Decay of Pm^{143} and Pm^{144}†

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Radioactive Pm^{143} and Pm^{144} were produced by the bombardment of Pr^{141} with α particles. Gamma rays at 475 kev, 610 kev, and 695 kev are found to be associated with the decay of Pm^{144} by K-capture to Nd^{144}. A 740-kev gamma transition is associated with the decay of Pm^{143} by K-capture to Nd^{143}. Levels of Nd^{144} are placed at 695 kev (2+), 1305 kev (4+), and 1780 kev (6+) above the ground level. Assignments of spins of levels and multipolarities of gamma-ray transitions are based on the results of measurements of conversion coefficients and gamma-gamma angular correlations. A value of $(13\pm4)\times10^{-11}$ sec was found for the mean life of the 1305-kev level of Nd^{144}. An upper limit of 3×10^{-11} sec was found for the mean life of the 695-kev level of Nd^{144}.

INTRODUCTION

ALL the published works, concerning the decay schemes of Pm^{144} and Pm^{143}, deal mainly with the identification of the isotopes and the determination of their lifetimes.[1,2] In these investigations it was found that Pm^{144} and Pm^{143} decay by K-capture to Nd^{144} and Nd^{143}, respectively, and that the half-lives of both are about 300 days. These experiments were carried out before the scintillation counter techniques were developed and they did not yield any information concerning the properties of the excited levels of Nd^{143} and Nd^{144} and of the emitted γ-rays.

In the present work the spectra of γ-rays accompanying the decays of Pm^{143} and Pm^{144} were measured. The spectra of coincidences with the various radiations were also measured. In addition, the conversion coefficients of the γ-rays and the angular correlations of 3 γ-ray cascades were determined. On the basis of the results of these measurements decay schemes were suggested and assignments for the spins of excited levels and multipolarities of the γ-radiations were made. Measurement of the lifetimes of two excited states of Nd^{144} were also carried out.

The radioactive sources were produced by the bombardment of Pr_2O_3 with α particles in the 60-in. cyclotron of Brookhaven National Laboratory. The α particles were degraded to suitable energies before striking the target. Alpha-ray energies of 17 Mev, 22 Mev, and 27 Mev were used. The possible reactions leading to the formation of radioactive isotopes in these bombardments are $Pr^{141}(\alpha,n)Pm^{144}$, $Pr^{141}(\alpha,2n)Pm^{143}$, and $Pr^{141}(\alpha,3n)Pm^{142}$. The half-life of Pm^{142} is about 30 sec.[3] As the measurements were carried out several weeks after the bombardment, the radioactivity of the source could come only from the decay of Pm^{143} and Pm^{144}. It was not found necessary to do any chemical separations in the source after the bombardment. By using the tables of semiempirical masses of atoms[4] it is found that the threshold of the reaction $Pr^{141}(\alpha,n)Pm^{144}$ is about 10 Mev and the threshold of the reaction $Pr^{141}(\alpha,2n)Pm^{143}$ is about 16 Mev. A comparison of the γ-ray spectrum obtained from a source which was bombarded by 17-Mev α particles with the spectrum obtained from a source which was bombarded by 27-Mev α particles showed clearly which γ-rays accompanied the decay of Pm^{144} and which accompanied the decay of Pm^{143}. Only one γ-ray (with an energy of 740 kev) accompanies the decay of Pm^{143}. All other γ-rays found in the spectrum accompany the decay of Pm^{144}.

EXPERIMENTAL PROCEDURE

1. Spectrum of Gamma Rays

The spectrum of γ-rays was examined with a 3 in. $\times$3 in. NaI(Tl) scintillation counter in combination with a hundred-channel analyzer. In curve A of Fig. 1 the shape of spectrum of γ-rays emitted from a source which was bombarded with 17-Mev α particles is shown. The spectrum obtained from a source bombarded with 27-Mev α particles is shown in curve B of Fig. 1. The clear conclusion is that the decay of Pm^{143} is accompanied by a 740-kev γ-ray and the decay of Pm^{144} is accompanied by 475-kev, 610-kev, and 695-kev γ-rays. An analysis of the spectra shows that no other γ-rays with intensities above 3×10^{-3} per disintegration are emitted.

2. Coincidence Studies

Gamma-gamma coincidence measurements were carried out with an apparatus employing two 3 in.$\times$3 in. NaI(Tl) crystals. Pulse-amplitude discrimination was made in the "gate"channel by a single-channel pulse-height analyzer and the coincidence spectrum was displayed on a 100-channel pulse-height analyzer. The coincidence circuit was operated at a resolving time (2τ) of 2×10^{-7} sec.

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* On leave from the Hebrew University, Jerusalem, Israel.
[1] V. K. Fisher, Phys. Rev. **87**, 859 (1952).
[2] G. Wilkinson and H. G. Hicks, University of California Radiation Laboratory Report UCRL-751, June, 1950 (unpublished).
[3] T. V. Marshal and J. O. Rasmussen (unpublished, 1958).
[4] A. G. W. Cameron, Chalk River Project Report CRP-690, 1957 (unpublished).

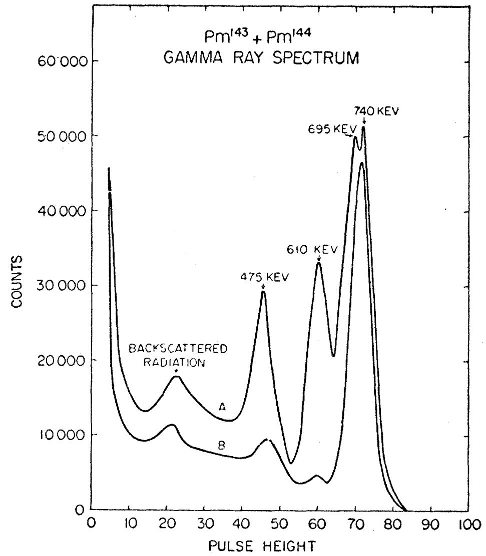

FIG. 1. Spectrum of $Pm^{143}+Pm^{144}$ gamma rays. (A) Source prepared by bombardment with 17-Mev α rays; (B) Source prepared by bombardment with 27-Mev α rays.

An analysis of the spectrum of coincidences with the 40-kev x-rays shows that its shape is identical with the $Pm^{143}+Pm^{144}$ singles spectrum. This proves that both isotopes decay by K-capture and that the lifetimes of the excited states are shorter than 10^{-7} sec. An analysis of the spectra of singles and coincidences obtained with the source prepared by bombardment with 27-Mev α-rays shows that $(45\pm5)\%$ of all the decays of Pm^{143} go to the 740-kev excited level of Nd^{143}. This value for the branching ratio was obtained from the comparison of the number of x-rays in the gating channel with the number of coincident pulses in the 740-kev peak. The same value was also obtained from the analysis of the "singles" spectrum. The decay scheme of Pm^{143} is given in Fig. 2.

The spectra of coincidences with the 475 kev, 610 kev, and 695 kev γ-rays were measured using a source

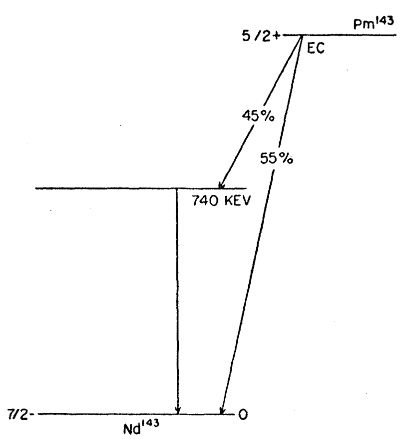

FIG. 2. Decay scheme of Pm^{143}.

prepared by bombardment with 17-Mev α particles, as shown in Fig. 3. The following conclusions have been drawn from the analysis of "singles" and coincidence spectra: The 610-kev and 695-kev transitions have equal intensities. The intensity of the 475-kev transitions is 45% of the intensities of the 610-kev and 695-kev transitions. The 610-kev and 695-kev transitions are in cascade. All the 475-kev γ-rays are coincident

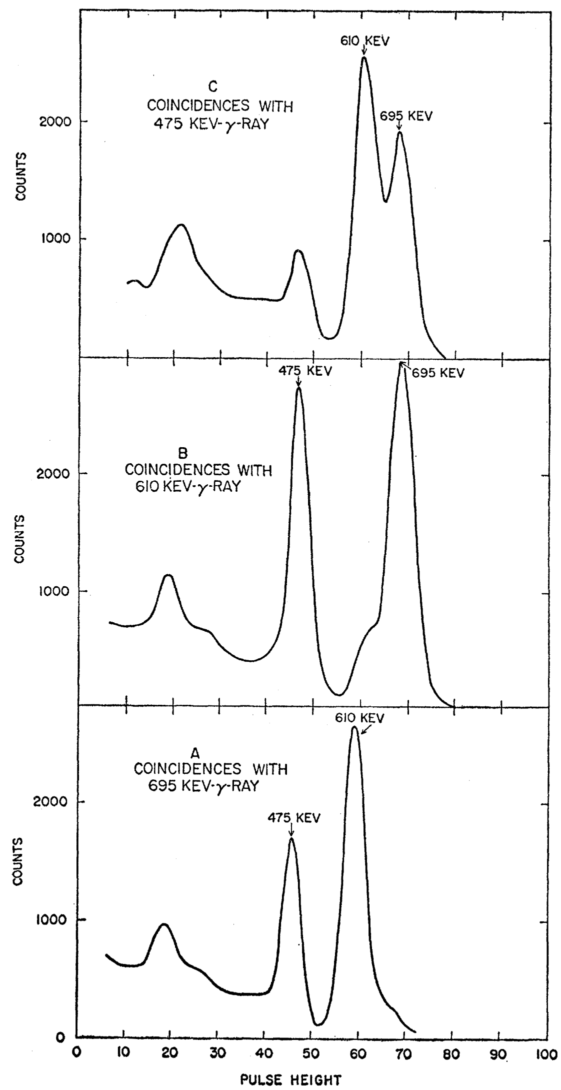

FIG. 3. Spectra of coincidences with 475 kev, 610 kev, and 695 kev γ rays of Pm^{144}.

with the 610-kev and 695-kev γ-rays. 45% of the 610-kev and 695-kev γ-rays are coincident with the 475-kev γ-ray. The 695-kev transition can be identified with the 695-kev transition following the β-decay of Pr^{144} to Nd^{144}.[5] A decay scheme of Pm^{144} consistent with the experimental results is given in Fig. 4.

[5] Hickok, McKinley, and Fultz, Phys. Rev. **109**, 113 (1958).

No 511 kev–511 kev coincidences could be found and the analysis of the measurements showed that for both Pm^{143} and Pm^{144} the ratio of β^+ decay to K-capture is smaller than 1/500.

3. Angular Correlations of Gamma-Ray Cascades

The angular correlations of the gamma-ray cascades following the decay of Pm^{144} were measured by using a standard fast-slow coincidence circuit and adjusting the channels of the pulse-height analyzers to select pulses belonging to the full peaks of the two γ-rays. The resolving time of the fast coincidence circuit was $(2\tau)=6\times10^{-8}$ sec. The radiations were detected by two 3 in.×3 in. NaI(Tl) scintillation counters. The distance between the source and the crystals was 14 cm. The angle between the counters was changed automatically every 5 minutes by 30° in the region between 90° and 270°. The number of single counts and coincidences were recorded at each angle on registers. The angular correlation function obtained for the 695 kev–610 kev cascade, after correcting for the finite solid angle of the detectors, was

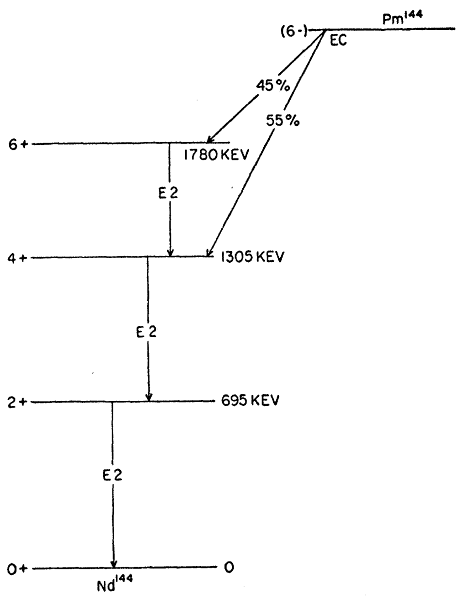

FIG. 4. Decay scheme of Pm^{144}.

$$P(\theta)=A[1+(0.103\pm0.010)P_2(\cos\theta)+(0.01\pm0.01)P_4(\cos\theta)]. \quad (1)$$

The angular correlation function obtained for the 475 kev–610 kev cascade was

$$P(\theta)=A[1+(0.11\pm0.01)P_2(\cos\theta)+(0.01\pm0.01)P_4(\cos\theta)]. \quad (2)$$

The function obtained for the 475 kev–695 kev cascade was

$$P(\theta)=A[1+(0.093\pm0.010)P_2(\cos\theta)+(0.02\pm0.01)P_4(\cos\theta)]. \quad (3)$$

4. Internal Conversion Coefficients

The source used for conversion electron studies was prepared by bombarding a Pr_2O_3 powder in the cylotron with 17-Mev α-particles for about 30 hours. The intensity of the beam was about 30 μa. The oxide was dissolved in hydrochloric acid. After evaporating the acid, the $PmCl_3$ salt was dissolved in water and the solution was evaporated onto a 1-mil thick Al foil over an area about 1 cm in diameter. The thickness of the source was about 1 mg/cm².

The measurements were performed with an iron-free intermediate-image spectrometer designed by Alburger.[6] The resolution setting of the spectrometer was 2.4% and its transmission about 4.5%. Four conversion lines with energies of 438 kev, 573 kev, and 703 kev were found. The lines could be identified as the K-conversion lines of the 475-kev, 610-kev, 695-kev, and 740-kev γ-transitions. The K-conversion coefficients were determined by comparing the relative intensities of the K-conversion lines with the relative intensities of the γ-rays. If one assigns a 2^+ character to the 695-kev level in Nd^{144} on the basis of its being the first excited state of an even-even nucleus and hence the multipole order $E2$ to the 695-kev γ-transition, the theoretical value of its conversion coefficient is 4.3×10^{-3}. Relying on this value and on the relative intensities of the γ transitions and their conversion lines, the K-conversion coefficients given in Table I were deduced.

5. Measurement of Lifetimes of the 695-kev and 1305-kev Levels of Nd^{144}

The lifetimes of the 695-kev and the 1305-kev levels of Nd^{144} were measured by using the electronic apparatus for lifetime measurements designed by Sunyar and described elsewhere.[7,8] Two plastic scintillators with thicknesses of about 2 cm were used for the

TABLE I. K-conversion coefficients of Pm^{143} and Pm^{144} γ transitions.

Energy of γ transition	Relative intensity of γ-line	Relative intensity of K-conversion line	Conversion coefficient
475 kev	45	90	$(1\pm0.2)\times10^{-2}$
610 kev	100	120	$(5.7\pm0.5)\times10^{-3}$
695 kev	100	100	4.3×10^{-3}
745 kev	150	195	$(6.5\pm1)\times10^{-3}$

[6] D. E. Alburger, Rev. Sci. Instr. **27**, 991 (1956).
[7] A. W. Sunyar, Bull. Am. Phys. Soc. Ser. II, **2**, 37 (1957).
[8] A. W. Sunyar, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958).

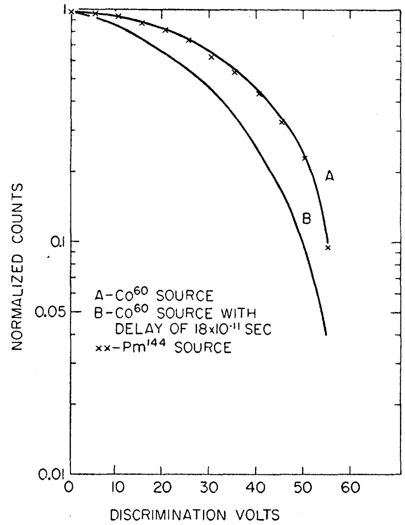

FIG. 5. Normalized integral spectra of coincidence pulse heights in the measurement of the 695-kev level lifetime.

measurement of the lifetime of the 695-kev level. The channel of one pulse-height analyzer was adjusted to select pulses corresponding to 695-kev γ rays detected in one plastic sctintillator. The channel of the second pulse-height analyzer was adjusted to select pulses corresponding to 610-kev γ rays (and higher energy γ rays but not 475-kev γ rays). The output pulses from the fast coincidence circuit were analyzed by a 100-channel pulse analyzer gated by the output of the slow triple-coincidence circuit. The points marked on curve *A* of Fig. 5 give the number of coincidences obtained with heights higher than the corresponding values given on the abscissa. The numbers were normalized so that the total number of coincidences was taken as 1. The spectrum of coincidences obtained was compared to the spectrum obtained with a Co^{60} source using the same channels as used with the Pm^{144} source. Curve *A* in Fig. 5 shows the integral spectrum obtained with the Co^{60} source. In order to measure the sensitivity of the shape of the coincidence spectrum to the lifetime of the intermediate state, an 18×10^{-11} sec delay cable was introduced between the 695-kev radiation detector and the fast-coincidence circuit, and the spectrum of the coincidences was measured using a Co^{60} source. In Fig. 5B the integral spectrum obtained with the 18×10^{-11} sec delay is shown. An analysis of the curves in Fig. 5 shows that the mean life of the 695-kev level is shorter than 3×10^{-11} sec.

The lifetimes of the 1305-kev level could not be measured by simply channeling the pulse-height analyzer on the 475-kev and the 610-kev lines respectively because in doing so the analyzers would also select pulses belonging to γ radiations of higher energy and not all the coincidences measured would belong to the 475 kev–610 kev or the 475 kev–695 kev cascades. The measurement of the lifetime of the 1305-kev level was carried out by using 2 plastic scintillators facing each other with the source between them and a 3 in. $\times$3 in. NaI scintillator perpendicular to the two plastic scintillators and as close to them as possible. The multichannel pulse analyzer was gated by the output of a fast-slow triple-coincidence circuit. The resolving time of this fast triple-coincidence circuit was about 10^{-7} sec. The outputs of the three scintillators were fed to the fast-slow triple-coincidence circuit. The analyzer of the pulses from the NaI scintillator was channeled on a region including the 610-kev and 695-kev photopeaks. The analyzer of the pulses of one of the plastic scintillator was channeled on a region including pulses corresponding to γ-rays with energies higher than 475 kev (610-kev and 695-kev γ-rays) and the third pulse-height analyzer was channeled on a region containing pulses corresponding to 475-kev γ rays (and higher energy γ rays) detected by the second plastic scintillator. The pulses from the plastic scintillators pulses were also fed to Sunyar's fast-coincidence circuit ($2\tau=3\times10^{-9}$ sec) and the coincidence spectrum was analyzed by the gated multichannel analyzer. In this way it was guaranteed that only coincidence pulses corresponding to cascades whose intermediate level is the 1305 kev would be analyzed. Curve *B* of Fig. 6 shows the integral spectrum of the coincidence pulses obtained with a Pm^{144} source. In curve *A* of Fig. 6 the integral curve obtained with a Co^{60} source (without requiring coincidences with the NaI detector) is shown. In curve *C* of Fig. 6 the integral curve obtained with a Co^{60} source after introducing a 20×10^{-11}-sec delay cable between the plastic scintillator channeled on the region above 475 kev and the fast-coincidence circuit is shown. An analysis of the curves of Fig. 6 shows that the mean life of the 1305-kev level of Nd^{144} is $(13\pm4)\times10^{-11}$ sec.

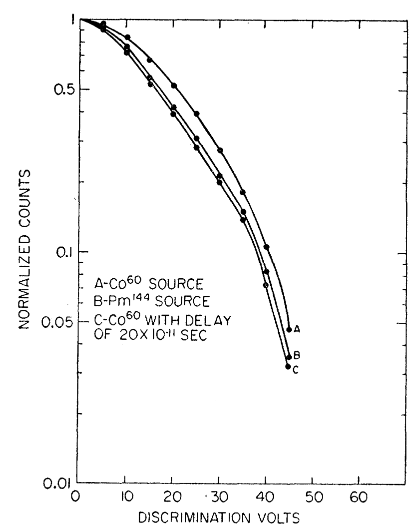

FIG. 6. Normalized integral spectra of coincidence pulse heights in the measurement of the 1305-kev level lifetime.

ANALYSIS OF EXPERIMENTAL RESULTS AND DISCUSSION

A 2+ classification is to be assigned to the 695-kev level of Nd^{144}, on the basis of being the first excited state of an even-even nucleus. The experimental K-conversion coefficients of the 610-kev and 475-kev transitions are in agreement with the theoretical conversion coefficients assuming that the transitions are of the $E2$ type. The nonexistence of a crossover transition from the 1305-kev level to the ground state of Nd^{144}, the value of the K-conversion coefficient of the 610-kev γ transition, and the angular correlation function obtained for the 610 kev–695 kev cascade are all in agreement with the assignment of spin and parity 4+ to the 1305-kev level. No other assignment can be reconciled with both the value of the K-conversion coefficient of the 610-kev level and the angular correlation function of the 695 kev–610 kev cascade. The most reasonable spin and parity assignment for the 1780-kev level of Nd^{144} is 6+. This assignment is based on the angular correlation function obtained for the 475 kev–610 kev cascade, on the value found for the K-conversion coefficient of the 475-kev transition and on the nonexistence of γ transition from the 1780-kev level to the ground state and first excited level of Nd^{144}. The angular correlation function obtained for the one-three gamma-gamma angular correlation of the 475-kev and the 695-kev γ rays is also in agreement with the theoretical function for a 6(2)4(2)2(2)0 cascade.[9]

The ground state characteristics of the odd-odd nucleus of Pm^{144} are not uniquely determined by the shell model,[10] but the predictions call for odd parity and a high spin. The odd neutron and odd proton most likely occupy $f_{7/2}$ and $d_{5/2}$ levels, respectively. According to Nordheim's "weak" rule, the angular momenta of the proton and the neutron tend to add, although not necessarily to the highest possible value. A high spin assignment for the ground state of Pm^{144} is also in agreement with the fact that Pm^{144} does not decay to the ground state or to the first excited state of Nd^{144} whose spins are 0 and 2, respectively. The transition energy from Pm^{144} to the ground level of Nd^{144}, as calculated from a semiempirical mass formula,[4] is about 1.9 Mev. The transition energy to the 1780-kev level is, therefore, about 120 kev and to the 1305-kev level about 600 kev. These calculated energies, are in agreement with the fact that within the accuracy of the measurements, no β^+ emission was found in the decay of Pm^{144}. Relying on these calculated transition energies, a value of 8.0 is found for the $\log ft$ of the decay to the 1305-kev level and a value of 6.8 is found for the $\log ft$ of the decay to the 1780-kev level. These $\log ft$ values lead to the conclusion that the most reasonable spin and parity assignment for the ground level of Pm^{144} is 6−, although an assignment 5− cannot be ruled out. In Fig. 4 the assignments of spins and multipolarities in the decay of Pm^{144} are given.

The measured spin of Nd^{143} is $\frac{7}{2}$,[11] and according to the shell model[12] it is a $f_{7/2}$ state with odd parity. The most reasonable spin and parity assignment, based on the shell model, for Pm^{143} is $\frac{5}{2}+$ (a $d_{5/2}$ state).[12] The decay of Pm^{143} to the ground level of Nd^{143} is therefore first forbidden. The calculated energy available for this transition is about 1.9 Mev[4] and its $\log ft$ value 8.8. This $\log ft$ value is not far from the average value for a first forbidden transition. The transition energy of the K-capture decay to the 740-kev level of Nd^{143} is 1.150 Mev and its calculated $\log ft$ value is 8.5. The experimental value of the conversion coefficient of the 740-kev γ transition is in agreement with the theoretical value for an $M1$ transition. The most likely spin and parity assignments for the 740-kev level are $\frac{9}{2}-$, $\frac{7}{2}-$ or $\frac{5}{2}-$.

Lifetime calculations, based on single proton transition probabilities,[13] yield a value of 10^{-10} sec for the mean life of the 1305-kev level and a value of 5×10^{-11} sec for the mean life of the 695-kev level. The experimental value obtained for the mean life of the 1305-kev level is in agreement with the calculated one whereas the lifetime of the 695-kev level is shorter than the calculated lifetime.‡

ACKNOWLEDGMENTS

I would like to thank Dr. D. E. Alburger for the use of his spectrometer, Dr. A. W. Sunyar for the use of his apparatus for short-lifetime measurements, and Dr. C. P. Baker for arranging the cyclotron bombardments.

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[12] M. Goeppert-Mayer, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), p. 433.

[13] S. A. Moszkowski, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), p. 391.

‡ *Note added in proof.*—Beta decay systematics indicate that the transition energy of the decay of Pm^{143} to the ground level of Nd^{143} is about 1.5 Mev and the $\log ft$ values of this decay and of the decay to the 740-kev level are about 8.5 and 8, respectively.