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u(2) ⊃ su*(2) ⊃ G symmetry adaptation for powers of E irreducible representations of point groups

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Abstract. Several problems in molecular spectroscopy involve two degrees of freedom to which a two dimensional irreducible representation of a point group is associated. We show that a unified treatment of such situations can be given within the $u(2)$ representation theory.

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1 Introduction

Since its early realization by Schwinger [1] in terms of two boson operators, the $u(2)$ algebra has found numerous applications in various areas of physics, ranging from atomic [2], nuclear [3] and molecular [4] physics to quantum optics [5]. In most cases only terms which are of low degree in the generators are considered and the standard $u(2) \supset su(2) \supset so(2)$ chain is used. Applications to molecular systems which admit double degenerate vibrational modes, and more generally two dimensional E type irreducible representations (*irreps*), do not explicitly use the power of the $u(2)$ representation theory; for instance couplings of elementary vibrational operators are performed in the molecular symmetry group [6]. The reason is that one usually introduces the molecular point group G as a subgroup of the $O(3)$ rotation group and there is no *irrep* which subduces to an E type one.

In a previous work [7] we showed that in fact a consistent treatment of doubly degenerate vibrational modes in molecules was possible through $u(2)$. These results are extended to allow more general problems to be considered. In particular some choices made in [7,8] are not convenient for the study of Jahn-Teller systems in doubly degenerate E electronic states. More precisely, such systems fall into two main categories according as the reduction of the symmetrized product $[E \times E]$ contains, besides the scalar *irrep* of G, another E type *irrep* or two B type ones; these are conventionally referred to as $E \otimes \varepsilon$ and $E \otimes (b_1 + b_2)$ cases when a linear coupling is present [9]. In both instances we have thus from the outset an $u_e(2)$ algebra associated with the electronic degrees of freedom. In the $E \otimes \varepsilon$ problems appears naturally an additional $u_v(2)$ algebra associated this time with the vibrational degrees of freedom and the study of vibronic interactions may be planned within $u_e(2) \oplus u_v(2)$. Although the $E \otimes (b_1 + b_2)$ systems do not seem to require the introduction of a $u_v(2)$ vibrational algebra there are some limiting cases where it is so; the obvious one is when both b_1 and b_2 modes have close frequencies [10]. Also we note that for such electronic states $E \otimes \varepsilon$ cases arise but without linear coupling with the vibrational ε mode. It is thus convenient to have a unified scheme for the construction of electronic as well as vibrational operators and the computation of their matrix elements.

In a first part we recall the main results of [7] in the standard $u(2) \supset su(2) \supset so(2)$ chain. Next we show how symmetry adaptation in a point group can be performed for arbitrary states and operators. The notation $u(2) \supset su^*(2) \supset G$ is used to emphasize that we deal with a non canonical symmetry adaptation. We consider first the case where the matrices $D^{(E)}(R)$, R being an element of G, have the real form commonly used in vibrational spectroscopy studies and next the case where some of these matrices are complex. The change of orientation is performed and determine new operators and states symmetrized in the whole chain. In each case our general results are illustrated with simple examples. Possible applications of our formalism are presented in the last section and correlations with previous studies made when possible. In particular we give complete sets of operators and states adapted to the study of orbital doublets of E symmetry type.

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Throughout this paper the notation E is retained for relations or properties which hold whatever the two dimensional *irrep* of G involved; when necessary additional indices are introduced.

2 The standard u(2) ⊃ su(2) ⊃ so(2) chain

We briefly recall the main results of [7,8] for the construction of the space of states and irreducible tensor operators.

We start from the well-known Schwinger realization [2,4] of $su(2)$ in terms of two boson operators:

$$J_+ = b_1^+ b_2, \quad J_- = b_2^+ b_1$$
$$J_z = \frac{1}{2}(N_1 - N_2) = \frac{1}{2}(b_1^+ b_1 - b_2^+ b_2). \tag{1}$$

These generators satisfy the angular momentum commutation relations:

$$[J_z, J_\pm] = \pm J_\pm, \qquad [J_+, J_-] = 2J_z. \tag{2}$$

$u(2)$ is obtained with the addition of the linear invariant $N = N_1 + N_2$. A basis for the space of states is given by:

$$|n_1, n_2\rangle = (n_1! n_2!)^{-1/2}\ b_1^{+n_1}\ b_2^{+n_2}|0,0\rangle \tag{3}$$

where $|0,0\rangle$ is the vacuum state.

2.1 The covariant 2d states and elementary operators

Keeping with previous conventions [11,12] covariant $su(2)$ states $|jm\rangle\rangle$ and operators $T_m^{(j)}$ are characterized by the relations $(m : -j, ..., j)$:

$$J_z\ |jm\rangle\rangle = -m\ |jm\rangle\rangle$$
$$J_\pm\ |jm\rangle\rangle = -[(j \pm m)(j \mp m + 1)]^{1/2}\ |jm \mp 1\rangle\rangle \tag{4}$$

and

$$[J_z,\ T_m^{(j)}] = -m\ T_m^{(j)}$$
$$[J_\pm,\ T_m^{(j)}] = -[(j \pm m)(j \mp m + 1)]^{1/2}\ T_{m\mp 1}^{(j)}. \tag{5}$$

Contravariant sets $((jm|$ and $T_{(j)}^m$, which satisfy the more usual Racah relations [13,14], may be obtained through the metric tensor[1]:

$$((jm| = \begin{pmatrix} m' & m \\ & j \end{pmatrix}\ |jm'\rangle\rangle = (-1)^{j+m}\ \delta_{m',-m}\ |jm'\rangle\rangle \tag{6}$$

$$T_{(j)}^m = \begin{pmatrix} m' & m \\ & j \end{pmatrix}\ T_{m'}^{(j)} = (-1)^{j+m}\ \delta_{m',-m}\ T_{m'}^{(j)}. \tag{7}$$

In the following we shall deal with covariant sets only.

States and tensor operators symmetrized in the whole $u(2) \supset su(2) \supset so(2)$ chain are further characterized by an additional $u(2)$ label $[m_{12}\, m_{22}] = [m_1\ -m_2]$ in Gel'fand notation [15–17] with $j = (m_1 + m_2)/2$ and

$$[N_i, {}^{[m_1\ -m_2]}T_{-j}^{(j)}] = m_{i2}{}^{[m_1\ -m_2]}T_{-j}^{(j)} \quad i = 1,2. \tag{8}$$

With equations (1) and (4, 8) it is easily checked that the states (3) determine a covariant standard $2d$ basis

$$|[n\,0]jm\rangle\rangle = e^{i\theta(j)}(-1)^{j-m}|j-m, j+m\rangle$$
$$= e^{i\theta(j)} \frac{(-1)^{j-m}}{[(j+m)!(j-m)!]^{1/2}}\, b_1^{+\,j-m} b_2^{+\,j+m}|0,0\rangle \tag{9}$$

with $u(2)$, $su(2)$ and $so(2)$ labels linked by:

$$j = \frac{n}{2} = \frac{n_1 + n_2}{2},\ m = -\frac{n_1 - n_2}{2}$$

$e^{i\theta(j)}$ is a phase factor to be fixed later. From relations (1, 5) and using known properties [15,17] of the $u(2) \supset su(2) \supset so(2)$ chain we first built two sets of irreducible tensor operators with symmetry $[m_1\, 0]$ and $[0\ -m_2]$ in $u(2)$

$${}^{[m_1\,0]}T_m^{(j)} = (-1)^{\frac{m_1}{2}+m}\ \mathcal{N}(m_1, 0)$$
$$\times \left[\frac{m_1!}{(\frac{m_1}{2} - m)!(\frac{m_1}{2} + m)!}\right]^{1/2}\ b_1^{+(\frac{m_1}{2}-m)} b_2^{+(\frac{m_1}{2}+m)} \tag{10}$$

with $j = m_1/2$, and

$${}^{[0\,-m_2]}T_m^{(j)} = \mathcal{N}(0, m_2)$$
$$\times \left[\frac{m_2!}{(\frac{m_2}{2} - m)!(\frac{m_2}{2} + m)!}\right]^{1/2}\ b_1{}^{(\frac{m_2}{2}+m)} b_2{}^{(\frac{m_2}{2}-m)} \tag{11}$$

with $j = m_2/2$. $\mathcal{N}(m_1, 0)$ and $\mathcal{N}(0, m_2)$ are normalization coefficients partly fixed with the condition that the operator (10) acting on the vacuum state gives a standard covariant $2d$ state:

$$|[n\,0]jm\rangle\rangle \equiv {}^{[n\,0]}T_m^{(j)}\ |0,0\rangle. \tag{12}$$

2.2 General operators

Two kinds of operators may be built from the previous (10, 11) ones. Those, denoted *pitos* in [7], which are symmetry adapted in the whole $u(2) \supset su(2) \supset so(2)$ chain:

$${}^{[m_1\ -m_2]}T_m^{(j)} = \left[{}^{[m_1\,0]}T^{(\frac{m_1}{2})} \times {}^{[0\,-m_2]}T^{(\frac{m_2}{2})}\right]_m^{(j)}$$
$$= C\begin{matrix} q_1 & q_2 & (j) \\ (\frac{m_1}{2} & \frac{m_2}{2}) & m \end{matrix}\ {}^{[m_1\,0]}T_{q_1}^{(\frac{m_1}{2})}\ {}^{[0\,-m_2]}T_{q_2}^{(\frac{m_2}{2})} \tag{13}$$

where $j = (m_1 + m_2)/2$ and C is a stretched $su(2)$ Clebsch-Gordan coefficient given in [14]. In some cases, for instance if one considers an E electronic state, these operators are

[1] When there is no ambiguity the Einstein summation convention for indices out of parentheses is used.

sufficient; however sometimes one needs more general operators as for instance when dealing with vibrational E modes. These operators, denoted *gitos* in [7], are obtained by allowing j to take all values allowed by the usual $su(2)$ rule:

$$j_{min} = \frac{|m_1 - m_2|}{2} \leq j \leq \frac{m_1 + m_2}{2} = j_{max}.$$

We have then

$$^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{m} = \left[{}^{[m_1\,0]}T^{(\frac{m_1}{2})} \times {}^{[0\,-m_2]}T^{(\frac{m_2}{2})}\right]^{(j)}_{m} \tag{14}$$

but this time the labeling $[m_1\;-m_2]$ is not linked to an $u(2)$ symmetry in Gel'fand notation. m_1 (or m_2) simply gives the power in elementary creation (or annihilation) operators in $^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{m}$ written in normal form. The operators (14) may also be written

$$^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{m} = {}^{\{m_1\,m_2\}}\mathrm{g}_j(N_1{+}N_2)\;\; {}^{[m_1'\,-m_2']}T^{(j)}_{m} \tag{15}$$

where $^{[m_1'\,-m_2']}T^{(j)}_{m}$ are the $2d$ standard operators (13). The labels m_1' and m_2' represent the "true" $u(2)$ symmetry of the operators (14) and are related to m_1, m_2 and j by

$$m_1' = \frac{m_1 - m_2}{2} + j,\; m_2' = \frac{m_2 - m_1}{2} + j. \tag{16}$$

It is easily checked that $j = (m_1' + m_2')/2$. $^{\{m_1\,m_2\}}\mathrm{g}_j$ is a polynomial function of the $u(2)$ linear invariant $N_1 + N_2$ given by[2]:

$$^{\{m_1\,m_2\}}\mathrm{g}_j(N_1{+}N_2) = \left[\frac{(2j+1)!}{(j_{max}+j+1)!(j_{max}-j)!}\right]^{1/2} \times (N_1 + N_2 + \frac{m_2 - m_1}{2} - j)^{[j_{max}-j]}. \tag{17}$$

2.3 Matrix elements in the 2d standard basis

Following previous conventions [11,12] the Wigner-Eckart theorem for covariant sets writes in terms of $su(2)$ Clebsch-Gordan coefficients:

$$\begin{aligned}\langle\langle [n''\,0]j''m''|{}^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{m}|[n'\,0]j'm'\rangle\rangle &= (2j''+1)^{-1/2} \\ \times C^{\,m\;m'\;\;(j'')*}_{\,(j\;j')\;\;m''} &\left([n''\,0]j''||{}^{[m_1\,-m_2]}\mathcal{T}^{(j)}||[n'\,0]j'\right) \\ = (2j''+1)^{-1/2} F^{\quad m \qquad\quad m' \qquad ([n''\,0]j'')*}_{([m_1'-m_2']j\;\;[n'\,0]j')\quad m''} \\ \times \left([n''\,0]j''||{}^{[m_1\,-m_2]}\mathcal{T}^{(j)}||[n'\,0]j'\right)\end{aligned} \tag{18}$$

where $*$ denotes complex conjugation. The operators are those given by equations (13, 14) of which those in equations (10, 11) are useful special cases. The F coefficients, which keep the full $u(2) \supset su(2)$ labels, are to be used in the next section. Reduced matrix elements for all operators may be obtained with:

$$\begin{aligned}\left([n''\,0]j''||{}^{[m_1\,-m_2]}\mathcal{T}^{(j)}||[n'\,0]j'\right) &= \delta_{n'',n'+m_1-m_2}\, i^{-m_2'} \\ \times \left[\frac{(2j+1)(n'+m_1'+1)!(n'-m_2')!}{(n'-m_2)!(n'-m_2)!(j_{max}+j+1)!(j_{max}-j)!}\right]^{1/2}&.\end{aligned} \tag{19}$$

2.4 Useful relations and examples

2.4.1 Phase choices

The results of the preceding sections involve several phase choices. In particular we set for the phase of states (9)

$$e^{i\theta(j)} = (-1)^{-j} = i^{-2j}, \tag{20}$$

and for the normalization coefficients

$$\begin{aligned}\mathcal{N}(m_1,m_2) &= \mathcal{N}(m_1,0)\,\mathcal{N}(0,m_2) \\ &= (-1)^{m_2}\; i^{m_1}[m_1!m_2!]^{-1/2}.\end{aligned} \tag{21}$$

These phase choices have been made in order to impose particular properties to the various operators under adjunction (†) and time reversal ($\mathcal{K}_t$):

$$\begin{aligned}{}^{[m_1\,-m_2]}\mathcal{T}^{\dagger(j)}_{m} &= (-1)^{j-m}\;\left({}^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{-m}\right)^{\dagger} \\ &= i^{m_1-m_2}\;{}^{[m_2\,-m_1]}\mathcal{T}^{(j)}_{m}\end{aligned} \tag{22}$$

$$\mathcal{K}_t\;{}^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{m}\;\;\mathcal{K}_t^{\;-1} = {}^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{-m} \tag{23}$$

and identical relations for those in (13) with the substitution of $\mathcal{T}$ by T.

2.4.2 Some examples

Three sets of quantities are of special importance in practical applications. Those involving the fundamental $[1\,0]$ *irrep* of $u(2)$ and its adjoint $[0\;-1]$ which subduce to $j = 1/2$ in $su(2)$ and the adjoint representation $[1\;-1]$ spanned by the generators. Their bosonic realizations are explicitly given below together with their reduced matrix elements.

The fundamental tensors. With equations (10, 11) and (21) one gets:

$$\begin{aligned}&\mathcal{N}(1,0) = i, &&\mathcal{N}(0,1) = -1 \\ &{}^{[1\,0]}T^{(1/2)}_{-1/2} = ib_1^+ &&{}^{[1\,0]}T^{(1/2)}_{1/2} = -ib_2^+ \\ &{}^{[0\,-1]}T^{(1/2)}_{-1/2} = -b_2 &&{}^{[0\,-1]}T^{(1/2)}_{1/2} = -b_1,\end{aligned} \tag{24}$$

and the two dimensional fundamental state is spanned by:

$$||[1\,0]\frac{1}{2}m\rangle\rangle = {}^{[1\,0]}T^{(1/2)}_{m}\;|0,0\rangle. \tag{25}$$

[2] For an arbitrary operator X we have $X^{[k]} = X \times (X-1) \times ... \times (X-k+1)$.

Within arbitrary states $|[n\,0]jm\rangle\rangle$ the reduced matrix elements are given by

$$\left([n'\,0]j'||^{[1\,0]}T^{(\frac{1}{2})}||[n\,0]j\right) = i\left([n\,0]j||^{[0\,-1]}T^{(\frac{1}{2})}||[n'\,0]j'\right) = [(n+2)(n+1)]^{1/2}\ \delta_{n',n+1} \quad (26)$$

Generators. With $\mathcal{N}(0,1) = -1$ and equation (13) we get:

$$^{[1\,-1]}T^{(1)}_{1} = iJ_{-},\quad {}^{[1\,-1]}T^{(1)}_{-1} = -iJ_{+}$$
$$^{[1\,-1]}T^{(1)}_{0} = -i\sqrt{2}J_z \quad (27)$$

with reduced matrix elements

$$\left([n'\,0]j'||^{[1\,-1]}T^{(1)}||[n\,0]j\right) = -\,i\left[\frac{(n+2)(n+1)n}{2}\right]^{1/2}\ \delta_{n',n}. \quad (28)$$

We recall that in equations (26, 28) the labels n and j are related by $j = n/2$.

3 Symmetry adaptation in a point group G

The natural subduction $\mathcal{D}^{(\frac{1}{2})} \downarrow G$, where G is a point group and $\mathcal{D}^{(\frac{1}{2})}$ the $su(2)$ fundamental *irrep*, gives a two dimensional spinor *irrep* (or two one dimensional ones). Yet $u(2)$ appears as a degeneracy algebra for a two dimensional oscillator [2] as well as an invariance algebra for an electronic E state [9]. This means that the mapping of an E *irrep* to a $D^{(\frac{1}{2})}$ one involves algebraic properties in configuration space and not geometrical ones; hence the notation $u(2) \supset su^{*}(2) \supset G$ used for this non canonical symmetry adaptation. We first consider the case of groups which admit an E *irrep* of integer type. This implies that the matrices $D^{(E)}(R)$ $(R \in G)$ may be transformed to real form [18]. Other groups for which the "E" *irrep* is in fact made of two one dimensional conjugate *irreps* will be treated separately.

3.1 Some fundamental properties

(i) We first recall that within our formalism [12,19] the covariant components of an irreducible tensor operator with symmetry Γ with respect to a group G are characterized by the transformation laws

$$P_R\,T^{(\Gamma)}_{\sigma}P_{R^{-1}} = [D^{(\Gamma)^{*}}(R)]^{\sigma'}_{\sigma}\,T^{(\Gamma)}_{\sigma'}. \quad (29)$$

We stress that in (29) the upper index in the matrix is a row index. Under adjunction equation (29) gives

$$P_R\,T^{(\Gamma)\dagger}_{\sigma}P_{R^{-1}} = [D^{(\Gamma)}(R)]^{\sigma'}_{\sigma}\,T^{(\Gamma)\dagger}_{\sigma'} \quad (30)$$

We may thus define the adjoint irreducible tensor operator by [12]

$$T^{\dagger(\Gamma)}_{\sigma} = e^{i\alpha_\Gamma}\begin{pmatrix}\Gamma\\ \sigma\ \sigma'\end{pmatrix}T^{(\Gamma)\dagger}_{\sigma'}, \quad (31)$$

where, within a phase, the symbol on the right-hand side is the U matrix which transforms Γ into its complex conjugate [18]. If the matrices for the *irrep* Γ are real the metric tensor may be, and usually is, chosen so that it reduces to the identity. So we have:

$$T^{\dagger(\Gamma)}_{\sigma} = T^{(\Gamma)\dagger}_{\sigma}. \quad (32)$$

(ii) As it is known any unitary transformation of the set $\{b_i\}_{i=1,2}$ preserves the bosonic commutation relations together with the operator $N = N_1 + N_2$. Assuming that the sets $\{b_i\}_{i=1,2}$ (or $\{b_i^{+}\}_{i=1,2}$) span an *irrep* of type E of G the quantities

$$\begin{cases} b^{(E)}_{1} = \alpha^1_1\,b_1 + \alpha^2_1\,b_2\\ b^{(E)}_{2} = \alpha^1_2\,b_1 + \alpha^2_2\,b_2\end{cases} \qquad \begin{cases} b^{(E)\dagger}_{1} = \alpha^{1*}_1\,b_1^{+} + \alpha^{2*}_1\,b_2^{+}\\ b^{(E)\dagger}_{2} = \alpha^{1*}_2\,b_1^{+} + \alpha^{2*}_2\,b_2^{+}\end{cases} \quad (33)$$

where (α^i_j) is a unitary matrix, span an equivalent E type *irrep* and the set $\left\{b^{(E)\dagger}_{\sigma}b^{(E)}_{\sigma'}\right\}_{\sigma,\sigma'=1,2}$ is then a generator system of $u(2)$ equivalent to the set (1) built from $\{b_i^{+}b_j\}_{i,j=1,2}$.

In order to restrict the arbitrariness of the α^i_j coefficients we must choose an orientation for the matrices $D^{(E)}(R)$ of the considered *irrep* E. Also for the systems we want to study the J_z operator, which is the $so(2)$ generator, transforms as the one dimensional *irrep* A_2 (or A_{2g}, A'_2) appearing in the antisymmetrized product $\{E \times E\}$.

3.2 The matrices $\mathbf{D^{(E)}(R)}$ are real

3.2.1 The orientation matrix

The assumption of real $D^{(E)}(R)$ matrices was made in [7,8]. For all groups considered here and for a given *irrep* E_r of type E the matrices for the generators X and Y, in the passive point of view, are usually taken as [20]

$$D^{(E_r)}(X) = \begin{pmatrix}\cos r\psi & \sin r\psi\\ -\sin r\psi & \cos r\psi\end{pmatrix}$$
$$D^{(E_r)}(Y) = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix} \quad (34)$$

and in some cases a third Z generator with matrix representative

$$D^{(E_{r_\alpha})}(Z) = \pm\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}, \quad (35)$$

in which case the substitution $E_r \to E_{r_\alpha}$ is also made in (34); α is an additional index needed for direct product groups. The choices for the elements X, Y, Z, the appropriate angle ψ, the possible r and α values appearing in equations (34, 35) are given in Appendix A. All matrices being real, and denoting by E any two dimensional *irrep* (E_r or E_{r_α}) of G, we can set (Eq. (32)):

$$b^{(E)\dagger}_{\sigma} = b^{\dagger(E)}_{\sigma} = b^{+(E)}_{\sigma}, \quad (36)$$

where in the right member we use the conventional notation. With equations (33, 36) and the condition that J_z be of symmetry A_2 (or A_{2g}, A_2') it may be shown that we have the set of equivalent solutions:

$$b_1 = \frac{e^{i\varphi}}{\sqrt{2}}(b^{(E)}_1 \pm i b^{(E)}_2), \quad b_1^+ = \frac{e^{-i\varphi}}{\sqrt{2}}(b^{+(E)}_1 \mp i b^{+(E)}_2)$$
$$b_2 = \frac{e^{i\kappa}}{\sqrt{2}}(b^{(E)}_1 \mp i b^{(E)}_2), \quad b_2^+ = \frac{e^{-i\kappa}}{\sqrt{2}}(b^{+(E)}_1 \pm i b^{+(E)}_2) \tag{37}$$

of which we choose in the following

$$b_1 = \frac{1}{\sqrt{2}}(b^{(E)}_1 + i b^{(E)}_2), \quad b_1^+ = \frac{1}{\sqrt{2}}(b^{+(E)}_1 - i b^{+(E)}_2)$$
$$b_2 = \frac{1}{\sqrt{2}}(b^{(E)}_1 - i b^{(E)}_2), \quad b_2^+ = \frac{1}{\sqrt{2}}(b^{+(E)}_1 + i b^{+(E)}_2). \tag{38}$$

This result allows one to determine easily the transformation laws of the tensor components (13) (as well as those of (15)). It appears in particular that each subspace characterized by $\ell = 2|m|$ spans a two dimensional representation of G (one dimensional for $\ell = 0$) whose reduction in G is straightforwardly found. As a result symmetry adapted tensor operators are obtained with:

$$^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma\sigma} = \, ^{[m_1\,-m_2]}G^{m}_{\ell\Gamma\sigma}\; ^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{m} \tag{39}$$

where the orientation matrix $^{[m_1\,-m_2]}G$ is given by

$$^{[m_1\,-m_2]}G^{\ell/2}_{\ell\Gamma\sigma} = \mathcal{N}(\ell)\,\mu^{(\Gamma\sigma)}(\ell)\, i^{\frac{m_2-m_1}{2}-\frac{\ell}{2}}$$
$$^{[m_1\,-m_2]}G^{-\ell/2}_{\ell\Gamma\sigma} = (-1)^{m_1}\,\Pi(\Gamma\sigma)\,\mathcal{N}(\ell) \times \mu^{(\Gamma\sigma)}(\ell)\, i^{\frac{m_2-m_1}{2}-\frac{\ell}{2}}. \tag{40}$$

with

$$\bullet \Pi(\Gamma_\sigma) = 1, \quad \mu^{(\Gamma_\sigma)}(\ell) = 1 \quad \text{if } \Gamma = A_{1_\tau}, B_{1_\tau} \text{ or } E_{r'_\tau}1$$
$$\bullet \Pi(\Gamma_\sigma) = -1, \quad \mu^{(\Gamma_\sigma)}(\ell) = \mu(\ell) \quad \text{if } \Gamma = A_{2_\tau}, B_{2_\tau} \text{ or } E_{r'_\tau}2. \tag{41}$$

Also $\mathcal{N}(\ell) = 1/\sqrt{2}$ (or 1) for $\ell \neq 0$ (or $\ell = 0$) and the τ label is needed only when $E = E_{r\alpha}$. The allowed symmetries Γ, for fixed ℓ, together with the phase factor $\mu(\ell)$ for common point groups and all possible E type *irrep* are given in Appendix B.

Likewise from the general tensor operators (15) we build symmetry adapted ones with

$$^{[m_1\,-m_2]}T^{(j)}_{\ell\Gamma\sigma} = \, ^{[m_1'\,-m_2']}G^{m}_{\ell\Gamma\sigma}\; ^{[m_1\,-m_2]}T^{(j)}_{m} = \, ^{\{m_1,m_2\}}\mathrm{g}_j(N_1+N_2)\; ^{[m_1'\,-m_2']}\mathcal{T}^{(j)}_{\ell\Gamma\sigma} \tag{42}$$

where m_1' and m_2' are given by (16) and the $^{[m_1'\,-m_2']}G^{m}_{\ell\Gamma\sigma}$ elements are those defined by equations (40, 41). Various phase choices lead to the following useful properties:

$$^{[m_1\,-m_2]}G^{m*}_{\ell\Gamma\sigma} = (-1)^{\frac{m_1+m_2}{2}+m}\, i^{m_2-m_1}\; ^{[m_2\,-m_1]}G^{-m}_{\ell\Gamma\sigma} = (-1)^{\frac{m_1+m_2}{2}-\frac{\ell}{2}}\; ^{[m_1\,-m_2]}G^{-m}_{\ell\Gamma\sigma} \tag{43}$$

$$^{[m_1\,-m_2]}G^{-m}_{\ell\Gamma\sigma} = (-1)^{m_1}\;\Pi(\Gamma\sigma)\; ^{[m_1\,-m_2]}G^{m}_{\ell\Gamma\sigma} \tag{44}$$

As a consequence we have the two properties upon adjunction and time reversal:

$$\left(^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma\sigma}\right)^\dagger = \, ^{[m_2\,-m_1]}\mathcal{T}^{(j)}_{\ell\Gamma\sigma} = \, ^{[m_1\,-m_2]}\mathcal{T}^{\dagger(j)}_{\ell\Gamma\sigma}$$
$$\mathcal{K}_t\; ^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma\sigma}\;\mathcal{K}_t^{\,-1} = (-1)^{j-\frac{\ell}{2}}\; ^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma\sigma} \tag{45}$$

with similar relations for operators in (39) with the substitution of $\mathcal{T}$ by T. Some $^{[m_1\,-m_2]}G^{m}_{\ell\Gamma\sigma}$ coefficients are given in Appendix C.

3.2.2 Matrix elements in the symmetry adapted basis

From equation (12) and the results of the previous section symmetry adapted states are given by:

$$|[n\,0]j\ell\Gamma\sigma\rangle\rangle = \, ^{[n\,0]}\mathcal{T}^{(j)}_{\ell\Gamma\sigma}\;|0,0\rangle = \sum_m \, ^{[n\,0]}G^{m}_{\ell\Gamma\sigma}\;|[n\,0]jm\rangle\rangle. \tag{46}$$

Starting from the expression of matrix elements (18) in the standard basis one obtains easily those in the symmetry adapted basis:

$$\langle\langle[n''\,0]j''\ell''\Gamma''\sigma''|^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma\sigma}|[n'\,0]j'\ell'\Gamma'\sigma'\rangle\rangle = (2j''+1)^{-\frac{1}{2}}\, F\begin{matrix} \ell\Gamma\sigma & \ell'\Gamma'\sigma' & ([n''\,0]j'')* \\ ([m_1'\,-m_2']j & [n'\,0]j') & \ell''\Gamma''\sigma'' \end{matrix} \times \left([n''\,0]j''||^{[m_1\,-m_2]}\mathcal{T}^{(j)}||[n'\,0]j'\right) \tag{47}$$

where the reduced matrix elements are those in (19) and where

$$F\begin{matrix} \ell\Gamma\sigma & \ell'\Gamma'\sigma' & ([n''\,0]j'') \\ ([m_1'\,-m_2']j & [n'\,0]j') & \ell''\Gamma''\sigma'' \end{matrix} = \sum_{m,m',m''} \, ^{[m_1'\,-m_2']}G^{m\,*}_{\ell\Gamma\sigma}\; ^{[n'\,0]}G^{m'\,*}_{\ell'\Gamma'\sigma'}\; ^{[n''\,0]}G^{m''}_{\ell''\Gamma''\sigma''} \times F\begin{matrix} m & m' & ([n''\,0]j'') \\ ([m_1'\,-m_2']j & [n'\,0]j') & m'' \end{matrix} \tag{48}$$

are symmetry adapted Clebsch-Gordan coefficients.

3.2.3 Some examples

We first consider the same operators than in Section 2.4.2 and give the expressions of their symmetry adapted components. Next we relate some of our results to another formalism used for spherical tops and C_{3v} molecules.

The fundamental tensors. We use equations (39–41) and the property which gives for any group G in Appendix A and any E type *irrep*

$$[m_1\ -m_2] = [1\,0] \text{ or } [0\ -1] \Rightarrow E_{r'} = E_r \quad \ell = 1 \quad \mu(\ell) = -i.$$

With the ${}^{[m_1\ -m_2]}G$ elements in Tables 6, 7 and equation (24) we have:

$$\begin{aligned} {}^{[1\,0]}T^{(1/2)}_{1E1} &= -\frac{1}{\sqrt{2}}(b_1^+ + b_2^+) = -b^{+(E)}_{\ 1} \\ {}^{[1\,0]}T^{(1/2)}_{1E2} &= -\frac{i}{\sqrt{2}}(b_1^+ - b_2^+) = -b^{+(E)}_{\ 2} \\ {}^{[0\,-1]}T^{(1/2)}_{1E1} &= -\frac{1}{\sqrt{2}}(b_1 + b_2) = -b^{(E)}_{\ 1} \\ {}^{[0\,-1]}T^{(1/2)}_{1E2} &= \frac{i}{\sqrt{2}}(b_1 - b_2) = -b^{(E)}_{\ 2}, \end{aligned} \tag{49}$$

and the two dimensional fundamental state is spanned by:

$$|[1\,0]\frac{1}{2}1E\sigma\rangle\rangle = {}^{[1\,0]}T^{(1/2)}_{1E\sigma}\ |0,0\rangle. \tag{50}$$

Generators. Likewise from equation (27) we get the general solutions:

$$\begin{aligned} &{}^{[1\,-1]}T^{(1)}_{0A_2} = -\sqrt{2}J_z, \\ &{}^{[1\,-1]}T^{(1)}_{2E_{r'}1} = \sqrt{2}J_x, \quad {}^{[1\,-1]}T^{(1)}_{2E_{r'}2} = -i\sqrt{2}\mu(2)J_y \end{aligned} \tag{51}$$

with:

- $r' = 2r$ when $2r \le (n-1)/2$ (n odd) ($2r \le n/2 - 1$ (n even)) for groups in $G_{(I)}$ and $\mu(2) = -i$,
- $r' = n - 2r$ when $2r > (n-1)/2$ (n odd) ($2r > n/2 - 1$ (n even)) for groups in $G_{(I)}$ and $\mu(2) = i$,
- for D_{nd} (n even) groups change n by $2n$,
- for groups in $G_{(II)}$ the same rules applies with in addition the labels $'$ or g whatever $\alpha = {}'$, $''$, u or g in $E_{r\alpha}$,
- for groups in $G_{(III)}$ $r' = 2r$ and $\mu(2) = -i$ with parity g in $D_{\infty h}$.

The special cases $r = n/4$ for C_{nv} and D_n groups ($r = n/2$ for D_{nd} (n even)) give:

$$\begin{aligned} &{}^{[1\,-1]}T^{(1)}_{0A_2} = -\sqrt{2}J_z, \\ &{}^{[1\,-1]}T^{(1)}_{2B_1} = \sqrt{2}J_x, \quad {}^{[1\,-1]}T^{(1)}_{2B_2} = \sqrt{2}J_y. \end{aligned} \tag{52}$$

For groups in $G_{(II)}$ add the parity $'$ or g whatever α.

We note here that if the operators in equation (49) are interpreted as creation and annihilation operators for a vibrational normal mode s with E symmetry type:

$$b^{(E)}_{\ \sigma} = \frac{1}{\sqrt{2}}({}_sq^{(E)}_{\ \sigma} + i\ {}_sp^{(E)}_{\ \sigma}),$$

where ${}_sq^{(E)}_{\ \sigma}$ and ${}_sp^{(E)}_{\ \sigma}$ are the dimensionless coordinates and their conjugate momenta, it is easily shown with equations (1, 49) that for the $su(2)$ generator J_z we have:

$$J_z = -\frac{1}{2}({}_sq^{(E)}_{\ 1}\,{}_sp^{(E)}_{\ 2} - {}_sq^{(E)}_{\ 2}\,{}_sp^{(E)}_{\ 1}) = -\frac{1}{2}\ {}_s\ell_z$$

where ${}_s\ell_z$ is the angular momentum of the doubly degenerate oscillator. We shall see that a different interpretation can be given when the operators in (49) are associated with an electronic state with the same E symmetry type.

Correlations with other formalisms. These can be made mainly in the case of groups C_{3v}, O, T_d and O_h for which general schemes have also been proposed [21,22]. In these cases we have only one E type *irrep* (two in O_h differing only in parity) which is assumed to be associated with a doubly degenerate vibration. General vibrational operators are written in the form:

$$[\mathcal{A}^{+n_s(k\Gamma)} \times \mathcal{A}^{n'_s(k'\Gamma')}]^{(C_s)}_{\sigma} \tag{53}$$

where $\mathcal{A}^{+n_s(k\Gamma)}$ (or $\mathcal{A}^{n'_s(k'\Gamma')}$) is built from n_s creation $a^{+(E)}_{\sigma}$ (or n'_s annihilation $a^{(E)}_{\sigma}$) operators. We note that the operators $\mathcal{A}^{+n_s(k\Gamma)}$ (as well as those $A^{+(lm0,0\Gamma)}_{\sigma}$ defined in [22]) differ from those denoted $A_2^{+n_s(k\Gamma)}$ in [6] by the internal coupling, performed in T_d, of the elementary creation operators $a^{+(E)}$. The schemes described in [21,22] allow to obtain functionally independent $\mathcal{A}^{+n_s(k\Gamma)}$ for arbitrary values of n_s which is not the case of those in [6]. Explicitly we have [21]:

$$\mathcal{A}^{+n(k,\Gamma)}_{\ \sigma} = \frac{\alpha(k)}{\sqrt{2^{\frac{n+k}{2}-1}}} \left(\left(a^{+(E)}_{\ 1}\right)^2 + \left(a^{+(E)}_{\ 2}\right)^2\right)^{\frac{n-k}{2}} \times \sum_{s=0}^{[\frac{k}{2}]} \binom{k}{2s} (-1)^s \left(a^{+(E)}_{\ 1}\right)^{k-2s} \left(a^{+(E)}_{\ 2}\right)^{2s} \tag{54}$$

for $\Gamma\sigma = A_1, E1$ and

$$\mathcal{A}^{+n(k,\Gamma)}_{\ \sigma} = \frac{\beta(k)}{\sqrt{2^{\frac{n+k}{2}-1}}} \left(\left(a^{+(E)}_{\ 1}\right)^2 + \left(a^{+(E)}_{\ 2}\right)^2\right)^{\frac{n-k}{2}} \times \sum_{s=0}^{[\frac{k-1}{2}]} \binom{k}{2s+1} (-1)^s \left(a^{+(E)}_{\ 1}\right)^{k-2s-1} \left(a^{+(E)}_{\ 2}\right)^{2s+1} \tag{55}$$

for $\Gamma\sigma = A_2, E2$. $\alpha(k)$ and $\beta(k)$ are real phase factors, denoted $c_1(k)$ and $c_2(k)$ respectively in [22]:

$$\mathcal{A}^{+n(k,\Gamma)}_{\ \sigma} = (\sqrt{2})^{\frac{n-k}{2}-1} A^{+(\frac{n+k}{2}\,\frac{n-k}{2}0,0\Gamma)}_{\ \sigma}. \tag{56}$$

The computation of the matrix elements of the operators in equations (53–55) is made [6,21] through a recursive process based upon the knowledge of reduced matrix elements of the elementary operators $a^{+(E)}$ (or $a^{(E)}$) and with formulas for tensor recouplings made in G. A different approach is made in [22] but the expressions given involve power coupling coefficients $\mathcal{Y}$ expressed in terms of coupling coefficients of G.

Assimilating the $a^{+(E)}$ with our $b^{+(E)}$ in equation (38) it may be shown that the $\mathcal{A}^{+n(k,\Gamma)}_{\ \sigma}$ are related to the $2d$

symmetrized *pitos* in Section 3.2.1 by:

$$^{[n\,0]}T^{(j)}_{\ell\Gamma\sigma} = i^{-\frac{n}{2}+\frac{\ell}{2}}\,(-1)^n\,(-1)^\Gamma\,\alpha(\ell) \times \left[2^{\frac{n-\ell}{2}}\left(\frac{n+\ell}{2}\right)!\left(\frac{n-\ell}{2}\right)!\right]^{-1/2}\mathcal{A}^{+n(\ell\Gamma)}_{\sigma} \quad (57)$$

where the factor $(-1)^\Gamma$ follows the usual convention

$$(-1)^\Gamma = 1 \text{ if } \Gamma = A_1, E \text{ and } (-1)^\Gamma = -1 \text{ if } \Gamma = A_2.$$

We recall that for the groups considered here we have:

$$\mu(\ell) = -i \quad \text{for} \quad \ell = 3p;\ 3p+1$$
$$\mu(\ell) = i \quad \text{for} \quad \ell = 3p+2. \quad (58)$$

As the matrix elements of the $^{[n\,0]}T^{(j)}_{\ell\Gamma\sigma}$ are known those of $\mathcal{A}^{+n(\ell\Gamma)}_{\sigma}$ (or $A^{+(\frac{n+\ell}{2}\,\frac{n-\ell}{2}0,0\Gamma)}_{\sigma}$) can be determined. However there is some ambiguity in the phases of states defined in [6,21]. We set:

$$|n\,\ell\,\Gamma\sigma\rangle = \left[2^{\frac{n-\ell}{2}}\left(\frac{n+\ell}{2}\right)!\left(\frac{n-\ell}{2}\right)!\right]^{-1/2}\mathcal{A}^{+n(\ell,\Gamma)}_{\sigma}|0,0\rangle.$$

As a consequence there might be a ± 1 phase difference according to the ℓ values *mod* 6 between these states and those of [6,22]. With equations (19, 47, 48) it may be checked that the matrix elements are given by:

$$\begin{aligned}\langle n''\,\ell''\,\Gamma''_{\sigma''}|\,\mathcal{A}^{+n(\ell,\Gamma)}_{\sigma}\,|n'\,\ell'\,\Gamma'_{\sigma'}\rangle &= \Pi(\Gamma_\sigma,\Gamma'_{\sigma'},\Gamma''_{\sigma''})\\ &\times\mathcal{N}(\ell)\mathcal{N}(\ell')\,\mathcal{N}(\ell'')\times\mu^{(\Gamma_\sigma)}(\ell)\,\mu^{(\Gamma'_{\sigma'})}(\ell')\mu^{(\Gamma''_{\sigma''})}(\ell'')\\ &\times(-1)^{\frac{\ell+\ell'-\ell''}{2}}(-1)^{\frac{n-\ell}{2}}\,(-1)^{\Gamma+\Gamma'+\Gamma''}\alpha(\ell)\,\alpha(\ell')\,\alpha(\ell'')\\ &\times(1+\Pi(\Gamma_\sigma)\,\Pi(\Gamma'_{\sigma'})\,\Pi(\Gamma''_{\sigma''}))\,\sqrt{2^{\frac{n-\ell}{2}}}\\ &\times\left[\frac{\left(\frac{n''+\ell''}{2}\right)!\left(\frac{n''-\ell''}{2}\right)!}{\left(\frac{n'+\ell'}{2}\right)!\left(\frac{n'-\ell'}{2}\right)!}\right]^{1/2}\delta_{n'',n+n'} \quad (59)\end{aligned}$$

with

$$\Pi(\Gamma_\sigma,\Gamma'_{\sigma'},\Gamma''_{\sigma''}) = \begin{cases}\Pi(\Gamma_\sigma) & \text{if } \ell''=\ell-\ell'\\ \Pi(\Gamma'_{\sigma'}) & \text{if } \ell''=\ell'-\ell\\ \Pi(\Gamma''_{\sigma''}) & \text{if } \ell''=\ell+\ell'\\ 0 & \text{else}\end{cases}.$$

Equation (59) holds for all label values except in the cases when $\ell = \ell' = \ell'' = 0$ for which the right member in (59) must be divided by two in order to agree with normalization conventions. We note that with equations (56, 59) and applying the Wigner-Eckart theorem in G one obtain the $\mathcal{Y}$ coefficients of equation (19) of [22].

3.3 The matrices $D^{(E)}(R)$ are complex

The orientation (34, 35) of the preceding section is that mostly used in vibrational studies [6,23,24]. However it is not well adapted to the treatment of vibronic interactions in $E\otimes\varepsilon$ and $G'\otimes\varepsilon$ Jahn-Teller systems [25,9]. We could fixed the new matrices for the E *irreps* and make the symmetry adaptation again. In fact it is much simpler (especially with regards to phase choices) to proceed through a change of basis from the known results.

3.3.1 The new orientation for E *irreps*

From an arbitrary covariant set $\{T^{(E_r)}_{\sigma}\}$ transforming according to equation (29) with $\Gamma = E_r$ we define:

$$T^{(E_r)}_{\bar{1}} = \frac{e^{i\mu}}{\sqrt{2}}\left(iT^{(E_r)}_{1} - T^{(E_r)}_{2}\right)$$
$$T^{(E_r)}_{\bar{2}} = \frac{e^{i\mu}}{\sqrt{2}}\left(-iT^{(E_r)}_{1} - T^{(E_r)}_{2}\right). \quad (60)$$

With equation (34) it is easily shown that we have now:

$$P_R\,T^{(E_r)}_{\bar{\sigma}}P_{R^{-1}} = [D^{(E_r)^*}(R)]^{\bar{\sigma}'}_{\bar{\sigma}}\,T^{(E_r)}_{\bar{\sigma}'}, \quad (61)$$

with

$$D^{(E_r)}(X) = \begin{pmatrix} e^{-ir\psi} & 0\\ 0 & e^{ir\psi}\end{pmatrix},\quad D^{(E_r)}(Y) = \begin{pmatrix}0 & -1\\ -1 & 0\end{pmatrix}.$$

When the additional Z generator is needed equation (35) is unchanged. Applying this to the fundamental *irrep* $[1\,0]$ associated with the considered E_r symmetry and choosing $e^{i\mu} = 1$ we obtain:

$$|[1\,0]\frac{1}{2}1E_r\bar{1}\rangle\rangle = |[1\,0]\frac{1}{2}\frac{1}{2}\rangle\rangle$$
$$|[1\,0]\frac{1}{2}1E_r\bar{2}\rangle\rangle = |[1\,0]\frac{1}{2}-\frac{1}{2}\rangle\rangle \quad (62)$$

which we can write more generally:

$$|[1\,0]\frac{1}{2}1E\bar{\sigma}\rangle\rangle = |[1\,0]\frac{1}{2}m\rangle\rangle = {}^{[1\,0]}T^{(\frac{1}{2})}_{m}\,|0,0\rangle \quad (63)$$

with the correspondence $\bar{1} \leftrightarrow m = 1/2$, $\bar{2} \leftrightarrow m = -1/2$. Equation (63) traduces that for the fundamental representation $[1\,0]$ the symmetry adapted basis is the standard basis.

3.3.2 The new G matrix and consequences

Symmetry adapted tensor operators in the whole $u(2) \supset su(2)^* \supset G$ are now given by (Eq. (39)):

$$^{[m_1\,-m_2]}T^{(j)}_{\ell\Gamma\bar{\sigma}} = {}^{[m_1\,-m_2]}\bar{G}^{m}_{\ell\Gamma\bar{\sigma}}\ {}^{[m_1\,-m_2]}T^{(j)}_{m}. \quad (64)$$

Obviously for *irreps* of type A or B in any group G equations (40, 41) are still valid and $^{[m_1\,-m_2]}\bar{G} = {}^{[m_1\,-m_2]}G$.

For E type representations we have:

$$^{[m_1\,-m_2]}\bar{G}^{m}_{\ell E\bar{\sigma}} = {}^{(E)}U^{\sigma}_{\bar{\sigma}}\ {}^{[m_1\,-m_2]}G^{m}_{\ell E\sigma} \quad (65)$$

where

$$^{(E)}U = \frac{1}{\sqrt{2}}\begin{pmatrix} i & -i\\ -1 & -1\end{pmatrix} \quad (66)$$

is the unitary matrix associated with the change of basis (Eq. (60) with $\mu = 0$). Straightforward computations lead to $(\sigma = 1, 2)$:

$$^{[m_1\,-m_2]}\bar{G}^{\ell/2}_{\ell E\bar{\sigma}} = \frac{i^\sigma}{\sqrt{2}}\ {}^{[m_1\,-m_2]}G^{\ell/2}_{\ell E\sigma}\,(1-(-1)^\sigma i\mu(\ell))$$

$$^{[m_1\,-m_2]}\bar{G}^{-\ell/2}_{\ell E\bar{\sigma}} = \frac{i^\sigma}{\sqrt{2}}\ {}^{[m_1\,-m_2]}G^{-\ell/2}_{\ell E\sigma}\,(1+(-1)^\sigma i\mu(\ell))$$

which show that, since $\mu(\ell) = \pm i$, in all cases the summation in (64) reduces to one term. Explicitly this gives:

- for $\mu(\ell) = -i$

$$^{[m_1\,-m_2]}T^{(j)}_{\ell E\bar{1}} = e^{i\varphi}\ ^{[m_1\,-m_2]}T^{(j)}_{\ell/2}$$
$$^{[m_1\,-m_2]}T^{(j)}_{\ell E\bar{2}} = (-1)^{m_1+1}e^{i\varphi}\ ^{[m_1\,-m_2]}T^{(j)}_{-\ell/2} \quad (67)$$

- for $\mu(\ell) = i$

$$^{[m_1\,-m_2]}T^{(j)}_{\ell E\bar{1}} = (-1)^{m_1}e^{i\varphi}\ ^{[m_1\,-m_2]}T^{(j)}_{-\ell/2}$$
$$^{[m_1\,-m_2]}T^{(j)}_{\ell E\bar{2}} = -e^{i\varphi}\ ^{[m_1\,-m_2]}T^{(j)}_{\ell/2} \quad (68)$$

where $e^{i\varphi} = i^{\frac{m_2-m_1}{2}-\frac{\ell}{2}+1}$. The preceding relations apply equally when the substitution $T \to \mathcal{T}$ is made. We note that for groups admitting several E type *irreps* the transformation (Eq. (60)) may be performed to all or only some E *irreps*. Defining for E *irrep*

$$\bar{\sigma} = \bar{1}, \quad -\bar{\sigma} = \bar{2}; \quad \bar{\sigma} = \bar{2}, \quad -\bar{\sigma} = \bar{1} \quad (69)$$

the properties of the $^{[m_1\,-m_2]}\bar{G}$ matrix elements may be written for any $\Gamma\bar{\sigma}$ by:

$$(\ ^{[m_1\,-m_2]}\bar{G}^{m}_{\ell\Gamma\bar{\sigma}})^* = (-1)^{\frac{m_1+m_2}{2}-\frac{\ell}{2}}\ ^{[m_1\,-m_2]}\bar{G}^{-m}_{\ell\Gamma-\bar{\sigma}}$$
$$= (-1)^{\frac{m_1+m_2}{2}+m}\ i^{m_2-m_1}\ ^{[m_2\,-m_1]}\bar{G}^{-m}_{\ell\Gamma-\bar{\sigma}} \quad (70)$$

The properties upon adjunction and time reversal (45) of the new symmetry adapted tensor operators become:

$$^{[m_1\,-m_2]}\mathcal{T}^{(j)\dagger}_{\ell\Gamma\bar{\sigma}} = {}^{[m_2\,-m_1]}\mathcal{T}^{(j)}_{\ell\Gamma-\bar{\sigma}} = {}^{[m_1\,-m_2]}\mathcal{T}^{\dagger(j)}_{\ell\Gamma-\bar{\sigma}}$$
$$\mathcal{K}_t\ ^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma\bar{\sigma}}\ \mathcal{K}_t^{\,-1} = (-1)^{j-\frac{\ell}{2}}\ ^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma-\bar{\sigma}}. \quad (71)$$

Matrix elements in the new symmetry adapted basis are given by relation (47) in which the substitution $\sigma \to \bar{\sigma}$ is made everywhere, the reduced matrix elements being unchanged. Likewise the new symmetry adapted Clebsch-Gordan coefficients are given by (48) with the substitution

$$^{[m_1\,-m_2]}G^{m}_{\ell\Gamma\sigma} \to {}^{[m_1\,-m_2]}\bar{G}^{m}_{\ell\Gamma\bar{\sigma}}. \quad (72)$$

3.3.3 Examples

We take again the cases of Sections 2.4.2, 3.2.3.

The fundamental tensors. With $\mu(1) = -i$ equations (67, 68) give:

$$^{[1\,0]}T^{(1/2)}_{1E\bar{1}} = {}^{[1\,0]}T^{(1/2)}_{1/2} = -ib_2^+$$
$$^{[1\,0]}T^{(1/2)}_{1E\bar{2}} = {}^{[1\,0]}T^{(1/2)}_{-1/2} = ib_1^+$$
$$^{[0\,-1]}T^{(1/2)}_{1E\bar{1}} = i^{[0\,-1]}T^{(1/2)}_{1/2} = -ib_1$$
$$^{[0\,-1]}T^{(1/2)}_{1E\bar{2}} = -i^{[0\,-1]}T^{(1/2)}_{-1/2} = ib_2 \quad (73)$$

and for the two dimensional fundamental state we still have:

$$|[1\,0]\frac{1}{2}1E\bar{\sigma}\rangle\rangle = {}^{[1\,0]}T^{(1/2)}_{1E\bar{\sigma}}\ |0,0\rangle. \quad (74)$$

Generators. For the two main cases (Sect. 3.2.3) we find:

– for $\mu(2) = -i$

$$^{[1\,-1]}T^{(1)}_{0A_2} = -i^{[1\,-1]}T^{(1)}_{0} = -\sqrt{2}J_z$$
$$^{[1\,-1]}T^{(1)}_{2E_{r'}\bar{1}} = {}^{[1\,-1]}T^{(1)}_{1} = iJ_-$$
$$^{[1\,-1]}T^{(1)}_{2E_{r'}\bar{2}} = {}^{[1\,-1]}T^{(1)}_{-1} = -iJ_+ \quad (75)$$

– for $\mu(2) = i$

$$^{[1\,-1]}T^{(1)}_{0A_2} = -i^{[1\,-1]}T^{(1)}_{0} = -\sqrt{2}J_z$$
$$^{[1\,-1]}T^{(1)}_{2E_{r'}\bar{1}} = -{}^{[1\,-1]}T^{(1)}_{-1} = iJ_+$$
$$^{[1\,-1]}T^{(1)}_{2E_{r'}\bar{2}} = -{}^{[1\,-1]}T^{(1)}_{1} = -iJ_-. \quad (76)$$

In the special cases equation (52) is unchanged.

4 Applications

The preceding formalism can be used in several areas of spectroscopy of which we consider below several cases.

4.1 Conventional vibrational studies

We start from the assumption that a given molecule has an E type mode in its full vibrational representation. The possible modes are given in Appendix A of [9] for all molecular structures. The results of the preceding sections determine all symmetry adapted states $|[n\,0]j\ell\Gamma\sigma\rangle\rangle$ (or $|[n\,0]j\ell\Gamma\bar{\sigma}\rangle\rangle$) and all vibrational operators $^{[m_1\,-m_2]}\mathcal{V}^{(j)}_{\ell\Gamma\sigma}$ (or $^{[m_1\,-m_2]}\mathcal{V}^{(j)}_{\ell\Gamma\bar{\sigma}}$) associated with that mode. In particular, as mentioned previously, the generator with symmetry A_2 is interpreted as the oscillator angular momentum:

$$^{[1\,-1]}T^{(1)}_{0A_2} = -i^{[1\,-1]}T^{(1)}_{0} = \frac{1}{\sqrt{2}}\,{}_s\ell_z$$

and the linear invariant is nothing but the usual number operator. Results in Appendix B give the symmetries of these operators, as a function of ℓ, for all common point groups. From this we can first build an effective vibrational Hamiltonian, linear combination of Hermitian and time-reversal invariant operators with $m_1 = m_2$:

$$\mathcal{H}_{vib} = \sum_{m_1 j\ell}\ {}^{m_1}t_{j\ell}\ ^{[m_1\,-m_1]}\mathcal{V}^{(j)}_{\ell A_1} \quad (77)$$

where the $^{m_1}t_{j\ell}$ are real parameters. In (77) $j = 0,...m_1$ and (Eqs. (45, 71)) $(-1)^{j-\frac{\ell}{2}} = 1$. Keeping only terms up

Table 1. Expressions for ${}^{[m_1\,-m_1]}\mathcal{T}^{(j)}_{\ell A_1}$ terms in equation (78).

${}^{[1\,-1]}\mathcal{V}^{(0)}_{0A_1}$	$\frac{1}{\sqrt{2}}(N_1+N_2)$
${}^{[2\,-2]}\mathcal{V}^{(0)}_{0A_1}$	$\frac{1}{2\sqrt{3}}(N_1+N_2)(N_1+N_2-1)$
${}^{[2\,-2]}\mathcal{V}^{(2)}_{0A_1}$	$\frac{1}{\sqrt{6}}(J^2-3J_z^2)$
${}^{[2\,-2]}\mathcal{V}^{(2)}_{4A_1}$	$\frac{1}{2\sqrt{2}}(J_+^2+J_-^2)$
${}^{[3\,-3]}\mathcal{V}^{(0)}_{0A_1}$	$\frac{1}{12}(N_1+N_2)(N_1+N_2-1)(N_1+N_2-2)$
${}^{[3\,-3]}\mathcal{V}^{(2)}_{0A_1}$	$\frac{1}{6}(N_1+N_2-2)(J^2-3J_z^2)$
${}^{[3\,-3]}\mathcal{V}^{(3)}_{6A_1}$	$\frac{1}{2\sqrt{3}}(J_+^3+J_-^3)$

to order three in the generators give us

$$\begin{aligned}\mathcal{H}_{vib} = {}&{}^1t_{00}\ {}^{[1\,-1]}\mathcal{V}^{(0)}_{0A_1} + {}^2t_{00}\ {}^{[2\,-2]}\mathcal{V}^{(0)}_{0A_1}\\ &+ {}^2t_{20}\ {}^{[2\,-2]}\mathcal{V}^{(2)}_{0A_1}\\ &+ {}^2t_{24}\ {}^{[2\,-2]}\mathcal{V}^{(2)}_{4A_1} \quad \text{(a)}\\ &+ {}^3t_{00}\ {}^{[3\,-3]}\mathcal{V}^{(0)}_{0A_1} + {}^3t_{20}\ {}^{[3\,-3]}\mathcal{V}^{(2)}_{0A_1}\\ &+ {}^3t_{36}\ {}^{[3\,-3]}\mathcal{V}^{(3)}_{6A_1} \quad \text{(b)} \end{aligned} \tag{78}$$

The conditions for the terms (a), (b) to be retained in the expansion for a given E_r mode are for groups in $G_{(I)}$:

$$\text{(a)}\begin{cases} r=n/4 & G=C_{nv},\ D_n\\ r=n/2 & G=D_{nd}\ (n\ \text{even})\end{cases}$$
$$\text{(b)}\begin{cases} r=n/6 & G=C_{nv},\ D_n\\ r=n/3 & G=C_{nv},\ D_n,\ O,\ T_d\,.\\ r=n/3,2n/3 & G=D_{nd}\ (n\ \text{even})\end{cases} \tag{79}$$

The same conditions apply for groups in $G_{(II)}$ for an $E_{r\alpha}$ mode for all α. Groups in $G_{(III)}$ have only E_1 ($E_{1\alpha}$) vibrational modes thus only terms with $\ell=0$, j even appear in the expansions (77) and (78). Explicit expressions for the terms in (78) are given in Table 1. Matrix elements of the various operators are easily computed with the results obtained in the preceding sections. As far as only totally symmetric operators are considered both orientation of Sections 3.2 and 3.3 are equivalent. In particular we note that all $\ell=0$ operators are diagonal since:

$$F\begin{matrix} 0A_1 & l'\Gamma'\sigma' & ([n''\,0]j'')\\ ([m_1\,-m_2]j & [n'\,0]j') & l''\Gamma''\sigma''\end{matrix} = C\begin{matrix}0 & \ell'/2 & (j'')\\ (j & j') & \ell''/2\end{matrix} \times \delta_{j'',j'+\frac{m_1-m_2}{2}}\ \delta_{\ell',\ell''}\ \delta_{\Gamma',\Gamma''}\ \delta_{\sigma',\sigma''}. \tag{80}$$

As an illustration we take again the case of spherical tops and C_{3v} molecules [6,21,22]. With equations (78, 79) we have:

$$\begin{aligned}\mathcal{H}_{vib} = {}&{}^1t_{00}\ {}^{[1\,-1]}\mathcal{V}^{(0)}_{0A_1} + {}^2t_{00}\ {}^{[2\,-2]}\mathcal{V}^{(0)}_{0A_1}\\ &+ {}^2t_{20}\ {}^{[2\,-2]}\mathcal{V}^{(2)}_{0A_1} + {}^3t_{00}\ {}^{[3\,-3]}\mathcal{V}^{(0)}_{0A_1}\\ &+ {}^3t_{20}\ {}^{[3\,-3]}\mathcal{V}^{(2)}_{0A_1} + {}^3t_{36}\ {}^{[3\,-3]}\mathcal{V}^{(3)}_{6A_1}. \end{aligned} \tag{81}$$

Within the same order of approximation and with the same assumption of an isolated E normal mode we have (Eq. (53)) using the notations of [21]:

$$\begin{aligned}\mathcal{H}_{vib} = {}& x_1\left(\mathcal{A}^{+1(1,E)}\times\mathcal{A}^{1(1,E)}\right)^{(A_1)}\\ &+x_2\left(\mathcal{A}^{+2(0,A_1)}\times\mathcal{A}^{2(0,A_1)}\right)^{(A_1)}\\ &+x_3\left(\mathcal{A}^{+2(2,E)}\times\mathcal{A}^{2(2,E)}\right)^{(A_1)}\\ &+x_4\left(\mathcal{A}^{+3(1,E)}\times\mathcal{A}^{3(1,E)}\right)^{(A_1)}\\ &+x_5\left(\mathcal{A}^{+3(3,A_1)}\times\mathcal{A}^{3(3,A_1)}\right)^{(A_1)}\\ &+x_6\left(\mathcal{A}^{+3(3,A_2)}\times\mathcal{A}^{3(3,A_2)}\right)^{(A_1)}. \end{aligned} \tag{82}$$

where the couplings are performed in $G=C_{3v}$ or $G=T_d, O_h$. Both models, in equations (81) and (82) are strictly equivalent. However at this stage several remarks and comparisons are in order concerning both expansions of $\mathcal{H}_{vib}$.

- The full characterization of operators in (81) requires less labels than in (82) (five instead of seven).
- The computation of matrix elements of any vibrational operators ${}^{[m_1\,-m_2]}\mathcal{V}^{(j)}_{\ell\Gamma\sigma}$ is straightforward which is not the case of those in equation (53).
- Contrarily to those in (82) all operators in (81), except ${}^{[3\,-3]}\mathcal{V}^{(3)}_{6A_1}$, are diagonal in the initial basis.
- The advantages linked to a normal form expansion of Hamiltonian operators [6,21,22] are preserved by the 2*d gitos* due to the coupling scheme (14) adopted. As such, in both formalisms the number of possible operators, acting within a vibrational sub-space characterized by a fixed value of $n=v$, is limited.
- The physical meaning of the various operators in (81) is easier to find. So if one refers to the quantum numbers v and ℓ characterizing in first approximation the Hamiltonian eigenstates (or energy levels): the operator ${}^{[1\,-1]}\mathcal{V}^{(0)}_{0A_1}$ gives the harmonic structure; ${}^{[2\,-2]}\mathcal{V}^{(0)}_{0A_1}$ and ${}^{[3\,-3]}\mathcal{V}^{(0)}_{0A_1}$ represent anharmonicity corrections to the equidistant structure obtained in zeroth order; ${}^{[2\,-2]}\mathcal{V}^{(2)}_{0A_1}$ and ${}^{[3\,-3]}\mathcal{V}^{(2)}_{0A_1}$ lead to the ℓ sub-structure of energy levels (which depends upon v for the operator ${}^{[3\,-3]}\mathcal{V}^{(2)}_{0A_1}$); finally ${}^{[3\,-3]}\mathcal{V}^{(3)}_{6A_1}$ gives the splitting of the A_1 and A_2 sub-levels for $\ell=3p$. Such a discussion is impossible with the operators in equation (82). In Figure 1 we illustrate with an energy level diagram the effect of the successive addition in $\mathcal{H}_{vib}$ of the various operators ${}^{[m_1\,-m_1]}\mathcal{V}^{(j)}_{\ell A_1}$.

The preceding discussion is in fact not restricted to spherical or C_{3v} molecules. This can be easily seen from equations (78, 79) and Table 1.

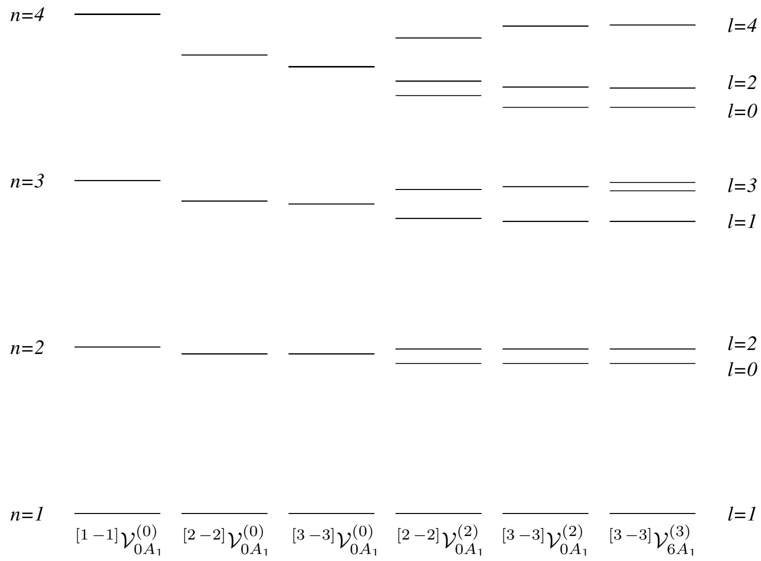

Fig. 1. Typical level scheme associated with $\mathcal{H}_{vib}$ (Eq. (81)).

4.2 The $\mathbf{U(p+1)}$ dynamical approach

The treatment of vibrational E modes presented in the preceding section implicitly assumes the possibility of an infinite number of energy levels: the n quantum number runs over all values from 0 up to ∞ and consequently there are also an infinite number of possible vibrational operators. In other words we deal with the noncompact $u(2,1)$ dynamical algebra of the two dimensional harmonic oscillator [2]. To overcome this somewhat unsatisfactory situation for real molecules, one may look for a compact dynamical algebra with an appropriate finite dimensional *irrep* which contains all the physical states. Various solutions have been proposed [26–29] more or less inspired by models of nuclear physics [30–32]. For our problem the $u(p+1)$ approach [8,29,33,34] is convenient especially in view of its application to linear $E \otimes \varepsilon$ Jahn-Teller systems and we show below that it is a natural extension of the $2d$ formalism. We assume that:

- we consider a molecule, with symmetry point group G (one of those introduced in previous sections), which admits an E-type mode in its full vibrational representation,
- within a $u(p+1)$ dynamical approach we have to introduce the non-invariance algebra $u(3)$ to which we associate a set of elementary boson operators $\{b_i^+, b_i\}_{i=1,2,3}$.

Thus we first deal with the chain:

$$u(3) \supset u(2) \supset su(2) \supset so(2). \tag{83}$$

The space of states is a carrier space for the so-called totally symmetric (or most degenerate) *irrep* $[N00] = [N\dot{0}]$ of $u(3)$ which subduces to $[n\,0]$ $(n = 0, 1, ...N)$ in $u(2)$ [15,17].

4.2.1 Tensors in the chain u(3) ⊃ u(2) ⊃ su(2) ⊃ so(2)

It is well-known that the quantities $E_{ij} = \{b_i^+ b_j\}$, with i and j running from 1 to 3, form a particular realization of the generators for the $u(3)$ algebra [2,16]. They satisfy:

$$[E_{ij}, E_{kl}] = \delta_{jk} E_{il} - \delta_{il} E_{kj}. \tag{84}$$

The E_{ij} generators with $i < j$, $i > j$, and $i = j$ are respectively raising, lowering and weight operators. The weight operators are diagonal in the $|n_1, n_2, n_3\rangle$ canonical basis of a three dimensional oscillator:

$$E_{ii}|n_1, n_2, n_3\rangle = N_i|n_1, n_2, n_3\rangle = n_i|n_1, n_2, n_3\rangle.$$

All operators which may act within an *irrep* $[N\,\dot{0}]$ have symmetry $[z\,0{-}z]$ with $z = 0, 1...N$. Now, it can be verified that all operators defined by

$$\mathrm{T}\begin{bmatrix} [z\ 0-z] \\ (max_c) \end{bmatrix} = \alpha_{z,N}\ b_1^{+z} b_3{}^z \tag{85}$$

are possible maximal components in $u(3)$ since their commutators with each E_{ij} raising generator is zero. So as to settle the value of the $\alpha_{z,N}$ coefficient we impose the normalization condition:

$$\langle n, 0, N-n|\ \mathrm{T}\begin{bmatrix} [z\ 0-z] \\ (max_c) \end{bmatrix}\ |0, 0, N\rangle = \delta_{z,n}. \tag{86}$$

The notation $|n_1, n_2, n_3\rangle$ for the states is that of the $u(3)$ canonical chain. $|0, 0, N\rangle$ and $|n, 0, N-n\rangle$ represent respectively the state with zero excitation quantum and the state with n excitation quanta maximal in $u(2)$. For condition (86) to be satisfied by operators in (85) we must have

$$\alpha_{z,N} = \left[\frac{(N-z)!}{N!\ z!}\right]^{1/2}. \tag{87}$$

Operators which are semi-maximal in $u(3)$ are deduced from those in equation (85) with appropriate lowering operators [35–37] and we obtain:

$$\mathrm{T}\begin{bmatrix} z & 0 & -z \\ & m_1 \;\; -m_2 & \\ & m_1 & \end{bmatrix} = \mathcal{G}(z,m_1,m_2)\; b_1^{+m_1}\; b_2{}^{m_2}, \quad (88)$$

where $\mathcal{G}(z,m_1,m_2)$ is an operator function invariant in $u(2)$

$$\mathcal{G}(z,m_1,m_2) = \left[\binom{z}{m_1}\binom{z}{m_2}\frac{(z+m_2+1)!(z+m_1+1)!(N-z)!}{(m_1+m_2+1)!(2z+1)!N!z!}\right]^{1/2} \times\left\{\sum_{t=0}^{u}(-1)^{t+m_2}\binom{z-m_1}{t}\frac{(m_1+m_2+1)!(z-m_2)!}{(m_1+m_2+1+t)!(z-m_2-t)!} \times (N_1+N_2-m_1)^{[t]}(N_3-z+m_1+u)^{[u-t]}\right\} \times b_3^{+z-m_1-u}\; b_3{}^{z-m_2-u}. \quad (89)$$

with $u = \inf(z-m_1, z-m_2)$. Now from the results of section 2 we have:

$$b_1^{+m_1}\; b_2{}^{m_2} = \mathcal{N}(m_1,m_2)\; {}^{[m_1\; -m_2]}T^{(j)}_{-j},$$

thus the operator in the left member of equation (88) is, within a phase factor, the minimal covariant component of an irreducible tensor in the $su(2) \supset so(2)$ chain. Consequently all necessary tensors oriented in $u(3) \supset u(2) \supset su(2) \supset so(2)$ are obtained if we define their minimal covariant component by:

$$\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & -j & \end{bmatrix} = e^{i\Psi(z,m_1,m_2)}\mathrm{T}\begin{bmatrix} z & 0 & -z \\ & m_1 \;\; -m_2 & \\ & m_1 & \end{bmatrix}.$$

Using the fact that the $su(2)$ generators (1) commute with $\mathcal{G}(z,m_1,m_2)$ it is easily checked that an arbitrary covariant tensor is obtained with:

$$\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & m & \end{bmatrix} = \frac{1}{\mathcal{N}(m_1,m_2)}\; e^{i\Psi(z,m_1,m_2)} \times \mathcal{G}(z,m_1,m_2)\; {}^{[m_1\; -m_2]}T^{(j)}_{m} \quad (90)$$

where the coefficients $\mathcal{N}(m_1,m_2)$ are those in equation (21). We recall the range of variation of each label specifying the operator in (90):

$$z = 0,1,...,N$$
$$m_1 = 0,1,...,z; \qquad m_2 = 0,1,...,z$$
$$j = \frac{m_1+m_2}{2}; \qquad m = -j,-j+1...,j-1,j. \quad (91)$$

The arbitrariness in the phase factor $e^{i\Psi(z,m_1,m_2)}$ can be raised by imposing that the $u(3)$ symmetry adapted tensors satisfy the same properties upon adjunction and time reversal than the standard $2d$ tensors:

$$\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & m & \end{bmatrix}^{\dagger} = (-1)^{j+m}\; i^{m_1-m_2} \times \mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_2 & -m_1] & (j) \\ & -m & \end{bmatrix} \quad (92)$$

$$\mathcal{K}_t\;\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & m & \end{bmatrix}\mathcal{K}_t^{-1} = \mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & -m & \end{bmatrix}. \quad (93)$$

If we set $\mathcal{K}_t\; b_3^+\;\; \mathcal{K}_t{}^{-1} = b_3^+$ it may be shown that an appropriate choice for $e^{i\Psi(z,m_1,m_2)}$ so that relations (92, 93) hold is:

$$e^{i\Psi(z,m_1,m_2)} = i^{m_1}. \quad (94)$$

4.2.2 Matrix elements

We denote $|[N\,\dot{0}][n\,0]jm\rangle\rangle$ the states associated with the *irrep* $[N\,\dot{0}]$ and symmetrized in $u(3) \supset u(2) \supset su(2) \supset so(2)$. They may be obtained from the zero quantum excitation state $|[N\,\dot{0}][0\,0]00\rangle\rangle \equiv |0,0,N\rangle$ with

$$|[N\,\dot{0}][n\,0]jm\rangle\rangle = \mathrm{T}\begin{bmatrix} n & 0 & -n \\ [n & 0] & (j) \\ & m & \end{bmatrix}\; |[N\,\dot{0}][0\,0]00\rangle\rangle. \quad (95)$$

Matrix elements of operators (90) within the basis (95) are given by an expression similar to (18):

$$\langle\langle [N\,\dot{0}]\,[n''\,0]j''m''|\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & m & \end{bmatrix}|[N\,\dot{0}]\,[n'\,0]j'm'\rangle\rangle = (2j''+1)^{-1/2} C^{\,m\;m'\;(j'')*}_{\,(j\;j')\;\;m''} \times\left([N\dot{0}]\,[n''\,0]j''||\mathrm{T}\begin{bmatrix} z\;0\;-z \\ [m_1\;-m_2]\;(j)\end{bmatrix}||[N\,\dot{0}]\,[n'\,0]j'\right). \quad (96)$$

The reduced matrix element in (96) is obtained once the contribution of the part $\mathcal{G}(z,m_1,m_2)$ in (90) is known. This is determined from the expanded expression (89). A rather straightforward computation leads to:

$$([N\,\dot{0}]\,[n''\,0]j''||\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 - m_2] & (j) \end{bmatrix}||[N\,\dot{0}]\,[n'\,0]j') = \left[\binom{z}{m_1}\binom{z}{m_2}\frac{(z+m_2+1)!(z+m_1+1)!(N-z)!}{(m_1+m_2+1)!(2z+1)!N!z!} \times\frac{m_1!m_2!(N-n')!(N-n'+m_2-m_1)!}{(N-n'-z+m_2+u)!(N-n'-z+m_2+u)!}\right]^{1/2} \times\left\{\sum_t(-1)^t\binom{z-m_1}{t}\frac{(m_1+m_2+1)!(z-m_2)!(n'-m_2)!}{(m_1+m_2+1+t)!(z-m_2-t)!} \times\frac{(N-n'+m_2-z+u)!}{(n'-m_2-t)!(N-n'+m_2-z+t)!}\right\} \times ([n''\,0]j''\;||{}^{[m_1\;-m_2]}T^{(j)}||\;[n'\,0]j') \quad (97)$$

where the reduced matrix element of the standard $2d$ operator is given by (19) with $m'_1 = m_1$, $m'_2 = m_2$.

4.2.3 Symmetry adaptation in a point group G

The behavior of operators (90) under an operation R of G is determined by that of the elementary bosons b_i^+ and b_i $(i = 1, 2, 3)$. In the following we keep first the assumptions of Section 3.2, in particular equation (38), for the boson operators with indices $i = 1, 2$ which span an *irrep* of type E of G. For a correct description, in terms of allowed symmetries in G, of the states within the representation $[N\dot{0}]$ in $u(3)$ we must impose that b_3^+, b_3 belong to the scalar representation of G. As a consequence the term $\mathcal{G}(z, m_1, m_2)$ given by (89) is invariant in G and the operators (90) transform under the action of the generators of G as the $2d$ standard operators. This property allows the symmetry adaptation in G of both kinds of tensors with the same orientation matrix. The $u(3)$ symmetry adapted tensors are thus given by

$$\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & \ell\Gamma\sigma & \end{bmatrix} = {}^{[m_1\,-m_2]}G^{m}_{\ell\Gamma\sigma}\,\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & m & \end{bmatrix}$$

where the ${}^{[m_1\,-m_2]}G$ matrix is that defined in Section 3.2.1. These new operators satisfy properties similar to those of operators ${}^{[m_1\,-m_2]}T^{(j)}_{\ell\Gamma\sigma}$. In particular it is easily checked with equations (92, 93) and the properties (43, 44) of the matrix ${}^{[m_1\,-m_2]}G$ that:

$$\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & \ell\Gamma\sigma & \end{bmatrix}^{\dagger} = \mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_2 & -m_1] & (j) \\ & \ell\Gamma\sigma & \end{bmatrix}$$

$$\mathcal{K}_t\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & \ell\Gamma\sigma & \end{bmatrix}\mathcal{K}_t^{-1} = (-1)^{j-\frac{\ell}{2}} \times \mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & \ell\Gamma\sigma & \end{bmatrix}. \quad (98)$$

Finally within the symmetry adapted states given by:

$$|[N\dot{0}][n\,0]j\ell\Gamma\sigma\rangle\rangle = \mathrm{T}\begin{bmatrix} n & 0 & -n \\ & [n\,0] & (j) \\ & \ell\Gamma\sigma & \end{bmatrix}|[N\dot{0}][0\,0]00\rangle\rangle, \quad (99)$$

the matrix elements are obtained with a relation similar to (47):

$$\langle\langle\Psi(\kappa'')|\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \\ & \ell\Gamma\sigma & \end{bmatrix}|\Psi(\kappa')\rangle\rangle =$$

$$(2j''+1)^{-1/2}F\begin{matrix} \ell\Gamma\sigma & \ell'\Gamma'\sigma' & ([n''\,0]j'')* \\ ([m_1\;-m_2]j & [n'\,0]j') & \ell''\Gamma''\sigma'' \end{matrix} \times\left([N\dot{0}]\,[n''\,0]j''|||\mathrm{T}\begin{bmatrix} z & 0 & -z \\ [m_1 & -m_2] & (j) \end{bmatrix}|||[N\,\dot{0}]\,[n'\,0]j'\right) \quad (100)$$

where we set $\kappa = [N\,\dot{0}][n\,0]j\ell\Gamma\sigma$. The reduced matrix elements are those in (97) and the symmetry adapted Clebsch-Gordan symbols those defined in (48) with the ${}^{[m_1\,-m_2]}G$ matrix appropriate for the G group and E mode considered. Obviously the results in this section can be transposed if the orientation for the *irreps* of G are those of Section 3.3. One only needs to make the substitution $\sigma \to \bar{\sigma}$ everywhere except in the right member of equation (98) where it is $\sigma \to -\bar{\sigma}$.

4.3 Electronic states and operators for an orbital doublet

For an orbital doublet or E term it is well-known that an $u(2)$ algebra is appropriate [9,38]. We shall denote E_r the *irrep* of G spanned by the electronic states (if needed the additional index $\alpha = {}'$ or ${}''$, u, g can be added in a trivial manner). So, from the results of the previous sections the electronic space of states is a carrier space for the *irrep* [10] of $u_e(2)$ which subduces to E_r in G with possible bases:

$$\begin{aligned} &|[10]\tfrac{1}{2}m\rangle\rangle \;\; u_e(2) \supset su_e(2) \supset so_e(2) \quad &(a)\\ &|[10]\tfrac{1}{2}E_r\,\sigma\rangle\rangle \;\; u_e(2) \supset su_e^*(2) \supset G \quad &(b)\\ &|[10]\tfrac{1}{2}E_r\,\bar{\sigma}\rangle\rangle \;\; u_e(2) \supset su_e^*(2) \supset G \quad &(c) \end{aligned} \quad (101)$$

where we indicate on the right the algebraic chain and where in case (b) (resp. (c)) the matrices for the *irrep* of type E are in real form (resp. complex). Also we recall that in fact basis (a) and (c) are identical.

A complete set of electronic operators is given by:

$${}^{[1\,-1]}\mathcal{E}^{(k)}_{p_e}\begin{cases} p_e = m & \text{case (a)} \\ p_e = \ell\,\Gamma\sigma & \text{case (b)} \\ p_e = \ell\,\Gamma\bar{\sigma} & \text{case (c)} \end{cases}. \quad (102)$$

Actually as ${}^{[1\,-1]}\mathcal{E}^{(0)}$ reduces to the $u(2)$ linear invariant $(N_1+N_2)/\sqrt{2}$ which is a constant within $[1\,0]$ we are left with ${}^{[1\,-1]}\mathcal{E}^{(1)} = {}^{[1\,-1]}E^{(1)}$. We give in Table 2 the expressions of the symmetry adapted electronic operators in terms of the $su_e(2)$ generators[3]. In this table the various subcases correspond to different symmetries of the electronic operators according to the rules given after equation (51):

(i) $r = n/4$ (or $r = n/2$);
(ii) $r' = 2r$;
(iii) $r' = n - 2r$ (or $r' = 2n - 2r$)

where the cases in parenthesis refer to D_{nd} (n even) groups. From these expressions it is easily checked that the

[3] For practical purposes the generators have been renormalized by a factor $\sqrt{2}$.

Table 2. Symmetry adapted electronic operators ${}^{[1\,-1]}E^{(1)}$.

(a)	${}^{[1\,-1]}E^{(1)}_{0} = -iS_z$	${}^{[1\,-1]}E^{(1)}_{1} = iS_-/\sqrt{2}$	${}^{[1\,-1]}E^{(1)}_{-1} = -iS_+/\sqrt{2}$
(b, c)(i)	${}^{[1\,-1]}E^{(1)}_{0A_2} = -S_z$	${}^{[1\,-1]}E^{(1)}_{2B_1} = S_x$	${}^{[1\,-1]}E^{(1)}_{2B_2} = S_y$
(b)(ii)	${}^{[1\,-1]}E^{(1)}_{0A_2} = -S_z$	${}^{[1\,-1]}E^{(1)}_{2E_{r'}1} = S_x$	${}^{[1\,-1]}E^{(1)}_{2E_{r'}2} = -S_y$
(b)(iii)	${}^{[1\,-1]}E^{(1)}_{0A_2} = -S_z$	${}^{[1\,-1]}E^{(1)}_{2E_{r'}1} = S_x$	${}^{[1\,-1]}V^{(1)}_{2E_{r'}2} = S_y$
(c)(ii)	${}^{[1\,-1]}E^{(1)}_{0A_2} = -S_z$	${}^{[1\,-1]}V^{(1)}_{2E_{r'}\bar{1}} = iS_-/\sqrt{2}$	${}^{[1\,-1]}V^{(1)}_{2E_{r'}\bar{2}} = -iS_+/\sqrt{2}$
(c)(iii)	${}^{[1\,-1]}E^{(1)}_{0A_2} = -S_z$	${}^{[1\,-1]}V^{(1)}_{2E_{r'}\bar{1}} = iS_+/\sqrt{2}$	${}^{[1\,-1]}V^{(1)}_{2E_{r'}\bar{2}} = -iS_-/\sqrt{2}$

symmetry adapted electronic operators satisfy the commutation rules of a "pseudo-spin" [9]:

$$\begin{aligned}\left[{}^{[1\,-1]}E^{(1)}_{0A_2},\ {}^{[1\,-1]}E^{(1)}_{2B_1}\right] &= -i\ {}^{[1\,-1]}E^{(1)}_{2B_2}\\ \left[{}^{[1\,-1]}E^{(1)}_{2B_1},\ {}^{[1\,-1]}E^{(1)}_{2B_2}\right] &= -i\ {}^{[1\,-1]}E^{(1)}_{0A_2}\\ \left[{}^{[1\,-1]}E^{(1)}_{2B_2},\ {}^{[1\,-1]}E^{(1)}_{0A_2}\right] &= -i\ {}^{[1\,-1]}E^{(1)}_{2B_1}\end{aligned} \tag{103}$$

for case (i) and

$$\begin{aligned}\left[{}^{[1\,-1]}E^{(1)}_{0A_2},\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}1}\right] &= i\eta\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}2}\\ \left[{}^{[1\,-1]}E^{(1)}_{2E_{r'}1},\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}2}\right] &= i\eta\ {}^{[1\,-1]}E^{(1)}_{0A_2}\\ \left[{}^{[1\,-1]}E^{(1)}_{2E_{r'}2},\ {}^{[1\,-1]}E^{(1)}_{0A_2}\right] &= i\eta\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}1}\end{aligned} \tag{104}$$

$$\begin{aligned}\left[{}^{[1\,-1]}E^{(1)}_{0A_2},\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}\bar{1}}\right] &= \eta\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}\bar{1}}\\ \left[{}^{[1\,-1]}E^{(1)}_{2E_{r'}\bar{1}},\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}\bar{2}}\right] &= \eta\ {}^{[1\,-1]}E^{(1)}_{0A_2}\\ \left[{}^{[1\,-1]}E^{(1)}_{2E_{r'}\bar{2}},\ {}^{[1\,-1]}E^{(1)}_{0A_2}\right] &= \eta\ {}^{[1\,-1]}E^{(1)}_{2E_{r'}\bar{2}}\end{aligned} \tag{105}$$

where $\eta = +1(-1)$ for cases (ii) and (iii).

4.3.1 Matrix elements of electronic operators

To facilitate comparisons with previous studies we give below the matrices of electronic operators, computed with the relations (18) for case (a), (47) for cases (b) and (c) with the substitution $\sigma \to \bar{\sigma}$ in this latter case. In all three cases we have $[m_1' - m_2']j = [0\,-0]0$ or $[m_1' - m_2']j = [1\,-1]1$ and

$$\begin{aligned}&\left([1\,0]\tfrac{1}{2}||{}^{[1\,-1]}\mathcal{E}^{(0)}||[1\,0]\tfrac{1}{2}\right) = \sqrt{2}\\ &\left([1\,0]\tfrac{1}{2}||{}^{[1\,-1]}E^{(1)}||[1\,0]\tfrac{1}{2}\right) = -i\sqrt{\tfrac{3}{2}}.\end{aligned}$$

To underline the differences between operators and their matrix representative we add a "∧" and all matrices are expressed in terms of the usual Pauli matrices:

$$\hat{\sigma}_z = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix},\quad \hat{\sigma}_x = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix},\quad \hat{\sigma}_y = \begin{pmatrix}0 & -i\\ i & 0\end{pmatrix} \tag{106}$$

or with $\hat{\sigma}_\pm = \hat{\sigma}_x \pm i\hat{\sigma}_y$; $\hat{\sigma}_0$ is the two dimensional identity matrix. In all cases we have:

$${}^{[1\,-1]}\hat{\mathcal{E}}^{(0)}_{0} = \frac{1}{\sqrt{2}}\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix} = \frac{1}{\sqrt{2}}\,\hat{\sigma}_0.$$

The $u_e(2) \supset su_e(2) \supset so_e(2)$ orientation

$$\begin{aligned}&{}^{[1\,-1]}\hat{E}^{(1)}_{1} = -\frac{i}{\sqrt{2}}\,\hat{\sigma}_+,\qquad {}^{[1\,-1]}\hat{E}^{(1)}_{-1} = \frac{i}{\sqrt{2}}\,\hat{\sigma}_-\\ &{}^{[1\,-1]}\hat{E}^{(1)}_{0} = \frac{i}{2}\,\hat{\sigma}_z.\end{aligned} \tag{107}$$

As it is normal the z component of the pseudo-spin S is diagonal but electronic states are not symmetrized in G.

The $u_e(2) \supset su_e^*(2) \supset G$ chain with orientation I

$$\begin{aligned}&{}^{[1\,-1]}\hat{E}^{(1)}_{2E_{r'}1} = \frac{1}{2}\,\hat{\sigma}_z,\qquad {}^{[1\,-1]}\hat{E}^{(1)}_{2E_{r'}2} = \frac{\eta}{2}\,\hat{\sigma}_x\\ &{}^{[1\,-1]}\hat{E}^{(1)}_{0A_2} = \frac{1}{2}\,\hat{\sigma}_y.\end{aligned} \tag{108}$$

with still $\eta = 1$ ($\eta = -1$) for case (ii) and (iii). The usual orientation for E symmetry types gives symmetry adapted states but the z component of the pseudo-spin S is no longer diagonal.

For the special case (i) the $E_{r'}$ generators are replaced by:

$${}^{[1\,-1]}\hat{E}^{(1)}_{2B_1} = \frac{1}{2}\,\hat{\sigma}_z,\qquad {}^{[1\,-1]}\hat{E}^{(1)}_{2B_2} = -\frac{1}{2}\,\hat{\sigma}_x. \tag{109}$$

In view of applications to $E \otimes (b_1 + b_2 + ...)$ cases [9,39] it appears immediately from (109) that it must be possible to define a new orientation for this case in which the generator ${}^{[1\,-1]}E^{(1)}_{2B_2}$ would be diagonal instead of the ${}^{[1\,-1]}E^{(1)}_{2B_1}$ one. The corresponding transformation is easily shown to be of the form:

$$\begin{aligned}T^{(E_r)}_{\bar{\bar{1}}} &= \frac{e^{i\theta}}{\sqrt{2}}\left(T^{(E_r)}_{\bar{1}} + iT^{(E_r)}_{\bar{2}}\right)\\ &= \frac{e^{i\theta}e^{i\pi/4}}{\sqrt{2}}\left(T^{(E_r)}_{1} - T^{(E_r)}_{2}\right)\\ T^{(E_r)}_{\bar{\bar{2}}} &= \frac{e^{i\mu}}{\sqrt{2}}\left(T^{(E_r)}_{\bar{1}} - iT^{(E_r)}_{\bar{2}}\right)\\ &= -\frac{e^{i\mu}e^{-i\pi/4}}{\sqrt{2}}\left(T^{(E_r)}_{1} + T^{(E_r)}_{2}\right).\end{aligned} \tag{110}$$

The choice $e^{i(\theta-\mu)} = i$ keeps real matrices with $D^{(E_r)}(X)$ identical to that in (34) and:

$$D^{(E_r)}(Y) = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{111}$$

The choice $e^{i\theta} = e^{-i\pi/4}$ leads to a real change of basis from orientation I:

$$T^{(E_r)}_{\bar{\bar{1}}} = \frac{1}{\sqrt{2}}\left(T^{(E_r)}_1 - T^{(E_r)}_2\right)$$
$$T^{(E_r)}_{\bar{\bar{2}}} = \frac{1}{\sqrt{2}}\left(T^{(E_r)}_1 + T^{(E_r)}_2\right). \tag{112}$$

Straightforward computations lead to a new $\bar{\bar{G}}$ matrix:

$${}^{[m_1\,-m_2]}\mathcal{T}^{(j)}_{\ell\Gamma\bar{\bar{\sigma}}} = {}^{[m_1\,-m_2]}\bar{\bar{G}}^m_{\ell\Gamma\bar{\bar{\sigma}}}\;{}^{[m_1\,-m_2]}\mathcal{T}^{(j)}_m \tag{113}$$

with elements:

$$\begin{array}{ll} \bar{\bar{\sigma}} & {}^{[m_1\,-m_2]}\bar{\bar{G}}^m_{\ell E_{r'}\bar{\bar{\sigma}}} \\ \bar{\bar{1}} \rightarrow & \frac{1}{\sqrt{2}}\left({}^{[m_1\,-m_2]}G^m_{\ell E_{r'}1} - {}^{[m_1\,-m_2]}G^m_{\ell E_{r'}2}\right) \\ \bar{\bar{2}} \rightarrow & \frac{1}{\sqrt{2}}\left({}^{[m_1\,-m_2]}G^m_{\ell E_{r'}1} - {}^{[m_1\,-m_2]}G^m_{\ell E_{r'}2}\right) \end{array} \tag{114}$$

and ${}^{[m_1\,-m_2]}\bar{\bar{G}} = {}^{[m_1\,-m_2]}G$ when Γ is of type A or B. The properties of these new coefficients are easily deduced from those given in (43, 44); likewise matrix elements of the new symmetry adapted tensors (113) are given by equation (47) with the substitution $\sigma \rightarrow \bar{\bar{\sigma}}$. In particular for the operators (109) we have now:

$${}^{[1\,-1]}\hat{E}^{(1)}_{2B_1} = \frac{1}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} = \frac{1}{2}\,\hat{\sigma}_x$$
$${}^{[1\,-1]}\hat{E}^{(1)}_{2B_2} = \frac{1}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} = \frac{1}{2}\,\hat{\sigma}_z. \tag{115}$$

Although the change of orientation can be made for any E type representation, it is mostly useful for $r = n/4$ ($r = n/2$) for D_n, C_{nv} groups with $n = 4p$ (D_{nd} (n even)) and similar cases for groups in $G_{(II)}$. In the following this new orientation will be referred to as orientation III.

The $u_e(2) \supset su^*_e(2) \supset G$ chain with orientation II

Case (i):

$${}^{[1\,-1]}\hat{E}^{(1)}_{2B_1} = -\frac{1}{2}\,\hat{\sigma}_x, \quad {}^{[1\,-1]}\hat{E}^{(1)}_{2B_2} = \frac{1}{2}\,\hat{\sigma}_y$$
$${}^{[1\,-1]}\hat{E}^{(1)}_{0A_2} = \frac{1}{2}\,\hat{\sigma}_z. \tag{116}$$

Case (ii):

$${}^{[1\,-1]}\hat{E}^{(1)}_{2E_{r'}\bar{1}} = -\frac{i}{\sqrt{2}}\,\hat{\sigma}_+, \quad {}^{[1\,-1]}\hat{E}^{(1)}_{2E_{r'}\bar{2}} = \frac{i}{\sqrt{2}}\,\hat{\sigma}_-$$
$${}^{[1\,-1]}\hat{E}^{(1)}_{0A_2} = \frac{1}{2}\,\hat{\sigma}_z. \tag{117}$$

Case (iii):

$${}^{[1\,-1]}\hat{E}^{(1)}_{2E_{r'}\bar{1}} = -\frac{i}{\sqrt{2}}\,\hat{\sigma}_-, \quad {}^{[1\,-1]}\hat{E}^{(1)}_{2E_{r'}\bar{2}} = \frac{i}{\sqrt{2}}\,\hat{\sigma}_+$$
$${}^{[1\,-1]}\hat{E}^{(1)}_{0A_2} = \frac{1}{2}\,\hat{\sigma}_z. \tag{118}$$

This orientation gives, in all cases, symmetry adapted states and the z-component of the pseudo-spin S is diagonal.

5 Conclusion

From a two boson realization of the $u(2)$ algebra, and assuming that these two bosons span an arbitrary two dimensional *irrep* of type E of a point group G, we showed that complete sets of states and operators symmetrized in the whole $u(2) \supset su^*(2) \supset G$ chain can be built. Several orientations for the matrices of E type *irrep* have been introduced whose relevance depends upon the problem at hand. Various physical interpretations of the corresponding degrees of freedom may be made. Examples related to vibrational and electronic spectroscopy problems have been presented. It would also be possible, with very few modifications, to use our results for rotational degrees of freedom in symmetric top molecules. This can be seen from the fact that the $su(2)$ and $so(3)$ algebras are isomorphic and that the $SO(3) \downarrow G$ symmetry subduction for the J_α elementary rotational operators is of type $A_2 + E$ for all groups considered here, except for cubic point groups associated with spherical tops. More involved applications are currently under development for the treatment of vibronic interactions in $E \otimes (b_1 + b_2 ...)$, $E \otimes \varepsilon$ and $G' \otimes \varepsilon$ Jahn-Teller systems.

Appendix A: Generators X, Y, Z

The various point groups admitting integer *irrep* of type E are separated into three categories:

- groups in $G_{(I)}$ are finite subgroups of $O(3)$ of rank two. We have thus the groups D_n, C_{nv}, D_{nd} (n even), O and T_d;
- groups in $G_{(II)}$ are direct product groups of an element in $G_{(I)}$ with C_s or C_I;
- groups in $G_{(III)}$ are $C_{\infty v}$ and $D_{\infty h}$.

The chosen elements X, Y, Z, the appropriate angle ψ, the possible r and α values are given for all groups in Tables 3, 4 and 5.

Appendix B: Symmetries of u(2) ⊃ su(2)* ⊃ G tensors for common point groups

For the various groups in $G_{(I)}$, $G_{(II)}$ the results have been established for arbitrary values of the principal axis order n. In order to facilitate applications we give below,

Table 3. Conventions for groups in $G_{(I)}$.

G	X	Y	r	ψ
D_n	C_n^z	$C_2'(Ox)$	$1...\frac{n-1}{2}$ n odd $1...\frac{n}{2}-1$ n even	$\frac{2\pi}{n}$
C_{nv}	C_n^z	$\sigma_v(Ox)$	$1...\frac{n-1}{2}$ n odd $1...\frac{n}{2}-1$ n even	$\frac{2\pi}{n}$
D_{nd} (n even)	S_{2n}^z	$C_2'(Ox)$	$1...n-1$	$\frac{\pi}{n}$
O	$C_{3(1,1,1)}$	C_4^z	1	$\frac{2\pi}{3}$
T_d	$C_{3(1,1,1)}$	$S_4^{z\,3}$	1	$\frac{2\pi}{3}$

Table 4. Conventions for groups in $G_{(II)}$.

G	Z	α	sign (Eq. (35))
$D_{nh} = D_n \times C_s$ (n odd)	σ_h	$'$ $''$	$+$ $-$
$D_{nh} = D_n \times C_i$ (n even)	I	g u	$+$ $-$
$D_{nd} = D_n \times C_i$ (n odd)	I	g u	$+$ $-$
$O_h = O \times C_i$	I	g u	$+$ $-$

Table 5. Conventions for groups in $G_{(III)}$.

G	X	Y	Z	r	α	sign (Eq. (35))
$C_{\infty v}$	C_ψ^z	$\sigma_v(Ox)$		$1, 2, ..., \infty$		
$D_{\infty h} = C_{\infty v} \times C_i$	C_ψ^z	$\sigma_v(Ox)$	I	$1, 2, ..., \infty$	g u	$+$ $-$

as a function of ℓ, the symmetries of operators or states one may build for common point groups. We use the notations of equations (39–42) for irreducible tensors built from quantities with symmetry E_r (or E_{r_α}). In applications to vibrational studies the substitution $\mathcal{T} \rightarrow \mathcal{V}$ is made; for electronic tensors and operators we use $\mathcal{T} \rightarrow \mathcal{E}$.

As whenever $\ell\Gamma$ is of type $A_1 + A_2$ (resp. $B_1 + B_2$) we have $\mu(\ell) = -i$ (resp. $\mu(\ell) = +i$), the phase $\mu(\ell)$ is specified below only for E type *irreps*.

• For all groups the $\ell = 0$ operators ${}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,0\Gamma)}$ are characterized by:

$$\begin{array}{ll} E_r(E_{r_\alpha}) & \Gamma\ (condition) \\ E_r & A_1\ (m_1\ \text{even}) A_2\ (m_1\ \text{odd}) \\ E_{r\alpha}(\alpha = {}'\ or\ {}'') & A_1'\ (m_1\ \text{even}) A_2'\ (m_1\ \text{odd}) \\ E_{r\alpha}(\alpha = g\ or\ u) & A_{1g}\ (m_1\ \text{even}) A_{2g}\ (m_1\ \text{odd}) \end{array} \quad \text{(B.1)}$$

with $\mu(0) = 1$ ($\mu(0) = -i$) when $\Gamma = A_1$ ($\Gamma = A_2$).

• $G = C_{3v}, D_3, O$ and T_d

$$\begin{array}{ll} {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,3pA_i)} & i = 1, 2 \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E)} & \ell = 3p+1\ (\mu(\ell) = -i) \\ & \ell = 3p+2\ (\mu(\ell) = i). \end{array} \quad \text{(B.2)}$$

• $G = D_{3d}, O_h, D_{3h}$
Operators ${}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell\Gamma_\tau)}$ have the same labels as in equation (B.2) with:

$$\begin{array}{llll} D_{3d},\ O_h & \text{for } E_\alpha = E_g & \tau = g & \\ & \text{for } E_\alpha = E_u & \tau = g & m_1 + m_2\ \text{even} \\ & & \tau = u & m_1 + m_2\ \text{odd} \\ D_{3h} & \text{for } E_\alpha = E' & \tau = {}' & \\ & \text{for } E_\alpha = E'' & \tau = {}' & m_1 + m_2\ \text{even} \\ & & \tau = {}'' & m_1 + m_2\ \text{odd}. \end{array} \quad \text{(B.3)}$$

• $G = D_{2d}, C_{4v}$ and D_4

$$\begin{array}{ll} {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,4pA_i)} & i = 1, 2 \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,4p+2B_i)} & i = 1, 2 \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E)} & \ell = 4p+1\ \ (\mu(\ell) = -i) \\ & \ell = 4p+3\ \ (\mu(\ell) = i). \end{array} \quad \text{(B.4)}$$

• $G = D_{4h}$
Operators ${}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell\Gamma_\tau)}$ have the same labels as in equation (B.4) with:

$$\begin{array}{l} \text{for } E_\alpha = E_g \tau = g \\ \text{for } E_\alpha = E_u \tau = g\ m_1 + m_2\ \text{even} \\ \qquad\qquad\quad \tau = u\ m_1 + m_2\ \text{odd}. \end{array} \quad \text{(B.5)}$$

• $G = D_{4d}$
$E_r = E_1$:

$$\begin{array}{ll} {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,8pA_i)} & i = 1, 2 \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,8p+4B_i)} & i = 1, 2 \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_1)} & \ell = 8p+1\ \ (\mu(\ell) = -i) \\ & \ell = 8p+7\ \ (\mu(\ell) = i) \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 8p+2\ \ (\mu(\ell) = -i) \\ & \ell = 8p+6\ \ (\mu(\ell) = i) \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_3)} & \ell = 8p+3\ \ (\mu(\ell) = -i) \\ & \ell = 8p+5\ \ (\mu(\ell) = i) \end{array} \quad \text{(B.6)}$$

$E_r = E_2$:

$$\begin{array}{ll} {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,4pA_i)} & i = 1, 2 \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,4p+2B_i)} & i = 1, 2 \\ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 4p+1\ \ (\mu(\ell) = -i) \\ & \ell = 4p+3\ \ (\mu(\ell) = i) \end{array} \quad \text{(B.7)}$$

$E_r = E_3$:

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,8pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,8p+4B_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_1)} & \ell = 8p+3 & (\mu(\ell) = -i) \\
 & \ell = 8p+5 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 8p+2 & (\mu(\ell) = i) \\
 & \ell = 8p+6 & (\mu(\ell) = -i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_3)} & \ell = 8p+1 & (\mu(\ell) = -i) \\
 & \ell = 8p+7 & (\mu(\ell) = i).
\end{array}
\quad (B.8)
$$

• $G = C_{5v}, D_5$

$E_r = E_1$:

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,5pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_1)} & \ell = 5p+1 & (\mu(\ell) = -i) \\
 & \ell = 5p+4 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 5p+2 & (\mu(\ell) = -i) \\
 & \ell = 5p+3 & (\mu(\ell) = i)
\end{array}
\quad (B.9)
$$

$E_r = E_2$:

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,5pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_1)} & \ell = 5p+2 & (\mu(\ell) = i) \\
 & \ell = 5p+3 & (\mu(\ell) = -i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 5p+1 & (\mu(\ell) = -i) \\
 & \ell = 5p+4 & (\mu(\ell) = i).
\end{array}
\quad (B.10)
$$

• $G = D_{5d}, D_{5h}$

Operators ${}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell\Gamma_\tau)}$ have the same labels as in equation (B.10) with:

$$
\begin{array}{llll}
D_{5d} & E_{r_\alpha} = E_{r_g} & \tau = g & \\
 & E_{r_\alpha} = E_{r_u} & \tau = g & m_1 + m_2 \text{ even} \\
 & & \tau = u & m_1 + m_2 \text{ odd} \\
D_{5h} & E_{r_\alpha} = E'_r & \tau = {}' & \\
 & E_{r_\alpha} = E''_r & \tau = {}' & m_1 + m_2 \text{ even} \\
 & & \tau = {}'' & m_1 + m_2 \text{ odd}.
\end{array}
\quad (B.11)
$$

• $G = C_{6v}, D_6$

$E_r = E_1$

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,6pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,6p+3B_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_1)} & \ell = 6p+1 & (\mu(\ell) = -i) \\
 & \ell = 6p+5 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 6p+2 & (\mu(\ell) = -i) \\
 & \ell = 6p+4 & (\mu(\ell) = i)
\end{array}
\quad (B.12)
$$

$E_r = E_2$

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,3pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 3p+1 & (\mu(\ell) = -i) \\
 & \ell = 3p+2 & (\mu(\ell) = i).
\end{array}
\quad (B.13)
$$

• $G = D_{6h}$

Operators ${}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell\Gamma_\tau)}$ have the same labels as in equation (B.13) with:

$$
\begin{array}{lll}
E_{r_\alpha} = E_{rg} & \tau = g & \\
E_{r_\alpha} = E_{ru} & \tau = g & m_1 + m_2 \text{ even.} \\
 & \tau = u & m_1 + m_2 \text{ odd.}
\end{array}
\quad (B.14)
$$

• $G = D_{6d}$

$E_r = E_1$

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,12pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,12p+6B_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_1)} & \ell = 12p+1 & (\mu(\ell) = -i) \\
 & \ell = 12p+11 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 12p+2 & (\mu(\ell) = -i) \\
 & \ell = 12p+10 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_3)} & \ell = 12p+3 & (\mu(\ell) = -i) \\
 & \ell = 12p+9 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_4)} & \ell = 12p+4 & (\mu(\ell) = -i) \\
 & \ell = 12p+8 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_5)} & \ell = 12p+5 & (\mu(\ell) = -i) \\
 & \ell = 12p+7 & (\mu(\ell) = i)
\end{array}
\quad (B.15)
$$

$E_r = E_2$:

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,6pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,6p+3B_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} & \ell = 6p+1 & (\mu(\ell) = -i) \\
 & \ell = 6p+5 & (\mu(\ell) = i) \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_4)} & \ell = 6p+2 & (\mu(\ell) = -i) \\
 & \ell = 6p+4 & (\mu(\ell) = i)
\end{array}
\quad (B.16)
$$

$E_r = E_3$:

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,4pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,4p+2B_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_3)} & \ell = 4p+1 & (\mu(\ell) = -i) \\
 & \ell = 4p+3 & (\mu(\ell) = i)
\end{array}
\quad (B.17)
$$

$E_r = E_4$:

$$
\begin{array}{lll}
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,3pA_i)} & i = 1,2 & \\
{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_4)} & \ell = 3p+1 & (\mu(\ell) = -i) \\
 & \ell = 4p+2 & (\mu(\ell) = i)
\end{array}
\quad (B.18)
$$

$E_r = E_5$:

$$
\begin{aligned}
&{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,12pA_i)} && i=1,2\\
&{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,12p+6B_i)} && i=1,2\\
&{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_1)} && \ell=12p+5\ \ (\mu(\ell)=-i)\\
& && \ell=12p+7\ \ (\mu(\ell)=i)\\
&{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_2)} && \ell=12p+2\ \ (\mu(\ell)=-i)\\
& && \ell=12p+10\ \ (\mu(\ell)=i)\\
&{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_3)} && \ell=12p+3\ \ (\mu(\ell)=-i)\\
& && \ell=12p+9\ \ (\mu(\ell)=i)\\
&{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_4)} && \ell=12p+4\ \ (\mu(\ell)=-i)\\
& && \ell=12p+8\ \ (\mu(\ell)=i)\\
&{}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_5)} && \ell=12p+1\ \ (\mu(\ell)=-i)\\
& && \ell=12p+11\ \ (\mu(\ell)=i). \qquad \text{(B.19)}
\end{aligned}
$$

• $G = C_{\infty v}$
For any E_r and $\ell \neq 0$

$$ {}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell E_{r'})}\ r' = r\ell. \qquad \text{(B.20)} $$

• $G = D_{\infty h}$
Operators ${}^{[m_1\,-m_2]}\mathcal{T}^{(j,\,\ell \Gamma_\tau)}$ have the same labels as in equation (B.20) with:

$$
\begin{aligned}
E_{r_\alpha} &= E_{rg} \quad \tau = g\\
E_{r_\alpha} &= E_{ru} \quad \tau = g \quad m_1+m_2 \text{ even}\\
& \qquad\quad\ \tau = u \quad m_1+m_2 \text{ odd}. \qquad \text{(B.21)}
\end{aligned}
$$

Appendix C: Some ${}^{[m_1\,-m_2]}G$ coefficients and symmetry adapted coupling symbols

(a) For the lowest values of m_1 and m_2 these coefficients can be obtained for arbitrary E_r *irrep*; for $E_{r\alpha}$ type *irreps* the values of the coefficients are the same and τ indices added following the rules given in Appendix B. These are given in Tables 6, 7 and 8 for $[m_1\ -m_2] = [1\,0]$, $[m_1\ -m_2] = [0\ -1]$ and $[m_1\ -m_2] = [1\ -1]$, respectively associated with the fundamental *irreps* of $u(2)$ and that spanned by the generators. The coefficients for $[m_1\ -m_2] = [2\,0]$ and $[m_1\ -m_2] = [0\ -2]$, useful for establishing connections with operators quadratic in the coordinates, are also given. In Table 6 appear coefficients which keep the same values in all orientations I, II and III; Tables 7, 8 gather those depending on the chosen orientation I or II.

(b) For the special orientation III determined by the change of basis (112) we only give below the ${}^{[m_1\,-m_2]}\bar{\bar{G}}$ for $[m_1\ -m_2] = [1\,0]$, $[m_1\ -m_2] = [0\ -1]$:

$$
\begin{aligned}
{}^{[1\,0]}\bar{\bar{G}}{}^{\frac12}_{1E_r\bar{\bar{1}}} &= \frac{\omega^*}{\sqrt2} & {}^{[1\,0]}\bar{\bar{G}}{}^{-\frac12}_{1E_r\bar{\bar{1}}} &= \frac{\omega}{\sqrt2}\\
{}^{[1\,0]}\bar{\bar{G}}{}^{\frac12}_{1E_r\bar{\bar{2}}} &= -\frac{\omega}{\sqrt2} & {}^{[1\,0]}\bar{\bar{G}}{}^{-\frac12}_{1E_r\bar{\bar{2}}} &= -\frac{\omega^*}{\sqrt2}\\
{}^{[0\,-1]}\bar{\bar{G}}{}^{\frac12}_{1E_r\bar{\bar{1}}} &= \frac{\omega}{\sqrt2} & {}^{[0\,-1]}\bar{\bar{G}}{}^{-\frac12}_{1E_r\bar{\bar{1}}} &= \frac{\omega^*}{\sqrt2}\\
{}^{[0\,-1]}\bar{\bar{G}}{}^{\frac12}_{1E_r\bar{\bar{2}}} &= \frac{\omega^*}{\sqrt2} & {}^{[0\,-1]}\bar{\bar{G}}{}^{-\frac12}_{1E_r\bar{\bar{2}}} &= \frac{\omega}{\sqrt2} \qquad \text{(C.1)}
\end{aligned}
$$

with $\omega = e^{i\frac{\pi}{4}}$. If $r = n/4$ ($r = n/2$) the ${}^{[1\,-1]}\bar{\bar{G}}$, ${}^{[2\,-0]}\bar{\bar{G}}$ and ${}^{[0\,-2]}\bar{\bar{G}}$ values are those in Table 6.

(c) The operators in equation (78) are determined with the following coefficients identical in all orientations:

- for any E_r mode ${}^{[2\,-2]}G^0_{0A_1} = 1$,
- for an E_r mode with $r = p$ in groups D_{4p}, C_{4pv}, D_{2pd}

$$ {}^{[2\,-2]}G^2_{4A_1} = -\frac{1}{\sqrt2}, \quad {}^{[2\,-2]}G^{-2}_{4A_1} = -\frac{1}{\sqrt2}, $$

- for an E_r mode with $r = p$ in groups D_{3p}, C_{3pv}, T_d, O or $r = p, 2p$ in D_{6p}, C_{6pv} or $r = 2p$, $4p$ in D_{6pd}

$$ {}^{[3\,-3]}G^3_{6A_1} = \frac{i}{\sqrt2}, \quad {}^{[3\,-3]}G^{-3}_{6A_1} = -\frac{i}{\sqrt2}. $$

(d) With the given ${}^{[m_1\,-m_2]}G$ matrix elements exact values of several symmetry adapted coupling symbols equation (48) can be obtained. In particular those used in Section 4.3.1

$$ F\begin{pmatrix} \ell\Gamma\gamma & 1E_r\gamma' & ([1\,0]1/2)\\ ([1\ -1]1\ [1\,0]1/2) & 1E_r\gamma'' & \end{pmatrix} \qquad \text{(C.2)} $$

Table 6. ${}^{[m_1\,-m_2]}G$ coefficients identical in all orientations.

Case	$[m_1, -m_2]$	ℓ	$\Gamma\sigma$	m	${}^{[m_1\,-m_2]}G^m_{\ell\Gamma\sigma}$
	[0 0]	0	A_1	0	1
	[1 − 1]	0	A_2	0	$-i$
all	[2 − 0]	0	A_1	0	$-i$
	[0 − 2]	0	A_1	0	i
	[1 − 1]	2	B_1	1	$-i/\sqrt2$
	[1 − 1]	2	B_1	−1	$i/\sqrt2$
	[1 − 1]	2	B_2	1	$1/\sqrt2$
	[1 − 1]	2	B_2	−1	$1/\sqrt2$
$r = n/4$	[2 − 0]	2	B_1	1	$-1/\sqrt2$
($r = n/2$)	[2 − 0]	2	B_1	−1	$-1/\sqrt2$
	[2 − 0]	2	B_2	1	$-i/\sqrt2$
	[2 − 0]	2	B_2	−1	$i/\sqrt2$
	[0 − 2]	2	B_1	1	$1/\sqrt2$
	[0 − 2]	2	B_1	−1	$1/\sqrt2$
	[0 − 2]	2	B_2	1	$i/\sqrt2$
	[0 − 2]	2	B_2	−1	$-i/\sqrt2$

Table 7. $^{[m_1\,-m_2]}G^{m}_{\ell\Gamma\sigma}$ coefficients in orientation I.

Case	$[m_1, -m_2]$	ℓ	$\Gamma\sigma$	m	G
all	$[0-1]$	1	E_r 1	$1/2$	$1/\sqrt{2}$
	$[0-1]$	1	E_r 1	$-1/2$	$1/\sqrt{2}$
	$[0-1]$	1	E_r 2	$1/2$	$-i/\sqrt{2}$
	$[0-1]$	1	E_r 2	$-1/2$	$i/\sqrt{2}$
	$[1-0]$	1	E_r 1	$1/2$	$-i/\sqrt{2}$
	$[1-0]$	1	E_r 1	$-1/2$	$i/\sqrt{2}$
	$[1-0]$	1	E_r 2	$1/2$	$-1/\sqrt{2}$
	$[1-0]$	1	E_r 2	$-1/2$	$-1/\sqrt{2}$
$r' = 2r$	$[1-1]$	2	$E_{r'}$ 1	1	$-i/\sqrt{2}$
	$[1-1]$	2	$E_{r'}$ 1	-1	$i/\sqrt{2}$
	$[1-1]$	2	$E_{r'}$ 2	1	$-1/\sqrt{2}$
	$[1-1]$	2	$E_{r'}$ 2	-1	$-1/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 1	1	$-1/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 1	-1	$-1/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 2	1	$i/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 2	-1	$-/i\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 1	1	$1/\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 1	-1	$1/\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 2	1	$-i/\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 2	-1	$/i\sqrt{2}$
$r' = n-2r$ $(r' = 2n-2r)$	$[1-1]$	2	$E_{r'}$ 1	1	$-i/\sqrt{2}$
	$[1-1]$	2	$E_{r'}$ 1	-1	$i/\sqrt{2}$
	$[1-1]$	2	$E_{r'}$ 2	1	$1/\sqrt{2}$
	$[1-1]$	2	$E_{r'}$ 2	-1	$1/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 1	1	$-1/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 1	-1	$-1/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 2	1	$-i/\sqrt{2}$
	$[2-0]$	2	$E_{r'}$ 2	-1	$/i\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 1	1	$1/\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 1	-1	$1/\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 2	1	$i/\sqrt{2}$
	$[0-2]$	2	$E_{r'}$ 2	-1	$-/i\sqrt{2}$

Table 8. $^{[m_1\,-m_2]}\bar{G}^{m}_{\ell\Gamma\bar{\sigma}}$ in orientation II.

Case	$[m_1, -m_2]$	ℓ	$\Gamma\bar{\sigma}$	m	$\bar{G}$
all	$[0-1]$	1	E_r $\bar{1}$	$1/2$	i
	$[0-1]$	1	E_r $\bar{1}$	$-1/2$	0
	$[0-1]$	1	E_r $\bar{2}$	$1/2$	0
	$[0-1]$	1	E_r $\bar{2}$	$-1/2$	$-i$
	$[1-0]$	1	E_r $\bar{1}$	$1/2$	1
	$[1-0]$	1	E_r $\bar{1}$	$-1/2$	0
	$[1-0]$	1	E_r $\bar{2}$	$1/2$	0
	$[1-0]$	1	E_r $\bar{2}$	$-1/2$	1
$r' = 2r$	$[1-1]$	2	$E_{r'}$ $\bar{1}$	1	1
	$[1-1]$	2	$E_{r'}$ $\bar{1}$	-1	0
	$[1-1]$	2	$E_{r'}$ $\bar{2}$	1	0
	$[1-1]$	2	$E_{r'}$ $\bar{2}$	-1	1
	$[2-0]$	2	$E_{r'}$ $\bar{1}$	1	$-i$
	$[2-0]$	2	$E_{r'}$ $\bar{1}$	-1	0
	$[2-0]$	2	$E_{r'}$ $\bar{2}$	1	0
	$[2-0]$	2	$E_{r'}$ $\bar{2}$	-1	i
	$[0-2]$	2	$E_{r'}$ $\bar{1}$	1	i
	$[0-2]$	2	$E_{r'}$ $\bar{1}$	-1	0
	$[0-2]$	2	$E_{r'}$ $\bar{2}$	1	0
	$[0-2]$	2	$E_{r'}$ $\bar{2}$	-1	$-i$
$r' = n-2r$ $(r' = 2n-2r)$	$[1-1]$	2	$E_{r'}$ $\bar{1}$	1	0
	$[1-1]$	2	$E_{r'}$ $\bar{1}$	-1	-1
	$[1-1]$	2	$E_{r'}$ $\bar{2}$	1	-1
	$[1-1]$	2	$E_{r'}$ $\bar{2}$	-1	0
	$[2-0]$	2	$E_{r'}$ $\bar{1}$	1	0
	$[2-0]$	2	$E_{r'}$ $\bar{1}$	-1	$-i$
	$[2-0]$	2	$E_{r'}$ $\bar{2}$	1	i
	$[2-0]$	2	$E_{r'}$ $\bar{2}$	-1	0
	$[0-2]$	2	$E_{r'}$ $\bar{1}$	1	0
	$[0-2]$	2	$E_{r'}$ $\bar{1}$	-1	i
	$[0-2]$	2	$E_{r'}$ $\bar{2}$	1	$-i$
	$[0-2]$	2	$E_{r'}$ $\bar{2}$	-1	0

with $\gamma = \sigma$, $\bar{\sigma}$, $\bar{\bar{\sigma}}$ respectively for orientations I, II and III and the appropriate $^{[m_1\,-m_2]}G$ matrix in equation (48).

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