

obtained by use of Einstein's relation and Eqs. (37) and (39).

The equipotentials for the small-signal case are given by

$$\Delta x = \frac{(D_n - D_p) \cdot 2y_0}{\theta D_0} \frac{\Delta \phi - \Delta \phi_2}{\Delta \phi_1 - \Delta \phi_2}, \quad (82)$$

as obtained by expanding the logarithms. This result may be written explicitly in terms of  $y$  by use of the appropriate expressions for  $\Delta \phi$  and  $E_x^{(oc)}$ . From it, the distance along the slab between intersections of an equipotential with the surfaces is given by

$$\Delta x = (D_n - D_p) \cdot 2y_0 / \theta D_0$$

for the small-signal case. This  $\Delta x$  is a direct measure

of  $\theta$ . It is large compared with the thickness  $2y_0$  of the slab, since  $\theta$  has been assumed small.

#### ACKNOWLEDGMENTS

Among the individuals to whose stimulating interest the author is indebted, particular mention should be made of W. H. Brattain, to whose awareness of difficulties attendant upon techniques for measuring recombination this work owes its inception; and of T. M. Buck, whose experimental studies have been a most valuable guide. They and J. A. Burton, R. C. Fletcher, and Conyers Herring made helpful suggestions concerning the manuscript. R. W. Hamming and Miss D. P. Booth capably provided invaluable help through numerical computations for the nonlinear cases.

## Hyperfine Structure and Nuclear Moments of Gadolinium\*

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Study of the optical hyperfine structure of several Gd I lines using enriched isotopes shows that the spins of the odd isotopes Gd<sup>155</sup> and Gd<sup>157</sup> each are  $\frac{3}{2}$ . The ratio of the magnetic moments is  $\mu_I(\text{Gd}^{155})/\mu_I(\text{Gd}^{157}) = 0.80 \pm 0.02$ . The magnetic moment for Gd<sup>157</sup> obtained from two lines is  $-0.37 \pm 0.04$  nm. Deviations from the interval rule in these two lines can be accounted for with a quadrupole moment of approximately  $1.0 \times 10^{-24}$  cm<sup>2</sup> for Gd<sup>157</sup> and  $1.1 \times 10^{-24}$  cm<sup>2</sup> for Gd<sup>155</sup>. The known anomalous isotope shift between neutron numbers 88 and 90 (Gd<sup>152</sup> and Gd<sup>154</sup>) is accurately measured for several lines.

### INTRODUCTION

SPECTROSCOPIC measurements of the nuclear moments of the heavier odd-neutron nuclei have been hampered by the presence in the corresponding elements of several even-even isotopes. With the availability of enriched samples, however, it has become possible to add to the rather scanty data on such nuclides for comparison with predictions of the unified shell model developed by Bohr<sup>1</sup> and by Bohr and Mottelson.<sup>2</sup> The recent work of Mottelson and Nilsson<sup>3</sup> shows that marked deviations are to be expected from the moments derived from the single-particle model in those regions of the periodic table where the nuclear deformations are large. Thus whereas a spin  $I=7/2$  was assigned by Klinkenberg<sup>4</sup> to the odd isotopes <sup>64</sup>Gd<sup>155</sup> and <sup>64</sup>Gd<sup>157</sup> on the basis of the shell model, it appears that if the nuclei are sufficiently deformed the lowest level should be either  $3/2^-$  or  $5/2^+$ .<sup>5</sup>

Previous studies<sup>6-9</sup> with natural gadolinium have been chiefly concerned with the isotope shifts, and Murakawa<sup>8</sup> has confirmed the anomalously large shift between isotopes 152 and 154 that was expected by analogy with Nd and Sm. This author, and somewhat earlier Suwa,<sup>9</sup> attempted to draw conclusions about the spins and magnetic moments of the odd isotopes of Gd from the unresolved structure underlying the strong components due to the even isotopes. Both investigators concluded that the spins were probably greater than  $3/2$ , and Murakawa assumed the value  $7/2$  in estimating the magnetic moments. It is therefore apparent that further study of the hyperfine structure of Gd with separated isotopes was needed. The very complete classification of the lines of both Gd I and Gd II by Russell<sup>10</sup> makes it possible to derive the nuclear moments from the observed splittings.

The composition of the four samples of enriched isotopes used in the present work, as compared with that of natural Gd, is shown in Table I.

\* Work supported by the National Science Foundation.

<sup>1</sup> A. Bohr, Kgl. Danske Videnskab. Selskab Mat.-fys. Medd. 26, No. 14 (1952).

<sup>2</sup> A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab Mat.-fys. Medd. 27, No. 16 (1953).

<sup>3</sup> B. R. Mottelson and S. G. Nilsson, Phys. Rev. 99, 1615 (1955).

<sup>4</sup> P. F. A. Klinkenberg, Revs. Modern Phys. 24, 63 (1952).

<sup>5</sup> Reference 3, Fig. 2, p. 1616.

<sup>6</sup> P. F. A. Klinkenberg, Physica 12, 33 (1946).

<sup>7</sup> P. Brix and H. D. Engler, Z. Physik 133, 362 (1925).

<sup>8</sup> K. Murakawa, Phys. Rev. 96, 1543 (1954).

<sup>9</sup> S. Suwa, J. Phys. Soc. (Japan) 8, 377 (1953) and Phys. Rev. 86, 247 (1952).

<sup>10</sup> H. N. Russell, J. Opt. Soc. Am. 40, 550 (1950).

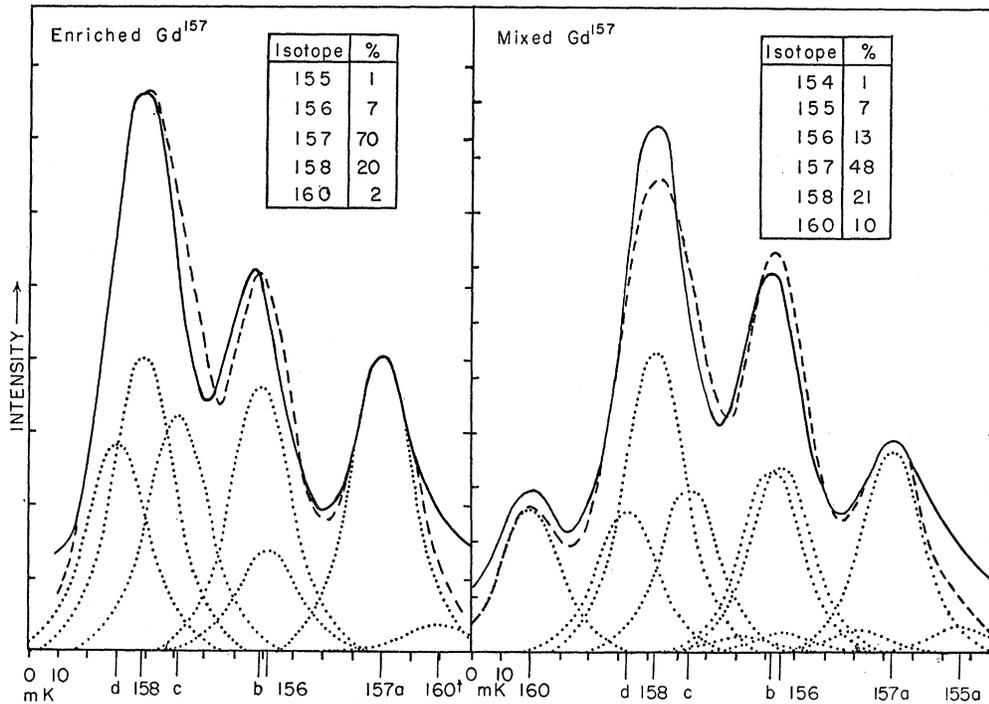


FIG. 1. Intensity distribution of Gd I  $\lambda 5015$  from two mixtures of the isotopes. The dotted curves indicate the positions assumed for the individual components. The dashed curves represent their sum.

In the present work the spectrum was excited in a Schuler tube with demountable cathode, cooled in liquid nitrogen, similar in design to that described by Arroe and Mack.<sup>11</sup> The hyperfine structure was resolved by means of a Fabry-Perot etalon crossed with a three-prism glass spectrograph in the external-beam mounting. The etalon mirrors were nine-layer dielectric films of ZnS and cryolite, which give a reflectance of over 95% and negligible absorption throughout the region of the spectrum from 4200 Å to 5900 Å. With the larger spacers used, these yield a calculated resolu-

ing power to excess of  $10^6$ , but the Doppler width of the lines limited the effective resolution to approximately  $8 \times 10^5$ .

**Gd<sup>155</sup> and Gd<sup>157</sup>**

On plates taken with the samples enriched in the odd isotopes the hfs of Gd<sup>155</sup> and Gd<sup>157</sup> were found to be very similar except that the Gd<sup>155</sup> splittings were about four fifths as large as those of Gd<sup>157</sup>. All observed lines with  $a^{11}F^0$  as the lower term show a structure consisting of three resolved components with the strongest toward the red. ( $\lambda 5015$  is typical of these.) Most lines with  $a^9D^0$  as the lower term show only a single slightly broadened component for the odd isotope and a weaker satellite which can be attributed to the most abundant even isotope in the sample. A few lines with this lower term however have structure with three or four resolved components. The unclassified line 4436.1 Å shows five components on Gd<sup>157</sup> plates and four barely resolved components on Gd<sup>155</sup> plates.

As shown in Table I, the samples enriched in the odd isotopes contained enough of the even isotopes to appreciably distort the observed patterns. This difficulty was partially overcome by adding a known (roughly equal) amount of the natural mixture. If the observed contour of the pattern then behaved as expected, the interpretation given to the original pattern was assumed to be correct. Essentially this is a method of extrapolating to zero concentration of the even isotopes.

Results of intensity measurements on the line  $\lambda 5015$

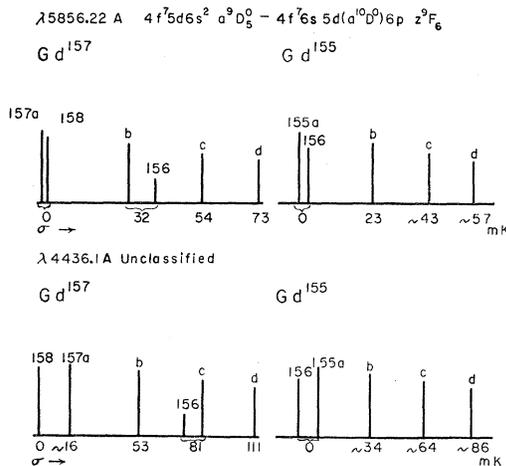


FIG. 2. Hfs of Gd I  $\lambda 4436.1$  and  $\lambda 5856$  from enriched isotope samples.

<sup>11</sup> O. H. Arroe and J. E. Mack, J. Opt. Soc. Am. 40, 386 (1950).

( $a^{11}F_8^\circ - z^{11}G_9$ ) for both the enriched and mixed Gd<sup>157</sup> samples are given in Fig. 1.<sup>12</sup> The dashed curve was obtained by assuming a value of 3/2 for the spin, giving each component its theoretical intensity and shifting their positions until the composite curve made the best fit with the observed contour (solid curve). The positions of the even isotopes were assigned in accordance with isotope shift data obtained using natural Gd. Only the diagonal components of the hfs needed to be considered since the intensity of the off-diagonal ones comprises only about two percent of the total intensity. No other assumed spin value will give a contour that fits the one observed. Results for Gd<sup>155</sup> were similar to those shown for Gd<sup>157</sup>. From this it is seen that the many lines with a three-component structure may be interpreted on the basis of a structure for the odd isotopes consisting of four components which degrade toward the red.

The interpretation of the structure of the lines  $\lambda 5856$  ( $a^9D_5^\circ - z^9F_6$ ) and  $\lambda 4436.1$  (unclassified) are given in Fig. 2. The presence of four components for the odd isotope confirms that its spin is 3/2 in each case. The evidence from  $\lambda 4436.1$  however is not conclusive because of the unknown  $J$ -values for this line. Thus all available evidence indicated that  $I=3/2$  for both Gd<sup>155</sup> and Gd<sup>157</sup>.

The hfs of the odd isotopes show marked deviations from the interval rule in lines where the positions of three components could be accurately measured or estimated. For example, using the interval rule and the separation of the first two components gives 110 mK for the total width of the structure for Gd<sup>157</sup> in  $\lambda 5015$ . This would place the fourth component well outside the observed contour and 20 mK from its position deduced from the intensity curve. The line  $\lambda 5103$  exhibits a similar behavior. There are no identified levels which could perturb the terms involved in these two lines so these deviations from the interval rule have been ascribed to a quadrupole moment.

The hfs of a term is expressed by the following equation<sup>13</sup>:

$$\Delta\sigma = \frac{1}{2}AK + B\{K(K+1) - (4/3)IJ(J+1)(I+1)\},$$

where  $K = F(F+1) - I(I+1) - J(J+1)$ , and

$$B = -\frac{3}{8} \frac{e^2Q}{hcIJ(2I-1)(2J-1)} \left( \frac{3 \cos^2\theta - 1}{r^3} \right)_{Av(M_J=J)}$$

$\Delta\sigma$  is the displacement of the hfs level from the center of gravity of the term. In order to use this equation for the evaluation of the nuclear moments the interval factor  $A$  must be expressed in terms of the coupling constants  $a_i$  for the individual electrons and

<sup>12</sup> The name kayser (abbreviated K) and the symbol  $\sigma$  are used for  $\text{cm}^{-1}$ . The abbreviation mK is used for the millikayser ( $10^{-3}\text{K}$ ).

<sup>13</sup> H. Kopfermann, *Kernmomente* (Akademische Verlagsgesellschaft M. B. H., Leipzig, 1940).

TABLE I. Composition of the enriched isotope samples compared with natural Gd.

	152	154	155	156	157	158	160
Nat. Gd	0.2	2.86	15.61	20.59	16.42	23.45	20.87
Gd <sup>155</sup>	0.5	1.2	72.3	17.7	4.6	2.9	0.8
Gd <sup>157</sup>	0.04	0.11	1.23	7.3	69.7	19.9	1.72
Gd <sup>152</sup>	14.9	9.7	27.3	19.3	10.1	11.7	7.0
Gd <sup>154</sup>	0.3	33.2	38.6	15.9	5.5	4.5	2.0

$[(3 \cos^2\theta - 1)/r^3]_{Av(M_J=J)}$  must be evaluated in  $B$ . With the assumption that the terms are formed by pure  $LS$ -coupling these calculations can be made from the  $LS$ -coupling wave functions. A summary of these calculations for the terms of the lines analyzed in the present work is given in the appendix.

The lines which were used for the determination of the magnetic and quadrupole moments were the lines  $\lambda 5015$  ( $4f^7 5d^2 6s a^{11}F_8^\circ - 4f^7 5d^2 6p z^{11}G_9$ ) and  $\lambda 5103$  ( $a^{11}F_7^\circ - z^{11}G_8$ ). These lines were chosen because they have a relatively wide hfs and are not disturbed by close neighboring lines. Also the  $LS$ -coupling wave functions for these levels may be easily determined using the method of Gray and Wills.<sup>14</sup>

Since only the positions of the diagonal components could be obtained from the intensity curves it was necessary to take into account the hfs of both the initial and final terms when calculating the moments from the hfs of a line. This was done by subtracting the hfs of the initial term from that of the final term. As an example the separation of the first and fourth components in  $\lambda 5015$  is given by

$$\Delta\sigma(1-4) = \{(51/2)A(a^{11}F_8^\circ) - (102B(a^{11}F_8^\circ)) - \{(57/2)A(z^{11}G_9) - 114B(z^{11}G_9)\}.$$

Substituting the values given in the appendix for the  $A$ 's and the  $B$ 's it was found in all cases that to within experimental accuracy the  $a_d$ 's could be neglected and that the ratio of the coefficients of  $a_s$  and  $a_p$  were constant for a given line. Results of calculations on the two lines considered along with the necessary data are

TABLE II. Summary of calculations to determine the nuclear moments from the structure of  $\lambda 5015$  and  $\lambda 5103$ .

		Gd <sup>157</sup>	Gd <sup>155</sup>
$\lambda 5015$	$\Delta\sigma(1-2)$ , mK	-41	-34
$\Delta\sigma(1-2)/0.594$	$\Delta\sigma(1-4)$ , mK	-90	-72
$= a_s - 1.70a_p - 12.0 \times 10^{-3}Q$	$a_s - 1.70a_p$	-57	-46
$\Delta\sigma(1-4)/1.59$	$Q(10^{-24} \text{ cm}^2)$	1.0	1.1
$= a_s - 1.70a_p + 0.67 \times 10^{-3}Q$			
$\lambda 5103$	$\Delta\sigma(1-2)$ , mK	-36	-31
$\Delta\sigma(1-2)/0.561$	$\Delta\sigma(1-4)$ , mK	-81	-62
$= a_s - 2.06a_p - 8.38 \times 10^{-3}Q$	$a_s - 2.06a_p$	-55	-42
$\Delta\sigma(1-4)/1.49$	$Q(10^{-24} \text{ cm}^2)$	1.1	1.4
$= a_s - 2.06a_p + 0.54 \times 10^{-3}Q$			

<sup>14</sup> N. M. Gray and L. A. Wills, Phys. Rev. 38, 248 (1931).

TABLE III. Summary of isotope shifts. The position of the components are given in mK with the 160 component taken as the origin.

	160	158	157	156	155	154	152
5015 ( $a^{11}F_8^{\circ} - z^{11}G_9$ )	0	42	~78	85	~112	141	262
5103 ( $a^{11}F_7^{\circ} - z^{11}G_8$ )	0	42	...	84	...	138	261
4374 ( $a^9D_4^{\circ} - \gamma^9F_3$ )	0	57	~104	~116	154	192	365
4402 ( $a^9D_6^{\circ} - \gamma^9F_6$ )	0	58	~107	~119	163	202	384
4436.1 (unclassified)	0	75	...	148	...	~242	492

summarized in Table II. The positions of the components in  $\lambda 5015$  are those obtained from the intensity curve. The positions in  $\lambda 5103$  are those estimated directly from the plates by setting with microscope and cross-hair and are thus more uncertain. Only the differences  $\Delta\sigma(1-2)$  and  $\Delta\sigma(1-4)$  were used in the calculations because these were considered to be the most accurately determined separations. However, the positions of the third component in  $\lambda 5015$  calculated from the derived values of  $a_s$ ,  $a_p$ , and  $Q$  agrees with the value obtained from the intensity trace to within 1 mK.

The values of  $a_s$  and  $a_p$  were determined from the two equations in the second column in Table II. The results for Gd<sup>157</sup> were

$$a_s = -65 \text{ mK},$$

$$a_p \approx -5 \text{ mK}.$$

The experimental uncertainty in  $a_s$  is estimated to be about 10%.

The Fermi-Segrè-Goudsmit formula was used to determine  $\mu_I$  from  $a_s$ :

$$a_s = -\frac{8R\alpha^2ZZ_a^2}{3 \cdot 1836n_a^3} \left(1 - \frac{d\sigma}{dn}\right) \frac{\mu_I}{I} \kappa(j, Z)(1-\delta)(1-\epsilon).$$

Since the spectrum of Gd is not extensively enough classified to allow a direct determination of

$$\frac{Z_a^2}{n_a^3} \left(1 - \frac{d\sigma}{dn}\right),$$

the value 0.319 for Gd I obtained by an indirect method by Brix<sup>15</sup> was used. The relativistic correction  $\kappa(\frac{1}{2}, 64) = 1.60$  was taken from tables in Kopfermann.<sup>13</sup> The Crawford-Schawlow correction<sup>16</sup> is  $(1-\delta) = 1 - 0.053$  and the Bohr-Weisskopf correction<sup>17</sup> is  $(1-\epsilon) = 1 + 0.016$ . Substituting these values into the above equation gives

$$a_s = 0.266\mu_I/I$$

or

$$I = -0.37 \text{ nm for Gd}^{157}.$$

<sup>15</sup> P. Brix, Z. Physik **132**, 579 (1952).<sup>4</sup>

<sup>16</sup> M. F. Crawford and A. L. Schawlow, Phys. Rev. **76**, 1310 (1949).

<sup>17</sup> A. Bohr and V. F. Weisskopf, Phys. Rev. **77**, 94 (1950).

The average value of the ratio of the magnetic moments obtained from Table II and the total splitting of the lines  $\lambda 5856$  and  $\lambda 4436$  is

$$\frac{\mu_I(\text{Gd}^{155})}{\mu_I(\text{Gd}^{157})} = 0.80 \pm 0.02.$$

This gives  $\mu_I = -0.30 \text{ nm}$  for Gd<sup>155</sup>.

The quadrupole moments deduced from the two lines are constant to within experimental accuracy which was estimated to be about  $\pm 30\%$ . Giving the determinations from  $\lambda 5015$  more weight in averaging, the best values are

$$Q = 1.0 \times 10^{-24} \text{ cm}^2 \text{ for Gd}^{157},$$

$$Q = 1.1 \times 10^{-24} \text{ cm}^2 \text{ for Gd}^{155}.$$

The correction factors for  $Q$  considered by Sternheimer<sup>18</sup> have been neglected. The experimental uncertainties given include only that part which arises from the accuracy of measurements. Additional uncertainties which might arise during the calculations such as deviations from  $LS$ -coupling and approximations made in calculating  $(r^{-3})_{Av}$  are much harder to estimate and have not been included.

The value for the nuclear spin  $I = 3/2$  is in agreement with the value predicted by Mottelson and Nilsson.<sup>3</sup> The values for the quadrupole moments are about thirty percent lower than the interpolated values used by Mottelson and Nilsson in making their prediction but using the present value one still obtains  $I = 3/2$  for the Gd isotopes from their level scheme.

#### ISOTOPE SHIFTS

Isotope shifts were measured in five lines on plates taken with enriched Gd<sup>152</sup> and Gd<sup>154</sup> samples. The results are summarized in Table III. It is confirmed that the shift Gd<sup>152</sup>-Gd<sup>154</sup> is anomalously large when compared to the other shifts caused by the addition of two neutrons in the same element. The average values for the ratios  $\Delta\sigma(158-160):\Delta\sigma(156-158):\Delta\sigma(154-156):\Delta\sigma(152-154)$  obtained from the five lines were 1.00:1.01:1.32:3.02. The center of gravity of the 155 and 157 patterns in the line  $\lambda 5015$  were estimated from the intensity curves of the line from enriched 155 and 157 samples. The 155 and 157 components appeared as single, slightly broadened components in the lines  $\lambda 4374$  and  $\lambda 4402$ . However the 157 component was never separated from the 156 component and the positions of these two components had to be estimated from the center of gravity of the composite line on plates taken with different mixtures of the isotopes. These more uncertain values are preceded by  $\sim$  in the table. All other values should be assigned a probable error of 1 mK.

<sup>18</sup> R. Sternheimer, Phys. Rev. **95**, 736 (1954).

## ACKNOWLEDGMENTS

The author wishes to express his gratitude to Professor F. A. Jenkins for his guidance and advice throughout the course of this work and to Dr. J. M. Stone for the preparation of the very excellent interferometer mirrors. Thanks are due also to Oak Ridge National Laboratory for the separated isotopes.

## APPENDIX

The factors  $A$  and  $B$  were evaluated for the terms  $4f^7 5d^2 6s$   $a^{11}F_8^{\circ}$ ,  $a^{11}F_7^{\circ}$ ,  $4f^7 5d^2 (a^{10}F) 6p z^{11}G_9$ ,  $z^{11}G_8$ . These calculations were made by first writing out the  $LS$ -coupling wave functions for the terms using the method of Gray and Wills.<sup>14</sup> The interval factors were obtained from the zero-order functions by evaluating the matrix elements of  $H_z$ , the  $z$ -component of the magnetic field produced at the nucleus by the electrons.<sup>19</sup> The results were

$$A(a^{11}F_8^{\circ}) = \frac{1}{16}a_s + \frac{37}{80}a_d' + \frac{3}{80}a_d'' + \frac{1}{5}a_d''',$$

$$A(a^{11}F_7^{\circ}) = \frac{74}{1120}a_s + \frac{1177}{2800}a_d' + \frac{143}{2800}a_d'' + \frac{51}{175}a_d''',$$

$$A(z^{11}G_9) = \frac{1}{6}a_p' + \frac{37}{90}a_d' + \frac{1}{30}a_d'' + \frac{8}{45}a_d''',$$

$$A(z^{11}G_8) = \frac{1341}{1860}a_p' + \frac{81}{8640}a_p'' + \frac{1}{40}a_p''' + \frac{25}{64}a_d' + \frac{227}{4800}a_d'' + \frac{409}{1800}a_d''',$$

$a_i'$ ,  $a_i''$ , and  $a_i'''$  are the single electron-coupling constants in the notation of Breit and Wills.<sup>20</sup>

The  $(3 \cos^2\theta - 1)_{Av} (M_J = J)$  were calculated from the zero-order functions using the same method used by Schmidt<sup>21</sup> in calculating and quadrupole moment of Ta.

<sup>19</sup> For a more detailed description of the method see S. Suwa, J. Phys. Soc. (Japan) 8, 734 (1953).

<sup>20</sup> G. Breit and L. Wills, Phys. Rev. 44, 470 (1933).

<sup>21</sup> T. Schmidt, Z. Physik 121, 63 (1943).

The results were

$$a^{11}F_8^{\circ} \left( \frac{3 \cos^2\theta - 1}{r^3} \right)_{Av} = \left\{ -\frac{84}{175}R' - \frac{14}{175}R'' + \frac{48}{175}S \right\} \left( \frac{1}{r_{6d}^3} \right)_{Av},$$

$$a^{11}F_7^{\circ} = \left\{ -\frac{143}{350}R' + \frac{13}{100}R'' + \frac{6}{35}S \right\} \left( \frac{1}{r_{6d}^3} \right)_{Av},$$

$$z^{11}G_9 = \left\{ -\frac{84}{175}R' - \frac{14}{175}R'' + \frac{48}{175}S \right\} \left( \frac{1}{r_{6d}^3} \right)_{Av}$$

$$- \frac{2}{5}R' \left( \frac{1}{r_{6p}^3} \right)_{Av},$$

$$z^{11}G_8 = \left\{ -\frac{916}{2625}R' + \frac{197}{2250}R'' + \frac{382}{2625}S \right\} \left( \frac{1}{r_{6d}^3} \right)_{Av} + \left\{ -\frac{47}{150}R' + \frac{54}{675}S \right\} \left( \frac{1}{r_{6p}^3} \right)_{Av},$$

$R'$ ,  $R''$ , and  $S$  are relativistic corrections tabulated by Kopfermann.<sup>12</sup>  $(r_{6d}^{-3})_{Av}$  and  $(r_{6p}^{-3})_{Av}$  were calculated from the fine structure separations using the relation

$$\left( \frac{a_0^3}{r_{nl}^3} \right)_{Av} = \frac{\zeta_{nl}}{R\alpha^2 Z_i \lambda(l, Z)},$$

where  $\zeta_{nl}$  is the spin-orbit parameter for an  $nl$  electron in the notation of Condon and Shortley.<sup>22</sup> The effective nuclear charge was taken to be  $Z_i = Z - 11$  for  $d$  electrons and  $Z_i = Z - 4$  for  $p$ -electrons. The values  $\zeta_{6d} = 864K$  and  $\zeta_{6p} = 1240K$  given by Murakawa<sup>8</sup> were used.  $\lambda(l, Z_i)$  is another relativistic correction given by Kopfermann. These values yield

$$(a_0^3/r_{6d}^3)_{Av} = 2.8 \quad \text{and} \quad (a_0^3/r_{6p}^3)_{Av} = 3.3.$$

<sup>22</sup> E. U. Condon and G. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, London, 1935).