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Publication Date

1960-11-22

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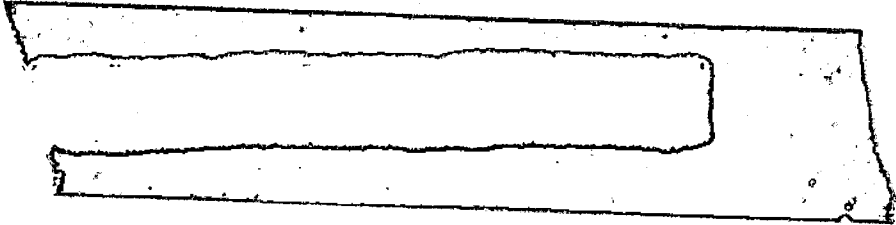
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ABSTRACT

A number of radioactive isotopes in the rare earth region have been investigated by using the atomic-beam magnetic-resonance technique. The total electronic angular momentum (J) and the atomic g value (g_J) have been determined for some low-lying levels in, Pm, Dy, Ho, and Er. These observations are consistent with the following ground-state assignments: PmI-- $(4f)^5(6s)^2$, $^6H_{5/2}$; DyI-- $(4f)^{10}(6s)^2$, 5I_8 ; HoI-- $(4f)^{11}(6s)^2$, $^4I_{15/2}$; and ErI-- $(4f)^{12}(6s)^2$, 3H_6 . The ground-level assignments agree with the Hund's-rule predictions, and the g values approximate the Landé values well. Our present experimental knowledge concerning electronic ground states in the rare-earth region is summarized. The following nuclear spin values have been obtained: Pm¹⁴⁹, $I = 7/2$; Pm¹⁵¹, $I = 5/2$; Gd¹⁵⁹, $I = 3/2$; Tb¹⁶⁰, $I = 3$; Dy¹⁶⁵, $I = 7/2$; Dy¹⁶⁶, $I = 0$; 27-hr Ho¹⁶⁶, $I = 0$; Er¹⁶⁹, $I = 1/2$, Er¹⁷¹, $I = 5/2$; and Tm¹⁷¹, $I = 1/2$. These values are compared with available beta-decay results. Most of these isotopes have large nuclear deformations, and the spin values are discussed in relation to the single-particle energy-level diagrams given by Nilsson. A few results are explained with the shell model.

ATOMIC-BEAM INVESTIGATIONS OF ELECTRONIC AND NUCLEAR
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INTRODUCTION

The elements with atomic numbers $Z = 57$ through 71 -- commonly known as the rare earth elements--exhibit a similarity in their chemical properties, and therefore occupy an unusual place in the periodic table. This similarity arises because successively added electrons in this range go into orbits that are embedded relatively deeply within the electronic cloud surrounding the nucleus. The chemical properties of elements are determined by the least tightly bound electrons and hence those that are outermost in the charge cloud. Therefore, the chemical properties are essentially uninfluenced by changing the number of electrons.

That the electrons in this region are embedded deeply in the core is a consequence of the peculiar effective potential seen by f electrons for high enough values of Z .¹ The orbits of electrons moving in this potential have been calculated in many ways in an effort to determine at what point in the periodic table the transition series involving $5f$ electrons begins^{1,2} and to determine the exact configuration of the rare earth ground states. From such a calculation, Hund³ postulated that the configurations were $(4f)^{n-1} (5d)^1 (6s)^2$.

*This work was done under the auspices of the U.S. Atomic Energy Commission and the Office of Naval Research.

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Experimental work by optical spectroscopy to determine the ground-state assignments of the rare earths has been inhibited because of the lack of well-separated rare earth samples and the enormous complexity of the observed spectrum, with the resultant difficulty in classifying lines. Hence, although spectroscopic measurements on the rare earths were made as early as 1912, by 1942 the ground-state configurations could be considered established for only seven of these elements.⁴ The war brought about the development of ion-exchange techniques for performing the separations necessary to obtain highly purified rare earth samples. However, although much progress has been made in classifying the lines of several rare earths and in measuring hyperfine structures and isotope shifts, only one additional configuration has been established--that of neodymium.⁵

The establishment of configurational assignments by the atomic-beams method rests on the ability to make direct measurements of the electronic angular momentum (J) and the g value for the state (g_J). This technique has been used effectively to establish the configuration of several elements in the actinide series.⁶ Because of the precision with which the g values can be obtained, the relative strengths of the spin-orbit and Coulomb interactions as well as the corrections brought about by relativity effects can be estimated.⁷

In this paper, we report the measurement of the J and g_J values of a number of rare earth elements and the configurational assignments inferred from them. In addition, a summary is given of some results obtained by other atomic-beam groups and by optical spectroscopy on the electronic structure of the rare earths. Finally a number of nuclear spins of neutron-produced isotopes scattered throughout the rare earth region are reported. Since in this region large deformations of nuclei become important, these spin values are expected to serve as a test for the applicability of the shell model and of the collective model.

EXPERIMENTAL METHOD

The measurements were all performed on neutron-activated radioactive isotopes by using the atomic beam method of Zacharias.⁸ By this method, resonances are observed when the frequency of the applied rf is equal to the precessional frequency of the atom in the applied magnetic field (H). At applied fields small enough that the nuclear spin (I) can be regarded as coupled to the electronic angular momentum (J) to form a total angular momentum (F), this precessional frequency (ν) is given by

$$\nu = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} \frac{\mu_0 H}{h}, \quad (1)$$

where g_J is the electronic g value and (μ_0/h) is the Bohr magneton divided by Planck's constant. A term in the nuclear magnetic moment has been neglected. Equation 1 provides the basis for the determination of all three quantities: g_J , J, and I. We note, however, that for a spin-zero nucleus, F equals J, and no information can be directly obtained concerning the J value.

Details of the apparatus and procedure used in these measurements are described elsewhere.⁹

BEAM PRODUCTION AND DETECTION

All the isotopes investigated were produced by neutron activation of some stable rare earth. In general, a quantity of the appropriate material (from 30 to 100 mg) was encapsulated in quartz and irradiated with an integrated neutron flux sufficient to produce about 100 mC of the desired activity. With one exception, the irradiated material was obtained commercially in the form of the pure metal with the natural isotopic abundance. In order to obtain increased specific activity, isotopically enriched Nd^{150} obtained in the oxide form was neutron-activated. Promethium-151 resulted from beta decay of the nucleus produced from the neutron activation.

The irradiated material was placed in a small tantalum cup with a sharp lip designed to prevent creep. The cup was placed in a tantalum oven which was then heated in the atomic-beam apparatus to beam temperatures. It was found, in all cases investigated, that materials produced by activation of the metal boil out of the oven in the form of a beam of atoms, with no further chemistry needed. Where the irradiated material was in the form of the oxide, a satisfactory chemical reduction was obtained by adding in the oven a lanthanum-cerium mixture known as "Mischmetall". The oven was heated by electron bombardment.

In all cases, the radioactive beam was collected on freshly flamed, uncooled, platinum foils. The collection efficiency is at least 25%, possibly 100%, and very highly reproducible. The radioactive deposit was measured by placing the foils in methane-filled proportional chambers and observing the beta decay.

EXPERIMENTAL RESULTS

The isotopes investigated include 50-hr Pm^{149} , 27-hr Pm^{151} , 18-hr Gd^{159} , 72-day Tb^{160} , 2.3-hr Dy^{165} , 82-hr Dy^{166} , 27-hr Ho^{166} , ¹⁰ 9.4-day Er^{169} , ¹¹ 7.8-hr Er^{171} , ¹¹ and 1.9-yr Tm^{171} . All observed resonances are listed in Table I, and a few examples of resonance curves are shown in Figs. 1 through 5. By use of Eq. (1), it is possible from these data to assign values of I, J, and F uniquely for almost all transitions, as indicated in the table. For Pm^{149} , where I equals J, the g_F values in the level observed are the same for all F states. Therefore, only one resonance is observed in this level. This is also the case for the 2.6-yr Pm^{147} , which has been investigated previously.¹² However, from the measurements on Pm^{151} , we can assign unambiguously the values of I and J in these cases. For Gd^{159} there are

a large number of states present in the beam, and some of the observed resonances might be a superposition of resonances originating from different states. Our observations fit very well the J and g values measured by Smith¹³ and a spin of $3/2$ (Fig. 5).

The average value of g_J and corresponding value of J for each observed level are summarized in Table II, and all spin values are given in Table III. In these tables we have also included the results obtained for 19-hr Pr¹⁴², 11-day Nd¹⁴⁷, 2.6-hr Pm¹⁴⁷, 47-hr Sm¹⁵³, and 129-day Tm¹⁷⁰, which have been reported separately.^{12, 14, 15, 16.}

The spin values are uniquely determined in all cases above except for the spin-zero results. With the technique employed here, one can not distinguish between a spin of zero and an extremely small magnetic moment. If the hyperfine structure is much smaller than the line-width, the resonance frequency becomes independent of the spin, within the experimental uncertainty. This means that if the spin of Dy¹⁶⁶ and Ho¹⁶⁶ is not zero, the moments can at most be of the order of 10^{-4} n. m.

ELECTRONIC GROUND STATES

In Table II we have given the most probable term assignments for the observed states. For Pr and Tm these are known to be the ground terms from optical and atomic-beam measurements.^{4, 17} The terms given for Pm, Dy, Ho, and Er are the Hund's-rule ground terms arising from configurations of the type $4f^n 6s^2$. In all these cases except Pm, the observed J values belong to the lowest level in each multiplet. The ground level of Pm should be $^6H_{5/2}$ according to Hund's rules, but the g value of this level is too small ($g_J \approx 0.28$) to be observed in our experiment.

The simple theoretical g values for pure Russell-Saunders coupling

(Landé values) are given in the fifth column of Table II. They agree well with the observed values. Therefore, there is very little doubt about the assignments made for the states observed in Pm, Dy, Ho, and Er. The observed deviations from the Landé values, which although small are quite significant, can be well explained by taking into account the effect of the spin-orbit coupling, the anomalous electron moment, and relativistic and diamagnetic effects.⁷ Since no indication of other states has been found, it is established almost beyond doubt that ground configurations in these cases are of the type $4f^n 6s^2$, and that the ground levels are in accordance with Hund's rules.

Of the elements not investigated here, all except terbium have well-established ground states. Values of J and g_J for some low-lying states in terbium have recently been measured by Penselin and Schlüpmann.¹⁸ In the accompanying paper⁷ it is shown that one of these states originates from the configuration $4f^9 6s^2$. The presence of other states which do not fit this picture indicates that another configuration, presumably $4f^8 5d 6s^2$, is very low-lying. The ground level of this configuration is probably $^8G_{15/2}$.

In Table IV we have collected the electronic ground states of all elements in this region according to the best available information at present. It is seen that the ground configurations show a striking regularity. The first element in each half shell definitely has a d electron in the ground state (La, Gd, and Lu). The second element seems to have a d electron at least in a very low-lying state (Ce and Tb). For all other elements, the ground configurations contain only f electrons. It is interesting to note that, when all electrons outside closed shells are equivalent, the most probable ground level is that predicted by Hund's rules. On the other hand, when two inequivalent groups of electrons are present, the coupling is not necessarily in accordance with these rules.

NUCLEAR SPIN VALUES

The observed spin values lie primarily in the region of mass numbers $150 < A < 190$, in which large nuclear deformations are known to occur. Therefore, our interpretation of the observed spins in this region are made on the collective model.^{19,20} In Figs. 6 and 7 the single-particle levels are plotted as a function of the deformation parameter, δ , for the odd-neutron and odd-proton cases, with $82 < N < 126$ and $50 < Z < 82$, respectively.²¹ The limit $\delta = 0$ gives the shell model, and the quantum numbers in this limit are the orbital angular momentum, l , and the total angular momentum, j . In the limit of large deformations, the appropriate quantum numbers are $[N n_z \Lambda] \Omega$, where N is the total oscillator quantum number, n_z is its component along the z axis, and Λ and Ω are respectively, the components of the orbital angular momentum and the total spin along the nuclear symmetry axis. In Table III, we give the measured spin for each isotope, the state of the last odd particle from the appropriate nuclear model, and, if available, the spin prediction from beta decay. The agreement with the beta decay is seen to be uniform.²²⁻³⁰ The spin assignments of Nd^{147} , Pr^{142} , and Sm^{153} are discussed elsewhere and will not be considered here.

The spins of the three isotopes Pm^{147} , Pm^{149} , and Pm^{151} with neutron numbers $N = 86, 88, \text{ and } 90$, respectively, are interesting in view of the spin change in going from 88 to 90 neutrons. At this point a sharp change is known to occur in the nuclear structure of europium as evidenced by the quadrupole-moment and isotope-shift data as well as the fact that the magnetic moment of Eu^{153} lies in the wrong Schmidt group.³¹ All of these properties can be accounted for on the assumption that Eu^{153} is highly deformed in relation to Eu^{151} . It seems reasonable, therefore, to assume that Pm^{151} is

highly deformed and that the sixty-first proton is in the state [532] 5/2. The spins of 7/2 for Pm^{147} and Pm^{149} can be explained on the assumption that they exhibit a deformation with $\delta \approx 0.05$ and that the state of the sixty-first proton is [404] 7/2, or that they go into the shell-model state g 7/2. However, the spin of ${}_{59}\text{Pr}^{141}$ is known to be 5/2,¹⁷ which indicates that the g 7/2 shell-model level lies lower than the f 5/2 level. This would seem to disagree with the shell-model assignment for the Pm isotopes. The nuclear moments of these isotopes are currently being measured to clarify the state assignments.

The spin 3/2 of Gd^{159} is the same as that of Gd^{155} and Gd^{157} . In view of the closeness of the orbitals [642] 5/2, [523], 5/2, and [521] 3/2, it has been suggested that the latter is the state of the ninety-fifth neutron.³⁴

The spin assignment of 3/2 for Tb^{159} has led to the assignment of the orbital [411] 3/2 for the sixty-fifth proton.²¹ On this basis, we have used the measured spin of Tb^{160} ($I=3$) to assign the orbital [521] 3/2 for the ninety-fifth neutron. This is the only plausible orbital giving agreement with the odd-odd coupling rules.³²

The measurement of spin zero for Ho^{166} would seem somewhat anomalous in view of the odd-proton and odd-neutron assignments. Both of these orbits are characterized by $\Omega = \Lambda + \Sigma$ and, according to the coupling rules for odd-odd nuclei, ought to exhibit a spin which is given by $\Omega_p + \Omega_n$.³² However, this may be a case where K equals 1/2, as discussed by Asaro et. al.³³

The spins of the erbium and dysprosium isotopes are in excellent agreement with the results of beta-decay measurements and the predictions of the collective model. The spin of Dy^{165} is the same as that of Er^{167} . Similarly Er^{169} and Er^{171} have the same spins as Yb^{171} and Yb^{175} ,

respectively. Thus the state assignment for the ninety-ninth, one-hundred-first, and one-hundred-third neutrons is very probably the same in these pairs of isotopes as those given in the ones in Table III. This shows that for deformations around $\delta = 0.3$, the order of the [633] $7/2$ and [521] $9/2$ levels is opposite to that in Fig. 7.

The observed spin of Tm^{171} ($I = 1/2$) further supports the assignment [411] $1/2$ as the state of the sixty-ninth proton.²¹ For the state of the one-hundred-first neutron, [521] $1/2$ has been assigned.³⁴ Since for both of these states Ω equals $\Lambda - \Sigma$, the spin of Tm^{170} is expected to be $\Omega_p + \Omega_n$,²⁸ which agrees with the measured value $I = 1$.

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Table I. Experimental observations

Z	El	A	I, J, F	H (gauss)	Observed frequency (Mc)	Calculated frequency (Mc)
61	Pm	149	(7/2, 7/2, all F)	2.818	1.650(70)	1.643
			(7/2, 7/2, all F)	5.567	3.250(50)	3.244
			(7/2, 7/2, all F)	20.755	12.060(50)	12.096
61	Pm	151	5/2, 7/2, 6	5.562	3.800(70)	3.785
			5/2, 7/2, 6	2.818	1.917(60)	1.916
			5/2, 7/2, 5	5.567	4.030(50)	4.001
			5/2, 7/2, 5	2.818	2.027(60)	2.026
64	Gd	159	(3/2, 3, 9/2)	4.201	8.000(200)	8.108
			(3/2, 4, 11/2)			7.861
			(3/2, 5, 13/2)			7.802
			(3/2, 2, 7/2)	8.900(200)	8.908	
			(3/2, 4, 9/2)		8.955	
			(3/2, 5, 11/2)		8.790	
			(3/2, 3, 7/2)	9.400(100)	9.273	
			(3/2, 2, 5/2)	9.700(100)	9.796	
			(3/2, 5, 9/2)	10.500(100)	10.448	
			(3/2, 4, 7/2)	11.100(100)	10.984	
(3/2, 3, 5/2)	11.700(100)	11.825				
66	Dy	165	7/2, 8, 23/2	8.248	10.120(60)	9.968
			7/2, 8, 21/2		10.630(40)	10.502
			7/2, 8, 19/2		11.400(100)	11.205
66	Dy	166	(0, 8, 8)	0.709	1.250(50)	1.241
			(0, 8, 8)	4.201	7.325(75)	7.299
			(0, 8, 8)	55.192	95.900(150)	95.899
			(0, 8, 8)	222.019	385.900(150)	385.774
			(0, 8, 8)	402.771	699.830(80)	699.843
67	Ho	166	(0, 15/2, 15/2)	0.709	1.175(60)	1.196
			(0, 15/2, 15/2)	5.567	9.350(100)	9.315
			(0, 15/2, 15/2)	55.192	92.420(100)	92.361
			(0, 15/2, 15/2)	93.043	155.700(150)	155.704
68	Er	169	1/2, 6, 13/2	0.709	1.070(50)	1.074
			1/2, 6, 13/2	1.418	2.150(50)	2.133
			1/2, 6, 13/2	4.201	6.375(75)	6.317
			1/2, 6, 13/2	8.248	12.450(50)	12.403
			1/2, 6, 13/2	15.920	23.960(50)	23.940
			1/2, 6, 13/2	25.387	38.150(150)	38.177
			1/2, 6, 11/2	0.709	1.250(50)	1.253
			1/2, 6, 11/2	1.418	2.475(50)	2.488
			1/2, 6, 11/2	4.201	7.400(50)	7.370
			1/2, 6, 11/2	8.248	14.600(50)	14.470
68	Er	171	5/2, 6, 17/2	4.201	4.875(50)	4.830
			5/2, 6, 17/2	8.248	9.560(40)	9.483
			5/2, 6, 15/2	4.201	5.280(30)	5.206
			5/2, 6, 15/2	8.248	10.325(40)	10.211
69	Tm	171	1/2, 7/2, 4	5.880	5.950(50)	5.872
			1/2, 7/2, 4	1.985	2.000(100)	1.982
			1/2, 7/2, 3	11.544	14.700(50)	14.823
			1/2, 7/2, 3	1.985	2.550(50)	2.548

Table II. Values of g_J and J for each observed level.

Z	El	Observed term	J	g_J (L-S)	g_J (exp't)
59	Pr	4_I	9/2	0.7273	0.7311 (2)
61	Pm	6_H	7/2	0.8254	0.831 (5)
			9/2	1.0707	1.068 (4)
66	Dy	5_I	8	1.2500	1.2414 (3) ^a
67	Ho	4_I	15/2	1.2000	1.1956 (12) ^a
68	Er	3_H	6	1.1667	1.164 (5) ^a
69	Tm	2_F	7/2	1.1428	1.1412 (2)

^aThese values agree with those independently obtained from spin-zero isotopes for Dy and Er by Spalding¹³ and for Ho by Goodman and Childs.¹⁰

Table III. Spin values.

Z	El	A	Measured spin	Nilsson state or shell-model state		Beta-decay predicted spin and parity
				Odd proton	Odd neutron	
59	Pr	142	2	4d _{5/2}	5f _{7/2}	2 ^{-b}
60	Nd	147	5/2 ^a		[5f _{7/2}] ⁻³ _{5/2}	
61	Pm	147	7/2	[404] 7/2		
		149	7/2	[404] 7/2		
		151	5/2	[532] 5/2		
62	Sm	153	3/2		[651] 3/2	3/2 ^c
					[521] 3/2	
64	Gd	159	3/2		[521] 3/2	3/2 ^{-d}
65	Tb	160	3	[411] 3/2	[521] 3/2	
66	Dy	165	7/2		[633] 7/2	7/2 ^{+e}
		166	0			
67	Ho	166	0	[523] 7/2	[633] 7/2	0 ^{-f}
68	Er	169	1/2		[521] 1/2	1/2 ^{-g}
		171	5/2		[512] 5/2	5/2 ^{-h}
69	Tm	170	1/2	[411] 1/2	[521] 1/2	1 ⁻ⁱ
		171	1/2	[411] 1/2		

^aAlso measured by means of paramagnetic resonance. (reference 22).

^bReference 23.

^cReference 24.

^dReference 25.

^eReference 26.

^fReference 27.

^gReference 28.

^hReference 29.

ⁱReference 30.

Table IV. Ground states of lanthanide elements.

Z	Element	Ground Configuration	Ground Level
57	La	5d	$2D_{3/2}^a$
58	Ce	(4f5d)	
59	Pr	4f ³	$4I_{9/2}^b$
60	Nd	4f ⁴	$5I_4^c$
61	Pm	4f ⁵	$6H(5/2)$
62	Sm	4f ⁶	$7F_0^d$
63	Eu	4f ⁷	$8S_{7/2}^e$
64	Gd	4f ⁷ 5d	$9D_2^d$
65	Tb	4f ⁹	$6H_{15/2}^f$
		(4f ⁸ 5d)	$(8G_{15/2})^f$
66	Dy	4f ¹⁰	$5I_8$
67	Ho	4f ¹¹	$4I_{15/2}$
68	Er	4f ¹²	$3H_6$
69	Tm	4f ¹³	$2F_{7/2}^g$
70	Yb	4f ¹⁴	$1S_0^h$
71	Lu	4f ¹⁴ 5d	$2D_{3/2}^h$

^aReference 34.^bReference 17.^cReference 5.^dReference 35.^eReference 36.^fReference 23 and 7.^gReference 4.^hReference 37.

FIGURE LEGENDS

- Fig. 1. Observed Zeeman resonances in Tb^{160} .
- Fig. 2. Zeeman resonances observed in the three highest F states of Dy^{165} .
- Fig. 3. Observed resonance in Ho^{166} . This frequency sweep covered the range of g_J values between zero and 1.600.
- Fig. 4. Resonance curves for Er^{171} . These resonances are assigned to the hyperfine-structure system $I=5/2$ and $J=6$.
- Fig. 5. Resonances observed in Gd^{159} . The arrows indicate the predicted frequencies corresponding to the statistical J values and a nuclear spin of $3/2$.
- Fig. 6. Energies of odd proton levels ($50 < Z < 82$) as a function of deformation parameter as calculated by Nilsson. [After S. G. Nilsson Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 8, 21 (1958).]
- Fig. 7. Nilsson diagram for odd neutrons for $82 < N < 126$. [After S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 8, 22 (1958).]

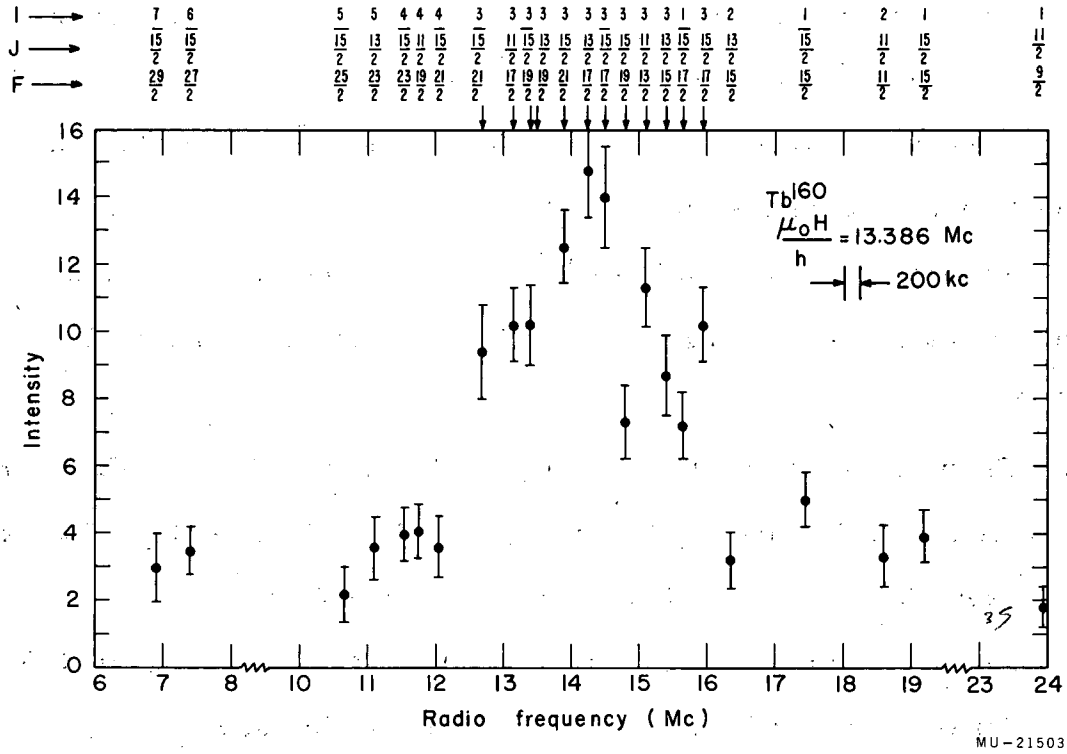
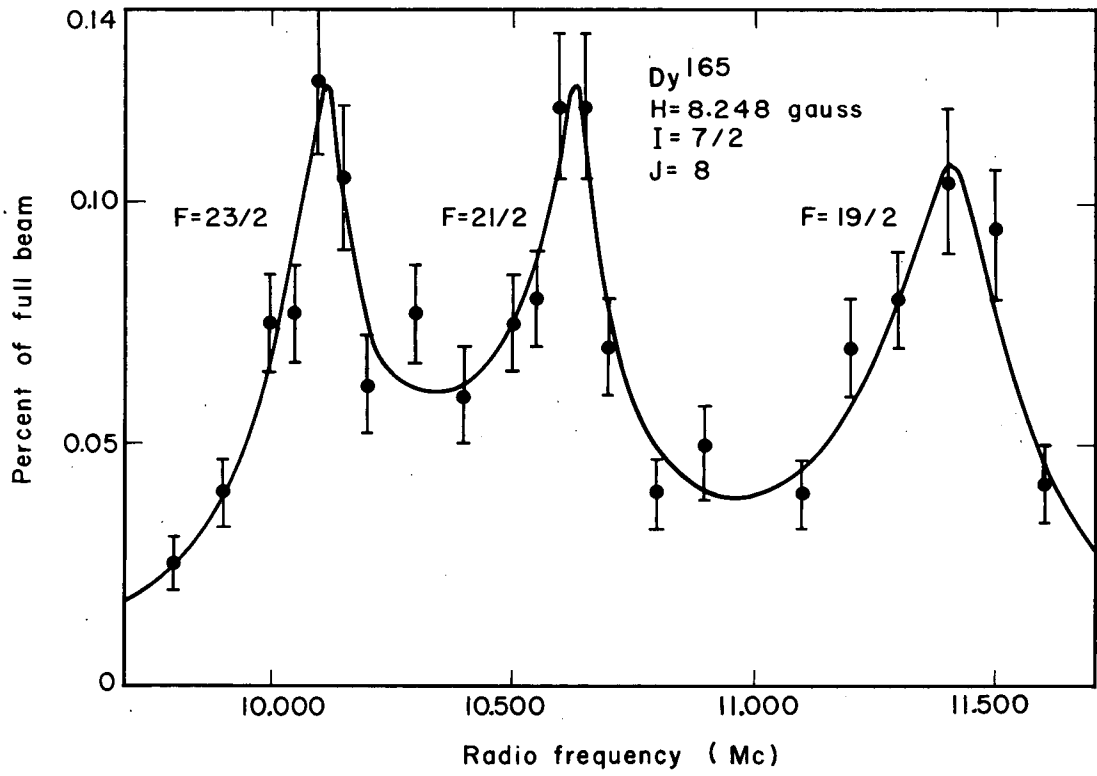
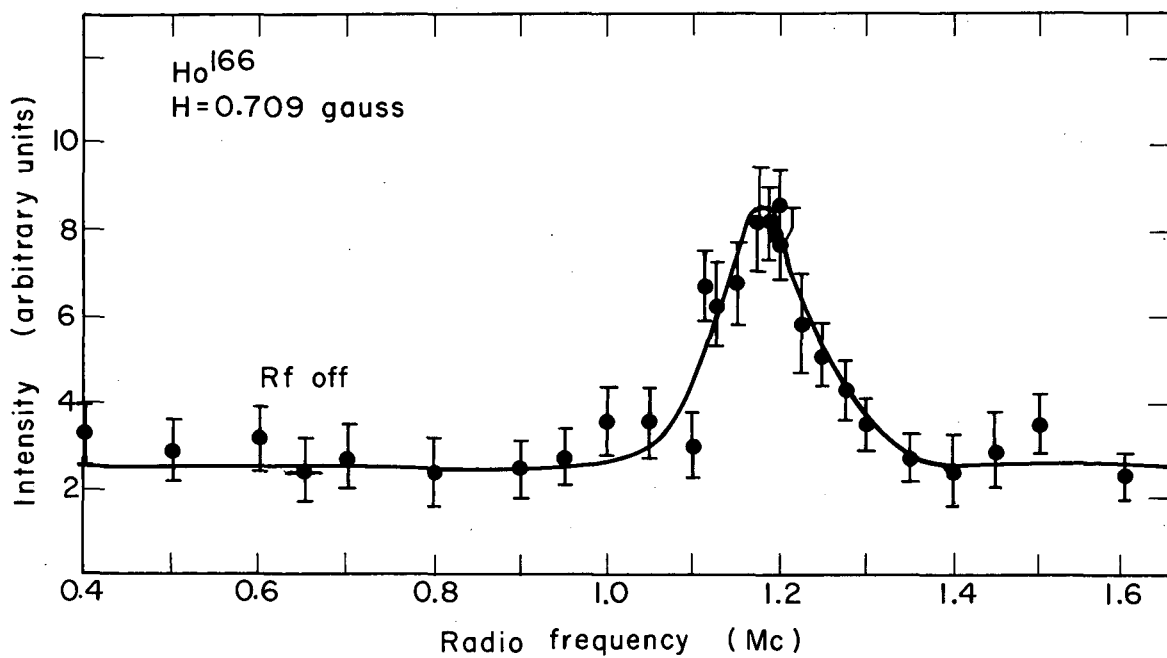


Fig. 1.



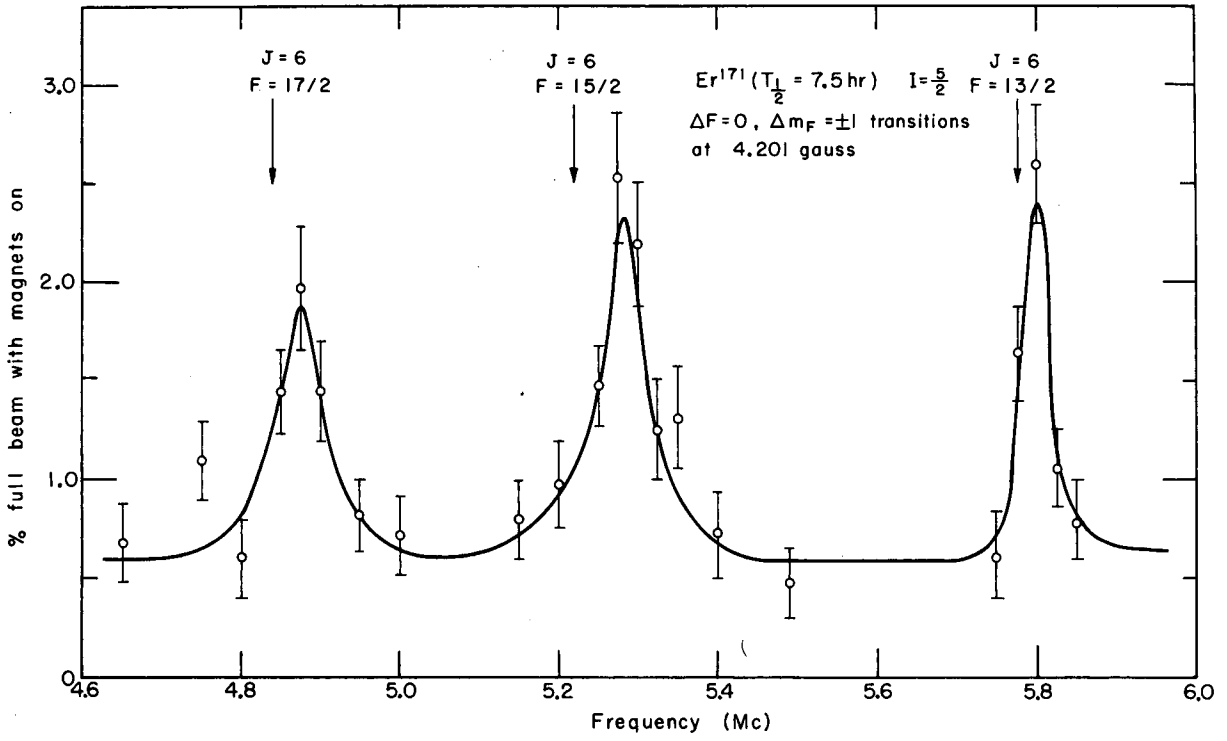
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Fig. 2.



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Fig. 3.



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Fig. 4.

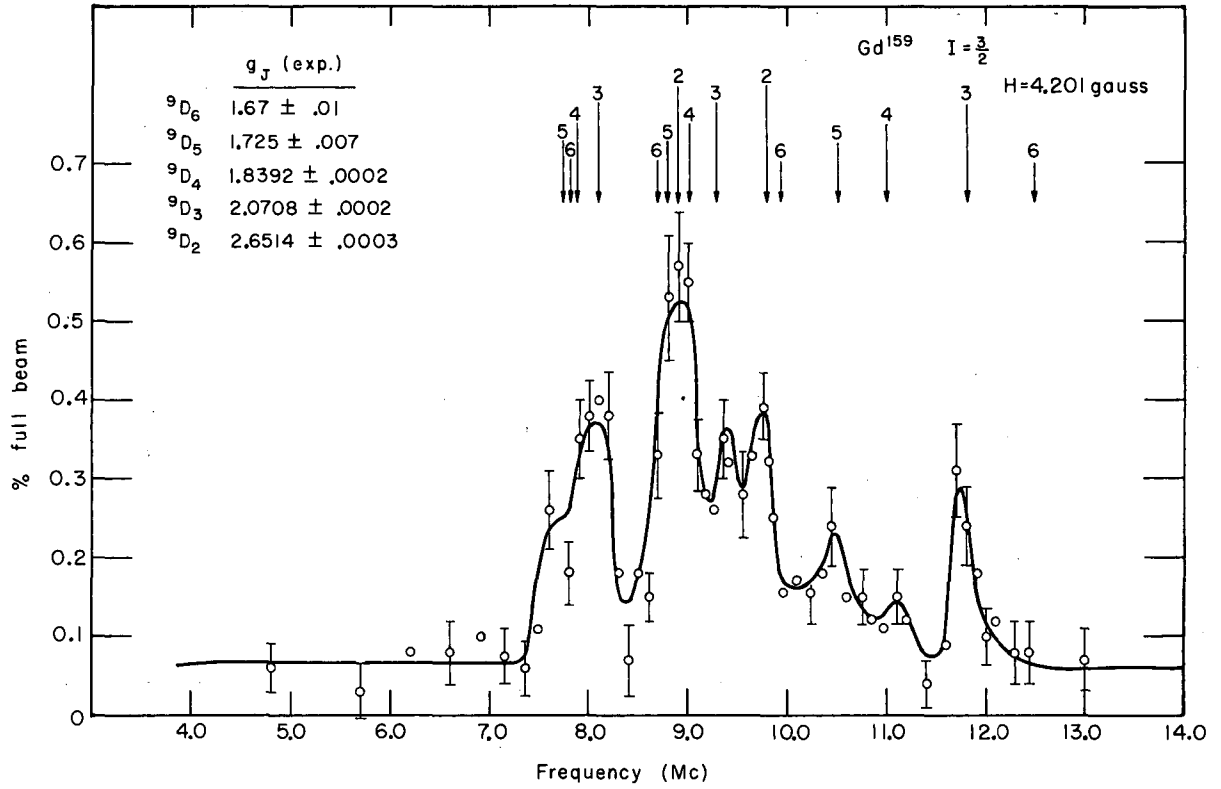
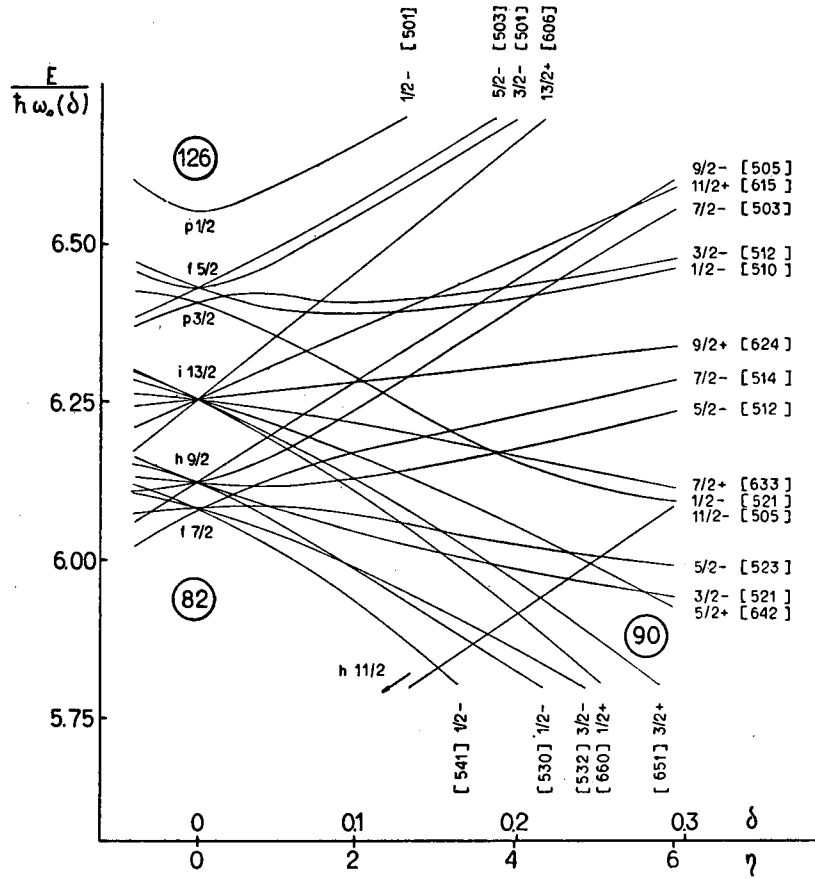


Fig. 5.



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Fig. 7.

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