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The Racah–Wigner approach to harmonic oscillations of the cluster of nodes of a cube

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Abstract. The Racah-Wigner approach is applied to demonstrate various group-theoretic labelling schemes for modes of oscillations and elasticity parameters of the cluster of nodes of a cube. It is shown that there exists a fundamental basis in the configuration space, closed under the action of the group O_h—the geometric symmetry group of the equilibrium. It provides a transparent interpretation of all basic invariants. The fibre bundle and tensor product structure of invariant bilinear forms over the configuration space yields three classification schemes for elasticity parameters in a way that resembles Racah recoupling of several angular momenta.

1. Introduction

The systematic Racah-Wigner approach to the problem of energy level structure of multi-electron atoms, based on exploitation of spherical symmetry, provides a satisfactory description of complex atomic spectra (Biederharn and Louck 1981). Some attempts have recently been made to extend such an approach to describe the electronic structure and small vibrations of multicentre systems (Chan and Newman 1983, Lulek *et al* 1985a, b, Biel *et al* 1987, Lulek and Lulek 1987, Lulek 1989). The spherical symmetry is replaced in these attempts by that of geometric distribution of atoms (Newman 1983, Lulek *et al* 1985a). In the present paper we aim to demonstrate this approach on an example of small oscillations of a system of identical material points, arranged at the corners of a cube. We refer hereafter to this system as the cluster of nodes.

Within the Racah-Wigner approach one distinguishes two essential parts: the single-particle description and correlations. The former part consists of a complete classification of some optimal electronic states or normal modes, whereas the latter is accounted for by a multiple coupling of appropriate single-particle spaces. In the case of a multi-electron atom the first part corresponds to a choice of the single-electron central field *nlms* states of a given configuration, whereas the second corresponds to appropriate *LSJ*-type coupling of orbital and spin angular momenta. Similarly, in the case of multicentre systems, the first part consists of a complete classification of molecular spin orbitals (for a molecule within the LCAO method), Bloch functions (for a crystal within the tight binding), or normal modes of oscillations, and in the second part the coupling of angular momenta is replaced by the Clebsch-Gordan procedure, in particular by the Mackey theorem (Altmann 1977) for transitive representations, as proposed by Chan and Newmann (1984), or Lulek *et al* (1985b).

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A classification of the single-particle multicentre states by irreps of the geometric symmetry group alone is, as a rule, not complete for reason of multiplicities of some irreps. A substantial enriching of this classification is provided by factorisation of the configuration space into the positional and polarisational factors, responsible respectively for spatial distribution of equilibrium positions of nodes, and the single-centre displacements (Lulek 1980, Newman 1981, 1983, Chan and Newman 1982, 1983, Lulek and Lulek 1984a, b, Ceulemans 1985). Due to the fibre structure of the configuration space (Biel *et al* 1987), the action of the symmetry group is also factorised and the factors are decomposed into positional and polarisational irreps, which are eventually coupled into resultant irreps. Such a description, bearing an evident analogy to the atomic *lsjmj* case, has been also proposed independently by other authors, in various formulations (Flurry 1973, Fieck 1977, 1978, Michel and Mozrzymas 1982, Butler 1983). In the present paper we proceed to discuss the second step of the Racah–Wigner approach on an example of bilinear invariants of the configuration space, i.e. the elasticity parameters of the cube. Various coupling schemes emerging from double factorisation yield appropriate classifications of these parameters. We discuss also the role of these schemes in the determination of energy level structure of vibrations.

The approach presented here incorporates in a systematic way all geometric selection rules, resulting from both distribution of nodes and the polarisation of excitations. It can be easily extended to electronic variables, as well as to coupled electron–vibration modes since, in the jargon of differential geometry, electronic and mechanical variables are characterised by the same base (the cluster of nodes), and differ by their typical fibres (the single-centre states, e.g. 1s, $2p_x$, $3d_{xy}$, etc, or the displacements x, y, z). The choice of mechanical variables in our paper is granted by a nice, clear geometric interpretation of appropriate elasticity parameters. These parameters can be substituted by, e.g., hybridisation channels in the theory of valence fluctuations (Wohlleben and Wittershagen 1985, Drzazga *et al* 1987), electron–phonon coupling parameters, Cooper pairs in the superconductivity theory (Lulek 1989), or other quantities, according to an actual physical need. All such quantities emerge from the construction of covariant bilinear (or, more generally, multilinear) forms over appropriate single-particle linear spaces. The Racah–Wigner approach offers thus a reasonable chance to achieve a systematic insight into mechanisms of many-body correlations, by a full account of geometric selection rules.

2. The structure of configuration space

Let L be configuration space of a cluster $\tilde{R} = \{1, \ldots, 8\}$ of nodes of a cube (figure 1), so that $\dim L = 3 \times 8 = 24$. This space can be presented as the tensor product

$$L = B \otimes W \tag{1}$$

of the positional space

$$B = \text{LC}\{e_r \mid r \in \tilde{R}\} \tag{2}$$

i.e. the space of all formal linear combinations (LC) of nodes of the cluster $\tilde{R}$ over the field C of complex numbers, by the space W of single-centre displacements from the equilibrium position, referred hereafter to as the polarisation space (cf Biel *et al* 1987 for details). Let M be the mechanical representation (rep), i.e. the linear rep of the

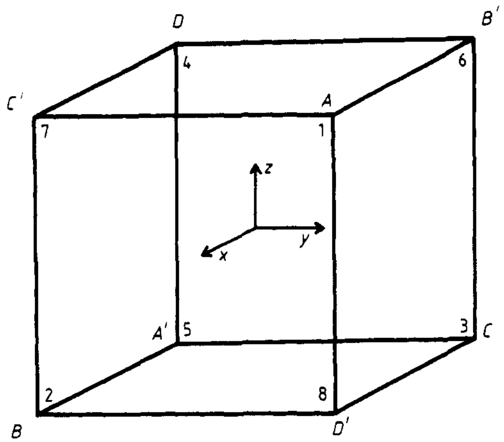

Figure 1. Notation for the cube.

octahedral group O_h of geometric symmetry of the cluster in equilibrium, describing the action of this group in the configuration space L.

Let

$$R \equiv R^{O_h:C_{3v}^A} \tag{3}$$

be the transitive rep of O_h on $\tilde{R}$, i.e. a permutation rep determined by the stability group $C_{3v}^A \subset O_h$ of the node $A \in \tilde{R}$ of figure 1 (cf appendix 1 for notation of elements of O_h), and V be the vector rep in the polarisation space W. Then

$$M \simeq R \otimes V \tag{4}$$

i.e. the action M is consistent with the factorisation (1), so that the group O_h acts separately on each factor B and W (respectively R and V).

Within the Racah–Wigner approach, we have to start with a definition of standard bases in the configuration space L, in analogy to known $|jm\rangle$ bases of angular momentum theory. Let

$$b_{\text{cart}}(L) = \{e_r^\alpha \mid \alpha \in \tilde{V}, r \in \tilde{R}\} \tag{5}$$

be the cartesian basis, with e_r^α being the unit vector for the displacement of rth node in the direction α, and $\tilde{V} = \{x, y, z\}$ the set of labels of cartesian coordinate axes, coinciding with fourfold axes of the group O_h. Then an irreducible basis is an arbitrary orthonormal basis consistent with the decomposoition

$$M \simeq A_{1g} \oplus E_g \oplus T_{1g} \oplus 2T_{2g} \oplus A_{2u} \oplus E_u \oplus 2T_{1u} \oplus T_{2u} \tag{6}$$

of the mechanical rep M into irreps of O_h, i.e. a basis.

$$b_{\text{irr}}(L) = \{e_\gamma^{\Gamma v} \mid \Gamma \in \tilde{O}_h, \gamma \in \tilde{\Gamma}, v \in \tilde{m}(M, \Gamma)\} \tag{7}$$

where $\tilde{O}_h$ is the set of irreps of O_h, $\tilde{\Gamma}$ is a standard basis for Γ, and $\tilde{m}(M,\Gamma) = \{1, \ldots, m(M,\Gamma)\}$ is the set of repetition indices for Γ in M, so that $m(M,\Gamma)$ is the multiplicity of Γ in M. Factorisation (1) enriches the classification of an irreducible basis in a sense that irreps entering the factors of the product (4) play the role of repetition indices, i.e.

$$\begin{aligned} &v = (\Lambda t, \Delta d, w) \\ &\Lambda, \Delta \in O_h' \qquad t \in \tilde{m}(R, \Lambda) \qquad d \in \tilde{m}(V, \Delta) \\ &w \in \tilde{m}(\Lambda \otimes \Lambda, \Gamma) \end{aligned} \tag{8}$$

(Lulek 1980, Newman 1981). In our case we have

$$R \simeq A_{1g} \oplus T_{2g} \oplus A_{2u} \oplus T_{1u}$$
$$V \simeq T_{1u} \tag{9}$$

so that most of the labels in (8) are redundant, and

$$v = \Lambda \tag{10}$$

i.e. the positional irrep provides a unique classification (Kuźma *et al* 1980). It is worth pointing out a similarity between the classification (7), (8) and that for atomic one-electron states, with the positional irrep Λ, polarisational Δ and resultant Γ being respectively analogues of orbital, spin and resultant angular momentum. This similarity suggests that we extend the routine Racah–Wigner methods for multi-electron systems in an atom to the case of multilinear forms over the configuration space L and, in particular, to invariant classification of elasticity parameters, discussed in section 3.

The irreducible basis (7), (8) is not very convenient for construction of multilinear invariants. It is better to introduce the third basis,

$$b_{\text{fund}}(L) = \{\varepsilon_r^\alpha \mid \alpha \in \tilde{V}, r \in \tilde{R}\} \tag{11}$$

where ε_r^α is the unit vector for displacements of rth node along the α axis, oriented *outside* the cube. This vector can be also looked at as an oriented edge of the cube. Elements ε_r^α coincide with the corresponding e_r^α of the cartesian basis (5) with an accuracy up to the sign, dependent on α and r. It is easy to observe that the basis (11) is a subset in L, closed and transitive under the action M of the group $\mathrm{O_h}$, so that the restriction

$$M|_{b_{\text{fund}}(L)} \simeq R^{\mathrm{O_h}:\mathrm{C}'_{1h}} = M' \tag{12}$$

is a transitive rep of the group $\mathrm{O_h}$, defined by the stability group $\mathrm{C}'_{1h} = \{E, \sigma_{\mathrm{CD}}\}$ of the edge $\mathrm{AB}' \in \varepsilon_{\mathrm{A}}^x$ in figure 1. Due to this property, we refer in the following to (11) as the fundamental basis.

The chain of subgroups (cf appendix 1 and table 1)

$$\mathrm{C}'_{1h} \subset \mathrm{C}_{3v}^{A} \subset \mathrm{O_h} \tag{13}$$

Table 1. Decomposition of the group $\mathrm{O_h}$ into left cosets with respect to the subgroup $\mathrm{C}'_{1h} = \{E, \sigma_{CD}\}$. Each left coset is classified by a pair (r, α), $r \in \tilde{R}$, $\alpha \in \tilde{V}$, according to the factorisation (16). Each row and each column of the table constitutes, respectively, a left coset with respect to the subgroup C_{3v}^{A} (the first row) and D_{4h}^{x} (the first column). First elements of the first column form the subgroup D_{2h}.

$r \in \tilde{R}$ \ $\alpha \in \tilde{V}$		x	y	z
1	A	E, σ_{CD}	C_{3A}, σ_{BC}	C_{3A}^{-1}, σ_{BD}
2	B	C_{2x}, σ_{AB}	C_{3C}, S_{4z}^{-1}	C_{3D}^{-1}, S_{4y}
3	C	C_{2y}, S_{4x}^{-1}	C_{3D}, S_{4z}	C_{3B}^{-1}, σ_{AC}
4	D	C_{2z}, S_{4x}	C_{3B}, σ_{AD}	C_{3C}^{-1}, S_{4y}^{-1}
5	A'	I, u_{CD}	S_{6A}^{-1}, u_{BC}	S_{6A}, u_{BD}
6	B'	σ_x, u_{AB}	S_{6C}^{-1}, C_{4z}	S_{6D}, C_{4y}^{-1}
7	C'	σ_y, C_{4x}	S_{6D}^{-1}, C_{4z}^{-1}	S_{6B}, u_{AC}
8	D'	σ_z, C_{4x}^{-1}	S_{6B}^{-1}, u_{AD}	S_{6C}, C_{4y}

with the stability group C_{3v}^{A} of the node A as an intermediate subgroup, defines a 'coarsening' of the orbit (11) of the transitive rep (12) into subsets $\{\varepsilon_r^\alpha \mid \alpha \in \tilde{V}\} \in \tilde{R}$, called imprimitivity systems (rows of table 1), each consisting of all vectors, corresponding to a node r of the cluster $\tilde{R}$. The transitive rep M' can be thus factorised as

$$M' = R^{\mathrm{O_h}:\mathrm{C}'_{1\mathrm{h}}} \simeq R^{\mathrm{O_h}:\mathrm{C}_{3\mathrm{v}}^{A}} \times R^{\mathrm{O_h}:\mathrm{D}_{4\mathrm{h}}^{x}} \equiv R \times V' \tag{14}$$

where R is given again by (3), and

$$V' = R^{\mathrm{O_h}:\mathrm{D}_{4\mathrm{h}}^{x}} \tag{15}$$

is the transitive rep, acting on the set $\tilde{V}$ of labels of the cartesian basis of the single-centre space W, i.e. on the set of columns of table 1.

Apparently, one can interpret the factorisation (14) as a permutation counterpart of the factorisation (4) with the orbit (11) being the cartesian product

$$b_{\mathrm{fund}}(L) \simeq \tilde{R} \times \tilde{V} \tag{16}$$

the analogy of the tensor product (1) for the configuration space L. A deeper insight shows, however, that such an analogy is superficial. We observe that whereas the positional factor R is the same in both factorisations, (4) and (14), the polarisation reps V and V' are inequivalent: $V \simeq T_{1\mathrm{h}}$ is a faithful irrep of $\mathrm{O_h}$ whereas $V' \simeq A_{1\mathrm{g}} \oplus E_{\mathrm{g}}$ is reducible, and moreover not faithful, with the non-trivial kernel

$$\operatorname{Ker} V' = \mathrm{D}_{2\mathrm{h}} = \{E, C_{2x}C_{2y}, C_{2z}, I, \sigma_x, \sigma_y, \sigma_z\} \lhd \mathrm{O_h} \tag{17}$$

($\lhd$ denoting normal subgroup) consisting of all first elements of the first columns of table 1.

Linear inequivalence of polarisational reps V and V' has its origin in the structure of the configuration space L. The point is that, despite apparently evident formulas (1) or (16), the positional and polarisational factors do not enter the configuration space on an equal footing. According to the paper of Biel *et al* (1987), the situation can be adequately displayed in terms of fibre bundles. The configuration space L is the space CsE of all sections of the bundle E with the base $\tilde{R}$ and the typical fibre W. The positional factor R has thus the 'absolute' meaning, emerging from the base of the bundle E, whereas the polarisational factor is 'relative' in the sense that it depends on the choice of basis vectors, which can be performed independently for each copy W_r of the typical fibre W, corresponding to a node $r \in \tilde{R}$. In particular, the presentation of L in the tensor product for (1) is not canonical, i.e. it does not result from any essential, structural properties of L, but merely from the choice of sets $\{e_r^\alpha \mid \alpha \in \tilde{V}\}$ for each fibre W_r as the parallel translation of the standard tetragonal basis $\{e^\alpha \mid \alpha \in \tilde{V}\}$ in the typical fibre W. On the other hand, the fundamental basis (11) is generated from the set $\{e_A^\alpha \mid \alpha \in \tilde{V}\}$ for the node A by means of geometric operations of the point group $\mathrm{D}_{2\mathrm{h}}$, given by (17). The fundamental basis (11) can be thus obtained from the cartesian basis (5) as the result of a sign modulation by the point group $\mathrm{D}_{2\mathrm{h}}$, or a gauge $\varepsilon_r^\alpha = \pm e_r^\alpha$, with a sign dependent on α and r.

Within such a picture, the action M of the group $\mathrm{O_h}$ in the configuration space L can be factorised only in some special cases. The reason is that the action on the base $\tilde{R}$ of the bundle E is determined canonically by the transitive rep R, whereas, in general one cannot define a total 'action on the fibre' due to an irregular dependence of basis vectors on the label r of a fibre W_r. Such total polarisational reps can be reasonably defined only for particular, coherent choices of basis vectors in several fibres. In the parallel translation case, i.e. the factorisation (1), the polarisational rep V entering the mechanical representation M as a factor in (4), is an ordinary geometric action of the group O_h in the space W of typical fibre. In the case of fundamental basis (11), i.e. the factorisation (16), the action of the group O_h is related to the semidirect product structure

$$O_h \equiv D_{2h} \square C^A_{3v} \tag{18}$$

shown in table 1, where passive group D_{2h} coincides with the kernel (17) of the new polarisational rep V', and the active group C^A_{3v} coincides with the stability group of the node A. Now the old polarisational rep $V \simeq T_{1u}$ can be looked at as the composition of the geometric action $V{\downarrow}D_{2h}$ of the passive group D_{2h}, consisting in the sign modulation of unit vectors e^x, e^y, e^z, and the purely permutational action V'' of the active group C^A_{3v}, given by the transitive representation

$$V'' \simeq R^{C^A_{3v}:C'_{1h}} \tag{19}$$

on the set $\tilde{V}$ (cf the description of permutation–inversion structure of the group O_h (18) by Florek *et al* (1988)). The permutation rep M' of the group O_h on the fundamental basis (11), with the factorisation (16), can be interpreted, according to (14), as the action which is modulated with respect to M in such a way that the sign modulation of the cartesian basis, i.e. the action $V{\downarrow}D_{2h}$ of the passive group of the semidirect product (18), is carried from the polarisation factor V to the positional R. As the result, the new polarisation rep V' of the group O_h is weakly equivalent (Michel and Mozrzymas 1978, 1981) to purely permutational action V'' of the group C^A_{3v}, i.e.

$$V' \simeq R^{O_h:D^x_{4h}} \approx V'' \simeq R^{C^A_{3v}:C'_{1h}} \equiv R^{(O_h/D_{2h}):(D^x_{4h}/D_{2h})} \tag{20}$$

where $\approx$ denotes weak equivalence. In other words, V' is an effective action, resulting from the effective action V'' by the extension (18) of the effective group C^A_{3v} by the neutral Kernel D_{2h}.

We observe therefore that the non-equivalence of polarisational reps V and V', associated respectively with the parallel translation and sign modulation of the single-centre cartesian basis is an effect of relativity, having its source in fibre structure of the configuration space. The configuration space L bears here a formal analogy to Galilean spacetime, i.e. to space of all sections of the bundle with the base and typical fibre being respectively the axis of an 'absolute' time and 'relative' three-dimensional Euclidean space (Ingarden and Jamiołkowski 1985; see also Biel *et al* 1987, Lulek 1989). The choice of the basis in each fibre W_r corresponds to a reference frame in Galilean spacetime, whereas a rigid adherence to the factorisation (1) is essentially similar to acceptation of Aristotelian spacetime, where both time and space are absolute, as a consequence of the prejudice for 'frame of absolute rest'.

3. The harmonic potential

3.1. *The space of elasticity parameters*

The potential energy describing interactions in the cluster $\tilde{R}$ in the harmonic approximation, is given by

$$U(u) = U_0 + \phi(u, u) \tag{21}$$

$$u \in L \tag{22}$$

where U_0 is the energy of equilibrium, and $\phi : L \times L \to C$ is a symmetric, O_h-invariant, complex-valued bilinear form over the configuration space L. The harmonic potential can therefore be constructed in terms of bilinear combinations of the displacement vector

$$u = \sum_{r \in \tilde{R}, \alpha \in \tilde{V}} x_r^\alpha e_r^\alpha = \sum_{\substack{\Gamma \in \tilde{O}_h \\ \gamma \in \tilde{\Gamma}, v \in \tilde{m}(M,\Gamma)}} q_\gamma^{\Gamma v} e_\gamma^{\Gamma v} \tag{23}$$

corresponding to invariants of the group O_h in the symmetric square $L^{\{2\}}$ of the configuration space L. An elementary character formula yields for our case

$$\dim L_{\mathrm{inv}}^{\{2\}} \equiv m(M^{\{2\}}, A_{1g}) = 12 \tag{24}$$

$$\dim L_{\mathrm{inv}}^{\{1^2\}} \equiv m(M^{\{1^2\}}, A_{1g}) = 2 \tag{25}$$

where $L^{\{1^2\}}$ is the antisymmetric part of $L^2 \equiv L \otimes L$. The small oscillations of the cluster $\tilde{R}$ can be thus described in the most general form by 12 independent elasticity parameters.

The elasticity parameters can be classified in several ways by the choice of a basis in the space $L^{\{2\}}$ (or in the space L_{inv}^2 followed by elimination of two antisymmetric invariants). Each such choice determines a classification scheme. In particular, the Racah–Wigner approach to factorisation (1) naturally suggests two classification schemes: irreducible and transitive (Lulek 1989). We discuss here also the fundamental scheme, emerging from the basis (11).

3.2. The irreducible scheme

The irreducible scheme, presented graphically in figure 2, consists of the construction of invariants $I_{vv'}(\Gamma)$ by the standard invariant pairing of two mutually conjugated Γ and Γ^* modes of the irreducible basis (7). The classification of elasticity parameters within this scheme is given in table 2.

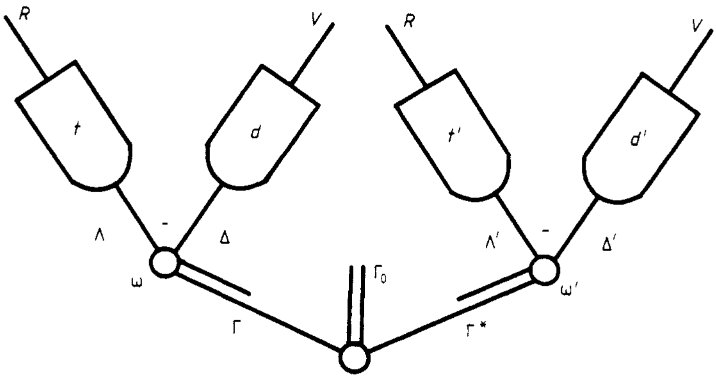

Figure 2. The irreducible scheme for the classification of invariant elasticity parameters (Γ_0 denotes the trivial irrep of the group G). See Lulek (1973) for details of graphical conventions.

Table 2. Classification of invariants $I_{\lambda\lambda'}(\Gamma)$ in the irreducible scheme.

Γ	A_{1g}	E_g	T_{1g}	T_{2g}				A_{2u}	E_u	T_{2u}	T_{1u}			
Λ	T_{1u}	T_{1u}	T_{1u}	A_{2u}		T_{1u}		T_{2g}	T_{2g}	T_{2g}	A_{1g}		T_{2g}	
Λ'	T_{1u}	T_{1u}	T_{1u}	A_{2u}	T_{1u}	A_{2u}	T_{1u}	T_{2g}	T_{2g}	T_{2g}	A_{1g}	T_{2g}	A_{1g}	T_{2g}

This scheme is the most convenient for calculations of secular equations (both classical and quantum mechanical), since basic invariants are adapted to the decomposition of configuration space L imposed by (6).

The physical meaning of invariants $I_{vv'}(\Gamma)$ is clearly imposed by their role in the secular problem. Invariants associated with a given $\Gamma \in \tilde{\mathrm{O}}_{\mathrm{h}}$ determine the spectrum of eigenfrequencies within the subspace of all normal modes of type Γ, *independently* of other subspaces. Thus the irreducible scheme explicitly accounts for all geometric selection rules.

In cases when $m(M, \Gamma) = 1$, there is a single, uniquely determined invariant $I(\Gamma)$, which is directly proportional to the square of the eigenfrequency of a normal mode. We have six such cases related to $\Gamma = A_{1g}, A_{2u}, E_g, E_u, T_{1g}, T_{2u}$.

When $m(M, \Gamma) = 2$, the dependence of the square of eigenfrequencies on elasticity parameters becomes nonlinear due to non-diagonal invariants $I_{vv'}(\Gamma)$, $v \neq v'$. These invariants can be recognised as parameters, responsible for the hybridisation between the modes $e_\gamma^{\Gamma v}$ and $e_\gamma^{\Gamma v'}$, of the same type, the hybridisation being the same for each $\gamma \in \tilde{\Gamma}$. For example $I_{A_{1g}T_{2g}}(T_{1u})$ is the parameter of hybridisation between translational $\Lambda\Gamma = A_{1g}T_{1u}$ and oscillational $T_{2g}T_{1u}$ modes.

The irreducible scheme clearly demonstrates the fact that knowledge of the vibration spectrum of a cluster (i.e. its eigenfrequencies) is, in general, not sufficient for the determination of elasticity parameters, due to the $m(M, \Gamma) > 1$ multiplicities. For example in our case there are 10 eigenfrequencies (possibly degenerate), determined by 12 elasticity parameters. To find these parameters we need to know not only the eigenfrequencies but also certain additional information on the hybridisation effects for both two-dimensional secular eigenproblems with $\Gamma = T_{2g}$ and T_{1u}.

3.3. *The transitive scheme*

In the transitive scheme (figure 3) we apply the factorisation (4) of the mechanical rep M to perform a recoupling according to the formula

$$M \otimes M \simeq (R \otimes V) \otimes (R \otimes V) \simeq (R \otimes R) \otimes (V \otimes V) \tag{26}$$

and make use of the permutation structure of the factor $R \otimes R$ by means of the Mackey theorem described in appendix 2. At the level of permutation reps we obtain

$$R \times R \equiv R^{\mathrm{O_h}:\mathrm{C}_{3v}^{A}} \times R^{\mathrm{O_h}:\mathrm{C}_{3v}^{A}} \simeq 2R^{\mathrm{O_h}:\mathrm{C}_{3v}^{A}} + 2R^{\mathrm{O_h}:\mathrm{C}_{1h}'} \tag{27}$$

$$\Omega \equiv \Omega(\mathrm{C}_{3v}^{A}, \mathrm{C}_{3v}^{A}) = \{\omega = 0, 1, 2, 3\} \tag{28}$$

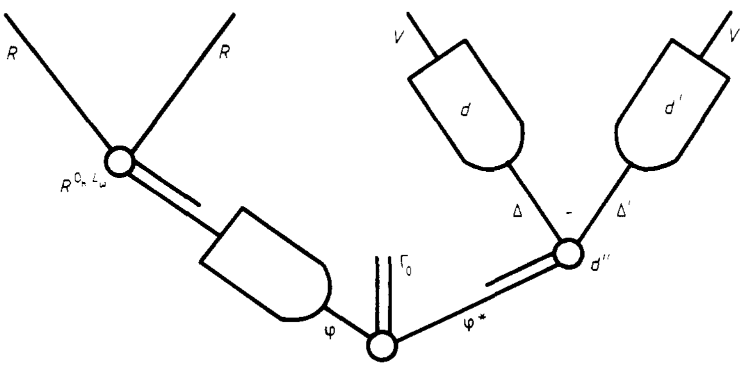

Figure 3. The transitive scheme.

and

$$L_\omega = \begin{cases} C_{3v}^{A} & \text{for } \omega = 0, 3 \\ C_{1h}' & \text{for } \omega = 1, 2. \end{cases} \tag{29}$$

Orbits $\omega = 0$, 1, 2, and 3 of appropriate resultant transitive reps in (27) correspond respectively to diagonal pairs $\{11, 22, \ldots, 88\} \equiv \{AA, BB, \ldots, D'D'\}$, pairs of first $\{16, 17, \ldots, 83\} \equiv \{AB', AC', \ldots, D'C\}$, second $\{12, 13, \ldots, 87\} \equiv \{AB, AC, \ldots, D'C'\}$ and third $\{15, 26, \ldots, 84\} \equiv \{AA', BB', \ldots, D'D\}$ neighbours. The orbit ω encloses $48/|L_\omega|$ pairs, i.e. 8, 24, 24, and 8 for $\omega = 0$, 1, 2, and 3, respectively.

Invariants within the transitive scheme are obtained by pairing of an irrep $\phi \in \tilde{O}_h$ enclosed in the resultant transitive rep $R^{G:L_\omega}$ of the Mackey decomposition (27) (i.e. arising from the base $\tilde{R} \times \tilde{R}$) with the complex conjugate counterpart ϕ^* from the fibre space $W \otimes W$ in accordance with figure 3. They are denoted as $I(\omega, \phi, f, \Delta d, \Delta' d', d'')$, $\omega \in \Omega$, ϕ, Δ, $\Delta' \in \tilde{O}_h$, $f \in \tilde{m}(R^{O_h:L_\omega}, \phi)$, d, $d' \in \tilde{m}(V, \Delta)$, $d'' \in \tilde{m}(\Delta \otimes \Delta', \phi^*)$.

In our case $\Delta = \Delta' = T_{1u}$, so that the indices Δd, $\Delta' d'$, d'' are redundant. Moreover, the decompositions of $R^{O_h:C_{3v}^{A}} \simeq R$ and $R^{O_h:C_{1h}'} \simeq M' \simeq M$ are given respectively by (9) and (6). As shown in detail by Lulek (1989), antisymmetric invariants enter only through the multiplicty indices $f \in \tilde{m}(R^{O_h:L_\omega}, \phi)$ (note that all double cosets in (28) are self-inverse, and $\Delta = T_{1u}$ is a tensor irrep). Since $m(R^{O_h:L_\omega}, \phi) \leqslant 2$, the index f is also redundant under the restriction to symmetric bilinear invariants. The basic invariants for the transitive scheme can thus be denoted by $I(\omega, \phi)$, where ω is the orbit of geometrically equivalent pairs of nodes of the cluster $\tilde{R}$ and ϕ is the irrep entering the resultant positional transitive rep $R^{O_h:L_\omega}$, associated with the cartesian square $\tilde{R} \times \tilde{R}$ of the base of the bundle E, and having its counterpart ϕ^* in the fibre space. The transitive scheme for the cluster $\tilde{R}$ is given in table 3.

Table 3. Classification of invariants $I(\omega, \phi)$ in the transitive scheme. The additional multiplicity label $f \in \tilde{m}(R^{O_h:L_\omega}, \phi)$, for $\omega = 1$, 2 and $\phi = T_{2g}$, distinguishes symmetric ($f = \{2\}$) and antisymmetric ($f = \{1^2\}$) bilinear forms.

ω	0		2					3		1				
ϕ	A_{1g}	T_{2g}	A_{1g}	E_g	T_{1g}	T_{2g}	T_{2g}	A_{1g}	T_{2g}	A_{1g}	E_g	T_{1g}	T_{2g}	T_{2g}
f	—	—	—	—	—	$\{2\}$	$\{1^2\}$	—	—	—	—	—	$\{2\}$	$\{1^2\}$

A nice feature of the transitive scheme is the simplicity, having its origin in the classification label ω of geometrically equivalent pairs of nodes. Invaraints within each orbit ω are further labelled by the irrep ϕ, responsible for a mutual compensation of effects of spatial distribution of nodes (ϕ enters the positional rep $R^{O_h:L_\omega}$) and polarisation effects (ϕ^* originates from the product $\Delta \otimes \Delta'$ (cf figure 3)). In particular, for $\phi = A_{1g}$ we obtain permutational invariants, i.e. quantities which are invariant under the action of the group O_h separately on the square $\tilde{R} \times \tilde{R}$ of the base and on $W \otimes W$ of the fibre of the bundle E. They can also be interpreted as 'isotropic' interactions between appropriate nodes, whereas those associated with $\phi \neq A_{1g}$ can be interpreted as various types of anisotropic couplings. We observe from table 3 that each orbit ω is associated with exactly one isotropic interaction, whereas the number of types of anisotropic couplings depends upon the orbit ω. There is just one type $\phi = T_{2g}$ of anisotropic coupling for $\omega = 0$ and $\omega = 3$ ('self-interaction' and third neighbours, respectively), whereas orbits $\omega = 1$ and 2 (first and second neighbours, respectively) are associated with three different types of anisotropic couplings, classified by $\phi = E_g$, T_{1g} and T_{2g}.

On the other hand the transitive scheme involves a suppression of the irreducible structure of the configuration space imposed by the decomposition (6) of a mechanical rep since a single basic invarant of this scheme exerts an influence on the structure of the spectrum of eigenfrequencies for various irreps Γ.

3.4. The fundamental scheme

It is worth observing that both irreducible and transitive schemes can be derived simply on the grounds of a non-canonical factorisation (1) of the configuration space, without resorting to its fibre structure. In this subsection we proceed to use explicitly this structure by exploiting the existence of the fundamental basis (11), with the closure property (12). The main advantage of the transitive scheme may be formally attributed to the application of the Mackey theorem (appendix 2) to two positional factors $R \times R$ of the direct square $M \otimes M$ of the mechanical rep. Using the fundamental basis, we are able to extend the powerful Mackey theorem for the whole square $M \otimes M$, or, more exactly, for its permutational counterpart $M' \times M'$, with M' given by (12). The Mackey decomposition

$$M' \times M' \simeq R^{O_h:C'_{1h}} \times R^{O_h:C'_{1h}} \simeq 4R^{O_h:C'_{1h}} + 10R^{O_h:C_1} \tag{30}$$

(cf figure 4(a)) yields a unique classification of 14 invariants in L^2_{inv} by a single label $\eta \in H(C'_{1h}, C'_{1h})$, where $H(C'_{1h}, C'_{1h})$ is the set of double cosets of O_h with respect to the pair (C'_{1h}, C'_{1h}) of subgroups (cf (A2.1) and (A2.2)). The set $H(C'_{1h}, C'_{1h})$ consists of 10 self-inverse double cosets and two pairs of mutually inversible ones, which yields 12 symmetric and 2 antisymmetric invariants (cf Lulek 1989).

The unique classification on invariants I_η, $\eta \in H(C'_{1h}, C'_{1h})$ in the fundamental scheme is based entirely on the action of the transitive rep $R^{O_h:C'_{1h}}$ on the basis $b_{\text{fund}}(L)$ of the configuration space L. We do not even need to introduce any linear structure—it is sufficient simply to count on one's fingers, i.e. to perform some combinatorial considerations on the cartesian square of the finite set (11). Each basic invariant I_η is now just an ordinary sum of products $\varepsilon^\alpha_r \varepsilon^{\alpha'}_{r'}$, corresponding to tetrads (r, r', α, α') running over all elements of the orbit $\eta \in H(C'_{1h}, C'_{1h})$ of the resultant transitive rep $R^{O_h:L_\eta}$.

Until now we have not used the factorisation (14) of M'. We proceed to use this factorisation in a way formally similar to the case of transitive scheme, (26), namely

$$M' \times M' \simeq (R \times V') \times (R' \times V') \simeq (R \times R) \times (V' \times V') \tag{31}$$

where the new polarisation factor V' accounts for the difference between fundamental (11) and cartesian (5) bases, emerging from the fibre structure of the configuration space as described in section 2. It is worth observing that the classification of fundamental invariants by the group action is already complete, so the factorisation (14) cannot yield any enriching of this classification. It can merely provide a more explicit meaning of the classification label η by a separation of positional and polarisational effects.

We apply the Mackey theorem in two stages: first to each factor on the RHS of (31) separately, and then to coupling the intermediate transitive reps to resultant ones (figure 4(b)).

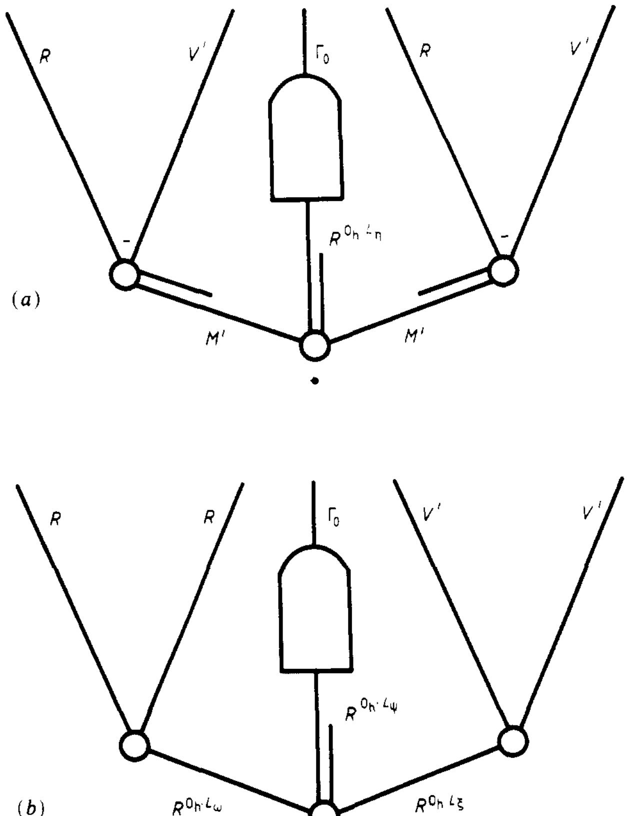

Figure 4. The fundamental scheme. $V' = R^{O_h:D_{4h}^x}$ and $M' = R^{O_h:C'_{1h}}$ are transitive retracts of polarisational and mechanical reps, respectively. (*a*) Immediate application of the Mackey theorem; (*b*) an application in two stages.

The first stage yields (27) for the positional factor and

$$V' \times V' \simeq R^{O_H:D_{4h}^x} + R^{O_h:D_{2h}} \tag{32}$$

for the polarisational one. The corresponding set of double cosets for the positional and polarisational factors is given respectively by Ω of (28) and by

$$\Xi \equiv \Xi(D_{4h}^x, D_{4h}^x) = \{\xi = 1, 2\}. \tag{33}$$

The latter set is associated with the decomposition

$$\tilde{V} \times \tilde{V} = \{xx, yy, zz\} \cup \{xy, xz, yx, yz, zx, zy\} \tag{34}$$

of the square of the orbit $\tilde{V}$ of $R^{O_h:D_{4h}^x}$ into diagonal ($\xi = 1$) and off-diagonal ($\xi = 2$) orbits.

The second stage yields

$$R^{O_h:L_\omega} \times R^{O_h:L_\xi} = \sum_{\psi \in \Psi(L_\xi, L_\omega)} R^{O_h:L_\psi} \tag{35}$$

which allows us to replace the single classification label η by the sequence

$$\eta = (\omega, \xi, \psi) \qquad \omega \in \Omega \qquad \xi \in \Xi \qquad \psi \in \Psi(L_\xi, L_\omega) \tag{36}$$

of labels of double cosets originating from appropriate Mackey decompositions. The full classification is achieved by using the following Mackey decompositions:

$$R^{O_h:C^A_{3v}} \times R^{O_h:D_{2h}} \simeq R^{O_h:C_1} \tag{37}$$

$$R^{O_h:C'_{1h}} \times R^{O_h:D^x_{4h}} \simeq R^{O_h:C'_{1h}} + R^{O_h:C_1} \tag{38}$$

$$R^{O_h:C'_{1h}} \times R^{O_h:D_{2h}} \simeq 3R^{O_h:C_1} \tag{39}$$

and (14), a particular case of the Mackey theorem (A2.1). The classification of invariants I_η, $\eta=(\omega, \xi, \psi)$ with in the fundamental scheme is given in table 4.

Table 4. Classification of invariants I_η, $\eta \equiv (\omega, \xi, \psi)$, in the fundamental scheme (g_η is a representative of the double coset $\eta \in H(C'_{1h}, C'_{1h})$).

ω	ξ	ψ	L_ω	L_ξ	L_ψ	g_η	η
0	1	—	C^A_{3v}	D^x_{4h}	C'_{1h}	E	1
	2	—	C^A_{3v}	D_{2h}	C_1	C_{3A}	2
2	1	1	C'_{1h}	D^x_{4h}	C'_{1h}	C_{2x}	3
		2	C'_{1h}	D^x_{4h}	C_1	C_{2y}	4
	2	1	C'_{1h}	D_{2h}	C_1	S^{-1}_{4y}	5
		2	C'_{1h}	D_{2h}	C_1	S_{4y}	6
		3	C'_{1h}	D_{2h}	C_1	C_{3B}	7
3	1	—	C^A_{3v}	D^x_{4h}	C'_{1h}	I	8
	2	—	C^A_{3v}	D_{2h}	C_1	S_{6A}	9
1	1	1	C'_{1h}	D^x_{4h}	C'_{1h}	σ_x	10
		2	C'_{1h}	D^x_{4h}	C_1	C_{4x}	11
	2	1	C'_{1h}	D_{2h}	C_1	C_{4y}	12
		2	C'_{1h}	D_{2h}	C_1	C^{-1}_{4y}	13
		3	C'_{1h}	D_{2h}	C_1	S_{6B}	14

The factorisation (14) provides thus an insight into the internal structure of the fundamental classification scheme. First of all, it encloses the label ω of pairs of geometrically equivalent nodes, the main advantage of the transitive scheme. Next, the label ξ distinguishes the class of diagonal (α, α), $(\xi=1)$ and off-diagonal (α, α'), $\alpha \neq \alpha'$ $(\xi=2)$ pairs of polarisation labels α, $\alpha' \in \tilde{V}$.

In the cases $\omega=0$ (self-interaction) and $\omega=3$ (interaction of third neighbours) the sequence (ω, ξ), $\xi \in \Xi$, already fully characterises the basic invaraiants $I_\eta \equiv I_{\omega\xi}$.

The situation is slightly more complex in cases where $\omega=1$ and $\omega=2$ (respectively first and second neighbours), since the Mackey decompositions (38) and (39) imply here a non-trivial interplay of positional and polarisational effects, involved in the third classification label $\psi \in \Psi(L_\xi, L_\omega)$. This label can easily be interpreted geometrically. For example let us consider the orbit $\omega=1$. Each pair (r_1, r_2) of nearest neighbours defines canonically a direction which is parallel to the one basic polarisation and perpendicular to the two others. For example, the pair $(A, B')=(1, 6)$ is parallel to $\alpha=x$, and perpendicular to y and z. This distinction yields two orbits $\psi \in \Psi(D^x_{4h}, C'_{1h})$ for the diagonal case $\xi=1$, and three orbits $\psi \in \Psi(D_{2h}, C'_{1h})$ for the off-diagonal case

$\xi=2$. All these five cases are demonstrated in figure 5, where continuous arrows denote the harmonic reaction of the cluster $\tilde{R}$, under the coupling $I_{1\xi\psi}$, to a displacement of the first node along the x axis (the broken arrow), assuming that elasticity parameters for all other invariants vanish. Similar considerations apply to the second neighbours ($\omega=2$), since each pair of such neighbours defines canonically a plane perpendicular to one axis, and parallel to the two others. Results are given in figure 6. Figures 5 and 6 provide a simple, transparent geometric interpretation of basic invariants in the fundamental scheme.

An additional advantage of this scheme is the simple form of the secular matrix in the fundamental basis (11). Each element of this matrix contains exactly one basic invariant, with the coefficient 1 (cf table 5). Formally it is a consequence of the fact that the set of elements of the potential matrix can be identified with the carrier set of the permutation rep $M'\times M'$ of the group O_h, and the invariants are associated with its orbits. It justifies the term 'fundamental scheme', since this scheme provides speaking somehow imprecisely, the optimal 'rectangular reference system' for the description of the bilinear harmonic invariants in the cluster $\tilde{R}$.

3.5. *The form of invariants*

The explicit form of invariants can be written down most easily in the fundamental scheme, since it is given immediately from table 5, e.g.

$$\begin{aligned}\tfrac{1}{2}I_3\equiv\tfrac{1}{2}I_{211}=\varepsilon_1^x\varepsilon_2^x+\varepsilon_1^y\varepsilon_3^y+\varepsilon_1^z\varepsilon_4^z+\varepsilon_3^x\varepsilon_4^x+\varepsilon_2^y\varepsilon_4^y+\varepsilon_2^z\varepsilon_3^z&\\ +\varepsilon_5^x\varepsilon_6^x+\varepsilon_5^y\varepsilon_7^y+\varepsilon_5^z\varepsilon_8^z+\varepsilon_7^x\varepsilon_8^x+\varepsilon_6^y\varepsilon_8^y+\varepsilon_6^z\varepsilon_7^z&\qquad(40)\end{aligned}$$

$$\tfrac{1}{2}I_4\equiv\tfrac{1}{2}I_{212}=\varepsilon_1^x\varepsilon_3^x+\varepsilon_1^x\varepsilon_4^x+\ldots+\varepsilon_7^y\varepsilon_8^y+\varepsilon_7^z\varepsilon_8^z \qquad (41)$$

etc.

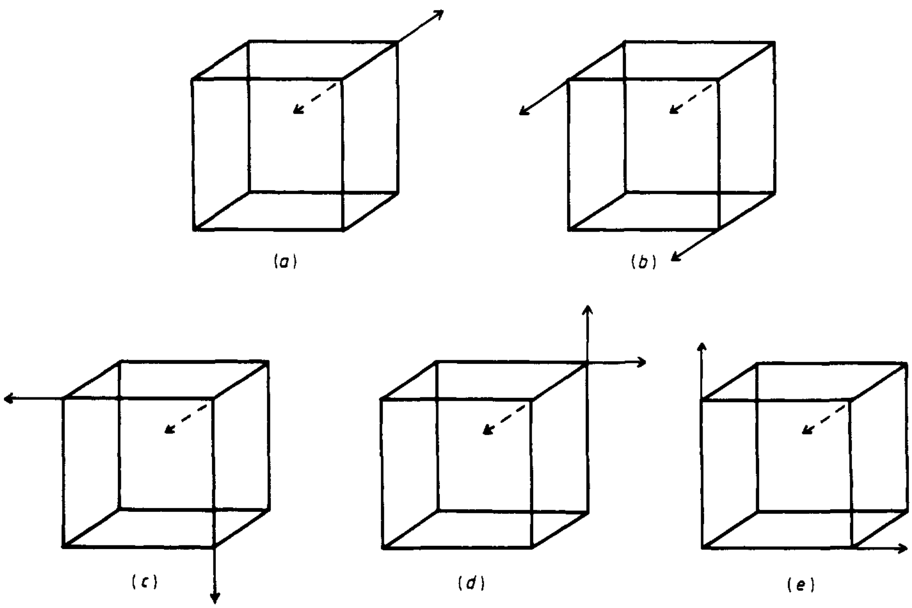

Figure 5. Geometric interpretation of basic invariants in the fundamental scheme for nearest neighbours ($\omega=1$). (a) I_{111}, (b) I_{112}, (c) I_{121}, (d) I_{122}, (e) I_{123}.

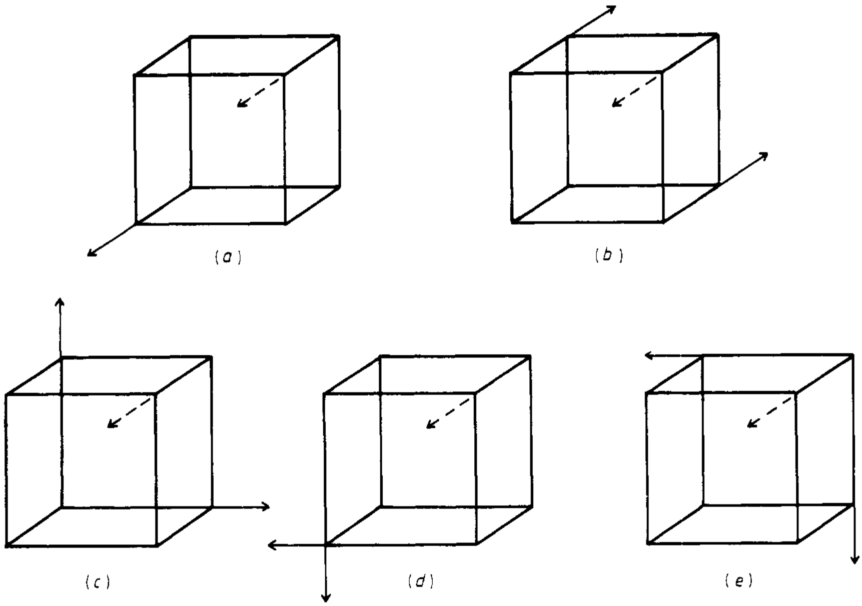

Figure 6. Basic invariants for the next-nearest neighbours ($\omega = 2$). (*a*) I_{211}, (*b*) I_{212}, (*c*) I_{221}, (*d*) I_{222}, (*e*) I_{223}.

Table 5. The matrix of the harmonic potential in the fundamental basis. Numbers in the table stand for elasticity parameters of fundamental basic invariants, labelled by η according to the last column in table 4. The table contains the first quarter of the matrix, corresponding to $r = 1$, 2, 3, 4 (enumeration of nodes according to figure 1). It is equal to the second diagonal quarter, whereas the two off-diagonal quarters are obtained from it by adding the number 7 to each quantity in the diagonal quarter.

	r'	1			2			3			4		
r	$\alpha \backslash \alpha'$	x	y	z	x	y	z	x	y	z	x	y	z
	x	1	2	2	3	6	6	4	5	7	4	7	5
1	y	2	1	2	5	4	7	6	3	6	7	4	5
	z	2	2	1	5	7	4	7	5	4	6	6	3
	x	3	6	6	1	2	2	4	7	5	4	5	7
2	y	5	4	7	2	1	2	7	4	5	6	3	6
	z	5	7	4	2	2	1	6	6	3	7	5	4
	x	4	5	7	4	7	5	1	2	2	3	6	6
3	y	6	3	6	7	4	5	2	1	2	5	4	7
	z	7	5	4	6	6	3	2	2	1	5	7	4
	x	4	7	5	4	5	7	3	6	6	1	2	2
4	y	7	4	5	6	3	6	5	4	7	2	1	2
	z	6	6	3	7	5	4	5	7	4	2	2	1

The transformation between basic invariants in the fundamental and transitive schemes is diagonal with respect to the label ω of the class of neighbours. For the zeroth and third neighbours the corresponding basic invariants coincide, i.e.

$$\begin{aligned} I_{\omega 1} &= I(\omega, A_{1g}) \\ I_{\omega 2} &= i(\omega, E_g) \qquad \omega = 0 \text{ or } 3. \end{aligned} \tag{42}$$

Thus, in particular, $I_{\omega 1}$ describes the isotropic coupling. In the case of nearest and next-nearest neighbours ($\omega = 1$ or 2) we have for the diagonal polarisations ($\xi = 1$)

$$\begin{aligned} I_{\omega 11} &= (I(\omega, A_{1g}) - I(\omega, E_g))/3 \\ I_{\omega 12} &= (2I(\omega, A_{1g}) + I(\omega, E_g))/3 \qquad \omega = 1 \text{ or } 2. \end{aligned} \tag{43}$$

Thus the isotropic coupling $I(\omega, A_{1g})$ is not a basic, but a composite, invariant in the fundamental scheme, i.e.

$$I(\omega, A_{1g}) = I_{\omega 11} + I_{\omega 12}. \tag{44}$$

For the off-diagonal polarisation ($\xi = 2$) we have

$$\begin{aligned} I_{\omega 21} &= [I(\omega, T_{1g}) - I(\omega, T_{2g}, \{1^2\})]/2 \\ I_{\omega 22} &= [I(\omega, T_{1g}) + I(\omega, T_{2g}, \{1^2\})]/2 \\ I_{\omega 23} &= I(\omega, T_{2g}, \{2\}) \qquad \omega = 1 \text{ or } 2. \end{aligned} \tag{45}$$

The first two invariants in (45) correspond to mutally inverse double cosets ($g_\eta = C_{4y}$ and C_{4y}^{-1} for $\omega = 1$, and similarly $g_\eta = S_{4y}$ and S_{4y}^{-1} for $\omega = 2$—cf table 4), and thus involve antisymmetric parts.

The transformation between fundamental and irreducible scheme is given in table 6. Invariants in the irreducible scheme, involve the linear structure of the configuration space, and look thus more complicated than those in the fundamental scheme.

Let us take as an example

$$I_{A_{2u}T_{1u}}(T_{2g}) = \frac{1}{8\sqrt{6}} \sum_{\substack{\omega \in \{0,3\} \\ \omega' \in \{1,2\}}} (I_{\omega 2} - I_{\omega' 23} - I_{\omega' 21} + I_{\omega' 22}). \tag{46}$$

Table 6. The matrix of the transformation between bilinear invariants in fundamental and irreducible schemes. The table contains the first quarter of the matrix. The whole matrix is of the form $\begin{pmatrix} A & A \\ A & -A \end{pmatrix}$, where A stands for the first quarter. Rows and columns of the whole matrix are labelled according to tables 2 and 4, respectively. (Each row of the matrix is to be multiplied by a constant c.)

			ω	0		2				
			ξ	1	2	1		2		
Γ	Λ	Λ'	c \ ψ	—	—	1	2	1	2	3
A_{1g}	T_{1u}	T_{1u}	$1/24$	1	1	1	1	1	1	1
E_g	T_{1u}	T_{1u}	$1/24\sqrt{2}$	2	-1	2	2	-1	-1	-1
T_{1g}	T_{1u}	T_{1u}	$1/16\sqrt{3}$	2	-1	-2	0	1	1	-1
T_{2g}	A_{2u}	A_{2u}	$1/8\sqrt{3}$	1	0	1	-1	0	0	0
		T_{1u}	$1/8\sqrt{6}$	0	1	0	0	-1	1	-1
	T_{1u}	A_{2u}	$1/8\sqrt{6}$	0	1	0	0	1	-1	-1
		T_{1u}	$1/16\sqrt{3}$	2	1	-2	0	-1	-1	1

This invariant, which plays the role of the hydridisation term in the secular equation, is the sum of the symmetric (first two terms in brackets) and antisymmetric (the two remaining terms) parts.

4. Final remarks and conclusions

We have applied the Racah-Wigner approach to the determination of the harmonic potential of a cube in the most general form admissible by the symmetry of the octahedral group O_h. The corresponding twelve independent elasticity parameters are classified in the irreducible, transitive, and fundamental schemes. This classification is the most transparent in the fundamental scheme (tables 4 and 5 and figures 4-6), since the coupling between any two elements of the fundamental basis $b_{\text{fund}}(L)$ in the configuration space L of the cube is given by a single parameter, which is the same within each orbit of the action of the group O_h on the cartesian square $b_{\text{fund}}(L) \times b_{\text{fund}}(L)$ of all elements of the potential matrix. Each such an orbit exhibits a simple geometric meaning as an interplay of positional and polarisational effects.

Extension of the Racah-Wigner approach to multicentre systems is associated with a factorisation of the configuration space into positional and polarisational factors which allows us to perform appropriate recouplings. It is, however, worth noting that such a factorisation is not canonical, even if it is consistent with the action of the symmetry group. We have shown here an example of the cube that adequate account of degree an arbitrariness of choice of a basis in the configuration space, along a covariant Racah-Wigner approach involving all geometric selection rules, is provided by the fibre structure of the configuration space. This structure, which at first sight introduces only a useless obfuscation of an apparently clear situation described by the factorisation (1) of the configuration space, allows us in fact to recognise that the positional factor in (1), (4), (14) or (16) has 'absolute' meaning, i.e. it is associated with the base $\tilde{R}$ of the bundle E, whereas the polarisation factor is 'relative', depending upon the choice of 'reference frame'—the set of basis vectors in each fibre W_r, $r \in \tilde{R}$. Actually, polarisation reps V and V', which enter respectively the transitive and fundamental schemes of classification of elasticity parameters, are mutally inequivalent, as a result of relativity on fibres.

It is worth mentioning that the factorisation (1) is associated with the cartesian basis (5), resulting from the parallel translation of a standard basis to each node. This basis is convenient in cases when the cube is a part of a crystal with translational symmetry. On the other hand, the factorisation (16) of the fundamental basis (11) is the best adapted to the octahedral symmetry itself. Both bases differ mutually by a sign modulation which imposes the linear inequivalence of polarisation reps V and V'.

Classification schemes presented here allow us to discuss the relationship between elasticity parameters and the spectrum of vibrations. In particular, the irreducible scheme reveals explicitly the connection between the hybridisation of modes Γ with non-trivial multiplicities $m(M, \Gamma) = 2$, and a nonlinear dependence of the corresponding eigenfrequencies upon the elasticity parameters.

The irreducible scheme of classification is best adapted to the secular eigenproblem, whereas transitive and fundamental ones allow us to single out the elementary 'fundamental' couplings, due to the powerful Mackey theorem.

The approach presented here can be extended to other clusters and other fibres, e.g. to study electron correlations or electron-phonon coupling in crystals. It is, however, important to note that the main advantage of the present case, i.e. the fundamental basis (11) with the closure property (12), does not exist in general. We hope to extend this concept to cover a larger class of physically significant cases. In any case, a systematic Racah–Wigner-type approach to multicentre systems provides an insight into the nature of many-body interactions.

Appendix 1. Notation of elements of the octahedral group

We assume that the axes x, y, z of the cartesian coordinate system coincide with the fourfold axes of the group $\mathrm{O_h}$, and the nodes of the cube are labelled according to figure 1. Then the elements of the group $\mathrm{O}\lhd\mathrm{O_h}$ (of rotations of a cube) are denoted as follows: the fourfold axes C_{4x}, C_{4y}^{-1}, etc., threefold C_{3A}, C_{3B}^{-1}, etc, twofold C_{2x}, C_{2y}, C_{2z}, and u_{AB}, u_{AC}, u_{BD} etc, so that the sense of a rotation is associated with the right-hand rule with the positive direction of a cartesian coordinate axis for the case of fourfold rotations, with the direction $A'A$, $B'B$, etc for the threefold axes, and with the twofold axis u_{AB} passing through the centres of the edges AB' and $A'B$. The mirror rotations are determined by the space inversion, so that e.g. $\sigma_x = IC_{2x}$, $\sigma_{AB} = Iu_{AB}$ are reflection planes, and again $S_{6A}^{-1} = IC_{3A}$, $S_{4x} = IC_{4x}^{-1}$ are the appropriate mirror rotations.

Let $\tilde{R}(\mathrm{G}:\mathrm{H})$ denote an orbit of a transitive rep of a group G, determined by the stability subgroup $\mathrm{H}\subset\mathrm{G}$ (with an accuracy up to an inner authomorphism of G—cf Lulek *et al* (1985a) for details). Using this notation, we are able to present concisely some geometric and combinational aspects of our constructions on the cluster $\tilde{R}$. In particular,

$$\tilde{R} \equiv \tilde{R}(\mathrm{O_h}:\mathrm{C}_{3\mathrm{v}}^{A}) = \{A, B, C, D, A', B', C', D'\} \tag{A1.1}$$

where

$$\mathrm{C}_{3\mathrm{v}}^{A} = \{E, C_{3A}, C_{3A}^{-1}, \sigma_{BC}, \sigma_{BD}, \sigma_{CD}\} \subset \mathrm{O_h} \tag{A1.2}$$

is the stability group of the node $A\in\tilde{R}$, and

$$\tilde{V} \equiv \tilde{R}(\mathrm{O_h}:\mathrm{D}_{4\mathrm{h}}^{x}) = \{x, y, z\} \tag{A1.3}$$

where $\mathrm{D}_{4\mathrm{h}}^{x}$ is the dihedral group with the fourfold x axis. Orbits (A1.1) and (A1.3) serve as labels of bases of the positional space B and the polarisation space W, respectively.

Decomposition of the manifold of the group $\mathrm{O_h}$ into left cosets with respect to the subgroup

$$\mathrm{C}_{1\mathrm{h}}' = \{E, \sigma_{CD}\} \tag{A1.4}$$

is given in table 1. This decomposition clearly exhibits the factorisation (16): rows and columns of this table constitute, respectively, left cosets of transitive factors $R^{\mathrm{O_h}:\mathrm{C}_{3\mathrm{v}}^{A}}$ and $R^{\mathrm{O_h}:\mathrm{D}_{4\mathrm{h}}^{x}}$ of the right-hand side of (16).

Appendix 2. The Mackey theorem

We formulate here the Mackey theorem (Altmann 1977) in a form adapted to an arbitrary pair of transitive reps, $R^{\mathrm{G}:\mathrm{H}}$ and $R^{\mathrm{G}:\mathrm{D}}$, of the group G, with stability groups H and D, respectively (Lulek *et al* 1985b). The direct product $R^{\mathrm{G}:\mathrm{H}}\times R^{\mathrm{G}:\mathrm{D}}$ of these reps acting on the cartesian product $\tilde{R}(\mathrm{G}:\mathrm{H})\times\tilde{R}(\mathrm{G}:\mathrm{D})$ of the corresponding orbits, in general is not transitive, but it decomposes into transitive reps according to the formula

$$R^{\mathrm{G:H}}\times R^{\mathrm{G:D}}\simeq\sum_{\omega\in\Omega(\mathrm{D,H})}R^{\mathrm{G:L}_\omega} \qquad (\mathrm{A}2.1)$$

where $\Omega(\mathrm{D},\mathrm{H})$ is the set of labels of double cosets of the group G with respect to the oriented pair (D, H) of its subgroups, related to the decomposition

$$G=\bigcup_{\omega\in\Omega(\mathrm{D,H})}\mathrm{D}g_\omega\mathrm{H} \qquad (\mathrm{A}2.2)$$

where g_ω is the double coset representative, and

$$L_\omega=\mathrm{D}\cap g_\omega\mathrm{H}g_\omega^{-1} \qquad \omega\in\Omega(\mathrm{D},\mathrm{H}) \qquad (\mathrm{A}2.3)$$

is the stability group of the resultant transitive rep $R^{\mathrm{G:L}_\omega}$. Clearly, A2.1 provides the transitive analogue for the Clebsch-Gordan decomposition of irreps. The orbit of the resultant transitive rep $R^{\mathrm{G:L}_\omega}$ consists of all such pairs (r_1, r_2), $r_1\in\tilde{R}(\mathrm{G}:\mathrm{H})$, $r_2\in R(\mathrm{G}:\mathrm{D})$ which are equivalent under the action of the group G (e.g. geometrically equivalent pairs of nearest, next-nearest etc, neighbours).

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