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Berkeley, California

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ATOMIC BEAM STUDIES OF NUCLEAR PROPERTIES
OF SOME RARE EARTH ISOTOPES

Burton Budick, Isaac Maleh, and Richard Marrus

March 25, 1964

INTRODUCTION

In this paper we report the results of a continuing investigation of the properties of nuclear ground states in the rare earth region. Interest in this region stems from the fact that a transition occurs from spherical to ellipsoidal nuclear cores. Therefore, the properties of rare earth nuclides can serve as tests for the various models of nuclear structure.

The method employed in this work is the atomic beam magnetic resonance method. For the application of this method to radioactive species the reader is referred to the many review articles.¹ The nuclear properties I (nuclear spin), μ_I (magnetic moment), and Q (quadrupole moment) are inferred quantities. The directly measured quantities are g_F , the splitting factor arising from the coupling of I and J ; the magnetic dipole hyperfine constant (A), and the electric quadrupole hyperfine constant (B). These are related by

$$g_F \approx g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)},$$

$$A = - \frac{1}{IJ} \mu_I \langle H_z \rangle_{m_J=J},$$

$$B = - e^2 \langle q_J \rangle Q,$$

where $\langle H_z \rangle_{m_J=J}$ is the magnetic field at the nucleus, $\langle q_J \rangle$ is the electric field gradient parameter, and J and g_J are the electronic angular momentum and the associated splitting factor. In order to infer nuclear properties, it is therefore necessary to know a considerable amount about the electronic state. For rare earth elements, this is a much investigated subject. One knows by now the values of J for almost all rare earth ground states and to within a few percent the values of g_J and $\langle H_z \rangle_{m_J=J}$.²

Unfortunately, there is still considerable uncertainty in the proper value of $\langle q_J \rangle$. No reliable information exists on the values of the quadrupole shielding and antishielding factors, although they are believed to be quite large.^{3,4} Therefore it is impossible to obtain reliable quadrupole moments from the hyperfine constants.

BEAM PRODUCTION, OBSERVATIONS, AND DATA ANALYSIS

Pr¹⁴³

The electronic structure of the low-lying states of Pr were found in earlier investigations.⁵ Schuurmanns and Meggers suggested that the ground state of Pr I was probably $f^3 s^2, {}^4I_{9/2}$.⁶ Lew confirmed this level assignment and established the g_J value as -0.731(2) in his work on the hfs and nuclear moments of Pr¹⁴¹.⁷ The hfs constants and nuclear moments of Pr¹⁴² (19 h) have also been measured.⁸

The only stable isotope of praseodymium has mass number 141, so that the production of Pr¹⁴³ proceeds through a double neutron capture on Pr¹⁴¹. Two successive high cross sections make this method feasible. Stable Pr was bombarded at the MTR in Arco, Idaho, for three weeks at a flux of 5×10^{14} neutrons/cm²-sec. Two weeks were allowed to elapse from the time of removal to permit the Pr¹⁴² to decay. A decay of a full beam foil showed that during the experiment Pr¹⁴³ was the dominant component in the beam. Two groups had reported half-life determinations of 13.76(5) and 13.59(4) days, respectively.⁹

Five flop-in transitions are observable. Three were seen at 2.818 G, two at 5.567 G, and one traced out at 10.865 G. They all correspond to a spin of 7/2. Some of the data are illustrated in Fig. 1.

Nd¹⁴⁹

The half-life and β spectrum of the neodymium isotope of mass number 149 had previously been measured.¹⁰ The half life was found to be 1.8 hours. Schuurmanns suggested the $f^4_s^2$ configuration and the Hund's rule ground term, 5I_4 .⁶ Smith and Spalding established the ground-level assignment and measured a g_J of -0.6032(1).¹¹ The even-Z character of this element implies that J is integral and the transitions are therefore of the multiple-quantum type.

A 4-hour neutron irradiation of natural Nd was used to produce Nd¹⁴⁹. The beam characteristics were complicated by the simultaneous production of Pm¹⁴⁹, Pm¹⁵¹, and Nd¹⁴⁷. At the inception of an experiment the counting rates of the 2-h component and of all the longer-lived components were approximately equal.

Transitions were observed in three J states at two settings of the magnetic field. All the data were consistent with a nuclear spin of 5/2. Sample results are shown in Fig. 2 and tabulated in Table I. It was important to establish that the resonances had a 1.8-h half life, since Nd¹⁴⁷ also has a spin of 5/2. Fortunately, the latter has a much longer half life and is easily distinguished.

Tb¹⁶¹

This radioactive isotope was produced by neutron bombardment on Gd metal at the MTR at Arco, Idaho. The reaction is $Gd^{160}(n, \gamma)Gd^{161} \xrightarrow[\beta]{3.7 \text{ min}} Tb^{161} (\tau = 6.8 \text{ d})$.¹² A 10-day bombardment enhances the ratio of the 6.8-day Tb¹⁶¹ to the longer-lived radioactive Gd isotopes, and performing the experiment 2 to 3 days after removal from the pile allowed time for the 18-h Gd¹⁵⁹ to decay. A satisfactory beam was

produced by electron-bombardment heating of a tantalum oven in which the Gd metal containing the radioactive isotope Tb^{161} was placed.

With the J and g_J values found at Heidelberg for Tb^{159} , ^{13, 14} a spin search was performed at two magnetic fields. Figure 3 shows the data at 5.566 gauss. Only the transitions in the $J = 11/2$ and $13/2$ were clearly seen. The result indicates $I=3/2$. A decay of one of the resonance buttons shows that the half life agrees with the reported value.

Ho^{161}

The purpose of investigating Ho^{161} was to determine both the nuclear and electronic angular momenta. The value of $g_J = -1.19516(10)$ was measured in conjunction with the spin of Ho^{166} . ¹⁵ This is close to the Russell-Saunders value -1.2 , for 11 f electrons coupling to a $^4I_{15/2}$ ground state. However, experiments on Ho^{166} could not establish the J value, and the possibility existed that the configuration was $4f^{10}5d$. In Russell-Saunders coupling this configuration gives rise to a $^6L_{21/2}$ ground state.

Even with J established as $15/2$, the $f^{10}d$ configuration cannot be ruled out because one need not assume R-S coupling between shells. The shells may be j - j coupled to one another, e. g., $(^5I_8 \ ^2D_{3/2})_{15/2}$. However, this coupling gives rise to a value of g_J that is far from the experimental value and is not plausible. In the light of these considerations the $J = 15/2$ measured in this experiment, together with the previously measured $g_J = -1.19516(10)$ lends strong support to the configuration assignment $4f^{11}6s^2$.

Natural dysprosium, in the form of pellets, was bombarded in the 60-inch Crocker cyclotron. Protons at 12 MeV and 30 μA produced the purest 2.5-h activity, at a level sufficiently intense for the experiment. The half life had been measured elsewhere. ¹⁶ In the early stages of the work the activity was observed in x-ray counters, the x rays accompanying the

electron-capture mode of decay. Subsequently, it proved both feasible and convenient to count the Auger electrons that are also emitted by the excited atom.

Figure 4 shows the results of a search at 5.567 gauss. Five of the eight flop-in transitions were observed at this field and three more were seen at 8.246 gauss. They all correspond to a spin of $7/2$ with $J = 15/2$.

Er¹⁶⁵

The electronic structure of erbium is discussed below in connection with Er¹⁷¹. To produce Er¹⁶⁵, 100%-abundant Ho¹⁶⁵ was bombarded in the 60-inch Crocker cyclotron. The products of the Ho¹⁶⁵(p, n)Er¹⁶⁵ reaction decayed with a pure 9.8 h half life. This is consistent with the 9.9(1) h half life that had been assigned previously.¹⁷ Holmium was in the form of discs and the erbium beam was produced by boiling directly out of the holmium.

Figure 5 shows data taken at 2.818 gauss. Another four flop-in transitions corresponding to $J = 6$, $I = 5/2$ were observed at 6.919 gauss.

Er¹⁷¹

By bombardment with 99.9% pure Er metal with thermal neutrons at the General Electric reactor at Vallecitos, California, for periods of 10 to 12 h, approximately 1 Ci of radioactive Er¹⁷¹ was produced. The reaction is Er¹⁷⁰(n, γ)Er¹⁷¹. The half life of this isotope ($\tau = 7.52$ h) was reported in 1958.¹⁸ The g_J value was determined initially by Smith¹¹ using stable ⁶⁸Er¹⁶⁶ while a more accurate value was obtained recently from the radioactive Er¹⁶⁹ [$g_J = -1.16381(5)$].¹⁹ The last value coincides very well with the prediction of Judd and Lindgren²⁰ using the configuration $(4f)^{12}(6s)^2$ and third-order spin-orbit corrections. The electronic ground state is then ³H₆, as predicted by Hund's rule. The nuclear spin of Er¹⁷¹ ($I = 5/2$) was

measured by Cabezas, using the atomic-beam magnetic resonance technique.¹⁴

A beam of the radioactive Er^{171} was obtained by electron-bombardment heating of a tantalum oven containing the metal. The possible radioactive contaminants in the beam included Er^{163} ($\tau = 75$ min), Er^{165} ($\tau = 10$ h), Er^{169} ($\tau = 9.4$ d), and Tm^{171} ($\tau = 1.9$ yr). The quantity of Er^{163} and Tm^{171} is negligible owing to their very short and very long half lives, respectively; the hfs on Er^{169} is known, and so it caused no trouble in the data analysis.¹⁹ Er^{165} , however, has the same spin ($I = 5/2$) as Er^{171} and therefore the same g_F factors. The half lives are similar, and although calculations predict the Er^{165} intensity to be $1/25$ or less than Er^{171} , resonances attributable to it were seen. Their amplitudes were $1/10$ or less of those of Er^{171} , and they decayed slightly differently. In this way they were separated from the Er^{171} data.

A total of 13 resonances were recorded. These were analyzed by the hyperfine computer program and values of A and B were obtained. The list of resonances with the corresponding magnetic fields (expressed in gauss as well as K^{39} resonance frequencies) is in Table II, and a hyperfine level diagram is shown in Fig. 6. A sample resonance is shown in Fig. 7. The results are:

$$|A| = 197.0(2.9) \text{ Mc,}$$

$$|B| = 3646(106) \text{ Mc,}$$

$$B/A > 0.$$

Since the χ^2 resulting from assuming a positive g_I in the hyperfine program is very similar to that resulting from a negative g_I , the average of the two convergences is quoted together with an uncertainty which includes both possibilities:

$$\chi^2 = \begin{pmatrix} 1.52 \\ 1.37 \end{pmatrix} \begin{pmatrix} \text{for } g_I > 0 \\ \text{for } g_I < 0 \end{pmatrix}.$$

Measurements on Er^{169} have determined directly the nuclear magnetic moment and the hyperfine constants. By use of these values and the Fermi-Segrè relation, the nuclear magnetic moment was determined to be $\mu_I = \pm 0.70(5) \text{ nm}$. If we assume that the electric field gradient $\langle q_J \rangle$ arises only from the coupling of the f electrons to the Hund's rule term, we infer for the quadrupole moment a value of $Q = 2.4(2)$ barns. However, it is well established that shielding and antishielding factors are quite large in the rare earth region and that this value may be in error by as much as a factor of two.^{3, 4} The value for the nuclear moment includes the corrections for diamagnetic shielding.

Figure 8 shows the hfs level ordering at zero magnetic field as a function of $\epsilon (= B/A)$.²¹ The experimental value of ϵ implies some inversion, as shown in Fig. 6.

Tm^{171}

A 3-week irradiation at the MTR at Arco, Idaho, on Er metal produced a sufficient quantity of radioactive Tm^{171} to permit the measurement of its hyperfine structure. The reaction is^{22, 23}

A few months were allowed to elapse before running to allow the 9-day Er^{169} to decay. Identification of the isotope was made by collecting the atoms on a platinum foil and taking a γ spectrum with a 200-channel Penco analyzer with a ThI crystal.

Using the same experimental technique as for Er^{171} , a total of nine resonances were found and served to give reasonable values of A , g_J , and g_I . Table III gives a list of the resonances and a χ^2 fit.

The results of these measurements are

$$A = \pm 372.1(5.9) \text{ Mc,}$$

$$g_J = -1.14116(10).$$

When published data²⁴ are used on Tm^{169} , the nuclear moment of Tm^{171} becomes

$$\mu_I(\text{corr}) = \pm 0.227(5) \text{ nm.}$$

The Fermi-Segrè relation was used to obtain the above values.

Figure 9 shows a typical resonance. Because of the long half life, the isotope was identified by examining a γ spectrum of a platinum button on which the beam was deposited. Figure 10 shows the γ spectrum of Tm^{171} . The 22.5-keV peak represents the iodine escape peak of the K x rays of Yb^{171} , i. e., the 67-keV level is highly converted, giving ≈ 53 -keV x rays (67 keV minus the binding energy of K electrons in Yb^{171}). These ≈ 53 -keV x rays sometimes knock out the K electrons of I in the ThI crystal, yielding electrons with energy ≈ 25 keV and x rays of iodine with 38 keV. The K x rays of iodine are not detected (escape), while the energy of the ≈ 25 -keV electrons is recorded. Thus, within the limits of the experiment, we see that the 22.5-keV peak corresponds to those electrons. The ≈ 84 -keV peak is strange, and may either represent a contaminant of the 99.9% pure Er or a real rotational level unreported by other researchers.^{22, 23}

DISCUSSION

Nuclear Spin Values

The last odd nucleon in Pr^{143} is the 59th proton. Two shell-model states, $d_{5/2}$ and $g_{7/2}$, are available. Evidence from other nuclei indicates that these levels are close together and that the actual ordering depends on the nucleus. For instance, the spin of Pr^{141} is $5/2$, indicating that the $g_{7/2}$ level lies lower. However, the spins of Pm^{147} and Pm^{149} are both $7/2$, implying that $d_{5/2}$ lies lower. For Pr^{143} , Martin et al.²⁵ argue in favor of a $d_{5/2}$ assignment, since the beta decay from the $h_{9/2}$ level of Ce^{143} to the ground state of Pr^{143} is not observed, and $\Delta l = 3$ would make this transition 1-forbidden. Kondaiah²⁶ predicts a spin of $7/2$ on the basis of x-ray multi-polarities, ft values, and the spin-orbit coupling model. The measured spin of $7/2$ indicates that the addition of two nucleons to Pr^{141} is sufficient to depress the $d_{5/2}$ shell below the $g_{7/2}$.

Neodymium-149 lies right in the transition region between single-particle and collective motions. A sharp change in nuclear structure is known to occur in going from 88 to 90 neutrons.²⁷ The measured spin of $5/2$ reflects the abruptness of this transition. No shell-model state with $j = 5/2$ is available in this region. On the other hand, an energy-level diagram for neutron numbers $82 < N < 126$ shows that a $[523] 5/2$ -state is available for small deformations.²⁸

The remaining isotopes are well within the collective region, with values of the deformation parameter $\delta \approx 0.3$. Their resultant nuclear angular momenta are due to the coupling of the last odd nucleon to the deformed nuclear core. In all cases their measured spins agree with those predicted.²⁷ The results are summarized in Table IV.

Magnetic Moments

Erbium-171 is far enough from closed shells that it can be nicely described by the strong-coupling approximations of the collective model. With a deformation ≈ 0.3 , which is about the same as other rare earths, the odd-particle state assignment is $[512] 5/2^-$. Using free nucleon g factors, we have calculated the moment for various deformations. The results are not very sensitive to the deformation, and yield

$$\mu_I = -0.95 \text{ nm}, \quad \delta \approx 0.3,$$

$$\mu_I = -0.91 \text{ nm}, \quad \delta \approx 0.2.$$

These are somewhat larger in magnitude than the measured value $\mu_I = \pm 0.70(5) \text{ nm}$. Although no determination of the sign was made in this experiment, we can infer it in the following way. Quadrupole moments in the rare earth region are usually positive. Using the positive sign for Q and the measured positive sign for B/A , we find that the magnetic moment is negative.

The magnetic moment of thulium can be determined in the same way. We must, however, know the decoupling parameter (a) as well as the deformation, since $I = 1/2$. The value of a has been found from the rotational spectra to be $a = -0.86$.²⁷ Using free nucleon g factors and assuming the state assignment to be $[411] 1/2^+$, we find

$$\mu = -0.300, \quad \delta \approx 0.3,$$

$$\mu = -0.132, \quad \delta \approx 0.2.$$

This is in good agreement with the measured value.

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Table I. Tabulation of all the observed resonances.

Z	Element	A	I	J	F	H (gauss)	Observed frequency	Calculated frequency
59	Pr	143	7/2	9/2	8	2.818	1.625	1.622
			7/2	9/2	7	2.818	1.676	1.674
			7/2	9/2	6	2.818	1.758	1.751
			7/2	9/2	8	5.567	3.210	3.203
			7/2	9/2	7	5.567	3.312	3.306
			7/2	9/2	8	10.865	6.253(50)	6.253
60	Nd	149	5/2	4	13/2	2.818	1.470	1.464
			5/2	4	11/2	2.818	1.565	1.564
			5/2	5	15/2	2.818	2.365	2.367
			5/2	5	13/2	2.818	2.550	2.550
			5/2	6	17/2	2.818	2.980	2.980
			5/2	4	13/2	5.567	2.891	2.891
			5/2	4	11/2	5.567	3.089	3.089
			5/2	4	9/2	5.567	3.417	3.417
			5/2	4	7/2	5.567	4.028	4.028
			5/2	5	15/2	5.567	4.675	4.675
			5/2	5	13/2	5.567	5.035	5.035
			5/2	6	17/2	5.567	5.885	5.591
65	Tb	161	3/2	11/2	7	10.865	18.110	18.112
			3/2	11/2	6	10.865	20.300	20.302
			3/2	13/2	8	5.566	9.265	9.264
			3/2	13/2	7	5.566	10.280	10.282

Table I (continued).

Z	Element	A	I	J	F	H (gauss)	Observed frequency	Calculated frequency
67	Ho	161	7/2	15/2	11	5.567	6.400(70)	6.352
			7/2	15/2	10	5.567	6.740(90)	6.691
			7/2	15/2	9	5.567	7.150(80)	7.143
			7/2	15/2	8	5.567	7.870(100)	7.760
			7/2	15/2	7	5.567	8.765(120)	8.800
			7/2	15/2	11	8.246	9.315(50)	9.411
			7/2	15/2	10	8.246	9.910	9.913
			7/2	15/2	9	8.246	10.580	10.582
68	Er	165	5/2	6	17/2	2.818	3.241	3.242
			5/2	6	15/2	2.818	3.493	3.494
			5/2	6	13/2	2.818	3.862	3.862
			5/2	6	11/2	2.818	4.431	4.430
			5/2	6	17/2	6.919	7.958	7.953
			5/2	6	15/2	6.919	8.577	8.572
			5/2	6	13/2	6.919	9.481	9.475
			5/2	6	11/2	6.919	10.879	10.870

Table II. Erbium-171 resonances and results of χ^2 test (g_I) positive and negative.

g_I	a (Mc)	Δa (Mc)	b (Mc)	Δb (Mc)	χ^2	
positive	196.300	2.163	3622.173	80.456	1.52	
negative	197.608	2.213	3669.895	82.169	1.37	
ν_K (Mc)	H(G)	ν_{exp} (Mc)	Residual (Mc)		Transition	Machine
			$g_I > 0$	$g_I < 0$		
20.000(40)	25.388(45)	58.460(28)	0.009	0.004	α	A
140.000(100)	117.684(58)	272.300(140)	0.137	0.124	α	B
170.000(130)	134.257(69)	310.700(140)	-0.048	-0.060	α	B
215.000(140)	157.118(68)	364.100(140)	0.016	0.007	α	B
249.900(140)	173.694(65)	402.900(140)	0.064	0.058	α	B
410.000(150)	242.716(61)	564.800(200)	-0.149	-0.135	α	B
80.000(100)	79.132(73)	197.500(160)	0.041	0.036	β	B
110.000(100)	99.549(64)	248.940(200)	0.048	0.047	β	B
140.000(100)	117.684(58)	294.900(200)	0.131	0.135	β	B
170.000(150)	134.257(80)	336.900(240)	0.040	0.050	β	B
140.000(100)	117.684(58)	330.850(200)	-0.022	-0.017	γ	B
170.025(150)	134.270(80)	380.000(300)	-0.119	-0.115	γ	B
210.075(150)	154.708(74)	442.400(250)	0.060	0.054	γ	B
<u>Transitions</u>						
	α	$F = 17/2$	$m_F = -3/2 \leftrightarrow -7/2$			
	β	$F = 15/2$	$m_F = -1/2 \leftrightarrow -5/2$			
	γ	$F = 13/2$	$m_F = 1/2 \leftrightarrow -3/2$			

Table III. Thulium-171 resonances and χ^2 fit.^a

a (Mc)	g_j	Δa	Δg_j	χ^2
372.0710	-1.141155	5.9403	0.000097	1.25

ν_K (Mc)	H (G)	ν_{exp} (Mc)	Residual	Transition
139.980(40)	117.672(23)	164.950(50)	-0.039	α
160.000(50)	128.875(27)	180.750(50)	-0.019	α
310.000(80)	200.654(35)	282.230(100)	-0.051	α
350.000(100)	217.808(42)	306.700(50)	0.049	α
80.030(50)	79.154(37)	142.360(50)	0.011	β
120.140(100)	105.890(61)	190.450(100)	0.003	β
140.000(40)	117.684(23)	211.650(50)	-0.011	β
209.950(50)	154.646(25)	278.125(50)	0.009	β
310.000(80)	200.654(35)	360.750(50)	0.020	β

$$\text{Residual} = \nu_{\text{exp}} - \nu_{\text{theo}}$$

Transitions α $F = 4; m_F = 0 \leftrightarrow -1$
 β $F = 3; m_F = 1 \leftrightarrow 0.$

^a Data taken on Machine B.

Table IV. Comparison of measured and theoretical spin values.

Z	Element	A	Measured spin	Nilsson state or shell- model state	
				Old proton	Odd neutron
59	Pr	143	7/2	48 7/2	
60	Nd	149	5/2		[523] $\frac{5}{2}^-$
65	Tb	161	3/2	[411] $\frac{3}{2}^+$	
67	Ho	161	7/2	[523] $\frac{7}{2}^-$	
68	Er	165	5/2		[523] $\frac{5}{2}^-$

FIGURE CAPTIONS

- Fig. 1. Two transitions in Pr^{143} .
- Fig. 2. Observed transitions in Nd^{149} .
- Fig. 3. Spin search for Tb^{161} at $\nu_K = 4.0$ Mc ($H = 5.566$ G).
- Fig. 4. Observations in Ho^{161} .
- Fig. 5. Four transitions observed in Er^{165} .
- Fig. 6. Hyperfine structure diagram (partial schematic) for Er^{171} .
- Fig. 7. Resonance due to an alpha transition at $\nu_K = 215.00$ Mc ($H \approx 157$ G).
- Fig. 8. Zero-field hfs level ordering for $(\frac{I}{J}) = 5/2$, $(\frac{J}{I}) = 6$, as a function of $\xi (= \frac{B}{A})$.
- Fig. 9. Alpha transition at $\nu_K = 350.000$ Mc ($H = 217.808$ G, $F = 4$, $m_F = 0 \leftrightarrow -1$).
- Fig. 10. Gamma spectra of Tm^{171} produced from $\text{Er}^{170} (n, \gamma) \text{Er}^{171} \xrightarrow[\beta^-]{7.5 \text{ h}} \text{Tm}^{171}$.

MU-26305

Fig. 1

MU-26438

Fig. 2

MU-27567

Fig. 3

Fig. 4

Fig. 5

Fig. 6

MU-28892

Fig. 7

MU-21398-A

Fig. 8

MU-28498

Fig. 9

MU-28998

Fig. 10

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