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# An operator formalism for the calculation of intensities of allowed and forbidden hyperfine transitions in EPR spectra

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**Abstract.** Operator expressions involving only the components of the electron and nuclear spin operators have been derived. The squares of the absolute values of the matrix elements of these operators between the respective zero-order eigenfunctions yield the intensities of the allowed ( $\Delta M = 1, \Delta m = 0$ ) and forbidden ( $\Delta M = 1, \Delta m = \pm 1, \pm 2$  and  $\Delta M = 2, \Delta m = 0, \pm 1$ ) EPR hyperfine transitions. (Here  $M, m$  are the electronic and nuclear magnetic quantum numbers, respectively.) The resulting expressions for the EPR line intensities are thus quite straightforward to calculate. The spin Hamiltonian considered consists of the electronic Zeeman, zero-field and hyperfine terms, wherein the principal axes of the various tensors  $\mathbf{g}$ ,  $\mathbf{D}$  and  $\mathbf{A}$ , all assumed to be anisotropic, are considered to be non-coincident; the orientations of the external Zeeman and excitation microwave fields are assumed to be arbitrary. Intensity expressions for the various transitions are specifically given. The present results are compared with those published previously. (The present results can be used to calculate the intensities of EPR transitions of purely electronic systems also by excluding the nuclear part of the spin Hamiltonian.)

## 1. Introduction

It has been suggested (Misra 1979) that the intensities of magnetic resonance transitions, alone, or in conjunction with the line positions, can be simultaneously fitted in a least-squares procedure to evaluate the spin-Hamiltonian (SH) parameters. The positions and intensities of the forbidden hyperfine (HF) lines have been used by Korkmaz and Aktas (1984) to determine the quadrupole interaction parameters, and by Mialhe (1973) to estimate the nuclear  $g$ -tensor. The mechanisms responsible for forbidden HF transitions and the importance of HF forbidden transitions in dynamic nuclear polarisation, discrete saturation, phase transition and liquid and glass structures has been pointed out in a recent review article by Misra and Upreti (1987a), which also includes a literature survey on the subject of HF forbidden transitions.

Forbidden hyperfine transitions ( $\Delta M = 1, \Delta m \neq 0$ ;  $\Delta M \neq 1, \Delta m \neq 0$ ) arise due to the admixing of nuclear states corresponding to different  $m$ -values. (Here  $M$  and  $m$  denote the electron and nuclear magnetic quantum numbers, respectively.) Bleaney

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and Rubins (1961) derived intensity expressions for the cases  $\Delta M = 1$ ,  $\Delta m = \pm 1, \pm 2, \pm 3$ , while the cases  $\Delta M = 1$ ,  $\Delta m = \pm 4, \pm 5$  were dealt with by Lupei *et al* (1972). These early investigations, based on perturbation theory, are inadequate because only admixing from the nearest-neighbouring eigenstates was taken into account in their derivation. More accurate expressions have been reported by Golding *et al* (1972) for the case  $\Delta M = 1$ ,  $\Delta m = 0, \pm 1, \pm 2$ , and by Golding and Tennant (1974) for  $\Delta M = 2$ ,  $\Delta m = 0, \pm 1$ . Fulton *et al* (1986) have described a diagram technique that can be used to calculate intensity expressions; however, they did not provide any specific expressions for the intensity of the HF transitions. Also, this technique does not appear to have been used by others. Bir (1964) calculated the angular variation of the EPR line intensity, assuming the HF interaction to be much smaller than the zero-field term, using the method of 'effective magnetic field'. In his method, the axis of quantisation for nuclear spin was taken to be along the direction of the effective magnetic field at the nucleus created by the electrons, being different for different electronic states. Mialhe and Erbeia (1973a, b) adapted the theory of Bir to derive intensity expressions for the cases  $\Delta M = 1$ ,  $\Delta m = 0, \pm 1$ . Their method consisted in expressing the admixture coefficients of the eigenstates in the form of operators so that the intensity of a magnetic resonance transition can be calculated to be proportional to the squared absolute value of their matrix element between the zero-order (unperturbed) eigenstates. An exhaustive, annotated review of the forbidden HF transitions has been given by Misra and Upreti (1987a). More recently, the subject has been reviewed briefly by Misra and Upreti (1987b) and by Weil (1987).

There are, however, certain errors in the operator expression given by Mialhe (1973a). On the other hand, the intensity expressions given by Golding *et al* (1972) and Golding and Tennant (1974) are incomplete, in that the admixture of all the possible eigenstates has not been taken into account. Apart from this, their results for  $\Delta M = 1$ ,  $\Delta m = \pm 2$  and  $\Delta M = 2$ ,  $\Delta m = 0$  are incorrect. Furthermore, they did not consider the case where the external Zeeman field is parallel to the microwave excitation field.

The intensity of a magnetic resonance transition is proportional to the squared absolute value of the matrix element of the Hamiltonian representing the interaction of the excitation microwave field with the magnetic moment of the electron between the states participating in resonance. Thus, the calculation of intensity requires perturbed eigenfunctions. Consequently, the intensity expressions consist of a large number of matrix elements between the unperturbed eigenstates. Methods to keep track of the required non-zero matrix elements have been described by Misra and Upreti (1987b), and by Skinner and Weil (1978).

It is the purpose of the present paper to describe a straightforward procedure to calculate the intensity of EPR lines by the use of operators that depend only on the components of the electron spin ( $S$ ) and the nuclear spin ( $I$ ). The operator expressions so derived in the present paper take into account complete admixtures of all the perturbed eigenstates up to second order. In order to compute the intensity using the operator expressions derived here, all one has to do is, then, simply to evaluate the matrix elements of these operators, which depend on the components of the operators  $S$  and  $I$ , between the zero-order simultaneous eigenvectors of the components of  $S$  and  $I$  along their respective axes of quantisation. The orientations of the Zeeman and excitation fields have, here, been chosen to be arbitrary. The spin Hamiltonian considered consists of the electronic Zeeman, zero-field and HF terms, appropriate to lowest symmetry, i.e., triclinic. For higher symmetries, one can simply put the appropriate coefficients of those spin operators, which should be absent for the particular symmetry, equal to zero (see

**Table 1.** Non-vanishing zero-field operator coefficients  $B_k^q$  in the spin Hamiltonian for the various symmetries. Also included is the relationship between the principal values of the HF tensor. For monoclinic symmetry  $C_2$  represents the orientation of the twofold axis, while  $x, y, z$  represent the magnetic axes.

Cubic:	$B_4^0, B_4^4 = 5B_4^0, B_6^4 = -21B_6^0; A_z = A_x = A_y$
Axial:	$B_2^0, B_4^0, B_6^0; A_z, A_x = A_y$
Hexagonal:	$B_2^0, B_4^0, B_6^0, B_6^6; A_z, A_x = A_y$
Tetragonal:	$B_2^0, B_4^0, B_4^4, B_6^0, B_6^4; A_z, A_x = A_y$
Trigonal:	$B_2^0, B_4^0, B_4^3, B_6^0, B_6^3, B_6^6; A_z, A_x = A_y$
Orthorhombic:	$B_k^n; k = 2, 4, 6; 0 \leq m \text{ (even)} \leq k; A_z, A_y, A_x$
Monoclinic	Orthorhombic + $B_1$ , where $\begin{cases} B_1(C_2 \parallel x) = B_k^{-(2n-1)} \\ B_1(C_2 \parallel y) = B_k^{2n-1} \\ B_1(C_2 \parallel z) = B_k^{-2n} \end{cases}$ and $k = 2, 4, 6; 1 \leq n \leq k/2; A_z, A_x = A_y$
Triclinic	$B_k^q; k = 2, 4, 6; -k \leq q \leq k; A_z, A_y, A_x.$

table 1). Nuclear Zeeman interaction can easily be taken into account by defining an equivalent HF term (Weil and Anderson 1961, Iwasaki 1974). The axis of quantisation for  $I$  is assumed to be along the conventionally chosen direction (Abragam and Bleaney 1970, Rockenbauer and Simon 1974), namely, along the effective magnetic field direction at the nucleus (Orton 1968). The extended Stevens spin operators  $O_k^q$  (Rudowicz 1985a), including those with  $q < 0$ , have been used to describe the zero-field part of the electronic spin Hamiltonian. The required matrix elements of the components of  $S$  and  $I$  can be easily evaluated, e.g., by utilising the tables listed by Al'tshuler and Kozyrev (1974).

Section 2 deals with the spin Hamiltonian, as well as its second-order perturbed eigenfunctions required for the evaluation of the operators for the calculation of the intensity. The expressions of the operators required to calculate the EPR line intensity are derived in § 3. Specific expressions for the EPR line intensities are developed in § 4 for a few illustrative examples. A comparison of the results derived here with the previously reported expressions is presented in § 5. Concluding remarks are made in § 6.

## 2. The spin Hamiltonian and its second-order perturbed eigenfunctions

The following spin Hamiltonian, consisting of the Zeeman ( $\mathcal{H}_Z$ ), zero-field ( $\mathcal{H}_{ZF}$ ) and the hyperfine ( $\mathcal{H}_{HF}$ ) terms, is considered:

$$\mathcal{H} = \mathcal{H}_Z + \mathcal{H}_{ZF} + \mathcal{H}_{HF}$$

where

$$\begin{aligned} \mathcal{H}_Z &= \mu_B \mathbf{S}^T \cdot \mathbf{g} \cdot \mathbf{B} & \mathcal{H}_{HF} &= \mathbf{S}^T \cdot \mathbf{A} \cdot \mathbf{I} \\ \mathcal{H}_{ZF} &= \sum_k \sum_{q=-k}^k \{B_k^q\} \{O_k^q\} & & (k \text{ even, } \leq 2S). \end{aligned} \tag{1}$$

In (1), the superscript T represents the transpose of a matrix, **B** is the external Zeeman field;  $\mu_B$  is the Bohr magneton; **g**, **A**,  $B_k^q$  are the usual SH parameters and  $O_k^q$  are the extended Stevens operators (Rudowicz 1985a). The relations between the conventional Stevens operators ( $O_l^m$ ), the tensor operators ( $T_l^m$ ) of Buckmaster *et al* (1972) and the  $O_k^q$  operators, used in (1), have been given by Rudowicz (1985b, 1987) and their transformations under rotations are listed by Rudowicz (1985a). The curly brackets around the parameters and spin operators in  $\mathcal{H}_{ZF}$  in (1) indicate that they are expressed in an original coordinate system.

It is found convenient to use special, different quantisation axes for the electron and nuclear spin operators (Iwasaki 1974, Rockenbauer and Simon 1974). These are obtained from the laboratory axes via multiplication by the matrix **R** to give a new set of axes ( $\xi, \eta, \zeta$ ) for the electron spin:

$$\{S\} = \mathbf{R}[S]_{\xi\eta\zeta} \tag{2}$$

and by the matrix **R'** to give the set of axes ( $\xi', \eta', \zeta'$ ) for the nuclear spin:

$$\{I\} = \mathbf{R}'[I]_{\xi'\eta'\zeta'}. \tag{3}$$

In the choice of the axes ( $\xi, \eta, \zeta$ ) and ( $\xi', \eta', \zeta'$ ), the axis of quantisation for **S** is chosen to be along

$$\hat{\zeta} = \hat{b}^T \cdot \mathbf{g}/g \tag{4}$$

and that for **I** along

$$\hat{\zeta}' = \hat{b}^T \cdot \mathbf{g} \cdot \mathbf{A}/gK. \tag{5}$$

In (4) and (5) a caret denotes a unit vector,

$$\hat{b} = B/B \quad g^2 = \hat{b}^T \cdot \mathbf{g}^T \cdot \mathbf{g} \cdot \hat{b} \quad g^2 K^2 = \hat{b} \cdot \mathbf{g}^T \cdot \mathbf{A}^T \cdot \mathbf{A} \cdot \mathbf{g} \cdot \hat{b}. \tag{6}$$

The transformed spin Hamiltonian can finally be expressed as a sum of zero-order Hamiltonian ( $\mathcal{H}_0$ ) and a perturbation ( $\mathcal{H}'$ ), treating the electronic Zeeman term as diagonal, as follows:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}' \tag{7}$$

where

$$\mathcal{H}_0 = \mu_B Hg[S_\zeta] + [B_2^0][O_2^0] + [B_4^0][O_4^0] + [B_6^0][O_6^0] + K[S_\zeta][I_{\zeta'}] \tag{8}$$

and

$$\begin{aligned} \mathcal{H}' = & \sum_{q=-2}^2 [B_2^q][O_2^q] + \sum_{q=-4}^4 [B_4^q][O_4^q] + \sum_{q=-6}^6 [B_6^q][O_6^q] + P[S_+][I_{\zeta'}] + P^*[S_-][I_{\zeta'}] \\ & + Q[S_+][I_+] + Q^*[S_-][I_-] + R[S_+][I_-] + R^*[S_-][I_+]. \end{aligned} \tag{9}$$

In (9), the prime on the summations indicates that the terms with  $q = 0$  have been omitted, while the asterisk indicates complex conjugate,

$$S_{\pm} = S_{\xi} \pm iS_{\eta} \quad I_{\pm} = I_{\xi'} \pm iI_{\eta'}$$

$$PP^* = \frac{1}{4}(\hat{b}^T \cdot \mathbf{g}^T \cdot \mathbf{A}^T \cdot \mathbf{A} \cdot \mathbf{A}^T \cdot \mathbf{A} \cdot \mathbf{g} \cdot \hat{b}/g^2 K^2 - K^2) \tag{10}$$

$$QQ^* = \frac{1}{16}[\text{Tr}(\mathbf{A}^T \cdot \mathbf{A}) - \hat{b}^T \cdot \mathbf{g}^T \cdot \mathbf{A}^T \cdot \mathbf{A} \cdot \mathbf{A}^T \cdot \mathbf{A} \cdot \mathbf{g} \cdot \hat{b}/g^2 K^2 - (2/K) \det(\mathbf{A})]$$

$$RR^* = \frac{1}{16}[\text{Tr}(\mathbf{A}^T \cdot \mathbf{A}) - \hat{b}^T \cdot \mathbf{g}^T \cdot \mathbf{A}^T \cdot \mathbf{A} \cdot \mathbf{A}^T \cdot \mathbf{A} \cdot \mathbf{g} \cdot \hat{b}/g^2 K^2 + (2/K) \det(\mathbf{A})].$$

The square brackets in (8) and (9) represent the spin operators and their coefficients in the transformed system of axes:  $(\xi, \eta, \zeta)$  for electron spin and  $(\xi', \eta', \zeta')$  for nuclear spin.

The second-order normalised perturbed eigenfunctions of  $\mathcal{H}$ , given by (7), can now be expressed as (Golding 1969)

$$|M, m\rangle = (1 + a_{M\pm N, m\pm n}^{M, m})|M, m\rangle^0 + \sum'_{N, n} c_{M\pm N, m\pm n}^{M, m}|M \pm N, m \pm n\rangle^0 \quad (11)$$

where  $N, n$  are positive integers and the prime on the summation sign indicates that the term with  $N = n = 0$  has been omitted.  $|M', m'\rangle^0$  are the zero-order (unperturbed) eigenvectors of  $\mathcal{H}_0$ , the  $a$ -term in the first bracket in (11) is required for normalisation, and the  $c$ s in (11) are the admixing coefficients of the participating zero-order states of  $S_z$  and  $I_z$ :  $S_z|M, m\rangle^0 = M|M, m\rangle^0$ ;  $I_z|M, m\rangle^0 = m|M, m\rangle^0$ , and are given by

$$a_{M\pm N, m\pm n}^{M, m} = -\frac{1}{2} \sum' \left| \frac{{}^0\langle M \pm N, m \pm n | \mathcal{H}' | M, m \rangle^0}{E_0^{M, m} - E_0^{M \pm N, m \pm n}} \right|^2 \quad (12)$$

and

$$c_{M\pm N, m\pm n}^{M, m} = \frac{{}^0\langle M \pm N, m \pm n | \mathcal{H}' | M, m \rangle^0}{E_0^{M, m} - E_0^{M \pm N, m \pm n}} + \frac{1}{E_0^{M, m} - E_0^{M \pm N, m \pm n}} \times \left( \sum'_{M', m'} \frac{{}^0\langle M \pm N, m \pm n | \mathcal{H}' | M', m' \rangle^{00} \langle M', m' | \mathcal{H}' | M, m \rangle^0}{E_0^{M, m} - E_0^{M', m'}} - \frac{{}^0\langle M \pm N, m \pm n | \mathcal{H}' | M, m \rangle^{00} \langle M, m | \mathcal{H}' | M, m \rangle^0}{E_0^{M, m} - E_0^{M \pm N, m \pm n}} \right). \quad (13)$$

In (12) and (13),

$$E_0^{M, m} = GM + [B_2^0] \mathbb{C}_{M, M}^{20} + [B_4^0] \mathbb{C}_{M, M}^{40} + [B_6^0] \mathbb{C}_{M, m}^{60} + KMm \quad (14)$$

where

$$G = \mu_B g B \quad \mathbb{C}_{M, M}^{k0} = {}^0\langle M | O_k^0 | M \rangle^0. \quad (15)$$

The prime on the summation sign in (12) and (13) indicates that the term with  $M' = M, m' = m$  is to be omitted. The matrix elements required in (12) and (13) can be evaluated using the methods described by Misra and Upreti (1987b), or Skinner and Weil (1978), and the relation (Vrehen and Volger 1965)

$${}^0\langle M' | [O_k^{-q}] | M \rangle^0 = \pm i {}^0\langle M' | [O_k^q] | M \rangle^0, \quad (16)$$

wherein the upper (+) and lower (-) signs apply to  $M' < M, M' > M$ , respectively. The values of some of the important admixture coefficients are given below:

$$\begin{aligned} c_{M+1, m}^{M, m} &= -(1/G)(b_2^1 \mathbb{C}_{M, M+1}^{21} + b_4^1 \mathbb{C}_{M, M+1}^{41} + b_6^1 \mathbb{C}_{M, M+1}^{61} + PS_{0,1} m) \\ c_{M-1, m}^{M, m} &= (1/G)(b_2^1 \mathbb{C}_{M, M-1}^{21} + b_4^1 \mathbb{C}_{M, M-1}^{41} + b_6^1 \mathbb{C}_{M, M-1}^{61} + P^* S_{0,-1} m) \\ c_{M+2, m}^{M, m} &= -(1/2G)(b_2^2 \mathbb{C}_{M, M+2}^{22} + b_4^2 \mathbb{C}_{M, M+2}^{42} + b_6^2 \mathbb{C}_{M, M+2}^{62}) \\ c_{M-2, m}^{M, m} &= (1/2G)(b_2^2 \mathbb{C}_{M, M-2}^{22} + b_4^2 \mathbb{C}_{M, M-2}^{42} + b_6^2 \mathbb{C}_{M, M-2}^{62}) \\ c_{M+1, m+1}^{M, m} &= -(1/G)QS_{0,1}I_{0,1} \\ c_{M-1, m-1}^{M, m} &= (1/G)Q^*S_{0,-1}I_{0,-1} \end{aligned} \quad (17)$$

$$c_{M+1, m-1}^{M, m} = -(R/G)S_{0,1}I_{0,-1}$$

$$c_{M-1, m+1}^{M, m} = (R^*/G)S_{0,-1}I_{0,1}.$$

Here,

$$\begin{aligned} b_k^q &= [B_k^q] + i[B_k^{-q}] & I_{ij}^2 &= [I(I+1) - (m+i)(m+j)] \\ S_{ij}^2 &= [S(S+1) - (M+i)(M+j)] & \mathcal{O}_{M, M \pm q}^{kq} &= \langle M \pm q | O_k^q | M \rangle. \end{aligned} \quad (18)$$

### 3. An operator for the calculation of the intensity of magnetic resonance transitions

The central idea behind the formulation of an operator to calculate the intensities of magnetic resonance transitions is based on the premise that the perturbed eigenstates  $|M, m\rangle$  can be expressed in terms of the zero-order eigenstates  $|M, m\rangle^0$  as follows:

$$|M, m\rangle = \mathcal{F}_{M, m} |M, m\rangle^0 \quad (19)$$

where the operator  $\mathcal{F}_{M, m}$  can be derived using the fact that in the expressions for the perturbed eigenstates, equations (11)–(16), one can write

$$|M \pm N, m \pm n\rangle^0 = \frac{(S_{\pm})^N (I_{\pm})^n |M, m\rangle^0}{\{S_{0, \pm 1} S_{\pm 1, \pm 2} \cdots S_{\pm(N-1), \pm N}\} \{I_{0, \pm 1} I_{\pm 1, \pm 2} \cdots I_{\pm(n-1), \pm n}\}}. \quad (20)$$

Combining (11), (19) and (20), one obtains

$$\mathcal{F}_{M, m} = (1 + a_{M \pm N, m \pm n}^{M, m}) + \sum' C_{M \pm N, m \pm n}^{M, m} (S_{\pm})^N (I_{\pm})^n \quad (21)$$

where

$$C_{M \pm N, m \pm n}^{M, m} = \frac{c_{M \pm N, m \pm n}^{M, m}}{\{S_{0, \pm 1} S_{\pm 1, \pm 2} \cdots S_{\pm(N-1), \pm N}\} \{I_{0, \pm 1} I_{\pm 1, \pm 2} \cdots I_{\pm(n-1), \pm n}\}}. \quad (22)$$

The prime on the summation sign in (21) indicates that the term  $N = n = 0$  is excluded. The coefficients  $c_{M', m'}^{M, m}$  are given by (12) and (13), and the denominators  $S_{ij}$  and  $I_{ij}$  are defined by (18).

The intensity,  $I_{M, m; M', m'}$ , of the transition between the perturbed states  $|M', m'\rangle$  and  $|M, m\rangle$  is proportional to the square of the absolute value of the matrix element of the Hamiltonian,  $\mathcal{H}_{\text{exc}}$ , describing the interaction of the electron magnetic moment with the excitation microwave field, i.e.

$$I_{M, m; M', m'} = \mathcal{H} |\langle M', m' | \mathcal{H}_{\text{exc}} | M, m \rangle|^2 \quad (23)$$

where  $\mathcal{H}$  is a constant, and

$$\mathcal{H}_{\text{exc}} = \mu_B \mathbf{S}^T \cdot \mathbf{g} \cdot \mathbf{B}_1. \quad (24)$$

In (24),  $\mathbf{B}_1$  is the amplitude of the excitation microwave magnetic field ( $= \mathbf{B}_1 \cos \omega t$ ).

Equation (23) can be expressed in terms of the zero-order eigenstates, using (19) and the expression for the bra vector

$$\langle M', m' | = {}^0 \langle M', m' | \mathcal{F}_{M', m'}^\dagger \quad (25)$$

derived from the form of the ket vector  $|M, m\rangle$  given by (19). The expression for the operator  $\mathcal{F}_{M', m'}$  can be written in the same way as that for  $\mathcal{F}_{M, m}$ , which is given by (21). In (25),  $\dagger$  denotes the adjoint of an operator.

Finally,

$$\langle M', m' | \mathcal{H}_{\text{exc}} | M, m \rangle = {}^0 \langle M', m' | \mathcal{F}_{M',m'}^\dagger \mathcal{H}_{\text{exc}} \mathcal{F}_{M,m} | M, m \rangle^0. \quad (26)$$

From (26), the operator whose squared absolute value of matrix-element is proportional to the intensity of the magnetic resonance line, is

$$\mathcal{P}_{M,m;M',m'} = \mathcal{F}_{M',m'}^\dagger \mathcal{H}_{\text{exc}} \mathcal{F}_{M,m}. \quad (27)$$

Equation (27) is the formal expression for the operator,  $\mathcal{P}_{M,m;M',m'}$ . It can be re-written in terms of the raising and lowering operators,  $S_+$  and  $S_-$ , as

$$\begin{aligned} \mathcal{P}_{M,m;M',m'} = & \mu_B [\frac{1}{2}(\mathbf{r}^T \cdot \mathbf{g} \cdot \mathbf{B}_1)(\mathcal{F}_{M',m'}^\dagger S_+ \mathcal{F}_{M,m}) + \frac{1}{2}(\mathbf{l}^T \cdot \mathbf{g} \cdot \mathbf{B}_1)(\mathcal{F}_{M',m'}^\dagger S_- \mathcal{F}_{M,m}) \\ & + (\hat{\zeta}^T \cdot \mathbf{g} \cdot \mathbf{B}_1)(\mathcal{F}_{M',m'}^\dagger S_\zeta \mathcal{F}_{M,m})] \end{aligned} \quad (28)$$

where

$$\mathbf{r} = \hat{\xi} - i\hat{\eta} \quad \mathbf{l} = \hat{\xi} + i\hat{\eta}. \quad (29)$$

In (29)  $\hat{\xi}$  and  $\hat{\eta}$  are the unit vectors along the  $\xi$  and  $\eta$  axes, respectively.

Thus, in order to calculate the EPR line intensity, all one has to do is to evaluate the matrix element of the operator  $\mathcal{P}_{M,m;M',m'}$ , given by (28), between the zero-order simultaneous eigenvectors of  $S_\zeta$  and  $I_\zeta$ ,  $|M', m'\rangle^0$  and  $|M, m\rangle^0$ .

To simplify the evaluation of the EPR line intensity for an arbitrary relative orientation of the external Zeeman and microwave excitation fields, it is convenient to express the intensity given by (23) as

$$I_{M,m;M',m'} = \mathcal{H} \sum_{\alpha,\beta} a_{\alpha\beta} W_{\alpha\beta} \quad (\alpha, \beta = +, - \text{ and } \zeta) \quad (30)$$

where

$$w_{\alpha\beta} = {}^0 \langle M, m | (\mathcal{F}_{M',m'}^\dagger S_\alpha \mathcal{F}_{M,m})^\dagger | M', m' \rangle^0 \langle M', m' | \mathcal{F}_{M',m'}^\dagger S_\beta \mathcal{F}_{M,m} | M, m \rangle^0 \quad (31)$$

and

$$a_{\alpha\beta} = (\mathbf{a}_\alpha^T \cdot \mathbf{g} \cdot \mathbf{B}_1)^* (\mathbf{a}_\beta^T \cdot \mathbf{g} \cdot \mathbf{B}_1). \quad (32)$$

In (32)

$$\mathbf{a}_+ = \frac{1}{2}\mathbf{r} \quad \mathbf{a}_- = \frac{1}{2}\mathbf{l} \quad \hat{\mathbf{a}}_\zeta = \hat{\zeta}. \quad (33)$$

#### 4. An illustrative example

Specific expressions for the intensity operator  $\mathcal{P}_{M,m;M',m'}$  will now be given for the various transitions. The evaluation of  $\mathcal{P}_{M,m;M',m'}$  will first be illustrated for the case of the allowed HF transition  $M - 1, m \leftrightarrow M, m$ ; using similar procedures expressions for  $\mathcal{P}_{M,m;M',m'}$  can be obtained for the forbidden HF transitions.

The calculation involves the evaluation of  $\mathcal{F}_{M-1,m}^\dagger S_\alpha \mathcal{F}_{M,m}$ ;  $\alpha = +, -$  and  $\zeta$ . Using (11) and (20) it is easily seen that

$$\begin{aligned} \mathcal{F}_{M,m} = & 1 + a_{M,m}^{M,m} + C_{M+1,m}^{M,m} S_+ + C_{M-1,m}^{M,m} S_- + C_{M+2,m}^{M,m} S_+^2 + C_{M-2,m}^{M,m} S_-^2 \\ & + (C_{M+1,m+1}^{M,m} S_+ + C_{M-1,m+1}^{M,m} S_- + C_{M,m+1}^{M,m}) I_+ + (C_{M+1,m-1}^{M,m} S_+ \\ & + C_{M-1,m-1}^{M,m} S_- + C_{M,m-1}^{M,m}) I_- \end{aligned} \quad (34)$$



$$\begin{aligned} \mathcal{F}_{M-1,m}^\dagger &= 1 + a_{M-1,m}^{M-1,m^*} + C_{M-1,m}^{M-1,m^*} S_- + C_{M-2,m}^{M-1,m^*} S_+ + C_{M+1,m}^{M-1,m^*} S_-^2 \\ &\quad + C_{M-3,m}^{M-1,m^*} S_+^2 + (C_{M,m+1}^{M-1,m^*} S_- + C_{M-2,m+1}^{M-1,m^*} S_+ + C_{M-1,m+1}^{M-1,m^*}) I_- \\ &\quad + (C_{M,m-1}^{M-1,m^*} S_- + C_{M-2,m-1}^{M-1,m^*} S_+ + C_{M-1,m-1}^{M-1,m^*}) I_+. \end{aligned} \quad (35)$$

Finally, one has, for use in (30) and (31),

$$\begin{aligned} \mathcal{F}_{M-1,m}^\dagger S_+ \mathcal{F}_{M,m} &= (1 + a_{M-1,m}^{M-1,m^*}) C_{M-2,m}^{M,m} S_+ S_-^2 + C_{M,m}^{M-1,m^*} C_{M-1,m}^{M,m} S_- S_+ S_- \\ &\quad + C_{M+1,m}^{M-1,m^*} (1 + a_{M,m}^{M,m}) S_-^2 S_+ + (C_{M,m+1}^{M-1,m^*} C_{M-1,m+1}^{M,m} S_- S_+ S_-) I_- I_+ \\ &\quad + (C_{M,m-1}^{M-1,m^*} C_{M-1,m-1}^{M,m} S_- S_+ S_-) I_+ I_- \end{aligned} \quad (36)$$

$$\begin{aligned} \mathcal{F}_{M-1,m}^\dagger S_- \mathcal{F}_{M,m} &= (1 + a_{M-1,m}^{M-1,m^*}) (1 + a_{M,m}^{M,m}) S_- + C_{M,m}^{M-1,m^*} C_{M+1,m}^{M,m} S_-^2 S_+ \\ &\quad + C_{M-2,m}^{M-1,m^*} C_{M-1,m}^{M,m} S_+ S_-^2 + C_{M+1,m}^{M-1,m^*} C_{M+2,m}^{M,m} S_-^3 S_+^2 \\ &\quad + C_{M-3,m}^{M-1,m^*} C_{M-2,m}^{M,m} S_+^2 S_-^3 + (C_{M,m+1}^{M-1,m^*} C_{M+1,m+1}^{M,m} S_-^2 S_+ \\ &\quad + C_{M-2,m+1}^{M-1,m^*} C_{M-1,m+1}^{M,m} S_+ S_-^2 + C_{M-1,m+1}^{M-1,m^*} C_{M,m+1}^{M,m} S_-) I_- I_+ \\ &\quad + (C_{M,m-1}^{M-1,m^*} C_{M+1,m-1}^{M,m} S_-^2 S_+ + C_{M-2,m-1}^{M-1,m^*} C_{M-1,m-1}^{M,m} S_+ S_-^2 \\ &\quad + C_{M-1,m-1}^{M-1,m^*} C_{M,m-1}^{M,m} S_-) I_+ I_- \end{aligned} \quad (37)$$

and

$$\begin{aligned} \mathcal{F}_{M-1,m}^\dagger S_\zeta \mathcal{F}_{M,m} &= (1 + a_{M-1,m}^{M-1,m^*}) C_{M-1,m}^{M,m} S_\zeta S_- + C_{M,m}^{M-1,m^*} (1 + a_{M,m}^{M,m}) S_- S_\zeta \\ &\quad + C_{M-2,m}^{M-1,m^*} C_{M-2,m}^{M,m} S_+ S_\zeta S_-^2 + C_{M+1,m}^{M-1,m^*} C_{M+1,m}^{M,m} S_-^2 S_\zeta S_+ \\ &\quad + (C_{M,m+1}^{M-1,m^*} C_{M,m+1}^{M,m} S_- S_\zeta + C_{M-1,m+1}^{M-1,m^*} C_{M-1,m+1}^{M,m} S_\zeta S_-) I_- I_+ \\ &\quad + (C_{M,m-1}^{M-1,m^*} C_{M,m-1}^{M,m} S_- S_\zeta + C_{M-1,m-1}^{M-1,m^*} C_{M-1,m-1}^{M,m} S_\zeta S_-) I_+ I_- \end{aligned} \quad (38)$$

The expressions for the operator  $\mathcal{F}_{M',m'}^\dagger S_\alpha \mathcal{F}_{M,m}$  similarly calculated for the forbidden transitions are given below.

(i) Transition  $M-2, m \leftrightarrow M, m$ :

$$\begin{aligned} \mathcal{F}_{M-2,m}^\dagger S_+ \mathcal{F}_{M,m} &= C_{M-3,m}^{M,m} S_+ S_-^3 + C_{M-1,m}^{M-2,m^*} C_{M-2,m}^{M,m} S_- S_+ S_-^2 \\ &\quad + C_{M,m}^{M-2,m^*} C_{M-1,m}^{M,m} S_-^2 S_+ S_- + C_{M+1,m}^{M-2,m^*} S_-^3 S_+ \end{aligned} \quad (39a)$$

$$\begin{aligned} \mathcal{F}_{M-2,m}^\dagger S_- \mathcal{F}_{M,m} &= C_{M-1,m}^{M,m} S_-^2 + C_{M-1,m}^{M-2,m^*} S_-^2 + C_{M-3,m}^{M-2,m^*} C_{M-2,m}^{M,m} S_+ S_-^3 \\ &\quad + C_{M,m}^{M-2,m^*} C_{M+1,m}^{M,m} S_-^3 S_+ + (C_{M-1,m+1}^{M-2,m^*} C_{M,m+1}^{M,m} S_-^2 \\ &\quad + C_{M-2,m+1}^{M-2,m^*} C_{M-1,m+1}^{M,m} S_-^2) I_- I_+ + (C_{M-1,m-1}^{M-2,m^*} C_{M,m-1}^{M,m} S_-^2 \\ &\quad + C_{M-2,m-1}^{M-2,m^*} C_{M-1,m-1}^{M,m} S_-^2) I_+ I_- \end{aligned} \quad (39b)$$

$$\begin{aligned} \mathcal{F}_{M-2,m}^\dagger S_\zeta \mathcal{F}_{M,m} &= C_{M-2,m}^{M,m} S_\zeta S_-^2 + C_{M,m}^{M-2,m^*} S_\zeta S_-^2 + C_{M-1,m}^{M-2,m^*} C_{M-1,m}^{M,m} S_\zeta S_-^2 \\ &\quad + C_{M-1,m+1}^{M-2,m^*} C_{M-1,m+1}^{M,m} S_\zeta S_-^2 I_- I_+ \\ &\quad + C_{M-1,m-1}^{M-2,m^*} C_{M-1,m-1}^{M,m} S_\zeta S_-^2 I_+ I_- \end{aligned} \quad (39c)$$

(ii) Transition  $M-2, m+1 \leftrightarrow M, m$ :

$$\begin{aligned} \mathcal{F}_{M-2,m+1}^\dagger S_+ \mathcal{F}_{M,m} &= (C_{M-3,m+1}^{M,m} S_+ S_-^3 + C_{M,m+1}^{M-2,m+1^*} C_{M-1,m+1}^{M,m} S_-^2 S_+ S_- \\ &\quad + C_{M-1,m}^{M-2,m+1^*} C_{M-2,m}^{M,m} S_- S_+ S_-^2 + C_{M+1,m}^{M-2,m+1^*} S_-^3 S_+) I_+ \end{aligned} \quad (40a)$$

$$\begin{aligned}
 \mathcal{F}_{M-2,m+1}^\dagger S_- \mathcal{F}_{M,m} &= (C_{M-1,m+1}^{M,m} S_-^2 + C_{M-1,m+1}^{M-2,m+1*} C_{M,m+1}^{M,m} S_-^2 \\
 &+ C_{M,m+1}^{M-2,m+1*} C_{M+1,m+1}^{M,m} S_-^3 S_+ + C_{M-3,m}^{M-2,m+1*} C_{M-2,m}^{M,m} S_+ S_-^3 \\
 &+ C_{M-1,m}^{M-2,m+1*} S_-^2 + C_{M-2,m}^{M-2,m+1*} C_{M-1,m}^{M,m} S_-^2) I_+ \\
 &+ C_{M-2,m-1}^{M-2,m+1*} C_{M-1,m-1}^{M,m} S_-^2 I_+^2 I_- .
 \end{aligned} \tag{40b}$$

$$\begin{aligned}
 \mathcal{F}_{M-2,m+1}^\dagger S_\zeta \mathcal{F}_{M,m} &= (C_{M-2,m+1}^{M,m} S_\zeta S_-^2 + C_{M-1,m+1}^{M-2,m+1*} C_{M-1,m+1}^{M,m} S_- S_\zeta S_- \\
 &+ C_{M,m+1}^{M-2,m+1*} C_{M,m+1}^{M,m} S_-^2 S_\zeta + C_{M-1,m}^{M-2,m+1*} C_{M-1,m}^{M,m} S_- S_\zeta S_- \\
 &+ C_{M-2,m}^{M-2,m+1*} C_{M-2,m}^{M,m} S_\zeta S_-^2 + C_{M,m}^{M-2,m+1*} S_-^2 S_\zeta) I_+ .
 \end{aligned} \tag{40c}$$

(iii) Transition  $M - 2, m - 1 \leftrightarrow M, m$ :

$$\begin{aligned}
 \mathcal{F}_{M-2,m-1}^\dagger S_+ \mathcal{F}_{M,m} &= (C_{M-3,m-1}^{M,m} S_+ S_-^3 + C_{M,m-1}^{M-2,m-1*} C_{M-1,m-1}^{M,m} S_-^2 S_+ S_- \\
 &+ C_{M-1,m}^{M-2,m-1*} C_{M-2,m}^{M,m} S_- S_+ S_-^2 + C_{M+1,m}^{M-2,m-1*} S_-^3 S_+) I_- .
 \end{aligned} \tag{41a}$$

$$\begin{aligned}
 \mathcal{F}_{M-2,m-1}^\dagger S_- \mathcal{F}_{M,m} &= (C_{M-1,m-1}^{M,m} S_-^2 + C_{M-1,m-1}^{M-2,m-1*} C_{M,m-1}^{M,m} S_-^2 \\
 &+ C_{M,m-1}^{M-2,m-1*} C_{M+1,m-1}^{M,m} S_-^3 S_+ + C_{M-1,m}^{M-2,m-1*} S_-^2 \\
 &+ C_{M-3,m}^{M-2,m-1*} C_{M-2,m}^{M,m} S_+ S_-^3 + C_{M-2,m}^{M-2,m-1*} C_{M-1,m}^{M,m} S_-^2) I_- \\
 &+ C_{M-2,m+1}^{M-2,m-1*} C_{M-1,m+1}^{M,m} S_-^2 I_-^2 I_+ .
 \end{aligned} \tag{41b}$$

$$\begin{aligned}
 \mathcal{F}_{M-2,m-1}^\dagger S_\zeta \mathcal{F}_{M,m} &= (C_{M-2,m-1}^{M,m} S_\zeta S_-^2 + C_{M-1,m-1}^{M-2,m-1*} C_{M-1,m-1}^{M,m} S_- S_\zeta S_- \\
 &+ C_{M,m-1}^{M-2,m-1*} C_{M,m-1}^{M,m} S_-^2 S_\zeta + C_{M-1,m}^{M-2,m-1*} C_{M-1,m}^{M,m} S_- S_\zeta S_- \\
 &+ C_{M-2,m}^{M-2,m-1*} C_{M-2,m}^{M,m} S_\zeta S_-^2 + C_{M,m}^{M-2,m-1*} S_-^2 S_\zeta) I_- .
 \end{aligned} \tag{41c}$$

(iv) Transition  $M - 1, m - 1 \leftrightarrow M, m$ :

$$\begin{aligned}
 \mathcal{F}_{M-1,m-1}^\dagger S_+ \mathcal{F}_{M,m} &= (C_{M-2,m-1}^{M,m} S_+ S_-^2 + C_{M,m-1}^{M-1,m-1*} C_{M,m-1}^{M,m} S_- S_+ S_- \\
 &+ C_{M,m}^{M-1,m-1*} C_{M-1,m}^{M,m} S_- S_+ S_- + C_{M+1,m-1}^{M-1,m-1*} C_{M-1,m}^{M,m} S_-^2 S_+ \\
 &+ C_{M+1,m}^{M-1,m-1*} S_-^2 S_+ + C_{M-1,m}^{M-1,m-1*} C_{M-2,m}^{M,m} S_+ S_-^2) I_- .
 \end{aligned} \tag{42a}$$

$$\begin{aligned}
 \mathcal{F}_{M-1,m-1}^\dagger S_- \mathcal{F}_{M,m} &= (C_{M,m-1}^{M,m} S_- + C_{M+1,m-1}^{M-1,m-1*} C_{M+1,m-1}^{M,m} S_-^2 S_+ \\
 &+ C_{M-2,m-1}^{M-1,m-1*} C_{M-1,m-1}^{M,m} S_+ S_-^2 + C_{M,m}^{M-1,m-1*} C_{M+1,m}^{M,m} S_-^2 S_+ \\
 &+ C_{M-2,m}^{M-1,m-1*} C_{M-1,m}^{M,m} S_+ S_-^2 + C_{M-1,m}^{M-1,m-1*} S_-) I_- \\
 &+ C_{M-1,m-2}^{M-1,m-1*} C_{M,m-2}^{M,m} S_- I_+ I_-^2 + C_{M-1,m+1}^{M-1,m-1*} C_{M,m+1}^{M,m} S_- I_-^2 I_+ .
 \end{aligned} \tag{42b}$$

$$\begin{aligned}
 \mathcal{F}_{M-1,m-1}^\dagger S_\zeta \mathcal{F}_{M,m} &= (C_{M-1,m-1}^{M,m} S_\zeta S_- + C_{M,m-1}^{M-1,m-1*} C_{M,m-1}^{M,m} S_- S_\zeta \\
 &+ C_{M+1,m-1}^{M-1,m-1*} C_{M+1,m-1}^{M,m} S_-^2 S_\zeta S_+ + C_{M,m}^{M-1,m-1*} S_- S_\zeta \\
 &+ C_{M-1,m}^{M-1,m-1*} C_{M-1,m}^{M,m} S_\zeta S_- + C_{M-2,m}^{M-1,m-1*} C_{M-2,m}^{M,m} S_+ S_\zeta S_-) I_- \\
 &+ C_{M,m-2}^{M-1,m-1*} C_{M,m-2}^{M,m} S_- S_\zeta I_+ I_-^2 \\
 &+ C_{M-1,m+1}^{M-1,m-1*} C_{M-1,m+1}^{M,m} S_\zeta S_- I_-^2 I_+ .
 \end{aligned} \tag{42c}$$

(v) *Transition*  $M - 1, m - 2 \leftrightarrow M, m$ :

$$\begin{aligned} \mathcal{F}_{M-1, m-2}^{\dagger} S_+ \mathcal{F}_{M, m} &= (C_{M-2, m-2}^{M, m} S_+ S_-^2 + C_{M+1, m}^{M-1, m-2^*} S_-^2 S_+ \\ &\quad + C_{M-1, m-2^*}^{M-1, m-2^*} C_{M-2, m}^{M, m} S_+ S_-^2 + C_{M, m-1}^{M-1, m-2^*} C_{M-1, m-1}^{M, m} S_- S_+ S_- \\ &\quad + C_{M+1, m-2}^{M-1, m-2^*} C_{M, m-2}^{M, m} S_-^2 S_+) I_-^2. \end{aligned} \tag{43a}$$

$$\begin{aligned} \mathcal{F}_{M-1, m-2}^{\dagger} S_- \mathcal{F}_{M, m} &= (C_{M, m-2}^{M, m} S_- + C_{M, m-1}^{M-1, m-2^*} C_{M+1, m-1}^{M, m} S_-^2 S_+ \\ &\quad + C_{M-2, m-1}^{M-1, m-2^*} C_{M-1, m-1}^{M, m} S_+ S_-^2 + C_{M-1, m-1}^{M-1, m-2^*} C_{M, m-1}^{M, m} S_- \\ &\quad + C_{M-1, m}^{M-1, m-2^*} S_-) I_-^2. \end{aligned} \tag{43b}$$

$$\begin{aligned} \mathcal{F}_{M-1, m-2}^{\dagger} S_{\zeta} \mathcal{F}_{M, m} &= (C_{M-1, m-2}^{M, m} S_{\zeta} S_- + C_{M, m-2}^{M-1, m-2^*} C_{M, m-2}^{M, m} S_- S_{\zeta} \\ &\quad + C_{M-1, m-1}^{M-1, m-2^*} C_{M-1, m-1}^{M, m} S_{\zeta} S_- + C_{M-1, m-1}^{M-1, m-2^*} C_{M-1, m}^{M, m} S_{\zeta} S_- \\ &\quad + C_{M, m}^{M-1, m-2^*} S_- S_{\zeta} + C_{M, m-1}^{M-1, m-2^*} C_{M, m-1}^{M, m} S_- S_{\zeta}) I_-^2. \end{aligned} \tag{43c}$$

(vi) *Transition*  $M - 1, m + 1 \leftrightarrow M, m$ :

$$\begin{aligned} \mathcal{F}_{M-1, m+1}^{\dagger} S_+ \mathcal{F}_{M, m} &= (C_{M-2, m+1}^{M, m} S_+ S_-^2 + C_{M, m+1}^{M-1, m+1^*} C_{M-1, m+1}^{M, m} S_- S_+ S_- \\ &\quad + C_{M+1, m+1}^{M-1, m+1^*} C_{M, m+1}^{M, m} S_-^2 S_+ + C_{M, m}^{M-1, m+1^*} C_{M-1, m}^{M, m} S_- S_+ S_- \\ &\quad + C_{M-1, m}^{M-1, m+1^*} C_{M-2, m}^{M, m} S_+ S_-^2 + C_{M+1, m}^{M-1, m+1^*} S_-^2 S_+) I_+ . \end{aligned} \tag{44a}$$

$$\begin{aligned} \mathcal{F}_{M-1, m+1}^{\dagger} S_- \mathcal{F}_{M, m} &= (C_{M, m+1}^{M, m} S_- + C_{M, m+1}^{M-1, m+1^*} C_{M+1, m+1}^{M, m} S_-^2 S_+ \\ &\quad + C_{M-2, m+1}^{M-1, m+1^*} C_{M-1, m+1}^{M, m} S_+ S_-^2 + C_{M-2, m}^{M-1, m+1^*} C_{M-1, m}^{M, m} S_+ S_-^2 \\ &\quad + C_{M, m}^{M-1, m+1^*} C_{M+1, m}^{M, m} S_-^2 S_+ + C_{M-1, m}^{M-1, m+1^*} S_-) I_+ \\ &\quad + C_{M-1, m+2}^{M-1, m+1^*} C_{M, m+2}^{M, m} S_- I_+^2 + C_{M-1, m-1}^{M-1, m+1^*} C_{M, m-1}^{M, m} S_- I_+^2 I_- . \end{aligned} \tag{44b}$$

$$\begin{aligned} \mathcal{F}_{M-1, m+1}^{\dagger} S_{\zeta} \mathcal{F}_{M, m} &= (C_{M-1, m+1}^{M, m} S_{\zeta} S_- + C_{M, m+1}^{M-1, m+1^*} C_{M, m+1}^{M, m} S_- S_{\zeta} \\ &\quad + C_{M+1, m+1}^{M-1, m+1^*} C_{M+1, m+1}^{M, m} S_-^2 S_{\zeta} S_+ + C_{M-2, m}^{M-1, m+1^*} C_{M-2, m}^{M, m} S_+ S_{\zeta} S_-^2 \\ &\quad + C_{M, m}^{M-1, m+1^*} S_- S_{\zeta} + C_{M-1, m}^{M-1, m+1^*} C_{M-1, m}^{M, m} S_{\zeta} S_-) I_+ \\ &\quad + C_{M, m+2}^{M-1, m+1^*} C_{M, m+2}^{M, m} S_- S_{\zeta} I_+^2 \\ &\quad + C_{M-1, m-1}^{M-1, m+1^*} C_{M-1, m-1}^{M, m} S_{\zeta} S_- I_+^2 I_- . \end{aligned} \tag{44c}$$

(vii) *Transition*  $M - 1, m + 2 \leftrightarrow M, m$ :

$$\begin{aligned} \mathcal{F}_{M-1, m+2}^{\dagger} S_+ \mathcal{F}_{M, m} &= (C_{M-2, m+2}^{M, m} S_+ S_-^2 + C_{M+1, m+2}^{M-1, m+2^*} C_{M, m+2}^{M, m} S_-^2 S_+ \\ &\quad + C_{M, m+1}^{M-1, m+2^*} C_{M-1, m+1}^{M, m} S_- S_+ S_- + C_{M-1, m}^{M-1, m+2^*} C_{M-2, m}^{M, m} S_+ S_-^2 \\ &\quad + C_{M+1, m}^{M-1, m+2^*} S_-^2 S_+) I_+^2. \end{aligned} \tag{45a}$$

$$\begin{aligned} \mathcal{F}_{M-1, m+2}^{\dagger} S_- \mathcal{F}_{M, m} &= (C_{M, m+2}^{M, m} S_- + C_{M-2, m+1}^{M-1, m+2^*} C_{M-1, m+1}^{M, m} S_+ S_-^2 \\ &\quad + C_{M, m+1}^{M-1, m+2^*} C_{M+1, m+1}^{M, m} S_-^2 S_+ + C_{M-1, m+1}^{M-1, m+2^*} C_{M, m+1}^{M, m} S_- \\ &\quad + C_{M-1, m}^{M-1, m+2^*} S_-) I_+^2. \end{aligned} \tag{45b}$$

$$\begin{aligned} \mathcal{F}_{M-1, m+2}^{\dagger} S_{\zeta} \mathcal{F}_{M, m} &= (C_{M-1, m+2}^{M, m} S_{\zeta} S_- + C_{M, m+2}^{M-1, m+2^*} C_{M, m+2}^{M, m} S_- S_{\zeta} \\ &\quad + C_{M, m+1}^{M-1, m+2^*} C_{M, m+1}^{M, m} S_- S_{\zeta} + C_{M-1, m+1}^{M-1, m+2^*} C_{M-1, m+1}^{M, m} S_{\zeta} S_- \\ &\quad + C_{M-1, m}^{M-1, m+2^*} C_{M-1, m}^{M, m} S_{\zeta} S_- + C_{M, m}^{M-1, m+2^*} S_- S_{\zeta}) I_+^2. \end{aligned} \tag{45c}$$

### 5. Comparison with previous results

In order to compare the present results with those reported previously, computed by the use of perturbed wavefunctions by Golding *et al* (1972), Golding and Tennant (1974), Mialhe and Erbeia (1973a) and Mialhe and Queduc (1976) (hereafter referred to as G1, G2, M1 and M2, respectively) the intensities were calculated for the various possible transitions using (30). G1 and G2 used the (conventional) direction  $\hat{b}^T \cdot \mathbf{g} \cdot \mathbf{A}$  as the axis of quantisation for the nuclear spin, whereas M1 and M2 used the direction of the effective magnetic field, which is different for different electronic states, defined by

$$B_{\text{eff}}^\beta = \sum_\alpha (\mu_n g_n)^{-1} A_{\alpha\beta} \langle M | S_\alpha | M \rangle \quad \alpha, \beta = x, y, z$$

as the axis of quantisation for the nuclear spin.

The intensities are now calculated, for the case where the microwave excitation field ( $\mathbf{B}_1$ ) is perpendicular to the Zeeman field ( $\mathbf{B}$ ), using (30), assuming the principal axes of  $\mathbf{g}$ ,  $\mathbf{D}$  and  $\mathbf{A}$  tensors to be coincident and assuming an axial  $\mathbf{g}$  tensor for the allowed transitions and an isotropic  $\mathbf{g}$  tensor for the forbidden transitions, in accordance with those used previously (G1, G2, M1, and M2). The explicit intensity expressions are listed as follows:

(i)  $\Delta M = 1, \Delta m = 0$

$$\begin{aligned}
 I_{M,m;M-1,m} &= \mathcal{H} |^0 \langle M-1, m | \mathcal{P} | M, m \rangle|^2 \\
 &= \mathcal{H} \left| \left\{ S_{0,-1} \left[ 1 + \frac{\rho}{2G} (S_{-1,-2}^2 - S_{0,1}^2) \right] + \left( \frac{\lambda(2M-1) + 2Pm}{4G^2} \right) \right. \right. \\
 &\quad \times \{ S_{0,1}^2 [\lambda(2M+1) + 2Pm] - 2S_{0,-1}^2 [\lambda(2M-1) + 2Pm] \\
 &\quad + S_{-1,-2}^2 [\lambda(2M-3) + 2Pm] \} \\
 &\quad + (\lambda/4G^2) \{ S_{-1,-2}^2 [\lambda(2M-3) + 2Pm] - S_{0,1}^2 [\lambda(2M+1) + 2Pm] \} \\
 &\quad + (\rho^2/8G^2) [S_{0,1}^2 (S_{1,2}^2 - S_{0,-1}^2) - S_{-1,-2}^2 (S_{0,-1}^2 - S_{-2,-3}^2)] \\
 &\quad + (Q^2/2G^2) [I_{0,1}^2 (S_{0,1}^2 - S_{0,-1}^2) - I_{0,-1}^2 (S_{0,-1}^2 - S_{-1,-2}^2)] \\
 &\quad + (R^2/2G^2) [I_{0,-1}^2 (S_{0,1}^2 - S_{0,-1}^2) - I_{0,1}^2 (S_{0,-1}^2 - S_{-1,-2}^2)] \\
 &\quad + (QR/2G^2) [(S_{0,1}^2 - 2S_{0,-1}^2 + S_{-1,-2}^2) (I_{0,1}^2 + I_{0,-1}^2)] \\
 &\quad + (\rho/2G^2) \{ S_{0,1}^2 (2\sigma M + Km) - S_{-1,-2}^2 [2\sigma(M-1) + Km] \} \\
 &\quad \left. \left. + \mathcal{C}_{M,m+1}^{M,m} \mathcal{C}_{M-1,m+1}^{M-1,m^*} + \mathcal{C}_{M,m-1}^{M,m} \mathcal{C}_{M-1,m-1}^{M-1,m^*} \right\} \right|^2 \tag{46}
 \end{aligned}$$

where  $\mathcal{H}$  is a constant of proportionality.

(ii)  $\Delta M = 1, |\Delta m| = 1$

$$\begin{aligned}
 I_{M,m;M-1,m-1} &= \mathcal{H} \left| S_{0,-1} I_{0,-1} \left\{ \frac{\lambda}{2G} \left[ (1 - a_{M-1,m-1}^{M-1,m-1}) \left( \frac{S(S+1) - 3M^2}{M} \right) \right. \right. \right. \\
 &\quad \left. \left. - (1 - a_{M,m}^{M,m}) \left( \frac{S(S+1) - 3(M-1)^2}{M-1} \right) \right] - \frac{\lambda A}{2G^2} (2M-1) \right\} \right|^2
 \end{aligned}$$

$$\begin{aligned} & \times [S(S+1) - M^2 + M - 1] - \frac{\lambda\rho}{4G^2} \left[ S_{0,1}^2 \left( \frac{S(S+1) - 3M^2}{M} \right) \right. \\ & \left. + S_{-1,-2}^2 \left( \frac{S(S+1) - 3(M-1)^2}{M-1} \right) \right] \Big|^2 \end{aligned} \tag{47}$$

$$\begin{aligned} I_{M,m;M-1,m+1} &= \mathcal{H} \left| S_{0,-1} I_{0,1} \left\{ \frac{\lambda}{2G} \left[ (1 - a_{M,m}^{M,m}) \left( \frac{S(S+1) - 3(M-1)^2}{M-1} \right) \right. \right. \right. \\ & \left. \left. - (1 - a_{M-1,m+1}^{M-1,m+1}) \left( \frac{S(S+1) - 3M^2}{M} \right) \right] - \frac{\lambda A}{2G^2} \right. \\ & \left. \times (2M-1)[S(S+1) - M^2 + M - 5] + \frac{\lambda\rho}{4G^2} \left[ S_{0,1}^2 \left( \frac{S(S+1) - 3M^2}{M} \right) \right. \right. \\ & \left. \left. + S_{-1,-2}^2 \left( \frac{S(S+1) - 3(M-1)^2}{M-1} \right) \right] \right\} \Big|^2. \end{aligned} \tag{48}$$

(iii)  $\Delta M = 1, |\Delta m| = 2$

$$\begin{aligned} I_{M,m;M-1,m-2} &= \mathcal{H} \left| -\frac{\lambda^2}{4G^2} S_{0,-1} I_{0,-1} I_{-1,-2} \right. \\ & \left. \times \left( \frac{S(S+1) - 3(M-1)^2}{M-1} \right) \left( \frac{S(S+1) - 3M^2}{M} \right) \right|^2 \end{aligned} \tag{49}$$

$$\begin{aligned} I_{M,m;M-1,m+2} &= \mathcal{H} \left| -S_{0,-1} I_{0,1} I_{1,2} \left[ \frac{\lambda^2}{4G^2} \left( \frac{S(S+1) - 3(M-1)^2}{M-1} \right) \right. \right. \\ & \left. \left. \times \left( \frac{S(S+1) - 3M^2}{M} \right) - \frac{R^2}{2G^2} [S(S+1) - M^2 + 3M] \right] \right|^2. \end{aligned} \tag{50}$$

(iv)  $\Delta M = 2, |\Delta m| = 0$

$$\begin{aligned} I_{M,m;M-2,m} &= \mathcal{H} \left| S_{0,-1} S_{-1,-2} \left\{ -\lambda/G + (\lambda\rho/4G^2)[S_{-2,-3}^2(2M-5) \right. \right. \\ & \left. \left. + S_{0,1}^2(2M+1)] + (\rho Pm/2G^2)(S_{-2,-3}^2 + S_{01}^2) \right. \right. \\ & \left. \left. - (\rho/12G^2)\{S_{0,1}^2[\lambda(6M-5) + 6Pm] + S_{-2,-3}^2[\lambda(6M-7) + 6Pm]\} \right. \right. \\ & \left. \left. + (1/G^2)[4\lambda\sigma(M-1) + (\lambda K + 2P\sigma)m] \right. \right. \\ & \left. \left. + (1/G)[I_{0,1}(R\mathcal{C}_{M-2,m+1}^{M-2,m*} - Q\mathcal{C}_{M,m+1}^{M,m}) \right. \right. \\ & \left. \left. + I_{0,-1}(Q\mathcal{C}_{M-2,m-1}^{M-2,m*} - R\mathcal{C}_{M,m-1}^{M,m}) \right] \right\} \Big|^2. \end{aligned} \tag{51}$$

(v)  $\Delta M = 2, |\Delta m| = 1$

$$\begin{aligned} I_{M,m;M-2,m-1} &= \mathcal{H} \left| S_{0,-1} S_{-1,-2} I_{0,-1} \right. \\ & \left. \times \left\{ \frac{\rho}{2G^2} (Q - R)(S_{0,1}^2 + S_{-2,-3}^2 - S_{0,-1}^2 - S_{-1,-2}^2) \right. \right. \end{aligned}$$

$$\begin{aligned}
 & -\frac{Q}{G^2}(2\sigma + K) + \frac{\lambda^2(Q + R)}{2G^2K} \left[ (2M - 3) \left( \frac{S(S + 1) - 3M^2}{M} \right) \right. \\
 & \left. + (2M - 1) \left( \frac{S(S + 1) - 3(M - 2)^2}{M - 2} \right) \right] \Bigg|^2 \tag{52}
 \end{aligned}$$

$$\begin{aligned}
 I_{M,m;M-2,m+1} = & \mathcal{H} \left| -S_{0,-1}S_{-1,-2}I_{0,1} \right. \\
 & \times \left\{ \frac{\rho}{2G^2}(Q - R)(S_{0,1}^2 + S_{-2,-3}^2 - S_{0,-1}^2 - S_{-1,-2}^2) \right. \\
 & + \frac{R}{G^2}(2\sigma - K) + \frac{\lambda^2(Q + R)}{2G^2K} \left[ (2M - 3) \left( \frac{S(S + 1) - 3M^2}{M} \right) \right. \\
 & \left. \left. + (2M - 1) \left( \frac{S(S + 1) - 3(M - 2)^2}{M - 2} \right) \right] \right\} \Bigg|^2. \tag{53}
 \end{aligned}$$

In (46)–(53)  $g_{\parallel}$ ,  $g_{\perp}$ ,  $D$ ,  $A$ ,  $B$  are the usual SH parameters,  $\theta$  is the angle between the Zeeman field and the principal magnetic  $z$  axis and

$$\begin{aligned}
 \sigma &= \frac{D}{2}(3 \cos^2 \varphi - 1) & g^2 &= g_{\parallel}^2 \cos^2 \theta + g_{\perp}^2 \sin^2 \theta \\
 \lambda &= D \sin \varphi \cos \varphi & g^2 K^2 &= g_{\parallel}^2 A^2 \cos^2 \theta + g_{\perp}^2 B^2 \sin^2 \theta \\
 \rho &= \frac{D}{4} \sin^2 \varphi & P &= g_{\parallel} g_{\perp} \frac{(A^2 - B^2)}{2G^2 K} \sin \theta \cos \theta \\
 \tan \varphi &= \frac{g_{\perp}}{g_{\parallel}} \tan \theta & Q &= \frac{B(A - K)}{4K} & R &= \frac{B(A + K)}{4K}
 \end{aligned}$$

$$\begin{aligned}
 c_{M,m+1}^{M,m} &= -(I_{0,1}/GKM) \{ (Q + R) [\lambda \{ S(S + 1) - 3M^2 \} - 2Pm] \\
 & \quad + P(S_{0,1}^2 Q - S_{0,-1}^2 R) \} \\
 c_{M,m-1}^{M,m} &= (I_{0,-1}/GKM) \{ (Q + R) [\lambda \{ S(S + 1) - 3M^2 \} - 2Pm] \\
 & \quad + P(S_{0,-1}^2 Q - S_{0,1}^2 R) \} \\
 a_{M,m}^{M,m} &= (1/8G^2) \{ S_{0,1}^2 [\lambda(2M + 1) + 2Pm]^2 + S_{0,-1}^2 [\lambda(2M - 1) + 2Pm]^2 \\
 & \quad + \rho^2 (S_{0,1}^2 S_{1,2}^2 + S_{0,-1}^2 S_{-1,-2}^2) + 4Q^2 (S_{0,1}^2 I_{0,1}^2 + S_{0,-1}^2 I_{0,-1}^2) \\
 & \quad + 4R^2 (S_{0,1}^2 I_{0,-1}^2 + S_{0,-1}^2 I_{0,1}^2) \}. \tag{54}
 \end{aligned}$$

The above results, given by (46)–(53), are now compared with those published previously ( $G_1, G_2, M_1$  and  $M_2$ ). A detailed comparison with  $G_1$  and  $G_2$  is entirely possible because the axis of quantisation for  $I$  used there is the same as that used in the present paper. Comparison with  $M_1, M_2$  can be made only for the transitions  $\Delta M = 1, \Delta m = 0$ ;  $\Delta M = 2, \Delta m = 0$ , because, as remarked earlier, the axis of quantisation for  $I$  used here is not the same as that used in  $M_1$  and  $M_2$ . It is to be noted that several extra terms appear in the intensity expressions given by (46)–(53) of the present paper, over and above those given by  $G_1$  and  $G_2$ . It is further to be noted that, contrary to what is stated in  $G_1$  and  $G_2$ , the intensity of the transition  $M', m + n \leftrightarrow M, m$  is not, in fact, equal to that for

the transition  $M', m - n \leftrightarrow M, m$ . With this general observation, detailed comparison of the present results with those of G1, G2, M1 and M2 is given below.

(i)  $\Delta M = 1, \Delta m = 0$ . The present result is in agreement with that of G1, except for the absence of six important second-order terms in G1, caused by some possible admixing coefficients not being considered in G1. The following terms in the present paper are absent in G1:

$$\begin{aligned} & - (\rho^2/8G^2)[S_{0,1}^2(S_{0,-1}^2 + S_{1,2}^2) + S_{-1,-2}^2(S_{0,-1}^2 + S_{-2,-3}^2)] \\ & + (\lambda^2/4G^2)[S_{0,1}^2(2M+1)(2M-1) + S_{-1,-2}^2(2M-1)(2M-3)] \\ & - (\lambda^2/8G^2)[2S_{0,-1}^2(2M-1)^2 + S_{0,1}^2(2M+1)^2 + S_{-1,-2}^2(2M-3)^2]. \end{aligned}$$

As for M1, their intensity expression is incorrect, since the operator expression in M1 is not correct. For, the intensity, calculated using the operator given in M1, does not simplify to the result of Bir (1964) for the case where the HF interaction is zero. This was further confirmed by an independent calculation, in which the axis of quantisation for  $I$  was chosen to be the same as that used in M1, namely, that along the effective magnetic field at the nuclear site. The correct operator expression of M1 should have been:

$$\begin{aligned} & \{1 + (\rho/G)(2S_z - 1) + (\lambda^2 A^2 B^2 / 2G^2 K^4) \\ & \quad \times [S_z^{-1}(S_z - 1)^{-1}(3S_z^2 - S^2)(3S_z^2 - 6S_z - S^2 + 3)](I^2 - I_z^2) \\ & \quad - (\lambda^2 A^2 B^2 / 4G^2 K^4)[S_z^{-2}(3S_z^2 - S^2)^2 \\ & \quad + (S_z - 1)^{-2}(3S_z^2 - 6S_z - S^2 + 3)^2](I^2 - I_z^2) \\ & \quad - (\lambda^2/G^2)(S^2 + 3S_z^2 - 3S_z) - (\rho^2/G^2)(S^2 - 3S_z^2 + 3S_z - \frac{3}{2})\}S_+. \end{aligned}$$

The fourth term of the above expression does not appear in (46) of the present paper. This occurs due to the use of a different axis of quantisation for  $I$  in M1 from that used in the present paper; the latter is described by (5).

(ii)  $\Delta M = 1, \Delta m = +1$ . The present result given by (47) agrees completely with that of G1.

(iii)  $\Delta M = 1, \Delta m = -1$ . The present result is not in agreement with that of G1, wherein  $I_{M,m;M-1,m+1} = I_{M,m;M-1,m-1}$ . This is because four extra terms that contribute in second order were omitted by G1.

(iv)  $\Delta M = 1, \Delta m = +2$ . The result of G1 is incorrect; the factor of four in G1 should not be present in the numerator.

(v)  $\Delta M = 1, \Delta m = -2$ . The present result is not in agreement with that of G1. In addition to a missing factor of four in the numerator in G1, the term

$$-S_{0,-1}I_{0,1}I_{1,2}(R^2/2G^2)[S(S+1) - M^2 + 3M]$$

of the present paper is absent in G1.

(vi)  $\Delta M = 2, \Delta m = 0$ . The factor of 8 in the denominator in G2 of the second-order terms should be 16. Moreover, not all the second-order admixture coefficients have been taken into account. The present result is, however, in agreement with that of M2, except that the last term of (51) of the present paper is absent in M2. This is because the axis of quantisation for  $I$  in the present paper is different from that used in M2.

(vii)  $\Delta M = 2$ ,  $\Delta m = +1$ . G2 is incorrect. Only some of the second-order admixture coefficients have been taken into account; also, in G2, the sign of the term in  $aA$  (in the notation of G2) is incorrect.

(viii)  $\Delta M = 2$ ,  $\Delta m = -1$ . Within limits of the approximations used, according to G2,  $I_{M,m;M-2,m+1} \neq I_{M,m;M-2,m-1}$ . The result in the present paper, in which all the second-order admixture coefficients are considered, agrees only with this finding of G2; the intensity expression of G2 for this case does not agree with the present result, given by (53).

(ix) *Diagram technique*. A technique involving the use of diagrams, which represent the various mathematical symbols and operators, to evaluate the admixture coefficients  $c_{M\pm N, m\pm n}^{M,m}$  required in the perturbation expressions, and the coefficients  $a_{\alpha\beta}$ , has been described by Skinner and Weil (1976, 1978) and by Fulton *et al* (1986, 1988). An advantage of this technique is that the various non-zero terms required for the calculation of the intensity can be identified in a systematic way. An apparent drawback of this method is that it takes a great deal of effort to learn—much more than is required for using the formalism proposed in the present paper. Furthermore, in spite of the modifications proposed by the authors, very similar diagrams are used to represent the matrix elements  $\langle M+1, m | S_+ S_z | M, m \rangle$ ,  $\langle M+1, m | S_z S_+ | M, m \rangle$ , and  $\langle M+1, m | S_+ I_z | M, m \rangle$ , which can lead to confusion. No specific intensity expressions were given by Fulton *et al* (1986), other than for the allowed transition  $M, m \leftrightarrow M-1, m$ , which is proportional to  $[S(S+1) - M(M-1)]$ . It was, therefore, not possible to compare the present results with those of Fulton *et al* (1986). However, the diagram technique was used by Skinner and Weil (1976) to determine specifically the third-order perturbed energy for the simple Hamiltonian  $\mathcal{H} = \mu_B \mathbf{S}^T \cdot \mathbf{g} \cdot \mathbf{B} + \mathbf{S}^T \cdot \mathbf{A} \cdot \mathbf{I}$ . The same energy calculated to third order in perturbation using the quantum mechanical prescription, as in the present paper, yielded the last term in (27) of Skinner and Weil (1976) to be  $-K(k^2 - K^2)Mm^3$  instead of  $K(k^2 - K^2)[S(S+1) - M^2]m^3$ . A further discrepancy is found in (20.4) of Skinner and Weil (1976), which should be corrected to read

$$6 \text{Det}(A_{pq}) = 2 \text{Tr}[A_{pq} \text{Adj}(A_{pq})].$$

## 6. Concluding remarks

Operator expressions have been developed in the present paper to calculate the intensities of the allowed and forbidden EPR HF transitions:  $\Delta M = 1$ ,  $\Delta m = 0, \pm 1, \pm 2$  and  $\Delta M = 2$ ,  $\Delta m = 0, \pm 1$ . These operator expressions are completely general, in that they can be used for the lowest symmetry and for arbitrary orientation of the external Zeeman field with respect to the excitation microwave field. The non-zero coefficients of the operators in the spin Hamiltonian conforming to specific symmetries are listed in table 1. The intensity operator is straightforward to use since all that one has to do here is to calculate its matrix elements with respect to the zero-order eigenstates corresponding to the (perturbed) eigenstates participating in the resonance. Using these general operators, explicit intensity expressions have been derived for the various commonly observed EPR transitions. The limitations as well as errors/omissions of the previously derived expressions have been discussed. By excluding the nuclear interaction terms in the spin Hamiltonian, the present results can be reduced to calculate the intensities of purely electronic transitions.



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## References

- Abragam A and Bleaney B 1970 *Electron Paramagnetic Resonance of Transition Ions* (Oxford: Clarendon) p 174
- Al'tshuler S A and Kozyrev B M 1974 *EPR in Compounds of Transition Elements* (New York: Wiley)
- Bir G L 1964 *Sov. Phys.—Solid State* **5** 1628
- Bleaney B and Rubins R S 1961 *Proc. Phys. Soc.* **78** 778
- Buckmaster H A, Chatterjee R and Shing Y H 1972 *Phys. Status Solidi* **a** **13** 9
- Fulton D B, Skinner R and Weil J A 1986 *J. Magn. Reson.* **69** 41
- 1988 unpublished
- Golding R M 1969 *Applied Wave Mechanics* (London: Van Nostrand) p 22
- Golding R M, Newman R H, Rae A D and Tennant W C 1972 *J. Chem. Phys.* **57** 1912
- Golding R M and Tennant W C 1974 *Mol. Phys.* **28** 167
- Iwasaki M 1974 *J. Magn. Reson.* **16** 417
- Korkmaz M and Aktas B 1984 *J. Phys. Chem. Solids* **45** 259
- Lupei V, Lupei A and Ursu I 1972 *Phys. Rev. B* **6** 4125
- Mialhe P 1973 *J. Phys. Chem. Solids* **34** 217
- Mialhe P and Erbeja A 1973a *Phys. Rev. B* **7** 4061
- 1973b *J. Phys. C: Solid State Phys.* **6** 1965
- Mialhe P and Quedec P 1976 *Phys. Rev. B* **14** 2757
- Misra S K 1979 *J. Phys. C: Solid State Phys.* **12** 5221
- Misra S K and Upreti G C 1987a *Magn. Reson. Rev.* **12** 1
- 1987b *Electronic Magnetic Resonance of the Solid State* ed. J A Weil (Ottawa: The Canadian Society for Chemistry) p 69
- Orton J W 1968 *Electron Paramagnetic Resonance* (London: Iliffe) p 68
- Rockenbauer A and Simon P 1974 *Mol. Phys.* **28** 1113
- Rudowicz C 1985a *J. Phys. C: Solid State Phys.* **18** 1415
- 1985b *J. Magn. Reson.* **63** 85
- 1987 *Magn. Reson. Rev.* **13** 1
- Skinner R and Weil J A 1976 *J. Magn. Reson.* **21** 271
- 1978 *J. Magn. Reson.* **29** 223
- Vrehen Q H F and Volger J 1965 *Physica* **31** 845
- Weil J A 1987 *Electronic Magnetic Resonance of the Solid State* ed J A Weil (Ottawa: The Canadian Society for Chemistry) p 1
- Weil J A and Anderson J H 1961 *J. Chem. Phys.* **35** 1410