

for copper, the residual nucleus in our (α, p) experiment, we obtain $a=4.7$, which is very good agreement with the value of 4.4 obtained in this experiment.

$S(\alpha, E)$ is related to the level density of the residual nucleus. For (p, p') the residual nucleus is even-even, while for (α, p) the residual nucleus is odd-even. The odd-even nucleus has a level density estimated to be five times that of an even-even nucleus.² Thus $S(\alpha, E)$ for (p, p') shows much structure, while $S(\alpha, E)$ for (α, p) is very smooth. However, if we average the (p, p') data over 1-Mev energy intervals, most of the detailed structure is smoothed out and we may attempt to obtain level density information by applying statistical theory. Figure 7 is a plot of $S(\alpha, E)$ for (p, p') scattering

on Ni as a function of \sqrt{E} . When this curve is fitted with a straight line, the result $a=2.4$ is obtained. This small value of a is consistent with smaller level densities that are observed for even-even nuclei.

ACKNOWLEDGMENTS

We would like to thank Alex Lorenz, Darrell Malone, Pete Stoering, and Henry Catron for helping to take data, and Natalie Groteguth, Richard Neifert, and James Doyle for the reduction of the data. We would also like to thank LeRoy Erickson, Donald Rawles, and the cyclotron crew for the operation of the Livermore cyclotron.

Nuclear Moment of Ce^{137m} by Nuclear Alignment*

J. N. HAAG, C. E. JOHNSON,† D. A. SHIRLEY, AND D. H. TEMPLETON

Lawrence Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California

(Received September 2, 1960)

Nuclei of Ce^{137} and Ce^{137m} have been aligned at low temperatures in a single crystal of neodymium ethylsulfate nonahydrate by means of the magnetic hfs coupling with the electrons of the Ce^{+3} ions. The anisotropy of their gamma radiation has been observed. The magnetic moment of Ce^{137m} is $|\mu_N| = 0.96 \pm 0.09$ nm. The spin of Ce^{137m} is established as 11/2.

1. INTRODUCTION

CERIUM-137 is one of a large group of nuclides which has an $h_{11/2}$ isomeric state that decays by emission of $M4$ radiation to a $d_{3/2}$ ground state. Brosi and Ketelle¹ have studied this isomeric transition and the electron-capture decay of the ground state to La^{137} by gamma-ray, coincidence, and conversion-electron-spectroscopic techniques. Their results lead to the energy-level scheme shown in Fig. 1. A $g_{7/2}$ orbital was assigned to the ground state of La^{137} from its observed second-forbidden beta decay to Ba^{137} (spin 3/2), and a $d_{5/2}$ state to the first excited state from the $M1$ character of the 10-kev gamma ray. The shell model is in good agreement with these assignments, and further predicts that the 455-kev level is either in a $s_{1/2}$ or a $d_{3/2}$ state.

We have measured the magnetic moment of Ce^{137m} by aligning Ce^{137m} nuclei and measuring the anisotropic distribution of the gamma radiation. Further information was obtained about the decay scheme of Ce^{137} , which was also aligned.

2. EXPERIMENTAL PROCEDURE

Cerium-137m was prepared by a $(p, 3n)$ reaction of 21-Mev protons on natural lanthanum (99.911% La^{139})

* Work performed under the auspices of the U. S. Atomic Energy Commission.

† Present address: Atomic Energy Research Establishment, Harwell, England.

¹ A. R. Brosi and B. H. Ketelle, Phys. Rev. **100**, 169 (1955); **103**, 917 (1956).

in the ORNL 86-inch cyclotron. Cerium was separated from the target material by oxidation to the +4 state, followed by solvent extraction,² which yielded about 10^{12} atoms of Ce^{137m} . The cerium was then reduced to the +3 state and grown into a single crystal of neodymium ethylsulfate nonahydrate so that it replaced some of the Nd^{+3} ions. The crystal was mounted in

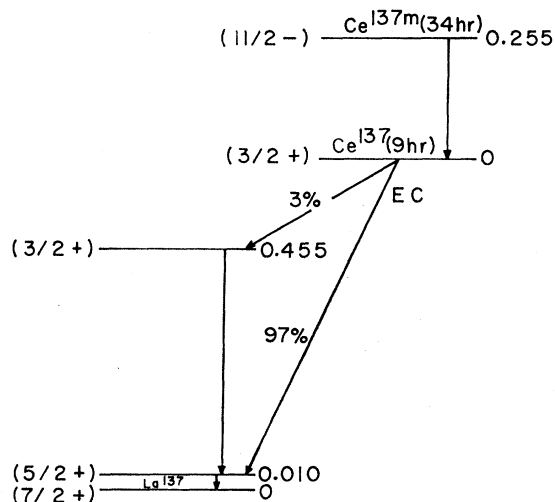


FIG. 1. Energy level scheme.

² L. E. Glendenin, Anal. Chem. **27**, 50 (1955).

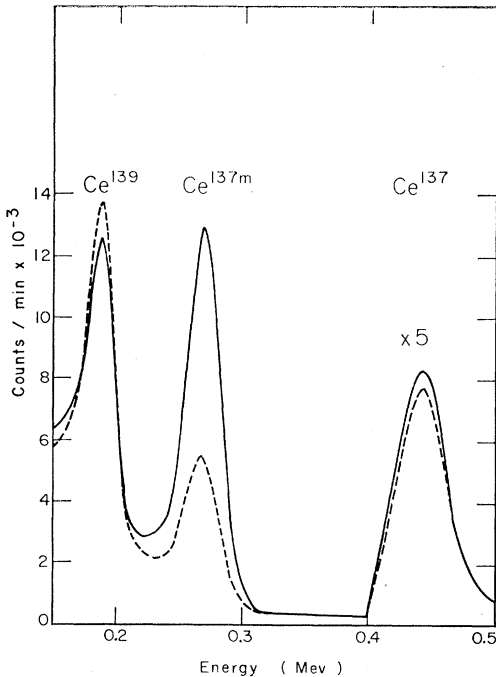


FIG. 2. Gamma-ray pulse-height spectrum at 1.1°K (solid line) and at 0.02°K (dashed line).

a demagnetization cryostat. Previous experiments^{3,4} on Ce^{139} and Ce^{141} had shown that nuclear alignment of the cerium isotopes was produced by cooling such a crystal to very low temperatures.

The crystal was cooled by adiabatic demagnetization from 1.1°K and fields of up to 18 000 gauss. The intensity of the gamma radiation was measured at several temperatures between 0.02- and 1.1°K for a series of angles θ defined by the direction of propagation of the gamma radiation with respect to the trigonal axis of the crystal. The gamma rays were counted using 3- \times -3-in. NaI(Tl) crystals and 100-channel pulse-height analyzers. The spectrum obtained is shown in Fig. 2. The peaks due to the 255-keV isomeric transition of Ce^{137m} , the 445-keV gamma ray of La^{137} , and the 165-keV gamma ray of La^{139} (from the decay of Ce^{139} , which was present as an impurity) are clearly resolved. The decay of these gamma rays was followed over 10 half-lives of the Ce^{137m} , and no other peaks were observed.

The magnetic temperature of the crystal after demagnetization was determined by measuring the mutual inductance of a pair of coils surrounding the crystal, using a 20-cycle/sec ac mutual-inductance bridge. The coils were calibrated in the liquid helium range of 4.2 to 1.1°K against a helium vapor-pressure thermometer. From the data of Meyer,⁵ the absolute temperatures

³ M. A. Grace, C. E. Johnson, R. G. Scurlock, and R. T. Taylor, *Phil. Mag.* (to be published).

⁴ C. F. M. Cacho, M. A. Grace, C. E. Johnson, A. C. Knipper, R. G. Scurlock, and R. T. Taylor, *Phil. Mag.* **46**, 1287 (1955).

⁵ H. Meyer, *Phil. Mag.* **2**, 521 (1957).

T reached after an adiabatic demagnetization from an initial temperature $T_i = 1.1^\circ\text{K}$, and various fields of H_i were known. A correlation between T and T^* was determined by extrapolating our value of the magnetic temperature T^* to the time of demagnetization.

The time taken for the temperature to rise from the lowest temperatures reached to that of the helium bath (1.1°K) was over an hour, but in order to avoid errors due to inhomogeneous heating of the crystal, the gamma-ray counting and the susceptibility measurements were continued for only one minute after the demagnetization. The crystal was then warmed to 1.1°K by the introduction of helium exchange gas. A further one-minute gamma-ray count at 1.1°K was then taken for normalization. The gamma radiation was isotropic within experimental error at this temperature. The gamma-ray counting rates were corrected for background and finite counter size effects,⁶ and the anisotropies $\epsilon = 1 - I(0^\circ)/I(90^\circ)$, were evaluated as a function of temperature.

3. RESULTS

The anisotropy of the 255-keV gamma ray of Ce^{137m} plotted versus $1/T$ is shown in Fig. 3.

The intensity of the 255-keV gamma ray at 0.018°K is shown as a function of θ in Fig. 4. This angular distribution, expressed in Legendre polynomials, was found to be

$$I(\theta) = 1 - (0.70 \pm 0.06)P_2(\cos\theta) + (0.05 \pm 0.01)P_4(\cos\theta). \quad (1)$$

At the same temperature, the intensity angular distribution of the 445-keV gamma ray was

$$I(\theta) = 1 - (0.10 \pm 0.02)P_2(\cos\theta),$$

and the 165-keV gamma ray of Ce^{139} showed an anisotropy of approximately -0.13 ± 0.03 . The latter result agrees with the data of Grace *et al.*³

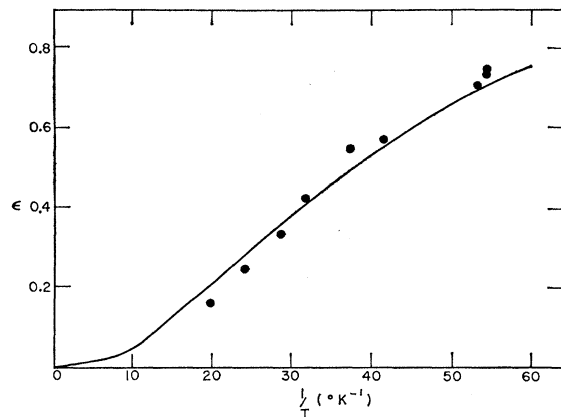


FIG. 3. Experimental values and corresponding theoretical fit for $|\mu_N| = 0.96 \text{ nm}$.

⁶ M. E. Rose, *Phys. Rev.* **91**, 610 (1953).

4. DISCUSSION

Determination of the Magnetic Moment of Ce^{137m}

The angular distribution of gamma radiation from aligned nuclei is given⁷ by

$$I(\theta) = 1 + B_2 U_2 F_2 P_2(\cos\theta) + B_4 U_4 F_4 P_4(\cos\theta) + \dots \quad (2)$$

The B_k 's are a measure of the degree of orientation of the parent nucleus. The U_k 's describe the amount of nuclear re-orientation that takes place during any unobserved beta or gamma transitions preceding the observed gamma ray. The F_k 's are constants determined by the multipolarity and the initial and final spins of the observed gamma transition.

The crystal field-theory of Ce^{+3} in the ethylsulfate lattice has been worked out in detail by Elliott and Stevens,⁸ and only a brief account will be given here.

The free ion Ce^{+3} has the configuration $4f^1$ and the ground term is ${}^2F_{5/2}$. In a trigonal crystalline field this term is split into doublets which may be characterized in the first approximation by $|\pm J_z\rangle$. In the ethylsulfate lattice, however, the lowest Kramers' doublet which is made mostly of the state $|\pm 5/2\rangle$, contains in addition, admixtures of other states from the ${}^2F_{5/2}$ ground term as well as from the next term ${}^2F_{7/2}$. It is, of course, essential that these admixtures be taken into account in calculating the nuclear magnetic-moment from hyperfine-structure constants.

The effective spin-Hamiltonian for the lowest Kramers' doublet of Ce^{137m} in the ethylsulfate is

$$\mathcal{H} = AS_z I_z + B(S_x I_x + S_y I_y) + P[I_z^2 - \frac{1}{3}I(I+1)].$$

The last term can be shown to have a negligible effect on nuclear alignment in this case, by using the theory

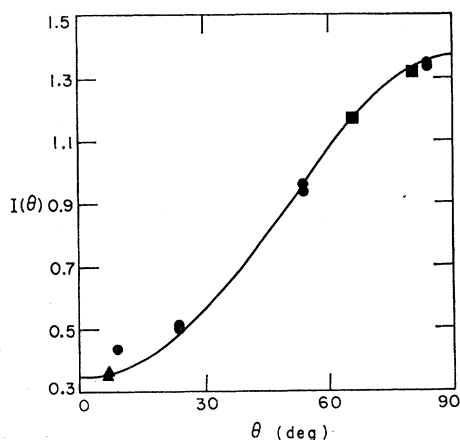


FIG. 4. Angular distribution of the 255-keV γ ray at 0.018°K. The line corresponds to $I(\theta) = 1 - 0.70 P_2(\cos\theta) + 0.05 P_4(\cos\theta)$. ● 1st quadrant, ■ 2nd quadrant, ▲ 4th quadrant.

⁷ T. P. Gray and G. R. Satchler, Proc. Phys. Soc. (London) **A68**, 349 (1955).

⁸ R. J. Elliott and K. W. H. Stevens, Proc. Roy. Soc. (London) **A215**, 437 (1952).

of Elliott and Stevens⁹ to calculate P and by using $Q=0.3$ barn for an $(h_{11/2})^9$ neutron configuration.¹⁰ The terms in B alter the energy levels of the hyperfine-structure multiplet slightly, and this has been taken into account. The energy levels of this multiplet then given approximately by twelve doublets $|\pm I_z\rangle$, separated by $A/2$. In going from 1.1 to 0.02°K the percentage of the Ce^{137m} nuclei occupying the lowest doublet changes from 8.3% to 37%.

For the 255-keV isomeric transition in Ce^{137m} there are no unobserved preceding transitions, and $U_2=U_4=1$. Thus, Eq. (2) becomes

$$I(\theta) = 1 - 0.8890 B_2 P_2(\cos\theta) + 0.4434 B_4 P_4(\cos\theta),$$

for the spin sequence $11/2 \rightarrow 3/2$ or

$$I(\theta) = 1 - 0.7444 B_2 P_2(\cos\theta) + 0.1693 B_4 P_4(\cos\theta)$$

for the spin sequence $9/2 \rightarrow 3/2$. The functions B_2 and B_4 depend on the single parameter $\beta = A/2kT$, and by varying A it is possible to fit the temperature dependence of the anisotropy for either spin sequence. Using the values of A which best fit the temperature dependence, we have calculated the angular distribution of the 255-keV γ ray at 0.018°K from each of the above expressions. The results are:

$$I(\theta) = 1 - 0.65 P_2(\cos\theta) + 0.04 P_4(\cos\theta), \quad \text{for } I=11/2, \quad (3)$$

$$I(\theta) = 1 - 0.60 P_2(\cos\theta) + 0.02 P_4(\cos\theta), \quad \text{for } I=9/2. \quad (4)$$

Comparison with Eq. (1) shows that (4) is in disagreement with it. Thus the spin possibility of 9/2 is eliminated for Ce^{137m} . We are not aware of any direct measurements of the spin of 11/2 for the $h_{11/2}-d_{3/2}$ isomers, therefore this measurement offers the most direct evidence available for this spin assignment.

The value for A obtained in (3) above is $|A| = 0.0129 \text{ cm}^{-1}$. By use of the theory of Elliott and Stevens for the ground doublet, together with the value of $\langle r^{-3} \rangle$ obtained by Judd and Lindgren,¹¹ we calculate

$$A = 0.074 \mu_N / I \text{ cm}^{-1}, \quad B = 0.002 \mu_N / I \text{ cm}^{-1}.$$

Comparison with our value for A yields

$$|\mu_N| = 0.96 \pm 0.09 \text{ nm}.$$

The limits of error were obtained from the scatter of the experimental points.

Because this is the first nucleus with $I=11/2$ for which the magnetic moment has been measured, we have included (Fig. 5) the Schmidt diagram for even-

⁹ R. J. Elliott and K. W. H. Stevens, Proc. Roy. Soc. (London) **A218**, 553 (1953).

¹⁰ Calculated using the method of H. Kopfermann, in *Nuclear Moments*, English edition (Academic Press Inc., New York, 1958), p. 398.

¹¹ B. R. Judd and I. P. K. Lindgren, Lawrence Radiation Laboratory Report UCRL-9188, April 25, 1960 (unpublished).

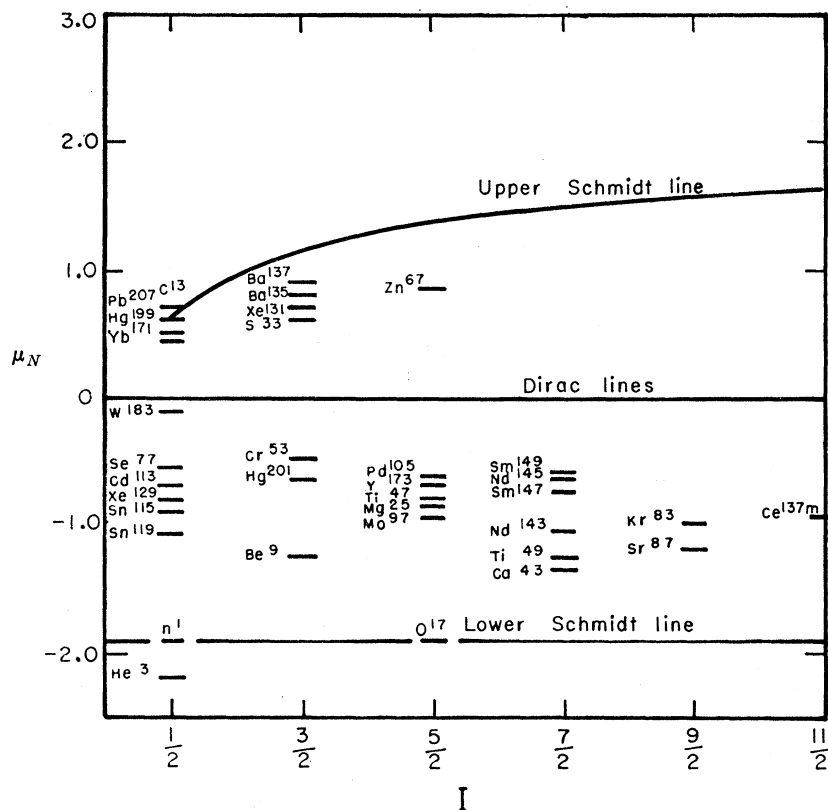


FIG. 5. Schmidt diagram for nuclei with an unpaired neutron.

odd nuclei. The moments for nuclei with $j < 11/2$ were taken from the Table of Isotopes.¹² We note that Ce^{137m} follows the trend in that the magnetic moment is about halfway between the Schmidt limit and the Dirac limit.

Nuclear Alignment of Ce^{137}

Since the half-life of Ce^{137} (9 hours) is long compared with the nuclear spin-lattice relaxation time, the anisotropy of its gamma radiation does not depend on the preceding isomeric transition of Ce^{137m} .

Our observation of an anisotropy in the 445-keV gamma ray immediately shows that the 455-keV state of La^{137} cannot have a spin of $1/2$, because this would show an isotropic gamma-ray distribution. Thus the

spins $3/2$ or $5/2$ are consistent with our data. This spin assignment and a determination of the magnetic moment of Ce^{137} could be made from a measurement of the plane polarization of the 445-keV gamma ray in addition to its anisotropy. From the present data it is concluded that if the 455-keV level has a spin of $3/2$, then the gamma ray must be a mixed $M1-E2$ radiation with $\delta(E2/M1) < 0$.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge several helpful discussions with Dr. Brian R. Judd concerning the theory of Elliott and Stevens. We thank Professor John O. Rasmussen for his suggestions and his continuing interest in this research. We are grateful to Dr. John L. Need and the crew of the 86-inch ORNL cyclotron for carrying out the bombardment for this experiment.

¹² D. Strominger, J. M. Hollander, and G. T. Seaborg, *Revs. Modern Phys.* **30**, 585 (1958).