

Supporting Information

Low-entropy lattices engineered through bridged DNA origami frames

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Materials

M13mp18 DNA was purchased from Bayou Biolabs, America. All short oligonucleotides were purchased from Sangon Biotech Co. Ltd (Shanghai, China). 10 nm AuNPs were purchased from Ted Pella Inc. Carbon-coated copper grids were purchased from Beijing Zhongjingkeyi Technology Co. Ltd (Beijing, China).

Design and synthesis of DNA origami frames

Octahedron, cube, hexagonal bipyramid and elongated octahedron DNA origami frames were designed by caDNAno software (downloaded from <http://cadnano.org/>). In the design, each edge of these frames was composed of a six-helix bundle. For the octahedral and cubic frames, the length of all the edges is 88 base pairs. For each hexagonal bipyramid, the bottom edges of the hexagonal pyramid are 46 base pairs in length, and the length of side edges was 67 base pairs. For an elongated octahedral frame, the length of four bottom edges is 88 base pairs that is equal to the octahedron's, and its side edges are 109 base pairs in length.

DNA origami frames were folded by mixing 10 nM M13mp18 scaffold DNA, 100 nM of each designed staple oligonucleotide (including a certain number of capture DNAs for encaging 10 nm AuNPs inside) in a buffer solution containing 12.5 mM magnesium acetate, 1 mM EDTA and 40 mM tris acetate with a pH around 8.3. The mixed solution was then cooled by a gradient according to the annealing procedure below:

1. 90 °C 25 sec
2. 90 °C ~ 85 °C -0.1 °C / 5 sec
3. 85 °C ~ 65 °C -0.1 °C / 30 sec
4. 65 °C ~ 40 °C -0.1 °C / 4 min
5. 40 °C ~ 32 °C -0.1 °C / 2 min
6. 32 °C ~ 20 °C -0.1 °C / 1 min
7. Hold at 20 °C

Prepared DNA origami frames were stored at room temperature.

DNA functionalization of gold nanoparticles

200 μM of 3'-thiolated oligonucleotides aqueous solution were incubated with tris-(2-carboxyethyl) phosphine (TCEP) in a ratio of 1:100 (DNA: TCEP). The mixed solution was kept in an ice bath for 1.5 hours, and then purified using a size exclusion column (G-25, GE Healthcare) to remove redundant TCEP. Treated thiolated oligonucleotides were mixed with 10 nm spherical gold nanoparticles with a ratio of 300:1, followed by buffering the solution to obtain 10 mM phosphate buffer (pH 7) after 1.5 hours. In the subsequent salt-aging process, 2

M NaCl solution was slowly added into the solution in five steps to reach a final NaCl concentration of 0.3 M. The solution was aged at room temperature for 18 hours. Excess oligonucleotides were then removed by centrifuging (20,000 rcf, 1h) the solution 4 times and resuspending with the washing buffer containing 0.1 M NaCl and 10 mM phosphate. The concentration of AuNPs was measured by UV-vis spectrometer and calculated with Lambert-Beer law.

Encaging AuNPs inside DNA origami frames for conjugated monomers

In order to insert AuNPs inside the DNA origami frames, functionalized AuNPs and prepared DNA origami frames were mixed at a ratio of 1.2:1. The mixed solution was then cooling from 50 °C to 20 °C at a rate of 0.6 °C h⁻¹ to obtain a high yield.

Fabrication of 3D lattices and 2D planar structures

According to the design of each system, corresponding bridging DNA strands and functionalized AuNPs were added into the prepared DNA origami frame solution. The molar ratio between the frames, bridged DNA strands and AuNPs was 1:1:1.2. To reduce the effect of dilution, a few of 125 mM magnesium acetate solution was added to adjust the magnesium salt concentration to original 12.5 mM. The mixed solution was then slowly cooled by a gradient according to the annealing procedure below:

1. 20 °C ~ 50 °C +1 °C / min
2. 50 °C ~ 20 °C -0.2 °C / 1 h
3. Hold at 20 °C

This procedure was repeated another two times to produce the final sample. Prepared 3D lattices and 2D planar structures were stored at 4 °C.

Silicification of DNA frameworks

The supernatant of the prepared 3D lattice solution was pipetted out slowly without disturbing bottom aggregates, followed by buffering with a solution containing 7.0 mM magnesium acetate, 1 mM EDTA and 40 mM tris acetate to remove free staple DNAs and AuNPs. 0.79 µL of 10 % (v/v) N-trimethoxysilylpropyl-N,N,N-trimethylammonium chloride (TMAPS) methanol solution was then mixed with the lattice solution and shaken at 20 °C for 20 min. Subsequently, 0.90 µL 10% (v/v) tetraethoxysilane (TEOS) methanol solution was added and kept shaking at 20 °C for 30 min.¹ After the shaking, the solution was allowed to stand for about 12 hours. The supernatant of the final solution was pipetted out and the aggregates were washed slowly with ultrapure water for several times.

TEM sample preparation and imaging

The sample solution of 5 μL was dropped onto a glow-discharged carbon-coated grid and deposited for 4 minutes. The droplet was then wicked away by a piece of filter paper along the edge of the grid. 5 μL of 2% (w/v) uranyl acetate aqueous solution was then dropped onto the carbon-coated grid. After staining for 8 seconds, the excess staining solution was wicked away in the same manner and dried in the air. The negative-stained sample was imaged by a JEM-2800 TEM operating at 200 kV.

SEM of siliconized samples

The silicon slice, as the SEM substrate, was carefully cleaned with ethanol and water in advance. 6 μL siliconized sample solution was dropped onto the silicon slice and dried under an infrared light. The sample was then imaged on the Zeiss Ultra Plus FE-SEM.

SAXS experimental methods

The SAXS measurements were conducted at the beamline BL19U2 at Shanghai Synchrotron Radiation Facility (SSRF). 50 μL of the sample solution was transferred to a glass capillary with an inner diameter of 1 mm. The capillary was sealed with wax to maintain the liquid phase and the aggregates sedimentated at its bottom. The 2D scattering data were collected on a Dectris Pilatus 2 M pixel-array detector with 12 keV (wavelength $\lambda = 0.932 \text{ \AA}$) incident X-rays.

Area images were integrated into a 1D $I(q)$ scattering curve according to the function $q = 4 \frac{\pi}{\lambda} \sin\left(\frac{\theta}{2}\right)$ of the scattering vector q , where θ is the scattering angle.² The structure factor $S(q)$ was obtained by dividing $I(q)$ by the corresponding particle form factor $P(q)$.

The geometric models for polyhedral DNA origami frames

The polyhedral DNA origami frame geometric models are shown in Figure S2, and described using d_1, d_2, L_1, L_2, L_3 and L_4 as the width and length of the polyhedral edges. The exact values are calculated based on the following constants: the helix width of DNA is 2.0 nm and the distance between adjacent base pairs is 0.34 nm. Stereo modeling shows the simplified frame geometry accurate to each double helix (represented by the cylinder). Potential deformations are not involved in these models.

Close-packed models and bridge pattern models for 3D lattices

According to the pre-designed arrangements of polyhedral frames, we constructed a series of close-packed 3D lattice models with no gaps (Figures S15-S17 and S34). Considering the

steric hindrance caused by adjacent structures, the vertices of DNA origami frames just touch each other. At bridged junctions, both the octahedral and cubic frames are illustrated with a pair of vertices as shown in Figures S6 and S8. Due to the difference in crystal growth directions, the hexagonal bipyramid and binary systems exhibit two non-identical crystallographic axis directions respectively (Figures S7 and S31). The lattice parameter values of these models calculated according to the space analytic geometry are as follows:

Geometric constants involved:

$$\mathbf{d}_1 = 5.5 \text{ nm}; \mathbf{d}_2 = 6.0 \text{ nm};$$

$$\mathbf{L}_1 = 29.9 \text{ nm}; \mathbf{L}_2 = 22.8 \text{ nm}; \mathbf{L}_3 = 15.7 \text{ nm}; \mathbf{L}_4 = 37.1 \text{ nm}$$

Octahedron model:

$$\mathbf{a} = \mathbf{b} = \mathbf{c} = \sqrt{2}(\mathbf{L}_1 + \mathbf{d}_1) = 50.1 \text{ nm}$$

Cube model:

$$\mathbf{a} = \mathbf{b} = \mathbf{c} = 2\left(\mathbf{L}_1 + \frac{\mathbf{d}_2}{2}\right) = 65.8 \text{ nm}$$

Hexagonal bipyramid model:

$$\mathbf{a} = \mathbf{b} = 2(\mathbf{L}_3 + \mathbf{d}_1) = 42.4 \text{ nm}$$

$$\mathbf{c} = 2\sqrt{\mathbf{L}_2^2 - \mathbf{L}_3^2} + \sqrt{2}\mathbf{d}_2 = 41.6 \text{ nm}$$

Binary system model:

$$\mathbf{a} = \mathbf{b} = \sqrt{2}(\mathbf{L}_1 + \mathbf{d}_1) = 50.1 \text{ nm}$$

$$\mathbf{c} = \frac{\sqrt{2}(\mathbf{L}_1 + \mathbf{d}_1)}{2} + \sqrt{(\mathbf{L}_4 + \mathbf{d}_1)^2 - \left(\frac{\sqrt{2}(\mathbf{L}_1 + \mathbf{d}_1)}{2}\right)^2} = 59.4 \text{ nm}$$

SAXS model fitting and lattice parameter calculation

The theoretical scattering data of the lattice structures was calculated by using PowderCell software. For the simple cubic and body-centered cubic lattices with defined structural parameters, we calibrated the first SAXS peak (q_1) of the experimental data with the first peak in theoretical spectrums to compare the peak positions and the intensity ratio. We then calculated the experimental lattice parameters in cubic lattices ($\mathbf{a} = \mathbf{b} = \mathbf{c}$) based on the peak position of q_1 . The three lattice parameters of the simple hexagonal and tetragonal structures are not equal ($\mathbf{a} = \mathbf{b} \neq \mathbf{c}$). We first calculated their experimental values using the first two SAXS peaks (q_1 and q_2), and then performed the calibration. The lattice parameter values calculated from the experimental data are as follows:

Simple cubic lattice, octahedron:

$$a = b = c = \frac{2\pi}{q_1} = 50.0 \text{ nm} (q_1 = 0.1258 \text{ nm}^{-1})$$

Body-centered cubic lattice, cube:

$$a = b = c = \frac{2\sqrt{2}\pi}{q_1} = 65.9 \text{ nm} (q_1 = 0.1349 \text{ nm}^{-1})$$

Simple hexagonal lattice, hexagonal bipyramid:

$$a = b = \frac{4\sqrt{3}\pi}{3q_2} = 43.0 \text{ nm} (q_2 = 0.1687 \text{ nm}^{-1})$$

$$c = \frac{2\pi}{q_1} = 42.1 \text{ nm} (q_1 = 0.1494 \text{ nm}^{-1})$$

Simple tetragonal lattice, binary system:

$$a = b = \frac{2\pi}{q_2} = 50.0 \text{ nm} (q_2 = 0.1258 \text{ nm}^{-1})$$

$$c = \frac{2\pi}{q_1} = 58.4 \text{ nm} (q_1 = 0.1076 \text{ nm}^{-1})$$

DNA Sequences

a. Staple strands of octahedral DNA origami frame

Oct-staple-1	TGTAGCATTCCAACGTTAGTAAATGAAGTGCCGCGCCACCCT
Oct-staple-2	CTTAAACAGCTTATATATTCGGTCGCTTGATGGGGAACAAGA
Oct-staple-3	AATAGCAATAGCACCAGAAGGAAACCTAAAGCCACTGGTAAT
Oct-staple-4	AGCTTTCATCAACGGATTGACCGTAAAATCGTATAATATTTT
Oct-staple-5	CTTCATCAAGAGAAATCAACGTAACAGAGATTTGTCAATCAT
Oct-staple-6	AAAGATTCATCAGGAATTACGAGGCATGCTCATCCTTATGCG
Oct-staple-7	ATAAATCATAATAAATCGGTTGTAAGTGTGCTGGCATGCCTG
Oct-staple-8	CAACGCTCAACAGCAGAGGCATTTTCAATCCAATGATAAATA
Oct-staple-9	AAACGAAAGAGGGCGAAACAAAGTACTGACTATATTCGAGCT
Oct-staple-10	AGAGCCTAATTTGATTTTTTTGTTTAAATCCTGAAATAAAGAA
Oct-staple-11	GACAGGAGGTTGAAACAAATAAATCCGCCCCCTCCGCCACCC
Oct-staple-12	TCATATGGTTTACGATTGAGGGAGGGAAACGCAATACATACA
Oct-staple-13	GGTAGCTATTTTAGAGAATCGATGAAAACATTAAATGTGTAG
Oct-staple-14	GAAACATGAAAGCTCAGTACCAGGCGAAAAATGCTGAACAAA
Oct-staple-15	ATCAAAATCATATATGTAAATGCTGAACAAACACTTGCTTCT
Oct-staple-16	AACGGGTATTAAGGAATCATTACCGCCAGTAATTCAACAATA
Oct-staple-17	ACTGTTGGGAAGCAGCTGGCGAAAGGATAGGTCAAGATCGCA
Oct-staple-18	CAGAATCAAGTTTCGGCATTTCGGTTAAATATATCACCAGT
Oct-staple-19	GCTCACAATTCCGTGAGCTAACTCACTGGAAGTAATGGTCAA

Oct-staple-20 TGATTGCTTTGAGCAAAAGAAGATGAAATAGCAGAGGTTTTG
 Oct-staple-21 GGCCCTGAGAGAAGCAGGCGAAAATCATTGCGTAGAGGCGGT
 Oct-staple-22 TTTGCGGATGGCCAACTAAAGTACGGGCTTGCAGCTACAGAG
 Oct-staple-23 CAAATGCTTTAAAAAATCAGGTCTTTAAGAGCAGCCAGAGGG
 Oct-staple-24 TTTGCGGAACAATGGCAATTCATCAATCTGTATAATAATTTT
 Oct-staple-25 GTCACCAGAGCCATGGTGAATTATCACCAATCAGAAAAGCCT
 Oct-staple-26 GGACAGAGTTACTTTGTGCGAAATCCGCGTGTATCACCGTACG
 Oct-staple-27 CAACATGATTTACGAGCATGGAATAAGTAAGACGACAATAAA
 Oct-staple-28 AACCAGACGCTACGTTAATAAAACGAACATACCACATTCAGG
 Oct-staple-29 TGACCTACTAGAAAAAGCCCCAGGCAAAGCAATTCATCTTC
 Oct-staple-30 TGCCGGAAGGGGACTCGTAACCGTGCATTATATTTTAGTTCT
 Oct-staple-31 AGAACCCCAAATCACCATCTGCGGAATCGAATAAAAATTTTT
 Oct-staple-32 GCTCCATTGTGTACCGTAACACTGAGTTAGTTAGCGTAACCT
 Oct-staple-33 AGTACCGAATAGGAACCCAAACGGTGTAACTCAGGAGGTTT
 Oct-staple-34 CAGTTTGAATGTTTAGTATCATATGCGTAGAATCGCCATAGC
 Oct-staple-35 AAGATTGTTTTTAAACCAAGAAACCATCGACCCAAAAACAGG
 Oct-staple-36 TCAGAGCGCCACCACATAATCAAAAATCAGAACGAGTAGTATG
 Oct-staple-37 GATGGTTGGGAAGAAAAATCCACCAGAAATAATTGGGCTTGA
 Oct-staple-38 CTCCTAACGTAGAAACCAATCAATAATTCATCGAGAACAGA
 Oct-staple-39 AGACACCTTACGCAGAACTGGCATGATTTTCTGTCCAGACAA
 Oct-staple-40 GCCAGCTAGGCGATAGCTTAGATTAAGACCTTTTTAACCTGT
 Oct-staple-41 CCGACTTATTAGGAACGCCATCAAAAATGAGTAACAACCCCA
 Oct-staple-42 GTCCAATAGCGAGAACCAGACGACGATATTCAACGCAAGGGA
 Oct-staple-43 CCAAATACAATATGATATTCAACCGTTAGGCTATCAGGTAA
 Oct-staple-44 AACAGTACTTGAAAACATATGAGACGGGTCTTTTTTAATGGA
 Oct-staple-45 TTTACCGCATTAAAGTCGGGAAACCTGATTTGAATTACCCA
 Oct-staple-46 GAGAATAGAGCCTTACCGTCTATCAAATGGAGCGGAATTAGA
 Oct-staple-47 ATAATTAATTTAAAAAACTTTTTCAAACTTTTAACAACGCC
 Oct-staple-48 GCACCCAGCGTTTTTTATCCGGTATTCTAGGCGAATTATTCA
 Oct-staple-49 GGAAGCGCCACAAACAGTTAATGCCCGACTCCTCAAGATA
 Oct-staple-50 GTTTGCCTATTACAGGCAGGTCAGACGCCACCACACCACC
 Oct-staple-51 CGCGAGCTTAGTTTTTCCAATTCTGCGCAAGTGTAAGCCT
 Oct-staple-52 AGAAGCAACCAAGCCAAAAGAATACACTAATGCCAAAACCTCC
 Oct-staple-53 ATTAAGTATAAAGCGGCAAGGCAAAGAACTAATAGGGTACC
 Oct-staple-54 CAGTGCCTACATGGGAATTTACCGTTCCACAAGTAAGCAGAT
 Oct-staple-55 ATAAGGCGCCAAAAGTTGAGATTTAGGATAACGGACCAGTCA
 Oct-staple-56 TGCTAAACAGATGAAGAAACCACCAGAATTTAAAAAAAGGCT
 Oct-staple-57 CAGCCTTGGTTTTGTATTAAGAGGCTGACTGCCTATATCAGA
 Oct-staple-58 CGGAATAATTC AACCCAGCGCCAAAGACTTATTTTAACGCAA
 Oct-staple-59 CGCCTGAATTACCCTAATCTTGACAAGACAGACCATGAAAGA

Oct-staple-60 ACGCGAGGCTACAACAGTACCTTTTACAAATCGCGCAGAGAA
 Oct-staple-61 CAGCGAACATTAAAAGAGAGTACCTTTACTGAATATAATGAA
 Oct-staple-62 GGACGTTTAATTTTCGACGAGAAACACCACCACTAATGCAGAT
 Oct-staple-63 AAAGCGCCAAAGTTTATCTTACCGAAGCCCAATAATGAGTAA
 Oct-staple-64 GAGCTCGTTGTAAACGCCAGGGTTTTCCAAAGCAATAAAGCC
 Oct-staple-65 AATTATTGTTTTTCATGCCTTTAGCGTCAGATAGCACGGAAAC
 Oct-staple-66 AAGTTTCAGACAGCCGGGATCGTCACCCTTCTGTAGCTCAAC
 Oct-staple-67 ACAAAGAAATTTAGGTAGGGCTTAATTGTATACAACGGAATC
 Oct-staple-68 AACAAAATAACTAGGTCTGAGAGACTACGCTGAGTTTCCCT
 Oct-staple-69 CATAACCTAAATCAACAGTTCAGAAAACGTCATAAGGATAGC
 Oct-staple-70 CACGACGAATTCGTGTGGCATCAATTCTTTAGCAAAATTACG
 Oct-staple-71 CCTACCAACAGTAATTTTATCCTGAATCAAACAGCCATATGA
 Oct-staple-72 GATTATAAAGAAACGCCAGTTACAAAATTTACCAACGTCAGA
 Oct-staple-73 AGTAGATTGAAAAGAATCATGGTCATAGCCGGAAGCATAAGT
 Oct-staple-74 TAGAATCCATAAATCATTTAACAATTTCTCCCGGCTTAGGTT
 Oct-staple-75 AAAGGCCAAATATGTTAGAGCTTAATTGATTGCTCCATGAGG
 Oct-staple-76 CCAAAGGAAAGGACAACAGTTTCAGCGAATCATCATATTCC
 Oct-staple-77 GAAATCGATAACCGGATACCGATAGTTGTATCAGCTCCAACG
 Oct-staple-78 TGAATATTATCAAATAATGGAAGGGTTAATATTTATCCCAA
 Oct-staple-79 GAGGAAGCAGGATTCGGGTAAAATACGTAAAACACCCCCAG
 Oct-staple-80 GGTGATTTTTCCAGCAGACAGCCCTCATTTCGTCACGGGATAG
 Oct-staple-81 CAAGCCCCACCCTTAGCCCGGAATAGGACGATCTAAAGTTT
 Oct-staple-82 TGTAGATATTACGCGGCGATCGGTGCGGGCGCCATCTTCTGG
 Oct-staple-83 CATCCTATTCAGCTAAAAGGTAAAGTAAAAGCAAGCCGTTT
 Oct-staple-84 CAGCTCATATAAGCGTACCCCGGTTGATGTGTCTGGATTCTCC
 Oct-staple-85 CATGTCACAAACGGCATTAAATGTGAGCAATTCGCGTTAAAT
 Oct-staple-86 AGCGTCACGTATAAGAATTGAGTTAAGCCCTTTTTAAGAAAG
 Oct-staple-87 TATAAAGCATCGTAACCAAGTACCGCACCGGCTGTAATATCC
 Oct-staple-88 ATAGCCCGCGAAAATAATTGTATCGGTTCCGCCACAATGAGT
 Oct-staple-89 AGACAGTTCATATAGGAGAAGCCTTTATAACATTGCCTGAGA
 Oct-staple-90 AACAGGTCCCGAAATTGCATCAAAAAGATCTTTGATCATCAG
 Oct-staple-91 ACTGCCCTTGCCCCGTTGCAGCAAGCGGCAACAGCTTTTTTCT
 Oct-staple-92 TCAAAGGGAGATAGCCCTTATAAATCAAGACAACAACCATCG
 Oct-staple-93 GTAATACGCAAACATGAGAGATCTACAACACTAGCTGAGGCCGG
 Oct-staple-94 GAGATAACATTAGAAGAATAACATAAAAAGGAAGGATTAGGA
 Oct-staple-95 CAGATATTACCTGAATACCAAGTTACAATCGGGAGCTATTTT
 Oct-staple-96 CATATAACTAATGAACACAACATACGAGCTGTTTCTTTGGGG
 Oct-staple-97 ATGTTTTGCTTTTGATCGGAACGAGGGTACTTTTTCTTTTGA
 Oct-staple-98 GGGGTGCCAGTTGAGACCATTAGATACAATTTTCACTGTGTG
 Oct-staple-99 CTTCGCTGGGCGCAGACGACAGTATCGGGGCACCGTCGCCAT

Oct-staple-100 TCAGAGCTGGGTAAACGACGGCCAGTGCGATCCCCGTAGTAG
 Oct-staple-101 TTAGCGGTACAGAGCGGGAGAATTAAGTTCGCTAATTTTCGGA
 Oct-staple-102 GATATTCTAAATTGAGCCGGAACGAGGCCCAACTTGGCGCAT
 Oct-staple-103 TGTCGTCATAAGTACAGAACC GCCACCCATTTTCACAGTACA
 Oct-staple-104 CGATTATAAGCGGAGACTTCAAATATCGCGGAAGCCTACGAA
 Oct-staple-105 AACATGTACGCGAGTGGTTTTGAAATACCTAAACACATTCTTA
 Oct-staple-106 GTCTGGATTTTTCGTTTTAAATGCAATGGTGAGAAATAAATT
 Oct-staple-107 GCCTTGAATCTTTTCCGGAACCGCCTCCAGAGCCCAGAGCC
 Oct-staple-108 CGCTGGTGCTTTTCTGAATCGGCCAACGAGGGTGGTGATTGC
 Oct-staple-109 TGATTATCAACTTTACAATAAGGAATCCAAAAAGTTTGAG
 Oct-staple-110 ACATAACTTGCCCTAACTTTAATCATTGCATTATAACAACAT
 Oct-staple-111 GTAGCGCCATTAAATTGGGAATTAGAGCGCAAGGCGCACCGT
 Oct-staple-112 TTATTTTACCGACAATGCAGAACGCGCGAAAAATCTTTCTT
 Oct-staple-113 TTTCAATAGAAGGCAGCGAACCTCCCGATTAGTTGAAACAAT
 Oct-staple-114 GGGCGACCCCAAAGTATGTTAGCAAATAAAAGAGTCACAA
 Oct-staple-115 AGCCGAAAGTCTCTCTTTTGATGATACAAGTGCCTTAAGAGC
 Oct-staple-116 GTGGGAAATCATATAAATATTTAAATTGAATTTTGTCTGGC
 Oct-staple-117 CCCACGCGCAAATGGTTGAGTGTTCGTGGACTTGCTTT
 Oct-staple-118 ATGACCACTCGTTTGGCTTTTGCAAAAGTTAGACTATATTCA
 Oct-staple-119 TCCAAATCTTCTGAATTATTTGCACGTAGGTTAACGCTAAC
 Oct-staple-120 GGGTTATTTAATTACAATATATGTGAGTAATTAATAAGAGTC
 Oct-capture-1 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGGGGTGCCAG
 TTGAGACCATTAGATACAATTTTCACTGTGTG
 Oct-capture-2 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTTCAGAGCTGG
 GTAAACGACGGCCAGTGCGATCCCCGTAGTAG
 Oct-capture-3 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTTTCAGCGGTAC
 AGAGCGGGAGAATTAAGTTCGCTAATTTTCGGA
 Oct-capture-4 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTTTCAGATTATCAA
 CTTTACAATAAAGGAATCCAAAAAGTTTGAG
 Oct-capture-5 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTAGCGCCAT
 TAAATTGGGAATTAGAGCGCAAGGCGCACCGT
 Oct-capture-6 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTAGCCGAAAGT
 CTCTCTTTTGATGATACAAGTGCCTTAAGAGC
 Oct-capture-7 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTGGGAAATC
 ATATAAATATTTAAATTGAATTTTGTCTGGC
 Oct-capture-8 ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTCCCACGCGCA
 AAATGGTTGAGTGTTCGTGGACTTGCTTT

Note: In order to position one gold nanoparticle in the body center of the octahedral frame, ‘ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTT’ sequence was added in the blue marked staple DNA strands.

b. Bridging strands of octahedral DNA origami frame

Oct-bridge-1 TGTAGCCTTTTGTGAGAGGGTAATTGAGAACACCAAATAG

Oct-bridge-2 CTATTATTCTTTTTGTAAAATTCGCATTATAAACGTAACTAG
 Oct-bridge-3 CATTATCATTTCAGGTCTAGAGCAAGCTTCAAGGCG
 Oct-bridge-4 AACATCCATTTTTTCACGTTGAAAATCTTGCGAATGGGATTT
 Oct-bridge-5 AACGCCTTTTGTAAAGATTCAAAAGGCCTGAGTTGACCCT
 Oct-bridge-6 CGGAGAGTTTTTCAGAGCCACCCTCTCAGAACTCGAGAG
 Oct-bridge-7 GCGCATTTTTATTGCGTAGATTTTCAAACAGATTGTTTTG
 Oct-bridge-8 GTCTTTCCTTTTCTCCAGCCAGCTTCCCCTCAGGACGTTGG
 Oct-bridge-9 TTCGCCTTTTAAGGGAACCGAAGTCTGAGCAGACGGTATCAT
 Oct-bridge-10 GCTGACTTTTAAGCCTTAAATCAAGACTTGCGGAGCAAAT
 Oct-bridge-11 ACAGGTAGTTTTGATAAGTCCTGAACAACCTGTTTAAAGAGAA
 Oct-bridge-12 ATTCCAAGTTTTATTTAAGAAGTGGCTTGAATTATCAGTGA
 Oct-bridge-13 GTATAAAGCTTTTGGTAATAGTAAAATGTAAGTTTTACTAT
 Oct-bridge-14 CCCCCTTTTAGGCGTTAATAAGAAGACCGTGTGCGAAG
 Oct-bridge-15 AGTGAATTTTTTTTCAAAGCGAACCAGACCGTTTTATATAGTC
 Oct-bridge-16 CCAACCTATTTTGTAAATCGTCGCTATTGAATAACTCAAGAA
 Oct-bridge-17 CAGCATTTTTTGTCTTGAGGACTAAAGAGCAACGGGGAGTT
 Oct-bridge-18 AGGTCATTTTTTTCAGAACCGCCACCCTCTCAGAGTATTAGC
 Oct-bridge-19 GTAGCGATTTTTAACCTGTTTAGCTATTTTCGCATTCATTC
 Oct-bridge-20 TGTATCCTTTTAGCACCATTACCATTACAGCAAATGACGGA
 Oct-bridge-21 CCGCCTTTTTTAAAGGTGGCAACATAGTAGAAAATAATAA
 Oct-bridge-22 AATAGAAAATTTTTTTCGCTATTGGGCGCCCGGGGTGCGCTC
 Oct-bridge-23 AACAAATGATTTTGTCCACTATTAAGAACCAGTTTTGGTTCC
 Oct-bridge-24 GTGAATTTTTTAAAGTTTTAACGGGGTTCGGAGTGTAGAATGG

c. Staple strands of cubic DNA origami frame

cub-staple-1 GAGTAACAGTGCCATGAAAGTATTAACACGCATAAAGACAGC
 cub-staple-2 TAAATAAGGCGTTAGAAAAAGCCTGTACTACCTACGCGAGAA
 cub-staple-3 ATAGCTGTTTCCATAAAGTGTAAGCTGTTGGGCCAGTCACG
 cub-staple-4 ATTAGACGGGAGGAGAGATAACCCACTTGATGGGGAACAAGA
 cub-staple-5 ACCAGTAGCACCCCGTAATCAGTAGCATTATACATGTTACTT
 cub-staple-6 ACGTAATGCCACCATCTTTGACCCCCAGGAGGAGTCTCTGA
 cub-staple-7 AATCGTCATAAAAGTTCAGAAAACGAATAACGCATAGCGAGA
 cub-staple-8 TACGCAGTATGTAAAGACACCACGGAAGAATTATTTTTCGGG
 cub-staple-9 CGTTTTAGCGAATTGCACCCAGCTACATCCCATGAACAAGCA
 cub-staple-10 ATCGCACTCCAGCCATTCAGGCTGCGGCCATCAGCGGATTGA
 cub-staple-11 GGTTTATCAGCTATGACAACAACCATTCATAGTGGAGTGAGA
 cub-staple-12 TTTGCGGAACAATGGCAATTCATCAATTATCCTATCCCAATC
 cub-staple-13 CTCAGAGCCACCAGCCGCCAGCAGAATCAAATCTTTTCA
 cub-staple-14 AGCTTAGATTAATAAATCATAGGTCTGACAAACAATATATGT
 cub-staple-15 GAACTAACGGAATTCAACTAATGCAGATTGCTGCAGTTGATT

cub-staple-16 ATTTAAATTGTATTTTTTAACCAATAGGGTGCCTCTGCATTAA
cub-staple-17 CTGATTGCCCTTAGCAGGCGAAAATCCCGGAGAATGAACGGT
cub-staple-18 TTGGGGCGCGAGATCATACAGGCAAGTGCTCATACTTTAATC
cub-staple-19 TGATTGCTTTGAGCAAAAGAAGATGATATCATACAACGCCAA
cub-staple-20 AGATTCAAAGGCTAGCTGATAAATTATTGAGTAGCAGATAG
cub-staple-21 AGAGAGTACCTTCGGATGGCTTAGAGGACCATAAGCCCGAAA
cub-staple-22 AGGCGCATAGGCAAATCAACGTAACAGTTTATTGAGGGAAGG
cub-staple-23 GACGACAATAAACTGAACAAGAAAAAATCCTGAAATAAAGAA
cub-staple-24 CCCAATAGGAACGCATTCCACAGACACTGAGACGTGTATCAC
cub-staple-25 GAGAAGGGCCTGTACCATGTACCGTAACCCACCCTCCACCCT
cub-staple-26 GCTATTACTTTTTTCATTTAACAATTTCCAGCTGGCGAAAAA
cub-staple-27 TAATAATTTGCTAATGTCGTCTTTCCAGATGCTTGATACCGA
cub-staple-28 GAAACAAAGCAGCAATTACCATTAGCAAATTTGGGCAATCAT
cub-staple-29 AATCAATATTACCCTGGCTGACCTTCATGAGGACATTAAGG
cub-staple-30 GAAATCGAATATCAAATTAAGTGAACACAGAATAATCCAACG
cub-staple-31 GACCATTCGGTGTGATGTTTTAAATATGGAATCAGTTGAGAT
cub-staple-32 AGGTCAGAAACACTTACGAAGGCACCAAGGAAGTTTACATGG
cub-staple-33 GTAACAAATCGTAACCGTGACCAGACCGGAAAATGTGAGCGA
cub-staple-34 AAGCAAAGACATCTGCCAGTTTGAGGGGCGCTTCTGGTGAG
cub-staple-35 TACAAATTACCTGAATACCAAGTTACAATCGGGAGTTGAGC
cub-staple-36 AATCAGGATTTTTGTAATTGCTCCTTTTGAAGCAATCGAGCT
cub-staple-37 TACGAGCTGCTATTCCTCCCGACTTGCGTTATCCGGAATCAT
cub-staple-38 TAACCCTGTAAAATCTCCAAAAAAAAGGATTTCTTAAACACA
cub-staple-39 TGAATATTATCAAATAATGGAAGGGTTGCGCCTGTTTATCA
cub-staple-40 AAATAAATGATACAAGACTTTTTTCATGACCTAAAACGAAAAA
cub-staple-41 AGGCCGCGGACTAAGGAGTGTACTGGTAAATGCCCCCTGCAC
cub-staple-42 TAAAGTAAGATACACAGTCAGGACGTTGGTAGAAAGATTAC
cub-staple-43 ATCTTCTTGATGCAGGGTTATATACTACTCAGTACCAGGCC
cub-staple-44 AATAATAAACCGTTGTGAGAAAGGCCGGCAATGCACCGAGG
cub-staple-45 CAGTAATGGGCTTAAGTATAAAGCCAACAGGCGAATTATTCA
cub-staple-46 CTTTTGATCCTCATGCCTTGATATTCACTTGAGGCAAAAGAA
cub-staple-47 TCGGTTGGAACCCCTCGGAATACCCAAAAAATACATACATAAA
cub-staple-48 GAATTACATTCTAGAGGATCCCGGGTAATCCGCTCACAATT
cub-staple-49 TGCATGCGATTAAGCTTCGCTATTACGCATTTCCACACAACA
cub-staple-50 GGTAATACGTTTACGTAAGAGCAACACTACGTTAGTAAATCT
cub-staple-51 AAACGCACTTACCGGAAACAATGAAATATACACCATCAATAT
cub-staple-52 ACCCCGGTGAGAGTCTACAAAGGCTATCTCGCAAGCGGTCCA
cub-staple-53 ATAAGGCCCAATAAACTGAAAAGGTGGCAAATAACCTTAAGAA
cub-staple-54 AAAAATGAGTTACAGCGTCTTTCCAGAGAATCATCATATTCC
cub-staple-55 ACCAACGCAGATGAAGAAACCACCAGAATTTAAAATAACGTC

cub-staple-56 TTCGCGTCCATTTCGCCAGCTTTCCGGCAACGACGAGTGTAGA
cub-staple-57 CTCCGGCTAATTACTAAATAAGAATAAAAATGGTTTAATTTTC
cub-staple-58 CCTGATACCGAACTCACCGACTTGAGCCGGCCGGAAACGTCG
cub-staple-59 TAGCTATATAATAACATATATTTTTAAATAGACAGTCAAATAA
cub-staple-60 TAAATGCGAACCGCCACCCTCAGAGCCAACTGAGTTTCGTTG
cub-staple-61 CCGGAACGGTCATAGTAGCGCGTTTTTCACGGCTGTCTTTTCCC
cub-staple-62 ATCGGTGCGGAAGCTGTGTGAAATTGTTCCGAGCTCCAAGCT
cub-staple-63 TTTAGCGCCACCAGACCCTCAGAGCCGCGAGCCGCGCCACCA
cub-staple-64 CTGGCTCAAATTTGGGACGAGAAACACCACAATAGTAGTAGCA
cub-staple-65 AACAAAATTTATCAGACGCTGAGAAGAGCTTAGAAAATCGTC
cub-staple-66 TTGCGTATTTCCAGTAATTGCGTTGCGCAGATTAAATTTTTG
cub-staple-67 TCAAAGGGAGATAGCCCTTATAAATCAACCCAGAGGGTAATT
cub-staple-68 ATAGCCC GCGAAAACAGCCTTTACAGAGCCTGAACAAAGTTA
cub-staple-69 AAGGGAAAATTTGTGCGGAGATTTGTATCAGCACCAATGAAAC
cub-staple-70 TAAGCAAGAAACGCTAGCAAACGTAGAAGAAGTGGGATAAAA
cub-staple-71 TGCTGTATACCACACAACATTATTACAGGGAAGAATTAGTTT
cub-staple-72 ACGATCTGCCGACATGCTTTTCGAGGTGACTCCAAATTGCGAA
cub-staple-73 TCAAAGCGCGGATTCCTGACTATTATAGTTCATCAACATTTA
cub-staple-74 AGAACGGCCAATAGCAAGCCTCCCTCACACTTATCATTCCA
cub-staple-75 TGGGCGCCCCGTCGTCCTGTAGCCAGCTTCCCGGAAACCAGG
cub-staple-76 TGCCCGCTTGGGCGTCAGAAAAGCCCCAGTTAAAATTCGCGT
cub-staple-77 CCAAAGATCACCGTGACCAACTTTGAAACAAGAGTAATCTCG
cub-staple-78 CATTTTCCGCAAATCAGATATAGAAGGCGGAGGTTTTGAAGG
cub-staple-79 AACAGTAAAGAGAACAGTACCTTTTACAAATCGCGCAGAGAA
cub-staple-80 GTCGAGAGCCACCCTCAGACCTAAATTTACGGATAAGTGCC
cub-staple-81 TGAATTACAAAAGGTCATATGGTTTACCATTGACAAGAACCG
cub-staple-82 ATTTTTATACCAAATCAGAGCATAAAGCGCAAGGTGGCAACA
cub-staple-83 GATTATAATAAGTCCAACATGTTTCAGCTAAAAGGTCGTCAGA
cub-staple-84 ATATTCGTCTGAAACCGTATAAACAGTTATAAGTTTACAGAG
cub-staple-85 AATTACGTTTTAACTATTCATTGAATCCAGACTGGCAGAGGG
cub-staple-86 GCAAGGCCTGCAGGTGCGACTAATTTTCTCGGGGGATGTGCT
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cub-staple-88 CATTGCCTTGATAACCAGGGTGGTTTTTTGAGAGAGTTGCAAC
cub-staple-89 TGGGATTTTTTTTACGTTGAAAATGTTTCCGAATTTTCTGTA
cub-staple-90 CCTACCAACAGTAATATAAAGTACCGACAATGCAGAACGCTC
cub-staple-91 GAGCTAAAGCTCATAACGTTAATATTTTAAAACAGAGGCGGT
cub-staple-92 ATTTGCCAAAATAGACCGTCTATCAAATGGAGCGGAATTAGA
cub-staple-93 TATTTTTTTGCCCCACCAGCTGGCCCTTTTTTACATATGT
cub-staple-94 GAGTAGTATTATACTTTTCGCAAATGGTCTCAATTCTACTAAC
cub-staple-95 CAGAACCGGGTTGATTAGCGGGGTTTTGTACACCAGTACAAA

hb-staple-10 CCGTAATCGTAAATAATCAGAAAAGCATGATATGATCTACAA
 hb-staple-11 AGCCGTTAGATTAGCGGGAGGTTTTGAAAAAAGAAATTCTGA
 hb-staple-12 GCCCGTATAAACTATTCTGAAACATGGCCAGAATTGATGATA
 hb-staple-13 AATAATGAATAAGGATAAGGGAACCGAATAATAAGCAGAGAG
 hb-staple-14 AAAAAAAGGCTCGCTTGCTTTCGAGGTTGTCGTTTCAACAGT
 hb-staple-15 TAAGTGCTGTTTCCCTGAGAAGAGTCCTTTTTTCAAAGGCGAT
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 hb-staple-17 TACATAAATCCGCGAAATACGTAATGACCGATTGAAAATACA
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 hb-staple-19 AGCATCGGAACGTAAAGACTTTTTTCATTTCTTACATAACCGA
 hb-staple-20 TAAAGGAGCTCTGTATGGGATTTTGTATAGAAAGTTCACGT
 hb-staple-21 GGCTTGATTAGGAAGGTAGAAAGATTCAACGTTAAGTCAGGA
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 hb-staple-23 TTACCTTGTATCGGGTAAATGAATTTCTATTTTCGGTCTGAAT
 hb-staple-24 TAGCGACTCAGAGCAGAGCCACCACCGGCGGTTTGTAGCCT
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 hb-staple-26 ATGTACCAGCTCAGAACCGCCACCCTCACCCAATAAGTTTTG
 hb-staple-27 TAGATACATTTCAATAACCTGTTTAGTATTAAGTAGCGGGGT
 hb-staple-28 AAGGGTGAGAAAAGTCAAATCACCATCACCTCCACCACCAG
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 hb-staple-30 TTCATCACCAACTGACCAACTTTGAAAGTAGGCTGACCGGAT
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 hb-staple-32 CCAGCTTCGACGACCCGTGCATCTGCCATCGGATATAATGGG
 hb-staple-33 ACGAGCCCCAGGGTGGAGAGGCGGTTTGGTCGTGCTGCCCGC
 hb-staple-34 TTGTATCATCGCCCATGTTACTTAGCGAAGTTTGAAAGAGGC
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 hb-staple-37 GAGCGGAATTATTCATCAATATAATCGTGGACTTATTAATC
 hb-staple-38 CCACCCTGAGAGACGGCCGTAAAAATTTTGAGTAAGGTTGAG
 hb-staple-39 CCACCACAGCCCTCAATTACGAGGCACCAATACTGAGAACCG
 hb-staple-40 CCGAAATGTGCCGGAGAGGGTAGCTATTGCCTGAGAAACAAG
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 hb-staple-42 TTATATAAAAATAGTGAATTTATCAAAACGGCTTACTAGAGG
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 hb-staple-44 TCAACAGAACATGTTTCTGTCCAGACGAAATAAGAAGTAATG
 hb-staple-45 CTTTAAAAAATTGCAGCGGGTCTGAACCATGTAGATAATCG
 hb-staple-46 CTTTTTTCGCGAGTAGCTCAACATGTAAGCCCGAAGCAAAC
 hb-staple-47 GGCGCATCGTAAAGTATCGGCCTCAGTGGCCTTATTCTCCGT
 hb-staple-48 CGAGCTCGAATTAATTGTTATCCGCTGCGATAGTCTGAGAGA
 hb-staple-49 GTCAATCCTTGCCCAAAGCTGCTCATTTCGACAAGAGCTGACC

hb-staple-50 AAAATCTTCGTAGGAATCATTACCGCGCATCCGGTCGCGAGG
 hb-staple-51 CGGGAGAAACAAGTTACAAAATCGCGTTGTTTGTGCACGTAA
 hb-staple-52 AGTTTCAGAGGCTTGATAGTTGCGCCCCGCCTCCCAGAATCA
 hb-staple-53 TTTGCGGTAGCCTTTAGCGTCAGACTGTCCCCTTACCATCTT
 hb-staple-54 TGCTGCAATATATTTAAAGAACGCGAGAACTATATTTGCATG
 hb-staple-55 TCCCAATCCAAATTTTTTGTTTAACGTGACCTAAAATAAGGC
 hb-staple-56 AATCAACGTAAGTACGAGAAACACCACGAGGCATGAACGGT
 hb-staple-57 TATACCACGAAAGGTGGCAACATATAAATTTGTCAAAAATTC
 hb-staple-58 TTCAATTTTCCCTTTGTAAATCGTCGCTTACATAAGAATTAC
 hb-staple-59 CATTATATTGGGCGGGAAGCATAAAGTTGAGGGGATCATCAA
 hb-staple-60 GATAACCCACAAAACAATGAAATAGCAAATGAAGGGAGAATT
 hb-staple-61 CGCAGTCAACCTATAGTTAATGCCCCCTATGGGGTTTAACTG
 hb-staple-62 AAGGTAAAGTAATCAGCTAATGCAGATATAAAGACATGTAAT
 hb-staple-63 GAAACAGATGAGAATCGCCATATTTAACTCGAGCCGAATATA
 hb-staple-64 CGAATAAAAAGTTCCAGTAAGCGTCATACATAAGTTCAGTGCC
 hb-staple-65 TAAAGGCAAGACAATGACAACAACCATCGGCTTGACCCTCA
 hb-staple-66 GCCAACGCGCGGGGTTTTTCTTTTCAATTCCACGAGCTAACT
 hb-staple-67 ATACCCAAAAGATTAAGACTCCTTATATCGCACCGGAAACCA
 hb-staple-68 ATCACCGGAACCCGCCACCCTCAGAAAGCACCGTTTCATCGG
 hb-staple-69 ATAGTAAAATGTGTCATAAATATTCAAGGTTTACACCCTCAT
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 hb-staple-71 GCTGATTGAACAAGTAGGGTTGAGTGTTTCGGCAAATCCTGTT
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 hb-staple-75 TATCATTCCAAGAAACCAAGTACCGCGCCTGTTATAATATCC
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 hb-staple-77 CACCCTCCGGAATCTTAGACTGGATAGCCAAAAGAGGAACCC
 hb-staple-78 AAActACAACGCGCGTAACGATCTAACAGATACTCGTTTACC
 hb-staple-79 CTTCTGAATACCAATAACGGATTGCGCTATATACAGATTTTC
 hb-staple-80 GCCTGTTTAGTATATACAAATCTTACTACAATTCTTTCCAG
 hb-staple-81 CAAAAGGATAGTTACTGTAGCATTCCACGTAACACGCGAGAG
 hb-staple-82 TTGAGTACATATCATACAAACAATTGATTAATTTACAAAGA
 hb-staple-83 CCCCCAGCACCCTACGAAGGCACCAACACACTCAAACAAAG
 hb-staple-84 TGATACCTGAGGACAGGGTAGCAACGGCGATCGTCAGGGAGT
 hb-staple-85 CGTTGGGGCTCAACTTTAATCATTGTGATATACCATAAAACG
 hb-staple-86 TTAATTTGCTTTGAATAATGGAAGGGTGCTGAATATCGCGTT
 hb-staple-87 CAAAGGGTGGCAATCATCATATTCCTGATTGCGGATAAAAAGT
 hb-staple-88 CCTCCCGACTTGTTGCTATTTTGCACATCGAGACAAGCAAAT
 hb-staple-89 AGAGCATAAAGCGTAATACTTTTTCGATTTTCATAACATCCA

hb-bridge-9 AAGATTCAATTTTCATCCTAATTTACGAGAAGAAAATATCAAC
hb-bridge-10 TCTTTCCTTTTTAGCCGCCGCCAGCATTCCAGAACAGAGCCA
hb-bridge-11 TCGATGAATTTTTTAGGCAGAGGCATTTAACGCCACCAACGC
hb-bridge-12 CCGACAATTTTAGGCTATCAGGTCATTTTTGAGATCAACCG
hb-bridge-13 CATTACCATTTTTAGCCTAATTTGCCAGTACGAGCGTTTATCC
hb-bridge-14 CTAGAAAAATTTTTTTGTTAAAATTCGCATTGTAAGAAGATT
hb-bridge-15 CATAATCAAATTTTCAGATATAGAAGGCTTCCAATAGACAAGCA
hb-bridge-16 TAGCGAATTTTCATTTTCGGTCATAGCAGCGCGTTAATCAG
hb-bridge-17 TTGACCATTTTTGTTAAATAAGAATAAAGTGTGATAATTTAA
hb-bridge-18 CATATTATTTATTTTTTTGCTCAGTACCAGGTTAGGATAGGCTGA
hb-bridge-19 AAGCCTCTTTTTAAAACGACGGCCAGTCCCAGTCGGGGATG
hb-bridge-20 TGCAAATCTTTTATAAATCATAACAGGCAGTAGCATTTTGGGG
hb-bridge-21 TCAGACGTTTTTAAAGTTACCAGAAGGAAAGCAGATACCGAA
hb-bridge-22 GTTGGGATTTTTTTTTAAATGCAATGCCTTAGAACTATTTCA
hb-bridge-23 GTAACAGTTTTTAACTGAACACCCTGAAATTAGACAATAGCA
hb-bridge-24 TATCAGAGATTTTCAGGAGTGTACTGGTAATGGCTTTGGAAAG
hb-bridge-25 CTTTTACATTTTTAGACGACGATAAAAAACATAACCCATAACGC
hb-bridge-26 ACCAGTACTTTTAAACAGAAATAAAGAAAAATTATTGATTATA
hb-bridge-27 ACCAGAAGTTTTTTCAGCGGAGTGAGAAAACAACCTCTTTCCA
hb-bridge-28 AATCTCCATTTTCTTTGCCCGAACGTTACAACCTCGCCAACGT
hb-bridge-29 ATGAATCGTTTTTAAAAGAATACACTAACTAAAACCCATTAA
hb-bridge-30 CGGAGATTTTTACATTAATTGCGTTGAATGAGTACAACAT
hb-bridge-31 CGGGTACTTTTGTACAGACCAGGCGCAAGGACAGGCAGACG
hb-bridge-32 ATTACCCATTTTCTACCTTTTTAACCTCTCATAGGCTTAGAT

g. Staple strands of elongated octahedral DNA origami frame

loct-staple-1 AAAGTACAACGGGTTACTTAGCCGGACTCAGCAATACGTAAT
loct-staple-2 TACTGCGGAATCTCAGGTCTTACCCTATTCTGGGGTTGATA
loct-staple-3 ATAGTTGCGCCGTTTTGCGGGATCGTGTTAGCGAGGAATTGC
loct-staple-4 ATTAAGCAATAAAATACTTTTGCGGGAGTTTCATATTTTCAT
loct-staple-5 TTTAATGCGCGAAAGATAAAACAGAGCCAGCCAACAGTAAT
loct-staple-6 TGGGATAGGTCAAGATCGCACTCCAGCGGTTGAAATAGGAAC
loct-staple-7 GTGAATTACCTTAACGGAACAACATTGGCGCAGGATATTCAT
loct-staple-8 TAATAAGTTTTAGCCTATTTCCGGAACCTTGATGGGGAACAAGA
loct-staple-9 TAAGAATAAACAAATCTTACCAGTACCTTATTGGAATAAGT
loct-staple-10 AACCAATCAATAGTTTTTATTTTCATGCCAACGTAATTCTGT
loct-staple-11 CGTGGCGAGAAAGTCACGCTGCGCGTCCACCACTCCTCATT
loct-staple-12 CGGATGGCTTAGTAAAGTACGGTGTCTTTCCGTCGGTGC GG
loct-staple-13 GAAATTGCGTAGGGAGAAACAATAACGTTATTAGCAATTCAT
loct-staple-14 ACCACCGGAACCTCAGAGCCGCCACCAAAATCACTTAGCGT

loct-staple-15 CGACGTTGTAAACGGGTACCGAGCTCTATTATAGAGCTTCAA
loct-staple-16 AGGTCATTGCCTTGTCAATCATATGTGCCTTTAGCCGGAGAC
loct-staple-17 TAACCGTTGTAGTCCAGAACAATATTTGCGCTGAACAAAATT
loct-staple-18 CAATAGATAATATAAAATCCTTTGCCCGGCGGTCTCAATCAAT
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loct-staple-20 AAGGTAAATATTTTGGGAATTAGAGCTTTTTAAGAAAACTTT
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loct-staple-25 ACAGCATGCTCCATAGATTTGTATCATCCCCAGCGAAACGAA
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loct-staple-30 TACATTTAATAGTACATCCAATAAATCAAAGCTAACCAAAAA
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loct-staple-33 GTTGGGATGAAAGAGGACAGATGAACGGAGTAGATCATTAGA
loct-staple-34 CTTTTTCAAAGAATACTCATCTTTGACCGCCTGATGAAATCC
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loct-staple-93 CCAGTCAGAGTAGTAAATTGGGCTTGAGACTGGCTCATTATA
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loct-staple-95 AACACTGCAGAACCTTGCAAAAGAAGTTTAGATACATGCAAA
loct-staple-96 CCACCCTAGGATTAGCGGGGTTTTGCTCGAGGTTTAGGGGT
loct-staple-97 GCTGAGATATGGTTGCTTTAGTAGAAGAGGCGAATAATTACC
loct-staple-98 AGTTTGAAAGCAAATATTTAAATTGTAAAGCCAGCAAATCTA
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loct-staple-117 TTATCCGGTATGCCGGAGAGGGTAGCTAAACAAGAGAATCGC
loct-staple-118 CTGACCTATAAGGCTTGCCCTGACGAGAGGGCGCATAGGCTGG
loct-staple-119 AACCAAGTAACAACGCCAACATGTAATTAACAAAGAAGGAGC
loct-staple-120 CATCAGTTAGCATTGCAAGCCCAATAGGCGCCACCAACCAAA
loct-staple-121 AGTTAATGCAAAATGGTTGAGTGTGTTTCGTGGACTGATACA
loct-staple-122 GAATACCATAAGAAATTAGACGGGAGAAAATTGAGATAGCTA
loct-staple-123 GCGACCTCGGAACGAGTTTCCATTAAACCAACCTAATTATAC
loct-staple-124 TATCGGCCCAAAAAAATCAGCTCATTTTCGCGTCTAACGGCG
loct-staple-125 TCGTAAATAAAAAATAGATTCAAAGGGTATATGATGAGATCT
loct-staple-126 CATTATGTGATTCCGGTCAATAACCTGTAAAGGTGAAGGCAA
loct-staple-127 AGAACAATTAATTGAAGTACCGACAAAAACAACATAATTTA
loct-staple-128 ATATGCGCAAACGTAAAGAAACGCAAAGAATAGAATGATAAA
loct-staple-129 CCGACTTTGGGTTAATCGCAAGACAAAGTTAATTTCAACCG
loct-staple-130 ACGCCTGTGAGATTAGGCATAGTAAGAGCGATAAACTCAGAG
loct-staple-131 CGCTGGTTTTTCTGTAATGAGTGAGCTACTTTCCAGCCAGGG
loct-staple-132 CGAACCAACGCTCAGGCAGATTCACCAGCCAACAGTTTTGAA
loct-staple-133 TAATAAAGGGAACCGAGTAATCTTGACAACAAAGCAATTTCA
loct-staple-134 CCTTGCTCAAGTTATGATGAAACAAACATTCATTTGTCTGTC

loct-staple-135	GAGTTAATTTGTCGAGAATAGAAAGGAAACGTTGACTTAAAC
loct-staple-136	TGGCAAGGCATTGATGATATTCACAAACCGCAGTCGACGGGG
loct-staple-137	TCGACAACCTGCAACTGAACCTCAAATATAATCAACACTAATA
loct-staple-138	AGCCTAAAGCAAGCAAGAACGCGAGGCGTGAAGCCGCTACAA
loct-staple-139	CAGAGCCACCATTATAGCGACAGAATCATTTCATCGAATCACC
loct-staple-140	TACCTTTAGTAACAATTCTGATTATCAGTTTGGACACGTAA
loct-staple-141	CTCTAGAGGATTGCTCAAATATCGCGTTAGCAAACGCCAGGG
loct-staple-142	TTTAAATGCCGGAACGCAACTGTTGGGAGCCAGCTTGATAAG
loct-staple-143	ATTTATCGCGCCGCGTTAGAATCAGAGTTTACTGTAAAT
loct-staple-144	CCATAAATAAGAGGGGCGGATAAGTGCCTAGGTGTTAGACTG
loct-capture-1	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTAGCCTAAAGCAA GCAAGAACGCGAGGCGTGAAGCCGCTACAA
loct-capture-2	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTTCGTAAATAAAA ATAGATTCAAAAGGGTATATGATGAGATCT
loct-capture-3	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTATATGCGCAAAC GTAAAGAAACGCAAAGAATAGAATGATAAA
loct-capture-4	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTCCGACTTTGGGTT AATCGCAAGACAAAGTTAATTTTCAACCG
loct-capture-5	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTAGTTAATGCAAA ATGGTTGAGTGTTCGTGGACTGATACA
loct-capture-6	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTTGGCAAGGCATT GATGATATTCACAAACCGCAGTCGACGGGG
loct-capture-7	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTCTCTAGAGGATT GCTCAAATATCGCGTTAGCAAACGCCAGGG
loct-capture-8	ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTTTTAAATGCCGG AACGCAACTGTTGGGAGCCAGCTTGATAAG

Note: In order to position one gold nanoparticle in the body center of the elongated octahedral frame, ‘ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTT’ sequence was added in the blue marked staple DNA strands.

f. Bridging strands of the binary system

bs-bridge-1	CGTCCAATTTTTTTCAGAACCGCCACCCTCTCAGAGTATTAGC
bs-bridge-2	CCAGTCATTTTAGCACCATTACCATTACAGCAAATGACGGA
bs-bridge-3	GTTTTTCTTTTTTTTTAAAGGTGGCAACATAGTAGAAAATAATAA
bs-bridge-4	TGTAAGGTTTTTAAGTTTTAACGGGGTCGGAGTGTAGAATGG
bs-bridge-5	CCGAAGCTTTTGCTTTGAGGACTAAAGAGCAACGGGGAGTT
bs-bridge-6	TATCCTGAATTTTTAACCTGTTTAGCTATTTTCGCATTCATTC
bs-bridge-7	GCATGTAGATTTTTTGCCTATTGGGCGCCCGCGGGGTGCGCTC
bs-bridge-8	GCGTTAAATTTTGTCCACTATTAAAGAACCAGTTTTGGTTCC
bs-bridge-9	GGGAGGGTTTTGTTAAATTCGCATTATAAACGTAAACTAG
bs-bridge-10	CGCTATTAATTTTCTCCAGCCAGCTTTCCCTCAGGACGTTGG
bs-bridge-11	GGCGAATTTTCAGGTCGACTCTAGAGCAAGCTTCAAGGCG
bs-bridge-12	CAGAGCCTTTTTGTAAAGATTCAAAGGCCTGAGTTGACCCT

bs-bridge-13 AGGCTATCTTTTGTCTAGAGGGTAATTGAGAACACCAAATAG
 bs-bridge-14 GACCGTAATTTTATTGCGTAGATTTTCAAACAGATTGTTTG
 bs-bridge-15 TCATTTTGTTTTTTCACGTTGAAAATCTTGCGAATGGGATT
 bs-bridge-16 ATTAGCAAATTTTCAGAGCCACCACCCTCTCAGAACTCGAGAG
 bs-bridge-17 CAGAAATAAATTTTATTTAAGAAGTGGCTTGAATTATCAGTGA
 bs-bridge-18 AGCCGTTTTTGGTAATAGTAAAATGTAAGTTTTACTAT
 bs-bridge-19 GCTATTAGTCTTTTTCAAAGCGAACCAGACCGTTTTATATAGTC
 bs-bridge-20 CGCAAATTTTTAAGGGAACCGAAGTGAAGCAGACGGTATCAT
 bs-bridge-21 GCGAAACTTTTGATAAGTCCTGAACAAGTGTAAAGAGAA
 bs-bridge-22 CTTTAATCATTTTTTAGGCGTTAAATAAGAAGACCGTGTGCAAG
 bs-bridge-23 ACCCTCATTTTGTAAATCGTCGCTATTGAATAACTCAAGAA
 bs-bridge-24 TGATACCGTTTTAAGCCTTAAATCAAGACTTGCGGAGCAAAT
 bs-bridge-25 AGGTCATTTTTTAACAAAGTCAGAGGGTTAACTGTTATCCC
 bs-bridge-26 TTGTTATCCTTTTGACTTGCGGGAGGTTTTTTAGCTTACCGC
 bs-bridge-27 CCGCCTTTTTCCAGACGACGACAATAGGTAAAGCTCAACA
 bs-bridge-28 GGTGAATTTTTTTTTATTTTGTCAATCACACCACACGCAGT
 bs-bridge-29 CCAGCATTTTTTAAGTATAGCCCGAAGTCGAGAAAACATG
 bs-bridge-30 GTAGCGATTTTAGCGAACCAGACCGGATTAATTCGTCAGAA
 bs-bridge-31 AATAGAAAATTTTTTGTGCTCACTGCCCAGTCAACATGGTC
 bs-bridge-32 AAACAATGATTTTGTCCACTATTAAGAACCAGTTTTGGTTCC
 bs-bridge-33 CTATTATTCTTTTTAGTCAAATCACCATCAGAGAAAGTTTCAAC
 bs-bridge-34 GTCTTTCCTTTTGCCATCAAAAATAATTTTAACTAATCAG
 bs-bridge-35 ACATTATCATTTTTGCCTCTTCGCTATTACAGGGCGAGCACCGC
 bs-bridge-36 CAACGCCTTTTTTGGGGCGGAGCTGATTAGCTATTCCATA
 bs-bridge-37 CTGTAGCCTTTTTCAAATATATTTTAGAACGCGACCTCCGG
 bs-bridge-38 TGCGCATTTTGGCCGATTAAAGGGATCGGGAGCCCCGCCG
 bs-bridge-39 AACATCCATTTAAGCCAGAATGGAAAGAAATAAACAGAGCC
 bs-bridge-40 CGGAGAGTTTTCAGACTGTAGCGGTTAGTTTGGCCAGTAG
 bs-bridge-41 CATTCCAAGTTTTGCCACTACGAAGGCACGGGTAAAGCGAAAG
 bs-bridge-42 GTATAAAGCTTTTTACCCAAATCAACGTAAGAACCAGCGTCA
 bs-bridge-43 AGTGAATTTTTTCTCGTTTACCAGACGACAACACTAAAGATT
 bs-bridge-44 ATTCGCCTTTTGAATAATAATTTTTTCCAATAATAACGAT
 bs-bridge-45 ACAGGTAGTTTCAATATAATCCTGATTGATGATGATTTTAA
 bs-bridge-46 TCCCCTTTTATCTGGTCAGTTGGCACAAACCCAGTATTA
 bs-bridge-47 CCAACCTATTTTAAAAGGGACATTCTGGTCACACGTTGCAAC
 bs-bridge-48 GGCTGACTTTTAATTACATTTAACAATTCAAGAAATTGCTT

g. Staple strands of anisotropy control

vex-capture-1 ATCCATCACTTCATACTATGACCACTCGTTTGGCTTTTGCAAAGTTAGACT
 ATATTCA

vex-capture-2	ATCCATCACTTCATACTCGATTATAAGCGGAGACTTCAAATATCGCGGAAG CCTACGAA
vex-capture-3	ATCCATCACTTCATACTACATAACTTGCCCTAACTTTAATCATTGCATTATA ACAACAT
vex-capture-4	ATCCATCACTTCATACTGATATTCTAAATTGAGCCGGAACGAGGCCCAACT TGGCGCAT
vex-capture-5	ATCCATCACTTCATACCATAACCTAAATCAACAGTTCAGAAAACGTCATAA GGATAGC
vex-capture-6	ATCCATCACTTCATACAGAAGCAACCAAGCCAAAAGAATACACTAATGCC AAAACCTCC
vex-capture-7	ATCCATCACTTCATACATAAGGCGCCAAAAGTTGAGATTTAGGATAACGGA CCAGTCA
vex-capture-8	ATCCATCACTTCATACCGCCTGAATTACCCTAATCTTGACAAGACAGACCA TGAAAGA
fc-staple-1	CGCCTGAATTACCCTAATCTT
fc-staple-2	CAGCCTTGGTTTTGTATTAAG
fc-staple-3	GTTTGCCTATTCACAGGCAGG
fc-staple-4	AGCCGAAAGTCTCTCTTTTGA
fc-staple-5	ACATAACTTGCCCTAACTTTA
fc-staple-6	TGTCGTCATAAGTACAGAACC
fc-capture-1	ATCCATCACTTCATACTTCAGACGCCACCACACCACC
fc-capture-2	ATCCATCACTTCATACTATCATTGCATTATAACAACAT
fc-capture-3	ATCCATCACTTCATACTGACAAGACAGACCATGAAAGA
fc-capture-4	ATCCATCACTTCATACTGCCACCCATTTTCACAGTACA
fc-capture-5	ATCCATCACTTCATACTTGATACAAGTGCCTTAAGAGC
fc-capture-6	ATCCATCACTTCATACTAGGCTGACTGCCTATATCAGA

Note: In order to position one gold nanoparticle near a vertex of the octahedral frame, ‘ATCCATCACTTCATACT’ sequence or ‘ATCCATCACTTCATAC’ sequence was added in the red marked staple DNA strands. In order to position one gold nanoparticle in the center of a triangular face of the octahedral frame, ‘ATCCATCACTTCATACT’ sequence was added in the green marked staple DNA strands.

h. Modified DNA sequence attached on gold nanoparticles

ThioM-1	GAAGTGATGGATGAT-SH
ThioM-2	GTAGAGTATGAAGTGATGGATGATGATGATGAT-SH

Figures S1-S35

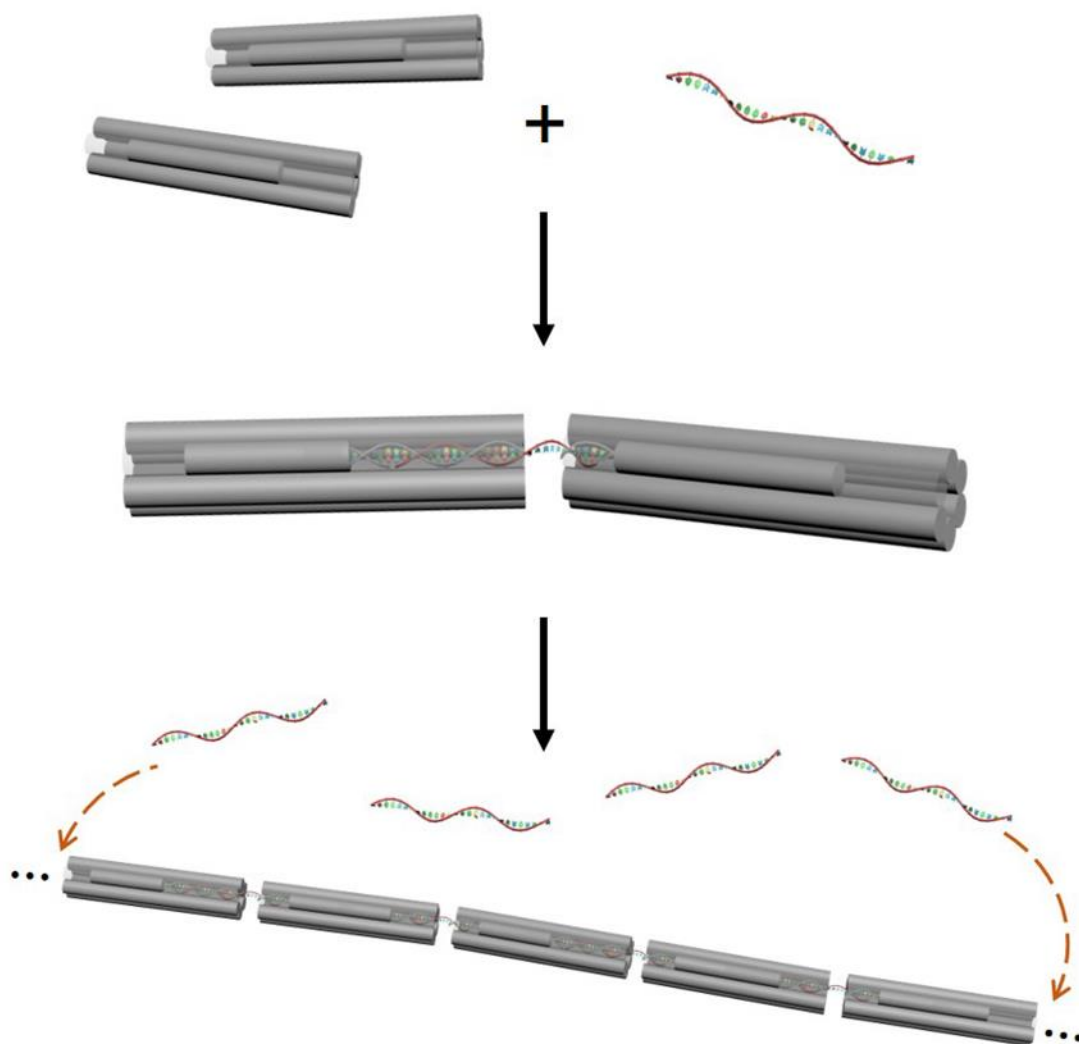


Figure S1. Schematics of the bridge pattern for a six-helix rod-shape model. Hybridization regions are designed inside the two ends of rods. Bridging strands hybridize with two adjacent rods to form a DNA bridge and further expand into a one-dimensional linear structure.

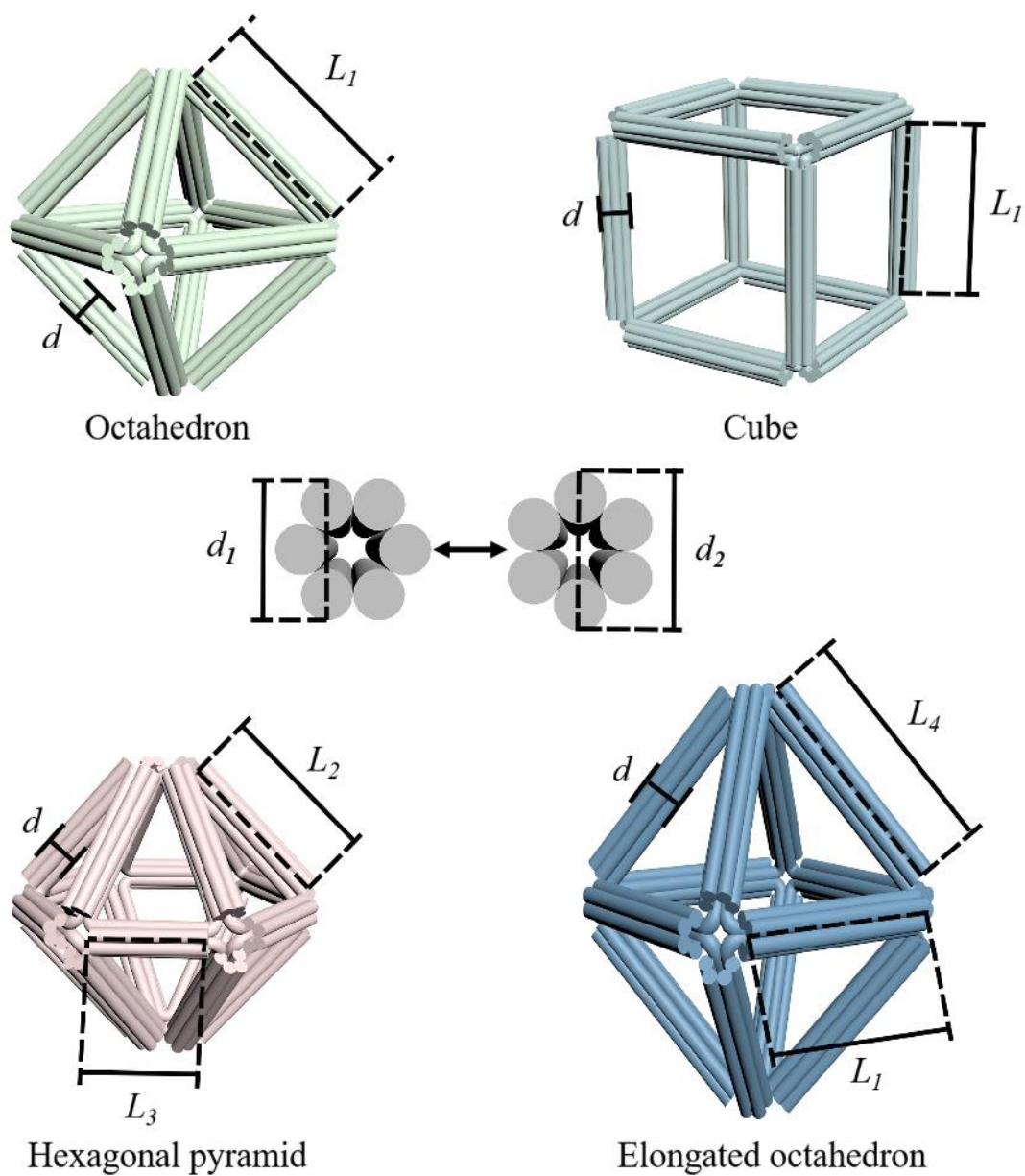


Figure S2. Geometric models for the four polyhedral DNA origami frames are octahedron, cube, hexagonal bipyramid, and elongated octahedron. d_1 , d_2 , L_1 , L_2 , L_3 and L_4 represent the width and length of the edges and their values are as follows: $d_1 = 5.5$ nm; $d_2 = 6.0$ nm; $L_1 = 29.9$ nm; $L_2 = 22.8$ nm; $L_3 = 15.7$ nm; $L_4 = 37.1$ nm.

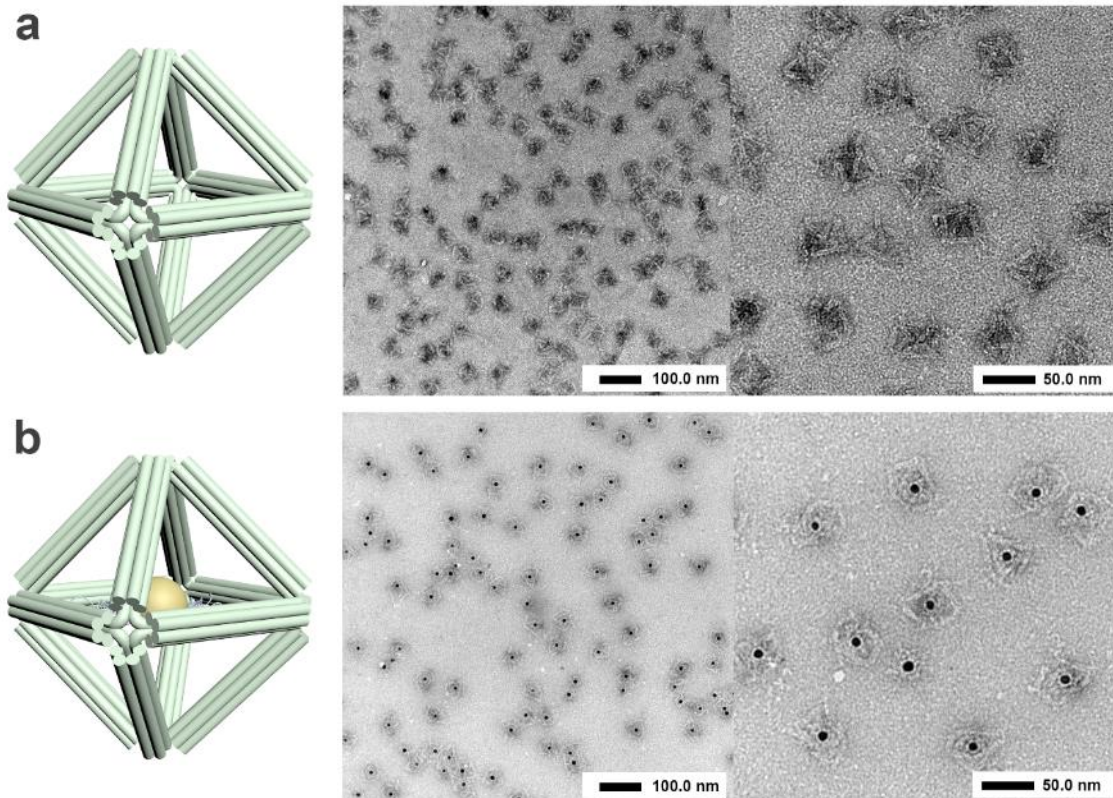


Figure S3. a) Representative negative-stained TEM images of octahedral DNA origami frames.
b) Representative negative-stained TEM images of AuNP-octahedron monomers.
Corresponding schematics are shown on the left.

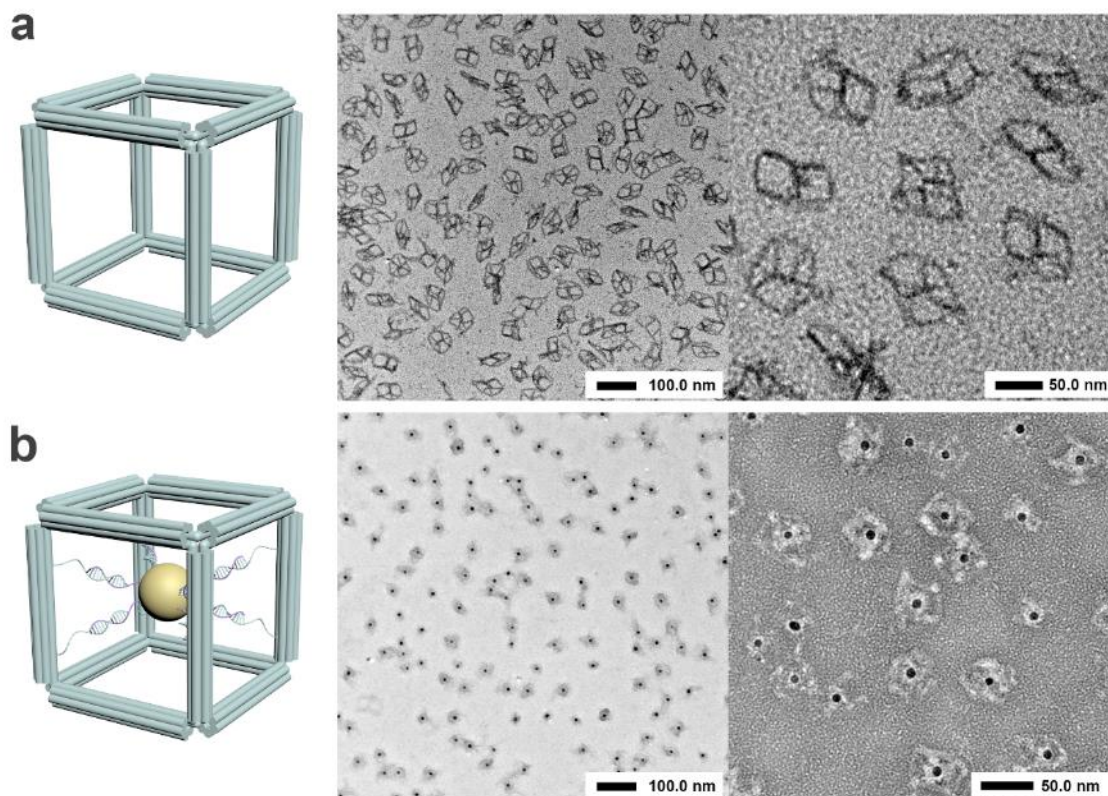


Figure S4. a) Representative negative-stained TEM images of cubic DNA origami frames. b) Representative negative-stained TEM images of AuNP-cube monomers. Corresponding schematics are shown on the left.

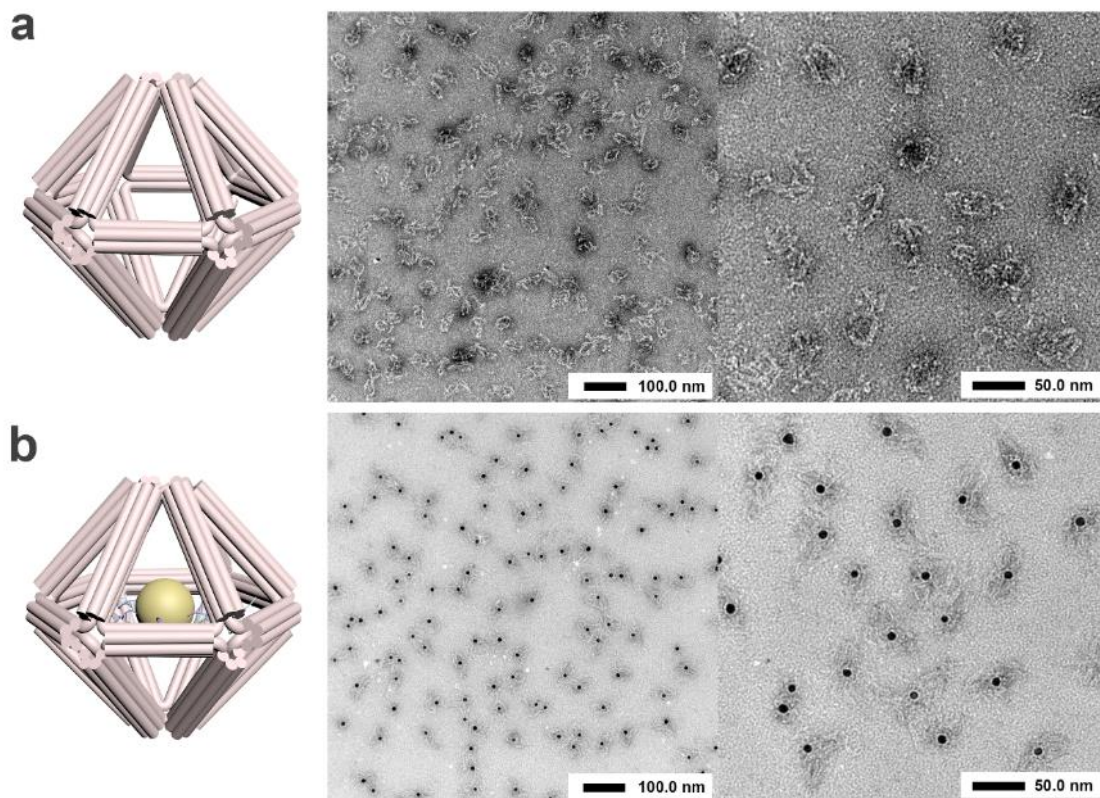


Figure S5. a) Representative negative-stained TEM images of hexagonal bipyramid (HB) DNA origami frames. b) Representative negative-stained TEM images of AuNP-HB monomers. Corresponding schematics are shown on the left.

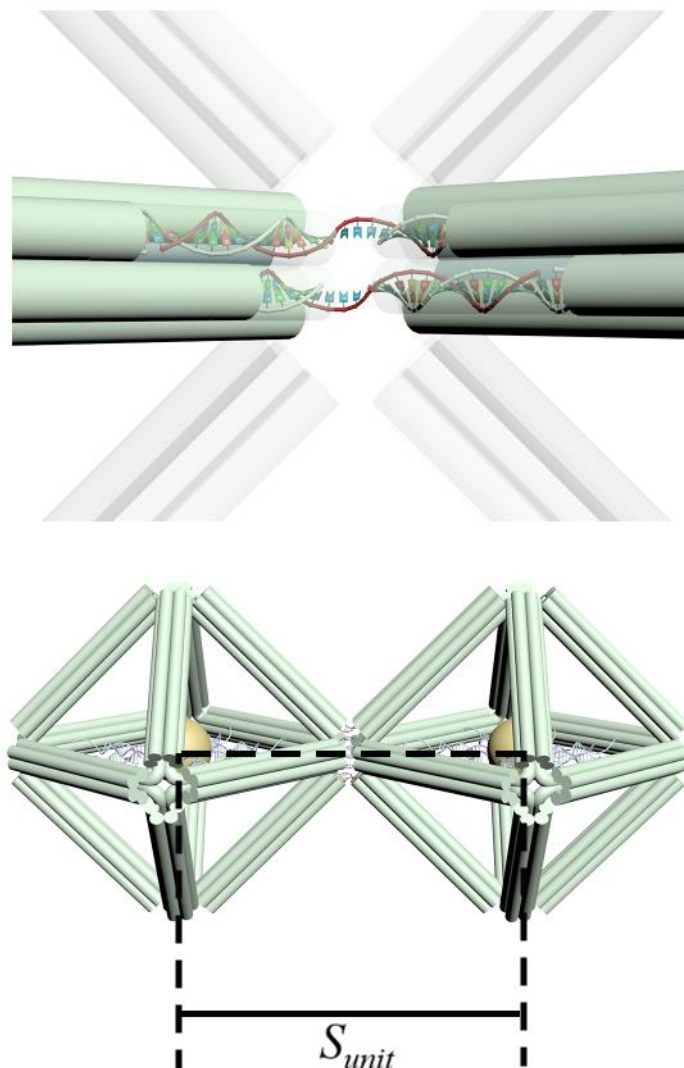


Figure S6. The magnified schematics of the bridge pattern for octahedral frames at vertex junctions. S_{unit} represents the distance between the body centers of adjacent frames in equivalent crystallographic axis directions.

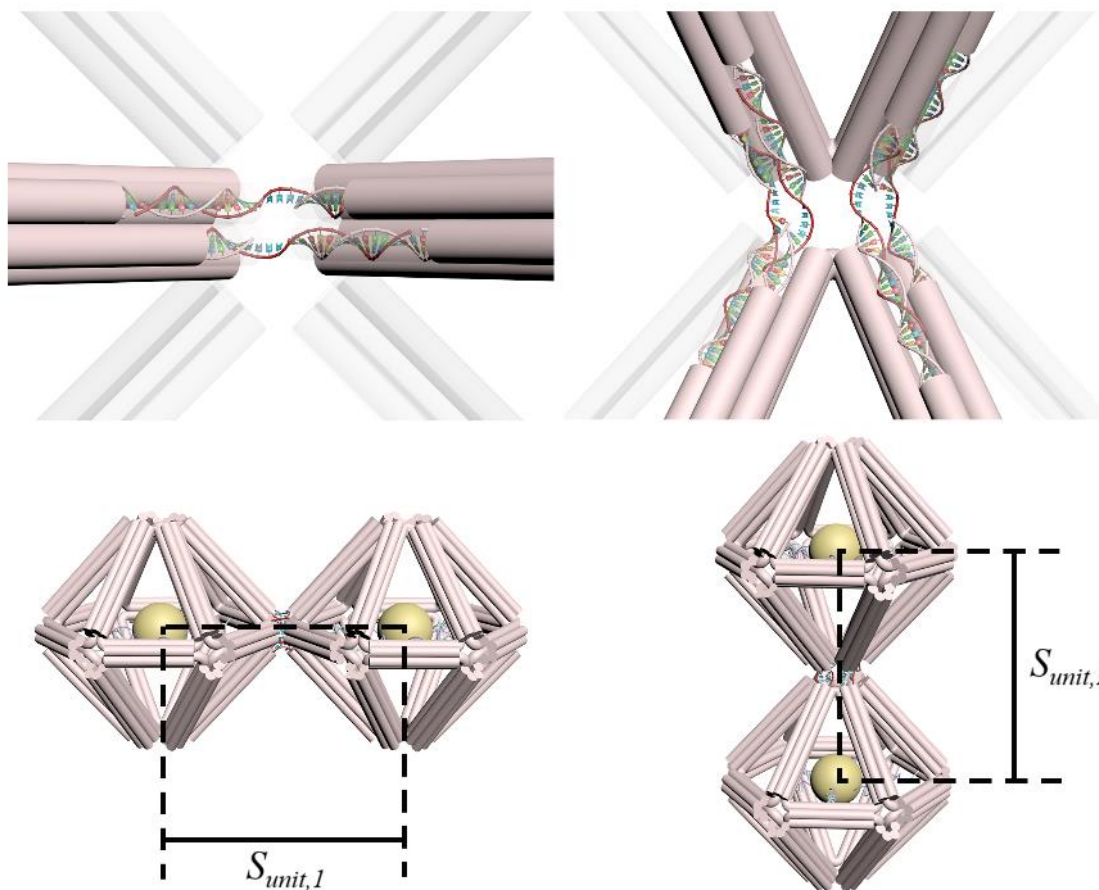


Figure S7. The magnified schematics of the bridge pattern for hexagonal bipyramid frames at vertex junctions. $S_{unit,1}$ and $S_{unit,2}$ respectively represents the distance between the body centers of adjacent frames in non-identical crystallographic axis directions.

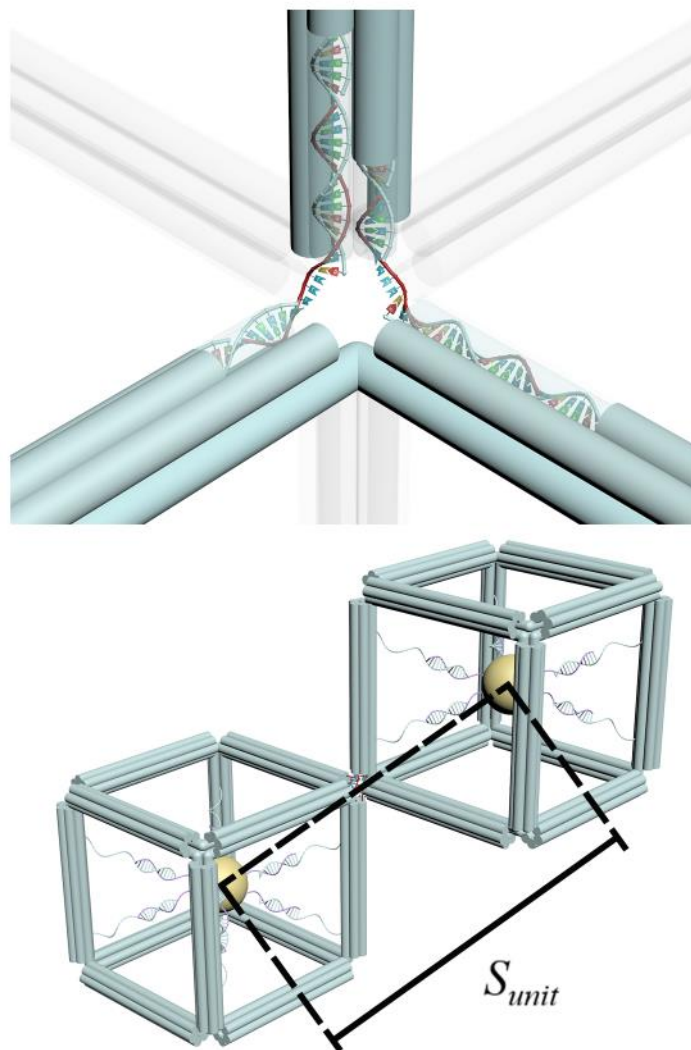


Figure S8. The magnified schematics of the bridge pattern for cubic frames at vertex junctions. S_{unit} represents the distance between the body centers of adjacent frames in equivalent crystallographic axis directions.

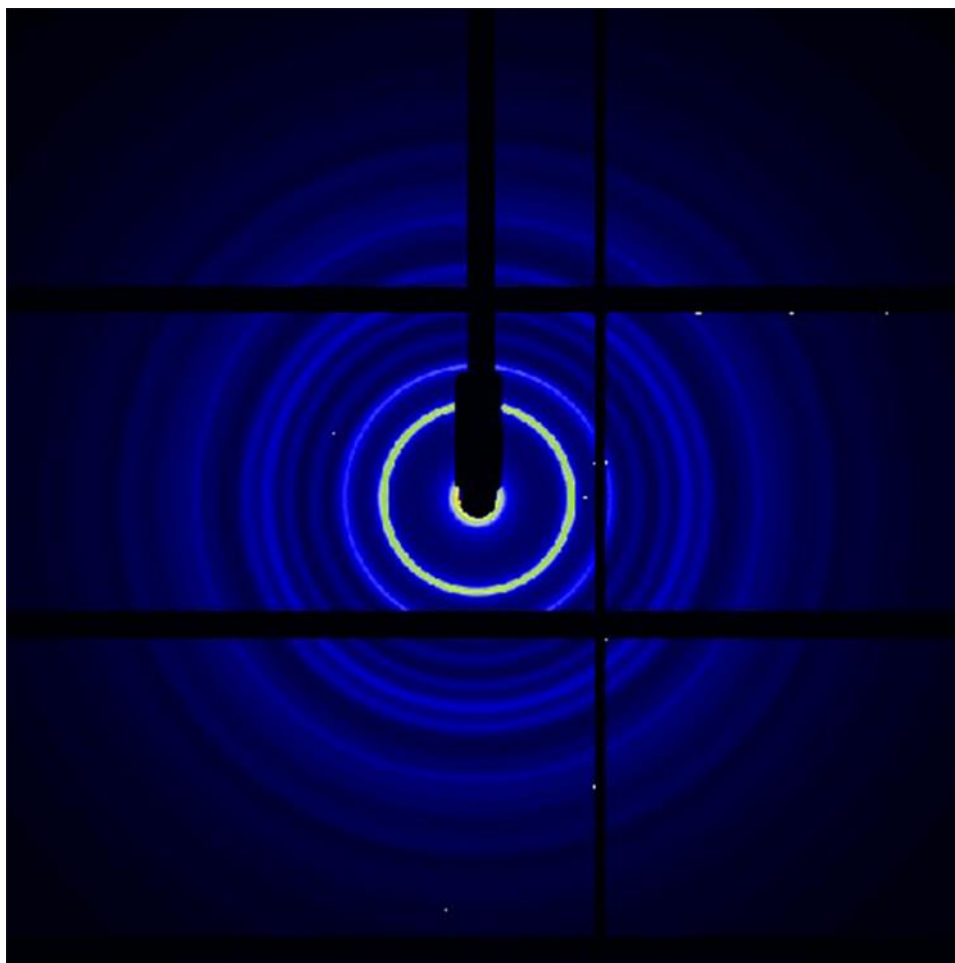


Figure S9. 2D pattern of the AuNP lattice formed by octahedral frames measured by SAXS.

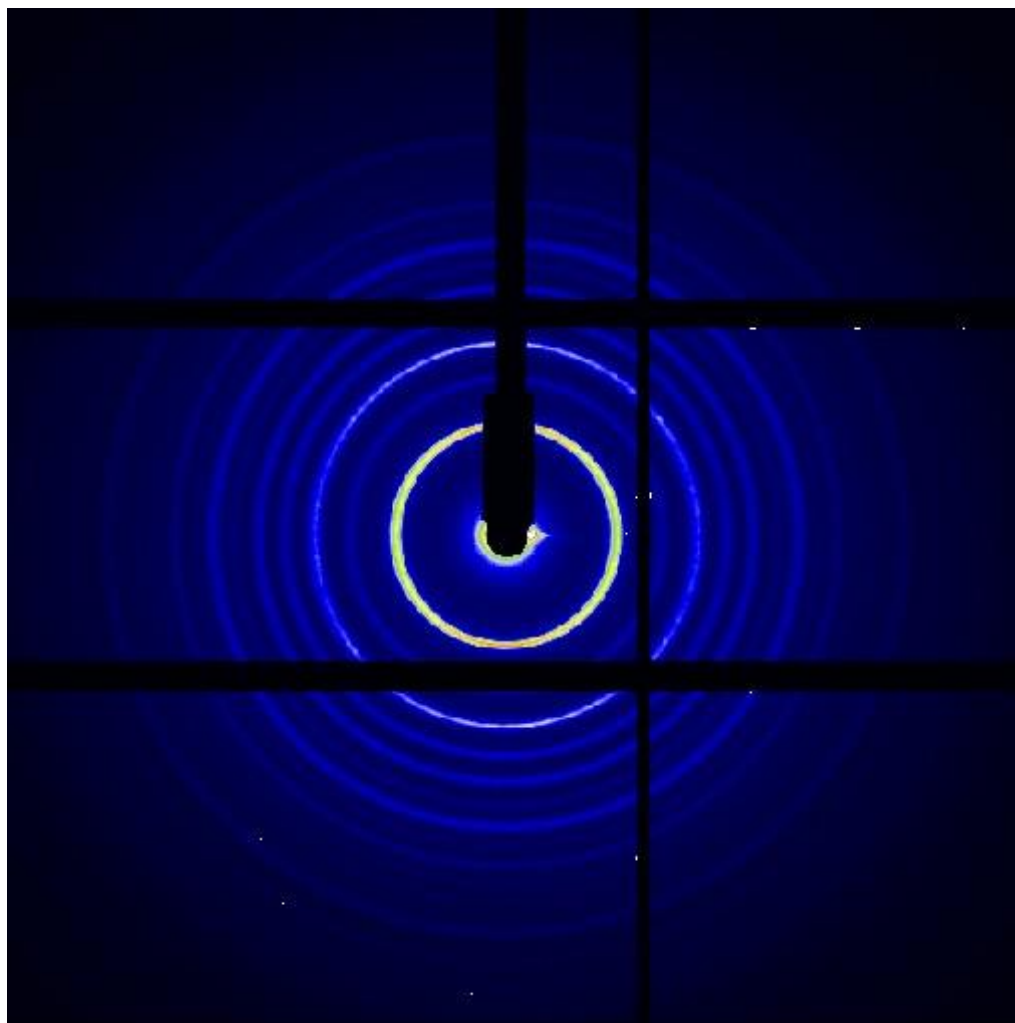


Figure S10. 2D pattern of the AuNP lattice formed by cubic frames measured by SAXS.

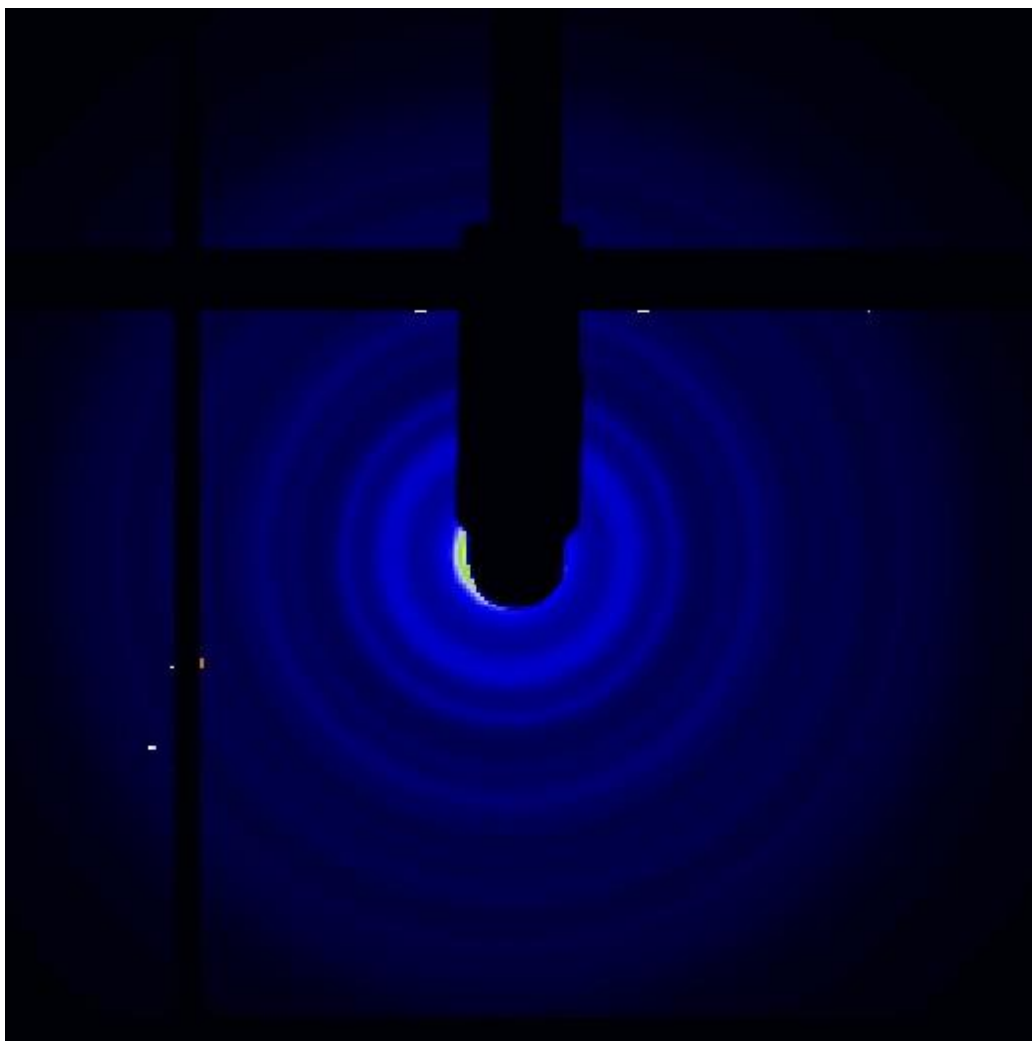


Figure S11. 2D pattern of the AuNP lattice formed by hexagonal bipyramid frames measured by SAXS.

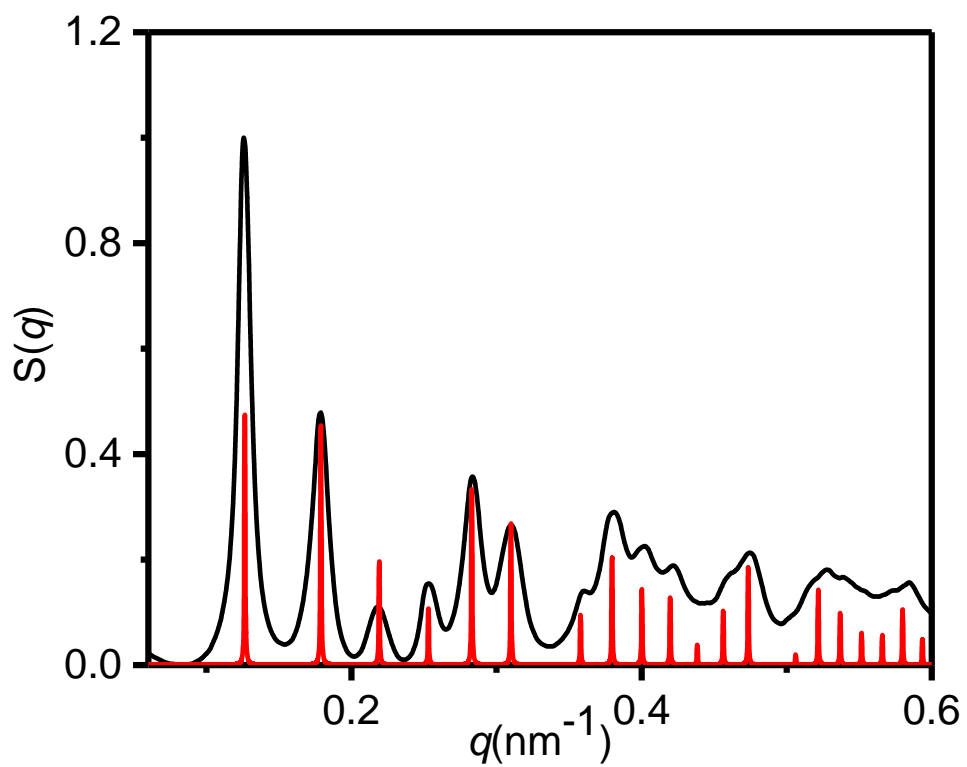


Figure S12. Enlargement of structure factor result for the simple cubic lattice shown in Figure 2d: black curve for the experimental data and red curve for the model.

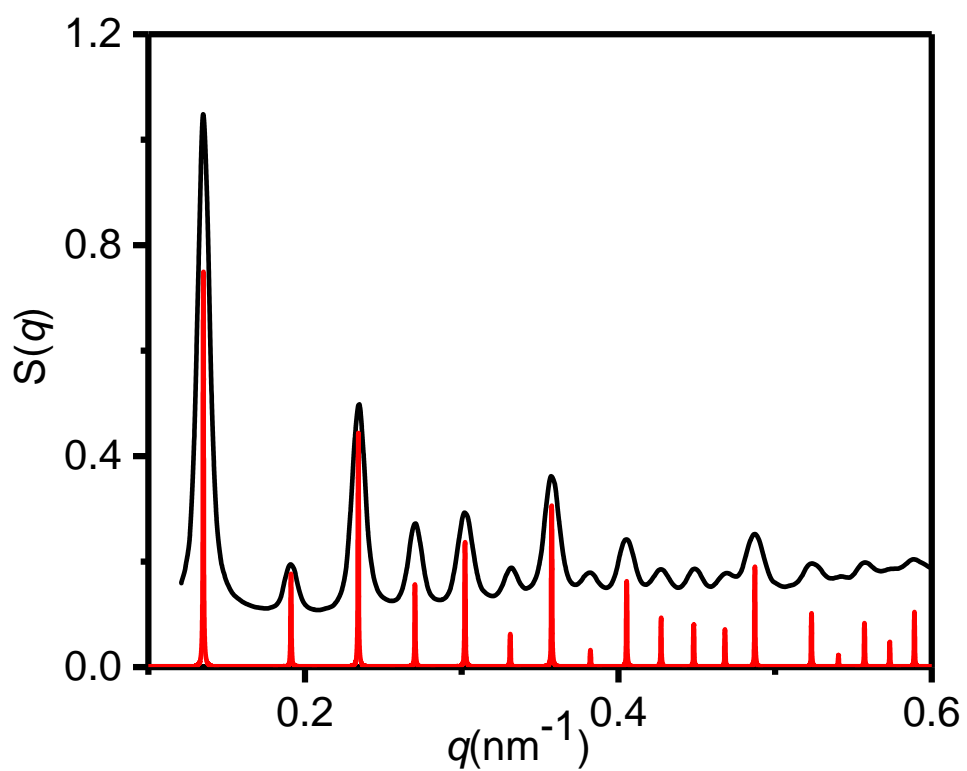


Figure S13. Enlargement of structure factor result for the body-centered cubic lattice shown in Figure 2e: black curve for the experimental data and red curve for the model.

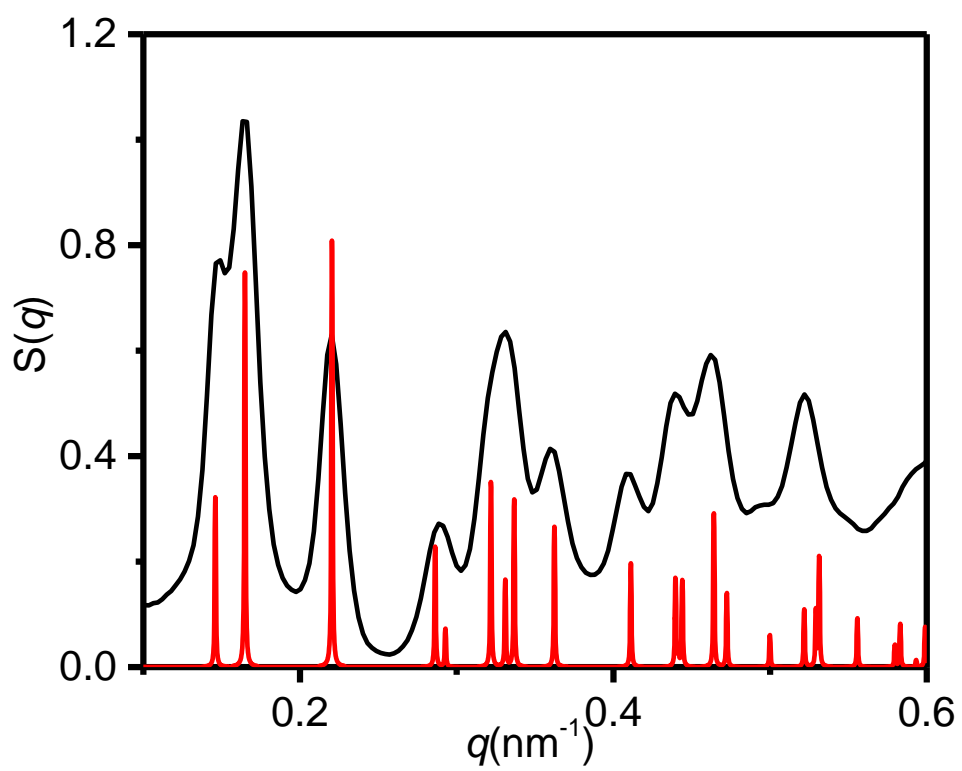


Figure S14. Enlargement of structure factor result for the simple hexagonal lattice shown in Figure 2f: black curve for the experimental data and red curve for the model.

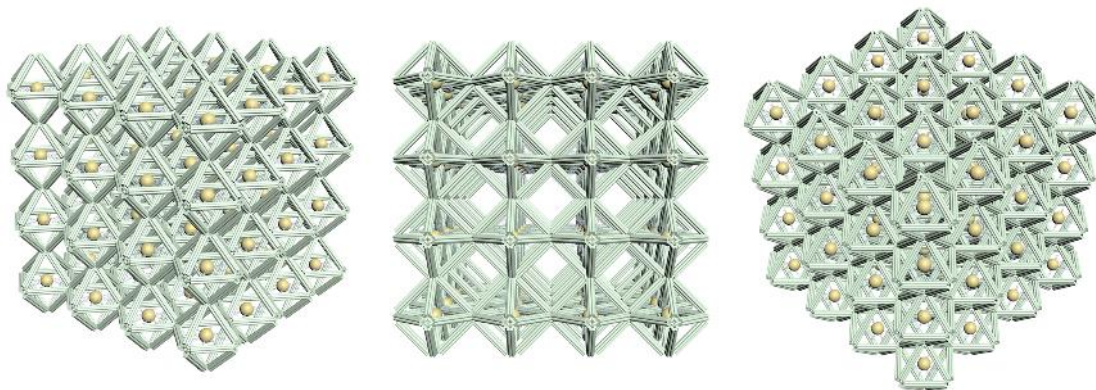


Figure S15. The close-packed models of simple cubic lattice formed by octahedral frames in different angles.

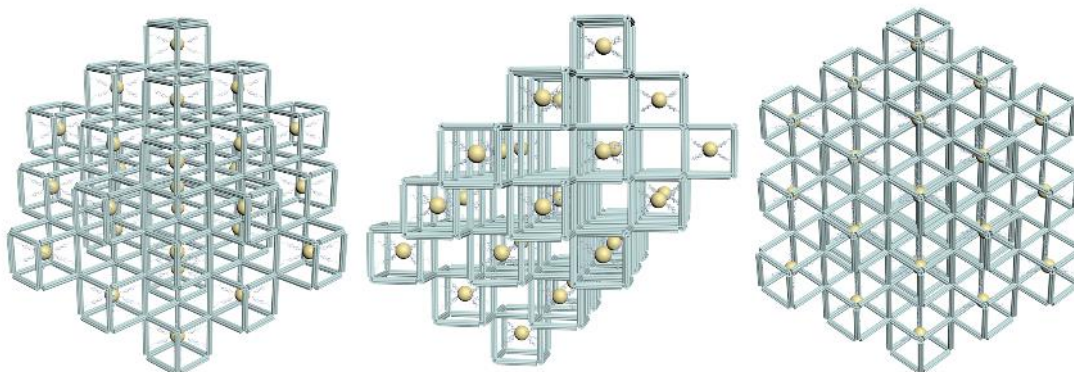


Figure S16. The close-packed models of body-centered cubic lattice formed by cubic frames in different angles.

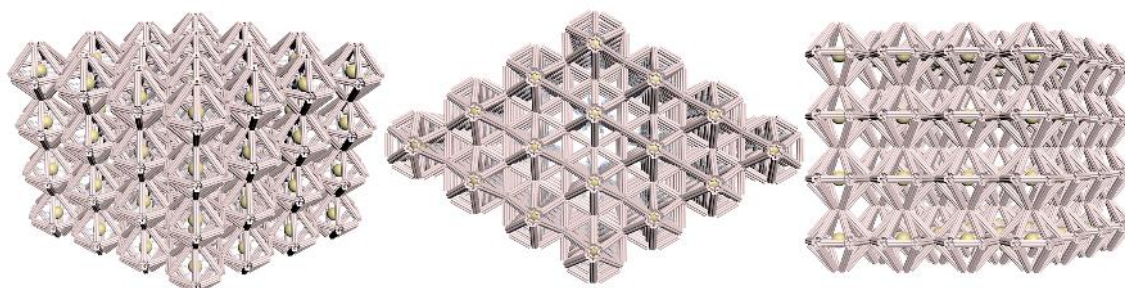


Figure S17. The close-packed models of simple hexagonal lattice formed by hexagonal bipyramid frames in different angles.

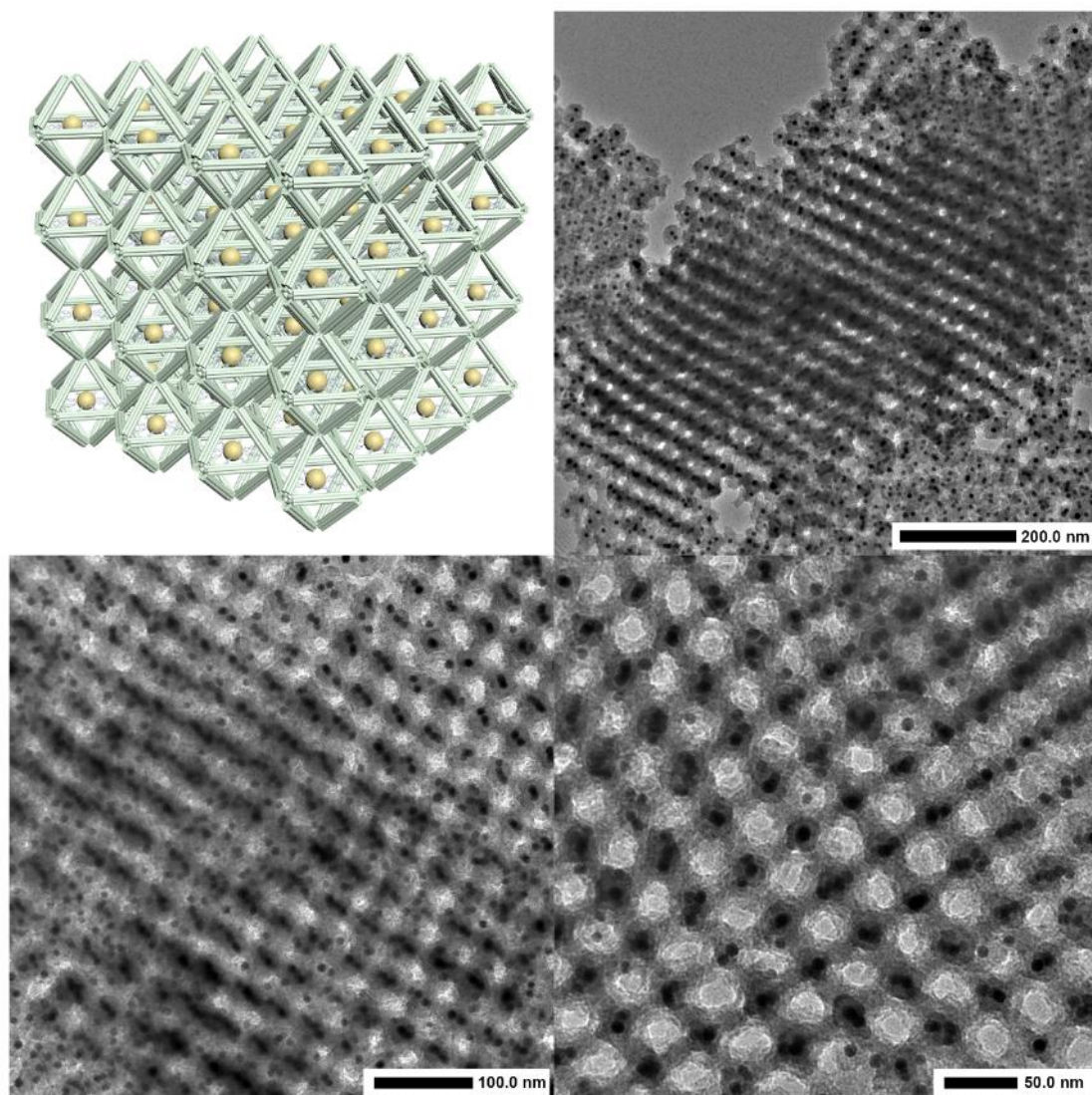


Figure S18. Representative TEM images of siliconized 3D lattices for octahedral frames. The schematic of the corresponding lattice model is shown on the upper left.

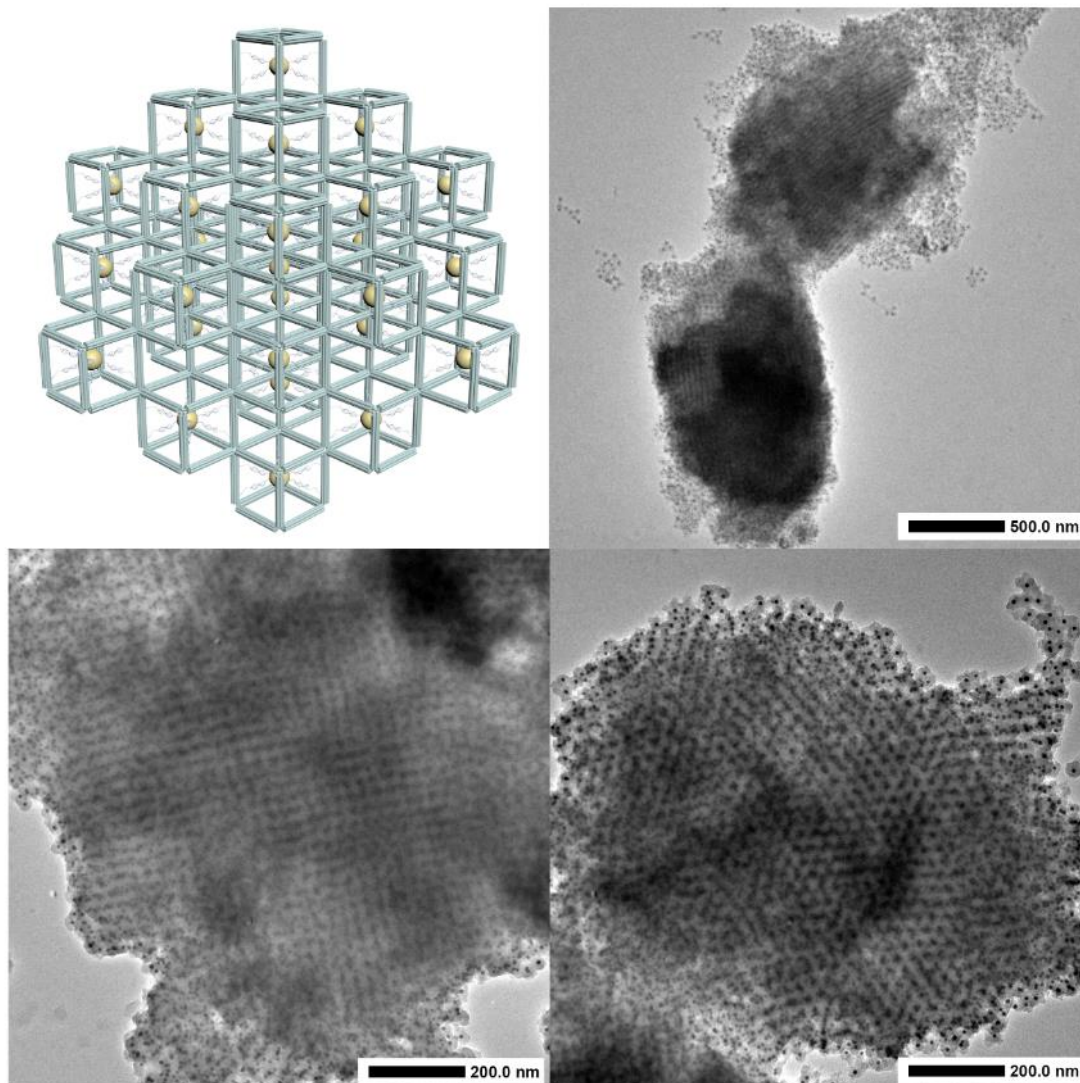


Figure S19. Representative TEM images of siliconized 3D lattices for cubic frames. The schematic of the corresponding lattice model is shown on the upper left.

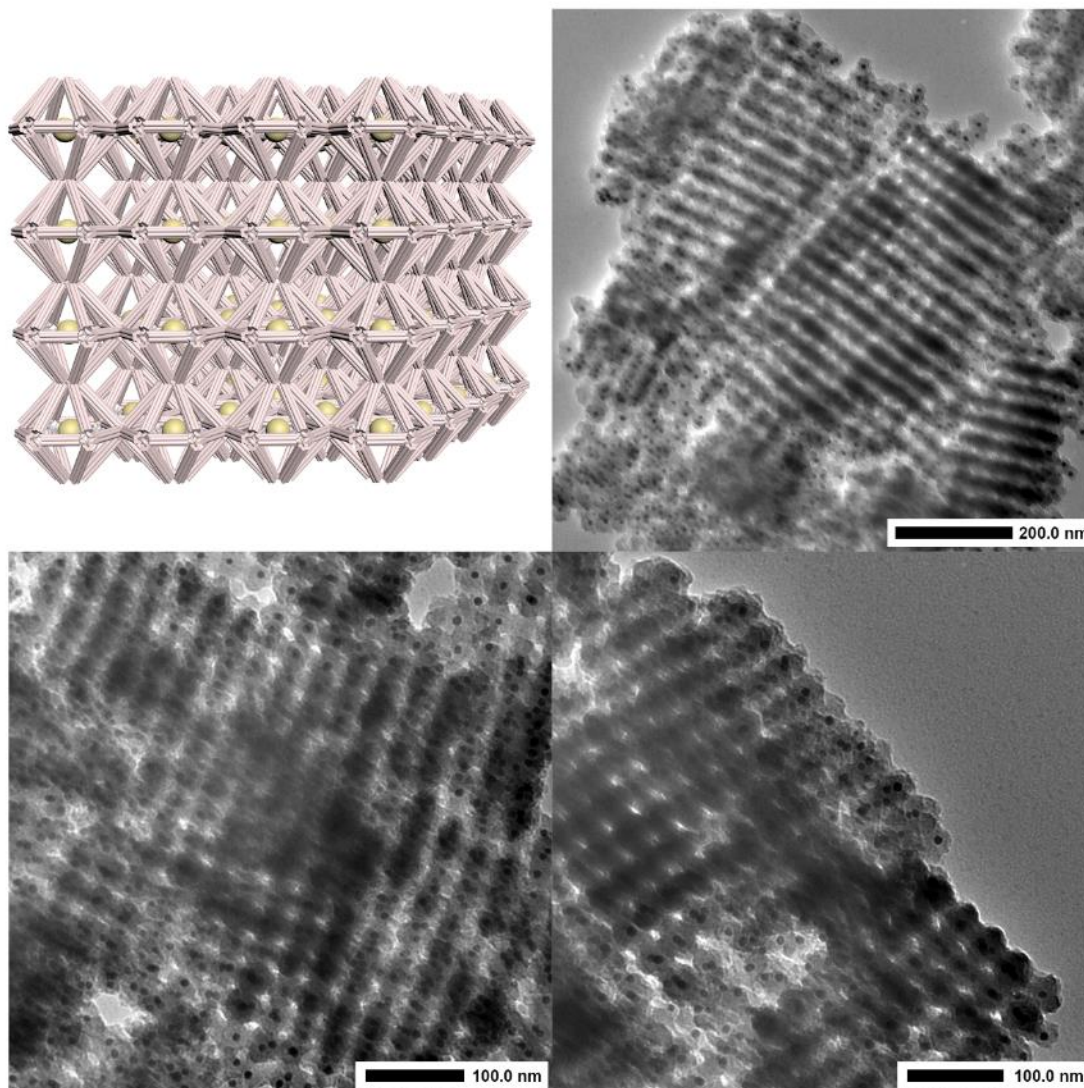


Figure S20. Representative TEM images of siliconized 3D lattices for hexagonal bipyramid frames. The schematic of the corresponding lattice model is shown on the upper left.

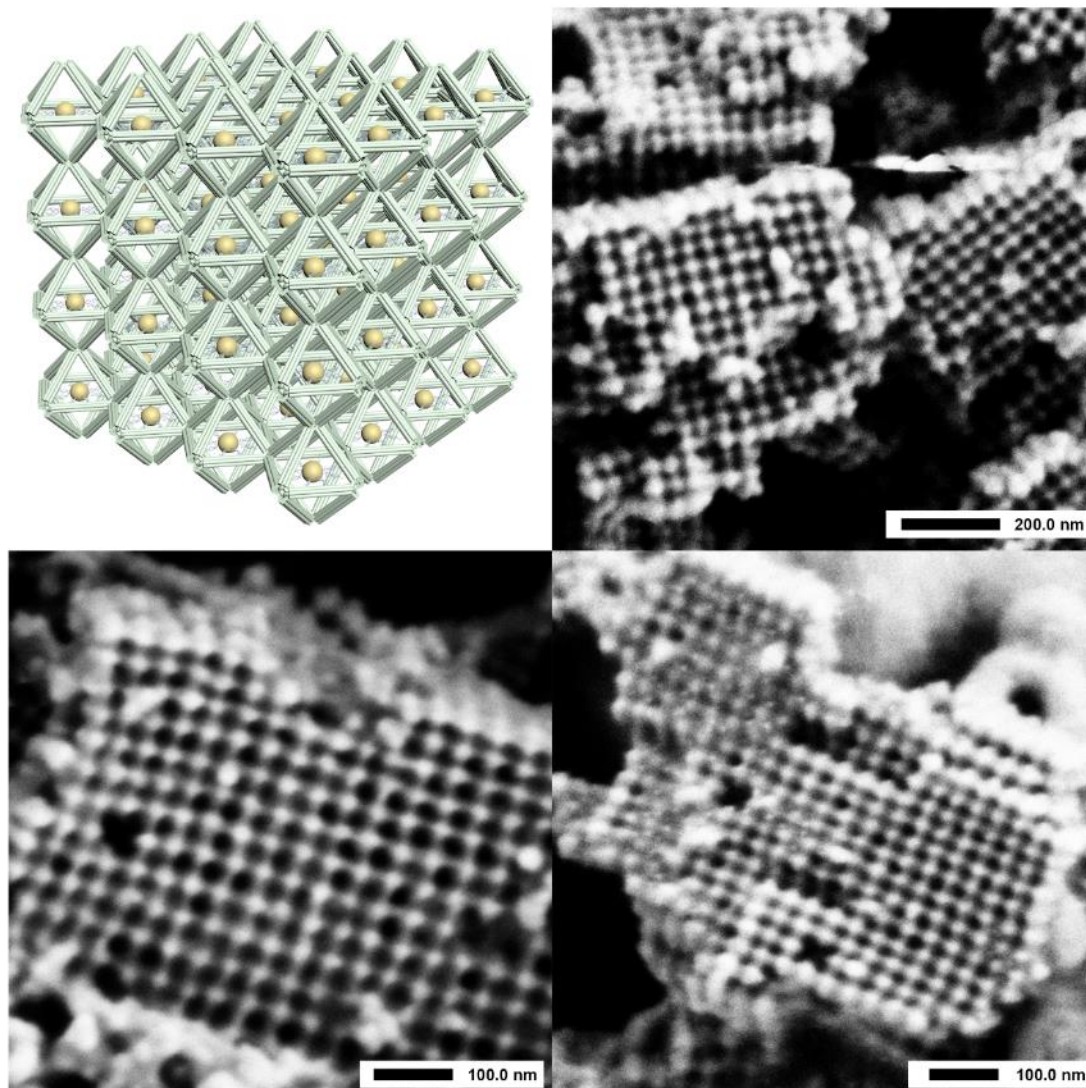


Figure S21. Representative SEM images of siliconized 3D lattices for octahedral frames. The schematic of the corresponding lattice model is shown on the upper left.

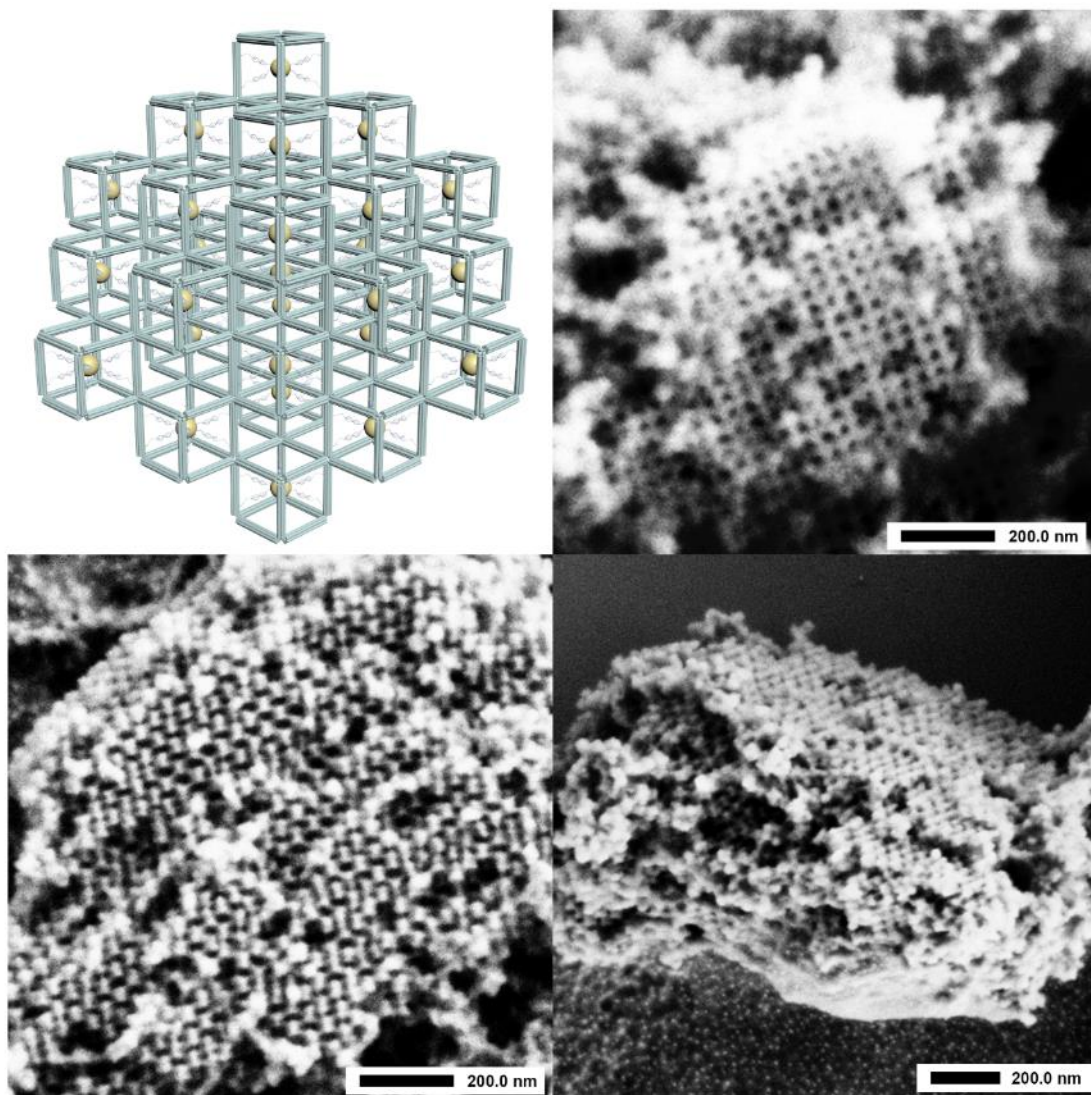


Figure S22. Representative SEM images of siliconized 3D lattices for cubic frames. The schematic of the corresponding lattice model is shown on the upper left.

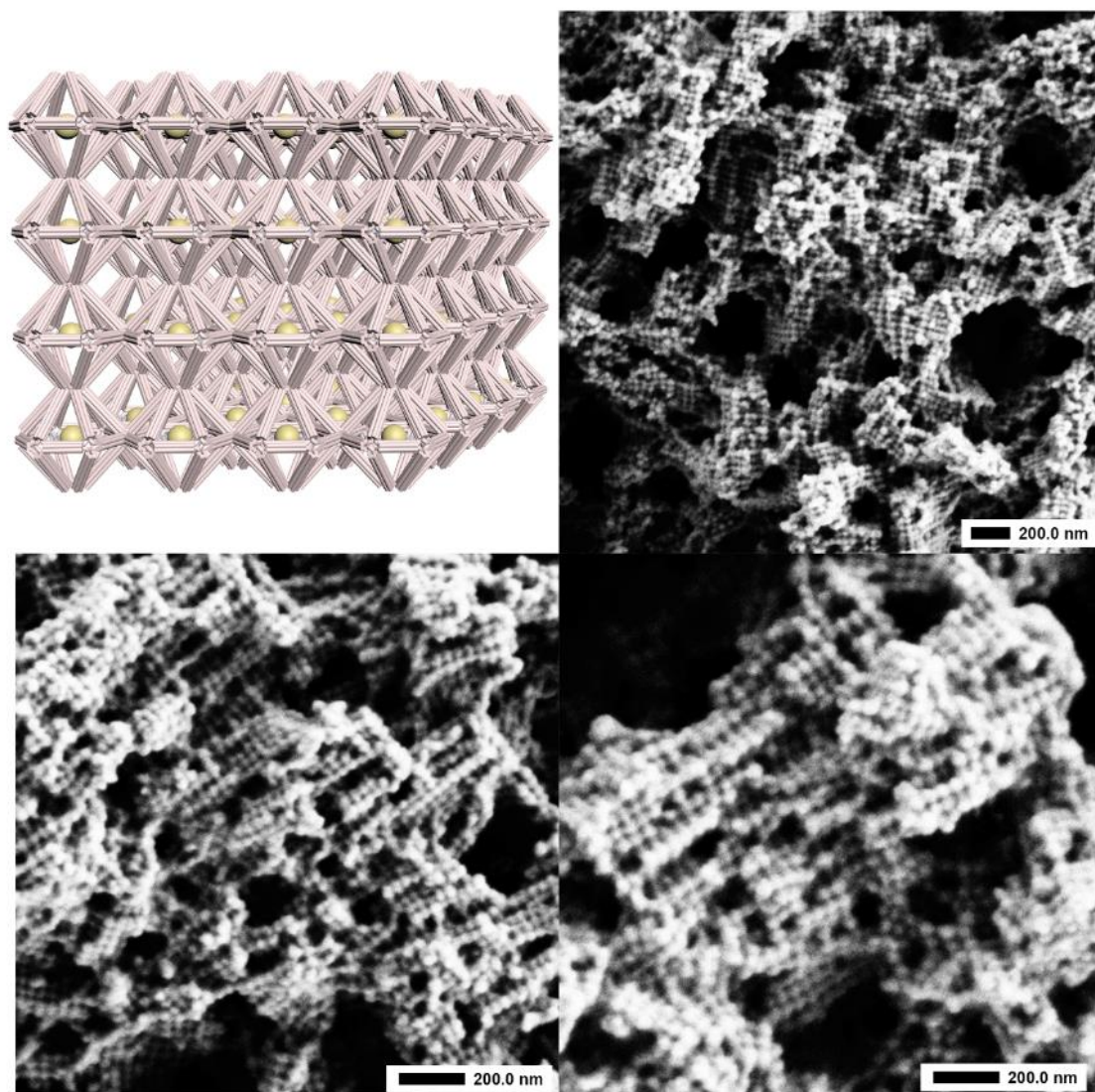


Figure S23. Representative SEM images of siliconized 3D lattices for hexagonal bipyramid frames. The schematic of the corresponding lattice model is shown on the upper left.

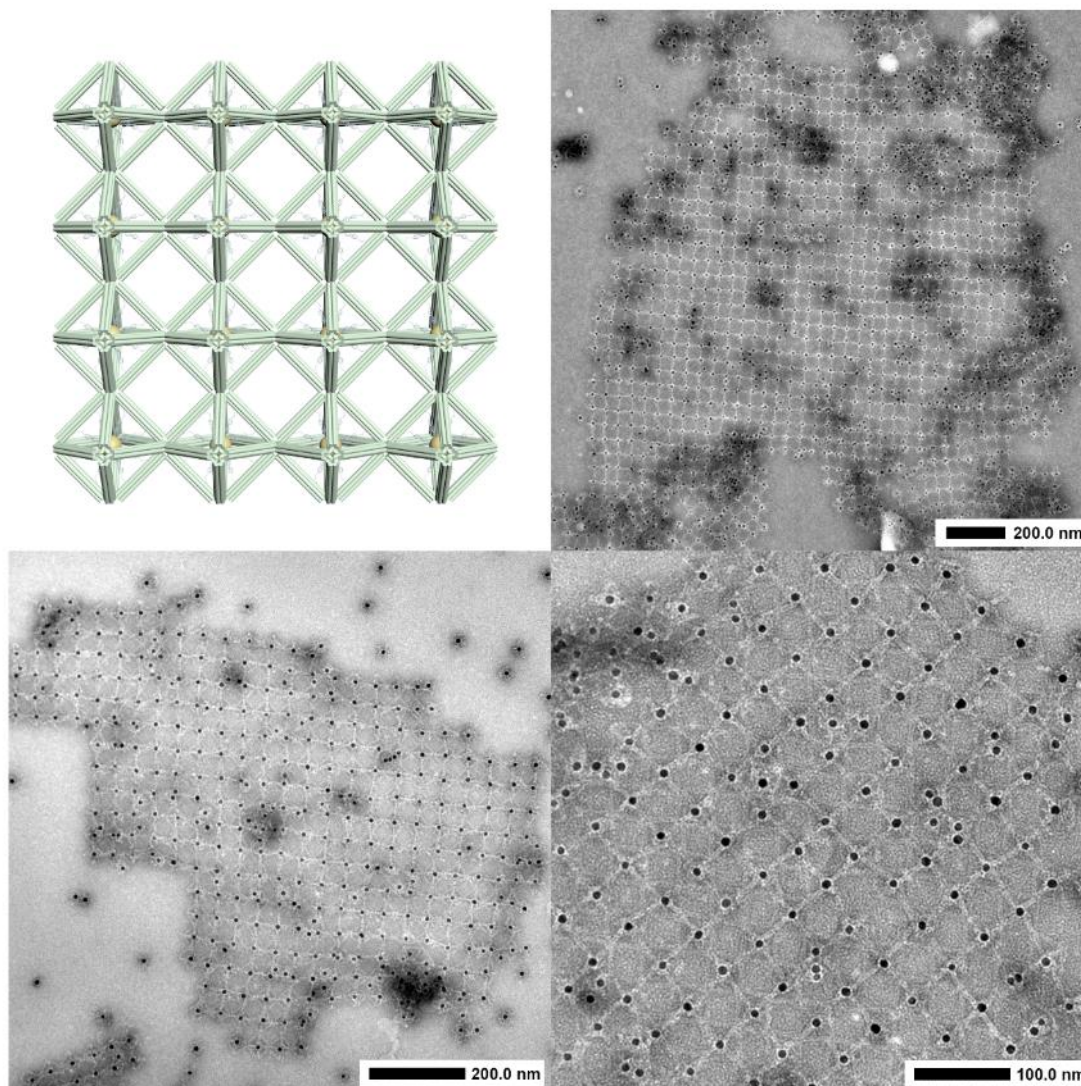


Figure S24. Representative negative-stained TEM images of 2D planar structures formed by AuNP-octahedron monomers. The corresponding schematic is shown on the upper left.

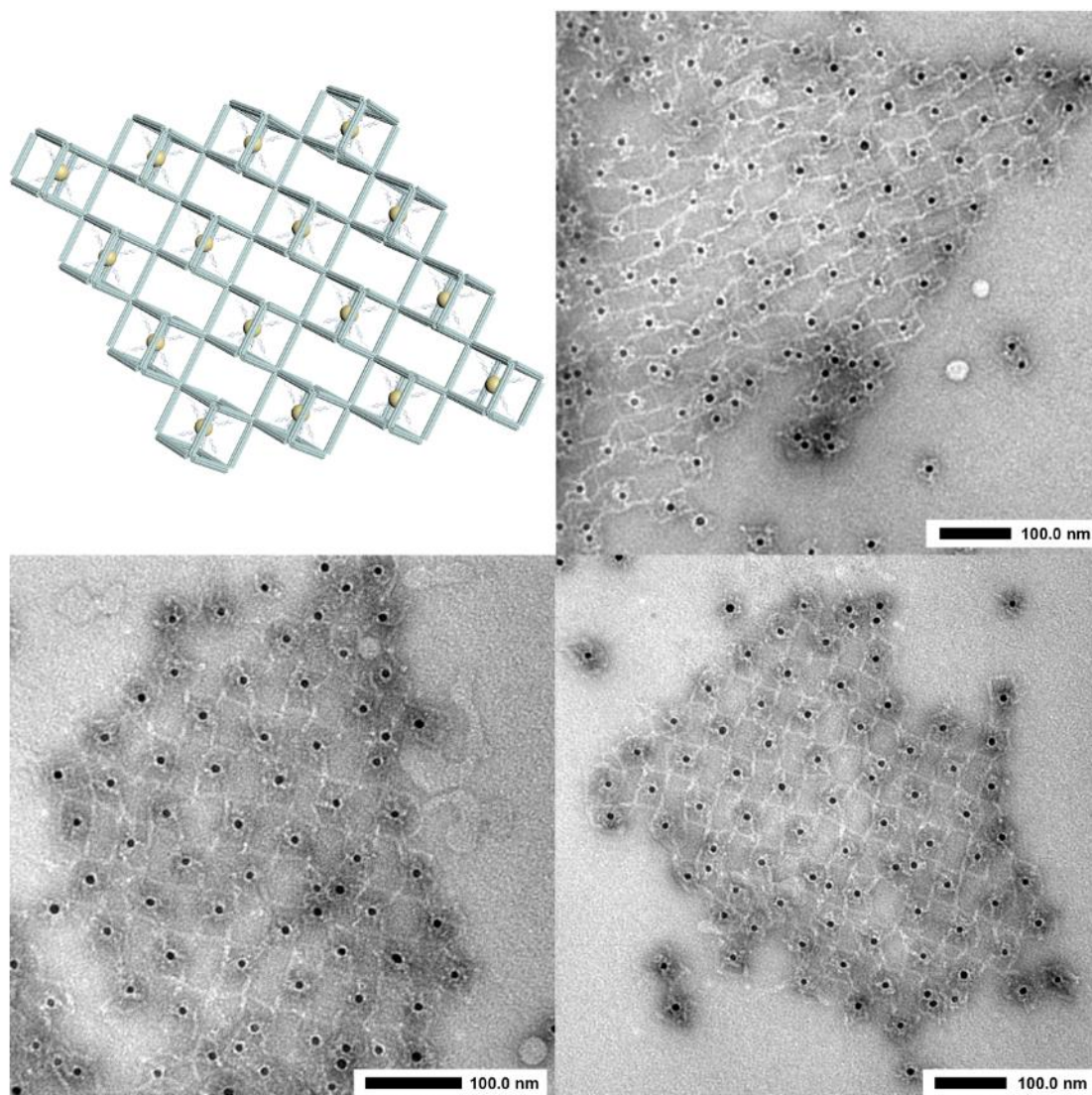


Figure S25. Representative negative-stained TEM images of 2D planar structures formed by AuNP-cube monomers. The corresponding schematic is shown on the upper left.

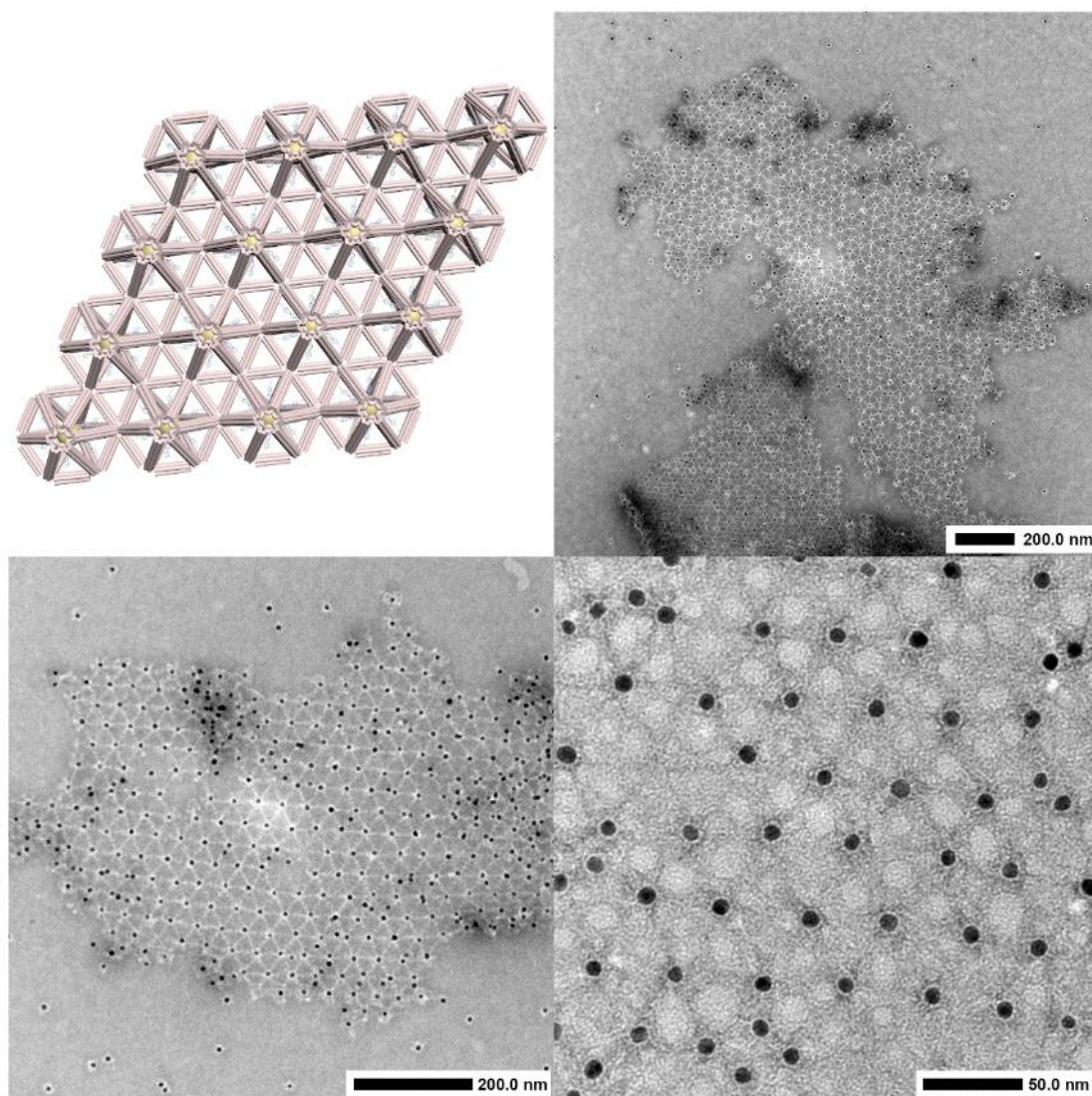


Figure S26. Representative negative-stained TEM images of 2D planar structures formed by AuNP-hexagonal bipyramid monomers. The corresponding schematic is shown on the upper left.

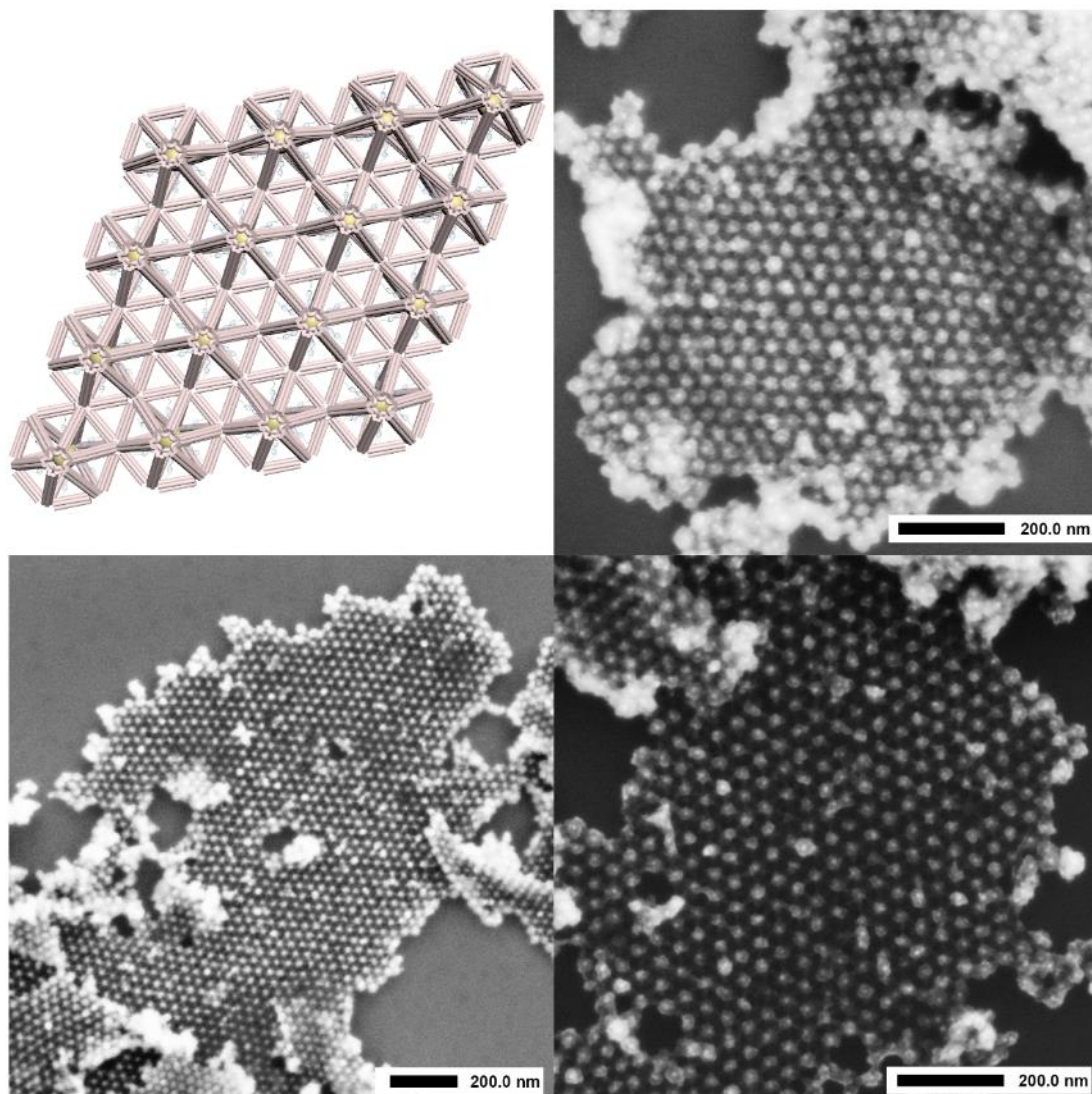


Figure S27. Representative SEM images of siliconized 2D planar structures formed by AuNP-hexagonal bipyramid monomers. The corresponding schematic is shown on the upper left.

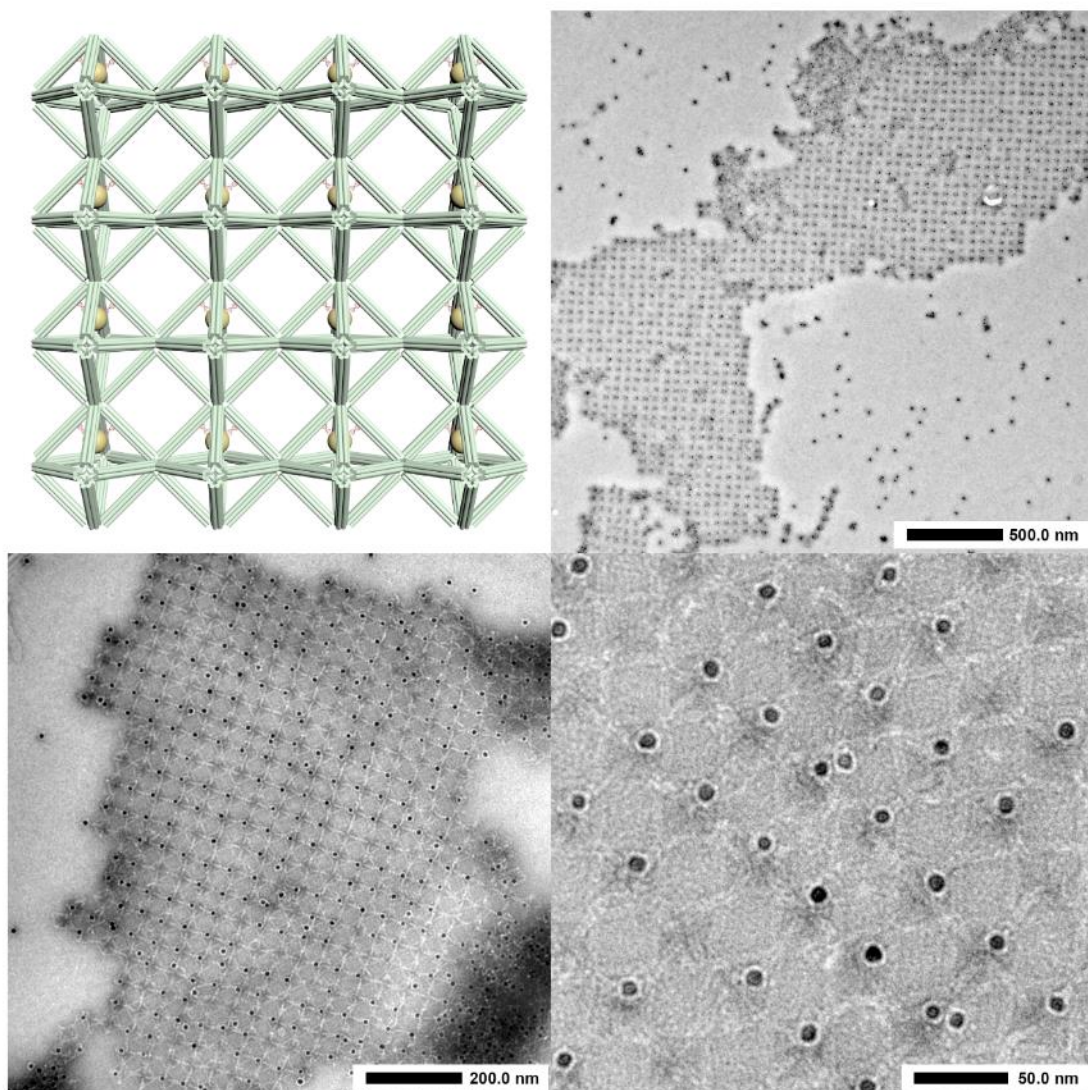


Figure S28. Representative negative-stained TEM images of 2D planar structures for the conjugated monomer with AuNPs near the vertex. The corresponding schematic is shown on the upper left.

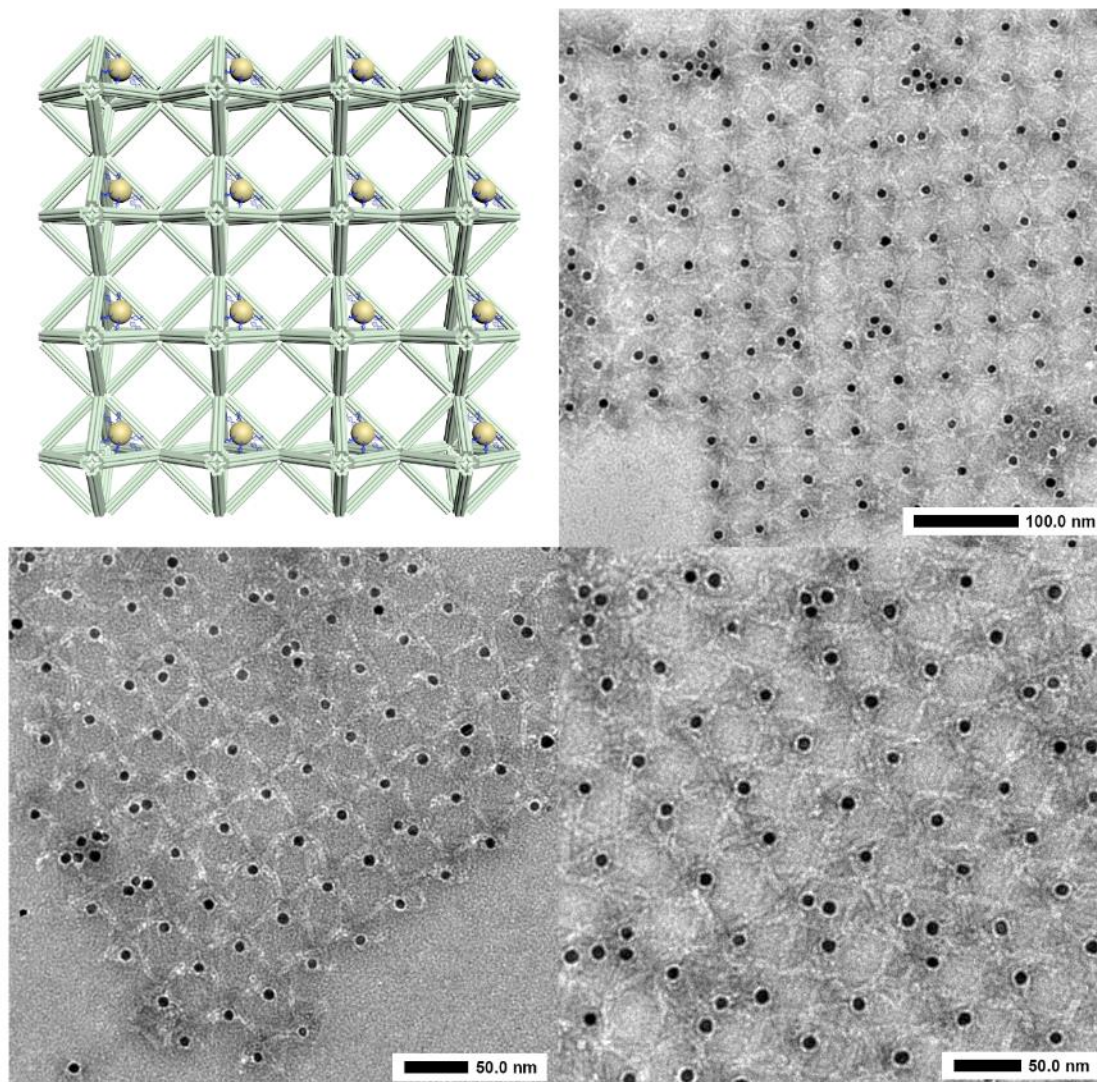


Figure S29. Representative negative-stained TEM images of 2D planar structures for the conjugated monomer with AuNPs in the face center. The corresponding schematic is shown on the upper left.

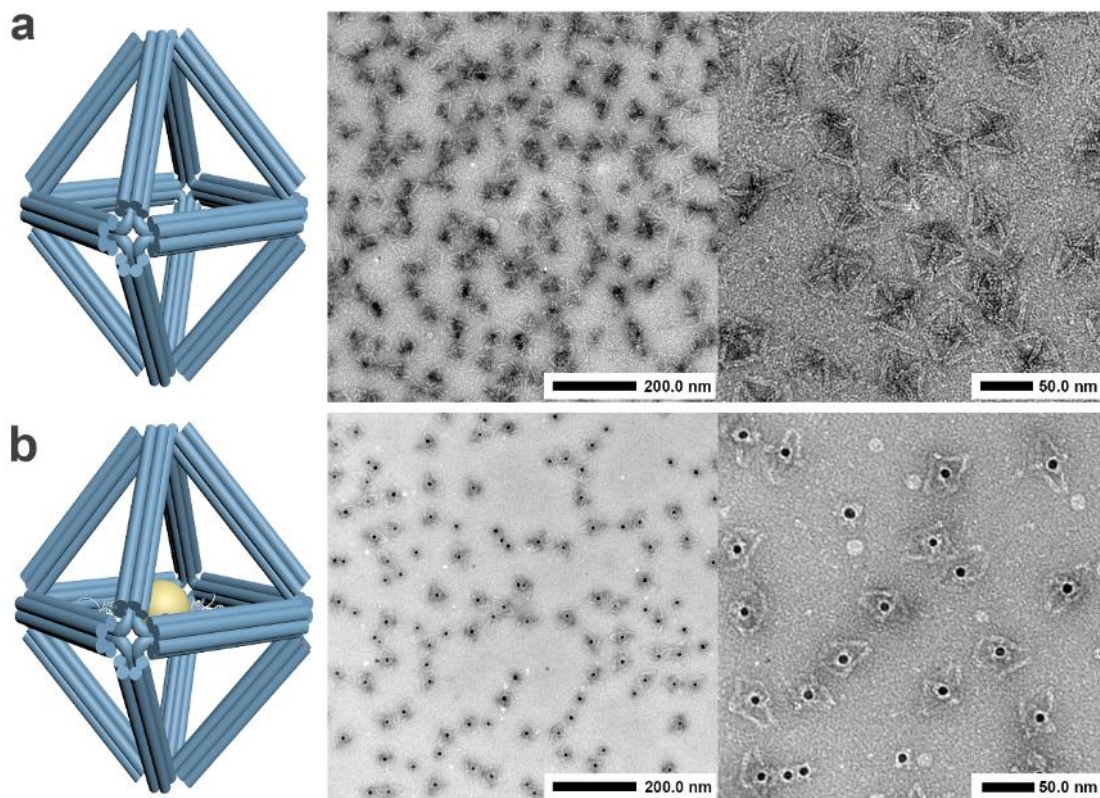


Figure S30. a) Representative negative-stained TEM images of elongated octahedral DNA origami frames. b) Representative negative-stained TEM images of AuNP-elongated octahedron monomers. Corresponding schematics are shown on the left.

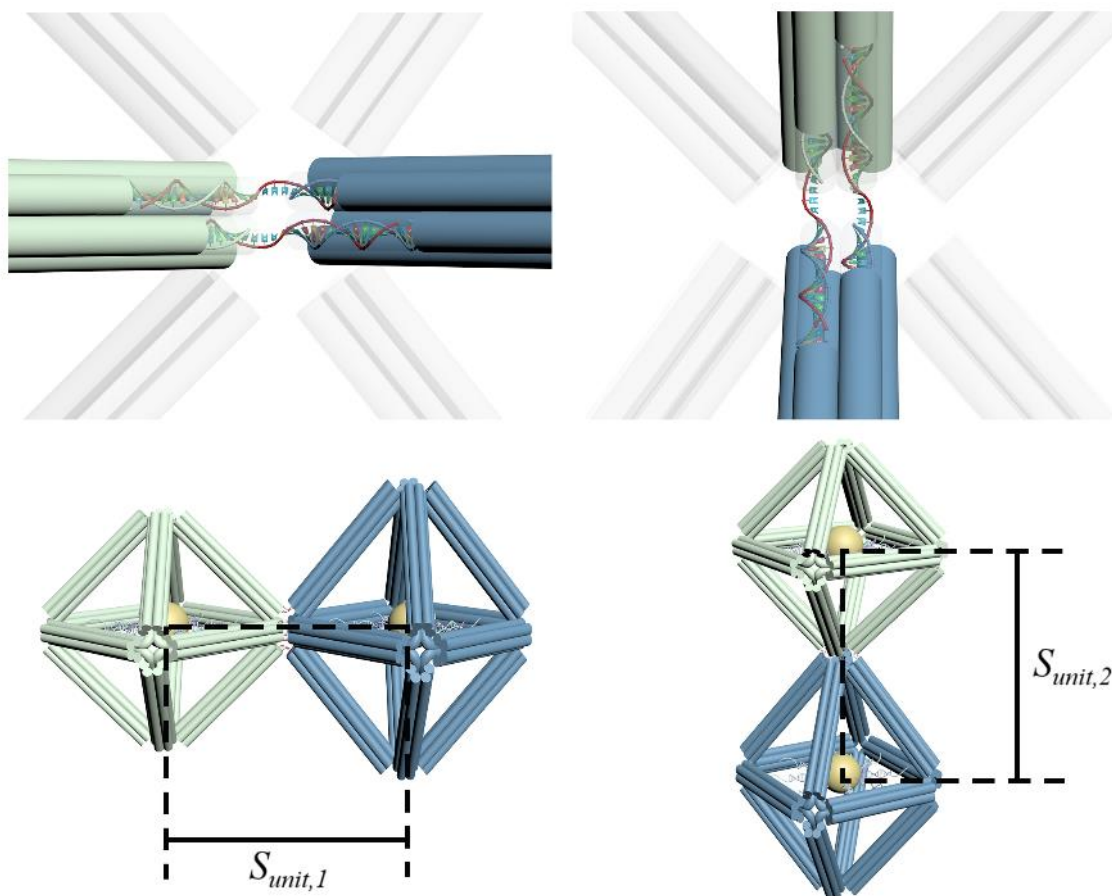


Figure S31. The magnified schematics of the bridge pattern for the binary system at vertex junctions. $S_{unit,1}$ and $S_{unit,2}$ respectively represents the distance between the body centers of adjacent frames in non-identical crystallographic axis directions.

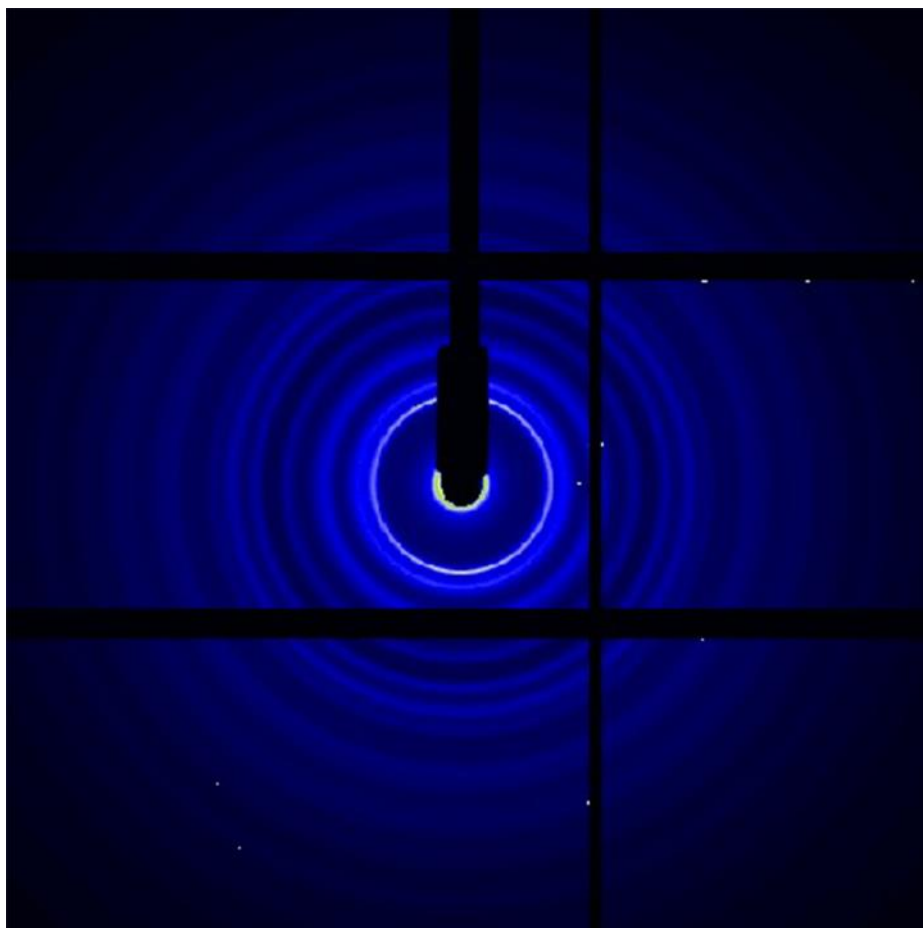


Figure S32. 2D pattern of the AuNP lattice in the binary system measured by SAXS.

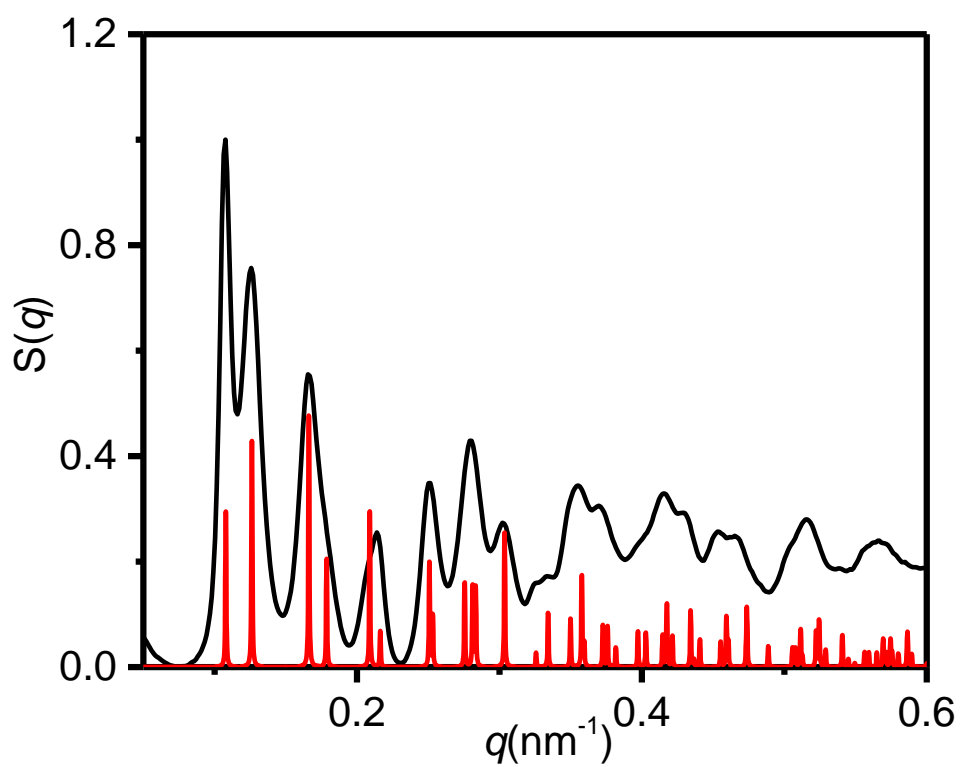


Figure S33. Enlargement of structure factor result for the tetragonal lattice shown in Figure 5b: black curve for the experimental data and red curve for the model.

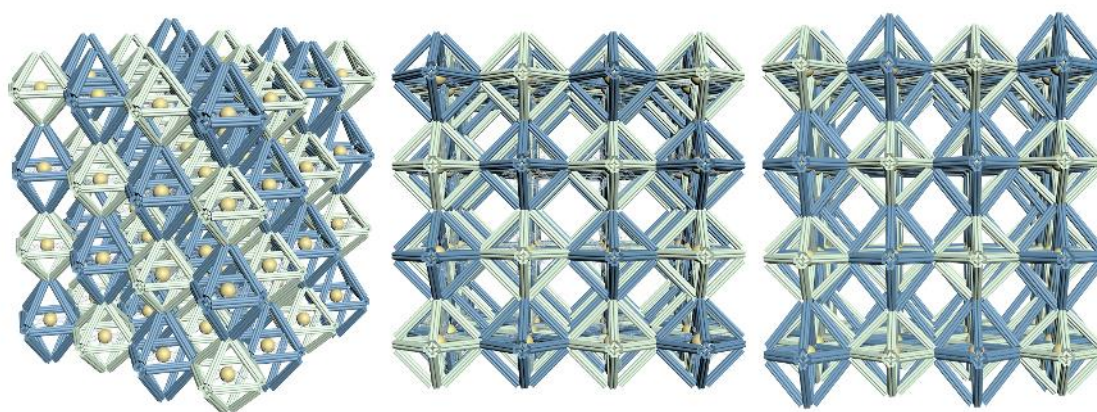


Figure S34. The close-packed models of tetragonal lattice of the binary system in different angles.

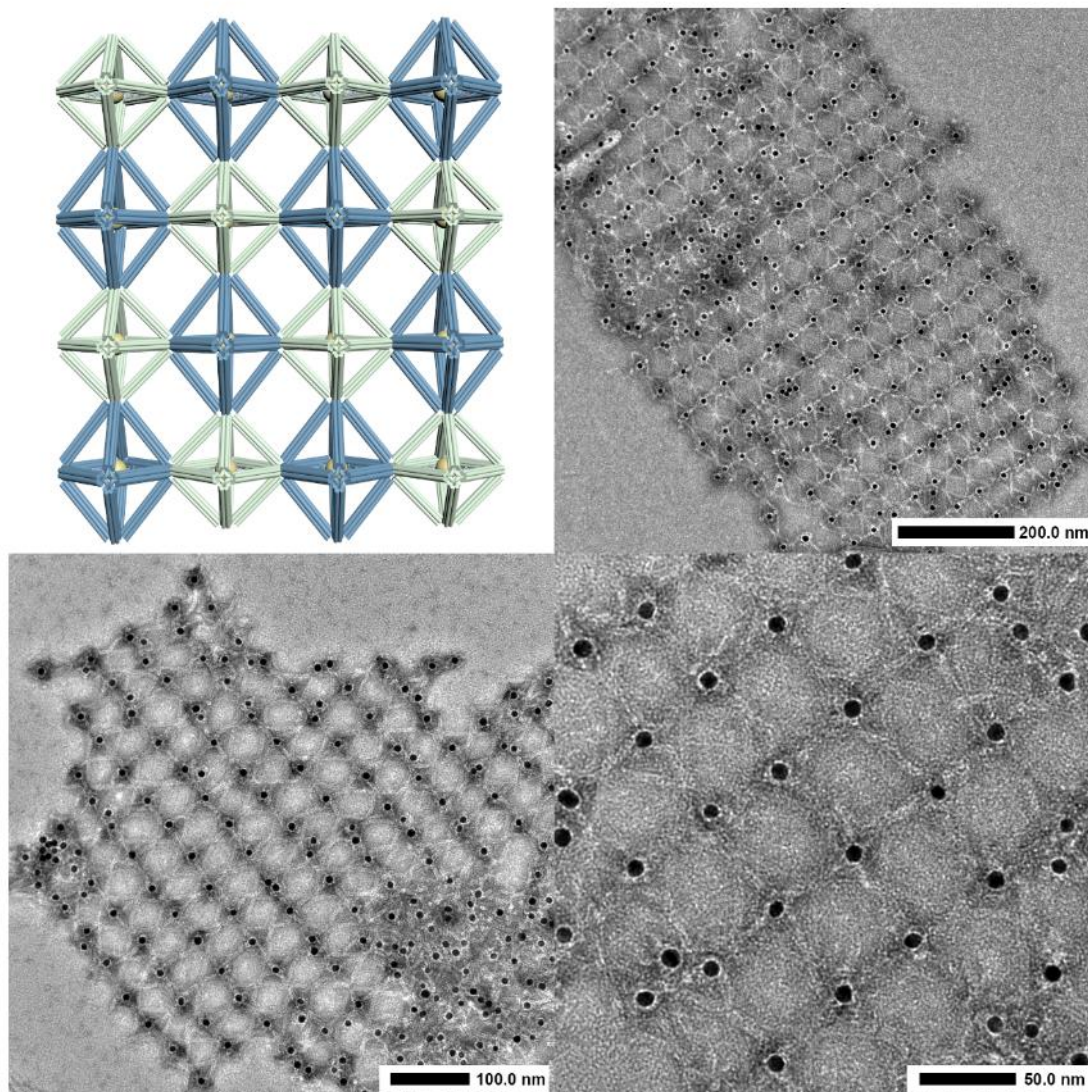


Figure S35. Representative negative-stained TEM images of 2D planar structures of the binary system. The corresponding schematic is shown on the upper left.

Assembly method	Bridge pattern		Sticky-end hybridization	
			(Nat. Mater., 2020)	
Frame shape	Octahedron	Cube	Octahedron	Cube
Lattice type	SC	BCC	SC	BCC
Identifiable peaks in SAXS	~15	~18	~25	~15
Average size of ordered grains	~ 1 μm		~ 3-5 μm	
Anisotropy control	Yes		No	

Table S1. Comparison of bridging method and sticky-end hybridization method (Nat. Mater., 2020, 19, 789-796.).

References

1. X. Liu, F. Zhang, X. Jing, M. Pan, P. Liu, W. Li, B. Zhu, J. Li, H. Chen, L. Wang, J. Lin, Y. Liu, D. Zhao, H. Yan and C. Fan, *Nature*, 2018, 559, 593-598.
2. Y. Tian, J. R. Lhermitte, L. Bai, T. Vo, H. L. Xin, H. Li, R. Li, M. Fukuto, K. G. Yager, J. S. Kahn, Y. Xiong, B. Minevich, S. K. Kumar and O. Gang, *Nat. Mater.*, 2020, 19, 789-796.