Supporting Information

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3D Chiral MetaCrystals

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Supporting Information

*Three-Dimensional Chiral MetaCrystals*

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S1. Enantiomeric handedness and circularly polarized light-dependence

The interaction between the circularly polarized light and a chiral element leads to a different signal between the two polarization components. In this work, the measured LCP and RCP scattering spectra are observed for couples of single enantiomers, which have identical sizes (right-handed (RH, red) and left-handed (LH, blue), respectively), fabricated with the same growth condition. When changing the structural handedness, the scattering signal of single right- and left-handed structures shows an opposite chiral behavior, Figures S1 a, b, display the scattering spectra of single 1-loop helices enantiomeric couples (RH and LH, respectively) with 550nm VP height. Their minute mismatches in the optical response of the RH- and LH-helices can be only related to small structural differences and fabrication tolerances\(^1\). For comparison, the scattering spectra for RCP and RCP incident light of an achiral pillar, are plotted in figure S1c. The optical investigation has been performed on a pillar with 550nm height. The spectra show that the pillar optical response for the two circularly polarized light components, is almost overlapped, indicating the absence of circular polarization-dependence.

Figure S1. LCP and RCP scattering spectra measured for couples of single enantiomers (right-handed and left- handed (blue line), respectively) with VP 550nm, Figure S1a, b, respectively. These spectra show that, for two enantiomeric couples, the chiral behavior is inverted for all the investigated enantiomeric couples. c) Scattering spectra for RCP and RCP incident light on a pillar with 550 nm height, framed in black in the figure. The spectra show that, for a non chiral element, like the pillar, the optical response for the two circularly polarized light components, is overlapped. The insets represent the enantiomeric helix couples and the achiral nanopillar with scale bar 100nm.

S2. Compositional profile of Platinum-based helix systems
Platinum based-helical nanostructures grown by Focused ion beam induced deposition, exhibit a compositional profile made of a platinum-carbon alloy with platinum nanograins (with averaged size of 5nm), embedded in an amorphous carbon matrix with a volume percentage of 50% Pt and 45% C and a low residual Ga content, <5%. Figure S2a is a schematical representation of a helix section where the carbon matrix is the green disk, while the platinum grains are the blue circles. The optical dispersion has been numerically calculated using a finite difference time-domain based software (Lumerical FDTD Solutions), considering the material composition retrieved by Transmission Electron Microscope and Energy Dispersive X-ray Spectroscopy in\textsuperscript{2}. Owe to the complex compositional profile of the Pt helix, an effective medium approach can be applied. Particularly, FDTD analysis has been carried out for a C host matrix, enclosing randomly distributed spheroids (representing Ga clusters and platinum nanoparticles) in a large FDTD box with periodic boundary conditions and illuminating the system with a plane wave with normal incidence. The reflection coefficients (r) of the overall compound are retrieved and the effective refractive index is calculated as:

\[
n(\lambda) = n_{\text{host}}(\lambda) \cdot \frac{1 - r(\lambda)}{1 - r(\lambda)}
\]

where \( n_{\text{host}} \) is air refractive index and \( r \) is the complex reflection coefficient. Using the dispersion values of Pt from\textsuperscript{3}, Ga from\textsuperscript{4} and Carbon from\textsuperscript{5}, the dispersion curves are retrieved and displayed in Figure S2b.
Figure S2. a) Schematic view of the wire cross section which shows that the structure is composed by Pt nanograins embedded in an amorphous C matrix. The blue points correspond to Pt grains, while the amorphous carbon matrix is represented by the green disk. d) Analytical dispersion values (n-blue line, k-red line) calculated for the Pt-based helix.

S3. Chiral Metacrystals as a function Array of Pt helices as a function of the unit cell

The transmission spectra measured for chiral metacrystals, as well as the SEM images depicting the metacrysal morphology, by tuning the unit cell, are observed in figure S3.

The overall trend shows a redshift of all the spectral features, when LP is fixed and varying VP, as observed for their circular dichroism spectra. When tuning the lattice periodicity, instead, beside the redshift of the main spectral range, the measured transmission spectra also exhibit an enlargement of the circular birefringence, together with the sharpening of other peaks corresponding to the diffractive orders owed to the in-plane interactions among the helices.
Figure S1. SEM Images of the built chiral metacrystals composed by right-handed helices with different periodicities: Top row: LP is fixed at 550nm while VP varies VP of 270nm, 350nm, 550nm, 600nm, 800nm, from left to right, respectively. Bottom Row: VP is fixed at 550nm with variable LPs of 435nm, 500nm, 550nm, 600nm, 700nm, from left to right, respectively. The scale bar is 100nm. In the central panel there is a schematic representation of the geometrical parameter of the unit cell: The Vertical Period (VP), External Diameter (ED), the wire Diameter (WD), the Lateral Period (LP). At the center: Transmission spectra for the Metacrystals imaged above a) With LP fixed at 550nm and varying VP and b) With VP fixed at 550nm and varying LP.

S4. Excitation efficiency of optical resonances

Following the analytical expression for transferred extinction power from ref. 20 in the main text:

\[ P_{\text{ext},n}^\pm = \frac{1}{4} m E_0 j_0 \times \{ \text{sinc}[mh(k \mp k_h - k_{FP})] - \text{sinc}[mh(k \mp k_h + k_{FP})] \} \]

And using our numerically calculated \( j_0 \), we derived the trend of extinction efficiency, for the helix with VP=550nm, for the two circular polarizations, respectively, as shown in figure S4. The figure shows how one polarization excites in similar way two resonances (RCP) while the other one (LCP)
efficiently excites a single resonance, in agreement with scattering spectra of figure 2 in the main text.

**Figure S4.** Calculated trend of extinction power for the two CPs, in the case of a single helix with VP=550nm.
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