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UNIVERSITY OF CALIFORNIA

Radiation Laboratory
Berkeley, California

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SPIN AND HYPERFINE STRUCTURE MEASUREMENTS
OF TWO NEUTRON-DEFICIENT GALLIUM ISOTOPES

John L. Worcester

(Thesis)

July 15, 1957

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SPIN AND HYPERFINE STRUCTURE MEASUREMENTS
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SPIN AND HYPERFINE STRUCTURE MEASUREMENTS OF TWO NEUTRON-DEFICIENT GALLIUM ISOTOPES

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July 15, 1957

ABSTRACT

The spins of two neutron-deficient radioactive gallium isotopes have been measured by the atomic-beam resonance method. They are

\[ I = 0 \text{ for gallium-66, } T_{1/2} = 9.4 \text{ hr, and} \]

\[ I = 3/2 \text{ for gallium-67, } T_{1/2} = 78 \text{ hr.} \]

The hyperfine structure constants, the magnetic dipole, and electric quadrupole moments of gallium-67 in the \(^2P_{3/2}\) electronic state are measured to be

\[ \Delta \nu_{32} = 597.2 \pm 1.3 \text{ Mc}, \]

\[ \Delta \nu_{21} = 278.8 \pm 2.0 \text{ Mc}, \]

\[ \mu_I = 1.84 \pm 0.01 \text{ nm, and} \]

\[ Q = 0.213 \pm 0.006 \times 10^{-24} \text{ cm}^2. \]

The direct transitions were not observed, but rather the hyperfine structures were determined by observing the transitions \( F = 3, m_F = -1 \leftrightarrow -2 \) and \( F = 2, m_F = 0 \leftrightarrow -1 \). The assignment of a positive sign to both the dipole and quadrupole moments is more consistent with the observed transition frequencies.
INTRODUCTION

Atomic Spectra

Atomic emission and absorption spectra can be interpreted very satisfactorily as the reorientation of atomic electrons in the quantized energy levels of the atom. Upon close examination, these spectral lines are found to be split into components whose energies are separated by as little as tenths of inverse centimeters. This "fine structure" splitting can be explained by assuming that electrons have a characteristic that can be interpreted classically as a rotation, or spin. Because of the mass and charge distribution of the "spinning electron", it will have an associated intrinsic angular momentum and magnetic moment.

With the advent of instruments of greater resolving power, some of these lines were found to be further split into components separated by as little as hundredths of inverse centimeters. This "hyperfine structure" splitting is explained by assuming that the nucleus of the atom also has spin and associated electric and magnetic multipole moments. In certain cases there may be electric and magnetic fields at the nucleus due to the electrons. It is the interaction of the nuclear moments with these electronic fields that gives the hyperfine structure splitting.
Atomic-Beam Spectroscopy

Atomic beam techniques provide excellent tools for observing the modification of the atomic energy levels. A beam of atoms is passed through a region of inhomogeneous magnetic field. If an atom in the beam has a net magnetic moment when in the field, it will be deflected. The amount of deflection will depend upon the magnitude of the moment, the degree of inhomogeneity, and the time the atom spends in the field. Because the effective moment of the atom as a whole is a function of the field strength, the deflection of the atom is intimately and exactly related to its atomic and nuclear properties.

Zero-Moment Technique

The degree of coupling between the magnetic moment of the nucleus and the magnetic field of the electrons is a function of any externally applied magnetic field. For certain values of external field, the coupling may be altered so that there is no net magnetic moment. Because there are different magnetic quantum states in which an atom may exist, there are also different values of magnetic field at which these "zero moments" may be observed. The number of zero moments, and the magnetic fields at which they are observed, can be used to determine the spin and the magnetic moment of the nucleus.

Resonance-Type Techniques

Atoms in different magnetic quantum states usually have different effective moments. A beam of atoms will have a particular trajectory in an inhomogeneous field, depending upon which quantum state the atoms are in. If a perturbation is applied to the atoms while they are in transit, changing them from one state to another, the trajectory of the beam will also be changed.

These transitions can be induced by illuminating the beam with radiation, or by allowing the beam to pass through regions of perturbing magnetic or electric fields. The change in the trajectory of the beam can cause a change of intensity at a suitably placed detector. The geometry of the apparatus can be arranged so that the detector is exposed to all of the beam, as in the "flop-out" type of experiment.
With this arrangement, the intensity at the detector is decreased when the proper transition takes place. Or, for the "flop-in" type, the detector is not exposed to any part of the beam until the transition takes place.

In the laboratories of the atomic-beam group at the University of California at Berkeley, with apparatus located both on the campus proper and at the Radiation Laboratory, experiments have been performed using all three techniques; zero moment, flop-in, and flop-out. The purpose of this paper is two-fold: to describe in part some of the theoretical considerations associated with atomic beam measurements, and to discuss in greater detail a portion of the experimental work done by this author, \textsuperscript{1,2} using apparatus at both of the above locations.

**BASIC THEORY ASSOCIATED WITH ATOMIC BEAM MEASUREMENTS**

**Shell Model of the Nucleus**

The nucleus of an atom may be thought of as having a shell-like structure very similar to the atomic electrons. The particles in the nucleus are, however, \( Z \) protons and \((A-Z)\) neutrons. The filling of the nuclear shells is associated with the so-called "magic numbers": 2, 8, 14, 20, 50, 82, and 126. These magic numbers are suggested by the discontinuities observed in isotopic and isomeric abundances, quadrupole moments, nuclear binding energies, neutron-capture cross sections, and others.

In the nucleus there is no known core providing a central force field, as there is in the atom. It is assumed then that, to a first approximation, an individual nucleon moves in a nuclear central force field resulting from the combined effects of the other nucleons. In this central force field, the orbital angular momentum is quantized and restricted to integral values of \( \mathbf{n} \). The exact shape of the potential well is unknown. Simple well shapes, such as rectangular-or parabolic-shaped wells, fail to give by themselves the proper energy-level sequence as indicated by the magic numbers. However, with the assumption of other odd-shaped wells, or with various degrees of spin-orbit coupling, the level sequence can be adjusted to agree with
experiment. Such a level scheme is shown in Fig. 1. It may be noted that Mayer and Jensen suggest two slightly different degrees of spin-orbit coupling to obtain the proper level sequences for protons and neutrons. 3

Nuclear Spins and Magnetic Moments

Like the electron, the neutron and the proton have intrinsic spins of 1/2 \( \hbar \). The total angular momentum of the nucleus will be the proper vector sum of the orbital and intrinsic moments of all the nucleons. Both the neutrons and the protons have intrinsic magnetic moments, while the protons may also have orbital magnetic moments. The nucleus as a whole will therefore have a net magnetic moment that is the proper sum of the intrinsic moments of the nucleons plus the orbital moments of the protons. Fortunately, the above-mentioned "proper sums" usually simplify. Shell-model theory assumes that even numbers of nucleons in quantum levels of the same total angular momentum annul the effects of each other. Therefore the only contribution to the net angular momentum and magnetic moment is attributed to any odd nucleons that may be in these levels. This assumption is immediately supported by the fact that no even-even isotope in the ground state has been observed to have any spin other than zero.

The relation between the spin and the magnetic moment is defined by

\[
\mu_I = g_I \mu_0 \bar{T}
\]

where \( \mu_I \) is the magnetic moment, \( \bar{T} \) is the angular momentum measured in units of \( \hbar \), \( \mu_0 \) is either a Bohr or nuclear magneton, depending upon choice of definition, and \( g_I \) is the proportionality factor.

The shell model predicts the spins of nuclei fairly satisfactorily, but anomalies have been found. The model is unable to predict the exact g-values, although limits can be placed upon them. Again, anomalies have also been found for the magnetic moments, some measured g's even having the wrong sign.
Fig. 1. Shell-model level scheme. Number of identical nucleons allowed in a level and total number in closed shells indicated on the right.
Nuclear Multipole Moments

The nucleus may also exhibit higher-order electric and magnetic moments because of the distribution of charge and currents in the nucleus. For example, if the nuclear charge distribution is not spherically symmetric, the nucleus may possess an electric quadrupole moment. From simple parity considerations, magnetic multipole moments are limited to odd-order, and electric multipole moments are limited to even-order. It can also be shown that a nucleus can have a multipole moment of order \( n \) only for \( n \leq 2I \). Although a nucleus may have a multipole moment, it will not be observable unless there is also the appropriate higher-order derivative of field at the nucleus. Therefore, there will be a modification of the atomic energy levels only for \( n \leq 2J \), where \( J \) is the total angular momentum of the electrons measured in units of \( \hbar \).

Interaction of a Nuclear Magnetic Moment with an Atomic Magnetic Field

It now is apparent that there is a modification of the atomic energy levels due to the interaction of any of the nuclear properties with magnetic and electric fields at the nucleus. If it is assumed that the nucleus has only a magnetic moment, then, in the absence of any externally applied fields, the energy of interaction can be written as

\[
W = -\overline{\mu}_I \cdot \overline{H}_J \tag{2}
\]

where \( \overline{H}_J \) is the magnetic field at the nucleus due to the electrons. Because \( \overline{H}_J \) can be taken as proportional to \( J \) (with certain qualifications), the energy can then be written as

\[
W = \hbar \, a \, \overline{I} \cdot \overline{J}. \tag{3}
\]

Some authors do not use Planck's constant \( \hbar \) in the above definition. If it is used, as above, then the interaction constant \( a \) is measured in units of frequency.

An approximate value for a single non-\( s \)-electron has been derived by various authors using semi-classical considerations. The expression for \( a \) is of the form:
where \( c \) is the velocity of light, \( R \) is Rydberg's constant, \( a \) is the fine-structure constant, \( Z \) is the atomic number, \( n, L, \) and \( J \) are the quantum numbers describing the electron, and \( M/m \) is the ratio of the mass of the proton to the electron.

In the absence of external fields, \( I \) and \( J \) are tightly coupled, their orientation completely described by the resultant vector \( \overrightarrow{F} = \overrightarrow{I} + \overrightarrow{J} \). The energy can then be written as

\[
W = \hbar a \cdot \frac{F(F + 1) - J(J + 1) - I(I + 1)}{2}
\]

where \( F \) can take on the values \( |I - J| \leq F \leq |I + J| \). For \( I \leq J \), there are \( 2I + 1 \) possible orientations of \( \overrightarrow{I} \) and \( \overrightarrow{J} \), and therefore \( 2I + 1 \) energy levels. If a transition can be induced, changing \( F \) to \( F-1 \), the energy change is given by the well-known interval rule

\[
W_F - W_{F-1} = \hbar a F.
\]

The energy separation between adjacent hyperfine levels is proportional to the larger \( F \).

Interaction of a Nuclear and Atomic Moment with an External Magnetic Field

In the presence of a weak, externally applied magnetic field, \( \overrightarrow{F} \) can take on \( 2F + 1 \) possible orientations with the field. This space quantization of angular momentum was first demonstrated by the now familiar Stern and Gerlach experiment.
Zeeman Effect

In a weak magnetic field, there is a further modification of the energy levels, the Zeeman effect, which is due to the interaction of the magnetic moments of the nucleus and the atom with the externally applied magnetic field. $\mathbf{I}$ and $\mathbf{J}$ couple to a total angular momentum $\mathbf{F}$ (Fig. 2a). The vector, $\mathbf{F}$, precesses about $\mathbf{H}$ with the familiar Larmor rotational frequency. This extra energy due to precession can be written as

$$W = -\mu_{\text{eff}} \cdot \mathbf{H} = -\mu_{\text{eff}} \frac{m_F H}{F} \quad (7)$$

where $\mu_{\text{eff}}$ is the effective magnetic moment of the atom and $g_F$ is the effective g-value.

Because of the rapid precession of $\mathbf{I}$ and $\mathbf{J}$, only their time-averaged value along $\mathbf{F}$ contributes to the energy of interaction. We may then write

$$W = -g_J \mu_0 \frac{\mathbf{J} \cdot \mathbf{F}}{F} \frac{\mathbf{F} \cdot \mathbf{H}}{F} - g_I \mu_0 \frac{\mathbf{I} \cdot \mathbf{F}}{F} \frac{\mathbf{F} \cdot \mathbf{H}}{F} \quad (8)$$

Comparing the two expressions we see that

$$g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} + g_I \frac{F(F+1) + I(I+1) - J(J+1)}{2F(F+1)} \quad (9)$$

But the magnetic moment of the nucleus is much smaller than that contributed by the electrons, $g_I \sim 1/2000 g_J$. Therefore, to a first approximation, we have

$$g_F \sim g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} \quad (10)$$
Fig. 2. I-J coupling scheme.
Paschen-Back Effect

As was previously mentioned, the above discussion is true only as long as \( \mathbf{I} \) and \( \mathbf{J} \) are tightly coupled. As soon as a perturbing force, such as a magnetic field, is applied, the coupling between \( \mathbf{I} \) and \( \mathbf{J} \) will be weakened. In the limit of a high magnetic field, \( \mathbf{I} \) and \( \mathbf{J} \) completely decouple and precess independently about \( \mathbf{H} \), Fig. 2b. In this region, the Paschen-Back region, the vector \( \mathbf{F} \) loses its meaning, and \( \mathbf{F} \) is no longer a good quantum number. The energy of a level is now completely described by \( m_1 \) and \( m_J \), the projections of \( \mathbf{I} \) and \( \mathbf{J} \) on the magnetic field \( \mathbf{H} \). The energy can then be written as

\[
W = -\mu_0 g_J \mathbf{J} \cdot \mathbf{H} - \mu_0 g_I \mathbf{I} \cdot \mathbf{H} \tag{11}
\]

For intermediate fields, neither of the above representations is correct. The problem cannot be solved explicitly in general. The eigenfunctions, from which the energies are obtained, are constructed from the proper linear combinations of the eigenfunctions of either the low-field or the high-field representations.

Intermediate Fields

The complete Hamiltonian in the high-field representation can now be written as

\[
\mathcal{H} = h \mathbf{I} \cdot \mathbf{J} - \mu_0 g_J \mathbf{J} \cdot \mathbf{H} - \mu_0 g_I \mathbf{I} \cdot \mathbf{H} . \tag{12}
\]

If either \( I \) or \( J \) is equal to one-half, the above expression can be solved exactly for arbitrary \( H \). When \( J = 1/2 \), the solution is the well-known Breit-Rabi formula,

\[
W_{Fm} = -\frac{\Delta W}{2(2I + 1)} - \frac{\mu_I H m}{I} \pm \frac{\Delta W}{2} \sqrt{1 - \frac{4m^2}{(2I + 1)(2I + 2)}} \tag{13}
\]

where we have
\[ \Delta W = \frac{\hbar a}{2} (2I + 1), \text{ and } x = \frac{(g_J - g_I)}{\Delta W} \mu_H, \]

and where \( F \) and \( m \) are the low-field representation quantum numbers. The plus sign is taken for the upper-level \( F = I + \frac{1}{2} \), while the minus sign is for the lower-level \( F = I - \frac{1}{2} \).

If \( I \) and \( J \) are both greater than one-half, the solution is in general most easily soluble by approximate methods. The Hamiltonian operator must also be modified to include any higher-order nuclear multipole moments. In particular, if the nucleus has an electric quadrupole moment, then the Hamiltonian becomes

\[ \mathcal{H} = -\mu_0 (g_J J + g_I \bar{I}) \cdot \bar{H} + \hbar a \mathcal{I} \cdot \mathcal{J} \]

The zero-field splitting between adjacent \( F \) levels no longer satisfies the simple interval rule. In particular, for \( J = \frac{3}{2} \), and \( I = \frac{3}{2} \),

\[ W_3 - W_2 = 3\hbar a + 2\hbar b \]
\[ W_2 - W_1 = 2\hbar a - 2\hbar b \]
\[ W_1 - W_0 = \hbar a - 2\hbar b. \]

The solution for \( J = \frac{3}{2} \) can best be done in the low-field representation in which \( F \) and \( m_F \) are good quantum numbers. In this representation, the matrix elements of \( (\mathcal{I} \cdot \mathcal{J})^n \), \( n = 0, 1, 2, 3, \ldots \), are diagonal, and are given by

\[ \langle F', m_F' | (\mathcal{I} \cdot \mathcal{J})^n | F, m_F \rangle = \left[ \frac{F(F+1) - J(J+1) - I(I+1)}{2} \right]^n \delta_{F,F'} \delta_{m_F,m_F'} . \]

The Hamiltonian can then be written as

\[ \mathcal{H} = \mathcal{H}_0 - \mu_0 (g_J J + g_I \bar{I}) \cdot \bar{H} \]

where \( \mathcal{H}_0 \) is the zero-field energy operator, containing the interaction constants \( a \) and \( b \).
The matrix elements for $\mathbf{J} \cdot \mathbf{H}$ and $\mathbf{I} \cdot \mathbf{H}$ are given by Condon and Shortley, and are of the form:

$$
\langle F m_F \mid \mathbf{J} \cdot \mathbf{H} \mid F m_F \rangle = \frac{F(F+1) + J(J+1) - I(I+1)}{2} H m_F \quad (18)
$$

and

$$
\langle F^* m_F \mid \mathbf{J} \cdot \mathbf{H} \mid F m_F \rangle = \langle F m_F \mid \mathbf{J} \cdot \mathbf{H} \mid F^* m_F \rangle \quad (19)
$$

where

$$
= \pm \left[ \frac{(F+J-I)(F-J+I)(J+1+F)(F+1+F)}{4F^2(2F+1)(2F-1)} \right]^{1/2} M^0 H.
$$

Expressions for $\mathbf{I} \cdot \mathbf{H}$ are obtained from the above by interchanging $I$ and $J$, and $g_I$ and $g_J$. The choice of the sign in Eq. 19 is somewhat arbitrary. Once a choice is made for $\mathbf{J} \cdot \mathbf{H}$, however, the opposite sign must be assigned the expression for $\mathbf{I} \cdot \mathbf{H}$.

For a particular value of $m_F$ (hereafter used interchangeably with $m$), the matrix of the Hamiltonian in the $F, m$ representation looks like:

$$
\begin{array}{cccccc}
F & F-1 & F-2 & F-3 & \ldots & I-J \\
F & H_{F,F} & H_{F,F-1} & 0 & 0 & 0 \\
F-1 & H_{F-1,F} & H_{F-1,F-1} & H_{F-1,F-2} & 0 & 0 \\
F-2 & 0 & H_{F-2,F-1} & H_{F-2,F-2} & H_{F-2,F-3} & 0 & 0 \\
F-3 & 0 & 0 & H_{F-3,F-2} & H_{F-3,F-3} & \ldots & \ldots \\
\ldots & 0 & 0 & 0 & \ldots & \ldots \\
I-J & 0 & 0 & 0 & 0 & \ldots & H_{I-J,I-J} \\
\end{array}
$$

The solution to the problem, then, is to diagonalize the above matrix. The energies $W$ of the various levels are those values of $W$ that make the following determinant zero:
For the particular case of $I = J = 3/2$, there are four $F$ levels, $F = 3, 2, 1, \text{ and } 0$. The complete solution for all of the energy levels then requires the diagonalization

\[
\begin{pmatrix}
H_{F, F-1} & H_{F, F-2} & 0 & 0 \\
H_{F-1, F} & H_{F-1, F-2} & 0 & 0 \\
0 & H_{F-2, F-1} & H_{F-2, F-2} & 0 \\
0 & 0 & H_{F-3, F-2} & H_{F-3, F-3} \\
0 & 0 & 0 & 0
\end{pmatrix} = 0.
\]

The associated energy levels can be solved exactly, in a simple form, only for $m = \pm 3$ and $\pm 2$.

**A General Description of Atomic Beam Flop-In Technique**

The measurements discussed in this paper were made using the conventional flop-in atomic-beam technique originated by Zacharias in his work with potassium. An atomic beam is formed by vaporizing the material of interest in an oven and effusing the atoms thus produced through a small aperture, which is usually in the shape of a narrow slit. If the substance in the vapor state is molecular, then some means must be used to dissociate the molecules. The rate of effusion is limited by the so-called Knudsen condition where the mean-free-path of the atoms in the oven must be greater than the dimensions of the oven aperture. If this condition is violated, the atoms will not effuse out, but will form a jet. This creates a cloud of atoms immediately in front of the oven slits, scattering the beam and effectively increasing the source slit dimensions.

* Beams of molecules can be used, but a discussion of this is outside the scope of this paper.
Atoms effusing out of the oven in a small solid angle pass through another slit system located further down the apparatus. The rest of the atoms are rejected by this collimator. The atoms thus selected are all moving in essentially the same direction. Because the resulting mean free path of the atoms in the beam is very large in comparison to the length of the apparatus, there are no atomic collisions in the beam. Any experiment performed on the beam is done on isolated atoms.

The beam thus formed passes successively through three regions of magnetic field (Fig. 3). These fields are designated "A", "C", and "B", respectively. All three fields are in the same direction, perpendicular to the beam. The C-field is homogeneous, while the A- and B-fields are inhomogeneous. Both inhomogeneities are in the same direction.

If the atoms in the beam have a magnetic moment, they will be deflected by the inhomogeneous fields. From Eq. 7, the average force on an atom is

\[ F_z = \frac{\partial W}{\partial z} = -\frac{\partial W}{\partial H} \frac{\partial H}{\partial z} = \mu_{\text{eff}} \frac{\partial H}{\partial z}, \tag{22} \]

where \( z \) is taken as the direction of inhomogeneity. The amount of deflection will then depend upon the effective magnetic moment, the strength of the inhomogeneity, and the time of transit.

Atoms emitted at a particular angle will be deflected in the A-field in such a manner that they will pass through the collimator slits. In the C-field there is no inhomogeneity so there is no deflection. In the B-field the atoms are further deflected in the same direction and therefore fail to hit the detector. No signal is observed at the detector if the effective magnetic moment is the same in the B- as in the A-field. If, in the region between these two fields, the moment is changed in magnitude or sign, then the trajectory of the beam will also be changed. For the apparatus used by this group, the physical and magnetic geometries of the machine are designed to refocus the beam to impinge upon the detector when the sign of the magnetic moment changes.

Because the deflection of the beam takes place in regions of strong magnetic field, \( \mathbf{I} \) and \( \mathbf{J} \) are to a large extent decoupled. From Eq. 11,
Fig. 3. Atomic-beam trajectories. Solid line indicates trajectory when proper refocusing conditions are satisfied. Positions of oven, collimator, stop wire, and detector are indicated by a, b, c, and d, respectively.
the effective magnetic moment of the atom is essentially \( \mu_0 g J m_J \). The beam will then be refocussed if a transition can be induced, changing \( m_J \) to \(-m_J\).

Transitions can be induced by applying a small rotating component to the C-field. This perturbing magnetic field, \( H' \), is perpendicular to the C-field, and rotates in the same sense as the precessional rotation of the atom. If the two frequencies of rotation are the same, then the atom effectively sees a stationary magnetic field at right angles to the C-field. The atom will then precess about \( H' \), changing the relative orientation of \( \vec{F} \) and \( \vec{H} \). In actuality, an oscillating magnetic field is applied perpendicularly to \( H \). This field can then be resolved into two circularly polarized components. One component rotates in the proper direction to induce a transition, the other component has only a very small effect upon the transition frequency.

The oscillating magnetic field is produced by inserting a small "hairpin" into the C-field and passing an rf current through it. With this type of rf field, and in a low C-field, the only quantum transitions favored are those obeying the selection rules \( \Delta F = 0 \) and \( \pm 1 \), and \( \Delta m_F = \pm 1 \).

Figure 4 is an energy diagram for \( I = J = 3/2 \), in which the energies of the various levels are plotted as a function of magnetic field. An examination of this diagram will show that the only allowed transitions that will be refocused are

\[
F = 3, \ m_F = -1 \leftrightarrow -2 \quad \text{and} \\
F = 2, \ m_F = 0 \leftrightarrow -1.
\]

This is one of the limitations of the flop-in type experiment, only a limited few of the allowable transitions are observable. However, this can also be considered as favorable because of the ease in identifying the observed transition.

For \( J = 3/2 \), and arbitrary \( I \), the observable transitions are

\[
F = I + 3/2, \ m_F = -I + 1/2 \leftrightarrow -I - 1/2 \quad \text{and} \\
F = I + 1/2, \ m_F = -I + 3/2 \leftrightarrow -I + 1/2.
\]
Fig. 4. Energy diagram for $I = J = 3/2$. The above diagram shows the two possible transitions that are refocused in a flop-in type experiment: (a) the upper transition, $F = 3$, $m_F = -1 \leftrightarrow -2$; (b) the lower transition, $F = 2$, $m_F = 0 \leftrightarrow -1$. 
Because the atoms in the beam are inhomogeneous in velocity, they will suffer different deflections depending upon the time they spend in the deflecting fields. Very slow atoms, greatly deflected, may strike the magnet pole faces. Very fast atoms, although not satisfying the refocusing conditions, may still be insufficiently deflected to miss the detector. These atoms contribute to the machine background. This background can be minimized by placing a stop wire in the B-field, so that the shadow of the stop wire falls on the detector. The refocused beam passes around the wire, while that part of the beam coming straight through strikes the wire.

Also contributing to the machine background are Majorana type resonances. As the atoms pass from the A- to the C- and then to the B-field, they pass through regions of changing magnetic field. If these changes are slow enough, no other effect takes place than an adiabatic change in their precessional velocity. If the change is not adiabatic, transitions may be induced, reorienting the atoms in the field. Some of these atoms may then satisfy refocusing conditions and be observed at the detector.

Because of some of the considerations mentioned, the design of an atomic beam machine must be optimized for a particular atomic species. In extreme cases even the temperature-vapor pressure relationship must be considered. The machines used for making the measurements of the gallium isotopes presented in this paper were optimized for atoms in the $^1S_{1/2}$ electronic state with $g_J = 2$. The ground state of gallium has an electronic state of $^2P_{1/2}$ with $g_J = 2/3$. The excited state, $^2P_{3/2}$ with $g_J = 4/3$ is about 826 cm$^{-1}$ above the ground state. At the oven temperature used, 1250°C, about 50% of each state is present in the oven. Refocusing is not optimum for either state, but is best for the upper state. This was later observed when transitions in both states were attempted.
APPARATUS

Flop-In Atomic Beam Machine

The apparatus employed in the work with gallium was the conventional flop-in type atomic-beam machine. Two different machines were used. The first few spin searches were done with machine "A", located in the physics building on the Berkeley campus; the hyperfine structure measurements were done with machine "B", located at the Radiation Laboratory at Berkeley. Machine "A" was built initially for work with the alkali isotopes, and is thoroughly described in the thesis of Robert Sunderland. 10 Machine "B", on which this author has done most of his work, was designed for use with the transuranic elements. The first measurements made with machine "B" were of the spins of some of the neutron-deficient thallium isotopes. 11 This work, and a complete description of the machine and its associated electronic circuitry, is recorded in the thesis of Gilbert Brink. 12 The pictures of the apparatus (Figs. 5 – 11) included in this paper were provided by Mr. Brink.

Novel features of this machine are:

1. A tapered system, where the width of the beam increases with distance from the oven. This gives a factor of 3 greater transmission than in conventional systems.
2. Rotation of the A-magnet pole faces, allowing either flop-in or flop-out types of measurement.
3. Introduction and removal of the oven without disturbing the vacuum of the system.
4. High impedance A- and B-magnets.
5. Electronic power supply for the A- and B-magnets, regulated to 1 part in 30,000.

The following specifications are abstracted from the thesis of Mr. Brink.
Fig. 5. Oven end of tank with glove box.
Fig. 7. Magnet system.
Fig. 10. Counting system.
Fig. 11. Chemistry box and lead shield.
Main Vacuum Tank

The tank is rolled from 1/8-in. stainless steel sheet and welded together. It has an inside diameter of 15-3/4 in. and a length of 59 in. The tank is internally separated into three sections; the oven, buffer, and magnet chambers.

Vacuum System

The three tank sections are pumped upon by individual oil-diffusion pumps; the oven chamber by one Consolidated Vacuum Company MCF 700, the buffer chamber by one MCF 300, and the magnet chamber by two MCF 700 pumps. All pumps are manifolded together and pumped on by another MCF 300, in turn backed by a Welch 1402 B mechanical pump. Vacuum obtained is in the order of 10^{-7} mm of Hg. in the magnet chamber. When the oven is heated, the oven-chamber pressure rises to 10^{-6} mm.

Magnet System

All magnets are water cooled, encased in copper, and separately pumped on to prevent any outgassing into the main vacuum chamber. The A- and B-magnets are equivalent two-wire Rabi systems.

The A-magnet has Permendur pole tips, and is wound with 1080 turns of No. 17 copper wire. The approximate dimensions are 6 in. high by 10 in. wide by 2 in. long. The radii of the pole tips are 0.080 in.

The B-magnet has Armco magnet-iron pole tips and is wound with 3024 turns of No. 17 copper wire. The approximate dimensions are 8-3/4 in. high by 13-3/4 in. wide by 21-1/4 in. long. The radii of the pole tips are 0.375 in.

The C-magnet has Armco magnet-iron pole faces, and is wound with 11,040 turns of No. 26 copper wire. The approximate dimensions are 7 in. high by 12 in. wide by 2 in. long. The magnet gap is 0.375 in.
Magnet Power Supplies

The A- and B-magnet power supply is a transformer-rectifier system electronically regulated to about 1 part in 30,000. It has a maximum current of 5 amp. The C-magnet supply is a 6-v automobile storage battery. The current is varied by a series resistance. With this voltage, the C-field can be varied up to about 120 gauss.

Radiofrequency System

Two oscillators were used for this work. For the low frequency work, we used a General Radio Company 805-C "Standard Signal Generator". For the high frequency searches, we used a General Radio Company 857-A "500-Mc Oscillator". Frequencies were measured with a Hewlett Packard Electronic Counter Model 524b.

Counting System

The counters were the same as used by this group for other radioactive measurements and are described in a paper by Hubbs and Nierenberg. A thallium-activated sodium iodide crystal is used in conjunction with an RCA 6655, or 5819 photomultiplier tube. The output of the multiplier, after being amplified, is fed into a pulse height analyzer. The analyzer is of a standard Radiation Laboratory design.
EXPERIMENTAL PROCEDURE

Isotope Production

The radioactive gallium isotopes discussed in this paper were produced by bombarding copper with energetic alpha particles in the Crocker Laboratory 60-in. cyclotron located on the Berkeley campus. Alpha particle energies up to the maximum of 48 Mev were used, giving the following reactions:

\[
\begin{align*}
\text{Cu}^{63} + \alpha & \rightarrow \text{Ga}^{66} + n & T_{1/2} = 9.4 \text{ hr} \\
\text{Cu}^{63} + \alpha & \rightarrow \text{Ga}^{65} + 2n & T_{1/2} = 15 \text{ min} \\
\text{Cu}^{63} + \alpha & \rightarrow \text{Ga}^{64} + 3n & T_{1/2} = 2.5 \text{ min} \\
\text{Cu}^{65} + \alpha & \rightarrow \text{Ga}^{68} + n & T_{1/2} = 68 \text{ min} \\
\text{Cu}^{65} + \alpha & \rightarrow \text{Ga}^{67} + 2n & T_{1/2} = 78 \text{ hr} \\
\text{Cu}^{65} + \alpha & \rightarrow \text{Ga}^{66} + 3n & T_{1/2} = 9.4 \text{ hr} \\
\text{Cu}^{65} + \alpha & \rightarrow \text{Ga}^{65} + 4n & T_{1/2} = 15 \text{ min}
\end{align*}
\]

Because of the time required in separating the gallium from the copper target, the only activities ever observed were those due to gallium-66, -67, and -68.

The cyclotron targets were cut from commercial sheet copper. These targets were usually mounted on a standard internal probe, although a few runs were made using an external target. The internal probe proved more satisfactory because of the larger beam current that could be used. The internal probe target measured 13-1/4" by 1-1/2" by 0.010". The copper target is water cooled on the back surface, and, as the water is under pressure, a minimum thickness of ten mils is required for mechanical strength.

The cyclotron beam was run at different currents, depending upon the energy of the beam. These currents ranged from 40 to 90 \( \mu \)a. The lower currents were required, when using the full energy of 48 Mev, to prevent puncturing of the copper foil. The higher
currents were permissible when lower beam energies were used.

The time of bombardment ranged from 0.5 to 10 hr, depending upon the isotope of interest. Some work was done with Ga$^{68}$, in which case the target was bombarded for a one-half hour. This kept the activity of Ga$^{66}$ down, and lessened the hazard of handling excessive amounts of activity. The target was removed immediately from the cyclotron after bombardment and transferred into a 2-in. lead-lined box in which the chemistry was performed. These targets measured as much as 1000 r/hr at a foot. When the target was inside of the lead box, the reading at the surface of the box was of the order of 100 mr/hr. In the process of transferring the target and doing the chemistry, the operator accumulated approximately 40 mr of body radiation.

The longer bombardments were used to increase the production of Ga$^{67}$. The targets were then allowed to cool for as much as 24 hr before handling, because most of the activity is due to the positron emission of the short half-life isotopes. No measurable amount of radiation was absorbed by the operator when working on these targets.

For the first few runs, used to determine the spins, the targets were bombarded with the maximum alpha energy to make as much activity as possible. Later, when it was desirable to enhance either the Ga$^{67}$ or Ga$^{68}$ isotopes, the probe was inserted into the cyclotron to radii of 21-3/4" or 19-1/2", decreasing the more energetic reactions.

**Gallium Separation**

A convenient way for separating the radioactive gallium from the copper target is by extraction from a 6 N solution of HCl with diethyl ether. Swift, in 1924, investigated the extraction by diethyl ether of various elements in 6 N HCl solution, and reported the following relative extraction efficiencies:$^{14}$

<table>
<thead>
<tr>
<th>Element</th>
<th>Extraction Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sb (SbCl$_3$)</td>
<td>6%</td>
</tr>
<tr>
<td>Sb (SbCl$_5$)</td>
<td>81%</td>
</tr>
<tr>
<td>As (AsCl$_3$)</td>
<td>68%</td>
</tr>
<tr>
<td>Ga (GaCl$_3$)</td>
<td>97%</td>
</tr>
<tr>
<td>Ge</td>
<td>40 to 60%</td>
</tr>
<tr>
<td>Au</td>
<td>95%</td>
</tr>
<tr>
<td>Element</td>
<td>Percentage</td>
</tr>
<tr>
<td>---------</td>
<td>------------</td>
</tr>
<tr>
<td>Ir</td>
<td>5%</td>
</tr>
<tr>
<td>Fe (FeCl₃)</td>
<td>99%</td>
</tr>
<tr>
<td>Mo (MoO₃)</td>
<td>80 to 90%</td>
</tr>
<tr>
<td>Te</td>
<td>34%</td>
</tr>
<tr>
<td>Tl (TlCl₃)</td>
<td>90 to 95%</td>
</tr>
<tr>
<td>Sn (SnCl₄)</td>
<td>17%</td>
</tr>
<tr>
<td>Sn (SnCl₂)</td>
<td>15 to 30%</td>
</tr>
</tbody>
</table>

The copper target is dissolved in about 15 cc of 10 N HNO₃ in which 20 to 40 mg of gallium carrier have been added. The carrier is added either as the metal or in the form of gallium nitrate. This solution is then boiled to drive off most of the liquid. Sixty cc of 6 N HCl is then added, the HCl having been previously saturated with ether. The solution is placed into an extraction flask with 50 cc of ether, previously saturated with HCl, and stirred vigorously. Because of the ether fumes in the chemistry box, a shaded-pole arcless stirring motor was used to turn a glass stirring rod. The gallium chloride, being more soluble in the ether, is extracted from the acid. The acid with the dissolved copper chloride is separated off. Because some of the copper also goes over into the ether, the ether is washed twice more with 25 cc and then 15 cc of 6 N HCl. This seems to remove essentially all of the copper. The gallium chloride is then extracted from the ether with 15 cc of H₂O. Sodium hydroxide is added to the water until the pH is about 5.5, at which time the gallium precipitates out as Ga(OH)₃. Because some HCl gets into the water, considerable NaOH must be added. Usually the pH of 5.5 is overshot and is adjusted with small amounts of acetic acid and ammonia water. The Ga(OH)₃ is centrifuged and the excess liquid poured off. The precipitate is then dissolved in the smallest possible amount of concentrated NaOH. Usually a few drops of NaOH would dissolve the greater part of the precipitate. Two platinum electrodes are introduced and the gallium electroplated out. For the electrodes used, 4 to 5 v at 0.5 amp seemed sufficient. About 50% of the gallium would plate out in 15 min, and then the plating would become slower. If speed was essential, as for the work with the 68-min activity, we would be satisfied with the 50%. However, for work with Ga⁶⁷ where we needed as much activity as possible, electroplating
was continued for an hr or longer.

The negative electrode is a 0.004-in. platinum wire encased in a Pyrex tube with about 1/8 in. of the wire exposed to the solution. This small electrode was necessary for efficient removal of the gallium. If a thick enough plate of gallium is deposited, it often will coalesce into globules, either at the top or the very bottom of the electrode. The smaller the wire, the more readily the gallium will coalesce. Dipping the wire into warm dilute HCl often helps this action. If the gallium coalesces on the end of the wire, it is removed simply by placing over the carbon oven and tapping gently. Usually the globule would fall directly into the oven. If the gallium would not coalesce, or the globules would not drop off readily, then a pointed glass rod was used to gently scrape off the major portion of the gallium. Any copper not removed from the solution electroplated onto the electrode before the gallium. This greatly hindered the coalescing of the gallium, justifying the extra time required for careful washing during the chemistry.

When the gallium coalesced readily, essentially all of the gallium electroplated was removed. If not, as much as 50% would be lost due to inefficient removal from the electrode. As the copper targets ranged in activity from 5 to 1000 r at a distance of 1 ft, all of the above procedure was carried out behind a 2-in. lead shield, using mechanical manipulators (Fig. 11). If easier handling were possible and speed unessential, the above procedure could yield up to 90% efficiency in extraction. Because of the techniques employed, however, an average extraction yielded about 50% while an exceptionally good one yielded 75%.

Beam Production

Because of the success of some previous work with thallium, we first attempted to distill the gallium directly out of the copper target. Slugs were punched from the center of the target where the beam had produced the greatest discoloration. These slugs were then placed in an oven and the temperature raised until the copper began to effuse. No appreciable beam of gallium was observed. We then tried electroplating the gallium onto thin platinum foils, with the idea of crumpling the whole foil into the oven and distilling the gallium off of the platinum. Again, we had negative results. The only way we could obtain a beam
was by placing the gallium metal into a carbon oven. Ovens made of iron and tantalum failed completely, due to the alloying of the gallium. This effect was also observed by Renzetti in his work with the stable isotopes. 16

To minimize the radiation hazard, we loaded the oven in the chemistry box. The oven was then placed in the oven loader, which allowed the introduction of the oven into the vacuum system through a vacuum lock. As previously mentioned, machine "B" had been used for investigating the heavy elements. This necessitated the complete isolation of any of the alpha activity introduced into the system. The oven loader was therefore enclosed in a glove box (Fig. 5). The inside of the glove box was contaminated with alpha activity which meant that any item put into the glove box could not be removed without decontamination. It was desirable to design a carbon oven that could be easily and economically machined so that it could be discarded after one run. Previously used carbon ovens had been replicas of the standard metal ovens used in atomic beam work. The end product is shown schematically in Fig. 12. All parts are circular, eliminating any milling operations. The cap and slit-snout are press fits and seemed to work very satisfactorily. The most difficult operation was the cutting of the slit at the end of the snout. This was done with a 0.005-in. slitting saw. Because of wobble, the carbon being too soft to hold the blade, the slit usually came out 0.007 in. wide. When made in quantity, these ovens took about 10 min apiece to machine.

The oven was heated by electron bombardment to temperatures ranging from 1200 to 1300°C. It usually was necessary to go to the upper limit, in order to get a large enough beam. The specific activity could have been increased by decreasing the carrier, but this would have made the handling of the metal more difficult. At 1300°C, the vapor pressure is about 0.6 mm of Hg, Fig. 13, and is getting close to the upper limit imposed by the Knudsen condition.

Because of the excessive radiation by the almost "black body" carbon oven, it was found necessary to use a tantalum heat shield. This was made by spot welding 3 mil tantalum foil into a tube which would just slide over the body of the oven. The snout was then inserted into the oven through a previously drilled hole in the heat shield.
Fig. 12. Carbon-oven schematic.
Fig. 13. Vapor-pressure curve for gallium.
Enough of the carbon was shielded to allow temperatures of up to 1300°C to be obtained from 100 w input by electron bombardment. Without the heat shield, over 200 watts were required to obtain the maximum temperature.

Detection of Beam

In work done by others with stable gallium isotopes, the beam was detected with a surface ionization detector. Because the ionization potential of gallium, 5.97 v, is higher than the work function of tungsten, 4.5 v, a continuously oxygenated tungsten filament had to be used, raising the work function to 6 v. For the radioactive isotopes, however, we can make use of this activity for detection purposes. The refocused beam is allowed to deposit upon a suitable collector which is then inserted into an appropriate counting device. The number of disintegrations per unit time is then a measure of the refocused beam. Further, the isotopes can be identified by observing the decay of the activity.

From past experience, sulfur has proved to be a good collector for many of the elements which have low vapor pressures at room temperature. As it also proved to be a good collector of gallium atoms, no further effort was made to find another collector. These collectors were prepared by packing powdered sulfur into recessed brass buttons, Fig. 14, and then heating on a hot plate until the sulfur just melted. Excessive heating darkens the sulfur, but apparently does not affect its collecting properties.

Beam Exposure

For exposure to the beam, the brass buttons had to be introduced into and removed from the apparatus without disturbing the vacuum. This was accomplished, again, by a vacuum lock device, Fig. 6. A typical exposure was 1 min for a direct beam, and 5 min for a refocused beam. Direct beam exposures with the stop-wire out and the magnetic fields on were taken every 15 to 20 min during a run to normalize for any oven-emission fluctuations. As was previously mentioned, the stop-wire is put into the path of the undeflected beam to eliminate any background due to molecules and extremely fast atoms.
Fig. 14. Brass buttons and counter.
This "partial" direct beam is then a measure of the total beam and can be used for normalization. For gallium we found that approximately 50% of the total beam was thrown out by the magnetic field as compared to about 99% for the alkalis.

Immediately after a direct beam exposure was made, the C-field was checked by observing an alkali resonance. Usually the field had not changed by more than a line width. Sometimes there was excessive drift, especially at high values of the magnetic fields. If such a drift was observed, the field was readjusted, and the amount of drift used to adjust the data.

**Spin Search**

For small values of $H$, the approximate expressions for the transition frequencies are given by Eq. 7, and Eq. 10. As previously mentioned, for $J = 3/2$, there are two observable transitions, one each in the upper two $F$-levels. For an atom with a $^2P_{3/2}$ electronic state, $g_J = -4/3$. Using this in the above mentioned equations, we obtain the two transition frequencies as a function of the spin $I$

$$\nu = \frac{4}{3} \frac{3}{2I + 3} \frac{\mu_0 H}{h} \quad F = I + 3/2$$

$$\nu = \frac{4}{3} \frac{2I + 9}{(2I+1)(2I+3)} \frac{\mu_0 H}{h} \quad F = I + 1/2 \quad (23)$$

For each spin $I$ there is, in general, a different transition frequency. Therefore, to determine the spin it is necessary only to take samples of any refocused beam at rf frequencies, given by the above equations for all possible values of $I$. For frequencies corresponding to the spins of any isotopes present in the beam, more atoms will be refocused than for any of the other frequencies.

It is important that the spin search be made at a large enough magnetic field, so that the frequencies given by Eq. 23 differ from each other by at least a line width of the apparatus. Otherwise, the two frequencies overlap and some ambiguity might be introduced. On the other hand, the field must be low enough that the transition is being induced in the Zeeman region so that Eq. 23 applies.
It may be noted that there is a lower limit to a measurable $C$-field. The field is calibrated by using a beam of atoms of known spin and magnetic moment. The uncertainty in measuring the center of the resonance of this calibrating beam is reflected in the uncertainty of the magnetic field.

The alkalis are quite often used for calibration purposes because they can be easily detected with a surface ionization detector and, because $J = 1/2$, their transition frequencies are exactly calculable. For an alkali in the Zeeman region, there is one observable transition given by

$$v = \frac{\mu_0 H}{h} \frac{1}{1 + 1/2}$$

Therefore, for a spin search, the exact value of $H$ need not necessarily be known explicitly. Rather, the search frequencies for the unknown isotope are given simply by the ratio of Eq. 23 to Eq. 24.

For our calibration beam, we used cesium or rubidium in machine "A" and potassium in machine "B". Potassium has a slight disadvantage, perhaps, over cesium and rubidium. Because of its small hyperfine structure (hfs), there is a relatively large quadratic shift from the Zeeman frequency even at low fields. At one Mc, the frequency is already shifted by 7 kc. For larger values of the magnetic field, the relation between field and frequency must be determined by using an exact expression. For convenience, the values of $H$ and $\mu_0 H/h$ were calculated on the University of California Radiation Laboratory's IBM 650 as a function of the potassium transition frequency. This was done for steps of 1 Mc for frequencies ranging from 1 Mc to 426 Mc.

There may be some ambiguity accidentally introduced if the atoms exist in the beam in different electronic states. The reason for this is that transition frequencies for the different $J$'s and $I$'s may be identical or be very close together. In this case, it may be necessary to look for other transitions, or to identify the line by observing the shift from the Zeeman frequency as a function of field. Such an ambiguity was encountered in the gallium measurements. Gallium-67 is made by the Cu($\alpha$, 2n) reaction, and Ga$^{66}$ by Cu($\alpha$, n) and Cu($\alpha$, 3n) reactions. It is therefore impossible to produce one without the other.
assignment presents no problem in this case because odd-mass isotopes must have half-integral spin, while even-mass isotopes have integral spin. Also, as these isotopes have measurable half-lives, decay curves were made of all spin buttons to further verify the proper mass assignment to spin.

**Hyperfine Structure Search**

Once the spin is known, the deviation from the Zeeman frequency is used to determine the hyperfine structure separation. From perturbation theory, * the expression for the energy of a level to second order is

\[ W_{Fm} = W_F + V_{FF}^m + \left( \frac{V_{F,F+1}^m}{W_{F-F+1}^m} \right)^2 + \left( \frac{V_{F,F-1}^m}{W_{F-F-1}^m} \right)^2 \]  

(25)

where the V's are matrix elements of \( \mu_0 H(g_J + g_I) \cdot \vec{H} \) in the F, m representation. \( W_{F} \) is the zero field energy of a particular F level.

For Ga\textsuperscript{67}, the spin proved to be 3/2. The frequencies corresponding to the two observable transitions for the \( ^2P_{3/2} \) electronic state are then

\[ \nu = \frac{-\mu_0 H(g_J+g_I)}{2h} + \left[ \frac{\mu_0 H(g_J-g_I)}{2h} \right]^2 \frac{0.6}{\Delta \nu_{32}} \quad F = 3 \]  

(26)

\[ \nu = \frac{-\mu_0 H(g_J-g_I)}{2h} + \left[ \frac{\mu_0 H(g_J-g_I)}{2h} \right]^2 \left( \frac{0.8}{\Delta \nu_{21}} - \frac{0.2}{\Delta \nu_{32}} \right) \quad F = 2 \]

The procedure for determining the hfs is to increase the field until a shift from the Zeeman frequency is observable. The upper transition depends only upon \( \Delta \nu_{32} \) as far as second order corrections are concerned. The shift in frequency in the upper transition determines an approximate value of \( \Delta \nu_{32} \). This value of \( \Delta \nu_{32} \) can then be used with the lower transition frequency to determine an approximate value for \( \Delta \nu_{21} \). These approximate \( \Delta \nu \) values can then be used to

* See Appendix
predict the transition frequencies at higher fields. A search is made in the vicinity of these predicted values. Resonance curves are then obtained, giving better values for the hyperfine structure intervals.

The uncertainty in the peak frequency of a resonance curve depends upon the width of the curve and its symmetry. For most of our resonances we have arbitrarily assigned an uncertainty equal to one-half of the line-width. This is evidently quite conservative. For some of the narrow symmetrical curves, we would be quite justified in using one-fourth of the line width.

The uncertainty in the center of the resonance curve is reflected in the uncertainty in the hfs intervals. This can be seen by differentiating Eq. 25 and keeping only the dominant term. Then,

\[
\frac{\delta (\Delta v)}{\Delta v} = \frac{\delta (\delta v)}{\delta v}
\]

where \( \delta (\Delta v) \) is the uncertainty in the hfs separation \( \Delta v \), \( \delta v \) is the shift from the Zeeman frequency, and \( \delta (\delta v) \) is the uncertainty in the peak of the resonance. To determine the \( \Delta v \)'s more precisely, we must use a narrower line or measure the resonance at a higher field where the quadratic shift is greater.

The line width is a function of the homogeneity of the C-field, the length of the transition-inducing rf field, and rf power. For gallium, the line width of machine "A" is about 200 kc and is, therefore, well suited for making rough searches. Machine "B" has a line width of about 50 kc and is, therefore, better suited for making precise measurements.

**Counting**

Immediately after exposure, the buttons were counted for 5 min. with a sodium iodide scintillation counter. All buttons were counted cyclically in four counters and their counting rates averaged. For the spin searches, buttons which showed activity were counted in this manner for from several hours to several days. The decay curves obtained were then used to interpret the data. For the hfs search, the buttons were counted later for longer periods of time to obtain better
Typical counting rates were 50 to 100 cpm for a partial direct beam and 0.5 to 5 cpm for a resonance point.

The photoelectric cross section for sodium iodide is shown in Fig. 15. For a 1-mm thick crystal, essentially all of the gallium K x-rays are stopped in the crystal. As the crystal has relatively small volume, it will have a small natural background. When the discriminator window was narrowed, we observed backgrounds as low as 0.5 cpm.

For the initial spin search, the counters were set to detect the K x-rays, because both Ga$^{66}$ and Ga$^{67}$ decay at least partially by electron capture. This was changed later when the hfs of Ga$^{67}$ was being investigated. The activity of Ga$^{66}$ always exceeded that of Ga$^{67}$ immediately after a bombardment and was comparable even after waiting some time. Because a usable beam of Ga$^{67}$ always had an appreciable background of Ga$^{66}$, it was desirable to enhance the fraction of Ga$^{67}$. This was done by waiting longer before running, giving the shorter half-life isotopes a chance to decay away. After about 60 hr, the activity of Ga$^{66}$ is down by a factor of one hundred, while Ga$^{67}$ is down 57% of its initial value. However, the Ga$^{67}$ activity was already so low that a typical 2-hr bombardment gave a running time of a little over an hour. Even at this, the counting rate of the refocused beam was so low that the buttons had to be counted for long periods to obtain convincing statistics. Fortunately, we were helped a little because in the decay of Ga$^{67}$ there occurs a 90- and 92-kev gamma-ray (Fig. 16). The photoelectric cross section is lower for these gamma-rays but is still large enough to give approximately 50% efficiency in detection.

Figures 17 and 18 are typical counter responses, showing the counting rate as a function of the high-voltage setting of the photomultiplier. Fig. 17 shows the response immediately after bombardment, and Fig. 18 is the same sample after a sufficient time has elapsed to allow the Ga$^{67}$ to become the more abundant isotope. The second figure clearly shows the 90- and 92-kev line, and the K x-ray. In Fig. 17 these peaks are hidden under the "hash" of the Ga$^{66}$ and Ga$^{68}$, probably due to the positron emission. Curves were also made at short time intervals to see if there was any essential difference between the counter response for Ga$^{66}$ and Ga$^{68}$. The only difference observed was that there was no K x-ray decay for Ga$^{68}$, as was expected.
Fig. 15. Sodium iodide cross section. Photoelectric cross section for sodium iodide crystal in region of the K edge.
Fig. 16. Decay scheme for gallium-67.
Fig. 17. Typical counter response for gallium-66. "A" is a region of "hash" probably due to the effect of the positrons emitted by Ga\textsuperscript{66} and Ga\textsuperscript{68}. "B" is a region of excessive counter noise.
Fig. 18. Typical counter response for gallium-67. "A" is the 90-and-92-keV gamma rays emitted by Ga$^{67}$, "B" is the K x-ray line, and "C" the region of excessive counter noise.
When working only with Ga\textsuperscript{67}, we set the counters on the peak of the 90- and 92-kev line, and the discriminator window to see only this line. By doing this, the counter would count a maximum of Ga\textsuperscript{67} and a minimum of the other isotopes.

From Figs. 17 and 18, a limitation of the counters is also made evident. As the high voltage is increased to detect K x-rays of lower energy, the background of the counters increases because of photomultiplier dark-current pulses.

**DATA AND RESULTS**

**Spins**

The only exposures that showed counting rates above background were those associated with the spins 0 and 3/2. The isotopes were identified by their half lives. The accepted half life of Ga\textsuperscript{66} is 9.4 hr and of Ga\textsuperscript{67} is 78 hr. The spin-0 button decayed with a pure 9.4-hr half life, as shown in Fig. 19. The spin of Ga\textsuperscript{66} is therefore 0.

The decay of the spin 3/2 button shows a composite decay not too dissimilar from the direct beam (see Fig. 20). Both curves can be resolved into two components with half lives of 9.4 hr and 78 hr. This ambiguity arises because the Zeeman frequencies for spin 0 in the \( ^2P_{1/2} \) state and spin 3/2 in the \( ^2P_{3/2} \) state are identical. As previously mentioned, there are atoms in both states present in the beam, although the machine favors those in the excited state. The spin of Ga\textsuperscript{67} is therefore 3/2. The similarity between the decay of the 3/2 button and the direct beam is entirely accidental.

Figure 21 shows the relative fraction of the direct beam for each of the isotopes as a function of rf frequency. Note that there is only a suggestion of a resonance for the ground state of Ga\textsuperscript{67} at spin 3/2.

Runs were also made to measure the spin of the 68-min isotope, Ga\textsuperscript{68}. No convincing resonance was ever observed, although all of the resonance buttons showed some activity due to Ga\textsuperscript{68}. Figure 21 does not include these exposures. It might be expected that Ga\textsuperscript{68} also has spin 0 because of the results obtained for Ga\textsuperscript{66}. We feel, however, that the only positive statement we can make about Ga\textsuperscript{68} is that its spin...
Fig. 19. Spin-0 decay curve.
Fig. 20. Spin-3/2 and direct-beam decay curves.
Fig. 21. Initial counting rate for all spin buttons.

("A") $^2P_{1/2}$, $I = 3/2$, Ga$^{67}$, $T_{1/2} = 78$ hr.

("B") $^2P_{1/2}$, $I = 0$, Ga$^{66}$, $T_{1/2} = 9.4$ hr.

("C") $^2P_{3/2}$, $I = 3/2$, Ga$^{67}$, $T_{1/2} = 78$ hr.

("D") $^2P_{3/2}$, $I = 0$, Ga$^{66}$, $T_{1/2} = 9.4$ hr.
is not zero. If it were, then the spin-0 exposure should decay like the direct beam less the 78-hr activity. The reason for this is that there would be no way of differentiating between the two species Ga\(^{68}\) and Ga\(^{66}\). No similarity was observed. Further work will be done in the future on this isotope.

The Uncertainty of the Spin-0 Assignment

There is some uncertainty to the assignment of spin 0 to Ga\(^{66}\). For very small hfs, the Paschen-Back transition frequencies for spins other than zero are nearly identical to the transition frequency for spin 0. This can be seen by comparing Fig. 22 with Fig. 23. Fig. 22 shows the energy levels for \(I = 0\), and Fig. 23 for \(I = 1\). The hfs for the spin 1 example is assumed to be so small that the transitions take place in the Paschen-Back region.

If we make the assumption that the spin really is 1, and that there is no quadrupole interaction, then the frequency of the observable transition is given approximately to second order by

\[
\nu = \nu_z + \frac{3}{8} \frac{a^2}{\nu_z}
\]

where \(a\) is the magnetic-moment interaction constant, and \(\nu_z\) is identical to the spin-0 transition frequency. The shift from the spin-0 frequency is then

\[
\delta \nu = \nu - \nu_z = \frac{3}{8} \frac{a^2}{\nu_z}
\]

Two resonance curves were obtained, one at high field, Fig. 24, and one at low field, Fig. 25. Other single exposures were taken at intermediate fields to establish that the transition frequency was linear with the field. Using the uncertainty in the resonance peak due to the line width, we can obtain an upper limit to the interaction constant \(a\). From Fig. 24, we obtain \(a < 7\) Mc, and therefore, \(\mu_I < 0.06\) nm.

The lower resonance (Fig. 25) where the shift would be greater, gives \(a < 400\) kc and \(\mu_I < 0.004\) nm.

These values for the magnetic moment seem highly improbable.
Fig. 22. Energy-level diagram for spin 0. Zeeman transition frequencies for spin zero:

"a" : \( \nu = \frac{4}{3} \frac{\mu_0 H}{h} \)

"b" : \( \nu = \frac{2}{3} \frac{\mu_0 H}{h} \)
Fig. 23. Energy-level diagram for spin 1. Paschen-Bach transition frequencies for spin 1:

("a") \( \nu = \frac{4}{3} \mu_0 H \hbar + a \)

("b") \( \nu = \frac{4}{3} \mu_0 H \hbar \)

("c") \( \nu = \frac{4}{3} \mu_0 H \hbar - a \)
Fig. 24. Spin-0 resonance at 30.054 gauss.
Fig. 25. Spin-0 resonance at 0.709 gauss.
It is therefore reasonable to assume that the spin-0 assignment is correct.

A further and more convincing reason for believing the spin-0 assignment is the relatively large resonances observed. Figure 21 shows that they are greater than 10% as compared to 3% for spin 3/2. Assuming 100% transition probability, 1/4 of the atoms partake in the observed transition, and about 1/2 clear the stop wire. This would overestimate the resonance at 12% of the direct beam. For spin 1, we would expect 1/3 of this and for higher spins still less. The evidence therefore supports the assignment of spin 0.

**Hyperfine Structure of Gallium-67**

The transitions for the two observable lines were followed up to magnetic fields as high as 108 gauss. The resonance curves obtained for each of these searches (Figs. 26 to 35) are summarized in Table I as follows:

<table>
<thead>
<tr>
<th>H (gauss)</th>
<th>(Mc)</th>
<th>(Mc)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.42</td>
<td>616 ± 75</td>
<td>286 ± 16</td>
</tr>
<tr>
<td></td>
<td>532 ± 75</td>
<td>286 ± 16</td>
</tr>
<tr>
<td>25.388</td>
<td>601 ± 27</td>
<td>278.3 ± 4.3</td>
</tr>
<tr>
<td></td>
<td>279.4 ± 4.3</td>
<td></td>
</tr>
<tr>
<td>36.198</td>
<td>607 ± 13</td>
<td>278.6 ± 2.1</td>
</tr>
<tr>
<td>75.432</td>
<td>598.6 ± 3.0</td>
<td></td>
</tr>
<tr>
<td>108.8476</td>
<td>596.9 ± 1.4</td>
<td></td>
</tr>
</tbody>
</table>

The search for the lower transition was not carried above 36 gauss because of the large sixth-order shift. Any higher field measurement would require evaluation of the eighth-order perturbation term. The average values for the hfs separations are
Fig. 26. Spin-3/2 resonances at 13.42 gauss.
Fig. 27. Spins-3/2 and-0 resonances at 13.42 gauss.
Fig. 28. Spin $3/2$, $F = 2$, resonance at 13.42 gauss.
Fig. 29. Spin 3/2, $F = 3$, resonance at 25.388 gauss.
Fig. 30. Spin $3/2$, $F = 2$, resonance at 25.388 gauss.
Fig. 31. Spin 3/2, F = 2, resonance at 25.388 gauss.
Fig. 32. Spin 3/2, F = 3, resonance at 36.198 gauss.
Fig. 33. Spin 3/2, F = 2, resonance at 36.198 gauss.
Fig. 34. Spin 3/2, F = 3, resonance at 75.432 gauss.
Fig. 35. Spin 3/2, F = 3, resonance at 108.848 gauss.
\[ \Delta \nu_{32} = 597.2 \pm 1.3 \text{ Mc} \] and
\[ \Delta \nu_{21} = 278.8 \pm 2.0 \text{ Mc}. \]

**Interaction Constants**

Using Eq. 15 and the above \( \Delta \nu \)'s, we obtain for the interaction constants:

\[ a = 175.2 \pm 0.5 \text{ Mc} \] and
\[ b = 35.8 \pm 1.0 \text{ Mc}. \]

At the time of the writing of this paper, Dr. William Nierenberg completed the program for diagonalizing the Hamiltonian for \( I = J = 3/2 \) on the IBM-650 computer. The values of the interaction constants, \( a \) and \( b \), obtained by this exact diagonalization were consistent with those obtained from sixth-order perturbation theory.

The two observable transition frequencies were then plotted on the automatic plotter associated with the IBM 650. In Fig. 36, these frequencies are plotted as a function of magnetic field. Generalized coordinates are used.

**Dipole and Quadrupole Moments**

The dipole moment and the quadrupole moment can be obtained from measured values of the stable isotopes \( \text{Ga}^{69} \) and \( \text{Ga}^{71} \) by taking the ratios of the \( a \) and \( b \) values. Using the data of Becker and Kusch, we obtain:

\[ \mu_1 = 1.84 \pm .01 \text{ nm} \] and
\[ Q = 0.213 \pm 0.006 \times 10^{-24} \text{ cm}^2. \]
Fig. 36. Transition frequencies for Ga$^{67}$, $I = 3/2$, $2P_{3/2}$.

The points of these curves were calculated and plotted in terms of generalized coordinates on the IBM-650 computer.
Sign of the Dipole and Quadrupole Moments

All of the above calculations and results are based on a positive sign for the magnetic dipole moment, this being consistent with the positive moments of the stable isotopes. If it is assumed that the sign is negative, then the values for the hfs separations are

\[ \Delta v_{32} = 602.0 \text{ Mc} \] 
\[ \Delta v_{21} = 280.5 \text{ Mc}. \]

Both sets of data imply that the sign of the quadrupole and the dipole moment are the same, either both negative or both positive. The experimental evidence favors a positive sign. Figs. 37 and 38 show the difference between the theoretical and the experimental frequencies as a function of the magnetic field H. In Fig. 37, where the sign of \( g_1 \) is assumed to be positive, the difference in all cases is less than the stated uncertainty. If in reality the sign was negative, we would expect these points to lie along the dashed line. In Fig. 38, the sign is assumed negative, and the dashed line is where the experimental points should fall if this assumption were incorrect. The data clearly favors a positive sign.

Discussion of Results

The assignment of spin 3/2 to Ga\(^{67}\) is to be expected from other considerations. In the simple shell-model theory, the 31st odd proton is assumed to be in a \( ^2P_{3/2} \) level. This is supported by the fact that the stable isotopes Ga\(^{69}\) and Ga\(^{71}\) have measured spins of 3/2. This would then suggest that Ga\(^{67}\) also has a spin of 3/2. The beta-ray and gamma-ray decay scheme for Ga\(^{67}\) has been shown to be consistent with the assignment of 3/2.\(^{19}\)

The shell model, however, predicts a spin greater than zero for Ga\(^{66}\). Both the odd proton and the odd neutron are expected to be in a \( ^2P_{3/2} \) level. Nordheim's weak rule states that the spins of the two nucleons should add in such a manner that the net spin is greater than their difference, or

\[ I > |j_n - j_p| = 0. \]
Fig. 37. Self-consistency of Ga$^{67}$ resonances assuming $g_1$ is positive. Difference of theoretical transition frequency, assuming a positive $g_1$, and experimentally observed transition frequency plotted against magnetic field. The dotted line represents the position of these differences if the sign of $g_1$ is actually negative.
Fig. 38. Self-consistency of Ga$^{67}$ resonances assuming $g_I$ is negative. Difference of theoretical transition frequency, assuming a negative $g_I$, and experimentally observed transition frequency plotted against magnetic field. The dotted line represents the position of these differences if the sign of $g_I$ is actually positive.
Mann, Meyerhof, and West have investigated the gamma beta decay scheme of Ga$^{66}$\textsuperscript{20}. They have found that it is not inconsistent with a spin assignment of either 0 or 1. Reasonable spin and parity assignments could be made for all except the three lowest levels of the excited state of Zn$^{66}$ using either spin 0 or 1. If we assume the spin is 1, the three lowest levels gave poor agreement to Nordheim's classification of the log ft values. The assignment of spin 0 is more favorable in this case, but less so with respect to beta decay to the ground state.

Recently, Fraunfelder, Hanson, Levine, Rossi, and DePasquali have measured the longitudinal polarization of the positron emitted from Ga$^{66}$ to the ground state of Zn$^{66}$\textsuperscript{21}. The beta decay, $0^+ \to 0^+$ obeys Fermi selection rules. No polarization was observed. This fact supports the suggestion of Alder, Stech, and Winther\textsuperscript{22} that parity is conserved in a pure Fermi transition.

The magnetic-dipole and electric-quadrupole moments and their signs are consistent with the values previously measured for the stable isotopes Ga$^{69}$ and Ga$^{71}$. 
CONCLUSIONS

The spins of two gallium isotopes have been measured. They are

\[ I = \frac{3}{2} \text{ for Ga}^{67}, \]

and, with some qualification,

\[ I = 0 \text{ for Ga}^{66}. \]

The hyperfine structure constants for Ga\(^{67}\) have been measured by making use of sixth-order perturbation theory, and verified by exact diagonalization on the IBM 650 computer. These values for the hfs give for the magnetic dipole and electric octupole moments the values

\[ \mu_I = 1.84 \pm 0.01 \text{ nm and} \]

\[ Q = 0.213 \pm 0.006 \times 10^{-24} \text{ cm}^2. \]
ACKNOWLEDGMENTS

At this time I would like to express my thanks to those with whom I have worked: the cyclotron crew for the many roentgens of radiation to which they surely must have been exposed, the transportation crew for their prompt delivery of the targets, and the health chemistry monitors for their assistance and efforts to make my work less hazardous.

I would also like to thank those other graduate students who have helped me from time to time. In particular, I would like to thank Howard Shugart for his example, in my opinion, of what a graduate student should be.

To Dr. John Hubbs, I am indebted for the experimental skills that he patiently tried to teach me.

But most of all, I would like to thank my research director, Dr. William Nierenberg. For his enthusiasm and abundant energy when I faltered, for his confidence and encouragement when I became discouraged, and just for his friendship, I am greatly indebted. It has been an extremely pleasant experience working with him, one which I hope some day can be continued.

And lastly, I must thank my wife, Kay, for the many hours spent alone in the evenings watching a defective television set and my neglected baby daughter, Laurie, who only recently realized she had a daddy.

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APPENDIX

Approximate Solution for $I = J = 3/2$ by Perturbation Methods

The exact solution for the energy levels of an atom with $J = 3/2$ in a magnetic field are the roots of equations up to the fourth order. The experimentally observed quantity is, however, the transition frequency, or essentially, the difference in energy of two of these levels. In the expressions for the energies appear the interaction constants $a$ and $b$, which are the final quest of the investigation. To solve for these constants by direct diagonalization would be somewhat tedious, not to mention the time involved. At this time a general procedure for solving the problem for $J = 3/2$ and arbitrary $I$ is being set up by Dr. William Nierenberg on the Radiation Laboratory IBM 650 machine.

The problem can be solved approximately by perturbation methods, at least to low order of perturbation correction. Condon and Shortley give the general solution to the problem, of which the evaluation of the higher-order corrections almost convinced the author that the more direct approach of exact diagonalization might have been more economical.

For the peculiar case $I = J$, the evaluation of the correction is greatly simplified because all diagonal terms of the perturbation are equal, i.e., the Zeeman frequencies for all transitions $m = \pm 1$ are equal. Because of this, the third- and fifth-order corrections were determined to be zero, suggesting that all odd orders above one are also zero.

Using the expansions given by Condon and Shortley, we obtain the expressions for the energy levels to the sixth order, for $F = 3$, and $F = 2$, and arbitrary $m$ which are as follows:

(a) $F = 3$.

\[
W^m_3 = W_3(0) - \mu_0(g_J + g_I) Hm + \frac{\langle 32 \rangle^2}{(32)^2} + \left\{ \frac{\langle 32 \rangle^2}{(32)^2} \left[ \frac{\langle 21 \rangle^2}{(31)^2} - \frac{\langle 32 \rangle^2}{(32)^2} \right] \right\} \\
+ \left\{ 2 \frac{\langle 32 \rangle^6}{(32)^5} - \frac{\langle 32 \rangle^4}{(32)^3} \frac{\langle 21 \rangle^2}{(31)^2} + \frac{1}{(31)^2} \right\} + \frac{\langle 32 \rangle^2}{(31)^2} \frac{\langle 21 \rangle^4}{(32)^3} + \frac{\langle 32 \rangle^2}{(32)^2} \frac{\langle 21 \rangle^2}{(31)^2} \frac{\langle 10 \rangle^2}{(30)}
\]
(b) $F = 2$.

$$W_m^2 = W_2(0) - \mu_0 (g_J + g_I) Hm + \left[ \frac{\langle 21 \rangle^2}{(21)} - \frac{\langle 32 \rangle^2}{(32)} \right]$$

$$+ \left\{ \frac{\langle 32 \rangle^4}{(32)^3} - \frac{\langle 21 \rangle^4}{(21)^3} + \frac{\langle 32 \rangle^2 \langle 21 \rangle^2}{(21)^2 (32)} \left[ \frac{1}{(21)} - \frac{1}{(32)} \right] - \frac{\langle 21 \rangle^2 \langle 10 \rangle^2}{(21)^2 (20)} \right\}$$

$$+ 2 \left\{ \frac{\langle 21 \rangle^6}{(21)^5} - \frac{\langle 32 \rangle^6}{(32)^5} - 3 \frac{\langle 21 \rangle^4 \langle 10 \rangle^2}{(21)^4 (20)} + 3 \frac{\langle 32 \rangle^4 \langle 21 \rangle^2}{(32)^4 (21)} \right\}$$

$$- 3 \frac{\langle 32 \rangle^2 \langle 21 \rangle^4}{(32) (21)^2} + \frac{\langle 21 \rangle^2 \langle 10 \rangle^4}{(21)^3 (20)^2} - \frac{\langle 21 \rangle^4 \langle 10 \rangle^2}{(21)^3 (20)^2} + 2 \frac{\langle 32 \rangle^2 \langle 21 \rangle^4 + \langle 32 \rangle^4 \langle 21 \rangle^2}{(32)^2 (21)^3}$$

$$- 2 \frac{\langle 32 \rangle^4 \langle 21 \rangle^2 + \langle 32 \rangle^2 \langle 21 \rangle^4}{(32)^3 (21)^2} + \frac{\langle 32 \rangle^2 \langle 21 \rangle^2 \langle 10 \rangle^2}{(32) (21)^2 (10)} \left[ \frac{1}{(20)} - \frac{1}{(32)} + \frac{2}{(21)} \right] \right\}.$$ 

In the above expressions, $W_{F(0)}$ is the absolute energy of the $F = 3, 2, 1, \text{and } 0 \text{ level in zero magnetic field.}$ The numbers in the denominators are convenient abbreviations for the hyperfine separation between $F$ levels, i.e.,

$$(32) = W_3(0) - W_2(0).$$

The expressions in the numerators are abbreviations for the matrix elements of the perturbation in the $F, m$ representation. We have left off the $m$ notation on all of these for convenience. Thus, we have

$$\langle 32 \rangle = \langle 3m \left| \mu_0 (g_J \vec{J} + g_I \vec{I}) \cdot \vec{H} \right| 2m \rangle.$$ 

The matrix elements are already quoted (Eq. 19). For $I = J = 3/2$, the off-diagonal matrix elements are given in Table II.
Table II

Off-diagonal matrix elements for $I = J = 3/2$

<table>
<thead>
<tr>
<th>matrix element</th>
<th>$m = 0$</th>
<th>$m = -1$</th>
<th>$m = -2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle 32 \rangle^2$</td>
<td>$9/5 x^2$</td>
<td>$8/5 x^2$</td>
<td>$x^2$</td>
</tr>
<tr>
<td>$\langle 21 \rangle^2$</td>
<td>$16/5 x^2$</td>
<td>$12/5 x^2$</td>
<td>$0$</td>
</tr>
<tr>
<td>$\langle 10 \rangle^2$</td>
<td>$5 x^2$</td>
<td>$0$</td>
<td>$0$</td>
</tr>
</tbody>
</table>

where

$$x^2 = (g_J - g_1)^2 (\mu_0 H)^2.$$
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