Hyperfine Structures of Re^{186} and Re^{188}†

RICHARD G. SCHLECHT,‡ MATTHEW B. WHITE,§ AND DOUGLAS W. MCCOLM
Lawrence Radiation Laboratory, University of California, Berkeley, California
(Received 7 December 1964)

The atomic-beam magnetic-resonance "flop-in" technique has been used to determine the hyperfine-structure separations of the isotopes Re^{186} and Re^{188}. The magnetic-dipole interaction constant a and the electric-quadrupole interaction constant b have been measured for these two radioactive isotopes in the $J=\frac{5}{2}$ ground state. Beams were produced by electron bombardment of irradiated rhenium wires. The spins of both isotopes had been determined previously to be 1. For the interaction constants of Re^{186} we obtained $a_{186}=\pm 78.3058(24)$ Mc/sec, $b_{186}=\mp 8.3601(50)$ Mc/sec. These give values for the two hyperfine-structure separations of $\Delta\nu_{186}(\frac{7}{2},\frac{5}{2})=\pm 265.292(14)$ Mc/sec, $\Delta\nu_{186}(\frac{5}{2},\frac{3}{2})=\pm 208.305(14)$ Mc/sec. For the interaction constants of Re^{188} we obtained $a_{188}=\pm 80.4320(32)$ Mc/sec, $b_{188}=\pm 7.7455(60)$ Mc/sec. These give values for the two hyperfine-structure separations of $\Delta\nu_{188}(\frac{7}{2},\frac{5}{2})=\pm 273.379(13)$ Mc/sec, $\Delta\nu_{188}(\frac{5}{2},\frac{3}{2})=\pm 212.698(17)$ Mc/sec. The nuclear moments of both isotopes were determined to be positive. Also obtained was an improved value for the electronic Landé g factor for rhenium: $g_J=-1.95203(8)$.

I. INTRODUCTION

THE atomic-beam magnetic-resonance method of Rabi[1] and Zacharias[2] has been used to measure the hyperfine-structure separations in the $(5d)^5(6s)^2\ {}^6S_{5/2}$ ground state of two radioactive isotopes of rhenium, Re^{186} and Re^{188}. The signs of the nuclear moments and an improved value of the rhenium g_J, which was first measured by Meggers,[3] were also obtained. The nuclear spins of Re^{186} and Re^{188} were determined to be 1 by Doyle and Marrus,[4] who also observed evidences of small hyperfine-structure separations. For a pure L-S coupling scheme, a ${}^6S_{5/2}$ state would have zero hyperfine structure. Perturbations to this coupling scheme give rise to the hyperfine structure in rhenium and would therefore be expected to be small. Other examples of this in atomic-beam experiments can be found in the papers of Sandars and Woodgate[5] and Marrus, Nierenberg, and Winocur.[6]

II. THEORY

The atomic-beam technique is a sensitive method for observing radio-frequency transitions between two energy states of a free atom in an external magnetic field. The Hamiltonian for such an atom is given by

$$\mathcal{H}=a\mathbf{I}\cdot\mathbf{J}+b\frac{3(\mathbf{I}\cdot\mathbf{J})^2+\frac{3}{2}(\mathbf{I}\cdot\mathbf{J})-I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}-g_I(\mu_0/h)\mathbf{I}\cdot\mathbf{H}_0-g_J(\mu_0/h)\mathbf{J}\cdot\mathbf{H}_0\,, \quad (1)$$

where a and b are the magnetic-dipole and electric-quadrupole hyperfine-structure interaction constants, respectively; $\mathbf{I}$ and $\mathbf{J}$ are the nuclear and electronic angular momenta in units of $\hbar$; $\mathbf{H}_0$ is the applied external magnetic field; and g_I and g_J are the nuclear and electronic g factors defined by μ_I/I and μ_J/J, respectively, where the magnetic moments μ_I and μ_J are in units of the Bohr magneton μ_0. Magnetic-octupole and higher order multipole-moment terms have been neglected. In the absence of an external magnetic field, the term energies resulting from this Hamiltonian are given by

$$W_F=a\frac{C}{2}+\frac{3b}{4}\frac{[C(C+1)-\frac{4}{3}I(I+1)J(J+1)]}{2I(2I-1)J(2J-1)}, \quad (2)$$

where

$$C=F(F+1)-J(J+1)-I(I+1), \quad (3)$$

and

$$\mathbf{F}=\mathbf{I}+\mathbf{J}. \quad (4)$$

The hyperfine-structure separation between two levels F and F' is given by

$$\Delta\nu(F,F')=W_F-W_{F'}, \quad (5)$$

where a, b, and $\Delta\nu$ are in the same units.

The qualitative features of the energy-level diagram for this Hamiltonian when $I=1$ and $J=5/2$ are given in Fig. 1. Also indicated are the five transitions that can be observed in each isotope with an atomic-beam machine with flop-in magnet geometry. Since a closed-form solution of the secular equation to the above Hamiltonian is, in general, not possible, a complete routine called HYPERFINE-3 (for the IBM 7090) was used to fit the experimental data to theory. The HYPERFINE routine is able to fit any combination of the five variables g_J, g_I, a, b, and c (the magnetic-octupole interaction constant) to the experimental data. Since

---

† This research was supported by the U. S. Atomic Energy Commission and the U. S. Navy.
‡ Present address: Westinghouse Research Laboratories, Pittsburgh 35, Pennsylvania.
§ Present address: Aeronutronic Division, Ford Motor Company, Newport Beach, California.
[1] I. I. Rabi, J. R. Zacharias, S. Millman, and P. Kusch, Phys. Rev. **53**, 318 (1938).
[2] J. R. Zacharias, Phys. Rev. **61**, 270 (1942).
[3] W. F. Meggers, J. Res. Natl. Bur. Std. **6**, 1027 (1931).
[4] W. M. Doyle and R. Marrus, Bull. Am. Phys. Soc. **7**, 605 (1962).
[5] P. G. H. Sandars and G. K. Woodgate, Proc. Roy. Soc. (London) **A257**, 269 (1960).
[6] R. Marrus, W. A. Nierenberg, and J. Winocur, Phys. Rev. **120**, 1429 (1960).

this routine is discussed elsewhere,[7,8,9] it is not reviewed here.

III. EXPERIMENTAL METHOD

Since the atomic-beam machine used in the course of this experiment was fully described elsewhere,[10] we will discuss only the essential features of this experiment. The isotopes Re^{186} and Re^{188} were produced by bombarding a 20-mil natural-rhenium wire with a neutron flux of 10^{14} neutrons/cm^2/sec. The isotopes Re^{186} and Re^{188} have half-lives of 90 h and 17 h, respectively, and could therefore be distinguished in a decay plot. The Re^{186} isotope was produced by bombarding the samples for 72 h and then allowing them to decay for 5 to 9 days. The amount of Re^{188} activity in the beam was less that 5%, as shown by decay plots of samples of the full beam and resonances. The Re^{188} isotope was preferentially produced by bombarding the samples for 3 to 4 h. As determined from decay plots, approximately 20% of the activity in the samples was Re^{186}. The beams were produced by heating the wires by electron bombardment. The atoms were collected on 1-mil fired-platinum foils which were then placed in a continuous-flow methane beta counter to measure their activity. A full-beam counting rate of from 600 to 1200 counts/min for a 1-min exposure was determined to be the most convenient. The magnitude of the homogeneous magnetic field H_0 was determined by observation of the Zeeman transition frequency in K^{39}.

IV. RESULTS

From Eq. (1) we see that the part of the energy due to the application of an external field H_0 in the z direction is given by the Hamiltonian

$$\mathcal{H}_{\text{ext}} = -g_J(\mu_0/h)J_zH_0 - g_I(\mu_0/h)I_zH_0. \tag{6}$$

In the weak-field case, **I** and **J** remain coupled and precess about **F**. The interaction energy due to the external field is then given by

$$W_{\text{ext}} = -g_F(\mu_0/h)m_FH_0, \tag{7}$$

where

$$g_F = g_J\frac{F(F+1)+J(J+1)-I(I+1)}{2F(F+1)} + g_I\frac{F(F+1)-J(J+1)+I(I+1)}{2F(F+1)}. \tag{8}$$

Therefore in the low-field approximation the transition

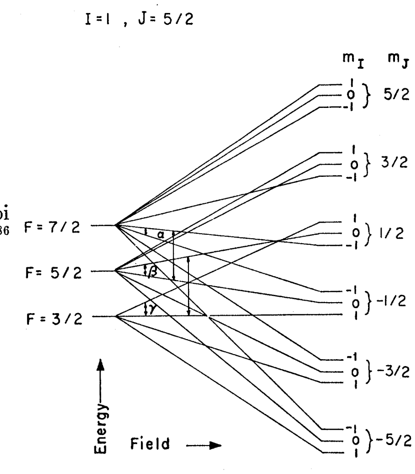

FIG. 1. Breit-Rabi diagram for Re^{186} and Re^{188}.

frequencies for the α, β, and γ transitions indicated in Fig. 1 ($\Delta F=0$, $\Delta m_F=\pm1$) are given by

$$\nu = -g_F(\mu_0/h)H_0. \tag{9}$$

Neglecting the nuclear term, the Zeeman transition frequency is

$$\nu = -\frac{(g_J\mu_0H_0)}{h}\frac{F(F+1)+J(J+1)-I(I+1)}{2F(F+1)}. \tag{10}$$

In the experiment the magnetic field H_0 was increased until the observed frequencies began to deviate from the Zeeman transition frequencies. Deviations were first seen at about 10 G. A third-order perturbation procedure was then used to predict the transition frequencies at higher fields. When enough points have been observed, the HYPERFINE-3 program was used to determine initial values of a and b. An IBM 650 computer program called JO-9 was then used to predict the transition frequencies at even higher fields, using the a and b values calculated by HYPERFINE-3. This procedure was continued until the values of a and b were known with sufficient accuracy that we could begin searching for the direct transitions ($F=7/2$, $m_F=-1/2$) $\leftrightarrow$ ($F=5/2$, $m_F=-1/2$) and ($F=5/2$, $m_F=1/2$) $\leftrightarrow$ ($F=3/2$, $m_F=1/2$).

Note that both direct transitions are σ transitions ($\Delta m_F=0$), whereas the α, β, and γ transitions are π transitions ($\Delta m_F=\pm1$). The π hairpin that was used to produce the α, β, and γ transitions was also used to produce the direct transitions. The part of this hairpin that carried the rf current consisted of a strip of metal whose plane was perpendicular to the external magnetic field. Therefore the only places where the radio-frequency magnetic field was parallel to the external magnetic field were at the edges of the strip. In this way separated oscillating fields, necessary for one to observe a Ramsey pattern, were produced. This type of hairpin construction was described by Schlecht and

[7] H. L. Garvin, T. M. Green, E. Lipworth, and W. A. Nierenberg, Phys. Rev. **116**, 393 (1959).
[8] W. A. Nierenberg, Lawrence Radiation Laboratory Report UCRL-3816 Rev., 1959 (unpublished).
[9] L. L. Marino, Lawrence Radiation Laboratory Report UCRL-8721, 1959 (unpublished).
[10] M. B. White, Lawrence Radiation Laboratory Report UCRL-10321, 1962 (unpublished).

TABLE I. Re^{186}: Magnetic-dipole, electric-quadrupole, and g variables.

a	b	g_J	$g_I\times10^4$	Error in a	Error in b	Error in g_J	Error in $g_I\times10^4$	χ^2
78.3058	−8.3601	−1.951997	11.444159	0.0012	0.0025	0.000044	6.939246	1.3444083

Energy levels and residuals

$b/a=-0.1068$

$\mu/h=1.399677$

$M_p/M_e=1836.12$

$\mu_I=2.101285$

Run No.	Frequency (Mc/sec)	Residual (Mc/sec)	Frequency error (Mc/sec)	F_1	M_1	F_2	M_2	H (G)	ΔH (G)	Weight factor
1	129.9300	0.0152	0.0150	5/2	1/2	5/2	−1/2	50.0003	0.0092	1122.8
2	480.1450	−0.0108	0.0150	7/2	−1/2	7/2	−3/2	200.0000	0.0044	2783.9
3	103.8550	0.0147	0.0150	7/2	−1/2	7/2	−3/2	50.0003	0.0092	1562.9
4	190.8450	0.0109	0.0150	3/2	3/2	3/2	1/2	50.0003	0.0092	810.8
5	78.3350	0.0045	0.0150	3/2	3/2	3/2	1/2	20.0000	0.0119	410.2
6	273.0000	0.0093	0.0150	5/2	1/2	5/2	−1/2	99.9995	0.0064	1807.8
7	221.4800	0.0087	0.0150	7/2	−1/2	7/2	−3/2	99.9499	0.0064	2119.9
8	204.1675	0.0000	0.0050	5/2	1/2	3/2	1/2	11.9999	0.0128	39862.6
9	268.6220	−0.0003	0.0070	7/2	−1/2	5/2	−1/2	12.0102	0.0128	14980.9

McColm.[11] Ramsey patterns were observed for both direct transitions in both isotopes. It was determined that the transition $(F=5/2,\ m_F=1/2)\leftrightarrow(F=3/2,\ m_F=1/2)$ attained a minimum transition frequency at approximately 12 G. Both direct transitions were observed for each isotope at this field in order to reduce the effect of the field error in the above transition.

Resonance curves representative of those obtained are shown in Figs. 2 and 3. A π transition is shown in Fig. 2 and a σ transition in Fig. 3. Since the oscillating fields are 180° out of phase, the Ramsey pattern in Fig. 3 shows a dip at the transition frequency rather than a peak.

Tables I and II give the final HYPERFINE-3 output for the isotopes Re^{186} and Re^{188}, respectively. A measure of how well the experimental points fit theoretical predictions based on the best values of a and b is the value of the goodness-of-fit parameter χ^2. Since χ^2 should have the value $N-N'$, where N is the number of observations and N' is the number of variables, for the Re^{188} data χ^2 should be 6 and for the Re^{186} χ^2 should be 5. The values 0.4 and 1.3, which were obtained by HYPERFINE-3 for Re^{188} and Re^{186}, respectively, indicate that the frequency errors were chosen pessimistically.

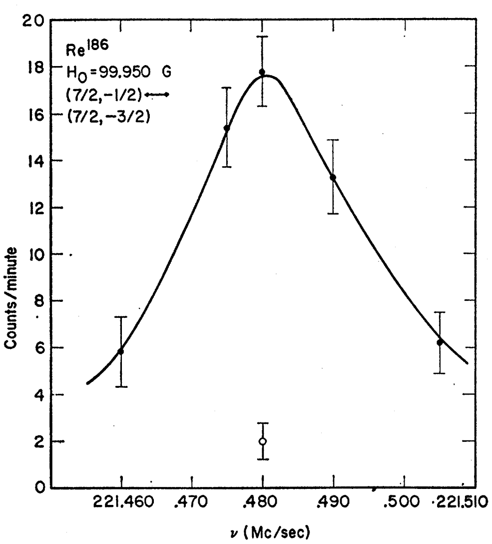

FIG. 2. Alpha transition in Re^{186} at 99.950 G.

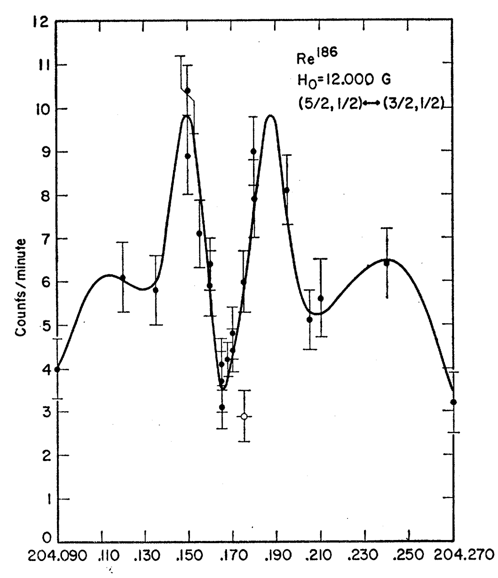

FIG. 3. $(\frac{5}{2},\frac{1}{2})\leftrightarrow(\frac{3}{2},\frac{1}{2})$ direct transition in Re^{186} at 12.000 G.

[11] R. G. Schlecht and D. McColm, Lawrence Radiation Laboratory Report UCRL-11773 (unpublished).

TABLE II. Re^{188}: Magnetic-dipole, electric-quadrupole, and g variables.

a	b	g_J	$g_I \times 10^4$	Error in a	Error in b	Error in g_J	Error in $g_I \times 10^4$	χ^2
80.4320	−7.7455	−1.952082	13.190500	0.0016	0.0030	0.000075	6.597555	0.38542005

Energy levels and residuals

$b/a = -0.0963$

$\mu/h = 1.399677$

$M_p/M_e = 1836.12$

$\mu_I = 2.421934$

Run No.	Frequency (Mc/sec)	Residual (Mc/sec)	Frequency error (Mc/sec)	F_1	M_1	F_2	M_2	H (G)	ΔH (G)	Weight factor
1	39.8850	0.0110	0.0150	7/2	−1/2	7/2	−3/2	20.0000	0.0119	1224.4
2	51.1350	0.0087	0.0150	5/2	1/2	5/2	−1/2	20.7547	0.0118	884.6
3	129.7250	−0.0041	0.0150	5/2	1/2	5/2	−1/2	50.0003	0.0092	1125.2
4	78.3300	0.0081	0.0150	3/2	3/2	3/2	1/2	20.0000	0.0119	409.8
5	191.2100	−0.0072	0.0150	3/2	3/2	3/2	1/2	50.0003	0.0092	802.2
6	272.9000	0.0037	0.0150	5/2	1/2	5/2	−1/2	99.9995	0.0064	1803.1
7	220.8750	−0.0008	0.0150	7/2	−1/2	7/2	−3/2	99.9995	0.0064	2129.8
8	103.6270	−0.0060	0.0150	7/2	−1/2	7/2	−3/2	50.0003	0.0092	1570.5
9	276.6650	−0.0001	0.0100	7/2	−1/2	5/2	−1/2	11.9999	0.0128	8548.5
10	208.4850	0.0000	0.0050	5/2	1/2	3/2	1/2	11.9999	0.0128	39973.2

Thus the frequency errors in a and b given by the HYPERFINE-3 program are probably too large. However, for security, twice these values have been used to set the uncertainty of the final results. Therefore we obtain for Re^{186} the values

$$a = \pm 78.3058(24)\ \text{Mc/sec},$$

$$b = \mp\ 8.3601(50)\ \text{Mc/sec},$$

and $g_I > 0$. For Re^{188} we obtain the values

$$a = \pm 80.4320(32)\ \text{Mc/sec},$$

$$b = \mp\ 7.7455(60)\ \text{Mc/sec},$$

and $g_I > 0$.

Taking the weighted average of the two values for g_J, we obtain

$$g_J = -1.95203(8).$$

For pure Russell-Saunders coupling, the g_J factor is given by[12]

$$g_J = 1 + (g_S - 1)\frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)},$$

where g_S is the g factor of the electron. Therefore, the g_J value for rhenium should be

$$g_J = g_S = -2.00229.$$

The major part of the discrepancy between this value and the experimental value come from the breakdown of Russell-Saunders coupling due to the spin-orbit and configuration interactions.

From Eqs. (2) and (5) we obtain, for the two hyperfine-structure separations in rhenium,

$$\Delta\nu(7/2,5/2) = (7/2)a + (21/20)b,$$

$$\Delta\nu(5/2,3/2) = (5/2)a - (3/2)b.$$

From this we obtain for the hyperfine-structure separations of Re^{186} the values

$$\Delta\nu_{186}(7/2,5/2) = \pm 265.292(14)\ \text{Mc/sec},$$

$$\Delta\nu_{186}(5/2,3/2) = \pm 208.305(14)\ \text{Mc/sec}.$$

For Re^{188} we obtain the values

$$\Delta\nu_{188}(7/2,5/2) = \pm 273.379(13)\ \text{Mc/sec},$$

$$\Delta\nu_{188}(5/2,3/2) = \pm 212.698(17)\ \text{Mc/sec}.$$

The application of these values in a more recent experiment is discussed in the next paper.[13]

[12] B. R. Judd, *Operator Techniques in Atomic Spectroscopy* (McGraw-Hill Book Company, Inc., New York, 1963).

[13] Lloyd Armstrong, Jr., and Richard Marrus, following paper, Phys. Rev. **138**, B310 (1965).