Decay of Pm^{150}†*

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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.

INTRODUCTION

IT has been shown by Long and Pool[1] and this author[2] that Pm^{150} decays to Sm^{150} by negatron emission with a 161-minute half-life. Hibdon and Muehlhause[3] 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.

Two considerations prompted a further study of the decay of Pm^{150}. First, the Bohr-Mottelson[4] 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 Kohman[5] 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 microampere. The (p,n) and $(p,2n)$ reactions occur with comparable cross sections (~2 millibarns), 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 them 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.

Apparatus

Grey Wedge Analyzer

The reader is referred to Bernstein, Chase, and Schardt[6] 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-μsec delay-line clipping necessary for the coincidence work and a cathode follower output.

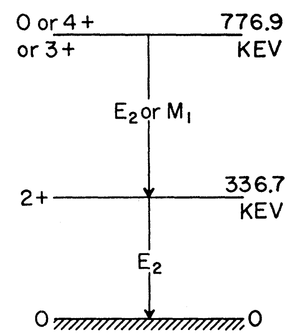

Fig. 1. Results of Hibdon and Muehlhause (see reference 4) for the lowest levels of $_{62}Sm^{150}$.

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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).

4 A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

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

6 Bernstein, Chase, and Schardt, Rev. Sci. Instr. **24**, 437 (1953).

The pulses are amplified by a standard nonoverloading UCRL linear amplifier. They then trigger the sweep of a Textronics 512AD oscilloscope, the output 16-μsec 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 μsec 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-μsec 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 Textronics oscilloscope is equipped with a 5*X*P11-*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. 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 $\frac{1}{2}$ inch long, was used for lower energy portions of the spectrum. Beryllium absorbers of 1500 mg/cm^2 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

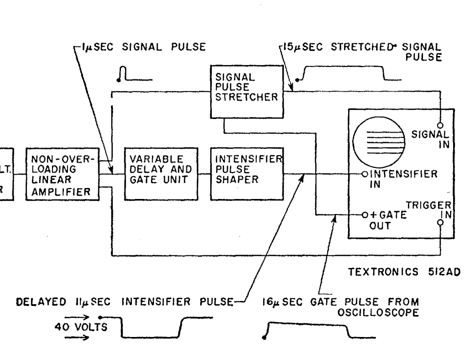

FIG. 2. Block diagram of grey wedge pulse height analyzer electronics.

[7] Obtained from the Harshaw Chemical Company, Cleveland 6, Ohio.

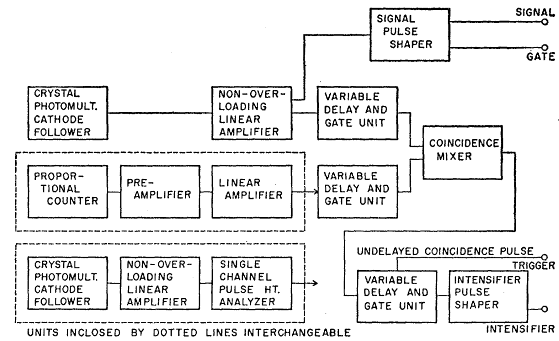

FIG. 3. Block diagram of coincidence experiment electronics.

checked roughly before each series of pictures for the given experimental conditions. Energy calibration was made before each experiment by using samples of Co^{56}, Co^{60}, Na^{22}, Cs^{137}, Cd^{109}, and Am^{241}, and some points were retaken at intervals to check for drifts. The distributions of the peaks in such pictures were 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 3 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 height analyzer which examines the pulse distribution from a second scintillation counter. The resolving time of the arrangement is ~ 3 μsec, placing a severe limitation on the count rate allowable. In all cases experiments were performed with maximum possible grometry: 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 coincidence 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 μsec (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 indicated the presence of gamma rays up to ~2.5 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 4 shows one of these pictures, and Fig. 5 contains the spectrum obtained with twice the linear amplifier gain of Fig. 4.

The results are summarized in Table I. Three dots 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.

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 $(160_{-60}{}^{+160})$ 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,

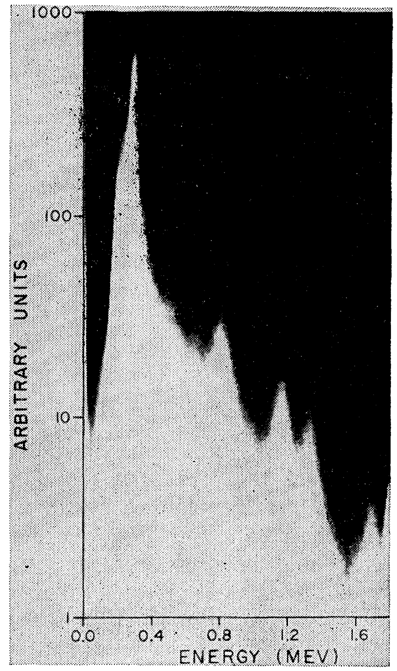

FIG. 5. Gamma-ray spectrum of Pm^{150}; intermediate energies.

no decay scheme agreeing with all the experimental results could be formulated. Therefore the old beta-ray spectroscopy results were re-examined 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. 6, 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 $(20\pm10$ percent) and $(80\pm10$ percent) respectively for the 3.00- and 2.01-Mev components.

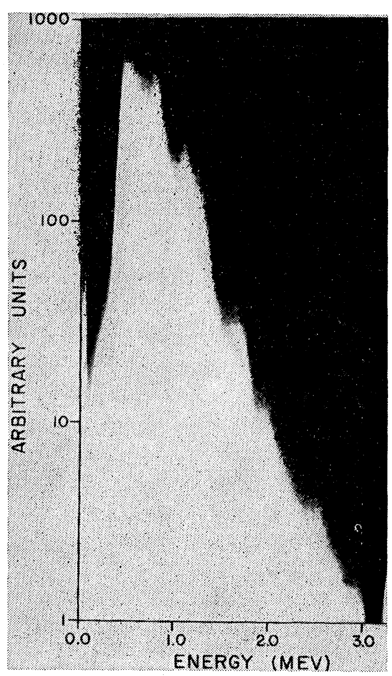

FIG. 4. Gamma-ray spectrum of Pm^{150}; higher energies.

CONCLUSIONS

Figure 7 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

$$E_{(3.0\text{ Mev})}-E_{(2.6\text{ Mev})}=0.40\pm0.05,$$
$$E_{(2.6\text{ Mev})}-E_{(2.0\text{ Mev})}=0.57\pm0.05.$$

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.

TABLE I. Energies of observed gamma rays.

Energy (Mev)	Approximate relative abundance	Occurrence in decay scheme	Observations: No coincidence	Coincidence 2-Mev $\beta-$	Coincidence 3-Mev $\beta-$	Coincidence 0.34-Mev γ
3.0 ± 0.1	0.004	γ_1	O	Distribution	···	Distribution
2.6 ± 0.1	0.008	γ_2	O	of pulses up to	···	of pulses up to
2.0 ± 0.1	0.004	$\gamma_3, \gamma_6, \gamma_{10}$	O	$\sim$ 2.5 Mev	···	$\sim$2.0 Mev
1.67 ± 0.05	0.008	γ_7, γ_{11}	O	O	···	O
1.32 ± 0.05	0.04	γ_4	O	O	O	O
?1.24	···	γ_{12}	···	···	O?	···
1.17 ± 0.05	0.04	$\gamma_8, \gamma_{12}, \gamma_{14}$	O	O	···	N
?0.96	···	γ_5	O?	···	···	···
0.82 ± 0.02	0.4	γ_{13}, γ_{15}	O	O	O	O
0.43 ± 0.02	0.2	γ_{17}	O	O	···	O
?0.39	···	γ_9, γ_{16}	···	···	O?	O?
0.34 ± 0.01	1.0	γ_9, γ_{18}	O	O	···	N

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's[8] 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's[9] rules for odd-odd isotopes are used in conjunction with Schwartz's[10] 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.

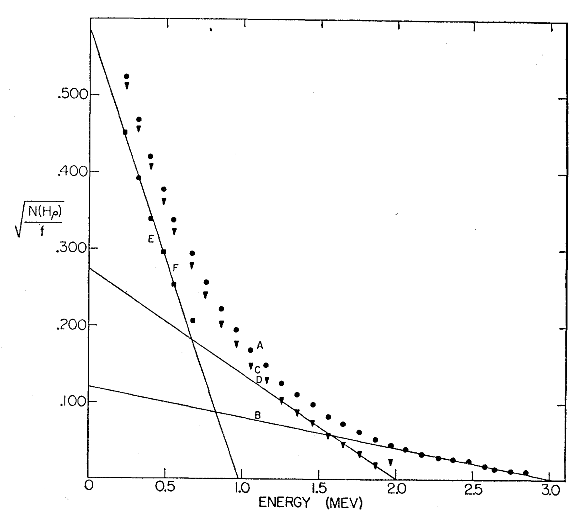

FIG. 6. Fermi-Kurie plot of beta-ray spectrometer data for the 161-minute and 54-hour half-life activities from $Nd^{150}+p$ bombardments: (a), from gross data corrected for 161-minute half-life decay; (b), 3-Mev component; (c), from gross data with 3-Mev component subtracted; (d), 2-Mev component; (e), from gross data with 2-Mev and 3-Mev components subtracted and corrected for 54-hour half-life decay; (f), 0.97-Mev component.

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 by 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.

On the basis of the experimental results no spin and parity assignments can be made. However, the observation that γ_1 and γ_2 have abundances of the same order of magnitude is not compatible with a difference of two or more between the spin changes occurring in these two transitions. This follows from a calculation of the transition probabilities as given in Blatt and Weisskopf.[11] In several recent compilations[4,12,13] of data on even-even isotopes, the first two excited levels of Sm^{150} are assigned to 2+ and 4+ states. This is the best interpretation of the results of Hibdon and Muehlhause,[3] but they do not rule out assignment of 3+ to the second excited level. In these papers[4,12,13] where this level is considered as a 4+ rotational state, the deviation of the ratio of the energies of the first two excited states from the value predicted by the

[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).

[11] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 627
[12] Aage Bohr, *Rotational States of Atomic Nuclei* (Ejnar Munksgaards Forlag, København, 1954).
[13] E. L. Church and M. Goldhaber (to be published, 1954).

formula for rotational levels,[12]

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

is accounted for by an increase in nuclear deformation due to a vibration-rotation type of interaction. The correction term[12] is

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

By calculating on the assumption that $E_2=337$ kev and $E_4=777$ kev, it is found that ΔE_I is 1.2 percent and 39 percent of E_I for E_2 and E_4 respectively. Thus the criterion for the existence of a rotational spectrum,[12] 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 second excited state of Sm^{150} is a 3+ rather than a 4+ state. Since the first excited state is 2+, the relative abundance of γ_1 and γ_2 is then much more easily explained.

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., ${}_{62}Sm^{150}-{}_{62}Sm^{152}$ and ${}_{63}Eu^{151}-{}_{63}Eu^{153}$,[14] and ${}_{60}Nd^{148}-{}_{60}Nd^{150}$,[15] suggest that some change in nuclear structure takes place between these neutron numbers. Rasmussen[16] has pointed out that the large quadrupole moment of ${}_{63}Eu^{153}$ 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.

Stability of Nd^{150}

Kohman[5] has pointed out that Nd^{150} would be expected to be unstable with respect to negatron decay to Pm^{150}. However, Mulholland and Kohman[17] 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 Duckworth[18] have obtained a Nd^{150}-Sm^{150} mass difference of 4.6 ± 0.8 Mev. Since the proposed Pm^{150}-Sm^{150} ground-state energy difference is 5.3 ± 0.15 Mev, the Nd^{150}-Pm^{150} difference is -0.7 ± 1.0 Mev and Nd^{150} may or may not be stable. If it is not, then it is unstable by at most 0.3 Mev. If one accepts the previous arguments for an assignment of negative parity and spin 5 or 6 to be the ground state of Pm^{150}, 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, Pm^{150} is unstable with respect to Nd^{150}, this mode of decay would not be detected. If one assumes the 4+ level of ${}_{60}Nd^{150}{}_{90}$ to be analogous to that of ${}_{62}Sm^{152}{}_{90}$, it would be at 0.37 Mev.[13] Assuming first-forbidden electron capture to such a level, a minimum half-life for decay of Pm^{150} to Sm^{150} is found to be twenty times greater than that known for the negatron decay to Nd^{150}.

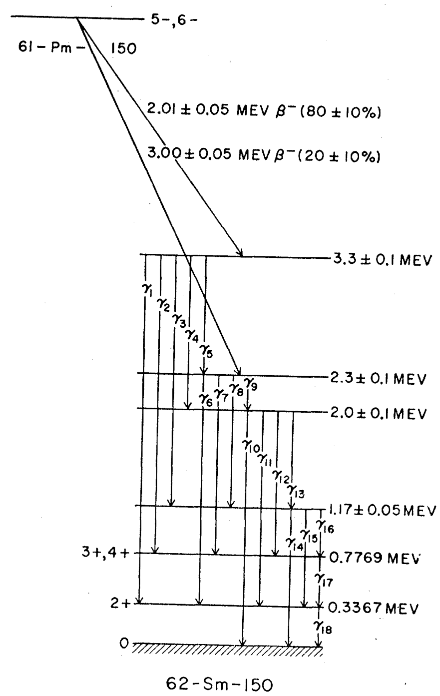

FIG. 7. Decay scheme proposed for Pm^{150}.

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[15] P. F. A. Klinkenberg, Physica **11**, 327 (1945).
[16] J. O. Rasmussen, Jr., Arkiv Fysik **7**, 185 (1953).
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[18] B. G. Hoagg and H. E. Duckworth, Can. J. Phys. **32**, 65 (1954).

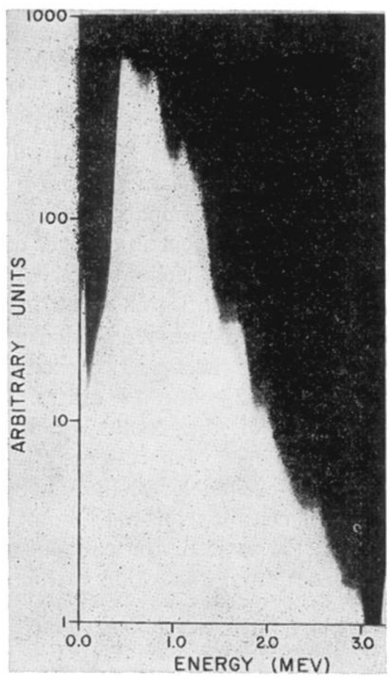

FIG. 4. Gamma-ray spectrum of Pm^{150}; higher energies.

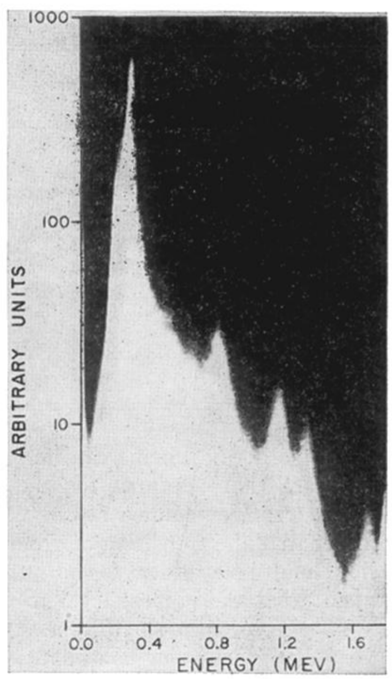

FIG. 5. Gamma-ray spectrum of Pm^{150}; intermediate energies.