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Editors' Suggestion

Classification of atomic-scale multipoles under crystallographic point groups and application to linear response tensors

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Four types of atomic-scale multipoles (electric, magnetic, magnetic toroidal, and electric toroidal multipoles) give a complete set to describe arbitrary degrees of freedom for coupled charge, spin, and orbital of electrons. We here present a systematic classification of these multipole degrees of freedom towards the application in condensed matter physics. Starting from the multipole description under the rotation group in real space, we generalize the concept of multipoles in momentum space with the spin degree of freedom. We show how multipoles affect the electronic band structure and linear responses, such as the magnetoelectric effect, magnetocurrent (magnetogyrotropic) effect, spin conductivity, piezoelectric effect, and so on. Moreover, we exhibit a complete table to represent the active multipoles under 32 crystallographic point groups. Our comprehensive and systematic analyses will give a foundation to identify enigmatic electronic order parameters and a guide to evaluate peculiar cross-correlated phenomena in condensed matter physics from the microscopic point of view.

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I. INTRODUCTION

The multipole moments characterize electric charge and current distributions, whose concept has been widely developed in various fields of physics at different length scales, such as classical electromagnetism [1–4], nuclear physics [5–8], solid-state physics [9–13], and metamaterials [14–18]. It was well-known that there are four types of fundamental multipoles according to their spatial inversion and time-reversal properties [4,19–21]: electric (E: polar/true tensor with time-reversal even), magnetic (M: axial/pseudotensor with time-reversal odd), magnetic toroidal (MT: polar/true tensor with time-reversal odd), and electric toroidal (ET: axial/pseudotensor with time-reversal even) multipoles. A recent study has shown that the four fundamental multipoles in atomic-scale constitute a complete set to span the Hilbert space under the space-time inversion group. They can be applied to not only a classical but also a quantum-mechanical picture [21]. The mutual relationship between four multipoles and their schematic pictures in the quantum-mechanical representation are shown in Fig. 1.

In condensed matter physics, the multipoles have been recognized as important quantities to describe multiple degrees of freedom of electrons in solids, e.g., charge, spin, and orbital, in a unified way. Especially, the atomic-scale multipoles have been extensively studied in f-electron systems, as the interplay between the Coulomb interaction and spin-orbit coupling due to its strongly localized nature gives rise to anisotropic charge and spin distributions [9,11,12]. In fact, higher-rank multipole orders beyond the conventional E and M dipole orders have been established by mutual interplay between experimental and theoretical investigations, for instance, E quadrupole in PrT_2X_{20}(T = Ir, Rh, V, Ti, Nb and X = Al, Zn) [22–30] and M octupole in $Ce_{1-x}La_xB_6$ [31–34].

On the other hand, such a multipole concept can be applied to a cluster consisting of several atomic sites, which is so-called a cluster multipole [35–46]. The complex electric and magnetic orderings in cluster are regarded as the ferroic arrangement of higher-order cluster multipoles from the symmetry aspect. For example, the all-in/all-out ordering of magnetic moments on the pyrochlore structure is regarded as an M octupole order (precisely speaking, M pseudoscalar in the point group) [47], while a staggered antiferromagnetic ordering on the zigzag chain is regarded as a ferroic order of MT dipole and M quadrupole [35,39,48]. More recently, the concept of multipoles is extended to multiple hybrid orbitals [21], and to the momentum space relevant with topologically nontrivial excitations through the Berry curvature [49–53].

In this way, studies of the multipoles are useful to cover various unconventional order parameters in a systematic manner and understand/expect physical phenomena from the symmetry viewpoint. For example, the nematic order in iron-based superconductors [54–59] and magnetic insulators [60–63], Pomeranchuk instability [64–69], anisotropic density wave including staggered flux phases [70–76], excitonic insulators [77–81], and spin chirality accompanying Berry phase [82–84] are also described by multipole terminology. It is advantageous for the multipole description that once a type of the multipoles is identified, it is easy to understand/predict how the band structure is deformed and what types of cross-correlated couplings and transport phenomena occur. For example, the magnetoelectric effects in

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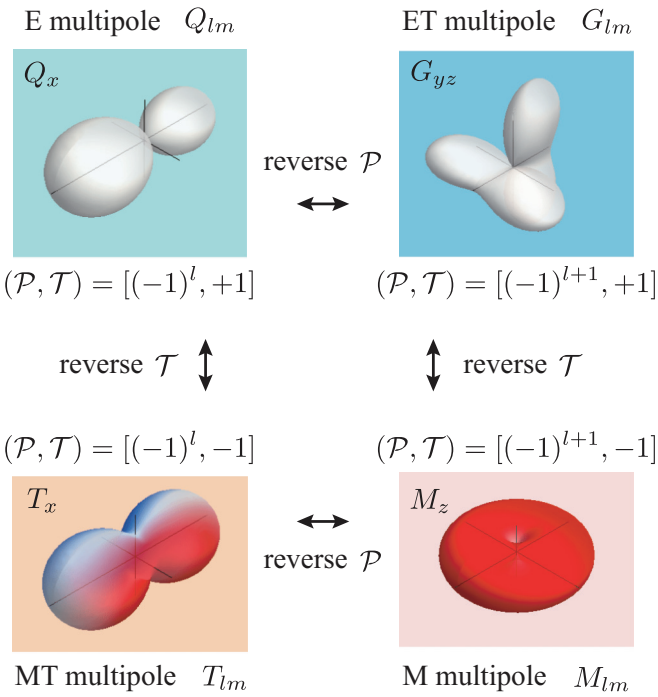

FIG. 1. Four types of multipoles under the space-time inversion group. The E (M) multipole transforms the MT (ET) multipole by reversing the time-reversal property, while the E (M) multipole transforms ET (MT) multipole by reversing the spatial-inversion property. Schematic pictures of the wave functions for the E dipole, M dipole, MT dipole, and ET quadrupole are shown in each panel. The shape and color map of the pictures represent the electric charge density and the z component of the orbital angular-momentum density, respectively.

Cr_2O_3 [85–87] and $Co_4Nb_2O_9$ [88–92] are understood by the M quadrupole, magnetocurrent effect in metallic UNi_4B by the MT dipole [93–95], valley splitting in the electronic band structure by the MT octupole [96], nonreciprocal magnon excitations in BiTeBr [97] and α-$Cu_2V_2O_7$ [98–102] by the E and MT dipoles, orbital Edelstein effect in Te by the ET monopole and quadrupole [103,104], and large anomalous Hall effect, Nernst effect, and Kerr rotation in Mn_3Sn by the M octupole [40,105–108]. Moreover, unconventional superconductivities are also discussed in the language of multipoles [109–112]. Since the microscopic quantum-mechanical definition of multipoles has been obtained recently [21], it enables us to describe a variety of these order parameters in a systematic way by means of the atomic-scale multipoles, and gives further understanding in cross-correlated responses.

In the present paper, we push forward these multipole studies for a comprehensive classification of multipoles in real and momentum spaces and to elucidate the physical properties brought by the multipole degrees of freedom. In order to deduce an unknown order parameter, a comprehensive classification is also very useful to narrow down a candidate. Starting from the multipole description under the rotation group in real space, we generalize the concept of multipoles in momentum space with the spin degree of freedom. We show how multipoles affect the electronic band structure and linear responses, such as the magnetoelectric and magnetocurrent (magnetogyrotropic) effects. Moreover, we demonstrate a complete table to display the active multipoles under 32 crystallographic point groups. Our systematic study will encourage a direction for material design based on multipole (electronic) degrees of freedom.

The organization of this paper is as follows. In Sec. II, we give a definition of four multipoles in both real and momentum spaces. In Sec. III, we show how the emergent multipoles affect the electronic band structure and linear responses including magnetoelectric and magnetocurrent (magnetogyrotropic) phenomena. After presenting the crystalline-electric-field potential under all the point groups, we clarify what types of multipoles are active and become potential order parameters under the point-group irreducible representation in Sec. IV. Section V is devoted to a summary of the present paper. In Appendix A, we show the atomic basis wave functions for s, p, d, and f orbitals. We discuss a possible extension of the momentum-space multipoles to multiorbital system in Appendix B. In Appendix C, we show transformation properties of linear response tensors under the spatial inversion and time-reversal operations. We present which multipoles are relevant with rank-3 linear response tensors, such as the spin conductivity tensor and piezoelectric(current) tensor in Appendix D. In Appendix E, we show the active multipoles for atomic basis functions for the cubic O_h and its subgroups. Appendix E contains various tables for the hexagonal D_{6h} and its subgroups to complete all the point groups. For completeness, we use here some of our results previously reported in Ref. [21].

II. DEFINITION OF MULTIPOLES

The concept of multipoles is useful to describe electronic degrees of freedom in a systematic way, and it is capable to express arbitrary type of order parameters in phase transition and physical responses to external fields at microscopic level [113]. As mentioned in Introduction, there are four types of multipoles, E (electric: Q_{lm}), M (magnetic: M_{lm}), MT (magnetic toroidal: T_{lm}), and ET (electric toroidal: G_{lm}), according to the time-reversal and spatial inversion properties [1,19,20].

In this section, we introduce microscopic expressions of four types of multipoles. First, we give the expressions in real space in Sec. II A, and then we give them in momentum space in Sec. II B. With these prescriptions, we summarize in Table III, which type of multipoles are activated in the atomic basis functions with the spin degree of freedom.

A. Multipoles in real space

Based on the standard multipole expansion of the electromagnetic potentials $\phi(\boldsymbol{r})$ and $\boldsymbol{A}(\boldsymbol{r})$ in the presence of the source electric current $\boldsymbol{j}_{\rm e}(\boldsymbol{r}) = c\nabla \times \boldsymbol{M}(\boldsymbol{r})$ and the magnetic current $\boldsymbol{j}_{\rm m}(\boldsymbol{r}) = c\nabla \times \boldsymbol{P}(\boldsymbol{r})$ ($\boldsymbol{M}$ and $\boldsymbol{P}$ are the magnetization and electric polarization) [114], and by inserting the quantum-mechanical expressions of $\boldsymbol{j}_{\rm e}$ and $\boldsymbol{j}_{\rm m}$ including spin contributions, two of the present authors have shown the quantum-mechanical expressions of multipoles as [21]

$$\hat{Q}_{lm} = -e\sum_j O_{lm}(\boldsymbol{r}_j), \tag{1}$$

$$\hat{M}_{lm} = -\mu_{\rm B}\sum_j \boldsymbol{m}_l(\boldsymbol{r}_j)\cdot\nabla O_{lm}(\boldsymbol{r}_j), \tag{2}$$

$$\hat{T}_{lm} = -\mu_{\rm B} \sum_j \boldsymbol{t}_l(\boldsymbol{r}_j) \cdot \boldsymbol{\nabla} O_{lm}(\boldsymbol{r}_j), \tag{3}$$

$$\hat{G}_{lm} = -e \sum_j \sum_{\alpha\beta}^{x,y,z} g_l^{\alpha\beta} \nabla_\alpha \nabla_\beta O_{lm}(\boldsymbol{r}_j), \tag{4}$$

where $\boldsymbol{m}_l$, $\boldsymbol{t}_l$, and $g_l^{\alpha\beta}$ are defined in terms of the dimensionless orbital and spin angular-momentum operators $\boldsymbol{l}_j$ and $\boldsymbol{\sigma}_j/2$ of an electron at $\boldsymbol{r}_j$ as

$$\boldsymbol{m}_l(\boldsymbol{r}_j) = \frac{2\boldsymbol{l}_j}{l+1} + \boldsymbol{\sigma}_j, \tag{5}$$

$$\boldsymbol{t}_l(\boldsymbol{r}_j) = \frac{\boldsymbol{r}_j}{l+1} \times \left(\frac{2\boldsymbol{l}_j}{l+2} + \boldsymbol{\sigma}_j \right), \tag{6}$$

$$g_l^{\alpha\beta}(\boldsymbol{r}_j) = m_l^\alpha(\boldsymbol{r}_j) t_l^\beta(\boldsymbol{r}_j). \tag{7}$$

We have introduced

$$O_{lm}(\boldsymbol{r}) = \sqrt{\frac{4\pi}{2l+1}} r^l Y_{lm}^*(\hat{\boldsymbol{r}}), \tag{8}$$

where $Y_{lm}(\hat{\boldsymbol{r}})$ are the spherical harmonics as a function of angles $\hat{\boldsymbol{r}} = \boldsymbol{r}/r$ [8,9] with the azimuthal and magnetic quantum numbers, l and m ($-l \leqslant m \leqslant l$). We adopt the phase convention so as to satisfy $Y_{lm}(\hat{\boldsymbol{r}}) = (-1)^m Y_{l-m}^*(\hat{\boldsymbol{r}})$. The index l represents the rank of multipoles: $l = 0$ (monopole), 1 (dipole), 2 (quadrupole), 3 (octupole), 4 (hexadecapole), and so on. Strictly speaking, the rank is meaningful as the irreducible representation only if rotational symmetry is preserved, otherwise, the same irreducible representations appear in different ranks in general, and they mix with each other. Hereafter, we omit "elementary charge" for multipoles, i.e., $-e$ and $-\mu_{\rm B}$, for notational simplicity.

In the rotation group, arbitrary linear combinations within the same rank can be taken as an irreducible basis set. It is often useful to introduce the tesseral harmonics $O_{lm}^{(\rm c)}(\boldsymbol{r})$ and $O_{lm}^{(\rm s)}(\boldsymbol{r})$, which correspond to the real expressions for the spherical harmonics [115]:

$$O_{l0}^{(\rm c)}(\boldsymbol{r}) \equiv O_{l0}(\boldsymbol{r}), \tag{9}$$

$$O_{lm}^{(\rm c)}(\boldsymbol{r}) \equiv \frac{(-1)^m}{\sqrt{2}} [O_{lm}(\boldsymbol{r}) + O_{lm}^*(\boldsymbol{r})], \tag{10}$$

$$O_{lm}^{(\rm s)}(\boldsymbol{r}) \equiv \frac{(-1)^m}{\sqrt{2}i} [O_{lm}(\boldsymbol{r}) - O_{lm}^*(\boldsymbol{r})], \tag{11}$$

for $m = 1, 2, \ldots, l$. Since the multipole operator $\hat{X}_{lm}$ ($X = Q, M, T, G$) transforms like $O_{lm}(\boldsymbol{r})$ by the rotational operation, similar linear combinations are applied to $\hat{X}_{lm}$ as well. In what follows, the symbols O_{lm} and $\hat{X}_{lm}$ are used to represent the set of harmonics in the real expression, either the tesseral harmonics or cubic harmonics, or hexagonal harmonics, and so on, with rank l. For later convenience, the even- and odd-parity cubic harmonics in $O_{\rm h}$ group are summarized in Tables I and II, respectively. The hexagonal harmonics in the $D_{6\rm h}$ group are also summarized in Tables XXII and XXIII in Appendix F.

These four multipole operators are clearly independent with each other under the space-time inversion operations as shown in Fig. 1. This is readily confirmed by using the facts that $O_{lm}(\boldsymbol{r})$ has parity $(-1)^l$ under the spatial inversion, and $\boldsymbol{l}$ and $\boldsymbol{\sigma}$ are odd under the time-reversal operation. $\hat{Q}_{lm}$ and $\hat{T}_{lm}$ in Eqs. (1) and (3) (see also the left panels in Fig. 1) represent the polar E and MT multipole operators, while $\hat{M}_{lm}$ and $\hat{G}_{lm}$ in Eqs. (2) and (4) (see also the right panels in Fig. 1) represent the axial M and ET multipole operators. Note that M monopole $\hat{M}_0 \equiv \hat{M}_{00}$, MT monopole $\hat{T}_0 \equiv \hat{T}_{00}$, ET monopole $\hat{G}_0 \equiv \hat{G}_{00}$, and ET dipole $\hat{G}_{1m} \equiv \hat{\boldsymbol{G}} = (\hat{G}_x, \hat{G}_y, \hat{G}_z)$ vanish in Eqs. (2)–(4) owing to the identity $\boldsymbol{\nabla} O_{00}(\boldsymbol{r}) = 0$ and $\nabla_\alpha \nabla_\beta O_{1m}(\boldsymbol{r}) = 0$.

In the spinless basis functions under the rotation group, (L, M) [$L = 0(s), 1(p), 2(d), 3(f)$ and $M = -L, -L+1, \ldots, L$] shown in Appendix A, the multipoles given by Eqs. (1)–(4) without spin contributions constitute a complete set to express an arbitrary degree of freedom [21]. In other words, the even-rank E and odd-rank M multipoles are active in nonhybrid (intra) orbitals, such as d-d and f-f orbitals. Meanwhile, the odd(even)-rank E and MT and even(odd)-rank M and ET multipoles are active in odd(even)-parity hybrid (inter) orbitals, such as p-d (s-d) and d-f (p-f) orbitals [21]. The total number of these independent active multipoles equals to the number of matrix elements in the relevant Hilbert space.

Since $\hat{X}_{lm}$ transforms like $O_{lm}(\boldsymbol{r})$ by the rotational operation, the matrix elements of four multipoles are related with each other. According to the Wigner-Eckart theorem, the matrix elements can be divided into the purely angular part and common part as

$$\langle L_1 M_1 | \hat{X}_{lm} | L_2 M_2 \rangle = \langle L_1 || \hat{X}_l || L_2 \rangle \langle L_1 M_1 | L_2 M_2; lm \rangle, \tag{12}$$

where $\langle L_1 || \hat{X}_l || L_2 \rangle$ and $\langle L_1 M_1 | L_2 M_2; lm \rangle$ are the reduced matrix element independent of M_1 and M_2, and the Clebsch-Gordan coefficient, respectively. Note that the reduced matrix elements for $\hat{Q}_{lm}$ and $\hat{T}_{lm}$ vanish when those for $\hat{M}_{lm}$ and $\hat{G}_{lm}$ are nonzero, and vice versa (see Table III, for example). Therefore the matrix elements for $\hat{T}_{lm}$ are proportional to those of $\hat{Q}_{lm}$ and the proportional coefficient, $R_l(L_1, L_2)$, is independent of M_1 and M_2, i.e.,

$$\langle L_1 M_1 | \hat{T}_{lm} | L_2 M_2 \rangle = R_l(L_1, L_2) \langle L_1 M_1 | \hat{Q}_{lm} | L_2 M_2 \rangle. \tag{13}$$

Similar proportionality holds between $\hat{G}_{lm}$ and $\hat{M}_{lm}$ as

$$\langle L_1 M_1 | \hat{G}_{lm} | L_2 M_2 \rangle = R_l(L_1, L_2) \langle L_1 M_1 | \hat{M}_{lm} | L_2 M_2 \rangle. \tag{14}$$

The proportional coefficients turn out to be common as

$$R_l(L_1, L_2) = \frac{\langle L_1 || \hat{T}_l || L_2 \rangle}{\langle L_1 || \hat{Q}_l || L_2 \rangle} = \frac{\langle L_1 || \hat{G}_l || L_2 \rangle}{\langle L_1 || \hat{M}_l || L_2 \rangle} = \frac{L_1(L_1+1) - L_2(L_2+1)}{(l+1)(l+2)} i. \tag{15}$$

The result is consistent with the fact that the toroidal multipoles, $\hat{T}_{lm}$ and $\hat{G}_{lm}$, vanish for nonhybrid orbitals, i.e., $L_1 = L_2$. For the spinful basis, as discussed below, a similar proportionality also holds, in which the spinless basis (L, M) is replaced by a spinful one (J, J_z). In other words, we can define arbitrary type of MT and ET multipoles in the total (orbital) angular-momentum basis through the proportionality, and it is sufficient to calculate the matrix elements of the E and M multipoles.

Next, let us extend to the situation where the total angular-momentum basis (J, J_z) is appropriate rather than (L, M) by

TABLE I. Even-parity cubic harmonics (E multipoles in unit of $-e$ in the cubic $O_{\rm h}$ group). The linear combinations of the tesseral harmonics are shown where (lm) and $(lm)'$ correspond to $O_{lm}^{(\rm c)}(\boldsymbol{r})$ and $O_{lm}^{(\rm s)}(\boldsymbol{r})$, respectively. In the irreducible representation (irrep.), the subscript represents the spatial parity (even:g, odd:u), while the superscript represents the time-reversal property (even: +, odd: −). A_1, A_2, E, T_1, and T_2 correspond to Γ_1, Γ_2, Γ_3, Γ_4, and Γ_5, respectively, in the Bethe notation.

rank	irrep.	symbol	definition	linear combination
0	A_{1g}^{+}	Q_0	1	(00)
2	E_g^{+}	Q_u, Q_v	$\frac{1}{2}(3z^2-r^2)$, $\frac{\sqrt{3}}{2}(x^2-y^2)$	(20), (22)
	T_{2g}^{+}	Q_{yz}, Q_{zx}, Q_{xy}	$\sqrt{3}yz$, $\sqrt{3}zx$, $\sqrt{3}xy$	$(21)'$, (21), $(22)'$
4	A_{1g}^{+}	Q_4	$\frac{5\sqrt{21}}{12}(x^4+y^4+z^4-\frac{3}{5}r^4)$	$(4)\equiv\frac{1}{2\sqrt{3}}[\sqrt{5}(44)+\sqrt{7}(40)]$
	E_g^{+}	Q_{4u}	$\frac{7\sqrt{15}}{6}[z^4-\frac{x^4+y^4}{2}-\frac{3}{7}r^2(3z^2-r^2)]$	$-\frac{1}{2\sqrt{3}}[\sqrt{7}(44)-\sqrt{5}(40)]$
		Q_{4v}	$\frac{7\sqrt{5}}{4}[x^4-y^4-\frac{6}{7}r^2(x^2-y^2)]$	$-(42)$
	T_{1g}^{+}	Q_{4x}^{α}	$\frac{\sqrt{35}}{2}yz(y^2-z^2)$	$-\frac{1}{2\sqrt{2}}[(43)'+\sqrt{7}(41)']$
		Q_{4y}^{α}	$\frac{\sqrt{35}}{2}zx(z^2-x^2)$	$-\frac{1}{2\sqrt{2}}[(43)-\sqrt{7}(41)]$
		Q_{4z}^{α}	$\frac{\sqrt{35}}{2}xy(x^2-y^2)$	$(44)'$
	T_{2g}^{+}	Q_{4x}^{β}	$\frac{\sqrt{5}}{2}yz(7x^2-r^2)$	$\frac{1}{2\sqrt{2}}[\sqrt{7}(43)'-(41)']$
		Q_{4y}^{β}	$\frac{\sqrt{5}}{2}zx(7y^2-r^2)$	$-\frac{1}{2\sqrt{2}}[\sqrt{7}(43)+(41)]$
		Q_{4z}^{β}	$\frac{\sqrt{5}}{2}xy(7z^2-r^2)$	$(42)'$
6	A_{1g}^{+}	Q_6	$\frac{231\sqrt{2}}{8}[x^2y^2z^2+\frac{r^2}{22}(x^4+y^4+z^4-\frac{3}{5}r^4)-\frac{r^6}{105}]$	$(6)\equiv\frac{1}{2\sqrt{2}}[-\sqrt{7}(64)+(60)]$
	A_{2g}^{+}	Q_{6t}	$\frac{\sqrt{2310}}{8}[x^4(y^2-z^2)+y^4(z^2-x^2)+z^4(x^2-y^2)]$	$(6t)\equiv\frac{1}{4}[-\sqrt{5}(66)+\sqrt{11}(62)]$
	E_g^{+}	Q_{6u}	$\frac{11\sqrt{14}}{4}[z^6-\frac{x^6+y^6}{2}-\frac{15}{11}r^2(z^4-\frac{x^4+y^4}{2})+\frac{5}{22}r^4(3z^2-r^2)]$	$\frac{1}{2\sqrt{2}}[(64)+\sqrt{7}(60)]$
		Q_{6v}	$\frac{11\sqrt{42}}{8}[x^6-y^6-\frac{15}{11}r^2(x^4-y^4)+\frac{5}{11}r^4(x^2-y^2)]$	$\frac{1}{4}[\sqrt{11}(66)+\sqrt{5}(62)]$
	T_{1g}^{+}	Q_{6x}^{α}	$\frac{3\sqrt{7}}{4}yz(y^2-z^2)(11x^2-r^2)$	$\frac{1}{8}[-\sqrt{22}(65)'-\sqrt{30}(63)'+2\sqrt{3}(61)']$
		Q_{6y}^{α}	$\frac{3\sqrt{7}}{4}zx(z^2-x^2)(11y^2-r^2)$	$\frac{1}{8}[\sqrt{22}(65)-\sqrt{30}(63)-2\sqrt{3}(61)]$
		Q_{6z}^{α}	$\frac{3\sqrt{7}}{4}xy(x^2-y^2)(11z^2-r^2)$	$(64)'$
	T_{2g}^{+}	$Q_{6x}^{\beta 1}$	$\frac{\sqrt{462}}{2}yz[y^4+z^4-\frac{5}{8}(y^2+z^2)^2]$	$\frac{1}{16}[\sqrt{3}(65)'+\sqrt{55}(63)'+3\sqrt{22}(61)']$
		$Q_{6y}^{\beta 1}$	$\frac{\sqrt{462}}{2}zx[z^4+x^4-\frac{5}{8}(z^2+x^2)^2]$	$\frac{1}{16}[\sqrt{3}(65)-\sqrt{55}(63)+3\sqrt{22}(61)]$
		$Q_{6z}^{\beta 1}$	$\frac{\sqrt{462}}{2}xy[x^4+y^4-\frac{5}{8}(x^2+y^2)^2]$	$(66)'$
	T_{2g}^{+}	$Q_{6x}^{\beta 2}$	$\frac{\sqrt{210}}{16}yz(33x^4-18x^2r^2+r^4)$	$\frac{1}{16}[\sqrt{165}(65)'-9(63)'+\sqrt{10}(61)']$
		$Q_{6y}^{\beta 2}$	$\frac{\sqrt{210}}{16}zx(33y^4-18y^2r^2+r^4)$	$\frac{1}{16}[\sqrt{165}(65)+9(63)+\sqrt{10}(61)]$
		$Q_{6z}^{\beta 2}$	$\frac{\sqrt{210}}{16}xy(33z^4-18z^2r^2+r^4)$	$(62)'$

taking into account the spin degree of freedom. In the one-electron state, the total angular momentum J is represented by $J=L-1/2$ or $L+1/2$ and $J_z=-J,-J+1,\ldots,J$. Similar to the argument for the spinless basis, the active multipoles are uniquely identified for the spinful basis as well from the following conditions: (1) the even-rank E multipoles and the odd-rank M multipoles are active in nonhybrid orbitals, such as s-s and p-p orbitals with the same total angular momentum J. (2) The even(odd)-rank E and MT multipoles and odd(even)-rank M and ET multipoles are active in the even(odd)-parity hybrid orbitals with different J or L. (3) The rank of active multipole l is determined by $|J_1-J_2|\leqslant l\leqslant J_1+J_2$, where J_1 and J_2 are total angular momenta in the bra- and ket-basis functions. For example, in p-d orbitals with the same total angular momentum $J=3/2$, the number of the independent active multipoles is 32 as indicated by the 11th row in Table III, which consist of M/ET monopole, E/MT dipole, M/ET quadrupole, and E/MT octupole without excess or deficiency. We summarize all the active multipoles in the spinful nonhybrid and hybrid orbitals in Table III, which will be useful to narrow down candidates of potential order parameters according to knowledge of relevant spin-orbital Hilbert space. In contrast to the cases for the spinless basis, $\hat{M}_0$, $\hat{G}_0$, and $\hat{\boldsymbol{G}}$ are active in the spinful basis.

The necessity of M and ET monopoles and ET dipole, which do not appear in the multipole expansion, implies that the corresponding quantum-mechanical operators $\hat{M}_0$, $\hat{G}_0$, and $\hat{\boldsymbol{G}}$ are definable in the spinful basis. To obtain explicit forms of these multipoles, we focus on the fact that the inner product between the M dipole $\hat{\boldsymbol{M}}=\boldsymbol{m}_1(\boldsymbol{r})$ and the position

TABLE II. Odd-parity cubic harmonics (E multipoles in unit of $-e$ in the cubic $O_{\rm h}$ group).

rank	irrep.	symbol	definition	linear combination
1	T_{1u}^{+}	$Q_x,\ Q_y,\ Q_z$	$x,\ y,\ z$	$(11),\ (11)',\ (10)$
3	A_{2u}^{+}	Q_{xyz}	$\sqrt{15}xyz$	$(32)'$
	T_{1u}^{+}	Q_x^{α}	$\frac{1}{2}x(5x^2-3r^2)$	$\frac{1}{2\sqrt{2}}[\sqrt{5}(33)-\sqrt{3}(31)]$
		Q_y^{α}	$\frac{1}{2}y(5y^2-3r^2)$	$-\frac{1}{2\sqrt{2}}[\sqrt{5}(33)'+\sqrt{3}(31)']$
		Q_z^{α}	$\frac{1}{2}z(5z^2-3r^2)$	(30)
	T_{2u}^{+}	Q_x^{β}	$\frac{\sqrt{15}}{2}x(y^2-z^2)$	$-\frac{1}{2\sqrt{2}}[\sqrt{3}(33)+\sqrt{5}(31)]$
		Q_y^{β}	$\frac{\sqrt{15}}{2}y(z^2-x^2)$	$\frac{1}{2\sqrt{2}}[-\sqrt{3}(33)'+\sqrt{5}(31)']$
		Q_z^{β}	$\frac{\sqrt{15}}{2}z(x^2-y^2)$	(32)
5	E_u^{+}	$Q_{5u},\ Q_{5v}$	$\frac{3\sqrt{35}}{2}xyz(x^2-y^2),\ -\frac{\sqrt{105}}{2}xyz(3z^2-r^2)$	$(54)',\ -(52)'$
	T_{1u}^{+}	$Q_{5x}^{\alpha 1}$	$\frac{x}{8}[8x^4-40x^2(y^2+z^2)+15(y^2+z^2)^2]$	$\frac{1}{8\sqrt{2}}[3\sqrt{7}(55)-\sqrt{35}(53)+\sqrt{30}(51)]$
		$Q_{5y}^{\alpha 1}$	$\frac{y}{8}[8y^4-40y^2(z^2+x^2)+15(z^2+x^2)^2]$	$\frac{1}{8\sqrt{2}}[3\sqrt{7}(55)'+\sqrt{35}(53)'+\sqrt{30}(51)']$
		$Q_{5z}^{\alpha 1}$	$\frac{z}{8}[8z^4-40z^2(x^2+y^2)+15(x^2+y^2)^2]$	(50)
	T_{1u}^{+}	$Q_{5x}^{\alpha 2}$	$\frac{3\sqrt{35}}{2}x[y^4+z^4-\frac{3}{4}(y^2+z^2)^2]$	$\frac{1}{16}[\sqrt{10}(55)+9\sqrt{2}(53)+2\sqrt{21}(51)]$
		$Q_{5y}^{\alpha 2}$	$\frac{3\sqrt{35}}{2}y[z^4+x^4-\frac{3}{4}(z^2+x^2)^2]$	$\frac{1}{16}[\sqrt{10}(55)'-9\sqrt{2}(53)'+2\sqrt{21}(51)']$
		$Q_{5z}^{\alpha 2}$	$\frac{3\sqrt{35}}{2}z[x^4+y^4-\frac{3}{4}(x^2+y^2)^2]$	(54)
	T_{2u}^{+}	Q_{5x}^{β}	$\frac{\sqrt{105}}{4}x(y^2-z^2)(3x^2-r^2)$	$\frac{1}{4\sqrt{2}}[-\sqrt{15}(55)-\sqrt{3}(53)+\sqrt{14}(51)]$
		Q_{5y}^{β}	$\frac{\sqrt{105}}{4}y(z^2-x^2)(3y^2-r^2)$	$\frac{1}{4\sqrt{2}}[\sqrt{15}(55)'-\sqrt{3}(53)'-\sqrt{14}(51)']$
		Q_{5z}^{β}	$\frac{\sqrt{105}}{4}z(x^2-y^2)(3z^2-r^2)$	(52)

vector $\boldsymbol{r}$ has the same symmetry as the M monopole in the rotation group. Thus we define the M monopole as

$$\hat{M}_0' \equiv \sum_j \boldsymbol{m}_1(\boldsymbol{r}_j)\cdot\boldsymbol{r}_j = \sum_j \boldsymbol{\sigma}_j\cdot\boldsymbol{r}_j, \tag{16}$$

where we have used $\boldsymbol{l}\cdot\boldsymbol{r}=0$, and the prime represents that it does not appear in the multipole expansion. $\hat{M}_0'$ in Eq. (16) clearly exhibits a property of a time-reversal-odd pseudoscalar and is valid only for the spinful basis. Similarly, by using the fact that the MT dipole $\hat{\boldsymbol{T}}=\boldsymbol{t}_1(\boldsymbol{r})$ has the same spatial

TABLE III. Active multipoles in nonhybrid (intra) and hybrid (inter) orbitals with the spin degree of freedom. The number of independent multipoles within the relevant Hilbert space is indicated in parentheses. The columns of $\mathcal{P}$ and l represent the spatial parity and the rank of the active multipoles, respectively. The column of "orbital" indicates the atomic orbital that consists of the basis.

basis	orbital	$\mathcal{P}$	$l=0$ (1)	1 (3)	2 (5)	3 (7)	4 (9)	5 (11)	6 (13)	7 (15)
1/2-1/2 (4)	s-s, p-p	+	E	M	–	–	–	–	–	–
3/2-3/2 (16)	p-p, d-d		E	M	E	M	–	–	–	–
5/2-5/2 (36)	d-d, f-f		E	M	E	M	E	M	–	–
7/2-7/2 (64)	f-f		E	M	E	M	E	M	E	M
1/2-3/2 (16)	s-d, p-p	+	–	M/ET	E/MT	–	–	–	–	–
1/2-5/2 (24)	s-d		–	–	E/MT	M/ET	–	–	–	–
3/2-5/2 (48)	p-f, d-d		–	M/ET	E/MT	M/ET	E/MT	–	–	–
3/2-7/2 (64)	p-f		–	–	E/MT	M/ET	E/MT	M/ET	–	–
5/2-7/2 (96)	f-f		–	M/ET	E/MT	M/ET	E/MT	M/ET	E/MT	–
1/2-1/2 (8)	s-p	−	M/ET	E/MT	–	–	–	–	–	–
3/2-3/2 (32)	p-d		M/ET	E/MT	M/ET	E/MT	–	–	–	–
5/2-5/2 (72)	d-f		M/ET	E/MT	M/ET	E/MT	M/ET	E/MT	–	–
1/2-3/2 (16)	s-p, p-d	−	–	E/MT	M/ET	–	–	–	–	–
1/2-5/2 (24)	s-f, p-d		–	–	M/ET	E/MT	–	–	–	–
1/2-7/2 (32)	s-f		–	–	–	E/MT	M/ET	–	–	–
3/2-5/2 (48)	p-d, d-f		–	E/MT	M/ET	E/MT	M/ET	–	–	–
3/2-7/2 (64)	d-f		–	–	M/ET	E/MT	M/ET	E/MT	–	–
5/2-7/2 (96)	d-f		–	E/MT	M/ET	E/MT	M/ET	E/MT	M/ET	–

TABLE IV. Operator expressions of even-parity multipoles up to $l = 4$ in the cubic O_h group. For a noncommute product, a symmetrized expression should be used, e.g., $AB \to (AB + B^\dagger A^\dagger)/2$. $\boldsymbol{m}_l$, $\boldsymbol{t}_l$, and $g_l^{\alpha\beta}$ are defined in Eqs. (5)–(7). The multipole with the prime requires complementary definitions other than Eqs. (1)–(4).

rank	type	irrep.	symbol	definition
0	E	A_{1g}^+	Q_0	1
	MT	A_{1g}^-	T_0'	i (off-diagonal element)
1	M	T_{1g}^-	$M_x,\ M_y,\ M_z$	$m_1^x,\ m_1^y,\ m_1^z$
	ET	T_{1g}^+	$G_x',\ G_y',\ G_z'$	$x\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma}),\ y\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma}),\ z\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma})$
2	E	E_g^+	$Q_u,\ Q_v$	$\frac{1}{2}(3z^2-r^2),\ \frac{\sqrt{3}}{2}(x^2-y^2)$
		T_{2g}^+	$Q_{yz},\ Q_{zx},\ Q_{xy}$	$\sqrt{3}yz,\ \sqrt{3}zx,\ \sqrt{3}xy$
	MT	E_g^-	$T_u,\ T_v$	$3zt_2^z-\boldsymbol{r}\cdot\boldsymbol{t}_2,\ \sqrt{3}(xt_2^x-yt_2^y)$
		T_{2g}^-	$T_{yz},\ T_{zx},\ T_{xy}$	$\sqrt{3}(yt_2^z+zt_2^y),\ \sqrt{3}(zt_2^x+xt_2^z),\ \sqrt{3}(xt_2^y+yt_2^x)$
3	M	A_{2g}^-	M_{xyz}	$\sqrt{15}(yzm_3^x+zxm_3^y+xym_3^z)$
		T_{1g}^-	$M_x^\alpha,\ M_y^\alpha,\ M_z^\alpha$	$3[\frac{1}{2}(3x^2-r^2)m_3^x-x(ym_3^y+zm_3^z)]$, (cyclic)
		T_{2g}^-	$M_x^\beta,\ M_y^\beta,\ M_z^\beta$	$\sqrt{15}[\frac{1}{2}(y^2-z^2)m_3^x+x(ym_3^y-zm_3^z)]$, (cyclic)
	ET	A_{2g}^+	G_{xyz}	$2\sqrt{15}(xg_3^{yz}+yg_3^{zx}+zg_3^{xy})$
		T_{1g}^+	$G_x^\alpha,\ G_y^\alpha,\ G_z^\alpha$	$9xg_3^{xx}-6(yg_3^{xy}+zg_3^{zx})-3x\sum_\alpha g_3^{\alpha\alpha}$, (cyclic)
		T_{2g}^+	$G_x^\beta,\ G_y^\beta,\ G_z^\beta$	$\sqrt{15}[2(yg_3^{xy}-zg_3^{zx})+x(g_3^{yy}-g_3^{zz})]$, (cyclic)
4	E	A_{1g}^+	Q_4	$\frac{5\sqrt{21}}{12}(x^4+y^4+z^4-\frac{3}{5}r^4)$
		E_g^+	$Q_{4u},\ Q_{4v}$	$\frac{7\sqrt{15}}{6}[z^4-\frac{x^4+y^4}{2}-\frac{3}{7}r^2(3z^2-r^2)],\ \frac{7\sqrt{5}}{4}[x^4-y^4-\frac{6}{7}r^2(x^2-y^2)]$
		T_{1g}^+	$Q_{4x}^\alpha,\ Q_{4y}^\alpha,\ Q_{4z}^\alpha$	$\frac{\sqrt{35}}{2}yz(y^2-z^2)$, (cyclic)
		T_{2g}^+	$Q_{4x}^\beta,\ Q_{4y}^\beta,\ Q_{4z}^\beta$	$\frac{\sqrt{5}}{2}yz(7x^2-r^2)$, (cyclic)
	MT	A_{1g}^-	T_4	$\frac{5\sqrt{21}}{3}[x^3t_4^x+y^3t_4^y+z^3t_4^z-\frac{3}{5}r^2(\boldsymbol{r}\cdot\boldsymbol{t}_4)]$
		E_g^-	T_{4u}	$\frac{\sqrt{15}}{3}[\sum_\alpha\alpha(5\alpha^2-3r^2)t_4^\alpha-3x(x^2-3y^2)t_4^x+3y(3x^2-y^2)t_4^y]$
			T_{4v}	$\sqrt{5}[x(x^2-3z^2)t_4^x-y(y^2-3z^2)t_4^y-3z(x^2-y^2)t_4^z]$
		T_{1g}^-	$T_{4x}^\alpha,\ T_{4y}^\alpha,\ T_{4z}^\alpha$	$\frac{\sqrt{35}}{2}[z(3y^2-z^2)t_4^y+y(y^2-3z^2)t_4^z]$, (cyclic)
		T_{2g}^-	$T_{4x}^\beta,\ T_{4y}^\beta,\ T_{4z}^\beta$	$6\sqrt{5}xyzt_4^x+\frac{\sqrt{5}}{2}(7x^2-r^2)(zt_4^y+yt_4^z)-\sqrt{5}yz(yt_4^y+zt_4^z)$, (cyclic)

inversion property as the E dipole, it is natural to define a time-reversal-even pseudoscalar $\hat{G}_0'$ as

$$\hat{G}_0' \equiv 6\sum_j \boldsymbol{m}_1(\boldsymbol{r}_j)\cdot\boldsymbol{t}_1(\boldsymbol{r}_j) = \sum_j \boldsymbol{l}_j\cdot(\boldsymbol{r}_j\times\boldsymbol{\sigma}_j), \tag{17}$$

where we have used $(\boldsymbol{r}\times\boldsymbol{l})\cdot\boldsymbol{l} = (\boldsymbol{r}\times\boldsymbol{\sigma})\cdot\boldsymbol{\sigma} = 0$ and the prefactor 6 for notational simplicity. Since $\hat{G}_0'$ acts as an elementary charge for the ET multipoles, the ET dipole $\hat{\boldsymbol{G}}' \equiv (\hat{G}_x', \hat{G}_y', \hat{G}_z')$ is obtained by multiplying the position vector $\boldsymbol{r}$ to $\hat{G}_0'$ as

$$\hat{\boldsymbol{G}}' \equiv \sum_j (\hat{G}_0')_j \boldsymbol{r}_j = \sum_j \boldsymbol{r}_j[\boldsymbol{l}_j\cdot(\boldsymbol{r}_j\times\boldsymbol{\sigma}_j)]. \tag{18}$$

It is noteworthy that $\hat{G}_0'$ and $\hat{\boldsymbol{G}}'$ are also valid only for the spinful basis.

On the other hand, the MT monopole is identically zero even by taking an inner product between the MT dipole $\boldsymbol{t}_1(\boldsymbol{r})$ and the position vector $\boldsymbol{r}$, since $\boldsymbol{t}_1(\boldsymbol{r})$ is perpendicular to $\boldsymbol{r}$. This is consistent with the fact that the MT monopole is unnecessary to span an arbitrary matrix in both nonhybrid and hybrid orbitals as shown in Table III. It is interesting to note that the MT monopole becomes active in hybrid orbitals with different additional quantum number within the same total angular momentum, e.g., $2p$-$4p$ and $3d$-$5d$ orbitals. From the correspondence between the E and MT multipoles, the expression of $\hat{T}_0'$ should be given by

$$\langle n_1 LM|\hat{T}_0'|n_2 LM\rangle \equiv i \quad (n_1 \neq n_2), \tag{19}$$

where n_1 and n_2 are the quantum numbers other than L and M, such as principal quantum numbers. Equation (19) means that $\hat{T}_0'$ represents the imaginary hybridization between the different orbitals with the same L and M.

Finally, let us make a remark on the further extension of multipoles in the spinful basis. In the spinful basis, the scalar product of $\boldsymbol{l}$ and $\boldsymbol{\sigma}$ can also be regarded as the E monopole, i.e.,

$$\hat{Q}_0' \equiv \sum_j \boldsymbol{l}_j\cdot\boldsymbol{\sigma}_j. \tag{20}$$

Then, by combining this elementary charge $\hat{Q}_0'$ with the polynomial $O_{lm}(\boldsymbol{r})$, we obtain the higher-rank E multipoles including the spin degree of freedom. Similar extensions for

TABLE V. Operator expressions of odd-parity multipoles up to $l = 4$ in the cubic $O_{\rm h}$ group.

rank	type	irrep.	symbol	definition
0	M	A_{1u}^-	M_0'	$\boldsymbol{r}\cdot\boldsymbol{\sigma}$
	ET	A_{1u}^+	G_0'	$\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma})$
1	E	T_{1u}^+	$Q_x,\ Q_y,\ Q_z$	$x,\ y,\ z$
	MT	T_{1u}^-	$T_x,\ T_y,\ T_z$	$t_1^x,\ t_1^y,\ t_1^z$
2	M	E_u^-	$M_u,\ M_v$	$3zm_2^z-\boldsymbol{r}\cdot\boldsymbol{m}_2,\ \sqrt{3}(xm_2^x-ym_2^y)$
		T_{2u}^-	$M_{yz},\ M_{zx},\ M_{xy}$	$\sqrt{3}(ym_2^z+zm_2^y),\ \sqrt{3}(zm_2^x+xm_2^z),\ \sqrt{3}(xm_2^y+ym_2^x)$
	ET	E_u^+	$G_u,\ G_v$	$3g_2^{zz}-\sum_\alpha g_2^{\alpha\alpha},\ \sqrt{3}(g_2^{xx}-g_2^{yy})$
		T_{2u}^+	$G_{yz},\ G_{zx},\ G_{xy}$	$2\sqrt{3}g_2^{yz},\ 2\sqrt{3}g_2^{zx},\ 2\sqrt{3}g_2^{xy}$
3	E	A_{2u}^+	Q_{xyz}	$\sqrt{15}xyz$
		T_{1u}^+	$Q_x^\alpha,\ Q_y^\alpha,\ Q_z^\alpha$	$\frac{1}{2}x(5x^2-3r^2)$, (cyclic)
		T_{2u}^+	$Q_x^\beta,\ Q_y^\beta,\ Q_z^\beta$	$\frac{\sqrt{15}}{2}x(y^2-z^2)$, (cyclic)
	MT	A_{2u}^-	T_{xyz}	$\sqrt{15}(yzt_3^x+zxt_3^y+xyt_3^z)$
		T_{1u}^-	$T_x^\alpha,\ T_y^\alpha,\ T_z^\alpha$	$3[\frac{1}{2}(3x^2-r^2)t_3^x-x(yt_3^y+zt_3^z)]$, (cyclic)
		T_{2u}^-	$T_x^\beta,\ T_y^\beta,\ T_z^\beta$	$\sqrt{15}[\frac{1}{2}(y^2-z^2)t_3^x+x(yt_3^y-zt_3^z)]$, (cyclic)
4	M	A_{1u}^-	M_4	$\frac{5\sqrt{21}}{3}[x^3m_4^x+y^3m_4^y+z^3m_4^z-\frac{3}{5}r^2(\boldsymbol{r}\cdot\boldsymbol{m}_4)]$
		E_u^-	M_{4u}	$\frac{\sqrt{15}}{3}[\sum_\alpha\alpha(5\alpha^2-3r^2)m_4^\alpha-3x(x^2-3y^2)m_4^x+3y(3x^2-y^2)m_4^y]$
			M_{4v}	$\sqrt{5}[x(x^2-3z^2)m_4^x-y(y^2-3z^2)m_4^y-3z(x^2-y^2)m_4^z]$
		T_{1u}^-	$M_{4x}^\alpha,\ M_{4y}^\alpha,\ M_{4z}^\alpha$	$\frac{\sqrt{35}}{2}[z(3y^2-z^2)m_4^y+y(y^2-3z^2)m_4^z]$, (cyclic)
		T_{2u}^-	$M_{4x}^\beta,\ M_{4y}^\beta,\ M_{4z}^\beta$	$6\sqrt{5}xyzm_4^x+\frac{\sqrt{5}}{2}(7x^2-r^2)(zm_4^y+ym_4^z)-\sqrt{5}yz(ym_4^y+zm_4^z)$, (cyclic)
	ET	A_{1u}^+	G_4	$-4\sqrt{21}(xyg_4^{xy}+yzg_4^{yz}+zxg_4^{zx})+\sqrt{21}\sum_\alpha(3\alpha^2-r^2)g_4^{\alpha\alpha}$
		E_u^+	G_{4u}	$\sqrt{15}[(3y^2-r^2)g_4^{xx}+(3x^2-r^2)g_4^{yy}+(3z^2-r^2)g_4^{zz}+4(2xyg_4^{xy}-xzg_4^{xz}-yzg_4^{yz})]$
			G_{4v}	$3\sqrt{5}[(x^2-z^2)g_4^{xx}+(z^2-y^2)g_4^{yy}+(y^2-x^2)g_4^{zz}-4(xzg_4^{xz}-yzg_4^{yz})]$
		T_{1u}^+	$G_{4x}^\alpha,\ G_{4y}^\alpha,\ G_{4z}^\alpha$	$3\sqrt{35}[yz(g_4^{yy}-g_4^{zz})+(y^2-z^2)g_4^{yz}]$, (cyclic)
		T_{2u}^+	$G_{4x}^\beta,\ G_{4y}^\beta,\ G_{4z}^\beta$	$3\sqrt{5}[4x(yg_4^{xz}+zg_4^{xy})+yz(2g_4^{xx}-g_4^{yy}-g_4^{zz})+(3x^2-r^2)g_4^{yz}]$, (cyclic)

$\hat{M}'_{lm}$, $\hat{T}'_{lm}$, and $\hat{G}'_{lm}$ are possible by combining the elementary charges, $\hat{M}'_0$, $\hat{T}'_0$, and $\hat{G}'_0$ with $O_{lm}(\boldsymbol{r})$.

The proper expressions of the even- and odd-parity multipole operators up to $l = 4$ in the cubic $O_{\rm h}$ group are summarized in Tables IV and V. For the hexagonal D_{6h} group, the proper expressions are also summarized in Tables XXIV and XXV in Appendix F. These expressions are sufficient to consider symmetry-classified multipoles in any point groups as they are subgroup of $O_{\rm h}$ or D_{6h} groups.

B. Multipoles in momentum space

The classification of multipoles is also applicable in momentum space. We present the momentum-based multipoles in single-band systems, which are expressed in terms of $(\boldsymbol{k},\ \sigma_0,\ \boldsymbol{\sigma})$, where $\boldsymbol{k}$ is the wave vector of electrons, and σ_0 and $\boldsymbol{\sigma}$ are the identity and Pauli matrices in the spin degree of freedom. The extension to multiband systems is not unique. We discuss a possible extension in Appendix B.

To construct the multipoles in momentum space, we first introduce the harmonics $O_{lm}(\boldsymbol{k})$ by replacing the position vector $\boldsymbol{r}$ with the wave vector $\boldsymbol{k}$ in $O_{lm}(\boldsymbol{r})$. Reflecting the polar vector of $\boldsymbol{k}$, the harmonics $\sigma_0 O_{lm}(\boldsymbol{k})$ represent the polar-type E and MT multipole degrees of freedom. As $\boldsymbol{k}$ is odd under time-reversal operation, the even-rank $\sigma_0 O_{lm}(\boldsymbol{k})$ represents the E multipoles, while the odd-rank $\sigma_0 O_{lm}(\boldsymbol{k})$ represents the MT multipoles. By multiplying $O_{lm}(\boldsymbol{k})$ by the operator $(\boldsymbol{k}\times\boldsymbol{\sigma})\cdot\boldsymbol{\nabla}_{\boldsymbol{k}}$, which reverses its time-reversal property while keeping its parity and rank, the expressions of the odd-rank E multipole and the even-rank MT multipole are obtained. Thus, the expressions of E and MT multipole in momentum space are given by

$$Q_{lm}(\boldsymbol{k}) \equiv \begin{cases} \sigma_0 O_{lm}(\boldsymbol{k}) & (l = 0, 2, 4, 6, \dots) \\ (\boldsymbol{k}\times\boldsymbol{\sigma})\cdot\boldsymbol{\nabla}_{\boldsymbol{k}} O_{lm}(\boldsymbol{k}) & (l = 1, 3, 5, \dots) \end{cases}, \tag{21}$$

$$T_{lm}(\boldsymbol{k}) \equiv \begin{cases} 0 & (l = 0) \\ (\boldsymbol{k}\times\boldsymbol{\sigma})\cdot\boldsymbol{\nabla}_{\boldsymbol{k}} O_{lm}(\boldsymbol{k}) & (l = 2, 4, 6, \dots). \\ \sigma_0 O_{lm}(\boldsymbol{k}) & (l = 1, 3, 5, \dots) \end{cases} \tag{22}$$

Note that T_0 vanishes owing to the derivative of $O_{lm}(\boldsymbol{k})$.

Similarly, the axial-type M and ET multipoles in momentum space are expressed by multiplying $\sigma_0 O_{lm}(\boldsymbol{k})$ by the operator $\boldsymbol{\sigma}\cdot\boldsymbol{\nabla}_{\boldsymbol{k}}$, which reverses its parity while keeping its time-reversal property and rank. Therefore the odd-rank M multipoles and the even-rank ET multipoles are given by $\boldsymbol{\sigma}\cdot\boldsymbol{\nabla}_{\boldsymbol{k}} O_{lm}(\boldsymbol{k})$. On the other hand, it is impossible to construct the even-rank M multipoles and the odd-rank ET multipoles within the single-band systems. They require the multiorbital or sublattice degree of freedom, as discussed in Appendix B. Thus the M and ET multipoles are

TABLE VI. Even-parity multipoles in momentum space up to $l = 4$ in the cubic O_h group. The higher-order expression with respect to $\boldsymbol{k}$ is also shown for Q_0 and (M_x, M_y, M_z) in the bracket. The multipole with the prime becomes active in the multiorbital systems as discussed in Appendix B.

rank	type	irrep.	symbol	definition
0	E	A_{1g}^+	Q_0	$\sigma_0\ [(k_x^2+k_y^2+k_z^2)\sigma_0]$
	MT	A_{1g}^-	T_0'	$i\sigma_0$
1	M	T_{1g}^-	$M_x,\ M_y,\ M_z$	$\sigma_x,\ \sigma_y,\ \sigma_z\ [(\boldsymbol{k}\cdot\boldsymbol{\sigma})k_x - \frac{1}{3}k^2\sigma_x$, (cyclic)]
	ET	T_{1g}^+	$G_x',\ G_y',\ G_z'$	$i\sigma_x,\ i\sigma_y,\ i\sigma_z$
2	E	E_g^+	$Q_u,\ Q_v$	$\frac{1}{2}(3k_z^2-k^2)\sigma_0,\ \frac{\sqrt{3}}{2}(k_x^2-k_y^2)\sigma_0$
		T_{2g}^+	$Q_{yz},\ Q_{zx},\ Q_{xy}$	$\sqrt{3}k_yk_z\sigma_0$, (cyclic)
	MT	E_g^-	$T_u,\ T_v$	$3k_zQ_z - \boldsymbol{k}\cdot\boldsymbol{Q},\ \sqrt{3}(k_xQ_x - k_yQ_y)$
		T_{2g}^-	$T_{yz},\ T_{zx},\ T_{xy}$	$\sqrt{3}(k_yQ_z + k_zQ_y)$, (cyclic)
3	M	A_{2g}^-	M_{xyz}	$\sqrt{15}(k_yk_z\sigma_x + k_zk_x\sigma_y + k_xk_y\sigma_z)$
		T_{1g}^-	$M_x^\alpha,\ M_y^\alpha,\ M_z^\alpha$	$3[\frac{1}{2}(3k_x^2-k^2)\sigma_x - k_x(k_y\sigma_y + k_z\sigma_z)]$, (cyclic)
		T_{2g}^-	$M_x^\beta,\ M_y^\beta,\ M_z^\beta$	$\sqrt{15}[\frac{1}{2}(k_y^2-k_z^2)\sigma_x + k_x(k_y\sigma_y - k_z\sigma_z)]$, (cyclic)
	ET	A_{2g}^+	G_{xyz}'	$\sqrt{15}i(k_yk_z\sigma_x + k_zk_x\sigma_y + k_xk_y\sigma_z)$
		T_{1g}^+	$G_x^{\alpha'},\ G_y^{\alpha'},\ G_z^{\alpha'}$	$3i[\frac{1}{2}(3k_x^2-k^2)\sigma_x - k_x(k_y\sigma_y + k_z\sigma_z)]$, (cyclic)
		T_{2g}^+	$G_x^{\beta'},\ G_y^{\beta'},\ G_z^{\beta'}$	$\sqrt{15}i[\frac{1}{2}(k_y^2-k_z^2)\sigma_x + k_x(k_y\sigma_y - k_z\sigma_z)]$, (cyclic)
4	E	A_{1g}^+	Q_4	$\frac{5\sqrt{21}}{12}(k_x^4+k_y^4+k_z^4-\frac{3}{5}k^4)\sigma_0$
		E_g^+	$Q_{4u},\ Q_{4v}$	$\frac{7\sqrt{15}}{6}[k_z^4 - \frac{k_x^4+k_y^4}{2} - \frac{3}{7}k^2(3k_z^2-k^2)]\sigma_0,\ \frac{7\sqrt{5}}{4}[k_x^4-k_y^4-\frac{6}{7}k^2(k_x^2-k_y^2)]\sigma_0$
		T_{1g}^+	$Q_{4x}^\alpha,\ Q_{4y}^\alpha,\ Q_{4z}^\alpha$	$\frac{\sqrt{35}}{2}k_yk_z(k_y^2-k_z^2)\sigma_0$, (cyclic)
		T_{2g}^+	$Q_{4x}^\beta,\ Q_{4y}^\beta,\ Q_{4z}^\beta$	$\frac{\sqrt{5}}{2}k_yk_z(7k_x^2-k^2)\sigma_0$, (cyclic)
	MT	A_{1g}^-	T_4	$\frac{5\sqrt{21}}{3}[k_x(k_x^2-\frac{3}{5}k^2)Q_x + k_y(k_y^2-\frac{3}{5}k^2)Q_y + k_z(k_z^2-\frac{3}{5}k^2)Q_z]$
		E_g^-	T_{4u}	$\frac{\sqrt{15}}{3}[\sum_\alpha k_\alpha(5k_\alpha^2-3k^2)Q_\alpha - 3k_x(k_x^2-3k_y^2)Q_x + 3k_y(3k_x^2-k_y^2)Q_y]$
			T_{4v}	$\sqrt{5}[k_x(k_x^2-3k_z^2)Q_x - k_y(k_y^2-3k_z^2)Q_y - 3k_z(k_x^2-k_y^2)Q_z]$
		T_{1g}^-	$T_{4x}^\alpha,\ T_{4y}^\alpha,\ T_{4z}^\alpha$	$\frac{\sqrt{35}}{2}[k_y(k_y^2-3k_z^2)Q_z - k_z(k_z^2-3k_y^2)Q_y]$, (cyclic)
		T_{2g}^-	$T_{4x}^\beta,\ T_{4y}^\beta,\ T_{4z}^\beta$	$\frac{\sqrt{5}}{2}[12k_xk_yk_zQ_x + k_z\{(5k_z^2-3k^2)-3(k_z^2-3k_x^2)\}Q_y + k_y\{(5k_y^2-3k^2)-3(k_y^2-3k_x^2)\}Q_z]$, (cyclic)

given by

$$M_{lm}(\boldsymbol{k}) \equiv \begin{cases} 0 & (l = 0, 2, 4, 6, \dots) \\ \boldsymbol{\sigma}\cdot\nabla_{\boldsymbol{k}} O_{lm}(\boldsymbol{k}) & (l = 1, 3, 5, \dots) \end{cases}, \tag{23}$$

$$G_{lm}(\boldsymbol{k}) \equiv \begin{cases} \boldsymbol{k}\cdot\boldsymbol{\sigma} & (l = 0) \\ \boldsymbol{\sigma}\cdot\nabla_{\boldsymbol{k}} O_{lm}(\boldsymbol{k}) & (l = 2, 4, 6, \dots). \\ 0 & (l = 1, 3, 5, \dots) \end{cases} \tag{24}$$

We complementarily define $G_0(\boldsymbol{k})$ as $\boldsymbol{k}\cdot\boldsymbol{\sigma}$ from the symmetry consideration. The even- and odd-parity multipole operators in momentum space in the O_h group up to $l = 4$ are summarized in Tables VI and VII. The hexagonal version is summarized in Tables XXVI and XXVII in Appendix F.

III. PHYSICAL PROPERTIES IN THE PRESENCE OF MULTIPOLES

When a thermodynamic average of a multipole degree of freedom $X_{lm} \equiv \langle \hat{X}_{lm} \rangle$, which we call a multipole moment, becomes nonzero, it generates anisotropic electric and/or magnetic fields via electromagnetic potentials. Moreover, it affects electronic states and leads to unconventional physical phenomena, such as the magnetocurrent effect through multipole-multipole interactions. In this section, we first give the formula for electric and magnetic fields in the presence of the multipole moment in Sec. III A. Then, we show how the multipole moment affects the electronic band structures in Sec. III B. We also classify linear response tensors, such as magnetoelectric and magnetocurrent tensors, from the multipole point of view in Sec. III C.

A. Electromagnetic potential and electric and magnetic fields

By the definition of the multipole expansion of the scalar and vector potentials with the Coulomb gauge $\nabla\cdot\boldsymbol{A} = 0$ [21], we obtain

$$\phi(\boldsymbol{r}) = \sum_{lm} \sqrt{\frac{4\pi}{2l+1}} Q_{lm} \frac{Y_{lm}(\hat{\boldsymbol{r}})}{r^{l+1}}, \tag{25}$$

$$\boldsymbol{A}(\boldsymbol{r}) = \sum_{lm} \left[i\sqrt{\frac{4\pi(l+1)}{(2l+1)l}} M_{lm} \frac{\boldsymbol{Y}_{lm}^{l}(\hat{\boldsymbol{r}})}{r^{l+1}} - \sqrt{4\pi(l+1)} T_{lm} \frac{\boldsymbol{Y}_{lm}^{l+1}(\hat{\boldsymbol{r}})}{r^{l+2}} \right], \tag{26}$$

where $\boldsymbol{Y}_{lm}^{l'}(\hat{\boldsymbol{r}})$ $(l' = l-1, l, l+1)$ is the vector spherical harmonics that transforms like $Y_{lm}(\hat{\boldsymbol{r}})$ under spatial rotation and l' is its orbital angular momentum [8,9]. By taking derivatives, we obtain the electric and magnetic fields in the presence of

TABLE VII. Odd-parity multipoles in momentum space up to $l = 4$ in the cubic $O_{\rm h}$ group.

rank	type	irrep.	symbol	definition
0	M	A_{1u}^-	M_0'	$i\boldsymbol{k}\cdot\boldsymbol{\sigma}$
	ET	A_{1u}^+	G_0	$\boldsymbol{k}\cdot\boldsymbol{\sigma}$
1	E	T_{1u}^+	$Q_x,\ Q_y,\ Q_z$	$k_y\sigma_z - k_z\sigma_y,\ k_z\sigma_x - k_x\sigma_z,\ k_x\sigma_y - k_y\sigma_x$
	MT	T_{1u}^-	$T_x,\ T_y,\ T_z$	$k_x\sigma_0,\ k_y\sigma_0,\ k_z\sigma_0$
2	M	E_u^-	$M_u',\ M_v'$	$i(3k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma}),\ \sqrt{3}i(k_x\sigma_x - k_y\sigma_y)$
		T_{2u}^-	$M_{yz}',\ M_{zx}',\ M_{xy}'$	$\sqrt{3}i(k_y\sigma_z + k_z\sigma_y)$, (cyclic)
	ET	E_u^+	$G_u,\ G_v$	$3k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma},\ \sqrt{3}(k_x\sigma_x - k_y\sigma_y)$
		T_{2u}^+	$G_{yz},\ G_{zx},\ G_{xy}$	$\sqrt{3}(k_y\sigma_z + k_z\sigma_y)$, (cyclic)
3	E	A_{2u}^+	Q_{xyz}	$\sqrt{15}(k_yk_zQ_x + k_zk_xQ_y + k_xk_yQ_z)$
		T_{1u}^+	$Q_x^\alpha,\ Q_y^\alpha,\ Q_z^\alpha$	$3[\frac{1}{2}(3k_x^2 - k^2)Q_x - k_x(k_yQ_y + k_zQ_z)]$, (cyclic)
		T_{2u}^+	$Q_x^\beta,\ Q_y^\beta,\ Q_z^\beta$	$\sqrt{15}[\frac{1}{2}(k_y^2 - k_z^2)Q_x + k_x(k_yQ_y - k_zQ_z)]$, (cyclic)
	MT	A_{2u}^-	T_{xyz}	$\sqrt{15}k_xk_yk_z\sigma_0$
		T_{1u}^-	$T_x^\alpha,\ T_y^\alpha,\ T_z^\alpha$	$\frac{1}{2}k_x(5k_x^2 - 3k^2)\sigma_0$, (cyclic)
		T_{2u}^-	$T_x^\beta,\ T_y^\beta,\ T_z^\beta$	$\frac{\sqrt{15}}{2}k_x(k_y^2 - k_z^2)\sigma_0$, (cyclic)
4	M	A_{1u}^-	M_4'	$\frac{5\sqrt{21}}{3}i[k_x(k_x^2 - \frac{3}{5}k^2)\sigma_x + k_y(k_y^2 - \frac{3}{5}k^2)\sigma_y + k_z(k_z^2 - \frac{3}{5}k^2)\sigma_z]$
		E_u^-	M_{4u}'	$\frac{\sqrt{15}}{3}i[\sum_\alpha k_\alpha(5k_\alpha^2 - 3k^2)\sigma_\alpha - 3k_x(k_x^2 - 3k_y^2)\sigma_x + 3k_y(3k_x^2 - k_y^2)\sigma_y]$
			M_{4v}'	$\sqrt{5}i[k_x(k_x^2 - 3k_z^2)\sigma_x - k_y(k_y^2 - 3k_z^2)\sigma_y - 3k_z(k_x^2 - k_y^2)\sigma_z]$
		T_{1u}^-	$M_{4x}^{\alpha'},\ M_{4y}^{\alpha'},\ M_{4z}^{\alpha'}$	$\frac{\sqrt{35}}{2}i[k_y(k_y^2 - 3k_z^2)\sigma_z - k_z(k_z^2 - 3k_y^2)\sigma_y]$, (cyclic)
		T_{2u}^-	$M_{4x}^{\beta'},\ M_{4y}^{\beta'},\ M_{4z}^{\beta'}$	$\frac{\sqrt{5}}{2}i[12k_xk_yk_z\sigma_x + k_z\{(5k_z^2 - 3k^2) - 3(k_z^2 - 3k_x^2)\}\sigma_y + k_y\{(5k_y^2 - 3k^2) - 3(k_y^2 - 3k_x^2)\}\sigma_z]$, (cyclic)
	ET	A_{1u}^+	G_4	$\frac{5\sqrt{21}}{3}[k_x(k_x^2 - \frac{3}{5}k^2)\sigma_x + k_y(k_y^2 - \frac{3}{5}k^2)\sigma_y + k_z(k_z^2 - \frac{3}{5}k^2)\sigma_z]$
		E_u^+	G_{4u}	$\frac{\sqrt{15}}{3}[\sum_\alpha k_\alpha(5k_\alpha^2 - 3k^2)\sigma_\alpha - 3k_x(k_x^2 - 3k_y^2)\sigma_x + 3k_y(3k_x^2 - k_y^2)\sigma_y]$
			G_{4v}	$\sqrt{5}[k_x(k_x^2 - 3k_z^2)\sigma_x - k_y(k_y^2 - 3k_z^2)\sigma_y - 3k_z(k_x^2 - k_y^2)\sigma_z]$
		T_{1u}^+	$G_{4x}^\alpha,\ G_{4y}^\alpha,\ G_{4z}^\alpha$	$\frac{\sqrt{35}}{2}[k_y(k_y^2 - 3k_z^2)\sigma_z - k_z(k_z^2 - 3k_y^2)\sigma_y]$, (cyclic)
		T_{2u}^+	$G_{4x}^\beta,\ G_{4y}^\beta,\ G_{4z}^\beta$	$\frac{\sqrt{5}}{2}[12k_xk_yk_z\sigma_x + k_z\{(5k_z^2 - 3k^2) - 3(k_z^2 - 3k_x^2)\}\sigma_y + k_y\{(5k_y^2 - 3k^2) - 3(k_y^2 - 3k_x^2)\}\sigma_z]$, (cyclic)

the multipole moment as

$$\boldsymbol{E}(\boldsymbol{r}) = -\nabla\phi = -\sum_{lm}\sqrt{4\pi(l+1)}Q_{lm}\frac{\boldsymbol{Y}_{lm}^{l+1}(\hat{\boldsymbol{r}})}{r^{l+2}}, \tag{27}$$

$$\boldsymbol{B}(\boldsymbol{r}) = \nabla\times\boldsymbol{A} = -\sum_{lm}\sqrt{4\pi(l+1)}M_{lm}\frac{\boldsymbol{Y}_{lm}^{l+1}(\hat{\boldsymbol{r}})}{r^{l+2}}. \tag{28}$$

Note that $\boldsymbol{E}(\boldsymbol{r})$ and $\boldsymbol{B}(\boldsymbol{r})$ have formally equivalent forms with Q_{lm} and M_{lm}, and no toroidal moments G_{lm} and T_{lm} appear in these expressions. Thus the toroidal moments G_{lm} and T_{lm} affect physical quantities not through classical electromagnetic interactions but through the quantum-mechanical multipole-multipole interactions. The latter also affects the phase of electrons through the vector potential, $e\boldsymbol{A}$.

It is interesting to note that by using the dual nature of electric and magnetic quantities, we can introduce dual potentials with opposite space-time-parity counterparts as

$$\phi^*(\boldsymbol{r}) \equiv \sum_{lm}\sqrt{\frac{4\pi}{2l+1}}M_{lm}\frac{Y_{lm}(\hat{\boldsymbol{r}})}{r^{l+1}}, \tag{29}$$

$$\boldsymbol{A}^*(\boldsymbol{r}) \equiv \sum_{lm}\left[i\sqrt{\frac{4\pi(l+1)}{(2l+1)l}}Q_{lm}\frac{\boldsymbol{Y}_{lm}^{l}(\hat{\boldsymbol{r}})}{r^{l+1}} - \sqrt{4\pi(l+1)}G_{lm}\frac{\boldsymbol{Y}_{lm}^{l+1}(\hat{\boldsymbol{r}})}{r^{l+2}}\right]. \tag{30}$$

Then, the electric and magnetic fields are given by $\boldsymbol{E} = \nabla\times\boldsymbol{A}^*$ and $\boldsymbol{B} = -\nabla\phi^*$. If a magnetic monopole charge e^* were present, G_{lm} could affect the phase of electrons via $e^*\boldsymbol{A}^*$. The expression in Eq. (30) indicates why the electric toroidal multipoles G_{lm} is the counterpart of magnetic toroidal multipoles T_{lm}, and represents a time-reversal-even axial degree of freedom in electrons.

B. Electronic band structure

By using the expressions of multipoles in momentum space in Sec. II B, we find the effect of multipoles on the electronic band structure from the microscopic viewpoint. Conversely, we can deduce what type of multipoles are activated when the electronic band structure is determined from the first-principles band calculations or detected by the ARPES and de Haas-van Alphen measurements, and so on.

A Hamiltonian in condensed matter physics must be totally symmetric and time-reversal even. Therefore the one-electron Hamiltonian must be in the scalar-product form as

$$\mathcal{H} = \sum_{X}^{Q,M,T,G}\sum_{\boldsymbol{k}\sigma\sigma'}\sum_{lm}X_{lm}^{\rm ext}X_{lm}^{\sigma\sigma'}(\boldsymbol{k})c_{\boldsymbol{k}\sigma}^\dagger c_{\boldsymbol{k}\sigma'}, \tag{31}$$

where $c_{\boldsymbol{k}\sigma}^\dagger$ ($c_{\boldsymbol{k}\sigma}$) is the creation (annihilation) operator of an electron with the wave vector $\boldsymbol{k}$ and spin $\boldsymbol{\sigma}$. Note that

TABLE VIII. Representative external fields. $\boldsymbol{u}$ represents a displacement vector field.

multipole	$\mathcal{P}$	$\mathcal{T}$	external field	symbol
$\boldsymbol{G}^{\rm ext}$	+	+	rotation	$\boldsymbol{\omega} = (\nabla \times \boldsymbol{u})/2$
$Q_0^{\rm ext}$, $Q_{2m}^{\rm ext}$		+	strain	$\varepsilon_{ij} = (\partial_i u_j + \partial_j u_i)/2$
$\boldsymbol{M}^{\rm ext}$		−	magnetic field	$\boldsymbol{H}$
$\boldsymbol{Q}^{\rm ext}$	−	+	electric field	$\boldsymbol{E}$
$\boldsymbol{T}^{\rm ext}$		−	electric current	$\boldsymbol{J}$, $\nabla \times \boldsymbol{H}$

we assume X_{lm} in real representation so that the Hermite conjugation is omitted in the scalar product. For example, the lower-rank contributions read from Tables VI and VII as

$$\mathcal{H} = \sum_{\boldsymbol{k}\sigma\sigma'} \Big[\frac{\hbar^2 \boldsymbol{k}^2}{2m}\sigma_0 + \boldsymbol{Q}^{\rm ext}\cdot(\boldsymbol{k}\times\boldsymbol{\sigma}) + \boldsymbol{M}^{\rm ext}\cdot\boldsymbol{\sigma} + \boldsymbol{T}^{\rm ext}\cdot\boldsymbol{k}\,\sigma_0 + G_0^{\rm ext}(\boldsymbol{k}\cdot\boldsymbol{\sigma}) + \cdots \Big]_{\sigma\sigma'} c^\dagger_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma'}, \quad (32)$$

where it has taken the form $Q_0^{\rm ext} = \hbar^2/2m$.

$X_{lm}^{\rm ext}$ ($X = Q, M, T,$ and G) represent "symmetry-breaking" fields, which bring about symmetry-breaking for the c-electron systems. The microscopic origins of $X_{lm}^{\rm ext}$ are the external fields applied to the systems, the crystalline electric field (CEF) from ligand ions, and molecular fields originating from multipole-multipole interactions by the spontaneous electronic orderings, and so on. For example, $\boldsymbol{M}^{\rm ext}$ arises from an external magnetic field $\boldsymbol{H}$ or molecular field of ferromagnetic ordering. The representative external fields are summarized in Table VIII.

In order to examine the effect on the band structure, the classification according to their spatial-time inversion properties is useful. In the single-band systems, the Hamiltonian in Eq. (31) can be divided as

$$\mathcal{H} = \sum_{\boldsymbol{k}\sigma\sigma'} \big[\varepsilon^{\rm S}(\boldsymbol{k})\delta_{\sigma\sigma'} + \varepsilon^{\rm A}(\boldsymbol{k})\delta_{\sigma\sigma'} + f^{\rm S}_{\sigma\sigma'}(\boldsymbol{k}) + f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k}) \big] c^\dagger_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma'}, \quad (33)$$

where $\varepsilon(f_{\sigma\sigma'})$ is the charge(spin) sector and the superscript S(A) represents symmetric(antisymmetric) contribution with respect to $\boldsymbol{k}$. As the even-rank $Q_{lm}(\boldsymbol{k})$, $T_{lm}(\boldsymbol{k})$, and the odd-rank $M_{lm}(\boldsymbol{k})$ are even function of $\boldsymbol{k}$, and other multipoles are odd function of $\boldsymbol{k}$, each coefficient is identified as

$$\varepsilon^{\rm S}(\boldsymbol{k}) = \sum_{lm}^{\rm even} Q_{lm}^{\rm ext} Q_{lm}(\boldsymbol{k}), \quad (34)$$

$$\varepsilon^{\rm A}(\boldsymbol{k}) = \sum_{lm}^{\rm odd} T_{lm}^{\rm ext} T_{lm}(\boldsymbol{k}), \quad (35)$$

$$f^{\rm S}_{\sigma\sigma'}(\boldsymbol{k}) = \sum_{lm}^{\rm odd} M_{lm}^{\rm ext} M_{lm}^{\sigma\sigma'}(\boldsymbol{k}) + \sum_{lm}^{\rm even} T_{lm}^{\rm ext} T_{lm}^{\sigma\sigma'}(\boldsymbol{k}), \quad (36)$$

$$f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k}) = \sum_{lm}^{\rm even} G_{lm}^{\rm ext} G_{lm}^{\sigma\sigma'}(\boldsymbol{k}) + \sum_{lm}^{\rm odd} Q_{lm}^{\rm ext} Q_{lm}^{\sigma\sigma'}(\boldsymbol{k}). \quad (37)$$

TABLE IX. Interplay between the multipole moments and space-time classified part of electronic structure. $\mathcal{P}$ and $\mathcal{T}$ represent the spatial inversion and time-reversal operations corresponding to $(\boldsymbol{k}, \sigma_0, \boldsymbol{\sigma}) \to (-\boldsymbol{k}, \sigma_0, \boldsymbol{\sigma})$ and $(\boldsymbol{k}, \sigma_0, \boldsymbol{\sigma}) \to (-\boldsymbol{k}, \sigma_0, -\boldsymbol{\sigma})$, respectively. S(A) represents the symmetric (antisymmetric) contribution arising in the band structure, and SS is the abbreviation of the spin splitting.

type	$\mathcal{P}$	$\mathcal{T}$	multipole	band structure
$\varepsilon^{\rm S}(\boldsymbol{k})$	+	+	$Q_{lm}^{\rm ext}$ (even)	S w/o SS
$f^{\rm S}_{\sigma\sigma'}(\boldsymbol{k})$		−	$M_{lm}^{\rm ext}$ (odd), $T_{lm}^{\rm ext}$ (even)	S w/ SS
$f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k})$	−	+	$G_{lm}^{\rm ext}$ (even), $Q_{lm}^{\rm ext}$ (odd)	A w/ SS
$\varepsilon^{\rm A}(\boldsymbol{k})$		−	T_{lm} (odd)	A w/o SS

When we consider the electronic band structure under symmetry-breaking fields, we apply symmetry operations only to the c-electron systems $X_{lm}(\boldsymbol{k})$ without acting on $X_{lm}^{\rm ext}$. Thus, for single-band systems, the spatial inversion operation $\mathcal{P}$ corresponds to $(\boldsymbol{k}, \sigma_0, \boldsymbol{\sigma}) \to (-\boldsymbol{k}, \sigma_0, \boldsymbol{\sigma})$, while the time-reversal operation $\mathcal{T}$ corresponds to $(\boldsymbol{k}, \sigma_0, \boldsymbol{\sigma}) \to (-\boldsymbol{k}, \sigma_0, -\boldsymbol{\sigma})$. The effects of the multipole moments on each part of the electronic structure are summarized in Table IX. We discuss coefficients in Eqs. (34)–(37) one by one below.

$\varepsilon^{\rm S}(\boldsymbol{k})$ in Eq. (34) represents the symmetric-type band dispersions without the spin splitting. This band structure is present when there are both the spatial inversion and time-reversal symmetries. For example, the kinetic energy of free electron, which is given by $\varepsilon^{\rm S}(\boldsymbol{k}) = \hbar^2\boldsymbol{k}^2/2m$, corresponds to the E monopole in terms of multipole terminology. The higher-rank quadrupole-type deformation in the band structure is attributed to the presence of $Q_{2m}^{\rm ext} = (Q_u^{\rm ext}, Q_v^{\rm ext}, Q_{yz}^{\rm ext}, Q_{zx}^{\rm ext}, Q_{xy}^{\rm ext})$. The band deformation caused by $Q_{xy}^{\rm ext}$ is shown in Fig. 2, for instance. This quadrupole-type deformation corresponds to orbital (nematic) orderings, which have been discussed in iron-based superconductors [116–121] and $Sr_3Ru_2O_7$ [122–124].

$f^{\rm S}_{\sigma\sigma'}(\boldsymbol{k})$ in Eq. (36) represents the symmetric-type band dispersions with the spin splitting. This band structure appears once the time-reversal symmetry is broken while the spatial inversion symmetry is still present. The odd-rank M multipoles and even-rank MT multipoles are categorized in this symmetry group. The band structure shows the spin polarization owing to the time-reversal symmetry breaking. The typical example is the band structure in the ferromagnetic ordering; the M dipole M_z leads to the symmetric spin splitting with the dispersions

$$\varepsilon_\sigma(\boldsymbol{k}) = \frac{\hbar^2\boldsymbol{k}^2}{2m} + \sigma H_z, \quad (38)$$

where $M_z^{\rm ext} = H_z$ and $\sigma = \pm 1$. The results indicate a uniform spin splitting in the band structure, as shown in Fig. 2. In the case of the higher-rank multipoles, e.g., the MT quadrupole T_{xy}, the band dispersions are given by

$$\varepsilon_\sigma(\boldsymbol{k}) = \frac{\hbar^2\boldsymbol{k}^2}{2m} + \sigma T_{xy}^{\rm ext}\sqrt{\big(k_x^2 - k_y^2\big)^2 + \big(k_x^2 + k_y^2\big)k_z^2}, \quad (39)$$

which indicates the quadrupole-type spin splitting, as shown in Fig. 2.

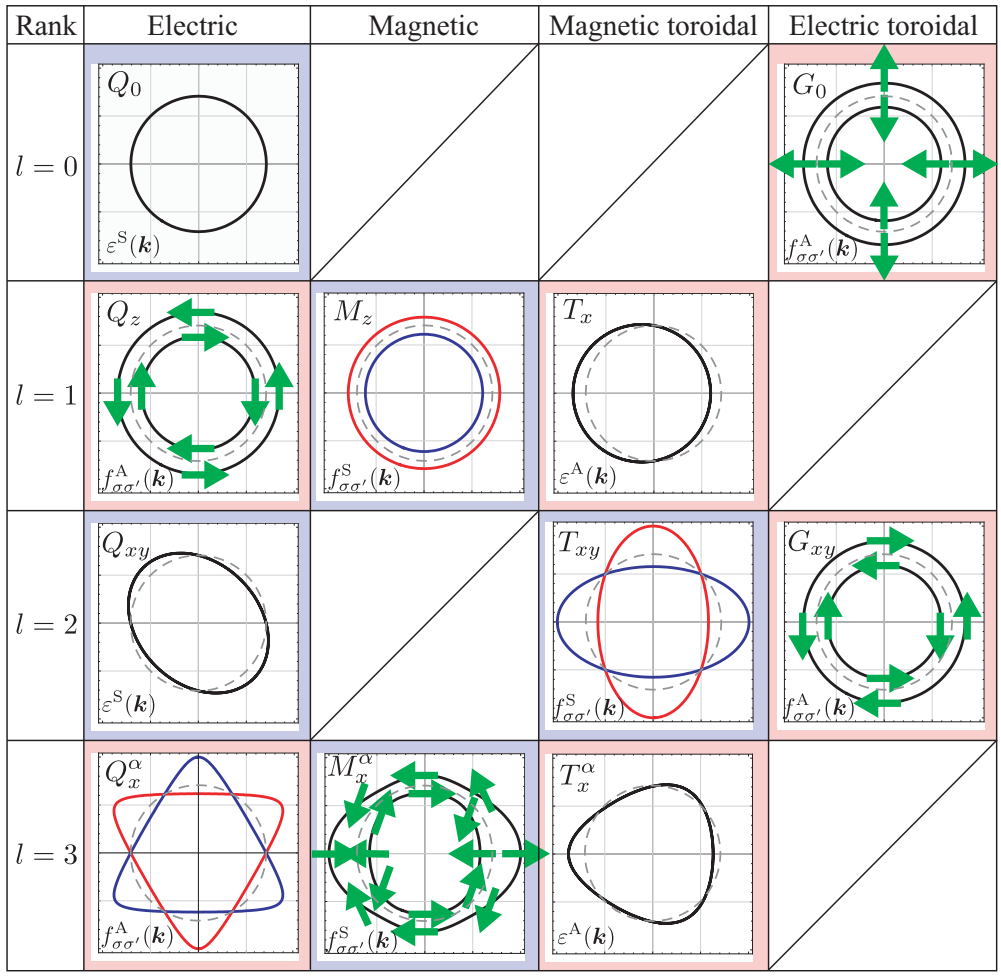

FIG. 2. Examples of the typical electronic band modulations in the k_x-k_y plane at $k_z = 0$ in the presence of the E, M, MT, and ET multipoles up to the rank $l = 3$ in the single-band picture. The blue (red) boxes stand for the band modulations by the even-parity (odd-parity) multipoles. The green arrows represent the in-plane spin moments at $\boldsymbol{k}$, while the red (blue) curves show the up (down)-spin moments along the out-of-plane direction. The dashed circles in each box represent the energy contours in the absence of the multipole, which corresponds to the energy contour for $Q_0^{\rm ext}$. The superscript "ext" of $X_{lm}^{\rm ext}$ is omitted for simplicity.

$f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k})$ in Eq. (37) represents the asymmetric-type band dispersions (the antisymmetric contribution to the background symmetric band dispersions) with the spin splitting. The odd-rank E multipoles and even-rank ET multipoles contribute to $f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k})$. This band structure is characterized by the breaking of the spatial inversion symmetry and is often realized in the presence of the spin-orbit coupling. For example, the Rashba-type spin-orbit coupling with the form of $k_y\sigma_x - k_x\sigma_y$ corresponds to the E dipole Q_z, while the Dresselhaus-type spin-orbit coupling $k_x(k_y^2 - k_z^2)\sigma_x + k_y(k_z^2 - k_x^2)\sigma_y + k_z(k_x^2 - k_y^2)\sigma_z$ corresponds to E octupole Q_{xyz}, as shown in Table VII. In other words, the Rashba-type (Dresselhaus-type) spin-orbit coupling appears when the corresponding odd-parity $\boldsymbol{Q}^{\rm ext}$ ($Q_{xyz}^{\rm ext}$) is present due to the crystal structure. To be specific, the E dipole component in $f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k})$ is induced by polar point groups (C_{4v}, C_4, C_{2v}, C_2, C_s, C_{6v}, C_6, C_{3v}, C_3, C_1), while ET monopole, E dipole, or the ET quadrupole component in $f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k})$ is induced by gyrotropic point groups (O, T, D_4, D_{2d}, C_{4v}, C_4, S_4, D_2, C_{2v}, C_2, C_s, D_6, C_{6v}, C_6, D_3, C_{3v}, C_3, C_1), as discussed in Sec. IV.

$\varepsilon^{\rm A}(\boldsymbol{k})$ in Eq. (35) represents the asymmetric-type band dispersions with the spin degeneracy. This band structure is obtained when the systems lack both the spatial inversion and time-reversal symmetries. The multipoles in this category are the odd-rank MT ones, which exhibit the odd-order $\boldsymbol{k}$ dispersions. For example, the MT dipole T_z order leads to the $\boldsymbol{k}$-linear dispersions,

$$\varepsilon(\boldsymbol{k}) = \frac{\hbar^2\boldsymbol{k}^2}{2m} + T_z^{\rm ext}k_z, \tag{40}$$

whereas the MT octupole leads to the third-order modulations with respect to $\boldsymbol{k}$; $k_x(5k_x^2 - 3\boldsymbol{k}^2)$ for $T_x^{\alpha\,\rm ext}$, which is schematically shown in Fig. 2.

Note that asymmetric band modulations are also obtained by combining the multipoles belonging to $f^{\rm S}_{\sigma\sigma'}(\boldsymbol{k})$ and $f^{\rm A}_{\sigma\sigma'}(\boldsymbol{k})$. For example, the noncentrosymmetric polar ferromagnets where the M dipole moment lies in the x direction

TABLE X. Relation between the components of the linear-response rank-2 tensors $\chi_{ij}^{(2)} = \chi_{ij}^{(\mathrm{J})} + \chi_{ij}^{(\mathrm{E})}$ and relevant multipoles in the presence ($\bigcirc$) or absence ($\times$) of $(\mathcal{P}, \mathcal{T}, \mathcal{PT})$, where the superscript J (E) represents the current driven dissipative (electric-field driven nondissipative) part. The superscripts S and A represent the symmetric and antisymmetric components of the linear-response tensor, respectively. The symbol "=" represents that there are no additional contributions from that in the presence of both spatial inversion and time-reversal symmetries. Note that we restrict our discussion only to the electronic contributions in the thermal conductivity although phonons usually contribute to it as well. The Seebeck coefficient corresponds to the symmetric components of β_{ij}.

tensor	type	$(\mathcal{P},\mathcal{T},\mathcal{PT}) = (\bigcirc,\bigcirc,\bigcirc)$	$(\bigcirc,\times,\times)$	$(\times,\bigcirc,\times)$	$(\times,\times,\bigcirc)$	$(\times,\times,\times)$
electric conductivity	polar	$\sigma_{ij}^{(\mathrm{J,S})}$	$+\sigma_{ij}^{(\mathrm{E,A})}$	=	=	both
thermoelectric conductivity [127–129]	polar	$\beta_{ij}^{(\mathrm{J})}$	$+\beta_{ij}^{(\mathrm{E})}$	=	=	both
thermal conductivity [127–130]	polar	$\kappa_{ij}^{(\mathrm{J})}$	$+\kappa_{ij}^{(\mathrm{E})}$	=	=	both
magnetoelectric(current) tensor	axial	–	–	$\alpha_{ij}^{(\mathrm{J})}$	$\alpha_{ij}^{(\mathrm{E})}$	both
symmetric (S) components		$Q_0,\ Q_{ij}$	$T_0,\ T_{ij}$	$G_0,\ G_{ij}$	$M_0,\ M_{ij}$	
antisymmetric (A) components		G_i	M_i	Q_i	T_i	

(M_x^{ext}) with the amplitude H_x and the E dipole moment lies in the z direction (Q_z^{ext}) with the amplitude of E_z. In this case, the Hamiltonian is written as

$$\mathcal{H} = \begin{pmatrix} \frac{\hbar^2 \boldsymbol{k}^2}{2m} & -E_z(k_y + ik_x) + H_x \\ -E_z(k_y - ik_x) + H_x & \frac{\hbar^2 \boldsymbol{k}^2}{2m} \end{pmatrix}, \quad (41)$$

where the eigenvalues are easily obtained as

$$\varepsilon_\sigma(\boldsymbol{k}) = \frac{\hbar^2 \boldsymbol{k}^2}{2m} \pm \sqrt{E_z^2\left(k_x^2 + k_y^2\right) + H_x^2 - 2E_z H_x k_y}. \quad (42)$$

The obtained band dispersions clearly show a shift of the band bottom along the k_y direction when $E_z H_x \neq 0$, which indicates the emergence of the MT dipole along the y direction $\boldsymbol{T}^{\mathrm{ext}} \equiv \boldsymbol{Q}^{\mathrm{ext}} \times \boldsymbol{M}^{\mathrm{ext}} = E_z H_x \boldsymbol{e}_y$, where $\boldsymbol{e}_y$ is the unit vector along the y direction. Note that there is no spin degeneracy in this situation because the symmetry of the product $\mathcal{PT}$ is broken. This result is also confirmed by the multipole-operator aspect in momentum space, since the direct product of $M_x \propto \sigma_x$ and $Q_z \propto k_x\sigma_y - k_y\sigma_x$, which are given in Tables VI and VII respectively, leads to the functional form of the MT dipole as $\{\sigma_x, k_x\sigma_y - k_y\sigma_x\} = -2k_y\sigma_0 \propto T_y$. Similar discussions can be straightforwardly extended to the antiferromagnets accompanied by spatial-inversion symmetry breaking. For example, the staggered-type antiferromagnetic orderings on the one-dimensional zigzag and two-dimensional honeycomb structures are regarded as the MT dipole and octupole orders, respectively [35,39,48,93].

C. Linear-response tensors

Next, we discuss the cross-correlated phenomena in terms of multipole degrees of freedom. We examine what types of linear responses are expected in the presence of multipoles on the basis of the Kubo formula. In the linear response $\langle \hat{A}_i \rangle = \sum_j \chi_{ij} F_j$, where F_j is an external field along the j direction ($i, j = x, y, z$), the coupling with the external perturbation is given by $\mathcal{H}_{\mathrm{ext}} = -\hat{C}_j F_j$, and $\hat{B}_j = d\hat{C}_j/dt$, the linear response tensor is generally written by

$$\chi_{ij}^{(2)} = -\frac{i}{V} \sum_{\boldsymbol{k},n,m} \frac{f_{n\boldsymbol{k}} - f_{m\boldsymbol{k}}}{\varepsilon_{n\boldsymbol{k}} - \varepsilon_{m\boldsymbol{k}}} \frac{\langle \psi_{n\boldsymbol{k}} | \hat{A}_i | \psi_{m\boldsymbol{k}} \rangle \langle \psi_{m\boldsymbol{k}} | \hat{B}_j | \psi_{n\boldsymbol{k}} \rangle}{\varepsilon_{n\boldsymbol{k}} - \varepsilon_{m\boldsymbol{k}} + i\gamma}, \quad (43)$$

where V is the system volume, γ is a broadening factor, $f_{m\boldsymbol{k}} = f(\varepsilon_{m\boldsymbol{k}})$ is the Fermi distribution function, and $\varepsilon_{m\boldsymbol{k}}$ and $\psi_{m\boldsymbol{k}}$ are the eigenvalue and eigenstate of the Hamiltonian. m and n are the band indices. This expression does not take account of the orbital motion due to the Lorentz force in the presence of the external magnetic field, which could be important for some response functions such as the normal Hall conductivity [125,126]. $\hat{A}_i$ and $\hat{B}_j$ are rank-1 tensor and Hermite operators and $\chi_{ij}^{(2)}$ is rank-2 tensor [the superscript (l) in χ_{ij} represents the rank-l tensor], although the following discussion is straightforwardly extended to higher-rank operators, such as the E quadrupole representing the elastic property, as discussed in Appendix D.

We decompose $\chi_{ij}^{(2)}$ in Eq. (43) into the energy degenerate (J: $\varepsilon_{n\boldsymbol{k}} = \varepsilon_{m\boldsymbol{k}}$) and nondegenerate (E: $\varepsilon_{n\boldsymbol{k}} \neq \varepsilon_{m\boldsymbol{k}}$) parts as

$$\chi_{ij}^{(2)} = \chi_{ij}^{(\mathrm{J})} + \chi_{ij}^{(\mathrm{E})},$$

$$\chi_{ij}^{(\mathrm{J})} = -\frac{\gamma}{V} \sum_{\boldsymbol{k}nm}^{=} \frac{f_{n\boldsymbol{k}} - f_{m\boldsymbol{k}}}{\varepsilon_{n\boldsymbol{k}} - \varepsilon_{m\boldsymbol{k}}} \frac{A_{i\boldsymbol{k}}^{nm} B_{j\boldsymbol{k}}^{mn}}{(\varepsilon_{n\boldsymbol{k}} - \varepsilon_{m\boldsymbol{k}})^2 + \gamma^2}, \quad (44)$$

$$\chi_{ij}^{(\mathrm{E})} = -\frac{i}{V} \sum_{\boldsymbol{k}nm}^{\neq} \frac{(f_{n\boldsymbol{k}} - f_{m\boldsymbol{k}}) A_{i\boldsymbol{k}}^{nm} B_{j\boldsymbol{k}}^{mn}}{(\varepsilon_{n\boldsymbol{k}} - \varepsilon_{m\boldsymbol{k}})^2 + \gamma^2}, \quad (45)$$

where $A_{i\boldsymbol{k}}^{nm} = \langle \psi_{n\boldsymbol{k}} | \hat{A}_i | \psi_{m\boldsymbol{k}} \rangle$ and $B_{j\boldsymbol{k}}^{mn} = \langle \psi_{m\boldsymbol{k}} | \hat{B}_j | \psi_{n\boldsymbol{k}} \rangle$, and $(f_{n\boldsymbol{k}} - f_{m\boldsymbol{k}})/(\varepsilon_{n\boldsymbol{k}} - \varepsilon_{m\boldsymbol{k}}) \to \partial f/\partial \varepsilon_{n\boldsymbol{k}}$. $\chi_{ij}^{(\mathrm{J})}$ is the dissipative (current driven) part, while $\chi_{ij}^{(\mathrm{E})}$ is the nondissipative (electric-field driven) part. As discussed in Appendix C [42], $\chi_{ij}^{(\mathrm{J})}$ and $\chi_{ij}^{(\mathrm{E})}$ have different transformation properties under the time-reversal operation, i.e., the latter (former) changes the sign when the time-reversal properties of $\hat{A}_i$ and $\hat{B}_j$ are the same (different). On the other hand, the spatial inversion changes the sign of $\chi_{ij}^{(\mathrm{J,E})}$ when the parities of $\hat{A}_i$ and $\hat{B}_j$ are different. These facts indicate that different multipoles contribute to $\chi_{ij}^{(\mathrm{J})}$ and $\chi_{ij}^{(\mathrm{E})}$. The relation between the 2nd-rank linear-response tensors and relevant multipoles is summarized in Table X.

The linear-response (rank-2) tensor $\chi_{ij}^{(2)}$ has nine independent components when $\hat{A}_i$ and $\hat{B}_j$ are rank-1 tensors. As will be discussed in Appendix D, each component is related with the multipole degree of freedom. $\chi_{ij}^{(2)}$ is spanned by the

multipoles in the following form:

$$\hat{\chi}^{(2)} = \begin{pmatrix} X_0 - X_u + X_v & X_{xy} + Y_z & X_{zx} - Y_y \\ X_{xy} - Y_z & X_0 - X_u - X_v & X_{yz} + Y_x \\ X_{zx} + Y_y & X_{yz} - Y_x & X_0 + 2X_u \end{pmatrix}, \tag{46}$$

where $X = Q$ or T (G or M) and $Y = G$ or M (Q or T) for the polar (axial) 2nd-rank tensor depending on their time-reversal property. Among 9 multipoles, rank-0 (X_0), rank-1 (Y_x, Y_y, Y_z), and rank-2 ($X_u, X_v, X_{yz}, X_{zx}, X_{xy}$) multipoles represent the isotropic component, and antisymmetric and symmetric traceless components, respectively.

Since $\chi_{ij}^{(\mathrm{J})}$ and $\chi_{ij}^{(\mathrm{E})}$ have definite parities under spatial inversion and time-reversal operations ($\chi_{ij}^{(\mathrm{J})}$ and $\chi_{ij}^{(\mathrm{E})}$ have opposite time-reversal property), the type of multipoles is determined so as to share the same parities. When the linear-response tensors $\chi_{ij}^{(\mathrm{J})}$ or $\chi_{ij}^{(\mathrm{E})}$ are both the time-reversal and spatial inversion even, i.e., time-reversal-even polar tensor, the relevant multipoles are the E monopole Q_0, ET dipole G_i, and E quadrupole Q_{ij}. When the time-reversal (spatial inversion) operation changes the sign of the linear-response tensors, i.e., time-reversal-odd polar tensor (time-reversal-even axial tensor), they are characterized by the MT monopole T_0, M dipole M_i, and MT quadrupole T_{ij} (ET monopole G_0, E dipole Q_i, and ET quadrupole G_{ij}), which become finite in the absence of the time-reversal (spatial inversion) symmetry. Moreover, the M monopole M_0, MT dipole T_i, and M quadrupole M_{ij} contribute to the time-reversal-odd axial tensors, and it can be finite in the case where there are no time-reversal and spatial inversion symmetries while their product is preserved. In the following, we discuss two fundamental examples of the linear-response tensors: one is the electrical conductivity tensor and the other is the magnetoelectric (current) tensor. We also discuss the other linear-response tensors, such as the spin conductivity tensor and piezoelectric (current) tensor in Appendix D.

1. Electric conductivity tensor

First, we consider the polar second-rank electric conductivity,

$$\boldsymbol{J} = \hat{\sigma}\,\boldsymbol{E} = (\hat{\sigma}^{(\mathrm{J})} + \hat{\sigma}^{(\mathrm{E})})\boldsymbol{E}, \tag{47}$$

namely, we adopt $\hat{A}_i = \hat{B}_i = \hat{J}_i$ in the general formula (43). As the electric current operator $\hat{J}_i$ is both time-reversal and spatial-inversion odd, the polar electric conductivity tensor $\hat{\sigma}^{(\mathrm{E})}$ (electric-field driven nondissipative part) becomes nonzero only when the time-reversal symmetry is broken, while there is no symmetry restriction for the current driven dissipative part $\hat{\sigma}^{(\mathrm{J})}$. Meanwhile, from Eqs. (45) and (44), $\hat{\sigma}^{(\mathrm{E})}$ is the antisymmetric tensor $\sigma_{ij}^{(\mathrm{E})} = -\sigma_{ji}^{(\mathrm{E})} \equiv \sigma_{ij}^{(\mathrm{E,A})}$ and $\hat{\sigma}^{(\mathrm{J})}$ is the symmetric tensor $\sigma_{ij}^{(\mathrm{J})} = \sigma_{ji}^{(\mathrm{J})} \equiv \sigma_{ij}^{(\mathrm{J,S})}$. According to these facts, the corresponding multipole degrees of freedom for $\hat{\sigma}^{(\mathrm{J,S})}$ and $\hat{\sigma}^{(\mathrm{E,A})}$ are identified as

$$\hat{\sigma}^{(\mathrm{J,S})} = \begin{pmatrix} Q_0 - Q_u + Q_v & Q_{xy} & Q_{zx} \\ Q_{xy} & Q_0 - Q_u - Q_v & Q_{yz} \\ Q_{zx} & Q_{yz} & Q_0 + 2Q_u \end{pmatrix},$$
$$\hat{\sigma}^{(\mathrm{E,A})} = \begin{pmatrix} 0 & M_z & -M_y \\ -M_z & 0 & M_x \\ M_y & -M_x & 0 \end{pmatrix}. \tag{48}$$

The result shows that the anomalous Hall conductivity corresponding to the antisymmetric nondissipative part becomes nonzero in the presence of the M dipole moment, and arises from the interband contribution.

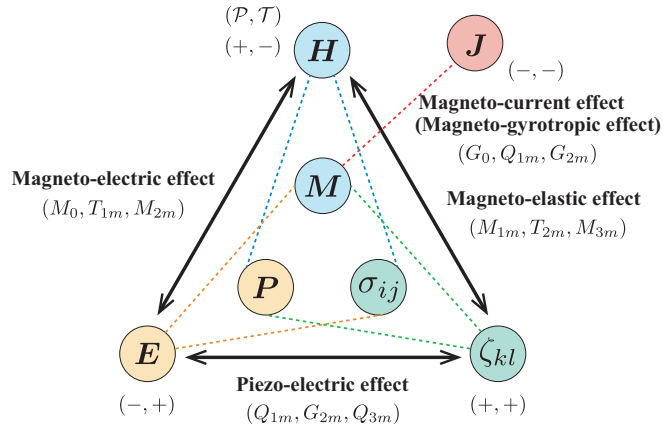

FIG. 3. Schematic picture of a revised Heckmann diagram showing the relation among electric, magnetic, and mechanical properties in solids. $\boldsymbol{E}$, $\boldsymbol{H}$, $\boldsymbol{J}$, and $\zeta_{kl} = \varepsilon_{kl} + \sum_m \epsilon_{klm}\omega_m$ are the external electric field, magnetic field, electric current, and strain-rotation field, while $\boldsymbol{P}$, $\boldsymbol{M}$, and σ_{ij} represent the electric polarization, magnetization, and stress tensor, respectively. The relevant multipoles in the linear-response tensor for each cross-correlated coupling are also shown.

2. Magnetoelectric(current) tensor

Let us consider the magnetoelectric (current) effect where the magnetization is induced by the electric field (current),

$$\boldsymbol{M} = \hat{\alpha}\,\boldsymbol{E} = (\hat{\alpha}^{(\mathrm{J})} + \hat{\alpha}^{(\mathrm{E})})\boldsymbol{E}. \tag{49}$$

The axial second-rank magnetoelectric (current) tensor $\hat{\alpha}$ is obtained by using $\hat{A}_i = \hat{M}_i$ and $\hat{B}_j = \hat{J}_j$ in Eq. (43). As $\hat{M}_i$ is time-reversal odd and spatial inversion even, the dissipative part $\hat{\alpha}^{(\mathrm{J})}$ becomes nonzero in the absence of only the spatial inversion symmetry. On the other hand, the nondissipative $\hat{\alpha}^{(\mathrm{E})}$ becomes nonzero only in the absence of both the time-reversal and spatial inversion symmetries. Thus $\hat{\alpha}^{(\mathrm{J})}$ and $\hat{\alpha}^{(\mathrm{E})}$ are identified as

$$\hat{\alpha}^{(\mathrm{J})} = \begin{pmatrix} G_0 - G_u + G_v & G_{xy} + Q_z & G_{zx} - Q_y \\ G_{xy} - Q_z & G_0 - G_u - G_v & G_{yz} + Q_x \\ G_{zx} + Q_y & G_{yz} - Q_x & G_0 + 2G_u \end{pmatrix},$$
$$\hat{\alpha}^{(\mathrm{E})} = \begin{pmatrix} M_0 - M_u + M_v & M_{xy} + T_z & M_{zx} - T_y \\ M_{xy} - T_z & M_0 - M_u - M_v & M_{yz} + T_x \\ M_{zx} + T_y & M_{yz} - T_x & M_0 + 2M_u \end{pmatrix}. \tag{50}$$

TABLE XI. Finite CEF parameters in terms of the tesseral harmonics under the point group except for triclinic crystals (C_1, C_i), where (lm), $(lm)'$, and (l) means the presence of $O_{lm}^{(c)}$, $O_{lm}^{(s)}$, and O_l (see Table I for the expressions of O_l). The checkmark in the columns N, P, C, and G represents the noncentrosymmetric (no inversion operation), polar (presence of rank-1 CEFs, see also rank-1 multipoles in Table XVI), chiral (no improper rotations, $\mathcal{PC}_n$), and gyrotropic (presence of less than rank-2 odd-parity multipoles in Table XVI, i.e., presence of χ_{ij}^{A}) point groups, respectively. We take the x axis as the C_2' rotation axis. For C_{3v}, we take the zx plane as the σ_v mirror plane. For monoclinic crystals, the standard orientation is taken.

crystal system	point group	N	P	C	G	even-parity CEF	odd-parity CEF
cubic	O_h					(4), (6)	–
	O	✓		✓	✓	(4), (6)	–
	T_d	✓				(4), (6)	$(32)'$
	T	✓		✓	✓	(4), (6), $(6t)$	$(32)'$
	T_h					(4), (6), $(6t)$	–
tetragonal	D_{4h}					(20), (40), (44), (60), (64)	–
	D_4	✓		✓	✓	(20), (40), (44), (60), (64)	$(54)'$
	D_{2d}	✓			✓	(20), (40), (44), (60), (64)	$(32)'$, $(52)'$
	C_{4v}	✓	✓		✓	(20), (40), (44), (60), (64)	(10), (30), (50), (54)
	C_{4h}					(20), (40), (44), (60), (64), $(44)'$, $(64)'$	–
	C_4	✓	✓	✓	✓	(20), (40), (44), (60), (64), $(44)'$, $(64)'$	(10), (30), (50), (54), $(54)'$
	S_4	✓			✓	(20), (40), (44), (60), (64), $(44)'$, $(64)'$	(32), (52), $(32)'$, $(52)'$
orthorhombic	D_{2h}					(20), (22), (40), (42), (44), (60), (62), (64), (66)	–
	D_2	✓		✓	✓	(20), (22), (40), (42), (44), (60), (62), (64), (66)	$(32)'$, $(52)'$, $(54)'$
	C_{2v}	✓	✓		✓	(20), (22), (40), (42), (44), (60), (62), (64), (66)	(10), (30), (32), (50), (52), (54)
monoclinic	C_{2h}					(20), (22), (40), (42), (44), (60), (62), (64), (66) $(22)'$, $(42)'$, $(44)'$, $(62)'$, $(64)'$, $(66)'$	–
	C_2	✓	✓	✓	✓	(20), (22), (40), (42), (44), (60), (62), (64), (66) $(22)'$, $(42)'$, $(44)'$, $(62)'$, $(64)'$, $(66)'$	(10), (30), (32), (50), (52), (54) $(32)'$, $(52)'$, $(54)'$
	C_s	✓	✓		✓	(20), (22), (40), (42), (44), (60), (62), (64), (66) $(22)'$, $(42)'$, $(44)'$, $(62)'$, $(64)'$, $(66)'$	(11), (31), (33), (51), (53), (55) $(11)'$, $(31)'$, $(33)'$, $(51)'$, $(53)'$, $(55)'$
hexagonal	D_{6h}					(20), (40), (60), (66)	–
	D_6	✓		✓	✓	(20), (40), (60), (66)	–
	D_{3h}	✓				(20), (40), (60), (66)	(33), (53)
	C_{6v}	✓	✓		✓	(20), (40), (60), (66)	(10), (30), (50)
	C_{6h}					(20), (40), (60), (66), $(66)'$	–
	C_6	✓	✓	✓	✓	(20), (40), (60), (66), $(66)'$	(10), (30), (50)
	C_{3h}	✓				(20), (40), (60), (66), $(66)'$	(33), (53), $(33)'$, $(53)'$
trigonal	D_{3d}					(20), (40), (60), (66), $(43)'$, $(63)'$	–
	D_3	✓		✓	✓	(20), (40), (60), (66), $(43)'$, $(63)'$	(33), (53)
	C_{3v}	✓	✓		✓	(20), (40), (43), (60), (63), (66)	(10), (30), (33), (50), (53)
	C_{3i}					(20), (40), (43), (60), (63), (66), $(43)'$, $(63)'$, $(66)'$	–
	C_3	✓	✓	✓	✓	(20), (40), (43), (60), (63), (66), $(43)'$, $(63)'$, $(66)'$	(10), (30), (33), (50), (53), $(33)'$, $(53)'$

The isotropic longitudinal magnetoelectric (current) response is realized in the presence of the M (ET) monopole, the antisymmetric transverse response in the presence of the MT (E) dipole, and the symmetric transverse and traceless longitudinal responses in the presence of the M (ET) quadrupoles.

The nondissipative $\hat{\alpha}^{(E)}$ originates from the nondegenerate (interband) contributions, while the dissipative $\hat{\alpha}^{(J)}$ mainly arises from the intraband contributions. In other words, $\hat{\alpha}^{(E)}$ plays an important role in insulating systems, while $\hat{\alpha}^{(J)}$ becomes important for metallic systems. Especially, in the presence of $\mathcal{PT}$ symmetry, the latter contribution is forbidden. From the above considerations, the former tensor is called magnetoelectric tensor and the latter tensor is called magnetocurrent (magnetogyrotropic) tensor, which has essentially the same origin as the so-call Edelstein effect [131,132]. The relations between the cross-correlated responses including the piezoelectric effect and the relevant multipoles are summarized in Fig. 3.

IV. POINT-GROUP IRREDUCIBLE REPRESENTATIONS

In the crystal systems, the rotational symmetry and inversion symmetry in some cases, are lost. In such cases, the components of the same rank (the irreducible representation of the rotational group) split into subgroups according to the point-group irreducible representation. In this section, we

TABLE XII. Multipoles under cubic, tetragonal, orthorhombic, and monoclinic crystals. The upper and lower columns represent even-parity and odd-parity multipoles, respectively. We take the x axis as the C_2' rotation axis.

E	ET	M	MT	$O_{\rm h}$	O	$T_{\rm d}$	$T_{\rm h}$	T	$D_{4\rm h}$	D_4	$C_{4\rm h}$	$D_{2\rm d}$	$C_{4\rm v}$	C_4	S_4	$D_{2\rm h}$	D_2	$C_{2\rm v}$	$C_{2\rm h}$	C_2	$C_{\rm s}$
$Q_0,\ Q_4$	–	–	$T_0,\ T_4$	A_{1g}	A_1	A_1	A_g	A	A_{1g}	A_1	A_g	A_1	A_1	A	A	A_g	A	A_1	A_g	A	A′
–	G_{xyz}	M_{xyz}	–	A_{2g}	A_2	A_2	A_g	A	B_{1g}	B_1	B_g	B_1	B_1	B	B	A_g	A	A_1	A_g	A	A′
$Q_u,\ Q_{4u}$	–	–	$T_u,\ T_{4u}$	E_g	E	E	E_g	E	A_{1g}	A_1	A_g	A_1	A_1	A	A	A_g	A	A_1	A_g	A	A′
$Q_v,\ Q_{4v}$	–	–	$T_v,\ T_{4v}$						B_{1g}	B_1	B_g	B_1	B_1	B	B	A_g	A	A_1	A_g	A	A′
Q^α_{4x}	$G_x,\ G^\alpha_x$	$M_x,\ M^\alpha_x$	T^α_{4x}	T_{1g}	T_1	T_1	T_g	T	E_g	E	E_g	E	E	E	E	B_{3g}	B_3	B_2	B_g	B	A″
Q^α_{4y}	$G_y,\ G^\alpha_y$	$M_y,\ M^\alpha_y$	T^α_{4y}													B_{2g}	B_2	B_1	B_g	B	A″
Q^α_{4z}	$G_z,\ G^\alpha_z$	$M_z,\ M^\alpha_z$	T^α_{4z}						A_{2g}	A_2	A_g	A_2	A_2	A	A	B_{1g}	B_1	A_2	A_g	A	A′
$Q_{yz},\ Q^\beta_{4x}$	G^β_x	M^β_x	$T_{yz},\ T^\beta_{4x}$	T_{2g}	T_2	T_2	T_g	T	E_g	E	E_g	E	E	E	E	B_{3g}	B_3	B_2	B_g	B	A″
$Q_{zx},\ Q^\beta_{4y}$	G^β_y	M^β_y	$T_{zx},\ T^\beta_{4y}$													B_{2g}	B_2	B_1	B_g	B	A″
$Q_{xy},\ Q^\beta_{4z}$	G^β_z	M^β_z	$T_{xy},\ T^\beta_{4z}$						B_{2g}	B_2	B_g	B_2	B_2	B	B	B_{1g}	B_1	A_2	A_g	A	A′
–	$G_0,\ G_4$	$M_0,\ M_4$	–	A_{1u}	A_1	A_2	A_u	A	A_{1u}	A_1	A_u	B_1	A_2	A	B	A_u	A	A_2	A_u	A	A″
Q_{xyz}	–	–	T_{xyz}	A_{2u}	A_2	A_1	A_u	A	B_{1u}	B_1	B_u	A_1	B_2	B	A	A_u	A	A_2	A_u	A	A″
–	$G_u,\ G_{4u}$	$M_u,\ M_{4u}$	–	E_u	E	E	E_u	E	A_{1u}	A_1	A_u	B_1	A_2	A	B	A_u	A	A_2	A_u	A	A″
–	$G_v,\ G_{4v}$	$M_v,\ M_{4v}$	–						B_{1u}	B_1	B_u	A_1	B_2	B	A	A_u	A	A_2	A_u	A	A″
$Q_x,\ Q^\alpha_x$	G^α_{4x}	M^α_{4x}	$T_x,\ T^\alpha_x$	T_{1u}	T_1	T_2	T_u	T	E_u	E	E_u	E	E	E	E	B_{3u}	B_3	B_1	B_u	B	A′
$Q_y,\ Q^\alpha_y$	G^α_{4y}	M^α_{4y}	$T_y,\ T^\alpha_y$													B_{2u}	B_2	B_2	B_u	B	A′
$Q_z,\ Q^\alpha_z$	G^α_{4z}	M^α_{4z}	$T_z,\ T^\alpha_z$						A_{2u}	A_2	A_u	B_2	A_1	A	B	B_{1u}	B_1	A_1	A_u	A	A″
Q^β_x	$G_{yz},\ G^\beta_{4x}$	$M_{yz},\ M^\beta_{4x}$	T^β_x	T_{2u}	T_2	T_1	T_u	T	E_u	E	E_u	E	E	E	E	B_{3u}	B_3	B_1	B_u	B	A′
Q^β_y	$G_{zx},\ G^\beta_{4y}$	$M_{zx},\ M^\beta_{4y}$	T^β_y													B_{2u}	B_2	B_2	B_u	B	A′
Q^β_z	$G_{xy},\ G^\beta_{4z}$	$M_{xy},\ M^\beta_{4z}$	T^β_z						B_{2u}	B_2	B_u	A_2	B_1	B	A	B_{1u}	B_1	A_1	A_u	A	A″

discuss such a reduction for multipoles. First, we discuss the CEF potential under all the point groups in Sec. IV A, which is nothing but the sum of the E multipoles in the totally symmetric A^+_{1g} representation. Then, we show how to classify multipoles under point-group irreducible representations in Sec. IV B. Such analyses offer a microscopic investigation of potential active multipole degrees of freedom in solids. We show several examples by considering specific basis functions in the tetragonal $D_{2\rm d}$ group.

Similar analysis can be applied to any point-group symmetry. For such purpose, we provide various tables for the parent cubic $O_{\rm h}$ group in the main text and the hexagonal $D_{6\rm h}$ group in Appendix F, and the compatible relations between these parent groups and subgroups in Tables XII and XXVIII.

A. CEF potential

Under the point-group symmetry, some components of the E multipoles reduce to the totally symmetric A^+_{1g} representation. These components constitute the CEF potentials. Therefore the CEF potential is represented by

$$V(\boldsymbol{r}) = \sum_{lm}^{\in A^+_{1g}} c_{lm} O_{lm}(\boldsymbol{r}), \tag{51}$$

where c_{lm} is the CEF parameter. Since we consider the largest orbital-angular momentum $L = 3$ corresponding to the f orbital in the one-electron state, going up to the rank $l = 2L = 6$ is sufficient in the summation. The even-parity (even-rank) CEF leads to even-parity hybridizations such as s-d and p-f orbitals, while the odd-parity (odd-rank) CEF leads to odd-parity hybridizations for s-p, p-d, d-f, and s-f orbitals at the same site. Note that the odd-parity CEF is present only in the lack of inversion center at lattice sites.

The nonzero CEF parameters under each point group except for triclinic crystals (C_1, C_i) are summarized in Table XI, which is constructed by reading the E multipoles belonging to A^+_{1g} representation in the reduction rules in Tables XII and XXVIII in Appendix F. The relevant harmonics are given in Tables I, II, XXII, and XXIII.

Now, let us consider a typical example by considering the system with the p-d hybridized orbitals under the $D_{2\rm d}$ group. From Table XI, the atomic CEF Hamiltonian is represented by

$$\mathcal{H}_{\rm CEF} = \mathcal{H}^{\rm even}_{\rm CEF} + \mathcal{H}^{\rm odd}_{\rm CEF} + \mathcal{H}_\Delta, \tag{52}$$

$$\mathcal{H}^{\rm even}_{\rm CEF} = c_{20} O^{\rm (c)}_{20} + c_{40} O^{\rm (c)}_{40} + c_{44} O^{\rm (c)}_{44}, \tag{53}$$

$$\mathcal{H}^{\rm odd}_{\rm CEF} = c'_{32} O^{\rm (s)}_{32}. \tag{54}$$

Note that c_{60}, c_{64}, and c'_{52} become zero for the p-d hybridized systems, as the maximum rank is given by $2\max(L_1 = 1, L_2 = 2) = 4$. $\mathcal{H}^{\rm even}_{\rm CEF}$ represents the even-parity CEF Hamiltonian, which splits three p orbitals (ϕ_x, ϕ_y, ϕ_z) into two orbitals (ϕ_x, ϕ_y) with the irreducible representation E and an orbital ϕ_z with the representation B_2; the five d orbitals $(\phi_u, \phi_v, \phi_{yz}, \phi_{zx}, \phi_{xy})$ split into three single orbitals ϕ_u with A_1, ϕ_v with B_1, and ϕ_{xy} with B_2, and two orbitals (ϕ_{yz}, ϕ_{zx}) with E. Meanwhile, $\mathcal{H}^{\rm odd}_{\rm CEF}$ leads to the odd-parity hybridization between p and d orbitals belonging to the same irreducible representation. In the present case, ϕ_z hybridizes with ϕ_{xy}, and (ϕ_x, ϕ_y) hybridize with (ϕ_{yz}, ϕ_{zx}). $\mathcal{H}_\Delta$ represents the atomic energy level for p and d orbitals where the energy difference is taken as $\Delta = \varepsilon_p - \varepsilon_d$.

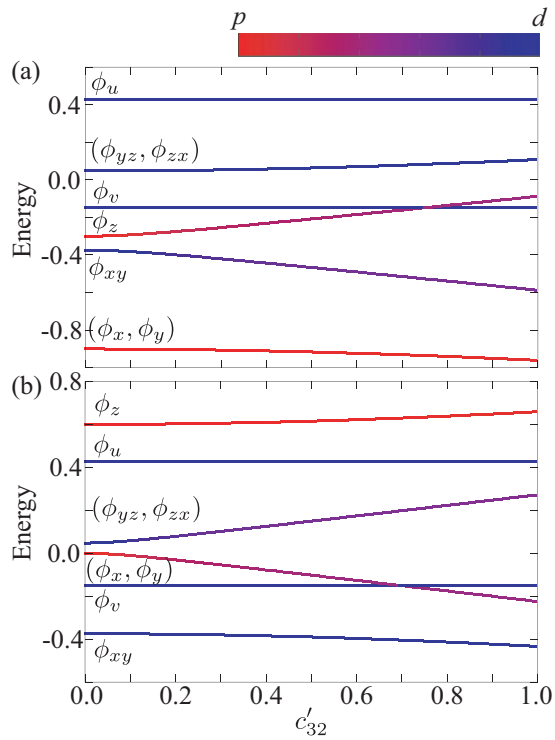

FIG. 4. Odd-parity CEF dependencies of the atomic energy level for p-d hybrid orbitals under the tetragonal D_{2d} group. The colors show the weights for p and d orbitals. The difference of the atomic levels for p and d orbitals is taken as (a) $\Delta = -0.7$ and (b) $\Delta = 0.2$, and the CEF parameters are $c_{20} = 1$, $c_{40} = 0.5$, and $c_{44} = 0.4$.

The CEF levels in Eq. (52) are shown in Fig. 4, which largely depends on the model parameters. Strong odd-parity hybridization between ϕ_z and ϕ_{xy} is expected for $\Delta = -0.7$ in Fig. 4(a), while that between (ϕ_x, ϕ_y) and (ϕ_{yz}, ϕ_{zx}) occurs for $\Delta = 0.2$ in Fig. 4(b).

B. Active multipoles under point groups

When a CEF splitting is large and one of CEF multiplets dominates the low-energy physics, the point-group irreducible representation is suitable to classify multipole moments. The classification is done by using the reduction rules, which are summarized in Tables XII and XXVIII in Appendix F for 32 point groups in seven crystal systems.

Tables XII and XXVIII are useful to investigate what type of multipoles are activated in the specific crystal structures, which stimulate microscopic understanding of physical phenomena induced by multipoles, as discussed in Sec. III. For example, the magnetocurrent effect, where the uniform magnetization is induced by electric current as was discussed in Sec. III C, can occur in the point groups $(O, T, D_4, C_4, D_6, C_6, D_3, C_3, D_2, D_{2d}, C_{4v}, S_4, C_2, C_s, C_{6v}, C_{3v}, C_{2v}, C_1)$, which is the so-called gyrotropic point groups. This is because any of the relevant multipoles for the magnetocurrent effect [G_0, (Q_x, Q_y, Q_z), and $(G_u, G_v, G_{yz}, G_{zx}, G_{xy})$] belong to the totally symmetric representation in the gyrotropic point groups.

Moreover, Tables XII and XXVIII show what multipoles are potential order parameters in the systems. Note that as the E (M) and ET (MT) multipoles belong to different irreducible representations in some point groups, the distinction of them should be essential in such point groups.

Let us demonstrate how to identify active multipole degrees of freedom in the low-energy multiplets by using the p-d hybridized CEF states in the tetragonal D_{2d} group as was discussed in the previous subsection. First, we consider the basis functions (ϕ_{yz}, ϕ_{zx}) in the representation E under the D_{4h} group, which is the parent group of D_{2d} and does not show orbital hybridization with different parities ($c'_{32} = 0$). By taking the direct product of the basis functions, the active multipole degrees of freedom is obtained as

$$\mathrm{E}_g \otimes \mathrm{E}_g = \mathrm{A}^+_{1g} \oplus \mathrm{A}^-_{2g} \oplus \mathrm{B}^+_{1g} \oplus \mathrm{B}^+_{2g}, \tag{55}$$

TABLE XIII. Multipoles under the tetragonal crystal system (D_{4h} and related groups) in the spinless basis. We introduce the abbreviation $(g,u)^\pm = \mathrm{A}^\pm_{1g,u} \oplus \mathrm{A}^\mp_{2g,u} \oplus \mathrm{B}^\pm_{1g,u} \oplus \mathrm{B}^\pm_{2g,u}$. The subscripts g, u are omitted for the D_4 group. The subscripts 1,2 are omitted for the C_{4h} group. $\mathrm{A}_{1u} \leftrightarrow \mathrm{B}_{1u}$ and $\mathrm{A}_{2u} \leftrightarrow \mathrm{B}_{2u}$, and then, the subscripts g, u are omitted for the D_{2d} group. $\mathrm{A}_{1u} \leftrightarrow \mathrm{B}_{1u}$ and $\mathrm{A}_{2u} \leftrightarrow \mathrm{B}_{2u}$, and then, the subscripts g, u and 1,2 are omitted for the S_4 group. $\mathrm{A}_{1u} \leftrightarrow \mathrm{A}_{2u}$ and $\mathrm{B}_{1u} \leftrightarrow \mathrm{B}_{2u}$, and then, the subscripts g, u are omitted for the C_{4v} group. $\mathrm{A}_{1u} \leftrightarrow \mathrm{A}_{2u}$ and $\mathrm{B}_{1u} \leftrightarrow \mathrm{B}_{2u}$, and then, the subscripts g, u and 1,2 are omitted for the C_4 group.

	(s) A_{1g}	(p) A_{2u}	E_u	(d) A_{1g}	B_{1g}	B_{2g}	E_g	(f) B_{1u}	A_{2u}	E_u	B_{2u}	E_u
(s) A_{1g}	A^+_{1g}	$\mathrm{A}^\pm_{2u}$	$\mathrm{E}^\pm_u$	$\mathrm{A}^\pm_{1g}$	$\mathrm{B}^\pm_{1g}$	$\mathrm{B}^\pm_{2g}$	$\mathrm{E}^\pm_g$	$\mathrm{B}^\pm_{1u}$	$\mathrm{A}^\pm_{2u}$	$\mathrm{E}^\pm_u$	$\mathrm{B}^\pm_{2u}$	$\mathrm{E}^\pm_u$
(p) A_{2u}		A^+_{1g}	$\mathrm{E}^\pm_g$	$\mathrm{A}^\pm_{2u}$	$\mathrm{B}^\pm_{2u}$	$\mathrm{B}^\pm_{1u}$	$\mathrm{E}^\pm_u$	$\mathrm{B}^\pm_{2g}$	$\mathrm{A}^\pm_{1g}$	$E^\pm_g$	$\mathrm{B}^\pm_{1g}$	$\mathrm{E}^\pm_g$
E_u			$(g)^+$	$\mathrm{E}^\pm_u$	$\mathrm{E}^\pm_u$	$\mathrm{E}^\pm_u$	$(u)^\pm$	$\mathrm{E}^\pm_g$	$\mathrm{E}^\pm_g$	$(g)^\pm$	$\mathrm{E}^\pm_g$	$(g)^\pm$
(d) A_{1g}				A^+_{1g}	$\mathrm{B}^\pm_{1g}$	$\mathrm{B}^\pm_{2g}$	$\mathrm{E}^\pm_g$	$\mathrm{B}^\pm_{1u}$	$\mathrm{A}^\pm_{2u}$	$\mathrm{E}^\pm_u$	$\mathrm{B}^\pm_{2u}$	$\mathrm{E}^\pm_u$
B_{1g}					A^+_{1g}	$\mathrm{A}^\pm_{2g}$	$\mathrm{E}^\pm_g$	$\mathrm{A}^\pm_{1u}$	$\mathrm{B}^\pm_{2u}$	$\mathrm{E}^\pm_u$	$\mathrm{A}^\pm_{2u}$	$\mathrm{E}^\pm_u$
B_{2g}						A^+_{1g}	$\mathrm{E}^\pm_g$	$\mathrm{A}^\pm_{2u}$	$\mathrm{B}^\pm_{1u}$	$\mathrm{E}^\pm_u$	$\mathrm{A}^\pm_{1u}$	$\mathrm{E}^\pm_u$
E_g							$(g)^+$	$\mathrm{E}^\pm_u$	$\mathrm{E}^\pm_u$	$(u)^\pm$	$\mathrm{E}^\pm_u$	$(u)^\pm$
(f) B_{1u}								A^+_{1g}	$\mathrm{B}^\pm_{2g}$	$\mathrm{E}^\pm_g$	$\mathrm{A}^\pm_{2g}$	$\mathrm{E}^\pm_g$
A_{2u}									A^+_{1g}	$\mathrm{E}^\pm_g$	$\mathrm{B}^\pm_{1g}$	$\mathrm{E}^\pm_g$
E_u										$(g)^+$	$\mathrm{E}^\pm_g$	$(g)^\pm$
B_{2u}											A^+_{1g}	$\mathrm{E}^\pm_g$
E_u												$(g)^+$

TABLE XIV. Multipoles under the tetragonal crystal system (D_{4h} and related groups) in the spinful basis. We introduce the abbreviation $(g,u)^{\pm} = A^{\pm}_{1g,u} \oplus A^{\mp}_{2g,u} \oplus E^{\mp}_{g,u}$, and $\langle g,u\rangle^{\pm} = B^{\pm}_{1g,u} \oplus B^{\pm}_{2g,u} \oplus E^{\pm}_{g,u}$. The subscripts g,u are omitted for the D_4 group. The subscripts 1,2 are omitted for the C_{4h} group. $A_{1u} \leftrightarrow B_{1u}$, $A_{2u} \leftrightarrow B_{2u}$, and $E_{1/2u} \leftrightarrow E_{3/2u}$, and then, the subscripts g,u are omitted for the D_{2d} group. $A_{1u} \leftrightarrow B_{1u}$, $A_{2u} \leftrightarrow B_{2u}$, and $E_{1/2u} \leftrightarrow E_{3/2u}$, and then, the subscripts g,u and 1,2 are omitted for the S_4 group. $A_{1u} \leftrightarrow A_{2u}$ and $B_{1u} \leftrightarrow B_{2u}$, and then, the subscripts g,u are omitted for the C_{4v} group. $A_{1u} \leftrightarrow A_{2u}$ and $B_{1u} \leftrightarrow B_{2u}$, and then, the subscripts g,u and 1,2 are omitted for the C_4 group.

		(s) $E_{1/2g}$	(p) $E_{1/2u}$	$E_{1/2u}$	$E_{3/2u}$	(d) $E_{1/2g}$	$E_{3/2g}$	$E_{3/2g}$	$E_{1/2g}$	$E_{3/2g}$	(f) $E_{3/2u}$	$E_{1/2u}$	$E_{1/2u}$	$E_{3/2u}$	$E_{3/2u}$	$E_{1/2u}$	$E_{3/2u}$
(s) $A_{1g} \otimes E_{1/2g} \rightarrow$	$E_{1/2g}$	$(g)^+$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$
(p) $A_{2u} \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$		$(g)^+$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$
$E_u \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$			$(g)^+$	$\langle g\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$
	$E_{3/2u}$				$(g)^+$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$
(d) $A_{1g} \otimes E_{1/2g} \rightarrow$	$E_{1/2g}$					$(g)^+$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$
$B_{1g} \otimes E_{1/2g} \rightarrow$	$E_{3/2g}$						$(g)^+$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$
$B_{2g} \otimes E_{1/2g} \rightarrow$	$E_{3/2g}$							$(g)^+$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$
$E_g \otimes E_{1/2g} \rightarrow$	$E_{1/2g}$								$(g)^+$	$\langle g\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$
	$E_{3/2g}$									$(g)^+$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$
(f) $B_{1u} \otimes E_{1/2g} \rightarrow$	$E_{3/2u}$										$(g)^+$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$
$A_{2u} \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$											$(g)^+$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$
$E_u \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$												$(g)^+$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$
	$E_{3/2u}$													$(g)^+$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$
$B_{2u} \otimes E_{1/2g} \rightarrow$	$E_{3/2u}$														$(g)^+$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$
$E_u \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$															$(g)^+$	$\langle g\rangle^{\pm}$
	$E_{3/2u}$																$(g)^+$

where the superscripts $+$ $(-)$ represent time-reversal even (odd), and all the multipoles are even-parity (the subscript g). Note that the symmetrized (antisymmetrized) product of bases corresponds to time-reversal even(odd) operators when the basis functions are spinless (the angular momentum is an integer). On the other hand, the symmetrized (antisymmetrized) product of the basis functions correspond to time-reversal odd(even) operators when the basis functions are spinful (the angular momentum is a half integer). The above irreducible decomposition for the spinless basis in the tetragonal crystal system is summarized in Table XIII.

The corresponding multipoles are given by

$$A^+_{1g}: \quad \hat{\rho}_0 \leftrightarrow Q_0, \tag{56}$$

$$A^-_{2g}: \quad \hat{\rho}_y \leftrightarrow M_z, \tag{57}$$

$$B^+_{1g}: \quad \hat{\rho}_z \leftrightarrow Q_v, \tag{58}$$

$$B^+_{2g}: \quad \hat{\rho}_x \leftrightarrow Q_{xy}, \tag{59}$$

where $\hat{\rho}_0$ and $\hat{\boldsymbol{\rho}}$ are 2×2 unit and Pauli matrices for doubly degenerate orbitals (ϕ_{yz}, ϕ_{zx}), respectively. Obviously, only even-parity E and M multipoles are active in these CEF multiplets.

Next, let us consider the basis functions (ϕ_z, ϕ_{xy}) belonging to the representation B_2 under the D_{2d} group. The multipole degrees of freedom are obtained by decomposing the direct product of the basis functions (using Table XIII) as

$$(B_2 \oplus B_2) \otimes (B_2 \oplus B_2) = (2A^+_1)_{\text{intra}} \oplus (A^+_1 \oplus A^-_1)_{\text{inter}}, \tag{60}$$

where $(\dots)_{\text{intra}}$ and $(\dots)_{\text{inter}}$ represent the multipoles activated in nonhybrid and hybrid orbitals, respectively. A set of independent operators in the basis (ϕ_z, ϕ_{xy}) is given by

$$A^+_1: \quad \{\hat{\tau}_0, \hat{\tau}_z\} \leftrightarrow \{Q_0, Q_u\}, \quad \hat{\tau}_x \leftrightarrow G_v, \tag{61}$$

$$A^-_1: \quad \hat{\tau}_y \leftrightarrow M_v, \tag{62}$$

where $\hat{\tau}_0$ and $\hat{\boldsymbol{\tau}}$ are respectively a 2×2 unit matrix and Pauli matrices in the basis (ϕ_z, ϕ_{xy}), and $\{\dots\}$ represent the appropriate linear combination. τ_0 and τ_z represent the intraorbital degree of freedom, while τ_x and τ_y represent the interorbital degree of freedom.

The expected linear responses are uniquely found once the multipole degrees of freedom are identified. In this case, the magnetocurrent effect is induced by the emergence of the ET quadrupole $G_v \propto k_x\sigma_x - k_y\sigma_y$; the induced magnetization M_x by the electric current in the x direction and M_y by the electric current in the y direction should have opposite sign and the same magnitude. The result implies that a larger magnetocurrent response is expected for larger ET quadrupole G_v. In a similar way, the magnetoelectric effect is anticipated once the time-reversal symmetry breaking occurs, as the antisymmetrized representation is the M quadrupole M_v.

In the spinful basis, possible multipoles are obtained by changing the irreducible representation of the basis function as $B_2 \otimes E_{1/2} \rightarrow E_{3/2}$, and the decomposition according to Table XIV gives

$$\begin{aligned}(E_{3/2} \oplus E_{3/2}) \otimes (E_{3/2} \oplus E_{3/2}) &= (2A^+_1 \oplus 2A^-_2 \oplus 2E^-)_{\text{intra}} \\ &\oplus (A^+_1 \oplus A^+_2 \oplus E^+ \oplus A^-_1 \oplus A^-_2 \oplus E^-)_{\text{inter}}.\end{aligned} \tag{63}$$

TABLE XV. Even-parity multipoles belonging to a totally symmetric representation in the point groups except for the triclinic ones (C_1, C_i). $\chi^{\rm P}_{ij}$ and $\chi^{\rm A}_{ijk}$ represent rank-2 polar and rank-3 axial tensors, respectively. $\chi^{\rm P}_{ij}$ ($\chi^{(2)}_{ij}$) has 9 components and $\chi^{\rm A}_{ijk}$ has 27 components at most. The components of $\chi^{\rm A}_{ijk}$ are decomposed into a symmetric monopole response denoted by 1 with 3 components ($\chi^{(1)}_{i}$), antisymmetric dipole response denoted by 2 with nine components ($\chi^{(2)}_{ij}$), and symmetric quadrupole response denoted by 3 with 15 components ($\chi^{(3)}_{ijk}$), respectively. The independent number of components in $\chi^{\rm P}_{ij}$ and $\chi^{\rm A}_{ijk}$ is shown for each point group. Responses in the absence of time-reversal symmetry, such as the magnetoelectric effect and Nernst effect, are obtained by replacing (Q_{lm}, G_{lm}) with (T_{lm}, M_{lm}).

						totally symmetric rep.					$\chi^{\rm P}_{ij}$	$\chi^{\rm A}_{ijk}$		
crystal system	point group	N	P	C	G	rank 0	rank 1	rank 2	rank 3	rank 4	2	1	2	3
cubic	$O_{\rm h}$					Q_0				Q_4	1		1	
	O	✓		✓	✓	Q_0				Q_4	1		1	
	$T_{\rm d}$	✓				Q_0				Q_4	1		1	
	T	✓		✓	✓	Q_0			G_{xyz}	Q_4	1		1	1
	$T_{\rm h}$					Q_0			G_{xyz}	Q_4	1		1	1
tetragonal	$D_{4\rm h}$					Q_0		Q_u		$Q_4,\ Q_{4u}$	2		2	1
	D_4	✓		✓	✓	Q_0		Q_u		$Q_4,\ Q_{4u}$	2		2	1
	$D_{2\rm d}$	✓			✓	Q_0		Q_u		$Q_4,\ Q_{4u}$	2		2	1
	$C_{4\rm v}$	✓	✓		✓	Q_0		Q_u		$Q_4,\ Q_{4u}$	2		2	1
	$C_{4\rm h}$					Q_0	G_z	Q_u	G^α_z	$Q_4,\ Q_{4u},\ Q^\alpha_{4z}$	3	1	3	3
	C_4	✓	✓	✓	✓	Q_0	G_z	Q_u	G^α_z	$Q_4,\ Q_{4u},\ Q^\alpha_{4z}$	3	1	3	3
	S_4	✓			✓	Q_0	G_z	Q_u	G^α_z	$Q_4,\ Q_{4u},\ Q^\alpha_{4z}$	3	1	3	3
orthorhombic	$D_{2\rm h}$					Q_0		$Q_u,\ Q_v$	G_{xyz}	$Q_4,\ Q_{4u},\ Q_{4v}$	3		3	3
	D_2	✓		✓	✓	Q_0		$Q_u,\ Q_v$	G_{xyz}	$Q_4,\ Q_{4u},\ Q_{4v}$	3		3	3
	$C_{2\rm v}$	✓	✓		✓	Q_0		$Q_u,\ Q_v$	G_{xyz}	$Q_4,\ Q_{4u},\ Q_{4v}$	3		3	3
monoclinic	$C_{2\rm h}$					Q_0	G_z	$Q_u,\ Q_v,\ Q_{xy}$	$G_{xyz},\ G^\alpha_z,\ G^\beta_z$	$Q_4,\ Q_{4u},\ Q_{4v},\ Q^\alpha_{4z},\ Q^\beta_{4z}$	5	1	5	7
	C_2	✓	✓	✓	✓	Q_0	G_z	$Q_u,\ Q_v,\ Q_{xy}$	$G_{xyz},\ G^\alpha_z,\ G^\beta_z$	$Q_4,\ Q_{4u},\ Q_{4v},\ Q^\alpha_{4z},\ Q^\beta_{4z}$	5	1	5	7
	$C_{\rm s}$	✓	✓		✓	Q_0	G_z	$Q_u,\ Q_v,\ Q_{xy}$	$G_{xyz},\ G^\alpha_z,\ G^\beta_z$	$Q_4,\ Q_{4u},\ Q_{4v},\ Q^\alpha_{4z},\ Q^\beta_{4z}$	5	1	5	7
hexagonal	$D_{6\rm h}$					Q_0		Q_u		Q_{40}	2		2	1
	D_6	✓		✓	✓	Q_0		Q_u		Q_{40}	2		2	1
	$D_{3\rm h}$	✓				Q_0		Q_u		Q_{40}	2		2	1
	$C_{6\rm v}$	✓	✓		✓	Q_0		Q_u		Q_{40}	2		2	1
	$C_{6\rm h}$					Q_0	G_z	Q_u	G^α_z	Q_{40}	3	1	3	3
	C_6	✓	✓	✓	✓	Q_0	G_z	Q_u	G^α_z	Q_{40}	3	1	3	3
	$C_{3\rm h}$	✓				Q_0	G_z	Q_u	G^α_z	Q_{40}	3	1	3	3
trigonal	$D_{3\rm d}$					Q_0		Q_u	G_{3a}	$Q_{40},\ Q_{4a}$	2		2	2
	D_3	✓		✓	✓	Q_0		Q_u	G_{3a}	$Q_{40},\ Q_{4a}$	2		2	2
	$C_{3\rm v}$	✓	✓		✓	Q_0		Q_u	G_{3b}	$Q_{40},\ Q_{4b}$	2		2	2
	$C_{3\rm i}$					Q_0	G_z	Q_u	$G^\alpha_z,\ G_{3a},\ G_{3b}$	$Q_{40},\ Q_{4a},\ Q_{4b}$	3	1	3	5
	C_3	✓	✓	✓	✓	Q_0	G_z	Q_u	$G^\alpha_z,\ G_{3a},\ G_{3b}$	$Q_{40},\ Q_{4a},\ Q_{4b}$	3	1	3	5

A set of independent operators in the basis (ϕ_z, ϕ_{xy}) is given by

$$\mathrm{A}_1^+ : \ \{\hat{\tau}_0\hat{\sigma}_0, \hat{\tau}_z\hat{\sigma}_0\} \leftrightarrow \{Q_0, Q_u\}, \quad \hat{\tau}_x\hat{\sigma}_0 \leftrightarrow G_v, \tag{64}$$

$$\mathrm{A}_2^+ : \ \hat{\tau}_y\hat{\sigma}_z \leftrightarrow G_{xy}, \tag{65}$$

$$\mathrm{E}^+ : \ (\hat{\tau}_y\hat{\sigma}_x, \hat{\tau}_y\hat{\sigma}_y) \leftrightarrow (Q_x, Q_y), \tag{66}$$

$$\mathrm{A}_1^- : \ \hat{\tau}_y\hat{\sigma}_0 \leftrightarrow M_v, \tag{67}$$

$$\mathrm{A}_2^- : \ \{\hat{\tau}_0\hat{\sigma}_z, \hat{\tau}_z\hat{\sigma}_z\} \leftrightarrow \{M_z, M^\alpha_z\}, \quad \hat{\tau}_x\hat{\sigma}_z \leftrightarrow M_{xy}, \tag{68}$$

$$\mathrm{E}^- : \ \{(\hat{\tau}_0\hat{\sigma}_x, \hat{\tau}_0\hat{\sigma}_y), (\hat{\tau}_z\hat{\sigma}_x, \hat{\tau}_z\hat{\sigma}_y)\} \leftrightarrow \{(M_x, M_y), (M^\alpha_x, M^\alpha_y)\}, \quad (\hat{\tau}_x\hat{\sigma}_x, \hat{\tau}_x\hat{\sigma}_y) \leftrightarrow (T_x, T_y), \tag{69}$$

In the case of the spinful basis functions, several odd-parity multipole degrees of freedom are potentially activated in addition to M_v and G_v in the spinless basis functions. For example, the MT dipole (T_x, T_y), which is the origin of the magnetoelectric effect, can be a primary order parameter when the thermodynamic averages of $(\hat{\tau}_x\hat{\sigma}_x, \hat{\tau}_x\hat{\sigma}_y)$ become nonzero. Interestingly, it is also possible to activate the MT dipole (T_x, T_y) by an external magnetic field or spontaneous magnetic ordering in the xy plane, since the M dipoles (M_x, M_y) belong to the same irreducible representation as (T_x, T_y). This fact indicates that E dipole $\boldsymbol{Q}$ is not necessary for the emergence of the MT dipole $\boldsymbol{T}$, which has never been clarified in previous interpretations of the MT dipole. Similarly, the M quadrupole M_{xy} becomes active under an external magnetic field along the z direction.

TABLE XVI. Odd-parity multipoles belonging to a totally symmetric representation in the point groups except for the triclinic ones (C_1, C_i) and other centrosymmetric point groups. $\chi^{\rm A}_{ij}$ and $\chi^{\rm P}_{ijk}$ represent rank-2 axial and rank-3 polar tensors, respectively. $\chi^{\rm A}_{ij}$ has nine components and $\chi^{\rm P}_{ijk}$ has 27 components at most. Note that the polar point groups (P) have rank-1 E multipoles ($\boldsymbol{Q}$), the chiral point groups (C) have rank-0 ET multipoles (G_0), and the gyrotropic point groups (G) have less than or equal to rank-2 E/ET multipoles (G_0, $\boldsymbol{Q}$, G_{2m}) yielding nonzero $\chi^{\rm A}_{ij}$. The gyrotropic point groups with only rank-1 E dipoles ($\boldsymbol{Q}$) in (C_{4v}, C_{6v}, C_{3v}) are called the weak gyrotropic point groups, which do not show a natural optical rotation.

						totally symmetric rep.					$\chi^{\rm A}_{ij}$	$\chi^{\rm P}_{ijk}$		
crystal system	point group	N	P	C	G	rank 0	rank 1	rank 2	rank 3	rank 4	2	1	2	3
cubic	O	✓		✓	✓	G_0				G_4	1		1	
	$T_{\rm d}$	✓							Q_{xyz}					1
	T	✓		✓	✓	G_0			Q_{xyz}	G_4	1		1	1
tetragonal	D_4	✓		✓	✓	G_0		G_u		G_4, G_{4u}	2		2	1
	$D_{2\rm d}$	✓			✓			G_v	Q_{xyz}	G_{4v}	1		1	2
	$C_{4\rm v}$	✓	✓		✓		Q_z		Q^α_z	G^α_{4z}	1	1	1	2
	C_4	✓	✓	✓	✓	G_0	Q_z	G_u	Q^α_z	G_4, G_{4u}, G^α_{4z}	3	1	3	3
	S_4	✓			✓			G_v, G_{xy}	Q_{xyz}, Q^β_z	G_{4v}, G^β_{4z}	2		2	4
orthorhombic	D_2	✓		✓	✓	G_0		G_u, G_v	Q_{xyz}	G_4, G_{4u}, G_{4v}	3		3	3
	$C_{2\rm v}$	✓	✓		✓		Q_z	G_{xy}	Q^α_z, Q^β_z	G^α_{4z}, G^β_{4z}	2	1	2	4
monoclinic	C_2	✓	✓	✓	✓	G_0	Q_z	G_u, G_v, G_{xy}	Q_{xyz}, Q^α_z, Q^β_z	G_4, G_{4u}, G_{4v}, G^α_{4z}, G^β_{4z}	5	1	5	7
	$C_{\rm s}$	✓	✓		✓		Q_x, Q_y	G_{yz}, G_{zx}	Q^α_x, Q^α_y, Q^β_x, Q^β_y	G^α_{4x}, G^α_{4y}, G^β_{4x}, G^β_{4y}	4	2	4	8
hexagonal	D_6	✓		✓	✓	G_0		G_u		G_{40}	2		2	1
	$D_{3\rm h}$	✓							Q_{3a}	G_{4a}				1
	$C_{6\rm v}$	✓	✓		✓		Q_z		Q^α_z		1	1	1	2
	C_6	✓	✓	✓	✓	G_0	Q_z	G_u	Q^α_z	G_{40}	3	1	3	3
	$C_{3\rm h}$	✓							Q_{3a}, Q_{3b}	G_{4a}, G_{4b}				2
trigonal	D_3	✓		✓	✓	G_0		G_u	Q_{3a}	G_{40}, G_{4a}	2		2	2
	$C_{3\rm v}$	✓	✓		✓		Q_z		Q^α_z, Q_{3a}	G_{4a}	1	1	1	3
	C_3	✓	✓	✓	✓	G_0	Q_z	G_u	Q^α_z, Q_{3a}, Q_{3b}	G_{40}, G_{4a}, G_{4b}	3	1	3	5

The final example is the basis functions $(\phi_x, \phi_y, \phi_{yz}, \phi_{zx})$ belonging to the E representation under the $D_{2\rm d}$ group. The direct product of the basis functions is given by

$$\begin{aligned}&(\mathrm{E}\oplus\mathrm{E})\otimes(\mathrm{E}\oplus\mathrm{E})\\&\quad=(2\mathrm{A}_1^+\oplus 2\mathrm{A}_2^-\oplus 2\mathrm{B}_1^+\oplus 2\mathrm{B}_2^+)_{\rm intra}\oplus(\mathrm{A}_1^+\oplus\mathrm{A}_2^+\\&\qquad\oplus\mathrm{B}_1^+\oplus\mathrm{B}_2^+\oplus\mathrm{A}_1^-\oplus\mathrm{A}_2^-\oplus\mathrm{B}_1^-\oplus\mathrm{B}_2^-)_{\rm inter}.\end{aligned}\tag{70}$$

This gives us a set of independent operators as

$$\mathrm{A}_1^+:\quad \{\hat\rho_0\hat\tau_0,\hat\rho_0\hat\tau_z\}\leftrightarrow\{Q_0,Q_u\},\quad \hat\rho_0\hat\tau_x\leftrightarrow G_v,\tag{71}$$

$$\mathrm{A}_2^+:\quad \hat\rho_y\hat\tau_y\leftrightarrow G_{xy},\tag{72}$$

$$\mathrm{B}_1^+:\quad \{\hat\rho_z\hat\tau_0,\hat\rho_z\hat\tau_z\}\leftrightarrow\{Q_v,Q_{4v}\},\quad \hat\rho_z\hat\tau_x\leftrightarrow G_u,\tag{73}$$

$$\mathrm{B}_2^+:\quad \{\hat\rho_x\hat\tau_0,\hat\rho_x\hat\tau_z\}\leftrightarrow\left\{Q_{xy},Q^\beta_{4z}\right\},\quad \hat\rho_x\hat\tau_x\leftrightarrow Q_z,\tag{74}$$

$$\mathrm{A}_1^-:\quad \hat\rho_0\hat\tau_y\leftrightarrow M_v,\tag{75}$$

$$\mathrm{A}_2^-:\quad \{\hat\rho_y\hat\tau_0,\hat\rho_y\hat\tau_z\}\leftrightarrow\left\{M_z,M^\alpha_z\right\},\quad \hat\rho_y\hat\tau_x\leftrightarrow M_{xy},\tag{76}$$

$$\mathrm{B}_1^-:\quad \hat\rho_z\hat\tau_y\leftrightarrow M_u,\tag{77}$$

$$\mathrm{B}_2^-:\quad \hat\rho_x\hat\tau_y\leftrightarrow T_z,\tag{78}$$

where $\hat\tau_0$ and $\hat{\boldsymbol{\tau}}$ are the unit and Pauli matrices acting on the different parity orbitals. In the case of the basis functions $(\phi_x, \phi_y, \phi_{yz}, \phi_{zx})$, there are several odd-parity multipole degrees of freedom even in the spinless basis functions, such as the E dipole Q_z and MT dipole T_z.

C. Linear response tensors

Finally, we show nonzero rank-2 and rank-3 tensors in each point group in terms of totally symmetric even-parity multipoles in Table XV and odd-parity multipoles in Table XVI. As described in Sec. III C, the rank-2 tensor $\chi^{(2)}_{ij}$ has nine independent components, which are characterized by the rank-0, rank-1, and rank-2 multipoles. Similarly, as discussed in Appendix D, the rank-2 tensor response by the rank-1 external field is described by the rank-3 tensor χ_{ijk} with 27 components, which are decomposed into a symmetric monopole response tensor with 3 components $\chi^{(1)}_i$, antisymmetric dipole response tensor with 9 components $\chi^{(2)}_{ij}$, and symmetric quadrupole response tensor with 15 components $\chi^{(3)}_{ijk}$. Moreover, the components of $\hat\chi^{(1,2,3)}$ are characterized by multipoles. For example, in the case of $D_{2\rm d}$ group, one component in the rank-2 axial tensor and three components of the rank-3 polar tensor become nonzero, as odd-parity multipoles G_v and Q_{xyz} belong to the totally symmetric representation in $D_{2\rm d}$ group as shown in Table XVI. Nonzero $\chi^{\rm A}_{ij}$ and $\chi^{\rm P}_{ijk}$ imply the emergence of the magnetoelectric effect,

TABLE XVII. Representative polar (P) and axial (A) tensors in condensed matter physics. The odd-parity tensors in the lower rows can be finite only when the spatial inversion symmetry is broken.

tensor	rank	$\mathcal{P}$	
χ_i^{A}	1	+	magnetocaloric coefficient
χ_{ij}^{P}	2		(thermo)electric/thermal conductivity
χ_{ijk}^{A}	3		spin conductivity, Nernst coefficient
χ_{ijkl}^{P}	4		elastic stiffness tensor
χ_i^{P}	1	−	electrocaloric coefficient
χ_{ij}^{A}	2		linear magnetoelectric tensor
χ_{ijk}^{P}	3		piezoelectric tensor
χ_{ijkl}^{A}	4		third-order magnetoelectric tensor

piezoelectric tensor, and so on. We summarize the representative tensors in condensed matter physics in Table XVII.

V. SUMMARY

In summary, we have investigated a general description of multipoles from the microscopic viewpoint. We have presented a definition of four multipoles in both real and momentum spaces, and how to apply to 32 point groups in seven crystal systems. We have demonstrated which multipole degrees of freedom become active in the tetragonal $D_{2\mathrm{d}}$ group as an example. We showed that physical properties in electron systems, such as the electromagnetic fields, band structures, and linear responses, are closely related with the multipoles, and hence, the multipole formulation gives a comprehensive and systematic understanding of physical phenomena in condensed matter physics at the microscopic level. Such a comprehensive investigation of multipoles could stimulate an identification of unknown order parameters, such as MT and ET multipoles, and a further exploration of cross-correlated couplings through multipole-multipole interactions, since we present more than 30 tables as a useful reference in order to cover most cases in all point groups.

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APPENDIX A: ATOMIC BASIS WAVE FUNCTIONS

We show the atomic spinless basis functions for s, p, d, and f orbitals as a function of angles $\hat{\boldsymbol{r}}/r$, which are used in the main text [133]. The basis functions are given by

$$\phi_0 = \frac{1}{\sqrt{4\pi}}, \tag{A1}$$

for an s orbital,

$$\phi_x = \sqrt{\frac{3}{4\pi}}\frac{x}{r}, \quad \phi_y = \sqrt{\frac{3}{4\pi}}\frac{y}{r}, \quad \phi_z = \sqrt{\frac{3}{4\pi}}\frac{z}{r}, \tag{A2}$$

for three p orbitals,

$$\begin{aligned} \phi_u &= \sqrt{\frac{5}{4\pi}}\frac{1}{2}\frac{3z^2-r^2}{r^2}, \quad \phi_v = \sqrt{\frac{5}{4\pi}}\frac{\sqrt{3}}{2}\frac{x^2-y^2}{r^2}, \\ \phi_{yz} &= \sqrt{\frac{5}{4\pi}}\sqrt{3}\frac{yz}{r^2}, \quad \phi_{zx} = \sqrt{\frac{5}{4\pi}}\sqrt{3}\frac{zx}{r^2}, \\ \phi_{xy} &= \sqrt{\frac{5}{4\pi}}\sqrt{3}\frac{xy}{r^2}, \end{aligned} \tag{A3}$$

for five d orbitals, and

$$\begin{aligned} \phi_{xyz} &= \sqrt{\frac{7}{4\pi}}\sqrt{15}\frac{xyz}{r^3}, \quad \phi_x^{\alpha} = \sqrt{\frac{7}{4\pi}}\frac{1}{2}\frac{x(5x^2-3r^2)}{r^3}, \\ \phi_y^{\alpha} &= \sqrt{\frac{7}{4\pi}}\frac{1}{2}\frac{y(5y^2-3r^2)}{r^3}, \quad \phi_z^{\alpha} = \sqrt{\frac{7}{4\pi}}\frac{1}{2}\frac{z(5z^2-3r^2)}{r^3}, \\ \phi_x^{\beta} &= \sqrt{\frac{7}{4\pi}}\frac{\sqrt{15}}{2}\frac{x(y^2-z^2)}{r^3}, \quad \phi_y^{\beta} = \sqrt{\frac{7}{4\pi}}\frac{\sqrt{15}}{2}\frac{y(z^2-x^2)}{r^3}, \\ \phi_z^{\beta} &= \sqrt{\frac{7}{4\pi}}\frac{\sqrt{15}}{2}\frac{z(x^2-y^2)}{r^3}. \end{aligned} \tag{A4}$$

for seven f orbitals. They are proportional to E multipoles in the cubic O_{h} representation in Tables I and II.

APPENDIX B: MOMENTUM-SPACE MULTIPOLES IN MULTIORBITAL SYSTEMS

As mentioned in Sec. II B, the extension of the momentum-space multipoles to multiorbital systems is not unique. Here, we discuss one possible extension.

As was discussed, in the single-band systems, the odd-rank M and even-rank ET multipoles are given by $\boldsymbol{\sigma}\cdot\nabla_{\boldsymbol{k}}O_{lm}(\boldsymbol{k})$ in Eqs. (23) and (24). By applying the time-reversal-conversion operator $(\boldsymbol{\sigma}\times\boldsymbol{k})\cdot\nabla_{\boldsymbol{k}}$, we obtain the expressions for the odd-rank ET and even-rank M multipoles in multiorbital systems as

$$\begin{aligned} G_{lm} &= (\boldsymbol{\sigma}\times\boldsymbol{k})\cdot\nabla_{\boldsymbol{k}}[\boldsymbol{\sigma}\cdot\nabla_{\boldsymbol{k}}O_{lm}(\boldsymbol{k})] \quad \text{(odd)}, \\ M_{lm} &= (\boldsymbol{\sigma}\times\boldsymbol{k})\cdot\nabla_{\boldsymbol{k}}[\boldsymbol{\sigma}\cdot\nabla_{\boldsymbol{k}}O_{lm}(\boldsymbol{k})] \quad \text{(even)}. \end{aligned}$$

These expressions are simplified by simple algebra as

$$M_{lm} = \begin{cases} i\boldsymbol{k}\cdot\boldsymbol{\sigma} & (l=0) \\ i\boldsymbol{k}\cdot\nabla_{\boldsymbol{k}}[\boldsymbol{\sigma}\cdot\nabla_{\boldsymbol{k}}O_{lm}(\boldsymbol{k})] & (l=2,4,\dots) \end{cases}, \tag{B1}$$

$$G_{lm} = i\boldsymbol{k}\cdot\nabla_{\boldsymbol{k}}[\boldsymbol{\sigma}\cdot\nabla_{\boldsymbol{k}}O_{lm}(\boldsymbol{k})] \quad (l=1,3,\dots), \tag{B2}$$

where we have used $\nabla^2 O_{lm}=0$. The specific form of the even-rank M and odd-rank ET multipoles up to $l=4$ and the MT monopole in momentum space is shown in Tables VI and VII.

APPENDIX C: SPACE-TIME INVERSION OPERATIONS OF LINEAR RESPONSE TENSOR

We discuss the space-time inversion properties of linear response tensors, which is also discussed in Ref. [42]. As discussed in Sec. III C, the linear response tensors in Eq. (43)

are generally decomposed into

$$\chi = \chi^{(\mathrm{J})} + \chi^{(\mathrm{E})},$$

$$\chi^{(\mathrm{J})} = -\frac{\gamma}{V}\sum_{knm}^{=} \frac{f_{nk}-f_{mk}}{\varepsilon_{nk}-\varepsilon_{mk}} \frac{A^{nm}_{k}B^{mn}_{k}}{(\varepsilon_{nk}-\varepsilon_{mk})^2+\gamma^2}, \tag{C1}$$

$$\chi^{(\mathrm{E})} = -\frac{i}{V}\sum_{knm}^{\neq} \frac{(f_{nk}-f_{mk})A^{nm}_{k}B^{mn}_{k}}{(\varepsilon_{nk}-\varepsilon_{mk})^2+\gamma^2}, \tag{C2}$$

where $\hat{A}$ and $\hat{B}$ are Hermite operators and these equations correspond to Eqs. (44) and (45), respectively. We assume that $\hat{A}$ and $\hat{B}$ have definite parties with respect to the spatial inversion and time-reversal operations as

$$\mathcal{P}(\hat{A}) = s\hat{A}, \quad \mathcal{T}(\hat{A}) = t\hat{A}, \quad (s,t=\pm 1), \tag{C3}$$

$$\mathcal{P}(\hat{B}) = s'\hat{B}, \quad \mathcal{T}(\hat{B}) = t'\hat{B}, \quad (s',t'=\pm 1). \tag{C4}$$

Moreover, for the Bloch states, $\mathcal{P}|n\boldsymbol{k}\rangle = |n-\boldsymbol{k}\rangle$ and $\mathcal{T}|n\boldsymbol{k}\rangle = |\bar{n}-\boldsymbol{k}\rangle$, where $\bar{n}$ is the time-reversal partner of n state. As $\mathcal{T}$ is antiunitary, these relations lead to $A^{nm}_{\boldsymbol{k}} = sA^{nm}_{-\boldsymbol{k}}$, $A^{nm}_{\boldsymbol{k}} = tA^{\bar{m}\bar{n}}_{-\boldsymbol{k}}$, $B^{nm}_{\boldsymbol{k}} = s'B^{nm}_{-\boldsymbol{k}}$, and $B^{mn}_{\boldsymbol{k}} = t'B^{\bar{n}\bar{m}}_{-\boldsymbol{k}}$.

Thus, in the presence of the spatial inversion symmetry, we show that

$$\begin{aligned}
\chi^{(\mathrm{J})} &= -\frac{\gamma}{V}\sum_{knm}^{=} \frac{f_{nk}-f_{mk}}{\varepsilon_{nk}-\varepsilon_{mk}} \frac{sA^{nm}_{-k}s'B^{mn}_{-k}}{(\varepsilon_{nk}-\varepsilon_{mk})^2+\gamma^2} \\
&= -\frac{\gamma}{V}\sum_{knm}^{=} \frac{f_{n-k}-f_{m-k}}{\varepsilon_{n-k}-\varepsilon_{m-k}} \frac{sA^{nm}_{k}s'B^{mn}_{k}}{(\varepsilon_{n-k}-\varepsilon_{m-k})^2+\gamma^2} \\
&= -\frac{\gamma}{V}\sum_{knm}^{=} \frac{f_{nk}-f_{mk}}{\varepsilon_{nk}-\varepsilon_{mk}} \frac{A^{nm}_{k}B^{mn}_{k}}{(\varepsilon_{nk}-\varepsilon_{mk})^2+\gamma^2} ss' \\
&= ss'\chi^{(\mathrm{J})},
\end{aligned}$$

where we have used $\varepsilon_{n-k} = \varepsilon_{nk}$. A similar discussion holds for $\chi^{(\mathrm{E})}$, and we obtain

$$\chi^{(\mathrm{J})} = ss'\chi^{(\mathrm{J})}, \quad \chi^{(\mathrm{E})} = ss'\chi^{(\mathrm{E})}. \tag{C5}$$

On the other hand, in the presence of the time-reversal symmetry, we have

$$\begin{aligned}
\chi^{(\mathrm{J})} &= -\frac{\gamma}{V}\sum_{knm}^{=} \frac{f_{nk}-f_{mk}}{\varepsilon_{nk}-\varepsilon_{mk}} \frac{tA^{\bar{m}\bar{n}}_{-k}t'B^{\bar{n}\bar{m}}_{-k}}{(\varepsilon_{nk}-\varepsilon_{mk})^2+\gamma^2} \\
&= -\frac{\gamma}{V}\sum_{knm}^{=} \frac{f_{\bar{m}-k}-f_{\bar{n}-k}}{\varepsilon_{\bar{m}-k}-\varepsilon_{\bar{n}-k}} \frac{tA^{nm}_{k}t'B^{mn}_{k}}{(\varepsilon_{\bar{m}-k}-\varepsilon_{\bar{n}-k})^2+\gamma^2} \\
&= -\frac{\gamma}{V}\sum_{knm}^{=} \frac{f_{mk}-f_{nk}}{\varepsilon_{mk}-\varepsilon_{nk}} \frac{A^{nm}_{k}B^{mn}_{k}}{(\varepsilon_{mk}-\varepsilon_{nk})^2+\gamma^2} tt' \\
&= tt'\chi^{(\mathrm{J})},
\end{aligned}$$

where we have used $\varepsilon_{\bar{n}-k} = \varepsilon_{nk}$. Similarly,

$$\begin{aligned}
\chi^{(\mathrm{E})} &= -\frac{i}{V}\sum_{knm}^{\neq} \frac{(f_{nk}-f_{mk})tA^{\bar{m}\bar{n}}_{-k}t'B^{\bar{n}\bar{m}}_{-k}}{(\varepsilon_{nk}-\varepsilon_{mk})^2+\gamma^2} \\
&= -\frac{i}{V}\sum_{knm}^{\neq} \frac{(f_{\bar{m}-k}-f_{\bar{n}-k})tA^{nm}_{k}t'B^{mn}_{k}}{(\varepsilon_{\bar{m}-k}-\varepsilon_{\bar{n}-k})^2+\gamma^2} \\
&= -\frac{i}{V}\sum_{knm}^{\neq} \frac{(f_{mk}-f_{nk})A^{nm}_{k}B^{mn}_{k}}{(\varepsilon_{mk}-\varepsilon_{nk})^2+\gamma^2} tt' \\
&= -tt'\chi^{(\mathrm{E})}.
\end{aligned}$$

Thus we obtain

$$\chi^{(\mathrm{J})} = tt'\chi^{(\mathrm{J})}, \quad \chi^{(\mathrm{E})} = -tt'\chi^{(\mathrm{E})}. \tag{C6}$$

Due to the above symmetry properties, in the case of $s \neq s'$ a breaking of the spatial inversion is necessary to obtain a finite $\chi^{(\mathrm{J,E})}$. Similarly, in the case of $t \neq t'$ ($t = t'$), a breaking of time-reversal symmetry is necessary to obtain a finite $\chi^{(\mathrm{J})}$ ($\chi^{(\mathrm{E})}$).

APPENDIX D: OTHER LINEAR RESPONSE TENSORS

In this section, we show a natural extension of linear-response tensors as discussed in Sec. III C. In the main text, we discuss the rank-2 linear-response tensors $\chi^{(2)}_{ij}$ with nine independent components when $\hat{A}_i$ and $\hat{B}_j$ are both rank-1 tensors. The extension of the linear response tensors when $\hat{A}_i$ and $\hat{B}_j$ are higher-rank tensors is straightforward.

Let us consider the rank-2 response to the rank-1 external field, namely,

$$A_{ij} = \sum_k \chi_{ijk}B_k. \tag{D1}$$

We can decompose A_{ij} into the monopole, dipole, and quadrupole contributions as

$$A_{ij} = \delta_{ij}A^{\mathrm{M}} + \sum_l \epsilon_{ijl}A^{\mathrm{D}}_l + A^{\mathrm{Q}}_{ij}, \tag{D2}$$

where ϵ_{ijl} is the totally antisymmetric tensor (Levi-Civita symbol), and A^{Q}_{ij} is symmetric and traceless. Note that A^{D}_i is polar (axial) when others are axial (polar) since ϵ_{ijk} and δ_{ij} are axial and polar tensors, respectively, and A^{M}, A^{D}_i, and A^{Q}_{ij} eventually have the same parity. For each contribution, the linear-response tensor is introduced as

$$A^{\mathrm{M}} = \frac{1}{3}\sum_i A_{ii} = \sum_k \chi^{(1)}_k B_k, \tag{D3}$$

$$A^{\mathrm{D}}_i = \frac{1}{2}\sum_{jk}\epsilon_{ijk}A_{jk} = \sum_k \chi^{(2)}_{ik}B_k \tag{D4}$$

$$A^{\mathrm{Q}}_{ij} = \sum_k \chi^{(3)}_{ijk}B_k, \tag{D5}$$

where $\chi^{(3)}_{ijk}$ is symmetric and traceless with respect to (i,j), i.e., $\chi^{(3)}_{ijk} = \chi^{(3)}_{jik}$ and $\sum_i \chi^{(3)}_{iik} = 0$. Note that $\chi^{(2)}_{ij}$ corresponds to Eq. (46) in the main text. By inserting these expressions into the decomposition of A_{ij}, we obtain the relation to χ_{ijk} as

$$A_{ij} = \sum_k \left(\delta_{ij}\chi^{(1)}_k + \sum_l \epsilon_{ijl}\chi^{(2)}_{lk} + \chi^{(3)}_{ijk}\right)B_k. \tag{D6}$$

Each linear-response function can be decomposed as

$$\chi_k^{(1)} = \chi_k^{\mathrm{D}}, \tag{D7}$$

$$\chi_{ij}^{(2)} = \delta_{ij}\chi^{\mathrm{M}} + \sum_k \epsilon_{ijk}\chi_k^{\mathrm{D}'} + \chi_{ij}^{\mathrm{Q}}, \tag{D8}$$

$$\chi_{ijk}^{(3)} = \delta_{ij}\chi_k^{\mathrm{D}''} + \sum_{lm}(\epsilon_{ikl}\delta_{jm} + \epsilon_{jkl}\delta_{im})\chi_{lm}^{\mathrm{Q}'} + \chi_{ijk}^{\mathrm{O}}, \tag{D9}$$

where χ_{ij}^{Q} is symmetric and traceless, and χ_{ijk}^{O} is totally symmetric with respect to any permutations of indices and $\sum_i \chi_{iik}^{\mathrm{O}} = -3\chi_k^{\mathrm{D}''}$.

Since χ^{M}, χ_k^{D}, χ_{ij}^{Q}, χ_{ijk}^{O} are characterized by Y_0 (monopole), X_i (dipole), Y_{ij} (quadrupole), and X_{ijk} (octupole), respectively, where X is polar (axial) and Y is axial (polar) when $\chi_{ijk}^{(3)}$ is polar (axial), we explicitly express the linear-response functions in terms of multipoles as

$$\chi^{(1)} = \begin{pmatrix} X_x & X_y & X_z \end{pmatrix}, \tag{D10}$$

$$\chi^{(2)} = \begin{pmatrix} Y_0 - Y_u + Y_v & Y_{xy} + X_z & Y_{zx} - X_y \\ Y_{xy} - X_z & Y_0 - Y_u - Y_v & Y_{yz} + X_x \\ Y_{zx} + X_y & Y_{yz} - X_x & Y_0 + 2Y_u \end{pmatrix}, \tag{D11}$$

$$\chi^{(3)} = \begin{pmatrix} X_x + Y_{yz} - X_x^\alpha - X_x^\beta & X_y - Y_{zx} - X_y^\alpha + X_y^\beta & -2X_z + 2X_z^\alpha \\ -3X_x + Y_{yz} + 3X_x^\alpha - X_x^\beta & 3X_y + Y_{zx} - 3X_y^\alpha - X_y^\beta & -2Y_{xy} + 2X_z^\beta \\ -3Y_u - Y_v + X_{xyz} & -3X_z - Y_{xy} - 2X_z^\alpha - 2X_z^\beta & -3X_y + Y_{zx} - 2X_y^\alpha + 2X_y^\beta \\ -3X_z + Y_{xy} - 2X_z^\alpha + 2X_z^\beta & 3Y_u - Y_v + X_{xyz} & -3X_x - Y_{yz} - 2X_x^\alpha - 2X_x^\beta \\ -3X_y - Y_{zx} - 2X_y^\alpha - 2X_y^\beta & -3X_x + Y_{yz} - 2X_x^\alpha + 2X_x^\beta & 2Y_v + X_{xyz} \end{pmatrix}, \tag{D12}$$

where we have put $\chi^{\mathrm{M}} = Y_0$, $\chi_i^{\mathrm{D}} = \chi_i^{\mathrm{D}'} = X_i$, $\chi_{ij}^{\mathrm{Q}} = \chi_{ij}^{\mathrm{Q}'} = Y_{ij}$, $\chi_{ijk}^{\mathrm{O}} = X_{ijk}$, and $\chi_i^{\mathrm{D}''} = 5X_i$ for notational simplicity. We have also used the relations

$$\sum_i Y_{ii} = 0, \quad Y_u = \frac{1}{6}(2Y_{zz} - Y_{xx} - Y_{yy}), \quad Y_v = \frac{1}{2}(Y_{xx} - Y_{yy}), \tag{D13}$$

for quadrupoles and

$$\begin{aligned} \sum_i X_{iik} &= -15X_k \quad (k = x, y, z), \\ X_x^\alpha &= \frac{1}{20}(2X_{xxx} - 3X_{yyx} - 3X_{zzx}), \\ X_y^\alpha &= \frac{1}{20}(2X_{yyy} - 3X_{zzy} - 3X_{xxy}), \\ X_z^\alpha &= \frac{1}{20}(2X_{zzz} - 3X_{xxz} - 3X_{yyz}), \\ X_x^\beta &= \frac{1}{4}(X_{yyx} - X_{zzx}), \\ X_y^\beta &= \frac{1}{4}(X_{zzy} - X_{xxy}), \\ X_z^\beta &= \frac{1}{4}(X_{xxz} - X_{yyz}), \end{aligned} \tag{D14}$$

for octupoles. The prefactors in the right-hand sides are chosen in order to simplify the resultant expressions in the linear-response tensors. In the expression of $\chi^{(3)}$, the rows correspond to $(i, j) = (u, v, yz, zx, xy)$, e.g., $\chi_{uk}^{(3)} = (2\chi_{zzk}^{(3)} - \chi_{xxk}^{(3)} - \chi_{yyk}^{(3)})/6$ according to Eq. (D13).

In order to complete the comprehensive lists, we also give the expression of $\chi^{(3)}$ in the hexagonal $D_{6\mathrm{h}}$ group as

$$\chi^{(3)} = \begin{pmatrix} X_x + Y_{yz} + 2X_{3u} & X_y - Y_{zx} + 2X_{3v} & -2X_z + 2X_z^\alpha \\ -3X_x + Y_{yz} + X_{3a} - X_{3u} & 3X_y + Y_{zx} + X_{3b} + X_{3v} & -2Y_{xy} + 2X_z^\beta \\ -3Y_u - Y_v + X_{xyz} & -3X_z - Y_{xy} - 2X_z^\alpha - 2X_z^\beta & -3X_y + Y_{zx} - 2X_y^\alpha + 4X_{3v} \\ -3X_z + Y_{xy} - 2X_z^\alpha + 2X_z^\beta & 3Y_u - Y_v + X_{xyz} & -3X_x - Y_{yz} - 2X_x^\alpha + 4X_{3u} \\ -3X_y - Y_{zx} + X_{3b} - X_{3v} & -3X_x + Y_{yz} - X_{3a} - X_{3u} & 2Y_v + X_{xyz} \end{pmatrix}, \tag{D15}$$

where we have used the relations

$$X_{3u} = -\tfrac{1}{2}\big(X_x^\alpha + X_x^\beta\big), \quad X_{3v} = -\tfrac{1}{2}\big(X_y^\alpha - X_y^\beta\big),$$
$$X_{3a} = \tfrac{1}{2}\big(5X_x^\alpha - 3X_x^\beta\big), \quad X_{3b} = -\tfrac{1}{2}\big(5X_y^\alpha + 3X_y^\beta\big). \tag{D16}$$

We present two examples in the following: spin conductivity and piezoelectric tensors.

1. Spin conductivity tensor

We here consider the axial third-rank spin conductivity tensor, which is defined as

$$J_{ij}^{\rm s} = \sum_k \sigma_{ijk}^{\rm s} E_k, \tag{D17}$$

where $\hat{A}_{ij} = \hat{J}_{ij}^{\rm s}$ and $\hat{B}_j = \hat{J}_j$ in the general formula in Eq. (43). Note that there are 9 independent components for the spin current tensor, as it is characterized by the product of rank-1 tensors, $\hat{J}_i$ and $\hat{\sigma}_j$ ($\hat{\sigma}$ is the spin degree of freedom). From the symmetry point of view, the spin conductivity tensor is decomposed as

$$J_{ij}^{\rm s} \equiv J_i \sigma_j = \delta_{ij} J^{\rm s,M} + \sum_k \epsilon_{ijk} J_k^{\rm s,D} + J_{ij}^{\rm s,Q}, \tag{D18}$$

where

$$J^{\rm s,M} = \frac{1}{3}\boldsymbol{J}\cdot\boldsymbol{\sigma}, \quad \boldsymbol{J}^{\rm s,D} = \frac{1}{2}(\boldsymbol{J}\times\boldsymbol{\sigma}), \quad \hat{J}^{\rm s,Q} = \frac{1}{2}\begin{pmatrix} [2J_z\sigma_z - J_x\sigma_x - J_y\sigma_y]/3 \\ J_x\sigma_x - J_y\sigma_y \\ J_y\sigma_z + J_z\sigma_y \\ J_z\sigma_x + J_x\sigma_z \\ J_x\sigma_y + J_y\sigma_x \end{pmatrix}. \tag{D19}$$

$J^{\rm s,M}$, $\boldsymbol{J}^{\rm s,D}$, and $\hat{J}^{\rm s,Q}$ possess the same symmetry as ET monopole, E dipole, and ET quadrupole, respectively. The corresponding tensors $\hat{\sigma}^{\rm s(1)}$, $\hat{\sigma}^{\rm s(2)}$, and $\hat{\sigma}^{\rm s(3)}$ are expressed as

$$\hat{\sigma}^{\rm s(1,J)} = (M_x \quad M_y \quad M_z), \quad \hat{\sigma}^{\rm s(1,E)} = (G_x \quad G_y \quad G_z), \tag{D20}$$

$$\hat{\sigma}^{\rm s(2,J)} = \begin{pmatrix} T_0 - T_u + T_v & T_{xy} + M_z & T_{zx} - M_y \\ T_{xy} - M_z & T_0 - T_u - T_v & T_{yz} + M_x \\ T_{zx} + M_y & T_{yz} - M_x & T_0 + 2T_u \end{pmatrix},$$
$$\hat{\sigma}^{\rm s(2,E)} = \begin{pmatrix} Q_0 - Q_u + Q_v & Q_{xy} + G_z & Q_{zx} - G_y \\ Q_{xy} - G_z & Q_0 - Q_u - Q_v & Q_{yz} + G_x \\ Q_{zx} + G_y & Q_{yz} - G_x & Q_0 + 2Q_u \end{pmatrix}, \tag{D21}$$

and

$$\hat{\sigma}^{\rm s(3,J)} = \begin{pmatrix} M_x + T_{yz} - M_x^\alpha - M_x^\beta & M_y - T_{zx} - M_y^\alpha + M_y^\beta & -2M_z + 2M_z^\alpha \\ -3M_x + T_{yz} + 3M_x^\alpha - M_x^\beta & 3M_y + T_{zx} - 3M_y^\alpha - M_y^\beta & -2T_{xy} + 2M_z^\beta \\ -3T_u - T_v + M_{xyz} & -3M_z - T_{xy} - 2M_z^\alpha - 2M_z^\beta & -3M_y + T_{zx} - 2M_y^\alpha + 2M_y^\beta \\ -3M_z + T_{xy} - 2M_z^\alpha + 2M_z^\beta & 3T_u - T_v + M_{xyz} & -3M_x - T_{yz} - 2M_x^\alpha - 2M_x^\beta \\ -3M_y - T_{zx} - 2M_y^\alpha - 2M_y^\beta & -3M_x + T_{yz} - 2M_x^\alpha + 2M_x^\beta & 2T_v + M_{xyz} \end{pmatrix},$$
$$\hat{\sigma}^{\rm s(3,E)} = \begin{pmatrix} G_x + Q_{yz} - G_x^\alpha - G_x^\beta & G_y - Q_{zx} - G_y^\alpha + G_y^\beta & -2G_z + 2G_z^\alpha \\ -3G_x + Q_{yz} + 3G_x^\alpha - G_x^\beta & 3G_y + Q_{zx} - 3G_y^\alpha - G_y^\beta & -2Q_{xy} + 2G_z^\beta \\ -3Q_u - Q_v + G_{xyz} & -3G_z - Q_{xy} - 2G_z^\alpha - 2G_z^\beta & -3G_y + Q_{zx} - 2G_y^\alpha + 2G_y^\beta \\ -3G_z + Q_{xy} - 2G_z^\alpha + 2G_z^\beta & 3Q_u - Q_v + G_{xyz} & -3G_x - Q_{yz} - 2G_x^\alpha - 2G_x^\beta \\ -3G_y - Q_{zx} - 2G_y^\alpha - 2G_y^\beta & -3G_x + Q_{yz} - 2G_x^\alpha + 2G_x^\beta & 2Q_v + G_{xyz} \end{pmatrix}. \tag{D22}$$

2. Piezoelectric tensor

Next, we consider a piezoelectric effect where the strain and rotation is induced by the electric field (or current). The polar third-rank piezoelectric tensor d_{ijk} is given by

$$\zeta_{ij} = \sum_k d_{ijk} E_k. \tag{D23}$$

$\zeta_{ij} \equiv \partial u_i/\partial x_j$, where $\boldsymbol{u}$ is a displacement vector field. It can be decomposed as

$$\zeta_{ij} = \delta_{ij}\varepsilon_0 + \sum_k \epsilon_{ijk}\,\omega_k + \tilde{\varepsilon}_{ij}, \tag{D24}$$

where $\varepsilon_0 = (\nabla \cdot \boldsymbol{u})/3$, $\boldsymbol{\omega} = (\nabla \times \boldsymbol{u})/2$, and $\varepsilon_{ij} = (\zeta_{ij} + \zeta_{ji})/2 = \delta_{ij}\varepsilon_0 + \tilde{\varepsilon}_{ij}$ represent the bulk modulus, rotation, and symmetric strain, respectively. Since ε_0, $(\omega_x, \omega_y, \omega_z)$, and $(\varepsilon_u, \varepsilon_v, \varepsilon_{yz}, \varepsilon_{zx}, \varepsilon_{xy})$ are the same symmetry as E monopole, ET dipole, and E quadrupole, respectively, the corresponding tensors $\hat{d}^{(1)}$, $\hat{d}^{(2)}$, and $\hat{d}^{(3)}$ are given by

$$\begin{aligned} \hat{d}^{(1,\mathrm{J})} &= (T_x \quad T_y \quad T_z), \\ \hat{d}^{(1,\mathrm{E})} &= (Q_x \quad Q_y \quad Q_z), \end{aligned} \tag{D25}$$

$$\hat{d}^{(2,\mathrm{J})} = \begin{pmatrix} M_0 - M_u + M_v & M_{xy} + T_z & M_{zx} - T_y \\ M_{xy} - T_z & M_0 - M_u - M_v & M_{yz} + T_x \\ M_{zx} + T_y & M_{yz} - T_x & M_0 + 2M_u \end{pmatrix},$$

$$\hat{d}^{(2,\mathrm{E})} = \begin{pmatrix} G_0 - G_u + G_v & G_{xy} + Q_z & G_{zx} - Q_y \\ G_{xy} - Q_z & G_0 - G_u - G_v & G_{yz} + Q_x \\ G_{zx} + Q_y & G_{yz} - Q_x & G_0 + 2G_u \end{pmatrix}, \tag{D26}$$

and

$$\hat{d}^{(3,\mathrm{J})} = \begin{pmatrix} T_x + M_{yz} - T_x^\alpha - T_x^\beta & T_y - M_{zx} - T_y^\alpha + T_y^\beta & -2T_z + 2T_z^\alpha \\ -3T_x + M_{yz} + 3T_x^\alpha - T_x^\beta & 3T_y + M_{zx} - 3T_y^\alpha - T_y^\beta & -2M_{xy} + 2T_z^\beta \\ -3M_u - M_v + T_{xyz} & -3T_z - M_{xy} - 2T_z^\alpha - 2T_z^\beta & -3T_y + M_{zx} - 2T_y^\alpha + 2T_y^\beta \\ -3T_z + M_{xy} - 2T_z^\alpha + 2T_z^\beta & 3M_u - M_v + T_{xyz} & -3T_x - M_{yz} - 2T_x^\alpha - 2T_x^\beta \\ -3T_y - M_{zx} - 2T_y^\alpha - 2T_y^\beta & -3T_x + M_{yz} - 2T_x^\alpha + 2T_x^\beta & 2M_v + T_{xyz} \end{pmatrix},$$

$$\hat{d}^{(3,\mathrm{E})} = \begin{pmatrix} Q_x + G_{yz} - Q_x^\alpha - Q_x^\beta & Q_y - G_{zx} - Q_y^\alpha + Q_y^\beta & -2Q_z + 2Q_z^\alpha \\ -3Q_x + G_{yz} + 3Q_x^\alpha - Q_x^\beta & 3Q_y + G_{zx} - 3Q_y^\alpha - Q_y^\beta & -2G_{xy} + 2Q_z^\beta \\ -3G_u - G_v + Q_{xyz} & -3Q_z - G_{xy} - 2Q_z^\alpha - 2Q_z^\beta & -3Q_y + G_{zx} - 2Q_y^\alpha + 2Q_y^\beta \\ -3Q_z + G_{xy} - 2Q_z^\alpha + 2Q_z^\beta & 3G_u - G_v + Q_{xyz} & -3Q_x - G_{yz} - 2Q_x^\alpha - 2Q_x^\beta \\ -3Q_y - G_{zx} - 2Q_y^\alpha - 2Q_y^\beta & -3Q_x + G_{yz} - 2Q_x^\alpha + 2Q_x^\beta & 2G_v + Q_{xyz} \end{pmatrix}. \tag{D27}$$

For example, the rotation by the electric field, i.e., nonzero $\hat{d}^{(2,\mathrm{E})}$, occurs for the gyrotropic point groups, and the symmetric strain by the electric field, i.e., nonzero $\hat{d}^{(3,\mathrm{E})}$, occurs for the noncentrosymetric point groups, as shown in Table XVI.

APPENDIX E: O_h AND ITS SUBGROUPS

We summarize the active multipoles for the basis for s, p, d, and f orbitals with/without the spin degree of freedom for the O_h and its subgroups. The active multipoles for cubic and orthorhombic crystals in the spinless (spinful) basis are shown in Table XVIII (XIX) and XX (XXI), respectively. The active multipoles for tetragonal crystals in the spinless (spinful) basis are shown in Table XIII (XIV) in the main text.

TABLE XVIII. Multipoles under the cubic crystal system (O_h) in the spinless basis. $(g,u)^\pm = \mathrm{A}^\pm_{1g,u} \oplus \mathrm{E}^\pm_{g,u} \oplus \mathrm{T}^\mp_{1g,u} \oplus \mathrm{T}^\pm_{2g,u}$; $\langle g,u\rangle^\pm = \mathrm{A}^\pm_{2g,u} \oplus \mathrm{E}^\pm_{g,u} \oplus \mathrm{T}^\pm_{1g,u} \oplus \mathrm{T}^\pm_{2g,u}$. The superscript $^\pm$ includes both time-reversal-even ($^+$) and time-reversal-odd ($^-$) operators. The subscripts g,u are omitted for the O group. $\mathrm{A}_{1u} \leftrightarrow \mathrm{A}_{2u}$ and $\mathrm{T}_{1u} \leftrightarrow \mathrm{T}_{2u}$ for the T_d group. The subscripts 1,2 are omitted for the T_h group. The subscripts g,u, 1, 2 are omitted for the T group.

	(s) A_{1g}	(p) T_{1u}	(d) E_g	T_{2g}	(f) A_{2u}	T_{1u}	T_{2u}
(s) A_{1g}	A^+_{1g}	$\mathrm{T}^\pm_{1u}$	$\mathrm{E}^\pm_g$	$\mathrm{T}^\pm_{2g}$	$\mathrm{A}^\pm_{2u}$	$\mathrm{T}^\pm_{1u}$	$\mathrm{T}^\pm_{2u}$
(p) T_{1u}		$(g)^+$	$\mathrm{T}^\pm_{1u}, \mathrm{T}^\pm_{2u}$	$\langle u\rangle^\pm$	$\mathrm{T}^\pm_{2g}$	$(g)^\pm$	$\langle g\rangle^\pm$
(d) E_g			$\mathrm{A}^+_{1g}, \mathrm{A}^-_{2g}, \mathrm{E}^+_g$	$\mathrm{T}^\pm_{1g}, \mathrm{T}^\pm_{2g}$	$\mathrm{E}^\pm_u$	$\mathrm{T}^\pm_{1u}, \mathrm{T}^\pm_{2u}$	$\mathrm{T}^\pm_{1u}, \mathrm{T}^\pm_{2u}$
T_{2g}				$(g)^+$	$\mathrm{T}^\pm_{1u}$	$\langle u\rangle^\pm$	$(u)^\pm$
(f) A_{2u}					A^+_{1g}	$\mathrm{T}^\pm_{2g}$	$\mathrm{T}^\pm_{1g}$
T_{1u}						$(g)^+$	$\langle g\rangle^\pm$
T_{2u}							$(g)^+$

TABLE XIX. Multipoles under the cubic crystal system ($O_{\rm h}$) in the spinful basis. $(g,u)^{\pm} = A^{\pm}_{1g,u} \oplus E^{\pm}_{g,u} \oplus T^{\pm}_{2g,u} \oplus A^{\mp}_{2g,u} \oplus 2T^{\mp}_{1g,u} \oplus T^{\mp}_{2g,u}$; $\langle g,u\rangle^{\pm} = E^{\pm}_{g,u} \oplus T^{\pm}_{1g,u} \oplus T^{\pm}_{2g,u}$. The subscripts g,u are omitted for the O group. $A_{1u} \leftrightarrow A_{2u}$, $T_{1u} \leftrightarrow T_{2u}$, and $E_{1/2u} \leftrightarrow E_{5/2u}$ for the $T_{\rm d}$ group. The subscripts 1,2 are omitted for the $T_{\rm h}$ group. The subscripts g,u and 1,2 are omitted for the T group.

	(s) $E_{1/2g}$	(p) $E_{1/2u}$	$G_{3/2u}$	(d) $G_{3/2g}$	$E_{5/2g}$	$G_{3/2g}$	(f) $E_{5/2u}$	$E_{1/2u}$	$G_{3/2u}$	$E_{5/2u}$	$G_{3/2u}$
(s) $A_{1g} \otimes E_{1/2g} \to E_{1/2g}$	A^{+}_{1g}, T^{-}_{1g}	$A^{\pm}_{1u}, T^{\pm}_{1u}$	$\langle u\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$A^{\pm}_{2g}, T^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$A^{\pm}_{2u}, T^{\pm}_{2u}$	$A^{\pm}_{1u}, T^{\pm}_{1u}$	$\langle u\rangle^{\pm}$	$A^{\pm}_{2u}, T^{\pm}_{2u}$	$\langle u\rangle^{\pm}$
(p) $T_{1u} \otimes E_{1/2g} \to E_{1/2u}$		A^{+}_{1g}, T^{-}_{1g}	$\langle g\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$A^{\pm}_{2u}, T^{\pm}_{2u}$	$\langle u\rangle^{\pm}$	$A^{\pm}_{2g}, T^{\pm}_{2g}$	$A^{\pm}_{1g}, T^{\pm}_{1g}$	$\langle g\rangle^{\pm}$	$A^{\pm}_{2g}, T^{\pm}_{2g}$	$\langle g\rangle^{\pm}$
	$G_{3/2u}$		$(g)^{+}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$
(d) $E_g \otimes E_{1/2g} \to G_{3/2g}$				$(g)^{+}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$
$T_{2g} \otimes E_{1/2g} \to E_{5/2g}$					A^{+}_{1g}, T^{-}_{1g}	$\langle g\rangle^{\pm}$	$A^{\pm}_{1u}, T^{\pm}_{1u}$	$A^{\pm}_{2u}, T^{\pm}_{2u}$	$\langle u\rangle^{\pm}$	$A^{\pm}_{1u}, T^{\pm}_{1u}$	$\langle u\rangle^{\pm}$
	$G_{3/2g}$					$(g)^{+}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$
(f) $A_{2u} \otimes E_{1/2g} \to E_{5/2u}$							A^{+}_{1g}, T^{-}_{1g}	$A^{\pm}_{2g}, T^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$A^{\pm}_{1g}, T^{\pm}_{1g}$	$\langle g\rangle^{\pm}$
$T_{1u} \otimes E_{1/2g} \to E_{1/2u}$								A^{+}_{1g}, T^{-}_{1g}	$\langle g\rangle^{\pm}$	$A^{\pm}_{2g}, T^{\pm}_{2g}$	$\langle g\rangle^{\pm}$
	$G_{3/2u}$								$(g)^{+}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$
$T_{2u} \otimes E_{1/2g} \to E_{5/2u}$										A^{+}_{1g}, T^{-}_{1g}	$\langle g\rangle^{\pm}$
	$G_{3/2u}$										$(g)^{+}$

APPENDIX F: D_{6h} AND ITS SUBGROUPS

Here, we summarize various tables for the hexagonal D_{6h} and its subgroups. Tables XXII and XXIII represent even- and odd-parity hexagonal harmonics. Tables XXIV and XXV represent the operator expressions of even- and odd-parity multipoles in real space. Tables XXVI and XXVII represent the even- and odd-parity multipoles in momentum space. Table XXVIII represents the relation of multipoles under the D_{6h} group and its subgroups. Tables XXIX (XXX) and XXXI (XXXII) represent the active multipoles under the hexagonal and trigonal crystals in the spinless (spinful) basis, respectively.

TABLE XX. Multipoles under the orthorhombic crystal system (D_{2h}) in the spinless basis. The subscripts g,u are omitted for the D_2 group. $A_g, B_{1u} \to A_1$, $A_u, B_{1g} \to A_2$, $B_{3g,u} \to B_1$, and $B_{2g,u} \to B_2$ for the C_{2v} group.

	(s) A_g	(p) B_{3u}	B_{2u}	B_{1u}	(d) A_g	A_g	B_{3g}	B_{2g}	B_{1g}	(f) A_u	B_{3u}	B_{2u}	B_{1u}	B_{3u}	B_{2u}	B_{1u}
(s) A_g	A^{+}_{g}	$B^{\pm}_{3u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$	$A^{\pm}_{g}$	$A^{\pm}_{g}$	$B^{\pm}_{3g}$	$B^{\pm}_{2g}$	$B^{\pm}_{1g}$	$A^{\pm}_{u}$	$B^{\pm}_{3u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$	$B^{\pm}_{3u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$
(p) B_{3u}		A^{+}_{g}	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$	$B^{\pm}_{3u}$	$B^{\pm}_{3u}$	$A^{\pm}_{u}$	$B^{\pm}_{1u}$	$B^{\pm}_{2u}$	$B^{\pm}_{3g}$	$A^{\pm}_{g}$	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$	$A^{\pm}_{g}$	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$
B_{2u}			A^{+}_{g}	$B^{\pm}_{3g}$	$B^{\pm}_{2u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$	$A^{\pm}_{u}$	$B^{\pm}_{3u}$	$B^{\pm}_{2g}$	$B^{\pm}_{1g}$	$A^{\pm}_{g}$	$B^{\pm}_{3g}$	$B^{\pm}_{1g}$	$A^{\pm}_{g}$	$B^{\pm}_{3g}$
B_{1u}				A^{+}_{g}	$B^{\pm}_{1u}$	$B^{\pm}_{1u}$	$B^{\pm}_{2u}$	$B^{\pm}_{3u}$	$A^{\pm}_{u}$	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$	$B^{\pm}_{3g}$	$A^{\pm}_{g}$	$B^{\pm}_{2g}$	$B^{\pm}_{3g}$	$A^{\pm}_{g}$
(d) A_g					A^{+}_{g}	$A^{\pm}_{g}$	$B^{\pm}_{3g}$	$B^{\pm}_{2g}$	$B^{\pm}_{1g}$	$A^{\pm}_{u}$	$B^{\pm}_{3u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$	$B^{\pm}_{3u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$
A_g						A^{+}_{g}	$B^{\pm}_{3g}$	$B^{\pm}_{2g}$	$B^{\pm}_{1g}$	$A^{\pm}_{u}$	$B^{\pm}_{3u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$	$B^{\pm}_{3u}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$
B_{3g}							A^{+}_{g}	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$	$B^{\pm}_{3u}$	$A^{\pm}_{u}$	$B^{\pm}_{1u}$	$B^{\pm}_{2u}$	$A^{\pm}_{u}$	$B^{\pm}_{1u}$	$B^{\pm}_{2u}$
B_{2g}								A^{+}_{g}	$B^{\pm}_{3g}$	$B^{\pm}_{2u}$	$B^{\pm}_{1u}$	$A^{\pm}_{u}$	$B^{\pm}_{3u}$	$B^{\pm}_{1u}$	$A^{\pm}_{u}$	$B^{\pm}_{3u}$
B_{1g}									A^{+}_{g}	$B^{\pm}_{1u}$	$B^{\pm}_{2u}$	$B^{\pm}_{3u}$	$A^{\pm}_{u}$	$B^{\pm}_{2u}$	$B^{\pm}_{3u}$	$A^{\pm}_{u}$
(f) A_u										A^{+}_{g}	$B^{\pm}_{3g}$	$B^{\pm}_{2g}$	$B^{\pm}_{1g}$	$B^{\pm}_{3g}$	$B^{\pm}_{2g}$	$B^{\pm}_{1g}$
B_{3u}											A^{+}_{g}	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$	$A^{\pm}_{g}$	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$
B_{2u}												A^{+}_{g}	$B^{\pm}_{3g}$	$B^{\pm}_{1g}$	$A^{\pm}_{g}$	$B^{\pm}_{3g}$
B_{1u}													A^{+}_{g}	$B^{\pm}_{2g}$	$B^{\pm}_{3g}$	$A^{\pm}_{g}$
B_{3u}														A^{+}_{g}	$B^{\pm}_{1g}$	$B^{\pm}_{2g}$
B_{2u}															A^{+}_{g}	$B^{\pm}_{3g}$
B_{1u}																A^{+}_{g}

TABLE XXI. Multipoles under the orthorhombic crystal system (D_{2h}) in the spinful basis. $(g,u)^{\pm} = A^{\pm}_{g,u} \oplus B^{\mp}_{1g,u} \oplus B^{\mp}_{2g,u} \oplus B^{\mp}_{3g,u}$. The subscripts g, u are omitted for the D_2 group. A_g, $B_{1u} \to A_1$, A_u, $B_{1g} \to A_2$, $B_{3g,u} \to B_1$, and $B_{2g,u} \to B_2$ for the C_{2v} group.

		(s) $E_{1/2g}$	(p) $E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$	(d) $E_{1/2g}$	$E_{1/2g}$	$E_{1/2g}$	$E_{1/2g}$	$E_{1/2g}$	(f) $E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$
(s) $A_g \otimes E_{1/2g} \to$	$E_{1/2g}$	$(g)^+$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$
(p) $B_{3u} \otimes E_{1/2g} \to$	$E_{1/2u}$		$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$
$B_{2u} \otimes E_{1/2g} \to$	$E_{1/2u}$			$(g)^+$	$(g)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$
$B_{1u} \otimes E_{1/2g} \to$	$E_{1/2u}$				$(g)^+$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$
(d) $A_g \otimes E_{1/2g} \to$	$E_{1/2g}$					$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$
$A_g \otimes E_{1/2g} \to$	$E_{1/2g}$						$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$
$B_{3g} \otimes E_{1/2g} \to$	$E_{1/2g}$							$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$
$B_{2g} \otimes E_{1/2g} \to$	$E_{1/2g}$								$(g)^+$	$(g)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$
$B_{1g} \otimes E_{1/2g} \to$	$E_{1/2g}$									$(g)^+$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$
(f) $A_u \otimes E_{1/2g} \to$	$E_{1/2u}$										$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$
$B_{3u} \otimes E_{1/2g} \to$	$E_{1/2u}$											$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$
$B_{2u} \otimes E_{1/2g} \to$	$E_{1/2u}$												$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$
$B_{1u} \otimes E_{1/2g} \to$	$E_{1/2u}$													$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$
$B_{3u} \otimes E_{1/2g} \to$	$E_{1/2u}$														$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$
$B_{2u} \otimes E_{1/2g} \to$	$E_{1/2u}$															$(g)^+$	$(g)^{\pm}$
$B_{1u} \otimes E_{1/2g} \to$	$E_{1/2u}$																$(g)^+$

TABLE XXII. Even-parity hexagonal harmonics (E multipoles in unit of $-e$ in the hexagonal D_{6h} group) up to $l = 6$. The correspondence to the tesseral harmonics is shown. A_1, A_2, B_1, B_2, E_1, and E_2 correspond to Γ_1, Γ_2, Γ_3, Γ_4, Γ_5, and Γ_6, respectively, in the Bethe notation. Note that $Q^{\beta 1}_{4v} = Q^{\alpha}_{4z}$, $Q^{\beta 2}_{4u} = -Q_{4v}$, $Q^{\beta 2}_{4v} = Q^{\beta}_{4z}$, $Q_{6s} = Q^{\beta 1}_{6z}$, $Q^{\beta 1}_{6v} = Q^{\alpha}_{6z}$, and $Q^{\beta 2}_{6v} = Q^{\beta 2}_{6z}$ in Table I.

rank	irrep.	symbol	definition	correspondence
0	A^+_{1g}	Q_0	1	(00)
2	A^+_{1g}	Q_u	$\frac{1}{2}(3z^2 - r^2)$	(20)
	E^+_{1g}	Q_{zx}, Q_{yz}	$\sqrt{3}zx$, $\sqrt{3}yz$	(21), (21)′
	E^+_{2g}	Q_v, Q_{xy}	$\frac{\sqrt{3}}{2}(x^2 - y^2)$, $\sqrt{3}xy$	(22), (22)′
4	A^+_{1g}	Q_{40}	$\frac{1}{8}(35z^4 - 30z^2r^2 + 3r^4)$	(40)
	B^+_{1g}	Q_{4a}	$\frac{\sqrt{70}}{4}yz(3x^2 - y^2)$	(43)′
	B^+_{2g}	Q_{4b}	$\frac{\sqrt{70}}{4}zx(x^2 - 3y^2)$	(43)
	E^+_{1g}	Q^{α}_{4u}, Q^{α}_{4v}	$\frac{\sqrt{10}}{4}zx(7z^2 - 3r^2)$, $\frac{\sqrt{10}}{4}yz(7z^2 - 3r^2)$	(41), (41)′
	E^+_{2g}	$Q^{\beta 1}_{4u}$, $Q^{\beta 1}_{4v}$	$\frac{\sqrt{35}}{8}(x^4 - 6x^2y^2 + y^4)$, $\frac{\sqrt{35}}{2}xy(x^2 - y^2)$	(44), (44)′
	E^+_{2g}	$Q^{\beta 2}_{4u}$, $Q^{\beta 2}_{4v}$	$\frac{\sqrt{5}}{4}(x^2 - y^2)(7z^2 - r^2)$, $\frac{\sqrt{5}}{2}xy(7z^2 - r^2)$	(42), (42)′
6	A^+_{1g}	Q_{60}	$\frac{1}{16}(231z^6 - 315z^4r^2 + 105z^2r^4 - 5r^6)$	(60)
	A^+_{1g}	Q_{6c}	$\frac{\sqrt{462}}{32}[x^6 - 15x^2y^2(x^2 - y^2) - y^6]$	(66)
	A^+_{2g}	Q_{6s}	$\frac{\sqrt{462}}{16}xy(3x^4 - 10x^2y^2 + 3y^4)$	(66)′
	B^+_{1g}	Q_{6a}	$\frac{\sqrt{210}}{16}yz(3x^2 - y^2)(11z^2 - 3r^2)$	(63)′
	B^+_{2g}	Q_{6b}	$\frac{\sqrt{210}}{16}zx(x^2 - 3y^2)(11z^2 - 3r^2)$	(63)
	E^+_{1g}	$Q^{\alpha 1}_{6u}$, $Q^{\alpha 1}_{6v}$	$\frac{3\sqrt{154}}{16}zx(x^4 - 10x^2y^2 + 5y^4)$, $\frac{3\sqrt{154}}{16}yz(5x^4 - 10x^2y^2 + y^4)$	(65), (65)′
	E^+_{1g}	$Q^{\alpha 2}_{6u}$, $Q^{\alpha 2}_{6v}$	$\frac{\sqrt{21}}{8}zx[5r^4 + 3z^2(11z^2 - 10r^2)]$, $\frac{\sqrt{21}}{8}yz[5r^4 + 3z^2(11z^2 - 10r^2)]$	(61), (61)′
	E^+_{2g}	$Q^{\beta 1}_{6u}$, $Q^{\beta 1}_{6v}$	$\frac{3\sqrt{7}}{16}(x^4 - 6x^2y^2 + y^4)(11z^2 - r^2)$, $\frac{3\sqrt{7}}{4}xy(x^2 - y^2)(11z^2 - r^2)$	(64), (64)′
	E^+_{2g}	$Q^{\beta 2}_{6u}$, $Q^{\beta 2}_{6v}$	$\frac{\sqrt{210}}{32}(x^2 - y^2)[r^4 + 3z^2(11z^2 - 6r^2)]$, $\frac{\sqrt{210}}{16}xy[r^4 + 3z^2(11z^2 - 6r^2)]$	(62), (62)′

TABLE XXIII. Odd-parity hexagonal harmonics (E multipoles in unit of $-e$ in the hexagonal D_{6h} group) up to $l = 6$. Note that $Q_{50} = Q^{\alpha 1}_{5z}$, $Q^{\beta 1}_{5u} = Q^{\alpha 2}_{5z}$, $Q^{\beta 1}_{5v} = Q_{5u}$, $Q^{\beta 2}_{5u} = Q^{\beta}_{5z}$, and $Q^{\beta 2}_{5v} = -Q_{5v}$ in Table II.

rank	irrep.	symbol	definition	correspondence
1	A^+_{2u}	Q_z	z	(10)
	E^+_{1u}	$Q_x,\ Q_y$	$x,\ y$	(11), (11)′
3	A^+_{2u}	Q^{α}_z	$\frac{1}{2}z(5z^2-3r^2)$	(30)
	B^+_{1u}	Q_{3a}	$\frac{\sqrt{10}}{4}x(x^2-3y^2)$	(33)
	B^+_{2u}	Q_{3b}	$\frac{\sqrt{10}}{4}y(3x^2-y^2)$	(33)′
	E^+_{1u}	$Q_{3u},\ Q_{3v}$	$\frac{\sqrt{6}}{4}x(5z^2-r^2),\ \frac{\sqrt{6}}{4}y(5z^2-r^2)$	(31), (31)′
	E^+_{2u}	$Q^{\beta}_z,\ Q_{xyz}$	$\frac{\sqrt{15}}{2}z(x^2-y^2),\ \sqrt{15}xyz$	(32), (32)′
5	A^+_{2u}	Q_{50}	$\frac{1}{8}z(63z^4-70z^2r^2+15r^4)$	(50)
	B^+_{1u}	Q_{5a}	$\frac{\sqrt{70}}{16}x(x^2-3y^2)(9z^2-r^2)$	(53)
	B^+_{2u}	Q_{5b}	$\frac{\sqrt{70}}{16}y(3x^2-y^2)(9z^2-r^2)$	(53)′
	E^+_{1u}	$Q^{\alpha 1}_{5u}, Q^{\alpha 1}_{5v}$	$\frac{3\sqrt{14}}{16}x(x^4-10x^2y^2+5y^4),\ \frac{3\sqrt{14}}{16}y(5x^4-10x^2y^2+y^4)$	(55), (55)′
	E^+_{1u}	$Q^{\alpha 2}_{5u}, Q^{\alpha 2}_{5v}$	$\frac{\sqrt{15}}{8}x[r^4+7z^2(3z^2-2r^2)],\ \frac{\sqrt{15}}{8}y[r^4+7z^2(3z^2-2r^2)]$	(51), (51)′
	E^+_{2u}	$Q^{\beta 1}_{5u}, Q^{\beta 1}_{5v}$	$\frac{3\sqrt{35}}{8}z(x^4-6x^2y^2+y^4),\ \frac{3\sqrt{35}}{2}xyz(x^2-y^2)$	(54), (54)′
	E^+_{2u}	$Q^{\beta 2}_{5u}, Q^{\beta 2}_{5v}$	$\frac{\sqrt{105}}{4}z(x^2-y^2)(3z^2-r^2),\ \frac{\sqrt{105}}{2}xyz(3z^2-r^2)$	(52), (52)′

TABLE XXIV. Operator expressions of even-parity multipoles up to $l = 4$ in the hexagonal D_{6h} group.

rank	type	irrep.	symbol	definition
0	E	A^+_{1g}	Q_0	1
	MT	A^-_{1g}	T'_0	i (off-diagonal element)
1	M	A^-_{2g}	M_z	m^z_1
		E^-_{1g}	$M_x,\ M_y$	$m^x_1,\ m^y_1$
	ET	A^+_{2g}	G'_z	$z\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma})$
		E^+_{1g}	$G'_x,\ G'_y$	$x\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma}),\ y\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma})$
2	E	A^+_{1g}	Q_u	$\frac{1}{2}(3z^2-r^2)$
		E^+_{1g}	$Q_{zx},\ Q_{yz}$	$\sqrt{3}zx,\ \sqrt{3}yz$
		E^+_{2g}	$Q_v,\ Q_{xy}$	$\frac{\sqrt{3}}{2}(x^2-y^2),\ \sqrt{3}xy$
	MT	A^-_{1g}	T_u	$3zt^z_2-\boldsymbol{r}\cdot\boldsymbol{t}_2$
		E^-_{1g}	$T_{zx},\ T_{yz}$	$\sqrt{3}(zt^x_2+xt^z_2),\ \sqrt{3}(yt^z_2+zt^y_2)$
		E^-_{2g}	$T_v,\ T_{xy}$	$\sqrt{3}(xt^x_2-yt^y_2),\ \sqrt{3}(xt^y_2+yt^x_2)$
3	M	A^-_{2g}	M^{α}_z	$3[\frac{1}{2}(3z^2-r^2)m^z_3-z(xm^x_3+ym^y_3)]$
		B^-_{1g}	M_{3a}	$3\frac{\sqrt{10}}{4}[(x^2-y^2)m^x_3-2xym^y_3]$
		B^-_{2g}	M_{3b}	$3\frac{\sqrt{10}}{4}[2xym^x_3+(x^2-y^2)m^y_3]$
		E^-_{1g}	$M_{3u},\ M_{3v}$	$\frac{\sqrt{6}}{4}[(5z^2-r^2)m^x_3+2x(5zm^z_3-\boldsymbol{r}\cdot\boldsymbol{m}_3)],\ \frac{\sqrt{6}}{4}[(5z^2-r^2)m^y_3+2y(5zm^z_3-\boldsymbol{r}\cdot\boldsymbol{m}_3)]$
		E^-_{2g}	$M^{\beta}_z,\ M_{xyz}$	$\sqrt{15}[\frac{1}{2}(x^2-y^2)m^z_3+z(xm^x_3-ym^y_3)],\ \sqrt{15}(yzm^x_3+zxm^y_3+xym^z_3)$
	ET	A^+_{2g}	G^{α}_z	$9zg^{zz}_3-6(xg^{zx}_3+yg^{yz}_3)-3z\sum_{\alpha}g^{\alpha\alpha}_3$
		B^+_{1g}	G_{3a}	$3\sqrt{\frac{5}{2}}[x(g^{xx}_3-g^{yy}_3)-2yg^{xy}_3]$
		B^+_{2g}	G_{3b}	$3\sqrt{\frac{5}{2}}[2xg^{xy}_3+y(g^{xx}_3-g^{yy}_3)]$
		E^+_{1g}	$G_{3u},\ G_{3v}$	$\sqrt{\frac{3}{2}}[x(5g^{zz}_3-\sum_{\alpha}g^{\alpha\alpha}_3)+2(5zg^{xz}_3-\sum_{\alpha}\alpha g^{x\alpha})],\ \sqrt{\frac{3}{2}}[y(5g^{zz}_3-\sum_{\alpha}g^{\alpha\alpha}_3)+2(5zg^{yz}_3-\sum_{\alpha}\alpha g^{y\alpha})]$
		E^+_{2g}	$G^{\beta}_z,\ G_{xyz}$	$\sqrt{15}[2(xg^{zx}_3-yg^{yz}_3)+z(g^{xx}_3-g^{yy}_3)],\ 2\sqrt{15}(xg^{yz}_3+yg^{zx}_3+zg^{xy}_3)$
4	E	A^+_{1g}	Q_{40}	$\frac{1}{8}(35z^4-30z^2r^2+3r^4)$
		E^+_{2g}	$Q^{\beta 2}_{4u}, Q^{\beta 2}_{4v}$	$\frac{\sqrt{5}}{4}(x^2-y^2)(7z^2-r^2),\ \frac{\sqrt{5}}{2}xy(7z^2-r^2)$
		B^+_{1g}	Q_{4a}	$\frac{\sqrt{70}}{4}yz(3x^2-y^2)$
		B^+_{2g}	Q_{4b}	$\frac{\sqrt{70}}{4}zx(x^2-3y^2)$
		E^+_{1g}	$Q^{\alpha}_{4u}, Q^{\alpha}_{4v}$	$\frac{\sqrt{10}}{4}zx(7z^2-3r^2),\ \frac{\sqrt{10}}{4}yz(7z^2-3r^2)$
		E^+_{2g}	$Q^{\beta 1}_{4u}, Q^{\beta 1}_{4v}$	$\frac{\sqrt{35}}{8}(x^4-6x^2y^2+y^4),\ \frac{\sqrt{35}}{2}xy(x^2-y^2)$

TABLE XXIV. (*Continued.*)

rank	type	irrep.	symbol	definition
	MT	A_{1g}^-	T_{40}	$\frac{1}{2}[3(5z^2-r^2)(5zt_4^z-\boldsymbol{r}\cdot\boldsymbol{t}_4)-40zt_4^z]$
		B_{1g}^-	T_{4a}	$\frac{1}{2}\sqrt{\frac{35}{2}}[6xyzt_4^x+3z(x^2-y^2)t_4^y+y(3x^2-y^2)t_4^z]$
		B_{2g}^-	T_{4b}	$\frac{1}{2}\sqrt{\frac{35}{2}}[3z(x^2-y^2)t_4^x-6xyzt_4^y+x(x^2-3y^2)t_4^z]$
		E_{1g}^-	$T_{4u}^\alpha, T_{4v}^\alpha$	$\boldsymbol{t}_4\cdot\nabla Q_{4u}^\alpha,\ \boldsymbol{t}_4\cdot\nabla Q_{4v}^\alpha$
		E_{2g}^-	$T_{4u}^{\beta1}, T_{4v}^{\beta1}$	$\frac{\sqrt{35}}{2}[x(x^2-3y^2)t_4^x-y(3x^2-y^2)t_4^y],\ \frac{\sqrt{35}}{2}[y(3x^2-y^2)t_4^x+x(x^2-3y^2)t_4^y]$
		E_{2g}^-	$T_{4u}^{\beta2}, T_{4v}^{\beta2}$	$\sqrt{5}[x(3z^2-x^2)t_4^x+y(y^2-3z^2)t_4^y+3z(x^2-y^2)t_4^z],\ \boldsymbol{t}_4\cdot\nabla Q_{4v}^{\beta2}$

TABLE XXV. Operator expressions of odd-parity multipoles up to $l=4$ in the hexagonal D_{6h} group.

rank	type	irrep.	symbol	definition
0	M	A_{1u}^-	M_0'	$\boldsymbol{r}\cdot\boldsymbol{\sigma}$
	ET	A_{1u}^+	G_0'	$\boldsymbol{l}\cdot(\boldsymbol{r}\times\boldsymbol{\sigma})$
1	E	A_{2u}^+	Q_z	z
		E_{1u}^+	$Q_x,\ Q_y$	$x,\ y$
	MT	A_{2u}^-	T_z	t_1^z
		E_{1u}^-	$T_x,\ T_y$	$t_1^x,\ t_1^y$
2	M	A_{1u}^-	M_u	$3zm_2^z-\boldsymbol{r}\cdot\boldsymbol{m}_2$
		E_{1u}^-	$M_{zx},\ M_{yz}$	$\sqrt{3}(zm_2^x+xm_2^z),\ \sqrt{3}(ym_2^z+zm_2^y)$
		E_{2u}^-	$M_v,\ M_{xy}$	$\sqrt{3}(xm_2^x-ym_2^y),\ \sqrt{3}(xm_2^y+ym_2^x)$
	ET	A_{1u}^+	G_u	$3g_2^{zz}-\sum_\alpha g_2^{\alpha\alpha}$
		E_{1u}^+	$G_{zx},\ G_{yz}$	$2\sqrt{3}g_2^{zx},\ 2\sqrt{3}g_2^{yz}$
		E_{2u}^+	$G_v,\ G_{xy}$	$\sqrt{3}(g_2^{xx}-g_2^{yy}),\ 2\sqrt{3}g_2^{xy}$
3	E	A_{2u}^+	Q_z^α	$\frac{1}{2}z(5z^2-3r^2)$
		B_{1u}^+	Q_{3a}	$\frac{\sqrt{10}}{4}x(x^2-3y^2)$
		B_{2u}^+	Q_{3b}	$\frac{\sqrt{10}}{4}y(3x^2-y^2)$
		E_{1u}^+	$Q_{3u},\ Q_{3v}$	$\frac{\sqrt{6}}{4}x(5z^2-r^2),\ \frac{\sqrt{6}}{4}y(5z^2-r^2)$
		E_{2u}^+	$Q_z^\beta,\ Q_{xyz}$	$\frac{\sqrt{15}}{2}z(x^2-y^2),\ \sqrt{15}xyz$
	MT	A_{2u}^-	T_z^α	$3[\frac{1}{2}(3z^2-r^2)t_3^z-z(xt_3^x+yt_3^y)]$
		B_{1u}^-	T_{3a}	$3\frac{\sqrt{10}}{4}[(x^2-y^2)t_3^x-2xyt_3^y]$
		B_{2u}^-	T_{3b}	$3\frac{\sqrt{10}}{4}[2xyt_3^x+(x^2-y^2)t_3^y]$
		E_{1u}^-	$T_{3u},\ T_{3v}$	$\frac{\sqrt{6}}{4}[(5z^2-r^2)t_3^x+2x(5zt_3^z-\boldsymbol{r}\cdot\boldsymbol{t}_3)],\ \frac{\sqrt{6}}{4}[(5z^2-r^2)t_3^y+2y(5zt_3^z-\boldsymbol{r}\cdot\boldsymbol{t}_3)]$
		E_{2u}^-	$T_z^\beta,\ T_{xyz}$	$\sqrt{15}[\frac{1}{2}(x^2-y^2)t_3^z+z(xt_3^x-yt_3^y)],\ \sqrt{15}(yzt_3^x+zxt_3^y+xyt_3^z)$
4	M	A_{1u}^-	M_{40}	$\frac{1}{2}[3(5z^2-r^2)(5zm_4^z-\boldsymbol{r}\cdot\boldsymbol{m}_4)-40zm_4^z]$
		B_{1u}^-	M_{4a}	$\frac{1}{2}\sqrt{\frac{35}{2}}[6xyzm_4^x+3z(x^2-y^2)m_4^y+y(3x^2-y^2)m_4^z]$
		B_{2u}^-	M_{4b}	$\frac{1}{2}\sqrt{\frac{35}{2}}[3z(x^2-y^2)m_4^x-6xyzm_4^y+x(x^2-3y^2)m_4^z]$
		E_{1u}^-	$M_{4u}^\alpha, M_{4v}^\alpha$	$\boldsymbol{m}_4\cdot\nabla Q_{4u}^\alpha,\ \boldsymbol{m}_4\cdot\nabla Q_{4v}^\alpha$
		E_{2u}^-	$M_{4u}^{\beta1}, M_{4v}^{\beta1}$	$\frac{\sqrt{35}}{2}[x(x^2-3y^2)m_4^x-y(3x^2-y^2)m_4^y],\ \frac{\sqrt{35}}{2}[y(3x^2-y^2)m_4^x+x(x^2-3y^2)m_4^y]$
		E_{2u}^-	$M_{4u}^{\beta2}, M_{4v}^{\beta2}$	$\sqrt{5}[x(3z^2-x^2)m_4^x+y(y^2-3z^2)m_4^y+3z(x^2-y^2)m_4^z],\ \boldsymbol{m}_4\cdot\nabla Q_{4v}^{\beta2}$
	ET	A_{1u}^+	G_{40}	$\frac{3}{2}[(3x^2+y^2-4z^2)g_4^{xx}+(x^2+3y^2-4z^2)g_4^{yy}+4(3z^2-r^2)g_4^{zz}+4xyg_4^{xy}-16z(xg_4^{xz}+yg_4^{yz})]$
		B_{1u}^+	G_{4a}	$3\sqrt{\frac{35}{2}}[yz(g_4^{xx}-g_4^{yy})+2x(zg_4^{xy}+yg_4^{xz})+(x^2-y^2)g_4^{yz}]$
		B_{2u}^+	G_{4b}	$3\sqrt{\frac{35}{2}}[zx(g_4^{xx}-g_4^{yy})-2y(xg_4^{yz}+zg_4^{xy})+(x^2-y^2)g_4^{xz}]$
		E_{1u}^+	$G_{4u}^\alpha, G_{4v}^\alpha$	$\sum_{\alpha\beta}g_4^{\alpha\beta}\nabla_\alpha\nabla_\beta Q_{4u}^\alpha,\ \sum_{\alpha\beta}g_4^{\alpha\beta}\nabla_\alpha\nabla_\beta Q_{4v}^\alpha$
		E_{2u}^+	$G_{4u}^{\beta1}, G_{4v}^{\beta1}$	$3\frac{\sqrt{35}}{2}[(x^2-y^2)(g_4^{xx}-g_4^{yy})-4xyg_4^{xy}],\ 3\sqrt{35}[xy(g_4^{xx}-g_4^{yy})+(x^2-y^2)g_4^{xy}]$
		E_{2u}^+	$G_{4u}^{\beta2}, G_{4v}^{\beta2}$	$3\sqrt{5}[(z^2-x^2)g_4^{xx}+(y^2-z^2)g_4^{yy}+(x^2-y^2)g_4^{zz}+4z(xg_4^{zx}-yg_4^{yz})],\ \sum_{\alpha\beta}g_4^{\alpha\beta}\nabla_\alpha\nabla_\beta Q_{4v}^{\beta2}$

TABLE XXVI. Even-parity multipoles in momentum space up to $l = 4$ in the hexagonal D_{6h} group. The higher-order representation with respect to $\boldsymbol{k}$ is also shown for Q_0 and (M_x, M_y, M_z) in the bracket.

rank	type	irrep.	symbol	definition
0	E	A_{1g}^+	Q_0	$\sigma_0\ [(k_x^2+k_y^2+k_z^2)\sigma_0]$
	MT	A_{1g}^-	T_0'	$i\sigma_0$
1	M	A_{2g}^-	M_z	$\sigma_z\ [(\boldsymbol{k}\cdot\boldsymbol{\sigma})k_z - \frac{1}{3}k^2\sigma_z]$
		E_{1g}^-	$M_x,\ M_y$	$\sigma_x,\ \sigma_y[(\boldsymbol{k}\cdot\boldsymbol{\sigma})k_x - \frac{1}{3}k^2\sigma_x, (\boldsymbol{k}\cdot\boldsymbol{\sigma})k_y - \frac{1}{3}k^2\sigma_y]$
	ET	A_{2g}^+	G_z'	$i\sigma_z$
		E_{1g}^+	$G_x',\ G_y'$	$i\sigma_x,\ i\sigma_y$
2	E	A_{1g}^+	Q_u	$\frac{1}{2}(3k_z^2-k^2)\sigma_0$
		E_{1g}^+	$Q_{zx},\ Q_{yz}$	$\sqrt{3}k_zk_x\sigma_0,\ \sqrt{3}k_yk_z\sigma_0$
		E_{2g}^+	$Q_v,\ Q_{xy}$	$\frac{\sqrt{3}}{2}(k_x^2-k_y^2)\sigma_0,\ \sqrt{3}k_xk_y\sigma_0$
	MT	A_{1g}^-	T_u	$3k_zQ_z - \boldsymbol{k}\cdot\boldsymbol{Q}$
		E_{1g}^-	$T_{zx},\ T_{yz}$	$\sqrt{3}(k_zQ_x + k_xQ_z),\ \sqrt{3}(k_yQ_z + k_zQ_y)$
		E_{2g}^-	$T_v,\ T_{xy}$	$\sqrt{3}(k_xQ_x - k_yQ_y),\ \sqrt{3}(k_xQ_y + k_yQ_x)$
3	M	A_{2g}^-	M_z^α	$3[\frac{1}{2}(3k_z^2-k^2)\sigma_z - k_z(k_x\sigma_x + k_y\sigma_y)]$
		B_{1g}^-	M_{3a}	$3\frac{\sqrt{10}}{4}[(k_x^2-k_y^2)\sigma_x - 2k_xk_y\sigma_y]$
		B_{2g}^-	M_{3b}	$3\frac{\sqrt{10}}{4}[2k_xk_y\sigma_x + (k_x^2-k_y^2)\sigma_y]$
		E_{1g}^-	$M_{3u},\ M_{3v}$	$\frac{\sqrt{6}}{4}[(5k_z^2-k^2)\sigma_x + 2k_x(5k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma})],\ \frac{\sqrt{6}}{4}[(5k_z^2-k^2)\sigma_y + 2k_y(5k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma})]$
		E_{2g}^-	$M_z^\beta,\ M_{xyz}$	$\sqrt{15}[\frac{1}{2}(k_x^2-k_y^2)\sigma_z + k_z(k_x\sigma_x - k_y\sigma_y)],\ \sqrt{15}(k_yk_z\sigma_x + k_zk_x\sigma_y + k_xk_y\sigma_z)$
	ET	A_{2g}^+	$G_z^{\alpha'}$	$3i[\frac{1}{2}(3k_z^2-k^2)\sigma_z - k_z(k_x\sigma_x + k_y\sigma_y)]$
		B_{1g}^+	G_{3a}'	$3i\frac{\sqrt{10}}{4}[(k_x^2-k_y^2)\sigma_x - 2k_xk_y\sigma_y]$
		B_{2g}^+	G_{3b}'	$3i\frac{\sqrt{10}}{4}[2k_xk_y\sigma_x + (k_x^2-k_y^2)\sigma_y]$
		E_{1g}^+	$G_{3u}',\ G_{3v}'$	$\frac{\sqrt{6}}{4}i[(5k_z^2-k^2)\sigma_x + 2k_x(5k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma})],\ \frac{\sqrt{6}}{4}i[(5k_z^2-k^2)\sigma_y + 2k_y(5k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma})]$
		E_{2g}^+	$G_z^{\beta'},\ G_{xyz}'$	$\sqrt{15}i[\frac{1}{2}(k_x^2-k_y^2)\sigma_z + k_z(k_x\sigma_x - k_y\sigma_y)],\ \sqrt{15}i(k_yk_z\sigma_x + k_zk_x\sigma_y + k_xk_y\sigma_z)$
4	E	A_{1g}^+	Q_{40}	$\frac{1}{8}(35k_z^4 - 30k_z^2k^2 + 3k^4)\sigma_0$
		B_{1g}^+	Q_{4a}	$\frac{\sqrt{70}}{4}k_yk_z(3k_x^2-k_y^2)\sigma_0$
		B_{2g}^+	Q_{4b}	$\frac{\sqrt{70}}{4}k_zk_x(k_x^2-3k_y^2)\sigma_0$
		E_{1g}^+	$Q_{4u}^\alpha,\ Q_{4v}^\alpha$	$\frac{\sqrt{10}}{4}k_zk_x(7k_z^2-3k^2)\sigma_0,\ \frac{\sqrt{10}}{4}k_yk_z(7k_z^2-3k^2)\sigma_0$
		E_{2g}^+	$Q_{4u}^{\beta1},\ Q_{4v}^{\beta1}$	$\frac{\sqrt{35}}{8}(k_x^4 - 6k_x^2k_y^2 + k_y^4)\sigma_0,\ \frac{\sqrt{35}}{2}k_xk_y(k_x^2-k_y^2)\sigma_0$
		E_{2g}^+	$Q_{4u}^{\beta2},\ Q_{4v}^{\beta2}$	$\frac{\sqrt{5}}{4}(k_x^2-k_y^2)(7k_z^2-k^2)\sigma_0,\ \frac{\sqrt{5}}{2}k_xk_y(7k_z^2-k^2)\sigma_0$
	MT	A_{1g}^-	T_{40}	$\frac{1}{2}[3(5k_z^2-k^2)(5k_zQ_z - \boldsymbol{k}\cdot\boldsymbol{Q}) - 40k_zQ_z]$
		B_{1g}^-	T_{4a}	$\frac{1}{2}\sqrt{\frac{35}{2}}[6k_xk_yk_zQ_x + 3k_z(k_x^2-k_y^2)Q_y + k_y(3k_x^2-k_y^2)Q_z]$
		B_{2g}^-	T_{4b}	$\frac{1}{2}\sqrt{\frac{35}{2}}[3k_z(k_x^2-k_y^2)Q_x - 6k_xk_yk_zQ_y + k_x(k_x^2-3k_y^2)Q_z]$
		E_{1g}^-	$T_{4u}^\alpha, T_{4v}^\alpha$	$\boldsymbol{Q}\cdot\nabla Q_{4u}^\alpha,\ \boldsymbol{Q}\cdot\nabla Q_{4v}^\alpha$
		E_{2g}^-	$T_{4u}^{\beta1}, T_{4v}^{\beta1}$	$\frac{\sqrt{35}}{2}[k_x(k_x^2-3k_y^2)Q_x - k_y(3k_x^2-k_y^2)Q_y],\ \frac{\sqrt{35}}{2}[k_y(3k_x^2-k_y^2)Q_x + k_x(k_x^2-3k_y^2)Q_y]$
		E_{2g}^-	$T_{4u}^{\beta2}, T_{4v}^{\beta2}$	$\sqrt{5}[k_x(3k_z^2-k_x^2)Q_x + k_y(k_y^2-3k_z^2)Q_y + 3k_z(k_x^2-k_y^2)Q_z],\ \boldsymbol{Q}\cdot\nabla Q_{4v}^{\beta2}$

TABLE XXVII. Odd-parity multipoles in momentum space up to $l = 4$ in the hexagonal D_{6h} group. The higher-order representation with respect to $\boldsymbol{k}$ is also shown for Q_0 and (M_x, M_y, M_z) in the bracket.

rank	type	irrep.	symbol	definition
0	M	A_{1u}^-	M_0'	$i\boldsymbol{k}\cdot\boldsymbol{\sigma}$
	ET	A_{1u}^+	G_0	$\boldsymbol{k}\cdot\boldsymbol{\sigma}$
1	E	A_{2u}^+	Q_z	$k_x\sigma_y - k_y\sigma_x$
		E_{1u}^+	$Q_x,\ Q_y$	$k_y\sigma_z - k_z\sigma_y,\ k_z\sigma_x - k_x\sigma_z$
	MT	A_{2u}^-	T_z	$k_z\sigma_0$
		E_{1u}^-	$T_x,\ T_y$	$k_x\sigma_0,\ k_y\sigma_0$
2	M	A_{1u}^-	M_u'	$3ik_z\sigma_z - i\boldsymbol{k}\cdot\boldsymbol{\sigma}$
		E_{1u}^-	$M_{zx}',\ M_{yz}'$	$\sqrt{3}i(k_z\sigma_x + k_x\sigma_z),\ \sqrt{3}i(k_y\sigma_z + k_z\sigma_y)$
		E_{2u}^-	$M_v',\ M_{xy}'$	$\sqrt{3}i(k_x\sigma_x - k_y\sigma_y),\ \sqrt{3}i(k_x\sigma_y + k_y\sigma_x)$
	ET	A_{1u}^+	G_u	$3k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma}$
		E_{1u}^+	$G_{zx},\ G_{yz}$	$\sqrt{3}(k_z\sigma_x + k_x\sigma_z),\ \sqrt{3}(k_y\sigma_z + k_z\sigma_y)$
		E_{2u}^+	$G_v,\ G_{xy}$	$\sqrt{3}(k_x\sigma_x - k_y\sigma_y),\ \sqrt{3}(k_x\sigma_y + k_y\sigma_x)$
3	E	A_{2u}^+	Q_z^α	$3[\frac{1}{2}(3k_z^2 - k^2)Q_z - k_z(k_xQ_x + k_yQ_y)]$
		B_{1u}^+	Q_{3a}	$3\frac{\sqrt{10}}{4}[(k_x^2 - k_y^2)Q_x - 2k_xk_yQ_y]$
		B_{2u}^+	Q_{3b}	$3\frac{\sqrt{10}}{4}[2k_xk_yQ_x + (k_x^2 - k_y^2)Q_y]$
		E_{1u}^+	$Q_{3u},\ Q_{3v}$	$\frac{\sqrt{6}}{4}[(5k_z^2 - k^2)Q_x + 2k_x(5k_zQ_z - \boldsymbol{k}\cdot\boldsymbol{Q})],\ \frac{\sqrt{6}}{4}[(5k_z^2 - k^2)Q_y + 2k_y(5k_zQ_z - \boldsymbol{k}\cdot\boldsymbol{Q})]$
		E_{2u}^+	$Q_z^\beta,\ Q_{xyz}$	$\sqrt{15}[\frac{1}{2}(k_x^2 - k_y^2)Q_z + k_z(k_xQ_x - k_yQ_y)],\ \sqrt{15}(k_yk_zQ_x + k_zk_xQ_y + k_xk_yQ_z)$
	MT	A_{2u}^-	T_z^α	$\frac{1}{2}k_z(5k_z^2 - 3k^2)\sigma_0$
		B_{1u}^-	T_{3a}	$\frac{\sqrt{10}}{4}k_x(k_x^2 - 3k_y^2)\sigma_0$
		B_{2u}^-	T_{3b}	$\frac{\sqrt{10}}{4}k_y(3k_x^2 - k_y^2)\sigma_0$
		E_{1u}^-	$T_{3u},\ T_{3v}$	$\frac{\sqrt{6}}{4}k_x(5k_z^2 - k^2)\sigma_0,\ \frac{\sqrt{6}}{4}k_y(5k_z^2 - k^2)\sigma_0$
		E_{2u}^-	$T_z^\beta,\ T_{xyz}$	$\frac{\sqrt{15}}{2}k_z(k_x^2 - k_y^2)\sigma_0,\ \sqrt{15}k_xk_yk_z\sigma_0$
4	M	A_{1u}^-	M_{40}'	$\frac{1}{2}i[3(5k_z^2 - k^2)(5k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma}) - 40k_z\sigma_z]$
		B_{1u}^-	M_{4a}'	$\frac{1}{2}\sqrt{\frac{35}{2}}i[6k_xk_yk_z\sigma_x + 3k_z(k_x^2 - k_y^2)\sigma_y + k_y(3k_x^2 - k_y^2)\sigma_z]$
		B_{2u}^-	M_{4b}'	$\frac{1}{2}\sqrt{\frac{35}{2}}i[3k_z(k_x^2 - k_y^2)\sigma_x - 6k_xk_yk_z\sigma_y + k_x(k_x^2 - 3k_y^2)\sigma_z]$
		E_{1u}^-	$M_{4u}^{\alpha'},\ M_{4v}^{\alpha'}$	$i\boldsymbol{\sigma}\cdot\nabla Q_{4u}^\alpha,\ i\boldsymbol{\sigma}\cdot\nabla Q_{4v}^\alpha$
		E_{2u}^-	$M_{4u}^{\beta1'},\ M_{4v}^{\beta1'}$	$\frac{\sqrt{35}}{2}i[k_x(k_x^2 - 3k_y^2)\sigma_x - k_x(3k_x^2 - k_y^2)\sigma_y],\ \frac{\sqrt{35}}{2}i[k_y(3k_x^2 - k_y^2)\sigma_x + k_x(k_x^2 - 3k_y^2)\sigma_y]$
		E_{2u}^-	$M_{4u}^{\beta2'},\ M_{4v}^{\beta2'}$	$\sqrt{5}i[k_x(3k_z^2 - k_x^2)\sigma_x + k_y(k_y^2 - 3k_z^2)\sigma_y + 3k_z(k_x^2 - k_y^2)\sigma_z],\ i\boldsymbol{\sigma}\cdot\nabla Q_{4v}^{\beta2}$
	ET	A_{1u}^+	G_{40}	$\frac{1}{2}[3(5k_z^2 - k^2)(5k_z\sigma_z - \boldsymbol{k}\cdot\boldsymbol{\sigma}) - 40k_z\sigma_z]$
		B_{1u}^+	G_{4a}	$\frac{1}{2}\sqrt{\frac{35}{2}}[6k_xk_yk_z\sigma_x + 3k_z(k_x^2 - k_y^2)\sigma_y + k_y(3k_x^2 - k_y^2)\sigma_z]$
		B_{2u}^+	G_{4b}	$\frac{1}{2}\sqrt{\frac{35}{2}}[3k_z(k_x^2 - k_y^2)\sigma_x - 6k_xk_yk_z\sigma_y + k_x(k_x^2 - 3k_y^2)\sigma_z]$
		E_{1u}^+	$G_{4u}^\alpha, G_{4v}^\alpha$	$\boldsymbol{\sigma}\cdot\nabla Q_{4u}^\alpha,\ \boldsymbol{\sigma}\cdot\nabla Q_{4v}^\alpha$
		E_{2u}^+	$G_{4u}^{\beta1}, G_{4v}^{\beta1}$	$\frac{\sqrt{35}}{2}[k_x(k_x^2 - 3k_y^2)\sigma_x - k_y(3k_x^2 - k_y^2)\sigma_y],\ \frac{\sqrt{35}}{2}[k_y(3k_x^2 - k_y^2)\sigma_x + k_x(k_x^2 - 3k_y^2)\sigma_y]$
		E_{2u}^+	$G_{4u}^{\beta2}, G_{4v}^{\beta2}$	$\sqrt{5}[k_x(3k_z^2 - k_x^2)\sigma_x + k_y(k_y^2 - 3k_z^2)\sigma_y + 3k_z(k_x^2 - k_y^2)\sigma_z],\ \boldsymbol{\sigma}\cdot\nabla Q_{4v}^{\beta2}$

TABLE XXVIII. Multipoles under hexagonal and trigonal crystals. The upper and lower columns represent even-parity (g) and odd-parity (u) multipoles. We take the x axis as the C_2' rotation axis. For C_{3v}, we take the zx plane as the σ_v mirror plane. $C_{3i} = S_6$. The basis functions are taken as $(x, y) \to x \pm iy$ for C_{6h}, C_6, C_{3h}, and C_{3i}.

E	ET	M	MT	D_{6h}	D_6	C_{6h}	C_{6v}	C_6	D_{3h}	C_{3h}	D_{3d}	D_3	C_{3v}	C_{3i}	C_3
$Q_0,\ Q_u,\ Q_{40}$	–	–	$T_0,\ T_u,\ T_{40}$	A_{1g}	A_1	A_g	A_1	A	A_1'	A′	A_{1g}	A_1	A_1	A_g	A
–	$G_z,\ G_z^\alpha$	$M_z,\ M_z^\alpha$	–	A_{2g}	A_2	A_g	A_2	A	A_2'	A′	A_{2g}	A_2	A_2	A_g	A
Q_{4a}	G_{3a}	M_{3a}	T_{4a}	B_{1g}	B_1	B_g	B_2	B	A_1''	A″	A_{1g}	A_1	A_2	A_g	A
Q_{4b}	G_{3b}	M_{3b}	T_{4b}	B_{2g}	B_2	B_g	B_1	B	A_2''	A″	A_{2g}	A_2	A_1	A_g	A
$Q_{zx},\ Q_{4u}^\alpha$	$G_x,\ G_{3u}$	$M_x,\ M_{3u}$	$T_{zx},\ T_{4u}^\alpha$	E_{1g}	E_1	E_{1g}	E_1	E_1	E″	E″	E_g	E	E	E_g	E
$Q_{yz},\ Q_{4v}^\alpha$	$G_y,\ G_{3v}$	$M_y,\ M_{3v}$	$T_{yz},\ T_{4v}^\alpha$												
$Q_v,\ Q_{4u}^{\beta1},\ Q_{4u}^{\beta2}$	G_{xyz}	M_{xyz}	$T_v,\ T_{4u}^{\beta1},\ T_{4u}^{\beta2}$	E_{2g}	E_2	E_{2g}	E_2	E_2	E′	E′	E_g	E	E	E_g	E
$Q_{xy},\ Q_{4v}^{\beta1},\ Q_{4v}^{\beta2}$	G_z^β	M_z^β	$T_{xy},\ T_{4v}^{\beta2},\ T_{4v}^{\beta2}$												
–	$G_0,\ G_u,\ G_{40}$	$M_0,\ M_u,\ M_{40}$	–	A_{1u}	A_1	A_u	A_2	A	A_1''	A″	A_{1u}	A_1	A_2	A_u	A
$Q_z,\ Q_z^\alpha$	–	–	$T_z,\ T_z^\alpha$	A_{2u}	A_2	A_u	A_1	A	A_2''	A″	A_{2u}	A_2	A_1	A_u	A
Q_{3a}	G_{4a}	M_{4a}	T_{3a}	B_{1u}	B_1	B_u	B_1	B	A_1'	A′	A_{1u}	A_1	A_1	A_u	A
Q_{3b}	G_{4b}	M_{4b}	T_{3b}	B_{2u}	B_2	B_u	B_2	B	A_2'	A′	A_{2u}	A_2	A_2	A_u	A
$Q_x,\ Q_{3u}$	$G_{zx},\ G_{4u}^\alpha$	$M_{zx},\ M_{4u}^\alpha$	$T_x,\ T_{3u}$	E_{1u}	E_1	E_{1u}	E_1	E_1	E′	E′	E_u	E	E	E_u	E
$Q_y,\ Q_{3v}$	$G_{yz},\ G_{4v}^\alpha$	$M_{yz},\ M_{4v}^\alpha$	$T_y,\ T_{3v}$												
Q_{xyz}	$G_v,\ G_{4u}^{\beta1},\ G_{4u}^{\beta2}$	$M_v,\ M_{4u}^{\beta1},\ M_{4u}^{\beta2}$	T_{xyz}	E_{2u}	E_2	E_{2u}	E_2	E_2	E″	E″	E_u	E	E	E_u	E
Q_z^β	$G_{xy},\ G_{4v}^{\beta1},\ G_{4v}^{\beta2}$	$M_{xy},\ M_{4v}^{\beta1},\ M_{4v}^{\beta2}$	T_z^β												

TABLE XXIX. Multipoles under the hexagonal crystal system (D_{6h}) in the spinless basis. $(g, u)^\pm = A_{1g,u}^\pm \oplus A_{2g,u}^\mp \oplus E_{2g,u}^\pm$ and $\langle g, u\rangle^\pm = B_{1g,u}^\pm \oplus B_{2g,u}^\pm \oplus E_{1g,u}^\pm$. The subscripts g, u are omitted for the D_6 group. The subscripts 1,2 in A and B are omitted for the C_{6h} group. $A_{1u} \leftrightarrow A_{2u}$ and $B_{1g} \leftrightarrow B_{2g}$, and then, the subscripts g, u are omitted for the C_{6v} group. The subscripts g, u and 1,2 in A and B are omitted for the C_6 group. $A_{1g}, B_{1u} \to A_1', A_{2g}, B_{2u} \to A_2', A_{1u}, B_{1g} \to A_1'', A_{2u}, B_{2g} \to A_2'', E_{2u}, E_{1g} \to E'', E_{2g}$, and $E_{1u} \to E'$ for the D_{3h} group. A_{1g}, $A_{2g}, B_{1u}, B_{2u} \to A', A_{1u}, A_{2u}, B_{1g}, B_{2g} \to A'', E_{2u}, E_{1g} \to E'', E_{2g}$, and $E_{1u} \to E'$ for the C_{3h} group.

	(s) A_{1g}	(p) A_{2u}	E_{1u}	(d) A_{1g}	E_{1g}	E_{2g}	(f) A_{2u}	B_{1u}	B_{2u}	E_{1u}	E_{2u}
(s) A_{1g}	A_{1g}^+	$A_{2u}^\pm$	$E_{1u}^\pm$	$A_{1g}^\pm$	$E_{1g}^\pm$	$E_{2g}^\pm$	$A_{2u}^\pm$	$B_{1u}^\pm$	$B_{2u}^\pm$	$E_{1u}^\pm$	$E_{2u}^\pm$
(p) A_{2u}		A_{1g}^+	$E_{1g}^\pm$	$A_{2u}^\pm$	$E_{1u}^\pm$	$E_{2u}^\pm$	$A_{1g}^\pm$	$B_{2g}^\pm$	$B_{1g}^\pm$	$E_{1g}^\pm$	$E_{2g}^\pm$
E_{1u}			$(g)^+$	$E_{1u}^\pm$	$(u)^\pm$	$\langle u\rangle^\pm$	$E_{1g}^\pm$	$E_{2g}^\pm$	$E_{2g}^\pm$	$(g)^\pm$	$\langle g\rangle^\pm$
(d) A_{1g}				A_{1g}^+	$E_{1g}^\pm$	$E_{2g}^\pm$	$A_{2u}^\pm$	$B_{1u}^\pm$	$B_{2u}^\pm$	$E_{1u}^\pm$	$E_{2u}^\pm$
E_{1g}					$(g)^+$	$\langle g\rangle^\pm$	$E_{1u}^\pm$	$E_{2u}^\pm$	$E_{2u}^\pm$	$(u)^\pm$	$\langle u\rangle^\pm$
E_{2g}						$(g)^+$	$E_{2u}^\pm$	$E_{1u}^\pm$	$E_{1u}^\pm$	$\langle u\rangle^\pm$	$(u)^\pm$
(f) A_{2u}							A_{1g}^+	$B_{2g}^\pm$	$B_{1g}^\pm$	$E_{1g}^\pm$	$E_{2g}^\pm$
B_{1u}								A_{1g}^+	$A_{2g}^\pm$	$E_{2g}^\pm$	$E_{1g}^\pm$
B_{2u}									A_{1g}^+	$E_{2g}^\pm$	$E_{1g}^\pm$
E_{1u}										$(g)^+$	$\langle g\rangle^\pm$
E_{2u}											$(g)^+$

TABLE XXX. Multipoles under the hexagonal crystal system (D_{6h}) in the spinful basis. $(g,u)^{\pm} = A^{\pm}_{1g,u} \oplus A^{\mp}_{2g,u} \oplus E^{\mp}_{1g,u}$, $(g',u')^{\pm} = A^{\pm}_{1g,u} \oplus A^{\mp}_{2g,u} \oplus B^{\mp}_{1g,u} \oplus B^{\mp}_{2g,u}$, and $\langle g,u\rangle^{\pm} = B^{\pm}_{1g,u} \oplus B^{\pm}_{2g,u} \oplus E^{\pm}_{2g,u}$. The subscripts g,u are omitted for the D_6 group. The subscripts 1,2 in A and B are omitted for the C_{6h} group. $A_{1u} \leftrightarrow A_{2u}$ and $B_{1g} \leftrightarrow B_{2g}$, and then, the subscripts g,u are omitted for the C_{6v} group. The subscripts g,u and 1,2 in A and B are omitted for the C_6 group. $A_{1g}, B_{1u} \to A'_1$, $A_{2g}, B_{2u} \to A'_2$, $A_{1u}, B_{1g} \to A''_1$, $A_{2u}, B_{2g} \to A''_2$, $E_{2u}, E_{1g} \to E''$, $E_{2g}, E_{1u} \to E'$, and $E_{1/2u} \leftrightarrow E_{3/2u}$ for the D_{3h} group. $A_{1g}, A_{2g}, B_{1u}, B_{2u} \to A'$, $A_{1u}, A_{2u}, B_{1g}, B_{2g} \to A''$, $E_{2u}, E_{1g} \to E''$, $E_{2g}, E_{1u} \to E'$, and $E_{1/2u} \leftrightarrow E_{3/2u}$ for the C_{3h} group.

		(s) $E_{1/2g}$	(p) $E_{1/2u}$	$E_{1/2u}$	$E_{5/2u}$	(d) $E_{1/2g}$	$E_{1/2g}$	$E_{5/2g}$	$E_{3/2g}$	$E_{5/2g}$	(f) $E_{1/2u}$	$E_{3/2u}$	$E_{3/2u}$	$E_{1/2u}$	$E_{5/2u}$	$E_{3/2u}$	$E_{5/2u}$
(s) $A_{1g} \otimes E_{1/2g} \to$	$E_{1/2g}$	$(g)^+$	$(u)^{\pm}$	$(u)^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(g)^{\pm}$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$\langle u\rangle^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$
(p) $A_{2u} \otimes E_{1/2g} \to$	$E_{1/2u}$		$(g)^+$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(u)^{\pm}$	$(u)^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$\langle u\rangle^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$
$E_{1u} \otimes E_{1/2g} \to$	$E_{1/2u}$			$(g)^+$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(u)^{\pm}$	$(u)^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$\langle u\rangle^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$
	$E_{5/2u}$				$(g')^+$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(u')^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(u')^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(g')^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(g')^{\pm}$
(d) $A_{1g} \otimes E_{1/2g} \to$	$E_{1/2g}$					$(g)^+$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$\langle u\rangle^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$
$E_{1g} \otimes E_{1/2g} \to$	$E_{1/2g}$						$(g)^+$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$\langle u\rangle^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$
	$E_{5/2g}$							$(g')^+$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(g')^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(u')^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(u')^{\pm}$
$E_{2g} \otimes E_{1/2g} \to$	$E_{3/2g}$								$(g)^+$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle u\rangle^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$\langle u\rangle^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(u)^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$
	$E_{5/2g}$									$(g')^+$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(u')^{\pm}$	$E^{\pm}_{1u}, E^{\pm}_{2u}$	$(u')^{\pm}$
(f) $A_{2u} \otimes E_{1/2g} \to$	$E_{1/2u}$										$(g)^+$	$\langle g\rangle^{\pm}$	$\langle g\rangle^{\pm}$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$
$B_{1u} \otimes E_{1/2g} \to$	$E_{3/2u}$											$(g)^+$	$(g)^{\pm}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$
$B_{2u} \otimes E_{1/2g} \to$	$E_{3/2u}$												$(g)^+$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(g)^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$
$E_{1u} \otimes E_{1/2g} \to$	$E_{1/2u}$													$(g)^+$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$\langle g\rangle^{\pm}$	$E^{\pm}_{1g}, E^{\pm}_{2g}$
	$E_{5/2u}$														$(g')^+$	$E^{\pm}_{1g}, E^{\pm}_{2g}$	$(g')^{\pm}$
$E_{2u} \otimes E_{1/2g} \to$	$E_{3/2u}$															$(g)^+$	$E^{\pm}_{1g}, E^{\pm}_{2g}$
	$E_{5/2u}$																$(g')^+$

TABLE XXXI. Multipoles under the trigonal crystal system (D_{3d}) in the spinless basis. $(g,u)^{\pm} = A^{\pm}_{1g,u} \oplus A^{\mp}_{2g,u} \oplus E^{\pm}_{g,u}$. The subscripts g,u are omitted for the D_3 group. The subscripts g,u are omitted, and $Q_{3a} \leftrightarrow Q_{3b}$ for the C_{3v} group. The subscripts 1,2 are omitted for the C_{3i} group. The subscripts g,u and 1,2 are omitted for the C_3 group.

	(s) A_{1g}	(p) A_{2u}	E_u	(d) A_{1g}	E_g	E_g	(f) A_{2u}	A_{1u}	A_{2u}	E_u	E_u
(s) A_{1g}	A^+_{1g}	$A^{\pm}_{2u}$	$E^{\pm}_u$	$A^{\pm}_{1g}$	$E^{\pm}_g$	$E^{\pm}_g$	$A^{\pm}_{2u}$	$A^{\pm}_{1u}$	$A^{\pm}_{2u}$	$E^{\pm}_u$	$E^{\pm}_u$
(p) A_{2u}		A^+_{1g}	$E^{\pm}_g$	$A^{\pm}_{2u}$	$E^{\pm}_u$	$E^{\pm}_u$	$A^{\pm}_{1g}$	$A^{\pm}_{2g}$	$A^{\pm}_{1g}$	$E^{\pm}_g$	$E^{\pm}_g$
E_u			$(g)^+$	$E^{\pm}_u$	$(u)^{\pm}$	$(u)^{\pm}$	$E^{\pm}_g$	$E^{\pm}_g$	$E^{\pm}_g$	$(g)^{\pm}$	$(g)^{\pm}$
(d) A_{1g}				A^+_{1g}	$E^{\pm}_g$	$E^{\pm}_g$	$A^{\pm}_{2u}$	$A^{\pm}_{1u}$	$A^{\pm}_{2u}$	$E^{\pm}_u$	$E^{\pm}_u$
E_g					$(g)^+$	$(g)^{\pm}$	$E^{\pm}_u$	$E^{\pm}_u$	$E^{\pm}_u$	$(u)^{\pm}$	$(u)^{\pm}$
E_g						$(g)^+$	$E^{\pm}_u$	$E^{\pm}_u$	$E^{\pm}_u$	$(u)^{\pm}$	$(u)^{\pm}$
(f) A_{2u}							A^+_{1g}	$A^{\pm}_{2g}$	$A^{\pm}_{1g}$	$E^{\pm}_g$	$E^{\pm}_g$
A_{1u}								A^+_{1g}	$A^{\pm}_{2g}$	$E^{\pm}_g$	$E^{\pm}_g$
A_{2u}									A^+_{1g}	$E^{\pm}_g$	$E^{\pm}_g$
E_u										$(g)^+$	$(g)^{\pm}$
E_u											$(g)^+$

TABLE XXXII. Multipoles under the trigonal crystal system (D_{3d}) in the spinful basis. $(g,u)^{\pm} = A^{\pm}_{1g,u} \oplus A^{\mp}_{2g,u} \oplus E^{\mp}_{g,u}$ and $(g',u')^{\mp} = A^{\mp}_{1g,u} \oplus A^{\pm}_{1g,u} \oplus 2A^{\pm}_{2g,u}$. The subscripts g, u are omitted for the D_3 group. The subscripts g, u are omitted, and $Q_{3a} \leftrightarrow Q_{3b}$ for the C_{3v} group. The subscripts 1,2 are omitted for the C_{3i} group. The subscripts g, u and 1,2 are omitted for the C_3 group.

		(s) $E_{1/2g}$	(p) $E_{1/2u}$	$E_{1/2u}$	$E_{3/2u}$	(d) $E_{1/2g}$	$E_{1/2g}$	$E_{3/2g}$	$E_{1/2g}$	$E_{3/2g}$	(f) $E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$	$E_{1/2u}$	$E_{3/2u}$	$E_{1/2u}$	$E_{3/2u}$
(s) $A_{1g} \otimes E_{1/2g} \rightarrow$	$E_{1/2g}$	$(g)^+$	$(u)^{\pm}$	$(u)^{\pm}$	$2E^{\pm}_u$	$(g)^{\pm}$	$(g)^{\pm}$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$2E^{\pm}_u$	$(u)^{\pm}$	$2E^{\pm}_u$
(p) $A_{2u} \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$		$(g)^+$	$(g)^{\pm}$	$2E^{\pm}_g$	$(u)^{\pm}$	$(u)^{\pm}$	$2E^{\pm}_u$	$(u)^{\pm}$	$2E^{\pm}_u$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$
$E_u \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$			$(g)^+$	$2E^{\pm}_g$	$(u)^{\pm}$	$(u)^{\pm}$	$2E^{\pm}_u$	$(u)^{\pm}$	$2E^{\pm}_u$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$
	$E_{3/2u}$				$(g')^+$	$2E^{\pm}_u$	$2E^{\pm}_u$	$(u')^{\pm}$	$2E^{\pm}_u$	$(u')^{\pm}$	$2E^{\pm}_g$	$2E^{\pm}_g$	$2E^{\pm}_g$	$2E^{\pm}_g$	$(g')^{\pm}$	$2E^{\pm}_g$	$(g')^{\pm}$
(d) $A_{1g} \otimes E_{1/2g} \rightarrow$	$E_{1/2g}$					$(g)^+$	$(g)^{\pm}$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$2E^{\pm}_u$	$(u)^{\pm}$	$2E^{\pm}_u$
$E_g \otimes E_{1/2g} \rightarrow$	$E_{1/2g}$						$(g)^+$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$2E^{\pm}_u$	$(u)^{\pm}$	$2E^{\pm}_u$
	$E_{3/2g}$							$(g')^+$	$2E^{\pm}_g$	$(g')^{\pm}$	$2E^{\pm}_u$	$2E^{\pm}_u$	$2E^{\pm}_u$	$2E^{\pm}_u$	$(u')^{\pm}$	$2E^{\pm}_u$	$(u')^{\pm}$
$E_g \otimes E_{1/2g} \rightarrow$	$E_{1/2g}$								$(g)^+$	$2E^{\pm}_g$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$(u)^{\pm}$	$2E^{\pm}_u$	$(u)^{\pm}$	$2E^{\pm}_u$
	$E_{3/2g}$									$(g')^+$	$2E^{\pm}_u$	$2E^{\pm}_u$	$2E^{\pm}_u$	$2E^{\pm}_u$	$(u')^{\pm}$	$2E^{\pm}_u$	$(u')^{\pm}$
(f) $A_{2u} \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$										$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$(g)^{\pm}$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$
$A_{1u} \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$											$(g)^+$	$(g)^{\pm}$	$(g)^{\pm}$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$
$A_{2u} \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$												$(g)^+$	$(g)^{\pm}$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$
$E_u \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$													$(g)^+$	$2E^{\pm}_g$	$(g)^{\pm}$	$2E^{\pm}_g$
	$E_{3/2u}$														$(g')^+$	$2E^{\pm}_g$	$(g')^{\pm}$
$E_u \otimes E_{1/2g} \rightarrow$	$E_{1/2u}$															$(g)^+$	$2E^{\pm}_g$
	$E_{3/2u}$																$(g')^+$

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