

elements in impure and compensated semiconductors. For example, a calculation of these quantities even for a one-dimensional model would be very helpful. It would also be useful to have a quantitative description of the density of states near the band edge for the difficult range of impurity concentrations in which the minimum in the density of states between the impurity level and the adjacent band disappears.

In spite of the many shortcomings of the model used in our calculations, we conclude that it gives a reasonable account of the stimulated emission of radiation in impure GaAs for the range of impurity and carrier concentrations for which the model can be expected to apply. For small-gap semiconductors, such as InAs

and InSb, the model may not be applicable directly, since small errors in the calculated position of the lasing peak lead to large errors in the factor E^{-2} of Eq. (4), an effect which is of minor importance in a semiconductor with a larger energy gap.

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Paramagnetic-Resonance Absorption in the Optically Populated State ${}^2F_{5/2}$, $E_{5/2}$ of Tm^{2+} in CaF_2

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Paramagnetic-resonance absorption was observed in the metastable state ${}^2F_{5/2}$, $E_{5/2}$ of $\text{CaF}_2:\text{Tm}^{2+}$ which was populated by an optical source. The measured g value of 1.453 ± 0.002 can be compared with the calculated value of $(-)$ 1.478. The g discrepancy of 0.025 is the same as found for the ground state. From the hyperfine constant for two values of J , a core-polarization correction of $(+)$ 14 ± 3 Mc/sec is derived for the ground state.

INTRODUCTION

PARAMAGNETIC-resonance absorption has been extensively used to analyze the structure of the ground state of paramagnetic ions and the interaction of these ions with their diamagnetic neighbors. Currently, this method is being exploited to examine the properties of excited states which can be populated by an optical source. Paramagnetic resonance absorption in a metastable state populated by optical pumping has already been reported for Dy^{2+} in CaF_2 .¹ We wish to report on the paramagnetic-resonance spectrum of an excited state of Tm^{2+} in CaF_2 , the emitting state of the laser, populated by incoherent unpolarized light. The optical spectrum of $\text{CaF}_2:\text{Tm}^{2+}$ has been observed by Kiss² and it was also demonstrated that this system could be operated as a laser³ at a wave number of about 9000 cm^{-1} . The ground-state paramagnetic-resonance spectrum and the ground-state nuclear-double-resonance spectrum have been investigated by Hayes and Twidell,⁴ and Bessent and Hayes,⁵ respectively. It has

also been shown⁶ that the broad $4f^{n-1}5d$ absorption bands show circular dichroism and that the ground-state Zeeman levels can be selectively pumped using polarized radiation. The ground configuration of Tm^{2+} consists of only a single term, 2F , and therefore any breakdown of L - S coupling due to the crystalline field can be readily taken into account. Because of the simplicity of the Tm^{2+} system, the combination of paramagnetic-resonance data for both excited and ground states yields information about covalent bonding and core polarization. It is felt that investigations of the divalent thulium ion hold the most promise for reaching some conclusions about covalent bonding and core polarization in rare-earth ions since it is theoretically the most straightforward system and a considerable amount of data can be obtained on both of the J states in the ground configuration.

OPTICAL SPECTRUM OF $\text{Tm}^{2+}:\text{CaF}_2$

The divalent thulium ion (isoelectronic with Yb^{3+}) has 13 electrons in the incomplete $4f$ shell. The spin-orbit coupling $\zeta\mathbf{L}\cdot\mathbf{S}$ splits the energy levels into two spin-orbit states ${}^2F_{5/2}$ and ${}^2F_{7/2}$, spaced by $\frac{7}{2}\zeta$ with the latter lower. An ideal cubic field further splits these two

¹ E. S. Sabisky and C. H. Anderson, Phys. Rev. Letters **13**, 754 (1964).

² Z. J. Kiss, Phys. Rev. **127**, 718 (1962).

³ Z. J. Kiss and R. C. Duncan, Jr., Proc. IEEE **50**, 1531 (1962).

⁴ W. Hayes and J. W. Twidell, J. Chem. Phys. **35**, 1521 (1961).

⁵ R. G. Bessent and W. Hayes, Proc. Roy. Soc. (London) **A285**, 430 (1965).

⁶ C. H. Anderson, H. A. Weakliem, and E. S. Sabisky, Phys. Rev. **143**, 223 (1966).

spin-orbit states as shown in Fig. 1. The measured values are those of Kiss.² The relative positions of the crystal-field split states are functions of two parameters, B_4 and B_6 . These two parameters are defined by the cubic potential given by

$$V_c = B_4(4\pi/9)^{1/2} \{ Y_4^0 + (5/14)^{1/2} (Y_4^4 + Y_4^{-4}) \} + B_6(4\pi/13)^{1/2} \{ Y_6^0 - (\frac{7}{2})^{1/2} (Y_6^4 + Y_6^{-4}) \},$$

where the Y_L^M are the normalized spherical harmonics. The coefficients B_4 and B_6 are related to those defined by Pappalardo and used by Kiss² by the relations $B_4(4\pi/9)^{1/2} = a\lambda = (4\pi/9)^{1/2}(7 \times 11)\lambda$ and

$$B_6 \left(\frac{4\pi}{13} \right)^{1/2} = b\mu = - \left(\frac{4\pi}{13} \right)^{1/2} \left(\frac{3 \times 6 \times 11 \times 13}{5} \right) \mu.$$

The relationship with the coefficients used by Bleaney⁷ are $b_4 = 7\lambda/3$, $b_6 = 6\mu$. For consistency, the parameters λ and μ will be used in this paper.

Second-order perturbation theory is used to take account of crystal-field mixing of the two $LSJM$ states. The calculations give the energy levels and eigenfunctions shown in Table I. In agreement with Bleaney, the observed optical spectrum can be fitted by taking $\zeta = 2513 \text{ cm}^{-1}$, $\lambda = 19.63 \text{ cm}^{-1}$, $\mu = 0.84 \text{ cm}^{-1}$. In addition $\delta W_1 = 21.3 \text{ cm}^{-1}$, $\delta W_2 = 0.18 \text{ cm}^{-1}$, $\alpha_a = 0.9988$, $\alpha_b = 0.0486$, $\alpha_c = 1.000$, $\alpha_d = 0.0045$.

PARAMAGNETIC-RESONANCE MEASUREMENTS

Paramagnetic-resonance absorption was observed in the metastable state (${}^2F_{5/2}$, $E_{5/2}$) of $\text{CaF}_2:\text{Tm}^{2+}$ populated by optical pumping. The resonance could be observed using scope presentation. The measurements were done using a superheterodyne system with phase-sensitive detection. The size of the samples were about

TABLE I. Eigenfunctions in terms of $|J, \Gamma, \mu\rangle$ and eigenvalues of the crystal-field states of Tm^{2+} in CaF_2 ; second-order crystal-field admixtures have been included.^a

Energy level	Calculated energy	Eigenfunction
$W_1; 7/2, E'_{5/2}\rangle$	$-18\lambda - 72\mu - \delta W_1$	$\alpha_a 7/2, E_{5/2}\rangle + \alpha_b 5/2, E_{5/2}\rangle$
$W_2; 7/2, G'\rangle$	$2\lambda + 96\mu - \delta W_2$	$\alpha_c 7/2, G\rangle - \alpha_d 5/2, G\rangle$
$W_3; 7/2, E_{1/2}\rangle$	$14\lambda - 120\mu$	$ 7/2, E_{1/2}\rangle$
$W_4; 5/2, E'_{5/2}\rangle$	$(7/2)\zeta - (44/3)\lambda + \delta W_1$	$\alpha_a 5/2, E_{5/2}\rangle - \alpha_b 7/2, E_{5/2}\rangle$
$W_5; 5/2, G'\rangle$	$(7/2)\zeta + (22/3)\lambda + \delta W_2$	$\alpha_c 5/2, G\rangle + \alpha_d 7/2, G\rangle$

^a Where: $\delta W_1 = \frac{\alpha_1^2}{W_4 - W_1}$; $\delta W_2 = \frac{\alpha_2^2}{W_5 - W_2}$; $\alpha_1 = -\frac{4}{\sqrt{3}}[5\lambda + 108\mu]$;

$$\alpha_2 = 4(\sqrt{5})[18\mu]; \quad \alpha^1 = \frac{\alpha_1}{W_1 - W_4}; \quad \alpha^2 = \frac{\alpha_2}{W_2 - W_5};$$

$$\alpha_a = \frac{1}{(1 + (\alpha^1)^2)^{1/2}}; \quad \alpha_b = \frac{\alpha^1}{(1 + (\alpha^1)^2)^{1/2}};$$

where the eigenfunctions in terms of $|J, M\rangle$ are:

$$\left\{ \begin{array}{l} |7/2, E_{5/2}\rangle = 1/2[\lambda(\sqrt{3}) \pm 5/2] - |\mp 3/2\rangle \\ |7/2, G\rangle = \left\{ \begin{array}{l} (7/12)^{1/2} [\pm 7/2] - (5/7)^{1/2} [\mp 1/2] \\ 1/2 [\pm 5/2] + (\sqrt{3}) [\mp 3/2] \end{array} \right\} \\ |7/2, E_{1/2}\rangle = (5/12)^{1/2} [\pm 7/2] + (7/5)^{1/2} [\mp 1/2] \end{array} \right\}$$

$$\left\{ \begin{array}{l} |5/2, E_{5/2}\rangle = (1/6)^{1/2} [\pm 5/2] - (\sqrt{5}) [\mp 3/2] \\ |5/2, G\rangle = \left\{ \begin{array}{l} (1/6)^{1/2} [(\sqrt{5}) [\pm 5/2] + | \mp 3/2] \\ | \mp 1/2 \rangle \end{array} \right\} \end{array} \right\}.$$

⁷ B. Bleaney, Proc. Roy. Soc. (London) A277, 289 (1964).

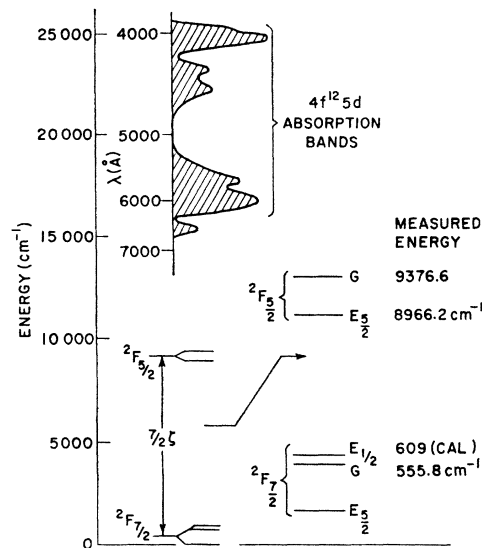


FIG. 1. Energy levels of $\text{CaF}_2:\text{Tm}^{2+}$.

0.02 cc. A number of crystals were used having a total thulium concentration varying from 0.01 to 0.2%. Approximately 50% or more of the thulium ions were reduced to the divalent state by calcium vapor baking. The samples were placed against the side wall of an X-band cavity operating in the TE_{101} mode. The cavity contained many slots for optical illumination. The pump lamp was either a 500-W dc mercury lamp or a 2500-W ac mercury lamp which was focussed on the crystal by a simple lens system. Radiation below 10000 cm^{-1} was filtered out. The temperature of the crystal was estimated to rise less than 0.2°K with application of the light. The excited-state resonance lines were observed from 1.5 to 4.2°K .

With the application of light, two resonance lines appear near 4700 G separated by about 570 G at X-band frequencies. From their g values, which are in agreement with optical measurements,⁸ these lines were attributed to the two allowed hyperfine transitions for the stable isotope ${}^{169}\text{Tm}$ ($I = \frac{5}{2}$) in the $E_{5/2}$ state of the $J = \frac{5}{2}$ spin-orbit state. These resonance lines could be made both absorptive, emissive, or one absorptive and the other emissive, depending on the optical pump rate and thulium concentration. This interesting phenomenon will be discussed in a future paper. The linewidth for a nominal thulium concentration of 0.1% was 13 G or about the same width as the ground-state resonance lines. The fluorine superhyperfine structure in samples containing about 0.01% thulium is easily resolved on the ground-state resonance lines but was not observed on the excited-state lines. This was probably due to the use of unoriented samples, and in the majority of cases, heavy doped samples for the excited-state measurements. From preliminary measurements of the pump down time on the ground-state resonance lines, the

⁸ Z. J. Kiss, Appl. Phys. Letters 2, 61 (1963); H. A. Weakliem and Z. J. Kiss, J. Chem. Phys. 41, 1507 (1964).

effective optical pump rates could be obtained using the measured spin-lattice relaxation rate of the ground state. Typical total pump rates of 8 sec^{-1} were obtained. The spin-lattice relaxation time of the ground state was a function of concentration and varied from 500 msec to 1.7 sec at 1.5°K and X-band frequencies. From these measurements, it was estimated that a few percent of the ions were being optically pumped into the excited state.

Preliminary comparative measurements of the spin relaxation time at 1.5°K at X-band frequencies by the cw saturation method between the ground and excited states were used to derive a relaxation time of about 10 msec for the Zeeman levels of the excited state. This value can be compared with the measured⁹ optical lifetime for this state of 6 msec. This implies that the actual population distribution of the Zeeman levels is primarily due to the optical lifetime while the spin-lattice relaxation time is comparatively much longer. Thus, thermalization between the Zeeman levels of the excited state is not reached and the spin temperature is considerably above the bath temperature. This in agreement with the observation of relatively weak paramagnetic-resonance signals even though the excited state in some experiments contained the order of 10^{16} spins. Using the spin-Hamiltonian formalism of Hayes and Twidell,⁴ the experimental results may be fitted using $|g| = 1.453 \pm 0.002$ and $|A| = (387 \pm 2) \times 10^{-4} \text{ cm}^{-1}$. The weak transition due to a simultaneous change of the spin and nuclear quantum numbers was not observed and prevented a determination of the pseudonuclear g factor. The experimental values⁹ for the ground state are $|g| = 3.452 \pm 0.003$ and $|A| = 1101.376 \pm 0.004 \text{ Mc/sec}$.

DISCUSSION

A. g Value

To compare the experimental and calculated g values the eigenfunctions which include second-order crystal-field admixtures given in Table I will be used. Application of the theory shows that for the excited state using the wave function $|\frac{5}{2}, E_{5/2}'\rangle = \alpha_a |\frac{5}{2}, E_{5/2}\rangle - \alpha_b |\frac{7}{2}, E_{5/2}\rangle$ the g value becomes

$$g_{5/2} = - (10/6)\Lambda_{5/2}\alpha_a^2 - (16/7\sqrt{3})\alpha_a\alpha_b + 3\Lambda_{7/2}\alpha_b^2$$

while for the ground state by interchanging α_a and α_b and changing the sign of the cross term

$$g_{7/2} = 3\Lambda_{7/2}\alpha_a^2 + (16/7\sqrt{3})\alpha_a\alpha_b - (10/6)\Lambda_{5/2}\alpha_b^2,$$

where $\Lambda_{5/2}$ and $\Lambda_{7/2}$ are the Lande g values for the $J = \frac{5}{2}$, $J = \frac{7}{2}$ states and α_a , α_b are positive values reflecting the amount of crystal-field admixtures between the two J states. From fitting the optical data, $\alpha_a = 0.9988$ and $\alpha_b = 0.0486$. It is seen that the excited state, $J = \frac{5}{2}$, has a negative g value while the ground-state g value is positive. The calculated values using $\Lambda_{7/2} = 1.1412$ and $\Lambda_{5/2} = 0.8548$ from Judd¹⁰ are $g_{5/2} = -1.478$ and $g_{7/2}$

$= +3.477$. The difference between theory and experiment for the $J = \frac{5}{2}$ state is $\Delta g_{5/2} = -0.025$ while a $\Delta g_{7/2} = +0.025$ is obtained using the published experimental g value for the ground state. Thus within the experimental accuracy, the magnitude of the g discrepancy is the same for both J states.

It is common practice to relate the g difference to the presence of a small degree of covalent bonding. If this is done by replacing the Zeeman orbital operator by kL , where k is the orbital reduction factor, the g value for the $J = \frac{5}{2}$ state becomes

$$g_{5/2} = - (10/6)[\Lambda_{5/2} + (k-1)(2-\Lambda_{5/2})]\alpha_a^2 - (16/7\sqrt{3}) \times (2-k)\alpha_a\alpha_b + 3[\Lambda_{7/2} + (k-1)(2-\Lambda_{7/2})]\alpha_b^2.$$

From this we find that $(1-k) = 0.014$ and 0.009 ± 0.001 for the $J = \frac{5}{2}$ and $\frac{7}{2}$ states, respectively. Thus the orbital factor is smaller or the amount of covalent bonding is larger for the excited state than for the ground state. Calculations have also shown that zero-point vibrational admixtures of excited crystal-field states may cause a small g shift. The calculated g shift¹¹ for the $J = \frac{7}{2}$ state of -0.01 is smaller than the observed difference of -0.025 but is of the correct sign and magnitude. To first order for this effect, one might expect the g shift of the excited state to be approximately the same as the ground state since

$$|\Delta g_{5/2}| \approx |\Delta g_{7/2}| \frac{\Lambda_{5/2} [W_{2,3} - W_1]}{\Lambda_{7/2} [W_5 - W_4]} \approx |\Delta g_{7/2}|.$$

Experimentally it has been found that the g discrepancy is the same for the two J states. Thus, covalent bonding and vibrational admixtures of excited crystal-field states are reasonable explanations of the g discrepancy. Further theoretical and experimental work on the Tm^{2+} ion in CaF_2 , SrF_2 , BaF_2 , and SrCl_2 hold the most promise for evaluating the magnitude of these two effects for the rare-earth ions.

B. Magnetic Hyperfine Constant

The interaction of the paramagnetic electrons with the nuclear spin can be expressed as

$$\mathcal{H}_{\text{hfs}} = p\mathbf{N} \cdot \mathbf{I} + \mathcal{K}\mathbf{S} \cdot \mathbf{I},$$

where

$$p = 2\beta\beta_N \frac{\mu_N}{I} \left\langle \frac{1}{r^3} \right\rangle, \quad \mathbf{N} = \sum_i \left\{ \mathbf{1}_i - \mathbf{s}_i + \frac{3(\mathbf{r}_i \cdot \mathbf{s}_i)\mathbf{r}_i}{r_i^2} \right\}$$

and \mathcal{K} is a constant representing the admixture of configurations with s electrons or which represents the effect of core polarization. $\langle r^{-3} \rangle$ denotes the average of $1/r^3$ for r an effective radius of the $4f$ shell. If the above Hamiltonian is assumed, measurements of the hyperfine constant for two different J states allows one to deduce the two unknown parameters, p and \mathcal{K} . $\langle 1/r^3 \rangle$ is taken to be a constant. From p , the value of $\langle r^{-3} \rangle$ can then be obtained provided μ_N is accurately known while \mathcal{K} gives the effect of core polarization.

⁹ G. J. Goldsmith (private communication, 1965).

¹⁰ B. R. Judd and I. Lindgren, Phys. Rev. **122**, 1802 (1961).

¹¹ M. Inoue, Phys. Rev. Letters **11**, 196 (1963).

TABLE II. Hyperfine constants for Tm^{2+} in CaF_2 for the ${}^2F_{5/2}$, $E_{5/2}$ and ${}^2F_{7/2}$, $E_{7/2}$ states.^a

	A (Mc/sec) Experimental	a (Mc/sec) Uncorrected	Corrected for second-order crystal-field admixture		Crystal-field and core-polarization corrections	
			a^1 (Mc/sec)	p^1 (Mc/sec)	a_f (Mc/sec)	a_c (Mc/sec)
Ground state $J = \frac{7}{2}$	(-) 1101.376 ± 0.004	(-) 367.12 ± 0.001	(-) 374.15	(-) 491.07 ± 0.001	(-) 389 ± 1.5	(+) 14 ± 3
Excited state $J = \frac{5}{2}$	(+) 1160 ± 5	(-) 696 ± 3	(-) 709	(-) 517 ± 2	(-) 700 ± 3	(-) 9 ± 1.5

^a Where: $a_f = p(J\|N\|J)$; $a_c = a - a_f(0.9812)$; $k = (+)87 \pm 13$ Mc/sec; $p = (-)510 \pm 2$ Mc/sec.

The measured magnetic hyperfine constant A is related to p and \mathcal{K} by the equation for the $J = \frac{7}{2}$ ground state.

$$\frac{A}{2\langle \frac{7}{2}, + | J_z | \frac{7}{2}, + \rangle} = p \langle \frac{7}{2} \| N \| \frac{7}{2} \rangle \left[\alpha_a^2 - \frac{\sqrt{3}}{6} \alpha_a \alpha_b + \alpha_b^2 \right] + \mathcal{K} \left[\frac{1}{7} \alpha_a^2 + \frac{16}{21\sqrt{3}} \alpha_a \alpha_b + \frac{5}{63} \alpha_b^2 \right]$$

and similarly for the $J = \frac{5}{2}$ state

$$\frac{A}{2\langle \frac{5}{2}, + | J_z | \frac{5}{2}, + \rangle} = p \langle \frac{5}{2} \| N \| \frac{5}{2} \rangle \left[\alpha_a^2 - \frac{\sqrt{3}}{6} \alpha_a \alpha_b - \alpha_b^2 \right] + \mathcal{K} \left[-\frac{1}{7} \alpha_a^2 + \frac{48}{35\sqrt{3}} \alpha_a \alpha_b - \frac{9}{35} \alpha_b^2 \right].$$

In the usual manner

$$\frac{A}{2\langle J, + | J_z | J, + \rangle} = a,$$

where a is the hyperfine constant when the hyperfine interaction is written as $a\mathbf{I} \cdot \mathbf{J}$. Using the above equations and experimental results, one can obtain the hyperfine constant a_f for the $4f$ electrons for the thulium ion and these are given in Table II. From Elliott and Stevens,¹²

$$\langle \frac{7}{2} \| N \| \frac{7}{2} \rangle = 2^4/21 \quad \text{and} \quad \langle \frac{5}{2} \| N \| \frac{5}{2} \rangle = (3 \times 2^4)/21.$$

In Table II, the values for a' and p' are those which only include corrections due to crystal-field admixtures. The parameter p' is seen to be different for the two J states and the difference is probably outside any expected changes in $\langle 1/r^3 \rangle$. By taking into account the effect of core polarization in the manner described by the previous two equations, the value of the two constants are found to be $p = (-)510 \pm 2$ Mc/sec and $\mathcal{K} = (+)87 \pm 13$ Mc/sec. The large error in \mathcal{K} reflecting the relatively large error in A for the $J = \frac{5}{2}$ state. Using the above two constants, the derived hyperfine constant a_f is (-389 ± 1.5) Mc/sec for the ground state and this can be compared to the value of (-374.14) Mc/sec for the thulium atom measured by Ritter¹³ and the value of (-382.4) Mc/sec for Tm^{2+} in CaF_2 obtained by Bleaney.⁷ The main difference between the result found in this paper and that of Bleaney arises because of the different values used for the core-polarization

¹² R. J. Elliott and K. W. H. Stevens, Proc. Roy. Soc. (London) **A218**, 553 (1953).

¹³ G. J. Ritter, Phys. Rev. **128**, 2238 (1962).

correction. The value obtained in this paper is $(+14 \pm 3)$ Mc/sec while Bleaney's estimate is $(+4.7 \pm 0.8)$ Mc/sec or a ratio of about 3. The large difference cast doubt on the present method which was used by Bleaney for estimating the core polarization for rare-earth ions. The present method is based on the assumption that core polarization produces practically the same magnetic field (per unit of spin) at the nucleus throughout the rare-earth group and, thereby, the value for Tm^{2+} can be deduced from the experimental results of S-state ions. The values obtained in this paper must also be used with care since no independent check on the constants was possible. In addition, corrections for covalent bonding and orbit-lattice interaction were not taken into account and must await further developments in these areas. These corrections would tend to shift p and \mathcal{K} to slightly higher values. The signs of the constants were obtained in the following manner: The sign of μ_N is negative from atomic-beam magnetic resonance, which implies a_f is negative. From the definition of A , the sign of A is negative for the $J = \frac{7}{2}$ state and positive for the $J = \frac{5}{2}$ state since the g values are positive and negative, respectively.

The a_f value of Tm^{2+} is 4% greater than the value measured by Ritter for TmI . Using Ritter's¹³ value for $\mu_n = -0.229 \pm 0.003$ n.m, the value of $\langle r^{-3} \rangle$ for the $4f$ electrons for Tm^{2+} is $11.7/a_0^3$, where a_0 is the Bohr radius. It is interesting to compare the experimental value of $\langle r^{-3} \rangle$ for Tm^{2+} to the calculated values of Lindgren¹⁴ who finds $\langle r^{-3} \rangle = 10.73$ and 11.20 atomic units for Tm I and Tm IV , respectively. Thus the calculated ratio in $\langle r^{-3} \rangle$ in going from atom to trivalent ion is 4.7% while the experimental ratio in $\langle r^{-3} \rangle$ is going from atom to divalent ion is 4%. With available information, it is too early to draw conclusions about the magnitude of the effect on the hyperfine interaction due to a removal of the two $6s$ electrons or a removal of a single $4f$ electron on the hyperfine interaction. The neutral Tm atom has the same configuration as Tm^{2+} differing only in having two extra electrons which are paired off in the $6s$ shell.

ACKNOWLEDGMENTS

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¹⁴ T. Lindgren, Nucl. Phys. **32**, 151 (1962).