

Nuclear Orientation of $\text{Pm}^{143}\dagger$

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Promethium-143 has been oriented in a crystal of neodymium ethylsulfate. The angular distribution of the 740-keV γ ray was found to be $W(\theta) = 1 - (0.065 \pm 0.006)P_2(\cos\theta)$ at 0.02°K . Values for the mixing ratio, δ , of the 740-keV γ ray of Nd^{143} were obtained as a function of the magnetic moment of the ground state of Pm^{143} . The spin of the excited state of Nd^{143} was assigned as $9/2^-$. An absolute lower limit of $|\mu| > 1.0$ was set on the magnetic moment of Pm^{143} . The mixing ratio of the γ ray of Nd^{143} was found to lie in the range $0.23 < \delta(E2/M1) < 0.35$.

INTRODUCTION

SEVERAL authors have described the decay of Pm^{143} . The most detailed work was done by Ofer.¹ This investigation included γ -ray spectra, γ -ray-x-ray coincidence spectra, and internal-conversion coefficients. Ofer observed only one γ ray, which had an energy of 740 keV, and he determined that 45% of electron capture went to the excited level and 55% directly to the spin- $7/2^-$ ground state of Nd^{143} .

Recent experiments have shown that promethium nuclei can be oriented in the ethylsulfate lattice,² and we have carried out low-temperature nuclear orientation experiments to study the decay of Pm^{143} .

EXPERIMENTAL

Praseodymium oxide, Pr_2O_3 , was bombarded with 35-MeV helium ions to produce an $(\alpha, 2n)$ reaction. This energy was chosen to minimize the amount of Pm^{144} produced. The Pm^{3+} was separated from the Pr^{3+} on a cation-exchange column by the method described by Thompson et al.³ The PmCl_3 solution was evaporated to dryness and taken up in a neodymium ethylsulfate solution. A crystal of $\text{Nd}(\text{C}_2\text{H}_5\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$ weighing about 5 g was grown, incorporating Pm^{3+} into the Nd^{3+} lattice sites.

The crystal was mounted in a demagnetization cryostat with the c axis horizontal. The counter was mounted on a table which rolled along tracks in the floor. After demagnetization, the counter was moved into position and the table locked in place. Counting was done at demagnetization temperatures and then, for normalization, at the helium bath temperature. A 3-in. by 3-in. cylindrical $\text{NaI}(\text{Tl})$ crystal and a 100-channel pulse-height analyzer were used. The magnetic temperature of the neodymium ethylsulfate crystal was measured with coils and an ac mutual-inductance bridge.

RESULTS

The angular distribution of the 740-keV γ ray was found to obey the equation $W(\theta) = 1 - (0.060 \pm 0.006)$

$\times P_2(\cos\theta)$ at 0.02°K (Fig. 1), where θ is the angle between the direction of propagation and the crystalline c axis. The ratio of cold to warm counting rate at $\theta = 0^\circ$ as a function of reciprocal temperature is shown in Fig. 2 and Table I. The absolute temperature was calculated from the magnetic temperature by using the data of Meyer⁴ with an appropriate correction for the demagnetization factor of the crystal.

DISCUSSION

The interpretation of nuclear orientation experiments is greatly facilitated by paramagnetic resonance data on atomic energy levels and hyperfine structure. In the absence of such work for promethium, we have resorted to interpolation of crystal-field parameters from neighboring rare-earth elements.

The energy levels of the tripositive Pm^{143} in neodymium ethylsulfate may be calculated by using the Hamiltonian:

$$\mathcal{H} = g_{11}\beta H_z S_z + A S_z I_z + \Delta_x S_x + \Delta_y S_y + P[I^2 - \frac{1}{3}I(I+1)] + c S_z(S_{1z} + S_{2z}).$$

The last term represents dipole-dipole interactions with the two nearest-neighbor Nd^{3+} ions,⁵ and the other

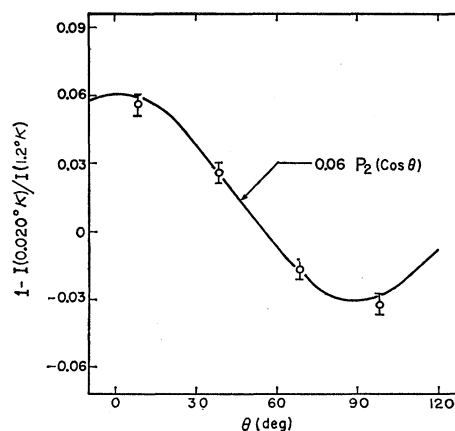


FIG. 1. Dependence of $1 - [I(0.020^\circ\text{K})/I(1.2^\circ\text{K})]$ for the 740-keV γ ray of Pm^{143} on the angle θ between the detector and the crystalline axis. The curve is $0.06P_2(\cos\theta)$.

[†] This work was done under the auspices of the U. S. Atomic Energy Commission.

¹ Shimon Ofer, *Phys. Rev.* **113**, 895 (1959).

² J. F. Schooley, D. A. Shirley, and J. O. Rasmussen, Lawrence Radiation Laboratory Report UCRL-9232, 1960 (unpublished).

³ S. G. Thompson, B. G. Harvey, G. R. Choppin, and G. T. Seaborg, *J. Am. Chem. Soc.* **76**, 6229 (1954).

⁴ H. Meyer, *Phil. Mag.* **2**, 521 (1957).

⁵ C. E. Johnson, J. F. Schooley, and D. A. Shirley, Lawrence Radiation Laboratory Report UCRL-9267, 1960 (unpublished); *Phys. Rev.* **120**, 2108 (1960).

TABLE I. Temperature dependence of the anisotropy.

$1/T^*$	$1/T$	$1 - \frac{I(0 \text{ deg}, T)}{I(0 \text{ deg}, 1^\circ\text{K})}$
11.2	11.4	0.008
11.4	11.9	0.004
11.7	12.3	0.003
17.3	19.4	0.017
21.9	24.7	0.019
23.1	27.0	0.028
25.6	29.6	0.026
26.7	31.8	0.042
29.5	34.4	0.031
30.5	35.6	0.040
31.5	38.6	0.049
33.5	42.6	0.051
33.1	44.8	0.055
33.8	47.6	0.061
34.4	50.0	0.068
34.6	51.0	0.072
35.7	54.4	0.071

terms have their usual significance. The term in P can be shown to be negligible in this case for purposes of nuclear alignment.⁶ A value of 0.0039 cm^{-1} was used for c , and a value of 0.014 cm^{-1} was used for Δ , where $\Delta^2 = \Delta_x^2 + \Delta_y^2$.

Tripositive promethium is a non-Kramers ion with the configuration $4f^4$. By Hund's rule, the ground term of the free ion is 5I_4 , and calculations indicate that in the ethylsulfate lattice the lowest level is a doublet composed of admixtures of the states $|J_z = \pm 4\rangle$ and $|J_z = \pm 2\rangle$.⁶ The magnetic hyperfine-structure constant, A , was calculated to be $(0.019 \pm 0.002)(\mu/I) \text{ cm}^{-1}$ by use of crystal-field theory.⁶

Experimentally, Ofer found the 740-keV γ ray to be predominantly $M1$,¹ but the experimental uncertainty precludes an accurate determination of how much $E2$ admixture may be present. The K -conversion coefficient reported is $(6.5 \pm 1) \times 10^{-3}$. The theoretical value inter-

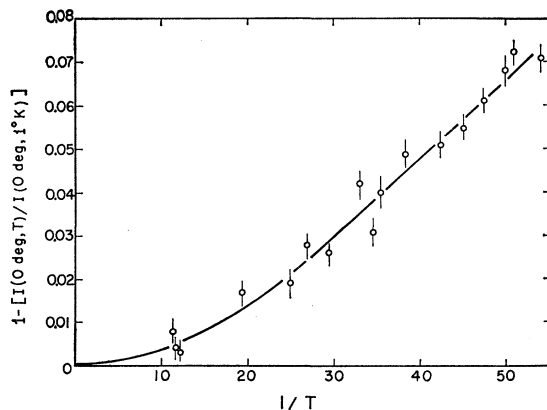


FIG. 2. Observed variation with $1/T$ of $1 - [I(0 \text{ deg}, T) / I(0 \text{ deg}, 1^\circ\text{K})]$ for the 740-keV γ ray of Pm^{143} .

⁶ J. F. Schooley, Ph.D. thesis, University of California, Berkeley, California, Lawrence Radiation Laboratory Report UCRL-9296, 1960 (unpublished).

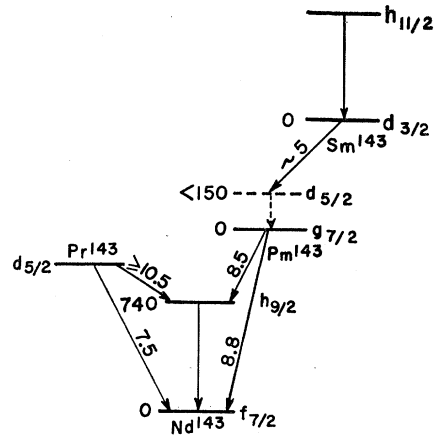


FIG. 3. Proposed level for several nuclei with $A = 143$. Numbers on arrows denote $\log ft$ values. Only indirect evidence is available for the excited state of Pm^{143} (see text and reference 11).

polated from the tables of Sliv and Band⁷ is 5.5×10^{-3} for an $M1$ transition and 3.4×10^{-3} from an $E2$ transition. Thus the spin and parity of the excited state of Nd^{143} may be $\frac{5}{2}^-$, $\frac{7}{2}^-$, or $\frac{9}{2}^-$.

The spins of Pm^{143} and of the excited state may be inferred from the following evidence: James and Bingham have found $M4$ isomerism in 81-neutron Sm^{143} ,⁸ which strongly suggests that the ground state of Sm^{143} is $d_{3/2}$ like its isotones, Nd^{141} , Ce^{139} , Ba^{137} , Xe^{135} , and Te^{133} . The $\log ft \sim 5$ for positron decay of Sm^{143} to Pm^{143} appears to indicate allowed decay and precludes a spin change of greater than one. Ofer gives an estimate of $\log ft = 8.8$ for the decay of Pm^{143} to the $\frac{7}{2}^-$ ground state of Nd^{143} , and $\log ft = 8.5$ for decay to the excited state, indicating first-forbidden decay.

No nuclear γ rays have been observed in the decay of Pr^{143} , and Starfelt and Cederlund set an upper limit of $10^{-30}\%$ on γ rays in the inner bremsstrahlung spectrum ($E_\gamma < 600 \text{ keV}$).⁹ A similar limit may be set on the 740-keV γ ray in this decay,¹⁰ and a lower limit of $\log ft \geq 10.5$ may thus be obtained for beta branching to the 740-keV state of Nd^{143} . This beta decay thus probably involves a spin change of at least 2. The most likely ground-state assignment of the odd proton in Pr^{143} is $\frac{5}{2}^+$. Then only spin and parity $\frac{5}{2}^-$ seem admissible for the excited state of Nd^{143} , involving the odd neutron in an $h_{9/2}$ orbit.

Way *et al.* have suggested that there is a low-lying $\frac{5}{2}^+$ level in Pm^{143} .¹¹ This would account very well for

⁷ L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 57ICC KI, issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)].

⁸ R. A. James and C. D. Bingham, Phys. Rev. **117**, 810 (1960).

⁹ N. Starfelt and J. Cederlund, Phys. Rev. **105**, 241 (1957).

¹⁰ R. W. Grant, Lawrence Radiation Laboratory, Berkeley, California (private communication).

¹¹ Nuclear Data Sheets, NRC 58-11-74 (1958) National Academy of Sciences, National Research Council (U. S. Government Printing Office, Washington, D. C.).

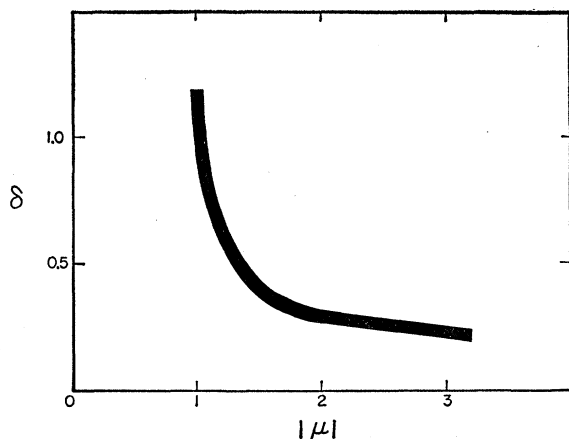


FIG. 4. Functional relationship between the magnetic moment of Pm^{143} and the $E2/M1$ mixing ratio of the 740-kev γ ray of Nd^{148} , as determined by this experiment. Width of line includes experimental error.

the fast beta decay from Sm^{148} , with subsequent γ -ray de-excitation to a $\frac{7}{2}^+$ ground state. These two states are close in energy in other odd promethium isotopes.¹¹ The only level scheme that is compatible with all the data is shown in Fig. 3, and the analysis of our results will be based on this scheme. We feel some degree of reservation in this interpretation, since the theoretical work of Kisslinger and Sorensen predicts that the ground state of Pm^{143} is $\frac{5}{2}^+$, with the $\frac{7}{2}^+$ level an excited state.¹²

On shell-model grounds it seems unlikely that the 740-kev state in 83-neutron Nd^{148} would be other than $\frac{9}{2}^-$, corresponding to the $h_{9/2}$ orbital. We cannot be quite so confident of our beta-decay ft -value arguments

that the ground state of Pm^{143} is $\frac{7}{2}^+$ and not $\frac{5}{2}^+$. The $\log ft$ of 8.5 for decay of Pm^{143} to the excited state is high enough that $\Delta I=2$, yes character is not precluded.

The analysis to follow is based on the assumption that the Pm^{143} spin is $\frac{7}{2}^+$. If the spin were to be measured as $\frac{5}{2}^+$, the necessary reinterpretation of our data would still give a relationship between δ and μ similar to that of Fig. 4.

The anisotropy of the radiation as a function of temperature was fitted to the theoretical function $W(\theta)=1+B_2(T)U_2F_2P_2(\cos\theta)$, where F_2 is the usual function of gamma multiplicities and initial and final spins for the gamma transition. The term U_2 depends on the unobserved preceding beta radiation and is a measure of the degree of realignment caused by the beta radiation. This notation is explained in the review article by Blin-Stoyle and Grace.¹³

At this point the interpretation of this work becomes somewhat tentative, because the anisotropy of the 740-kev γ ray depends on the mixing ratio, $\delta(E2/M1)$, and on the magnetic moment of the ground state of Pm^{143} . In Fig. 4 is shown the functional relationship between δ and $|\mu|$ as derived from the anisotropy data. Clearly, lower limits of $\delta > 0.23$ and $|\mu| > 1.0$ may be set from this work alone. According to the conversion-coefficient data, this transition is essentially pure $M1$, with the limits of error just including this multipolarity. The results presented here necessitate an $E2$ admixture of at least 5%. Indeed a pure $M1$ transition would require the anisotropy to have a sign opposite to that observed. Assuming that the magnetic moment of Pm^{143} lies between the Schmidt and Dirac limits of 1.72 and 3.11, respectively, we find $0.23 < \delta(E2/M1) < 0.35$. Thus the transition is $(8 \pm 3)\%$ quadrupole.

¹² L. F. Kisslinger and R. Sorensen; Kgl. Danske Videnskab Selskab, Mat.-fys. Skrifter (to be published).

¹³ R. J. Blin-Stoyle and M. A. Grace, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 555.