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INTERNAL CONVERSION OF GAMMA RADIATION IN THE L SUBSHELLS (Thesis)

By
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March 30, 1954

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ABSTRACT

Electron spectra from conversion of low energy nuclear gamma radiation (up to around 350 kev) in several isotopes of the heavier elements have been investigated using a double focusing beta spectrometer previously described.[1] The following isotopes were studied: Am^{241}, Am^{242m}, Cm^{242}, Pa^{228}, Pa^{230}, Tl^{198m}, Np^{238}, and Np^{236}. Decay schemes, some tentative, have been proposed for some of the above nuclides.

Miscellaneous data on the following isotopes, Pu^{241}, Pu^{240}, Pm^{150}, and Fr^{223}, are summarized in Appendix I.

A twin lens coincidence beta spectrometer, now in the assembly stage, is briefly described in Appendix II.

Agreement of experimentally determined L conversion ratios with the most recent theoretical calculations were generally very good except for the electric dipole case, where about twice the expected ($L_I + L_{II}$) conversion was found.

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I. INTRODUCTION

A study of conversion electron spectra in the heavier elements was undertaken with the following objectives in mind:

1. The assignment of the multipolarity of transitions between nuclear energy states by comparison of $L_I:L_{II}:L_{III}$ conversion ratios with theoretical values.
2. The comparison of multipolarities assigned in this manner with those assigned from other types of experiments.
3. The elucidation of a more complete decay scheme for each of the nuclides studied.

II. EXPERIMENTAL APPARATUS AND TECHNIQUES

A. The Double-Focusing Beta Spectrometer

The instrument used in these investigations has been previously described[1] so only the few modifications made are described here. Briefly, however, the double focusing spectrometer consists of a pancake-shaped iron magnet about 3 feet in diameter and about 1 foot in thickness with the center hollowed out so that the gap between the top and bottom pole pieces is smaller at the center than at the periphery. Thus, in addition to a co-axial magnetic field there exists a co-radial component of magnetic field. The focusing properties of these two components of magnetic field (hence the name double-focusing) are such that an image of the sample is formed at an angle of 255° from the sample position. A Geiger counter at this image position is

used as a detector. The magnetic field is varied by changing the current passing through the coil which is situated just inside the hollowed-out portion, but outside the vacuum chamber. The top half of the "pancake" may be unbolted and removed for making adjustments inside the chamber. Most of the changes made were in connection with the associated equipment such as the vacuum pumping system, the counter, the counter gas supply, the scaling unit, and the technique of sample preparation.

1. The vacuum pumping system. -- When a thin plastic counter window ruptures or a counter becomes "fatigued", it is necessary to reduce the vacuum chamber to atmospheric pressure and to raise the 700 pound top half of the iron "pancake" using an overhead block and tackle in order to gain access to the counter. Therefore a 2-inch modified Crane valve was placed between the vacuum chamber and the diffusion pump, allowing the latter to be kept operating while this operation is carried out. An additional forepump is used after reassembly until a pressure is reached at which the diffusion pump can again be opened to the system.

2. The counting system. -- End window counters were not very reliable so it was decided early to try the side window type. It was rumored that the latter type were far less subject to counter gas contaminants, counter wire type, and counter wall irregularities; and that if a length to diameter ratio of four or larger were maintained, usable Geiger voltage plateaus could be expected. All five side window counters designed and put in use counted satisfactorily even though almost none of the traditional recipes for making counters was

strictly followed. That is, no particular effort was made to keep the inside counter wall or central wire scrupulously clean or free from irregularities. Tungsten, molybdenum, platinum, and stainless steel central wires were tried with equal success. Stainless steel is recommended since it is of reasonable cost and is easily soldered. Brass was used for the counter body for three of the counters and copper for the fourth. Brass is recommended.

One of the more formidable problems in low energy beta spectrometry is the making of counter windows. The window must be of a thickness less than 100 $\mu g/cm^2$ and preferably below 50 $\mu g/cm^2$ if it is to have 100 percent transmission for electrons with energies below 20 kev. It must, of course, also be able to withstand the counter gas pressure of about 10 to 15 cm of mercury.

Support grids upon which are laid several layers of formvar, nylon, collodion, or plastic films are commonly used, but these grids often mask as much as 40 percent of the usable counter window area. With the kind assistance of Mr. Earl Hostetter of this laboratory a window grid was designed which masks less than 6 percent of the usable window area. The grids were made on the same principle as those designed by Mr. Hostetter for producing the electrostatic field in a time-of-flight isotope separator.[2] In the present application, 0.0005-inch diameter tungsten wire was pressed into a small, grooved, copper cylinder which had been soldered to a brass disk of an appropriate size. The brass disk contained an O-ring groove for the vacuum seal to the counter.

For the window material itself the writer has had consistent success with a vinyl chloride - vinyl acetate copolymer No. 1080 supplied by the Industrial Plastics Corporation, Oakland, California. The films were made in the traditional manner of allowing a drop of the liquid plastic (diluted 1:1 with No. 1080 thinner) to spread on a water surface. Distilled water to which a very small amount of concentrated ammonia had been added seemed to give the best results although the conditions did not seem to be critical. One three-layer window made in this fashion lasted for over a year of constant use at 15 cm total counter gas pressure. Its thickness was such that the transmission factor for 20 kev electrons was about 100 percent and the cutoff was approximately 4 kev. One factor in the unusually long life of this window may be the practice of keeping the counter filled with gas only when in use.

The use of a 10 percent ethylene - 90 percent argon gas mixture for the counter has been continued and gives satisfactory counter operation even with as much as 1 percent air contamination. Of course, it is more desirable to keep air contamination at a minimum. The more air contamination the higher the total gas pressure necessary to maintain the same Geiger region voltage plateau. The present counter has a 100 volt plateau at 12.5 cm total gas pressure.

3. Energy calibration of the spectrometer. -- The residual magnetic field in the iron of the spectrometer can be reproduced with some degree of confidence by carrying out a magnetization cycle before each experiment. The cycle consists of reversing the direction of the current and operating at maximum current for three

to five minutes; then for four times as long a time operating at maximum current with current flow in the same direction as is to be used in the experiment. This is followed by a momentary demagnetization with 1.5 amperes of alternating current. Even with this cycle of operations, several disturbing shifts have been noted in certain energy regions. Until such time as the spectrometer is equipped with a precise field measuring device, the instrument, in the writer's opinion, will be primarily useful for conversion line ratios and energy differences at high resolution.

Nuclides which have been useful in calibrating the instrument are Cs^{137}, I^{131}, Ta^{182}, Ir^{192}, and Am^{241}.

4. Semi-automatic operation. --The magnet current can be varied continuously very slowly by a motor and gear system. This makes possible the use of a traffic counter set to stamp at intervals of time small enough to allow the assumption of a point count for each stamp and long enough to allow the collection of enough counts for reasonably good statistics. This system was used to collect the data described below for Am^{241}, Cm^{242}, Pa^{228}, and Pa^{230}.

B. Sample Preparation

Probably the most critical of all techniques in beta spectrometry is the preparation of the radioactive samples. A much used sample-making procedure is the evaporation of a drop of water containing the radioactivity on backings of plastic films similar to those used for counter windows. The writer has had generally more favorable results using thin gold, palladium, or aluminum leaf. The advantages of these metal leaves have been two. They allow the leakage of

electric charges arising from radioactive decay which an insulating plastic film would build up to spectra-distorting proportions, and they may be more strongly heated than the plastic films. The thicknesses of these were found to be 87, 157, and 177 $\mu g/cm^2$, respectively, by weighing a known area of leaf. The strength of two layers of gold leaf is about equal to one of the palladium or aluminum. The double layer of gold leaf has been used with intermittent success as a backing for samples contained in reagents such as concentrated HF. Cases where the gold leaf failed may have been due to the choice of inferior leaves. Much more consistent success was obtained with the aluminum and palladium leaf so long as solutions of <0.1 N HNO_3 and <1 M HCl, respectively, were used to transfer the radioactivity to the backings. Even these solutions weakened the leaf, however, and it is recommended that the activity be transferred to distilled water before evaporating from these backings. In cases where a macro amount (of the order of several milligrams) of material had to be mounted, a 0.00025-inch thick platinum counting disk was used since sample thickness has a greater distorting effect on an electron spectrum than does the backing thickness.

At the present time a high geometry sublimator is in use in which the aluminum and palladium leaves are especially useful. High geometry is achieved by the use of a V-shaped filament. Sublimation of the radioactivity onto the thin metal foils assures both a uniformly thin deposit and a thin backing. The metal foils have an advantage over plastic films in their ability to withstand the heat of a nearby filament more readily. The investigations of the Tl^{198m} and

Np^{238} spectra were performed with samples sublimated onto palladium leaf.

The new sublimator is mounted inside a glove box for work with the high level activities necessary in beta spectrometry. Of course, carrier free chemistry must still be carried out on the radioactive sample to eliminate as much extraneous mass as possible. This mass does not readily sublimate onto the leaf and thereby greatly reduces the sublimation yield. It may, of course, in certain instances be possible to use the sublimation process as a means of separating certain elements from others, but to the writer's knowledge no extensive study has been made of its potentialities along this line.

III. EXPERIMENTAL DATA

A. Americium 241

1. L lines of 59.6, 43.4, 99, and 33.2 kev gamma rays. -- The electron spectrum of a sample of ~2 x 10^8 alpha counts per minute of Am^{241} on an aluminum leaf backing is shown in Figs. 1a and 1b. The electron data are summarized in Table 1. The Am^{241} was obtained very pure by milking from the Pu^{241} parent. The sample was evaporated onto the backing from a solution of the activitiy in distilled water. Since samples of intense alpha activity such as this one can not be flamed to make the material adhere to the backing, great care must be used in transferring the samples between the glove box where these samples are prepared and the spectrometer. The newer technique of high geometry sublimation reduces this great hazard since in this case the activity forms a uniform, adherent layer.

The L lines of the 59.6 kev gamma ray are indicated by the numbers 9 and 11 in Fig. 1a. It is seen that in the case of the L_I-L_{II} line the normally almost vertical forward edge gives the indication of a hip, about one-seventh of the way down from the peak. This may be interpreted as evidence that the abundance of the L_{II} line is perhaps somewhat less than that of the L_I line. The L_{III} line was clearly resolved. The abundance of the L_I-L_{II} line was corrected for the presence of the other spectral line known to occur at the same energy, namely the 43.4_M line. A similar correction was made for the 43.4_N line since it coincides in energy with the $59.6_{L_{III}}$. The abundances of the 43.4_M and 43.4_N lines were inferred from the $43.4_{\Sigma L}$ abundance and the L/M and L/N ratios observed for the 44.1 kev gamma ray in the decay of Cm^{242}. Of course, the chief difficulty in procuring reliable abundance measurements lies in the uncertainties involved with the extrapolation of the low energy tail of a given line. The author's policy has been to use well resolved lines as models for determining the tail of a line which was not so well resolved. In this case the $59.6_{L_{III}}$ line was so used. The $(L_I + L_{II})/L_{III}$ ratio for the 59.6 kev gamma ray is 4.4 ± 1. This value is in fair agreement with the value of around 6 obtained by Wolfson.[3]

From an experiment using Cm^{242} the transmission of the spectrometer was determined (see Section III-C). From this value of the transmission (0.3 percent), an absolute alpha count of the Am^{241} sample, and the assumption of 0.40 59.6 kev photons per alpha,[4] the total conversion coefficient of the 59.6 kev gamma was determined to be 0.92 ± 0.1. A previously reported value for the total conversion

Table 1

Americium 241 Electron Lines

Designation in Fig. 1a	Gamma energy (kev)	Electron energy (kev)	Conversion shell	Intensity e^-/α
5	26.3	25.0	N_{II}	0.0068
1	33.1	10.6	L_I	0.067
2		11.5	L_{II}	
7		27.3	M_I	0.018
8		28.7	M_{III}	0.0012
		31.7	N_I	0.004
3	43.4	20.8	L_I	0.017
4		21.8	L_{II}	0.035
6		25.8	L_{III}	0.039
10	56.4	38.8	L_{III}	0.0015
9	59.6	37.1	L_I-L_{II}	0.236
11		42.0	L_{III}	0.054
12		54.4	M_{II}	0.064
13		59.2	N	0.014
(Figure 1b)	99.5 ± 1	77.9	L_I-L_{II}	0.00062
		81.5	L_{III}	0.00037
		95	M	0.00031

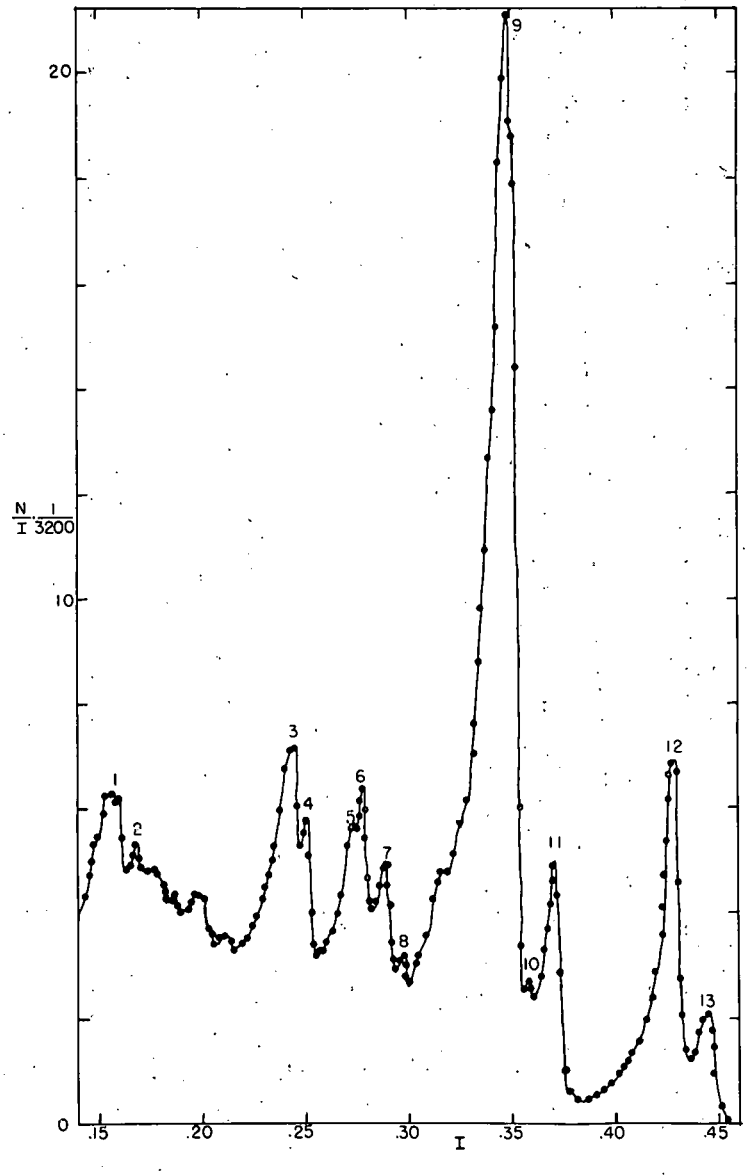


Fig. 1a — Electron spectrum of Am^{241}.

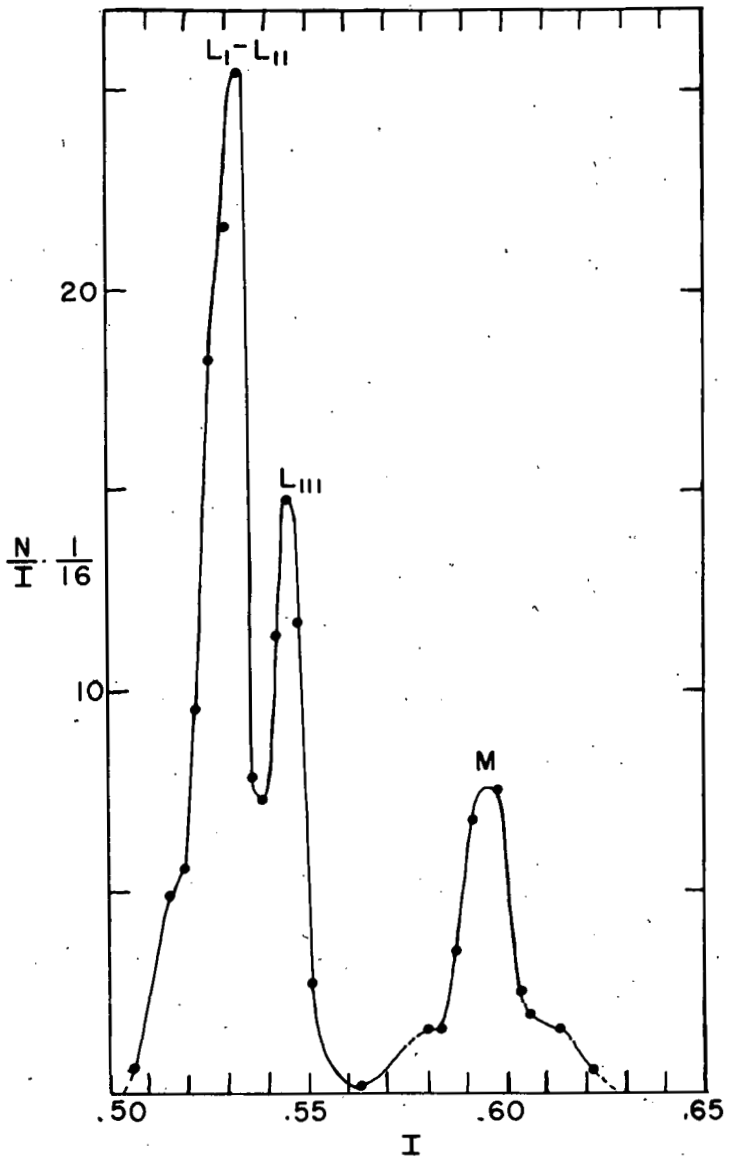


Fig. 1b—Electron spectrum of Am^{241}.

coefficient is <1.5.[5] O'Kelley[1] reports the total L conversion coefficient to be about 0.7 which compares very favorably with our value of 0.72 ± 0.07. The 59.6 kev gamma ray is given an E1 assignment on the basis of its total L conversion coefficient.

Three lines of the spectrum were assignable to the L_I, L_{II}, and L_{III} lines, respectively, of a 43.4 ± 0.5 kev gamma ray. There are large uncertainties in the abundances listed in Table 1, but it appears that the conversion ratios are not inconsistent with the interpretation of the gamma as a mixture of E2 and M1 radiation.[6] As can be seen from Fig. 1a, a line assigned as 26.3_N (line 5) appears superimposed on the tail of the line assigned as $43.4_{L_{III}}$ (line 6). From the very uncertain abundance of this 26.4 N line and the assumption that the M/N ratio is the same for the 26.3 kev as for the 59.6 kev transition (both have been given E1 assignments),[5] the abundance of the 26.4_M (line 3) line may be calculated. This value is then subtracted from the value obtained for the one line which represents both the 26.4_M and 43.4_{L_I} (line 3) to give the intensity of the 43.4_{L_I} line. In spite of the very large uncertainties involved, it is felt that a not insignificant fraction of the 43.4 kev transitions are converting in the L_I shell. Since no gamma quanta of this energy have been observed to a very low limit, (<1 percent of the 59.6 kev photon),[4,7] the assignment cannot be E1 nor can it be of very high multipolarity since all of the alpha particles are in fast coincidence (<0.15 μsec)[5] with the 59.6 kev photon. Thus we are left with a very strong probability that the radiation is either E2 or a mixture of M1 and E2.

Lines assignable as the $L_I + L_{II}$, L_{III}, and M lines of a 99.5 ± 1 kev gamma ray were observed as shown in Fig. 1b. The $(L_I + L_{II})/L_{III}$ ratio is 1.7. Jaffe has observed a 102 kev gamma ray on a scintillation spectrometer.[7] Wolfson[3] reports electron lines of a gamma ray of 99 kev with an $(L_I + L_{II})/L_{III}$ ratio of about 2. Church[8] reports a gamma ray converting in neptunium in a sample containing Am^{241} of 98.9 ± 0.3 kev, which he assigns as E2 from the L_{II}/L_{III} ratio. Since the expected[6] $(L_I + L_{II})/L_{III}$ ratio is 1.85, the E2 assignment is reasonably certain.

Assignment of lines of the 33.2 kev gamma ray was more difficult. Since Jaffe[7] has determined the energies of one of the two cascading gamma rays de-exciting the 59.62 kev level in Np^{237} to be 26.38 ± 0.04 kev, the energy of the other by difference would be (59.62 ± 0.06) - (26.38 ± 0.04) = 33.24 ± 0.10 kev. The fact that the 26.4 kev and 33.2 kev gamma rays parallel the 59.6 kev gamma ray has been established by the complex alpha spectrum.[9] As can be seen from Table 1 and Fig. 1a, lines assignable to the L_I, L_{II}, M_I, M_{III}, and N_I conversion of the 33.1 kev gamma ray have been found. The M1 assignment was made from the fact that the L_I/L_{II} ratio was >5 and that no line appeared where one would expect the $33.1_{L_{III}}$. Of course, the presence of Auger lines in that region preclude our setting any sort of limit on L_{III} conversion. Line intensities below about 17 kev are attenuated because of absorption in the window of the detector. Since the 33.2_{L_I} line appears at 10.6 kev, it is evident that only the order of magnitude of abundances are reliable. The fact that the line assigned as 33.1_{L_I} stands out so prominently in spite of the increased window

absorption gives one added confidence in the M1 assignment. This assignment is consistent with the designation of both the 59.6 and 26.4 kev gamma rays as E1 transitions.

2. Decay scheme of Am^{241} -- levels in Np^{237}. -- A decay scheme including all the presently available data is shown in Fig. 2. The positions of the 33.2, 59.6, 103, and 159 kev levels were established from the complex alpha spectrum.[9] More recent work[10] on this alpha spectrum indicates that the presence of a previously reported[9] alpha group 11 kev below the level marked 0 in Fig. 2 is due to an instrumental effect. The 270 kev level is found in the beta decay of U^{237}.[11]

Jaffe has observed gamma rays of ~210, 168, 128, and 102 kev in a scintillation spectrometer study.[7] The alpha groups populating the 270 kev and 438 kev levels would have been in too small abundance to have been observed by Asaro.[9] There is considerable doubt about the position of the 168 and 128 kev gamma rays although the indicated ones seem quite reasonable.

The L_{III} line (line 10) of a 56.4 kev gamma was observed as shown in Fig. 1a and from its abundance one can deduce a total abundance of the 56.4 kev transition (assuming E2 character which is consistent with failure to observe the gamma quanta[4, 7]). The 56 kev transition is very likely an E2 since its L_{III} peak was observed. Of course it is not at all impossible that the 56.4 kev transition includes a significant fraction of M1 radiation since the L_I peak could not be resolved.

The maximum intensity possible for this gamma transition is 0.0142 per alpha since only 1.42 percent of the alphas populate the 158.5 kev state level. The 99.5 ± 1 kev gamma ray is the other

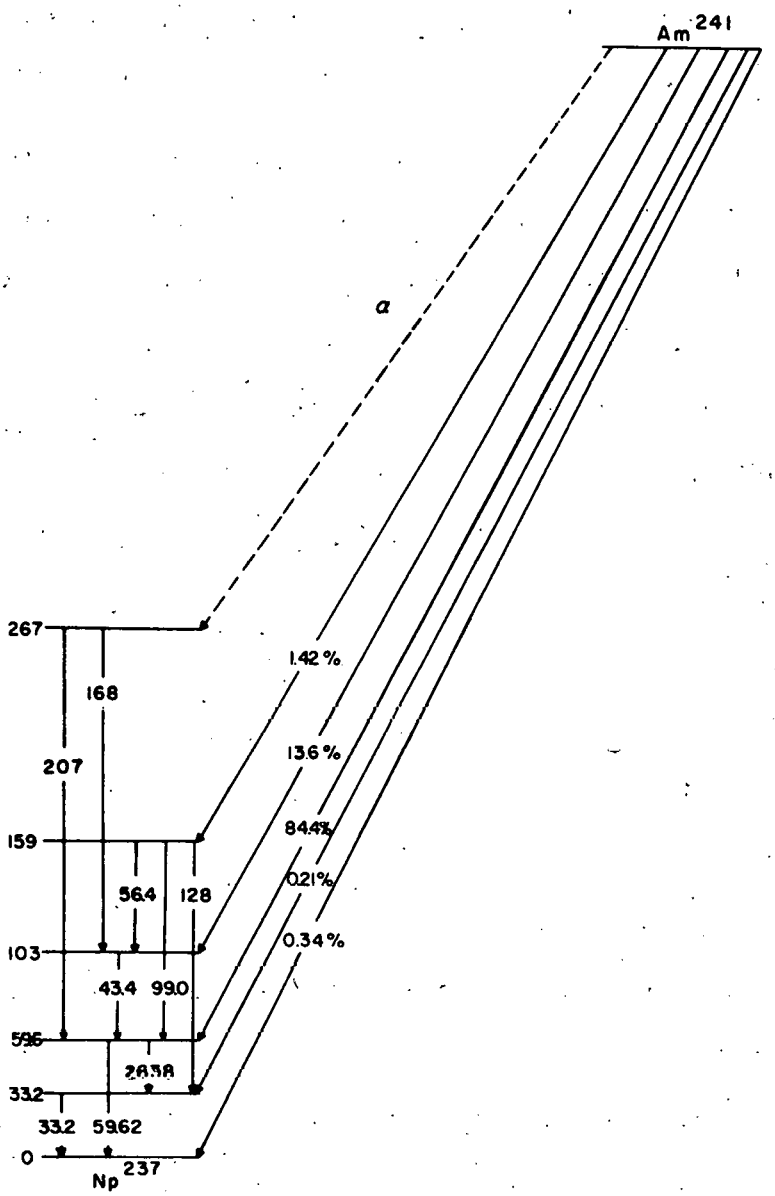
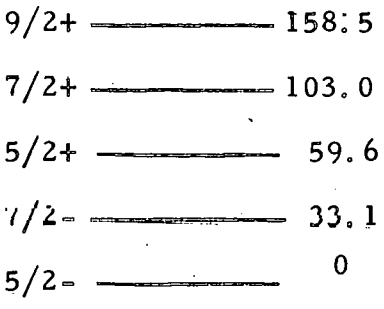


Fig. 2—Decay scheme of Am^{241}.

possible means of de-exciting the 158.5 kev level. The total abundance of the 99.5 ± 1 kev transition; i.e., photons plus electrons per alpha, was determined to be 0.0018 from the value of 0.40 ± 0.015 59.6 kev photons per alpha,[7] the 59.6 kev gamma conversion coefficient of 0.92 ± 0.10, and a scintillation spectrometer experimental value of 0.00056 photons per alpha.[7] Thus we may conclude that the 56.4 kev - 43.4 kev gamma cascade is the chief means whereby the 158.5 kev level is de-excited, but that the 98.9 kev crossover transition is not of insignificant abundance. The limits of error on the absolute abundance of the 56.4 kev transition's electrons are sufficient to encompass the value of 0.012 per alpha, which is the required value imposed by the alpha population to the 158.5 kev level and the abundance of the 98.9 kev transition.

The 26.4 kev transition has been assigned E1 character on the basis of its total conversion coefficient. This value (4.75 ± 1.5) was deduced from alpha populations of each level, the value of 0.40 ± 0.015 59.6 kev photons per alpha,[4] the total conversion coefficient of the 59.6 kev gamma ray (0.92 ± 0.10), the value of 0.04 26.4 kev photons per alpha deduced from the work of Beling and co-workers,[4] and the assumption that the decay scheme shown in Fig. 2 is correct for all states below the 158.5 kev level. From this value of the total conversion coefficient and the L/M + N ratio of the 59.6 kev E1 transition (assuming they are the same for both transitions), the total L conversion coefficient may be calculated to be 3.75 ± 1.2. This rules out an M1 or E2 assignment and is in fair agreement with that expected for an E1 assignment (2.3).[6]

Assuming the assignments of E2 character (with a possible admixture of M1) for the 98.9, 43.4, and 56.4 kev transitions, E1 character for the 59.6 and 26.4 kev transitions, and M1 character for the 33.1 kev transition are correct, we may speculate as to spin designations for the levels in Np^{237}. The ground state spin has been measured and found to be 5/2.[12,13] If the parity of the ground state is assumed to be odd, then the parity of the 33.1, 59.6, 103, and 158.5 kev levels must be odd, even, even, and even, respectively. If the first excited state is the only observed level of an odd parity "rotational band"[14] and the next three higher levels are part of an even parity "band", then the levels are as follows:



The 128 and 168 kev gamma rays which have been observed in very low abundance (0.000056 per alpha and 6.3×10^{-6} per alpha, respectively)[7] might possibly represent the transitions from the 11/2+ level (expected at 227 kev above ground) of the even parity band to the 103 kev and 59.6 kev levels respectively. The next higher negative parity level is expected about 76 kev above the ground state. This level has not been observed from the alpha decay, but its abundance is expected to be low from the trends in hindrance factors.[14]

Electron lines were observed which could be assigned to the L_I-L_{II} conversion of gamma rays of 168.5 and 209.8 kev. However, these lines were of such low and uncertain intensity that no detailed arguments can

be made from the data with any reasonable confidence. An unassigned electron line of 118 kev was also observed.

B. Americium 242m and 242

1. L lines of the 41.0 and the 43.3 kev transitions in the decay of Am^{242m}. -- The two isomers of Am^{242} with half-lives of 16 hours and about 100 years, respectively, were first observed as neutron capture products of Am^{241}.[15-17] The low energy electron lines and beta spectrum of the 16-hour isomer were studied previously by O'Kelley et al.[18] Their work is summarized in Figs. 3 and 4.

The sample used in the present investigation was produced by a neutron bombardment of Am^{241} in the MTR reactor at Arco, Idaho. The low energy line spectrum observed from this sample appears in Fig. 6. The general features of this spectrum and its interpretation were discussed by Hoff.[19] However, a revised set of abundances for the various lines will be presented, inasmuch as the relative L shell conversion coefficients are of primary interest here. This revised list of line abundances appears in Table 2. It was obtained from the same data Hoff[19] used, but is based upon a more detailed analysis of the line shapes. It is, however, still subject to very large uncertainties for several reasons. The first is the uncertainty in the exact level of scattered electron background. It is here assumed to be equal to the level of the spectrum at 0.38 amperes. A second factor is the unknown abundance of L Auger electrons which accentuate the intensity of the low energy tail of the 17.3 kev line. The third and probably most serious factor is the uncertainty involved in determining the low energy tail of unresolved lines.

Table 2

Revised Intensities of Am^{242m} Electron Lines

Gamma energy (kev)	Electron energy (kev)	Conversion shell	Revised intensities (Arbitrary Units)
41.0	17.3	CmL_{II}	520
	22.0	CmL_{III}	380
	35.5	CmM_{II}	380
	36.5	CmM_{III} or M_{IV}	
	39.9	CmN (PuM?)	100
43.3	20.9	PuL_{II}	210
	25.5	PuL_{III}	150
CmL X-rays	~16.5	CmM and N	500

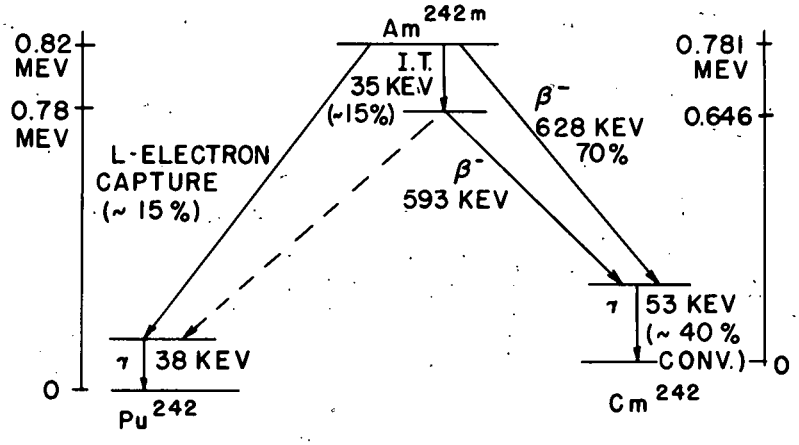


Fig. 3 — Decay scheme of Am^{242m} proposed by O'Kelley et al.[18].

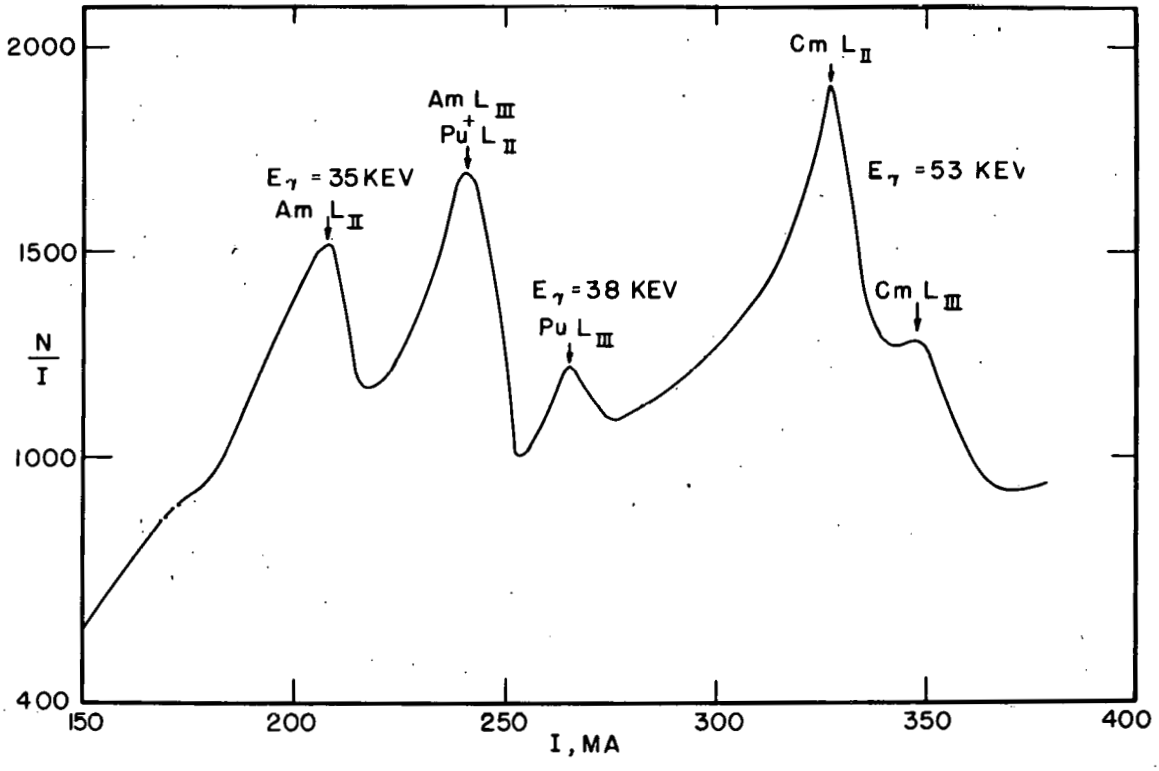


Fig. 4 — Electron spectrum observed by O'Kelley et al.[18].

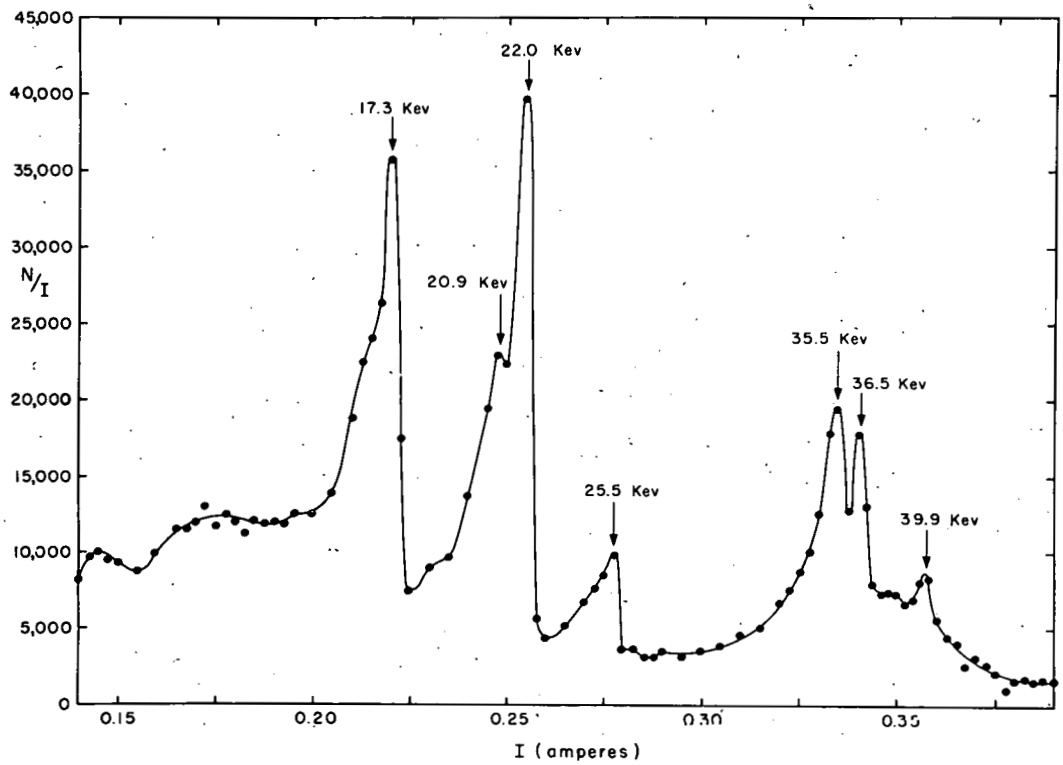


Fig. 5 — Electron spectrum of Am^{242m}.

With such uncertainties as these involved, the arguments based upon line intensities must be cautiously used. The L_{II}/L_{III} conversion ratios are 1.37 ± 0.4 and 1.4 ± 0.4 for the 41.0 and 43.3 kev gamma rays respectively. Of course, the fact that the L_{II} and L_{III} conversion electrons are of largest intensity indicates the radiation is quadrupole or higher multipole electric radiations since El and all magnetic radiations are expected to exhibit marked L_I conversion lines.[6] The assignment of E2 character to both the 41.0 and 43.3 kev radiations is consistent with the regularities noted among first excited states of even-even nuclei[20, 21] and the theoretical calculations of Gellman et al.[6]

2. Miscellaneous data on Am^{242m} and Am^{242}.--The beta spectra of both the isomers of Am^{242} were run with separate samples. Both gave straight line Fermi-Kurie plots within the limits of experimental error. The end point of the 16 hour Am^{242m} beta spectrum occurred at 620 ± 10 kev while that of the long-lived Am^{242} occurred at 585 ± 10 kev. No electron lines were observed in the long-lived Am^{242} spectrum which could not reasonably be ascribed to the Am^{241} and Cm^{242} also present in the sample.

The integrated intensity of the Am^{242m} beta spectrum at the time the electron spectrum shown in Fig. 5 was taken amounted to ~1900 ± 500 arbitrary units. This gives the value 0.7 ± 0.2 for the fraction of the Am^{242m} beta decay populating the first excited state of Cm^{242}. Hoff's[19] estimate of this fraction was larger (~1.0) since he included the low-energy shoulder of the $41.0\ L_{II}$ line with the abundance of that line. The subtraction of this shoulder from the $41.0\ L_{II}$ line

abundance is reasonably justified by the expected presence of L Auger lines in this region. Using the value of 0.57 for the L_{II} shell fluorescence yield given by Kinsey[22] (which gives 0.43 for the L_{II} Auger yield), the expected abundance of all the L_{II} Augers from the L_{II} line abundances would be about 500 arbitrary units. The presence of L_I Augers in the same region from L_I electron capture[19] added to the abundance of this line also. Thus, one can conclude that there are present sufficient L_I or L_{II} Auger electrons to account for the abundance assigned to the line in question.

3. Decay scheme of Am^{242m} and Am^{242}. -- Some very recent coincidence experiments performed by Stephens[23] at this laboratory indicate 48 ± 5 percent and 52 ± 5 percent branchings of Am^{242m} beta decay to the first excited and ground states of Cm^{242}, respectively. These experiments lend added support to the necessity for a reinterpretation of the 41.0 L_{II} line intensity. Similar experiments performed by Stephens[23] on the long-lived Am^{242} indicate a branching of 42 ± 5 percent for its beta decay to the first excited state of Cm^{242}, the remainder again populating the ground state. The limits of error quoted above for the percent of branching do not include possible errors from uncertainties in 1) the value 0.5 used for the L Auger yield and, 2) the value 0.8 used for the fraction (L conversion electrons)/(total conversion electrons) for the 41.0 ± 2 kev gamma transition.

Recent experiments performed by Church[8] give energies of 42.2 ± 0.3 and 44.6 ± 0.3 kev, respectively, for the gamma rays following Am^{242m} beta and electron capture decay, respectively. These agree with the values reported here within the limits of our absolute

error which because of calibration difficulties was ±2 kev. Consistent with the work of the author, Church also failed to observe any electron lines which could be ascribed to the isomeric transition.

The absence of any electron lines ascribable to the isomeric transition and the low intensities of americium L x-rays[7] leads one to set an upper limit of about 6 percent on the relative intensity of the isomeric transition. Church[24] interprets this absence of any observeable isomeric transition as indicating a spin difference of at least three between the two isomers. This interpretation is not at all consistent with the recent results of Stephens[23] which shows that both isomers beta decay primarily to the ground (0+) and first excited states (2+) of Cm^{242}. This peculiar situation leads one to think that the spin of both isomers might be zero and that they have different parities. This is reasonably consistent with the ft values only if the Gamow-Teller selection rules are assumed to apply. A gamma transition between a 0+ and a 0- state is absolutely forbidden both to emission of orbital electrons and to emission of gamma quanta.

Of course, another consequence of Stephens' results is that the beta spectrum maximum energies observed for each isomer quite probably correspond to the beta group populating the ground states of Cm^{242} in each case. Thus the revised decay scheme given by Hoff[19] must again be revised in light of these more recent experiments. There are sufficient uncertainties remaining, however, so as to preclude the drawing of a unique decay scheme.

C. Curium 242

1. L lines of 44.1 ± 0.5 and 102.0 ± 1 kev gamma rays. -- Previous studies on the electron spectra have been made by Prohaska,[25] Dunlavey and Seaborg,[26] and O'Kelley.[1] Prohaska found coincidences between electrons of 37.5 and 25 kev and alpha particles. Using the photographic emulsion technique, Dunlavey and Seaborg observed L and M electrons corresponding to a gamma ray of about 45 kev in coincidence with alpha particles. Using the same instrument as the author, O'Kelley observed L and M electrons corresponding to a gamma ray of 43 kev. Asaro[9] has investigated the complex alpha spectrum of Cm^{242} using a magnetic spectrograph. The present study was undertaken with the purpose of obtaining L_{II}/L_{III} conversion ratios for the 44.1 and the 102.0 kev gamma rays.

Two samples were made from about one microgram of Cm^{242}. The curium had been obtained from neutron bombardments of Am^{241} and was chemically very pure. Both samples were evaporated from a solution on a palladium leaf backing.

The electron spectra obtained with the two samples are presented in Figs. 6a and 6b. The more intense sample (about 5×10^9 alpha disintegrations per minute) was used for the very low intensity electron lines from the 102 kev transition. The lines of the more abundant 44 kev transitions were run on an accurately alpha counted sample of 2.1×10^8 disintegrations per minute. From the integrated abundance of the lines of the 44 kev transition (assuming 100 percent conversion),[9] a knowledge of the limiting inherent resolution of the spectrometer at the time of the experiment (1 percent), the accurate alpha count of the

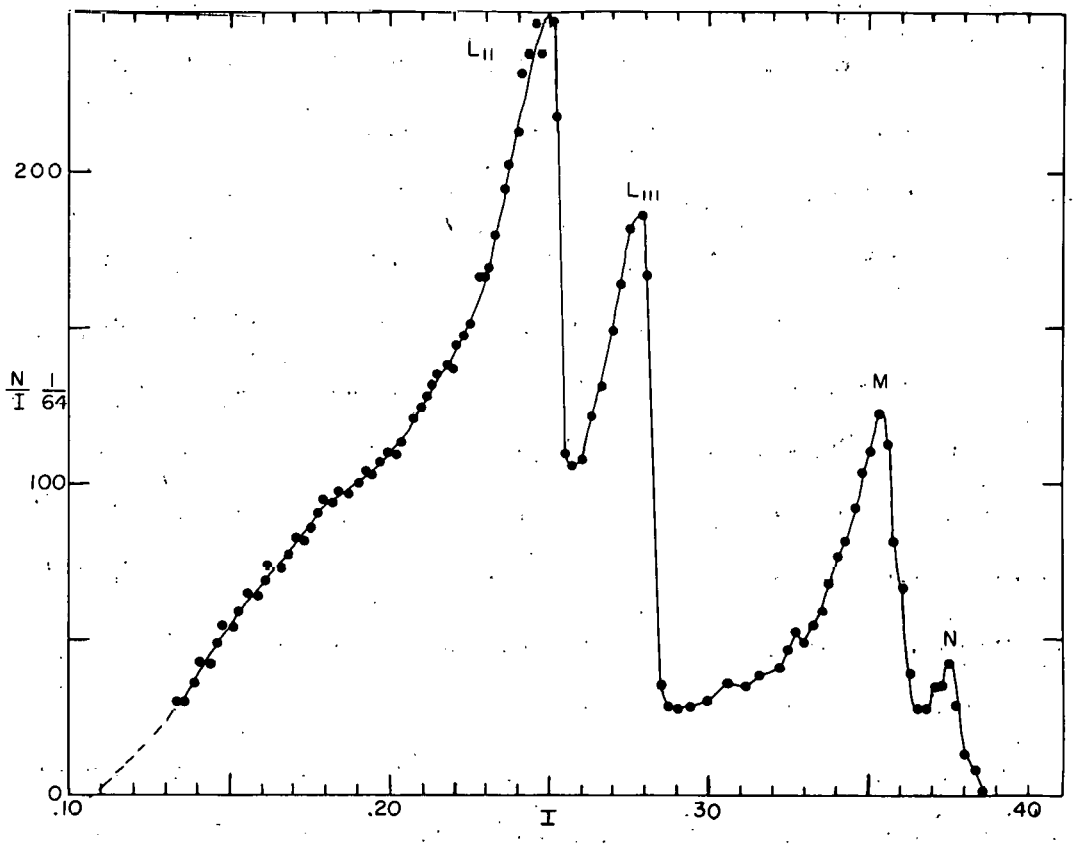


Fig. 6a — Electron spectrum of Cm^{242}.

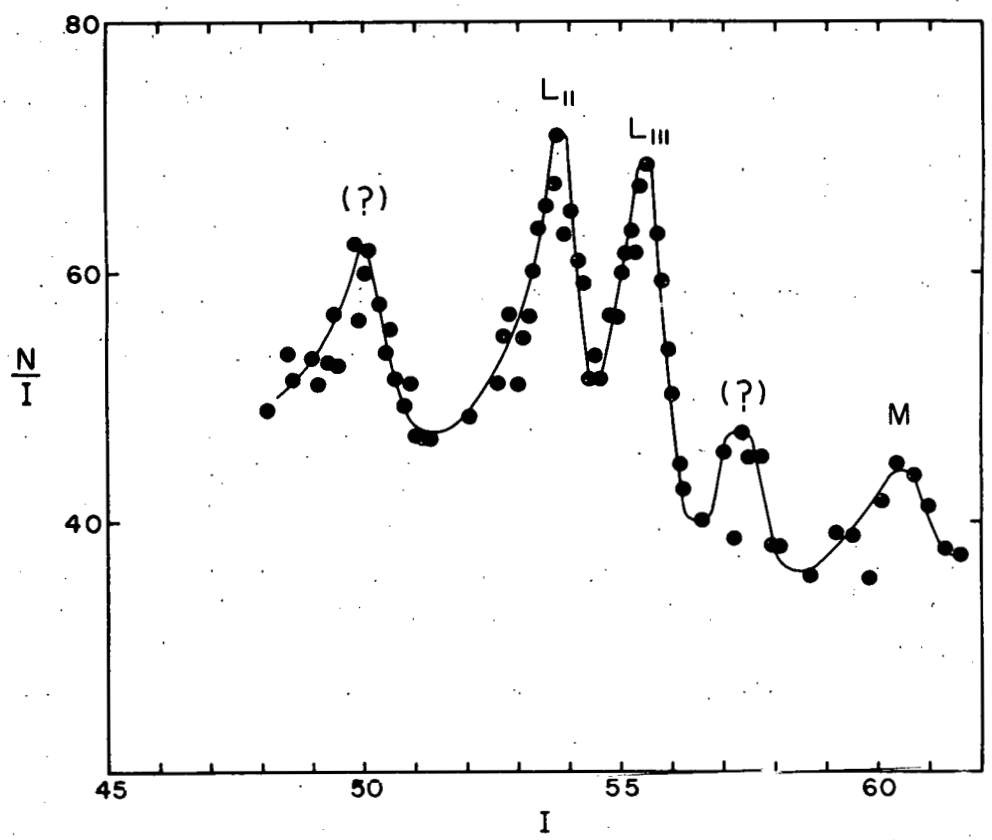


Fig. 6b — Electron spectrum of Cm^{242}.

sample, and a knowledge of the alpha population to the levels in Cm^{242} (see Fig. 7), the transmission of the instrument can be calculated. The value thus obtained was 0.3 percent of 4π.

Table 3 presents the data obtained from the two samples of Cm^{242}. The line abundances were normalized on the 44.1_N line which was observed for both samples. The L_{II}/L_{III} conversion ratio of the 44.1 kev gamma ray calculated from the data is 1.43 ± 0.2. Uncertainties in determining the low energy tails of the incompletely resolved L_{II} and L_{III} lines are the chief reason for the large limits of error quoted above. The presence of L Auger electrons in the region of the low energy tail of the $44.1_{L_{II}}$ line adds even more uncertainty to the abundance of this line.

Table 3

Curium 242 Electron Lines

Gamma transition (kev)	Electron energy (kev)	Conversion shell	Abundance (Arbitrary Units)
44.1	21.7	PuL_{II}	12.1
	26.0	PuL_{III}	8.4
	39.0	PuM_{II}	5.1
	42.8	PuN_{II}	0.9
102.0	79.7	PuL_{II}	~0.001
	84.0	PuL_{III}	~0.001

The L_{II} and L_{III} lines of the 102.0 kev transitions are seen to be in roughly the same intensity from the present work. The errors are quite large in these relative intensity measurements. This same gamma transition has been investigated from a sample of Np^{238} by

Slätis et al.,[27] and by the author (see Np^{238}). As can be seen from Fig. 6b, the scattered electron background is very high. This high scattered electron background is due to the very high intensity of electrons from the 44.1 kev transition relative to those of the 102.0 kev transition. Slätis et al.,[27] report an L_{II}/L_{III} ratio for the 102.0 kev transition of 1.55.

2. Decay scheme -- levels in Pu^{238}. --The present work is consistent with an E2 assignment for both the 44.1 kev and 102.0 kev transitions. The high total conversion coefficients (640 and 5, respectively[9]) definitely rule out E1 assignments and the absence of appreciable L_I conversion[27] in each case rule out any magnetic multipole assignments.[6] Since the lifetimes of the first two excited states of Pu^{238} have been found by coincidence experiments on Np^{238} [28] to be of the order of micro seconds, it can be assumed that the two radiations emitted in the decay of these two excited states are E1, E2, M1, or M2. Thus we can consider the E2 assignment established for both the 102.0 and the 44.1 kev transitions. Work by the author on Np^{238} (see Np^{238}) sets a low limit on the amount of M1 radiation admixed with the well-established E2 radiations.

The energies determined by Slätis et al.,[27] for these two gamma rays were 44.1 ± 0.1 kev and 102.1 ± 0.2 kev. These values are in excellent agreement with those obtained in the present study, namely 44.1 ± 0.5 kev and 102.1 ± 1 kev. The limits of error in the latter values were based upon uncertainties in calibration of the spectrometer. The use of three significant figures is justified by the much smaller uncertainties in energy differences.

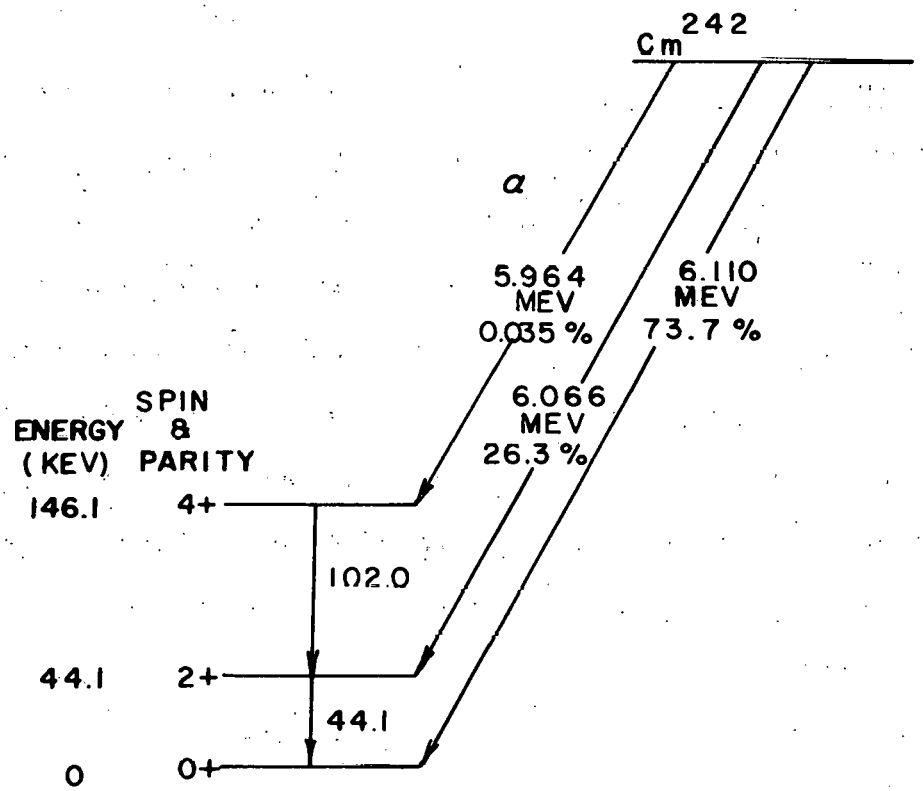


Fig. 7 — Decay scheme of Cm^{242}.

The author was not able to assign a number of electron lines of low intensity, two of which are shown in Fig. 6b. It is not impossible, energy-wise, that some of these lines arise from an unknown amount of Cm^{243} contamination or from the 157 kev radiation observed by Asaro[9] in Cm^{242} decay. The abundance of the line (not shown) which might be assigned as one of the L lines of a 157 kev gamma ray is of about the intensity expected from the work of Asaro.[9]

A decay scheme which incorporates the latest available data is shown in Fig. 7. The spin of the second excited state could equally well be 0+ or 2+.

D. Neptunium 238

1. L lines of the 44.1 and 102.0 kev gamma rays. --Neptunium 238 was first produced by Seaborg et al.[29] in bombardments of uranium with 16 Mev deuterons. Of the four groups of workers who have subsequently investigated the radiations of Np^{238} [27, 28, 30, 31] two have used methods similar to those used in the present study.[27, 28]

The purpose of the present investigation was to obtain more accurate measurements of the L conversion ratios for the 44.1 kev and 102.0 kev gamma rays.

The sample of Np^{238} used was produced by neutron bombardment of around one milligram of Np^{237} in the MTR reactor at Arco, Idaho. Chemical purification was achieved by using procedures which included an oxidation-reduction cycle and a final anion column separation. It should be noted that a significant amount of Np^{239} was produced by the second order neutron capture reaction in this bombardment. The Np^{239} was detected by the observation of certain of its more intense electron

lines. (See Fig. 8). The unmarked lines are those from Np^{239}.

The Np^{238} sample was prepared by sublimation of the activity from a tantalum filament onto a palladium leaf backing. Samples prepared in this way were invisible.

A momentum plot of part of the low energy electron spectrum of Np^{238} is shown in Fig. 8. Lines of the 102.0 kev gamma are not shown. The striking feature of the spectrum is the absence of low energy tails on the 44.1 L_{II} and 44.1 L_{III} lines. The use of the sublimation technique of sample preparation is responsible for the absence of the low energy tails. The electron line data are summarized in Table 4.

Table 4

Neptunium 238 Electron Lines

Gamma energy (kev)	Electron energy (kev)	Conversion shell	Intensity (Arbitrary Units)
44.1 ± 0.5	21.0	PuL_I	<5
	21.8	PuL_{II}	156
	26.0	PuL_{III}	124
	38.7	PuM_{II}	37
	39.7	PuM_{III}	27
	42.6	$Pu\Sigma N$	20
102.0 ± 1	78.9	PuL_I	<2
	79.7	PuL_{II}	6.5
	83.9	PuL_{III}	4.0

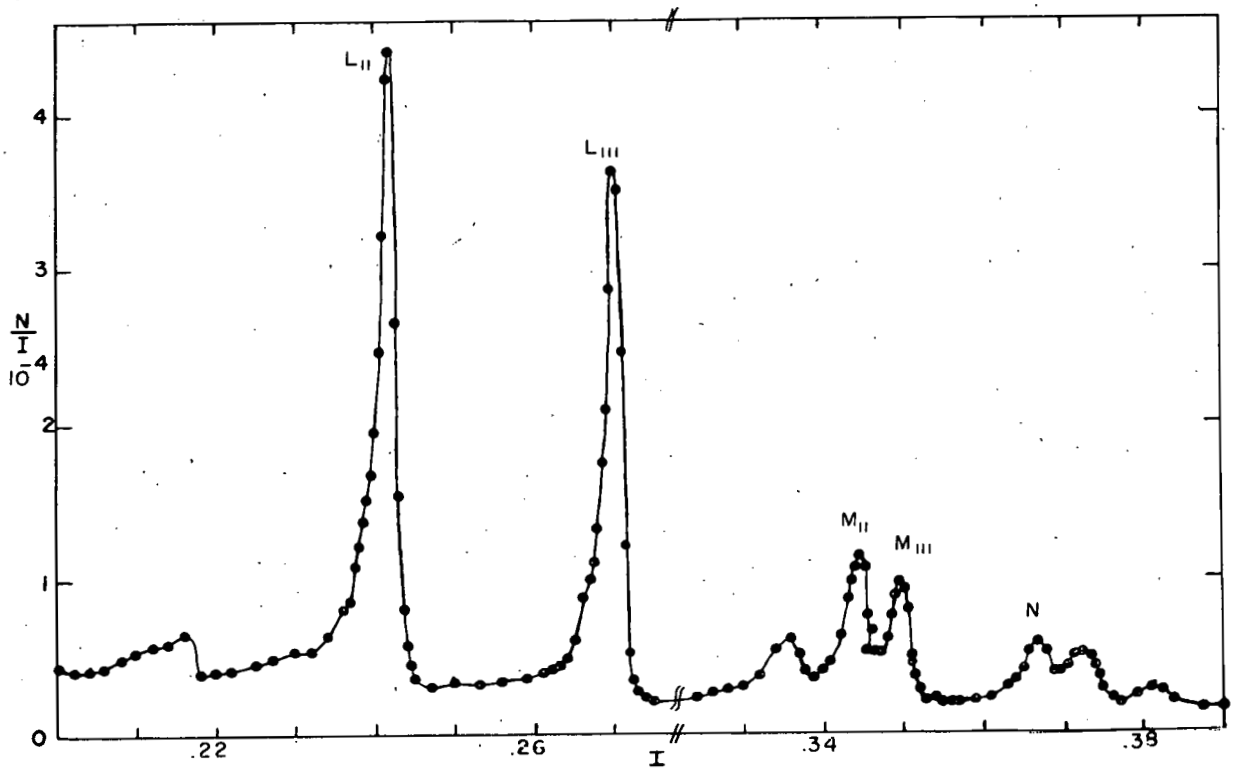


Fig. 8—Electron lines of Np^{238}.

The value (obtained in the present study) of 1.26 ± 0.05 for the 44.1 kev L_{II}/L_{III} ratio is in reasonably good agreement with the value 1.35 obtained by Slätis et al.[27] and in only fair agreement with the value 1.43 obtained for the same transition following Cm^{242} alpha decay. It should be noted that in the present study there was almost no uncertainty in the delineation of the low energy tails of the $44.1_{L_{II}}$ and $44.1_{L_{III}}$ lines. In none of the previous investigations was this uncertainty absent. Thus the line intensity determinations in the present work are subject to much smaller errors than those of previous studies.

Because of the presence of lines of the Np^{239} impurity and K Auger lines in the same energy region, the measured intensities of the $102_{L_{II}}$ and $102_{L_{III}}$ lines are somewhat uncertain. However, the value 1.62 ± 0.2 for the 102.0 kev L_{II}/L_{III} ratio is in good agreement with the value given by Slätis et al.[27] (1.56), but in poor agreement with the value (about 1) obtained for the same transition following Cm^{242} alpha decay. The poor agreement with the Cm^{242} results can probably be ascribed to the use of an inferior method of sample preparation and the large uncertainties in line shapes in the Cm^{242} investigation.

As indicated in Table 4, limits of <3 percent and <15 percent can be set for the amount of L_I conversion of the 44.1 kev and the 102.0 kev gamma rays, respectively. These values are in agreement with the calculations of Gellman et al.[6] for electric quadrupole radiations. Thus an E2 assignment for both these gamma rays is justified. The absence of appreciable L_I conversion in each case rules out the M1 and E1 assignments.

The present work confirms the spin assignment made for the first two excited states of Pu^{238} made by Asaro[9] and Slätis et al.[27] A

unique decay scheme cannot be drawn which is consistent with all the available data. However, it is felt that the ground, first excited, and second excited states in Pu^{238} have been established as shown in Fig. 7, with the possibility existing that the second excited state has a spin of 0+ or 2+.

E. Thallium 198m

1. L lines of the 48.4, 261.5, and 284 kev gamma rays. -- A 1.9-hour isotope of thallium was first observed and assigned to Tl^{198} by Orth et al.[32] Recently this activity has been independently assigned by two different groups of investigators to the decay of an isomeric state. The previously unobserved ground state decays by electron capture to Hg^{198} with a half-life of 5.3 ± 0.5 hours.

Michel and Templeton[33] of this laboratory produced these activities by the $Au^{197}(d, 3n)Tl^{198}$ reaction in the laboratory's 60-inch cyclotron. Mass separation was made on a time-of-flight isotope separator[2] and the 1.75-hour and 5.3-hour activities shown to be Tl^{198}. Bergström, Hill, and DePasquali[34] at the University of Illinois produced the same activities by bombarding mercury with 11.5-Mev deuterons. Among the many electron lines they observed were several approximately 1.9-hour lines assignable to two gamma rays converting in thallium with energies of 282.4 and 260.7 kev, and a third gamma ray of 48.7 kev whose assignment was not unambiguous. The authors suggested that all three gamma rays were in cascade from an isomeric state having the unusually high spin of 9 with odd parity. Because of the unusual decay scheme suggested, the ambiguity of the 48.7 kev gamma ray's assignment, and the uncertainties in the photographically determined

intensities, further work on this isomer was thought to be desirable.

In this investigation 1 mil gold foil was bombarded in the laboratory's 60-inch cyclotron with 40 Mev helium ions to produce the Tl^{198m} by the (d, 3n) reaction. This choice of foil thickness and bombarding energy was effective in minimizing the production of Tl^{199}.

The thallium was chemically separated from the gold target by the following process: 1) the gold was dissolved in aqua regia leaving the gold and thallium in solution as Au^{+3} and Tl^{+3} ions, respectively; 2) sulfur dioxide gas was bubbled through the solution to reduce the Tl^{+3} to Tl^{+1} and the gold to the metal; 3) the solution was centrifuged and supernatant containing Tl^{+1} and traces of Au^{+3} carried through varying purification procedures including an ethyl acetate extraction and an anion column separation; 4) the final step in the purification procedure was the use of a column containing Dowex A-2 anion resin. Hydrochloric acid was used as the eluant. The solution containing thallium was passed through the column in 6 M HCl after first oxidizing the Tl^{+} to Tl^{+3} with persulfate. The Tl^{+3} sticks at the top under these conditions. Successively more dilute hydrochloric acid solutions were used for washing out impurities, 5) the Tl^{+3} was finally stripped from the column with water saturated with sulfur dioxide gas. This carrier-free solution was used to make the spectrometer samples.

The only sample prepared which gave well resolved electron peaks was one in which the thallium was vaporized onto a palladium leaf from a tungsten filament. The palladium leaf had a thickness of 157 $\mu g/cm^2$. This vaporized sample had the dimensions 1 mm x 6 mm.

The experimental data are summarized in Table 5. The spectrum itself is shown in Figs. 9a and 9b. The K Auger lines are discussed below. The L Auger lines were also observed, but no inferences can be made therefrom because of the large and uncertain window absorption correction necessary at such low energies. In Table 5 the gamma ray energies are also given. Except for the 48.4 kev gamma ray, the energy values of the Illinois group[34] are more accurate and will be used in the following discussion.

It should be mentioned that there was initially present a very small percentage of 7.4 hour Tl^{199}. The only Tl^{199} electron line which coincided with any of those from Tl^{198m} was the 50.0_{L_I}. This line was of insignificant abundance at the time the $48.4_{L_{II}}$ and $48.4_{L_{III}}$ lines of Tl^{198m} were observed, but several hours later it was a useful calibration point for determining the energy of the 48.4 kev gamma ray.

The ratios given in Table 5 depend considerably upon the half-life assumed when the decay corrections were made. The limits of error do not necessarily encompass those introduced by the unknown uncertainty in half-life. We used a half-life value of 1.75 hours, which was measured by Michel and Templeton[33] on a separated sample.

Bergström and co-workers[34] found that the 260.7 kev gamma ray was definitely the cascade initiator, being of the M3 or M4 type with a possible admixture of electric radiation. Our K/L ratio (1.0) is in excellent agreement with that for pure M4 radiation.[35-37] According to the curves of Tralli and Lowen,[38] where L_{III}/L_I is plotted as a function of Z^2/E, the isomeric transition would be of M3 type (L_{III}/L_I = 0.80 for M3 and 1.4 for M4). However, these curves are based on

Table 5

Thallium 198m Electron Lines

Gamma energy (kev)	Electron energy (kev)	Conversion shell	Abundance (Arbitrary Units)	Conversion ratios
48.4 ± 0.2	(33.1)	Tl L_I	<1.5	L_{II}/L_{III} = 1.11 ± 0.1
	33.7	Tl L_{II}	37.3	
	35.8	Tl L_{III}	33.6	
	45.2	Tl M_{II}	18.9	
	45.6	Tl M_{III}		
	47.4	Tl N_I	5.3	
261.5 ± 2	177	Tl K	44.3	K/L = 1.0 ± 0.1
	246	Tl L_I	26.0	
	249	Tl L_{III}	17.8	
	257	Tl M N	11.9	
284 ± 3	199	Tl K	19.1	K/L = 9 ± 1
	268	Tl L_I	2.1	

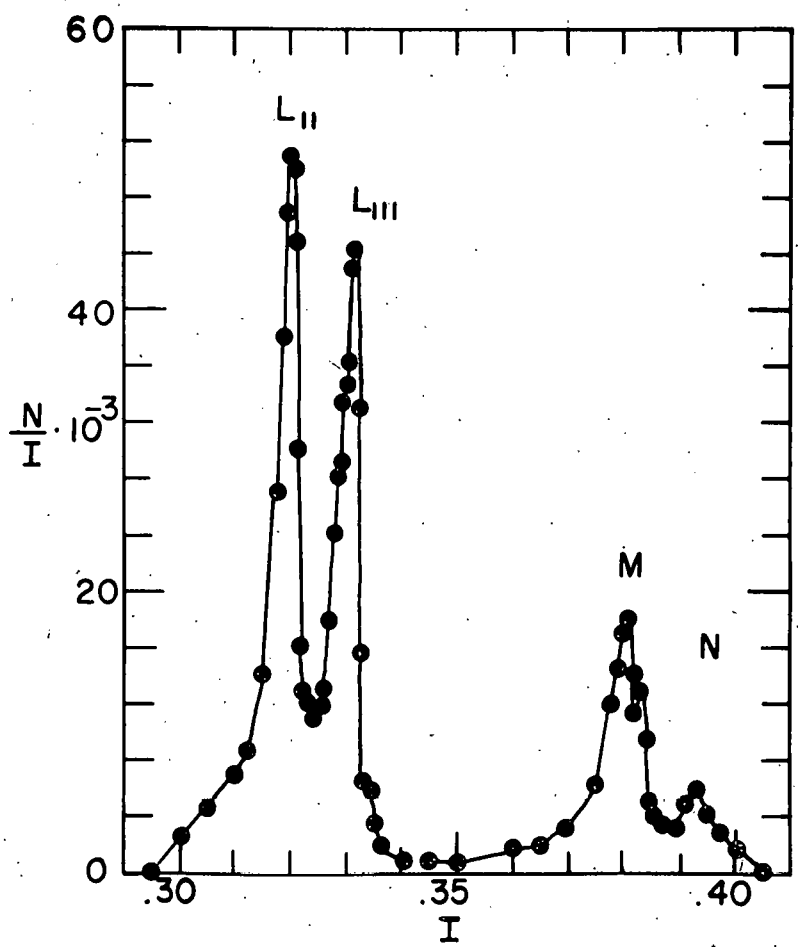


Fig. 9a — Thallium 198m electron spectrum.

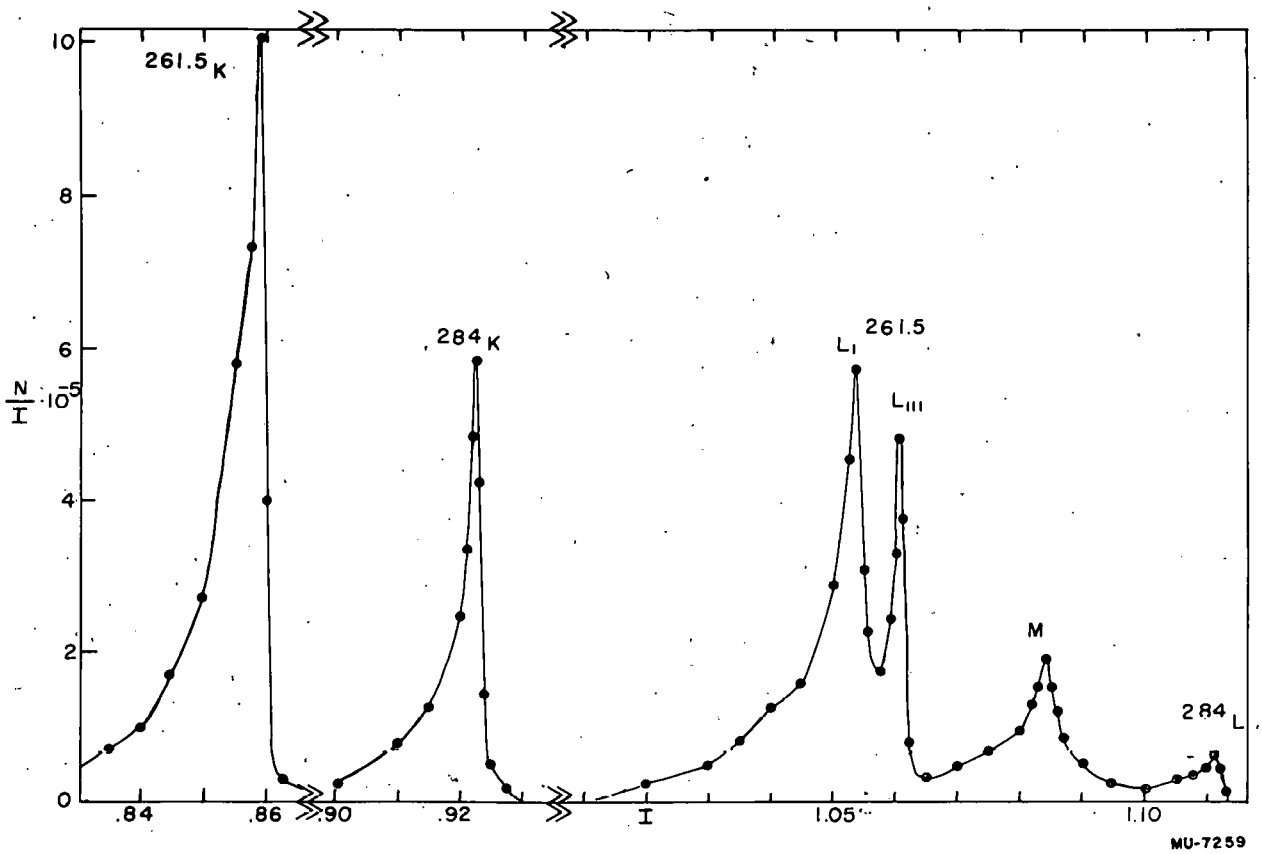


Fig. 9b — Thallium 198m electron spectrum.

approximate calculations and were performed for Z = 35. Mihelich[39] has reported a summary of experimental L_{III}/L_I conversion ratios for some M4 transitions in the region of atomic number around thallium. Fig. 10 shows this data with the inclusion of our value of 0.68 ± 0.07 for the 260.7 kev gamma ray of Tl^{198m}. It is apparent that a smooth curve could be drawn through the experimental points shown, supporting an M4 isomeric transition for Tl^{198m}.

The experimental mean life of the 260.7 kev gamma ray may be calculated using the theoretical K conversion coefficients for M4 radiation from the tables of Rose et al.[40] (18), our $260.7_K/\Sigma 260$ ratio (0.44), and the half-life of the isomeric state (1.75 hours). The mean life thus calculated is 3.7×10^5 seconds. The theoretical value one obtains using the nomogram of Weisskopf's lifetime-energy-spin formula prepared by Montalbetti[41] is 5×10^5 seconds. This agreement may be fortuitous. However, the M4 assignment is reasonable from the comparisons which have been made with existing theories and empirical correlations.

Because of the high intensity of the Tl^{199} 50.0_{L_I} line, Bergström et al.[34] were unable to resolve the $48.4_{L_{III}}$ line. As a result they designated the $48.4_{L_{II}}$ line as 48.7_{L_I}. The two lines observed in this study can only be an L_{II}-L_{III} pair, and using the 50.0_{L_I} line of Tl^{199} as a standard, one arrives at an energy of 48.4 ± 0.2 kev for the gamma ray. The L_{II}/L_{III} ratio is in better agreement with E2 (1.6) than with E1 (about 1.8) or M1 (about 900), although only M1 can be ruled out on this basis.[6] The E1 assignment was eliminated by a scintillation spectrometer experiment which determined the total conversion coefficient of the gamma ray to be

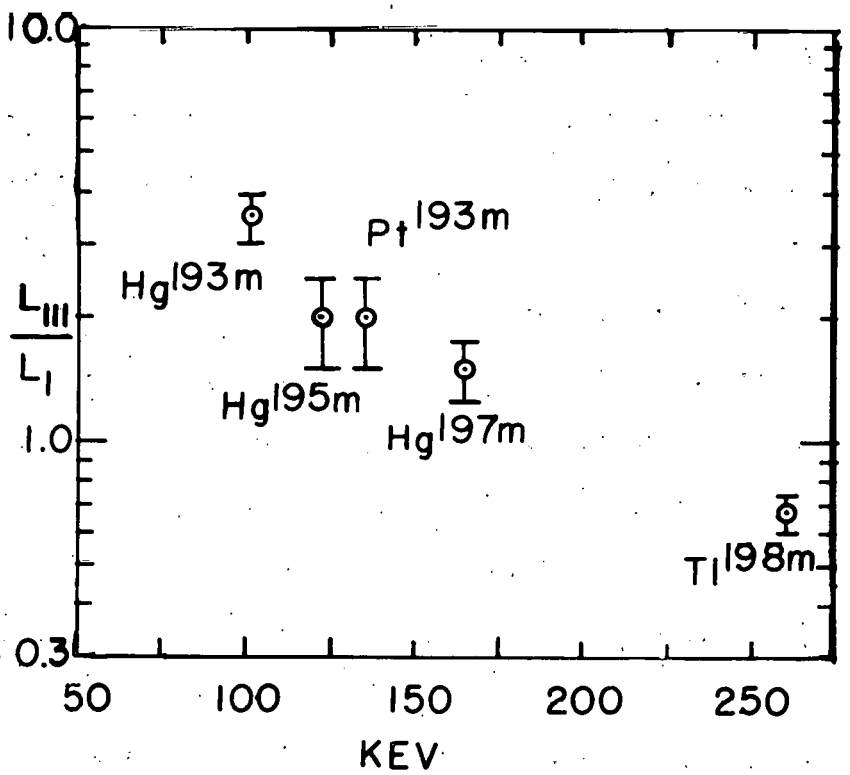


Fig. 10 — Experimental L_{III}/L_I ratios for M4 transitions in the region of atomic numbers near 80 versus energy in kev.

greater than 10. Therefore, the E2 assignment seems to be the only reasonable one.

The Illinois group has shown the 260.7 and 282.4 kev gamma rays to be converting in thallium. Coincidence measurements performed by Stephens[23] of this laboratory have shown the 282.4 kev gamma ray to be in coincidence with K x-rays. Thus it appears that the 282.4 kev and 260.7 kev gamma rays are in cascade. Assuming no electron capture from excited states of Tl^{198} the K conversion coefficient of the 282.4 kev gamma ray can be calculated from the intensity ratio $(282_K)/(260_{K+L+M...})$. The value (0.24) thus obtained may indicate a mixture of E2 (0.076) and M1 (0.52) radiations.[40] The high K/L ratio (9 ± 1) of this gamma ray supports an M1 assignment.[37]

If the 48.4 kev gamma ray is emitted in cascade with the other two and is assumed to be E2 radiation, the total intensity of its conversion electrons should equal the total intensity of those of the 260.7 kev gamma ray. As can be seen from Table 5, these intensities are equal within experimental error. This does not exclude the possibility of an approximately 50 percent electron capture branching from the 1.75 hour isomeric state, with the 48 kev transition taking place in Hg^{198}. The L_{II}-L_{III} binding energy differences are too similar in mercury and thallium to allow an assignment on that basis. However, if electron capture were occurring, one would expect gamma rays from levels in Hg^{198} in high intensity with a 1.75 hour half-life. A scintillation spectrometer experiment gave no indication of such gamma rays. Thus, it may be concluded that the three gamma rays are very probably emitted in cascade as suggested by the Illinois group.[34]

2. Spin assignments -- Decay scheme of Tl^{198m}. --If one assumes that the assignments of multipolarity are correct and that the three gamma rays are in cascade, a tentative decay scheme can be constructed (see Fig. 11) in which the spin difference between the 1.75 hour Tl^{198m} and the 5.3 hour Tl^{198} can be as high as 7 with a parity change. The fact that no crossover radiation was observed supports this large spin difference. The positions of the 282.4 kev and the 48.4 kev transitions may be as shown in Fig. 11 -- or reversed. The spin alternative to the left in Fig. 11 is based on the assumption of a 2- ground state for Tl^{198}.[34] The alternative to the right is based on the assumption of a coupling between the spins of the 81st proton and the 117th neutron. The 260.7 kev M4 transition would then represent a transition of the 81st proton from an $h_{11/2}$ to a $d_{3/2}$ configuration, the 117th neutron remaining in an $f_{5/2}$ configuration; the 282.4 kev M1 + E2 transition may perhaps represent a transition of the same proton between the $d_{3/2}$ and $s_{1/2}$ configurations, the neutron again remaining in the $f_{5/2}$ configuration; the 48.4 kev E2 transition then represents a transition of the odd neutron from the $f_{5/2}$ to a $p_{1/2}$ configuration, the proton remaining in an $s_{1/2}$ configuration. This interpretation, of course, is extremely speculative.

Also shown in Fig. 11 is the electron capture decay from Pb^{198} which was observed by Neumann and Perlman.[42] If the 25 minute Pb^{198} parent of Tl^{198m} is the 0+ ground state of Pb^{198}, then Tl^{198m} must have a low spin and Tl^{198} must have a high spin. The assignment of a high spin to Tl^{198} is in sharp disagreement with the interpretation of its electron capture decay given by Bergström and co-workers.[34]

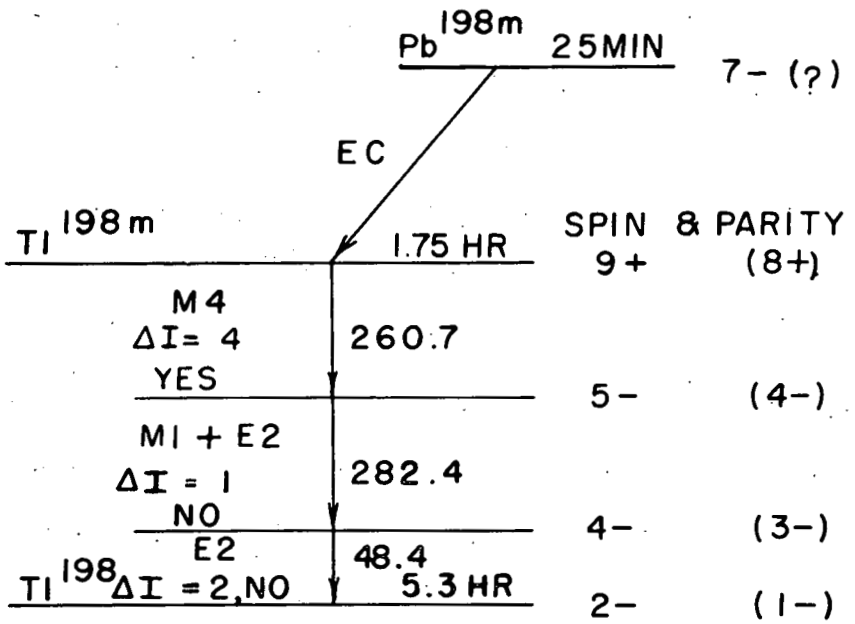


Fig. 11 — Decay scheme of Tl^{198m}.

These workers' data indicate that the electron capture decay of Tl^{198} goes predominantly to the 2+ first excited state of Hg^{198}. Thus of the two isomeric states of Tl^{198} the 1.75 hour Tl^{198m} most probably has the higher spin. The fact that other even-even lead isotopes have high spin isomers suggests that Pb^{198} might also have one. For example, the isomeric state of Pb^{204} has been suggested to be 6+[43] or 7-.[44] Very recently Maeder and Wapstra[45] have found indication of a 9- isomeric state in Pb^{202}. Alburger[46] assigned the isomeric level in Pb^{206} as 7-.

If Pb^{198m} has a similar high spin, direct electron capture decay from a high spin Pb^{198m} to a high spin Tl^{198m} would be possible. It may, therefore, be concluded that there is indeed another case of isomerism in Pb^{198}.

3. Auger electron spectrum. -- The K Auger electrons emitted in the K electron capture of the 5.3 hour Tl^{198} daughter of Tl^{198m} were investigated. Although some K x-rays of thallium were present from the K conversion of the 260.7 kev and the 282.4 kev gamma rays, greater than 70 percent of those present were those of mercury. Because of the presence of the K Auger lines of two adjacent elements the assignment of resolved lines becomes somewhat ambiguous. However, reliable relative intensities of the KL_pL_q, KL_pY_q and KX_pY_q (where X and Y refer to the M, N, etc. atomic orbitals) groups of lines can be obtained from the data. These relative intensities would correspond to those for an atomic number between 80 and 81.

The K Auger spectrum uncorrected for decay is shown in Fig. 12. The decay corrections were not large since all the lines were observed within a period of 1.5 hours. Using a half-life of four hours for decay

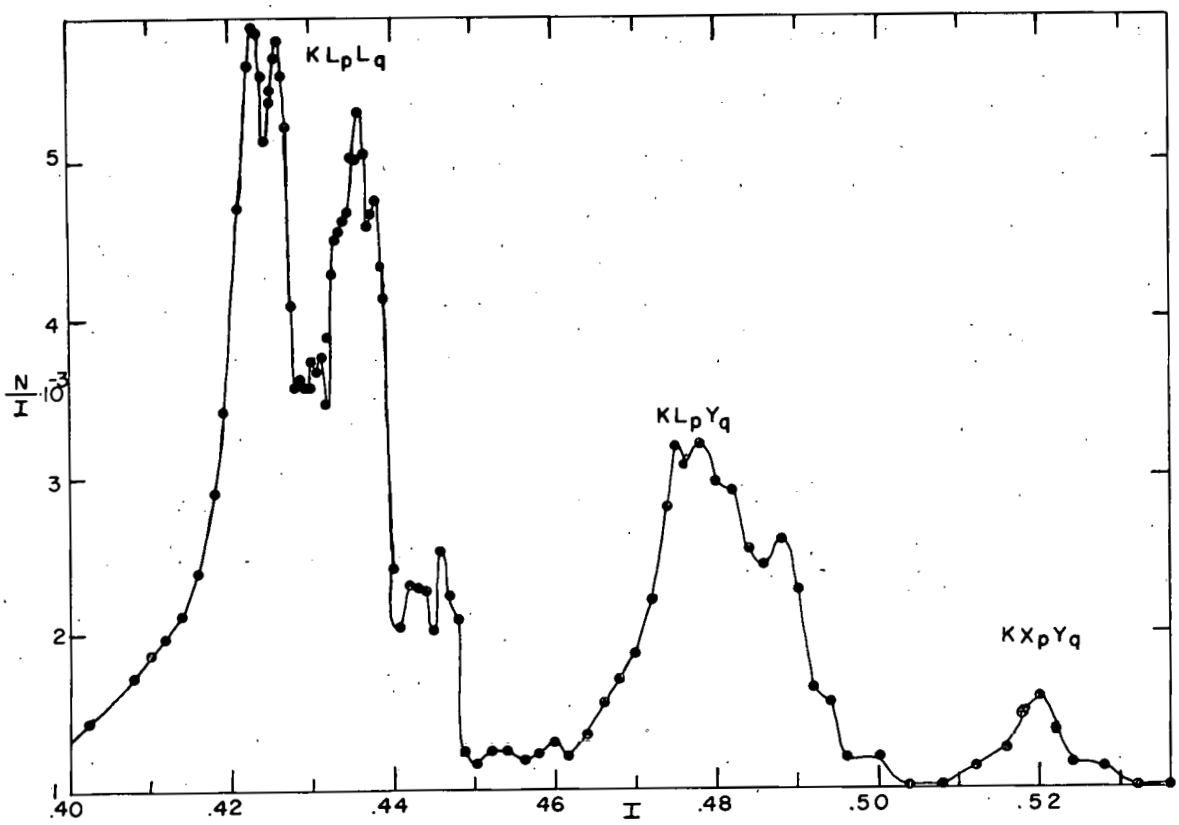


Fig. 12—K Auger electrons of Tl^{198}-Tl^{198m}.

corrections, the ratio KL_pL_q:KL_pY_q:KX_pY_q becomes 1.0:0.53±0.05: 0.063 ± 0.02. The half-life used for decay correction was taken as the observed decay period of one of the initial points on the spectrum.

Broyles[47] reports the value 1.0:0.7±0.15 for the KL_pL_q:KL_pY_q of mercury. The limits of error of Broyles' and the present work mutually encompass the ratio 1.0:0.56. Thus the present work is in fair agreement with Broyles' results. However, it is not in good agreement with the theoretical calculations of Pincherle.[48] Pincherle's value for the KL_pL_q:KL_pY_q:KX_pY_q ratio is 1.00:0.716:0.103 using non-relativistic unscreened, hydrogen-like wave functions. These ratios are supposed to be good for all atomic numbers. However, since relativistic effects are much greater at larger atomic numbers one would not expect close agreement with Pincherle's values. Massey and Burhop[49] have found that consideration of relativity increases the theoretical KLL Auger yield. If similar relativistic effects have less influence on the KL_pY_q and KX_pY_q Auger yields, the discrepancy between the results of the present work and the calculations of Pincherle[48] would be resolved.

F. Protactinium 228

1. L lines assigned to 57.8 and 130 kev gamma rays. -- Protactinium 228 was first produced by Ghiorso et al.[50] in bombardments of thorium with 80 Mev deuterons. These workers found the EC/α branching ratio to be about 50. No previous work has been reported on the electron spectrum of Pa^{228} since the investigators mentioned above studied only the alpha particle radiations of these isotopes.

A sample of Pa^{228} also containing some Pa^{232}, Pa^{230}, and Pa^{229} was obtained from a bombardment of thorium with 115 Mev protons. Because of the presence of a visible amount of solid material in the final protactinium fraction, it was decided to use a 0.001-inch thick platinum counting disk for the sample backing.

Because of the presence of Pa^{229}, Pa^{230}, and Pa^{232} no unambiguous assignments can be made for four prominent lines appearing at 22, 26, 30, and 33 kev. However, relatively certain assignments can be made for four electron lines at 38.2, 41.5, 110, and 114 kev, respectively. As seen in Table 6 these lines have been assigned to gamma rays in Th^{228} which have been previously observed in Ac^{228} beta minus decay.[51] The gamma rays are also observed in U^{232} alpha decay. The L_{II}/L_{III} ratio for each gamma ray is consistent with an E2 assignment.[6]

Table 6

Electron Lines from Pa^{228} Decay

Gamma energy (kev)	Electron energy (kev)	Intensity (Arbitrary Units)	e^-/β^- disintegration of Ac^{228} from Ref. 51
57.8 ± 1	38.2	28 ± 5	0.238
	41.5	20 ± 5	0.202
130 ± 2	110	3 ± 0.5	0.033
	114	2 ± 0.5	0.023

2. Decay scheme of Pa^{228}.--It is of interest to note that the electron capture decay of Pa^{228} populates directly or indirectly the first two excited states of Th^{228} in about the same ratio of intensities as does the beta decay of Ac^{228}.[51] The closed cycles calculation[52]

of the decay energy for Pa^{228} is 2.06 Mev. Since this value is not far different from the decay energy exhibited by Ac^{228}, it is not improbable that the electron capture of Pa^{228} populates the excited states of Th^{228} in much the same manner as Ac^{228}. Thus the similarities noted above for the first two excited states are not surprising.

G. Protactinium 230

1. Electron spectrum. -- Protactinium 230 was first identified by Studier and Hyde[53] in bombardments of Th^{232} with deuterons. The EC/β^- branching ratio is about 92:8[54] and alpha branching occurs to the extent of 0.003 percent.[55] Measurements of the electron and gamma radiations were made by Osborne et al.[56] by absorption techniques. They report a gamma ray of 0.94 Mev and K and L x-rays with the relative abundances 14:9:5. Osborne et al.[56] also observed beta particles with energy maxima at 0.22 Mev and 0.43 Mev with relative intensities of around four to one.

The present work was undertaken to study in greater detail the electron and gamma radiations emitted in the decay of Pa^{230}. The activity was produced by the reaction $Th^{232}(p, 3n)Pa^{230}$ using around 100 Mev protons from the laboratory's 184-inch synchro-cyclotron. The Pa^{232}, Pa^{228}, and Pa^{229} also produced in the bombardment were allowed to decay for about one month before chemical separations were performed. The Pa^{230} was separated from the thorium target by di-isopropyl-ketone extraction techniques.[57, 58] It was apparent some time after the beta spectrometer samples were made that for some reason a large amount of Zr^{95} and Nb^{95} activity had followed protactinium in the

chemical separation. The sample used also contained an unknown amount of the U^{230} decay chain.

The sample was mounted on a double layer of gold leaf by evaporation of an HF solution containing the activity. The material was visible on the gold leaf so the reason for the relatively poor resolution obtained is probably sample thickness and not back-scattering. The spectra were obtained using the semi-automatic system described previously.

A beta continuum underlying the electron lines corresponds well with that which would be expected from the beta particles of Zr^{95} and Nb^{95}. The presence of these nuclides in high abundance was shown by gamma scintillation spectrometer studies which will be discussed below. The lines for which the interpretation is fairly certain are listed in Table 7. Since electron capture is about ten times as frequent as beta minus decay in this nuclide, all of the lines have been assigned to gamma transitions in Th^{230}.

The electron lines assigned (see Table 7) to 52.2_M and 52.2_N also fit energywise with the expected L_{II} and L_{III} lines of a 69 kev gamma ray. A line appears in the spectrum which might be assigned to the M + N line of this gamma ray. Thus the high abundance of the lines assigned as 52.2_M and 52.2_N relative to the $52.2_{L_{II}}$ and $52.2_{L_{III}}$ lines is perhaps due to the presence in the sample of considerable U^{230}. However, this explanation is not consistent with the relatively high intensity of the line postulated as 69_{M+N}, nor with the absence of lines from the 110 kev gamma ray of Th^{226}.[9] It is possible that the approximately 69 kev gamma ray follows the beta decay of Pa^{230} although the first excited state of U^{230} is expected to be at a lower energy.[21]

Table 7

Electron Lines of Pa^{230}

Gamma energy (kev)	Electron energy (kev)	Conversion shell	Intensity (Arbitrary Units)
52.2 ± 1	33.3	ThL_{II}	1.5 ± 0.7
	35.9	ThL_{III}	1.0 ± 0.5
	48.5	ThM	1.4 ± 0.5*
	52.9	ThN	1.0 ± 0.5*
90 → 94	76.7	ThL	0.2 ± 0.1
293 ± 10	183	ThK	1.0 ± 0.5
	276	ThL	0.1 ± 0.05
305 ± 10	194	ThK	5.7 ± 0.5
	284	ThL	1.0 ± 0.2
	302	Th_{M+N}	0.2 ± 0.2

*See text.

The most reasonable explanation of this discrepancy is that self-absorption within the sample is attenuating the L_{II} and L_{III} lines with respect to the M and N lines.

The K and L lines of gamma rays of 293 ± 10 kev and 305 ± 10 kev decay with the half-life of Pa^{230} within the limits of error of the measurement. The K/L ratios of each are large (about 10 and about 6, respectively) which is indicative of M1 or E1 radiation. The high intensity of the electron peaks of these radiations with respect to those of the 52 kev gamma ray make the M1 assignment seem more likely.

The beta spectrum of Pa^{230} was not observed with any reasonable certainty because of the relatively high intensity of the Zr^{95} and Nb^{95} beta spectra. The maximum energy of the Pa^{230} beta spectrum is expected to be 430 kev from closed cycle calculations.[52]

2. Scintillation spectrometry of Pa^{230} gamma rays. -- The gamma rays emitted by Pa^{230} were observed in the 50-channel scintillation spectrometer of this laboratory. The spectra obtained are shown in Figs. 13a and 13b. In agreement with the absorption experiments of Osborne et al.,[56] a gamma ray of 940 ± 20 kev thorium K x-rays, and thorium L x-rays were observed. In addition to these radiations, however, gamma rays with energies 305 ± 10 kev, 1000 ± 40 kev, 1060 ± 30 kev, 1180 ± 40 kev (?), 460 ± 15 kev, and 540 ± 20 kev were observed. Of course, one of the more prominent peaks in the spectrum was that from the 721 kev gamma ray of Zr^{95} and the 745 kev gamma ray of Nb^{95}. The assignment of the 750 kev peak to these activities was proved by its asymmetry and by its increasing intensity relative to other peaks in the spectrum with time. A summary of the data on the gamma rays is given in Table 8. The corrected intensities were calculated using the

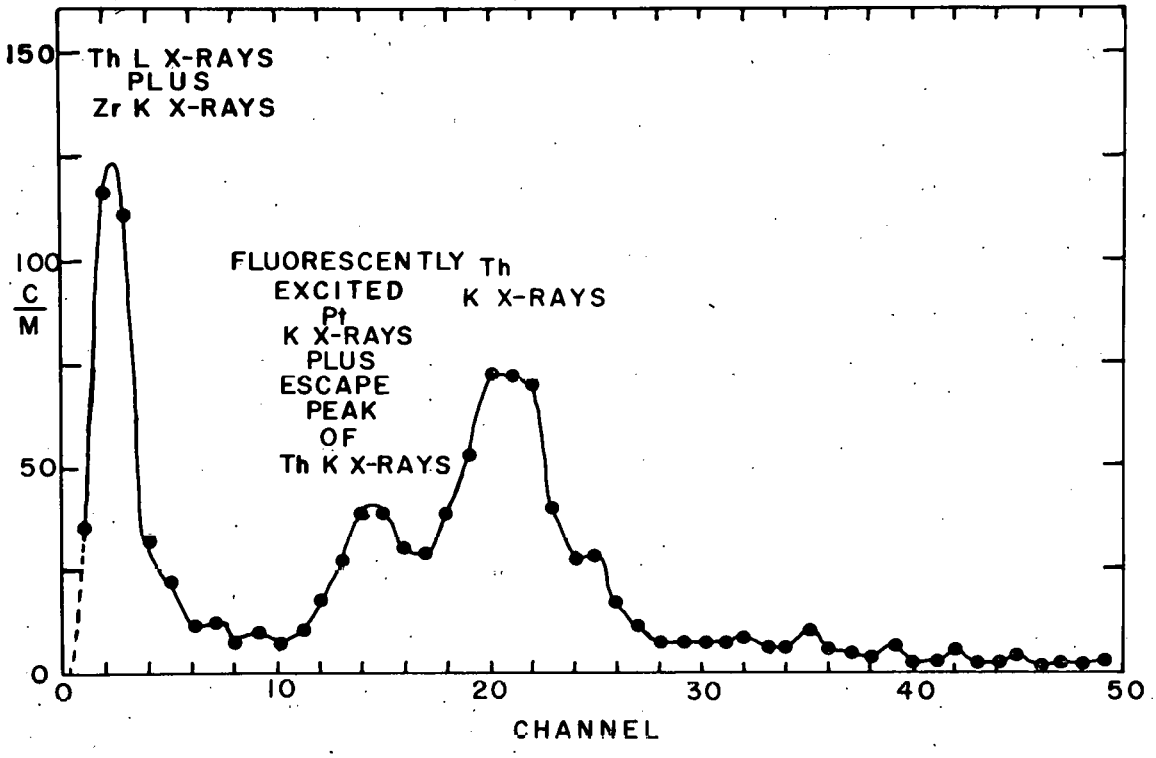


Fig. 13a—Gamma spectrum of Pa^{230}.

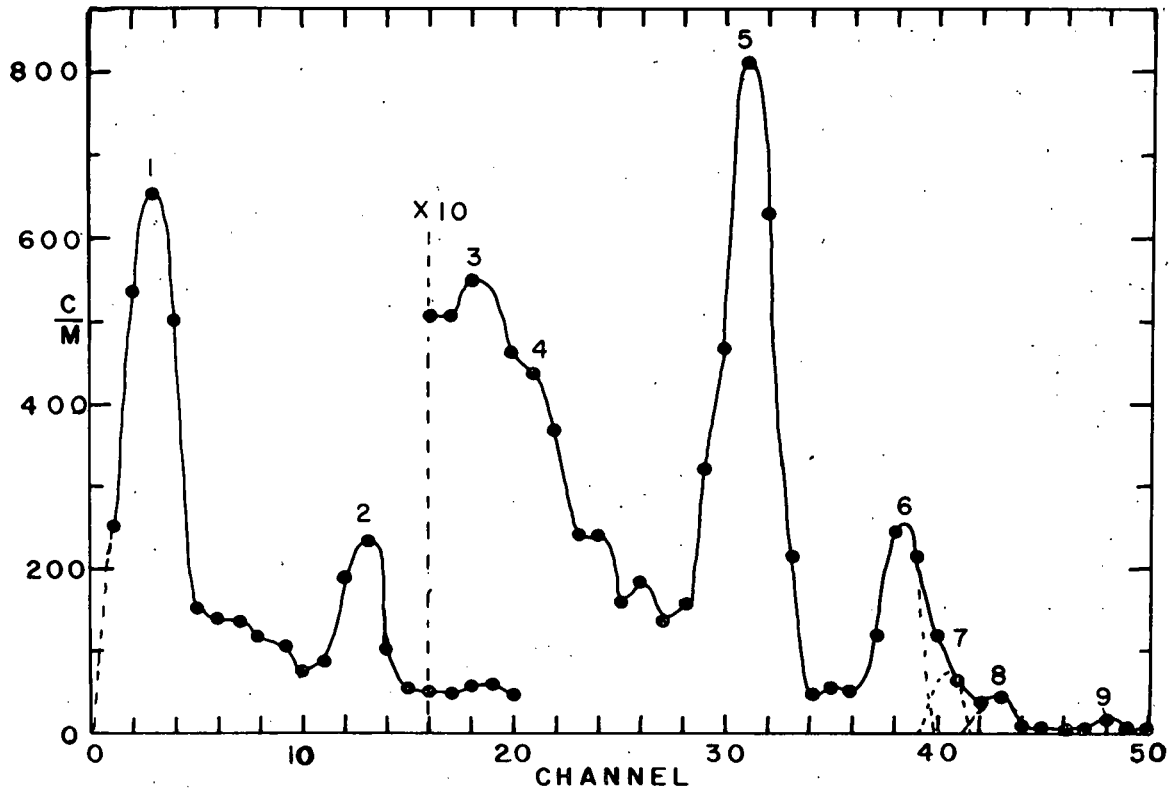


Fig. 13b — Gamma spectrum of Pa^{230}.

curve of counting efficiency for sodium iodide crystals given by McLaughlin and O'Kelley.[59]

Table 8

Protactinium 230 Gamma Rays

Gamma energy (kev)	Uncorrected relative intensity (Arbitrary Units)	Corrected* intensity
14 ± 4 (L x-rays)	1800	1800
94 ± 5 (K x-rays)	2000	2000
305 ± 10	460	820
460 ± 15	100	360
540 ± 20	Low	Low
940 ± 20	50	830
1000 ± 40	7	130
1060 ± 30	7	140
1180 ± 40 (?)	1	20

*See text.

3. Decay scheme of Pa^{230}--levels in Th^{230}.-- From the present work no unambiguous decay scheme can be drawn. However, reasoning from relative intensities of gamma rays and electron lines separately, the following inferences can be made: 1) at least 40 ± 10 percent of the electron capture decay populates one or a pair of levels about 940 kev above the ground state; 2) at least 40 percent of the electron capture disintegrations give rise to a 310 ± 10 kev gamma ray. This gamma ray is very likely of M1 or E1 character from the K/L conversion ratio; 3) a significant fraction of the electron capture disintegrations directly or indirectly populate the first excited state of Th^{230}; 4) no appreciable

fraction of the electron capture decay gives rise to gamma radiation of the energy difference between the second and first excited states of Th^{230} (120 kev).[9]

H. Neptunium 236

1. L lines assigned to 43.4 kev and 44.2 kev gamma rays. -- Neptunium 236 was first produced and mass assigned by James, et al.[60] from bombardments of uranium with deuterons. In the present study, uranium foil containing >99 percent U^{235} was bombarded with 12.5 Mev deuterons in the 60-inch cyclotron of this laboratory. The chemical separation and purification included an oxidation-reduction cycle and a final anion exchange column separation. The sample for the present investigation was prepared by evaporation of a drop of distilled water containing the activity from a palladium leaf backing.

The only previous study of the radiations of 22-hour Np^{236} is that of Orth and O'Kelley.[61] These workers report an EC/β^- branching ratio of around 2, a two component beta spectrum with maximum energies of 360 and 510 kev, respectively, and a 150 kev gamma ray. An electron spectrum was observed in the present work which was similar to that observed in the previous work.[61] However, certain lines in the spectrum showed greater complexity than before. The appearance of this complexity, coupled with the results of recent scintillation spectrometer studies[62] requires a reinterpretation of the electron data.

The lines designated as L Augers by O'Kelley were resolved into four lines which correspond well energywise to the L_{II} and L_{III} lines of gamma rays with energies 43.5 ± 1 kev and 44.2 ± 1 kev, respectively. These energies depend upon the arbitrary assumption that the slightly

more intense pair of L lines arise from conversion in plutonium following beta decay, and that the less intense pair arise from conversion in uranium following electron capture. The electron data are summarized in Table 9.

Table 9

Electron Lines of Np^{236}

Gamma energy (kev)	Electron energy (kev)	Conversion shell	Intensity (Arbitrary Units)
43.5 ± 1	21.1	PuL_{II}	
	25.4	PuL_{III}	
44.2 ± 1	23.2	UL_{II}	48
	27.0	UL_{III}	
44 ± 2	37.9	$Pu\Sigma M$ and $U\Sigma M$	18
43 ± 1	41.7	$Pu\Sigma N$ and $U\Sigma N$	3
K x-rays	88 ± 5	$U\Sigma L$ and $U\Sigma M$	10
———	500 ± 30 (β^- maximum)	———	200

The line designated 150_K by Orth and O'Kelley[61] also may be interpreted as the unresolved ΣM conversion lines of the 43.5 and 44.2 kev gamma rays. A line also appeared which may be assigned to the unresolved N line of the two gamma rays. K Auger lines were also observed.

2. Decay scheme of Np^{236}. --The beta spectrum was observed both on the double focusing spectrometer where the intensity was too low for a Fermi analysis and on a magnetic lens spectrometer. A Fermi analysis by O'Kelley of the data from the magnetic lens spectrometer gave a maximum beta energy of 518 ± 10 kev.[63] The Fermi-Kurie plot exhibited a marked concavity toward the abscissa which is indicative of a forbidden beta transition. However, it is not impossible that the forbidden shape was caused by some variation in counting efficiency of the detector used.

Scintillation spectrometer experiments performed by Jaffe et al.[62] indicate that approximately 1.66 times as many K x-rays as L x-rays are emitted in Np^{236} decay. No significant amount of gamma radiation was observed. A 4π count of the beta particles from the same sample used in the scintillation spectrometer experiments gave an EC/β^- ratio of (43 ± 3):(57 ± 3), assuming only K electrons are captured. From the relative K and L x-ray intensities, the number of K and L vacancies produced per 100 disintegrations can be calculated by using the following assumptions: 1) that 72 percent of the K vacancies are filled by L electrons[64]; 2) that the K Auger yield is 0.03 at the atomic number 92; 3) that the L Auger yield in this region of atomic number is 0.5;[22] 4) that 43 percent of the disintegrations take place by capture of K electrons; 5) that the intensity of the K x-rays is 1.66 times that of the L x-rays. From this calculation one can conclude that for each 100 disintegrations, 21 L vacancies arise from either L electron capture or gamma ray conversion.

From the very approximate values of electron intensities one can conclude that the abundance of conversion electrons is about 35 percent

that of the beta spectrum. Since 57 percent of the disintegrations take place by beta particle emission, one is led to a value of 20 conversion electrons per 100 disintegrations. Thus within the limits of error of these intensity measurements, all of the L vacancies arising by means other than by the filling of K vacancies arise from L conversion of the 43.4 and 44.2 kev gamma rays. However, the limits of error are such that as many as 5 to 10 L vacancies per 100 disintegrations may be arising from L capture. It appears safe to conclude that the lower limit for the K/L capture ratio is around 4.

The intensities of the L lines of the 43.4 and the 44.2 kev gamma rays are roughly comparable. Thus one can set an upper limit of about 10 events per 100 disintegrations which populate each of the first excited states of Pu^{236} and U^{236}. The intensities given in Table 9 are subject to considerable uncertainty, but the values should be reliable to ±25 percent.

The interpretations of the data from the present study are in sharp disagreement with those of Orth and O'Kelley.[61] One reason for the large discrepancy between the two sets of data may be the different relative amount of Np^{234} present in each case. Hoff[19] reports an upper limit of 1.0 for the K/L capture ratio for Np^{234}. Since considerable amounts of Np^{234} were present in the samples of Np^{236} investigated by Orth and O'Kelley, one would expect their K/L capture ratio to be somewhat in error. Also, the absorption methods which they used are less reliable for intensity measurements than the scintillation spectrometer used in the present study. The discrepancy in the interpretation of the electron line spectrum resulted from

spectrometer calibration uncertainties in the low energy region at the time the previous experiments were performed.

A decay scheme which is consistent with the available data is shown in Fig. 14.

IV. DISCUSSION OF RESULTS

A. Theoretical Calculations

The use of the term "conversion coefficient" is misleading. Theoretical interpretation of the processes of radiative (emission of gamma quanta) and non-radiative (emission of orbital electrons) transitions indicate that they may be considered nearly independent of each other.[65] That is, if radiative transitions in nuclei are assumed to arise from a radiator (electric dipole, electric quadrupole, magnetic dipole, etc.) located at the center of the nucleus, then it is found that the radiative emission rate is almost independent of the non-radiative emission rate for a given transition. Thus a nuclear transition rate is the sum of the rates of gamma and electron emission, the two processes occurring side by side with nearly independent rates and there is little if any so-called "conversion" of gamma radiation into electron "radiation".

The reasoning leading to the above conclusions is briefly as follows:[65] If the nucleus were stripped of its orbital electrons, the total nuclear transition rate would equal the rate of emission of gamma quanta. However, in the presence of the orbital electrons, the nuclear transition rate is augmented by the possibility of another mode of nuclear de-excitation, namely orbital electron emission. The emission of orbital electrons arises from the interaction of the radiation field of the nuclear radiator with the orbital electron cloud. This interaction

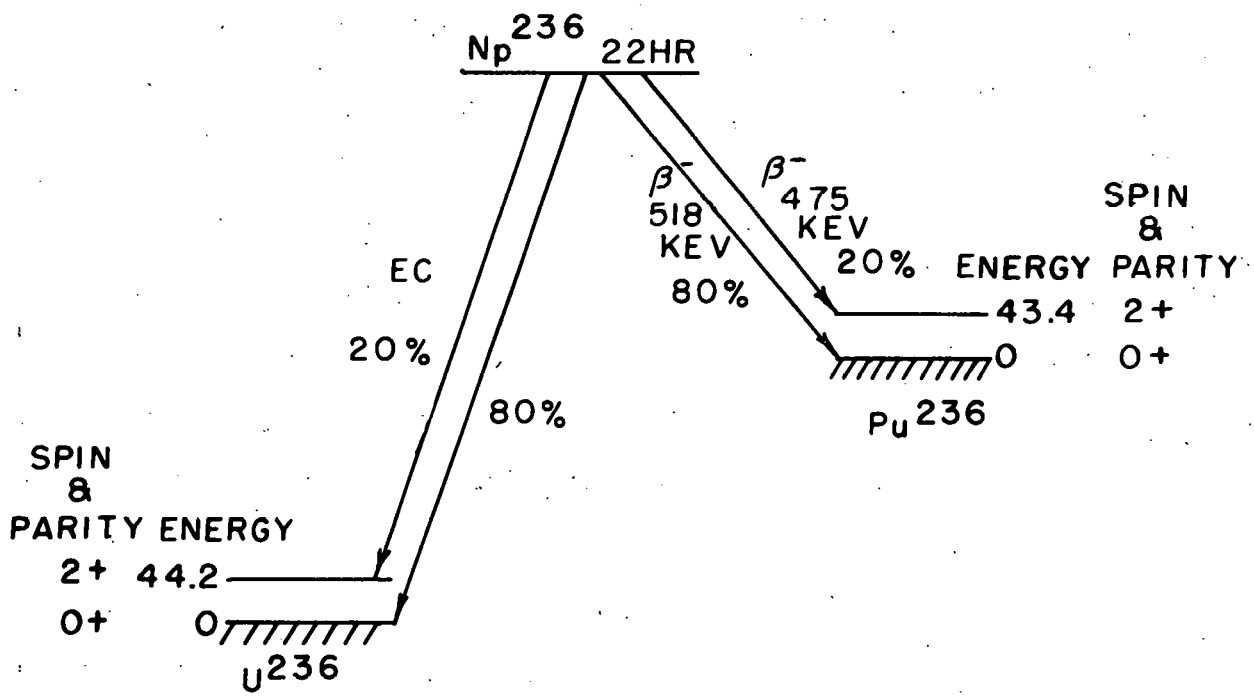


Fig. 14—Decay scheme of Np^{236}.

thus provides a mechanism in addition to gamma emission for the occurrence of the nuclear transition.

The so-called "conversion coefficient", then, has been most commonly defined as the orbital electron emission rate divided by the gamma emission rate. Some writers, however, still use the term "conversion coefficient" to indicate the fraction of the total nuclear transitions which take place by orbital electron emission. The former, commonly used definition will be used in this discussion.

Theoretical calculations of L conversion coefficients for the heavier elements have been performed by Fisk[66] and Gellman et al.[67,6] Since the calculations made by Gellman et al.[67,6] are more extensive than any previously made for this region of atomic number, and were made using exact relativistic equations, the author has chosen to present comparisons of experimental values with these calculations only. Figures 15a through 16i are based on these calculations. No exact calculations exist at present for L shell conversion coefficients (for Z >80) where the effects of screening were taken into account. However, Reitz[68] has found the screening correction for the K shell conversion coefficient to be a positive one to thirteen percent of the "unscreened" values. Whether the same or a larger screening correction holds for any of the L shells remains to be seen. Rose et al.,[69] believe the screening correction will be appreciable for the L shells, especially at high atomic number.

B. Empirical Correlations

Mihelich[70] has reviewed the experimental data on L conversion ratios through early 1952. His multipolarity assignments based upon earlier, less extensive, and in some cases less exact calculations are

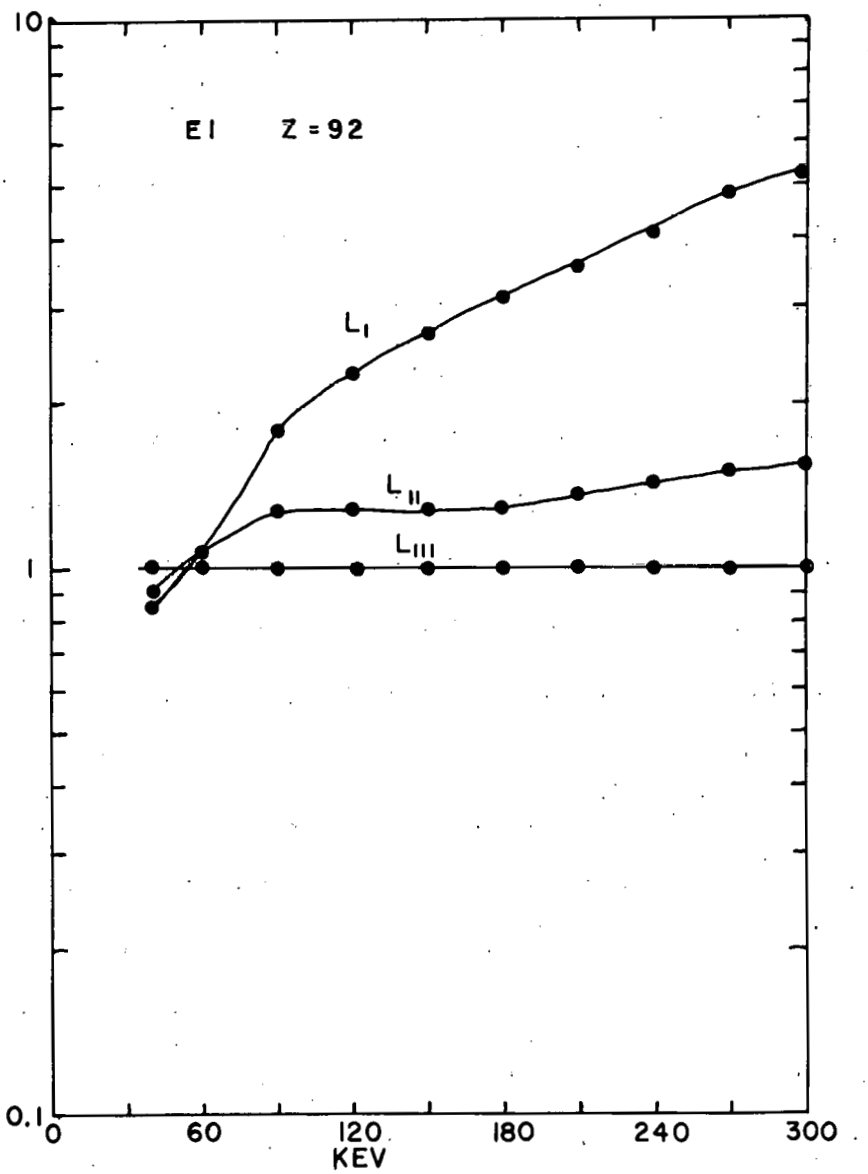


Fig. 15a — L_I:L_{II}:L_{III} ratios based on the calculations of Gellman et al.[6]

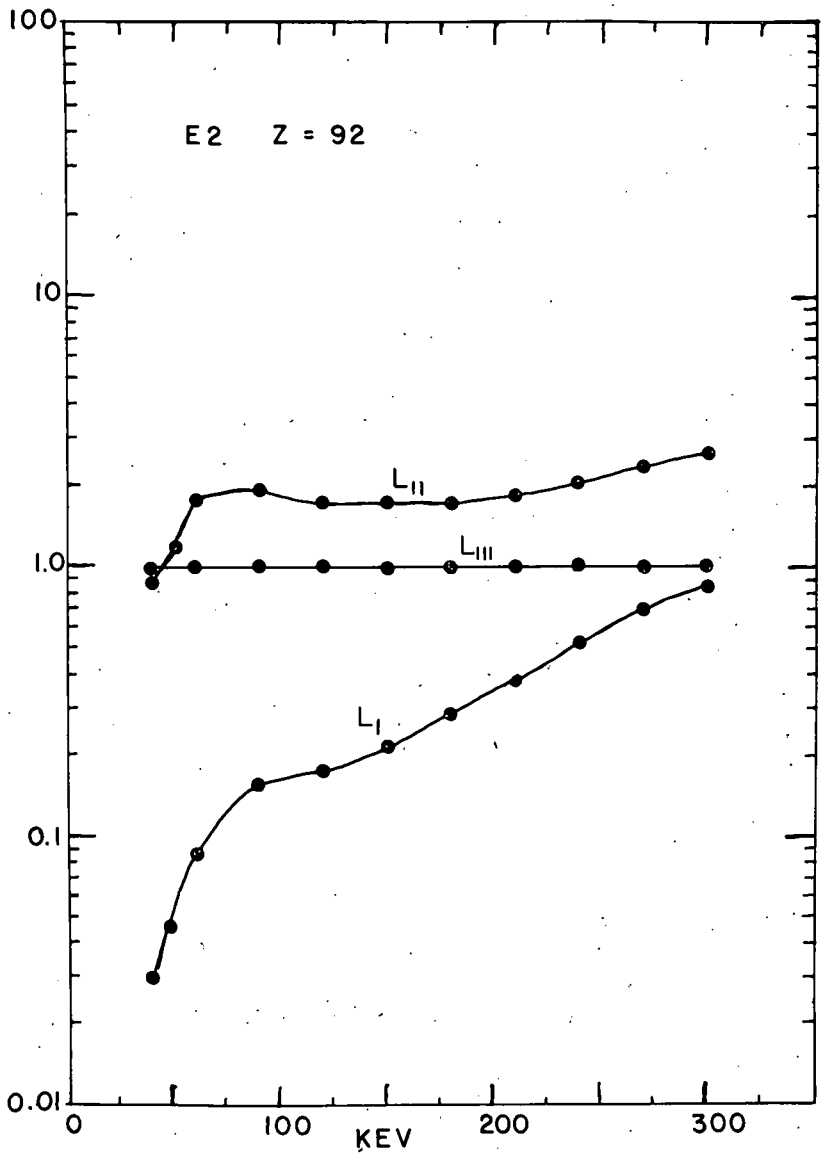


Fig. 15b — L_I:L_{II}:L_{III} ratios based on the calculations of Gellman et al.[6]

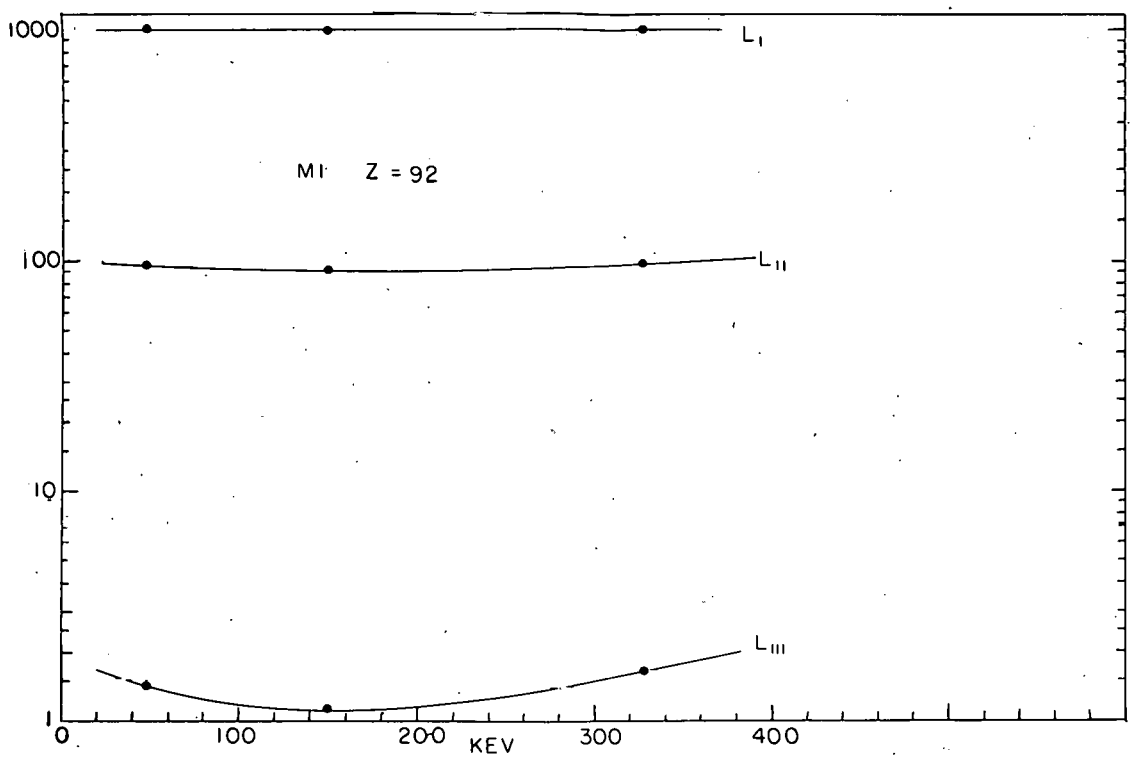


Fig. 15c — L_I:L_{II}:L_{II} ratios based on the calculations of Gellman et al.[6]

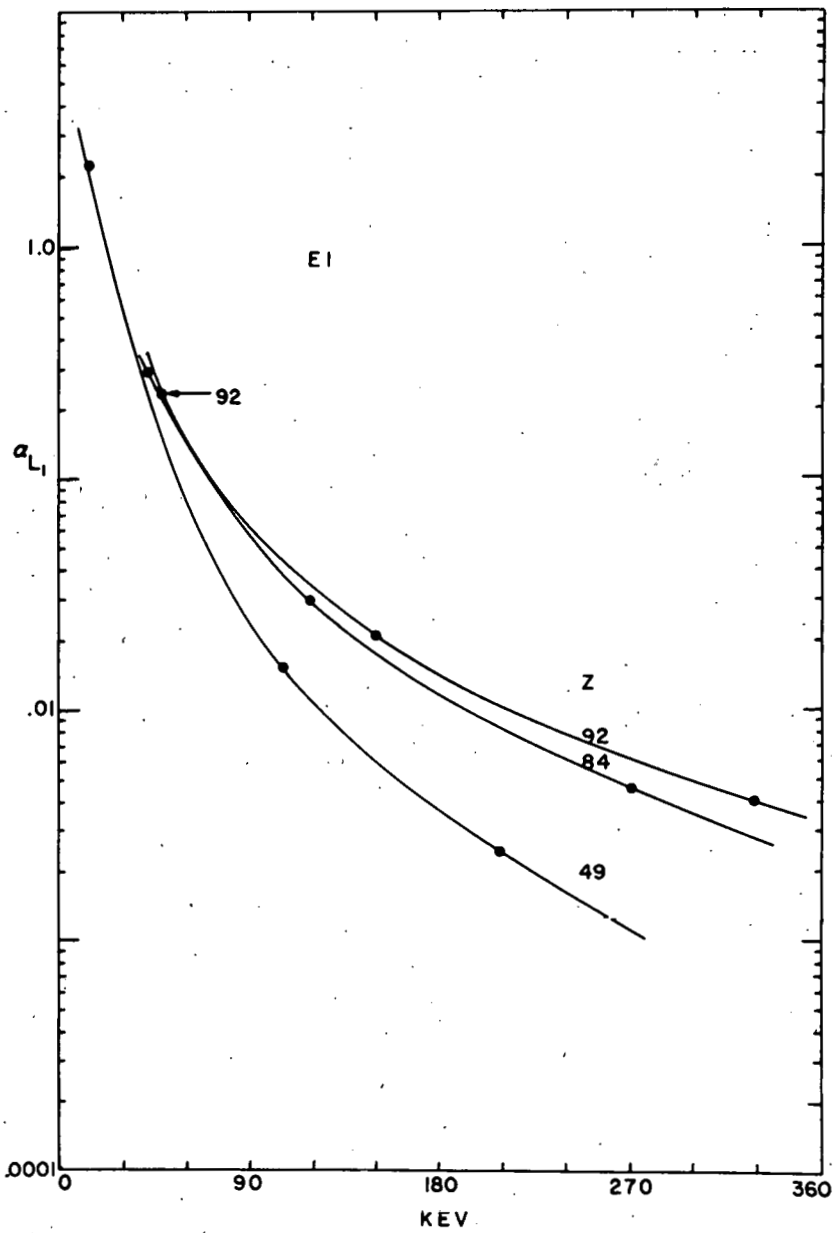


Fig. 16a — Curves for interpolating the tables of Gellman et al.[6].

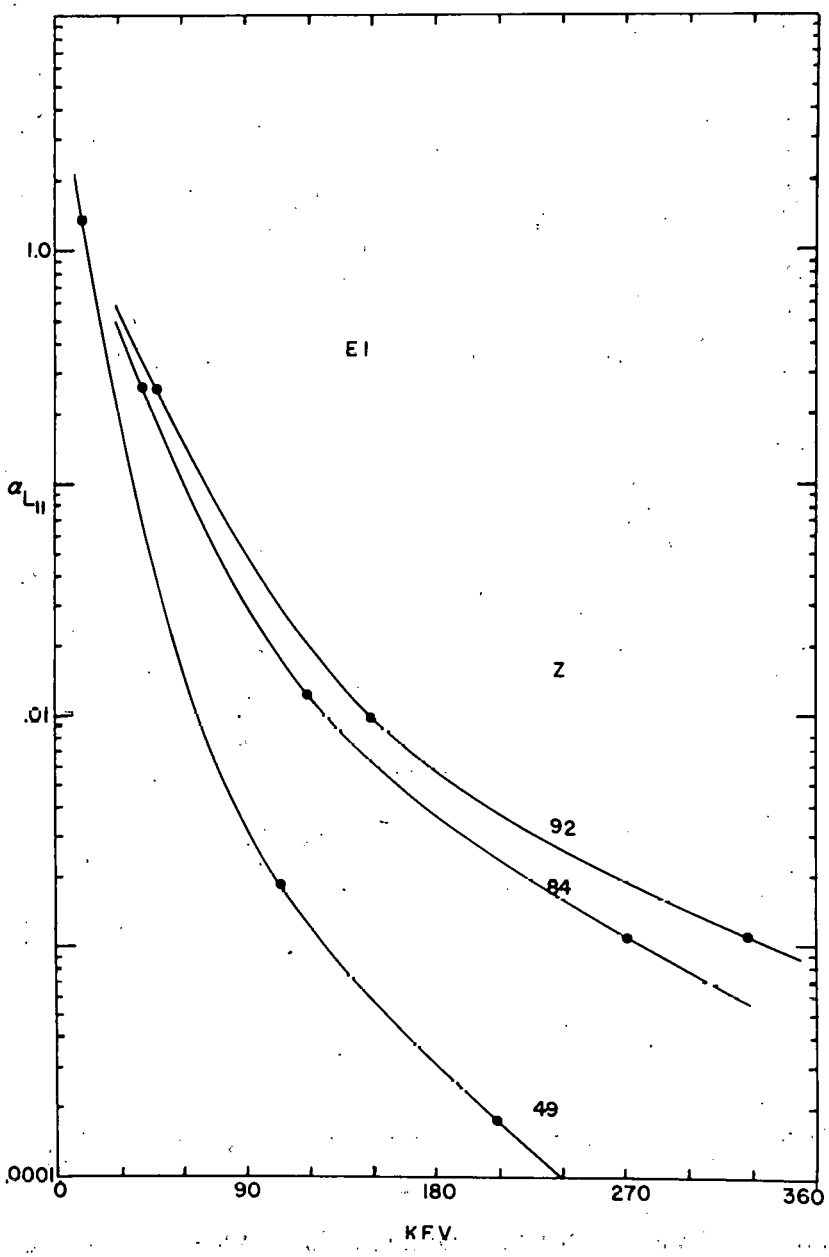


Fig. 16b—Curves for interpolating the tables of Gellman et al.[6].

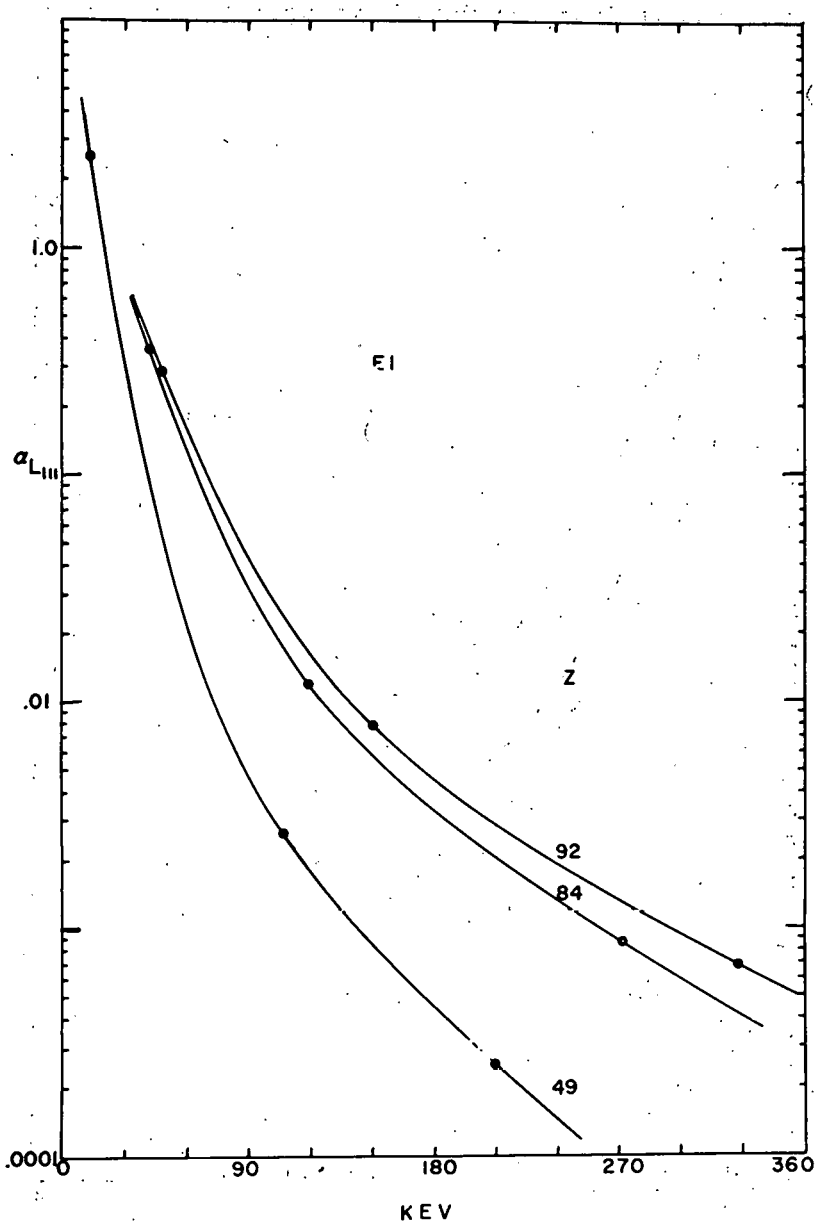


Fig. 16c.—Curves for interpolating the tables of Gellman et al.

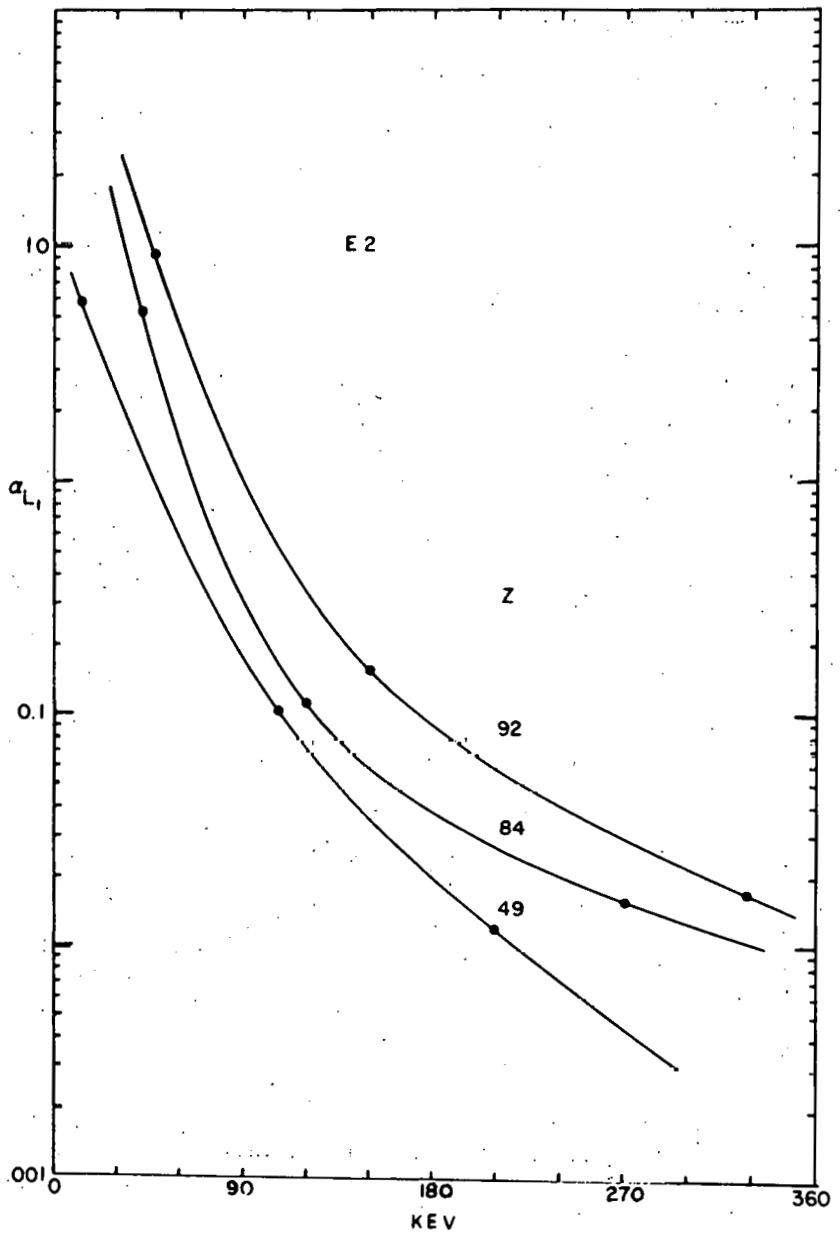


Fig. 16d—Curves for interpolating the tables of Gellman et al.[6].

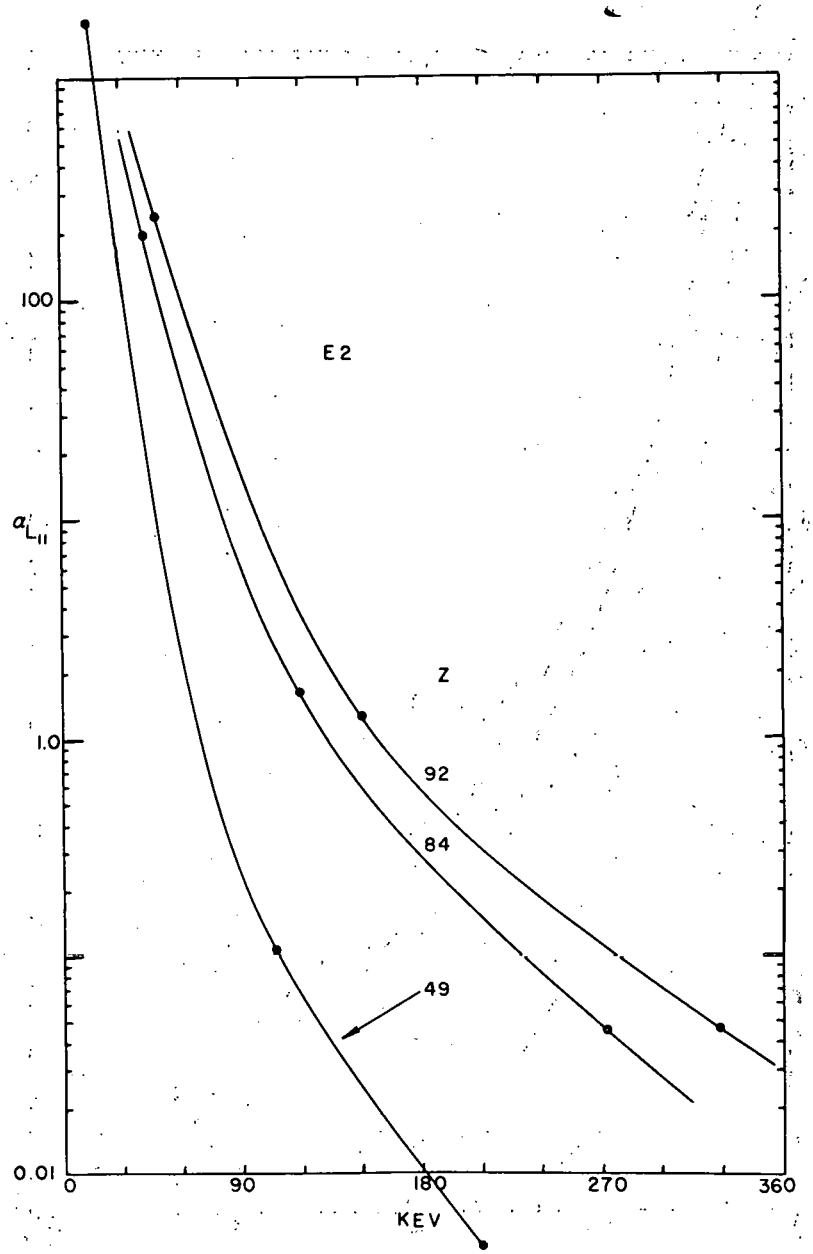


Fig. 16e — Curves for interpolating the tables of Gellman et al.[6].

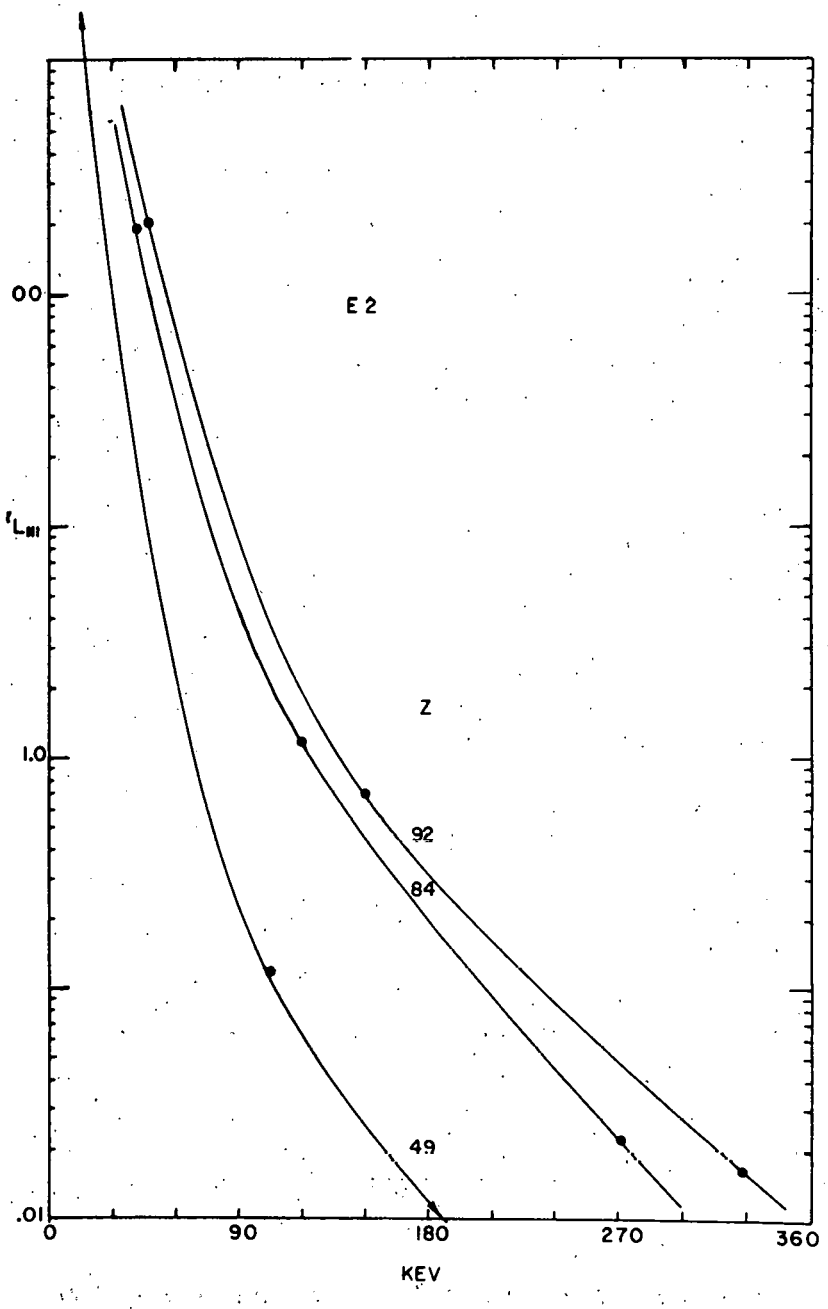


Fig. 16f—Curves for interpolating the tables of Gellman et al.[6].

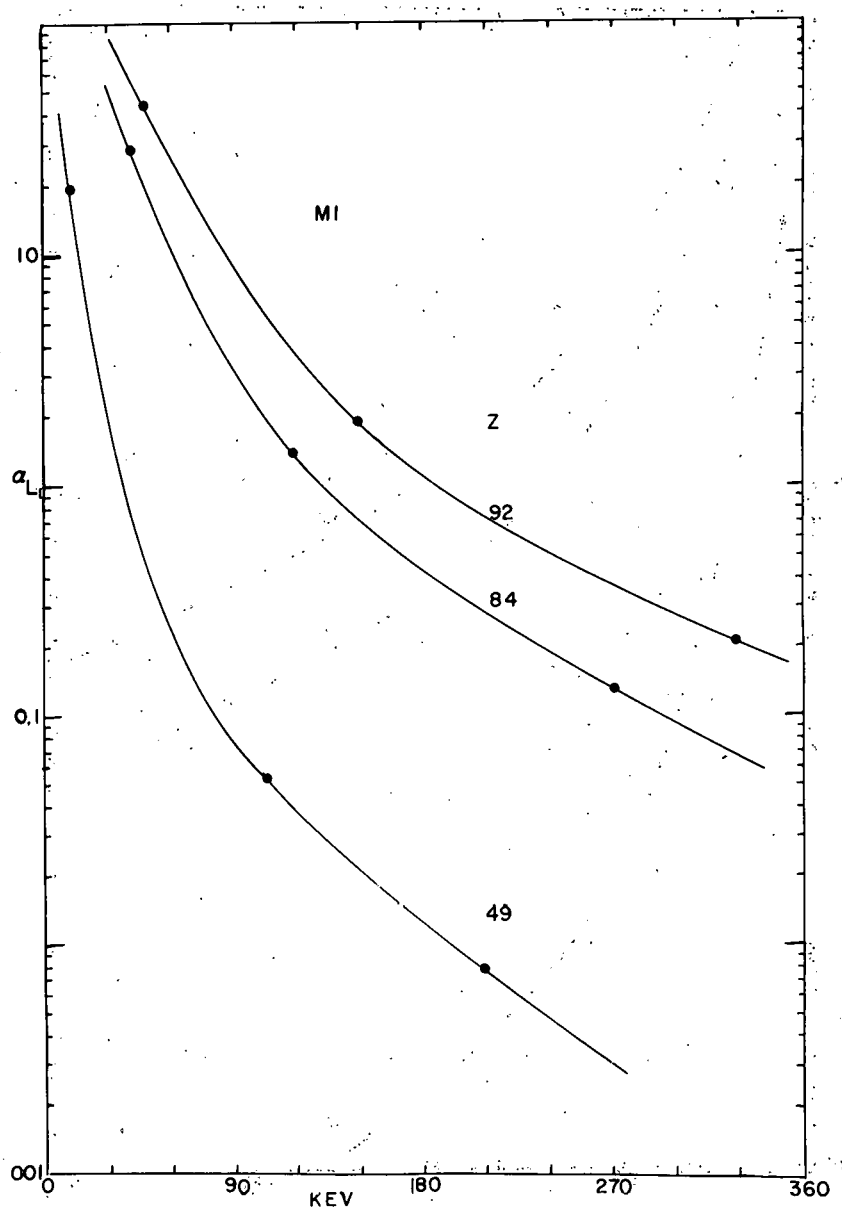


Fig. 16g — Curves for interpolating the tables of Gellman et al.[6].

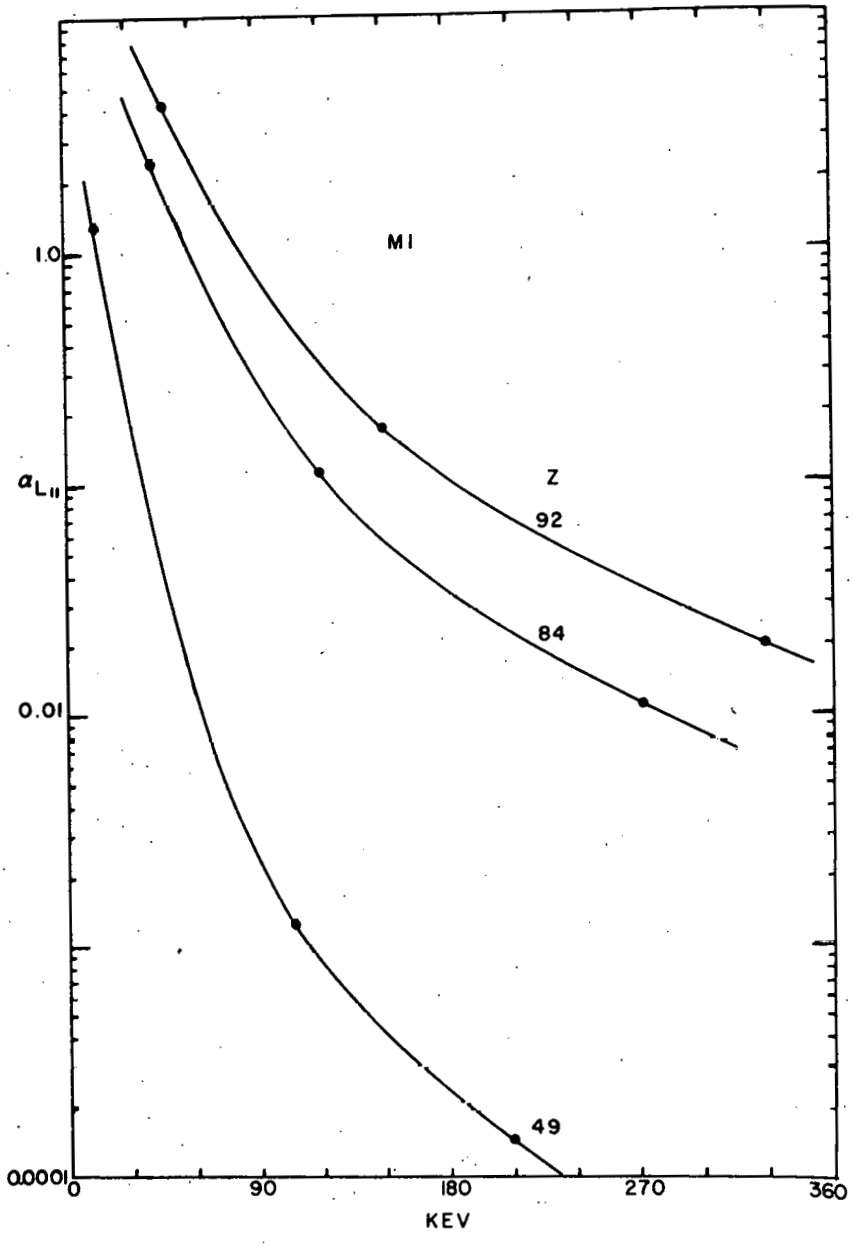


Fig. 16h—Curves for interpolating the tables of Gellman et al.[6].

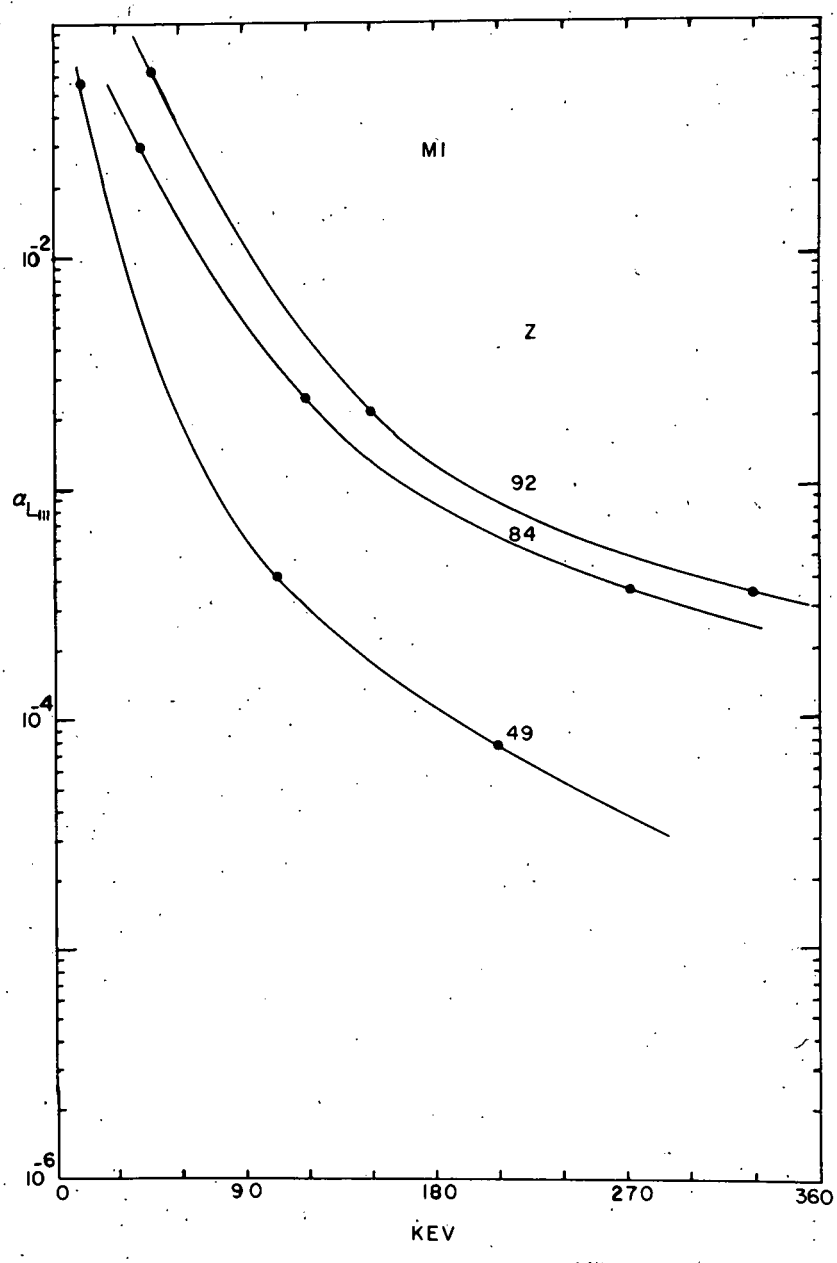


Fig. 16i — Curves for interpolating the tables of Gellman et al.[6].

in good general agreement with those which would be predicted by a comparison with the more recent tables of Gellman et al.[6]

Swan and Hill[71] have found experimental data on several electric quadrupole radiations to be in reasonably good agreement with the predictions of Gellman et al.[6]

Mihelich[39] has summarized experimental conversion ratios for M4 transitions in a recent paper. These data are the bases for Fig. 10 (see Tl^{198m}) in which the author has compared results for an M4 transition in this work with those reviewed by Mihelich.

C. Comparison of Experimental Data with Predictions.

1. Electric dipole transitions. -- The 59.6 and 26.4 kev gamma transitions which occur in the alpha decay of Am^{241} have been characterized with reasonable certainty as electric dipole radiations.[5,9] The 59.6 kev gamma ray has a total conversion coefficient of 0.92 ± 0.1 and a total L conversion coefficient of 0.72 ± 0.07. From the $L_I + L_{II}$:L_{III} ratio of 4.4, the L_{III} conversion coefficient is 0.17. This value is in excellent agreement with the value 0.15 from interpolation of the tables of Gellman et al.[6] The sum of the conversion coefficients for the L_I and L_{II} subshells is $0.72 - 0.17 = 0.55$. This value is in very poor agreement with the value 0.32 from interpolation of the tables of Gellman et al.[6] Some, but certainly not all of the difference between these values can be ascribed to errors inherent in interpolation. The only obvious explanation for this discrepancy is the possibility that the screening correction for either or both the L_I and L_{II} subshells is much larger percentagewise than those found by Reitz[68] for the K shell.

A priori the screening correction would be expected to be very similar percentagewise for the $L_{II}(p_{1/2})$ and $L_{III}(p_{3/2})$ shells. This seems borne out by the good agreement of L_{II}/L_{III} ratios of E2 transitions

with the "unscreened" calculations of Gellman et al.[6] Similarly, the percentagewise screening correction for the L_I ($s_{1/2}$) shell might be expected to be of different magnitude from those of the L_{II} and L_{III} shells. Thus L_I:L_{II}:L_{III} ratios for radiations (E1 and all magnetic radiations) which convert appreciably in the L_I shell[6] would not be expected to agree as well with the unscreened ratios as those converting only in the L_{II} and L_{III} shells (E2, E3, E4, etc.). That this is so is borne out by the data on the 59.6 kev E1 gamma ray under consideration here and the 46.5 kev M1 gamma ray following RaD decay.

The $(L_I + L_{II})/L_{III}$ ratio expected[6] for the 59.6 kev gamma ray is 1.9. The experimentally determined value is 4.4. From the above considerations it would seem that the screening effect for the L_I shell changes the "unscreened" conversion coefficient by a factor of about 3. This, of course, assumes that the percentagewise screening correction of the L_{II} shell is the same as that of the L_{III} shell.

Wu et al.[72] have obtained an accurate L_I:L_{II}:L_{III} ratio for the 46.5 kev M1 radiation following RaD beta minus decay. The value (140:10.7:1.0) they obtain is in very poor agreement with that (915:99:1.0) calculated from the tables of Gellman et al.[6] It seems that both the L_{II} and L_I intensities are about seven times the expected intensities with respect to the L_{III} intensity. This would indicate that the screening correction for the L_{II} shell is more similar to that of the L_I shell than it is to that of the L_{III} shell. Thus one might conclude that perhaps a screening effect is not the only effect operating in these two cases to alter the L_I:L_{II}:L_{III} ratios from those expected.[6]

The 26.4 kev gamma ray is given an E1 assignment from its deduced total L conversion coefficient of 3.75 ± 1.2. The value obtained from an extrapolation of the tables of Gellman et al.[6] is 2.3. The difference between these values is probably not significant since extrapolation of the theoretical curves involves great uncertainty. It is interesting, however, that the experimental value is larger than the theoretical value as in the case of the 59.6 kev gamma transition.

2. Magnetic dipole radiation. --The only transition given an M1 assignment was the 33.1 kev gamma ray of Am^{241}. Although this assignment is by no means certain, it appears to be the only one consistent with all the available data. The experimental value for the L_I:L_{II} ratio of >5 is to be compared with the theoretical value of 10 obtained by extrapolating the tables of Gellman et al.[6] Failure to observe the L_{III} line is consistent both with theoretical[6] and empirical evidence[70] on other M1 transitions.

3. Electric quadrupole transitions. -- Most of the gamma transitions for which data is reported in this thesis have been given E2 assignments. Conversion has been observed in the L_{II} and L_{III} shells only. In two cases (Np^{238} and Tl^{198m}) limits can be set for the relative amount of L_I conversion. The experimental L_{II}:L_{III} ratios are compared with the theoretical ones in Table 10.

Of course, the theoretical values are not necessarily accurate because of the fact that only three values were determined by Gellman et al.[6] for each L shell and given Z. Thus the interpolations are subject to errors of significant magnitude. The accuracy of the various experimental ratios are given in the section on experimental data. Within the conservative limits of error of both the interpolated

theoretical values and the experimental ones, the agreement in every case is good. In the three cases (7, 8, 11 in Table 10) where a resolution of the lines was sufficiently good to set limits on L_I conversion, the upper limits set are about equal to the values interpolated from the tables of Gellman et al.[6]

Table 10

Summary of Data on Electric Quadrupole Transitions

	Gamma energy (kev)	Observed in decay of:	Experimental $L_I:L_{II}:L_{III}$	Theoretical $L_I:L_{II}:L_{III}$ (E2)
1.	99.5 ± 1	Am^{241}	___:2:1	0.016:1.85:1
2.	41.0 ± 2	Am^{242m}	___:1.37:1	0.034:1.0:1
3.	43.3 ± 2	Am^{242m}	___:1.4:1	0.04:1.1:1
4.	57.8 ± 1	Pa^{228}	___:1.4:2	0.075:1.6:1
5.	130 ± 2	Pa^{228}	___:1.5:1	0.018:1.8:1
6.	52.2 ± 1	Pa^{230}	___:1.5:1	0.06:1.3:1
7.	44.1 ± 1	Np^{238}	<0.04:1.26:1	0.04:1.1:1
8.	102.0 ± 1	Np^{238}	<0.3:1.6:1	0.17:1.8:1
9.	42.4 ± 1	Np^{236}	___:1:1*	0.04:1.1:1
10.	44.2 ± 1	Np^{236}	___:1:1*	0.04:1.1:1
11.	48.4 ± 0.2	Tl^{198m}	<0.05:1.11:1.0	0.05:1.3:1

*See Section III-H.

It is clear that a theoretical calculation of L conversion coefficients at smaller intervals of energy and atomic number is required if one desires a quantitative check of theoretical ratios of L conversion. The best values in Table 10 are those for the 44.1 kev gamma ray of Np^{238} and the 48.4 kev gamma ray of Tl^{198m}. The

limits of error of these two values are probably less than those of the interpolated theoretical ones. These two determinations should provide an excellent check for a more extensive set of theoretical calculations than those of Gellman et al.[6]

4. Mixtures of magnetic dipole and electric quadrupole radiation. -- Only two transitions of all those investigated gave any reasonable indication of being an M1-E2 mixture. The first is the 43.4 kev transition following Am^{241} alpha decay. Here the $L_I:L_{II}:L_{III}$ ratio was 0.5:1.0:1.0. If all the L_I conversion arises from the M1 radiations present, a 20 percent M1, 80 percent E2 mixture is indicated. However, the uncertainty in the ratios is rather large because of the fact that the 43.4 kev L lines were situated on the low energy tail of the 59.6 kev L lines in the spectrum.

The other case is that of the 282.4 kev gamma ray of Tl^{198m}. Here the deduced K conversion coefficient (0.24) was lower than that expected (0.52) from the tables of Rose et al.[40] for pure M1 radiation. Since the expected conversion coefficient[40] for E2 radiation of this same energy is 0.076, a 64 percent E2 - 36 percent M1 mixture is indicated.

5. Higher multipole radiation. --The only transition of higher multipolarity which was investigated was the 260.7 kev gamma ray of Tl^{198m}. As can be seen in Fig. 10 the L_{III}/L_I ratio for this gamma ray would fit on a smooth curve with those of other M4 transitions in the same region of atomic number. The conversion of magnetic radiation primarily in the L_I and L_{III} shells as predicted by Tralli and Lowen[38] seems substantiated. Other criteria of comparison (K/L

ratios and lifetime) are in excellent agreement with those of well known M4 transitions (see Section III-E). Thus qualitative comparisons with the theory (for Z = 35) of Tralli and Lowen[38] and quantitative comparisons with theoretical lifetimes,[41] and empirical correlations of K/L[37] and L_{III}/L_I[39] ratios lead to an M4 assignment for this transition.

6. Conclusion. -- The results of the investigations presented in this thesis thus indicate very good agreement with the theoretical predictions of Gellman et al.[6] except for the one case of electric dipole radiation. The unexpectedly large L_I and/or L_{II} conversion coefficients found may possibly arise from a large screening effect on these shells. Such a large screening effect for L shells would not have been expected from the calculations of Reitz[68] for the K shell. It may be that screening is not the only effect altering the L_I:L_{II}:L_{III} ratios in this case.

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APPENDIX I

Miscellaneous Data on the Isotopes $Pu^{241, 240}$, Pm^{150}, and Fr^{223}

1. Plutonium 241, 240. -- The beta spectrum of Pu^{241} from a 0.5 mg sample of pile irradiated plutonium was observed on the double focusing beta spectrometer. A mass analysis of the sample indicated the following isotopic composition:

Mass number	Percent by mass
239	23.86
240	49.43
241	16.93
242	9.78

The sample was mounted on a single gold leaf (87 $\mu g/cm^2$).

A Fermi-Kurie plot of the beta spectrum is shown in Fig. 17. The fact that the plot is straight back to about 12 kev probably does not indicate that the counter window transmits 100 percent of all electrons down to 12 kev. Previous experiments seemed to indicate that the window transmitted electrons down to about 17 kev with 100 percent efficiency. The delay of the sharp downward turn of the plot is ascribed to extensive self-absorption and back scattering in the sample. These effects tend to accentuate the lower energy portions of a beta spectrum and hence in this case presumably compensated the loss due to window absorption as far down as 12 kev.

The window energy cut-off determined from this Fermi-Kurie plot is about 4.5 kev. Thus the common supposition that the energy of electrons transmitted 100 percent is between four and five times the window cut-off energy seems substantiated.

Previous work on this isotope has characterized it as a 14 year beta emitter with a maximum beta energy of 20.5 kev.[73,76] Its decay by alpha emission has been observed and the energy reported as 4.91[65] and 4.893 Mev.[9] The maximum beta energy determined from this experiment is 20.5 ± 1 kev, in excellent agreement with Freedman et al.[76]

In addition to the beta spectrum, a reasonably definite indication of the L and M lines of a 43 ± 3 kev gamma ray was observed. This gamma ray very probably arises from Pu^{240} alpha decay to the first excited state in U^{236}. This assignment is based on its abundance and energy. Other values given for this gamma ray are 49.6 kev,[76] 47 kev,[76] 48 kev,[76] 45.0 ± 0.2 kev,[77] and 44 ± 2 kev.[9]

2. Promethium 150.--A sample of ~10^7 disintegrations per minute of Pm^{150} was obtained from a proton bombardment of neodymium enriched in mass 150 in the laboratory's 60-inch cyclotron. The neodymium was bombarded as the oxide in a pistol grip holder. The beta spectrometer sample was prepared by simply taking up the oxide as a slurry in conductivity water and evaporating the material on a 0.001 inch platinum plate. The mass of the neodymium oxide on this sample was of the order of two milligrams.

The half-life of this isotope has been determined as 2.7 hours by Long and Pool,[78] Fisher,[79] and Kurbatov and Pool.[80] Beta particles of 2.01 Mev (about 70 percent) and 3.00 Mev (about 30 percent) and gamma rays of 1.4 Mev and 0.3 Mev have been reported by Fisher.[79] An absorption measurement reported by Long and Pool[78] gives the beta energy as 2.4 Mev.

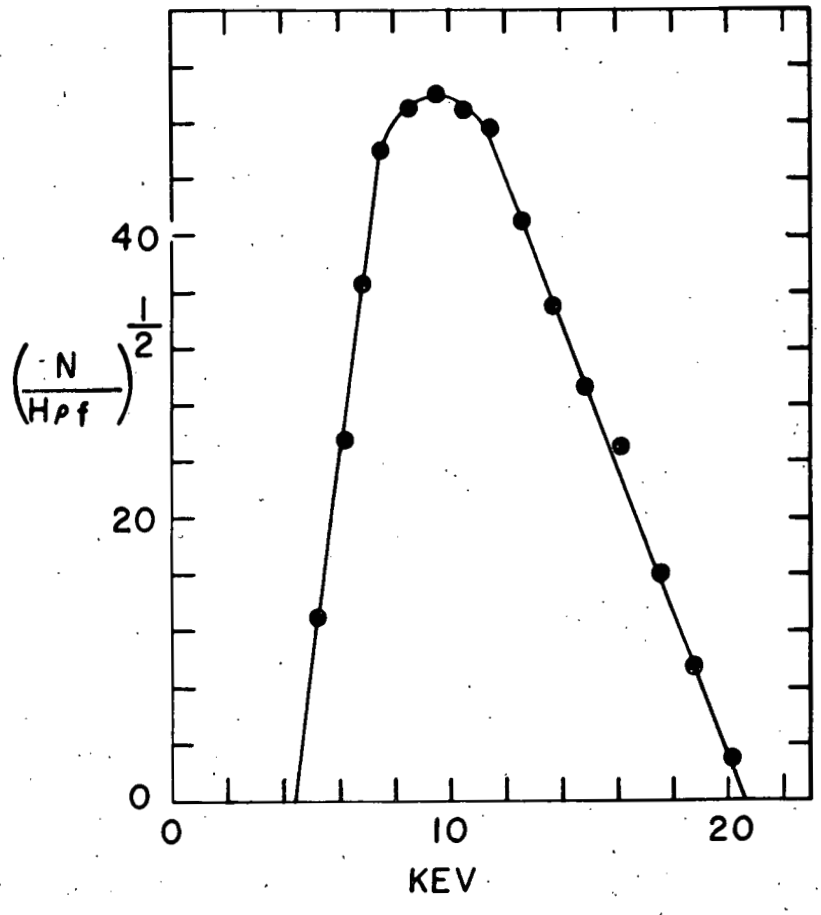

Fig. 17 — Fermi-Kurie plot of Pu^{241} beta spectrum.

Because of the low intensity of the sample and the relatively short half-life, only one electron line of Pm^{150} was characterized. No electron line of any appreciable abundance appeared below this line at 290.6 kev. This line was interpreted as a K conversion line of a 337 ± 2 kev gamma ray (a slight indication of an L line was observed). The region in which the L line occurred was not swept with sufficiently close intervals to allow a relative abundance measurement to be made.

Hibdon and Muehlhause[81] have reported gamma rays from the neutron capture of Sm^{149} to be 336.7 kev and 440.2 kev. The energy of the former is in excellent agreement with the above value from this experiment. Thus it appears that a significant proportion of the beta decay of Pm^{150} decays through the level giving rise to this 337 kev gamma ray.

Scharff-Goldhaber[20] has interpreted the 336.7 kev gamma ray as the transition between the first excited state of Sm^{150} and the ground state. From the observation of a 440.2 kev gamma ray, the second excited state is supposed[20] to be at 777 kev. From the data on Pm^{150}, it can be said that the 440_K line is less than about 1/3 of the 337_K line. Recent experiments at this laboratory indicate that several high-energy gamma rays (around 1.5 Mev) of high intensity are also emitted in Pm^{150} beta decay.[82] Thus one might reasonably conclude that the primary route of decay is to levels about 1.5 Mev or higher above the ground state from which are emitted the several high energy gamma rays, a significant fraction of which cascade through the first excited state to ground. Apparently the second excited state is involved to a smaller extent in these cascades than the first excited state. It is

of interest to note that this pattern of decay is noted among other odd-odd beta emitters in the medium and heavy elements where sufficient decay energy exists, for example, Np^{238} and Ac^{228}.

3. Francium 223 (AcK). --Several experiments were attempted to determine the beta spectrum of Fr^{223}. The sample was prepared by E. K. Hyde of this laboratory using a procedure involving a silicotungstic acid precipitation.[83] The activity was milked from a 20 millicurie source of Ac^{227} which had originally been made in a pile bombardment of Ra^{226} by the reactions:

$$\underset{1622\ y}{Ra^{226}}(n, \gamma)\underset{41.2\ min}{Ra^{227}}\ \beta^- \rightarrow \underset{22.0\ y}{Ac^{227}}.$$

The Fr^{223} arises from the 1.2 percent alpha decay branching of Ac^{227}. The half-life of Fr^{223} is 21 minutes.[84]

Previous work on the beta spectrum of Fr^{223} indicates a beta energy of 1.2 Mev.[85-87] The data here obtained indicates a two-component beta spectrum, the most abundant component having an energy of 1.0 ± 0.1 Mev. Because of the much lower intensity of the higher energy component the limits of error for its energy are greater than 0.1 Mev. The value obtained was 1.3 ± 0.15 Mev. The large limits of error quoted are due primarily to spectrometer calibration uncertainties at the time of the experiment.

Electron lines which might be assigned to gamma rays with energies 45 ± 7 kev and 230 ± 20 kev, were observed in a very fast sweep of the spectrum. Recent work by Hyde[88] using a scintillation spectrometer shows that gamma transitions occur at the energies 49.8 ± 0.2, 80 kev, 215 kev, and 310 kev. Previous workers had found by absorption methods gamma transitions of 90,[89, 90] ~330,[87]

and 48.6 kev.[87]

From the high intensity of the 49.8 kev photon relative to the beta particles[88] it is quite reasonable to assume that it is of E1 character. Hyde[88] found that none of the other gamma rays observed are in coincidence with the 49.8 kev gamma ray. A unique decay scheme cannot be constructed for Fr^{223} without further information.

APPENDIX II

A Twin Lens Beta Spectrometer for Coincidence Measurements

A. Purpose and Design of the Instrument

The decision to construct a back-to-back double beta spectrometer was based upon the usefulness of such an instrument in elucidating decay schemes, especially among heavy element isotopes where a large proportion of the gamma transitions take place primarily with the ejection of orbital electrons. Thus this unit was designed to be an electron-electron coincidence spectrometer, consisting essentially of two thin lens beta spectrometers connected at the sample end so that both could simultaneously receive radiation from the same sample, yet operate independently. The design arrived at was a modification of one described by K. Siegbahn.[91] A schematic diagram of the instrument and a block diagram of its associated equipment is given in Fig. 18.

B. Brief Description of Major Components

1. Lens coils. -- The lens coils are designed for low voltage - high current, motor-generator power and hence require cooling. The cooling system is similar to that described by Freedman.[92] Units consisting of copper tubing sandwiched between two copper plates

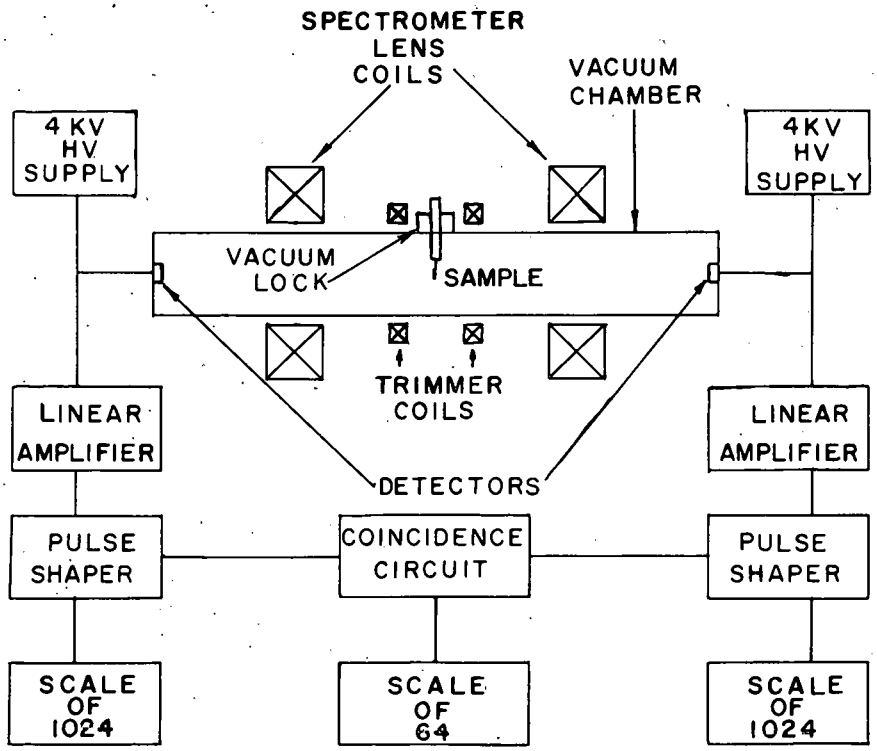

Fig. 18—Block and schematic diagram of twin lens beta spectrometer and associated equipment.

were built pancake fashion at two positions between the coil windings. Copper cooling coils are also soldered to the external surface of the lens spool. The coils are supported independently on a welded aluminum stand.

2. Trimmer lenses. -- A smaller lens coil near the sample is connected in series opposition with the lens on the same side to cancel the effect of that spectrometer on the other.

3. Vacuum chamber. -- The vacuum chamber was made by rolling and welding 0.250 inch aluminum alloy sheet. It is supported at each end by a three-point suspension coming from the coil mount and in addition, at the center by a two point mount coming from the aluminum stand. The latter is necessary for the making of initial adjustments.

4. Sample entrance section. -- The sample vacuum lock entrance is contained on an independent section of the vacuum chamber and may be altered or removed easily without changing the two remaining longer sections. Thus, if it is desired later to make the instrument an electron-electron angular correlation instrument, the sample entrance section may be changed to a bellows and a new support stand made, no change being necessary in the longer vacuum chamber sections or other components. The sample entrance section also has, at 180° from the sample, an entrance for a light pipe in case triple, alpha-electron or photon-electron coincidence experiments are desired. A single channel recording pulse-analyzer has been built to accommodate such experiments.

5. Counting equipment. -- The counters, amplifiers, and coincidence circuits have been designed for resolving times down to 0.1 μsec. The detectors are ordinary side window counters to be used as low pressure proportional counters. Should it be desirable to change to a scintillation counter system, alternate end plates have been constructed for the vacuum chamber to accommodate a standard laboratory photo-tube, light-pipe assembly. To take full advantage of the shorter resolving times of a scintillation counter, however, a new pair of amplifiers and a new coincidence circuit would be required. The advantage of the present system over the scintillation counters is that it counts with 100 percent efficiency down to energies below 20 kev whereas the counting efficiency of most scintillation counter systems declines rapidly below 100 kev.

6. Automatic counting control. -- One of the two spectrometers may be automatically operated step-wise over any given spectrum while the other remains focused on only one energy region. Thus the long time counts often necessary may be taken without the presence of a human operator. The automatic count control may be set to collect any given number of counts at each of 100 points at large or small intervals on any part of the spectrum. An over-ride system insures against the system's spending more than any given maximum time (<30 minutes) at any point on the spectrum. The design of this system is a copy of that designed by Olsen and O'Kelley[93] of the California Research and Development Company.

C. Expected Operating Characteristics

Each end of the spectrometer has been designed to transmit about 1 percent of the total electron radiation from the sample at any given energy at a resolution of about 2 percent if ring focusing is used. Without ring focusing the resolution will probably be closer to 3 or 4 percent. The latter is sufficient resolution for a great many experiments which the instrument is expected to perform.

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