Complete Multipole Basis Set for Single-Centered Electron Systems

Hiroaki Kusunose¹, Rikuto Oiwa¹, and Satoru Hayami²

¹Department of Physics, Meiji University, Kanagawa 214-8571, Japan
²Department of Applied Physics, The University of Tokyo, Bunkyo, Tokyo 113-8656, Japan

A whole series of expressions for four species of multipoles (electric, magnetic, magnetic toroidal, and electric toroidal) is provided as a complete basis set to describe arbitrary single-centered spinful electron systems. A compact formula to calculate matrix elements of these multipoles is also derived. A visualization method of an electronic state characterized in terms of multipoles is proposed. The complete basis set is useful to narrow down a candidate order parameter of electron systems in phase transition, to describe a property of cross-correlated phenomena, to analyze spectra of x-ray scattering in magnetically ordered state, and so on. We demonstrate a usage of the complete basis set by taking monopole and toroidal dipole orderings, and the mutual relationship among three distinct magnetic dipoles (orbital, spin angular momenta and anisotropic dipole) in a spin-orbit coupled system as prime examples.

1. Introduction

A concept of multipole is widely used in various fields of physics such as classical electromagnetism,¹⁻³ nuclear physics,⁴⁻⁶ solid-state physics,⁷⁻¹⁵ meta-materials,¹⁶⁻¹⁸ and so on. In addition to well-known electric (E) and magnetic (M) multipoles in elementary electromagnetism, there exist magnetic toroidal (MT) and electric toroidal (ET) multipoles, which have common spatial parity to their time-reversal counterparts.¹³⁻¹⁴,¹⁹,²⁰ Since these four species of multipoles are sufficient to describe arbitrary degrees of freedom of electromagnetic properties, they have been utilized to express multiple degrees of freedom of electrons in solids in accordance with symmetry point of view.

In order to discuss electronic states microscopically in terms of multipoles, their quantum mechanical operator expressions are required. In spinless systems, the operator expressions of E, M and MT multipoles have been obtained on the basis of the so-called multipole expansions of electromagnetic potentials.¹³⁻¹⁵ As for the remaining of ET multipole, which does not appear in the multipole expansion, its operator expression can be deduced from the time-reverting operator to M multipole.¹³ Moreover, such multipole expansions are straightforwardly extended to spinful systems by including the spin contribution to the electric current.

However, the multipoles introduced through the multipole expansions do not constitute a complete set, and quite a few multipoles are missing to satisfy the closure relation, especially for a spinful space. Motivated by this circumstances, we provide a systematic definition of spinful multipoles of four species in this paper. We derive a compact formula to calculate the matrix elements of a series of multipole operators with respect to total angular momentum basis or direct product of orbital and spin angular momentum bases. Since 32 crystallographic point groups are subgroups of rotational group symmetry are obtained merely by an appropriate linear combination of the expressions in the rotational group derived in this paper.

They are also utilized to describe the multipoles in momentum space,¹⁴ since the hopping integrals are essentially single-centered quantity from their hopping origins. The matrix elements of these augmented multipoles defined over a cluster are also obtained by the mapping between a sub-lattice and a molecular orbital basis of a cluster.

The organization of this paper is as follows. In §2, we first introduce the definition of spinless multipoles and their matrix elements in orbital angular momentum basis, and then we extend the discussion to spinful multipoles and their matrix elements. We give explicit expressions of spinful multipoles up to rank 1. In §3, we discuss the relation between multipoles defined by the multipole expansions and those in a complete set derived in the previous section. It becomes clear which multipoles are missing in the multipole expansions. In §4, we propose two complementary ways of visualization of an electronic state, which are useful to grasp anisotropy of electronic states, and the mutual relationship among distinct multipole degrees of freedom. In §5, we demonstrate a practical usage of the complete basis set by taking the simplest system with total angular momenta, J ≡ 1/2 and 3/2, in the s and p orbitals. The final section summarizes the paper. In three Appendices, we give detailed derivations of the matrix elements, and the relation between the multipoles in the expansions and those in a complete set.

2. Complete Multipole Basis Set

2.1 For spinless systems

First, let us summarize a complete multipole basis set for spinless systems. We have already discussed in the literature that four species of multipole operators can describe arbitrary electronic degrees of freedom in orbital states, which are characterized by the orbital angular momentum, L, and its component, M. They are defined as

\[
\hat{O}_{l,m}^{(\text{orb})} = \hat{O}_{l,m},
\]

\[
\hat{\mu}_{l,m}^{(\text{orb})} = \frac{1}{2l+1} \left[ (\nabla \hat{O}_{l,m}) \cdot \hat{l} + \hat{l} \cdot (\nabla \hat{O}_{l,m}) \right],
\]

\[
\hat{I}_{l,m}^{(\text{orb})} = \frac{2}{2l+1} \left[ (\nabla \hat{O}_{l,m}) \cdot (r \times \hat{l}) - (\hat{l} \times r) \cdot (\nabla \hat{O}_{l,m}) \right],
\]

in this paper, they are also useful to express cluster and bond extensions of multipoles.²⁴⁻⁳¹
(4)

where \( \hat{L} = -i(r \times \nabla) \) is the dimensionless orbital angular momentum operator, and the prefactor 1/2 is due to symmetrization of the operators. Note that in these expressions (\( \nabla \)) should be understood that \( \nabla \) acts only on \( O_{j,m} \), and

\[
O_{j,m}(r) = \sqrt{\frac{4\pi}{2l+1}} r^l Y_{1,0,0}^l(\hat{r}), \quad \hat{r} = r.
\]

is proportional to the spherical harmonics of the orbital angular momentum (rank of multipole), \( l = 0, 1, 2, \ldots \) and its z-component, \( m = -l, -l+1, \ldots, l \). We adopt the Racah normalization and the Condon-Shortley phase convention, where \( \hat{\rho}_{l,m} \) and \( \hat{\rho}_{l,m} \) represent their time-reversal parities (even or odd).

By considering the fact that \( Y_{1,0,0}^l \) transforms like \( Y_{1,0,0}^l \) with respect to the spatial rotation, the matrix element of the orbital angular momentum basis, \( |\eta, L, M \rangle \) (\( l = 1, 2 \)), can be decomposed as

\[
\langle n_1 L_1 M_1 | Y_{1,0,0}^l | n_2 L_2 M_2 \rangle = (-1)^{m_1} \left( \frac{L_1}{M_1 M_2} \right) \langle n_1 L_1 | Y_{1,0,0}^l | n_2 L_2 \rangle,
\]

where the parenthesis represents the Wigner’s 3j symbol, and \( \langle n_1 L_1 | Y_{1,0,0}^l | n_2 L_2 \rangle \) is the so-called reduced matrix element. Their explicit expressions are given below. The additional index \( n \) indicates quantum numbers other than \( L_1, M_1 \) such as the principal quantum number. This is a consequence of the Wigner-Eckart theorem.

The reduced matrix elements for \( X = Q, M \) and \( G \) are given by

\[
\langle n_1 L_1 M_1 | Q_{l,m}^{(Q)} | n_2 L_2 \rangle = (-1)^{l_1} \left( \frac{l_2}{l_1 l_2} \right) \sqrt{2l + 1} \left( l_1 l_2 l \right) \langle n_1 L_1 | Q_{l,m}^{(Q)} | n_2 L_2 \rangle,
\]

where the curly bracket is the Wigner’s 6j symbol, and

\[
\langle l \rangle_{12} = \int_0^\infty dr r^l Y_{1,0,0}^l(r) Y_{1,0,0}^l(r),
\]

is the matrix element in the radial part.

### Table 1. Correspondence between \( X_{i,m}^{(l)} \) and \( X_{i,m}^{(Q)} \) in spin sector.

| \( X_{i,m}^{(l)} \) | \( k = 0 \) | \( k = \pm 1 \) |
|-----------------|-----------|-----------|
| \( Q_{l,m}^{(Q)} \) | \( T_{l,m}^{(Q)} \) | \( M_{l,m}^{(Q)} \) |
| \( T_{l,m}^{(Q)} \) | \( G_{l,m}^{(Q)} \) | \( G_{l,m}^{(Q)} \) |

The reduced matrix elements for \( X = T \) and \( G \) are proportional to those for \( Q \) and \( M \) as

\[
\langle n_1 L_1 | T_{l,m}^{(Q)} | n_2 L_2 \rangle = i R_{L_1, L_2} \langle n_1 L_1 | Q_{l,m}^{(Q)} | n_2 L_2 \rangle,
\]

where the common proportional coefficient is given by

\[
R_{L_1, L_2} = \frac{L_1(L_1+1) - L_2(L_2+1)}{(l+1)(l+2)}. \tag{12}
\]

From this expression, it is apparent that \( T_{l,m}^{(Q)} \) and \( G_{l,m}^{(Q)} \) are non-active for \( L_1 = L_2 \). Since the Wigner’s symbols and the radial matrix elements are real, the matrix elements of \( Q_{l,m}^{(Q)} \) and \( M_{l,m}^{(Q)} \) are real, while those of \( T_{l,m}^{(Q)} \) and \( G_{l,m}^{(Q)} \) are pure imaginary. The detailed derivation is given in Appendix A.

### 2.2 For spinful systems

Next, we extend the complete multipole basis set to spinful (two-component spinor) systems. The spinful space can be decomposed into charge and spin sectors; the 2 \( \times 2 \) identity matrix \( \sigma_0 \) acts on charge sector, while the Pauli matrices (half of them are the dimensionless spin operators) \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) act on spin sector, respectively. Since \( \sigma_0 \) and \( \sigma \) are regarded as rank 0 and 1 tensors, respectively, it is natural to construct the spinful multipole operators by composing \( \sigma_0 \) and \( \sigma \) with \( X_{l,m}^{(Q)} \) in accordance with the addition rule of angular momentum. The definition of the composed spinful multipole operators is given by

\[
X_{l,m}^{(s)} \equiv \rho^{ik}(k=1, \ldots, 10) \sqrt{2l + 1} \left( l_1 l_2 l \right) \langle n_1 L_1 M_1 | X_{l,m}^{(Q)} | n_2 L_2 \rangle \times \prod_{n=1}^{l} \left( \frac{l + k}{m - n} \right) \sigma_{l,m}^{(s)} \sigma_{l,m}^{(s)} \sigma_{l,m}^{(s)} \sigma_{l,m}^{(s)} \tan \theta \right)^{\frac{1}{2}}.
\]

Here, the indices \( s = 0 \) and \( k = 0 \) specify a multipole in charge sector with \( \sigma_{0,0} = \sigma_0 \), while \( s = 1 \) and \( k = -1, 0, 1 \) specify that in spin sector where three spin components (\( n = 0, \pm 1 \)) are defined as \( \sigma_{i,j} = \sigma_i \) and \( \sigma_{i,j} = \pm (\sigma_i \pm i \sigma_j)/\sqrt{2} \). Thanks to the phase factor \( i^{s+k} \) the spinful multipole operator also satisfies

\[
\left( X_{l,m}^{(s)} \right)^\dagger = (-1)^{m} X_{l,-m}^{(s)}.
\]

It is easily confirmed that \( X_{l,m}^{(s)}(0) = X_{l,m}^{(Q)} \sigma_0 \) in charge sector.

Since \( \sigma \) is the time-reversal odd axial vector, the time-reversal parity of \( X_{l,m}^{(s)} \) is opposite to that of \( X_{l,m}^{(Q)} \) for \( k = 0, \pm 1 \), and the spatial parity is opposite as well for \( k = \pm 1 \) components. The correspondence is summarized in Table I.

In the presence of spin-orbit coupling, it is natural to use the
eigenstates \( |JM; L \rangle \) of the total angular momentum operator \( \hat{J} = J + \sigma/2 \) as a spinful basis. They are explicitly given by

\[
|JM; L \rangle = (-1)^{J-M} \sqrt{2J+1} \sum_{m=0}^{\pm 1} \left( \begin{array}{ccc} L & J & 1/2 \\ M & -\sigma & \sigma \end{array} \right) |L, M - \sigma \rangle |\sigma \rangle.
\]

(15)

\( J \) is positive half integer, and \( M = J, J + 1, \cdots, J \). The orbital angular momentum \( L \) in \( |JM; L \rangle \) (non-negative integer) is omitted in what follows for notational simplicity.

In this basis, the matrix element of \( X_{lm}^{(s)}(k) \) is also decomposed as eq. (7) by the Wigner-Eckart theorem, since \( X_{lm}^{(s)}(k) \) transforms like \( Y_{lm} \) under the spatial rotation:

\[
\langle n_1 J_1 M_1 | X_{lm}^{(s)}(k) | n_2 J_2 M_2 \rangle
\]

\[
= (-1)^{J_1-M_1} \left( \begin{array}{ccc} J_1 & J_2 & l \\ M_1 & M_2 & m \end{array} \right) \langle n_1 J_1 | X_{lm}^{(s)}(k) | n_2 J_2 M_2 \rangle.
\]

(16)

The reduced matrix elements \( \langle n_1 J_1 | X_{lm}^{(s)}(k) | n_2 J_2 \rangle \) can be expressed in terms of \( \langle n_1 L_1 || X_{lm}^{(orb)} || n_2 L_2 \rangle \) as defined in eqs. (8), (9), and (11). The detailed derivation is given in Appendix B, and the results are given as follows. In charge sector, it is given by

\[
\langle n_1 J_1 | X_{lm}^{(0)}(0) | n_2 J_2 \rangle = (-1)^{J_1+2l_2+1} \sqrt{2l_1 + 1 \left( 2l_2 + 1 \right)} \langle n_1 L_1 || X_{lm}^{(orb)} || n_2 L_2 \rangle \times \left( \begin{array}{ccc} L_1 & J_1 & l_2 \\ J_2 & l_1 & 1/2 \end{array} \right).
\]

(17)

In spin sector,

\[
\langle n_1 J_1 | X_{lm}^{(1)}(k) | n_2 J_2 \rangle
\]

\[
= -i^{-k+1} P_l^{(k)}(J_1, J_2; L_1, L_2) \langle n_1 L_1 || X_{lm}^{(orb)} || n_2 L_2 \rangle,
\]

(18)

with

\[
P_l^{(k)}(J_1, J_2; L_1, L_2) = \sqrt{6(2l_1 + 1)(2l_2 + 1)} \times \left( \begin{array}{ccc} J_1 & J_1 & 1/2 \\ L_1 & L_2 & 1/2 \end{array} \right),
\]

(19)

where the curly bracket is the Wigner's 9j symbol. Using these expressions, eq. (11) and Table I, we obtain explicit relations between \( \langle n_1 J_1 || X_{lm}^{(1)}(k) || n_2 J_2 \rangle \) and \( \langle n_1 L_1 || Q_{lm}^{(orb)} || n_2 L_2 \rangle \) or \( \langle n_1 L_1 || M_{lm}^{(orb)} || n_2 L_2 \rangle \) for \( k = 0 \) and \( k = \pm 1 \) as

\[
\langle n_1 J_1 || Q_{lm}^{(1)}(0) || n_2 J_2 \rangle = R_l P_l^{(0)}(n_1 L_1 || Q_{lm}^{(orb)} || n_2 L_2),
\]

\[
\langle n_1 J_1 || M_{lm}^{(1)}(0) || n_2 J_2 \rangle = R_l P_l^{(0)}(n_1 L_1 || M_{lm}^{(orb)} || n_2 L_2),
\]

\[
\langle n_1 J_1 || T_{lm}^{(1)}(0) || n_2 J_2 \rangle = -i P_l^{(0)}(n_1 L_1 || Q_{lm}^{(orb)} || n_2 L_2),
\]

\[
\langle n_1 J_1 || G_{lm}^{(1)}(0) || n_2 J_2 \rangle = -i P_l^{(0)}(n_1 L_1 || M_{lm}^{(orb)} || n_2 L_2),
\]

(21)

\[
\langle n_1 J_1 || Q_{lm}^{(1)}(\pm 1) || n_2 J_2 \rangle = \pm P_l^{(\pm 1)}(n_1 L_1 || M_{lm}^{(orb)} || n_2 L_2),
\]

\[
\langle n_1 J_1 || M_{lm}^{(1)}(\pm 1) || n_2 J_2 \rangle = \pm P_l^{(\pm 1)}(n_1 L_1 || Q_{lm}^{(orb)} || n_2 L_2),
\]

\[
\langle n_1 J_1 || T_{lm}^{(1)}(\pm 1) || n_2 J_2 \rangle = \pm i R_l P_l^{(\pm 1)}(n_1 L_1 || M_{lm}^{(orb)} || n_2 L_2),
\]

\[
\langle n_1 J_1 || G_{lm}^{(1)}(\pm 1) || n_2 J_2 \rangle = \pm i R_l P_l^{(\pm 1)}(n_1 L_1 || Q_{lm}^{(orb)} || n_2 L_2),
\]

\[
\langle n_1 J_1 || Q_{lm}^{(1)} || n_2 J_2 \rangle = \frac{1}{\sqrt{3}}(t \times \sigma),
\]

(25)

\[
M^{(1)}(l) = \frac{1}{\sqrt{3}}(r \cdot \sigma),
\]

(26)

\[
G^{(1)}(l) = \frac{1}{\sqrt{5}}(t \cdot \sigma),
\]

(27)

\[ Q^{(0)}(0) = \sigma_0, \quad Q^{(1)}(0) = \frac{1}{\sqrt{2}}(l \cdot \sigma), \]

(28)

\[ R^{(0)}(0) = \sigma_0, \quad R^{(1)}(0) = \frac{1}{\sqrt{2}}(r \times \sigma), \]

\[ [Q^{(1)}(l)]^2 = \frac{2}{3\sqrt{10}}\left(3(r \times \sigma_0 \sigma_x + 3(r \times \sigma_0 \sigma_y)ight). \]
The ET vector $(\mathbf{r} \cdot \sigma) r - \frac{p^2}{3} \sigma$, (29)

\[ T^{(0)}(0) = i \sigma_0, \quad T^{(1)}(0) = \frac{1}{\sqrt{2}} (\sigma \times r), \]

\[ [T^{(1)}(1)]_z = \frac{1}{2 \sqrt{10}} \left[ 3(\ell z)_z \sigma_x + 3(\ell t)_z \sigma_y \right. \]

\[ + 2[2(l_zz) - (l)_xz - (l)_yz] \sigma_z \] (cyclic), (30)

\[ G^{(1)}(1) = \frac{1}{\sqrt{2}} (\sigma \times \ell), \]

\[ [G^{(1)}(1)]_z = \frac{1}{2 \sqrt{10}} \left[ 3(\ell r)_z \sigma_x + 3(\ell r)_z \sigma_y \right. \]

\[ + 2[2(l_zr) - (l)_xz - (l)_yz] \sigma_z \] (cyclic). (31)

Here, $e_{1z} = e_z$ and $e_{1 \pm z} = \pm (e_x \pm e_y)/\sqrt{2}$, and $(AB)_ij = (A_i B_j + A_j B_i + B_j A_i + B_i A_j)/4$, and $e_i (i = x, y, z)$ are unit vectors in the Euclidean coordinate.

The ET monopole $Q^{(1)}(1)$ is nothing but the atomic spin-orbit coupling, and the M monopole $M^{(1)}(1)$ is the atomic limit of the so-called magnetic flux. The ET monopole $G^{(1)}(1)$ is an essential ingredient for chiral and strong gyrotropic point groups.

$M^{(1)}(1)$ represents the anisotropic magnetic dipole operator, which is independent of the primary orbital and spin angular momentum operators, $M^{(0)}(0) = I$ and $M^{(1)}(1) = \sigma_0$. This type of the anisotropic magnetic dipole has been known to appear in the context of the x-ray magneto-circular dichroism (XMCD), referred to as $T$-vector in literatures.32-35

The MT dipole $T^{(1)}(k)$ is relevant to the time-reversal odd axial tensor such as the linear magneto-electric effect.21 The ET vector $G^{(1)}(k)$ appears in the off-diagonal elements of the time-reversal even polar tensor such as Seebeck effect.14

3. Relation to Multipoles Defined by Potential Expansion

Multipoles usually appear in the multipole expansions of the scalar and vector potentials.6,8,20,36 In other words, the multipoles $X_{lm}$ are defined through the expansions. In the Coulomb gauge $\nabla \cdot \mathbf{A} = 0$, the expansions in unit of $-e = -\mu_B = 1$ are given by

\[ \phi(r) = \sum_{lm} \sqrt{\frac{4\pi}{2l+1}} \langle Q_{lm} \rangle Y_{lm}^{*}(\hat{r}) \frac{r^l}{r^{l+1}}, \] (32)

\[ A(r) = \sum_{lm} i \sqrt{\frac{4\pi}{2l+1}} \langle M_{lm} \rangle Y_{lm}^{*}(\hat{r}) \frac{r^l}{r^{l+1}} \]

\[ - \sqrt{\frac{4\pi}{(l+1)(l+2)}} \langle T_{lm} \rangle Y_{lm+1}^{*}(\hat{r}) \frac{r^{l+1}}{r^{l+2}} \], (33)

where $Y_{lm}^{*}(\hat{r}) (k = 0, \pm 1)$ is the vector spherical harmonics, whose definition is given by eq. (C-1), and $(X_{lm})$ indicates an appropriate thermal average of the multipole operator $X_{lm}$.

Note that which component of multipoles appears in the expansions depends on the gauge fixing condition. The ET multipole $G_{lm}$ does not appear in the expansion. In order to define the ET multipole operator in a systematic way, the operator $R_T = t_3 \cdot \nabla$, which reverts the time-reversal parity with keeping the spatial parity, is used as $G_{lm} \equiv R_T Q_{lm}$, where $t_3$ is the elementary MT dipole.13

The multipole operators $X_{lm}$ in the context of the potential expansions do not constitute a complete basis set, and quite a few spinful multipoles are missing. In fact, the multipole $X_{lm}$ hereby can be expanded by the spinful multipole $X_{lm}^{(s)}(k)$ as follows (see Appendix C in detail):

\[ Q_{lm} = Q_{lm}^{(s)} \sigma_0 = Q_{lm}^{(0)}(0), \]

\[ M_{lm} = M_{lm}^{(s)} \sigma_0 + (\nabla O_{lm}) \cdot \sigma \]

\[ = M_{lm}^{(0)}(0) + \sqrt{l(2l-1)} M_{lm}^{(1)}(-1), \]

\[ T_{lm} = T_{lm}^{(s)} \sigma_0 \]

\[ + \frac{1}{2l+1} \left[ (\nabla O_{lm}) \cdot (r \times \sigma) + (r \times \sigma) \cdot (\nabla O_{lm}) \right] \]

\[ = T_{lm}^{(0)}(0) - \frac{l}{2l+1} T_{lm}^{(1)}(0), \]

\[ G_{lm} = G_{lm}^{(s)} \sigma_0 + \frac{1}{2} \sum_{ij} \left[ (\nabla_i \nabla_j O_{lm}) g_{ij} + \hat{s}_{ij}(\nabla_i \nabla_j O_{lm}) \right] \]

\[ = G_{lm}^{(0)}(0) + \frac{l\sqrt{l(2l-1)}}{l+2} G_{lm}^{(1)}(-1) - \frac{l}{l+1} \frac{G_{lm}^{(1)}(1)}{l+2}, \] (37)

where

\[ g_{ij} = \frac{2(r \times \ell)_i (r \times \ell)_j}{(l+1)(l+2)} + \frac{2(r \times \sigma)_i (r \times \sigma)_j}{(l+1)^2}. \] (38)

Although there exists the term proportional to $(r \times \sigma)_i \sigma_j$ in the definition of $G_{lm}$, it is shown to vanish identically.

It should be emphasized that the expressions of $Q_{lm}$ were obtained by assuming that the sources of $\phi(r)$ and $A(r)$ are ordinary density of electron charge $\rho(r)$ and orbital and spin contributions to the electric current $j(r) = j_{\text{ord}}(r) + j_{\text{spin}}(r)$. However, other multipoles which do not appear in the expansions are also able to be an order parameter through many-body interactions. Once such an order parameter appears, it induces other multipoles belonging to the same irreducible representation of it. Eventually, other multipoles such as $G_{lm}^{(1)}(k)$ and $G_{lm}^{(1)}(-1)$ must contribute to $\phi(r)$, and $M_{lm}^{(1)}(l+1)$, and $T_{lm}^{(1)}(\pm 1)$ to $A(r)$ in this sense.

4. Visualization of Electronic States

When a thermal average of specific multipole operator becomes finite, the corresponding anisotropy appears. Thus, it is useful to visualize the anisotropy that characterizes an electronic state in the presence of multipoles.38 We show two complementary ways to visualize the anisotropy of a system having eigenstates $|\psi_i\rangle$ with the energy $E_i$.

4.1 Based on charge and angular-momentum distributions

A thermal average of an operator $A$ at inverse temperature $\beta$ is given by

\[ \langle A \rangle = \frac{1}{Z} \sum_{\gamma} e^{-\beta E_i} \langle \psi_i | A | \psi_i \rangle, \quad Z = \sum_{\gamma} e^{-\beta E_i}. \] (39)
By using the total angular-momentum basis \( (i = nJM; L) \) as 
\[ |\psi_p\rangle = \sum_n U_{i_n}^\dagger |i_1\rangle |J\rangle |M\rangle |\sigma\rangle, \]
the expectation value of \( A \) with respect to the \( \gamma \) eigenstate is expressed as

\[
\langle \psi_\gamma | A | \psi_\gamma \rangle = \sum_{i_1,i_2} U_{i_1}^\dagger U_{i_2} \sum_i \int d\tilde{r} \psi_{i_1}^* (\tilde{r}, \sigma) \psi_i (\tilde{r}, \sigma) \langle i_1 | A | i_2 \rangle
\]

\[
= \int \frac{d\tilde{r}}{4\pi} \bar{A}_\gamma (\tilde{r}),
\]
(40)

where we have inserted the completeness relation, 1 = \( \sum_\sigma \int d\tilde{r} |\tilde{r}\rangle \langle \tilde{r}| \sum_i |i_1\rangle \langle i_1|, \)

\[ \psi_i (\tilde{r}, \sigma) = (\tilde{r}|i\rangle = (-1)^{i+M} \sqrt{2J+1} \]

\[ \times \left( \frac{L}{M-\sigma} - J \right) ^{1/2} \sigma \right) Y_{LM-M} (\tilde{r}). \]
(41)

Then, the angular distribution is given by

\[ \bar{A}_\gamma (\tilde{r}) = \sum_{i_1} \text{Re} \left[ U_{i_1}^\dagger \sum_i \int d\tilde{r} \psi_{i_1}^* (\tilde{r}, \sigma) \psi_i (\tilde{r}, \sigma) \right] \]

\[ = (-1)^{i+M+iJ+M_1+1} 4\pi \int (2J+1)(2J_1+1) \]

\[ \times \left( \frac{L_1}{M_1-\sigma} - J_1 \right) ^{1/2} \sigma \right) Y_{LM-M}^* (\tilde{r}) Y_{L_1M_1-M_1}(\tilde{r}). \]
(42)

The angular distribution of the thermal average \( \langle A \rangle \) is thus given by

\[ \bar{A}(\tilde{r}) = \frac{1}{2} \sum_\gamma e^{-\beta E_\gamma} \bar{A}_\gamma (\tilde{r}). \]
(43)

In the case of spinless systems, we replace \( i \) and \( P_{i_1}^\dagger (\tilde{r}) \) in eq. (42) with \( i = nLM \) and \( P_{i_1} (\tilde{r}) = 4\pi Y_{LM}(\tilde{r}) Y_{L_1M_1}(\tilde{r}) \).

In order to visualize an electronic state, the angular distribution of specific multipole operator, such as the E charge \( \rho = Q (0) \), and the M dipoles \( I = M^0 (0) \), \( \sigma = M^{(1)} (-1) \), are useful. These operators are given in §2.4. For instance, \( \bar{A}(\tilde{r}) \) is used to express the shape of the wavefunction, and its magnetic property is displayed by the M dipole distributions.

### 4.2 Based on multipole charge densities

The thermal average of the multipoles is related with the polarization or magnetization density \( X(\tilde{r}) \) as

\[ \langle X_{lm} \rangle = \int d\tilde{r} \bar{X}(\tilde{r}) \cdot (\nabla \delta_{lm}) = \int d\tilde{r} \bar{X}_{lm}(\tilde{r}) \delta_{lm}, \]
(44)

where we have introduced the corresponding multipole “charge” density as \( \rho_{X}(\tilde{r}) = -\nabla \cdot \bar{X}(\tilde{r}) \). Here, \( X_{lm} \) is either \( X^{(0)}(k) \), \( X^{(0)}(0) \) or \( X_{lm} \). This is utilized to visualize an electronic state as follows. To this end, we decompose \( \rho_{X}(\tilde{r}) \) into the radial part \( \rho_{X}(\tilde{r}) \) and angular part \( \bar{X}_{lm}(\tilde{r}) \) as \( \rho_{X}(\tilde{r}) = \rho_{X}(\tilde{r}) \bar{X}_{lm}(\tilde{r}) / 4\pi \). The latter can be extracted by the completeness relation of the spherical harmonics as

\[ \bar{X}_{lm}(\tilde{r}) = \sum_{lm} (2l+1) \frac{X_{lm}(\tilde{r}) \delta_{lm}}{\langle r^2 \rangle}, \]
(45)

where we have introduced the radial average \( \langle r^2 \rangle \equiv \int_0^\infty dr r^2 \rho_{X}(r) \), which is roughly given by the average \( \langle r^2 \rangle_{12} \) over relevant orbitals, \( L_1 \) and \( L_2 \). For example, \( \bar{X}_{lm}(\tilde{r}) \) is used to express the shape of the wavefunction, and its M, MT, and ET charge distributions, i.e., \( \bar{X}_{M}(\tilde{r}) \), \( \bar{X}_{MT}(\tilde{r}) \), and \( \bar{X}_{ET}(\tilde{r}) \) are plotted as a color map on the surface of the shape.

5. Examples

In order to demonstrate the complete multipole basis set, we consider \( J = 1/2 \) and \( J = 3/2 \) systems with \( L = 0 (s) \) and \( 1 (p) \) orbitals as an example. The eigenstates of the total angular momentum \( |J, M; L) \) are given in terms of the linear combinations of the direct product of the orbital \( |L, M \pm \sigma) \) and spin \( |\sigma) \) states as

\[
\begin{align*}
|\frac{1}{2}, \frac{1}{2} ; s \rangle & = |0, 0) \rangle, \\
|\frac{1}{2}, -\frac{1}{2} ; s \rangle & = |0, 0) \rangle,
\end{align*}
(46)

\[
\begin{align*}
|\frac{1}{2}, \frac{3}{2} ; p \rangle & = \sqrt{\frac{2}{3}} |1, +1) \rangle - \sqrt{\frac{1}{3}} |1, 0) \rangle, \\
|\frac{1}{2}, \frac{1}{2} ; p \rangle & = -\sqrt{\frac{1}{2}} |1, -1) \rangle + \sqrt{\frac{1}{3}} |1, 0) \rangle, \\
|\frac{3}{2}, \frac{3}{2} ; p \rangle & = |1, +1) \rangle, \\
|\frac{3}{2}, -\frac{3}{2} ; p \rangle & = \sqrt{\frac{2}{3}} |1, +1) \rangle + \sqrt{\frac{1}{3}} |1, 0) \rangle, \\
|\frac{3}{2}, -1 ; p \rangle & = \sqrt{\frac{1}{3}} |1, -1) \rangle + \sqrt{\frac{2}{3}} |1, 0) \rangle, \\
|\frac{3}{2}, -\frac{1}{2} ; p \rangle & = |1, -1) \rangle.
\end{align*}
(47)

The wavefunctions of the basis \( |J, M; L) \) are visualized in Fig. 1. where \( \bar{X}_{M}(\tilde{r}) \) and \( \bar{X}_{MT}(\tilde{r}) \) are used for the shape and colormap, respectively.

![Fig. 1. Charge and magnetic charge densities of the wavefunctions](image_url)
Table II. Active multipoles in $J = 1/2(s)$, $1/2(p)$, and $3/2(p)$. The upper (lower) off-diagonal represents electric (magnetic) multipoles. We use the abbreviations, $\rho = Q^0(0)$, $G_\sigma = G^{p1}(1)$, $M_s = M^{s1}(1)$, $M_\sigma = M^{s1}(1)$, $r = Q^0(0)$, $Q_\sigma = Q^{p1}(0)$, $l = M^{s1}(1)$, $s = M^{s1}(1)$, $t = T^0(0)$, $T_\sigma = T^{p1}(0)$, and $Q_s = G^{s1}(0)$. $X^{(1)}(k)$ and $X^{(1)}(k)$ represent quadrupole and octupole, respectively.

| $J$     | 1/2 ($s$) | 1/2 ($p$) | 3/2 ($p$) |
|---------|-----------|-----------|-----------|
| 1/2 ($s$) | $\rho$ | $G_\sigma$ | $Q_\sigma$ |
|          | $\sigma$ | $r$        | $G^{s1}_{(1)}(1)$ |
| 1/2 ($p$) | $M_\sigma$ | $\rho$ | $G_\sigma$ |
|          | $t$ | $l$ | $Q^{p1}_{(1)}(1)$ |
| 3/2 ($p$) | $M^{s1}_{(1)}(1)$ | $M_s$ | $Q^{s1}_{(1)}(1)$ |
|          | $T_\sigma$ | $T^{p1}_{(2)}(0)$ | $\sigma$, $M^{s1}_{(1)}(1)$ |

Table III. Angular distributions for the M- and ET-monopole, and the MT- and ET-dipole eigenstates. $X$ is the essential vector that characterizes the wavefunction.

| $X_{j_{\mu}(k)}$ | $\mathbf{p} (\mathbf{r})$ | $X$ | $\overline{X} (\mathbf{r})$ |
|------------------|---------------------------|-----|---------------------|
| $M_\sigma$       | 1                         | $\sigma$ | $\mathbf{r}$ |
| $G_\sigma$       | 1                         | $G_\sigma$ | $-\frac{1}{\sqrt{2}} \mathbf{p}$ |
| $T^{v}_\sigma$   | $\frac{1}{4} (5 - 3 \hat{z}^2)$ | $\sigma$ | $\frac{\sqrt{6}}{2} (\hat{y}, -\hat{x}, 0)$ |
| $G^{v}_\sigma$   | $\frac{1}{4} (3 \hat{z}^2 - 1)$ | $Q_\sigma$ | $\frac{1}{12\sqrt{2}} (-\hat{y} - \sqrt{2} \hat{x}, \hat{y} + \sqrt{2} \hat{x}, 0)$ |

5.1 The M and ET monopoles

First, we consider the ground states of the following Hamiltonians,

$$H_{M0} = -M_\sigma, \quad H_{ET0} = -G_\sigma.$$  (49)

The ground states are doubly degenerate, and they are given by

$$M_\sigma : | \pm \rangle = \frac{1}{\sqrt{2}} \left[ \left| \frac{1}{2}, \frac{1}{2}, \pm 1; s \right> - \left| \frac{1}{2}, \frac{1}{2}, \pm 1; p \right> \right],$$  (50)

$$G_\sigma : | \pm \rangle = \frac{1}{\sqrt{2}} \left[ \left| \frac{1}{2}, \frac{1}{2}, \pm 1; s \right> + i \left| \frac{1}{2}, \frac{1}{2}, \pm 1; p \right> \right],$$  (51)

respectively.

For these ground states, angular distributions are summarized in the upper rows in Table III. Both in the M- and ET-monopole ground states, the angular distribution of $\rho$ is isotropic due to the nature of $J = 1/2$ wavefunctions in Fig. 1. In the M-monopole $M_\sigma$ ground state, the monopole flux appears in $\mathbf{p} (\mathbf{r})$ as shown in Fig. 2(a). On the other hand, in the ET-monopole $G_\sigma$ ground state, there are no characteristic angular distributions in ordinary physical quantities such as the E dipole $r$, and the M dipoles $l$ and $\sigma$. Since the ET monopole is pseudoscalar, there must exist a monopole flux. Indeed, it appears in the angular distribution of $G_\sigma = (\sigma \times l) / \sqrt{2}$ as shown in Fig. 2(b). Although it is difficult to observe directly $G_\sigma$ itself, the ET monopole flux results in hedgehog-type spin polarization in momentum space in periodic systems, since $G_\sigma \cdot r \sim (\sigma \times l) \cdot r = (l \times r) \cdot \sigma - t \cdot \sigma$ has the same symmetry as $k \cdot \sigma$ where $k$ is the wave vector. Thus, an observation of the hedgehog spin textures in momentum space is helpful to identify the emergence of the ET monopole.

5.2 The MT and ET dipoles

Next, we consider the ground states of the following Hamiltonians,

$$H_{MT1} = -T^{v}_\sigma, \quad H_{ET1} = -G^{v}_\sigma.$$  (52)

The ground states are doubly degenerate, and they are given by

$$T^{v}_\sigma : | \pm \rangle = \frac{1}{\sqrt{3}} \left[ \left| \frac{1}{2}, \pm 1; s \right> \pm i \frac{1}{\sqrt{2}} \left| \frac{1}{2}, \pm 1; p \right> \right],$$  (53)

$$G^{v}_\sigma : | \pm \rangle = \frac{1}{\sqrt{3}} \left[ \left| \frac{1}{2}, \pm 1; s \right> \pm i \frac{3}{\sqrt{2}} \left| \frac{1}{2}, \pm 1; p \right> \right],$$  (54)

respectively.

In the MT-dipole $T^{v}_\sigma$ ground state, the vortex-like angular distribution of $\sigma$ arises perpendicular to $T^{v}_\sigma$ as shown in Fig. 2(c). It is characterized by the vorticity, $\int d\mathbf{r} \mathbf{r} \times \mathbf{v}$, which is $-\mathbf{e}_z$. The charge distribution becomes anisotropic since the ground state is the superposition of anisotropic wavefunctions in Fig. 1. The angular dependences are given in the lower rows in Table III. Similarly, the wavefunction of the ET-dipole $G^{v}_\sigma$ ground state becomes anisotropic as shown in Fig. 2(d). However, there are no indications of the toroidal nature in ordinary physical quantities. Interestingly, it appears in less ordinary E-dipole involving spin degrees of freedom, namely, $Q_\sigma = (\sigma \times t) / \sqrt{2}$. By looking at this quantity, we realize the toroidal nature in this ground state as shown in Fig. 2(d).

In this way, the spinful multipoles are required to characterize the multipole ordered states in general.
5.3 The anisotropic M dipole

The anisotropic M dipole is often significant in analysis of XMCD spectra.\(^{32-35,37}\) In such a context, the relation among orbital wavefunction in a certain principal axis and three distinct M dipoles, \(I\), \(\sigma\), and \(M_o\) \(M^{(1)}(1)\) provides useful information.

To elucidate the relation between them, let us consider a simplified situation in a magnetically ordered state, which is described by the following Hamiltonian,

\[
H_{M1} = -\sqrt{3} \lambda Q^{(1)}(1) - \epsilon Q^{(0)}(0) - h_s \cdot \sigma - h_o \cdot I,
\]

(55)

where the first two terms represents the spin-orbit coupling \(I \cdot \sigma\) and the locking potential with sufficiently large \(\epsilon\) that makes the z axis of \(p_z\) orbital be the principal axis, namely, the crystalline electric field (CEF) potential. The last two terms are the Zeeman coupling with the spin and orbital angular momenta which represent the molecular fields from the surrounding magnetic moments. We set \(\epsilon = 1\) and assume the molecular magnetic field in \(zx\) plane, i.e., \(h_{s,o} = h_{s,o}(\sin \theta, 0, \cos \theta)\) in the following discussion.

First, we consider the case without the spin-orbit coupling, \(\lambda = 0\), and dominant spin ordering, \(h_s > 0\) and \(h_o = 0\). The ground state energy is given by \(E_{gs} = -2/5 - h\), and its eigenstate is

\[
|0\rangle = \frac{1}{\sqrt{3}} \left[ \cos \frac{\theta}{2} |1, 1, 1; p\rangle - \sin \frac{\theta}{2} |1, 1, -1; p\rangle - \sqrt{2} \cos \frac{\theta}{2} \sin \frac{\theta}{2} |1, 0, 0; p\rangle \right].
\]

(56)

Note that the wavefunction is independent of the magnitude of \(h_s\). In this ground state, the charge and three M-dipole distributions are given by

\[
\overline{a}(\rho) = 3 \rho^2,
\]

\[
\overline{\sigma}(\rho) = 3 \rho^2 (\sin \theta, 0, \cos \theta),
\]

\[
\overline{I}(\rho) = 0,
\]

\[
\overline{M}_s(\rho) = \frac{3 \rho^2 \cos \theta}{5 \sqrt{10}} (3 \hat{x} - 2 \hat{z} \tan \theta, 3 \hat{y}, 4 \hat{z} + 3 \hat{x} \tan \theta).
\]

(57)

The expectation values are obtained by the angular average as

\[
\overline{p} = 1,
\]

\[
\overline{\sigma} = (\sin \theta, 0, \cos \theta),
\]

\[
\overline{M}_s = -\frac{2}{5 \sqrt{10}} (\sin \theta, 0, -2 \cos \theta).
\]

(58)

Note that \(\overline{\sigma}\) and \(\overline{M}_s\) become perpendicular with each other at the so-called magic angle \(\theta_0 = \cos^{-1}(1/\sqrt{3}) \approx 54.7356^\circ\). These angular distributions at \(\theta = \theta_0\) are shown in Fig. 3(a).

When we switch on the molecular field to \(I\), the relative directions of three M dipoles are altered. Figure 3(b) shows three M-dipole angular distributions for \(h_s = h_o = 1\) at the magic angle \(\theta = \theta_0\). \(I\) and \(\sigma\) tend to align in the same direction of the magnetic field because of the Zeeman couplings, while \(M_s\) tends to direct the opposite direction to \(I\) and \(\sigma\). Note that the angular average of the M dipole distributions lies in \(zx\) plane. The shape of the wavefunction is also deformed. As shown in this example, a careful consideration of the contributions from three distinct M dipoles is necessary in analyzing XMCD spectra.

Moreover, we discuss the influence of the spin-orbit coupling. To this end, let us consider the limit of strong spin-orbit coupling, \(\lambda = \infty\). For simplicity, we put \(h_o = 0\). In this case, we obtain the explicit analytical solution only for \(\theta = \theta_0\), and the ground state energy is given by

\[
E_{gs} = -\frac{1}{15} \sqrt{9 + 125 h_s^2 + 10 h_s \sqrt{27 + 100 h_s^2}}.
\]

(59)

The corresponding eigenstate is also obtained analytically, however we omit it as it is rather lengthy. The angular distributions of three M dipoles for weak magnetic field \(h_s = 0.1\) and strong one \(h_s = 1\) are shown in Fig. 4. The angular average of the M dipole distributions also lies in \(zx\) plane. Interestingly, the effect of the spin-orbit coupling on the mutual interplay among three M dipoles is similar to that shown in Fig. 3(b) in which the molecular fields are applied both on \(I\) and \(\sigma\) in the absence of \(\lambda\). This is because the effective coupling among three M dipoles arises through the E quadrupole \(Q^{(0)}(0)\). Namely, there are the 3rd order couplings in the free energy,

\[
(M_s^2 \left[ Q^{(1)}(1) + Q^{(0)}(0) \right], \quad I, \sigma, M_s Q^{(0)}(0)).
\]

(60)

The combination of these couplings gives rise to the effective coupling among three M dipoles, the spin-orbit coupling, \(I, \sigma\), and the CEF potential, \(Q^{(0)}(0)\). The general treatment of the algebra of multipoles and the Landau expansion in terms of them are discussed in Supplemental Materials in detail.

![Fig. 3. Angular distributions of the spin \(\sigma\), anisotropic M dipole \(M_s\), and orbital angular momentum \(I\) in the absence of the spin-orbit coupling. The magnetic field \(h\) is applied in \(zx\) plane, and \(\theta\) is fixed at the magic angle \(\theta_0\). (a) \(h_o = 0\), (b) \(h_o = h_s = 1\).](image-url)
where we have used the integration formula for the product of vector spherical harmonics. Then, we obtain eq. (9) by the comparison with eq. (7).

A.2 Relation between E, M and MT, ET multipoles

Moreover, we discuss the relation between the multipoles and toroidal multipoles. First, let us consider \( T_{l,m}^{(orb)} \). By the relation, \( r \times (\nabla \mathbf{O}_{l,m}) = \mathbf{i} \ell \mathbf{O}_{l,m}^{(orb)} \), the following expression appearing in the definition of \( T_{l,m}^{(orb)} \) is reexpressed as

\[
\frac{1}{2} \left[ (\mathbf{O}_{l,m}) \cdot (r \times l) - (l \times r) \cdot (\mathbf{O}_{l,m}) \right] = \frac{1}{2} \left[ -(r \times \nabla \mathbf{O}_{l,m}) - l \cdot l \cdot (r \times \nabla \mathbf{O}_{l,m}) \right] = -\frac{1}{2} \left[ \hat{F} \mathbf{O}_{l,m} - \mathbf{O}_{l,m} \hat{F} \right],
\]

where we have used the identities,

\[
\mathbf{O}_{l,m} = \mathbf{O}_{l,m}^{(orb)} - \mathbf{O}_{l,m} \hat{l}. \]

Note that the parenthesis specifies on what range the operators \( l \) and \( \nabla \) act. Acting on the bra (ket) state on the first (second) term in the above expression, we obtain eq. (11) for \( T_{l,m}^{(orb)} \) and \( \mathbf{G}_{l,m}^{(orb)} \).

Similarly, by comparing both definitions of \( \mathbf{G}_{l,m}^{(orb)} \) and \( \mathbf{M}_{l,m}^{(orb)} \), we obtain

\[
\mathbf{G}_{l,m}^{(orb)} = \frac{4i}{2(l+1)^2} \left[ (\nabla \mathbf{O}_{l,m}) \cdot \hat{l} \hat{F} - \hat{F} \hat{l} \cdot (\nabla \mathbf{O}_{l,m}) \right] = -i \left[ \frac{1}{(l+1)^2} \right] \left[ \mathbf{M}_{l,m}^{(orb)} - \mathbf{M}_{l,m}^{(orb)} \hat{l} \right].
\]

Thus, the same relation, eq. (11), holds for \( \mathbf{G}_{l,m}^{(orb)} \) and \( \mathbf{M}_{l,m}^{(orb)} \).

Appendix B: Derivation of Reduced Matrix Elements of \( X_{l,m}^{(s)}(k) \)

In this appendix, the derivation of the reduced matrix elements of the spinful multipole operator \( X_{l,m}^{(s)}(k) \) is given.

Let us begin with the Wigner-Eckart theorem, eq. (16). By using the completeness relation for 3j symbol, we can revert eq. (16) as

\[
\langle J_1| \mathbf{X}_{l,m}^{(s)}(k) |J_2 \rangle = \sum_{m_1m_2} (-1)^{L_1+M_1} \langle J_1 M_1 | \mathbf{X}_{l,m}^{(s)}(k) |J_2 M_2 \rangle.
\]
From the definition of $X_{lm}(k)$, eq. (13) and by applying the Wigner-Eckart theorem separately for the orbital and spin parts, we obtain the matrix element in the right-hand-side of eq. (B-1) as

$$\langle J_1 M_1 | X_{lj}^{(k)}(k) | J_2 M_2 \rangle = i^{l+k} \sum_{M'_1 M_2 \sigma_1, \sigma_2} (-1)^{l+m_1+J_1 M_1 + J_2 M_2} \times \sqrt{(2l+1)(2J_1+1)(2J_2+1)} \langle L_1 M_1' | X_{lj}^{(orb)}(k) | L_2 M_2' \rangle \times \langle \sigma_1 | \sigma_1 \rangle \langle \sigma_2 | \sigma_2 \rangle \times \left( \frac{L_2}{M_2'} \begin{array}{c} J_2 \ 1/2 \\ M_2' \ -M_2 \end{array} \right) \left( \frac{L_1}{M_1'} \begin{array}{c} J_1 \ 1/2 \\ M_1' \ -M_1 \end{array} \right) \langle l+k \ l \ s \rangle \langle m' \ -m \ n \rangle.$$

Then, we substitute this expression into eq. (B-1), and using the definition of $j$ symbol in terms of the summation of product of six $j$ symbols, we obtain

$$\langle J_1 | X_{lj}^{(k)}(k) | J_2 \rangle = i^{l+k} (-1)^l \langle L_1 | X_{lj}^{(orb)}(k) | L_2 \rangle \times \sqrt{(2l+1)(2J_1+1)(2J_2+1)(1-s)!(2+s)!} \times \left( \frac{L_1}{J_1} \begin{array}{c} 1/2 \\ 1/2 \\ 0 \end{array} \right) \left( \frac{L_2}{J_2} \begin{array}{c} 1/2 \\ 1/2 \\ 0 \end{array} \right) \langle l+k \ l \ s \rangle \langle m' \ -m \ n \rangle.$$

This expression is eq. (18) for spin sector ($s=1$). For charge sector, by putting $s = k = 0$ and using the identity,

$$\left( \begin{array}{c} L_1 \ J_1 \ 1/2 \\ L_2 \ J_2 \ 1/2 \end{array} \right) = \frac{(-1)^{l+1/2+l_2+l} \sqrt{2(l+1)}}{(l+1)l} \left( \begin{array}{c} L_1 \ L_2 \ l \end{array} \begin{array}{c} 1/2 \\ 1/2 \end{array} \right),$$

we obtain eq. (17).

**Appendix C: Relation between Multipoles through Potential Expansion and Spinful Multipoles**

In this appendix, we express the multipole $X_{lm}$ defined through the electromagnetic potentials in eqs. (32) and (33) in terms of spinful multipole basis set, eq. (13). Since $X_{lm}$ always contains the orbital part, we discuss the remaining part $\delta X_{lm} = X_{lm} - X_{lm}^{(0)}$ containing the spin operator $\sigma$.

Let us begin with $\delta M_{lm} = \sigma \cdot (\nabla O_{lm})$. Since $\nabla O_{lm} = r^{l+1} Y_{lm}$ and by comparing the definitions of $M_{lm}^{(1)}(-1)$ with that of the vector spherical harmonics,

$$Y_{lm}^{(k)}(\hat{r}) = (-1)^{l+m} \sqrt{2l+1} \sum_n (l+k \ l \ s \ -m \ m' \ n) e_{lm}^{(k)}(\hat{r}) \sigma^{lm},$$

we obtain eq. (35) as

$$\delta M_{lm} = \sigma \cdot (\nabla O_{lm}) = \sqrt{(2l-1)} M_{lm}^{(1)}(-1).$$

For the MT multipole, $\delta T_{lm}$ is proportional to $(r \times \sigma) \cdot (\nabla O_{lm})$, which can be deformed as $(r \times \sigma) \cdot (\nabla O_{lm}) = -(r \times \nabla O_{lm}) \cdot \sigma = -i \langle \sigma | (O_{lm}) \rangle$. With $H_{lm}^{\mu} = \sqrt{(l+1)} Y_{lm}^{(1)}(0)$ and the comparison of the definitions between $Y_{lm}^{(1)}(0)$ and $T_{lm}^{(1)}(0)$, we have the relation, $\sigma \cdot (\nabla O_{lm}) = -i \sqrt{(l+1)} T_{lm}^{(1)}(0)$. Thus, we obtain eq. (36) as

$$\delta T_{lm} = -\sqrt{(l+1)} T_{lm}^{(1)}(0).$$

As for the ET multipole, first we consider the first part of eq. (38). This part can be deformed as

$$\frac{1}{2} \sum_{ij} ((\nabla \nabla O_{ij})(r \times l_1) - (l \times r)(\nabla \nabla O_{ij})) | \sigma \rangle = \frac{1}{2} \sqrt{(2l-1)} \sum_{ij} [(r \times \nabla \nabla O_{ij}) l_i + l_i (r \times \nabla \nabla O_{ij})] | \sigma \rangle = -i \sqrt{(l+1)} T_{lm}^{(1)}(0)$$

By using $(l \nabla O_{ij}) = l_i (\nabla \nabla O_{ij}) - (\nabla \nabla O_{ij}) l_i$, we have

$$\frac{1}{2} \sum_{ij} [(l \nabla \nabla O_{ij}) l_i + l_i (\nabla \nabla O_{ij})] | \sigma \rangle.$$

Then, the first term becomes

$$\delta G_{lm}^{(1st)} = -i \sqrt{(2l-1)} \sum_{ij} [(l \nabla \nabla O_{ij}) l_i + l_i (\nabla \nabla O_{ij})] | \sigma \rangle.$$

By noticing the identity derived from eqs. (20) and (21),

$$i R_{l1,l2} (L_1 | M_{lm}^{(1)}(-1) | L_2 M_2) = (J_1 M_1 | G_{lm}^{(1)}(-1) | J_2 M_2),$$

we obtain

$$\delta G_{lm}^{(1st)} = i \sqrt{(2l+1)} \sum_{ij} [(l \nabla \nabla O_{ij}) l_i + l_i (\nabla \nabla O_{ij})] | \sigma \rangle.$$
Therefore, we have
\[ -\frac{i}{2} \sum_{ij} \left[ l_i (\nabla_j O_{lm}) + (\nabla_j O_{lm}) l_i \right] \sigma_i \]
and the second term becomes
\[ \delta G^{(2nd)}_{lm} = -\frac{1}{2} \sqrt{l(l+1)} G^{(1)}_{lm}(0). \] (C.5)

Finally, we obtain the relation as in eq. (37).

1) V. Dubovik and A. Cheshkov: Sov. J. Part. Nucl. 5 (1975) 318.
2) L. D. Landau and E. M. Lifshitz: The Classical Theory of Fields, 4th ed. (Butterworth-Heinemann, Oxford, 1980).
3) S. Nanz: Toroidal Multipoles in Classical Electrodynamics: An Analysis of Their Emergence and Physical Significance (Springer, 2016).
4) R. J. Blin-Stoyle: Rev. Mod. Phys. 28 (1956) 75.
5) I. B. Zel’dovich: Sov. Phys. J. Exp. Theor. Phys. 6 (1958) 1184.
6) J. M. Blatt and V. F. Weisskopf: Theoretical Nuclear Physics (Dover Publications, New York, 1991).
7) R. Shina, H. Shiba, and P. Thalmeier: J. Phys. Soc. Jpn. 66 (1997) 1741.
8) H. Kusunose: J. Phys. Soc. Jpn. 77 (2008) 064710.
9) Y. Kuramoto: Prog. Theor. Phys. Suppl. 176 (2008) 77.
10) P. Santini, S. Carretta, G. Amoretti, R. Ciaccia, N. Magnani, and G. H. Lander: Rev. Mod. Phys. 81 (2009) 807.
11) Y. Kuramoto, H. Kusunose, and A. Kiss: J. Phys. Soc. Jpn. 78 (2009) 072001.
12) M.-T. Suzuki, H. Ikeda, and P. M. Oppeneer: J. Phys. Soc. Jpn. 87 (2018) 041008.
13) S. Hayami and H. Kusunose: J. Phys. Soc. Jpn. 87 (2018) 033709.
14) S. Hayami, M. Yatsushiro, Y. Yanagi, and H. Kusunose: Phys. Rev. B 98 (2018) 165110.
15) H. Watanabe and Y. Yanase: Phys. Rev. B 98 (2018) 245129.
16) T. Kaelberer, V. Fedotov, N. Papasimakis, D. Tsai, and N. Zheludev: Science 330 (2010) 1510.
17) V. Savinov, V. A. Fedotov, and N. I. Zheludev: Phys. Rev. B 89 (2014) 205112.
18) N. Papasimakis, V. Fedotov, V. Savinov, T. Raybould, and N. Zheludev: Nat. Mater. 15 (2016) 263.
19) V. Dubovik, L. Tосunyan, and V. Tugushev: Zh. Eksp. Teor. Fiz. 90 (1986) 590.
20) V. Dubovik and V. Tugushev: Phys. Rep. 187 (1990) 145.
21) N. A. Spaldin, M. Fiebig, and M. Mostovoy: J. Phys.: Condens. Matter 20 (2008) 434203.
22) Y. V. Kopaev: Physics-Uspekhi 52 (2009) 1111.
23) T. Y. Inui, T and Y. Ondera: Group Theory and Its Applications in Physics (Springer, Berlin, 1996) 2nd ed.
24) C. Ederer and N. A. Spaldin: Phys. Rev. B 76 (2007) 214404.
25) Y. Yanase: J. Phys. Soc. Jpn. 83 (2014) 014703.
26) S. Hayami, H. Kusunose, and Y. Motome: J. Phys.: Condens. Matter 28 (2016) 395601.
27) M.-T. Suzuki, T. Koretsune, M. Ochi, and R. Arita: Phys. Rev. B 95 (2017) 094406.
28) M. Matsumoto, K. Chimata, and M. Koga: J. Phys. Soc. Jpn. 86 (2017) 034704.
29) M.-T. Suzuki, T. Nomoto, R. Arita, Y. Yanagi, S. Hayami, and H. Kusunose: Phys. Rev. B 99 (2019) 174407.
30) F. Thölle and N. A. Spaldin: Philos. Trans. R. Soc. A 376 (2018) 20170450.
31) S. Hayami, Y. Yanagi, H. Kusunose, and Y. Motome: Phys. Rev. Lett. 122 (2019) 147602.
32) P. Carra, B. T. Thole, M. Altarelli, and X. Wang: Phys. Rev. Lett. 70 (1993) 694.
33) J. Stöhr: J. Electron Spectrosc. Relat. Phenom. 75 (1995) 253.
34) J. Stöhr and H. König: Phys. Rev. Lett. 75 (1995) 3748.
35) J. Crocombette, B. Thole, and F. Jollet: J. Phys.: Condens. Matter 8 (1996) 4095.
36) C. Schwartz: Phys. Rev. 97 (1955) 380.
37) Y. Yamauchi, H. Nakao, and T.-h. Arima: arXiv:1909.08179 (2019).
Supplemental Materials

We here provide the algebra of multipoles, and the Landau expansion are derived in order to discuss mutual coupling among multipoles. Then, the matrix elements of all active multipoles in $J = 1/2$ and $J = 3/2$ with $L = 0, 1$ used in the main text. Moreover, the irreducible basis functions of cubic $O_h$ and hexagonal $D_{6h}$ groups are also given. The expressions of the multipole operators and their matrix elements in 32 crystallographic point groups are obtained by using the coefficient of linear combination of the tesseral basis functions.

Appendix A: Algebra and Coupling of Multipoles

A.1 The property of multipole operators

Let us consider a complete multipole basis set, $\{X_i\}$. The multipole operator $X_i$ is $d \times d$ matrix and hermite, $X_i = X_i^\dagger$. There are $d^2$ mutually independent matrices, and they satisfy

$$\text{Tr}(X_iX_j) = d \delta_{ij} \quad (i, j = 0, 1, 2, \cdots d^2 - 1).$$ \hfill (A-1)

The 0-th component ($i = 0$) represents the unit matrix, $X_0 \equiv I$, and the other matrices are traceless $\text{Tr}(X_i) = 0 \,(i \neq 0)$.

A.2 The matrix product

Since $\{X_i\}$ constitute a complete set, we expand a product of multipoles as

$$X_iX_j = \sum_k h_{ijk}X_k,$$ \hfill (A-2)

where the coefficient $h_{ijk}$ is complex in general. By this relation and eq. (A-1), we obtain

$$h_{ijk} = \frac{1}{d}\text{Tr}(X_iX_jX_k).$$ \hfill (A-3)

Thus, the cyclic permutation of $h_{ijk}$ is identical, $i.e.$, $h_{ijk} = h_{jki} = h_{kij}$. Moreover, taking hermite conjugate of eq. (A-2) gives

$$X_jX_i = \sum_k h^*_{ijk}X_k \Rightarrow h_{ijk} = h^*_{jki}.$$ \hfill (A-4)

When we decompose $h_{ijk}$ into real and imaginary parts as $h_{ijk} = g_{ijk} + if_{ijk}$, each part satisfies

$$g_{ijk} = g_{jki} = g_{kji}, \quad f_{ijk} = f_{jki} = f_{kji} = -f_{kji} = -f_{jki} = -f_{ijk}.$$ \hfill (A-5)

Therefore,

$$X_iX_j = \sum_{k=0}^{d^2-1} (g_{ijk} + if_{ijk})X_k.$$ \hfill (A-6)

By tracing the above relation, we obtain

$$d \delta_{ij} = (g_{i00} + if_{i00})d.$$ \hfill (A-7)

Namely, we have

$$g_{i00} = \delta_{ij}, \quad f_{i00} = 0.$$ \hfill (A-8)

By using eq. (A-2) and its hermite conjugate, we obtain

$$g_{ijk} = \frac{1}{2d}\text{Tr}\left(X_iX_jX_k\right), \quad f_{ijk} = \frac{1}{2d^2}\text{Tr}\left(X_iX_j\right).$$ \hfill (A-9)

where $[A, B]_\pm = AB \pm BA$.

A.3 Free energy and Landau expansion

A.3.1 The free energy in the mean-field approximation

Let us consider a generalized exchange Hamiltonian,

$$H = -\frac{1}{2} \sum_{X'} \sum_{X''} \sum_{i,j} D^{\alpha\beta}_{ij} X'^\alpha_i X''^\beta_j,$$ \hfill (A-10)
where $s$ represents a site of the operators. We assume that $D_{ij}^{ss'} = D_{ij}^{ss'}$, $D_{ij}^{st} = D_{ij}^{st} = 0$, $\sum_{i}^{s} = \sum_{i}^{t}$ indicates that the summation excludes $i = 0$ component.

We consider the one-body trial Hamiltonian, $H_0(\phi) = -\sum_{s} \sum_{i} \phi_{ii} X_{is}^{s} \equiv \sum_{s} \sum_{i} H_{0s}(\phi_{is})$, and adopt the Feynman’s variational principle, we obtain the trial free energy as

$$F(\phi) = \sum_{s} F_{0s}(\phi_{is}) - \frac{1}{2} \sum_{s, s'} \sum_{ij} \frac{\partial^2 F_{0s}}{\partial \phi_{ij}^{s} \partial \phi_{ij}^{s'}} + \frac{1}{2} \sum_{s} \sum_{i} \phi_{ii} X_{is}^{s} , \quad F_{0s}(\phi_{is}) = -\frac{1}{\beta} \ln \text{Tr}(e^{-\beta H_0}) . \quad (A-11)$$

Here, the expectation value w.r.t. the trial Hamiltonian is given by

$$\langle X_{is}^{s} \rangle = \frac{\text{Tr}(e^{-\beta H_0} X_{is}^{s})}{\text{Tr}(e^{-\beta H_0})} = -\frac{\partial F_{0s}}{\partial \phi_{ij}^{s}} (i \neq 0) . \quad (A-12)$$

The variational condition of $F(\phi)$ gives

$$\phi_{ij}^{s} = \sum_{s} \sum_{i} D_{ij}^{s} \langle X_{is}^{s} \rangle \quad (i \neq 0) . \quad (A-13)$$

This is nothing but the self-consistent equation in the mean-field approximation.

### A.3.2 Landau expansion

Let us expand the free energy, $F_{0s}(\phi_{is})$, with respect to $\phi_{ij}^{s}$. Using the relations,

$$\frac{1}{d} \text{Tr}(\phi_{is} X_{is}^{s} X_{is}) = h_{ij} , \quad \frac{1}{d} \text{Tr}(\phi_{is} X_{is}^{s} X_{is}^{s'}) = \sum_{m} h_{ijkl} h_{mkl} ,$$

we obtain

$$\frac{1}{d} \text{Tr}(e^{-\beta H_0}) = 1 + \frac{\beta^2}{2} \sum_{i} \phi_{ii}^{s} + \frac{\beta^3}{3!} \sum_{ik} h_{ik} \phi_{ii}^{s} \phi_{ij}^{s} + \frac{\beta^4}{4!} \sum_{ijkl} h_{ijkl} \phi_{ii}^{s} \phi_{ij}^{s} \phi_{ij}^{s'} + \cdots .$$

Therefore, the free energy is expanded as

$$F_{0s}(\phi_{is}) = -\frac{1}{\beta} \ln d - \frac{\beta}{2} \sum_{i} \phi_{ii}^{s} + \frac{\beta^2}{3!} \sum_{ik} g_{ik} \phi_{ii}^{s} \phi_{ij}^{s} - \frac{\beta^3}{4!} \sum_{ijkl} \epsilon_{ijkl} \phi_{ii}^{s} \phi_{ij}^{s} \phi_{ij}^{s'} + \cdots . \quad (A-14)$$

In this expression, as $\phi_{ii}^{s} \phi_{ij}^{s} \phi_{ij}^{s'}$ is completely symmetric, the contributions of $f_{ijk}$ in 3rd order vanish, and only the contributions from $g_{ijk}$ remain. Similarly, since $\phi_{ii}^{s} \phi_{ij}^{s}$ or $\phi_{ij}^{s} \phi_{ij}^{s'}$ is symmetric with the permutation of $(i, j)$ or $(k, l)$, there remains $\sum_{m} g_{ijkl} g_{mkl}$ in $\sum_{m} h_{ijkl} h_{mkl}$ in 4th order. Therefore,

$$F_{0s}(\phi_{is}) = -\frac{1}{\beta} \ln d - \frac{\beta}{2} \sum_{i} \phi_{ii}^{s} - \frac{\beta^2}{3!} \sum_{ik} g_{ik} \phi_{ii}^{s} \phi_{ij}^{s} - \frac{\beta^3}{4!} \sum_{ijkl} \epsilon_{ijkl} \phi_{ii}^{s} \phi_{ij}^{s} \phi_{ij}^{s'} + \cdots . \quad (A-15)$$

Here, we have introduced

$$\epsilon_{ijkl} = \sum_{m} g_{ijkl} g_{mkl} - 3 \delta_{ij} \delta_{kl} + \sum_{m} g_{ijkl} g_{mkl} - 2 \delta_{ij} \delta_{kl} . \quad (A-15)$$

By the expansion of $F_{0s}(\phi_{is})$ and appropriate replacement of dummy indices, we obtain the relation,

$$\langle X_{is}^{s} \rangle = \beta \phi_{ii}^{s} + \frac{\beta^2}{2} \sum_{jk} g_{jk} \phi_{ii}^{s} \phi_{ij}^{s} + \frac{\beta^3}{3!} \sum_{ijkl} \epsilon_{ijkl} \phi_{ii}^{s} \phi_{ij}^{s} \phi_{ij}^{s'} + \cdots . \quad (A-16)$$

In order to invert this relation for $\phi_{ij}^{s}$, we assume the relation,

$$\beta \phi_{ij}^{s} = \langle X_{is}^{s} \rangle - \frac{\beta^2}{2} \sum_{jk} g_{jk} \phi_{ii}^{s} \phi_{ij}^{s} - \frac{\beta^3}{3!} \sum_{ijkl} \epsilon_{ijkl} \phi_{ii}^{s} \phi_{ij}^{s} \phi_{ij}^{s'} + \cdots ,$$

and insert it iteratively, then we obtain

$$\beta \phi_{ij}^{s} = \langle X_{is}^{s} \rangle - \frac{\beta^2}{2} \sum_{jk} g_{jk} \langle X_{is}^{s} \rangle - \frac{\beta^2}{2} \sum_{jk} g_{jk} \phi_{ii}^{s} \phi_{ij}^{s} \left[ \langle X_{is}^{s} \rangle - \frac{\beta^2}{2} \sum_{kl} g_{kl} \phi_{ii}^{s} \phi_{ij}^{s} \right] .$$
where we have introduced

\[ L_{ijkl} = \frac{1}{2} \left[ 3 \sum_m \hat{g}_{ijkl} g_{mkl} - \epsilon_{ijkl} \right] = \sum_m \hat{g}_{ijkl} g_{mkl} + \delta_{ijkl} \delta_{klm} = \sum_m \hat{g}_{ijkl} g_{mkl}. \]

Inserting this expression into eq. (A-11), we finally obtain the Landau expansion of the free energy

\[ F(X) = -N T \ln d + \frac{1}{2} \sum_{x'x} \sum_{ij} \left[ T \delta_{x'i} \delta_{ij} - D_{ij}^{x'} \right] \langle X_i' \rangle \langle X_j' \rangle \]

\[ - \frac{T}{3} \sum_s \sum_{ij} \hat{g}_{ijkl} \langle X_i' \rangle \langle X_j' \rangle + \frac{2T}{4!} \sum_s \sum_{ijkl} L_{ijkl} \langle X_i' \rangle \langle X_j' \rangle \langle X_k' \rangle \langle X_l' \rangle + \cdots, \]

where we have introduced

\[ \hat{L}_{ijkl} = \frac{1}{2} \left[ 3 \sum_m \hat{g}_{ijkl} g_{mkl} - \epsilon_{ijkl} \right] = \sum_m \hat{g}_{ijkl} g_{mkl} + \delta_{ijkl} \delta_{klm} = \sum_m \hat{g}_{ijkl} g_{mkl}. \]

Inserting this expression into eq. (A-11), we finally obtain the Landau expansion of the free energy

\[ F(X) = -N T \ln d + \frac{1}{2} \sum_{x'x} \sum_{ij} \left[ T \delta_{x'i} \delta_{ij} - D_{ij}^{x'} \right] \langle X_i' \rangle \langle X_j' \rangle \]

\[ - \frac{T}{3} \sum_s \sum_{ij} \hat{g}_{ijkl} \langle X_i' \rangle \langle X_j' \rangle + \frac{2T}{4!} \sum_s \sum_{ijkl} L_{ijkl} \langle X_i' \rangle \langle X_j' \rangle \langle X_k' \rangle \langle X_l' \rangle + \cdots, \]

where \( N \) is the number of the sites. Note that the lowest order coupling is given by \( g_{ijkl} \), i.e., eq. (A-9).

The coupling among multipoles is also obtained by the quantity,

\[ \text{Tr}(e^{-\beta H_{\text{int}} X_i'}) = \sum_{k=0}^{\infty} \frac{(-\beta)^k}{k!} \text{Tr}(H_{\text{int}}^k X_i'). \]

which becomes finite if the coupling to \( X_i' \) exists.

**Appendix B: \( J = 1/2 \) and \( 3/2 \) with \( L = 0 \) and 1 System**

**B.1 Basis**

The total angular-momentum basis \( | J, M; L \rangle \) used in the main text is given by

\[ \left| \frac{1}{2}, \frac{1}{2}; s \right> = |0, 0 \rangle \uparrow, \quad \left| \frac{1}{2}, -\frac{1}{2}; s \right> = |0, 0 \rangle \downarrow, \]

\[ \left| \frac{1}{2}, \frac{1}{2}; p \right> = \sqrt{\frac{2}{3}} |1, +1 \rangle \downarrow - \sqrt{\frac{1}{3}} |1, 0 \rangle \downarrow, \quad \left| \frac{1}{2}, -\frac{1}{2}; p \right> = - \sqrt{\frac{2}{3}} |1, -1 \rangle \uparrow + \sqrt{\frac{1}{3}} |1, 0 \rangle \uparrow, \]

\[ \left| \frac{3}{2}, \frac{3}{2}; p \right> = |1, +1 \rangle \uparrow, \quad \left| \frac{3}{2}, -\frac{1}{2}; p \right> = \frac{1}{\sqrt{3}} |1, +1 \rangle \uparrow + \sqrt{\frac{2}{3}} |1, 0 \rangle \uparrow, \]

\[ \left| \frac{3}{2}, -\frac{3}{2}; p \right> = \frac{1}{\sqrt{3}} |1, -1 \rangle \uparrow + \sqrt{\frac{2}{3}} |1, 0 \rangle \downarrow, \quad \left| \frac{3}{2}, -\frac{3}{2}; p \right> = |1, -1 \rangle \downarrow. \]

**B.2 Symmetry of multipoles**

The angle dependence of the multipoles used in the main text is given by

\( (x, y, z) : x, y, z \)

\( (u, v, w, z) : \frac{1}{2} (3z^2 - r^2), \frac{\sqrt{3}}{2} (x^2 - y^2), \sqrt{3} yz, \sqrt{3} zx, \sqrt{3} xy \)

\( (xyz, ax, ay, az, bx, by, bz) : \)

\[ \sqrt{15} xz y, \frac{1}{2} x (5x^2 - 3r^2), \frac{1}{2} y (5y^2 - 3r^2), \frac{1}{2} z (5z^2 - 3r^2), \frac{\sqrt{15}}{2} x (y^2 - z^2), \frac{\sqrt{15}}{2} y (z^2 - x^2), \frac{\sqrt{15}}{2} z (x^2 - y^2) \]
B.3 Matrix Element

B.3.1 Electric multipole (orbital)

rank 0

\[
Q^{(0)}(0) = \begin{pmatrix}
1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 1
\end{pmatrix}
\]

rank 1

\[
Q^{(1)}_1(0) = \begin{pmatrix}
0 & 0 & 0 & -\frac{1}{2} & -\frac{\sqrt{3}}{3} & 0 & \frac{1}{2} & 0 \\
0 & 0 & -\frac{1}{2} & 0 & 0 & -\frac{\sqrt{3}}{6} & 0 & \frac{1}{\sqrt{6}} \\
0 & -\frac{1}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{1}{2} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 0 & 0
\end{pmatrix}
\]

\[
Q^{(1)}_2(0) = \begin{pmatrix}
0 & 0 & 0 & -\frac{1}{2} & -\frac{\sqrt{3}}{3} & 0 & \frac{1}{2} & 0 \\
0 & 0 & -\frac{1}{2} & 0 & 0 & -\frac{\sqrt{3}}{6} & 0 & \frac{1}{\sqrt{6}} \\
0 & -\frac{1}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{1}{2} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 0 & 0
\end{pmatrix}
\]

\[
Q^{(1)}_3(0) = \begin{pmatrix}
0 & 0 & 0 & -\frac{1}{2} & -\frac{\sqrt{3}}{3} & 0 & \frac{1}{2} & 0 \\
0 & 0 & -\frac{1}{2} & 0 & 0 & -\frac{\sqrt{3}}{6} & 0 & \frac{1}{\sqrt{6}} \\
0 & -\frac{1}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{1}{2} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 0 & 0
\end{pmatrix}
\]

\[
Q^{(1)}_4(0) = \begin{pmatrix}
0 & 0 & 0 & -\frac{1}{2} & -\frac{\sqrt{3}}{3} & 0 & \frac{1}{2} & 0 \\
0 & 0 & -\frac{1}{2} & 0 & 0 & -\frac{\sqrt{3}}{6} & 0 & \frac{1}{\sqrt{6}} \\
0 & -\frac{1}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \frac{1}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{1}{2} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 0 & 0
\end{pmatrix}
\]
rank 2

\[ Q^{(0)}_s(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{2} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -\frac{1}{2} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{1}{2} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} \]

\[ Q^{(0)}_c(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{3} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -\frac{1}{3} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{1}{3} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} \]

\[ Q^{(0)}_v(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{6} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -\frac{1}{6} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{1}{6} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} \]

\[ Q^{(0)}_s(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} \]
### B.3.2 Electric multipole (spin)

#### rank 0

\[ Q^{(1)}(1) = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\frac{2\sqrt{3}}{9} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -\frac{2\sqrt{3}}{9} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{\sqrt{3}}{3} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{3} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \frac{\sqrt{3}}{3} \\ \end{pmatrix} \]

#### rank 1

\[ Q^{(1)}(0) = \begin{pmatrix} 0 & 0 & 0 & \frac{\sqrt{3}}{9} & -\frac{\sqrt{3}}{18} & 0 & \frac{1}{18} & 0 \\ 0 & \frac{\sqrt{3}}{9} & 0 & 0 & -\frac{1}{18} & 0 & \frac{\sqrt{3}}{18} & 0 \\ 0 & -\frac{\sqrt{3}}{9} & 0 & 0 & 0 & 0 & 0 & 0 \\ \frac{1}{18} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \end{pmatrix} \]

\[ Q^{(1)}(0) = \begin{pmatrix} 0 & 0 & 0 & -\frac{\sqrt{3}}{9} & -\frac{\sqrt{3}}{18} & 0 & -\frac{1}{18} & 0 \\ 0 & \frac{\sqrt{3}}{9} & 0 & 0 & \frac{1}{18} & 0 & -\frac{\sqrt{3}}{18} & 0 \\ 0 & -\frac{\sqrt{3}}{9} & 0 & 0 & 0 & 0 & 0 & 0 \\ \frac{1}{18} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \end{pmatrix} \]

\[ Q^{(1)}(0) = \begin{pmatrix} 0 & 0 & 0 & \frac{\sqrt{3}}{9} & 0 & 0 & \frac{1}{18} & 0 \\ 0 & 0 & -\frac{\sqrt{3}}{9} & 0 & 0 & \frac{1}{18} & 0 & 0 \\ \frac{\sqrt{3}}{9} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \end{pmatrix} \]
\[ Q^{(1)}_{\xi}(-1) = \left( \begin{array}{cccccccc} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{array} \right) \]

\[ Q^{(1)}_{\eta}(-1) = \left( \begin{array}{cccccccc} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{array} \right) \]

\[ Q^{(1)}_{\xi}(-1) = \left( \begin{array}{cccccccc} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{array} \right) \]

\[ Q^{(1)}_{\eta}(-1) = \left( \begin{array}{cccccccc} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{array} \right) \]
### B.4 Magnetic multipole (orbital)

**rank 1**

\[
M^{(0)}_s(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{2}{3} & \frac{-\sqrt{6}}{9} & 0 & \frac{1}{\sqrt{3}} & 0 \\
0 & 0 & \frac{2}{3} & 0 & 0 & 0 & \frac{\sqrt{3} \gamma}{\sqrt{2}} & 0 \\
0 & 0 & 0 & \frac{-\sqrt{6}}{9} & \frac{1}{3} & 0 & 0 & \frac{1}{\sqrt{3}} \\
0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{\sqrt{3}} & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{-\sqrt{6}}{9} & 0 & \frac{1}{\sqrt{3}} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

**rank 0**

\[
M^{(0)}_s(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{2}{3} & \frac{-\sqrt{6}}{9} & 0 & \frac{1}{\sqrt{3}} & 0 \\
0 & 0 & \frac{2}{3} & 0 & 0 & 0 & \frac{\sqrt{3} \gamma}{\sqrt{2}} & 0 \\
0 & 0 & 0 & \frac{-\sqrt{6}}{9} & \frac{1}{3} & 0 & 0 & \frac{1}{\sqrt{3}} \\
0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{\sqrt{3}} & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{-\sqrt{6}}{9} & 0 & \frac{1}{\sqrt{3}} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

### B.5 Magnetic multipole (spin)

**rank 0**

\[
M^{(0)}(1) = \begin{pmatrix}
0 & 0 & \frac{-\sqrt{6}}{9} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{-\sqrt{6}}{9} & 0 & 0 & 0 & 0 \\
\frac{-\sqrt{6}}{9} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]
\[ M_1^{(1)}(1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 2\sqrt{10} & \sqrt{15} & 0 & -\sqrt{15} & 0 \\
0 & 0 & \sqrt{15} & 0 & 0 & -\sqrt{10} & 0 & 0 \\
0 & 0 & 0 & \sqrt{15} & 0 & -\sqrt{10} & 0 & 0 \\
0 & 0 & 0 & -\sqrt{10} & 0 & 0 & -2\sqrt{10} & 0 \\
0 & 0 & 0 & 0 & 0 & -\sqrt{15} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{10} & 0 \\
\end{pmatrix} \]

\[ M_2^{(1)}(1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 2\sqrt{10} & \sqrt{15} & 0 & -\sqrt{15} & 0 \\
0 & 0 & \sqrt{15} & 0 & 0 & -\sqrt{10} & 0 & 0 \\
0 & 0 & 0 & \sqrt{15} & 0 & -\sqrt{10} & 0 & 0 \\
0 & 0 & 0 & -\sqrt{10} & 0 & 0 & -2\sqrt{10} & 0 \\
0 & 0 & 0 & 0 & 0 & -\sqrt{15} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{10} & 0 \\
\end{pmatrix} \]

\[ M_3^{(1)}(1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 2\sqrt{10} & \sqrt{15} & 0 & -\sqrt{15} & 0 \\
0 & 0 & \sqrt{15} & 0 & 0 & -\sqrt{10} & 0 & 0 \\
0 & 0 & 0 & \sqrt{15} & 0 & -\sqrt{10} & 0 & 0 \\
0 & 0 & 0 & -\sqrt{10} & 0 & 0 & -2\sqrt{10} & 0 \\
0 & 0 & 0 & 0 & 0 & -\sqrt{15} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{10} & 0 \\
\end{pmatrix} \]

\[ M_4^{(1)}(-1) = \begin{pmatrix}
0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\
1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -\sqrt{10} & \sqrt{15} & 0 & -\sqrt{10} & 0 \\
0 & 0 & \sqrt{10} & 0 & 0 & -\sqrt{15} & 0 & 0 \\
0 & 0 & 0 & -\sqrt{10} & \sqrt{15} & 0 & -\sqrt{10} & 0 \\
0 & 0 & 0 & \sqrt{10} & 0 & 0 & -\sqrt{15} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{15} \\
\end{pmatrix} \]

\[ M_5^{(1)}(-1) = \begin{pmatrix}
0 & -i & 0 & 0 & 0 & 0 & 0 & 0 \\
i & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -\sqrt{10} & \sqrt{15} & 0 & -\sqrt{10} & 0 \\
0 & 0 & \sqrt{10} & 0 & 0 & -\sqrt{15} & 0 & 0 \\
0 & 0 & 0 & -\sqrt{10} & \sqrt{15} & 0 & -\sqrt{10} & 0 \\
0 & 0 & 0 & \sqrt{10} & 0 & 0 & -\sqrt{15} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{15} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{15} \\
\end{pmatrix} \]
\[
M_{x}^{(1)}(-1) = \begin{pmatrix}
1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -\frac{1}{3} & 0 & 0 & -\frac{2\sqrt{3}}{9} & 0 & 0 \\
0 & 0 & 0 & \frac{1}{3} & 0 & 0 & -\frac{2\sqrt{3}}{3} & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & -\frac{2\sqrt{3}}{3} & 0 & 0 & \frac{1}{3} & 0 & 0 \\
0 & 0 & 0 & -\frac{2\sqrt{3}}{3} & 0 & 0 & \frac{1}{3} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 \\
\end{pmatrix}
\]

rank 2

\[
M_{x}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & \frac{\sqrt{3}}{3} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{3} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{\sqrt{3}}{3} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{y}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{3} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{\sqrt{3}}{3} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{z}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{3} & 0 & -\frac{1}{2} & 0 \\
0 & 0 & 0 & 0 & \frac{1}{2} & 0 & 0 & \frac{\sqrt{3}}{3} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{1}{2} & 0 & 0 & \frac{\sqrt{3}}{3} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{1}{2} & 0 & 0 & \frac{\sqrt{3}}{3} \\
\end{pmatrix}
\]

\[
M_{yz}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{3} & 0 & -\frac{1}{2} & 0 \\
0 & 0 & 0 & 0 & \frac{1}{2} & 0 & 0 & \frac{\sqrt{3}}{3} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{1}{2} & 0 & 0 & \frac{\sqrt{3}}{3} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{1}{2} & 0 & 0 & \frac{\sqrt{3}}{3} \\
\end{pmatrix}
\]
\[
M_{\alpha \alpha}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{\sqrt{15}}{3} \\
0 & 0 & 0 & 0 & 0 & \frac{\sqrt{15}}{3} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

rank 3

\[
M_{\alpha \alpha}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{\alpha x}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{\alpha y}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & -2 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 3 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 3 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 3 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{\alpha z}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{\alpha x}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{\alpha y}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
M_{\alpha z}^{(1)}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]
B.6 Magnetic toroidal multipole (orbital)

rank 1

\[ T^{(1)}_x(0) = \begin{pmatrix}
0 & 0 & 0 & -\frac{i}{9} & -\frac{\sqrt{5}}{18} & 0 & \frac{\sqrt{5}}{18} & 0 \\
0 & 0 & 0 & -\frac{i}{9} & 0 & 0 & -\frac{\sqrt{5}}{18} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 
\end{pmatrix} \]

\[ T^{(1)}_y(0) = \begin{pmatrix}
0 & 0 & 0 & -\frac{i}{9} & -\frac{\sqrt{5}}{18} & 0 & \frac{\sqrt{5}}{18} & 0 \\
0 & 0 & 0 & -\frac{i}{9} & 0 & 0 & -\frac{\sqrt{5}}{18} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 
\end{pmatrix} \]
B.7 Magnetic toroidal multipole (spin)

rank 1

\[
T^{(0)}(0) = 
\begin{pmatrix}
0 & 0 & -\frac{i}{\sqrt{3}} & 0 & 0 & \frac{\sqrt{3}}{3} & 0 & 0 \\
0 & 0 & 0 & \frac{i}{\sqrt{3}} & 0 & 0 & \frac{\sqrt{3}}{\sqrt{3}} & 0 \\
\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
\frac{-\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
e
\end{pmatrix}
\]

\[
T^{(1)}(0) = 
\begin{pmatrix}
0 & 0 & 0 & -\frac{i}{\sqrt{3}} & \frac{\sqrt{3}}{3} & 0 & -\frac{i}{\sqrt{3}} & 0 \\
0 & 0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & \frac{i}{\sqrt{3}} & 0 & -\frac{-\sqrt{3}}{\sqrt{3}} \\
\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
\frac{-\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
e
\end{pmatrix}
\]

\[
T^{(2)}(0) = 
\begin{pmatrix}
0 & 0 & 0 & -\frac{i}{\sqrt{3}} & \frac{\sqrt{3}}{3} & 0 & -\frac{i}{\sqrt{3}} & 0 \\
0 & 0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & \frac{i}{\sqrt{3}} & 0 & -\frac{-\sqrt{3}}{\sqrt{3}} \\
\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
\frac{-\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
e
\end{pmatrix}
\]

\[
T^{(3)}(0) = 
\begin{pmatrix}
0 & 0 & 0 & -\frac{i}{\sqrt{3}} & \frac{\sqrt{3}}{3} & 0 & -\frac{i}{\sqrt{3}} & 0 \\
0 & 0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & \frac{i}{\sqrt{3}} & 0 & -\frac{-\sqrt{3}}{\sqrt{3}} \\
\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
\frac{-\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
e
\end{pmatrix}
\]

\[
T^{(4)}(0) = 
\begin{pmatrix}
0 & 0 & 0 & -\frac{i}{\sqrt{3}} & \frac{\sqrt{3}}{3} & 0 & -\frac{i}{\sqrt{3}} & 0 \\
0 & 0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & \frac{i}{\sqrt{3}} & 0 & -\frac{-\sqrt{3}}{\sqrt{3}} \\
\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{i}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
\frac{-\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{\sqrt{3}}{\sqrt{3}} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
e
\end{pmatrix}
\]
rank 2

\[
T^{(1)}_e(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{\sqrt{2}}{2} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{\sqrt{2}}{2} & 0 & 0 \\
0 & 0 & \frac{\sqrt{6}}{2} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{\sqrt{6}}{2} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
T^{(1)}_t(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \frac{\sqrt{2}}{2} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{\sqrt{2}}{2} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
T^{(1)}_m(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{\sqrt{6}}{2} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{\sqrt{6}}{2} & 0 & 0 \\
0 & 0 & \frac{3}{10} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{3}{10} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
T^{(1)}_s(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{3}{10} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{3}{10} & 0 & 0 \\
0 & 0 & \frac{3}{10} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{3}{10} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
T^{(1)}_v(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{\sqrt{6}}{2} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{\sqrt{6}}{2} & 0 & 0 \\
0 & 0 & \frac{3}{10} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{3}{10} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]
B.8 Electric toroidal multipole (spin)

rank 0

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & -\frac{\sqrt{2}}{2} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -\frac{\sqrt{2}}{2} & 0 & 0 & 0 & 0 \\
\frac{\sqrt{2}}{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{\sqrt{2}}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 1

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 2

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 3

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 4

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 5

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 6

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 7

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 8

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 9

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$

rank 10

$$G^{(1)}(0) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}$$
rank 2

\[
G^{(1)}_n(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & \frac{\sqrt{3}}{9} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -\frac{\sqrt{9}}{9} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
-\frac{\sqrt{9}}{9} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{\sqrt{9}}{9} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
G^{(1)}_s(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{9} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{\sqrt{3}}{9} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
-\frac{\sqrt{9}}{9} & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
G^{(1)}_{s'}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
G^{(1)}_{c'}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
G^{(1)}_{c}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
G^{(1)}_{c'}(-1) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]
Appendix C: Cubic and Hexagonal Harmonics

We introduce the tesseral harmonics as

\[
O_{l,m} = \sqrt{\frac{4\pi}{2l+1}} r^l Y_{l,m} = (-1)^m O_{l,-m}^*,
\]

\[
C_{l,0} = O_{l,0}, \quad C_{l,m} = \frac{(-1)^m}{\sqrt{2}} (O_{l,m} + O_{l,-m}^*), \quad S_{l,m} = \frac{(-1)^m}{\sqrt{2}i} (O_{l,m} - O_{l,-m}^*).
\]

Then, the cubic \(O_h\) and hexagonal \(D_{6h}\) harmonics are expressed by the linear combinations of the tesseral harmonics. The expressions for other point group are obtained by using the compatibility relation of the irreducible representations.

In the following tables, we abbreviate \(C_{lm} \to C[m]\) and \(S_{lm} \to S[m]\). We use the multiplicity label for the basis functions belonging to the same irreducible representation in which \(m\) is descending order except for \(C[0]\). This convention sometimes differs from those used in literatures. Note that the basis functions are orthogonal with each other.

In cubic point group, \((x, y, z)\) components in \(T\) and \((u, v)\) component in \(E\) are related with each other by \(C_3\) rotation along [111] direction.

In hexagonal group, we use the principal axes as

\[
a = e_x, \quad b = -\frac{1}{2} e_x + \frac{\sqrt{3}}{2} e_y, \quad c = e_z.
\]

We take \(e_y\) axis as \(C_2'\) rotation axis. \((x, y)\) components in \(E\) correspond to \(C_{l,m}\) and \(S_{l,m}\), respectively.
### C.1 Cubic multipoles up to rank 11

#### rank 0

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1g}$ | $Q_0$ | $C[0]$ |

#### rank 1

| irrep. | symbol | definition |
|--------|--------|------------|
| $T_{1u}$ | $Q_x$ | $C[1]$ |
| $Q_y$ | $S[1]$ |
| $Q_z$ | $C[0]$ |

#### rank 2

| irrep. | symbol | definition |
|--------|--------|------------|
| $E_g$ | $Q_u$ | $C[0]$ |
| $Q_v$ | $C[2]$ |
| $T_{2g}$ | $Q_{yz}$ | $S[1]$ |
| $Q_{zx}$ | $C[1]$ |
| $Q_{xy}$ | $S[2]$ |

#### rank 3

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{2u}$ | $Q_3^u$ | $S[2]$ |
| $T_{1u}$ | $Q_{3x}^u$ | $- \frac{\sqrt{5} C[1]}{4} + \frac{\sqrt{15} C[3]}{4}$ |
| $Q_{3y}^u$ | $- \frac{\sqrt{5} C[1]}{4} - \frac{\sqrt{15} C[3]}{4}$ |
| $Q_{3z}^u$ | $C[0]$ |
| $T_{2u}$ | $Q_{3x}^u$ | $- \frac{\sqrt{10} C[1]}{4} - \frac{\sqrt{5} C[3]}{4}$ |
| $Q_{3y}^u$ | $\frac{\sqrt{10} C[1]}{4} - \frac{\sqrt{5} C[3]}{4}$ |
| $Q_{3z}^u$ | $C[2]$ |

#### rank 4

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1g}$ | $Q_4$ | $\frac{\sqrt{21} C[0]}{6} + \frac{\sqrt{15} C[4]}{6}$ |
| $E_g$ | $Q_{4u}$ | $\frac{\sqrt{15} C[0]}{6} - \frac{\sqrt{21} C[4]}{6}$ |
| $Q_{4v}$ | $-C[2]$ |
| $T_{1g}$ | $Q_{4x}^\gamma$ | $- \frac{\sqrt{14} C[1]}{4} - \frac{\sqrt{2} C[3]}{4}$ |
| $Q_{4y}^\gamma$ | $\frac{\sqrt{14} C[1]}{4} - \frac{\sqrt{2} C[3]}{4}$ |
| $Q_{4z}^\gamma$ | $S[4]$ |
| $T_{2g}$ | $Q_{4x}^\delta$ | $- \sqrt{5} C[1] + \sqrt{14} C[3]$ |
| $Q_{4y}^\delta$ | $- \sqrt{5} C[1] - \sqrt{14} C[3]$ |
| $Q_{4z}^\delta$ | $S[2]$ |

#### rank 5

| irrep. | symbol | definition |
|--------|--------|------------|
| $E_u$ | $Q_{5u}$ | $S[4]$ |
| $Q_{5v}$ | $-S[2]$ |
| $T_{1u}$ | $Q_{5x}^{2u}$ | $\frac{\sqrt{15} C[1]}{8} - \frac{\sqrt{70} C[3]}{16} + \frac{3 \sqrt{14} C[5]}{16}$ |
| $Q_{5y}^{2u}$ | $\frac{\sqrt{15} C[1]}{8} + \frac{\sqrt{70} C[3]}{16} + \frac{3 \sqrt{14} C[5]}{16}$ |
| $Q_{5z}^{2u}$ | $C[0]$ |
| $T_{2u}$ | $Q_{5x}^{2u}$ | $\frac{\sqrt{10} C[1]}{8} + \frac{9 \sqrt{2} C[3]}{16} + \frac{\sqrt{10} C[5]}{16}$ |
| $Q_{5y}^{2u}$ | $- \frac{\sqrt{10} C[1]}{8} + \frac{9 \sqrt{2} C[3]}{16} - \frac{\sqrt{10} C[5]}{16}$ |
| $Q_{5z}^{2u}$ | $C[2]$ |
### rank 6

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1g}$ | $Q_6^{A}$ | $\sqrt{3} C[0]/4 - \sqrt{14} C[4]/4$ |
| $A_{2g}$ | $Q_6^{A}$ | $\sqrt{17} C[2]/4 - \sqrt{5} C[6]/4$ |
| $E$ | $Q_{6u}$ | $\sqrt{14} C[0]/4 + \sqrt{2} C[4]/4$ |
| | $Q_{6v}$ | $\sqrt{7} C[2]/4 + \sqrt{1} T C[6]/4$ |
| $T_{1g}$ | $Q_{6x}^{a}$ | $\sqrt{3} S[1]/8 - \sqrt{3} S[3]/8 - \sqrt{2} S[5]/8$ |
| | $Q_{6y}^{a}$ | $\sqrt{3} S[1]/8 - \sqrt{3} S[3]/8 + \sqrt{2} S[5]/8$ |
| | $Q_{6z}^{a}$ | $S [4]$ |
| $T_{2g}$ | $Q_{6x}^{a}$ | $3 \sqrt{5} S[1]/16 + \sqrt{3} S[3]/16 + S [5]/16$ |
| | $Q_{6y}^{a}$ | $3 \sqrt{5} S[1]/16 - \sqrt{3} S[3]/16 + S [5]/16$ |
| | $Q_{6z}^{a}$ | $S [6]$ |

### rank 7

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{2u}$ | $Q_7^{A}$ | $\sqrt{7} S[2]/12 + \sqrt{6} S[6]/12$ |
| $E_u$ | $Q_{7u}$ | $S [4]$ |
| | $Q_{7v}$ | $\sqrt{6} S[2]/12 - \sqrt{7} S[6]/12$ |
| $T_{1u}$ | $Q_{7x}^{a}$ | $3 \sqrt{2} C[1]/32 + 3 \sqrt{3} C[1]/32 - \sqrt{3} S[1]/32 + \sqrt{2} S[1]/32$ |
| | $Q_{7y}^{a}$ | $-3 \sqrt{2} C[1]/32 + \sqrt{3} C[1]/32 - \sqrt{3} S[1]/32 - \sqrt{2} S[1]/32$ |
| | $Q_{7z}^{a}$ | $C[0]$ |
| $T_{2u}$ | $Q_{7x}^{a}$ | $-3 \sqrt{2} C[1]/32 - \sqrt{3} C[1]/32 + 3 \sqrt{5} C[5]/32 + \sqrt{3} C[7]/32$ |
| | $Q_{7y}^{a}$ | $-3 \sqrt{2} C[1]/32 + \sqrt{3} C[1]/32 + 3 \sqrt{5} C[5]/32 - \sqrt{3} C[7]/32$ |
| | $Q_{7z}^{a}$ | $C[4]$ |
| $T_{2u}$ | $Q_{7x}^{a}$ | $- \sqrt{5} S[1]/64 - 3 \sqrt{3} S[3]/64 - 5 \sqrt{5} S[5]/64 - \sqrt{3} C[7]/64$ |
| | $Q_{7y}^{a}$ | $\sqrt{5} S[1]/64 - 3 \sqrt{3} S[3]/64 + 5 \sqrt{5} S[5]/64 - \sqrt{3} C[7]/64$ |
| | $Q_{7z}^{a}$ | $C[6]$ |
| $T_{2u}$ | $Q_{7x}^{a}$ | $-15 \sqrt{6} C[1]/64 + 19 \sqrt{1} C[3]/64 - \sqrt{2} C[5]/64 - \sqrt{2} 0022 C[7]/64$ |
| | $Q_{7y}^{a}$ | $15 \sqrt{6} C[1]/64 + 19 \sqrt{1} C[3]/64 + \sqrt{2} C[5]/64 + \sqrt{2} 0022 C[7]/64$ |
| | $Q_{7z}^{a}$ | $C[2]$ |
### rank 8

| irrep. symbol | definition |
|---------------|------------|
| \( A_{1g} \) \( Q_8 \) | \( \sqrt{33} \mathbf{c}[0] + \sqrt{21} \mathbf{c}[4] + \sqrt{195} \mathbf{c}[8] \) |
| \( E_g \) \( Q_{8u}^{1} \) | \( -\frac{\sqrt{266}}{32} \mathbf{c}[0] + \frac{\sqrt{182}}{16} \mathbf{c}[4] + \frac{\sqrt{106}}{32} \mathbf{c}[8] \) |
| \( Q_{8v}^{1} \) | \( C[6] \) |
| \( E_g \) \( Q_{8u}^{2} \) | \( -\frac{\sqrt{210}}{32} - \frac{\sqrt{130}}{48} + \frac{\sqrt{960}}{96} \mathbf{c}[8] \) |
| \( Q_{8v}^{2} \) | \( C[2] \) |
| \( T_{1g} \) \( Q_{8x}^{\alpha 1} \) | \( \frac{\sqrt{75} \mathbf{c}[1]}{32} - \frac{\sqrt{75} \mathbf{c}[3]}{32} - \frac{\sqrt{75} \mathbf{c}[5]}{32} - \frac{\sqrt{555} \mathbf{c}[7]}{32} \) |
| \( Q_{8y}^{\alpha 1} \) | \( \frac{\sqrt{75} \mathbf{c}[1]}{32} - \frac{\sqrt{75} \mathbf{c}[3]}{32} + \frac{\sqrt{75} \mathbf{c}[5]}{32} - \frac{\sqrt{555} \mathbf{c}[7]}{32} \) |
| \( Q_{8z}^{\alpha 1} \) | \( S[8] \) |
| \( T_{1g} \) \( Q_{8x}^{\alpha 2} \) | \( -\frac{\sqrt{75} \mathbf{c}[1]}{32} + \frac{5 \sqrt{75} \mathbf{c}[3]}{32} - \frac{3 \sqrt{75} \mathbf{c}[5]}{32} - \frac{\sqrt{555} \mathbf{c}[7]}{32} \) |
| \( Q_{8y}^{\alpha 2} \) | \( \frac{\sqrt{75} \mathbf{c}[1]}{32} + \frac{5 \sqrt{75} \mathbf{c}[3]}{32} + \frac{3 \sqrt{75} \mathbf{c}[5]}{32} - \frac{\sqrt{555} \mathbf{c}[7]}{32} \) |
| \( Q_{8z}^{\alpha 2} \) | \( S[4] \) |
| \( T_{2g} \) \( Q_{8x}^{\beta 1} \) | \( -\frac{\sqrt{855} \mathbf{c}[1]}{64} + \frac{\sqrt{910} \mathbf{c}[3]}{64} + \frac{7 \sqrt{725} \mathbf{c}[5]}{64} + \frac{3 \sqrt{305} \mathbf{c}[7]}{64} \) |
| \( Q_{8y}^{\beta 1} \) | \( -\frac{\sqrt{855} \mathbf{c}[1]}{64} - \frac{\sqrt{910} \mathbf{c}[3]}{64} + \frac{7 \sqrt{725} \mathbf{c}[5]}{64} - \frac{3 \sqrt{305} \mathbf{c}[7]}{64} \) |
| \( Q_{8z}^{\beta 1} \) | \( S[6] \) |
| \( T_{2g} \) \( Q_{8x}^{\beta 2} \) | \( -\frac{\sqrt{705} \mathbf{c}[1]}{64} + \frac{3 \sqrt{665} \mathbf{c}[3]}{64} - \frac{\sqrt{1430} \mathbf{c}[5]}{64} + \frac{\sqrt{2005} \mathbf{c}[7]}{64} \) |
| \( Q_{8y}^{\beta 2} \) | \( -\frac{\sqrt{705} \mathbf{c}[1]}{64} - \frac{3 \sqrt{665} \mathbf{c}[3]}{64} + \frac{\sqrt{1430} \mathbf{c}[5]}{64} - \frac{\sqrt{2005} \mathbf{c}[7]}{64} \) |
| \( Q_{8z}^{\beta 2} \) | \( S[2] \) |
### rank 9

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1u}$ | $Q_9$ | $\frac{\sqrt{102}}{12} - \frac{\sqrt{42}}{12}$ |
| $A_{2u}$ | $Q'_9$ | $\frac{\sqrt{3}}{4} - \frac{\sqrt{3}}{4}$ |
| $E_u$ | $Q_{9u}$ | $\frac{\sqrt{102}}{12} + \frac{\sqrt{42}}{12}$ |
| $Q_{9v}$ | | $\frac{\sqrt{102}}{12} + \frac{\sqrt{42}}{12}$ |
| $T_{1u}$ | $Q^{a1}_{9x}$ | $21 \sqrt{2310C[1]} - 3 \sqrt{1430C[7]} + 3 \sqrt{24310C[9]}$ |
| $Q^{a1}_{9y}$ | | $21 \sqrt{2310S[3]} - 3 \sqrt{1430S[7]} + 3 \sqrt{24310S[9]}$ |
| $Q^{a1}_{9z}$ | | $C[0]$ |
| $T_{1u}$ | $Q^{a2}_{9x}$ | $21 \sqrt{2310C[1]} + 8 \sqrt{2310S[3]} - 7 \sqrt{510C[7]} + 3 \sqrt{24310C[9]}$ |
| $Q^{a2}_{9y}$ | | $21 \sqrt{2310S[3]} + 8 \sqrt{2310C[1]} - 7 \sqrt{510S[7]} + 3 \sqrt{24310S[9]}$ |
| $Q^{a2}_{9z}$ | | $C[8]$ |
| $T_{1u}$ | $Q^{a3}_{9x}$ | $21 \sqrt{1001C[1]} - 3 \sqrt{170C[5]} + 23 \sqrt{24310C[9]}$ |
| $Q^{a3}_{9y}$ | | $21 \sqrt{1001S[3]} - 3 \sqrt{170S[5]} + 23 \sqrt{24310S[9]}$ |
| $Q^{a3}_{9z}$ | | $C[4]$ |
| $T_{2u}$ | $Q^{b1}_{9x}$ | $21 \sqrt{858C[1]} + 8 \sqrt{2310C[1]} - 7 \sqrt{510C[7]} - 3 \sqrt{24310C[9]}$ |
| $Q^{b1}_{9y}$ | | $21 \sqrt{858S[3]} + 8 \sqrt{2310S[3]} - 7 \sqrt{510S[7]} - 3 \sqrt{24310S[9]}$ |
| $Q^{b1}_{9z}$ | | $C[6]$ |
| $T_{2u}$ | $Q^{b2}_{9x}$ | $21 \sqrt{858C[1]} - 8 \sqrt{2310C[1]} - 7 \sqrt{510C[7]} - 3 \sqrt{24310C[9]}$ |
| $Q^{b2}_{9y}$ | | $21 \sqrt{858S[3]} - 8 \sqrt{2310S[3]} - 7 \sqrt{510S[7]} - 3 \sqrt{24310S[9]}$ |
| $Q^{b2}_{9z}$ | | $C[2]$ |
### Rank 10

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1g}$ | $Q_{10}$ | $\sqrt{390}\tau_{10} - \sqrt{72}\tau_{14} - \sqrt{1125}\tau_{8}$ |
| $A_{2g}$ | $Q_{10}^a$ | $\sqrt{85}\tau_{10}$ + $\sqrt{1482}\tau_{12}$ + $\sqrt{57}\tau_{6}$ |
| $E_g$ | $Q_{10a}$ | $11\sqrt{20189}\tau_{10}$ + $\sqrt{827645}\tau_{14}$ + $\sqrt{146055}\tau_{8}$ |
| $E_g$ | $Q_{10r}$ | $\sqrt{10099}\tau_{10}$ + $\sqrt{190095}\tau_{12}$ |
| $T_{1g}$ | $Q_{10x}^{a1}$ | $\sqrt{2225}\tau_{1} - \sqrt{1025}\tau_{3} - \sqrt{1015}\tau_{5} - 11\sqrt{55}\tau_{7} - \sqrt{355}\tau_{9}$ |
| $T_{1g}$ | $Q_{10y}^{a1}$ | $\sqrt{2225}\tau_{1} - \sqrt{1025}\tau_{3} + \sqrt{1015}\tau_{5} - 11\sqrt{55}\tau_{7} - \sqrt{355}\tau_{9}$ |
| $T_{1g}$ | $Q_{10z}^{a1}$ | $\sqrt{2225}\tau_{1} - \sqrt{1025}\tau_{3} + \sqrt{1015}\tau_{5} - 11\sqrt{55}\tau_{7} - \sqrt{355}\tau_{9}$ |
| $T_{2g}$ | $Q_{10x}^{b1}$ | $\sqrt{1990}\tau_{1} + \sqrt{4845}\tau_{3} + \sqrt{969}\tau_{5} + \sqrt{285}\tau_{7} + \sqrt{55}\tau_{9}$ |
| $T_{2g}$ | $Q_{10y}^{b1}$ | $\sqrt{1990}\tau_{1} + \sqrt{4845}\tau_{3} + \sqrt{969}\tau_{5} + \sqrt{285}\tau_{7} + \sqrt{55}\tau_{9}$ |
| $T_{2g}$ | $Q_{10z}^{b1}$ | $\sqrt{1990}\tau_{1} + \sqrt{4845}\tau_{3} + \sqrt{969}\tau_{5} + \sqrt{285}\tau_{7} + \sqrt{55}\tau_{9}$ |
| $T_{2g}$ | $Q_{10x}^{b2}$ | $9\sqrt{78}\tau_{1} + 69\tau_{3} - \sqrt{55}\tau_{5} + 43\sqrt{17}\tau_{7} + \sqrt{355}\tau_{9}$ |
| $T_{2g}$ | $Q_{10y}^{b2}$ | $9\sqrt{78}\tau_{1} + 69\tau_{3} - \sqrt{55}\tau_{5} + 43\sqrt{17}\tau_{7} + \sqrt{355}\tau_{9}$ |
| $T_{2g}$ | $Q_{10z}^{b2}$ | $9\sqrt{78}\tau_{1} + 69\tau_{3} - \sqrt{55}\tau_{5} + 43\sqrt{17}\tau_{7} + \sqrt{355}\tau_{9}$ |
| $T_{2g}$ | $Q_{10x}^{b3}$ | $7\sqrt{5}\tau_{1} + 7\sqrt{5}\tau_{3} + 5\sqrt{13}\tau_{5} + 7\sqrt{142}\tau_{7} + \sqrt{2519}\tau_{9}$ |
| $T_{2g}$ | $Q_{10y}^{b3}$ | $7\sqrt{5}\tau_{1} + 7\sqrt{5}\tau_{3} + 5\sqrt{13}\tau_{5} + 7\sqrt{142}\tau_{7} + \sqrt{2519}\tau_{9}$ |
| $T_{2g}$ | $Q_{10z}^{b3}$ | $7\sqrt{5}\tau_{1} + 7\sqrt{5}\tau_{3} + 5\sqrt{13}\tau_{5} + 7\sqrt{142}\tau_{7} + \sqrt{2519}\tau_{9}$ |
| $Q_{10c}$ | | $S[2]$ |
### irrep. symbol definition

| irrep. | symbol | definition |
|--------|--------|------------|
| \(A_{2u}\) | \(Q_{11}^g\) | \[\sqrt{798S[10]} \frac{48}{3} + \sqrt{255S[2]} \frac{24}{3} + 3 \sqrt{5S[6]} \frac{16}{3}\] |
| \(E_u\) | \(Q_{11u}^1\) | \(S[8]\) |
| \(E_u\) | \(Q_{11v}^2\) | \(S[4]\) |
| \(T_{1u}\) | \(Q_{11x}^{\text{el}}\) | \[-\sqrt{88179S[11]} 512 - 21 \sqrt{665S[1]} 512 - 15 \sqrt{135S[5]} 512 - 77 \sqrt{178S[7]} 512 - 39 \sqrt{13S[9]} 512\] |
| \(T_{1u}\) | \(Q_{11y}^{\text{el}}\) | \[-\sqrt{88179S[11]} 512 - 21 \sqrt{665S[1]} 512 - 15 \sqrt{135S[5]} 512 - 77 \sqrt{178S[7]} 512 - 39 \sqrt{13S[9]} 512\] |
| \(T_{1u}\) | \(Q_{11z}^{\text{el}}\) | \(C[0]\) |
| \(T_{2u}\) | \(Q_{11x}^{\text{el}}\) | \[-\sqrt{213S[11]} 1024 - \sqrt{2939S[1]} 512 - 9 \sqrt{1615S[3]} 512 - 5 \sqrt{1356S[5]} 1024 - 7 \sqrt{1330S[7]} 1024 - 9 \sqrt{13S[9]} 1024\] |
| \(T_{2u}\) | \(Q_{11y}^{\text{el}}\) | \[-\sqrt{213S[11]} 1024 - \sqrt{2939S[1]} 512 - 9 \sqrt{1615S[3]} 512 - 5 \sqrt{1356S[5]} 1024 - 7 \sqrt{1330S[7]} 1024 - 9 \sqrt{13S[9]} 1024\] |
| \(T_{2u}\) | \(Q_{11z}^{\text{el}}\) | \(C[10]\) |
| \(T_{2u}\) | \(Q_{11x}^{\text{el}}\) | \[-\sqrt{213S[11]} 1024 - 15 \sqrt{277S[1]} 512 - 5 \sqrt{59S[3]} 512 - 105 \sqrt{70S[5]} 1024 - 61 \sqrt{11S[7]} 1024\] |
| \(T_{2u}\) | \(Q_{11y}^{\text{el}}\) | \[-\sqrt{213S[11]} 1024 - 15 \sqrt{277S[1]} 512 - 5 \sqrt{59S[3]} 512 - 105 \sqrt{70S[5]} 1024 - 61 \sqrt{11S[7]} 1024\] |
| \(T_{2u}\) | \(Q_{11z}^{\text{el}}\) | \(C[6]\) |

### C.2 Hexagonal multipoles up to rank 11

| irrep. | symbol | definition |
|--------|--------|------------|
| \(A_{1g}\) | \(Q_0\) | \(C[0]\) |

### irrep. symbol definition

| irrep. | symbol | definition |
|--------|--------|------------|
| \(E_{1u}\) | \(Q_x\) | \(C[1]\) |
| \(Q_y\) | \(S[1]\) |
### Rank 2

| Irrep.  | Symbol | Definition |
|---------|--------|------------|
| $A_{1g}$ | $Q_{x}$ | $C[0]$ |
| $E_{1g}$ | $Q_{yz}$ | $S[1]$ |
|          | $Q_{zx}$ | $C[1]$ |
| $E_{2g}$ | $Q_{xy}$ | $S[2]$ |
|          | $Q_{y}$ | $C[2]$ |

### Rank 3

| Irrep.  | Symbol | Definition |
|---------|--------|------------|
| $A_{2u}$ | $Q_{3u}^{\alpha}$ | $C[0]$ |
| $B_{1u}$ | $Q_{3u}^{\gamma}$ | $S[3]$ |
| $B_{2u}$ | $Q_{3v}^{\gamma}$ | $C[3]$ |
| $E_{1u}$ | $Q_{3u}^{\alpha}$ | $C[1]$ |
|          | $Q_{3v}^{\alpha}$ | $S[1]$ |
| $E_{2u}$ | $Q_{3u}^{\beta}$ | $C[2]$ |
|          | $Q_{3v}^{\beta}$ | $S[2]$ |

### Rank 4

| Irrep.  | Symbol | Definition |
|---------|--------|------------|
| $A_{1g}$ | $Q_{4}^{\alpha}$ | $C[0]$ |
| $B_{1g}$ | $Q_{4}^{\beta}$ | $C[3]$ |
| $B_{2g}$ | $Q_{4u}^{\beta}$ | $S[3]$ |
| $E_{1g}$ | $Q_{4u}^{\alpha}$ | $S[1]$ |
|          | $Q_{4v}^{\alpha}$ | $C[1]$ |
| $E_{2g}$ | $Q_{4u}^{\beta}$ | $S[4]$ |
|          | $Q_{4v}^{\beta}$ | $C[4]$ |
| $E_{2g}$ | $Q_{4u}^{\gamma}$ | $S[2]$ |
|          | $Q_{4v}^{\gamma}$ | $C[2]$ |

### Rank 5

| Irrep.  | Symbol | Definition |
|---------|--------|------------|
| $A_{2u}$ | $Q_{5}^{\alpha}$ | $C[0]$ |
| $B_{1u}$ | $Q_{5}^{\beta}$ | $S[3]$ |
| $B_{2u}$ | $Q_{5u}^{\beta}$ | $C[3]$ |
| $E_{1u}$ | $Q_{5u}^{\alpha}$ | $S[5]$ |
|          | $Q_{5v}^{\alpha}$ | $C[5]$ |
| $E_{1u}$ | $Q_{5u}^{\gamma}$ | $C[1]$ |
|          | $Q_{5v}^{\gamma}$ | $S[5]$ |
| $E_{2u}$ | $Q_{5u}^{\delta}$ | $C[4]$ |
|          | $Q_{5v}^{\delta}$ | $S[4]$ |
| $E_{2u}$ | $Q_{5u}^{\epsilon}$ | $S[2]$ |
|          | $Q_{5v}^{\epsilon}$ | $C[2]$ |
### rank 6

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1g}$ | $Q_{6}^1$ | $C[0]$ |
| $A_{1g}$ | $Q_{6}^2$ | $C[6]$ |
| $A_{2g}$ | $Q_{6}^{\beta}$ | $S[6]$ |
| $B_{1g}$ | $Q_{6}^{\gamma}$ | $C[3]$ |
| $B_{2g}$ | $Q_{6}^{\delta}$ | $S[3]$ |
| $E_{1g}$ | $Q_{6u}^{\alpha_1}$ | $S[5]$ |
|        | $Q_{6v}^{\alpha_1}$ | $C[5]$ |
| $E_{1g}$ | $Q_{6u}^{\alpha_2}$ | $S[1]$ |
|        | $Q_{6v}^{\alpha_2}$ | $C[1]$ |
| $E_{2g}$ | $Q_{6u}^{\beta_1}$ | $S[4]$ |
|        | $Q_{6v}^{\beta_1}$ | $C[4]$ |
| $E_{2g}$ | $Q_{6u}^{\beta_2}$ | $S[2]$ |
|        | $Q_{6v}^{\beta_2}$ | $C[2]$ |

### rank 7

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1u}$ | $Q_{7}^{1}$ | $S[6]$ |
| $A_{2u}$ | $Q_{7}^{2}$ | $C[0]$ |
| $A_{2u}$ | $Q_{7}^{2}$ | $C[6]$ |
| $B_{1u}$ | $Q_{7}^{\gamma}$ | $S[3]$ |
| $B_{2u}$ | $Q_{7}^{\delta}$ | $C[3]$ |
| $E_{1u}$ | $Q_{7u}^{\alpha_1}$ | $C[7]$ |
|        | $Q_{7v}^{\alpha_1}$ | $S[7]$ |
| $E_{1u}$ | $Q_{7u}^{\alpha_2}$ | $C[5]$ |
|        | $Q_{7v}^{\alpha_2}$ | $S[5]$ |
| $E_{1u}$ | $Q_{7u}^{\alpha_3}$ | $C[1]$ |
|        | $Q_{7v}^{\alpha_3}$ | $S[1]$ |
| $E_{2u}$ | $Q_{7u}^{\beta_1}$ | $C[4]$ |
|        | $Q_{7v}^{\beta_1}$ | $S[4]$ |
| $E_{2u}$ | $Q_{7u}^{\beta_2}$ | $C[2]$ |
|        | $Q_{7v}^{\beta_2}$ | $S[2]$ |
### rank 8

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1g}$ | $Q_8^1$ | $C[0]$ |
| $A_{1g}$ | $Q_8^2$ | $C[6]$ |
| $A_{2g}$ | $Q_8^8$ | $S[6]$ |
| $B_{1g}$ | $Q_8^y$ | $C[3]$ |
| $B_{2g}$ | $Q_8^s$ | $S[3]$ |
| $E_{1g}$ | $Q_{8u}^{a1}$ | $S[7]$ |
|          | $Q_{8v}^{a1}$ | $C[7]$ |
| $E_{1g}$ | $Q_{8u}^{a2}$ | $S[5]$ |
|          | $Q_{8v}^{a2}$ | $C[5]$ |
| $E_{1g}$ | $Q_{8u}^{a3}$ | $S[1]$ |
|          | $Q_{8v}^{a3}$ | $C[1]$ |
| $E_{2g}$ | $Q_{8u}^{b1}$ | $C[8]$ |
|          | $Q_{8v}^{b1}$ | $S[8]$ |
| $E_{2g}$ | $Q_{8u}^{b2}$ | $C[4]$ |
|          | $Q_{8v}^{b2}$ | $S[4]$ |
| $E_{2g}$ | $Q_{8u}^{b3}$ | $C[2]$ |
|          | $Q_{8v}^{b3}$ | $S[2]$ |

### rank 9

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1u}$ | $Q_9$ | $S[6]$ |
| $A_{2u}$ | $Q_9^a$ | $C[0]$ |
| $A_{2u}$ | $Q_9^b$ | $C[6]$ |
| $B_{1u}$ | $Q_9^c$ | $S[9]$ |
| $B_{1u}$ | $Q_9^2$ | $S[3]$ |
| $B_{2u}$ | $Q_9^1$ | $C[9]$ |
| $B_{2u}$ | $Q_9^2$ | $C[3]$ |
| $E_{1u}$ | $Q_{9u}^{a1}$ | $C[7]$ |
|          | $Q_{9v}^{a1}$ | $S[7]$ |
| $E_{1u}$ | $Q_{9u}^{a2}$ | $C[5]$ |
|          | $Q_{9v}^{a2}$ | $S[5]$ |
| $E_{1u}$ | $Q_{9u}^{a3}$ | $C[1]$ |
|          | $Q_{9v}^{a3}$ | $S[1]$ |
| $E_{2u}$ | $Q_{9u}^{b1}$ | $C[8]$ |
|          | $Q_{9v}^{b1}$ | $S[8]$ |
| $E_{2u}$ | $Q_{9u}^{b2}$ | $C[4]$ |
|          | $Q_{9v}^{b2}$ | $S[4]$ |
| $E_{2u}$ | $Q_{9u}^{b3}$ | $C[2]$ |
|          | $Q_{9v}^{b3}$ | $S[2]$ |
### rank 10

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1g}$ | $Q_{10}^{1}$ | C[0] |
| $A_{1g}$ | $Q_{10}^{2}$ | C[6] |
| $A_{2g}$ | $Q_{10}^{3}$ | S[6] |
| $B_{1g}$ | $Q_{10}^{4}$ | C[9] |
| $B_{1g}$ | $Q_{10}^{5}$ | C[3] |
| $B_{2g}$ | $Q_{10}^{6}$ | S[9] |
| $B_{2g}$ | $Q_{10}^{7}$ | S[3] |
| $E_{1g}$ | $Q_{10u}^{8}$ | S[7] |
|           | $Q_{10v}^{8}$ | C[7] |
| $E_{1g}$ | $Q_{10u}^{9}$ | S[5] |
|           | $Q_{10v}^{9}$ | C[5] |
| $E_{1g}$ | $Q_{10u}^{10}$ | S[1] |
|           | $Q_{10v}^{10}$ | C[1] |
| $E_{2g}$ | $Q_{10u}^{11}$ | S[10] |
|           | $Q_{10v}^{11}$ | C[10] |
| $E_{2g}$ | $Q_{10u}^{12}$ | S[8] |
|           | $Q_{10v}^{12}$ | C[8] |
| $E_{2g}$ | $Q_{10u}^{13}$ | S[4] |
|           | $Q_{10v}^{13}$ | C[4] |
| $E_{2g}$ | $Q_{10u}^{14}$ | S[2] |
|           | $Q_{10v}^{14}$ | C[2] |

### rank 11

| irrep. | symbol | definition |
|--------|--------|------------|
| $A_{1u}$ | $Q_{11}^{1}$ | S[6] |
| $A_{2u}$ | $Q_{11}^{3}$ | C[0] |
| $A_{2u}$ | $Q_{11}^{4}$ | C[6] |
| $B_{1u}$ | $Q_{11}^{5}$ | S[9] |
| $B_{1u}$ | $Q_{11}^{6}$ | S[3] |
| $B_{2u}$ | $Q_{11}^{7}$ | C[9] |
| $B_{2u}$ | $Q_{11}^{8}$ | C[3] |
| $E_{1u}$ | $Q_{11u}^{9}$ | C[11] |
| $E_{1u}$ | $Q_{11v}^{9}$ | C[11] |
| $E_{1u}$ | $Q_{11u}^{10}$ | C[7] |
| $E_{1u}$ | $Q_{11v}^{10}$ | S[7] |
| $E_{1u}$ | $Q_{11u}^{11}$ | C[5] |
| $E_{1u}$ | $Q_{11v}^{11}$ | S[5] |
| $E_{1u}$ | $Q_{11u}^{12}$ | C[1] |
| $E_{1u}$ | $Q_{11v}^{12}$ | S[1] |
| $E_{2u}$ | $Q_{11u}^{13}$ | C[10] |
| $E_{2u}$ | $Q_{11v}^{13}$ | S[10] |
| $E_{2u}$ | $Q_{11u}^{14}$ | C[8] |
| $E_{2u}$ | $Q_{11v}^{14}$ | S[8] |
| $E_{2u}$ | $Q_{11u}^{15}$ | C[4] |
| $E_{2u}$ | $Q_{11v}^{15}$ | S[4] |
| $E_{2u}$ | $Q_{11u}^{16}$ | C[2] |
| $E_{2u}$ | $Q_{11v}^{16}$ | S[2] |