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ANOMALOUS ELECTRIC DIPOLE CONVERSION COEFFICIENTS IN ODD-MASS ISOTOPES OF THE HEAVY ELEMENTS

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ABSTRACT

A detailed review is given of experimental data on the anomalous L- and M-shell conversion coefficients of low-energy electric dipole transitions observed in the decays of odd-A nuclei of high atomic number.

The data are consistent in every case with the interpretation that the E1 conversion coefficients in the L_{III} shell agree with the theoretical, model-independent coefficients calculated by Sliv and Band and by Rose. It is definitely established in several well-measured cases that the L_I and L_{II} coefficients are substantially larger than the theoretical values. The most striking anomaly occurs in the 84.2-kev transition in Pa^{231}, where the L_I and L_{II} coefficients are 21 and 15 times larger than the theoretical values, respectively.

The experimental L_I and L_{II} coefficients are correlated with the lifetimes of the transitions, and it is shown that the magnitude of the anomaly (L_I plus L_{II}) is proportional to the retardation in gamma-ray lifetime over that calculated from the single-proton formulas. No systematic trend has been observed in the deviations of the L_I and L_{II} coefficients individually.

*Work done under the auspices of the U.S. Atomic Energy Commission.

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INTRODUCTION

A number of electric dipole (E1) transitions have been identified in the decay schemes of the trans-lead isotopes. It has been established that several of these E1 transitions in odd-mass nuclei have measurable lifetimes and, in fact, are longer-lived by many orders of magnitude than would be expected on the basis of "single-particle" transition-probability formulas. The first such case noted was a 59.6-kev transition in Np^{237} found by Beling, Newton, and Rose[1] to have a half life of 6×10^{-8} seconds, which is more than 10^5-fold slower than the value calculated from the usual lifetime formulas. In the meantime, it has also been noted that the over-all conversion coefficient[2] and the L and M subshell conversion ratios[3,4] for this transition have values which are definitely at variance with the theoretically calculated conversion coefficients of M. E. Rose.[5] More recently, Ewan, Knowles, and MacKenzie[6] noted that the 106-kev E1 transition in Pu^{239} has L_I and L_{III} conversion coefficients distinctly different from the theoretical values. It has also been suggested by Rosenblum, Valadares, and Milsted[7] that the abnormal conversion ratios of the 59.6-kev transition in Np^{237} may be related to the slowness of the transition.

The purpose of this paper is to review some of the data on electric dipole transitions, to demonstrate the existence of additional anomalous conversion coefficients in the L and M subshells, and to correlate the magnitudes of the anomalies with the lifetimes of the transitions. Some of the results of this work have been presented in the theoretical paper on this subject by Nilsson and Rasmussen.[8]

EXPERIMENTAL DATA

The data discussed in the following sections are summarized in Table I.

Np^{237}

Np^{237}. 59.6-kev transition. Total and L-shell conversion coefficients.

This transition has been observed in the decays of Am^{241}, U^{237}, and Pu^{237}. It was identified from Am^{241} decay as E1 on the basis of its low conversion coefficient ($\leq$1.5). Since then, more detailed data have been obtained which permit a more precise calculation to be made of the conversion coefficient. The position of this gamma ray in the level scheme of Np^{237} is well known, and Fig. 1 shows the pertinent part of the level structure.

Jaffe, Passell, Browne, and Perlman[2], in a study of the radiations of Am^{241}, calculated values 0.92 ± 0.10 and 0.72 ± 0.07 for the total and L-conversion coefficients, respectively, from their measured absolute abundances of the conversion lines of the 59.6-kev transition and from the photon abundance, 0.406 ± 0.015, determined by Beling, Newton, and Rose.[1] If we use the more recent photon intensity measurement by Magnusson[9] (0.359 ± 0.007 photons per alpha) and the electron intensities of Jaffe et al,[2] the total and L-conversion coefficients become 1.0 ± 0.1 and 0.80 ± 0.08. Similarly, by use of the electron intensity data of Turner[3], the L-conversion coefficient is found to be 0.71 ± 0.03.

The total conversion coefficient may also be determined from a knowledge of the decay scheme of Am^{241}, the abundance of the 59.6-kev photon, and the relative intensities of the conversion lines of the 33.2- and 59.6-kev transitions. In the decay of Am^{241}, 99.5% of the alpha transitions populate (directly or indirectly) the 59.6-kev level[7,10,11] and this state de-excites to ground either by the 59.6-kev transition or by the cascading 26.4- and 33.2-kev transitions (see Fig. 1). Since the 33.2-kev gamma ray is highly converted, the sum of the abundances of the 59.6-kev photon plus its conversion electrons and those of the 33.2-kev transition must add up to 99.5%. From the known absolute abundance of the 59.6-kev photon and the relative abundances of the conversion lines of the 59.6- and 33.2-kev transitions, one can then calculate the conversion coefficient of the 59.6-kev transition. The relative electron abundances are available from the spectroscopic study of Am^{241} decay by Baranov and Shlyagin.[12] The total conversion coefficient of the 59.6-kev gamma

Table I

Summary of L-shell E1 conversion coefficient data.

Nucleus	Transition Energy (kev)	Experimental Conversion Coefficients $\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(T)$	Theoretical Conversion Coefficients (Sliv and Band/Rose) $\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	Conversion* Anomaly Factor	Photon Retardation Factor ($t_{exp}/t_{s.\ proton}$)
Np^{237}	59.6	0.22 ± 0.02	0.46 ± 0.05	0.12 ± 0.03	1.0 ± 0.1	0.13 /0.11	0.12 /0.10	0.13 /0.13	1.1 ± 0.2	3.1×10^5
Np^{237}	26.4	2.0	3.9	1.2	10 ± 2	0.55 /0.22	1.1 /0.55	1.4 /1.3	1.3 ± 0.5	3.8×10^5
Np^{239}	74.6	0.08 ± 0.02	0.06 ± 0.02	0.06 ± 0.02	0.31	0.084/0.072	0.066/0.055	0.063/0.061	$0.04 \begin{smallmatrix}+0.15\\-0.04\end{smallmatrix}$***	$\sim 5 \times 10^3$
Am^{243}	83.9	0.047 ± 0.011	0.057 ± 0.013	0.041 ± 0.009	0.20 ± 0.04	0.068/0.054	0.052/0.042	0.046/0.045	0.17 ± 0.10***	1.3×10^4
Pu^{239}	106.1	0.062 ± 0.007	0.071 ± 0.007	------	------	0.041/0.035	0.026/0.021	0.021/0.021	0.75 ± 0.11	2.4×10^6
Pu^{239}	61.4	0.4	------	------	------	0.13//0.10	------	------	------	----
Pa^{231}	84.2	1.3 ± 0.2	0.65 ± 0.15	0.046 ± 0.014	2.8 ± 0.4	0.064/0.055	0.042/0.037	0.039/0.039	12.8 ± 2.1	2.8×10^6
Pa^{231}	25.7	------	------	------	4.8 ± 1.0	------	------	------	0.18 ± 0.07**	4.5×10^4
Pa^{233}	86.3	0.35 ± 0.15	0.57 ± 0.26	0.08 ± 0.08	1.9 ± 0.7	0.060/0.052	0.039/0.034	0.036/0.036	6.4 ± 3.0	1.4×10^6
Pa^{233}	29.3	------	------	------	3.0 ± 0.8	------	------	------	$0.076 \begin{smallmatrix}+0.18\\-0.076\end{smallmatrix}$**	7.2×10^4
Ra^{223}	50.0	------	------	------	0.7 ± 0.2	------	------	------	$0.045 \begin{smallmatrix}+0.09\\-0.045\end{smallmatrix}$***	1.1×10^3
Ac^{227}	27.5	------	------	------	$\alpha(L)$ 2.8 ± 0.3	0.55 /0.28	1.2 /0.53	0.84 /1.1	0.24 ± 0.11**	3.3×10^4
Ac^{225}	40.0	0.23 ± 0.07	0.26 ± 0.09	0.41 ± 0.13	------	0.29 /0.21	0.32 /0.25	0.40 /0.37	0.13 ± 0.08***	$\leq 4.7 \times 10^3$

* Compared with theoretical conversion coefficients of Sliv and Band.
** From M-subshell ratios
*** From L-subshell ratios

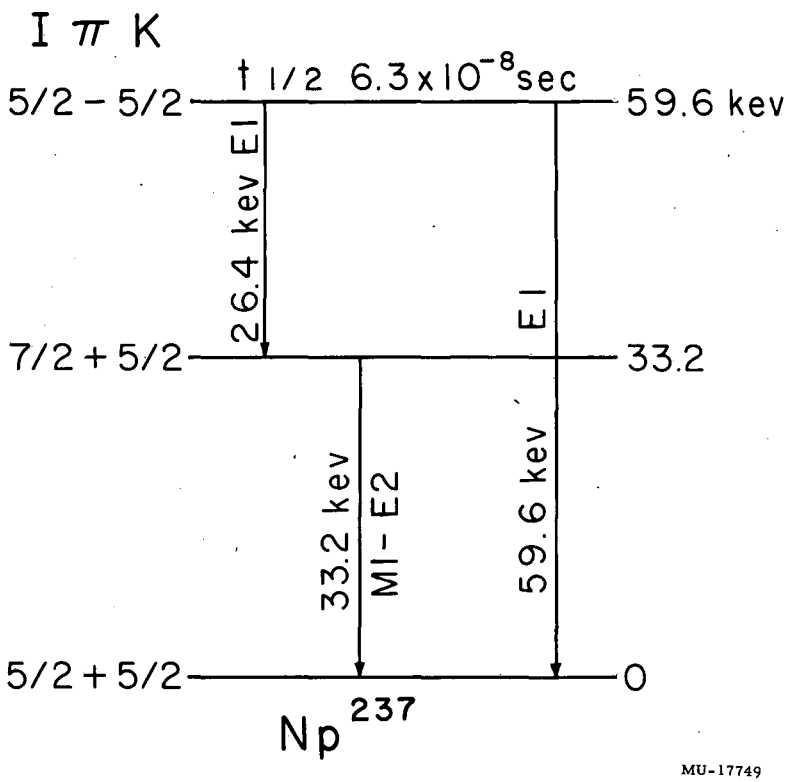

Fig. 1. Partial level scheme of Np^{237}.

ray, $\alpha(T)_{59.6}$, is then given by the expression:

$$\alpha(T)_{59.6} = \frac{0.995 - \gamma_{59.6} - e_{33.2}}{\gamma_{59.6}} = \frac{0.995 - \gamma_{59.6} - \dfrac{0.995 - \gamma_{59.6}}{1 + e_{59.6}/e_{33.2}}}{\gamma_{59.6}} .$$

Here $\gamma_{59.6}$, $e_{59.6}$, and $e_{33.2}$ are the intensities, respectively, of the 59.6-kev photon and of the conversion lines of the two transitions indicated. Unfortunately, the intensities of all of the individual conversion lines are not known with precision; in particular, the prominent L_I line of the 33.2-kev transition is very soft (~11 kev) and may be attenuated in the source and window of the detector. Since the M-shell lines have higher energies and are absorbed to a lesser extent, it was considered better to use these for comparison, with the assumption that the ratio of M lines for the two transitions is approximately the same as the ratio of total conversion-line intensities. This means that in the above expression the value for $e(M)_{59.6}/e(M)_{33.2}$ is substituted for $e_{59.6}/e_{33.2}$. Examination of available information regarding the validity of this assumption leads to the conclusion that an error as great as 10% could be introduced in the calculated $\alpha(T)_{59.6}$. This point will be explored further in later parts of this paper.

The intensity ratio $e(M)_{59.6}/e(M)_{33.2}$ taken from the graph and tables of Baranov and Shlyagin[12] is 1.7 and, when substituted along with other known quantities, gives $\alpha(T)_{59.6} = 1.1$. Within the limits of uncertainty this is in agreement with the value 1.0 recalculated, as mentioned, from the data of Jaffe et al.[2] and this value, as well as $\alpha(L)_{59.6} = 0.80 \pm 0.08$, will be used henceforth in this paper.

The first point of interest is to compare this experimental conversion coefficient with theory. The tables of Rose[5] and Sliv and Band[13] of relativistic, screened conversion coefficients which include the effects of finite nuclear size give, for a 59.6-kev E1 transition in $Z = 93$, $\alpha(L) = 0.34$ and 0.38 respectively. The discrepancy of a factor of two for $\alpha(L)_{59.6}$ (0.80, experimental vs. 0.38-0.34, theoretical) will be discussed further in the next section where the L-subshell conversion coefficients are considered.

Np^{237}. 59.6-kev transition. Subshell conversion coefficients.

The relative conversion coefficients of the 59.6-kev transition in the L-subshells have been studied by a number of different workers with results which we summarize in Table II.

Table II

Ratio of $L_I/L_{II}/L_{III}$ conversion of the 59.6-kev transition in Np^{237}.

Authors	Relative abundances $L_I/L_{II}/L_{III}$	Relative abundances $(L_I+L_{II})/L_{III}$	Parent Activity	Limits of error
Hollander, Smith, and Rasmussen[4]	1.5/3.3/1.0		Am^{241}	25%
Baranov and Shlyagin[12]	2.2/4.7/1.0		Am^{241}	
Canavan[14]	2.4/4.7/1.0		Am^{241}	
Rasmussen, Canavan, and Hollander[15]	1.6/3.2/1.0		U^{237}	
Rosenblum, Valadares, and Milsted[7]	1.7/3.3/1.0		Am^{241}	
Jaffe et al.[2]		4.4/1.0	Am^{241}	23%
Wolfson[16]		6.4/1.0	Am^{241}	20%
Turner[3]		6.4/1.0	Am^{241}	12%

All of the data in Table II have been used to arrive at the following mean value for the ratio $L_I/L_{II}/L_{III}$ = 1.9/3.8/1.0. The corresponding theoretical value is 1.1/1.0/1.1, which can be seen to be distinctly different. Now if we employ the experimental total L-shell conversion coefficient, $\alpha(L) = 0.80$, the absolute L-subshell coefficients may be determined. The results are listed in the top line of Table III and are compared with theory. It is seen that

Table III

Absolute L-subshell conversion coefficients of the 59.6-kev transition in Np^{237}.

	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(L)$
Experimental composite	0.22 ± 0.02	0.46 ± 0.05	0.12 ± 0.03	0.80 ± 0.08
Theoretical values				
(Rose)	0.11	0.10	0.125	0.34
(Sliv and Band)	0.13	0.12	0.13	0.38

agreement is good for $\alpha(L_{III})$ and that the experimental value is definitely greater for $\alpha(L_I)$ and much greater for $\alpha(L_{II})$.

Let us consider as a source of this anomaly the possibility of admixtures of multipoles other than E1 in this transition. If the experimental $\alpha(L_{III})$ is taken to be 0.15 (the highest value consistent with the error as stated), one calculates the maximum contribution of M2 radiation to be 0.015%. This amount of admixture would raise the calculated $\alpha(L_I)$ to 0.20 but would not appreciably affect $\alpha(L_{II})$. It is clear, as pointed out by Hollander, Smith, and Rasmussen,[4] that no proportion of E1 and M2 mixing can reproduce the observed predominance of L_{II} conversion because M2 radiation converts least in the L_{II} subshell. Likewise, the explanation cannot lie in E3 admixture; the maximum amount of E3 radiation, from the experimental $\alpha(L_{III})$, is 1.5×10^{-3}%, which would raise the calculated $\alpha(L_{II})$ only to 0.16 and $\alpha(L_I)$ not at all.

These anomalies are also apparent in the higher atomic shells. The ratio of conversion coefficients in the M-shells was found by Baranov and Shlyagin[12] to be $M_I/M_{II}/M_{III} = 1.3/2.8/1.0$, and the values of Rasmussen, Ganavan, and Hollander[15] are $M_I/M_{II}/M_{III}/M_{IV+V} = 1.7/3.6/1.0/0.1$. These are to be compared with Rose's[5] theoretical, point-nucleus ratios $M_I/M_{II}/M_{III}/M_{IV+V} = 1.1/0.9/1.0/0.4$. In Table IV, the M-subshell conversion coefficients of the 59.6-kev transition are given. These are calculated from the value $\alpha(T) = 1.0$ discussed above and the relative electron intensities found by various workers.

Table IV

M-subshell conversion coefficients of the 59.6-kev transition in Np^{237}.

	M_I	M_{II}	M_{III}	M_{IV+V}
Experimental composites				
(Baranov and Shlyagin)	0.051	0.11	0.039	-----
(Hollander, Smith, and Rasmussen)	~0.07	0.14	0.037	0.004
Theoretical unscreened point-nucleus value (Rose)	0.044	0.037	0.041	0.016

Anomalies in M-shell conversion are similar to those in the L-shell. The conversion of $p_{3/2}$ electrons (M_{III}) appears to agree with theory but conversion of the $p_{1/2}$ electrons (M_{II}) is definitely high and the $s_{1/2}$ electrons (M_I)

possibly so. It is also worth pointing out that the $M_{IV} + M_V$ conversion coefficient seems to be about fourfold lower than the theoretical value (see Table IV). Data are also available from the work of Rasmussen et al.[15] on N-shell conversion. The approximate subshell ratios are $N_I/N_{II}/N_{III} = 1.5/3.0/1.0$. If we assume that the theoretical values of the subshell ratios should be approximately equal as they are for L- and M-shells, it is seen that these data are consistent with anomalously high values for the N_I- and particularly the N_{II}-subshells. It appears to be the general case in the heavy-element region that conversion ratios for s and p electrons in the M- and N-shells are similar to those in the L-shell.

Np^{237}. 26.4-kev E1 transition.

It may be seen from the decay scheme in Fig. 1 that the conversion coefficient of the 26.4-kev electric dipole transition can be deduced from a knowledge of the photon intensities of it and the 59.6-kev transition together with the conversion coefficient of the 59.6-kev transition. The intensity of the 26.4-kev photon has been given by Magnusson[9] as 0.025 photons per Am^{241} disintegration. The conversion coefficient is then

$$\alpha(T)_{26.4} = \frac{e_{26.4}}{\gamma_{26.4}} = \frac{0.995 - (\gamma_{59.6} + e_{59.6} + \gamma_{26.4})}{\gamma_{26.4}} =$$

$$\frac{0.995 - [0.359 + (1.0 \times 0.359) + 0.025]}{0.025} = 10 \pm 2$$

The error of 20% includes a 10% error in the intensity of the 26.4-kev photon.

For this transition, the theoretical point-nucleus E1 conversion coefficients for the L- and M-shells are 3.1 and 1.3, giving a total of 4.4. (The M-shell value is not corrected for screening.) Although conversion in the N- and higher shells will add slightly (~0.5) to this theoretical value, the experimental number is definitely larger by about a factor of two, just as in the case of the 59.6-kev transition. The anomaly is even more pronounced if comparison is made to the finite-size nucleus theoretical coefficients. Taking the theoretical total L-shell coefficient from the tables of Sliv and Band[13], $\alpha(L) = 3.1$, and estimating the ratio of $\alpha(L)/\alpha(T)$ to be ~0.7 (as has been found

in general for higher energy transitions) we end up with a total theoretical coefficient of ~4.4. This is less than one-half of the experimental value. Comparison with the finite-size nucleus values of Rose would make the discrepancy more pronounced. These conversion coefficients are listed in Table V, where comparison can also be found for L-subshells. The L-subshell coefficients

Table V

Conversion coefficients for the 26.4-kev E1 transition in Np^{237}.

	$\alpha(T)$	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(L)$
Experimental*	10 ± 2	2.0	3.9	1.2	7.1
Theoretical values					
(Rose)		0.22	0.55	1.25	2.0
(Sliv and Band)		0.55	1.1	1.4	3.1

*None of the L-subshell coefficients were obtained directly from experimental data. See the text for explanation of the assumptions which went into the calculations.

were estimated indirectly according to the following description and are entered in Table V. Baranov and Shlyagin[12] reported the ratios $L_I/L_{II}/L_{III} = 0.7/1.5/1.0$, and Rasmussen, Canavan, and Hollander[15] reported experimental ratios $N_I/N_{II}/N_{III} = 1.7/3.3/1.0$. The difference between the experimental L ratios and N ratios may be due to error in the relative intensities of the L lines; the problem of measuring the intensities of such low-energy electrons is a very difficult one, because of extreme source- and window-thickness effects. In particular, the L_I (4.0-kev) and L_{II} (4.8-kev) electrons are expected to be attenuated with respect to the L_{III} (8.8-kev) line. Since the energy of all three N lines is about 25 kev, the relative N-subshell intensities are considered the more reliable. If we make the assumptions that the N-subshell ratios are the same as the L ratios (as found for the 59.6-kev transition[15]) and that the ratio $\alpha(L)/\alpha(T)$ is about 0.7, as is generally found,[17] we calculate coefficients of 2.0, 3.9, and 1.2 for the L_I-, L_{II}-, and L_{III}-subshells, respectively. The theoretical values of Sliv and Band and of Rose are shown for comparison in Table V. Even if we allow considerable uncertainties because of the assumptions made in arriving at the "experimental" figures, it is obvious that the anomalously high conversion coefficients originate in conversions of $s_{1/2}$ and $p_{1/2}$ electrons (L_I, L_{II}, M_I, M_{II}, etc.)

The discrepancy between the experimental and theoretical values cannot be explained by admixtures of other multipoles; no amount of M2 or E3 admixture can explain the high L_I/L_{III} conversion ratio which is deduced since the theoretical L_I/L_{III} ratio is 1.1 for M2 radiation and 0.01 for E3 radiation. Furthermore, admixture of E3 radiation cannot explain the L_{II}/L_{III} ratio of 3.3 since the theoretical L_{II}/L_{III} ratio for E3 is 1.0.

Np^{237}. Lifetimes of the 59.6- and 26.4-kev transitions.

The half life of the 59.6-kev state in Np^{237} has been measured to be $6.3\ (\pm 0.5) \times 10^{-8}$ seconds.[1] From the knowledge of the 59.6-kev photon abundance (0.359 per alpha) and of the population of this state (99.5%), one calculates the half life of the radiative transition to be 1.75×10^{-7} seconds. This value is a factor 3.1×10^5 greater than the half life calculated from the formula of Moszkowski[18] for single-proton transitions. The 26.4-kev photon, which also depopulates the 59.6-kev state, has an abundance of 0.025 per alpha; the photon half life is thus 2.5×10^{-6} seconds and the corresponding retardation factor 3.8×10^5.

Np^{239}.

A partial level scheme for Np^{239} is shown in Fig. 2 and the lowest three states are seen to be identical in assignment (and to differ only slightly in spacing) with those of Np^{237} (Fig. 1). The other level in Fig. 2 is also found in Np^{237} and is entered here only because the transitions from this state will be used in estimating the lifetimes for the E1 transitions from the 74.6-kev state.

We shall be concerned with the E1 transitions of 74.6- and 43.1-kev, but it might be mentioned that two other E1 transitions have been identified[19] and one of these (the 118-kev transition) is shown in Fig. 2. The conversion coefficients will not be discussed because accurate and detailed data are not available.

Np^{239}. 74.6-kev E1 transition. Total L-shell conversion coefficient.

The alpha spectrum of Am^{243} and associated gamma spectrum show that 99% of the transitions go through the 74.6-kev state.[20, 21] The photon

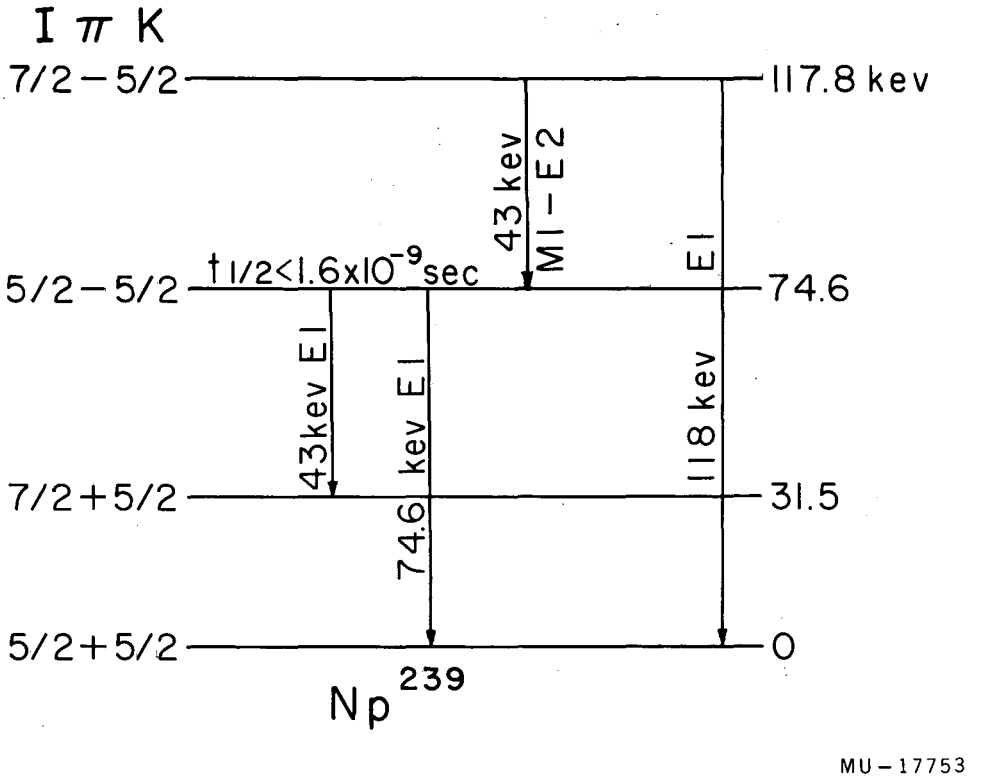

Fig. 2. Partial level scheme of Np^{239}.

intensities of the 74.6-, 43-, and 118-kev transitions are 0.69 ± 0.03, 0.04 ± 0.01, and 0.005, respectively.[19] It remains to estimate the conversion coefficient of the 43-kev transition, after which the total conversion coefficient, $\alpha(T)_{74.6}$, may be calculated by the expression

$$\alpha(T)_{74.6} = \frac{0.99 - [\gamma_{74.6} + \gamma_{43}(1 + \alpha(T)_{43})]}{\gamma_{74.6}} .$$

The value for $\alpha(T)_{43}$ is taken to be 1.2, which was obtained by using the theoretical E1 value[13] for $\alpha(L)_{43}$ (0.83) and adding an additional factor (0.35) for M, N, -- shell conversion. Although this may be inaccurate, the effect on $\alpha(T)_{74.6}$ will be only 15% for a factor-of-two error in $\alpha(T)_{43}$. From this we calculate 0.31 for $\alpha(T)_{74.6}$ and, using the value $\Sigma e_L/\Sigma e_{L+M} = 0.65 \pm 0.07$ measured by Hollander,[17] we obtain $\alpha(L)_{74.6} = 0.20$. This is to be compared (see Table VI) with the theoretical values, 0.19 and 0.21. It is seen that within the uncertainty of these measurements (probably ~20%) the experimental and theoretical values agree.

Table VI

Absolute L-subshell conversion coefficients of the 74.6-kev E1 transition in Np^{239}.

	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(L)$
Experimental composite	0.08 ± 0.02	0.06 ± 0.02	0.06 ± 0.02	0.20 ± 0.05
Theoretical values				
(Rose)	0.072	0.055	0.061	0.19
(Sliv and Band)	0.084	0.066	0.063	0.21

An independent experimental value for $\alpha(T)_{74.6}$ was given as 0.18 by Slätis,[22] who compared the intensities of the L and M conversion lines with that of the beta continuum of U^{239}. It is difficult to assess the possible uncertainties in this measurement. Similarly, Kahn[23] determined an L-shell conversion coefficient of 0.15 - 0.20 by comparing the intensities of the photons

and the L x-rays from U^{239} decay. This measurement has some uncertainties of unknown magnitude because of the absorption of some of the L x-rays in the source, estimation of the L x-ray fluorescence yield and the contributions of L x-rays resulting from transitions parallel to the 74.6-kev transition.

Np^{239}. 74.6-kev transition. Subshell conversion coefficients.

The L-subshell ratios of this transition have been measured from Am^{243} decay by Hollander[17] with a photographic-recording beta spectrograph. The results, obtained by visual comparison with intensity standards, are $L_I/L_{II}/L_{III}$ = 1.25/1.0/1.0 with an accuracy of ±20%. From these and the experimental total L-conversion coefficient (0.20) we obtain the subshell values $\alpha(L_I)$, 0.08 ± 0.02, $\alpha(L_{II})$ = 0.06 ± 0.02, and $\alpha(L_{III})$ = 0.06 ± 0.02. These are to be compared with the theoretical values in Table VI and show agreement within the experimental uncertainty.

Np^{239}. 74.6-kev transition. Lifetime.

An experimental upper limit on the half life of the 74.6-kev state has been set[24] as 1.6×10^{-9} seconds. It is also possible to estimate the lifetime roughly by making comparisons with competing transitions whose lifetimes are presumably calculable. Examination of Fig. 2 reveals two rotational bands between which are the two E1 transitions of 118-kev and 74.6 kev and, in addition, there should be an E2-M1 transition of 43 kev between the spin 7/2 and 5/2 states of the 5/2-band.

The half life for the 43-kev E2-M1 transition can be estimated in the manner to be described, and, by making use of the population of the 118-kev state (11.5%) and the intensity of the 118-kev photon (0.5%), the half life for this transition is readily calculated. Finally, the branching ratio rules of Alaga and co-authors[25] for transitions between members of one rotational band and one energy level of another, permit calculation of the lifetime for the 75-kev transition when that for the 118-kev transition is known.

The half life for the 43-kev E2-M1 transition required for the above is estimated as follows: The E2 radiative lifetime of a transition between adjacent members of a rotational band such as this is known from Coulomb excitation studies[26] to be about 100 times shorter than the value given by the

single-proton formula. Then, by using the theoretical E2 conversion coefficient, the E2 transition lifetime is determined. The composite half life of the E2-M1 mixture is then determined by assuming 57% E2 branching in conformity with the branching of the corresponding transition in Np^{237}.[4]

This method of estimation gives a half life of 2×10^{-9} seconds for the lifetime of the 74.6-kev state, which value gives reason for believing that the measured upper limit, 1.6×10^{-9} seconds, is not far from the actual value. If we take a round number of 10^{-9} seconds, this half life corresponds to a retardation of 5000 from the value calculated with the single-proton formula of Moszkowski.[18] From similar reasoning, the 44-kev E1 transition can be shown to be retarded by a factor of 2×10^4.

Am^{243}.

Am^{243}. 83.9-kev E1 transition. Total L-shell conversion coefficient.

The partial level scheme for Am^{243}, consisting of states seen from the study of Pu^{243} decay, is shown in Fig. 3. The spins and parities are those assigned by Stephens, Asaro, and Perlman.[27] Freedman and co-workers[28] reported the following photon and electron intensities relative to total Pu^{243} decay events: 21% and 1% for the photons of 84 and 42 kev, respectively; and 4% and 16% for the corresponding electrons. These data have been reexamined[29] and a total conversion coefficient for the 84-kev transition obtained, $\alpha(T) = 0.20 \pm 0.04$. The conversion line intensity ratios were given as $(L_I + L_{II})/L_{III}/(M+N) = 2.8/1.0/1.3$ with an estimated error of about 10%.[29] From these we calculate that $\alpha(L)/\alpha(T) = 0.745 \pm 0.015$ and $\alpha(L) = 0.149 \pm 0.03$. For this transition Stephens, Asaro, and Perlman[27] found $\alpha(L)/\alpha(T) = 0.69 \pm 0.03$. If we combine this with the above-mentioned value for $\alpha(T)$, we find $\alpha(L) = 0.138 \pm 0.03$. The weighted average of the two partially independent values is $\alpha(L) = 0.145 \pm 0.03$ and will be used henceforth. This compares with the theoretical value of $\alpha(L) = 0.166$ (Sliv and Band). Within experimental uncertainty there is no discrepancy between theory and experiment for $\alpha(L)$, but it will be seen that the subshell coefficients are not in agreement. These data, as well as the subshell coefficients, are summarized in Table VII.

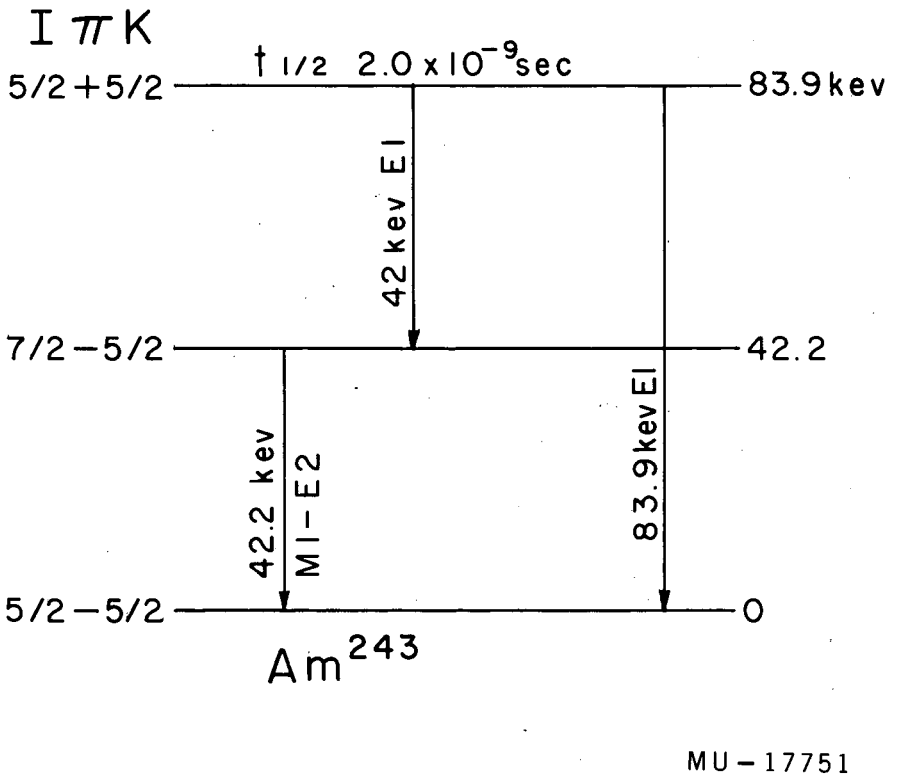

Fig. 3. Partial level scheme of Am^{243}.

Table VII

Absolute L-shell conversion coefficients of the 83.9-kev E1 transition in Am^{243}.

	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(L)$
Experimental composite	0.047 ± 0.011	0.057 ± 0.013	0.041 ± 0.009	0.145 ± 0.03
Theoretical				
(Rose)	0.054	0.042	0.045	0.141
(Sliv and Band)	0.068	0.052	0.046	0.166

Am^{243}. 83.9-kev E1 transition. Subshell conversion coefficients.

The subshell conversion coefficient ratios measured by Stephens *et al.*[27] are $L_I/L_{II}/L_{III}$ = 1.15/1.4/1.0, with an accuracy of ±20%. For comparison, the theoretical values (Sliv and Band) are $L_I/L_{II}/L_{III}$ = 1.48/1.13/1.00. It will be noted that theory has L_I conversion more prominent than L_{II}, whereas the measured values are the opposite. Other relations are also anomalous.

The absolute subshell coefficients can be obtained from these subshell ratios and the total L-shell coefficient (0.145 ± 0.03). These are listed in Table VII and compared with the theoretical values. It is seen that the experimental $\alpha(L_{III})$ agrees with theory, $\alpha(L_I)$ is possibly low, and $\alpha(L_{II})$ possibly high.

Am^{243}. 42-kev E1 transition. Photon intensity.

It is seen from Fig. 3 that there are two transitions of approximately 42 kev, of which one is mixed M1-E2 de-exciting the first rotational state and the other is an electric dipole. The electron and photon abundances of Freedman *et al.* cited above, do not distinguish these two transitions but it is easily demonstrated that essentially all of the photon intensity belongs to the electric dipole transition. That is, the assumption that the entire electron intensity, 16%, belongs to the M1-E2 transition coupled with the smallest conversion coefficient expected for an M1-E2 transition (that of a pure M1, for which $\alpha(L) = 70$) leads to the conclusion that the maximum photon intensity of the M1-E2 transition

is ~0.1% or only about one-tenth of the observed photon intensity. Since it has not been possible to determine conversion coefficients for the E1 transition, no comparison can be made with theoretical values.

Am^{243}. 84- and 42-kev transitions. Lifetime.

The half life of the 84-kev state has been measured[29] as 2.0 (±0.3) x 10^{-9} seconds. If we take the measured conversion coefficient of the 84-kev transition, the theoretical value for the 42-kev transition, and the relative intensities of the two photons, we calculate gamma-ray half lives for the 84-kev and 42-kev transitions to be 2.6 (±0.5) x 10^{-9} seconds and 5 x 10^{-8} seconds, respectively. These values correspond to retardation factors over the single-particle estimates of 1.3 (±0.3) x 10^4 and 3 x 10^4, respectively.

Pu^{239}.

Pu^{239}. 106.1-kev transition. Total and subshell coefficients.

This transition, observed from the decays of Np^{239} and Cm^{243}, has been interpreted as an electric dipole on the basis of the L-shell conversion coefficient[30] and total conversion coefficient.[31] Its position in the Pu^{239} level scheme is well known, and is shown in Fig. 4.

Ewan, Knowles, and MacKenzie[6] have obtained the most precise values of $\alpha(L_I)$ and $\alpha(L_{II})$ from their study of the beta decay of Np^{239}. Their values are: $\alpha(L_I) = 0.062 \pm 0.007$ and $\alpha(L_{II}) = 0.071 \pm 0.007$. It was not possible to measure $\alpha(L_{III})$ because of interference by an intense electron line of another transition. These authors noted that their values were distinctly higher than the point-nucleus theoretical coefficients. These and the finite-size values are shown in Table VIII for comparison with the experimental data. Ewan et al. also pointed out that the discrepancies could not be explained by M2 admixture.

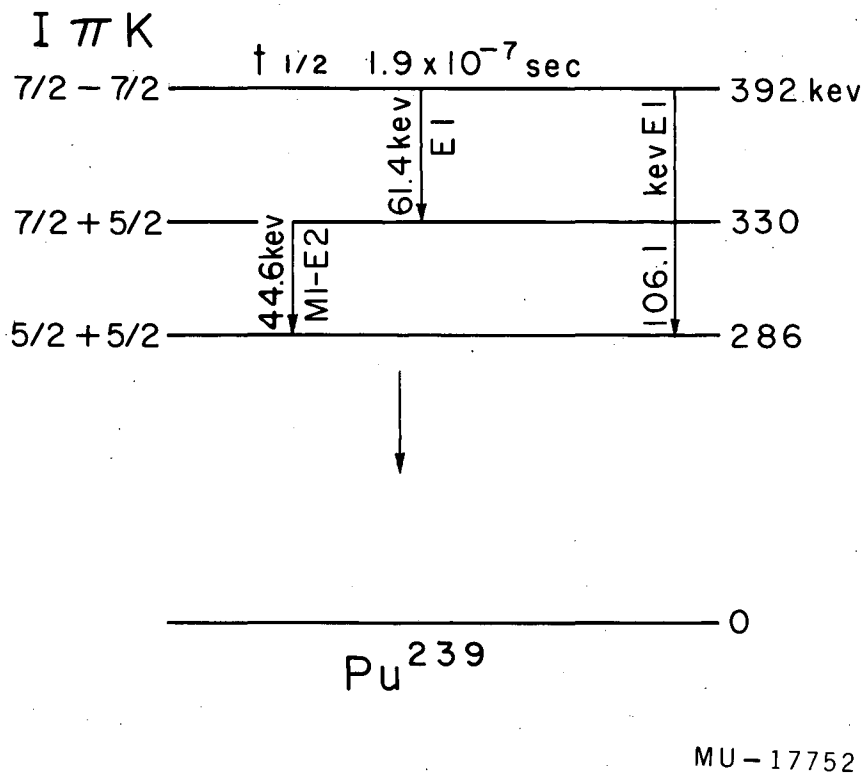

Fig. 4. Partial level scheme of Pu^{239}.

Table VIII

Absolute L-subshell conversion coefficients of the 106.1-kev transition in Pu^{239}.

	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$
Experimental	0.062 ± 0.007	0.071 ± 0.007	------
Theoretical			
Point-nucleus	0.042	0.024	0.021
Finite-size nucleus (Sliv and Band)	0.041	0.026	0.021
(Rose)	0.035	0.021	0.021

Pu^{239}. 61.4-kev transition.

The conversion coefficients for this transition (see Fig. 4) have not yet been determined with accuracy, but something can be said about the L_I subshell coefficient. It will be seen that the value we adopt is $\alpha(L_I) \simeq 0.4$, which is to be compared with the theoretical values for finite-size nucleus, 0.13 (Sliv and Band) or 0.10 (Rose).

Photons and electrons of this transition have been observed in studying the decay of Np^{239}. Using electron intensities of Fulbright[32] and photon intensities of Day[33], Engelkemeir and Magnusson[30] estimated that the total L conversion coefficient lies in the range 0.4 - 0.9 and classified the transition as E1 on this basis. However, Baranov and Shlyagin[34] showed that the L_{II} and L_{III} lines are masked by electron lines of other more intense transitions. Hollander, Smith, and Mihelich[35] also came to this conclusion but were able to obtain an approximate measurement of the L_I line intensity.

The conversion coefficient $\alpha(L_I)$ is given in terms of the following expression:

$$\alpha(L_I)_{61} = \frac{e(L_I)_{61}}{e(L)_{57}} \cdot \frac{e(L)_{57}}{\gamma_{57}} \cdot \frac{\gamma_{57}}{\gamma_{61}} .$$

The intensity ratio of the L_I line of the 61-kev transition to the L line of the 57-kev transition is given by Hollander and co-workers as ~0.012. The next ratio in the expression above is the conversion coefficient for the 57-kev E2 transition for which the theoretical value (α_L = 170) is adopted. The photon intensity ratio was measured by Jaffe[36] as γ_{57}/γ_{61} = 0.20. From these data, $\alpha(L_I)_{61}$ = 0.4. Because of the uncertainty in the conversion electron intensity ratio, this figure is probably reliable to little better than a factor of two. Partially independent calculations of $\alpha(L_I)_{61}$ can be made using other data, but these are probably even more uncertain.

Pu^{239}. 106.1-kev transition. Lifetime.

The half life of the state which de-excites by the 106- and 61-kev transitions has been measured by Engelkemeir and Magnusson[30] as 1.93×10^{-7} seconds. In order to obtain the partial half life for the 106-kev photon, correction must be made for decay by internal conversion and by the competing 61-kev transition. An intensity ratio γ_{61}/γ_{106} was sought in the alpha decay of Cm^{243} by observing γ-γ coincidences with γ_{277} and a value <0.06 was obtained.[31] Similar measurements with Np^{239} as the source gave the value 0.04 ± 0.02.[37] From this value and 0.15 for the conversion coefficient of the 106-kev transition, the photon lifetime is 2.4×10^{-7} seconds. This value is 2.4×10^{6} times longer than the half life calculated for a single-proton transition. The retardation with respect to the half life calculated for a single-neutron transition[18] would be somewhat smaller.

Pa^{231} and Pa^{233}.

The low-lying excited states of these two isotopes have certain similarities both in their energies and in their decay properties, as shown in Fig. 5. Hence Pa^{231} and Pa^{233} are discussed together in this section.

The energy levels of Pa^{231} have been studied from the beta decay of Th^{231} and from the electron-capture decay of U^{231} by Hollander, Stephens, Asaro, and Perlman.[38] Those of Pa^{233} were examined by Stephens, Asaro, and Perlman[39] by means of the Np^{237} alpha decay. The spin assignments in both cases are based upon energy-level spacings, transition multipolarities, and half lives. Also, in Pa^{231}, Newton[40] has observed the 58-kev E2 photon by Coulomb excitation.

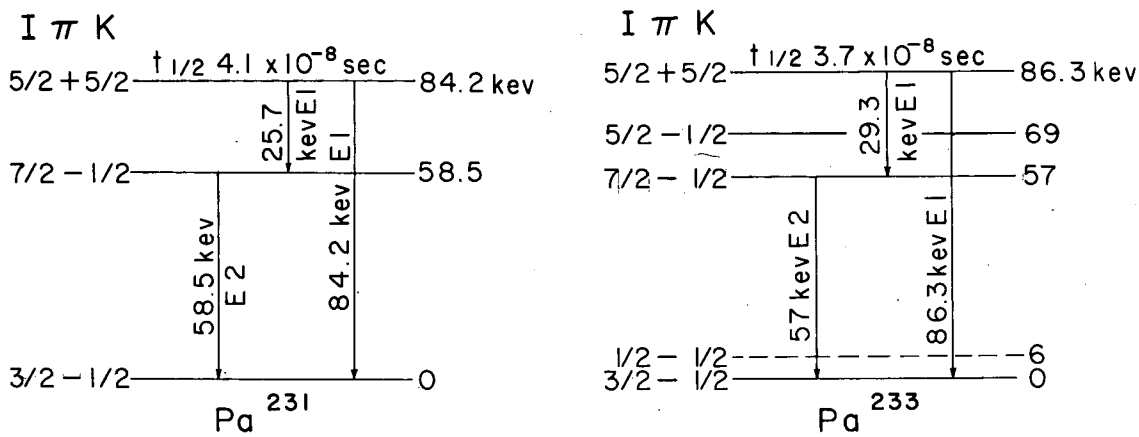

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Fig. 5. Partial level schemes of Pa^{231} and Pa^{233}.

Pa^{231}. 84.2-kev transition. Total conversion coefficient.

The 84-kev photon is prominent in the spectrum of Th^{231} and U^{231}, and the conversion lines of this transition are also strong. Coincidence studies[38] indicate that essentially all of the Th^{231} beta decay processes go through the 84-kev level and the intensity of the photon is 7.2 (±1)% relative to total Th^{231} decay intensity. (The U^{231} electron-capture decay apparently proceeds by the same path because the photon intensity noted was 7.3 (±1)%.) With this information on the decay scheme and some additional intensity data, the total conversion coefficient, $\alpha(T)_{84.2}$, may be calculated by the following expression:

$$\alpha(T)_{84.2} = \frac{1.00 - \gamma_{84} - e_{58}}{\gamma_{84}} = [(1.00/\gamma_{84}) - 1] / [1 + (e_{58}/e_{84})]$$

where γ_{84} is the intensity of the photon and e_{58} and e_{84} refer to the total intensity of conversion electrons of the 58- and 84-kev transitions. The validity of this expression is based upon the fact that the 58-kev transition is E2, hence e_{58} represents substantially all of the events which depopulate the 84-kev state in the cascade process (Fig. 5).

The ratio e_{84}/e_{58} was measured in a photographic recording spectrograph as 3.6 and 3.5 from Th^{231} and U^{231} decay, respectively.[38] A similar measurement on Th^{231} using Geiger-counter detection[41] was 3.7. We take an average value, 3.6 ± 0.3; the limit of error is chosen to be ±10% in view of the usual uncertainty in such intensity measurements. With these data, the total conversion coefficient, $\alpha(T)_{84.2}$, is 2.8 ± 0.4.

Pa^{231}. 84.2-kev transition. L-shell and subshell conversion coefficients.

The total L-shell coefficient, $\alpha(L)_{84.2}$, is readily obtained from the value of $\alpha(T)_{84.2}$ and the ratio $e(L)_{84.2}/e_{84.2}$. This ratio was found by Hollander and co-workers[38] to be 0.76 and 0.72 from Th^{231} and U^{231} decay, respectively; and Juliano[41] reported the value 0.69 from Th^{231} decay. The value we will adopt is 0.72 ± 0.04. The L-shell coefficient, $\alpha(L)_{84.2}$, then becomes 2.0 ± 0.3, which is more than an order of magnitude greater than the theoretical value, 0.14. As seen in Table I, this transition has the greatest factor of discrepancy yet noted for E1 conversion. The experimental value (2.0) is actually closer to the theoretical M1 coefficient (~6) than it is to

the E1 value, but the transition almost surely involves parity change because the 58.5-kev and 25.7-kev transitions are, respectively, E2 and E1.

The L-subshell coefficients are readily obtained from the data of Hollander and co-workers[38] and Juliano[41] on electron line intensities. Hollander et al. found the ratio $e(L_I)/e(L_{II}) = 1.6$ from measurements on U^{231} decay and 1.9 from Th^{231}. Juliano reported the same ratio as 2.5 from Th^{231} decay. We shall adopt the average value 2.0 ± 0.5. Similarly, Hollander et al. reported $e(L_{III})/e(L_I) = 0.035 \pm 0.009$. Employing $\alpha(L) = 2.0 \pm 0.3$, the following subshell coefficients result: $\alpha(L_I) = 1.3 \pm 0.2$, $\alpha(L_{II}) = 0.65 \pm 0.15$, $\alpha(L_{III}) = 0.046 \pm 0.014$. As seen from Table IX, both $\alpha(L_I)$ and $\alpha(L_{II})$ are much higher than the theoretical values, whereas $\alpha(L_{III})$ is in agreement.

Table IX

Absolute L-subshell conversion coefficients of the 84.2-kev transition in Pa^{231}.

	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(L)$
Experimental	1.3 ± 0.2	0.65 ± 0.15	0.046 ± 0.014	2.0 ± 0.3
Theoretical				
(Rose)	0.055	0.037	0.039	0.131
(Sliv and Band)	0.064	0.042	0.039	0.145

M2 admixture can, in this case also, be shown not to be the cause of the anomalously high L_I and L_{II} coefficients. If we take the maximum value of the experimental L_{III} coefficient consistent with the error limits, 0.060, and the theoretical E1 coefficient, 0.039, we find the contribution of M2 radiation to be at the most 0.02%. With this amount of M2 admixture, the theoretical mixed E1-M2 coefficient for the L_I shell becomes 0.13, still a factor of ten lower than the experimental value. The effect on the L_{II} coefficient of this amount of admixture is negligible.

Hollander et al. obtained intensities of the M, N, and O lines from U^{231} decay. The values are shown in Table X.

Table X

M, N, and O conversion coefficients for the 84.2-kev gamma.

Shell	M_I	M_{II}	M_{III}	N_I	O_I
Experimental values	0.34	0.21	0.009	0.17	0.031
Theoretical unscreened point-nucleus values	0.021	0.014	0.014	0.009*	0.005*

* These values are non-relativistic extrapolations of $\alpha(M_I)$.

As in the case of the 59.6-kev transition in Np^{237}, $\alpha(M_{III})$ is not far from the corresponding theoretical number, while $\alpha(M_I)$ and $\alpha(M_{II})$ are in distinct disagreement. It might be worth-while to note that both $\alpha(N_I)$ and $\alpha(O_I)$ are larger than the theoretical value for $\alpha(M_I)$. Brysk and Rose[42] showed for the electron-capture process (in a non-relativistic approximation) that the transition probability for $s_{1/2}$ electrons should vary approximately as the probability density of the radial wave functions (of a hydrogen-like atom) within the nucleus. If we make the same assumption for the internal conversion process, the conversion coefficients would vary as the inverse cube of the principal quantum number. With the value of 0.021 for $\alpha(M_I)$ as the basis, the non-relativistic values for $\alpha(N_I)$ and $\alpha(O_I)$ are given in Table X.

As discussed previously, the anomalously high conversion coefficient appears to originate in the $s_{1/2}$ and $p_{1/2}$ shells with no detectable anomaly in the $p_{3/2}$ shell.

Pa^{231}. 25.7-kev transition. Total conversion coefficient.

A value can be calculated for the conversion coefficient in the same way as was done in the case of the 26.4-kev transition in Np^{237}. From the measured[38] photon intensity, 12.5 ± 2%, and from our knowledge that essentially all of the beta decay of Th^{231} gives rise to the 84-kev level, we calculate $\alpha(T)_{26}$ = 4.8 ± 1.0. If the assumption of 100% population of the 84-kev state is incorrect -- for example, if there is some direct population of the 58.5-kev state -- then the actual value of the conversion coefficient will be lower than we calculate here. The sum of the theoretical L and M coefficients is 4.5, in good agreement with the experimental number. There seems little doubt that this transition is E1 because the next lowest coefficient, M1, is about 50-fold greater.

Pa^{231}. 25.7-kev transition. Subshell conversion coefficients.

Only M-subshell ratios are available for this low-energy transition. The results are summarized in Table XI.

Table XI

M-subshell conversion coefficient ratios of the 25.7-kev transition in Pa^{231}.

		$M_I/M_{II}/M_{III}/M_{IV}/M_V$
Hollander *et al.*[38]	(Th^{231} decay)	0.45/0.83/1.00/0.23/0.22
Juliano[41]	(Th^{231} decay)	0.69/0.74/1.00/0.3 /0.3
Hollander *et al.*[38]	(U^{231} decay)	0.38/0.61/1.00/0.38/0.38
Theoretical (unscreened point nucleus)[5]		0.66/0.76/1.00/0.41/0.49

The measurement on U^{231} decay should be the most accurate because the electron lines were not as distorted by source thickness as was the case with the Th^{231} sources. Accepting this and assuming that the intensities are known to about 25%, we see that the M_I/M_{III} ratio may be different from the theoretical ratio.

Pa^{231}. Lifetimes of the 84- and 26-kev transitions.

Several measurements of the half life of the 84-kev state have been made. From Th^{231} decay, Strominger and Rasmussen[43] obtained the value $4.1 \pm 0.4 \times 10^{-8}$ seconds and Mize and Starner[44] report $4.5 \pm 0.3 \times 10^{-8}$ seconds. From U^{231} decay Hollander, Stephens, Asaro, and Perlman[38] obtained the half life 4.1×10^{-8} seconds, and Hoff, Olsen, and Mann[45] report $3.7 \pm 0.4 \times 10^{-8}$ seconds from Np^{235} decay. We shall adopt the average of these values, 4.1×10^{-8} seconds.

With the photon intensities as given above, the partial half life of the 84-kev photon is 5.7×10^{-7} seconds and that of the 26-kev photon is 3.3×10^{-7} seconds. These lifetimes are longer than the single-particle estimates[18] by factors of 2.8×10^{6} and 4.5×10^{4}, respectively.

Pa^{233}. 86.3-kev transition. Total conversion coefficient.

As seen in Fig. 5, this transition is analogous to the 84-kev transition in Pa^{231}. In the present case, the level structure has been determined from the study of Np^{237} alpha decay.

The absolute abundances of the conversion electrons of $\gamma_{86.3}$ are not known, but the ratio of electron intensities of the 86-kev transition to the 57-kev E2 transition has been measured. From this ratio and the intensity of the 86-kev photon as well as some knowledge of the decay scheme, the conversion coefficient can be determined.

Magnusson and co-workers[46], studying the alpha decay of Np^{237}, found the intensity of γ_{86} to be 0.14 of the total alpha particles and the intensity of K x-rays, 0.05. (Consistent with these values are the results of Stephens and co-workers[47], who found the combined K x-ray-γ_{86} peak to have an intensity of 0.18.) We assign, somewhat arbitrarily, a limit of error of ±25% to the gamma-ray intensity. Stephens et al.[39] have interpreted most of the low-energy levels of Pa^{233} in terms of three rotational bands. This interpretation coupled with the alpha particle abundances and reinforced with gamma-gamma coincidence measurements[47] led to the figure 90 ± 5% for the amount of alpha disintegrations which give rise to the 86-kev state. It is also estimated[39] that the 57-kev state receives 3% population by paths other than from the decay of the 86-kev state. Since the 57-kev state is essentially completely de-excited by internal conversion, it is possible to derive the following expression for the conversion coefficient of the 86-kev transition:

$$\alpha(T)_{86} = [(0.93/\gamma_{86}) - 1]/[1 + (e_{57}/e_{86})] = 1.9 \pm 0.7.$$

(The intensity of γ_{86} used here has already been mentioned and the value for the conversion electron ratio (e_{57}/e_{86}) was found[39] to be 2.0 ± 0.6.)

The ratio of L-shell conversion to total conversion in this case was found to be 0.54 ± 0.11,[39] hence the coefficient $\alpha(L)_{86}$ is 1.0 ± 0.4. The theoretical value for $\alpha(L)$ is 0.135; thus there is a large discrepancy, although not as large as for the corresponding transition in Pa^{231}. The question of whether this transition in Pa^{233} is indeed E1 should be answered. The evidence is good that the cascading 29.3- and 56.9-kev are, respectively, E1 and E2; therefore, the 86.4-kev state is of opposite parity from the ground state, and with a measured $\alpha(L)$ of 1.0 only an E1 assignment is possible.

Pa^{233}. 86.3-kev transition. Subshell conversion coefficients.

The L-conversion ratios have been measured by Stephens et al.[39] as $L_I/L_{II}/L_{III}$ = 4.2/6.9/1. There are several sources of large error here: the L_{III} line is not resolved from a conversion line from the daughter isotope U^{233}. Assigning limits of error on this basis, the subshell coefficients are calculated and compared with the theoretical values in Table XII.

Table XII

L-subshell conversion coefficients of the 86.3-kev transition in Pa^{233}.

	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(L)$
Experimental composite	0.35 ± 0.15	0.57 ± 0.26	0.08 ± 0.08	1.0 ± 0.4
Theoretical				
(Rose)	0.052	0.034	0.036	0.122
(Sliv and Band)	0.060	0.039	0.036	0.135

If we assume 0.26% M2 admixture $\alpha(L_I)$ and $\alpha(L_{III})$ can be brought into agreement but $\alpha(L_{III})$ is raised only to 0.05. One can, therefore, say that $\alpha(L_{II})$ is definitely high by at least a factor of ten, $\alpha(L_I)$ is probably high, and that $\alpha(L_{III})$ is consistent with theory within a large limit of error.

Pa^{233}. 29.3-kev transition. Total conversion coefficient.

The conversion coefficient of this transition is calculated exactly as was that of the 26-kev transition in Pa^{231}. The photon intensity has been measured by Stephens et al.[47] as 0.11 and by Magnusson et al.[46] as 0.14; we shall use the average value, 0.125 ± 0.02. From this, from the fractional population of the 86-kev state (0.90 ± 0.05), and from the conversion coefficient of the 86-kev transition (1.9), the conversion coefficient of the 29-kev transition is calculated to be 3.0 ± 0.8. The theoretical $\alpha(L) + \alpha(M)$ value for an E1 transition is 3.2 with the L values of Sliv and Band or 2.5 with those of Rose; both are in agreement with the experimental number. Any assignment for this transition other than E1 is ruled out because the conversion coefficient would be more than 50-fold greater than that measured.

Pa^{233}. 29.3-kev transition. M-subshell ratios.

The L conversion lines have energies which are too low to permit them to be measured readily, but Stephens et al.[39] were able to see the M lines from a long exposure (9 months) of a Np^{237} source in a permanent-magnet spectrograph. The relative intensities on the photographic plate were compared visually, and the values for $M_I/M_{II}/M_{III}/M_{IV} + M_V$ are 0.8/0.9/1.0/0.6. The corresponding theoretical values are 0.72/0.79/1.00/0.79. The experimental intensities are reliable only to within about a factor of two because the lines were broadened by sample thickness. Within the limits of uncertainty, the experimental and theoretical values are seen to be in good agreement.

Pa^{233}. Lifetimes of the 86- and 29.3-kev transitions.

The lifetime of the 86-kev state was determined by Engelkemeir and Magnusson[48] to be 3.7×10^{-8} seconds. The partial lifetimes of the 86- and 29.3-kev photons, 2.6×10^{-7} seconds and 3.0×10^{-7} seconds, correspond respectively to retardation factors of 1.4×10^6 and 7.2×10^4 over the calculated single-proton E1 lifetimes.

Ra^{223}.

Ra^{223}. 50.0-kev transition. Total conversion coefficient.

This gamma ray is well known in the decay of Fr^{223} and Th^{227} and was shown to be an E1 transition by Pilger.[49] The level structure of Ra^{223} is extremely complex and only the part pertinent to these discussions is shown in Fig. 6. As reported by Hyde,[50] Stephens had found that the 50-kev photon was in coincidence with a prominent photon of 236 kev. Pilger showed by coincidence counting that there were 0.6 (±0.1) 50-kev photons per 236-kev photon and that the 50-kev state probably decays only to the ground state. The total conversion coefficient for γ_{50} is therefore $[(1 - 0.6)/0.6] = 0.7 \pm 0.2$. The theoretical value of $\alpha(L) + \alpha(M)$ is 0.75 (Sliv and Band L values) or 0.61 (Rose), both of which agree well with the experimental value.

Ra^{223}. 50.0-kev transition. Subshell conversion coefficients.

The L-subshell ratios were measured by Pilger as $L_I/L_{II}/L_{III}$ = 1.07/0.85/1.00. The precision of the intensity measurements is here about ±20%,

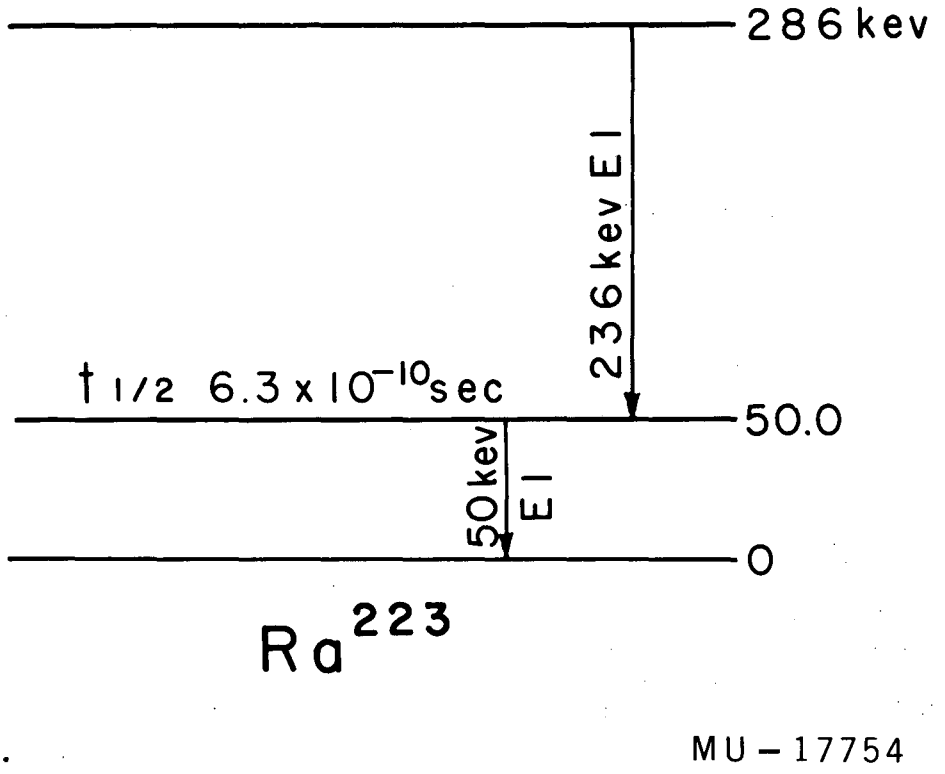

Fig. 6. Partial level scheme of Ra^{223}

but because of the possibility that there are transitions of the daughter isotope Ra^{223} which were unresolved from the lines under discussion, the accuracy of these intensities is in doubt. Bearing in mind this uncertainty, the experimental values are in excellent agreement with the theoretical ratios for an E1 transition: $L_I/L_{II}/L_{III}$ = 1.00/0.91/1.00 (Sliv and Band) or 0.93/0.85/1.00 (Rose).

Ra^{223}. Lifetime of 50-kev transition.

The half life of the 50-kev transition has been measured by Vartapetian[51] to be 6.3 (±0.7) x 10^{-10} seconds. This value represents a photon half life of 1.1 x 10^{-9} seconds, and a retardation factor of 1.1 x 10^{3} over the single-proton lifetime.

Ac^{227}.

Ac^{227}. 27.5-kev transition. Total conversion coefficient.

This transition has been observed in studies of the alpha decay of Pa^{231} and the beta decay of Ra^{227}. It was assigned as E1 by Teillac, Riou, and Desneiges[52], who obtained the value 7 for the conversion coefficient. The L-shell conversion coefficient was determined by Stephens, Asaro, and Perlman[39] by comparing the intensities of the 28-kev photon with the L x-rays from the internal conversion of this transition. This could be done by measuring alpha-photon delayed coincidences in the decay of Pa^{231} in view of the measurable lifetime of the 27.5-kev state (see below). The figure 0.52 was taken as the L-shell fluorescence yield and with the coincidence data the value $\alpha(L)$ turned out to be 2.8 ± 0.3. This is to be compared with the theoretical $\alpha(L)$ of 2.66 (Sliv and Band) or 1.74 (Rose). There is a discrepancy between the experimental value and the theoretical value of Rose.

Ac^{227}. 27.5-kev transition. Subshell conversion coefficients.

The M-subshell conversion ratios are available, and they do not agree in detail with the theoretical expectations for an E1 transition. However, in this case, it is possible to bring about agreement by assuming 0.003% M2 admixture. This comparison is summarized in Table XIII.

Table XIII

M-subshell conversion coefficient ratios of the 27.5-kev transition in Ac^{227}.

	M_I /M_{II} /M_{III}/M_{IV} + M_V
Experimental	0.9 /0.5 /1.0 /0.6
Theoretical	
(E1)	0.61/0.77/1.00/0.96
(E1 + 0.003% M2)	0.85/0.62/1.00/0.75

It will be noted that $M_I > M_{II}$ experimentally but for a pure E1 transition the reverse should be true. Although the precision of the measurements is limited (~±25%), a qualitative observation of this kind is probably reliable. It can therefore be said that if there is no M2 admixture the theory and experiment do not agree in detail but that the discrepancy can be eliminated by assuming a small M2 contribution. However, as pointed out below, there may be difficulties in reconciling this explanation with the lifetime of the transition.

Ac^{227}. Lifetime of 27.5-kev transition.

The half life of the state which de-excites by the 27.5-kev transition has been measured by Teillac *et al.*[52] as 4.2×10^{-8} seconds and by Foucher *et al.*[53] as 3.7×10^{-8} seconds. No limits of error were stated, so we shall use the average value, 4.0×10^{-8} seconds. With the assumptions that the measured delay is that of the 27.5-kev transition and that there are no other transitions from this state, we calculate the photon lifetime to be 2.0×10^{-7} seconds (a total conversion coefficient of 4.0 was used, which assumes $\alpha(L)/[\alpha(L) + \alpha(M) --] = 0.7$). This photon lifetime is longer than the single-proton E1 value by the factor 3.3×10^4. If, as mentioned above, there may be 0.003% M2 admixture, the corresponding M2 half life would be $\sim 10^{-2}$ seconds, which is just the calculated single-proton value. However, the few measured M2 lifetimes which have been reported are delayed by factors of 100 or more.

Ac^{225}.

Ac^{225}. 40.0-kev transition. Total conversion coefficient.

This transition was observed by Perlman, Stephens, and Asaro[54] and by Magnusson, Wagner, Engelkemeir, and Freedman[55] from the beta decay of Ra^{225} and assigned the multipolarity E1 on the basis of its small conversion coefficient. The value 0.9[47] was obtained for the L-conversion coefficient by a comparison of photon and L x-ray intensities in the scintillation counter spectrum, which contained only these two radiations. An L x-ray fluorescence yield of 0.5 was assumed in the calculation. The value 0.9, accurate to 30%, is in close agreement with the theoretical L-conversion coefficients of 1.01 (Sliv and Band) or 0.83 (Rose).

Ac^{225}. 40.0-kev transition. Subshell conversion coefficients.

The L-conversion ratio was measured[39] to be $L_I/L_{II}/L_{III}$ = 0.55/0.64/1.0, with a precision ±25%. The resulting absolute L-coefficients are shown in Table XIV and are seen to be in good agreement with the theoretical values.

Table XIV

L-subshell conversion coefficients of the 40.0-kev transition in Ac^{225}.

	$\alpha(L_I)$	$\alpha(L_{II})$	$\alpha(L_{III})$	$\alpha(L)$
Experimental composite	0.23 ± 0.07	0.26 ± 0.09	0.41 ± 0.13	0.9 ± 0.3
Theoretical				
(Rose)	0.21	0.25	0.37	0.83
(Sliv and Band)	0.29	0.32	0.40	1.01

Ac^{225}. Lifetime of the 40-kev transition.

The state which de-excites by this transition has a half life less than 4×10^{-9} seconds, according to Rasmussen and Stephens.[56] Using the L-conversion coefficient 0.9 and the value $\alpha(L)/[\alpha(L) + \alpha(M) + --] = 0.7$, we calculate a maximum photon half life 9×10^{-9} seconds, which corresponds to a maximum delay over the single-proton lifetime of 4.7×10^3.

DISCUSSION

We have presented in the foregoing sections a detailed account of experimental data on L-shell conversion coefficients of low-energy electric dipole transitions observed in the decays of odd-A nuclei of high atomic number. In every case in which L-subshell coefficients could be determined, the experimental data are consistent with the interpretation that the E1 conversion coefficients in the L_{III} subshell agree with the theory. In the case of the 106.1-kev transition in Pu^{239}, the L_{III} conversion coefficient is not available.

In three cases where the L conversion coefficients are known with relatively small error, it is definitely established that the experimental L_I and L_{II} coefficients are substantially larger than the theoretical values. These transitions occur in Np^{237}, Pa^{231}, and Pa^{233}. In the most striking example, the 84.2-kev transition in Pa^{231}, the L_I and L_{II} coefficients are 21 and 15 times larger than the theoretical values,[13] respectively, and in the 86.3-kev transition of Pa^{233}, the same factors are 6 and 15. These two cases are further interesting because, despite the fact that the two transitions appear to take place between the same intrinsic odd-proton states, the L_I/L_{II} ratios differ by more than a factor of three. For the 59.6-kev transition of Np^{237}, the experimental coefficients are factors 1.7 and 3.8 greater than the theoretical for the L_I and L_{II} shells, respectively.

Analysis of the data indicates a definite correlation of the anomalies with the lifetimes of the E1 photons; the more retarded the electromagnetic radiation, the greater the disparity between experimental and theoretical coefficients for the L_I and L_{II} shells.

The existence of anomalies of this type was predicted by Church and Weneser[57] in a theoretical discussion of magnetic dipole matrix elements. They point out that the finite nuclear size can give rise to additional nuclear matrix elements for the process of electron ejection which are different from that for gamma-ray emission. The connection with the correlation noted in this study is that the electron-ejection matrix element need not vanish when that for gamma-ray emission does, hence the anomaly in conversion coefficients may be related to the retardation in lifetime for the radiative transition. The theory for this problem for E1 transitions has been dealt with in some detail by Nilsson and Rasmussen.[8] Since the anomaly in conversion coefficients

is nuclear model dependent, it is not surprising that a complete description will, of necessity, be complex and involve selection rules appropriate to the nuclear model.

In Fig. 7 we have plotted a function of the L-subshell conversion coefficient anomalies against the retardation of the photon lifetime.

We have been unable to discern any systematic trends in the deviations of the L_I and L_{II} subshells individually. Hence in presenting these deviations graphically as a function of photon transition probability we define the following "total anomaly factor":

$$f = \frac{|\alpha(L_I)_{exp} - \alpha(L_I)_{theor}| + |\alpha(L_{II})_{exp} - \alpha(L_{II})_{theor}| + |\alpha(L_{III})_{exp} - \alpha(L_{III})_{theor}|}{\Sigma\alpha L_{theor}}$$

Because there seems to be no anomaly in L_{III} conversion, the last term in f is equated to zero. We have evaluated this factor for each of the transitions discussed here, and we plot these factors against the photon retardation factors ($t_{exp}/t_{theor\ single\text{-}proton}$) in Fig. 7. (In the use of the Moszkowski single-proton formula for photon lifetimes, the statistical factor was taken to be unity.) It appears from this graph that the conversion anomaly as defined here is roughly proportional to the photon retardation. The theoretical values of $\alpha(L)$ used in the calculation were those of Sliv[illegible] and Band.[13]

In several cases where only experimental M-shell coefficients are available, we have evaluated the "total anomaly factors" from M-subshell ratios alone, by equating the experimental M_{III} relative electron intensity to the theoretical M_{III} conversion coefficient. This is unsatisfactory in the sense that the theoretical unscreened, point-nucleus M-subshell ratios may not be valid, but it is the only direct comparison with theory one can presently make.

The errors shown in Fig. 7 have been derived from the error limits quoted in the text by standard statistical methods, with the assumption that all errors are standard deviations.

It is seen that in those cases for which the information is most reliable (high retardation factors and large anomalies) the relation is linear with a slope of unity. It is not possible to justify fully such a simple function in terms of the theory developed by Church and Weneser[57] and by Nilsson and Rasmussen.[8] Barring fortuitous cancellations, this relationship does seem to mean that for

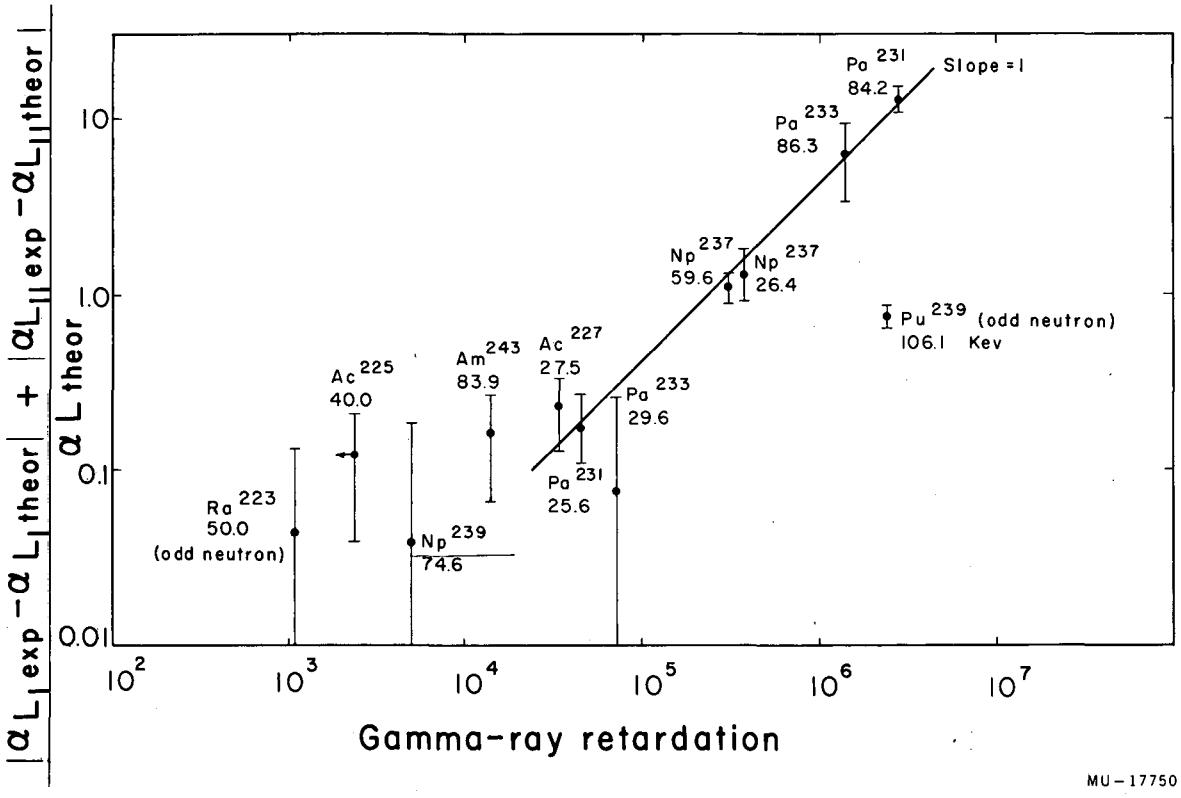

Fig. 7. El conversion coefficient anomaly vs. gamma-ray retardation. Retardation = (experimental partial photon half life) ÷ (theoretical single proton half life).

the cases examined the anomalous part of the electron-ejection matrix element does not change rapidly when that for gamma-ray emission becomes severely attenuated.

REFERENCES

1. Beling, Newton, and Rose, Phys. Rev. 87, 670 (1952); 86, 797 (1952).
2. Jaffe, Passell, Browne, and Perlman, Phys. Rev. 97, 142 (1955).
3. J. F. Turner, Phil. Mag. 46, 687 (1955).
4. Hollander, Smith, and Rasmussen, Phys. Rev. 102, 1372 (1956).
5. M. E. Rose, Internal Conversion Coefficients (North Holland Publishing Co., Amsterdam; Interscience Publishing Co., New York, 1958) and earlier privately circulated tables of conversion coefficients.
6. Ewan, Knowles, and MacKenzie, Phys. Rev. 108, 1308 (1957).
7. Rosenblum, Valadares, and Milsted, J. phys. et radium 18, 609 (1957).
8. S. G. Nilsson and J. O. Rasmussen, Nuclear Phys. 5, 617 (1958).
9. L. B. Magnusson, Phys. Rev. 107, 161 (1957).
10. Asaro, Reynolds, and Perlman, Phys. Rev. 87, 277 (1952); Asaro and Perlman, Phys. Rev. 93, 1423 (1954).
11. Gol'din, Tret'yakov, and Novikova, Conference of the U. S. S. R. Academy of Sciences on the Peaceful Uses of Atomic Energy, Phys. Math. Sci., p. 226 (July 1955).
12. S. A. Baranov and K. N. Shlyagin, Conference of the U. S. S. R. Academy of Sciences on the Peaceful Uses of Atomic Energy, July 1 - 5, 1955 (Akademiia Nauk, S. S. S. R., Moscow, 1955) translated by Consultants Bureau, New York, 1955, Vol. 1, p. 183.
13. L. A. Sliv and I. M. Band, Table of γ-ray Conversion Coefficients, Part 2, Physico-Technical Institute, Academy of Science, Leningrad, U. S. S. R. (1958).
14. F. L. Canavan, unpublished data (1956) reported by Hollander et al.[4]
15. Rasmussen, Canavan, and Hollander, Phys. Rev. 107, 141 (1957).
16. J. L. Wolfson (private communication cited in Ref. 2).
17. J. M. Hollander, unpublished data.
18. S. A. Moszkowski, Beta- and Gamma-Ray Spectroscopy, Kai Siegbahn, Ed. Ch. XIII (Interscience Publishers, Inc., New York; North Holland Publishing Co., Amsterdam, 1955).
19. Asaro, Stephens, and Perlman, unpublished data (1957).
20. J. P. Hummel, Ph. D. Thesis, Univ. of Calif. Rad. Lab. Report UCRL-3456 (July 1956) (unpublished).
21. Stephens, Hummel, Asaro, and Perlman, Phys. Rev. 98, 261 (1955)(A).
22. H. Slätis, Arkiv. Mat. Astron. Fysik 35A, No. 3 (1948).
23. J. H. Kahn, Oak Ridge National Laboratory Report ORNL-1089 (Nov. 1951).

24. D. Strominger, Ph. D. Thesis, Univ. of Calif. Rad. Lab. Report UCRL-3374 (June 1956) (unpublished).

25. Alaga, Alder, Bohr, and Mottelson, Dan. Mat. Fys. Medd. 29, No. 9 (1952).

26. Alder, Bohr, Huus, Mottelson, and Winther, Revs. Mod. Phys. 28, 432 (1956).

27. Stephens, Asaro, and Perlman, unpublished data (1956).

28. Freedman, Porter, Wagner, Day, and Engelkemeir, private communication to J. M. Hollander (Nov. 1957), reported in Strominger, Hollander, and Seaborg, Revs. Mod. Phys. 30, 585 (1958).

29. D. W. Engelkemeir, private communication (1958).

30. D. W. Engelkemeir and L. B. Magnusson, Phys. Rev. 99, 135 (1955).

31. Asaro, Thompson, Stephens, and Perlman, unpublished data (1957).

32. A. W. Fulbright, unpublished data, reported by Engelkemeir and Magnusson.[30]

33. P. P. Day, unpublished data, reported by Engelkemeir and Magnusson.[30]

34. S. A. Baranov and K. N. Shlyagin, Atomnaya Energia 1, 52 (1956).

35. Hollander, Smith, and Mihelich, Phys. Rev. 102, 740 (1956).

36. H. Jaffe, Ph. D. Thesis, Univ. of Calif. Rad. Lab. Report UCRL-2537 (April 1954).

37. Asaro, Stephens, and Perlman, unpublished data (1958).

38. Hollander, Stephens, Asaro, and Perlman, to be published.

39. Stephens, Asaro, and Perlman, unpublished data (1957-1958).

40. J. O. Newton, Nuclear Phys. 3, 345 (1957); 5, 218 (1958).

41. J. O. Juliano, Ph. D. Thesis, Univ. of Calif. Rad. Lab. Report UCRL-3733 (April 1957).

42. H. Brysk and M. E. Rose, Revs. Mod. Phys. 30, 1169 (1958).

43. D. Strominger and J. O. Rasmussen, Nuclear Phys. 3, 197 (1957).

44. J. P. Mize and J. W. Starner, Bull. Am. Phys. Soc., Series II, 1, 171 (1956).

45. Hoff, Olsen, and Mann, Phys. Rev. 102, 805 (1956).

46. Magnusson, Engelkemeir, Freedman, Porter, and Wagner, Phys. Rev. 100A, 1237 (1955).

47. F. S. Stephens, Jr., Ph. D. Thesis, Univ. of Calif. Rad. Lab. Report UCRL-2970 (June 1955).

48. D. Engelkemeir and L. B. Magnusson, Phys. Rev. 94, 1395 (1954).

49. R. C. Pilger, Ph. D. Thesis, Univ. of Calif. Rad. Lab. Report UCRL-3877 (1957).

50. E. K. Hyde, Phys. Rev. 94, 1221 (1954).

51. H. Vartapetian, Compt. rendus 246, 1109 (1958).

52. Teillac, Riou, and Desneiges, Compt. rendus 237, 41 (1953).

53. Foucher, Dick, Perrin, and Vartapetian, J. Phys. et radium 17, 581 (1956).

54. Perlman, Stephens, and Asaro, Phys. Rev. 98, 262A (1955). See also Ref. 47.

55. Magnusson, Wagner, Engelkemeir, and Freedman, Argonne National Laboratory Report ANL-5386 (Jan. 1955).

56. J. O. Rasmussen and F. S. Stephens, unpublished data (1954) reported in Refs. 54 and 47.

57. E. L. Church and J. Weneser, Phys. Rev. 104, 1382 (1956).

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