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Hyperfine Structure and Nuclear Moments of Thulium-166†

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The hyperfine structure of 7.7-h Tm^{166} has been studied using the atomic-beam magnetic resonance technique. The results are $|A|=19\pm6$ Mc/sec, $|B|=7700\pm300$ Mc/sec, and $B/A<0$. Values of the nuclear moments are calculated from the hyperfine interaction constants A and B for Tm^{166} using known values for A, B, μ_I, Q for Tm^{170}. Results for Tm^{166} are $|\mu_I|=0.05\pm0.03$ nm and $|Q|=4.6\pm0.7$ b with $Q/\mu_I>0$. These results are discussed from the standpoint of the deformed-nucleus model of Mottelson and Nilsson.

INTRODUCTION

IN a previous paper[1] we described work performed at this laboratory to measure the nuclear spins of the ground states of Tm^{166} and Tm^{167}. The ground state of 7.7-h Tm^{166} was found to have $I=2$.

We have determined the magnetic dipole moment and electric quadrupole moment of this Tm^{166} ground state by the atomic-beam magnetic resonance method. These determinations were made from the values of the hyperfine interaction constants A and B which were determined by observing multiple quantum transitions at several static field values. We have made use of determinations of the electronic ground-state configuration of thulium[2] and measurements of g_J, A, B, Q, and μ_I for Tm^{170} made elsewhere.[3] Using these measurements of atomic and nuclear properties for Tm^{170} and our values of A and B, we have obtained values for the nuclear moments of Tm^{166}.

THEORY

For the Hamiltonian of a free atom in an external magnetic field of magnitude H_c we may write[4]

$$\mathcal{H}=g_J\mu_0H_cJ_z+g_I\mu_0H_cI_z+hA(\mathbf{I}\cdot\mathbf{J})+hB\frac{3(\mathbf{I}\cdot\mathbf{J})^2+\frac{3}{2}(\mathbf{I}\cdot\mathbf{J})-I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}. \quad (1)$$

We have adopted the convention which defines the g_J of a free electron as positive. Octupole and higher-order interactions have been omitted. In this equation,

$$A=-1/h(\mu_I/I)[H(0)]/J,\qquad B=eQ(\partial^2V/\partial z^2)_0, \quad (2)$$

in which $[H(0)]$ is the magnetic field at the nucleus produced by the atomic electrons and $(\partial^2V/\partial z^2)_0$ is the electric field gradient at the nucleus due to the electrons.

At zero external field, the eigenvalues of the above Hamiltonian are the hyperfine energy levels. The energies labeled by total angular momentum quantum number F are

$$\begin{aligned}E_{11/2}&=7hA+(14/56)hB,\\E_{9/2}&=(3/2)hA-(19/56)hB,\\E_{7/2}&=-3hA-(16/56)hB,\\E_{5/2}&=-(13/2)hA+(5/56)hB,\\E_{3/2}&=-9hA+(30/56)hB.\end{aligned}$$

These energy levels are each $(2F+1)$-fold degenerate for zero external field. The hyperfine structure separations are defined as $\Delta\nu_F=(E_F-E_{F-1})/h$. From the above, we have

$$\begin{aligned}\Delta\nu_{11/2}&=(11/2)A+(33/56)B,\\\Delta\nu_{9/2}&=(9/2)A-(3/56)B,\\\Delta\nu_{7/2}&=(7/2)A-(21/56)B,\\\Delta\nu_{5/2}&=(5/2)A-(25/56)B.\end{aligned} \quad (3)$$

For small values of the external field, we may use perturbation techniques to obtain the energy levels. Using the notation of Christensen,[5] we get the following:

$$E(F,m)=E_F+Rg_J\mu_0H_cm+\left\{\frac{Q(F^2-m^2)}{E_F-E_{F-1}}-\frac{P(F+1)^2-m^2}{E_{F+1}-E_F}\right\}(g_J\mu_0H_c)^2+\cdots. \quad (4)$$

Here,

$$P\equiv\frac{(F+1-J+I)(F+1+J-I)(J+I+F+2)(J+I-F)}{4(F+1)^2(2F+1)(2F+3)},$$

$$Q\equiv\frac{(F-J+I)(F+J-I)(J+I+1+F)(J+I+1-F)}{4F^2(2F-1)(2F+1)},$$

$$R\equiv\frac{J(J+1)-I(I+1)+F(F+1)}{2F(F+1)}.$$

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* Now at Clarendon Laboratory, Oxford, England.
[1] J. C. Walker and D. L. Harris, Phys. Rev. **121**, 224 (1961).
[2] W. F. Meggers, Revs. Modern Phys. **14**, 96 (1942).
[3] A. Cabezas and I. Lindgren, Phys. Rev. **120**, 920 (1960).
[4] N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956).
[5] R. L. Christensen, D. R. Hamilton, H. G. Bennewitz, J. B. Reynolds, and H. H. Stroke, Phys. Rev. **122**, 1302 (1961).

In (4), we have omitted terms containing g_I/g_J.[6] The present experiment was restricted to values of H_c sufficiently low so that terms in (4) higher than second order in H_c make negligible contributions to $E(F,m)$. That is, these contributions are much smaller than the energy widths of observed resonances.

The transitions observed in this experiment were those for which $\Delta F=0$. A single quantum transition obeys the dipole selection rule $\Delta m=\pm 1$. Other transitions may occur between two states which differ in m by more than one. These transitions involve the emission or absorption of more than one quantum. These multiple-quantum transitions are made up of a series of dipole transitions of the same frequency in which intermediate transitions are made to virtual states.[7] Thus, the frequency of an N-quantum transition is

$$\nu_{NQ}=\frac{E(F,m+N)-E(F,m)}{Nh}.$$

Using (4), this becomes

$$\nu=Rg_J\mu_0H_c/h+\left\{\frac{P}{\Delta\nu_{F+1}}-\frac{Q}{\Delta\nu_F}\right\}\times(N+2m)\left(\frac{g_J\mu_0H_c}{h}\right)^2+\cdots,$$

in which m is appropriate to the state on which the transition ends. Note that the frequency spacings among multiple-quantum transitions associated with the same m are equal to second order in H_c:

$$\delta\nu=\left\{\frac{P}{\Delta\nu_{F+1}}-\frac{Q}{\Delta\nu_F}\right\}\left(\frac{g_J\mu_0H_c}{h}\right)^2+\cdots.$$

If we define $\nu_0\equiv Rg_J\mu_0H_c/h$, we may write

$$\nu-\nu_0=(N+2m)\delta\nu=(N+2m)\times\left(\frac{P}{\Delta\nu_{F+1}}-\frac{Q}{\Delta\nu_F}\right)\left(\frac{g_J\mu_0H_c}{h}\right)^2. \quad (5)$$

From measurements of $\nu-\nu_0$ and the frequency separation of adjacent multiple quantum resonances the value of $|N+2m|$ can be inferred. g_J for thulium has been measured to be 1.14122 ± 0.00015.[3] The values of A and B can, therefore, be determined from the observations of multiple-quantum resonances at several values of H_c, using (3) and (5).

A radial wavefunction for the thulium ground-state electronic configuration has been calculated by Cabezas and Lindgren.[3] With this wavefunction, values of μ_I and Q for Tm^{170} were obtained from measurements of A and B. Using (2), we may write

$$\frac{A_{170}}{A_{166}}=\frac{(\mu_I/I)_{170}}{(\mu_I/I)_{166}} \quad\text{and}\quad \frac{B_{170}}{B_{166}}=\frac{Q_{170}}{Q_{166}}. \quad (6)$$

With these relationships and the values of A, B, μ_I, and Q for Tm^{170}, we can determine μ_I and Q for Tm^{166} from our measurements of A and B.

APPARATUS AND EXPERIMENT

The focusing atomic beam apparatus used for this experiment was built by Lemonick, Pipkin, and Hamilton.[8] It has been modified to incorporate a self-normalizing "flop-in" detection system described in more detail elsewhere.[9] With this system atoms which have undergone a transition from a state for which $m_J>0$ to a state for which $m_J<0$ are collected on an outer disk 2 in. in diam, while the atoms of the remainder of the beam are collected on a concentric inner button 0.6 in. in diam. After exposure in the apparatus, the button and disk are removed and counted separately in standard NaI scintillation counters. Resonances appear as increases in D/B, where D is the disk count rate and B the button count rate. Fluctuations of the beam intensity at the oven end of the apparatus do not affect the value of D/B.

The radioactive thulium was produced by two different nuclear reactions. For the initial experiments, Tm^{166} was produced by a (p,n) reaction on naturally occurring erbium (of which 33% is Er^{166}). For these bombardments the 18-MeV Princeton cyclotron was used. Subsequent bombardments using 40-MeV alpha particles were made with the cyclotron at the Brookhaven National Laboratory. The reaction of interest is Ho^{165} $(\alpha,3n)$ Tm^{166}.[10] Thulium beams were produced by evaporating the thulium from the holmium at a temperature of about 1060°C (as determined by a Leeds and Northrup optical pyrometer). When erbium was the parent material, the corresponding temperature was about 1150°C. A typical alpha bombardment of 12 μa h produced enough Tm^{166} for approximately 30 button-disk exposures each of 5-min duration. Such an exposure ordinarily produced several thousand counts per minute on the button portion of the collector. The collector surfaces were cleaned copper which was at room temperature.

We observed multiple quantum $\Delta F=0$ transitions for $F=11/2$, 9/2, and 7/2. In addition to multiple-quantum transitions corresponding to different values $N+2m$, there were multiple-quantum transitions with different values of N and m separately but with the same $N+2m$. These transitions occur at the same frequency to second order in H_c. The situation is illustrated in Fig. 1. The arrows represent transitions observed.

[6] The measured value of μ_I for Tm^{166} leads to a value of $(g_I)_{166}\cong10^{-5}$. This is smaller, by a factor of 10 than the uncertainty in the value of g_J used.

[7] M. N. Hack, Phys. Rev. **104**, 84 (1956).

[8] A. Lemonick, F. M. Pipkin, and D. R. Hamilton, Rev. Sci. Instr. **26**, 1112 (1955).

[9] O. Ames, A. M. Bernstein, M. H. Brennan, and D. R. Hamilton, Phys. Rev. **123**, 1793 (1961).

[10] G. Wilkinson and H. G. Hicks, Phys. Rev. **75**, 1370 (1949).

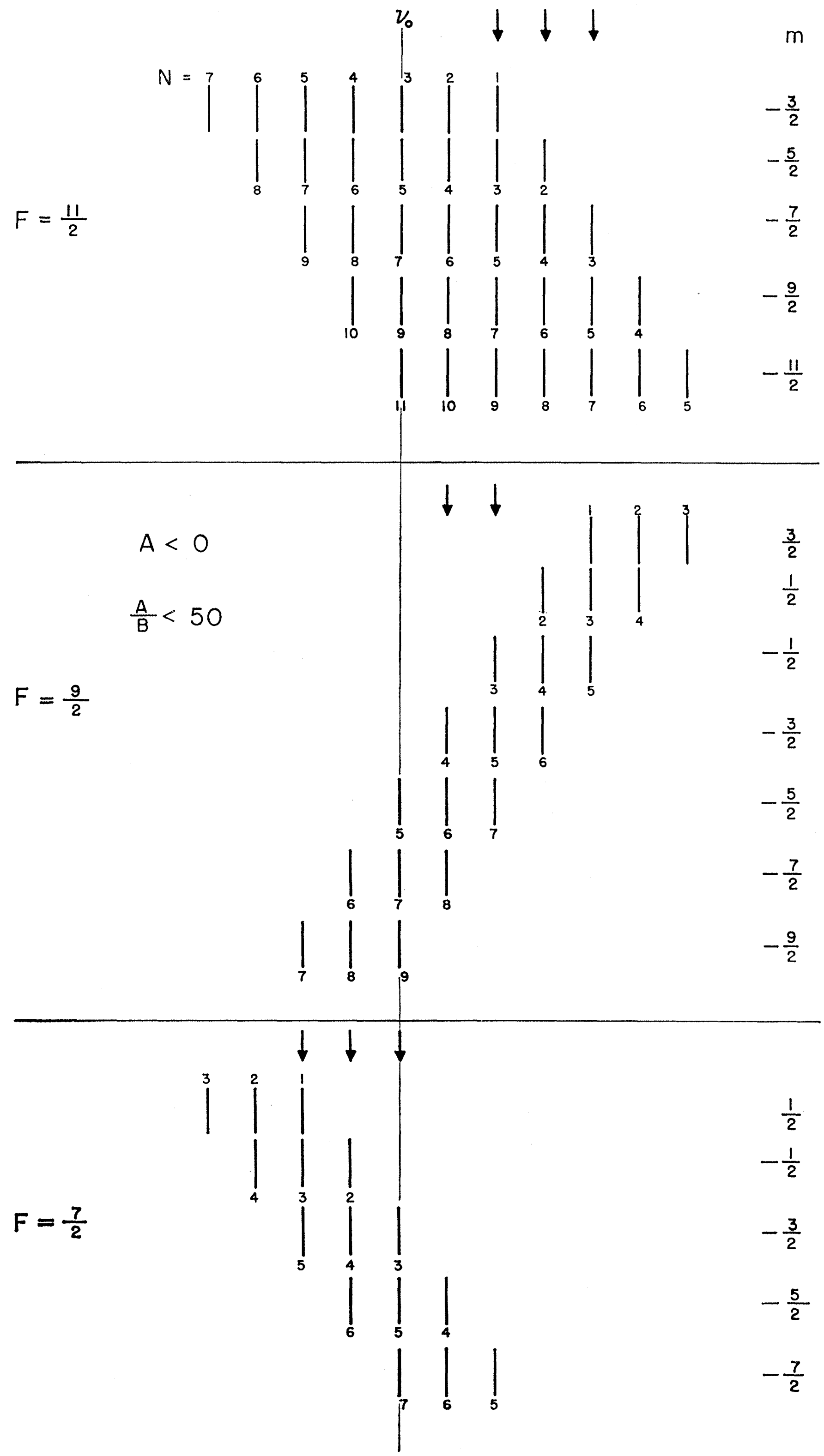

FIG. 1. Schematic multiple quantum spectra for three F values. Small numbers indicate quantum multiplicity, and rows are labeled by the m_F of the final state in the transition. Arrows represent resonances observed in this experiment.

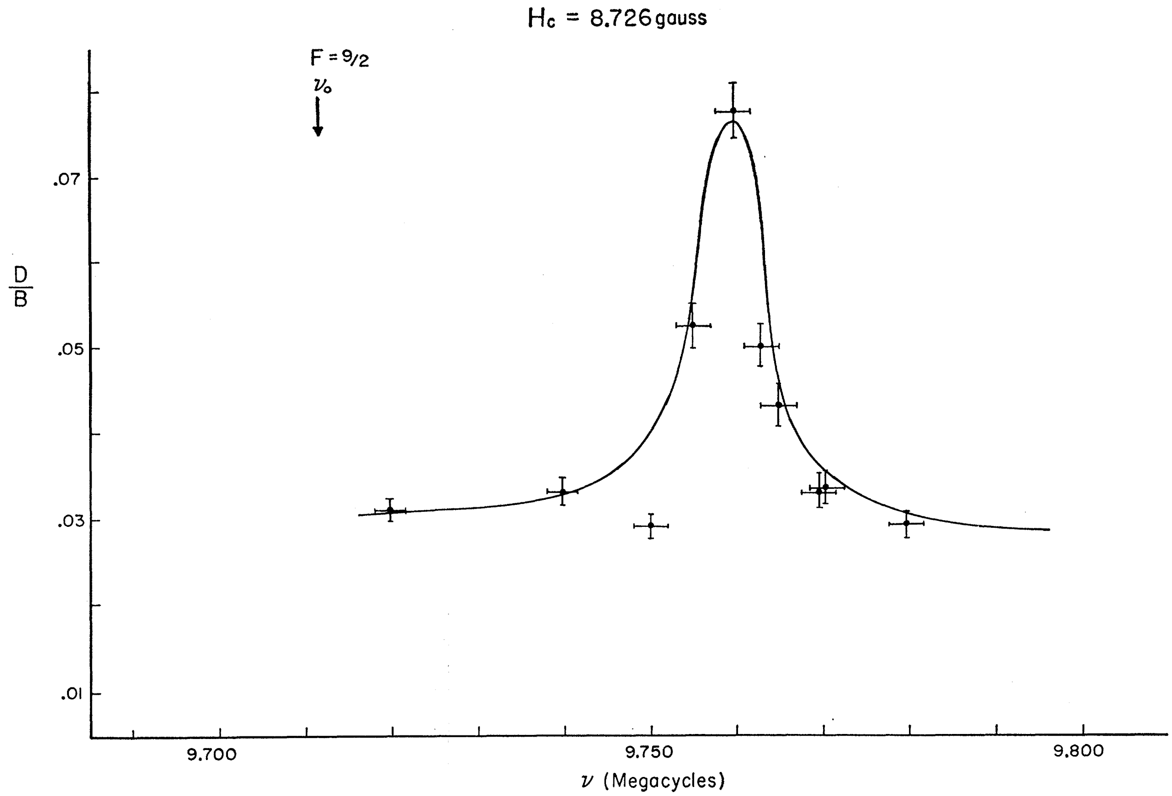

FIG. 2. Typical $\Delta F=0$ resonance in region of quadratic field dependence.

The degeneracy of these multiple-quantum transitions with respect to $N+2m$ raises some questions as to what value of rf field to use, as different multiple quantum transitions require different values of rf field to saturate the transition.[7] It was noticed during the spin measurements that too high a value of the rf field caused an anomalous shifting of the resonance to a slightly lower frequency. The appropriate H_{rf} for "seeing" multiple-quantum transitions in Tm^{166} was determined empirically. That is, the appropriate rf field value was chosen by trial and error starting from those values of H_{rf} which gave large unshifted resonances in the region of linear field dependence ($\nu-\nu_0\approx 0$). Care was taken to minimize the danger of power "pulling" of the multiple-quantum resonances which were observed in the region of quadratic field dependence.

The resonances observed in this experiment were quite narrow, particularly those for which $\nu-\nu_0$ differed appreciably from zero. A typical resonance is shown in Fig. 2. In some cases the width of the resonance at half maximum was ≈ 5 kc/sec. This small resonance width in principle allows more accurate determination of transition frequencies, but makes more stringent the requirement of a constant static magnetic field H_c. Most of the uncertainties in our values of $\nu-\nu_0$ (Fig. 3) come from uncertainties in the value of H_c (which result in uncertainties in ν_0). Resonance frequency shifts due to changes in H_c were sometimes larger than the frequency width of the resonances and made necessary rather frequency checks of the value of the static field.

RESULTS

Approximate values of $|A|$, $|B|$, and the sign of B/A were determined from Eqs. (3) and (5) and the experimental data by the method of least squares.[11] The least squares fit does not depend on the sign of A assumed, so that only the relative signs of A and B can be determined from the present data. The integral values of $|N+2m|$ were determined from spacings among multiple-quantum resonances observed at the same value of H_c. These $|N+2m|$ assignments were verified by comparing least squares fits for different sets of $|N+2m|$. The experimental data were consistent only for the particular values of $|N+2m|$ determined previously from the multiple-quantum spacings. Figure 3 shows the results of the least squares fit with values of $|A|$ and $|B|$ so determined. Including the errors as obtained from the least squares procedure, the results are

$$|A|=19\pm 6\ \text{Mc/sec},$$

$$|B|=7700\pm 300\ \text{Mc/sec},$$

$$B/A<0.$$

[11] J. Topping, *Errors of Observation and Their Treatment* (Institute of Physics Monograph, London, 1958).

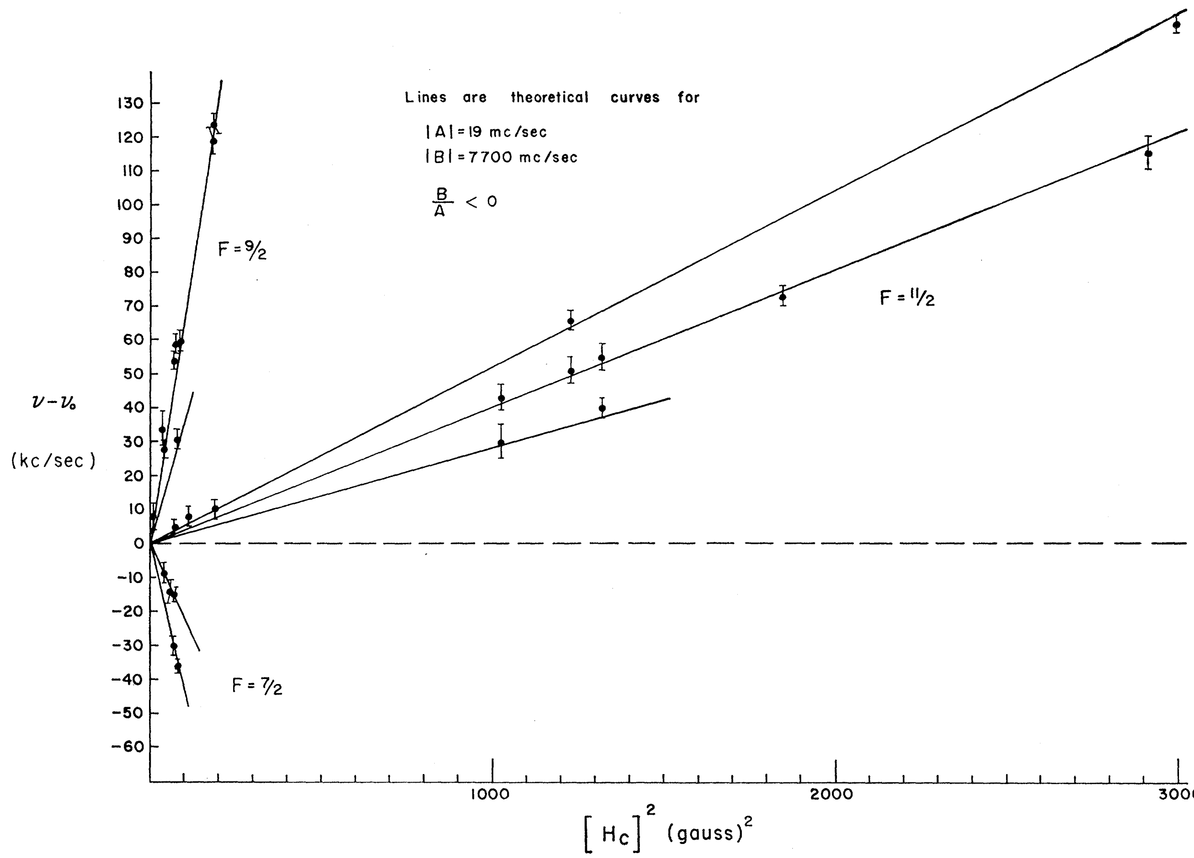

FIG. 3. Results of least-squares fit.

A curious result of the unusually large value of B/A is the negative value of $\nu-\nu_0$ for the observed $F=7/2$ resonances over the range of H_c used in this experiment (Figs. 1 and 3). The unusual zero-field hyperfine level ordering (Fig. 4) results from the large electric quadrupole interaction.

Using Eqs. (6), the values of the nuclear moments and $|A|$, $|B|$, and B/A for Tm^{170} we can determine[3] $|Q|$, $|\mu_I|$, and the sign of Q/μ_I for Tm^{166}. The results are

$$|\mu_I| = 0.05\pm0.03 \text{ nm},$$

$$|Q| = 4.6\pm0.7 \text{ b},$$

and $Q/\mu_I>0$.

DISCUSSION

In the case of Tm^{166} the quadrupole moment arises entirely from a deformation of the whole nucleus from sphericity. The quadrupole moment of a uniformly charged ellipsoid of revolution is

$$Q_0=(4/5)Zr_0^2\delta[1+(7/6)\delta+\cdots]. \quad (7)$$

The deformation parameter δ is defined as $(a-b)/r_0$, where a and b are major and minor axes of the ellipsoid, and r_0 is the radius of a shpere with volume equal to that of the ellipsoid. Z is the ratio of the total charge to the charge of a proton. However, a nuclear ellipsoid has rotational degrees of freedom and the Q measured in the laboratory is not the Q_0 seen by an observer at rest with respect to the ellipsoid. For nuclear ground states, Q and Q_0 are related by[12]

$$Q=[I(2I-1)/(I+1)(2I+3)]Q_0. \quad (8)$$

Using (7) and (8) and the measured values of Q and I for Tm^{166}, we may get an estimate of δ. In this case $Z=69$, and we use $r_0\simeq1.2A^{1/3}\times10^{-13}$ cm. The result is $\delta=0.43$. This is significantly larger than the theoretical estimate of 0.29 by Mottelson and Nilsson.[13] The value of δ for Tm^{166} is twice as large as the empirical value of $\delta_{170}=0.20$ ($|Q|=0.61$ b, $I=1$) for Tm^{170}. Only Er^{167} with $I=7/2$, $|Q|=10\pm2$ b, shows a larger deformation than Tm^{166}.

From the work of Hooke,[14] we obtain the following

[12] A. Bohr and B. R. Mottleson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

[13] B. R. Mottleson and S. G. Nilsson, Kgl. Danske Videnskab. Selskabs, Mat.-fys. Skrifter **1**, No. 8 (1958).

[14] W. M. Hooke, Phys. Rev. **115**, 453 (1959).

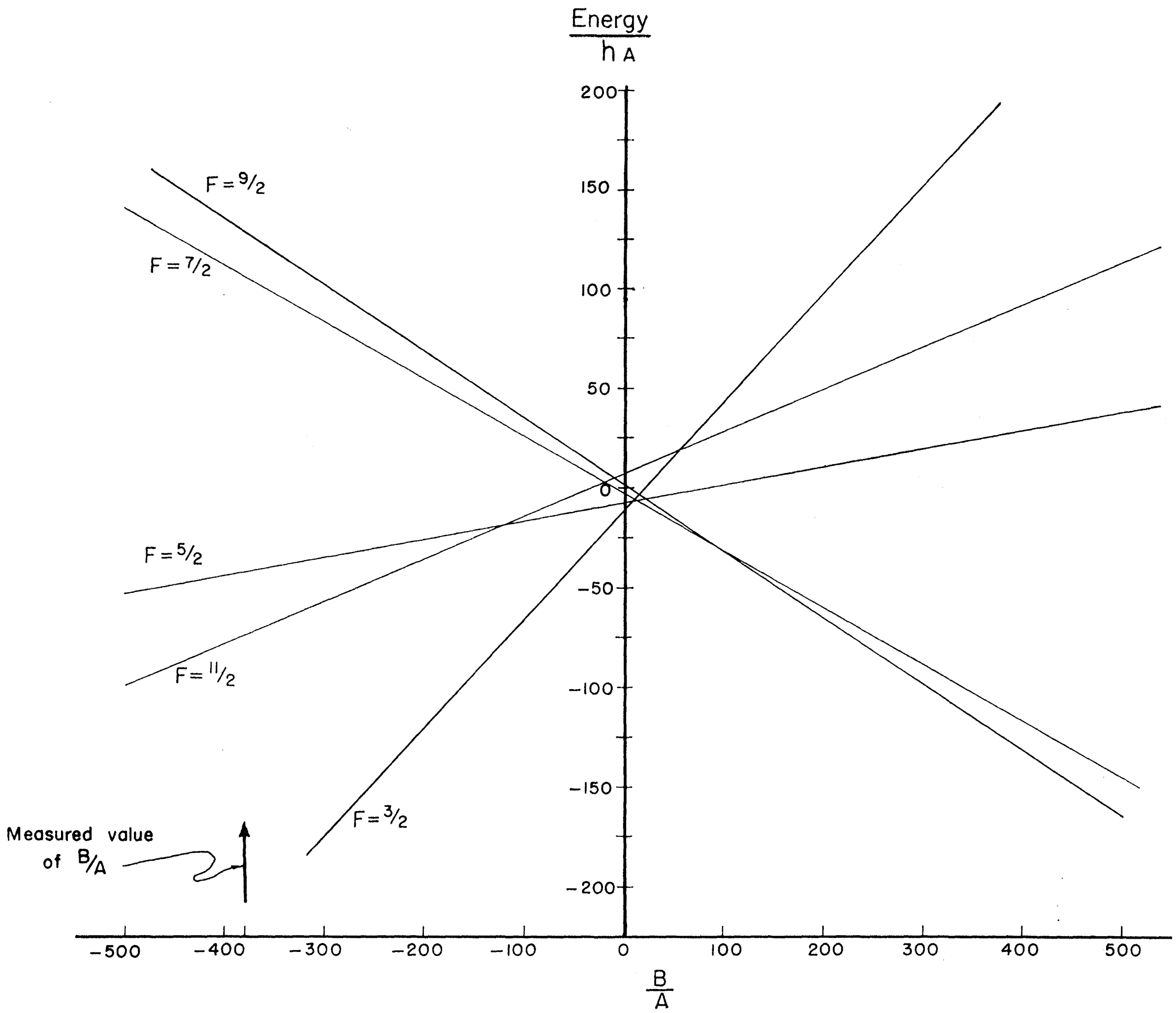

FIG. 4. Hyperfine level ordering vs B/A. The experimental value of B/A for Tm^{166} is indicated.

expression for μ_I for a deformed odd-odd nucleus:

$$\mu_I=[I/(I+1)][g_{sp}\langle S_{pz'}\rangle+g_{sn}\langle S_{nz'}\rangle+g_{lp}\langle l_{pz'}\rangle+g_R]. \quad (9)$$

In this equation $g_{sp}=+5.585$, $g_{lp}=1$, $g_{sn}=-3.826$. The classical value of g_R for a uniformly charged nucleus is Z/A. $\langle S_{pz'}\rangle$ is the expectation value of the projection of the **s** of the odd proton on the body symmetry axis of the nucleus. Gallagher and Moszkowski[15] have used the asymptotic Nilsson wavefunctions[16] in an equivalent expression for μ_I for odd-odd nuclei. In this case, the expectation values in (9) are replaced by the appropriate values of Σ and Λ. Σ and Λ are eigenvalues of the projections of **s** and **l** on the body symmetry axis for the case in which the terms containing $\mathbf{l}\cdot\mathbf{s}$ are omitted from the nuclear Hamiltonian. This makes the calculations particularly easy as the Nilsson orbitals are generally labeled by these asymptotic quantum numbers. That is, the $[411\frac{1}{2}]$ orbital has $\Lambda=1$, $\Omega=(\Lambda+\Sigma)=\frac{1}{2}$, so $\Sigma=-\frac{1}{2}$. Similarly, the $[642\frac{5}{2}]$ orbital has $\Lambda=2$, $\Omega=(2+\frac{1}{2})$; therefore $\Sigma=+\frac{1}{2}$. We expect to obtain a reasonable value for μ_I using the asymptotic Nilsson wavefunctions in cases in which δ is greater than 0.3. In this case, the asymptotic wavefunctions differ from the actual wavefunctions by less than 10%.[13]

According to the Nilsson diagram the $[411\frac{1}{2}]$ orbital should be assigned to the 69th proton for all thulium isotopes. This assignment is confirmed by the measured spins of 1/2 for Tm^{167}, Tm^{169}, and Tm^{171}. The assignment of a Nilsson orbital for the 97th neutron in Tm^{166} is more difficult. For odd-odd nuclei, where there is strong coupling of the odd nucleons to the deformed core, the ground-state spin is given by[12]

$$I=|\Omega_p\pm\Omega_n|.$$

[15] C. J. Gallagher and S. A. Moszkowski, Phys. Rev. **111**, 1282 (1958).

[16] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.- fys. Medd. **29**, No. 16 (1955).

As $\Omega_p = 1/2$, we expect either 5/2 or 3/2 for Ω_n. Mottelson and Nilsson[13] suggest that the 97th neutron orbital is $[523\frac{5}{2}]$ on the basis of the measured ground-state spin and moment of ${}_{66}Dy_{97}{}^{163}$. The $[642\frac{5}{2}]$ orbital which lies near the $[523\frac{5}{2}]$ orbital on the Nilsson diagram for $\delta \approx 0.3$ offers another possibility. We should also consider the $[521\frac{3}{2}]$ orbital which is nearby.

Gallagher and Moszkowski[15] have proposed the following coupling rules for deformed odd-odd nuclei:

$$I = \Omega_p + \Omega_n \quad \text{if} \quad \Omega_p = \Lambda_p \pm \tfrac{1}{2} \quad \text{and} \quad \Omega_n = \Lambda_n \pm \tfrac{1}{2},$$
$$I = |\Omega_p - \Omega_n| \quad \text{if} \quad \Omega_p = \Lambda_p \pm \tfrac{1}{2} \quad \text{and} \quad \Omega_n = \Lambda_n \mp \tfrac{1}{2}.$$

The $[642\frac{5}{2}]$ orbital for the 97th neutron is the only choice which is consistent with these coupling rules if the $[411\frac{1}{2}]$ orbital is assigned to the 69th proton.

Table I shows the "asymptotic" values of μ_I, calculated using (9), for each of the above possibilities for the odd-neutron orbital. Only the choice of the $[642\frac{5}{2}]$ orbital gives reasonable agreement with the measured value of the moment. The choice of the $[642\frac{5}{2}]$ state for the 97th neutron implies that the parity of the ground state of Tm^{166} is positive.

We can infer that the sign of the measured magnetic moment is positive since Q/μ_I is positive. The calculations of Mottelson and Nilsson[13] and observations of large positive values of Q in the rare-earth region imply that Q is positive for Tm^{166} also, and therefore that μ_I is positive.

It is also interesting to note that a relatively large uncertainty in $\mu_{I\,\text{asymptotic}}$ comes from an uncertainty in the value of g_R. We have used the classical value $g_R = Z/A \approx 0.40$. However, g_R can be determined from the measurement of the $M1$ transition rate between different states of a rotational band, if the value of $g_\Omega = (1/\Omega)[g_s\langle S_{z'}\rangle + g_l\langle l_{z'}\rangle]$ is known for the lower state since the $M1$ rate depends on $(g_\Omega - g_R)^2$. Such measurements yield $g_R = 0.2$ for Nd^{150}, Sm^{142}, and Sm^{154}.[17] With the value $g_R = 0.2$, the asymptotic μ_I for the $[411\frac{1}{2}]$ and $[642\frac{5}{2}]$ odd nucleon states is equal to $+0.05$ nm. We are still uncertain as to what value of g_R we should use for Tm^{166}, however, as these neodymium and samarium nuclei show no indications of very large deformations.

TABLE I. Theoretical values of μ_I using asymptotic Nilsson wavefunctions.

Proton orbital	Neutron orbital	I asymptotic
[411 1/2]	[642 5/2]	+0.18 nm
[411 1/2]	[523 5/2]	+2.74 nm
[411 1/2]	[521 3/2]	−1.87 nm
		Measured value: $\lvert\mu_I\rvert = 0.05 \pm 0.03$ nm

ACKNOWLEDGMENTS

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[17] R. P. Sharenberg and G. Goldring, Bull. Am. Phys. Soc. 3, 55 (1958).