Van der Waals stacking-induced diverse topological phases in higher-order topological insulators

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Abstract

Two-dimensional (2D) tetragonal group-V materials have attracted considerable interest, while their topological properties remain largely unexplored. Using density functional theory, topological quantum chemistry theory, and a tight-binding model, we show that monolayer tetragonal antimonene (T-Sb) realizes a higher-order topological insulator (HOTI). A layer-dependent analysis reveals an even-odd oscillation between HOTI and trivial phases with varying layer numbers. Upon stacking these 2D layers into a three-dimensional structure, successive phase transitions from a metal to a strong topological insulator and eventually to a weak HOTI are driven by tuning the interlayer van der Waals (vdW) coupling. Tight-binding (TB) model analysis shows that these transitions are closely related to the vdW-modulated vertical hopping strength and onsite energy of Sb p_z orbitals. Moreover, the TB model predicts a weak topological insulator phase within an appropriate parameter regime of interlayer vdW interaction and spin-orbit coupling, as verified in T-Sb materials. These results highlight vdW engineering as an experimentally accessible strategy for designing diverse topological phases based on 2D HOTIs.

Result1: Higher-order topology in monolayer T-Sb

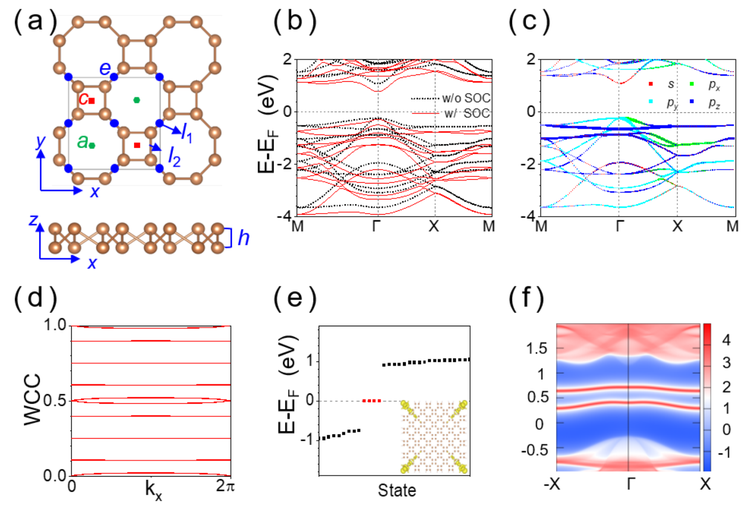

Fig. 1: (a) Top and side views of the T-Sb monolayer. The maximal Wyckoff positions (2a, 2c, and 4e) are also given. (b) Band structures of the 1L T-Sb. (c) Projected band structures of Sb *s* and *p* orbitals in the 1L T-Sb without SOC. (d) Evolution of the Wannier charge centers of 1L T-Sb without SOC. (e) Energy levels of a finite 1L T-Sb flake calculated by using DFT, where red dots denote corner states. The bottom-right inset shows the distribution of the corner states in the T-Sb flake. (f) Edge states of a semi-infinite 1L T-Sb without SOC.

Result2: Even-odd oscillation behavior in few-layer T-Sb

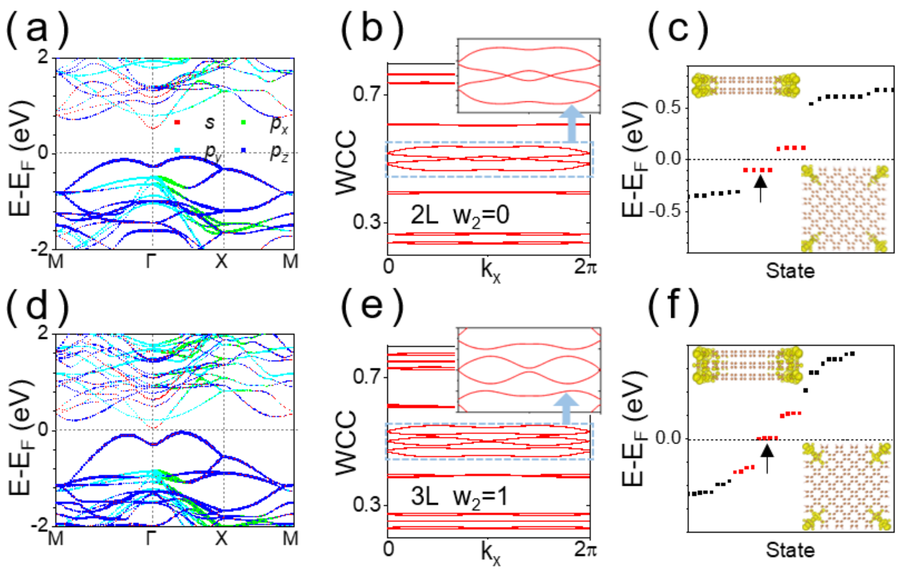

Fig. 2: (a) Projected band structures of the 2L T-Sb without SOC. (b) Evolution of the WCCs in the 2L T-Sb. The inset shows an enlarged view of the region marked by the blue dashed rectangle. (c) Energy levels of a 2L T-Sb flake calculated by using DFT. The black arrow marks the occupied corner states. The two insets illustrate the side and top views of the corner states in the 2L T-Sb flake. (d-f) Same as (a-c), respectively, but for 3L T-Sb.

Result3: Diverse topological phases in 3D T-Sb

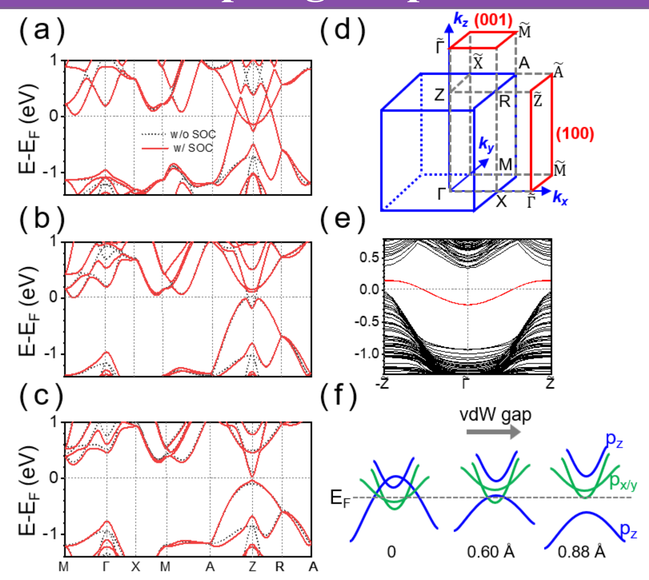

Fig. 3: (a-c) DFT-calculated band structures of 3D T-Sb with interlayer expansions along the *z* direction of ΔH = 0.0 Å, 0.60 Å, and 0.88 Å, respectively. The SOC is not taken into account. (d) The 3D Brillouin zone with its projected (001) and (100) planes. (e) DFT-calculated hinge states: four degenerate states near E_F. (f) Schematic illustrating the shifting trend of $p_{x/y}$ and p_z orbitals with decreasing vdW coupling, based on DFT calculations.

Result4: Tight-binding model for 3D T-Sb

A 3D TB model involving the $p_{x/y}$ and p_z orbitals on each Sb atom is constructed to elucidate the mechanism responsible for the rich topological phases in the 3D T-Sb. The TB Hamiltonian is written as

$$H_{3D} = H_{\text{onsite}} + H_{\text{in}} + H_{\text{ver}} + H_{\text{soc}},$$

where H_{onsite} describes the orbital-dependent onsite energies,

$$H_{\text{onsite}} = \varepsilon_{p_{x/y}} \sum_i c^{\dagger}_{p^i_{x/y}} c_{p^i_{x/y}} + \varepsilon_{p_z} \sum_i c^{\dagger}_{p^i_z} c_{p^i_z}.$$

In Eq. (6), $c^{\dagger}_{\alpha^i}$ (c_{α^i}) represents a creation (annihilation) operator for an electron with α ($\alpha = p_{x/y}$ and p_z) orbitals on site i. ε_α is the onsite energy for orbital α. The H_{in} term in Eq. (5) denotes the intralayer Hamiltonian,

$$H_{\text{in}} = \sum_{i \neq j} V_{ij} (c^{\dagger}_i c_j + h.c.),$$

where V_{ij} indicates the hopping integral between sites i and j within each layer.

According to the DFT band structure, the interlayer coupling is dominated by the p_z orbital, whereas the p_x and p_y orbitals contribute primarily to the intralayer interactions and are thus negligible for the interlayer hopping. Therefore, the H_{ver} term in Eq. (5), which describes the vertical interlayer hopping, is only considered for the p_z orbital:

$$H_{\text{ver}} = \sum_{\substack{i \in 4,5,6,7 \\ j^+ \in 0,1,2,3}} J_z m_{p^i_z, p^{j+}_z} c^{\dagger}_i c_{j+} + \sum_{\substack{i \in 0,1,2,3 \\ j^- \in 4,5,6,7}} J_z m_{p^i_z, p^{j-}_z} c^{\dagger}_i c_{j-} + h.c.,$$

where i labels the intralayer atomic sites within a given Sb layer, while j^+ and j^- denote the corresponding atomic sites in the adjacent upper and lower Sb layers, respectively. The J_z and $m_{i,j}$ indicate the interlayer interaction coefficient and hopping integral via the p_z orbitals, respectively. Only the nearest-neighbor hopping interactions are considered in the model. The H_{soc} term in Eq. (5) represents the onsite SOC term. Excellent agreement with the DFT band gap and dispersion is achieved by using three fitting parameters: J_z, ε_{p_z}, and λ, where λ denotes the SOC strength.

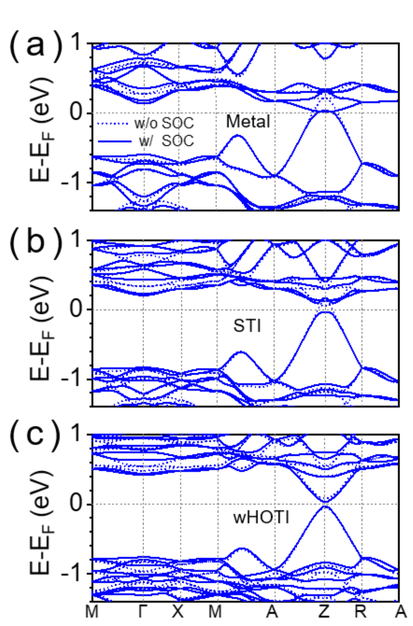

Fig. 4: Band structures of the TB model without SOC ($\lambda = 0$ eV, dotted curves) and with SOC ($\lambda = 0.11$ eV, solid curves). The parameters (in eV) are: (a) $J_z = 0.38, \varepsilon_{p_z} = 1.21$, (b) $J_z = 0.30, \varepsilon_{p_z} = 0.64$. (c) $J_z = 0.25, \varepsilon_{p_z} = 0.27$. Panels (a–c) correspond to metal, STI, and wHOTI phases, respectively. The trend is consistent with the DFT results shown in Figs. 3(a-c).

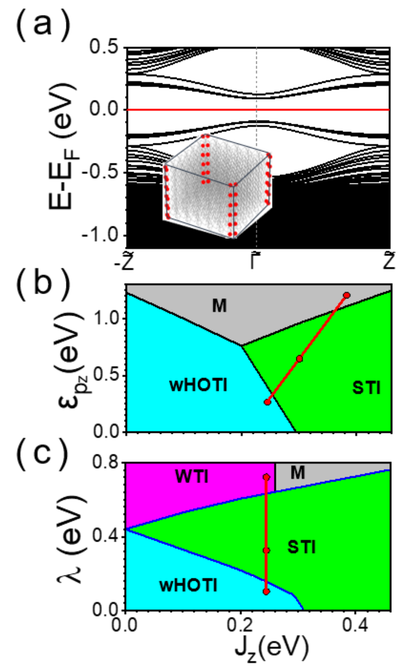

Fig. 5: (a) Hinge (red) and bulk (black) states from the TB model with parameters as in Fig. 5(c). The inset in (a) shows the hinge-state charge distribution. (b, c) Phase diagrams of 3D T-Sb from the TB model, showing the wHOTI (blue), STI (green), WTI (magenta), and metallic (gray) phases. In (b) and (c), the λ and ε_{p_z} are fixed at 0.11 eV and 0.27 eV, respectively. In (b), the three red points on the red line (from right to left) correspond to Figs. 5(a–c), respectively. In (c), the three red points on the red line (from bottom to top) correspond to Figs. S10(a, b, and d) in the SM[45], respectively.

Conclusions

We demonstrate that monolayer group-V T-Sb is a higher-order topological insulator, characterized by corner states originating from a filling anomaly at the 4*e* Wyckoff position. Layer-dependent calculations reveal an even–odd oscillatory topological transition between HOTI and trivial insulating phases, with its oscillation period dictated by the denominator of the corner charge (*e*/2) in the monolayer limit. By engineering the stacking of 2D HOTI layers with tunable interlayer coupling, we realize various topological phases in 3D T-Sb, including STIs and wHOTIs. Notably, the metal-STI–wHOTI topological transitions in 3D T-Sb is quantitatively captured by the established TB model, arising from the reduced vdW interlayer hopping and onsite Sb p_z orbital energy. The interplay between the vdW interlayer coupling strength and the SOC can also give rise to rich topological phases. Intriguingly, a novel WTI phase predicted from the TB model is confirmed in 3D T-Sb by DFT calculations. Our work identifies T-Sb as a versatile functional platform for tailoring tunable topological phases and provides a practical design principle for engineering layered topological materials.

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References

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