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Igor Proskurin^{1,2}, Alexander S Ovchinnikov^{2,3}, Pavel Nosov² and Jun-ichiro Kishine⁴¹ Faculty of Science, Hiroshima University, Higashi-Hiroshima, Hiroshima 739-8526, Japan² Institute of Natural Sciences, Ural Federal University, Ekaterinburg 620002, Russia³ Institute for Metal Physics, RAS, 620137, Ekaterinburg, Russia⁴ Division of Natural and Environmental Sciences, The Open University of Japan, Chiba 261-8586, JapanE-mail: iprosk@ouj.ac.jp**Keywords:** optical chirality, nongeometric symmetries, Lipkin's zilch, gyrotropic crystals, natural optical activity**Abstract**

We discuss optical chirality in different types of gyrotropic media. Our analysis is based on the formalism of nongeometric symmetries of Maxwell's equations in vacuum generalized to material media with given constituent relations. This approach enables us to directly derive conservation laws related to nongeometric symmetries. For isotropic chiral media, we demonstrate that like a free electromagnetic field, both duality and helicity generators belong to the basis set of nongeometric symmetries that guarantees the conservation of optical chirality. In gyrotropic crystals, which exhibit natural optical activity, the situation is quite different from the case of isotropic media. For light propagating along a certain crystallographic direction, there arises two distinct cases: (1) the duality is broken but the helicity is preserved, or (2) only the duality symmetry survives. We show that the existence of one of these symmetries (duality or helicity) is enough to define optical chirality. In addition, we present examples of low-symmetry media, where optical chirality cannot be defined.

1. Introduction

The notion of chirality, the term originally coined by Lord Kelvin for any object that cannot be superimposed onto its mirror image [1], is perhaps one of the most fundamental concepts in nature ranging from particle physics to biology [2]. After the discovery of a new conservation law for Maxwell's equations in vacuum by Lipkin [3], it was realized that not only material objects but also fields can be characterized by a certain chirality [4, 5]. In particular, we can construct the conserving pseudoscalar for a free electromagnetic field

$$C_\chi = \frac{1}{2} \int d^3r (\epsilon_0 \mathbf{E} \cdot \nabla \times \mathbf{E} + \mu_0^{-1} \mathbf{B} \cdot \nabla \times \mathbf{B}), \quad (1)$$

which is even under time-reversal (T) and odd under spatial inversion (P) transformations. These symmetry properties are consistent with the definition of *true chirality* proposed by Barron [6, 7], who stressed that we should distinguish it from *false chirality* with broken T -symmetry. In this respect, this quantity is eligible to be called optical chirality, which was originally coined *zilch* by Lipkin [3].

Later it was realized that besides classical conservation laws arising from the invariance under the space-time Poincaré group, a free electromagnetic field is invariant under the eight-dimensional Lie algebra of nongeometric symmetry transformations. This algebra results in an infinite number of integro-differential conservation laws, which include zilch as a particular case [8]. Together with the approach based on nongeometric symmetries [8], these conservation laws were studied using the Lie theory and the Noether theorem [9–14], using the analogy between the Maxwell and Dirac equations [15], and through gauge symmetry [16].

For a free electromagnetic field, it was demonstrated that the existence of duality symmetry for Maxwell's equations, i.e. the linear transformation that mixes electric and magnetic fields, automatically engenders the preservation of optical helicity, i.e. the projection of total angular momentum on the direction of linear momentum [17–20].

The renewed interest in optical chirality has been stimulated through interdisciplinary studies in molecules and metamaterials [21–26]. Notable progress in the field of optical angular momentum is also relevant to these directions [27, 28]. The relationship between optical chirality, helicity, and spin angular momentum of light was discussed in a number of papers [29–37]. A recent review on the general space-time symmetries of Maxwell’s equations can be found in [38].

For an electromagnetic field in media, the problem of optical chirality has been considered in [16, 39, 40]. In dispersive media, the generalization of Lipkin’s zilch was established by Philbin [16] by means of Noether’s theorem applied to the specific gauge transformation of the magnetic vector potential. In isotropic chiral media, first-order electromagnetic conservation laws were heuristically constructed by Ragusa in the relativistically noncovariant [39] and covariant forms [40].

The purpose of this paper is to develop a systematic approach to the optical chirality in gyrotropic media. To provide a theoretical basis for our treatment, we invoke the formalism of nongeometric symmetries in vacuum [8], and generalize it to media taking heed of the corresponding constituent equations. The key idea is to find the invariance algebra of nongeometric symmetries in media, and to investigate whether the basis set of this algebra includes the transformations of duality and helicity.

First, we analyze nongeometric symmetries in isotropic chiral media. Like an electromagnetic field in vacuum [17, 18], an isotropic chiral medium is self-dual, which automatically means that helicity is preserved [41]. However, as we explicitly show, in chiral media, the original eight-dimensional invariance algebra of the free electromagnetic field is broken down to its four-dimensional subalgebra due to lack of inversion symmetry. Duality and helicity are two essential generators of this subalgebra. For a spatially nonuniform chiral medium, we find that the actual expression for optical chirality depends on the choice of constituent relations in order to guarantee continuity of the chirality flow.

Next, we consider optical chirality in gyrotropic crystals, where surrounding symmetry is more restrictive, and, globally, neither duality nor helicity symmetry transformations are allowed. Although these symmetries can survive along the principal crystalline axes, the equivalence between duality and helicity that holds in isotropic media is lost. We find that in crystals with gyrotropic birefringence both duality and helicity operations are allowed along the principal axis. Crystals possessing natural optical activity and belonging to point groups C_n or C_{nv} ($n \geq 3$) provide an example, where helicity is preserved along the principal axis [42] while the duality symmetry is broken. The opposite situation is realized in achiral materials with natural optical activity, where the duality symmetry along the principal axis is preserved without helicity. The existence of either duality or helicity transformations along a certain direction leads to the conservation law for optical chirality, which is consistent with underlying symmetries.

Conclusively, we consider low-dimensional crystals, where we encounter the invariance algebra with neither preserved duality nor preserved helicity, which makes optical chirality ill-defined.

2. Free electromagnetic field

We begin with a brief review of the latest developments in symmetry analysis of the electromagnetic field in vacuum. We give a pedagogical introduction in the method of nongeometric symmetries, which is generalized in subsequent sections to an electromagnetic field in media.

The discovery of a conservation law in equation (1) stimulated the discussion of the related ‘hidden’ symmetries for the electromagnetic field. Historically, it had been established shortly after the formulation of electrodynamics that Maxwell’s equations in free space

$$\nabla \times E = -\partial_t B, \quad \nabla \times B = \partial_t E, \quad (2)$$

$$\nabla \cdot E = 0, \quad \nabla \cdot B = 0, \quad (3)$$

(in this section we use $c = 1$ and $\hbar = 1$) remain invariant under the duality transformation

$$E \rightarrow E \cos \theta + B \sin \theta, \quad (4)$$

$$B \rightarrow -E \sin \theta + B \cos \theta, \quad (5)$$

which can be viewed as ‘rotation’ in the pseudospace of E and B vectors (for a review and historical background, see [12, 13]).

In contrast to Maxwell’s equations, the standard Lagrangian formulation of electrodynamics is not symmetric under the duality transformation, and, to mitigate this obstacle, a duality symmetric form of the Lagrangian density was proposed [12, 13]. The duality symmetric form, on the basis of the Noether theorem, ties up the transformation in equations (4) and (5) with conservation of optical helicity,

$$C_{\text{hel}} = \frac{1}{2} \int d^3r (A \cdot \nabla \times A + C \cdot \nabla \times C), \quad (6)$$

written in terms of magnetic (A) and electric (C) vector potentials, which are determined by $E = -\nabla \times C = -\partial_t A$ and $B = \nabla \times A$, and satisfy the symmetry transformation in equations (4) and (5).

With respect to the optical helicity, Lipkin's zilch in equation (1) can be regarded as the next-order term in the infinite hierarchy of higher-order conserving zilches [5, 32]. Although being different from optical helicity in general, for monochromatic fields, Lipkin's zilch becomes proportional to the helicity. Both quantities are determined by the difference between left and right polarized photon numbers [30, 31]. Tang and Cohen proposed to use Lipkin's 00-zilch as a measure of *optical chirality* in light-matter interactions [21, 23].

A different approach to zilch conservation laws has been developed in [9–11] on the basis of the Lie–Noether analysis. Recently, Philbin explicitly demonstrated that zilch conservation can be obtained from the standard electromagnetic Lagrangian $L = (E^2 - B^2)/2$ by applying a specific 'hidden' gauge symmetry transformation of the magnetic vector potential, $A \rightarrow A + \eta \nabla \times \partial_t A$, with infinitesimal parameter η [16], similar to Calkin's original arguments [17].

Another powerful tool, different from the Lagrangian–Noether approach, that we used throughout this paper, is a method of the nongeometric symmetries developed by Fushchich and Nikitin [8]. The advantage of this method is that it is solely based on the analysis of the equations of motion and, therefore, does not rely on any ambiguity in specific gauge choice or Lagrangian representation. This fact makes it possible to generalize this approach to Maxwell's equations in media with given constituent relations.

For the symmetry analysis, it is essential to find convenient representation of Maxwell's equations. We use the Silberstein–Bateman form, which is convenient to work in the momentum space. The transformation to the momentum space is reached by

$$E(t, \mathbf{r}) = \frac{1}{(2\pi)^{3/2}} \int d^3p e^{i\mathbf{p}\cdot\mathbf{r}} E(t, \mathbf{p}), \quad (7)$$

$$B(t, \mathbf{r}) = \frac{1}{(2\pi)^{3/2}} \int d^3p e^{i\mathbf{p}\cdot\mathbf{r}} B(t, \mathbf{p}). \quad (8)$$

In the Silberstein–Bateman representation, the first pair of Maxwell's equations in equation (2) is expressed in terms of a Shroedinger-like equation for a six-component vector column $\phi(t, \mathbf{p}) = (E, B)^T$

$$i \frac{\partial \phi(t, \mathbf{p})}{\partial t} = \mathcal{H} \phi(t, \mathbf{p}), \quad (9)$$

where \mathcal{H} is the Hermitian matrix given by

$$\mathcal{H} = -\sigma_2 \otimes (\hat{S} \cdot \mathbf{p}) = \begin{pmatrix} 0 & i(\hat{S} \cdot \mathbf{p}) \\ -i(\hat{S} \cdot \mathbf{p}) & 0 \end{pmatrix}, \quad (10)$$

which can be considered as an analog of the quantum-mechanical Hamiltonian. Here, we introduced 3×3 spin matrices \hat{S}_α with matrix elements $(\hat{S}_\alpha)_{\beta\gamma} = -i\epsilon_{\alpha\beta\gamma}$, where $\epsilon_{\alpha\beta\gamma}$ is the Levi-Civita symbol, \otimes means the Cartesian product, and σ_μ ($\mu = 1, 2, 3$) are 2×2 Pauli matrices. In what follows, we hold the following notations. The 'hat' is used to distinguish 3×3 matrices. Calligraphic style is reserved for the 6×6 matrices. σ_0 and \hat{I} denote two- and three-dimensional unit matrices, and Greek indices run over the three-dimensional space.

The second pair of Maxwell's equation (3) is equivalent to the additional constraint imposed on $\phi(t, \mathbf{p})$ [8]:

$$\mathcal{L} \phi(t, \mathbf{p}) = 0, \quad \mathcal{L} = p^2 - (\mathcal{S} \cdot \mathbf{p})^2, \quad (11)$$

where $\mathcal{S} \equiv \sigma_0 \otimes \hat{S}$, which accounts for the transversal character of the electromagnetic field. For real $\phi(t, \mathbf{p})$, one should also require $\phi^*(t, \mathbf{p}) = \phi(t, -\mathbf{p})$.

Now let us find all the transformations in the \mathbf{p} -space given by matrices $\mathcal{Q}_A(\mathbf{p})$ that transform a solution of Maxwell's equations $\phi(t, \mathbf{p})$ into another solution $\phi'(t, \mathbf{p}) = \mathcal{Q}_A(\mathbf{p}) \phi(t, \mathbf{p})$. Following [8], we will call $\mathcal{Q}_A(\mathbf{p})$ *nongeometric symmetry* transformations. The total number of such $\mathcal{Q}_A(\mathbf{p})$ is given in the theorem, which claims that Maxwell's equations in vacuum are invariant under the eight-dimensional Lie algebra:

$$\mathcal{Q}_1 = \sigma_3 \otimes (\hat{S} \cdot \tilde{\mathbf{p}}) \hat{D}, \quad \mathcal{Q}_2 = i\sigma_2 \otimes \hat{I}, \quad (12)$$

$$\mathcal{Q}_3 = -\sigma_1 \otimes (\hat{S} \cdot \tilde{\mathbf{p}}) \hat{D}, \quad \mathcal{Q}_4 = -\sigma_1 \otimes \hat{D}, \quad (13)$$

$$\mathcal{Q}_5 = \sigma_0 \otimes (\hat{S} \cdot \tilde{\mathbf{p}}), \quad \mathcal{Q}_6 = -\sigma_3 \otimes \hat{D}, \quad (14)$$

$$\mathcal{Q}_7 = \sigma_0 \otimes \hat{I}, \quad \mathcal{Q}_8 = i\sigma_2 \otimes (\hat{S} \cdot \tilde{\mathbf{p}}), \quad (15)$$

where $\tilde{\mathbf{p}} = \mathbf{p}/p$, and $\hat{D} = \hat{D}_0 + \hat{D}_1$ with $(\hat{D}_0)_{\alpha\beta} = (\delta - f)p_\alpha p_\beta / (p^2 \delta)$ and

$$\hat{D}_1 = \frac{1}{\delta} \begin{pmatrix} f - 2p_2^2 p_3^2 & p_1 p_2 p_3^2 & p_1 p_3 p_2^2 \\ p_1 p_2 p_3^2 & f - 2p_1^2 p_3^2 & p_2 p_3 p_1^2 \\ p_1 p_3 p_2^2 & p_2 p_3 p_1^2 & f - 2p_1^2 p_2^2 \end{pmatrix}, \quad (16)$$

where $f = p_1^2 p_2^2 + p_1^2 p_3^2 + p_2^2 p_3^2$ and $\delta = [p_1^4 (p_2^2 - p_3^2)^2 + p_2^2 p_3^2 (p_1^2 - p_2^2)(p_1^2 - p_3^2)]^{1/2}$. The basis elements form the algebra, which is isomorphic to the Lie algebra of the group $U(2) \otimes U(2)$ [8].

All the basis elements in equations (12)–(15) commute with \mathcal{H} in equation (10) and act as generators for the continuous symmetry transformations

$$\phi(t, \mathbf{p}) \rightarrow \exp(\mathcal{Q}_A \theta_A) \phi(t, \mathbf{p}), \quad (17)$$

where θ_A are real parameters.

Analogy with quantum mechanics suggests that one can find conserving quantities related to the symmetry transformations \mathcal{Q}_A , which can be conveniently formulated in terms of bilinear forms:

$$\langle \mathcal{Q}_A \rangle = \frac{1}{2} \int d^3 p \phi^\dagger(t, \mathbf{p}) \mathcal{M} \mathcal{Q}_A \phi(t, \mathbf{p}), \quad (18)$$

where \mathcal{M} is any operator commutative with \mathcal{H} .

Some symmetry transformations in equations (12)–(15) have transparent physical interpretations. For example, \mathcal{Q}_7 is the identity transformation. The corresponding conserving quantity is the electromagnetic energy

$$\langle \mathcal{Q}_7 \rangle = \frac{1}{2} \int d^3 p \phi^\dagger(t, \mathbf{p}) \mathcal{Q}_7 \phi(t, \mathbf{p}) = \frac{1}{2} \int d^3 p (E^2 + B^2). \quad (19)$$

The duality transformation in equations (4) and (5) also belongs to the class on nongeometric symmetries. This symmetry is generated by \mathcal{Q}_2 , in accordance with equation (17).

Electromagnetic chirality can be expressed as a conserving quantity that corresponds to the operator $p \mathcal{Q}_5$. Indeed, the expression

$$C_\chi = \frac{1}{2} \int d^3 p \phi^\dagger(t, \mathbf{p}) (\mathbf{S} \cdot \mathbf{p}) \phi(t, \mathbf{p}) \quad (20)$$

is transformed into equation (1) in the real space. In what follows, we would refer to \mathcal{Q}_5 as a *helicity operator*, since electromagnetic helicity in equation (6) can be also expressed in terms of a bilinear form containing this operator acting in the space (A, C) .

By noting that $p \mathcal{Q}_5 \mathcal{Q}_2 = -i\mathcal{H} = \partial_t$, we can find an alternative form of the optical chirality expressed via the duality operator

$$C_\chi = -\frac{i}{2} \int d^3 p \phi^\dagger(t, \mathbf{p}) \mathcal{Q}_2 \partial_t \phi(t, \mathbf{p}). \quad (21)$$

However, as we show in the next sections, the identity above between \mathcal{Q}_5 , \mathcal{Q}_2 , and \mathcal{H} does not necessary hold in crystals, where it is possible that either \mathcal{Q}_2 or \mathcal{Q}_5 is allowed symmetry along a certain crystallographic direction, but not both simultaneously.

Using the ambiguity in the choice of \mathcal{M} in equation (18), we can identify the hierarchy of higher-rank conserving zilches, which contain high-order derivatives of electromagnetic fields. Substituting $\mathcal{M} = (-1)^{n+m} p^{2n} \mathcal{H}^{2m+1}$ into equation (21) (apparently, it commutes with \mathcal{H}), we obtain the following conserving quantities:

$$C_\chi^{(m,n)} = \frac{1}{2} \int d^3 r (\mathbf{B} \cdot \nabla^{2n} \partial_t^{2m+1} \mathbf{E} - \mathbf{E} \cdot \nabla^{2n} \partial_t^{2m+1} \mathbf{B}), \quad (22)$$

which were found previously by other methods (see e.g. [16, 19, 20]).

3. General formalism in media

The formalism of nongeometric symmetries can be generalized to Maxwell's equations in medium, where symmetry of constituent relations imposes additional constraints on the form of conservation laws. In general, this leads to the reduction of the original eight-dimensional invariance algebra A_8 to a smaller number of elements. In this section, we analyze the situation when the electromagnetic field propagates in a time-independent dielectric medium. We show that, basically, A_8 shrinks to one of its commutative subalgebras A_4 with four basis elements. In the subsequent sections, we demonstrate that this situation is common for isotropic chiral media as far as for chiral gyrotropic crystals when light spreads along the principal symmetry direction.

It should be mentioned that the existence of A_4 symmetry does not guarantee the conservation of optical chirality, as we discuss at the end of this section.

On a macroscopic level, Maxwell's equations in dielectric media can be expressed as follows:

$$\nabla \times \mathbf{E} = -\partial_t \mathbf{B}, \quad \nabla \times \mathbf{H} = \partial_t \mathbf{D}, \quad (23)$$

$$\nabla \cdot \mathbf{D} = 0, \quad \nabla \cdot \mathbf{B} = 0, \quad (24)$$

which should be accompanied by constituent relations. In what follows, we consider the following form of constituent relations:

$$\begin{pmatrix} \mathbf{D}(t, \mathbf{p}) \\ \mathbf{B}(t, \mathbf{p}) \end{pmatrix} = \mathcal{A}(\mathbf{p}) \begin{pmatrix} \mathbf{E}(t, \mathbf{p}) \\ \mathbf{H}(t, \mathbf{p}) \end{pmatrix}, \quad (25)$$

where the momentum representation is used. The matrix $\mathcal{A}(\mathbf{p})$ is supposed to be time-independent and determined by the properties of the medium.

For symmetry analysis in the medium, it is convenient to introduce the Silberstein–Bateman vector $\psi(t, \mathbf{p}) = (\mathbf{D}, \mathbf{B})^T$. In this case, the first pair of Maxwell's equations is written as

$$\mathcal{L}^{(A)} \psi = 0, \quad \mathcal{L}^{(A)} = i\partial_t - \mathcal{H}^{(A)}, \quad (26)$$

where

$$\mathcal{H}^{(A)} = -\sigma_2 \otimes (\hat{\mathbf{S}} \cdot \mathbf{p}) \mathcal{A}^{-1}, \quad (27)$$

and the constraint on $\psi(t, \mathbf{p})$ imposed by the second pair of Maxwell's equations is the same as in equation (11).

For a common situation when constituent relations do not mix up electric and magnetic fields, equation (27) is reduced to the following expression:

$$\mathcal{H}^{(A)} = \begin{pmatrix} 0 & i(\hat{\mathbf{S}} \cdot \mathbf{p}) \hat{\mu}^{-1}(\mathbf{p}) \\ -i(\hat{\mathbf{S}} \cdot \mathbf{p}) \hat{\varepsilon}^{-1}(\mathbf{p}) & 0 \end{pmatrix}, \quad (28)$$

where $\hat{\varepsilon}^{-1}(\mathbf{p})$ and $\hat{\mu}^{-1}(\mathbf{p})$ are inverse permittivity and permeability tensors.

3.1. Nongeometric symmetries

In general, it may be a tedious problem to find symmetry transformations for Maxwell's equations in a medium. However, the task is alleviated along the directions where transverse electromagnetic waves can propagate. Mathematically, it corresponds to $\mathcal{A}(\mathbf{p})$ being commutative with \mathcal{L} in equation (11) along such directions. In this case, to identify possible symmetry transformations, we apply a transformation $\bar{\psi} = \mathcal{N}^{-1}\psi$ to the basis, where $\mathcal{H}^{(A)}$ and \mathcal{L} are both diagonal:

$$\bar{\mathcal{H}}^{(A)} = \mathcal{N}^{-1}\mathcal{H}^{(A)}\mathcal{N} = \text{diag}(\omega_1, \omega_2, 0, \omega_4, \omega_5, 0), \quad (29)$$

$$\bar{\mathcal{L}} = \mathcal{N}^{-1}\mathcal{L}\mathcal{N} = \text{diag}(0, 0, p^2, 0, 0, p^2), \quad (30)$$

where parameters $\omega_i = \omega_i(\mathbf{p})$ are real-valued in the absence of dissipation. Note that, since in the original basis $\mathcal{H}^{(A)}$ is not necessary a Hermitian matrix, the transformation \mathcal{N} may be nonunitary.

For the diagonal operators in equations (29) and (30), it is easy to find the invariance algebra. The number of basis elements in the invariance algebra depends on the symmetry relations between ω_i (see appendix A for mathematical aspects of the derivation). In the absence of any degeneracies between ω_i , all symmetry transformations in the $\bar{\psi}$ -basis are given by four diagonal operators. The basis in this four-dimensional linear space can be chosen as follows:

$$\bar{\mathcal{Q}}_a = \text{diag}(1, 1, 0, -1, -1, 0), \quad (31)$$

$$\bar{\mathcal{Q}}_b = \text{diag}(-1, 1, 0, -1, 1, 0), \quad (32)$$

$$\bar{\mathcal{Q}}_c = \text{diag}(1, 1, 0, 1, 1, 0), \quad (33)$$

$$\bar{\mathcal{Q}}_d = \text{diag}(-1, 1, 0, 1, -1, 0), \quad (34)$$

which can be conveniently expressed by the Kronecker product of $\{\sigma_0, \sigma_3\}$ and $\{\hat{I}, \hat{\Gamma}\}$, where $\hat{\Gamma} = \text{diag}(-1, 1, 0)$, which forms the Klein four-group isomorphic to the direct sum $\mathbb{Z}_2 \oplus \mathbb{Z}_2$ [43]. The form of these operators in the original basis ψ is reached by the inverse transformation $\psi = \mathcal{N}\bar{\psi}$.

From equations (31)–(34), we conclude that a radiation field in media is invariant at least under the four commutative symmetry transformations, which we will refer to as A_4 symmetries. This group has two trivial elements that correspond to the identity and $i\partial_t \equiv \mathcal{H}^A$. The latter simply states that in a time-independent medium the time derivative of the solution is again the solution. The physical meaning of the other two elements is determined by the constituent relations encoded into the transformation \mathcal{N} . Let us note that A_4 is a minimal

symmetry. In the case of additional degeneracies between different ω_i in equation (29), A_4 becomes a subalgebra of a larger invariance algebra.

For illustration, we consider a free electromagnetic field. In vacuum, the diagonal form of \mathcal{H} in equation (10) is reached by unitary transformation $\psi = \mathcal{U}\bar{\psi}$, where $\mathcal{U} = U_2 \otimes \hat{U}_\Lambda$ combines the transformation to the helicity basis:

$$\hat{U}_\Lambda = \begin{pmatrix} \frac{p_1 p_3 + i p_2 p}{\sqrt{2} p p_\perp} & \frac{p_1 p_3 - i p_2 p}{\sqrt{2} p p_\perp} & \frac{p_1}{p} \\ -\frac{p_2 p_3 - i p_1 p}{\sqrt{2} p p_\perp} & \frac{p_2 p_3 + i p_1 p}{\sqrt{2} p p_\perp} & \frac{p_2}{p} \\ \frac{p_1}{\sqrt{2} p} & -\frac{p_1}{\sqrt{2} p} & \frac{p_3}{p} \end{pmatrix}, \quad (35)$$

where $(\hat{S} \cdot \mathbf{p})$ is diagonal, and with $SU(2)$ rotation in the pseudospace of D and B vectors

$$U_2 = \frac{1}{\sqrt{2}}(\sigma_0 - i\sigma_1). \quad (36)$$

Comparing the resulting diagonal form of \mathcal{H} ,

$$\tilde{\mathcal{H}} = \mathcal{U}^\dagger \mathcal{H} \mathcal{U} = p\sigma_3 \otimes \hat{\Gamma} = \text{diag}(-p, p, 0, p, -p, 0), \quad (37)$$

with equation (29), we find that $\tilde{\mathcal{H}}$ has two degeneracies between ω_i , namely, $\omega_1 = \omega_5 = -p$ and $\omega_2 = \omega_4 = p$. The existence of these degeneracies, according to (A6), means that the symmetry transformation \bar{Q}_A in (A5) contains in total eight free parameters: $q_{11}, q_{22}, q_{44}, q_{55}, q_{15}, q_{24}, q_{51}$, and q_{42} . In the original basis $\psi = \mathcal{U}^{-1}\bar{\psi}$, it gives rise to the eight-dimensional algebra A_8 in equations (12)–(15). Under the inverse transformation, four diagonal operators in equations (31)–(34) transform into $\bar{Q}_a \rightarrow \bar{Q}_2, \bar{Q}_b \rightarrow \bar{Q}_5, \bar{Q}_c \rightarrow \bar{Q}_7$, and $\bar{Q}_d \rightarrow \bar{Q}_8$.

3.2. Optical chirality

The generalization of the conservation laws in equation (18) in media is straightforward. We define conserving quantities as

$$\langle \mathcal{Q}_A \rangle = \frac{1}{2} \int d^3p \psi^\dagger(t, \mathbf{p}) \rho(\mathbf{p}) \mathcal{Q}_A(\mathbf{p}) \psi(t, \mathbf{p}), \quad (38)$$

where \mathcal{Q}_A is one of the symmetry transformations. The scalar product is modified by $\rho = (\mathcal{N}^{-1})^\dagger \mathcal{N}^{-1}$ for nonunitary \mathcal{N} . For any Hermitian matrix \mathcal{H}^A , the property $\rho = 1$ is restored.

In general, the existence of the symmetries defined in equations (31)–(34) does not automatically ensure the conservation law for optical chirality. It can be introduced if the invariance algebra contains an element that yields a conserving pseudoscalar C_χ , which is simultaneously even under time-reversal and odd under spatial inversion symmetries.

The existence of C_χ is justified in a medium that has duality symmetry [41, 44]. In this case, the duality transformation $\mathcal{Q}_{\text{dual}} = i\rho^{-1}(\sigma_2 \otimes \hat{I})$ is one of the symmetries, and C_χ can be introduced as a conservation of $\mathcal{Q}_A = \mathcal{Q}_{\text{dual}}\partial_t$ in equation (38) that eventually leads to the following form of optical chirality:

$$C_\chi^{(\text{dual})} = \frac{1}{2} \int d^3p (B^* \cdot \partial_t D - D^* \cdot \partial_t B). \quad (39)$$

The general form of the constituent relations in media, which preserves the duality transformation was obtained in [41]. It was demonstrated that a necessary and sufficient condition for the system to remain self-dual is commutativity of the duality transformation generator and the matrix of constituent relations.

Unlike a free electromagnetic field, where conservation of $\mathcal{Q}_{\text{dual}}\partial_t$ is equivalent to the helicity conservation (see equations (20) and (21)), in media it is possible that the helicity is conserved even if the system does not have dual symmetry. In this case, we define C_χ as a conservation of the $(\mathcal{S} \cdot \mathbf{p})$ operator in equation (38). A general criterion for the helicity conservation is commutativity of the $(\mathcal{S} \cdot \mathbf{p})$ operator with the matrix of constituent relations \mathcal{A} .

Note that there is no ambiguity in the definition of optical chirality if the system is invariant under both the duality symmetry and the helicity transformations. In this case, these two elements belong to the same set of transformations in equations (31)–(34), which means that the product $\mathcal{Q}_{\text{dual}}\partial_t$ is a linear combination of other symmetry elements of A_4 that include $(\mathcal{S} \cdot \mathbf{p})$.

4. Optical chirality in media

In dielectric media, optical activity is a usual manifestation of microscopic structural chirality. This effect, in general, is related to noncentrosymmetry and is shared by both chiral and achiral materials [45]. On a macroscopic level, optical activity can be described by proper constituent relations. Historically, constituent

relations describing natural optical activity were first developed in the Born–Drude–Fedorov (BDF) form [46–49]:

$$\mathbf{D} = \hat{\varepsilon}\varepsilon_0(\mathbf{E} + \hat{\alpha}\nabla \times \mathbf{E}), \quad (40)$$

$$\mathbf{B} = \hat{\mu}\mu_0(\mathbf{H} + \hat{\alpha}^T\nabla \times \mathbf{H}), \quad (41)$$

where $\hat{\varepsilon}$ and $\hat{\mu}$ are the electric permittivity and the magnetic permeability tensors, and $\hat{\alpha}$ is the gyration tensor ($\hat{\alpha}^T$ means transposed $\hat{\alpha}$).

Another form of constituent relation that features optical rotation, which we will refer to as chiral magnetoelectric (CME) constituent relations, comes from the general relativity covariance principle [49, 50] and can be written as follows:

$$\mathbf{D} = \hat{\varepsilon}\varepsilon_0\mathbf{E} + i\hat{\varkappa}\mathbf{H}, \quad (42)$$

$$\mathbf{B} = \hat{\mu}\mu_0\mathbf{H} - i\hat{\varkappa}^T\mathbf{E}, \quad (43)$$

where $\hat{\varkappa}$ is the magnetoelectric tensor [51]. We note that these relations are usually formulated in the frequency domain for time-harmonic electromagnetic fields with the frequency factor ω being included in $\hat{\varkappa}$. In what follows, when we use CME relations, we imply that the formalism of complex time-harmonic fields is used, where time dependencies are given by $\exp(-i\omega t)$.

The CME form of constituent relations is frequently used in chiral metamaterials [26, 52] and in crystals with gyrotropic birefringence [53, 54]. The mutual relation between the BDF and CME equations has been studied by several authors [55–57].

4.1. Isotropic chiral media

Let us first consider optical chirality in isotropic chiral media characterized by constituent relations of either BDF or CME types. Both types share a number of common features, and therefore, we carry out the discussion in parallel. To avoid redundant complications, we use the units where $\varepsilon\varepsilon_0 = \mu\mu_0 = 1$ and restore SI units whenever necessary.

The matrix form of constituent relations in the momentum space is given by equation (25) with

$$\mathcal{A}(\mathbf{p}) = \begin{cases} \sigma_0 \otimes (\hat{I} + \alpha(\hat{\mathbf{S}} \cdot \mathbf{p})), & \text{BDF,} \\ (\sigma_0 - \varkappa\sigma_2) \otimes \hat{I}, & \text{CME,} \end{cases} \quad (44)$$

where the upper (lower) line is for constituent relations in equations (40) and (41) (equations (42) and (43)), which thus provides the Hermitian matrix

$$\mathcal{H}^{(A)} = \begin{cases} -\sigma_2 \otimes \frac{(\hat{\mathbf{S}} \cdot \mathbf{p}) - \alpha(\hat{\mathbf{S}} \cdot \mathbf{p})^2}{1 - \alpha^2 p^2}, & \text{BDF,} \\ -(\sigma_2 + \varkappa\sigma_0) \otimes \frac{(\hat{\mathbf{S}} \cdot \mathbf{p})}{1 - \varkappa^2}, & \text{CME,} \end{cases} \quad (45)$$

which is invariant under both duality and helicity transformations.

To find the complete set of nongeometric symmetries, we diagonalize $\mathcal{H}^{(A)}$ by applying the same unitary transformation as in vacuum, $\mathcal{U} = U_2 \otimes \hat{U}_\Lambda$, where \hat{U}_Λ and U_2 are defined in equations (35) and (36), respectively, which leads to the diagonal form

$$\tilde{\mathcal{H}}^{(A)} = \text{diag}(-p_\pm, p_\pm, 0, p_\pm, -p_\pm, 0), \quad (46)$$

with the upper (lower) sign for the BDF (CME) constituent relation, and $p_\pm = p(1 \mp \alpha p)^{-1}$ ($p_\pm = p(1 \mp \varkappa)^{-1}$) for BDF (CME).

In chiral media, symmetry breaking between left and right polarized states removes the degeneracy between the eigenvalues of $\tilde{\mathcal{H}}^{(A)}$, and according to equations (29) and (A5), the set of nongeometric symmetries is reduced to four elements with the following basis:

$$\mathcal{Q}_2 = i\sigma_2 \otimes \hat{I}, \quad \mathcal{Q}_5 = \sigma_0 \otimes (\hat{\mathbf{S}} \cdot \tilde{\mathbf{p}}), \quad (47)$$

$$\mathcal{Q}_7 = \sigma_0 \otimes \hat{I}, \quad \mathcal{Q}_8 = i\sigma_2 \otimes (\hat{\mathbf{S}} \cdot \tilde{\mathbf{p}}), \quad (48)$$

which includes both duality (\mathcal{Q}_2) and helicity (\mathcal{Q}_5) transformations. Therefore, in isotropic chiral media, similar to the case of a free electromagnetic field, we can say that duality symmetry is related to the helicity conservation.

Since duality symmetry is preserved, Lipkin's zilch is directly obtained from equation (39), which in the \mathbf{r} -space is written as

$$C_{\chi}^{(\text{iso})} = \frac{1}{2} \int d^3r (B^* \cdot \partial_t D - D^* \cdot \partial_t B). \quad (49)$$

Here, and in equations (50)–(55), we hold the following convention. For BDF constituent relations one has to remove complex conjugations for the fields in r -space. In contrast, for CME relations all fields are supposed to be time-harmonic complex fields, and ∂_t should be replaced by $-i\omega$ in final expressions. The transformation to SI units in equation (49) is rendered by substitutions

$$\begin{aligned} D &\rightarrow D(\varepsilon\varepsilon_0)^{-1}, & B &\rightarrow B(\varepsilon\varepsilon_0\mu\mu_0)^{-\frac{1}{2}}, \\ H &\rightarrow H\left(\frac{\mu\mu_0}{\varepsilon\varepsilon_0}\right)^{\frac{1}{2}}, & t &\rightarrow t(\varepsilon\varepsilon_0\mu\mu_0)^{-\frac{1}{2}}, \end{aligned}$$

supplemented by $C_{\chi}^{(\text{iso})} \rightarrow \varepsilon\varepsilon_0 C_{\chi}^{(\text{iso})}$.

We emphasize that in infinite homogeneous medium optical chirality can be expressed in several equivalent forms. For example, instead of the operator $\mathcal{Q}_2\partial_t$ that gives the conservation law in equation (49), we can consider another symmetry operation $\mathcal{Q}_2\mathcal{A}^{-1}\partial_t$, which leads to

$$C_{\chi}^{(\text{iso})} = \frac{1}{2} \int d^3r (B^* \cdot \partial_t E - D^* \cdot \partial_t H). \quad (50)$$

However, in the realistic case, we should also care about the conservation of chirality flow across the boundaries separating different media, which removes this ambiguity.

The situation with several forms of optical chirality is not new. A similar situation occurs with the energy density in chiral materials. In the absence of boundaries, energy density can be also expressed in several equivalent forms. However, only one form guarantees proper energy balance across the boundary between two chiral media [49]. It was demonstrated by Fedorov [49] that the physical form of energy density depends on the choice of constituent relations as follows:

$$\mathfrak{E} = \frac{1}{2} \begin{cases} (\varepsilon\varepsilon_0)^{-1}D \cdot D + (\mu\mu_0)^{-1}B \cdot B, & \text{BDF,} \\ E^* \cdot D + H^* \cdot B, & \text{CME.} \end{cases} \quad (51)$$

We anticipate that similar to energy density, optical chirality for BDF and CME constituent relations should be taken in different forms.

The situation becomes more transparent in the spatially nonuniform space where $\varepsilon(\mathbf{r})$, $\mu(\mathbf{r})$, $\alpha(\mathbf{r})$, and $\varkappa(\mathbf{r})$ depend on the local position. Looking for the proper form of zilch density in the real space, we have settled on the following choice:

$$\rho_{\chi} = \frac{1}{2} \begin{cases} B \cdot \partial_t D - D \cdot \partial_t B, & \text{BDF} \\ \varepsilon\varepsilon_0 B^* \cdot \partial_t E - \mu\mu_0 D^* \cdot \partial_t H, & \text{CME,} \end{cases} \quad (52)$$

which corresponds to $C_{\chi}^{(\text{iso})}$ given by equation (49) (equation (50)) for a BDF (CME) medium.

In the nonuniform space, the conservation law for ρ_{χ} is violated by the source term on the right-hand side of the continuity equation

$$\partial_t \rho_{\chi} + \nabla \cdot J_{\chi} = F(t, \mathbf{r}). \quad (53)$$

However, both expressions for zilch density in equation (52) are related to the same zilch flow

$$J_{\chi} = \frac{\varepsilon_0\varepsilon}{2} E^* \times \partial_t E + \frac{\mu_0\mu}{2} H^* \times \partial_t H, \quad (54)$$

and the source term

$$F(t, \mathbf{r}) = \frac{\varepsilon_0}{2} \nabla \varepsilon \cdot E^* \times \partial_t E + \frac{\mu_0}{2} \nabla \mu \cdot H^* \times \partial_t H. \quad (55)$$

The source term contains only gradients of ε and μ . In this regard, we mention [58] where it has been demonstrated that in isotropic time-independent media the mixing of helicity occurs only in the presence of the space-dependent ‘resistance’ proportional to $\sqrt{\mu(\mathbf{r})/\varepsilon(\mathbf{r})}$. Similar to [58], the absence of the gradients of gyrotropic constants in $F(t, \mathbf{r})$ justifies continuity of J_{χ} between two chiral media if $\sqrt{\mu/\varepsilon}$ remains the same across the boundary⁵.

We emphasize that the absence of $\nabla \alpha$ or $\nabla \varkappa$ in the source term takes place only for the form of ρ_{χ} in equation (52).

⁵ If the relation $\mu(\mathbf{r})/\varepsilon(\mathbf{r}) = \text{Const}$ holds in medium with constituent relations in equations (40)–(43), this medium is self-dual according to [41]. In this case, we can also rewrite equation (53) in the form of conservation law for redefined ρ_{χ} and J_{χ} .

Table 1. Parameters and point groups in crystals with gyrotropic birefringence.

Tensors	Point groups
$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_{\perp} & -ia & 0 \\ ia & \varepsilon_{\perp} & 0 \\ 0 & 0 & \varepsilon_{\parallel} \end{pmatrix}, \quad \hat{\mu} = \begin{pmatrix} \mu_{\perp} & -ib & 0 \\ ib & \mu_{\perp} & 0 \\ 0 & 0 & \mu_{\parallel} \end{pmatrix}$	Tetragonal: $C_4(4)$, $C_{4h}(4/m)$, $S_4(\bar{4})$ Trigonal: $C_3(3)$, $S_6(6)$ Hexagonal: $C_6(6)$, $C_{3h}(\bar{6})$, $C_{6h}(6/m)$
$\hat{\varepsilon}\hat{\mu} = \hat{\mu}\hat{\varepsilon}, \quad \mathbf{a}\parallel\mathbf{b}\parallel\hat{z}$	

Table 2. Gyration tensor in different point groups.

Gyration tensor $\hat{\alpha}$	Point groups	Gyration tensor $\hat{\alpha}$	Point groups
$\begin{pmatrix} \alpha_0 & \alpha_2 & 0 \\ \alpha_2 & -\alpha_0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	Tetragonal: $S_4(\bar{4})$	$\begin{pmatrix} 0 & \alpha_2 & 0 \\ \alpha_2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	Tetragonal: $D_{2d}(\bar{4}2m)$
$\begin{pmatrix} \alpha_0 & \alpha_2 & 0 \\ -\alpha_2 & \alpha_0 & 0 \\ 0 & 0 & \alpha_1 \end{pmatrix}$	Tetragonal: $C_4(4)$ Trigonal: $C_3(3)$ Hexagonal: $C_6(6)$	$\begin{pmatrix} 0 & \alpha_2 & 0 \\ -\alpha_2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	Tetragonal: $C_{4v}(4mm)$ Trigonal: $C_{3v}(3m)$ Hexagonal: $C_{6v}(6mm)$
$\begin{pmatrix} \alpha_0 & 0 & 0 \\ 0 & \alpha_0 & 0 \\ 0 & 0 & \alpha_1 \end{pmatrix}$	Tetragonal: $D_4(422)$ Trigonal: $D_3(32)$ Hexagonal: $D_6(622)$	$\begin{pmatrix} \alpha_0 & 0 & 0 \\ 0 & \alpha_0 & 0 \\ 0 & 0 & \alpha_0 \end{pmatrix}$	Cubic: $T(23)$, $O(432)$

4.2. Optical chirality in crystals

In crystals, nonequivalent directions have different symmetries that are encoded in the structure of material tensors. Therefore, our general formalism should be applied with respect to certain crystalline directions. We note that, in this section, under the duality and helicity symmetries, we mean the symmetry transformations with respect to the principal axis. This resembles the situation with forward and backward scattering symmetry theorems, where the explicit form of the Mueller matrix for light scattering shows features that are related to the crystal symmetry of the dielectric scatter [59].

Fundamentally, anisotropic gyrotropic media are split into two different groups, namely, crystals with gyrotropic birefringence and media with natural optical activity, which show different behavior with respect to mirror reflections [49].

Let us first briefly focus on crystals with gyrotropic birefringence. These materials are characterized by the following constituent relations [53, 54]:

$$\mathbf{D} = (\hat{\varepsilon}_s + \mathbf{ia} \times) \mathbf{E} = \hat{\varepsilon} \mathbf{E}, \quad (56)$$

$$\mathbf{B} = (\hat{\mu}_s + \mathbf{ib} \times) \mathbf{H} = \hat{\mu} \mathbf{H}, \quad (57)$$

with \mathbf{a} and \mathbf{b} being the gyrotropic vectors, and $\hat{\varepsilon}_s$ ($\hat{\mu}_s$) stands for the diagonal part of $\hat{\varepsilon}$ ($\hat{\mu}$). We consider the case when \mathbf{a} and \mathbf{b} are parallel to the high-symmetry direction taken as the z -axis. The explicit form of $\hat{\varepsilon}$ and $\hat{\mu}$ for some point groups is given in table 1. As is well known, this kind of gyrotropy is prohibited in cubic crystals [49].

The constituent relations in equations (56) and (57) preserve the duality symmetry [41]. Apparently, along the z -axis the helicity operator \hat{S}_z is also a symmetry transformation, since it commutes with the constituent relations. Taking into account broken inversion symmetry, we conclude that similar to the isotropic chiral media, the set of nongeometric symmetries in birefringent crystals for $\mathbf{p}\parallel\hat{z}$ is four-dimensional. It contains identity, $i\partial_t$, \hat{S}_z , and duality $\mathcal{Q}_{\text{dual}}$. The latter guarantees conservation of chirality in the form of equation (39).

Maxwell's equations in crystals with natural optical activity are given by equations (26)–(28) with the following permittivity and permeability tensors:

$$\hat{\varepsilon}(\mathbf{p}) = \hat{\varepsilon}_s(\hat{I} + \hat{\alpha}(\hat{S} \cdot \mathbf{p})), \quad (58)$$

$$\hat{\mu}(\mathbf{p}) = \hat{\mu}_s(\hat{I} + \hat{\alpha}^T(\hat{S} \cdot \mathbf{p})). \quad (59)$$

Henceforth, we point \mathbf{p} along the symmetry axis, $\mathbf{p} = p\hat{z}$. Different forms of the gyration tensor for point groups in cubic, tetragonal, and hexagonal crystal families are listed in table 2. In these crystal families, $\hat{\varepsilon}_s$ and $\hat{\mu}_s$ are given by the diagonal parts of $\hat{\varepsilon}$ and μ in table 1. Apparently, the cubic case is identical to isotropic media.

All point groups in table 2 are noncentrosymmetric and break down into chiral and achiral parts. The former are represented by the point groups of 11 enantiomorphic pairs of chiral space groups, namely, T , O , C_n , and D_n ($n \geq 3$) [60], while the latter are given by S_4 , D_{2d} and C_{nv} . For a review of natural optical activity in achiral materials see, for instance, [45, 49].

Let us consider symmetry transformations in crystals with natural optical activity. According to the conditions for dual systems (see equation (11) in [41]), we find that the constituent relations in equations (58)

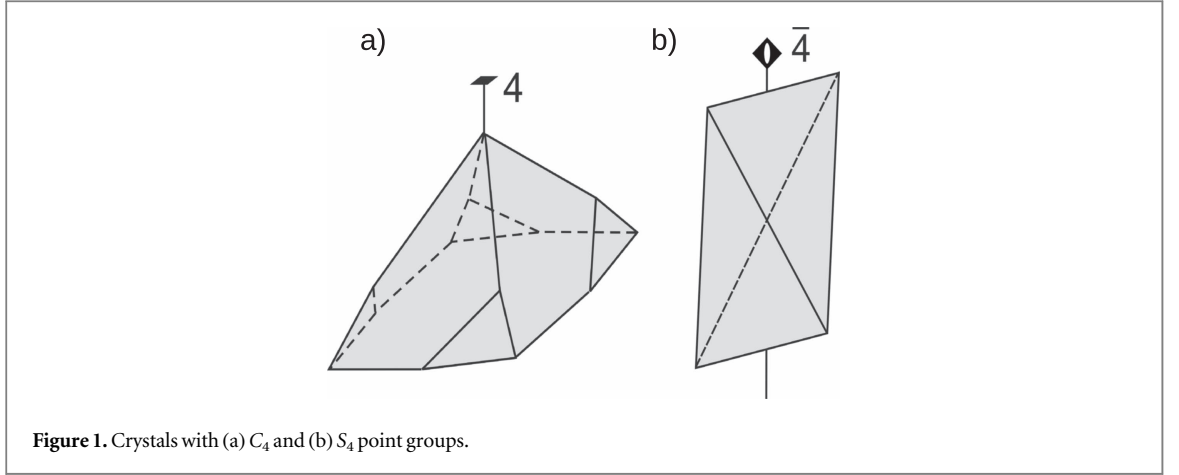


Figure 1. Crystals with (a) C_4 and (b) S_4 point groups.

and (59) preserve the duality symmetry only if $\hat{\alpha} = \hat{\alpha}^T$. This means that the duality symmetry is broken in C_n and C_{nv} ($n \geq 3$) point groups (see table 2).

At the same time, $\hat{\alpha}$ commutes with the helicity transformation \hat{S}_z in C_n and C_{nv} ($n \geq 3$), while in D_{2d} and S_4 commutativity between $\hat{\alpha}$ and \hat{S}_z does not hold. The conservation of helicity in C_n and C_{nv} is supported by the scattering theorem, which states that electromagnetic forward scattering in linear systems with the discrete rotational symmetry ($n \geq 3$) can be only helicity preserving when light spreads along the principal axis [42].

The examples above demonstrate that in crystals with natural optical activity it is possible that either the duality or helicity operator is the symmetry transformation but not both of them at the same time. We consider these two cases in more detail in the following sections.

4.2.1. Conservation of helicity with broken duality

Let us consider nongeometric symmetry transformations in C_n point groups ($n \geq 3$). The gyration tensor $\hat{\alpha}$ is given in table 2. For illustration, a chiral crystal belonging to the C_4 point group is shown in figure 1(a).

The diagonal form of $\mathcal{H}^{(A)}$ in equation (28) is brought forth by the unitary transformation

$$\psi(t, \mathbf{p}) = \mathcal{U}_C \bar{\psi}(t, \mathbf{p}) \quad (60)$$

(see appendix B for the explicit form of \mathcal{U}_C) that brings about

$$\bar{\mathcal{H}}^A = \text{diag}(p_+, -p_+, 0, -p_+, p_+, 0), \quad (61)$$

with $p_{\pm} = p[(1 \mp \alpha_0 p)^2 + \alpha_2^2 p^2]^{-1/2}$.

The form of equation (61) suggests that in C_n point groups the invariance algebra for the light spread along the high-symmetry direction is four-dimensional A_4 . In the transformed frame, the basis elements are given in equations (31)–(34). However, in achiral point groups C_{nv} , we have additional constraint $\alpha_0 = 0$ (see table 2), which restores the symmetry between left and right polarized states, $p_+ = p_-$, and the resulting invariance algebra becomes eight-dimensional, which is in agreement with equation (A6).

In order to define optical chirality, we construct the following operator in the transformed frame:

$$\bar{\mathcal{Q}}_{\chi}(\mathbf{p}) = \frac{p}{2p_-}(\bar{\mathcal{Q}}_a + \bar{\mathcal{Q}}_d) + \frac{p}{2p_+}(\bar{\mathcal{Q}}_a - \bar{\mathcal{Q}}_d), \quad (62)$$

which in the original basis is written as

$$\mathcal{Q}_{\chi}(\mathbf{p}) = \begin{pmatrix} 0 & -i\hat{l} - i\hat{\alpha}(\hat{S} \cdot \mathbf{p}) \\ i\hat{l} + i\hat{\alpha}^T(\hat{S} \cdot \mathbf{p}) & \end{pmatrix}. \quad (63)$$

We can use this operator to define optical chirality. Using $\mathcal{Q}_A = i\mathcal{Q}_{\chi}\partial_t$ in equation (38), optical chirality in point groups C_n and C_{nv} is obtained as

$$C_{\chi}^{(c)} = \frac{1}{2} \int d^3p (D^* \cdot \partial_t B - B^* \cdot \partial_t D) + \frac{i}{2} \int d^3p (D^* \cdot \hat{\alpha} \mathbf{p} \times \partial_t B - B^* \cdot \hat{\alpha}^T \mathbf{p} \times \partial_t D). \quad (64)$$

By using the constituent relations in equations (58) and (59), this equation can be rewritten in a compact form

$$C_\chi^{(c)} = -\frac{i}{2} \int d^3p (D^* \cdot \varepsilon_s^{-1} \mathbf{p} \times D + B^* \cdot \mu_s^{-1} \mathbf{p} \times B), \quad (65)$$

which is nothing but the conservation law for the helicity operator ($\mathcal{S} \cdot \tilde{\mathbf{p}}$) (see equation (B9) in appendix B).

4.2.2. Duality symmetry without helicity transformation

The gyration tensor in point groups S_4 and D_{2d} is given in table 2. We take $\hat{\alpha}$ in the form that corresponds to the S_4 group. The case of the D_{2d} group is obtained by setting $\alpha_0 = 0$. An example of an achiral crystal with an S_4 point group is demonstrated in figure 1(b).

The unitary transformation that diagonalizes $\mathcal{H}^{(A)}$ in S_4 and D_{2d} is defined as

$$\psi(t, \mathbf{p}) = \mathcal{U}_S \bar{\psi}(t, \mathbf{p}), \quad (66)$$

where \mathcal{U}_S is specified in appendix C, which leads to the following diagonal form in equation (29)

$$\tilde{\mathcal{H}}^{(A)} = \text{diag}(-p_1, p_1, 0, p_1, -p_1, 0), \quad (67)$$

where $p_1 = p[1 + (\alpha_0^2 + \alpha_2^2)p^2]^{-1/2}$.

Equation (67) shows that unlike C_n point groups, the symmetry properties of $\mathcal{H}^{(A)}$ in S_4 and D_{2d} for \mathbf{p} along the principal axis, are similar to the case of C_{nv} point groups. The invariance algebra of the symmetry transformations in S_4 and D_{2d} is eight-dimensional since the symmetry between left and right polarized states remains unbroken, which is related to the absence of optical rotation along the symmetry direction in achiral crystals [45].

Since in S_4 and D_{2d} we have $\hat{\alpha} = \hat{\alpha}^T$ for $\mathbf{p} \parallel \hat{z}$, the constituent relations preserve the duality symmetry, which means that $\mathcal{Q}_{\text{dual}}$ enters the invariance algebra. This means that optical chirality in S_4 and D_{2d} groups is given by equation (39).

We emphasize that the helicity operator ($\mathcal{S} \cdot \tilde{\mathbf{p}}$) does not belong to the symmetry transformations in S_4 and D_{2d} point groups even for $\mathbf{p} \parallel \hat{z}$. Instead, the role of the helicity operator is played by $\mathcal{Q}_h = -p^{-1} \mathcal{Q}_{\text{dual}} \partial_t$, whose explicit form is given by

$$\mathcal{Q}_h = \begin{pmatrix} (\mathcal{S} \cdot \tilde{\mathbf{p}}) \hat{\varepsilon}^{-1}(\mathbf{p}) & 0 \\ 0 & (\mathcal{S} \cdot \tilde{\mathbf{p}}) \hat{\mu}^{-1}(\mathbf{p}) \end{pmatrix}, \quad (68)$$

where we used the identity $i\partial_t \equiv \mathcal{H}^{(A)}$ together with equation (28).

4.3. Lack of duality symmetry and helicity

In systems with low crystalline symmetry, the definition of optical chirality meets with difficulties. For illustration, we consider a simple example of the nongyrotropic system that belongs to the orthorhombic crystal class. In this case, the electric permittivity and magnetic permeability tensors in principal axes are given by diagonal matrices $\hat{\varepsilon} = \text{diag}(\varepsilon_1, \varepsilon_2, \varepsilon_3)$ and $\hat{\mu} = \text{diag}(\mu_1, \mu_2, \mu_3)$.

For the diagonal $\hat{\varepsilon}$ and $\hat{\mu}$, the symmetry analysis developed in section 3 remains valid along the principal axes. We consider one particular direction taken as the \hat{z} -axis, and fix $\mathbf{p} = p\hat{z}$. To implement the general formalism in equations (26)–(28), we use the nonunitary transformation $\psi = \mathcal{N}_2 \bar{\psi}$ (\mathcal{N}_2 is specified in appendix D), which leads to the diagonal form,

$$\tilde{\mathcal{H}}^{(A)} = \left(\frac{-p}{\sqrt{\varepsilon_1 \mu_2}}, \frac{p}{\sqrt{\varepsilon_2 \mu_1}}, 0, \frac{p}{\sqrt{\varepsilon_1 \mu_2}}, \frac{-p}{\sqrt{\varepsilon_2 \mu_1}}, 0 \right), \quad (69)$$

prescribed by equation (29). In accordance with equation (A6), the symmetry transformations in the transformed frame are given in equations (31)–(34).

However, neither duality nor helicity operators can be expressed in this basis. To illustrate this fact, we apply the inverse transformation to the initial basis, $\bar{\psi} = \mathcal{N}_2^{-1} \psi$, to the operators in equations (31)–(34). Straightforward calculation yields the following basis operators in the initial frame:

$$\mathcal{Q}'_3 = \rho \mathcal{N} \bar{\mathcal{Q}}_a \mathcal{N}_2^{-1}, \quad \mathcal{Q}'_6 = \rho \mathcal{N} \bar{\mathcal{Q}}_b \mathcal{N}_2^{-1}, \quad (70)$$

$$\mathcal{Q}'_7 = \rho \mathcal{N} \bar{\mathcal{Q}}_c \mathcal{N}_2^{-1}, \quad \mathcal{Q}'_8 = \rho \mathcal{N} \bar{\mathcal{Q}}_d \mathcal{N}_2^{-1}, \quad (71)$$

where the explicit form of these matrices is given in appendix D. Now if we take the limiting case of the isotropic system, $\varepsilon_i = \mu_i = 1$ ($i = 1, 2$), we will find that equations (70) and (71) are mapped to the commutative

⁶ Here, under the helicity we mean the projection of the spin onto the direction of propagation, ($\mathcal{S} \cdot \tilde{\mathbf{p}}$). We note that to discuss the physical helicity expressed through the difference in population of left and right polarized photons, one has to construct a photon wave function in chiral crystals with S_4 and D_{2d} groups, which is, however, beyond the scope of our symmetry analysis.

subalgebra $\{Q_3, Q_6, Q_7, Q_8\}$ of A_8 (see equations (12)–(15)), which contains neither Q_2 (duality) nor Q_5 (helicity) operators.

From equations (70) and (71), we find that the only possible T -even and P -odd combinations are $p_z Q'_6$ and $p_z Q'_7$. However, substitution of these expressions into equation (38) gives zero. Therefore, we make a conclusion that it is not possible to construct optical chirality in the system, where both duality and helicity are absent. This result is supported by the conclusions in [42] that helicity-conserving scattering theorems do not exist for systems with one- and two-fold principal rotational axes.

5. Summary

We examined the symmetry properties of Maxwell's equations in various types of gyrotropic media with a particular focus on the conservation of optical chirality. For this purpose, we extended the formalism of nongeometric symmetries in vacuum to a medium with given constituent relations. Within this approach, a conclusion about the conservation of optical chirality is reduced to the analysis of the invariance algebra of the nongeometric symmetries and establishing possible isomorphism between some elements of this algebra and operators of the helicity and duality symmetries in vacuum. The advantage of this approach is that it suggests a straightforward way to derive various conservation laws related to the invariance algebra of Maxwell's equations.

Using this method, we constructed the conservation law for optical chirality in isotropic chiral media, as well as in trigonal, tetragonal, and hexagonal crystals along the symmetry direction. In particular, we demonstrated that in the gyrotropic crystals with natural optical activity, which belong to the point groups C_n or C_m , only the optical helicity remains along the principal axis; whereas in the case of achiral optically active crystals of the point symmetry S_4 or D_{2d} , only the duality transformation remains. In all presented examples, except the achiral materials, we deal with a reduction of the original eight-dimensional invariance algebra in the vacuum to the four-dimensional basis set. Additionally, we give an example of a medium where none of these symmetries is conserved.

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Appendix A. Derivation of the invariance algebra in medium

We highlight the derivation of the invariance algebra for Maxwell's equations in (23) and (24). Any symmetry transformation $Q_A(\mathbf{p})$ that transforms a solution $\psi(t, \mathbf{p}) = (D(t, \mathbf{p}), B(t, \mathbf{p}))$ of equations (23) and (24) into another solution $\psi' = Q_A \psi$ should satisfy the following invariance conditions:

$$[\mathcal{L}^{(A)}, Q_A] = g_{11} \mathcal{L}^{(A)} + g_{12} \mathcal{L}, \quad (\text{A1})$$

$$[\mathcal{L}, Q_A] = g_{21} \mathcal{L}^{(A)} + g_{22} \mathcal{L}, \quad (\text{A2})$$

where \mathcal{L} and $\mathcal{L}^{(A)}$ are determined in equations (11) and (26), and g_{ij} denotes some arbitrary operators acting on ψ .

For the diagonal operators in equations (29) and (30), we can find the invariance algebra. Given the fact that $Q_A(\mathbf{p})$ depends only on \mathbf{p} , the invariance conditions in the transformed frame take the following reduced form:

$$[\bar{\mathcal{H}}^{(A)}, \bar{Q}_A] = \bar{g}_{12} \bar{\mathcal{L}}, \quad (\text{A3})$$

$$[\bar{\mathcal{L}}, \bar{Q}_A] = \bar{g}_{22} \bar{\mathcal{L}}, \quad (\text{A4})$$

where $\bar{Q}_A = \mathcal{N}^{-1} Q_A \mathcal{N}$, and \bar{g}_{12} and \bar{g}_{22} denote some redefined operators.

The most general form of \bar{Q}_A imposed by equation (A4) is rendered as

$$\bar{Q}_A = \begin{pmatrix} q_{11} & q_{12} & 0 & q_{14} & q_{15} & 0 \\ q_{21} & q_{22} & 0 & q_{24} & q_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ q_{41} & q_{42} & 0 & q_{44} & q_{45} & 0 \\ q_{51} & q_{52} & 0 & q_{54} & q_{55} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} + \mathcal{F}\bar{\mathcal{L}}, \quad (\text{A5})$$

with some operator \mathcal{F} . The last term in this equation can be safely dropped since it does not contribute to finding the invariance algebra [8]. Then, the first invariance condition given by equation (A3) is satisfied for commutative $\bar{\mathcal{H}}^{(A)}$ and \bar{Q}_A .

To identify the invariance algebra, we explicitly calculate the commutator in equation (A3):

$$[\bar{\mathcal{H}}^{(A)}, \bar{Q}_A] = \begin{pmatrix} 0 & q_{12}(\omega_2 - \omega_1) & 0 & q_{14}(\omega_4 - \omega_1) & q_{15}(\omega_5 - \omega_1) & 0 \\ q_{21}(\omega_1 - \omega_2) & 0 & 0 & q_{24}(\omega_4 - \omega_2) & q_{25}(\omega_5 - \omega_2) & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ q_{41}(\omega_1 - \omega_4) & q_{42}(\omega_2 - \omega_4) & 0 & 0 & q_{45}(\omega_5 - \omega_4) & 0 \\ q_{51}(\omega_1 - \omega_5) & q_{52}(\omega_2 - \omega_5) & 0 & q_{54}(\omega_4 - \omega_5) & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}, \quad (\text{A6})$$

where equation (29) is used. The number of basis elements in the invariance algebra depends on the symmetry relations between ω_i that occur in equation (29). When there are no degeneracies between ω_i (the lowest symmetry case), all the matrices \bar{Q}_A , which commute with $\bar{\mathcal{H}}^{(A)}$, have diagonal form with four free parameters

$$\bar{Q}_A = \text{diag}(q_{11}, q_{22}, 0, q_{44}, q_{55}, 0), \quad (\text{A7})$$

which corresponds to the set of basis operators in equations (32)–(34).

Appendix B. Transformations in point groups C_n , C_{nv} and D_n , $n \geq 3$

In the point groups C_n , C_{nv} , and D_n , the inverse tensors $\hat{\varepsilon}^{-1}$ and $\hat{\mu}^{-1}$ in equation (28) are given by

$$\hat{\varepsilon}^{-1} = \frac{1}{\varepsilon_{\perp} d} \begin{pmatrix} 1 + i\alpha_2 p & i\alpha_0 p & 0 \\ -i\alpha_0 p & 1 + i\alpha_2 p & 0 \\ 0 & 0 & 0 \end{pmatrix} + \frac{1}{\varepsilon_{\parallel}} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (\text{B1})$$

$$\hat{\mu}^{-1} = \frac{1}{\mu_{\perp} d^*} \begin{pmatrix} 1 - i\alpha_2 p & i\alpha_0 p & 0 \\ -i\alpha_0 p & 1 - i\alpha_2 p & 0 \\ 0 & 0 & 0 \end{pmatrix} + \frac{1}{\mu_{\parallel}} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (\text{B2})$$

where $d = 1 - |a|^2 + 2i\alpha_2 p$ and $a = (\alpha_0 + i\alpha_2)p$. In what follows, we will use the units where $\varepsilon_{\perp} = \mu_{\perp} = 1$.

The matrix $\mathcal{H}^{(A)}$ in equation (28) is diagonalized in two steps. First, we make a transformation to the helicity basis

$$\mathcal{H}_{\Lambda}^{(A)} = \mathcal{U}_{\Lambda}^{\dagger} \mathcal{H}^{(A)} \mathcal{U}_{\Lambda}, \quad (\text{B3})$$

where $\mathcal{U}_{\Lambda} = \sigma_0 \otimes \hat{U}_{\Lambda}$ (see equation (35)), which gives

$$\mathcal{H}_{\Lambda}^{(A)} = \begin{pmatrix} 0 & i\hat{M}^* \\ -i\hat{M} & 0 \end{pmatrix}, \quad (\text{B4})$$

where $\hat{M} = \text{diag}(-p/(1 + a^*), p/(1 + a), 0)$. Second, we apply a unitary transformation

$$\bar{\mathcal{H}}^{(A)} = \mathcal{U}_M^{\dagger} \mathcal{H}_{\Lambda}^{(A)} \mathcal{U}_M, \quad (\text{B5})$$

with

$$\mathcal{U}_M = \frac{1}{\sqrt{2}} \begin{pmatrix} -\frac{ip}{p_+(1-a)} & 0 & 0 & \frac{ip}{p_+(1-a)} & 0 & 0 \\ 0 & -\frac{ip}{p_-(1+a^*)} & 0 & 0 & \frac{ip}{p_-(1+a^*)} & 0 \\ 0 & 0 & \sqrt{2} & 0 & 0 & 0 \\ 1 & 0 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & \sqrt{2} \end{pmatrix}, \quad (\text{B6})$$

which gives the diagonal form equation (61). Altogether, the whole transformation in equation (60) is rendered by

$$\mathcal{U}_C = \mathcal{U}_\Lambda \mathcal{U}_M. \quad (\text{B7})$$

Let us note that the conservation law for the optical chirality in equation (65) is obtained directly. If we notice that the inverse transformation of $\bar{\mathcal{Q}}_b$ in equation (32) gives the helicity operator

$$(\mathcal{S} \cdot \tilde{\mathbf{p}}) = \mathcal{U}_C \bar{\mathcal{Q}}_b \mathcal{U}_C^\dagger, \quad (\text{B8})$$

then optical chirality can be defined similar to equation (20):

$$C_\chi^{(c)} = \frac{1}{2} \int d^3p \psi^\dagger(t, \mathbf{p}) (\mathcal{S} \cdot \mathbf{p}) \psi(t, \mathbf{p}), \quad (\text{B9})$$

which corresponds to equation (65).

Appendix C. Transformations in point groups S_4 and D_{2d}

In achiral point groups S_4 and D_{2d} , we have the following $\hat{\varepsilon}^{-1}$ and $\hat{\mu}^{-1}$ in equation (28):

$$\hat{\varepsilon}^{-1} = \frac{1}{\varepsilon_\perp(1+|a|^2)} \begin{pmatrix} 1 - i\alpha_2 p & i\alpha_0 p & 0 \\ i\alpha_0 p & 1 + i\alpha_2 p & 0 \\ 0 & 0 & 0 \end{pmatrix} + \frac{1}{\varepsilon_\parallel} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (\text{C1})$$

$$\hat{\mu}^{-1} = \frac{1}{\mu_\perp(1+|a|^2)} \begin{pmatrix} 1 - i\alpha_2 p & i\alpha_0 p & 0 \\ i\alpha_0 p & 1 + i\alpha_2 p & 0 \\ 0 & 0 & 0 \end{pmatrix} + \frac{1}{\mu_\parallel} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (\text{C2})$$

In order to diagonalize $\mathcal{H}^{(A)}$ in equation (28), we apply a sequence of unitary transformations

$$\mathcal{U}_S = \mathcal{U}_\Lambda \mathcal{U}_N \mathcal{U}_2, \quad (\text{C3})$$

where $\mathcal{U}_\Lambda = \sigma_0 \otimes \hat{U}_\Lambda$, $\mathcal{U}_N = \sigma_0 \otimes \hat{U}_N$, and $\mathcal{U}_2 = \hat{U}_2 \otimes I$ with

$$\hat{U}_\Lambda = \begin{pmatrix} -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\ \frac{i}{\sqrt{2}} & \frac{i}{\sqrt{2}} & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \hat{U}_N = \begin{pmatrix} \frac{|a|}{\sqrt{2}a^*} \frac{1 - \sqrt{1+|a|^2}}{\sqrt{1+|a|^2} - \sqrt{1+|a|^2}} & \frac{|a|}{\sqrt{2}a^*} \frac{1 + \sqrt{1+|a|^2}}{\sqrt{1+|a|^2} + \sqrt{1+|a|^2}} & 0 \\ \frac{|a|}{\sqrt{2}} \frac{1}{\sqrt{1+|a|^2} - \sqrt{1+|a|^2}} & \frac{|a|}{\sqrt{2}} \frac{1}{\sqrt{1+|a|^2} + \sqrt{1+|a|^2}} & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (\text{C4})$$

Note that \mathcal{U}_N commutes with $\bar{\mathcal{L}} = \mathcal{U}_\Lambda^\dagger \mathcal{L} \mathcal{U}_\Lambda$.

Appendix D. Orthorhombic crystal

The diagonalization in equation (29) is carried out by applying

$$\mathcal{N} = \begin{pmatrix} -\sqrt{\frac{\varepsilon_1}{\varepsilon_1 + \mu_2}} & 0 & 0 & \sqrt{\frac{\varepsilon_1}{\varepsilon_1 + \mu_2}} & 0 & 0 \\ 0 & -\sqrt{\frac{\varepsilon_2}{\varepsilon_2 + \mu_1}} & 0 & 0 & \sqrt{\frac{\varepsilon_2}{\varepsilon_2 + \mu_1}} & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & \sqrt{\frac{\mu_1}{\mu_1 + \varepsilon_2}} & 0 & 0 & \sqrt{\frac{\mu_1}{\mu_1 + \varepsilon_2}} & 0 \\ \sqrt{\frac{\mu_2}{\mu_2 + \varepsilon_1}} & 0 & 0 & \sqrt{\frac{\mu_2}{\mu_2 + \varepsilon_1}} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}, \quad (\text{D1})$$

which commutes with \mathcal{L} in equation (11).

The explicit form of Q'_A in equations (70) and (71) is given by the following expressions:

$$Q'_3 = \begin{pmatrix} 0 & 0 & 0 & 0 & -\frac{\varepsilon_1 + \mu_2}{2\sqrt{\varepsilon_1\mu_2}} & 0 \\ 0 & 0 & 0 & -\frac{\varepsilon_2 + \mu_1}{2\sqrt{\varepsilon_2\mu_1}} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\frac{\varepsilon_2 + \mu_1}{2\sqrt{\varepsilon_2\mu_1}} & 0 & 0 & 0 & 0 \\ -\frac{\varepsilon_1 + \mu_2}{2\sqrt{\varepsilon_1\mu_2}} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix},$$

$$Q'_8 = \begin{pmatrix} 0 & 0 & 0 & 0 & \frac{\varepsilon_1 + \mu_2}{2\sqrt{\varepsilon_1\mu_2}} & 0 \\ 0 & 0 & 0 & -\frac{\varepsilon_2 + \mu_1}{2\sqrt{\varepsilon_2\mu_1}} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\frac{\varepsilon_2 + \mu_1}{2\sqrt{\varepsilon_2\mu_1}} & 0 & 0 & 0 & 0 \\ \frac{\varepsilon_1 + \mu_2}{2\sqrt{\varepsilon_1\mu_2}} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}, \quad (\text{D2})$$

$$Q'_6 = \text{diag}\left(-\frac{\varepsilon_1 + \mu_2}{2\varepsilon_1}, \frac{\varepsilon_2 + \mu_1}{2\varepsilon_2}, 0, \frac{\varepsilon_2 + \mu_1}{2\mu_1}, -\frac{\varepsilon_1 + \mu_2}{2\mu_2}, 0\right),$$

$$Q'_7 = \left(\frac{\varepsilon_1 + \mu_2}{2\varepsilon_1}, \frac{\varepsilon_2 + \mu_1}{2\varepsilon_2}, 0, \frac{\varepsilon_2 + \mu_1}{2\mu_1}, \frac{\varepsilon_1 + \mu_2}{2\mu_2}, 0\right). \quad (\text{D3})$$

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