Rotation and electric-field responses and absolute enantioselection in chiral crystals

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Microscopic origin of chirality and possible electric-field induced rotation and rotation-field induced electric polarization are investigated. By building up a realistic tight-binding model for elemental Te crystal in terms of symmetry-adopted basis, we identify the microscopic origin of the chirality and essential couplings among polar and axial vectors with the same time-reversal properties. Based on this microscopic model, we elucidate quantitatively that the inter-band process, driven by the nearest-neighbor spin-dependent imaginary hopping, is the key factor in the electric-field induced rotation and its inverse response. From the symmetry point of view, these couplings are characteristic common to any chiral material, leading to a possible experimental approach to achieve absolute enantioselection by simultaneously applied electric and rotation fields, or magnetic field and electric current, and so on, as a conjugate field of the chirality.

Introduction.— Chirality is three-dimensional geometric property exhibiting ubiquitously in nature. Handedness or enantiomer in chiral materials is characterized by a quantity having time-reversal $T$-even pseudoscalar (spatial-inversion $P$ odd) property $[1, 2]$, whose sign corresponds to the left and right handednesses. This significant quantity in chiral materials, however, has not fully been understood at microscopic level. Thus, clarifying the microscopic origin of a $T$-even pseudoscalar inherent in chiral materials is essential to unveil the heart of chirality and to achieve absolute enantioselection in chiral materials.

In this Letter, we begin with a phenomenological discussion of the features of $T$-even pseudoscalar and important coupling to it based on the symmetry argument. Then, to confirm the existence of the coupling, we quantitatively investigate the expected responses by using the specific microscopic model of the elemental Te crystal. Lastly, we propose a possible experimental approach to realize absolute enantioselection in chiral materials, by the external fields that is accessible to $T$-even pseudoscalar via the elucidated coupling.

Chirality and related quantities.— From the symmetry point of view, a $T$-even pseudoscalar can be decomposed into the inner product of polar and axial vectors with the same $T$ property. Such a decomposition is clearly carried out by the concept of electronic multipole basis $[3, 4]$. Namely, a $T$-even pseudoscalar corresponds to an electric toroidal (ET) monopole $G_0$ with $(P, T) = (-, +)$, and is decomposed into the inner product of a magnetic (M) dipole $M$, $(P, T) = (+, -)$, and magnetic toroidal (MT) dipole $T$, $(P, T) = (-, -)$, as

$$G_0 \rightarrow T \cdot M.$$ (1)

Since a conjugate field of $T$ is an electric current $J$, Eq. (1) indicates that linear current-induced magnetic response (Edelstein effect) can occur in chiral crystals. This is because an invariant coupling $G_0(T \cdot M)$ exists in the free energy of chiral crystals with finite $G_0$. Indeed, the Edelstein effect $[7, 8]$ and current-induced optical activity $[9, 10]$ have been observed in elemental Te crystal $[11, 12]$. In addition, as the symmetry of $T$ is common as a wave vector $k$, the hedgehog spin texture observed around the H point of the Brillouin zone in Te $[13, 14]$ is nothing but a consequence of the $k$-space representation of the ET monopole, $k \cdot \sigma$ $[4]$.

An ET monopole $G_0$ can be decomposed in another way:

$$G_0 \rightarrow Q \cdot G,$$ (2)

where $Q$ is an electric (E) dipole, $(P, T) = (-, +)$, such as a position vector $R$, while $G$ represents an ET dipole with $(P, T) = (+, +) [4]$. Equation (2) implies that $G$-flux structure exists in chiral crystals as shown in Fig. (a). (b) Classical view of the ET dipole, which is a vortex-like alignment of the E dipoles, $Q$. (c) The essential coupling existing in chiral crystals. The conjugate fields of $G$ and $Q$ are given in the parenthesis.

FIG. 1. (a) ET monopole $G_0$ in terms of the flux structure of the ET dipoles $G$, whose direction determines the handedness of the chirality. (b) Classical view of the ET dipole, which is a vortex-like alignment of the E dipoles, $Q$. (c) The essential coupling existing in chiral crystals. The conjugate fields of $G$ and $Q$ are given in the parenthesis.

With the above consideration, we find that chiral crystals...
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substantially possess the third-order coupling in the free energy as shown in Fig (c):
\[ F^{(3)} = g_1 G_0^{(1)} G^{(2)}_z G_z Q_z + g_2 G_0^{(2)} G_z Q_z, \]
where the coupling constants satisfy \( g_z = g_{\perp} \) in cubic crystals, otherwise \( g_z \neq g_{\perp} \) (\( z \) is along the screw axis). Here, \( G_0^{(1)} \)
and \( G_0^{(2)} \) can be independent ET monopoles in general. This coupling gives rise to a conversion from a polar field such as the electric field or temperature gradient into an axial response such as a rotation of the lattice, and vice versa. In other words, an electric-field induced rotation (EIR) and its inverse response, i.e., a rotation-field induced electric polarization (RIP), could appear in chiral crystals. Note that one can apply a lattice rotation field by using transverse ultrasonic wave as it generates both the strain and rotation fields. In this context, it is proposed that the temperature gradient gives rise to the lattice rotation in Te [24].

In what follows, we elucidate the microscopic origin of \( G_0 \) and related responses by taking Te as a specific example. First, we construct the realistic tight-binding (TB) model of Te by using the results of the density-functional (DF) calculation. Since the obtained TB model is expressed in terms of the symmetry-adopted electronic multipole basis, we can easily identify the microscopic origin of \( G_0 \), and evaluate the relevant couplings to it quantitatively. We then propose a possible experimental approach to achieve absolute enantioselectivity, keeping the coupling, Eq. (3), in mind.

**Tight-binding model for Te.**— Let us first consider the specific TB model for Te. As shown in Fig (a), the bulk Te crystal consists of the threefold-symmetric helical chains, which contain A, B, and C sublattices in a unit cell as shown in Fig. (b). The space group of the right- and left-handed Te are P3\(_1\)21 (#152, \( D_3^1 \)) and P3\(_2\)21 (#154, \( D_3^2 \)), respectively. Hereafter, we focus on the right-handed Te. The lattice constants are \( a = 4.458 \text{ Å}, c = 5.925 \text{ Å} \), and relaxed value is \( u = r/a = 0.274 \) for the dimensionless helix parameter [25], where \( r \) denotes the radius of the helix.

Since the electronic states near the band edges (Fermi level) in Te mainly consist of three \( p \) orbitals, \( p_x, p_y, \) and \( p_z \) [26], we consider the spinful TB Hamiltonian in 18x18 matrix. The TB Hamiltonian is constructed by using the symmetry-adopted multipole basis \( Z_\alpha \) as \( H = \sum_\alpha z_\alpha Z_\alpha \), where \( Z_\alpha \) are the independent multipole basis, which satisfy the normalization \( \text{Tr}(Z_\alpha Z_\alpha) = 1 \), and belonging to the identity irreducible representation of \( D_3^1 \). Each \( Z_\alpha \) is expressed by the direct product of the cluster or bond basis [27, 28] and atomic basis as shown later [29, 30], which are generated automatically by the symbolic computation with symmetry operations [31]. Then, the coefficients \( z_\alpha (\Delta^Q_i, A^Q_i, t^Q_i, \alpha^Q_i, \ldots \) etc.) are determined so as to reproduce the band dispersions obtained by the DF calculation with the help of the machine-learning technique [31, 32]. In this model construction, terms containing \( G_0 \) are particularly important.

The constructed TB Hamiltonian is expressed as \( H_0 = H_{\text{CEF}} + H_{\text{SOC}} + \sum_{i=1}^8 \lambda_{\alpha}^{(i)} \), where \( H_{\text{CEF}} \) and \( H_{\text{SOC}} \) are the crystalline electric field (CEF) and spin-orbit coupling (SOC) within the unit cell, and \( \lambda_{\alpha}^{(i)} \) is the \( i \)th neighbor hopping term. By taking up to the 8th neighbors, we achieved the accuracy less than \( 10^{-4} \) of the mean squared error between the normalized energy eigenvalues of our TB model and DF result [31]. The comparison of the energy dispersions is shown in Fig. (a). We have also confirmed that the orbital and spin characters of the obtained electronic states roughly reproduce those obtained by DF calculation. There are 255 independent parameters \( z_\alpha \) in total, and 30 parameters are within the nearest-neighbor (NN) hopping; four CEF parameters, five SOC parameters, and twenty one NN intra-chain hopping parameters [31].

Among these multipole basis, the most dominant contributions containing the ET monopole are \( G_0^{(ca)} \) in \( H_{\text{SOC}} \) and \( G_0^{(ba)}, G_0^{(ba)} \) in \( H^{(i)}_\text{CEF} \), which are given by

\[ G_0^{(ca)} = \frac{1}{\sqrt{2}} \left( Q^c_x \otimes G^c_x + Q^c_y \otimes G^c_y \right), \]
\[ G_0^{(ba)} = T^{(b)}_z \otimes \sigma^a_z, \]
\[ G_0^{(ba)} = \frac{1}{\sqrt{2}} \left( T^{(b)}_z \otimes \sigma^a_z + T^{(b)}_y \otimes \sigma^a_y \right), \]
where the superscripts (c,b,a) represent cluster, bond, and atomic basis, respectively. The weight \( z_\alpha \) of \( G_0^{(ca)} \) is \( \lambda^{Q2} = 1.718 \text{ eV} \) which is the most dominant contribution among \( H_{\text{CEF}} + H_{\text{SOC}} \), and that of \( G_0^{(ba)} \) is \( \alpha^{Q2} = 1.749 \text{ eV} \) which is the most dominant contribution among the ET monopoles in \( H^{(i)}_\text{CEF} \), while that of \( G_0^{(ba)} \) is \( \alpha^{Q2} = 0.5854 \text{ eV} \). Note that \( G_0^{(ca)} \) does not appear because the \( z \) component of the E dipole \( Q^{(c)} \) identically vanishes in the present Hilbert space [31]. We have confirmed that the magnitude of the parameters decreases for further neighbor hoppings [31]. Here, \( G^{(c)} = I \times \sigma \) is the atomic ET dipole (\( I \) and \( \sigma \) are the dimensionless orbital and half of spin angular momenta, respectively). \( Q^c_\mu (\mu = x, y) \) and \( T^{(b)}_\mu (\mu = x, y, z) \) are the \( \mu \) component of the cluster E dipole and bond MT dipole, which are defined in the ABC.
nomenclaturally, a conversion between polar and axial degrees of freedom is expected to occur in chiral materials via Eq. (3). To demonstrate it microscopically, we investigate the electric-field induced ET dipole response based on the present TB model. Before showing the results, we define explicitly the local E- and ET-dipole operators that describe the input and output of the response. The E dipole \(Q^{(ca)}_z\) is given by the similar expression of Eq. (4) with a minus sign for the second term, which is schematically shown in Fig. 3(c), i.e., the vortex-like alignment of \(G^{(a)}_z\) and \(G^{(a)}_y\) (cf. the roles of \(G\) and \(Q\) are reverted in Fig. 3(b)). The output rotation is also described by \(G^{(ca)}_z = Q^{(ca)}_0 \otimes G^{(a)}_z\).

Using the Kubo formula, the response function in \(G^{(ca)}_z = d_{zz} E_z\) is expressed as \((c = \text{the lattice constant})\)

\[
d_{zz}^{(c)} = d_{zz}^{(P)}[Q] + d_{zz}^{(VV)}[Q] + d_{zz}^{(VV)}[v],
\]

\[
d_{zz}^{(P)}[Q] = -\frac{e e}{N} \sum_{k \epsilon} \frac{\partial}{\partial \epsilon_{nk}} G_{nm}^{(a)} Q_{mk}^{(a)},
\]

\[
d_{zz}^{(VV)}[Q] = \frac{e e}{N} \sum_{k \epsilon} \epsilon_{nk} f_{nk} - \epsilon_{mk} f_{mk} G_{nm}^{(a)} \psi_{zk},
\]

\[
d_{zz}^{(VV)}[v] = \frac{e \hbar}{N} \sum_{k \epsilon} f_{nk} - f_{mk} \epsilon_{nk} - \epsilon_{mk} G_{nm}^{(a)} \psi_{zk}.
\]

Here, the matrix element of an operator \(\hat{O}\) is \(\langle \psi_{nk} | \hat{O} | \psi_{mk} \rangle\), \(f_{nk} = f(\epsilon_{nk})\) is the Fermi distribution function, \(e(>0)\) is the elementary charge, and \(N\) is the number of lattice sites. The responses \(d_{zz}^{(P)}\) and \(d_{zz}^{(VV)}\) represent the intra-band Pauli contribution proportional to the density of states (DOS), and inter-band van Vleck contributions, respectively. \([Q]\) and \([v]\) indicate the contributions arising from the local E dipole and itinerant hopping process via the velocity operator, \(v_k = \partial H_0 / \partial \epsilon_k\), respectively. Note that \(d^{(P)}_{zz}[v]\) vanishes identically by the symmetry. We have used \(N = 64^3\) and the temperature \(T = 0.01\) eV in the following results.

Figure 4(b) shows the chemical potential \(\mu\) dependence of the responses. The inter-band contribution from the itinerant hopping process, \(d_{zz}^{(VV)}[v]\), is always dominant irrespective of \(\mu\), and the EIR occurs even in the insulator. Analyzing the essential parameters to exhibit the finite response \(d_{zz}^{(P)}\), it turns out that the lowest-order of the response is proportional to the highest-weight term \(G^{(ba)}_y\) of \(H_0\) with the coefficient \(\alpha_2^{(a)}\), which is consistent with the fact that \(d_{zz}^{(VV)}[v]\) is dominant in numerical result. Thus, the itinerant ET monopole \(G^{(ba)}_y\) is the key component of the EIR response in Te. Note that the inverse RIP process is also expected to occur in both metals and insulators, as their response functions are common with Eqs. (11) and (12).

Although we have concentrated on the electronic responses in the above, the actual lattice rotation should occur via the electron-lattice coupling. When we restrict our discussion to the pure lattice rotation with the angle \(\omega_z\) with respect to \(z\) screw axis, the electron-lattice coupling can be evaluated by rotating inversely the electronic system by the angle \(-\omega_z\) [33].

Electric-field induced rotation.— As already discussed, the sublattice space as

\[
Q^{(c)}_x = \frac{1}{\sqrt{6}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 2 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad Q^{(c)}_y = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix},
\]

(7)

\[
Q^{(c)}_0 = \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad T^{(b)}_x = \frac{1}{\sqrt{6}} \begin{pmatrix} 0 & -i & i \\ i & 0 & -i \\ -i & 0 & 0 \end{pmatrix},
\]

(8)

\[
T^{(b)}_y = \frac{1}{2\sqrt{3}} \begin{pmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 2i \end{pmatrix}, \quad T^{(b)}_z = \frac{1}{2\sqrt{3}} \begin{pmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 2i \end{pmatrix}.
\]

(9)

Although there are also the spinless version of \(G^{(ba)}_y\) and \(G^{(ba)}_0\) which are given by replacing \(\sigma\) with \(I\) in Eq. 5, the weight of these multipole basis is much smaller than that for the spinful ones. As shown in Fig. 3(a), \(G^{(ca)}_0\) is the local ET monopole having the \(G^{(a)}\)-flux structure in the unit cell as Eq. 4. On the other hand, \(G^{(ba)}_y\) in Fig. 3(b) is the itinerant ET monopole which is a kind of SOC with the spin-dependent imaginary hopping. From Figs. 3(a), (b), the relation between the sign of \(G_0\) and the handedness is apparent. For example, in Fig. 3(b), \(T_x^{(b)}\) corresponds to the imaginary hopping directed to \(+z\) direction. If we consider the left-handed system, \(T_x^{(b)}\) directs to \(-z\) direction, and the sign of \(G_0\) is inverted.

These ET monopoles, \(G^{(ca)}_0\) and \(G^{(ba)}_0\), play roles of \(G^{(1)}\) and \(G^{(2)}\) in Eq. 4, respectively. As discussed below, both of them give dominant contributions to the EIR and RIP. In addition, the itinerant ET monopole \(G^{(ba)}_y\) is also the main origin of the Edelstein effect observed in Fe [11-12]. Moreover, the Fourier transform of \(G^{(ba)}_0\) together with \(G^{(ba)}_y\) give rise to the hedgehog spin texture around the H point in the momentum space [13-14, 31].
Absolute enantioselection by rotation and electric fields.—

Finally, we propose a possible experimental approach to achieve absolute enantioselection in chiral materials. As was shown phenomenologically and microscopically, there is a proper coupling among $G_0$, $G$, and $Q$ in chiral materials. The conjugate field of $Q$ is an electric field $E$ and that of $G$ is a rotation field $\omega$ such as a rotation of electric field or equivalently time-dependent magnetic field. Thus, the conjugate field of the chirality, that is $G_0$, is a product of polar and axilal vector fields, $\vec{E}_\mu$ and $\omega_\mu$. In the electromagnetism, it is known as the quantity called zilch, which describes optical chirality. Therefore, as shown in Fig. 4 in the case of $\mu = z$, absolute enantioselection can be achieved by simultaneous application of electric $(\vec{E}_\mu)$ and rotation $(\omega_\mu)$ fields for instance. The sign of $\omega_\mu E_\mu$ controls the preferred handedness during crystallization process, as shown in the left-most and right-most panels in Fig. 4. It should be emphasized that a time-dependent magnetic field is favorable with the constant time derivative or net accumulation with definite sign. In addition to this, both electric and magnetic fields must be parallel with each other. Moreover, as was discussed in Eq. (1), $G_0$ could also couple with $M_\mu T_\mu$. Thus, the combined static magnetic field and time-dependent electric field (i.e., $\nabla \times \vec{B}$, which is a conjugate field of $T_\mu$), or the combined static magnetic field and electric current, can be used to achieve absolute enantioselection as well. This generic approach is applicable to any chiral material.

Conclusion.— We have unveiled the microscopic origin of chirality and possible electric-field induced rotation and its inverse response, rotation-field induced electric polarization. First, we have demonstrated that the chirality corresponds to the electric toroidal monopole given by Eq. (2), in terms of symmetry. We have also found the essential coupling to it as shown in Eq. (3), which is the key element for both the electric-field induced rotation and its inverse response. Then, using the realistic tight-binding model for elemental Te crys-
tal, we have elucidated that the inter-band process, driven by
the itinerant electric toroidal monopole shown in Fig. 3(b), is
the crucial factor in these response functions. These responses
occur even in the insulators, which is in marked contrast to
the Edelstein effect observed in Te. Lastly, we have proposed
a generic experimental approach to realize the absolute enan-
tioselection in chiral materials by the conjugate field of the
chirality, such as simultaneously applied electric and rotation
fields, or magnetic field and electric current, and so on.

The authors would like to thank Yuki Yanagi for provid-
ing us the data of the DF calculation. We also thank Michito
Suzuki, Hiroaki Ikeda, Yoshihiko Togawa, Junichiro Kishine,
Koichi Izawa, Hiroshi Amitsuka, Tatsuya Yanagisawa, Hi-
royuki Hidaka, and Satoru Hayami for fruitful discussions.
This work was supported by JSPS KAKENHI Grants Num-
bers JP19K03752, JPJP20J21838. A part of numerical and
symbolic calculations was performed in the supercomputing
systems in the MAterial science Supercomputing system for
Advanced MUlti-scale simulations towards NExt-generation-
Institute for Materials Research (MASAMUNE-IMR) of the
Center for Computational Materials Science, Institute for Ma-
terials Research, Tohoku University.

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adopted multipole basis and the explicit expressions of the TB
Hamiltonian in terms of them are given in detail. A brief de-
scription of the process of parameter fitting, and the obtained
values of the 30 parameters within the nearest-neighbor hop-
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