

## The Hyperfine Structure of the Stable Isotopes of Indium\*

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The atomic beams magnetic resonance method has been applied to the study of the h.f.s. of the metastable  ${}^2P_{3/2}$  state of both  $\text{In}^{113}$  and  $\text{In}^{115}$ . It has been found that the separations of the energy levels at zero external magnetic field are completely described by the equation  $W = haC/2 + hbC(C+1)$ , where  $C = F(F+1) - I(I+1) - J(J+1)$ , and  $a$  and  $b$  are the magnetic dipole and electric quadrupole interaction constants, respectively. No evidence has been found to indicate the existence of a nuclear moment of higher order than the quadrupole moment. These are the heaviest atoms in which a critical search for such moments has been made.

The numerical values of the constants for  $\text{In}^{115}$  are:

$$a^{115} = 242.165 \pm 0.002 \text{ mc/sec.}$$

$$b^{115} = 1.56098 \pm 0.00006 \text{ mc/sec.}$$

The ratio  $a^{115}/a^{113}$  has been determined by Hardy and Millman

to be  $1.00224 \pm 0.00010$ . From this value, and the results of our measurements, the numerical values of the constants for  $\text{In}^{113}$  are:

$$a^{113} = 241.624 \pm 0.024 \text{ mc/sec.}$$

$$b^{113} = 1.53855 \pm 0.00015 \text{ mc/sec.}$$

where the uncertainties in  $a^{113}$  and  $b^{113}$  follow from the uncertainty in the ratio of the  $a$ 's. The numerical values of the electric quadrupole moments are:

$$Q^{115} = 1.161 \times 10^{-24} \text{ cm}^2$$

$$Q^{113} = 1.144 \times 10^{-24} \text{ cm}^2.$$

It is seen from the ratios  $I^{115}/I^{113} = 1$ ,  $a^{115}/a^{113} = 1.00224$ ,  $b^{115}/b^{113} = 1.0146$ , that in these measurable nuclear properties  $\text{In}^{113}$  and  $\text{In}^{115}$  exhibit the greatest degree of similarity observed for any pair of isotopes of odd mass differing in mass by two units.

### INTRODUCTION

THE methods of atomic beams have been applied to the study of the h.f.s. of indium in several previous experiments. The first of these experiments was that of Millman, Rabi, and Zacharias,<sup>1</sup> in which the atomic beam zero moment method was applied to the measurement of the h.f.s. separation of the  ${}^2P_{3/2}$  state, the nuclear spin, and the magnetic moment of  $\text{In}^{115}$ . This was followed by the work of Hamilton,<sup>2</sup> who used the zero moment method to study the h.f.s. of the  ${}^2P_{3/2}$  state of  $\text{In}^{115}$ . Hamilton verified the previously determined value of the nuclear spin, and measured the constants,  $ha^{115}$ , the energy of interaction of the magnetic dipole moment of the nucleus with the magnetic field of the orbital electrons, and,  $hb^{115}$ , the energy of interaction of the nuclear electric quadrupole moment with the gradient of the electric field at the nucleus. The uncertainty in these measurements was about 1 percent. Hamilton further set an upper limit on the value of  $c^{115}$ , the octupole moment interaction constant. It is of interest to note that in neither of these experiments was any structure found which could be attributed to the rarer of the two stable indium isotopes,  $\text{In}^{113}$ , even though the abundance of the isotope is sufficiently great so that structure arising from it could have been observed. Somewhat later, the atomic beam magnetic resonance method was utilized by Hardy and Millman<sup>3</sup> to measure the nuclear spin and h.f.s. separation ( $\Delta\nu$ ) of the  ${}^2P_{3/2}$  state of  $\text{In}^{113}$ . Hardy and Millman also remeasured the same quantities for the  ${}^2P_{3/2}$  state of  $\text{In}^{115}$  with greater precision than that of the previous experiment.<sup>1</sup> Finally, Kusch and Foley<sup>4</sup>

and Taub and Kusch,<sup>5</sup> in their work on the magnetic moments of the electron and proton, respectively, have measured the ratio of the  $g_J$  of indium in the  ${}^2P_{3/2}$  state to that of sodium in the  ${}^2S_{1/2}$  state, the h.f.s. separation of the  ${}^2P_{3/2}$  state of  $\text{In}^{115}$ , and the ratio  $g_I(\text{In}^{115})/g_J({}^2P_{3/2})$  with high precision.

The results of these experiments that are of primary interest here are as follows: (1) The nuclear spin of  $\text{In}^{115}$  and that of  $\text{In}^{113}$  are both equal to  $9/2$ , and (2) the h.f.s. separation of the ground state of  $\text{In}^{115}$  is equal to that of  $\text{In}^{113}$  within about 1 part in 500 ( $\Delta\nu_{115}/\Delta\nu_{113} = 1.00224 \pm 0.00010$ ). The very small difference between these  $\Delta\nu$ 's requires for its observation the high resolution characteristic of the magnetic resonance method. The ratio  $\Delta\nu_{115}/\Delta\nu_{113}$  is equal to the ratio  $a^{115}/a^{113}$ , except for possible perturbations, e.g., that which might arise from the existence of matrix elements between the two states  $J=1/2$  and  $J=3/2$ . This perturbation, if it exists, will affect the levels of the two isotopes in exactly the same way, except for the difference in their  $\Delta\nu$ 's, and, consequently, since the  $\Delta\nu$ 's are so nearly the same, it may be neglected. With these facts available, it is of interest to know the ratio of the electric quadrupole interaction constants of the two indium isotopes with at least as great a precision as that with which the ratio of the magnetic dipole interaction constants is known. The remainder of this paper will describe experiments, utilizing atomic beams magnetic resonance methods, on the h.f.s. of the  ${}^2P_{3/2}$  state of both  $\text{In}^{115}$  and  $\text{In}^{113}$ , from which this information is obtained.

### DESCRIPTION OF THE ENERGY LEVELS

The Hamiltonian expression for the interactions of an atom with nuclear spin  $\mathbf{I}$ , total electronic angular momentum  $\mathbf{J}$ , with a magnetic field may be written

\* H. Taub and P. Kusch, Phys. Rev. **75**, 1481 (1949).

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<sup>1</sup> Millman, Rabi, and Zacharias, Phys. Rev. **53**, 384 (1938).

<sup>2</sup> D. R. Hamilton, Phys. Rev. **56**, 30 (1939).

<sup>3</sup> T. C. Hardy and S. Millman, Phys. Rev. **61**, 459 (1942).

<sup>4</sup> P. Kusch and H. M. Foley, Phys. Rev. **74**, 250 (1948).

as:<sup>6</sup>

$$\mathcal{H} = a\mathbf{I} \cdot \mathbf{J} + b2\mathbf{I} \cdot \mathbf{J}(2\mathbf{I} \cdot \mathbf{J} + 1) + \mu_0 H(g_J J_z + g_I I_z) \quad (1)$$

where the first two terms arise from the interaction of the nuclear magnetic dipole and nuclear electric quadrupole, respectively, with the orbital electrons. For a spherically symmetric charge distribution of the orbital electrons, i.e., for  $J=0$  or  $1/2$ , the quadrupole term vanishes. The last two terms in (1) are the interaction of the external field, taken in the  $z$  direction, with the electrons and nucleus, respectively. Terms due to moments of a higher order than the quadrupole moment are omitted in (1) for simplicity. For the case of the  $^2P_{3/2}$  state of indium ( $I=9/2$ ), the resulting secular determinant of forty rows and columns factors according to the total magnetic quantum number, giving rise to two secular equations of the first, second, and third degrees, and seven quartic equations. These equations may be solved for the energies of the levels in terms of the constants  $a$ ,  $b$ ,  $g_J$ ,  $g_I$  and  $H$ . The higher degree equations may be simplified by use of the substitution

$$W/a = W_0/a + \delta W/a, \quad (2)$$

where  $W_0/ha$  is the weak field, first-order approximation to the energy of the level. Using Hamilton's<sup>2</sup> values of  $a$  and  $b$ , and the value of  $g_I$  obtained by Hardy and

Millman,<sup>3</sup> several selected energy levels are plotted as a function of  $x = \mu_0 g_J H/ha$  in Fig. 1. The levels are labelled in both the weak field ( $F, m_F$ ) and strong field ( $m_I, m_J$ ) notations. The energies of these particular levels (which comprise less than one-fourth of the total number) were calculated in detail primarily because the information was necessary to the measurement of the ratio  $g_J(^2P_{3/2}\text{In}^{115})/g_J(^2P_{3/2}\text{In}^{113})$ , which is described in the following paper. In the apparatus used in the present series of experiments, it is possible to observe only those lines which are designated in the high field notation by  $\Delta m_J = \pm 1, \Delta m_I = 0$ . It is possible to observe the lines at a field strength at which  $m_J$  and  $m_I$  are not appropriate quantum numbers; however, the transitions observable at low and intermediate fields must be capable of an "adiabatic" transformation to the high field case. The states shown in Fig. 1 are, therefore, the particular states which give rise to the observable transitions  $\Delta F = 0, \Delta m_F = \pm 1$  at low and intermediate magnetic fields. No need has arisen in any part of this work to calculate at intermediate fields the energies of any other levels.

For zero external magnetic field, the first two terms in (1) are diagonal in the  $F, m_F$  representation ( $F = I + J, \dots, I - J$ ), with matrix elements given by  $aC/2$

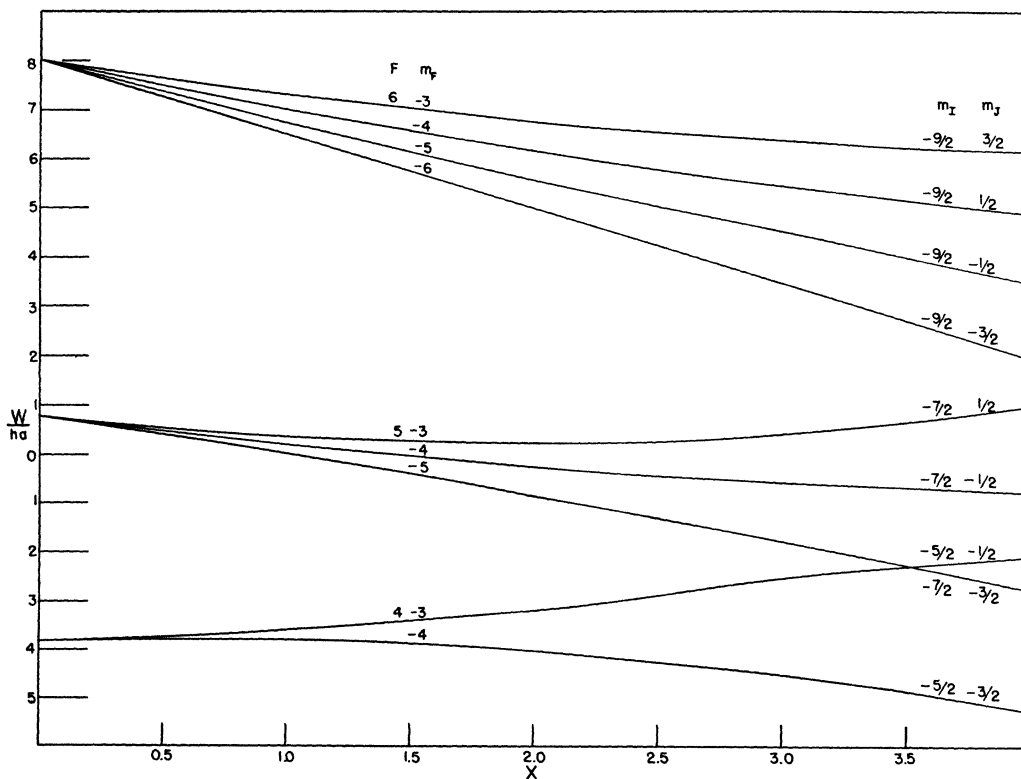


FIG. 1. The variation with magnetic field of certain energy levels of  $\text{In}^{115}$  in the  $^2P_{3/2}$  state. The energies of the levels have been calculated exactly for the values of  $x$  shown in the figure. A magnetic field strength of approximately 130 gauss corresponds to  $x = 1$ .

<sup>6</sup>H. B. G. Casimir, *On the Interaction between Atomic Nuclei and Electrons* (Teyler's Tweede Genootschap, Netherlands, 1936).

+ $bC(C+1)$  where  $C=F(F+1)-I(I+1)-J(J+1)$ . There are, then, four levels corresponding to the four possible values of  $F$ . If  $C$  is evaluated for each  $F$ , one obtains expressions for the energy of each level in terms of  $a$  and  $b$ . The allowed frequencies ( $\Delta F=\pm 1$ ) corresponding to the energy differences between these levels are:

$$\begin{aligned} (W_6 - W_5)/h &= f_6 = 6a + 192b \\ (W_5 - W_4)/h &= f_5 = 5a - 60b \\ (W_4 - W_3)/h &= f_4 = 4a - 192b, \end{aligned} \quad (3)$$

where  $a$  and  $b$  are expressed in frequency units. These frequencies are, for the  $^2P_{3/2}$  state of indium, in the range 650-1750 mc/sec.

In general, atomic beam transitions cannot be observed in zero magnetic field. The necessity for a field arises from the fact that transitions are detected by observing changes in magnetic moment which accompany them. In order to detect changes in magnetic moment, the atoms must pass through inhomogeneous magnetic fields before and after undergoing a transition. If the field strength in the region in which the transition takes place is zero, there will be induced "non-adiabatic transitions." A field of sufficient magnitude to avoid this must therefore be present in the transition region.

At very low magnetic fields, the splitting of each  $F$ -level into  $(2F+1)$  levels is adequately described, without recourse to the exact solutions of the secular equations, by the first-order (Zeeman) approximation,

$$W_{F,m_F} = W_F + \mu_0 m_F g_F H \quad (4)$$

where

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

Since  $|g_I/g_J| \approx 0.0005$  for the  $^2P_{3/2}$  state of both indium isotopes, the second term in the expression for  $g_F$  may be neglected within the precision of the present experiment. The pattern of the line frequencies arising from transitions between the levels described by Eq. (4) is obtained by applying the selection rules for magnetic dipole radiation, namely:  $\Delta F=0, \pm 1$ ;  $\Delta m_F=0, \pm 1$ .<sup>†</sup> At very weak magnetic fields the transitions for which  $\Delta F=0, \Delta m=\pm 1$ , give rise to four lines, since each  $F$ -level is characterized by a particular value of  $g_F$ . Observation of these lines contributes no information about the nuclear interaction constants, but does aid in determining the nuclear spin, and if the spin is known, can serve to determine the magnetic field at which observations have been made. The transitions for which  $\Delta F=\pm 1, \Delta m=\pm 1$  ( $\pi$ -lines) are those caused by the component of the oscillating magnetic field perpendicular to the constant field,  $H$ , while transitions

<sup>†</sup> Henceforth  $m_F$  will be referred to as  $m$ .

for which  $\Delta F=\pm 1, \Delta m=0$  ( $\sigma$ -lines) result from the component parallel to  $H$ . Figure 2 shows the field dependence of the frequencies of the  $\pi$ -lines in the Zeeman region. The figure is, of course, applicable only to the case for which  $I=9/2$ . For convenience, the diagram has been cut off at  $H=1$  gauss. The lines are labelled in accord with the convention  $(F,m) \leftrightarrow (F',m')$ . The observable lines are indicated by solid lines. Certain of the  $\pi$ -lines cannot be observed because the corresponding high field lines are those for which  $\Delta m_J=0, \Delta m_I=\pm 1$ . These are the dashed lines in Fig. 2.

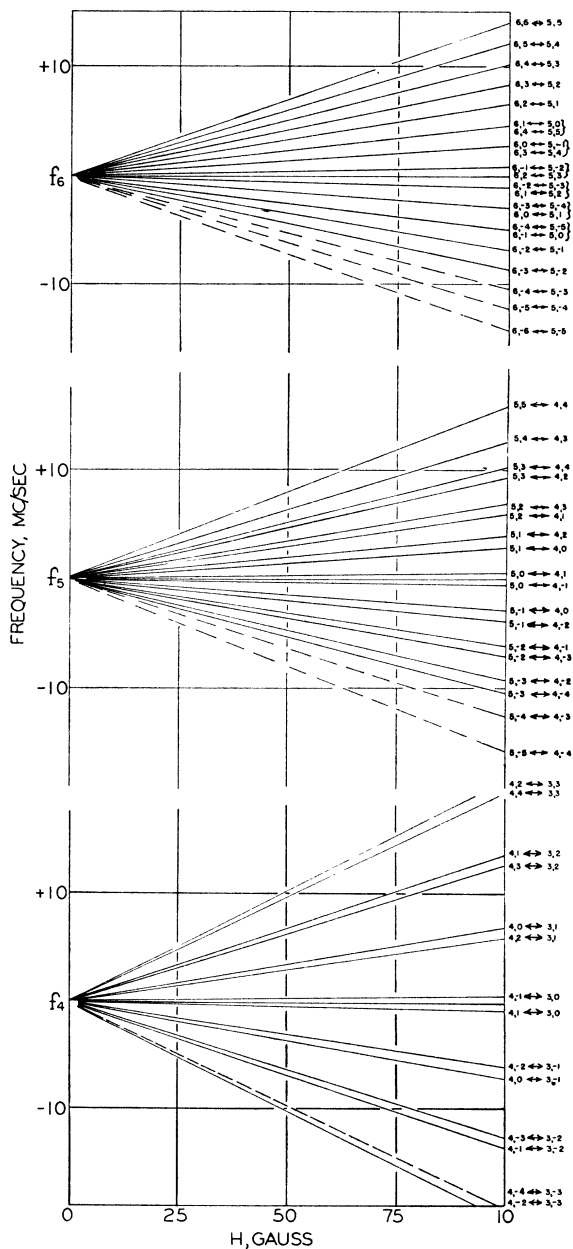


Fig. 2. The field dependence in the Zeeman region of the frequencies of the  $\pi$ -lines ( $\Delta F=\pm 1, \Delta m_F=\pm 1$ ) in the spectrum of the  $^2P_{3/2}$  state of  $\text{In}^{115}$ .

TABLE I. Results of the measurements of  $f_4^{115}$ .

Transition	Observed frequency, mc	Approx. $H$ , gauss	$f_4^{115}$ mc
4, -1 ↔ 3, 0	669.102	2.10	668.969
4, 1 ↔ 3, 0	668.890		
4, 2 ↔ 3, 1	670.275		668.974
4, -2 ↔ 3, -1	667.721		
4, 0 ↔ 3, 1	670.469		668.976
4, 0 ↔ 3, -1	667.538		
4, 3 ↔ 3, 2*	671.625		668.963
4, 1 ↔ 3, 2*	671.825		
4, -1 ↔ 3, 0	669.187	3.30	668.952
4, 1 ↔ 3, 0	668.854		
4, 2 ↔ 3, 1	670.996		668.945
4, -2 ↔ 3, -1	667.014		
4, 0 ↔ 3, 1	671.332		668.946
4, 0 ↔ 3, -1	666.695		
4, -1 ↔ 3, 0	669.527	6.60	668.954
4, 1 ↔ 3, 0	668.911		
			Mean 668.960

\* These lines do not constitute a symmetrical pair. A value of  $f_4$  was computed from each of these lines using the field strength,  $H$ , obtained from the other lines observed in the same run. The mean of the two values of  $f_4$  is entered in the last column.

The validity of Eq. (4) at any given field strength can be tested simply by determining whether or not, within experimental error, the line frequency pattern does have the predicted symmetry. For fields at which the departures from this symmetry become evident, but are small, the second-order approximation can be used to introduce a term in  $H^2$  to Eq. (4), which then becomes

$$W_{F,m} = W_F + \mu_0 m g_F H + \left( \frac{\alpha^2}{W_F - W_{F+1}} + \frac{\beta^2}{W_F - W_{F-1}} \right) (\mu_0 g_F H)^2, \quad (5)$$

where

$$\alpha^2 = \frac{(F+1-I+J)(F+1+I-J)(I+J+2+F)}{4(F+1)^2(2F+1)(2F+3)} \times (I+J-F)[(F+1)^2 - m^2]$$

$$\beta^2 = \frac{(F-I+J)(F+I-J)(I+J+1+F)}{4F^2(2F-1)(2F+1)} \times (I+J+1-F)(F^2 - m^2)$$

In this experiment, all line frequencies were observed at magnetic fields at which the second-order terms were small corrections to the total frequency (approximately

1 part in  $10^4$  or less). The lines occur as pairs,  $F, m \leftrightarrow F', (m \pm 1)$  and  $F, -m \leftrightarrow F', -(m \pm 1)$ . To the first order, the mean frequency of a pair is equal to the zero field frequency,  $(W_F - W_{F'})/h$ . The magnetic field can readily be determined by noting that the difference in the frequencies of any symmetrical pair of lines is  $2[mg_F - (m \pm 1)g_{F'}]\mu_0 H/h$ . The magnetic field can be calculated from these differences with sufficient accuracy to enable the second-order correction to be made within the error of the observations.

## APPARATUS AND PROCEDURE

### General Discussion

The principal difficulty encountered in this experiment may be seen from elementary considerations of the properties of the atoms involved. The relative abundances of the indium isotopes are 95.5 percent for  $\text{In}^{115}$ , and 4.5 percent for  $\text{In}^{113}$ . The metastable  $^2P_{3/2}$  state of the atoms lies  $2212.6 \text{ cm}^{-1}$  ( $=\Delta\nu'$ ) above the ground state. The ratio of the number of atoms in the higher state to the number in the lower state is given by the Boltzmann factor multiplied by the ratio of the statistical weights, that is, by  $2e^{-hc\Delta\nu'/kT}$ . At an oven temperature of about  $1500^\circ\text{K}$ , required to give a beam of indium by use of conventional techniques, the expected abundance of the metastable state is about 20 percent. Since there are  $(2J+1)(2I+1)$ , or 40, magnetic levels in the  $^2P_{3/2}$  state, and a radiofrequency transition involves only two levels, with a maximum of about 75 percent<sup>7</sup> of the atoms in these levels reorienting, the maximum intensity change due to a transition in  $\text{In}^{115}$  is approximately  $95.5 \times 0.2 \times 0.05 \times 0.75$ , or 0.72 percent of the total beam intensity. The maximum change resulting from a transition in  $\text{In}^{113}$  is  $4.5 \times 0.72/95.5$ , or 0.034 percent of the total intensity. Changes in intensity of the order of those expected in  $\text{In}^{115}$  can, in general, be observed under the conditions of beam steadiness usually obtained in an atomic beam apparatus. However, intensity changes of the magnitude expected in  $\text{In}^{113}$  require the use of some method which removes from the beam a large fraction of the atoms in states not involved in the transitions to be studied, and thus increases the signal to noise ratio at the detector to a value at which measurements are possible. There have been employed in the past several different methods for this purpose. The primary considerations in the selection of one of these methods in a particular application are the properties of the atoms to be studied, and the information to be derived from the experiment. As stated previously, the constants,  $a^{115}$  and  $b^{115}$ , were measured by Hamilton<sup>2</sup> with a precision of about 1 percent, and a correspondingly high upper limit placed on  $c^{115}$ . In order to accomplish the purpose of the present experiment, it was necessary to remeasure all of these same quantities with considerably greater precision. In the event that the more precisely meas-

<sup>7</sup> H. C. Torrey, Phys. Rev. **59**, 293 (1941).

ured  $a^{115}$  and  $b^{115}$  are sufficient to describe the h.f.s., then  $a^{113}$  may be calculated directly from the known<sup>3</sup> ratio  $a^{115}/a^{113}$ , and there remains only  $b^{113}$  to be measured. For this reason, in addition to intensity considerations, different procedures were employed in the study of the two isotopes.

### Method—Indium<sup>115</sup>

The main features of the apparatus and techniques used in this experiment on In<sup>115</sup> have been described in detail in previous papers.<sup>3,8,9</sup> Essentially, a beam of atoms issues from a narrow source slit, and is first deflected in an inhomogeneous magnetic field,  $H_A$ . Some of these atoms then pass through a collimating slit and an homogeneous magnetic field,  $H_C$ , on which is superimposed a weak radiofrequency oscillating field. Finally, the atoms are deflected in the opposite direction to that in magnet  $A$  by a second inhomogeneous field,  $H_B$ , and, if the deflection produced at the detector by  $B$  is just equal to that produced by  $A$ , strike a detector filament. The condition for an atom to have zero net deflection at the detector in the present apparatus requires that its effective dipole moment be the same in both the  $A$  and  $B$  magnets. If an atom is caused to undergo a transition in the  $C$  magnet from one energy level to another, the condition for zero net deflection usually no longer holds, and the result is a diminution of beam intensity at the detector. It will be seen from the expressions for the energy levels in the previous section that the frequency required to produce a transition depends only on  $\| H_C$ , and the properties of the atoms in the beam. Consequently, for a given value of  $H$ , the line frequencies are completely determined, and are the only data of the experiment.  $\|$  The particular magnitude of  $H$  at which observations are made depends on the information desired from the experiment. The magnitudes of  $H_A$  and  $H_B$  are chosen to satisfy the deflection conditions imposed by the properties of the atoms and the dimensions of the apparatus, and are, in general, not critical. The lengths of the  $A$  and  $B$  magnets used in this experiment were 10.4 and 16.5 cm, respectively, with a ratio of gradient to field equal to 3.2 cm<sup>-1</sup>. The widths of the oven and collimating slits and of the detector were about 0.02 mm. The actual length of the  $C$  magnet in which transitions took place was about 2 cm, and therefore the theoretical half-width of the lines was about 50 kc/sec. This width was observed in the experiments.

With this arrangement, and the oscillating magnetic field designed to be perpendicular to the homogeneous field, almost all transitions,  $\Delta F = \pm 1$ ;  $\Delta m = \pm 1$ , were

<sup>8</sup> Kusch, Millman, and Rabi, Phys. Rev. **57**, 765 (1940).

<sup>9</sup> G. Becker and P. Kusch, Phys. Rev. **73**, 584 (1948).

$\|$   $H_C$  will henceforth be denoted by  $H$ .

$\|$  In order to apply the second-order corrections (discussed above) to the measured frequencies, it is necessary to know only the approximate value of  $H$ , which may be calculated from the measured frequencies of certain lines.

TABLE II. Results of the measurements of  $f_0^{115}$ .

Transition	Observed frequency, mc	Approx. $H$ , gauss	$f_0^{115}$ mc
5,0 $\leftrightarrow$ 4,1	1117.501	7.27	1117.150
5,0 $\leftrightarrow$ 4,-1	1116.823		
5,0 $\leftrightarrow$ 4,1	1117.416	5.44	1117.155
5,0 $\leftrightarrow$ 4,-1	1116.908		
			Mean 1117.153

TABLE III. Results of the measurements of  $f_0^{115}$ .

Transition	Observed frequency, mc	Approx. $H$ , gauss	$f_0^{115}$ mc
(6,-1 $\leftrightarrow$ 5,-2)(6,2 $\leftrightarrow$ 5,3)	1753.088	4.10	1752.714
(6,1 $\leftrightarrow$ 5,2)(6,-2 $\leftrightarrow$ 5,-3)	1752.331		
(6,1 $\leftrightarrow$ 5,0)(6,4 $\leftrightarrow$ 5,5)	1754.613		1752.695
(6,-1 $\leftrightarrow$ 5,0)(6,-4 $\leftrightarrow$ 5,-5)	1750.795		
(6,0 $\leftrightarrow$ 5,-1)(6,3 $\leftrightarrow$ 5,4)	1753.854		1752.702
(6,0 $\leftrightarrow$ 5,1)(6,-3 $\leftrightarrow$ 5,-4)	1751.558		
(6,-1 $\leftrightarrow$ 5,-2)(6,2 $\leftrightarrow$ 5,3)	1753.288	6.26	1752.712
(6,1 $\leftrightarrow$ 5,2)(6,-2 $\leftrightarrow$ 5,-3)	1752.120		
(6,-1 $\leftrightarrow$ 5,-2)(6,2 $\leftrightarrow$ 5,3)	1753.460	8.44	1752.688
(6,1 $\leftrightarrow$ 5,2)(6,-2 $\leftrightarrow$ 5,-3)	1751.885		
			Mean 1752.702

observable.\*\* Hence all frequencies occurring in Eqs. (3) could be measured, and, in consequence, the constants  $a$ ,  $b$  and  $c$  for In<sup>115</sup> could be evaluated.

The radiofrequency current was supplied by a grounded grid coaxial line oscillator (Radar Jammer T-85/APT-5) using a 3C22 lighthouse tube. This oscillator was used for all measurements in the frequency range 650-1750 mc. Frequency measurements were made with a General Radio heterodyne frequency meter, Type 620A. No attempt was made to determine the shapes of the resonance lines, but the continuously variable oscillator was set at the frequency producing maximum change in beam intensity, and the frequency meter was simultaneously tuned to zero beat. The frequency of the quartz crystal against which the frequency meter was calibrated was in turn calibrated against a signal from WWV, and proper corrections made in the measured values of the transition frequencies.

\*\* In general, the  $A$  and  $B$  magnets are designed to operate at a fairly high magnetic field. This serves to keep the beam trajectory short, and thus to give a high level of beam intensity and improved stability. In the high fields, however, the  $m_J$  and  $m_I$  quantization is appropriate, and a significant moment change occurs only when  $\Delta m_J = \pm 1$ . No loss of information results from this circumstance since in the low magnetic field of the transition region many more lines can be observed than are required to determine the desired constants.

TABLE IV. Results of the measurements of  $f_6^{113}$  and  $f_6^{115}$ .

I	II	III	IV	V	VI
Transition	Observed frequency 115, mc	$f_6^{115}$ , mc	Observed frequency 113, mc	$f_6^{113}$ , mc	(II-IV) mc
5,2 $\leftrightarrow$ 4,3	1123.834		1122.494		1.340
5,-2 $\leftrightarrow$ 4,-3	1110.724	1117.165	1109.384	1115.825	1.340
5,2 $\leftrightarrow$ 4,1	1122.779		1121.480		1.299
5,-2 $\leftrightarrow$ 4,-1	1111.453	1117.126	1110.142	1115.821	1.311
5,1 $\leftrightarrow$ 4,2	1120.727		1119.365		1.362
5,-1 $\leftrightarrow$ 4,-2	1113.699	1117.154	1112.343	1115.796	1.356
5,1 $\leftrightarrow$ 4,1	1120.174		1118.892		1.282
5,-1 $\leftrightarrow$ 4,-1	1114.155	1117.152	1112.821	1115.844	1.334
5,1 $\leftrightarrow$ 4,0	1119.753		1118.413		1.340
5,-1 $\leftrightarrow$ 4,0	1114.526	1117.143	1113.186	1115.780	1.340
5,0 $\leftrightarrow$ 4,1	1117.615		1116.253		1.362
5,0 $\leftrightarrow$ 4,-1	1116.720	1117.132	1115.346	1115.803	1.374
5,0 $\leftrightarrow$ 4,0*	1117.154	1117.149	1115.786	1115.781	1.368
	Mean	1117.148		1115.807	

\* This is the center line of the pattern, and, consequently, gives the zero field frequency directly after the application of a quadratic field correction.

### Method—Indium<sup>113</sup>

An attempt was made to observe the spectrum of In<sup>113</sup> by utilizing a mass spectrometer as the detecting element of the atomic beam apparatus. If the use of the mass spectrometer does not result in an appreciable loss of intensity, then the problem of observing the spectrum of In<sup>113</sup> with the mass spectrometer is identical to the problem of observing the spectrum of In<sup>115</sup> in the total beam. However, a substantial loss of intensity does occur in the mass spectrometer, and the spectrometer itself introduces new instabilities to the detected beam, so that this method was found to be less adequate than the method subsequently described for a careful analysis of the spectrum of In<sup>113</sup>. Nevertheless, it should be mentioned that observation, by use of the mass spectrometer, of certain lines at intermediate magnetic fields did serve to determine a value of  $b^{113}$  which is in agreement with that determined by other means, but of somewhat lower precision.

The experimental arrangement finally used for the study of In<sup>113</sup> was almost identical with that first used by Zacharias<sup>10</sup> in the measurement of the spin and h.f.s. separation of  $K^{40}$ . In this method the deflections produced by the two inhomogeneous magnetic fields are not equal and opposite for an atom in a fixed state. Thus the beam intensity observed at the detector is effectively zero. Atoms are refocused at the detector only when a transition occurs in the transition region

<sup>10</sup> J. R. Zacharias, Phys. Rev. **61**, 270 (1942).

which results in some particular moment change. In general, relatively few transitions are observable for any given instrumental arrangement, and the amount of information which may be obtained from an experiment is considerably more limited than in the case of the experiment described for In<sup>115</sup>. The arrangement employed in the present instance could detect transitions which resulted in a change in the sign of the moment but no change in the magnitude. Since the atoms are substantially in the Paschen-Back region in the deflecting fields, only transitions whose high field equivalent is  $(m_J = \frac{1}{2}, m_I) \leftrightarrow (m_J = -\frac{1}{2}, m_I)$  could be detected. Of the transitions  $\Delta F = \pm 1, \Delta m = 0, \pm 1$ , only those arising between the levels  $F = 4, 5$  were observable. Other lines of the type  $\Delta F = 0, \Delta m = \pm 1$  were also observable; in the present case these lines were not investigated principally because of the difficulty in locating a line of small absolute intensity by exploration. It should be emphasized that a rearrangement of components of the apparatus would make it possible to observe other transitions as well.

The actual experimental arrangement differed from that used for In<sup>115</sup> in only two essentials. First, the direction of the gradient of the  $B$  magnet was changed to be the same as that in the  $A$  magnet. This caused most of the atoms in the  $^2P_{3/2}$  state and some in the  $^2P_{1/2}$  state to be so strongly deflected that they did not strike the detector. Second, a stop-wire was placed in the direct line of the beam to eliminate the remainder of the atoms which, because of their very small moments or very high velocities, were not deflected sufficiently to miss the detector. Under these conditions, the background beam intensity was about 0.1 percent of the total beam striking the detector with the magnets off and the wire not in the beam path. If a transition took place in the  $C$  magnet which was accompanied only by a change of sign of the effective magnetic moment, then the atoms undergoing that transition were deflected by the  $B$  magnet in the proper direction to enable them to strike the detector, providing their deflection was great enough to cause them to miss the stop-wire. The diameter of the stop-wire, and its position in the  $B$  magnet, which would allow passage of the reoriented atoms were calculated from the deflection properties of the apparatus.

## RESULTS

### Indium<sup>115</sup>

The results of the experiments on In<sup>115</sup> are presented in Tables I-III. The first column in each table gives the observed transition, labelled by the weak field quantum numbers  $F$  and  $m$ . It is to be noted that coincidences occur for the lines  $(6, m) \leftrightarrow (5, m')$ . The line frequencies actually differ by terms of the magnitude of  $g_I \mu_0 H / h$ , but at the magnetic fields at which measurements were made these terms are of the order of a few kc/sec., and the lines cannot be resolved. In the

second column are shown the observed frequencies of line pairs which are symmetrically situated (except for second-order corrections) about the zero field frequency. The third column shows the approximate field strength at which the observations were made. Each pair of lines determines a value of the field, as discussed previously. The mean of these values, with due weight given to the fact that the accuracy of a value depends directly on the frequency difference between symmetrical lines, is given in the third column. It should be emphasized that these values of  $H$  are used only to apply second-order corrections to the observed frequencies. Finally, the last column in each table lists the zero field frequency obtained from each line pair after all corrections are made. From general experimental considerations, we estimate the uncertainty in the absolute values of the zero field frequencies to be 1 part in  $5 \times 10^4$ . The relative values are, however, to a first approximation independent of any error in the calibrating crystal of the frequency meter, and consequently we assign an uncertainty of 1 part in  $7.5 \times 10^4$  to the relative values of the zero field frequencies.

Using the mean values of the zero field frequencies, and Eqs. (3), the constants  $a$  and  $b$  may be calculated. The results of a least squares solution are  $a^{115} = 242.165 \pm 0.002$  mc/sec., and  $b^{115} = 1.56098 \pm 0.00006$  mc/sec. If these values are substituted in Eqs. (3), the calculated frequencies agree with those observed within the precision of the measurements, i.e., within 1 part in  $7.5 \times 10^4$ .

It is also possible to place an upper limit on the magnitude of interactions due to nuclear moments of a higher order than the quadrupole moment. The inclusion of the interaction of a magnetic octupole moment with the orbital electrons requires the addition of the term<sup>11</sup>  $c(C^3 + 4C^2 + 4C/5)$  to the equation for the energy of the h.f.s. levels at zero external field. The terms  $3084.4c$ ,  $345.5c$ , and  $3186.6c$  must then be added to the expressions for  $f_4$ ,  $f_5$ , and  $f_6$ , respectively. The three equations then determine  $a$ ,  $b$ , and  $c$  to be  $a = 242.163$ ,  $b = 1.56098$ , and  $c = 6.8 \times 10^{-6}$ , all in mc/sec. This value of  $c^{115}$  is too small to be meaningful, and thus it may be concluded that no observable octupole interaction occurs to within the precision of the measurements.

### Indium<sup>113</sup>

The results of the run on In<sup>113</sup> are shown in Table IV. Since lines in the spectrum of the abundant isotope were observed as a check on the procedure, these observations are included in the table. As before, the first column lists the transitions. The second and fourth columns give the observed frequencies of line pairs in In<sup>115</sup> and In<sup>113</sup>, respectively. The mean field strength calculated from the observed frequencies is 9.36 gauss. In the third and fifth columns are shown the values of

the zero field frequencies of the two isotopes obtained from the observed line pairs. Finally, the sixth column gives the frequency separation  $f^{115} - f^{113}$  for each of the observed lines. This separation is practically independent of the quadratic field correction. It serves to indicate the consistency of the interpretation of the spectra, and the precision of measurement. It will be noted that the mean value of  $f_5^{115}$  in Table IV is in good agreement with that of Table II.

Using  $a^{115}/a^{113} = 1.00224 \pm 0.00010$ , and  $a^{115} = 242.165$  mc/sec., we have,  $a^{113} = 241.624 \pm 0.024$  mc/sec. From the equation for  $f_5$ , we then obtain,  $b^{113} = 1.53855 \pm 0.00015$  mc/sec., and therefore,  $b^{115}/b^{113} = 1.0146 \pm 0.0001$ , where the uncertainty in  $a^{113}$  and  $b^{113}$  is that of the ratio of the  $a$ 's.

It is of interest to note that  $\sigma$ -lines were observed, and are listed in the data of Table IV, although the oscillating magnetic field at the position at which a perfectly aligned beam traverses the r-f region is perpendicular to the homogeneous field, and only  $\pi$ -lines had been observed previously. Since the  $\sigma$ -lines also exhibited a complex structure, a careful investigation was made to account for their presence. Figure 3 shows a detailed plot of a portion of the In<sup>115</sup> spectrum, taken under the same conditions as the data in Table IV. The two outside lines are  $\pi$ -lines whose width is determined by the transit time of the atoms in the r-f region. The mean frequency of the symmetrical struc-

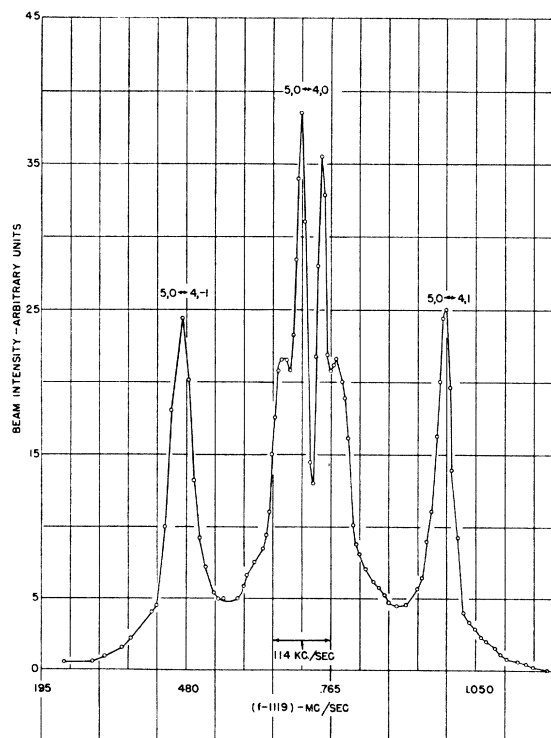


FIG. 3. A detailed plot of a portion of the spectrum of In<sup>115</sup>. This curve was obtained under the special conditions described in the text.

<sup>11</sup> H. B. G. Casimir and H. Korsching, *Zeits. f. Physik* **103**, 434 (1936).

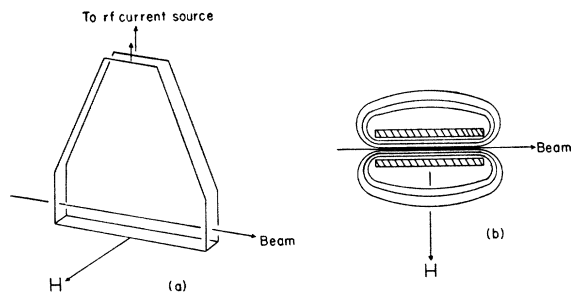


FIG. 4. The arrangement for superimposing the oscillating magnetic field,  $H_{r-t}$ , on the constant field,  $H$ . (a) The general arrangement of the r-f circuit and the beam. (b) A cross section of the circuit in a plane traversed by the beam.

ture in the center is that of a  $\sigma$ -line. It was found that the relative intensities of the  $\pi$ - and  $\sigma$ -lines were critically dependent on the strength of the oscillating magnetic field. At an r-f current of approximately one-quarter that at which the data in Fig. 3 were taken, the center structure had almost entirely disappeared, and the  $\pi$ -lines increased in intensity by a factor of about two. The positions of the  $\pi$ -lines remained undisturbed. The relative intensities were also found to be dependent on the position and direction of the beam path in the oscillating magnetic field. Changes in the position and direction of the beam path produced effects similar to those resulting from changes in the r-f current.

The presence of the  $\sigma$ -lines, and the details of their structure can be explained by considerations due to Ramsey,<sup>12</sup> who has calculated the resonance spectrum to be expected when the oscillating magnetic field is concentrated in two regions at the beginning and end of the homogeneous field, and of a length small compared to the distance between the two regions. The general arrangement of the r-f circuit in space is shown in Fig. 4a. A cross section of the circuit in a plane traversed by the beam is shown in Fig. 4b. It is evident from the latter of these figures that a beam which traverses the circuit symmetrically does not pass through a region in which the r-f magnetic field is parallel to the constant field,  $H$ . However, if either the circuit or the beam is misaligned such as to cause an asymmetrical traversal, the beam will pass through regions in which the r-f field has a component parallel to  $H$ . In general, both the length of and the r-f amplitude in such regions will be so small that the probability of transition is also small. If, however, the r-f amplitude is much larger than the optimum required to produce  $\pi$ -transitions in the center region of the r-f circuit, it is possible to induce  $\sigma$ -transitions in the two end regions. The complex structure observed in the  $\sigma$ -line is then essentially the result of interference between the probabilities of transition in each of the two short, end

regions. The exact details of the pattern depend on the relative amplitudes of the r-f in the two regions which contain the appropriate r-f field, and on the relative phases. If the amplitudes are equal, and the fields are out of phase by  $180^\circ$ , a structure such as that shown in Fig. 3 is observed. The normal frequency of the center of the  $\sigma$ -line corresponds to the central minimum of the pattern in Fig. 3, and a number of equally spaced maxima appear on either side of the center. The separation of the two central peaks is calculated to be approximately  $1.2\alpha/l$ , where  $\alpha$  is the most probable velocity of atoms in the oven, and  $l$  is the distance between the oscillating fields. In the present case, this quantity is estimated to be about 30 kc/sec., which compares favorably with the observed value of 40 kc/sec.

With the arrangement as shown in Fig. 4, the details of the structure of the  $\sigma$ -line depend critically on accidental factors in the geometry of the apparatus. In the most general case, effects of the indicated type arising from instrumental errors would result in a distortion of the line shape not subject to easy interpretation. It is evident that with the present arrangement it is not possible to observe similar effects for the  $\pi$ -lines.

#### THE QUADRUPOLE MOMENTS

The relations between the interaction constants and the nuclear moments have been given by Casimir.<sup>6</sup> The main difficulty in the application of these relations arises from the necessity of evaluating the terms  $\langle\langle r^{-3} \rangle\rangle_{Av}$ , and  $\langle\langle [3 \cos^2\theta - 1]/r^3 \rangle\rangle_{J,J} \rangle_{Av}$ , which occur in the expressions for  $a$  and  $b$ , respectively. It has been pointed out by Davis, Feld, Zabel, and Zacharias,<sup>13</sup> that, in the approximation in which the electron wave function is separable, the dependence of both  $a$  and  $b$  on  $r$  is through the same factor  $\langle\langle r^{-3} \rangle\rangle_{Av}$ , and the ratio  $b/a$  is therefore independent of the radial factor. One can then write

$$Q = -\frac{8\mu_0^2}{3e^2} \frac{F b I(2I-1)(2J-1)(L+1)(2L+3)}{g_I R a (J+1)}, \quad \text{for } J=L+\frac{1}{2} \quad (6)$$

where  $R$  and  $F$  are small relativistic corrections given by Casimir. This result is the same as that given by Davis *et al.* for chlorine, except that  $b$  is defined differently here,<sup>††</sup> and that the change in the sign of  $b$  required for chlorine is not required in this case. The sign changes occur because in the case of chlorine an electron shell which lacks a single electron of being closed is considered, while for indium there is a single valence electron ( $5p$ ) beyond the ( $5s$ ) closed shell.

Observation<sup>6</sup> of the spectrum of the  $^2P_{3/2}$  state of  $\text{In}^{115}$  has yielded directly the ratio  $g_I(\text{In}^{115})/g_J(^2P_{3/2}) = -9.95$

<sup>13</sup> Davis, Feld, Zabel, and Zacharias, *Phys. Rev.* **76**, 1076 (1949). We are also indebted to the group at M.I.T. for sending us a prepublication copy of their paper.

<sup>††</sup> If  $b'$  denotes the constant used by the M.I.T. group, and  $b$  the constant used here, then  $b' = 8/3IJ(2I-1)(2J-1)b$ .

<sup>12</sup> N. F. Ramsey, *Phys. Rev.* **75**, 1326A (1949); **76**, 996 (1949). We are also indebted to Professor Ramsey for a private communication.



$\times 10^{-4}$ , where  $g_J(^2P_{3/2})=0.66599$ . For indium  $F/R$  is 0.94930. Insertion of the numerical quantities in Eq. (6) gives  $Q^{115}=1.161 \times 10^{-24}$  cm<sup>2</sup>.

It is to be noted that the diamagnetic correction customarily applied to observed nuclear  $g$  values has not been applied here. It is not at all clear from the present state of the theory that this correction should be made. It seems doubtful that the theory is sufficiently good to enable one to calculate quadrupole moments with a precision limited only by uncertainties in observed quantities. It should also be noted that in several instances the measured ratio of the h.f.s. separations of the same states of two isotopes is not simply derived from the measured ratio of the nuclear magnetic dipole moments.<sup>14</sup> The presence of such an effect in indium would cause the calculated quadrupole moment to be in error. For atoms with the same spin, the magnitude of the effect presumably is small compared to the error arising from uncertainties in the application of the diamagnetic correction.

<sup>14</sup> J. E. Nafe and E. B. Nelson, *Phys. Rev.* **73**, 718 (1948); F. Bitter, *Phys. Rev.* **76**, 150 (1949); P. Kusch and A. K. Mann, *Phys. Rev.* **76**, 707 (1949).

To a very high order of approximation

$$Q^{115}/Q^{113}=b^{115}/b^{113}=1.0146$$

which leads to the value  $Q^{113}=1.144 \times 10^{-24}$  cm<sup>2</sup>, where the diamagnetic correction has not been made.

### CONCLUSIONS

The h.f.s. of the  $^2P_{3/2}$  state of indium can be described exactly in terms of two interaction constants. No experimental evidence has been found to indicate the existence of a nuclear moment of higher order than the quadrupole moment. This is the heaviest atom in which a critical search has been made for higher order nuclear moments.

The addition of two neutrons to In<sup>113</sup> produces an isotope which differs only very slightly from In<sup>113</sup> in certain measurable nuclear properties. The spins are identical, the magnetic dipole moments differ by 0.22 percent, and the electric quadrupole moments differ by 1.46 percent. This is the greatest degree of similarity observed for any pair of isotopes of odd mass differing in mass by two units.

## Further Data on the Spin Gyromagnetic Ratio of the Electron\*†

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The atomic beam magnetic resonance method has been used to compare the  $g_J$  values of In in the  $^2P_{3/2}$  and  $^2P_{1/2}$  states in order to obtain a value of the spin gyromagnetic ratio of the electron. The determination of the  $g_J$  ratio involved only the measurement of frequencies of lines in the h.f.s. spectra of both states at constant magnetic field. Lines with widely different frequency dependence on field were selected to avoid possible systematic errors arising from inhomogeneities in the field. The result of these measurements is  $g_J(^2P_{3/2})/g_J(^2P_{1/2})=2(1.00200 \pm 0.00006)$ . Assuming Russell-Saunders coupling, and  $g_L=1$ , the spin gyromagnetic ratio of the electron may be calculated. The result is

$g_S=2(1.00133 \pm 0.00004)$ , which is to be compared with the value  $g_S=2(1.00119 \pm 0.00005)$  obtained by Kusch and Foley. The discrepancy is greater than the sum of the experimental errors. The  $^2P_{1/2}$  state of In is believed to be free of significant perturbations which might affect the total electronic  $g$  value. However, in view of possible perturbations of the  $^2P_{3/2}$  state, the agreement must be considered as very good. The present result confirms the conclusions of the previous experiment, both as to the existence of the intrinsic magnetic moment of the electron, and as to its approximate magnitude.

### INTRODUCTION

IN a recent experiment, Kusch and Foley<sup>1</sup> have measured the ratio of the electron spin  $g$  value ( $g_S$ ) to the orbital  $g$  value ( $g_L$ ) by comparing the total electronic  $g$  values of gallium in the  $^2P_{1/2}$  and  $^2P_{3/2}$  states, indium in the  $^2P_{1/2}$  state, and sodium in the  $^2S_{1/2}$  state. Three independent intercomparisons gave three values for the ratio  $g_S/g_L$  which are in agreement to within 7

parts in  $10^5$ . In addition, the experimental result is in good agreement with the result obtained by Schwinger<sup>2</sup> from a first-order theoretical investigation.

The method employed by Kusch and Foley avoids the difficulty of producing a magnetic field which is known to a high order of precision in terms of absolute standards, and which is required for a precision measurement of the absolute value of a gyromagnetic ratio. However, the validity of any individual determination of the ratio  $g_S/g_L$  by the method of Kusch and Foley may be limited by deviations of the properties of the atomic systems from the simple description implicit in

\* A brief account of the material in this paper was given at the April, 1949 meeting of the American Physical Society.

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‡ Now at the University of Pennsylvania, Philadelphia, Pennsylvania.

<sup>1</sup> P. Kusch and H. M. Foley, *Phys. Rev.* **74**, 250 (1948).

<sup>2</sup> J. Schwinger, *Phys. Rev.* **73**, 416 (1948).