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Valley photonic molecular crystals

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Abstract

Photonics have been considered as versatile platforms for emulating condensed-matter physics, particularly topological phases, owing to the highly controllable band structures, directly observing the wave dynamics, and flexibly engineering topological transport beyond the limitations of natural materials. Molecular-crystal topological insulators have recently attracted growing interest as structurally programmable platforms, in which lattice geometry, orbital coupling, and molecular rotation can all be engineered by design. Inspired by this concept, we have introduced the molecular-rotational degree of freedom into valley photonic crystals to implement a valley photonic molecular crystal (VPMC). Because random molecular rotations break the periodicity of the VPMC, conventional band-based analysis of valley topology becomes inapplicable. To address this, we have proposed an effective topological indicator to characterize the valley topology and validate it by adopting a real-space Spectral Localizer operator. Furthermore, we have investigated the evolution of the valley topology under varying degrees of random molecular rotation and demonstrated that the valley topology would persist even under strong disorder. This result updates the conventional knowledge that valley-topological systems can only withstand weak and local disorder, and demonstrates their robustness to withstand strong and global disorder. This work would also be helpful to establish the VPMC as a programmable photonic platform for emulating molecular-crystal inspired topology and manipulating artificial gauge fields.

Keywords Topological insulators, Photonic crystals, Molecular crystals, Disorder systems

Introduction

Topological insulators (TIs) have inspired a paradigm shift in physics due to their topological bandgaps and robust edge states against disorder, and have been extended to other systems, such as photonic and phononic topological insulators [1–6]. Recently, molecular-crystal topological insulators have attained much attention as they represent a novel topological phase of matter and possess the potential for structural tunability, spintronics, valleytronics and flexible electronics. Currently, most organic topological insulators are realized through a class of molecular crystal known as metal–organic frameworks (MOFs) [7–18]. In MOFs,

metal atoms and their connecting organic molecules are situated at periodic lattice sites. Unlike inorganic crystals, the molecules in MOFs possess rotational degrees of freedom, which is responsible to the torsional potential energy [19–21]. It has been reported that such molecular rotations have significant impact on the adsorption, optical, and mechanical properties of MOFs [19–24]. However, the impact of molecular rotations on topological properties remains largely unexplored, as nonperiodic systems require more complex topological characterization and there are no well-defined conventional momentum-space invariants. Therefore, it is not only essential but also highly meaningful to investigate the influence of molecular rotation on the topological properties.

Photonic crystals feature band structures and topological properties analogous to electronic solids, while the photonic bands and wave functions can be readily engineered, so that they could serve as an ideal platform for emulating condensed-matter systems, particularly topological phases [25–29]. Inspired by the concept of

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molecular-crystal topological insulators, we have introduced rotational degree of freedom in to valley photonic crystals to implement a valley photonic molecular crystal (VPMC). In the VPMC, the molecules are localized at the periodic sites with random oriental angles, which is consistent with those in molecular crystals. Introducing random molecular rotations breaks the original periodicity, creating a disordered system, where conventional valley-topological analysis based on band theory is no longer directly applicable. To address this issue, we have developed an effective Hamiltonian model for the VPMC, proposed an effective topological indicator to characterize the corresponding valley topology, and further verified the valley-topological properties of the VPMC by adopting a real-space Spectral Localizer operator. Based on it, we have investigated the evolution of the valley topology of the VPMC under different degrees of rotational disorder. Theoretical modeling, numerical simulations, and experiments consistently demonstrate that the valley topology of the VPMC is preserved even under strong disorder. In addition, under strong disorder (random rotation ranges of 40° and 60°), we have observed a competition between valley-topologically protected edge states and Anderson-localized states.

This work has systematically investigated how molecular-rotational degrees of freedom influence the valley topology of the VPMC and the robustness against rotational disorder has been verified. The tolerance of the

VPMC to strong global disorder fully demonstrates the robustness of valley-topological systems and suggests its potential for realizing highly robust integrated photonic circuits. The results of this work are not only restricted to the molecule structure, but also are applicable to all C_3 -symmetric molecules. In addition, the molecular-rotation degree of freedom in the VPMC can serve as an additional tuning dimension, enabling programmable topological photonic systems and various forms of synthetic gauge fields, such as pseudo-electric and pseudo-magnetic fields. Beyond valley-topological systems, the proposed framework can also be extended to spin-Hall systems and may further inspire analogous studies in condensed-matter, acoustic, and cold-atom platforms.

Design of the photonic molecular valley crystal

The scheme of the periodic VPMC is shown in Fig. 1a, the molecule in the honeycomb-shaped unitcell is C_3 -symmetric and composed of six silicon (Si) rods with two different radii R_A and R_B . The structural parameters are defined as follows: the height of Si rods is $h = 1 \mu\text{m}$, the radii of two different Si rods are $R_A = 0.15a$, $R_B = 0.08a$, the distance between the centers of rods and the unit cell is $d = 0.6a/\sqrt{3}$, and the lattice constant is $a = 900 \text{ nm}$. Each molecule is embedded in air and can rotate freely around its center, defined by the orientational angle θ . Fig. 1b, and c are corresponding to two contrast configurations: a periodic VPMC, where all molecules share

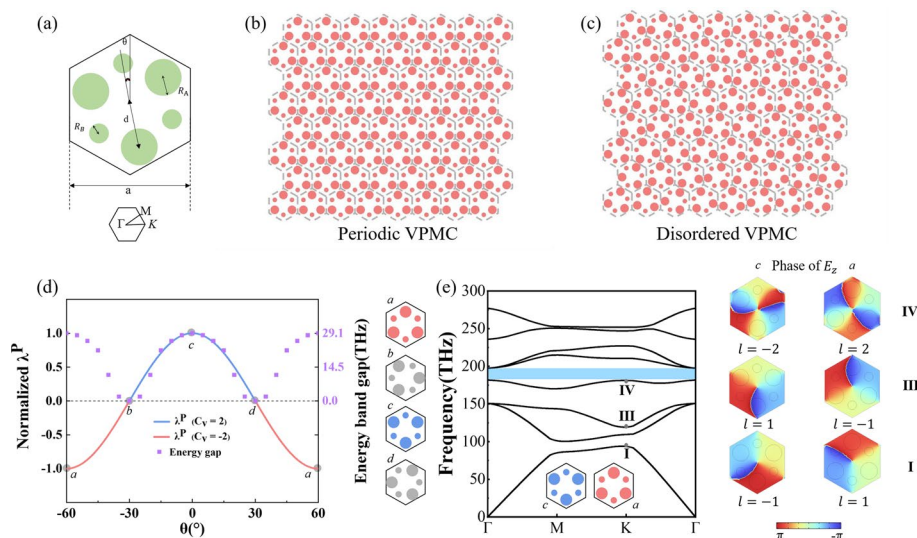


Fig. 1 Valley photonic molecular crystal. **a** Scheme of the unitcell and the first Brillouin zone of the periodic VPMC. **b**, **c** Schemes of the periodic and disordered VPMCs, the dashed gray hexagonal boxes mark the unitcells. **d** Phase diagram of the periodic VPMC. The purple solid squares are valley energy gaps obtained from finite element method (FEM) simulations; red (blue) lines are normalized λ^P with valley Chern number -2 (2). The unitcells of the VPMCs with $\theta = \pm 60^\circ$ (a), -30° (b), 0° (c) and 30° (d) are also provided in the right column. **e** Band structure of the periodic molecular crystal with $\theta = 0^\circ$ (blue) and 60° (red), a bandgap with $C_v = 2$ for $\theta = 0^\circ$ ($C_v = -2$ for $\theta = 60^\circ$) is marked by blue. At K valley, three modes (marked by gray dots) are with non-zero valley Chern number. Phases of E_z profiles of the three marked modes are shown on the right, the OAM charge of the modes are represented by l

a uniform orientation angle θ , and a disordered VPMC, where θ is randomly distributed.

In the periodic VPMC, the energy bands around the K and K' valleys can be described by the Dirac Hamiltonian:

$$\hat{H} = f_D + v_D(\hat{\sigma}_x \hat{\tau}_z \delta k_x + \hat{\sigma}_y \delta k_y) + \lambda^P \hat{\sigma}_z, \quad (1)$$

where $\hat{\sigma}_i$, $\hat{\tau}_i$ are the Pauli matrices acting on orbit and valley, and λ^P is the mass term, $|\lambda^P|$ and $\text{sgn}(\lambda^P)$ describe energy gap and valley Chern number (C_v) at K/K' valley, respectively [30–34]. Such Hamiltonian is analogous to the tight-binding one, which is previously adopted to describe molecular topological insulators [9, 24]. Interestingly, the effective mass term is related to the orientational angle θ :

$$\lambda^P \propto \cos(3\theta). \quad (2)$$

Detailed derivation of Eqs. (1) and (2) are provided in the Supplemental Material. A. The phase diagram with respect to θ is calculated by Eq. (2) and shown in Fig. 1d. The analytical results are quite consistent with the finite-element method (FEM) simulations of energy gap at K point (purple solid squares in Fig. 1d), validating our theoretical model. Surprisingly, the λ^P – θ relationship shown in Fig. 1d is highly consistent with the relation between the torsional potential energy and the orientation angle of the molecule in a C_3 -symmetric molecular crystal [21, 24]. Thus, it is validated to emulate molecular-rotation degree of freedom by introducing random twisting into the VPMC.

Moreover, the phase diagram of Fig. 1d could be divided to two regions: when $|\theta| < 30^\circ$, $\lambda^P > 0$, the valley Chern number is $C_v = 2$; when $|\theta| > 30^\circ$, $\lambda^P < 0$, $C_v = -2$. The energy bands and mode profiles of the periodic VPMC with $\theta = 0^\circ$ and 60° are shown in Fig. 1f. Although the energy bands are identical due to the equal $|\lambda^P|$, the phase profiles exhibit opposite orbital angular momentum (OAM) charges due to the opposite λ^P (Details in Supplemental Material. B, their valley-topological properties are shown in Supplemental Material. C). Such phase diagram highlights the significance of the molecular orientation angle θ in VPMC: it determines the system's effective mass term and the corresponding valley-topological properties. Therefore, it is imperative to investigate the influence of molecular orientation angles in quasi-periodic VPMCs with variable orientation angles.

Valley-topological properties in molecular crystals

To demonstrate of the pivotal role of molecular rotation, we have investigated the evolution of valley-topological properties by introducing varying degrees of random rotations into an initially periodic VPMC. The periodic

VPMCs with $\theta = 0^\circ$ and 60° are utilized as the initial systems, as they correspond to the valley-topological phases with the largest bandgaps (Fig. 1d). Random molecular rotations are introduced by applying a random twist angle θ_i to each lattice site i , where θ_i follows the uniform distribution:

$$\theta_i \sim U\left(-\frac{\theta_d}{2}, \frac{\theta_d}{2}\right). \quad (3)$$

Here, θ_d is the maximum range of the random molecular twist and represents the strength of disorders. Therefore, the VPMC with random twists could be described by two parameters: (θ_0, θ_d) . θ_0 is the initially uniform orientation angle of the period VPMC, and θ_d indicates the range of rotation angles of the molecules within the VPMC. To investigate the valley-topological properties of the VPMC, we construct a heterostructure interface by stitching together two types of VPMCs: $(0^\circ, \theta_d)$ and $(60^\circ, \theta_d)$. Fig. 2a, b shows the schemes of VPMCs of $(0^\circ, 0^\circ)|(60^\circ, 0^\circ)$ and $(0^\circ, 40^\circ)|(60^\circ, 40^\circ)$, respectively (the sign means the interface).

When random molecular twists are introduced, the periodicity of the system is broken, rendering band theory-based calculations invalid. To inspect the topological properties of this quasi-periodic system, we have employed two different methods: effective Hamiltonian approach and spectral localizer analysis. The effective Hamiltonian approach is as follows: since the lattice of the VPMC remains periodic, and the orientational angle of the molecule modify its mass term λ^P , the quasi-periodic VPMC could be approximately treated as a periodic crystal with an average effective mass term. The effective Hamiltonian could be formulated by averaging the λ^P of each unit cell:

$$\hat{H} = f_D + v_D(\hat{\sigma}_x \hat{\tau}_z \delta k_x + \hat{\sigma}_y \delta k_y) + \sum_{i=1}^N \frac{\lambda_i^P(\theta_0 + \theta_i)}{N} \hat{\sigma}_z, \quad (4)$$

where λ_i^P is the λ^P within each unitcell and determined by the corresponding oriental angle $\theta_0 + \theta_i$ [35]. The averaged efficient mass term is given by

$$\lambda_e^P = \sum_{i=1}^N \frac{\lambda_i^P(\theta_0 + \theta_i)}{N}. \quad (5)$$

$|\lambda_e^P|$ and $\text{sgn}(\lambda_e^P)$ describe the effective valley energy gap and valley Chern number, respectively. As a result, the effective valley Chern number of $\text{sgn}(\lambda_e^P)$ can serve as a topological invariant for characterizing the valley-topological properties of the VPMC. According to Eq. (3), the variation of λ_e^P with respect to θ_d is shown in Fig. 2c. When $\theta_d = 0^\circ$, $|\lambda_e^P|$ achieves the maximum and decreases as θ_d increases, while the $\text{sgn}(\lambda_e^P)$ and valley Chern

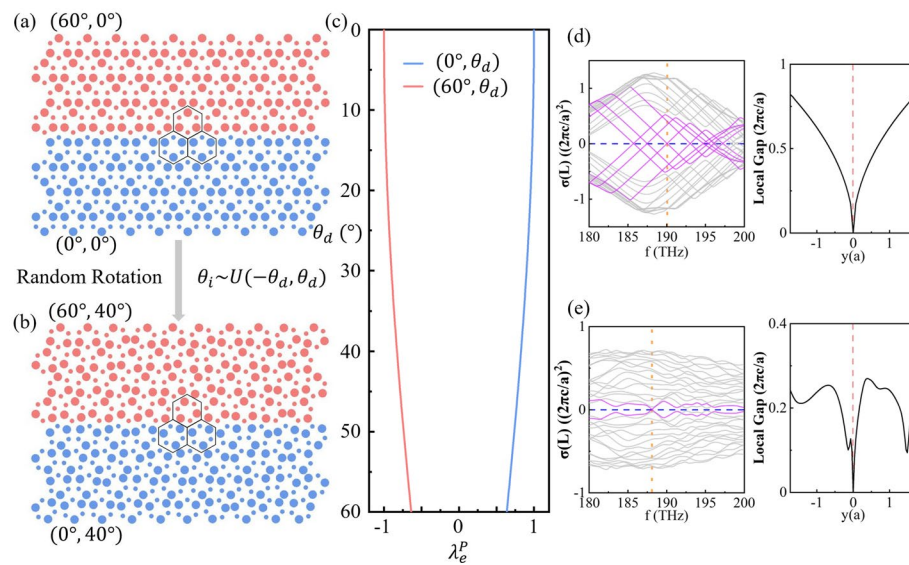


Fig. 2 Topological properties of the disordered VPMCs. **a, b** Scheme of the VPMCs with $(0^\circ, 0^\circ)|(60^\circ, 0^\circ)$ and $(0^\circ, 40^\circ)|(60^\circ, 40^\circ)$, respectively. **c** Evolution of the normalized λ_e^P with respect to the rotation range θ_d for the two VPMCs $(0^\circ, \theta_d)$ and $(60^\circ, \theta_d)$. **d, e** Spectral localizer results of the molecular crystals in **a** and **b**, respectively. Left: the spectrum of the spectral localizer, purple lines in the spectrum are the eigenvalues crossing zero. The frequency marked by the orange dashed line is selected to calculate the local gap. Right: the local gaps of the spectral localizer at different y positions, and the red dashed line marks the interfaces

number of the VPMCs $(0^\circ, \theta_d)$ and $(60^\circ, \theta_d)$ is remained. With the strongest disorder ($\theta_d = 60^\circ$), $|\lambda_e^P|$ remains more than 0, so that the bulk gap is still remained and topological edge states are maintained within the bulk gap.

The second method is to analyze the presence of topological edge states by using the spectral localizer. Spectral localizer is a real-space operator that can diagnose the presence of topological edge states using local topological indicators, such as the local gap and local index [36–38]. It is particularly suitable for investigating the topological properties of disordered systems as it is defined in the real space. For the system shown in Fig. 2a, the spectrum of its spectral localizer at the interface is calculated and plotted in the left of Fig. 2d. In the frequency range of 180–200 THz, there are four sets of eigenvalues cross the zero line, as highlighted by the purple lines. The eigenvalues that cross the zero line indicating that there is topological edge state at the interface. Then, the local gap is utilized to analyze the localization of the edge state. At the frequency point, where the eigenvalue of the spectral localizer is zero (marked by orange dashed line), its local gap is shown in the right part of Fig. 2d. It could be observed that the local gap vanishes at the interface and is much more than zero at other y position. It is a clear sign that the topological edge state is tightly localized at the interface. After introducing random twist, the spectral localizer of the system shown in Fig. 2b is also calculated and plotted in Fig. 2e. It could be seen that in

the spectrum of the spectral localizer, there are still eigenvalues cross zero (purple dashed lines), indicating that topological states exist in the disordered system. Similar to the disorder-free system, the local gap at the interface is zero, while at other locations, it is larger than zero, proving the edge state is localized at the interface. Detailed calculation and more results are shown in Supplemental Material. D.

The above analysis has demonstrated that the disordered VPMC can still support valley-topologically protected edge states. In valley-topological systems, the suppression of intervalley scattering is also essential. In the VPMC, introducing random molecular rotations would break the C_3 symmetry, so that well-defined K and K' valleys no longer exist in momentum space. Accordingly, the effective Hamiltonian in Eq. (4) describes the band properties of effective K and K' valleys. As a result, intervalley scattering in the VPMC is hard to be directly extracted. Nevertheless, the propagation direction of valley-topologically protected edge states at the VPMC interface is locked to the effective valley of the VPMCs on both sides. Therefore, the presence or absence of intervalley scattering can be examined through the propagation behavior of the valley-topologically protected edge states. A detailed analysis is provided in the Supplemental Material. E. The results show that there is no appreciable inter-valley scattering in the VPMC within a disorder range of 60° .

Samples and experimental measurement

In the previous section, both the effective Hamiltonian and the analysis of spectral localizer demonstrate that the valley-topological properties of the VPMCs are preserved within the range of $\theta_d \leq 60^\circ$. In success, we have fabricated samples and conducted measurements to verify their topological properties. The structure of the samples is shown in Fig. 3a. Two VPMCs with $(0^\circ, \theta_d)$ and $(60^\circ, \theta_d)$ are arranged to form a Z-shaped interface, while the two ends of the interface are connected to straight waveguides and grating couplers (fabrication processes are shown in Supplemental Material. H). The interface is set in a Z-shape to verify the properties of the valley-topological edge state, demonstrating that the valley topology is protected by navigating through such sharp bends. Our experimental setup employs a tunable laser and a photodetector coupled via optical fibers to the two grating couplers. The transmission spectrum is obtained by normalizing the total measured power over the reference grating couplers to account the coupling losses (details in Supplemental Material. I).

The SEM photographs of the samples with $\theta_d = 0^\circ$ and $\theta_d = 40^\circ$ are shown in Fig. 3b, e, respectively. For the sample with $\theta_d = 0^\circ$, the two VPMCs are periodic. As a result, the Z-shaped interface is very clear. In addition, the simulated field profile of the edge state transmission is shown in Fig. (c). Here, the simulated field profiles are obtained from two-dimensional FEM simulations. This approach provides an effective approximation to the three-dimensional structure, substantially reducing the

computational cost while maintaining high in-plane simulation accuracy and yielding high-precision mode-field distributions. In this work, all simulated electric-field distributions presented in the main text and Supplemental Materials are calculated using such method. The valley-topologically protected edge states transmit along the Z-shaped interface, and is immune to the sharp bend. For the sample with $\theta_d = 40^\circ$, the VPMCs are highly disordered due to the large range of molecular twisting angles θ_d , and the Z-shaped interface cannot be clearly observed. However, the simulation result in Fig. 3f shows that valley-topologically protected edge states still exist at the Z-shaped interface of the two VPMCs. The edge state is tightly confined at the interface, without extending into the bulk, and there is no leakage of the mode at the corners. This result indicates that the topological edge states are well-preserved even under 40° of disorder. The experimental transmission spectra of the two systems show good agreement with our simulations, as shown in Fig. 3d, g, respectively. Here, the simulated transmission spectra are obtained from three-dimensional finite difference time domain (FDTD) calculations to yield broadband transmission data consistent with the experimental configuration. The simulated transmission spectra presented in the main text and Supplemental Materials are calculated by using such approach. The transmission peaks corresponding to the topological edge states are highlighted with colored boxes. A noticeable high transmission peak exists near the wavelength of 1570 nm in both the disorder-free and disordered cases,

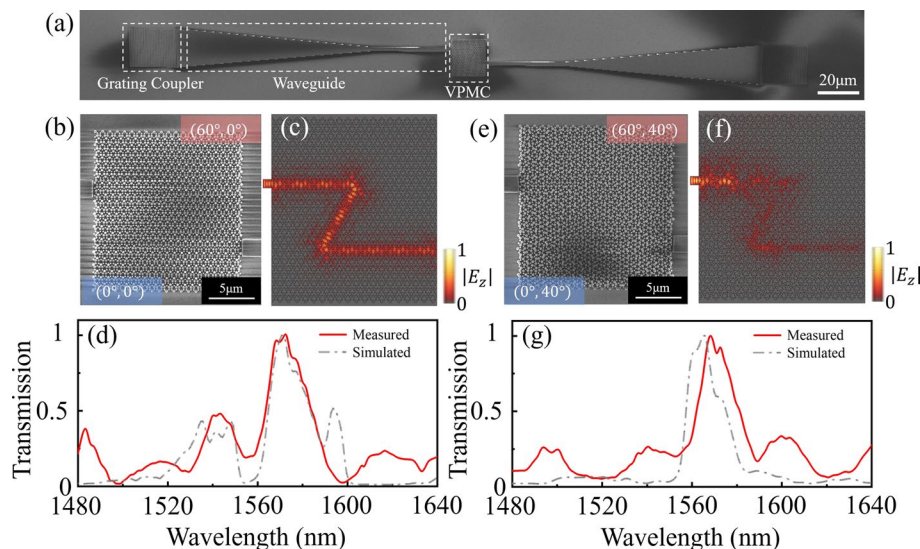


Fig. 3 Experimental setups. **a** Overview SEM image showing the overall structure of the sample. **b, e** SEM photo of the samples with $\theta_d = 0^\circ$ and 40° , respectively. **c, f** Simulated transmission profile of $|E_z|$ (by using FEM) at the wavelength 1570 nm with $\theta_d = 0^\circ$ and 40° , respectively. **d, g** Normalized simulated (gray dashed line, by using finite difference time domain (FDTD) method) and measured (red solid line) transmission spectra of the sample shown in **b** and **e**, respectively

which corresponds to the transmission of the valley-topologically protected edge state.

Evolution of the valley-topological properties in VPMCs

The valley-topological properties of the VPMC manifest in two aspects. First, the VPMC possesses a global bulk bandgap, as inter-valley scattering cannot be suppressed in a gapless system. Second, valley-topologically protected edge states emerge at the interface between two VPMCs with different valley Chern numbers. In this section, we would investigate the evolution of the bulk bandgap and the valley-topological edge states as the disorder strength increases, and demonstrate the impact of disorder on the VPMC, including its valley-topological edge states as well as disorder-induced Anderson localization states.

First, the phase diagram of the VPMC under different degrees of disorder θ_d is calculated by examining the bulk-mode transmission spectrum of the VPMCs under varying disorder strengths and plotted in Fig. 4a (Details in Supplemental Material. G). Each column in this diagram corresponds to the bulk transmission spectrum under a given disorder strength θ_d and the color represents the magnitude of the transmission. It could be seen that within the disorder range of $\theta_d \leq 60^\circ$, the transmission within the frequency range of 185–195 THz remains very low, indicating that the bulk bandgap is remained under the increasing disorder.

Then, to analyze the valley-topological edge states and Anderson localization states in the VPMCs, we consider the system in Fig. 2b, in which a straight interface

is formed by two VPMCs with $(0^\circ, \theta_d)$ and $(60^\circ, \theta_d)$. Such two VPMCs have opposite valley Chern numbers and valley-topological edge states would emerge at the interface. The energy spectra of these systems with $\theta_d = 0^\circ, 20^\circ, 30^\circ, 40^\circ$ and 60° are plotted in Fig. 4b. First, the energy spectrum of the $\theta_d = 0^\circ$ case could serve as a benchmark: it could be seen that, there are two kinds of modes in the spectrum: bulk modes (black circles) and edge modes (red stars). In the frequency range of 181–197 THz (marked by the purple box), there is no bulk modes, and this frequency range is corresponding to the bulk bandgap, while the frequencies of bulk modes are continuous outside the bulk bandgap. Exactly within the bulk bandgap, there are several edge modes localized at the interface, corresponding to the valley-topological edge states. With increasing disorder, the width of the bulk bandgap (purple boxes) decreases, but the phenomenon that edge states lie exactly within the bulk bandgap is maintained. The upper and lower frequency bounds of the bulk bandgap are obtained from Fig. 2b and plotted in Fig. 2a (the gray dots) to give a direct comparison with the bulk transmission diagram. It could be seen that these two results are well-matched, confirming the dynamics that bandgap gradually closes under increasing disorder.

Another important phenomenon is that Anderson localization states emerge in the VPMCs under relatively strong disorder ($\theta_d = 40^\circ$ and 60°). In the energy spectra of these two disordered VPMCs, sparsely distributed Anderson localization states (two Anderson localization states in total 150 eigenstates) lies both inside and outside the bulk bandgap. A representative mode profile of the Anderson localization state is provided in Fig. 2b, it

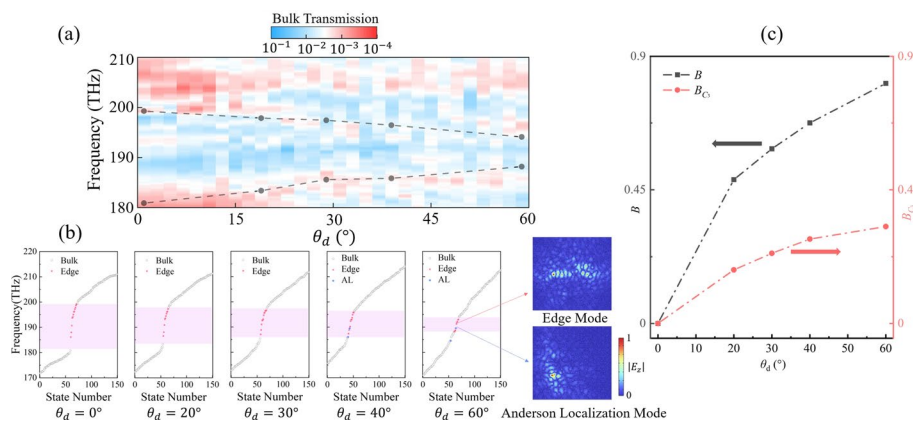


Fig. 4 Evolution of the valley-topological properties. **a** Simulated (by using FDTD method) bulk transmission diagram of the VPMC under different degrees of disorder. The bulk bandgaps of the VPMCs obtained from the energy spectra in **b** is also provided as the gray dots, and are connected by the dashed lines. **b** Energy spectra (by using FEM) of the VPMCs with $\theta_d = 0^\circ, 20^\circ, 30^\circ, 40^\circ$ and 60° , respectively. The bulk modes, edge modes and Anderson localization modes (AL modes) are marked by black circles, red stars and blue dots, respectively. For the VPMC with $\theta_d = 60^\circ$, mode profiles of an edge mode and an Anderson localization mode are also provided. **c**, Scatter plot of the parameter B (black) as well as B_{C_3} (red) for five selected values of $\theta_d = 0^\circ, 20^\circ, 30^\circ, 40^\circ$ and 60°

could be seen that this strong disorder induced Anderson localization state manifests as a random micro-cavity, and the spatial distribution of this mode is random, originating from the random relative permittivity distribution in the VPMCs [39–44]. A detailed analysis of the Anderson localization states is provided in the Supplemental Material. F. To quantitatively characterize the degree of disorder in the VPMCs, we define the parameter B to measure the disorder of the refractive-index distribution, and the parameter B_{C_3} to quantify the degree of C_3 -symmetry breaking in the system. For the VPMC with disorder degree of θ_d , its relative permittivity distribution is $\epsilon(\mathbf{r}, \theta_d)$, and B is defined as

$$B(\theta_d) = \frac{\|\epsilon(\mathbf{r}, \theta_d) - \epsilon(\mathbf{r}, 0^\circ)\|}{\|\epsilon(\mathbf{r}, 0^\circ)\|}, \quad (6)$$

in which $\|\cdot\|$ donates the L_2 norm. $B(\theta_d)$ represents the degree of disorder of the VPMC with θ_d compared with the ordered VPMC. A larger B indicates a higher degree of disorder in the system. Calculated $B(\theta_d)$ is plotted in Fig. 4c. As seen, B increases with θ_d , reaching 0.68 and 0.81 at $\theta_d = 40^\circ$ and 60° , respectively, which are already close to 1. This result indicates that the degree of disorder in the VPMC is already high at these disorder strengths, which also explains the emergence of Anderson-localized states under these two conditions. In addition to the overall degree of disorder in the system, the degree of C_3 -symmetry breaking B_{C_3} also merits attention, as the Dirac Hamiltonian and the valley-topological properties are closely related to C_3 symmetry. The definition of B_{C_3} is as follow: for the VPMC with θ_d , its C_3 -symmetric component $\epsilon_{C_3}(\theta_d)$ could be calculated by rotating the $\epsilon(\mathbf{r}, \theta_d)$ by 0° , 120° , and 240° , respectively, and then averaging them:

$$\epsilon_{C_3}(\mathbf{r}, \theta_d) = \frac{1}{3} \sum_{i=0}^2 \epsilon(R_{2\pi i/3} \mathbf{r}, \theta_d), \quad (7)$$

in which $R_{2\pi i/3}$ is the rotation operator with the rotation angle $2\pi i/3$. Then B_{C_3} could be calculated by

$$B_{C_3}(\theta_d) = \frac{\|\epsilon(\mathbf{r}, \theta_d) - \epsilon_{C_3}(\mathbf{r}, \theta_d)\|}{\|\epsilon(\mathbf{r}, \theta_d)\|}, \quad (8)$$

in which $\|\cdot\|$ donates the L_2 norm. Calculated $B_{C_3}(\theta_d)$ is also plotted in Fig. 4c. For the periodic VPMC ($\theta_d = 0^\circ$), B_{C_3} is zero as the C_3 symmetry is preserved. Then, B_{C_3} increases as θ_d increasing, and gradually approaches to the saturation when $\theta_d \geq 40^\circ$. Compared with the large B , B_{C_3} is much smaller. This is attributed to the fact that the lattice sites in the VPMC remain fixed, while only the molecules undergo random rotations, thereby preserving a certain degree of C_3 symmetry. The relatively

small value of B_{C_3} indicates that the C_3 -symmetry breaking in the system is not severe, which ensures that the disordered VPMC still retains its valley-topological properties and supports valley-topological edge states.

To validate the valley-topological properties of the VPMCs under different degrees of disorder, we have conducted extensive of simulations and experiments for $\theta_d = 0^\circ, 20^\circ, 30^\circ, 40^\circ$ and 60° , respectively. After introducing random twisting in VPMCs, the arrangements of the molecules are random. As a result, multiple samples with the same θ_d but different realizations are considered to demonstrate that the valley-topological properties of the VPMCs are universal and independent of specific realizations. Ten samples are considered in simulations and three samples are fabricated and experimentally measured for the four cases except $\theta_d = 0^\circ$.

The SEM photographs of the samples with different θ_d are shown in Fig. 5a. As θ_d increases, the disorder of the VPMCs gradually increases, and the periodicity of the VPMCs is gradually disrupted. Especially when $\theta_d = 40^\circ$ and 60° , the lattice points of VPMC and the Z-shaped interface are difficult to distinguish. The simulated transmission mode profiles of these samples are shown in Fig. 5b. It can be seen that when $\theta_d = 0^\circ$, the valley-topological edge state mode is periodic, corresponding to the periodicity of the VPMC. At $\theta_d = 20^\circ, 30^\circ$, and 40° , the valley-topologically protected edge states remain tightly bound at the Z-shaped interface, but the edge of the mode field is gradually blurred. This is caused by the irregular molecular arrangement on both sides of the interface. At $\theta_d = 60^\circ$, the topological edge state is still distributed along the edges of the Z-shaped interface. However, compared with the previous cases, the corresponding mode field is more diffused and blurred, and some localized states are present. The formation of localized states arises from the phenomenon of Anderson localization that occurs under strong disorder, reflecting the competition between topological edge states and Anderson localization, as mentioned above.

The simulated and measured transmission spectra are plotted in Fig. 5c, d (waterfall plots are shown in Supplemental Material. G and I). It can be observed that for all θ_d values, both the simulated and measured spectra show high transmission peaks in the wavelength range of 1560–1580 nm, which correspond to the transmission of valley-topological edge states, as mentioned in previous section. For different samples with the same θ_d , the transmission peaks of their topological edge states are generally similar, while the spectra outside the transmission peaks appear more randomness. It is due to the scattering of bulk modes caused by structural randomness. These simulation and experimental results collectively

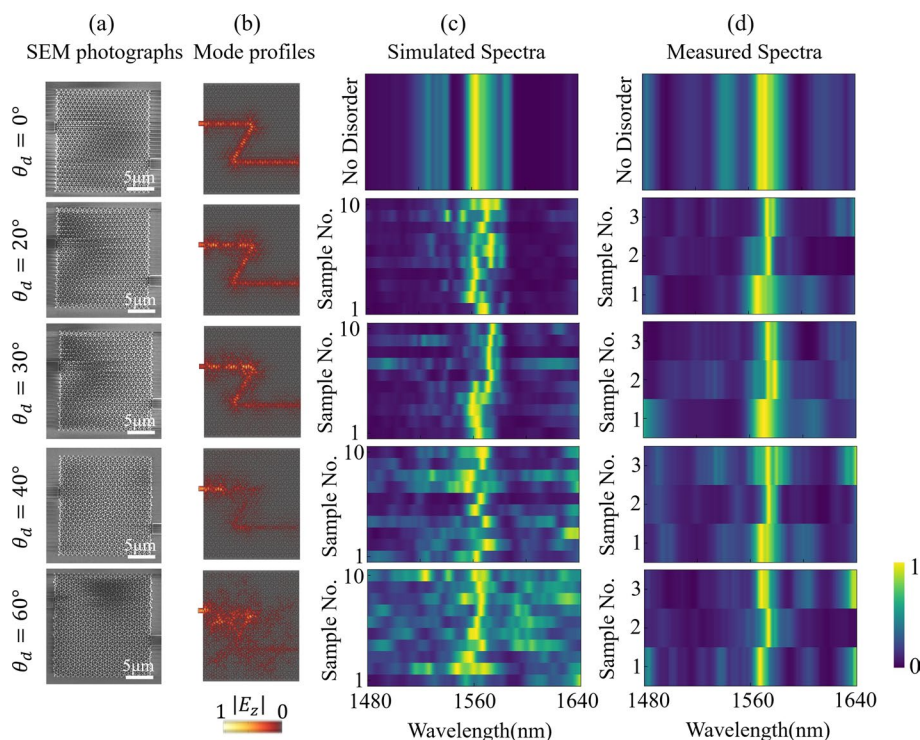


Fig. 5 Experimental demonstration of the VPMCs under different degrees of disorder. **a** SEM photographs of the samples with different degrees of disorder. **b**, Simulated (by using FEM) transmission mode profiles of the samples in **a**. **c**, **d**, Normalized simulated (by using FDTD method) and experimentally measured transmission spectra of the samples under different degrees of disorder. For **a**, **b**, **c** and **d**, from top to bottom, the corresponding disorder levels are: $\theta_d = 0^\circ, 20^\circ, 30^\circ, 40^\circ$ and 60°

demonstrate that the valley-topological properties of the VPMC are preserved within a disorder range of 60° .

Conclusion

In this work, we have proposed valley photonic molecular crystals (VPMCs) based on the valley photonic crystal on the silicon platform. By introducing different degrees of molecular-rotational disorder into the initially periodic VPMC, we have systematically investigated the influence of molecular rotation on the valley-topological properties of the VPMC. Using an effective Hamiltonian approach, we derived a valley-topological invariant for the VPMCs. Such invariant enables a simple and efficient evaluation of the effective valley Chern number of the VPMC, thereby addressing the difficulty of calculating topological invariants in VPMCs. We have also demonstrated the evolution of the valley-topological properties of the VPMC s under different degrees of disorder, revealing its robustness to withstand strong rotational disorder. In addition, the competition between valley-topological edge states and Anderson localization states is also discussed under strong rotational disorder. We believe that this work fills an important gap in the study of organic topological insulators. It may stimulate further investigations into organic

topological insulators and can be extended to a broader range of topological systems, such as condensed-matter systems, cold-atom systems, and acoustic systems.

Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1007/s44519-026-00017-3>.

Supplementary file 1.

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Author contributions

Zhe Li, Yongzhao Li and Xue Feng conceived the idea. Zhe Li designed and performed the calculations, simulations, experiments and data analysis. Yiheng Fan, Ziming Chen, Deyang Kong and Kaiyu Cui provided useful discussions and comments. Yiheng Fan contributed significantly to the calculation of the Spectral Localizer. Zhe Li and Xue Feng wrote this paper. Yongzhao Li and Yidong Huang revised this paper. The manuscript was written through contributions of all authors.

Data availability

The data in this study are available from the corresponding author upon reasonable request.

Declarations

Competing of interests

The authors declare no competing interest.

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References

1. Qi X-L, Zhang S-C (2011) Topological insulators and superconductors. *Rev Mod Phys* 83:1057–1110
2. Hasan MZ, Kane CL (2010) *Colloquium*: Topological insulators. *Rev Mod Phys* 82:3045–3067
3. Kane CL, Mele EJ (2005) Quantum Spin Hall Effect in Graphene. *Phys Rev Lett* 95:226801
4. Lu L, Joannopoulos JD, Soljačić M (2014) Topological photonics. *Nat Photonics* 8:821–829
5. Ozawa T et al (2019) Topological photonics. *Rev Mod Phys* 91:015006
6. Haldane FDM, Raghu S (2008) Possible realization of directional optical waveguides in photonic crystals with broken time-reversal symmetry. *Phys Rev Lett* 100:013904
7. Zhang LZ et al (2016) Intrinsic Two-dimensional organic topological insulators in metal-dicyanoanthracene lattices. *Nano Lett* 16:2072–2075
8. Wang ZF, Liu Z, Liu F (2013) Organic topological insulators in organometallic lattices. *Nat Commun* 4:1471
9. Sun H, Tan S, Feng M, Zhao J, Petek H (2018) Deconstruction of the electronic properties of a topological insulator with a two-dimensional noble metal-organic Honeycomb–Kagome band structure. *J Phys Chem C* 122:18659–18668
10. Osada T, Kiswandhi A (2020) Possible current-induced phenomena and domain control in an organic dirac fermion system with weak charge ordering. *J Phys Soc Jpn* 89:103701
11. Hu T, Zhong W, Zhang T, Wang W, Wang ZF (2023) Identifying topological corner states in two-dimensional metal-organic frameworks. *Nat Commun* 14:7092
12. Falsafi N, Abedinpour SH, Nazari F, Illas F (2025) Tuning topologically nontrivial states in the BHT-Ni metal-organic framework. *J Phys Chem C* 129:2556–2569
13. Shi X, Qu T, Wang M, Zhang L, Chen J (2024) Coexistence of chiral edge and localized states in a high Chern number topological photonic alloy. *ACS Photonics* 11:5180–5187
14. Zhang T, Su N, Hu T, Wang W, Wang Z (2024) Topological band engineering of one-dimensional π - d conjugated metal-organic frameworks. *J Am Chem Soc* 146:30539–30547
15. Yin Y, Gao Y, Zhang L, Zhang Y-Y, Du S (2024) Prediction of intrinsic two-dimensional topological insulators in Y3-/X3Y4-tetrahydroxybenzene metal-organic networks. *Sci China Mater* 67:1202–1208
16. Wang L et al (2025) Metal-organic frameworks: Strain-tunable quantum phase transition and high-order topological insulator and emergent fermions. *Phys Rev B* 111:035102
17. Li C et al (2025) Programmable higher-order topological phases in open-shell metal-organic frameworks. *J Am Chem Soc* 147:39662–39670
18. Drichko N, Thoi VS, Armitage NP (2026) Much ado about MOFs: metal-organic-frameworks as quantum materials. *Npj Quantum Mater* 11:24
19. Gonzalez-Nelson A et al (2021) Emergence of coupled rotor dynamics in metal-organic frameworks via tuned steric interactions. *J Am Chem Soc* 143:12053–12062
20. Gonzalez-Nelson A, Coudert F-X, Van Der Veen MA (2019) Rotational dynamics of linkers in metal-organic frameworks. *Nanomaterials* 9:330
21. Karlen SD et al (2010) Symmetry and dynamics of molecular rotors in amphidynamic molecular crystals. *Proc Natl Acad Sci* 107:14973–14977
22. Pizzanelli S et al (2020) A close view of the organic linker in a MOF: structural insights from a combined ^1H NMR relaxometry and computational investigation. *Phys Chem Chem Phys* 22:15222–15230
23. Meier D et al (2021) Rotation in an enantiospecific self-assembled array of molecular raffle wheels. *Angew Chem Int Ed* 60:26932–26938
24. Liu B et al (2023) Imaging the dynamic influence of functional groups on metal-organic frameworks. *Nat Commun* 14:4835
25. Dai T et al (2024) A programmable topological photonic chip. *Nat Mater* 23:928–936
26. Mazanov M et al (2024) Photonic molecule approach to multiorbital topology. *Nano Lett* 24:4595–4601
27. Liu G-G et al (2025) Photonic axion insulator. *Science* 387:162–166
28. Feng X, Wu T, Ge L, Feng L (2026) Co-propagating photonic topological interface states with hybridized pseudo-spins. *Nat Phys* 22:559–565
29. Dong Z, Chen X, Yuan L (2025) $\text{SL}(\mathbb{Z}, \mathbb{C})$ non-Abelian gauge fields in a photonic molecule array. *Nat Commun* 16:10166
30. Ma T, Shvets G (2016) All-Si valley-Hall photonic topological insulator. *New J Phys* 18:025012
31. Chen X-D, Zhao F-L, Chen M, Dong J-W (2017) Valley-contrasting physics in all-dielectric photonic crystals: orbital angular momentum and topological propagation. *Phys Rev B* 96:020202
32. Khanikaev AB et al (2013) Photonic topological insulators. *Nat Mater* 12:233–239
33. Borges-Silva D, Costa CH, Carvalho BR, Bezerra CG (2023) Pseudospin topological behavior and topological edge states in a two-dimensional photonic crystal composed of Si rods in a triangular lattice. *Phys Rev B* 107:075406
34. Yan B et al (2024) Multifrequency and Multimode Topological Waveguides in a Stampfli-triangle photonic crystal with large valley Chern numbers. *Laser Photonics Rev* 18:2300686
35. Chen X-D et al (2024) Realization of time-reversal invariant photonic topological Anderson insulators. *Phys Rev Lett* 133:133802
36. Cerjan A, Loring TA, Schulz-Baldes H (2024) Local Markers for Crystalline Topology. *Phys Rev Lett* 132:073803
37. Cerjan A, Loring TA (2022) An operator-based approach to topological photonics. *Nanophotonics* 11:4765–4780
38. Cerjan A, Loring TA (2022) Local invariants identify topology in metals and gapless systems. *Phys Rev B* 106:064109
39. Segev M, Silberberg Y, Christodoulides DN (2013) Anderson localization of light. *Nat Photonics* 7:197–204
40. Anderson PW (1958) Absence of Diffusion in Certain Random Lattices. *Phys Rev* 109:1492–1505
41. Zhou P et al (2020) Photonic amorphous topological insulator. *Light Sci Appl* 9:133
42. Rosiek CA et al (2023) Observation of strong backscattering in valley-Hall photonic topological interface modes. *Nat Photonics* 17:386–392
43. Karbasi S et al (2014) Image transport through a disordered optical fibre mediated by transverse Anderson localization. *Nat Commun* 5:3362
44. Groth CW, Wimmer M, Akhmerov AR, Tworzydło J, Beenakker CWJ (2009) Theory of the topological Anderson insulator. *Phys Rev Lett* 103:196805

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