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PRECISION MEASUREMENT OF X-RAYS AND GAMMA RAYS IN RADIOACTIVE DECAY

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(Thesis)

June, 1952

Berkeley, California

PRECISION MEASUREMENT OF X-RAYS AND GAMMA RAYS IN RADIOACTIVE DECAY

by

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DISSERTATION

Submitted in partial satisfaction of the requirements for the degree of

DOCTOR OF PHILOSOPHY

in

Chemistry

in the

GRADUATE DIVISION

of the

UNIVERSITY OF CALIFORNIA

Approved:

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Committee in Charge

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ABSTRACT

In the course of the work described in this dissertation, a 10 inch bent crystal spectrometer was used to examine the spectra of the radiation emitted in the decay of the following nuclides: Nd^{140}, Am^{241}, Pb^{210}, Pu^{239}, U^{237}, Pa^{232}, and Pa^{233}. For the greater portion of the research, the range of the instrument was limited to 10-70 kev, but improvements in collimation increased this range to 10-120 kev in the case of the Pa^{233} experiment. The radiations observed were thus for the most part L x-rays and very soft gamma rays. Calculations are made on the relative quantum yields and relative number of vacancies in the L levels, and the results compared with theoretical calculations in the literature. Wherever possible, the L_1 conversion coefficients of the gamma rays are calculated and the multipole orders deduced.

It is observed that the beta decay of U^{237} and the alpha decay of Am^{241} apparently lead to the same excited state in the daughter, Np^{237}, since a gamma ray of identical energy is found in the two cases.

An apparent anomaly is found in the energies of the uranium x-rays emitted in the decay of Pa^{232} and Pa^{233}, since the energies of the x-rays observed in these cases are higher than the values found in the case of Pu^{239}, which agree well with the accepted values from electron bombardment.

The x-ray data in the decay of Pa^{232} are used to calculate a lower limit for the branching ratio in the decay of this nuclide.

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INTRODUCTION

The measurement by diffraction methods of the x-rays and soft gamma radiation given off in radioactive decay provides a means of obtaining information of considerable interest in several respects. The higher accuracy of diffraction methods, as contrasted with absorption techniques, and even pulse analysis methods, makes it possible to determine gamma ray energies with sufficient accuracy to give much greater confidence to decay schemes and, indeed, to calculate nuclear level spacings to a much higher degree of refinement. Further, the high resolution of the method makes it possible to detect the presence of gamma radiation of energy within the range of the L x-rays of heavy elements. In the case of the transuranium elements, it is of interest to measure the values of the x-rays themselves. Measurement of the relative intensity of the L series x-rays permits calculation, when apropos, of the relative amounts of conversion in the L levels, and hence, at least in principle, of the multipole order of the gamma ray undergoing conversion.

To all these ends, the bent-crystal Cauchois-type spectrometer (Fig. 1) previously described by Barton *et al.*[1] has been modified by replacing the quartz diffraction crystal with topaz, increasing the effective crystal area, and replacing the proportional counter with a scintillation counter. The topaz crystal employed was found by standard x-ray methods to be the $Al_2F_2SiO_4$ modification, and the instrument was so adjusted as to measure reflections from the 303 planes of this

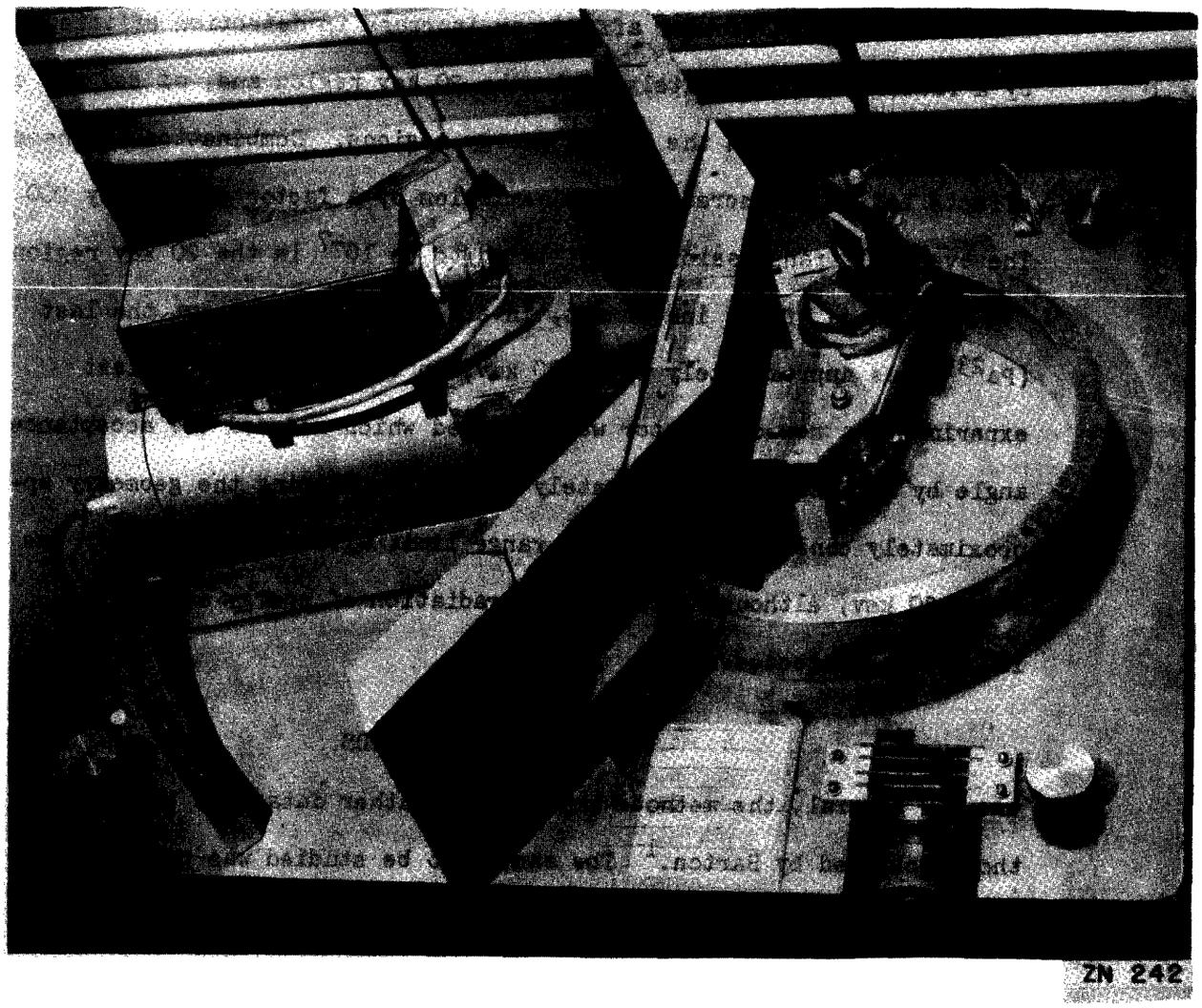

Fig. 1. Ten inch Cauchois bent crystal spectrometer.

crystal. Under these conditions, the reflectivity was found to be some five times greater than that from the 100 planes of quartz.

The effective crystal area was increased to 5.5 cm^2, or approximately three times that used by Barton, by altering the design of the crystal holder itself. The use of an NaI(Tl) - RCA 5819 photomultiplier combination in the counter affords an increase in detection efficiency by a factor of approximately 15 in the 20 kev region and, of course, a much higher factor in the higher energy regions. Combination of these effects led to an increase in transmission by a factor of roughly 200. The over-all transmission is estimated to be 10^{-7} in the 20 kev region.

The range of the instrument, in all experiments except the last (Pa^{233}) was approximately 10 to 80 kev; in the course of the last experiment, a new collimator was employed which reduced the acceptance angle by a factor of approximately two, while keeping the geometry approximately constant. The upper range limit with this collimator was some 130 kev, although very intense radiation of energy as high as 180 kev was detected.

EXPERIMENTAL METHODS

In general, the methods employed to gather data were the same as those followed by Barton.[1] The sample to be studied was prepared as a powder and mounted in a Pyrex capillary as narrow in diameter as practical (usually about 0.02 inch). Although no attempt has been made to determine the inherent resolution of the spectrometer, there is adequate reason to believe that in the experiments described here the limit of resolution lay in the width of the sample itself, for calculations of the angular width presented by the sample check well with

observed line widths.

The capillary was mounted on a lucite holder, either in a hole drilled in the lucite or in a channel on the lucite surface, and placed in the spring clips of the spectrometer sample mount (Fig. 2) which has provisions for adjustment of the sample radially, laterally, vertically, and in rotation about the radial axis. Provision is made in the design of the spectrometer for the sample mount to rotate as it sweeps through the angular range, so that the sample always presents the same face to the crystal; care was taken in adjustment of the sample to ensure that it was at the center of rotation of the mount, and that it was as nearly vertical as could be determined.

Counting pulses were led from the scintillation counter along two paths, one of which led to a counting rate meter and an Esterline-Angus recorder, which was used for monitoring and ready reference purposes only. The other path led to a standard scaling and register circuit, which was used to drive a traffic counter which periodically recorded the number of scales on a paper tape. The stamping interval of the traffic counter was set to correspond to an angular interval of either 0.01 or 0.005 units on the scale in which the spectrometer is graduated, which would correspond to 0.02 or 0.01 degrees in Bragg angle. The Microflex timer unit commonly employed in traffic counters was found to be undependable in reproducing intervals as short as 0.8 minute, and was replaced by a synchronous motor-microswitch system.

In observing the location of line peaks, angular readings were taken with a vernier capable of reading to 0.005 unit. The gear drive on the instrument cannot be relied upon to move the sample uniformly, so that readings of angle were taken every few stamps over the peaks

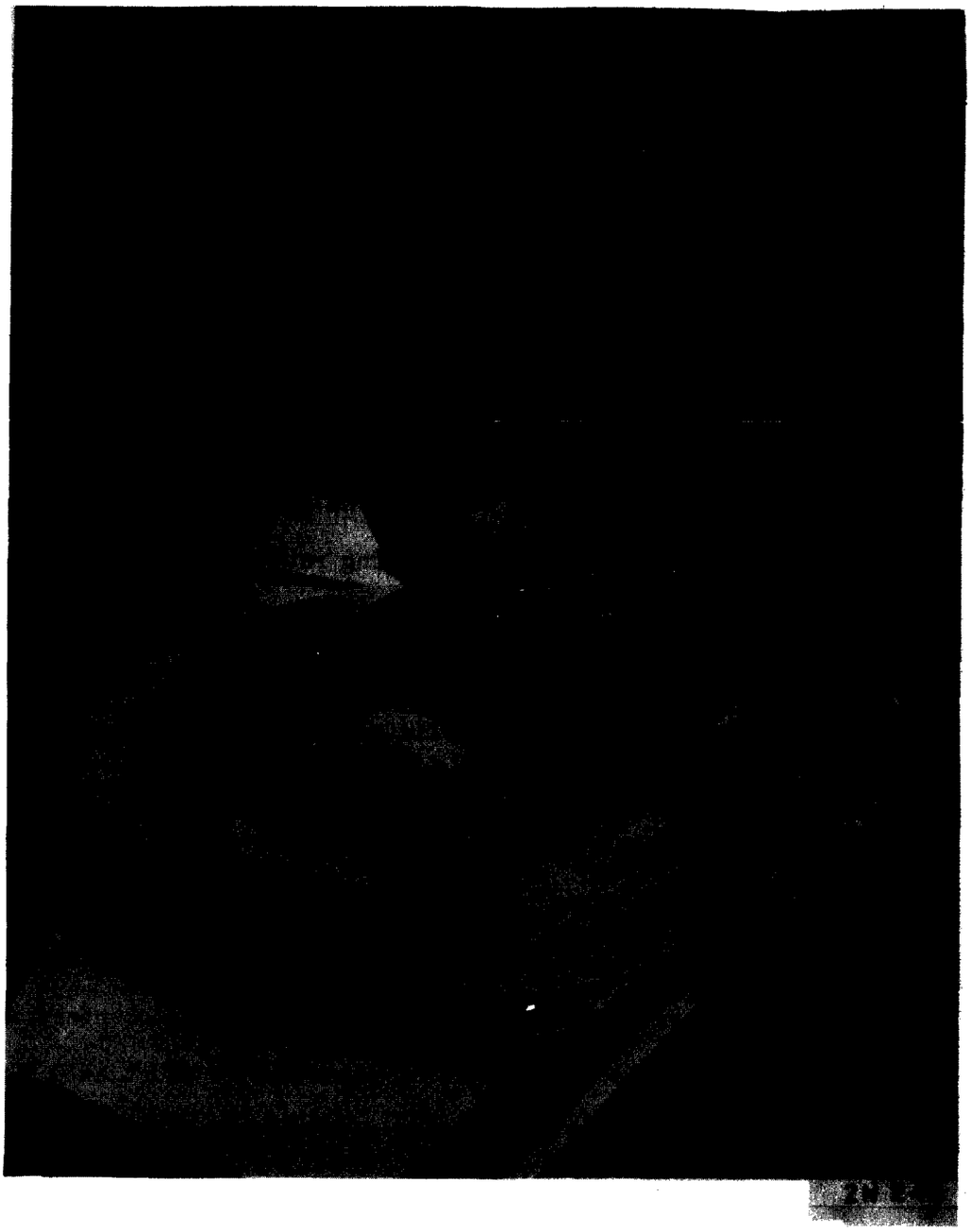

Fig. 2. Spectrometer sample mount

of lines swept.

The introduction of a scintillation counter has the disadvantage of having a background count appreciably larger than that of a proportional counter. The mounting of the scintillation crystal itself was made up of an aluminum ring approximately 1 cm high, covered at one end with 0.005 inch beryllium foil. All inner surfaces were highly polished. The scintillation crystal itself was covered with a thin film of mineral oil, placed in this ring, and taped to the surface of the RCA 5819 photomultiplier tube. This arrangement proved highly satisfactory in all respects save the fact that the pulse resolution was poor, and thus precluded the advantageous employment of the pulse discriminator circuits of the spectrometer. The background of the counter was 20-30 counts per minute.

In plotting spectra, partial smoothing of the background variations was attempted by plotting values averaged over periods somewhat longer than the stamping intervals referred to above. A common smoothing interval employed was 0.05 unit, which would correspond to a 20 minute count on the slow sweep, or a 4 minute count on the faster sweep. When half-lives permitted, the spectrum was usually explored on a sweep of one unit per 80 minutes, then more carefully examined on a sweep of one unit per 400 minutes, and finally individual lines were reswept at the slower rate. Energies were generally determined as the average of several sweeps over the line; energy uncertainties were calculated from the estimated angular uncertainty and the Bragg equation.

In practice, the Bragg angle of a line was determined by observation of the location of the line on each side of the undiffracted beam.

The angular difference between the two positions represents twice the Bragg angle, and should be unaffected by minor errors of setting of the sample with respect to the diffraction planes. The location of the midpoint of the undiffracted beam, as determined by this method, was used as a criterion for the accuracy of the spectrometer alignment. When the sample and counter were properly aligned, the midpoint was constant within experimental error over the complete spectrum; poor alignment procedure (probably in the centering of the sample in the sample mount) led to variations as high as 0.02 unit in midpoint location.

The use of topaz as a diffraction crystal is accompanied by the disadvantage that the 303 planes are oriented at an angle of $28^{\circ}59'$ from the cleavage face. Inasmuch as the spectrometer was originally designed for quartz, the angular distance through which the sample may be moved is centered about a perpendicular to the center of the crystal, and encompasses an arc of approximately 46 degrees. A 30 degree displacement of midpoint, then, leaves only some 16 degrees available on the side toward which the midpoint is displaced. Thus, lines of energy less than some 16.4 kev could be observed on one side of the midpoint only. To measure the energy of these lines, the midpoint established by higher energy radiation was assumed.

The spacing for the 303 planes of topaz, upon which all energy measurements reported here are based, was measured by x-ray methods to be 1.356 A in the unbent crystal, in good agreement with the Strukturbericht value. All conversion factors employed were based upon the values of the physical constants reported by DuMond and Cohen.[2] A calculation of the correction of the Bragg angle due to the index

of refraction of topaz indicated that it was not significant within the limits of accuracy of the instrument.

The estimation of relative intensities of the observed radiation poses a much more serious problem than the measurement of the energies and was, in general, much more uncertain. The data as obtained were subjected to three corrections: reflectivity of the crystal, absorption by matter in the path between the sample and the counter, and self-absorption in the sample itself. The reflectivity correction was made by taking the variation in reflectivity with energy as proportional to the reciprocal of the square of the energy. This relation was determined by comparison of relative intensities of lines widely removed in energy as they were reflected from quartz and topaz and assumption of the $1/E^2$ law for quartz as reported by Lind et al.[3] The correction for absorbers in the path was based on the measured thickness of these absorbers and the values for the mass absorption coefficients of the elements, from the data of S. J. M. Allen, as reported by Compton and Allison.[4] Corrections for self-absorption in the sample were made on the basis of a cylindrical sample, which is a good assumption, and were based on the relationship reported by Dixon[5] for such sources. The principal uncertainties lay in the estimation of absorption coefficients for the elements of the sample. It is unfortunately true that many members of the L x-ray series of a given element will have energies between the L_1 and L_3 edges of the neighboring elements. Data on absorption coefficients for radiation of energy between the L_1 and L_3 edge are very sparse indeed for the most thoroughly investigated elements, and were for the most part obtained by interpolation and extrapolation methods.

Thus in all experiments in which the sample had appreciable mass, or where it was isotopic with the carrier, relative x-ray intensities are rendered uncertain to the extent of the error in the approximation methods employed. This situation is aggravated even further in the cases of the transuranium elements, for which no absorption data at all are available.

For the transuranium elements, values of absorption coefficients at energies above the L_1 and below the L_3 edges were estimated from Jönsson's "universal absorption curve."[6] These values were used to construct a log-log plot of absorption coefficient versus energy which was extrapolated, on an assumption of linearity,[7] to the energies of the L_1 and L_3 edges. The magnitude of the L absorption jumps were estimated by extrapolating the data available for elements of lower atomic number.[8] Values for the absorption coefficient between the L edges were estimated by assuming a linear variation on a log-log plot of absorption coefficient versus energy and connecting the points corresponding to the bottom of the jump for one edge with the top of the jump for the next. The same general method was followed in obtaining absorption coefficient values for elements of lower atomic number, except that it was generally not necessary to employ the "universal absorption curve" to arrive at data above and below the L edge region. Such experimental values as were employed were taken from the data of S. J. M. Allen, as tabulated by Compton and Allison.[4]

The energies of the L edges of the transuranium elements were calculated by applying the correspondence principle to values of the K and L x-rays (Table I) and the L_3 edge (Table II) obtained from a Moseley extrapolation of the values tabulated by Siegbahn[9] for uranium and thorium.

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Table I. Extrapolated values of the K and L x-ray energies of the transuranium elements.

Transition	X-ray	Energy (kev)									
		Th	U	Np	Pu	Am	Cm	Bk	Cf	99	100
$K - L_2$	K α_2	90.44	94.48	96.53	98.61	100.70	102.82	104.97	107.13	109.32	111.53
$K - L_3$	α_1	93.51	97.88	100.10	102.34	104.61	106.90	109.22	111.56	113.93	116.32
$K - M_{2,3}$	β_1-β_3	105.83	110.59	113.01	115,46	117.93	120.43	122.95	125.50	128.08	130.68
$K - N_{2,3}$	β_2	109.10	114.11	116.65	119.23	121.83	124.46	127.11	129.80	132.50	135.25
$L_1 - M_2$	L β_4	15.62	16.57	17.06	17.56	18.06	18.57	19.08	19.61	20.14	20.67
$L_1 - M_3$	β_3	16.42	17.46	17.98	18.52	19.06	19.61	20.17	20.74	21.31	21.90
$L_1 - N_2$	γ_2	19.31	20.49	21.09	21.70	22.32	22.96	23.59	24.24	24.90	25.56
$L_1 - N_3$	γ_3	19.51	20.72	21.34	21.97	22.60	23.25	23.90	24.57	25.24	25.92
$L_1 - O_2$	γ_4'	20.24	21.51	22.16	22.81	23.48	24.16	24.84	25.54	26.24	26.96
$L_1 - O_3$	γ_4	20.30	21.57	22.22	22.88	23.55	22.22	24.91	25.61	26.32	27.04
$L_1 - P_{2,3}$	γ_{13}	20.46	21.75	22.40	23.07	23.75	24.43	25.12	25.83	26.55	27.28
$L_2 - M_1$	η	14.51	15.40	15.85	16.32	16.79	17.26	17.75	18.23	18.73	19.23
$L_2 - N_1$	γ_5	18.37	19.51	20.09	20.68	21.28	21.88	22.50	23.12	23.75	24.39
$L_2 - O_1$	γ_8	19.41	20.63	21.25	21.89	22.53	23.18	23.84	24.51	25.19	25.88
$L_2 - M_4$	β_1	16.20	17.22	17.74	18.27	18.80	19.35	19.90	20.46	21.02	21.60
$L_2 - N_4$	γ_1	18.98	20.16	20.77	21.38	22.00	22.63	23.27	23.92	24.58	25.25
$L_2 - O_4$	γ_6	19.60	20.85	21.49	22.13	22.79	23.46	24.13	24.82	25.51	26.22
$L_3 - M_1$	ℓ	11.12	11.62	11.87	12.13	12.39	12.65	12.91	13.18	13.45	13.73
$L_3 - N_1$	β_6	14.97	15.72	16.11	16.50	16.89	17.29	17.70	18.10	18.52	18.93
$L_3 - O_1$	β_7	16.01	16.84	17.27	17.70	18.13	18.57	19.02	19.47	19.92	20.38
$L_3 - M_4$	α_2	12.81	13.44	13.76	14.08	14.41	14.74	15.08	15.42	15.76	16.11
$L_3 - M_5$	α_1	12.97	13.61	13.94	14.28	14.61	14.96	15.30	15.65	16.01	16.36
$L_3 - N_4$	β_{15}	15.58	16.33	16.79	17.20	17.62	18.05	18.47	18.91	19.35	19.79
$L_3 - N_5$	β_2	15.62	16.43	16.84	17.25	17.68	18.10	18.53	18.97	19.41	19.86
$L_3 - O_{4,5}$	β_5	16.21	17.07	17.51	17.95	18.40	18.86	19.32	19.79	20.26	20.74
$L_1 - M_4$	β_{10}	16.98	18.03	18.56	19.10	19.65	20.21	20.77	21.35	21.93	22.52
$L_1 - M_5$	β_9	17.14	18.21	18.75	19.31	19.87	20.44	21.02	21.60	22.20	22.53
$L_3 - M_2$	t	11.47	11.98	12.24	12.50	12.78	13.03	13.29	13.57	13.84	14.12
$L_3 - M_3$	s	12.26	12.87	13.18	13.49	13.81	14.08	14.45	14.78	15.11	15.45

Table II. Extrapolated values of the K and L edges of the transuranium elements.

Edge	Th	U	Np	Pu	Am	Cm	Bk	Cf	99	100
K	109.79	115.04	117.72	120.42	123.15	125.92	128.71	131.54	134.40	137.28
L_1	20.45	21.74	22.40	23.07	23.75	24.44	25.13	25.84	26.56	27.29
L_2	19.68	20.94	21.59	22.24	22.91	23.58	24.27	24.96	25.67	26.38
L_3	16.28	17.16	17.61	18.06	18.52	18.99	19.45	19.93	20.41	20.90

In a typical experiment, such as that on Am^{241}, the correction factors for the neptunium $L\gamma_1$ (20.86 kev) with respect to the neptunium $L\beta_1$ x-ray (17.79 kev) are: reflectivity, 1.4; path absorbers, 0.8; self-absorption, 1/1.7. For the $L\alpha_1$ (13.98 kev) the corresponding corrections are: reflectivity, 0.62; path absorbers, 1.9; self-absorption, 1.9. For the 59.8 kev gamma ray the corrections are: reflectivity, 11; path absorbers, 0.56; self-absorption, 0.81. These data illustrate the fact that the self-absorption correction constitutes a large portion of the over-all intensity correction, so that large uncertainties in this correction must be reflected in concomitant uncertainties in relative intensity values when lines of energy between those of the L edges are compared. In the experiments described here intensities are calculated relative to the $L\beta_1$ x-ray, since this was usually the dominant feature of the x-ray spectrum. In general, the values obtained are considered to be accurate to no more than 30-40 percent. The intensities relative to each other of lines of energies greater than the L_1 edge of the sample absorber may be taken with greater confidence, perhaps to 10 percent.

It has been the practice of Barton and other workers in the field to compare observed relative intensities of x-rays arising from a given L shell vacancy (such as the $L\beta_1$ and $L\gamma_1$, both of which arise from an L_2 vacancy) with the relative intensities of these lines as reported by Compton and Allison[10] from electron bombardment experiments. This practice will not be followed in this work. The data tabulated by Compton and Allison have not been corrected for self-absorption in their targets, and while their relative intensities agree surprisingly well with the values obtained here before self-absorption corrections are applied, such agreement must necessarily be fortuitous and rather meaningless since there is no reason why the self-absorption corrections should be equal in the two cases.

EXPERIMENTAL RESULTS

The Decay Chain Nd^{140} - Pr^{140} - Ce^{140}

The decay chain Nd^{140} - Pr^{140} - Ce^{140} provides an almost unique opportunity for the determination of the K/β^+ ratio in Ce^{140} and the fluorescent yields of the K x-rays in both praseodymium and cerium. Nd^{140} has been reported previously[11] as an electron capturer with no evident positrons and a half-life of 3.3 days, while its daughter, Pr^{140}, is an electron capturer and positron emitter with a half-life of 3.4 minutes. No gamma radiation has been observed in either case and on this basis there can be no x-rays arising from internal conversion processes. Therefore, if a sample of Nd^{140} is permitted to equilibrate with its daughter and the x-ray spectrum of the joint decay measured, the ratio of the intensity of praseodymium K x-rays to those of cerium should give the fraction of the Pr^{140} decay which

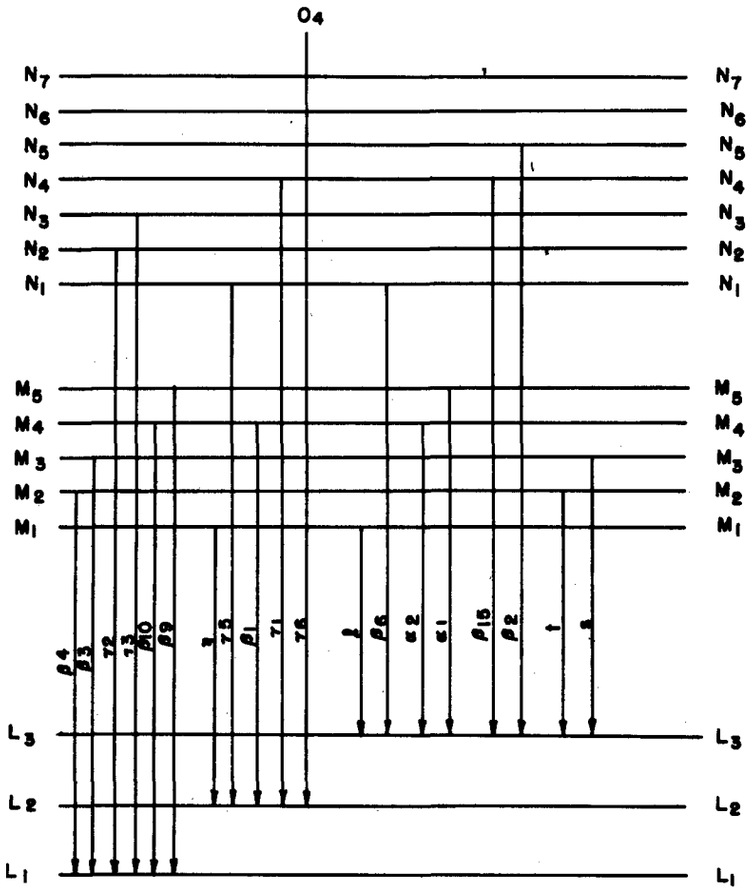

Fig. 3. Diagram of L x-ray nomenclature

(level spacings not to scale).

takes place by electron capture. Also, if the intensity of the Auger electrons can be measured with respect to the intensity of the positrons, a ratio of K x-radiation to K Auger electrons can be derived which will represent the mean fluorescent yield for praseodymium and cerium.

Nd^{140} was prepared by bombardment of 300 milligrams of praseodymium oxide with 19 Mev deuterons on the Crocker 60-inch cyclotron. Chemical purification was accomplished through the use of citrate elutions from a Dowex 50 cation exchange resin column at 83° C.[12] The purified sample was studied on the double focusing beta ray spectrometer described elsewhere by O'Kelley,[13] the 10 inch bent crystal spectrometer, and a scintillation crystal pulse analyzer.

The Bethe and Bacher approximation was used to calculate the Fermi function employed in a Fermi plot of the Pr^{140} positron spectrum which exhibited good linearity with an endpoint energy of 2.23 ± 0.02 Mev (Fig. 4). The data do not exclude the possibility of a low abundance lower energy component (ca., 1 Mev). The electron spectrum was investigated up to Hρ values corresponding to an electron energy of 1 Mev and showed only one discernible line at 28.9 ± 0.5 kev which was interpreted as electrons from Auger conversion of K shell vacancies in either praseodymium or cerium or both, as the resolution of the line does not permit this distinction.

The gamma ray spectrum from the scintillation counter showed no lines in addition to K x-rays, annihilation radiation, and Compton radiation in intensity as high as 10 percent of the K x-rays; there was meager indication of radiation of energy 1.0 - 1.2 Mev.

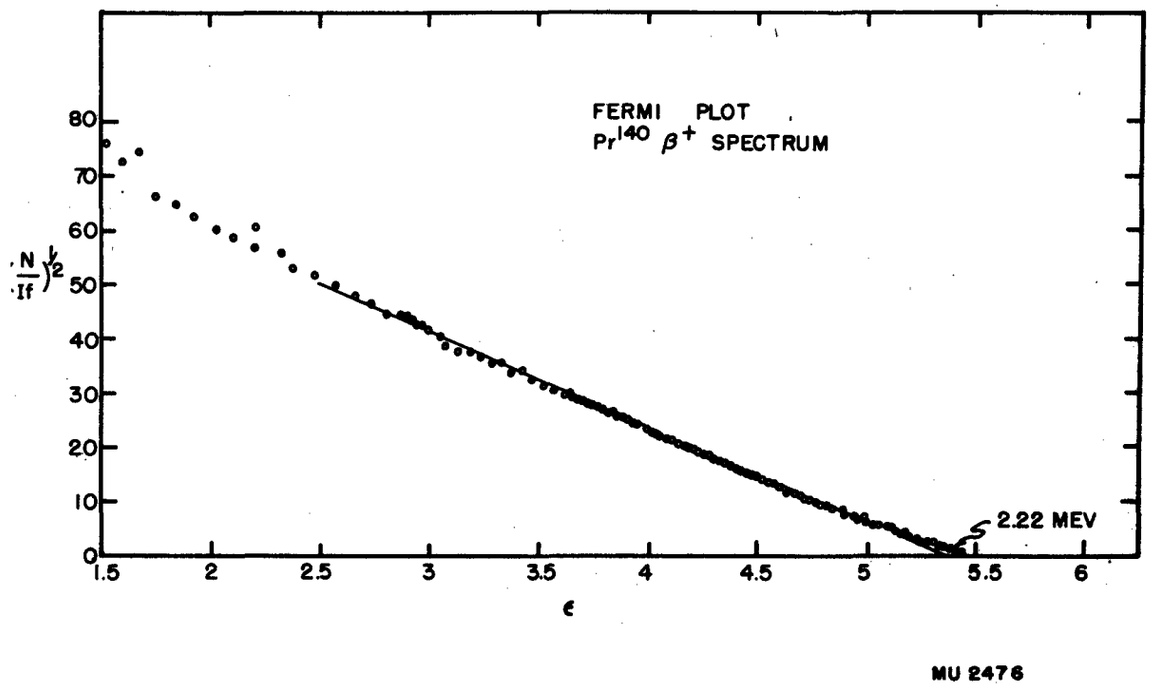

Fig. 4. Positron spectrum of Pr^{140}.

The x-ray spectrum (Fig. 5) showed only the $K\alpha_1\alpha_2$ (unresolved) and the $K\beta_1$ x-rays of both praseodymium and cerium in the following intensity ratios:

$$Pr\ K\alpha_1\alpha_2 : Ce\ K\alpha_1\alpha_2 : Pr\ K\beta_1 : Ce\ K\beta_1 = 100 : 50 : 28 : 11 .$$

Since no sources of K x-rays other than electron capture could be detected, all observed x-rays could be attributed to that source. On this basis, the electron capture to positron ratio in the decay of Pr^{140} may be computed. If we consider only K capture for the first estimation, and assume that the fluorescent yields of K x-rays are very nearly equal in praseodymium and cerium, then, since the chain is in equilibrium, the ratio of the intensity of the cerium x-rays to that of the praseodymium x-rays is a direct measure of the fraction of K capture in Pr^{140}. The K/β^+ ratio in Pr^{140} is, then, 1.0 ± 0.1.

The value of ft for the Pr^{140} decay was calculated from the theoretical values of Feenberg and Trigg[14] for $(f_k + f_+)$ to be 2×10^4, which leads to the conclusion that the transition is allowed and if the TAM (total angular momentum) of Ce^{140} is taken as 0, then the ΔI must be, by Gamow-Teller rules, unity. The ratio of f_k/f_+ is given by Feenberg and Trigg values, under these conditions, as 1.0 in good agreement with the value obtained above. If L capture is taken into consideration, these values must be changed somewhat. An internally consistent solution based upon Rose's theoretical L/K ratios,[15] the above data, and theoretical values for f_k leads to the conclusion that Nd^{140} decays by 26 percent L capture and 74 percent K capture; that Pr^{140} decays 58 percent by β^+ emission, 37 percent by K capture, and 5 percent by L capture; from which the ratio of β^+ to K in Pr^{140} is

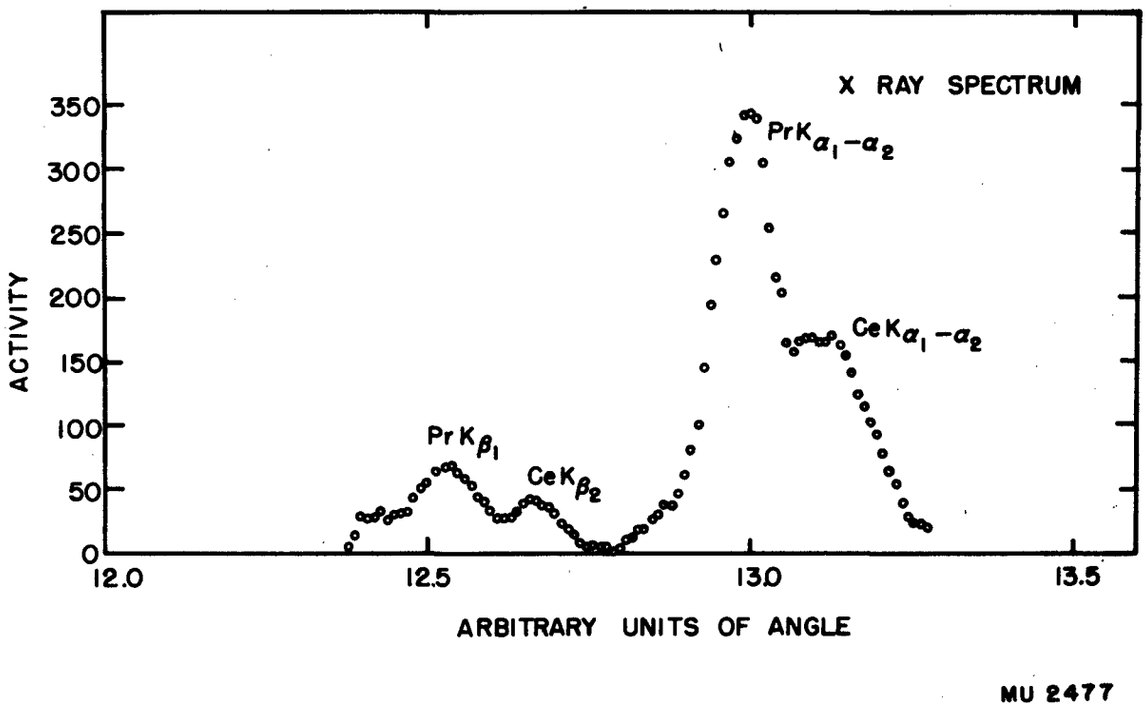

Fig. 5. X-ray spectrum of Pr^{140} and Nd^{140}.

approximately 1.6 to 1. Since the $Nd^{140} \longrightarrow Pr^{140}$ transition is also presumably allowed, the decay energy of Nd^{140} can be obtained from its half-life and comes out to be 105 ± 40 kev.

The ground state of Ce^{140} is very probably S_0 so that the state of Pr^{140}, by Nordheim's rule[16] that $\Delta \ell = 0$ for an allowed transition, must be S_1. Both the odd nucleons in Pr^{140} fall into the shell $50 \leq N \leq 82$ with possible states, by the Mayer model, of $g_{7/2}$, $d_{5/2}$, $d_{3/2}$, $s_{1/2}$, and $h_{11/2}$. Since the TAM is low, Nordheim's rule 9 should apply to Pr^{140}, leading to six possible combinations for the states describing the odd neutron and proton. However, the TAM of Pr^{141} (59 protons) is known to be 5/2 and that of Ba^{137} (81 neutrons) is 3/2, suggesting the probable choice of $d_{5/2}$ as the state of the proton, and $d_{3/2}$ as that of the neutron which is one of the combinations admitted by Nordheim's rule. If, then, the angular momenta couple antiparallel as Nordheim implies, the resulting over-all state would be S_1 in agreement with the conclusion reached earlier.

Integration of the positron and electron spectra leads to a ratio of positrons to electrons of approximately 50, leading in turn to a K/e^- ratio of 100. This implies a very high figure (0.99) for the mean K fluorescent yield for $Z = 58$ and 59. Considering that an error of a factor of ten (a very high estimate indeed) in the integrations through poor estimations of self-absorption, backscattering, etc. would reduce the value of the fluorescent yield to only 0.90, it seems justifiable to set the lower limit for W_K in this region as 0.90.

Americium 241

It is becoming generally recognized that odd A alpha emitters frequently exhibit alpha ray spectra at sharp variance with simple alpha decay theory in that the most abundant alpha particle group is not of the highest energy. Americium 241 has been found by Asaro and Perlman[17] to be a prime example of this effect, since one group comprising 84 percent of the emitted alpha particles has an energy some 70 kev less than the highest energy group. The decay of this nuclide is further characterized by the emission of abundant gamma rays of some 60 kev, as well as x-rays of approximately 20 kev.[18] The conversion electrons from this gamma ray have been measured by O'Kelley,[13] who reported the gamma ray energy as 59.4 ± 1.0 kev.

It is apparent that the accurate determination of the energies of the gamma rays emitted in the course of the decay would be of great value in the establishment of a decay scheme. In this respect, a high resolution instrument offers particular advantages, since the gamma radiation from alpha emitters will usually be very soft, and might well be indistinguishable from the L series x-rays in experiments with instruments of lower resolution.

The Am^{241} used in these experiments was mounted as the trifluoride powder in a capillary within a lucite holder. The sample contained less than 0.5 percent impurities of alpha activity. In the course of the experiments, the sample was removed from the spectrometer, replaced, and reoriented several times to find any errors due to poor orientation. In general, the spectrum was taken off topaz except that in the final observation, a quartz crystal was used to measure the energies of the

most energetic gamma ray and the $L\beta_1$ x-ray. The positions and intensities of all peaks were checked several times.

The X-Rays.-- Fig. 6 shows a typical spectrum in the region 10-22 kev, and Fig. 7 shows in greater detail the resolution of the $L\alpha_1$ and $L\alpha_2$. The reality of some of the peaks illustrated in Fig. 6 was determined on the basis of their reappearance in consecutive runs and on both sides of the undiffracted beam. In many cases, what would appear at first examination to be a peak would not reappear in subsequent runs, and in several instances could be traced to the presence of large radioactive sources in nearby laboratories.

Table III presents the energies and intensities of the lines which have been assigned as neptunium x-rays, such assignment being based upon the propinquity of the observed energy to the expected energies of neptunium x-rays from extrapolation of the Moseley relation, and upon rough consistency in relative intensity with that observed in other experiments for x-rays arising for a given vacancy. Column 3 of Table III presents the extrapolated values for neptunium x-rays, and column 4 shows the values obtained by Barton[1] for some of the lines observed here. The agreement with extrapolated values is good in all cases except the $L\beta_2$ and the $L\gamma_1$, while the agreement with Barton's values is excellent. The relative intensities were calculated on the bases indicated above.

On the basis of these intensities and line assignments, the relative quantum yields, i.e., photon intensities, of the three L levels may be calculated. This is done by summing the intensities of all lines arising from a given shell vacancy. Since not all possible lines

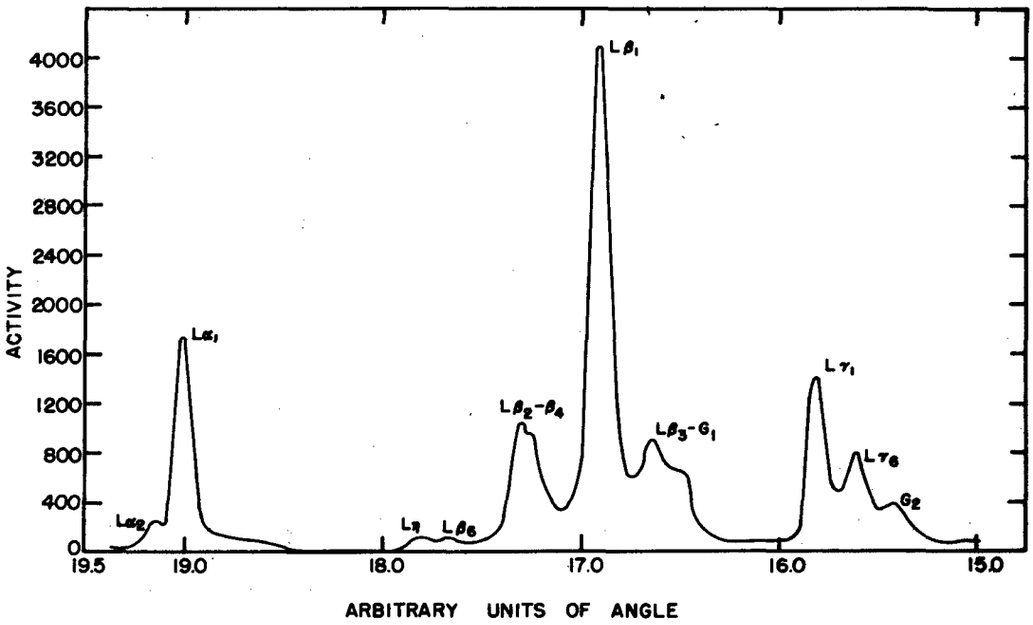

Fig. 6. Spectrum of neptunium x-rays in Am^{241} decay.

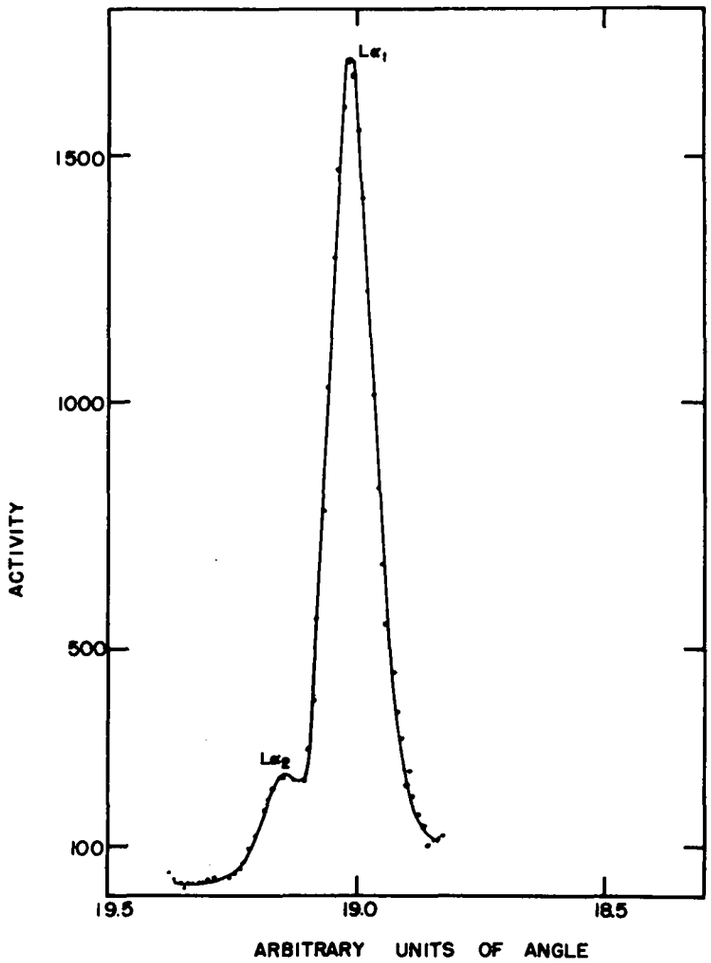

Fig. 7 Resolution of the neptunium $L\alpha_1$ and $L\alpha_2$ x-rays in Am^{241} decay.

Table III. Energies and intensities of neptunium x-rays from Am^{241} decay.

Line	Transition	Energy observed here (kev)	Energy observed by Barton (kev)	Extrapolated energy value (kev)	Relative intensity
$L\alpha_2$	$L_3 \longrightarrow M_4$	13.79 ± 0.02		13.76	10
$L\alpha_1$	$L_3 \longrightarrow M_5$	13.98 ± 0.02	13.98 ± 0.03	13.94	70
$L\eta$	$L_2 \longrightarrow M_1$	15.89 ± 0.04		15.85	5
$L\beta_2$	$L_3 \longrightarrow N_5$	16.98 ± 0.02	16.94 ± 0.05	16.84	30
$L\beta_4$	$L_1 \longrightarrow M_2$	17.12 ± 0.04		17.06	20
$L\beta_1$	$L_2 \longrightarrow M_4$	17.80 ± 0.02	17.79 ± 0.03	17.74	100
$L\gamma_1$	$L_2 \longrightarrow N_4$	20.86 ± 0.02		20.77	60
$L\gamma_6$	$L_2 \longrightarrow O_4$	21.45 ± 0.08		21.49	30

are observed, for reasons of low intensity, resolution, etc., those which are observed must be weighted by a factor corresponding to the fraction of total intensity which they represent as described below. This is particularly necessary for the L_I level, since only the $L\beta_4$ was observed. To obtain this weighting factor, the results of this and other experiments were combined. For example, in the experiment with Pa^{233} to be described below, the series of lines resulting from an L_1 vacancy were much more prominent than in the present case. Accordingly, the weighting factor for the $L\beta_4$ was based principally upon the Pa^{233} results. The ratios of quantum yields, after these calculations, are:

$$L_1 : L_2 : L_3 = 1 : 3.0 : 2.5 .$$

If there were available data on the fluorescent yields of the L levels, the relative number of vacancies of these levels in the decay could be calculated from the quantum yields above. No such data are available, but Kinsey[19] has arrived at approximate figures for the fluorescent yields on the basis of a combination of such data as are pertinent (width of the levels, etc.) and theoretical considerations. His published values for the fluorescent yields of the uranium L_1, L_2, and L_3 levels are 0.16, 0.59, and 0.41, respectively. These yields should be applicable to the neptunium case, since the values do not change very rapidly with atomic number in this region, according to Kinsey's calculations. On this basis, the relative number of vacancies in the L shells is:

$$L_1 : L_2 : L_3 = 1.0 : 0.8 : 0.97 .$$

These figures take no cognizance of the phenomenon known as the Coster-Kronig effect, in which an L_1 vacancy is filled by an L_3 electron. This is usually not a radiative transition, since the $L_1 - L_3$ energy difference is used to eject one of the M electrons, leaving the atom in a doubly ionized state. This transition is followed by the filling of the L_3 and M vacancies with electrons from the outer shells, with the emission of the appropriate x-rays. The x-rays are slightly different in energy from those normally obtained from the filling of an L_3 vacancy, since the double ionization of the atom changes the energetics of the situation and constitute satellites to the normal lines. Detection of the effect was made possible through the observation and explanation of these satellite lines. It is not at all certain that the $L_1 - L_3M$ Coster-Kronig transition is highly favored over the

$L_2 - L_3M$, or $L_2 - L_3N$, as the energetics may permit, but Kinsey has chosen this possibility in the absence of any data to the contrary. On the bais of this assumption, he has calculated values for the coefficient for the $L_1 - L_3M$ transition, and arrives at a figure of 60 percent for atomic numbers in the vicinity of uranium. If the relative vacancy data mentioned above are modified on this basis, one obtains:

$$L_1 : L_2 : L_3 = 1.0 : 0.8 : 0.4$$

which is in excellent agreement with O'Kelley's data from beta spectroscopy:

$$L_1 : L_2 : L_3 = 1.0 : 0.8 : 0.3 .$$

It is clear that in this instance the ratio of vacancies in the L_1 and L_2 shells is quite different from that observed by Arnoult[20] in the case of the decay of Bi^{212}, where he found a ratio of

$$L_1 : L_2 = 10 : 1 .$$

The Gamma Rays.-- The photon spectrum in the energy region 22-80 kev is shown in Fig. 8, and Table IV presents the energies and relative intensities of the lines not attributable to neptunium x-rays. The intensities (relative to each other) of the lines of energy greater than 22 kev should be more dependable than those of the softer radiation since the absorption corrections to these intensities are not subject to the large uncertainties arising from the L edges of americium.

It is interesting to note that the line labelled G_1 corresponds closely in energy to the extrapolated value for the $L\beta_1$ x-ray of americium, and that labelled G_2 is fairly close to the extrapolated value for the americium $L\gamma_1$ x-ray. If these are indeed americium x-rays, there is at this time no explanation for their presence.

The line labelled G_4 is seen to have almost exactly half the energy of the prominent 59.8 kev line, G_7. This line was tested for

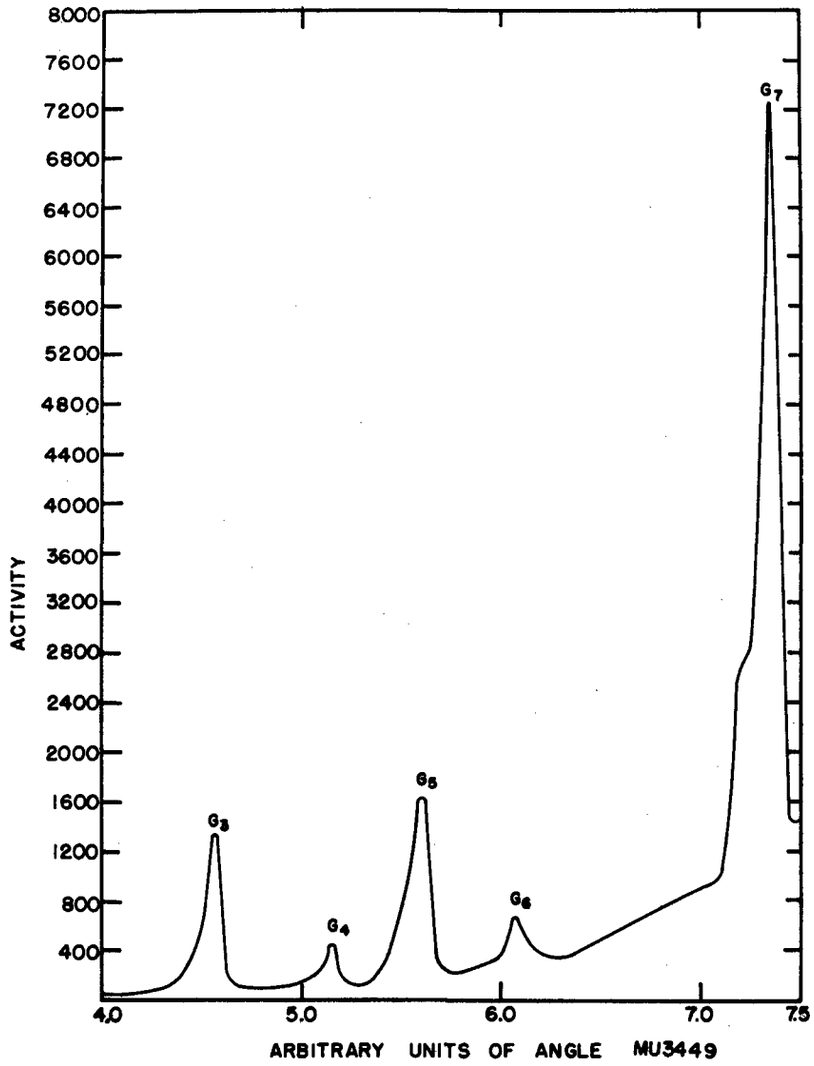

Fig. 8. Spectrum of the energy region 22-80 kev in Am^{241} decay.

Table IV. Energies and intensities of the gamma rays in Am^{241} decay.

	Energy (kev)	Intensity (relative to $L\beta_1$)
G_1	18.83 ± 0.03	15
G_2	22.20 ± 0.08	10
G_3	26.43 ± 0.03	55
G_4	29.82 ± 0.05	20
G_5	33.36 ± 0.03	50
G_6	38.00 ± 0.08	10
G_7	59.78 ± 0.04	180

second order reflection of the 59.8 line by interposing absorbers between the source and diffraction crystal in the spectrometer, and it was found that the intensity of G_4 decreased very closely as the intensity of the more energetic G_7. It may therefore be assumed that most, if not all, of G_4 arises as a second order reflection of G_7. Thus the lines which may be taken as nuclear gamma rays in Am^{241} decay are those labelled G_3, G_5, G_6, and G_7.

In earlier unpublished experiments with a proportional counter, Crane and Ghiorso[21] have observed 0.4 "60" kev gamma rays per alpha disintegration. This datum, coupled with Asaro's figure of 84 percent of total alpha decay to the level which may be taken as the source of the 59.8 kev gamma ray, leads to the conclusion that of the alpha transitions to that level, decay by emission of a 59.8 kev gamma ray takes place approximately 50 percent of the time. If one now considers O'Kelley's[13] approximate figure that the gamma ray is 50 percent

converted, it may be concluded that the decay from this level is predominantly by a 59.8 kev transition, either radiative or otherwise, and hence that this gamma ray may be taken as the principal source by far of L vacancies.

On this basis, the conversion coefficient in the L_1 shell may be computed from the number of L_1 vacancies and the intensity of the gamma ray. Such a calculation leads to the result $\alpha_{L_1} = 2.5$. On the basis of the relativistic, non-screened calculations of Gellman et al.,[22] the conversion coefficients in the L_1 shell for a gamma ray of this energy are: electric dipole, 0.17; electric quadripole, 4.5; magnetic dipole, 30. Choosing the closest of these figures, the 59.8 kev gamma ray may be taken as an electric quadripole, from which the vector spin change is 2, with no parity change. However, considering the ratio of L_1 to L_2 vacancies and the later publication of Gellman et al.,[22] it seems probable that the radiation is an $E_2 - M_1$ mixture.

Fig. 9 presents a partial decay scheme for Am^{241}. The level energies are based upon Asaro's data from the alpha spectrum, and it may be seen that the 59.78 kev value for the principal gamma ray is in good agreement with the level energy differences from the alpha work. The energies of the lines labelled G_3 and G_5 add almost exactly to 59.78 kev, so that these lines are assumed to be in cascade with the principal gamma ray. The energies of these gamma rays, also, are in good agreement with the corresponding level energy differences for the alpha decay. The 38.00 kev gamma ray has not been located on this diagram, since no corresponding level differences appear in the alpha spectrum. It would not be surprising, however, if alpha transitions

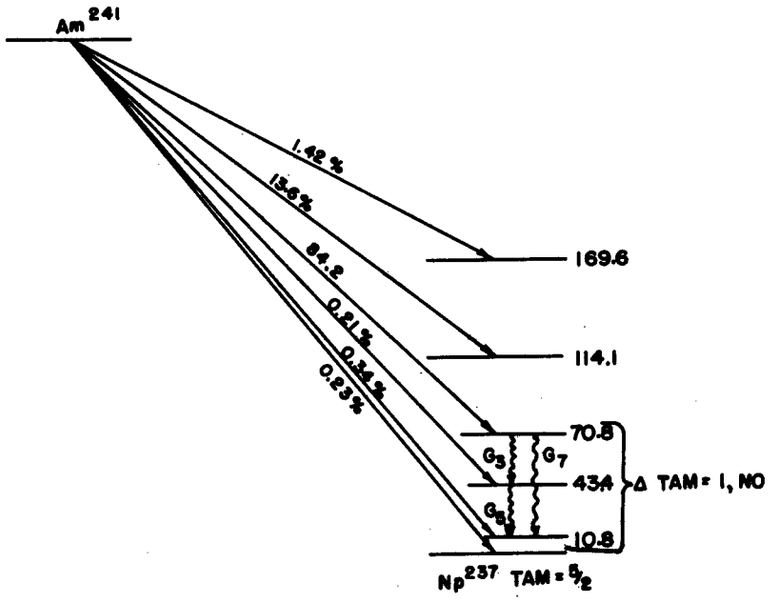

Fig. 9. Partial decay scheme in Am^{241}.

corresponding to this energy difference did take place, since several of those observed were barely measurable.

The coincidence measurements of Prohaska[23] strengthen the location of the 59.8 kev gamma ray in the decay scheme, since his high alpha conversion electron coincidence rate demands that this gamma ray be located between levels lower than that sought by the most abundant alpha transition.

Some aspects of these data may be used as a basis of comparison with other experiments. Thus, if only the L_1 and L_2 vacancies are considered, the 59.8 kev gamma ray is, by these data, 80 percent converted in the L shell. On the basis of Crane and Ghiorso's figure of 0.4 59.8 gamma rays per alpha particle, and Asaro's data, the gamma ray would be 50 percent converted, while the data of Prohaska and O'Kelley lead to a figure of 60 percent conversion. Also, the data here lead to a ratio of total L x-ray quanta to 59.8 kev quanta of 2.1. This is to be compared with the result of Crane and Ghiorso[21] that this ratio is 1.8, while Martin[24] finds a ratio of 1.7.

It is noteworthy that no gamma rays were discernible which could be assigned to transitions to the ground state and, further, that no gamma ray of approximately 41 kev was detected, since the energy interval between the most highly populated alpha transition and the next most abundant (14 percent) corresponds to this value.

Lead 210

Lead 210, or RaD, offers the opportunity to compare data from this spectrometer with the results obtained by many other workers. The activity is available in large quantities as a natural decay product,

and has thus received the intense scrutiny to which most members of the naturally occurring radioactivities have been subjected. Frilley,[25,26] Tsien,[27] and other workers in the Curie laboratory have studied its radiations by bent-crystal spectrometer and cloud chamber methods; Kinsey[28] has studied its x-radiation by absorption methods; Feather[29] has reviewed the results reported by the many other workers, and recently Bannerman and Curran[30] have studied the gamma rays on a scintillation spectrometer.

In the work to be described here, some 5 millicuries of RaD from radon decay were separated from RaE and RaF by repeated precipitations as $PbCrO_4$. At some time in its previous history, large quantities of inactive lead carrier had been added to the sample. While the presence of the carrier simplified the chemistry considerably, its presence introduced into the intensity calculations the difficulties noted in the Introduction. The separation factor from RaE was determined by counting of alpha particles to be approximately 100 by activity. The mounted sample contained some 2 milligrams of inactive lead.

The X-Rays.-- Table V presents the observed energies and relative intensities of the lines assigned as bismuth x-rays, and compares the energies with the values tabulated by Siegbahn[9] for bismuth x-rays from electron bombardment. Column 6 lists the relative intensities as observed by Frilley[26] from RaD decay. Fig. 10 shows a typical spectrum in the x-ray region.

It is noteworthy that the spectrum contains, in high relative abundance, x-rays arising from vacancies in the L_1 shell. The appearance of these lines is rather a novelty in this laboratory inasmuch as they have been observed in reasonable intensity only in the case of RaD and Pa^{233}.

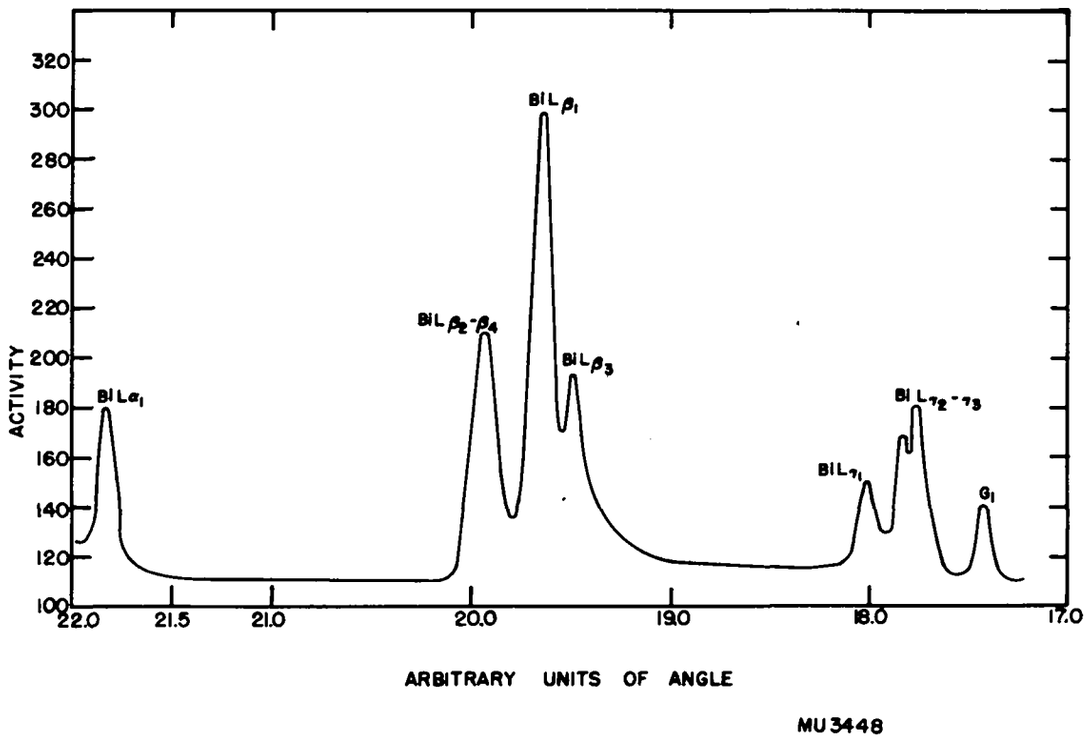

Fig. 10. Spectrum of the bismuth x-rays in Pb^{210} decay.

Table V. Energies and intensities of the bismuth x-rays from Pb^{210} decay.

Line	Transition	Energy observed here	Siegbahn's energy value	Intensity here	Intensity (Frilley)
$L\alpha_1$	$L_3 - M_5$	10.86 ± 0.03	10.84	125	125
$L\beta_4$	$L_1 - M_2$	12.71 ± 0.02	12.69	50	31
$L\beta_1$	$L_2 - M_4$	13.04 ± 0.02	13.02	100	100
$L\beta_3$	$L_1 - M_3$	13.23 ± 0.03	13.21	120	50
$L\gamma_1$	$L_2 - N_4$	15.30 ± 0.05	15.25	80	
$L\gamma_2$	$L_1 - N_2$	15.58 ± 0.06	15.58	55	
$L\gamma_3$	$L_1 - N_3$	15.71 ± 0.06	15.71	54	

The agreement of the observed energies with the values tabulated by Siegbahn is generally excellent; only the $L\gamma_1$ shows deviation beyond that expected from experimental uncertainties.

The relative quantum yields of the L shells, calculated on the basis of the intensities listed in Table V, are:

$$L_1 : L_2 : L_3 = 4 : 3 : 2 .$$

If Kinsey's values[19] for the fluorescent yields of the L_1 and L_2 shells (0.12 and 0.57, respectively) and the experimental value of 0.37 for the fluorescent yield of the L_3 shell are employed, the number of relative vacancies in the L shells is:

$$L_1 : L_2 : L_3 = 1.0 : 0.14 : 0.16 .$$

The number of L_3 vacancies must again be corrected for the effect of the Coster-Kronig transition $L_1 - L_3M$. If we attribute all the L_3 vacancies to this type transition, the coefficient for the transition

is 16 percent which must represent an upper limit. Such a value is in very poor agreement with the value of 70 percent estimated by Kinsey.[19]

The agreement with Frilley's results is not very good at all. The initial discrepancy in relative intensities is probably due to the method of calculation of self-absorption in the source, since this correction is particularly critical in the case of the $L\beta_3$, which lies between the L_1 and L_3 edges of lead. Frilley does not indicate his method employed to calculate his reported intensities, or whether heavy self-absorption corrections were necessary. Even using this data, however, it is not at all clear how his reported ratio of L_1 to L_2 excitation was determined, since use of fluorescent yields as reported by Kinsey will lead to a ratio not nearly as high as Frilley's value of 18 : 1.

If the data obtained here are used to compute the ratio of intensity of the Lβ x-rays to the Lα group, a figure of 2.1 is obtained which is to be compared with Frilley's value of 1.0 and the values of Kinsey[28] and Riou[31] of 1.4 and 1.5, respectively.

The Gamma Rays.-- Table VI presents the energies and intensities of the gamma radiation observed here and lists the energies and intensities of the gamma rays observed by Tsien[32] in cloud chamber experiments. The agreement in energies of the lines common to the two lists is seen to be very good. Insofar as the intensities of the lines observed by Tsien but not found here are concerned, an upper limit of 5 percent of the 46.7 kev intensity can be placed on the intensity of the 42.6 kev line; such a value is not inconsistent with Tsien's data. There were meager indications of radiation in the vicinity of 30 kev, but of such low intensity as to be indistinguishable from the background variations.

Table VI. Energies and intensities of the gamma rays in Pb^{210} decay.

Energy here	Energy (Tsien)	Intensity (relative to $L\beta_1$)	Intensity (Tsien) (normalized so that $I_{46.7}$ is equal)
16.33 ± 0.05	16.1 ± 0.4	50	
23.31 ± 0.03	23.2 ± 0.6	50	285
46.69 ± 0.05	46.7	800	800

The line listed as 23.31 kev was shown by interposition of absorbers to be a second order reflection of the 46.7 kev line, since their intensities were decreased by the absorbers at almost exactly the same rate, which agreed well with the decrease expected from the mass absorption coefficients of S. J. M. Allen. The identification of this radiation as a second order reflection leads to a sharp disagreement with Tsien's value for intensity of this radiation. Tsien observes, in the cloud chamber, radiation of energy very nearly 23.31 kev in intensity approximately 35 percent of the 46.7 kev line. If it is assumed that a line of this energy is present and is close enough in energy to the second order reflection of the 46.7 kev line to make the two unresolvable in the experiments described here, then the intensity observed here should be greater than that observed by Tsien to the extent of the contribution of the second order reflection; however, this is decidedly not the case. The intensity observed here is some 6 percent of that observed for the 46.7 kev line (a very reasonable value for a second order reflection) which is less than Tsien's value by a factor of six.

On the basis of the intensities of the x-rays and gamma rays observed here, the ratio of total L vacancies to 46.7 kev quanta is 3.6, and if the assumption is made that this is the only gamma ray undergoing conversion in the L shell (an apparently very good assumption, considering the intensities of the other gamma rays capable, energy-wise, of producing conversion), it may be concluded that the gamma ray is 78 percent converted in the L shell. If Richardson's value[33] is accepted that 84 percent of total conversion takes place in the L shell, the gamma ray is calculated to be 95 percent converted, which is in good agreement with Stahel's value[34] of 97 percent.

From these data the conversion coefficient in the L_1 shell may also be calculated leading to a value for α_{L_1} of 3.1 in excellent agreement with Tsien's value from beta ray spectroscopy of 2.9. On this basis, and the calculations of Gellman et al.,[22] it may be concluded that the 46.7 kev gamma ray is an electric quadripole - magnetic dipole mixture.

At the present time the generally accepted decay scheme for RaD[29,30] pictures the decay by beta emission (E = 18 kev) to a 46.7 kev level in RaE, with several modes of de-excitation of this level (Fig. 11). This model of the decay scheme has been strengthened considerably by the coincidence work of Bannerman and Curran,[30] and accounts for all gamma radiation reported (with the exception of the source of the bismuth K x-rays reported at one time by Tsien) by the numerous workers with this activity.

If we assume that RaD has zero total angular momentum (TAM) and even parity, since it is an even-even nucleus, and that RaF has the

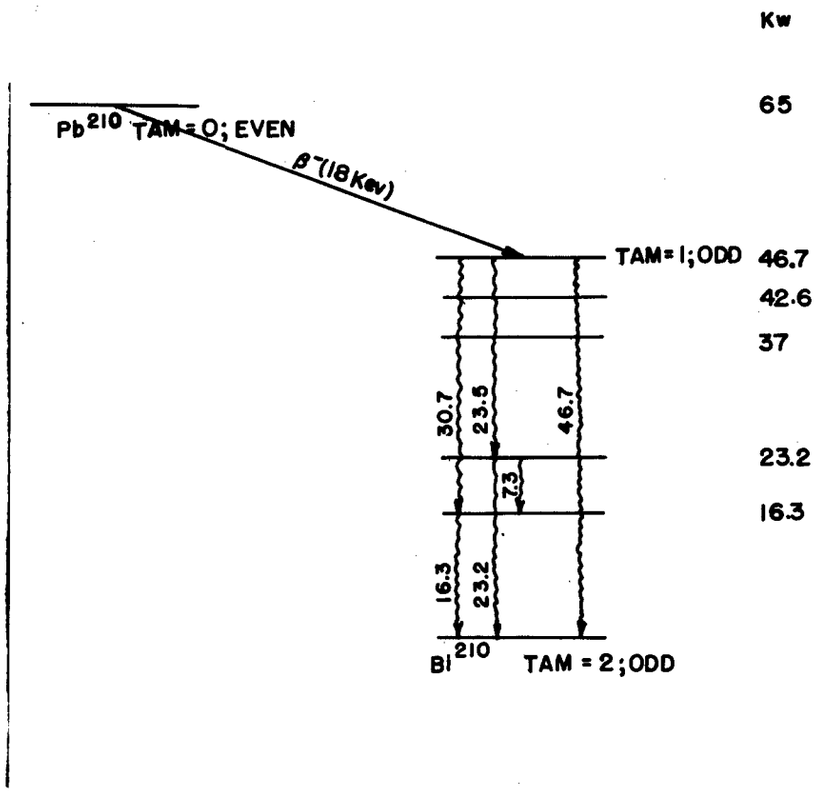

Fig. 11. Decay scheme of Pb^{210}.

same characteristics, then the classification of the beta decay of RaE into the first forbidden category ($\Delta I = 2$, yes) by Nordheim[35] leads to the conclusion that RaE has TAM of 2 and odd parity. If the 46.7 kev gamma ray is, as the data here imply, an electric quadripole - magnetic dipole mixture, then it involves a change of one unit in TAM and no parity change. Reasoning on the basis of ft values, the beta decay of RaD would appear to be either an allowed transition ($\Delta I = 0$, no) or a first forbidden transition ($\Delta I = 1$, yes). Choice of the latter for the beta transition and of the ($\Delta I = 1$, no) rules for the electric quadripole - magnetic dipole permit an assignment to the 46.7 kev level of TAM = 1 and odd parity.

Plutonium 239

Plutonium 239, like americium 241, is an odd A alpha emitter, and might be expected to exhibit a complex alpha particle decay scheme. Asaro[17] and others have shown that such is, in fact, the case. Examination of the spectrum of Pu^{239} thus offers the same interest as Am^{241}, in that measurement of the energy of any soft gamma radiation present would be of great value in the establishment of a decay scheme, and the determination of multipole orders, if possible, could be used to considerable advantage in extension of alpha systematics.

In the present work, approximately 10 milligrams of isotopically pure Pu^{239} were precipitated as the fluoride and mounted in the usual way. The very low specific activity of the sample resulted in such low intensity of all the observable lines that quantitative estimations of intensities were not worthwhile. The positions of the observable lines were quite reproducible, however, so that the energy values

should be reasonably reliable.

Table VII presents the energies of the radiation observed, and compares these energies with Siegbahn's[9] values for the x-rays of uranium. The agreement is very good in the cases of the $L\beta_1$ and $L\beta_3$. The lack of agreement in the case of the $L\alpha_1$ is not surprising for the case of a very low-lying line visible on only one side of the undiffracted beam. The presence of the $L\beta_3$ with no discernible $L\beta_4$ can be explained only on the basis of higher absorption in the case of the latter. The $L\beta_3$ itself was barely discernible, and since the $L\beta_4$ energy is lower, and thus further from the L_3 edge in plutonium, its absorption would be expected to be considerably greater.

Table VII. Energies of uranium x-rays in Pu^{239} decay.

Line	Transition	Energy here	Energy (Siegbahn)
$L\beta_1$	$L_2 - M_4$	17.24 ± 0.04	17.22
$L\beta_3$	$L_1 - M_3$	17.44 ± 0.08	17.45
$L\gamma_1$	$L_3 - M_5$	13.69 ± 0.07	13.61

No radiation assignable as gamma radiation was visible, though particularly close search was made in the vicinity of 14 kev, since Asaro's data on the alpha particle spectrum of Pu^{239} show levels of 14 kev separation in U^{235}. On the basis of the measurement of West and Dawson[36] of 0.04 L x-rays per alpha particle emitted, it is concluded that the number of radiative 14 kev transitions is less than 0.01 per alpha particle.

As mentioned above, the generally low intensity of the x-radiation makes calculation of the relative line intensities subject to so great an error as to be impractical. It is noteworthy, however, that the $L\beta_1$ was once more the dominant feature of the spectrum.

Uranium 237

Uranium 237, a 6.5 day beta emitter, has the same daughter nucleus, Np^{237}, as that formed in the alpha decay of Am^{241}. It is of considerable interest to discover which, if any, of the excited states in the daughter nucleus in beta decay are the same as those reached in the alpha decay. Melander and Slatis[37] report a gamma ray of 56.7 kev energy in U^{237} decay on the basis of beta spectroscopy on the conversion electrons. Such a result raises the question that a more accurate examination of this radiation might reveal it to be identical in energy with the 59.8 kev line observed in Am^{241} decay.

The uranium 237 was prepared by irradiation in the Argonne pile of a sample of some 2 milligrams of uranium oxide, of which the uranium was 95% U^{236}, 4% U^{235}, and 1% U^{234} and U^{238} by weight. This sample was irradiated for approximately 2 weeks, and upon its return was processed by repeated extractions of the uranium into diethyl ether and precipitations with ammonium hydroxide. The final hydroxide precipitate was dried and mounted in the usual way. The activity was unfortunately considerably less than that expected - presumably due to low pile flux - and gave only some 7000 counts per minute in the undiffracted beam of the spectrometer. For a complete and thorough examination of the spectrum of an activity of this half-life, the sample should be more active by a factor of at least five.

The X-Rays.-- Table VIII presents the energies and relative intensities of the observed radiation assignable as neptunium x-rays. The accuracy of the intensity values is rather lower than usual, because of the comparatively low ratio of peak heights to background. For example, the $L\alpha_1$ line was only some 4 counts per minute over background at the peak. The energies, however, compare very well indeed with those values obtained for corresponding lines in the decay of Am^{241}. As in the case of Am^{241}, the $L\beta_1$ was the dominant line of the x-ray spectrum, while x-rays arising from L_1 vacancies were not observable at all. So few lines were observed that it was considered advisable, in the calculation of relative level quantum yields and intensities, to obtain the total relative intensity from a given level by dividing the sum of the intensities of the observable lines characteristic of that level by the fraction of the total level radiation which these lines should contain, basing this last figure on other samples, principally Pa^{232} and Pa^{233}, for reasons which will be noted below. This procedure seems completely justifiable, since the choice of M, N, or O electrons to fill a given L vacancy should be completely independent of the method of formation of that vacancy, and should be, according to the data of Compton and Allison, a very slowly varying function of atomic number.

Table VIII. Energies and intensities of the neptunium x-rays in U^{237} decay.

Line	Transition	Energy here	Energy Am^{241}	Energy extrapolated	Intensity
$L\alpha_1$	$L_3 - M_5$	13.98 ± 0.04	13.98 ± 0.02	13.94	27
$L\beta_1$	$L_2 - M_4$	17.81 ± 0.04	17.80 ± 0.02	17.74	100
$L\gamma_1$	$L_2 - N_4$	20.85 ± 0.06	20.86 ± 0.02	20.77	42
$L\gamma_6$	$L_2 - O_4$	21.51 ± 0.08	21.45 ± 0.08	21.49	31

No L_1 lines were discernible, as noted above, so that all figures for that level are upper limits, and are based upon an estimation of the highest possible intensity which would be indiscernible from background variations. On this basis, the relative quantum yields are:

$$L_1 : L_2 : L_3 = 1.0 : 3.9 : 1.2 .$$

Employing, as usual, Kinsey's quasi-empirical values for the fluorescent yields, the ratio of vacancies is:

$$L_1 : L_2 : L_3 = 1.0 : 1.1 : 0.47$$ (ignoring the Coster-Kronig effect).

The situation is complicated in this case by the presence in high intensity of K x-rays, presumably from conversion of high energy gamma rays. It was estimated[24] from data taken with a scintillation spectrometer that the K x-ray intensity was equal to that of the only gamma ray visible in the spectrum. On this basis, and employing fluorescent yield for the K x-rays of ~100%,[38] the L vacancies can be corrected for vacancies arising from the filling of vacancies in the K shell. (This process will not affect the L_1 level.) The results of this correction lead to a ratio of L vacancies from other processes of:

$$L_1 : L_2 : L_3 = 1.0 : 0.7 : 0.3 .$$

These data cannot be wisely employed to compute either multipole orders through the α_{L_1}, or give values for the Coster-Kronig coefficient for the $L_1 - L_3M$ process, since the L_1 number represents only a rough upper limit.

The Gamma Rays.-- Only one line was discernible which could not be assigned as a neptunium x-ray. An average of several sweeps over this line leads to the conclusion that the gamma ray has an energy of

59.78 ± 0.08 kev, and an intensity of 210, relative to an $L\beta_1$ intensity of 100. The agreement with the energy value determined in Am^{241} is exact, an event probably fortuitous in nature, as the angular uncertainties give rise to fairly high energy uncertainties in this energy region. It may be stated with considerable confidence, however, that the transition in Np^{237} involves the same two levels in this case and in the decay of Am^{241}. Unfortunately, the low sample activity precluded the establishment or denial of the existence of the gamma rays of 26.5 and 33.3 kev, whose presence is demanded by the partial decay scheme put forth for Am^{241}.

The agreement of these data with those from Am^{241} is generally quite poor. For example, the ratio of the total x-ray intensity to the intensity of the 59.8 kev gamma ray is in Am^{241}, 1.9, while in U^{237} it is only 1.2 before correction for the K x-rays. Unless the decay scheme is quite different in the two cases and the agreement in energy of the two gamma rays is entirely fortuitous, one would expect the ratio of x-rays to the 59.8 gamma ray to be the same in the two cases (after correction for the K x-rays) since the conversion coefficient of the 59.8 kev gamma ray should be the same, and it might be assumed to be the principal converter in the L shell. Even if the higher energy gamma rays are appreciably converted in the L shell, this effect would lead to higher values of the ratio of x-rays to the gamma in the case of the U^{237}, rather than in Am^{241}, which is the implication of the data. The intensities of all lines were generally low, however, so that the experiment should be repeated with more activity before any significant conclusions may be drawn.

Protactinium 232

In the cases in which a nuclide undergoes decay by two modes, examination of the x-rays emitted in the decay offers an excellent method of determination of branching ratio, provided that both modes of decay lead to excitation of the x-rays of the daughter and that some method can be used to arrive at a determination of the number of x-ray events per disintegration. This last figure need not, of course, be on an absolute basis as long as it offers a ratio of the number of x-ray events per disintegration by one mode to the number by the other. A case in point is that of Pa^{232}, a 1.32 day beta emitter. Although branching has never been observed in this decay, calculations based upon closed cycles of radioactive decay lead to the conclusion that the decay energy in electron capture is approximately 0.6 Mev. Further calculations based on ft values obtained from theoretical calculations of f_K and f_L, and grouping of the ft values for the known electron capturers in this region, lead to the conclusions that the electron capture half-life should be between 2 and 20 hours if the transition is allowed and 300 to 3000 hours if first forbidden.

The former possibility, at least, offers the opportunity to see the thorium x-rays in the decay. To establish this, Pa^{232} was prepared by bombardment of Th^{232} with protons on the Crocker 60-inch cyclotron. Purification and separation of the Pa^{232} was accomplished by successive extractions into di-isopropyl ketone from an 8 M hydrochloric acid solution and return into a solution approximately 0.2 M in perchloric acid. The procedure was established as bringing

about a very good separation and purification by following the decay of the sample. A semi-logarithmic plot of activity versus time exhibited very good linearity, and decayed into background with no visible "tail" of longer-lived activities.

The method offers the additional advantage that the Pa^{232} can be obtained in "weightless" form, so that a carrier can be chosen for the precipitation and mounting which has a K edge far below the energies of the L x-rays of uranium. By this means, the intensity calculations need not have the uncertainties characteristic of the samples in which the carrier was isotopic with the activity. In the present experiment, iron was chosen as the carrier, and precipitation of the hydroxide with ammonium hydroxide carried the protactinium activity very satisfactorily.

The X-Rays.-- Table IX presents the energies and relative intensities of the lines observed in the x-ray region. Column 3 lists the results of the first bombardment, and those of another bombardment undertaken to confirm the first. It is seen that the energies are uniformly higher than the values tabulated by Siegbahn for uranium x-rays. In order to ascertain whether this effect was instrumental, the sample was re-alined in the spectrometer for each of the determinations shown in Table IX. To eliminate the possibility of a change in the instrument, Am^{241} was rerun after each of the bombardments. The energy of the neptunium $L\beta_1$ x-ray was found in each case to be identical with the value previously determined.

Table IX. Energies and intensities of the uranium x-rays in Pa^{232} decay.

Line	Transition	Energy here	Energy (Siegbahn)	Intensity
$L\beta_1$	$L_2 - M_4$	17.32 ± 0.07		100
		17.31 ± 0.04	17.22	
		17.30 ± 0.06		
		17.29 ± 0.02		
		17.29 ± 0.02		
$L\beta_2$	$L_3 - N_5$	16.48 ± 0.02	16.43	17
		16.51 ± 0.06		
$L\alpha_1$	$L_3 - M_5$	13.65 ± 0.02	13.61	28
$L\alpha_2$	$L_3 - M_4$	13.46 ± 0.02	13.44	23
$L\gamma_1$	$L_2 - N_4$	20.26 ± 0.04	20.16	40
		20.26 ± 0.09		
$L\gamma_6$	$L_2 - O_4$	20.92 ± 0.06	20.85	15
$L\beta_3$	$L_1 - M_3$	17.54 ± 0.07	17.46	10

No explanation of this anomaly has yet been reached. The good agreement of the experimental values for the bismuth x-rays in RaD decay with Siegbahn's tabulated values, plus the agreement of the value obtained for the uranium $L\beta_1$ x-ray in the decay of Pu^{239}, add considerable weight to the validity of the data as they stand. Attempts at explanations based upon the isotope shift, or upon the interaction of an excited state of the nucleus with the electron levels, fall short by orders of magnitude in explaining the approximate

60 volt shift implied by the data. The most promising explanation would seem to be one based upon a double ionization, which is capable of causing shifts of this order of magnitude, but such explanations demand so many conditions as to seem unlikely.

No x-rays were observable which could be attributed to thorium. Consideration of the maximum intensity which could have escaped detection leads to an upper limit for the intensity ratio of the thorium $L\beta_1$ to that of the uranium $L\beta_1$ of 1:34.

The intensity data of Table IX lead to a ratio of quantum yields of

$$L_1 : L_2 : L_3 = 1 : 3.9 : 1.3$$

and, if Kinsey's data for the fluorescent yields are employed, the vacancy ratios are

$$L_1 : L_2 : L_3 = 1 : 1.05 : 0.49 \text{ (ignoring the Coster-Kronig transitions).}$$

These figures are not very meaningful, however, since a large number of K x-rays is present, which will lead to L vacancies through K shell filling. On the bais of observations on a scintillation crystal pulse analyzer, the ratio of K to L x-rays is about unity.[24] Taking the K fluorescence yield as about 100 percent at this atomic number,[38] the number of K vacancies may be taken as equal to the number of L x-rays from which the number of L vacancies may be corrected for those arising from K vacancies, assuming the ratios of the K x-rays to be

$$K\alpha_1 : K\alpha_2 : K\beta_1\beta_3 : K\beta_2 = 100 : 50 : 45 : 20$$

by extrapolating the data listed in Compton and Allison.[39] This correction leads to the ratios of L vacancies from L conversion processes:

$$L_1 : L_2 : L_3 = 1 : 0.82 : 0.03$$

again ignoring the Coster-Kronig transitions.

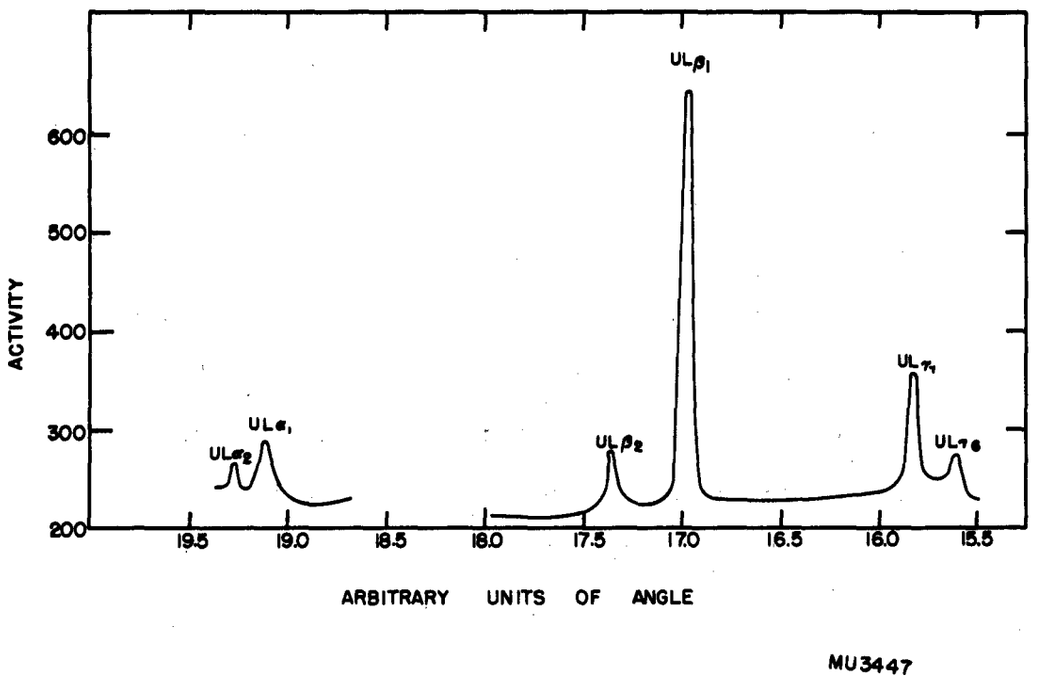

Fig. 12. Spectrum of the uranium x-rays in Pa^{232} decay.

Theoretical calculations on the capture process at this atomic number and with this decay energy lead to the conclusion that the process should be 80 percent K capture in an allowed transition. Using Kinsey's fluorescent yields, the K x-ray ratios noted above, and the relative x-ray yields from a given L shell as derived from the data on Pa^{233} (see Table X), there should be 0.085 visible thorium $L\beta_1$ x-rays per capture event (assuming all L capture from the L_1 shell only).

Analysis and integration of the e^- spectrum of Pa^{232} in a double focusing beta ray spectrometer leads to a value of 0.15 K conversion electrons per beta particle emitted. From the measured ratio of K to L x-rays mentioned above and the observed number of $L\beta_1$ x-rays, the number of observed $L\beta_1$ x-rays of uranium per beta particle is calculated to be 0.062. Taking the lower limit of thorium $L\beta_1$ x-rays to uranium $L\beta_1$ x-rays as 34 to 1, the number of beta event per capture event must be at least 46 to 1. Such a branching ratio leads to a lower limit for the capture half-life of 1400 hours, which is within the limits set for a first forbidden transition.

Protactinium 233

The inexplicably high values for uranium x-rays observed in the decay of Pa^{232} suggested observation of Pa^{233} as a quasi-definitive experiment in determining whether the effect were real or instrumental. The experiment would be definitive, of course, only if the effect were real and confined to Pa^{232}; in such a case, the uranium x-rays from Pa^{233} decay would have normal energies, and comparison of Pa^{232} and Pa^{233} samples in succession would eliminate the possibility of

instrumental changes as a function of time. Pa^{233} offers several advantages and interesting aspects in its own right, since it may be produced in very high specific activity by pile bombardment of Th^{232}, has a long enough half-life (27.4 days) for convenient experimentation, and has been reported[40] to have a number of soft gamma rays of energies within the range of the bent-crystal spectrometer.

Some 10 grams of thorium were bombarded as thorium nitrate in the Oak Ridge pile for a period of approximately 2 weeks. The protactinium was separated and purified by the same chemical methods described in the section on Pa^{232}, and the activity mounted as coprecipitant on ferric hydroxide. The activity of the mounted sample was approximately 10^{11} disintegrations per minute. In the course of the examination of the spectrum, a new collimator was mounted on the spectrometer. The finest collimator previously available consisted of lead plates 4 inches long, 1 inch high, and 2 millimeters thick, with 2 millimeter spacing between plates at the end of the collimator nearest the crystal. (Inasmuch as the beam is slightly divergent as it comes off the diffraction crystal, the collimator plates are not mounted parallel, but diverge to approximately the same degree as the beam of diffracted radiation.) The new collimator was constructed of tantalum plates 0.020 inch in thickness, spaced 0.020 inch apart. The transparency of the collimator thus remains constant, while the acceptance angle is markedly reduced. The use of the new collimator permitted observation of radiation of considerably higher energy.

The X-Rays.-- Table X presents the energies of the observed radiation attributed to uranium K and L x-rays, the energies of uranium x-rays as tabulated by Siegbahn, and the observed intensities as compared to the $L\beta_1$. It is apparent that the same deviation from tabulated values is present as in the case of the uranium x-rays from Pa^{232} decay and, indeed, the agreement with the energy values found in the Pa^{232} case is acceptable as within experimental error. As a result, the experiment casts no great amount of additional light upon the problem. If the shift is due to a double ionization effect, it is not surprising to find it in Pa^{233}, since this nuclide is notorious for the complexity of its decay scheme with regard to the number of emitted gamma rays.

Included in Table X with the energies of the radiation attributed to uranium K x-rays are two lines which are considered to be second order reflections of the K x-rays. Although absorption experiments were not performed, the agreement of the energies of these lines with one-half the energies of the K x-rays is considered to be fairly conclusive evidence of the second order relationship, particularly in view of the fact that the appearance of two gamma rays having an energy fitting this relation can be taken as a very remote possibility. Inasmuch as the range of uncertainty in energy is considerably less at the energies of the second order reflections, the energies of the second order reflections may be taken as a better measure of the K x-ray energies than the observations made on the x-rays themselves. On this basis, the observed energies of the K x-rays are seen to be notably higher than the energies tabulated by Siegbahn.

Table X. Energies and intensities of the uranium x-rays in Pa^{233} decay.

Line	Transition	Energy here	Energy Pa^{232}	Energy (Siegbahn)	Intensity
$K\alpha_1$	K - L_3	99.0 ± 0.8 (1st order)	--	97.88	380
		99.0 ± 0.4 (2nd order)	--		9
$K\alpha_2$	K - L_2	95.1 ± 0.7 (1st order)	--	94.48	190
		94.9 ± 0.4 (2nd order)	--		6
$L\alpha_2$	L_3 - M_4	13.47 ± 0.02	13.46 ± 0.02	13.44	7.2
$L\alpha_1$	L_3 - M_5	13.65 ± 0.01	13.65 ± 0.02	13.61	56
$L\eta$	L_2 - M_1	15.45 ± 0.02	--	15.40	2
$L\beta_6$	L_3 - N_1	15.75 ± 0.02	--	15.72	2
$L\beta_2$	L_3 - N_5	16.49 ± 0.02	16.48 ± 0.02	16.43	35
$L\beta_4$	L_1 - M_2	16.60 ± 0.03	--	16.57	31
$L\beta_1$	L_2 - M_4	17.27 ± 0.01	17.29 ± 0.02	17.22	100
$L\beta_3$	L_1 - M_3	17.51 ± 0.01	17.54 ± 0.02	17.46	23
$L\beta_{10}$	L_1 - M_4	18.09 ± 0.03	--	18.03	2
$L\beta_9$	L_1 - M_5	18.27 ± 0.02	--	18.21	2
$L\gamma_5$	L_2 - N_1	19.59 ± 0.03	--	19.51	1.5
$L\gamma_1$	L_2 - N_4	20.25 ± 0.02	20.26 ± 0.04	20.16	37
$L\gamma_2$	L_1 - N_2	20.55 ± 0.02	--	20.49	13
$L\gamma_3$-$L\gamma_6$	L_1 - N_3	20.85 ± 0.04	--	20.72	16
	L_2 - O_4			20.85	
$L\gamma_4$	L_1 - O_3	21.62 ± 0.02	--	21.57	6

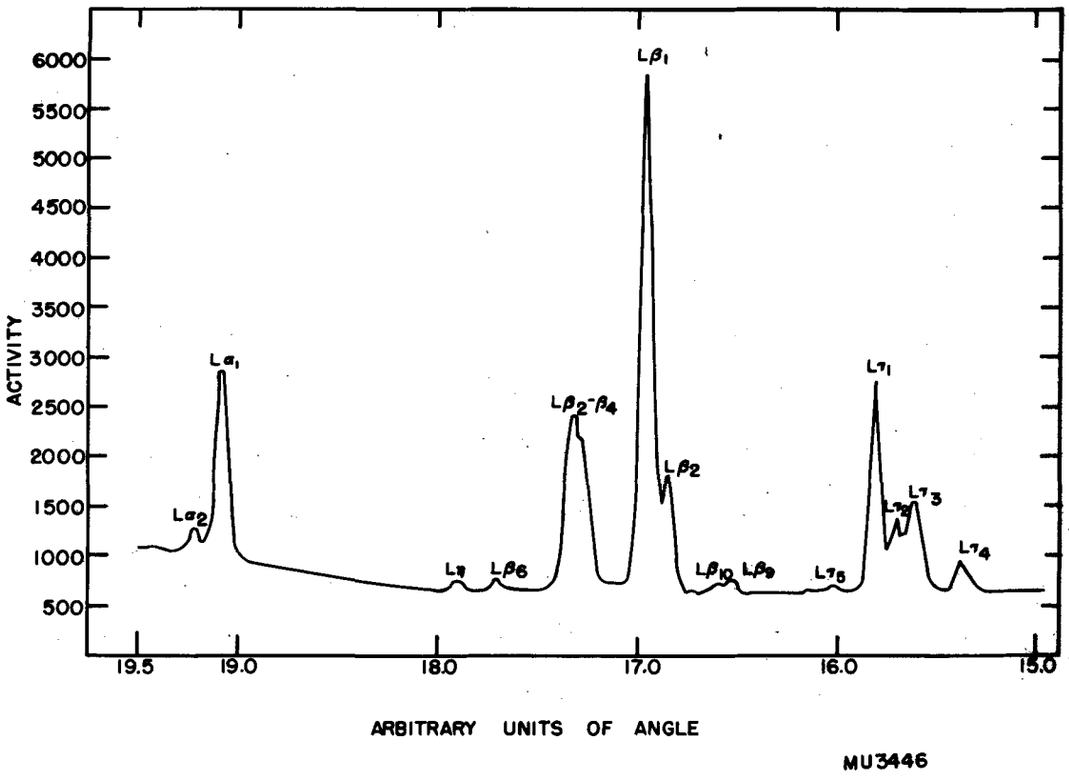

Fig. 13. Spectrum of the uranium x-rays in Pa^{233} decay.

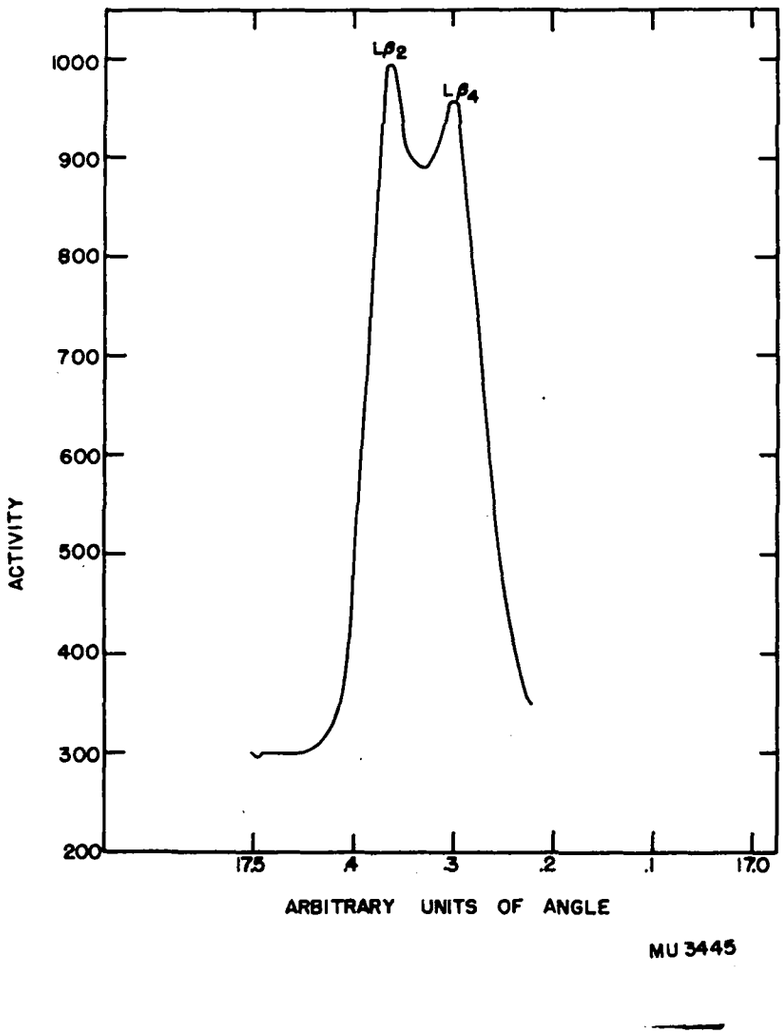

Fig. 14. Resolution of the uranium $L\beta_2$ and $L\beta_4$ x-rays in Pa^{233} decay.

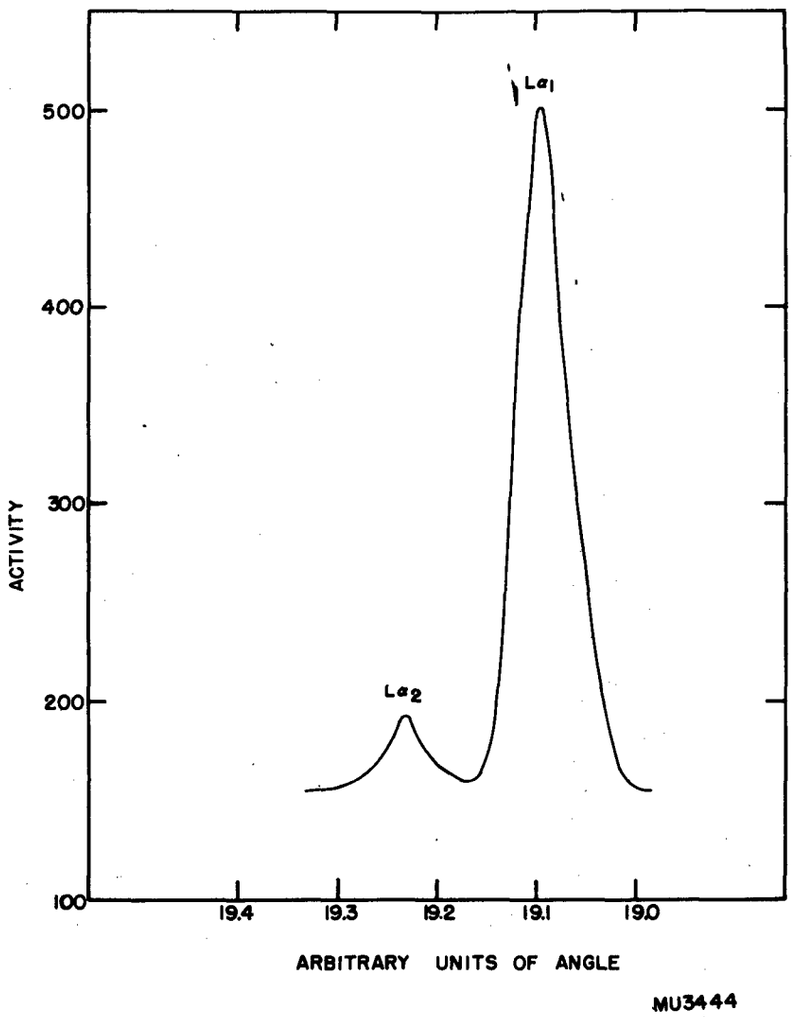

Fig. 15. Resolution of the uranium La_1 and La_2 x-rays in Pa^{233} decay.

In attempting an explanation of this discrepancy, a Moseley extrapolation was made on the basis of the tabulated K x-ray energies of lead and bismuth. The extrapolated value for the thorium $K\alpha_2$ was 90.48 kev, and that of the $K\alpha_1$, 93.54 kev, in very good agreement (0.05 percent error) with the tabulated values of 90.43 kev and 93.51 kev, respectively. In the case of uranium the comparison was much less satisfactory. The extrapolated values for the uranium $K\alpha_2$ and $K\alpha_1$ were 95.22 and 98.53 kev, while the tabulated values are 94.47 and 97.87 kev, a difference of some 0.7 percent. It is difficult to understand such a sharp change in the accuracy of the Moseley relation, and, indeed the results suggest at once that the tabulated data on uranium x-rays may be in error. On the strength of this possibility, an extrapolation was made based upon the values for thorium K x-rays, using the $\Delta(\nu/R)^{1/2}$ obtained from the data on lead and bismuth. This calculation leads to the result that the $K\alpha_2$ of uranium is 95.17 kev and the $K\alpha_1$ 98.50 kev. When these figures are compared with the experimental results (averaging the first and second order reflections) of 95.0 ± 0.2 and 99.0 ± 0.2, agreement is reached to 0.1 percent in the case of the $K\alpha_2$ and to 0.5 percent in the case of the $K\alpha_1$, within the limits of experimental error in the case of the $K\alpha_2$ and barely outside these limits for the $K\alpha_1$. It seems justifiable to conclude that the tabulated values for uranium K x-rays are in error, and that the values obtained here may be taken as the K x-ray energies for uranium. A tempting explanation of the high values for the L x-ray energies is that the emission of the converted K electron is followed very rapidly (within electron readjustment times)

by a conversion of another gamma ray in the L shell, and that the L vacancies are filled while the K vacancy is empty, or during the process of its filling. The change in screening constant values in such a process would result in the increase of the L x-ray energies. Such an explanation demands that conversion in the L shell take place only under these conditions (otherwise the "normal" L x-rays would be seen) and, indeed, such a contingency seems unlikely in view of the numerous gamma rays capable of L conversion.

The high intensity of the source permits observation of a greater number of lines in the x-ray spectrum than in any previous sample; the high intensity of the principal lines, plus the advantage of a low atomic number carrier, permits calculation of the relative intensities of the x-ray lines with a fairly high degree of confidence in the result. Table XI presents the relative intensities of x-ray lines arising from a given x-ray vacancy, and compares these figures with those tabulated by Compton and Allison.[10] It is noteworthy that the greatest deviations occur in just those cases in which the effect of the L edges of uranium would be expected to affect Compton and Allison's data most seriously. For example, consider the $L\beta_3$ and the other x-rays arising from this type vacancy. The energy of the $L\beta_3$ line lies just above the L_3 edge of uranium, while those of the $L\gamma_2$ and $L\gamma_3$ lie just below the L_2 edge, and that of the $L\gamma_4$ lies just below the L_1 edge. As a result, one would expect the $L\beta_3$ to be absorbed somewhat more heavily than the other lines of the series and, in consequence, should be relatively reduced in data uncorrected for this effect. In comparing the Compton and Allison data with those

Table XI. Relative intensities of x-rays arising from a given L vacancy.

Vacant shell	Line	Transition	Relative intensity of x-radiation arising from this shell vacancy		
			Compton and Allison	Pa^{233}	Pa^{232}
L_1	$L\beta_4$	$L_1 - M_2$	98	135*	--
	$L\beta_3$	$L_1 - M_3$	100	100	--
	$L\gamma_2$	$L_1 - N_2$	36	55	--
	$L\gamma_3$	$L_1 - N_3$	33	(66)**	--
	$L\gamma_4$	$L_1 - O_3$	0	26	--
	$L\beta_{10}$	$L_1 - M_4$	0	7	--
	$L\beta_9$	$L_1 - M_5$	0	10	--
L_2	$L\beta_1$	$L_2 - M_4$	100	100	100
	$L\eta$	$L_2 - M_1$	2	2	--
	$L\gamma_5$	$L_2 - N_1$	0	1.5	--
	$L\gamma_1$	$L_2 - N_4$	24	36	37
	$L\gamma_6$	$L_2 - O_4$	4.4	15	15
L_3	$L\alpha_1$	$L_3 - M_5$	100	100	--
	$L\alpha_2$	$L_3 - M_4$	11	13	--
	$L\beta_6$	$L_3 - N_1$	1.6	2	--
	$L\beta_2$	$L_3 - N_5$	28	35	--
	$L\ell$	$L_3 - M_1$	3.4	--	--
	$L\beta_7$	$L_3 - O_1$	0.4	--	--
	$L\beta_5$	$L_3 - O_{4,5}$	6.4	--	--

*Doubtful through difficulty of resolution from the $L\beta_2$.

**Doubtful through poor resolution from $L\gamma_6$.

taken here, it is surprising to note that the deviation is in the opposite direction, i.e., that in the data uncorrected for self-absorption the $L\beta_3$ is more intense with respect to the other lines than in the case of the data for which this effect has presumably been corrected out. No explanation of this anomaly has been attempted, although if one bears in mind the high correction factors of relatively uncertain validity applied to the data of Compton and Allison, as well as to the data here, it becomes remarkable that any quantitative agreement on intensities is reached. The soft lines of the L series, however, do show remarkable agreement under the circumstances. The agreement between the $L\beta_1$ to $L\gamma_1$ ratios in the cases of the Pa^{233} and Pa^{232} runs is also noteworthy. If the usual values for the fluorescent yields of the L shells are applied to these data, the resulting ratios, ignoring for the moment the Coster-Kronig effect for the $L_1 - L_3M$ shift, are:

$$L_1 : L_2 : L_3 = 1.0 : 0.46 : 0.18 .$$

To have any significance in regard to L conversion processes, these data must be corrected for the L vacancies produced by L electrons filling the K vacancies produced by K conversion events. Basing these corrections on the observed intensities of the K x-rays and taking the K fluorescent yield as approximately 100 percent,[38] the L vacancy ratios are:

$$L_1 : L_2 : L_3 = 1.0 : 0.13 : 0.0$$

where the $K\alpha_1$ abundance demands that all observed L_3 vacancies arise from that source and, indeed, the agreement between the $K\alpha_1$ intensity and the L_3 vacancy intensity is well within experimental error.

Such a result brings up additional difficulties, for it implies that the Coster-Kronig coefficient for the transition $L_1 - L_3M$ is essentially zero.

The Gamma Rays.-- The gamma radiation of Pa^{233} has been investigated thoroughly by Keller and Cork[40] using the methods of beta spectroscopy. They find some 13 gamma rays varying in energy from 28.9 to 416.4 kilovolts. Many of these lines are within the range of the bent-crystal spectrometer with the improved collimation mentioned above; Table XII presents the energies and intensities of the lines which were observed, and compares the energies with the values reported by Keller and Cork. It is seen that the agreement is, in general, excellent. All the lines of energy less than 100 kilovolts reported by Keller and Cork were observed, with the possible exception of one doubtful case (the line of energy 58.1 kev) and, in addition, two more low energy gamma rays were found. It is significant that the 17.4 kev line demanded by the decay scheme proposed by Keller and Cork was not observable, although it is, of course, possible that the transition is so highly converted as to be unobservable as electromagnetic radiation.

Table XII. Energies and intensities of the gamma rays in Pa^{233} decay.

Line	Energy here	Energy (Keller)	Intensity
G_1	28.67 ± 0.02	28.9	2.5
G_2	40.47 ± 0.1	40.6	2.0
G_3	75.4 ± 0.2	75.7	55
G_4	87.0 ± 0.3	87.1	72

DISCUSSIONS AND CONCLUSIONS

One of the more disappointing aspects of the data gathered here is that they illustrate the rather poor state of knowledge of extranuclear processes, when these processes are considered in detail. It is clear from even the small number of cases cited that Kinsey's generalization on the ratios of conversion in the L shells was not justified, and that these ratios will vary with the energy and multipole order of the gamma ray converted. The Coster-Kronig transitions pose a more difficult problem; in the case of Am^{241}, Kinsey's estimate of 60 percent for the $L_1 - L_3M$ transition agrees quite well with the data and yet, in the case of Pa^{233} and Pa^{232}, this coefficient must be taken as essentially zero on the basis of the data. Such a startling change within one atomic number is not accounted for by the theory. In the case of Pb^{210}, where an upper limit of 16 percent was set to the $L_1 - L_3M$ coefficient, it is possible to account for the change, at least qualitatively, by noting that the $L_1 - L_3$ energy difference in the uranium region is large enough to permit ejection of an M_3 electron, while at bismuth the $L_1 - L_3$ difference is sufficient for ejection of an M_4 electron only. A further point in this respect is that for atomic numbers beyond 90, the $L_2 - L_3$ difference is sufficient for ejection of an M_4 electron. Inasmuch as $L_2 - L_3M$ transitions were not considered in the calculations, the rise of the coefficient for this transition would result in an apparent increase in the $L_1 - L_3M$ coefficient.

The values for relative intensities of x-rays arising from a given L vacancy are still at odds with the theory. While the values

tabulated in the text are believed to be generally better than those listed by Compton and Allison, by virtue of a correction for sample absorption, they are not sufficiently different to overcome the theoretical difficulties which these authors point out, in that wave-mechanical calculations of the relative abundances of doublets will lead to values quite different from those experimentally obtained.

The apparent anomaly in the energies of the uranium x-rays from the decay of Pa^{233} and Pa^{232} remains unexplained, since it is difficult, as noted in the text, to dismiss the effect as one of instrumental error. A most interesting experiment would be the observation of the uranium x-rays from Pu^{238} decay; if these should agree in energy with Siegbahn's values and the values obtained from Pu^{239} decay, the reality of the shift in the x-ray energies in the protactinium decay would be established beyond doubt, and a more comprehensive search for an explanation would be justifiable.

The few experiments described here actually do little more than to point up the nature of the problems to be solved in this type of experimentation and to illustrate the very great capabilities of a precision instrument in attacking problems in the nuclear field of research. A spectrometer of the type described here, used in conjunction with a high resolution beta ray spectrometer, makes it possible to assign multipole orders and parities to the nuclear levels in even such a complicated decay scheme as that of Pa^{233}; used in conjunction with an alpha ray spectrometer, it can be of tremendous aid in analyzing the decay schemes of complex alpha spectra. The extension of these techniques to decay schemes involving isomeric

transitions should hold great promise, and it is only by the use of such an instrument that a comprehensive experimental attack can be made on the problems of electron capture, for only by this means can the ratios of K electron capture to capture from the various L levels be ascertained. The introduction of coincidence methods to the present techniques of the spectrometer would call for no great amount of elaboration in apparatus and would widen even further the potentialities of the spectrometer. The proper application of bent crystal spectroscopy to the fields of nuclear research will be striking in the extension of knowledge.

ACKNOWLEDGMENTS

This research was made possible largely through the interest, encouragement, and guidance extended by Professor I. Perlman throughout the course of the experiments described here. The author recalls with pleasure the cooperation and assistance of Drs. W. W. T. Crane and R. E. Batzel in much of the chemistry involved in sample preparation, of Dr. G. D. O'Kelley and Messrs. H. P. Robinson and A. Hartzell in solving the electronic and instrumental problems, of Mrs. E. B. King and Miss H. L. Jensen in gathering the data, and of Mrs. N. B. Browne in plotting the spectra and in aiding the researcher in many respects.

This work was supported by the Atomic Energy Commission. The author worked under the auspices of the United States Air Force Institute of Technology.

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