensity of $L\alpha$ x rays. The ratio $L\alpha/L_3$ is

TABLE V. Relative intensities of $L\alpha$:$L\beta$:$L\gamma$ measured with the proportional counter.

α decay	$L\alpha$	$L\alpha$	$L\gamma$
$Cm^{244} \rightarrow Pu^{240}$	0.951±0.092	1.0	0.207±0.028
$Pu^{240} \rightarrow U^{236}$	0.776±0.040	1.0	0.194±0.023
$Pu^{238} \rightarrow U^{234}$	0.792±0.028	1.0	0.192±0.034

independent of the method of producing vacancies and has been derived from curved-crystal spectra of L x rays from plutonium[9,32] and uranium.[33–35] The second method uses the ratio $L\gamma/L_2$ to give a value for the relative intensity of L_2 x rays. Since L_1 x rays have a negligible intensity, either method provides a value for F_3'.

The third method required a knowledge of how the $L\beta$ group is divided between L_2 and L_3 x rays. Since $L\beta_1$ alone originates in the L_2 subshell, the ratio $(L\beta-L\beta_1)/(C_3'L\beta_1)$ is a function of Z only provided always that L_1-subshell vacancies have a negligible intensity. This ratio has been determined for plutonium from values of C_3' and curved-crystal L x-ray spectra published by Salgueiro *et al.*[9] and by Barton *et al.*[32] for the decay of Cm^{242}. The value for uranium has been derived from results reported here on the decay of Pu^{238}.

The relevant data for the determination of F_3' from the proportional-counter measurements are summarized in Table VI. The values of F_3' are listed in Table VII. In the case of U^{234}, F_3' was also determined by an independent method based on curved-crystal spectra alone.

B. Curved-Crystal Measurements

The curved-crystal spectrograph, of the Cauchois approximate focusing type, had a Mica crystal, 0.25 mm in thickness, bent to a cylinder of 20-cm radius. The uranium L x-ray spectrum was recorded on Ilford G5 nuclear emulsion plates, 200 μ thick. The emulsion was covered with a single layer of black paper. Cochran[34] had earlier calibrated the sensitivity of the spectrograph as a function of energy. His calibration in the energy range 9–36 keV was carried out under the same conditions as in the present experiment using the K x rays of thirteen elements.

The measurement of F_3' was made using a 2.6-mCi source of Pu^{238} painted uniformly on a thin, stainless-steel disk. The source material covered a circular area, 14 mm in diam. A small amount of Pu^{239}, 4.5% by weight, was present in the source. Since the half-life of Pu^{239} is approximately 2.4×10^4 years, it contributed only 0.017% of the total source activity. Hence no correction was required for the presence of this impurity. The source was covered with a layer of polyvinyl Formar of superficial density 247 $\mu g/cm^2$. A Mica window, 2.47 $\mu g/cm^2$ thick, was also interposed between the source and the spectrograph.

The nuclear emulsion was exposed to the uranium L x rays for 70 days. It was then developed using the temperature cycle development technique[36] to ensure uniform development throughout the depth of the emulsion. The developer used was an elon based developer as used by Cochran.[34] After development, the plate was microdensitometered using an automatic recording microdensitometer, made by Joyce, Loebl, and Co., Ltd. Unfortunately, the part of the plate containing the $L\alpha$ lines was spoiled during development. As a result it was not possible to determine F_3' using all of the lines. However, a measure of F_3' could still be obtained from the relative intensities of the $L\beta_1$ and $L\beta_2$ lines.

Several traverses were made with the microdensitom-

TABLE VI. Curved-crystal data required for evaluation of F_3'.

$L\alpha/L_3$	$L\gamma/L_2$	$(L\beta-L\beta_1)/C_3'L\beta_1$	$L\beta_1/L_2$	$L\beta_2/L_2$	Reference
		Plutonium			
0.7441	0.192	0.3740			9
0.7381	0.228				32
0.741±0.003	0.210±0.018	0.3740			Average
		Uranium			
0.7652	0.196		0.8045	0.1664	33
0.7671	0.157		0.8244	0.1789	34
0.7605	0.220		0.7304	0.1843	36
		0.3348			Present work
0.764±0.005	0.191±0.026	0.3348	0.786±0.040	0.1765±0.0075	Average

TABLE VII. Measured values of F_3'.

	F_3' from proportional-counter measurements			F_3' from curved-crystal measurements	
α-decay	Method 1	Method 2	Method 3		Weighted mean
$Cm^{244} \rightarrow Pu^{240}$	1.47±0.14	1.19±0.16	1.27±0.21	···	1.332±0.096
$Pu^{240} \rightarrow U^{236}$	1.06±0.05	0.94±0.20	1.07±0.20	···	1.072±0.047
$Pu^{238} \rightarrow U^{234}$	1.09±0.03	0.98±0.26	1.02±0.21	1.08±0.08	1.081±0.035

[32] G. W. Barton, H. P. Robinson, and I. Perlman, Phys. Rev. **81**, 208 (1951).
[33] A. H. Compton and S. K. Allison, *X Rays in Theory and Experiment* (D. Van Nostrand, Inc., New York, 1935).
[34] A. J. Cochran, Ph.D. thesis, University of Edinburgh, 1955 (unpublished).
[35] M. Goldberg, J. Phys. Radium. **22**, 743 (1961).
[36] C. C. Dilworth, G. Occhialini, and L. Vermaesen, Bull. Centre Phys. Nucl. Univ. Libre Bruxelles **13A, Part 1, 1** (1950).

TABLE VIII. Data required for determination of ω_2, f_{23}, and a_2.

α decay	10^2F	F_3'	C_3'	I	ω_3
$Cm^{244} \rightarrow Pu^{240}$	9.42±0.14	1.332±0.096	0.920±0.037	0.1666±0.0017	0.462±0.010
$Pu^{240} \rightarrow U^{236}$	10.41±0.05	1.072±0.047	0.945±0.128	0.1826±0.006	0.437±0.010
$Pu^{238} \rightarrow U^{234}$	12.83±0.14	1.081±0.035	0.798±0.12	0.2230±0.002	0.437±0.010

TABLE IX. The L_2 subshell yields in uranium and plutonium. Two values are listed for each experimentally determined yield. Those in the upper (lower) row are derived from the experimental (theoretical) value of C_3'.

ω_2	f_{23}	a_2	α decay	C_3'	Reference
			Uranium		
0.535±0.042	0.37±0.07	0.09±0.11	$Pu^{240} \rightarrow U^{236}$	0.945(E)	Present work
0.513±0.021	0.40±0.07	0.09±0.10		0.863(T)	
0.497±0.035	0.43±0.06	0.07±0.07	$Pu^{238} \rightarrow U^{234}$	0.798(E)	Present work
0.505±0.012	0.38±0.06	0.11±0.06		0.868(T)	
0.54 ±0.10	0.11±0.04	0.35±0.14	...	...	3
			Plutonium		
0.466±0.023	0.42±0.08	0.11±0.08	$Cm^{244} \rightarrow Pu^{240}$	0.920(E)	Present work
0.466±0.021	0.42±0.09	0.11±0.09		0.920(T)	
0.413±0.02	0.22±0.08	0.37±0.08	$Cm^{242} - Pu^{238}$	0.80(E)	9
0.430±0.02	0.21±0.08	0.35±0.08		0.925(T)	
0.48 ±0.10	0.24±0.10	0.28±0.20	...	...	3

eter at different heights on the plate, and the results were summed. The observed relative intensities of the two lines were then corrected using Cochran's sensitivity-energy calibration curve, and the ratio of the intensities of the $L\beta_2$ and $L\beta_1$ lines was determined to be 0.243±0.004.

A number of determinations have been reported[33–35] for the intensities of $L\beta_1$ and $L\beta_2$ in uranium relative to the total intensities of L x rays originating in the L_2 and L_3 subshells, respectively. The values obtained are listed in Table VI and the results indicate that the relative intensities of $L\beta_1$ and $L\beta_2$ must be multiplied by 4.46±0.30 in order to obtain F_3'. The final value of $F_3'=1.08\pm0.08$ for Pu^{238} is compared with the proportional-counter measurements in Table VII.

6. L_2-SUBSHELL YIELDS IN U AND Pu

The L_2-subshell yields in uranium and plutonium may be derived from the data assembled in Table VIII by application of relations (4), (5), and (2b). The results are listed in Table IX together with values of the yields based on other experimental or theoretical work.

Some remarks should first be made relating to the principal sources of error in these results. In the case of the measurements on Pu^{240}, the largest error stems from the determination of F_3' resulting from a lack of precision in the measurement of the relative intensity of the $L\alpha$ group using the proportional counter. Another consequence of this fact is that the value of F_3', obtained by the first method, which in the cases of U^{234} and U^{236} received by far the greatest weight in the final estimate, has received a much smaller weight in the result for Pu^{240}.

The lack of precise knowledge of C_3' is the main source of error in the results obtained for U^{236} and U^{234} and this represents an important limitation for all three nuclei studied. For this reason the values of the L_2-subshell yields listed in Table IX are supplemented by values obtained using estimates of C_3' based on the internal conversion coefficients of Sliv,[13] and corrected for L_1-subshell conversion as described in Sec. 3.

We consider first the values obtained for the L_2-subshell fluorescence yields. The values of $\omega_2=0.535\pm0.042$ and $\omega_2=0.497\pm0.035$, found for the uranium isotopes U^{236} and U^{234}, are consistent with each other and with the theoretical predictions of Listengarten. The agreement improves significantly in both respects when the computation is carried out using the theoretical estimates of C_3' listed at the end of Sec. 3. The results therefore tend to support not only the theoretical work of Listengarten but also the theoretical internal conversion coefficients of Sliv.

The value of $\omega_2=0.466+0.023$ obtained for Pu^{240} is consistent with, but about 3% smaller than, the value predicted by Listengarten. The value 0.413+0.02 reported by Salgueiro *et al.* from observations on Pu^{238}, is 14% below that predicted theoretically and differs significantly from that obtained in the present work.

It is possible to arrive at a value of $\omega_2=0.444+0.02$ for plutonium from the data published by Salgueiro *et al.*, by using theoretical estimates of C_3' and a value of $I=0.1968$ based on the most recent[21] data on the α decay of Cm^{242}. This result lies just outside the range of values predicted by Listengarten and differs by only one standard deviation from the result obtained for Pu^{240} in the present experiments. Nevertheless, an examination of the systematic behavior of F and I with

atomic number and mass number for the nuclei studied here, strongly suggests that the value of F reported by Salgueiro *et al.* for Pu^{238} is too low. It is this factor, rather than small uncertainties in the values of I and C_3', which is responsible for the discrepancy between the values of ω_2 reported for Pu^{240} and Pu^{238}.

The experimental evidence relating to the Coster-Kronig yields in Pu^{240}, U^{236}, and U^{234} provides at least qualitative support for the prediction of the Listengarten theory of a rapid increase in f_{23} above $Z=91$; the onset of L_2-L_3 $M_{4,5}$ Coster-Kronig transitions is thus conclusively demonstrated. The measured values of f_{23} are however about twice as large as those predicted by theory and are almost as large as consistency with the measured values of ω_2 allows. It should be emphasized again that all the experimental values of f_{23} depend on assuming a value for ω_3 which has not itself been derived directly from experiment.

The combined results for the fluorescence and Coster-Kronig yields obtained in the present series of experiments tend to suggest that the radiative widths have been correctly estimated in the Listengarten theory, but that the normal Auger effect is suppressed relative to the Coster-Kronig effect near threshold. At the time Listengarten's theory was put forward, experimental results on nonradiative transitions near the Coster-Kronig threshold were almost nonexistent. Since that time, however, the observations of Ferreira *et al.*[37] on L-series satellites have confirmed the essential correctness of the predicted behavior of f_{13} above $Z=73$. As has been stated in Sec. 1, the theory of nonradiative transitions above $Z=91$ rests squarely on the assumption that Γ_{23} varies for $Z\geq 91$ exactly as Γ_{13} varies for $Z\geq 73$. The present experimental results therefore cast doubt on the validity of this assumption.

An analysis of the dynamics of the Coster-Kronig transitions at $Z=73$ and $Z=91$ reveals several reasons why Γ_{23} might be expected to increase more rapidly for $Z\geq 91$ than Γ_{13} increases for $Z\geq 73$. In the first place Coster-Kronig transition probabilities depend strongly on the energy of the ejected electron. L_1-L_3 $M_{4,5}$ Coster-Kronig electrons have energies in the range 10–20 eV when these transitions become possible below $Z=50$ and above $Z=73$, and these energies increase to about 80–120 eV for values of Z at which the transition probability is maximum. L_2-L_3 $M_{4,5}$ Coster-Kronig electrons have energies of the order 50 eV at $Z=91$ and these energies increase rapidly with larger values of Z. This difference in the energy scale may itself be sufficient to explain the more rapid increase observed in f_{23} above $Z=91$.

[37] J. G. Ferreria, M. O. Costa, M. I. Goncalves, and L. Salgueiro, J. Phys. (Paris) **26**, 5 (1965).

There is a second, and possibly more important, effect which may contribute to the observed increase. Transfer of ionization from the L_1 subshell to the L_3 subshell involves a change in parity in the electron wave function. This has the consequence that, ignoring velocity-dependent contributions to the atomic potential, only p-wave and f-wave Coster-Kronig electrons can be ejected. No parity change is involved in transfer of ionization from the L_2 subshell to the L_3 subshell and the predominant emission consists of s-wave Coster-Kronig electrons for which the transition probability is greatest.

7. MEAN L-SHELL FLUORESCENCE YIELDS

In the notation of Ross *et al.*[11] the mean L-shell fluorescence yield is defined as $\bar{\omega}_L=F/I$, where F and I are not corrected for ionization in the L_1 subshell. F is therefore taken from the column of uncorrected results in Table IV, and I is redefined as the sum of the vacancies in all three subshells. The resulting values are as follows:

$$\bar{\omega}_L=0.566\pm0.010 \quad \text{for} \quad Pu^{240},$$
$$\bar{\omega}_L=0.570\pm0.019 \quad \text{for} \quad U^{236},$$
$$\bar{\omega}_L=0.576\pm0.015 \quad \text{for} \quad U^{234}.$$

Halley and Engelkemeir[22] report the values $\bar{\omega}_L=0.540\pm0.009$ and $\bar{\omega}_L=0.478\pm0.009$ for Pu^{240} and U^{234}, respectively. As has been noted in Sec. 4, the disagreement between these results and those given here stems entirely from the conflicting values of F found in the two sets of experiments. Salgueiro *et al.*[9] obtained the value $\bar{\omega}_L=0.486\pm0.01$ for Pu^{238} following the decay of Cm^{242}, while Akalaev *et al.*[38] quote a value of $\bar{\omega}_L=0.73\pm0.10$ based on observations of L x-ray emission from sources containing mixtures of Cm^{242} and Cm^{244}. Recognizing that $\bar{\omega}_L$ is a quantity of very limited significance, it is nevertheless clear that there is no real agreement between the various experimental values fo $\bar{\omega}_L$ reported for uranium and plutonium.

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[38] G. G. Akalaev, N. A. Vartanov, and P. S. Samoilov, Izv. Akad. Nauk SSSR, Ser. Fiz. **28**, 1260 (1964).