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EFFECT ON HYPERFINE STRUCTURE

Robert W. Schmieder

February 1970

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MATRIX ELEMENTS OF THE QUADRATIC STARK EFFECT ON HYPERFINE STRUCTURE

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February 1970

ABSTRACT

The matrix elements for the second-order perturbation of hyperfine levels by a uniform static electric field are written in a form that does not assume the field is in the z-direction. Use of vector coupling coefficients separates the effects into monopole and quadrupole interactions. Some numerical values of the elements are given, and the energy levels of typical atoms obtained by diagonalization are plotted as a function of the field.

### INTRODUCTION

It is well-known that the linear Stark effect is identically zero in nearly all atoms.<sup>1</sup> This is because nearly all atoms, unlike most molecules, have nondegenerate energy eigenstates that have well-defined parity, and the average value of the Stark operator

$$V_{\epsilon} = - \vec{\epsilon} \cdot \vec{p} \quad (1)$$

where  $\vec{p}$  is the electric dipole moment and  $\vec{\epsilon}$  is the static uniform electric field, is zero. Thus, the energy levels are perturbed only in second (and higher orders) and the energy level shifts are proportional to  $\epsilon^2$  (and higher powers of  $\epsilon$ ). Because of this, the level shifts at reasonable fields (< 500 kV/cm) are very small ( $10^{-10^3}$  MHz) and only in the past few years have non-spectroscopic techniques<sup>2</sup> been developed capable of measuring such small shifts.

The analysis of a typical Stark effect measurement requires knowledge of the kinematical effects of the electric field on the atom--how the energy levels split, whether they cross, their behavior at very large fields, and so on. When the atom has hyperfine structure (hfs), it is necessary to diagonalize the perturbation hamiltonian (quadratic Stark effect plus hfs), and this obviously requires computation of the matrix elements. The matrix elements of the hf interaction are well known and available many places,<sup>3</sup> but those of the quadratic Stark effect are more complicated, and numerical values have not been published.

It is usual to use a computer to evaluate these matrix elements and perform the diagonalization, and this is by far the easiest way in practice.

However, much insight can be gained from a numerical table of matrix elements arranged as a matrix. Rather than assisting in the analysis of an experiment, such a table is useful for understanding the symmetries of the interaction, identifying relationships, and spotting simplifications.

We present in this paper the matrix elements of the quadratic Stark operator in the representation in which  $\vec{F} = \vec{I} + \vec{J}$  and  $M = M_I + M_J$  are constants of the motion. The matrix elements in the  $|M_I M_J\rangle$  representation are not as often needed, and we do not present these. Some examples are given of energy level shifts in the hfs of alkali atoms. The alkali atoms (Li, Na, K, Rb, Cs) are particularly important because they can be treated like one-electron atoms.<sup>4</sup>

## THEORY

The effects of the field  $\vec{\epsilon}$  on a degenerate state in second order are given by the matrix elements of the effective operator<sup>5</sup>

$$V_{\epsilon\epsilon} = \vec{\epsilon} \cdot \vec{p} \lambda \vec{\epsilon} \cdot \vec{p} \quad (2)$$

where  $\lambda = \sum_i P_i / (E - E_i)$  is a factor involving the energy difference  $E - E_i$  between the state being perturbed and the  $i^{\text{th}}$  intermediate state, and projection operators  $P_i$  for each  $i^{\text{th}}$  state. Using the algebra of irreducible spherical tensors,<sup>6</sup> it is possible to rewrite Eq. (2) in the form

$$V_{\epsilon\epsilon} = -\frac{1}{2} \alpha_0 \epsilon^2 - \frac{1}{2} \alpha_2 \epsilon^2 Q \quad (3a)$$

where  $\alpha_0, \alpha_2$  are two constants that involve  $\lambda$  and the matrix elements of  $\vec{p}$ , and

$$Q = \left[ \frac{3\hat{n} \cdot \vec{J} \hat{n} \cdot \vec{J} - \vec{J} \cdot \vec{J}}{J(2J-1)} \right]. \quad (3b)$$

In Eq. (3b),  $\hat{n}$  is a unit vector in the direction of  $\vec{\epsilon}$ , and  $\vec{J}$  is the angular momentum operator of magnitude  $\vec{J} \cdot \vec{J} = J(J+1)$ .

By rewriting Eq. (2) as Eq. (3) we have separated the interaction into two parts, one that is a simple number, independent of  $\hat{n}$  and  $\vec{J}$  and termed a "scalar interaction", and the other involving  $\hat{n}$  and  $\vec{J}$  in a form similar to the Legendre polynomial  $P_2 = \frac{1}{2} (3\mu^2 - 1)$ , and termed a "tensor interaction". The numbers  $\alpha_0, \alpha_2$  are called the scalar and tensor polarizabilities,<sup>7</sup> and are given in units<sup>8</sup> of  $\text{cm}^3$  or  $\text{MHz}/(\text{kV}/\text{cm})^2$ . These numbers can be numerically

calculated for simple atoms like the alkalis and the alkali isoelectronic sequence, and good agreement between such computed values and experimental ones has been obtained.<sup>9</sup> The polarizabilities are the same type of quantity as a magnetic moment, i.e., a constant of proportionality between energy and field. The dynamical structure of the interaction is contained in these constants and the magnitude of the field, whereas the kinematical structure is represented by the multipole operators  $1$  (the identity operator) and  $Q$ .

MATRIX ELEMENTS

Since we wish to study the effects of Eq. (3) on the hfs of an atom, we require matrix elements of  $V_{\epsilon\epsilon}$  between two hf states  $|n(I(s\ell)J)FM\rangle$  and  $|n'(I'(s'\ell')J')F'M'\rangle$ . However, since  $V_{\epsilon\epsilon}$  operators only on electronic coordinates (not nuclear or electronic spin), we know that  $I' = I$  and  $s' = s$ . Now suppose we always keep the field  $\vec{\epsilon}$  small enough so that the level shifts, whatever they might be, are always much smaller than the fine structure separations  $E(n(s\ell)J) - E(n(s\ell)J')$ . Under this condition, the elements off-diagonal in  $n, \ell, J$  will be very small, so only  $n' = n, \ell' = \ell, J' = J$  elements are important. Thus, we have only elements between states  $|n(I(s\ell)J)FM\rangle \equiv |FM\rangle$  and  $|n(I(s\ell)J)F'M'\rangle \equiv |F'M'\rangle$  to consider, i.e.,  $\langle FM|V_{\epsilon\epsilon}|F'M'\rangle$ .

The matrix elements of  $V_{\epsilon\epsilon}$  can be computed straightforwardly from Eq. (3) if we write

$$\hat{n} \cdot \vec{J} = -n_{+1}J_{-1} + n_0J_0 - n_{-1}J_{+1} \quad (4)$$

where  $n_{\pm 1} = \mp(n_x \pm in_y)/\sqrt{2}$ ,  $n_0 = n_z$ , (same form for  $J_{\pm 1}, J_0$ )

and using the matrix elements<sup>10</sup> ( $\mu = \pm 1, 0$ ),

$$\langle FM|F'M'\rangle = \delta_{FF'} \delta_{MM'} \quad (5)$$

$$\langle FM|J_{\mu}|F'M'\rangle = (-1)^{F-M} \begin{pmatrix} F & 1 & F' \\ -M & \mu & M' \end{pmatrix} \quad (6)$$

$$\times (-1)^{I+J+F+1} \sqrt{(2F+1)(2F'+1)} \begin{Bmatrix} F & 1 & F' \\ J & I & J \end{Bmatrix} \\ \times \sqrt{J(J+1)(2J+1)} .$$



Specific forms of the (3-j) and {6-j} coefficients appearing in Eq. (6) are listed in many places,<sup>11</sup> and numerical values are available, so using Eqs. (4), (5), and (6) in Eq. (3) is direct, though tedious.

A much easier way is to use the fact that the operator  $Q$  in Eq. (3b) is a scalar product of a second rank tensor constructed from the three components of  $\hat{n} = (n_{+1}, n_0, n_{-1})$  and a second rank tensor operator, constructed from the three components of  $\vec{J} = (J_{+1}, J_0, J_{-1})$ . The following relation can be proved by direct multiplication:

$$Q = \frac{3}{J(2J-1)} \left( \hat{n}\hat{n} - \frac{1}{3} \vec{J}\vec{J} \right) : \left( \frac{\vec{J}\vec{J} + (\vec{J}\vec{J})^\dagger}{2} - \frac{1}{3} \vec{J} \cdot \vec{J} \vec{J}\vec{J} \right) \quad (7a)$$

Now  $\hat{X} \equiv \hat{n}\hat{n} - \frac{1}{3} \vec{J}\vec{J}$  is a dyadic, which represents a second-rank irreducible tensor  $X^2$ , which has components

$$X^2_\mu = \sum_{qq'} n_q n_{q'} \begin{pmatrix} 1 & 2 & 1 \\ q & \mu & q' \end{pmatrix} \sqrt{5} (-1)^\mu \quad (7b)$$

Likewise, the rest of  $Q$  in Eq. (7a) forms a second-rank irreducible tensor operator  $T^L_\mu$  with components  $T^L_\mu$ . Thus, Eq. (7a) can be written<sup>12</sup>

$$Q = \sum_\mu (-1)^\mu X^2_{-\mu} T^2_\mu \equiv X^2 \cdot T^2 \quad (7c)$$

which is in the form of a scalar product of two tensors.

The matrix elements of tensor operators are particularly simple,<sup>13</sup> and from Eq. (7c) it is not difficult to show that

$$\begin{aligned} \langle FM | V_{\epsilon\epsilon} | F'M' \rangle &= -\frac{1}{2} \alpha_0 \epsilon^2 \delta_{FF'} \delta_{MM'} \\ &\quad - \frac{1}{2} \alpha_2 \epsilon^2 Q_{FF';MM'} \end{aligned} \quad (8a)$$

where

$$\begin{aligned} Q_{FF';MM'} &= \sqrt{\frac{15}{2}} \left[ \frac{(J+1)(2J+1)(2J+3)}{J(2J-1)} \right]^{\frac{1}{2}} \sum_{\mu} \sum_{qq'} n_q n_{q'} \begin{pmatrix} 1 & 2 & 1 \\ q & -\mu & q' \end{pmatrix} \\ &\quad \times (-1)^{I+J+F-F'-M} \sqrt{(2F+1)(2F'+1)} \begin{pmatrix} F & 2 & F' \\ M & \mu & -M' \end{pmatrix} \begin{Bmatrix} F & 2 & F' \\ J & I & J \end{Bmatrix}. \end{aligned} \quad (8b)$$

In Eq. (8b) the sum over  $\mu, q, q'$  result from writing the operator as a scalar product of two tensors. The (3-j) symbols yield the two conditions  $\mu = q + q' = M' - M$ , which means there is actually only one independent sum in Eq. (8b).

The matrix elements of Eq. (8a,b) are valid when the electric field is in any direction, specified by the components of  $\hat{n}$ . If for some reason<sup>14</sup> we find it desirable to establish a z-axis in some direction other than along  $\vec{\epsilon}$ , these general elements will be required, and  $V_{\epsilon\epsilon}$  is clearly not diagonal in  $M$ . However, if we can chose  $\vec{\epsilon} = \epsilon \hat{e}_z$ , we have  $\hat{n} = (0, 1, 0)$  and

$$\begin{pmatrix} 1 & 2 & 1 \\ 0 & 0 & 0 \end{pmatrix} = \sqrt{\frac{2}{15}}, \text{ and since } \mu = 0 \text{ the second (3-j) symbol in Eq. (8) is}$$

zero unless  $M = M'$ . Thus, defining  $Q_{FF';M} \equiv Q_{FF';MM}$ , we find

$$Q_{FF';M} = \left[ \frac{(J+1)(2J+1)(2J+3)}{J(2J+1)} \right]^{\frac{1}{2}} (-1)^{I+J+F-F'-M} \quad (9)$$

$$\times \sqrt{(2F+1)(2F'+1)} \begin{pmatrix} F & 2 & F' \\ M & 0 & -M \end{pmatrix} \begin{Bmatrix} F & 2 & F' \\ J & I & J \end{Bmatrix}$$

Thus, the matrix  $Q$  is strictly diagonal in  $M$  when  $\epsilon$  is in the  $z$ -direction; for every value of  $M$ , there is a matrix with rows and columns labelled by  $F$  and  $F'$ , respectively. In Fig. 1 we illustrate the forms of these matrices for an atom with  $I = 5/2$ ,  $J = 3/2$ .

## NUMERICAL VALUES

In Table I we list numerical values of  $Q_{FF';M}$  for the important case of  $J = 3/2$  state and  $I = 3/2, 5/2, 7/2$ . These values are appropriate to the  $^2P_{3/2}$  states of the stable alkali atoms,  $^7\text{Li}$ ,  $^{23}\text{Na}$ ,  $^{39}\text{K}$ ,  $^{85}\text{Rb}$ ,  $^{87}\text{Rb}$ ,  $^{133}\text{Cs}$ , as well as others. For such atoms, the  $^2P_{3/2}$  states are split by the hyperfine interaction into four hf states labelled by  $F = I + 3/2, I + 1/2, I - 1/2, I - 3/2$ , each with  $2F + 1$  magnetic substates. Since only substates with the same  $M$  (but possibly different  $F$ ) can perturb each other, matrices  $Q_{FF';M}$  will be at most  $4 \times 4$ , and for some  $M$ , smaller than  $4 \times 4$ .

There are several interesting observations to be made on Table I:

- 1) The ( $F = F' = 3, M = 2$ ) elements are zero for all  $I$ ;
- 2) The ( $F = F' = 2, I = 3/2$ ) elements are zero for all  $M$ ;
- 3) The  $F = F' = 0$  element = 0 (this is the  $0 \neq 0$  selection rule);
- 4) No elements besides those of 1) 2) 3) are zero, except for  $|F - F'| > 2$ ;
- 5) The  $M = 0$  matrices are reducible from  $4 \times 4$  to two  $2 \times 2$ , and the energy eigenvalues are roots of quadratics instead of quartics. This follows from the invariance of the determinant to reordering of the rows and/or columns:

$$\begin{aligned}
 \det \begin{bmatrix} a & 0 & \delta & 0 \\ 0 & b & 0 & \eta \\ \delta & 0 & c & 0 \\ 0 & \eta & 0 & d \end{bmatrix} &= \det \begin{bmatrix} a & \delta & 0 & 0 \\ \delta & b & 0 & 0 \\ 0 & 0 & c & \eta \\ 0 & 0 & \eta & d \end{bmatrix} \\
 &= \det \begin{bmatrix} a & \delta \\ \delta & b \end{bmatrix} \det \begin{bmatrix} c & \eta \\ \eta & d \end{bmatrix} \\
 &= (ab - \delta^2)(cd - \eta^2) .
 \end{aligned} \tag{10}$$

Thus, the energy eigenvalues  $E$  satisfy

$$\begin{aligned}(a - E)(b - E) - \delta^2 &= 0 \\(c - E)(d - E) - \eta^2 &= 0\end{aligned}\tag{11}$$

and therefore the curves  $E(|\vec{\epsilon}|)$  are parabolas.

- 6) All diagonal elements are rational, and nearly all off-diagonal elements are irrational.

The fact that the  $\pm M$  states have the same matrix elements means the quadratic Stark interaction will not separate these two substates.

## ENERGY LEVELS

Although the main purpose of this paper is to present the matrix elements of  $V_{\epsilon\epsilon}$ , the main purpose of the matrix elements is the computation of energy levels versus  $\epsilon$ . Since we are considering hfs and Stark effect perturbations of roughly equal magnitude, we must find the energy shifts as the eigenvalues of

$$V = V_{\text{hfs}} + V_{\epsilon\epsilon} \quad (12)$$

But from Eq. (8a) the scalar part of  $V_{\epsilon\epsilon}$  (involving  $\alpha_0$ ) is not only diagonal in the (FF';MM') subspace but actually a multiple of the unit matrix. Thus, every magnetic substate will be shifted by exactly the same amount by this term, namely  $-\frac{1}{2}\alpha_0\epsilon^2$ , and we can ignore it when diagonalizing Eq. (12). Furthermore, since we are neglecting elements off-diagonal in I and J we can write out  $V_{\text{hfs}}$  explicitly, so Eq. (12) becomes

$$V' = a \vec{I} \cdot \vec{J} + b \left[ \frac{3(\vec{I} \cdot \vec{J})^2 + \frac{3}{2} \vec{I} \cdot \vec{J} - \vec{I} \cdot \vec{I} \vec{J} \cdot \vec{J}}{2I(2I-1)J(2J-1)} \right] - \frac{1}{2}\alpha_2\epsilon^2 Q \quad (13)$$

where a,b are the dipole, quadrupole hfs constants, and we have dropped the  $\alpha_0$  term.

The operator Eq. (13) represents the quadrupole hfs quadratic Stark effect, and the energy levels may be found as a function of  $\epsilon$  by evaluating  $\det\{V'-E\} = 0$ . We have used a computer to diagonalize Eq. (13) for a number of cases of interest. Figure 2 shows a typical result, for the  $7^2P_{3/2}$  state of  $^{133}\text{Cs}$  (I = 7/2). The 0 on the vertical scale is the energy of the  $7^2P_{3/2}$  state for  $V = 0$ . The four unperturbed hf levels at  $\epsilon = 0$  have  $F = 5, 4, 3, 2$ ,

and for very large  $\epsilon$ , the  $m_J = \pm 3/2, \pm 1/2$  substates are well separated. In order to mark the horizontal scale in units of  $[\epsilon] = \text{kV/cm}$ , it is necessary to know  $a, b$  and  $\alpha_2$ . The former are generally known from measurements,<sup>15</sup> and values of  $\alpha_2$  are currently being computed and measured.<sup>16</sup> We used  $a = 18.6 \text{ MHz}$ ,  $b = -0.11 \text{ MHz}$ , and the theoretical value<sup>17</sup>

$$\alpha_2 = -1.072 \text{ MHz}/(\text{kV/cm})^2.$$

Several aspects of Fig. 2 may be noticed:

- 1) The center-of-gravity of the energy levels remains at zero for all  $\epsilon$ . This is a consequence of the zero trace (average) of all non-zero rank tensors. Note that the scalar part of  $V_{\epsilon\epsilon}$ , excluded from Fig. 2, is a zero rank tensor, and adds to each curve the parabola  $-\frac{1}{2}\alpha_0\epsilon^2$ . Since  $\alpha_0 > 0$  and  $|\alpha_0| \gg |\alpha_2|$  (usually) the actual levels are bent downward sharply;
- 2) All substates  $M \neq 0$  are doubly degenerate ( $\pm M$ );
- 3) Level crossings occur at  $\epsilon > 0$ . These crossings, and those occurring at  $\epsilon = 0$ , form the basis for electric field level crossing experiments.<sup>18</sup>

It might be constructive to compare Fig. 2 with a similar plot of the Zeeman effect (Fig. 3). This plot was computed in the same way as Fig. 2, using Eq. (11) with  $g_J\mu_0\mathcal{H}_z$  instead of  $-\frac{1}{2}\alpha_2\epsilon^2$ . The main differences are in degeneracy ( $M$  vs.  $\pm|M|$ ) and field dependence ( $\mathcal{H}$  vs.  $\epsilon^2$ ); both effects have constant center-of-gravity, and at large fields the levels separate into groups labelled by  $m_J$  (i.e., Paschen-Back effect).

Finally, in Fig. 4 we plot the energy levels of the  $3^2P_{3/2}$  state of  ${}^7\text{Li}$  vs.  $\epsilon$ . This atom is interesting because the dipole hfs constant  $a$  is negative, a unique occurrence among the alkali atoms. Figure 4 makes use of the extrapolated values  $a = -0.935$  MHz,  $b = -0.04$  MHz, and the theoretical<sup>17</sup> value  $\alpha_2 = -0.377$  MHz/(kV/cm)<sup>2</sup>. The result of  $a < 0$  is that the crossing near 3.25 kV/cm is exceptionally sharp (compare Fig. 2). This fact is currently being exploited in a level-crossing experiment to obtain precise values of the hfs constants  $a, b$  in this state.<sup>19</sup>



FOOTNOTES AND REFERENCES

\* Work supported under the auspices of the U. S. Atomic Energy Commission.

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8. The relationship between the two units is found by writing Eq. (3a) in cgs-esu

$$V_{\epsilon\epsilon}[\text{erg}] = -\frac{1}{2} \alpha[\text{cm}^3](\epsilon[\text{esu}])^2$$

and using

$$V_{\epsilon\epsilon}[\text{MHz}] = \frac{10^{-6}}{h} V_{\epsilon\epsilon}[\text{erg}]$$

$$\epsilon[\text{kV/cm}] = 10^3 \cdot \frac{1}{300} \epsilon[\text{esu}]$$

so we find

$$V_{\epsilon\epsilon}[\text{MHz}] = -\frac{1}{2} \left( \frac{1}{h(300)^2} \alpha[\text{cm}^3] \right) (\epsilon[\text{kV/cm}])^2$$

from which

$$\alpha[\text{cm}^3] = 596 \times 10^{-24} \alpha[\text{MHz}/(\text{kV/cm})^2]$$

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## FIGURE CAPTIONS

Fig. 1. Arrangement of the matrix elements of  $Q$  for an atom with  $I = 5/2$ ,  $J = 3/2$ , grouped according to the value of  $M$ . The separation of different  $M$  matrices is possible only if  $\vec{\epsilon}$  is in the z-direction. For different values of  $I$ , there are more (or less)  $4 \times 4$  matrices, as seen in Table I.

Fig. 2. Energy levels vs.  $\epsilon$  of the  $7^2P_{3/2}$  state of the  $Cs^{133}$  atom. The constant shift  $-\frac{1}{2}\alpha_0\epsilon^2$  of all levels has not been included.

Fig. 3. Energy levels of the same atomic state as Fig. 2, as a function of magnetic field. The differences between this plot and Fig. 2 are indicative of the symmetry differences of the Stark and Zeeman effects.

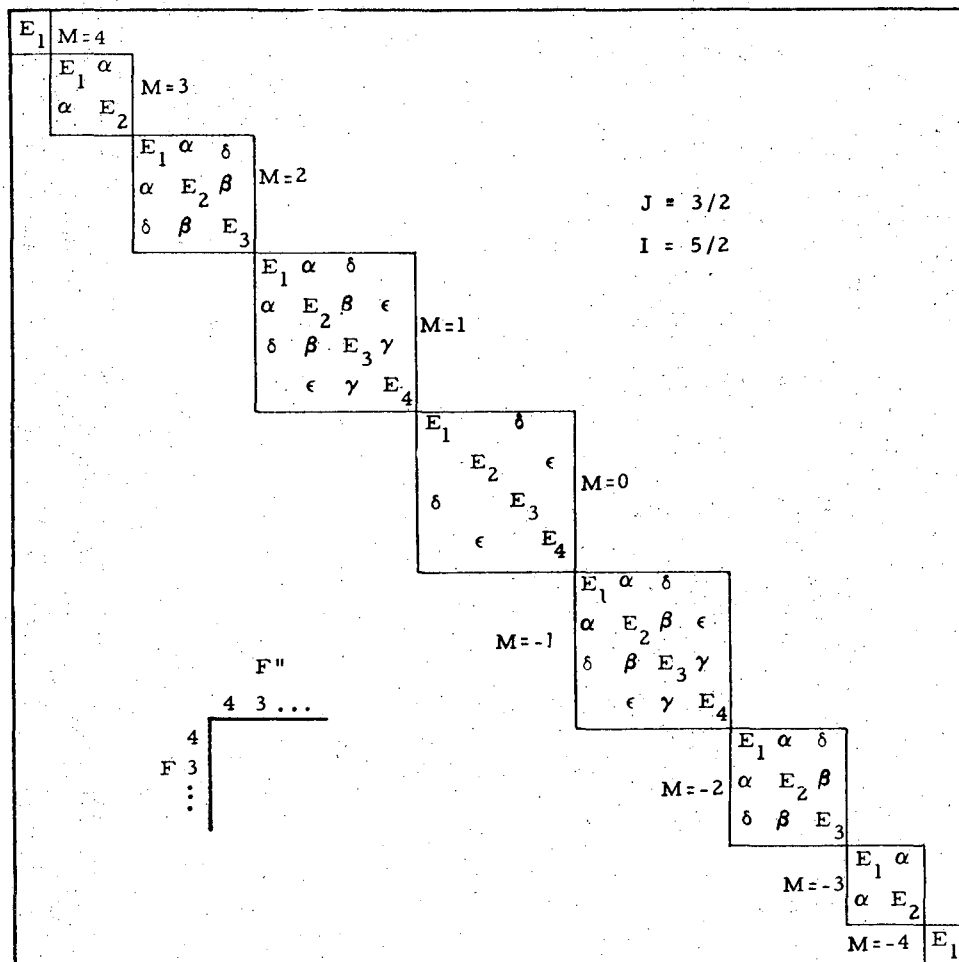
Fig. 4. Energy levels vs.  $\epsilon$  of the  $3^2P_{3/2}$  state of  $Li^7$ . As in Fig. 2, if we include the constant shift  $-\frac{1}{2}\alpha_0\epsilon^2$ , we find the actual levels bend downward sharply.

I=3/2	I=5/2	I=7/2																											
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XBL 703-501

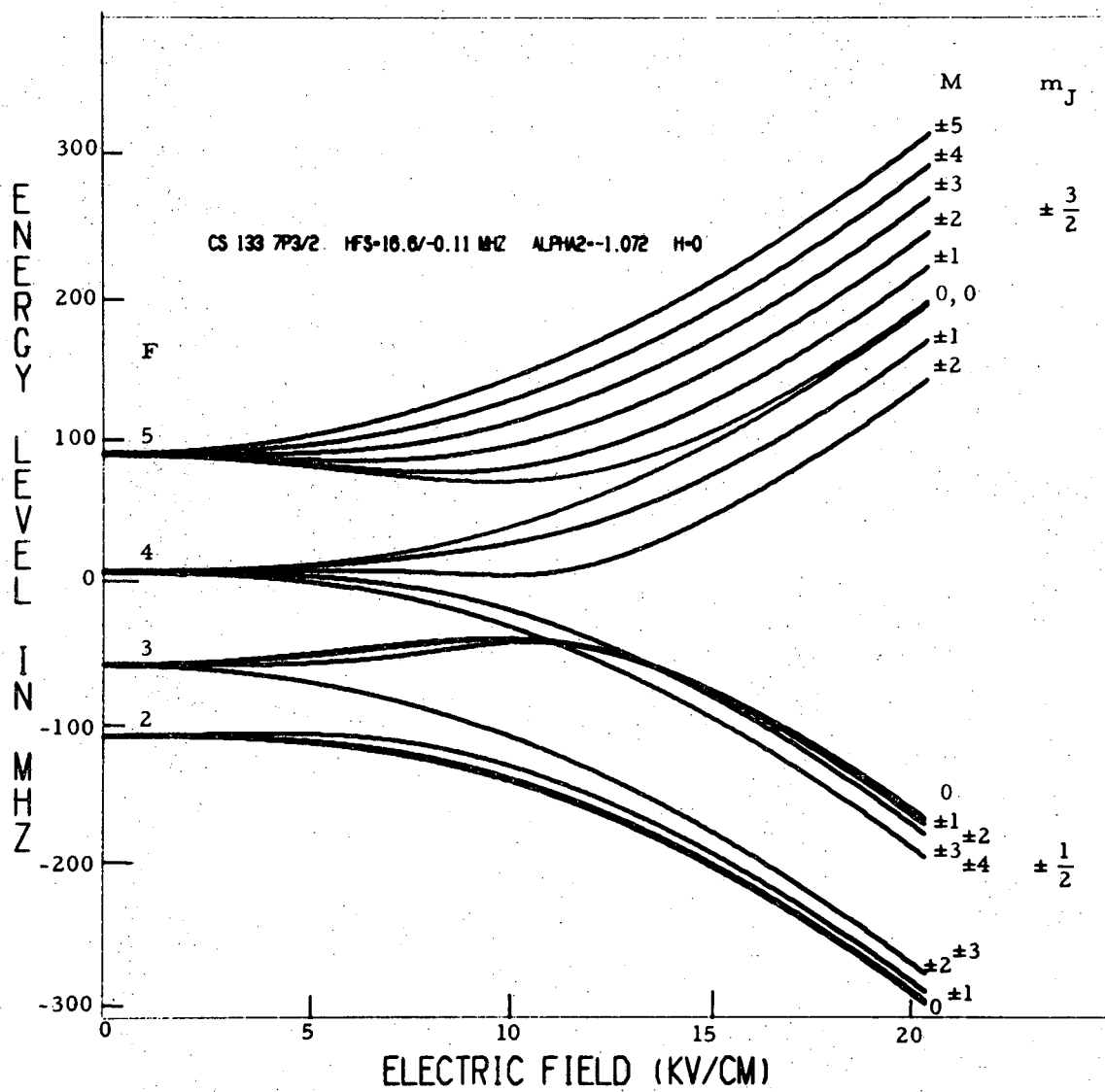
Table I.

Matrix Elements of the Second-Order Stark Operator.



XBL 703-505

Fig. 1.



XBL 703-504

Fig. 2.

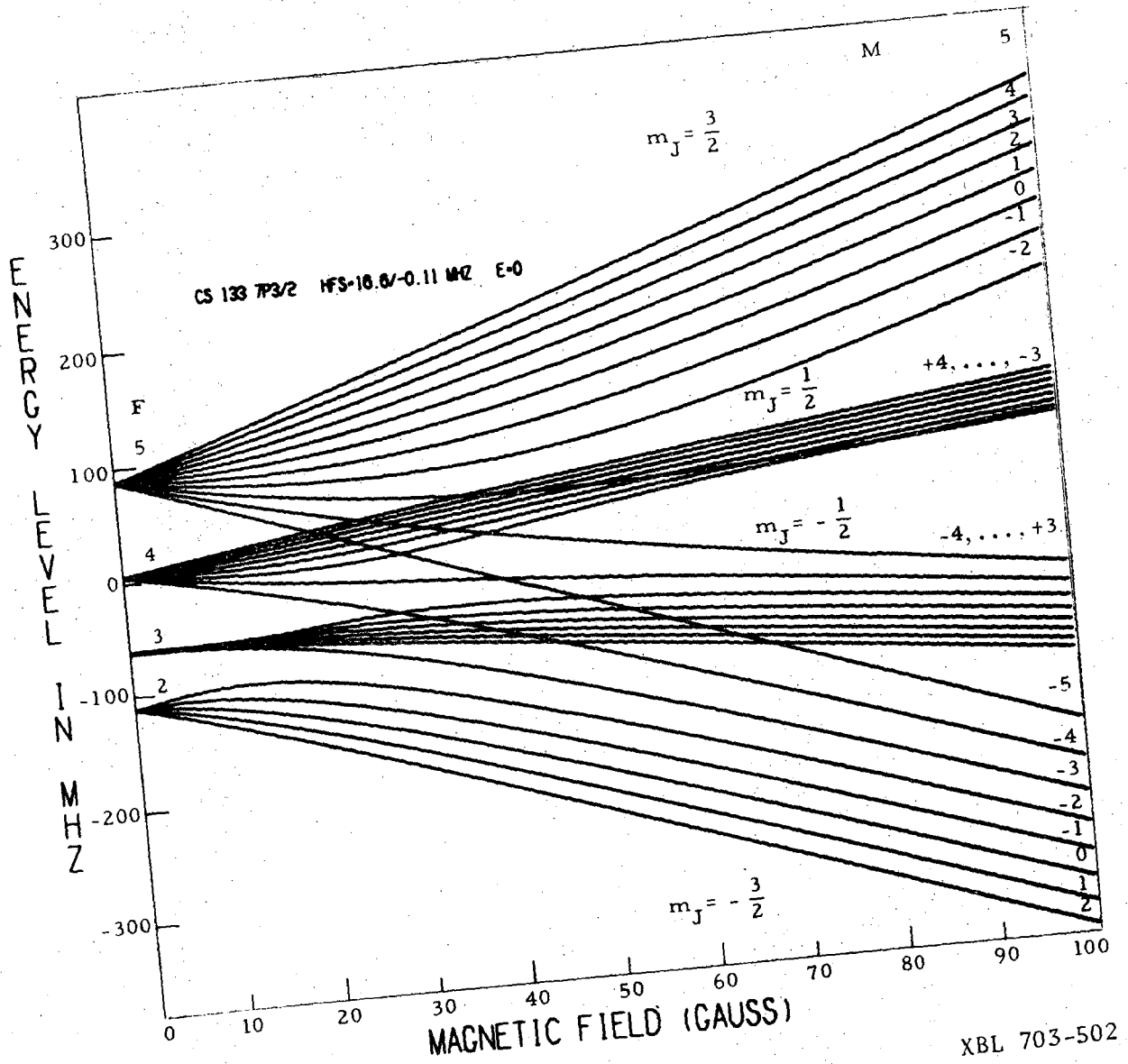
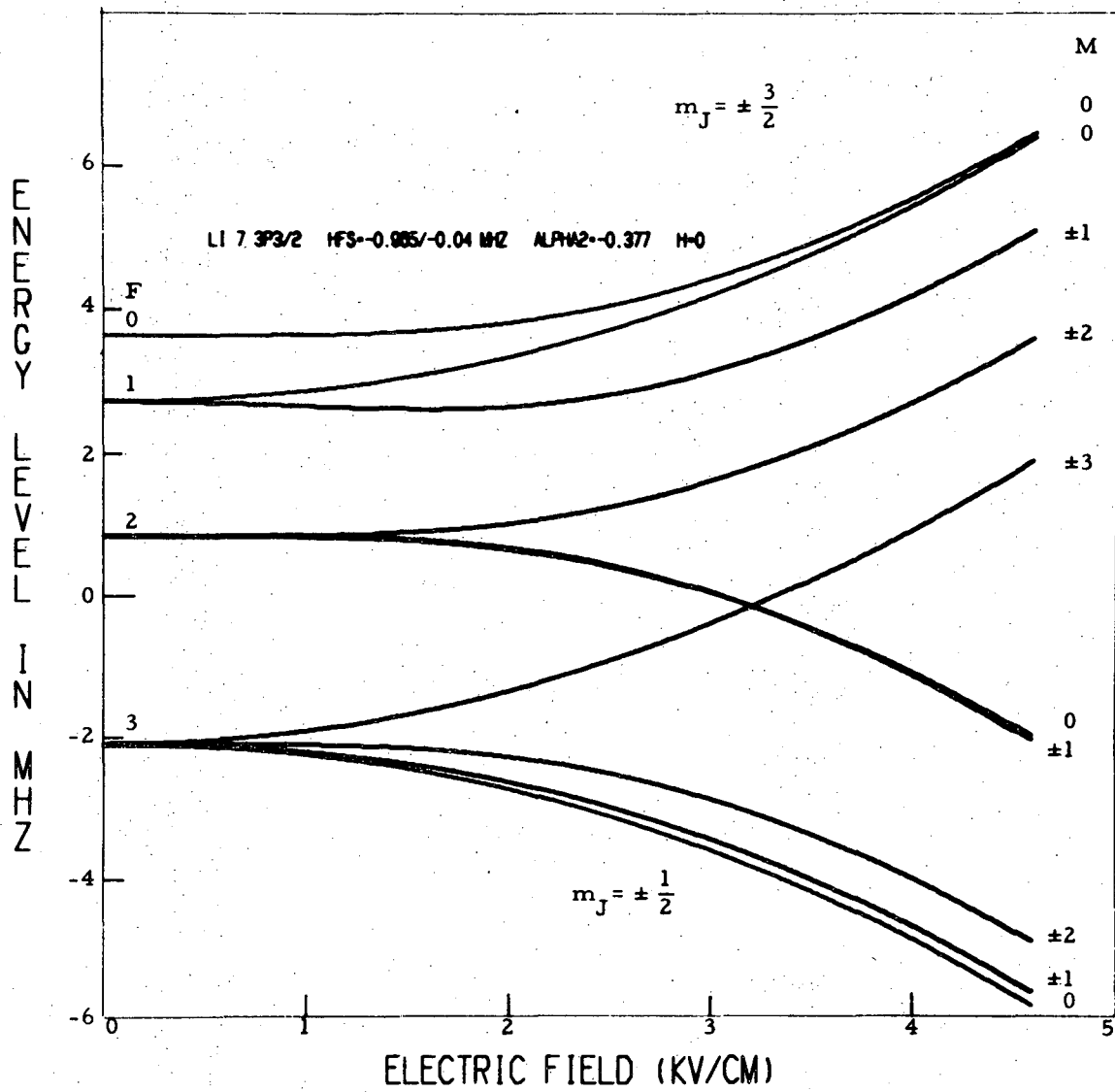


Fig. 3.





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Fig. 4.

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