

being here defined as the number of coincidences in the pulse-height range corresponding to gamma-ray energies from 1.6 to 5.5 Mev in the center crystal per unit incident flux. Relative values for the response to 4.4-Mev gamma rays, to 14-Mev neutrons, and to 2.5-Mev neutrons were found to be 1.0, 0.08, and 0.008 respectively. From these measurements and available data on 14-Mev neutron elastic and inelastic scattering, it is estimated that the effect on the pulse-height distribution due to neutrons scattered by the sample into the detector is not more than a few percent for the geometries used.

A thick zirconium-tritium target bombarded by 130-kev deuterons from a Cockcroft-Walton accelerator provided the 14-Mev neutrons whose source strength was measured by a calibrated long counter. The long counter readings were corrected for the effect of neutron scattering by the graphite sample which was in the

form of a ring and located so that the plane of the ring lay immediately behind the target. The spectrometer was placed 26 cm in front of the target, a tungsten bar providing shielding of the center crystal from the direct neutron beam.

Figure 1 shows the pulse-height distribution, scatterer in minus scatterer out, obtained for 14-Mev neutrons incident on carbon. The  $\times$  points represent the net result after correction for radiative loss and electron escape obtained by normalization from the Pu-Be data, and the curve drawn shows the corrected pulse-height distribution. This distribution is consistent with the assignment of a single-energy line at 4.4 Mev in the 1.6–5.5 Mev region. The calculated value for the production of 4.4-Mev gamma rays from 14-Mev neutron bombardment of C<sup>12</sup>, taking into account the attenuation of incident neutrons and resulting gamma rays in the scatterer, is  $245 \pm 35$  millibarns.

## Coincidence Studies of the Disintegration of Pm<sup>151</sup> and Nd<sup>147</sup>†

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Chemically pure Nd<sub>2</sub>O<sub>3</sub> was irradiated by slow neutrons on five successive occasions in the Brookhaven pile. The 27.5-hour Pm<sup>151</sup> was found to emit gamma rays of energies 64, 100, 163, 177, 240, 275, 340, 440, 650, and 700 kev. The 11-day Nd<sup>147</sup> was shown to emit quanta of energies 92, 165, 280, 320, 410, 440, 530, and 690 kev. The relative intensities of the various quantum radiations have been determined, and coincidence measurements have been performed to ascertain the various sequential relationships between pairs of gamma rays. Partial decay schemes for both radionuclides have been indicated.

### INTRODUCTION

IT was originally reported by Law, Pool, Kurbatov, and Quill<sup>1</sup> that 2.3-hr, 47-hr, and 11-day activities can be produced when neodymium is irradiated by certain nuclear particles. The mass number of the 11-day activity has been shown to be 147.<sup>2,3</sup> The 2.3-hr activity has been identified as the neodymium parent of the 47-hr promethium,<sup>2,4</sup> the mass numbers being 149.<sup>5</sup> Beta-gamma coincidence studies, and absorption measurements were carried out by Mandeville *et al.*<sup>6</sup> to establish the principal features and general pattern of

the decay schemes of the 11-day neodymium and the 50-hr<sup>7</sup> Pm<sup>149</sup>. Kondaiah<sup>8</sup> has reported in the decay of the 11-day activity gamma rays of energies 520, 391, 309, and 91 kev and three beta-ray spectra having end points at 350, 470, and 780 kev. This author found no gamma rays to be emitted in the decay of the 50-hr Pm<sup>149</sup>. Emmerich and Kurbatov<sup>9</sup> have investigated the disintegration of the 11-day activity and report gamma rays at 91.5, 320, and 534 kev and beta-ray energies of 380, 600, and 825 kev. More recently, Cork *et al.*<sup>7</sup> have reported for Nd<sup>147</sup> gamma rays of energies 91.2, 120.5, 168.1, 197.1, 231.2, 259.8, 273.3, 300.8, 318.1, 398.4, 441.4, and 532.3 kev. These latter authors have also reported the presence of Pm<sup>151</sup> ( $T_{1/2} = 27.5$  hr) in samples of irradiated Nd which had been enriched in Nd<sup>150</sup>. This activity is apparently grown from the 12-minute Nd<sup>151</sup>. Pm<sup>151</sup> was reported<sup>7</sup> to emit gamma rays of energies 64.7, 65.8, 69.6, 100.0, 105.2, 116.2, 144.0, 163.1, 168.0, 177.1, 208.3, 231.9, 239.9, 275.2, 340.2, and 715 kev.

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<sup>1</sup> Law, Pool, Kurbatov, and Quill, *Phys. Rev.* **59**, 936 (1941).

<sup>2</sup> Marinsky, Glendenin, and Coryell, *J. Am. Chem. Soc.* **69**, 2781 (1947).

<sup>3</sup> R. J. Hayden, *Phys. Rev.* **74**, 650 (1948).

<sup>4</sup> J. A. Marinsky and L. E. Glendenin, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 7.543, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, 1264 (1951).

<sup>5</sup> Ingram, Hess, and Hayden, *Phys. Rev.* **71**, 743 (1947).

<sup>6</sup> C. E. Mandeville and E. Shapiro, *Phys. Rev.* **79**, 391 (1950); *Proc. Nat. Inst. Sci. India* **17**, No. 1, 45 (1951); C. E. Mandeville and M. V. Scherb, *Phys. Rev.* **76**, 186 (1949).

<sup>7</sup> Rutledge, Cork, and Burson, *Phys. Rev.* **86**, 775 (1952).

<sup>8</sup> E. Kondaiah, *Phys. Rev.* **81**, 1056 (1951).

<sup>9</sup> W. S. Emmerich and J. D. Kurbatov, *Phys. Rev.* **83**, 40 (1951).

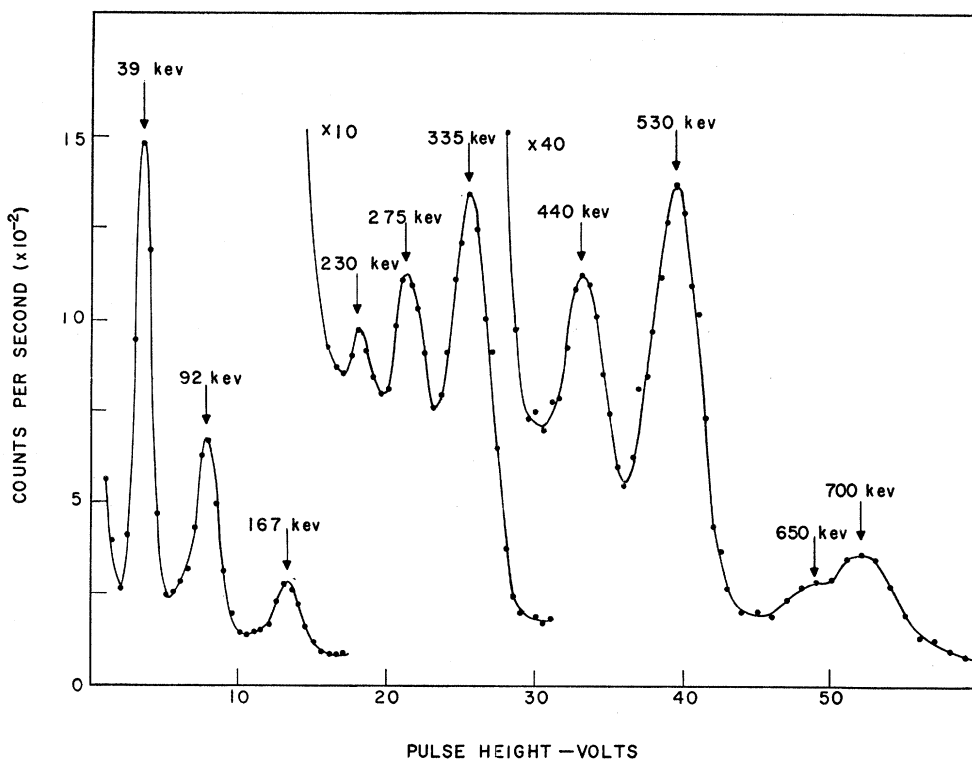


FIG. 1. Spectrum of gamma rays emitted in the decay of  $\text{Nd}^{147}$ ,  $\text{Pm}^{149}$ , and  $\text{Pm}^{151}$  sixty hours after cessation of irradiation.

In none of the measurements described above were scintillation counting methods employed in coincidence studies of the decay of the several radionuclides of neodymium. Consequently, such investigations were undertaken for the purpose of obtaining additional information concerning the decay schemes in question. In all, five successive exposures of samples<sup>10</sup> of very pure  $\text{Nd}_2\text{O}_3$  were carried out in the Brookhaven pile. The times of irradiation varied from two hours to two weeks in length, and measurements were usually commenced within a few hours after the target material had been removed from the reactor.

#### $\text{Pm}^{151}$

For the study of the radiations of the relatively short-lived activities of neodymium, exposure times of a few hours were employed so as to suppress any contributions of the 11-day period. The gamma-ray spectrum of  $\text{Pm}^{149}$ – $\text{Pm}^{151}$  is shown in Fig. 1. The spectrum was observed to decay for the most part with a half-life of 27.5 hours. Only in the vicinity of 280 keV were any contributions of an activity of half-period intermediate between 27.5-hr and 11-days in evidence. After four days of observations, it became apparent that in this spectral region, decay was proceeding at a somewhat slower rate than could be associated with a 27.5-hr

period. Photopeaks are seen to occur which correspond to quantum energies of 39, 92, 167, 230, 275, 335, 440, 530, 650, 700, and  $\sim 1500$  keV (not shown). From the measurements of Cork *et al.*<sup>7</sup> and from results to be subsequently discussed in this paper, it is clear that some of the above-listed peaks are composite ones corresponding to more than one gamma ray. In particular, observations about one week after cessation of irradiation showed that the radiations of the 11-day activity contributed appreciably only to the photopeaks at 92 keV and 530 keV. Of the full energy peaks observed in the spectrum of Fig. 1, the decay of the ordinate values related the following to the 27.5-hour period: 167, 230, 275, 335, 440, 650, and 700 keV with relative intensities of 70, 42, 55, 100, 40, 18, and 26.

In order to ascertain the complexity of the various peaks, coincidence measurements were undertaken. With the axes of two scintillation counters at  $90^\circ$ , a strong coincidence rate was observed when the channel of a differential pulse-height analyzer used in conjunction with either detector was placed at  $\sim 167$  keV. Thus, at least two quanta in coincidence of energies about 167 keV are emitted in the decay of the 27.5-hour activity.

Coincidences between 230-keV quanta and the remainder of the spectrum are shown in Fig. 2 where photopeaks in the coincidence rate occur at 64 and 100 keV. Similarly, these two quanta were found to be

<sup>10</sup> Five grams of  $\text{Nd}_2\text{O}_3$  were kindly supplied by Dr. F. H. Spedding of the Institute for Atomic Research, Iowa State College, Ames, Iowa.

coincident with the 275-keV gamma ray as shown in Fig. 3A. In Fig. 3B are shown photopeaks of the two gamma rays of energies 39 and 100 keV which are coincident with gamma rays of energies greater than 530 keV.

In addition to the data depicted in Figs. 2 and 3, several other coincidence measurements were performed. With the channel of one analyzer fixed at 100 keV, the other was moved through the region of 275 keV. No 275-keV gamma rays were found in coincidence with the 100-keV radiation. The coincidences at 100 keV shown in Fig. 3A must then arise from Compton recoils of high-energy gamma rays coincident with the gamma ray at 100 keV. A curve of coincidences was also obtained by fixing the channel of one pulse height analyzer at 100 keV and moving the channel of the other through the energy interval about 167 keV. A pronounced peak appeared, but when the reverse procedure was followed of fixing at 167 keV and moving through 100 keV, no definite peak appeared. From these measurements it was concluded that the 100 keV–167 keV coincidences arose from coincidences between the 100-keV gamma ray and backscattered quanta deriving from the hard gamma rays of energies greater than 530 keV. Still another coincidence experiment was carried out when the channel of one analyzer was placed at 64 keV and the channel of the other moved through the region of 275 keV. In this case, genuine coincidences were distinctly observed. This peak of coincidences was so broad as to suggest the 64-keV radiation to be coincident with the interval of quantum energies extending from 230 keV to 280 keV. This conclusion is partially borne out by the data of Fig. 2. No two gamma rays of energies greater than 230 keV were found to be in coincidence.

The various coincidence studies of the preceding paragraphs combined with the spectrometric data of Cork *et al.*<sup>7</sup> suggest the decay scheme of Fig. 4. For example, the intense coincidence rate observed in the

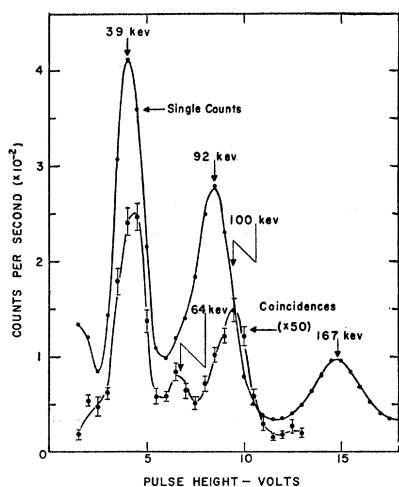


Fig. 2. Spectrum of gamma rays coincident with the 230-keV quanta of  $\text{Pm}^{151}$ .

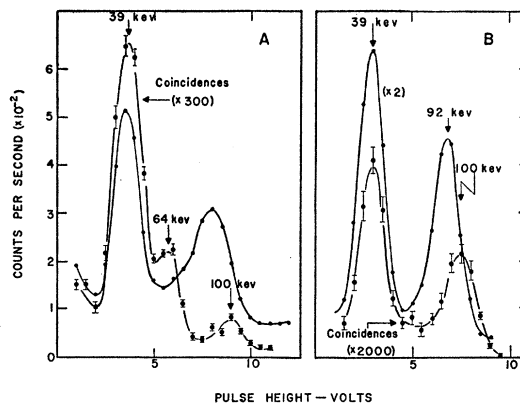


Fig. 3. A, spectrum of gamma rays coincident with the 275-keV gamma ray of  $\text{Pm}^{151}$ . B, spectrum of gamma rays coincident with quanta of energies greater than 530 keV.

present investigation between gamma rays of energies about 167 keV is assumed to arise from the quanta reported<sup>7</sup> at 163 and 177 keV. The 275 keV–65 keV cascade and the associated cross-over transition at 340 keV and the 240 keV–100 keV cascade are also consistent with the data of Cork *et al.*<sup>7</sup> Only in the case of the 240 keV–100 keV cascade is the order of emission of the gamma rays known. This particular sequence was established by a consideration of the fact that coincidences were found between the 100-keV radiation and the hard gamma rays of energies greater than 530 keV. The weak hard quanta at 1500 keV were previously assigned<sup>7</sup> to the 50-hr activity. In the present instance, this hard gamma ray was observed to decay with a half-life of 27.5 hours. After obtaining the data of Fig. 3B, the channel of the analyzer selecting pulses of higher energies was moved through the interval extending from 530 keV to 750 keV. Coincidences were detected for gamma-ray energies greater than 530 keV and less than 650 keV. Transitions from the 715- and 650-keV levels to the 100-keV level could account for them. These

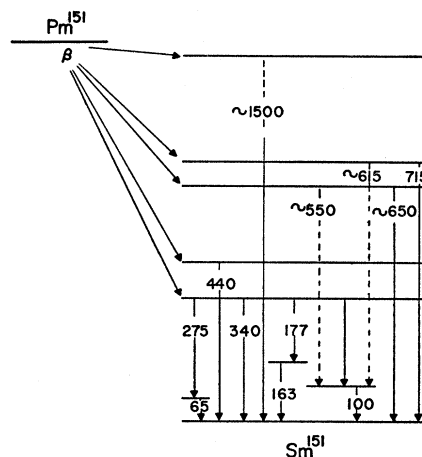


Fig. 4. Nuclear energy levels of  $\text{Sm}^{151}$ .

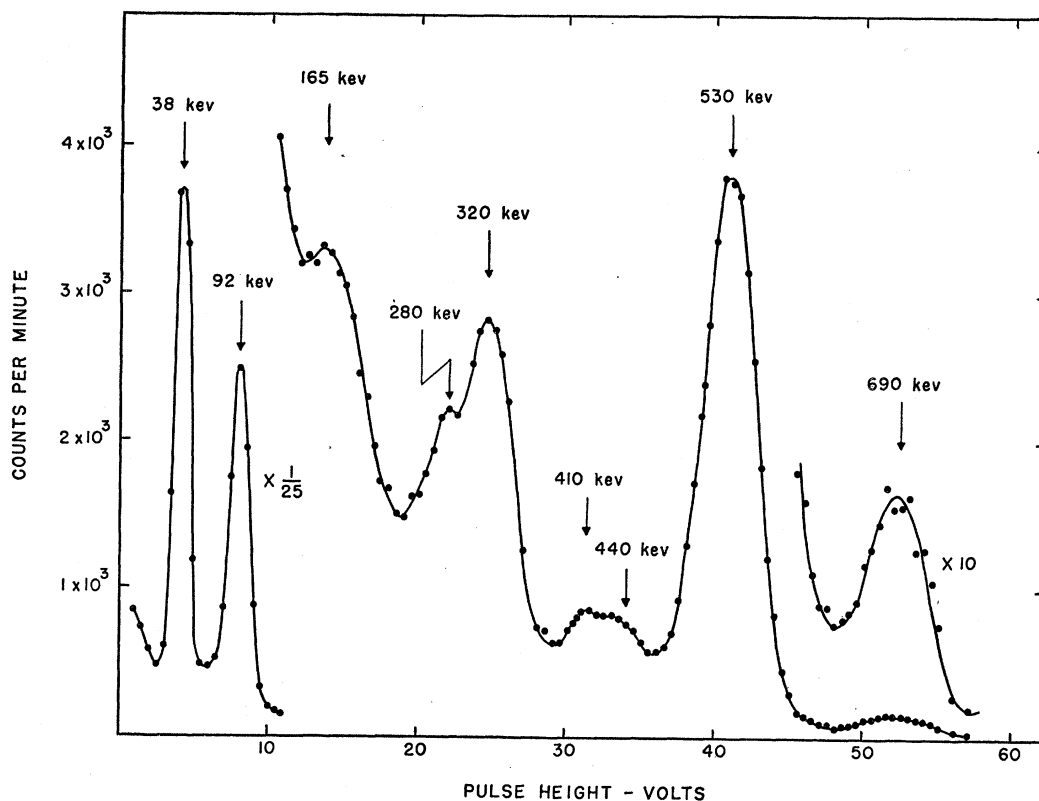


Fig. 5. Spectrum of gamma rays emitted in decay of  $\text{Nd}^{147}$ .

implied transitions are indicated by broken lines in Fig. 4.

Because of the poorer resolution of scintillation spectroscopy, it was not possible to account for all the gamma rays detected in the magnetic spectrometers.

#### $\text{Nd}^{147}$

The spectrum of the gamma rays emitted in the decay of the 11-day  $\text{Nd}^{147}$  is shown in Fig. 5 where peaks corresponding to quantum energies of 92, 165, 280, 320, 410, 440, 530, and 690 keV are indicated with relative intensities of 55, 1.5, 2.5, 4.5, 3.0, 3.5, 25, and 1.5. Earlier results<sup>7-9</sup> show that the regions of quantum energies about 165 keV and 320 keV contain contribu-

tions in either case from several gamma rays, more than indicated by the scintillation spectrometer. Cork *et al.*<sup>7</sup> report seven quanta in the entire energy interval extending from 165 keV to 320 keV.

The presumably complex photopeak at 320 keV was found to be coincident with gamma rays at 92, 120, and 280 keV as shown in Fig. 6. The 120-keV radiation was evidently of too low an intensity to make an appreciable contribution to the curve of single counts shown in Fig. 5. To obtain the data of Fig. 6, the channel of one analyzer was fixed at 320 keV while that of the other was moved through the region of lower energies. Similarly, the 92-keV gamma ray was found to be in coincidence with quanta of energies of 120, 280, 320, 440, and 600 keV as shown in Fig. 7. The 120-keV and hardest coincident quantum were not of sufficient intensity to appear in the curve of single counts shown in Fig. 5. Not described in any figure is the additional fact that coincidences between the 280- and 410-keV gammas were also noted.

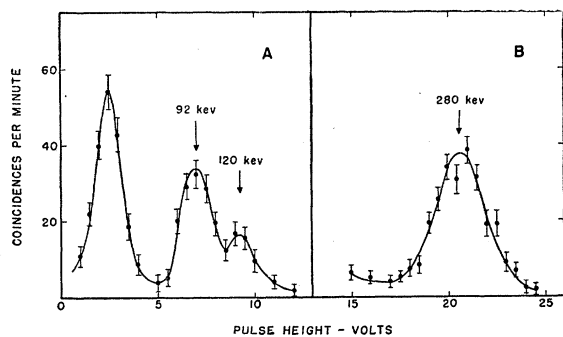


Fig. 6. Gamma rays coincident with 320-keV radiation of  $\text{Nd}^{147}$

The disintegration scheme of  $\text{Nd}^{147}$  is shown in Fig. 8 where the coincidence studies of Figs. 6 and 7 have been combined with the information contained in the spectrum of single counts given in Fig. 5. The relative intensities of the 92-keV and 120-keV peaks of Fig. 6A show that in the triple cascade, the order of emission of the gamma rays is 120 keV  $\rightarrow$  320 keV  $\rightarrow$  92 keV. In

reaching this conclusion, it has been assumed that the conversion coefficient of the 120-keV radiation is not greater than that of the 92-keV line. Further confirmation of the suggested order of emission lies in the fact that the quantum energy of the 410-keV gamma ray is such as to indicate that it might be emitted in the cross-over transition of the 320 keV–92 keV cascade. Assuming little conversion of the 440-keV radiation, it can likewise be argued that because of its small intensity, it precedes the 92-keV radiation in the decay scheme. From energy considerations, it has been assumed that the 530-keV gamma ray is emitted in the cross-over transition of the 440 keV–92 keV cascade. The gamma ray of energy 690 keV has been taken as the cross-over quantum of the 280 keV–320 keV–92 keV cascade.

To obtain the *K*-shell conversion coefficient of the 92-keV gamma ray, the areas under the x-ray peak and

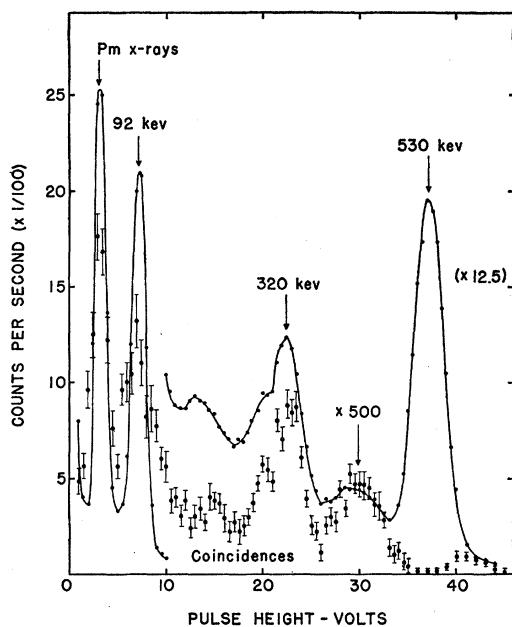


FIG. 7. Gamma rays coincident with 92-keV radiation of Nd<sup>147</sup>.

under the full-energy peak of the 92-keV gamma ray were carefully determined in a crystal arrangement presenting practically no preabsorption to quanta of either energy. The counts in both peaks were properly corrected for escape from the crystal of the *K*-line of iodine. The *K*-shell conversion coefficient thus obtained was found to be  $1.6 \pm 0.2$ . In arriving at this result, it has been assumed that contributions to the x-ray peak arising from conversion of any other gamma rays are small. This assumption can be justified from a consideration of the data of Figs. 5 and 7.

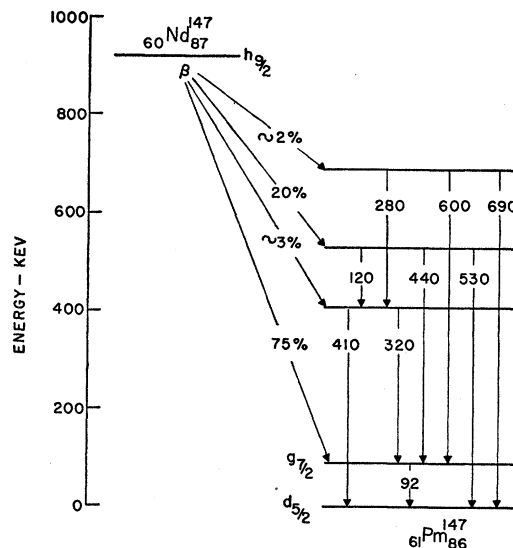


FIG. 8. Nuclear energy levels of Pm<sup>147</sup>.

The measured<sup>11</sup> ground state spin of Sm<sup>147</sup> is 7/2 in agreement with the orbital value of  $f_{7/2}$  predicted by the nuclear shell model. The transition Pm<sup>147</sup> →  $\beta$  → Sm<sup>147</sup> has been classified<sup>12</sup> as first forbidden,  $d_{5/2} \rightarrow f_{7/2}$ . All measured *K/L* ratios and *K*-shell conversion coefficients<sup>7,8,13,14</sup> favor identification of the 92-keV gamma transition as *M1* so that the orbital of the first excited state of Pm<sup>147</sup> may be reasonably taken as  $g_{7/2}$ . This latter assignment is furthermore consistent with an assignment of  $h_{9/2}$  for the ground state of Nd<sup>147</sup> and with the fact that the bulk of the beta rays are contained in a spectrum which terminates at the 92-keV level rather than at the ground state of orbital  $d_{5/2}$ . The values of  $\log ft$  calculated for the various beta spectra shown in Fig. 8 are in the order of increasing end point energy 7.05, 7.00, 8.22, and 7.44. Thus, the *ft* value of the hardest spectrum is consistent with the orbital assignments. It seems inadvisable to attempt interpretation beyond this point, because gamma rays have been reported<sup>7</sup> which cannot be resolved by the methods employed in these measurements.

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<sup>11</sup> C. S. Bogle and H. E. D. Scovil, Proc. Roy. Soc. (London) **A65**, 360 (1952); Kiyoshi Murakawa, Phys. Rev. **93**, 1232 (1954).

<sup>12</sup> Mayer, Moszkowski, and Nordheim, Revs. Modern Phys. **23**, 315 (1951).

<sup>13</sup> A. B. Smith and A. C. G. Mitchell, Phys. Rev. **87**, 1128 (1952).

<sup>14</sup> R. L. Graham and R. E. Bell, Can. J. Phys. **31**, 377 (1953).