Laser spectroscopy of ^{170}Tm

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The magnetic moment, quadrupole moment, and isotope shift of the radioactive isotope ^{170}Tm have been measured by the atomic-beam resonance-fluorescence technique. Hyperfine structures of the 563-, 568-, 590-, and 597-nm transitions were observed with a resolution of 12 MHz. The magnetic moment is found to be $\mu^{(170)}=+0.2458(17)\ \mu_N$; the spectroscopic quadrupole moment, $Q_s^{(170)}=0.72(5)$ b. The isotope shifts $\delta\nu^{169,170}$ for the above lines are $-469(2)$, $-471(6)$, $+462(6)$, and $-461(8)$ MHz, respectively.

I. INTRODUCTION

The nucleus ^{170}Tm ($Z=69$, $N=101$) appears to be unique in that both the odd proton and the odd neutron are in Nilsson orbits having $K_n=K_p=\frac{1}{2}$ for the ground state.[1] This ground state is the lowest member of a $K=1$ band, but is strongly Coriolis coupled to a $K=0$ band. The magnitudes of the magnetic and quadrupole moments of ^{170}Tm (half-life 129 days, spin $I=1$) have been measured by Cabezas and Lindgren[2] with the atomic-beam magnetic-resonance technique. Here more accurate values of these quantities, as well as the signs of the moments, and the isotope shift are measured. The hyperfine structures of the four transitions given in Table I have been measured. All transitions originate from the $4f^{13}6s^2\,{}^2F^o_{7/2}$ Tm I ground state. In three cases the transition is that of an s electron into a p state; in the fourth case an f electron moves into a d state.

II. EXPERIMENTAL METHOD

In the laser-atomic-beam technique used, the laser beam intersected a collimated ^{170}Tm atomic beam at right angles. Resonantly scattered photons were detected by a phototube in a direction perpendicular to both the laser and atomic beams (see Fig. 1).

The beam from an actively stabilized ring dye laser was passed through two sets of three knife-edge collimators inside the entrance and exit of the vacuum chamber. A lens in front of the chamber focused the beam to a diameter of about 2 mm in the interaction region. Photons were collected by a condensing lens system and focused onto a cooled photomultiplier tube. Interference filters were used to reduce background from oven light. To eliminate most of the background from laser-scattered light, the laser beam was chopped on and off by a 200-MHz acousto-optic modulator. Beams were on for periods ranging from 150 to 450 ns with cycle times ranging from 1.5 to 4.5 μs, set depending on the lifetime of the atomic state involved. A gate on a time-to-amplitude converter accepted photon counts during the beam-off periods only. Remaining background counts were generated primarily by phototube dark current.

The ^{170}Tm sample was made by exposing 60 mg of natural ^{169}Tm to a flux of about 10^{14} neutrons/cm^2 s for 30 h, giving a ^{170}Tm sample of about 15 μg (hence a ratio of ^{169}Tm to ^{170}Tm of about 4000), as measured by observing the 84-keV γ rays from the ^{170}Tm decay with a germanium detector. This sample was heated in the tantalum crucible of a resistively heated atomic-beam oven. The collimation ratio of the atomic beam was 1:200. Peak widths (full width half maximum) were typically 12 MHz, dominated by Doppler broadening.

No isotope separation was applied to the sample. The laser beams suffered from weakly interfering laser modes, giving satellite peaks equally spaced from the ^{169}Tm resonances at the one part in 30 000 level. This necessitated actively filtering the laser beam prior to the interaction region. The filter consisted of a scanning etalon locked to the laser frequency by stabilizing the transmitted power. The power of the laser beam in the interaction region during the beam-on period ranged from 0.3 to 1.3 mW.

At each of the laser frequencies, data were acquired from several scans, typically 7 GHz in width and lasting 6 min each, during oven-on times lasting a total of several hours. A temperature-controlled confocal etalon was used to place frequency markers every 150 MHz on a simultaneously acquired spectrum. Also simultaneously acquired were the laser power (used to normalize the spectra) and the power transmitted by the 7-GHz free-spectral-range etalon in the laser wave meter (to check for laser-mode hops). One of the 568-nm scans is shown in Fig. 2. This spectrum is one of seven acquired, and represents 30% of the 568-nm data. In our configuration, the ratio of ^{169}Tm to ^{170}Tm in the atomic beam could not be measured accurately by laser spectroscopy, due to laser-saturation effects and overloading of the electronics during the intense ^{169}Tm fluorescence, but the ratios were within a factor of 3 of those measured by γ rays. The 150-MHz etalon free-spectral range was measured by running a sodium atomic beam and observing the $D1$ line hyperfine structure, both before and after the set of ^{170}Tm measurements. A ^{23}Na splitting of 1960.13 MHz (Ref. 3) between the lowest- and highest-frequency lines was used.

Five out of six ^{170}Tm peaks were observed for the 563- and 568-nm transitions; seven out of seven, for 590 nm; three out of seven for 597 nm. A nonlinear least-squares

TABLE I. Magnetic dipole coupling constants A, electric quadrupole coupling constants B (both for ^{170}Tm upper atomic levels), and isotope shifts $\delta\nu^{169,170}=\nu^{170}-\nu^{169}$ measured in this work. The coupling constants for the common lower level (the ground state, $4f^{13}6s^2\,{}^2F^o_{7/2}$) were determined to be $A=199.0(6)$ MHz and $B=-1006.7(1.8)$ MHz. The angular momentum J and configuration refer to the upper level of the transition. The A value for the 597.1-nm line was inferred, not measured directly.

Line (nm)	J	Configuration	A (MHz)	B (MHz)	$\delta\nu^{169,170}$ (MHz)
563.1	$\frac{5}{2}$	$4f^{13}6s6p$	124.9(1.0)	−616.4(2.8)	−469(2)
567.6	$\frac{9}{2}$	$4f^{13}6s6p$	335.4(6)	−1004.5(3.1)	−471(6)
589.6	$\frac{7}{2}$	$4f^{12}5d_{5/2}6s^2$	261.2(6)	−531.1(2.4)	462(6)
597.1	$\frac{7}{2}$	$4f^{13}6s6p$	[392.0(2.0)]	−799(11)	−461(8)

fit to each scan was performed, where the variables were the peak amplitudes, the isotope shift, the excited-state magnetic dipole constant A (with the one exception noted below), the excited-state electric quadrupole interaction constant B, a peak width, a constant background, and four parameters describing tails from ^{169}Tm peaks. The frequency calibration was obtained by fitting the centroids of the 150-MHz transmission peaks, allowing a quadratic term in the primarily linear dependence of laser frequency on scan time. In several of the 568-nm scans it was possible to vary the A and B of the ground state as well, and thus to determine more accurate values of the constants A and B determined by Cabezas and Lindgren.[2] The present results are $A=+199.0(6)$ MHz and $B=-1006.7(1.8)$ MHz, where the signs have now been determined and the magnitudes are in good agreement with the previous values. The isotope shifts were obtained by comparing the centroids from these fits with the peak positions from fits to the weaker ^{169}Tm lines. (Electronic-saturation effects prevented use of the two stronger ^{169}Tm peaks.) In the case of the 597-nm line, only two of the ^{170}Tm peaks were clearly observable; the third line was barely visible. In this case the value of the upper-state A coupling constant was fixed such that the ratio of upper- to lower-state A values was the same for ^{169}Tm (Refs. 4 and 5) and ^{170}Tm (using the lower-state A as measured in this work). An error of 2 MHz on the 597-nm A value for the upper state was included in assigning the upper-state B value error and the isotope shift error for this transition.

Spectra for ^{169}Tm were acquired at lower oven temperatures and were fit to get A's for the excited states, the ground-state A being fixed at the value determined by Giglberger and Penselin.[4] These A values, having errors of about 1 MHz, agree with those of van Leeuwen *et al.*[5] and Pfeufer,[6] within errors.

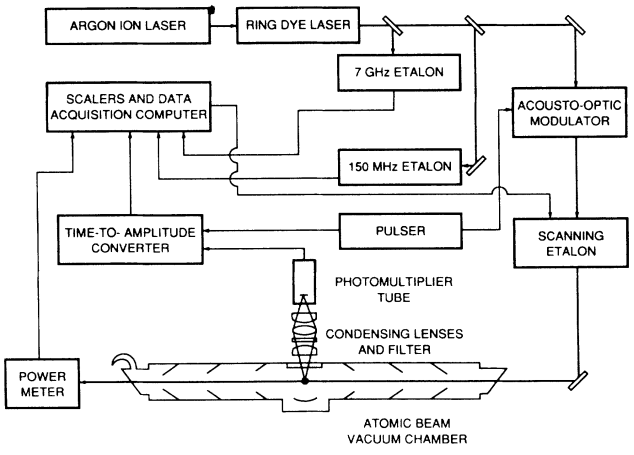

FIG. 1. Schematic diagram of the apparatus.

III. RESULTS AND DISCUSSION

The final isotope shifts and interaction constants for the four excited states are given in Table I. Errors are primarily the standard deviation of the distribution of values from the several scans at each wavelength, but also include propagation of errors in ground-state A and B values, when these were fixed in the fits, and in the calibration of the 150-MHz etalon. As the only stable thulium isotope (^{169}Tm) has nuclear spin $\frac{1}{2}$, and thus zero spectroscopic quadrupole moment, the various B values

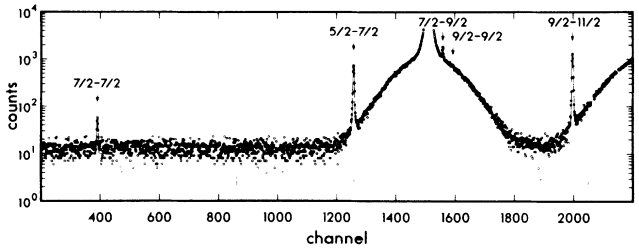

FIG. 2. Hyperfine spectrum measured for the 568-nm transition. The peaks are labeled by the angular momentum F of the ground state followed by that of the excited state. (The $\frac{9}{2}$-$\frac{9}{2}$ transition is observed, but is not visible on this scale. The $\frac{9}{2}$-$\frac{7}{2}$ transition, having an intensity 50 times lower than the most intense peak, was not observed.) The counts, normalized to laser power, are plotted as a function of scan time (120 ms/channel) with frequency increasing to the right. The solid lines show results of a fit to the data. The frequency markers below the data show the centroids of transmission peaks for an etalon of 149.7 MHz free-spectral range. The intense peaks are ^{169}Tm lines. The isotope shift was obtained from the peak position of a less intense ^{169}Tm peak at a frequency above the range of this plot (at channel 2894).

here represent new atomic information for thulium and may be useful in parametrizations of rare-earth hyperfine structure. For the first three transitions (563, 568, and 590 nm), it is found that the ratios of excited-state to ground-state A values for ^{170}Tm are the same as those for ^{169}Tm,[4,5,6] within stated errors. For the case of the 590-nm $4f$-$5d$ transition, there is no change in the number of s electrons, but the isotope shift is still large (though opposite in sign to those of the $6s$-$6p$ transitions). As noted by Pfeufer,[7] this $4f$-$5d$ shift arises from the change in effective screening of the $6s$ electrons by the $4f$ electrons.

The ^{170}Tm nuclear magnetic moment $\mu^{(170)}$ can be obtained from the ^{169}Tm magnetic moment $\mu^{(169)}$ and the A values for the two isotopes with the formula

$$\mu^{(170)}=[I^{(170)}/I^{(169)}][A^{(170)}/A^{(169)}]\mu^{(169)} , \quad (1)$$

where the I's are nuclear spins ($I=1$ for ^{170}Tm and $I=\frac{1}{2}$ for ^{169}Tm). No correction for a hyperfine anomaly is used here. Such a correction is expected to be small for the atomic ground state, as there are no unpaired s electrons. The ^{169}Tm magnetic moment and atomic ground-state A have been measured by Giglberger and Penselin[4] to be $\mu^{(169)}=-0.2310(15)$ μ_N and $A^{(169)}=-374.137\,661(3)$ MHz. Their magnetic moment, which has been diamagnetically corrected, was determined directly by atomic beam magnetic resonance at high magnetic fields and does not depend on electronic properties of the Tm atom. The atomic ground-state value $A^{(170)}=199.0(6)$ MHz of the present work thus gives $\mu^{(170)}=+0.2458(17)$ μ_N, where the error arises primarily from the uncertainty in the ^{169}Tm nuclear magnetic moment. This result is in good agreement with the value $\mu^{(170)}=0.247(5)$ μ_N derived by Ekström and Lamm,[8] using the data of Cabezas and Lindgren,[2] where the sign was derived from the experimental μ/Q ratio and the expected positive value of Q for nuclei in this region of deformation. The present value is also in good agreement with the Nilsson model value 0.23 μ_N of Ekström and Lamm,[8] where the ratios of the nucleon spin-g factors to the free values were taken to be 0.6 in the calculation.

For the quadrupole moment, an estimate may be obtained from the atomic ground state B value, as this ground state has the configuration of a single f electron hole in otherwise closed shells. The traditional method is used, wherein a nonrelativistic value of Q_s is obtained and then Sternheimer corrections are applied. Using the precise values of A and μ for ^{169}Tm,[4] the value $\langle r^{-3}\rangle=11.14(7)\,a_0^{-3}$ (for the $4f$ electron, where a_0 is the Bohr radius) is obtained from the relations given in the Appendix of Lindgren and Rosen.[9] In the absence of a hyperfine anomaly, the same value holds for ^{170}Tm. Nonrelativistically, the $\langle r^{-3}\rangle$ and the measured B for ^{170}Tm result in an uncorrected spectroscopic quadrupole moment of 0.577(4) b. The errors quoted thus far include only errors in the experimental values of A, B, and μ. Recently Pfeufer[7] determined Sternheimer-like quadrupole shielding correction factors R_{4f}^{01} for a series of rare-earth elements. Taking a mean value of $R_{4f}^{01}=0.20(5)$ for the $4f^n6s^2$ correction factor gives $Q_s^{(170)}=0.577/(1-R_{4f}^{01})=0.72(5)$ b. The uncertainty assigned to the shielding correction is responsible for most of the uncertainty in the final result.

For the case of axially symmetric deformation and strong coupling of the odd proton, the relationship between intrinsic and spectroscopic quadrupole moments is given by[10]

$$Q_s=Q_0\frac{3K^2-I(I+1)}{(I+1)(2I+3)} . \quad (2)$$

For ^{170}Tm, $K=I=1$, so $Q_0=7.2(5)$ b is obtained. By comparison, Ekström and Lamm[8] used the data of Cabezas and Lindgren[2] and arrived at $Q_0=6.25(51)$ b, the difference with the current value being predominantly due to the different shielding correction assumed. The current value is somewhat higher (though within errors) than the macroscopic-microscopic model value of 6.8 b calculated by Möller and Nix.[11] In contrast, it is somewhat lower than the value of 7.78(16) b obtained by Fransson *et al.*[12] from $B(E2)$ values measured by Coulomb excitation. Such a difference may arise from uncertainties in the Tm atomic structure, in particular the Sternheimer correction, or from the nuclear-model dependencies invoked in the two determinations of Q_0.

Given Q_0, the deformation β_q (assuming a quadrupoloid with a constant volume) may be found from[13]

$$Q_0=\frac{3}{\sqrt{5\pi}}ZA^{2/3}r_0^2\beta_q\left[1+\frac{2}{7}\sqrt{5/\pi}\beta_q+\frac{1}{14\pi}\beta_q^2\right] . \quad (3)$$

For $Q_0=7.2(5)$ b and $r_0=1.2$ fm, a β_q of 0.282(18) is obtained. [The Fransson value[12] $Q_0=7.78(16)$ b gives $\beta_q=0.303(6)$.] The present value is consistent with ground-state deformations found throughout the deformed rare-earth region.

As there exist no isotope shift measurements derived from K x-ray measurements or from muonic atoms, it is necessary to use empirical atomic factors to obtain $\delta\langle r^2\rangle$ values from the isotope shifts. Here the 568-nm line is considered, as the transition is s^2-sp and the statistical accuracy of the spectrum is highest in this case. The mass shift is taken to be 10(5) MHz.[14] This gives a field shift of $-481(8)$ MHz. To convert this field shift to $\delta\langle r^2\rangle$, the formulation of Blundell *et al.*[15] is used, in which the field shift is divided into electronic and nuclear parts

$$FS=k\Delta N_{\rm tot}\lambda , \quad (4)$$

where

$$k=(2\pi/3)(Ze^2/4\pi\epsilon_0) \quad (5)$$

and

$$\lambda=\sum_{n=2}^{6}S_n\delta\langle r^n\rangle , \quad (6)$$

where the S_n are Seltzer coefficients.[16] The value of S_2 is 1.0; for thulium, $S_4=-9.854\times10^{-4}$ fm^{-2} and $S_6=2.382\times10^{-6}$ fm^{-4}, as tabulated in Ref. 15. The quantity $\Delta N_{\rm tot}$ is the change in total (relativistic) electron probability density at the origin from one level of the transition to another. Since in the present case the prob-

ability density is greater for the lower level, $\Delta N_{\rm tot}$ is negative. It is the product of N, the probability density for a single valence s electron, and $\gamma_{\rm FS}$, a screening factor:

$$\Delta N_{\rm tot}=\gamma_{\rm FS}N \ . \tag{7}$$

The magnitude of $\gamma_{\rm FS}$ is of order unity, but its sign is taken to be negative, to give $\Delta N_{\rm tot}$ negative. The value of N may be estimated from the magnetic dipole coupling constant $a_s^{\rm exp}$ for the $6s$ electron. This constant has been derived by Pfeufer[6] via a parametric analysis of the hyperfine structure of many Tm I lines, with the result $a_s^{\rm exp}=-3865(88)$ MHz for ^{169}Tm. To take into account many-body effects, another factor $\gamma_{\rm HFS}$ is introduced to connect the experimental $a_s^{\rm exp}$ with that for a single valence s electron a_s:

$$a_s^{\rm exp}=\gamma_{\rm HFS}a_s \ . \tag{8}$$

The relation between the single-electron quantities is then

$$N=a_s\eta/(1-\epsilon)\pi K \ , \tag{9}$$

where $\eta=2.807$ is tabulated in Ref. 15, ϵ is the hyperfine anomaly correction factor (expected to be less than a few percent), and K is given by

$$K=(\tfrac{4}{3})\alpha^2R_\infty(m/m_p)g_sg_Ia_0^3 \ . \tag{10}$$

Here g_I is the nuclear g factor, and the other quantities are the usual fine-structure constant, Rydberg constant, ratio of electron to proton mass, electron g factor, and Bohr radius. With $g_I=(I)^{-1}(\mu/\mu_N)$ derived from the ^{169}Tm value above[4] and $\epsilon=0$, it is found that $\lambda=-0.0482\gamma_{\rm HFS}/\gamma_{\rm FS}$. If the values $\gamma_{\rm HFS}=1.0$ and $\gamma_{\rm F}=-0.73$ are taken, in the latter case the magnitude calculated by Coulthard[17] for Eu (justified by the small variation of screening ratios of several of the rare-earth elements[14]), the approximate value $\lambda=0.066$ fm^2 is obtained.

In order to deal with the higher-order terms in the series expansion for λ, calculations assuming a constant-volume quadrupoloid [radius[13] $R=r_0A^{1/3}(1+\alpha_{00}Y_{00}+\beta_qY_{20})$] were performed. This approximation to the nuclear shape, using the deformation $\beta_q=0.282$ derived above, indicates that the higher-order terms decrease the nuclear factor λ by not quite 6%. This implies $\delta\langle r^2\rangle^{169,170}=0.070(14)$ fm^2, where the error in determining the various atomic and nuclear factors in going from the field shift to the change in mean-square radius has been estimated to be 20%.

Considering trends prevalent in neighboring isotope chains, this value of $\delta\langle r^2\rangle^{169,170}$(Tm) may be compared to that estimated from the isotones in Yb ($Z=70$) and Er ($Z=68$). Muonic isotope shift data tabulated by Engfer *et al.*[18] show $\delta\langle r^2\rangle^{170,171}(Yb)=0.067$ fm^2. For Er a direct measurement does not exist, but $\delta\langle r^2\rangle^{168,169}$(Er) may be estimated by taking one-half of $\delta\langle r^2\rangle^{168,170}$(Er) (Ref. 19) to get $\delta\langle r^2\rangle^{168,169}(Er)=0.061$ fm^2. This number should be reduced to account for odd-even staggering, wherein the isotope shift in going from an even-N nucleus to an odd-N nucleus is normally less than one-half the shift between two even-N nuclei. The value $\delta\langle r^2\rangle^{169,170}(Tm)=0.070(14)$ fm^2 from this work is slightly higher but not inconsistent with the mean of the Yb and Er values, since odd-even staggering may be playing a role. Unfortunately no universally accepted formula (or interpretation) exists to reliably predict what the relative staggerings should be.

The droplet model[20] predicts a change in mean-square radius between ^{169}Tm and ^{170}Tm of 0.056 fm^2 for a deformation of $\beta_2=0.282$ (β_2 and β_q are approximately equal). It is in fact possible to exactly reproduce the central value of the $\delta\langle r^2\rangle$ derived here by letting the deformation vary between the two isotopes. Less than a 2% change in the magnitude of β_2 in going from mass 169 to 170 will account for the shift seen here. Caution must be exercised with this interpretation, however, since with the large deformations present in the rare earths the contributions due to β_4 deformations cannot truly be neglected, as they often are for other nuclei. These deformations are known to reach a minimum magnitude in Er but typically have large relative uncertainties,[21] thereby making accurate model predictions impossible. Similarly, $\delta\langle r^2\rangle$ values are insufficient to uniquely derive a value for both β_2 and β_4. (Contrary to the prediction of Okamura and Matsuki,[22] odd-even staggering as a result of changing nuclear deformation cannot be ruled out in Er based solely on the β_2 values. Not only will the droplet model reproduce the staggering seen for ^{167}Er within the limits of error for each β_2 they list, but also small changes in β_4 may likewise be contributing to the observed $\delta\langle r^2\rangle$.)

In preparing this paper, it was learned that Mishin *et al.*[23] have measured the hyperfine structure of the 590- and 597-nm transitions for $^{157-172}$Tm, using resonance photoionization. Their atomic ground-state A and B values are in agreement with those of the present work, though of lower accuracy. Their isotope shift for the 597-nm transition is also in agreement, though again of lower accuracy [$-487(50)$ vs $-461(8)$ MHz]. Their $\delta\langle r^2\rangle$, however, is 30% below the present value, primarily due to a different choice for the atomic wave function at the nucleus. Mishin *et al.* obtained this wave function from an average of two values derived from two semiempirical methods based on (1) the Goudsmit-Fermi-Segrè (GFS) formula and (2) the magnetic hyperfine structure (HFS) constant of the valence s electron. The analysis of the present paper is roughly that of the HFS method. As Mishin *et al.* do not quote the individual values of the two numbers averaged, it is difficult to make a comparison. The GFS method gives a result close to the value of Mishin *et al.*, but it is difficult to see how such a high value can be obtained by the HFS method. The $a_s^{\rm exp}$ value used by Mishin *et al.* was taken from earlier work, which gives a larger wave function than does the value of Pfeufer[6] used in the present work. However, this explains only about a quarter of the difference. No interpretation is offered here to resolve the remaining discrepancy.

IV. CONCLUSIONS

The hyperfine structures for several transitions in the ^{170}Tm atom have been measured by laser spectroscopy.

The nuclear magnetic moment and quadrupole moment (including the signs) were determined; they are in general agreement with theory and with other measurements. From the isotope shift, a value of $\delta\langle r^2\rangle^{169,170}=0.070$ fm^2 is obtained, which is somewhat above the droplet-model result and a small amount above the value estimated from neighboring erbium and ytterbium isotope shifts. Further atomic calculations or measurements are required to reduce the uncertainty (about 20%) in converting between measured isotope shift and $\delta\langle r^2\rangle$.

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