

## An Atomic Beam Study of the Hyperfine Structure of the Metastable $^2P_{3/2}$ State of $\text{In}^{115}\text{I}$

### The Electric Quadrupole Moment of $\text{In}^{115*}$

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The zero-moment method of atomic beams has been used to investigate the hyperfine structure of the metastable  $^2P_{3/2}$  state of  $\text{In}^{115}\text{I}$ . Thirteen zero-moment peaks have been observed, and it is found that their behavior is completely described by the equation for the h. f. s. energy levels,  $E = \frac{1}{2}aC + bC(C+1)$ , together with a knowledge of the nuclear  $g$  factor.  $a$  and  $b$  are the interval rule and quadrupole interaction constants, respectively; the numerical values are  $b/a = (6.432 \pm 0.04) \times 10^{-3}$ ;  $a = (8.10 \pm 0.10) \times 10^{-3} \text{ cm}^{-1}$ ;  $b = (0.0521 \pm 0.0009) \times 10^{-3} \text{ cm}^{-1}$ . From this the total h. f. s. separation is  $(0.1184 \pm 0.0015) \text{ cm}^{-1}$  and the quadrupole moment of  $\text{In}^{115}$  is  $0.84 \times 10^{-24} \text{ cm}^2$ . The nuclear spin is verified to be  $9/2$ . If there is an additional term  $c(C^3 + 4C^2 + 4/5C)$  in the h. f. s. energy level equation, as suggested by Tolansky in the case of iodine, an upper limit of  $5 \times 10^{-6}$  may be set on  $c/a$ .

THE electric quadrupole moments of a number of nuclei have been measured spectroscopically in recent years by observing deviations from the interval rule in hyperfine structure (h. f. s.). The interval rule arises from the cosine interaction between the nuclear magnetic moment and the magnetic field produced at the nucleus by the electrons. The nuclear quadrupole moment,  $Q$ , is defined as the average of  $(3z^2 - r^2)$  over the nuclear charge density for the state  $M_I = I$ , i.e.,  $eQ = \int \rho(3z^2 - r^2) d\tau$ , where  $\int \rho d\tau = Ze$ .  $Q$  is thus a measure of the deviation of the nucleus from spherical symmetry and gives rise to a cosine squared interaction with a properly asymmetrical electronic charge density. The position of the h. f. s. levels is given by

$$E = \frac{1}{2}aC + bC(C+1) \\ (C = F(F+1) - I(I+1) - J(J+1)), \quad (1)$$

where  $C$  is the quantum-mechanical analog of the cosine. The first term gives the interval rule and the second arises from the nuclear quadrupole.  $b$  is proportional to  $Q$  and to  $(3 \cos^2 \theta - 1)/r^3$  averaged over the electronic state  $M_J = J$ . Perturbations by other states may also give rise to deviations from the interval rule.

With the zero-moment method of atomic beams,<sup>1</sup> which is used in the present experiment,

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<sup>1</sup> V. W. Cohen, Phys. Rev. **46**, 713 (1934).

one observes in a magnetic field the behavior of the h. f. s. levels of the state in which the atoms of the beam exist. If one follows the course of these levels as the field is increased from zero through the region of transition between the h. f. s. anomalous Zeeman and Paschen-Back effects, it is found that for each of a number of levels there exists a value of the field at which  $dE/dH = 0$ . An atom for which this is true will experience no net translational force in an inhomogeneous magnetic field of this value, or in other words its effective magnetic moment is zero. If, now, an atomic beam traverses a transversely inhomogeneous deflecting field and the intensity of the beam at the position of zero deflection is observed as a function of magnetic field, "peaks" will be observed at the zero-moment fields of the various states. The fields at which such peaks occur are the observed quantities of the experiment.

Because of the limitations of the surface ionization beam detection method, set by the ionization potential of the atom being detected, zero-moment experiments have so far been limited to the alkali metals and indium. The ground states of all these elements have electronic charge distributions for which  $[\langle (3 \cos^2 \theta - 1)/r^3 \rangle_{Av}]_{M_J=J}$  is zero, hence a behavior not influenced by a nuclear quadrupole.

However, in the absolute moment experiments of Millman, Rabi and Zacharias carried out on

the  ${}^2P_{1/2}$  ground state of indium<sup>2</sup> (and not to be confused with the more recent magnetic resonance moment experiments), it was found possible to observe peaks of the metastable  ${}^2P_{3/2}$  state, which lies  $2212.6 \text{ cm}^{-1}$  above the ground level. The measurements with which this paper is concerned were made on these peaks with a new apparatus which has two advantages, for this purpose, over the one used by Millman, Rabi and Zacharias: first, increased resolving power, giving more and better peaks; second, use of the well-known two-wire type deflecting magnet<sup>3</sup> which has no iron and thus makes it possible to avoid magnetization curve difficulties. At the source temperature of about  $1500^\circ\text{K}$ , the expected abundance of the metastable state is about 20 percent according to Boltzmann statistics; since this level has 40 magnetic states, each peak should have about 0.5 percent of the intensity of the entire beam. This statement is verified by the authors cited and in the experiment to be described. Since  $[\langle(3 \cos^2 \theta - 1)/r^3\rangle_{av}]_{M_J=J}$  is not zero for the  ${}^2P_{3/2}$  state, the fields at which the peaks occur depend on  $Q$  and a means of measuring  $Q$  is thus provided.

This occurrence, for certain states, of a value of the magnetic field at which the effective moment of an atom is zero is a result of the progressive reorientation of  $\mathbf{J}$  by the external field, which uncouples  $\mathbf{I}$  and  $\mathbf{J}$ . In this reorientation process there is some value of the magnetic field at which the time average direction of  $\mathbf{J}$  is perpendicular to  $\mathbf{H}$ , and the effective moment of the atom is then zero. The strength of the field which is necessary to bring this about is directly proportional to the strength of the interaction between the electrons and the nuclear magnetic dipole moment if one neglects the influence of the quadrupole interaction. The coupling of the nuclear quadrupole to the asymmetrical electron charge distribution brings into play another torque on  $\mathbf{J}$  with a different dependence on the relative orientations of  $\mathbf{I}$  and  $\mathbf{J}$ . This helps or hinders the rearrangement process, as the case may be, and causes the zero moment to occur at a higher or lower value of the field than in the absence of the quadrupole. Thus, observation of

the relative values of a number of zero-moment fields provides a measure of the relative strengths of the magnetic dipole and electric quadrupole interactions.

To find the exact theoretical peak positions and their dependence on  $Q$ , one solves the secular equation for the energy of the atom in an external field  $H$  directed, say, along the  $z$  axis. Omitting constant terms, the Hamiltonian is given by<sup>4</sup>

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

(The terms are, in order, the interaction between the external field and the electrons and nucleus, and the electron-nuclear interactions due to the nuclear magnetic dipole and electric quadrupole.) Since  $I = 9/2$  and  $J = 3/2$ , the h. f. s. levels in zero field are characterized by  $F$  values of 3, 4, 5, 6; in a magnetic field each of these splits into  $2F+1$  states, a total of 40 in all, giving a 40-rowed secular determinant.

However, in either the  $F, M_F$  representation or the  $M_I, M_J$  representation  $H'$  is diagonal in  $M_F$ , the  $z$  component of the total angular momentum; since at most four states have the same  $M_F$  the secular determinant factors into a series of smaller determinants having at most four rows, the solution of which leads to quartic equations in the energy. The  $F, M_F$  representation is the more convenient since in it the last two terms in Eq. (2) are diagonal with matrix elements given by  $\frac{1}{2} a C + b C(C+1)$ , from which Eq. (1) arises. The matrix elements of  $I_z$  and  $J_z$  are well known and are given by Condon and Shortley<sup>5</sup> or, in a notation which is the same as ours if  $m = M_F$ , in the appendix to reference 2.

It is convenient for calculation to express energies in terms of  $a$ ; defining

$$x = g_J \mu_0 H / a, \quad (3)$$

we then have

$$H' = x \left( J_z + \frac{g_I}{g_J} I_z \right) + \mathbf{I} \cdot \mathbf{J} + \frac{b}{a} - 2 \frac{b}{a} (\mathbf{I} \cdot \mathbf{J}) [2(\mathbf{I} \cdot \mathbf{J}) + 1]. \quad (4)$$

<sup>4</sup> H. B. G. Casimir, "On the Interaction between Atomic Nuclei and Electrons," Prize Essay published by Teyler's Tweede Genootschap (1936).

<sup>5</sup> E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* (Cambridge, 1935).

<sup>2</sup> Millman, Rabi and Zacharias, *Phys. Rev.* **53**, 384 (1938).

<sup>3</sup> Rabi, Kellogg and Zacharias, *Phys. Rev.* **46**, 157 (1934).

After calculating the numerical values of the matrix elements involving the angular momenta, the secular equation contains as parameters  $x$ ,  $g_I/g_J$ , and  $b/a$ . Given any particular state, one desires the value of  $x$ , say  $x_0$ , at which  $\partial E/\partial x=0$  (provided  $x_0$  exists).  $x_0$  will depend on the values of  $g_I/g_J$  and  $b/a$ ; numerical calculation of this dependence is necessary—i.e., one assumes values for  $g_I/g_J$  and  $b/a$ , calculates  $\partial E/\partial x$  for several values of  $x$ , and interpolates to find  $x_0$ . This process must be repeated to obtain  $x_0$  for as many values of  $b/a$  and  $g_I/g_J$  as are necessary.

It is found that there is one value of the field at which the moment is zero for each of the following states, denoted by its  $(M_F, F)$  value:  $(-4, 4)$ ,  $(-4, 6)$ ,  $(0, 4)$ , and the twelve states for  $M_F = -1, -2, -3, F=3, 4, 5, 6$ . In addition there are two different zero-moment values of the field for the state  $(1, 4)$ .

The effect of  $g_I/g_J$  is very small, and, as will be discussed later,  $g_I/g_J$  is already known with an accuracy higher than can be obtained for it from the present experiment. In all the calculations we have therefore taken  $g_I = -0.000774$ , corresponding to  $\mu_I = 6.40$  nuclear magnetons,<sup>2</sup> and  $g_J = 4/3$  (see also later). For each peak, preliminary calculations of  $x_0$  were made for  $b/a = 0$  and  $0.006$  and  $dx_0/d(b/a)$  at  $b/a = 0$  was found. More exact locations of the zero moments were later calculated for  $b/a = 0.0056, 0.0060, 0.0064$ . The results are shown in Fig. 1. No exact calculations were made on the low zero-moment field of the state  $(1, 4)$  since it occurs at too low a field to be observed with the present apparatus.

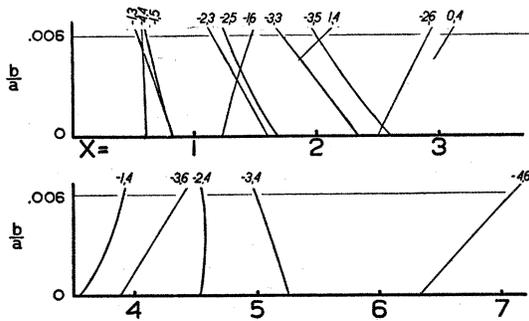


FIG. 1. Theoretical peak position as function of quadrupole interaction constant  $b/a$ .  $x \sim 7.6 \times 10^{-3} H$ ; see Eq. (2). Peaks denoted by  $(M_F, F)$  values.

#### APPARATUS AND PROCEDURE

A schematic top view of the apparatus is shown in Fig. 2. Some features are similar to those discussed in reference 2, but to obtain a design with maximum resolution an analysis of the half-widths of zero-moment peaks was carried out. Unlike previous calculations these included the variation of the moment of the atoms in the beam across the width of the beam, an important factor at high resolution. We summarize briefly the analysis and its results.

Let  $H_0$  be the value of the field at which the moment of the state under consideration is zero. To a close approximation, the absolute value of the deflecting field,  $H$ , varies linearly across the width of the beam and the magnetic moment at the field  $H$  varies linearly with  $(H-H_0)$ , i.e., with  $y$ , the distance from the zero-moment plane. (The locus of points for which  $H=H_0$  is the "zero-moment plane," while the "beam plane" is determined by the centers of the source and collimating slits and of the detector. The two planes are parallel, and coincide at a zero-moment peak.) If  $d^2E/dH^2 > 0$  for  $H=H_0$ , the zero-moment plane is a position of minimum energy and the atom is attracted to it by a force proportional to  $y$ . Such a state is called a "focusing" state. If  $d^2E/dH^2 < 0$  ("nonfocusing" state), the atom is repelled from the zero-moment plane by a force proportional to  $y$ . From this it is easily shown that the atomic trajectories in the deflecting field are given by

$$\begin{aligned} y &= y_0 \cos \beta t + (\dot{y}_0/\beta) \sin \beta t & (\text{focusing}) \\ y &= y_0 \cosh \beta t + (\dot{y}_0/\beta) \sinh \beta t. & (\text{nonfocusing}) \end{aligned}$$

$y_0$  and  $\dot{y}_0$  are the values of  $y$  and the transverse velocity  $\dot{y}$  at  $t=0$ ;

$$\beta^2 = \left| \frac{R^2 H_0^2}{m} \left( \frac{d^2 E}{dH^2} \right)_{H=H_0} \right|;$$

$m$  is the mass of the atom and  $R$  is the ratio of field gradient to field.

As the value of the field at the beam plane is increased from less than to more than  $H_0$ , the deflection of a beam of atoms of the state in question decreases until it is zero for  $H=H_0$  (and thus gives a peak in the intensity received at the detector) and then increases in the opposite direction. From the width of the deflected

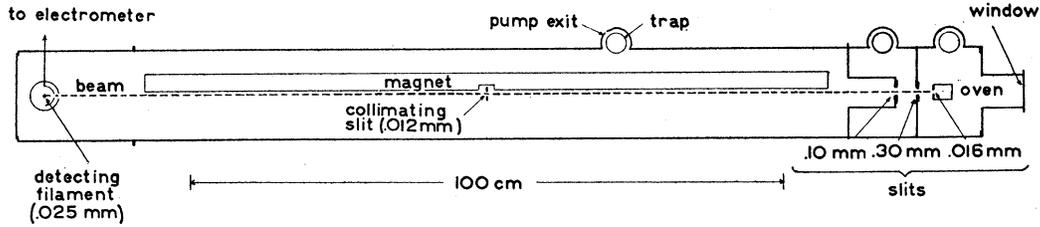


FIG. 2. Schematic top view of apparatus.

beam and the variation of its deflection with the value of  $H$  at the beam plane, both of which are easily calculated from the geometry of the apparatus and the above trajectories, the half-width of the peak (width at half the peak intensity), may be found for any given velocity of beam atoms.

The expression thus obtained is minimized with respect to the position of the collimating slit and the relative amounts of field-free space at each end of the magnet, with the result that the resolution is found to be best with a completely symmetrical arrangement such as that shown in Fig. 2. With this arrangement the half-width is given, for nonfocusing states, by the larger of the two following quantities:

$$\begin{aligned}
 (\Delta H/H)_c &= RW_c \left[ 1 - \frac{\lambda \gamma \cosh \gamma/2 + 2 \sinh \gamma/2}{\lambda \gamma \cosh \gamma + (1 + \lambda^2 \gamma^2/4) \sinh \gamma} \right]^{-1}; \\
 (\Delta H/H)_s &= \frac{RW_s}{2} \left[ 1 - \frac{\lambda \gamma \cosh \gamma + (1 + \lambda^2 \gamma^2/4) \sinh \gamma}{\lambda \gamma \cosh \gamma/2 + 2 \sinh \gamma/2} \right]^{-1}.
 \end{aligned} \quad (5)$$

For focusing peaks one substitutes  $\sin$  for  $\sinh$ ,  $\cos$  for  $\cosh$ , and changes the sign before the term  $\lambda^2 \gamma^2/4$ .  $W_s$  and  $W_c$  are the source and collimating slit widths, respectively;  $\lambda$  is the ratio of total field-free region to length of the deflecting field,  $l$ ;  $\gamma = \beta l/v$  where  $v$  is the atomic velocity. The physical significance of  $\gamma$  is obvious; for  $\gamma = \pi/2$ , an atom of a focusing state which is traveling in the beam plane will, on entering the deflecting region, just reach the zero-moment plane when it leaves the deflecting region.

These equations are useful in finding the best practical combination of the remaining adjustable variables  $R$ ,  $l$ ,  $\lambda$ ,  $W_s$  and  $W_c$ . The irregularities in the field itself set a limit to  $W_s$  and  $W_c$ .

of about 0.01 mm, below which any decrease in slit widths serves merely to decrease the intensity without helping the resolution. The final choice of  $R$ ,  $l$  and  $\lambda$  is determined by such factors as the obtainable length for an accurately machined magnet, the length of beam convenient as to intensity of the beam and stability of the apparatus, etc. Roughly speaking, for good results one should have  $\gamma \geq 1.2$  for focusing peaks and  $\gamma \geq 1.5$  for nonfocusing peaks, with  $\lambda \sim 0.3$ . The half-width increases very rapidly as  $\gamma$  falls below  $l$ , unless  $\lambda$  is increased rapidly, which is usually impracticable.

A cross section of the deflecting magnet at right angles to the beam plane is shown in Fig. 3. The magnet consists of two copper tubes 0.144" OD and 0.066" ID set one above the other in the face of a Duralumin block 3.5 × 5 × 115 cm. (For such a magnet the quantity  $R$  previously referred to is approximately  $1/a$  where  $a$  is the radius of the wires. All other quantities being constant, the resolution increases with  $R$ .) The tubes, insulated from one another and from the block by mica, carry the same current in opposite directions and are water cooled. After placing the wires in the block and before placing the block in the apparatus, the average deviation of the surface tangent to the wires from the mean tangent plane was about 0.007 mm. For the excitation of the field, currents up to 800 amperes (hereafter referred to as the "field current") were supplied by a six-volt bank of storage batteries of 3500 ampere-hours capacity. To measure the field current a Leeds & Northrup type K potentiometer and a 1500-ampere, 50-millivolt shunt were used. No direct measurements of the magnetic field were necessary; a much easier and more accurate calibration was made with an atomic beam of caesium, as will be described later. Because of this way of calibrating

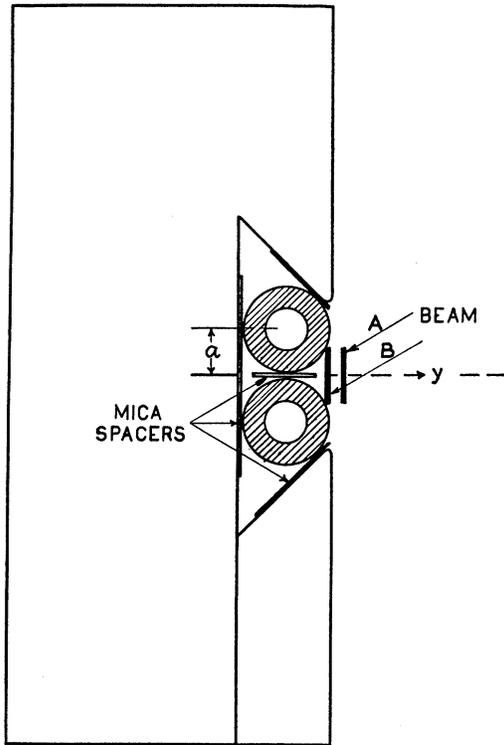


FIG. 3. Cross section of deflecting magnet. Field wires enlarged relative to Duralumin magnet block

the field, the calibration of the shunt does not affect the results.

The beam is defined by the oven and collimating slits, about 0.016 and 0.012 mm wide, respectively. The tungsten detecting filament is 0.025 mm in diameter. The height of the beam is limited to 2 mm by stops at each end. The oven and collimating slits are accurately vertical and may be moved under vacuum so that the beam may be placed parallel to the face of the magnet block and at any desired distance from it; the details of the adjustment are similar to those given in reference 2. One of the advantages of the symmetrical arrangement used here is that any error, within wide limits, in placing the beam plane parallel to the plane of the field wires has no effect whatsoever on the position or the half-widths of the observed peaks. (This does not apply, of course, to the verticality of the beam plane.) In the course of an experiment, the position of the beam at zero field was noted between each peak in order to avoid error caused by the walking of the beam; the position of the filament

and hence the relative position of the beam could be observed to better than 0.0005 mm. Under good conditions the average shift of the beam during the observation of one or two peaks was 0.001 mm or less, and while corrections for this were calculated, they were in most cases negligible.

Data were taken over a number of runs on three different line-ups, the first two with the beam in position A as shown in Fig. 3, about 0.6 mm outside the plane defined by the surface of the field wires and the third 0.1 mm inside, at position B.

#### EXPERIMENTAL DATA

Figure 4 shows the complete course of the beam intensity as a function of field current. The large background is due to  ${}^2P_{1/2}$  atoms, largely in the state  $M_F=0$ , which have smaller over-all moments than the  ${}^2P_{3/2}$  and therefore are not swept out of the field as readily. It is known from the work of Millman, Rabi and Zacharias on the

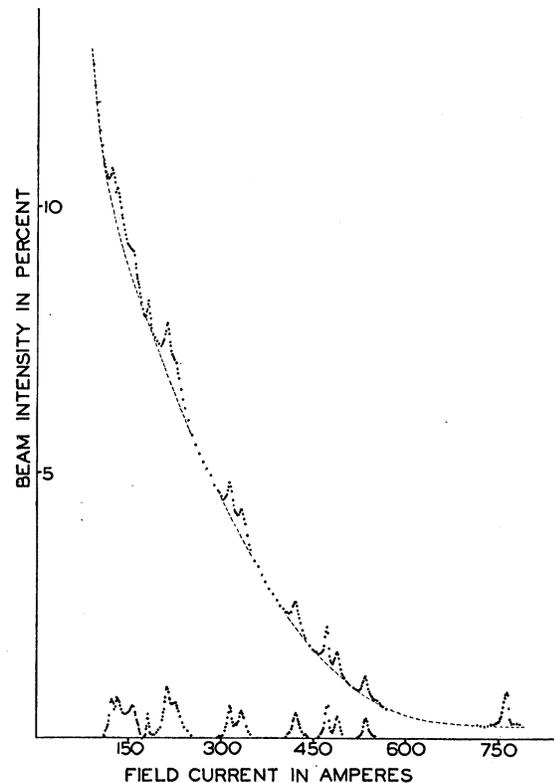


FIG. 4. Beam intensity at position of zero deflection as function of field current. Background of  ${}^2P_{1/2}$  atoms is included.

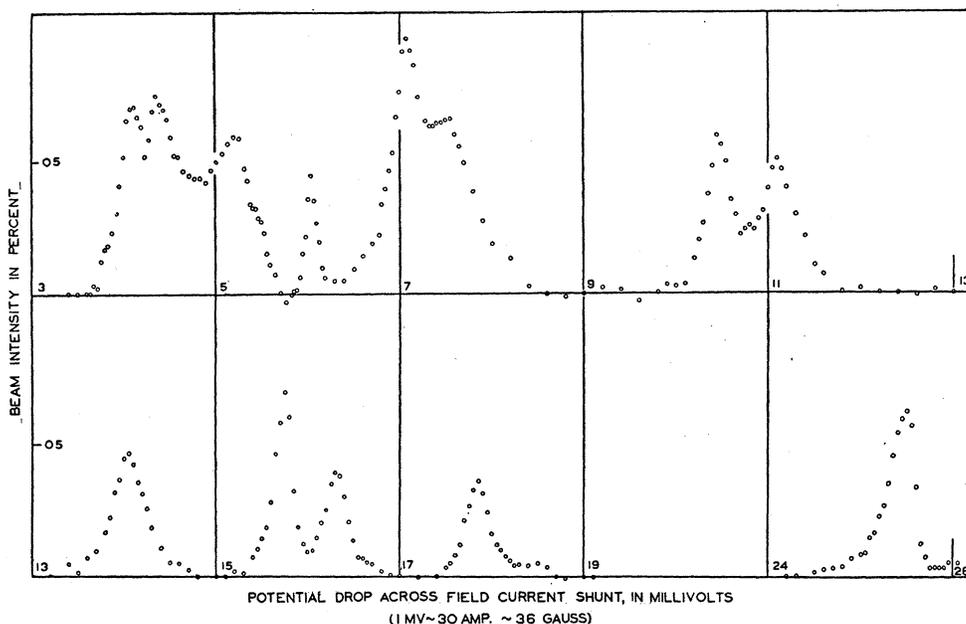


FIG. 5. Beam intensity, at position of zero deflection, of  $^2P_{3/2}$  atoms alone.

ground state that there are no  $^2P_{3/2}$  peaks at higher fields than those shown. In locating the  $^2P_{3/2}$  peaks the  $^2P_{1/2}$  background is subtracted by running a smooth curve through the points adjacent to the peaks. The peaks thus obtained are shown in Fig. 4 and in more detail in Fig. 5.

The unequal intensity of the peaks is not caused by unequal population of the corresponding states. At a peak arising from a focusing state the atoms travel down the middle of a long potential valley to the detector and are focused in to the detector to give an intense peak. For a nonfocusing peak the atoms are trying to travel a straight and narrow path along the top of a potential ridge and much fewer reach the detector. The area under a peak is not a measure of population either, as the half-widths are not necessarily greater for the nonfocusing than for the focusing peaks. This may be seen with reference to Eqs. (5).

An asymmetry may also be observed for several peaks, notably the last one. This occurs because these curves were taken on line-up *B* as shown in Fig. 3 so that the wires constitute, in effect, an asymmetrical height stop; because of this, focusing peaks have their high field intensity, the nonfocusing peaks their low field intensity reduced. This asymmetry has been

taken into account in locating the peaks. It does not appear in the curves taken with the line-ups for which the beam is clear of the wires.

The directly observed quantity which gives the position of a peak is the potential drop, in millivolts, across the field current shunt, hence there were three observed positions of a given peak corresponding to the three different selections of position of the beam with respect to the field wires. In order to compare the peaks of the first two line-ups with those of the third line-up, for which the resolution was best, the ratios of the positions (in millivolts) of each peak on line-up 1 to the positions of the same peaks on line-up 3 were calculated and a weighted average over all peaks gave a conversion factor changing millivolts on line-up 1 to millivolts on line-up 3. A similar procedure was followed for line-up 2. The resulting comparison showed no discrepancies, beyond the limits of error which had previously been assigned for the location of the peak positions in millivolts, between the results obtained on the three line-ups, except in one case. Here there had been a systematic error in locating two peaks which were poorly resolved on both the first two line-ups.

From the three positions of each peak and the corresponding limits of error a final position and

limit of error was set. The numbers which enter into the final results of the experiment were based on an average of 16 observations for each peak, distributed over 10 runs. The results are shown in Table I.

### RESULTS

The relative positions at which the peaks occur depend only on the ratio  $b/a$  and the nuclear and electronic total angular momenta,  $I$  and  $J$ . The absolute value of the magnetic field at each peak is proportional to  $a/g_J$ , i.e., to the relative strength of the electron-nuclear and electron-external field magnetic interactions. From the numbers upon which Fig. 1 is based, the dependence on  $b/a$  of a number of ratios of peak positions was calculated and from the experimentally observed values of these ratios the corresponding values of  $b/a$  were determined. Of the resulting values of  $b/a$ , those having limits of error of less than three percent are shown in Fig. 5. The weighted average of these gives  $b/a = (6.432 \pm 0.04) \times 10^{-3}$ . Here, as in all further results, the plus or minus represents a reasonable limit of error. It will be noted that the value of  $b/a$  is entirely independent of any measurement of absolute value of magnetic field.

#### The hyperfine structure separation

Knowing  $b/a$ , we may now determine from the calculations on which Fig. 1 is based the value of  $x$  at which any given peak occurs. A measure of the field at this peak then gives  $a/g_J$  directly, by use of Eq. (3). The most accurate way of calibrating the field is to run another element whose peaks occur at known fields. In this case caesium was used. The  $\Delta\nu$  of the  $^2S_{1/2}$  ground state of caesium is known to be  $0.3067 \pm 0.0004$

$\text{cm}^{-1}$  spectroscopically;<sup>6</sup> Millman and Fox,<sup>7</sup> using atomic beam methods, found  $0.307 \pm 0.003 \text{ cm}^{-1}$  for  $\Delta\nu$  and  $7/2$  for the spin. Given the  $\Delta\nu$ , for which we take the spectroscopic value, and the spin, the field at any peak of caesium is then known. It was not found feasible to obtain caesium and indium beams from the same source oven, so the caesium run was bracketed between two indium runs. The difference in the position of the  $(-3, 4)$  indium peak (the most accurately reproducible of the indium peaks) on the two runs was at most 0.1 percent and the maximum spread of locations of this peak on three indium runs preceding the caesium run over a period of three weeks was 0.06 percent. Experimentally, the ratio of the  $(-3, 4)$  peak to the first caesium peak was found to be  $0.7913 \pm 0.0022$ . The  $(-3, 4)$  peak comes at  $x = 4.9839 \pm 0.0025$  for the value of  $b/a$  found above. Making use of these two numbers and the  $\Delta\nu$  and spin of caesium we obtain, from Eq. (3) and the analogous equation for caesium,  $a/g_J = (6.087 \pm 0.028) \times 10^{-3} \text{ cm}^{-1}$ . If one assumes for  $g_J$  the value for a pure  $^2P_{3/2}$  state,  $4/3$ , this gives  $a = (8.116 \pm 0.037) \times 10^{-3} \text{ cm}^{-1}$ . By way of comparison, Schüler and Schmidt,<sup>8</sup> who also investigated the h. f. s. of this state, give  $a = 7.97 \times 10^{-3} \text{ cm}^{-1}$ , obtained from the two largest separations of the h. f. s. pattern. Their separations are stated to  $0.001 \text{ cm}^{-1}$ , and an error of this amount in one of the measured separations corresponds to an error of  $0.15 \times 10^{-3} \text{ cm}^{-1}$  in  $a$ . The two values are not in contradiction in view of their respective limits, but because of the possible influence of perturbations on  $g_J$  (see later), we increase the limit of error and take  $a = (8.10 \pm 0.10) \times 10^{-3} \text{ cm}^{-1}$ . If there were no quadrupole moment the h. f. s. separation would then be given by  $\Delta\nu = 15a = (0.1215 \pm 0.0014) \text{ cm}^{-1}$ . Because of the presence of the quadrupole, which shifts the two extreme levels by different amounts, the actual separation of these levels is  $(0.1184 \pm 0.0015) \text{ cm}^{-1}$ , making use of the value of  $b = (0.0521 \pm 0.0009) \times 10^{-3} \text{ cm}^{-1}$  deduced from the above values of  $b/a$  and  $a$ . Schüler and Schmidt found  $b = 0.048 \times 10^{-3} \text{ cm}^{-1}$ , a value which is altered by ten percent by a

TABLE I. Final determinations of peak positions.

PEAK ( $M_F, F$ )	PEAK POSITION MV ON LINE-UP 3	PEAK ( $M_F, F$ )	PEAK POSITION MV ON LINE-UP 3
-2,3	$4.068 \pm 0.035$	0,4	$11.122 \pm 0.040$
-2,5	$4.410 \pm 0.04$		
-1,6	$5.215 \pm 0.06$	-1,4	$14.020 \pm 0.025$
-3,3	$6.040 \pm 0.015$	-3,6	$15.760 \pm 0.020$
		-2,4	$16.300 \pm 0.025$
-3,5	$7.080 \pm 0.020$	-3,4	$17.836 \pm 0.023$
1,4	$7.500 \pm 0.035$	-4,6	$25.44 \pm 0.10$
-2,6	$10.463 \pm 0.026$		

<sup>6</sup> L. P. Granath and R. K. Stranathan, Phys. Rev. **48**, 725 (1935).

<sup>7</sup> S. Millman and M. Fox, Phys. Rev. **50**, 220 (1936).

<sup>8</sup> H. Schüler and T. Schmidt, Zeits. f. Physik **104**, 468 (1937).

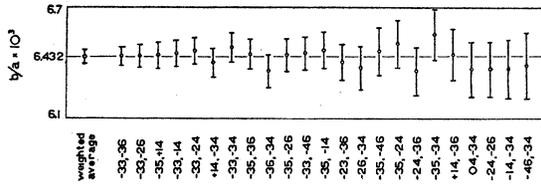


FIG. 6. Values of  $b/a$  calculated from observed peak ratios. The two peaks contributing to a given value of  $b/a$  are specified by their  $(M_F, F)$  values.

change of  $0.001 \text{ cm}^{-1}$  in one of their measured separations.

### Nuclear spin and nuclear $g$ factor

The fact that an unambiguous identification of observed and theoretical peaks may be made serves to determine the correctness of the previously measured value  $9/2$  for the nuclear spin  $I$ , upon which the calculations were based.

Strictly speaking, the ratio of the nuclear and electronic  $g$  factors, which occurs in Eq. (4), affects slightly the relative positions of the peaks. It was because of this fact that Millman, Rabi and Zacharias<sup>2</sup> were able to find the nuclear  $g$  factor of indium directly rather than through calculations based upon h. f. s. separations. To a very close approximation, the shift of a peak by the term in  $g_I$  may be obtained from the following considerations. If  $g_I$  (and hence the nuclear moment) is zero then the zero-moment value of the field is that for which  $\mathbf{J}$  is, on the average, perpendicular to  $\mathbf{H}$ —i.e., for which  $\bar{M}_J = 0$ . Since  $M_F$  is a constant of the motion and  $M_F = M_I + M_J$ , it follows that  $\bar{M}_I = M_F$ . If now  $g_I$  is not zero, the moment of the atom at this particular field is  $-g_I \bar{M}_I \mu_0 = -g_I M_F \mu_0$ , which is of the order of  $0.001$ – $0.002$  Bohr magneton for the case in question. If one knows  $d\mu/dH$ , which is easily calculated, then  $\Delta H_0$ , the shift in the zero-moment value of the field due to  $g_I$ , is given by  $\Delta H_0 = g_I M_F / (d\mu/dH)$ . The electronic moment is 2 Bohr magnetons for the  ${}^2P_{3/2}$  state as compared with  $\frac{1}{3}$  of a Bohr magneton for the  ${}^2P_{1/2}$ , and the effect of this factor in increasing  $d\mu/dH$  is to make the percent shift of the peaks correspondingly smaller for the  ${}^2P_{3/2}$  state. Since the  $g_I$  of Millman, Rabi and Zacharias is therefore more accurate than one which could be obtained from the present experiment, we have used their value in calculating the theoretical

peak positions shown in Fig. 1. If we set  $g_I = 0$  in the theoretical calculations, the resulting changes in the values of  $b/a$  as deduced from the experimental data are in no case changed by more than the limits of error shown in Fig. 6. The consistency of the values of  $b/a$ , however, is made noticeably worse, indicating that the assumed sign and order of magnitude of  $g_I$  are correct.

### The quadrupole moment

For this particular state, the relation between  $Q$  and  $b$  is<sup>4, 8</sup>

$$Q = b \frac{I(2I-1)Z_i H C}{\delta R'} \frac{60\mu_0^2}{e^2},$$

Here  $\delta$  is the doublet separation,  $2212.6 \text{ cm}^{-1}$ , which enters since  $\bar{r}^{-3}$  is proportional to it.  $H$ ,  $R'$ , and  $C$  are relativistic correction factors which enter because of the use of  $\delta$  to give  $\bar{r}^{-3}$ . Calculations of these quantities have been made by Casimir,<sup>4</sup> from whose tables we obtain  $H = 1.050$  and  $R' = 1.088$ .  $C$  is a correction for the fact that in calculating  $H$  and  $R'$  the difference in the normalization integrals of the  ${}^2P_{1/2}$  and  ${}^2P_{3/2}$  states, caused by the difference in the energy of these two states, was neglected. Casimir gives an analytical expression for this effect from which we obtain  $C = 1.025$ .  $Z_i$  is the "effective" nuclear charge. On the basis of the relativistic wave functions from which  $H$ ,  $R'$  and  $C$  are calculated, Casimir obtains an expression for  $\delta$  which depends on  $Z_i$ . From the doublet separations and term values,  $E_n$ , of the series  $5s^2np$ , a value of  $Z_i$  may be obtained for each  $n$ . Averaging the values obtained from  $n = 6$  to  $10$  inclusive we find  $Z_i = 44.8 = Z - 4.2$ , with an average deviation of  $1.0$ . (The value for  $n = 5$  was omitted since its calculation involved a rather crude estimation of  $dE_n/dn$ , which is the form in which  $E_n$  enters the formula for  $\delta$ .) Finally, in the calculation of  $\mu_0/e$ , in which  $\mu_0$  and  $e$  are the Bohr magneton and the electronic charge, we have taken  $h/e = 1.3759 \times 10^{-7}$  e. s. u. and  $e/m = 1.7591 \times 10^7$  e. m. u.  $b$  and  $\delta$  are expressed in  $\text{cm}^{-1}$ .

Substitution of these numerical values gives  $Q = 0.837 \times 10^{-24} \text{ cm}^2$ ; we take

$$Q = 0.84 \times 10^{-24} \text{ cm}^2.$$

Some attention must be paid to perturbations of the  $5s^25p \text{ } {}^2P_{3/2}$  state in question by electrostatic

coupling with states of the same total angular momentum in higher configurations, notably the three states with  $J = \frac{3}{2}$  in the  $5s5p6s$  configuration. As was shown by Fermi and Segré<sup>9</sup> in the closely analogous case of thallium, such coupling is of the right order of magnitude to explain the anomalously small  $\Delta\nu$  of this state, which is about half what it should be on the basis of the known value for the magnetic moment. While the levels of the  $5s5p6s$  configuration have not been observed spectroscopically, the calculations of Fermi and Segré indicate that it lies roughly  $50,000 \text{ cm}^{-1}$  above the ground level, with a matrix element of the electrostatic energy of the order of  $5000 \text{ cm}^{-1}$  coupling the two configurations. This means that properties of the states in the  $5s5p6s$  configuration mix with those of the lower level in the ratio of about 1 : 100. Since the upper configuration has two unbalanced  $s$  electrons the  $\Delta\nu$ 's of the various multiplet levels will be very large. If we represent the total effect by a single  $\Delta\nu$  with a reasonable value of  $12 \text{ cm}^{-1}$  and an inverted pattern, the  $\Delta\nu$  of the lower state is cut in half as is observed. The situation is quite different as regards the quadrupole moment, however;  $s$  electrons, having spherically symmetrical wave functions, do not exhibit the quadrupole effect in h. f. s. The quadrupole contributions to the energy levels of both configurations arise from the  $5p$  electron and hence will be of the same order of magnitude so that perturbations in the value of  $Q$  observed for the lower state will have an order of magnitude of at most one percent. As concerns  $g_J$ , previously referred to, the  $g$  sum rule shows that the average  $g_J$  for the states  $J = \frac{3}{2}$  in the upper level is  $22/15 = 1.47$  as compared with  $4/3$  for the state in question, so that  $g_J$  might possibly be increased by 0.3 percent.

#### DISCUSSION

Schüler and Schmidt, whose work on the same state has already been mentioned, obtained for  $0.8 \pm 0.2$ . The large probable error arises from the fact, previously noted, that a  $0.001\text{-cm}^{-1}$  error in one of their h. f. s. level separations causes a ten-percent error in  $b$ , and hence in  $Q$ . Bacher and Tomboulia,<sup>10</sup> working with the

$5s6p \ ^1P_1$  state of In II, found  $Q = 0.82$ . The agreement with our value of 0.84 is very pleasing; but in view of the fact that the method of calculating  $Q$  from h. f. s. is very similar to that used for nuclear moments, which usually show a wide spread, the agreement may be in part fortuitous. Much more significant is the internal consistency of our results, as indicated by the 0.6 percent limit of error on  $b/a$ . Because there are thirteen observables in this experiment as compared with the much smaller number of h. f. s. energy differences observable in a spectroscopic investigation of a single state, this consistency provides a much more complete check on the sufficiency of the assumed interaction between nucleus and electrons.

In this connection, Tolansky<sup>11</sup> has recently reported that he finds it necessary, for several states of iodine, to add to Eq. (1) a term  $c(C^3 + 4C^2 + 4/5C)$  representing a cosine<sup>3</sup> interaction between nucleus and electrons. The constant  $c$  is presumably proportional to a nuclear magnetic octopole moment, and  $c/a$  is of the order of magnitude of  $2 \times 10^{-4}$ . We have calculated the effect which such a term would have on our peaks; we find that if  $c/a = 5 \times 10^{-6}$ , and if the term in  $c/a$  is neglected in calculating the theoretical dependence of peak position on  $b/a$ , then some of the more accurately determined experimental values of  $b/a \times 10^3$  shown in Fig. 6 should be inconsistent by  $1\frac{1}{2}$  percent, which is not so for the values in question. Furthermore, such inconsistencies as appear in Fig. 6 are not bettered by any choice of  $c/a$ . Hence  $5 \times 10^{-6}$  seems a good upper limit to  $c/a$  in this state.

In conclusion, the author desires to acknowledge his indebtedness to Professor I. I. Rabi, who suggested this application of molecular beam methods to the measurement of quadrupole moments and whose advice was very helpful. He also wishes to express his deepest gratitude to Mr. Nicholas A. Renzetti for his continuously invaluable assistance during the course of the research. The other members of the molecular beam laboratory have been of great aid in contributing the benefits of their previous experience.

<sup>9</sup> E. Fermi and E. Segré, *Zeits. f. Physik* **82**, 729 (1933).

<sup>10</sup> R. F. Bacher and D. H. Tomboulia, *Phys. Rev.* **52**, 836 (1937).

<sup>11</sup> S. Tolansky, *Proc. Roy. Soc.* **A170**, 205 (1939).