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Some properties of the coupling coefficients of real spherical harmonics and their relation to Gaunt coefficients[1]

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Abstract

Spherical harmonics are of considerable importance for computations involving basis functions corresponding to large values of the angular momentum quantum number ℓ. Their use allows efficient coding of programs involving such basis functions because the formulae of the coupling coefficients are simple. The choice of real spherical harmonics allows one to avoid the use of complex quantities in computer programs that increase storage and CPU time requirements. In this paper, certain properties of the coupling coefficients for real spherical harmonics are derived that are necessary for an efficient computation of coupling terms.

Keywords: Ab initio program; Basis function; Coupling coefficient; Gaunt coefficient; Selection rule; Symmetry relation

1. Introduction

Basis sets including functions corresponding to larger values of the angular momentum quantum number ℓ are necessary in linear combination of atomic orbital (LCAO) calculations in order to approach the self-consistent field (SCF) basis set limit, and to increase the accuracy of the correlation energy approximation. Such basis functions are also important for scattering calculations and for the description of Rydberg states. For Cartesian-type functions, the angular momentum coupling of such basis functions becomes more difficult the higher ℓ is. Thus, the coding of ab initio programs for these functions becomes rather complex in the case of high angular momentum quantum numbers. On the contrary, the coupling of spherical harmonics is governed by simple formulae even for high values of ℓ. Thus, it is advantageous to use basis sets based on spherical harmonics if high ℓ values are necessary. Coding of programs [1–3] is much simpler in this way.

Complex spherical harmonics are often used. However, the use of complex quantities in computer programs increases storage and CPU time requirements. In this respect, it is advantageous to use a unitary transformation to real spherical harmonics. Although this idea is a rather old one, it seems that there is no description to date of the properties of the coupling coefficients for real spherical harmonics in the literature. The present contribution aims at closing this gap.

In the following section, various quantities are defined and then some general properties of real

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spherical harmonics are listed. In Section 3, their coupling coefficients are studied. Symmetry relations and selection rules will be given and the relation to the coupling coefficients of the complex spherical harmonics, i.e. to Gaunt coefficients and their generalization, will be established.

2. General properties of real spherical harmonics

The usual complex spherical harmonics (more precisely, complex spherical surface harmonics) (see [4], p. 3, Eq. (1.2-1))

$$Y_\ell^m(\theta,\phi) = i^{m+|m|}\left[\frac{2\ell+1}{4\pi}\frac{(\ell-|m|)!}{(\ell+|m|)!}\right]^{1/2} \times P_\ell^{|m|}(\cos\theta)\exp(im\phi) \qquad (1)$$

that are defined in terms of associated Legendre functions P_ℓ^m, obey the relation

$$[Y_\ell^m(\theta,\phi)]^* = (-1)^m Y_\ell^{-m}(\theta,\phi) \qquad (2)$$

The definition (eqn (1)) is consistent with the phase convention of Condon and Shortley ([5], p. 487, Eq. (3)). One can define real spherical harmonics as

$$X_\ell^\mu(\theta,\phi) = \sum_m U_{\ell m}^\mu Y_\ell^m(\theta,\phi) \qquad (3)$$

for $\mu \in \{-\ell, -\ell+1, \ldots, \ell-1, \ell\}$. Here, $U_\ell = \{U_{\ell m}^\mu\}$ denotes for fixed ℓ a $(2\ell+1) \times (2\ell+1)$ unitary matrix. Thus, we have

$$\sum_m [U_{\ell m}^\mu]^* U_{\ell m}^{\mu'} = \delta_{\mu\mu'}, \qquad \sum_\mu [U_{\ell m}^\mu]^* U_{\ell m'}^\mu = \delta_{mm'} \qquad (4)$$

and hence

$$Y_\ell^m(\theta,\phi) = \sum_\mu [U_{\ell m}^\mu]^* X_\ell^\mu(\theta,\phi) \qquad (5)$$

In the following, we also use the notation $\Omega = (\theta,\phi)$. It follows from Eq. (3) that the parity of $X_\ell^\mu(\Omega)$ is $(-1)^\ell$, i.e. identical to that of the corresponding complex spherical harmonics with the same ℓ. In order to obtain real functions, i.e. to ensure

$$[X_\ell^\mu(\Omega)]^* = X_\ell^\mu(\Omega) \qquad (6)$$

the equation

$$\sum_m U_{\ell m}^\mu Y_\ell^m(\Omega) = \sum_m [U_{\ell m}^\mu]^*[Y_\ell^m(\Omega)]^* \qquad (7)$$

or equivalently

$$\sum_m \{(-1)^m [U_{\ell -m}^\mu]^* - U_{\ell m}^\mu\} Y_\ell^m(\Omega) = 0 \qquad (8)$$

has to hold. Since the complex spherical harmonics are linearly independent, the elements of the unitary matrix U_ℓ have to obey

$$(-1)^m [U_{\ell -m}^\mu]^* = U_{\ell m}^\mu \qquad (9)$$

or equivalently

$$[U_{\ell m}^\mu]^* = (-1)^m U_{\ell -m}^\mu \qquad (10)$$

We chose as real spherical harmonics the functions

$$X_\ell^\mu(\Omega) = \begin{cases} \sqrt{2}\,\Re(Y_\ell^{|\mu|}(\Omega)) & \text{for } \mu > 0 \\ Y_\ell^0(\Omega) & \text{for } \mu = 0 \\ \sqrt{2}\,\Im(Y_\ell^{|\mu|}(\Omega)) & \text{for } \mu < 0 \end{cases} \qquad (11)$$

where $\Re(.)$ denotes the real part and $\Im(.)$ the imaginary part. The corresponding unitary matrix U_ℓ has elements

$$U_{\ell m}^\mu = \delta_{m0}\delta_{\mu 0} + \frac{1}{\sqrt{2}}(\Theta(\mu)\delta_{m\mu} + \Theta(-\mu)(+i)(-1)^m\delta_{m\mu} + \Theta(-\mu)(-i)\delta_{m-\mu} + \Theta(\mu)(-1)^m\delta_{m-\mu}) \qquad (12)$$

where δ_{mn} is the usual Kronecker symbol, and

$$\Theta(m) = \begin{cases} 1 \text{ for } m > 0 \\ 0 \text{ for } m \le 0 \end{cases} \qquad (13)$$

Thus

$$U_{\ell m}^\mu = 0 \text{ for } |\mu| \neq |m| \qquad (14)$$

Dropping the arguments θ and ϕ for the moment, we may thus write

$$\begin{pmatrix} X_\ell^\ell \\ \vdots \\ X_\ell^1 \\ X_\ell^0 \\ X_\ell^{-1} \\ \vdots \\ X_\ell^{-\ell} \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & & & & & & (-1)^\ell \\ & \ddots & & & & \cdot^{\cdot^{\cdot}} & \\ & & 1 & & -1 & & \\ & & & \sqrt{2} & & & \\ & & -i & & -i & & \\ & \cdot^{\cdot^{\cdot}} & & & & \ddots & \\ -i & & & & & & (-1)^\ell i \end{pmatrix} \begin{pmatrix} Y_\ell^\ell \\ \vdots \\ Y_\ell^1 \\ Y_\ell^0 \\ Y_\ell^{-1} \\ \vdots \\ Y_\ell^{-\ell} \end{pmatrix} \tag{15}$$

Note that $U_{\ell m}^\mu$ is real for $\mu \geq 0$, and purely imaginary for $\mu < 0$. All other possible choices of real spherical harmonics can be obtained from our choice by applying a suitable real orthogonal transformation.

As a direct consequence of the unitarity of U_ℓ, we note the identity

$$\sum_\mu X_\ell^\mu(\Omega_1) X_\ell^\mu(\Omega_2) = \sum_m Y_\ell^m(\Omega_1)[Y_\ell^m(\Omega_2)]^* \tag{16}$$

As a first application of this formula, we note that it allows one to obtain the following form of the Laplace expansion of the Coulomb potential:

$$\frac{1}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|} = \sum_{\ell\mu} \frac{4\pi}{2\ell+1} \frac{r_<^\ell}{r_>^{\ell+1}} X_\ell^\mu(\Omega_1) X_\ell^\mu(\Omega_2) \tag{17}$$

with

$$r_< = \min(|\boldsymbol{r}_1|, |\boldsymbol{r}_2|) \quad r_> = \max(|\boldsymbol{r}_1|, |\boldsymbol{r}_2|) \tag{18}$$

A more important consequence of eqn (16) is the equation (compare [6], p. 91)

$$\sum_{\ell\mu} X_\ell^\mu(\Omega_1) X_\ell^\mu(\Omega_2) = \sum_{\ell m} Y_\ell^m(\Omega_1)[Y_\ell^m(\Omega_2)]^* = \delta(\Omega_1 - \Omega_2) \tag{19}$$

with

$$\delta(\Omega_1 - \Omega_2) = (\sin\theta_1)^{-1} \delta(\theta_1 - \theta_2) \delta(\phi_1 - \phi_2) \tag{20}$$

Since complex spherical harmonics form a complete orthonormal system (CONS) we have ($d\Omega = \sin\theta \, d\theta \, d\phi$)

$$\int X_\ell^\mu(\Omega) X_{\ell'}^{\mu'}(\Omega) d\Omega = \sum_{mm'} [U_{\ell m}^\mu]^* U_{\ell' m'}^{\mu'} \times \int [Y_\ell^m(\Omega)]^* Y_{\ell'}^{m'}(\Omega) d\Omega = \delta_{\ell\ell'} \sum_m [U_{\ell m}^\mu]^* U_{\ell m}^{\mu'} = \delta_{\ell\ell'} \delta_{\mu\mu'} \tag{21}$$

because, again, the matrices U_ℓ are unitary. In addition, completeness of the real spherical harmonics is a consequence of Eqs. (3) and (5). Thus, the real spherical harmonics also form a CONS.

Finally, we note that one can express the real spherical harmonics in terms of sine and cosine functions of ϕ according to ($\mu > 0$)

$$X_\ell^\mu(\theta, \phi) = (-1)^\mu \left[\frac{2\ell+1}{2\pi} \frac{(\ell-\mu)!}{(\ell+\mu)!} \right]^{1/2} P_\ell^\mu(\cos\theta) \cos\mu\phi$$

$$X_\ell^{-\mu}(\theta, \phi) = (-1)^\mu \left[\frac{2\ell+1}{2\pi} \frac{(\ell-\mu)!}{(\ell+\mu)!} \right]^{1/2} P_\ell^\mu(\cos\theta) \sin\mu\phi \tag{22}$$

3. Coupling coefficients of real spherical harmonics

In this section, we study coupling coefficients of real spherical harmonics, i.e. we want to express products of real spherical harmonics of the same argument by linear combinations of such functions. This can be done in close analogy to the treatment of properties of coupling coefficients of complex spherical harmonics given in Appendix A. These properties will be used in the following.

We saw in the last section that real spherical harmonics form a CONS, as do their complex counterparts. Thus, the product of two real spherical harmonics of the same argument can be represented as a linear combination of real spherical harmonics according to

$$X_{\ell_1}^{\mu_1}(\Omega)X_{\ell_2}^{\mu_2}(\Omega) = \sum_{\ell_3\mu_3} \langle \ell_3\mu_3|\ell_1\mu_1|\ell_2\mu_2\rangle_{\mathrm{R}} X_{\ell_3}^{\mu_3}(\Omega) \tag{23}$$

Similar to the complex case, the orthonormality of the real spherical harmonics allows one to compute the coefficients in this sum according to

$$\langle \ell_3\mu_3|\ell_1\mu_1|\ell_2\mu_2\rangle_{\mathrm{R}} = \int X_{\ell_3}^{\mu_3}(\Omega)X_{\ell_1}^{\mu_1}(\Omega)X_{\ell_2}^{\mu_2}(\Omega)\mathrm{d}\Omega \tag{24}$$

We call them "coupling coefficients of real spherical harmonics" or "R-Gaunt coefficients". Their notation differs from that of the Gaunt coefficients defined in Eq. (A2) in Appendix A by the subscript "R".

In the remainder of this section, we will study the properties of the R-Gaunt coefficients. We are interested in their symmetries and selection rules, and how these coefficients can be computed. Most of these questions can be answered by deriving explicit and compact formulae that express R-Gaunt coefficients in terms of Gaunt coefficients and the unitary matrices U_ℓ.

From Eq. (24), one notes immediately that R-Gaunt coefficients are invariant under all permutations of the pairs (ℓ_1,μ_1), (ℓ_2,μ_2) and (ℓ_3,μ_3). For instance, the following symmetry relation holds.

$$\langle \ell_3\mu_3|\ell_1\mu_1|\ell_2\mu_2\rangle_{\mathrm{R}} = \langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_{\mathrm{R}} \tag{25}$$

Since essentially only the ϕ-dependent parts of real and complex harmonics are different, it must be possible to relate R-Gaunt to Gaunt coefficients. In principle, this can be done by using Eqs. (1) and (22) in the defining integrals, and comparing the resulting expressions. However, a large number of different cases—according to the various sign patterns of the μ_i or their disappearance—then have to be distinguished and treated separately.

Less cases are required in the following approach based on the unitary matrices U_ℓ, introduced in the previous section.

Using Eq. (3) twice and Eq. (7) once in Eq. (24) in combination with Eq. (A2) in Appendix A, one obtains

$$\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_{\mathrm{R}} = \sum_{m_1m_2m_3} [U_{\ell_1m_1}^{\mu_1}]^* U_{\ell_2m_2}^{\mu_2} U_{\ell_3m_3}^{\mu_3} \times \langle \ell_1m_1|\ell_2m_2|\ell_3m_3\rangle \tag{26}$$

Use of the selection rule Eq. (A3) in Appendix A allows this to be simplified to

$$\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_{\mathrm{R}} = \sum_{m_2m_3} [U_{\ell_1m_2+m_3}^{\mu_1}]^* U_{\ell_2m_2}^{\mu_2} U_{\ell_3m_3}^{\mu_3} \times \langle \ell_1m_2+m_3|\ell_2m_2|\ell_3m_3\rangle \tag{27}$$

This formula will be the basis for the following considerations. We study three cases corresponding to various values of the μ_i $(i = 1,2,3)$:

Case A	No μ_i vanishes.
Case B	Exactly one μ_i vanishes. Then, using the symmetry relation Eq. (25), it suffices to study the case that $\mu_3 = 0$, $\mu_1 \neq 0$, $\mu_2 \neq 0$.
Case C	Two or more μ_i vanish. Here, it suffices to consider $\mu_2 = \mu_3 = 0$.

In Cases A and B, $\mu_2 \neq 0$. Then, according to Eq. (12), only the two terms with $m_2 = \pm\mu_2$ will contribute to the sum over m_2 in Eq. (27). One obtains

$$\begin{aligned}\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_{\mathrm{R}} = {} & \sum_{m_3} [U_{\ell_1\mu_2+m_3}^{\mu_1}]^* U_{\ell_2\mu_2}^{\mu_2} U_{\ell_3m_3}^{\mu_3} \\ & \times \langle \ell_1\mu_2+m_3|\ell_2\mu_2|\ell_3m_3\rangle \\ & + \sum_{m_3} [U_{\ell_1m_3-\mu_2}^{\mu_1}]^* U_{\ell_2-\mu_2}^{\mu_2} U_{\ell_3m_3}^{\mu_3} \\ & \times \langle \ell_1m_3-\mu_2|\ell_2-\mu_2|\ell_3m_3\rangle \end{aligned} \tag{28}$$

In Case A, $\mu_3 \neq 0$. Then, according to Eq. (12), only the two terms with $m_3 = \pm\mu_3$ will contribute to the sums over m_3 in Eq. (28). One obtains

$$\begin{aligned}\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_{\mathrm{R}} = {} & [U_{\ell_1\mu_2+\mu_3}^{\mu_1}]^* U_{\ell_2\mu_2}^{\mu_2} U_{\ell_3\mu_3}^{\mu_3} \\ & \times \langle \ell_1\mu_2+\mu_3|\ell_2\mu_2|\ell_3\mu_3\rangle \\ & + [U_{\ell_1\mu_2-\mu_3}^{\mu_1}]^* U_{\ell_2\mu_2}^{\mu_2} U_{\ell_3-\mu_3}^{\mu_3} \\ & \times \langle \ell_1\mu_2-\mu_3|\ell_2\mu_2|\ell_3-\mu_3\rangle \\ & + [U_{\ell_1\mu_3-\mu_2}^{\mu_1}]^* U_{\ell_2-\mu_2}^{\mu_2} U_{\ell_3\mu_3}^{\mu_3} \\ & \times \langle \ell_1-\mu_2+\mu_3|\ell_2-\mu_2|\ell_3\mu_3\rangle \\ & + [U_{\ell_1-\mu_2-\mu_3}^{\mu_1}]^* U_{\ell_2-\mu_2}^{\mu_2} U_{\ell_3-\mu_3}^{\mu_3} \\ & \times \langle \ell_1-\mu_2-\mu_3|\ell_2-\mu_2|\ell_3-\mu_3\rangle \end{aligned} \tag{29}$$

Since Gaunt coefficients are unchanged if the signs of all magnetic quantum numbers are reversed simultaneously according to Eq. (A9) in Appendix A, the last equation can be simplified to

$$\begin{aligned}\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_R = &\langle \ell_1\mu_2+\mu_3|\ell_2\mu_2|\ell_3\mu_3\rangle \\ &\times \{[U^{\mu_1}_{\ell_1\mu_2+\mu_3}]^* U^{\mu_2}_{\ell_2\mu_2} U^{\mu_3}_{\ell_3\mu_3} \\ &+ [U^{\mu_1}_{\ell_1-\mu_2-\mu_3}]^* U^{\mu_2}_{\ell_2-\mu_2} U^{\mu_3}_{\ell_3-\mu_3}\} \\ &\times \langle \ell_1\mu_2-\mu_3|\ell_2\mu_2|\ell_3-\mu_3\rangle \\ &\times \{[U^{\mu_1}_{\ell_1\mu_2-\mu_3}]^* U^{\mu_2}_{\ell_2\mu_2} U^{\mu_3}_{\ell_3-\mu_3} \\ &+ [U^{\mu_1}_{\ell_1\mu_3-\mu_2}]^* U^{\mu_2}_{\ell_2-\mu_2} U^{\mu_3}_{\ell_3\mu_3}\} \quad (30)\end{aligned}$$

The terms in braces are identified using Eq. (10) as real parts according to

$$\begin{aligned}\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_R = &2\langle \ell_1\mu_2+\mu_3|\ell_2\mu_2|\ell_3\mu_3\rangle \Re \\ &\times \left\{[U^{\mu_1}_{\ell_1\mu_2+\mu_3}]^* U^{\mu_2}_{\ell_2\mu_2} U^{\mu_3}_{\ell_3\mu_3}\right\} \\ &+ 2\langle \ell_1\mu_2-\mu_3|\ell_2\mu_2|\ell_3-\mu 3\rangle \Re \\ &\times \left\{[U^{\mu_1}_{\ell_1\mu_2-\mu_3}]^* U^{\mu_2}_{\ell_2\mu_2} U^{\mu_3}_{\ell_3-\mu_3}\right\} \quad (31)\end{aligned}$$

This is the final result for Case A.

In Case B, $\mu_3 = 0$. Then, according to Eq. (12), only the terms with $m_3 = 0$ will contribute to the sums over m_3 in Eq. (28). Proceeding similarly as before, one obtains

$$\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3 0\rangle_R = 2\langle \ell_1\mu_2|\ell_2\mu_2|\ell_3 0\rangle \Re\left\{[U^{\mu_1}_{\ell_1\mu_2}]^* U^{\mu_2}_{\ell_2\mu_2}\right\} \quad (32)$$

as the final result in Case B.

In Case C, the final result is

$$\langle \ell_1\mu_1|\ell_2 0|\ell_3 0\rangle_R = \delta_{\mu_1 0}\langle \ell_1 0|\ell_2 0|\ell_3 0\rangle \quad (33)$$

Since programs for the computation of Gaunt coefficients are available [1], the computation of R-Gaunt coefficients can be based on Eqs. (31)–(33).

Selection rules of the R-Gaunt coefficients are direct consequences of Eqs. (31)–(33).

Only if, for given μ_2 and μ_3, the value of μ_1 satisfies

$$\mu_1 \in \{\mu_2+\mu_3, \mu_2-\mu_3, -\mu_2+\mu_3, -\mu_2-\mu_3\} \quad (34)$$

can the R-Gaunt coefficient $\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_R$ be nonzero. In Cases A and B, this follows by applying Eq. (14) to the first in the product of matrix elements of the unitary matrices in Eq. (31) and Eq. (32), respectively.

In any case, the R-Gaunt coefficient under consideration is directly proportional to a single Gaunt coefficient (or zero). In Case A, this is a consequence of the fact that only one term in Eq. (31) can be nonzero for a given combination of quantum numbers: either the condition $\mu_1 = \pm(\mu_2+\mu_3)$ or $\mu_1 = \pm(\mu_2-\mu_3)$ holds as required in Eq. (34).

We conclude that the selection rules of this single Gaunt coefficient have to be obeyed. In particular, the R-Gaunt coefficient $\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_R$ is zero unless

$$\ell_1+\ell_2+\ell_3 = 2n \quad n: \text{ integer} \quad (35)$$

holds. This, together with the general coupling rules for angular momenta and the selection rules Eqs. (A5), (A6) and (A7), implies that the R-Gaunt coefficient $\langle \ell_1\mu_1|\ell_2\mu_2|\ell_3\mu_3\rangle_R$ is zero unless

$$\ell_1 \in \{\ell_{\max}, \ell_{\max}-2, \ldots, \ell_{\min}\} \quad (36)$$

where

$$\ell_{\max} = \ell_2+\ell_3 \quad (37)$$

and

$$\ell_{\min} = \begin{cases} \kappa(\ell_2,\ell_3,\mu_2,\mu_3) & \text{if } \kappa(\ell_2,\ell_3,\mu_2,\mu_3)+\ell_{\max} \text{ is even} \\ \kappa(\ell_2,\ell_3,\mu_2,\mu_3)+1 & \text{if } \kappa(\ell_2,\ell_3,\mu_2,\mu_3)+\ell_{\max} \text{ is odd}\end{cases} \quad (38)$$

$$\kappa(\ell_2,\ell_3,\mu_2,\mu_3) = \max(|\ell_2-\ell_3|, \min(|\mu_2+\mu_3|, |\mu_2-\mu_3|))$$

A further general selection rule is that all R-Gaunt coefficients with an odd number of negative μ_i are necessarily zero. This can be read off from Eqs. (31)–(33) since $U^{\mu}_{\ell m}$ with $\mu < 0$ are purely imaginary or zero (compare Eq. (12)).

Generalized R-Gaunt coefficients can be defined as

$$\langle \ell_0\mu_0|\ell_1\mu_1\ldots\ell_{k-1}\mu_{k-1}|\ell_k\mu_k\rangle_R = \int X^{\mu_0}_{\ell_0}(\Omega) X^{\mu_1}_{\ell_1}(\Omega)\ldots X^{\mu_k}_{\ell_k}(\Omega)\,d\Omega \quad (39)$$

They are invariant under all permutations of the pairs (ℓ_i,μ_i). As in the case of generalized Gaunt coefficients, they can be reduced to sums over products of R-Gaunt coefficients using Eq. (23) repeatedly. For instance, we have

$$\langle \ell_0\mu_0|\ell_1\mu_1\ell_2\mu_2|\ell_3\mu_3\rangle_R = \sum_{\ell'\mu'} \langle \ell'\mu'|\ell_1\mu_1|\ell_2\mu_2\rangle_R \langle \ell_0\mu_0|\ell'\mu'|\ell_3\mu_3\rangle_R \quad (40)$$

Note that the sum over μ' can be evaluated using the selection rule Eq. (34) for each of the R-Gaunt coefficients. In addition, the summation over ℓ' is restricted by the requirements

$$\ell' \in \{\ell_1+\ell_2, \ell_1+\ell_2-2, \ldots, |\ell_1-\ell_2|(+1)\} \quad (41)$$

and

$$\ell' \in \{\ell_0+\ell_3, \ell_0+\ell_3-2, \ldots, |\ell_0-\ell_3|(+1)\} \quad (42)$$

Hence, both $\ell_1+\ell_2$ and $\ell_0+\ell_3$ must be even or odd for a non-zero result. In combination with the above-mentioned symmetry of the generalized R-Gaunt coefficients, this implies that in the selection rule the number of odd ℓ_i must be even. Equivalently,

$$\ell_0+\ell_1+\ell_2+\ell_3 = 2n \quad n: \text{ integer} \quad (43)$$

has to hold.

As a further selection rule for the generalized Gaunt coefficient it may be noted that the numbers of negative elements in the sets $\{\mu_1,\mu_2\}$ and $\{\mu_0,\mu_3\}$ have to be either both even or both odd. In combination with the above-mentioned symmetry of the generalized R-Gaunt coefficients, this implies that the total number of negative μ_i must be even as for the R-Gaunt coefficients.

It should be noted that the last selection rule also follows from the definition Eq. (39) in combination with Eq. (22) and the observation that real spherical harmonics are for $\mu < 0$ ($\mu \geq 0$) odd (even) functions of ϕ with respect to $\phi = \pi$. Likewise, the selection rules Eqs. (35) and (43) follow from the defining integrals and the parity $(-1)^\ell$ of $X_\ell^\mu(\Omega)$.

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APPENDIX A: Some properties of Gaunt coefficients

Here we state some properties of coupling coefficients of complex spherical harmonics.

Since complex spherical harmonics are a CONS, the product of two complex spherical harmonics can be represented as a linear combination of complex spherical harmonics according to

$$Y_{\ell_1}^{m_1}(\Omega)Y_{\ell_2}^{m_2}(\Omega) = \sum_{\ell m} \langle \ell m|\ell_1 m_1|\ell_2 m_2\rangle Y_\ell^m(\Omega) \quad (A1)$$

The coefficients in this sum are from the orthonormality of the complex spherical harmonics given by

$$\langle \ell m|\ell_1 m_1|\ell_2 m_2\rangle = \int [Y_\ell^m(\Omega)]^* Y_{\ell_1}^{m_1}(\Omega) Y_{\ell_2}^{m_2}(\Omega)\mathrm{d}\Omega \quad (A2)$$

They are called Gaunt coefficients. These coefficients satisfy a number of selection rules which simplify the range of the summation over ℓ and m in the last formula considerably. The selection rules for $\langle \ell m|\ell_1 m_1|\ell_2 m_2\rangle$ are the following [1]. The magnetic quantum numbers have to satisfy the equation

$$m_1+m_2 = m \quad (A3)$$

For the angular momentum quantum numbers, we have that

$$\ell+\ell_1+\ell_2 = 2n \quad n: \text{integer} \quad (A4)$$

must hold. This, together with the general coupling rules for angular momenta, implies that

$$\ell \in \{\ell_{\max}, \ell_{\max}-2, \ldots, \ell_{\min}\} \quad (A5)$$

where

$$\ell_{\max} = \ell_1+\ell_2 \quad (A6)$$

and

$$\ell_{\min} = \begin{cases} k(\ell_1,\ell_2,m_1,m_2) \text{ if } k(\ell_1,\ell_2,m_1,m_2)+\ell_{\max} \\ \quad \text{is even} \\ k(\ell_1,\ell_2,m_1,m_2)+1 \text{ if } k(\ell_1,\ell_2,m_1,m_2)+\ell_{\max} \\ \quad \text{is odd} \end{cases} \quad (A7)$$

$$k(\ell_1,\ell_2,m_1,m_2) = \max(|\ell_1-\ell_2|, |m_1+m_2|)$$

If the selection rules are not satisfied, the Gaunt coefficient is necessarily zero. Hence, one may also write

$$Y_{\ell_1}^{m_1}(\Omega)Y_{\ell_2}^{m_2}(\Omega) = \sum_{\ell=\ell_{min}}^{\ell_{max}} (2)\,\langle \ell m_1 + m_2 | \ell_1 m_1 | \ell_2 m_2 \rangle \times Y_{\ell}^{m_1+m_2}(\Omega) \quad \text{(A8)}$$

where $\sum^{(2)}$ indicates summation in steps of two.

Gaunt coefficients are real. They have the following symmetries:

$$\begin{aligned} \langle \ell m | \ell_1 m_1 | \ell_2 m_2 \rangle &= \langle \ell m | \ell_2 m_2 | \ell_1 m_1 \rangle \\ &= \langle \ell - m | \ell_1 - m_1 | \ell_2 - m_2 \rangle \\ &= (-1)^{m+m_1} \langle \ell_1 - m_1 | \ell - m | \ell_2 m_2 \rangle \end{aligned} \quad \text{(A9)}$$

Generalized Gaunt coefficients ([7], Eq. (10)) are defined as

$$\langle \ell m | \ell_1 m_1 \ldots \ell_{k-1} m_{k-1} | \ell_k m_k \rangle = \int [Y_{\ell}^{m}(\Omega)]^* Y_{\ell_1}^{m_1}(\Omega) \ldots Y_{\ell_k}^{m_k}(\Omega) \mathrm{d}\Omega \quad \text{(A10)}$$

They can be reduced to sums over products of Gaunt coefficients using Eq. (A1) repeatedly. For instance, we have

$$\langle \ell m | \ell_1 m_1 \ell_2 m_2 | \ell_3 m_3 \rangle = \sum_{\ell' m'} \langle \ell' m' | \ell_1 m_1 | \ell_2 m_2 \rangle \langle \ell m | \ell' m' | \ell_3 m_3 \rangle \quad \text{(A11)}$$

Note that the sum over m' can be evaluated using the selection rule Eq. (A3) twice and that this gives rise to a selection rule for the generalized Gaunt coefficient.

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