Peer Review File

Manuscript Title: Topological Chern vectors in three-dimensional photonic crystals

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:

Referee #1 (Remarks to the Author):

In this manuscript, the authors study a photonic 3D Chern insulator in a gyromagnetic photonic crystal in the RF regime. The 3D Chern insulator is constructed by stacking layers of the 2D Haldane model. Due to the existence of Interlayer couplings, the topological phase transition between a 3D trivial insulator and a 3D topological insulator is now intermediated by a gapless Weyl phase. Both bulk and boundary properties are studied theoretically and experimentally. I find the topic very interesting and the paper is well-written. Both experimental and theoretical data are well presented. However, I still have some concerns about the novelty part. I wonder whether the current bulk hamiltonian can be continuously deformed to that of a trivial stacking, which has zero interlayer coupling. Thus, the phase with a nontrivial Chern vector here is more or less the same thing as those discussed in Ref. 15. Personally, I find the intermediate Weyl phase very interesting and may deserve more discussion, but that's a quite different way to build the whole story. Currently, I would recommend this paper be transferred to a more specific journal instead of Nature, but I'm very glad to hear about the opinions of other referees.

Here are some suggestions on technical parts,

In Fig. 1b, the authors use (ta = 2, tb = -1.2) for the tight-binding model. I wonder why ta and tb carry a different sign. I feel that they should carry the same sign since the layers are AA-stacked. I'm unconcern about this part since the mode profiles are not presented. I just wonder whether this is the cause of the difference between b) and e) since it will largely change the total interlayer coupling.
In Fig. 1b)e), does the range of B (-0.6,0.6) in panel e) match the range \phi (-pi,pi)? I guess not, so I would recommend clarifying how to map B to \phi clearly here.

Referee #2 (Remarks to the Author):

In the manuscript titled "Photonic realization of a three-dimensional Haldane modoel", the authors have fabricated a three-dimensional photonic lattice that harbors the so-called "three-dimensional Chern insulator phases". These states are characterized by three Chern numbers, and host chiral boundary modes at certain surface terminations. The authors indeed observed these topological modes, and tested their robustness to perturbations. The project has been well executed, with expected results, and the manuscript is well written. Therefore, this work should definitely be published in some form.

However, I am not convinced that this work should be published in Nature. On the one hand, this so-called “three-dimensional Chern insulator phases” are topologically equivalent to a straightforward stacking of two-dimensional Chern insulators. Although the authors argued that there is a fundamental difference that the surface states in their three-dimensional Chern insulator phases have kz dispersion, while those of a trivial stacking do not, this is merely a minor difference. In fact, we can easily add some small coupling between the stacking layers so that the surface states do have a small kz dispersion. Therefore, no fundamental difference exists between the stacking states the one observed here.

There is another important reason why I do not recommend this work to Nature. Even if the significance of “three-dimensional Chern phases” were advocated, the present photonic realization is not novel enough to be published in Nature, because the three-dimensional quantum Hall states have been published two years ago [F. Tang, et al, Three-dimensional quantum Hall effect and metal–insulator transition in ZrTe5, Nature 569, 537–541 (2019); Ref. 14 of this manuscipt]. These states are topologically equivalent to the photonic 3D Chern insulators.

To summarize, I believe that this work is well executed and well written, and should be published in some good journals. However, I do not believe that it is sufficiently novel and significant to warrant publication in Nature.

Referee #3 (Remarks to the Author):

It seems like topological photonics will never stop surprising with its demonstrations of increasingly exotic topological phases. In this work, the authors for the first-time design and experimentally realize a topological phase, 3D Chern insulator, which was predicted theoretically, but was never observed, and whose implementation demands symmetry reductions requiring the use magnetic materials for the time-reversal symmetry breaking, and a subtle design for controllable inversion symmetry breaking, all combined into a structure with isolated Weyl points to allow a complete topological bandgap.
From my point of view, the reported results represent another milestone in demonstrating both the power of photonics and the illusive topological phase which further broadens landscape of attainable topological materials. And a transition between three distinct 3D topological phases tuned by magnetic field, makes this work even more groundbreaking. I am confident that this work will be of significant interest to the broad readership of Nature, from condensed matter physics to photonics communities, and physics in general. The paper is overall very well written, and I have only two minor comments/suggestion on how the presentation could be improved. From my point of view the paper deserves publication in Nature.

Minor comments/suggestions
1) While from the general symmetry point of view the arguments are clear, the transition from the

tight-binding model to the photonic crystal is not smooth and not easy to understand. Specifically, the author should explain in more detail how the photonic model maps to their TBM. Some form of field distributions in resonators, overlap integrals between nearby cites, should be somehow connected to the TBM. Alternatively, the continuous limit could be used to mimic the TBM Hamiltonian near the high-symmetry points in the Brillouin zone, but the physics should be understandable from the electromagnetics point of view. See for example Nature Photonics 11, 130–136 (2017) and its supplement. I also believe that the last work, although reporting a different 3D topological phase, should be cited, along with the work reporting its experimental realization by some of the coauthors of the current paper [Nature 565, 622–626 (2019)].
2) Comparison between the subplots Fig 4. (a) and (b) raises some questions. Why, the diffraction in (b) is less pronounced than in (a) despite the longer propagation distance travelled by the surface wave? I would expect a more diffracted beam in (b) due to the longer propagation distance behind the metallic obstacles and scattering by the obstacles. Is it due to the negative refraction across the interfaces behind the obstacles? If this is the case, if would be good to illustrate this by numerical modelling and by an additional sublot showing the unfolded boundary behind the obstacle and the field distribution on it.

Author Rebuttals to Initial Comments:

Response Letter to Referees

We are grateful for the constructive comments on the manuscript (2021-12-19301) from three referees, which have guided us in significantly improving the paper.

In the text below, referee comments are quoted in *italics* and followed by our detailed response. We have also revised the manuscript based on the referee comments, and these updates are highlighted in blue and by a vertical red line in the left margin in those files. In the text below, the references to these updates are highlighted in a similar way.

GENERAL COMMENTS FROM REFEREE #1:

In this manuscript, the authors study a photonic 3D Chern insulator in a gyromagnetic photonic crystal in the RF regime. The 3D Chern insulator is constructed by stacking layers of the 2D Haldane model. Due to the existence of Interlayer couplings, the topological phase transition between a 3D trivial insulator and a 3D topological insulator is now intermediated by a gapless Weyl phase. Both bulk and boundary properties are studied theoretically and experimentally. I find the topic very interesting and the paper is well-written. Both experimental and theoretical data are well presented.

Response from Authors:

We thank the referee for considering our work "*very interesting*" and "*well-written*". In the following, we will address the referee's specific comments point-by-point.

SPECIFIC COMMENTS FROM REFEREE #1:

Referee #1 -- Comment 1:

However, I still have some concerns about the novelty part. I wonder whether the current bulk Hamiltonian can be continuously deformed to that of a trivial stacking, which has zero interlayer coupling. Thus, the phase with a nontrivial Chern vector here is more or less the same thing as those discussed in Ref. 15. Personally, I find the intermediate Weyl phase very interesting and may deserve more discussion, but that's a quite different way to build the whole story. Currently, I would recommend this paper be transferred to a more specific journal instead of Nature, but I'm very glad to hear about the opinions of other referees.

Response from Authors:

We thank the referee for this critical comment, and would like to address it from several angles.

We note that such an argument is applicable to other systems, such as a 1D Su-Schrieffer–Heeger (SSH) model whose properties can be understood by deforming it into a collection of decoupled molecules, as shown below in Fig. R1. Yet, individual molecules are 0D objects, and these lower-dimensional "building blocks" do not fully capture the topological physics of the full 1D SSH chain.

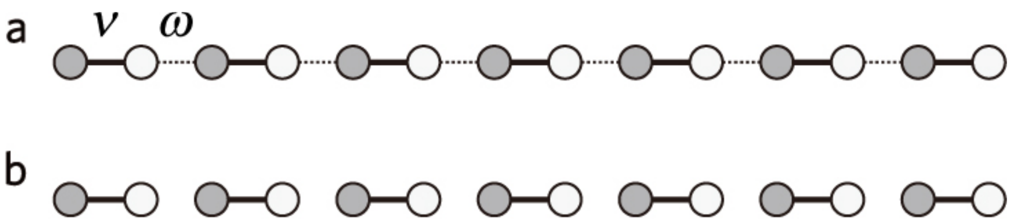

Fig. R1 | 1D SSH model. a, The 1D SSH chain with coupling $\omega < v$. **b,** The chain dissolves into individual molecules when $\omega = 0$.

Similarly, we can justify the novelty of our photonic 3D Chern insulator based on the following properties.

Property 1: The Chern vectors are directional

It is true that an isolated sample with a fixed Chern vector can be understood by deforming it into a stack of 2D systems with a scalar Chern number (for example, if the Chern vector is $\vec{C} = m\hat{z}$, we can deform the system into 2D sheets of scalar Chern number m stacked along z). However, there is no such simple interpretation when we consider interfaces between 3D samples with Chern vectors pointing in different directions.

In the revised manuscript, we have added a study of the surface states between two samples with *perpendicular* Chern vectors, $\vec{C}_1 = 2\hat{z}$ and $\vec{C}_2 = 2\hat{x}$ (Fig. R2). The surface states form a (2, 2)-torus link, or Hopf link, in the surface Brillouin zone, as shown in Fig. R2b.

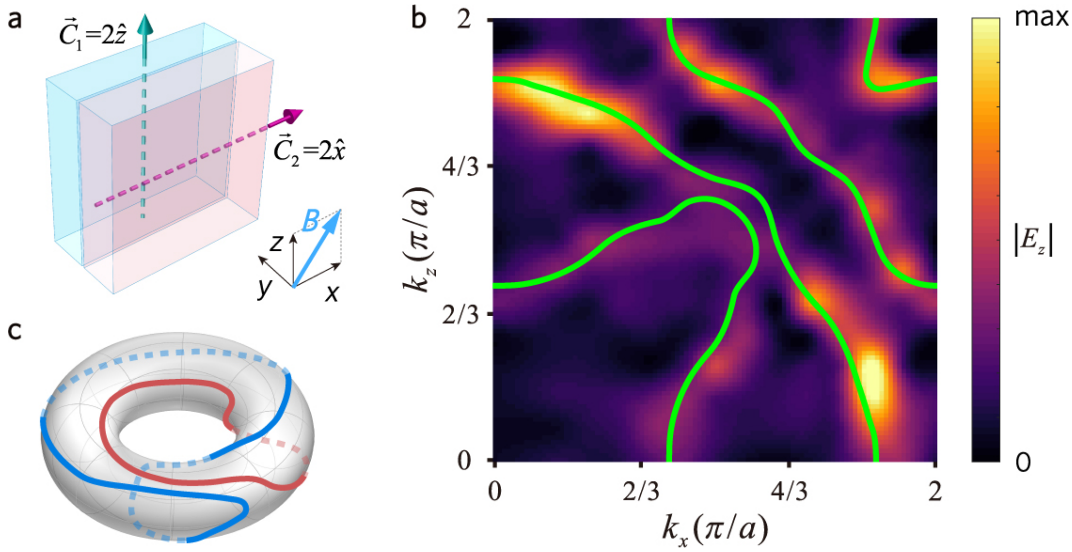

Fig. R2. | Hopf-link surface states from two perpendicular Chern vectors. a, Illustration of an interface between two photonic 3D Chern insulators with perpendicular Chern vectors $\vec{C}_1 = (0,0,2) = 2\hat{z}$ and $\vec{C}_2 = (2,0,0) = 2\hat{x}$, respectively. The magnetic field B is applied along $\hat{x} + \hat{z}$. **b,** Measured surface intensity at 19.6 GHz on the interface in **a**. The green lines indicate the simulated Fermi loop surface states. **c,** Simulated Fermi loops wrap around the surface BZ in a torus geometry and form a (2, 2)-torus link, or Hopf link. The blue and red colors distinguish the two components of the torus link.

To understand the origin of this Hopf link, we need (1) **the generalization of Chern-type bulk-boundary correspondence from 1D to 2D**, and (2) **torus knot theory**. Let us consider the general case with $\vec{C}_1 = m\hat{z}$ and $\vec{C}_2 = n\hat{x}$, which applies to two 3D Chern insulators with a common *x*-*z* interface. The first Chern vector $\vec{C}_1 = m\hat{z}$ induces m Fermi loops that wind along the median of the surface torus, while the second Chern vector $\vec{C}_2 = n\hat{x}$ induces n Fermi loops that wind along the longitude of the surface torus. According to knot theory, the combination of m and n forms a torus knot if m and n are coprime, and a torus link if m and n are not coprime (the number of components in the link is the greatest common divisor between m and n), as illustrated in Fig. R3.

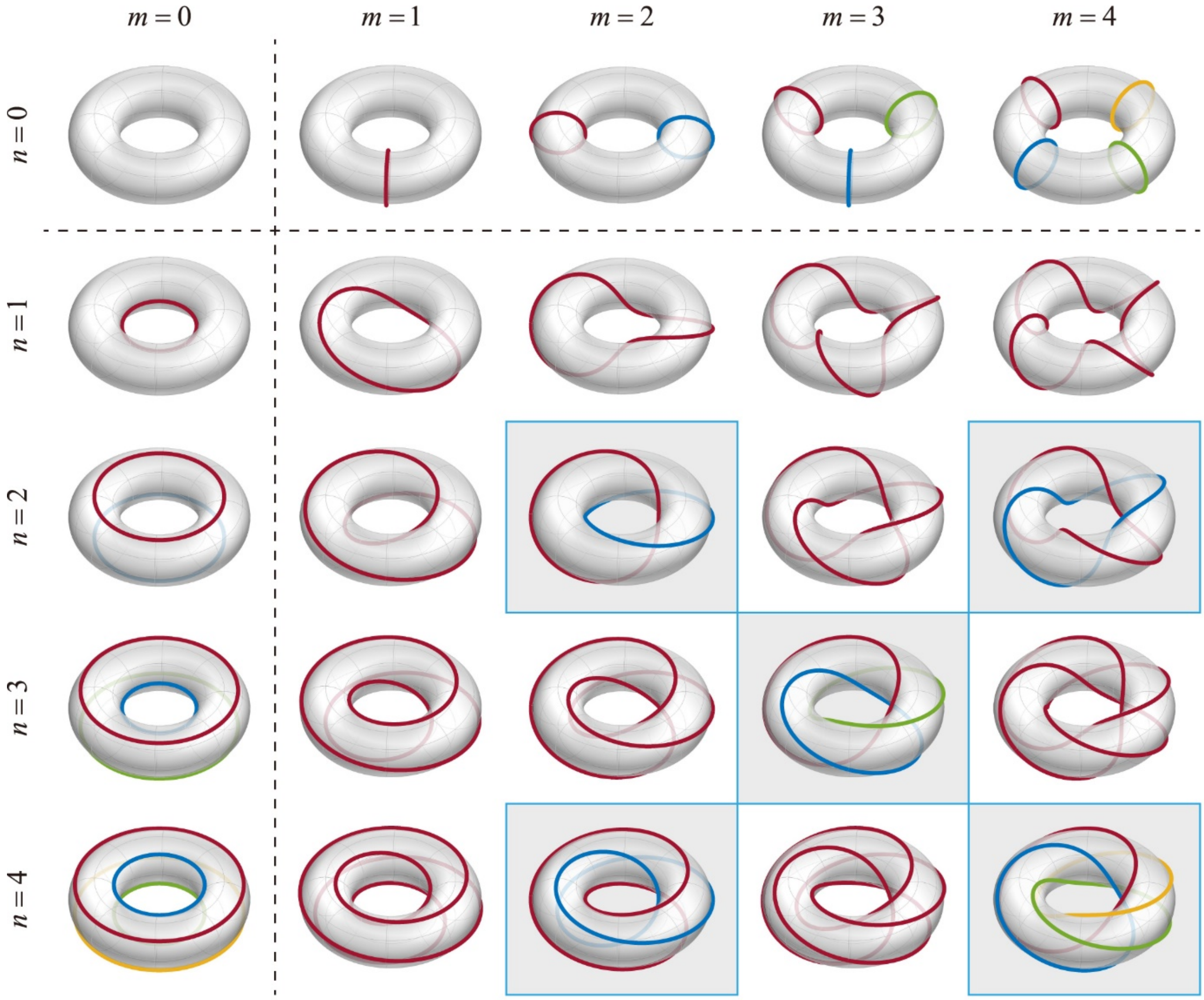

Fig. R3 | Summary of $T(m, n)$-torus knots/links with different combinations of m and n. The colors of red, blue, green and yellow represent the first, second, third and fourth loops that wrap around the torus surface without crossing. The links with non-coprime m and n are highlighted with grey background. The simplest link is the $T(2, 2)$-torus link, or the Hopf link on the torus surface.

To the best of our knowledge, this is the first time a $T(2, 2)$ torus link (or a Hopf link) is demonstrated for topological surface states. In other topological materials, the topology generally refers to the topology of the bulk bands, not the topology of loops in the surface BZ, so this is a novel instance of the application of topological ideas.

In the revised manuscript, Fig. R2 and Fig. R3 have been included as Fig. 4 and Extended Data Fig. 9, respectively. Some other related figures have been added as Extended Data Figs. 5e, 10, 11. The discussions of the generalization of Chern-type bulk-boundary correspondence from

1D to 2D and the surface torus/link have been included in part of the abstract (line 31-38), the introduction (line 59-62, 72-76), the main text (line 136-139, 165-186), the conclusion (line 187-194), the references (ref. 29), and the methods (line 369-389).

Property 2: The Chern vector magnitude is tunable

Even though each layer has a unit Chern number (similar to the 2D Chern insulator in the 1988 Haldane model), the stacking scheme allows us to form Chern vectors with various magnitudes. In fact, we can achieve a Chern vector component exceeding all previously realized "large Chern number" experiments done on 2D Chern insulators.

In a set of experiments newly described in the revised manuscript, we fix the Chern vector along the z axis, e.g., $\vec{C} = m\hat{z}$, and modulate the arrangement of the interlayer coupling holes to tune m. Specifically, we maintain the hole radii as $r_1 = 2$ mm and $r_2 = 1.3$ mm, but arrange the stacked layers in a staggered manner, as shown in Fig. R4a-e for unit cells constructed by 2, 3, 4, 5 and 6 layers, which correspond to the Chern vector component $C_z = 2, 3, 4, 5$ and 6, respectively. These values of C_z were experimentally verified by counting the number of Fermi loops, as shown in Fig. R4f-j. **Our demonstrated value of $C_z = 6$ is larger than all previously realized scalar Chern numbers in both gapped and gapless topological materials**.

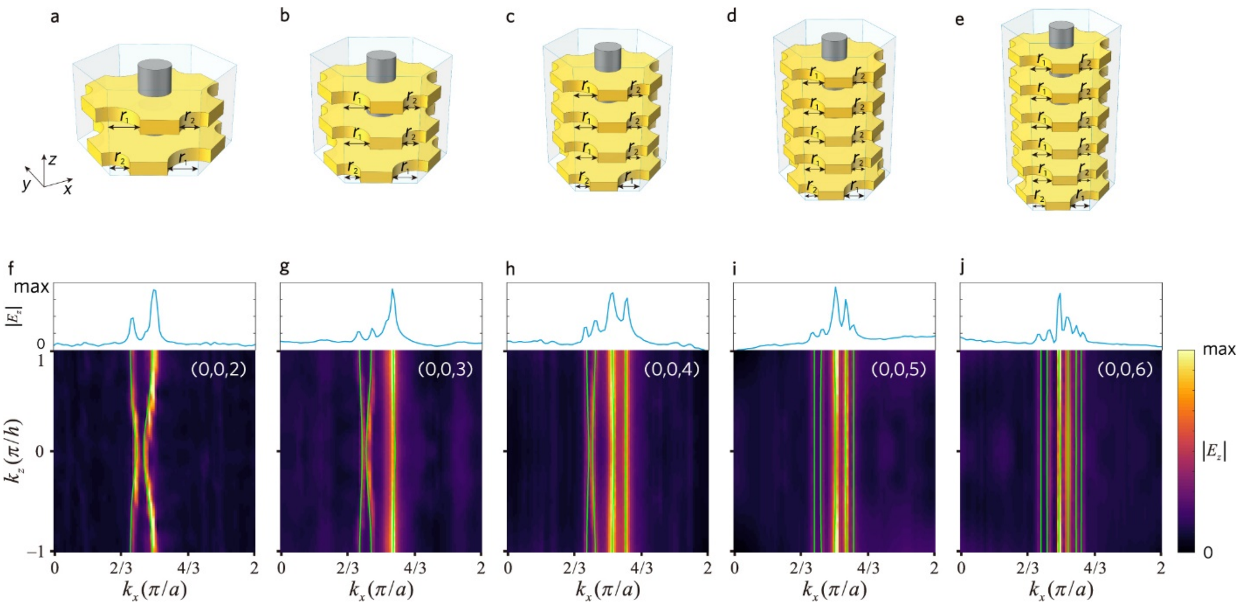

Fig. R4 | Photonic 3D Chern insulators with large Chern vectors. a-e, Unit cell of the photonic crystal with Chern vector (0,0,2), (0,0,3), (0,0,4), (0,0,5), and (0,0,6), respectively. The prorated holes have radii $r_1 = 2$ mm and $r_2 = 1.3$ mm. **f-j,** Lower panel: Measured surface intensity for biasing magnetic field 0.45 T at frequency 19.6 GHz. The green lines are the simulated Fermi loops. Upper panel: Measured surface intensity extracted from the lower panel at $k_z = \pi/h$, where h is the periodicity along z for each unit cell.

To our knowledge, in previous work, the largest Chern number found is 5 in gapped topological materials [*Nature* **588,** 419-423 (2020)], and 4 in gapless topological materials [*arXiv* 2203.10722 (2022)]. Since the Chern number is one of the most fundamental quantities in topological physics, our work is notable in this regard as the largest Chern number observed in real and artificial topological materials.

In the revised manuscript, Fig. R4 has been included as Fig. 3f-j. Some other related figures have been added as Extended Data Fig. 8f-j. The discussions of the tunable Chern vector magnitude have been included in part of the abstract (line 30-31), the introduction (line 70-72), and the main text (line 156-164).

Property 3: The 3D Chern insulator has phase transitions to Weyl phases

Topological semimetals and topological insulators, although conceptually connected, have never been demonstrated in the same platform. **Our work demonstrates the phase transition between 3D Chern insulating phases and Weyl semimetal phases—including a Weyl phase with a single Fermi arc, which had never been realized before** [*PRL* **107,** 127205 (2011)]. (A single Fermi arc is possible only in magnetic Weyl semimetals.)

Moreover, the choice of stacking scheme can further enrich the phase diagram. We have demonstrated two distinct Weyl phases, as shown in Fig. 3 in the revised manuscript, excerpted below as Fig. R5. We can see that the two Weyl phases can be selectively accessed with different magnetic fields. Both Weyl phases carry a single Fermi arc, but the latter one has an extra Fermi loop. **This kind of magnetic control over Fermi arcs has never previously been observed**.

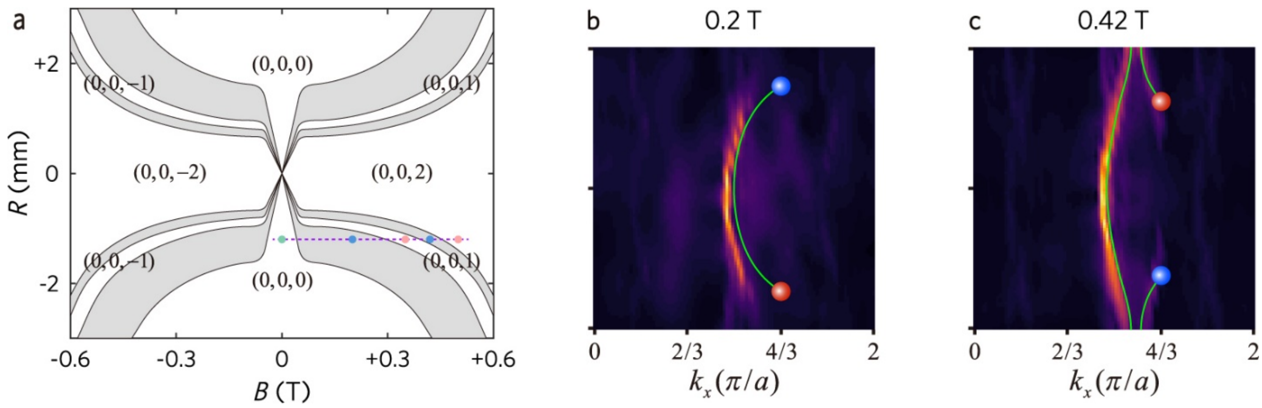

Fig. R5 | Phase transition of a photonic 3D Chern insulators with C_z = 2. a, Phase diagram. There are two Weyl phases (grey regions) and three insulating phases labeled by different Chern vectors. Increasing the magnetic field along the purple dashed line can access all five phases. **b,** A single Fermi arc at 0.2 Tesla in the first Weyl phase. **c,** Another single Fermi arc at 0.42 Tesla in the second Weyl phase; this Fermi arc is associated with a Fermi loop. The green lines indicate the simulated Fermi arc or Fermi loop surface states, while the red and blue dots are the projected WPs carrying opposite topological charges.

In the revised manuscript, Fig. R5 has been included as part of Fig. 3. Some other related figures have been added as Figs. 3a, 3c-e and Extended Data Figs. 2d-h, 5d, 8a-e. The discussions of the phase transition of a 3D Chern insulators with $C_z = 2$ have been included in part of the main text (line 140-155) and methods (line 305-306).

The above three properties go significant beyond the physics of decoupled 2D Chern insulators, and we hope this will convince the referee on the novelty of our 3D Chern insulator platform.

Referee #1 -- Comment 2:

Here are some suggestions on technical parts.
In Fig. 1b, the authors use (ta = 2, tb = -1.2) for the tight-binding model. I wonder why ta and tb carry a different sign. I feel that they should carry the same sign since the layers are AA-stacked. I'm unconcern about this part since the mode profiles are not presented. I just wonder

whether this is the cause of the difference between b) and e) since it will largely change the total interlayer coupling.
In Fig. 1b)e), does the range of B (-0.6,0.6) in panel e) match the range \phi (-pi,pi)? I guess not, so I would recommend clarifying how to map B to \phi clearly here.

Response from Authors:
We thank the referee for the suggestions.

We are sorry for the confusion caused by the sign of interlayer coupling in the tight-binding model. We originally set $t_a = 2$, $t_b = -1.2$ to emphasize t_a and t_b are different, but even when we set both to be positive (for instance, $t_a = 3$, $t_b = 0.5$), the phase diagram is still qualitatively the same, as shown in the comparison in Fig. R6 below.

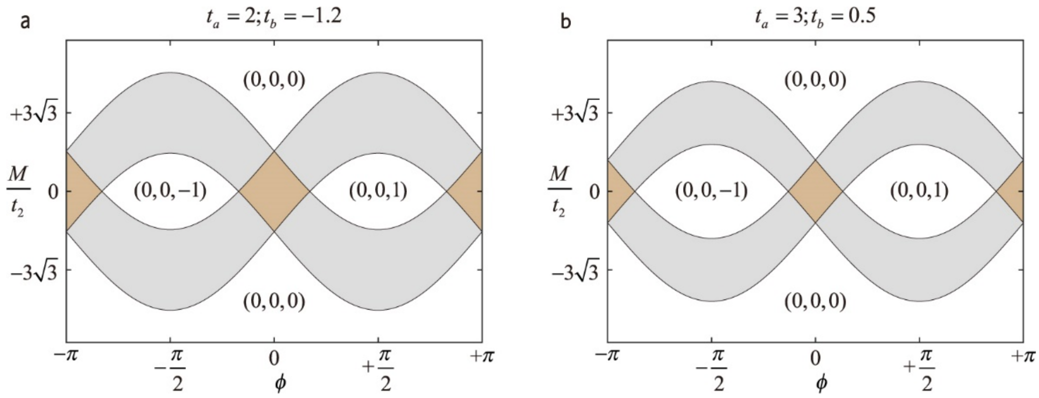

Fig. R6 | Comparison between positive and negative interlayer coupling. a, Phase diagram with $t_a = 2$, $t_b = -1.2$. **b,** Phase diagram with $t_a = 3$, $t_b = 0.5$.

The referee is certainly right that both t_a and t_b should have the same sign. To avoid potential confusion, we have replaced the phase diagram with the one with $t_a = 3$, $t_b = 0.5$ in the revised manuscript. Since the tight-binding model is only meant to serve as a qualitative guide to our actual photonic system, we have moved all discussions of the tight-binding models to Methods and Extended Data Fig. 3.

Regarding whether "*the range of B (-0.6,0.6) in panel e) match the range \phi (-pi,pi)*", we can confirm with the referee that the answer is no. B and ϕ are used to characterize the strength of T breaking in the photonic crystal and in the tight-binding model, respectively. In our studied magnetic fields range (0 T~0.6 T), the strength of T breaking increases when B increases. In a tight-binding model, however, the strength of T breaking increases when $0<\phi<\pi/2$, but decreases when $\pi/2<\phi<\pi$.

Our study is inspired by the tight-binding model, but ultimately the purpose of the tight-binding model is to help us interpret the topological phases and potential phase transitions. It is not meant to map to specific photonic structures with realistic parameters. This kind of qualitative model is common in both condensed matter and photonic systems; for example, it is nearly impossible to find out a clear mapping from the tight-binding model to a realistic photonic crystal. In the tight-binding model, when ϕ varies, all other parameters stay intact (even in the coupling $t_2\exp(i\phi)$, only its phase changes, but not its amplitude). However, in a realistic photonic crystal, when B varies, it will change all the couplings. This has imposed a fundamental difficulty in obtaining a one-to-one mapping between B and ϕ.

In view of the qualitative role of the tight-binding models, the title of the manuscript has been changed to "Photonic realization of 3D Chern insulators" to emphasize the topological phase, rather than the tight-binding model.

GENERAL COMMENTS FROM REFEREE #2:

In the manuscript titled "Photonic realization of a three-dimensional Haldane modoel", the authors have fabricated a three-dimensional photonic lattice that harbors the so-called "three-dimensional Chern insulator phases". These states are characterized by three Chern numbers, and host chiral boundary modes at certain surface terminations. The authors indeed observed these topological modes, and tested their robustness to perturbations. The project has been well executed, with expected results, and the manuscript is well written. Therefore, this work should definitely be published in some form.

Response from Authors:

We thank the referee for commenting that our work is "*well executed*," "*well written*," and "*should definitely be published in some form.*" We will address the referee's concerns in the following.

SPECIFIC COMMENTS FROM REFEREE #2:

Referee #2 -- Comment 1:

However, I am not convinced that this work should be published in Nature. On the one hand, this so-called "three-dimensional Chern insulator phases" are topologically equivalent to a straightforward stacking of two-dimensional Chern insulators. Although the authors argued that there is a fundamental difference that the surface states in their three-dimensional Chern insulator phases have kz dispersion, while those of a trivial stacking do not, this is merely a minor difference. In fact, we can easily add some small coupling between the stacking layers so that the surface states do have a small kz dispersion. Therefore, no fundamental difference exists between the stacking states the one observed here.

Response from Authors:

This comment from Referee #2 questions the fundamental difference between 3D Chern insulators and 2D Chern insulators. It is similar to Comment 1 from Referee #1.

We have explained in detail in response to Comment 1 from Referee #1. Here we provide a summary of the fundamental differences.

1) Our demonstrated 3D Chern insulator phases are characterized by Chern vectors. The vectorial nature of the Chern vector gives rise to distinct configurations of topological surface states described by knot theory, a relationship that has (to our knowledge) never previously been noted. We demonstrate the surface states induced by two *perpendicular* Chern vectors, which form a (2, 2)-torus link, or a Hopf link in the surface Brillouin zone.

2) Even though each layer has a unit Chern number (similar to the 2D Chern insulator in the 1988 Haldane model), different stacking schemes allow us to generate Chern vectors of different magnitude. We have used this to demonstrate Chern vector magnitudes of up to 6, which to our knowledge, exceeds the largest Chern numbers previously observed in experiments (5 in gapped [*Nature* **588,** 419-423 (2020)] and 4

in gapless [*arXiv* 2203.10722 (2022)] topological materials).

3) The 3D Chern insulator phase exhibits phase transition to Weyl semimetal phases, including a hypothesized one with a single Fermi arc [*PRL* **107,** 127205 (2011)]. The single Fermi arc is a fundamental difference between magnetic and non-magnetic Weyl semimetals, but has never been observed in any system. Our work has allowed a single Fermi arc to be observed in a real lattice for the first time. Furthermore, we have demonstrated magnetic control over Fermi arcs and Weyl points, for example, selective excitation of Fermi arcs. Such control has never been demonstrated in Weyl semimetals.

All the above properties cannot be understood from the physics of a 2D Chern insulator. We hope Referee #2 can agree on the novelty of our work in conclusively demonstrating the 3D Chern insulator phase characterized by Chern vectors.

Referee #2 -- Comment 2:
There is another important reason why I do not recommend this work to Nature. Even if the significance of "three-dimensional Chern phases" were advocated, the present photonic realization is not novel enough to be published in Nature, because the three-dimensional quantum Hall states have been published two years ago [F. Tang, et al, Three-dimensional quantum Hall effect and metal–insulator transition in ZrTe5, Nature 569, 537–541 (2019); Ref. 14 of this manuscipt]. These states are topologically equivalent to the photonic 3D Chern insulators.
To summarize, I believe that this work is well executed and well written, and should be published in some good journals. However, I do not believe that it is sufficiently novel and significant to warrant publication in Nature.

Response from Authors:
First, we note that the referee's comment above is equally applicable to 2D, specifically with respect to the difference between the 2D quantum Hall effect and the 2D Chern insulator (quantum anomalous Hall effect). The 2D quantum Hall effect arises from Landau quantization of electron gas, while the band topology in a Chern insulator is an intrinsic material property and does not involve Landau levels. (This difference was emphasized in Haldane's 1988 PRL, which proposed the first Chern insulator model: "Model for a Quantum Hall Effect without Landau Levels…" [*PRL* **61,** 2015-2018 (1988)].)

The 3D quantum Hall system, which, as the referee pointed out, was achieved in 2019 [F. Tang, et al. Three-dimensional quantum Hall effect and metal–insulator transition in ZrTe5, *Nature* **569,** 537–541 (2019)], relies on Landau level quantization (and also strong electron-electron interaction). By contrast, our 3D Chern insulator does not exhibit Landau levels but originates in the lattice (it is a "material property").

We have stressed this point explicitly in the revised manuscript from line 41, which reads as:

"Unlike the quantum Hall effect, which is based on Landau level quantization induced by a strong external magnetic field, the Haldane model describes how a Chern insulator can arise via time-reversal-symmetry (*T*) breaking in a crystal."

Second, regarding topology, we agree that both the 3D quantum Hall effect and the 3D Chern insulator can be characterized with a Chern vector. However, note that when the Chern vector is perpendicular to a surface, it does not induce topological surface states on that surface.

Therefore, when applying the bulk-boundary correspondence principle to this system, at most two components in the Chern vector are relevant. If one sticks to only one component of the Chern vector, the bulk-boundary correspondence, as both referee #1 and referee #2 have noticed, is "*more or less the same thing*" as in a 2D Chern insulator. To demonstrate the vectorial nature of the Chern vector, one needs to use two components in the Chern vector in bulk-boundary correspondence.

In the published 2019 Nature paper, the 3D quantum Hall effect is demonstrated, but only one component in the Chern vector plays a relevant role. Our work is the first to demonstrate the vectorial nature of the Chern vector. We observe topological surface states forming a torus link, induced by assigning perpendicular Chern vectors to the two bulk samples on either side of the interface. This demonstrates the significance of the vectorial nature of the Chern vector, generalizes the Chern-type bulk-boundary correspondence from 1D to 2D, and gives rise to novel topological features for the resulting surface states.

With the above justifications, we hope the referee will agree on the novelty and significance of our work.

<u>GENERAL COMMENTS FROM REFEREE #3:</u>

It seems like topological photonics will never stop surprising with its demonstrations of increasingly exotic topological phases. In this work, the authors for the first-time design and experimentally realize a topological phase, 3D Chern insulator, which was predicted theoretically, but was never observed, and whose implementation demands symmetry reductions requiring the use magnetic materials for the time-reversal symmetry breaking, and a subtle design for controllable inversion symmetry breaking, all combined into a structure with isolated Weyl points to allow a complete topological bandgap.
From my point of view, the reported results represent another milestone in demonstrating both the power of photonics and the illusive topological phase which further broadens landscape of attainable topological materials. And a transition between three distinct 3D topological phases tuned by magnetic field, makes this work even more groundbreaking. I am confident that this work will be of significant interest to the broad readership of Nature, from condensed matter physics to photonics communities, and physics in general. The paper is overall very well written, and I have only two minor comments/suggestion on how the presentation could be improved. From my point of view the paper deserves publication in Nature.

<u>Response from Authors:</u>

We thank the referee for his/her positive comments and favorable recommendation. In the following, we fully address the specific comments point-by-point.

<u>SPECIFIC COMMENTS FROM REFEREE #3:</u>

<u>Referee #3 -- Comment 1:</u>

While from the general symmetry point of view the arguments are clear, the transition from the tight-binding model to the photonic crystal is not smooth and not easy to understand. Specifically, the author should explain in more detail how the photonic model maps to their TBM. Some form of field distributions in resonators, overlap integrals between nearby cites, should be somehow connected to the TBM. Alternatively, the continuous limit could be used to mimic the TBM Hamiltonian near the high-symmetry points in the Brillouin zone, but the physics should be understandable from the electromagnetics point of view. See for example Nature Photonics 11, 130–136 (2017) and its supplement. I also believe that the last work, *although reporting a different 3D topological phase, should be cited, along with the work reporting its experimental realization by some of the coauthors of the current paper [Nature 565, 622–626 (2019)].*

Response from Authors:

We thank the referee for the suggestion. We have cited both papers [*Nat. Photonics* **11,** 130–136 (2017)] and [*Nature* **565,** 622–626 (2019)] as ref. 32 and 33 in the revised manuscript.

In our study, the tight-binding model is meant to serve as a qualitative guide to the possible topological phases and their phase transitions of the photonic system; it is not meant to map to specific photonic structures with realistic parameters. In [*Nat. Photonics* **11,** 130–136 (2017)], the tight-binding model is workable near certain points in the 2D Brillouin zone when bandgap opening is considered as a small perturbation, but not for the entire Brillouin zone. In our systems, however, the magnetic field varies in a wide range, and the Weyl points migrate through the 3D Brillouin zone. Such parameter variations cannot be considered small perturbations, so it is unreasonable to expect a direct mapping between the tight-binding results and full-wave simulations of realistic photonic structures.

In view of the qualitative role of the tight-binding model, we have moved it to methods part and Extended Data Fig. 3. The title of the manuscript has been changed to "Photonic realization of 3D Chern insulators" to emphasize the topological phase, rather than the tight-binding model.

Referee #3 -- Comment 2:

Comparison between the subplots Fig 4. (a) and (b) raises some questions. Why, the diffraction in (b) is less pronounced than in (a) despite the longer propagation distance travelled by the surface wave? I would expect a more diffracted beam in (b) due to the longer propagation distance behind the metallic obstacles and scattering by the obstacles. Is it due to the negative refraction across the interfaces behind the obstacles? If this is the case, if would be good to illustrate this by numerical modelling and by an additional sublot showing the unfolded boundary behind the obstacle and the field distribution on it.

Response from Authors:

We thank the referee for his/her careful reading of the paper. The diffraction of the surface waves comes from the dispersion of the chiral surface states along the *z*-axis. To exhibit the surface dispersion near the frontal (010) surface, we construct a supercell shown in Fig. R7a and plot its dispersion curve near 19.6 GHz related to k_z in Fig. R7b. The eigenstate plotted in Fig. R7c shows that waves are strongly localized near the frontal (010) surface and exhibit pronounced dispersion along the *z*-direction.

If we insert several copper pillars near the fontal (010) surface (see Fig. R7d), the waves are strongly localized near the periphery of copper pillars (Fig. R7f). Moreover, the eigenfrequency dispersion along the k_z is much weaker than the case in Fig. R7a-c.

In summary, even though the surface wave in Fig. 4b experiences a longer propagation distance than it in Fig. 4a, the waves are mainly confined at their individual layers when passing around the copper pillars, due to the weak dispersion along the *z*-axis.

In the revised manuscript, we have moved Fig. 4a, b to Extended Data Fig. 7, and added one sentence at its caption from line 517, which reads as:

"The surface waves are mainly confined at their individual layers when passing around the copper pillars due to the weak dispersion along the *z*-axis."

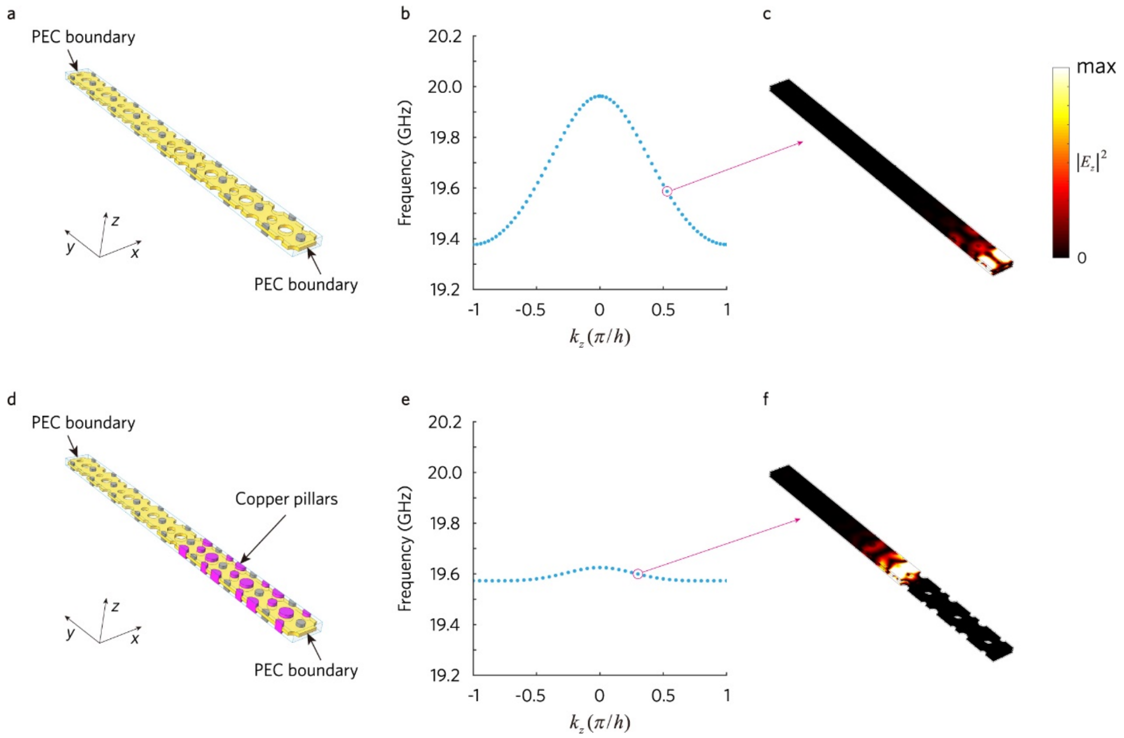

Fig. R7 | Surface dispersion without (a-c) and with (d-f) copper pillars. a, d, Supercell utilized to analyze surface dispersion. Two (010) boundaries are set as PECs and the other boundaries as periodic boundary conditions. **b, e,** Dispersion of eigenfrequency around 19.6 GHz for supercell in **a**, **d**, with $k_x = 1.1\ \pi/a$ and $0.665\ \pi/a$, respectively. **c, f,** Eigenstates for **b**, **e**, respectively.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

All my concerns have been convincingly addressed in the response. I think the current manuscript can meet the standard of Nature and I'm very glad to support the publishment of it.

Referee #2 (Remarks to the Author):

I apprecite the authors' effort to clarify the raised issues and revise their manuscript. The manuscript has been improved, and the work has been enriched. However, I am not convinced regarding my main criticism, namely that the states realized here are topologically equivalent to a straightforward stacking of 2D Chern insulators. Referee #1 also raised the same comment, and I do not think that this vital point has been satisfactorily addressed. The knot structure of the surface Fermi loop is an interesting feature but, as far as I can see, this is not an intrinsic feature of surfaces states of 3D Chern insulators. In fact, such knot loop can be found in purely two-dimensional band strucutres. It is not even a unique feature of topological surface states. If not phrased in terms of ``knot'', such ``knot states'' look quite ordinary in two-dimensional band structures. It is of course good to realize it at the interfaces between two 3D Chern insulators, but this does not demonstrates that there is essential difference between the realized states and a straightforward stacking of 2D states. Therefore, I still think that this work, while quite interesting, does not meet the high standard of Nature.

Referee #3 (Remarks to the Author):

The authors have fully answered my questions raised in the first review round. In addition, the manuscript was substantially revised to add new even more exciting results and to address all concerns of all reviewers. Some criticism from the other two reviewers was understandable as, indeed, the topological phase demonstrated in previous version represented a quasi-2D phase with the vector nature of the topological invariant being rather irrelevant. However, since this is the first experimental demonstration of 3D Chen phase in photonics, form my point of view, even that original demonstration was novel enough for Nature, despite somewhat similar (but not identical) phase previously demonstrated in a condensed matter system. The revised version has been enhanced significantly where now originality is no longer a question. Specifically, the authors demonstrate features which are unique to the 3D topological systems and employ differently oriented Chern vectors to form new types of topological boundaries, which has never been studies before experimentally, and this work reports the first observation of “Hopf link” surface states. I am confident that the paper reports truly original results which will be of interest to broad community readership, form solid state physics to photonics and acoustics. I therefore strongly recommend this paper for publication in its present form.

Author Rebuttals to First Revision:

Response Letter to Referees

We are grateful for the constructive comments on this manuscript (2021-12-19301A) from three referees. In the text below, referee comments are quoted in italics and followed by our response.

COMMENTS FROM REFEREE #1:

All my concerns have been convincingly addressed in the response. I think the current manuscript can meet the standard of Nature and I'm very glad to support the publishment of it.

Response from Authors:

We thank the referee for recommending the publication.

COMMENTS FROM REFEREE #2:

I apprecite the authors' effort to clarify the raised issues and revise their manuscript. The manuscript has been improved, and the work has been enriched. However, I am not convinced regarding my main criticism, namely that the states realized here are topologically equivalent to a straightforward stacking of 2D Chern insulators. Referee #1 also raised the same comment, and I do not think that this vital point has been satisfactorily addressed. The knot structure of the surface Fermi loop is an interesting feature but, as far as I can see, this is not an intrinsic feature of surfaces states of 3D Chern insulators. In fact, such knot loop can be found in purely two-dimensional band strucutres. It is not even a unique feature of topological surface states. If not phrased in terms of ``knot'', such ``knot states'' look quite ordinary in two-dimensional band structures. It is of course good to realize it at the interfaces between two 3D Chern insulators, but this does not demonstrates that there is essential difference between the realized states and a straightforward stacking of 2D states. Therefore, I still think that this work, while quite interesting, does not meet the high standard of Nature.

Response from Authors:

We thank the referee for the appreciation on our "*effort to clarify the raised issues and revise their manuscript*" and the comments of "*The manuscript has been improved, and the work has been enriched*" and "*this work… [is]… quite interesting*".

The Hopf link/knot structure of topological surface states is a previously unappreciated consequence of Chern vectors, and they are intrinsically tied to the vectorial nature of Chern vectors. Our work for the first time demonstrated a Hopf link for topological surface states, and thus demonstrated the vectorial nature of Chern vectors. We note that in a different context, a Hopf link can be constructed in the 3D Brillouin zone of a topological semimetal [as recently published in *Nature* 604, 647 (2022), when our work was in the second round review]. Our work is fundamentally different as it is in a topological insulating phase, and the demonstrated Hopf link exists in the surface Brillouin zone (thus a "torus link").

In terms of topology, the referee has pointed out in the last round of review that our demonstrated photonic states are topologically equivalent to the 3D quantum Hall states (as quoted in the following: "*the three-dimensional quantum Hall states have been published… these states are topologically equivalent to the photonic 3D Chern insulators.*") This was one of the two criticisms in the last round. We have explained in detail in the last response letter that, while both the 3D quantum Hall effect and our 3D photonic crystals can be characterized by the Chern vector, it is our work that uniquely demonstrates the vectorial nature of the Chern vector. The referee has accepted our explanation.

The referee still insisted on the other criticism that "*the states realized here are topologically equivalent to a straightforward stacking of 2D Chern insulators*." We would like to further clarify this issue in the following.

The key question here is not on the "*straightforward stacking*" itself (note that our demonstrations are much more complex than "*straightforward stacking*"), but whether the "*straightforward stacking*" has created a nontrivial topology that is different from the 2D Chern insulator phase characterized by the scalar Chern number. In other words, the question is: is the Chern vector $m\hat{z}$ topologically equivalent to the scalar Chern number m?

To answer this question, let us consider the experimental setup in Fig. 4a in the main text, which is duplicated below as Fig. R1 in this letter. We have demonstrated in the main text the topological surface states at the interface between two photonic crystals with perpendicular Chern vectors $\vec{C}_1 = 2\hat{z}$ and $\vec{C}_2 = 2\hat{x}$.

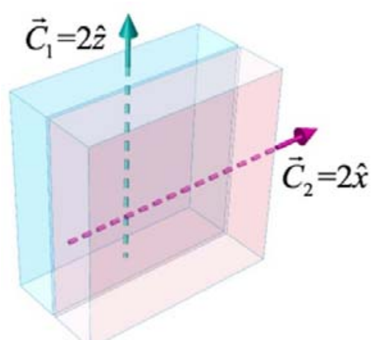

Fig. R1. Illustration of an interface between two photonic crystals with perpendicular Chern vectors $\vec{C}_1 = 2\hat{z}$ and $\vec{C}_2 = 2\hat{x}$.

Now, if the Chern vector $\vec{C}_1 = 2\hat{z}$ is topologically equivalent to the scalar Chern number 2, so shall be the Chern vector $\vec{C}_2 = 2\hat{x}$. Then, the two Chern vectors $\vec{C}_1 = 2\hat{z}$ and $\vec{C}_2 = 2\hat{x}$ shall be topologically equivalent. As a result, there shall be no topological surface states at the interface. However, this contradicts to our experimental demonstration. In fact, it is exactly the topological non-equivalence between $\vec{C}_1 = 2\hat{z}$ and $\vec{C}_2 = 2\hat{x}$ that gives rise to the demonstrated Hopf-link topological surface states, which, according to the comments of Referee #3, are "*more exciting*" and "*truly original*".

Therefore, we can conclude that the Chern vector $m\hat{z}$ is topologically non-equivalent to the scalar Chern number *m*. This is irrelevant to how the Chern vector is constructed, either by "*straightforward stacking*" or by any other approach.

We hope the above further explanations have resolved the concern of the referee.

COMMENTS FROM REFEREE #3:

The authors have fully answered my questions raised in the first review round. In addition, the manuscript was substantially revised to add new even more exciting results and to address all concerns of all reviewers. Some criticism from the other two reviewers was understandable as, indeed, the topological phase demonstrated in previous version represented a quasi-2D phase with the vector nature of the topological invariant being rather irrelevant. However, since this is the first experimental demonstration of 3D Chen phase in photonics, form my point of view, even that original demonstration was novel enough for Nature, despite somewhat similar (but not identical) phase previously demonstrated in a condensed matter system. The revised version has been enhanced significantly where now originality is no longer a question. Specifically, the authors demonstrate features which are unique to the 3D topological systems and employ differently oriented Chern vectors to form new types of topological boundaries, which has never been studies before experimentally, and this work reports the first observation of "Hopf link" surface states. I am confident that the paper reports truly original results which will be of interest to broad community readership, form solid state physics to photonics and acoustics. I therefore strongly recommend this paper for publication in its present form.

Response from Authors:

We thank the referee for recommending the publication.