Supporting Information

Formation of Free-Standing Supercrystals from the Assembly of Polyhedral Gold Nanocrystals by Surfactant Diffusion in the Solution

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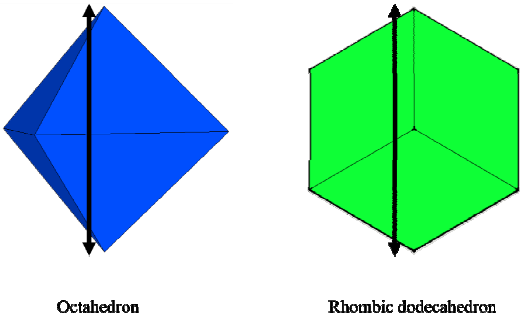

Figure S1. Drawings of an octahedron and a rhombic dodecahedron. The arrows indicate the measured particle sizes.

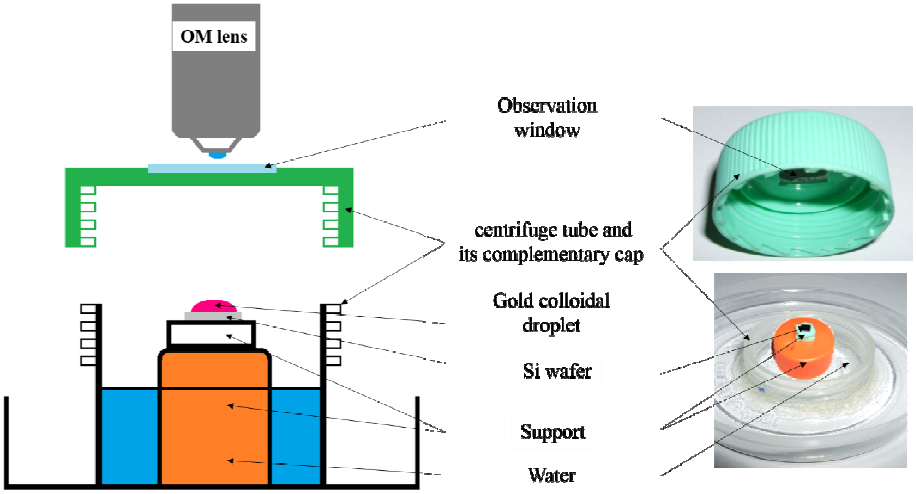

Figure S2. Illustration of the setup designed to reduce the Au colloid droplet evaporation rate during optical microscopy observation. The specimen chamber is made from a centrifuge tube by cutting its lower part and gluing the top portion to a plastic plate underneath. The cap has an opening with a microscope cover slide attached for optical observation. The sample chamber is designed to minimize the distance between the specimen and the cover glass for high magnification observation. The cap was rotated to a certain position such that the colloidal droplet does not touch the cover glass, and the working distance is still large enough for microscopic observation. A camara is attached to the microscope for video recording.

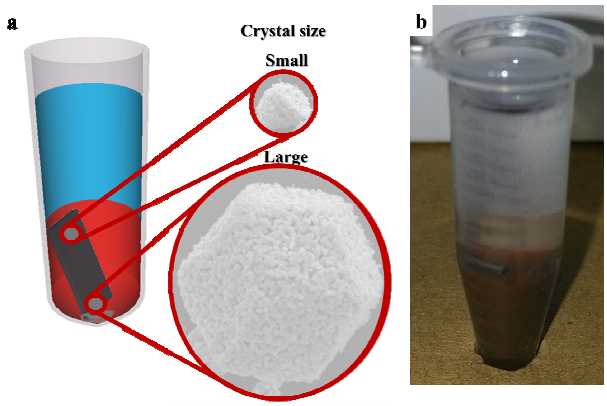

Figure S3. (a) Illustration of the method used to grow small and large supercrystals over a substrate. (b) Photograph showing an Eppendorf tube loaded with a slanted Si substrate. The lower layer is the gold colloidal solution, while the upper transparent layer is the CTAC solution.

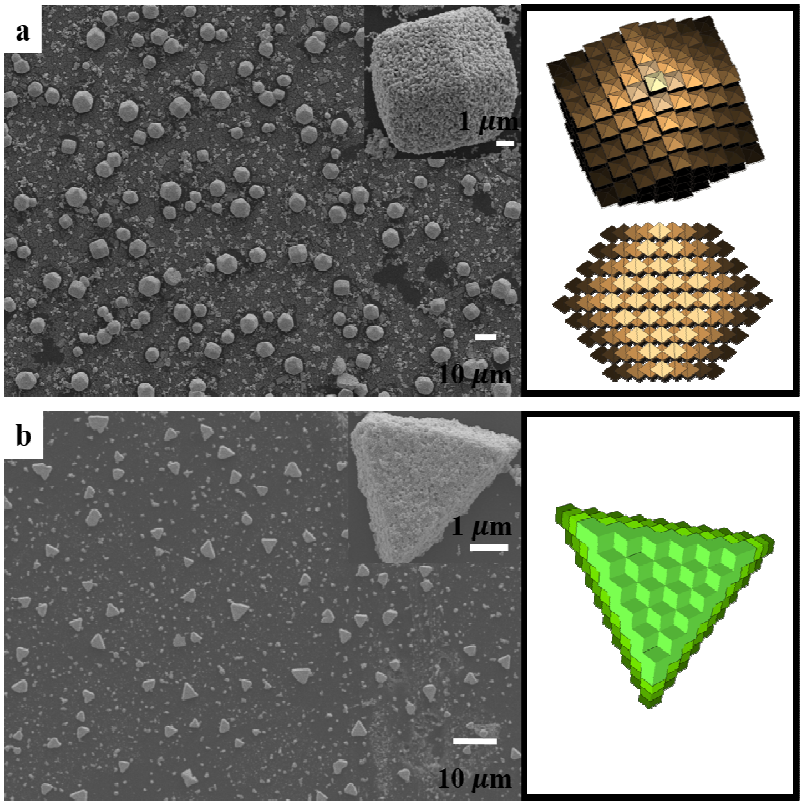

Figure S4. SEM images of supercrystals assembled by gold (a) octahedra and (b) rhombic dodecahedra at 90 ºC by the droplet evaporation approach. Insets give the enlarged images of single supercrystals. Schematic drawings of the nanocrystal packing structures are also shown. Two viewing orientations are given for the rhombic dodecahedral supercrystals assembled from octahedral particles.

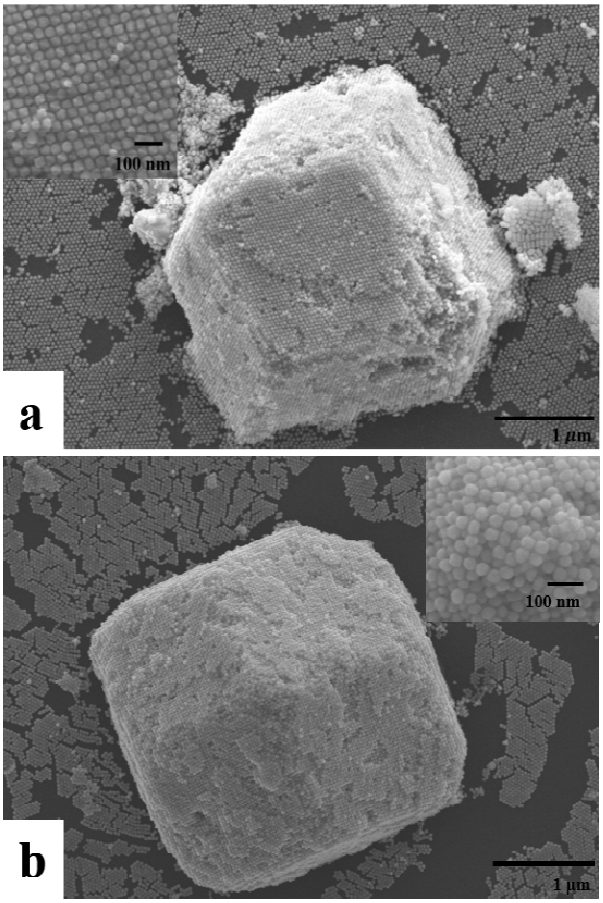

Figure S5. (a, b) SEM images of a single supercrystal formed through assembly of Au octahedra on an evaporating substrate at 90 ºC. No interparticle fusion can be observed. Inset images show clear octahedral particles without fusion. Disk-like byproduct particles also form their own assembled structure, confirming that the particle assembly process is strongly shape-guided.

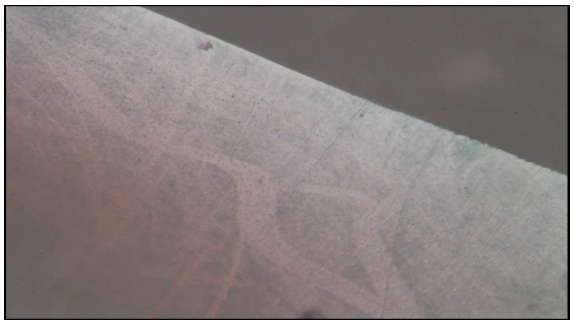

Figure S6. Optical image of an edge portion of the substrate during the supercrystal formation process. Supercrystals are formed first near the edge of the droplet or substrate and this region reveals more of the color of the substrate. At this point the central region of the droplet still contains suspended gold rhombic dodecahedra, so the lower left region maintains a purplish red color of Au colloidal solution.

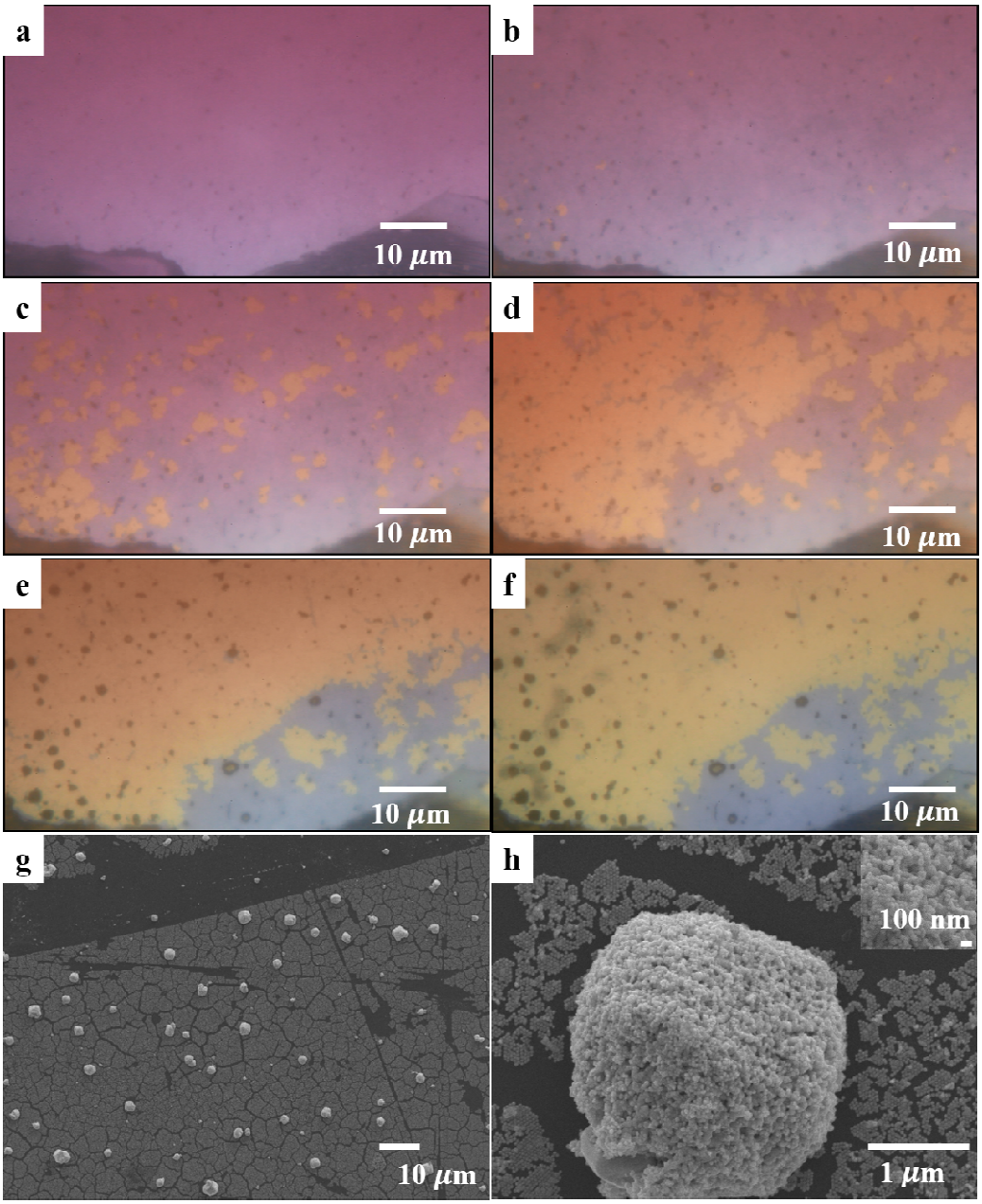

Figure S7. (a–f) Optical microscopy images of the supercrystal formation process taken at (a) 3, (b) 12, (c) 13, (d) 15, (e) 18, and (f) 20 min in the droplet evaporation process. Gold octahedra were used as the building blocks. The yellowish brown color comes from a monolayer film of assembled octahedra. (g, h) Large-area and enlarged SEM images of the synthesized supercrystals. Inset shows a close-up view of the assembled octahedra. Some particle fusion may have occurred, so the particle shape is more difficult to distinguish.

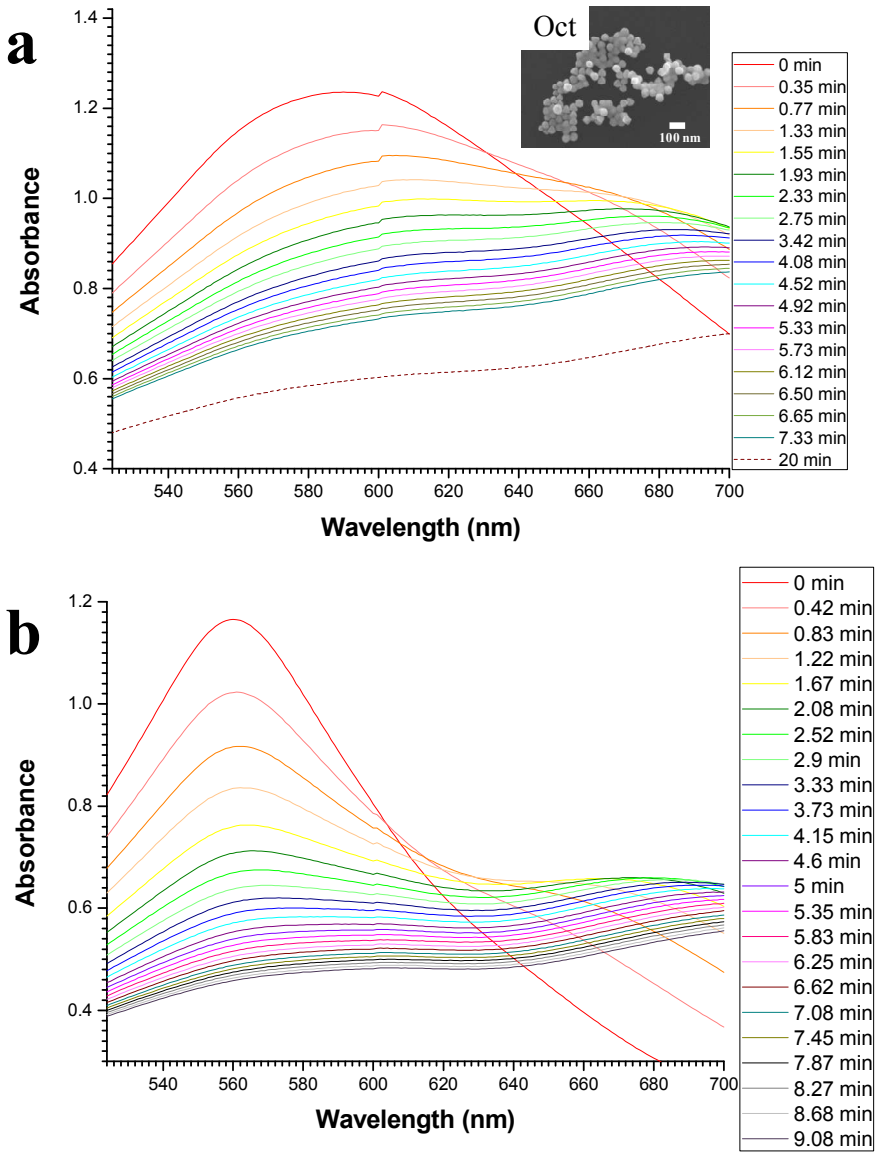

Figure S8. Time-dependent UV–vis absorption spectra of octahedral Au nanocrystals dispersed in (a) 0.7 and (b) 0.3 M CTAC solutions. Inset shows SEM image of the resulting aggregated octahedra.