Rotation Matrices for Real Spherical Harmonics. Direct Determination by Recursion

Joseph Ivanic and Klaus Ruedenberg*

Ames Laboratory, U.S. Department of Energy, and Department of Chemistry, Iowa State University, Ames, Iowa 50011

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A recurrence procedure is derived for constructing the rotation matrices between real spherical harmonics directly in terms of the elements of the original 3×3 rotation matrix without the intermediary of any parameters. The procedure furnishes a simple, efficient, and general method for the formal as well as numerical evaluation of these representation matrices.

1. Introduction

In the context of *atomic* calculations, *complex* spherical harmonics are more useful than real spherical harmonics because they provide a more effective basis for the full exploitation of all representational aspects of the full rotation group in the elucidation of atomic structure and spectra. The $(2l+1)\times(2l+1)$ representation matrices, according to which the complex spherical harmonics of order l transform among each other when the coordinate axes are rotated, therefore occupy a central place in the theory of atoms and have been studied in depth. Wigner's classical formulas express them in terms of the Euler angle parameters of the basic rotations.[1] Recently, Lynden-Bell and Stone[2] have pursued work with alternative parameters, namely those of Klein−Caley−Euler:

$$q_o = \cos \gamma/2 \qquad q_1 = d_1 \sin \gamma/2$$
$$q_2 = d_2 \sin \gamma/2 \qquad q_3 = d_3 \sin \gamma/2$$

where $\mathbf{d} = \{d_1,d_2,d_3\}$ is the unit vector along the axis of rotation, γ is the angle of the right-hand rotation around this axis, and $\sum_j q_j^2 = 1$. These four parameters can be considered as quaternion components and the algebra of the rotation group is known to be isomorphic to that of the quaternions.[3] Using the latter, these authors have formulated the complex rotation matrices $D^l_{mm'}$ in this parametrization.

In molecular calculations, on the other hand, real atomic orbitals are more useful than complex atomic orbitals as building blocks for molecular wave functions. This is because the latter are *intrinsically real* for the overwhelming majority of polyatomic molecules. In this context too, however, there arises a need for locally rotating the atomic orbitals from which a molecular wave function is constructed. We were confronted with this task in our work on the identification and analysis of atomic valence states in the context of molecular ab initio calculations. Spherical harmonics also form the basis for electrostatic multipole expansions in molecules so that their rotational transformations are encountered when multipoles are optimally oriented with respect to local molecular surroundings. In the context of molecular dynamics, too, rotations of spherical harmonics can be useful.

For all such rotations of real spherical harmonics, the representation matrices mentioned in the first paragraph present certain inconveniences: they apply to the complex rather than directly to the real harmonics; they require the expression of each rotation in terms of some parameter set such as, e.g., the Euler angles; and they are given by explicit expressions which increase in complexity for higher l-values such as are becoming more and more common for accurate molecular wave function expansions.

Nonetheless, we found only one publication dealing with the transformation matrices of real spherical harmonics, namely that by Sherman and Grinter[4] who give explicit expressions of these matrices in terms of Euler angles for d and f orbitals. The reason for this paucity is perhaps that only limited efforts have been expended so far on conceptual local analyses of ab initio calculations and that, for the sheer determination of the wave functions, all necessary operations, in particular the integral evaluations, can be carried out satisfactorily with unrotated parallel-displaced atomic orbitals. However, in as much as multipolar expansions may find wider use for energy integrals in large molecules, rotations of spherical harmonics may also prove valuable in that context.

Departing from the aforementioned traditional use of parametric representations, we noted that the rotation matrices for the real spherical harmonics of degree l must, in fact, be directly expressible as polynomials of degree l in terms of the elements of the original 3×3 rotation matrices. We therefore began by determining these polynomials by brute force: The real spherical harmonics were expressed in terms of homogeneous products of the Cartesian coordinates, the latter were transformed by the tensor products of the 3×3 rotation matrices, and finally, the rotated Cartesian products were back-transformed to the rotated spherical harmonics. Assisted by the algebraic manipulation program in Mathematica, we thus obtained explicit expressions for the rotation matrix elements of the d and f harmonics in terms of those of the p harmonics. Recognizing the increasing complexity of this procedure with increasing l values, however, we turned to the development of the recurrence procedure which is reported in this note.

About halfway through this work, we learned that A. J. Stone had recently also derived explicit formal expressions of higher order real rotation matrices in terms of those for p functions.[5] Starting from the complex representation matrices in the Klein−Caley parametrization[2] and using an algebraic manipulation program, he obtained explicit expressions of the real rotation matrices for d, f, and g harmonics in terms of the 3×3 rotation matrices. Dr. Stone graciously sent us a computer listing of his explicit formulas so that we had the benefit of a three-way check: his explicit expressions, our explicit expressions, and our recurrence scheme.

The recursive approach described below *avoids the detour over any parametrization of the rotations and it also bypasses the complex representation.* The relations derived in sections

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6 and 7 and summarized in section 8 express the elements $R^l_{mm'}$ of the rotation matrix for the real harmonics of order l *directly* in terms of those of order ($l-1$), i.e. $R^{l-1}_{mm'}$, and those of the original 3×3 rotation matrix, $R_{mm'}$. These recurrence relations can be applied with equal ease formalistically or numerically. Both kinds of usages are straightforward and, at the same time, open ended with regard to going up to arbitrary values of l. We believe that this recursion offers an extremely efficient and general access to the real rotation matrices.

2. Real Spherical Harmonics

For the purpose of the subsequent discussions, it is expedient to use the following notation regarding spherical coordinates

$$\mathbf{x} = (x,y,z) = r(\xi_1,\xi_{-1},\xi_0) = r\boldsymbol{\xi} \tag{2.1}$$

$$\xi_1 = \sin\theta\cos\phi \qquad \xi_{-1} = \sin\theta\sin\phi \qquad \xi_0 = \cos\theta \tag{2.2}$$

We define the real spherical harmonics as follows

$$Y_{lm}(\xi) = P_l^{|m|}(\cos\theta)\,\Phi_m(\phi) \tag{2.3}$$

where

$$\Phi_0 = (2\pi)^{-1/2}$$

$$\Phi_m = \pi^{-1/2}\cos m\phi \qquad \text{for } m > 0 \tag{2.4}$$

$$= \pi^{-1/2}\sin m\phi \qquad \text{for } m < 0$$

and

$$P_l^m(t) = \left[\frac{2l+1}{2}\,\frac{(l-m)!}{(l+m)!}\right]^{1/2} P_l^m(t) \tag{2.5a}$$

$$P_l^m(t) = (2^l l!)^{-1}(1-t^2)^{1/2}(\partial/\partial t)^{l+m}(t^2-1)^l \tag{2.5b}$$

with $t = \cos\theta$. The definition (2.5) is, in fact, also applicable for negative values of m and it is readily shown that, then,

$$P_l^{-m}(t) = (-1)^m P_l^m(t) \tag{2.6}$$

The real spherical harmonics form an orthonormal complete basis

$$\langle Y_{lm}|Y_{\lambda\mu}\rangle = \delta_{l\lambda}\delta_{m\mu} \tag{2.7}$$

for all integer values of m and μ, positive, negative, and zero.

3. Recurrence Relations for Real Spherical Harmonics

All subsequent derivations are based on the following three recurrence relations[6] for the normalized Legendre functions of (2.5):

$$\cos\theta\, P_l^m = A_l^m P_{l-1}^m + A_{l+1}^m P_{l+1}^m \tag{3.1}$$

with

$$A_l^m = [(l+m)(l-m)/(2l+1)(2l-1)]^{1/2} \tag{3.2}$$

and

$$\sin\theta\, P_l^m = B_l^m P_{l-1}^{m-1} - B_{l+1}^{-m+1} P_{l+1}^{m-1} \tag{3.3}$$

$$\sin\theta\, P_l^m = -B_l^{-m} P_{l-1}^{m+1} + B_{l+1}^{m+1} P_{l+1}^{m+1} \tag{3.4}$$

with

$$B_l^m = [(l+m)(l+m-1)/(2l+1)(2l-1)]^{1/2} \tag{3.5}$$

By virtue of (2.6), it is seen that, if (3.1) is valid for positive m values, then it is also valid for negative m values. On the other hand, with the help of (2.6), one deduces (3.4) for negative m values from (3.3) for positive m values and vice versa. Thus, (3.1), (3.3), and (3.4) are valid for all integer values of m, positive, negative, and zero.

Multiplication of (3.1) by the appropriate trigonometric factors, $\cos m\phi$ and $\sin m\phi$, yields the corresponding recurrence relation for the real spherical harmonics

$$\xi_0 Y_{lm} = A_l^m Y_{l-1,m} + A_{l+1}^m Y_{l+1,m} \tag{3.6}$$

for any integer value of m.

In the case of (3.3) and (3.4), we multiply these equations, for $m \geq 0$, by the identities

$$\cos\phi\cos m\phi \pm \sin\phi\sin m\phi = \cos(m \mp 1)\phi$$

$$\cos\phi\sin m\phi \pm \sin\phi\cos m\phi = \sin(m \pm 1)\phi$$

Attention must also be paid to the difference in the normalization factors for $m = 0$ and $m \neq 0$ [see (2.4)]. One obtains the following equations.

For $m = 0$:

$$\xi_1 Y_{l0} = \{-B_l^0 Y_{l-1,1} + B_{l+1}^1 Y_{l+1,1}\}/\sqrt{2} \tag{3.7}$$

$$\xi_{-1} Y_{l0} = \{-B_l^0 Y_{l-1,-1} + B_{l+1}^1 Y_{l+1,-1}\}/\sqrt{2} \tag{3.8}$$

For $m > 0$:

$$\xi_1 Y_{lm} + \xi_{-1} Y_{l,-m} = (1+\delta_{m1})^{1/2}\{B_l^m Y_{l-1,m-1} - B_{l+1}^{-m+1} Y_{l+1,m-1}\} \tag{3.9a}$$

$$\xi_1 Y_{lm} - \xi_{-1} Y_{l,-m} = -B_l^{-m} Y_{l-1,m+1} + B_{l+1}^{m+1} Y_{l+1,m+1} \tag{3.9b}$$

$$\xi_1 Y_{l,-m} + \xi_{-1} Y_{l,m} = -B_l^{-m} Y_{l-1,m+1} - B_{l+1}^{m+1} Y_{l+1,-m-1} \tag{3.9c}$$

$$\xi_1 Y_{l,-m} - \xi_{-1} Y_{l,m} = -(1-\delta_{m1})\{B_l^m Y_{l-1,-m+1} - B_{l+1}^{-m+1} Y_{l+1,-m+1}\} \tag{3.9d}$$

Addition and subtraction of the first two and the last two of these equations yields

$$2\xi_1 Y_{l,m} = (1+\delta_{m1})^{1/2}\{B_l^m Y_{l-1,m-1} - B_{l+1}^{-m+1} Y_{l+1,m-1}\} - B_l^{-m} Y_{l-1,m+1} + B_{l+1}^{m+1} Y_{l+1,m+1} \tag{3.10a}$$

$$2\xi_1 Y_{l,-m} = (1-\delta_{m1})\{B_l^m Y_{l-1,-m+1} - B_{l+1}^{m+1} Y_{l+1,-m+1}\} - B_l^{-m} Y_{l-1,-m-1} + B_{l+1}^{m+1} Y_{l+1,-m-1} \tag{3.10b}$$

$$2\xi_{-1} Y_{l,m} = (1-\delta_{m1})\{B_{l+1}^{-m+1} Y_{l+1,-m+1} - B_l^m Y_{l-1,-m+1}\} - B_l^{-m} Y_{l-1,-m-1} + B_{l+1}^{m+1} Y_{l+1,-m-1} \tag{3.11a}$$

$$2\xi_{-1} Y_{l,-m} = (1+\delta_{m1})^{1/2}\{B_l^m Y_{l-1,m-1} - B_{l+1}^{-m+1} Y_{l+1,m-1}\} + B_l^{-m} Y_{l-1,m+1} - B_{l+1}^{m+1} Y_{l+1,m+1} \tag{3.11b}$$

As mentioned above, *m is assumed to be a positive number in* (3.9), (3.10), and (3.11).

4. Integral Formulas

From the preceding recurrence relations, explicit expressions can be derived for the integrals $\langle Y_{l+1,m}|\xi_i|Y_{l,\mu}\rangle$ which will be needed in sections 6 and 7. From (3.6) one finds immediately

$$\langle Y_{l+1,m}|\xi_0|Y_{l\mu}\rangle = A^{\mu}_{l+1}\delta_{m\mu} \tag{4.1}$$

because of the orthonormality of the real spherical harmonics. For the same reasons, one finds from (3.7), (3.8), (3.10), and (3.11) that the *only nonvanishing* integrals involving ξ_1 and ξ_{-1} are the following:

$$\begin{array}{lll} m & \mu & \langle Y_{l+1,m}|\xi_1|Y_{l,\mu}\rangle \\ \geq 1 & m+1 & -B^{-m}_{l+1}/2 \\ \geq 1 & m-1 & (1+\delta_{m,1})^{1/2}B^{m}_{l+1}/2 \\ 0 & 1 & -B^{0}_{l+1}/\sqrt{2} \\ \leq -1 & m+1 & (1-\delta_{m,-1})B^{-m}_{l+1}/2 \\ \leq -1 & m-1 & -B^{m}_{l+1}/2 \end{array} \tag{4.2}$$

and

$$\begin{array}{lll} m & \mu & \langle Y_{l+1,m}|\xi_{-1}|Y_{l,\mu}\rangle \\ \geq 1 & -m-1 & -B^{-m}_{l+1}/2 \\ \geq 1 & -m+1 & -(1-\delta_{m,1})B^{m}_{l+1}/2 \\ 0 & -1 & -B^{0}_{l+1}/\sqrt{2} \\ \leq -1 & -m-1 & (1+\delta_{m,-1})^{1/2}B^{-m}_{l+1}/2 \\ \leq -1 & -m+1 & B^{m}_{l+1}/2 \end{array} \tag{4.3}$$

It is manifest that the integrals $\langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle$ are closely related to the Gaunt coefficients $\langle Y^{c}_{l+1,m}|Y^{c}_{1i}|Y^{c}_{l\mu}\rangle$, where the Y^{c}_{lm} denote complex spherical harmonics.[7]

5. Rotation of Spherical Harmonics

If the rotations and rotation−reflections of the orthonormal unit vectors along the *x,y,z* axis are given by

$$\hat{\mathbf{e}}_k = \sum_i \mathbf{e}_i R_{ik} \qquad \mathbf{R}^{\dagger} = \mathbf{R}^{-1} \tag{5.1}$$

then the coordinates of any vector in terms of these basis vectors will transform correspondingly. Since $r = (x^2 + y^2 + z^2)^{1/2}$ is invariant under such orthogonal transformations $\mathbf{R}$, these coordinate transformations yield the following transformations between the quantities ξ_1, ξ_{-1}, ξ_0 defined in (2.1) and (2.2):

$$\hat{\xi}_k = \sum_{i=-1}^{1} \xi_i R_{ik} \tag{5.2}$$

Since the real spherical harmonics for $l = 1$ are given by

$$Y_{1m}(\xi) = (3/4\pi)^{1/2}\xi_m \qquad m = 0, \pm 1 \tag{5.3}$$

they transform similarly under such axis rotations, i.e.,

$$\hat{Y}_{1m'} = \sum_{m=-1}^{1} Y_{1m}R^{1}_{mm'} \tag{5.4}$$

where we have defined

$$\hat{Y}_{lm} = Y_{lm}(\hat{\xi}) \tag{5.5}$$

and

$$R^{1}_{mm'} = R_{mm'} \tag{5.6}$$

For any other arbitrary l value, too, the spherical harmonics span a representation of the rotation−reflection group and, hence, transform among each other. In the case of complex spherical harmonics, Y^{c}_{lm}, the representation matrices are conventionally denoted as $\mathbf{D}^{l}_{mm'}$, i.e. we have

$$\hat{Y}^{c}_{lm'} = Y^{c}_{lm'}(\hat{\xi}) = \sum_{m=-l}^{l} Y^{c}{}_{lm}(\xi)\mathbf{D}^{l}_{mm'} = \sum_{m=-l}^{l} \mathrm{Y}^{c}{}_{lm}\mathbf{D}^{l}_{mm'} \tag{5.7}$$

and expressions for the complex, unitary matrices $\mathbf{D}^{l}$ have been given by Wigner in terms of the Euler angles through which the original rotation matrices $\mathbf{R}$ can be parametrically expressed.[1] Lynden-Bell and Stone have expressed them in terms of the Klein−Caley−Euler parameters.[2]

Here we are interested in the real orthogonal matrices R^{l} for the transformation of the real spherical harmonics, i.e.

$$\hat{Y}_{lm'} = \sum_{m=-l}^{l} Y_{lm}R^{l}_{mm'} \tag{5.8}$$

Since the real spherical harmonics of order l are homogeneous polynomials of degree l in ξ_1, ξ_{-1}, and ξ_0, it is apparent that the elements of $\mathbf{R}^{l}$ must be homogeneous polynomials of degree l in terms of the nine elements of the matrix $\mathbf{R}$. In the next two sections, we shall derive a direct construction of the $\mathbf{R}^{l}$ by recursion without the use of Euler angles or any other parameters. It is valid for the cases of det($\mathbf{R}$) = +1 as well as −1.

6. Recurrence Relations for the Rotation Matrices of Real Spherical Harmonics. I

The recurrence relations established in section 3 apply to the spherical harmonics in the rotated coordinate frame, i.e. $Y_{lm}(\hat{\xi}) = \hat{Y}_{lm}$, as well as to those in the unrotated coordinate frame, i.e. $Y_{lm}(\xi) = Y_{lm}$.

Multiplying (3.6) in the *rotated* frame by the *unrotated* $Y_{l+1,m}$ and integrating, one notes that the first term on the right hand side vanishes because, by virtue of (5.8), the rotated $\hat{Y}_{lm}$ can be expressed as linear combinations of the unrotated Y_{lm} which, in turn, are orthogonal to the $Y_{l+1,m}$. One obtains therefore

$$\langle Y_{l+1,m}|\hat{\xi}_0|\hat{Y}_{lm'}\rangle = A^{m'}_{l+1}\langle Y_{l+1,m}|\hat{Y}_{l+1,m'}\rangle \tag{6.1}$$

Expanding now the rotated quantities on both sides of this equation in terms of the unrotated quantities, we obtain the recurrence relation

$$R^{l+1}_{mm'} = (A^{m'}_{l+1})^{-1}\sum_i\sum_\mu \langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle R_{i0}R^{l}_{\mu m'} \tag{6.2}$$

which expresses the matrix $\mathbf{R}^{l+1}$ in terms of the matrices $\mathbf{R}^{l}$ and $\mathbf{R} = \mathbf{R}^{1}$.

The double summation present in (6.2) goes over $6l+3$ terms. However, by virtue of the explicit expressions derived in section 4 for the integrals occurring in (6.2), most of these terms vanish, leaving at most five non-zero terms. Thus inserting the expressions of (4.1), we find the following explicit formulas. For $m = 0$, we obtain

$$R^{l+1}_{0m'} = a^{l+1}_{mm'}R_{00}R^{l}_{0m'} - b^{l+1}_{mm'}(R_{10}R^{l}_{1m'} + R_{-10}R^{l}_{-1m'})/\sqrt{2} \tag{6.3}$$

where

$$a^{l}_{mm'} = A^{m}_{l}/A^{m'}_{l} = [(l+m)(l-m)/(l+m')(l-m')]^{1/2} \tag{6.4}$$

$$b^{l}_{mm'} = B^{m}_{l}/A^{m'}_{l} = [(l+m)(l+m-1)/(l+m')(l-m')]^{1/2} \tag{6.5}$$

For $m > 0$, we obtain

$$R^{l+1}_{mm'} = a^{l+1}_{mm'} R_{00} R^l_{mm'} + b^{l+1}_{mm'}[(1+\delta_{m1})^{1/2} R_{10} R^l_{m-1,m'} - (1-\delta_{m1}) R_{-10} R^l_{-m+1,m'}]/2 - b^{l+1}_{-mm'}[R_{10} R^l_{m+1,m'} + R_{-10} R^l_{-m-1,m'}]/2 \quad (6.6)$$

and

$$R^{l+1}_{-mm'} = a^{l+1}_{mm'} R_{00} R^l_{-mm'} + b^{l+1}_{mm'}[(1+\delta_{m1})^{1/2} R_{-10} R^l_{m-1,m'} - (1-\delta_{m1}) R_{10} R^l_{-m+1,m'}]/2 + b^{l+1}_{-mm'}[R_{-10} R^l_{m+1,m'} - R_{10} R^l_{-m-1,m'}]/2 \quad (6.7)$$

It is also possible to derive recurrence relations expressing the matrix $\mathbf{R}^{l-1}$ in terms of the matrices $\mathbf{R}^l$ and $\mathbf{R}$. By multiplying (3.6) in the *rotated* frame with the *unrotated* $Y_{l-1,m}$ and integrating, one obtains

$$\langle Y_{l-1,m}|\hat{\xi}_0|\hat{Y}_{lm'}\rangle = A^{m'}_l \langle Y_{l-1,m}|\hat{Y}_{l-1,m'}\rangle \quad (6.8)$$

Expanding, as before, the rotated quantities on both sides of this equation in terms of the unrotated quantities one finds the recurrence relation

$$R^{l-1}_{mm'} = (A^{m'}_l)^{-1} \sum_i \sum_\mu \langle Y_{l\mu}|\xi_i|Y_{l-1,m}\rangle R_{i0} R^l_{\mu m'} \quad (6.9)$$

The relations are however of lesser interest for two reasons. First, they are useless for the successive buildup of the representation matrices of order l from those of lower order. Secondly, while we know that the elements of $\mathbf{R}^{l-1}$ are expressible as polynomials to degree of $(l-1)$ in terms of the elements of $\mathbf{R}$, the direct application of (6.8) yields polynomials of degree $(l+1)$. To obtain the simpler expression, it is therefore necessary to simplify the resulting expressions by means of the orthonormality relations $\mathbf{R}\mathbf{R}^\dagger = \mathbf{R}^\dagger\mathbf{R} = \mathbf{I}$.

7. Recurrence Relations for the Rotation Matrices of Real Spherical Harmonics. II

The equations of the preceding section do not cover the cases whree $|m'| = l+1$ because the original recurrence relation (3.1) does not do so. Recursion formulas applicable to these cases are however obtained by analogous derivations starting from the earlier relations (3.7) to (3.10).

In analogy to (6.1), we deduce from (3.7) and (3.8) that

$$\langle Y_{l+1,m}|\hat{\xi}_1|\hat{Y}_{l0}\rangle = B^1_{l+1}\langle Y_{l+1,m}|\hat{Y}_{l+1,1}\rangle/\sqrt{2} \quad (7.1a)$$

$$\langle Y_{l+1,m}|\hat{\xi}_{-1}|\hat{Y}_{l0}\rangle = B^1_{l+1}\langle Y_{l+1,m}|\hat{Y}_{l+1,-1}\rangle/\sqrt{2} \quad (7.1b)$$

while from (3.9b) and (3.9c), we deduce for $m > 0$, that

$$\langle Y_{l+1,m}|\hat{\xi}_1|\hat{Y}_{lm'}\rangle - \langle Y_{l+1,m}|\hat{\xi}_{-1}|\hat{Y}_{l,-m'}\rangle = B^{m'+1}_{l+1}\langle Y_{l+1,m}|\hat{Y}_{l+1,m'+1}\rangle \quad (7.2a)$$

$$\langle Y_{l+1,m}|\hat{\xi}_1|\hat{Y}_{l,-m'}\rangle + \langle Y_{l+1,m}|\hat{\xi}_{-1}|\hat{Y}_{lm'}\rangle = B^{m'+1}_{l+1}\langle Y_{l+1,m}|\hat{Y}_{l+1,-m'-1}\rangle \quad (7.2b)$$

Expanding again all rotated quantities in these equations in terms of the unrotated quantities, now yields recurrence relations for $R^{l+1}_{m,m'+1}$ and $R^{l+1}_{m,-m'-1}$ in terms of $R^l_{mm'}$ and R_{ij}. From (7.1) we obtain

$$R^{l+1}_{m1} = (2^{1/2}/B^1_{l+1}) \sum_i \sum_\mu \langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle R_{i1} R^l_{\mu 0} \quad (7.3a)$$

$$R^{l+1}_{m,-1} = (2^{1/2}/B^1_{l+1}) \sum_i \sum_\mu \langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle R_{i,-1} R^l_{\mu 0} \quad (7.3b)$$

and from (7.2) we find, for $m' > 1$,

$$R^{l+1}_{m,m'+1} = (B^{m'+1}_{l+1})^{-1} \sum_i \sum_\mu \langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle [R_{i1} R^l_{\mu m'} - R_{i,-1} R^l_{\mu,-m'}] \quad (7.4a)$$

$$R^{l+1}_{m,-m'-1} = (B^{m'+1}_{l+1})^{-1} \sum_i \sum_\mu \langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle [R_{i1} R^l_{\mu,-m'} + R_{i,-1} R^l_{\mu m'}] \quad (7.4b)$$

These equations permit the calculation of the elements $R^{l+1}_{m,l+1}$ and $R^{l+1}_{m,-l-1}$ from $\mathbf{R}^l$ and $\mathbf{R}$, which were the cases not covered in the preceding section.

By inserting in (7.4a) the integral expressions of section 4 and taking account of the vanishing integrals, we obtain for $m = 0$:

$$R^{l+1}_{0,m'+1} = c^{l+1}_{0,m'+1}(R_{01} R^l_{0,m'} - R_{0,-1} R^l_{0,-m'}) - d^{l+1}_{0,m'+1}[R_{11} R^l_{1m'} - R_{1,-1} R^l_{1,-m'} + R_{-11} R^l_{-1m'} - R_{-1,-1} R^l_{-1,-m'}]/\sqrt{2} \quad (7.5)$$

where

$$c^l_{mm'} = A^m_l / B^{m'}_l = [(l+m)(l-m)/(l+m')(l+m'-1)]^{1/2} \quad (7.6)$$

$$d^l_{mm'} = B^m_l / B^{m'}_l = [(l+m)(l+m-1)/(l+m')(l+m'-1)]^{1/2} \quad (7.7)$$

For $m > 0$, we find

$$R^{l+1}_{m,m'+1} = c^{l+1}_{m,m'+1}(R_{01} R^l_{mm'} - R_{0,-1} R^l_{m,-m'}) + d^{l+1}_{m,m'+1}[(1+\delta_{m1})^{1/2}(R_{11} R^l_{m-1,m'} - R_{1,-1} R^l_{m-1,-m'}) - (1-\delta_{m1})(R_{-11} R^l_{-m+1,m'} - R_{-1,-1} R^l_{-m+1,-m'})]/2 - d^{l+1}_{-m,m'+1}[(R_{11} R^l_{m+1,m'} - R_{1,-1} R^l_{m+1,-m'}) + (R_{-11} R^l_{-m-1,m'} - R_{-1,-1} R^l_{-m-1,-m'})]/2 \quad (7.8a)$$

and

$$R^{l+1}_{-m,m'+1} = c^{l+1}_{m,m'+1}(R_{01} R^l_{-mm'} - R_{0,-1} R^l_{-m,-m'}) + d^{l+1}_{m,m'+1}[(1+\delta_{m1})^{1/2}(R_{-11} R^l_{m-1,m'} - R_{-1,-1} R^l_{m-1,-m'}) + (1-\delta_{m1})(R_{11} R^l_{-m+1,m'} - R_{1,-1} R^l_{-m+1,-m'})]/2 + d^{l+1}_{-m,m'+1}[(R_{-1,1} R^l_{m+1,m'} - R_{-1,-1} R^l_{m+1,-m'}) - (R_{11} R^l_{-m-1,m'} - R_{1,-1} R^l_{-m-1,-m'})]/2 \quad (7.8b)$$

We can similarly simplify (7.4b) by inserting the integrals of section 4. This yields for $m = 0$:

$$R^{l+1}_{0,-m'-1} = c^{l+1}_{0,m'+1}(R_{0,1}R^l_{0m'} + R_{0,1}R^l_{0,-m'}) - d^{l+1}_{0,m'+1}[R_{1,-1}R^l_{1m'} + R_{1,1}R^l_{1,-m'} + R_{-1,-1}R^l_{-1m'} + R_{-1,1}R^l_{-1,-m'}]/\sqrt{2} \quad (7.9a)$$

whereas for $m > 0$, we have

$$R^{l+1}_{m,-m'-1} = c^{l+1}_{m,m'+1}(R_{0,-1}R^l_{mm'} + R_{01}R^l_{m,-m'}) + d^{l+1}_{m,m'+1}[(1 + \delta_{m1})^{1/2}(R_{1,-1}R^l_{m-1,m'} + R_{11}R^l_{m-1,-m'}) - (1 - \delta_{m1})(R_{-1,-1}R^l_{-m+1,m'} + R_{-11}R^l_{-m+1,-m'})]/2 - d^{l+1}_{-m,m'+1}[(R_{1,-1}R^l_{m+1,m'} + R_{11}R^l_{m+1,-m'}) + (R_{-1,-1}R^l_{-m-1,m'} - R_{-11}R^l_{-m-1,-m'})]/2 \quad (7.9b)$$

and

$$R^{l+1}_{-m,-m'-1} = c^{l+1}_{m,m'+1}(R_{0,-1}R^l_{-mm'} + R_{01}R^l_{-m,-m'}) + d^{l+1}_{m,m'+1}[(1 + \delta_{m1})^{1/2}(R_{-1,-1}R^l_{m-1,m'} + R_{-1,1}R^l_{m-1,-m'}) + (1 - \delta_{m1})(R_{1,-1}R^l_{-m+1,m'} - R_{11}R^l_{-m+1,-m'})]/2 + d^{l+1}_{-m,m'+1}[(R_{-1,-1}R^l_{m+1,m'} + R_{-11}R^l_{m+1,-m'}) - (R_{1,-1}R^l_{-m-1,m'} + R_{11}R^l_{-m-1,-m'})]/2 \quad (7.9c)$$

In analogy to (6.8), one can also deduce recurrence relations for $R^{l-1}_{m,m'+1}$ and $R^{l-1}_{m,-m'-1}$ in terms of $R^l_{mm'}$ and R_{ij}.

From (3.7) and (3.8), one obtains

$$\langle Y_{l-1,m}|\hat{\xi}_1|\hat{Y}_{l0}\rangle = -B^0_l\langle Y_{l-1,m}|\hat{Y}_{l-1,1}\rangle/\sqrt{2} \quad (7.10a)$$

$$\langle Y_{l-1,m}|\hat{\xi}_{-1}|\hat{Y}_{l0}\rangle = -B^0_l\langle Y_{l-1,m}|\hat{Y}_{l-1,-1}\rangle/\sqrt{2} \quad (7.10b)$$

and from (3.9b) and (3.9c), we find for $m > 0$ that

$$\langle Y_{l-1,m}|\hat{\xi}_1|\hat{Y}_{lm'}\rangle - \langle Y_{l-1,m}|\hat{\xi}_{-1}|\hat{Y}_{l,-m'}\rangle = -B^{-m'}_l\langle Y_{l-1,m}|\hat{Y}_{l-1,m'+1}\rangle \quad (7.11a)$$

$$\langle Y_{l-1,m}|\hat{\xi}_1|\hat{Y}_{l,-m'}\rangle + \langle Y_{l-1,m}|\hat{\xi}_{-1}|\hat{Y}_{lm'}\rangle = -B^{-m'}_l\langle Y_{l-1,m}|\hat{Y}_{l-1,-m'-1}\rangle \quad (7.11b)$$

As before, we expand the rotated quantities in terms of the unrotated quantities. From (7.10), we obtain for $m = 0$:

$$R^{l-1}_{m1} = -(2^{1/2}/B^0_l)\sum_i\sum_\mu\langle Y_{l-1,m}|\xi_i|Y_{l,\mu}\rangle R_{i1}R^l_{\mu0} \quad (7.12a)$$

$$R^{l-1}_{m,-1} = -(2^{1/2}/B^0_l)\sum_i\sum_\mu\langle Y_{l-1,m}|\xi_i|Y_{l,\mu}\rangle R_{i,-1}R^l_{\mu0} \quad (7.12b)$$

and from (7.8) we obtain for $m > 0$:

$$R^{l-1}_{m,m'+1} = -(B^{-m'}_l)^{-1}\sum_i\sum_\mu\langle Y_{l-1,m}|\xi_i|Y_{l,\mu}\rangle[R_{i1}R^l_{\mu m'} - R_{i,-1}R^l_{\mu-m'}] \quad (7.13a)$$

$$R^{l-1}_{m,-m'-1} = -(B^{-m'}_l)^{-1}\sum_i\sum_\mu\langle Y_{l-1,m}|\xi_i|Y_{l,\mu}\rangle[R_{i1}R^l_{\mu-m'} + R_{i,-1}R^l_{\mu m'}] \quad (7.13b)$$

For the reasons discussed at the end of section 6, the relations (7.12) and (7.13) are again of less interest.

8. Implementation

The heart of our computer code is the calculation of the elements of $\mathbf{R}^l$ from those of $\mathbf{R}^{l-1}$ and $\mathbf{R}$. For all elements $R^l_{mm'}$ with $|m'| < l$, the recurrence relations (6.3) to (6.7) are used. For the elements $R^l_{m,l}$ and $R^l_{m,-l}$ the recurrence relations (7.5) to (7.9) are used. All cases can be cast in the following operational form:

$$R^l_{mm'} = u^l_{mm'}U^l_{mm'} + v^l_{mm'}V^l_{mm'} + w^l_{mm'}W^l_{mm'} \quad (8.1)$$

where the $u^l_{mm'}$, $v^l_{mm'}$, $w^l_{mm'}$ are numerical coefficients and $U^l_{mm'}$, $V^l_{mm'}$, $W^l_{mm'}$ are functions of $\mathbf{R}^{l-1}$ and $\mathbf{R}$. For the various values of l, m and m', they are defined by the expressions given in Tables 1 and 2.

Two programs were implemented: One for the calculation of $\mathbf{R}^l$ from $\mathbf{R}^{l-1}$ and $\mathbf{R}$; the other for the calculation of the

TABLE 1: Definitions of the Numerical Coefficients $u^l_{mm'}$, $v^l_{mm'}$, and $w^l_{mm'}$, Occurring in Eq 8.1

	$\|m'\| < l$	$\|m'\| = l$
$u^l_{mm'}$	$\left[\dfrac{(l+m)(l-m)}{(l+m')(l-m')}\right]^{1/2}$	$\left[\dfrac{(l+m)(l-m)}{(2l)(2l-1)}\right]^{1/2}$
$v^l_{mm'}$	$\dfrac{1}{2}\left[\dfrac{(1+\delta_{m0})(l+\|m\|-1)(l+\|m\|)}{(l+m')(l-m')}\right]^{1/2}(1-2\delta_{m0})$	$\dfrac{1}{2}\left[\dfrac{(1+\delta_{m0})(l+\|m\|-1)(l+\|m\|)}{(2l)(2l-1)}\right]^{1/2}(1-2\delta_{m0})$
$w^l_{mm'}$	$-\dfrac{1}{2}\left[\dfrac{(l-\|m\|-1)(l-\|m\|)}{(l+m')(l-m')}\right]^{1/2}(1-\delta_{m0})$	$-\dfrac{1}{2}\left[\dfrac{(l-\|m\|-1)(l-\|m\|)}{(2l)(2l-1)}\right]^{1/2}(1-\delta_{m0})$

TABLE 2: Definitions of the Functions $U^l_{mm'}$, $V^l_{mm'}$, and $W^l_{mm'}$ Occurring in Eq 8.1

	$m = 0$	$m > 0$	$m < 0$
$U^l_{mm'}$	${}_0P^l_{0m'}$	${}_0P^l_{mm'}$	${}_0P^l_{mm'}$
$V^l_{mm'}$	${}_1P^l_{1m'} + {}_{-1}P^l_{-1m'}$	${}_1P^l_{m-1,m'}(1+\delta_{m1})^{1/2} - {}_{-1}P^l_{-m+1,m'}(1-\delta_{m1})$	${}_1P^l_{m+1,m'}(1-\delta_{m,-1}) + {}_{-1}P^l_{-m-1,m'}(1+\delta_{m,-1})^{1/2}$
$W^l_{mm'}$	–	${}_1P^l_{m+1,m'} + {}_{-1}P^l_{-m-1,m'}$	${}_1P^l_{m-1,m'} - {}_{-1}P^l_{-m+1,m'}$

where the funcrtions ${}_iP^l_{\mu m'}$, are given in terms of the matrix elements R_{ij} and $R^{l-1}_{\mu,m'}$, as follows:

	$\|m'\| < l$	$m' = l$	$m' = -l$
${}_iP^l_{\mu m'}$	$R_{i,0}R^{l-1}_{\mu,m'}$	$R_{i,1}R^{l-1}_{\mu,m'} - R_{i,-1}R^{l-1}_{\mu,-m'}$	$R_{i,1}R^{l-1}_{\mu,m'} + R_{i,-1}R^{l-1}_{\mu,-m'}$

TABLE 3: Rotation Matrix for Spherical Harmonics with $l = 2$ in Terms of the Elements of R[a,b]

	$\hat{Y}_{2,0}$	$\hat{Y}_{2,1}$	$\hat{Y}_{2,-1}$	$\hat{Y}_{2,2}$	$\hat{Y}_{2,-2}$
$Y_{2,0}$	$(3R_{00}^2 - 1)/2$	$\sqrt{3}R_{01}R_{00}$	$\sqrt{3}R_{0,-1}R_{00}$	$\sqrt{3}(R_{01}^2 - R_{0,-1}^2)/2$	$\sqrt{3}R_{01}R_{0,-1}$
$Y_{2,1}$	$\sqrt{3}R_{10}R_{00}$	$R_{11}R_{00} + R_{10}R_{01}$	$R_{1,-1}R_{00} + R_{10}R_{0,-1}$	$R_{11}R_{01} - R_{1,-1}R_{0,-1}$	$R_{11}R_{0,-1} + R_{1,-1}R_{01}$
$Y_{2,-1}$	$\sqrt{3}R_{-10}R_{00}$	$R_{-11}R_{00} + R_{-10}R_{01}$	$R_{-1,-1}R_{00} + R_{-10}R_{0,-1}$	$R_{-11}R_{01} - R_{-1,-1}R_{0,-1}$	$R_{-11}R_{0,-1} + R_{-1,-1}R_{01}$
$Y_{2,2}$	$\sqrt{3}(R_{10}^2 - R_{-10}^2)/2$	$R_{11}R_{10} - R_{-11}R_{-10}$	$R_{1,-1}R_{10} - R_{-1,-1}R_{-10}$	$(R_{11}^2 - R_{1,-1}^2 - R_{-11}^2 + R_{-1,-1}^2)/2$	$R_{11}R_{1,-1} - R_{-11}R_{-1,-1}$
$Y_{2,-2}$	$\sqrt{3}R_{10}R_{-10}$	$R_{11}R_{-10} + R_{10}R_{-11}$	$R_{1,-1}R_{-10} + R_{10}R_{-1,-1}$	$R_{11}R_{-11} - R_{1,-1}R_{-1,-1}$	$R_{11}R_{-1,-1} + R_{1,-1}R_{-11}$

[a] $Y_{2,m} = Y_{2,m}(\xi)$, $\hat{Y}_{2,m} = Y_{2,m}(\hat{\xi})$, $\hat{\xi}_k = \sum_i \xi_i R_{ik}$. [b] In this table, the symbol $R_{mm'}^2$ means $(R_{mm'})^2$, i.e. the superscript denotes the square and not $l = 2$.

matrices $\mathbf{R}^l$ for *all* values $l = 0, 1, 2, ... L$ from a given 3×3 rotation matrix $\mathbf{R}$.

9. Discussion

If the recursion code is used to calculate all elements of one matrix $\mathbf{R}^l$, *for a given l*, from those of $\mathbf{R}$, then the number of numerical operations is comparable to that needed when the explicit formulas for $\mathbf{R}^l$ are used. In practice, one typically requires, however, the transformation matrices for *all l* values up to a some maximum value L. The accomplishment of this task by the recursion route will require considerably less operations than would be needed if all matrices were independently calculated by the explicit formulas.

In view of the simple structure of the recurrence relations, it is also straightforward to use them to construct explicit *formal expressions* for the rotation matrices in terms of the elements of the 3×3 matrix. As an example we display the formulas for the rotation matrix of the d functions in Table 3. It should be noted, however, that these explicit formulas are not unique. This is because the nine components R_{ij} are not independent but satisfy the six orthonormality conditions. The latter can therefore be used to modify and, sometimes, to simply the expressions for the $R^l_{mm'}$. For example, direct application of the recurrence relations to generate the element $R^2_{00} = \langle Y_{2,0}|\hat{Y}_{2,0}\rangle$ yields $R^2_{00} = (R_{00})^2 - [(R_{10})^2 + (R_{-10})^2]/2$. Subsequent application of the orthonormalization relations, $\mathbf{R}^\dagger\mathbf{R} = \mathbf{R}\mathbf{R}^\dagger = \mathbf{I}$ will then yield the simpler expression given in Table 3 in the upper left corner.

We conclude that, for the formal as well as the numerical evaluation of the rotation matrices between real spherical harmonics, the described recursion scheme offers an eminently practical route combining simplicity, efficiency, and generality.

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