Electronic Supplementary Information for

Controlling Size and Circular Dichroism of Chiral Gold Helicoid

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Supporting figure S1. Circular dichroism and extinction spectra of 180 nm sized 432 helicoid III (A) Circular dichroism spectrum and (B) extinction spectrum of 180 nm sized 432 helicoid III nanoparticle, which are used for calculation of *g*-factor.



Supporting figure S2. Graphical modelling of size-controlled 432 helicoid III

Based on the structural analysis, various sizes of chiral nanoparticles have been modelled. For the 100 nm sized 432 helicoid III, nanoparticle with only chiral motif has been used as the model. For the 120 nm to 180 nm sized 432 helicoid III, nanoparticles have been modified only in the edge length, considering the chiral gap structures remained relatively similar to size variation of nanoparticles.



Supporting figure S3. Simulated circular dichroism of size-controlled 432 helicoid III Circular dichroism of 432 helicoid III with size variation. Legends indicate the edge length of simulated 432 helicoid III nanoparticles.



	120 nm 432 helicoid III	180 nm 432 helicoid III
Edge length (L) (nm)	<mark>122.3 (±8.8)</mark>	<mark>178.6 (±10.5)</mark>
Gap depth (d) (nm)	56.2 (±6.1)	<mark>71.3 (±9.0)</mark>
Gap width (w) (nm)	21.5 (±4.0)	<mark>23.8 (±2.9)</mark>
Gap angle (t) (deg)	44.0 (±8.0)	<mark>49.7 (±8.6)</mark>

Chiral gap geometry

(Counted nanoparticles : 50)

Supporting table S1. Detailed gap structure analysis of size-controlled 432 helicoid III

Statistical analysis of chiral nanoparticles and gap structures from analyzing 50 randomly selected nanoparticles. The structural analysis was conducted based on the SEM images of 120 nm and 180 nm sized 432 helicoid III nanoparticles, according to the defined geometric parameters as shown in the schematic.