Nuclear Spin of Ho^{166}†

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The hyperfine structure of Ho^{166} has been examined by means of the atomic-beam magnetic-resonance technique. The atomic ground state appears to be $^4I_{15/2}$, and g_J has the measured value 1.19509 ± 0.00007, in close agreement with the Russell-Saunders value of 6/5. Only one resonance is observed, and its transition frequency for $0.15\leqslant H\leqslant 150$ gauss is proportional to the magnetic field strength to within experimental error. The simplest interpretation is that the nuclear spin is $I=0$. It is shown that if $I=1$, the magnetic hyperfine interaction constant a must be less than 5 kc/sec.

INTRODUCTION

ISOTOPES in the rare earth region have received a considerable amount of attention from nuclear physicists during the last few years. One of the most striking results of this research was the discovery of large collective effects.

The application of atomic-beam magnetic-resonance techniques to these isotopes, although hindered until recently by a number of experimental difficulties, is now proceeding. In particular, recent research[1,2] on the beta-gamma spectroscopy of the decay products of Ho^{166} indicated the desirability of a direct measurement of the spin.

GENERAL PRINCIPLES

The general features of the atomic-beam magnetic-resonance technique for determining nuclear spins have been extensively treated. Accordingly, only a brief outline will be given here.

In the scheme of Rabi,[3] as modified by Zacharias,[4] the material to be investigated is vaporized in an oven with a narrow slit. The emerging beam then passes in turn through three magnetic fields. The first and third are strong and inhomogeneous while the central field has no gradient. The inhomogeneous fields and their gradients are arranged in such a way that the deflections suffered by an atom of the beam are equal in magnitude and in the same direction (away from the detector) unless a suitable change in the effective magnetic moment of the atom occurs between the deflections. If such a change of state occurs, the second deflection will be reversed in direction and will refocus the atom to a central detector. Such observable transitions can be induced by an appropriately oriented rf magnetic field of the proper frequency, in the central, homogeneous magnetic field. The resonant frequency is a function of all the parameters that appear in the Hamiltonian (see "Theory of the Method," below) and measurement of the frequency at a number of known intensities of the homogeneous magnetic field can yield information on I if enough is known of the other parameters.

SOURCE PREPARATION

The 27-hr Ho^{166} source was formed by neutron capture (cross section = 64 b) in 100% abundant Ho^{165}. Overnight irradiations were arranged at a flux of 2×10^{13} neutrons cm^{-2} sec^{-1} in the Argonne research reactor *CP*-5. About 250 mg of Ho provided a source strong enough to be useful for about a week.

EXPERIMENTAL DETAILS

Ovens. Since it was anticipated that a relatively high temperature would be required to produce a Ho^{166} beam of adequate intensity, tantalum was selected for the oven material. A small amount of sodium metal was added to the irradiated Ho to facilitate beam alignment. It was found that a temperature of about 1100°C was sufficient to give a strong beam of radioactive Ho^{166} on the first day of operation. As the sample became less active, however, higher temperatures were required.

A second oven, containing sources of both K and Cs, was left in position at all times for magnetic field calibration. It was mounted above and a little to one side of the centrally located Ho oven.

Homogeneous magnetic field. Prior to investigation of the Ho, several changes were made in the C magnet to improve its homogeneity. The procedure followed and the results achieved have been described previously.[5]

Calibration of the homogeneous field, conventionally called the "C" field, is achieved by means of the Cs^{133} or K^{39} beams from the auxiliary oven mentioned above. The alkali beams are detected by an off-center hot tungsten wire used as a surface ionization detector. Radio-frequency resonances can be observed in either K^{39} or Cs^{133} and thus the C field can be monitored continuously without interfering with the radioactive beam.

Two rf loops are available in the C field, and they can be cross-calibrated by observing the same resonance in turn with each loop. Such studies indicate that there is seldom a difference in frequency of as much as 2 kc/sec

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[1] R. L. Graham, J. L. Wolfson, and M. A. Clark, Phys. Rev. **98**, 1173 (1955).
[2] R. G. Helmer and S. B. Burson, Phys. Rev. **119**, 788 (1960).
[3] I. I. Rabi, J. R. Zacharias, S. Millman, and P. Kusch, Phys. Rev. **53**, 318 (1938).
[4] J. R. Zacharias, Phys. Rev. **61**, 270 (1942).
[5] L. S. Goodman, Rev. Sci. Instr. (to be published).

at frequencies up to 100Mc/sec. Although an exhaustive investigation has not been made (and need not be made if the loops are cross-checked prior to each run), it appears that varying the magnet history does not increase the difference in field at the two loop positions (2 cm apart) by more than 1–3 milligauss. Much larger effects were common before altering the magnet assembly as described previously.[5]

Operation at very low fields. The Majorana flop-in pattern at zero field was investigated by use of a beam of stable Ga. It was found that no flop-in occurred at fields above 0.1 gauss, and that above 0.075 gauss the amount occurring was too small to interfere with resonance work. Accordingly, it was decided to begin the search for Ho^{166} transitions at 0.15 gauss where the searching required would be nearly a minimum. The calibration resonances in Cs^{133} and K^{39} at 0.15 gauss were clean and easy to use.

Collection technique. The Ho^{166} beam was detected by allowing it to condense on copper-coated steel collectors, which were removed from the vacuum system after exposure and counted in gas-flow Geiger counters. The copper surface was deposited on the steel by momentarily dipping the collector, after preparation by etching, into a dilute solution of $CuSO_4$. The collectors were then kept in alcohol until immediately before insertion into the vacuum system for use. No special care of the collectors was required after removal from the vacuum.

THEORY OF THE METHOD

The Hamiltonian describing the energy levels of an atom, with nuclear spin I and electronic angular momentum J, in an external magnetic field may be written[6]

$$\mathcal{H}=ha\mathbf{I}\cdot\mathbf{J}+g_JJ_z\mu_0H+g_II_z\mu_0H+hbQ_{\text{op}}, \qquad (1)$$

where terms of order higher than electric quadrupole have been omitted. In this expression a and b are parameters to be determined, and g_I and g_J are the nuclear and electronic g factors, respectively, and are defined by $g_I=-\mu_I/I$ and $g_J=-\mu_J/J$. In addition, μ_0 is the Bohr magneton, H the external magnetic field, and Q_{op} is the electric quadrupole interaction operator. The z axis is chosen to coincide with the direction of the external magnetic field. All quantities are defined as in reference 6.

It should be noted that although the Hamiltonian contains the atomic inputs J and g_J, neither of these quantities was known for the atomic ground state of Ho. It was suggested[7] to the authors that Ho would most likely have eleven f electrons, and an inverted ^4I configuration. Thus, the ground state anticipated was $^4I_{15/2}$, for which the Russell-Saunders g_J value is calculated to be 6/5.

If the nuclear spin were indeed 0, as proposed by the nuclear spectroscopists,[1,2] the Hamiltonian would immediately yield the eigenvalues $g_Jm_J\mu_0H$, so that the observable transition frequency ($\Delta m_J=\pm1$) becomes $g_J\mu_0H$ as expected. Any departure of the transition frequency from linearity as the magnetic field is varied would indicate a spin $I>0$.

If the nuclear spin were 1, three zero-field hyperfine levels, characterized by total angular momenta $F=17/2$, 15/2, and 13/2, would be expected from the $^4I_{15/2}$ ground state, and each of these would produce an observable transition at nonzero field. If the nuclear moments (and consequently, the hyperfine interaction constants a and b) were very small, all nonlinearity of the observed frequencies would occur only at very weak fields, and consequently the spin might appear to be zero. Similarly, if the hyperfine separations were very large, the linear Zeeman region would extend to strong fields. For most atoms with nonzero spin, a situation between these two extremes holds, and the nonlinearity can be measured precisely so that definite spin assignments can be deduced. It is clear, however, that if no nonlinearity is detected, one cannot make a definitive spin determination; rather one can only hope to place limits on a, and b, and thus indirectly on μ_I and Q.

MEASUREMENTS

Table I summarizes the results of the measurements on Ho^{166}, together with the data associated with calibration of the homogeneous field in which the transitions were induced. It is seen that, within experimental error, no nonlinearity could be detected, and thus the most reasonable interpretation is that the nuclear spin of Ho^{166} is $I=0$. Figure 1 displays the data graphically.

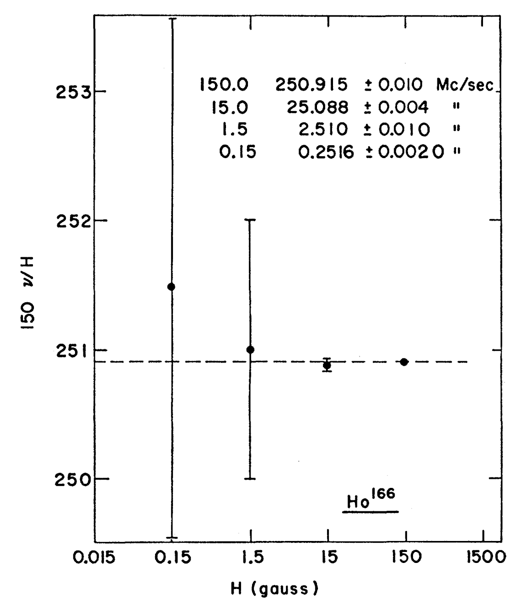

FIG. 1. Graphical presentation of the Ho^{166} data to display the observed linearity of resonance frequency with field strength. The resonance frequency for each value of the magnetic field is given in the figure.

[7] M. Fred (private communication, 1960).

[6] N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956), p. 272.

The observed resonance at 0.15 gauss (shown in Fig. 2) at 0.2516 ± 0.0020 Mc/sec is to be compared with the frequency of 0.2519 Mc/sec that was calculated for spin 0 on the basis of the Russell-Saunders value of g_J for a ${}^4I_{15/2}$ state. This result suggests that the atomic ground state is ${}^4I_{15/2}$ as anticipated and that its g_J value is close to the Russell-Saunders value. Some evidence that some of the atoms in the beam were in the ${}^4I_{13/2}$ and ${}^4I_{11/2}$ states was found at 0.15 gauss when operating at greater rf power than for the run shown in Fig. 2.

Although no activity other than that of Ho^{166} was expected in the source, a check was made on the half-life of atoms collected on resonance at 150 gauss to provide more positive identification of the atoms undergoing resonance. The 28 ± 1 hr half-life obtained agrees closely with the reported[8] value of 27.3 hr for Ho^{166}.

DISCUSSION

The simplest interpretation of the data is that the nuclear spin of Ho^{166} is 0. As mentioned above, the transition frequency is then given by $g_J\mu_0H$, from which

TABLE I. Summary of the data obtained on Ho^{166}. All frequencies are in Mc/sec. The resonance at 0.15 gauss was observed five times, and that at 15 gauss twice. The $(4, -3 \leftrightarrow 4, -4)$ transition in Cs^{133} was used for field calibration. The value[a] $g_J(\text{Cs})=2.002577$ was used in calculating the Cs frequencies, and the g_J value quoted for Ho is thus relative to the Cs value.

H (gauss)	Cs^{133} frequency	Ho^{166} frequency
0.15	0.0525	0.2516 ± 0.0020
1.5	0.525	$2.510\ \pm0.010$
15	5.269	$25.088\ \pm0.004$
150	54.661	$250.915\ \pm0.010$

[a] See reference 9.

g_J can be simply extracted. The value obtained in this way is

$$g_J(\text{Ho}^{166})=1.19509\pm0.00007,$$

relative to $g_J(\text{Cs})=2.002577$.[9]

The observed linearity of the resonance frequency with magnetic field strength does not, however, establish that the spin is 0. If the spin were 1, then very small or very large values of a could also be consistent with the data. If the spin is 1 and we make the reasonable assumption that $J=15/2$, F may take on the values 17/2, 15/2, and 13/2. (The justification of the assignment $J=15/2$ rests on the close agreement between the g_J value deduced experimentally for either $I=0$ or $I=1$ and that calculated for the expected Russell-Saunders ${}^4I_{15/2}$ state.) An observable $\Delta F=0$ transition may be expected to arise from each of these three hyperfine levels, and the resonance frequencies can be calculated as functions of a, b, g_J, and H. In addition, approximate

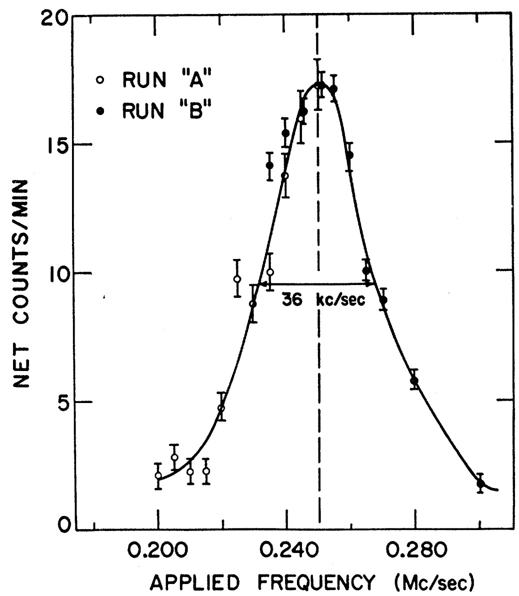

FIG. 2. The Ho^{166} resonance as observed at 0.15 gauss at low rf power. At higher power levels, there is some evidence of resonances associated with other ${}^4I$ states.

values for the relative transition probabilities of the three resonances may be obtained. An extensive program of such calculations was carried out on the digital computer GEORGE. The calculations were made for a wide choice of values of a and b/a, so that all four possible zero-field hyperfine orderings were considered.

The calculations show that a linear field dependence of transition frequencies may be expected either for very small a (where the external field H is effectively very strong and I and J are decoupled) or for very large a (where the external field is effectively very weak and I and J are tightly coupled).

Calculations at 0.15 gauss show that for $a\geqslant 150$ kc/sec, the three resonances should occur at frequencies $0.238g_J$ $(F=13/2)$, $0.207g_J$ $(F=15/2)$,[10] and $0.185g_J$ $(F=17/2)$, all in Mc/sec, unless the ratio b/a is very close to one of the values -10, 70/9, or 140. When b/a approaches any one of these three values, two of the three zero-field hyperfine (F) levels approach the same energy, and the

[8] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).

[9] V. W. Hughes in *Recent Research in Molecular Beams*, edited by I. Estermann (Academic Press, Inc., New York, 1959), p. 87.

[10] It is interesting to note that this resonance $(F=15/2)$ is the only one of the three whose frequency, for strong effective magnetic field, is proportional to H. Since the resonance frequency of the observed transition is also proportional to H for $0.15\leqslant H\leqslant 150$ gauss (see Table I), it is tempting to assume this identification, for $I=1$, and use it to place an upper limit on a. There are two dangers in this procedure, however. The resonance frequency for the $F=15/2$ transition at 0.15 gauss has been calculated to be $0.210g_J$ for a strong effective field ($a\leqslant 30$ kc/sec), and $0.207g_J$ for a weak effective field ($a\geqslant 150$ kc/sec). The small a dependence of this resonance frequency and the lack of precise knowledge of g_J (g_J is determined only after establishing that a is small) lead to the conclusion that comparison of the observed frequency with the extremes calculated is, without other supporting arguments, a poor criterion for placing limits on a. The second objection is still more serious. The frequencies calculated above are valid only if the value of b/a is not too close to either -10 or 70/9, and thus a systematic investigation of the dependence of all three transition frequencies on both a and b/a becomes desirable.

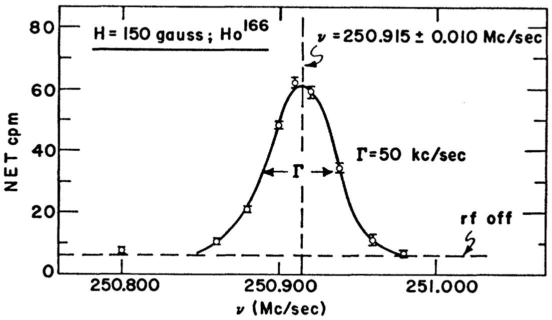

FIG. 3. The Ho^{166} resonance as observed at 150 gauss. The observed width is 50 kc/sec.

two corresponding resonances approach each other in frequency, while the third resonance frequency remains unaffected (to within 1 kc/sec).

Since only one resonance is observed at 0.15 gauss, it may be identified with any of the three anticipated. When this is done for $I=1$, and $a \geqslant 150$ kc/sec, g_J can be evaluated for every value of b/a and the resonance frequencies for the other two transitions can be calculated. The relative transition probabilites of the three transitions can also be calculated. The frequency range covered in the search for resonances at 0.15 gauss included, on both sides of the single observed transition, all frequencies at which the other two transitions might be expected to appear. Comparison of the observed spectrum containing the single resonance at 0.2516 ± 0.0020 Mc/sec (Fig. 2), with the detailed calculations shows that there is no value of b/a for which the experimental results are consistent with the calculated frequencies for $I=1$ and $a \geqslant 150$ kc/sec. It is thus clear that if $I=1$, $a \leq 150$ kc/sec regardless of the value of b.

Thus, if $I=1$, it has been shown that $a \leq 150$ kc/sec. Calculations show that for a as small as this, 150 gauss represents a very strong effective field. At such a strong field, the frequencies of the three $\Delta F=0$ transitions may be accurately approximated by the relation

$$h\nu = g_J \mu_0 H + ham_I, \tag{2}$$

where m_I, the projection of I on the field axis, may assume the values 0 and ± 1. This expression is valid for arbitrary b. It is thus possible to evaluate g_J as

$$g_J = (h/\mu_0) d\nu/dH. \tag{3}$$

The measured value of g_J is thus the same for $I=1$ as that obtained under the interpretation $I=0$. When this value for g_J is used at 0.15 gauss, the quantity $g_J\mu_0 H$ has the value 0.2509 Mc/sec compared with the observed frequency 0.2516 ± 0.0020 Mc/sec. On substitution of these two numbers into Eq. (2), it is clear that $ham_I \leq 3$ kc/sec. Consequently, either $m_I=0$ for the transition observed, or $a \leq 3$ kc/sec.

Since the spacings between the three frequencies predicted above must be a, one may search the frequency range on either side of the observed transition for companions. This was done at 150 gauss over a region 350 kc/sec immediately below resonance. The corresponding search above resonance was unnecessary since it is established that $m_I=0$ for the transition observed. The appearance of the resonance at 150 gauss is shown in Fig. 3.

Since no companion resonance was found, one may conclude either that $a \geqslant 350$ kc/sec or that all three resonances are contained within the observed transition. If the observed resonance, which has a width of 50 kc/sec, does contain all three transitions, we must have $a \leq 15$ kc/sec. Since we have already shown that a cannot be larger than 150 kc/sec, we may conclude that if $I=1$, $a \leq 15$ kc/sec regardless of b. Actually, examination of the transition as observed at 15 gauss (Fig. 4), where the full width at half maximum is 14 kc/sec, makes it possible to reduce the upper limit on a to 5 kc/sec for $I=1$.

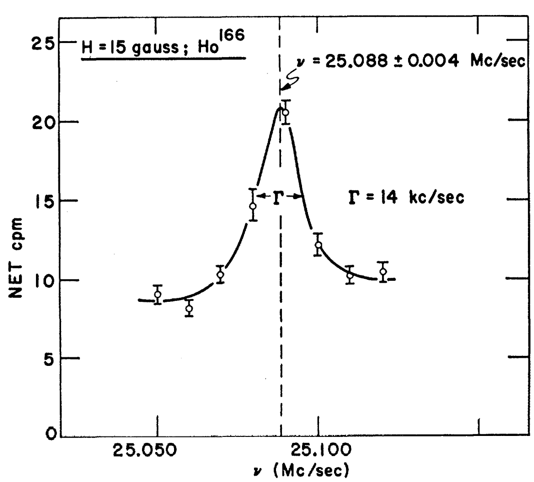

FIG. 4. The Ho^{166} transition as observed at 15 gauss. The observed width is 14 kc/sec.

CONCLUSIONS

The simplest interpretation of the present data is that $I=0$ for Ho^{166}. If the spin is 1, it has been shown that $a \leq 5$ kc/sec. The possibility of a spin larger than 1 was not investigated, but has been excluded on other grounds.[1,2]

The measured value for g_J (for $J=15/2$) is 1.19509 ± 0.00007, a result which is valid for either $I=0$ or $I=1$ (if $I=0$, the result for g_J is independent of J). The closeness of this value to 6/5 implies strongly that the atomic ground state is a Russell-Saunders $^4I_{15/2}$ state, although the present work does not constitute a measurement of J.[11]

The essential conclusions presented above were reached independently by R. Marrus *et al.*[12]

[11] L. S. Goodman and K. Schlüpmann (private communication) at Heidelberg have recently established that $J=15/2$ for the atomic ground state of Ho. This result confirms the tentative determination of J in this paper.

[12] L. S. Goodman, W. J. Childs, R. Marrus, I. P. K. Lindgren, and A. Y. Cabezas, Bull. Am. Phys. Soc. 5, 344 (1960).