Remote control of spin polarization of topological corner states

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In two-dimensional higher-order topological insulators, the corner states are separated by a non-negligible distance. The crystalline symmetries protect the robustness of their corner states with long-range entanglement, which are robust against time-reversal-breaking perturbations. Here, we demonstrate the possibility of direct control of the topological corner states by introducing the spin degree of freedom in a rhombus-shaped Kekulé nanostructure with local magnetization and local electric potential. By applying a local magnetization on one corner, the other corner can also be strongly spin polarized. By further applying a local electric potential at the same corner, the sign of the spin polarization can be reversed at both corners. We also prove the robustness of the control of the spin polarization under the disorder. Moreover, we demonstrate the material realization in a γ-graphyne nanostructure with Mn adsorption and Si replacement at one corner by using first-principles calculations. Other higher-order lattices and the shape of the nanostructure are also discussed. Our studies give a showcase of the remote correlation of quantum states in higher-order topological materials for spintronic and quantum applications.

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Research interest in topological insulators (TIs) [1] has stayed extremely high as their gapless edge or surface states are robust against perturbations and hence have potential for diverse technological innovations [2–5]. Recently, the proposal of electric multipole insulators [6] inspired explorations of a new class of topological materials, so-called higher-order topological insulators (HOTIs) [7–14], which show lower-dimensional topological corners or hinge states. Several two-dimensional (2D) lattices have been proposed for the realization of the HOTI phase, including a square lattice [6,8–12], a breathing Kagome lattice [15,16], a Kekulé lattice [17–25], and nonperiodic quasicrystals [26]. The difference between the intercell and intracell hopping parameters drives these systems into the higher-order topological phase by opening a gap for the edge states and generating more localized topological corner states. In analogy to the conventional bulk-edge correspondences [1,27,28] in 2D TIs, the higher-order corner states manifest the bulk-corner correspondence [15–25,29,30] or the edge-corner correspondence [6,8–12,29,31]. As the corner states are protected by certain crystalline symmetries, the higher-order topology is preserved in the presence of size variations, defects, and disorder, even with the time-reversal symmetry breaking. The robustness of the topological corner states has been proved both theoretically and experimentally in electronic crystals [19,32–36], photonic crystals [25,37–40], acoustic networks [41,42], and bound states in the continuum [43].

It is known that the topological edge states in 2D TIs lead to the quantum spin Hall effect (QSHE) [44–46] or the quantum anomalous Hall effect (QAHE) [47–50], and their possible applications in quantum computing and spintronic devices have been extensively investigated [51–54]. Comparatively, the potential use of the corner states in 2D HOTIs has rarely been discussed because even their realization in real materials remains a challenge. Experimental studies of HOTIs to date were mostly based on metamaterials [37,39–41,55–61], and some new phenomena have been reported such as the valley-selectivity of corner states of a sonic crystal [62]. In this regard, it is essential to find a real solid HOTI, establish a profound understanding, and design conceptual devices for benefiting from the discovery of this new topological phase. Since spin is one of the most important factors in all topological studies [47–50], it is natural to perceive that the control of the spin degree of freedom of the topological corner states is crucial for the application of HOTIs. Although the artificial pseudospin degree of freedom has been introduced for the topological corner state [20], the study of their real-spin degree of freedom is still barren.

In this paper, we propose the possibility of making remote magnetoelectric control in rhombus-shaped Kekulé nanostructure, through theoretical studies using the tight-binding (TB) method and computational studies using first-principles calculations with tunable local magnetization $M$ and local electric potential $V$. By magnetizing one corner, the other corner separated by a non-negligible distance can also be spin polarized. Furthermore, electric manipulation at the same corner may alter the sign of spin polarization (SP) at both corners due to the newly generated corner state with the opposite spin, as in the schematic figure shown in Fig. 1. The higher-order topology ensures the existence of the corner states within the edge gap, giving us opportunities to generate new corner states by applying local electric potential. Accordingly, the control of SP is robust under disorder, which is also discussed by considering edge and bulk disorder. Importantly, we demonstrate the material realization of HOTI...
FIG. 1. Schematic figure of remote control of spin polarization in a higher-order topological insulator. By applying local magnetization $M$ and electric potential $V$ on one corner, the other corner can be spin-polarized and the sign of the spin polarization can be reversed on both corners. The red and blue arrows represent the positive and negative spin polarization, respectively.

using a $\gamma$-graphyne nanostructure with density-functional theory (DFT) calculations which show identical responses to the manipulations of $M$ and $V$ as predicted by the TB model. Here, $M$ and $V$ are applied by using Mn adsorption and Si substitution at one corner of the $\gamma$-graphyne flake. We also show similar results in a breathing Kagome lattice model with a triangular-shaped nanostructure. This study extends the understanding of HOTIs and gives a showcase of using the topological corner states in quantum and spintronic devices.

We start from the TB model Hamiltonian for the Kekulé lattice:

$$H_0 = t_0 \sum_{\langle ij \rangle \alpha} c_{i \alpha}^\dagger c_{j \alpha} + t_1 \sum_{\langle i'j' \rangle \alpha} c_{i' \alpha}^\dagger c_{j' \alpha}. \quad (1)$$

Here, $c_{i \alpha}^\dagger$ is the operator of the electron creation on-site $i$ of spin $\alpha$, $\langle ij \rangle$ and $\langle i'j' \rangle$ represent the nearest-neighbor hopping for intracell ($t_0$) and intercell ($t_1$), respectively, as shown in the inset of Fig. 2(a). As reported in previous studies [17–25], a HOTI phase is created on the Kekulé lattice when $t_1 > t_0$. Here, we choose $t_1/t_0 = 1.25$ in the following discussions. The gapped edge states and parity calculations confirmed the higher-order topology with $Z_2 = 1$ (see Supplemental Material, Sec. I [63]). To show the topological corner states, we construct a rhombus-shaped nanostructure with an $11 \times 11$ Kekulé lattice [Fig. 2(a)]. There is a pair of states located in the gap of the bulk and edge states around the Fermi level, representing the bonding and antibonding corner states ($C_0$ and $C_0'$) in Fig. 2(b), which can be understood as the constructive and destructive interference of the wave functions at two corners. This means that the wave functions of bonding and antibonding corner states are the hybridization of the wave functions at each independent corner. As the two corners are geometrically identical, the hybridized wave functions of the bonding and antibonding corner states are evenly distributed at two corners, as shown in Figs. 2(c) and 2(d). Note that the topological local states only appear at the 120$^\circ$ corners due to the chiral charge cancellation at the 60$^\circ$ corners for the bipartite lattices [19].
Next, we consider the interplay between $M$ and $V$ on the corner states. The total Hamiltonian becomes

$$H_{\text{tot}} = H_0 - \lambda_z \sum_{\alpha} c_\alpha^\dagger (\sigma_{\alpha} \otimes \tau) c_\alpha + V \sum_{\alpha} c_\alpha^\dagger (I_2 \otimes \tau) c_\alpha,$$

where $\lambda_z$ represents exchange splitting, $V$ represents electric potential, $\alpha$ is the spin index, $\sigma_\alpha$ is the Pauli matrix and $I_2$ stands for a $2 \times 2$ identity matrix. $\tau$ represents $2 \times 2$ matrix acting on the two corners, where

$$\tau = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$$

represents $M$ or $V$ applied on the bottom-right corner including six atomic sites on each corner [Figs. 2(e) and 2(i)]. The SP of the corner states is defined as $SP = |\langle \alpha_1 | \psi \rangle|^2 - |\langle \alpha_2 | \psi \rangle|^2$, where $\psi$ is the wave function of the corner states. We first choose parameters $\lambda_z = 0.5I_0$, and $V = I_0$ to illustrate the influence of $M$ and $V$.

When the bottom-right corner is magnetized, as shown in Fig. 2(e), the spin degeneracy and geometrical symmetry are broken for the corner states. The wave functions of the corner state become unevenly distributed at two corners, as shown in Figs. 2(f) and 2(g). The positive $M$ applied on the bottom-right corner induces a spin-down polarization at the top-left corner spontaneously. The SP shows antiparallel couplings for the two corners [Fig. 2(h)]. Interestingly, when $V$ $(V > V_c$, where $V_c$ is the critical point of generating a new corner state) is applied on the same corner, as shown in Fig. 2(i), the spin-up bonding corner state becomes localized at the top-left corner [Fig. 2(j)], and a new spin-down corner state is generated at the bottom-right corner [Fig. 2(k)]. As a result, the sign of the SP is reversed at both corners, comparing Fig. 2(h) with Fig. 2(i). In a finite-size system, each bonding or antibonding corner state contains information on both corners, so that the manipulation of one corner can change the weight of the wave-function distribution at both corners. Besides, the higher-order topology of the bulk or edge states endows the existence of the topological corner states within the edge gap due to the bulk-corner [15–25,29,30] or the edge-corner correspondence [6,8–12,29,31]. This anomalous property gives us opportunities to lift or lower the energy level of the corner states to rearrange the wave-function distributions and generate new corner states within the edge gap. Consequently, the manipulation of $M$ and $V$ at one corner not only induce the SP at the other corner but also reverse the sign of the SP at both corners. This offers an innovative way to remotely control the magnetization of HOTI flakes.

To understand these important findings, we investigate the evolution of the corner states with $M$ and $V$, respectively. First, let us consider the change of $\lambda_z$ when $V = 0$, as shown in Fig. 3(a). When a positive $M$ is applied, the spin-up branch of $C_0$ is gradually lowered in energy and becomes $C_1$ more localized at the bottom-right corner. Meanwhile, the spin-down branch of $C_0$ becomes $C_2$ more localized at the top-left corner. Similarly, $C_0'$ also splits into two localized states, $C_2$ and $C_3'$, with opposite spins. The wave function distributions of these four corner states are shown in Fig. 3(a) for $\lambda_z = 0.5I_0$. With increasing $\lambda_z$, the spin splitting is enlarged and the change of energy of the bottom-right corner states ($C_1$ and $C_4'$) is much faster than the other corner states ($C_2$ and $C_3$). When $\lambda_z$ is further increased, the corner states $C_1$ and $C_4'$ merge into the edge states, and $C_2$ and $C_3$ switch their occupation statuses. Correspondingly, the edge states adjacent to the bottom-right corner become more and more localized and generate the new corner states ($C_7$ and $C_8$) in a concave shape, as shown by their wave-function distributions in Fig. 3(a) for $\lambda_z = 2I_0$. The evolution of energy levels in a larger regime can be found in the Supplemental Material, Sec. II [63]. The other two newly generated states ($C_7$ and $C_8$) are trivial localized states belonging to the corner hexagon on which $M$ is applied [see the two wave-function distributions at the bottom right of Fig. 3(a)]. Due to the strong localization, their energy-changing rate $\partial E/\partial \lambda_z$ is much larger than the topological corner states that contain edge components. More details of the trivial localized states are discussed in the Supplemental Material, Sec. III [63].

Next, we set $\lambda_z = 0$ and focus on the influence of $V$. As shown in Fig. 3(b), both wave-function distributions of $C_0$ and $C_0'$ are weighted more toward one corner as the symmetry of the two corners is broken by $V$. The wave functions of new

![FIG. 3. Discrete energy levels of the corner states (a) as a function of local magnetism $\lambda_z$ with $V = 0$, and (b) as a function of local electric potential $V$ with $\lambda_z = 0$. The red, blue, and gray circles represent the spin-up, spin-down, and spin-degenerate energy levels, respectively. The wave function distributions on the right panels correspond to the typical levels labeled as the solid circles on the left plots.](image-url)
corner states \((C_{0}^{(1)}\) and \(C_{0}^{(2)}\)) are unevenly distributed at the two corners with the addition of \(V_{c}\), as shown in Fig. 3(b) for \(V = 0.5t_{0}\). When \(V\) further increases, \(C_{0}^{(2)}\) merges into the edge states and a new corner state \(C_{0}^{(3)}\) emerges from the valence edge states. The distribution of \(C_{0}^{(3)}\) shows a concave shape, as shown in Fig. 3(b) for \(V = 2t_{0}\). Again, a trivial localized state \(C_{0}^{(4)}\) also forms at the bottom-right tip hexagon [see the bottom right of Fig. 3(b)]. It is important to note that the persistence of having the corner states in our model against either \(M\) or \(V\) is another clear indication of its HOTI nature.

Now we investigate the response of the topological corner states to the interplay of \(M\) and \(V\). For simplicity, we plot the schematic figure of energy evolutions of corner states under the influence of \(M\) and \(V\) in Fig. 4(a). Without \(M\) and \(V\), the wave functions of \(C_{0}\) and \(C_{0}^{'t}\) are evenly distributed on both corners. With a positive \(M\) at the bottom-right corner, they split into four states, as already shown in Fig. 3(a). If we further add a local positive \(V\) at the same corner, all states at the bottom-right corner (\(C_{1}\) and \(C_{4}\)) are lifted, and the energies of the states at the top-left corner (\(C_{2}\) and \(C_{3}\)) are also slightly affected, as sketched in Fig. 4(b). The different lifting rates cause a crossing of \(C_{1}\) and \(C_{3}\) at \(V = \lambda_{z}\) (labeled \(C_{1}\) and \(C_{3}\) in Fig. 4), which restores their even distributions at two corners. After the crossing, \(C_{3}\) is gradually localized again at the top-left corner and becomes a bonding corner state. In contrast, \(C_{1}\) with the wave function localized more at the bottom-right corner becomes the antibonding part of the \(C_{1}\) and \(C_{3}\) rehybridization. For the spin-down channel, the lifting of \(C_{4}\) is faster than \(C_{2}\) so they never cross or rehybridize with positive \(V\). When \(V > V_c\), a new spin-down state \(C_{5}\) is lifted into the edge gap jointly by \(V\) and \(M\). This newly generated state \(C_{5}\) is a topological corner state protected by higher-order topology. Spontaneously, \(C_{5}\) crosses the Fermi level and becomes unoccupied. \(C_{4}\) merges into the conduction edge states. The spins of the two occupied corner states \(C_{3}\) and \(C_{5}\) are reversed from those for small \(V\) \((C_{1}\) and \(C_{2}\)). As a result, the SP changes its sign at both corners across \(V_c\), as shown in Figs. 2(h) and 2(l).

In addition, we investigate the robustness of existence and control of the topological corner states by introducing randomized on-site disorder \([19,32,36]\). The random on-site potential \(\epsilon_{n}\) is added to \(H_{tot}\) in Eq. (2) for all atoms except the three outmost atoms on each corner. Here, \(\epsilon_{n}\) denotes the additional potential for site \(n\) and is randomized in a range of \([-W/2, W/2]\), corresponding to Anderson disorder with a strength of \(W\) \([76]\). Figure 5 shows the disorder configurations with \(W = 0.2t_{0}\) and \(W = 0.5t_{0}\), which are in the same energy scale with the edge and bulk band gaps. Using the same parameters as results in Fig. 2, we find that the inclusion of \(\epsilon_{n}\) makes no qualitative change. With and without applying the electric potential, the sign of the SP is reversed at both corners when the disorder strength goes up to the bulk band gap. The corresponding energy levels of the corner states are barely changed. More details can be seen in the Supplemental Material, Sec. IV \([63]\).

From the discussions above, there are three key messages from our systematic studies for the control of spin-polarized corner states: (1) Magnetizing one corner can induce the SP of the other corner separated by a significant distance. (2) Raising the electric potential of one corner can change the wave-function distributions of the bonding and antibonding corner states. (3) The manipulation of \(V\) with \(M\) on the same corner can reverse the sign of SP at both corners. The use of negative \(V\) shows the same conclusions (see Supplemental Material, Sec. V \([63]\)). Besides, the shape or the size of the nanostructure is not constrained as the higher-order topology ensures the robustness of the corner states due to the bulk-boundary correspondence. A larger size of Kekulé nanostructure shows similar manifestations (see Supplemental Material, Sec. VI \([63]\)). We also investigate a hexagonal-shaped Kekulé nanostructure with six topological corners. The correspondence between all the corners needs to be considered if we have more than two topological corners. And the main conclusion for the remote control of corner states is still valid for a more complex shaped system (see Supplemental Material, Sec. VII for more details \([63]\)). Furthermore, we investigate the case having \(M\) at both corners.
with ferromagnetic and antiferromagnetic orders. With $V$ applied on one corner, a transition happens between parallel and antiparallel SPs at two corners (see Supplemental Material, Sec. VIII [63]).

To confirm the applicability of the remote control to general HOTTIs, we also investigate breathing Kagome lattice in a triangular-shaped nanostructure. We choose $t_1 = 4t_0$, where $t_0$ and $t_1$ represent the hopping of upward and downward triangles of the breathing Kagome lattice. As shown in Fig. 6(a), if we apply $M$ on the bottom-right one corner, the clockwise correspondence of corners will induce spin-down SP for the bottom-left corner and spin-up SP for the top corner, while the counterclockwise correspondence of corners induces spin-up SP for the bottom-left corner and spin-down SP for the top corner. As a result, the SPs at the bottom-left and top corners are canceled out due to the geometrical frustration [77]. To get spin-polarized corner states, we apply $M$ on two bottom corners, as shown in Fig. 6(b). Both left and right bottom corners will induce a spin-down SP at the top corner, leading to the opposite SP for the remote corner. The calculated SP for the bonding corner states with $\lambda_z = 0.5t_0$ on one or two corners are shown in Figs. 6(c) and 6(d), which is consistent with the schematic figures in Figs. 6(a) and 6(b). Then we apply $V$ on the bottom two corners. All the corners can have opposite SP in an appropriate regime of $V$, as shown in Fig. 6(e) for $V = 3.5t_0$, which is consistent with the results in the Kekulé lattice we discussed above. More detailed results for a breathing Kagome lattice are shown in the Supplemental Material, Sec. IX [63]. We conclude that the remote control of the corner states can be applied to different lattices with higher-order topology and different geometry shapes of the nanostructures.

Last but not least, we discuss the possible realizations and advantages of our model using the first-principles calculations. The $\gamma$-graphyne nanostructures [78,79] are known as the HOTI due to the different intracell C-C and intercell C–C≡C–C bonds [19]. We build a $4 \times 4$ rhombus-shaped two-dimensional nanostructure, as shown in Fig. 7(a). The calculation method can be found in the Supplemental Material, Sec. X [63]. Two corner states ($C_0$ and $C'_0$) within the edge gap are mainly distributed at the two 120° corners. To introduce $M$, we adsorb a Mn atom at the side of the bottom-right corner, which replaces two hydrogen adsorptions, as shown in Fig. 7(b). The system becomes spin polarized with a magnetic momentum of $0.94 \mu_B$. The bonding (antibonding) corner state $C_0$ ($C'_0$) split into $C_1$ and $C_2$ ($C_3$ and $C_4$), which distribute at the bottom-right and top-left corners (top-left and bottom-right corners), respectively. The Mn atom adsorbed on the bottom-right corner induces the sizable SP at both
FIG. 7. Material realization of the $\gamma$-graphyne. Discrete energy levels of (a) $4 \times 4 \gamma$-graphyne nanostructure (b) with Mn adsorption at the bottom-right corner (c) with Si dopant at the same corner, and the corresponding partial charge density for nonmagnetic (gray), spin-up (yellow), and spin-down (blue) corner states (a) C0 and C0′, (b) C1–C4, (c) and C1–C5. The spin polarization (SP) is calculated for the spin-polarized bonding and antibonding corner states in panels (b) and (c). Isovalue $= 0.001/\text{Å}$.

corners, as shown in the right panel of Fig. 7(b), consistent with what is depicted in Fig. 4 for $\lambda_z \neq 0$ and $V = 0$. The SP for bonding and antibonding corner states have opposite signs at both corners. Then, to mimic the addition of V in DFT, we replace one C atom with Si to lift the on-site potential for both spin-up and spin-down states at the bottom-right corner, as shown in Fig. 7(c). C3 becomes a bonding corner state localized at the top-left corner, and C1 becomes an antibonding corner state localized at the bottom-right corner, which is again comparable to the model results with $\lambda_z \neq 0$ and $V > V_c$ in Fig. 4. Spontaneously, a new spin-down corner state (C5) is generated, similar to C5 in Fig. 4, except the alternation by Si makes C5 higher than C3. Interestingly, the sign of SP is reversed for both corners by $V$, as shown in the right panel of Fig. 7(c), which shows similar manifestations as we discussed in Figs. 2(h) and 2(l).

To this end, we perceive that not only $\gamma$-graphyne but also any 2D HOTIs should manifest the topological features revealed in our model. The local magnetization can be applied by adding magnetic atoms near the corners, e.g., Mn, Cr, or Mo [80,81]; and the local electric potential can be easily controlled by the gate electrode [82,83] or STM tips [84–86] in experiments. For device applications, the voltage can be AC with a typical frequency of MHz to GHz. It is known that electrons in solids may adjust their distribution in a femtosecond timescale under external stimuli, much faster than the period of the AC. Therefore, the static corner-corner responses we discussed in this paper can be directly extended for AC case without the issue of waiting for oscillation spread. This is much different from HOTIs from metamaterials especially the acoustic or photonic lattice for which the frequency of excitation is comparable to the resonance frequency [25,37–42].

In conclusion, we demonstrate that the local magnetization applied on one corner of a HOTI nanoflake can induce a sizable SP at the other corners. Furthermore, manipulation of electric potential at the same corner may switch the sign of SP of both corners. Importantly, this remote control of topological corner states revealed from the TB model of Kekulé nanostructures is confirmed in a real material $\gamma$-graphyne through DFT calculations. The robust and remotely tunable spin-polarized corner states provide great advantages for diverse applications such as spin filtering, quantum gates, and information storage.

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