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Rapid and stable determination of rotation matrices between spherical harmonics by direct recursion

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Recurrence relations are derived for constructing rotation matrices between complex spherical harmonics directly as polynomials of the elements of the generating 3×3 rotation matrix, bypassing the intermediary of any parameters such as Euler angles. The connection to the rotation matrices for real spherical harmonics is made explicit. The recurrence formulas furnish a simple, efficient, and numerically stable evaluation procedure for the real and complex representations of the rotation group. The advantages over the Wigner formulas are documented. The results are relevant for directing atomic orbitals as well as multipoles.
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I. INTRODUCTION

Spherical harmonics play important roles in many areas of theoretical and applied physics. In quantum chemistry, they occur for instance as factors of atomic orbitals and as factors in multipole expansions. Our current interest derives from their use in the identification of atoms in molecules and in the fast multipole method (FMM)[1] for the calculation of Fock matrices. In this as in many other contexts, it is often necessary or expedient to rotate the spatial coordinate axes and there then arises the need to express the spherical harmonics defined with respect to one coordinate axis system in terms of the spherical harmonics defined with respect to the other.

It follows from group theory[2] that the two sets of harmonics associated with the two axis systems are related to each other by a transformation that is *block-diagonal* with respect to the azimuthal quantum number l, i.e.,

$$\hat{Y}_{lm'} = \sum_{m=-1}^{l} Y_{lm} D^{l}_{mm'}, \tag{1.1}$$

for complex spherical harmonics Y_{lm} and

$$\hat{\boldsymbol{Y}}_{lm'} = \sum_{m=-l}^{l} \boldsymbol{Y}_{lm} R^{l}_{mm'}, \tag{1.2}$$

for real spherical harmonics[3] $\boldsymbol{Y}_{lm}$, where the summations do *not* go over l. The $D^{l}_{mm'}$ are complex unitary matrices and the $R^{l}_{mm'}$ are real orthogonal matrices. These so-called *rotation matrices* are determined by the 3×3 orthogonal matrix $\boldsymbol{R}$ that defines the original rotation between the basis vectors of the two axis systems,

$$\hat{\mathbf{e}}_k = \sum_i \mathbf{e}_i R_{ik}. \tag{1.3}$$

Manifestly, Y_{lm}, $\boldsymbol{Y}_{lm}$ refer to the basis $\mathbf{e}_i$ whereas $\hat{Y}_{lm}$, $\hat{\boldsymbol{Y}}_{lm}$ refer to the basis $\hat{\mathbf{e}}_k$. Wigner[2] has given explicit direct formulas for the elements of the complex rotation matrices $D^{l}_{mm'}$ in terms of the Euler angles of the matrix $\boldsymbol{R}$.

In practical applications, one typically deals *simultaneously* with the spherical harmonics of *all* azimuthal quantum numbers $l=1,2,3,...L$, where L is the length of some initially presumed expansion. In such a context, the application of Wigner's formulas is inefficient because it entails the independent calculation of the rotation matrices for every l, a procedure embodying a large amount of duplication in the calculation of factorial factors. It moreover loses significant figures for larger values of L. Recurrence relations with respect to m for fixed l were given by Edmonds[4] and recently implemented by White and Head-Gordon[5] for use in fast multipole method calculations. They are unstable in the vicinity of particular polar angles and, although the instability can be partially remedied through alternative algorithms, problems remain with regard to the consistent calculation of all terms to the same accuracy.

Ivanic and Ruedenberg[3] have recently shown that the rotation matrices between *real* spherical harmonics obey a set of recurrence relations that allow for a much more efficient determination of the $R^{l}_{mm'}$. Their analysis differs from the aforementioned approaches in two respects:

(i) It is based on the recognition that the elements of the rotation matrices $R^{l}_{mm'}$ can be directly expressed as polynomials of degree l in terms of the matrix elements R_{jk} of the original 3×3 axis rotation.

(ii) These polynomials can be obtained recursively because the elements $R^{l}_{mm'}$ can be represented as bilinear expressions in terms of the elements $R^{l-1}_{mm'}$ and the elements R_{jk}.

Since the original axis rotation $\boldsymbol{R}$ is typically defined in terms of a number of interatomic distance vectors in a molecule, this approach also avoids the detour over the Euler angles. The procedure has since been used for image analysis at the University of Uppsala, Sweden, and in electronic engineering at the University of Auckland, New Zealand.

In the present article, we establish the analogous system of recurrence relations for the *complex* rotation matrices $D^l_{mm'}$. The formal reasoning as well as the organization of the material follow the paper of Ivanic and Ruedenberg.[3] The required background mathematics is assembled and laid out in Secs. II to V. The heart of the investigation is Sec. VI which presents three sets of recursion relations, analogous to those in Ref. 3. The derivation is contained in Sec. VI B. For the execution of actual calculations, the complex identities of Sec. VI are transformed into real equations in Sec. VII. In Sec. VIII, the quantitative relations between the complex rotation matrices $D^l_{mm'}$ of Eq. (1.1) and the real rotation matrices $R^l_{mm'}$ of Eq. (1.2) are formalized. The final section provides information about the computational implementation and a documentation of the advantages of the new approach as regards speed and accuracy.

II. DEFINITION OF COMPLEX SPHERICAL HARMONICS

Using the spherical coordinate definition

$$(x,y,z)=r(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta), \tag{2.1}$$

and adopting the phase conventions of Condon and Shortley,[6] we define the complex spherical harmonics by the equations

$$Y_{lm}(\theta,\phi)=(-1)^m\boldsymbol{P}_l^m(\cos\theta)e^{im\phi}/\sqrt{2\pi}. \tag{2.2}$$

with the normalized Legendre functions as defined by Bethe[7]

$$\boldsymbol{P}_l^m(t)=\left[\frac{(2l+1)(l-m)!}{2(l+m)!}\right]^{1/2}P_l^m(t), \tag{2.3}$$

where $t=\cos\theta$ and the standard Legendre functions are given by

$$P_l^m(t)=\frac{1}{2^l l!}(1-t^2)^{m/2}\frac{d^{l+m}}{dt^{l+m}}(t^2-1)^l. \tag{2.4}$$

By virtue of these definitions, it is readily verified that

$$\boldsymbol{P}_l^{-m}(t)=(-1)^m\boldsymbol{P}_l^m(t). \tag{2.5}$$

Since the so-called ''solid'' spherical harmonics r^lY_{lm} are well known to be *homogeneous* polynomials of degree l in the Cartesian coordinates x,y,z, the ''surface'' harmonics Y_{lm} can be expressed as homogenous polynomials

$$Y_{lm}=Y_{lm}(\xi), \tag{2.6}$$

in terms of the complex components ξ of the real unit vector in the basis e_1,e_2,e_3,

$$\xi=(\xi_-,\xi_0,\xi_+), \tag{2.7}$$

where

$$\begin{aligned}\xi_+&=-e^{i\phi}\sin\theta/\sqrt{2}=(-x-iy)/\sqrt{2}r,\\ \xi_-&=e^{-i\phi}\sin\theta/\sqrt{2}=(x-iy)/\sqrt{2}r,\\ \xi_0&=\cos\theta=z/r,\end{aligned} \tag{2.8}$$

which satisfy

$$\xi_0^2-\xi_-\xi_+=|\xi_-|^2+|\xi_0|^2+|\xi_+|^2=(x^2+y^2+z^2)/r^2=1. \tag{2.9}$$

For example, for the azimuthal quantum number $l=1$, one has

$$\begin{aligned}Y_{11}(\xi)&=(4\pi/3)^{1/2}\xi_+,\\ Y_{1,-1}(\xi)&=(4\pi/3)^{1/2}\xi_-,\\ Y_{10}(\xi)&=(4\pi/3)^{1/2}\xi_0.\end{aligned} \tag{2.10}$$

Because of the identity (2.9), the homogeneous form (2.6) can of course be converted into various nonhomogeneous forms, but we shall consider these as ''nonstandard.'' In what follows, we shall always think of the spherical harmonics as the *standard* homogeneous functions (2.6) of the Cartesian coordinates ξ in a given frame defined by basis vectors $\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3$ [see Eq. (1.3)], rather than as functions of the corresponding angles θ and ϕ as is conventionally done.

III. RECURRENCE RELATIONS FOR COMPLEX SPHERICAL HARMONICS

All subsequent derivations are deduced from the following three recurrence relations[7] for the normalized Legendre functions of Eq. (2.2):

$$\cos\theta\,\boldsymbol{P}_l^m=A_l^m\boldsymbol{P}_{l-1}^m+A_{l+1}^m\boldsymbol{P}_{l+1}^m, \tag{3.1}$$

$$\sin\theta\,\boldsymbol{P}_l^m=B_l^m\boldsymbol{P}_{l-1}^{m-1}-B_{l+1}^{-m+1}\boldsymbol{P}_{l+1}^{m-1}, \tag{3.2}$$

$$\sin\theta\,\boldsymbol{P}_l^m=-B_l^{-m}\boldsymbol{P}_{l-1}^{m+1}+B_{l+1}^{m+1}\boldsymbol{P}_{l+1}^{m+1}, \tag{3.3}$$

where

$$A_l^m=\left[\frac{(l+m)(l-m)}{(2l+1)(2l-1)}\right]^{1/2}, \tag{3.4}$$

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

Multiplication of Eq. (3.1) by $(-1)^me^{im\phi}/\sqrt{2\pi}$ yields

$$\xi_0Y_{lm}=A_l^mY_{l-1,m}+A_{l+1}^mY_{l+1,m}, \tag{3.6}$$

and similar multiplications for Eqs. (2.3) and (2.4) yield

$$\sqrt{2}\xi_-Y_{lm}=-B_l^mY_{l-1,m-1}+B_{l+1}^{-m+1}Y_{l+1,m-1}, \tag{3.7}$$

$$\sqrt{2}\xi_+Y_{lm}=-B_l^{-m}Y_{l-1,m+1}+B_{l+1}^{m+1}Y_{l+1,m+1}. \tag{3.8}$$

Equations (3.1), (3.2), (3.3) as well as Eqs. (3.6), (3.7), (3.8) are valid for m being positive, negative, or zero. In accordance with the remarks at the end of Sec. II, we perceive Eqs. (3.6) to (3.8) as identities between the standard homogeneous polynomial representations of the Y_{lm} in terms of ξ_-,ξ_0,ξ_+.

IV. INTEGRAL FORMULAS

Explicit expressions will be needed in the subsequent sections for the transition moment integrals $\langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle$ where the implied integration is defined to extend over the unit sphere in the Cartesian space. By virtue of the orthonormality of the spherical harmonics, they are readily derived from the recurrence relations of the previous section. From Eq. (3.6) one obtains

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

and from Eqs. (3.7) and (3.8) follows that, for l given and fixed, the only nonvanishing integrals involving ξ_+ and ξ_- are

$$\langle Y_{l+1,m}|\xi_+|Y_{l\mu}\rangle = \frac{1}{\sqrt{2}} B_{l+1}^{\mu+1}\delta_{m,\mu+1}, \tag{4.2}$$

and

$$\langle Y_{l+1,m}|\xi_-|Y_{l\mu}\rangle = \frac{1}{\sqrt{2}} B_{l+1}^{-\mu+1}\delta_{m,\mu-1}. \tag{4.3}$$

Eqs. (4.1), (4.2), and (4.3) are valid for m being positive, negative, or zero.

V. ROTATION OF COMPLEX SPHERICAL HARMONICS

In the present section, we collect some elementary relations regarding rotation matrices that will be used in the subsequent sections.

A. Decomposition into real and imaginary parts

The rotation of the basis vectors in real three-dimensional space, introduced by Eq. (1.3), implies the following transformation between the coordinates of any one vector with respect to the two bases:

$$(\hat{x},\hat{y},\hat{z}) = (x,y,z)\begin{pmatrix} R_{xx} & R_{xy} & R_{xz} \\ R_{yx} & R_{yy} & R_{yz} \\ R_{zx} & R_{xy} & R_{zz} \end{pmatrix} = (x,y,z)\boldsymbol{R}. \tag{5.1}$$

By virtue of the invariance of $r=(x^2+y^2+z^2)^{1/2}$, the complex components of the unit vector with respect to the two bases transform therefore as follows:

$$\hat{\xi} = (\hat{\xi}_-,\hat{\xi}_0,\hat{\xi}_+) = (\xi_-,\xi_0,\xi_+)\boldsymbol{D} = \xi\boldsymbol{D}, \tag{5.2}$$

with

$$\boldsymbol{D} = \boldsymbol{F} + i\boldsymbol{G} \tag{5.3}$$

where the real and imaginary parts of $\boldsymbol{D}$ are given by the following expressions in terms of the elements of $\boldsymbol{R}$:

$$\begin{pmatrix} F_{-,-} & F_{-,0} & F_{-,+} \\ F_{0,-} & F_{0,0} & F_{0,+} \\ F_{+,-} & F_{+,0} & F_{+,+} \end{pmatrix} = \begin{pmatrix} (R_{yy}+R_{xx})/2 & R_{xz}/\sqrt{2} & (R_{yy}-R_{xx})/2 \\ R_{zx}/\sqrt{2} & R_{zz} & -R_{zx}/\sqrt{2} \\ (R_{yy}-R_{xx})/2 & -R_{xz}/\sqrt{2} & (R_{yy}+R_{xx})/2 \end{pmatrix}, \tag{5.4}$$

$$\begin{pmatrix} G_{-,-} & G_{-,0} & G_{-,+} \\ G_{0,-} & G_{0,0} & G_{0,+} \\ G_{+,-} & G_{+,0} & G_{+,+} \end{pmatrix} = \begin{pmatrix} (R_{yx}-R_{xy})/2 & R_{yz}/\sqrt{2} & -(R_{yx}+R_{xy})/2 \\ -R_{zy}/\sqrt{2} & 0 & -R_{zy}/\sqrt{2} \\ (R_{yx}+R_{xy})/2 & R_{yz}/\sqrt{2} & (R_{xy}-R_{yx})/2 \end{pmatrix}. \tag{5.5}$$

In analogy to Eq. (5.3), all complex rotation matrices of Eq. (1.1) can be resolved into their real and complex parts, i.e.,

$$D^l_{mm'} = F^l_{mm'} + iG^l_{mm'}. \tag{5.6}$$

Since, from Eqs. (1.1), (2.2), and (2.5) one readily deduces

$$D^l_{-m,-m'} = (-1)^{m+m'}(D^l_{mm'})^*, \tag{5.7}$$

it follows that

$$\begin{aligned} F^l_{-m,-m'} &= (-1)^{m+m'}F^l_{mm'}, \\ G^l_{-m,-m'} &= -(-1)^{m+m'}G^l_{mm'}. \end{aligned} \tag{5.8}$$

These general identities account in particular for the relationships seen to exist between the matrix elements in the case of Eqs. (5.4), (5.5).

B. General rotation matrices $D^l_{mm'}$ as homogeneous polynomials of $D_{mm'}$

In analogy to Eq. (2.6), the transformed harmonics on the left hand side of Eq. (1.1) are defined as the standard homogeneous polynomials in terms of the transformed complex coordinates $(\hat{\xi}_-,\hat{\xi}_0,\hat{\xi}_+)$, i.e.,

$$\hat{Y}_{lm} = Y_{lm}(\hat{\xi}). \tag{5.9}$$

The transformation (1.1) can therefore be determined by first substituting the transformation (5.2) into the polynomials given by Eq. (5.9) and, then, transforming back to the $Y_{lm}(\xi)$ using one of the possible inverses of Eq. (2.6). From this reasoning, it is apparent that the elements of the general rotation matrices $\boldsymbol{D}^l$ can be expressed as homogenous polynomials of degree l in terms of the elements of $\boldsymbol{D}$. They can thus be calculated directly from the elements of the rotation matrix $\boldsymbol{R}$ without the detour over the Euler angles. In particular, Eq. (2.10) shows that the matrix for $l=1$ is identical with the matrix $\boldsymbol{D}$ given by Eqs. (5.3) to (5.5), i.e.,

$$\boldsymbol{D}^1 = \boldsymbol{D}. \tag{5.10}$$

C. Rotation matrices as integrals

By virtue of Eq. (1.1), the rotation matrices can also be expressed as the integrals

$$D^l_{mm'} = \langle Y_{lm}|\hat{Y}_{lm'}\rangle, \tag{5.11}$$

where the integration goes over the invariant Cartesian unit sphere and the arguments of Y_{lm} and $\hat{Y}_{lm'}$, respectively are the components of the same unit vector relative to the two fixed Cartesian bases $\{\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3\}$ and $\{\hat{\mathbf{e}}_1,\hat{\mathbf{e}}_2,\hat{\mathbf{e}}_3\}$ that are connected by $\boldsymbol{R}$ [Eq. (13)].

VI. RECURRENCE RELATIONS FOR COMPLEX ROTATION MATRICES

As seen in the preceding section, the rotation matrix elements $D^l_{mm'}$ can be obtained as homogeneous polynomials of the elements of $\boldsymbol{D}$. We shall now build up these polynomials by recursion, starting with $\boldsymbol{D}^1=\boldsymbol{D}$. From the identities in Sec. III, a variety of recurrence relations can be deduced between the rotation matrices for the harmonics of order l and those of order $(l-1)$. Here, we derive three sets that prove useful for the quantitative evaluation of the matrices.

A. Recurrence relation for $(-l+1)\leqslant m'\leqslant(+l-1)$

This recurrence relation is derived from the recurrence relation (3.6) between spherical harmonics. It yields the recurrence relation

$$D^{l}_{mm'}=a^{l}_{m,m'}D_{00}D^{l-1}_{m,m'}+b^{l}_{m,m'}D_{10}D^{l-1}_{m-1,m'}+b^{l}_{-m,m'}D_{-10}D^{l-1}_{m+1,m'}, \tag{6.1}$$

for the rotation matrices, where

$$a^{l}_{mm'}=A^{m}_{l}/A^{m'}_{l}=\left[\frac{(l+m)(l-m)}{(l+m')(l-m')}\right]^{1/2}, \tag{6.2}$$

$$b^{l}_{mm'}=B^{m}_{l}/(\sqrt{2}A^{m'}_{l})=\left[\frac{(l+m)(l+m-1)}{2(l+m')(l-m')}\right]^{1/2}. \tag{6.3}$$

It should be noted that the cases $m'=\pm l$ are not covered and that

$$a^{l}_{mm'}=0 \quad \text{for} \quad m=\pm l, \tag{6.4}$$

$$b^{l}_{mm'}=0 \quad \text{for} \quad m=-l \quad \text{and} \quad m=-l+1 \tag{6.5}$$

which, in certain cases, eliminates one or two terms in Eq. (6.1).

B. Proof of Eq. (6.1)

The recurrence relation (3.6) applies to the spherical harmonics in the rotated coordinate frame as well as to those in the unrotated coordinate frame. Multiplying (3.6) in the *rotated* frame by the *unrotated* $Y_{l+1,m}$ and integrating over the unit sphere in Cartesian space, as discussed at the end of the last section, yields

$$\langle Y_{l+1,m}|\hat{\xi}_0|\hat{Y}_{lm'}\rangle=A^{m'}_{l}\langle Y_{l+1,m}|\hat{Y}_{l-1,m'}\rangle+A^{m'}_{l+1}\langle Y_{l+1,m}|\hat{Y}_{l+1,m'}\rangle. \tag{6.6}$$

The rotated quantities on both sides of this equation are now expanded in terms of the unrotated quantities, using Eq. (1.1) for the harmonics and Eq. (5.2) for $\hat{\xi}_0$. It is noted that the first term on the right hand side vanishes, because the rotated $\hat{Y}_{l-1,m}$ can be expressed as linear combinations of the unrotated $Y_{l-1,m}$ which, in turn, are orthogonal to the $Y_{l+1,m}$. One obtains therefore

$$D^{l+1}_{mm'}=(A^{m'}_{l+1})^{-1}\sum_{i=-}^{+}\sum_{\mu=-l}^{l}\langle Y_{l+1,m}|\xi_i|Y_{l\mu}\rangle D_{i0}D^{l}_{\mu,m'}, \tag{6.7}$$

which expresses the matrix $\boldsymbol{D}^{l+1}$ in terms of the matrices $\boldsymbol{D}^{l}$ and $\boldsymbol{D}$. Inserting now, for the moment integrals occurring in this equation, the explicit expressions derived in Sec. IV, one finds, for any value of m, the formula

$$D^{l+1}_{mm'}=b^{l+1}_{-m,m'}D_{-10}D^{l}_{m+1,m'}+b^{l+1}_{m,m'}D_{10}D^{l}_{m-1,m'}+a^{l+1}_{m,m'}D_{00}D^{l}_{m,m'}, \tag{6.8}$$

where the coefficients are those defined by Eqs. (6.2) and (6.3). Replacement of l by $(l-1)$ in this equation yields Eq. (6.1).

C. Recurrence relation for $-l\leqslant m'\leqslant(l-2)$

This recurrence relation is derived from the recurrence relation (3.7) for spherical harmonics. Starting with Eq. (3.7), a derivation that is entirely analogous to that just discussed yields

$$D^{l}_{m,m'}=c^{l}_{m,-m'}D_{0,-1}D^{l-1}_{m,m'+1}+d^{l}_{m,-m'}D_{1,-1}D^{l-1}_{m-1,m'+1}+d^{l}_{-m,-m'}D_{-1,-1}D^{l-1}_{m+1,m'+1}, \tag{6.9}$$

where

$$c^{l}_{mm'}=\sqrt{2}A^{m}_{l}/B^{m'}_{l}=\left[\frac{2(l+m)(l-m)}{(l+m')(l+m'-1)}\right]^{1/2}, \tag{6.10}$$

$$d^{l}_{mm'}=B^{m}_{l}/B^{m'}_{l}=\left[\frac{(l+m)(l+m-1)}{(l+m')(l+m'-1)}\right]^{1/2}. \tag{6.11}$$

It should be noted that the case $m'=l$ and $m'=(l-1)$ are not covered by Eq. (6.8) and that

$$c^{l}_{mm'}=0 \quad \text{for} \quad m=\pm l, \tag{6.12}$$

$$d^{l}_{mm'}=0 \quad \text{for} \quad m=-l \quad \text{and} \quad m=(-l+1). \tag{6.13}$$

D. Recurrence relation for $(-l+2)\leqslant m'\leqslant +l$

This recurrence relation is derived from the recurrence relation (3.8) for spherical harmonics. Starting with Eq. (3.8), a derivation that is entirely analogous to that used in Sec. VI B yields

$$D^{l}_{m,m'}=c^{l}_{m,m'}D_{0,1}D^{l-1}_{m,m'-1}+d^{l}_{m,m'}D_{11}D^{l-1}_{m-l,m'-1}+d^{l}_{-m,m'}D_{-1,1}D^{l-1}_{m+1,m'-1}, \tag{6.14}$$

where the coefficients $c^{l}_{mm'}$ and $d^{l}_{mm'}$ are again those defined by Eqs. (6.10) and (6.11). This recurrence relation does not cover the cases $m'=-l$ and $m'=(-l+1)$.

E. Comment

A knowledgeable referee has called the author's attention to a very general relationship between Wigner $\boldsymbol{D}$-functions with arbitrary sub- and superscripts which is listed, for instance, as Eq. (5) in Sec. 4.6.2 of the comprehensive formula collection for the quantum theory of angular momenta by Varshalovich *et al.*[8] Due to its complete generality, this identity is complex, containing numerous Clebsch–Gordan coefficients. For the case that three superscripts in this equation are chosen as $j_1=1$, $j_2=j-1$, $j_3=j$, the general equation collapses in fact into our relations (6.1), (6.9), (6.14). It is because this case is so elementary, that we were able to reach the recurrence relations given here by a line of reasoning that is simpler than the body of derivations required to establish the general theory of the Wigner $\boldsymbol{D}$-functions. One has to assume that it is because of the large number of general formulas in that theory and because of the complexity of most of them that the practical usefulness of our particular identities for evaluating rotation matrices has so far escaped notice.

VII. OPERATIONAL ALGORITHM

In the context of the practical quantitative use of rotation matrices, evaluations of real quantities are ultimately required. For the numerical execution it is therefore advantageous to recast the complex recurrence scheme of the preceding section in terms of a real recurrence scheme for the real and imaginary components $\boldsymbol{F}$ and $\boldsymbol{G}$ introduced through Eq. (5.6). This is accomplished by inserting this resolution as well as Eq. (5.3) into Eqs. (6.1), (6.9), (6.14) and then separating the real and the imaginary parts. The resulting formulas become more transparent by use of the intermediary quantities

$$H^l_{m.m'}(i,j)=F_{ij}F^{l-1}_{m,m'}-G_{ij}G^{l-1}_{m,m'}, \tag{7.1}$$

$$K^l_{m,m'}(i,j)=F_{ij}G^{l-1}_{m,m'}+G_{ij}F^{l-1}_{m,m'}, \tag{7.2}$$

where the F_{ij} and G_{ij} are defined in Eqs. (5.4) and (5.5). With these definitions, one deduces from Eqs. (6.1), (6.9), (6.14) the following recurrence relations for $F^l_{mm'}$ and $G^l_{mm'}$.

A. Recurrence relation for $(-l+1)\leqslant m'\leqslant(+l-1)$

$$F^l_{mm'}=a^l_{m,m'}H^l_{m,m'}(0,0)+b^l_{m,m'}H^l_{m-1,m'}(+,0)+b^l_{-m,m'}H^l_{m+1,m'}(-,0), \tag{7.3}$$

$$G^l_{mm'}=a^l_{m,m'}K^l_{m,m'}(0,0)+b^l_{m,m'}K^l_{m-1,m'}(+,0)+b^l_{-m,m'}K^l_{m+1,m'}(-,0). \tag{7.4}$$

B. Recurrence relation for $-l\leqslant m'\leqslant(l-2)$

$$F^l_{m,m'}=c^l_{m,-m'}H^l_{m,m'+1}(0,-)+d^l_{m,-m'}H^l_{m-1,m'+1}(+,-)+d^l_{-m,-m'}H^l_{m+1,m'+1}(-,-), \tag{7.5}$$

$$G^l_{m,m'}=c^l_{m,-m'}K^l_{m,m'+1}(0,-)+d^l_{m,-m'}K^l_{m-1,m'+1}(+,-)+d^l_{-m,-m'}K^l_{m+1,m'+1}(-,-). \tag{7.6}$$

C. Recurrence relation for $(-l+2)\leqslant m'\leqslant+l$

$$F^l_{m,m'}=c^l_{m,m'}H^l_{m,m'-1}(0,+)+d^l_{m,m'}H^l_{m-1,m'-1}(+,+)+d^l_{-m,m'}H^l_{m+1,m'-1}(-,+), \tag{7.7}$$

$$G^l_{m,m'}=c^l_{m,m'}K^l_{m,m'-1}(0,+)+d^l_{m,m'}K^l_{m-1,m'-1}(+,+)+d^l_{-m,m'}K^l_{m+1,m'-1}(-,+). \tag{7.8}$$

The coefficients $a^l_{mm'},b^l_{mm'},c^l_{mm'},d^l_{mm'}$ occurring in these equations are those defined by Eqs. (6.2) to (6.5) and (6.10) to (6.13). We note that every one of them is of the form $(\alpha\cdot\beta/\gamma\cdot\delta)$ where α, β, γ, δ are all square-roots of integers not larger than $(2L+1)$ with L being the largest azimuthal quantum number considered.

VIII. RELATION TO REAL SPHERICAL HARMONICS AND THEIR ROTATION MATRICES

In agreement with most authors,[9] Ivanic and Ruedenberg[3] define the real spherical harmonics as

$$\boldsymbol{Y}_{lm}(\theta,\phi)=\boldsymbol{P}^{|m|}_l(\cos\theta)\Phi_m(\phi), \tag{8.1}$$

where

$$\begin{aligned}\Phi_m(\phi)&=(2\pi)^{-1/2} && m=0,\\ \Phi_m(\phi)&=\pi^{-1/2}\cos m\phi && m>0,\\ \Phi_m(\phi)&=\pi^{-1/2}\sin m\phi && m<0.\end{aligned} \tag{8.2}$$

They are related to the complex harmonics of Eq. (2.2) by

$$(Y_{l,-l}\cdots Y_{l,-m}\cdots Y_{l,0}\cdots Y_{l,m}\cdots Y_{l,l})=(\boldsymbol{Y}_{l,-l}\cdots\boldsymbol{Y}_{l,-m}\cdots\boldsymbol{Y}_{l,0}\cdots\boldsymbol{Y}_{l,m}\cdots Y_{l,l})\boldsymbol{W}^l, \tag{8.3}$$

where m is presumed to be a positive integer and the unitary matrices $\boldsymbol{W}^l$ are

$$\boldsymbol{W}^l=\begin{pmatrix} W_{-l,-l} & & & & & & W_{-l,l}\\ & \ddots & & & & ⋰ & \\ & & W_{-m,-m} & & W_{-m,m} & & \\ & & & W_{0,0} & & & \\ & & W_{m,-m} & & W_{m,m} & & \\ & ⋰ & & & & \ddots & \\ W_{l,-l} & & & & & & W_{l,l}\end{pmatrix}. \tag{8.4}$$

TABLE I. Accuracy of various methods for calculating matrix elements $D^l_{0,0}$ for the Euler angle β=90°.

	l=30	l=40	l=50	l=100
Wigner, exact[a]	−0.144 464 448 094 368	0.125 370 687 619 579	−0.112 275 172 659 217	0.079 589 237 387 178 7
Recursion, d.p.[b]	−0.144 464 448 094 367	0.125 370 687 619 579	−0.112 275 172 659 217	0.079 589 237 387 178 6
Wigner, d.p.[c,d]	−0.144 464 *454 621 912*	0.125 37*1 770 238 445*	−0.1*01 160 073 432 271*	Inaccessible[e]

[a]Calculated by Eq. (9.1) using MATHEMATICA[10] with the specification of 15 or more significant figures for the result.
[b]Calculated by the recursion procedure of Sec. VII of this paper in double precision arithmetic.
[c]Calculated using a Wigner formula program in double precision arithmetic (see Acknowledgments).
[d]Incorrect numbers due to loss of significant figures are indicated by underlined italics.
[e]Because in excess of 16 significant figures are lost.

As indicated, they contain nonzero elements only on the two diagonals; all other elements vanish. The nonvanishing elements are given by

$$W_{00}=1, \tag{8.5}$$

and for $m>0$:

$$\begin{pmatrix} W_{-m,-m} & W_{-m,m} \\ W_{m,-m} & W_{m,m} \end{pmatrix} = \begin{pmatrix} -i/\sqrt{2} & i(-1)^m/\sqrt{2} \\ 1/\sqrt{2} & (-1)^m/\sqrt{2} \end{pmatrix}. \tag{8.6}$$

It follows that the complex rotation matrices $\boldsymbol{D}^l$ of the present investigation and the rotation matrices $\boldsymbol{R}^l$ for the real spherical harmonics[3] are related by the similarity transformation

$$\boldsymbol{D}^l=(\boldsymbol{W}^l)^\dagger \boldsymbol{R}^l \boldsymbol{W}^l, \tag{8.7}$$

where $(\boldsymbol{W}^l)^\dagger$ denotes the hermitian conjugate of $\boldsymbol{W}^l$. Insertion of Eq. (5.8) into the left hand side of Eq. (8.7) and of Eqs. (8.4), (8.5), (8.6) into the right hand side of Eq. (8.7) yields, after separation of the real and imaginary parts, the relations

$$2F^l_{mn}=\alpha_m\alpha_n R^l_{|m|,|n|}+\beta_m\beta_n R^l_{-|m|,-|n|}, \tag{8.8}$$

$$2G^l_{mn}=\alpha_m\beta_n R^l_{|m|,-|n|}-\beta_m\alpha_n R^l_{-|m|,|n|}, \tag{8.9}$$

where m and n can be positive, zero, or negative and the factors α_m, β_m are given by

$$\begin{aligned} \alpha_m &= [(1+\delta_{m0})(-1)^{(|m|+m)}]^{1/2}, \\ \beta_m &= \mathrm{sign}(m)(1-\delta_{mo})\alpha_m. \end{aligned} \tag{8.10}$$

The inverse identities are

$$R^l_{m,n}=\alpha_m\alpha_n F^l_{m,n}-\beta_m\beta_{-n}F^l_{m,-n}, \tag{8.11}$$

$$R^l_{-m,-n}=\alpha_m\alpha_n F^l_{m,n}+\beta_m\beta_{-n}F^l_{m,-n}, \tag{8.12}$$

$$R^l_{m,-n}=\alpha_m\beta_n G^l_{m,n}-\beta_m\alpha_{-n}G^l_{m,-n}, \tag{8.13}$$

$$R^l_{-m,n}=-\alpha_m\beta_n G^l_{m,n}-\beta_m\alpha_{-n}G^l_{m,-n}, \tag{8.14}$$

where m and n are presumed to denote nonnegative integers.

It should be noted that the matrix $\boldsymbol{R}^l$ for $l=1$ is not identical with the matrix $\boldsymbol{R}$ of Eq. (5.1) but differs from it by a permutation of rows and columns as follows

$$\begin{pmatrix} R^1_{-1,-1} & R^1_{-1,0} & R^1_{-1,1} \\ R^1_{0,-1} & R^1_{0,0} & R^1_{0,1} \\ R^1_{1,-1} & R^1_{1,0} & R^1_{1,1} \end{pmatrix} = \begin{pmatrix} R_{yy} & R_{yz} & R_{yx} \\ R_{zy} & R_{zz} & R_{zx} \\ R_{xy} & R_{xz} & R_{xx} \end{pmatrix}. \tag{8.15}$$

IX. IMPLEMENTATION AND ASSESSMENT

In the implementation of the code, we have used Eqs. (7.3), (7.4) to calculate all matrix elements with $|m'|\neq l$. Equations (7.5), (7.6) were used to calculate the matrix elements with $m'=-l$, and Eqs. (7.7), (7.8) were used to calculate the matrix elements with $m'=+l$. The program input consists of the axis rotation matrix $\boldsymbol{R}$ defined by Eq. (5.1) and the highest quantum number L. The program then finds the real and imaginary parts of the complex rotation matrices $D^l_{mm'}$ as well as using Eqs. (8.11) to (8.14), the real rotation matrices $R^l_{mm'}$, for all quantum numbers $l=0,1,2,3,...L$. The repeated calculation of square-roots is avoided by generating a square-root table for all integers up to $(2L+1)$ before beginning the recursion.

The accuracy of the program was tested by comparing our quantitative results with those obtained with the Ivanic–Ruedenberg program[3] for rotation matrices of real spherical harmonics for a number of cases. The results found by the two methods agreed to 14 significant figures up to $L=40$. Since the two procedures go through very different sequences of extended numerical arithmetic, it can be inferred that no significant figures are lost by either one of the two algorithms. We confirmed this inference by additionally calculating the elements to sufficient accuracy using MATHEMATICA[10] and Wigner's formulas. We also found that the identities (5.8) were satisfied to full accuracy by the values of $F^{40}_{mm'}$ and $F^{40}_{-m,-m'}$.

Computations with the Wigner formulas have the problem that the $D^l_{mn'}$, all of which have absolute values less than unity, are obtained as sums of very large positive and negative numbers. A transparent example is the expression for $m=m'=0$ (which is independent of α and γ):

$$\begin{aligned} &D^l_{00}(\alpha,\beta,\gamma)=\sum_k (-1)^k[l!/k!(l-k)!]^2 x^k(1-x)^{l-k}, \\ &x=\sin^2\beta/2. \end{aligned} \tag{9.1}$$

Its examination (using Stirling's formula) shows that, for large l and β near $\pi/2$, a loss of approximately $[\log(2^{l+1}/\pi l)]$ significant figures (where log denotes the decimal logarithm) is to be expected, suggesting a loss of about 7, 10, 13, 16, and 29 significant figures for l=30, 40, 50, 60, and 100, respectively. These predictions are in fact confirmed by a comparison of the numerical results displayed in the three rows of Table I which list D^l_{00} values obtained in three dif-

TABLE II. Execution times and loss of significant figures for Wigner formulas. (Euler angles $\alpha=\beta=\gamma=\pi/4$).

L =Highest Value of l	Ratio of CPU Times[a]	Matrix elements $F^{l}_{mm'}$ with largest errors for $l=L$: Magnitudes of Largest Errors[b]	Number of Elements[b]	Magnitudes of Elements
5	16	0.6 to 6×10^{-16}	70	0 to 0.5
10	25	0.6 to 6×10^{-15}	71	0.03 to 0.3
20	47	0.2 to 2×10^{-12}	140	0.01 to 0.2
30	77	0.1 to 1×10^{-9}	141	0.004 to 0.2
40	115	0.5 to 5×10^{-7}	191	0.002 to 0.1

[a](Execution time for Wigner method/execution time for present method.) The time is that used for the calculation of *all* rotation matrices up to L.
[b]Only the matrices for the largest values $l=L$ are considered. Calculations are executed with a machine accuracy of 15 significant figures. The errors quoted are the differences between the results obtained by using the Wigner formulas and the present method.

ferent ways, viz: (i) From Eq. (9.1) with MATHEMATICA,[10] exact to 15 significant figures, (ii) with our recurrence procedure of Sec. VII executed in double-precision arithmetic, and (iii) with a general Wigner formula program executed in double-precision arithmetic. Striking in particular are the results for $l=100$ inasmuch as they imply that no significant figures whatsoever are lost by our recursion in a case where, even with quadruple precision arithmetic, all significance is lost by the Wigner representation.

A comparison of the performance, regarding speed as well as accuracy, of the present method with that of the evaluation by Wigner's formulas is provided by Table II which lists some statistics for the rotation matrices for $L=5$, 10, 20, 30, 40, deduced from the rotation $\boldsymbol{R}$ with the Euler angles $\alpha=\beta=\gamma=\pi/4$. Displayed are the ratios of the execution times (for the evaluation of all matrices from $l=1$ to $l=L$) taken by the two methods, as well as some information pertaining to those matrix elements for the highest value $l=L$ which have the largest errors in the Wigner procedure. Listed are the orders-of-magnitude of the largest errors found, the number of elements having such an error, and the magnitude of the elements themselves. Discrepancies of the same order-of-magnitude were also found when inserting matrix elements obtained by the Wigner method into the identities (5.8).

The quoted quantitative results exhibit the advantages of the described recursion.

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