

Supporting Information for

# Uniform Chiral Gap Synthesis for High Dissymmetry Factor in Single Plasmonic Gold Nanoparticle

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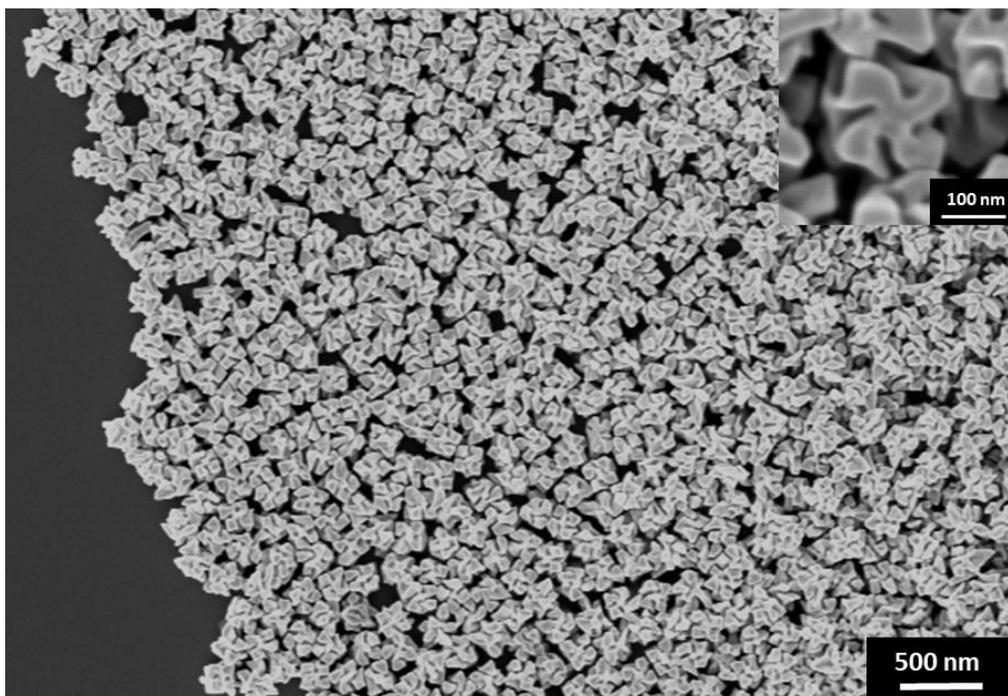
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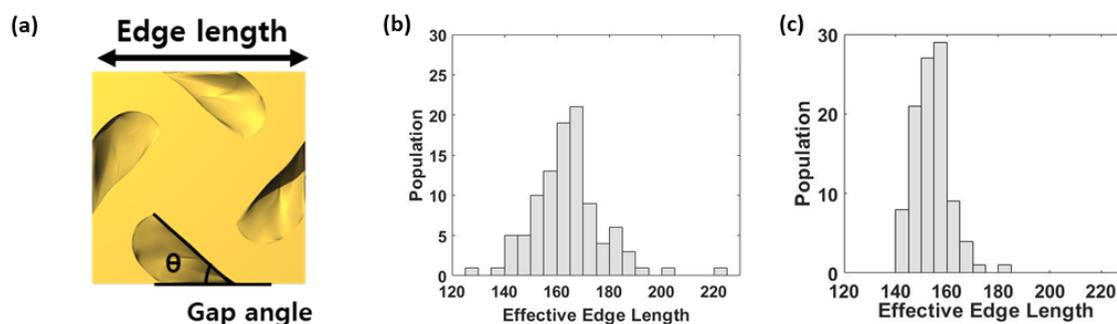
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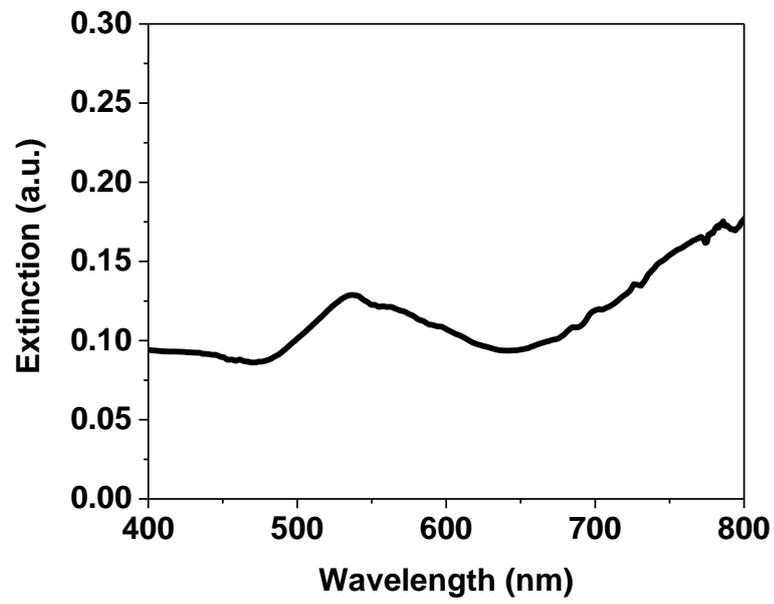
**Figure S2.** Low magnification and high magnification SEM image of 432 helicoid III nanoparticles by conventional synthetic method.



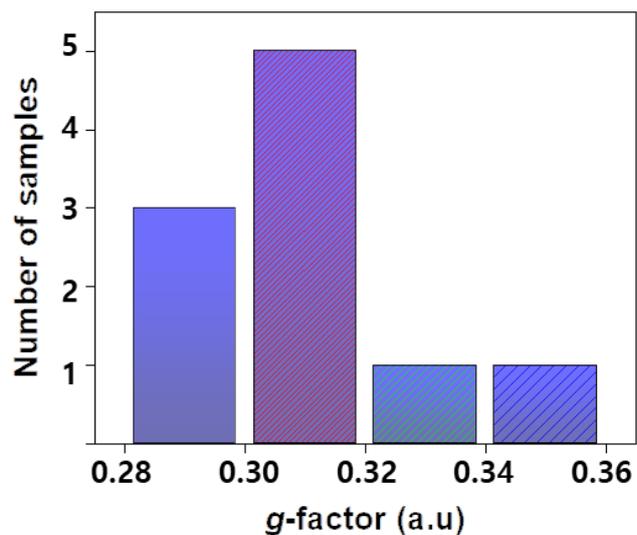
**Figure S2.** Graphical modelling and size distributions of 432 helicoid III. a) Graphical modelling of 432 helicoid III with high uniformity viewed from  $\langle 100 \rangle$  direction. Geometrical parameters of chiral components in uniform 432 helicoid III model are indicated. Effective edge length distribution results of 100 particles synthesized via (b) single-step method and (c) multi-chirality-evolution step method

**Table S1.** Statistical comparison of gap angle between single-step 432 helicoid III and Multi-Chirality-Evolution 432 helicoid III.

	<b>Single-step Helicoid III</b>	<b>Multi-chirality-evolution Helicoid III</b>
<b>Average gap angle (deg°)</b>	<b>51.51 (<math>\pm 8.31</math>)</b>	<b>41.00 (<math>\pm 9.21</math>)</b>



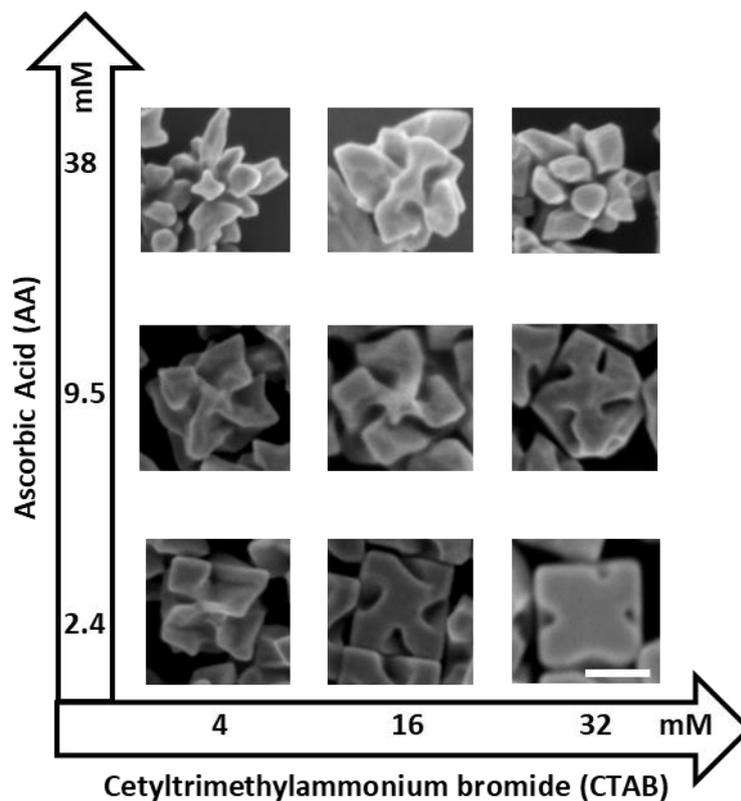
**Figure S3.** Extinction spectrum of 432 helicoid III with high uniformity.



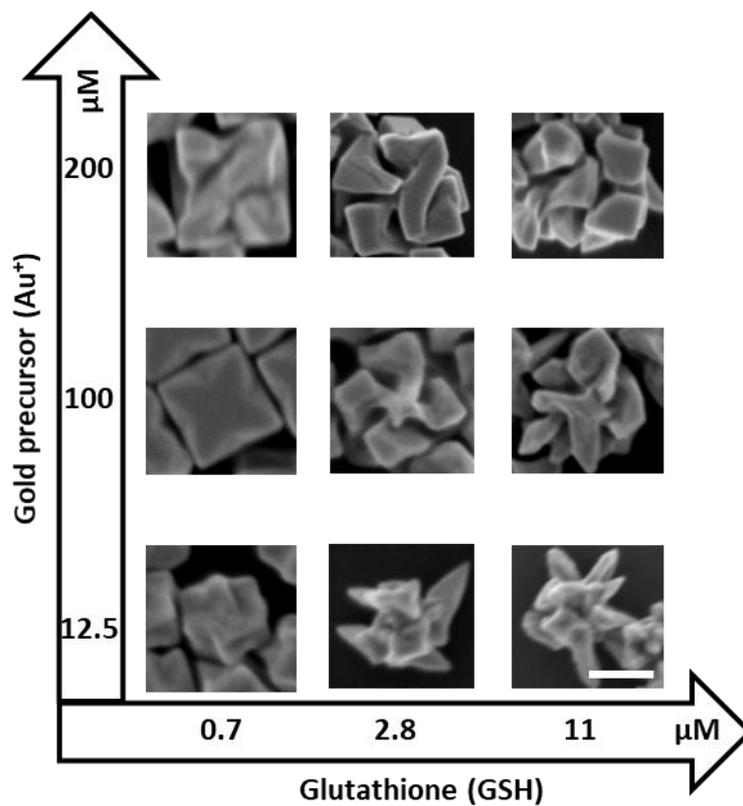
**Figure S4.** Histogram of ten samples'  $g$ -factors adopting multi-step synthesis

**Table S2.** Average FWHD and  $g$ -factor for 432 helicoid III via single-step and multi-step method.

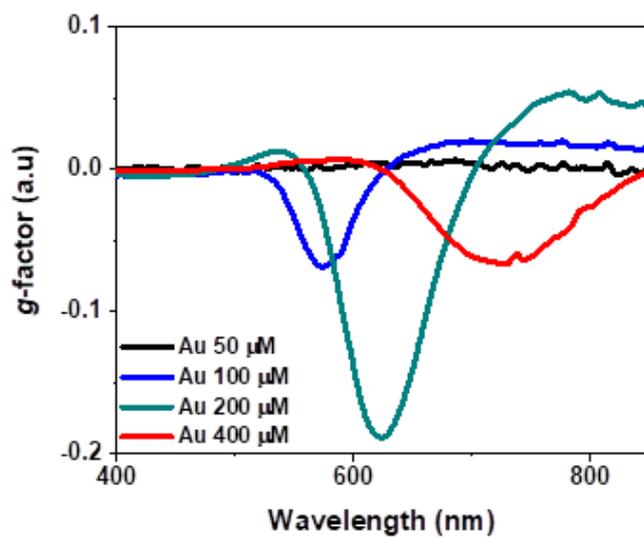
	Single-step Helicoid III	Multi-step Helicoid III
Average FWHD (nm)	74.45	56.50 ( $\pm 2.27$ )
Average $g$ -factor (a.u)	0.20	0.31 ( $\pm 0.02$ )



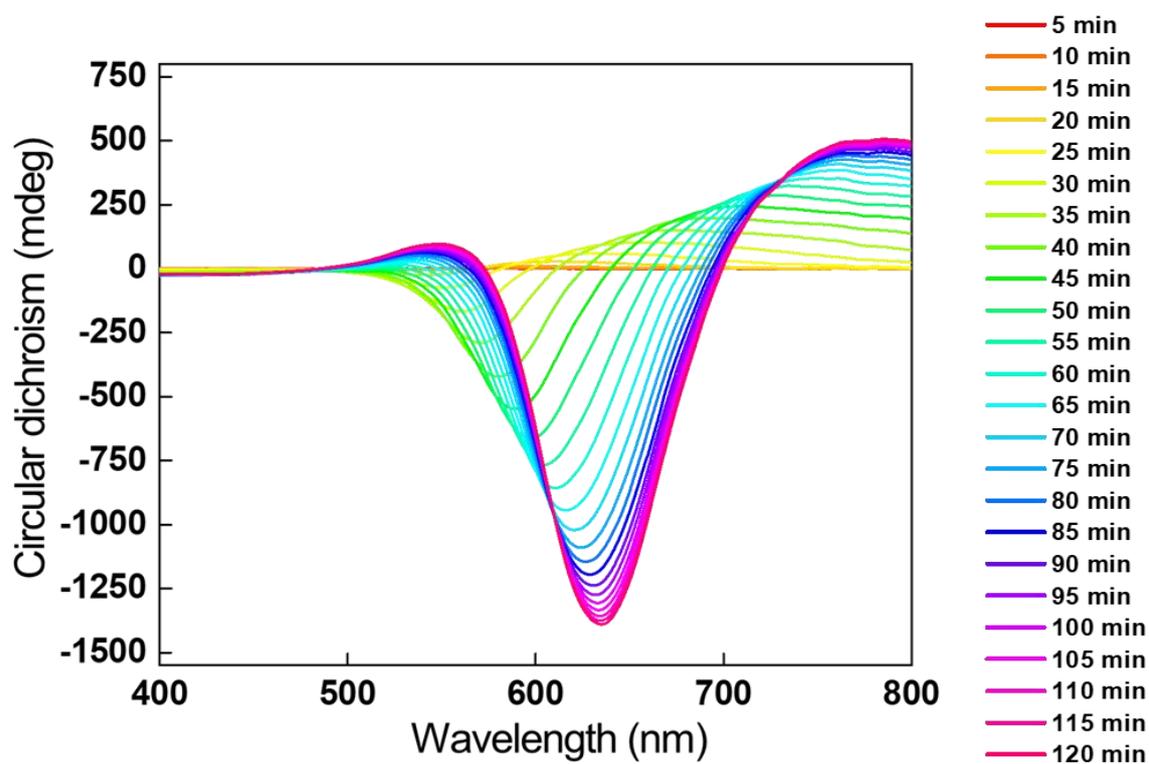
**Figure S5.** Extended chiral morphology diagram respect to change in AA and CTAB concentration during 432 helicoid III synthesis. Increase in CTAB promotes  $\{100\}$  facet stabilization and decrease in gap depth while increase in AA promotes dendritic morphology with increase in gap depth (Scale bar =100 nm).



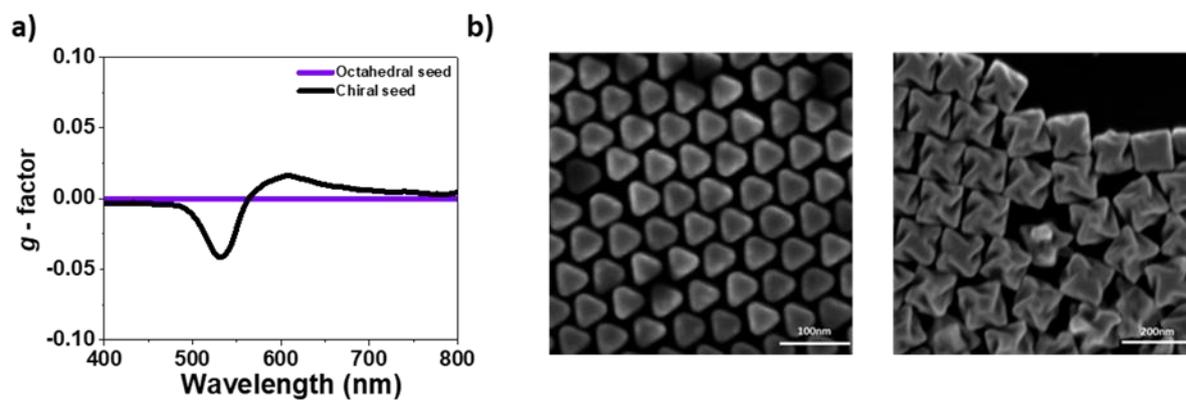
**Figure S6.** Extended chiral morphology diagram respect to change in Gold precursor and GSH concentration during 432 helicoid III synthesis. Increase in GSH promotes dendritic morphology and while increase in Gold precursor promotes achiral morphology with increase in nanoparticle size. (Scale bar =100 nm).



**Figure S7.** *g*-factor spectra of 432 helicoid III for different amount of Au precursor in the growth solution All particles synthesized by single step method.



**Figure S8.** Time variant CD measurement of 432 helicoid III nanoparticle during its growth process. Chirality progressively increases with shift of resonance to larger wavelength.



**Figure S9.** Chiroptic responses and SEM images of seed nanoparticles. a) Chiroptic response of octahedral seed nanoparticles and intermediate chiral seed nanoparticles. b) Corresponding SEM images of octahedral seed nanoparticles and