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Theoretical Analysis of Level Crossing in a ^2P Atomic State

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Two approaches to the analysis of level crossing are presented. A theoretical approach utilizes values of the hyperfine coupling constants to predict the entire level structure of a system, including all level-crossing data of interest. A second approach suggests a procedure for determining hyperfine parameters with reasonable accuracy, based on three or four measured quantities. Results of this procedure in 2 ^2P lithium give the contact, dipole, and orbital hyperfine coupling constants as -9.806 ± 0.117, -1.909 ± 0.034, and 8.638 ± 0.039 MHz, respectively. This work reemphasizes the fact that an accurate determination of magnetic hfs requires three parameters to fully define the three independent magnetic hyperfine interactions.

I. INTRODUCTION

Level crossing is one of the most widely used techniques for investigating the fine and hyperfine structure of atoms. It is based on the fact that often, when an atom is placed in a variable magnetic field, two levels may be tuned to the same energy or "crossed" and that the magnetic field value of the crossing may be detected experimentally. The level structure and thus the crossing fields are clearly related to hyperfine interactions in the particular atomic system. Thus, in theory this technique may be used to determine hyperfine constants. In practice, however, experimentalists have accurately determined the level-crossing data and then obtained values for hyperfine constants from

it in a rather crude way. Theorists, on the other hand, have been content to calculate hyperfine constants when in fact they could have gone on to predict in detail the level-crossing data.

The situation is illustrated quite clearly in the case of the $2\,^2P$ state of lithium. Accurate level-crossing data for this system have been obtained by Brog, Eck, and Wieder.[1] However, their calculation of hyperfine constants from the measured data is based on a two-parameter theory which is not strictly justified from the form of the hyperfine Hamiltonian. Recently, accurate values of the hyperfine constants have been calculated by two different many-body methods.[2-4] These papers, on the other hand, made no attempt to relate their results to quantities which are measurable in a level-crossing experiment. Thus the accurate analysis necessary to make a detailed comparison of these results has not been presented.

It is the purpose of this paper to carry out the theoretical analysis of level crossing in two ways. First, it is possible to obtain all information about level structure and crossings from first principles. Thus based on values for the fine-structure splitting, hyperfine constants, and quadrupole coupling constant, all level-crossing data may be predicted. In the second approach, a procedure is developed to predict hyperfine constants of a ^2P state quite accurately based on three or four experimental parameters. These two procedures are demonstrated by application to the case of $2\,^2P$ lithium.

II. THEORETICAL APPROACH

A. Matrix Elements

In order to reproduce theoretically the level-crossing structure which is observed through experiment, it is necessary to include a number of interactions in the Hamiltonian. The terms required are the spin-orbit interaction, electronic Zeeman interaction, the three magnetic hyperfine interactions, the nuclear Zeeman interaction, and the nuclear electric quadrupole interaction. This may be written symbolically

$$\mathcal{H}' = \mathcal{H}'_{so} + \mathcal{H}'_{eZ} + \mathcal{H}'_{c} + \mathcal{H}'_{orb} + \mathcal{H}'_{dip} + \mathcal{H}'_{NZ} + \mathcal{H}'_{Q}\,, \tag{1}$$

where $\mathcal{H}'_c$, $\mathcal{H}'_{orb}$, and $\mathcal{H}'_{dip}$ are the contact, orbital, and dipolar hyperfine interactions, respectively. The explicit expressions for these interactions are

$$\mathcal{H}'_{so} = \zeta \vec{L}\cdot\vec{S}\,, \tag{2}$$

$$\mathcal{H}'_{eZ} = \mu_L \vec{L}\cdot\vec{H} + \mu_S \vec{S}\cdot\vec{H}\,, \tag{3}$$

$$\mathcal{H}'_{c} = \frac{8\pi}{3}\frac{\mu_S\mu_I}{a^3}\vec{I}\cdot\sum_{i=1}^{N_e}\vec{s}_i\,\delta(\vec{r}_i)\,, \tag{4}$$

$$\mathcal{H}'_{orb} = 2\frac{\mu_L\mu_I}{a^3}\vec{I}\cdot\sum_{i=1}^{N_e}\frac{\vec{l}_i}{r_i^3}\,, \tag{5}$$

$$\mathcal{H}'_{dip} = \frac{\mu_S\mu_I}{a^3}\vec{I}\cdot\sum_{i=1}^{N_e}\left(\frac{3(\vec{s}_i\cdot\vec{r}_i)}{r_i^5}\vec{r}_i - \frac{\vec{s}_i}{r_i^3}\right), \tag{6}$$

$$\mathcal{H}'_{NZ} = -\mu_I\vec{I}\cdot\vec{H}\,, \tag{7}$$

$$\mathcal{H}'_{Q} = -\sum_{i=1}^{N_e}\sum_{p=1}^{N_p}\frac{r_p^2}{r_i^3}P_2(\cos\theta_{ip})\,. \tag{8}$$

Here ζ is the fine-structure splitting, $\vec{\mu}_L = \mu_L\vec{L}$ is the electron orbital magnetic moment, $\vec{\mu}_S = \mu_S\vec{S}$ is the electron spin magnetic moment, $\mu_I = \mu_I\vec{I}$ is the nuclear magnetic moment, a is the Bohr radius in the appropriate system, and N_e and N_p are the number of electrons and protons in the atom, respectively. These quantities may be further expressed in terms of fundamental constants. If the angular momenta are measured in terms of $\hbar$, then the magnetic moments expressed in terms of Bohr magnetons μ_B or nuclear magnetons μ_n are

$$\mu_L = g_M\mu_B\,, \tag{9}$$

$$\mu_S = g_S\mu_B\,, \tag{10}$$

$$\mu_I = (\mu_N/I)\mu_n\,, \tag{11}$$

where $g_M = 1 - M_e/M_I$ with M_I the isotopic mass, $g_S = 2.002\,319$, and μ_N the nuclear magnetic moment in nuclear magnetons. Also, in terms of the Bohr radius for hydrogen a_0, we have

$$a = (1 + m_e/M_I)a_0\,. \tag{12}$$

For an atom in a magnetic field, the quantities L^2, S^2, and I^2 are constant and independent of the field. Thus the eigenfunctions of $\mathcal{H}'$ can be expressed as linear combinations of M_L, M_S, M_I states for any value of the field. Therefore, it is necessary to know the matrix elements of the interactions in $\mathcal{H}'$ in the M_L, M_S, M_I representation. These matrix elements may be obtained by expressing each of the operators in terms of irreducible tensor components and applying the Wigner-Eckart theorem. The reduced matrix elements may then be expressed in terms of commonly used physical quantities by evaluating the general matrix element in the special case $M_L = L$, $M_S = S$, $M_I = I$.

Evaluating the matrix elements is trivial for the two Zeeman interactions. Assuming the direction of the z axis is defined by the external field $\vec{H}$, the only nonvanishing matrix elements are

$$\langle M_L, M_S, M_I|\mathcal{H}'_{eZ}|M_L, M_S, M_I\rangle = (g_M\mu_B M_L + g_S\mu_B M_S)H\,, \tag{13}$$

$$\langle M_L, M_S, M_I|\mathcal{H}'_{NZ}|M_L, M_S, M_I\rangle = -(\mu_N/I)\mu_n M_I H\,. \tag{14}$$

The nonvanishing matrix elements of $\mathcal{H}'_{so}$ are also easily shown to be

$$\langle M_L, M_S, M_I | \mathcal{H}'_{so} | M_L, M_S, M_I \rangle = M_L M_S \zeta , \quad (15)$$

$$\langle M_L \pm 1, M_S \mp 1, M_I | \mathcal{H}'_{so} | M_L, M_S, M_I \rangle = \tfrac{1}{2} L_\pm S_\mp \zeta , \quad (16)$$

where

$$L_\pm = [(L \mp M_L)(L \pm M_L + 1)]^{1/2}$$

and likewise for $S_\pm$.

The four remaining interactions are somewhat more complex, each being a scalar product of tensor operators. Using the Wigner-Eckart theorem and the tensor product formula[5]

$$T^M_J = (-1)^{j_2 - j_1 - M} (2J+1)^{1/2} \times \sum_{m_1, m_2} \begin{pmatrix} j_1 & j_2 & J \\ m_1 & m_2 & -M \end{pmatrix} T^{m_1}_{j_1} T^{m_2}_{j_2} ; \quad (17)$$

these interactions may be expressed in terms of reduced matrix elements. In the case of $\mathcal{H}'_c$ this leads to

$$\langle M_L, M_S, M_I, | \mathcal{H}'_c | M_L, M_S, M_I \rangle = 4 M_I M_S \frac{(I || \vec{\mu}_I || I)(S || \vec{H}_c || S)}{[(2I)_3 (2S)_3]^{1/2}} , \quad (18)$$

$$\langle M_L, M_S \pm 1, M_I \mp 1 | \mathcal{H}'_c | M_L, M_S, M_I \rangle = 2 S_\pm I_\mp \frac{(I || \vec{\mu}_I || I)(S || \vec{H}_c || S)}{[(2I)_3 (2S)_3]^{1/2}} \quad (19)$$

where Pockhammer's symbol $(a)_k$ is defined by[6]

$$(a)_0 = 1, \quad (a)_k = a(a+1) \cdots , (a+k-1) , \quad (20)$$

and $\mathcal{H}'_c$ has been expressed in the form

$$\mathcal{H}'_c = \vec{\mu}_I \cdot \vec{H}_c . \quad (21)$$

The reduced matrix elements are evaluated by using the empirical hfs Hamiltonian

$$\mathcal{H}'_i = a_{i, I, J} \vec{I} \cdot \vec{J} . \quad (22)$$

Here the subscript i designates one of the three magnetic hyperfine interactions. In particular for $i = c$ we have

$$\langle J = L+S, M_J = J, M_I = I | a_{c, I, J} \vec{I} \cdot \vec{J} | J = L+S, M_J = J, M_I = I \rangle = a_{c, I, L+S} I (L+S) \quad (23)$$

and

$$\langle M_L = L, M_S = S, M_I = I | \mathcal{H}_c | M_L = L, M_S = S, M_I = I \rangle = 4 I S \frac{(I || \vec{\mu}_I || I)(S || \vec{H}_c || S)}{[(2I)_3 (2S)_3]^{1/2}} . \quad (24)$$

Since these two matrix elements must be equal, the reduced matrix elements are found to be

$$\frac{(I || \vec{\mu}_I || I)(S || \vec{H}_c || S)}{[(2I)_3 (2S)_3]^{1/2}} = \frac{L+S}{4S} a_{c, I, L+S} . \quad (25)$$

With this result the nonvanishing matrix elements of $\mathcal{H}'_c$ are given by

$$\langle M_L, M_S, M_I | \mathcal{H}'_c | M_L, M_S, M_I \rangle = M_S M_I (L+S) S^{-1} a_{c, I, L+S} , \quad (26)$$

$$\langle M_L, M_S \pm 1, M_I \mp 1 | \mathcal{H}'_c | M_L, M_S, M_I \rangle = S_\pm I_\mp \tfrac{1}{2} (L+S) S^{-1} a_{c, I, L+S} . \quad (27)$$

The corresponding procedure for $\mathcal{H}'_{orb}$ and $\mathcal{H}'_{dip}$ gives

$$\langle M_L, M_S, M_I | \mathcal{H}'_{orb} | M_L, M_S, M_I \rangle = M_L M_I (L+S) L^{-1} a_{orb, I, L+S} , \quad (28)$$

$$\langle M_L \pm 1, M_s, M_I \mp 1 | \mathcal{H}'_{orb} | M_L, M_S, M_I \rangle = L_\pm I_\mp \tfrac{1}{2} (L+S) L^{-1} a_{orb, I, L+S} , \quad (29)$$

$$\langle M_L, M_S, M_I | \mathcal{H}'_{dip} | M_L, M_S, M_I \rangle = [3 M_L^2 - L(L+1)] M_S M_I \times (L+S)[L(2L-1)S]^{-1} a_{dip, I, L+S} , \quad (30)$$

$$\langle M_L \pm 1, M_S \mp 1, M_I | \mathcal{H}'_{dip} | M_L, M_S, M_I \rangle = \tfrac{3}{4} (2 M_L \pm 1) L_\pm S_\mp M_I \times (L+S)[L(2L-1)S]^{-1} a_{dip, I, L+S} , \quad (31)$$

$$\langle M_L \pm 2, M_S \mp 1, M_I \mp 1 | \mathcal{H}'_{dip} | M_L, M_S, M_I \rangle = \tfrac{3}{4} L_\pm^2 S_\mp I_\mp (L+S)[L(2L-1)S]^{-1} a_{dip, I, L+S} , \quad (32)$$

$$\langle M_L \pm 1, M_S, M_I \mp 1 | \mathcal{H}'_{dip} | M_L, M_S, M_I \rangle = \tfrac{3}{4} (2 M_L \pm 1) L_\pm M_S I_\mp \times (L+S)[L(2L-1)S]^{-1} a_{dip, I, L+S} , \quad (33)$$

$$\langle M_L, M_S \pm 1, M_I \mp 1 | \mathcal{H}'_{dip} | M_L, M_S, M_I \rangle = -\tfrac{1}{4} [3 M_L^2 - L(L+1)] \times S_\pm I_\mp (L+S)[L(2L-1)S]^{-1} a_{dip, I, L+S} , \quad (34)$$

where

$$L_\pm^2 = [(L \mp M)(L \pm M + 1)(L \mp M - 1)(L \pm M + 2)]^{1/2} .$$

The same procedure may be followed in evaluating matrix elements of the electric quadrupole interaction. The reduced matrix elements may be evaluated by comparing with a matrix element of the spin Hamiltonian[7]

$$\mathcal{H}'_Q = \frac{b}{2I(2I-1)J(2J-1)}\left[3(\vec{I}\cdot\vec{J})^2+\tfrac{3}{2}(\vec{I}\cdot\vec{I})(\vec{J}\cdot\vec{J})\right] \tag{35}$$

in the special case $J=L+S$, $M_J=J$, $M_I=I$. The resulting matrix elements are

$$\langle M_L,\ M_S,\ M_I|\mathcal{H}'_Q|M_L,\ M_S,\ M_I\rangle = \tfrac{1}{4}b\frac{[3M_L^2-L(L+1)][3M_I^2-I(I+1)]}{L(2L-1)I(2I-1)}, \tag{36}$$

$$\langle M_L\pm1,\ M_S,\ M_I\pm1|\mathcal{H}'_Q|M_L,\ M_S,\ M_I\rangle = \tfrac{3}{8}b\frac{(2M_L\pm1)(2M_I\mp1)L_\pm I_\mp}{L(2L-1)I(2I-1)}, \tag{37}$$

$$\langle M_L\pm2,\ M_S,\ M_I\mp2|\mathcal{H}'_Q|M_L,\ M_S,\ M_I\rangle = \tfrac{3}{8}b\frac{L_\pm^2 I_\mp^2}{L(2L-1)I(2I-1)}. \tag{38}$$

B. Level Structure

In general, the theoretical calculation of level structure is quite straightforward. The $\mathcal{H}'$ matrix in the M_L, M_S, M_I representation is calculated and its eigenvalues obtained as a function of the magnetic field H. From this data any quantities of interest may be deduced or the entire level structure may be plotted. When carried out on a computer, this is a trivial problem requiring mainly the diagonalization of an 18×18 or 24×24 matrix for the ^2P states of Li^6 and Li^7, respectively.

A simple computer program has been written to carry out this procedure. This program requires as input the parameters ζ, μ_L, μ_S, μ_I, $a_{c,I,L+S}$, $a_{orb,I,L+S}$, $a_{dip,I,L+S}$, b, L, S, and I. It then evaluates and diagonalizes the $\mathcal{H}'$ matrix. The program will produce either a table of eigenvalues of $\mathcal{H}'$ as a function of magnetic field or it will determine a crossing or anticrossing field value to a specified accuracy.

The level structure of a ^2P state is well known at zero magnetic field, very low field, or very high field. The region of interest, however, is the area in between where hyperfine levels become rearranged into different groups and crossings occur. By plotting all eigenstates as a function of H, the entire level structure may be displayed. The over-all picture is shown in Fig 1. This is a plot of the fine-structure levels from zero field out to the strong-field region. Incorporating the hfs into this picture for the most part simply replaces each of the fine-structure lines by a closely spaced group of $2I+1$ lines. In other words, if one were to examine in greatly expanded scale the actual structure of one of the lines in Fig. 1 at almost any point, one would observe a set of $2I+1$ lines either converging linearly toward or diverging linearly from the fine-structure line as required by $\mathcal{H}_{NZ}$.

The points of particular interest, then, in Fig. 1 are those places where the hfs behavior is different from this; all such points are indicated on Fig. 1. There are three classes of situations. The first is the weak-field area including the low-field transition from (F, M_F) states to (M_J, M_I) states. These regions are from 0 to about 10 G for the $J=\frac{3}{2}$ level and up to about 100 G for the $J=\frac{1}{2}$ level. These regions, marked $T1$ and $T2$, respectively, on Fig. 1, have been examined in detail in the past[8] and the weak-field level structure of a ^2P state is well understood. The second type of special case in Fig. 1 is a point where the hyperfine levels within a fine-structure line are interchanged. There are three instances of this in a ^2P state and they are marked $I1$, $I2$, and $I3$ in Fig. 1. An example of this behavior is shown in detail in Fig. 2 for $I=\frac{3}{2}$ where the deviations of the hfs levels from the fine-structure line are plotted against H near the point $I1$. The final points of interest are the actual crossings of fine-structure levels. There are two of these in a ^2P atomic state as shown on Fig. 1 at $C1$ and $C2$ and examples of these are pictured in detail in Figs. 3 and 4. As seen in Fig. 3, the levels cross uneventfully at $C1$. However, at $C2$ there are several pairs of levels which interact, resulting in the anticrossings seen as deflected pairs of lines in Fig. 4. Experimentally observable quantities are indicated on Figs. 3 and 4.

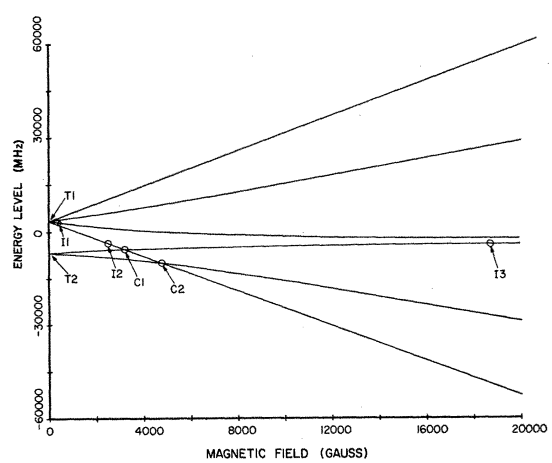

FIG. 1. Fine structure of Li 2 ^2P from zero-field to the strong-field region with points of interest indicated.

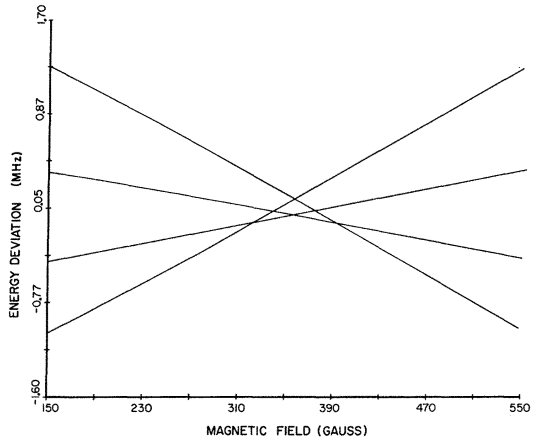

FIG. 2. Hyperfine structure of the $J=\frac{3}{2}$, $M_J=-\frac{1}{2}$ level in Li^7 2 ^2P at $I1$ of Fig. 1 plotted as deviations from the fine-structure line.

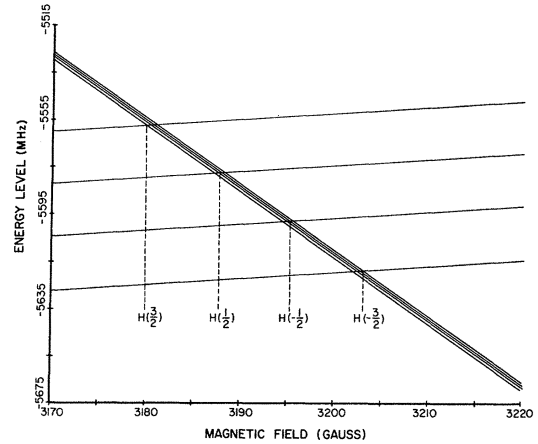

FIG. 3. Crossings of hyperfine levels at $C1$ in 2 ^2P Li^7 with crossing field values $H(M_I)$ of observable crossings indicated.

C. Application to 2 ^2P Lithium

As an example of a calculation of level-structure results in a ^2P state, we have carried out the analysis described above for the case of lithium in the 2 ^2P state. The values used for the hfs parameters are those previously obtained by the Brueckner-Goldstone many-body perturbation theory.[2] These and the values used for the other parameters required by the computer program are listed for reference in Table I. Based on these input values we have calculated all quantities which might be observable in 2 ^2P lithium.

The quantities of experimental interest at hfs level crossings such as those at H_{C1} are the field intervals between crossings of hyperfine components with the same M_I value. There are four such crossings at H_{C1} in Li^7 and three in Li^6. Our calculated field intervals between these crossings are given in Table II. The measured intervals of Brog, Eck, and Wieder[1] in Li^7 corrected for overlap of the signals are listed for comparison. As seen in Table II, all calculated results agree with experiment to within 1%, the stated accuracy of the hyperfine constants used.[2]

At H_{C2}, where a number of anticrossings occur, the quantities of interest are the field intervals between anticrossings as well as the interaction matrix element V which causes the anticrossing. There are three anticrossings at H_{c2} in Li^7 and two in Li^6. The corresponding field intervals and matrix elements are given in Table III. The anticrossing signals are not well resolved experimentally in 2 ^2P lithium, making it difficult to determine the anticrossing field intervals and matrix elements. Recently Eck and Smith[13] have developed a sophisticated computer analysis of the measured data which will yield values for these quantities. A first estimate from this analysis indicates that the field interval between the two anticrossings in Li^6 is about 3–4 G. This is somewhat larger than our calculated value of 2.9 G but could easily come into good agreement as their procedure is perfected. Their preliminary value for the average interaction matrix element appears to be fairly stable at (6.783 ± 0.069) MHz. This is in quite good agreement with our average value of 6.703 MHz.

In Table IV we have given the field values of the hyperfine crossings in the three hfs inversions indicated at $I1$, $I2$, and $I3$ in Fig. 1. It would be very interesting if an experimental technique could be devised to observe these crossings. They could be valuable in providing additional checks on the determined magnetic hyperfine and quadrupole coupling constants.

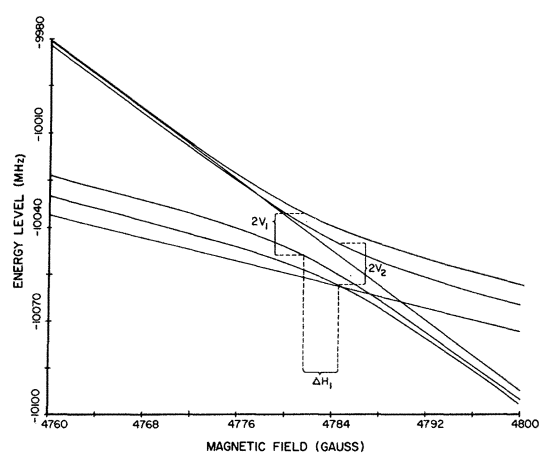

FIG. 4. Crossings and anticrossings of hyperfine levels in 2 ^2P Li^6 at $C2$. Observable anticrossing interaction matrix elements V_i and crossing field interval ΔH_i are indicated.

TABLE I. Parameter values used in calculation of level structure of Li 2 ^2P.

	Units	Ref.	$I=1$	$I=\frac{3}{2}$
μ_L	μ_B	a	0.999 908 8	0.999 921 8
μ_S	μ_B	b	2.002 319 3	2.002 319 3
μ_I	μ_n	c	2.170 903	0.822 019
ζ	MHz	d	6701.84	6702.16
$a_{c,I,3/2}$	MHz	e	−3.6269	−9.5788
$a_{\mathrm{dip},I,3/2}$	MHz	e	−0.7180	−1.8964
$a_{\mathrm{orb},I,3/2}$	MHz	e	3.2838	8.6727
b	MHz	f	−0.003 8541	−0.20276

[a]$\mu_L = g_M \mu_B$, where $g_M = 1 - m_e/M_I$. m_e and μ_B values used are from Ref. 9 while isotopic mass values are from Ref. 10.

[b]$\mu_S = g_S \mu_B$, where g_s and μ_B values used are from Ref. 9.

[c]$\mu_I = (\mu_N/I)$, where μ_n is from Ref. 9 and μ_N values for Li^6 and Li^7 are from Ref. 11.

[d]Values used are from Ref. 1.

[e]Values used are from Ref. 2.

[f]$b \doteq qQ$, where q is from Ref. 2 and Q is from Ref. 12.

III. EXPERIMENTAL APPROACH: PERTURBATION THEORY ANALYSIS

Experimentalists could utilize the same procedure described above to obtain values for the hyperfine parameters from measured data. In this case they would start with some trial values of a_{IIJ} and b and employ the procedure iteratively to obtain the best fit to the level-crossing data. Good trial values might be from Hartree-Fock calculations or more sophisticated results where available. Alternatively, an experimentalist can utilize an approximate analysis to make a direct calculation of the hfs parameters. These may be either for use as trial values for the above procedure or if the approximate analysis is reasonably sophisticated they would be of value in themselves.

We present here a perturbation procedure which is a generalization of that utilized by Brog, Eck, and Wieder[1] and is expected to yield hfs parameters of useful accuracy. The error introduced by the analysis should not be more than a few percent.

The fact that three independent parameters are required to characterize the three magnetic hyperfine interactions dictates that at least three equations relating the hfs parameters to measured quantities must be known. The electric quadrupole interaction requires a fourth independent parameter; however, the procedure for treating it depends on its relative magnitude. If the quadrupole interaction is comparable to the three magnetic hyperfine interactions, then four equations must be obtained and solved for the four unknown parameters. In cases such as Li 2 ^2P where the quadrupole interaction is significantly smaller than the magnetic hfs, a stepwise procedure may be used to obtain b after first solving for a_{IIJ}.

The analysis will be developed here as specifically applicable to the current experimental situation in 2 ^2P lithium. It should be emphasized, however, that the same approach is generally applicable in any atomic system. Some measurable quantities which derive from the hfs parameters are the hyperfine coupling constant in a specific J state, the interaction matrix element between two anticrossing hyperfine levels, and the average field interval between hyperfine crossings or anticrossings in a fine-structure crossing. The theoretical quantities which are used here and which fully define the hfs are $a_{c,3/2,3/2}$, $a_{\mathrm{orb},3/2,3/2}$, $a_{\mathrm{dip},3/2,3/2}$, and b.

In the 2 ^2P state of lithium the two allowed J states are $J=\frac{3}{2}$ and $J=\frac{1}{2}$. In the $J=\frac{3}{2}$ state, a_{IJ} has

TABLE II. Field intervals between hfs crossings at H_{C1} in Li^6 and Li^7.

I	M_I	$\Delta H(M_I) = H(M_I - 1) - H(M_I)$ (G) Present calculation	Expt[a]
$\frac{3}{2}$	$\frac{3}{2}$	7.6003	7.5416 ± 0.0078
$\frac{3}{2}$	$\frac{1}{2}$	7.6490	7.5957 ± 0.0070
$\frac{3}{2}$	$-\frac{1}{2}$	7.7107	7.6379 ± 0.0127
$\frac{3}{2}$	av	7.6533	7.5916 ± 0.0164
1	1	2.9111	
1	0	2.206	
1	av	2.9159	

[a]See Ref. 1. The values quoted above are those from Ref. 1 less the "off-diagonal hfs corrections" quoted therein.

TABLE III. Field intervals and interaction matrix elements of anticrossings at H_{C2} in Li^6 and Li^7.

	Li^6	Li^7
ΔH_1	2.8686 G	7.7571 G
ΔH_2		7.6116 G
V_1	6.6996 MHz	21.5542 MHz
V_2	6.7067 MHz	25.0273 MHz
V_3		21.7941 MHz

TABLE IV. Field values of hfs crossings at H_{I1}, H_{I2}, and H_{I3} in Li^6 and Li^7 in G.

		$I1$	$I2$	$I3$
Li^7	H_1	320.3027	2458.5962	18 797.5097
Li^7	H_2	338.5100	2510.2758	18 810.1120
Li^7	H_3	356.5468	2562.6140	18 822.6596
Li^7	H_4	356.9958	2563.0157	18 824.3199
Li^7	H_5	375.0787	2616.3140	18 837.6418
Li^7	H_6	393.4256	2670.8920	18 852.5679
Li^6	H_1	355.7966	2535.5376	18 816.9852
Li^6	H_2	359.8517	2548.4474	18 817.9094
Li^6	H_3	363.9010	2561.4609	18 818.8349

not been measured. However, the value of $a_{I,1/2}$ is known experimentally, so it can serve as one of the three measured quantities. The relation between $a_{i,3/2,1/2}$ and the $a_{i,3/2,3/2}$ can be found simply by evaluating the diagonal matrix elements of the appropriate hyperfine interaction in both the J, M_J representation and the M_L, M_S representation and equating them. Thus we find

$$a_{c,3/2,1/2} = -a_{c,3/2,3/2}\ , \tag{39}$$

$$a_{\mathrm{dip},3/2,1/2} = -10 a_{\mathrm{dip},3/2,3/2}\ , \tag{40}$$

$$a_{\mathrm{orb},3/2,1/2} = 2 a_{\mathrm{orb},3/2,3/2}\ . \tag{41}$$

And the desired relation is

$$\begin{aligned} a_{3/2,1/2} &= a_{c,3/2,1/2} + a_{\mathrm{dip},3/2,1/2} + a_{\mathrm{orb},3/2,1/2} \\ &= -a_{c,3/2,3/2} - 10 a_{\mathrm{dip},3/2,3/2} + 2 a_{\mathrm{orb},3/2,3/2}\ . \end{aligned} \tag{42}$$

This relation between $a_{3/2,1/2}$ and the $a_{i,3/2,3/2}$ is exact. Unfortunately simple algebraic relations between other measurable quantities and the hfs parameters can only be obtained approximately. In this approach the two approximations used are (a) treating the hyperfine interactions as perturbations to the spin-orbit and electronic Zeeman effects and (b) neglecting the effect on the wave function of variation in the magnetic field over the range of hfs crossings in a fine-structure crossing. Both of these approximations are well justified since they represent changes of not more than 1%.

The perturbation procedure requires the complete analysis of the spin-orbit and electronic Zeeman level structure which is a straightforward problem. Solving for the eigenvalues and eigenvectors of this zero-order Hamiltonian requires at most the diagonalization of a 2×2 matrix. The eigenvalues $E_0(J, M_J, M_I)$ thus obtained are

$$E_0(\tfrac{3}{2}, \tfrac{3}{2}, M_I) = [\zeta/2 + (g_M + g_S/2)\mu_B H]M_I\ , \tag{43}$$

$$\begin{aligned} E_0(\tfrac{3}{2}, \tfrac{1}{2}, M_I) = \{ &-\zeta/2 + g_M \mu_B H \\ &+ [9\zeta^2/4 + \zeta(g_S - g_M)\mu_B H \\ &+ (g_S - g_M)^2 \mu_B^2 H^2]^{1/2}\} M_I/2\ , \end{aligned} \tag{44}$$

$$\begin{aligned} E_0(\tfrac{3}{2}, -\tfrac{1}{2}, M_I) = \{ &-\zeta/2 - g_M \mu_B H \\ &+ [9\zeta^2/4 - \zeta(g_S - g_M)\mu_B H \\ &+ (g_S - g_M)^2 \mu_B^2 H^2]^{1/2}\} M_I/2\ , \end{aligned} \tag{45}$$

$$E_0(\tfrac{3}{2}, -\tfrac{3}{2}, M_I) = [\zeta/2 - (g_M + g_S/2)\mu_B H]M_I\ , \tag{46}$$

$$\begin{aligned} E_0(\tfrac{1}{2}, \tfrac{1}{2}, M_I) = \{ &-\zeta/2 + g_M \mu_B H \\ &- [9\zeta^2/4 + \zeta(g_S - g_M)\mu_B H \\ &+ (g_S - g_M)^2 \mu_B^2 H^2]^{1/2}\} M_I/2\ . \end{aligned} \tag{47}$$

$$\begin{aligned} E_0(\tfrac{1}{2}, -\tfrac{1}{2}, M_I) = \{ &-\zeta/2 - g_M \mu_B H \\ &- [9\zeta^2/4 - \zeta(g_S - g_M)\mu_B H \\ &+ (g_S - g_M)^2 \mu_B^2 H^2]^{1/2}\} M_I/2\ , \end{aligned} \tag{48}$$

and the eigenvectors $\Psi_0(J, M_J, M_I)$ may be expressed in terms of M_L, M_S, M_I basis states as

$$\Psi_0(\tfrac{3}{2}, \tfrac{3}{2}, M_I) = |1, \tfrac{1}{2}, M_I\rangle\ , \tag{49}$$

$$\begin{aligned} \Psi_0(\tfrac{3}{2}, \tfrac{1}{2}, M_I) = {} & A(H)|0, \tfrac{1}{2}, M_I\rangle \\ & + B(H)|1, -\tfrac{1}{2}, M_I\rangle\ , \end{aligned} \tag{50}$$

$$\begin{aligned} \Psi_0(\tfrac{3}{2}, -\tfrac{1}{2}, M_I) = {} & C(H)|-1, \tfrac{1}{2}, M_I\rangle \\ & + D(H)|0, -\tfrac{1}{2}, M_I\rangle\ , \end{aligned} \tag{51}$$

$$\Psi_0(\tfrac{3}{2}, -\tfrac{3}{2}, M_I) = |-1, -\tfrac{1}{2}, M_I\rangle\ , \tag{52}$$

$$\begin{aligned} \Psi_0(\tfrac{1}{2}, \tfrac{1}{2}, M_I) = {} & -B(H)|0, \tfrac{1}{2}, M_I\rangle \\ & + A(H)|1, -\tfrac{1}{2}, M_I\rangle\ , \end{aligned} \tag{53}$$

$$\begin{aligned} \Psi_0(\tfrac{1}{2}, -\tfrac{1}{2}, M_I) = {} & -D(H)|-1, \tfrac{1}{2}, M_I\rangle \\ & + C(H)|0, -\tfrac{1}{2}, M_I\rangle\ . \end{aligned} \tag{54}$$

At zero magnetic field these coefficients are easily found to be

$$A(0) = D(0) = (\tfrac{2}{3})^{1/2}\ , \tag{55}$$

$$B(0) = C(0) = (\tfrac{1}{3})^{1/2}\ . \tag{56}$$

The fields at which these fine-structure levels cross may be determined by equating the eigenvalues for the appropriate levels and solving for H. The crossing at H_{C1} is between the $J=\frac{3}{2}$, $M_J=-\frac{3}{2}$ and the $J=\frac{1}{2}$, $M_J=\frac{1}{2}$ lines. Thus we have the crossing equation

$$E_0(\tfrac{3}{2}, -\tfrac{3}{2}, M_I) - E_0(\tfrac{1}{2}, \tfrac{1}{2}, M_I) = 0\ , \tag{57}$$

and solving for H_{C1} we obtain

$$H_{C1} = \frac{g_M + \frac{1}{2}g_S}{g_M(g_M + g_S)} \frac{\zeta}{\mu_B}\ . \tag{58}$$

In the same way, the crossing field at $C2$ is obtained from

$$E_0(\tfrac{3}{2}, -\tfrac{3}{2}, M_I) - E_0(\tfrac{1}{2}, -\tfrac{1}{2}, M_I) = 0\ , \tag{59}$$

resulting in

$$H_{C2} = \frac{g_M + \frac{1}{2}g_S}{g_M g_S}\, \zeta/\mu_B\ . \tag{60}$$

To the accuracy inherent in this approximate procedure, these two expressions may be replaced by

$$H_{C1} = 2\zeta/3\mu_B \,, \tag{61}$$

$$H_{C2} = \zeta/\mu_B \,. \tag{62}$$

The coefficients in the eigenvectors are found at these field values to be

$$A(H_{C1}) = \sqrt{\tfrac{9}{11}} \,, \tag{63}$$

$$B(H_{C1}) = \sqrt{\tfrac{2}{11}} \,, \tag{64}$$

$$C(H_{C1}) = 6/[73-(73)^{1/2}]^{1/2} \tag{65}$$

$$D(H_{C1}) = [37-(73)^{1/2}]^{1/2}/[73-(73)^{1/2}]^{1/2} \,, \tag{66}$$

$$A(H_{C2}) = 2/[17-3(17)^{1/2}]^{1/2} \,, \tag{67}$$

$$B(H_{C2}) = [13-3(17)^{1/2}]^{1/2}/[17-3(17)^{1/2}]^{1/2} \,, \tag{68}$$

$$C(H_{C2}) = \sqrt{\tfrac{2}{3}} \,, \tag{69}$$

$$D(H_{C2}) = \sqrt{\tfrac{1}{3}} \,. \tag{70}$$

With these zero-order eigenvalues and eigenvectors, it is possible to calculate simple relations between measurable quantities and the $a_{i,I,J}$ using first-order perturbation theory. In the case of lithium 2 ^2P there are two additional quantities that have been measured to good precision. These are the average interaction matrix element of the anticrossing in Li^6 and the average field interval between hfs crossings at H_{C1} in Li^7. Using the approximations stated above, expressions may be obtained relating these two quantities to the theoretical parameters.

The average anticrossing interaction matrix element at H_{C2} in Li^6 is defined by

$$V_{\text{av}} = (1/2I) \sum_{M_I=-I}^{I-1} \times \langle \Psi(\tfrac{3}{2}-\tfrac{3}{2},\ M_I+1) | \mathcal{H}' | \Psi(\tfrac{1}{2},\ -\tfrac{1}{2},\ M_I) \rangle \,. \tag{71}$$

This may be evaluated using the zero-order wave functions and the M_L, M_S, M_I matrix elements given above, and reduces to

$$V_{\text{av}} = \sqrt{\tfrac{3}{2}}\,(-a_{\text{c},1,3/2} + 2a_{\text{dip},1,3/2} + a_{\text{orb},1,3/2}) \,, \tag{72}$$

to the approximation used here,

$$a_{i,1,3/2} = \tfrac{3}{2}(\mu_N^6/\mu_N^7)\, a_{i,3/2,3/2} \,, \tag{73}$$

so that Eq. (72) may be rewritten

$$V_{\text{av}} = (\tfrac{3}{2})^{3/2}(\mu_N^6/\mu_N^7)(-a_{\text{c},3/2,3/2} + 2a_{\text{dip},3/2,3/2} + a_{\text{orb},3/2,3/2}) \,. \tag{74}$$

In order to obtain an expression for the average field interval between crossings, it is necessary to first determine the crossing fields. Hyperfine crossing field values may be found by solving for H in the hfs level-crossing equation to first order in the perturbation

$$\sum_{i=0}^{1} [E_i(\tfrac{3}{2},\ -\tfrac{3}{2},\ M_I) - E_i(\tfrac{1}{2},\ \tfrac{1}{2},\ M_I)] = 0 \,. \tag{75}$$

The subscript i here refers to the order in the perturbation. For each value of M_I Eq. (75) may be solved for the crossing field $H(M_I)$ by the same procedure and approximations used for Eq. (71). The average field interval between crossings is found from

$$(\Delta H)_{\text{av}} = (1/2I) \sum_{M_I=-I+1}^{I} [H(M_I-1) - H(M_I)] \,, \tag{76}$$

giving

$$(\Delta H)_{\text{av}} = \frac{3a_{\text{c},3/2,3/2} - 15a_{\text{dip},3/2,3/2} + 15a_{\text{orb},3/2,3/2}}{(10g_M + g_S)\mu_B} \,. \tag{77}$$

It should be noted here that the field interval between crossings depends only on the hfs parameters and the quadrupole interaction. In the average over M_I the quadrupole contribution vanishes. In other words, the average field interval is dependent only on the magnetic hfs parameters while deviations from the average are dependent also on the quadrupole coupling constant.

Equations (42), (74), and (77) comprise three equations in the three unknowns $a_{i,3/2,3/2}$ for i = c, dip, orb. Inverting these relations gives

$$\begin{aligned} a_{\text{c},3/2,3/2} &= (40C_1 + 14C_2 + 15C_3)/69 \,, \\ a_{\text{dip},3/2,3/2} &= (-7C_1 + C_2 + 6C_3)/69 \,, \\ a_{\text{orb},3/2,3/2} &= (-5C_1 + 4C_2 + C_3)/23 \,, \end{aligned} \tag{78}$$

where

$$\begin{aligned} C_1 &= -(\tfrac{2}{3})^{3/2}(\mu_N^7/\mu_N^6)\, V_{\text{av}} \,, \\ C_2 &= (10g_M + g_S)\,\mu_B(\Delta H)_{\text{av}}/3 \\ C_3 &= -a_{3/2,1/2} \,. \end{aligned} \tag{79}$$

Measured values for the three experimental parameters are[1,4]

$$V_{\text{av}} = 6.783 \pm 0.069 \text{ MHz},$$

$$(\Delta H)_{\text{av}} = 7.5916 \pm 0.0164 \text{ G},$$

and[14]

$$a_{3/2,1/2} = 46.17 \pm 35 \text{ MHz} \,.$$

Substituting these values into Eq. (78) yields the results shown in the first column of Table V.

If these values are used in the procedure of Sec. II as a test of consistency, the results obtained for V_{av} and $(\Delta H)_{\text{av}}$ are not in very good agreement with experiment. However, following Brog, Eck, and Wieder, we can improve this procedure further. The most precise of the experimental quantities is $(\Delta H)_{\text{av}}$. However, the relation obtained for $(\Delta H)_{\text{av}}$

TABLE V. Hyperfine coupling constant results from experimental approach expressed in MHz.

	Eq. (78)	Eq. (78) with 2nd order	LPD[a]
$a_{c,3/2,3/2}$	-9.890 ± 0.117	-9.806 ± 0.117	-9.888 ± 0.099
$a_{dip,3/2,3/2}$	-1.915 ± 0.034	-1.909 ± 0.034	-1.869 ± 0.019
$a_{orb,3/2,3/2}$	8.566 ± 0.039	8.638 ± 0.039	8.728 ± 0.087
$a_{3/2,3/2}$	-3.240 ± 0.128	-3.077 ± 0.128	-3.029 ± 0.133

[a]See Ref. 2.

is the least accurate since neglected effects are largest at H_{C1}. In view of this a significant improvement can be made by considering the second-order perturbation effects of the magnetic hyperfine interactions on $(\Delta H)_{av}$. The straightforward approach might seem to require solving a level-crossing equation to second order for the crossing fields; however, the resulting equation for $(\Delta H)_{av}$ is too complex to be useful. An alternative approach which preserves the simplicity of Eq. (77) is to calculate the changes in the crossing field values due to second-order perturbation terms and remove these increments from the measured field values. Thus the remaining part of the measured value of $(\Delta H)_{av}$ should be a more appropriate quantity to substitute into Eq. (77).

These second-order "corrections" to the measured field values may be derived starting with the second-order level-crossing equation

$$\sum_{i=0}^{2}[E_i(\tfrac{3}{2},\ -\tfrac{3}{2},\ M_I)-E_i(\tfrac{1}{2},\ \tfrac{1}{2},\ M_I)]=0\ . \qquad (80)$$

This equation is solved for the crossing field in the same approximations as used for Eq. (75). Using $g_M=1$ and $g_S=2$ the change in $H(M_I)$ resulting from the addition of second-order terms to the level-crossing equation is

$$\delta H(M_I)=[E_2(\tfrac{3}{2},\ -\tfrac{3}{2},\ M_I)-E_2(\tfrac{1}{2},\ \tfrac{1}{2},\ M_I)]/$$

$$[(2+B(H)^2)\,\mu_B]\,. \qquad (81)$$

The calculation of $E_2(J,\ M_J,\ M_I)$ is straightforward, requiring again the use of the zero-order eigenfunctions and eigenvalues and the M_L, M_S, M_I matrix elements given above.

The results for the second-order "corrections," Eq. (81), to be removed from $H(M_I)$ are given in Table VI. These values were obtained by using the results in the first column of Table V for $a_{i,3/2,3/2}$. Also included in this table are similar results obtained by Brog, Eck, and Wieder by their two-parameter calculation. It is seen here that the effect of using only two parameters to describe the hfs represents a major change. In addition, the procedure of Sec. II was used to calculate approximately these second-order effects of the magnetic hyperfine interactions. This was done by eliminating off-diagonal hfs matrix elements from the calculation. These results are also given in Table VI for comparison.

The second-order effects were removed from the measured $(\Delta H)_{av}$, Eq. (78) reapplied, and the resulting values given in the second column of Table V. These values are much more self-consistent. That is, when these values for $a_{i,3/2,3/2}$ are used in the complete procedure of Sec. II, the results for $(\Delta H)_{av}$ and V_{av} are

$$(\Delta H)_{av}=7.5902\ \text{G},$$

$$V_{av}=6.781\ \text{MHz}\ ,$$

in very good agreement with the measured values used to obtain the $a_{i,3/2,3/2}$. Also, if these new values are used to repeat the calculation of second-order terms, the results are identical to those obtained initially so that iterative procedure is clearly converged. In addition, calculated values of $a_{i,3/2,3/2}$ are listed in Table V to show that agreement is within assumed error ranges.

The procedure given by Brog, Eck, and Wieder for the determination of b is quite adequate. Using our second-order corrections and their measured values of the crossing field intervals at H_{C1} in 2 ^2P lithium, we obtain $b=-0.25\pm0.20$. This corresponds to their result of $b=-0.18\pm0.12$, the only difference being the choice between the use of columns 1 or 2 in Table VI in the calculation. This emphasizes the fact that the experimental precision is really not adequate to determine the quadrupole moment in lithium. However, in systems with significantly larger electric quadrupole interaction there should be no difficulty.

IV. CONCLUSIONS

We have discussed two alternative ways to link experimental level-crossing data with a theoretical analysis. From the theoretical point of view one can utilize calculated values of the hyperfine coupling constants to calculate the entire level structure including all crossing data of interest. The experi-

TABLE VI. Comparison of second-order changes in crossing-field values at H_{C1} in G.

	BEW[a]	Eq. (81)	Sec. II[b]
$\delta H(\frac{3}{2})$	0.085	0.0713	0.0624
$\delta H(\frac{1}{2})$	-0.056	-0.0735	-0.0718
$\delta H(-\frac{1}{2})$	-0.136	-0.1477	-0.1642
$\delta H(-\frac{3}{2})$	-0.148	-0.1512	-0.1780

[a]See Ref. 1.

[b]Total effect on crossing field values of leaving out off-diagonal hfs matrix elements in level-structure calculation.

mental approach, on the other hand, offers two possibilities. One procedure is to obtain three or four equations relating the appropriate hyperfine parameters to observable quantities and solve them as in Sec. III above. If this procedure does not yield adequate values for the parameters, then the results can be used iteratively in the procedure described in Sec. II until values are obtained which produce level structure consistent with that observed. Of course, this iterative approach could be used with any initial set of hyperfine parameters available for the system of interest. It should be stressed again that the perturbation approach of Sec. III may be easily modified for any system in which three or four experimental quantities are known. Also, as theorists calculate hyperfine constants for new systems, they might be encouraged to provide, in addition, the predicted level-crossing data to assist future experimental work.

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