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A group theoretical study of f^n systems in an environment of approximately icosahedral symmetry

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Group theoretical methods are developed to determine in a generalised format for an f^n ion in a crystal field environment of icosahedral symmetry, the effect on the energy level scheme when a crystal field distortion is considered parallel to any direction. As an illustration, the effect on the g-tensor components are examined as a function of the magnitude and the direction of the crystal field distortion. All appropriate reduced matrix elements in group theoretical terminology are evaluated for the f^n-ion ground states. Specific results are given for the f^3-ion case and compared with electron paramagnetic resonance, optical, and magnetic susceptibility data.

Key words: f^n-systems—icosahedral symmetry—g-tensors—generalised crystal field distortions—e.p.r.

1. Introduction

Often in the interpretation of e.p.r. and magnetic susceptibility data, for a specific f^n-system the f^n-ion is assumed to be in an environment defined by a low symmetry crystal field potential expressed as a series of spherical harmonics with a number of unknown parameters.

The parameters are adjusted to reflect the experimental results but usually the solution is not unique. Another approach is to examine the data assuming the crystal field environment is close to octahedral or icosahedral symmetry. By

commencing with a high symmetry and then applying a small distortion a generalised approach using group theoretical methods is possible. This approach enables a quick analysis of the experimental data and indicates if there is a need for more detailed analysis. In this paper we develop a group theoretical method which generalises the g-tensor for f^n systems in a crystal field environment of approximately icosahedral symmetry.

The idea of using icosahedral symmetry in handling f^n-systems was first suggested by Judd [1], who discussed the possibility that the appropriate symmetry in the rare-earth double nitrates, was predominantly that of the icosahedral group which is, to a small extent, modified to the symmetry of its subgroup C_{3v}. Judd based his assumption on the relationships found for the crystal field parameters rather than on the detailed structure of these compounds.

He found that the last three terms in the C_{3v} crystal field hamiltonian

$$\begin{aligned}\mathcal{H} = {} & A_2^0(3z^2-r^2)+A_4^0(35z^4-30r^2z^2+3r^4)\\ & +A_4^3z(x^3-3xy^2)+A_6^0(231z^6-315r^2z^4+105r^4z^2-5r^6)\\ & +A_6^3(11z^3-3zr^2)(x^3-3xy^2)\\ & +A_6^6(x^6-15x^4y^2+15x^2y^4-y^6)\end{aligned} \tag{1}$$

were major interactions whose ratios were quite close to those found for a crystal field of icosahedral symmetry i.e.

$$A_6^3/A_6^0 = \pm 14\sqrt{5}, \qquad A_6^6/A_6^0 = 14.$$

The other terms played a much smaller role. More recent studies [2–5] by X-ray analysis have shown a number of lanthanide and actinide compounds with symmetry close to icosahedral symmetry. Indeed, the deviations from icosahedral symmetry of the cerium compound described by Dexter and Silverton [4] are smaller than the deviations often neglected when studying compounds with octahedral symmetry.

2. Theory

The crystal field hamiltonian of icosahedral symmetry may be expressed as

$$\mathcal{H} = \left\{\frac{\sqrt{11}}{5}\,Y_{60}(\Theta,\phi) - \frac{\sqrt{7}i}{5}\left[Y_{65}(\Theta,\phi)+Y_{6-5}(\Theta,\phi)\right]\right\}\frac{6\sqrt{143}\sqrt{\pi}\Delta}{35} \tag{2}$$

where Δ is the appropriate crystal field parameter such that when spin-orbit coupling is neglected, the splitting of the f^1-system by the crystal field is Δ. The crystal field hamiltonian given by (2) defines the z-axis along the fivefold axis and is the appropriate form for use in group theory methodology. Nevertheless, Eq. (2) may be rotated so that the z-axis is parallel to a threefold axis, and this

yields the following Hamiltonian:

$$D(0,-\beta_3,-\pi/2)\mathscr{H}=\frac{-\sqrt{11}}{9}\left\{Y_{60}(\Theta,\phi)-\frac{\sqrt{7}}{\sqrt{3}}[Y_{63}(\Theta,\phi)-Y_{6-3}(\Theta,\phi]\right.$$
$$\left.-\frac{2\sqrt{7}}{\sqrt{33}}[Y_{66}(\Theta,\phi)+Y_{6-6}(\Theta,\phi)]\right\}\frac{6\sqrt{143}\sqrt{\pi}\Delta}{35} \tag{3}$$

where

$$\beta_3=\cos^{-1}\left\{\frac{1}{3}+\frac{2}{3\sqrt{5}}\right\}^{1/2}.$$

Similarly, choosing the z-axis along a two-fold axis yields the following Hamiltonian.

$$D(0,-\beta_2,-\pi/2)\mathscr{H}=\frac{\sqrt{11}}{16}\left\{Y_{60}(\Theta,\phi)+\frac{\sqrt{21}}{2}[Y_{62}(\Theta,\phi)+Y_{6-2}(\Theta,\phi)]\right.$$
$$-\frac{\sqrt{7}}{\sqrt{2}}[Y_{64}(\Theta,\phi)+Y_{6-4}(\Theta,\phi)]$$
$$\left.-\frac{\sqrt{105}}{2\sqrt{11}}[Y_{66}(\Theta,\phi)+Y_{6-6}(\Theta,\phi)]\right\}\frac{6\sqrt{143}\sqrt{\pi}\Delta}{35} \tag{4}$$

where $\beta_2=\frac{1}{2}\cos^{-1}(-1/\sqrt{5})$ and $D(\alpha,\beta,\gamma)$ is the rotation operator defined in Ref. [6].

One method for examining a distortion from icosahedral symmetry is to consider a crystal field distortion component, expressed in terms of spherical harmonics, as

$$\mathscr{H}=a_2Y_{20}(\Theta,\phi)+a_4Y_{40}(\Theta,\phi)+a_6Y_{60}(\Theta,\phi). \tag{5}$$

To examine the effect of this distortion it is first necessary to determine the eigenfunctions, for a specific f^n case, of Eqs. (2), (3) or (4), depending on whether the distortion is to be considered parallel to a five, three or twofold axis respectively.

To generalise the calculations by using group theoretical symmetry coupling coefficients [7] it is necessary to define that the z-axis is parallel to the fivefold axis and consider the crystal field distortion component at a general angle to the axis system defined by Eq. (2).

Note that in dealing with even J-values, in which only the case where $\gamma=0$ need be considered, account must be taken of terms given in the expression

$$D(\alpha,\beta,0)Y_{J0}(\Theta,\phi)=C_J(Aa)|Aa\rangle+C_J(U\kappa)|U\kappa\rangle+C_J^*(U\kappa)|U\nu\rangle$$
$$+C_J(U\lambda)|U\lambda\rangle+C_J^*(U\lambda)|U\mu\rangle$$
$$+C_J(V2)|V2\rangle+C_J^*(V2)|V-2\rangle$$
$$+C_J(V1)|V1\rangle+C_J^*(V1)|V-1\rangle+C_J(V0)|V0\rangle \tag{6}$$

where $C_J^*(\Gamma a)$ is the complex conjugate of $C_J(\Gamma a)$.

The crystal field distortion matrix elements may be generalised using

$$\langle \Gamma_1 a | J' \Gamma_3 c | \Gamma_2 b \rangle = \sum_{J'} \left[(-1)^{\Gamma_1 - a} V \begin{pmatrix} \Gamma_1 & \Gamma_2 & J'\Gamma_3 \\ (-a) & b & c \end{pmatrix} \langle \Gamma_1 || J' \Gamma_3 || \Gamma_2 \rangle \right] \tag{7}$$

where the V-coefficients are defined in Ref. [7].

In this paper, the general expressions for the matrix elements $\langle W' a' | \mathcal{H}_D^J | W' b' \rangle$ and $\langle U' a' | \mathcal{H}_D^J | U' b' \rangle$, where

$$\mathcal{H}_D^J = a_J D(\alpha, \beta, 0) Y_{J0}(\Theta, \phi), \tag{8}$$

are determined and given in Appendix A. In Appendix A the reduced matrix elements for a specific J-term in Eq. (7) are defined as follows:

$$\begin{aligned} &\langle W' || A || W' \rangle = K_J; \quad && \langle W' || 4U || W' \rangle = E_J \\ &\langle W' || 2V || W' \rangle = A_J: && \langle U' || A || U' \rangle = K'_J \\ &\langle W' || 4V || W' \rangle = B_J; && \langle U' || 2V || U' \rangle = D_J. \end{aligned} \tag{9}$$

The $C_J(\Gamma a)$ coefficients are given in Appendix B.

The reduced matrix elements (9) may be determined by equating the appropriate matrix elements for a specific J-value of (8) for each J' in the $^{2S+1}L_{J'}$ level for the $^{2S+1}L_{J'}$ term of a general f^n configuration. These J'-values range from $\frac{5}{2}$ to $\frac{15}{2}$. The results for $J = 2, 4$ and 6 are given in Appendix A(c) and A(d). Knowledge of these coefficients provides a very convenient method for determining the eigenvalues and eigenfunctions for the crystal field distortion along any axis.

Once the effect of the generalised form of the crystal field interaction on the energy level scheme is known, a range of properties may be readily calculated. Let us examine the dependence of the g-tensor on the crystal field environment. The W' level is split into three doublets and the U'-level into two doublets; the energy separation of these doublets depends on the angles α and β and the a_2, a_4 and a_6 values in Eq. (8). To determine the g-values of these doublets, an applied magnetic field interaction, $\boldsymbol{B}$, is considered. When calculating the g-tensor components, it is more appropriate to define

$$\begin{aligned} B_{x'} &= D(\alpha, \beta, 0) B_x = \cos\alpha \cos\beta B_x + \sin\alpha \cos\beta B_y - \sin\beta B_z \\ B_{y'} &= D(\alpha, \beta, 0) B_y = -\sin\alpha B_x + \cos\alpha B_y \\ B_{z'} &= D(\alpha, \beta, 0) B_z = \cos\alpha \sin\beta B_x + \sin\alpha \sin\beta B_y + \cos\beta B_z \end{aligned}$$

where the z'-axis is parallel to the crystal field distortion axis. Since the magnetic field transforms as T_1 the magnetic field interaction may be expressed in terms of the reduced matrix elements, $\langle \Gamma_1 || T_1 || \Gamma_2 \rangle$. Here only the case where $\Gamma_1 = \Gamma_2$ is considered.

Since

$$|J'\Gamma a\rangle = \sum_M C(J'M\Gamma a)|J'M\rangle$$

$$\langle J'\Gamma a|L_z + 2S_z|J'\Gamma a\rangle = g'_J \sum_M |C(J'M\Gamma a)|^2 M$$

therefore, the reduced matrix elements follow from the equation,

$$\sum_{J'} (-1)^{\Gamma - a} V \begin{pmatrix} \Gamma & \Gamma & J'T_1 \\ -(a) & a & 0 \end{pmatrix} \langle \Gamma||J'T_1||\Gamma\rangle = g_{J'} \sum_M |C(J'M\Gamma a)|^2 M. \tag{10}$$

The complete list of the magnetic field interaction matrix elements in terms of the direction cosines l, m and n and the reduced matrix elements are given in Appendix C.

From the knowledge of these reduced matrix elements the g-tensor may readily be determined for any $^{2S+1}L_{J'}$ level where the f^n ion is in a crystal field environment of predominantly icosahedral symmetry, with a distortion component given by Eq. (8) at any specific direction. A few examples are given in the next section.

Further, for the case in which the f^n system is in a crystal field environment of exact icosahedral symmetry, the e.p.r. spectrum expected for W' and U' levels may be derived from the generalised form of the magnetic field interaction within these levels. Using the axis system defined by the crystal field interaction given by the Hamiltonian (2), the energies and the intensities of the transitions can be determined, in general terms, when the applied magnetic field is parallel to the x, y and z directions. For the U'-level, the e.p.r. spectrum is a single isotropic line, where

$$g_{xx} = g_{yy} = g_{zz} = \frac{1}{\sqrt{15}} \langle U'||T_1||U'\rangle. \tag{11}$$

The results for the W'-level are not so simple; the transitions are set out in detail below.

(a) *Applied magnetic field parallel to the x-axis*
Transitions $i \rightarrow j$ ($i, j = 1, 2$ and 3) parallel to the y-axis

$$\text{Energy difference} = |\varepsilon_i + \varepsilon_j| \mu_B B.$$

$$\text{Relative intensity} = \left((\alpha_i\beta_j + \beta_i\alpha_j) \frac{X}{2\sqrt{105}} + (\beta_i\gamma_j + \gamma_i\beta_j) \times \left\{ \frac{2a}{\sqrt{105}} - \frac{b}{2\sqrt{21}} \right\} + \gamma_i\gamma_j \left\{ \frac{\sqrt{3}a}{\sqrt{70}} + \frac{b}{\sqrt{42}} \right\} \right)^2.$$

Transitions $i \rightarrow j$ ($i, j = 1$, 2 and 3) parallel to the z-axis

$$\text{Energy difference} = |\varepsilon_i + \varepsilon_j| \mu_B B.$$

$$\text{Relative intensity} = \left\{ \alpha_i\alpha_j \left\{ -\frac{\sqrt{5}a}{\sqrt{42}} + \frac{b}{5\sqrt{42}} \right\} + \frac{(10a^2 + 2\sqrt{5}ab - 24b^2)}{X^2} \right.$$
$$\left. \times \left(\beta_i\beta_j \left\{ \frac{\sqrt{3}a}{\sqrt{70}} + \frac{b}{\sqrt{42}} \right\} + \gamma_i\gamma_j \left\{ -\frac{a}{\sqrt{210}} + \frac{\sqrt{2}b}{\sqrt{21}} \right\} \right) \right\}^2$$
$$+ \left\{ \alpha_i\alpha_j \left\{ \frac{\sqrt{7}b}{5\sqrt{6}} \right\} + \frac{7(2\sqrt{5}a + b)b}{X^2} \right.$$
$$\left. \times \left(\beta_i\beta_j \left\{ \frac{\sqrt{3}a}{\sqrt{70}} + \frac{b}{\sqrt{42}} \right\} + \gamma_i\gamma_j \left\{ -\frac{a}{\sqrt{210}} + \frac{\sqrt{2}b}{\sqrt{21}} \right\} \right) \right\}^2.$$

(b) *Applied magnetic field parallel to the y-axis*
Transitions $i \to j$ ($i, j = 1$, 2 and 3) parallel to the x-axis

$$\text{Energy difference} = |\varepsilon_i + \varepsilon_j| \mu_B B.$$

$$\text{Relative intensity} = \left\{ (\alpha_i\beta_j + \beta_i\alpha_j) \frac{X}{2\sqrt{105}} + (\beta_i\gamma_j + \gamma_i\beta_j) \left\{ \frac{2a}{\sqrt{105}} - \frac{b}{2\sqrt{21}} \right\} \right.$$
$$\left. + \gamma_i\gamma_j \left\{ \frac{\sqrt{3}a}{\sqrt{70}} + \frac{b}{\sqrt{42}} \right\} \right\}^2.$$

Transitions $i \to j (i, j = 1$, 2 and 3) parallel to the z-axis

$$\text{Energy difference} = |\varepsilon_i - \varepsilon_j| \mu_B B.$$

$$\text{Relative intensity} = \frac{\alpha_i^2\alpha_j^2 7b^2}{150X^4} [60a^2 + 5\sqrt{5}ab - 25b^2]^2.$$

$$\text{Energy difference} = |\varepsilon_i + \varepsilon_j| \mu_B B.$$

$$\text{Relative intensity} = \left\{ \frac{\alpha_i\alpha_j}{5\sqrt{42}X^2} \{-50\sqrt{5}a^3 - 40a^2b + 220\sqrt{5}ab^2 + 25b^3\} \right.$$
$$\left. + \beta_i\beta_j \left\{ \frac{\sqrt{3}a}{\sqrt{70}} + \frac{b}{\sqrt{42}} \right\} + \gamma_i\gamma_j \left\{ -\frac{a}{\sqrt{210}} + \frac{\sqrt{2}b}{\sqrt{21}} \right\} \right\}^2.$$

(c) *Applied magnetic field parallel to the z-axis*
Transitions perpendicular to the z-axis

$$\text{Energy difference} = |E_1 - E_2| \mu_B B.$$

$$\text{Relative intensity} = 2 \left(\rho \left\{ \frac{a}{\sqrt{42}} + \frac{b}{2\sqrt{210}} \right\} + \frac{t\sqrt{7}b}{2\sqrt{30}} \right)^2.$$

$$\text{Energy difference} = |E_1 + E_2| \mu_B B.$$

$$\text{Relative intensity} = 2 \left(\rho \frac{\sqrt{7}b}{2\sqrt{30}} - t \left\{ \frac{a}{\sqrt{42}} + \frac{b}{2\sqrt{210}} \right\} \right)^2.$$

$$\text{Energy difference} = |E_2 - E_3| \mu_B B.$$

$$\text{Relative intensity} = 2\left(-\frac{2a}{\sqrt{105}}+\frac{b}{2\sqrt{21}}\right)^2.$$

$$\text{Energy difference} = |2E_3|\mu_B B.$$

$$\text{Relative intensity} = \left(\frac{\sqrt{3}a}{\sqrt{70}}+\frac{b}{\sqrt{42}}\right)^2.$$

ε_i and ε_j are the eigenvalues with the corresponding eigenfunctions ϕ_i and ϕ_j ($\phi_h = \alpha_h\phi_1 + \beta_h\phi_2 + \gamma_h\phi_3$) of the following matrix:

$$\begin{array}{c|ccc} & \phi_1 & \phi_2 & \phi_3 \\ \phi_1 & 0 & X/2\sqrt{105} & 0 \\ \phi_2 & X/2\sqrt{105} & 0 & -2a/\sqrt{105}+b/2\sqrt{21} \\ \phi_3 & 0 & -2a/\sqrt{105}+b/2\sqrt{21} & \sqrt{3}a/\sqrt{70}+b/\sqrt{42} \end{array}$$

where

$$X = \{10a^2 + 2\sqrt{5}ab + 25b^2\}$$

$$a = \langle W'||T_1||W'\rangle, \qquad b = \langle W'||5T_1||W'\rangle$$

$$E_1 = \left[\left\{-\frac{\sqrt{5}a}{\sqrt{42}}+\frac{b}{5\sqrt{42}}\right\}^2+\frac{7b^2}{150}\right]^{1/2}$$

$$E_2 = -\frac{\sqrt{3}a}{\sqrt{70}}-\frac{b}{\sqrt{42}}$$

$$E_3 = -\frac{a}{\sqrt{210}}+\frac{\sqrt{2}b}{\sqrt{21}}$$

$$\rho^2 = \frac{1}{2}+\left\{-\frac{\sqrt{5}a}{\sqrt{42}}+\frac{b}{5\sqrt{42}}\right\}\Big/2E_1$$

$$t^2 = \frac{1}{2}-\left\{-\frac{\sqrt{5}a}{\sqrt{42}}+\frac{b}{5\sqrt{42}}\right\}\Big/2E_1$$

and $\rho t = \sqrt{7}b/(10\sqrt{6}E_1)$.

Except when the applied field is parallel to the y-axis and the transitions are parallel to the z-axis, the sum of the intensities is proportional to $(a^2+b^2)/6$. In the case of the exception, when half the intensity of the zero transition is added the sum is also proportional to $(a^2+b^2)/6$.

In general the e.p.r. results are complex and not isotropic; specific cases are examined in the next section.

3. Results and discussion

If the crystal field environment has exact icosahedral symmetry, a number of e.p.r. transitions are allowed within the sixfold degenerate W' state. The situation,

which then becomes rather complex, is illustrated by two examples, the $J=\frac{7}{2}$ and $J=\frac{9}{2}$ cases. In Tables 1 and 2, the expected energy and its corresponding relative intensity are given for the case when the applied magnetic field is along the x, y and z axes and the exciting field is perpendicular to these axes.

Tables 1 and 2 illustrate the interesting result that the e.p.r. spectrum is anisotropic and in general up to 14 transitions occur. Hence this sixfold degenerate level is the only level which yields an anisotropic e.p.r. spectrum when the crystal field

Table 1. The allowed e.p.r. transitions and their relative intensities for an f^n ion in a crystal field environment of icosahedral symmetry, with a $J=\frac{7}{2}$ ground state

Applied field	x-axis		y-axis		z-axis
transitions	y-axis	z-axis	z-axis	x-axis	x & y axes
Energy ($g_J\mu_B B$)					
5.2426	0.8217	0.6577	0.6250	0.8217	
4.5000					1.8000
4.2426			0.3815		
3.6213	0.2095	0.5156	0.0686	0.2095	
3.2426	0.9318	1.9496	0.8378	0.9318	
2.6213			2.6050		
2.5000					2.2500
2.0000	4.2388	1.6955	0.1696	4.2388	
1.6213			0.4469		
1.0000	2.2500	1.3235	0.9420	2.2500	4.0000
0.6213	2.0483	4.3582	1.7532	2.0483	
0.5000					2.4500
0.0000			5.3408		

Table 2. The allowed e.p.r. transitions and their relative intensities for an f^n ion in a crystal field environment of icosahedral symmetry, with a $J=\frac{9}{2}$ ground state

Applied field	x-axis		y-axis		z-axis
transitions	y-axis	z-axis	z-axis	x-axis	x & y axes
Energy ($g_J\mu_B B$)					
6.2000					0.4900
5.9391	0.3464	0.6093	0.4097	0.3464	
5.6785			0.5284		
5.4180	0.9282	0.3586	0.0090	0.9282	
3.8000					5.7600
3.4090	5.1634	6.8620	6.3032	5.1634	
3.2000					1.2800
3.1484			0.7394		
2.5301			0.5588		
2.2696	8.4079	5.9145	5.1751	8.4079	
1.8000					8.8200
0.8789	1.1555	1.8276	1.4366	1.1555	
0.2606	0.3487	0.7780	0.2496	0.3487	
0.0000			1.8804		

environment for an f^n-ion is of octahedral or of higher symmetry. Therefore, if the e.p.r. spectrum for an f^n-ion is anisotropic it does not rule out the possibility that the crystal field environment may be of icosahedral or very nearly icosahedral symmetry. We shall explore next the effect on the system by calculating the e.p.r. spectrum when a crystal field distortion of the form given by Eq. (5) is added to the crystal field potential of icosahedral symmetry.

Here we shall neglect, as a first approximation, the crystal field distortion interaction between the various energy levels and confine our analysis to the effect of the distortion on the icosahedral W'-level. The crystal field distortion matrix element of the form given by Eq. (5), where the distortion axis given in Eq. (5) may be at any angle to the icosahedral axis system - the angles α and β of Eq. (6) define this direction - are given in Appendix A(a). The $C_J(\Gamma a)$ coefficients are given in Appendix B for a specific J-value and the angles α and β. The four reduced matrix elements A_J, B_J, E_J, and K_J are given in Appendix A(c). The eigenvalues of the 6×6 crystal field distortion matrix yield three twofold degenerate levels. From the eigenfunctions of the matrix the e.p.r. spectrum may be determined using the magnetic field interaction matrix elements given in Appendix C.

To illustrate the variation of the principal g-values for the three W'-doublets as the direction of the crystal field distortion changes we have chosen $J=\frac{9}{2}$, $\alpha=\pi/2$ and varied β for the $Y_{20}(\Theta,\phi)$ and $Y_{60}(\Theta,\phi)$ component of (5). The angular dependence of the three principal g-values in units of g_J for the three W'-doublets for the $Y_{20}(\Theta,\phi)$ component of (5) are shown in Fig. 1.

In Fig. 1, there is a fivefold axis at $\beta=0°$ and 116.5651°, a threefold axis at $\beta=37.7774°$ and 79.1876°, and a twofold axis at $\beta=58.2825°$ and 148.2825°. It will be seen that in general the three principal g-values are anisotropic for each doublet. Along the fivefold and threefold axes, the g-tensor for each doublet is axially symmetric. This is usual for the distortion Hamiltonian of the form given by Eq. (5) and Fig. 2 illustrates the case for the three W' doublets for the $Y_{60}(\Theta,\phi)$ component of (5).

In addition, the g-values are very dependent on angle and very small changes in β may have a very significant effect on the results. This is particularly noticeable near the threefold axis in Fig. 1. For other J'-values this marked angular dependence near the threefold axis is even more pronounced. It is noted that for a specific doublet there are other possible β angles which yield an axially symmetric g-tensor and, in some cases, the g-tensor is very nearly isotropic. Further information is provided in Tables 3 and 4 which give the major g-values at the principal rotation axes when each of the distortion components is considered separately. In all cases, the z direction is taken to be parallel to the rotation axis of interest. The distortion parameters a_2, a_4, and a_6 were taken as being positive, and sufficiently large to regard the doublets as isolated Kramers doublets. From Tables 3 and 4, and for all other cases, when the distortion is along the fivefold axis the g-values of the three doublets for each $Y_{J0}(\Theta,\phi)$ distortion operator are the same but the relative separation of the doublets are different. (For the $J=\frac{7}{2}$

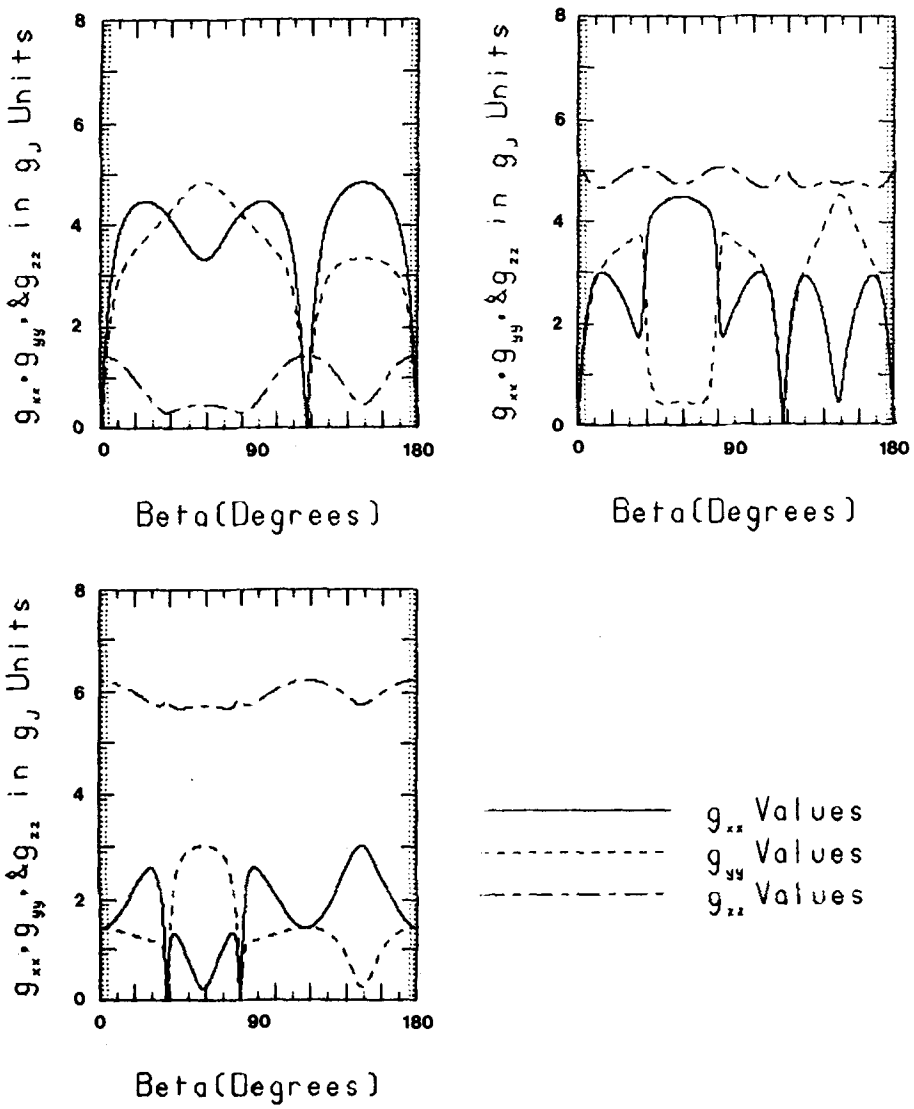

Fig. 1. The variation of the principal g-values when the direction of the $Y_{20}(\Theta, \phi)$ component is changed by the angle β

case - Table 3 - the degeneracy of the W'-level is not lifted completely to yield the three doublets by the $Y_{60}(\Theta, \phi)$ distortion component.) In contrast when the distortion is along the threefold axis only one of the Kramers doublets for the three separate distortion operators has the same principal g-values and when the distortion is along the twofold axis all the Kramers doublets have different principal g-values.

We shall examine next the experimental e.p.r. data for Nd^{3+} in the double nitrate, $Nd_2Mg_3(NO_3)_{12}\cdot 24H_2O$, an f^3 ion with a $^4I_{9/2}$ ground state. In their analysis of

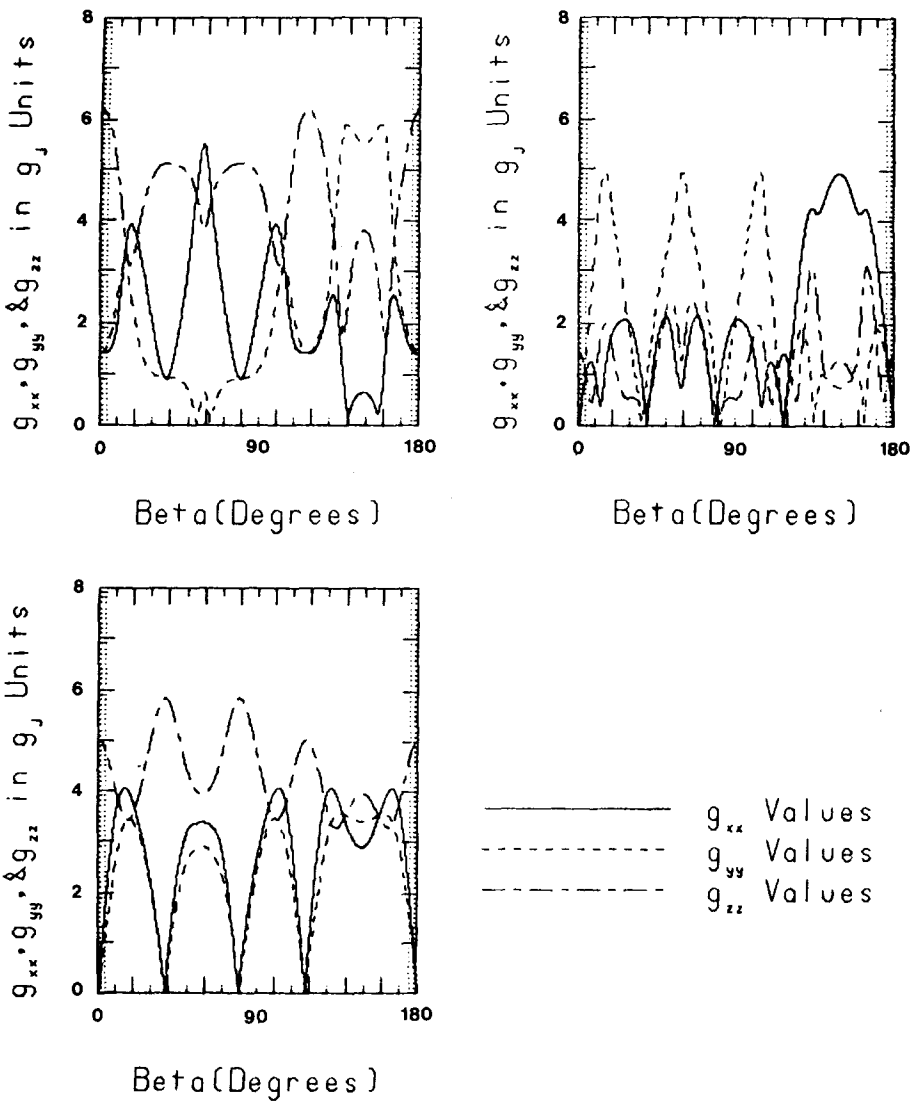

Fig. 2. The variation of the principal g-values when the direction of the $Y_{60}(\Theta, \phi)$ component is changed by the angle β

the experimental data for the neodymium compound, Earney et al. [8] found wavefunctions which satisfactorily explained the e.p.r. and optical data, but not the magnetic susceptibility data. They assumed that the distortion from icosahedral symmetry was parallel to a threefold axis. The g-value results given by Earney et al. [8] are: the ground state Kramers doublet $g_{\|} = 0.39$ and $g_{\perp} = 2.70$; the other doublet 33 cm^{-1} above the ground state $g_{\|} = 3.4$ and $g_{\perp} = 0$.

These results may be interpreted as arising from a distortion given by Eq. (5) from icosahedral symmetry along the threefold axis where the W' level is split

Table 3. The principal g values along the various rotation axes, when the distortion components are considered separately, for $J = \frac{7}{2}$

5-fold axis			3-fold axis			2-fold axis			
Energy	$g_{xx}(g_{yy})$	g_{zz}	Energy	$g_{xx}(g_{yy})$	g_{zz}	Energy	g_{xx}	g_{yy}	g_{zz}
$a_2 Y_{20}(\Theta, \phi)$									
0.138	0	4	0.182	2.265	4.215	0.177	2.570	1.993	4.112
0.035	0	5	−0.078	1.735	1.215	−0.050	1.977	4.962	1.112
−0.173	4	1	−0.104	0.000	3.000	−0.127	4.594	2.954	1.000
$a_4 Y_{40}(\Theta, \phi)$									
0.121	4	1	0.104	1.414	4.630	0.106	2.186	1.479	3.292
0.054	0	4	−0.040	0.000	3.000	−0.005	0.755	2.624	2.453
−0.175	0	5	−0.064	2.586	1.630	−0.101	1.059	5.145	1.745
$a_6 Y_{60}(\Theta, \phi)$									
0.058	0	4	0.154	0	3	0.132	2.553	0.241	2.953
−0.085	—	—	−0.006	2	5	−0.011	0.553	2.241	4.047
−0.085	—	—	−0.085	2	2	−0.085	2.000	2.000	3.000

g-values in units of g_J.
Energy in units of $a_J \langle 7/2 || Y_J || 7/2 \rangle$

Table 4. The principal g values along the various rotation axes, when the distortion components are considered separately, for $J = \frac{9}{2}$

5-fold axis			3-fold axis			2-fold axis			
Energy	$g_{xx}(g_{yy})$	g_{zz}	Energy	$g_{xx}(g_{yy})$	g_{zz}	Energy	g_{xx}	g_{yy}	g_{zz}
$a_2 Y_{20}(\Theta, \phi)$									
0.125	1.4	6.2	0.070	0.0	5.828	0.090	0.211	3.012	5.701
−0.039	0.0	5.0	0.057	2.817	5.077	0.033	4.526	0.442	4.734
−0.086	0.0	1.4	−0.127	4.217	0.277	−0.123	3.337	4.855	0.433
$a_4 Y_{40}(\Theta, \phi)$									
0.112	1.4	6.2	0.069	0.0	5.828	0.110	2.420	4.022	3.955
−0.006	0.0	1.4	0.063	4.524	0.067	0.005	4.895	0.151	1.437
−0.106	0.0	5.0	−0.131	3.124	4.733	−0.115	3.875	2.773	3.991
$a_6 Y_{60}(\Theta, \phi)$									
0.108	0.0	5.0	0.048	0.0	5.828	0.069	3.380	2.890	3.926
0.035	0.0	1.4	−0.019	0.494	0.307	−0.043	0.728	4.929	1.265
−0.001	1.4	6.2	−0.107	0.906	5.107	−0.070	5.508	0.639	3.791

g-values in units of g_J.
Energy in units of $a_J \langle 9/2 || Y_J || 9/2 \rangle$

into three Kramers doublets. With

$$a_2 = 44.8/\langle 9/2||Y_2||9/2\rangle \text{ cm}^{-1}$$

$$a_4 = 398.4/\langle 9/2||Y_4||9/2\rangle \text{ cm}^{-1}$$

$$a_6 = 259.4/\langle 9/2||Y_6||9/2\rangle \text{ cm}^{-1}$$

the following energy level diagram and the corresponding g-values are obtained.

Energy (cm^{-1})	$g_\parallel$	$g_\perp$
−44.98	0.392	2.768
−11.85	3.400	0

Another approach in examining the e.p.r. data is to choose the crystal field interaction Hamiltonian in the form

$$\begin{aligned}\mathcal{H} = {} & a_2^0 Y_{20}(\Theta,\phi) + a_4^0 Y_{40}(\Theta,\phi) + a_4^3[Y_{4-3}(\Theta,\phi) - Y_{43}(\Theta,\phi)] \\ & + a_6^0 Y_{60}(\Theta,\phi) + a_6^3[Y_{6-3}(\Theta,\phi) - Y_{63}(\Theta,\phi)] \\ & + a_6^6[Y_{6-6}(\Theta,\phi) + Y_{66}(\Theta,\phi)]. \end{aligned} \tag{12}$$

From the experimental g data [8], the ground state wavefunction must be of the form

$$l|\tfrac{9}{2} \quad \tfrac{7}{2}\rangle + m|\tfrac{9}{2} \quad \tfrac{1}{2}\rangle + n|\tfrac{9}{2} \quad -\tfrac{5}{2}\rangle$$

and the first excited state of the form

$$\alpha|\tfrac{9}{2} \quad \tfrac{9}{2}\rangle + \beta|\tfrac{9}{2} \quad \tfrac{3}{2}\rangle + \gamma|\tfrac{9}{2} \quad -\tfrac{3}{2}\rangle + \delta|\tfrac{9}{2} \quad -\tfrac{9}{2}\rangle.$$

In addition, the temperature-independent term in the magnetic susceptibility has been given as $\alpha_\parallel = 8.77 \times 10^{-3}$ $\text{cm}^3\,\text{mol}^{-1}$ and $\alpha_\perp = 30.8 \times 10^{-3}$ $\text{cm}^3\,\text{mol}^{-1}$ [8].

By adjusting the parameters in (12), an acceptable solution yields the following results:

	Energy (cm^{-1})	$g_\parallel$	$g_\perp$	$\alpha_\parallel$ (10^{-3} $\text{cm}^3\,\text{mol}^{-1}$)	$\alpha_\perp$ (10^{-3} $\text{cm}^3\,\text{mol}^{-1}$)
(1)	−122.3	0.398	2.752	5.72	20.63
	−89.3	3.261	0	-	-
(2)	−81.5	0.398	2.752	8.57	30.94
	−59.5	3.261	0	-	-

The following crystal field parameters were used for the first set of results: $a_2^0 = 1.8$, $a_4^0 = -1.65$, $a_4^3 = -2.1$, $a_6^0 = -3.75$, $a_6^3 = 12.9$, $a_6^6 = 6.6$, all in units of $\langle \frac{9}{2}||Y_J||\frac{9}{2}\rangle/\sqrt{2145}$. The second set of results were obtained with crystal field parameters $\frac{2}{3}$ of the first set values. The g_J-value is 0.663 compared with $\frac{8}{11}$ (0.727) for $J = \frac{9}{2}$.

It is possible, by using a crystal field of predominantly icosahedral symmetry, to obtain a satisfactory theoretical account of the experimental data [8] although, in the final analysis, these solutions depend very much on the amount of confidence that can be placed on the experimental data.

The group-theoretical method enables a generalised approach in handling calculations for f^n systems in a wide range of crystal field environments. Not only may the results be generalised but the work involved in calculating the required matrix element is simplified and reduced greatly. For example, the effect of a distortion component to the crystal field environment of icosahedral symmetry on the sixfold degenerate W'-level may be determined from only 8 different matrix elements for any W'-level for any f^n-configuration. In this paper we have concentrated on showing that calculations of the e.p.r. spectra for f^n ions in crystal field environments of icosahedral symmetry or with a distortion component at a specific direction to the icosahedral axis system may readily be carried out from the generalised matrix elements. The results give a much greater wealth of information than may be determined from a single calculation for a specific f^n system in a pre-determined crystal field environment.

Appendix A. All the required matrix elements for the Hamiltonian of Eq. (8)

(a) The matrix elements for the W' irreducible representation.

$$\langle W'\tau'|\mathcal{H}_D^J|W'\tau'\rangle = \langle W'\omega'|\mathcal{H}_D^J|W'\omega'\rangle = C_J(Aa)\frac{K_J}{\sqrt{6}} + \frac{C_J(V0)}{2\sqrt{105}}[5A_J + \sqrt{3}B_J]$$

$$\langle W'\tau'|\mathcal{H}_D^J|W'\nu'\rangle = \langle W'\psi'|\mathcal{H}_D^J|W'\omega'\rangle = \frac{C_J(V1)}{\sqrt{14}}\left[A_J - \frac{4B_J}{5\sqrt{3}}\right] + C_J(U\lambda)\frac{iE_J}{2\sqrt{15}}$$

$$\langle W'\tau'|\mathcal{H}_D^J|W'\phi'\rangle = \langle W'\chi'|\mathcal{H}_D^J|W'\omega'\rangle = \frac{C_J(V2)}{\sqrt{7}}\left[\frac{A_J}{2} + \frac{\sqrt{3}}{10}B_J\right] + C_J^*(U\kappa)\frac{i\sqrt{3}}{2\sqrt{10}}E_J$$

$$\langle W'\tau'|\mathcal{H}_D^J|W'\chi'\rangle = -\langle W'\phi'|\mathcal{H}_D^J|W'\omega'\rangle = -C_J^*(V2)\frac{i\sqrt{7}}{5\sqrt{3}}B_J + C_J(U\kappa)\frac{E_J}{2\sqrt{30}}$$

$$\langle W'\tau'|\mathcal{H}_D^J|W'\psi'\rangle = -\langle W'\nu'|\mathcal{H}_D^J|W'\omega'\rangle = C_J^*(V1)\frac{i\sqrt{7}}{5\sqrt{6}}B_J + C_J^*(U\lambda)\frac{E_J}{\sqrt{15}}$$

$$\langle W'\nu'|\mathcal{H}_D^J|W'\nu'\rangle = \langle W'\psi'|\mathcal{H}_D^J|W'\psi'\rangle = C_J(Aa)\frac{K_J}{\sqrt{6}} - \frac{C_J(V0)}{2\sqrt{105}}[A_J + 3\sqrt{3}B_J]$$

$$\langle W'\nu'|\mathcal{H}_D^J|W'\phi'\rangle = \langle W'\chi'|\mathcal{H}_D^J|W'\psi'\rangle = \frac{C_J(V1)}{\sqrt{105}}[\sqrt{3}A_J + 2B_J] - C_J(U\lambda)\frac{iE_J}{2\sqrt{6}}$$

$$\langle W'\nu'|\mathscr{H}_D^J|W'\chi'\rangle = -\langle W'\phi'|\mathscr{H}_D^J|W'\psi'\rangle$$
$$= \frac{C_J(V2)i}{2\sqrt{105}}[3\sqrt{3}A_J - B_J] + C_J^*(U\kappa)\frac{E_J}{2\sqrt{6}}.$$

$$\langle W'\phi'|\mathscr{H}_D^J|W'\phi'\rangle = \langle W'\chi'|\mathscr{H}_D^J|W'\chi'\rangle = C_J(A_a)\frac{K_J}{\sqrt{6}} + \frac{C_J}{\sqrt{105}}[\sqrt{3}B_J - 2A_J]$$

(b) The matrix elements for the U' irreducible representation.

$$\langle U'\kappa'|\mathscr{H}_D^J|U'\kappa'\rangle = \langle U'\nu'|\mathscr{H}_D^J|U'\nu'\rangle$$
$$= \frac{C_J(Aa)K_J'}{2} + \frac{C_J(V0)D_J}{2\sqrt{5}}$$

$$\langle U'\kappa'|\mathscr{H}_D^J|U'\lambda'\rangle = \langle U'\mu'|\mathscr{H}^J|U'\nu'\rangle = \frac{C_J(V1)D_J}{\sqrt{10}}$$

$$\langle U'\kappa'|\mathscr{H}_D^J|U'\mu'\rangle = -\langle U'\lambda'|\mathscr{H}_D^J|U'\nu'\rangle = \frac{C_J(V2)iD_J}{\sqrt{10}}$$

$$\langle U'\lambda'|\mathscr{H}_D^J|U'\lambda'\rangle = \langle U'\mu'|\mathscr{H}_D^J|U'\mu'\rangle$$
$$= \frac{C_J(Aa)K_J'}{2} - \frac{C_J(V0)D_J}{2\sqrt{5}}$$

(c) The reduced matrix elements, $\langle J'W'||\Gamma||J'W'\rangle$, for $J' = \frac{5}{2}$ *to* $\frac{15}{2}$

RME	$\frac{5}{2}$	$\frac{7}{2}$	$\frac{9}{2}$	$\frac{11}{2}$
A_2	1	$\frac{1}{2}(2)^{1/2}$	$\frac{-13}{5(77)^{1/2}}$	$\frac{-31}{(1001)^{1/2}}$
A_4	0	$\frac{-5}{2}\left(\frac{5}{66}\right)^{1/2}$	$-2\left(\frac{13}{231}\right)^{1/2}$	$\frac{-9}{7}\left(\frac{15}{286}\right)^{1/2}$
A_6	0	$\frac{1}{2}\left(\frac{5}{143}\right)^{1/2}$	$\frac{-14}{5}\left(\frac{2}{243}\right)^{1/2}$	$-7\left(\frac{5}{2431}\right)^{1/2}$
B_2	0	$\frac{-1}{2}\left(\frac{3}{2}\right)^{1/2}$	$2\left(\frac{3}{77}\right)^{1/2}$	$\frac{12}{7}\left(\frac{3}{143}\right)^{1/2}$
B_4	$\frac{(5)^{1/2}}{3}$	$\frac{-1}{6}\left(\frac{5}{22}\right)^{1/2}$	$\frac{1}{3}\left(\frac{11}{91}\right)^{1/2}$	$\frac{44}{21}\left(\frac{5}{286}\right)^{1/2}$
B_6	0	$\frac{3}{2}\left(\frac{15}{143}\right)^{1/2}$	0	0
E_4	$-\frac{2}{3}$	$\frac{-1}{3}\left(\frac{2}{11}\right)^{1/2}$	$\frac{14}{3}\left(\frac{7}{715}\right)^{1/2}$	$\frac{-10}{3(286)^{1/2}}$
E_6	0	$\left(\frac{42}{143}\right)^{1/2}$	$2\left(\frac{21}{715}\right)^{1/2}$	$-3\left(\frac{42}{2431}\right)^{1/2}$
K_6	0	$\frac{-1}{2(13)^{1/2}}$	$\left(\frac{2}{65}\right)^{1/2}$	$\frac{5}{2(221)^{1/2}}$

RME	$\frac{13}{2}$	$\frac{15}{2}$
A_2	$\frac{-43}{70}\left(\frac{3}{26}\right)^{1/2}$	$\frac{1}{140(17)^{1/2}}\left\{107 \pm \frac{341}{2}\left(\frac{11}{71}\right)^{1/2}\right\}$
A_4	$\frac{3}{14}\left(\frac{15}{4862}\right)^{1/2}$	$\frac{1}{56}\left(\frac{5}{12\,597}\right)^{1/2}\left\{858 \pm 117\left(\frac{11}{71}\right)^{1/2}\right\}$
A_6	$\frac{-713}{5}\left(\frac{3}{646\,646}\right)^{1/2}$	$\frac{-1}{20(8398)^{1/2}}\left\{308 \mp 323\left(\frac{11}{71}\right)^{1/2}\right\}$
B_2	$\frac{-27}{14(26)^{1/2}}$	$\frac{1}{28}\left(\frac{3}{17}\right)^{1/2}\left\{11 \pm \frac{51}{2}\left(\frac{11}{71}\right)^{1/2}\right\}$
B_4	$\frac{-227}{42}\left(\frac{5}{4862}\right)^{1/2}$	$-\frac{1}{168}\left(\frac{5}{4199}\right)^{1/2}\left\{146 \mp 1165\left(\frac{11}{71}\right)^{1/2}\right\}$
B_6	$\frac{195}{(646\,646)^{1/2}}$	$\pm\frac{5}{4}\left(\frac{429}{45\,866}\right)^{1/2}$
E_4	$\frac{-46}{3(4862)^{1/2}}$	$\frac{1}{6(4199)^{1/2}}\left\{67 \mp 170\left(\frac{11}{71}\right)^{1/2}\right\}$
E_6	$\frac{-24}{(230\,945)^{1/2}}$	$\frac{1}{2}\left(\frac{21}{20\,995}\right)^{1/2}\left\{11 \pm 14\left(\frac{11}{71}\right)^{1/2}\right\}$
K_6	$\frac{37}{2}\left(\frac{6}{146\,965}\right)^{1/2}$	$\frac{1}{2(461\,890)^{1/2}}\left\{121 \pm 284\left(\frac{11}{71}\right)^{1/2}\right\}$

The reduced matrix elements (RME) are in units of $\langle J'||Y_J||J'\rangle$
E_2, K_2 and K_4 are all zero

(d) The reduced matrix elements, $\langle J'U'||\Gamma||J'U'\rangle$, for $J' = \frac{9}{2}$ to $\frac{15}{2}$

RME	$\frac{9}{2}$	$\frac{11}{2}$	$\frac{13}{2}$	$\frac{15}{2}$
D_2	$\frac{2}{5}\left(\frac{3}{11}\right)^{1/2}$	$-\frac{17}{(3003)^{1/2}}$	$-\frac{6}{5}\left(\frac{2}{91}\right)^{1/2}$	$\frac{1}{10}\left(\frac{51}{7}\right)^{1/2}$
D_4	$\frac{-6}{(143)^{1/2}}$	$-\left(\frac{10}{1001}\right)^{1/2}$	$\left(\frac{110}{1547}\right)^{1/2}$	$\frac{-11}{2}\left(\frac{5}{29\,393}\right)^{1/2}$
D_6	$\frac{1}{5}\left(\frac{42}{143}\right)^{1/2}$	$-4\left(\frac{35}{7293}\right)^{1/2}$	$\frac{-187}{5}\left(\frac{2}{46\,189}\right)^{1/2}$	$\frac{11}{5}\left(\frac{21}{8398}\right)^{1/2}$
K_6'	$-\left(\frac{3}{65}\right)^{1/2}$	$-\left(\frac{2}{663}\right)^{1/2}$	$\frac{-33}{(146\,965)^{1/2}}$	$\frac{-11}{2}\left(\frac{33}{20\,995}\right)^{1/2}$

The reduced matrix elements, RME, are in units of $\langle J'||Y_J||J'\rangle$
K_2', and K_4' are all zero

Appendix B. The list of $C_J(\Gamma a)$ coefficients for $J = 2$, 4 and 6

$J = 2$

$$C_2(Aa) = C_2(U\kappa) = C_2(U\lambda) = 0$$

$$C_2(V2) = \frac{-\sqrt{3}}{2\sqrt{2}} \sin^2\beta \exp(-i2\alpha)$$

$$C_2(V1) = \frac{i\sqrt{3}}{\sqrt{2}} \sin\beta \cos\beta \exp(-i\alpha)$$

$$C_2(V0) = (3\cos^2\beta - 1)/2$$

$J = 4$

$$C_4(Aa) = 0$$

$$C_4(U\kappa) = \frac{-\sqrt{7}}{4\sqrt{3}} \sin^2\beta \{\sin\beta \cos\beta \exp(-i3\alpha) - i(7\cos^2\beta - 1)\exp(i2\alpha)\}$$

$$C_4(U\lambda) = \frac{-\sqrt{7}}{4\sqrt{3}} \sin\beta \{i\sin^3\beta \exp(i4\alpha) - \cos\beta(3 - 7\cos^2\beta)\exp(-i\alpha)\}$$

$$C_4(V2) = \frac{-1}{4\sqrt{6}} \sin^2\beta \{14i \sin\beta \cos\beta \exp(i3\alpha) + (7\cos^2\beta - 1)\exp(-i2\alpha)\}$$

$$C_4(V1) = \frac{-1}{8\sqrt{6}} \sin\beta \{7\sin^3\beta \exp(i4\alpha) - 8i\cos\beta(3 - 7\cos^2\beta)\exp(-i\alpha)\}$$

$$C_4(V0) = (35\cos^4\beta - 30\cos^2\beta + 3)/8$$

$J = 6$

$$C_6(Aa) = \frac{-\sqrt{11}}{80} \{42\sin^5\beta \cos\beta \sin 5\alpha - 231\cos^6\beta + 315\cos^4\beta - 105\cos^2\beta + 5\}$$

$$C_6(U\kappa) = \frac{\sqrt{21}}{32} \sin^2\beta \{4\sin\beta \cos\beta(3 - 11\cos^2\beta)\exp(-i3\alpha)$$
$$+ i(33\cos^4\beta - 18\cos^2\beta + 1)\exp(i2\alpha)\}$$

$$C_6(U\lambda) = \frac{\sqrt{21}}{160} \sin\beta \{i11\sin^5\beta \exp(-i6\alpha) + i6\sin^3\beta(11\cos^2\beta - 1)\exp(i4\alpha)$$
$$+ 8\cos\beta(33\cos^4\beta - 30\cos^2\beta + 5)\exp(-i\alpha)\}$$

$$C_6(V2) = \frac{\sqrt{21}}{16} \sin^2\beta \{i\sin\beta \cos\beta(3 - 11\cos^2\beta)\exp(i3\alpha)$$
$$- (33\cos^4\beta - 18\cos^2\beta + 1)\exp(-i2\alpha)\}$$

$$C_6(V1) = \frac{\sqrt{21}}{160} \sin\beta \{-11\sin^5\beta \exp(-i6\alpha) + 9\sin^3\beta(11\cos^2\beta - 1)\exp(i4\alpha)$$
$$- i2\cos\beta(33\cos^4\beta - 30\cos^2\beta + 5)\exp(-i\alpha)\}$$

$$C_6(V0) = \frac{-\sqrt{14}}{80} \{33\sin^5\beta \cos\beta \sin 5\alpha + 231\cos^6\beta - 315\cos^4\beta + 105\cos^2\beta - 5\}$$

Appendix C. The non-zero magnetic field interaction matrix elements in terms of the direction cosines 1, *m*, and *n* and the reduced matrix elements

(a) The matrix elements for the W' irreducible representation.

$$a = \langle W' || T_1 || W' \rangle;\ b = \langle W' || 5T_1 || W' \rangle$$

$$\langle W'\tau' | B' | W'\tau' \rangle = -\langle W'\omega' | B' | W'\omega' \rangle = n/\sqrt{42}\{-\sqrt{5}a + b/5\}$$

$$\langle W'\tau' | B' | W'\nu' \rangle = -\langle W'\psi' | B' | W'\omega' \rangle = -(i1 + m)/\sqrt{42}\{a + b/2\sqrt{5}\}$$

$\langle W'\tau'|B'|W'\psi'\rangle = \langle W'\nu'|B'|W'\omega'\rangle = (1+im)\sqrt{7}b/2\sqrt{30}$

$\langle W'\tau'|B'|W'\omega'\rangle = -in\sqrt{7}b/5\sqrt{6}$

$\langle W'\nu'|B'|W'\nu\rangle = -\langle W'\psi'|B'|W'\psi'\rangle = -n\{\sqrt{3}a/\sqrt{70}+b/\sqrt{42}\}$

$\langle W'\nu'|B'|W'\phi'\rangle = -\langle W'\chi'|B'|W'\psi'\rangle = (i1+m)\{-2a/\sqrt{105}+b/2\sqrt{21}\}$

$\langle W'\phi'|B'|W'\phi'\rangle = -\langle W'\chi'|B'|W'\chi\rangle = n\{-a/\sqrt{210}+\sqrt{2}b/\sqrt{21}\}$

$\langle W'\phi'|B'|W'\chi'\rangle = (1-im)\{\sqrt{3}a/\sqrt{70}+b/\sqrt{42}\}.$

(b) The matrix elements for the U' irreducible representation

$c = \langle U'||T||U'\rangle$

$\langle U'\kappa'|B'|U'\kappa'\rangle = -\langle U'\nu'|B'|U'\nu'\rangle = -\sqrt{3}cn/2\sqrt{5}$

$\langle U'\kappa'|B'|U'\lambda'\rangle = \langle U'\nu'|B'|U'\mu'\rangle = -c(i1+m)/2\sqrt{5}$

$\langle U'\lambda'|B'|U'\lambda'\rangle = -\langle U'\mu'|B'|U'\mu'\rangle = -cn/2\sqrt{15}$

$\langle U'\lambda'|B'|U'\mu'\rangle = c(l-im)/\sqrt{15}.$

(c) The reduced matrix elements for the magnetic field interaction, given in units of g_J.

J'-value	$\langle W'\|\|T_1\|\|W'\rangle$	$\langle W'\|\|5T_1\|\|W\rangle$	$\langle U'\|\|T_1\|\|U'\rangle$
$\frac{5}{2}$	$-\sqrt{105}/\sqrt{2}$	0	—
$\frac{7}{2}$	$\sqrt{105}/\sqrt{2}$	$\sqrt{21}/\sqrt{2}$	—
$\frac{9}{2}$	$17\sqrt{3}/\sqrt{70}$	$-10\sqrt{6}/\sqrt{7}$	$9\sqrt{3}/\sqrt{5}$
$\frac{11}{2}$	$-\sqrt{15}/\sqrt{14}$	$12\sqrt{6}/\sqrt{7}$	$\sqrt{5}/\sqrt{3}$
$\frac{13}{2}$	$-31\sqrt{3}/\sqrt{70}$	$-15\sqrt{3}/\sqrt{14}$	$-7\sqrt{3}/\sqrt{5}$
$\frac{15}{2}$	$\sqrt{3}\{14\pm 341\sqrt{11}/\sqrt{71}\}/2\sqrt{70}$	$\mp 45\sqrt{33}/2\sqrt{994}$	$-17\sqrt{3}/\sqrt{5}$

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