Nuclear Spin and Moments of 14-hr Ga^{72}†

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The hyperfine structure of the 14-hr isotope Ga^{72} has been investigated by use of atomic-beam magnetic-resonance techniques. The nuclear spin is found to be 3, the magnetic dipole moment $\mu_I=-0.132196 \pm 0.000013$ nm (subject to correction for a possible hyperfine anomaly), and the electric quadrupole moment $Q=+0.718\pm0.007$ barn.

INTRODUCTION

HISTORICALLY, the first elements to be investigated by atomic-beam magnetic-resonance techniques were those belonging to group I. In many respects, group-III atoms were also found to be well suited to this method of study. At the start of the present research, many of the stable and several of radioactive group-III isotopes had been investigated, and it was felt that the Argonne atomic-beam machine was well suited to the study of the 14-hr group-III isotope Ga^{72}.

The odd-odd nuclide Ga^{72}, with $Z=31$ and $N=41$, lies within the region in which the shell-model encounters ambiguity in the assignment of ground-state spin values. The difficulty is caused by the closely competing $g_{9/2}$, $p_{1/2}$, $f_{5/2}$, and $p_{3/2}$ levels for both, protons and neutrons. The value of new information on isotopes in this region is clear.

The most interesting aspect of the present experiment, however, was not anticipated. The isotope Ga^{72} has a large quadrupole moment and a small dipole moment, so that the hyperfine structure is very unusual and presents interesting variations from normal atomic beam studies.

GENERAL PRINCIPLES

The method used is the well known scheme of Zacharias[1] as modified for work with radioactive species. A collimated, ribbon-shaped beam of neutral atoms is allowed to pass through a series of three magnetic fields. The first and third magnetic fields (conventionally called the A and B fields, respectively) are strong and inhomogeneous. In these strong fields, m_J (the projection of the electronic angular momentum J on the magnetic field axis) is a good quantum number. In the two inhomogeneous fields, atoms with the same m_J are deflected away from the detector in the same direction.

The A and B fields and their gradients are such that if, in the region between these two deflecting fields, an atom suffers a change of state such that $m_J(A) = -m_J(B)$ [where $m_J(A)$ and $m_J(B)$ are the values of m_J in the A and B fields, respectively], then the atom will undergo a deflection in the B magnet opposite to the deflection in the A magnet, and will have a net deflection of zero at the position of the detector slit.

Such changes of state can be induced by an oscillating magnetic field of the appropriate resonance frequency ν in the uniform field of the second magnet (conventionally called the C magnet). For resonance to occur, $h\nu$ should be equal to the energy difference of the two states of the atom in the magnetic field and the oscillating field should be perpendicular to the direction of the C field for $\Delta m_F=\pm1$ and parallel to it for $\Delta m_F=0$. The quantum numbers F, m_F are often used to describe the state of an atom in a finite magnetic field, although it should be understood that this is the zero-field state from which the state in question is developed by the application of the magnetic field.

Several such allowed, observable transitions are indicated on the hyperfine structure diagrams (Figs. 1 and 2) of the two atomic states of Ga^{72} which are present

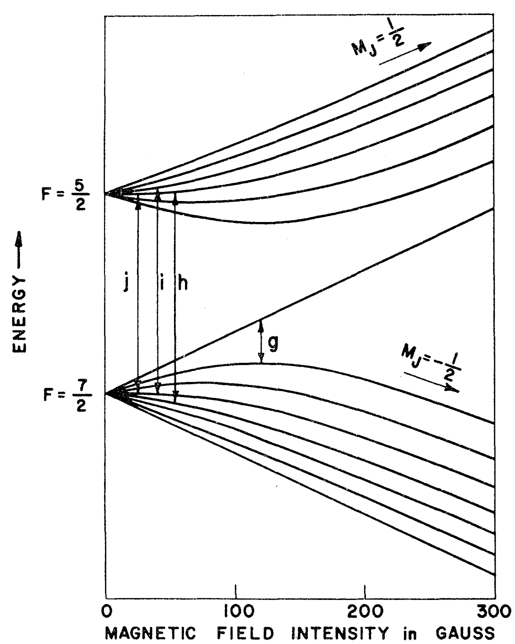

FIG. 1. Hyperfine structure diagram of the atomic $P_{1/2}$ ground state of Ga^{72}. Four of the observable transitions are indicated.

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

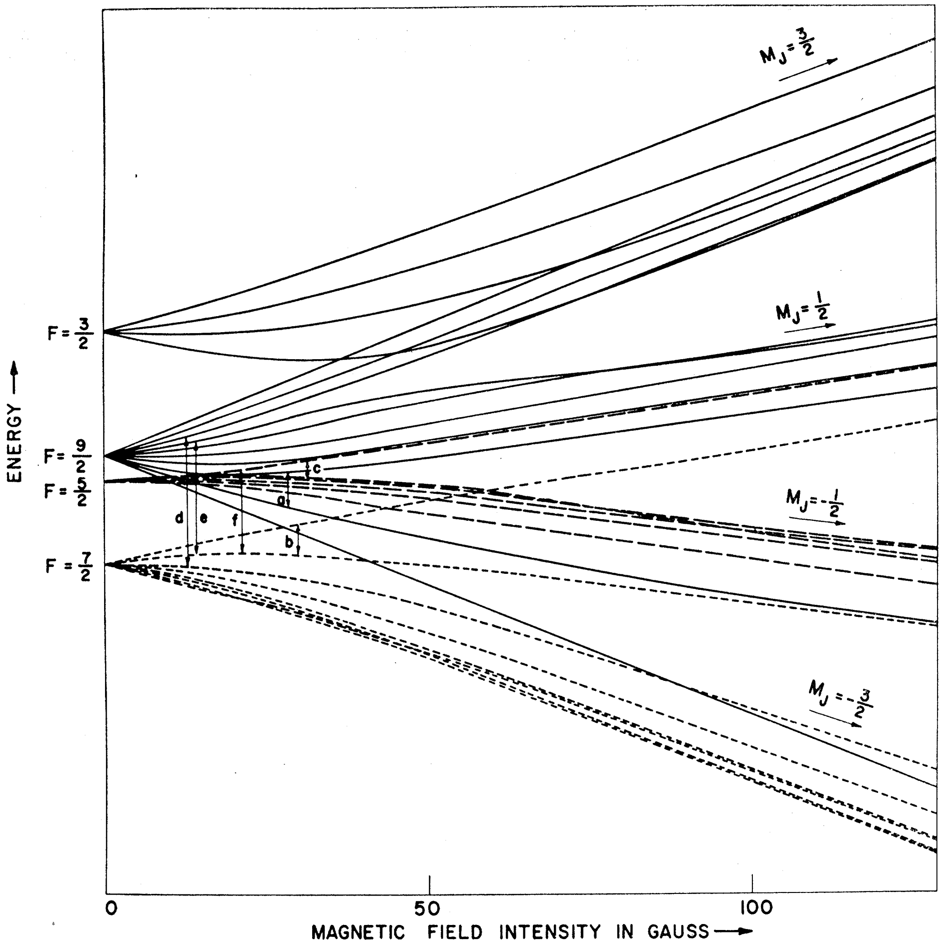

FIG. 2. Hyperfine structure diagram of the atomic $P_{3/2}$ metastable state of Ga^{72}. Six observable transitions are indicated.

in the atomic beam. The justification of the ordering of the hyerpfine levels will be discussed elsewhere in the text.

PREPARATION OF SOURCE

Preparation of the source was straightforward. About 400 mg of gallium metal was placed in the Argonne CP-5 Research Reactor overnight at a flux of 2×10^{13} neutrons cm^{-2} sec^{-1}. Since the thermal-neutron capture cross section in the 40% abundant Ga^{71} is 4.6 barns, sources of adequate strength could be obtained easily. The 21-min isotope Ga^{70} was relatively depleted by the time collections were under way and, in any case, the identity of atoms collected could be easily determined from half-life measurements. Such measurements on atoms collected at resonance showed only the 14-hr decay of Ga^{72}.

A typical sample, prepared in the way described, could be used for two days before it became too weak. A large number of such samples were required for collection of the data presented.

EXPERIMENTAL DETAILS

The atomic beam machine used for the present experiment has been described previously.[2] A number of the components of the instrument have, however, been extensively changed, as described briefly below.

The oven system was reduced in size and provided with improved heat shielding to facilitate attainment of the higher temperature required to produce a beam of gallium. For most of the data taking, a triple-walled Ta shield surrounded a Ta block in which the oven was placed. The block was heated by electron bombardment from W filaments which were also within the shield. The oven itself was a graphite cylinder 1 in. long$\times\frac{1}{2}$-in. o.d., sealed with a tapered graphite plug on one end. The slit was 0.003 in. wide and was parallel to the axis of the cylinder.

With the present arrangement of the three magnetic fields, a large part of the field (about 30 gauss) in the homogeneous magnet is due to stray flux from the A and B magnets. Under these circumstances, it soon became clear that good regulation of the A and B

[2] L. S. Goodman and S. Wexler, Phys. Rev. **99**, 192 (1955).

TABLE I. Observations on the $\Delta F=0$ ("g") transition in the atomic $P_{1/2}$ state of Ga^{72}. For calibration of the magnetic field, the $(2,-1\leftrightarrow 2,-2)$ transition in the $P_{1/2}$ state of Ga^{69} and Ga^{71} was used except at 100 gauss, where improved resolution made it possible to use the Ga^{69} transition alone. The observed frequency at 40.874 gauss is the combined result of two independent measurements. All frequencies are in Mc/sec except that the differences between theoretical and experimental frequencies in lines 3 and 4 are in kc/sec.

H(gauss)	8.175	20.437	40.874	100.000
Calibration frequency	1.901	4.767	9.581	23.842
Observed Ga^{72} frequency	1.130±0.013	3.045±0.013	6.831±0.007	24.172±0.013
$\nu_{\mathrm{th}}-\nu_{\mathrm{exp}}(\mu_I>0)$	6	−8	0	−2
$\nu_{\mathrm{th}}-\nu_{\mathrm{exp}}(\mu_I<0)$	7	−7	2	2
$\lvert\Delta\nu\rvert;\mu_I>0$	175±40	150±5	153.6±0.6	153.63±0.13
$\lvert\Delta\nu\rvert;\mu_I<0$	175±40	150±5	153.8±0.6	153.67±0.13

magnets was a prerequisite for the necessary stability of the C field. This problem was met by providing the flux for each of the inhomogeneous magnets from Alnico V slugs which could be rotated in such a way as to vary the division of flux between the gap and a magnetic shunt. The resulting A and B fields are continuously variable and reversible, even though maintained solely by permanent magnet sources. The details have been published elsewhere.[3]

For stable isotopes of materials such as Ga or K, an oxidized hot tungsten wire can be used as a surface-ionization detector to indicate the intensity of the beam which passes through the detector slit.

The Ga^{72}, produced by neutron capture in stable 40% abundant Ga^{71}, could be distinguished from the much more abundant Ga^{71} only by making use of its radioactivity. Detection was accomplished by allowing the beam to condense on a suitable collector. The disintegrations of the atoms on such collectors were then counted in windowless gas-flow Geiger counters external to the main apparatus. Thin sulfur-coated steel collectors were used for most of the work.

The problem of rapid insertion and removal of the

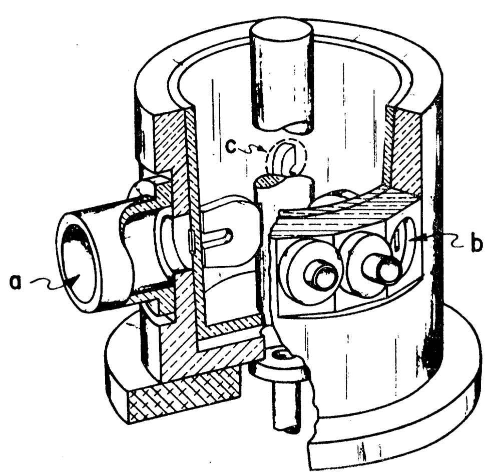

FIG. 3. Rotatable brass "stopcock" used for introducing collectors into the vacuum system rapidly.

[3] J. A. Dalman and L. S. Goodman, Rev. Sci. Instr. **28**, 961 (1957).

collectors from the vacuum system was met by constructing a tapered differentially-pumped brass "stopcock." The unit is shown in Fig. 3. Three small equally-spaced holes (120° apart) pierce the stationary female part. One hole (a), at the left of the figure, is directly on the beam line so that the beam may pass through and strike a collector attached to the rotatable male part. The two holes (b and c) in the female part are for loading and unloading samples. Several additional holes provide for the differential pumping. The hollow male part has three depressions, spaced 120° apart, large enough to receive individual collectors which are held in place by small magnets. With this arrangement it has been found possible to change collectors, at a vacuum of 3×10^{-7} mm of Hg, in about 10 sec.

The radiofrequency equipment used is standard, mostly from commercial sources. The Berkeley Model 5570 frequency counter, with the Model 5575 vhf converter, was used for the frequency standard. For signal generation, a General Radio 805-C and Hewlett-Packard 606A and 608C were used together with homemade amplifiers. A Collins 51J-4 receiver and Hewlett-Packard 540A transfer oscillator were also of great value. Modification of the rf loops required for the "flop-out on flop-in" part of the experiment is discussed below.

The magnetic field was measured by observing resonances in one or more of the isotopes Ga^{69}, Ga^{71}, K^{39}, and Cs^{133}, for which the field dependence of all transition frequencies is known. Atoms of the first two isotopes were present in the beam at all times, and the K or Cs beam was produced from an auxiliary oven which could be moved into position whenever desired for a field measurement.

MEASUREMENTS

$P_{\frac{1}{2}}$ Atomic State

Observed resonance frequencies of Ga^{72} in the $P_{1/2}$ atomic state for four values of the magnetic field are listed in Table I. These data are only consistent with a nuclear spin $I=3$ and a hyperfine separation $\Delta\nu=153.63\pm0.13$ Mc/sec or 153.67 ± 0.13 Mc/sec for the respective assumptions of a positive or a negative nuclear dipole moment.

TABLE II. Observations of $\Delta F=1$ transitions in the atomic $P_{1/2}$ state of Ga^{72}. The transitions used for field calibration in Ga^{69} and Ga^{71}, and in Cs^{133}, were the $(2, -1 \leftrightarrow 2, -2)$ and the $(4, -3 \leftrightarrow 4, -4)$ transitions, respectively. The key to identification of the Ga^{72} transitions is given in Table III. All frequencies are in Mc/sec.

H (gauss)	5	5
Calibration frequency	1.162	1.752
Calibration isotope	$Ga^{69,71}$	Cs^{133}
Observed Ga^{72} frequency	153.052±0.010	153.722±0.001
Identification of Ga^{72} resonance	j	h,i
$\|\Delta\nu\|$, corrected to zero field	153.648±0.010	153.653±0.001

Table II lists the observations of the $\Delta F=1$ transitions in which (with correction back to zero field) the hyperfine splitting is measured directly. The transitions h and i, which are identified in Table III, are identical in frequency except for a small difference, $2g_I\mu_0H$. For the magnetic moment inferred from the small hfs observed, this amounts to only 0.3 kc/sec at 5 gauss. To first order, when the nuclear contribution is neglected, the transitions h and i are independent of field since the energies of the upper and lower states for each transition have the same field dependence. This accounts for the relatively small field correction compared to that for transition j.

From the observed hfs, $\Delta\nu=153.653\pm0.001$ Mc/sec, the magnetic moment of the nucleus can be calculated with the Fermi-Segrè relation[4] and the published data[5] on Ga^{69} and Ga^{71}. Ignoring hyperfine anomalies, one calculates $|\mu|=0.132196\pm0.000013$ nm. One cannot determine the sign of μ from our data on the $P_{1/2}$ atomic ground state.

TABLE III. Convention for identification of transitions in Ga^{72}. All of the listed transitions except e have been observed, although h and i were unresolved. In addition to those listed, 14 others in the $P_{1/2}$ state should be observable with the present arrangement. As mentioned in the text, the classification of definite levels as "observable" implies a choice of b/a. The experimentally observed value was used in preparing this table.

Transition	Label	ΔF
a. Metastable $P_{3/2}$ state		
$(\frac{9}{2}, -\frac{5}{2} \leftrightarrow \frac{9}{2}, -\frac{7}{2})$	a	0
$(\frac{7}{2}, \frac{7}{2} \leftrightarrow \frac{7}{2}, \frac{5}{2})$	b	0
$(\frac{5}{2}, \frac{5}{2} \leftrightarrow \frac{5}{2}, \frac{3}{2})$	c	0
$(\frac{9}{2}, \frac{5}{2} \leftrightarrow \frac{7}{2}, \frac{3}{2})$	d	1
$(\frac{9}{2}, \frac{3}{2} \leftrightarrow \frac{7}{2}, \frac{5}{2})$	e	1
$(\frac{5}{2}, \frac{5}{2} \leftrightarrow \frac{7}{2}, \frac{5}{2})$	f	1
b. Atomic $P_{1/2}$ (ground) state		
$(\frac{7}{2}, \frac{7}{2} \leftrightarrow \frac{7}{2}, \frac{5}{2})$	g	0
$(\frac{5}{2}, \frac{1}{2} \leftrightarrow \frac{7}{2}, -\frac{1}{2})$	h	1
$(\frac{5}{2}, -\frac{1}{2} \leftrightarrow \frac{7}{2}, \frac{1}{2})$	i	1
$(\frac{5}{2}, \frac{1}{2} \leftrightarrow \frac{7}{2}, \frac{1}{2})$	j	1

[4] E. Fermi and E. Segrè, Z. Physik **82**, 729 (1933).
[5] N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956).

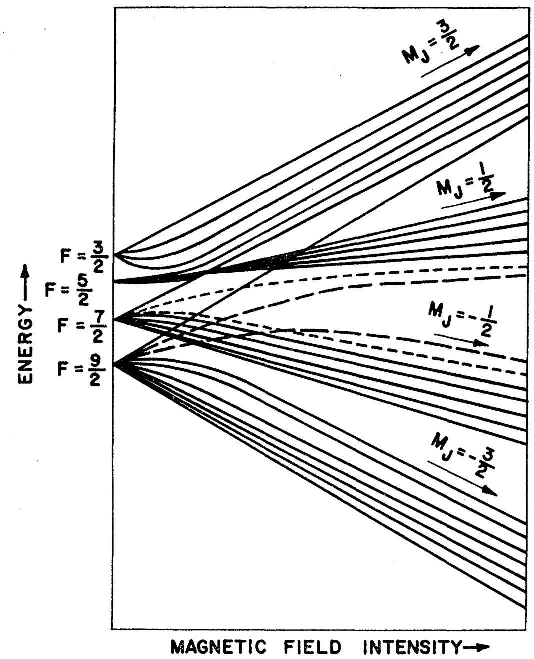

FIG. 4. Schematic hyperfine structure diagram of the metastable atomic $P_{3/2}$ state of Ga^{72} on the assumption that $|b|\ll|a|$. This shows the so-called normal ordering of the hyperfine levels.

$P_{\frac{3}{2}}$ Atomic State

Although the atomic ground state of Ga is $P_{1/2}$, the relatively high temperature (about 1100°C) in the source of the atomic beam excites about 50% of the atoms in the beam to the metastable $P_{3/2}$ atomic state at 826 cm^{-1}. For atoms in this atomic state, both nuclear magnetic dipole and electric quadrupole contributions to the hyperfine interaction must be considered. The Hamiltonian is written

$$\mathcal{H}=ha\mathbf{I}\cdot\mathbf{J}+hbQ_{op}+g_J\mu_0\mathbf{J}\cdot\mathbf{H}+g_I\mu_0\mathbf{I}\cdot\mathbf{H},$$

where Q_{op} is the nuclear electric quadrupole operator, h is Planck's constant, μ_0 is the Bohr magneton, and H is the external field. The symbols g_J and g_I represent the electronic and nuclear g factors, respectively, and are defined by the relations $g_J=-\mu_J/J$ and $g_I=-\mu_I/I$, where both μ_J and μ_I are expressed in Bohr magnetons. The quantities a and b are the nuclear magnetic dipole and electric quadrupole interaction constants, respectively, and are to be determined experimentally. The definitions of b and Q_{op} are those of Ramsey.[5]

For the usual case encountered in hyperfine interaction studies, $|b|\ll|a|$ and the ordering of the hyperfine levels at zero field is determined by the magnetic dipole interaction and is given by $\langle F|\mathbf{I}\cdot\mathbf{J}|F\rangle$.

A schematic hyperfine interaction diagram for $b=0$ and $I=3$ is shown in Fig. 4. There are two allowed observable transitions for $\Delta F=0$, $\Delta m_F=1$ (where the quantum numbers are those for the zero-field condition from which the states in question are developed). The

TABLE IV. Observations on the $\Delta F=0$ transitions in the metastable $P_{3/2}$ state of Ga^{72}. The transitions a, b, and c are identified in Table III. The transitions α, β, and γ are the $(3,-1\leftrightarrow 3,-2)$, $(2,0\leftrightarrow 2,-1)$, and the $(2,-1\leftrightarrow 2,-2)$ transitions, respectively. All frequencies are in Mc/sec and frequency differences in kc/sec. In a few cases, the observed frequencies listed for Ga^{72} represent the weighted averages of several measurements.

	Calibration data				Ga^{72} data		
H (gauss)	Isotope	Atomic state	Transition	Observed frequency	Transition	Observed frequency	$\nu_{obs}-\nu_{calc}$
4.087	$Ga^{69,71}$	$P_{3/2}$	α and β	3.835	a	2.627 ± 0.020	-2
4.087	$Ga^{69,71}$	$P_{3/2}$	α and β	3.835	b	2.220 ± 0.020	11
4.827	$Ga^{69,71}$	$P_{3/2}$	α and β	4.532	b	2.723 ± 0.020	29
8.175	$Ga^{69,71}$	$P_{1/2}$	γ	1.901	a	5.436 ± 0.030	-32
8.175	$Ga^{69,71}$	$P_{1/2}$	γ	1.901	b	5.255 ± 0.020	27
12.262	$Ga^{69,71}$	$P_{1/2}$	γ	2.854	a	8.752 ± 0.030	4
12.262	$Ga^{69,71}$	$P_{1/2}$	γ	2.854	b	9.090 ± 0.025	34
16.350	K^{39}	$P_{1/2}$	γ	12.349	a	13.411 ± 0.030	-91
16.350	K^{39}	$P_{1/2}$	γ	12.349	b	13.587 ± 0.030	-53
16.350	$Ga^{69,71}$	$P_{1/2}$	γ	3.810	c	2.941 ± 0.010	10
18.393	$Ga^{69,71}$	$P_{1/2}$	γ	4.289	a	16.990 ± 0.030	25
18.393	$Ga^{69,71}$	$P_{1/2}$	γ	4.289	b	16.239 ± 0.030	52
18.393	$Ga^{69,71}$	$P_{1/2}$	γ	4.289	c	3.914 ± 0.020	27
30.656	Ga^{71}	$P_{3/2}$	β	29.911	a	44.010 ± 0.050	-98
30.656	Ga^{71}	$P_{3/2}$	β	29.911	b	34.280 ± 0.050	56
30.656	Ga^{71}	$P_{3/2}$	β	29.911	c	$13.830{+0.030 \atop -0.040}$	-12

first connects the substates indicated by the dotted lines in the hyperfine level $F=9/2$, and the second connects the dotted $F=\frac{7}{2}$ substates. These are the only low-frequency allowed transitions between two states which have equal and opposite values of m_J at high field. In general, for the $P_{3/2}$ atomic state with normal ordering (i.e., with $|b|\ll|a|$) and $I\neq0$, there are two allowed observable transitions.

In the more general case in which the ratio b/a may take on any value, the zero-field energies of the hyperfine levels will have a variety of possible orders. Figure 5 shows how the ordering of the zero-field eigenvalues of the $P_{3/2}$ atomic state of Ga^{72} depends on b/a. The algebraic sign of a has been assumed to be negative. The assumption of a positive sign would cause each energy level to be reflected through the zero-energy axis.

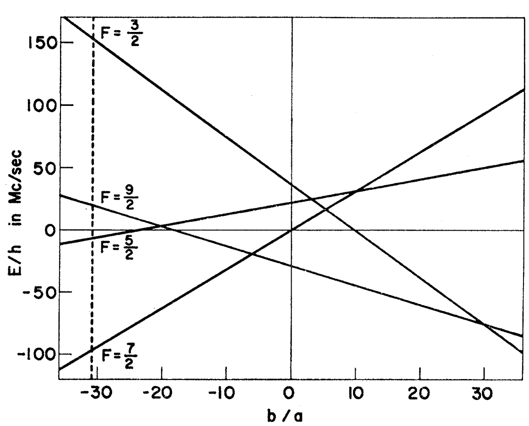

FIG. 5. Energy ordering of the zero-field hyperfine levels in the $P_{3/2}$ state of Ga^{72} as a function of b/a. The quadrupole moment is assumed to be positive.

The zero-field hyperfine levels may occur in seven different orders. If the hyperfine structure diagram is drawn for each condition, it will be seen not only that the observable transitions can be between different magnetic substates of F than for the usual case, but also that the number of observable transitions may vary. Indeed, for three out of the seven possible orderings, a transition is also observable in the $F=\frac{5}{2}$ hyperfine state.

Referring again to Fig. 5, one observes at the far left the ordering given by a pure nuclear electric-quadrupole interaction of positive sign; at the far right, the same with negative sign; and in the center, pure nuclear magnetic-dipole coupling with negative sign. The dashed line is at the value of b/a determined for Ga^{72} in this experiment.

Table IV lists the observations on the $\Delta F=0$ transition in the $P_{3/2}$ atomic state of Ga^{72} along with the data taken to set the homogeneous magnetic field. The observed transition c establishes that the order is indeed abnormal. This transition was not observable at low magnetic fields so that the interpretation of the early data was somewhat difficult. The reason for this phenomenon is of interest in itself and will be discussed under transition probabilities.

The data in Table IV are consistent with the nuclear spin and magnetic dipole moment determined in the studies of the $P_{1/2}$ atomic state, and with a ratio $b/a=-30.95\pm0.02$.

This was close enough to allow a search for the observable $\Delta F=1$ transitions from which one can calculate a and b more accurately. Table V lists these observed frequencies from which the values of $|a|$, $|b|$, and b/a can be deduced.

Transition d is unique in that the high-field states that have equal and opposite effective magnetic

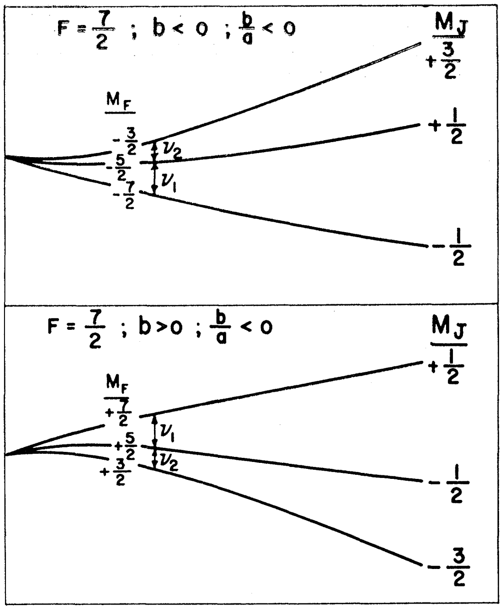

FIG. 6. Magnetic substates of the $F=\frac{7}{2}$, $J=\frac{3}{2}$ hyperfine level of Ga^{72} used in determining the algebraic signs of the moments.

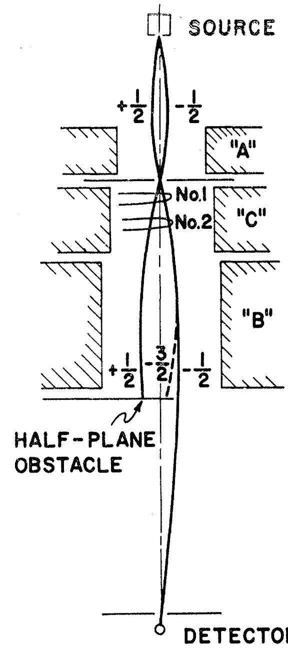

FIG. 7. Schematic diagram of the atomic beam setup used in determining the algebraic signs of the moments.

moments have high-field quantum numbers $m_J=\pm\frac{3}{2}$ instead of the usual $m_J=\pm\frac{1}{2}$. At high fields this transition is strongly forbidden ($\Delta m_J=3$); but at low fields, transitions between the states from which these $m_J=\pm\frac{3}{2}$ states are developed are allowed.

Algebraic Sign of the Moments

The algebraic sign of the moments was determined with a slight modification of the "flop-out on flop-in" method of King and Jaccarino.[6] The method allows one to determine the zero-field ordering of the hyperfine levels.

For Ga^{72} the contribution to the hyperfine structure from the nuclear magnetic dipole moment is small compared to the effect of the nuclear electric quadrupole moment. The determination of the zero-field ordering of the hyperfine levels therefore allows one to infer the sign of the nuclear electric quadrupole moment. The sign of the nuclear magnetic dipole moment then follows from the previously determined negative sign of b/a.

Figure 6 shows pertinent details of the hyperfine structure diagrams for the $F=\frac{7}{2}$, $J=\frac{3}{2}$ state of Ga^{72}, drawn with the two assumptions of positive and negative nuclear electric quadrupole moments. One sees that the observable $F=\frac{7}{2}$ transition should take place between $m_F=\frac{7}{2}$ and $\frac{5}{2}$ for the positive sign of b, and $m_F=-\frac{7}{2}$ and $-\frac{5}{2}$ for the negative.

Figure 7 is a schematic drawing of the trajectories of atoms involved in the experiment. In the particular situation shown, atoms which have $m_J=\frac{1}{2}$ in the A magnet and $m_J=-\frac{1}{2}$ in the B magnet will reach the detector. The atoms which have $m_J(A)=-\frac{1}{2}$ and $m_J(B)=\frac{1}{2}$ and which would ordinarily reach the detector slit are stopped by a half-plane obstacle. The beam passes in sequence through rf loops No. 1 and No. 2.

Suppose that $b<0$ so the situation at the top of Fig. 6 applies. Note that ν_1 is the transition frequency for $F=\frac{7}{2}$, $m_F=\pm\frac{7}{2}\leftrightarrow m_F=\pm\frac{5}{2}$ for either sign of b, and ν_2 is the transition frequency for $F=\frac{7}{2}$, $m_F=\frac{5}{2}\leftrightarrow m_F=\pm\frac{3}{2}$. Both frequencies are calculated for the correct negative value of b/a, but are essentially independent of the algebraic sign of b or a. If rf power is applied in loop No. 1 at frequency ν_1, some atoms will change state.

TABLE V. Observations of the $\Delta F=1$ transitions in the metastable $P_{3/2}$ state of Ga^{72}. All frequencies are in Mc/sec. Identification of the transitions "d" and "f" may be made by referring to Table III. The $(4, -3\leftrightarrow 4, -4)$ transition in Cs^{133} was used to calibrate the field.

H(gauss)	Calibration frequency	Ga^{72} transition	Observed Ga^{72} frequency	$\lvert\Delta\nu\rvert$ corrected to zero field
1	0.350	d	118.048±0.003	117.103±0.003
1	0.350	f	88.770±0.015	89.700±0.015

[6] J. G. King and V. Jaccarino, Phys. Rev. 94, 1610 (1954).

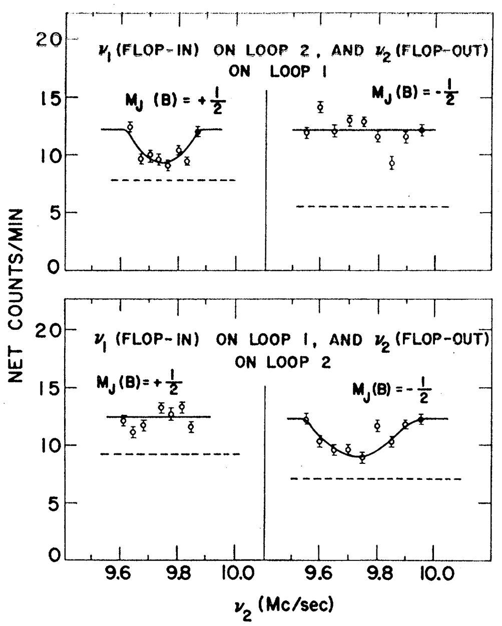

FIG. 8. Results of the "flop-out on flop-in" experiment to determine the algebraic signs of the moments.

Of the atoms undergoing this transition, only those changing from $m_J(A)=+\frac{1}{2}$ to $m_J(B)=-\frac{1}{2}$ will pass through the detector slit. If rf power at the frequency ν_2 is now applied in loop No. 2, it can only affect those atoms which would have $m_J(B)=+\frac{1}{2}$ or $+\frac{3}{2}$, and would therefore not affect those going through the slit.

Suppose now that the frequencies fed to the two loops are interchanged. The application of frequency ν_1 in loop 2 will cause some atoms to change state from $m_J(A)=+\frac{1}{2}$ to $m_J(B)=-\frac{1}{2}$ so they will pass through the detector slit, just as when this frequency was applied to loop 1.

However, the application of ν_2 in loop 1 will now cause some of the atoms with $m_J(A)=+\frac{1}{2}$ to change to $m_J(B)=+\frac{3}{2}$ so ν_1 in loop 2 can no longer focus them on the detector slit. Thus, if a transition takes place in an rf field at frequency ν_1 in loop No. 1, applying power at ν_2 in loop No. 2 will have no effect. If, on the other hand, the same transition takes place with ν_1 in loop No. 2, then applying ν_2 in loop No. 1 will diminish the detected beam ("flop-out on flop-in").

If on the other hand $b>0$ and ν_1 is applied to loop No. 1, applying ν_2 in loop No. 2 will spoil the refocusing of some of the atoms because the change from $m_J=+\frac{1}{2}$ to $-\frac{1}{2}$ in loop No. 1 may be followed by a further change to $-\frac{3}{2}$ in loop No. 2. A determination of the order of application of the two frequencies ν_1 and ν_2 for which "flop-out on flop-in" occurs thus determines the sign of b.

If the half-plane is on the other side of the center line of the machine, and shields the detector from those atoms with $m_J(B)<0$, it can be seen by like reasoning that for the same sign of b the opposite sequence of ν_1 and ν_2 would be required to produce the same effect.

The experiment was first run for the stable isotope Ga^{69} which has a positive nuclear magnetic dipole moment,[5] the hyperfine interaction of which governs the ordering of the hyperfine levels. It was found necessary to isolate the two loops from each other with a double-walled copper shield which had slits to allow the transmission of the beam. Without the isolator, the effect was confused and dependent on power because of the presence of fields of more than one frequency at the position of either loop.

With the method confirmed for Ga^{69}, radioactive Ga^{72} was investigated and the data of Fig. 8 were obtained. At a field of 15 gauss, a frequency $\nu_1=12.050$ Mc/sec was fed into the indicated loop and the refocused fraction of the radioactive beam was observed. Frequency ν_2 was then fed into the other loop and swept in steps as shown.

The data show some scatter, which is apparently caused by collection efficiency and contamination, but on top of this is a definite observation of the "flop-out on flop-in" from which one infers that $b>0$. Since b/a was known to be negative from a fit of the hyperfine interaction constants, it can also be inferred that $a<0$.

COMPUTATIONS

Calculation of Transition Frequencies

A program has been written for the Argonne digital computer GEORGE to calculate the eigenvalues of the Hamiltonian of the hyperfine interaction. Input

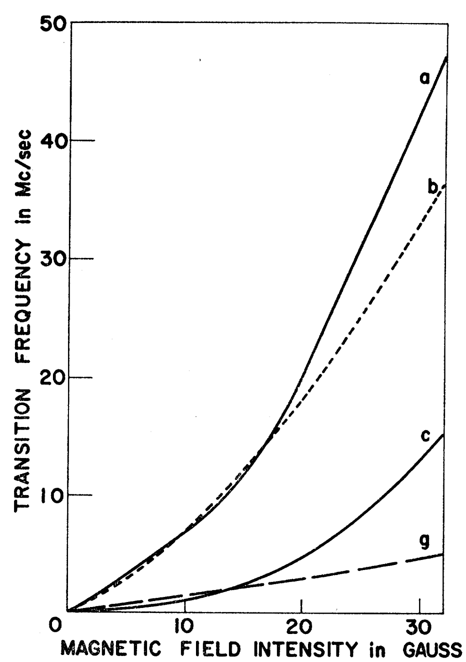

FIG. 9. Magnetic field dependence of the $\Delta F=0$ transitions in Ga^{72}. Identification of the transitions is given in Table III.

data consist of I, J, a, b, g_J, g_I, the states between which transitions occur, and a sequence of magnetic field values for which the calculation is to be made. The machine forms all of the necessary matrices in the (F,m_F) representation, calculates the necessary eigenvalues and eigenvalue differences for the pairs of states chosen, and the (F,m_F) eigenvector for each state. Such calculations are of inestimable value in atomic beam research with radioactive isotopes since the search for a resonance is time-consuming, and the source is decaying. Usually the $\Delta F=0$ transitions have to be measured with rather good precision at relatively high fields. The hyperfine interaction constants can then be calculated with sufficiently small uncertainties so that a search for $\Delta F=1$ transitions becomes practical.

In order to predict even the $\Delta F=0$ transitions accurately, good calculations are needed. Figure 9, which shows the $\Delta F=0$ transition frequencies as a function of magnetic field strength, illustrates the need for precise computations for identification of the observed transitions.

Relative Transition Probabilities

If the transition probability P at resonance is sufficiently small, it is proportional to

$$M^2=|\langle\psi_f|\mathbf{H}_{\mathrm{rf}}\cdot\boldsymbol{\mu}|\psi_i\rangle|^2.$$

Although in atomic beam experiments with radioactive isotopes one seldom finds it practical to work at the limit of small P, the quantity M^2 is still a useful guide in the investigation of accidentally forbidden transitions. The phrase "accidentally forbidden" is used to describe the situation in which no selection rules are violated but M^2 nevertheless turns out to be very small. Within the above framework M_a^2/M_b^2 will be called the transition probability of case a relative to case b.

In the perturbation term $\mathbf{H}_{\mathrm{rf}}\cdot\boldsymbol{\mu}$, the major contribution to the magnetic moment is the $\mu_0 g_J J$ of the electronic system. With this approximation $\mathbf{H}_{\mathrm{rf}}\cdot\boldsymbol{\mu}=-g_J\mu_0\mathbf{H}_{\mathrm{rf}}\cdot\mathbf{J}$.

For $\Delta m_F=\pm1$, only an oscillating rf magnetic field perpendicular to H_0 (the homogeneous field) can supply the necessary angular momentum and

$$\mathbf{H}_{\mathrm{rf}}\cdot\mathbf{J}=H_{\mathrm{rf}}\tfrac{1}{2}\sqrt{2}(J_x\pm iJ_y).$$

For $\Delta m_F=0$, only that component of $\mathbf{H}_{\mathrm{rf}}$ parallel to $\mathbf{H}_0$ is effective and $\mathbf{H}_{\mathrm{rf}}\cdot\mathbf{J}=H_{\mathrm{rf}}J_z$. For a given rf magnetic field intensity in the appropriate direction, the relative transition probability is thus reduced to the calculation of $|M/H_{\mathrm{rf}}|^2=\frac{1}{2}|\langle\psi_f|J_x\pm iJ_y|\psi_i\rangle|^2$ for $\Delta m_F=\pm1$ and $|\langle\psi_f|J_z|\psi_i\rangle|^2$ for $\Delta m_F=0$.

Figure 10 shows the relative transition probabilities for the three observable $\Delta F=0$ transitions of the $P_{3/2}$ atomic state of Ga^{72}. The point of interest is the small ratio of the transition probability of transition c to transition a (about 0.001) at zero field. When this number had increased to 0.02 at a field of 16 gauss, the transition was observable.

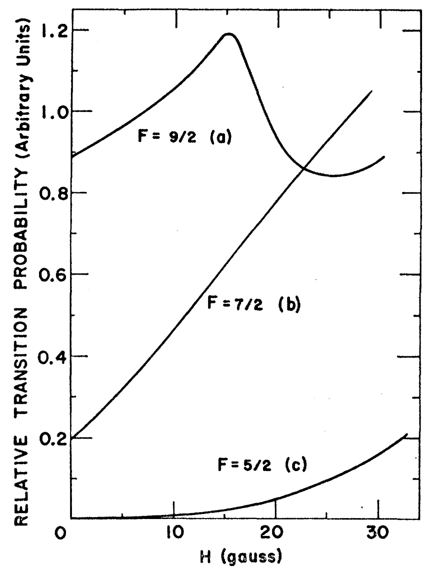

FIG. 10. Relative transition probabilities of the three observable $\Delta F=0$ transitions in the $P_{3/2}$ state of Ga^{72} as a function of magnetic field strength.

RESULTS

The quantities measured directly in this experiment are the nuclear spin I and the zero-field hyperfine intervals $\Delta\nu$. Thus we have

$$I=3,$$

and

$$\Delta\nu(F=\tfrac{7}{2}\leftrightarrow F=\tfrac{5}{2})=\tfrac{7}{2}|a(P_{1/2})|=153.653\pm0.001\text{ Mc/sec},$$

in the $P_{1/2}$ atomic ground state. In the metastable $P_{3/2}$ state we find the same nuclear spin and

$$\Delta\nu(F=\tfrac{5}{2}\leftrightarrow F=\tfrac{7}{2})=-\tfrac{7}{2}a+(7/20)b=89.700\pm0.015\text{ Mc/sec},$$

and

$$\Delta\nu(F=(9/2)\leftrightarrow F=\tfrac{7}{2})=+(9/2)a+\tfrac{3}{4}b=117.103\pm0.003\text{ Mc/sec}.$$

From these hyperfine intervals, the hyperfine interaction constant in the $P_{1/2}$ state is calculated to be

$$a(P_{1/2})=-43.9009\pm0.0003\text{ Mc/sec},$$

and in the $P_{3/2}$ state we find

$$a(P_{3/2})=-6.2593\pm0.0027\text{ Mc/sec},$$

$$b(P_{3/2})=+193.693\pm0.016\text{ Mc/sec},$$

and consequently

$$(b/a)(P_{3/2})=-30.945\pm0.013.$$

By use of the hyperfine interaction constants and the nuclear spin, approximate values for the nuclear moments may be deduced from the Fermi-Segrè relation,[4] since the necessary measurements have been made on Ga^{69} and Ga^{71}.[5] From the hyperfine interval in the $P_{1/2}$ state the value deduced for the nuclear magnetic moment is

$$\mu_I(P_{1/2}) = -0.132196 \pm 0.000013 \text{ nm},$$

subject to any possible hyperfine anomaly. In the same way, a value for the nuclear dipole moment can be extracted from the $P_{3/2}$ data. One thus finds, again subject to a hyperfine anomaly,

$$\mu_I(P_{3/2}) = -0.13227 \pm 0.00006 \text{ nm},$$

which is consistent with the result for the $P_{1/2}$ state.

Similarly, if one assumes that the ratio of the quadrupole moments of two isotopes (of the same Z) is proportional to the ratio of their quadrupole interaction constants b, one can deduce a value for Q. Comparing Ga^{72} with Ga^{69} or Ga^{71} in this way, one finds

$$Q(\text{Ga}^{72}, P_{3/2}) = +0.718 \pm 0.007 \text{ barn},$$

where the uncertainty quoted results from those given for the quadrupole moments of Ga^{69} and Ga^{71}.[7,8] A direct method for evaluating Q is also available. From reference 7,

$$ha = -\mu_0^2 g_I \mathfrak{F} \frac{2L(L+1)}{J(J+1)} \langle r^{-3} \rangle_{\text{av}},$$

$$hb = +e^2 Q \mathfrak{R} \frac{2L}{2L+3} \langle r^{-3} \rangle_{\text{av}},$$

where L and J are the quantum numbers describing the electronic state, and $\mathfrak{F}$ and $\mathfrak{R}$ are known relativistic correction factors close to unity. On combining these two expressions with the definition for g_I, an explicit expression independent of $\langle r^{-3} \rangle_{\text{av}}$ is obtained for Q, namely,

$$Q = +\frac{8}{3}\frac{b}{a}\left(\frac{\mu_0}{e}\right)^2 \frac{m}{M}\frac{\mu_I}{I}\frac{\mathfrak{F}}{\mathfrak{R}},$$

where m/M is the ratio of the mass of the electron to that of the proton, and $\mathfrak{F}/\mathfrak{R}$ has the value[7] 0.97960 for gallium. For Ga^{72}, Q is thus found to have the value

$$Q(\text{Ga}^{72}, P_{3/2}) = +0.724 \text{ b},$$

with a relative uncertainty equal to that of μ_I and of the same origin (a possible hyperfine anomaly). It is seen that this result is in good agreement with that found above.

It should be noted that the procedures outlined above serve to determine magnitudes only. The algebraic signs of the derived quantities were measured independently as discussed above.

DISCUSSION

It is of interest to reconcile the measured nuclear spin and moments of Ga^{72} with the conventional nuclear models. It is immediately clear that it is difficult to understand a quadrupole moment as large as that measured for Ga^{72} without introducing some collective effects.

One successful approach was recently suggested[9] to the authors. Let the protons in ${}_{31}\text{Ga}_{41}{}^{72}$ be arranged just as in ${}_{31}\text{Ga}_{40}{}^{71}$ with $I=\frac{3}{2}$, and let the neutrons be arranged like those in ${}_{34}\text{Se}_{41}{}^{75}$ with $I=\frac{5}{2}$. The quadrupole moments for both of these isotopes are known. If one then folds the neutron and proton configurations together to form ${}_{31}\text{Ga}_{41}{}^{72}$, requiring only that $I(\text{Ga}^{72})=3$ as observed, the value $+0.58$ b is predicted for the quadrupole moment of Ga^{72}. This is in fair agreement with the measured value of $+0.72$ b.[10]

Unfortunately, the magnetic dipole moment for Se^{75} has not been measured so that one cannot predict a dipole moment for Ga^{72}. One can, however, work backwards, combining the known dipole moment of Ga^{71} and Ga^{72} to predict a dipole moment of -1.91 nm for Se^{75}. The validity of the interpretation is thus subject to test by measurements on Se^{75}.

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[7] L. Davis, B. T. Feld, C. W. Zabel, and J. R. Zacharias, Phys. Rev. **76**, 1076 (1949).

[8] G. E. Becker and P. Kusch, Phys. Rev. **73**, 584 (1948).

[9] B. J. Raz (private communication, 1960).

[10] V. J. Ehlers, in a private communication, pointed out to the authors that G. F. Koster [Phys. Rev. **86**, 148 (1952)] has re-evaluated the quadrupole moments of Ga^{69} and Ga^{71} after examination of the effects of configuration interaction. His results, when combined with the present value of b, lead to the result $Q(\text{Ga}^{72}) = (+0.59 \pm 0.03)b$, in close agreement with the prediction of Raz.