

Nuclear Decay of Br^{84} and the Level Scheme in Kr^{84}

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The decay properties of fission product Br^{84} have been studied by scintillation methods. Its half-life was found to be 31.80 ± 0.08 minutes. Gamma-ray spectra showed gamma rays of energy 0.27, 0.35, 0.43, 0.47, 0.52, 0.61, 0.74, 0.81, 0.879 ± 0.007 , 1.01, 1.21, 1.47, 1.57, 1.74, 1.90 ± 0.02 , 2.05, 2.17, 2.47 ± 0.03 , 2.82, 3.03, 3.28, and 3.93 ± 0.03 Mev. Single-crystal and coincidence spectrometry established beta-ray groups at 4.71, 3.83, 2.80, 1.81, 1.39, and possibly 0.83 Mev. From these data a level scheme in Kr^{84} has been formulated with excited levels at 0.88, 1.90, 2.17, 2.36, 2.62, 2.71, 2.91, 3.35, 3.70, 3.91, and 4.18 Mev. The mass difference between the ground state of Br^{84} and Kr^{84} is represented by the 4.71-Mev beta ray which includes 32% of the beta-ray transitions.

I. INTRODUCTION

SHORTLY after the discovery of fission, Dodson and Fowler¹ and Hahn and Strassmann² reported a bromine fission product of half-life 30 to 40 minutes. Born and Seelmann-Eggebert³ assigned the 30-minute bromine activity to mass 84 by the reaction $\text{Rb}^{87}(n,\alpha)\text{Br}^{84}$.

The beta spectrum of Br^{84} has been studied with a magnetic lens spectrometer by Duffield and Langer,⁴ who reported beta groups of 4.679 ± 0.010 , 3.56, 2.53, and 1.72 Mev, the highest energy group being 40% abundant, having a $\log ft$ value of 7.75, and showing an allowed shape. Their coincidence experiments indicated that this 4.68-Mev group was the ground-state transition and presented an inconsistency with the most likely expectations of simple shell-model theory. Their subsequent search for an isomeric level in Kr^{84} gave negative results. In the decay of Rb^{84} , Huddleston and Mitchell⁵ observed a single level in Kr^{84} at 0.890 Mev. Additional work by Duffield and Langer⁴ on the Rb^{84} decay disclosed two gamma rays of 0.890 and 1.89 Mev. Recently Welker and Perlman⁶ have studied the decay of Rb^{84} and assigned the first two excited states in Kr^{84} at 0.89 and 1.91 Mev. From Coulomb excitation of krypton, Temmer and Heydenburg⁷ have found the first excited level in Kr^{84} at 0.880 Mev and have given it a $2+$ assignment.

The high decay energy of Br^{84} available for excitation of nuclear levels in Kr^{84} suggested that a number of very energetic gamma rays might be present, which in addition to its high fission yield, made an investigation of the decay of Br^{84} interesting and important. Thus the

present study was initiated, using scintillation methods to obtain both the beta- and gamma-ray spectra.

II. SOURCE PREPARATION AND HALF-LIFE DETERMINATION

The Br^{84} samples used in this study were prepared by neutron irradiation of UO_2SO_4 in the Oak Ridge Graphite Reactor. After irradiation, the targets were dissolved in dilute HNO_3 containing carrier Br^- and I^- , and following KMnO_4 oxidation, the Br_2 and I_2 were extracted into CCl_4 . After several cycles of selectively reducing the Br_2 with $\text{NH}_2\text{OH} \cdot \text{HCl}$ and extracting the I_2 , the Br^- was precipitated as AgBr .

From two separate experiments very closely agreeing half-lives have been determined for Br^{84} . In one the sample was mounted 9.3 cm above a 3-inch \times 3-inch NaI crystal and the integral gamma-ray decay followed. Here the pulse-height selector was set to discriminate against the low-energy gamma rays of Br^{83} which were also present. The resulting decay curve was exponential over about eight half-lives and a least squares analysis of the data gave a half-life value of 31.81 ± 0.04 minutes. In a second experiment a twenty-channel analyzer was set to cover the most intense gamma-ray peak (0.879 Mev). By a least squares analysis the peak was found to decay with a half-life of 31.79 ± 0.04 minutes. It is felt that an accurate representation of the Br^{84} half-life is given by 31.80 ± 0.08 minutes where the quoted deviation is an estimate of the combined standard and systematic errors. This is in agreement with the value 31.6 ± 0.2 minutes recently determined by Fix and Schindewolf.⁸

III. SINGLE CRYSTAL SPECTROMETER RESULTS

The gamma-ray scintillation spectrometer consisted of a 3-inch \times 3-inch cylindrical NaI crystal mounted on a Dumont 6363 photomultiplier tube by the method described by Lazar and Klema.⁹ Sources were centered 9.3 cm above the top surface of the crystal. To eliminate

* Operated by Union Carbide Nuclear Company for the U. S. Atomic Energy Commission.

¹ R. W. Dodson and R. D. Fowler, *Phys. Rev.* **55**, 880 (1939).

² O. Hahn and F. Strassmann, *Naturwissenschaften* **27**, 533 (1939).

³ H. J. Born and W. Seelmann-Eggebert, *Naturwissenschaften* **31**, 86 (1943).

⁴ R. B. Duffield and L. M. Langer, *Phys. Rev.* **81**, 203 (1951).

⁵ C. M. Huddleston and A. C. G. Mitchell, *Phys. Rev.* **88**, 1350 (1952).

⁶ J. P. Welker and M. L. Perlman, *Phys. Rev.* **100**, 74 (1955).

⁷ G. M. Temmer and N. P. Heydenburg, *Phys. Rev.* **104**, 967 (1956).

⁸ R. C. Fix and U. Schindewolf, Massachusetts Institute of Technology Annual Progress Report, June 1, 1955 to May 31, 1956 (unpublished), p. 34.

⁹ N. H. Lazar and E. D. Klema, *Phys. Rev.* **98**, 710 (1955).

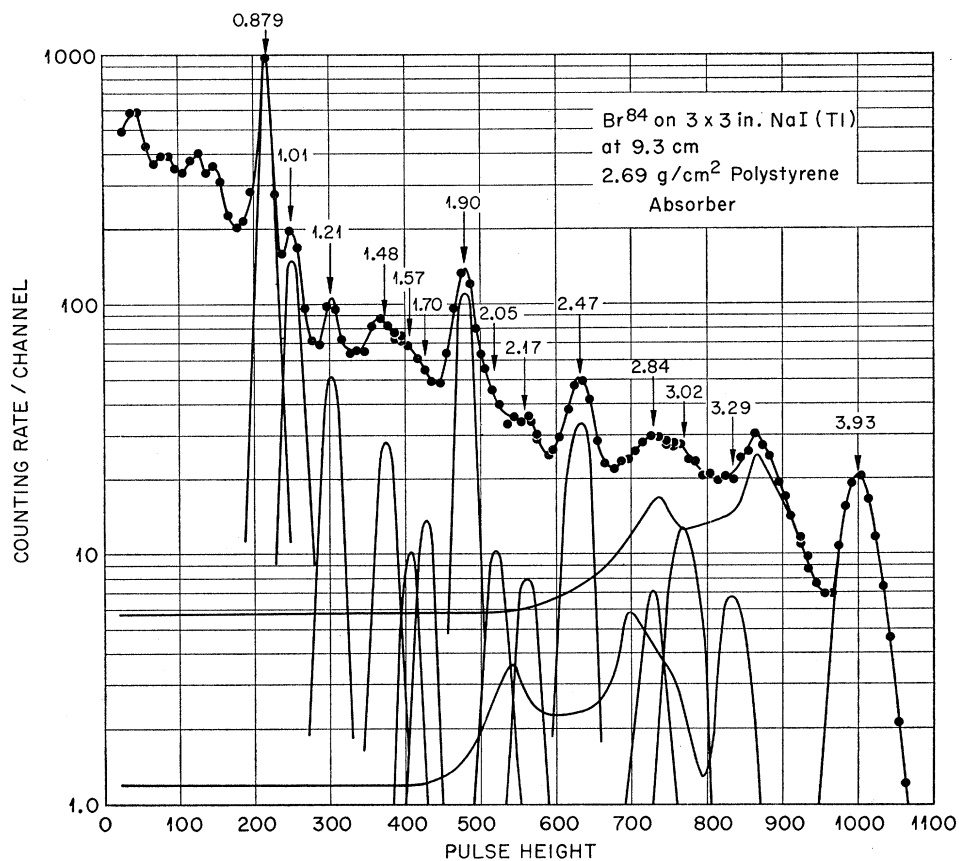


FIG. 1. High-energy portion of gamma-ray spectrum from a Br^{84} source 9.3 cm above a 3-inch \times 3-inch NaI crystal.

the beta background it was necessary to place a 2.69 g/cm² polystyrene absorber between the sample and the crystal. For the pulse-height analysis a twenty-channel analyzer designed by Bell, Kelley, and Goss¹⁰ was used.

A single crystal gamma-ray spectrum (high-energy portion) of Br^{84} is shown in Fig. 1. Although a complete analysis was performed to resolve the full-energy peaks, only the 3.93- and 3.28-Mev gamma rays¹¹ have their complete pulse-height distributions shown in order to avoid possible confusion as a result of the complexity of the fully analyzed spectrum. The full-energy peaks are drawn in with a Gaussian shape, whereas the shapes for the Compton and pair peaks are obtained from the pulse-height distributions of various energy gamma-ray standards run under conditions similar to this experiment. The lower energy portion of the gamma-ray spectrum run at expanded amplifier gain is displayed in Fig. 2. The analyzed components properly accounted for the gross pulse-height distributions except for a slight asymmetry on the high-energy side of the 3.28-Mev gamma ray and for the somewhat excessive widths of the 1.01- and 1.21-Mev full-energy peaks. Attempts

to sum the "backscatter" peaks (large-angle Compton scattering of the photons from the surroundings into the crystal) from the many gamma rays are subject to large error and thus make further analysis below about 0.35 Mev very impractical. However, it is later shown in coincidence experiments that this region does contain additional gamma rays.

Since 6363 photomultiplier tubes are known to exhibit a gain shift with counting rate,¹² the energies of the most prominent gamma rays were measured by counting the Br^{84} simultaneously with appropriately selected standards. The Bi^{207} 0.569- and 1.063-Mev peaks just bracket the most intense Br^{84} gamma ray whose energy was thereby determined as 0.879 ± 0.007 Mev. The highest energy gamma ray was determined as 3.93 ± 0.03 Mev, using ThC'' (2.615 Mev) and the zero-energy intercept of a calibration curve. By comparing with Na^{24} (1.368 and 2.753 Mev) two other prominent gamma rays were found to have energies of 1.90 ± 0.02 and 2.47 ± 0.03 Mev. The energies of the remaining Br^{84} gamma rays were obtained from these four most prominent Br^{84} peaks as internal standards.

Before calculating the relative gamma-ray intensities, it was necessary to make certain assumptions about the decay scheme to check for possible coincident summing

¹⁰ Bell, Kelley, and Goss, Oak Ridge National Laboratory Report ORNL-1278, 1951 (unpublished).

¹¹ Energies of gamma rays discussed in the text are from Table I which lists the best energy values from several experiments. The figures show values determined in the particular experiments.

¹² Bell, Davis, and Bernstein, Rev. Sci. Instr. 26, 726 (1955).

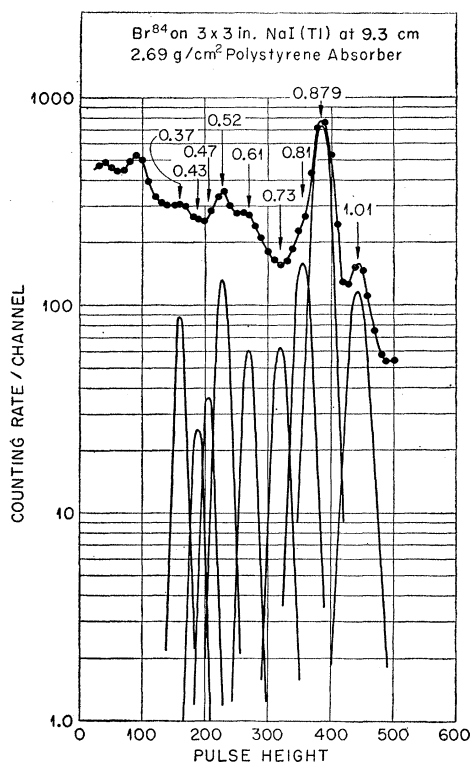


FIG. 2. Low-energy portion of a gamma-ray spectrum from Br^{84} at expanded amplifier gain. Other conditions are the same as Fig. 1.

of cascading gamma rays (the decay scheme assumed was based on the principal observations from coincidence experiments). From calculations of the type described by Lazar and Klema,⁹ the loss in gamma-ray intensity from coincident summing was found to be of

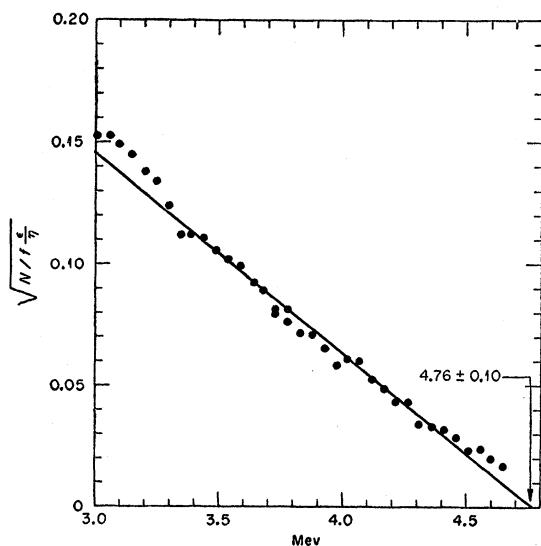


FIG. 3. Fermi analysis of high-energy portion of Br^{84} beta-ray spectrum measured on a $1\frac{1}{4}$ -inch \times 1-inch anthracene scintillation spectrometer.

the order of one percent and therefore negligible. The full-energy peak areas of the gamma rays in Figs. 1 and 2 were corrected for absorption losses in the polystyrene and for the peak efficiencies and geometry of the crystal used.¹³ The best values of the gamma-ray energies and the relative gamma-ray intensities are shown in Table I.

The beta-ray spectrum was measured with an anthracene scintillation spectrometer using a $1\frac{1}{4}$ -inch \times 1-inch cylindrical crystal. A thin source of the activity in the form of silver bromide precipitate was mounted on 0.8 mg/cm^2 polystyrene film. As the beta spectrum was already known to be complex,⁴ only the

TABLE I. Summary of Br^{84} gamma-ray data.

Gamma-ray energy, Mev	Intensities relative to 0.879-Mev gamma ray as 100 units		Gamma ray appeared in coincidence with:			
	From singles data	Best value from coincidence data	0.879 Mev	1.90 Mev	2.47 Mev	3.93 Mev
0.27 ± 0.04	a	1.1 ± 0.4	Yes	Yes	?	Yes
0.35 ± 0.03	3.5 ± 1.7	2.5 ± 1.0	Yes	Yes	?	No
0.43 ± 0.04	$\sim 1^b$	5.0 ± 2.0	Yes	Yes	No	No
0.47 ± 0.04	2.0 ± 1.0	2.1 ± 0.5	?	Yes	?	No
0.52 ± 0.03	7.3 ± 2.0	5.7 ± 1.7	Yes	Yes	?	No
0.61 ± 0.03	5 ± 2	5.4 ± 1.6	Yes	?	?	No
0.74 ± 0.03	6.7 ± 2.5	3.7 ± 1.0^c	Yes	No	?	No
0.81 ± 0.03	20 ± 4	15 ± 4	Yes	Yes	No	No
0.879 ± 0.007	100	100	No	No	Yes	No
1.01 ± 0.02	20 ± 3	21 ± 2	Yes	? ^d	No	No
1.21 ± 0.02	7.6 ± 0.8	7.7 ± 1.0	Yes	Yes	No	No
1.47 ± 0.03	4.2 ± 1.1	4.1 ± 0.8	Yes	No	No	No
1.57 ± 0.05	2.3 ± 1.1	2.3 ± 1.1	Yes	Yes	No	No
1.74 ± 0.05	3.6 ± 1.3	4.9 ± 1.0	Yes	No	No	No
1.90 ± 0.02	36 ± 4	36 ± 4	No	No	No	No
2.05 ± 0.05	3.8 ± 1.3	3.8 ± 1.0	?	Yes	No	No
2.17 ± 0.04	3.9 ± 1.0	3.9 ± 1.0	No	No	No	No
2.47 ± 0.03	18 ± 3	13 ± 3	Yes	No	No	No
2.82 ± 0.05	4.0 ± 1.6	4.5 ± 1.3	Yes	No	No	No
3.03 ± 0.05	9.8 ± 3.0	7.2 ± 1.5	Yes	No	No	No
3.28 ± 0.05	5.8 ± 2.0	5.6 ± 1.2	Yes	No	No	No
3.93 ± 0.03	25 ± 4	25 ± 4	No	No	No	No

^a This peak appears so near the "backscatter" peak that its analysis was not possible.

^b Since it is difficult to subtract the Compton background, no attempt was made to determine the limit of error in the intensity.

^c Another 0.74-Mev gamma ray which would not have appeared in any of the coincidence data is proposed elsewhere in the decay scheme and is discussed in the text.

^d There were slight indications for a very small coincidence contribution from a 1.01-Mev gamma ray which could be accommodated at several positions above the 1.90-Mev level.

end-point region of the maximum beta-ray component was studied. The Fermi analysis shown in Fig. 3 yielded a value of 4.76 ± 0.10 Mev which is probably not as accurate as the energies assigned to the other beta-rays from coincidence experiments, since in the single crystal spectrum the random summing of Compton electrons from high-energy gamma rays causes considerable distortion near the end point. Energy calibration was obtained from the 0.625-Mev line of Ba^{137m} and the 0.976-Mev line of Bi^{207} . The linearity of the spectrometer above 0.976 Mev was checked with

¹³ Lazar, Davis, and Bell, Trans. Inst. Radio Engrs., Nuclear Sci. NS-3, 136 (1956).

a precision pulse generator and by a measurement of the P^{32} beta-ray spectrum.

IV. COINCIDENCE SPECTROMETRY

A. Gamma-Gamma

Gamma-gamma coincidence measurements were made using a "fast-slow" coincidence circuit whose resolving time was $2\tau = 0.17 \mu\text{sec}$. The radiations were viewed by two 3-inch \times 3-inch NaI crystals at 180° to each other and the sources were located at the center of a collimating anti-Compton shield similar to that described by Bell.¹⁴ In separate experiments the single-channel window was set on the principal gamma peaks and the coincident gamma-ray spectrum in each case recorded on the multichannel analyzer.

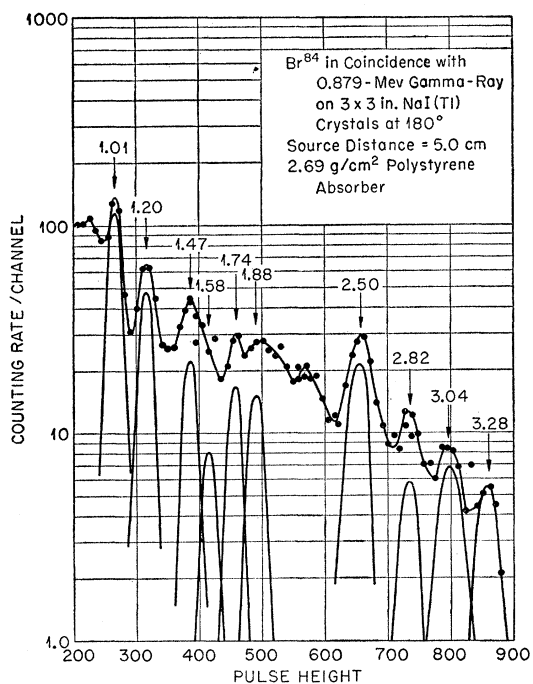


FIG. 4. High-energy region of Br^{84} gamma-ray spectrum in coincidence with 0.879-Mev gamma rays.

The gamma-ray spectra in coincidence with 0.879 Mev are shown in Figs. 4 and 5. The full-energy peaks were determined from analyses of the curves in the same manner as described for the single-crystal spectra. In Fig. 5 the Compton and pair contributions from the gamma rays above 1.01 Mev were normalized from the data of Fig. 4.

In Fig. 4 the 3.28-, 3.03-, 2.82-, and 2.47-Mev peaks are clearly resolved and thus provide a straightforward analysis. However, in the region of 2.0–2.3 Mev the situation is more complex; there are some indications

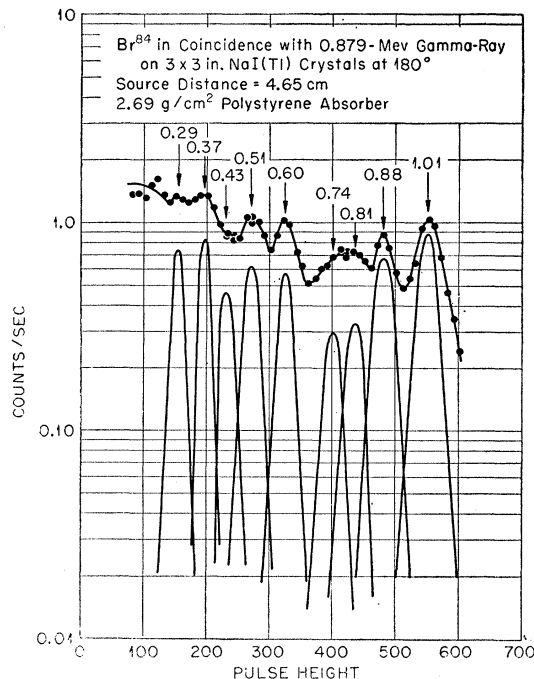


FIG. 5. Br^{84} gamma-ray spectrum (at expanded amplifier gain) in coincidence with 0.879-Mev gamma rays.

for gamma rays of about 2.0 and 2.2 Mev, but the very low intensity and poor statistics do not warrant their insertion. That there is a 2.05-Mev gamma ray in coincidence with 0.879 Mev, is, however, later verified on the basis of its coincidence with the 1.90-Mev transition. The peak at 1.57 Mev is easily resolved although the poor statistics of the gross spectrum in this region may cause an error in its intensity of as much as 50%.

The lower energy portion of the 0.879-coincident gamma-ray spectrum at expanded amplifier gain is shown in Fig. 5. At 0.51 Mev a point-by-point subtraction gives a peak much too broad which implies that this is probably a composite peak containing both the 0.47- and 0.52-Mev gamma rays observed in the single crystal spectrum. Similarly at 0.35 Mev a peak much broader than the normal Gaussian distribution can probably be assumed to contain additional gamma rays of about this energy.

To determine the intensities of the gamma rays in Figs. 4 and 5 relative to the 0.879-Mev peak in the window, use is made of the following general equation:

$$P(\gamma_1) = \epsilon_p(\gamma_1)\Omega(\gamma_1)e^{-\mu d}C_w(fA + \sum_i f_i' B_i)\bar{W}(\theta), \quad (1)$$

where $P(\gamma_1)$ is the coincidence peak area of the gamma-ray coincident with the counts in the single-channel window, $\epsilon_p(\gamma_1)$ and $\Omega(\gamma_1)$ are the peak efficiency and solid angle for the detection of γ_1 , $e^{-\mu d}$ is the fraction of γ_1 transmitted through the beta absorber, and C_w is the observed counting rate in the window of the single-channel analyzer. A is the fraction of the observed rate in the window due to the gamma ray with which co-

¹⁴ P. R. Bell in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 5.

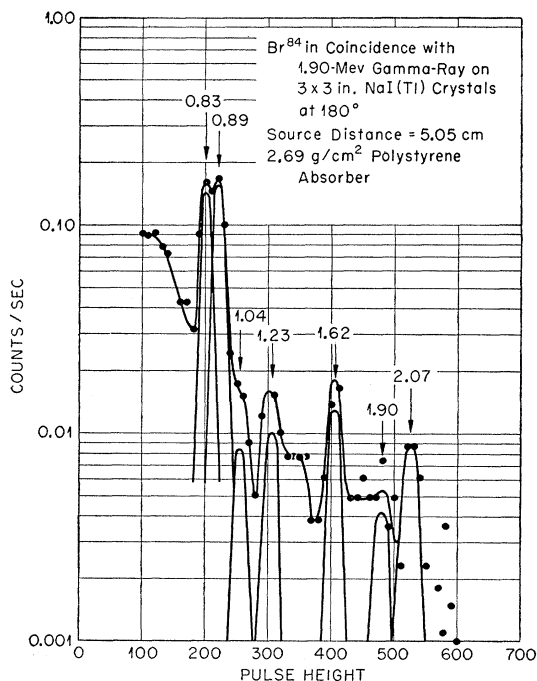


FIG. 6. Br^{84} gamma-ray spectrum coincident with 1.90-Mev gamma rays.

incidences are being measured and may be obtained from the single-crystal spectrum. The factor f is the number of coincidence processes relative to the number of gamma rays of interest in the window. The term $\sum_i f_i' B_i$ corrects for Compton pulses in the window from higher energy gamma rays which are also in coincidence with γ_1 . B_i is the fraction of counts in the window due to γ_i and f_i' the relative number of γ_1 coincident with γ_i , as determined from the decay scheme. $\bar{W}(\theta)$ is the angular distribution function of the two gamma rays integrated over the face of the crystal.¹⁵ Except for the case of the 0.879–1.01-Mev coincidences, the value of $\bar{W}(\theta)$ was unknown and hence was assumed to be unity. By using Eq. (1), the 0.88- and 1.9-Mev peaks were shown to arise from coincidences with Compton pulses in the window.

The gamma-ray spectrum coincident with 1.90 Mev is shown in Fig. 6 and as in the spectrum coincident with 0.88 Mev, there is an undefined distribution in the region of 2.2 Mev. Also at 1.75 Mev there may be a coincident peak, but because of poor statistics no attempt has been made to determine its intensity. Although the spectrum below about 0.6 Mev is not shown in Fig. 6, it was obtained from an experiment at expanded amplifier gain from which an analysis of the coincident gamma rays up to 1 Mev was made. As was the case in the 0.879-Mev coincidence experiment, the appearance of the 0.88- and 1.90-Mev peaks in Fig. 6

¹⁵ M. E. Rose, Phys. Rev. **91**, 610 (1953).

can be accounted for by the other components in the window.

In the gamma-ray spectrum coincident with the 2.47-Mev gamma ray, a very prominent peak was observed at 0.88 Mev. There was also some evidence for very weak gamma rays at 0.74, 0.60, 0.50, and 0.35 Mev. With the single-channel window set on the 3.93-Mev gamma ray, there appeared a single coincident peak at 0.27 Mev.

A summary of the intensities measured in various gamma-gamma coincidence experiments is included in Table I, in which the coincidence ratios [f in Eq. (1)] have been corrected to the same basis as the single-crystal intensities using the decay scheme and gamma-ray branching ratios calculated from the single-crystal intensities.

B. Beta-Gamma

Whereas considerable error may be introduced in determining the lower energy beta-ray components from a complex of beta groups, it is possible to simplify the Fermi plot by looking at the beta spectra in coincidence with appropriately selected gamma rays, and hence to obtain the various beta-ray end points more accurately. Beta-gamma coincidence spectrometry is also very helpful in determining the positions of gamma rays in a decay scheme. In the beta-gamma coincidence experiments of this study the same "fast-slow" coincidence circuitry mentioned previously was employed. A $1\frac{1}{4}$ -inch \times 1-inch anthracene crystal was used in the multi-

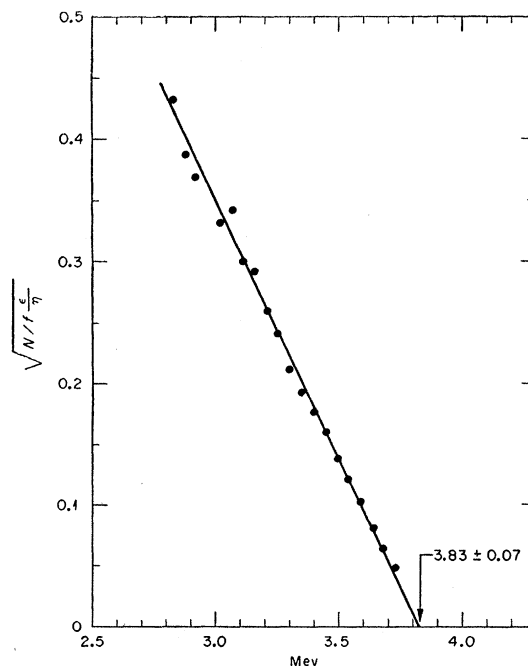


FIG. 7. Fermi analysis of beta-ray spectrum in coincidence with 0.879-Mev gamma rays.

channel circuit and a 3-inch \times 3-inch NaI crystal in the single-channel side.

Figures 7, 8, and 9 show Fermi analyses of beta-ray spectra in coincidence with gamma rays of 0.879, 1.90, and 2.47 Mev, respectively. A beta-ray group of 3.83 ± 0.07 Mev was found in coincidence with 0.879 Mev; two groups of energy 2.80 ± 0.10 and 1.81 ± 0.06 Mev were in coincidence with 1.90 Mev; a group of 1.39 ± 0.10 and possibly 0.83 ± 0.08 Mev was in coincidence with the 2.47-Mev gamma ray. The 0.83-Mev component may be questionable, since backscatter from the thick source used probably introduced a considerable distortion of the low-energy beta-ray spectrum.

To determine if the 4.76-Mev beta ray of Fig. 3 was the ground state transition, the window of the single-channel analyzer was set to cover that portion of the 4.76-Mev beta spectrum beyond the end point of the 3.83-Mev component. No gamma rays appeared in coincidence, however.

V. DECAY SCHEME

A decay scheme consistent with the majority of the Br^{84} observations is presented in Fig. 10. It is assumed that since no gamma rays were in prompt coincidence with the beta ray measured as 4.76 Mev, this group represents the ground-state transition. As mentioned previously, the beta-ray energy obtained from the single-crystal spectrum is less reliable than the other beta-ray energies measured by the coincidence method; hence, the preferred mass difference between Br^{84} and Kr^{84} is taken as 4.71 ± 0.08 Mev, by using the beta-ray

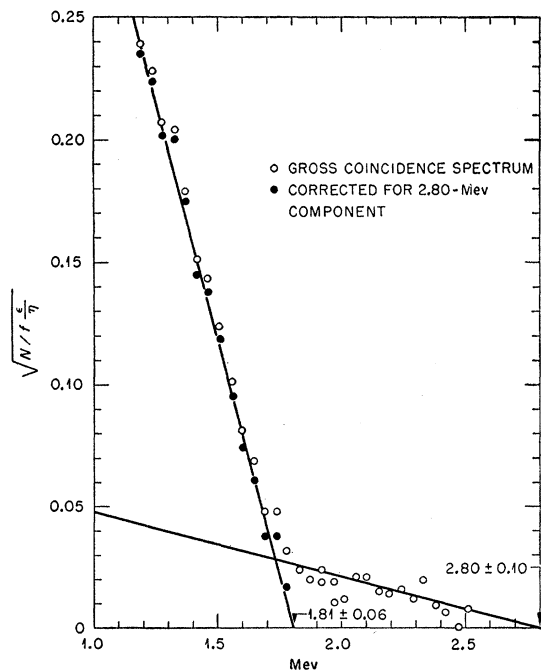


FIG. 8. Fermi analysis of beta-ray spectrum coincident with 1.90-Mev gamma rays.

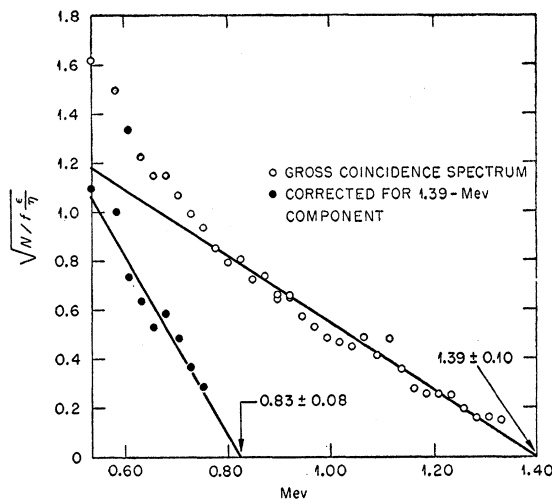


FIG. 9. Fermi analysis of beta-ray spectrum in coincidence with 2.47-Mev gamma rays.

energy of 3.83 ± 0.07 Mev in coincidence with the 0.879 ± 0.007 -Mev gamma ray. A coincidence between the 1.90-Mev gamma peak and a 2.80-Mev beta ray defines the second excited level at 1.90 Mev.

Although Figs. 4 and 6 give slight indications for coincidences between the 1.90- and 2.17-Mev gamma rays, such can be discounted on the basis of intensity arguments. The latter is therefore assumed to proceed to the ground state of Kr^{84} and to bring about a level at 2.17 Mev.

Levels at 2.36 and 2.62 Mev may be inferred since the 1.47- and 1.74-Mev gamma rays are in coincidence with the 0.879-, but not with the 1.90-Mev peak. There is no direct experimental evidence for the 2.71-Mev level. However, its insertion helps account for the 1.21-, 0.81-, 0.61- and 0.35-Mev gamma rays while maintaining consistency with the beta-gamma coincidence data. Strong coincidences of a 1.81-Mev beta ray with the 1.90-Mev gamma ray lead to a level assignment at 2.91 Mev. Gamma rays in coincidence with the 0.879-Mev gamma ray determine the levels at 3.35, 3.70, 3.91, and 4.18 Mev. A beta-ray group of 1.39 Mev also distinguishes the level at 3.35 Mev, and the single peak at 0.27 Mev in coincidence with the 3.93-Mev gamma ray further verifies the 3.91- and 4.18-Mev levels.

It seems noteworthy to point out that in the above arguments for eleven excited states in Kr^{84} only the 2.71-Mev level was invoked on grounds other than from coincidence experiments. However, it is not to be implied that the scheme of Fig. 10 gives all the levels excited in the Br^{84} decay. The multitude of gamma rays encountered in this problem makes it quite likely that additional transitions of low intensity have not been detected and perhaps are related to additional levels. In fact, the gamma-ray spectrum in coincidence with the 2.47-Mev peak gives some indications for the possibility of additional excited levels at high energies.

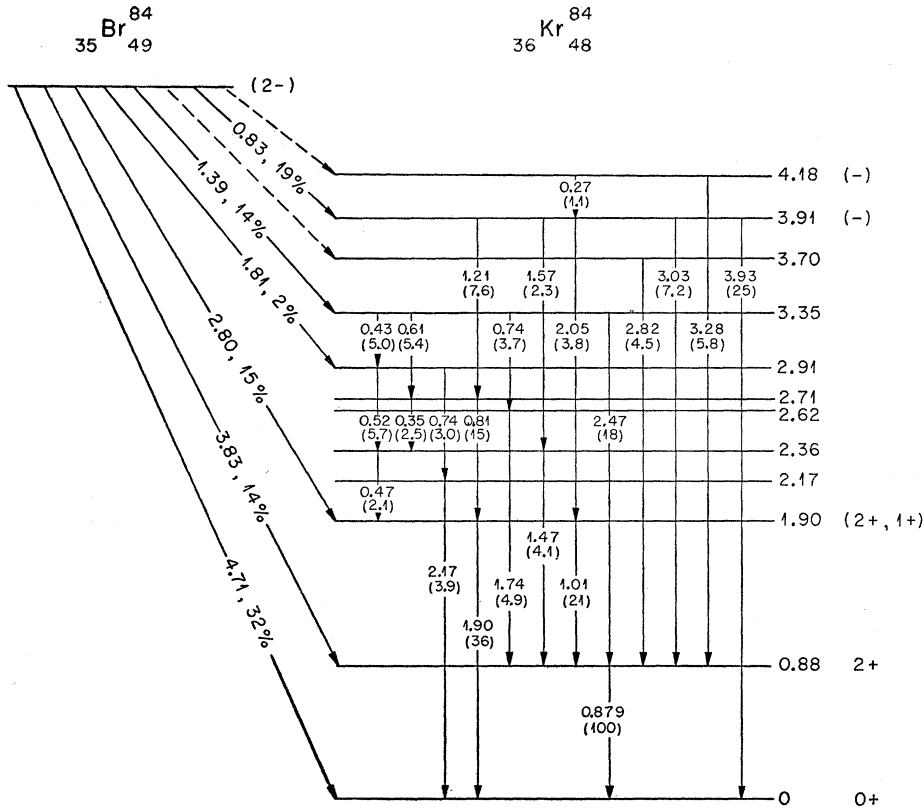


FIG. 10. Decay scheme proposed for Br^{84} . All energies are in Mev, and numbers in parentheses beneath the gamma-ray energies are the relative intensities from Table I.

Placement of the 0.879-, 1.01-, 1.47-, 1.74-, 1.90-, 2.17-, 2.47-, 2.82-, 3.03-, and 3.28-Mev gamma rays in the scheme of Fig. 10 is dictated by the coincidence results with 0.879 and 1.90 Mev. Although a faint peak at about 1 Mev was detected in coincidence with 1.90 Mev, the intensity of this gamma ray would be less than one percent of the intensity of the 0.879-Mev gamma ray. Hence, the 1.01-Mev gamma ray must be assigned as shown in the decay scheme, which is inconsistent with the observations of Welker and Perlman⁶ on the decay of Rb^{84} . While the 1.90-Mev gamma ray was detected, they did not observe the 1.01-Mev cascade transition in either the single-crystal or coincidence spectra. To clarify this situation, the single-crystal and 90° coincidence spectra of Rb^{84} were measured in this laboratory with spectrometer arrangements similar to those described earlier. In Fig. 11(a) the low-energy region of the Rb^{84} single-crystal spectrum is shown whereas Fig. 11(b) shows the 1.01-Mev gamma ray in coincidence with 0.879 Mev.

An analysis of the Rb^{84} single-crystal spectrum gave a ratio $I(0.88)/I(1.90) = 90$ compared with the value 71 calculated from the data of Welker and Perlman. Using the anisotropy $[\bar{W}(180^\circ) - \bar{W}(90^\circ)]/\bar{W}(90^\circ) = -0.246$ for the 1.01-0.879 Mev cascade measured in a Br^{84} angular correlation experiment, it was possible to determine the value of $\bar{W}(\theta)$ for both the 90° and 180° coincidence experiments. From two closely agreeing Rb^{84} coincidence experiments, a value of $I(1.01)/$

$I(1.90) = 0.59$ was calculated by use of Eq. (1), compared with an upper limit of 0.33 from the data of Welker and Perlman. With this same treatment the value in the case of the Br^{84} decay was $I(1.01)/I(1.90) = 0.60$. Therefore, since less than 1.01-Mev separation exists between the 1.90-Mev Kr^{84} excited level and the ground state of Rb^{84} ,⁶ and as the 0.88- and 1.90-Mev levels have been established as the first two excited states of Kr^{84} , this very close agreement confirms that the 1.01-Mev transition cascades directly into the 0.88-Mev Kr^{84} level.

The 2.05-Mev transition appeared in coincidence with 1.90 Mev, and as there is no strong evidence for an excited Kr^{84} level above 4.18 Mev, assignment of this transition from the 3.91- to the 1.90-Mev level is reasonable.

The single-crystal analysis for the 0.74-Mev gamma ray yielded an intensity of 6.7 units compared with the value 3.7 determined from its coincidence with 0.879 Mev. It is assumed, therefore, that 3.7 units of the 0.74-Mev gamma-ray cascade via the 1.74-Mev path to the 0.88-Mev level and the remaining 3 units cascade into the 2.17-Mev level, accounting for the intensity of the 2.17-Mev transition. In placing the 0.81-Mev gamma ray as shown, its relative intensities measured from single-crystal spectra and 1.90-Mev coincidence experiments agree; but the relative intensity from the 0.879-Mev coincidence data disagrees with the other results, probably because of difficulties associated with

analyzing the coincidence pulse-height spectrum in this region. The remaining gamma rays have been placed in the scheme in the manner most consistent with intensity arguments based on both the single-crystal and coincidence data, as well as with the available beta-ray data.

In an experiment designed to establish the beta-ray branching to the levels of Kr^{84} , the beta disintegration rate of an aliquot of Br^{84} was measured on a 4π proportional counter, correcting for a small contribution from Br^{83} and its daughter Kr^{83m} . Simultaneously the absolute rate of 0.879-Mev gamma rays in another aliquot was determined by gamma-ray spectrometry. Then using the decay scheme of Fig. 10 and the absolute beta- and gamma-ray rates, the beta branching to the levels of Kr^{84} was calculated. In this manner the beta branching to the ground state was found to be 0.319 ± 0.050 . The various beta groups are listed in Table II together with their associated intensities and comparative half-lives, the latter computed from Moszkowski's nomographs.¹⁶ Errors in intensities were assigned from an analysis of the errors in the gamma-ray intensities, allowing for the possibility of additional weak gamma rays.

VI. DISCUSSION

For Br^{84} the ground-state proton configuration predicted by the shell model is $\dots(p_{3/2})^4(f_{5/2})^3$ with a possibility for $\dots(p_{3/2})^3(f_{5/2})^4$. The expected neutron configuration is $\dots(p_{1/2})^2(g_{9/2})^{-1}$, and less probably, $\dots(p_{1/2})^1(g_{9/2})^{10}$. When one uses Nordheim's rules,¹⁷ the most likely spin and parity possibilities are $2-$ and $1+$, the former being preferred, since the measured spins of Rb^{85} , Kr^{83} , and Sr^{87} as well as the spin and

TABLE II. Intensities and comparative half-lives of Br^{84} beta-ray transitions. (Beta-ray subscripts denote final states.)

Beta-ray group	Experimental energies, Mev	Intensity	$\log ft$
β_0	4.71 ± 0.08	0.319 ± 0.050	7.7 ± 0.1
$\beta_{0.88}$	3.83 ± 0.07	0.144 ± 0.029	7.7 ± 0.1
$\beta_{1.90}$	2.80 ± 0.10	0.150 ± 0.060	$7.1_{-0.1}^{+0.2}$
$\beta_{2.17}$		~ 0	...
$\beta_{2.36}$		~ 0	...
$\beta_{2.62}$		~ 0	...
$\beta_{2.71}$		~ 0	...
$\beta_{2.91}$	1.81 ± 0.06	$(0.015)^a$	(7.3)
$\beta_{3.35}$	1.39 ± 0.10	0.136 ± 0.035	5.9 ± 0.1
$\beta_{3.70}$		$(0.019)^a$	(6.0)
$\beta_{3.91}$	0.83 ± 0.08	0.187 ± 0.038	5.0 ± 0.1
$\beta_{4.18}$		0.029 ± 0.010	$5.4_{-0.1}^{+0.2}$
β_{total}		1.00	

^a Intensity value uncertain due to possibility of additional weak gamma rays decaying to or from this level.

parity assignments of Rb^{84} and Rb^{86} can be attributed to $f_{5/2}$ proton and $g_{9/2}$ neutron orbitals. The ground and first excited states of Kr^{84} are known to be $0+$ and $2+$, respectively, and each of the beta branches to these two levels has a $\log ft$ of 7.7. Accordingly, the $2-$ assignment appears reasonable for the Br^{84} ground state. This, however, is not in agreement with the interpretation of the Br^{84} beta spectrum by Duffield and Langer⁴ who reported an allowed shape for the ground-state transition. On examining their data it seems possible that the expected unique shape may have been masked by the lower energy beta rays. Their analysis of the beta-ray spectrum using an allowed shape for the most energetic group possibly accounts for their having reported a larger ground-state branching and lower energies than those of this research.

For the second excited level in Kr^{84} either a $1+$, $2+$, or $3+$ assignment is compatible with $\log ft=7.1$ for the beta branch to this 1.90-Mev state. Using the Weisskopf formula¹⁸ and estimates of the matrix elements based on the lifetime systematics of Goldhaber and Sunyar,¹⁹ ratios of the intensities of the 1.90- to the 1.01-Mev gamma rays were calculated for the possible spin and parity assignments and transition multiplicities. Comparison with experiment narrows the assignment to the 1.90-Mev level as either $1+$ or $2+$.

If one assumes that the second excited state in Kr^{84} is $1+$ and that the 1.01- and 0.88-Mev gamma rays form a pure $M1 \rightarrow E2$ cascade, an anisotropy of -0.26 is calculated by using the tables of Biedenharn and Rose.²⁰ This is in close agreement with the measured value -0.25 , whereas all other pure multipole assignments give answers quite remote from the experimental observation. If, however, this level is $2+$ and the 1.01-Mev transition is $E2$ with an $M1$ admixture of only

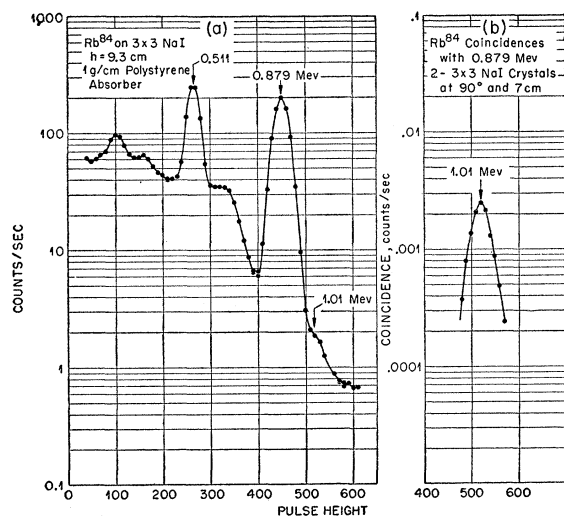


FIG. 11. (a) Low-energy portion of gamma-ray spectrum from a Rb^{84} source 9.3 cm above a 3-inch \times 3-inch NaI crystal. (b) Rb^{84} gamma-ray spectrum in the region of 1 Mev coincident with 0.879-Mev gamma rays.

¹⁶ S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).

¹⁷ L. W. Nordheim, Revs. Modern Phys. **23**, 322 (1951).

¹⁸ J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1954), Chap. XII.

¹⁹ M. Goldhaber and A. W. Sunyar, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. XVI.

²⁰ L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).

about 0.1%, a value in accord with experiment is also obtained. Hence, it is not possible to clearly discriminate against either a 1+ or 2+ assignment although the existing information on second excited states of even-even nuclei would tend to favor 2+.

Negative-parity assignments to the 3.91- and 4.18-Mev levels are based on the $\log ft$ values of about 5.

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Beta Spectrum of Th^{233} †

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The beta spectrum of Th^{233} has been measured in a ring-focus solenoidal spectrometer and was not found to exhibit an anomalous shape, as had been previously reported. The end point is 1.23 ± 0.01 Mev in agreement with the earlier value. It is shown that Th^{233} decays predominantly to the ground state of Pa^{233} . Half-life measurements yield a value of (22.4 ± 0.1) min for Th^{233} , somewhat shorter than the previously reported values.

IN an earlier investigation,¹ it was found that the beta spectrum of Th^{233} appeared to have an anomalous shape which was not explainable in terms of the generally accepted theory of beta decay.² The momentum distribution exhibited a deficiency of low-energy electrons which became manifest in that the Fermi-Kurie plot of the data fell below the usual straight line, in the energy region below 600 kev. No satisfactory explanation of this peculiar behavior was available at the time.

In the present work, the spectrum of Th^{233} was re-investigated under somewhat improved conditions. Measurements were made in a new ring-focus solenoidal spectrometer patterned after the instrument designed by Schmidt³ and operating at a transmission and resolution of about one percent. The electrons were detected with a methane-flow (10 cm Hg pressure) proportional counter having a 0.9-mg/cm^2 aluminized Mylar window. The sources were in the form of $\frac{1}{8}$ -in. diameter disks mounted on 0.9 mg/cm^2 aluminized Mylar backing. Three different sources were prepared by slow-neutron capture in the Los Alamos "Water

Boiler" reactor. One was a uniform, 3-mg/cm^2 slurry deposit of ThO_2 on a thick 0.002-in. Al backing. A second run was obtained with a source of 1.2 mg/cm^2 of ThO_2 bonded with dilute Zapon, mounted on 0.9-mg/cm^2 Mylar. A third run was obtained with a vacuum evaporated deposit of ThF_4 0.36 mg/cm^2 thick, on a backing of 0.00025-in. aluminum. Measurements were started only after all 2.3-min Al^{28} activity had died out. It was then found that all parts of the measured distributions decayed with the same half-life.

The Fermi-Kurie plots of the data are shown in Fig. 1. It is clear that there is here no anomalous behavior but only the usual low-energy deviation which is to be expected from such relatively thick sources and backings. The end point is 1.23 ± 0.01 Mev, in agreement with the earlier result.

It is difficult, in retrospect, to account for the results which were observed in the earlier work. The original

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¹ Bunker, Langer, and Moffat, *Phys. Rev.* **80**, 468 (1950).

² A somewhat similar anomalous beta spectrum (i.e., an apparent deficiency of electrons at low energy) was reported for Ru^{103} by E. Kondaiah in *M. Siegbahn Commemorative Volume* (Almqvist and Wiksells Boktryckeri AB, Uppsala, 1951), p. 411. Recent measurements on the beta spectrum of Ru^{103} by R. L. Robinson and L. M. Langer (to be published) yield a normal, linear Fermi-Kurie plot with an end point of 227 kev for the spectrum of beta particles measured in coincidence with the 498-kev gamma ray. It would appear that the apparent anomalous behavior, in this case too, was probably instrumental.

³ F. H. Schmidt, *Rev. Sci. Instr.* **23**, 361 (1952). A detailed description of the spectrometer used in this investigation will appear in another paper.

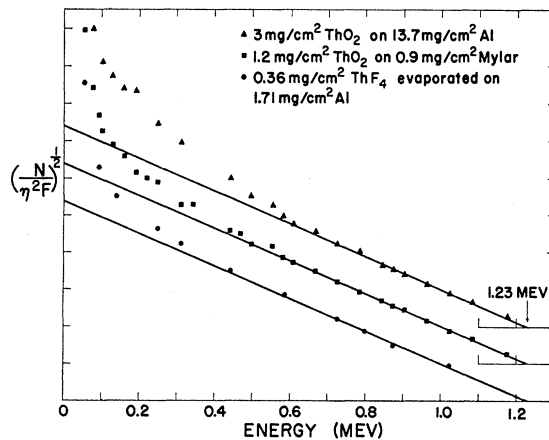


FIG. 1. Fermi-Kurie plots of the Th^{233} beta spectrum.