Atomic Beam Study of the Hyperfine Structure of Thulium-170*

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(Received June 3, 1960)

The atomic-beam magnetic-resonance technique has been used to measure atomic and nuclear quantities of the 129-day isotope Tm^{170} as follows: $J = 7/2$, $g_J = 1.14122 \pm 0.00015$, $I = 1$, $|A| = 200 \pm 3$ Mc/sec, and $|B| = 1010 \pm 15$ Mc/sec. The values of J and g_J are consistent with the ground-state assignment $^2F_{7/2}$. Values of the nuclear moments are calculated from the hyperfine-structure interaction constants A and B by use of a two-parameter radial wave function, in which one parameter is determined from comparison with Hartree functions and the other parameters from the experimental spin-orbit coupling constant. Uncorrected values are obtained, as follows: $|\mu_I| = 0.26 \pm 0.02$ nm and $|Q| = 0.61 \pm 0.05$ barn, with the two moments of the same sign. The same wave function is used to calculate the relativistic and diamagnetic corrections to the atomic g value, and the result is in excellent agreement with the experiment.

INTRODUCTION

THE work reported here is a part of a more general program for investigations of radioactive isotopes in the rare-earth region (lanthanides) by the method of atomic beams. This technique involves hyperfine-structure (hfs) measurements and therefore gives information about the nucleus as well as the electronic structure. For the isotope reported here the nuclear spin (I), the magnetic dipole and electric quadrupole interaction constants (A, B), the total electronic angular momentum (J), and the atomic g value (g_J) have been measured.[1] From the hfs interaction constants approximate values of the nuclear moments have been calculated by use of an improved radial wave function.

This isotope has also been investigated by beta spectroscopists,[2] and their spin assignment is in agreement with ours.

The electronic ground state of thulium has been determined by optical methods,[3] and is (with spectroscopic notations) $4f^{13}6s^2$, $^2F_{7/2}$, which is consistent with our results. The atomic g value, however, has not been accurately measured before, and is found to differ significantly from the classical Landé value. Since the state above is essentially a single-electron state the admixture of other states is very small. However, for a heavy atom like thulium the relativistic and diamagnetic effects become quite important. It is shown that when these effects are taken into account, as well as the anomalous moment of the electron, excellent agreement with the experimental g value is obtained.

EXPERIMENTAL METHOD AND RESULTS

The method employed in this experiment is the conventional atomic-beam flop-in technique, which has been described in detail in several articles.[4] The principles are briefly the following. A beam of free atoms is allowed to pass through a homogeneous magnetic field, and transitions are observed between different hfs levels. Transitions within the same F state are followed up from the linear Zeeman region, where they, in principle, give the nuclear spin, into intermediate fields, where information about the hfs is obtained. More accurate determinations of the hfs separations can then be made by observations of direct transitions between different F states.

The 129-day isotope Tm^{170} was produced by irradiation of thulium metal in a neutron flux of 2×10^{13} n/cm^2 sec for a few weeks in the pool-type reactor at the Lawrence Radiation Laboratory, Livermore. The metal piece was then put directly into the tantalum oven of the atomic beam apparatus[5] and heated up to about 600 to 800°C. A few hundred milligrams was bombarded each time, and this gave a stable beam for several days. The beam was collected on clean platinum foils, which were subsequently counted in a continuous-flow proportional counter.

The Hamiltonian for a free atom in an external magnetic field H can be written[6]

$$\mathcal{H} = hA\mathbf{I}\cdot\mathbf{J} + hBQ_{op} - g_J\mu_0\mathbf{J}\cdot\mathbf{H} - g_I\mu_0\mathbf{I}\cdot\mathbf{H}, \quad (1)$$

where

$$Q_{op} = \frac{3(\mathbf{I}\cdot\mathbf{J})^2 + \frac{3}{2}(\mathbf{I}\cdot\mathbf{J}) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}.$$

Octupole and higher order interactions are here omitted. The hfs energy levels are shown schematically in Fig. 1 for Tm^{170} ($I = 1$, $J = 7/2$), in which case there are three $\Delta F = 0$ transitions (α, β, γ) and two $\Delta F = 1$ transitions (δ, ϵ) observable with a flop-in arrangement.

[1] I. Lindgren, A. Cabezas, and W. A. Nierenberg, Bull. Am. Phys. Soc. **5**, 273 (1960).

[2] R. L. Graham, J. L. Wolfson, and R. E. Bell, Can. J. Phys. **30**, 459 (1952).

[3] W. F. Meggers, Revs. Modern Phys. **14**, 96 (1942).

[4] J. R. Zacharias, Phys. Rev. **61**, 270 (1942); L. Davis, D. E. Nagle, and J. R. Zacharias, Phys. Rev. **76**, 1068 (1949).

[5] G. O. Brink, J. C. Hubbs, W. A. Nierenberg, and J. L. Worcester, Phys. Rev. **107**, 189 (1957).

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

The energy levels at zero field are, from Eq. (1),

$$W_{9/2} = (7/2)hA + (1/4)hB,$$

$$W_{7/2} = -hA - (5/7)hB,$$

$$W_{5/2} = -(9/2)hA + (15/28)hB.$$

The relative positions of these levels are shown in Fig. 2 as a function of the ratio B/A. The level order is normal in the region $-4.667 < B/A < 2.8$. The experimental ratio is -5.05, which means that the $F = 9/2$ and $F = 7/2$ levels are inverted. Since this ratio is very close to one of the critical values, the two hfs separations become very different in magnitude (73 and 1960 Mc/sec, respectively) and this gives the three $\Delta F = 0$ transitions quite different behavior as the magnetic field increases. Figure 3 shows the frequency divided by $\mu_0 H/h$ for these transitions. At low fields the frequencies are approximately given by

$$\nu \cong g_F \mu_0 H/h,$$

where

$$g_F \cong g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)}.$$

Therefore, in this diagram the curves start at the g_F values and have a slope at the beginning corresponding to the second-order term. The alpha transition has no quadratic term and actually starts with a zero slope. Since the separation between $F = 9/2$ and $F = 7/2$ ($\Delta \nu_1$) is so small, however, the higher-order terms become significant at a relatively low field. Also, for the beta transition the higher-order terms very soon
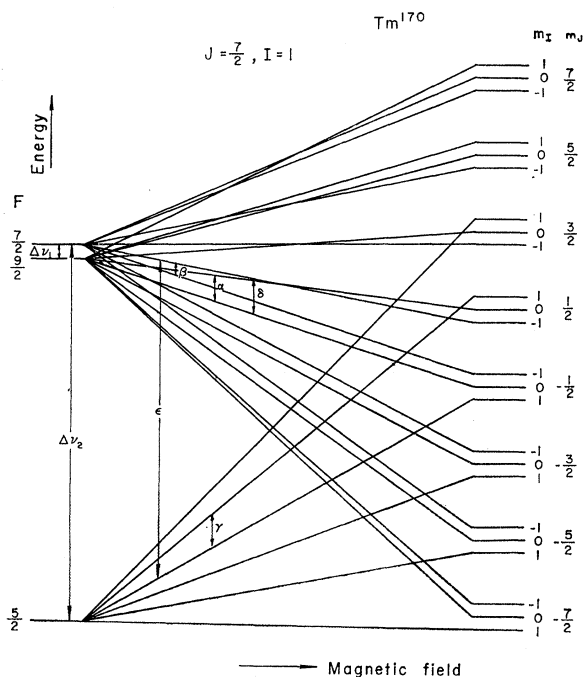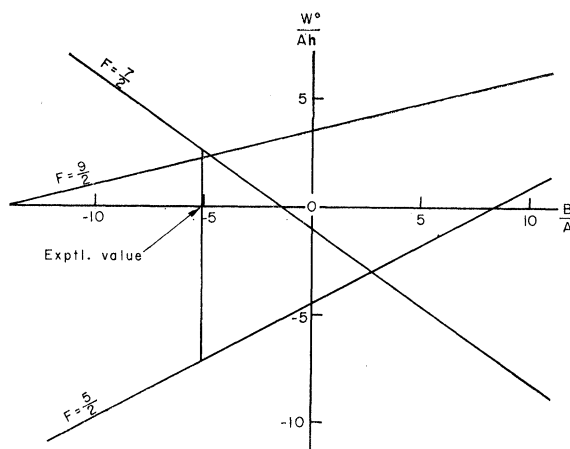

FIG. 2. Relative positions of the energy levels at zero field *versus* the B/A ratio.

become predominant, and the total shift from the linearity turns negative. The gamma transition, on the other hand, is independent of $\Delta \nu_1$ in the first approximation and consequently has a much smaller relative shift. The points in Fig. 3 correspond to the experimental resonance frequencies, and the curves show the corresponding values calculated for the best fit of the three parameters A, B, and g_J.

The three $\Delta F = 0$ transitions have been followed up to about 300 gauss, and one of the $\Delta F = 1$ transitions, δ, has been observed at two low fields. The other $\Delta F = 1$ transition, ϵ, occurs at an inconveniently high frequency (approx 1960 Mc/sec) and has not been looked for. The resonance curves for each of the $\Delta F = 0$ transitions at the highest field are shown in Fig. 4 together with one curve for the $\Delta F = 1$ transition. The latter transition is of σ type ($\Delta m = 0$) and the resonance curve is therefore double-peaked. The resonance frequency corresponds to the center of the dip. In general, the uncertainty in the resonance frequency has been taken to be about $\pm 1/4$ of the half-width of the resonance curve.

The experimental data have been analyzed on the IBM 704 computer, with a program described elsewhere.[7] A least-squares fit is made of the three parameters (A, B, and g_J) and also a correction for the small g_I term which appears in Eq. (1). The sign of the nuclear moment, however, has to be chosen in advance and can be determined only from comparison between the fits with opposite sign assumptions.

Table I shows the data processed by the computer. This gives the resonance frequencies for the radioactive isotope and the corresponding magnetic field. The differences between the experimental and calculated frequencies are also included. A positive sign of the nuclear moment gives a smaller χ^2 but the difference is

FIG. 1. Schematic hfs energy-level diagram for Tm^{170}.

[7] R. Marrus, W. A. Nierenberg, and J. Winocur, University of California Radiation Laboratory Report UCRL-9207 (to be published).
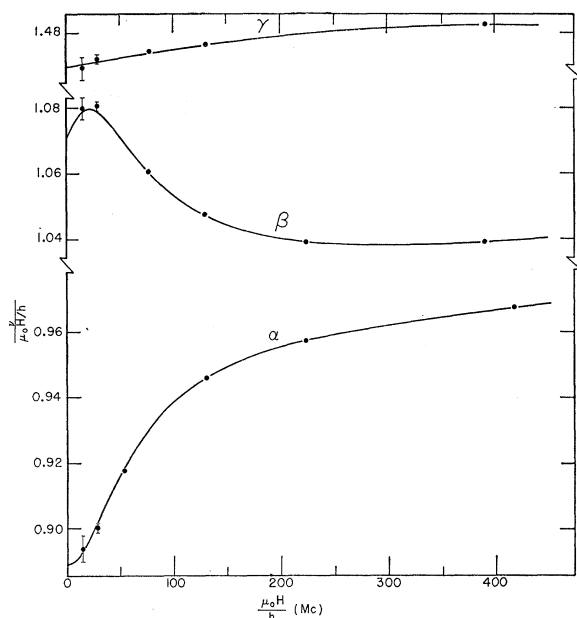
FIG. 3. Resonance frequencies divided by $\mu_0 H/h$ for the $\Delta F=0$ transitions *versus* the magnetic field.

not significant. This means that the moment is too small to allow a definite sign determination from this experiment. However, the relative sign of the dipole and quadrupole moments can be uniquely determined from the sign of the B/A ratio.

The final results are

$J=7/2$, $g_J=1.14122\pm0.00015$,

$I=1$, $|A|=200\pm3$ Mc/sec,

$\qquad\qquad |B|=1010\pm15$ Mc/sec, $(B/A<0)$.

We have here stated larger errors than obtained from the computer in order to include possible systematic errors.

CALCULATION OF THE NUCLEAR MOMENTS

Since the electronic configuration of thulium consists of completely filled shells minus one electron, the relations between the hfs interaction constants and the nuclear moments are given by[8]

$$A=c\ \mathrm{Ry}\alpha^2 g_I\frac{l(l+1)}{j(j+1)}F\left\langle\frac{a_0^3}{r^3}\right\rangle,$$

$$B=-c\ \mathrm{Ry}\frac{Q}{a_0^2}\frac{2j-1}{j+1}R\left\langle\frac{a_0^3}{r^3}\right\rangle,$$
$$\tag{2}$$

where Ry is the Rydberg constant, α the fine-structure constant, and a_0 the first Bohr radius. The relativistic correction factors F and R are, for f electrons, very close to unity and are here discarded.

[8] L. Davis, B. T. Feld, C. W. Zabel, and J. R. Zacharias, Phys. Rev. **76**, 1076 (1949).

In order to estimate $\langle r^{-3}\rangle$ one needs some approximate radial wave function. In most applications hydrogenic wave functions have been used, but these cannot be expected to be good approximations, except for electrons moving very close to the nucleus. This is clearly demonstrated by self-consistent-field (SCF) calculations.

With the wave function discussed in the Appendix, which is a modification of the hydrogenic wave function to better agreement with SCF calculations, we get, in atomic units (a.u.)

$$\kappa=0.40:\ \langle r^{-3}\rangle=10.6,$$

$$\kappa=0.44:\ \langle r^{-3}\rangle=10.4.$$

This shows, as one would expect, that the shape of the wave function is not critical when $\langle r^{-3}\rangle$ is determined from the experimental spin-orbit coupling constant. Ridley[9] gives, for Tm^{3+}, 11.5 a.u., which should be slightly higher than for the neutral atom, since the removal of the outer electrons pushes the other electrons a little closer to the nucleus. The very crude hydrogenic formula for the spin-orbit coupling constant,

$$\zeta=hc\ \mathrm{Ry}\alpha^2 Z_{\mathrm{eff}}\langle r^{-3}\rangle=hc\ \mathrm{Ry}\alpha^2\frac{Z_{\mathrm{eff}}^4}{n^3 l(l+\frac{1}{2})(l+1)},\tag{3}$$

which is frequently used by spectroscopists, gives $\langle r^{-3}\rangle=13.1$ a.u., which is certainly too high.
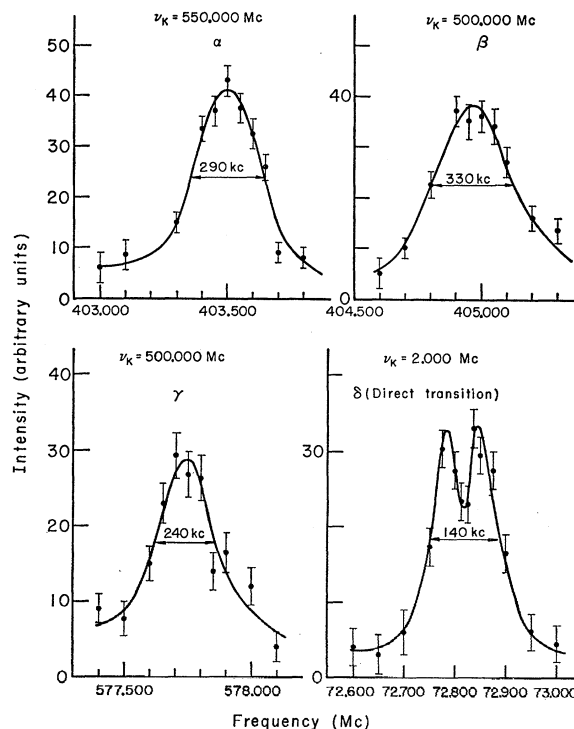

FIG. 4. Resonance curves for the $\Delta F=0$ transitions and one $\Delta F=1$ transition.

[9] E. C. Ridley, Proc. Cambridge Phil. Soc. **56**, 41 (1960).

With $\langle r^{-3}\rangle = 10.5$ a.u. we get, for the nuclear moments (uncorrected values)

$$|\mu_I| = 0.26 \pm 0.02 \text{ nm},$$

$$|Q| = 0.61 \pm 0.05 \text{ barn},$$

with the two moments of the same sign. The error in the magnetic moment is large enough to include diamagnetic corrections. For the quadrupole moment, on the other hand, corrections of the Sternheimer type,[10] which have not been considered here, might make the corrected value fall outside the given limits.

The hfs of the stable isotope Tm¹⁶⁹ has been investigated optically by Lindenberger,[11] and he gives for the magnetic moment

$$\mu_I{}^{169} = -0.20_5 \pm 0.02 \text{ nm}.$$

Although he uses hydrogenic wave functions, he gets, surprisingly enough, consistent results from the hfs constants for the $4f$ and $6s$ electrons. With our value of $\langle r^{-3}\rangle$ for the $4f$ electron, which we believe is more accurate, we obtain from his data

$$\mu_I{}^{169} = -0.25 \text{ nm},$$

which is outside the given limits of error.

CORRECTIONS TO THE ATOMIC g VALUE

Since the ground state of thulium is essentially a single-electron state, the admixture of other states is

TABLE I. Data processed by the computer for Tm¹⁷⁰.

Data No.	H (gauss)	ν_{obs} (Mc/sec)	$\nu_{obs}-\nu_{calc}$ (Mc/sec)	Transition
1	0.711(71)	0.950	+0.066	α
2	1.418(70)	1.760	−0.002	α
3	10.865(39)	13.600	+0.039	α
4	20.754(59)	26.100	−0.055	α
5	38.243(50)	49.070	−0.007	α
6	93.043(33)	123.000	−0.021	α
7	0.711(71)	1.100	+0.035	β
8	1.418(70)	2.125	+0.000	β
9	10.865(39)	16.400	+0.020	β
10	20.754(59)	31.350	+0.061	β
11	55.192(43)	81.830	+0.048	β
12	93.043(33)	136.150	−0.012	β
13	0.711(71)	1.470	+0.008	γ
14	10.865(39)	22.310	−0.019	γ
15	20.754(59)	42.700	+0.024	γ
16	55.192(43)	113.745	+0.054	γ
17	159.545(24)	213.460	−0.045	α
18	159.545(24)	231.715	−0.034	β
19	278.798(20)	577.740	+0.030	γ
20	278.798(20)	404.970	+0.017	β
21	298.380(19)	403.505	−0.011	α
22	0.740(42)	72.855	+0.002	δ
23	2.818(42)	72.815	−0.003	δ
24	93.043(33)	192.010	+0.016	γ

$(F,m) \leftrightarrow (F',m')$

α: $(9/2,1/2) \leftrightarrow (9/2,-1/2)$
β: $(7/2,-1/2) \leftrightarrow (7/2,-3/2)$
γ: $(5/2,3/2) \leftrightarrow (5/2,1/2)$
δ: $(7/2,-1/2) \leftrightarrow (9/2,-1/2)$

[10] R. Sternheimer, Phys. Rev. **86**, 316 (1952); **95**, 736 (1954).
[11] K. H. Lindenberger, Z. Physik **141**, 476 (1955).

very small. Furthermore, the electrostatic interaction can mix only states with the same S, L, and J and hence has no effect on the g value. An estimate of the configuration interaction caused by the spin-orbit coupling shows that its effect is quite negligible compared with the experimental uncertainty. Therefore, all the measurable deviation from the classical Landé value must be due to (a) the anomalous magnetic moment of the electron and (b) relativistic and diamagnetic effects. By relativistic effects we mean here the change of the interaction between the atomic moment and the external field, due to the velocity of the electron, and the change of the spin-orbit coupling, due to the external field. These corrections follow directly from the Dirac equation for a single electron, and are proportional to the kinetic energy T in the first approximation. The diamagnetic correction is caused by changes in the spin-other-orbit and orbit-orbit interactions, due to the external field. This correction depends essentially on the electron density in the core.

The relativistic correction to the magnetic moment of a single electron has been calculated by Breit[12] and Margenau[13] and can be written

$$\delta g_1 = \alpha^2 \frac{(j+\frac{1}{2})^2}{j(j+1)}\langle T\rangle. \tag{4}$$

All radial integrals are here expressed in atomic units. This correction is usually referred to as the Breit-Margenau correction.

In their discussion of the Zeeman effect in atomic oxygen, Abragam and Van Vleck[14] have calculated the diamagnetic correction, assuming spherically symmetric electron density. From their expressions we get for the diamagnetic correction to the Zeeman energy for a single electron in the state (nlm_sm_l),

$$\delta Z = -\mu_0 H\alpha^2[(m_l+2m_s)\langle Y\rangle - m_s\langle \sin^2\theta\rangle\langle U\rangle], \tag{5}$$

where

$$U = \frac{1}{r^3}\int_0^r r'^2\rho(r')dr', \quad Y = \frac{1}{3}\left[U + \int_r^\infty \frac{\rho(r')}{r'}dr'\right],$$

and

$$\langle \sin^2\theta\rangle = 2\frac{l(l+1)-1+m_l^2}{(2l-1)(2l+3)}.$$

Here $\rho(r')$ is the radial density of all electrons, except the one we are taking the average for.

From Eqs. (4) and (5) we get the total correction for an f electron in the state $^2F_{7/2}$,

$$\delta g = -\alpha^2\left[\frac{64}{63}\langle T\rangle + \frac{8}{7}\langle Y\rangle - \frac{8}{63}\langle U\rangle\right].$$

[12] G. Breit, Nature **122**, 649 (1928).
[13] H. Margenau, Phys. Rev. **57**, 383 (1940).
[14] A. Abragam and J. H. Van Vleck, Phys. Rev. **92**, 1448 (1953). More general treatments are given by K. Kembe and J. H. Van Vleck, Phys. Rev. **96**, 66 (1954), and F. R. Innes and C. W. Ufford, Phys. Rev. **111**, 194 (1958).

TABLE II. Corrections to atomic g value, as calculated with modified and unmodified hydrogenic functions.

	Hydrogenic wave function ($\kappa=0$)	Modified hydrogenic wave functions	
		$\kappa=0.40$	$\kappa=0.44$
Landé value	1.14286	1.14286	1.14286
Schwinger correction	+0.00033	+0.00033	+0.00033
Breit-Margenau correction	−0.00166	−0.00134	−0.00126
Diamagnetic correction	−0.00084	−0.00070	−0.00066
Theoretical value	1.14069	1.14115	1.14127
Experimental value	1.14122±0.00015		

With the wave function described in the Appendix and the electron density from the Thomas-Fermi model, we obtain the following values of the radial integrals:

$$\kappa=0.40: \langle T\rangle=24.7, \quad \langle U\rangle=16.5, \quad \langle Y\rangle=13.3 \text{ a.u.;}$$

$$\kappa=0.44: \langle T\rangle=23.3, \quad \langle U\rangle=15.6, \quad \langle Y\rangle=12.5 \text{ a.u.}$$

In table II we have summarized all the corrections and for comparison have also given the corresponding values obtained with a hydrogenic wave function.

It is seen that the agreement between the experimental and calculated g values is extremely good with κ around 0.4, the value obtained by comparison with SCF wave functions (see Appendix).

Since all wave functions used here are fitted to the experimental spin-orbit coupling constant with the same potential, the difference in result is entirely due to the difference in shape. The experimental deviation from the Landé value together with the spin-orbit coupling therefore constitutes a measure of the shape of the wave function. Although the accuracy here is not very high, it definitely shows that the hydrogenic wave function is too sharp. The hydrogenic wave function used above has been fitted to the experimental spin-orbit coupling constant by means of the Thomas-Fermi potential. If Z_{eff} is instead determined from Eq. (3) the agreement becomes even much poorer.

ACKNOWLEDGMENTS

APPENDIX. ANALYTIC RADIAL WAVE FUNCTIONS

For numerical calculations it is very convenient to have an approximate analytic expression for the wave functions obtained by the SCF method. This also makes it possible to interpolate between such functions. A suitable form is the Slater-Löwdin approximation,[15]

[15] J. C. Slater, Phys. Rev. 42, 33 (1932); P.-O. Löwdin, Phys. Rev. 90, 120 (1953).

which for functions of the $4f$ type (single maximum) is

$$R(r)=r^n(c_1e^{-a_1r}+c_2e^{-a_2r}+c_3e^{-a_3r}+\cdots).$$

With three terms in this expansion the agreement with the original wave function is extremely good. For our purpose, however, we prefer to use a two-parameter function, and choose the symmetric form

$$R(r)=Nr^ne^{-ar}\cosh[\kappa(ar-n)]$$
$$=\tfrac{1}{2}Nr^n(e^{-\kappa n}e^{-a(1-\kappa)r}+e^{\kappa n}e^{-a(1+\kappa)r}). \quad (A\text{-}1)$$

For this function the position of the maximum depends only on a, and the other parameter, κ, determines essentially the shape. A function of this type fitted to the SCF wave function for Tm^{3+} is shown in Fig. 5. One could easily determine both parameters in Eq. (A-1) by interpolation or extrapolation from existing SCF calculations, but we believe that more reliable wave functions are obtained if one of the parameters
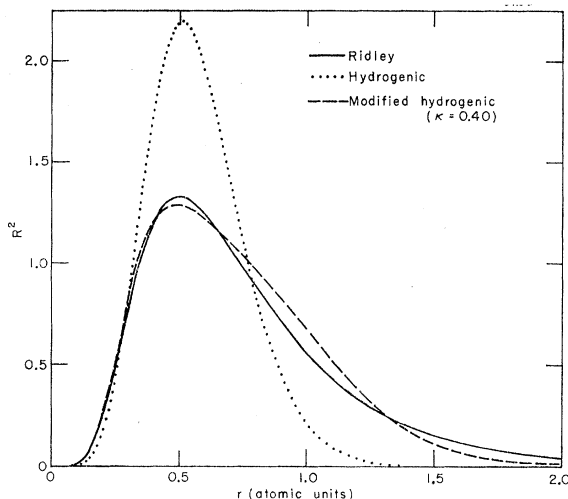
FIG. 5. Radial wave functions for Tm^{3+}.
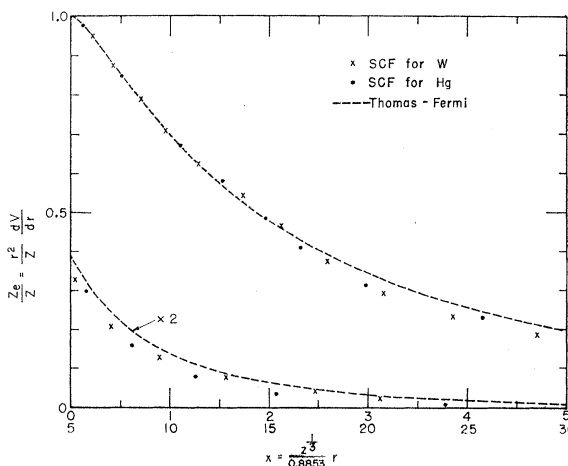
FIG. 6. r^2dV/dr from the Thomas-Fermi potential and from SCF calculations in tungsten and mercury.

is determined from the experimental spin-orbit coupling constant. Since the shape of the wave function changes very little from element to element, we have determined κ by comparison with SCF wave functions and a from the spin-orbit coupling constant. In the latter case we have used the Thomas-Fermi potential, which is accurate enough for this purpose. This potential is particularly close to SCF potentials near the nucleus, where the main contribution to the spin-orbit coupling originates (see Fig. 6).

No SCF calculations are available for any rare-earth atoms but some have recently been carried out for the Pr^{3+} and Tm^{3+} ions.[9] The difference in shape between the $4f$ wave functions for these ions is very small, and both correspond to a κ value slightly greater than 0.4. Since one would not expect the shape to differ much between the ions and the atoms, this should be a reasonable value also for the atoms. This is in agreement with the value obtained by extrapolation from heavier atoms like W and Hg.

For the wave function (A-1) the following formulas are easily verified (subscript hy indicates hydrogenic value).

$$N^2 = \frac{(2a)^{2n+1}}{(2n)!}\frac{1}{C_{2n+1}} = \frac{N_{hy}^2}{C_{2n+1}},$$

$$\langle r^{-m}\rangle = \frac{(2a)^m(2n-m)!}{(2n)!}\frac{C_{2n+1-m}}{C_{2n+1}} = \langle r^{-m}\rangle_{hy}\frac{C_{2n+1-m}}{C_{2n+1}},$$

$$\langle T\rangle = \frac{1}{2}\left[n(n-1)\langle r^{-2}\rangle - \left\langle \frac{1}{R}\frac{d^2R}{dr^2}\right\rangle\right]$$

$$= \frac{1}{2}a^2\left[1-\kappa^2-2\kappa\frac{D_{2n}}{C_{2n+1}}\right],$$

where

$$C_s = \tfrac{1}{4}\left[e^{-2n\kappa}(1-\kappa)^{-s}+2+e^{2n\kappa}(1+\kappa)^{-s}\right],$$

and

$$D_s = \tfrac{1}{4}\left[e^{-2n\kappa}(1-\kappa)^{-s}-e^{2n\kappa}(1+\kappa)^{-s}\right].$$

Preformation Factor in Emission of Complex Particles from Nuclear Reactions*

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The reason why complex particles (alphas, tritons, Li^6 nuclei, etc.) are emitted in nuclear reactions as frequently as nucleons (after corrections for Coulomb barrier penetration and energetics), whereas ice crystals are never emitted from evaporating water droplets is investigated. It is shown that the difference is entirely explained by the fact that a nucleus is a highly degenerate system subject to Fermi-Dirac statistics, whereas an evaporating water droplet is a nondegenerate statistical system.

IF one considers an excited compound nucleus to be a conglomeration of neutrons and protons similar to a liquid drop, it is intuitively appealing to assume that neutrons and protons should be emitted most readily in nuclear reactions, and tritons, alpha particles, Li^6 nuclei, etc., should be impeded by a preformation factor, f, relative to the emission of nucleons. (We ignore here other factors affecting emission such as Coulomb barrier penetration factors, energetics, etc.) The problem seems to be analogous to that of an evaporating droplet of water, where the evaporation of a sizeable crystal of ice is certainly very much less probable than the evaporation of water molecules one at a time.

This view indeed prevailed in early treatments of the subject, the best known of which is Bethe's many body theory of alpha decay.[1] However, when it was found experimentally that alpha particles are frequently

emitted from nuclear reactions so that f is close to unity, the use of a preformation factor became unfashionable, although it is still occasionally discussed or referred to.[2] Proofs have been offered to show that if complex nuclei are captured with geometric cross sections in experiments where they bombarded nuclei, application of the principle of detailed balance indicates that f must be unity. However, this cannot explain the difference between a decaying compound nucleus and an evaporating water droplet. In the latter case, an ice crystal striking the droplet would certainly be absorbed with the geometric cross section, and detailed balance is essentially an expression of invariance under time reversal which is a classical as well as a quantum mechanical principle.

It is the purpose of this paper to clearly elucidate the difference between the two cases. It will be shown that it is due solely to the fact that a nucleus is a highly

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[1] H. A. Bethe, Revs. Modern Phys. **9**, 69 (1937).

[2] See, for example: J. J. Devaney, Phys. Rev. **91**, 587 (1953); H. A. Toelhoek and P. J. Brussaard, Physics **21**, 449 (1955); G. H. McCormick, H. G. Blosser, B. L. Cohen, and T. H. Handley, J. Inorg. & Nuclear Chem. **2**, 269 (1956).