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ABSTRACT

The nuclear magnetic dipole and nuclear electric quadrupole interaction constants 'a' and 'b' have been measured in 8-day iodine-131 by an atomic beam magnetic resonance experiment. The results are

$$a = 575.903 \pm 0.007 \text{ Mc}$$

$$b = 578.866 \pm 0.075 \text{ Mc} \quad .$$

The zero-field hyperfine splittings computed from the above values of a and b are

$$\Delta\nu(5,4) = 3292.99 \pm 0.09 \text{ Mc}$$

$$\Delta\nu(4,3) = 2138.22 \pm 0.05 \text{ Mc}$$

$$\Delta\nu(3,2) = 1314.24 \pm 0.07 \text{ Mc} \quad .$$

The nuclear magnetic dipole moment and nuclear electric quadrupole moment are calculated as

$$\mu_{131} = 2.738 \pm 0.001,$$

$$Q_{131} = -0.41 \pm 0.01 \times 10^{-24} \text{ cm}^2 \quad .$$

The value of μ_{131} obtained here differs slightly from that obtained by other workers, while the value of Q confirms an earlier measurement made by a different method.

ATOMIC-BEAM MEASUREMENT OF THE HYPERFINE STRUCTURE AND NUCLEAR MOMENTS OF IODINE-131*

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INTRODUCTION

Atomic-beam magnetic-resonance techniques are currently being applied to the measurement of the nuclear spins and static nuclear moments of radioactive halogen nuclei.[1] This paper reports a measurement by the method of atomic beams of the hyperfine structure and nuclear moments of 8-day I^{131}. The value of the nuclear magnetic moment obtained differs slightly from that measured by Fletcher and Amble using microwave absorption spectroscopy.[2] The values of the nuclear spin and nuclear quadrupole moment confirm earlier measurements by Livingston et al.[3]

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EXPERIMENTAL METHOD AND THEORY

The atomic-beam apparatus and the experimental techniques used in this experiment have been described in detail elsewhere.[1] Radioactive I^{131} is mixed with stable iodine carrier and dissociated in a radio-frequency discharge tube or a heated platinum dissociation tube.[4] The atoms are passed through an atomic-beam apparatus designed to observe "flop-in-in"-type transitions in the way first proposed by Zacharias.[5] The theory and method of the present experiment are similar to those used by Davis et al.[6] in their study of the stable chlorine isotopes and by King and Jaccarino[7] on the stable bromine isotopes, and is identical with that described in connection with a recent experiment on bromine-82.[8] For completeness a brief description of the method of obtaining the interaction constants is given below.

The interaction Hamiltonian of an atom in a magnetic field $\vec{H}$ can be written[9]

$$\mathcal{H} = h.\, a\, (\vec{I} \cdot \vec{J}) + hbQ_{op} - \mu_0 (g_J \vec{J} \cdot \vec{H} + g_I \cdot \vec{I} \cdot \vec{H}), \qquad (1)$$

where

$$\vec{I} \cdot \vec{J} = \frac{1}{2} \left[F(F+1) - J(J+1) - I(I+1) \right],$$

$$Q_{op} = \frac{3(\vec{I} \cdot \vec{J})^2 + \frac{3}{2}(\vec{I} \cdot \vec{J}) - I(I+1)\, J(J+1)}{2I(2I-1)\, J(2J-1)}$$

I is the nuclear spin, J the electronic angular momentum and $\sqrt{F(F+1)}$ is the total angular momentum in units of $\hbar$; g_J and g_I are the electronic and nuclear g values; μ_0 is the Bohr magneton, and <u>a</u> and <u>b</u> are the magnetic dipole and electric quadrupole hyperfine interaction constants.

This Hamiltonian neglects interactions between the nucleus and electrons of higher order than electric quadrupole, and no account has been taken of perturbations due to possible configuration interaction. The constants a and b are related to the nuclear moments by the expressions[10,6]

$$\underline{a} = \frac{\mu_0^2}{h} g_I \mathcal{J} \frac{2\ell(\ell+1)}{J(J+1)} \langle \gamma^{-3} \rangle , \tag{2a}$$

$$\underline{b} = -\frac{e^2}{h} Q \mathcal{R} \frac{2\ell}{2\ell+3} \langle \gamma^{-3} \rangle ; \tag{2b}$$

g_I is given by $g_I = \frac{m}{M} \frac{\mu}{I}$, where μ is the nuclear magnetic dipole moment in nuclear magnetons; Q is the nuclear electric quadrupole moment. The relativistic correction factors $\mathcal{J}$ and $\mathcal{R}$ have the values $\mathcal{J} = 1.062$ and $\mathcal{R} = 1.128$ for iodine. Other symbols have their usual meanings.

The nuclear spin of I^{131} is 7/2,[3] and the electronic ground state is $^2P_{3/2}$. The energy levels of I^{131} are shown plotted versus magnet field in Fig. 1. The levels were obtained by solving the Hamiltonian (1) by means of an IBM 650 computer and with a and b values close to those finally obtained for I^{131}. In order to determine the hyperfine splittings the two transitions labelled (α) and (β) in Fig. 1 are observed with increasing magnetic field values and at each point the Hamiltonian is solved to yield approximate values of a and b. These values of a and b are used to predict the frequencies of the transitions at still higher fields, and the process is continued until a and b are known well enough so that a search can be made for the direct ($\Delta F = \pm 1$) hyperfine transitions at low field. As the computational labor involved in such a procedure is great, two IBM computer programs have been developed to facilitate the calculations. These programs have been described in detail elsewhere.[8] The first program calculates transition frequencies at any arbitrary magnetic field for given values of a and b, the second program makes a least-squares fit of the Hamiltonian (1) to any observed resonance data in order to provide best values of a and b. The latter program makes its fit with both signs of g_I and tests the goodness of fit by the χ^2 test of significance in both two cases.[11] It can thus be used to determine the sign of g_I and hence the sign of the nuclear moment if the data allow this.

The values quoted below for a and b are the values computed by the second of the two programs from all available input data (α, β and $\Delta F=\pm 1$ transitions) and the values of $\Delta\nu$ quoted below are those computed from these best values of a and b.

RESULTS

Table I contains all resonances that have been observed in I^{131} during the course of this experiment. The values of a and b computed from the 17 resonances with g_I taken positive are a = 575.903 ± 0.007 Mc, b = 578.866 ± 0.075 Mc. The uncertainties quoted are three times those actually resulting from the machine computations. The value of χ^2 with $g_I > 0$ is 3.4, but if g_I is assumed negative the value of χ^2 is 30 . Consultation of tables of χ^2 shows that the probability that g_I is positive is greater than 98%. Indeed, statistical arguments would require that if the choice of the uncertainties in the resonance centers had been made in the best way allowed by the data, the resulting value of χ^2 should be equal to the number of observations minus the number of degrees of freedom (i. e. 17-2=15). The small value of χ^2 obtained above for $g_I > 0$ therefore indicates there is justification for reducing the assigned uncertainties below the values we have chosen by a factor $\sqrt{\frac{15}{3.4}} \approx 2$. This would have the result of further increasing the probability that g_I is positive. In the last column of Table I we have listed the residuals, i. e., the differences between the observed resonance frequencies and those calculated using the best values of _a_ and _b_, and in Fig. 2 we have plotted the results in a way which shows very clearly the improved fit to the Hamiltonian if g_I is taken positive rather than negative.

Corrections to _a_ due to the finite size of the nuclear charge and magnetic-moment distribution are negligible for atoms of moderate Z in pure $P_{3/2}$ ground states and can be neglected, as can corrections arising from the mixing in of the $P_{1/2}$ state by the applied magnetic field.

The zero-field hyperfine-structure separation can best be obtained by solving the Hamiltonian (1) with I = 7/2 and J = 3/2. We find

$$\Delta\nu(5,4) = 5a + \frac{15}{21}\,b = 3292.99 \pm 0.09 \text{ Mc/sec},$$

$$\Delta\nu(4,3) = 4a - \frac{2}{7}\,b = 2138.22 \pm 0.05 \text{ Mc/sec},$$

$$\Delta\nu(3,2) = 3a - \frac{15}{21}\,b = 1314.24 \pm 0.07 \text{ Mc/sec}.$$

NUCLEAR MOMENTS OF I^{131}

A. Magnetic Moment

Following the method first proposed by Davis et al.,[6] $\left\langle \frac{1}{r^3} \right\rangle$ may be eliminated from Eq. (2a) by taking ratios between isotopes of the same element. We have

$$\frac{a^{131}}{a^{s}} = \frac{\mu^{131}}{\mu^{s}} \quad \frac{I^{s}}{I^{131}}, \qquad (4)$$

where s refers to the comparison isotope.

The magnetic moment of I^{127} has been measured by Walchli in a nuclear magnetic resonance experiment ($\mu^{127} = 2.8090 \pm 0.0004$, diamagnetically corrected),[12] and the _a_ value for I^{127} has been measured by Jaccarino, King, and Stroke in an atomic-beam experiment ($a^{127} = 827.265 \pm 0.003$).[13] Introducing these results into Eq. (4), we obtain

$$\mu^{131} = 2.738 \pm 0.001 \text{ nm}.$$

This value differs from the value 2.56 ±0.12 nm obtained by Fletcher and Amble.[2]

B. Electric Quadrupole Moment

From Eq. (2) we find

$$Q \equiv -\frac{8}{3}\left(\frac{\mu_o}{e}\right)^2\left(\frac{m}{M}\right)\frac{\mu}{I}(\mathcal{F}/\mathcal{R})\frac{b}{a} .$$

Using the known values of a μ, and I for the stable isotope I^{127} and the measured value of b for I^{131}, we find

$$Q = -\ 0.40 \text{ barn.}$$

In order to obtain the true nuclear quadrupole moment Q_t, a correction factor C, such that $Q_t = CQ$, is introduced. This factor C, first calculated by Sternheimer,[14] allows for the changed interaction of the valence electron with the inner core of electrons in the presence of the polarizing field due to the nuclear quadrupole moment. This constant has been calculated for iodine by Sternheimer but with the neglect of certain antishielding corrections. C is 1.029, but in view of the uncertainty in the exact value of C we have chosen to assign an uncertainty to Q_t equal to the value of the correction itself. Thus

$$Q_t = -\ 0.41 \pm 0.01 \text{ barn} .$$

This result is in agreement with an earlier measurement made by Livingston, Cox, andGordy, using microwave absorption spectroscopy.[3]

The authors wish to acknowledge the assistance of Mr. Larry M. Cohen who aided in the operation of the atomic beam apparatus. Members of the Radiation Laboratory Chemistry Department and Health Chemistry Group have made substantial conbributions to the success of this work.

Table I

Compilation of experimental resonance data observed in I^{131}

Resonance type	Frequency, f (Mc)	Calibration frequency, ν (Mc)	Alkali employed	δ_ν^a (Mc)	δ_f^a (Mc)	Residual (Mc) $g_I > 0$
α	49.600	90.0	K	0.080	0.073	0.024
β	34.900	90.0	K	0.080	0.063	0.004
β	48.175	137.0	K	0.100	0.047	-0.017
α	169.900	500.0	K	0.152	0.057	0.025
β	133.300	500.0	K	0.152	0.080	0.045
α	517.000	320.0	Cs	0.050	0.417	-0.277
β	529.400	320.0	Cs	0.050	0.203	0.006
α	355.750	220.0	Cs	0.088	0.250	-0.056
β	320.950	220.0	Cs	0.088	0.230	-0.025
α	805.800	500.0	Cs	0.175	0.480	-0.036
(4, 0)-(3, 0)	2138.230	0.5	K	0.025	0.030	0.005
(4, 0)-(3, 0)	2138.220	3.0	K	0.025	0.029	-0.003
(4, 3)-(3, 2)	2170.090	10.0	Cs	0.025	0.060	0.043
(4, 3)-(3, 3)	2170.880	10.0	Cs	0.025	0.070	0.107
(5, -3)-(4, -3)	3289.090	5.0	K	0.025	0.043	-0.035
(5, -3)-(4, -3)	3291.440	2.0	K	0.025	0.034	0.024
(4, 0)-(3, 0)	2138.210	2.0	K	0.025	0.029	-0.014

[a]The quantities $\delta\nu$ and δf are 1/4 and 1/3 of the resonance widths at half height of the calibrating beam and iodine resonances respectively.

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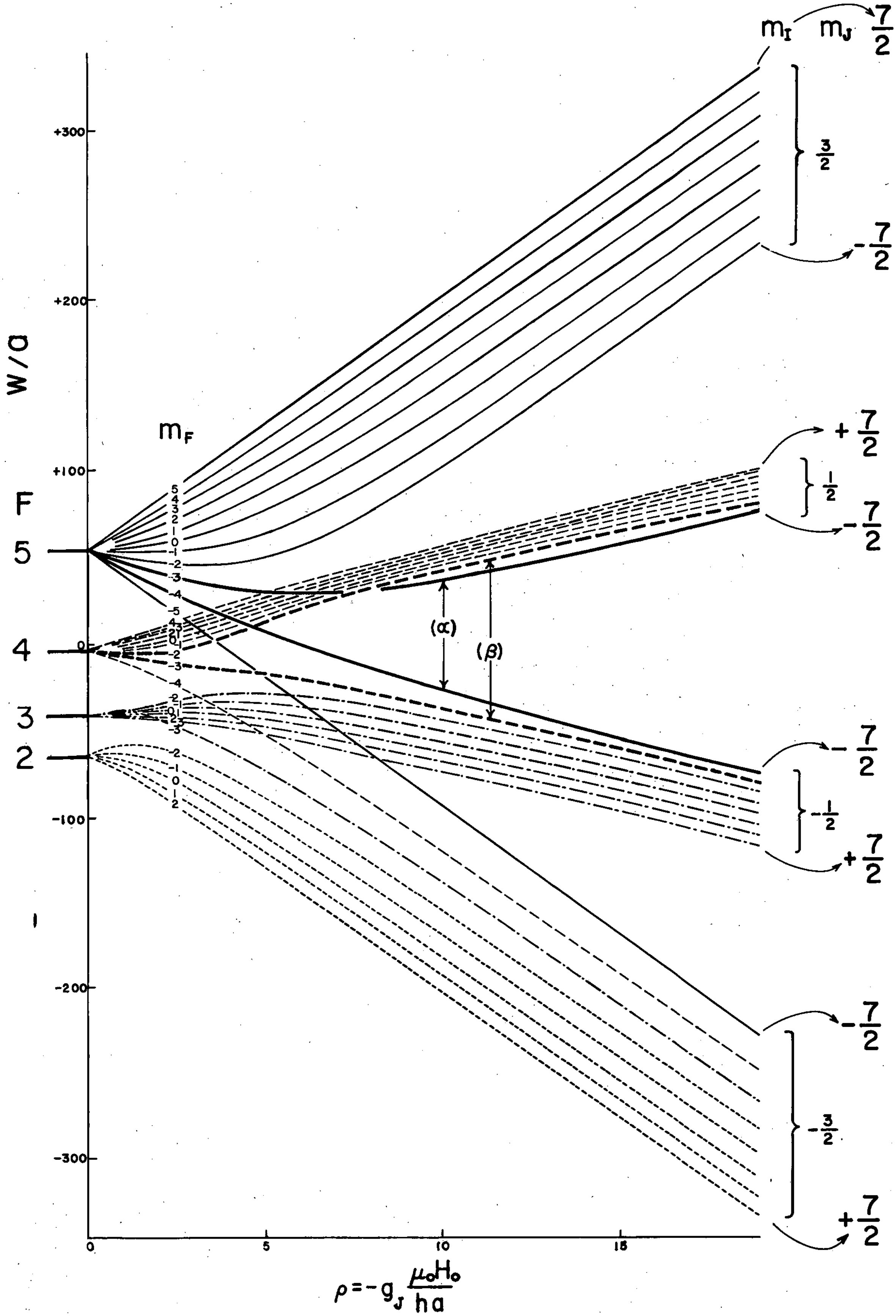
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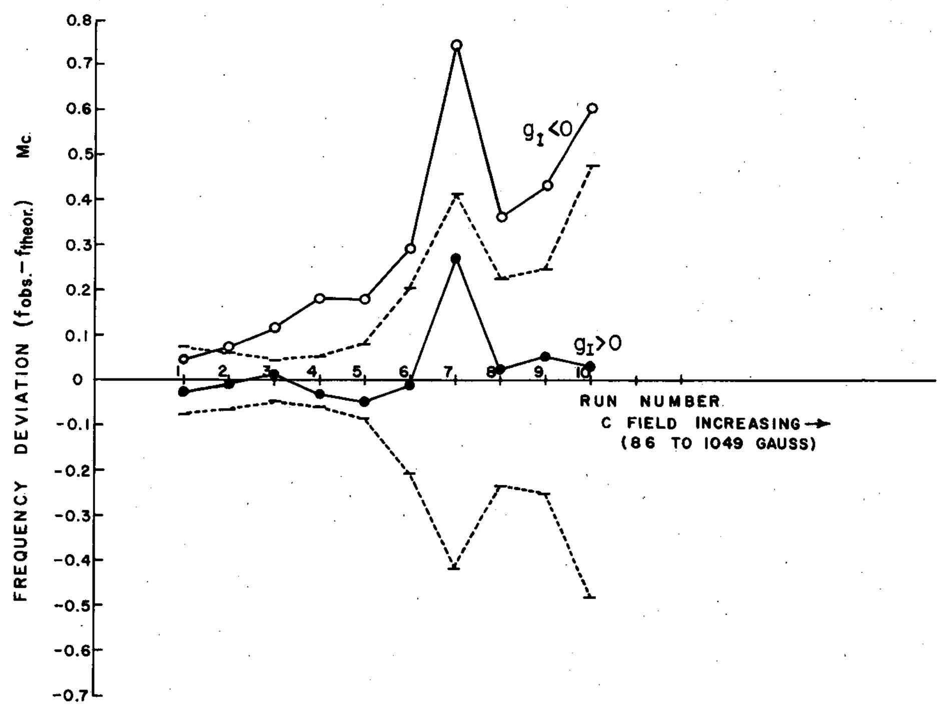

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Fig. 2. Test for the sign of g_I

$(f_{obs.} - f_{theor.})$ for $g_I < 0$

$(f_{obs.} - f_{theor.})$ for $g_I > 0$

Uncertainty of measurement

Plotted above are the differences between the observed resonance frequencies, and the frequencies calculated with the best values of a and b, for two signs of g_I. The dotted lines exhibit the experimental uncertainty. It is seen that $g_I > 0$ gives a consistent fit to the data.