(i) $K'+K=-3$ ($\Delta J=\pm 3$, $\Delta l=\pm 2$)

S_{ijk}	a	b	$\{144\kappa'(\kappa'+1)(\kappa'+2)/[(2\kappa'+1)(2\kappa'+3)(2\kappa'+5)]\}(H^2S-h^2s)^2$
	b	a	$\{144\kappa'(\kappa'-1)(\kappa'-2)/[(2\kappa'-1)(2\kappa'-3)(2\kappa'-5)]\}(H^2V-h^2v)^2$.

Where the cross terms occur, their magnitude is given simply by

$$|M_\Omega M_{\Omega'}^*|^2=|M_\Omega|^2|M_{\Omega'}|^2$$

and the sign of the cross term, i.e., the sign of each of the real expressions $(-iM_rM_\alpha^*)$, etc. . . ., is as follows:

Group	ψ	ψ'	ΔJ	Sign
$K'-K=\pm 1$			1	+
			−1	−
$K'+K=0$	a	b		+
	b	a		−
$K'-K=\pm 2$	a	a		+
	b	b		−

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Hyperfine Structure Anomalies in the $^2P_{\frac{1}{2}}$ State of Tl^{205} and Tl^{203}†

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The atomic beam magnetic resonance method has been used to make a precision measurement of the hyperfine structure separation of the ground state of Tl^{205} and Tl^{203}. The experimentally determined ratio $\Delta\nu^{205}/\Delta\nu^{203}=1.00974\pm 0.00003$ is to be compared with the ratio of the magnetic moments, determined by nuclear induction techniques, of $g_I^{205}/g_I^{203}=1.00986\pm 0.00005$. It is shown that the difference between the two ratios can be accounted for by consideration of effects predicted by Bohr and Weisskopf and by Crawford and Schawlow. In particular, the agreement between theory and experiment can be construed as evidence of the reality of the effects postulated by Crawford and Schawlow, which have not been heretofore directly observed.

INTRODUCTION

A PREVIOUS determination[1] of the ratio of the hyperfine structure separations ($\Delta\nu$) of the $^2P_{\frac{1}{2}}$ ground states of Tl^{205} and Tl^{203} differed by about 40 parts in 10^5 from the ratio of the magnetic moments of the two isotopes. This discrepancy was approximately four times as large as could be accounted for on the basis of any extant theories of hfs anomalies. Unfortunately, the combined experimental uncertainties of the two ratios was 25 parts in 10^5, with the principal contribution to the uncertainties arising from the ratio of the $\Delta\nu$'s. Since an ability to account for a hfs anomaly in Tl would be a stringent test, of a type not heretofore made, of the validity of certain aspects of current theories on the subject, a new and more precise determination of the ratio of the hfs separations has been made by use of atomic beam techniques.

THEORY

A direct determination of the $\Delta\nu$'s of Tl^{203} and Tl^{205}, from observation of the transitions $\Delta F=\pm 1$, would require frequencies that are inconveniently high (~21,300 Mc). The value of the $\Delta\nu$'s may, however, be determined from measurements of the $\Delta F=0$, $\Delta m_F=\pm 1$ transitions $(1, 1\leftrightarrow 1, 0)$ and $(1, 0\leftrightarrow 1, -1)$. The expressions for the frequencies of these lines at an arbitrary magnetic field are[2]

$$f_1=\tfrac{1}{2}\Delta\nu[(1+x)-(1+x^2)^{\frac{1}{2}}+2x(g_J/g_I'-1)^{-1}] \quad \text{for } (1, 1\leftrightarrow 1, 0), \quad (1)$$

$$f_2=\tfrac{1}{2}\Delta\nu[(1+x^2)^{\frac{1}{2}}-(1-x)+2x(g_J/g_I'-1)^{-1}] \quad \text{for } (1, 0\leftrightarrow 1, -1), \quad (2)$$

where $x=(g_J-g_I')\mu_0H/h\Delta\nu$, and g_I' differs slightly from the nuclear g value as determined by nuclear resonance methods because of a partial decoupling of the **L** and **S** vectors in the $^2P_{\frac{1}{2}}$ state.

If the transitions are observed at a fixed magnetic field, then a measurement of the frequencies of the two lines will be sufficient to determine both x and $\Delta\nu$, provided that the value of the ratio g_J/g_I' is known.

The value of $\Delta\nu$ deduced from the Breit-Rabi formula will not be the true hfs separation. Nonvanishing matrix elements of the electron-nucleus interaction operator, which are not diagonal in J, lead to a per-

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[1] Berman, Kusch, and Mann, Phys. Rev. **77**, 140 (1950).

[2] Millman, Rabi, and Zacharias, Phys. Rev. **55**, 384 (1938).

turbation of levels of the same F (in this case $F=1$) in the $P_{\frac{1}{2}}$ and $P_{\frac{3}{2}}$ fine structure levels. Thus, if $\Delta\nu'$ is the true hfs separation and $\Delta\nu$ the hfs deduced from the Breit-Rabi formula, then

$$\Delta\nu'=\Delta\nu-\frac{|\langle J=\frac{1}{2}|H'|J=\frac{3}{2}\rangle|^2}{E_{\frac{3}{2}}-E_{\frac{1}{2}}},$$

where H' is the operator connecting the fine structure levels. This relation can be shown[3] to reduce to the form

$$\Delta\nu'=\Delta\nu(1-k\Delta\nu/\delta),$$

where k is a constant and δ is the separation between the ${}^2P_{\frac{3}{2}}$ and ${}^2P_{\frac{1}{2}}$ levels. This correction has been calculated by Robert Frosch for a number of elements. In the case of thallium it amounts to 15.2 kc for Tl^{205} and 14.9 kc for Tl^{203}. Within the precision of this experiment the effect may be neglected.

For an atom in a state for which $J=\frac{1}{2}$ and $I\geqslant 1$, doublets occur in the hfs spectrum, from which the value of g_J/g_I' can be determined. When $I=\frac{1}{2}$, as in the case of both Tl isotopes, this ratio cannot be determined simply by measurements of lines in the hfs spectrum.

No precision measurement of the g_J value of the ${}^2P_{\frac{1}{2}}$ state of Tl has been reported. However, it may be expected that to an accuracy of 1 part in 10^4 the g_J values of atoms are characteristic of the atomic state rather than of the particular atom. Indeed, the measured values of $g_J({}^2P_{\frac{1}{2}}\text{ In})$ and $g_J({}^2P_{\frac{1}{2}}\text{ Ga})$ reported by Kusch and Foley[4] do agree to within 1 part in 10^5, although the stated precision is not quite as great. For the similar electronic configuration of Tl, it is plausible to assume that $g_J({}^2P_{\frac{1}{2}}\text{ In})=g_J({}^2P_{\frac{1}{2}}\text{ Tl})$. Using the value

$$g_H/g_J({}^2P_{\frac{1}{2}})=-45.6877\times10^{-4} \quad (3)$$

given by Taub and Kusch[5] and the values

$$\begin{aligned} g_I(\text{Tl}^{203})/g_H&=0.571499\pm0.00005,\\ g_I(\text{Tl}^{205})/g_H&=0.577135\pm0.00005 \end{aligned} \quad (4)$$

obtained by Poss[6] by means of nuclear resonance techniques, it is then possible to determine a value of g_J/g_I for each isotope.

Foley[7] has considered the effect of the partial decoupling of the **L** and **S** vectors in the ${}^2P_{\frac{1}{2}}$ state. This effect was shown to reduce the apparent value of the magnetic moment of the nucleus, as deduced from observation of the Zeeman effect of hfs to a value below that determined by nuclear resonance experiments. If g_I' is the apparent value of the nuclear gyromagnetic ratio, then

$$g_I'/g_I=1-\Delta\nu/[g_I(2I+1)6\delta], \quad (5)$$

where δ is the fine structure separation between the ${}^2P_{\frac{1}{2}}$ and ${}^2P_{\frac{3}{2}}$ states. For the case of Tl the magnitude of the values of g_I used in computing the ratio g_J/g_I' must accordingly be reduced by 0.429 percent from the values determined by the nuclear resonance method.

[3] Robert Frosch, private communication.
[4] P. Kusch and H. M. Foley, Phys. Rev. **74**, 250 (1948).
[5] H. Taub and P. Kusch, Phys. Rev. **75**, 1481 (1949).
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METHOD

A field of 3000 gauss, in the vicinity of which most of the measurements of this experiment were made, corresponds to $x\approx0.14$. An expansion of (1) and (2) for small values of x leads to the approximate relationships

$$f_{1,2}=g_F\mu_0H/h\pm\tfrac{1}{4}x^2\Delta\nu+\cdots, \quad (6)$$

$$f_2-f_1=\tfrac{1}{2}x^2\Delta\nu+\cdots, \quad (7)$$

where $g_F\cong g_J/2$, $g_J=\frac{2}{3}$, and $\mu_0/h=1.3998\times10^6\text{ sec}^{-1}$.

Equation (6) indicates that the primary field dependence of the observed lines is through the term $g_F\mu_0H/h$ and is 0.467 Mc/gauss. Dependence of the frequencies on $\Delta\nu$ enters only through a small term quadratic in the field.

The product $x\Delta\nu$, which is proportional to (g_J-g_I'), can in general be determined to a high degree of precision since it is determined from the sum of the two frequencies. However, the same fractional error will occur in $\Delta\nu$ as occurs in the difference of the two frequencies. At $x=0.14$, for example, the absolute error in $\Delta\nu$ will then be about 100 times as great as the absolute error in f_2-f_1. The conditions under which the experiment was performed, and the attainable accuracy of the results were largely determined by the considerations of these implications of Eqs. (6) and (7).

Stringent requirements of stability and homogeneity were necessarily imposed on the magnetic field in which the transitions occurred. The current of 160 amperes, required for the "C" field coils, was supplied by two two-volt submarine batteries. This current had to be maintained constant to within at least 0.0005 ampere for extended periods of time in order that measurements of f_2 and f_1 could be made at values of the fields which did not differ by more than 1 part in 300,000. A greater shift in the field between successive observations of f_1 and f_2 would result in an error $>\frac{1}{2}$ Mc in the value of $\Delta\nu$. The necessary stability was obtained most readily by maintaining a continuously monitored voltage drop across a 0.001-ohm resistor constant, by the introduction of a small manually controlled bucking current.

A small fraction of the absolute magnitude of the "C" field arose from stray fields from the "A" and "B" magnets. As no attempt was made to stabilize these fields against drift, an approximately monotonic drift of 1 part in 60,000 per hour occurred in the magnitude of the "C" field. However, by a proper sequential observation of the transitions the effect of this drift, as well as effects arising from drifts in the "C" field not dependent on fluctuations in the field coil currents, could be averaged out.

The rf region used in this experiment was 1.25 cm long. Since an oven temperature of 750°C is necessary to generate the beam, the theoretical half-widths of the resonances is 20 kc. Although extreme care was taken to obtain as small half-widths as possible, inhomogeneities in the magnetic field increased the half-widths of the lines to about three times the theoretical value.

In view of the sensitivity of the values of $\Delta\nu$ to the accuracy of measurement of the position of the center of the lines, it was essential that no significant overlap between the lines occurred. If any overlap did occur, it would cause an asymmetry in the shapes of the lines which would result in a mismeasurement of the position of the center of the lines. At values of the magnetic field corresponding to $x=0.14$, the separations between the same line for each isotope are $\sim$950 kc for the f_1 lines and $\sim$1050 kc for the f_2 lines. Thus, the resonances observed in this experiment appeared as two completely resolved doublets.

In the experiment reported by Berman, Kusch, and Mann,[1] the $\Delta\nu$ was determined at fields corresponding to an $x\approx 0.1$. At such fields the separations between the lines corresponding to the same transition for each isotope was $\sim$450 kc. The possibility of systematic errors arising from an overlapping of the "tails" of the resonance peaks was, therefore, considerably greater than in this experiment. As can be seen from Eq. (7), at such a value of x, the same absolute error of measurement of f_2-f_1, would result in twice as great an error in $\Delta\nu$ as would result at a value of $x=0.14$.

RESULTS

Six runs, productive of significant data, were obtained in the present set of experiments. Each run was taken at a somewhat different value of the magnetic field. A check on the internal consistency of the data may be obtained from consideration of the fact that the ratio,

$$\frac{f_2^{203}+f_1^{203}}{f_2^{205}+f_1^{205}}=\frac{g_J+g_I'^{203}}{g_J+g_I'^{205}}\equiv\omega,$$

should be a constant for all values of the magnetic field. The values of $\Delta\nu^{205}$, $\Delta\nu^{203}$, and ω calculated in each of these runs are given in Table I.

The value of $\omega=1.0000278\pm0.0000014$ is in good agreement with the value $\omega=1.000026$ which may be calculated from the values of g_I^{205}/g_H, g_I^{203}/g_H, and $g_H/g_J(^2P_{\frac{1}{2}})$ given in Eqs. (3) and (4).

The values of ω given in Table I indicate that both g_I are negative. Since it is known that $|g_I^{205}|>|g_I^{203}|$, a value of $\omega<1$ indicates that both g_I are positive, while a value of $\omega>1$ indicates that both g_I are negative. The possibility that only one of the g_I's is negative may be precluded by the good agreement of the observed and calculated values.

The mean values of $\Delta\nu$ obtained from the experimental data are

$$\Delta\nu(\mathrm{Tl}^{205})=21311.48\pm0.19\times10^6\ \mathrm{sec}^{-1},$$
$$\Delta\nu(\mathrm{Tl}^{203})=21106.06\pm0.49\times10^6\ \mathrm{sec}^{-1},$$

TABLE I. Values of $\Delta\nu^{205}$ and $\Delta\nu^{203}$ obtained from measurements of the transitions $F=0$, $m_F=\pm1$, together with corresponding values of $\omega=(g_J+g_I'^{203})/(g_J+g_I'^{205})$. Frequencies are given in Mc.

$\Delta\nu^{203}$	$\Delta\nu^{205}$	ω
21106.21	21311.05	1.000025
21107.52	21311.32	1.000024
21106.31	21311.32	1.000028
21105.61	21311.64	1.000022
21106.15	21311.26	1.000029
21104.81	21311.53	1.000031
Weighted average of all data		
21106.06±0.49	21311.48±0.19	1.0000278±0.0000014

which lead to a value of the ratio,

$$\Delta\nu^{205}/\Delta\nu^{203}=1.00974\pm0.00003.$$

The ratio of the uncertainties of $\Delta\nu^{203}$ and $\Delta\nu^{205}$ is approximately the same as the ratio of the isotope abundances, indicating that the differential uncertainties may be attributed to the differential intensities of the resonance lines for each isotope.

The quoted errors are purely statistical in nature. A systematic error is possible, as the values given for $\Delta\nu^{205}$ and $\Delta\nu^{203}$ are directly dependant upon the assumed values of g_J/g_I'. However, it may be seen from (1) and (2) that at magnetic fields corresponding to an $x=0.14$, an error as great as 0.1 percent in the assumed value of g_J/g_I' will cause a maximum error in the resultant $\Delta\nu$ of 0.001 percent. An error of this magnitude, if assumed to be in the same directions for both isotopes, will cause an error in the ratio $\Delta\nu^{205}/\Delta\nu^{203}$ of only 1 part in 10^6. Hence any reasonable departure of g_J(Tl) from g_J(In) or an effect arising from an incomplete calculation of g_I'/g_I will have no significant effect on the important datum of this experiment, the ratio $\Delta\nu^{205}/\Delta\nu^{203}$. Similarly, a systematic error of this nature should not appreciably affect the value of the isotope shift $(\Delta\nu^{205}-\Delta\nu^{203})$, which the results of this experiment indicate to be 205.4±0.4 Mc.

Poss[6] has reported that the ratio of the magnetic moments is

$$g_I^{205}/g_I^{203}=1.00986\pm0.00005.$$

Defining

$$\Delta\equiv g_I^{205}/g_I^{203}-\Delta\nu^{205}/\Delta\nu^{203},$$

gives

$$\Delta=0.00012\pm0.00008.$$

DISCUSSION OF RESULTS

First-order theory of hyperfine structure predicts a null value for Δ. The theory is, however, based on two assumptions. The first assumes that the Coulomb attraction between the electron and the nucleus becomes infinite as $r\to0$, while the second assumes that the hfs splittings arise from the interaction between the magnetic field produced by the orbital electrons and a point dipole located at the center. For electrons whose wave functions do not have appreciable values in the nuclear region, the error introduced by these assumptions should be negligible. However, for electrons in the $P_{\frac{1}{2}}$ and $S_{\frac{1}{2}}$ states a nonzero value of Δ may be expected, because the radial wave functions of these states remain finite in the nuclear region. The expected value of Δ would be composed of two parts, Δ_1 and Δ_2. Δ_1 arises from the cutoff of the Coulomb potential, while Δ_2 arises from the finite distribution of the magnetic moment in the nucleus.

If it is assumed that for a nucleus of finite radius the Coulomb field is cut off in some appropriate manner, then the wave functions for electrons in the $P_{\frac{1}{2}}$ and $S_{\frac{1}{2}}$ states will differ markedly within the nuclear region from values corresponding to a pure Coulomb field. Rosenthal and Breit[8] have pointed out, that since the coupling of the electron and the nuclear magnetic moment is proportional to the integral

$$\int \phi_1\phi_2 y^{-2}dy = I, \tag{8}$$

where ϕ_1 and ϕ_2 are the Darwin-Gordon radial functions, and $y=2Zr/a_H$, special consideration of the behavior of the wave functions at values of $r\leqslant r_0$, the nuclear radius, is necessary. It is evident from the form of (8), that the values of the product $\phi_1\phi_2$ are weighted most heavily for small values of y where, for a nucleus of finite size, ϕ_1 and ϕ_2 depart most from their values in a pure Coulomb field. Taking into account the actual perturbed nature of the wave functions at values of $r<r_0$, Rosenthal and Breit were led to a correction factor for nuclear magnetic moments deduced from hfs splittings of the form

$$\Delta\nu \sim g_I(1-KA^{(2\rho-1)/3}), \tag{9}$$

where $\rho=(1-Z^2\alpha^2)^{\frac{1}{2}}$.

The factor K in Eq. (9) is a function of the electronic state, the atomic number, an assumed variation of the nuclear radius of $r_0=1.5\times10^{-13}A^{\frac{1}{3}}$ cm, and the exact form of the electrostatic potential within the nuclear region. It is clear from the form of Eq. (9) that for two isotopes of the same atom a differential correction will exist. Thus

$$\Delta\nu_1/\Delta\nu_2=(g_{I^1}/g_{I^2})\{1-K(A_1{}^{(2\rho-1)/3}-A_2{}^{(2\rho-1)/3})+\cdots\},$$

or

$$\Delta_1=g_{I^1}/g_{I^2}K(A_1{}^{(2\rho-1)/3}-A_2{}^{(2\rho-1)/3}). \tag{10}$$

Crawford and Schawlow[9] have calculated the factor K for two specific models of nuclear charge distribution. For the case of a uniform distribution of charge and negligible nonelectrical forces in the nucleus, they are led to a value of

$$\Delta_1({}^2P_{\frac{1}{2}}\,\mathrm{Tl}^{203,205})=9.3\times10^{-5},$$

while for the case of charge concentrated on the surface of the nucleus and similarly negligible nonelectrical forces within, they are led to a value of

$$\Delta_1({}^2P_{\frac{1}{2}}\,\mathrm{Tl}^{203,205})=10.4\times10^{-5}.$$

Bohr and Weisskopf[10] have considered the interaction of $S_{\frac{1}{2}}$ and $P_{\frac{1}{2}}$ electrons with a magnetic moment that has a finite distribution over the nuclear region. They are led to expect a nonzero value of Δ_2 which depends not only on the size of the nucleus, but also on the intrinsic structure of the nuclear magnetic moments. Assuming that the spin g-factor is that of the proton, and that the orbital momentum is that of the odd particle in the nucleus, they were able to account for observed anomalies[11,12] in K and Rb. Using the same assumptions, the maximum expected value on the basis of this effect alone will be

$$\Delta_2({}^2P_{\frac{1}{2}}\,\mathrm{Tl})=1.4\times10^{-5}.$$

The smallness of the expected effect results from the great similarity in the magnitudes of the magnetic moments of the two isotopes, and also from the fact that the effect will in general be considerably less for $P_{\frac{1}{2}}$ states than for $S_{\frac{1}{2}}$ states.

A combination of the predictions of the theories of Crawford and Schawlow, and of Bohr and Weisskopf leads to

$$\Delta_{\mathrm{calc}}=11\pm1\times10^{-5}.$$

This is to be compared with the experimental value of

$$\Delta_{\mathrm{obs}}=12\pm8\times10^{-5}.$$

The agreement seems to be quite good but, unfortunately, the combined precision to which the two experimental ratios is known precludes any significant discrimination between the proposed nuclear models. However, the general accordance of theory and experiment may be construed as further evidence of the essential validity of current theories of hfs anomalies.

In particular, as the principal effect observed was that of Rosenthal and Breit, this experiment may be considered as an additional indication of the accuracy of the correction factors calculated by Crawford and Schawlow, which are to be applied when magnetic moments are deduced from spectroscopic measurements of hyperfine structure splittings in heavy elements.

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