Hyperfine Structure and Nuclear Moments of the Stable Bromine Isotopes*

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The hyperfine structure of the $^2P_{\frac{3}{2}}$ state of atomic Br^{79} and Br^{81} has been investigated by the atomic-beam, magnetic-resonance method. The following values have been obtained for the nuclear magnetic dipole interaction constant, a, and the nuclear electric quadrupole interaction constant, b:

$$a^{79}=884.810\pm0.003 \text{ Mc/sec}, \quad a^{81}=953.770\pm0.003 \text{ Mc/sec},$$
$$b^{79}=-384.878\pm0.008 \text{ Mc/sec}, \quad b^{81}=-321.516\pm0.008 \text{ Mc/sec}.$$

From these data, an upper limit of 0.0001 Mc/sec can be placed on the nuclear magnetic octupole interaction constant, c. Applying the results of recent theoretical calculations by Sternheimer and Koster, the following values of the nuclear quadrupole moments were computed:

$$Q^{79}=(0.33\pm0.02)\times10^{-24} \text{ cm}^2, \quad Q^{81}=(0.28\pm0.02)\times10^{-24} \text{ cm}^2.$$

The ratios (a^{79}/a^{81}) and (b^{79}/b^{81}) are in satisfactory agreement with the ratios (μ^{79}/μ^{81}) and (eqQ^{79}/eqQ^{81}), respectively, as measured by other methods.

An experimental technique is given for combining many of the advantages of the "flop-out" resonance method with those of the "flop-in" resonance method in the analysis of complex spectra.

Increased importance can be assigned to the hyperfine structure interaction constants of pairs of isotopes in view of the available information on the ground states of nuclei and on nuclear structure, and the fact that new methods exist for precise measurements of the ratio of nuclear moments.

I. INTRODUCTION

THE present work represents an extension of the study of the hyperfine structure of various halogen atoms by the atomic-beam method to the stable isotopes of bromine. Refinements in technique have made it possible to produce and detect a steady atomic beam of bromine. The analysis of the spectra of the two almost equally abundant isotopes ($Br^{79}=50.6$ percent; $Br^{81}=49.4$ percent) is facilitated by using a mass spectrometer to separate one from the other in the negative ion beam from the surface ionization detector.

Because of the similarity in theory and method of this experiment to that performed with the stable chlorine isotopes by Davis, Feld, Zabel, and Zacharias (DFZZ), it is suggested that the reader refer to the report of their work[1] whenever further amplification of our discussion is desired. A detailed description of methods for simplifying the study of hyperfine structure and for determining the sign of the nuclear moments is given.

The atomic ground state of bromine is a $^2P_{\frac{3}{2}}$ configuration. The energy difference between the $^2P_{\frac{3}{2}}$ state and the $^2P_{\frac{1}{2}}$ metastable state is 3685 wave numbers (1.105×10^8 Mc/sec). (The inversion of the fine structure doublets in the halogens is a result of the fact that the valence electron is a "missing" electron.) As is known, and has been confirmed in this experiment, the nuclear spin I, of both isotopes is $\frac{3}{2}$ in units of $\hbar$. Thus, with $I=J=\frac{3}{2}$, the Zeeman effect of the hfs interaction of bromine is similar to that of the two isotopes of chlorine, a discussion of which is given in DFZZ.

The Hamiltonian of the hfs interaction in a magnetic field H may be expressed as[2]:

$$\mathbf{H}=ha\mathbf{I}\cdot\mathbf{J}+hbQ_{op}+hcO_{op}+\mu_0(g_J\mathbf{J}\cdot\mathbf{H}+g_I\mathbf{I}\cdot\mathbf{H}), \quad (1)$$

where

$$\mathbf{I}\cdot\mathbf{J}=[f(f+1)-j(j+1)-i(i+1)]/2, \quad (2)$$

$$Q_{op}=\frac{3(\mathbf{I}\cdot\mathbf{J})^2+\frac{3}{2}(\mathbf{I}\cdot\mathbf{J})-i(i+1)j(j+1)}{2i(2i-1)j(2j-1)}. \quad (3)$$

The hfs interaction constants a and b for the case $j=l+\frac{1}{2}$ may be written as follows:

$$a=-\frac{\mu_0^2}{h}g_I\mathfrak{F}\frac{2l(l+1)}{j(j+1)}\langle r^{-3}\rangle, \quad (4)$$

$$b=-\frac{e^2}{h}QR\frac{2l}{2l+3}\langle r^{-3}\rangle. \quad (5)$$

Where $g_I=-(m/M)(\mu/I)$, μ being the nuclear magnetic moment in nuclear magnetons, Q is the nuclear electric quadrupole moment in units of 10^{-24} cm^2, $\mathfrak{F}=1.04$ and $R=1.05$ are two relativistic correction factors,[3] and the other symbols have their usual meanings. (This choice of the diagonal form for the Hamiltonian, that is, the assumption that the eigenvalues of J^2, J_z, I^2, and I_z are good quantum numbers, presupposes that the effects of neighboring electronic and nuclear levels on the ground state are small compared to the hfs interaction in the ground state, that is, in particular

$$E_{\text{fine structure}}(^2P_{\frac{3}{2}}\leftrightarrow{}^2P_{\frac{1}{2}})\gg E_{\text{hfs}}(^2P_{\frac{3}{2}}).$$

For $I=\frac{3}{2}$, multipole interactions of higher order than the magnetic octupole are forbidden.

For atoms with $I=J=\frac{3}{2}$, the calculated hfs energy levels in an external magnetic field are plotted, for $b=0$, in Figs. 1(a) and 1(b), where μ_I is assumed positive and negative, respectively.

II. APPARATUS

Since the apparatus used in this experiment is the same as that used in the chlorine experiments,[1,4–6] the

* This work was supported in part by the Signal Corps, the Air Materiel Command, and the U. S. Office of Naval Research.

[1] Davis, Feld, Zabel, and Zacharias (referred to as DFZZ), Phys. Rev. **76**, 1076 (1949).

[2] N. F. Ramsey in *Experimental Nuclear Physics* edited by E. Segrè (John Wiley and Sons, Inc., New York, 1953), Vol. 1, Chap. 3, pp. 358–467.

[3] H. B. G. Casimir, *On the Interaction Between Atomic Nuclei and Electrons* (DeErven F. Bohn N. V., Haarlem, 1936).

[4] Davis, Nagle, and Zacharias, Phys. Rev. **76**, 1068 (1949).

[5] V. Jaccarino and J. G. King, Phys. Rev. **83**, 471 (1951).

[6] J. G. King and V. Jaccarino, Phys. Rev. **84**, 852 (1951).

papers describing the latter experiments may be consulted for general information not found in the description of the modified details of the apparatus peculiar to the bromine experiment.

A. Atomic Beam Source and Detector

The microwave-discharge atom source that was used for chlorine was found adequate for bromine. Since, however, the needle valves or metallic leaks used to control the gas flow to the discharge in the case of chlorine operated erratically after exposure to bromine, the arrangement shown in Fig. 2 was adopted. The flow of bromine vapor was determined both by the vapor pressure of the liquid bromine in the reservoir, which was maintained at a constant temperature by a water bath with an adjustable thermostat, and by a fixed leak consisting of a 40-cm length of capillary into which a wire was inserted to reduce the cross-section area to 2×10^{-4} mm^2; the entire leak was kept at 70°C to prevent condensation of the vapor. The ease with which bromine could be trapped anywhere in the system with dry ice eliminated the need for many troublesome valves.

The consumption of liquid bromine was about 0.1 cc per hour. From this value and the measured detector ion currents, the detection efficiency of the hot tungsten ribbon for bromine was estimated to be 2×10^{-5}, in agreement with previous results.[7]

It was later found that detection efficiencies more closely approaching unity could be achieved for the halogens by using a properly activated thoriated tungsten ribbon. The low detection efficiency made it desirable to increase the sensitivity of the electrometer system (particularly since we intend to investigate the hyperfine structure of the $^2P_{\frac{1}{2}}$ metastable state).

To achieve greater electrometer sensitivity, an FP54 electrometer in a conventional circuit was coupled by means of an amplifier to an L and N galvanometer of sensitivity 0.00014 μa/mm. Part of the increased sensitivity that results from the use of the amplifiers is sacrificed in an equalizer network that reduces the effective time constant of the system.

With this arrangement the sensitivity and time constant could easily be adjusted so that changes in ion beam intensity comparable to the thermal fluctuations in the grid resistor of the FP54 could be observed. A simplified schematic of the system is given in Fig. 3. The batteries shown were continuously charged at a rate that maintained them near full charge.

B. Generation and Measurement of Transition Frequencies

The low-frequency $\Delta F=0$ transitions were observed with General Radio 805-C signal generators below 50 Mc/sec. In the range from 2200 to 2600 Mc/sec, a 707-B klystron was used with a set of simple cylindrical cavities chosen to cover each of the regions predicted from the low-frequency data in which it was necessary to search for high-frequency $\Delta F=\pm1$ transitions. Two other $\Delta F=\pm1$ transitions near 1300 Mc/sec were observed with a F4800 jamming transmitter using a 2C39-A lighthouse tube. All these high-frequency oscillators were operated by batteries or by well-filtered power supplies to minimize the output signal width, which was in all cases less than 1 kc/sec. Adequate output was obtained from all oscillators when optimally

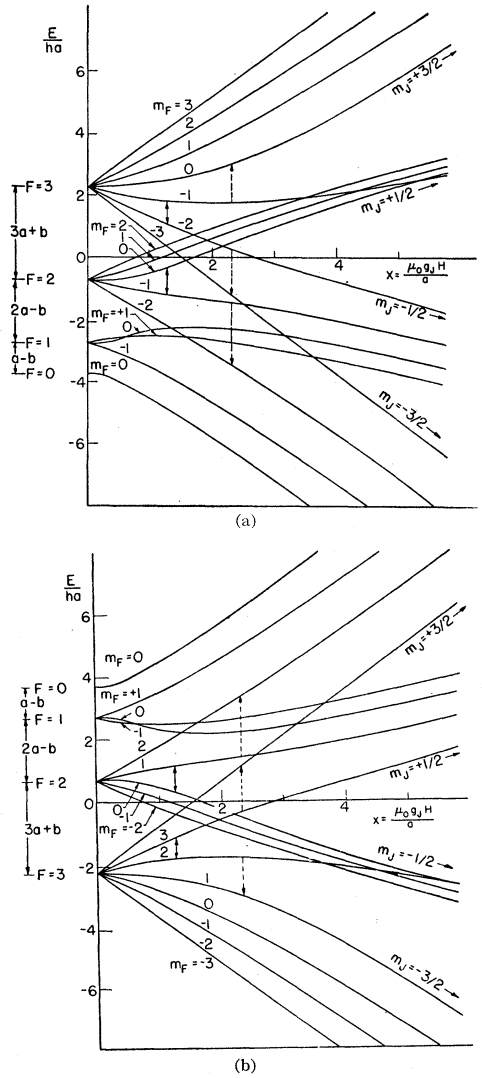

FIG. 1. Calculated hfs energy level diagrams for atoms with $I=J=(3/2)\hbar$ in an external magnetic field H. In plotting both curves, the quadrupole interaction constant b has been set equal to zero. Figure 1(a) was plotted with μ_I assumed positive, and Fig. 1(b) with μ_I assumed negative. The double arrows represent the normal $\Delta F=0$ "low-frequency," "flop-in" transitions; and the dashed arrows, the transitions observed as "flop-out on flop-in."

[7] H. H. Stroke, Massachusetts Institute of Technology, Master's thesis, 1952 (unpublished).

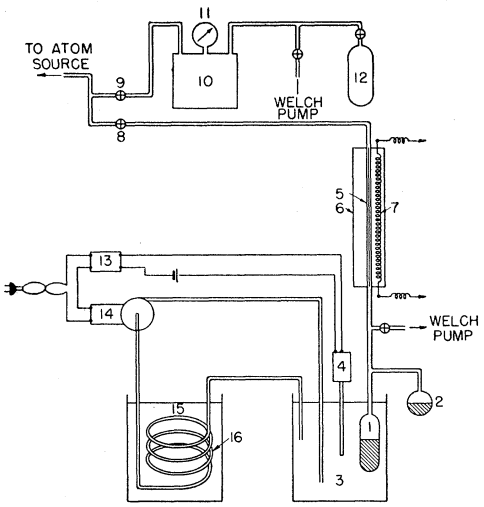

FIG. 2. Bromine handling system: (1) Br reservoir, (2) drying agent, (3) constant temperature bath, (4) thermostat, (5) capillary leak, (6) and (7) leak heating bath and heater, (8) Br shut-off valve, (9) chlorine needle valve, (10), (11), and (12) chlorine system, (13) relay, (14) motor driven pump, (15) ice bath, (16) heat exchanger coil.

matched to the transition field loop, although only the F4800 was capable of overflopping a transition.

Preliminary measurements were made with a Cardwell TS-175/U heterodyne wave meter, which was also used to prevent gross errors in making more precise measurements. Increased precision was obtained by using a General Radio 620-A heterodyne wave meter to produce a beat between the signal causing the transition and the nearest harmonic of a 120 Mc/sec, 150 Mc/sec, or 180 Mc/sec signal generated by multiplying the output of a stable 5-Mc/sec crystal, which was maintained within ± 1 cps of the 5-Mc/sec signal transmitted by WWV. The total maximum error introduced by the frequency measuring techniques is less than one part in 10^6.

III. EXPERIMENTAL PROCEDURE

A. Nuclear Spin

From observations of the Zeeman effect of the hfs interaction in weak magnetic fields, the spin I for both bromine isotopes was determined. In weak fields the frequencies corresponding to transitions for which $\Delta F=0$ may be expressed as

$$\nu=g_F\mu_0 H/h, \tag{6}$$

where

$$g_F=[(\mathbf{F}\cdot\mathbf{J})g_J+(\mathbf{F}\cdot\mathbf{I})g_I]/[f(f+1)]. \tag{7}$$

Since $g_I \ll g_J$,

$$\nu=\frac{f(f+1)+j(j+1)-i(i+1)}{2f(f+1)}g_J\mu_0 H/h. \tag{8}$$

For the case of $I=J$ the Zeeman frequencies are identical for all F levels. $\Delta F=0$ transitions at a given weak field were observed at the same frequency in both bromine isotopes and in chlorine, whence, since $g_J=4/3$ and $J=\frac{3}{2}$ for all halogens, the value of $I=\frac{3}{2}$ for both Br^{79} and Br^{81} was immediately established.

B. Determination of the HFS Intervals

In order to observe transitions which satisfy the selection rule $\Delta F=0$, $\Delta m_F=\pm 1$, a further condition must be imposed on the value of the effective magnetic moment of the atom (m_J) in the large fields ($x>7$) of the deflecting (**A**) and refocusing (**B**) magnets, depending on the relative orientation of the gradients of the two magnets. In these experiments the **A** and the **B** magnets have their gradients in the same direction, so that an atom arrives at the detector only if the sign of its effective moments is reversed by a transition in the intervening homogeneous field produced by the **C** magnet. Transitions can therefore be observed free of background due to atoms, except for that resulting from scattering. The background of molecules which have no net atomic magnetic moment, is conveniently removed by a stopping wire placed at the end of the **B** magnet. This is the "flop-in" method first used by Zacharias on K^{40}.[8] Alternatively the **A** and **B** magnets can have their gradients oppositely directed, so that all atoms reach the detector, except those whose effective magnetic moment changes sign or magnitude as a result of a transition in the **C** field. Transitions must, therefore, be observed as a small decrease of the total beam. This is the "flop-out" method.

"Flop-in," though preferable to "flop-out" when it is necessary to work with small or unsteady beams, does not allow as many transitions to be observed as can be observed with "flop-out," because of the more stringent requirements that must be satisfied by the atoms in the **A** and **B** fields. For instance, in the case

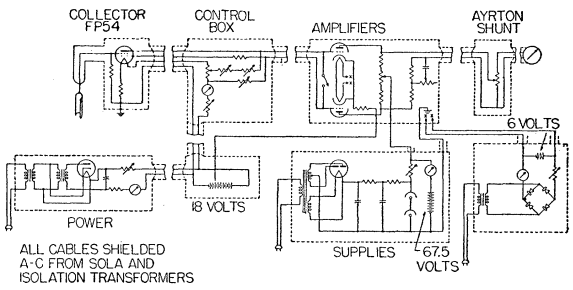

FIG. 3. Electrometer system. The total increase in sensitivity resulting from the amplifiers shown is a factor of about 100 partly due to improved matching to the galvanometer. An equalizer with an insertion loss of 10 is used to speed up the initial response of the galvanometer so that the presence of a transition may be more easily ascertained when the radio-frequency oscillator is turned on and off.

[8] J. R. Zacharias, Phys. Rev. **61**, 270 (1942).

of $I=J=\frac{3}{2}$, two transitions can be seen in "flop-in," whereas six can be seen in "flop-out."

If two independent transition wires carrying rf currents of different frequency are used, both producing oscillating magnetic fields so oriented as to induce $\Delta m_F=\pm 1$ transitions, most of the advantages of the "flop-out" method may be realized in a "flop-in" apparatus. The two independent wires for inducing $\Delta m_F=\pm 1$ transitions were placed on either side of the wire used for inducing $\Delta m_F=0$ transitions as shown schematically in Fig. 4. As an illustration, consider the transition $(F=3,\ m_F=-1\leftrightarrow F=3,\ m_F=-2)$ as indicated in Fig. 1(a).

If an rf current is passed through the wire labelled f_i (see Fig. 4) at a frequency $\nu_1=E(3,\ -1\leftrightarrow 3,\ -2)/h$, "flop-in" will be observed as an increase in the negative ion current from the detector. Now when a current of frequency $\nu_2=E(3,\ -2\leftrightarrow 3,\ -3)/h$ is passed through the wire labelled f_0 with the current of frequency $E(3,\ -1\leftrightarrow 3,\ -2)/h$ still present in the wire f_i, those atoms which were in the state $(F=3,\ m_F=-1,\ m_J=\frac{1}{2})$ in the **A** magnet will be in the state $(3,\ -1,\ \frac{1}{2})$ in the **B** magnet and will consequently not be refocused at the detector.[9] The transitions induced by the current in f_0 will, therefore, be observed as a decrease of one half in the amount of "flop-in" initially observed. Reference to Fig. 1(a) shows that besides the two normal "flop-in" transitions, three additional "flop-out" transitions (shown in dashed lines) may be observed by this method. It is worth mentioning that the "flop-out on flop-in" method as described above corresponds selectively to observing either induced emission or absorption (but not to both) depending on the sign of the nuclear moment and the particular transition involved; further discussion of this point will be given in Sec. III-C.

These "flop-out on flop-in" techniques are expected to be useful in investigating radioactive nuclides such as Br^{80} because they combine the high discrimination against background of normal "flop-in" techniques with some of the flexibility of "flop-out" techniques. Using a mass spectrometer as well, should compensate for the isotopic dilution characteristic of many such experiments.

The "flop-out on flop-in" method is particularly valuable in the example given above since a relatively precise value of the zero field interval $F=3\leftrightarrow F=2$,[10] Δ, may be obtained by observing the frequency of the transition as a function of **C** field and using the exact

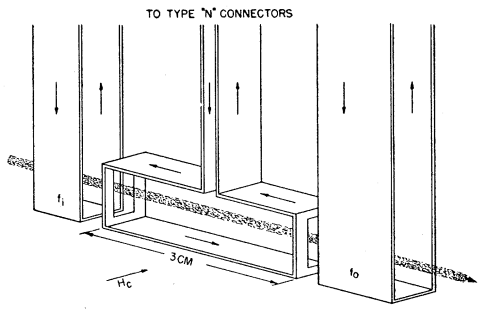

FIG. 4. Radio-frequency transition wire system. The direction of the steady homogeneous field of the **C** magnet is indicated by H_c, and is perpendicular to the direction of the beam. The wires f_i and f_0 generate oscillating field components in a direction perpendicular to that of H_c, whereas the middle wire generates components parallel to H_c.

expression:

$$\nu_2(3,\ -2\leftrightarrow 3,\ -3)=\tfrac{1}{2}(1-\gamma)ax-\tfrac{1}{2}\Delta+[\Delta^2+(1+\gamma)^2a^2x^2]^{\frac{1}{2}},\quad (9)$$

where $\gamma=-(g_Im)/(g_JM)$, $x=(\mu_0 g_J H)/ha$, and the other symbols have their usual meanings.

The field is measured by observing the same transition in Cl^{35}, for which the interaction constants are known, and by assuming that $g_J(\mathrm{Cl})=g_J(\mathrm{Br})$.

The relative complexity of the expressions for the various $\Delta m=\pm 1$ lines can be seen from the form of the solution of the general secular equation. The secular equation, for $I=J=\frac{3}{2}$, factors into one-fourth-order determinant, and pairs of first-order, second-order and third-order determinants corresponding to the low field quantum number $m_F=0,\pm 3,\pm 2,\pm 1$, respectively. Only in the case of the transition $(3,\ -2\leftrightarrow 3,\ -3)$ can the transition frequency be expressed as a function of H and the zero field interval Δ in simple closed form so that a relatively accurate value of Δ can be calculated from the experimentally observed quantities. It is, therefore, not necessary to solve numerically the rather involved secular determinants as in the method of DFZZ. Evidently the present method can be used in general to simplify the prediction of the zero field intervals from low-frequency data, since a "flop-out" transition expressible by an equation of lower order than the equation for the normal "flop-in" transition can always be found.

The zero field interval $(F=2\leftrightarrow F=1)$, Γ, was estimated by observing the quadratic departure from the linear Zeeman effect of the three lines:

$$\nu_1(3,\ -1\leftrightarrow 3,\ -2)=\tfrac{1}{2}(1-\gamma)ax+[(3/20)/\Delta](1+\gamma)^2a^2x^2,\quad (10)$$

$$\nu_3(2,\ 0\leftrightarrow 2,\ -1)=\tfrac{1}{2}(1-\gamma)ax+[(1/5)/\Gamma-(1/20)/\Delta](1+\gamma)^2a^2x^2,\quad (11)$$

$$\nu_4(2,\ -1\leftrightarrow 2,\ -2)=\tfrac{1}{2}(1-\gamma)ax+[(3/5)/\Gamma-(3/20)/\Delta](1+\gamma)^2a^2x^2,\quad (12)$$

[9] The quantum number m_I is not specified since (a) it is uniquely determined by $m_I=m_F-m_J$; (b) the physically important quantum number is m_J, which determines the path of the atom in the high fields of the **A** and **B** magnets.

[10] The following notation will be used throughout for the zero field intervals:

hfs intervals	Symbol
$(F=3\leftrightarrow F=2)$	Δ
$(F=2\leftrightarrow F=1)$	Γ
$(F=1\leftrightarrow F=0)$	Ω.

TABLE I. Frequencies of various $\Delta F=\pm 1$ transitions observed in a weak magnetic field.

	Br^{79}			Br^{81}		
Transitions used	$(1, 0\leftrightarrow 0, 0)$	$(2, 0\leftrightarrow 1, 0)$ $(2, 1\leftrightarrow 1, 1)$	$(3, -1\leftrightarrow 2, -1)$	$(1, 0\leftrightarrow 0, 0)$	$(2, 0\leftrightarrow 1, 0)$ $(2, 1\leftrightarrow 1, 1)$	$(3-1\leftrightarrow 2-1)$
Observed frequency (Mc/sec) corrected to $x=0$	1269.702	2154.499	2269.552	1275.271	2229.056	2539.794
Theoretical splitting	$a-b+100.8c$	$2a-b-50.4c$	$3a+b+14.4c$	$a-b+100.8c$	$2a-b-50.4c$	$3a+b+14.4c$

where the expressions represent the frequencies expanded to second order in the field parameter x, and the value of Δ obtained from Eq. (1) is used. A knowledge of the approximate values of the zero-field intervals facilitated finding the three corresponding $\Delta F=\pm 1$, $\Delta m_F=0$ transitions in each isotope. A transition wire which produced an oscillating magnetic field principally in the direction of the **C** field (see Fig. 4) was used. Since no "flop-in" transition exists between the levels $F=1$ and $F=0$, the ingenious method of DFZZ was used, with slight modification. The "flop-in" transition $(F=2,\ m_F=1,\ m_J=\frac{1}{2}\leftrightarrow F=1,\ m_F=0,\ m_J=-\frac{1}{2})$ was induced by loop f_i, and the transition $(1, 0, \frac{1}{2}, \rightarrow 0, 0, \frac{3}{2})$ induced by the z-component wire was observed as "flop-out" in a fashion completely analogous to the method described above in its application to $\Delta F=0$ transitions.

The $\Delta F=\pm 1$, $\Delta m=0$ transitions were observed in very weak fields (approximately 1 gauss) and, since these transitions are field independent to first order, the natural line widths were obtained. Correction of each of the measured frequencies to zero field was made as described in DFZZ. The results are summarized in Table I. A full resonance curve was obtained for each transition as a precautionary measure in interpreting the results. Representative "flop-in" and "flop-out on flop-in" curves are shown in Fig. 5. Several of the narrow field-independent transitions exhibited structure similar to that observed and interpreted by Hughes, Tucker, Rhoderick, and Weinreich in their work with atomic helium.[11] It was found possible to enhance or eliminate the structure observed in the bromine transitions by changing the strength of the oscillating field and the range of velocities of the observed atoms, but in all cases the center of the structure was unshifted in frequency as predicted by the Majorana theory.[11,12]

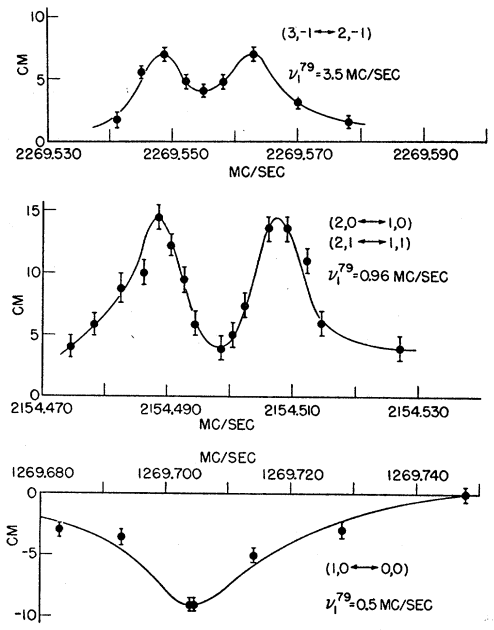

FIG. 5. Representative resonance curves in Br^{79}. These correspond to $\Delta F=\pm 1$ transitions as labeled. The field at which each transition was observed was calibrated by measuring a $\Delta F=0$ transition at the same time. The bottom curve was observed as "flop-out" on the (2,1↔1,0) "flop-in" transition. A galvanometer deflection of 1 cm corresponds to a collector ion current of 1.5 $\times 10^{-16}$ ampere.

C. Determination of the Sign of the Nuclear Magnetic Moments

The hfs energy level diagrams for $I=J=\frac{3}{2}$ appear in Figs. 1(a) and 1(b), respectively, for positive and negative nuclear moments. The important differences between these two diagrams are that the order of the hfs level is inverted with respect to energy and that all magnetic quantum numbers are reversed in sign. For purposes of this discussion the interaction of the nuclear magnetic moment with the external field can be neglected since $\mu_I \ll \mu_0$. Since the path followed by atoms in a given state in the strong fields of the **A** and **B** magnets depends on the direction of the gradient of the magnetic fields and the sign of the effective moment (e.g., atoms with $m_J>0$ are deflected toward weaker fields), atoms in a given m_J state may be removed from the observed beam by inserting an obstacle, at the exit of the **B** magnet, that extends outward from the center of the beam as indicated in Fig. 6.

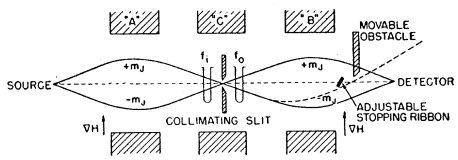

FIG. 6. Atom paths drawn exaggerated to illustrate how the sign of a nuclear moment was determined as described in Sec. IIIC of the text.

[11] Hughes, Tucker, Rhoderick, and Weinreich, Phys. Rev. **91**, 828 (1953).

[12] E. Majorana, Nuovo cimento **9**, 43 (1932).

The method used here to determine the sign of the nuclear moment requires only the observation of relatively low-frequency $\Delta F=0$ transitions. Consider the one observable $\Delta F=0$, "flop-in" transition in the $F=3$ level. Depending on the sign of the nuclear magnetic moment, this transition is $\{3, -1, \frac{1}{2}\leftrightarrow 3, -2, -\frac{1}{2}\}$ for positive moment, and $\{3, 1, -\frac{1}{2}, \leftrightarrow 3, 2, \frac{1}{2}\}$ for negative moment, as can be seen from Figs. 1(a) and 1(b). There are, in addition, the corresponding transitions $\{3, -2, -\frac{1}{2}, 3, -3, -\frac{3}{2}\}$, $(\mu_I>0)$, or $\{3, 2, \frac{1}{2}\leftrightarrow 3, 3, \frac{3}{2}\}$, $(\mu_I<0)$, observable as "flop-out on flop-in" as previously described.

The $\Delta F=0$, "flop-in" transition was reduced to half its maximum value by inserting the obstacle from the low **B** field side of the beam, so that no atoms with $m_J>0$ in the **B** magnet reach the detector. Now to determine whether the remaining atoms were in the state $(3, -2, -\frac{1}{2})$ or in the state $(3, 1, -\frac{1}{2})$, the frequency corresponding to the $\Delta F=0$, "flop-out" transition was introduced in the wire, whereupon the remaining "flop-in" disappeared. The atoms were therefore in the state $(3, -2, -\frac{1}{2})$, and it may be concluded that diagram 1(a) applies and the nuclear magnetic moment is positive. With the obstacle on the high **B** field side of the center of the beam, no "flop-out" was observed. Had the nuclear magnetic moment been negative, the same effects would have been observed with the obstacle on the high **B** field side of the beam.

IV. RESULTS

A. The Interaction Constants

From the measured intervals given in Table I the following values for the interaction constants may be calculated:

$$a_{\frac{3}{2}}^{79}=884.810\pm0.003 \text{ Mc/sec},$$
$$b_{\frac{3}{2}}^{74}=384.878\pm0.008 \text{ Mc/sec},$$
$$c_{\frac{3}{2}}^{79}\leqq 0.0001 \text{ Mc/sec},$$
$$a_{\frac{3}{2}}^{81}=953.770\pm0.003 \text{ Mc/sec},$$
$$b_{\frac{3}{2}}^{81}=321.516\pm0.008 \text{ Mc/sec},$$
$$c_{\frac{3}{2}}^{81}\leqq 0.0001 \text{ Mc/sec}.$$

Thus, as in all previous atomic beam hfs experiments, the observed nuclear-electron interaction may be described, to a high degree of accuracy, in terms of only two electromagnetic interaction parameters. This is not unexpected, since a simple theory predicts that the nuclear magnetic octupole moment should be, in order of magnitude $(a_0/R_K)^2\approx 10^8$ as small as the nuclear magnetic dipole moment.

B. The Ratio of the Magnetic Dipole Interaction Constants

From the data above the ratio $a_{\frac{3}{2}}^{81}/a_{\frac{3}{2}}^{79}$ may be calculated. This ratio can be compared with the ratios, given in Table II, of the nuclear magnetic dipole moments as measured by nuclear resonance techniques. Two of the values listed agree within the experimental uncertainties with the value reported here. The result of Sheriff and Williams, however, disagrees by 18 parts in 10^5. Since the electronic wave function density for a $P_{\frac{3}{2}}$ electron at the position of the nucleus is extremely small and the two bromine isotopes have similar g_I values, the magnetic hyperfine structure anomaly to be expected in this case is far smaller than the above discrepancy (certainly less than 1 part in 10^6).[13] Professor Williams has pointed out[14] that if an error in sideband identification in the measurements of Sheriff and Williams had been made, the necessary correction would bring their result into agreement with those listed in Table II. Otherwise, it is difficult to reconcile these results.

TABLE II. Ratios of the nuclear magnetic dipole interactions.

Method	Ratio
Nuclear resonance in NaBr, LiBr solution[a]	$\mu^{81}/\mu^{79}=1.0778\pm0.0003$
Nuclear resonance in NaBr solution[b]	$\mu^{81}/\mu^{79}=1.07775\pm0.00005$
Nuclear resonance in NaBr solution[c]	$\mu^{81}/\mu^{79}=1.07796\pm0.00002$
Atomic beam magnetic resonance[d]	$a_{\frac{3}{2}}^{81}/a_{\frac{3}{2}}^{79}=1.077938\pm0.000020$

[a] R. V. Pound, Phys. Rev. **72**, 1273 (1947).
[b] R. E. Sheriff and D. Williams, Phys. Rev. **82**, 651 (1951).
[c] H. E. Walchli, Oak Ridge National Laboratory Report ORNL-1469, 1953 (unpublished), pp. 36 and 37. See reference WL 52–5 to unpublished work of H. E. Walchli.
[d] This paper.

C. The Ratio of the Electric Quadrupole Interaction Constants

In a similar fashion, the ratio $b_{\frac{3}{2}}^{79}/b_{\frac{3}{2}}^{81}$ may be compared with the ratio of the electric quadrupole moments as observed by pure nuclear quadrupole resonance in solids. (See Table III.) Since both methods are capable of great precision and are free from molecular binding effects, the agreement of these results, unique in the comparison of the quadrupole interaction ratios for two isotopes, indicates that the hfs interaction is well understood.

TABLE III. Ratios of the nuclear electric quadrupole interactions.

Method	Ratio
Quadrupole resonance in Br_2[a]	$\nu^{79}/\nu^{81}=1.1968\pm0.0002$
Molecular beams (KBr)[b]	$Q^{79}/Q^{81}=1.1977\pm0.0008$
Microwave spectroscopy[c] (average of 10 values taken with 6 compounds)	$Q^{79}/Q^{81}=1.19707\pm0.00003$
Atomic beam magnetic resonance[d]	$b^{79}/b^{81}=1.19707\pm0.00003$

[a] H. G. Dehmelt, Z. Physik **130**, 480 (1951).
[b] Fabricand, Carlson, Lee, and Rabi, Phys. Rev. **91**, 1403 (1953).
[c] A. L. Schawlow (private communication).
[d] This paper.

[13] A. Bohr and V. F. Weisskopf, Phys. Rev. **77**, 94 (1950).
[14] D. R. Williams (private communication).

TABLE IV. Nuclear electric quadrupole moments (in units of 10^{-24} cm^2).

Method	Values: Uncorrected Br^{79}	Uncorrected Br^{81}	With Sternheimer correction[d] Br^{79}	With Sternheimer correction[d] Br^{81}
Microwave spectroscopy[a]	0.31	0.26	0.32	0.27
Quadrupole resonance in Br_2[b]	0.30	0.25	0.31	0.26
Atomic beam magnetic resonance[c]				
(1) calculated from Eq. (13)	0.322±0.019	0.269±0.016	0.335±0.020	0.280±0.017
(2) calculated from Eqs. (5) and (14)	0.296±0.018	0.247±0.015	0.308±0.018	0.257±0.016

[a] Gordy, Simmons, and Smith, Phys. Rev. **74**, 243 (1948).
[b] H. G. Dehmelt, Z. Physik **130**, 480 (1951).
[c] This paper.
[d] The results of Sternheimer (see reference 16) indicate that the correction should be applied to the values of Q obtained by these methods.

D. The Nuclear Electric Quadrupole Moments

From the values of $b_{\frac{1}{2}}$, $a_{\frac{1}{2}}$, μ, and I, Q may be calculated using the method of DFZZ, where it is assumed only that the angular and radial parts of the wave functions for the electron are separable, and that $\langle r^{-3}\rangle$ is the same in the electric quadrupole and magnetic dipole interactions. (Because the excited atomic states of the halogens are different from those of the gallium family it is possible, as it was not in the case of gallium,[15] to neglect the effects of configuration interaction in obtaining Q to the desired accuracy, which is limited by the relativistic corrections which are known to approximately one percent). The magnitude of Q must however be corrected for the polarization of the inner electron core by the nuclear quadrupole moment. This correction has been evaluated by Sternheimer in several articles.[16] The result may be expressed as

$$Q=-(8/3)(\mu_0/e)^2(m/M)(\mu/I)(\mathfrak{F}/R)(b/a)C, \quad (13)$$

where $C=1.040$ is the Sternheimer correction and $\mathfrak{F}/R=0.99$ is a relativistic correction.[3]

It should be noted that the sign of the nuclear magnetic moment previously confirmed to be positive (see Sec. III C) enters in determining the sign of Q.

An independent method for calculating the quadrupole moments utilizes the fact that $\langle r^{-3}\rangle$ may be evaluated from the fine structure separation δ by using the relation

$$\delta=(\mu_0^2/hc)Z_i(2l+1)H\langle r^{-3}\rangle. \quad (14)$$

For the effective value of the nuclear charge Z_i, the result of recent work of Smith and Barnes[17] for p electrons is used to write $Z_i=Z-n$, n being the radial quantum number. H here is a relativistic correction factor tabulated by Casimir.[3] Using the above relation and Eq. (5), the Q's have been calculated and are listed in Table IV for comparison with the values obtained by the aforementioned method. The agreement is good and indicates the apparent validity of quadrupole moment calculation by either method in the case of the halogens. Considering the accuracy of the experiments and the detail of the theory, we believe that ±6 percent represents an upper limit for the uncertainty in these moments.

TABLE V. The anomalous spin gyromagnetic ratio of the $P_{2/3}$ electron in bromine.

$$g_J(\mathrm{Br})/g_J(\mathrm{Cl})=0.9997\pm0.0004$$
$$g_S(\mathrm{Br})=2(1.0008\pm0.0005)$$

E. The Anomalous Spin Gyromagnetic Ratio of the $P_{3/2}$ Electron in Bromine

The ratio $g_J(\mathrm{Br})/g_J(\mathrm{Cl})$ may be determined from measurements of the transition $(3,-2\leftrightarrow 3,-3)$ in each element in the same magnetic field, once the interaction constants are known. As can be seen in Table V, the g_J's are similar in bromine and chlorine, the accuracy of the determinations being severely limited by the broadening of the transitions caused by the inhomogeneity of the C-field. From a previous investigation of the g_J's in the $J=\frac{3}{2}$ and $J=\frac{1}{2}$ states in chlorine,[18] and setting $g_L=1$, the value of g_S listed in Table V for the $P_{\frac{3}{2}}$ electron in bromine was obtained.

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[15] G. F. Koster, M.I.T. Doctoral thesis (1951); Phys. Rev. **86**, 148 (1952).
[16] R. Sternheimer, Phys. Rev. **84**, 244 (1951); **86**, 316 (1952).
[17] W. V. Smith and R. G. Barnes, Phys. Rev. **91**, 457 (1953).
[18] J. G. King and V. Jaccarino, Phys. Rev. **87**, 228 (1952), and Quarterly Progress Report, Research Laboratory of Electronics, M.I.T. April 15, 1952 (unpublished), p. 33.