Engineering Weyl phases and nonlinear Hall effects in T_d-MoTe_2

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MoTe_2 has recently attracted much attention due to the observation of pressure-induced superconductivity, exotic topological phase transitions, and nonlinear quantum effects. However, there has been debate on the intriguing structural phase transitions among various observed phases of MoTe_2, and their connection to the underlying topological electronic properties. In this work, by means of density-functional theory (DFT+U) calculations, we describe the structural phase transition between the polar T_d and nonpolar 1T′ phases of MoTe_2 in reference to a hypothetical high-symmetry T_0 phase that exhibits higher-order topological features. In the T_d phase we obtain a total of 12 Weyl points, which can be created/annihilated, dynamically manipulated, and switched by tuning a polar phonon mode. We also report the existence of a tunable nonlinear Hall effect in T_d-MoTe_2, and propose possible ways to observe this effect in experiments. In particular, we identify a configuration in which a nonlinear surface response current is predicted. The potential technological applications of the tunable Weyl phase and the nonlinear Hall effect are discussed.

Owing to its intriguing structural and electronic phase transitions and novel technological applications, MoTe_2 remains in the active area of research [1–9]. Specifically, the experimentally tunable structural phase transition between the 1T′ and T_d-phases [10–12] allows for the exploitation of the topological electronic properties and the electronic phase transitions, yielding various novel quantum phenomena such as extremely large magnetoresistance [13–16], Berry-curvature-induced planar Hall effect [17], nonlinear anomalous Hall effect [18], quantum spin Hall effect [19], spin-photogalvanic effect [20], quantum nonlinear Hall effect [21], large spin Hall conductance with high spin Hall angles [22], complex Fermiology [23–25], and tunable polar/phase domain walls in MoTe_2 [12]. Recent theoretical and experimental work has confirmed the existence of type-I and type-II Weyl fermions in T_d-MoTe_2, and reported topological quantum oscillations and pressure-enhanced superconductivity [7, 10, 23, 26–35]. However, the total number and location of Weyl fermions in MoTe_2 are still under debate [23–26, 29, 32, 36]. Strikingly, a higher-order topological phase has been predicted in 1T′-MoTe_2 [37–39].

Despite the recent advances in understanding the topological electronic properties of MoTe_2 [10, 23, 26–29, 31–37], the link between the higher-order topological phase (1T′) and the Weyl phase (T_d) remains elusive. Kim et al. theoretically showed that the interlayer coupling is pivotal in determining distinct structural phase transitions in MoTe_2 [4]. Recent experiments further confirm the essential role of interlayer shear modes in governing the T_d-1T′ phase transition in MoTe_2 [4, 40, 41]. Notably, controlled mechanical [10], optical [41], dimensional [42, 43], and temperature [7, 44] -driven T_d-1T′ phase transitions have been experimentally demonstrated. A centrosymmetric phase (T_d′) with no Weyl nodes has also been recently observed at the T_d-1T′ phase boundary [45, 46]. We find that most of the reported theoretical and experimental studies mainly focus on the characterization of distinct phases of MoTe_2. A systematic connection among these phases, in the context of the potential energy surface profile and crystal symmetries, has not been clarified in the literature.

In this work, by means of ab-initio density-functional theory (DFT) calculations, we first describe the intriguing structural phase transition between the polar T_d and nonpolar 1T′ phases of MoTe_2 by defining a high-symmetry reference phase T_0, which has a higher-order topological nature. We then study the evolution of the Weyl fermions in the polar phase along the polarity reversal path. We find that additional Weyl points (WPs) get created/annihilated via pair creation/annihilation process as we tune the spatial-inversion symmetry breaking parameter (λ). The WPs switch their chirality as we go from one to another polar variant of the T_d phase. Since this structural phase transition has recently been experimentally realized using electron beams [12], and notably, is completely reversible, we believe that the switching and pairwise creation/annihilation of Weyl fermions can be experimentally accessed in MoTe_2. We further demonstrate that a higher-order topological phase naturally appears when all WPs annihilate each other at λ = 0. Finally, we report on the existence of a tunable nonlinear Hall effect and discuss the role of dimensionality on this effect. We notice that the surface termination along the (001) direction leads to the manifestation of a nonlinear surface response current solely arising due to the broken symmetries at the surface. Such a tunable nonlinear Hall effect could lead to electrically switchable circular photogalvanic [20, 47], bulk rectification [48], and chiral polaritonic effects [49].

MoTe_2 crystallizes in three distinct phases: (i) 2H (hexagonal, space group P6_3/mmc), (ii) 1T′ (monoclinic, space group P2_1/m), and (iii) T_d (orthorhombic, space group Pnam_2_1) [3, 4, 10, 40, 44, 50–52]. In all three phases, Mo and Te atoms form Te-Mo-Te triple layers, which stack along the c-axis and interact via weak van der Waals interactions. The Te atoms form symmetrical octahedra in the hexagonal 2H phase, whereas these octahedra...
FIG. 1. Crystal structure of MoTe$_2$ in (a) 1T’, (b) T$_0$, and (c) T$_d$ phases. Green arrows denote the interlayer displacement direction parallel (+) or antiparallel (−) to $\vec{a}$, and $\lambda$ represents the interlayer displacement parameter (see text). Hollow circles ‘.’ mark the inversion centers in the 1T’ phase along $\vec{c}$. (d) The phonon band structure of T$_0$ phase shown within [-20, 80] cm$^{-1}$. (e) The double-well potential energy profile of T$_0$ phase as a function of the inversion symmetry breaking parameter $\lambda$. (f) An schematic representation showing the link between all T$_d$ and 1T’ phases via the reference structure T$_0$.

dra are markedly distorted in the 1T’ and T$_d$ phases [12]. Fig. 1(a,c) shows the crystal structure of MoTe$_2$ in the 1T’ and T$_d$ phases. Both phases are quite similar, except for the fact that 1T’ is monoclinic ($\beta \neq 90$) while T$_d$ is orthorhombic ($\alpha = \beta = \gamma = 90$). In both phases, Mo atoms dimerize forming long-short bonds along the $\vec{a}$ lattice vector and zigzag Mo-Mo metallic bonds running along the $\vec{b}$ direction.

We notice an interesting symmetry between the Mo-Te polyhedra (highlighted using light grey rectangles in Fig. 1(c)) of alternating triple layers. These polyhedra alternatively adopt either clockwise or counterclockwise twist (as viewed along $\vec{b}$) in the alternating triple layers. In the T$_d$ phase, adjacent layers are connected by $M_x \{ T (\frac{a}{2} (1 + \lambda)) \}$ symmetry operation, where $M_x$ is a vertical mirror, $T (\frac{a}{2} / 2)$ denotes translation by $\vec{a}/2$, and $\lambda$ denotes an interlayer displacement along $\vec{a}$, as shown in Fig. 1(c). The main cause of the nonzero $\lambda$ is the presence of steric interactions between Te atoms in the adjacent triple layers, which drive an in-plane shift of the alternating layers along $\vec{a}$ so as to increase the separation between these atoms. Taking the above facts into account, we define a nonpolar high-symmetry phase T$_0$ (space group $Pnma$) having $\lambda = 0$, as shown in Fig. 1(b).

Fig. 1(d) shows an enlarged phonon spectrum of the T$_0$ phase. The full phonon spectra of T$_0$, 1T’, and T$_d$ phases together with all the theoretical details are given in the supplemental material (SM) [53]. We notice only two phonon instabilities in the T$_0$ phase: (i) an unstable optical zone center phonon mode ($\Gamma_2^-$), and (ii) a linearly-dispersing unstable phonon branch along $\Gamma$-X direction indicating an elastic instability ($\Gamma_+^4$). The first instability, $\Gamma_2^-$, breaks the inversion symmetry of the T$_0$ phase and corresponds to an in-plane optical vibration of the alternating triple layers. By modulating T$_0$ phase along $\Gamma_2^-$ mode, we obtain a double-well potential energy profile with two local minima at $\lambda = \pm 0.50 \text{Å}$, as shown in Fig. 1(e). These local minima belong to the two polar variants of the T$_d$ phase, which we refer as T$_d$-A and T$_d$-B. The interlayer displacement pattern of the alternating Mo-Te triple layers in the T$_d$-A and T$_d$-B phases is $- + - +$ and $+ + + +$, respectively, thus, ensuring the orthogonality of the T$_d$ phase.

On the other hand, the elastic instability ($\Gamma_+^4$ mode) causes a shear distortion of the unit cell, making the $\vec{a}$ and $\vec{c}$ lattice vectors nonorthogonal. By modulating the T$_0$ phase along the $\Gamma_+^4$ mode, we obtain a similar potential energy profile as shown in Fig. 1(e) with two local minima at cell angles $\beta = 93.6^\circ$ and $\beta = 87.4^\circ$ (see SM [53]). These two local minima mark the two ferroelastic twin phases, 1T’-I and 1T’-II, of the monoclinic 1T’ phase. The obtained value of $\beta$ is in excellent agreement with previous reports [4, 10, 23, 26-29, 31-34, 36, 50]. The corresponding interlayer displacement pattern of Mo-Te triple layers in the 1T’-I and 1T’-II phases is: $++ + +$ and $--- ---$, respectively. Fig. 1(f) schematically represents the connection between two polar T$_d$ phases and two ferroelastic 1T’ phases in reference to the unstable high symmetry T$_0$ phase. An electron beam induced tunable reversible structural phase transition among these phases has recently been experimentally realized [12].

Due to the broken inversion symmetry requirement, the path connecting the 1T’-I and 1T’-II phases cannot access the Weyl phase. Therefore, we focus on the T$_d$-A $\rightarrow$ T$_d$-B path, investigating the subtle changes in the electronic band structure that occur there. Fig. 2(a) shows the electronic band structure of T$_d$-MoTe$_2$ calculated with inclusion of spin-orbit coupling (SOC). The states near the Fermi energy ($E_F$) are mainly composed of Mo-$4d$ and Te-$5p$ orbitals [54]. Without SOC, the lowest conduction band and the highest valence band
at Ed ordinates are given in the SM [53]. The total 12 WPs in BZ is shown in Fig. 2(c), and their co-
linearly cross each other near the Fermi level, forming type-I and type-II nodal loops above and below $E_F$, as marked in Fig. 2(b). Inclusion of SOC destroys the nodal loops and results in discrete gapless WPs formed slightly
away from the high-symmetry directions near the Fermi level [29]. The type-I WPs (W1) are located near the
Gamma point (on the $k_z = 0$ plane) above the Fermi level at $E_F + 0.108$ eV, whereas type-II WPs (W2) are located
relatively farther from the Gamma point (off the $k_z=0$ plane) at $E_F - 0.038$ eV. A careful investigation of the electronic
band structure reveals a total of 12 WPs (4 W1 and 8 W2) in the full Brillouin zone (BZ). The distribution of
all 12 WPs in BZ is shown in Fig. 2(c), and their coordinates are given in the SM [53]. The $T_d$-$A$ and $T_d$-$B$ phases host exactly the same number of WPs at the same coordinates in momentum and energy, but with reversed chirality. This is expected since the $T_d$-$A$ and $T_d$-$B$ phases are related by inversion symmetry.

Motivated by the above results, we investigate the evolution of the WPs along the $T_d$-$A$ to $T_0$ to $T_d$-$B$ path as a function of $\lambda$. We observe that WPs get created in pairs as we move away from the $T_d$-$A$ phase (see the SM [53]). Note that the newly created pairs of WPs are of type I. The total number of WPs increases from 12 to 16 and then 20, 24, 28, and 32, as we vary $|\lambda/\lambda_0|$ from 1.0 to 0.92, 0.88, 0.79, 0.72, and 0.63, respectively ($\lambda_0 = 0.50$ Å). The maximum number of obtained WPs is 32. This finding explains why previous authors reported such different counts of the number of Weyl points [23–26, 29, 32, 36], and reveals that the total number of WPs in MoTe$_2$ is very sensitive to the lattice distortions. As we further tune $|\lambda/\lambda_0|$, the WPs move towards their opposite partners in momentum space and start pair-annihilating, leaving no remaining WPs at $|\lambda/\lambda_0| = 0$ (at $T_0$ phase).

Due to the absence of WPs and the presence of a double band inversion at the Gamma point, the $T_0$ phase turns into a second-order topological insulator, similar to the 1T$'$ phase [37, 38]. Notably, we find that both the $T_0$ and 1T$'$ phases belong to a strong topological class 20 as classified in Ref. [55] having topological invariant $z_2=2$. As we cross the $T_0$ phase and migrate towards the $T_d$-$B$ phase, the WPs systematically start reappearing, and the aforementioned process repeats but with the switched chirality of WPs (details in the SM [53]). An animation showing the evolution of WPs as a function of $|\lambda/\lambda_0|$ is provided in the SM [53]. The pairwise creation/annihilation of WPs causes abrupt changes in the Berry curvature and Fermi-surface geometry yielding a nonzero Berry curvature dipole moment (BCDM) and, as a result, a nonlinear Hall effect in the $T_d$ phase, as we discuss below.

In the study of the nonlinear quantum Hall effect [56], a transverse current is predicted to be generated by a harmonically oscillating electric field $E_c = \text{Re}\{E_c e^{i\omega t}\}$ in the absence of inversion symmetry. The response current up to second order reads $j_a = \text{Re}\{j_a^0 + j_a^2 e^{2i\omega t}\}$, where a rectified current $j_a^0 = \chi_{abc} E_b E_c$ and a second-harmonic current $j_a^{2\omega} = \chi_{abc} E_b E_c$ depend on the nonlinear conductivity tensor $\chi_{abc}$, where $a, b, c \in \{x, y, z\}$. The nonlinear conductivity tensor associated with the BCDM ($D_{bd}$) can be written as

$$
\chi_{abc}(\omega) = -\varepsilon_{abc} \frac{e^{3\tau}}{2(1+i\omega \tau)} D_{bd},
$$

where $\varepsilon_{abc}$ is the rank-three Levi-Civita symbol and $\tau$ is the relaxation time. The $D_{bd}$ is obtained by integrating Berry curvature, weighted by the Cartesian component of the group velocity on the Fermi surface according to

$$
D_{bd} = \oint_{\text{FS}} \frac{d^2k}{(2\pi)^3} \sum_n v_n^a(k) \Omega_n^a(k),
$$
where \( v^n_y(k) = \partial_{k_y} E_{n k} / |\nabla_k E_{n k}| \) is a normalized group velocity component for band \( n \), and \( \Omega^n \) is the Berry curvature pseudovector defined via \( \Omega^n_{\alpha} = \varepsilon_{\alpha bc} \Omega^n_{bc} \). The superscripts represent band indices. We compute the Berry curvature using the Kubo formula

\[
\Omega^n_{\alpha}(k) = -2\hbar^2 \sum_{m \neq n} \text{Im} \frac{\langle m k | \hat{v}_n | m k \rangle \langle m k | \hat{v}_b | n k \rangle}{(E_{n k} - E_{m k})^2 + \delta^2},
\]

where \( \hat{v}_n \) is the velocity operator and \( \delta = 0.1 \text{meV} \) is a broadening term (see [57] for numerical details).

![FIG. 3. Calculated Berry curvature (a) \( \Omega_x \) and (b) \( \Omega_y \) on the Fermi surface of MoTe2 in \( T_d-A \) phase. Yellow (Blue) color represents positive (negative) Berry curvature. (c) Calculated BCDM of MoTe2 in \( T_d-A \) phase. The non-vanishing \( D_{xy} \) and \( D_{yz} \) terms are plotted with respect to the chemical potential.](image)

In the presence of inversion symmetry, i.e., the case of 1T'-MoTe2, the BCDM completely vanishes. Instead, in the polar \( T_d \) phase, a non-vanishing BCDM is allowed [56, 58]. MoTe2 in the \( T_d \) phase exhibits simple mirror \( M_y \) and glide mirror \( M_x T(\bar{c}/2) \) symmetries, excepting constraints on the BCDM tensor. For instance, \( M_y \), a mirror plane normal to the chain direction, forces the group velocity \( v_y \) and Berry curvature \( \Omega_y \) to obey

\[
M_y : (v_x, v_y, v_z) \rightarrow (v_x, -v_y, v_z) \quad (\Omega_x, \Omega_y, \Omega_z) \rightarrow (-\Omega_x, \Omega_y, -\Omega_z).
\]

Here, \( v_i \) denotes group velocity for a particular band at a particular \( k \). Thus, under the \( M_y \) symmetry operation, all components of the BCDM tensor vanish except for the \( D_{xy} \), \( D_{zx} \), \( D_{yz} \), and \( D_{zy} \) terms. A further consideration of \( M_x T(\bar{c}/2) \) symmetry eliminates the \( D_{xz} \) and \( D_{xy} \) terms as well. Thus, only two terms, \( D_{xy} \) and \( D_{yz} \), survive in \( T_d-MoTe2 \).

Fig. 3(a-b) shows the calculated Berry curvature \( \Omega_x \) and \( \Omega_y \) projected on the Fermi surface. The central bulb, a hole pocket at the \( \Gamma \) point, is pinched by two electron pockets along the \( k_z \) direction. From the Berry curvature distribution plot on the Fermi surface, one can anticipate the non-vanishing nature of the \( D_{xy} \) and \( D_{yz} \) terms. Because of the complex metallic bands with anisotropic group velocities in type-II Weyl semimetals, the Fermi surface has significant Berry curvature even away from the WPs (see the SM [53]). This renders the BCDM more sensitive to the chemical potential than for type-I Weyl semimetals [21, 59]. In the latter, the major contribution to the BCDM comes from the WPs, whereas the rest of the Fermi surface makes a negligible contribution due to the isotropic group velocities near the WPs. Therefore, we notice a considerable change in the BCDM with respect to the chemical potential in \( T_d-MoTe2 \).

The nonvanishing \( D_{xy} \) and \( D_{yz} \) terms in the \( T_d-A \) phase, computed as a function of the chemical potential, are shown in Fig. 3(c). \( D_{yz} \) is peaked near the Fermi level, while \( D_{xy} \) exhibits oscillating behavior. (Note that the BCDM is a dimensionless quantity in three dimensions.) At \( E_F \), \( D_{xy} \) and \( D_{yz} \) are estimated to be 1.6 and 6.7, respectively. These values are several times larger than the corresponding \( D_{xy} = 0.8 \) and \( D_{yz} = -0.7 \) reported for \( T_d-MoTe2 \) by Zhang et al. [21]. The main reason behind this difference is the strong sensitivity of the Fermi surface to the on-site Hubbard \( U \) of Mo 4d electrons [25, 36], which was not taken into account in the previous study [21]. Indeed, only four WPs are reported in their study, in contrast to the twelve (8 type-II and 4 type-I) obtained in our case.

From Eq. 1, the nonlinear conductivity tensor has nonzero terms \( \chi_{zzz} = -\chi_{zzz} \) associated with \( D_{xy} \), and \( \chi_{yyz} = -\chi_{yyz} \) associated with \( D_{yx} \). In view of the significant peak in \( D_{yz} \) near \( E_F \), one interesting measurement would be the observation of a transverse current \( j_z \) induced by an oscillating electric field along \( y \) direction. In the \( \omega \rightarrow 0 \) limit, an external electric field applied along the \( y \) direction, i.e., the chain direction, generates an out-of-plane current \( j_z^0 = 2\chi_{yyz} |E_y|^2 \). If one can raise the electron chemical potential via gating, the transverse current \( j_z^0 \) is predicted to rapidly reach its maximum and then decrease, and eventually reverse its sign.

Here, we stress that a structural transition from the \( T_d-A \) to \( T_d-B \) phase flips the sign of \( D_{ab} \) while keeping its magnitude intact, thus, allowing one to distinguish between the two variants of polar \( T_d \) phases. For this purpose, observation of \( D_{xy} \) via \( j_z^0 = 2\chi_{xyz} |E_x|^2 \) may be most suitable, since the sign of \( D_{xy} \) is less sensitive to the electron chemical potential. We do not notice significant differences in the magnitudes of \( D_{ab} \) for the intermediate structures along the polarity reversal path, although new pairs of WPs get created/annihilated as a function of \( \lambda \). This is due to the fact that the newly created WPs belong to the type-I category which have relatively smaller tilt of Weyl cone compared to type-II category, thus yielding minimal changes to the overall BCDM.
An interesting aspect of the nonlinear Hall conductivity in this system is that, because the surfaces have lower symmetry than the bulk, new components of the $D$ tensor are activated at the surface. In particular, the glide mirror $M \tau T (\vec{c}/2)$ is broken at the (001) cleavage surface. Recall that the $D_{yz}$ and $D_{zy}$ tensor elements were argued to vanish in the bulk because of this glide mirror, but they need not vanish at the surface. Thus, response currents associated with the conductivity tensor elements $\chi_{yxx} = -\chi_{xyy}$ and $\chi_{zzz} = -\chi_{zzz}$ are allowed. While we can confidently predict the existence of such currents, we are not currently in a position to compute the surface $D$ tensors quantitatively. This observation thus provides a challenge for future efforts at both theoretical prediction and experimental detection of surface nonlinear Hall responses.

We may also consider the symmetries that remain in the exfoliated single-layer limit. In fact, the $\chi_{yxx} = -\chi_{xyy} \propto D_{yz}$ tensor elements are the only ones to survive in this limit. The other terms, proportional to $\Omega_x$ or $\Omega_y$, are not well defined in two dimensions. Therefore, measuring the in-plane nonlinear Hall conductivity of MoTe$_2$ with respect to the film thickness may reveal a noticeable transition from the film to the surface responses.

In principle, one can utilize the nonlinear response current generated due to the rapid fluctuation of $D_{yx}$ and its sign reversal near the Fermi level as a function of chemical potential to devise a nonlinear Hall transistor for practical applications. Moreover, recent experiments [41, 60] demonstrated an ultrafast optical control over $T_d$ and $1T'$ structural phase transitions, hence, an ultrafast topological optical switch can be designed using the nonlinear quantum Hall property of MoTe$_2$, where $T_d$ (1T') phase can act as an ON (OFF) state.

In summary, we explain the intricate structural phase transitions in MoTe$_2$ by defining a high-symmetry nonpolar phase $T_0$ that exhibits a higher-order topology. We unveil the connection between the Weyl phase and the higher-order topological phase in MoTe$_2$. We report that WPs can be readily created/annihilated, manipulated, and switched by controlling the structural phase transitions between the two polar variants of the $T_d$ phase. Since this structural phase switching has already been experimentally achieved, and is shown to be reversible [12], MoTe$_2$ offers a promising platform to harness the dynamics of Weyl fermions for technological applications. We also report on the presence of a tunable nonlinear Hall effect in $T_d$-MoTe$_2$, and discuss the potential applications of this effect in designing ultrafast topological optical switches and transistors. We further predict that an emergent nonlinear surface response current, arising solely due to the surface effects, can be observed in a slab geometry. One can quantify the magnitude of the emergent surface response current by reducing the dimensionality of the system. The synergy between the possible experimental control over the atomic arrangements, Weyl phase, and the nonlinear quantum Hall effects in MoTe$_2$ paves the way for realization of Weyltronics.

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