

Hyperfine Structure and Nuclear Moments of Promethium-148 and Erbium-165†

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The hyperfine structure of the ${}^6H_{7/2}$ level in 5.3-day Pm^{148} and the 3H_6 level in 10-h Er^{165} have been measured by atomic-beam magnetic resonance. The spins of these isotopes have been verified to be $I(\text{Pm}^{148})=1$ and $I(\text{Er}^{165})=\frac{3}{2}$. The electronic g factor for the ${}^6H_{7/2}$ level in Pm^{148} is found to be $g_J=-0.8278(1)$. The hyperfine constants and inferred nuclear moments are,

$$\begin{array}{ll} \text{for } \text{Pm}^{148}: & A = +1038(75) \text{ Mc/sec}, & B = -98(103) \text{ Mc/sec}, \\ & \mu_I = +2.07(21) \text{ nm}, & Q = +0.2(2) \text{ b}; \\ \text{and for } \text{Er}^{165}: & |A| = 195(6) \text{ Mc/sec}, & |B| = 3502(115) \text{ Mc/sec}, \quad B/A < 0, \\ & |\mu_I| = 0.65(3) \text{ nm}, & |Q| = 2.2(1) \text{ b}, \quad Q/\mu_I > 0. \end{array}$$

The nuclear moments of Pm^{148} were calculated from the hyperfine constants and assumptions made concerning the electronic fields at the nucleus; these assumptions include a 2% correction for the breakdown of Russell-Saunders coupling, but do not include corrections for the Sternheimer effect. The stated errors include a 5% uncertainty for $\langle 1/r^3 \rangle$. The Fermi-Segrè formula and the direct measurement of the magnetic moment of Er^{167} were used to calculate the dipole moment of Er^{165} . The quadrupole moment of Er^{165} has not been corrected for the Sternheimer effect. The measurements indicate that Pm^{148} may be described by the shell model, but that Er^{165} is deformed and must be interpreted by the collective model.

INTRODUCTION

THE occurrence of large nuclear electric-quadrupole moments in the rare-earth region provides evidence for the existence of strong correlations in nucleon motions. Experiments have shown that these collective effects become important in nuclei with neutron numbers N greater than about 88. This research on Pm^{148} and Er^{165} is part of a continuing program to determine whether the onset of collective effects for $N > 88$ is valid for rare-earth isotopes in general and to test the applicability of various nuclear models to the specific nuclei.

Precision measurements of the quantities A (magnetic-dipole hyperfine constant) and B (electric-quadrupole hyperfine constant) by the method of atomic-beam magnetic resonance may yield information about the nuclear properties μ_I (magnetic dipole moment) and Q (electric quadrupole moment). The relations between the directly measured and inferred quantities are

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

and

$$B = - e^2 \langle q_J \rangle_{m_J=J} Q,$$

where I is the nuclear spin, J is the electronic angular momentum, $\langle H_z \rangle_{m_J=J}$ is the magnetic field produced at the nucleus by the electrons, and $\langle q_J \rangle_{m_J=J}$ is the electric-field gradient at the nucleus. To infer reliable values for μ_I and Q from the hyperfine constants requires knowledge of the electronic parameters. Values of J and, to within a few percent, values of $\langle H_z \rangle_{m_J=J}$ are available

for the ground states of almost all the rare earths.¹ However, large uncertainty in the quadrupole shielding and antishielding factors,² and therefore in $\langle q_J \rangle_{m_J=J}$, prevent our obtaining reliable values of Q from the hyperfine constant B .

EXPERIMENTAL METHOD, OBSERVATIONS, AND DATA ANALYSIS

Discussion of the principles and techniques involved in the application of the atomic-beam magnetic-resonance flop-in method to the study of radioactive nuclei, and details of the apparatus used are given in Refs. 1 and 3. A beam of atoms is produced by electron-bombardment heating of a tantalum oven containing the sample material. Atoms are detected by collection on platinum foil and subsequent counting by continuous-flow beta counters.

At low magnetic fields where I and J couple strongly to a total angular momentum F , the transition frequency (Zeeman frequency) between magnetic sub-states of the same hyperfine structure level is

$$\nu = g_F (\mu_0 H / h),$$

where

$$g_F \approx g_J [F(F+1) + J(J+1) - I(I+1)] / 2F(F+1).$$

At intermediate fields where the Zeeman approximation is no longer valid, information about the hyperfine constants may be obtained. The theoretical transition

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¹ For a summary, see Richard Marrus and William A. Nierenberg, in *Proceedings of the International School of Physics Enrico Fermi, Course XVII* (Academic Press Inc., New York, 1962), pp. 118-156.

² R. Sternheimer, *Phys. Rev.* **84**, 244 (1951) and **86**, 316 (1952).

³ William A. Nierenberg, *Ann. Rev. Nucl. Sci.* **7**, 349 (1957).

frequency must be determined from the Hamiltonian

$$\mathcal{H} = A\mathbf{I} \cdot \mathbf{J} + B \frac{3(\mathbf{I} \cdot \mathbf{J})^2 + \frac{3}{2}(\mathbf{I} \cdot \mathbf{J}) - I(I+1)J(J+1)}{2IJ(2I-1)(2J-1)} - g_J \mu_0 (\mathbf{J} \cdot \mathbf{H}) - g_I \mu_0 (\mathbf{I} \cdot \mathbf{H}).$$

Direct diagonalization of \mathcal{H} is performed by an IBM 7090 program described elsewhere.⁴ The observed resonance frequencies are fitted to the theoretical frequencies calculated from \mathcal{H} by adjustment of the parameters A , B , and g_J . As part of its output, the computer program yields final values of A and B , a goodness-of-fit parameter (χ^2), and frequency residuals. These residuals are the differences between the observed and theoretical frequencies.

Promethium-148

The ground electronic configuration of promethium is $4f^6 6s^2$, and Russell-Saunders coupling of the electrons gives rise to a 6H ground term.⁵ Only the $J = \frac{7}{2}$ level of the ground term was observed; for this level Budick and Marrus measured $g_J = -0.8279(4)$.⁶ Promethium-148 has a 5.3-day half-life⁷ and a spin of $I = 1$.⁸

A chloride solution containing curie amounts of 2.7-yr Pm¹⁴⁷ was obtained from the Oak Ridge National Laboratory.⁹ The chloride was converted to nitrate by adding an excess of nitric acid to the solution and heating it to dryness. This nitrate powder (≈ 1 mg) was irradiated for 3 weeks at a flux of 5×10^{14} neutrons/cm² sec to produce a sample of Pm¹⁴⁸. After irradiation the powder was dissolved in a weak solution of nitric acid, transferred to the inner liner of an oven, and heated in air to 700°F to convert the nitrate to oxide. A sharp-tipped tantalum-oven inner liner prevented creeping. A beam of promethium atoms was obtained by adding misch metal—primarily a mixture of lanthanum and cerium—to the liner and heating the oven in the beams machine to about 1000°C. The presence of Pm¹⁴⁷ and the atoms in the $J = \frac{5}{2}$ ground electronic level gave rise to such a large background that the signal-to-noise ratio was only 2:1.

⁴ Hugh L. Garvin, Thomas M. Green, Edgar Lipworth, and William A. Nierenberg, Phys. Rev. **116**, 393 (1959).

⁵ Amado Y. Cabezas, Ingvar Lindgren, and Richard Marrus, Phys. Rev. **122**, 1796 (1961).

⁶ Burton Budick and Richard Marrus, Phys. Rev. **132**, 723 (1963).

⁷ G. W. Parker, P. M. Lantz, M. G. Inghram, D. C. Hess, Jr., and R. J. Hayden, Phys. Rev. **72**, 85 (1947); J. D. Kurbatov and M. L. Pool, Phys. Rev. **63**, 463A (1943).

⁸ Dolores E. Ali, Ph.D. thesis, Lawrence Radiation Laboratory Report UCRL-11536, 1964 (unpublished).

⁹ Union Carbide Nuclear Company, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

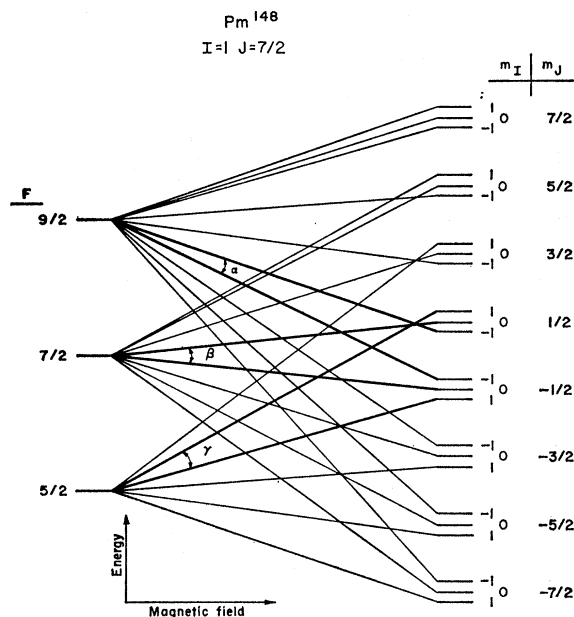


FIG. 1. Schematic Breit-Rabi diagram for Pm¹⁴⁸ ($I=1, J=\frac{7}{2}$).

The three observable flop-in transitions for Pm¹⁴⁸ are illustrated in the schematic Breit-Rabi diagram, Fig. 1. Typical resonance curves are shown in Figs. 2 and 3. Resonance data at fields between 67 and 375 G yield

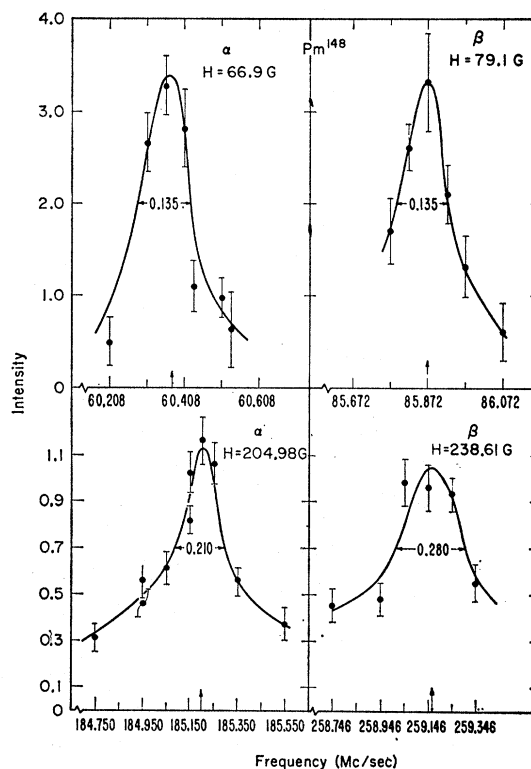


FIG. 2. Observed α and β transitions in Pm¹⁴⁸.

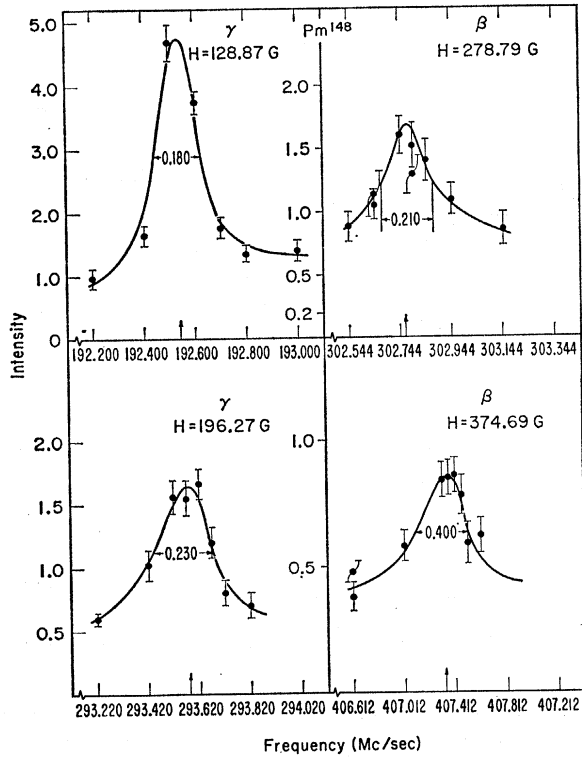


FIG. 3. Observed γ and β transitions in Pm^{148} .

final values of

$$A = +1038(75) \text{ Mc/sec,}$$

and

$$B = -98(103) \text{ Mc/sec,}$$

and a more accurate

$$g_J = -0.8277(2),$$

$$\begin{matrix} \text{Er}^{165} \\ I=5/2 \quad J=6 \end{matrix}$$

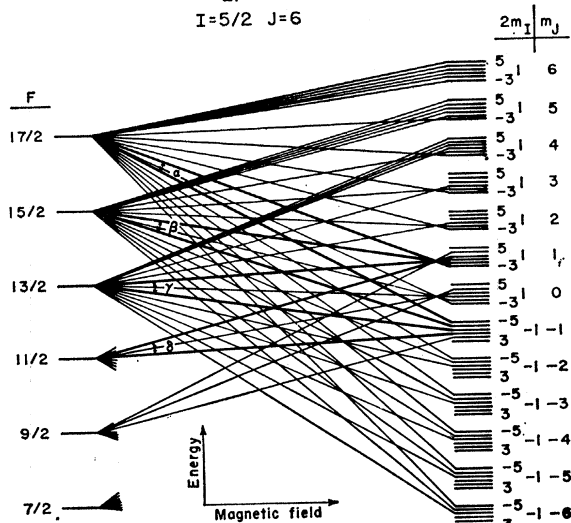


FIG. 4. Schematic Breit-Rabi diagram for Er^{165} ($I=5/2, J=6$).

where the quoted errors represent standard deviations. Table I contains the resonance data and results of the χ^2 test. This g_J , when combined with those obtained in experiments on Pm^{147} and Pm^{151} (Ref. 6), gives a weighted mean of $g_J = -0.8278(1)$ for the $J=7/2$ level.

The nuclear dipole and quadrupole moments inferred from the hyperfine constants are

$$\mu_I = +2.07(21) \text{ nm}$$

and

$$Q = +0.2(2) \text{ b.}$$

The Judd and Lindgren value of $\langle 1/r^3 \rangle = 4.87/a_0^3$ was used to estimate the fields at the nucleus.¹⁰ Our data are sufficiently sensitive to the $-\mathbf{u}_I \cdot \mathbf{H}$ term in the Hamiltonian that the sign of μ_I can be unambiguously inferred to be positive. This value for the magnetic

TABLE I. Promethium-148 resonances and results of χ^2 test (g_I positive).

ν_K (Mc/sec)	H (G)	ν_{exp} (Mc/sec)	Residual (Mc/sec)	Transition ^a
$A = +1038(75) \text{ Mc/sec}, B = -98(103) \text{ Mc/sec},$ $g_I = +(8.1 \pm 4.2) \times 10^{-4}, \chi^2 = 9.0.$				
64.000(148)	66.914(112)	60.368(40)	0.034	α
140.000(94)	117.682(54)	106.163(50)	-0.026	α
210.000(73)	154.669(36)	139.692(50)	0.049	α
240.000(80)	169.072(38)	152.609(55)	-0.071	α
280.000(87)	187.403(39)	169.300(50)	0.017	α
320.000(95)	204.989(41)	185.200(40)	-0.019	α
360.000(105)	222.017(44)	200.697(70)	0.037	α
400.000(113)	238.617(46)	215.726(50)	0.005	α
80.000(148)	79.131(108)	85.872(35)	0.016	β
160.000(94)	128.873(51)	139.851(75)	0.006	β
210.000(100)	154.669(49)	167.800(80)	-0.056	β
280.000(87)	187.403(39)	203.368(85)	-0.046	β
400.000(113)	238.617(46)	259.156(65)	0.070	β
500.000(115)	278.795(45)	302.764(75)	-0.036	β
750.000(187)	374.692(70)	407.332(70)	0.015	β
90.000(148)	86.252(102)	128.781(80)	0.046	γ
120.000(100)	105.803(61)	158.179(100)	0.202	γ
160.000(94)	128.873(51)	192.540(60)	0.031	γ
200.000(73)	149.711(37)	223.746(80)	0.022	γ
260.000(83)	178.346(38)	266.500(80)	-0.153	γ
300.000(90)	196.276(40)	293.580(60)	0.027	γ

^a Transitions— α : $F=3/2, M_F=-1/2 \leftrightarrow -3/2$.
 β : $F=3/2, M_F=1/2 \leftrightarrow -1/2$.
 γ : $F=5/2, M_F=3/2 \leftrightarrow 1/2$.

moment is in agreement with that obtained for Pm^{148} by Grant and Shirley in low-temperature nuclear-alignment experiments, $|\mu_I| = 1.82 \pm 0.19 \text{ nm}$.¹¹

The quoted error in our μ_I includes a 2% correction for the breakdown of Russell-Saunders coupling⁶ and a 5% uncertainty for $\langle 1/r^3 \rangle$. However, there is some indication that the Freeman and Watson $\langle 1/r^3 \rangle$ values are more appropriate for the earlier elements in the rare-earth region, whereas those of Judd and Lindgren are more appropriate for the heavier elements.¹² So the uncertainty in $\langle 1/r^3 \rangle$ may be greater than 5%. Corrections for the Sternheimer effect have not been included.²

¹⁰ B. R. Judd and I. Lindgren, Phys. Rev. **122**, 1802 (1961).

¹¹ R. W. Grant and D. A. Shirley, Phys. Rev. **130**, 1100 (1963).

¹² A. J. Freeman and R. E. Watson, Phys. Rev. **127**, 2058 (1962).

Erbium-165

Erbium has a ground electronic configuration of 4f¹²6s² (Ref. 5). In experiments on radioactive Er¹⁶⁹, Doyle and Marrus obtained $g_J = -1.16381(5)$ for the ³H₆ Russell-Saunders ground-state term.¹³

The half-life of Er¹⁶⁵ is 9.9(1) h,¹⁴ and its spin has been shown to be $I = \frac{5}{2}$.¹⁵

Either 1 g of the natural metal or 200 mg of an enriched erbium oxide (30 to 40% enriched in Er¹⁶⁴)¹⁶ was irradiated for 20 h at a flux of 9×10^{13} neutrons/cm² sec to produce a sample of Er¹⁶⁵. Misch metal proved effective in reducing the oxide.

The experiments on Er¹⁶⁵ were complicated by the presence of Er¹⁷¹ in the beam. Not only did this isotope contribute to machine background, but also because it has the same spin as Er¹⁶⁵ ($I = \frac{5}{2}$) and a similar half-life ($\tau_{\frac{1}{2}} = 7.5$ h), its resonances were difficult to distinguish from those of Er¹⁶⁵. For these reasons the oxide samples

TABLE II. Erbium-165 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	194.316	5.044	-3492.875	105.802	1.39
negative	195.181	5.042	-3510.431	105.721	2.15

νR (Mc/sec)	H (G)	ν_{exp} (Mc/sec)	Residual (Mc/sec)		Trans- sition ^a
			$g_I > 0$	$g_I < 0$	
20,000(50)	25.387(57)	59.782(200)	0.002	0.004	α
50,000(75)	55.192(66)	133.044(120)	0.048	0.037	α
140,000(140)	117.682(81)	291.900(200)	0.074	0.064	α
170,000(100)	134.255(53)	334.670(180)	0.013	0.006	α
210,000(120)	154.669(59)	387.670(150)	-0.066	-0.068	α
20,000(50)	25.387(57)	62.324(120)	0.052	0.049	β
50,000(75)	55.192(66)	133.984(140)	0.095	0.090	β
20,000(50)	25.387(57)	69.564(100)	-0.004	-0.007	γ
300,000(130)	196.276(57)	537.188(80)	-0.075	-0.102	γ
350,000(140)	217.804(59)	595.856(150)	-0.120	-0.150	γ
410,000(175)	242.712(72)	663.844(140)	0.014	-0.020	γ
20,000(50)	25.387(57)	79.958(140)	0.165	0.164	δ
300,000(130)	196.276(57)	617.092(140)	-0.115	-0.111	δ
350,000(140)	217.804(59)	685.240(140)	0.079	0.088	δ

^a Transitions— α : $F = 17/2, M_F = -\frac{3}{2} \leftrightarrow -\frac{1}{2}$.
 β : $F = 15/2, M_F = -\frac{1}{2} \leftrightarrow -\frac{3}{2}$.
 γ : $F = 13/2, M_F = \frac{1}{2} \leftrightarrow -\frac{3}{2}$.
 δ : $F = 11/2, M_F = \frac{3}{2} \leftrightarrow -\frac{1}{2}$.

containing less than 25% Er¹⁷¹ were used exclusively in the later experiments.

Flop-in transitions in the four highest F -states were observed. (See the schematic Breit-Rabi diagram, Fig. 4.) Typical resonance curves are shown in Figs. 5 and 6. Confirmation that the observed resonances correspond to those of 10-h Er¹⁶⁵ was obtained by decay of the resonance exposures. Resonance data were obtained at fields between 25 and 243 G; these data yield final

¹³ Walter M. Doyle and Richard Marrus, Phys. Rev. **131**, 1586 (1963).

¹⁴ F. D. S. Butement, Proc. Phys. Soc. (London) **A63**, 775 (1950); D. N. Kundu, J. D. Service, and M. L. Pool, Phys. Rev. **87**, 203A (1952).

¹⁵ Burton Budick, Isaac Maleh, and Richard Marrus, Phys. Rev. **135**, B1281 (1964).

¹⁶ The erbium oxide used in these experiments was obtained from Union Carbide Nuclear Company, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

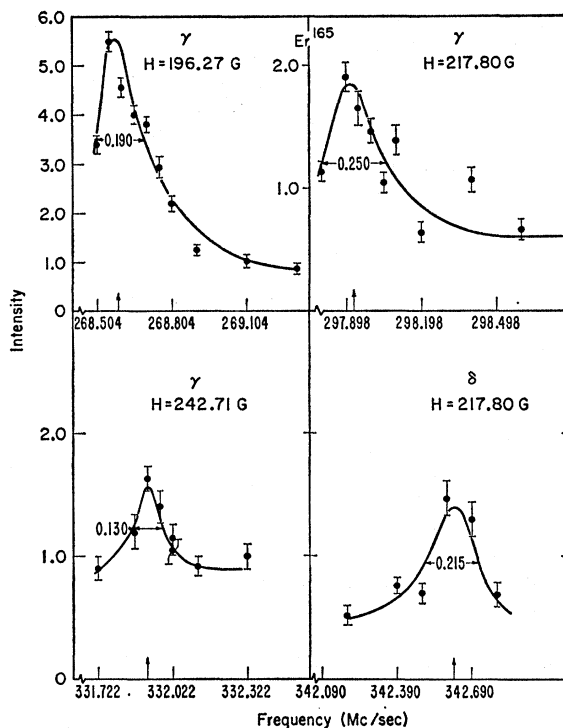


FIG. 5. Observed γ and δ transitions in Er¹⁶⁵ (metal).

values of

$$A = \pm 195(6) \text{ Mc/sec}$$

and

$$B = \mp 3502(115) \text{ Mc/sec.}$$

Table II contains the resonance data and results of the χ^2 test. The quoted A and B are averages of the values for g_I positive and g_I negative; quoted errors include the extremes of both positive and negative g_I .

From the measured hyperfine constants, A and B , the nuclear moments of Er¹⁶⁵ are $\mu_I = \pm 0.65(3)$ nm, and $Q = \pm 2.2(1)$ b with $Q/\mu_I > 0$.

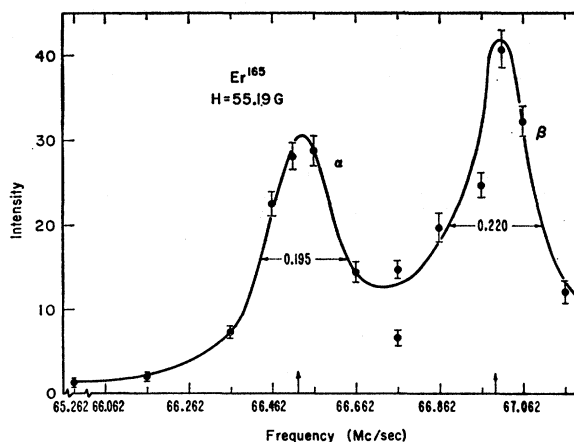


FIG. 6. Observed α and β transitions in Er¹⁶⁵ (oxide).

The Fermi-Segrè formula and the measured moment of Er^{167} (Ref. 17) were used to calculate the magnetic moment of Er^{165} . The quadrupole moment was calculated by use of $\langle 1/r^3 \rangle = 9.97(15)/a_0^3$ from the Er^{167} results to estimate the field at the nucleus. This value of $\langle 1/r^3 \rangle$ compares favorably with the Judd and Lindgren value, $9.66(48)/a_0^3$ (Ref. 10).

DISCUSSION

Nuclear Moments of Promethium-148

Comparison of the moments of Pm^{148} ($N=87$) with those of Pm^{147} and Pm^{151} (Ref. 6) confirms that collective effects do not become important for the promethium isotopes until $N \geq 88$. Shell-model state assignments of $g_{7/2}$ for the odd proton and $[(f_{7/2})^{-3}]_{5/2}$ for the odd neutrons are consistent with the measured spin ($I=1$).

Assuming j - j coupling among the odd nucleons, one calculates the Schmidt limit for the magnetic moment to be

$$\mu_I = +1.796 \text{ nm if free nucleon } g \text{ factors are used}$$

or

$$\mu_I = +1.934 \text{ nm if quenched } g \text{ factors are used.}^{18}$$

These values compare favorably with the empirical moment, $\mu_I = +2.07(21)$ nm.

If there is pure j - j coupling and the state assignments are $g_{7/2}[(f_{7/2})^{-3}]_{5/2}$, the ratio of the quadrupole moments of Pm^{148} and Pm^{147} (Ref. 6) should be

$$\frac{Q_{148}}{Q_{147}} = \frac{3C(C-1) - 4J_p(J_p+1)I(I+1)}{J_p(2J_p-1)(2I+1)(2I+3)} = 0.286,$$

where $J_p = \frac{7}{2}$, $J_n = \frac{5}{2}$, and $C = I(I+1) + J_p(J_p+1) - J_n(J_n+1)$. This ratio and the value $Q_{147} = 0.7(3)$ b yield the prediction that $Q_{148} = +0.20(9)$ b, which is close to the empirical value of $+0.2(2)$ b. It is apparent that the assumption of pure j - j coupling among the nucleons, which is inherent in the single-particle shell model, gives an excellent description of the nuclear moments.

¹⁷ K. F. Smith (private communication).

¹⁸ Lung-wen Chiao, Ph.D. thesis, Lawrence Radiation Laboratory Report UCRL-9648, 1961 (unpublished).

Nuclear Moments of Erbium-165

Quadrupole moments in the rare-earth region are expected to be positive. Since $Q/\mu_I > 0$ for Er^{165} , it follows that the magnetic moment of Er^{165} is probably positive. A collective-model state assignment $\frac{5}{2}^- [523]$ for the 97th neutron gives a positive moment, as well as the correct spin ($I = \frac{5}{2}$). However, the magnitude of the moment predicted by the collective model is considerably greater than the empirical moment $\mu_I = (+)0.65(3)$ nm. The Er^{165} nucleus has a deformation $\delta \approx 0.24$ (estimated from the quadrupole moment), so the collective-model prediction for the magnetic moment lies between

$$\mu_I = +1.223 \text{ nm; } \delta \approx 0.3$$

and

$$\mu_I = +1.122 \text{ nm; } \delta \approx 0.2.$$

Since Er^{165} and Er^{171} have the same spin, their hyperfine structures were expected to be similar. This is confirmed by the results for these two isotopes. (See Ref. 15 for the Er^{171} results.) But, interestingly, $Q/\mu_I < 0$ for Er^{171} (Ref. 15), indicating that its magnetic moment is negative. The difference in signs for the magnetic moments of these two isotopes is explained by the different state assignments for the 97th and 103rd neutrons.

The asymptotic expression for the collective-model magnetic moment is (except for $\Omega = \frac{1}{2}$)

$$\mu_I = (I/I+1)[g_i \Lambda \pm g_s(\frac{1}{2}) + g_R] \text{ nm,}$$

where $g_R \approx Z/A$, ($g_i = 0$, $g_s = -3.826$) for the neutron, and Λ and Ω are quantum numbers. (See Ref. 19.) The dominant term in the equation for an odd-neutron nucleus is $\pm g_s(\frac{1}{2})$, where the upper sign holds if $\Omega = \Lambda + \frac{1}{2}$ and the lower sign holds if $\Omega = \Lambda - \frac{1}{2}$. Since the state assignment $\frac{5}{2}^- [523]$ for the 97th neutron of Er^{165} has $\Omega = \Lambda - \frac{1}{2}$, whereas the state assignment $\frac{5}{2}^- [512]$ for the 103rd neutron of Er^{171} has $\Omega = \Lambda + \frac{1}{2}$, their moments are predicted to differ in sign.

ACKNOWLEDGMENTS

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¹⁹ B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Skrifter I, No. 8 (1959).