Title
NUCLEAR MOMENT OF Ce137m BY NUCLEAR ALIGNMENT

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

Publication Date
1960-07-01

UCRL 9299

UNIVERSITY OF CALIFORNIA

Ernest O. Lawrence Radiation Laboratory

BERKELEY, CALIFORNIA

To be published in Physical Review

UCRL-9299

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

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

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

July, 1960

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

July, 1960

ABSTRACT

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.

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

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

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 M4 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

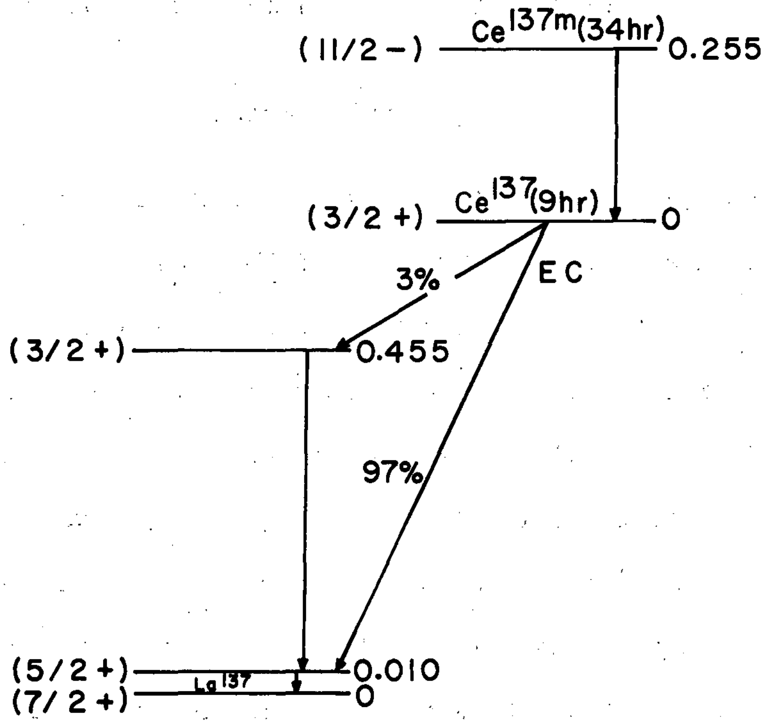
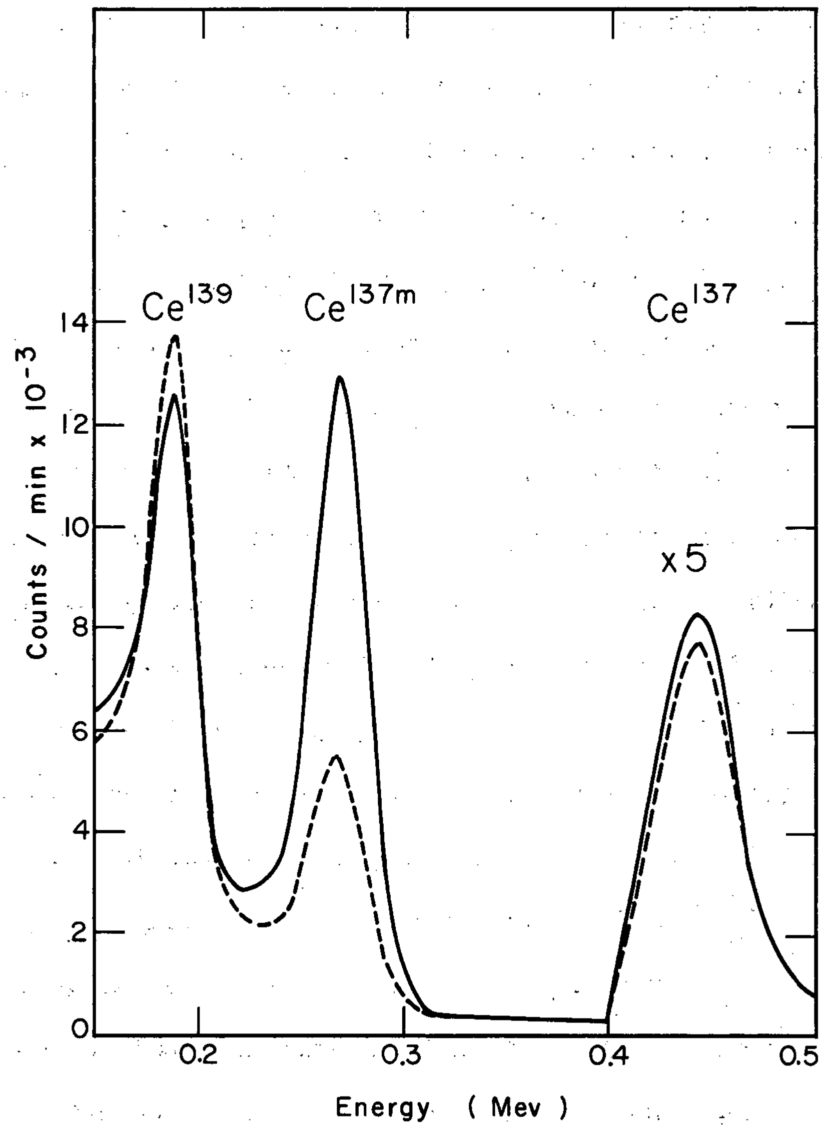


Fig. 1. Energy Level Scheme.

ethylsulfate 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°K and fields of up to 18000 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- 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°K 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°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.



MU-20988

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,[6] and the anisotropies $\epsilon = 1 - I(0\ \text{deg})/I(90\ \text{deg})$, 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 θ in Fig. 4. This angular distribution, expressed in Legendre polynomials, was found to be

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

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]

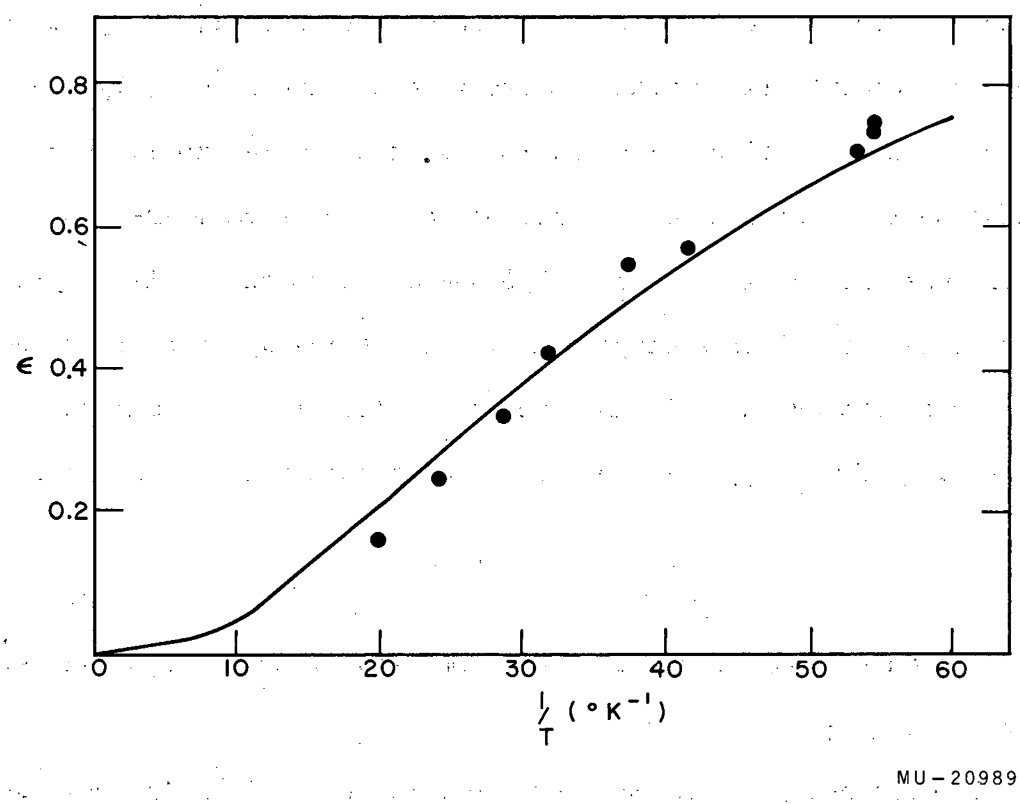
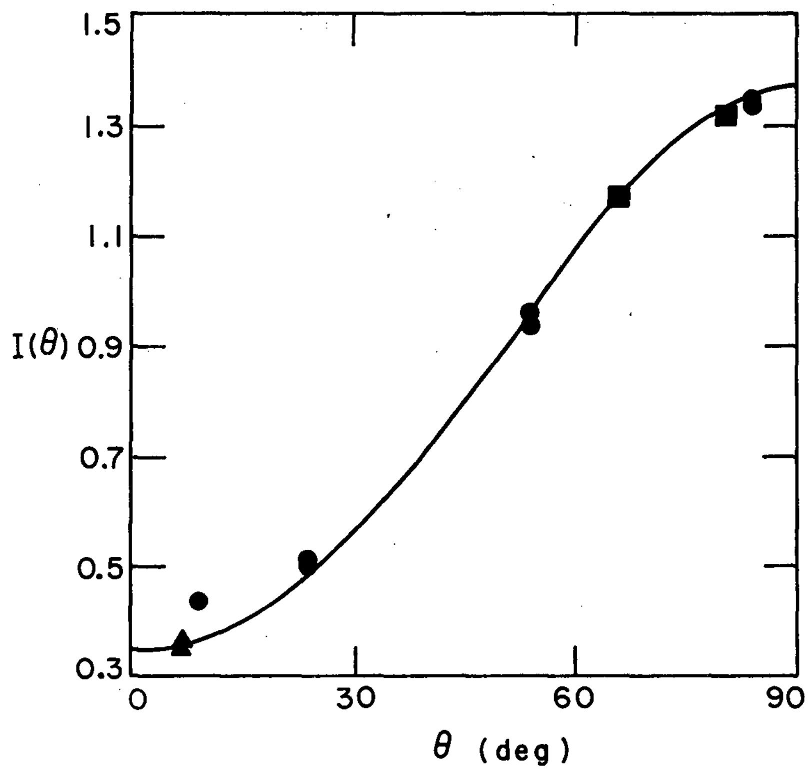


Fig. 3. Experimental values and corresponding theoretical fit for $|\mu_N| = 0.96$ nm.



MU-20990

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[7] by

$$I(\theta) = 1 + B_2U_2F_2P_2\ (\cos\theta) + B_4U_4F_4P_4\ (\cos\theta) + \ldots \quad (2)$$

The B_k's are a measure of the degree of orientation of the parent nucleus. The U_k's describe the amount oof 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

$$\mathcal{H} = AS_zI_z + B(S_xI_x + S_yI_y) + P[I_z^2 - \frac{1}{3}I\ (I + 1)].$$

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[9] to calculate P and by using Q = 0.3 barns 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, equation (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 $11/2 \xrightarrow{M4} 3/2$ or

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

for the spin sequence $9/2 \xrightarrow{M4} 3/2$. 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 γ-ray at 0.018°K from each of the above expressions. The results are:

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

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

Comparison with equation (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 $|A| = 0.0129\ cm^{-1}$. 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

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

Comparison with our value for A yields

$$|\mu_N| = 0.96 \pm 0.09\ nm.$$

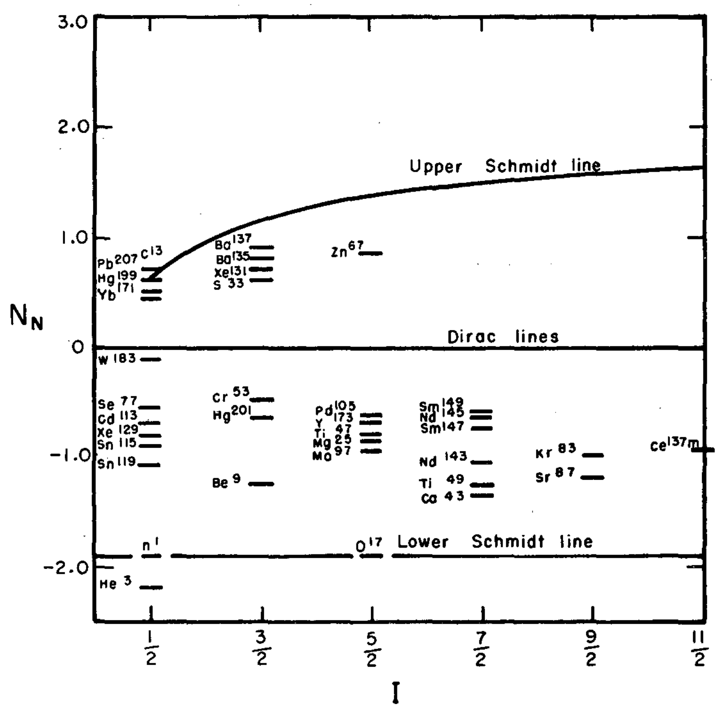
The limits of error were obtained from the scatter of the experimental points.

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.[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.

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



MU-20991

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 $\delta(E2/M1) < 0$.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge several helpful discussion 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.

REFERENCES

1. A. R. Brosi and B. H. Ketelle, Phys. Rev. 100, 169 (1955); Phys. Rev. 103, 917 (1956).
2. L. E. Glendenin, Anal. Chem. 27, 50 (1955).
3. M. A. Grace, C. E. Johnson, R. G. Scurlock, and R. T. Taylor, Phil. Mag. (to be published, 1960).
4. C.F.M. Cacho, M. A. Grace, C. E. Johnson, A. C. Knipper, R. G. Scurlock, and R. T. T ylor, Phil. Mag. 46, 1287 (1955).
5. H. Meyer, Phil. Mag. 2, 521 (1957).
6. M. E. Rose, Phys. Rev. 91, 610 (1953).
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).
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 ed., Academic Press Inc., New York, 1958), p. 398.
11. B. R. Judd and I. P. K. Lindgren, Theory of Zeeman Effects in the Ground Multiplets of Rare Earth Atoms, Lawrence Radiation Laboratory Report, UCRL-9188, April 25, 1960.
12. D. Strominger, J. M. Hollander, and G.T. Seaborg, Rev. Mod. Phys. 30, 585 (1958).