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Authors
Haag, J.N.
Johnson, C.E.
Shirley, D.A.
et al.

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Ernest O. Lawrence

Radiation Laboratory

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NUCLEAR MOMENT OF $^{137m}$Ce BY NUCLEAR ALIGNMENT

J. N. Haag, C. E. Johnson, D. A. Shirley, and D. H. Templeton

July, 1960
NUCLEAR MOMENT OF $^{137}_{\text{Ce}}$ 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

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ABSTRACT

Nuclei of $^{137}_{\text{Ce}}$ and $^{137m}_{\text{Ce}}$ 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 $^{3+}_{\text{Ce}}$ ions. The anisotropy of their gamma radiation has been observed. The magnetic moment of $^{137m}_{\text{Ce}}$ is $|\mu_N| = 0.96 \pm 0.09$ nm. The spin of $^{137m}_{\text{Ce}}$ is established as $1\frac{1}{2}$.

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Nuclear Moment of Ce\(^{137m}\) by Nuclear Alignment

J. N. Haag, C. E. Johnson, D. A. Shirley, and David H. Templeton
Lawrence Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

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 M\(^4\) radiation to a \(d_{3/2}\) ground state. Brosi and Ketelle\(^1\) 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\(^{139}\)) 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.
Fig. 1. Energy Level Scheme.
ethyl sulfate nonahydrate so that it replaced some of the Nd$^{+3}$ ions. The crystal was mounted in 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 $1.1^0K$ and fields of up to 18000 gauss. The intensity of the gamma radiation was measured at several temperatures between 0.02- and $1.1^0K$ for a series of angles $\Theta$ 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- x 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 $1.1^0K$ against a helium vapor-pressure thermometer. From the data of Meyer,$^5$ the absolute temperatures T reached after an adiabatic demagnetization from an initial temperature $T_i = 1.1^0K$, 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.
Fig. 2. Gamma-ray pulse-height spectrum at 1.1°K (solid line) and at 0.02°K (dashed line).
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, and the anisotropies $\epsilon = I(0\,\text{deg})/I(90\,\text{deg})$, 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 $\theta$ in Fig. 4. This angular distribution, expressed in Legendre polynomials, was found to be

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

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 $- 0.13 \pm 0.03$. The latter result agrees with the data of Grace, et al. $^3$
Fig. 3. Experimental values and corresponding theoretical fit for $|\mu_N| = 0.96 \text{ nm}$.
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.
4. DISCUSSION

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

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

$$I(\theta) = 1 + B_2 U_2 P_2 (\cos \theta) + B_4 U_4 P_4 (\cos \theta) + \ldots$$  \hspace{1cm} (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, 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 $|\pm \frac{5}{2}\rangle$. In the ethylsulfate lattice, however, the lowest Kramers' doublet which is made mostly of the state $|\pm \frac{5}{2}\rangle$, 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 of Elliott and Stevens\textsuperscript{9} to calculate $P$ and by using $Q = 0.3$ barns for an $(h_{11/2})^9$ neutron configuration.\textsuperscript{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\textsuperscript{0}K the percentage of the Ce\textsuperscript{137m} nuclei occupying the lowest doublet changes from 8.3\% to 37\%.

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

$$I(\theta) = 1 - 0.8890 B_{12} P_2 (\cos \theta) + 0.4434 B_{24} P_4 (\cos \theta),$$

for the spin sequence $11/2 \overset{M_4}{\rightarrow} 3/2$ or

$$I(\theta) = 1 - 0.7444 B_{12} P_2 (\cos \theta) + 0.1693 B_{24} P_4 (\cos \theta)$$

for the spin sequence $9/2 \overset{M_4}{\rightarrow} 3/2$. The functions $B_2$ and $B_4$ depend on the single parameter $\beta = \frac{A}{2kT}$, 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 $\gamma$-ray at 0.018\textsuperscript{0}K from each of the above expressions. The results are:

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

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

Comparison with equation (1) shows that (4) is in disagreement with it. Thus the spin possibility of $9/2$ is eliminated for Ce\textsuperscript{137m}. We are not aware of any
direct measurements of the spin of 11/2 for the \( \frac{11}{2} - \frac{3}{2} \) 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, 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-odd nuclei. The moments for nuclei with \( j < 11/2 \) were taken from the Table of Isotopes. We note that Ce\(^{137m}\) follows the trend in that the magnetic moment is about halfway between the Schmidt limit and the Dirac limit.

The 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
Fig. 5. Schmidt diagram for nuclei with an unpaired neutron.
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 $\delta(E2/M1) < 0$.

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REFERENCES

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