Nanoscale topological corner states in nonlinear optics

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Topological states of light have received significant attention due to the existence of counter-intuitive nontrivial boundary effects originating from the bulk properties of optical systems. Such boundary states, having their origin in topological properties of the bulk, are protected from perturbations and defects, and they show promises for a wide range of applications in photonic circuitry. The bulk-boundary correspondence relates the N-dimensional bulk modes to (N-1)-dimensional boundary states. Recently, the bulk-boundary correspondence was generalized to higher-order effects such that an N-dimensional bulk defines its (N-M)-dimensional boundary states. Prominent examples are topological corner states of light in two-dimensional structures that have been realized at the micrometer-scale. Such corner states, due to their tight confinement in all directions, provide a novel route towards topological cavities. Here we bring the concept of topological corner states to the nanoscale for enhancing nonlinear optical processes. Specifically, we design topologically nontrivial hybrid metasurfaces with C6-symmetric honeycomb lattices supporting both edge and corner states. We report on direct observations of nanoscale topology-empowered localization of light in corner states revealed via a nonlinear imaging technique. Nanoscale topological corner states pave the way towards on-chip applications in compact classical and quantum nanophotonic devices.

INTRODUCTION

Topological phases of matter possessing quantized invariants have been a subject of intense studies in various fields ranging from condensed matter physics to photonics and acoustics, due to a plethora of novel physics and potential applications [1], [2]. Thus far, topological systems have been applied to lasing [6]–[9], quantum computing platforms [10]–[12] excitons [13], exciton-polaritons [14], communications, and robust signal transmission in various systems [15]. The higher-order bulk-boundary correspondence in topologically nontrivial structures, including topological corner states, has recently come into focus throughout various branches of physics including condensed matter [16]–[23], electrical systems [24], phononics and mechanics [25], [26], acoustics [27]–[30] and, microwaves [31]–[35] among others. In optics, topological corner states have been realized in arrays of waveguides [3], [5] and in coupled ring resonators [4]. However, both types of systems employ building blocks that are an order of magnitude larger than the wavelength of light. Such building blocks are unsuitable for applications in nanophotonics, where strong confinement of light at subwavelength scales is required for enhancing light-matter interactions. There have been growing efforts to bring topological photonics to the nanoscale. Particularly interesting candidates for this are nanostructures made of high-index dielectric materials with judiciously designed subwavelength resonant elements supporting both electric and magnetic Mie resonances. Without suffering absorption loss in contrast to their metallic counterparts, they are promising for implementations of the topological order for light, and have been successfully employed for the simplest forms of the bulk-boundary correspondence, such as 1D states in 2D structures [15], [36] and 0D states in 1D structures [37]–[39].
RESULTS AND DISCUSSIONS

Here we observe 0D corner states in a 2D nanophotonic topological insulator. In our system, nontrivial topology arises from the band-folding topological insulator geometry of periodic photonic structures [40]. The corner states further emerge in a cascaded process that can be conceptually illustrated in the momentum space. First, a bandgap is opened in a spectrum of 2D bulk modes. The bandgap is bridged by a pair of 1D topological edge modes. Second, a mini band-gap is opened in 1D modes due to the violation of C6 rotational symmetry at the boundary that can be described by the recently found fragile topology [41]. The mini-band gap hosts 0D corner states via the nontrivial fractional corner charge localization [42]. We realize this concept by using resonant dielectric metasurfaces. The metasurfaces exhibit a topological phase transition and band inversion above the light line, similar to an earlier theoretical proposal [2]. We design two arrays of silicon nanopillars hexamers — expanded and shrunken hexamers — as shown in Fig. 1a, characterized by distinct topological invariants. In this approach, the dielectric hexamers reside on a metallic mirror, creating a hybrid metal-dielectric structure. The metallic mirror, to a first approximation, doubles the effective height of the pillars for transverse magnetic (TM)-like modes while cutting off the first order transverse electric (TE)-like modes [35]. This design approach, while being compatible with standard nanofabrication technologies, leads to the creation of pure TM-like modes at frequencies of interests with only negligible contributions from TE-like and higher-order modes.

![Figure 1. Hybrid metasurfaces for nanoscale topological corner states of light.](image)

Topological corner states are hosted at the corners that are separating topologically dissimilar domains. Such corner localization of light in photonic systems can be associated with similar effects in solid-state physics where the nontrivial topology of the corner states was found recently to arise from the charge filling anomalies for electrons in crystals [42]. Specifically, at charge neutrality, the arrangements of electrons at the maximal Wyckoff positions in crystals inevitably break the intrinsic symmetries (C6 symmetry in our design), and extra electrons or holes have to be added onto corners of the crystal to restore the crystalline symmetry. However, this causes the charge neutrality to be broken, and introduces a new quadrupole moment term Q into the well-established corner charge expression: Qc=q_{c1}+q_{c2}, and the original equation is modified to Qc=Q+q_{c1}+q_{c2}, where q_{c1} and q_{c2} are the edge polarizations corresponding to the edges forming the corner and Qc is the total corner charge localization. This results in the 120-degree corner having topological protected e/2 fractional charge localization [42]. The 240-degree corner also naturally holds e/2 fractional charge of opposite sign because the 120-degree corner can be cut out from one infinite crystal with charge neutrality. The corner charge distribution for both the 120 and the 240-degree corner configurations is schematically illustrated by a ribbon configuration shown in Fig. 1c. We further note here that the discussed topological corner localization exists for corners formed by the domain walls with an armchair shape (from a perspective of the arrangement of individual pillars, otherwise a zig-zag shape from a perspective of the boundaries of unit cells. We will further use the perspective of individual pillars, and hence will refer to this type of domain wall as an “armchair” type).
Figure 2. Observation of bulk, edge, and corner topological states in metasurfaces. (a) False-color scanning electron microscopy image of the fabricated sample featuring a 240° corner between the two topologically dissimilar areas. (b) Close-up image of the corner with the two marked unit-cells featuring the shrunken and the expanded lattice designs. (c-e) Surface images of the nonlinear optical signal at the third harmonic wavelength measured for (c) 1730 nm, (d) 1590 nm, and (e) 1615 nm excitation wavelengths revealing the bulk mode, the edge state and the corner state correspondingly.

We proceed with full-wave 3D simulations with COMSOL Multiphysics with details provided in the Methods Section. The structure consists of Si cylinders 180 nm in diameter and 580 nm in height organized into hexamer clusters with a lattice constant of 1100 nm and shrunken/expanded coefficients of 0.764 and 1.12 correspondingly. The Si nanostructure rests on an Al mirror substrate.

We fabricate the designed structures with standard electron beam lithography followed by plasma etching (see an electron microscopy image in Fig. 1b and details in the Methods). To observe experimentally optical states in the metasurfaces, we employ a nonlinear imaging approach [6,7] (see details in the Methods). We note that the experimental characterization of topological corner states becomes more challenging at the nanoscale compared to its micrometer-scale counterparts. Nonlinear optical interactions in topological nanostructures provide unique opportunities to perform direct high-contrast visualizations of optical topological states. In our experiments, a short-pulse light of high peak power is illuminated onto a broad area of the sample to generate third-harmonic signals through the nonlinear interaction with Si hexamers. The wavelength of the illumination beam ranges within 1550-1750 nm spectral band, covering the region of the bulk modes, the edge and the corner states in the near-infrared. The corresponding nonlinear third-harmonic generation process converts the infrared radiation into a visible light at one third of the illumination wavelength between 517 and 583 nm. Importantly, visible light generated at third harmonic frequencies scatters freely into the far-field carrying a background-free information about the optical states at the illumination frequencies in the infrared. Since the third harmonic intensity is proportional to a third power of the illumination intensity, the third harmonic signal highlights areas of light localizations revealing the bulk modes, the edge and the corner states. In addition, a shorter wavelength of the nonlinear signal enhances the resolution of the far-field imaging by bringing down the wavelength-dependent diffraction limit.
Figure 3. Theoretical analysis of localizations of light in topological corner states. (a-c) Energy spectra of the systems hosting three types of corner states. (a) Spectrum of a metasurface hosting a 120° angle between arm-chair domain walls featuring a mini-bandgap. (b) 240° angle between arm-chair domain walls. (c) 240° angle between zig-zag domain walls featuring no mini-bandgap. (d-f) Real-space distributions of light around the corners corresponding to red dots marked in a-c. Note that 120° (d) has odd mirror parity, while 240° (e) has even mirror parity.

In our system of shrunken and expanded hexamers the optical eigenstates are circularly polarized, as determined by the structure symmetry. Therefore, in our experiments we employ a right-circularly polarized incident beam to couple efficiently from the far-field to the edge states and to the corner states. We further scan the angles of incidence of the illumination beam within approximately a 40° cone to find experimentally incident wavevectors that are optimal for coupling to the edge and the corner states from the far-field.

We first consider a 240-degree corner formed by two armchair domain walls (see Figs. 2 a,b). Detection of the third-harmonic signal reveals 2D bulk modes (Fig. 2c), 1D edge states (Fig. 2d) and 0D corner states (Fig. 2e) for different wavelengths of excitation.

Next, we proceed with theoretical and experimental studies of an alternative 120-degree corner formed by the same armchair domain walls and contrast these experiments with a study of a 240-degree corner formed by an alternative, zig-zag, domain wall. Figures 3a-c show the spectra of the three types of corners retrieved from full-wave numerical simulations. The two topological corners formed by armchair domain walls feature mini-gaps in their spectra with a corner state positioned in the mid-gap. The third type of corner formed by the zig-zag domain walls does not exhibit a mini-gap in its spectrum (band diagram can be found in Supplementary information Figure S1). The gaplessness of the edge state on the zig-zag domain, is essentially a direct consequence of the 120 and 240-degree corner states. The edge Hamiltonian is a one-dimensional Hamiltonian with alternating-sign coupling coefficient whose gap size is determined by the difference in on-site energies. (Supplementary information Note 1). Figures 3d-f show numerically simulated real-space field distributions at around the three types of corners at frequencies marked correspondingly in Figs. 3a-c.

Figure 4 shows experimentally observed topological light localization at the 120 and 240-degree corners (Figs. 4 a,b) contrasted to the light distribution around a non-topological corner (Fig. 4c). The non-topological corner in Fig. 4c exhibits relatively low confinement of light that spreads over multiple unit cells in a random fashion. The light tends to be distributed additionally along the domain wall, as in this case the structure does not feature a mini-gap as can be seen...
in Fig. 3c. In a sharp contrast, both topologically protected corner states in Figs. 4a,b show tight light localizations in our experiments. The signal is largely confined within an area of just a single hexamer unit cell. In this scenario, no significant coupling to the edge states along the domain walls was detected, as expect for structures exhibiting a corner state in the centre of the mini-gap (as per Figs. 3a,b). We attribute this difference in light localizations between Figs. 4a,b and Fig. 4c to the topological protection of the mid-gap corner states.

Figure 4. Experimentally observed localization of light around three types of corners. Superposition of the scanning electron microscopy images of the sample (grayscale background) with the spatial distribution of optical signal at the third-harmonic frequency captured by a visible camera (false-color intensity colormap). (a) Light localization at around the topological mid-gap corner state at the 120° angle of the two arm-chair domain walls. (b) Topological mid-gap corner state at the 240° angle of the two arm-chair domain walls. (c) Localization of light around 240° armchair corner with no mini-bandgap in the spectrum. Precision of the alignment of the electron microscopy image with an optical camera image is determined by the diffraction limit of the far-field camera imaging.

In summary, we have observed experimentally the manifestation of photonic topological corner states in nonlinear nanophotonics, where strong localization of light in subwavelength corner states is realized in disorder-immune optical structures revealed via the generation of optical harmonics. We have observed pronounced third-harmonic generation signals from the topological corner states due to strong topology-induced confinement of light that enhances nonlinear frequency conversion. The topology-driven localization of light, when combined with optical nonlinearities of constituent materials, can boost substantially nonlinear effects and provide topological protection against certain classes of disorder. Nonlinear nanoscale topological states might find their applications in compact on-chip classical and quantum nanophotonic devices.

**MATERIALS AND METHODS**

**Theoretical analysis**

For numerical simulations, we use the finite-element-method solver in COMSOL Multiphysics in the frequency domain. The near-field distributions are simulated using the eigenmode solver in COMSOL Multiphysics. All calculations were realized for metasurfaces placed on a semi-infinite substrate.

The tight binding model is used to analyze the semi-analytical counter-part of the studied model. In the tight binding model, each silicon pillar was assumed to hold an s orbital wave function. By changing the on-site potential of three sites in the corner unit-cell. The corner states can be transferred throughout the entire band-gap, forming a gapless spectrum flow.

**Fabrication**

The fabrication process of the topological metasurfaces begins with the deposition of a layer of Al on Si substrate using an electron-beam evaporation (Temescal BJD-2000) followed by a deposition of hydrogenated amorphous Si film using
plasma-enhanced chemical vapor deposition (Oxford Plasmalab System 100). The thickness was measured using the ellipsometer JA Woollam M-2000D. For the next step, the electron beam resist layer ZEP 520A (2:1) anisole was spin-coated. Next, a metasurface pattern was defined via electron-beam lithography (Raith-150 EBL system), the area dose of the exposure was 140 μAs/cm². A hard Al mask was created via metal deposition and a lift-off process. Then the pattern was transferred onto the material using an inductively-coupled plasma reactive ion etching (Oxford Plasmalab System 100) with CHF3, SF6 and Ar gases, and a subsequent removal of the hard mask. Electron microscope images of the resulting sample were obtained using an FEI Verios scanning electron microscope.

Optical experiments

The sample was excited with the light of a tunable optical parametric amplifier MIROPA from Hotlight Systems pumped by a pulsed laser Ekspla Femtolux. The excitation regime used 4 ps pulses with 5 MHz repetition rate and 300 mW of average power. Lasing spectra were controlled with a spectrometer NIR-Quest by Ocean Optics. The spectrum of the nonlinear signal generated from the sample was measured by a spectrometer QE Pro by Ocean Optics confirming that the signal corresponds to a third-harmonic wavelength. Spatial distributions of light across the samples were imaged at the third harmonic frequency by a cooled camera (Trius-SX694 Starlight Xpress Ltd) paired with an infinity-corrected f=150 mm achromatic doublet lens from Thorlabs. To illuminate the sample, a collimated laser beam was narrowed by two lenses: a f=100 mm achromatic doublet from Thorlabs, and an objective f=150 mm achromatic doublet lens from Thorlabs. To illuminate the sample, a collimated laser beam was narrowed by two lenses: a f=100 mm achromatic doublet from Thorlabs, and an objective lens X100 0.7NA from Mitutoyo. The same objective lens was collecting the third-harmonic signal. A dichroic mirror was used to separate the pump beam and the backward-emitting third harmonic signal. The polarization of the pump beam was controlled after the dichroic mirror to avoid its possible influence on the polarization state. In reflection, the imaging system residuals of the pump beam were filtered with an FGS900 filter.

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The 60 and 300 degree corners with arm chair boundaries however has no fractional charge, however we are still able to find two localized corner modes. Fig. 2 c,g h show the spectrum and field distribution of the 60 degree corner. The 60 degree corner has trivial only corner states. The topological index can be computed by:

\[ Q = \frac{e}{3} (\#K_2^{(3)} - \#\Gamma_2^{(3)}) \]

in which \(\#K_2^{(3)}\) is the number of bands below the band gap, with \(C_3\) eigen value \(e^{i \frac{2\pi}{3}}\) at \(K(\Gamma)\) point, which is zero in tight binding model.

Figure S1. Experimental study of edge and corner localization of light in topological metasurfaces. (a) A close-up image of the studied domain wall corner. The images obtained with an electron microscope. False colors visualize two topologically dissimilar domains. (b) Enlarged field of view of the corresponding samples. (c,d) Third-harmonic optical signal measured from the corresponding sample areas featuring (c) the edge state at the pump pulse wavelength centered around 1590 nm and (d) the corner localization at the pump pulse wavelength centered around 1610 nm.
We will demonstrate below that the gaplessness feature of the zig-zag edge states can be derived from the corner states of armchair edges. As is shown in Fig. S3(a) the zig-zag edge can be visualized as combinations of the 120 and 240 degree corners of armchair edges, shown as the A and B sites. N is the number of unit cells between the corner sites A and B. As is shown in Figure 3. (d-e) 120 and 240 degree corner states has opposite pairty according to their mirror planes. Hence around the corner states energy, the zig-zag edge can be modeled equivalent to a 1-D tight binding model shown in Fig. S3(b). Note that the hopping strength between adjacent atoms are alternating in signs, due to the parities of the corner modes. This tight binding model construct a 2-band Hamiltonian, whose gap size is only determined by the difference between the on-site energies $t_1$ and $t_2$, as is shown in Figure. S3(d). Note that in both tight binding model and the full wave simulations the differences of eigen energy of the two corners are diminishingly small, which guarantees the gaplessness of the two edge bands. Figure. S3(c) gives the band diagram with N equals 3. As is shown, the edge band diagram shows even higher similarities to the tight binding model in Figure. S3(d).

Figure. S3. Corner states induced band crossing for zig-zag edges. (a) Schematic of the zig-zag edge. (b) Effective 1-D tight binding model for the zig-zag edge Hamiltonian (c) Band diagram for zig-zag edge when the number of unit cells one arm N is 3. (d) Tight binding model’s band diagram with W equals 2 and different on-site potentials.
Figure S4. Full-wave theoretical calculations and analysis of corner localization of light in topological metasurfaces. (a) Energy spectrum of systems hosting a 300° angle between the two arm-chair domain walls featuring a mini-bandgap with a mid-gap corner state. (b,c) Real-space distribution of the two corner states marked in (a) as red dots Q1 and Q2 correspondingly.