

# Gamma-Ray Anisotropies from Oriented $\text{Pm}^{144}\dagger$

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Radioactive  $\text{Pm}^{144}$  was oriented at low temperatures in a single crystal of neodymium ethylsulfate. The gamma rays at 475, 615, and 695 kev were found to be anisotropic. The results confirm the decay scheme previously proposed as well as crystal field calculations for  $\text{Pm}^{+3}$  in this lattice. It is not possible to decide between spin 5 and 6 for  $\text{Pm}^{144}$ . Values were obtained for  $|A|/k$  and  $|\mu|$  of  $0.0091^\circ\text{K}$  and  $1.68 \pm 0.14$  nm for  $I=5$ , or  $0.0079^\circ\text{K}$  and  $1.75 \pm 0.14$  nm for  $I=6$ . The lowest doublet of  $\text{Pm}^{+3}$  was found to be split, presumably due to Jahn-Teller distortion.

## INTRODUCTION

THE known isotopes of promethium cover the range from a closed neutron shell at  $N=82$  into the region where nuclei exhibit collective rotational behavior. Because there are no stable isotopes, relatively little is known about the atomic spectroscopy, and hence the hyperfine structure, of this element. Tripositive  $\text{Pm}^{+3}$  has the configuration  $4f^4$  and, according to Hund's rules, the ground term should be  $^5I_4$ . Using the theory of Elliott and Stevens<sup>1</sup> with crystal field parameters interpolated from other rare earth ethylsulfates, it can be predicted that the ground state of  $\text{Pm}^{+3}$  in the ethylsulfate lattice is a doublet composed of an admixture of  $|J_z=\pm 4\rangle$  and  $|J_z=\mp 2\rangle$ . If the hyperfine structure is large enough, the sensitive techniques of low-temperature nuclear orientation can be employed to study the nuclear properties of promethium. Ofer,<sup>2</sup> and Toth and Nielson<sup>3</sup> have studied the 300-day electron capture isotope,  $\text{Pm}^{144}$ , and the nuclear level scheme of  $\text{Nd}^{144}$  has been established. In the present work promethium-144 was aligned and polarized in a crystal of neodymium ethylsulfate.

## EXPERIMENTAL

Promethium-144 was made by bombarding a thick target of  $\text{Pr}^{141}$  with 20-Mev helium ions in the Berkeley 60-inch cyclotron. The promethium was separated from the praseodymium by elution of the tripositive ions with  $0.4M$  alpha hydroxyisobutyric acid adjusted to  $\text{pH}\sim 4.0$  from a Dowex 50 cation exchange column.<sup>4</sup> About  $10\mu\text{C}$  of  $\text{Pm}^{144}$  activity were grown into a 5-g single crystal of  $\text{Nd}(\text{C}_2\text{H}_5\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$ . The crystal was mounted on a 2-mm glass rod to which a thermal guard of compressed  $\text{MnSO}_4 \cdot (\text{NH}_4)_2\text{SO}_4 \cdot 24\text{H}_2\text{O}$  was attached, and the whole assembly was placed in a demagnetization cryostat and cooled to  $1.1^\circ\text{K}$  by contact with liquid helium. The magnetic field used in cooling the crystal by adiabatic demagnetization was applied parallel to

the crystalline  $c$  axis. In order to obtain the temperature dependence of the anisotropies, the crystal was demagnetized from various magnetic field strengths. Thus, the counting could be done immediately following demagnetization, minimizing errors caused by inhomogeneous warming of the crystal. During each counting period the absolute temperature changed by no more than 1%.

The temperature of the crystal was measured through its paramagnetic susceptibility by means of an ac mutual inductance bridge. This "magnetic temperature" was corrected to absolute temperature using the data of Meyer<sup>5</sup> and an appropriate demagnetization correction.

The gamma radiation was detected by  $3\times 3$ -in. cylindrical  $\text{Na}(\text{Tl})$  crystals with a 100-channel differential pulse-height analyzer. The sequence followed during most runs was (1) demagnetization, (2) immediate counting for a short period of time (generally five minutes), simultaneously determining the magnetic temperature of the crystal, (3) warming the crystal to the temperature of the liquid helium reservoir by admitting helium gas to the cryostat, and (4) counting of the now isotropic gamma radiation for the same period as before. Although warming of the crystal by stray heat leaks was slight, a check on the possibility of inhomogeneous warming was made by determining the anisotropy in successive counting periods of one minute and of twenty-five minutes duration. The anisotropy was the same in each case. Additional important information was gained by making anisotropy measurements on the magnetically-cooled crystal while a small polarizing field was applied along the  $c$  axis, using a battery-powered iron-free magnet. Fields of 100, 200, and 400 gauss were used in these polarization measurements. Finally, the experiment was repeated using a second crystal, and consistent results were obtained.

## RESULTS

The gamma-ray spectrum of  $\text{Pm}^{144}$  consists of three gamma rays of similar intensities and of energies 475, 615, and 695 kev. Pulse-height analysis of the spectrum with a 100-channel analyzer (Fig. 1) permitted the

<sup>†</sup> Work performed under the auspices of the U. S. Atomic Energy Commission.

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<sup>1</sup> R. S. Elliott and K. W. H. Stevens, Proc. Roy. Soc. (London) **A218**, 553 (1953).

<sup>2</sup> S. Ofer, Phys. Rev. **113**, 895 (1959).

<sup>3</sup> K. S. Toth and O. B. Nielson, Phys. Rev. **115**, 1004 (1959).

<sup>4</sup> S. G. Thompson *et al.*, J. Am. Chem. Soc. **76**, 6229 (1954).

<sup>5</sup> Horst Meyer, Phil. Mag. **2**, 521 (1957).

anisotropies of the individual gamma rays to be determined simultaneously allowing direct comparison among them. It was found that the anisotropies  $\epsilon = [W(90^\circ) - W(0)]/W(90^\circ)$  of the 615- and 695-keV gamma rays were identical, while that of the 475-keV gamma ray was about 20% less. The temperature dependence of the anisotropy of the two higher energy gamma rays is shown in Fig. 2. The square root of the anisotropy is plotted against the reciprocal of the absolute temperature to show that the anisotropy is very nearly a linear function of  $(1/T)^2$ .

Figure 3 shows the anisotropy as a function of the angle between the crystalline  $c$  axis and the direction of propagation at the lowest temperature. The uncertainties indicated for the data points plotted in Figs. 1, 2, and 3 are just those due to the statistical error in each count. In each case any necessary corrections have been made for background, finite source and counter size, drift in the scintillation spectrometer energy calibration, and change in block time of the 100-channel pulse-height analyzer. The uncertainty in the corrections was much less than the statistical error, and the corrections were never more than 1% of the total counting rate.

### DISCUSSION

The relevant spin Hamiltonian<sup>6</sup> for Pm<sup>143</sup> is

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

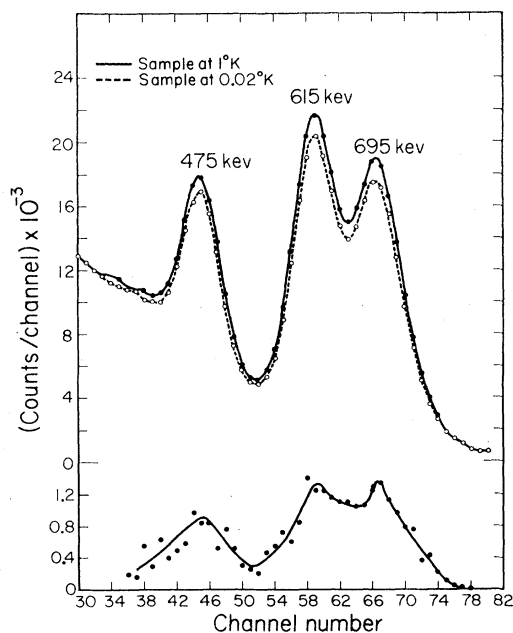


FIG. 1. Gamma-ray spectrum of Pm<sup>144</sup> taken during a typical run. The counter was placed along the crystalline  $c$  axis. The lower curve is a plot of the difference between "warm" and "cold" counting rates.

<sup>6</sup> J. M. Baker and B. Bleaney, Proc. Roy. Soc. (London) **A245**, 156 (1958).

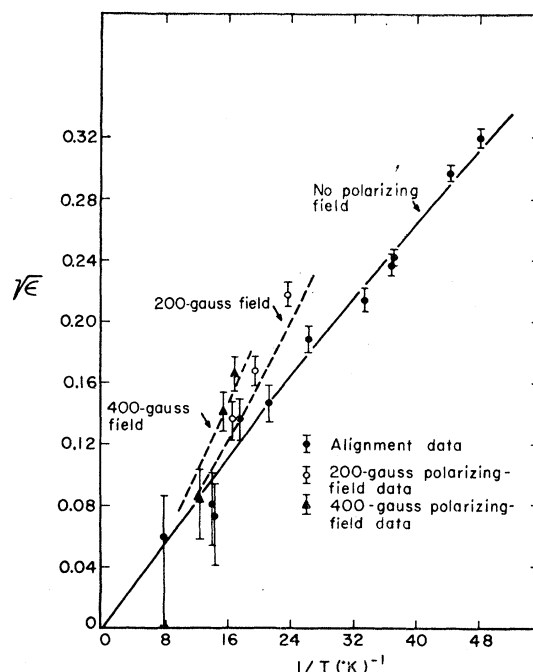


FIG. 2. Temperature dependence of anisotropy of the two higher energy gamma rays. Error flags represent rms statistical errors.

where the last term represents dipole-dipole interaction with the nearest neighbor neodymium ions and the other terms have their usual significance.

For a quadrupole moment of  $\sim 0.5$  barn, estimated from nuclear systematics,  $P$  is easily shown to be negligible on the theory of Elliott and Stevens, and may be neglected. Using this theory and interpolated<sup>7</sup> crystal field parameters with estimated limits of error  $A_2^0 \bar{r}^2 = (60 \pm 10) \text{ cm}^{-1}$ ,  $A_4^0 \bar{r}^4 = -(80 \pm 10) \text{ cm}^{-1}$ ,  $A_6^0 \bar{r}^6 = -(40 \pm 10) \text{ cm}^{-1}$ , and  $A_6^6 \bar{r}^6 = (600 \pm 20) \text{ cm}^{-1}$ , the ground state is calculated to be  $0.63 |J_z = \pm 4\rangle + 0.78 |J_z = \mp 2\rangle$ , with  $g_{11} = 0.47 \pm 0.04$ , and  $A/k = (0.027 \pm 0.002) |\mu|/I$ . Here  $\mu$  and  $I$  are the nuclear magnetic moment and spin, respectively, and the off-diagonal terms ( $\Delta_x S_x + \Delta_y S_y$ ) are due to the Jahn-Teller distortion effect,<sup>6</sup> and they tend to decrease the nuclear orientation. These terms split the lowest doublet and cause the hyperfine structure splitting to be unequally spaced. In the limit of  $A \ll \Delta$  the energies of the hyperfine structure levels are proportional to  $I_z^2$  rather than as  $I_z$  as in the case  $\Delta = 0$ . It is easily shown<sup>8</sup> that the nuclear alignment parameter  $B_2$  is unaffected by application of an external magnetic field along the crystalline  $c$  axis in the absence of off-diagonal terms in the Hamiltonian. In the presence of a Jahn-Teller interaction, however, the alignment can be substantially increased, with a consequent enhancement of the gamma-ray

<sup>7</sup> B. R. Judd, Proc. Roy. Soc. (London) **A251**, 407 (1959).

<sup>8</sup> J. M. Daniels, Proc. Phys. Soc. (London) **A66**, 673 (1953).

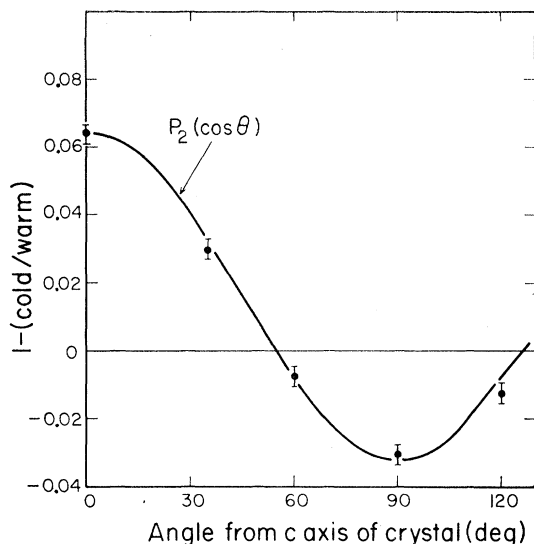


FIG. 3. Angular distribution of the two higher energy gamma rays at 0.02°K.

anisotropy, on application of a magnetic field along the  $c$  axis.

The term in  $c$  is introduced to account for spin-spin interactions between the promethium ion and the two nearest neighbor neodymium ions which lie at  $d = 7.07$  Å in either direction along the  $c$  axis. The six next nearest neighbors lie at angles giving rise to negligible spin-spin interaction.  $S_{1z}$  and  $S_{2z}$  are the effective electronic spin projections of these neodymium ions (taken as  $\pm \frac{1}{2}$  according to "spin Hamiltonian" convention), and  $c = -2g_{11}(\text{Pm})g_{11}(\text{Nd})\beta^2/d^3$ . The value of  $c$  can be calculated from existing data<sup>10,11</sup> and the value of  $g_{11}(\text{Pm})$  derived earlier in this paper. The value of  $c/k$  used in all our calculations is 0.0056°K.

Using the above Hamiltonian, and neglecting  $P$ , the energies of the  $2^3(2I+1)$  hyperfine states of the lowest doublet ( $S_z = \pm \frac{1}{2}$ ) are given by the formula

$$E(S_z S_{1z} S_{2z} I_z) = S_z \{ \Delta^2 + [g_{11}\beta H_z + A I_z + c(S_{1z} + S_{2z})]^2 \}^{\frac{1}{2}}, \quad (2)$$

where  $\Delta^2 = \Delta_x^2 + \Delta_y^2$ . The effects of the various terms governing the level spacing are illustrated in Fig. 4.

The gamma-ray intensity in this type of experiment depends on both the temperature of the crystal and the angle  $\theta$  between the direction of propagation and the crystalline  $c$  axis. Thus it is always necessary to separate these functional dependences experimentally before the maximum information can be obtained from such an experiment. The procedure employed in this laboratory is described below. First it is noted that in most nuclear alignment experiments the angular distribution of

gamma radiation is given by the expression

$$W(\theta) = 1 + B_2 U_2 F_2 P_2(\cos\theta) + B_4 U_4 F_4 P_4(\cos\theta).$$

The parameters  $B_\nu$ ,  $U_\nu$ , and  $F_\nu$  are described elsewhere,<sup>12</sup> and  $P_\nu$  is the Legendre polynomial of order  $\nu$ . The important point here is that the entire dependences on  $T$  and  $\theta$  are contained in  $B_\nu$  and  $P_\nu$ , respectively. The experimental procedure is the following: (1) Measure the complete angular distribution at the lowest temperature attainable, where the relative contribution of the term in  $B_4$  is expected to be greatest, (2) calculate from these data the approximate magnitudes of the  $B_2$  and  $B_4$  terms at all temperatures, and (3) measure the temperature dependence of  $W(\theta)$ . If, as is often the case, the  $B_4$  term is negligible even at the lowest attainable temperature, then the angular distribution

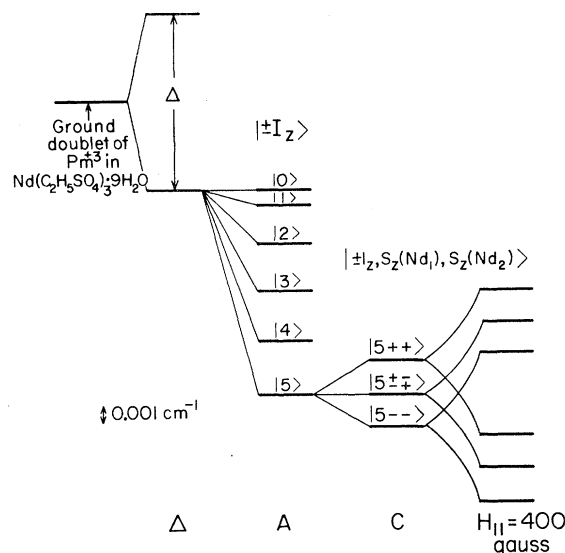


FIG. 4. Partial energy level diagram of  $\text{Pm}^{144}$  in neodymium ethylsulfate, showing the effect of each term in the spin Hamiltonian.

goes as  $P_2(\cos\theta)$  at all higher temperatures and need not be studied as extensively at these temperatures.

In the present experiment it was found that the term in  $P_2(\cos\theta)$  provided an adequate description of the angular distribution at the lowest temperature (Fig. 3).

It is clear from the form of the spin Hamiltonian that states of maximum  $|I_z|$  lie lowest. Thus the signs of the anisotropies are meaningful for establishing the signs of the  $F_2$  coefficients. Then from the signs and relative magnitudes of the anisotropies alone it is possible to confirm substantially the level scheme of  $\text{Nd}^{144}$  proposed by Ofer<sup>2</sup> (Fig. 5). The spins and parities of the ground state and first excited states are well established as  $0+$  and  $2+$  because the nucleus is even-even and the 695-keV transition is  $E2$ . The sign of the anisot-

<sup>9</sup> M. A. Grace, C. E. Johnson, R. G. Scurlock, and R. T. Taylor, *Phil. Mag.* **3**, 456 (1958).

<sup>10</sup> K. D. Bowers and J. Owen, *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 304.

<sup>11</sup> J. A. A. Ketelaar, *Physica* **4**, 619 (1937).

<sup>12</sup> R. J. Blin-Stoyle and M. A. Grace, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 555.

ropy of the 695-keV gamma ray also supports these assignments.

The sign and relative magnitude of the anisotropy of the 615-keV  $E2$  gamma ray proceeding from the 1.31-MeV level definitely rules out all spin and parity assignments other than  $3+$  or  $4+$  for this level. Spins and parities of  $1+$  or  $2+$  would require  $F_2 > 0$  contrary to experiment for both the 615- and the 695-keV transitions, and a spin and parity of  $0+$  would of course allow no anisotropy. Ofer obtained an unambiguous assignment of  $4+$  for this level.

Interpretation of the anisotropy of the 475-keV gamma ray is less straightforward because the unobserved preceding electron capture transition affects this anisotropy in a different way than those of the other gamma rays. The observed anisotropy is 20% less than that of the other two gamma rays, and the only plausible expla-

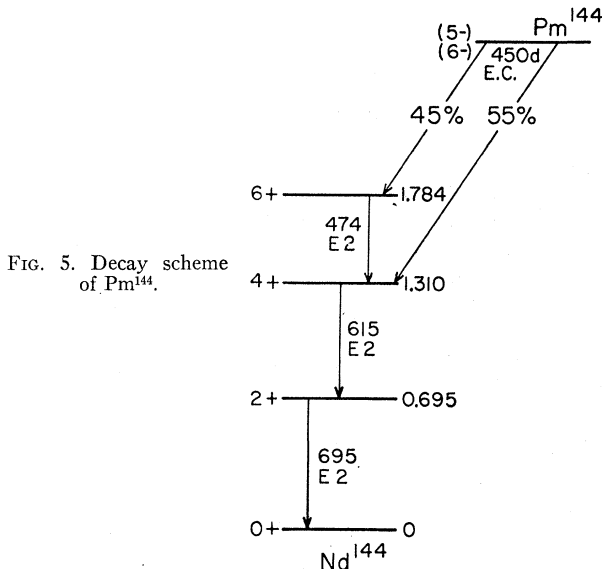


FIG. 5. Decay scheme of  $\text{Pm}^{144}$ .

nations seem to be (a) the 1780-keV level has a lifetime of the order of  $10^{-9}$  second and the changing extranuclear fields following electron capture decay attenuate the correlation by a factor  $G_2 \cong 0.7$ , or (b) a considerable fraction of the decay to the 1780-keV level is of the  $L=2$  type, thus reducing the alignment in this state substantially. If (b) is correct, the sequence  $5 \xrightarrow{L=2} 6$  for the electron capture decay to the 1780-keV state would fit the alignment data quantitatively. Regardless of this ambiguity, the anisotropy definitely excludes all possible spins and parities other than  $6+$  and  $2+$ , in support of Ofer, who obtained an unambiguous assignment of  $6+$ .

The existence of a Jahn-Teller distortion is established by an increased anisotropy in the presence of an external axial magnetic field. The accuracy of the temperature measurement in the field, which involves a 10% saturation correction, was checked by observing

TABLE I. Magnetic moments of  $\text{Pm}^{144}$ .

Proton state	$I$	$\mu_{\text{theo}}$
$d_{5/2}$	5	2.85
$d_{5/2}$	6	2.25
$g_{7/2}$	5	-0.138
$g_{7/2}$	6	-0.165

the anisotropies of gamma rays following the decay of  $\text{Nd}^{147}$  in the same lattice in several fields. The effect of magnetic fields on this anisotropy is readily calculated, and good agreement was obtained.

The anisotropy data were fitted using the Hamiltonian [Eq. (1)] and the decay scheme proposed by Ofer<sup>2</sup> (Fig. 5). The value of  $A$  depends on which spin is assumed for  $\text{Pm}^{144}$ . The results are

$$|A|/k = (0.0091 \pm 0.0003)^\circ\text{K}, \text{ for } I=5,$$

or

$$|A|/k = (0.0079 \pm 0.0003)^\circ\text{K}, \text{ for } I=6,$$

$$\Delta/k = (0.020 \pm 0.010)^\circ\text{K}.$$

The nuclear magnetic moment of  $\text{Pm}^{144}$  may be calculated from  $A$  using the theory of Elliott and Stevens and the value of  $\langle 1/r^3 \rangle = 36.8 \times 10^{24} \text{ cm}^{-3}$ .<sup>13</sup> The result is  $|\mu| = 1.68 \pm 0.14 \text{ nm}$  for  $I=5$  or  $|\mu| = 1.75 \pm 0.14 \text{ nm}$  for  $I=6$ .

The error in the calculated value of  $|\mu|$  is principally due to considerable uncertainty in the estimation of the crystal field parameters. Since  $A$  is proportional to  $g_{11}$ , a direct determination of  $g_{11}$  for any promethium isotope in the ethylsulfate lattice by paramagnetic resonance would be very desirable. The present experiment verifies the approximate value  $g_{11} \cong 0.47$  and the finite value of  $\Delta$  suggests that the transition probabilities for resonance should not be too small.

Shell-model considerations suggest that the odd neutron is in an  $f_{7/2}$  orbital and that the odd proton is in either a  $d_{5/2}$  or a  $g_{7/2}$  state. The magnetic moments expected on the simple vector-coupling model are listed in Table I. Thus the  $d_{5/2}$  proton state is favored, although a strong admixture of  $g_{7/2}$  is likely in view of the proximity of these two states in the heavier odd isotopes of promethium.<sup>14</sup>

#### ACKNOWLEDGMENTS

We wish to thank Dr. Brian R. Judd for several helpful discussions and for communication of the value of  $\langle 1/r^3 \rangle$  for  $\text{Pm}^{143}$ , G. A. Westenbarger for assistance with the measurements on  $\text{Nd}^{147}$ , and Mrs. Mab Tocher for assistance with preparation of the source.

<sup>13</sup> B. R. Judd and I. P. K. Lindgren, University of California Radiation Laboratory Report UCRL-9188, April 25, 1960 (unpublished).

<sup>14</sup> Nuclear Data Sheets, NRC 58-11-74 (National Research Council, 1958).