Intensities of X-Rays and γ Rays in Am^{241} Alpha Decay*

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Electromagnetic radiation intensities from levels of Np^{237} were remeasured with NaI scintillation detectors and an argon proportional chamber. By absolute and coincidence counting the photon intensities per alpha particle are 0.376, 0.025, 0.00073, and 0.359 for the L x-rays and the 26.4-, 43.4-, and 59.6-kev gamma rays, respectively. Gamma rays with 99- and 103-kev energy were found by α-γ coincidence. The 99-kev γ ray is emitted from the 159-kev level with an intensity of 2.3×10^{-4}. The 103-kev γ ray, of intensity 1.9×10^{-4} per alpha particle, is a de-excitation of the 103-kev level. The intensity of L x-rays from levels above 59.6 kev is 0.064. Coincidence intensities of 26-kev γ rays with L x-rays from conversion of the 33.2-kev γ ray and of L x-rays from these transitions are 0.01 and 0.031 per alpha particle, respectively.

IN association with calibrations of the counting efficiencies of some sodium iodide scintillation crystals, Engelkemeir and the author have essayed absolute measurements of electromagnetic radiation intensities from the levels of Np^{237}.[1] The observed L x-ray intensities following Am^{241} alpha decay did not fit expectations derived from the decay scheme[2,3] and recent measurements of the conversion-electron spectrum.[4] Disagreement was also noted with prior direct measurements.[5,6] Extension of the measurements to alpha-particle, gamma-ray, and L x-ray coincidence and improved measurements of absolute photon intensities are reported in this paper.

PROCEDURES AND RESULTS

Preliminary values for the intensities of L x-rays, 26.4-kev and 59.6-kev gamma rays have been published.[1] Since the final results for the total L x-rays and 59.6-kev gamma ray are believed to be reliable to within about 2%, the experimental procedure will be given in detail sufficient for evaluation. Relative intensities of the l, α, β, and γ groups of L x-rays were measured with an argon proportional chamber. Absolute intensities were determined in beams collimated to enter only the central portion of the face of a scintillation crystal to avoid uncertainties in the counting efficiency and geometry introduced by the crystal edge and the distance from sample to crystal.

A. Samples

Isotopically pure Am^{241} was deposited on a 1-mil $\times\frac{3}{4}$-in. aluminum disk over an area of $\frac{1}{8}$-in. diameter by evaporation in vacuum from a hot filament. The deposit was nearly invisible, the weight of americium alone being about 0.04 μg. By precise low-geometry measurement the alpha-disintegration rate was found to be 2.80×10^5 per minute. From less precise alpha-59.6 kev gamma coincidence measurements the disintegration rate was found to be 2.78×10^5. A further check on the disintegration rate was obtained by 2π counting. The 2π-counting rate corrected for resolution loss would yield a disintegration rate agreeing with that from the low-geometry measurement if one assumed a counting yield of 51%.

For analysis of photon pulses, the sample disk was clamped in a set of aluminum rings similar to that described elsewhere.[7] Three rings were punched from 22-mil aluminum sheet with outer diameters of $1\frac{3}{4}$, $\frac{15}{16}$, and $1\frac{3}{4}$ and inner diameters of $\frac{3}{4}$, $\frac{9}{16}$, and 1 in., respectively. The rings, sample disk and an 0.8 mg/cm^2 aluminum cover foil of $1\frac{3}{4}$ inch diameter were stacked on centers in the order: $1\frac{3}{4}$ in.$\times\frac{3}{4}$ in. ring, $\frac{15}{16}$ in.$\times\frac{9}{16}$ in. ring, sample disk with active deposit facing up, cover foil, and $1\frac{3}{4}$ in.$\times$1 in. ring. The stack is held together with six No. 2 brass screws spaced near the peripheries of the large rings.

The purity with respect to other alpha and gamma emitters was confirmed by alpha-gamma coincidence and alpha-pulse analysis. For resolution of the alpha-particle spectrum the cover foil in the ring clamp described above was replaced by a 2-mil-thick electroformed screen.[7] There was no loss of sample by contact with the screen.

A larger sample (1.18×10^7 disintegrations/min) was required for some of the proportional chamber counting. This sample, previously used for conversion-electron measurements,[8] was also deposited in vacuum over an area of about $\frac{1}{8}$-inch diameter on a 0.1-mil$\times\frac{3}{4}$-inch aluminum foil. The foil was cemented to a $1\frac{3}{4}$-inch aluminum ring of $\frac{1}{16}$-inch cross section.

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.
[1] L. B. Magnusson and D. W. Engelkemeir, Bull. Am. Phys. Soc. Ser. II, 1, 171 (1956).
[2] F. Asaro and I. Perlman, Phys. Rev. 93, 1423 (1954).
[3] Jaffe, Passell, Browne, and Perlman, Phys. Rev. 97, 142 (1955).
[4] S. A. Baranov and K. N. Shlyagin, *Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, July 1–5, 1955*, Session of the Division of Physical and Mathematical Sciences (Akademiia Nauk S.S.S.R., Moscow, 1955), translated by Consultants Bureau, New York, 1955, Vol. 1, p. 183.
[5] Beling, Newton, and Rose, Phys. Rev. 86, 797 (1952).
[6] P. P. Day, Phys. Rev. 97, 689 (1955); and private communication.
[7] D. W. Engelkemeir and L. B. Magnusson, Rev. Sci. Instr. 26, 295 (1955).
[8] Freedman, Porter, and Wagner (unpublished work).

B. Collimator

The solid angle for admitting photons to the detecting crystal or proportional-counter chamber was defined by a drilled, cylindrical aperture (0.3563-inch diameter) in a lead plate of thickness 3.0 g/cm^2 sufficient to be opaque to the 59.6-kev gamma ray (Fig. 1). The aperture-to-sample distance was fixed by a simple aluminum box, open on two sides, which supported both collimator and sample. The removable base of the box was machined with a $1\frac{3}{4}$-inch diameter inset to center the rings holding the small sample foil. Variations of the sample mounting will be described in following sections. The active side of the small sample foil either faced away from the detector or was covered with 0.8-mg/cm^2 Al foil to permit depth gauge measurements through the collimator hole without contacting the active deposit.

C. Proportional Spectrometry

The proportional chamber is a conventional brass cylinder of $3\frac{5}{8}$-inch inside diameter. Radiations are admitted through a 1-inch hole covered by a 40 mg/cm^2 beryllium window sealed in with an O ring. Transmission by the beryllium was measured with beams of x-rays from Fe^{55} (5.9 kev) and Se^{75} (10.5 kev). The mass absorption coefficients found for these rays were 2.8 and 0.82 cm^2/g, respectively. The chamber gas is 90% argon, 10% methane vented through a glass frit dipping into the mercury in one arm of a U tube. The other arm of the tube terminates in a closed glass bulb which was filled with air to give a pressure of 762 mm Hg at 23° at the frit with gas flowing. The U tube maintains constant density in the chamber gas. The sample collimator is held in a shelf support below the beryllium window. For measurements of the large sample the aluminum base of the collimator was replaced by a $\frac{1}{16}$-inch thick copper sheet having a $\frac{1}{4}$-inch hole coaxial with the collimator hole. The sample ring was centered over the $\frac{1}{4}$-inch hole.

A typical pulse spectrum for the sample of 1.18×10^7 disintegrations/min as recorded by a 20-channel analyzer is shown in Fig. 2. No Pb fluorescent radiation appears in the spectrum indicating that the scattered contribution from the wall of the collimator hole is negligible. The detection of Np M x-rays (about 3 kev) is supporting evidence for a relatively small amount of absorption in the system. The L x-ray spectrum is resolved as α, β, and γ groups with the l line contributing to a shoulder on the low-energy side of the α group. The L x-ray region was counted repeatedly for both Am samples to obtain the relative intensities of the groups. The results for the L x-ray groups agreed well for the two samples. The M intensity from the larger sample, however, was about half that from the other, indicating self absorption. From the appearance of the large sample it seemed likely that a small amount of an impurity was present, perhaps tantalum from the evaporation filament. Intensities relative to background were not sufficient from the smaller sample to give good accuracy for the $L\gamma$, 26.4-kev, and 59.6-kev rays. The peak intensities are summarized in Table I. Overlapping portions of the peaks were unfolded by fitting normal frequency distributions. The observed intensities were corrected for 5% loss by escape of argon K x-rays as measured with a beam of Mo $K\alpha$ fluorescent x-rays. The error in the observed intensities of the peaks with energies greater than the l line is estimated to be less than 5%.

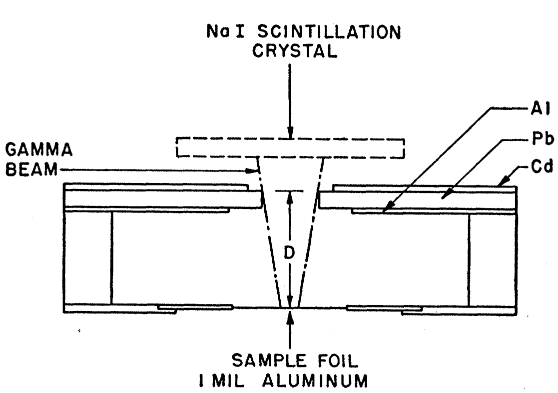

FIG. 1. Section of the photon collimator. The beam-hole diameter is 0.3563 inch.

TABLE I. Intensities of the Np L x-rays and gamma rays from the argon proportional chamber.

Group	Intensity[a] counts/min/α	Transmission[b]	Counting efficiency[c]	Corrected intensity photons/α
L l x-rays	0.0031	0.959	0.392	0.0082
α	0.0363	0.970	0.272	0.1375
β	0.0268	0.982	0.1454	0.1878
γ	0.00441	0.986	0.0882	0.0507
			Total	=0.384
26.4-kev γ-rays	0.00112	0.995	0.0460	0.025
59.6 kev	0.00121	1.00	0.00370	0.33

[a] The tabulated intensities are the observed peak intensities corrected for 5% argon escape and divided by the geometry and alpha emission rate. The 59.6-kev full-energy peak was corrected for 17.5% loss by the wall effect [see West, Dawson, and Mandleberg, Phil. Mag. **43**, 875 (1952).]
[b] The transmission is that over-all for 0.8-mg/cm^2 Al cover foil, air, and window.
[c] See text.

The counting efficiencies in column 4 of Table I were derived from a plot of mass absorption coefficients in argon given by Colvert,[9] Allen,[10] Victoreen,[11] and White[12] and the relative intensities for individual L lines from the data of Day.[6] Photoelectric events in argon only were assumed to contribute to the observed peaks. Scattering corrections to the total absorption coefficients were interpolated from the data of White.[12]

[9] W. W. Colvert, Phys. Rev. **36**, 1619 (1930).
[10] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935).
[11] J. A. Victoreen, J. Appl. Phys. **20**, 1141 (1949).
[12] G. R. White, National Bureau of Standards Report 1003, May, 1952 (unpublished).

Day has reported relative intensities of the Np L x-rays at high resolution with a bent-topaz crystal spectrometer. Recently Day has determined the topaz reflectivity more accurately and has recalculated the relative intensities of the Np L x-rays. The newer values are given in column 4 of Table II. In estimating the sample self-absorption, Day used the theoretical absorption coefficients for lead, thorium, and uranium given by White.[12] These theoretical values are about 30% smaller than older experimental values. A few measurements of the attenuation of the neptunium x-ray groups by lead absorbers indicated that the older data is more accurate in the region of energy less than the americium L_{III} edge. Absorption coefficients for americium were accordingly extrapolated from data given by Allen[13] for tungsten, platinum, gold, and lead and by Stephenson for uranium.[14] The absorption coefficients increase approximately as Z^3. Column 5 of Table II gives the relative intensities calculated from Day's data by the author. For energies greater than the americium L_{III} edge, the use of Allen's data may give absorption coefficients which are too large. Lead absorption coefficients measured for 22 kev (Ag^{109}) and 32 kev (Ba^{137}) K x-rays were about 15% smaller than interpolated from Allen's values. The relative intensities of the $L\gamma$ x-rays and the gamma rays in column 5 of Table II are expected to be too high. The self-absorption correction is obviously a large source of error in the absolute intensities which may be derived from Day's data but the relative intensities of lines within the narrow energy range included in a group peak in the proportional spectrum are little effected by change in the correction. The group counting efficiencies in column 4 of Table I were calculated by summing the products of the counting efficiencies and relative intensities of the individual lines within each group. The η and γ_5 lines were assumed from the method of resolution to contribute equally to adjacent groups as indicated in Table II. The total intensity of L x-rays, 0.384 per disintegration, agrees well with the more accurate value of 0.376 from scintillation pulse spectra (Sec. D). The agreement supports the belief that the relative intensities of the α, β, and γ groups are known to within about 5%. The intensity of the 59.6-kev gamma ray from the proportional data (Table I) is about 10% lower than that from the scintillation data. The error is probably in the argon cross section and the escape effect,[15] i.e., the number of photoelectrons which enter the counter wall with more than about 3-kev residual energy. The 26.4-kev gamma-ray intensity from the proportional data should be reliable to better than 10%. The exact agreement between the recalculated value of Day and that of Beling *et al.*,[5] (Table II) for the intensity of the 26.4-kev gamma ray is fortuitous. The extrapolated value of the americium absorption coefficient, 73 cm²/g at 26.4 kev, could be as much as 15% too large. The self-absorption correction factor for Day's sample was 2.3 for the 26.4-kev gamma ray. The values of Beling *et al.* are consistently about 12% larger than those of the present work.

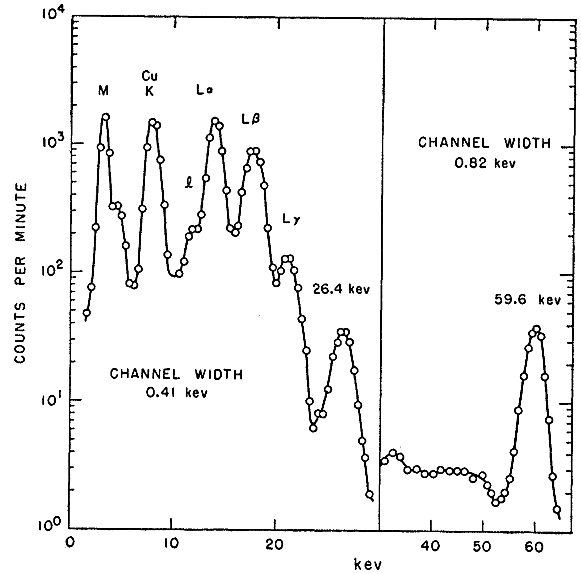

FIG. 2. A scan of the pulse spectrum from the argon proportional chamber. The sample was 1.18×10^7 disintegrations/min of Am^{241} mounted in the collimator. The distance from the top of the collimator hole to the sample was 0.686 inch.

D. Scintillation Spectrometry

The collimator and detector were aligned on a horizontal axis with a fixed mount for the collimator and a roller track for the detector unit. The detecting apparatus was shielded by 6 inches of steel. No scattering around the collimator could be detected with the beam hole closed by a $\frac{3}{4}$-inch diameter lead shutter. To insure that the apparent photon intensities were not dependent on the detector, three types of crystals were used. One was a commercial model (National Radiac) of 2.2-mm thickness and $1\frac{1}{2}$-inch diameter with a 0.5-mil aluminum window. The expected transmission of the window was confirmed approximately by comparing the intensities of the neptunium L x-rays in beams directed at the crystal face at angles of 40 and 90 degrees. Another, assembled in this laboratory, was $\frac{1}{8}$-inch thick and $1\frac{1}{4}$-inch in diameter with an 18-mil beryllium window. The third, assembled by D. W. Engelkemeir of this laboratory, was a cleaved rectangular section of crystal, $\frac{1}{8}$-inch thick with 0.84-inch and 0.86-inch sides. The aluminum window, 3.5 mg/cm², was weighed and measured for transmission before assembly with K x-rays from Fe^{55} and Se^{75}.

[13] S. J. M. Allen, Phys. Rev. **24**, 1 (1924); **27**, 266 (1926); **28**, 907 (1926); *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Company, Cleveland, 1956) thirty-eighth edition.
[14] R. J. Stephenson, Phys. Rev. **43**, 527 (1933).
[15] West, Dawson, and Mandleberg, Phil. Mag. **43**, 875 (1952).

TABLE II. Summary of the intensities of the Np L x-rays and gamma rays.

Group	Line	Energy[a] kev	Relative intensity[b] Bent crystal spect. Day[c]	Day[d]	Proportional spect. Beling *et al.*[e]	Present[f]	Absolute intensity[g] photon/alpha	
Ll	l	11.89	0.83	1.32	...	2.2_4	0.008	0.008
	α_2	13.78	1.89	2.61				0.013
	α_1	13.96	17.2	24.4				0.119
	$\frac{1}{2}\eta$	15.88	0.39	0.49				0.0024
$L\alpha$			19.5	27.5	42.0	37.5	0.135	
	$\frac{1}{2}\eta$	15.88	0.39	0.49				0.0018
	β_6	16.14	0.38	0.50				0.0018
	β_2	16.86	4.60	6.03				0.022
	β_4	17.08	3.04	3.90				0.015
	β_5	17.52	0.74	0.99				0.0037
	β_1	17.76	26.1	34.2				0.127
	β_3	18.00	2.41	3.17				0.012
	$\frac{1}{2}\gamma_5$	20.12	0.11	0.15				0.0006
$L\beta$			37.8	49.5	66.0	51.2	0.184	
	$\frac{1}{2}\gamma_5$	20.12	0.10	0.13				0.0005
	γ_1	20.80	6.42	7.92				0.031
	γ_2	21.11	0.71	0.86				0.0033
	γ_3	21.34	0.81	0.96				0.0037
	γ_6	21.48	1.73	2.01				0.0078
	γ_4	22.20	0.78	0.92				0.0036
$L\gamma$			10.6	12.8	17.7	13.8	0.050	
		26.36	5.93	7.5	7.5	7.0	0.025	
		33.20	0.40	0.40	...	...	0.0011	
		43.46	0.22	0.26	...	0.20[h]	0.0007	
		59.56	100	100	100	100[h]	0.359	
		99.0 }	0.10[h]		...	0.064[h]	0.00023	
		103.0 }				0.053[h]	0.00019	

[a] Reference 6.
[b] Photons per 100 59.6-kev photons.
[c] Relative intensities of reference 6 recalculated by Day with the topaz reflectivity dependence equal to $1/E^{1.35}$.
[d] Relative intensities of column 4 recalculated with mass absorption coefficients for the sample self-absorption extrapolated from the data in references 13 and 14.
[e] Reference 5.
[f] The preliminary intensities for the L x-ray groups in column 5, Table I, were normalized to the absolute intensities of 0.376 L x-ray/α and 0.359 59.6-kev gamma-ray/α determined by NaI crystal spectrometry (Sec. D).
[g] The relative group intensities of column 7 were distributed by the relative line intensities within each group as given by column 5 and normalized to give the absolute total of 0.376 L x-ray/α.
[h] These values were measured by scintillation spectrometry.

The principal neptunium radiations produce pulse spectra with two peaks which are roughly equal in intensity. The lower energy peak is a composite of the L x-rays with the 26.4-kev gamma ray and escape peak from the 59.6-kev gamma ray appearing as a prominent shoulder. The full width at half-height of the 59.6-kev peak was typically about 11 kev in the present work. The ratio of the peak height to the valley on the low-energy side was 100. The intensity ratio of escape peak to full-energy peak for each of the detectors was measured in the pulse spectra from 59.6-kev radiation transmitted by a lead absorber (130 mg/cm^2) placed between the collimator and detector. Of the total pulses, $(11.0\pm0.2)\%$ appeared in the escape peak. About 11.5% would be expected from an interpolation of the data calculated by Axel.[16]

From the spectrum transmitted by a tantalum absorber (17 mg/cm^2) the intensity of the 26.4-kev gamma ray was estimated to be 0.025 ± 0.002 per alpha particle. The escape-peak contribution from the 59.6-kev radiation was determined by adding the lead absorber (130 mg/cm^2) and multiplying the escape-peak intensity by the intensity ratio of the 59.6-kev peak without and with the lead absorber.

The peak intensity producing the shoulder at 26 kev in the difference spectrum was obtained by trial fitting with a normal frequency distribution. The full width at half-height (7.1 kev) of the trial peak was interpolated from widths of other low-energy peaks (22-kev Ag^{109} and 32-kev Ba^{137} x-rays). The interpolated value[10] for the absorption of 26-kev radiation by tantalum and the absorber thickness were substantiated by measuring the absorption of 22-kev Ag^{109} x-rays with a series of tantalum absorbers.

Results of intensity measurements of the L x-rays and 59.6-kev gamma ray are given in Table III. Some variations in the method of measurement were introduced to assess the contribution of scattered radiation to the direct beam. The small diameter of the sample disk, $\frac{3}{4}$-inch, required that the active deposit be relatively close to scattering material, namely the aluminum supporting rings. The wall of the collimator hole was also considered to be a possible source of detectable

[16] P. Axel, Rev. Sci. Instr. 25, 391 (1954).

scattered radiation. The ratio of photons entering the wall to those traversing the hole was about 0.3. In the first five measurements in Table III the detector-to-collimator distance was increased stepwise close to the maximum permitted by the diameter of the crystal. No significant changes in the numbers of photons intercepted by the crystal were observed. It was concluded that the detection of scattered and fluorescent radiation from the wall of the collimator hole was negligible. The conclusion was confirmed by later measurements with the proportional chamber reported above in Sec. C. This fortunate circumstance probably resulted from the microscale roughness of the wall which shades the detector. The scattered contribution from material in the plane of the sample disk and more than about $\frac{3}{4}$-inch away from the active deposit was evidently also negligible. The scattering from the aluminum near the sample was estimated from the cross sections tabulated by White.[12] For the 59.6-kev radiation, the scattered contribution to the direct beam was of the order of 2%, of which 0.5% came from the sample disk. For the L x-rays the total scattered contribution of only 0.1% came from the sample disk.

That the scattered contributions are relatively small is supported by the last ten measurements in Table III in which the mass of scattering material near the sample was substantially reduced. The sample disk was centered on an aluminum ring, 40 mg/cm^2 thick, and secured with a small amount of Apiezon wax. The supporting ring was $1\frac{3}{4}$-inch in diameter with a $\frac{9}{16}$-inch hole. With the detector-to-collimator distance in the range of $\frac{1}{4}$- to $1\frac{1}{8}$-inch the counting rate of 59.6-kev photons was essentially constant within the reliable (0.9) error of 0.6%. The apparent intensity of 59.6-kev photons was about 1% less than calculated from the measurements using the heavier supporting rings (fourth column, Table III). With the detector-to-collimator distance increased to $1\frac{1}{2}$-inch, the intensities decrease as the fringe of the beam misses the crystal.

The two measurements with detector C were similar except that the active side of the sample foil was reversed between the two and not covered for the second measurement. The purpose of the experiment was to see if the recoil following alpha emission was altering the measured geometry. The effect was not detectable.

To calculate the apparent L x-ray intensities given in the last column of Table III the observed intensities of low energy pulses were corrected for the contributions from the 26.4-kev gamma ray and the escape peak from the 59.6-kev gamma ray, the sample spread,[17] and the transmission by the detector window, air, and aluminum foil. For the transmission corrections, the L x-ray spectrum was assumed to consist of radiations in the relative abundances determined by the proportional spectrometer measurements of Sec. C. The apparent L x-ray intensity with detector B was about 3% lower than obtained with the other two detectors. The window transmission for detector B was not tested so the results indicate that the beryllium is not pure which agrees with some earlier rough transmission measurements. From the indicated transmission of 95% for the L x-rays it may be safely assumed that the impurity is not important in the transmission of the 59.6-kev gamma ray. The average of the nine results for the L x-ray intensity with detectors A and C is 0.376 photon per alpha particle. The absolute intensities of individual lines given in column 9 of Table II were calculated from this value combined with the relative group intensities from the proportional spectrometer and the relative line intensities (columns 7 and 5, Table II).

The upper edge of the collimator hole (Fig. 1) is not an infinitely sharp cutoff for 59.6-kev photons. The transmission, I/I_0, of the edge for a point source on the

[17] A. H. Jaffey, Rev. Sci. Instr. 25, 349 (1954).

TABLE III. Intensities of the Np L x-rays and the 59.6 kev gamma ray measured with sodium iodide detectors.

Detector[a]	Detector-to-collimator distance (in.)	Sample-to-collimator distance[b] (in.)	Apparent 59.6-kev intensity[c] (photons/α)	Apparent L x-ray intensity[d] (photons/α)
Active side of sample foil mounted on 22-mil Al rings, covered by 0.8 mg/cm^2 Al and facing detector.				
A	$\frac{5}{16}$	0.748	0.367	0.377
	$\frac{5}{8}$		0.365	0.379
	1		0.366	0.376
	$1\frac{1}{8}$		0.364	0.379
	$1\frac{1}{4}$		0.365	0.377
	$\frac{3}{8}$	0.720	0.366	0.374
	$\frac{3}{8}$	0.741	0.367	0.376
B	$\frac{1}{2}$	0.748	0.365	0.368
	$\frac{3}{4}$		0.366	0.367
	1		0.363	0.367
C	$\frac{3}{8}$	0.752	0.362	0.372
Active side of sample foil, with no cover, facing away from detector.				
C	$\frac{3}{8}$	0.748	0.366	0.373
Active side of sample foil, mounted on 6-mil Al ring, facing away from detector.				
B	$\frac{1}{8}$	0.776	0.365	
	$\frac{5}{16}$		0.363	0.366
	$\frac{1}{2}$		0.362	
	$\frac{5}{8}$		0.362	
	$\frac{3}{4}$		0.361	
	$\frac{7}{8}$		0.360	
	1		0.360	
	$1\frac{1}{8}$		0.359	0.365
	$1\frac{1}{4}$		0.356	0.366
	$1\frac{1}{2}$		0.338	0.355

[a] The NaI detectors were as follows: A, 2.2 mm thick $\times 1\frac{1}{2}$ in. diam crystal with 0.5 mil Al window; B, $\frac{1}{8}$ in. thick $\times 1\frac{1}{4}$ in. diam crystal with 18 mil Be window; C, $\frac{1}{8}$ in. thick $\times$0.84 in. $\times$0.86 in. cleaved crystal with 3.5 mg/cm^2 Al window.

[b] The sample-to-collimator distance and the collimator hole diameter (0.3563 inch) define the solid angle of the beam from a point source.

[c] The 59.6-kev photon intensity was calculated from the observed counting rate of the full-energy peak, the disintegration rate of the sample (2.80×10^5 disintegrations/min) and the measured geometry with corrections for the escape peak, sample spread, transmission by the edge of the collimator hole, and transmissions by the detector window and Al foil. The measured escape peak intensities for detectors A, B, and C were 11.2, 10.9, and 11.1%, respectively, of the total 59.6-kev gamma-ray pulses.

[d] The L x-ray intensity was calculated from the total counting rate of low-energy pulses and the measured geometry with corrections for the contributions from the 59.6-kev escape peak and 26.4-kev pulses (0.025 per alpha), sample spread, and transmissions by the detector window, air, and Al foil.

axis of the hole is

$$I/I_0=\exp[\mu(R/\sin\theta-D/\cos\theta)], \quad (1)$$

where μ is the linear absorption coefficient, R is the hole radius, θ is the angle from the axis to the photon path, and D is the distance along the axis from source to the top of the collimator. The effective geometry, g, for the detection of photons passing through the edge is

$$g=\frac{1}{2}\int(I/I_0)\sin\theta d\theta. \quad (2)$$

The detection of scattered photons is neglected in Eq. (2); however, about 80% of the attenuation in lead is photoelectric. To compensate for what would be a relatively small error, an empirical μ, 138 inch^{-1}, was determined by measuring the intensity of the *ca* 59-kev radiation as a function of the amount of lead absorber placed between collimator and detector. The geometry for detecting scattered radiation from the absorber was similar to that for radiation scattered in the edge of the hole.

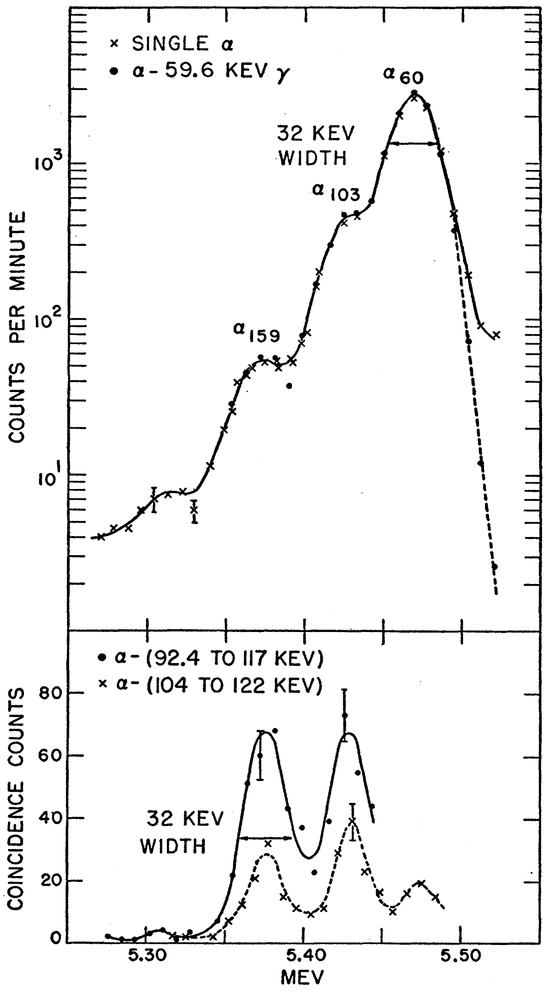

FIG. 3. Alpha-particle pulse spectra recorded by 20-channel analyzer from 2.80×10^5 disintegrations/min of Am^{241} collimated with 2-mil Lektromesh screen. The single alpha pulse spectrum in the upper section of the figure is a composite from 5-minute runs. The counting time for the α—59.6-kev γ coincidence was 23 minutes, the single-channel rate of the 59.6-kev γ being 14 740 counts/min. The counting times for the solid points and the crosses of the lower section of the figure were 1000 and 961 minutes, respectively.

Equation (2) must be summed numerically. For the conditions of the present measurements an approximate solution proved to be sufficiently accurate. The transmission decreases exponentially with θ by virtue of the fact that the photon path length, the factor in parentheses of Eq. (1), is very nearly a linear function of θ. Since $\sin\theta$ is essentially constant, the geometry is given approximately by

$$g=\tfrac{1}{2}\sin^2\theta/\mu D\theta. \quad (3)$$

The magnitude of the edge correction in the data of Table III is 2%.

From the work of Bell *et al.*[18] and McGowan[19] on the relative intensities of full-energy peaks and total pulse spectra, it appears that all the pulses from 59.6-kev radiation, except those resulting from the escape of iodine K x-rays, should be included in the full-energy peak under the conditions of the present measurements. The range of summation of full energy pulses included about half of the pulses from the 43.5-kev gamma first observed by Day;[6] however this error is less than 0.1% (see Sec. E). The sum of escape and full-energy intensities for the 59.6-kev gamma ray with the appropriate transmission corrections yields the apparent photon intensities in column 4, Table III. The value indicated by the last series of measurements in the table corrected for 0.5% scattered contribution from the sample disk gives the final result of 0.359 photon per alpha particle.

E. Coincidence Spectrometry

Further information on the neptunium transition intensities was secured from double-coincidence measurements with sodium iodide crystals, the proportional chamber, and an ion chamber as detectors. Pulses were sorted with single- and 20-channel instruments. When two scintillation detectors were being used, the Am^{241} sample proved to be a convenient source of triggering pulses for the measurement of resolving time. With both detectors close to the sample, the iodine K x-ray, which may escape after the photoelectric absorption of a 59.6-kev gamma ray in either crystal, can be caught by the other crystal. If the discriminators of both analyzers are adjusted to accept pulses in a broad band about 30 kev, the coincident events are detected with high efficiency.

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[18] Bell, Heath, and Davis, Oak Ridge National Laboratory Report ORNL-1415, September 20, 1952 (unpublished).
[19] F. K. McGowan, Phys. Rev. 93, 163 (1954).

1. 43.4-Kev Gamma Ray

The intensity was determined from the spectrum coincident with the 59.6-kev gamma ray by using sodium iodide detectors. The 69-mμsec half-life[20] of the 59.6-kev level presents a minor experimental difficulty. As the resolving time is lengthened to catch most of the pulses following the delayed state, the chance rates of 59.6-kev full-energy and escape-peak pulses increase in the spectrum of radiations preceding the delayed state. With a resolving time of 140 mμsec and a delay of 30 mμsec for the 43.4-kev pulses, the full-energy peak of the latter was resolved in a 16-hour measurement. With corrections for resolving time, escape peak loss,[16] and geometry as determined from the 59.6-kev peak intensity in each detector, the intensity of the 43.4-kev gamma ray is $(7.3\pm0.7)\times10^{-4}$ per alpha particle.

2. 99- and 103-kev Gamma Rays

Radiations with energies of 99 (or 103), 113, 128, 159 (or 168) 210, 270, 328, and 370 kev have been reported by Jaffe *et al.*[3] and by Day.[6] A more detailed investigation of the 100-kev region by α-γ coincidence appeared feasible. The alpha-pulse analyzer has been described elsewhere.[7] For detection of the gamma rays the $\frac{1}{8}$-inch thick NaI crystal with associated photomultiplier circuits was mounted below a beryllium window in the base of the ion chamber. Other than the ion chamber, there was no shielding for the gamma detector and the background was relatively high. The pulse rate in the range 92-114 kev in one of the measurements, for example, was 13 counts/min over a background of 61 counts/min. The singles rate so determined was intended only to be a rough confirmation of rates calculated from the coincidence data.

Alpha-particle pulse spectra are shown at the top of Fig. 3. That the resolution was not quite so good as obtained with a smaller sample (30-kev width at half-height) was caused by the high counting rate (12 400 counts/min with screen collimator). The alpha particles emitted in low intensity in decay to the 33-kev and ground states are not resolved. The particles emitted in decays to the 59.6-, 103.0-, and 158.6-kev levels are clearly evident. A low-energy group is suggested at about 5.31 Mev in agreement with a recent result from magnetic spectrometry.[21] Approximate group intensities from fitting normal frequency distributions to the pulse spectrum from a smaller sample are listed in Table IV showing reasonable agreement with magnetic spectrometer values. Analyzer performance for coincident events, $\alpha-59.6$ kev γ, is indicated by the solid points of Fig. 3. The incidence rate in each channel has been multiplied by the disintegration rate and the fraction (0.994) of the decays populating the 59.6-kev and higher levels and divided by the gamma rate. The rates derived in this manner should match the single rates if there is no cross-over of the 59.6-kev level. Actually the derived rates are generally slightly higher than the singles rates. The effect is caused by a small distortion of the alpha-pulse spectrum by the addition of conversion-electron ionization.[7] The approximate constancy of the relative heights of the groups in the singles and coincidence spectra verifies that there is little cross-over of the 59.6-kev level.

Alpha-pulse spectra coincident with selected gamma energy bands in the region of 100 kev are given at the bottom of Fig. 3. The bias scale and band width of the single-channel gamma analyzer were calibrated with the 59.6-kev peak with small corrections for the nonlinear pulse height of the $\frac{1}{8}$-inch crystal. The possibility of simultaneous detection of lower energy radiation and 59.6-kev gamma was eliminated by covering the gamma detector with copper and silver absorbers. To maintain reasonable counting rates in the 100-kev region, the amount of absorber was relatively small; the pulse-height discrimination being sufficiently good to prevent interference from the 59.6-kev gamma rays. The subtracter bias of the alpha-pulse amplifier and the analyzer channels were calibrated by the position of the main alpha peak, 5.480 Mev minus an estimated pulse-height loss in the collimator screen of 10 kev.[7] Calibrations were run before and after each coincidence run. No detectable drift, i.e., less than 4 kev, occurred through the series of measurements which extended over a period of 40 hours. Slightly different segments of the coincident alpha-pulse spectrum were presented to the twenty-channel analyzer in the two runs. The contribution from chance coincidence and α–59.6-kev γ coincidence is shown by the last three points of the dashed line spectrum of Fig. 3. The chance and 59.6-kev γ coincidences for the solid line spectrum were recorded in an overcount register. The chance and 59.6-kev γ contribution is clearly negligible in the region below 5.45 Mev. The alpha groups which populate the 158.6- and 103.0-kev levels are prominent in both coincidence spectra. The approximately equal intensities of the two groups compared to the relative intensities of these groups in the singles spectrum shows that photons of about 100 kev are emitted from each

TABLE IV. Alpha-particle intensities (percent).

Np^{237} level kev	Asaro and Perlman[a]	Goldin *et al.*[b]	Pulse analysis
0	0.3	0.39	...
33.2	0.2	0.24	...
59.6	84.0	85.0	84.6
103.0	14.0	12.84	13.5
158.6	1.4	1.66	1.3

[a] See reference 2.
[b] See reference 21.

[20] D. W. Engelkemeir (unpublished data).

[21] Goldin, Tretyakov, and Novikova, *Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy July 1–5, 1955*, Session of the Division of Physical and Mathematical Sciences (Akademiia Nauk S.S.S.R., Moscow, 1955), translated by Consultants Bureau, New York, 1955, Vol. 1, p. 249.

TABLE V. Efficiency factors for detection of 99- and 103-kev gamma rays and $K\alpha_1$ and $K\alpha_2$ x-rays with $\frac{1}{8}$-inch thick, 1.2-inch diam NaI crystal.

Detector band (kev)		104–122				92.4–117		
α singles, $N\epsilon_\alpha$ (counts/min)		12 400				12 400		
Coincident α	α_{103}		α_{159}		α_{103}		α_{159}	
Coincidence, counts/min$\times 10^2$	13 ± 1		10 ± 1		26.4 ± 1.6		28.8 ± 1.6	
Coincident photons (kev)	103	99	97	101	103	99	97	101
Copper transmission[a]	0.97	0.97	0.97	0.97	0.97	0.97	0.97	0.97
Silver transmission[a]	0.91	0.90	0.89	0.91	0.83	0.81	0.80	0.82
Effective geometry[b]	0.170	0.171	0.171	0.171	0.170	0.171	0.171	0.171
1-I escape peak[c]	0.96	0.96	0.96	0.96	0.96	0.96	0.96	0.96
Counting efficiency[a,d]	0.86	0.90	0.92	0.88	0.86	0.90	0.92	0.88
Fraction of peak in band	0.447	0.250	0.159	0.343	0.998	0.807	0.712	0.862
Net efficiency	0.0554	0.0323	0.0358		0.113	0.0937	0.0930	
$(\gamma/\alpha)\times 10^4$	1.9	2.3	2.0		1.9	2.3	2.3	

[a] Actual absorber or crystal thickness multiplied by 1.2 to correct for poor geometry.
[b] Apparent solid angle calculated from 59.6-kev gamma-ray intensity.
[c] See reference 16.
[d] Calculated from photoelectric absorption coefficients of NaI from reference 12.

of the two levels. Previous measurements of conversion electrons have led to the assignment of a 99-kev transition from the 158.6-kev level.[3] Measurements of the unconverted gamma radiation have consistently yielded energies somewhat greater than 99 kev. Day was prompted to assign the gamma radiation to a transition from the 103.0-kev level to ground.[6] In apparent contradiction to Day's assignment is the reported coincidence of about 100-kev radiation with the 59.6-kev photon.[3] These interpretations are reconciled by the recognition of at least two transitions. One is certainly the emission of 103 kev in decay to the ground state (K conversion of the 103-kev level cannot occur). Possibilities for the other are emission of a 99-kev gamma ray from the 158.6-kev level and K x-rays (α_1, 101 kev; α_2, 97 kev) following conversion of energy between the 158.6 and 33.2 kev or ground levels.

Knowing the energy, one can calculate the absolute intensity of the 103-kev gamma ray from the equation

$$G=N\gamma\epsilon_\gamma\epsilon_\alpha, \tag{4}$$

where G is the coincidence rate, N is the disintegration rate, γ is the number of photons per disintegration, and ϵ subscript is the net counting efficiency. The factors of ϵ_γ and the counting rates are detailed in Table V. The least reliable is that in the third row from the bottom which is the fraction of the full-energy pulses included in the bias band of the single-channel analyzer. To calculate the fraction, the pulse heights of the 103-kev gamma ray were assumed to give a normal frequency distribution with a standard deviation of 7.4 kev. The peak width for photons of about 100 kev was extrapolated from widths observed for the 59.6-kev gamma ray and the 88-kev gamma ray from Ag^{109}. Both coincidence runs yielded 1.9×10^{-4} for the intensity of the 103-kev gamma ray.

Treatment of the data for transitions from the 158.6-kev level is less straightforward. The coincidence rate is given by

$$G=N\epsilon_\alpha(b_\alpha b_{99}\epsilon_{99}+b_\alpha b_{55}b_{103}\epsilon_{103}), \tag{5}$$

where b_α is the alpha intensity (0.014) populating the 158.6-kev level, b_{99} is the fraction emitting 99-kev gamma rays or K x-rays, b_{55} is the fraction which decays to the 103-kev level, b_{103} is the fraction of the 103-kev level emitting 103-kev gamma rays, and ϵ is the counting efficiency. From conversion-electron data[4,8] the factor b_{55} is estimated to be about 0.8. The data for the two runs (Table V) are in good agreement for the absolute intensity of the 99-kev gamma ray, $b_\alpha b_{99}$ from Eq. (4).

The third, fourth, seventh, and eighth columns of numbers in Table V give the factors for computing the net counting efficiency for K x-rays. The efficiency was derived by assuming an intensity ratio of 2 for the α_1 and α_2 lines. The two runs do not agree well for K conversion. Further evidence indicating that the K x-ray intensity must be less than about 10^{-5} per disintegration was obtained by measuring the radiation of about 100-kev energy coincident with the 59.6-kev gamma ray. The absolute intensity calculated from the observed peak was 2.4×10^{-4} per alpha particle in good agreement with the intensity derived above for the 99-kev gamma ray. Radiation of this energy and intensity cannot be emitted from any level above the 158.6-kev level for it would have been revealed by the coincident alpha spectrum. The energy difference between the 59.6- and 158.6-kev levels is not enough for K conversion so the radiation coincident with the 59.6-kev gamma ray must be the 99.0-kev gamma ray.

The radiation of about 128-kev energy reported previously[3,6] with an intensity of 5×10^{-5} is not coincident with the 59.6-kev gamma ray, supporting the previous assignment to a transition from the 158.6-kev level to the 33.2-kev level. The limit on the K x-ray intensity characterizes this 125.4-kev gamma ray as $E1$, the assignment being consistent with level spins and parities as discussed by Jaffe *et al.*[3]

TABLE VI. Intensities of the Np L x-rays coincident with the 59.6-kev gamma ray.

Group	Singles[a] counts/min	Coincidence[a,b] counts/min	Counting efficiency	Intensity photon/alpha Proportional	Scintillation
l	...	...	...	(0.0015)	
α	1277	14.6	0.0338	0.0261	
β	939	8.83	0.0182	0.0294	
γ	153	1.13	0.0109	0.0063	
ΣL				0.063	0.064

[a] Corrected for 5% argon escape peak.
[b] The intensity of 59.6-kev gamma rays in the single-channel gate was 16 500 counts/min. The coincidence rate was corrected for chance coincidence.

3. L X-Rays

The L x-rays coincident with the 59.6-kev gamma ray were measured with both the proportional chamber and a scintillation detector. The resolving time, τ, with the gas detector was 4 μsec and the geometry for L x-rays was about 10%. Counting efficiencies for the L x-ray groups were calculated from the observed peak intensities in the singles spectrum and the absolute intensities from the collimated-beam measurements (Table II). The graphical resolution of the l line was not repeated in these measurements but the shoulder on the low-energy side of the $L\alpha$ group appeared and was assumed to have the same intensity relative to the α group as in the collimator measurements. The data for the coincidence spectrum are given in Table VI. The ratio of the $L\alpha$ to $L\beta$ groups was clearly larger than in the singles spectrum. The ratio of $L\beta$ to $L\gamma$ appeared somewhat larger although the difference is not beyond experimental error.

The absolute intensity of L x-rays was measured with the sample in the collimator (Sec. B), the detectors being the 2-mm sodium iodide crystal for L x-rays and the $\frac{1}{8}$-inch crystal for the 59.6-kev gamma rays. That no radiations of energy greater than about 3 kev are in prompt coincidence with the 59.6-kev gamma ray was evident from delay-line curves. There appears to be no published report of this evidence supporting the accepted decay scheme. The resolving time, 2τ, was 400 mμsec with 180 mμsec delay of the L x-ray pulse to maximize the coincidence rate. Transmission corrections were derived from the relative intensities from the proportional chamber (Table VI). With corrections for the loss of delayed coincidences (2.2%) and the counting efficiency, the number of L x-rays preceding the 59.6-kev level is 0.064 per alpha particle. Several confirmatory measurements with different resolving and delay times agreed within 3%. Beling *et al.*[5] have reported that (6±0.8)% of the total L x-rays are coincident with the 59.6-kev gamma ray in agreement with the present value of (6.1±0.2)%. The coincidence rate of 26.4-kev gamma rays with L x-rays is the sum of the prompt and delayed rates,

$$G_p = N\gamma_{26}L_{33}\epsilon_A\epsilon_B \quad (6)$$

and

$$G_d = N\gamma_{26}L_p\epsilon_A\epsilon_B(1-e^{-\lambda t}), \quad (7)$$

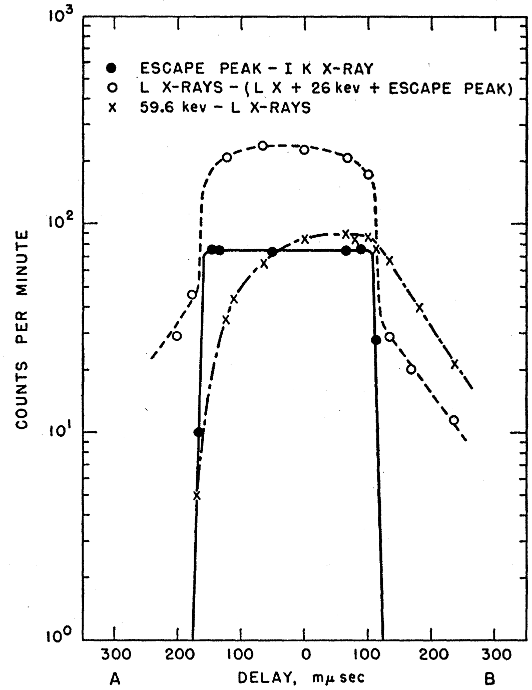

FIG. 4. Coincidence-counting rates as functions of pulse delay times. Detector A was a $\frac{1}{2}$-inch thick×1.15-inch diameter sodium iodide crystal with a 9-mil aluminum window. Detector B was a $\frac{1}{8}$-inch thick×1.22-inch diameter crystal with an 18-mil beryllium window.

where γ_{26} is the 26-kev gamma intensity per disintegration, L_{33} is the fraction of the 33-kev transitions which emit L x-rays, L_p is the intensity of L x-rays preceding the 59.6-kev level, ϵ is the counting efficiency, and $1-e^{-\lambda t}$ is the fraction of the delayed coincidences detected. The 26.4-kev gamma ray was detected by the proportional counter or by scintillation counter, the sample being covered by absorber to discriminate against x-rays. Neither type of detector gave good accuracy for the low-intensity gamma ray, the proportional counter having relatively high backgrounds of extraneous activity and chance coincidence and the scintillation counter having inadequate resolution. What appeared to be the best measurement yielded a value of 0.40 for the fraction, L_{33}, of the 33-kev transition which is L converted with the emission of L x-rays. From the scatter in the values from other measurements the uncertainty is estimated to be about 15%.

The prompt coincidence rate of L x-rays with L x-rays is a measure of the number of L-shell vacancies following conversion of the 26.4- and 33.2-kev transitions. From consideration of the decay scheme and conversion-electron data, the contribution to the prompt rate from other transitions is negligible. The magnitude of the contribution of delayed events to the coincidence rate of L x-rays is indicated by the delay-line data of Fig. 4. The coincidence rates are the sums of prompt and delayed rates, G_p and G_d, with

$$G_p = 2NbL_{33}\epsilon_A\epsilon_B, \quad (8)$$

and the delayed rates having the form

$$G_d = NL_pL_f(1-e^{-\lambda t})\epsilon_A\epsilon_B, \quad (9)$$

where b is the branching from the 59.6-kev level leading through the 26.4-kev transition, L is the fraction of the transition which emits L x-rays with the subscripts p and f designating transitions preceding and following the delayed state, respectively, and t is the decay time for the delayed state. The counting efficiencies are assumed equal for all transitions and are calculated from the singles rates of L x-rays in the two detectors. Corrected for the 26.4-kev gamma ray, escape peak and delayed coincidence contributions, the data of Fig. 4 give a value of 0.032 for the factor, $bL_{26}L_{33}$, in Eq. (8). In better measurements the sample was mounted in the collimator box with a $\frac{1}{2}$-inch collimator hole. The detector for the beam transmitted by the hole was the 2-mm thick crystal with 0.5-mil aluminum window. The other detector, facing the sample disk and the base of the box at a distance of about $\frac{5}{8}$-inch, was the $\frac{1}{8}$-inch crystal with 18-mil beryllium window. Measurements taken with resolving times of 98, 123, and 400 mμsec all gave the value 0.031 for the factor, $bL_{26}L_{33}$.

CONCLUSION

The absolute electromagnetic radiation intensities may be correlated with relative conversion-electron intensities to derive absolute electron intensities for the major transitions in the Np^{237} levels. None of the absolute electron intensities are reliably known at present by reason of the complexity of the spectrum and the usual difficulties in calibrating the spectrometer transmission. A derivation of absolute electron intensities which is independent of spectrometer transmission and the absorption of low-energy lines will be presented in a subsequent paper.

Preliminary values for two total L-shell conversion coefficients are implicit in the photon intensities. From the conversion-electron spectrum[4,8] it is clear that more than 90% of the L x-rays preceding the 59.6 kev level are from conversion of the 43.4-kev transition. Since the fluorescence yield for neptunium is expected to be about 0.5,[22] the total L conversion coefficient is near 180. Comparison with theoretical values[23] of 660 for an $E2$ transition and 80 for an $M1$ transition indicates mixing in qualitative agreement with observations of L-subshell conversion.[24] Similarly, the ratio of L x-rays from the prompt-coincidence measurement and the gamma-ray intensity is a measure of the conversion of the 26.4-kev transition. With the approximate correction for fluorescence yield, the L conversion coefficient is about 6 in agreement with previous estimates.[3,5]

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[22] B. B. Kinsey, Can. J. Research **26**, 404 (1948).
[23] M. E. Rose (privately circulated tables).
[24] Hollander, Smith, and Rasmussen, Phys. Rev. **102**, 1372 (1956).