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Atomic-Beam Studies of Nuclear Properties of Some Rare-Earth Isotopes*

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A study has been made of the nuclear spins I and hyperfine structures of several radioactive isotopes in the rare-earth region. The nuclear-spin determinations are Pr^{143} ($I=7/2$), Nd^{149} ($I=5/2$), Tb^{161} ($I=3/2$), Ho^{161} ($I=7/2$), and Er^{169} ($I=5/2$). The measured hyperfine constants are: for Er^{171} , $|A|=197.0(2.9)$ Mc/sec, $|B|=3646(106)$ Mc/sec, $B/A > 0$; and for Tm^{171} , $|A|=372.1(5.9)$ Mc/sec; and the moments inferred from them are: for Er^{171} , $|\mu_I|=0.70(5)$ nm, and for Tm^{171} , $|\mu_I|=0.227(5)$ nm. A discussion of these values in terms of the various nuclear models is included.

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

Pr^{143}

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

² For a summary, see R. Marrus and W. A. Nierenberg, in *Proceedings of the International School of Physics, Enrico Fermi, Course 17* (Academic Press Inc., New York, 1962), pp. 118-156.

³ A. J. Freeman and R. E. Watson, *Phys. Rev.* **131**, 2566 (1963).

⁴ S. DeBenedetti, G. Lang, and R. Ingalls, *Phys. Rev. Letters* **6**, 60 (1961).

⁵ N. Rosen, G. R. Harrison, and J. R. MacNally, *Phys. Rev.* **60**, 722 (1941).

* This work was done under the auspices of the U. S. Atomic Energy Commission.

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¹ See, for instance, W. A. Nierenberg, *Ann. Rev. Nucl. Sci.* **7**, 349 (1957).

fields. Figure 3 shows the data at 5.566 G. 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.

Ho¹⁶¹

The purpose of investigating Ho¹⁶¹ was to determine both the nuclear and electronic angular momenta. The

TABLE I. Tabulation of all the observed resonances.

Element	Z	A	I	J	F	H (G)	Observed frequency	Calculated frequency
59 Pr	143		$\frac{7}{2}$	9/2	8	2.818	1.625	1.622
				9/2	7	2.818	1.676	1.674
				9/2	6	2.818	1.758	1.751
				9/2	8	5.567	3.210	3.203
				9/2	7	5.567	3.312	3.306
60 Nd	149		$\frac{7}{2}$	9/2	8	10.865	6.253(50)	6.253
				4	13/2	2.818	1.470	1.464
				4	11/2	2.818	1.565	1.564
				5	15/2	2.818	2.365	2.367
				5	13/2	2.818	2.550	2.550
				6	17/2	2.818	2.980	2.980
				4	13/2	5.567	2.891	2.891
				4	11/2	5.567	3.089	3.089
				4	9/2	5.567	3.417	3.417
				4	7/2	5.567	4.028	4.028
65 Tb	161		$\frac{7}{2}$	11/2	7	10.865	18.110	18.112
				11/2	6	10.865	20.300	20.302
				13/2	8	5.566	9.265	9.264
				13/2	7	5.566	10.280	10.282
				15/2	11	5.567	6.400(70)	6.352
				15/2	10	5.567	6.740(90)	6.691
				15/2	9	5.567	7.150(80)	7.143
				15/2	8	5.567	7.870(100)	7.760
67 Ho	161		$\frac{7}{2}$	15/2	7	5.567	8.765(120)	8.800
				15/2	11	8.246	9.315(50)	9.411
				15/2	10	8.246	9.910	9.913
				15/2	9	8.246	10.580	10.582
				6	17/2	2.818	3.241	3.242
				6	15/2	2.818	3.493	3.494
				6	13/2	2.818	3.862	3.862
				6	11/2	2.818	4.431	4.430
68 Er	165		$\frac{7}{2}$	6	17/2	6.919	7.958	7.953
				6	15/2	6.919	8.577	8.572
				6	13/2	6.919	9.481	9.475
				6	11/2	6.919	10.879	10.870

value of $g_J = -1.19516(10)$ was measured in conjunction with the spin of Ho¹⁶⁶.¹⁵ This is close to the Russell-Saunders (R-S) value -1.2 , for 11 f electrons coupling to a $^4I_{15/2}$ ground state. However, experiments on Ho¹⁶⁶ could not establish the J value, and the possibility

¹⁵ L. S. Goodman, W. J. Childs, R. Marrus, I. P. K. Lindgren, and A. Y. Cabezas, *Bull. Am. Phys. Soc.* **5**, 344 (1960); W. J. Childs and L. S. Goodman, *Phys. Rev.* **122**, 591 (1961).

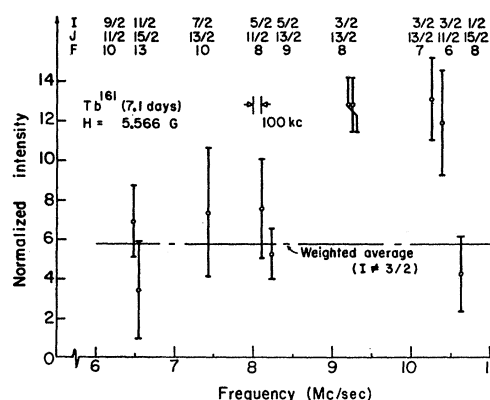


FIG. 3. Spin search for Tb¹⁶¹ at $\nu_K=4.0$ Mc/sec ($H=5.566$ G).

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-in. Crocker cyclotron. Protons at 12 MeV and 30 μ A 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 G. Five of the eight flop-in transitions were observed at this field and three more were seen at 8.246 G. They all correspond to a spin of $\frac{7}{2}$ 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-in. 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 disks and the erbium beam was produced by boiling directly out of the holmium.

¹⁶ T. H. Handley and E. L. Olson, *Phys. Rev.* **93**, 524 (1954).

¹⁷ F. D. S. Butement, *Proc. Phys. Soc. (London)* **A63**, 775 (1950).

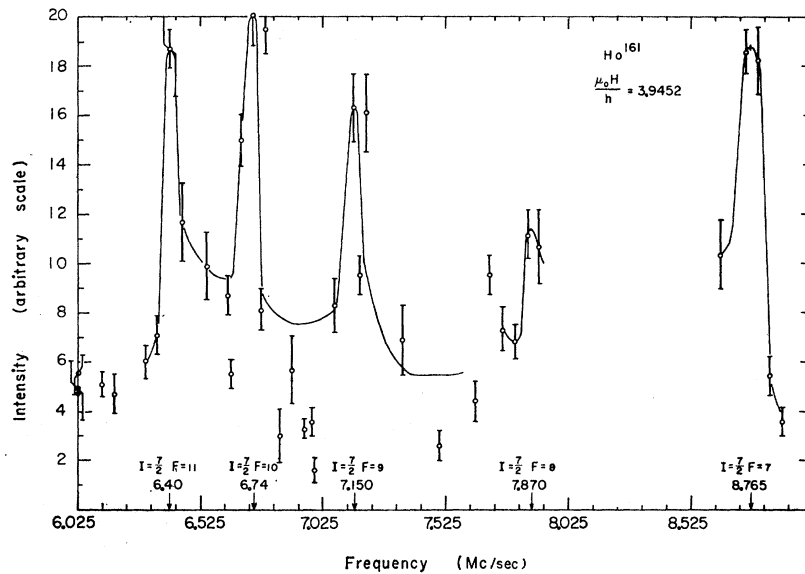
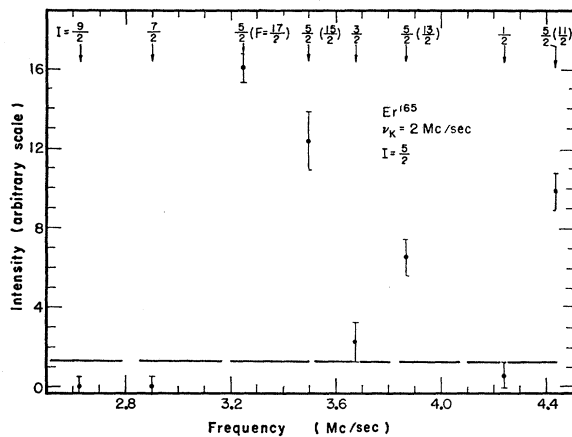
FIG. 4. Observations in Ho^{161} .

Figure 5 shows data taken at 2.818 G. Another four flop-in transitions corresponding to $J=6$, $I=\frac{5}{2}$ were observed at 6.919 G.

Er^{171}

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 Er^{171} was produced. The reaction is $\text{Er}^{170}(n,\gamma)\text{Er}^{171}$. 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 $^{68}\text{Er}^{166}$ while a more accurate value was obtained recently from the radioactive Er^{169} [$g_J=-1.16381(5)$].¹⁹ The last value coincides very well with the prediction of Judd and

FIG. 5. Four transitions observed in Er^{166} .

¹⁸ F. P. Cranston, Jr., M. E. Bunker, and J. W. Starner, Phys. Rev. **110**, 1427 (1958).

¹⁹ W. M. Doyle and R. Marrus, Phys. Rev. **131**, 1586 (1963).

Lindgren²⁰ using the configuration $(4f)^{12}(6s)^2$ and third-order spin-orbit corrections. The electronic ground state is then 3H_6 , as predicted by Hund's rule. The nuclear spin of Er^{171} ($I=\frac{5}{2}$) 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} ($\tau=9.4$ days), 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 ($I=\frac{5}{2}$) 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 $\frac{1}{10}$ or less 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:

$$\begin{aligned} |A| &= 197.0(2.9) \text{ Mc/sec,} \\ |B| &= 3646(106) \text{ Mc/sec,} \\ B/A &> 0. \end{aligned}$$

An attempt was made to determine the sign of the magnetic moment by inserting first a positive and then

²⁰ B. R. Judd and I. P. K. Lindgren, Phys. Rev. **122**, 1802 (1961).

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

g_I	a (Mc/sec)	Δa (Mc/sec)	b (Mc/sec)	Δb (Mc/sec)	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/sec)	H (G)	ν_{exp} (Mc/sec)	Residual (Mc/sec)		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	

Transitions α $F=17/2$ $m_F = -3/2 \leftrightarrow -7/2$
 β $F=15/2$ $m_F = -1/2 \leftrightarrow -5/2$
 γ $F=13/2$ $m_F = 1/2 \leftrightarrow -3/2$

a negative value of g_I in the computer program. The parameter χ^2 , which measures the accuracy of the least-squares fit, had values of 1.52 and 1.37 for $g_I > 0$ and $g_I < 0$, respectively. The proximity of these values makes a sign assignment impossible. Results for the hyperfine constants quoted above are averages of results from the

two least-squares fits. The stated uncertainty also includes both possibilities.

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 $\mu_I = \pm 0.70(5)$ 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 $Q = 2.4(2)$ b. 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 2.^{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.

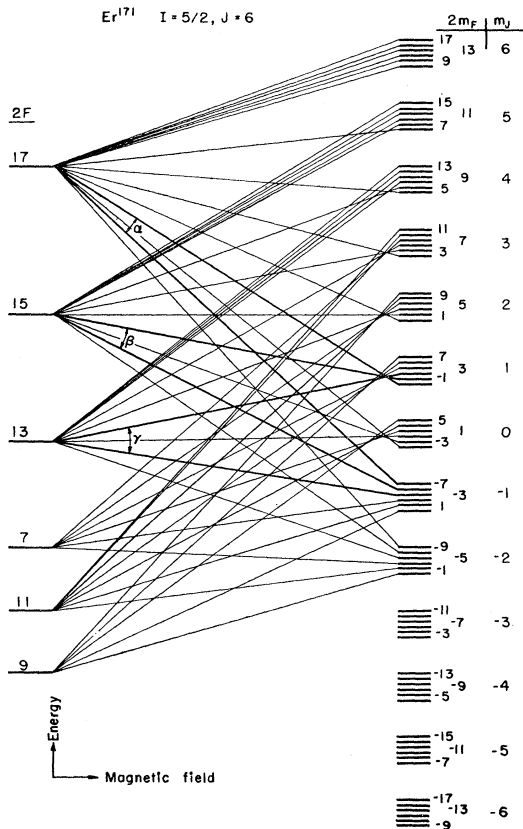


FIG. 6. Hyperfine structure diagram (partial schematic) for Er^{171} .

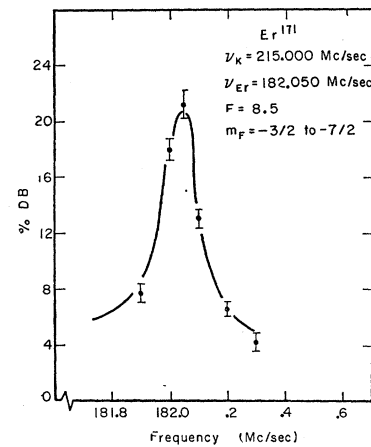


FIG. 7. Resonance due to an alpha transition at $\nu_K = 215.00$ Mc/sec ($H \approx 157$ G).

²¹ F. M. Baker, University of California Lawrence Radiation Laboratory Report UCRL-9364, August 1960 (unpublished).

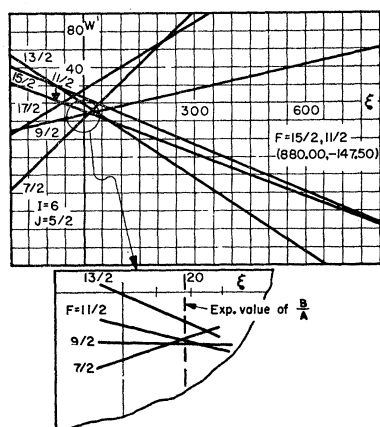
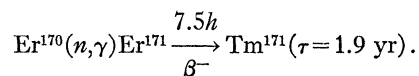


FIG. 8. Zero-field hfs level ordering for $(I/J)=\frac{5}{2}$, $(J/I)=6$, as a function of $\xi (=B/A)$.

Tm¹⁷¹

A 3-week irradiation at the Materials Testing Reactor at Arco, Idaho, on Er metal produced a sufficient quantity of radioactive Tm¹⁷¹ 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 Er¹⁶⁹ 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 Er¹⁷¹, 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.

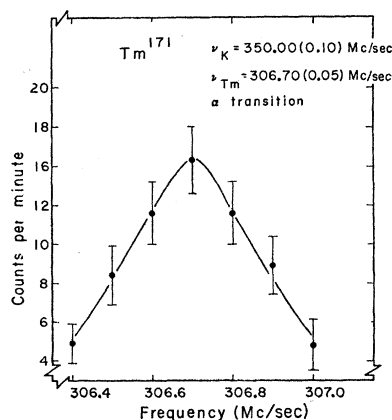


FIG. 9. Alpha transition at $\nu_K = 350.000 \text{ Mc/sec}$ ($H = 217.808 \text{ G}$, $F = 4$, $m_F = 0 \leftrightarrow -1$).

²² W. G. Smith, R. L. Robinson, J. H. Hamilton, and L. M. Langer, Phys. Rev. **107**, 1314 (1957).

²³ B. L. Sharma, Nucl. Phys. **25**, 175 (1961).

The results of these measurements are

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

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

When published data²⁴ are used on Tm¹⁶⁹, the nuclear moment of Tm¹⁷¹ 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 Tm¹⁷¹. The 22.5-keV peak represents the iodine escape peak of the K x rays of Yb¹⁷¹, i.e., the 67-keV level is highly converted, giving ≈ 53 -keV x rays (67 keV minus the binding energy of K electrons in Yb¹⁷¹). These ≈ 53 -keV x rays sometimes knock out the K electrons

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

a (Mc/sec)	g_J	Δa	Δg_J	χ^2
ν_K (Mc/sec)	H (G)	ν_{exp} (Mc/sec)	Residual Transition	
372.0710	-1.141155	5.9403	0.000097	1.25
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

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.

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 Pr¹⁴³ 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 Pr¹⁴¹ is $\frac{5}{2}$, indicating

²⁴ G. J. Ritter, Phys. Rev. **128**, 2238 (1962).

that the $g_{7/2}$ level lies lower. However, the spins of Pm^{147} and Pm^{149} are both $\frac{7}{2}$, 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 $\Delta 1 = 3$ would make this transition 1 forbidden. Kondaiah²⁶ predicts a spin of $\frac{7}{2}$ on the basis of x-ray multiplicities, ft values, and the spin-orbit coupling model. The measured spin of $\frac{7}{2}$ 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 $\frac{5}{2}$ reflects the abruptness of this transition. No shell-model state with $j = \frac{5}{2}$ is available in this region. On the other hand, an energy-level diagram for neutron numbers

TABLE IV. Comparison of measured and theoretical spin values.

Z	Element	A	Measured spin	Nilsson state or shell-model state	
				Odd proton	Odd neutron
59	Pr	143	spin-orbit coupling	$4g_{7/2}$	
60	Nd	149			$[523]_{\frac{5}{2}}^-$
65	Tb	161		$[411]_{\frac{3}{2}}^+$	
67	Ho	161		$[523]_{\frac{5}{2}}^-$	
68	Er	165			$[523]_{\frac{5}{2}}^-$

$82 < N < 126$ shows that a $[523]_{\frac{5}{2}}^-$ 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 approxi-

²⁵ P. W. Martin, M. K. Brice, J. M. Cork, and S. B. Burson, *Phys. Rev.* **101**, 182 (1956).

²⁶ E. Kondaiah, *Phys. Rev.* **83**, 471 (1951).

²⁷ B. R. Mottelson and S. G. Nilsson, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Skrifter I*, No. 8 (1959).

²⁸ S. G. Nilsson, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **29**, No. 16 (1960).

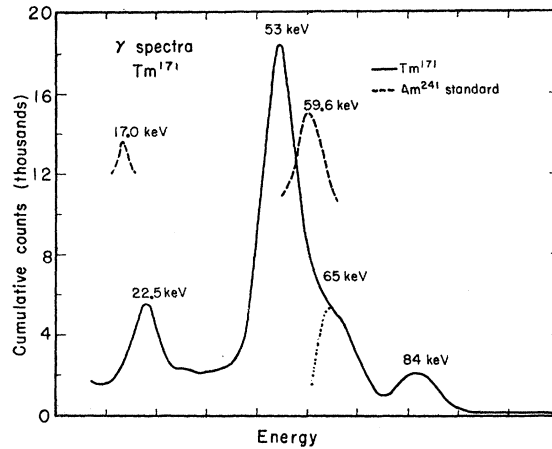


FIG. 10. Gamma spectra of Tm^{171} produced from $\text{Er}^{170}(n, \gamma)\text{Er}^{171} \xrightarrow{\beta^-} \text{Tm}^{171}$.

mations of the collective model. With a deformation ≈ 0.3 , which is about the same as other rare earths, the odd-particle state assignment is $[512]_{\frac{5}{2}}^-$. 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 $\mu_I = \pm 0.70(5)$ 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 $I = \frac{7}{2}$. 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 $[411]_{\frac{3}{2}}^+$, we find

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

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

The measured value, $|\mu| = 0.227(5)$, lies within these limits and corresponds to an intermediate value of the deformation parameter.