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Supplementary Material

Realization of Giant Superstructural Chirality at Broadband Optical Wavelengths via Perovskite Dielectric Metasurfaces

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Fig. S1 Ellipsometry data for low-loss high-index dielectric: (a) hydrogenated amorphous silicon $(a-Si:H)^1$ and the perovskite and (b) cesium lead bromide $(CsPbBr_3)^2$. a-Si:H depicts the significant transparency at the visible wavelength used for nanostrucutral engineering. CsPbBr₃ also shows a lower extinction coefficient at the working wavelengths of visible ranges.

Parameters	Optimal values
Length (L ₁)	205nm
Length (L ₂)	210nm
Width (W ₁)	105nm
Width (W ₂)	85nm
Nanostructures thickness (t_1)	400nm
Perovskite thickness (t_2)	100nm
Substrate thickness (t_3)	200nm
Periodicity in x-direction	650nm
Periodicity in y-direction	260nm
Relative rotation (δ)	45°

Table S1 Optimized design parameters



Fig. S2 Comparison of chiro-optical responses for CP incident light, i.e., (*without*) when no perovskite layer is used, (*middle*) when the perovskite is placed as a middle layer, (*top*) when the perovskite layer is coated on the top of nanostructures.



Fig. S3 Spectral absorption for the L-Enan superstructure. Maximum absorption is demonstrated for the RCP incident light and minimum for the LCP illumination.



Fig. S4 Spectral reflection for the L-Enan superstructure, which shows some reflective amplitude for RCP illumination.



Fig. S5 Circular dichroism at the working wavelengths for L-Enan and R-Enan, plotted as a function of the thickness of the perovskite layer.



Fig. S6 Perovskite used as a bottom layer: electric and magnetic field's distributions in the *xz*-plane at the working wavelengths of 488 nm, 532 nm, and 633 nm.



Fig. S7 Perovskite used as a top layer: electric and magnetic field's distributions in the *xz*-plane at the working wavelengths of 488 nm, 532 nm, and 633 nm.

Fabrication Process:

For Type I design, we anticipate a two-step fabrication process. Firstly, a controlled deposition of a few perovskite layers will be carried out utilizing a solution-based process, such as spin-coating or dip-coating. These methods allow for the controlled deposition of perovskite solution. To achieve uniformity, optimization of the solution concentration, deposition speed, and post-treatment processes will be implemented. Subsequently, nanopillars will be grown on the perovskite layer through techniques like nanosphere lithography or electron-beam lithography, followed by a selective etching process. The etching will define the nanopillar pattern with precision.

For Type II design, nanopillars will first be grown on the glass layer through electron-beam lithography, followed by a selective etching process. Subsequently, the perovskite layer will be placed using the abovementioned methods, such as spin-coating or dip-coating. Additionally, many other techniques can be explored to enhance uniformity over a large area surface.

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