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# Molien generating functions and integrity bases for the action of the $SO(3)$ and $O(3)$ groups on a set of vectors

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**Abstract.** A method for the construction of generalized integrity bases of invariant and covariant polynomials built from a set of three dimensional vectors under the  $SO(3)$  and  $O(3)$  symmetry is presented. The work is a follow-up to our previous article [G. Dhont and B. I. Zhilinskií 2013. The action of the orthogonal group on planar vectors: invariants, covariants and syzygies. *J. Phys. A: Math. Theor.*, **46**, 455202] that dealt with a set of two dimensional vectors under the action of the  $SO(2)$  and  $O(2)$  groups. The Molien generating functions for invariant rings and covariant modules under  $SO(3)$  are derived. Their expressions as one rational function are a useful guide to build integrity bases for rings of invariants and free modules of covariants. We show how to deal with the Molien generating functions for non-free modules of covariants and how to take advantage of their expressions as a sum of rational functions to obtain generalized integrity bases. The  $O(3)$  invariants and covariants bases are deduced from the  $SO(3)$  ones. The results can be used in quantum chemistry to describe the potential energy or multipole moment hypersurfaces of molecules. In particular, the generalized integrity bases that are useful for the electric and magnetic quadrupole moment hypersurfaces of tetratomic molecules are given for the first time.

*Keywords:* orthogonal group, invariant theory, integrity basis, free module, non-free module, dipole moment, quadrupole moment

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## 1. Introduction

The concept of symmetry pervades many branches of physics and has been formalized by the mathematical framework of group theory [1, 2]. In particular, the irreducible representations of a compact Lie group  $G$  classify the different ways a set of objects can transform among themselves under the action of the elements  $g$  of the group. We define as invariant an object that is unmodified under the elements of the symmetry group  $G$ , *i.e.* that follows the one dimensional totally symmetric representation  $\Gamma_0$  of the group. An irreducible representation  $\Gamma \neq \Gamma_0$  of dimension  $n$  corresponds to a set of  $n$   $\Gamma$ -covariants which transform according to the  $n \times n$  representation matrix of the element  $g$ . Group theory gives methods to build symmetry-adapted objects, however the projector and shift operators [3] are inefficient tools because the resulting expressions are sometimes redundant or null. Invariant theory [4, 5, 6, 7] takes care of the algebraic structure in order to produce powerful methods to construct invariants and covariants.

Concretely, given a representation  $V$  of  $G$  (associated for example to  $SO(3)$  acting on the coordinates of a set of vectors), the action of  $G$  extends to the corresponding algebra  $A$  of polynomials. An integrity basis (for invariants) is then a set of polynomials that generates the algebra  $A_0$  of invariant polynomials. This algebra  $A_0$  is Cohen–Macaulay, that is, it is a free module over a polynomial subalgebra: it can be written  $A_0 = \bigoplus_{i=1}^n \mathbf{C}[g_1, \dots, g_m] \cdot f_i$ . The  $g_i$  are called primary invariants and form an h.s.o.p. (homogeneous system of parameters), the  $f_i$  are called secondary invariants. The decomposition and the h.s.o.p. are not unique but the choice of the h.s.o.p. does not matter for our forthcoming considerations. To each irreducible representation  $\Gamma \neq \Gamma_0$  is associated a finitely generated  $A_0$ -module of covariants,  $A_\Gamma$ . If it is free over  $\mathbf{C}[g_1, \dots, g_m]$ ,  $A_\Gamma$  is called a Cohen–Macaulay module. When  $G$  is finite, all the  $A_\Gamma$  are Cohen–Macaulay and the notion of integrity basis for covariants generalizes immediately: it splits into the h.s.o.p. (invariant polynomials) and a family of covariant polynomials that generate freely  $A_\Gamma$  as a  $\mathbf{C}[g_1, \dots, g_m]$ -module.

This concept of integrity basis has been leveraged in molecular physics [8, 9, 10, 11, 12, 13, 14], solid state physics [15, 16], physics of deformable bodies [17, 18, 19, 20], high energy physics [21], quantum information [22, 23, 24] and general multivariate interpolation [25]. In recent years, the combination of computer algebraic techniques with artificial intelligence tools to derive primary invariants and fit symmetry-adapted quantum mechanical quantities has been a field of particularly active research [26, 27, 28].

When  $G$  is continuous, the Cohen–Macaulay property does not hold in general for  $A_\Gamma$  [29]. The existence of such non-free modules over an h.s.o.p. is one of the noteworthy features unseen when dealing with finite point groups that we want to point out; it requires the introduction of the notion of *generalized integrity basis*. We investigate this phenomenon in detail in the present article.

The present work on the invariants and covariants built from the coordinates of  $N$

vectors of the three-dimensional space under the SO(3) and O(3) groups stems from three previous articles [30, 31, 32], where the action of the SO(2) and O(2) groups on the coordinates of a set of vectors of the plane was considered. These papers put forward the problem we mentioned of dealing with non-free modules of covariants over an h.s.o.p. that occasionally arises when working with continuous groups. We propose integrity bases for the ring of invariants and free modules of covariants under the SO(3) group and develop generalized integrity bases in the case of non-free modules of covariants over an h.s.o.p.. As in our previous paper [30], the Molien function [33] plays a central role in the conception of the integrity bases and their generalization.

The article is organized as follows: in Section 2, explicit expressions of the Molien generating functions are provided up to  $N = 5$  vectors. In Section 3, relying on these expressions, we formulate two conjectures of practical importance to derive useful bases in the perspective of fitting symmetry-adapted quantum mechanical quantities. In Section 4, we explain a general method to actually build generalized integrity bases and apply it to the non trivial case of 3 vectors. We provide results for both SO(3) and O(3) groups. However, we argue that despite the fact that molecular systems can be considered as invariant under spatial inversion, it is usually more useful to fit molecular potential energy and multipole moment hypersurfaces with plain SO(3) symmetry. We conclude with our prospects for this work. In particular, the generalized integrity basis explicitly constructed for the first time for the non-free ( $L = 2$ )-covariant module in the case of  $N = 3$  vectors will be useful to represent electric and magnetic quadrupole moment hypersurfaces of tetratomic molecules.

## 2. Construction of the Molien generating function for $N$ vectors

### 2.1. General Molien generating function

In a polyatomic molecule with  $N + 1$  nuclei, it is always possible to define  $N$  relative vectors that are invariant under any translation [34]. These relative vectors can be Jacobi vectors, Radau vectors [35] or simply differences of the vector positions of two nuclei,  $\vec{r}_i - \vec{r}_j$ . From now on, by *vector* we mean one of these relative vectors.

In an active transformation, a rotation of angle  $\omega$  around a rotation axis whose direction is given by the unit vector  $\hat{n} = n_x \hat{e}_x + n_y \hat{e}_y + n_z \hat{e}_z$  transforms the vector  $\vec{r} = x \hat{e}_x + y \hat{e}_y + z \hat{e}_z$  into the vector  $\vec{r}' = x' \hat{e}_x + y' \hat{e}_y + z' \hat{e}_z$ . The initial and final coordinates are related by [36]:

$$\begin{pmatrix} x' \\ y' \\ z' \end{pmatrix} = R(\omega, \hat{n}) \begin{pmatrix} x \\ y \\ z \end{pmatrix}, \quad R(\omega, \hat{n}) = 1_3 + N \sin \omega + N^2 (1 - \cos \omega),$$

where  $1_3$  is the  $3 \times 3$  identity matrix and  $N$  is the  $3 \times 3$  matrix defined as:

$$N = \begin{pmatrix} 0 & -n_z & n_y \\ n_z & 0 & -n_x \\ -n_y & n_x & 0 \end{pmatrix}.$$

Using spherical coordinates to give the orientation of the rotation axis  $\hat{n}$  gives  $n_x = \sin \theta \cos \varphi$ ,  $n_y = \sin \theta \sin \varphi$  and  $n_z = \cos \theta$ .

Each value of the angle of rotation  $\omega \in [0, \pi]$  defines an equivalence class of the SO(3) group, with all the rotations of the same angle but different rotation axes belonging to the same class. The character of the class for the  $(2L + 1)$ -dimensional irreducible representation  $(L)$ ,  $L \in \mathbb{N}$ , of SO(3) is given by [37]:

$$\chi^{(L)}(\omega) = \frac{\sin \left[ (2L + 1) \frac{\omega}{2} \right]}{\sin \frac{\omega}{2}}.$$

The 3 coordinates of one vector span the irreducible representation (1) of SO(3) and the  $3N$  coordinates of the  $N$  vectors span the reducible representation  $\underbrace{(1) \oplus \cdots \oplus (1)}_{N \text{ times}}$ , called

from now on the initial representation. The group SO(3) acts diagonally on the direct sum of the various symmetric tensor powers of this representation or, equivalently, on polynomials in the coordinates. The Molien generating function associated to a given irreducible representation  $(L)$  (called the final representation) is then the formal power series in a variable  $\lambda$  where the coefficient of  $\lambda^n$  is the multiplicity of  $(L)$  in the SO(3)-module of degree  $n$  polynomials in the coordinates.

The Molien generating function for computing the number of linearly independent invariants (*i.e.* “(0)-covariants”) or  $(L)$ -covariants ( $L > 0$ ) of a given degree built from  $N$  vectors is given by

$$g_{\text{SO}(3)}(N, L; \lambda) = \frac{1}{2\pi^2} \int_0^\pi \int_0^{2\pi} \int_0^\pi \frac{\chi^{(L)}(\omega)^*}{\det(I - \lambda D(\omega, \hat{n}))} \sin \theta d\theta d\varphi \sin^2 \frac{\omega}{2} d\omega. \quad (1)$$

$D(\omega, \hat{n})$  is the  $3N \times 3N$  block matrix representation of the rotation operation:

$$D(\omega, \hat{n}) = \begin{pmatrix} R(\omega, \hat{n}) & 0 & \cdots & 0 \\ 0 & R(\omega, \hat{n}) & \cdots & 0 \\ \vdots & \vdots & \ddots & 0 \\ 0 & 0 & \cdots & R(\omega, \hat{n}) \end{pmatrix},$$

and one easily finds that

$$\det(I - \lambda D(\omega, \hat{n})) = [(1 - \lambda)(1 - 2\lambda \cos \omega + \lambda^2)]^N.$$

Equation (1) then reduces to

$$g_{\text{SO}(3)}(N, L; \lambda) = \frac{2}{\pi} \frac{1}{(1 - \lambda)^N} \int_0^\pi \frac{\sin \left[ (2L + 1) \frac{\omega}{2} \right] \sin \frac{\omega}{2}}{(1 - 2\lambda \cos \omega + \lambda^2)^N} d\omega. \quad (2)$$

The integral in (2) can be evaluated by the use of the product-to-sum identity  $\sin[(2L+1)\omega/2]\sin(\omega/2) = \{\cos(L\omega) - \cos[(L+1)\omega]\}/2$  and the tabulated formula [38]:

$$\begin{aligned} & \int_0^\pi \frac{\cos nx \, dx}{(1 - 2a \cos x + a^2)^m} \\ &= \frac{a^{2m+n-2}\pi}{(1-a^2)^{2m-1}} \sum_{k=0}^{m-1} \binom{m+n-1}{k} \binom{2m-k-2}{m-1} \left(\frac{1-a^2}{a^2}\right)^k, \quad a^2 < 1. \end{aligned} \quad (3)$$

Collins and Parsons [39] determined the Molien generating function for the invariants with a parametrization of the rotations based on Euler angles. Our approach with the rotation angle  $\omega$  and rotation axis  $\hat{n}$ , while giving the same result for the invariants, is more amenable to a generalization for covariants because our parametrization gives a straightforward integral over the single variable  $\omega$  while calculations would not be so direct using the Euler angles.

### 2.2. Relationship with the Molien generating functions of SO(2)

The Molien generating function for SO(3) can be written as the difference between Molien generating functions for SO(2) that were determined in [30]:

$$\begin{aligned} g_{\text{SO}(3)}(N, L; \lambda) &= \frac{1}{(1-\lambda)^N} (g_{\text{SO}(2)}(N, L; \lambda) - g_{\text{SO}(2)}(N, L+1; \lambda)), \\ &= \frac{1}{(1-\lambda)^N} \left\{ \frac{1}{\pi} \int_0^\pi \frac{\cos(L\omega) \, d\omega}{(1-2\lambda \cos \omega + \lambda^2)^N} - \frac{1}{\pi} \int_0^\pi \frac{\cos[(L+1)\omega] \, d\omega}{(1-2\lambda \cos \omega + \lambda^2)^N} \right\}, \end{aligned} \quad (4)$$

where  $g_{\text{SO}(2)}(N, L; \lambda)$  is the generating function for the number of covariants with irreducible representation ( $L$ ) of SO(2) that can be built from  $N$  two dimensional vectors.

### 2.3. Molien generating function for one vector

The three coordinates  $\{x, y, z\}$  of one vector span an irreducible three-dimensional representation: (1). Combining the general Molien function formula (4) with integral (3) for the case  $N = 1$ , we obtain

$$g_{\text{SO}(3)}(1, L; \lambda) = \frac{\lambda^L}{1-\lambda^2}. \quad (5)$$

The formal expansion of this generating function gives  $\lambda^L + \lambda^{L+2} + \dots$ , which means that we can build one set of  $2L+1$  ( $L$ )-covariants of degree  $p \geq L$  when  $p-L$  is even.

As a verification, we can sum over all the possible representations  $L$  the numbers of ( $L$ )-covariants. Remembering that we have for a given  $L$  a set of  $2L+1$  polynomials that transform among themselves, we get

$$\sum_{L=0}^{\infty} (2L+1) g_{\text{SO}(3)}(1, L; \lambda) = \frac{1}{(1-\lambda)^3} = 1 + 3\lambda + 6\lambda^2 + 10\lambda^3 + \dots$$

where the coefficients 1, 3, 6, 10 are respectively the total number of polynomials in three variables of degree 0, 1, 2, 3 without any symmetry consideration. This result generalizes to any number of vectors:

$$\sum_{L=0}^{\infty} (2L+1) g_{SO(3)}(N, L; \lambda) = \frac{1}{(1-\lambda)^{3N}} = \sum_{k=0}^{\infty} \binom{3N+k-1}{k} \lambda^k,$$

and means that the weighted sum of the Molien generating functions  $g_{SO(3)}(N, L; \lambda)$  over all the possible final representations reduces to the Molien generating function for the problem when the symmetry is not taken into account.

#### 2.4. Recursive formula for two or more vectors

The Molien generating function for  $N \geq 2$  can be determined from formula (2). There is however an other approach, which gives more insight in the forthcoming construction of the integrity bases. According to the triangular conditions of the theory of angular momentum, the coupling of a set of  $(L_1)$ -covariants with a set of  $(L_2)$ -covariants generates one set of  $(L)$ -covariants, with  $|L_1 - L_2| \leq L \leq L_1 + L_2$ . Correspondingly the Molien generating function for  $N_1 + N_2$  vectors and final representation  $(L)$  can be computed from the Molien generating functions for  $N_1$  and  $N_2$  vectors, see equation (43) of [7]:

$$g_{SO(3)}(N_1 + N_2, L; \lambda_1, \lambda_2) = \sum_{L_1=0}^{\infty} \sum_{L_2=0}^{\infty} \Delta(L_1, L_2, L) g_{SO(3)}(N_1, L_1; \lambda_1) g_{SO(3)}(N_2, L_2; \lambda_2), \quad (6)$$

where  $\Delta(L_1, L_2, L) = 1$  if  $|L_1 - L_2| \leq L \leq L_1 + L_2$  and 0 otherwise.

#### 2.5. Molien generating function for two vectors

The six coordinates  $\{\mathbf{x}_1, \mathbf{x}_2\}$  of two vectors span the reducible six-dimensional representation  $(1) \oplus (1)$ . Rather than making use of Eqs. (4) and (3), the generating functions for two vectors can be deduced by coupling, according to (6), the generating functions for one vector obtained in (5). Distinguishing the two Molien generating functions for one vector by their variables  $\lambda_1$  and  $\lambda_2$ , one obtains:

$$g_{SO(3)}(2, L; \lambda_1, \lambda_2) = \frac{\sum_{i=0}^L \lambda_1^i \lambda_2^{L-i} + \lambda_1 \lambda_2 \sum_{i=0}^{L-1} \lambda_1^i \lambda_2^{L-i-1}}{(1-\lambda_1^2)(1-\lambda_2^2)(1-\lambda_1 \lambda_2)}, \quad (7)$$

with the convention that the second term in the numerator is zero for  $L = 0$ . If we do not distinguish between the two vectors, we can set  $\lambda_1 = \lambda_2 = \lambda$  and get:

$$g_{SO(3)}(2, L; \lambda) = \frac{(L+1)\lambda^L + L\lambda^{L+1}}{(1-\lambda^2)^3}. \quad (8)$$

### 2.6. Molien generating function for three vectors

The reducible initial representation  $(1) \oplus (1) \oplus (1)$  contains nine variables. Again, the Molien generating functions for three vectors can be obtained by coupling the generating function for one vector and the generating function for two vectors, according to (6). One obtains the following expression:

$$g_{\text{SO}(3)}^{(a)}(3, L; \lambda) = \frac{\mathcal{N}_{\text{SO}(3)}^{(a)}(3, L; \lambda)}{(1 - \lambda^2)^6}, \quad (9)$$

$$\begin{aligned} \mathcal{N}_{\text{SO}(3)}^{(a)}(3, L; \lambda) &= \frac{(L+2)(L+1)}{2} \lambda^L + (L+2)L\lambda^{L+1} \\ &\quad - (L+1)(L-1)\lambda^{L+3} - \frac{L(L-1)}{2} \lambda^{L+4}. \end{aligned} \quad (10)$$

The coefficients in the numerator (10) are all non-negative if  $L = 0$  or  $L = 1$ . Negative coefficients appear for  $L \geq 2$ . This means that the symbolic interpretation of the numerator coefficients as the numbers of covariants of each degree in the integrity basis does not hold anymore. Instead, the first negative coefficient indicates the number of syzygies of lowest degree and diagnoses the fact that the  $(L)$ -component of the ring of polynomials over the ring of primary invariants is not a free module.

However, the Molien generating function can be recast in the form (11) suitable for a symbolic interpretation in term of a generalized integrity basis (whose notion will be introduced later):

$$\begin{aligned} g_{\text{SO}(3)}^{(b)}(3, L; \lambda) &= \frac{(2L+1)\lambda^L + (2L+1)\lambda^{L+1}}{(1 - \lambda^2)^6} \\ &\quad + \frac{\frac{L(L-1)}{2}\lambda^L + (L+1)(L-1)\lambda^{L+1} + \frac{L(L-1)}{2}\lambda^{L+2}}{(1 - \lambda^2)^5}, \end{aligned} \quad (11)$$

where all the coefficients in the numerators are positive coefficients for  $L \geq 2$ . The generating functions suitable for a symbolic interpretation in term of a generalized integrity basis are given in Table 1 up to  $L = 6$ .

We have proceeded in a similar fashion to obtain the generating functions for four vectors, see Appendix A.1 and five vectors, see Appendix A.2. Any number of vectors can be achieved by recursion.

### 2.7. Extension to the $\text{O}(3)$ group

The matrix representation of the inversion operation for the problem of  $N$  vectors is  $-\mathbf{1}_{3N}$ , where  $\mathbf{1}_{3N}$  is the  $3N \times 3N$  identity matrix. Denoting the irreducible representations of  $\text{O}(3)$  by  $(L^\epsilon)$ ,  $L \in \mathbb{N}$ ,  $\epsilon = \pm 1$ , the corresponding Molien functions for  $N$  vectors under the  $\text{O}(3)$  group are obtained through:

$$g_{\text{O}(3)}(N, L^\epsilon; \lambda) = \frac{1}{2} [g_{\text{SO}(3)}(N, L; \lambda) + \epsilon g_{\text{SO}(3)}(N, L; -\lambda)].$$

**Table 1.** Expressions for  $0 \leq L \leq 6$  and three vectors of the Molien functions that admit a symbolic interpretation in term of (generalized) integrity bases.

$x$	$L$	$g_{\text{SO}(3)}^{(x)}(3, L; \lambda)$
$a$	0	$\frac{1+\lambda^3}{(1-\lambda^2)^6}$
$a$ or $b$	1	$\frac{3\lambda+3\lambda^2}{(1-\lambda^2)^6}$
$b$	2	$\frac{5\lambda^2+5\lambda^3}{(1-\lambda^2)^6} + \frac{\lambda^2+3\lambda^3+\lambda^4}{(1-\lambda^2)^5}$
$b$	3	$\frac{7\lambda^3+7\lambda^4}{(1-\lambda^2)^6} + \frac{3\lambda^3+8\lambda^4+3\lambda^5}{(1-\lambda^2)^5}$
$b$	4	$\frac{9\lambda^4+9\lambda^5}{(1-\lambda^2)^6} + \frac{6\lambda^4+15\lambda^5+6\lambda^6}{(1-\lambda^2)^5}$
$b$	5	$\frac{11\lambda^5+11\lambda^6}{(1-\lambda^2)^6} + \frac{10\lambda^5+24\lambda^6+10\lambda^7}{(1-\lambda^2)^5}$
$b$	6	$\frac{13\lambda^6+13\lambda^7}{(1-\lambda^2)^6} + \frac{15\lambda^6+35\lambda^7+15\lambda^8}{(1-\lambda^2)^5}$
$b$	$\vdots$	$\vdots$

### 3. Two conjectures around the Molien generating function

#### 3.1. First conjecture

Based on the results of section 2, we conjecture that for any number  $N$  of three dimensional vectors and any final representation ( $L$ ) of  $\text{SO}(3)$ , there exists a positive integer  $l_{\max}(N, L)$  with  $1 \leq l_{\max}(N, L) \leq 3N - 2$ , such that the Molien function  $g$  can be cast in the form of a sum of rational functions:

$$g_{\text{SO}(3)}(N, L; \lambda) = \sum_{l=1}^{l_{\max}(N, L)} \frac{\mathcal{N}_{N, L}^l(\lambda)}{(1 - \lambda^2)^{3N-2-l}}, \quad (12)$$

with the  $l_{\max}(N, L)$  numerator polynomials  $\mathcal{N}_{N, L}^l(\lambda) = \sum_{n=0}^{n_{\max}(N, L, l)} c_{N, L}^{l, n} \lambda^{L+n}$  having only non-negative coefficients  $c_{N, L}^{l, n}$ . The exponents on  $\lambda$  in the numerator polynomials start at  $L$ , since ( $L$ )-covariants built from vectors are at least of total degree  $L$ .

The case  $L = 0$  corresponds to the ring of invariant polynomials  $A_0$  under the action of the compact  $\text{SO}(3)$  group. We have seen that it is a finitely generated, free module over the algebra  $\mathbb{R}[g_1, \dots, g_m]$  for any maximal set of homogeneous, algebraically independent, invariant polynomials,  $\{g_1, \dots, g_m\}$ , the primary invariants [40, 41]. That is to say, there exists a finite set of secondary invariants  $\{f_1, \dots, f_p\}$  such that,

$$A_0 = \mathbb{R}[g_1, \dots, g_m]f_1 \oplus \dots \oplus \mathbb{R}[g_1, \dots, g_m]f_p. \quad (13)$$

This relationship constitutes an ‘‘Hironaka decomposition’’ of  $A_0$ . The conjecture holds in that case as a consequence of [42]: the Molien function for invariant polynomials can be cast in the form (12) with a single rational function, *i.e.*  $l_{\max}(N, 0) = 1$ . This corresponds to the fact that one can choose a h.s.o.p. with  $3N - 3$  primary or denominator invariant polynomials of degree 2 and  $c_{N, 0}^{1, n}$  secondary or numerator invariant polynomials of degree  $L + n$  with  $0 \leq n \leq n_{\max}(N, 0, 1)$ .

If the module of  $(L)$ -covariants ( $L > 0$ ) is Cohen–Macaulay, that is, free over an arbitrary h.s.o.p., the situation is similar to the ring of invariant polynomials: the form (12) is then a single rational function whose denominator can be chosen to be the same as the one for  $L = 0$ . It is suited to an interpretation in term of integrity basis as the terms of the numerator correspond to the secondary covariants, and the conjecture holds again.

An interesting issue occurs when the form (12) has  $l_{\max}(N, L) > 1$ . It corresponds to the case where the module of  $(L)$ -covariants is not free over an h.s.o.p., which is known to happen [43]. We already proposed to interpret this situation by introducing generalized integrity bases in our previous work on  $SO(2)$  [30]. The non-free module of  $(L)$ -covariants is then a sum of  $l_{\max}(N, L)$  submodules. Each of these submodules corresponds to a  $l^{\text{th}}$  rational function,  $1 \leq l \leq l_{\max}(N, L)$ , which conjecturally admits a symbolic interpretation in terms of integrity basis. Each submodule is a direct sum of free modules on rings of invariants generated by  $3N - 2 - l$  primary invariants. In the cases we investigated (see the forthcoming sections), the submodule has a stronger property: it is free on a single ring of invariants (that is, the primary invariants can be chosen to be the same for all free modules in the direct sum). The number of linearly independent, secondary covariants of degree  $L + n$  generating these free modules is given by  $c_{N,L}^{l,n}$ . The family of all these data, encoded by pairs of a subset of the h.s.o.p. and a covariant polynomial defines a generalized integrity basis. Our conjecture is closely related to that of Stanley, which has a more general scope: conjecture 5.1 of Ref. [44].

In practice, in the examples treated later on, the numerators in the sum of expression (12) have been determined algorithmically for a given pair  $(N, L)$  by starting with the Molien generating function written as a single rational function with numerator  $\mathcal{N}_{SO(3)}^{(a)}(N, L; \lambda)$ . If for the value of  $L$  considered, the numerator  $\mathcal{N}_{SO(3)}^{(a)}(N, L; \lambda)$  has only non-negative coefficients, then the form is suitable for an interpretation in term of integrity basis and we can start the construction of the integrity basis. Otherwise we perform a modified polynomial long division of the numerator by  $1 - \lambda^2$ . The modification is in the halt criterion: the division process is stopped not when the degree of the remainder  $r_1$  is less than 2 as one would do in the usual polynomial division, but when the coefficients of the remainder  $r_1$  become all non-negative for the value of  $L$  considered. Then the numerator is rewritten as:

$$\mathcal{N}_{SO(3)}^{(a)}(N, L; \lambda) = r_1 + (1 - \lambda^2) q_1.$$

The remainder  $r_1$  serves as the numerator of the first ( $l = 1$ ) rational function in the right-hand side of (12). If all the coefficients in the quotient  $q_1$  are non-negative for the value of  $L$  considered, the algorithm stops and the final form of the Molien function is then:

$$g_{SO(3)}^{(b)}(N, L; \lambda) = \frac{r_1}{(1 - \lambda^2)^{3N-3}} + \frac{q_1}{(1 - \lambda^2)^{3N-4}}.$$

If at least one coefficient in the quotient  $q_1$  is negative, the modified polynomial division procedure is applied to  $q_1$ . The quotient  $q_1$  is decomposed as  $q_1 = r_2 + (1 - \lambda^2) q_2$  and

the new remainder  $r_2$  with non-negative coefficients will constitute the numerator of the second ( $l = 2$ ) rational function. The modified polynomial division is repeated until the quotient has also only non-negative coefficients. The statement that the algorithm indeed converges, is a stronger formulation of the conjecture.

For example, the single rational function (9) with its numerator (10) is suitable for  $N = 3$  and  $L \in \{0, 1\}$ . For  $L \geq 2$ , we obtain successively by the modified polynomial division of the numerator (10):

$$\begin{aligned}
& \frac{(L+2)(L+1)}{2}\lambda^L + (L+2)L\lambda^{L+1} - (L+1)(L-1)\lambda^{L+3} - \frac{L(L-1)}{2}\lambda^{L+4} \\
= & (1-\lambda^2) \left( \frac{L(L-1)}{2}\lambda^{L+2} \right) + \frac{(L+2)(L+1)}{2}\lambda^L + (L+2)L\lambda^{L+1} - \frac{L(L-1)}{2}\lambda^{L+2} - (L+1)(L-1)\lambda^{L+3} \\
= & (1-\lambda^2) \left( (L+1)(L-1)\lambda^{L+1} + \frac{L(L-1)}{2}\lambda^{L+2} \right) + \frac{(L+2)(L+1)}{2}\lambda^L + (2L+1)\lambda^{L+1} - \frac{L(L-1)}{2}\lambda^{L+2} \\
= & (1-\lambda^2) \left( \frac{L(L-1)}{2}\lambda^L + (L+1)(L-1)\lambda^{L+1} + \frac{L(L-1)}{2}\lambda^{L+2} \right) + (2L+1)\lambda^L + (2L+1)\lambda^{L+1}. \quad (14)
\end{aligned}$$

The remainder  $(2L+1)\lambda^L + (2L+1)\lambda^{L+1}$  has only non-negative coefficients and is the numerator of the first rational function we are seeking for. All the coefficients of the quotient  $\frac{L(L-1)}{2}\lambda^L + (L+1)(L-1)\lambda^{L+1} + \frac{L(L-1)}{2}\lambda^{L+2}$  are non-negative for  $L \geq 2$ . This quotient is the numerator of the second fraction. This procedure gives again the expression (11) of the Molien function written as a sum of two rational functions with non-negative coefficients in the numerators.

For  $N = 4$ , all the coefficients of the numerator  $\mathcal{N}_{\text{SO}(3)}^{(a)}(4, L; \lambda)$  of (A.2) are non-negative for  $L < 3$ . For  $L \geq 3$ , we obtain successively by the modified polynomial division of the initial numerator:

$$\begin{aligned}
& \frac{(L+3)(L+2)(L+1)}{6}\lambda^L + \frac{(L+3)(L+2)L}{2}\lambda^{L+1} + \frac{(L+3)(L+2)(L+1)}{6}\lambda^{L+2} - \frac{(L+3)(L-2)(5L+4)}{6}\lambda^{L+3} \\
& - \frac{(L+3)(L-2)(5L+1)}{6}\lambda^{L+4} + \frac{L(L-1)(L-2)}{6}\lambda^{L+5} + \frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+6} + \frac{L(L-1)(L-2)}{6}\lambda^{L+7} \\
= & (1-\lambda^2) \left( -\frac{L(L-1)(L-2)}{6}\lambda^{L+5} \right) + \frac{(L+3)(L+2)(L+1)}{6}\lambda^L + \frac{(L+3)(L+2)L}{2}\lambda^{L+1} + \frac{(L+3)(L+2)(L+1)}{6}\lambda^{L+2} \\
& - \frac{(L+3)(L-2)(5L+4)}{6}\lambda^{L+3} - \frac{(L+3)(L-2)(5L+1)}{6}\lambda^{L+4} + \frac{L(L-1)(L-2)}{3}\lambda^{L+5} + \frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+6} \\
= & (1-\lambda^2) \left( -\frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+4} - \frac{L(L-1)(L-2)}{6}\lambda^{L+5} \right) + \frac{(L+3)(L+2)(L+1)}{6}\lambda^L + \frac{(L+3)(L+2)L}{2}\lambda^{L+1} \\
& + \frac{(L+3)(L+2)(L+1)}{6}\lambda^{L+2} - \frac{(L+3)(L-2)(5L+4)}{6}\lambda^{L+3} - \frac{(L-2)(L^2+8L+3)}{3}\lambda^{L+4} + \frac{L(L-1)(L-2)}{3}\lambda^{L+5} \\
= & (1-\lambda^2) \left( -\frac{L(L-1)(L-2)}{3}\lambda^{L+3} - \frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+4} - \frac{L(L-1)(L-2)}{6}\lambda^{L+5} \right) + \frac{(L+3)(L+2)(L+1)}{6}\lambda^L \\
& + \frac{(L+3)(L+2)L}{2}\lambda^{L+1} + \frac{(L+3)(L+2)(L+1)}{6}\lambda^{L+2} - \frac{(L-2)(L^2+7L+4)}{2}\lambda^{L+3} - \frac{(L-2)(L^2+8L+3)}{3}\lambda^{L+4} \\
= & (1-\lambda^2) \left( \frac{(L-2)(L^2+8L+3)}{3}\lambda^{L+2} - \frac{L(L-1)(L-2)}{3}\lambda^{L+3} - \frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+4} - \frac{L(L-1)(L-2)}{6}\lambda^{L+5} \right) \\
& + \frac{(L+3)(L+2)(L+1)}{6}\lambda^L + \frac{(L+3)(L+2)L}{2}\lambda^{L+1} - \frac{(L^3+6L^2-37L-18)}{6}\lambda^{L+2} - \frac{(L-2)(L^2+7L+4)}{2}\lambda^{L+3} \\
= & (1-\lambda^2) \left( \frac{(L-2)(L^2+7L+4)}{2}\lambda^{L+1} + \frac{(L-2)(L^2+8L+3)}{3}\lambda^{L+2} - \frac{L(L-1)(L-2)}{3}\lambda^{L+3} - \frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+4} \right. \\
& \left. - \frac{L(L-1)(L-2)}{6}\lambda^{L+5} \right) + \frac{(L+3)(L+2)(L+1)}{6}\lambda^L + 4(2L+1)\lambda^{L+1} - \frac{(L^3+6L^2-37L-18)}{6}\lambda^{L+2} \quad (15)
\end{aligned}$$

$$\begin{aligned}
&= (1 - \lambda^2) \left( \frac{(L^3 + 6L^2 - 37L - 18)}{6} \lambda^L + \frac{(L-2)(L^2 + 7L + 4)}{2} \lambda^{L+1} + \frac{(L-2)(L^2 + 8L + 3)}{3} \lambda^{L+2} - \frac{L(L-1)(L-2)}{3} \lambda^{L+3} \right. \\
&\quad \left. - \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+4} - \frac{L(L-1)(L-2)}{6} \lambda^{L+5} \right) + 4(2L+1)\lambda^L + 4(2L+1)\lambda^{L+1}. \tag{16}
\end{aligned}$$

For  $L \in \{3, 4\}$ , the algorithm stops at (15), where all coefficients of the remainder are non-negative, whereas for larger values of  $L$  the coefficient  $-(L^3 + 6L^2 - 37L - 18)/6$  of  $\lambda^{L+2}$  is negative in the remainder of (15) and one must stop a step later at (16). Let us consider first the cases  $L = 3$  and  $L = 4$ . The quotient has negative coefficients and must be divided again by  $(1 - \lambda^2)$ ,

$$\begin{aligned}
&\frac{(L-2)(L^2 + 7L + 4)}{2} \lambda^{L+1} + \frac{(L-2)(L^2 + 8L + 3)}{3} \lambda^{L+2} - \frac{L(L-1)(L-2)}{3} \lambda^{L+3} - \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+4} - \frac{L(L-1)(L-2)}{6} \lambda^{L+5} \\
&= (1 - \lambda^2) \left( \frac{L(L-1)(L-2)}{6} \lambda^{L+3} \right) + \frac{(L-2)(L^2 + 7L + 4)}{2} \lambda^{L+1} + \frac{(L-2)(L^2 + 8L + 3)}{3} \lambda^{L+2} - \frac{L(L-1)(L-2)}{2} \lambda^{L+3} \\
&\quad - \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+4} \\
&= (1 - \lambda^2) \left( \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+2} + \frac{L(L-1)(L-2)}{6} \lambda^{L+3} \right) + \frac{(L-2)(L^2 + 7L + 4)}{2} \lambda^{L+1} - \frac{(L-2)(L^2 - 16L - 9)}{6} \lambda^{L+2} \\
&\quad - \frac{L(L-1)(L-2)}{2} \lambda^{L+3} \\
&= (1 - \lambda^2) \left( \frac{L(L-1)(L-2)}{2} \lambda^{L+1} + \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+2} + \frac{L(L-1)(L-2)}{6} \lambda^{L+3} \right) + 2(L-2)(2L+1)\lambda^{L+1} \\
&\quad - \frac{(L-2)(L^2 - 16L - 9)}{6} \lambda^{L+2}. \tag{17}
\end{aligned}$$

Both remainder and quotient have non-negative coefficients for  $L \in \{3, 4\}$ , so we stop here and obtain again the numerators of the second and third fractions of  $g_{\text{SO}(3)}^{(b)}(N, L; \lambda)$  in (A.3). For  $L > 4$ , we have to divide the quotient of (16) by  $(1 - \lambda^2)$ ,

$$\begin{aligned}
&\frac{(L^3 + 6L^2 - 37L - 18)}{6} \lambda^L + \frac{(L-2)(L^2 + 7L + 4)}{2} \lambda^{L+1} + \frac{(L-2)(L^2 + 8L + 3)}{3} \lambda^{L+2} - \frac{L(L-1)(L-2)}{3} \lambda^{L+3} \\
&\quad - \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+4} - \frac{L(L-1)(L-2)}{6} \lambda^{L+5} \\
&= (1 - \lambda^2) \left( \frac{L(L-1)(L-2)}{6} \lambda^{L+3} \right) + \frac{(L^3 + 6L^2 - 37L - 18)}{6} \lambda^L + \frac{(L-2)(L^2 + 7L + 4)}{2} \lambda^{L+1} + \frac{(L-2)(L^2 + 8L + 3)}{3} \lambda^{L+2} \\
&\quad - \frac{L(L-1)(L-2)}{2} \lambda^{L+3} - \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+4} \\
&= (1 - \lambda^2) \left( \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+2} + \frac{L(L-1)(L-2)}{6} \lambda^{L+3} \right) + \frac{(L^3 + 6L^2 - 37L - 18)}{6} \lambda^L + \frac{(L-2)(L^2 + 7L + 4)}{2} \lambda^{L+1} \\
&\quad - \frac{(L-2)(L^2 - 16L - 9)}{6} \lambda^{L+2} - \frac{L(L-1)(L-2)}{2} \lambda^{L+3} \\
&= (1 - \lambda^2) \left( \frac{L(L-1)(L-2)}{2} \lambda^{L+1} + \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+2} + \frac{L(L-1)(L-2)}{6} \lambda^{L+3} \right) + \frac{(L^3 + 6L^2 - 37L - 18)}{6} \lambda^L \\
&\quad + 2(L-2)(2L+1)\lambda^{L+1} - \frac{(L-2)(L^2 - 16L - 9)}{6} \lambda^{L+2} \tag{18}
\end{aligned}$$

$$\begin{aligned}
&= (1 - \lambda^2) \left( \frac{(L-2)(L^2 - 16L - 9)}{6} \lambda^L + \frac{L(L-1)(L-2)}{2} \lambda^{L+1} + \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+2} + \frac{L(L-1)(L-2)}{6} \lambda^{L+3} \right) \\
&\quad + 2(L-3)(2L+1)\lambda^L + 2(L-2)(2L+1)\lambda^{L+1}. \tag{19}
\end{aligned}$$

For  $5 \leq L \leq 16$  the algorithm stops at (18), when all coefficients of the remainder are non-negative. For larger values of  $L$ , the coefficient of  $\lambda^{L+2}$  is negative in the remainder

of (18) and the algorithm must continue up to (19). In both cases, the quotient has only non-negative coefficients, so the remainders and quotients of Eqs. (18) and (19) gives the numerators of the second and third fractions of  $g_{\text{SO}(3)}^{(c)}(N, L; \lambda)$  and  $g_{\text{SO}(3)}^{(d)}(N, L; \lambda)$  in Eqs. (A.4) and (A.5).

### 3.2. Second conjecture

The relevant covariant module for the electric dipole moment hypersurface corresponds to the (1) irreducible representation and will always be a free module over an h.s.o.p.. This conjecture is based on the observation that for a given  $N$ , the generating Molien function admits a symbolic interpretation in term of integrity basis with a single rational function for all  $L$  if  $N \in \{1, 2\}$  and for all  $L < N$  if  $5 \geq N > 2$ .

## 4. Construction of the integrity bases for $N$ vectors

The main goal of this section is to propose a general method to write integrity bases of SO(3) covariants, and in particular the generalized integrity bases of a non-free module. The approach takes advantage of the interpretable form of the Molien function seen in the previous sections. The simplest case of a non-free module occurs for  $N = 3$  vectors and the irreducible representation  $L = 2$ . We first provide integrity bases for the free cases corresponding to  $N \in \{1, 2\}$ ,  $L \in \{0, 1, 2\}$  and  $N = 3$ ,  $L \in \{0, 1\}$ . Then, we show how to deal with the simplest non-free case:  $N = 3$ ,  $L = 2$ . Finally, we explain how to generalize the latter construction.

### 4.1. One vector

The initial representation (1) is spanned by the three variables  $x_1, y_1, z_1$ . Equation (5) indicates that the polynomials belonging to the free module of ( $L$ )–covariants can be univoquely built from one primary invariant of degree 2 and one set of  $(2L + 1)$  secondary ( $L$ )–covariants of degree  $L$ . The primary invariant is of course the scalar product  $Q_{1,1} = x_1^2 + y_1^2 + z_1^2$ . The  $(2L + 1)$  secondary ( $L$ )–covariants are naturally chosen as the real solid harmonics  $\tilde{\mathcal{Y}}_{L,M}(x_1, y_1, z_1)$ , see Appendix B where the expressions of the real solid harmonics for  $L$  up to 3 are given.

### 4.2. Two vectors

The Molien generating function for the ring of invariants ( $L = 0$ ) is

$$g_{\text{SO}(3)}(2, 0; \lambda_1, \lambda_2) = \frac{1}{(1 - \lambda_1^2)(1 - \lambda_2^2)(1 - \lambda_1\lambda_2)}.$$

The ring of invariant is well-known [42] and the integrity basis consists in the 3 scalar products  $Q_{1,1} = x_1^2 + y_1^2 + z_1^2$ ,  $Q_{2,2} = x_2^2 + y_2^2 + z_2^2$ ,  $Q_{1,2} = x_1x_2 + y_1y_2 + z_1z_2$  as the primary polynomials of degree 2 and 1 as the secondary polynomial of degree 0. The totally invariant quantities such as potential energy hypersurfaces of triatomic molecules

can be expanded as polynomials in these three primary polynomials.

For  $L = 1$ , the Molien generating function is

$$g_{\text{SO}(3)}(2, 1; \lambda_1, \lambda_2) = \frac{\lambda_1 + \lambda_2 + \lambda_1 \lambda_2}{(1 - \lambda_1^2)(1 - \lambda_2^2)(1 - \lambda_1 \lambda_2)}.$$

The two sets of (1)–covariants of degree 1 can be taken as  $\sqrt{\frac{4\pi}{3}}\tilde{Y}_{1,M}(\mathbf{x}_i)$ ,  $i \in \{1, 2\}$ ,  $M \in \{1, 0, -1\}$ , and the set of 3 (1)–covariants of degree 2 can be chosen as their cross–product:

$$\begin{pmatrix} f_{1,1}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{1,0}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{1,-1}(\mathbf{x}_1, \mathbf{x}_2) \end{pmatrix} = \begin{pmatrix} y_1 z_2 - z_1 y_2 \\ x_1 y_2 - y_1 x_2 \\ z_1 x_2 - x_1 z_2 \end{pmatrix}.$$

(The subscripts of the  $f$ –symbols are respectively  $L$ , and  $M$ .)

The electric dipole moment function,  $\bar{\mu}^{\text{el}}$ , transforms as the irreducible representation ( $1^-$ ) of the group O(3). The Molien generating function is:

$$g_{\text{O}(3)}(2, 1^-; \lambda) = \frac{2\lambda}{(1 - \lambda^2)^3}.$$

The cross–products do not enter the integrity basis because they are invariant with respect to the inversion operation. Therefore, the components  $\mu_M^{\text{el}}$ ,  $M \in \{1, 0, -1\}$  of the electric dipole moment function of an ABC molecule have the form:

$$\begin{aligned} \mu_M^{\text{el}}(\mathbf{x}_1, \mathbf{x}_2) &= P_1^{\text{el}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times \sqrt{\frac{4\pi}{3}}\tilde{Y}_{1,M}(\mathbf{x}_1) \\ &+ P_2^{\text{el}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times \sqrt{\frac{4\pi}{3}}\tilde{Y}_{1,M}(\mathbf{x}_2), \end{aligned} \quad (20)$$

where  $P_1^{\text{el}}$  and  $P_2^{\text{el}}$  are two polynomials in the three primary invariants  $Q_{1,1}$ ,  $Q_{2,2}$ , and  $Q_{1,2}$ . Thus, only two polynomials in three variables need to be fitted with respect to quantum chemistry data to determine the dipole moment surface functions. In contrast, magnetic dipole moment function,  $\bar{\mu}^{\text{mag}}$ , transforms as the irreducible representation ( $1^+$ ) of the group O(3), so its components have the form

$$\mu_M^{\text{mag}}(\mathbf{x}_1, \mathbf{x}_2) = P_1^{\text{mag}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times f_{1,M}(\mathbf{x}_1, \mathbf{x}_2), \quad (21)$$

where  $P_1^{\text{mag}}$  is a polynomial in the three primary invariants.

For  $L = 2$ , the Molien generating function is:

$$g_{\text{SO}(3)}^{(1)}(2, 2; \lambda) = \frac{3\lambda^2 + 2\lambda^3}{(1 - \lambda^2)^3}. \quad (22)$$

The first two sets of (2)–covariants of degree 2 are chosen as  $\sqrt{\frac{4\pi}{5}}\tilde{Y}_{2,M}(\mathbf{x}_i)$ ,  $i \in \{1, 2\}$ ,  $M \in \{2, 1, 0, -1, -2\}$ . The last set of (2)–covariants of degree 2 is chosen as:

$$\begin{pmatrix} f_{2,2}^{(2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,1}^{(2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,0}^{(2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,-1}^{(2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,-2}^{(2)}(\mathbf{x}_1, \mathbf{x}_2) \end{pmatrix} := \begin{pmatrix} \frac{\sqrt{3}}{2}(x_1x_2 - y_1y_2) \\ \frac{\sqrt{3}}{2}(x_1z_2 + z_1x_2) \\ \frac{1}{2}(2z_1z_2 - x_1x_2 - y_1y_2) \\ \frac{\sqrt{3}}{2}(y_1z_2 + z_1y_2) \\ \frac{\sqrt{3}}{2}(x_1y_2 + y_1x_2) \end{pmatrix}.$$

The two sets of (2)–covariants of degree three can be constructed by substituting one set of coordinates in the last expression by the ( $L = 1$ )–cross–product covariant,

$$\begin{pmatrix} f_{2,2}^{(3,1)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,1}^{(3,1)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,0}^{(3,1)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,-1}^{(3,1)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,-2}^{(3,1)}(\mathbf{x}_1, \mathbf{x}_2) \end{pmatrix} := \begin{pmatrix} \frac{\sqrt{3}}{2}(x_1(y_1z_2 - z_1y_2) - y_1(z_1x_2 - x_1z_2)) \\ \frac{\sqrt{3}}{2}(x_1(x_1y_2 - y_1x_2) + z_1(y_1z_2 - z_1y_2)) \\ \frac{1}{2}(2z_1(x_1y_2 - y_1x_2) - x_1(y_1z_2 - z_1y_2) - y_1(z_1x_2 - x_1z_2)) \\ \frac{\sqrt{3}}{2}(y_1(x_1y_2 - y_1x_2) + z_1(z_1x_2 - x_1z_2)) \\ \frac{\sqrt{3}}{2}(x_1(z_1x_2 - x_1z_2) + y_1(y_1z_2 - z_1y_2)) \end{pmatrix},$$

and

$$\begin{pmatrix} f_{2,2}^{(3,2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,1}^{(3,2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,0}^{(3,2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,-1}^{(3,2)}(\mathbf{x}_1, \mathbf{x}_2) \\ f_{2,-2}^{(3,2)}(\mathbf{x}_1, \mathbf{x}_2) \end{pmatrix} := \begin{pmatrix} \frac{\sqrt{3}}{2}((y_1z_2 - z_1y_2)x_2 - (z_1x_2 - x_1z_2)y_2) \\ \frac{\sqrt{3}}{2}((y_1z_2 - z_1y_2)z_2 + (x_1y_2 - y_1x_2)x_2) \\ \frac{1}{2}(2(x_1y_2 - y_1x_2)z_2 - (y_1z_2 - z_1y_2)x_2 - (z_1x_2 - x_1z_2)y_2) \\ \frac{\sqrt{3}}{2}((z_1x_2 - x_1z_2)z_2 + (x_1y_2 - y_1x_2)y_2) \\ \frac{\sqrt{3}}{2}((y_1z_2 - z_1y_2)y_2 + (z_1x_2 - x_1z_2)x_2) \end{pmatrix}.$$

(The superscripts of the  $f$ –symbols are respectively the degree of the covariant and an integer index when there are several secondary covariants of the same degree.)

The electric quadrupole moment function,  $q^{\text{el}}$ , transforms as the irreducible representation ( $2^+$ ) of the group O(3), so, its components have the form

$$\begin{aligned} q_M^{\text{el}}(\mathbf{x}_1, \mathbf{x}_2) &= P_1^{\text{el}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times \sqrt{\frac{4\pi}{5}}\tilde{Y}_{2,M}(\mathbf{x}_1) + P_2^{\text{el}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times \sqrt{\frac{4\pi}{5}}\tilde{Y}_{2,M}(\mathbf{x}_2) \\ &\quad + P_3^{\text{el}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times f_{2,M}^{(2)}(\mathbf{x}_1, \mathbf{x}_2), \end{aligned} \quad (23)$$

while its magnetic counterpart,  $q^{\text{mag}}$ , transforms as the irreducible representation ( $2^-$ ) and its components have the form

$$q_M^{\text{mag}}(\mathbf{x}_1, \mathbf{x}_2) = P_1^{\text{mag}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times f_{2,M}^{(3,1)}(\mathbf{x}_1, \mathbf{x}_2) + P_2^{\text{mag}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) \times f_{2,M}^{(3,2)}(\mathbf{x}_1, \mathbf{x}_2). \quad (24)$$

## 4.3. Three vectors

The Molien generating function for the invariant polynomials ( $L = 0$ ) is

$$g_{\text{SO}(3)}^{(a)}(3, 0; \lambda_1, \lambda_2, \lambda_3) = \frac{1 + \lambda_1 \lambda_2 \lambda_3}{(1 - \lambda_1^2)(1 - \lambda_2^2)(1 - \lambda_3^2)(1 - \lambda_1 \lambda_2)(1 - \lambda_1 \lambda_3)(1 - \lambda_2 \lambda_3)}.$$

The 6 primary invariant polynomials of degree 2 are the 6 scalar products  $Q_{i,j} := x_i x_j + y_i y_j + z_i z_j$  for  $1 \leq i \leq j \leq 3$  [42]. The secondary invariant of zero degree is the constant,  $f_1(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) := 1$  and the secondary invariant of degree 3 is the determinant

$$f_2(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) := \begin{vmatrix} x_1 & x_2 & x_3 \\ y_1 & y_2 & y_3 \\ z_1 & z_2 & z_3 \end{vmatrix}. \quad \text{The latter transforming as the irreducible representation } (0^-) \text{ of the group } \text{O}(3),$$

and the potential energy hypersurfaces of tetratomic molecules being totally symmetrical under the O(3)-group, they can be expanded as polynomials in the primary invariant polynomials only.

The Molien generating function for  $L = 1$  is

$$g_{\text{SO}(3)}^{(a)}(3, 1; \lambda_1, \lambda_2, \lambda_3) = \frac{\lambda_1 + \lambda_2 + \lambda_3 + \lambda_1 \lambda_2 + \lambda_1 \lambda_3 + \lambda_2 \lambda_3}{(1 - \lambda_1^2)(1 - \lambda_2^2)(1 - \lambda_3^2)(1 - \lambda_1 \lambda_2)(1 - \lambda_1 \lambda_3)(1 - \lambda_2 \lambda_3)}.$$

The three sets of (1)-covariants of degree 1 can be taken as  $\sqrt{\frac{4\pi}{3}} \tilde{\mathcal{Y}}_{1,M}(\mathbf{x}_i)$ ,  $i \in \{1, 2, 3\}$ . The three sets of (1)-covariants of degree 2 are the cross-products  $f_1(\mathbf{x}_1, \mathbf{x}_2)$ ,  $f_1(\mathbf{x}_1, \mathbf{x}_3)$ , and  $f_1(\mathbf{x}_2, \mathbf{x}_3)$ . The components of the electric dipole moment function of an ABCD molecule have the form:

$$\begin{aligned} \mu_M^{\text{el}}(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) = & P_1^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times \sqrt{\frac{4\pi}{3}} \tilde{\mathcal{Y}}_{1,M}(\mathbf{x}_1) \\ & + P_2^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times \sqrt{\frac{4\pi}{3}} \tilde{\mathcal{Y}}_{1,M}(\mathbf{x}_2) \\ & + P_3^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times \sqrt{\frac{4\pi}{3}} \tilde{\mathcal{Y}}_{1,M}(\mathbf{x}_3), \end{aligned} \quad (25)$$

where  $P_1^{\text{el}}$ ,  $P_2^{\text{el}}$ , and  $P_3^{\text{el}}$  are polynomials in the 6 primary invariants  $Q_{i,j}$ , and those of the magnetic dipole moment function are similarly,

$$\begin{aligned} \mu_M^{\text{mag}}(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) = & P_1^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times f_{1,M}(\mathbf{x}_2, \mathbf{x}_3) \\ & + P_2^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times f_{1,M}(\mathbf{x}_1, \mathbf{x}_3) \\ & + P_3^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times f_{1,M}(\mathbf{x}_1, \mathbf{x}_2). \end{aligned} \quad (26)$$

For  $L = 2$ , when the three representations associated with the three vectors are distinguished by the three variables  $\lambda_i$ , the numerator of the Molien function is:

$$\mathcal{N}_{\text{SO}(3)}^{(a)}(3, 2; \lambda_1, \lambda_2, \lambda_3) = \lambda_1^2 + \lambda_2^2 + \lambda_3^2 + \lambda_1 \lambda_2 + \lambda_1 \lambda_3 + \lambda_2 \lambda_3 +$$

$$\begin{aligned} & \lambda_1^2 \lambda_2 + \lambda_1^2 \lambda_3 + \lambda_1 \lambda_2^2 + \lambda_1 \lambda_3^2 + \lambda_2^2 \lambda_3 + \lambda_2 \lambda_3^2 + 2\lambda_1 \lambda_2 \lambda_3 \\ & - \lambda_1^2 \lambda_2^2 \lambda_3 - \lambda_1^2 \lambda_2 \lambda_3^2 - \lambda_1 \lambda_2^2 \lambda_3^2 - \lambda_1^2 \lambda_2^2 \lambda_3^2. \end{aligned} \quad (27)$$

Its symbolic interpretation in term of generators and syzygies suggests the existence of 6 sets of (2)–covariant generators of degree 2 and 8 sets of (2)–covariant generators of degree 3. These generators are related by 3 syzygies of degree 5 and 1 syzygy of degree 6. Such an expression of the numerator is not suitable for an interpretation of the generating function in term of an integrity basis. However, the expression of the generating function can be rewritten as a sum of two rational functions:

$$g_{SO(3)}^{(b)}(3, 2; \lambda) = \frac{5\lambda^2 + 5\lambda^3}{(1 - \lambda^2)^6} + \frac{\lambda^2 + 3\lambda^3 + \lambda^4}{(1 - \lambda^2)^5}. \quad (28)$$

The symbolic interpretation of the first rational function of  $g_{SO(3)}^{(b)}(3, 2; \lambda)$  specifies that only 5 sets of secondary (2)–covariants of degree 2 and 5 sets of secondary (2)–covariants of degree 3 should be used to generate a free module  $\mathcal{M}_1^{A_1}$  over the ring  $\mathcal{A}_1$  generated by the six primary invariants  $\{Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}\}$ . However, the free module  $\mathcal{M}_1^{A_1}$  is only a submodule of the module of all the (2)–covariant polynomials in the coordinates  $(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3)$ . The second rational function of  $g_{SO(3)}^{(b)}(3, 2; \lambda)$  indicates that a second module  $\mathcal{M}_2^{A_2}$  is needed, with one set of (2)–covariants of degree 2, three sets of (2)–covariants of degree 3 and one set of (2)–covariants of degree 4. The module  $\mathcal{M}_2^{A_2}$  should be free over a subring  $\mathcal{A}_2 \subsetneq \mathcal{A}_1$  spanned by only five primary invariants.

These pieces of information can be used to propose a generalized integrity basis of the non–free module of (2)–covariants in 3 vectors. For  $1 \leq i \leq j \leq 3$ , we define the  $D_{2,M}^{i,j}$ 's as:

$$D_{2,M}^{i,j} = f_{2,M}^{(2)}(\mathbf{x}_i, \mathbf{x}_j).$$

The 6  $D_{2,M}^{i,j}$ 's are the secondary (2)–covariants of degree 2 that we are looking for. Next, we construct a similar expression by substituting  $(x_j, y_j, z_j)$  with a  $(L = 1)$ –covariant of degree 2:

$$\begin{pmatrix} f_{2,2}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k) \\ f_{2,1}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k) \\ f_{2,0}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k) \\ f_{2,-1}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k) \\ f_{2,-2}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k) \end{pmatrix} = \begin{pmatrix} \frac{\sqrt{3}}{2}(x_i(y_j z_k - z_j y_k) - y_i(z_j x_k - x_j z_k)) \\ \frac{\sqrt{3}}{2}(x_i(x_j y_k - y_j x_k) + z_i(y_j z_k - z_j y_k)) \\ \frac{1}{2}(2z_i(x_j y_k - y_j x_k) - x_i(y_j z_k - z_j y_k) - y_i(z_j x_k - x_j z_k)) \\ \frac{\sqrt{3}}{2}(y_i(x_j y_k - y_j x_k) + z_i(z_j x_k - x_j z_k)) \\ \frac{\sqrt{3}}{2}(x_i(z_j x_k - x_j z_k) + y_i(y_j z_k - z_j y_k)) \end{pmatrix}.$$

However, only 8 can be linearly independent, since  $\forall i, j, k \in \{1, 2, 3\}$  and  $M \in \{2, 1, 0, -1, -2\}$ :

$$f_{2,M}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k) + f_{2,M}^{(3)}(\mathbf{x}_i, \mathbf{x}_k, \mathbf{x}_j) = 0,$$

and

$$f_{2,M}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k) + f_{2,M}^{(3)}(\mathbf{x}_j, \mathbf{x}_k, \mathbf{x}_i) + f_{2,M}^{(3)}(\mathbf{x}_k, \mathbf{x}_i, \mathbf{x}_j) = 0.$$

To shorten the notation, we define  $T_{2,M}^{i,j,k} = f_{2,M}^{(3)}(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_k)$ . For a given  $i$  and  $M$ , we retain only those  $T_{2,M}^{i,j,k}$  with  $j < k$ , and for  $i, j, k$  all distinct, we decide to discard

$$T_{2,M}^{2,1,3} = T_{2,M}^{1,2,3} + T_{2,M}^{3,1,2}. \quad (29)$$

This means that we select

$$T_{2,M}^{1,1,2}, T_{2,M}^{1,1,3}, T_{2,M}^{1,2,3}, T_{2,M}^{2,1,2}, T_{2,M}^{2,2,3}, T_{2,M}^{3,1,2}, T_{2,M}^{3,1,3}, T_{2,M}^{3,2,3},$$

as the 8 linearly independent  $T_{2,M}^{i,j,k}$ .

The three syzygies of degree 5 are found to be, by solving linear systems of equations:

$$Q_{2,3}T_{2,M}^{1,1,2} - Q_{2,2}T_{2,M}^{1,1,3} - Q_{1,3}T_{2,M}^{2,1,2} - Q_{1,1}T_{2,M}^{2,2,3} + Q_{1,2}(2T_{2,M}^{1,2,3} + T_{2,M}^{3,1,2}) = 0, \quad (30)$$

$$Q_{1,3}T_{2,M}^{2,2,3} + Q_{3,3}T_{2,M}^{2,1,2} - Q_{1,2}T_{2,M}^{3,2,3} + Q_{2,2}T_{2,M}^{3,1,3} - Q_{2,3}(2T_{2,M}^{3,1,2} + T_{2,M}^{1,2,3}) = 0, \quad (31)$$

$$Q_{1,1}T_{2,M}^{3,2,3} + Q_{2,3}T_{2,M}^{1,1,3} - Q_{1,2}T_{2,M}^{3,1,3} - Q_{3,3}T_{2,M}^{1,1,2} + Q_{1,3}(2T_{2,M}^{3,1,2} - T_{2,M}^{2,1,3}) = 0. \quad (32)$$

The last one can be rewritten by using the relation (29) as

$$Q_{1,1}T_{2,M}^{3,2,3} + Q_{2,3}T_{2,M}^{1,1,3} - Q_{1,2}T_{2,M}^{3,1,3} - Q_{3,3}T_{2,M}^{1,1,2} + Q_{1,3}(T_{2,M}^{3,1,2} - T_{2,M}^{1,2,3}) = 0. \quad (33)$$

Similarly, the syzygy of degree 6 is found to be:

$$\begin{aligned} & (Q_{2,3}^2 - Q_{2,2}Q_{3,3})D_{2,M}^{1,1} + (Q_{1,3}^2 - Q_{1,1}Q_{3,3})D_{2,M}^{2,2} + (Q_{1,2}^2 - Q_{1,1}Q_{2,2})D_{2,M}^{3,3} + \\ & 2((Q_{1,2}Q_{3,3} - Q_{1,3}Q_{2,3})D_{2,M}^{1,2} + (Q_{1,3}Q_{2,2} - Q_{1,2}Q_{2,3})D_{2,M}^{1,3} + (Q_{1,1}Q_{2,3} - Q_{1,2}Q_{1,3})D_{2,M}^{2,3}) = 0. \end{aligned} \quad (34)$$

There is some arbitrariness in the choice of the primary invariant to be removed from the ring of invariants  $\mathcal{A}_1 = \mathbb{C}[Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}]$  to define the subring  $\mathcal{A}_2 \subsetneq \mathcal{A}_1$  of the second submodule  $\mathcal{M}_2^{A_2}$ . Let us remove  $Q_{2,3}$ , so that  $\mathcal{A}_2$  is the subring of  $\mathcal{A}_1$  generated by  $\{Q_{1,1}, Q_{2,2}, Q_{3,3}, Q_{1,2}, Q_{1,3}\}$ . It is then convenient to eliminate the secondary covariant  $D_{2,M}^{1,1}$  of degree 2 from  $\mathcal{M}_1^{A_1}$  and to choose  $D_{2,M}^{1,1}$  and covariant  $Q_{2,3} \times D_{2,M}^{1,1}$  of degree 4 as secondary covariants for  $\mathcal{M}_2^{A_2}$ , because relation (34) allows one to reexpress  $Q_{2,3} \times D_{2,M}^{1,1}$  in terms of elements of either the  $\mathcal{M}_1^{A_1}$  or the  $\mathcal{M}_2^{A_2}$  module, whatever choice of degree 3 secondary covariant partitioning is made. In fact, the same is true for any product  $(Q_{2,3}^n \times D_{2,M}^{1,1})$  with  $n > 1$  by a repeated use of the syzygy (34). It remains to select the three secondary covariants of order 3 to exclude from  $\mathcal{M}_1^{A_1}$  and to include in  $\mathcal{M}_2^{A_2}$ . A natural choice is  $T_{2,M}^{1,1,2}$ ,  $(2T_{2,M}^{3,1,2} + T_{2,M}^{1,2,3})$  and  $T_{2,M}^{1,1,3}$ , since  $Q_{2,3}T_{2,M}^{1,1,2}$ ,  $Q_{2,3}(2T_{2,M}^{1,2,3} + T_{2,M}^{2,2,3})$  and  $Q_{2,3}T_{2,M}^{1,1,3}$ , are easily re-expressed with terms either in  $\mathcal{M}_1^{A_1}$  or in  $\mathcal{M}_2^{A_2}$ , by means of the syzygies (30), (31) and (33) respectively. The 5 secondary covariants of degree 2 and the 5 secondary covariants of degree 3 spanning  $\mathcal{M}_1^{A_1}$  can be chosen to be  $D_{2,M}^{1,2}$ ,  $D_{2,M}^{1,3}$ ,  $D_{2,M}^{2,2}$ ,  $D_{2,M}^{2,3}$ ,  $D_{2,M}^{3,3}$ ,  $T_{2,M}^{1,2,3}$ ,  $T_{2,M}^{2,1,2}$ ,  $T_{2,M}^{2,2,3}$ ,  $T_{2,M}^{3,1,3}$ , and  $T_{2,M}^{3,2,3}$ .

With this choice, the electric quadrupole moment components have the form,

$$q_M^{\text{el}}(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) = P_1^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times D_{2,M}^{1,2}$$

$$\begin{aligned}
& + P_2^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times D_{2,M}^{1,3} \\
& + P_3^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times D_{2,M}^{2,2} \\
& + P_4^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times D_{2,M}^{2,3} \\
& + P_5^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times D_{2,M}^{3,3} \\
& + P_6^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{3,3}) \times D_{2,M}^{1,1} \\
& + P_7^{\text{el}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{3,3}) \times Q_{2,3} D_{2,M}^{1,1}, \tag{35}
\end{aligned}$$

and those of the magnetic dipole moment function can be expressed as:

$$\begin{aligned}
q_M^{\text{mag}}(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) & = P_1^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times T_{2,M}^{1,2,3} \\
& + P_2^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times T_{2,M}^{2,1,2} \\
& + P_3^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times T_{2,M}^{2,2,3} \\
& + P_4^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times T_{2,M}^{3,1,3} \\
& + P_5^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{2,3}, Q_{3,3}) \times T_{2,M}^{3,2,3} \\
& + P_6^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{3,3}) \times T_{2,M}^{1,1,2} \\
& + P_7^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{3,3}) \times (2T_{2,M}^{3,1,2} + T_{2,M}^{1,2,3}) \\
& + P_8^{\text{mag}}(Q_{1,1}, Q_{1,2}, Q_{1,3}, Q_{2,2}, Q_{3,3}) \times T_{2,M}^{1,1,3}. \tag{36}
\end{aligned}$$

These expansions are new and potentially useful, for they are as compact as possible.

#### 4.4. Generalization to higher $N$ and $L$ values

The steps of the construction of the generalized integrity basis presented in the subsection 4.3 for  $N = 3$  and  $L = 2$  can be generalized to higher values of  $N$  and  $L$ . The decomposition of non-free modules into a sum of free modules on subrings of the ring of primary invariants may have more terms. Table A1 suggests a decomposition of the generating function in three rational functions for  $N = 4$  and  $L \geq 3$ , while Table A2 suggests a decomposition in four rational functions for  $N = 5$  and  $L \geq 4$ . One is tempted to conjecture that decompositions in  $(N - 1)$  rational functions will occur for  $N$ -vectors and  $L \geq N - 1$ , for all  $N \geq 2$ . The generalized integrity bases are not unique, since, as we have seen, there is some arbitrariness in the choice of the generators of the primary invariant subrings and of the covariant generators of the modules on these subrings (although some choices are more practical than others). So, it is difficult to give a general and explicit construction. We only sketch the main lines.

The construction of a generalized integrity basis of a non-free module follows the steps of the algorithm presented in section 3.1. To the sum in the right-hand side of (12) corresponds a decomposition of the non-free module in a sum of  $l_{\max}(N, L)$  submodules:

$$\mathcal{M} = \bigoplus_{i=1}^{l_{\max}(N, L)} \mathcal{M}_i^{\mathcal{A}_i}. \tag{37}$$

The number and degrees of the secondary covariants which are the basis of each free submodule  $\mathcal{M}_i^{\mathcal{A}_i}$  in the decomposition (37) are given by the successive remainders of the

divisions by  $(1 - \lambda^2)$  and the last quotient with only positive coefficients. To find the successive sets of syzygies, it is enough to solve linear systems in the primary invariant ring and its successively selected subrings. The numbers of independent syzygies to write and their degrees are given by the negative coefficients in the expressions of the numerator  $\mathcal{N}_{\text{SO}(3)}^{(a)}(N, L, \lambda)$  and its successive quotients by  $(1 - \lambda^2)$  appearing while following the algorithm of section 3.1. For example, for  $N = 4$ ,  $L = 3$ , (A.2) tells us that there will be 19 syzygies of degree 6 and 16 of degree 7 to be used in order to select  $\mathcal{A}_2$  and the 20 secondary covariants of degree 3, 28 secondary covariants of degree 4 and 8 secondary covariants of degree 5 of  $\mathcal{M}_1^{\mathcal{A}_1}$  according to the rest in (15). This first set of syzygies is to be obtained by solving a linear system in  $\mathcal{A}_1$ . Then, the quotient of (15) tells us that a second set of 2 syzygies of degree 6, 4 of degree 7 and 1 of degree 8 is to be obtained by solving linear systems in the subring  $\mathcal{A}_2$ , previously selected. Finally, the remainder in (17), tells us that there are 14 secondary covariants of degree 4 and 8 of degree 5 to be chosen for  $\mathcal{M}_2^{\mathcal{A}_2}$ , and the quotient in (17) that, once  $\mathcal{A}_3$  has been selected, there will be 3 secondary covariants of degree 4, 4 of degree 5 and 1 of degree 6 to be found for  $\mathcal{M}_3^{\mathcal{A}_3}$ .

Since the  $\mathcal{S}_N$  permutation group action on vector indices preserves partial degrees, one can take advantage of the Molien functions with distinguished representation arguments to obtain information about the partial degrees of the variables in these syzygies. This reduces significantly the size of the linear systems to be solved. For example, in the  $N = 3$  and  $L = 2$  case, the syzygy of order 6 is found to be of partial degrees  $n_1 = n_2 = n_3 = 2$  from the last term in (27). We however have not systematically reported such detailed expressions for  $N > 2$  because the closed formulas are not polynomial. For example, for  $N = 3$ , we have obtained the following numerator

$$\begin{aligned} \mathcal{N}_{\text{SO}(3)}^{(a)}(3, L; \lambda_1, \lambda_2, \lambda_3) = & \frac{1}{(\lambda_1 - \lambda_2)(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} [\lambda_2 \lambda_3 (-\lambda_3^{1+L} - \lambda_2 \lambda_3^{1+L} + \lambda_2^{1+L} (1 + \lambda_3)) + \\ & \lambda_1^3 \lambda_2 \lambda_3 (-\lambda_3^{1+L} - \lambda_2 \lambda_3^{1+L} + \lambda_2^{1+L} (1 + \lambda_3)) + \lambda_1^{2+L} (-\lambda_3 (1 + \lambda_3) - \lambda_2^3 \lambda_3 (1 + \lambda_3) + \lambda_2 (1 + \lambda_3^3) + \\ & \lambda_2^2 (1 + \lambda_3^3)) + \lambda_1 (\lambda_3^{2+L} + \lambda_2^3 \lambda_3^{2+L} - \lambda_2^{2+L} (1 + \lambda_3^3)) + \lambda_1^2 (\lambda_3^{2+L} + \lambda_2^3 \lambda_3^{2+L} - \lambda_2^{2+L} (1 + \lambda_3^3))] \end{aligned} \quad (38)$$

which we only managed to simplify into interpretable polynomial expressions such as (27), when  $L$  takes specific values. Such interpretable detailed expressions are useful to provide the partial degrees of the secondary covariants of the modules, after division by the factor in the denominator corresponding to the primary invariant excluded from the next subring in the decomposition: for example, if the invariant  $Q_{2,3}$  is excluded one has to divide by  $(1 - \lambda_2 \lambda_3)$ .

## 5. Conclusion

A general expression of the Molien generating function for SO(3) has been given as well as a recursive process which provides more insight for the practical construction of integrity and generalized integrity bases. So, there are two independent paths to obtain explicit formulas for Molien generating function as soon as  $N > 1$  vectors are

considered. The first relies on the computation of the Molien integral [33] and requires the matrix representation of the group action on the  $N$  vectors. The second path considers the Molien generating function for only one vector as the elementary building material from which the Molien generating functions for more than one vector can be worked out recursively. Both paths have been followed to verify the expressions up to  $N = 5$  vectors provided in this work.

Such Molien generating function expressions are useful guides for the construction of invariant and covariant (possibly generalized) integrity bases of the  $SO(3)$  symmetry group for  $N$  vectors of a three dimensional Euclidean space. The extension to the  $O(3)$  group is direct. When the module of covariants is non-free over an h.s.o.p., an algorithm has been proposed to transform the Molien function into a form amenable to a symbolic interpretation in term of a generalized integrity basis. The same algorithm can guide step by step the construction of such a generalized integrity basis. Within this approach, the non-free module is decomposed as a direct sum of submodules. Each submodule is associated with a rational function indicating the numbers of primary invariants and secondary covariants, the former decreasing in the consecutive rational functions.

The case of the invariant ( $L = 0$  covariants) module is always free [29]. In quantum physics, an integrity basis can be useful to express  $SO(3)$ -totally invariant observables such as the so-called potential energy (hyper)surface (PES) in quantum chemistry [13], when it is not possible or appropriate to separate out rotational from internal coordinates. This is the case of very floppy molecules such as  $CH_5^+$ , in order to use its full symmetry group [45]. Note that PES are actually  $O(3)$ -totally invariant, but for polyatomic molecules of four atoms and more, it is more practical to parametrize them by using coordinates that are  $SO(3)$ - but not  $O(3)$ -invariants, such as dihedral angles.

We have conjectured that the module of  $L = 1$  covariants will always be free as well. We have provided explicit integrity bases up to  $N = 3$ . In quantum physics, these integrity bases can be useful to express observables such as a dipole moment hypersurface, used in theoretical spectroscopy to calculate dipolar transition intensities. If there is a finite group action on the vector variables in addition to the  $SO(3)$  action, we can take advantage of it too as was argued in [13] for the particular case of invariants. For example, for  $N = 2$ , related to the case of a triatomic molecule ABC, if the origin of the two vectors is A and if atoms B and C are equivalent, then the action of the permutation group  $\mathcal{S}_2$  on the vectors  $\overrightarrow{AB} = x_1 \hat{e}_x + y_1 \hat{e}_y + z_1 \hat{e}_z$  and  $\overrightarrow{AC} = x_2 \hat{e}_x + y_2 \hat{e}_y + z_2 \hat{e}_z$  can be exploited to simplify the expression of the physical observables such as the dipole moment surface functions. In this particular case, we deduce for example that the polynomials of (20) must satisfy  $P_1^{\text{el}}(Q_{1,1}, Q_{2,2}, Q_{1,2}) = P_2^{\text{el}}(Q_{2,2}, Q_{1,1}, Q_{1,2})$ .

The ( $L = 2$ )-case can be useful in theoretical physics and chemistry to expand quadrupole moments. One of the main achievement of this work has been to provide compact expansions for quadrupole moment hypersurfaces, which take full advantage of the structure of the non-free covariant module through the use of generalized integrity basis. However, the elimination of some primary invariants breaks the permutational symmetry of the subrings used for the modules  $\mathcal{M}_2^{A^2}$ ,  $\mathcal{M}_3^{A^3}$ ,  $\dots$ , of the decomposition

of the module of covariants. So, unfortunately in such a case, further permutational symmetry adaptation may be unpractical in general.

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## Appendix A. Molien generating functions for four and five vectors

### Appendix A.1. Molien generating function for four vectors

By coupling the Molien generating functions for one vector and three vectors, the Molien generating function for four vectors can be written as a single rational function:

$$g_{\text{SO}(3)}^{(a)}(4, L; \lambda) = \frac{\mathcal{N}_{\text{SO}(3)}^{(a)}(4, L; \lambda)}{(1 - \lambda^2)^9}, \quad (\text{A.1})$$

with the numerator:

$$\begin{aligned} \mathcal{N}_{\text{SO}(3)}^{(a)}(4, L; \lambda) = & \frac{(L+3)(L+2)(L+1)}{6} \lambda^L + \frac{(L+3)(L+2)L}{2} \lambda^{L+1} \\ & + \frac{(L+3)(L+2)(L+1)}{6} \lambda^{L+2} - \frac{(L+3)(L-2)(5L+4)}{6} \lambda^{L+3} \\ & - \frac{(L+3)(L-2)(5L+1)}{6} \lambda^{L+4} + \frac{L(L-1)(L-2)}{6} \lambda^{L+5} \\ & + \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+6} + \frac{L(L-1)(L-2)}{6} \lambda^{L+7}. \end{aligned} \quad (\text{A.2})$$

The Molien generating function (A.1) has non-negative coefficients in its numerator for  $L \in \{0, 1, 2\}$ . Negative coefficients appear for  $L \geq 3$ . However, the Molien function can be rewritten as (A.3), which has only non-negative coefficients in the numerator for  $L \in \{2, 3, 4\}$ ,

$$\begin{aligned} & g_{\text{SO}(3)}^{(b)}(4, L; \lambda) \\ = & \frac{\frac{(L+3)(L+2)(L+1)}{6} \lambda^L + 4(2L+1) \lambda^{L+1} + (-\frac{1}{6}L^3 - L^2 + \frac{37}{6}L + 3) \lambda^{L+2}}{(1 - \lambda^2)^9} \\ & + \frac{2(L-2)(2L+1) \lambda^{L+1} - \frac{(L-2)(L^2-16L-9)}{6} \lambda^{L+2}}{(1 - \lambda^2)^8} \\ & + \frac{\frac{L(L-1)(L-2)}{2} \lambda^{L+1} + \frac{(L+1)(L-1)(L-2)}{2} \lambda^{L+2} + \frac{L(L-1)(L-2)}{6} \lambda^{L+3}}{(1 - \lambda^2)^7}, \end{aligned} \quad (\text{A.3})$$

as (A.4), which has only non-negative coefficients in the numerator for  $L$  between 5 and 16,

$$g_{\text{SO}(3)}^{(c)}(4, L; \lambda)$$

$$\begin{aligned}
&= \frac{4(2L+1)\lambda^L + 4(2L+1)\lambda^{L+1}}{(1-\lambda^2)^9} \\
&+ \frac{(\frac{1}{6}L^3 + L^2 - \frac{37}{6}L - 3)\lambda^L + 2(L-2)(2L+1)\lambda^{L+1} - \frac{(L-2)(L^2-16L-9)}{6}\lambda^{L+2}}{(1-\lambda^2)^8} \\
&+ \frac{\frac{L(L-1)(L-2)}{2}\lambda^{L+1} + \frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+2} + \frac{L(L-1)(L-2)}{6}\lambda^{L+3}}{(1-\lambda^2)^7}, \tag{A.4}
\end{aligned}$$

and as (A.5), which has only non-negative coefficients in the numerator for  $L \geq 17$ ,

$$\begin{aligned}
&g_{\text{SO}(3)}^{(d)}(4, L; \lambda) \\
&= \frac{4(2L+1)\lambda^L + 4(2L+1)\lambda^{L+1}}{(1-\lambda^2)^9} \\
&+ \frac{2(L-3)(2L+1)\lambda^L + 2(L-2)(2L+1)\lambda^{L+1}}{(1-\lambda^2)^8} \\
&+ \frac{\frac{(L-2)(L^2-16L-9)}{6}\lambda^L + \frac{L(L-1)(L-2)}{2}\lambda^{L+1} + \frac{(L+1)(L-1)(L-2)}{2}\lambda^{L+2} + \frac{L(L-1)(L-2)}{6}\lambda^{L+3}}{(1-\lambda^2)^7}. \tag{A.5}
\end{aligned}$$

The expressions of the Molien generating functions suitable for a symbolic interpretation in term of a (generalized) integrity basis are given in Table A1 for selected values of  $L$ .

### Appendix A.2. Molien generating function for five vectors

By coupling the Molien generating functions for one vector and four vectors, the Molien generating function for five vectors can be written as a single rational function:

$$g_{\text{SO}(3)}^{(a)}(5, L; \lambda) = \frac{\mathcal{N}_{\text{SO}(3)}^{(a)}(5, L; \lambda)}{(1-\lambda^2)^{12}}, \tag{A.6}$$

with the numerator:

$$\begin{aligned}
\mathcal{N}_{\text{SO}(3)}^{(a)}(5, L; \lambda) &= \frac{(L+4)(L+3)(L+2)(L+1)}{24}\lambda^L + \frac{(L+4)(L+3)(L+2)L}{6}\lambda^{L+1} \\
&+ \frac{(L+4)(L+3)(L+2)(L+1)}{8}\lambda^{L+2} - \frac{(L+4)(L+3)(L^2-3L-\frac{5}{2})}{3}\lambda^{L+3} \\
&- \frac{(L+4)(L+3)(L-3)(7L+2)}{12}\lambda^{L+4} - \frac{(L+4)(L-3)(2L+1)}{2}\lambda^{L+5} \\
&+ \frac{(L+4)(L-2)(L-3)(7L+5)}{12}\lambda^{L+6} + \frac{(L-2)(L-3)(L^2+5L+\frac{3}{2})}{3}\lambda^{L+7} \\
&- \frac{L(L-1)(L-2)(L-3)}{8}\lambda^{L+8} - \frac{(L+1)(L-1)(L-2)(L-3)}{6}\lambda^{L+9} \\
&- \frac{L(L-1)(L-2)(L-3)}{24}\lambda^{L+10}. \tag{A.7}
\end{aligned}$$

**Table A1.** Expressions of the  $g_{\text{SO}(3)}^{(x)}(4, L; \lambda)$  Molien generating functions for four vectors and selected final irreducible representations  $L$  suitable for a symbolic interpretation in term of a (generalized) integrity basis.

$x$	$L$	$g_{\text{SO}(3)}^{(x)}(4, L; \lambda)$
$a$	0	$\frac{1+\lambda^2+4\lambda^3+\lambda^4+\lambda^6}{(1-\lambda^2)^9}$
$a$	1	$\frac{4\lambda+6\lambda^2+4\lambda^3+6\lambda^4+4\lambda^5}{(1-\lambda^2)^9}$
$a$ or $b$	2	$\frac{10\lambda^2+20\lambda^3+10\lambda^4}{(1-\lambda^2)^9}$
$b$	3	$\frac{20\lambda^3+28\lambda^4+8\lambda^5}{(1-\lambda^2)^9} + \frac{14\lambda^4+8\lambda^5}{(1-\lambda^2)^8} + \frac{3\lambda^4+4\lambda^5+\lambda^6}{(1-\lambda^2)^7}$
$b$	4	$\frac{35\lambda^4+36\lambda^5+\lambda^6}{(1-\lambda^2)^9} + \frac{36\lambda^5+19\lambda^6}{(1-\lambda^2)^8} + \frac{12\lambda^5+15\lambda^6+4\lambda^7}{(1-\lambda^2)^7}$
$c$	5	$\frac{44\lambda^5+44\lambda^6}{(1-\lambda^2)^9} + \frac{12\lambda^5+66\lambda^6+32\lambda^7}{(1-\lambda^2)^8} + \frac{30\lambda^6+36\lambda^7+10\lambda^8}{(1-\lambda^2)^7}$
$c$	6	$\frac{52\lambda^6+52\lambda^7}{(1-\lambda^2)^9} + \frac{32\lambda^6+104\lambda^7+46\lambda^8}{(1-\lambda^2)^8} + \frac{60\lambda^7+70\lambda^8+20\lambda^9}{(1-\lambda^2)^7}$
$c$	7	$\frac{60\lambda^7+60\lambda^8}{(1-\lambda^2)^9} + \frac{60\lambda^7+150\lambda^8+60\lambda^9}{(1-\lambda^2)^8} + \frac{105\lambda^8+120\lambda^9+35\lambda^{10}}{(1-\lambda^2)^7}$
$c$	$\vdots$	
$c$	14	$\frac{116\lambda^{14}+116\lambda^{15}}{(1-\lambda^2)^9} + \frac{564\lambda^{14}+696\lambda^{15}+74\lambda^{16}}{(1-\lambda^2)^8} + \frac{1092\lambda^{15}+1170\lambda^{16}+364\lambda^{17}}{(1-\lambda^2)^7}$
$c$	15	$\frac{124\lambda^{15}+124\lambda^{16}}{(1-\lambda^2)^9} + \frac{692\lambda^{15}+806\lambda^{16}+52\lambda^{17}}{(1-\lambda^2)^8} + \frac{1365\lambda^{16}+1456\lambda^{17}+455\lambda^{18}}{(1-\lambda^2)^7}$
$c$	16	$\frac{132\lambda^{16}+132\lambda^{17}}{(1-\lambda^2)^9} + \frac{837\lambda^{16}+924\lambda^{17}+21\lambda^{18}}{(1-\lambda^2)^8} + \frac{1680\lambda^{17}+1785\lambda^{18}+560\lambda^{19}}{(1-\lambda^2)^7}$
$d$	17	$\frac{140\lambda^{17}+140\lambda^{18}}{(1-\lambda^2)^9} + \frac{980\lambda^{17}+1050\lambda^{18}}{(1-\lambda^2)^8} + \frac{20\lambda^{17}+2040\lambda^{18}+2160\lambda^{19}+680\lambda^{20}}{(1-\lambda^2)^7}$
$d$	18	$\frac{148\lambda^{18}+148\lambda^{19}}{(1-\lambda^2)^9} + \frac{1110\lambda^{18}+1184\lambda^{19}}{(1-\lambda^2)^8} + \frac{72\lambda^{18}+2448\lambda^{19}+2584\lambda^{20}+816\lambda^{21}}{(1-\lambda^2)^7}$
$d$	19	$\frac{156\lambda^{19}+156\lambda^{20}}{(1-\lambda^2)^9} + \frac{1248\lambda^{19}+1326\lambda^{20}}{(1-\lambda^2)^8} + \frac{136\lambda^{19}+2907\lambda^{20}+3060\lambda^{21}+969\lambda^{22}}{(1-\lambda^2)^7}$
$d$	$\vdots$	

The six next alternative expressions of the Molien generating function are written as a sum over four rational functions:

$$g_{\text{SO}(3)}^{(x)}(5, L; \lambda) = \frac{\mathcal{N}_{\text{SO}(3)}^{(x),1}(5, L; \lambda)}{(1-\lambda^2)^{12}} + \frac{\mathcal{N}_{\text{SO}(3)}^{(x),2}(5, L; \lambda)}{(1-\lambda^2)^{11}} + \frac{\mathcal{N}_{\text{SO}(3)}^{(x),3}(5, L; \lambda)}{(1-\lambda^2)^{10}} + \frac{\mathcal{N}_{\text{SO}(3)}^{(x),4}(5, L; \lambda)}{(1-\lambda^2)^9}, \quad x \in \{b, c, d, e, f, g\},$$

and provide for any value of  $L$  at least one expression with only non-negative coefficients in the numerators. An algorithm to derive these numerators is presented in section 3.1.

#### Appendix A.2.1. Numerators of the Molien generating function (b)

$$\begin{aligned} & \mathcal{N}_{\text{SO}(3)}^{(b),1}(5, L; \lambda) \\ &= \frac{(L+4)(L+3)(L+2)(L+1)}{24} \lambda^L + 20(2L+1)\lambda^{L+1} \\ & \quad + \left(-\frac{1}{24}L^4 - \frac{5}{12}L^3 - \frac{35}{24}L^2 + \frac{455}{12}L + 19\right) \lambda^{L+2} \\ & \mathcal{N}_{\text{SO}(3)}^{(b),2}(5, L; \lambda) \\ &= \left(\frac{1}{6}L^4 + \frac{3}{2}L^3 + \frac{13}{3}L^2 - 36L - 20\right) \lambda^{L+1} \end{aligned}$$

$$\begin{aligned}
& - \frac{(L-3)(L^3 + 13L^2 - 406L - 208)}{24} \lambda^{L+2} \\
& - \frac{(L-3)(L^3 + 12L^2 - 58L - 30)}{6} \lambda^{L+3} \\
\mathcal{N}_{\text{SO}(3)}^{(b),3}(5, L; \lambda) & \\
& = - \frac{(L-2)(L-3)(L^2 - 81L - 40)}{24} \lambda^{L+2} - \frac{(L-2)(L-3)(L^2 - 10L - 6)}{6} \lambda^{L+3} \\
\mathcal{N}_{\text{SO}(3)}^{(b),4}(5, L; \lambda) & \\
& = \frac{L(L-1)(L-2)(L-3)}{4} \lambda^{L+2} + \frac{(L+1)(L-1)(L-2)(L-3)}{6} \lambda^{L+3} \\
& + \frac{L(L-1)(L-2)(L-3)}{24} \lambda^{L+4}
\end{aligned}$$

Appendix A.2.2. Numerators of the Molien generating function (c)

$$\begin{aligned}
\mathcal{N}_{\text{SO}(3)}^{(c),1}(5, L; \lambda) & \\
& = \frac{(L+4)(L+3)(L+2)(L+1)}{24} \lambda^L + 20(2L+1) \lambda^{L+1} \\
& + \left( -\frac{1}{24} L^4 - \frac{5}{12} L^3 - \frac{35}{24} L^2 + \frac{455}{12} L + 19 \right) \lambda^{L+2} \\
\mathcal{N}_{\text{SO}(3)}^{(c),2}(5, L; \lambda) & \\
& = 5(2L+1)(2L-7) \lambda^{L+1} - \frac{(L-3)(L^3 + 13L^2 - 406L - 208)}{24} \lambda^{L+2} \\
\mathcal{N}_{\text{SO}(3)}^{(c),3}(5, L; \lambda) & \\
& = \frac{(L-3)(L^3 + 12L^2 - 58L - 30)}{6} \lambda^{L+1} \\
& - \frac{(L-2)(L-3)(L^2 - 81L - 40)}{24} \lambda^{L+2} \\
& - \frac{(L-2)(L-3)(L^2 - 10L - 6)}{6} \lambda^{L+3} \\
\mathcal{N}_{\text{SO}(3)}^{(c),4}(5, L; \lambda) & \\
& = \frac{L(L-1)(L-2)(L-3)}{4} \lambda^{L+2} + \frac{(L+1)(L-1)(L-2)(L-3)}{6} \lambda^{L+3} \\
& + \frac{L(L-1)(L-2)(L-3)}{24} \lambda^{L+4}
\end{aligned}$$

Appendix A.2.3. Numerators of the Molien generating function (d)

$$\begin{aligned}
\mathcal{N}_{\text{SO}(3)}^{(d),1}(5, L; \lambda) & \\
& = 20(2L+1) \lambda^L + 20(2L+1) \lambda^{L+1} \\
\mathcal{N}_{\text{SO}(3)}^{(d),2}(5, L; \lambda) & \\
& = \left( \frac{1}{24} L^4 + \frac{5}{12} L^3 + \frac{35}{24} L^2 - \frac{455}{12} L - 19 \right) \lambda^L + 5(2L+1)(2L-7) \lambda^{L+1}
\end{aligned}$$

$$\begin{aligned}
& - \frac{(L-3)(L^3 + 13L^2 - 406L - 208)}{24} \lambda^{L+2} \\
\mathcal{N}_{SO(3)}^{(d),3}((L); \Gamma_5; \lambda) & \\
& = \frac{(L-3)(L^3 + 12L^2 - 58L - 30)}{6} \lambda^{L+1} - \frac{(L-2)(L-3)(L^2 - 81L - 40)}{24} \lambda^{L+2} \\
& - \frac{(L-2)(L-3)(L^2 - 10L - 6)}{6} \lambda^{L+3} \\
\mathcal{N}_{SO(3)}^{(d),4}(5, L; \lambda) & \\
& = \frac{L(L-1)(L-2)(L-3)}{4} \lambda^{L+2} + \frac{(L+1)(L-1)(L-2)(L-3)}{6} \lambda^{L+3} \\
& + \frac{L(L-1)(L-2)(L-3)}{24} \lambda^{L+4}
\end{aligned}$$

Appendix A.2.4. Numerators of the Molien generating function (e)

$$\begin{aligned}
\mathcal{N}_{SO(3)}^{(e),1}(5, L; \lambda) & \\
& = 20(2L+1)\lambda^L + 20(2L+1)\lambda^{L+1} \\
\mathcal{N}_{SO(3)}^{(e),2}(5, L; \lambda) & \\
& = \left(\frac{1}{24}L^4 + \frac{5}{12}L^3 + \frac{35}{24}L^2 - \frac{455}{12}L - 19\right)\lambda^L + 5(2L+1)(2L-7)\lambda^{L+1} \\
& - \frac{(L-3)(L^3 + 13L^2 - 406L - 208)}{24} \lambda^{L+2} \\
\mathcal{N}_{SO(3)}^{(e),3}(5, L; \lambda) & \\
& = (L-3)(2L-7)(2L+1)\lambda^{L+1} - \frac{(L-2)(L-3)(L^2 - 81L - 40)}{24} \lambda^{L+2} \\
\mathcal{N}_{SO(3)}^{(e),4}(5, L; \lambda) & \\
& = \frac{(L-2)(L-3)(L^2 - 10L - 6)}{6} \lambda^{L+1} + \frac{L(L-1)(L-2)(L-3)}{4} \lambda^{L+2} \\
& + \frac{(L+1)(L-1)(L-2)(L-3)}{6} \lambda^{L+3} + \frac{L(L-1)(L-2)(L-3)}{24} \lambda^{L+4}
\end{aligned}$$

Appendix A.2.5. Numerators of the Molien generating function (f)

$$\begin{aligned}
\mathcal{N}_{SO(3)}^{(f),1}(5, L; \lambda) & \\
& = 20(2L+1)\lambda^L + 20(2L+1)\lambda^{L+1} \\
\mathcal{N}_{SO(3)}^{(f),2}(5, L; \lambda) & \\
& = 5(2L+1)(2L-9)\lambda^L + 5(2L+1)(2L-7)\lambda^{L+1} \\
\mathcal{N}_{SO(3)}^{(f),3}(5, L; \lambda) & \\
& = \frac{(L-3)(L^3 + 13L^2 - 406L - 208)}{24} \lambda^L + (L-3)(2L+1)(2L-7)\lambda^{L+1} \\
& - \frac{(L-2)(L-3)(L^2 - 81L - 40)}{24} \lambda^{L+2}
\end{aligned}$$

$$\begin{aligned}
& \mathcal{N}_{\text{SO}(3)}^{(f),4}(5, L; \lambda) \\
&= \frac{(L-2)(L-3)(L^2-10L-6)}{6} \lambda^{L+1} + \frac{L(L-1)(L-2)(L-3)}{4} \lambda^{L+2} \\
&+ \frac{(L+1)(L-1)(L-2)(L-3)}{6} \lambda^{L+3} + \frac{L(L-1)(L-2)(L-3)}{24} \lambda^{L+4}
\end{aligned}$$

Appendix A.2.6. Numerators of the Molien generating function (g)

$$\begin{aligned}
& \mathcal{N}_{\text{SO}(3)}^{(g),1}(5, L; \lambda) \\
&= 20(2L+1)\lambda^L + 20(2L+1)\lambda^{L+1} \\
& \mathcal{N}_{\text{SO}(3)}^{(g),2}(5, L; \lambda) \\
&= 5(2L+1)(2L-9)\lambda^L + 5(2L+1)(2L-7)\lambda^{L+1} \\
& \mathcal{N}_{\text{SO}(3)}^{(g),3}(5, L; \lambda) \\
&= 2(L-3)(L-6)(2L+1)\lambda^L + (L-3)(2L+1)(2L-7)\lambda^{L+1} \\
& \mathcal{N}_{\text{SO}(3)}^{(g),4}(5, L; \lambda) \\
&= \frac{(L-2)(L-3)(L^2-81L-40)}{24} \lambda^L + \frac{(L-2)(L-3)(L^2-10L-6)}{6} \lambda^{L+1} \\
&+ \frac{L(L-1)(L-2)(L-3)}{4} \lambda^{L+2} + \frac{(L+1)(L-1)(L-2)(L-3)}{6} \lambda^{L+3} \\
&+ \frac{L(L-1)(L-2)(L-3)}{24} \lambda^{L+4}
\end{aligned}$$

The expressions of the Molien generating functions suitable for a symbolic interpretation in term of a (generalized) integrity basis are given in Table A2 for selected values of  $L$ .

**Table A2.** Expressions of the  $g_{\text{SO}(3)}^{(x)}(5, L; \lambda)$  Molien generating functions for five vectors and selected final irreducible representations  $L$  suitable for a symbolic interpretation in term of a (generalized) integrity basis.

$x$	$L$	$g_{\text{SO}(3)}^{(x)}(5, L; \lambda)$
$a$	0	$\frac{1+3\lambda^2+10\lambda^3+6\lambda^4+6\lambda^5+10\lambda^6+3\lambda^7+\lambda^9}{(1-\lambda^2)^{12}}$
$a$	1	$\frac{5\lambda+10\lambda^2+15\lambda^3+30\lambda^4+30\lambda^5+15\lambda^6+10\lambda^7+5\lambda^8}{(1-\lambda^2)^{12}}$
$a$	2	$\frac{15\lambda^2+40\lambda^3+45\lambda^4+45\lambda^5+40\lambda^6+15\lambda^7}{(1-\lambda^2)^{12}}$
$a$	3	$\frac{35\lambda^3+105\lambda^4+105\lambda^5+35\lambda^6}{(1-\lambda^2)^{12}}$
$b$	4	$\frac{70\lambda^4+180\lambda^5+110\lambda^6}{(1-\lambda^2)^{12}} + \frac{44\lambda^5+65\lambda^6+\lambda^7}{(1-\lambda^2)^{11}} + \frac{29\lambda^6+10\lambda^7}{(1-\lambda^2)^{10}} + \frac{6\lambda^6+5\lambda^7+\lambda^8}{(1-\lambda^2)^9}$
$c$	5	$\frac{126\lambda^5+220\lambda^6+94\lambda^7}{(1-\lambda^2)^{12}} + \frac{165\lambda^6+149\lambda^7}{(1-\lambda^2)^{11}} + \frac{35\lambda^6+105\lambda^7+31\lambda^8}{(1-\lambda^2)^{10}} + \frac{30\lambda^7+24\lambda^8+5\lambda^9}{(1-\lambda^2)^9}$
$c$	6	$\frac{210\lambda^6+260\lambda^7+50\lambda^8}{(1-\lambda^2)^{12}} + \frac{325\lambda^7+245\lambda^8}{(1-\lambda^2)^{11}} + \frac{135\lambda^7+245\lambda^8+60\lambda^9}{(1-\lambda^2)^{10}} + \frac{90\lambda^8+70\lambda^9+15\lambda^{10}}{(1-\lambda^2)^9}$
$d$	7	$\frac{300\lambda^7+300\lambda^8}{(1-\lambda^2)^{12}} + \frac{30\lambda^7+525\lambda^8+345\lambda^9}{(1-\lambda^2)^{11}} + \frac{330\lambda^8+465\lambda^9+90\lambda^{10}}{(1-\lambda^2)^{10}} + \frac{210\lambda^9+160\lambda^{10}+35\lambda^{11}}{(1-\lambda^2)^9}$
$d$	8	$\frac{340\lambda^8+340\lambda^9}{(1-\lambda^2)^{12}} + \frac{155\lambda^8+765\lambda^9+440\lambda^{10}}{(1-\lambda^2)^{11}} + \frac{655\lambda^9+780\lambda^{10}+110\lambda^{11}}{(1-\lambda^2)^{10}} + \frac{420\lambda^{10}+315\lambda^{11}+70\lambda^{12}}{(1-\lambda^2)^9}$
$d$	9	$\frac{380\lambda^9+380\lambda^{10}}{(1-\lambda^2)^{12}} + \frac{335\lambda^9+1045\lambda^{10}+520\lambda^{11}}{(1-\lambda^2)^{11}} + \frac{1149\lambda^{10}+1204\lambda^{11}+105\lambda^{12}}{(1-\lambda^2)^{10}} + \frac{756\lambda^{11}+560\lambda^{12}+126\lambda^{13}}{(1-\lambda^2)^9}$
$d$	10	$\frac{420\lambda^{10}+420\lambda^{11}}{(1-\lambda^2)^{12}} + \frac{581\lambda^{10}+1365\lambda^{11}+574\lambda^{12}}{(1-\lambda^2)^{11}} + \frac{1855\lambda^{11}+1750\lambda^{12}+56\lambda^{13}}{(1-\lambda^2)^{10}} + \frac{1260\lambda^{12}+924\lambda^{13}+210\lambda^{14}}{(1-\lambda^2)^9}$
$e$	11	$\frac{460\lambda^{11}+460\lambda^{12}}{(1-\lambda^2)^{12}} + \frac{905\lambda^{11}+1725\lambda^{12}+590\lambda^{13}}{(1-\lambda^2)^{11}} + \frac{2760\lambda^{12}+2430\lambda^{13}}{(1-\lambda^2)^{10}} + \frac{60\lambda^{12}+1980\lambda^{13}+1440\lambda^{14}+330\lambda^{15}}{(1-\lambda^2)^9}$
$e$	12	$\frac{500\lambda^{12}+500\lambda^{13}}{(1-\lambda^2)^{12}} + \frac{1320\lambda^{12}+2125\lambda^{13}+555\lambda^{14}}{(1-\lambda^2)^{11}} + \frac{3825\lambda^{13}+3255\lambda^{14}}{(1-\lambda^2)^{10}} + \frac{270\lambda^{13}+2970\lambda^{14}+2145\lambda^{15}+495\lambda^{16}}{(1-\lambda^2)^9}$
$e$	13	$\frac{540\lambda^{13}+540\lambda^{14}}{(1-\lambda^2)^{12}} + \frac{1840\lambda^{13}+2565\lambda^{14}+455\lambda^{15}}{(1-\lambda^2)^{11}} + \frac{5130\lambda^{14}+4235\lambda^{15}}{(1-\lambda^2)^{10}} + \frac{605\lambda^{14}+4290\lambda^{15}+3080\lambda^{16}+715\lambda^{17}}{(1-\lambda^2)^9}$
$e$	14	$\frac{580\lambda^{14}+580\lambda^{15}}{(1-\lambda^2)^{12}} + \frac{2480\lambda^{14}+3045\lambda^{15}+275\lambda^{16}}{(1-\lambda^2)^{11}} + \frac{6699\lambda^{15}+5379\lambda^{16}}{(1-\lambda^2)^{10}} + \frac{1100\lambda^{15}+6006\lambda^{16}+4290\lambda^{17}+1001\lambda^{18}}{(1-\lambda^2)^9}$
$f$	15	$\frac{620\lambda^{15}+620\lambda^{16}}{(1-\lambda^2)^{12}} + \frac{3255\lambda^{15}+3565\lambda^{16}}{(1-\lambda^2)^{11}} + \frac{\lambda^{15}+8556\lambda^{16}+6695\lambda^{17}}{(1-\lambda^2)^{10}} + \frac{1794\lambda^{16}+8190\lambda^{17}+5824\lambda^{18}+1365\lambda^{19}}{(1-\lambda^2)^9}$
$f$	16	$\frac{660\lambda^{16}+660\lambda^{17}}{(1-\lambda^2)^{12}} + \frac{3795\lambda^{16}+4125\lambda^{17}}{(1-\lambda^2)^{11}} + \frac{390\lambda^{16}+10725\lambda^{17}+8190\lambda^{18}}{(1-\lambda^2)^{10}} + \frac{2730\lambda^{17}+10920\lambda^{18}+7735\lambda^{19}+1820\lambda^{20}}{(1-\lambda^2)^9}$
$f$	17	$\frac{700\lambda^{17}+700\lambda^{18}}{(1-\lambda^2)^{12}} + \frac{4375\lambda^{17}+4725\lambda^{18}}{(1-\lambda^2)^{11}} + \frac{910\lambda^{17}+13230\lambda^{18}+9870\lambda^{19}}{(1-\lambda^2)^{10}} + \frac{3955\lambda^{18}+14280\lambda^{19}+10080\lambda^{20}+2380\lambda^{21}}{(1-\lambda^2)^9}$
$f$	$\vdots$	$\vdots$
$f$	79	$\frac{3180\lambda^{79}+3180\lambda^{80}}{(1-\lambda^2)^{12}} + \frac{118455\lambda^{79}+120045\lambda^{80}}{(1-\lambda^2)^{11}} + \frac{1715985\lambda^{79}+1824684\lambda^{80}+48279\lambda^{81}}{(1-\lambda^2)^{10}} + \frac{5310690\lambda^{80}+9015006\lambda^{81}+6086080\lambda^{82}+1502501\lambda^{83}}{(1-\lambda^2)^9}$
$f$	80	$\frac{3220\lambda^{80}+3220\lambda^{81}}{(1-\lambda^2)^{12}} + \frac{121555\lambda^{80}+123165\lambda^{81}}{(1-\lambda^2)^{11}} + \frac{1804726\lambda^{80}+1896741\lambda^{81}+30030\lambda^{82}}{(1-\lambda^2)^{10}} + \frac{5599594\lambda^{81}+9489480\lambda^{82}+6405399\lambda^{83}+1581580\lambda^{84}}{(1-\lambda^2)^9}$
$f$	81	$\frac{3260\lambda^{81}+3260\lambda^{82}}{(1-\lambda^2)^{12}} + \frac{124695\lambda^{81}+126325\lambda^{82}}{(1-\lambda^2)^{11}} + \frac{1896830\lambda^{81}+1970670\lambda^{82}+10270\lambda^{83}}{(1-\lambda^2)^{10}} + \frac{5900115\lambda^{82}+9982440\lambda^{83}+6737120\lambda^{84}+1663740\lambda^{85}}{(1-\lambda^2)^9}$
$g$	82	$\frac{3300\lambda^{82}+3300\lambda^{83}}{(1-\lambda^2)^{12}} + \frac{127875\lambda^{82}+129525\lambda^{83}}{(1-\lambda^2)^{11}} + \frac{1981320\lambda^{82}+2046495\lambda^{83}}{(1-\lambda^2)^{10}} + \frac{11060\lambda^{82}+6212560\lambda^{83}+10494360\lambda^{84}+7081560\lambda^{85}+1749060\lambda^{86}}{(1-\lambda^2)^9}$
$g$	83	$\frac{3340\lambda^{83}+3340\lambda^{84}}{(1-\lambda^2)^{12}} + \frac{131095\lambda^{83}+132765\lambda^{84}}{(1-\lambda^2)^{11}} + \frac{2057440\lambda^{83}+2124240\lambda^{84}}{(1-\lambda^2)^{10}} + \frac{34020\lambda^{83}+6537240\lambda^{84}+11025720\lambda^{85}+7439040\lambda^{86}+1837620\lambda^{87}}{(1-\lambda^2)^9}$
$g$	84	$\frac{3380\lambda^{84}+3380\lambda^{85}}{(1-\lambda^2)^{12}} + \frac{134355\lambda^{84}+136045\lambda^{85}}{(1-\lambda^2)^{11}} + \frac{2135484\lambda^{84}+2203929\lambda^{85}}{(1-\lambda^2)^{10}} + \frac{58671\lambda^{84}+6874470\lambda^{85}+11577006\lambda^{86}+7809885\lambda^{87}+1929501\lambda^{88}}{(1-\lambda^2)^9}$
$q$	$\vdots$	$\vdots$

## Appendix B. Complex and real solid harmonics

The complex solid harmonics are homogeneous polynomials of degree  $l$  in  $x$ ,  $y$ , and  $z$ . Their expression is given by [46]:

$$\mathcal{Y}_{l,m}(\mathbf{x}) = \sqrt{\frac{2l+1}{4\pi}} (l+m)!(l-m)! \sum_k \frac{(-x-iy)^{k+m} (x-iy)^k z^{l-2k-m}}{2^{2k+m} (k+m)!k!(l-m-2k)!},$$

where  $\mathbf{x}$  is the triplet of coordinates  $(x, y, z)$ . The complex solid harmonics are complex-valued functions for  $m \neq 0$  and satisfy the property

$$\mathcal{Y}_{l,m}(\mathbf{x})^* = (-1)^m \mathcal{Y}_{l,-m}(\mathbf{x}).$$

Biedenharn and Louck [36] define the real solid harmonics  $\bar{\mathcal{Y}}_{l,m}$  through the linear combinations of complex solid harmonics [47]:

$$\begin{aligned} \bar{\mathcal{Y}}_{l,m}(\mathbf{x}) &= -\frac{1}{\sqrt{2}} [\mathcal{Y}_{l,m}(\mathbf{x}) + (-1)^m \mathcal{Y}_{l,-m}(\mathbf{x})], \\ \bar{\mathcal{Y}}_{l,0}(\mathbf{x}) &= \mathcal{Y}_{l,0}(\mathbf{x}), \\ \bar{\mathcal{Y}}_{l,-m}(\mathbf{x}) &= \frac{i}{\sqrt{2}} [\mathcal{Y}_{l,m}(\mathbf{x}) - (-1)^m \mathcal{Y}_{l,-m}(\mathbf{x})], \end{aligned}$$

with  $m \in \{l, l-1, \dots, 1\}$ .

Other linear combinations of complex solid harmonics exist in the literature. Steinborn [48] or Blanco *et al.* [49] use:

$$\begin{aligned} \tilde{\mathcal{Y}}_{l,m}(\mathbf{x}) &= \frac{(-1)^m}{\sqrt{2}} [\mathcal{Y}_{l,m}(\mathbf{x}) + (-1)^m \mathcal{Y}_{l,-m}(\mathbf{x})], \\ \tilde{\mathcal{Y}}_{l,0}(\mathbf{x}) &= \mathcal{Y}_{l,0}(\mathbf{x}), \\ \tilde{\mathcal{Y}}_{l,-m}(\mathbf{x}) &= \frac{(-1)^m}{i\sqrt{2}} [\mathcal{Y}_{l,m}(\mathbf{x}) - (-1)^m \mathcal{Y}_{l,-m}(\mathbf{x})], \end{aligned}$$

with  $m \in \{l, l-1, \dots, 1\}$ . The two definitions of real solid harmonics are identical except for  $m$  even where the two definitions give expressions with opposite sign. In the main text, we use  $\tilde{\mathcal{Y}}_{l,m}$  as real solid harmonics. Their expression for up to  $L = 3$  are given below:

$$\begin{aligned} \sqrt{4\pi} \tilde{\mathcal{Y}}_{0,0}(\mathbf{x}) &= 1 \\ \sqrt{\frac{4\pi}{3}} \begin{pmatrix} \tilde{\mathcal{Y}}_{1,1}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{1,0}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{1,-1}(\mathbf{x}) \end{pmatrix} &= \begin{pmatrix} x \\ z \\ y \end{pmatrix} \\ \sqrt{\frac{4\pi}{5}} \begin{pmatrix} \tilde{\mathcal{Y}}_{2,2}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{2,1}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{2,0}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{2,-1}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{2,-2}(\mathbf{x}) \end{pmatrix} &= \begin{pmatrix} \frac{\sqrt{3}}{2} (x^2 - y^2) \\ \sqrt{3}xz \\ \frac{1}{2} (2z^2 - x^2 - y^2) \\ \sqrt{3}yz \\ \sqrt{3}xy \end{pmatrix} \end{aligned}$$

$$\sqrt{\frac{4\pi}{7}} \begin{pmatrix} \tilde{\mathcal{Y}}_{3,3}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{3,2}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{3,1}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{3,0}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{3,-1}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{3,-2}(\mathbf{x}) \\ \tilde{\mathcal{Y}}_{3,-3}(\mathbf{x}) \end{pmatrix} = \begin{pmatrix} \frac{\sqrt{10}}{4}x(x^2 - 3y^2) \\ \frac{\sqrt{15}}{2}z(x^2 - y^2) \\ \frac{\sqrt{6}}{4}(4z^2 - x^2 - y^2)x \\ \frac{1}{2}z(2z^2 - 3x^2 - 3y^2) \\ \frac{\sqrt{6}}{4}(4z^2 - x^2 - y^2)y \\ \sqrt{15}xyz \\ \frac{\sqrt{10}}{4}y(3x^2 - y^2) \end{pmatrix}$$

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