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Jahn-Teller Effect of an Orbital Triplet Coupled to Both E_g and T_{2g} Modes of Vibrations: Symmetry of the Vibronic States

R. Romestain
Laboratoire de Spectroscopie Hertzienne de l' ENS, Faculté des Sciences, Paris, France

and

Y. Merle d'Aubigné
Laboratoire de Spectrométrie Physique, Faculté des Sciences, Grenoble, France

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When an orbital triplet in cubic symmetry is equally coupled to both E_g and T_{2g} modes, the Hamiltonian describing the Jahn-Teller effect has a symmetry much higher than cubic. This fact allows to reduce to a great extent the number of vibrational states coupled to an electronic state. The general expression for the fractional parentage coefficients involved in the calculation of the matrix elements of the interaction is given. A general relation between Ham factors is deduced from symmetry considerations. This model seems to apply very well to the excited level of the F^+ center in CaO.

I. INTRODUCTION

In a recent paper, O'Brien[1] showed that a very special kind of dynamical Jahn-Teller effect might occur when a T_1 orbital level in cubic symmetry is coupled to both E and T_2 modes of vibration. As pointed out by Ham,[2] a quenching of physical quantities such as the orbital angular momentum occurs in the ground vibronic state as a consequence of the Jahn-Teller interaction: This quenching is described by reduction factors or Ham factors K which in many problems depend only on the symmetry of the associated operator. If modes of E symmetry are the only coupled ones, then it is found that parameters of E symmetry are not quenched at all; i.e., $K(E)=1$ but $K(T_2)$ and $K(T_1)$ both decrease to zero as the strength of the coupling is increased. On the other hand, a coupling to T_2 modes leads to a quenching of T_1 and E parameters, but $K(T_2)$ is left larger than $\frac{2}{3}$. One would have guessed that equal coupling to both E and T_2 modes would have reduced the three kinds of parameters. Instead, O'Brien[1] found, using the adiabatic approximation, that this is not the case: Though the Ham factor $K(T_1)$ is zero, as expected in this approximation, $K(E)$ and $K(T_2)$ have the large value of $\frac{2}{5}$. At about the same time experimental work showed that this model might very well apply to the case of the CaO:F$^+$-center problem. The zero-phonon line did not show any spin-orbit splitting, though Kemp *et al.*[3] and Bessent *et al.*[4] measured values of λ of the order of 40 cm^{-1} in the broad band by the Faraday rotation technique. This indicates that $K(T_1)$ is smaller than 0.05. On the other hand, the stress effect on the zero-phonon line[5] showed that the coupling coefficients to strains of E and T_2 symmetry were not much reduced.

This led Hughes[6] to apply the O'Brien model to this system. He showed that strong coupling to E and T_2 modes could explain the qualitative features of the absorption band shape and the relative intensity of the no-phonon line. Merle d' Aubigné and Roussel,[7] through a moment analysis of the magnetic circular dichroism, measured $K(T_1)=0.02$ and showed that indeed the contribution of noncubic (E and T_2) modes to the width of the band is much stronger than that of the cubic (A_{1g}) modes. More recently measurements of the linear dichroism led Duran *et al.*[8] to the values of $K(T_2)\simeq K(E)\simeq 0.4$.

However, as pointed out by Hughes[6] and measured by Merle d' Aubigne and Roussel,[7] the value of E_{JT} was not large enough, as compared to the energy $\hbar\omega$ of the vibrational modes, to make the adiabatic approximation valid. Only a purely quantum-mechanical solution of this problem could give such factors as $K(T_1)$, which would be zero otherwise, or the zero-phonon line intensity, which requires the knowledge of the exact vibronic wave function.

We shall present here a calculation which uses the fact that the complete Hamiltonian may be written in such a way that it behaves as though it had a symmetry much higher than cubic, as soon as E and T_2 modes are considered to have equal energy and coupling strength to a T_1 electronic state. We shall show that, without going into the numerical calculations, a very simple relation is readily found between the two factors $K(T_1)$ and $K(E)=K(T_2)$. This relation is to be compared to the one[9] which links $q=K(E)$ and $p=K(A_2)$ in the case of E-mode coupling to E electronic state. In order to determine the vibronic eigenfunctions and eigenvalues, one would have to diagonalize the Hamiltonian by numerical techniques. The formalism we describe allows us to reduce to a large extent the order of the matrix to be diagonalized. We present in the Appendix the calculation of the fractional parentage coefficients which are necessary to determine the matrix elements.

II. FORM AND SYMMETRIES OF HAMILTONIAN

When treating the coupling of an electronic state to its neighborhood, one has to make many simplifications in order to reduce the number of degrees of freedom of the problem, which would otherwise be untractable. Very often one can limit oneself to the even modes of vibration of the surrounding octahedra which span the $A_1 + E + T_2$ irreducible representations (IR) of the cubic symmetry group. If more modes are involved, one tries to find the linear combination of the same symmetry modes which carry most of the coupling strength: These are known as the interaction modes.[10] We shall thus make the assumption that we have been able to analyze the problem in terms of two degenerate modes of E symmetry and three of T_2 symmetry (the A_1 mode, having only diagonal elements, is very simple to handle) and, in addition, that these have the same frequency and coupling strength.

The Hamiltonian representing the vibration of the five harmonic oscillators can be written as

$$\mathcal{H}_0 = \sum_i \hbar\omega (a_i^\dagger a_i + \tfrac{1}{2}) ,$$

where i stands for $E\theta$, $E\epsilon$, $T_2\xi$, $T_2\eta$, $T_2\zeta$, or any linear combination of them, and $a^\dagger$ and a are the phonon creation and annihilation operators.

The interaction Hamiltonian is

$$\mathcal{H}_1 = \sum_i K\, \mathcal{O}_i (a_i^\dagger + a_i) .$$

K is related to the parameters V_E, V_T, and μ defined in Abragam and Bleaney[11] by

$$K = V_E \left(\frac{5}{4} \frac{\hbar}{\mu\omega} \right)^{1/2} = V_T \left(\frac{5}{3} \frac{\hbar}{\mu\omega} \right)^{1/2} .$$

The $\mathcal{O}_i$'s are electronic operators having the same symmetry as i. The matrix elements of these five operators are the ones which can be found in calculating the matrix elements of the five real components of a second-rank irreducible tensor in a triplet P state as defined in the full rotation group R_3. Indeed, it is well known that a set of operators spanning $\mathcal{D}_2$ is split in $E + T_2$ by a cubic field and the P state is left unchanged. If the K and $\hbar\omega$ parameters are the same for E and T_2, the restriction of the symmetry does not appear in the mathematical form of the Hamiltonian. Instead of considering a real basis for the phonon states, it is worth using some of their linear combinations so that the five $\mathcal{O}_i$'s can be written as the five standard components of a second-rank irreducible tensor. Each phonon can be thought of as having an orbital angular momentum equal to 2, the projection of which is equal to i; the $a_i^\dagger$ appearing in $\mathcal{H}_0$ are then the creation operators for such phonons.

The full Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$ has been transformed in such a way that it seems to exhibit the symmetry of R_3, though the physical problem is certainly not invariant under the elements of R_3. However, it would be if we were considering the coupling of a P state with an isotropic continuum of matter surrounding the defect. Then the deformation would be represented by a nine-component tensor,

$$\epsilon_{ij} = \frac{\partial u_i}{\partial x_j} ,$$

of which the even part spans the $\mathcal{D}_0 + \mathcal{D}_2$ IR of the R_3 group. The vibration and coupling of five modes of $\mathcal{D}_2$ would be represented by exactly the same Hamiltonian we have written.

III. CLASSIFICATION OF EIGENFUNCTION OF VIBRATIONAL HAMILTONIAN $\mathcal{H}_0$

We have seen that the vibronic Hamiltonian $\mathcal{H}$ has a symmetry higher than cubic; here we shall show the vibrational part has a still higher symmetry. It is indeed a simple matter to demonstrate that $\mathcal{H}_0$ is invariant upon the application of the 25 operators $a_i^\dagger a_j$ (their effect is just to move one phonon from state j to state i, thus leaving energy unaffected). These can be thought of as infinitesimal operators of the continuous U_5 group of which R_3 is a subgroup.[12] The eigenfunctions can be classified according to the IR of this group: The states which correspond to the excitation of n phonons span the $[n, 0, 0, 0, 0]$ IR which have

$$C_{n+4}^4 = \frac{(n+1)(n+2)(n+3)(n+4)}{1 \times 2 \times 3 \times 4} \text{ dimensions.}$$

(We should label IR by their highest weight as usual, but shall sometimes drop the zeros as all IR of the problem have this same form.) In just the same way the three-dimensional harmonic oscillator is invariant under U_3, which explains the additional degeneracies one observes in that case.

In order to classify the states, it is worthwhile to consider the chain of reduction $U_5 \rightarrow R_5 \rightarrow R_3$. In reducing U_5 to R_5 one chooses as infinitesimal operators

$$W_{\mu\nu} = a_\mu^\dagger a^\nu (-)^\nu - a_{-\nu}^\dagger a^{-\mu} (-)^\mu , \quad \mu = 1\text{–}5, \quad \nu = 1\text{–}5 .$$

Among these 25 only 10 are linearly independent. The $[n, 0, 0, 0, 0]$ can be shown[13] to split according to

$$[n, 0, 0, 0, 0] \rightarrow [n, 0] + [n-2, 0] + \cdots + [1, 0] \text{ or } [0, 0] ,$$

where the $[\alpha, \beta]$ IR of R_5 are labeled by their highest weight as usual.

Again this reduction is comparable to the $U_3 \rightarrow R_3$ reduction in the three-dimensional oscillator in which

$$[n, 0, 0] \to \mathfrak{D}_n + \mathfrak{D}_{n-2} + \cdots + \mathfrak{D}_1 \text{ or } \mathfrak{D}_0 .$$

The remaining reduction consists of restricting oneself to the three operators of R_3,

$$L_{+1} = (\sqrt{\tfrac{2}{10}})W_{21} - (\sqrt{\tfrac{3}{10}})W_{10} ,$$

$$L_0 = (\sqrt{\tfrac{4}{10}})W_{22} - (\sqrt{\tfrac{1}{10}})W_{11} ,$$

$$L_{-1} = (\sqrt{\tfrac{2}{10}})W_{-2-1} - (\sqrt{\tfrac{3}{10}})W_{-10} .$$

We get the splitting

$$[n, 0] \to \textstyle\sum_L \mathfrak{D}_L ,$$

where the set of the L cannot be expressed in a simple way but has very interesting properties[14]: (a) $L = 1$ never occurs and (b) $L = 0$ and $L = 2$ occur only separately, the first when n is a multiple of 3, the second in all other cases.

We are now able to label any of the vibrational wave functions by the set of quantum numbers

$$|n, \alpha, L, m\rangle ,$$

where n stands for the U_5 IR, i.e., the number of phonons, α stands for the R_5 IR, and L and m specify the $\mathfrak{D}_L$ IR and its component in R_3. L is the momentum resulting from the coupling of n orbital angular momenta, each being equal to 2. It is the same problem we solve when we calculate the different terms of a d^n electronic configuration, but here we are dealing with spinless bosons instead of fermions.

IV. INTRODUCTION OF COUPLING

We are in a position to calculate the effect of the interaction by searching for the vibronic wave functions which diagonalize $\mathcal{H}_0 + \mathcal{H}_1$. Since $\mathcal{H}$ is R_3 invariant, they span different IR of R_3, but for the physical problem we are involved in, it is enough to consider only the P-like state for the following reason. The ground state of the F center is an S state and optical absorption can only reach the $J = 1$ state; during emission at sufficiently low temperature only the ground vibronic state will be populated and this state is expected to keep the same symmetry as the unperturbed electronic one. The symmetry is now an over-all symmetry including electronic and vibrational coordinates; that is, we have to couple two orbital angular momenta. The electronic one is equal to 1; the other must be chosen so that the resulting momentum is also 1: It has to be either 2, 1, or 0.

As $L = 1$ does not appear, the only vibrational wave function which will enter the problem can be written as

$$|n, \alpha, D, m\rangle , \quad |n, \alpha, S, 0\rangle$$

where S and D stand for $L = 0$ and $L = 2$. To respect the symmetry of the resulting $|P\mu\rangle\rangle$ state, the eigenvectors which we shall obtain have to be of the form

$$|P\mu\rangle\rangle = \sum_{n,\, \alpha = 3\beta} \gamma(n, \alpha)|n, \alpha, S\rangle |P\mu\rangle + \sum_{n,\, \alpha \neq 3\beta;\, \nu} \gamma(n, \alpha)|n, \alpha, D, m\rangle\, |P\nu\rangle\langle 21 m\nu | 1\mu\rangle .$$

The values of the Clebsch-Gordan coefficient $\langle 21m\nu | 1\mu\rangle$ are found in tables.[15]

We use the double ket $|P\mu\rangle\rangle$ notation to keep in mind that it involves both electron and vibrations coordinates.

The values of $\gamma(n, \alpha)$ are to be found numerically by calculating and diagonalizing the matrix of $\mathcal{H}$ in the $|P\mu\rangle\rangle$ base. This involves the calculation of such elements as $\langle n, \alpha, L \| a^\dagger \| n', \alpha', L'\rangle$ which are directly related to the fractional parentage coefficients. Their derivation can be found in the Appendix.

Replacing $\sum \gamma(n, \alpha)|n, \alpha, S\rangle$ by $|S\rangle$ and $\sum \gamma(n, \alpha)|n, \alpha, D, m\rangle$ by $|D, m\rangle$, one can write

$$|P1\rangle\rangle = \{ |S\rangle + (\sqrt{\tfrac{1}{10}})|D0\rangle\} |P1\rangle - (\sqrt{\tfrac{3}{10}})|D1\rangle |P0\rangle + (\sqrt{\tfrac{3}{5}})|D2\rangle |P-1\rangle ,$$

$$|P0\rangle\rangle = (\sqrt{\tfrac{3}{10}})|D-1\rangle |P1\rangle + \{ |S\rangle - (\sqrt{\tfrac{2}{5}})|D0\rangle\} |P0\rangle + (\sqrt{\tfrac{3}{10}})|D+1\rangle |P-1\rangle .$$

Because of the presence of the γ's, $|D\rangle$ and $|S\rangle$ are not normalized states but the $|P\mu\rangle\rangle$ are, so

$$S^2 + D^2 = 1 ,$$

where $S^2 = \langle S | S\rangle$ and $D^2 = \langle Dm | Dm\rangle$.

V. CALCULATION OF REDUCTION FACTORS

The simple form the vibronic wave functions display enables us to calculate very easily the matrix elements of any electronic operator in its irreducible tensor form T_k^q. Thus the Ham factor will appear in a nearly analytical way and depends only on the symmetry of the operator. One finds that the vibronic $\langle\langle P \| T_k \| P\rangle\rangle$ double-barred matrix element is equal to the electronic $\langle P \| T_k \| P\rangle$ multiplied by the factor K_k, which is

$$K_1 = S^2 - \tfrac{1}{2} D^2 ,$$

$$K_2 = S^2 + \tfrac{1}{10} D^2 .$$

[The numerical factors appearing in K_k might have been calculated directly from Eq. (3.37) of Judd.[12] They are just equal to the 6j's

$$\begin{Bmatrix} 1 & k & 1 \\ 1 & L & 1 \end{Bmatrix}$$

multiplied by $3(-)^k$.]

Taking into account that $S^2 + D^2 = 1$, one finds that the following simple relation holds:

$$\tfrac{3}{5} K_1 = + K_2 - \tfrac{2}{5} .$$

K_1 is usually called $K(T_1)$ and K_2 represent $K(E)$ or $K(T_2)$, which in the R_3 symmetry have to be equal.

VI. CONCLUSION

The simple form of the general vibronic wave function has been established without any calculation, by using only the fact that equal coupling to E and T_2 modes of equal energy introduces extra symmetry. It leads to an expression of the Ham reduction factors K in terms of one parameter, and then to a relation between these K independent of the strength of the coupling. In the limit of strong coupling where $K(T_1)$ is expected to be very small, a value of $K(E)=K(T_2)=0.4$ is found, in agreement with adiabatic theory of O'Brien.[1]

The formalism we have presented can be best used in the calculation of the eigenvectors and eigenvalues of the Hamiltonian. In order to set up the matrix elements of $\mathcal{H}$, we calculated all the coefficients of fractional parentage (cfp) necessary to this problem. As we were planning to do the numerical calculations we received a report, prior to publication, of work by O'Brien,[16] in which she gives the numerical results of the diagonalization. She obtained the cfp numerically. We thought it useful, for future developments of the theory, to publish our way of deriving these coefficients and to show how the relation between $K(E)$ and $K(T_1)$ comes out from pure symmetry considerations.

We shall now apply these results to the case of the F^+ centers in CaO. When comparing the calculated and the observed shapes of the absorption band, one has to take a full account of the effect of the cubic modes of vibration. They (i) reduce the intensity of the zero-phonon line by an amount e^{-S_c}, (ii) shift the zero-phonon line towards low energies by $S_c(\hbar\omega_c)$, and (iii) contribute to the second moment of the band by $S_c(\hbar\omega_c)^2$, where the Huang-Rhys factor S_c is defined as the average number of phonons emitted when a photon is absorbed. The contributions of the cubic and noncubic modes of vibration to the second moment of the absorption band were measured by Merle d' Aubigné and Roussel[7] (see their Table II c), so that S_c and S_{nc}, the Huang-Rhys factor for noncubic modes, are known. Using the numerical results of O'Brien,[16] it is then possible to calculate $K(T_1)=0.025$ and the intensity of the zero-phonon line which is given by an effective Huang-Rhys factor $S^*=5.2$. These compare very well to the experimental values[7] of 0.02 and 5.6.

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APPENDIX

The calculation of the matrix of $\mathcal{H}_1$ is straightforward as soon as the matrix elements of the type

$$M=\langle n', \alpha', L', m' | a_\mu^\dagger | n, \alpha, L, m\rangle$$

are found. n, α, L, m label the successive IR in the chain of reduction $U_5 \to R_5 \to R_3 \to R_2$.

We first apply the extension of Wigner-Eckart theorem to U_5, noting that $a_\mu^\dagger$ transforms as the 1, 2, μ component of the $[1,0,0,0,0]$ IR of U_5. M can be written as

$$M=\langle n' \| a^\dagger \| n\rangle \langle n', \alpha', L', m' | 112\mu + n, \alpha, L, m\rangle , \tag{A1}$$

where the second factor is a generalized Clebsch-Gordan coefficient.

The double-barred matrix is easily evaluated by choosing the particular wave function $|n, n, 2n, 2n\rangle$, where all phonons are in the same $i=2$ state, and letting $a_2^\dagger$ act on it. As the Clebsch-Gordan coefficient is obviously 1, we get

$$\langle n' \| a^\dagger \| n\rangle = (n+1)^{1/2}\delta(n', n+1) .$$

The generalized Clebsch-Gordan coefficient can be factorized according to a theorem due to Racah,[17] which we use twice:

$$\begin{aligned}\langle n', \alpha', L', m' & | 112\mu + n, \alpha, L, m\rangle \\ &= \langle n', \alpha' | 11 + n, \alpha\rangle \langle \alpha', L' | 12 + \alpha, L\rangle \\ &\quad \times \langle L', m' | 2\mu + L, m\rangle .\end{aligned} \tag{A2}$$

The last factor is a R_3 Clebsch-Gordan coefficient, which is usually written as

$$\langle L', m' | 2\mu + L, m\rangle = \langle 2, L, L', m' | 2, L, \mu, m\rangle ;$$

the product of the remaining two is a coefficient of fractional parentage,[12]

$$\begin{aligned}\langle n', \alpha', L' [\![n, \alpha, L\rangle &= \langle n', \alpha' | 11 + n, \alpha\rangle \\ &\quad \times \langle \alpha', L' | 12 + \alpha, L\rangle .\end{aligned} \tag{A3}$$

The $\langle n', \alpha' | 11 + n, \alpha\rangle$'s have been calculated by Hecht.[13] He finds that the only nonzero coefficients are

$$\langle n+1, \alpha+1 | 11 + n, \alpha\rangle = \left(\frac{(n+\alpha+5)(\alpha+1)}{(2\alpha+5)(n+1)}\right)^{1/2} , \tag{A4a}$$

$$\langle n+1, \alpha-1 | 11 + n, \alpha\rangle = \left(\frac{(n-\alpha+2)(\alpha+2)}{(2\alpha+1)(n+1)}\right)^{1/2} . \tag{A4b}$$

These selection rules derive from the fact that in R_5, $[n, 0]\times[1, 0]=[n+1, 0]+[n, 1]+[n-1, 0]$. Figure 1 summarizes them by showing the only existing connections between IR.

Recursion Formula for Calculating Coefficients of Fractional Parentage

We are left with the $R_5 \to R_3$ coefficient, which we will better evaluate as a cfp:

$$\langle n, n, L \rrbracket n+1, n+1, L' \rangle = \langle n, L+12 | n+1, L' \rangle \ .$$

We shall use the Redmond formula as given in Judd,[18] slightly modified by the fact that we are dealing with spinless bosons:

$$(n+1) \sum_{\psi'} (\psi' \ \bar{\psi})(\bar{\psi}' \rrbracket \psi')\delta(L, L')$$

$$= (-1)^{\bar{L}+\bar{L}'} n(2\bar{L}+1)^{1/2}(2\bar{L}'+1)^{1/2}$$

$$\times \sum_{\bar{\bar{\psi}}} (\bar{\bar{\psi}} \rrbracket \bar{\psi})(\bar{\psi}' \llbracket \bar{\bar{\psi}}) \begin{Bmatrix} \bar{L} & l & \bar{\bar{L}} \\ \bar{L}' & l & L \end{Bmatrix} + \delta(\bar{\psi}, \bar{\psi}') \quad . \qquad \text{(A5)}$$

$\{\,\}$ is a usual $6j$ coefficient; l is the orbital angular momentum of each phonon, i.e., 2; ψ' is any $(n+1)$-phonon wave function $|n+1, \alpha, L'\rangle$, where $L' = L$; $\bar{\psi}\bar{\psi}'$ are n-phonon wave functions $|n, \beta, L\rangle$ or $|n, \beta, \bar{L}'\rangle$; and $\bar{\bar{\psi}}$ is any $(n-1)$-phonon wave function $|n-1, \gamma, \bar{\bar{L}}\rangle$ connected to both $\bar{\psi}$ and $\bar{\psi}'$. In this formula $\bar{\psi}$ and $\bar{\psi}'$ are arbitrary, as well as L.

Let us choose $\bar{\psi}$ and $\bar{\psi}'$ among the $|n, n, L\rangle$ functions. Then, due to selection rules included in Eq. (A4) or in Fig. 1, one sees that $\bar{\bar{\psi}}$ has to be one of the $|n-1, n-1, \bar{\bar{L}}\rangle$. The possible values of ψ' are either

$$\psi'_1 = |n+1, n+1, L\rangle$$

or

$$\psi'_2 = |n+1, n-1, L\rangle \quad .$$

But the $(\bar{\psi} \rrbracket \psi'_2)$ can be related to a $(\bar{\bar{\psi}} \rrbracket \bar{\psi})$ through the relation of symmetry of the R_5 Clebsch-Gordan coefficient,[17] and use of Eqs. (A3) and (A4b):

$$(n, n, L \rrbracket n+1\, n-1\, L') = \left(\frac{2(n+2)}{(n+1)(2n+1)}\right)^{1/2} \langle n, L+12 | n-1, L' \rangle$$

$$= \left(\frac{2}{n+1} \frac{n+2}{(2n+1)}\right)^{1/2} (-)^{L'-L} \left(\frac{2L+1}{2L'+1} \frac{g(n-1)}{g(n)}\right)^{1/2} \langle n-1, L'+12 | n, L \rangle$$

$$= (-)^{L'-L} \left(\frac{2L+1}{2L'+1} \frac{2}{n+1} \frac{n}{2n+3}\right)^{1/2} (n-1, n-1, L' \rrbracket n, n, L) \ , \qquad \text{(A6)}$$

where $g(n) = \frac{1}{6}(n+2)(n+1)(2n+3)$ is the dimension of the IR$[n, 0]$ of R_5.

One can use (A6) to rewrite Redmond's formula:

$$(n+1) \sum_{\psi'_1 = |n+1, n+1, L'\rangle} (\bar{\psi} \rrbracket \psi'_1)(\psi'_1 \llbracket \bar{\psi}')\delta(L, L') = \delta(\bar{\psi}, \bar{\psi}') + n(-)^{\bar{L}+\bar{L}'}(2\bar{L}+1)^{1/2}(2\bar{L}'+1)^{1/2}$$

$$\times \sum_{\bar{\bar{\psi}} = |n-1, n-1, \bar{\bar{L}}\rangle} (\bar{\psi} \llbracket \bar{\bar{\psi}})(\bar{\bar{\psi}} \rrbracket \bar{\psi}') \left[\begin{Bmatrix} \bar{L} & l & \bar{\bar{L}} \\ \bar{L}' & l & L \end{Bmatrix} - \frac{2\delta(L, \bar{\bar{L}})}{(2n+3)(2L+1)} \right] \quad . \qquad \text{(A7)}$$

The $\sum_{\psi'_1}$ is reduced to a single term wherever L appears only once in the reduction of $[n+1, 0]$ to R_3. If so, the $(\bar{\psi} \rrbracket \psi)$ can be readily calculated as soon as the $(\bar{\bar{\psi}} \rrbracket \bar{\psi})$ are known; in fact, (A7) together with normality relations provides too many conditions, and this overdetermined system will let us calculate explicit values of the cfp in some simple cases.

Explicit Values of Some Coefficients of Fractional Parentage

We will now show that the cfp relative to $L=0$ or $L=2$ can be explicitly calculated. This is mostly due to the fact that many small L terms are missing.

Figure 2 shows the nine kinds of cfp linking a $L=2$ or $L=0$ to the other states. Using formula (A7) gives the nth-order cfp functions α^2, $\alpha\beta$, $\alpha\gamma$, δ^2, λ^2, and μ^2 as functions of the $(n-1)$th-order cfp. ϵ^2 is simply given by the normality condition $\epsilon^2 + \delta^2 = 1$.

α^2 is a simple function of the $(D \rrbracket S)$ cfp, which is obviously 1, and is

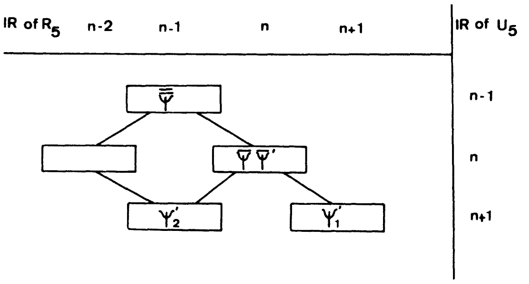

FIG. 1. Functions involved in Redmond's formula. The bar indicates a connection between two Ψ; i.e., the cfp is nonzero.

TABLE I. Values of the coefficients of fractional parentage linking an S or a D term.

$N=3k$	$(NS\rrbracket N+1\,D)=\left(\frac{1}{5}\,\frac{2N+5}{2N+3}\,\frac{N+3}{N+1}\right)^{1/2}=\alpha$
	$(NF\rrbracket N+1\,D)=\left(\frac{1}{2}\,\frac{2N+5}{2N+3}\,\frac{N}{N+1}\right)^{1/2}=\beta$
	$(NG\rrbracket N+1\,D)=\left(\frac{3}{10}\,\frac{2N+1}{2N+3}\,\frac{N}{N+1}\right)^{1/2}=\gamma$
$N=3k+1$	$(ND\rrbracket N+1\,D)=\left(\frac{2}{7}\,\frac{2N+5}{N+1}\right)^{1/2}=\delta$
	$(ND\rrbracket N+1\,G)=\left(\frac{5}{21}\,\frac{2N+5}{2N+3}\,\frac{N+5}{N+1}\right)^{1/2}=\eta$
	$(NG\rrbracket N+1\,D)=\left(\frac{3}{7}\,\frac{N-1}{N+1}\right)^{1/2}=\epsilon$
$N=3k+2$	$(ND\rrbracket N+1\,S)=1$
	$(ND\rrbracket N+1\,F)=\left(\frac{5}{14}\,\frac{N+4}{N+1}\right)^{1/2}=\gamma$
	$(ND\rrbracket N+1\,G)=\left(\frac{1}{6}\,\frac{2N+7}{2N+3}\,\frac{N+2}{N+1}\right)^{1/2}=\mu$

$$\alpha^2=\frac{1}{5}\,\frac{(2N+5)(N+3)}{(N+1)(2N+3)}\quad .$$

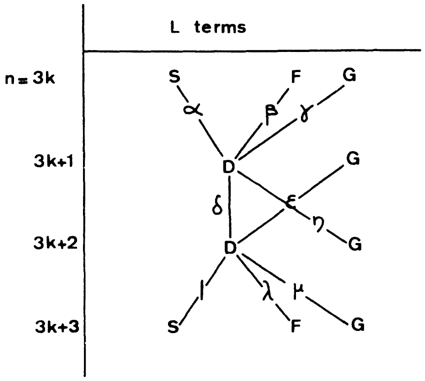

FIG. 2. Schematic definition of the various cfp calculated (see Table I).

The normality of $|3k+1,D\rangle$ imposes the condition $\alpha^2+\beta^2+\gamma^2=1$. Using the recursion formulas, this can be written as a condition on δ. Thence

$$\delta^2=\frac{2}{7}\,\frac{2N+5}{N+1}\quad .$$

For the sake of completeness the nine cfp are given in Table I. The relative phases of the $|n,n,L\rangle$ can be chosen so that they all appear as positive square roots.

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