Connecting Higher-Order Topology with the Orbital Hall Effect in Monolayers of Transition Metal Dichalcogenides

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Monolayers of transition metal dichalcogenides (TMDs) in the 2H structural phase have been recently classified as higher-order topological insulators (HOTIs), protected by $C_3$ rotation symmetry. In addition, theoretical calculations show an orbital Hall plateau in the insulating gap of TMDs, characterized by an orbital Chern number. We explore the correlation between these two phenomena in TMD monolayers in two structural phases: the noncentrosymmetric 2H and the centrosymmetric 1T. Using density functional theory, we confirm the characteristics of 2H TMDs and reveal that 1T TMDs are identified by a $Z_4$ topological invariant. As a result, when cut along appropriate directions, they host conducting edge states, which cross their bulk energy-band gaps and can transport orbital angular momentum. Our linear response calculations thus indicate that the HOTI phase is accompanied by an orbital Hall effect. Using general symmetry arguments, we establish a connection between the two phenomena with potential implications for orbitronics and spin orbitronics.

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Introduction.—The orbital Hall effect (OHE) refers to the transverse flow of orbital angular momentum (OAM) in response to a longitudinally applied electric field. It resembles the spin Hall effect (SHE) [1–4], but unlike the latter, it does not require spin-orbit coupling (SOC). Characteristics of the OHE and the physical mechanisms underlying it are currently under investigation [5–10]. For instance, signatures of the OHE in 3D metallic systems were recently observed [11,12], paving the way for possible orbitronic applications [1,13,14].

Lately, the OHE in two-dimensional (2D) materials has received a great deal of attention [15–25]. Theoretical calculations predict the existence of orbital textures in some 2D materials, which can give rise to the OHE [17,18,26,27]. They have been observed in insulating transition metal dichalcogenides (TMDs) [28,29], where OHE plateaus are predicted [17–20]. Furthermore, it is possible to attribute an orbital Chern number to this insulating phase [19,20], indicating a connection with nontrivial topology.

The topological nature of layered TMDs is mainly focused on the distorted structural phase 1T’ that hosts topological insulators [30–32]. Less is known about the topology of octahedral (1T) and trigonal prismatic (2H) structures, where most monolayers exhibit an insulating character [33]. MoS$_2$ is an archetype of a 2H TMD that exhibits a large energy band gap. Their zigzag nanoribbons display metallic edge states that cross the bulk gap [34,35], suggesting an underlying topology, despite being trivial with respect to the $Z_2$ index [36].

References [37,38] have revealed that some 2H TMDs are 2D higher-order topological insulators (HOTIs), not previously identified by the SHE signature [39]. Triangular nanoflakes with armchair edges present in-gap corner states with fractional charge, protected by $C_3$ symmetry. Besides being one of the striking features of a 2D-HOTI [39–44], they explain the presence of metallic edge states in the zigzag edges and connect them to the topology of the 2D material. Their topological bulk polarization is perpendicular to the zigzag edges, leading to charge accumulation and metallic edge states.

Here, we use density functional theory (DFT) and linear-response calculations to study the interplay between the OHE and HOTI phases in monolayers of TMDs respecting the rotation symmetry $C_3$. We uncover that centrosymmetric 1T TMDs can also be HOTIs. We then correlate the appearance of a HOTI phase in the two different structures with an orbital Hall insulating phase. We discuss the existence of a pseudo-time-reversal symmetry, which originates from the crystalline symmetries of the lattice.
In analogy with the photonic quantum spin Hall effect, eigenstates of the orbital angular momentum play the role of pseudospins. They are also eigenstates of the $C_3$ rotation operator that protects the HOTI. Because of the orbital nature of the pseudospins, the higher-order topological phase can be witnessed by the OHE. We use a well-known low energy model for HOTIs [45] and the three $d$-orbitals model for 2$H$ TMDs [46] to explicitly show this connection [47].

**OHE calculations for TMDs in the 2H and 1T structural phases.**—We begin by presenting the band structure and the OHE for the two families. For that purpose, we performed DFT calculations for 1$T$- and 2$H$-TMD monolayers ($MX_2$, where $M$ is a transition metal and $X$ is a chalcogen). The TMD structures were obtained from the C2DB database [61]. We also adopted their criteria for dynamic (phonons) and thermodynamic stability. We fully optimize the structural parameters to obtain the wave functions. Then, we construct a PAO Hamiltonian using the pseudo-atomic-orbital (PAO) projection method [62–65] for each compound. This method is implemented in the PAOFLOW code [66,67]; for technical details see Supplemental Material (SM).

Once the PAO Hamiltonian $H_{PAO}(k)$ is built, we calculate the spin Hall (SH) and orbital Hall (OH) conductivities to linear order on the external electric field [17–19]

\[
\sigma_{\eta,SH}^{\eta} = \frac{e}{(2\pi)^2} \sum_n \int_{BZ} \hat{d}^2 k f_n \Omega_{\eta,n}^{X}, \tag{1}
\]

where $\sigma_{\eta,SH}^{\eta}$ is the OH (SH) dc conductivity with polarization along the $\eta$ direction, $f_n$ is the Fermi-Dirac distribution, and $\Omega_{\eta,n}^{X}$ is the angular momentum projected Berry curvature in the intra-atomic approximation [17–19] (see SM [47]).

Figures 1 and 2 show the fully relativistic band structures, OH conductivities, and the orbital Berry curvatures calculated for different TMD monolayers. In Fig. 1 we depict results for 2$H$ TMDs CrS$_2$, MoS$_2$, and WS$_2$. We note that within their insulating gaps these systems exhibit OH conductivity (OHC) plateaus with values $\sigma_z$ = 1.89, 2.65, and 1.43 in units of $(e/2\pi)$, respectively. Conversely, $\sigma_x$ and $\sigma_y$ vanish for all three systems. Their corresponding orbital Berry curvatures are similar. They are peaked around the $K$ point with similar maxima for the three systems, giving a large positive contribution to the OHC. Around $M$, the Berry curvature becomes negative.
TABLE I. Main characteristics of the 1T-TMD monolayers insulating phases of NiS$_2$, PdS$_2$, and PtS$_2$. $Z_M$ is the atomic number of the constituent transition metal atom. The columns $\sigma_{\text{OH}}$, $\sigma_{\text{OH}}^2$, $\sigma_{\text{OH}}^3$, and $|\sigma_{\text{OH}}|$ show their OHE in-gap values in units of $e/(2\pi)$. $E_g$ is the energy band gap and the last column shows the values of the topological invariant $Z_4$.

| TMD (Z$_M$) | $\sigma_{\text{OH}}$ | $\sigma_{\text{OH}}^2$ | $\sigma_{\text{OH}}^3$ | $|\sigma_{\text{OH}}|$ | $E_g$ (eV) | $Z_4$ |
|-------------|-----------------|-----------------|-----------------|-----------------|-------------|------|
| NiS$_2$ (28) | 0.123 | -1.78 | 2.05 | 0.54 | 2 |
| PdS$_2$ (46) | 0.73 | -1.22 | 1.85 | 1.14 | 2 |
| PtS$_2$ (78) [69] | 0.51 | -0.77 | 0.92 | 1.72 | 2 |

and has different values for the three compounds, resulting in the plateaux variations obtained for the OHCs when we go from Cr to W. Since the top valence bands of the TMDs have predominantly $d$ character, the chalcogen element does not significantly affect the OHC. Table 1 of the SM [47] shows that the in-gap OHC does not change much when S is replaced by Se or Te.

Figure 2 presents results for the 1T-TMDs monolayers NiS$_2$, PdS$_2$, and PtS$_2$. Differently from the 2H TMDs, the OHCs for the 1T TMDs with $y$ polarization are not zero, and thus contribute to the entire OHC. We follow Ref. [68] and define the absolute value of the OHC as $|\sigma_{\text{OH}}| = \sqrt{(\sigma_{\text{OH}}^2)^2 + (\sigma_{\text{OH}}^3)^2 + (\sigma_{\text{OH}}^4)^2}$. Within each energy band gap, we clearly see that the dominant contribution to $|\sigma_{\text{OH}}|$ is $\sigma_{\text{OH}}^1$ for all three systems, and the OHC plateaux reduces as we move from Ni to Pt.

Table I summarizes the main findings depicted in Fig. 2. It is noteworthy that both 2H- and 1T-TMD monolayers exhibit finite OH conductivity plateaus within their insulating energy gaps. This shows that the OHE in 2D materials is not constrained by spatial inversion symmetry and can also appear in centrosymmetric monolayers. Our results are in line with recent predictions showing that centrosymmetric 2H-TMD bilayers also display OHC plateaus [18–20]. We note that $|\sigma_{\text{SH}}| = 0$ inside the band gap for both sets, which is consistent with the fact that TMDs do not exhibit a quantum spin Hall effect (QSHE) in the structural phases studied here. In the SM, we include tables containing several 2H and 1T semiconducting TMD monolayers [47]. We present their electronic band structures and orbital-weighted Berry curvatures for the valence bands and discuss some of their features.

HOTI phase.—We now proceed to the characterization of the HOTI phases. Recent works showed that triangular nanoflakes of 2H-TMD monolayers with armchair edges present in-gap corner states with fractional charge ($-\frac{1}{2} |e|$), protected by $C_3$ symmetry [37,38]. We begin our analysis by calculating the topological indicators for this TMD family.

For noncentrosymmetric materials, the HOTI phase is protected by a $C_4$ rotation symmetry. It can be identified by the symmetry representations of the occupied energy bands at special high-symmetry points (HSPs) of the first Brillouin zone (BZ) [44]. For $C_3$ rotation symmetry, we take $|K_2^C| = |K_2^C| - |K_2^C|$, where $|K_2^C|$ and $|K_2^C|$ represent the number of occupied bands with symmetry eigenvalue $e^{2\pi i (p-1)/3}$ (for $p = 1, 2, 3$) at the $K$- and $\Gamma$-high-symmetry points, respectively. The final topological indicator $\chi^C$ and corner charge $Q^C$ are given by

$$\chi^C = \langle |K_1^C|, |K_2^C| \rangle \mod e, \quad Q^C = 2\pi e/3. \quad \text{(2)}$$

where $e$ is the elemental charge. We use the software IReP [70] to calculate the symmetry eigenvalues of the occupied DFT energy bands. With them, we calculate the topological indicator and the corner charge with the expressions above. The three 2H-TMD monolayers presented here have the same topological indicator $\chi^C = [-1, 2]$ and corner charge $Q^C = 2e/3$. A table for several 2H TMDs is included in the SM [47].

To complement the analysis based on the eigenstates of the rotation operator, we use DFT to examine 2H-TMD triangular flakes with armchair edges and confirm the presence of in-gap corner states, as shown in Fig. 3. 2H TMDs also display an electronic dipole $P = \left(\frac{1}{3}, \frac{2}{3}\right)$, which is perpendicular to the zigzag direction [37]. As a result, if the system is cut in the zigzag direction, there is charge accumulation at the edges, leading to metallic edge states.

Differently from the 2H TMDs, 1T TMDs are HOTIs protected by inversion symmetry. Hence, they are characterized by the $Z_4$ indicator, which can be calculated from the inversion parities of occupied bands [71,72]:

$$Z_4 = \sum_{k \in \text{TRIM}_i} n_-(k_i) \mod 4,$$

where $n_-(k_i)$ is the number of odd parity occupied Kramer pairs at the time-reversal invariant momenta (TRIM) points $k_i$ in the BZ.

The index $Z_4 = 2$ warrants that 1T TMDs also present conducting edge states capable of carrying OAM currents. This can be confirmed from the energy bands of a PtS$_2$ nanoribbon with zigzag edges portrayed in Fig. 4. This figure also highlights the orbital projection of the edge states. Because of the inversion symmetry, Bloch states of 1T TMDs do not exhibit net OAM but can still display an

FIG. 3. Monolayer MoS$_2$ nanoflake (0D) geometry. (a) Fully relativistic calculation eigenvalues. The corner states are highlighted in red. (b) Real space projection of the eigenfunctions for the corner states highlighted in panel (a). Isosurface value of 0.003 eÅ$^{-3}$. 

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For 2H TMDs, the conduction and valence bands at K and K' are mainly composed of d_{x^2} and d_{y^2} orbitals, which are eigenstates of L_z. This is a consequence of the D_{3h} point group symmetry of the crystal: the d_{x^2} orbitals belong to the unidimensional irreducible representation A_1, while the d_{xy} and d_{z^2-y^2} belong to E'. Since the representation E' is two dimensional, the linear combinations d_{\pm} can be treated as pseudospins that transform under a pseudo-time-reversal symmetry operator related to the rotation operators. In the SM [47], we show that the Chern number associated with the \( T_p \) symmetry leads to the same result previously obtained [19].

In the case of the 1T TMDs, the identification of the pseudospins is more subtle. The D_{3d} symmetry imposes that the \( p \) orbitals of the X atoms and the \( d \) orbitals from the M atom should be dominant for the bulk energy states near \( \Gamma \). Our first-principles calculations [47] evince a strong energy splitting in \( \Gamma \) between the \( p_x \), \( p_y \), and \( p_z \) orbitals, as previously reported by Yao et al. for PtSe_{2} [76]. The valence and conduction bands near the gap have a strong contribution of linear combinations of \( p_x \) and \( p_y \) orbitals of the X atom that form \( p_{\pm} \), which transform as \( T_p^2 = -1 \). They are also composed of \( d_{\pm} \). The sizable contribution of \( d_{xz} \) and \( d_{yz} \) orbitals explains the \( L_y \) component of the OHE.

When the system is cut into a ribbon, these orbitals participate in the formation of edge states, as shown in Fig. 4(b). Therefore, in contrast with 2H TMDs, these edge states will be mainly composed of combinations of three pseudospins, one formed by the \( p_x \), \( p_y \) orbitals and two composed of the \( d_{xz} \), \( d_{yz} \), \( d_{z^2-y^2} \), and \( d_{xy} \) orbitals.

In principle, similar to photonics systems, one could use these eigenstates of \( L_z \) to emulate topological phases in fermionic materials: a system which is invariant under inversion and \( T_p \), must have pseudospins forming Kramers’ pairs. However, differently from bosons, fermions have half-integer spins, and two spin-degenerate states for each pseudospin. Therefore, if one tries to construct a fermionic system without spin-orbit coupling where the pseudospins emulate a quantum spin Hall insulator, the system has spin-degenerate pseudospins Kramers’ pairs. This results in an even number of Kramers’ pairs and the system cannot be indexed by a \( Z_2 \) = 1, although it can be a HOTI.

To illustrate these ideas, we use the low energy Hamiltonian presented in Ref. [45] to model HOTIs protected by \( C_3 \) rotation and inversion symmetries (see SM [47]). This model consists of a block diagonal Hamiltonian containing basically the superposition of two copies of the Bernevig-Hughes-Zhang (BHZ) model. It is well known that each BHZ Hamiltonian presents a \( Z_2 \) topological phase. Its eigenstates can also be written in terms of pseudospins that are eigenstates of \( L_z \). Surprisingly, we show they present an orbital Hall plateau in their topological gap. When the two BHZ copies are
taken into account, the system is not a topological insulator but, as shown in Ref. [45], it is a HOTI that has twice the number of edge states of the BHZ model. As expected, the HOTI still presents an orbital Hall plateau and the orbital current can be carried by the in-gap edge states.

To strengthen this link, we used another model to show the onset of a HOTI phase in systems without inversion symmetry, following the ideas presented in Ref. [77]. We considered the simplified three orbital tight-binding Hamiltonian in a triangular lattice that describes the low-energy properties of $2H$ TMDs. We begin with a case with inversion symmetry and orthogonality between the orbitals in different representations. Under this condition, a strong spin-orbit coupling opens a gap in the system, leading to a trivial insulator phase with a vanishing OHE and the absence of in-gap edge states. From this, we identify that if inversion symmetry is broken and hopping between orthogonal orbitals is allowed, as in the case of $2H$ TMD, there is a topological transition to a HOTI that presents zigzag metallic edge states and a large OH plateau, which is independent of the SOC [47].

**Conclusions.**—We employed DFT and linear response transport calculations to study the interplay between the orbital Hall effect and higher-order topological phases. We analyzed all stable $2H$ or $1T$ monolayer TMDs and found that they are HOTIs, protected by either $C_3$ rotation symmetry ($2H$) or inversion symmetry ($1T$). Simultaneously, they all display a plateau in the orbital Hall conductivity inside the band gap.

Recent works start to uncover the role of orbital hybridization in HOTIs and the advent of orbital effects [77–79]. Here, we connect the HOTI phase to the existence of pseudo-time-reversal operators and associated pseudospinors. As these pseudospinors are eigenstates of the orbital angular momentum, HOTI phases can generate an OHE. More importantly, HOTIs with edges that are perpendicular to their bulk polarization present in-gap metallic edge states that can carry the orbital angular momentum in the orbital Hall insulating phase. This can be employed for efficient orbital current injection in novel spin-orbitronics devices. Furthermore, the OHE in 2D HOTIs may be used in machine learning strategies for spotting potentially useful materials for orbitronic applications [80–82].

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