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The relationship of the hyperspherical harmonics to *SO*(3), *SO*(4) and orientation distribution functions

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The expansion of an orientation distribution function as a linear combination of the hyperspherical harmonics suggests that the analysis of crystallographic orientation information may be performed entirely in the axis–angle parameterization. Practical implementation of this requires an understanding of the properties of the hyperspherical harmonics. An addition theorem for the hyperspherical harmonics and an explicit formula for the relevant irreducible representatives of $SO(4)$ are provided. The addition theorem is useful for performing convolutions of orientation distribution functions, while the irreducible representatives enable the construction of symmetric hyperspherical harmonics consistent with the crystal and sample symmetries.

1. Introduction

The materials science community, for the most part, describes the orientation of a crystal by a triplet of Euler angles, and an orientation distribution function (ODF) as a linear combination of the generalized spherical harmonics (Bunge, 1993). Although alternative descriptions for the orientations of individual crystals appear throughout the literature, the general sentiment of the field has been that there is no alternative to the analytical expression of an ODF as a linear combination of the generalized spherical harmonics. This continues to encourage the use of Euler angles, despite repeated observations of the benefits of using the other descriptions (Grimmer, 1974; Frank, 1988; Heinz & Neumann, 1991; Morawiec & Field, 1996). Recently, Mason & Schuh (2008) pointed out that an alternative analytical expression for the ODF could be derived from the description of crystal orientations as normalized quaternions. By interpreting quaternions as vectors in a four-dimensional vector space over the field of real numbers, a collection of crystal orientations may be mapped to a collection of points residing on the unit sphere S^3 in four dimensions. Since the harmonic functions on S^3 provide a complete orthonormal basis for the expansion of a square-integrable function, the ODF describing these orientations is written as a linear combination of harmonic functions in the form

$$f(\omega,\theta,\varphi)=\sum_{n=0,2\ldots}^{\infty}\sum_{l=0}^{n}\sum_{m=-l}^{l} z^n_{l,m} Z^n_{l,m}(\omega,\theta,\varphi), \qquad (1)$$

where the function $Z^n_{l,m}(\omega,\theta,\varphi)$ is one of the hyperspherical harmonics.[1] The index n is restricted to even integers by the trivial symmetry of three-dimensional space, and the angles $0\le\omega\le 2\pi$, $0\le\theta\le\pi$ and $0\le\varphi<2\pi$ correspond to the angle of rotation and the polar and azimuthal angles of the axis of rotation, respectively, of a rotation of three-dimensional space.

The fact that equation (1) gives the ODF as a function of quantities relating directly to the angle and axis of rotation is one of the more noticeable benefits of this expansion compared with the generalized spherical harmonic expansion. A description of rotations by an angle and axis is not only more intuitive (Frank, 1988), but describing rotation distributions as functions of these quantities allows the derivation of certain results that are practically inaccessible within the generalized spherical harmonic formalism. For instance, the derivation of the misorientation angle distribution function is, in principle, as simple as writing the expansion of the misorientation distribution function (MDF) and integrating out the axis information. Nevertheless, analytic expressions for the misorientation angle distribution function appear in the literature only for the case of random textures (Handscomb, 1957; Mackenzie, 1958; Grimmer, 1979; Morawiec, 1995). This absence may historically be attributed to the purely practical difficulty of separating the misorientation angle dependence from an MDF expressed as a function of Euler angles, although calculating the misorientation angle distribution function from an MDF expressed in the form of equation (1) is as simple as integrating out the dependence on θ and φ.

More generally speaking, while the expansion of an ODF as a linear combination of basis functions gives an analytical expression to an otherwise arbitrary square-integrable function, the main motivation for this technique is that the expanded function inherits the properties of the basis functions. That is, the expansion allows the well known properties

[1] The arrangement of indices on the hyperspherical harmonics differs from that of Mason & Schuh (2008) owing to the consideration that the index n identifies the set of hyperspherical harmonics that form a basis for an irreducible representation of $SO(4)$, while the indices l and m identify individual members of this set; this difference in significance and function encourages the separation of n from l and m.

of the basis functions to be used in the analysis of an experimental ODF. Since the use of the hyperspherical harmonics as basis functions has only recently been proposed, there is a need to improve our understanding of the properties of the hyperspherical harmonics and thereby to more completely realize the utility of the expansion in equation (1).

Specifically, the first purpose of this paper is to derive an addition theorem for the hyperspherical harmonics. Practically speaking, the addition theorem simplifies the summation of products of the hyperspherical harmonics, and is expected to allow the convolution of ODFs as is required to calculate distribution functions of orientation differences. The second purpose of this paper is to provide explicit expressions for the matrices that determine the effect of rotations on the hyperspherical harmonics. Since a three-dimensional rotation of the sample changes the crystal orientations relative to the reference orientation, this operation changes the basis functions of the expansion of the ODF as well. More specifically, a three-dimensional rotation of the sample engenders a four-dimensional rotation of the hyperspherical harmonics. The effect of this rotation on the hyperspherical harmonics is given in matrix form by the irreducible representatives of $SO(4)$, the four-dimensional rotation group. An explicit expression for the irreducible representatives allows, for example, the reference orientation of a collection of orientation measurements to be changed in order to clearly reveal the statistical sample symmetry introduced by the processing history. The irreducible representatives further allow the construction of symmetric basis functions consistent with the crystallographic symmetry and the statistical sample symmetry; the expansion of an ODF as a linear combination of the symmetric hyperspherical harmonics is often significantly more efficient than the expansion in equation (1).

Results corresponding to these for the generalized spherical harmonics appear in a variety of sources (Bunge, 1993; Gel'fand *et al.*, 1963; Vilenkin, 1968) and follow directly from group-theoretical considerations and knowledge of the relationship of these functions to the irreducible representatives of $SO(3)$, the three-dimensional rotation group. Furthermore, restricted forms of the addition theorem for the hyperspherical harmonics appear in the quantum-mechanics literature (Avery & Wen, 1982; Bander & Itzykson, 1966; Domokos, 1967) and the general form of the irreducible representatives of $SO(4)$ follows from basic considerations of the quantum theory of angular momentum. Nevertheless, the literature generally presents these results without following an explicit (or, in some cases, consistent) set of conventions. To the author's knowledge, the results contained herein do not appear elsewhere in the literature in a form that is consistent with the conventions of this paper, particularly as they apply to the field of texture analysis and to the expansion of the ODF given in equation (1).

2. Conventions

While definitions of the hyperspherical harmonics appear throughout the literature (Bander & Itzykson, 1966; Biedenharn, 1961; Domokos, 1967; Meremianin, 2006), there is no general agreement on the phase. Since changing the phase of the hyperspherical harmonics amounts to a similarity transformation of the irreducible representatives of $SO(4)$ for which these form a basis, some care should be exercised to ensure that the phase of the hyperspherical harmonics is consistent with the explicit formula for the irreducible representatives of $SO(4)$. Otherwise, these representatives do not transform the elements of the basis correctly. The definition and phase convention of the complex hyperspherical harmonics used in this paper is

$$Z^n_{l,m}(\omega,\theta,\varphi) = (-i)^l \frac{2^{l+1/2} l!}{2\pi}\left[(2l+1)\frac{(l-m)!}{(l+m)!}\frac{(n+1)(n-l)!}{(n+l+1)!}\right]^{1/2} \times [\sin(\omega/2)]^l C^{l+1}_{n-l}(\cos(\omega/2)) P^m_l(\cos\theta)\exp(im\varphi), \quad (2)$$

with integer indices $0 \le n$, $0 \le l \le n$ and $-l \le m \le l$, and where C^{l+1}_{n-l} is a Gegenbauer polynomial and P^m_l is an associated Legendre function (Bateman & Erdélyi, 1953; Gradshtein *et al.*, 2000). While different from the convention of Mason & Schuh (2008), this definition is preferable for the purposes of this paper and is consistent with the definitions of the hyperspherical harmonics provided in, *e.g.*, Biedenharn (1961), Hicks & Winternitz (1971), Muljarov *et al.* (2000) and Meremianin (2006).

With respect to the irreducible representatives of the rotation group, this paper follows the same conventions as Altmann (1986). That is, a rotation operation is considered as an active rotation of configuration space, rather than a passive rotation of the coordinate system. An irreducible representative left-multiplies the column vector of the coordinates of a point and right-multiplies the row vector of the components of the basis. With these conventions established, a formula for the matrix element in row m' and column m of the $(2l+1)$-dimensional irreducible representative of $SU(2)$ is found in Appendix A to be

$$U^l_{m',m}(\omega,\theta,\varphi) = \sum_\lambda \sum_\mu \frac{[2(2\lambda+1)]^{1/2}\pi}{2l+1} C^{l,m}_{l,m',\lambda,\mu} Z^{2l}_{\lambda,\mu}(\omega,\theta,\varphi), \quad (3)$$

with integer or half-integer indices $0 \le l$, $-l \le m' \le l$ and $-l \le m \le l$, and where the properties of the Clebsch–Gordan coefficients $C^{l,m}_{l,m',\lambda,\mu}$ constrain the summation indices to the values $0 \le \lambda \le 2l$ and $\mu = m - m'$. Wigner (1959), Vilenkin (1968), Rose (1995) and Varshalovich *et al.* (2008) provide some discussion of the definitions and properties of the Clebsch–Gordan coefficients. The notation for the Clebsch–Gordan coefficients used in this paper follows that of Varshalovich *et al.* (2008). This expansion appears elsewhere in the literature (Varshalovich *et al.*, 1974, 2008), but does not seem to be well known.

3. An addition theorem

The addition theorem derived in this section for the hyperspherical harmonics is analogous to the addition theorems for, *e.g.*, the Gegenbauer polynomials or the associated Legendre functions, and is used to simplify certain summations of products of the hyperspherical harmonics. The formula derives from the observation that if a rotation described by the parameters ω_1, θ_1 and φ_1 is followed by a rotation described by the parameters ω_2, θ_2 and φ_2, then the result is equivalent to that of some single rotation described by the parameters ω, θ and φ. This is equivalent to the matrix multiplication of the corresponding irreducible representatives, or

$$U^l_{m',m}(\omega,\theta,\varphi)=\sum_{m''}U^l_{m',m''}(\omega_2,\theta_2,\varphi_2)U^l_{m'',m}(\omega_1,\theta_1,\varphi_1). \quad (4)$$

Although equation (4) may be expanded with equation (3), the procedure is simplified by initially applying some of the symmetry properties of the Clebsch–Gordan coefficients to equation (3) to obtain

$$U^l_{m',m}(\omega,\theta,\varphi) =\sum_{\lambda}\sum_{\mu}(-1)^{-\lambda+l-m'}\frac{(2)^{1/2}\pi}{(2l+1)^{1/2}}C^{\lambda,\mu}_{l,-m',l,m}Z^{2l}_{\lambda,\mu}(\omega,\theta,\varphi). \quad (5)$$

This is substituted into equation (4) to give

$$\begin{aligned}\sum_{\lambda}\sum_{\mu}&(-1)^{-\lambda+l-m'}\frac{(2)^{1/2}\pi}{(2l+1)^{1/2}}C^{\lambda,\mu}_{l,-m',l,m}Z^{2l}_{\lambda,\mu}\\ &=\sum_{m''}\sum_{\lambda_2}\sum_{\mu_2}(-1)^{-\lambda_2+l-m'}\frac{(2)^{1/2}\pi}{(2l+1)^{1/2}}C^{\lambda_2,\mu_2}_{l,-m',l,m''}Z^{2l}_{\lambda_2,\mu_2}\\ &\quad\times\sum_{\lambda_1}\sum_{\mu_1}(-1)^{-\lambda_1+l-m''}\frac{(2)^{1/2}\pi}{(2l+1)^{1/2}}C^{\lambda_1,\mu_1}_{l,-m'',l,m}Z^{2l}_{\lambda_1,\mu_1}, \end{aligned} \quad (6)$$

where $Z^{2l}_{\lambda_i,\mu_i}$ is written for $Z^{2l}_{\lambda_i,\mu_i}(\omega_i,\theta_i,\varphi_i)$ for the sake of brevity. After multiplying by the Clebsch–Gordan coefficient $C^{\lambda',\mu'}_{l,-m',l,m}$ and summing over the indices $-m'$ and m, this becomes

$$\begin{aligned}\sum_{\lambda}\sum_{\mu}&(-1)^{-\lambda}Z^{2l}_{\lambda,\mu}\left[\sum_{-m'}\sum_{m}C^{\lambda',\mu'}_{l,-m',l,m}C^{\lambda,\mu}_{l,-m',l,m}\right]\\ &=\frac{(2)^{1/2}\pi}{(2l+1)^{1/2}}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{-\lambda_2-\lambda_1}Z^{2l}_{\lambda_2,\mu_2}Z^{2l}_{\lambda_1,\mu_1}\\ &\quad\times\left[\sum_{m''}\sum_{-m'}\sum_{m}(-1)^{l-m''}C^{\lambda',\mu'}_{l,-m',l,m}C^{\lambda_2,\mu_2}_{l,-m',l,m''}C^{\lambda_1,\mu_1}_{l,-m'',l,m}\right]. \end{aligned} \quad (7)$$

The quantity in brackets in the first line is $\delta_{\lambda',\lambda}\delta_{\mu',\mu}$ by the unitarity of the Clebsch–Gordan coefficients, where δ is the Kronecker delta, while the quantity in brackets in the third line is found to be (Varshalovich *et al.*, 2008)

$$\begin{aligned}\sum_{m''}\sum_{-m'}\sum_{m}&(-1)^{l-m''}C^{\lambda,\mu}_{l,-m',l,m}C^{\lambda_2,\mu_2}_{l,-m',l,m''}C^{\lambda_1,\mu_1}_{l,-m'',l,m}\\ &=(-1)^{-\lambda}(2\lambda_2+1)^{1/2}(2\lambda_1+1)^{1/2}C^{\lambda,\mu}_{\lambda_2,\mu_2,\lambda_1,\mu_1}\begin{Bmatrix}\lambda&\lambda_2&\lambda_1\\ l&l&l\end{Bmatrix}. \end{aligned} \quad (8)$$

The quantity in braces is the Wigner $6j$ symbol, and is defined in, *e.g.*, Wigner (1959) and Varshalovich *et al.* (2008). Simplification of the left side of equation (7) and substitution of equation (8) into the right side of equation (7) gives

$$\begin{aligned}Z^{2l}_{\lambda,\mu}&=\frac{(2)^{1/2}\pi}{(2l+1)^{1/2}}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{-\lambda_2-\lambda_1}(2\lambda_2+1)^{1/2}\\ &\quad\times(2\lambda_1+1)^{1/2}Z^{2l}_{\lambda_2,\mu_2}Z^{2l}_{\lambda_1,\mu_1}C^{\lambda,\mu}_{\lambda_2,\mu_2,\lambda_1,\mu_1}\begin{Bmatrix}\lambda&\lambda_2&\lambda_1\\ l&l&l\end{Bmatrix} \end{aligned} \quad (9)$$

as an addition theorem for the hyperspherical harmonics. This is closely related to an addition theorem for the generalized characters of the irreducible representations of the rotation group as reported by Alper (1971) and Varshalovich *et al.* (1974), and is a generalization of the more restricted addition theorems reported by Bander & Itzykson (1966), Domokos (1967) and Avery & Wen (1982).

4. The irreducible representatives of $SO(4)$

The effect of a four-dimensional rotation on the hyperspherical harmonics is defined by the irreducible representatives of $SO(4)$, with bases given by sets of the hyperspherical harmonics. The local isomorphism of $SO(4)$ to the direct product group $SU(2)\otimes SU(2)$ (Racah, 1959; Biedenharn, 1961; Bander & Itzykson, 1966; Hicks & Winternitz, 1971) allows the irreducible representatives of $SO(4)$ to be constructed from the direct product of the irreducible representatives of $SU(2)$ (Roman, 1961; Sharp, 1968). A suitable similarity transformation to the coupled basis by means of the Clebsch–Gordan coefficients then changes the bases of these irreducible representatives to sets of the hyperspherical harmonics. The resulting irreducible representatives of $SO(4)$ determine the effect of a three-dimensional rotation of the sample on the hyperspherical harmonic expansion of an ODF, in view of the considerations in Mason & Schuh (2008).

The purpose of this section is to find an alternative expression for the $(2l+1)^2$-dimensional irreducible representatives of $SO(4)$, the elements $R^{2l}_{\lambda',\mu',\lambda,\mu}$ of which are constructed in Mason & Schuh (2008) as

$$\begin{aligned}R^{2l}_{\lambda',\mu',\lambda,\mu}&(\omega_2,\theta_2,\varphi_2,\omega_1,\theta_1,\varphi_1)\\ &=\sum_{m'''}\sum_{m''}\sum_{m'}\sum_{m}C^{\lambda',\mu'}_{l,m'',l,m'}U^{l*}_{m''',m''}(\omega_2,\theta_2,\varphi_2)\\ &\quad\times U^l_{m',m}(\omega_1,\theta_1,\varphi_1)C^{\lambda,\mu}_{l,m''',l,m}, \end{aligned} \quad (10)$$

with integer or half-integer index $0\le l$, and integer indices $0\le\lambda'\le 2l$, $-\lambda'\le\mu'\le\lambda'$, $0\le\lambda\le 2l$ and $-\lambda\le\mu\le\lambda$. The indices λ' and μ' label the rows of the representative in increasing values of λ' and in decreasing values of μ' for a particular value of λ', while the indices λ and μ label the columns of the representative in increasing values of λ and in decreasing values of μ for a particular value of λ. The summation indices m''', m'', m' and m range from $-l$ to l, subject to the constraints $m''+m'=\mu'$ and $m'''+m=\mu$. The assertion of the author is that the irreducible representative in equation (10) of the current paper correctly transforms the hyperspherical harmonics in equation (2). While an explicit demonstration of this is not the purpose of the current paper, this issue will be addressed in future publications.

The irreducible representatives of $SU(2)$ in equation (10) may be expanded by means of equation (3) to find

$$R^{2l}_{\lambda',\mu',\lambda,\mu} = \sum_{m'''}\sum_{m''}\sum_{m'}\sum_{m} C^{\lambda',\mu'}_{l,m'',l,m'} C^{\lambda,\mu}_{l,m''',l,m} \times \sum_{\lambda_2}\sum_{\mu_2} \frac{[2(2\lambda_2+1)]^{1/2}\pi}{2l+1} C^{l,m''}_{l,m''',\lambda_2,\mu_2} Z^{2l*}_{\lambda_2,\mu_2} \times \sum_{\lambda_1}\sum_{\mu_1} \frac{[2(2\lambda_1+1)]^{1/2}\pi}{2l+1} C^{l,m}_{l,m',\lambda_1,\mu_1} Z^{2l}_{\lambda_1,\mu_1}, \quad (11)$$

where $Z^{2l}_{\lambda_i,\mu_i}$ and $R^{2l}_{\lambda',\mu',\lambda,\mu}$ are written for $Z^{2l}_{\lambda_i,\mu_i}(\omega_i,\theta_i,\varphi_i)$ and $R^{2l}_{\lambda',\mu',\lambda,\mu}(\omega_2,\theta_2,\varphi_2,\omega_1,\theta_1,\varphi_1)$ for the sake of brevity. Since complex conjugation of equation (2) reveals that

$$Z^{n*}_{l,m}(\omega,\theta,\varphi) = (-1)^{l+m} Z^{n}_{l,-m}(\omega,\theta,\varphi), \quad (12)$$

some rearrangement of equation (11) and substitution of equation (12) gives

$$R^{2l}_{\lambda',\mu',\lambda,\mu} = \frac{2\pi^2}{(2l+1)^2}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{\lambda_2+\mu_2}(2\lambda_2+1)^{1/2} \times (2\lambda_1+1)^{1/2} Z^{2l}_{\lambda_2,-\mu_2} Z^{2l}_{\lambda_1,\mu_1} \times \sum_{m'''}\sum_{m''}\sum_{m'}\sum_{m} C^{\lambda',\mu'}_{l,m'',l,m'} C^{\lambda,\mu}_{l,m''',l,m} C^{l,m''}_{l,m''',\lambda_2,\mu_2} C^{l,m}_{l,m',\lambda_1,\mu_1}. \quad (13)$$

The symmetry properties of the Clebsch–Gordan coefficients, along with substitution of the index $-\mu_2$ for μ_2, allow equation (13) to be written as

$$R^{2l}_{\lambda',\mu',\lambda,\mu} = \frac{2\pi^2}{(2l+1)^2}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{\lambda_2-\mu_2+\mu_1} \times (2\lambda_2+1)^{1/2}(2\lambda_1+1)^{1/2} Z^{2l}_{\lambda_2,\mu_2} Z^{2l}_{\lambda_1,\mu_1} \times \left[\sum_{m'''}\sum_{m''}\sum_{m'}\sum_{m} C^{\lambda',\mu'}_{l,m'',l,m'} C^{\lambda,\mu}_{l,m''',l,m} C^{l,m''}_{l,m''',\lambda_2,-\mu_2} C^{l,m'}_{l,m,\lambda_1,-\mu_1}\right]. \quad (14)$$

The quantity in brackets is found to be (Varshalovich *et al.*, 2008)

$$\sum_{m'''}\sum_{m''}\sum_{m'}\sum_{m} C^{\lambda',\mu'}_{l,m'',l,m'} C^{\lambda,\mu}_{l,m''',l,m} C^{l,m''}_{l,m''',\lambda_2,-\mu_2} C^{l,m'}_{l,m,\lambda_1,-\mu_1} = \sum_{k}\sum_{\kappa}(2l+1)(2\lambda+1)^{1/2}(2k+1)^{1/2} \times C^{k,\kappa}_{\lambda_2,-\mu_2,\lambda_1,-\mu_1} C^{\lambda',\mu'}_{\lambda,\mu,k,\kappa} \begin{Bmatrix} \lambda' & l & l \\ \lambda & l & l \\ k & \lambda_2 & \lambda_1 \end{Bmatrix}, \quad (15)$$

where the quantity in braces in equation (15) is the Wigner $9j$ symbol, and is defined in, *e.g.*, Wigner (1959) and Varshalovich *et al.* (2008). With this, the expression for the matrix element of the irreducible representative becomes

$$R^{2l}_{\lambda',\mu',\lambda,\mu} = \frac{2\pi^2(2\lambda+1)^{1/2}}{2l+1}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{\lambda_2-\mu_2+\mu_1} \times (2\lambda_2+1)^{1/2}(2\lambda_1+1)^{1/2} Z^{2l}_{\lambda_2,\mu_2} Z^{2l}_{\lambda_1,\mu_1} \times \sum_{k}\sum_{\kappa}(2k+1)^{1/2} C^{k,\kappa}_{\lambda_2,-\mu_2,\lambda_1,-\mu_1} C^{\lambda',\mu'}_{\lambda,\mu,k,\kappa} \begin{Bmatrix} \lambda' & l & l \\ \lambda & l & l \\ k & \lambda_2 & \lambda_1 \end{Bmatrix}, \quad (16)$$

which is an alternative form for the elements of the irreducible representatives of $SO(4)$ described by equation (10).

5. Implications of the formula for the irreducible representatives of $SO(4)$

The utility of equation (16) is that certain properties of the irreducible representatives of $SO(4)$ follow more readily from this form than from that provided in equation (10). For instance, examine the elements of the row identified by the indices $\lambda'=0$ and $\mu'=0$. Applying this restriction to equation (16) gives

$$R^{2l}_{0,0,\lambda,\mu} = \frac{2\pi^2(2\lambda+1)^{1/2}}{2l+1}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{\lambda_2-\mu_2+\mu_1} \times (2\lambda_2+1)^{1/2}(2\lambda_1+1)^{1/2} Z^{2l}_{\lambda_2,\mu_2} Z^{2l}_{\lambda_1,\mu_1} \times \sum_{k}\sum_{\kappa}(2k+1)^{1/2} C^{k,\kappa}_{\lambda_2,-\mu_2,\lambda_1,-\mu_1} C^{0,0}_{\lambda,\mu,k,\kappa} \begin{Bmatrix} 0 & l & l \\ \lambda & l & l \\ k & \lambda_2 & \lambda_1 \end{Bmatrix}. \quad (17)$$

Since the Clebsch–Gordan coefficient $C^{0,0}_{\lambda,\mu,k,\kappa}$ is equal to $(-1)^{\lambda-\mu}\delta_{\lambda,k}\delta_{\mu,-\kappa}(2\lambda+1)^{-1/2}$, this simplifies to

$$R^{2l}_{0,0,\lambda,\mu} = \frac{2\pi^2(2\lambda+1)^{1/2}}{2l+1}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{\lambda+\lambda_2}(2\lambda_2+1)^{1/2} \times (2\lambda_1+1)^{1/2} Z^{2l}_{\lambda_2,\mu_2} Z^{2l}_{\lambda_1,\mu_1} C^{\lambda,-\mu}_{\lambda_2,-\mu_2,\lambda_1,-\mu_1} \times \begin{Bmatrix} 0 & l & l \\ \lambda & l & l \\ \lambda & \lambda_2 & \lambda_1 \end{Bmatrix}, \quad (18)$$

where the requirement that $\mu = \mu_1 + \mu_2$, as enforced by the remaining Clebsch–Gordan coefficient, cancels some of the factors of (-1). This expression is simplified further by observing that the $9j$ symbol may be reduced to a $6j$ symbol through the relation (Varshalovich *et al.*, 2008)

$$\begin{Bmatrix} 0 & l & l \\ \lambda & l & l \\ \lambda & \lambda_2 & \lambda_1 \end{Bmatrix} = \frac{(-1)^{\lambda+\lambda_2}}{(2l+1)^{1/2}(2\lambda+1)^{1/2}} \begin{Bmatrix} \lambda & \lambda_2 & \lambda_1 \\ l & l & l \end{Bmatrix}. \quad (19)$$

This allows equation (18) to be written as

$$R^{2l}_{0,0,\lambda,\mu} = \frac{2\pi^2}{(2l+1)^{3/2}}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(2\lambda_2+1)^{1/2}(2\lambda_1+1)^{1/2} \times Z^{2l}_{\lambda_2,\mu_2} Z^{2l}_{\lambda_1,\mu_1} C^{\lambda,-\mu}_{\lambda_2,-\mu_2,\lambda_1,-\mu_1} \begin{Bmatrix} \lambda & \lambda_2 & \lambda_1 \\ l & l & l \end{Bmatrix}, \quad (20)$$

or, once more using the symmetry properties of the Clebsch–Gordan coefficients,

$$R^{2l}_{0,0,\lambda,\mu} = (-1)^{-\lambda}\frac{(2)^{1/2}\pi}{2l+1}\left[\frac{(2)^{1/2}\pi}{(2l+1)^{1/2}}\sum_{\lambda_2}\sum_{\mu_2}\sum_{\lambda_1}\sum_{\mu_1}(-1)^{\lambda_2+\lambda_1} \times (2\lambda_2+1)^{1/2}(2\lambda_1+1)^{1/2} Z^{2l}_{\lambda_2,\mu_2} Z^{2l}_{\lambda_1,\mu_1} \times C^{\lambda,\mu}_{\lambda_2,\mu_2,\lambda_1,\mu_1} \begin{Bmatrix} \lambda & \lambda_2 & \lambda_1 \\ l & l & l \end{Bmatrix}\right]. \quad (21)$$

The quantity in brackets, by the addition theorem provided in equation (9), is one of the hyperspherical harmonics. That is, the expression for the elements of the row identified by the indices $\lambda' = 0$ and $\mu' = 0$ of the irreducible representative of $SO(4)$ defined in equation (10) is given by

$$R^{2l}_{0,0,\lambda,\mu} = (-1)^{\lambda}\frac{(2)^{1/2}\pi}{2l+1} Z^{2l}_{\lambda,\mu}, \quad (22)$$

where the arguments ω, θ and φ of the hyperspherical harmonic describe the rotation resulting from following a rotation described by ω_1, θ_1 and φ_1 by a rotation described by ω_2, θ_2 and φ_2. Similar considerations reveal that the elements of the column of the irreducible representative identified by the indices $\lambda = 0$ and $\mu = 0$ may be written as

$$R^{2l}_{\lambda',\mu',0,0} = \frac{(2)^{1/2}\pi}{2l+1} Z^{2l*}_{\lambda',\mu'}, \quad (23)$$

where the arguments ω, θ and φ of the hyperspherical harmonic describe the rotation resulting from following a rotation described by ω_2, θ_2 and φ_2 by a rotation described by ω_1, θ_1 and φ_1. The author emphasizes that these results are obtained much more simply from equation (16) than from equation (10).

6. Discussion and conclusions

One of the benefits of the hyperspherical harmonic expansion of an ODF is that this expansion is given in terms of angular quantities intuitively related to the axis and angle of a rotation, with the practical and mathematical benefits that this brings. Nevertheless, the utility of this expansion is currently restricted by our limited understanding of the properties of the hyperspherical harmonics.

From the definition of the hyperspherical harmonics in equation (2) and the expansion of the matrix elements of the irreducible representatives of $SO(3)$ in equation (3), this paper derives two mathematical results that are expected to considerably increase the usefulness of the hyperspherical harmonic expansion. The first of these, given in equation (9), is an addition theorem for the hyperspherical harmonics. This is closely related to the generalized spherical harmonic addition theorem which enables the convolution of ODFs expressed in the generalized spherical harmonic expansion, and the author expects that the hyperspherical harmonic addition theorem will serve a similar purpose. The second result, given in equation (16), is an analytical form for the irreducible representatives of $SO(4)$ that does not require prior construction of the irreducible representatives of $SU(2)$. This allows certain properties of the irreducible representatives of $SO(4)$ to be more easily observed, and provides a route to a deeper understanding of the matrices required to change the reference orientation for an ODF in the form of equation (1), or to construct a basis of symmetric hyperspherical harmonics that reflects the crystal and sample symmetries, as in Mason & Schuh (2008).

The author hopes that the results provided in this paper will increase the utility of the hyperspherical harmonic expansion presented for the crystallography community, and for the materials science community as a whole.

APPENDIX A
Determination of the functions $U^l_{m',m}(\omega,\theta,\varphi)$

The function $U^l_{m',m}(\omega,\theta,\varphi)$ is the matrix element in row m' and column m of one of the $(2l+1)$-dimensional irreducible representatives of $SU(2)$. An analytical expression for these functions may be found by converting an expression for the matrix elements in terms of the Cayley–Klein parameters a and b into an equivalent expression in terms of the angular quantities ω, θ and φ. The formula for the matrix elements of the irreducible representatives of $SU(2)$ using the Cayley–Klein parameters is (Wigner, 1959; Vilenkin, 1968; Altmann, 1986; Rose, 1995)

$$R^{l}_{m',m}(a,b) = [(l+m')!(l-m')!(l+m)!(l-m)!]^{1/2} \times \sum_{k} \frac{a^{l+m-k}(a^*)^{l-m'-k} b^{m'-m+k}(-b^*)^{k}}{(l+m-k)!(l-m'-k)!(m'-m+k)!k!}, \tag{24}$$

with integer or half-integer indices $0 \le l$, $-l \le m \le l$ and $-l \le m' \le l$. The index m' labels the rows of the representative sequentially from l to $-l$, and m labels the columns sequentially from l to $-l$. The index k ranges over all values for which the factorials are finite. While the meaning and use of this representative varies subtly among the cited references, the interpretation of the author follows that of Altmann (1986).

The conversion from the Cayley–Klein parameters to the angles ω, θ and φ is accomplished by comparing the matrix elements of the two-dimensional complex representatives of $SU(2)$ in the respective parameterizations. For the Cayley–Klein parameters, the constraints of unitarity and unit determinant require the representative to be of the form

$$R^{1/2}(a,b) = \begin{pmatrix} a & b \\ -b^* & a^* \end{pmatrix}, \tag{25}$$

where a and b satisfy the condition $|a|^2 + |b|^2 = 1$. The Cayley–Klein parameters may be written as linear combinations of the components of a normalized quaternion (Altmann, 1986), and through them expressed as functions of the rotation angles ω, θ and φ by means of a polar parameterization (Mason & Schuh, 2008). With these relations, the irreducible representative in equation (25) becomes

$$U^{1/2}(\omega,\theta,\varphi) = \begin{pmatrix} \cos(\omega/2) - i\sin(\omega/2)\cos(\theta) & -i\sin(\omega/2)\sin(\theta)\exp(-i\varphi) \\ -i\sin(\omega/2)\sin(\theta)\exp(i\varphi) & \cos(\omega/2) + i\sin(\omega/2)\cos(\theta) \end{pmatrix}. \tag{26}$$

Comparison of the matrix elements of equations (25) and (26) then allows the Cayley–Klein parameters to be written as functions of ω, θ and φ.

The unitarity of $R^{1/2}(a,b)$ in equation (25) indicates, by inspection, that the inverse of $R^{1/2}(a,b)$ is $R^{1/2}(a^*,-b)$. Insertion of the quantities a^* and $-b$ into equation (24) in the place of a and b provides

$$R^{l}_{m',m}(a^*,-b) = (-1)^{m-m'} R^{l}_{-m,-m'}(a,b) \tag{27}$$

for the matrix elements of the inverse of $R^l(a,b)$. An analogous expression holds for the matrix elements of the inverse of $U^l(\omega,\theta,\varphi)$ as well. Although other symmetries exist, this one shall be of particular use in the following.

While an expression for the functions $U^{l}_{m',m}(\omega,\theta,\varphi)$ may be found by direct substitution of the matrix elements of $U^{1/2}(\omega,\theta,\varphi)$ from equation (26) into equation (24), a more elegant expression may be found by considering the operation described by the representative $U^l(\omega,\theta,\varphi)$ to be the result of three distinct rotations (Varshalovich *et al.*, 1974). The first rotation, described by the representative $U^l(\theta,\pi/2,\varphi-\pi/2)$, brings the point on the sphere described by the spherical angles θ and φ to the z axis. The second rotation, described by the representative $U^l(\omega,0,0)$, performs a rotation by the angle ω about the z axis. The third rotation, described by the representative $U^l(\theta,\pi/2,\varphi+\pi/2)$, returns the point on the z axis to the point on the sphere described by the spherical angles θ and φ, and is the inverse of the first rotation. The representative $U^l(\omega,\theta,\varphi)$ is then constructed as the product of three representatives by

$$U^{l}_{m',m}(\omega,\theta,\varphi) = \sum_{m'''}\sum_{m''} U^{l}_{m',m'''}(\theta,\pi/2,\varphi+\pi/2) \times U^{l}_{m''',m''}(\omega,0,0) U^{l}_{m'',m}(\theta,\pi/2,\varphi-\pi/2), \tag{28}$$

with integer or half-integer indices $-l \le m''' \le l$ and $-l \le m'' \le l$. Observing that the third representative is the inverse of the first, a symmetry relation equivalent to equation (27) is used to find

$$U^{l}_{m',m}(\omega,\theta,\varphi) = \sum_{m'''}\sum_{m''}(-1)^{m'''-m'} U^{l}_{m''',m''}(\omega,0,0) U^{l}_{-m''',-m'}(\theta,\pi/2,\varphi-\pi/2) \times U^{l}_{m'',m}(\theta,\pi/2,\varphi-\pi/2). \tag{29}$$

Since two of the representatives in equation (29) share the same arguments, the product of these representatives may be expanded in a series using the Clebsch–Gordan coefficients (Wigner, 1959; Vilenkin, 1968; Rose, 1995; Varshalovich *et al.*, 2008). This gives

$$U^{l}_{m',m}(\omega,\theta,\varphi) = \sum_{m'''}\sum_{m''}(-1)^{m'''-m'} U^{l}_{m''',m''}(\omega,0,0) \times \sum_{\lambda}\sum_{\nu,\mu} C^{\lambda,\nu}_{l,-m''',l,m''} U^{\lambda}_{\nu,\mu}(\theta,\pi/2,\varphi-\pi/2) C^{\lambda,\mu}_{l,-m',l,m}, \tag{30}$$

with integer indices $0 \le \lambda \le 2l$, $-\lambda \le \nu \le \lambda$ and $-\lambda \le \mu \le \lambda$, and where $\nu = -m''' + m''$ and $\mu = -m' + m$.

The quantity $U^{l}_{m''',m''}(\omega,0,0)$ is evaluated by comparing equation (26) with equation (25), and observing that $a = \exp(-i\omega/2)$ and $b = 0$. Insertion of these quantities into equation (24) reveals that the individual terms of the summation therein vanish for $k \ne 0$, and that $U^{l}_{m''',m''}(\omega,0,0)$ vanishes for $m''' \ne m''$. Simplification of the remaining quantity provides the expected result, that

$$U^{l}_{m''',m''}(\omega,0,0) = \delta_{m''',m''}\exp(-im''\omega). \tag{31}$$

With this, the expression for $U^{l}_{m',m}(\omega,\theta,\varphi)$ becomes

$$U^{l}_{m',m}(\omega,\theta,\varphi) = \sum_{m''}(-1)^{m''-m'}\exp(-im''\omega) \times \sum_{\lambda}\sum_{\mu} C^{\lambda,0}_{l,-m'',l,m''} U^{\lambda}_{0,\mu}(\theta,\pi/2,\varphi-\pi/2) C^{\lambda,\mu}_{l,-m',l,m}. \tag{32}$$

The quantity $U^{\lambda}_{0,\mu}(\theta,\pi/2,\varphi-\pi/2)$ is evaluated similarly. Comparison of equation (26) with equation (25) indicates that, for this representative, $a = \cos(\theta/2)$ and $b = \sin(\theta/2)\exp(-i\varphi)$. Substitution into equation (24) yields

$$U^{\lambda}_{0,\mu}(\theta, \pi/2, \varphi - \pi/2) = (-1)^{\mu}\left[\frac{(\lambda+\mu)!}{(\lambda-\mu)!}\right]^{1/2}\left[(-1)^{\mu}\lambda!(\lambda-\mu)!\left(\frac{1+\cos\theta}{2}\right)^{\lambda} \times \left(\frac{1-\cos\theta}{1+\cos\theta}\right)^{-\mu/2}\sum_{k}\frac{(-1)^{k}}{(\lambda+\mu-k)!(\lambda-k)!(-\mu+k)!k!} \times \left(\frac{1-\cos\theta}{1+\cos\theta}\right)^{k}\right]\exp(i\mu\varphi) \quad (33)$$

after some rearrangement and collection of terms. The trigonometric functions of half-angles in this equation have been converted to functions of full angles. Recognizing the quantity in brackets as the associated Legendre function $P^{-\mu}_{\lambda}(\cos\theta)$ (Vilenkin, 1968), the function $U^{\lambda}_{0,\mu}(\theta, \pi/2, \varphi - \pi/2)$ is instead written as

$$U^{\lambda}_{0,\mu}(\theta, \pi/2, \varphi - \pi/2) = \left[\frac{(\lambda-\mu)!}{(\lambda+\mu)!}\right]^{1/2} P^{\mu}_{\lambda}(\cos\theta)\exp(i\mu\varphi). \quad (34)$$

The expression for $U^{l}_{m',m}(\omega, \theta, \varphi)$ then becomes

$$U^{l}_{m',m}(\omega, \theta, \varphi) = \sum_{\lambda}\sum_{\mu}(-1)^{-m'}C^{\lambda,\mu}_{l,-m',l,m}\left[\frac{(\lambda-\mu)!}{(\lambda+\mu)!}\right]^{1/2} \times P^{\mu}_{\lambda}(\cos\theta)\exp(i\mu\varphi)\sum_{m''}(-1)^{m''}C^{\lambda,0}_{l,-m'',l,m''}\exp(-im''\omega). \quad (35)$$

The symmetry properties of the Clebsch–Gordan coefficients (Vilenkin, 1968; Rose, 1995; Varshalovich *et al.*, 2008) allow the factors of $(-1)^{-m'}$ and $(-1)^{m''}$ to be cancelled by rearranging the indices, giving

$$U^{l}_{m',m}(\omega, \theta, \varphi) = \sum_{\lambda}\sum_{\mu}(-i)^{\lambda}C^{l,m}_{l,m',\lambda,\mu}\,\frac{2\lambda+1}{2l+1}\left[\frac{(\lambda-\mu)!}{(\lambda+\mu)!}\right]^{1/2} \times P^{\mu}_{\lambda}(\cos\theta)\exp(i\mu\varphi)\left[i^{\lambda}\sum_{m''}C^{l,m''}_{l,m'',\lambda,0}\exp(-im''\omega)\right]. \quad (36)$$

The quantity in brackets is referred to as the generalized character of the irreducible representations of the rotation group and is written as $\chi^{l}_{\lambda}(\omega)$. An alternate expression for this function is (Varshalovich *et al.*, 2008)

$$\chi^{l}_{\lambda}(\omega) = 2^{\lambda}\lambda!\left[\frac{(2l+1)(2l-\lambda)!}{(2l+\lambda+1)!}\right]^{1/2}[\sin(\omega/2)]^{\lambda}C^{\lambda+1}_{2l-\lambda}(\cos(\omega/2)), \quad (37)$$

where $C^{\lambda+1}_{2l-\lambda}(\cos(\omega/2))$ is a Gegenbauer polynomial (Bateman & Erdélyi, 1953; Gradshtein *et al.*, 2000). Substitution of equation (37) into equation (36) and some rearrangement yields

$$U^{l}_{m',m}(\omega, \theta, \varphi) = \sum_{\lambda}\sum_{\mu}\frac{[2(2\lambda+1)]^{1/2}\pi}{2l+1}C^{l,m}_{l,m',\lambda,\mu}\Bigg\{(-i)^{\lambda}\frac{2^{\lambda+1/2}\lambda!}{2\pi} \times \left[(2\lambda+1)\frac{(\lambda-\mu)!}{(\lambda+\mu)!}\frac{(2l+1)(2l-\lambda)!}{(2l+\lambda+1)!}\right]^{1/2} \times [\sin(\omega/2)]^{\lambda}C^{\lambda+1}_{2l-\lambda}(\cos(\omega/2))P^{\mu}_{\lambda}(\cos\theta)\exp(i\mu\varphi)\Bigg\}. \quad (38)$$

The quantity in braces is the hyperspherical harmonic $Z^{2l}_{\lambda,\mu}(\omega, \theta, \varphi)$, as defined in equation (2) of the current paper. This allows a compact expression for the matrix elements in the form

$$U^{l}_{m',m}(\omega, \theta, \varphi) = \sum_{\lambda}\sum_{\mu}\frac{[2(2\lambda+1)]^{1/2}\pi}{2l+1}C^{l,m}_{l,m',\lambda,\mu}Z^{2l}_{\lambda,\mu}(\omega, \theta, \varphi). \quad (39)$$

This result is identical to the one obtained for $U^{l}_{m',m}(\omega, \theta, \varphi)$ in Varshalovich *et al.* (1974, 2008). This derivation is provided in the current paper to verify that equation (39) is consistent with the author's current conventions.

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