Hfs of the 5 $^2P_{\frac{1}{2}}$ State of In^{115} and In^{113}: Hfs Anomalies in the Stable Isotopes of Indium*†

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The hfs splittings of the 5 $^2P_{\frac{1}{2}}$ states of In^{115} and In^{113} have been measured by the atomic-beam magnetic-resonance method. We find:

$$\Delta\nu(In^{115}) = [11\,409.7506 \pm (20)] \times 10^6 \text{ sec}^{-1},$$
$$\Delta\nu(In^{113}) = [11\,385.4300 \pm (20)] \times 10^6 \text{ sec}^{-1}.$$

A comparison of the ratio of the dipole coupling constants of the two isotopes in the $P_{\frac{1}{2}}$ state and in the $P_{\frac{3}{2}}$ state with the ratio of the nuclear g_I factors yields the following hfs anomalies:

$$\Delta_{\frac{1}{2}} = (7.5 \pm 1.3) \times 10^{-6}, \quad \Delta_{\frac{3}{2}} = -(23.8 \pm 1.3) \times 10^{-6}.$$

The fractional change in the nuclear radius between In^{113} and In^{115}, $\delta R/R$, is found to be 3.66×10^{-3} as compared to 5.85×10^{-3} calculated on the assumption of an incompressible nucleus.

INTRODUCTION

INDIUM has two stable isotopes, In^{115} and In^{113}, each of which has a nuclear spin of 9/2. Within the precision of previous determinations of the ratio of their nuclear magnetic moments[1] (4 parts in 10^5) and of the ratio of their magnetic dipole interaction constants in the $P_{\frac{1}{2}}$ state[2] (1 part in 10^4), these two isotopes have shown no hfs anomaly. Rice and Pound[3] have recently measured the magnetic moment ratio to a precision of one part in 10^6 by use of the method of nuclear magnetic resonance. We have measured the zero-field hfs splitting in the $P_{\frac{1}{2}}$ states of In^{115} and In^{113} to about one part in 5×10^6. These measurements yield the hfs anomaly in the 5 $^2P_{\frac{1}{2}}$ state of indium, approximately 7 parts in 10^6.

The present work was undertaken as part of a detailed investigation of the hfs of the $P_{\frac{1}{2},\frac{3}{2}}$ ground state doublet of In^{115} and In^{113}. In the course of the investigation the magnetic dipole coupling constants in the $P_{\frac{3}{2}}$ state have been determined.[4] We shall compare the experimental and theoretical values of the anomaly for these two states and from this comparison obtain an estimate of the variation in nuclear radius in going from In^{113} to In^{115}.

THEORY

In the presence of an external magnetic field the energies of the magnetic hfs levels of the $P_{\frac{1}{2}}$ state of indium are given to a good approximation by the Breit-Rabi formula.[5] Deviations from this formula appear principally as small additional terms in $\Delta\nu$ which do not occur through the coefficient $a_{\frac{1}{2}}$ in the $\mathbf{I}\cdot\mathbf{J}$ interaction and as a change in the apparent value of g_I, the nuclear gyromagnetic ratio. These deviations, which arise from perturbations of the $P_{\frac{1}{2}}$ state by the $P_{\frac{3}{2}}$ state, have been considered by Clendenin[6] for a nucleus possessing only a magnetic dipole moment. The modification of the theory to include the effects of configuration interaction and its extension to the case where the nucleus possesses an electric quadrupole moment are given in reference 4.

The transitions observed in this experiment were those for which $\Delta F = \pm 1$, $\Delta m = \pm 1$, where $F = I \pm \frac{1}{2}$. For small fields these transitions give rise in the case of each isotope of indium to 18 lines. There are 8 doublets ($I+\frac{1}{2}, m \leftrightarrow I-\frac{1}{2}, m-1$; $I+\frac{1}{2}, m-1 \leftrightarrow I-\frac{1}{2}, m$) each with a frequency separation $2g_I\mu_0 H/h$ and with a mean frequency given with sufficient precision for our purposes by

$$f\begin{pmatrix} F, m \leftrightarrow F-1, m-1 \\ F, m-1 \leftrightarrow F-1, m \end{pmatrix} = \Delta\nu\left\{1 + \left(\frac{2m-1}{2I+1}\right)x + \left[\frac{1}{2} - \frac{(2m^2-2m+1)}{(2I+1)^2}\right]x^2\right\}, \quad (1)$$

where

$$x = (g_J - g_I)\mu_0 H/h\Delta\nu.$$

These doublets are entirely unresolved at the magnetic fields at which the observations of the present experiment were made. The frequencies of the two single lines 5, 5↔4, 4 and 5, −5↔4, −4 are given by the same expression with the additional term $\pm g_I\mu_0 H/h$ for the two lines respectively.

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APPARATUS AND PROCEDURE

The measurements of $\Delta\nu$ for In^{115} and In^{113} were made on an atomic-beam magnetic-resonance apparatus of conventional design, the general features of which have previously been described.[7] The small relative abundance of In^{113} (4.2%) led to the choice of an arrangement of the deflecting fields in which both the gradient and the field in the A and B magnets were in the same direction. An atom is then refocused at the detector only when it has made a transition in the C field which results in a reversal in the sign of the magnetic moment at high fields. An A field of about 15 000 gauss and a B field of about 10 000 gauss left only a small background of intensity at the detector in the absence of transitions and gave satisfactory refocusing of atoms which had made the transition 5, 5↔4, 4 or any of the transitions which lead to the doublets. The line 5, −5↔4, −4 was observed for In^{115} as a small negative intensity peak corresponding to a reduction in the background beam. We attribute this to the fact that the 4, −4 state has a very small effective moment for the B field which was used.

The high-frequency power was generated by a Varian X-13 klystron, stabilized in a manner previously discussed.[8] The klystron frequency was measured by conventional methods against our secondary frequency standard which was in turn checked against WWV. That part of the high-frequency rf circuit in which the transitions occurred (the hairpin) was constructed from X-band wave guide tapered along its short dimension to permit insertion into the 0.635 cm gap of the C magnet. The oscillating magnetic field was then perpendicular to the C field and only π transitions were induced. The C field never exceeded 5 gauss. It was large enough so that nonadiabatic transitions among the magnetic levels did not occur in the region between the two deflecting fields. It also gave excellent resolution between adjacent lines since the interval between such lines increases at the rate of 187 kc sec^{-1} $gauss^{-1}$. Finally, it was low enough so that terms in the expression for the frequency, quadratic in the field, were very small. The natural line width of approximately 20 kc/sec did not allow the resolution of the doublets, since at the highest field, 5 gauss, the doublet separation is only 9 kc/sec.

A set of data consisted of a number of measurements of the klystron frequency corresponding to several of the peaks. All the measurements of a set were taken at a nominally identical C field and those sets for which there was evidence of appreciable C field drift were discarded. A value of $\Delta\nu$ was found by a least-squares adjustment of the measured frequencies in the set to the expressions for the frequencies [Eq. (1)].

[7] J. R. Zacharias, Phys. Rev. **61**, 270 (1942).
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TABLE I. The results of measurement in separate runs of the zero field hfs splitting of indium in the $^2P_{\frac{1}{2}}$ state.

Isotope	Number of peaks measured	$10^{-6}\times\Delta\nu$ (sec^{-1})
115	9	11 409.7520±(13)
115	9	11 409.7496±(10)
115	10	11 409.7502±(27)
113	9	11 385.4294±(12)
113	9	11 385.4307±(15)

RESULTS

Three sets of data for In^{115} and two for In^{113} were taken under good observational conditions and free of obvious systematic error such as that introduced by a drift of the C field. The values of $\Delta\nu$ obtained from these sets are tabulated in Table I. Here, as elsewhere in this paper, the number in parentheses is the uncertainty in the last-quoted figure of the result. The uncertainty assigned to each value of $\Delta\nu$ is the probable error given by the least-squares calculation. We give

$$\Delta\nu(In^{115}) = [11\,409.7506\pm(20)]\times10^6\ sec^{-1},$$
$$\Delta\nu(In^{113}) = [11\,385.4300\pm(20)]\times10^6\ sec^{-1}.$$

To the stated precision, possible systematic errors in the comparison of our frequency standard with WWV and possible errors in WWV itself are negligible.

The value of $\Delta\nu(In^{113})$ agrees within experimental error with its best previous determination[2] of $[11\,387\pm(3)]\times10^6\ sec^{-1}$. In the case of In^{115}, however, there is a discrepancy between the present value and a value $[11\,409.50\pm(10)]\times10^6\ sec^{-1}$ previously obtained[9] from measurements at 17 000 gauss that is two and one-half times the stated uncertainty. This discrepancy is discussed in the following paper and is found to arise from field-dependent deviations from the Breit-Rabi formula that are negligible at very weak fields but not for a field of 17 000 gauss. When these deviations are taken into account, the zero-field and intermediate-field determinations of $\Delta\nu(In^{115})$ are in good agreement.

HFS ANOMALY: EXPERIMENTAL

The hfs anomaly constant, Δ, is defined by the expression

$$\Delta \equiv \left[\frac{a'(1)}{a'(2)}\right]\left[\frac{g_I(2)}{g_I(1)}\right]-1, \qquad (2)$$

where 2 refers to the heavier of the two isotopes, a' is the dipole coupling constant for the electronic state considered, and g_I is the nuclear gyromagnetic ratio. The primed notation is that adopted in the following paper and indicates that the corrections due to $P_{\frac{1}{2},\frac{3}{2}}$ mutual perturbations and to configuration interaction have been taken into account. The expression relating the dipole interaction constant in the $P_{\frac{1}{2}}$ state of indium to the measured value of $\Delta\nu$ is given in the following

[9] H. Taub and P. Kusch, Phys. Rev. **75**, 1481 (1949).

paper. From this expression we obtain

$$a_{\frac{1}{2}}'(\mathrm{In}^{115})=[2281.95536\pm(40)]\times10^{6}\ \mathrm{sec}^{-1},$$
$$a_{\frac{1}{2}}'(\mathrm{In}^{113})=[2277.09117\pm(40)]\times10^{6}\ \mathrm{sec}^{-1},$$
$$a_{\frac{1}{2}}'(\mathrm{In}^{113})/a_{\frac{1}{2}}'(\mathrm{In}^{115})=0.99786841\pm(28).$$

For the $P_{\frac{3}{2}}$ state we have[4]

$$a_{\frac{3}{2}}'(\mathrm{In}^{113})/a_{\frac{3}{2}}'(\mathrm{In}^{115})=0.99783716\pm(28).$$

When combined with g_I ratio,[3]

$$g_I(\mathrm{In}^{115})/g_I(\mathrm{In}^{113})=1.0021437\pm(12),$$

the dipole interaction constant ratios give

$$\Delta_{\frac{1}{2}}=(7.5\pm1.3)\times10^{-6},\quad \Delta_{\frac{3}{2}}=-(23.8\pm1.3)\times10^{-6}.$$

The uncertainties in $\Delta_{\frac{1}{2}}$ and $\Delta_{\frac{3}{2}}$ come principally from the uncertainty in the g_I ratio. The quantity

$$\Delta_{\frac{1}{2}}-\Delta_{\frac{3}{2}}=[a_{\frac{1}{2}}'(\mathrm{In}^{113})/a_{\frac{1}{2}}'(\mathrm{In}^{115})-a_{\frac{3}{2}}'(\mathrm{In}^{113})/a_{\frac{3}{2}}'(\mathrm{In}^{115})]\times[g_I(\mathrm{In}^{115})/g_I(\mathrm{In}^{113})]=(31.32\pm0.40)\times10^{-6},$$

which is independent of the exact value of the g_I ratio, is the most useful quantity for comparison with the detailed theory of the hfs anomaly in indium.

HFS ANOMALY: THEORETICAL

For a point charge, point magnetic dipole moment nucleus, Δ would have a null value. Nonzero values of Δ arise from the finite distribution of both the charge and the magnetization within the nuclear volume. The former effect has been treated by Breit and Rosenthal[10] and the latter by Bohr and Weisskopf[11] and by Bohr.[12] A calculation of the distributed moment anomaly based on the single particle model of the nucleus[11] gives a value that is about 9% of the observed anomaly for indium. Since the uncertainty in this calculation is large and the apparent magnitude of the distributed moment anomaly is less than the uncertainty in the theoretical value of Δ_{BR}, the anomaly due to the distributed charge, we shall neglect the distributed moment anomaly in the following discussion.

Using the expression given in reference 10 for the magnetic dipole interaction for a finite charge distribution in terms of that for a point charge, we obtain the following theoretical expression for Δ_{BR}:

$$\Delta_{BR}=\frac{2(j-\rho)\rho(2\rho+1)}{(2j-1)[\Gamma(2\rho+1)]^2}\left(\frac{2Zp}{a_{\mathrm{H}}}\right)^{2\rho-1}\times[R(2)^{2\rho-1}-R(1)^{2\rho-1}],$$

where a_{H} is the radius of the first Bohr orbit in hydrogen, Z is the atomic number, $\rho=(1-Z^2\alpha^2)^{\frac{1}{2}}$, α is the fine structure constant, $R(2)$ is the nuclear radius of the heavier isotope, j is the quantum number taking the values $-1, 1, -2, 2, \cdots$ for $s_{\frac{1}{2}}, p_{\frac{1}{2}}, p_{\frac{3}{2}}, d_{\frac{3}{2}}, \cdots$ electrons, respectively. The parameter p is defined by the requirement that the dipole interaction integral from $r=0$ for $r=\infty$ for a distributed nuclear charge be equal to the integral from $r=pR$ to $r=\infty$ for a point-charge nucleus. Crawford and Schawlow[13] find $p\cong8/9$ for a uniform distribution of nuclear charge and $p\cong10/9$ for all the charge on the surface of the nucleus. Setting $R(2)=R(1)+\delta R$ and expanding to terms linear in $\delta R/R$, we have

$$\Delta_{BR}=\frac{2(j-\rho)\rho(2\rho+1)(2\rho-1)}{(2j-1)[\Gamma(2\rho+1)]^2}\left(\frac{2ZpR}{a_{\mathrm{H}}}\right)^{2\rho-1}\frac{\delta R}{R}. \quad (3)$$

Since $\delta R/R$ is of the order of 10^{-3}, higher order terms in the expansion are negligible.

Only $s_{\frac{1}{2}}$ and $p_{\frac{1}{2}}$ electrons will contribute to the anomaly, since only for these states does the electronic wave function have an appreciable density within the nuclear volume. Assuming a uniform charge distribution ($p=8/9$) and taking $R=1.2\times10^{-13}A^{\frac{1}{3}}$ cm, we find for indium

$$\Delta_{BR}(p_{\frac{1}{2}})=1.41\times10^{-3}\delta R/R,\quad \Delta_{BR}(s)=16.81\times10^{-3}\delta R/R.$$

The value of $\Delta_{BR}(p_{\frac{1}{2}})$ has been reduced by a factor of 1.22 from the value given by Eq. (3) to take account of the shielding of the $p_{\frac{1}{2}}$ electron by the core electrons.[14]

Since a $p_{\frac{3}{2}}$ electron has a negligible density at the nucleus, the anomaly in the $P_{\frac{3}{2}}$ state of indium must be due to the lack of purity of this electronic state. Schwartz[15] has discussed the anomalies in the $4\,^2P_{\frac{1}{2},\frac{3}{2}}$ states of gallium assuming that all of the $P_{\frac{3}{2}}$ anomaly is due to the admixture of electronic configurations in which one of the $4s$ electrons of gallium is excited to a higher s state, s'. The theory is equally applicable to indium and thallium. Under the stated assumption Schwartz derives the following expressions for the effective electron density at the nucleus for the $P_{\frac{1}{2}}$ and $P_{\frac{3}{2}}$ states in terms of $b_{p\frac{1}{2}}$ and b_s, the density at the

TABLE II. Comparison of theoretical and experimental hfs anomaly ratios in the ground state of group III elements.

Isotopes	θ	$a_{\frac{1}{2}}/a_{\frac{3}{2}}$	$b_s/b_{p\frac{1}{2}}$[a]	$b_{\frac{3}{2}\,\mathrm{eff}}/(b_{\frac{1}{2}\,\mathrm{eff}}-b_{\frac{3}{2}\,\mathrm{eff}})$	$\Delta_{\frac{3}{2}}/(\Delta_{\frac{1}{2}}-\Delta_{\frac{3}{2}})$
$\mathrm{Ga}^{69,71}$	1.10[b]	7.02[b]	35.05	−0.79	−0.80 ±0.09[c,d]
$\mathrm{In}^{113,115}$	1.30[b]	9.423	11.90	−0.762	−0.760±0.043
$\mathrm{Tl}^{203,205}$	2.416[e]	80.5[e]	3.41	−0.939	−0.939±0.001[c,e]

[a] $b_s/b_{p\frac{1}{2}}=k(1+\rho)/3(1-\rho)$, where k is the correction factor which takes account of the shielding of the $p_{\frac{1}{2}}$ electron.[14]
[b] C. Schwartz, Phys. Rev. **97**, 380 (1955).
[c] See reference 8.
[d] R. T. Daly, Jr., and J. H. Holloway, Phys. Rev. **96**, 539 (1954).
[e] G. Gould, Phys. Rev. **101**, 1828 (1956).

[10] G. Breit and J. E. Rosenthal, Phys. Rev. **41**, 459 (1932).
[11] A. Bohr and V. F. Weisskopf, Phys. Rev. **77**, 94 (1950).
[12] A. Bohr, Phys. Rev. **81**, 331 (1951).
[13] M. F. Crawford and A. L. Schawlow, Phys. Rev. **76**, 1310 (1949).
[14] C. Schwartz, Phys. Rev. **105**, 173 (1957).
[15] C. Schwartz, Phys. Rev. **99**, 1035 (1955).

nucleus for a $p_{\frac{1}{2}}$ and $s_{\frac{1}{2}}$ electron, and $a_{\frac{1}{2}}/a_{\frac{3}{2}}$, the experimental ratio of the dipole interaction constant in the $P_{\frac{1}{2}}$ state to that in the $P_{\frac{3}{2}}$ state:

$$b_{\frac{1}{2}\text{ eff}} = (1-\beta_{\frac{1}{2}})b_{p\frac{1}{2}} + \beta_{\frac{1}{2}}b_s, \quad b_{\frac{3}{2}\text{ eff}} = \beta_{\frac{3}{2}}b_s,$$

where

$$\beta_{\frac{1}{2}} = (1-5\theta a_{\frac{3}{2}}/a_{\frac{1}{2}})/(1+5\theta), \quad \beta_{\frac{3}{2}} = -(a_{\frac{1}{2}}/a_{\frac{3}{2}})\beta_{\frac{1}{2}}.$$

θ is a relativistic correction factor. The applicability of the theory can be tested by comparing the experimental value of $\Delta_{\frac{3}{2}}/(\Delta_{\frac{1}{2}}-\Delta_{\frac{3}{2}})$ with the calculated value of

$$b_{\frac{3}{2}\text{ eff}}/(b_{\frac{1}{2}\text{ eff}} - b_{\frac{3}{2}\text{ eff}}) = \beta_{\frac{3}{2}}/[\beta_{\frac{1}{2}} - \beta_{\frac{3}{2}} + (1-\beta_{\frac{1}{2}})b_{p\frac{1}{2}}/b_s].$$

In Table II we have listed the results for gallium, indium, and thallium along with the values of the quantities necessary for the calculation. The good agreement between the calculated and experimental ratios is strong support for Schwartz conclusion[14] that it is the $(s\rightarrow s')$ excitation of the s electrons that is most important and that the alternative mode of excitation $(s\rightarrow d)$, which may be of importance in boron, can safely be ignored in the heavier group III elements.

In order to obtain a numerical value for the theoretical anomaly it is necessary to know how the nuclear radius changes in going from In^{113} to In^{115}, i.e., we must be able to evaluate $\delta R/R$. Though the relation $R = 1.2\times10^{-13}A^{\frac{1}{3}}$ cm may be expected to give the gross behavior of R as a function of A, we cannot expect it to give the correct value of the variation of R between two isotopes. An apparent value of $\delta R/R$ can be obtained by equating the theoretical and experimental values of $\Delta_{\frac{1}{2}}-\Delta_{\frac{3}{2}}$. Using

$$\Delta_{\frac{1}{2}}-\Delta_{\frac{3}{2}} = (1-\beta_{\frac{1}{2}})\Delta_{BR}(p_{\frac{1}{2}}) + (\beta_{\frac{1}{2}}-\beta_{\frac{3}{2}})\Delta_{BR}(s)$$

we obtain for indium

$$\delta R/R = 3.66\times10^{-3}$$

or about 0.6 times the value of 5.85×10^{-3} obtained by assuming an incompressible nucleus $[(\delta R/R) = (\delta A/3A)]$. This decrease in $\delta R/R$ is in agreement with the simple theory of nuclear compressibility as formulated by Wilets, Hill, and Ford.[16]

It should be pointed out that while there is no disagreement in the case of indium within experimental error and theoretical uncertainty between the hfs anomaly data and the present theory of nuclear compressibility, there is considerable difficulty in interpreting the results for thallium. The experimental value of $\Delta_{\frac{1}{2}}-\Delta_{\frac{3}{2}}$ for thallium[8,17] is $(1729.4\pm0.7)\times10^{-6}$. If we again neglect the distributed moment anomaly (which is only 12% of the observed anomaly), we obtain, assuming $R = 1.2\times10^{-13}A^{\frac{1}{3}}$ cm and $p = 8/9$, $\delta R/R = 4.7\times10^{-3}$. This is 1.4 times the value of 3.3×10^{-3} obtained by assuming an incompressible nucleus. The theory of reference 16 predicts a value of $\delta R/R$ equal to approximately $\frac{1}{2}$ the incompressible nucleus value. Isotope shift data for thallium[13] gives $\delta R/R = 1.7\times10^{-3}$, in good agreement with reference 16 and in marked disagreement with the hfs anomaly value of $\delta R/R$.

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[16] Wilets, Hill, and Ford, Phys. Rev. **91**, 1488 (1953).
[17] G. Gould, Phys. Rev. **101**, 1828 (1956).