nature portfolio

Peer Review File

Unidirectional Chiral Emission via Twisted Bi-layer Metasurfaces

Corresponding Author: Professor Lin Wu

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The article by Gromyko et. Al. presents experimental observations of unidirectional chiral emission from twisted bilayer silicon discs with integrated perovskite quantum dots (QDs). Using three degrees of freedom, the study investigates multidimensional control of chirality and directionality and reveals nearly uniform circular dichroism and significant differences in reflection in reciprocal space. The metasurfaces offer potential applications in various photonic applications by exploiting twisted configurations, changes in interlayer spacing, and lateral shifts.
The thesis represents a novel and significant contribution to the field of photonics and metasurface technology. The experimental results and methods are very good, and their relevance to Nature Communications is clear. I recommend that the manuscript be considered for publication with minor revisions to improve clarity and accessibility to a wider audience.
Suggestions for improvement
1 The work addresses a key challenge in modern photonics by demonstrating unidirectional chiral emission with high circular dichroism of QDs on metasurfaces. The authors use a well-known approach to tailor the reflectivity of bilayer silicon wafers, leading to a (chiral) change in QD emission. The authors substantiate their impressive results with the radiation properties of the resonant modes in reciprocal space. In addition, the manuscript is very well structured with appropriate sections, and the depth of supporting information leaves no doubt about the authors' understanding of physics. To better understand the reader, the authors should consider presenting chiral emission and unidirectional chiral emission side by side when presenting the results.
2. The use of technical terminology and explanations is appropriate for the intended audience, although some sections could benefit from further clarification to make them more accessible. The introduction is heavily enumerative and would benefit from a revision that interprets the results in the context of the literature.
3. The double alignment lithography is a precise but potentially complex and time-consuming technique. Scaling up this process for mass production could lead to issues in maintaining alignment accuracy and consistency over large areas. Electron beam lithography is used for metasurface patterning and is known for its high resolution but low throughput. A transition to higher throughput lithography techniques, such as photolithography or nanoimprint lithography, may be required to achieve commercial scalability. The use of materials such as silicon and fused silica in nanostructures is well established, but integrating these materials with active materials such as perovskite quantum dots at large scale could pose issues with the homogeneity and stability of the materials. The summary and outlook would benefit from a discussion of the aspect and challenge of scalability.

Reviewer #2

(Remarks to the Author)
The paper by Dmitrii Gromyko et. al. introduces a class of twisted bilayer photonic lattice structures that exhibit directional circular dichroism and chiral photoluminescence when coated with a layer of perovskite quantum dots. The authors include a comprehensive theoretical analysis of the corresponding photonic lattice resonance properties after varying the vertical layer separation and the lateral layer displacement. This analysis is then backed up by parametrically varied nanostructure fabrication followed by polarization and angle-resolved reflection spectra measurements. The study culminates in measurements of angular resolved photoluminescence, revealing that the emission occurs predominantly in a single quadrant in k-space and is dominated by LCP. The experimental results utilize doublet alignment lithography (DAL) for precise control over critical parameters such as twist angle, interlayer distance, and lateral displacement, leading to CD of

0.94. While polarization and direction selective light emission is important and the results are described clearly for the most part with well presented data, I'm not convinced of the novelty of resonant chiral emission from a metasurface. The authors claim that "addressing emission directionality remains unexplored", that existing designs "often retain certain mirror symmetries, resulting in symmetric polarization and intensity patterns of emission" and "experimental observations of directional chiral radiation from resonant metasurfaces with near-field-coupled emitters remain obscure". However, directional chiral photoluminescence from DCM coated 3D chiral metasurfaces (ref 31) and nonlinear chiral emission (third harmonic generation) from a 2D chiral silicon metasurface (ref 29) has been reported. Chiral resonances and thermal chiral emission have also been described theoretically in bilayer twisted high Q metasurfaces. While the platform introduced here certainly seems new, I don't see evidence that unidirectional chiral emission is new, especially compared to ref 31. If the phenomenon has been observed but the new platform improves performance either in brightness or polarization discrimination, this should be discussed, but I don't believe it has been. I therefore do not find the paper in its current form suitable for Nature Communications. To reach the bar for Nature Communications, the authors should explain clearly the benefits of their platform compared to that of Ref 31.

Detailed comments are also provided below.

1. In the introduction, particularly in the second paragraph, the authors should provide a brief review of the mechanism behind unidirectional resonant modes enhancing chiral emission, rather than merely mentioning extended structure and near-field emission enhancement. Additionally, there should be a connection from intrinsic to extrinsic chirality. A concluding sentence in the second paragraph should comprehensively summarize past results, highlighting the absence of research on intrinsic chirality and its application in emission control, to be introduced in the third paragraph.

2. In Figure 1(b), the unit cell of D2, which features a twisted angle and lateral shifts, significantly increases dimensions of the Fabry-Perot cavity. How is this unit cell defined in the simulation and how it changes the background Fabry-Perot resonance?

3. In line 180 and the following paragraph, although the authors explain and demonstrate in the nearfield regime, there is minimal environmental impact between the "open structure" and fully silica encapsulation, what is the fundamental experimental limitation in creating an "open structure"? Is it feasible to further evaporate silica after drop-casting the quantum dots (QD)?

4. Why do the dimensions shown in Figure 3, such as A1 and A2, slightly differ from those of the near-field and intermediate structures depicted in Figure 2? What is the possible reason for the increased left-handed circular polarization (LCP) with a lower resonance wavelength in the experiment compared to the simulation?

5. In line 218, what specific fabrication irregularities are being referred to? Please specify whether these irregularities occur within a single layer or between the two layers.

6. In line 248, how does lateral displacement affect the coupling model discussed in the design section?

7. In the Methods and Quantum Emitter Synthesis section, how did the authors ensure the uniform distribution of the $FAPbI_3$ QD solution after drop-casting, without aggregation on a specific spot on a disk with a notch?

Reviewer #3

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4

(Remarks to the Author)
In the manuscript "Unidirectional Chiral Emission via Twisted Bi-layer Metasurfaces"by D. Gromyko et al. the authors propose a bi-layer nanostructure consisting of dielectric discs that exhibits sharp resonances of high chirality. The reported chiroptical response of the structure originates from the resonant modes simultaneously excited in the upper and lower layers of the structure. In isolated single-layer metasurfaces, these modes exhibit linear far-field polarizations, however, when coupled within a bi-layer structure, they act as scatterers of elliptically polarized light. The authors provide a coupling model to describe the amplitude, phase and direction of the light scattered by both layers. I can recommend the publication of the manuscript in Nature Communications after several necessary revisions.
1) The authors claim that the resonance is unidirectional based on the radiative losses in the upper half-space evaluated for the resonance at different points in the reciprocal space. However, smaller radiative losses of the resonance may be associated with smaller energy losses and, hence, larger enhancement of the near field. Therefore, a resonance with smaller losses might produce emission with greater amplitude. The relation of the radiative losses and unidirectionality of the resonance should be clarified.
2) The chirality of the emission is demonstrated for the structures with displacement D1 and D2. It would be beneficial to add similar emission spectra for the structure without displacement.
3) The fabrication technique used in the manuscript is called "doublet alignment lithography". While the structure indeed

requires a single alignment operation between two layers, it remains unclear what the meaning of “doublet alignment” is. To which pair does the noun 'doublet' refer? It also would be beneficial to explain how this approach to alignment differs from the previously reported techniques such as https://doi.org/10.1038/ncomms14180, https://pubs.acs.org/doi/10.1021/acsphotonics.7b01460, https://pubs.acs.org/doi/10.1021/acsnano.0c07295.
4) The authors claim that the bi-layer design is essential for achieving chirality. However, the nanostructure comprising two layers of notched discs looks rather complex. Is it possible to achieve a similar performance in a structure with a simplified geometry?
5) It should be explained in more details if it possible to achieve this effect at larger separation distance.
6) For metasurface A2 the resonant peaks in reflectance are much smaller compared to those for metasurface A1. It is claimed that the nonradiative losses dominate over the radiative losses in structure A2. However, the authors use an objective with a significantly large NA=0.6, which should result in excitation of resonant modes in a broad area of the reciprocal space at nonzero incidence angles. This dilutes the contribution of the sharp quasi-BIC mode and normal incidence and decreases the amplitude of the resonant reflectance. The angular properties of the excitation beam should be clarified, experimental data of the reflectance spectra in dependence on the excitation beam aperture size are required.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The quality of the manuscript has been significantly improved by the review process. All questions and comments of the reviewers have been thoroughly and comprehensively addressed, and the suggested revisions have been successfully implemented. The manuscript now meets the high scientific standards and is recommended for publication.

Reviewer #2

(Remarks to the Author)
The authors have addressed my comments in detail and have clarified the novelty of their findings. I'm still not fully convinced that the chiral momentum space structure they claim provides unique opportunities for unidirectional chiral emission is fundamentally different from previous 3D chiral lattice resonances. However, I support publishing what I consider to be interesting and carefully examined measurements.

Reviewer #3

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4

(Remarks to the Author)
I agree with the authors' responses and I recommend the manuscript for publication in the Journal.

Response to reviewers for manuscript: NCOMMS-24-38824

We sincerely thank the reviewers for their constructive feedback, which has been invaluable in enhancing this manuscript. We have provided detailed responses to the reviewers' comments, along with a summary of all changes made. We believe that the revisions have significantly strengthened the manuscript and that it now meets the publication standards of Nature Communications.

Summary of changes (highlighted in the revised manuscript in yellow):

Main manuscript:

1. Page 1: Added affiliation for Zhaogang Dong: SUTD SMT and changed the email to zhaogang_dong@sutd.edu.sg
2. Pages 3/4: Revised Introduction according to the reviewers' suggestions.
3. Page 5: Modify Fig. 1 to display intrinsic chiral emission and unidirectional chiral emission side by side.
4. Page 6: Added a qualitative explanation of the coupling model in the design section
5. Page 8: Added a reference to the discussion of pure far-field coupling at larger separation distances.
6. Page 9: Clarified the origin of the fabrication irregularities.
7. Page 13: Added a paragraph to the Discussion section addressing the future aspects of large-scale nanofabrication, including commercial scalability.
8. Page 15: Revised the Fabrication section in Methods to provide a detailed account of the fabrication process for the "doublet alignment lithography."
9. Page 16: Added the data availability statement.

Supplemental information:

1. S5: Added Figure S5: Chiral emission from bi-layer metasurface A1 with perfect alignment.
2. S6: Added theoretical analysis of pure far-field coupling to explore the possibilities of achieving chirality at larger separation distances.
3. S7: Added the discussion of the radiation asymmetry in the up-down direction and its relation to the Q-factor.
4. S10: Added a SEM image of the metasurface with deposited QDs (Figure S19).

New References:

21 Hsu, C. W., Zhen, B., Stone, A. D., Joannopoulos, J. D. & Soljačić, M. Bound states in the continuum. *Nature Reviews Materials* **1**, 1-13 (2016).

32 Chen, W. *et al.* Uncovering Maximum Chirality in Resonant Nanostructures. *Nano Letters* **24**, 9643-9649 (2024).

52 Park, J.-S. *et al.* All-glass, large metalens at visible wavelength using deep-ultraviolet projection lithography. *Nano Letters* **19**, 8673-8682 (2019).

53 Kim, J. *et al.* Scalable manufacturing of high-index atomic layer–polymer hybrid metasurfaces for metaphotonics in the visible. *Nature Materials* **22**, 474-481 (2023).

54 Dong, Z. *et al.* Second-harmonic generation from sub-5 nm gaps by directed self-assembly of nanoparticles onto template-stripped gold substrates. *Nano Letters* **15**, 5976-5981 (2015).

55 Xu, J. *et al.* Multiphoton upconversion enhanced by deep subwavelength near-field confinement. *Nano Letters* **21**, 3044-3051 (2021).

Detailed point-to-point replies can be found below. We appreciate your help and look forward to hearing from you.

Yours sincerely,
Cheng-Wei Qiu, Zhaogang Dong, & Lin Wu
On behalf of all authors

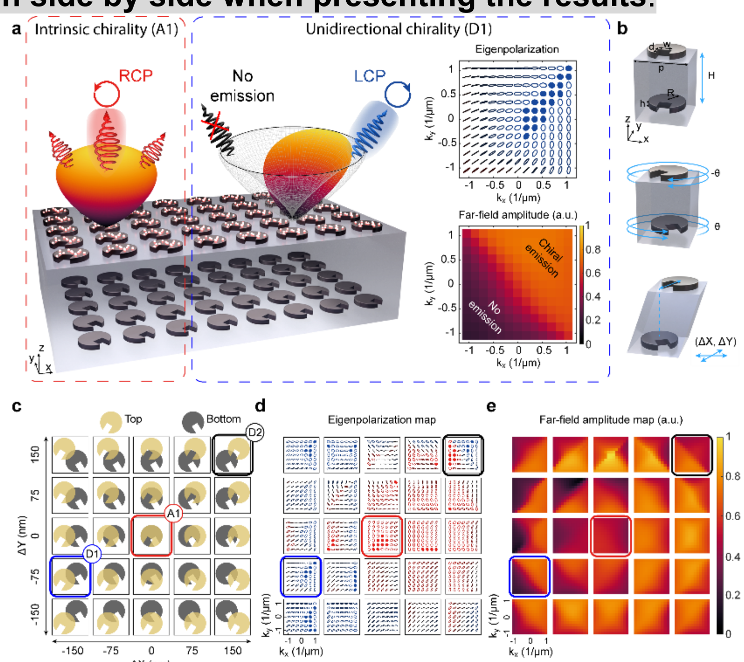
Reviewer #1:

The article by Gromyko et. Al. presents experimental observations of unidirectional chiral emission from twisted bilayer silicon discs with integrated perovskite quantum dots (QDs). Using three degrees of freedom, the study investigates multidimensional control of chirality and directionality and reveals nearly uniform circular dichroism and significant differences in reflection in reciprocal space. The metasurfaces offer potential applications in various photonic applications by exploiting twisted configurations, changes in interlayer spacing, and lateral shifts. The thesis represents a novel and significant contribution to the field of photonics and metasurface technology. The experimental results and methods are very good, and their relevance to Nature Communications is clear. I recommend that the manuscript be considered for publication with **minor revisions to improve clarity and accessibility to a wider audience**.

Response: We thank the Reviewer for the positive feedback on our work and for the detailed, constructive suggestions that will enhance the interpretation and presentation of our results, making them more accessible to the diverse audience of Nature Communications.

Q1 The work addresses a key challenge in modern photonics by demonstrating unidirectional chiral emission with high circular dichroism of QDs on metasurfaces. The authors use a well-known approach to tailor the reflectivity of bilayer silicon wafers, leading to a (chiral) change in QD emission. The authors substantiate their impressive results with the radiation properties of the resonant modes in reciprocal space. In addition, the manuscript is very well structured with appropriate sections, and the depth of supporting information leaves no doubt about the authors' understanding of physics. To better understand the reader, the authors should consider **presenting chiral emission and unidirectional chiral emission side by side when presenting the results**.

Response & Revisions: We thank the Reviewer for this valuable suggestion. To highlight the main results achieved in this work, we modified Fig. 1 (shown on the right) which now presents intrinsic chiral emission and unidirectional chiral emission side by side.



Q2 The use of technical terminology and explanations is appropriate for the intended audience, although some sections could benefit from **further clarification** to make them more accessible. The introduction is heavily enumerative and would benefit from a revision that interprets the results in the context of the literature.

Response: Thank you for your valuable suggestions for improvement.

Revisions:

(1) We have revised the 2nd paragraph of the **Introduction** to give a background of dielectric metasurfaces with either unidirectional resonant modes or chiral resonances.

> Dielectric metasurfaces with unidirectional resonant modes[17-20], which emit solely to one side, offer a promising solution for controlling radiation direction. Randomly polarized emitters distributed across the metasurface can couple to these resonant modes through near-field interactions, radiating according to the resonance's far-field properties. Unidirectional resonances can be derived from symmetry-protected bound-in-continuum (BIC) resonances[21] by breaking up-down symmetry[17,18]. This symmetry breaking splits the BIC resonance into two points of opposite chirality in momentum space. Further increasing the asymmetry merges these chirality points, thereby restricting resonant radiation to one side. However, the impact of unidirectional resonance polarization properties on the opposite side of the metasurface remains an unexplored area. Meanwhile, various resonant nanostructures with strong chiral effects at normal light incidence (*i.e.*, intrinsic chirality) have been explored, including multi-height metasurfaces[22-25], flat metasurfaces[26-32], slant-geometry metasurfaces[33,34], stacked and twisted bi-layer nanostructures[35-39], and chiral nanocavities[40,41]. Efforts have also been made to achieve absolute chirality at oblique light incidence (*i.e.*, extrinsic chirality) in flat metasurfaces with broken symmetry[42-44] or precise engineering of synthetic valleys in the folded Brillouin zone[45]. Nonetheless, these designs often retain certain mirror symmetries, resulting in symmetric polarization and intensity patterns of emission. In general, the transition from extrinsic to intrinsic chirality is achievable by tilting the walls of the structure which results in a continuous migration of the chirality points in the momentum space[33,34], similar to the recipe of unidirectionality. In most of these cases, chiral resonances in metasurfaces evolve from the achiral modes found in flat metasurfaces and, thus, are considered indivisible entities, missing one or several degrees of design freedom. On the contrary, stacked designs with vertical and lateral offsets naturally introduce a phase shift into the electromagnetic wave scattering problem, allowing for chirality[24] and directionality engineering[46]. Ultimately, while both unidirectional and chiral resonances were demonstrated, observations of unidirectional chiral remain obscure, likely due to the challenge of achieving multidimensional precise control.

(2) We have included a more comprehensive comparison of our metasurfaces with the previously reported metasurfaces.

> We term this effect "unidirectional chiral emission". The radiation pattern is influenced by a strong asymmetry in the up-down direction, which is inverted along the lateral directions. This results in off-normal chiral emission, distinct from the highly collimated chiral emission reported previously[33]. In that study, the emission is confined to the normal direction due to the rapid increase in the quasi-BIC Q-factor near the Γ-point. In contrast, our metasurfaces exhibit superior performance compared to other designs, which typically feature symmetrically positioned points of opposite chirality in momentum space and radiate over a broad solid angle[42-44]. This work suggests the potential for full multi-dimensional control of polarization response, enabling reconfigurable metasurface design via its integration with nano-electro-mechanical systems[47] or via scaling to the terahertz frequencies[48].

(3) We have incorporated a quantitative description of the coupling model into the theoretical analysis section in the Results.

> Tuning θ and H is crucial for achieving absolute intrinsic chirality in these modes. While each isolated metasurface exhibits linear polarization determined by the notch angle, interlayer coupling induces elliptical polarization in each layer. Consequently, the far-field polarization of the bi-layer metasurface is shaped by two arrays of elliptically polarized dipoles that conform to C_2 symmetry. The vertical offset of these dipoles introduces an additional propagation phase, which, under certain conditions, can result in purely circular polarization of the hybrid resonant mode.

Q3 The double alignment lithography is a precise but potentially complex and time-consuming technique. Scaling up this process for mass production could lead to issues in maintaining alignment accuracy and consistency over large areas. Electron beam lithography is used for metasurface patterning and is known for its high resolution but low throughput. **A transition to higher throughput lithography techniques, such as photolithography or nanoimprint lithography, may be required to achieve commercial scalability.** The use of materials such as silicon and fused silica in nanostructures is well established, but integrating these materials with active materials such as perovskite quantum dots at large scale could pose issues with the homogeneity and stability of the materials. **The summary and outlook would benefit from a discussion of the aspect and challenge of scalability**.

Response: We agree with the referee that the double alignment electron beam lithography is a precise but potentially complex and time-consuming technique. It is important to scale up this process for mass production via the transition to higher throughput lithography techniques, such as photolithography or nanoimprint lithography with commercial scalability.

Revisions: In the revised manuscript, we have added a paragraph to the **Discussion** section addressing the future aspects of large-scale nanofabrication, including commercial scalability. We also discuss potential challenges, such as the deposition of perovskite quantum dots over large areas using the directed self-assembly method.

> Doublet alignment lithography (DAL) is a precise and powerful technique for rapid prototyping in proof-of-concept experiments. However, it is time-consuming and typically limited to patterning areas of a few mm^2. For scaling up to mass production and commercial applications, it is crucial to develop higher throughput lithography techniques, such as photolithography with DUV/EUV[52] or nanoimprint[53] lithography with accurate alignment capabilities. Additionally, achieving uniform deposition of perovskite quantum dots over large areas remains a challenge. Advancing the directed self-assembly method, which has been optimized for uniform deposition of Au[54] and up-conversion nanoparticles[55], may provide a viable solution.

Accordingly, we added 4 new references:

52 Park, J.-S. et al. All-glass, large metalens at visible wavelength using deep-ultraviolet projection lithography. Nano Letters 19, 8673-8682 (2019).

53 Kim, J. et al. Scalable manufacturing of high-index atomic layer–polymer hybrid metasurfaces for metaphotonics in the visible. Nature Materials 22, 474-481 (2023).
54 Dong, Z. et al. Second-harmonic generation from sub-5 nm gaps by directed self-assembly of nanoparticles onto template-stripped gold substrates. Nano Letters 15, 5976-5981 (2015).
55 Xu, J. et al. Multiphoton upconversion enhanced by deep subwavelength near-field confinement. Nano Letters 21, 3044-3051 (2021).

Reviewer #2

The paper by Dmitrii Gromyko et. al. introduces a class of twisted bilayer photonic lattice structures that exhibit directional circular dichroism and chiral photoluminescence when coated with a layer of perovskite quantum dots. The authors include a comprehensive theoretical analysis of the corresponding photonic lattice resonance properties after varying the vertical layer separation and the lateral layer displacement. This analysis is then backed up by parametrically varied nanostructure fabrication followed by polarization and angle-resolved reflection spectra measurements. The study culminates in measurements of angular resolved photoluminescence, revealing that the emission occurs predominantly in a single quadrant in k-space and is dominated by LCP. The experimental results utilize doublet alignment lithography (DAL) for precise control over critical parameters such as twist angle, interlayer distance, and lateral displacement, leading to a CD of 0.94. While polarization and direction-selective light emission is important and the results are described clearly for the most part with well-presented data, I'm not convinced of the novelty of resonant chiral emission from a metasurface. The authors claim that "addressing emission directionality remains unexplored", that existing designs "often retain certain mirror symmetries, resulting in symmetric polarization and intensity patterns of emission" and "experimental observations of directional chiral radiation from resonant metasurfaces with near-field-coupled emitters remain obscure". However, directional chiral photoluminescence from DCM-coated 3D chiral metasurfaces (ref 31) and nonlinear chiral emission (third harmonic generation) from a 2D chiral silicon metasurface (ref 29) have been reported. Chiral resonances and thermal chiral emission have also been described theoretically in bilayer twisted high Q metasurfaces. **While the platform introduced here certainly seems new, I don't see evidence that unidirectional chiral emission is new, especially compared to ref 31**. If the phenomenon has been observed but the new platform improves performance either in brightness or polarization discrimination, this should be discussed, but I don't believe it has been. I therefore do not find the paper in its current form suitable for Nature Communications. To reach the bar for Nature Communications, the authors should **explain clearly the benefits of their platform compared to that of Ref 31**.

Response: Thank you for your keen eye in identifying the major contributions of this work regarding the new platform. We are excited to share that **radiation direction control is just one of many potential applications for the proposed bi-layer platform**.

For instance, in our other work [**R1**], we utilize bi-layer chiral metasurfaces with high rotational symmetry to achieve nontrivial nonlinear responses, such as converting linearly polarized light into circularly polarized third and second harmonic signals. Additionally, the bi-layer design provides new opportunities for investigating resonant modes in moiré structures. Furthermore, bi-layer metasurfaces integrated with nano-electro-mechanical systems (NEMS) have the potential for chirality switching. While the slanted pillar metasurface discussed in [**R2**], (Ref. 31 in the previous version of the main text) shows a high Q-factor and directional chiral emission (a direct comparison will be provided below), **our bi-layer metasurfaces offer greater versatility in both properties and**

applications. This includes advantages like unidirectional chirality, wide-angle intrinsic chirality, and a natural extension to moiré designs, due to their **multi-dimensional control**.

In particular, regarding "**radiation direction control**," a directional chiral emission beam was indeed observed in [**R2**]. However, several significant differences distinguish our approach from that work, as summarized in the **Table** below. The primary advantage of our platform is that **it enables directional emission beyond the normal direction**, facilitating **asymmetric chiral emission in the upper half-space** (for instance, LCP light emitted to the right, with no emission to the left). This feature, combined with the up-down radiation asymmetry, allows our proposed platform to function as a unidirectional grating coupler that is selectively responsive to circularly polarized waves.

	[R2] (Ref. 31)	**This work**
Underlying Mechanism	The high directionality of emission is conditioned by the **steep variation of the Q-factor.**	The observed emission asymmetry is due to the **unidirectional nature of the resonant mode in momentum space**.
Radiation Properties	The highest emission intensity occurs at **normal incidence**. **Limitation**: This method is limited to normal incidence, preventing its use for achieving directional off-gamma chiral emission. This restricts the potential applications of the metasurface, such as in chiral grating couplers or asymmetric light routers.	We observe circularly polarized light resonantly emitted along the positive x-axis, with the intensity peaking at approximately **10-12 degrees**. Emission is significantly suppressed in the opposite direction.

References:

R1. Gromyko, D., Loh, J. S., Feng, J., Qiu, C.-W. & Wu, L. Enabling all-to-circular polarization upconversion by nonlinear chiral metasurfaces with rotational symmetry. *arXiv preprint arXiv:2407.19293* (2024).

R2. Zhang, X., Liu, Y., Han, J., Kivshar, Y. & Song, Q. Chiral emission from resonant metasurfaces. *Science* **377**, 1215-1218 (2022).

Revisions: To clarify our position, we have added the following statement to the **Introduction**:

> We term this effect "unidirectional chiral emission". The radiation pattern is influenced by a strong asymmetry in the up-down direction, which is inverted along the lateral directions. This results in off-normal chiral emission, distinct from the highly collimated chiral emission reported previously[33]. In that study, the emission is confined to the normal direction due to the rapid increase in the quasi-BIC Q-factor near the Γ-point. In contrast, our metasurfaces exhibit superior performance compared to other designs, which typically feature symmetrically positioned points of opposite chirality in momentum space and radiate over a broad solid angle[42-44]. This work suggests the potential for full multi-dimensional control of polarization response, enabling reconfigurable metasurface design via its integration with nano-electro-mechanical systems[47] or via scaling to the terahertz frequencies[48].

Q1. In the introduction, particularly in the second paragraph, the authors should provide a brief review of the mechanism behind unidirectional resonant modes enhancing chiral emission, rather than merely mentioning extended structure and near-field emission enhancement. Additionally, there should be a connection between intrinsic to extrinsic chirality. A concluding sentence in the second paragraph should comprehensively summarize past results, highlighting the absence of research on intrinsic chirality and its application in emission control, to be introduced in the third paragraph.

Response & Revisions: We sincerely appreciate your improvement suggestions. Following your guidelines, we have revised the **Introduction** section accordingly.

> Dielectric metasurfaces with unidirectional resonant modes[17-20], which emit solely to one side, offer a promising solution for controlling radiation direction. Randomly polarized emitters distributed across the metasurface can couple to these resonant modes through near-field interactions, radiating according to the resonance's far-field properties. Unidirectional resonances can be derived from symmetry-protected bound-in-continuum (BIC) resonances[21] by breaking up-down symmetry[17,18]. This symmetry breaking splits the BIC resonance into two points of opposite chirality in momentum space. Further increasing the asymmetry merges these chirality points, thereby restricting resonant radiation to one side. However, the impact of unidirectional resonance polarization properties on the opposite side of the metasurface remains an unexplored area. Meanwhile, various resonant nanostructures with strong chiral effects at normal light incidence (*i.e.*, intrinsic chirality) have been explored, including multi-height metasurfaces[22-25], flat metasurfaces[26-32], slant-geometry metasurfaces[33,34], stacked and twisted bi-layer nanostructures[35-39], and chiral nanocavities[40,41]. Efforts have also been made to achieve absolute chirality at oblique light incidence (*i.e.*, extrinsic chirality) in flat metasurfaces with broken symmetry[42-44] or precise engineering of synthetic valleys in the folded Brillouin zone[45]. Nonetheless, these designs often retain certain mirror symmetries, resulting in symmetric polarization and intensity patterns of emission. In general, the transition from extrinsic to intrinsic chirality is achievable by tilting the walls of the structure which results in a continuous migration of the chirality points in the momentum space[33,34], similar to the recipe of unidirectionality. In most of these cases, chiral resonances in metasurfaces evolve from the achiral modes found in flat metasurfaces and, thus, are considered indivisible entities, missing one or several degrees of design freedom. On the contrary, stacked designs with vertical and lateral offsets naturally introduce a phase shift into the electromagnetic wave scattering problem, allowing for chirality[24] and directionality engineering[46]. Ultimately, while both unidirectional and chiral resonances were demonstrated, observations of unidirectional chiral remain obscure, likely due to the challenge of achieving multidimensional precise control.

Q2 In Figure 1(b), the unit cell of D2, which features a twisted angle and lateral shifts, significantly increases the dimensions of the Fabry-Perot cavity. **How is this unit cell defined in the simulation** and **how does it change the background Fabry-Perot resonance**?

Response: The top view of the unit cell is schematically illustrated in **Fig. R1**. The unit cell maintains consistent dimensions across all shifts and twist-angles between the disc notches. For large shifts, the unit cell is defined using the discs that are divided into several segments to satisfy periodicity conditions. For instance, with perfect alignment at (dX,dY)=(0nm,0nm) and a displacement of (dX,dY)=(200nm,100nm), the unit cells are defined as follows.

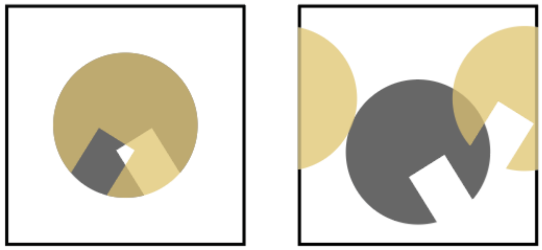


Fig. R1 Examples of unit cells of the metasurfaces with perfect alignment at (dX,dY)=(0nm,0nm) and a displacement of (dX,dY)=(200nm,100nm).

Lateral shifts have no significant effect on the background Fabry-Perot resonance. For example, the calculated and measured reflectance spectra for the metasurface D2 (**Supplementary material SI-8**, copied below) demonstrate a near-zero background, similar to that of metasurface A1 with perfect alignment.

8 Extrinsic chirality of configuration D2

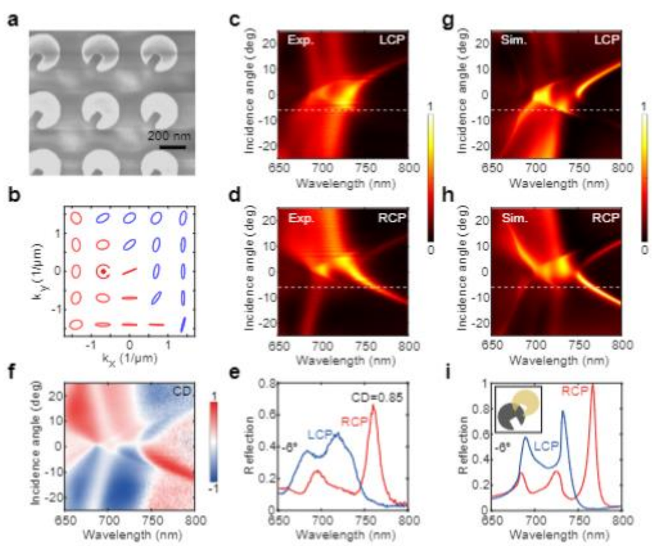


Figure S14: Reflection properties of bi-layer metasurface D2 with extrinsic chirality. **a,** SEM image of the bi-layer twisted metasurface with the designed lateral displacement $\Delta X = 180$ nm, $\Delta Y = 120$ nm. **b,** Calculated eigenpolarization map of the anti-symmetric resonance. **c, d,** Measured angle-resolved reflection spectra under LCP and RCP illumination. **e,** Measured reflection spectra at 6 degrees incidence corresponding to the dashed lines in **c** and **d**. **f,** Angle-resolved circular dichroism extracted from the reflection spectra in **c**, **d**. **g-i,** Simulated reflection spectra corresponding to **c-e**.

Additionally, we provide simulated reflectance spectra of the “open” structure (same parameters as we used for structures A1) with lateral shifts dY=0, dX=±100 nm. From **Fig. R2**, we observe that the background reflectance stays below 10%.

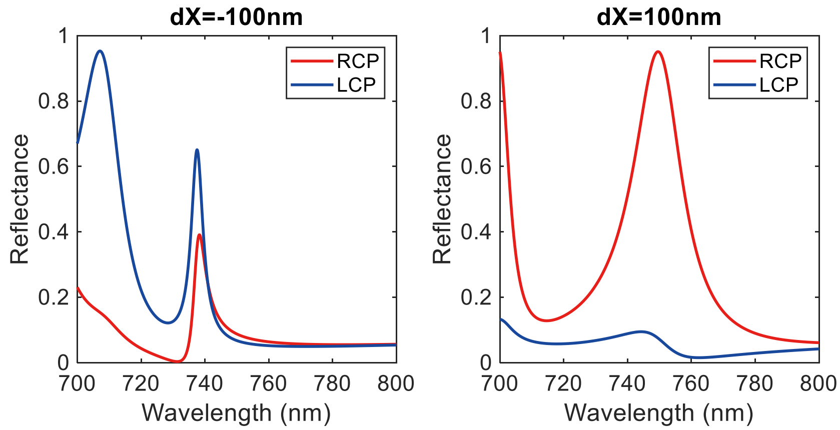


Fig. R2 Reflectance spectra of bi-layer metasurface with lateral shifts dY=0, dX=±100 nm.

Q3 In line 180 and the following paragraph, although the authors explain and demonstrate in the nearfield regime, that there is minimal environmental impact between the "open structure" and fully silica encapsulation, what is the fundamental experimental limitation in creating an "open structure"? **Is it feasible to further evaporate silica after drop-casting the quantum dots (QD)**?

Response: There are no fundamental experimental limitations to creating an "open structure," which is actually simpler than achieving complete silica encapsulation. It is feasible to deposit additional silica onto quantum dots (QDs) after drop-casting them, using methods such as e-beam evaporation or spin coating a thick layer of FOX resist, provided that these deposition processes do not adversely affect the QDs.

Q4 Why do the dimensions shown in Figure 3, such as A1 and A2, slightly differ from those of the near-field and intermediate structures depicted in Figure 2? What is the possible reason for the increased left-handed circular polarization (LCP) with a lower resonance wavelength in the experiment compared to the simulation?

Response: The parameters differ slightly, as illustrated in **Fig. 2** in the main text, where we explore options for achieving both high Q-factor and high chirality of the resonant modes. A high Q-factor enhances the resonant mode's strength and allows for the spectral separation of two resonant modes in the intermediate regime. However, due to unavoidable nonradiative losses, **there is always a tradeoff between resonant amplitude and Q-factor in practical samples**. Consequently, we used slightly modified system parameters for fabrication, resulting in a larger notch-to-radius ratio of the discs. This modification leads to greater symmetry breaking and, thus, increased radiative losses. When we reduced the notch size in structure A2 (Intermediate regime) compared to structure A1 (Near field regime), we observed a significant drop in resonant amplitude while the Q-factor improved.

To illustrate the impact of nonradiative losses on resonant amplitudes, we performed reflectance simulations for structures A1 and A2 with imaginary parts of the refractive index
k=0 and k=0.01, as depicted in **Fig. R3**. The simulations reveal that material losses broaden the resonances and reduce the resonant amplitude. For metasurface A2, which has narrower resonances and smaller radiative losses, the decrease in resonant amplitude is more pronounced. This is because nonradiative losses (from material absorption or sample imperfections) become more significant compared to radiative losses when they dominate. Conversely, metasurface A1, with its larger notch and consequently higher radiative losses, shows a smaller reduction in resonant amplitude due to additional material losses. Our simulations with k=0.01 align well with the experimentally measured spectra, confirming their accuracy.

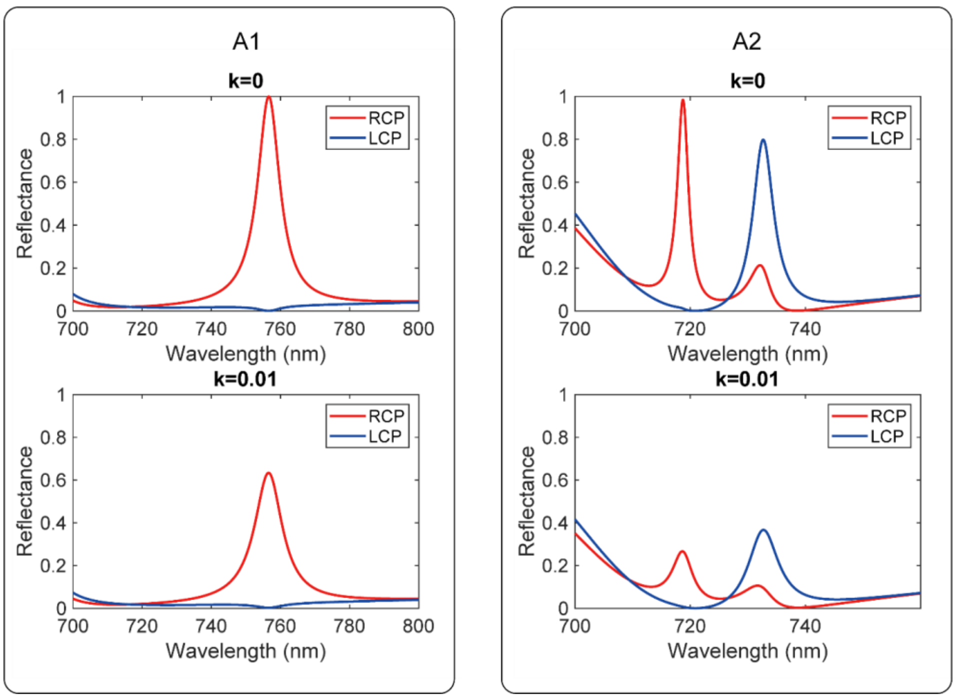


Fig. R3 Simulated reflectance spectra for the metasurfaces A1 and A2 with dielectric discs characterized by the imaginary part of the refractive index k=0 and k=0.01 respectively.

The **simulated** (assuming k=0.01) and **measured** reflectance spectra for metasurface A2 are compared in **Fig. R4**. The observed **increased** left-handed circular polarization at the lower resonance wavelength in the experimental data can be attributed to **(1)** additional background reflections at the flat air-SiO□ boundary, and **(2)** minor variations in the thickness of the SiO□ separation layer in the fabricated sample. These factors may contribute to slight deviations from the ideal chiral response predicted by the simulations.

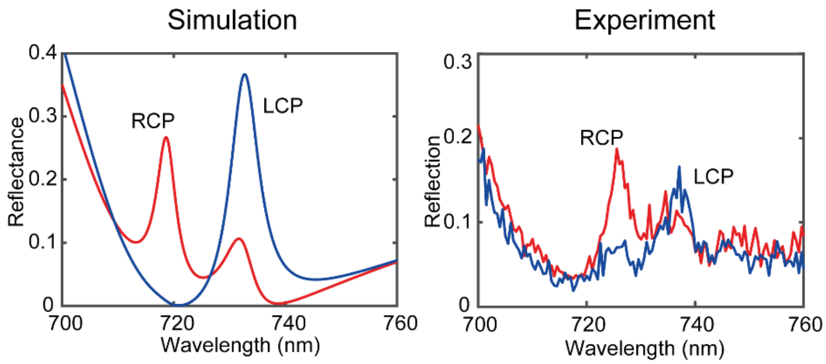


Fig. R4 Reflectance spectra for the metasurface A2: simulated with k=0.01 (left) and measured (right).

Q5 In line 218, what specific fabrication irregularities are being referred to? Please specify whether these irregularities occur within a single layer or between the two layers.

Response & Revision: Thank you for your question. These irregularities occur both within a single layer (*i.e.*, surface roughness of silicon discs), as well as between the two layers (*i.e.*, thickness variation of the SiO_2 spacing layer). To clarify this point, we have included the following sentence in the manuscript:

> A1. This should be attributed to small radiative losses of the resonance and the presence of nonradiative losses, which are inevitable even in all-dielectric structures due to the finite size of the sample and scattering on fabrication irregularities. In the bi-layer structure, these irregularities are caused by the surface roughness of the silicon discs in each layer and variations in the thickness of the SiO_2 separation layer. Nevertheless, configuration A2 demonstrates a chiral response at a normal incidence of illumination associated with the symmetric mode (Fig. 3i), which accords well with the predictions of the dipole model.

Q6 In line 248, how does lateral displacement affect the coupling model discussed in the design section?

Response: The coupled dipole model presented in our manuscript is valid when **the eigenmode problem exhibits C□ symmetry**. Under these conditions, the resonant modes in the top and bottom metasurfaces are excited with equal amplitudes, either in phase or out of phase, as detailed in **SI-4**, which includes the equations **S26**, **S29**, and **S30** (shown below). This behavior is a direct consequence of the symmetry of the full coupling Hamiltonian [**R1**, **R2**]:

$$\mathcal{H} = \begin{pmatrix} \Omega^{a} + V_{aa} & V_{ab} \\ V_{ba} & \Omega^{b} + V_{bb} \end{pmatrix} = \begin{pmatrix} \Omega^{a} + \left\langle I_u^{a} \middle| \mathcal{P}\tilde{S}^{b}_{ud}\mathbb{D}_{dd}\mathcal{P} \middle| O_d^{a} \right\rangle & \langle I_u^{a}|\mathbb{D}_{uu}\mathcal{P}|O_u^{b}\rangle \\ \langle I_d^{b}|\mathbb{D}_{dd}\mathcal{P}|O_d^{a}\rangle & \Omega^{b} + \left\langle I_d^{b} \middle| \mathcal{P}\tilde{S}^{a}_{du}\mathbb{D}_{uu}\mathcal{P} \middle| O_u^{b} \right\rangle \end{pmatrix} \tag{S26}$$

In our model, the dipoles represent the resonant input and output vectors in the far-field channel, as addressed in the Theory of Coupled Mode Theory (TCMT) [**R3**]. **However, introducing lateral shifts breaks the C□ symmetry, leading to two primary effects**:

1. **Asymmetric Mode Excitation**: The coupling Hamiltonian no longer retains its symmetry, resulting in the top and bottom layers being excited with unequal amplitudes and with a phase difference that is not necessarily a multiple of π.
2. **Modification of Scattering and Fabry-Perot Operators**: The background scattering matrices $\tilde{S}$ and Fabry-Perot operators D interact with and modify the resonant input and output vectors of the top and bottom metasurfaces.

$$\langle I^{\mathrm{c}}| = X^{-1}\begin{pmatrix} \langle I_d^{\mathrm{a}}| + \langle I_u^{\mathrm{a}}|\,\mathcal{P}\tilde{\mathrm{S}}_{ud}^{\mathrm{b}}\mathbb{D}_{dd}\mathcal{P}\tilde{\mathrm{S}}_{dd}^{\mathrm{a}} & \langle I_u^{\mathrm{a}}|\,\mathbb{D}_{uu}\mathcal{P}\tilde{\mathrm{S}}_{uu}^{\mathrm{b}} \\ \langle I_d^{\mathrm{b}}|\,\mathbb{D}_{dd}\mathcal{P}\tilde{\mathrm{S}}_{dd}^{\mathrm{a}} & \langle I_u^{\mathrm{b}}| + \langle I_d^{\mathrm{b}}|\,\mathcal{P}\tilde{\mathrm{S}}_{du}^{\mathrm{a}}\mathbb{D}_{uu}\mathcal{P}\tilde{\mathrm{S}}_{uu}^{\mathrm{b}} \end{pmatrix}, \qquad \text{(S29)}$$

$$|O^{\mathrm{c}}\rangle = \begin{pmatrix} |O_u^{\mathrm{a}}\rangle + \tilde{\mathrm{S}}_{uu}^{\mathrm{a}}\mathcal{P}\tilde{\mathrm{S}}_{ud}^{\mathrm{b}}\mathbb{D}_{dd}\mathcal{P}\,|O_d^{\mathrm{a}}\rangle & \tilde{\mathrm{S}}_{uu}^{\mathrm{a}}\mathbb{D}_{uu}\mathcal{P}\,|O_u^{\mathrm{b}}\rangle \\ \tilde{\mathrm{S}}_{dd}^{\mathrm{b}}\mathbb{D}_{dd}\mathcal{P}\,|O_d^{\mathrm{a}}\rangle & |O_d^{\mathrm{b}}\rangle + \tilde{\mathrm{S}}_{dd}^{\mathrm{b}}\mathcal{P}\tilde{\mathrm{S}}_{du}^{\mathrm{a}}\mathbb{D}_{uu}\mathcal{P}\,|O_u^{\mathrm{b}}\rangle \end{pmatrix} X. \qquad \text{(S30)}$$

These effects occur even if the metasurface is composed of stacked discs without notches. Once a lateral shift is introduced, the C□ symmetry is violated, causing the coupling between BIC modes of isolated metasurfaces to result in radiative modes of the bilayer metasurface with the applied shift.

Thus, to fully account for the effects of notches and lateral shifts in the metasurface, the comprehensive coupling model should incorporate calculations of the complete resonant input and output vectors using the Fourier Modal Method [**R4**]. This approach includes not only the far-field amplitudes but also considers multiple Fourier harmonics to account for the evanescent near fields. The process involves solving the eigenproblem for the coupling Hamiltonian to determine the resonant amplitudes, which are then used to evaluate the resonant input and output vectors of the coupled system. The relative lateral shift of the bottom structure by distances D_x and D_y is accounted for using the following transformation:

$$\tilde{\mathrm{S}}^{\mathrm{b}} \to \tilde{\mathrm{S}}^{\mathrm{b}\prime} = P^{+}\tilde{\mathrm{S}}^{\mathrm{b}}P$$
$$\left|O_n^{\mathrm{b}}\right\rangle \to \left|O_n^{\mathrm{b}\prime}\right\rangle = P^{\dagger}\left|O_n^{\mathrm{b}}\right\rangle, \left\langle I_n^{\mathrm{b}}\right| \to \left\langle I_n^{\mathrm{b}\prime}\right| = \left\langle I_n^{\mathrm{b}}\right|P,$$

where P is the shift matrix

$$P = \mathrm{diag}\{\exp(iK_xD_x + iK_yD_y)\},$$

with $K_{x,y}$ being the x and y projections of the wavevectors for the Fourier harmonics:

$$K_x = k_x + G_x, K_y = k_y + G_y$$
$$\mathbf{G} = \left\{\frac{2\pi}{p_x}g_x, \frac{2\pi}{p_y}g_y, 0\right\}, g_{xy} = 0, \pm 1, \pm 2, \dots$$

Here, $k_{x,y}$ are the lateral projections of the incoming wave wavevector, $p_{x,y}$ are the structure periods, and $\mathbf{G}$ is the reciprocal lattice. This comprehensive approach was demonstrated in [**R5**] and is similarly employed in recent work [**R6**], which investigates stacked membranes with C□ symmetry in the constituent layers.

References:

R1 Gippius, N. A.; Weiss, T.; Tikhodeev, S. G.; Giessen, H. Resonant mode coupling of optical resonances in stacked nanostructures. Optics Express 2010, 18, 7569–7574.

R2 Weiss, T.; Gippius, N. A.; Granet, G.; Tikhodeev, S. G.; Taubert, R.; Fu, L.; Schweizer, H.; Giessen, H. Strong resonant mode coupling of Fabry–Perot and grating resonances in stacked two-layer systems. Photonics and Nanostructures-Fundamentals and Applications 2011, 9, 390–397.

R3 Fan, S.; Suh, W.; Joannopoulos, J. D. Temporal coupled-mode theory for the Fano resonance in optical resonators. JOSA A 2003, 20, 569–572.

R4 Tikhodeev, S. G.; Yablonskii, A.; Muljarov, E.; Gippius, N. A.; Ishihara, T. Quasiguided modes and optical properties of photonic crystal slabs. Physical Review B 2002, 66, 045102

R5 Gromyko, D.; Dyakov, S.; Tikhodeev, S.; Gippius, N. Resonant mode coupling approximation for calculation of optical spectra of stacked photonic crystal slabs. Part I. Photonics and Nanostructures – Fundamentals and Applications 2023, 53, 101109.

R6 Ni, X. et al. Three-dimensional reconfigurable optical singularities in bilayer photonic crystals. Physical Review Letters 132, 073804 (2024).

Q7 In the Methods and Quantum Emitter Synthesis section, how did the authors ensure the uniform distribution of the FAPbI3 QD solution after drop-casting, without aggregation on a specific spot on a disk with a notch?

Response: We thank the referee for the insightful question. The referee's observation is valid: when using the drop-casting method to deposit FAPbI□ quantum dots (QDs) onto a disk with a notch, a uniform distribution of the QDs cannot be ensured.

Revision: In response to this, we have included an SEM image in the Supplementary Materials (**Fig. S19**) to illustrate the distribution of FAPbI□ QDs.

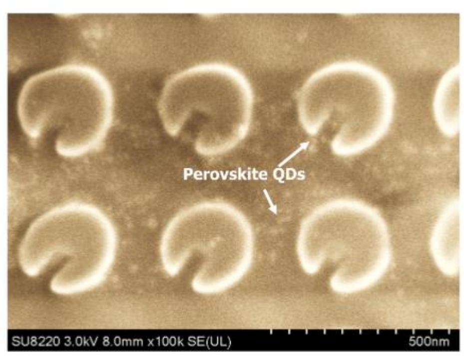


Figure S19: SEM image of the bi-layer metasurface, top layer with deposited perovskite quantum dot emitters.

Meanwhile, to prevent potential confusion for future readers, we have revised the "**Fabrication**" section in the Methods:

Similarly, the bi-layer metasurface with displacement can be realized through the intentional shift of exposure position (Fig. S18h). The $FaPbI_3$ QDs were deposited onto the top silicon disc array via the drop-casting method to realize unidirectional spin emission (Fig. S18j). An SEM image of the metasurface with deposited QDs is shown in Fig. S19.

Reviewer #3:

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response: We are delighted to extend our gratitude for your role as a distinguished reviewer for our manuscript. We have carefully addressed the suggestions provided by all the referees and have made significant revisions to enhance the manuscript. We kindly invite you to review the revised version, which we hope meets the high standards set by Nature Communications. Your feedback is greatly valued, and we are eager to hear your thoughts.

Reviewer #4:

In the manuscript “Unidirectional Chiral Emission via Twisted Bi-layer Metasurfaces” by D. Gromyko et al. the authors propose a bi-layer nanostructure consisting of dielectric discs that exhibit sharp resonances of high chirality. The reported chiroptical response of the structure originates from the resonant modes simultaneously excited in the upper and lower layers of the structure. In isolated single-layer metasurfaces, these modes exhibit linear far-field polarizations, however, when coupled within a bi-layer structure, they act as scatterers of elliptically polarized light. The authors provide a coupling model to describe the amplitude, phase, and direction of the light scattered by both layers. I can recommend the publication of the manuscript in Nature Communications after several necessary revisions.

Response: We appreciate the referees' constructive comments, which have significantly enhanced our manuscript. We have carefully revised the manuscript in response to your suggestions and are confident that these improvements meet the high standards of Nature Communications. We hope the revised version is both compelling and satisfactory.

Q1 The authors claim that the resonance is unidirectional based on the radiative losses in the upper half-space evaluated for the resonance at different points in the reciprocal space. However, smaller radiative losses of the resonance may be associated with smaller energy losses and, hence, larger enhancement of the near field. Therefore, a resonance with smaller losses might produce an emission with greater amplitude. **The relation between the radiative losses and the unidirectionality of the resonance should be clarified**.

Response & Revision: Thank you very much for your insightful comment. To clarify our findings, we have added the following discussion to **Section SI-7** of the Supplementary Materials:

7 Unidirectional resonances in bi-layer metasurfaces with lateral displacement

At each point in the reciprocal space, the total radiative losses of the resonance can be represented as a sum of losses in the top and bottom half-spaces: $\gamma_r = \gamma_{top} + \gamma_{bot}$. The radiative losses are related to the radiative Q-factor of the resonance $Q = \omega/2\gamma_r$ (Fig. S12b), which can be extracted from the eigenmode simulation of a nonglossy metasurface. At the same time, the radiative losses are proportional to the squared absolute value of the far-field amplitude of the resonant mode, which we illustrate in Fig. 1e of the main text and Fig. S13. At the same time, a unidirectional resonance exhibits large radiation asymmetry in the up-down direction (Fig. S12c), which is $\gamma_{top}/\gamma_{bot} \ll 1$ or $\gamma_{top}/\gamma_{bot} \gg 1$.

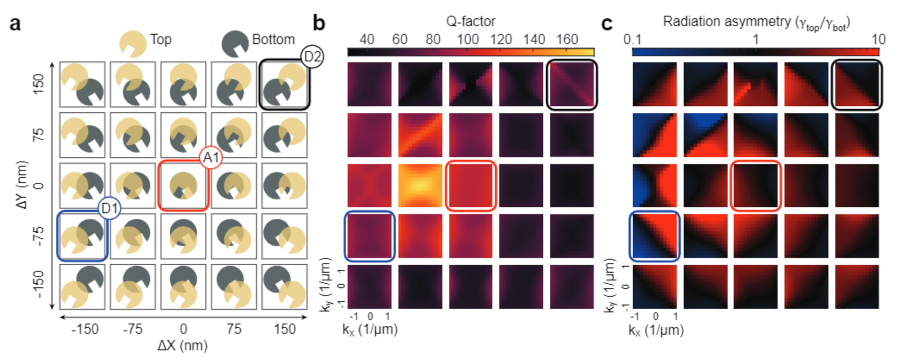


Figure S12: **a,** Unit cell (top view) of bi-layer metasurfaces with possible lateral shifts ranging from -150 nm to 150 nm in both directions. **b, c,** Corresponding maps of Q-factor and up-down radiation asymmetry $\gamma_{top}/\gamma_{bot}$ for the anti-symmetric (lowest) resonant mode.

Q2 The chirality of the emission is demonstrated for the structures with displacement D1 and D2. It would be beneficial to add similar emission spectra for the structure without displacement.

Response & Revisions:

Thank you for your suggestion. We added the measured emission spectra for a sample with an A1 design, *i.e.*, without displacement to **Section SI-5** of the Supplementary Materials.

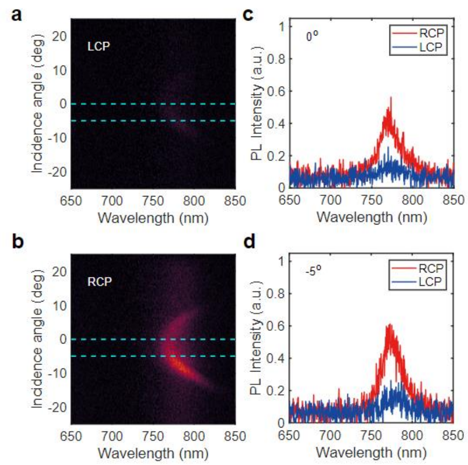


Figure S5: Chiral emission from bi-layer metasurface A1 with perfect alignment (zero lateral displacement) along x direction. **a, b,** Measured angle-resolved spectra of the LCP and RCP photoluminescence emission. **c, d,** Emission spectra at 0° and $-5°$ emission angles.

Q3 The fabrication technique used in the manuscript is called "doublet alignment lithography". While the structure indeed requires a single alignment operation between two layers, it remains unclear what the meaning of "doublet alignment" is. To which pair does the noun 'doublet' refer? It also would be beneficial to explain how this approach to alignment differs from the previously reported techniques such as https://doi.org/10.1038/ncomms14180, https://pubs.acs.org/doi/10.1021/acsphotonics.7b01460, https://pubs.acs.org/doi/10.1021/acsnano.0c07295.

Response: The referee is correct that our description of the "doublet alignment lithography" in the Methods section was not sufficiently clear.

Specifically, our nanofabrication process involves the following steps: (1) We first fabricate alignment markers using Cr/Au; (2) Next, we perform the first alignment and etch the bottom layer silicon nanostructures; (3) We then spin coat FOX resist to create a flat dielectric layer; (4) Finally, we carry out the second alignment to fabricate the top layer silicon nanostructures.

Our process for aligned nanofabrication follows the procedure outlined in the paper [**R1**], where alignment markers are fabricated first using electron beam lithography (EBL). However, the references [**R2**] and [**R3**] do not clearly describe the fabrication of alignment markers. It is unclear whether these references fabricated the alignment markers first or if the markers were created simultaneously with the first layer.

References:

R1 Zhao, Y. et al. Chirality detection of enantiomers using twisted optical metamaterials. Nature Communications 8, 14180 (2017).

R2 Fasold, S. et al. Disorder-enabled pure chirality in bilayer plasmonic metasurfaces. ACS Photonics 5, 1773-1778 (2018).

R3 Tanaka, K. et al. Chiral bilayer all-dielectric metasurfaces. ACS Nano 14, 15926-15935 (2020).

Revision: We have revised this section to provide a detailed account of the fabrication process for the "doublet alignment lithography."

evaporation and lift-off process (Fig. S18c). After that, the bottom Si disks with notches were patterned based on the 1st aligned EBL process (Fig. S18d). The patterned a-Si sample was then etched using a chlorine (Cl_2)-based inductively-coupled-plasma reactive-ion etching (ICP RIE, Oxford Instruments Plasmalab System 100) with an ICP power of 300 watts, an RF power of 100 watts under a pressure of 5 mTor and a temperature of 0°C. Then, the middle SiO_2 was uniformly distributed by spin coating of commercially available Fox 16 resist at a speed of 5000 rpm without ramping up the process (Fig. S18e). As a result, the notches of the bottom disks could be filled while the top surface of SiO_2 is flat, which promises a fully embedded environment. After that, CF_4 gas-based RIE was performed to thin the middle SiO_2 layer to 60 (280) nm for the configurations in near-field (intermediate) regimes. The thickness of the final SiO_2 layer was confirmed by a thin film analyzer (FILMETRICS, F50). Subsequently, another 50 nm a-Si layer was deposited using ICP CVD to prepare for the top disk (Fig. S18f). In the next step, we carried out the 2nd aligned EBL process to pattern the top silicon notch nanostructures and the Cl_2-based etching (Fig. S18g). To fabricate the embedded structure, another SiO_2 layer was covered on top of the open structure by spin-coating (Fig. S18i). Similarly, the bi-layer metasurface with displacement can be realized through the intentional shift of exposure position (Fig. S18h). The $FaPbI_3$ QDs were deposited onto the top silicon disc array via the drop-casting method to realize unidirectional spin emission (Fig. S18j). An SEM image of the metasurface with deposited QDs is shown in Fig. S19.

Q4 The authors claim that the bi-layer design is essential for achieving chirality. However, the nanostructure comprising two layers of notched discs looks rather complex. Is it possible to achieve a similar performance in a structure with **a simplified geometry**?

Response: Achieving chirality generally requires breaking all mirror symmetries of structures, including out-of-plane (up-down) symmetry. This is commonly achieved in multi-layer structures [**R1**, **R2**], multi-height designs [**R3**], or those with slanted geometries [**R4**, **R5**]. Recent research has shown that maximum chirality can also be attained in monolayer structures with straight walls through environmental asymmetry of the substrate and superstrate [**R6**, **R7**]. Remarkably, chiral effects have been observed even in up-down symmetric free-standing photonic membranes, which either involve simultaneous excitation of TE and TM resonances [**R8**] or exhibit birefringent behavior [**R9**]. Despite their geometric simplicity, **it is still uncertain whether additional unidirectionality of resonances can be achieved in monolayer chiral structures**. Conversely, unidirectional resonances have been demonstrated in bi-layer structures with simple geometries, such as arrays of parallel bars shifted along the periodicity direction [**R10**].

In contrast to these chiral metasurfaces, our design operates on the hybridization of two sharp resonant modes found in the upper and lower layers. While the dipole approximation is used to describe the far-field properties of these hybrid resonant modes, **our structure is not merely a stack of elliptical dielectric bars**, which could be seen as a simplified version. In structures with dielectric bars, the resonance Q-factor depends on the bar size. Small bars act as perturbations to a homogeneous waveguiding layer, creating weak gratings with quasi-guided resonances. These resonances exhibit a Q-factor inversely proportional to the square of the perturbation strength (*i.e.*, the bar size) [**R11**, **R12**]. Although such resonances can have high Q-factors, they result in weak spectral separation and poor spatial localization of modes. Without the dielectric bars, resonant modes do not exist.

On the contrary, our design utilizes large dielectric discs whose resonant modes are present regardless of notch size. The large discs scatter light more efficiently within the periodic layer, creating a high-contrast grating that allows for strong spectral separation of the resonant modes, some of which are bound states in the continuum (BIC). The addition of small notches opens radiation channels and influences the far-field coupling properties of the resonances at normal incidence, while minimally affecting the near-field distribution and spectral positions of the quasi-BIC resonances.

We are excited to highlight that **chirality and radiation direction control are just a few of the many potential applications for the proposed bi-layer platform**. For example, in our other work [**R13**], we use bi-layer chiral metasurfaces with high rotational symmetry to achieve nontrivial nonlinear effects, such as the conversion of linearly polarized light into circularly polarized third and second harmonic signals. Additionally, the bi-layer design opens new avenues for exploring resonant modes in moiré structures. Furthermore, bi-layer metasurfaces integrated with nano-electro-mechanical systems (NEMS) could enable chirality switching. **Overall, our bi-layer metasurface platform provides exceptional versatility**, offering benefits such as unidirectional chirality, wide-angle intrinsic chirality, and inherent adaptability to moiré designs due to their **multi-dimensional control**.

Meanwhile, we are exploring **simplified geometries** that could potentially offer similar performances, at least for certain applications.

References:

R1 Overvig, A., Yu, N. & Alù, A. Chiral quasi-bound states in the continuum. Physical Review Letters 126, 073001 (2021).

R2 Tanaka, K. et al. Chiral bilayer all-dielectric metasurfaces. ACS nano 14, 15926-15935 (2020).

R3 Kühner, L. et al. Unlocking the out-of-plane dimension for photonic bound states in the continuum to achieve maximum optical chirality. Light: Science & Applications 12, 250 (2023).

R4 Chen, Y. et al. Observation of intrinsic chiral bound states in the continuum. Nature 613, 474-478 (2023).

R5 Zhang, X., Liu, Y., Han, J., Kivshar, Y. & Song, Q. Chiral emission from resonant metasurfaces. Science 377, 1215-1218 (2022).

R6 Chen, W. et al. Uncovering Maximum Chirality in Resonant Nanostructures. Nano Letters 24, 9643-9649 (2024).

R7 Gorkunov, M. V., Antonov, A. A., Mamonova, A. V., Muljarov, E. A. & Kivshar, Y. S. Substrate-induced maximum optical chirality of planar dielectric structures. arXiv preprint arXiv:2408.10742 (2024).

R8 Semnani, B., Flannery, J., Al Maruf, R. & Bajcsy, M. Spin-preserving chiral photonic crystal mirror. Light: Science & Applications 9, 23 (2020).
R9 Shi, T. et al. Planar chiral metasurfaces with maximal and tunable chiroptical response driven by bound states in the continuum. Nature Communications 13, 4111 (2022).
R10 Zeng, Y., Hu, G., Liu, K., Tang, Z. & Qiu, C.-W. Dynamics of topological polarization singularity in momentum space. Physical Review Letters 127, 176101 (2021).
R11 Rosenblatt, D., Sharon, A. & Friesem, A. A. Resonant grating waveguide structures. IEEE Journal of Quantum Electronics 33, 2038-2059 (1997).
R12 Huang, L. et al. Ultrahigh-Q guided mode resonances in an All-dielectric metasurface. Nature Communications 14, 3433 (2023).
R13 Gromyko, D., Loh, J. S., Feng, J., Qiu, C.-W. & Wu, L. Enabling all-to-circular polarization upconversion by nonlinear chiral metasurfaces with rotational symmetry. arXiv preprint arXiv:2407.19293 (2024).

Q5 It should be explained in more detail if it is possible to achieve this effect at a larger separation distance.

Response & Revision: The resonant modes of the chiral bi-layer metasurfaces with significant separation distances exhibit strong overlap, which may render them impractical. To clarify the coupling of modes at large distances, we have extended **Section SI-6** to include a derivation of the resonant energies using the far-field coupling Hamiltonian.

Let us examine the possibility of achieving maximum chirality in the purely far-field regime. At large H we can analytically evaluate the effective coupling Hamiltonian (S26) while neglecting near-field coupling. Using $A^2 = i\gamma(r+t)$ (see[8]), in the far-field approximation, it reads:

$$H = \begin{pmatrix} \Omega + i\gamma + \dfrac{i\gamma e^{2i(\Phi-\phi)}|r|(|r|+i|t|)}{1-|r|^2 e^{2i(\Phi-\phi)}} & \cos(2\theta)\dfrac{i\gamma e^{i(\Phi-\phi)}(|r|+i|t|)}{1-|r|^2 e^{2i(\Phi-\phi)}} \\ \cos(2\theta)\dfrac{i\gamma e^{i(\Phi-\phi)}(|r|+i|t|)}{1-|r|^2 e^{2i(\Phi-\phi)}} & \Omega + i\gamma + \dfrac{i\gamma e^{2i(\Phi-\phi)}|r|(|r|+i|t|)}{1-|r|^2 e^{2i(\Phi-\phi)}} \end{pmatrix}, \tag{S39}$$

where we explicitly write the real and imaginary part of the single-layer resonant frequencies $\omega_1^{a,b} = \Omega + i\gamma$, while the reflection phase Φ and propagation phase ϕ were defined in (S11) and (S25). Since maximum chirality can only be achieved at the background Fabry-Perot resonance (S16), we put $e^{i(\Phi-\phi)} = \pm 1$. The resonant energies of the purely far-field coupled modes are eigenvalues of the Hamiltonian:

$$\omega_{A,S} = \omega - \frac{|r||t|}{1-|r|^2} + i\frac{\gamma}{1-|r|^2} \pm \cos(2\theta)\frac{\gamma|t|}{1-|r|^2} \mp i\cos(2\theta)\frac{\gamma|r|}{1-|r|^2} \tag{S40}$$

This addition has been referenced in the **main text** accordingly.

near fields that resemble those of the unperturbed BIC mode. However, in this regime, *e.g.*, ($H = 280$ nm, $\theta = 45^o$), both symmetric and anti-symmetric quasi-BIC modes are accessible and exhibit high chirality while coupling to waves with opposite circular polarizations (Fig. 2f). When H exceeds the metasurface period, purely far-field coupling determines the properties of the coupled resonances. The energy splitting becomes insufficient, and two chiral resonances spectrally overlap (see Fig. 2a,g), no matter how θ changes (SI-6).

Full discussion and numerical simulations are presented in **SI-6**. We show that while in the far-field regime the response can be fully described with just several resonant parameters (the resonant frequency and radiative losses, the resonant in- and out-vectors, and background scattering coefficients in TCMT formalism), variation of these parameters does not allow for separation of chiral resonant modes.

Q6 For metasurface A2 the resonant peaks in reflectance are much smaller compared to those for metasurface A1. It is claimed that the nonradiative losses dominate over the radiative losses in structure A2. However, the authors use an objective with a significantly large NA=0.6, which should result in the excitation of resonant modes in a broad area of the reciprocal space at nonzero incidence angles. This dilutes the contribution of the sharp quasi-BIC mode at normal incidence and decreases the amplitude of the resonant reflectance. The angular properties of the excitation beam should be clarified, and experimental data of the reflectance spectra in dependence on the excitation beam aperture size are required.

Response: Thank you for this valuable comment. In our experiments, we used an objective with a numerical aperture (NA) of 0.6 to cover a broad range of angles, enabling us to simultaneously observe resonances across an angle range of approximately -37° to 37°. We appreciate the referee's point that this approach might reduce the signal-to-noise ratio, potentially causing some important details to be less discernible. However, as demonstrated in Fig. 3 of the main text, all resonances predicted by the simulations are clearly visible with the current setup. While reducing the effective NA of the objective by narrowing the incident beam aperture in k-space could improve the signal-to-noise ratio at smaller angles of incidence, we believe that the current signal-to-noise ratio is sufficiently high. Therefore, such a modification is unlikely to provide significant additional insight or enhance the qualitative results of our observations.

Reviewer #1 (Remarks to the Author):

The quality of the manuscript has been significantly improved by the review process. All questions and comments of the reviewers have been thoroughly and comprehensively addressed, and the suggested revisions have been successfully implemented. The manuscript now meets the high scientific standards and is recommended for publication.

Reviewer #2 (Remarks to the Author):

The authors have addressed my comments in detail and have clarified the novelty of their findings. I'm still not fully convinced that the chiral momentum space structure they claim provides unique opportunities for unidirectional chiral emission is fundamentally different from previous 3D chiral lattice resonances. However, I support publishing what I consider to be interesting and carefully examined measurements.

Reviewer #3 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4 (Remarks to the Author):

I agree with the authors' responses and I recommend the manuscript for publication in the Journal.

Reply to the reviewers:

We sincerely thank all the reviewers for their valuable comments and suggestions, which have greatly contributed to the improvement of our manuscript. Your insights and thoughtful feedback have been instrumental in refining our work, and we deeply appreciate the time and effort you invested in the peer-review process.