Radioactive Decay of Pm^{143}, Pm^{144}, and Pm^{146}†

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The decay of Pm^{143}, Pm^{144}, and Pm^{146} has been studied with permanent magnet spectrographs, an intermediate-image beta spectrometer, and scintillation counters. Results on the electron-capture decay of Pm^{143} and Pm^{144} confirm the findings of previous investigators. The results of gamma-gamma directional correlation measurements are consistent with the spin assignments for the Nd^{144} levels made by Ofer. Pm^{146} ($T_{\frac{1}{2}}\sim710$ days) is found to decay both by beta emission to a level of 749 kev in Sm^{146} and by electron capture to levels of 453 and 1198 kev in Nd^{146}. The percentage branchings are 35%, 35%, and 30%, respectively. Gamma-ray transitions of 453 and 745 kev in cascade in Nd^{146} and 749 kev in Sm^{146} are observed. The end-point energy for the beta-decay branch is measured to be 779 kev. Directional correlation and internal conversion coefficient measurements are most consistent with a 3+ assignment for the 1198-kev level in Nd^{146}, with a 2+ assignment being possible, but less likely, since the cross-over transition is absent or extremely weak.

I. INTRODUCTION

CONSIDERABLE data are available on the decay schemes and level systematics of nuclei whose low-lying excitation spectra may be explained by either the single-particle or the collective model. Much less data are available for the nuclei in the so-called transition regions, i.e., those nuclei of mass 140–152 and 180–200. We have been investigating some of the level schemes in the region of mass 143 to 148 and it is the purpose of this paper to present some of our results on the decay of Pm^{143}, Pm^{144}, and Pm^{146}.

Early investigations of Pm^{143} and Pm^{144} by Fisher[1] and Wilkinson and Hicks[2] indicated that Pm^{143} and Pm^{144} decay by electron capture, with the half-lives of each being about 300 days.

Recently, Ofer[3] has investigated these decays in detail and proposed decay schemes for both. He produced his sources by bombardment of Pr isotopes with alpha particles. He found that Pm^{143} decays by electron capture to the ground state and to a state at 740 kev in Nd^{143}, with the branching being 55% and 45%, respectively. The conversion coefficient of the 740-kev transition was in agreement with that for an $M1$ transition.

Ofer found three gamma rays in cascade in Nd^{144} following the electron-capture decay of Pm^{144}. These gammas have energies of 695, 610, and 475 kev, giving a sequence of levels at 695, 1305, and 1780 kev in Nd^{144}. Ofer's directional correlation and conversion coefficient measurements established a sequence of spins and parities of 0+, 2+, 4+, and 6+ for these levels. He found that 55% of the electron capture decay occurs to the 1305-kev level and 45% to the 1780-kev level, and log ft values were consistent with a spin of 6- or 5- for Pm^{144}. A value of $(13\pm4)\times10^{-11}$ sec and an upper limit of 3×10^{-11} sec were found for the mean lifetimes of the 1305- and 695-kev levels, respectively. Toth and Nielsen[4] have reported data on Pm^{144} which are in agreement with those of Ofer. They found a half-life of about 450 days for Pm^{144}. Neutron capture work by Campion *et al.*[5] showed capture gamma rays populating levels at 696, 1313, and 1556 kev as well as many other high-lying levels in Nd^{144}. Numerous investigations of the beta decay of Pr^{144} to Nd^{144} have been made.[6] Beta decay populates the 2+ level at 695 kev and a 1− level at 2180 kev.

Some early work on the decay of Pm^{146} was carried out by Fisher[1] and by Long, Pool, and Kundu.[7] These investigations indicated that Pm^{146} decays to Sm^{146} by emission of beta particles with a maximum energy of about 750 kev and a half-life of between 1 and 2.5 years. The results of our investigation establish that Pm^{146} also decays by electron capture to levels in Nd^{146}.

Data on the levels in Nd^{146} were obtained by Bernstein *et al.*[8], who studied the beta decay of Pr^{146} to Nd^{146}. Their data indicated levels in Nd^{146} at 455, 1200, and 1950 kev and one possibly at 1045 kev. More complete data on this decay have been obtained by Hoffman and Daniels.[9] Coulomb excitation experiments by Heydenburg and Temmer[10] established a level at 455 kev which is presumably 2+. Neutron capture gamma rays of 460 kev and 605 kev have been reported by Hickok and Draper.[11]

Information on the low-lying levels in Sm^{146} has been obtained from studies of the decay of Eu^{146} (5 day)[12,13]

† Work accomplished in part under contract with the U. S. Atomic Energy Commission.

[1] V. K. Fisher, Phys. Rev. **87**, 859 (1952).
[2] G. Wilkinson and H. G. Hicks, University of California Radiation Laboratory Report UCRL-751, 1950 (unpublished).
[3] S. Ofer, Phys. Rev. **113**, 895 (1959).
[4] K. S. Toth and O. B. Nielsen, Phys. Rev. **115**, 1004 (1959).
[5] P. J. Campion, J. W. Knowles, and G. A. Bartholomew, Bull. Am. Phys. Soc. **4**, 247 (1959).
[6] *Nuclear Data Sheets*, edited by C. L. McGinnis (National Research Council), No. 59-1-113.
[7] J. K. Long, M. L. Pool, and D. N. Kundu, Phys. Rev. **88**, 171(A) (1952).
[8] W. Bernstein, S. S. Markowitz, and S. Katcoff, Phys. Rev. **93**, 1073 (1954).
[9] D. C. Hoffman and W. R. Daniels, Bull. Am. Phys. Soc. **4**, 372 (1959), and private communication.
[10] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **100**, 150 (1955).
[11] R. L. Hickok and J. E. Draper, Bull. Am. Phys. Soc. **3**, 382 (1958).
[12] N. M. Antonieva, A. A. Bashilov, B. S. Dzhelepov, and V. A. Sergienko, Nuclear Phys. **14**, 438 (1960).
[13] E. G. Funk, C. F. Schwerdtfeger, J. W. Mihelich, and B.

which indicate levels at 749 kev and 1384 kev. Sm^{146} is radioactive ($T_{\frac{1}{2}}=5\times10^7$ years),[14] and there are no Coulomb-excitation data for this nucleus.

II. PROCEDURE

The sources were produced by proton irradiation of enriched neodymium isotopes in the ORNL 86-in. cyclotron. Internal conversion electron spectra were studied with 180° permanent-magnet spectrographs. The limit of error on energy measurements from these spectrographs was about 0.2%. Gamma-ray scintillation spectra were obtained using 2 in.×2 in. NaI(Tl) crystals coupled to RCA 6342A photomultipliers and a 3 in.×3 in. NaI(Tl) crystal coupled to a Dumont 6363 photomultiplier. By utilizing the scintillation spectra of the 662-kev gamma ray of Cs^{137} and the 742-kev gamma ray of Pm^{143} the complex spectra were broken apart into individual photopeaks and Compton distributions. Gamma-ray intensities were then obtained by correcting for crystal efficiency and peak-to-total ratio.

Coincidence and directional-correlation measurements were carried out with a fast-slow coincidence circuit having a resolving time of $\tau=0.067$ μsec. The directional correlations were run in the normal double quadrant sequence. The sources were in the form of promethium chloride in hydrochloric acid. The liquid source material was contained in a cylindrical Lucite source holder $\frac{1}{8}$ in. in diameter and $\frac{3}{8}$ in. in length. Since the lifetimes of the intermediate states involved in all the correlations are presumably short, the full correlation is expected. For the Pm^{144} directional correlations, the source to crystal distance was 10 cm and data were taken every 15°. For the Pm^{146} correlation the source to crystal distance was 7 cm and data were taken every 30°. The counters were shielded frontally with $\frac{1}{8}$ in. of aluminum but no lateral lead shielding was employed since coincidences due to scattering were eliminated by differential discrimination. In all cases the real to accidental coincidence ratio was greater than 5 to 1. The data were analyzed by the least squares method as described by Rose[15] and the resulting coefficients were corrected for finite angular resolution using the results of Arns, Sund, and Wiedenbeck.[16]

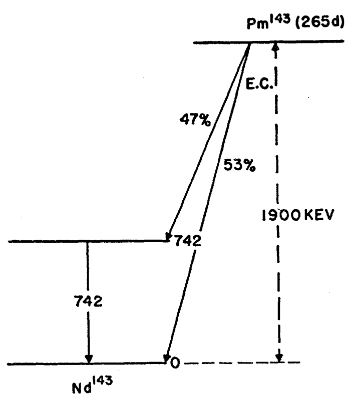

FIG. 1. Decay scheme for Pm^{143}.

The Pm^{146} beta spectrum was first studied with a $\frac{1}{4}$ in. thick by 1-in. diameter Pilot *B* plastic phosphor coupled to an EMI 9536*B* photomultiplier (energy resolution∼12% for 624-kev electrons). After completion of our intermediate-image beta spectrometer of the Slätis-Siegbahn type (momentum resolution∼2–4%), improved data were obtained.

III. RESULTS

Our results on the decay of Pm^{143} and Pm^{144} are mainly confirmatory in nature, but are included and discussed first because the source of Pm^{146} also contained these activities. Our transition energy measurements for Pm^{143} and Pm^{144} are believed to be more precise than those previously reported.

A. Pm^{143}

The source of Pm^{143} was made by a $(p,2n)$ reaction on enriched Sm^{144}, producing Eu^{143}. This in turn decayed to the long-lived Pm^{143} and the Pm was chemically extracted. The source contained less than 1% of Pm^{144}. The decay has been followed for several years giving a half-life of 265±15 days.

Our data for this activity confirm Ofer's results (see Fig. 1). The scintillation spectrum, consisting of a single photopeak at about 740 kev, is shown in Fig. 2(d). An accurate value of 741.8 kev for this transition energy was obtained from the permanent magnet spectrograph. The percentage of electron capture to the 742 kev and ground states was found to be (47±3)% and (53±3)%, respectively. These data were obtained by comparing the number of *K* x rays and 742-kev gamma rays in a scintillation counter geometry calibrated using a Cs^{137} source. Since the ratio of Ba *K* x rays to 662-kev gamma rays in the Cs^{137} source is known accurately, one may obtain a good measurement of the ratio of Pm *K* x rays and 742-kev gamma rays. Corrections were made for fluorescence yield and escape peak. A value of 7.5 was taken for the ratio of *K* capture to *L* capture to the levels in Nd^{143}. This was found from the results of Brysk and Rose[17] assuming Cameron's prediction of 1900 kev[18] for the energy difference between Pm^{143} and Nd^{143}.

B. Pm^{144}

The Pm^{144} source was produced by a $(p,3n)$ reaction on enriched Nd^{146}. About 95% of the source consisted of Pm^{144}. It contained some Pm^{143}, Pm^{145}, and Pm^{146}.

Harmatz, Bull. Am. Phys. Soc. 5, 254 (1960), and unpublished work.

[14] D. C. Dunlavey and G. T. Seaborg, Phys. Rev. 92, 206 (1953).

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

[16] R. G. Arns, R. Sund, and M. L. Wiedenbeck, privately circulated report.

[17] H. Brysk and M. E. Rose, Revs. Modern Phys. 30, 1169 (1958).

[18] A. G. W. Cameron, Atomic Energy of Canada Limited Report AECL-433, 1957 (unpublished).

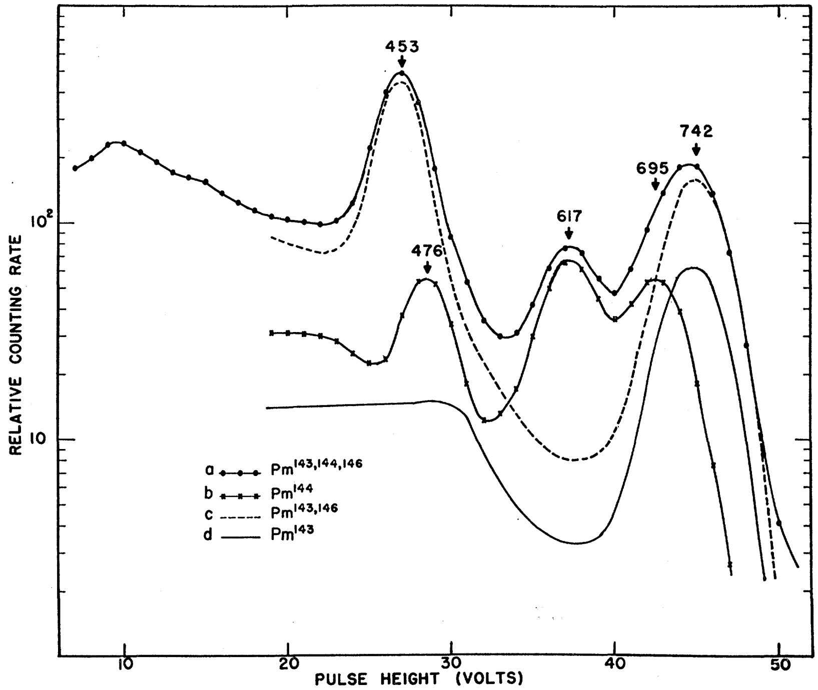

FIG. 2. Gamma-ray spectra obtained with a 2 in. by 2 in. NaI(Tl) scintillation counter. (a) Composite source consisting of Pm^{143}, Pm^{144}, and Pm^{146}; (b) Pm^{144} source; (c) composite source spectrum minus Pm^{144} spectrum; (d) Pm^{143} source.

The presence of the 18-yr Pm^{145} made a half-life measurement difficult, but the presence of the Pm^{143} allowed a comparison of Pm^{144} and Pm^{143} gamma-ray intensities at various times. The result showed that Pm^{144} has a half-life larger than that of Pm^{143}, probably of the order of 350–450 days. The Pm^{145} in the source caused no difficulty since it decays to Nd^{145} with the subsequent emission of two transitions with energies of 67 and 72 kev.[19]

The scintillation counter spectrum consisting of three prominent photopeaks at about 475, 620, and 695 kev is shown in Fig. 2(b). Relative photon intensities obtained from analysis of the spectrum are given in Table I, together with transition energies determined by the permanent magnet spectrograph. Gamma-gamma coincidence measurements showed that the 696-, 617-, and 476-kev gamma rays are in cascade and K x ray coincidences reproduced the singles curve exactly. The decay scheme of Pm^{144} is shown in Fig. 3, together with the decay scheme of Pr^{144},[6] which is included for completeness.[19a] An additional weak transition of about 210 kev was detected in the permanent magnet spectrographs and tentative evidence for a gamma-ray of about this energy was found in the scintillation spectrum and coincidence studies.

TABLE I. Transition data for the decay of Pm^{144}.

K-conversion electron energy (kev)	Transition energy (kev)	Relative photon intensity
652.4	696.0	100
573.5	617.1	100
432.1	475.7	40

[19] A. R. Brosi, B. H. Ketelle, H. Thomas, and R. J. Kerr, Phys. Rev. **113**, 239 (1959).

Directional-correlation measurements were carried out on the 617–696 kev, 476–617 kev, and 476–696 kev

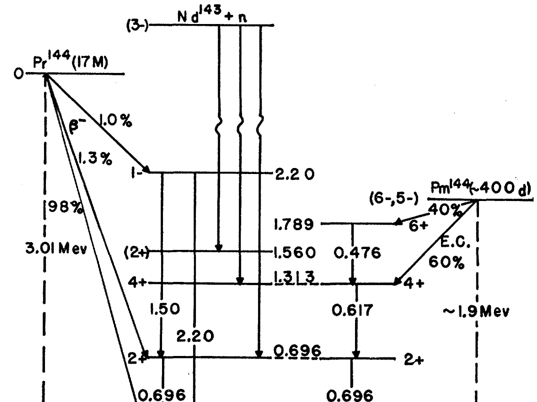

FIG. 3. Decay schemes for Pm^{144} and Pr^{144}. The Pr^{144} scheme is taken from reference 6. Neutron-capture gamma-ray data are taken from reference 5.

[19a] *Note added in proof*. The 1560 kev state in Nd^{144} has now been established as 3+, as reported in the abstracts to the Kingston Conference on Nuclear Structure, August 1960, by P. J. Campion, J. W. Knowles, and G. A. Bartholomew. No evidence was found for a low-lying second 2+ state.

TABLE II. Directional correlation results for Pm^{144}.

Correlation	Experimental coefficients A_2	A_4
617 kev–696 kev	0.0940±0.0085	0.003±0.012
476 kev–617 kev	0.1031±0.0098	0.016±0.014
476 kev–696 kev	0.0856±0.0097	−0.007±0.020
Theoretical: 4(Q)2(Q)0 or 6(Q)4(Q)2	0.1020	0.0091

cascades employing differential discrimination. The coefficients A_2 and A_4 describing the directional correlations are listed in Table II. These coefficients have been corrected for finite angular resolution. The 617–696 kev and 476–617 kev correlation results are in good agreement with the theoretical results for 4(Q)2(Q)0 and 6(Q)4(Q)2 cascades, respectively. The coefficient A_2 for the 476–696 kev correlation is slightly low for agreement with this sequence, but interference from the negative asymmetry 742–453 kev correlation in Pm^{146} could account for the discrepancy. Thus the directional correlation measurements are in agreement with the sequence of spins shown in Fig. 3 and confirm the results of Ofer.[3]

C. Pm^{146}

The source of Pm^{146} was produced by a $(p,3n)$ reaction using a target of enriched Nd^{148}. The experiment was begun after the 42-day Pm^{148} had decayed to a negligible amount. At this time the relative amounts of Pm^{146}, Pm^{144}, and Pm^{143} present were about 75%, 15%, and 10%, respectively. A large amount of 2.6-yr Pm^{147} was also present, but since only a single beta transition of 230 kev and negligible gammas ($<10^{-5}$ per disintegration) are involved in the Pm^{147} decay[20] this caused no great difficulty. This source will henceforth be referred to as the "composite" source. Due to the relatively small cross section for the $Nd^{146}(p,n)$ reaction it was not possible to produce a source of any higher isotopic purity than the source employed.

The half-life of Pm^{146} was determined to be 710±70 days. This value was obtained with an 80-mg/cm² absorber between source and Geiger counter to eliminate any contribution from the low-energy Pm^{147} beta particles.

The NaI scintillation spectrum from 200 to 800 kev for the "composite" source is shown in Fig. 2(a). No photopeaks of appreciable intensity were detected above 800 kev. When a 742-kev contour [Fig. 2(d)] was subtracted from this, a pair of peaks at 695 kev and 620 kev were obtained. The resulting spectrum was identical to that of Pm^{144} in the 600–750 kev region. When a normalized Pm^{144} spectrum [Fig. 2(b)] was subtracted from the composite spectrum [Fig. 2(a)], the spectrum shown in Fig. 2(c) was obtained. This difference spectrum appears to consist of two photopeaks at about 453 and 745 kev. As will be discussed below, the 745-kev peak was found to be triply composite. The K x-ray peak is not shown in Fig. 2, but a careful comparison of the K x-ray peak with the x-ray peak obtained from the Pm^{143} source established that more than 90% of the K x rays from the composite source were Nd x rays.

Beta spectra obtained with both a ¼-in. thick Pilot B plastic phosphor and the intermediate-image beta spectrometer indicated two beta components with end points of approximately 780 and 230 kev. The 230-kev component did not show coincidences with any of the observed photons and was attributed to the Pm^{147} present in the source.

γ-γ coincidence measurements established that the 453-kev photons coincide with photons of about 745 kev. Coincidences obtained by gating with K x rays and sweeping the gamma-ray spectrum indicated that the 453-kev photons and some of the 745-kev photons coincide strongly with K x rays as shown in Fig. 4. The number of K x ray—745-kev gamma ray coincidences was far too large to be accounted for by the Pm^{143} present in the source. On the basis of these coincidence data, it was concluded that levels of 453 and 1198 kev in Nd^{146} are populated by the electron-capture decay of Pm^{146}. These are presumably the same levels which are populated by the beta decay of Pr^{146},[8,9] with the 453-kev state being the 2+ state observed also by Coulomb excitation of Nd^{146}.[10]

From an analysis of the K x ray—γ ray coincidence data (Fig. 4) the ratio of 453- and 745-kev photons in

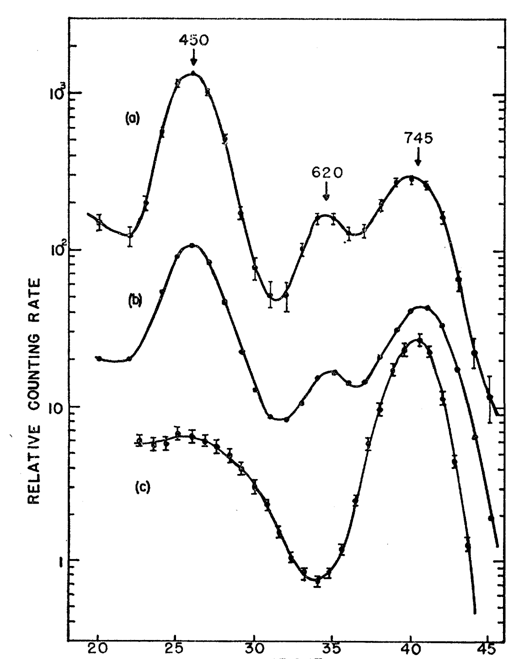

FIG. 4. Coincidence spectra obtained with Pm^{146} composite source. (a) Coincidences with K x rays; (b) singles spectrum; (c) coincidences with 500–800 kev electrons.

[20] L. M. Langer, J. W. Motz, and H. C. Price, Jr., Phys. Rev. 77, 798 (1950).

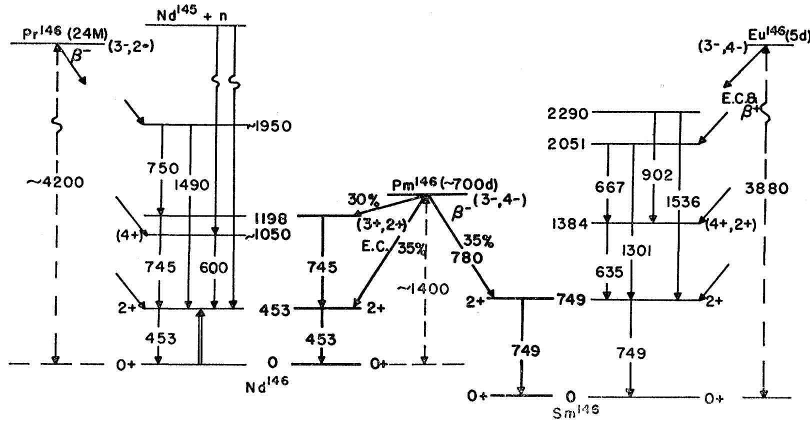

FIG. 5. Proposed decay scheme of Pm^{146}. Also shown are the partial decay schemes of Pr^{146} as proposed by Bernstein *et al.* and Hoffman and Daniels (see references 8 and 9) and Eu^{146} which is based on data taken in our laboratory (see reference 13). Energies are given in kev.

coincidence with K x rays was found to be 1.9 times their intensity ratio in singles, indicating that either the 745-kev gamma transition is fed by an electron-capture branch with a low K-capture to L-capture ratio or that an appreciable precentage of the 745-kev transitions follow the beta decay of Pm^{146}. Beta-gamma coincidence measurements carried out with Pilot B and NaI scintillation counters confirmed the latter hypothesis. Coincidences with electrons of energy between 500 and 800 kev showed a spectrum consisting of a single gamma ray of about 750 kev (Fig. 4), and a scintillation counter coincidence spectrum gating with the gamma-ray counter on the 745-kev photopeak yielded a beta spectrum with an end point of about 780 kev.

Hence, Pm^{146} decays both by electron capture to levels of 453 and 1198 kev in Nd^{146} and by beta decay to a level of 749 kev in Sm^{146}. The 749-kev level is presumably the first excited state in Sm^{146} which is also populated by the decay of Eu^{146}.[13] The proposed decay scheme is presented in Fig. 5. For completeness, the partial decay schemes of Pr^{146} [8,9] and Eu^{146} [13] are shown also in Fig. 5.

Two weak internal conversion electron lines were detected on the permanent magnet spectrograph films. The first, at an electron energy of 409.1 kev, was the K line for a 452.7-kev transition in Nd, and the other, at an electron energy of about 700 kev, was probably a composite K line due to the 745-kev transition in Nd^{146} and the 749-kev transition in Sm^{146}. The latter energy has been measured accurately as 748.8 kev from a study of the decay of Eu^{146}.[13]

From analysis of the singles spectrum shown in Fig. 2(c) the ratio of the intensities of ~745-kev and 453-kev photons from the composite source was determined to be 1.05±0.05. The 745-kev photopeak was of course due to three gamma transitions: the 742-kev transition in Nd^{143}, the 745-kev transition in Nd^{146} and the 749-kev transition in Sm^{146}.

A search for higher energy gamma rays (>1 Mev) with a 3 in.×3 in. NaI crystal showed no peaks other than addition peaks due to summing of two coincident gamma rays in the crystal. An upper limit for the intensity ratio of a possible 1198-kev cross-over transition to the 745-kev transition in Nd^{146} was found to be 0.003.

A calibrated-coincidence measurement was carried out to determine the electron-capture branching ratios to the 1198- and 453-kev states in Nd^{146}. Two 2 in.×2 in. NaI scintillation counters were placed at an angle of 90° to one another and the source positioned at a distance of 5 cm from each counter. Aluminum frontal shields ($\frac{1}{8}$ in.) and lateral lead shields (15 g/cm^2) were employed. Coincidence spectra were run with both the Pm^{144} and the composite Pm^{146} sources, in each case gating on the region of 450–475 kev. The coincidence spectra are shown in Fig. 6. The Pm^{144} source provided a calibration of the geometry since the decay scheme and the relative gamma intensities for Pm^{144} are known. By comparing the intensity of the 745-kev coincidence peak (due to Pm^{146}) with the intensity of the 696-kev coincidence peak in Pm^{144}, after normalization for the number of pulses in the gate, the ratio of the intensities of 745-kev and 453-kev photons following electron capture of Pm^{146} was found to be 0.46±0.05. An identical ratio was obtained by comparing the 696- and 745-kev peaks in the coincidence spectrum of the "composite" source alone, knowing the relative number of gate pulses which were due to 453-, 476-, and 617-kev photons. No correction for angular correlation effects was made since the solid angle subtended was rather large (~6%) and the asymmetries of the angular correlations involved are less than 17%. Thus, the 1198-kev state in Nd^{146} is fed by 46% of the electron-capture decays and the 453-kev state by 54% of the electron-capture decays if one assumes no decay to the ground state of Nd^{146}.

The beta spectrum above 300 kev obtained with the intermediate image spectrometer is shown in Fig. 7. The strongest available source strength was about 0.05 microcurie. The source was deposited on a 1-mg/cm^2 aluminized-Mylar backing. The diameter of the source was 3 mm and the source thickness about 2 mg/cm^2. Under these conditions, the momentum resolution of the spectrometer was 3.6% as determined from the 453-kev K-conversion peak.

A Fermi analysis was carried out using only those

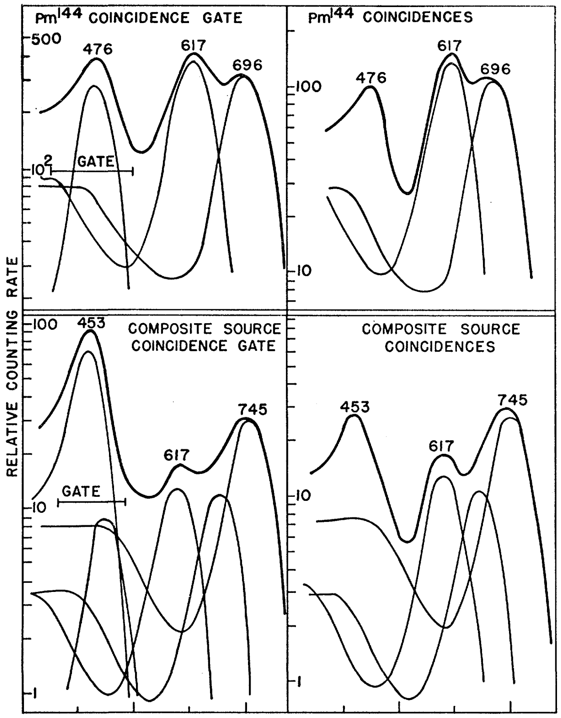

FIG. 6. Results of calibrated coincidence measurements using sources of Pm^{144} and the composite source. Left half of figure shows the gates in each case and the right-hand portion the coincidence spectra. Heavier lines indicate observed spectra, lighter lines the individual gamma-ray components.

points between the internal-conversion electron peaks. The results are shown in Fig. 8. The data were least-squares fitted to a straight line and an end point of 779±15 kev obtained. Because of the relatively few experimental points and the large statistical errors associated with these points, it is difficult to rule out the possibility of a forbidden shape for this component. This measured end-point energy leads to an energy difference of 1528±15 kev between Pm^{146} and Sm^{146}, as compared to the value of 1660 kev predicted by Cameron's empirical mass formula.[18]

Assuming an allowed shape for the 779-kev beta spectrum, the ratio of the intensities of the 779-kev beta spectrum and 453-kev K-conversion electrons can be obtained by comparison of the area under the beta spectrum to the area of the 453-kev conversion peak. This ratio is found to be 44. Using the theoretical value of 0.0125 for the K-conversion coefficient for a 453-kev transition obtained from Rose's tables,[21] a value of 0.35±0.02 is found for the ratio of 779-kev beta transitions to total 453-kev transitions. Since there is no observed beta-decay branch to the 0+ ground state of Sm^{146}, it is likely that there is no electron-capture decay to the 0+ ground state of Nd^{146}. If one assumes no ground-state electron-capture branch, the percentage beta branching for the Pm^{146} decay is 35% and the electron-capture branchings to the 1198- and 453-kev states in Nd^{146} are 30% and 35%, respectively.

The K x ray-gamma ray coincidence data served as a check on the branching ratios for the decay of Pm^{146}. As was previously mentioned, the ratio of the intensities of 453- and 745-kev gamma rays in coincidence with K x rays was found to be 1.9±0.1 times their intensity ratio in singles. If the K- to L-capture ratio were approximately the same for the electron-capture branches to both states in Nd^{146} and for the Pm^{143} electron capture, this result would imply that 47±5% of the ~745-kev gamma rays from the composite source follow the beta decay. This, in conjunction with the fact that the ratio of total "745"-kev photon intensity to 453-kev photon intensity is 1.05±0.05, leads to a beta branching ratio of 33±3%, in agreement with the result of 35±2% obtained from the beta spectrometer data. We are assuming no ground-state electron-capture branch.

The energy available for electron-capture decay of Pm^{146} is predicted by Cameron to be 897 kev.[18] Therefore, the actual available energy is probably not more than a few hundred kev greater than 1198 kev, the energy of the highest state populated by electron capture. One might expect considerable L capture to this state. The inaccuracy of the K x ray peak intensity due to the isotopic complexity of the source made a direct determination of the K to L-capture ratio impossible. However, no appreciable L x ray peak was observed with a 2-mm NaI crystal, indicating that there was probably no strong L-capture branch.

To determine the effect of the relative amount of L capture on the deduced branching ratios, the following analysis was carried out: let us call $\mathbf{x}$ the ratio of K- to L-electron capture in the decay to the level of 1198 kev in Nd^{146}, $\mathbf{y}$ the fractional decay of Pm^{146} to the 749-kev level of Sm^{146}, and $\mathbf{z}$ the ratio of electron capture to the 1198- and 453-kev levels in Nd^{146}. We assume a K- to

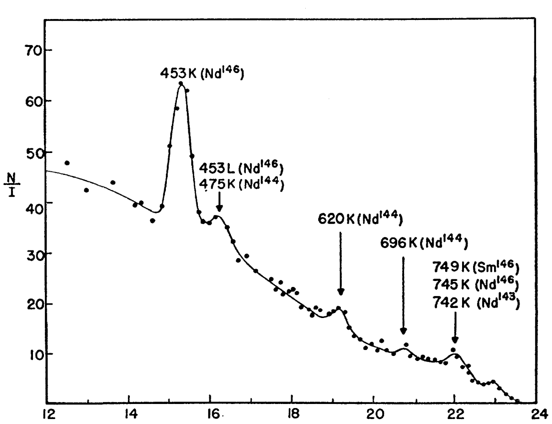

FIG. 7. Pm^{146} beta spectrum obtained with an intermediate-image spectrometer (momentum resolution = 3.6%).

[21] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

L-capture ratio of 7 to 1 for the decay to the 453-kev state and for the Pm^{143} decay. One may define a ratio

$$R(\mathbf{x},\mathbf{y},\mathbf{z})=\frac{(453/745)_{K\text{ x coincidence}}}{(453/745)_{\text{singles}}}$$

with the numerator being the ratio of the intensities of 453- and ~745-kev photons in coincidence with K x rays, and the denominator being the ratio of these photon intensities in singles. This ratio R can be expressed in terms of the quantities **x**, **y**, and **z**. The result of the calibrated coincidence measurement gives **z** directly, and R is determined experimentally to be 1.9±0.1 as discussed previously. Thus we may derive an expression relating **y** to **x**. For values of **x** equal to 6, 1, 0.1, and 0, the values of **y**, the beta branching ratio, are found to be 33±3%, 30±3%, 29±3%, and 26±3%, respectively. These K- to L-capture ratios correspond to electron-capture energies of approximately 200 kev, 65 kev, 50 kev, and <44 kev as determined from the theoretical predictions of Brysk and Rose.[17]

It can be seen that the deduced beta branching ratio is not very sensitive to the amount of L capture to the 1198-kev state. Considering all the experimental data and Cameron's predicted energy, it is most probable that the total energy difference between Pm^{146} and Nd^{146} is between 1260 and 1500 kev.

From the electron spectrum shown in Fig. 7, the ratio of K-conversion electrons from the ~745-kev transitions and the 453-kev transition was found to be 2.55±0.25. Since the branching ratios and the relative numbers of ~745-kev transitions in the triply composite gamma-ray peak have now been determined, one can obtain the K-internal conversion coefficient for the 745-kev transition in Nd^{146}. Using Rose's theoretical K-conversion coefficients[21] for the 749-kev $E2$ transition in Sm^{146}, the 742-kev $M1$ transition in Nd^{143},[3] and the 453-kev $E2$ transition in Nd^{146}, one obtains a value of 0.0055±0.0011 for the K-conversion coefficient of the 745-kev transition. The theoretical values for various multipole orders are[21]: $E1$ (0.0014), $E2$ (0.0036), $M1$ (0.0059), and $M2$ (0.0160). The experimental conversion coefficient is consistent with the 745-kev gamma ray being pure $M1$ or a mixture of $M1$ and $E2$ with a quadruple content less than 65%. It is also compatible with an $E1-M2$ mixture with a quadrupole content of between 18 and 36%.

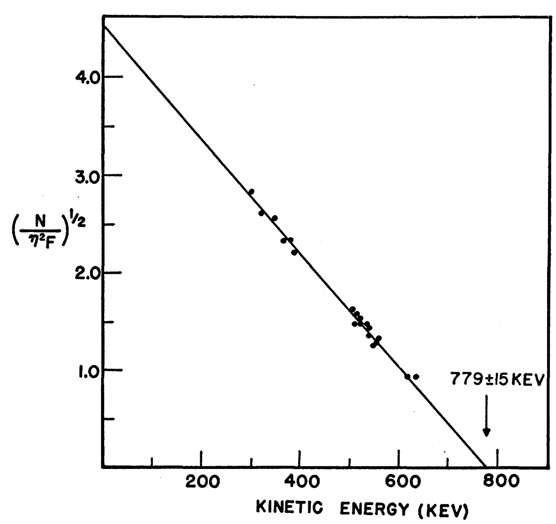

FIG. 8. Fermi plot for Pm^{146} beta spectrum.

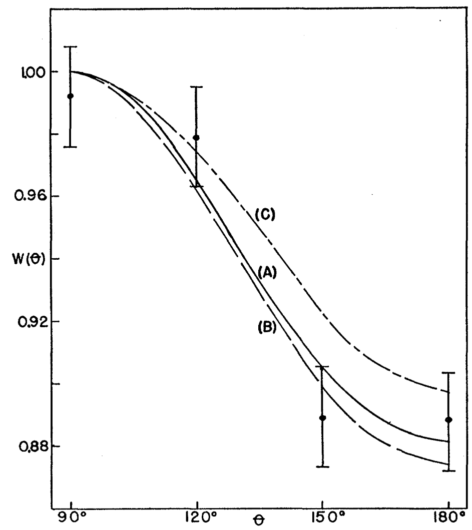

FIG. 9. Angular correlation data for 745 kev-453 kev gamma-ray cascade in Nd^{146}. (A) Observed correlation function, after correction for finite angular resolution of detectors. (B) Correlation function after correction for interference from Pm^{144} gamma-ray cascades. (C) Theoretical curve for a $3(D)2(Q)0$ sequence.

A directional correlation measurement was carried out on the 745–453 kev gamma cascade in Nd^{146} and the resulting curve is shown in Fig. 9. Because of the Pm^{144} present in this source the correlation included a contribution from the 696–476 kev and 696–617 kev correlations in Nd^{144}. With the differential discriminator set on the high side of the 745-kev peak this contribution was kept to about 2%. The coefficients for this correlation, after correction for finite angular resolution and interference from the known Nd^{144} correlations, are $A_2=-0.091\pm0.025$ and $A_4=0.008\pm0.050$. These experimental coefficients are consistent with a $3(D)2(Q)0$ sequence for which the theoretical values are $A_2=-0.071$ and $A_4=0$. If one considers a dipole-quadrupole mixture in the 745-kev transition, a limit of less than 0.05% quadrupole is obtained from the mixture curves of Arns and Wiedenbeck.[22] The coefficients are also in agreement with a $2(D,Q)2(Q)0$ sequence with a quadrupole content of 18±2% and a sequence of $1(D,Q)2(Q)0$ with a quadrupole content of 2±1%.

The possible spins and parities for the 1198-kev state and the corresponding multiple orders for the 745-kev transition which are consistent with both the angular

[22] R. G. Arns and M. L. Wiedenbeck, privately circulated report.

TABLE III. Possible spin and parity values for 1198-kev state of Nd^{146} and multipolarity of 745-kev transition.

1198-kev state I, π	Multipolarity of 745-kev transition
3+	Pure $M1$ or $M1-E2$ mixture with <0.05% $E2$
2+	82% $M1$, 18% $E2$
1+	98% $M1$, 2% $E2$
2−	82% $E1$, 18% $M2$

TABLE IV. Log ft values for Pm^{146} decay.

Type of decay	Transition energy (kev)	Log ft
Beta decay	779	9.7
		$\log[(W_0^2-1)ft]=10.4$
Electron-capture decay		
Available energy≅1400 kev	945	9.0
	200	7.6
Available energy≅1300 kev	845	8.9
	100	7.0

correlation and conversion coefficient results are listed in Table III. The apparent absence of a cross-over transition of 1198 kev would tend to rule out a 1+ assignment. A 2− assignment would be very unusual since low-lying 2− states have not been observed in even-even nuclei. A 3+ assignment is favored over the 2+ because of the absence of the cross-over transition and the relatively small $E2$ admixture (~18%) in the 745-kev transition. In the observed cases where the cross-over transition from the second 2+ state to the 0+ ground state is extremely weak compared to the 2+ → 2+ transition, the 2+ → 2+ transition contains a large quadrupole content (>90%).[23]

Log ft values for the electron-capture and beta-decay branches were obtained from Moszkowski's graphs,[24] using the experimentally determined branching ratios and a half-life of 710 days for Pm^{146}. The resulting log ft values are presented in Table IV. Values for the electron-capture branches are given for two possible $Pm^{146}-Nd^{146}$ energy differences, 1300 kev and 1400 kev.

The spin and parity of the Pm^{146} ground state should be either 3− or 4− since: (1) no beta transition to the 0+ ground state of Sm^{146} is observed, and (2) levels of 2+ and 3+ in Nd^{146} and Sm^{146} are populated by transitions with log ft values in the first-forbidden range, requiring a spin change of 0, 1, or 2 and a change of parity. The shell model predicts a negative parity and a spin of between 1 and 6 for the odd-odd nucleus Pm^{146} since the 85th neutron is most likely in an $f_{7/2}$ state and the 61st proton in a $d_{5/2}$ state.

If the Pm^{146} ground state were 4−, the 779-kev beta branch and the high-energy electron-capture branch would be unique first-forbidden transitions. This would require the beta-spectrum Fermi plot to exhibit the characteristic α shape. As mentioned previously, the experimental data are not precise enough to determine whether the Fermi plot deviates from linearity. The values of $\log ft=9.7$ and $\log[(W_0^2-1)ft]=10.4$ for the 779-kev beta branch and the log ft of about 9 for the high-energy electron-capture branch are consistent with the expected values for unique first-forbidden transitions, but it is also quite possible that these are ordinary first-forbidden transitions with slightly high log ft values. Consequently, no choice can be made between the possible 3− or 4− assignments for Pm^{146}.

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IV. DISCUSSION

The decay schemes of Pm^{143} and Pm^{144} have been discussed in detail by Ofer,[3] Toth and Nielsen,[4] and Mallmann and Porter.[25] Therefore, this section will be concerned only with a discussion of a few interesting aspects of the decay scheme of Pm^{146} and a consideration of the energy level systematics for even-even nuclei having neutron numbers from 84 to 88.

It is of interest to consider why the electron-capture decay of Pm^{146} does not populate the ~1050-kev level in Nd^{146}. This state has been observed in investigations of the decay of Pr^{146},[9] and in neutron-capture gamma-ray studies.[11] The spin of this state is given as (4+) on the nuclear data sheets.[6] No firm experimental evidence is available for this assignment, but the absence of a cross-over transition of ~1050 kev indicates that the spin is probably 4+ and not 2+. Furthermore, a 4+ second excited state is observed in Sm^{148},[26] a nucleus having the same neutron number as Nd^{146} ($N=86$), and most of the other even-even nuclei in this mass region are observed to have 4+ second excited states. (Energy level data for even-even nuclei with $N=84$ to 88 are presented in Table V.[5,13,9,26–29])

An electron-capture transition to the presumed 1050-kev 4+ state must have a log ft value greater than about 9.5 or we would have detected the presence of the 600-kev gamma transition in both the gamma-ray scintillation spectrum [Fig. 2(c)] and the 453-kev gamma ray coincidence spectrum (Fig. 6). For a log ft of 9.5, the branching ratio for electron-capture decay to the 1050-kev level would be about 1% and the intensity of the 600-kev gamma transition approximately 2% of the intensity of the 453-kev gamma transition.

If the spin and parity of the Pm^{146} ground state are 3−, the 779-kev beta transition and the electron-capture transitions to the 453-kev, 1050-kev, and 1198-

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kev states would be ordinary first-forbidden transitions. Since the experimental log *ft* values are 9.7, 9.0, >9.5, and 7.6, respectively, the first three transitions must be retarded while the transition to the 1198-kev level is normal. If the spin and parity of the Pm^{146} ground state are 4−, the first two transitions mentioned above would have to be unique first-forbidden and the other two, ordinary first-forbidden. For this case, the log *ft* values indicate that the only retarded transition would be that to the 1050-kev state in Nd^{146}. In either case, the electron-capture transition to the 1050-kev state in Nd^{146} must be retarded by a factor of 100 or more. This could possibly be due to some selection rule analogous to the *K* forbiddenness observed in the neighboring deformed nuclei.[30]

The fact that a beta transition was not observed to the 1384-kev Sm^{146} state (4+ or 2+) is readily explained. The available energy is only 144 kev as compared to 779 kev for the transition to the 749-kev state, and one would expect only a 1% branch to the 1384-kev state if the log *ft* were as low as 8.[24]

An interpretation of the energy levels of Nd^{146} in terms of one of the proposed nuclear models[31,32] is not presently feasible because relatively few states are populated by the Pm^{146} decay. Results of Coulomb excitation experiments show that the reduced transition probability for the 453-kev *E*2 transition is about 11 times the single-particle estimate,[33] indicating that the 453-kev state may be of collective nature. The 3+ level at 1198 kev may be an intrinsic state rather than collective since the 745-kev transition is almost pure *M*1, and one might expect a large *E*2 admixture if the state were due to a collective excitation.

As seen in Table V, most (and possibly all) of the even-even nuclei with $N=84$, 86, or 88 are observed to have a 4+ second excited state. The ratio of the energy of the first 4+ state to the energy of the first 2+ state lies between 1.85 and 2.35. For the transition region of mass number 192 to 200, the second excited state is found to be 2+ in most of the observed cases, with the ratio of the energies of the second 2+ state and the first 2+ state being about 2.[6]

Table V. Available data on low-lying states of even-even nuclei with $N=84$, 86, and 88.

N	Nuclide	E_2^1 [a]	E_4^1	E_2^2	E_4^1/E_2^1	E_2^2/E_2^1	References
84	$_{58}Ce^{142}$	630		(1500)[b]		(2.38)	27
	$_{60}Nd^{144}$	696	1313		1.89		This work, 3
	$_{62}Sm^{146}$	749	(1384)		(1.85)		13
86	$_{60}Nd^{146}$	453	(1060)		(2.34)		This work, 9
	$_{62}Sm^{148}$	551	1181		2.14		26
88	$_{62}Sm^{150}$	340	(780)		(2.29)		28
	$_{64}Gd^{152}$	344	(757)		(2.19)		29

[a] E_2^1, E_4^1, and E_2^2 are the energies (in kev) of the first 2+ state, first 4+ state, and second 2+ state, respectively.
[b] Parentheses indicate that the spin is not certain.

Some success has been achieved in describing the low-lying excited states for nuclei in the $A=192$ to 200 region either by quadrupole vibrations about a spherical equilibrium shape[31] or by the asymmetric rotor model.[32,34] The Davydov-Filippov asymmetric rotor model[32,34] predicts that the ratio of the energies of the second excited state and the first 2+ excited state should never be less than 2 and that the ratio of the energies of the first 4+ state and the first 2+ state should not be less than 2.67. This model therefore fails for some of the even-even nuclei in the $N=84$ to 88 region. An asymmetric rotor model currently being developed by Mallmann[35] overcomes these two difficulties and may possibly provide an explanation for the level schemes in the transition regions as well as for many other nuclei.

It is hoped that further studies of the level schemes of nuclei with neutron number 84 to 88 will provide sufficient data to test the usefulness of proposed nuclear models for this region.

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