MASTER

UNIVERSITY OF
CALIFORNIA
Ernest O. Lawrence
Radiation
Laboratory

SPINS, MOMENTS, AND HYPERFINE
STRUCTURES OF SOME BROMINE ISOTOPES

BERKELEY, CALIFORNIA

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

UCRL-8730
Physics and Mathematics

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

SPINS, MOMENTS, AND HYPERFINE STRUCTURES OF
SOME BROMINE ISOTOPES

Thomas Myer Green, III
(Thesis)

June 1959

Printed for the U. S. Atomic Energy Commission

Printed in USA. Price \$2.50. Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

SPINS, MOMENTS, AND HYPERFINE STRUCTURES OF
SOME BROMINE ISOTOPES

Contents

Abstract	3
I. Introduction	4
II. Theory	5
III. Experimental Procedure	24
IV. Apparatus and Equipment	30
V. Discussion of Individual Isotopes:	
Bromine 82, 80m, 80, 77, 76	50
VI. Summary of Data	95
Acknowledgments	100

SPINS, MOMENTS, AND HYPERFINE STRUCTURES OF SOME BROMINE ISOTOPES

Thomas Myer Green, III

Lawrence Radiation Laboratory and Department of Physics
University of California
Berkeley, California

June 1959

ABSTRACT

This paper presents the results of measurements on a series of bromine isotopes by the method of atomic beams. Experimental techniques, theory, and apparatus employed are described, together with a discussion of the results.

Original results presented here are:

Br^{82} ($t_{1/2} = 35.5 \text{ hr}$) $I = 5$ $a = 205.04 \pm 0.05$ $b = -870.66 \pm 0.90 \text{ Mc}$

$\Delta\nu_{12} = 766.82 \pm 0.60$; $\Delta\nu_{23} = 1287.32 \pm 0.43 \text{ Mc}$

$\Delta\nu_{34} = 1488.6 \pm 1.1 \text{ (calc.)}$

$\mu(\text{calc.}) = 1.624(5)$ $Q(\text{calc.}) = 0.76(3)$

$\text{Br}^{80\text{m}}$ (4.5 hr) $I = 5$

Br^{80} (18m) $I = 1$

Br^{77} (57 hr) $I = 3/2$

Br^{76} (17 hr) $I = 1$ $a = 345.417 \pm 0.07$; $b = 314.329 \pm 0.11 \text{ Mc}$

$\Delta\nu_{12} = 1256.45 \pm 0.07$; $\Delta\nu_{23} = -189.115 \pm 0.07 \text{ Mc}$

$\mu(\text{calc.}) = \pm 0.5480(2)$ $Q(\text{calc.}) = \mp 0.27(2)$

I. INTRODUCTION

This paper describes measurements of the spins, moments, and hyperfine structures of a series of bromine isotopes by the method of atomic beams. This investigation is being continued and is part of a more general program of investigation of the radioactive halogens by the atomic-beam technique.

The measured values of the spins, moments, and hyperfine structures have been published and are available in the literature (Refs. 1 through 9). Other results obtained in the course of the work, but which are not discussed here, can be found in Refs. 10 through 13. The purpose of the present report is to gather the bromine results together and relate them to current nuclear-structure theories, as well as to describe the experimental work in detail.

The main body of the paper is divided into sections describing theory, experimental procedure, and equipment used. These are followed by discussions of the results on individual isotopes and a concluding summary of the data and their relationship to nuclear theory.

II. THEORY

The theory of hyperfine structure deals with the interaction of nuclear multipole moments with fields acting on the nucleus. Basic atomic structure of a heavy positively charged nucleus surrounded by a relatively extended collection of electrons was established in the early 1900's. Bohr's theory of electronic quantum states appeared in 1913 and with subsequent improvements explained spectral line frequencies with high accuracy. Fine structure in spectral lines was explained by assigning a magnetic moment to the orbital electrons by Uhlenbeck and Goudsmit in 1925.¹³

Even finer structure in spectral lines was known and had been observed before 1900. This hyperfine structure was attributed to a nuclear magnetic moment in the same decade as the discovery of the electron moment¹⁴ and became well established by the work of Back and Goudsmit in 1927-8.¹⁵

Although the present work is discussed in terms of these historic angular momenta, these angular momenta are used as tools for an investigation of a totally different kind. The measurements made here are undertaken to contribute to a knowledge of the internal structure of the nucleus. Because the bromine nucleus has only about 80 nucleons, interpretation will be in terms of discrete particle orbits as visualized by the "shell model."¹⁶

From a different point of view, very precise measurements of transition frequencies indicate deviations from the gross hyperfine-structure theory and are interpreted in terms of a finite nuclear size and distribution of moment over the nucleus. A mention of the spirit of these calculations seems desirable, even though they are not utilized to any great extent in this paper. Work by Sternheimer has dealt with corrections to the measured quadrupole moments of nuclei.¹⁷ The correction results from a polarization of the closed electron shells by the electrons in incomplete, and hence non-spherically symmetric, shells. This may be of either sign and amounts to a few percent.

When the nucleus is not assumed to be a point, a variation in the electron wave functions over the nuclear dimensions must be considered and will give a changed interaction energy. Calculations based on various distributions of electric charge over the nuclear volume were made by Breit as long ago as 1932¹⁸ and later by Breit and Rosenthal.¹⁹ Crawford and Schawlow have considered the effect of a number of different charge distributions.²⁰ Distribution of magnetic moment over the nuclear size has been considered by Bohr and Weisskopf.²¹

Recent very high precision measurements have inspired a general re-evaluation of the theory of hyperfine structure by Schwartz²² in which he considers second-order effects causing coupling between different multipole orders and configuration interactions.

A number of experimental results concerning the nucleus, which will be here taken as postulates, must be stated. They are:

a. The nuclear charge, Ze , and mass is confined to a region of the order of 10^{-12} cm. Measurements of nuclear size give results consistent with $R = (1.1-1.5)A^{1/3} 10^{-13}$ cm, where the first factor varies somewhat with the method of measurement.

b. The nucleus is composed of neutrons and protons. This implies that the interaction of the particles with one another is much less than their rest energy so that it makes sense to talk about individual nucleons. Their average binding energy is, in fact, about 8 Mev as compared with approximately 931 Mev of rest mass.

c. The nucleus possesses an angular momentum, I , which has all the properties associated with a quantum-mechanical angular-momentum vector.

d. If A is odd/even the nucleus obeys Fermi-Dirac/Bose-Einstein statistics. This result is established by the observation of alternating intensities of lines in the band spectra of homonuclear molecules.²³

- e. I is half-integral or integral as A is odd or even.
- f. The nucleus can exhibit electric and magnetic moments.

If an atom has no nuclear angular momentum and no electronic angular momentum, it may be described in terms of a charged nucleus and electrons moving in the central Coulomb field. If there is an odd number of neutrons, protons, or both, the nucleus will in general possess a quantized angular momentum with the properties listed above.

In addition, if the electronic angular momentum is non-zero, the atom will possess hyperfine structure as a result of the electron-nucleus interaction. These hyperfine levels are the eigenvalues of $\vec{F}$ where $\vec{F} = \vec{I} + \vec{J}$ and $|\vec{I} - \vec{J}| \leq \vec{F} \leq |\vec{I} + \vec{J}|$. The eigenvalues are separated by an energy much less than kT at room temperature so the level populations are determined solely by quantum-mechanical considerations.

An atom in field-free space is independent of spatial coordinates and orientation. Therefore, the Hamiltonian describing the system must be a scalar and independent of external coordinates. Both the nuclear and electronic wave functions can be written in terms of spherical harmonics in which the angular dependence is a function of the angular momentum involved; thus so we are led to expect a Hamiltonian involving the scalar product $\vec{I} \cdot \vec{J}$, e. g.

$$\mathcal{H} = \sum_{i=0}^x a_i (\vec{I} \cdot \vec{J})^i \quad (1)$$

where $x \neq \infty$ as would be expected classically, but is limited to $x = 2I$ (or $2J$ if J is less than I). One must consider both electrostatic and magnetic interactions between the nucleus and electrons, but parity considerations require terms of a given symmetry to contain only one of the two interactions. This can be seen in the electric case as follows: Assuming that, a. all nuclear electrical effects arise from electric charge, and the b. there is no degeneracy of nuclear states with different parity, we have (by writing the electric multipole moment of the nucleus classically):

$$\mathcal{H}_j^{(i)} = \int \rho_n x_j^i dV,$$

which in the quantum mechanical case becomes a quantity:

$$\langle \psi | \chi^i | \psi \rangle = \langle \psi | \chi^i \psi \rangle.$$

If i is odd, the whole integrand will be odd and hence equal to zero. Therefore, only electrical monopole, quadrupole, etc. terms are allowed.

To obtain the equivalent expressions for the magnetic nuclear moments, ρ_n is replaced by $-\nabla_n \cdot \vec{m}_n$ where, $\vec{j} = c \nabla \times \vec{m}_n$. This is shown, for example, on pp. 68-70 of Ref. (24). Since $-\nabla_n \cdot \vec{m}_n$ is a pseudoscalar, the parity considerations are reversed from those of the electric case, and only magnetic dipole, octupole, etc., terms appear.

This gives the interesting result that the electric and magnetic terms alternate--beginning, of course, with the electric monopole.

Since it can be shown that the transition from the electric to magnetic interaction can be made by the substitution²⁴

$$\rho_e \rightarrow \nabla_e \cdot \vec{m}_e \quad \rho_m \rightarrow -\nabla_n \cdot \vec{m}_n, \quad (2)$$

the physical situation can be made evident by working with the electric case, retaining the moments of both parities and making the substitution for the ones which should be magnetic.

The general electrostatic interaction, including a finite nuclear size, may be written:

$$\mathcal{V} = \int_{\tau_e} \int_{\tau_n} \frac{\rho_e \rho_n d\tau_e d\tau_n}{r}, \quad (3)$$

and by writing

$$\frac{1}{r} = \frac{1}{\sqrt{r_e^2 + r_n^2 - 2r_e r_n \cos \theta}} = \frac{1}{r_e} \sum_{k=0}^{\infty} \left(\frac{r_n}{r_e} \right)^k P_k(\cos \theta)$$

from the well known expansion, written here for $r_e > r_n$ one may separate terms in $\mathcal{H}$:

$$\mathcal{H}_e = \sum_k \mathcal{H}_{ek} ; \mathcal{H}_{ek} \equiv \int_{\tau_e} \int_{\tau_n} \frac{\rho_e^{(e)} \rho_n}{r_e} \left(\frac{r_n}{r_e} \right)^k P_k(\cos \theta) d\tau_e d\tau_n . \quad (4)$$

The expressions here are restricted to charges external to the nucleus, indicated by $\rho_e^{(e)}$. This integral is difficult to use as θ involves the coordinates of both electrons and nucleus and therefore cannot be separated conveniently. Separation may be accomplished by use of spherical-harmonic addition theorem and the analysis carried through for the general case in terms of tensor products:

$$\mathcal{H}_{ek} = Q^{(k)} \cdot F^{(k)} \quad (5)$$

as may be found in Refs. 22 or 24.

While this analysis may be carried through for general field configurations, a more illuminating picture, physically, results from another approach, making use of the symmetry of the fields involved. ²⁵

Let primed axes be nuclear coordinates and unprimed be electron coordinates. Both nuclear and atomic fields are axially symmetric so the interaction energy can only depend on the angle between their axes. Assume, without loss of generality, that the y, y' axes coincide (Fig. 1). Then write the energy of interaction:

$$W = \int \rho_n \phi dV .$$

Take $\phi = \phi(x', y', z')$ and expand to:

$$\begin{aligned} W = & \phi(0) \int \rho_n dV' + \\ & + \frac{\partial \phi(0)}{\partial x'} \int \rho_n x' dV' + \dots \\ & + \frac{1}{2} \left\{ \frac{\partial^2 \phi(0)}{\partial x'^2} \int x'^2 \rho_n dV' + \frac{\partial^2 \phi(0)}{\partial x' \partial y'} \int x' y' \rho_n dV' + \dots \right. \end{aligned} \quad (6)$$

The first term is not angle-dependent and so contributes only a constant term. Terms on line 2 will actually equal zero because of the argument presented earlier. But making here the substitutions $\rho_n \rightarrow -\nabla_n \cdot \vec{m}_n$ and $\phi \rightarrow \phi_{\text{mag}}$ gives:

$$W_d = - \frac{\partial \phi_m}{\partial x'} \mu_x - \frac{\partial \phi_m}{\partial y'} \mu_y - \frac{\partial \phi_m}{\partial z'} \mu_z = - \nabla \phi_m \cdot \vec{\mu} = - \vec{\mu} \cdot \vec{H}(0)$$

for the magnetic dipole term.

For the terms on the third line of Eq. (6), one may write the integrals as the components of a tensor:

$$eQ_{x'x'} = \int \rho_n x'^2 dV' \quad \text{etc.}$$

Taking the nuclear symmetry axis as z' , the tensor defined here will have no off-diagonal terms and $Q_{x'x'} = Q_{y'y'}$. Therefore in line 3 of Eq. (6) all the terms ϕ_{ij} with $i \neq j$ are multiplied by zero and only diagonal terms appear in the product.

For non-s electrons we have $\nabla^2 \phi(0) = 0$ and therefore the quadrupole interaction energy becomes

$$\begin{aligned} W_Q &= \frac{1}{2} \left\{ \frac{\partial^2 \phi}{\partial x'^2} eQ_{x'x'} + \frac{\partial^2 \phi}{\partial y'^2} eQ_{y'y'} + \frac{\partial^2 \phi}{\partial z'^2} eQ_{z'z'} \right\} \\ &= \frac{e}{2} \left[Q_{x'x'} \left\{ \frac{\partial^2 \phi}{\partial x'^2} + \frac{\partial^2 \phi}{\partial y'^2} \right\} + Q_{z'z'} \frac{\partial^2 \phi}{\partial z'^2} \right] \\ &= \frac{e}{2} \left[Q_{x'x'} (\nabla^2 \phi) + Q_{z'z'} \frac{\partial^2 \phi}{\partial z'^2} - Q_{x'x'} \frac{\partial^2 \phi}{\partial z'^2} \right] \\ &= \frac{e}{2} \frac{\partial^2 \phi}{\partial z'^2} (Q_{z'z'} - Q_{x'x'}) = \frac{e}{2} \frac{\partial^2 \phi}{\partial z'^2} \int \rho_n (z'^2 - x'^2) dV'. \end{aligned}$$

MU-17593

Fig. 1. Nuclear and electron coordinate axes.

Again by symmetry, we have $r^2 = z'^2 + 2x'^2$ and

$$\frac{e}{2} \frac{\partial^2 \phi}{\partial z'^2} \int \rho_n (z'^2 - \frac{1}{2} (r^2 - z'^2)) dV = \frac{e}{4} \frac{\partial^2 \phi}{\partial z'^2} \int \rho_n (3z'^2 - r^2) dV.$$

Q is defined as

$$Q = \int \rho_n (3z'^2 - r^2) dV, \quad (8)$$

which, in the quantum-mechanical case, is taken in the state $m_I = I$, the maximum observable component. Re-express $\frac{\partial^2 \phi}{\partial z'^2}$ in terms of electron coordinates, $\frac{\partial z}{\partial z'} = \cos \theta$; $\frac{\partial x}{\partial z'} = \sin \theta$

$$\frac{\partial^2 \phi}{\partial z'^2} = \frac{\partial^2 \phi}{\partial z^2} \cos^2 \theta + \frac{\partial^2 \phi}{\partial z^2} \sin^2 \theta \quad \text{and} \quad \frac{\partial^2 \phi}{\partial z^2} = -2 \frac{\partial^2 \phi}{\partial x^2} = -2 \frac{\partial^2 \phi}{\partial y^2}.$$

$$\therefore \frac{\partial^2 \phi}{\partial z'^2} = \frac{1}{2} \frac{\partial^2 \phi}{\partial z^2} (3 \cos^2 \theta - 1) \quad \text{and}$$

$$W_Q = \frac{e}{4} Q \frac{\partial^2 \phi}{\partial z^2} \frac{1}{2} (3 \cos^2 \theta - 1). \quad (9)$$

It is perhaps worth mentioning that the quadrupole interaction in s states is not equal to zero, as is often said. It is merely unobservable by atomic beams, as the field symmetry is such that the interaction energy has no angle-dependent terms. This can be most easily seen by referring to Fig. 2. There it may be seen that the value of the wave function is smaller at the larger values of r , occupied by the distorted nucleus, than in the regions left vacant by introducing the distortion. Hence the energy shift is positive and the quadrupole shape is less tightly bound.

Writing the quantum-mechanical Hamiltonian as a series in $(I \cdot J)^n$ as was made plausible earlier--one sets, for the dipole term

$$\mathcal{H}_{m_I} = -\vec{\mu}_I \cdot \vec{H}(0) = h a \vec{I} \cdot \vec{J}, \quad (10)$$

so that the magnetic-dipole interaction constant, a , is

$$a = \frac{-\vec{\mu}_I \cdot \vec{H}(0)}{h \vec{I} \cdot \vec{J}} \quad (11)$$

MU-17592

Fig. 2. Illustration of the energy shift resulting from nuclear distortion.

For the electric quadrupole interaction²⁶ we have

$$\mathcal{H}_{E_2} = \frac{e \left\langle \frac{\partial^2 \phi}{\partial z^2} \right\rangle_{m_J=J}}{2I(2I-1)J(2J-1)} \left[3(\vec{I} \cdot \vec{J})^2 + \frac{3}{2} (\vec{I} \cdot \vec{J}) - I(I+1) J(J+1) \right] \quad (12)$$

The quadrupole-interaction constant b is defined as

$$b = \frac{e \left\langle \frac{\partial^2 \phi}{\partial z^2} \right\rangle_{m_J=J} Q}{2I(2I-1)J(2J-1)} \quad (13)$$

For the zero-field Hamiltonian we have

$$\mathcal{H} = h a \vec{I} \cdot \vec{J} + h b Q_{op} \quad (14)$$

In the presence of an external magnetic field this becomes

$$\mathcal{H} = h a \vec{I} \cdot \vec{J} + h b Q_{op} - g_J \mu_0 \vec{J} \cdot \vec{H} - g_I \mu_0 \vec{I} \cdot \vec{H}, \quad (15)$$

while in a small field

$$\mathcal{H} = h a \vec{I} \cdot \vec{J} + h b Q_{op} - g_F \mu_0 \vec{F} \cdot \vec{H} \quad (16)$$

is an approximate form.

The use of J in the quantum-mechanical description of the system implies that J is a constant of the motion and therefore that the mixture of states of other J is negligibly small. For bromine, the first excited level is $^2P_{1/2}$, lying 3685 cm^{-1} above the $^2P_{3/2}$ ground state. This corresponds to about $11 \times 10^7 \text{ Mc}$, while the maximum hyperfine separation measured in the course of the work has been less than 3000 Mc , giving a ratio greater than 36×10^3 to 1. Measurements of g_J on stable bromine show deviations from the theoretical value for Russell-Saunders coupling that are negligible for this work.²⁷

From a and b, the nuclear magnetic-dipole and electric-quadrupole moments must be calculated. When the electronic configuration of the halogens, closed shell minus one, and our definition of the moments as $+g_I \bar{I}$ etc., are taken into account, the desired expressions are^{28, 29}

$$a = \frac{\mu_0^2}{h} g_I \mathcal{F} \frac{2L(L+1)}{J(J+1)} \left\langle \frac{1}{r^3} \right\rangle, \quad (17)$$

$$b = \frac{-e^2}{h} Q \mathcal{R} \frac{2L}{2L+3} \left\langle \frac{1}{r^3} \right\rangle, \quad (18)$$

$\mathcal{F}$ and $\mathcal{R}$ are relativistic correction terms tabulated in Ref. 30 and are, for bromine: $\mathcal{F} = 1.0261$, $\mathcal{R} = 1.0535$. Each of the equations (17) and (18) involves $\left\langle \frac{1}{r^3} \right\rangle$, which is sensitive to the value of the wave functions chosen. Values of $\left\langle \frac{1}{r^3} \right\rangle$ for these calculations have been obtained from "Electric Field Gradients of Atomic p Electrons" by Barnes and Smith.³¹

From Eqs. (17) and (18), with $L = 1$, and $J = 3/2$, we have

$$Q = - \frac{8}{3} \left(\frac{\mu_0}{e} \right)^2 \left(\frac{m}{M} \right) \left(\frac{\mu}{I} \right) \left(\frac{\mathcal{F}}{\mathcal{R}} \right) \frac{b}{a} C, \quad (19)$$

where C is the Sternheimer correction for the polarization of the closed electron shells by the electrons in incompletely filled shells.¹⁷

A value for $\left\langle \frac{1}{r^3} \right\rangle$ can be obtained from knowledge of the fine-structure separation δ and Formula (17):

$$\delta = \frac{\mu_0^2}{r C} Z_i (2L+1) H \left\langle \frac{1}{r^3} \right\rangle, \quad (20)$$

where H is a relativistic correction factor tabulated in Ref. 30 and Z_i is the effective Z acting on a valence electron. Barnes and Smith have shown that $Z_i = Z - n$, where n is the radial quantum number, is a reasonable choice for Z_i . For bromine $n = 4$, giving $Z_i = 31$.

Measurements of a and b have been made on the stable bromine isotopes with assigned errors of 3 and 8 kc for the a 's and b 's respectively.²⁷ By writing Eqs. (17) and (18) for two isotopes and dividing, one obtains

$$\frac{a}{a'} = \frac{g_I}{g_{I'}} = \frac{\mu_I}{\mu_{I'}} \frac{I'}{I} \quad (21)$$

and

$$\frac{b}{b'} = \frac{Q}{Q'} \quad (22)$$

Comparisons of the various measured values can then be made in order to verify the consistency of the measurements with one another. The value of a/a' is subject to the hyperfine-structure anomaly, defined by

$$\frac{\Delta\nu_{\text{obs}}}{\Delta\nu_{\text{pt}}} = 1 + \epsilon, \quad (23)$$

but this is expected to be small for $P_{3/2}$ states because of the smallness of the electron wave functions at the nucleus. It is certainly much less than the assigned errors in this work and will be ignored. Corrections to quadrupole moments are less well understood, and hence uncorrected quadrupole moments will be explicitly exhibited as well as corrected values.

Application of Theory of Experiment

As the external magnetic field approaches zero, the interaction of $\vec{I}$ and $\vec{J}$ with the external field becomes much less than their interaction with each other. This is equivalent to the remark that they are tightly coupled to each other, as in Fig. 3a, so that $\vec{F} = \vec{I} + \vec{J}$ is a good quantum-mechanical vector and so that m_F is a good quantum number. Therefore we may label the wave function by F and m_F , and for the low-field interaction energy write³²

$$W = - \vec{\mu}_F \cdot \vec{H}, \quad (24)$$

where $\mu_F = g_F \mu_0 \vec{F}$, and g_F is derived from the vector model,

$$g_F \mu_0 F = g_J \mu_0 \frac{\vec{J} \cdot \vec{F}}{F} + g_I \mu_0 \frac{\vec{I} \cdot \vec{F}}{F}. \quad (25)$$

At the initial point of the experiment g_I is unknown, but since the mechanical angular momentum of nuclei is known to be of the order of $\hbar$, while nuclear moments are of the order of nuclear magnetons, one can infer that $g_I \cong \frac{m}{M} g_J$ and may be neglected at low fields. Therefore we have

$$g_F \cong g_J \frac{\vec{J} \cdot \vec{F}}{F^2} = \frac{1}{2} g_J \frac{F(F+1) - I(I+1) + J(J+1)}{F(F+1)}. \quad (26)$$

For a $^2P_{3/2}$ ground state g_J equals 4/3 and previous work on stable isotopes of bromine have shown only minor deviations from this.²⁷ Now, we have

$$g_F = \frac{2}{3} \frac{F(F+1) - I(I+1) + J(J+1)}{F(F+1)}, \quad (27)$$

and by observing that a flop-in transition exists in each of the two F states $\vec{F} = \vec{I} + \vec{J}$ and $\vec{F} = \vec{I} + \vec{J} - 1$ (the transitions being referred to

(a) LOW FIELD COUPLING

(b) HIGH FIELD COUPLING

MU-13646

Fig. 3. The two extreme coupling models for $\vec{I}$ and $\vec{J}$.

hereafter as α and β respectively) a discrete set of g_F 's may be calculated. Two g_F 's result for each value of I . The predicted transition frequencies for a given spin and magnetic field can then be obtained by calculating $\Delta E = h\nu = \langle F, m | -\vec{\mu}_F \cdot \vec{H} | F, m \rangle - \langle F, m' | -\vec{\mu}_F \cdot \vec{H} | F, m' \rangle$.

For convenience, the direction of the external field is usually selected to be the z axis. Then introduction of an oscillating magnetic field in the xy plane produces transitions between m values differing by one. We therefore require $(m-m')=1$ and

$$h\nu = -g_F \mu_0 \left\{ \langle F, m | \vec{F} \cdot \vec{H} | F, m \rangle - \langle F, m-1 | \vec{F} \cdot \vec{H} | F, m-1 \rangle \right\},$$

$$\nu = -g_F \frac{\mu_0 H}{h} \left\{ m - (m-1) \right\} = -g_F \frac{\mu_0 H}{h}. \quad (28)$$

In principle, therefore, the spin I can then be uniquely established by one low-field observation, the only limitation being the possibility of overlap between one spin frequency and another due to finite line width or shifts due to small values of a . Overlap occurs only at very low fields and is usually easy to detect, as the approximate resonance line width of the apparatus is known. Observation of both the α and β transitions of an isotope at a given field greatly decreases the possibility of error, and observations are repeated at several values of field before definitely I is assigned.

In conjunction with the spin determination, positive isotope identification must be made. This can be a difficult procedure, but for the bromines is quite easy on the basis of half life. Decays of both the full beam³³ and resonance exposures are taken for this purpose. Full-beam decays establish the isotopic composition of the beam, while the resonance-button decays insure that the given I is assigned to the proper isotope.

While I is uniquely determined by the above observations, no information regarding the interaction constants a and b can be inferred³⁴. This information principally comes from a transition of another sort, which approaches a nonzero frequency for zero magnetic field. Such "direct"

transitions between adjacent hyperfine levels involve $\Delta F = \pm 1$.

Before searching for these transitions, whose frequencies cannot be predicted at the start, one must obtain information as to their approximate value. This may derive from optical spectroscopy, but a previous estimate of the hyperfine structure is practically never available in the case of a radioactive nucleus and the information must be obtained in the course of the experiment.

Observations of the α and β transitions must be made at higher and higher values of magnetic field. When the field becomes great enough to perturb the coupling between I and J by a significant amount, the observed resonance frequency will no longer be equal to that calculated by $-g_F \frac{\mu_0 H}{h}$. The linear dependence on magnetic field assumes an infinite hyperfine-structure interval and, consequently, the deviations from linearity give information regarding the actual hyperfine structure. By second-order perturbation theory one may obtain, for the α transition in a $^2P_{3/2}$ atom,

$$\delta\nu = \frac{2}{3} \frac{I^2}{I+1} \frac{\nu_0^2}{\Delta\nu_{12}}, \quad (29)$$

and for the β ,

$$\delta\nu = \left[\frac{(2I+1)^2 (2I+3)^2}{(2I+9)^2} \right] \left[\frac{-6I(I-1)}{(I+1)(2I+3)^2 \Delta\nu_{12}} + \frac{2(2I+3)(2I-1)(I-1)}{I(I+1)(2I+1)^2 \Delta\nu_{23}} \right] \nu_0^2, \quad (30)$$

where

$$\nu_0 = -g_F \frac{\mu_0 H}{h}, \quad \delta\nu = \nu_{\text{obs.}} - \nu_0,$$

$\Delta\nu_{12}$ = frequency separation of $F, F-1$ levels,

$\Delta\nu_{23}$ = frequency separation of $F-1, F-2$ levels, where

$F = F_{\text{max}}$ and levels are in normal order.

When estimates of the $\Delta\nu_s$ have been made as above, values for a and b can be derived. The Hamiltonian is first written in the form

$$\left(\frac{\delta}{ha}\right)_F = (\vec{I} \cdot \vec{J})_F + \left(\frac{b}{a}\right) Q_{OPF}, \quad (31)$$

where the subscript F indicates the level for which a solution is obtained. All factors in the equation can be calculated except b/a . For $b/a = 0$, only a dipole interaction exists and the hyperfine intervals are, from highest F to lowest, proportional to $F, F-1, F-2$, etc., a relation known as the interval rule. Since estimates of the $\Delta\nu_s$ are available, one may write

$$\Delta\nu_{F \rightarrow F-1} = a \left\{ \left(\frac{\delta}{ha}\right)_F - \left(\frac{\delta}{ha}\right)_{F-1} \right\}. \quad (32)$$

This gives two equations in two unknowns so that a and b can be calculated, with an accuracy depending on the magnitude of the δ and the resonance line width, i. e., the relative accuracy in the $\delta\nu$'s.

These preliminary values allow one to predict the resonance frequencies at even higher values of H . Once resonances have been obtained at the higher values of H , higher-order perturbation terms may become appreciable and a second-order calculation is not sufficient. Further computations are performed on an IBM-650 computer by the use of two programs which have resulted in an enormous decrease in hand calculation. These programs have been described in detail in Ref. (7) and it will suffice here to describe their function.

The first program computes the magnetic-field dependence of a given transition. The required input information is: $I, J, b/a, F_1, m_1, F_2, m_2$, and $\delta\rho$, the increment of (dimensionless)

magnetic field between calculated points. Output cards will then be punched, starting at zero field, in increasing steps of $\delta\rho$. They contain: the transition frequency divided by a ; transition frequency divided by $a\rho$; the energy values of the levels involved; and the partial derivative of transition frequency vs field.

It is then a matter of only a few minutes of computer time to calculate the predicted frequency for any given resonance when estimates of a and b are available.

The second program performs a least-squares fit to resonance data. Inputs to the program are: I, J, a , and b , where a and b need be only rough estimates. For each observation an input card must be supplied containing resonance frequency; magnetic field; m_1, m_2 ; a weighting factor for the resonance; and the energy values of the levels involved. Given this information, the program computes new values of a and b , the errors in these, and a goodness-of-fit parameter χ^2 . The parameter χ^2 decreases with each iteration as the program converges to a best fit. When the operator observes that χ^2 has ceased to decrease, he can cause the computer to punch out cards containing the input information, except that the level energies will be changed to the best values given by the newly calculated a and b values, and the frequency difference between the observed resonance and the position at which the least-squares fit predicts the resonance to be will be included. Finally, a card is punched giving the best values of a and b , the errors in each, and χ^2 .

Any transition may be used as input to either program whether it satisfies a selection rule or not. Experimental work can then proceed as follows:

From predictions made by the first program, resonances can be observed at higher magnetic fields. These are utilized in the second program, giving better values of a and b , which may then be used in the first program to carry on to even higher fields. This process is continued, always making as large an increase in magnetic field between observations as possible without causing the uncertainty in

position of the predicted frequency to be so large that the search becomes unmanageable. When the errors in a and b have become small enough so that the calculated errors in $\Delta\nu$ are only a few megacycles, a search is made for the $\Delta F = 1$ transitions.

When these have been observed, the errors in a and b are reduced to a minimum and the only undetermined parameter still accessible to the experiment is the sign of g_I . As discussed in the calculation of g_F , the value of g_I is about $1/2000$ that of g_J and is unobservable at low fields. To determine this, observations of a resonance must be made at such a high field that the term $-(\Delta m_I)g_I \frac{\mu_0 H}{h}$ contributes a shift in frequency comparable to the apparatus line width. Two frequency predictions are then made, one for each sign of g_I . Observation of a resonance at one of these two frequencies then completes the determination. Unfortunately, on the present apparatus it has not been possible to determine the signs of any moments to date. The rather large line width of the apparatus implies that the field for observation must be very large, and it has been found that the line width increases with magnetic field at such a rate as to almost cancel any gains made.

As will be seen in Section V, interpretation of the measurements will be made in terms of the nuclear-shell model and in most cases the single-particle model. An adequate discussion of the shell model would be too long to include here and, because there are a number of very readable discussions available, would be superfluous. Reference is therefore made to Ref. 16 and Refs. 35, 36, and 37.

III. EXPERIMENTAL PROCEDURE

The experimental work reported here has been done on both neutron- and alpha-induced activities, and there are, therefore, two different approaches to sample preparation.

Bromine-82, with a 35-hour half life, was the first isotope of the series on which work was done. This was selected primarily because of the ease with which samples could be obtained. It was purchased from Oak Ridge National Laboratory in units of 1.2 grams of KBr powder, each containing approximately 100 Mc of bromine-82. Samples were bombarded for one week and shipped by air, arriving about 30 hours after being removed from the pile. No activity other than bromine-82 was observed in these samples.

Pile-produced bromine-80m and -80 with half-lives of 4.5 hours and 18 minutes respectively, were obtained by bombardment of KBr powder in a flux of about 10^{12} neutrons/cm²/sec at the Livermore pool type reactor (LPTR) recently put into operation. Samples were sealed in Pyrex or quartz capsules and enclosed in plastic for the actual bombardment. Bombardments were for two to three hours. Samples arrived at Berkeley about 1-1/2 hours after the end of bombardment.

All of the cyclotron-produced isotopes for which experimental results are reported here were produced by the reaction $As^{75}(\alpha, xn)$, where $x = 2$ or 3 . For producing bromine-76 by means of the $3n$ reaction, the full cyclotron beam energy of about 46 Mev at 30 microamperes was used. Reasonable atomic-beam intensities resulted when the integrated flux was greater than 70 microampere-hours ($\mu a-hr$).

In the case of 57-hour bromine-77, a beam degraded by 8 Mev was selected to minimize the production of bromine 76. On these bombardments, 300 $\mu a-hr$ is desirable, and negligible bromine-76 activity has been detected in beams produced within 12 hours after concluding the bombardment.

A "cat's eye" target was used for all cyclotron bombardments. These targets consist of an aluminum block with internal water-cooling passages, and a cover plate with a rectangular aperture through which the beam passes. Arsenic powder was placed in a small depression pressed in a 0.010-in. aluminum plate and covered by two 0.001-in. aluminum foils. In use, the back plate is in contact with the water circulating inside of the target block, while the front foils are cooled by an atmosphere of helium. The whole sandwich is held onto the block by the front plate, which in turn is held down by two screws.

One of the principal advantages of this target assembly was that it could easily be unloaded remotely in a lead-shielded cave. This was done in a lead holder with one end cut down in such a way that after removal of the front hold-down plate the entire sandwich could be slid gently into a test tube. Once in the test tube, the backing foil was scraped with a spatula to remove any caked powder, and then discarded. The same was then done with the inside front foil by holding it with tweezers and sweeping the arsenic off with a stiff brush. Once all of the foils were scraped and removed, a stopper was inserted and the test tube transferred to a glove box where the chemical separation was performed.

Previously it was the practice to process the sample with one or more of the aluminum foils in the reaction vessel. On a number of occasions this caused trouble and the foils should be discarded as described above. Inclusion of aluminum seems to have been the cause of a gelatinous mass which sometimes formed in the reaction vessel and acted as a trap for much of the activity.

The chemical procedure used to prepare the samples for the beam apparatus has undergone continuous modification of detail. Modifications have been introduced with a view to reducing personal exposure by increasing the reliability and simplicity of the chemistry. Increased reliability has resulted in less handling of the glassware during processing and in a much more rapid process, which becomes important in work on bromine-80m-80 where the half lives are short.

Because bromine is such a volatile material, a malfunction of the chemistry apparatus usually caused loss of the sample. An earlier form of the chemical procedure is described in Ref. 7, while the most recent approach will be described here. Glassware used for this process is shown in Figs. 4 and 5.

In the event that further work may be done on bromine, the preparation of the samples will be described in rather great detail.

This final chemical procedure is equally effective for a sample of either arsenic powder or potassium bromide. Natural bromine, for carrier, was placed in the reaction vessel as a weighed amount of KBr powder before introduction of the active material. After the active sample had been placed in the vessel and the stopper assembly put in place, dry helium was passed through the glassware to remove moist air. A small flow of helium was maintained through the glassware during the entire process.

After letting the helium flow for a short while, the right-hand vial was cooled in liquid nitrogen and a few cc of concentrated sulfuric acid slowly introduced into the left-hand vessel. When the evolution of hydrogen bromide, bromine, and sulfur dioxide had ceased, more acid was added until the addition of acid produced no more gas. At this point, sufficient acid was added to cover the sample by about a half inch, or more. Then, by adding hydrogen peroxide in very small increments, the remainder of the bromine could be released by the reaction

and carried into the cold vial by the helium flow. At the conclusion, the liquid in the reaction vessel should be practically clear. It is probably worth noting that the first additions of peroxide produced only small amounts of bromine, while some later addition released almost all of the bromine at a rapid rate. It is because of this that the peroxide must be added so slowly; otherwise bromine would be evolved so rapidly that it could carry on past the cold surface in appreciable amounts or even force the glass joints apart.

MU-17594

Fig. 4. Glassware used for separation of bromine from targets. Reaction vessel is at left and connecting tube contains P_2O_5 to absorb water vapor. The vial at right is ultimately closed and removed to the atomic-beam apparatus.

ZN-2171

Fig. 5. Vial for radioactive bromine samples. Gas flows in through the side tube and out through the center tube to avoid clogging of the center tube. Center-tube diameter and distance from bottom of vial must be approximately as shown for optimum performance.

At this point the vial contained a light yellow or white substance in considerable quantity, in addition to the bromine. This was probably SO_2 , a decomposition product of the acid. To remove it, the vials were placed in a dewar full of ice water and the volatile materials evaporated off leaving the bromine. Unfortunately, evaporation under these conditions was so rapid that some bromine was carried away.

When the vial was at ice-water temperature, it was sealed by closing the stopcock and inserting a glass stopper in the 7/15 joint. Then the vial was recooled in liquid nitrogen and quickly transported to the atomic-beam apparatus. It should be noted that sealing of the vial must be done before cooling in liquid nitrogen. Otherwise, cold air will be inside of the vial and will expand during the transfer to the beam apparatus, causing the stopcock to blow out of the vial and allowing the possible escape of bromine.

IV. APPARATUS AND EQUIPMENT

An atomic-beam apparatus for work on radioactive samples must generally be designed so that the intensity of the refocused beam at the detector is a maximum. This is modified only by the need for a low radioactive background.

Although there are of the order of 10^7 atoms in the beam at any instant, the interactions of the atoms with each other are orders of magnitude less than in the solid or liquid states. Consequently, the field in which the atom is observed is at the control of the experimenter and is selected to be a homogeneous magnetic field.

Upon emerging from the beam source, an atom passes through an inhomogeneous magnetic field, which serves as a state selector, and then through the homogeneous field where a perturbing radiofrequency field may be introduced. Finally the atoms pass through another inhomogeneous field that focuses atoms, which have made the desired transition, onto the detector as in Fig. 6. This is known as a flop-in configuration, inasmuch as only atoms that underwent a transition are refocused. This arrangement is the one usually used for radioactive beams since it gives a relatively large increase in counting rate on a small background, rather than a decrease on a very large background, as is the case in flop-out experiments.

An atomic-beam apparatus is equivalent, geometrically, to a Schlieren optical system in that only particles describing a particular curved path can reach the detector. The resemblance is even more marked in the case of atomic-beam equipment designed with cylindrical symmetry,³⁸ inspired by the increased intensity available at the detector from increased solid angle. Reduction of apparatus background is as important as high intensity. Low background is generally achieved by maintenance of low pressures, efficient trapping, and accurate geometric alignment. While the first two factors reduce scattering out of the beam and the possibility of a scattered atom

MIJ-13185

Fig. 6. Trajectories of atoms in an atomic-beam apparatus.

reaching the detector, the last item may not be easily handled. If, for example, there exists an appreciable variation of $\text{grad}H$ across the beam, the trajectories will not be symmetric. That is, the center line of the beam with the magnets off will not coincide with that when the magnets are on, and the focal point of the refocused beam will lie off the center of the apparatus. Moving the detector over to intercept the maximum refocused beam will raise the background, as the detector will no longer be in line with the stop wire. Similarly, moving the stop wire over in line with the new detector position will cause it to intercept a considerable amount of the refocused beam, and the signal-to-noise ratio suffers no matter which solution is attempted. The usual cause of this effect is the use of a very short and narrow-gap A magnet in which the beam occupies a large percentage of the gap. Thus, the gain in solid angle by diminishing the size of the A magnet does not always result in an increased signal.

When all other factors have been considered, there still remains a residual background resulting from Majorana transitions. These arise from changes of magnetic field. If, in the coordinates of a beam atom, these changes contain appreciable Fourier components near a transition frequency, the atom may make a transition. A special case of this occurs if, at some point in the trajectory, the magnetic field passes through zero and changes direction, causing the atoms to lose their quantum mechanically fixed m_j . Upon re-entering a magnetic field they become reoriented among the various m 's, resulting in a considerable refocused beam. In a cylindrical apparatus the field configuration is so complex, and the possible locations for pumps and traps so few, that it is not obvious that backgrounds can be controlled to the extent that their theoretical advantages are realized.

Many published descriptions of atomic-beam installations are available and no attempt will be made here to describe the equipment in detail. Instead, the discussion will be restricted to aspects of the apparatus that are nonstandard.

An isometric drawing of the apparatus is shown in Fig. 7 and illustrates the principal features with a minimum of distracting detail. The total length of the beam itself is about one meter.

The vacuum system has been designed to incorporate a minimum of internal volume so that it may be rapidly pumped from atmospheric pressure to a working pressure of about 10^{-6} mm Hg. To achieve this end, the spaces through which the beam passes have cross sections of only a few square inches. In the A- and B-magnet regions, the sides of the vacuum chamber are formed by the pole faces, while the C-magnet poles are outside of the vacuum. Originally, it was intended to have all of the pole faces outside of the vacuum and to pass the beam down a small tube placed between the poles. This design was found to be unworkable and is described in Ref. 9. In the present arrangement there is ample area adjacent to the beam for liquid-nitrogen-cooled traps and for pumping away scattered atoms.

Valves, marked J in the figure, are mounted at the high-vacuum end of each diffusion pump. These may be closed and the pumps left running when it is desired to bring the apparatus up to atmospheric pressure. With this arrangement it is possible to pump the system down to less than 10^{-5} mm Hg in about ten minutes. In the drawing, diffusion pumps can be seen on the oven can, detector can, and A and C magnets. The B magnet is pumped through the detector can. It has been observed that the background counting rate decreases appreciably with the length of time the apparatus has been under vacuum, even though the indicated pressures show little change. For this reason a 1/16-in. stainless steel slide valve has been installed in the oven-can exit so that sources may be changed without bringing the remainder of the apparatus up to atmospheric pressure. Although impossible to incorporate in the present oven can, a desirable modification would be an arrangement whereby the calibrating oven F could also remain under vacuum, or be brought up to atmospheric pressure and reloaded separately.

MU-13930

Fig. 7. Isometric drawing of the atomic beam apparatus used for the measurements on bromine.

A, B, and C. magnets; D, detector-can window for optical alignment of apparatus; E, "oven" or source can; F, alkali oven for magnetic-field calibration. For use, it is raised and lowered by a motor and chain (not shown); G, elevator for introduction of beam-collection buttons into vacuum chamber (pumping lines to this assembly are not shown); H, stop-wire assembly; I, liquid nitrogen traps; J, gate valves to close off diffusion pumps from vacuum system.

All of the magnet windings and flux return paths are entirely outside of the vacuum system and do not become contaminated with radioactivity. This enables one to replace the windings and make repairs with relative ease. Each section of the vacuum system is connected to adjacent sections by means of flexible bellows and O-ring seals so that they may be individually removed for repair, leak testing, etc.

The A and B magnets are designed to be run in series at 300 volts and 5 amp. This is supplied by a regulated 3-phase power supply which is stable to about 1 part in 30,000. The circuit of this supply is shown in Ref. 39.

Current to the C field can easily be supplied by automobile storage batteries, as 480 ma corresponds to about 1450 gauss. On recent work, an electronically regulated current supply has been used. The control element in this supply is a 2N174 transistor mounted on a copper plate and cooled by forced air. In use, the drift of this instrument is undetectable, and on bench tests the drift has been found to be about 1 part in 10,000 over a period of 15 minutes.⁴⁰ For reference, the circuit is reproduced in Fig. 8.

The C magnet poles are $2\frac{1}{4} \times 1\frac{3}{4}$ -in. bars of Armco steel and are entirely external to the vacuum chamber. When carefully aligned with one another, a resonance-line width of about 80 kc is available on potassium. Greater line widths can be immediately obtained by misaligning the pole pieces. This feature has been found to be of great value. When searching for a transition whose frequency is inaccurately known, the rate at which a frequency range can be covered is inversely proportional to the line width, so that a greater line width means a faster sweep rate. Once the desired transition has been located, the poles may be realigned in about five minutes and the resonance traced out with the best available accuracy.

Slits defining the beam width are located at the exit of the C field and are adjustable externally with the apparatus under vacuum.

The stop wire is of a rectangular cross section and may be adjusted in width across the beam from 0.053 to 0.103 in. and moved across the beam from one pole face to the other without disturbing the vacuum.

The performance of the deflecting magnets can be estimated most easily by making measurements on the alkali calibrating beam. An atom in a beam will suffer a deflection proportional to the force of deflection divided by the absolute temperature of the source. Since the effective source temperatures for the bromine and potassium beams are not widely different, one may assume that potassium is deflected about 1.5 times the deflection of bromine. In the case of potassium, we have $I = 3/2$ and $J = 1/2$, so there are eight hyperfine levels, of which two contribute to the allowed flop-in transition, and a maximum resonance of 25% of the full beam would be theoretically possible. Tests on the present apparatus indicate a resonance height of about 8%, which is reduced to half of this upon inserting the stop wire. Setting the magnets to their optimum current for bromine, one would expect that the 50% greater deflections of the alkali would carry a significant portion of it into the pole faces. Lowering the magnet current then should result in an increase in the resonance height until, with the stop wire out, a refocused beam of the order of 25% should be observed. In actuality a maximum of less than 12% occurs. The small increase in resonance height upon field decrease, together with the 50% decrease in resonance height upon inserting the stop wire, seems to imply that the deflecting power of the magnets is undesirably low.

There is also evidence that the trajectories of the atoms are not symmetric about the geometric center line of the apparatus. Upon setting the stop wire for minimum background a maximum of refocused beam should occur, as atoms could be deflected around the wire on either side whereas an off-center stop would intercept refocused atoms in greater numbers. With the present magnets the resonance is largest when the stop is near the convex pole, decreasing to a minimum

as the wire reaches a point about two-thirds of the way across to the concave pole. The resonance increases abruptly as the stop approaches closer to the concave pole. It therefore appears that many more atoms are being refocused around the concave-pole side of the stop wire than on the convex side, when the stop wire is set for minimum background on full beam.

With 50% of the potassium beam resonance hitting the stop wire, considerably more than this amount of the bromine resonance must be stopped. In view of this it does not seem unreasonable to expect that an improved magnet system could result in a gain in signal-to-noise of two or three.

In the case of an atomic bromine beam, the source is a quartz discharge tube which is shown in its latest form in Fig. 9. The forward part, carrying the electrodes and slit, is made of quartz, while the remainder is of Pyrex with metal where it is soldered into the brass plate. A slit is formed in the end of the tube by blowing it out to a thin shell and then sawing with a fine wire and abrasive. Ground joints 1 and 2 are sealed in place with black wax so they may be adjusted or replaced without removal from the glove box. This is done by heating them with a piece of Nichrome tape operated by filament-supply leads.

Bromine molecules are dissociated into atoms by a radio-frequency discharge maintained in the quartz tube. Radiofrequency leads to the tube make friction contact with 0.003-in. nickel foils wrapped around the tube. The foils are spot-welded to the proper size and slid into position. Coupling to the tube is enhanced by painting the quartz with Aquadag in the regions where the foils are mounted. The discharge is excited by a 450-kc resonant circuit through a Steinmetz regulator as shown in Fig. 10. Analysis of the regulator circuit shows that, at resonance, the current through the tube is independent of the resistance at the tube. Thus, constant discharge current can be maintained without attention during a run.

MU-17252

Fig. 9. Schematic top view of source-end of atomic-beam apparatus, showing position of calibrating oven, discharge tube, and slide valve.

The whole source assembly is carried on a 4×4-in. brass plate which may be translated both horizontally and vertically without disturbing the vacuum in the machine. Alignment of the tube in the apparatus is done visually by striking an iodine discharge and sighting down the apparatus with a telescope. Iodine is convenient for the purpose, as its vapor pressure at room temperature is suitable for a discharge.

To obtain a stable discharge and reasonable rate of effusion, the pressure inside of the discharge tube should be of the order of a few tenths of a millimeter. At room temperature the vapor pressure of bromine is much greater than this and so the effusion rate must be controlled by either refrigerating the vial or use of a slow leak.⁴⁰

A leak was selected for the entire control of the flow rate although previous workers have used combinations of leaks and cooling systems in work on chlorine, which has an even higher vapor pressure than does bromine.^{41, 29} Several sintered-glass leaks were constructed and the one most nearly satisfying our requirements was used. These leaks were made by packing powdered glass into a tube of about 3-mm diameter to a length of about 1/8 in. Then, using a soft flame, the tubes were heated until the powdered glass was seen to adhere to the outer tube. Preliminary testing, to reject completely closed tubes, was done by bubbling air through under pressure. With a given leak rate, the beam intensity and duration could still be varied by a factor of 3 or more by varying the amount of carrier added during the chemical processing. Final adjustments of beam were made by use of an infrared lamp run from a variable transformer or by cooling in a constant-temperature bath, such as ice and water.

Although the leak originally selected served well for over a year, the most recent work has been done with leaks of a different design. The new leaks are constructed and their thruput estimated as suggested by Gordon.⁴² In this work a leak with an "apparent diameter" of 2.3 microns is used and gives an effusion rate of the order of 0.3 cc per hour of liquid bromine.

RF DISCHARGE TUBE

DISCHARGE STABILIZER

MU-14243

Fig. 10. Steinmetz regulator circuit and old-style of discharge tube.

After a vial of active bromine has been passed into the glove box on the atomic-beam apparatus, a very high radiation level is usually present in front of the box. To obtain a beam with the minimum of handling and exposure, the following procedure has been developed and will be described in detail for the convenience of any future experimenter.

The vial is placed on the leak, the other end of which is attached to the ground joint as indicated in Fig. 9. Then, with stopcock A opened to the forevac and with a bypass stopcock of the leak open, the glassware is pumped out as far as the closed stopcock on the oven vial, which is full of helium and bromine at approximately atmospheric pressure. While the glassware is being pumped out, the vial is cooled by a dry-ice and alcohol mixture. After the bromine is frozen, the vial stopcock can be opened and the vial pumped down to forevac pressure. After closing the leak-bypass stopcock, stopcock A is opened to the oven can. If the pressure in the can indicates that there are no leaks in the glassware, the vial is removed from the cold bath. A steady beam is obtained about twenty minutes after removing the vial from the alcohol mixture and no further attention to the sample should be necessary for the duration of the run.⁴³

Beam atoms are detected by their radioactive decay after collection on a silver surface. These surfaces are vacuum-evaporated onto brass or stainless steel "buttons" and stored under vacuum until use. Several views of such buttons are shown in Fig. 11. Their dimensions are $3/4 \times 5/8 \times 1/4$ in. In the counters or beam apparatus they are held in position by a spring-loaded ball which fits into the slot on the side of the button and which may be removed with long-nose pliers whose jaws fit into the two holes in the rear of the button.

A review article dealing with the subject of collection of beams has recently appeared.⁴⁴ In the article no mention of chemical combination appears explicitly, yet there is reason to believe that this may be playing a role in the present work. In order to discover

ZN-1732

Fig. 11. Brass beam-collection buttons.

whether bromine would evaporate off of an exposed button and possibly contaminate the counters, the following experiment was performed. An exposed button, of high counting rate, was counted and then heated on a hotplate until too hot to hold with the bare hand and recounted after cooling. No change in the counting rate was observed, and in view of the very volatile nature of bromine it must be assumed that the bond to the surface was considerably stronger than simple condensation. As AgBr is a very stable compound, and since buttons which have been exposed for a few hours to air collect with only about 50% the efficiency of fresh surfaces, chemical formation of AgBr seems to be a reasonable explanation.

Buttons are introduced and removed from the apparatus by an elevator which passes through O-ring seals. Intermediate spaces are pumped by a mechanical vacuum pump, and buttons may be changed in about 10 sec without introducing excessive gas into the detector can.

After exposure the buttons are counted in small-volume continuous-gas-flow beta counters as shown in Fig. 12. Methane is used as a filling gas and the counting head is a small loop of 0.001 to 0.003-in. tungsten wire maintained at 2900 volts with respect to the housing. The very small volume of the counter makes possible a relatively low background counting rate. When in use the heads are completely enclosed within lead housings and the background is from one to six counts per minute, depending on the condition of the loop, discriminator and attenuator settings, and the intensity of radiation in the room. Pulses are amplified and counts displayed on scalars with decimal read out.

To make the recording of information on decay rates more convenient, an automatic count-recording device has been assembled. The basis of this device is an electrically operated timing and print-out machine constructed by the Clary Company.⁴⁵ This machine is equipped with a timing motor which generates digital time in minutes and hundredths of a minute that may be printed on a paper tape upon receipt of an electrical command. In addition there are two rows of

MU-17401

Fig. 12. Drawing of small-volume continuous-gas-flow beta counters as used in this experiment.

numerical inputs which may be used for identification information. For the operation of the device we have constructed an external circuit which causes the machine to print a counter identification number along with the time at which the print command is received. The two scalars used with the circuit have modified mechanical registers containing microswitches that close on the 10th, 100th, or 1000th impulse received by the register. Since the scalars in question may be set so that the registers receive an impulse every 2^n counts, where n is from 1 to 7, all counting rates encountered in the course of this work have been easily accommodated.

During the course of a run a rough plot of the counting rate of the buttons is maintained, usually using the result from just one of the two counters. For final resonance plots the important exposures are usually recounted to improve counting statistics, the results of both counters averaged, a probable error calculated, and the results plotted. For the rapid calculation of errors within the counting range of our usual interest, the nomograms reproduced in Figs. 13 and 14 were constructed. If, however, a great many buttons are to be corrected for decay, counter efficiency, and several recounts, it is usually more convenient to punch the information on IBM cards and do the arithmetic by use of an IBM-650 computer. A program has been devised to accomplish this and the time spent in constructing a program has been rapidly repaid in decreased numerical drudgery.

As discussed in Section II, continuous-wave radiofrequency signals are used to induce transitions among the hyperfine levels. Frequencies used in this work have varied from essentially zero up to about 3,000 Mc. Frequency determinations up to 220 Mc were made by a Hewlett-Packard (H. P.) model-524B frequency counter and associated plug-in converters. For higher frequencies an H. P. transfer oscillator model-540A was used between the signal source and frequency counter. At less than 50 Mc a General Radio model-805C and a Tektronix Type-190 signal generator were used. From 10 to 500 Mc a H. P. model-608A oscillator was available, and to 1200 Mc

MU-17588

Fig. 13. Nomogram for the solution of the equation

$$\sigma = \sqrt{\bar{C}/T}$$

MU-17589

Fig. 14. Nomogram for the solution of the equation

$$x = 1/2 \sqrt{\sigma_a^2 + \sigma_b^2}$$

a H. P. model-612A oscillator was used. An Airborne Instruments Laboratory type-124C oscillator was obtained during the course of the work and subsequently used for all work in the range of 200 to 3000 Mc. At first this oscillator suffered from excessive frequency modulation, but after modification to battery-operated filament and an external stabilized plate supply, the instrument could be held to within ± 5 kc for 5 minutes, a negligible fraction of the apparatus line width.

V. DISCUSSION OF INDIVIDUAL ISOTOPES

Bromine-82

The initial exposures were made in the linear Zeeman region to determine the spin. Figure 15 shows the results of an early spin-search at fields of 2 and 4 gauss; spin 5 is clearly indicated. Both the α and β resonances were observed at five different values of magnetic field up to 417.4 gauss. Figure 16 shows an α resonance observed at a field of 83.83 gauss ($\nu_{\text{Cs}} = 30$ Mc), and Fig. 17 a β resonance observed at a field of 164.03 gauss ($\nu_{\text{Cs}} = 60$ Mc). All the resonances observed are consistent with an assignment of 5 for the nuclear spin of bromine-82. In a typical run, for a ten-minute button exposure the full-beam counting rate was 2500 cpm, the resonance heights approximately 35 cpm, and the apparatus background (i. e., the button counting rate with the A and B fields switched on and the stopwire inserted into the path of the beam) was 10 cpm. The dissociation efficiency of the discharge was in a typical instance $\sim 80\%$. The α and β resonances observed at and below 417.4 gauss enabled the constants a and b to be calculated with sufficient accuracy so that a search for the direct hyperfine transitions could be attempted. The searches were made with a broad line which was narrowed down once a resonance had been spotted. Figure 18 shows the $\Delta M_F = 0$ resonance between the upper pair of hyperfine states at a field of 56.3 gauss ($\nu_{\text{Cs}} = 20$ Mc). The line is a little broader than expected from measurements of the cesium resonance line-width, probably because the resonance was observed with the rf loop oriented to observe $\Delta M_F = \pm 1$ transitions.

Figure 19 exhibits $\Delta M_F = \pm 1$ resonances between the two hyperfine states $F = 11/2$ and $F = 9/2$ at a field of 0.95 gauss ($\nu_{\text{Cs}} = 0.3$ Mc). The line is broad owing to the fact that there are 17 possible $\Delta M_F = \pm 1$ transitions between these two hyperfine states, all coincident at zero field but partially resolved at 0.95 gauss. In order to increase the precision of measurement of the $F = 11/2 \rightarrow F = 9/2$ interval, one of these resonances [the ($F=11/2$, $M_F=-7/2 \rightarrow F=9/2$, $M_F=-5/2$) transition] was followed to fields high enough so that it

MU-14318

Fig. 15. Bromine 82 spin-search exposures at 2 and 5 gauss.

Fig. 16. Bromine 82 α resonance at 83.8 gauss.

MU-17270

Fig. 17. Bromine 82 β resonance at 164.0 gauss.

MU-17269

Fig. 18. Bromine 82 $\Delta\nu_{12}$ resonance at 56.3 gauss.

MU-17268

Fig. 19. Bromine 82 $\Delta \nu_{23}$ resonance at 0.95 gauss.
 Resonance curve is a composite of 17 almost
 superimposed transitions.

became completely resolved from its partners, the measured g_F value being used for positive identification. The resonance ($F = 11/2$, $M_F = -5/2$) $\rightarrow$ ($F = 9/2$, $M_F = -5/2$) has also been observed at a field of 28.37 gauss ($\nu_{Cs} = 10$ Mc). The frequencies and fields of all transitions that have been observed in bromine-82 are tabulated in Table I together with the uncertainties assigned to the positions of the resonance centers. The uncertainties have been computed by assigning an uncertainty of one-third the resonance width at half maximum both to the bromine and cesium resonances, and compounding these errors by using the equation

$$\omega_i = \frac{1}{(\Delta f_i)^2 + \left(\frac{\partial f_i}{\partial H}\right)^2 (\Delta H)^2}$$

as defined in Appendix I of Ref. 7.

Table I

Frequencies and fields of transitions observed in bromine-82

Resonance type	Resonance frequency (Mc)	Compounded uncertainty (Mc)	Magnetic field (gauss)	Cesium frequency (Mc)	Weighting factor (W)
α	1.25	0.06	2.86	1	259
α	6.26	0.06	14.24	5	251
β	3.54	0.05	14.24	5	340
α	12.81	0.07	28.37	10	192
β	7.19	0.05	28.37	10	474
α	6.31	0.06	14.24	5	325
α	41.13	0.11	83.83	30	83
β	22.15	0.08	83.83	30	163
α	90.30	0.24	164.02	60	18
α	90.25	0.11	164.02	60	81
β	173.05	0.52	417.38	164	3.6
β	173.05	0.66	417.38	164	2.3
α	304.70	0.43	417.38	164	5.6
β	46.95	0.07	164.02	60	224
$(13/2, -9/2) \leftrightarrow (11/2, -9/2)$	727.50	0.21	56.31	20	22
$(11/2, -7/2) \leftrightarrow (9/2, -5/2)$	1281.50	0.18	5.71	2	57
$(11/2, -7/2) \leftrightarrow (9/2, -5/2)$	1273.10	0.16	14.24	5	41
$(11/2, -7/2) \leftrightarrow (9/2, -5/2)$	1281.60	0.14	5.71	2	72
$(11/2, -7/2) \leftrightarrow (9/2, -5/2)$	1281.60	0.10	5.71	2	100
$(11/2, -7/2) \leftrightarrow (9/2, -5/2)$	1273.03	0.10	14.24	5	110
$(11/2, -5/2) \leftrightarrow (9/2, -5/2)$	1265.91	0.10	28.37	10	100

Results and Discussion

As previously described the hyperfine constants were evaluated by using an IBM digital computer. The machine calculation gives

$$a = 205.04 \pm .05 \text{ Mc}$$

and

$$b = - 870.66 \pm .90 \text{ Mc.}$$

The values of χ^2 with g_I assumed positive and negative are

$$\chi^2 (g_I > 0) = 3.5$$

and

$$\chi^2 (g_I \leq 0) = 6.4.$$

The uncertainties quoted are five times the uncertainties calculated on the basis of the error assignments discussed above. This factor of five is intended to allow for unknown systematic sources of error that might be present. A value of $g_J(\text{Br}) = -1.3338 \pm 0.0003$, obtained from the measurements of King and Jaccarino on stable bromine, has been used in the data reduction.²⁷ The effect of the uncertainty in g_J upon the final values of a and b was obtained by reducing the data with a slightly different value of g_J . The effect on the uncertainty vs a and b is negligible.

In order to check the internal consistency of the data, the α and β resonances ($\Delta F = 0$, $\Delta m = \pm 1$), and the $\Delta F = \pm 1$ resonances were run separately through the IBM program. The results are shown in Table II. The uncertainties quoted are those resulting directly from the calculation. The quantity g_I has been assumed positive.

Table II

Results of resonance computations for bromine-82		
Resonance group	a (Mc)	b (Mc)
All α, β	205.28 ± 0.31	-871.63 ± 1.98
All $\Delta F = \pm 1$	205.038 ± 0.007	-870.66 ± 0.18

The internal consistency of the data is well demonstrated.

No measurement appears to have been made of the magnetic interaction constant $a_{1/2}$ in the $P_{1/2}$ state of stable bromine. It is therefore difficult to say with certainty what the effect of configuration interaction is upon the present measurement of $a_{3/2}$ in Br^{82} . The effect, however, is probably quite small and has not been included as a correction. In addition, corrections to $a_{3/2}$ due to the finite size of the nuclear charge distribution²⁰ and magnetic-moment distribution²¹ are negligible for atoms in pure $P_{3/2}$ ground states and can be neglected. Perturbations of the $P_{3/2}$ ground state by admixture of the $P_{1/2}$ state do not affect the results significantly.

Magnetic Moment

We have, from Eq. (21);

$$\frac{a_{3/2}(\text{Br}^{82})}{a_{3/2}(\text{Br}^s)} = \frac{\mu^{82}}{\mu^s} \frac{I^s}{I^{82}}, \quad (21)$$

where μ^s is the nuclear magnetic moment and I^s the nuclear spin of the stable bromine isotope Br^s . Using the results of King and Jaccarino²⁷ for the $a_{3/2}$ values of Br^{79} and Br^{81} , together with the nuclear moments of the same isotopes given by Walchli,⁴⁶ we find for both comparison isotopes

$$\mu^{82} = \pm 1.6264(5) \text{ nm}, (\text{Br}^{79})$$

$$\mu^{82} = \pm 1.6264(5) \text{ nm}, (\text{Br}^{81}).$$

The difference between the two values of χ^2 listed above is not sufficient to permit a definite statement as to the sign of the magnetic

moment of Br^{82} . Consultation of the table of χ^2 in Ref. 47 shows that the data are fitted at better than the 95% level of significance with either choice of the sign of g_I .⁴⁷ The reason for this is simply that the resonance lines with the existing C-field magnet are not narrow enough to permit the discrimination required for the determination of the sign. This point is illustrated in Fig. 20. Several attempts were made to observe Br^{82} resonances at a field of 1464 gauss ($\nu_{\text{Cs}} = 800 \text{ Mc}$) with a narrow line, but all were unsuccessful. While a direct measurement of the sign must await improvement of the C field, there are two reasons for believing that the sign is positive:

(a) The magnetic moment of Rb^{82} has been measured to be $+1.50 \text{ nm}$.⁴⁸ The spin of Rb^{82} is 5, the same as Br^{82} , and it seems reasonable that isotopes with the same spin differing by pairs of neutrons and protons might have very nearly the same magnetic moment.¹⁵

(b) The magnetic moment calculated for Br^{82} on the basis of a simple jj-coupling model is positive.

The magnetic moment of an odd-odd nucleus using a single-particle description is given by the expression¹⁶

$$\mu = 1/2 \left[(g_p + g_n) I + (g_p - g_n) \frac{j_p(j_p + 1) - j_n(j_n + 1)}{(I + 1)} \right], \quad (33)$$

where g_p and g_n are the g factors of the odd neutron and proton, j_p and j_n are the proton and neutron angular momenta, and I is the nuclear spin. If the mean of the nuclear moments of Br^{79} and Br^{81} (2.188 nm) and the observed spins of $3/2$ are used to define an effective g_p for the proton part of the core of Br^{82} , we find $g_p = 1.459$. An effective g_n for the neutron part of the core can be obtained from the known spins and moments of the neighboring odd-A odd-N nuclei $^{34}_{43}\text{Se}^{77}$, $^{36}_{45}\text{Se}^{79}$ and $^{36}_{47}\text{Kr}^{83}$. Setting $I = 5$ and making use of Eq.(33), we obtain the following table:

MU-16565

Fig. 20. Plot showing the agreement of the bromine-82 observations with the two choices of sign for g_I .

Table III

Comparison of calculated Br^{82} magnetic moments with measured moment				
Isotope	Spin	Configuration	g_n	$\mu(\text{Br}^{82})_{\text{calc.}}$ (nm)
Se^{77}	1/2	$p_{1/2}$	1.068	6.42
Se^{79}	7/2	$(g_{9/2})^7_{7/2}$	-.291	1.17
Kr^{83}	9/2	$g_{9/2}$	-.215	0.18

$\mu(\text{Br}^{82})_{\text{obs}} = 1.63 \text{ nm}$

It is thus likely that the neutron part of Br^{82} does not couple to $J = 9/2$; coupling to $J = 7/2$ is a possibility. With the assignment $(g_{9/2})^7_{7/2}$, Nordheim's weak rule⁴⁹ applies and predicts for Br^{82} a nuclear spin of 5 or less. It is significant that for all reasonable choices of the neutron configuration the calculated magnetic moment is positive. This fact lends weight to the conclusion of the previous paragraph, i. e., that μ is positive.

Quadrupole Moment

The uncorrected quadrupole moment Q_e can be calculated from Eq. (19):

$$Q_e = 8/3 \left(\frac{\mu_0}{e} \right) \frac{m}{M} \left(\frac{\mu}{I} \right) \left(\frac{7}{2} \right) \frac{b}{a} . \quad (19)$$

Introducing the measured values of a and b and the value of μ obtained above, we find $Q_e = 0.728$ barn. In order to obtain the true nuclear quadrupole moment Q_c , a correction factor C such that $Q_e = CQ_c$ is introduced. This factor, first introduced by Sternheimer 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. The constant C has been calculated for bromine by Sternheimer¹⁷ but with the neglect of certain anti-shielding corrections. C is 1.040, but in view of the uncertainty associated with the exact value of C we have chosen to assign an uncertainty to Q_t equal to the value of the correction itself. Thus:

$$Q_t (\text{Br}^{82}) = 0.76 \pm 0.03 \text{ barn.}$$

If μ is positive, the sign of the quadrupole moment is also positive, since b/a is negative.

Q can be calculated, using an alternative procedure, by making use of the fine-structure separation δ to obtain a value of $\langle 1/r^3 \rangle$. The quantity δ is related to $\langle 1/r^3 \rangle$ by the expression²⁸

$$\delta = \frac{\mu_0^2}{hc} Z_i^2 (2l + 1) H \left\langle \frac{1}{r^3} \right\rangle . \quad (20)$$

Here Z_i is the effective value of the nuclear charge and $H(Z_i)$ is a relativistic correction factor tabulated in Ref. 30. Barnes and Smith³¹ have shown that $Z_i = (Z - n)$ approximately, n being the radial quantum number of the valence electron. For bromine we have $n = 4$ and $Z_i = 31$; the value of Q_e obtained by using $\langle 1/r^3 \rangle$

from Eq. (20) is 0.69 barn; and the corrected quadrupole moment Q_t is 0.72 barn. The uncertainty in this value of Q_t arises both from the uncertainty in Z_i and the uncertainty in the Sternheimer correction. An examination of the results of Barnes and Smith and a comparison with some similar calculations by Koster⁵⁰ indicate that Z_i is probably known to about 5%. Thus we have finally $Q_t = 0.72 \pm 0.06$ barn.

The agreement between these two methods of calculation is satisfactory.

Hyperfine Separations

The hyperfine separations in Br^{82} are easily calculated from Eq. (32).⁵¹ The results are

$$\Delta\nu (13/2, 11/2) = 766.82 \pm 0.60 \text{ Mc,}$$

$$\Delta\nu (11/2, 9/2) = 1287.32 \pm 0.43 \text{ Mc,}$$

$$\Delta\nu (9/2, 7/2) = 1488.6 \pm 1.1 \text{ Mc.}$$

A plot of these levels as a function of magnetic field is shown in Fig. 21.

MU-17271

Fig. 21. Hyperfine levels of bromine 82 as a function of magnetic field.

Bromine-80m, Bromine-80

Bromine-80m and -80 were produced by bombardment of KBr powder in a neutron flux of 2 to 5×10^{12} neutrons/cm²/sec. Bombardments were done at UCRL Livermore for about 3 hours and samples arrived at Berkeley approximately 1-1/2 hours after bombardment. About 3 grams of powder, sealed in quartz or Pyrex capsules, was used for irradiation, and no carrier was added during the chemical separation. Activity from these samples was sufficient for runs of 4 to 8 hours in duration.

Spins of 5 and 1 were observed, and representative resonances of each are exhibited as Figs. 22 and 23. Assignment of the spins to the respective isomers was made on the basis of their decay rates. A spin-1 decay is shown in Fig. 24. Negligible bromine-82 activity was observed in these samples.

Inasmuch as the 4.5-hour isomer decays into the 18-minute one, a continuous source of 18-minute bromine-80 is available in the vial, and this source decays with a 4.5-hour half life. At the time of writing, this is believed to be the shortest-half-life isotope that has been measured by atomic beams.

The decay scheme of this pair has an interesting consequence in regard to the counting of beam-collection buttons. Since the 4.5-hour bromine-80m decays by γ emission, to which our counters are quite insensitive, only the β decay of bromine-80 is detected. Therefore, when the radiofrequency is set on a bromine-80m transition frequency, a low initial counting rate would be expected which would rise as shown in Fig. 25, reaching its peak in about 80 minutes and thereafter decreasing with a 4.5-hour half life.

No estimate of the interaction constants a and b can be made from the present data on these isomers.

Considerable previous work had been done on bromine-80m and -80, a well known pair of isomers in the first "island of isomerism." In particular, indirect evidence indicated that the nuclear spins differed

Fig. 22. Bromine 80^m , 5(α) resonance at 10.9 gauss.

Fig. 23. Bromine 80m, 1 (α) resonance at 5.6 gauss.

Fig. 24. Bromine 80m, 1(α) exposure decay-plot.

Fig. 25. Calculated counting rate as a function of time for a bromine 80m ($I = 5$) exposure.

by 4 and were probably 5(-) and 1(+).^{52, 53} Briefly these conclusions, which were verified by the direct measurement described above, resulted from;

(a) Assignment of the bromine-80m 4.5-hour decay as M3 on the basis of lifetime and conversion-coefficient measurements.^{54, 55, 56}

(b) Conversion-coefficient measurements which assigned the 0.37-kev transition as E1.^{55, 56}

(c) Study of the shape and log ft values for the β^- and β^+ spectra of bromine-80 implied a spin 1(+).

(d) Angular correlation between conversion electrons of the 49- and 37-kev transitions which showed that the spin of the intermediate state could not be zero.

The decay scheme of these isomers is shown in Fig. 26.

A spin change of 4 between isomer and ground state is also expected here from the very closely spaced $g_{9/2}$ and $p_{1/2}$ levels in the shell model, and the respective values of 5 and 1 would be predicted from the measured spins of bromine-82 and -76.

Table IV

Bromine-80m and bromine-80 resonances

Resonance type	Frequency (Mc)	Resonance uncertainty (Mc)	H (gauss)	Ref. beam	Freq. (Mc)
1	6.25	0.18	5.57	K	4
1	7.65	0.20	5.57	K	4
1	12.3	0.35	10.87	K	8
5	2.4	0.20	5.57	K	4
5	4.8	0.18	10.87	K	8
5	2.72	0.09	10.87	K	8

MU-17590

Fig. 26. Decay scheme for bromine 80m, 80.

Bromine-77

Production of bromine-77 proceeds by an $\text{As}^{75}(\alpha, 2n)\text{Br}^{77}$ reaction. Details of the bombardments have been discussed in Section 4. Good production of bromine-76 is obtained with full energy from the external beam of the cyclotron. In order to minimize the 17-hour activity and enhance production by the $(\alpha, 2n)$ reaction, a beam of 8 Mev less than full energy was used for bombardments to produce bromine-77. An 8-Mev attenuation was selected as corresponding approximately to the energy difference between the $k = 3$ and $k = 2$ peaks on the available $\text{Br}(\alpha, kn)\text{Rb}$ excitation curves [Fig. 4 of Ref. (57)]. Negligible bromine-76 production has been observed on these targets.

$I = 3/2$ was expected for this isotope, since it differs by pairs of neutrons from bromine-79 and -81, whose spins were already known to be $3/2$ ⁽²⁾. This spin was established on the first run, and observations were made at four different fields. Figure 27 shows decays of both the full beam and the $I = 3/2$ buttons, giving in each case a pure 57-hour half life, within the probable errors, which is in agreement with the previously assigned half life. In Fig. 28 is shown the resonance from which the decay buttons were selected. The exposure used for the decay is marked by a square.

In the case in which $I = J = 3/2$, the two flop-in transitions occur at the same frequency in the low-field limit. The expression for g_F here reduces to

$$g_F \simeq g_J \frac{-I(I+1) + J(J+1) + F(F+1)}{2F(F+1)} = g_J \frac{+F(F+1)}{2F(F+1)},$$

since $I = J$. The terms in F cancel, so that we obtain $g_F \simeq +1/2 g_J = -2/3$ for all allowed values of F . Therefore both the flop-in transitions are coincident at low fields, giving twice the resonance height above background that would otherwise occur. A further consequence of the equality of the g_F 's is that the $\Delta F = \pm 1$, $\Delta m_F = 0$ transitions are field-independent to first order so that narrow lines will be expected for these transitions, as were observed for the stable bromine isotopes. ²⁷

Fig. 27. Bromine 77. Decay plot of full-beam and resonance exposures. (See Fig. 28).

Fig. 28. Bromine 77. α and β resonance at 20.3 gauss.
Exposure at 19.4 Mc, marked by square, was used for
Fig. 27 line B.

The last observation listed in Table V indicates a shift from linear dependence of resonance frequency upon H , but the data available are not sufficient to make a reliable estimate of a and b .

Table V

Bromine-77 resonances ^a					
Resonance type	Frequency (Mc)	Resonance uncertainty (Mc)	H (gauss)	Ref. beam	Freq. (Mc)
α, β	10.25	0.15	10.87	K	8
α, β	19.35	0.20	20.75	K	16
$\alpha?$	(45.6)	?	48.89	K	43
$\beta?$	(46.0) ^b				

^aVery-low-field spin-search exposures have been omitted.

^bAssignment of two peaks tentative. Data are needed at higher values of field to establish this.

Bromine-76

Low-field observations on bromine-76 established the spin as 1, and Fig. 29 shows both the α and β transitions well resolved at 2.86 gauss. All subsequent resonances have been consistent with a spin assignment of 1.

Figures 30 and 31 show an α and a β transition, respectively, obtained at higher values of H . At $H = 110.9$ gauss, the α resonance shows a shift from linear prediction of 4.4 Mc, while at only 17.1 gauss the β is shifted 3.1 Mc. Reference to Eq. (30) shows that for $I = 1$ the second-order perturbation term for the β transition vanishes so that the frequency shift should be small and vary as H^3 . Examination of the three β 's at 16.0, 23.9, and 38.7 gauss showed unexpectedly large shifts which were not consistent with a single hyperfine assignment, but showed a progressive shift of the apparent b/a value.⁵⁸

Preliminary values of b/a were about +0.6 and, as may be seen by Fig. 32, the two lower hyperfine levels invert at $b/a = 2/3$. If this occurs, the m_F values involved in the transition change so that the second-order term becomes nonzero. Furthermore, a third flop-in transition (in the $F = 1/2$ state) exists in this case and allows an immediate check to be made as to whether the levels are, in fact, reversed. A low-field search in the region corresponding to $g_F = 2.22$ verified the existence of the transition, designated γ in Table VI. The transition was observed at three values of H , of which the resonance obtained at 6.9 gauss is shown in Fig. 33. The γ transitions obtained showed very little shift from linearity. This was expected, since the second-order perturbation term is zero for the γ transition when the $F = 1/2$ level is above the $F = 3/2$ level.

Substitution of new m values corresponding to the hyperfine ordering 5/2, 1/2, 3/2 into the least-squares computer routine gave a good fit to the data. Observations of the α and β transitions were extended to fields of 265.7 and 64.6 gauss respectively, at which point a search for the direct $\Delta F = \pm 1$ transitions was made at low field. Figure 34 shows the transitions $F = 3/2$ to $F = 1/2$, giving

MU-15474

Fig. 29. Bromine 76. α and β resonances at 2.9 gauss.

Fig. 30. Bromine 76. α resonances at 111.0 gauss.

Fig. 31. Bromine 76. β resonances at 17.1 gauss.

MU-15559

Fig. 32. Plot of (b/a) vs (energy divided by a) for $I = 1$, $J = 3/2$.

Fig. 33. Bromine 76. α resonance at 6.9 gauss.

MU-17253

Fig. 34. Bromine 76. $\Delta\nu_{23}$ resonance at 19.6 gauss.

a hyperfine separation at zero field of -189.115 Mc. The $F = 5/2$ to $F = 3/2$ transition was obtained at two fields, of which the one obtained at 1.42 gauss is shown as Fig. 35. At zero field the $5/2$ to $3/2$ frequency is 1256.454 Mc.

All the resonances used in the least-squares computer calculation are shown in Table VI with their uncertainties. Low-field resonances giving no information on hyperfine structure have been omitted.

In summary, the measurements are:

$$I = 1 \quad \begin{aligned} (F=5/2 \text{ to } F=3/2) &= 1256.454 \pm 0.07 \text{ Mc,} \\ (F=3/2 \text{ to } F=1/2) &= -189.115 \pm 0.07 \text{ Mc,} \end{aligned}$$

where the indicated errors result from multiplying the calculated errors in a and b , as given by the computer, by 5. The relationships between the hyperfine levels and the a and b values are:

$$\begin{aligned} (5/2-3/2) &= 5/2 a + 5/4 b, \\ (3/2-1/2) &= 3/2 a - 9/4 b, \end{aligned}$$

as plotted in Fig. 32. The hyperfine levels, as a function of magnetic field are plotted in Fig. 36 for $b/a = 0.908$.

For input to the least-squares routine, $1/3$ of the resonance half widths were used. For all the resonances of Table VI, the results were:

$$\begin{aligned} a &= 345.417 \pm 0.014 \text{ Mc,} \\ b &= 314.329 \pm 0.022 \text{ Mc,} \\ b/a &= 0.91, \end{aligned}$$

where these values are the averages of those calculated for $g_I > 0$ and $g_I < 0$. For these two cases we have

$$\begin{aligned} \text{for } g_I > 0, \quad \chi^2 &= 4.0617, \\ \text{for } g_I < 0, \quad \chi^2 &= 3.5620. \end{aligned}$$

Since the values for χ^2 are so nearly equal, they give no reliable indication of the sign of g_I . Consistency of the data is indicated by Table VIII in which the first column indicates the input data to the computer.

MU-17254

Fig. 35. Bromine 76. $\Delta\nu_{12}$ resonance at 1.4 gauss.

From Eq. (21) we obtain

$$\frac{a_{3/2}(76)}{a_{3/2}(81)} = \frac{\mu(76)}{\mu(81)} \frac{I_{81}}{I_{76}} \quad (21)$$

From Eq. (21) the moment of bromine-76 becomes

$$\mu = 0.5480 (2) \text{ nm},$$

calculated by using the moment of bromine-81, and

$$\mu = 0.5480 (2) \text{ nm},$$

by using the moment of bromine-79.

Calculation of the magnetic moment from Eq. (17), where $\left\langle \frac{1}{r^3} \right\rangle = 0.365 \times 10^{26} \text{ cm}^{-3}$ from Eq. (20), and the values for δ and

Z_i contained in Ref. 31, gives

$$g_I = (2.906 \pm 0.105) \times 10^{-4}$$

and

$$\mu = 0.534 \pm 0.027 \text{ nm},$$

which is in reasonable agreement with the above values.

By Eq. (19) the quadrupole moment for bromine-76 is

$$Q = \frac{8}{3} \frac{\mu_0}{e} \frac{m}{M} \frac{\mu}{I} \frac{b}{a} = 0.261 \times 10^{-24} \text{ cm}^2.$$

Multiplying by the Sternheimer correction ($C=1.04$) and assigning 4% uncertainty, as for bromine-82 we obtain

$$Q_e = cQ = (0.27 \pm 0.01) \times 10^{-24} \text{ cm}^2.$$

Direct calculation of the uncorrected quadrupole moment from Eqs. (18) and (19) gives

$$Q = (0.26 \pm 0.02) \times 10^{-24} \text{ cm}^2$$

and

$$Q_e = (0.27 \pm 0.03) \times 10^{-24} \text{ cm}^2,$$

MU-17402

Fig. 36. Hyperfine levels of bromine 76 as a function of magnetic field.

where the uncertainties have been assigned in the same way as for bromine-82. From the expression $Q/Q' = b/b'$ for comparison with bromine-79 and -81, the values obtained are 0.270 and 0.274 barn, respectively, so all calculations give consistent results.

Here a and b have the same sign; therefore μ and Q must have opposite signs. The spins of the even-neutron bromine isotopes 81, 79, and 77 are all $3/2$, and from the positive Q values of 79 and 81, the proton configuration is assigned as $(p_{3/2})^{-1}_{3/2}$. The consistent appearance of $l_p = 3/2$ makes this assignment the logical one to assume for the protons of bromine-76.

In the case of the neutron levels in the region of 41, the situation is not so clear-cut. For reference, the behavior of the odd-neutron even-proton isotopes with neutron numbers in the $p_{1/2}$, $g_{9/2}$ region is given in Table VIII.

In terms of the shell model, the level $g_{9/2}$ is only slightly above the $p_{1/2}$ level; they differ by 4 units of angular momentum and are responsible for the first "island of isomers" (see Fig. 37).

Filling of the levels in order would put the 41st neutron in the $g_{9/2}$ level, as with Ge_{41}^{73} , where a spin of $9/2$ is encountered. Because this assignment would restrict the spin of bromine-76 to values between 3 and 6, it is clearly ruled out.

Pairing, however, occurs in Se_{43}^{75} , giving $I = 1/2$. From this configuration Nordheim's "strong rule" would predict the observed value of 1 for bromine-76. Calculation of μ on the basis of a $p_{1/2}$ state for the odd neutron gives a magnetic moment about $3-1/2$ times that observed, and the calculated Q has the same sign as μ . Since the two signs are known to be opposite, this configuration seems unlikely.

In a number of cases, nucleons couple to a J which is one less than the individual nucleon j . This seems to be the case in bromine-82 and also appears in Se_{45}^{79} , but this is inconsistent with the measured spin in bromine-76.

The observed spin of $5/2$ for Se_{41}^{75} is, however, most extraordinary and leads one to conclude that the 41st neutron is in

the $f_{5/2}$ state, which should have been filled with the 38th neutron and which should not exhibit holes due to pairing above it. This neutron state is consistent with the measured spin of bromine-76 and has interesting implications.

If we assume $(f_{5/2})^{-1}_{5/2}$ for the neutron configuration, g_n is 0.218, as calculated in Ref. 59. Then by using $g_p = 1.459$, derived from the stable bromine isotopes, we find the calculated μ for bromine-76 is -0.72 nm, which is in good agreement with the magnitude of the observed value. Moreover, a positive Q results from application of Eq. 9.6 of Ref. 37.

The very large quadrupole moment of selenium-75 suggests a deformed-core effect, and it is interesting to investigate the consequences of this consideration. From Eq. 17 of the paper by Nilsson,⁶⁰ the intrinsic quadrupole moment of bromine-76 may be calculated. For $I = 1$ we have

$$Q = \frac{I(2I-1)}{(I+1)(2I+3)} Q_0$$

and

$$Q_0 = \frac{1}{10} Q_0,$$

so that $Q_0 = 2.7$ barns. Comparison with $Q_{Se} = \frac{10}{28} Q_0 = 1.1$ gives $Q_{0, Se} = 3.1$, which is reasonably close to the value calculated for bromine-76.

Calculation of the deformation parameter, δ , to second order by Eq. (16) of Ref. 6,

$$\left[Q_0 \approx 0.8ZR_z^2 \delta \left(1 + \frac{2}{3} \delta\right) = 0.8Z(1.2 \times 10^{-13} A^{1/3})^2 \delta \left(1 + \frac{2}{3} \delta\right) \right],$$

gives $\delta = +0.3$. Reference to the levels calculated by Nilsson as a function of δ (Fig. 5 of Ref. 6) predicts that the 41st particle should be in an $f_{5/2}$ level, which is in agreement with the observations on both bromine-76 and selenium-75.

It appears then that the nuclear distortion is larger than the fluctuations present, that the observations cannot be fitted in terms of the simple shell model, and that a distorted core must be assumed. It is unfortunate that the magnetic moment of selenium-75 is unknown, as it would give further insight into the situation. Also the values of the moments for bromine-80 should be of interest as it also has a spin of 1.

Table VI.

Bromine-76 resonances						
Resonance type	Frequency (Mc)	Compounded uncertainty (Mc)	Ref. beam	Freq. (Mc)	H (gauss)	Weighting factor ω
β	16.95	0.01909	Cs	4	11.40	52.38
β	26.3	0.0482	Cs	6	17.07	20.74
α	64.25	0.02346	Cs	20	56.31	42.63
α	128.6	0.02403	Cs	40	110.95	41.61
α	325.7	0.01736	Cs	100	265.74	57.6
β	44.9	0.02886	K	22	27.63	34.65
γ	41.6	0.03561	K	10	13.42	28.08
γ	86.5	0.13176	Cs	10	28.37	7.59
γ	86.5	0.15846	Cs	10	28.37	6.31
β	78.6	0.26542	Cs	16	45.18	3.77
β	78.7	0.00822	Cs	16	45.18	11.34
β	118.8	0.11352	Cs	23	64.61	8.81
$\Delta\nu_{23}$	213.6	0.00732	K	15	19.57	136.61
$\Delta\nu_{12}$	1254.75	0.00282	K	1	11.42	354.61
$\Delta\nu_{12}$	1248.36	0.00438	K	5	6.92	228.31

Table VII

Results of resonance computations for bromine-76			
Resonances	g_I	a (Mc)	b (Mc)
All ($\alpha, \beta, \gamma, \Delta v$)	+	345.418 ± 0.014	314.327 ± 0.022
	-	345.416 ± 0.014	314.331 ± 0.022
All $\Delta F = \pm 1$	+	345.418 ± 0.014	314.327 ± 0.022
	-	345.416 ± 0.014	314.332 ± 0.022
$\alpha \beta \gamma$ only	+	344.344 ± 1.05	312.840 ± 0.90
	-	345.044 ± 1.06	313.288 ± 0.91

Table VIII

Observed nucleon data for isotopes of odd-n from 41 to 47			
Isotope	I	μ (nm)	Q (barn)
Ge ₄₁	9/2	-0.877	-0.2
Se ₄₁	5/2	?	+1.1
Se ₄₃	1/2	0.534	
Se ₄₅	7/2	-1.0	+0.9
Kr ₄₇	9/2	-1.0	+0.22

VI. SUMMARY

A summary of the results for bromine isotopes obtained by atomic beams is shown in Table IX. Data obtained by King and Jaccarino on the stable isotopes of bromine are included for completeness.

Table IX

Bromine atomic-beam results						
Isotope	Half life	I	a (Mc)	b (Mc)	μ (nm)	Q (barn)
$^{82}_{47}\text{Br}$	34 hr	5	± 205.04 (5)	∓ 870.66 (90)	1.6264 (5)	0.76 (3)
$^{81}_{46}\text{Br}$	stable	3/2	∓ 953.770 (3)	∓ 321.516 (8)	2.26958 (3)	0.28 (2)
$^{80}_{45}\text{Br}$	4.5 hr	5				
$^{80}_{45}\text{Br}$	18 min	1				
$^{79}_{44}\text{Br}$	stable	3/2	$+884.810$ (3)	-384.878 (8)	2.10555 (30)	0.33 (2)
$^{77}_{42}\text{Br}$	57 hr	3/2				
$^{76}_{41}\text{Br}$	17 hr	1	± 345.42 (7)	± 314.33 (11)	± 0.5480 (2)	∓ 0.27 (2)

Proton and neutron levels

4						
				1 g 9/2	[10]	(50)
				2 p 1/2	[2]	(40)
3				1 f 5/2	[6]	(38)
				2 p 3/2	[4]	(32)
3						
				1 f 7/2	[8]	(28)

This section presents a summary of some of earlier experimental work on stable bromine. Most of the results were obtained by optical or microwave spectroscopy, but some of molecular-beam determinations, not previously mentioned, are also included. In all cases the years listed are those of publication, and papers of the same year are not necessarily in order of their publication during that year.

As a starting point, references from R. Bacher, Atomic Energy States from Optical Spectra and C. Moore, Atomic Energy Levels were consulted, but many references led to others, so that no single reference list can be quoted. In the following, several useful bibliographies will be listed explicitly. No information on radioactive isotopes has been included because of space limitations. Publications dealing with unstable isotopes may be most easily located by consulting the Table of Isotopes compiled by Strominger, Hollander, and Seaborg [Revs. Modern Phys. 30, 585 (1958)].

Before 1920

According to Kimura (see 1920), Berndt attempted to observe the Zeeman effect in bromine, but failed as a result of insufficient intensity from his source. Following Berndt's work in 1902, work was done by Nutting in 1904, and Goldstein in 1907, but apparently without great success.

1920: Isotopy of Bromine. First Extensive Spectra

Aston, using a methyl bromide beam in his mass spectrometer, discovered the existence of two almost equally abundant isotopes of bromine: 79 and 81. This was regarded as a surprise, since the combining weight of bromine was almost equal to 80.

Kimura observed the band spectra and lines of both the arc and spark spectra. He tabulated the lines but gave no interpretation.

In Part II of the same paper he studied some complex lines at $\lambda = 6632, 6560, 6351, \text{ and } 6150 \text{ \AA}$ and found that they all gave triplets in a strong magnetic field. He also noticed that the Zeeman effect on the single lines gave triplets.

1925

Ruark and Chenault attempted to interpret the measurements of Kimura. Their conclusions were that the fine structure was not due to isotopy or the effect of a nuclear moment. They claimed that the results might be explained by "slight quantized variations in the configuration of loosely bound underlying electron shells."

1926: Ionization potential, δ

Turner tabulated lines, obtained with a vacuum spectrograph, from $\lambda = 1230$ to $2050 \text{ \AA}$. By referring to a suggestion due to Pauli, that a hole in an electron shell should be equivalent to one electron outside a filled shell, Turner inferred that the halogens should exhibit a 2P ground state. He observed the recurring difference 3685 cm^{-1} in the lines and correctly attributed this to the fine-structure interval.

The longest line he observed was $1633.6 \text{ \AA}$, from which he assigned 8.01 ev for the "radiating potential" of bromine.

Nakamura observed quadruplets, triplets, and doublets but made no theoretical interpretations or derivation of series until a later paper of 1926. In the later paper he attempted to fit series from observed absorption edges by equations of the type

$\nu = an + bn^2 + \dots$, where n takes on values $0, 1, 2, \dots$, but obtained only poor agreement.

1927: Isotopic mass determination, screening constant

L. and E. Block observed 1100 lines between $\lambda = 6700$ and $2250 \text{ \AA}$ and remarked that 700 were new.

Aston, using a new mass spectrograph with an accuracy of 1 in 10,000, determined the masses of bromine-79 and -81 and their packing fractions to be

Br^{79}	78.929	-9.0	} parts in 10^4
Br^{81}	80.926	-8.6	

He again used methylene bromide for source material and concluded that the isotopic abundance of each bromine was the same within his accuracy of determination.

From the optical data of Turner and from x-ray spectra, Laporte obtained values of the screening constant s . The values given, and their origins, are:

From relativistic x-ray doublets, $s = 17.0$;

From optical data of Turner, $s = 18.13$,

where the x-ray value was calculated from the $N_{II}-N_{III}$ doublet.

1930: Ground state, δ confirmation, I, ionization potentials of Br II and Br III

Kiess and de Bruin classified many lines of bromine and assigned the configuration $4s^2 4p^5: ^2P_{3/2}$ as the ground state. On the basis of their assignments they confirmed the value of 3685 cm^{-1} as the 2P separation.

They refer to a bibliography of 88 papers on bromine in Kayser, Handbuch der Spektroskopie (1924) vols. V, VII.

Deb attempted to classify lines of BrII through Br V by using data from L. and E. Bloch, and Kayser. He obtained additional data from $\lambda = 4200$ to $7500 \text{ \AA}$ and assigned the ionization potentials

Br II = 19.1 ev,

Br III = 25.7 ev.

De Bruin, from observations of four hyperfine-structure components on four strong lines, assigned $I = 3/2$. No mention of the existence of two isotopes was made here. His findings were published in two journals, of which *Naturwissenschaften* is the more complete. Here he fitted the interval rule to his observations by assuming $J = 5/2$, $I = 3/2$.

1931: Mass redetermination and isotopic abundance

Aston gave redetermined mass values for the two isotopes which agreed with his previous values of 1927. By correcting to the chemical scale of mass, by 1.25 parts in 10,000, he gave

Atomic wt. of bromine = 79.9165 ± 0.002 ,
which agreed with the chemically determined value, for which no
reference was given.

Tolansky assigned $I = 3/2$ to both isotopes of bromine on the
basis that he observed no isotope effect in any lines except a small
mass shift in "at least one" line. His observations were of lines from
7500 to 4400 $\text{\AA}$. A more detailed paper describing the work appeared
in 1932 and contained a good theoretical discussion, giving considerable
insight into the 1932 viewpoint.

A review article in 1931 by Kronig and Frisch gave a table of
the moments and spins known at that time together with references.

1934

La Croute studied the Zeeman effect of bromine. He classified
26 lines of Br II and gave g values for 14 of them.

1935: Isotopic composition

Blewett determined the isotopic composition of the two bromine
isotopes to be

$$\frac{\text{Br}^{79}}{\text{Br}^{81}} = 0.975 \pm 0.025.$$

1937

Rao and Krishnamurty argued that the level assignments made
by Deb in 1930 were incorrect. This was a result of an error having
been found in his reference element's level assignment. These men
gave the ionization potential of Br III as 35.7 ev, which is 10 v too high.

1938: Packing fractions, magnetic moment

Schmidt, from Tolansky's work, obtained $^4\text{P}_{5/2} \delta = 495$,
 $^4\text{P}_{3/2} \delta = 252$, $^2\text{P}_{3/2} \delta = 210$, and calculated coupling constants from
theoretical expressions due to Breit and Wills. From these calculations

he assigned the magnetic moment as 2.6 nm and suggested that the values for both isotopes were about the same, since Tolansky saw little isotope effect of any kind. Also Schmidt gives a table of the known I and μ values and references.

Aston measured the packing fractions and isotopic masses:

Br^{79} -7.4 78.9417 ± 0.0020

Br^{81} -7.4 80.9400 ± 0.0020

1946: Isotopic composition

Williams and Yuster remeasured the isotopic composition to be

$$\frac{\text{Br}^{79}}{\text{Br}^{81}} = 0.979 \pm 0.004,$$

and, using the value of the packing fraction given by Aston in 1938, assigned the atomic weight to be 79.908. This work set an upper limit of one part in 10,000 to the abundance of the isotopes 78, 80, and 82 relative to the abundance of bromine-79.

1947: Q couplings, magnetic moments

Townes, Holden, Bardeen, and Merritt confirmed $I = 3/2$ for both isotopes, determined that Q was positive, and gave, for quadrupole coupling constants

Br^{79} $720 \pm 10 \text{ Mc}$,

Br^{81} $556 \pm 10 \text{ Mc}$

by absorption measurements on BrCN . These values were determined for the $J = 2$ to $J = 3$ transition.

By a molecular-beam experiment on CsBr and LiBr , Brody, Nierenberg, and Ramsey assigned magnetic moments of 2.110 ± 0.021 and $2.271 \pm 0.023 \text{ nm}$, but were not able to detect which isotope was associated with which moment.

Gordy, Smith, Smith, and Ring observed the $J = 3$ to $J = 4$ transition in BrCN and calculated the bond stretching from excitation. From the effect on their lines they assigned "large" Q and, for the coupling constants measured,

$$\text{Br}^{79} \quad 567 \pm 15 \text{ Mc,}$$

$$\text{Br}^{81} \quad 670 \pm 15 \text{ Mc,}$$

Pound saw lines in aqueous solutions of LiBr and NaBr by nuclear paramagnetic absorption. At a field of 3000 gauss he obtained lines of different widths for bromine-79 and -81, therefore was able to assign the smaller Q to bromine-81.

Using Na^{23} as an intermediate standard with $\mu(\text{Na}^{23}) = 2.217 \pm 0.002 \text{ nm}$, and his measured frequency ratios,

$$\frac{\nu(81)}{\nu(79)} = 1.0778 \pm 0.0003,$$

$$\frac{\nu(79)}{\nu(23)} = 1.0209 \pm 0.0003,$$

he assigned

$$\mu(81) = 2.2706 \pm 0.003,$$

$$\mu(79) = 2.1066 \pm 0.003.$$

1948: Quadrupole moment

Gordy, Simmons, and Smith looked at rotational levels of CH_3Br and gave

$$Q(79)/Q(81) = 1.197.$$

Smith, Ring, Smith, and Gordy gave, for the quadrupole coupling in BrCN ($J = 2$ to 3), $\text{Br}(79) = 686 \text{ Mc}$,
 $\text{Br}(81) = 573 \text{ Mc}$.

In a review article, Gordy, using the data above, quotes actual values of Q (in barns) as follows:

	<u>a</u>	<u>b</u>
Br ⁷⁹	0.28	0.24
Br ⁸¹	0.23	0.19

where column a results from the Smith, Ring, Smith, and Gordy paper and a private communication from Townes, Holden, and Merritt, and column b results from data by Gordy, Simmons, and Smith (above). These seem to be the first values for Q given specifically. References are given to theories regarding the evaluation of $\partial^2 V / \partial Z^2$, but they make no statement as to how the calculation was done.

1949

Bitter quoted the magnetic-resonance frequency of bromine-81 in relation to that of hydrogen to be $\text{Br}/\text{H} = 0.27003 \pm 0.03\%$.

In a publication later in the year Zimmerman and Williams compared bromine frequencies (NaBr solution) with protons in the same magnetic field and gave

$$\begin{array}{ll} \text{Br}^{79} & 0.25059 \pm 0.02\% , \\ \text{Br}^{81} & 0.27014 \pm 0.02\% . \end{array}$$

1951

Dehmelt and Kruger, using polycrystalline methyl bromide at 83°K, saw quadrupole resonances and assigned the ratio of the quadrupole moments to be

$$Q_{79}/Q_{81} = 1.1973 \pm 0.0006.$$

Kuiper and van Zoonen assigned an upper limit to the ionization potential of bromine as less than 11.6 ev.

1951

By NMR Sheriff and Williams determined the magnetic moments to be

$$\begin{array}{l} \mu(79) = 2.10574 \pm 0.00010, \\ \mu(81) = 2.26947 \pm 0.00013 \text{ nm.} \end{array}$$

These measurements were made on NaBr solutions and used Sc^{45} as an intermediate standard. The quoted results include a diamagnetic correction ($\times 1.00308$) and are relative to the proton magnetic moment, which has been taken as 2.79255 ± 0.00010 nm.

1953

Honig, Stitch, and Mandel observed the $J = 9$ to $J = 10$, 10 to 11 , and 11 to 12 transitions in CsBr and calculated the mass ratio of the two isotopes to be

$$\frac{M(79)}{M(81)} = 0.9753068 \pm 0.0000045.$$

By molecular beams Fabricand, Carlson, Lee, and Rabi, from measurements on KBr, assigned

$$\frac{M(79)}{M(81)} = 0.9853088 \pm 0.0000020.$$

1954

Kuiper and van Zoonen improved their 1953 work and assigned ionization potential limits for bromine (Br_2) as greater than 9.91 eV and less than 11.55 eV.

Honig, Mandel, Stitch, and Townes redetermined the mass ratio of the two isotopes to be

$$\frac{M(79)}{M(81)} = 0.9752999 \pm 0.0000065.$$

1955

Cameron and Lippert used bromine compounds from various geographical areas to determine if there was any variation in isotopic composition with area. They found no significant variation and their average ratio was 1.0217 ± 0.0002 , which they concluded was reasonable agreement with the earlier assignment by Williams and Yuster.

Reference List of Section VI

1902 Berndt	Ann. Physik (4) <u>8</u> , 625 (1902).
1904 Nutting	Astrophys. J. <u>19</u> , 239 (1904).
1907 Goldstein	Verhandl. deut. physik. Ges. <u>9</u> ,
321, (1907).
1920 Aston	Phil. Mag. <u>40</u> , 628 (1920).
Kimura	Mem. Coll. Sci. Univ. Kyoto <u>4</u> , 127
(1920); <u>4</u> , 139 (1920).
1925 Ruark and Chenault	Phil. Mag. <u>50</u> , 937 (1925).
1926 Turner	Phys. Rev. <u>27</u> , 400 (1926).
Nakamura	Mem. Coll. Sci. Univ. Kyoto <u>9</u> , 335
(1926).
1927 Bloch and Bloch	Ann. Phys. <u>7</u> , 205 (1927).
Aston	Proc. Roy. Soc. (London) <u>A115</u> ,
487 (1927).
Laporte	Phys. Rev. <u>29</u> , 650 (1927).
1930 Kiess and de Bruin	J. Research Natl. Bur. Standards <u>4</u> ,
667 (1930).
Deb	Proc. Roy. Soc. (London) <u>A127</u> , 197
(1930).
de Bruin	Nature <u>125</u> , 414 (1930).
de Bruin	Naturwiss. <u>18</u> , 265 (1930).
1931 Aston	Proc. Roy. Soc. (London) <u>A132</u> , 487
(1931).
Tolansky	Nature <u>127</u> , 855 (1931).
Kronig and Frisch	Physik. Z. <u>32</u> , 457.
1932 Tolansky	Proc. Roy. Soc. (London) <u>A136</u> , 585
(1932).
1934 La Croute	Ann. Phys. <u>3</u> , 5 (1934).
1935 Blewett	Phys. Rev. <u>49</u> , 900 (1935).
1937 Rao and Krishnamurty	Proc. Roy. Soc. (London) <u>A161</u> ,
38 (1937).
1938 Schmidt	Z. Physik <u>108</u> , 408 (1938).
Aston	Nature <u>141</u> , 1096 (1938).

- 1946 Williams and Yuster Phys. Rev. 69, 556 (1946).
- 1947 Townes, Holden Bardeen,
 and Merritt Phys. Rev. 71, 644 (1947).
- Brody, Nierenberg, and
 Ramsey Phys. Rev. 72, 258 (1947).
- Gordy, Smith, Smith,
 and Ring Phys. Rev. 72, 259 (1947).
- Pound Phys. Rev. 72, 1273 (1947).
- 1948 Gordy, Simmons, and
 Smith Phys. Rev. 74, 243 (1948).
- Smith, Ring, Smith,
 and Gordy Phys. Rev. 74, 370 (1948).
- Gordy Revs. Modern Phys. 20, 668 (1948).
- 1949 Bitter Phys. Rev. 75, 1326 (1949).
- Zimmerman and Williams Phys. Rev. 76, 350 (1949).
- 1951 Dehmelt and Kurger Z. Physik. 129, 401 (1951).
- Sheriff and Williams Phys. Rev. 82, 651 (1951).
- 1953 Honig, Stith, and
 Mandel Phys. Rev. 92, 901 (1953).
- Fabricand, Carlson,
 Lee and Rabi Phys. Rev. 91, 1403 (1953).
- Kuiper and van Zoonan Appl. Sci. Research B3, 390 (1953).
- 1954 Kuiper and van Zoonan Appl. Sci. Research B4, 235 (1954).
- Honig, Mandel, Stith,
 and Townes Phys. Rev. 96, 629 (1954).
- 1955 Cameron and Lippert Science 121, 136 (1955).

ACKNOWLEDGMENTS

The role of graduate-student-wife is never easy and often impossible. Consequently, any acknowledgment is inadequate and I can only say that my appreciation for my wife's help and encouragement is unbounded.

It is so impossible to adequately express my appreciation to my parents that any attempt here is unfeasible.

Professor William A. Nierenberg developed the computer programs used in processing the experimental data. Many thanks are due him for his enthusiasm, theoretical discussions, and efficient direction of the beam group as a whole, all of which have contributed greatly to the advancement of the research.

Dr. Edgar Lipworth designed and constructed the atomic-beam apparatus on which the work was done. It has been a pleasure to work daily with him for almost two years, and his good humor, patience, and sound knowledge of physics have been greatly appreciated.

Many helpful discussions of both theory and experimental practice have been contributed by Professor K. Smith during his year's visit. In particular, the design of the valve between the oven can and A magnet was suggested by him.

I would also like to acknowledge a very illuminating discussion with Visiting Professor B. Mottelson regarding the interpretations of the results on bromine-76.

Mr. Doug Macdonald's services to the group have been both universal and expert.

Henry Lancaster designed and developed the transistorized magnet regulator shown in Section III.

My relations with the glass shop have been mainly with Mr. H. Powell, who capably directs the shop, and with Mr. W. Berland. They have both made valuable design suggestions and constructed the leaks and other glassware used in this work.

The available assistance at the Lawrence Radiation Laboratory is so great that it is not possible to mention individually all those whose help contributed to the advancement of the experimental work. This state of affairs can, perhaps, be fully appreciated only by one who has done previous work on a do-it-yourself basis.

In conclusion, I would like to thank the other students in the atomic-beam group for their assistance and discussions.

This work was performed under the auspices of the U. S. Atomic Energy Commission.

REFERENCES AND NOTES

1. Garvin, Green, and Lipworth Bull. Am. Phys. Soc. 2, 344, (1957).
2. Green, Garvin, Lipworth, and Nierenberg, Bull. Am. Phys. Soc. 2, 383 (1957).
3. Green, Garvin, and Lipworth, Bull. Am. Phys. Soc. 3, 318 (1958).
4. Garvin, Green, Lipworth, and Nierenberg, Phys. Rev. Letters 1, 293 (1958).
5. Lipworth, Garvin, and Green, Bull. Am. Phys. Soc. 4, 11 (1959).
6. Green, Garvin, Lipworth, and Smith, Bull. Am. Phys. Soc. 4, 250 (1959).
7. Garvin, Green, Lipworth, and Nierenberg, Hyperfine Structure and Nuclear Moments of Bromine-82, UCRL-8680, March 9, 1959 (to be published in Phys. Rev.).
8. Green, Lipworth, and Nierenberg, Hyperfine Structure and Nuclear Moments of Bromine-76 (in preparation for Phys. Rev.)
9. Garvin, Green and Lipworth, Phys. Rev. 111, 534 (1958).
10. Garvin, Green, and Lipworth, Bull. Am. Phys. Soc. 2, 383 (1957).
11. Garvin, Green, Lipworth, and Nierenberg, Phys. Rev. Letters 1, 74 (1958); Bull. Am. Phys. Soc. 3, 319 (1958).
12. Garvin, Green, and Lipworth, Phys. Rev. Letters 1, 292 (1958); Bull. Am. Phys. Soc. 3, 370 (1958).
13. G. E. Uhlenbeck and S. A. Goudsmit, Naturwiss. 13, 953 (1925); Nature 117, 264 (1926).
14. According to H. E. White [Introduction to Atomic Spectra (McGraw-Hill, New York, 1934), p. 353] this suggestion was due to Pauli [(Naturwiss. 12, 741, (1924))] and, independently, H. N. Russell.
15. S. A. Goudsmit and E. Back, Z. Physik. 43, 321 (1927); 47, 174 (1928).
16. M. G. Mayer and J. H. D. Jenson, Elementary Theory of Nuclear Shell Structure (Wiley, New York, 1955).

17. R. Sternheimer, Phys. Rev. 86, 316 (1952); 95, 736 (1954).
18. Bartlett, Nature 128, 408 (1931).
19. G. Breit and J. Rosenthal, Phys. Rev. 41, 459 (1932).
20. M. F. Crawford and A. L. Schawlow, Phys. Rev. 76, 1310 (1949).
21. A. Bohr and V. F. Weisskopf, Phys. Rev. 77, 94 (1950).
22. C. Schwartz, Phys. Rev. 97, 380 (1955).
23. H. A. Bethe, Elementary Nuclear Theory (Wiley, New York, 1947).
24. N. F. Ramsey, Molecular Beams (Oxford University Press, Oxford 1955),
25. This development follows that of H. G. Dehmelt, Am. J. Phys. 22, 110 (1954), except for some details.
26. N. F. Ramsey, Nuclear Moments (Wiley, New York, 1953).
27. J. G. King and V. Jaccarino, Phys. Rev. 94, 1610 (1954).
28. H. B. G. Casimir, On the Interaction Between Atomic Nuclei and Electrons (Teylers Tweede Genootschap, Haarlem, 1936).
29. Davis, Feld, Zabel, and Zacharias, Phys. Rev. 76, 1076 (1949).
30. H. Kopfermann, Nuclear Moments, E. E. Schneider Trans. (Academic Press, New York, 1958).
31. R. G. Barnes and W. V. Smith, Phys. Rev. 93, 95 (1954).
32. Each state should be labeled by a complete set of observables, but here all those labels not entering the calculation have been dropped for convenience in notation.
33. The full beam is that beam falling on the detector with the deflecting fields switched off and the stop wire withdrawn.
34. If a were very small, the dependence of the resonance frequency upon magnetic field would be observably nonlinear, even at low field. So, in actuality, the observations referred to exclude this possibility and it is not strictly true that no information is obtained concerning the value of a .
35. E. Feenberg, Shell Theory of the Nucleus (Princeton University Press, Princeton, 1955).
36. Elliott and Lane, Handbuch der Physik. 39, 241 (1957).
37. R. J. Blin-Stoyle, Theories of Nuclear Moments (Oxford University Press, Oxford, 1957); Revs. Modern Phys. 28, 75 (1956).

38. Lemonik, Pipkin, and Hamilton, Rev. Sci. Instr. 26, 1112 (1955);
I. Lindgren, Nuclear Instr. 3, 1 (1958).
39. G. O. Brink, Nuclear Spins of Thallium-197, Thallium-198m,
Thallium-199, and Thallium-204, (Thesis) UCRL-3642, June
1957.
40. See Acknowledgments.
41. J. G. King and V. Jaccarino (Phys. Rev. 94, 1610 (1954)) used a
combination of temperature control and leak, but for radioactive
work the combination seemed to add more complication than
would be desirable.
42. S. A. Gordon, Rev. Sci. Instr. 29, 501 (1958).
43. Alcohol and dry ice are a convenient combination as they will
completely evaporate in a reasonably short time and therefore
need not be cleaned up in event of spillage. This is advantageous
during a run as the high radiation field and possibility of breaking
the glassware make extra manipulations in the glove box very
undesirable.
44. S. Wexler, Res. Modern Phys. 30, 402 (1958).
45. Clary Corporation, 408 Junipero, San Gabriel, California.
46. H. E. Walchi, A Table of Nuclear Moment Data, ORNL-469,
Suppl. II, Feb. 1 1955.
47. R. A. Fisher, Statistical Methods for Research Worker, (Hafner,
New York, 1954).
48. Hubbs, Nierenberg, Shugart, Silsbee, and Sunderland, Phys. Rev.
107, 723 (1957).
49. L. W. Nordheim, Revs. Modern Phys. 23, 322 (1951).
50. G. F. Koster, Phys. Rev. 86, 148 (1952).
51. For bromine-82, Eq. (32) becomes
$$\begin{aligned} [F = (13/2 - 11/2)] &= 13/2 a + 351/540 b \\ (F = 11/2 - 9/2) &= 11/2 a - 99/540 b \\ F = 9/2 - 7/2 &= 9/2 a - 351/540 b. \end{aligned}$$
52. M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 179 (1952).
53. E. Segré and A. C. Helmholtz, Rev. Modern Phys. 21, 271 (1949).

54. A. Berthelot, Ann. Physik. 19, 219 (1944).
55. R. Rothwell and D. West, Proc. Phys. Soc., (London) 63A, 539 (1950).
56. Lidovsky, Macklin, and Wu, Phys. Rev. 78, 318A (1950).
57. W. A. Nierenberg, Ann. Rev. Nuclear Sci. 7, p. 349, (1957).
58. The third-order perturbation theory expression is

$$\delta \nu_{\beta}^{(3)} = \nu_0^3 \left\{ \frac{-0.1217}{(\Delta \nu_{12})^2} + \frac{1.316}{(\Delta \nu_{23})^2} \right\}$$

59. Phys. Rev. 98, 1224 (1955).
60. S. G. Nilsson, Binding States of Individual Nucleons in Strongly Deformed Nuclei, Kgl. Danske Videnskab. Selskab Mat.-fys. Medd. 29, No. 16 (1955).

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.