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THE DECAY OF PM^{150}

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Berkeley, California

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Radiation Laboratory, Department of Physics,
University of California, Berkeley, California,
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U. S. Naval Radiological Defense Laboratory,
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ABSTRACT

A grey wedge pulse-height analyzer was used to study the decay of Pm^{150} with and without coincidence arrangements. Gamma rays 3.0, 2.6, 2.0, 1.67, 1.32, 1.17, 0.82, 0.43 and 0.34 Mev in energy were observed, some in coincidence with 2.01- and 3.00-Mev negatrons which had previously been shown to be emitted. A decay scheme is suggested and some remarks on the beta stability of Nd^{150} are made.

* This work was sponsored in part by the U. S. Atomic Energy Commission.

† The completion of this study was made possible by the Sarah Berliner fellowship (1953-54) of the American Association of University Women.

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INTRODUCTION

Pm^{150} has been shown by Long and Pool¹ and this author² to decay to Sm^{150} by negatron emission with a 161-minute half life. Hibdon and Muehlhause³ have studied the conversion electrons of gamma rays from excited states of Sm^{150} produced by neutron capture by Sm^{149} . They found that 336.7- and 440.2-keV gamma rays were emitted, and proposed the level assignments shown in Fig. 1.

Previous studies with a double-focusing beta-ray spectrometer indicated that Pm^{150} decays by emitting 2.01- and 3.00-MeV negatrons.² Lead absorption studies showed at least two gamma rays ~ 1.4 and ~ 0.3 MeV in energy to be present. In the course of the present work Dr. T. Passell of this laboratory (University of California) examined a sample with the same instrument for conversion electrons. Peaks due to a 336-keV gamma ray were seen, in agreement with Hibdon and Muehlhause, but none from a 440-keV gamma ray. However, the sample was so weak that peaks less than one third as abundant as those observed would not have been detected.

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+ The completion of this study was made possible by the Sarah Berliner fellowship (1953-54) of the American Association of University Women.

1. J. K. Long and M. L. Pool, Phys. Rev. 85, 137 (1952).
2. Vera Kistiakowsky, Phys. Rev. 87, 859 (1952).
3. C. T. Hibdon and C. O. Muehlhause, Phys. Rev. 88, 943 (1952).

Two considerations prompted a further study of the decay of Pm^{150} . First, the Bohr-Mottelson⁴ collective model of the nucleus which successfully treats excited states in even-even nuclei as rotational states, uses Sm^{150} as one example. This makes further knowledge of its levels seem desirable. Second, a knowledge of the Pm^{150} - Sm^{150} ground-state energy difference, in conjunction with the known Nd^{150} - Sm^{150} mass difference, might permit a verification of the suggestion by Kohman⁵ that Nd^{150} , which occurs in nature, is probably beta unstable.

Sample Preparation

The samples studied were prepared by bombarding Nd_2O_3 enriched with Nd^{150} with ~ 9 -Mev protons from the 60-inch cyclotron at Crocker Radiation Laboratory for one hour at an average external beam current of one micro-ampere. The (p, n) and (p, 2n) reactions occur with comparable cross sections (~ 2 millibarn), and at the end of bombardment ~ 5 percent of the disintegrations are those of Pm^{149} , a negatron emitter with a 54-hour half life.

The samples were purified from non-rare-earth activities by dissolving in dilute nitric acid from which the rare earth fluoride was precipitated. This was dissolved in concentrated boric and nitric acids and the hydroxide was precipitated. The sample was then mounted on a platinum disk for study.

4. A. Bohr and B. R. Mottelson, Dan. Mat. Fys. Medd. 27, No. 16 (1953).

5. T. P. Kohman, Phys. Rev. 73, 16 (1948); private communication (1954).

Apparatus

Grey Wedge Analyzer

The reader is referred to Bernstein, Chase, and Schardt⁶ for a discussion of the principles and problems of grey wedge pulse-height analysis. However, since the arrangement used was developed at this laboratory (University of California) and differs from that published in the reference given above, it is discussed briefly here.

A block diagram is given in Fig. 2. The first unit is a conventional NaI(Tl) - DuMont 6292 phototube package incorporating 1 μ sec delay-line clipping necessary for the coincidence work and a cathode follower output. The pulses are amplified by a standard nonoverloading UCRL linear amplifier. They then trigger the sweep of a Tektronics 512AD oscilloscope, the output 16- μ sec gate pulse of which is sent to the pulse-stretcher unit. This consists basically of a normally conducting diode which is clamped for the duration of the gate pulse. The linear amplifier pulses going into this unit are delayed 1.25 μ sec and then charge a condenser which discharges only when the diode again becomes conducting. The stretched pulse which goes to the oscilloscope signal input is constant in amplitude to ~ 2 percent and the device is linear to ~ 2 percent in the operating range of 5-95 volts.

A third output of the linear amplifier is used to trigger a standard UCRL variable delay and gate unit. A suitably delayed 12- μ sec positive gate pulse from this unit is amplified, inverted, and clipped to a constant amplitude of -40 volts by the intensifier pulse shaper, and is then used to intensify the oscilloscope trace.

The 512AD Tektronics oscilloscope is equipped with a 5XP11-M tube and modified to be used with an external high-voltage supply. The traces were photographed by a 4-by-5-inch view camera with a 127-mm f 4.5 lens.

6. W. Bernstein, R. L. Chase and A. W. Schardt, Rev. Sci. Inst. 24, 437 (1953).

A 4-by-5 inch grey wedge was mounted in the back of the camera directly in front of the film. After experiments with several types of film, Kodak Super Ortho Press was settled upon, because it combines workable film-speed and contrast qualities. The latter were emphasized by overdeveloping in Kodak D-19.

The NaI (Tl) crystals ⁷ used were packaged with a MgO diffuse reflector. A crystal 1.5 inches in diameter by 2 inches long was used for high-energy gamma-ray studies, while another, 1.5 inches in diameter by 1/2 inch long, was used for lower-energy portions of the spectrum. Beryllium absorbers of 1500 mg/cm² were used to remove the negatron spectra.

Pictures were enlarged, and corrected for a slight barrel distortion of the pulses on the oscilloscope face by reading them from a grid. A relative calibration of exposure amplitude versus counting rate was made and checked roughly before each series of pictures for the given experimental conditions. Energy calibration was made before each experiment using samples of Co⁵⁶, Co⁶⁰, Na²², Cs¹³⁷, Cd¹⁰⁹, and Am²⁴¹, and some points were retaken at intervals to check for drifts.

Figure 3 shows a typical grey wedge picture taken for energy-calibration purposes. The distribution of the peaks in such pictures was also used in interpreting the unknown spectra.

The limits of error quoted on the gamma-ray energies include small uncertainties in the energy calibration as well as the uncertainties of reading the pictures.

Coincidence Experiments

Figure 4 is a block diagram of the coincidence experiment electronics. Pulses from the scintillation counter are coincidized either with those from a proportional counter or with those from a single-channel pulse-

7. Obtained from The Harshaw Chemical Co., Cleveland 6, Ohio

height analyzer which examines the pulse distribution from a second scintillation counter. The resolving time of the arrangement is $\sim 3 \mu\text{sec}$, placing a severe limitation on the count rate allowable. In all cases experiments were performed with maximum possible geometry: 16 percent for each of the scintillation counters and 25 percent for the proportional counter. Samples of various strengths were used to adjust the count rate to an optimum value with respect to both chance coincidences and statistics.

The coincidence signal is fed into a variable delay and gate unit. An undelayed pulse from the discriminator in the gate input is used to trigger the grey wedge analyzer oscilloscope, while the delayed gate pulse is used for intensification as before.

No attempt is made to achieve actual pulse height-to-energy correspondence for beta particles in the gas counter. It is operated in the proportional region rather than the Geiger region for the sake of resolution time, and its pulses are RC clipped to $1 \mu\text{sec}$ (decay from 90 percent to 10 percent maximum). Decisions as to beta-gamma coincidences were made by quantitatively comparing spectra in coincidence with the proportional counter with various thicknesses of absorber in front of it.

RESULTS

The first noncoincidence experiments and all the coincidence experiments indicate the presence of gamma rays up to $\sim 2.5 \text{ Mev}$ in the decay of Pm^{150} . Later experiments, in which several long exposures of the high-energy spectrum were taken, showed another peak at 3.0 Mev and were better in high-energy calibration. Figure 5 shows one of these pictures, and Fig. 6 contains the spectrum obtained with twice the linear amplifier gain of Fig. 5.

The results are summarized in Table I. The blank spaces represent those points on which no conclusions could be drawn, owing to insufficient data. "O" stands for observed, "N" stands for observed to be absent, and "?", for uncertainty. The approximate relative abundances were calculated by comparison with the spectra obtained for samples having known relative abundances. Owing to uncertainties involved in this method the numbers given are good only to an order of magnitude.

Table I - Energies of Observed Gamma Rays

Energy (Mev)	Approximate Relative Abundance	Occurrence in Decay Scheme	Observations			
			No Coincidence	Coincidence 2-Mev β^-	Coincidence 3-Mev β^-	Coincidence 0.34-Mev γ
3.0 \pm 0.1	0.004	γ_1	0	Distribution of pulses up to \sim 2.5 Mev		Distribution of pulses up to \sim 2.0 Mev
2.6 \pm 0.1	0.008	γ_2	0			
2.0 \pm 0.1	0.004	$\gamma_3, \gamma_6, \gamma_{10}$	0			
1.67 \pm 0.05	0.008	γ_7, γ_{11}	0	0		0
1.32 \pm 0.05	0.04	γ_4	0	0	0	0
? 1.24		γ_{12}			0 ?	
1.17 \pm 0.05	0.04	$\gamma_8, \gamma_{12}, \gamma_{14}$	0	0		N
? 0.96		γ_5	0 ?			
0.82 \pm 0.02	0.4	γ_{13}, γ_{15}	0	0	0	0
0.43 \pm 0.02	0.2	γ_{17}	0	0		0
? 0.39		γ_9, γ_{16}			0 ?	0 ?
0.34 \pm 0.01	1.0	γ_9, γ_{18}	0	0		N

All the gamma-ray peaks observed in the noncoincidence experiments stay in the same ratios to one another during the first nine hours of decay, and during this period the gross decay has a half life of 161 minutes. After that the decay rate decreases and peaks belonging to the 285-kev and 1-Mev gamma rays of Pm^{149} change the spectrum. It was found that the number of gamma rays in the energy interval 3.0 ± 0.25 Mev decreases with a half life of 160^{+160}_{-60} minutes. Unfortunately it was impossible to make a sufficiently active sample to permit a more accurate determination.

Before the 3.0-Mev gamma ray had been discovered, no decay scheme agreeing with all the experimental results could be formulated. Therefore the old beta-ray spectroscopy results were reexamined to determine if they could be in error. Four independent sets of data give Fermi-Kurie plots which resolve to give 2.01- and 3.00-Mev components. One, shown in Fig. 7, also exhibits a 0.97-Mev negatron due to Pm^{149} . However, the author would like to revise her earlier calculation of the relative intensities to $(20 \pm 10 \text{ percent})$ and $(80 \pm 10 \text{ percent})$ respectively for the 3.00- and 2.01-Mev components.

CONCLUSIONS

Figure 8 shows the simplest decay scheme in agreement with all the results of these and previous experiments. It is not thought that all the transitions indicated occur. They are just listed to show how they could be explained by the observed energy spectrum.

At first it was postulated that the 3.0-Mev gamma ray arises from a transition to ground state. There are three objections to this. First, although the energies of the highest-energy gamma rays are uncertain by 0.1 Mev, their differences are certain to the accuracy of reading the pictures. Thus

$$E_{(3.0 \text{ Mev})} - E_{(2.6 \text{ Mev})} = 0.40 \pm 0.05$$

$$E_{(2.6 \text{ Mev})} - E_{(2.0 \text{ Mev})} = 0.57 \pm 0.05$$

If the 3.0-, 2.6-, and 2.0-Mev gamma rays corresponded to transitions to the ground state and first and second excited states, one would expect differences of 0.34 and 0.44 Mev respectively.

Second, no simple decay scheme can be formulated from this postulate that accounts for the observed abundance of the 1.17-Mev gamma ray.

Third, in order that the transition of ground have a sufficiently low multipole order to be probable, it must be assumed that the highest level to which the Pm^{150} negatron decays has a small spin. This seems unlikely in view of the following considerations. From Klinkenberg's⁸ tables the 61st proton of Pm^{150} is assigned to a $d_{5/2}$ state, and the 89th neutron to a $f_{7/2}$ state. If Nordheim's⁹ rules for odd-odd isotopes are used in conjunction with Schwartz's¹⁰ remarks, the ground state of Pm^{150} is expected to have odd parity and spin, J ; $1 < J \leq 6$. The log ft values indicate that the negatron transitions are first-forbidden, and thus the levels in Sm^{150} to which the decay leads should have even parity and spin differing by zero or one unit from that of the ground state of Pm^{150} . Thus if the highest levels in Sm^{150} have spin less than 4 and Pm^{150} has spin less than 5, negatron transitions to intermediate levels of spin of at least 3 would be expected to occur. Q. E. D.

These three arguments are satisfied if it is assumed that the 3.0-Mev gamma ray arises from a transition to the first excited state. The energies of this and the second level are taken from the work previously mentioned. The remaining levels represent the most obvious choices satisfying the experimental results. The three most important justifications should be mentioned, however, although a detailed discussion would take too long. First, if the highest level is at 3.3 ± 0.1 Mev, there must be another, 1 Mev below, to account for the negatron decay. Second, the level at 1.17 ± 0.05 Mev accounts for the 1.17-Mev gamma ray, which is not in coincidence with the 0.34-Mev gamma ray. Third, the level at 2.0 ± 0.1 Mev accounts for the abundance of the 1.67-Mev gamma ray in coincidence with the 2-Mev negatron.

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8. P. F. A. Klinkenberg, Revs. Modern Phys. 24, 63 (1952).
 9. L. Nordheim, Phys. Rev. 78, 294 (1950).
 10. C. Schwartz, Phys. Rev. 94, 95 (1954).

On the basis of the experimental results no spin and parity assignments can be made. However, the following remarks seem pertinent. In several recent compilations^{4, 11, 12} of data on even-even isotopes, the second excited state of Sm^{150} is assigned spin 4 and even parity. This is the best interpretation of the results of Hibdon and Muehlhause, but an assignment of 3+ to this level is not ruled out. A survey of the Table of Isotopes¹³ gave no preference for the assignment of 4+ to the second excited states of even-even nuclei. Of thirteen cases with $Z > 20$ for which spin assignments have been established, one has spin 0; one, spin 1; five, spin 2; three¹⁴, spin 3; and three, spin 4. In the previously mentioned papers^{4, 11, 12} thirteen additional examples where a tentative assignment of spin 4 can be made are given. Sm^{150} is one of these examples and the deviation of the ratio of the energies of the first two excited states from the value predicted by the simple formula for rotational levels¹¹

$$E_I = \frac{\hbar^2}{2 \mathcal{J}} I (I + 1)$$

by an increase in nuclear deformation due to a vibration-rotation type of interaction. The correction term¹¹ is

$$\Delta E_I \approx -2 \left(\frac{1}{\hbar \omega_{\text{vib}}} \right)^2 \left(\frac{\hbar^2}{\mathcal{J}} \right)^3 I^2 (I + 1)^2 .$$

Calculating the parameters on the assumption that $E_2 = 337$ kev and $E_4 = 777$ kev, it is found that ΔE_I is 1.2% and 39% of E_I for E_2 and E_4 respectively. Extrapolating, it would be 82% for E_6 . Thus the criterion for the existence of a rotational spectrum,¹¹ that ΔE_I be small compared to E_I , is not fulfilled for $I \geq 4$. It is not unreasonable, therefore, to suggest that the

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11. "Rotational States of Atomic Nuclei." Aage Bohr. Ejnar Munksgaards Forlag, Copenhagen (1954).
 12. E. L. Church and M. Goldhaber, to be published (1954).
 13. J. M. Hollander, I. Perlman, and G. T. Seaborg, Revs. Modern Phys. 25, 469 (1953).
 14. One of these is Pb^{206} . D. E. Alburger, Washington, D. C., meeting of the American Physical Society (1954).

second excited state of Sm^{150} is a 3+ rather than a 4+ state, an assignment which would agree more satisfactorily with the observed gamma-ray abundances.

There are other indications that Sm^{150} is not a strong-coupling case. The large isotope shifts observed between spectra of isotopes with 82 + 6 neutrons and those with 82 + 8 neutrons (e. g. ${}_{62}\text{Sm}^{150} - {}_{62}\text{Sm}^{152}$ and ${}_{63}\text{Eu}^{151} - {}_{63}\text{Eu}^{153}$ ^(15.), and ${}_{60}\text{Nd}^{148} - {}_{60}\text{Nd}^{150}$ ^(16.)), suggest that some change in nuclear structure takes place between these neutron numbers. Rasmussen ¹⁷ has pointed out that the large quadrupole moment of ${}_{63}\text{Eu}^{153}$ indicating a large spheroidal distortion of the nucleus suggests applicability of the strong-coupling model. It seems reasonable, therefore, to suppose that the change that occurs is from intermediate to strong coupling.

15. P. Brix and H. Kopferman, Phys. Rev. 85, 1050 (1952).

16. P. F. A. Klinkenberg, Physica 11, 327 (1945).

17. J. O. Rasmussen, Jr.; Arriv. For Fysik, Band 7, 185 (1953).

Stability of Nd¹⁵⁰

Kohman⁵ has pointed out that Nd¹⁵⁰ would be expected to be unstable with respect to negatron decay to Pm¹⁵⁰. However, Mulholland and Kohman¹⁸ did not observe any appreciable activity in neodymium, and placed a lower limit of 2×10^{15} years on the possible half life of such decay.

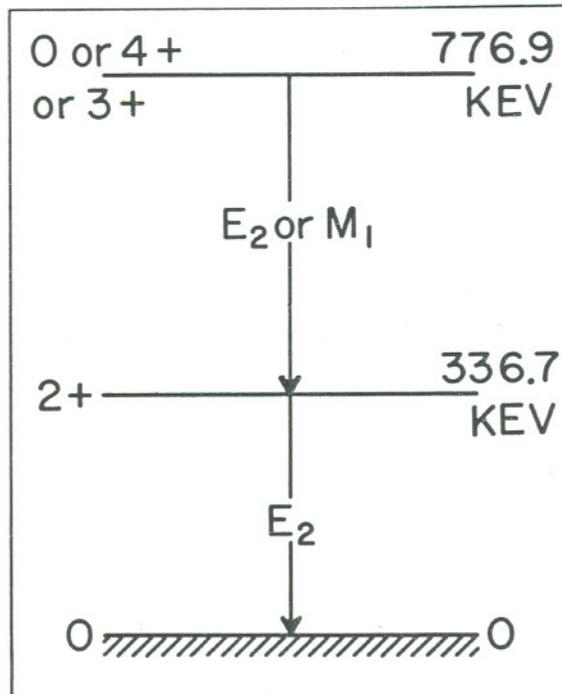
Hoagg and Duckworth¹⁹ have obtained a Nd¹⁵⁰ - Sm¹⁵⁰ mass difference of 4.6 ± 0.8 Mev. Since the proposed Pm¹⁵⁰ - Sm¹⁵⁰ ground-state energy difference is 5.3 ± 0.15 Mev, the Nd¹⁵⁰ - Pm¹⁵⁰ difference is -0.7 ± 1.0 Mev and Nd¹⁵⁰ may or may not be stable. If it is not, then it is unstable by at most 0.3 Mev. Accepting the previous arguments for an assignment of negative parity and spin 5 or 6 to be the ground state of Pm¹⁵⁰, the negatron decay would be at least fifth-forbidden. A reasonable choice of log ft would be 26, and from this a lower limit of 10^{18} years can be set on the half life.

If, on the other hand, Pm¹⁵⁰ is unstable with respect to Nd¹⁵⁰, this mode of decay would not be detected. Assuming the 4+ level of ${}_{60}\text{Nd}^{150}_{90}$ to be analogous to that of ${}_{62}\text{Sm}^{152}_{90}$, it would be at 0.37 Mev¹². Assuming first-forbidden electron capture to such a level, a minimum half life for decay of Pm¹⁵⁰ to Sm¹⁵⁰ is found to be twenty times greater than that known for the negatron decay to Nd¹⁵⁰.

ACKNOWLEDGMENTS

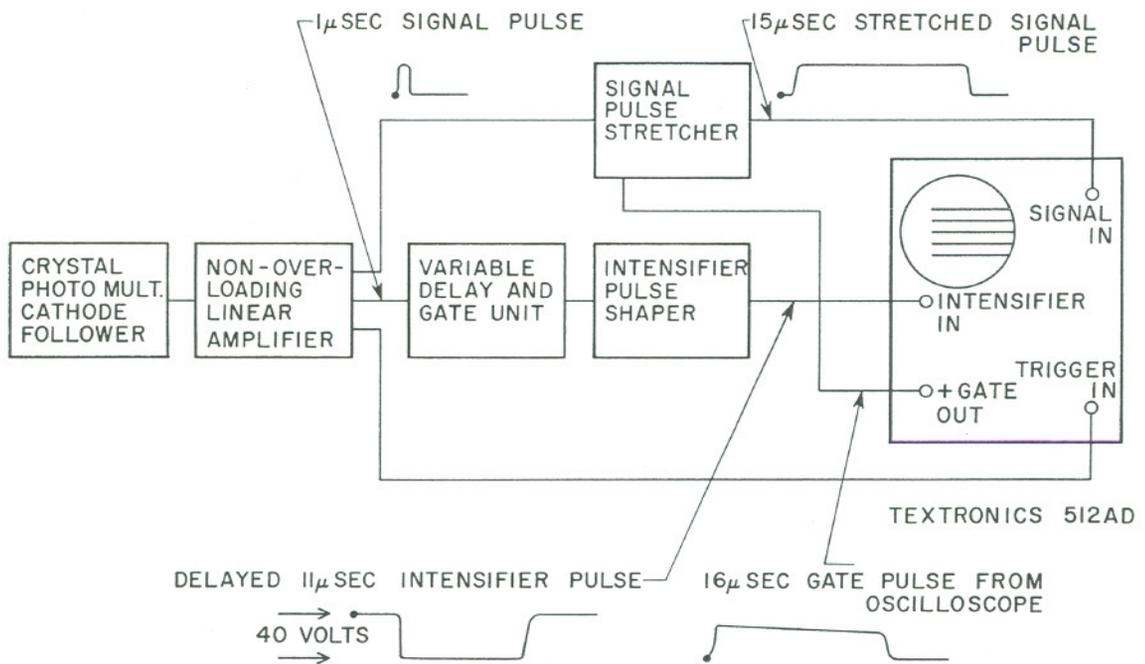
It is a pleasure to acknowledge some valuable discussions with Professor J. O. Rasmussen, Jr., and a fruitful correspondence with Professor T. P. Kohman. I also wish to thank Dr. B. T. Youtz and Mr. L. T. Kerth for the grey wedge analyzer design and the Isotope Research and Development Division, Oak Ridge, Tennessee, for making available to me the enriched isotope that made this work possible.

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18. G. I. Mulholland and T. P. Kohman, Phys. Rev. 85, 144 (1952).
 19. B. G. Hoagg and H. E. Duckworth, Can. J. Phys. 32, 65 (1954).



MU-7907

Fig. 1 Results of Hibdon and Muehlhause⁴ for the lowest levels of $^{150}_{62}\text{Sm}$.



MU-7417

Fig. 2 Block diagram of grey wedge pulse-height analyzer electronics.

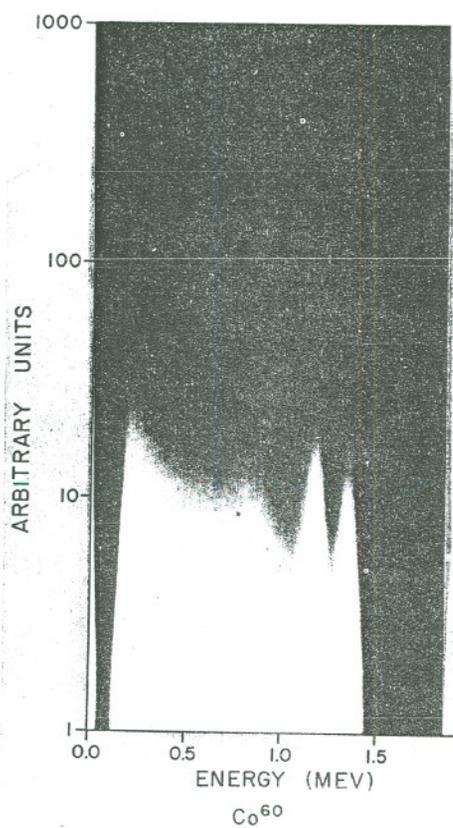
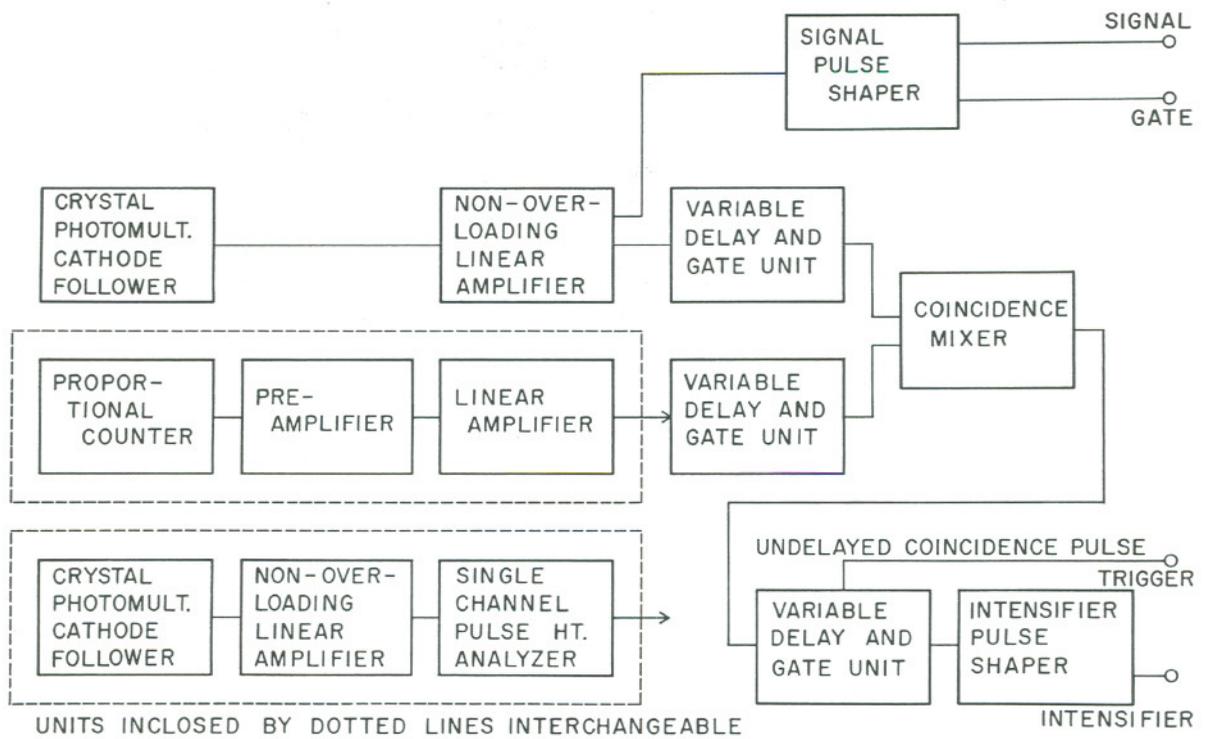


Fig. 3 Gamma-ray spectrum of Co^{60} .



MU-7416

Fig. 4 Block diagram of coincidence experiment electronics.

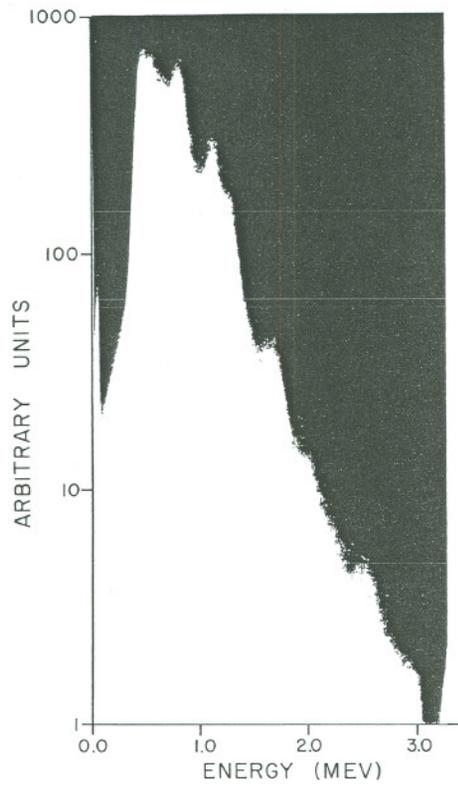
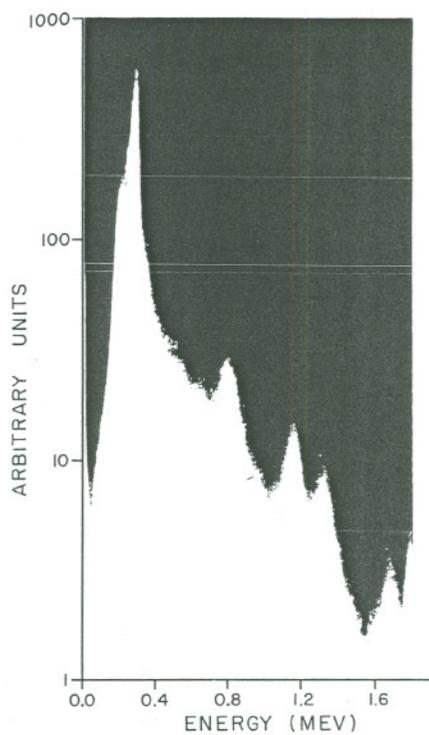


Fig. 5

Gamma-ray spectrum of Pm^{150} ;
higher energies.



MU-7419

Fig. 6 Gamma-ray spectrum of Pm^{150} ; intermediate energies.

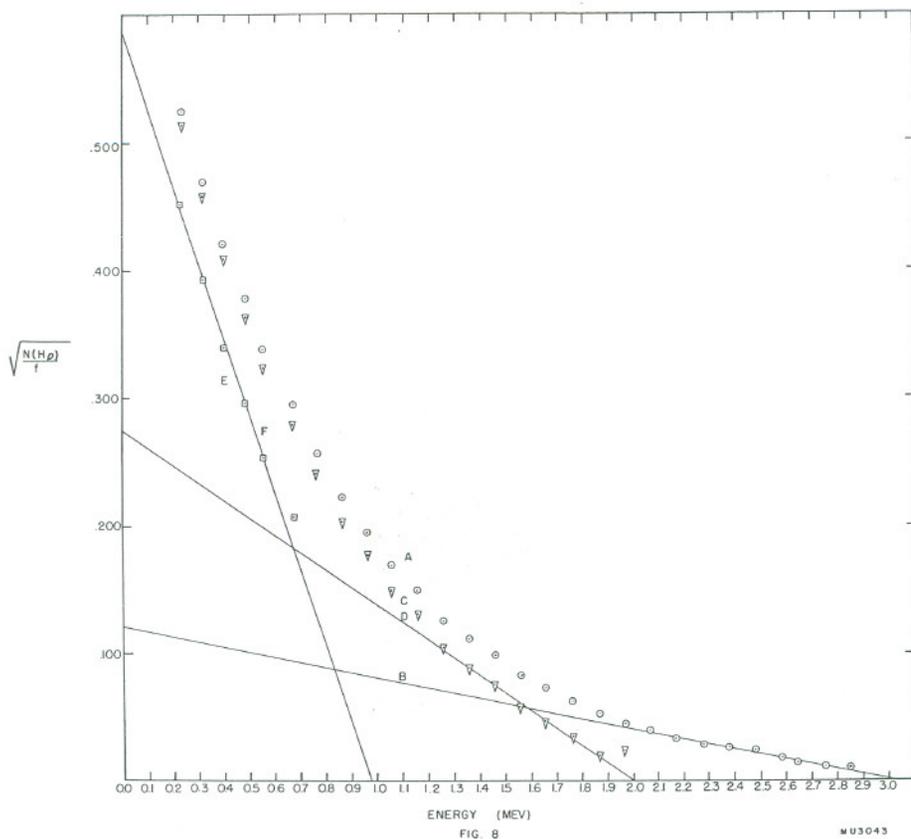
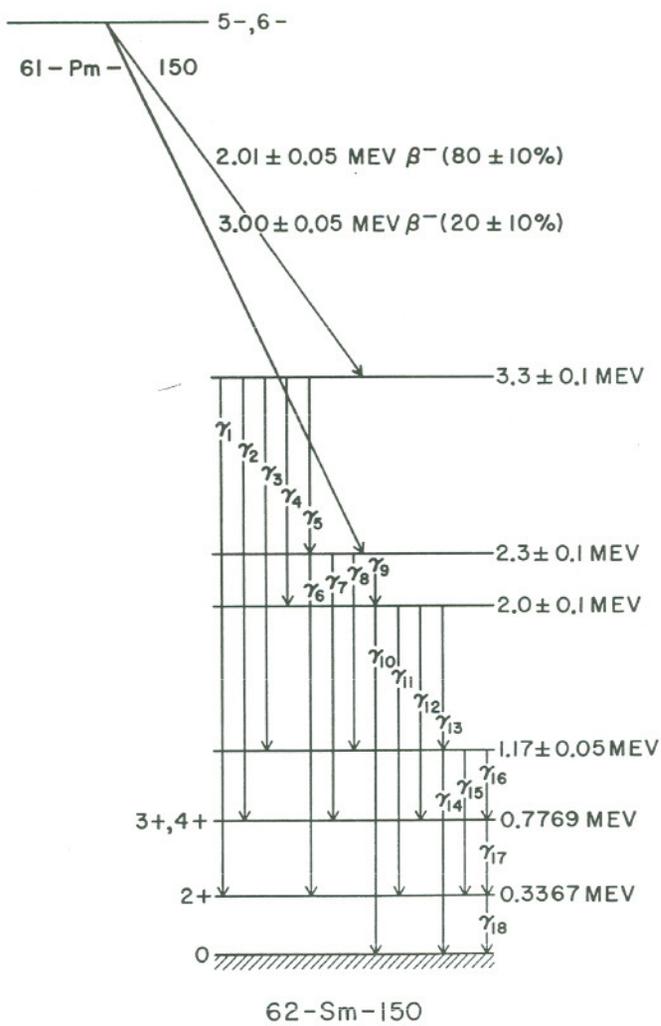


Fig. 7 Fermi-Kurie plot of beta-ray spectrometer data for the 161-minute and 54-hour half life activities from $Nd^{150} + p$ bombardments.

- (a) From gross data corrected for 161-minute half life decay.
- (b) 3-Mev component.
- (c) From gross data with 3-Mev component subtracted.
- (d) 2-Mev component.
- (e) From gross data with 2-Mev and 3-Mev components subtracted and corrected for 54-hour half life decay.
- (f) 0.97-Mev component.



MI-7908

Fig. 8 Decay scheme proposed for Pm^{150} .