Directional Correlation of Gamma Transitions in Promethium-147

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The angular correlations of five different gamma-ray cascades, involving the states of Pm^{147}, excited in the decay of Nd^{147}, have been studied. The observed correlation functions for the various cascades are as follows:

$$W(_{599}\theta_{91}) = 1+(0.056\pm0.028)P_2(\cos\theta)-(0.049\mp0.034)P_4(\cos\theta);$$
$$W(_{442}\theta_{91}) = 1+(0.065\pm0.020)P_2(\cos\theta)-(0.035\mp0.025)P_4(\cos\theta);$$
$$W(_{199}\theta_{400}) = 1-(0.054\pm0.020)P_2(\cos\theta)+(0.043\mp0.025)P_4(\cos\theta);$$
$$W(_{199}\theta_{491}) = 1+(0.095\pm0.028)P_2(\cos\theta)+(0.014\mp0.034)P_4(\cos\theta);$$

and

$$W(_{120}\theta_{322}) = 1+(0.035\pm0.020)P_2(\cos\theta)-(0.001\mp0.025)P_4(\cos\theta).$$

The analysis of the above correlation functions with the ground-state spin of Pm^{147} as $\frac{7}{2}$, and the consideration of the logft values for the beta transitions from Nd^{147} state of spin $\frac{5}{2}$, give the spin values for the 91, 413, 491, 533, and 690-kev excited states as $\frac{5}{2}$, $\frac{7}{2}$, $\frac{7}{2}$, $\frac{5}{2}$, and $\frac{5}{2}$, respectively. The data of Bishop *et al.* and Ambler *et al.* on nuclear alignment experiments have been reanalyzed. The results are consistent with the above spin assignment of $\frac{5}{2}$ for both the 91- and 533-kev levels.

INTRODUCTION

THE excited states of Pm^{147} at 91, 413, 491, 533, 690, and 723 kev, which are populated in the decay of Nd^{147}, are shown[1] in Fig. 1. The ground-state spin of Nd^{147} has been measured by Kedzie *et al.*[2] by paramagnetic resonance method and by Cabezas *et al.*[3] using the atomic beam method. Both the authors have assigned the spin $\frac{5}{2}$ for this nucleus. The ground-state spin of Pm^{147} has been measured by Klinkenberg and Tomkins[4] by optical spectroscopy as $\frac{7}{2}$. The angular correlation of the 322–91 kev gamma-ray cascade was studied by Lindquist and Karlsson.[5] Recently, Bodenstedt *et al.*[6] have investigated the angular correlation of seven different gamma-ray cascades; and have assigned the spin values for the excited states at 91, 413, 491, 533, and 690 kev as $\frac{5}{2}$, $\frac{5}{2}$ or $\frac{7}{2}$, $\frac{5}{2}$ or $\frac{7}{2}$, $\frac{3}{2}$ or $\frac{7}{2}$, and $\frac{3}{2}$ or $\frac{7}{2}$, respectively. It has been shown that there is an unresolved gamma-ray peak at 310 kev, between the 277- and 322-kev gamma-ray peaks in the scintillation spectrum, and that this gamma ray is in coincidence with the 91, 322, and 413-kev gamma rays.[1] Hence some of the correlations of the previous workers[6] are distorted by the interfering cascades and their conclusions could be in error.

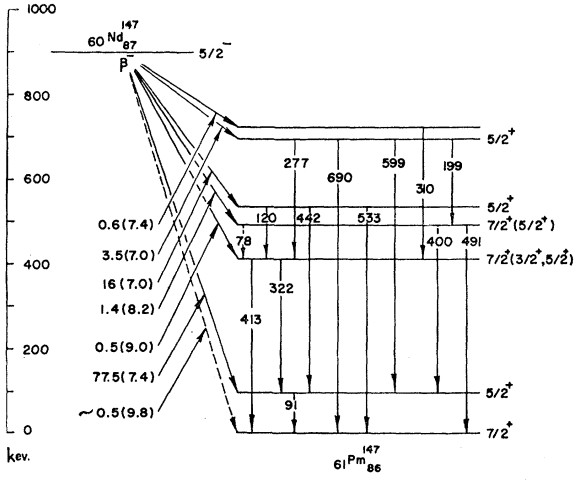

FIG. 1. Energy levels of Pm^{147} excited in the decay of Nd^{147}, with the probable spin values. The intensities of the beta transitions in percent followed by logft values in parentheses are also given.

The angular distribution of the 91- and 533-kev gamma rays, emitted from aligned nuclei of Nd^{147}, has been studied by Ambler *et al.*[7] The polarization asymmetry and anisotropy of the above gamma rays emitted from the aligned state have been studied by Bishop *et al.*[8] For their analysis, these authors used the spin values of 9/2 and $\frac{5}{2}$ for the ground states of Nd^{147} and Pm^{147}. In view of the now measured spin values of $\frac{5}{2}$ and $\frac{7}{2}$ for these nuclei, respectively, their conclusions regarding the spins of the 91- and 533-kev excited states, and other properties of the gamma transitions and nuclear states, are in error.

In view of this situation, a study of the angular correlations of relatively clean gamma-ray cascades was undertaken; and the results of the nuclear alignment experiments have been reanalyzed and interpreted.

[1] M. R. Gunye, R. Jambunathan, and B. Saraf, Phys. Rev. **123**, 172 (1961), preceding paper.
[2] R. W. Kedzie, M. Abraham, and C. D. Jeffries, Phys. Rev. **108**, 54 (1957).
[3] A. Cabezas, I. Lindgren, E. Lipworth, R. Marrus, and M. Rubinstein, Nuclear Phys. **20**, 509 (1960).
[4] P. F. A. Klinkenberg and F. S. Tomkins, Physica **26**, 103 (1960).
[5] T. Lindquist and E. Karlsson, Arkiv Fysik **12**, 519 (1957).
[6] E. Bodenstedt, H. J. Körner, F. Friesius, D. Hovestadt, and E. Gerdau, Z. Physik **160**, 33 (1960).
[7] E. Ambler, P. R. Hudson, and G. M. Temmer, Phys. Rev. **97**, 1212 (1955).
[8] G. R. Bishop, M. A. Grace, C. E. Johnson, H. R. Lemmer, and J. Perez y Jorba, Phil. Mag. **2**, 534 (1957).

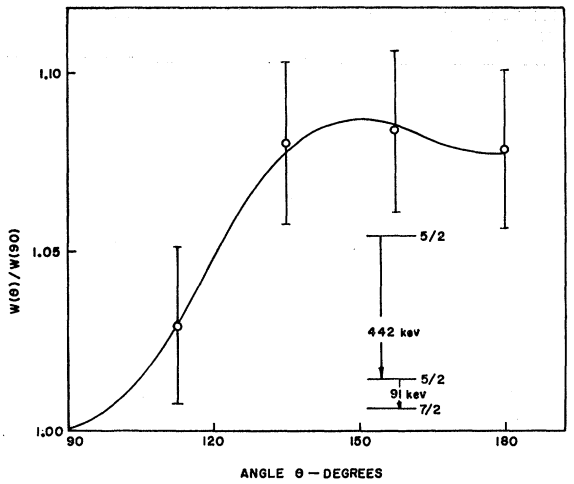

FIG. 2. The asymmetry parameter $W(\theta)/W(90°)$ at various angles and the least-squares fit curve of the form, $W(\theta)=1+0.234\cos^2\theta-0.157\cos^4\theta$, for the 442–91 kev gamma-ray cascade.

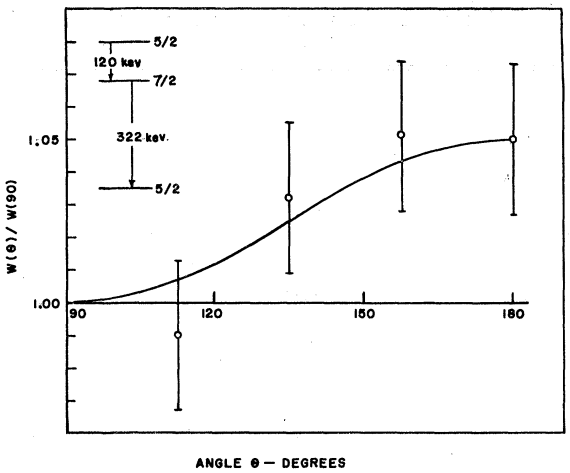

FIG. 4. The asymmetry parameter $W(\theta)/W(90°)$ at various angles and the least-squares fit curve, $W(\theta)=1+0.055\cos^2\theta-0.004\cos^4\theta$, for the 120–322 kev gamma-ray cascade.

EXPERIMENTAL PROCEDURE

Sources of Nd^{147} were obtained by irradiating specpure samples of neodymium oxide in Apsara reactor (Trombay) for a period of 12 to 15 days. In some cases, the active samples were purified in an ion exchange column to remove other active impurities; while in other cases the samples were allowed to decay for over a month to suppress the Pm^{149} and Pm^{151} activities.

The angular correlation studies have been carried out with two scintillation spectrometers having 1.75-in. diam×2-in. thick NaI(Tl) crystals as gamma detectors and single-channel analyzers for selection of pulse heights in the proper energy region. A standard fast-slow coincidence circuit with resolving time $2\tau=0.04$ μsec was employed in the experiments. The crystals were surrounded with lead cones of thickness 1.5 cm, having a conical aperture of half-angle about 8° viewing the source mounted along the axis of the moving arm. Active neodymium chloride solution, in water, in a Perspex container with a cavity of 5-mm diam and 2-mm wall thickness was used as source in these experiments. Thus the perturbation of the angular correlation, if any, where the 91-kev level having a half-life of 2.4×10^{-9} sec forms the intermediate state, was avoided. In the study of angular correlations of the 199–400 kev and the 199–491 kev gamma-ray cascades, a 3-mm lead absorber was placed just upon the crystal detecting the 400-kev or 491-kev gamma rays. In this way the coincidences due to the back-scattering radiation from the crystal detecting the higher energy gamma rays, into the other crystal, were minimized.

In all, five different cascades of gamma transitions,

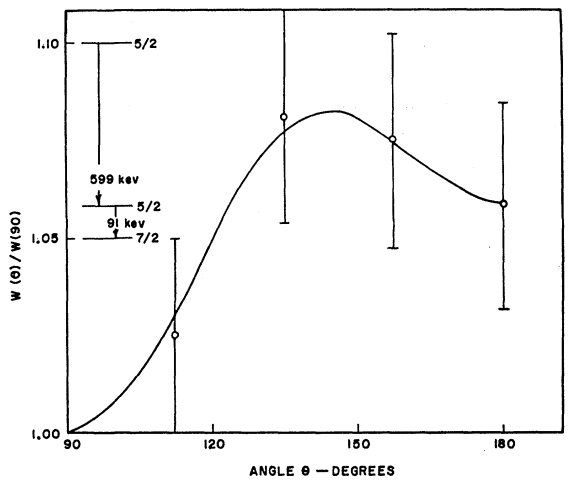

FIG. 3. The asymmetry parameter $W(\theta)/W(90°)$ at various angles and the least-squares fit curve, $W(\theta)=1+0.250\cos^2\theta-0.191\cos^4\theta$, for the 599–91 kev gamma-ray cascade.

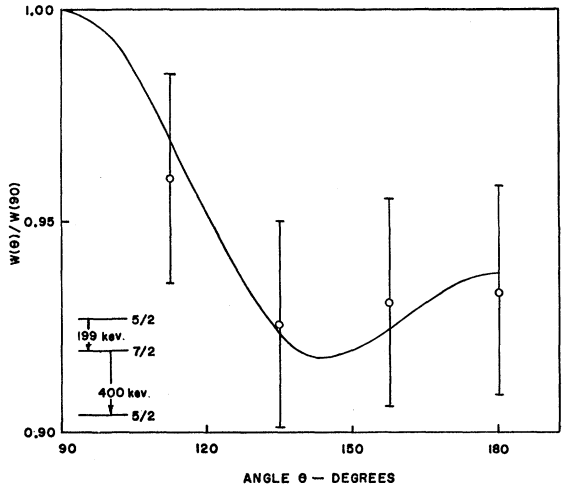

FIG. 5. The asymmetry parameter $W(\theta)/W(90°)$ at various angles and the least-squares fit curve, $W(\theta)=1-0.210\cos^2\theta+0.156\cos^4\theta$, for the 199–400 kev gamma-ray cascade.

TABLE I. Details of the angular correlation studies of the various gamma-ray cascades and the corrected coefficients of the $P_2(\cos\theta)$ and $P_4(\cos\theta)$ terms in the corresponding correlation function $W(\theta)$.

Gamma-ray cascade	Accepted energy intervals in channels—kev (A)	(B)	N_{12}[a]	Corrected coefficients of the terms in the correlation functions $W(\theta)$. $P_2(\cos\theta)$	$P_4(\cos\theta)$
442– 91	80– 95	430–460	5000	$+(0.065\pm0.020)$	$-(0.035\mp0.025)$
599– 91	85– 95	585–615	2000	$+(0.056\pm0.028)$	$-(0.049\mp0.034)$
199–400	190–210	390–420	5000	$-(0.054\pm0.020)$	$+(0.043\mp0.025)$
199–491	190–210	475–500	2000	$+(0.095\pm0.028)$	$+(0.014\mp0.034)$
120–322	120–135	300–340	5000	$+(0.035\pm0.020)$	$-(0.001\mp0.025)$

[a] N_{12} denotes the total number of coincidences collected at each angle.

involving the five excited states of Pm^{147}, have been studied. In the study of all the correlations, the number of coincidences were recorded at each of the angles in succession for 10 minutes and the set repeated. The influence of any drift in the equipment is thus minimized; also the correction for the source decay was reduced to less than 0.3%. The integrated values of asymmetry, $W(\theta)/W(90°)$, at various angles for the different cascades, are shown in Figs. 2 to 6. The data were analyzed by the least-squares fit method for the $P_2(\cos\theta)$ and $P_4(\cos\theta)$ terms in the correlation functions, which were further corrected for the angular resolution of the detectors. For each correlation study, the number of coincidences collected at each angle, the accepted energy intervals in both the channels, and the coefficients of the $P_2(\cos\theta)$ and $P_4(\cos\theta)$ terms obtained from the analysis are summarized in Table I. The coincidence spectra reported in reference 1 show that there are no major contributions of the gamma-ray coincidences other than those of interest, when the detection channels are fixed in the regions shown in the table. In one particular cascade of 442–91 kev gamma rays, the interference due to the 400–91 kev gamma-ray cascade is suppressed by fixing the channel above 430 kev. However, no correction has been made for the little interference that might be present.

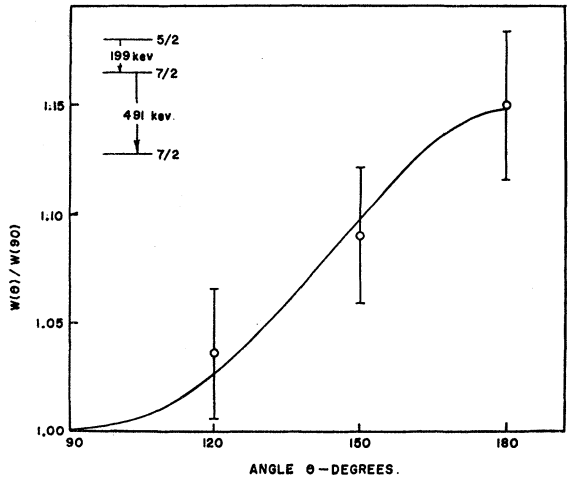

FIG. 6. The asymmetry parameter $W(\theta)/W(90°)$ at various angles and the least-squares fit curve, $W(\theta)=1+0.095\cos^2\theta+0.052\cos^4\theta$, for the 199–491 kev gamma-ray cascade.

TABLE II. Results of the analysis of the various gamma-gamma angular correlation functions. Nature of the gamma transitions for the three possible spin sequences are given.

Energy level (kev)	Gamma rays (kev)	Range of the permissible $E2$ components in gamma transitions for different spin sequences: Sequence A Spin	Sequence A $E2$%	Sequence B Spin	Sequence B $E2$%	Sequence C Spin	Sequence C $E2$%	$(E2\%)$[a]
690	599	$\frac{5}{2}$	98–100	$\frac{5}{2}$	98–100	$\frac{9}{2}$	80–90	
	199		10– 30		20– 40		15–25	10
533	442	$\frac{5}{2}$	98–100	$\frac{5}{2}$	98–100	$\frac{9}{2}$	85–95	50
	120		10– 25		0– 20		100	2
			or 80–100		or 90– 95	(if 413-kev level is $\frac{5}{2}$)		
		(if 413-kev level is $\frac{7}{2}$)						
							0– 10	
			0– 10				or 94–100	
			or 100			(if 413-kev level is $\frac{7}{2}$)		
		(if 413-kev level is $\frac{3}{2}$)						
491	491	$\frac{7}{2}$	70– 85	$\frac{5}{2}$	80– 90	$\frac{7}{2}$	60–80	...
	400		97–100		95–100		85–95	...
413	322	$\frac{7}{2}$	80–100	$\frac{5}{2}$	0– 20	$\frac{7}{2}$	94–100	
			or 10– 25		or 90– 95		or 0– 10	
		or $\frac{3}{2}$	100		...	or $\frac{5}{2}$	100	
			or 0– 10					
91	91	$\frac{5}{2}$	25–100	$\frac{5}{2}$	25–100	$\frac{9}{2}$	5– 25	1
0	0	$\frac{7}{2}$	...	$\frac{7}{2}$	...	$\frac{7}{2}$	...	

[a] The values of $E2$ components are recently reported by Ewan *et al.*[9] for some of the gamma transitions from the study of the internal conversion measurements.

TABLE III. Results of the analysis of Bishop's work on aligned Nd^{147} nuclei. The values of p/ϵ for the 533- and 91-kev transitions were taken as $-0.68_{-0.20}^{+0.08}$ and $+0.05\pm0.025$, respectively.

Spin of the 533- or 91-kev level	Permissible values of δ for the 533-kev radiation and corresponding values of $B_2(T)$		Permissible values of δ for the 91-kev radiation and the corresponding values of $B_2(T)$	
	δ	$B_2(T)$	δ	$B_2(T)$
$\frac{5}{2}$	-0.64[a]	0.38	0.0 to 0.12	0.75 to 0.34
$\frac{7}{2}$	1.6 to 1.85	0.16 to 0.17	(Solution suppressed)	
9/2	0.56 to 0.9	0.24 to 0.13	-0.13 to 0.07	0.15 to 0.19

[a] Solution was obtained for only lowest limit of $p/\epsilon=-0.88$.

ANALYSIS OF THE DATA

The ground-state spin of Nd^{147} is $\frac{5}{2}$. The $\log ft$ values for the beta transitions to the various excited states of Pm^{147} lie between 7.0 and 9.0. Hence it appears that all beta transitions are of first-forbidden nature, of the type $\Delta I=0$ or ±1, yes; and the spin of the various levels could be $\frac{3}{2}$, $\frac{5}{2}$ or $\frac{7}{2}$. To allow for the possibility of some abnormally low $\log ft$ value for a $\Delta I=\pm2$, yes type of first-forbidden transition, the spin values of $\frac{1}{2}$ and 9/2 were also considered for any of the states. The observed intensities of the gamma transitions from all the excited states to the ground state of spin $\frac{7}{2}$, rule out the spin value $\frac{1}{2}$ for any of the excited levels. Hence a detailed analysis of the angular correlation data with the spin values $\frac{3}{2}$, $\frac{5}{2}$, $\frac{7}{2}$, or 9/2 for each state has been carried out. The gamma transitions were considered as mixtures of $M1$ and $E2$ wherever possible.

The correlation function for the 442–91 kev gamma-ray cascade is analyzed first. The positive sign of the coefficient of $P_4(\cos\theta)$ term is outside the error region, since the maximum possible value is -0.01. This eliminates the possibility of spin $\frac{3}{2}$ for the 91-kev level and $\frac{3}{2}$ or $\frac{7}{2}$ for the 533-kev level. Similarly, the negative sign of the coefficient of $P_4(\cos\theta)$ term in the correlation function for the 599–91 kev gamma ray cascade, suggests the spin value $\frac{5}{2}$ or 9/2 alone for the 690-kev level. On the other hand, the positive sign of the coefficient of $P_4(\cos\theta)$ term in the 199–400 kev gamma-ray correlation function rules out the spin value $\frac{7}{2}$ for the 91-kev level, if the 690-kev level is assigned the spin value $\frac{5}{2}$ or 9/2, irrespective of the spin value for the 491-kev level. Thus there are four spin sequences possible for the 442–91 and 599–91 kev gamma-ray cascades, which are as follows: (a) $\frac{5}{2}-\frac{5}{2}-\frac{7}{2}$; (b) $9/2-9/2-\frac{7}{2}$; (c) $\frac{5}{2}-9/2-\frac{7}{2}$; and (d) $9/2-\frac{5}{2}-\frac{7}{2}$. In the cases (c) and (d), the first transition is pure $E2$ in nature; and no possible admixture of $M1$ and $E2$ in the 91-kev transition yields simultaneously the measured values of the coefficients of the $P_2(\cos\theta)$ and $P_4(\cos\theta)$ terms, within the permissible errors for the 442–91 and 599–91 kev gamma-ray correlation functions. Thus the spin values for the 91, 533, and 690-kev states could all be either $\frac{5}{2}$ or 9/2.

The observed correlation function for the 120–322 kev gamma-ray cascade does not allow the spin value 9/2 for the 413-kev state, if the 91- and 533-kev levels are both assigned the spin value $\frac{5}{2}$. Unfortunately all other spin values, namely $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$ give a suitable correlation function for proper admixture of $M1$ and $E2$ in the two transitions. Similarly, if the 91- and 533-kev levels are both assigned the spin value 9/2, the 413-kev state could have a spin value $\frac{5}{2}$, $\frac{7}{2}$, or 9/2.

The nonzero coefficient of the $P_4(\cos\theta)$ term in the 199–400 kev gamma-ray correlation function rules out the spin value $\frac{3}{2}$ for the 491-kev level. Since the coefficient of the $P_2(\cos\theta)$ term in the above correlation function is negative, the spin value 9/2 for the 491-kev level is not permissible if the 91- and 690-kev levels are both assigned the spin $\frac{5}{2}$; similarly, a spin $\frac{5}{2}$ for the 491-kev level is not permissible if the 91- and 690-kev states are both assigned the spin value 9/2. In the latter case, a spin 9/2 for the 491-kev level is ruled out by a detailed analysis of the 199–400 and 199–491 kev gamma-ray correlation functions. Hence, the spin of the 491-kev state could be $\frac{5}{2}$ or $\frac{7}{2}$, if the 91- and 690-kev levels are assigned the spin $\frac{5}{2}$; or it can have a spin $\frac{7}{2}$, if the latter two levels are assigned the spin value 9/2.

The results of the analysis of all the correlation functions are summarized in Table II. The possible admixtures of $M1$ and $E2$ in the various transitions are given. The nature of the various gamma transitions, as obtained from the internal conversion data by Ewan *et al.*,[9] are also given in the same table for comparison.

Since the information derived from the study of the 533- and 91-kev gamma emissions from the aligned nuclei of Nd^{147}, as reported,[7,8] is not tenable, the observed data of these experiments have been reanalyzed. A summary of the observed quantities and the information derived from them with the now known spin values of $\frac{5}{2}$ and $\frac{7}{2}$ for the Nd^{147} and Pm^{147} nuclei, respectively, is reported here.

ANALYSIS OF THE NUCLEAR ALIGNMENT EXPERIMENTS ON Nd^{147}

The Nd^{147} nuclei were aligned by Bishop *et al.*[8] in a crystal of neodymium ethyl sulfate along the axis. At the low temperature of 1/40°K, the anisotropy ϵ [defined as $1-I(0)/I(\pi/2)$], for the 533- and 91-kev gamma radiations, was found to be $+0.200\pm0.005$ and -0.093 ± 0.005, respectively. The polarization parameter p (obtained after taking into account the efficiency

[9] G. T. Ewan, R. L. Graham, and J. S. Geiger, Bull. Am. Phys. Soc. **6**, 238 (1961).

TABLE IV. The observed parameters of the 533-kev gamma emission from aligned Nd^{147} nuclei by Ambler *et al.* at $T=0.00308°K$, are compared with the calculated values for the permissible values of spin for the 533-kev level and the δ for the gamma emission and at $T=0°K$.

Spin of 533-kev level	δ for the 533-kev gamma ray	Theoretically calculated values[a] for total alignment at $T=0°K$. $I(0°)$	$I(90°)$	$-\epsilon(90°)$	$-\epsilon(50°)$
$\frac{5}{2}$	-0.64	1.31	0.85	0.54	0.26 (for $\Delta I_\beta=1$)
		1.51	0.77	0.96	0.41 (for $\Delta I_\beta=0$)
$\frac{7}{2}$	1.60–1.84	1.44–1.40	0.94–0.97	0.53–0.44	0.50–0.48
9/2	0.56–0.90	1.75–1.55	0.72–0.78	1.43–1.03	0.71–0.47
Experimentally observed[b] values at 0.00308°K		1.21	0.87	0.39	0.16

[a] $I(\theta)$ is defined as the ratio of gamma ray intensity at angle θ with the reference axis from the aligned system, to that from isotropic system. $\epsilon(\theta)$ is defined as $[1-I(0°)/I(\theta)]$.
[b] The experimentally observed values of $I(0°)$ and $I(90°)$ are estimated from Fig. 11 of reference 7.

of the polarimeter) was measured as $-0.136_{-0.040}^{+0.017}$ and -0.055 ± 0.025 for the same gamma transitions, respectively. For different spin values of the 533- and 91-kev excited states, the mixing parameter δ for the $M1$ and $E2$ mixture in the gamma transitions as obtained from the above-measured quantities, and the corresponding values of the $B_2(T)$ functions,[10] are shown in Table III. Only those solutions are given for which an approximately common value of $B_2(T)$ function is obtained from the values of ϵ and p for the 533- and 91-kev radiations. In case of the 533-kev state having a spin $\frac{5}{2}$, it was observed that the mean value of $p/\epsilon(=-0.68)$ does not provide a real solution for δ. However, it has been found that just near the lower limit of the value for $p/\epsilon(=-0.88)$, a real solution is permissible and that is given in the table. The value of $B_2(T)$ functions, as given in Table III, are to be compared with the value of 0.39 which is calculated from the values of A and B, the hfs constants, for Nd^{147} in neodymium ethyl sulfate crystal as estimated by Kedzie *et al.*,[2] and for temperature $T=1/40°K$. However, it should be mentioned that the temperature value of 1/40°K is taken from Figs. 2 and 3 of reference 8 for proper values of p and ϵ; but in the text of the same reference the same values of p and ϵ are mentioned to be at $T=1/25°K$. Corresponding to this value of temperature, the value of $B_2(T)$ function is 0.23.

Ambler *et al.*[7] produced the alignment of the Nd^{147} nuclei in a single crystal of cerium magnesium nitrate. In this experiment the alignment was produced in a plane. The anisotropy of the 533- and 91-kev gamma radiation at the recorded temperature of 0.00308°K was studied. It was observed that the 91-kev gamma radiation is emitted isotropically within the limits of 5%. The 533-kev gamma radiation shows an enhanced emission of 21% along the direction normal to the plane of alignment. The anisotropy $\epsilon(90°)$, as defined earlier, of about -0.39 was observed. The anisotropy $\epsilon(50°)$ was found to be -0.16. For different spin values of the 533-kev level, the $E2$ and $M1$ mixing parameter δ was chosen from Bishop's work as given in Table III; and corresponding to those values of δ, the values of $I(0°)$, $I(90°)$, $\epsilon(90°)$, and $\epsilon(50°)$ were calculated for the ideal case of total alignment, i.e., $T=0°K$. These quantities are compared with the measured values in Table IV.

[10] N. R. Steenberg, Proc. Phys. Soc. (London) **A66**, 399 (1953); **A65**, 791 (1952).

CONCLUSIONS

The mixing ratios for the gamma transitions of energy 120, 199, and 322 kev, given by the analysis of the angular correlation data for the sequences A and B of Table II, are in fair agreement with the values obtained from the internal conversion data.[9] The value of 20% $E2$ component in the 322-kev transition is inconsistent with that for spin sequence "C." The mixing of $E2$ components in the 91- and 442-kev transitions, as obtained from angular correlation studies, is too large in comparison to the values obtained from internal conversion measurements. It has not been found possible to improve upon the accuracy of the 442–91 kev and 599–91 kev gamma-ray correlation functions to determine the $P_4(\cos\theta)$ terms more accurately.

A spin value 9/2 for the 91, 533, and 690-kev levels would make the beta transitions to these levels of the form $\Delta I=2$, yes; and consequently the observed $\log ft$ values of 7.4, 7.0, and 7.0 for these transitions would be all too low. This supports the assignment of spin sequence "A" and "B" of Table II.

Ambler's observations[7] for the gamma-ray intensity distribution, as given in Table IV, rule out the spin value $\frac{7}{2}$ for the 533-kev state; but the spin values $\frac{5}{2}$ and 9/2 are equally probable. The calculated value of 0.39 for the $B_2(T)$ function (for $T=1/40°K$), if compared with the values given in Table III, favors the spin value $\frac{5}{2}$ for both the 91- and 533-kev states.

On the basis of all these considerations, the preferred spin values for the excited states of Pm^{147} at 91, 413, 491, 533, and 690 kev energy are $\frac{5}{2}$, $\frac{7}{2}$, $\frac{7}{2}$, $\frac{5}{2}$, and $\frac{5}{2}$; and they are shown in Fig. 1. The sequence "B" of Table II with spin $\frac{5}{2}$ for all the states, although possible, is unlikely.

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