Supporting Information

Tuning Dipolar and Multipolar Resonances of Chiral Silicon Nanostructures for Control of Near field Superchirality.

Dominic J.P. Koyroytsaltis-McQuire^{1*}, Rahul Kumar¹, Tamas Javorfi², Giuliano Siligardi², Nikolaj Gadegaard³, Malcolm Kadodwala^{1*}

¹ School of Chemistry, University of Glasgow, Glasgow, G12 8QQ, UK

² Diamond Light Source Ltd., B23 Beamline, Harwell Science and Innovation Campus, Didcot, United Kingdom

³School of Engineering, Rankine Building, University of Glasgow, Glasgow G12 8LT, UK

* Corresponding Authors

E-mail: Malcolm.kadodwala@glasgow.ac.uk

E-mail: <u>d.koyroytsaltis-mcquire.1@research.gla.ac.uk</u>

S1 Sample Heights

A *Bruker Dektak Surface Profiler* was used to verify the heights of the samples of **figure 2**, shown in **S1**.



S1: Scan data from surface profile measurements of the arrays from each sample.

S2 Simulated Enantiomorphic Reflectance

As reflectance is a non-chiroptical measurement, enantiomorphic structures generate equal reflectance spectra from numerical simulations with idealised structures, shown in **S2**.



S2: Simulated reflectance for 160 nm LH and RH structures of single and 4x S structure unit cells.

S3 Modelling of Racemic Array RS

The repeating unit of the RS array contains two RH and two LH structures. The width and depth of the model were therefore doubled, with the height remaining the same, shown in **S3.** To validate the model, these simulations were also performed for 4xLH and 4xRH structures to compare against single structure simulations.



S3: An example of the unit cell of a four-structure simulation, in this case for an RS array.

S4 Equivalent Field Maps Between Single and 4x Structure Simulations

The field map behaviour between the single and 4x S structure simulations show good agreement, shown in **S4**.

		Reflectance Peak A		Reflectance Peak D	
		Single RH	4x RH	Single RH	4x RH
<i>E</i> /Vm ⁻¹	30 20 10 0				
<i>H</i> /Am ⁻¹	0.3 0.2 0.1 0				

S4: A comparison of field distributions between single and 4x S structure simulations for peaks A and D of a 180 nm sample. Plots are taken from the midpoint of the structures.

S5 Idealised vs AFM models



Field maps of idealised and 'real' AFM models shown in **S5** display similar character.

S5: Reflectance comparison between experimental and simulated 'real' AFM and 'ideal' structures with a 180nm sample, for a) y- and b) x-polarised LPL excitation. c) Field comparisons between 'ideal' and 'real' simulated structures for peaks A and D, taken from the midpoint of the structures.

S6 Field Distributions of Enantiomorphic Structures

Given that the numerical simulations of idealised enantiomorphic structures yield identical reflectance spectra. this implies that their field intensities and distributions must be mirror image equivalents, shown in **S6**.



S6: A comparison of field distributions between LH and RH structures for peaks A and D of a 180 nm sample. Plots are taken from the midpoint of the structures.

S7 Circulating Electric Currents of the Silicon Nanostructures

The $|\mathbf{E}|$ and $|\mathbf{H}|$ field distributions are characteristic of silicon nanoparticles, in which the electric field circulates around a magnetic resonance. The circulating electric currents can be observed by generating arrow plots of electric field and material current densities, shown in **S7**.



S7: Electric field and current density arrow plots for the non-periodic resonances of a 180 nm sample, taken from the midpoint of the structures.

S8-10 Mueller Matrix Polarimetry of Silicon Metasurfaces

Full Mueller Matrix Polarimetry was performed on each of the four arrays of a 210 nm sample to determine their optical properties, which are shown in **S8-10**. The optical properties include Circular Dichroism (CD), Linear Dichroism (LD), Circular Birefringence (CB), Linear Birefringence (LB), Linear Dichroism at $\pm 45^{\circ}$ (LD'), Linear Birefringence at $\pm 45^{\circ}$ (LB'), Absorption, G-factor and Transmission.



S8: Optical Properties for LH (black) and RH (red) arrays.



S9: Optical properties of the RA array.



S10: Optical properties of the RS array.