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Supplementary Information

Stepwise Assembly of Nanocluster Guided by DNA Origami Frames with High-throughput

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1. Materials and methods:

Folding DNA origami frames: Octahedral DNA origami frames (DOFs) were folded by mixing long scaffold DNA chains and ~120 different short staple DNA sequences by one-step annealing process. Briefly, 10 nM scaffold DNA (M13mp18, Bayou Biolab) were mixed with 100 nM of each staple DNA in 1×TAE buffer containing 12.5 mM Mg^{2+}. The mixture was firstly heated to 90 °C rapidly and then slowly cooling down to 20 °C over 21 hours using polymerase chain reaction (PCR) machine. The product was then stored at 4 °C and ready for use. Note that the DOFs used in the 1st and 2nd strategies were slightly different in the extracted DNA sticky ends from vertices, as shown in the sequence section.

DNA functionalized on Au nanoparticles: Au colloids we purchased from Ted Pella were 10 nm gold nanoparticles (NPs) functionalized with citric acid. The Au nanoparticles (AuNPs) used in both two strategies were modified with different thiolated DNA: the 1st strategy used NPs functionalized with thiolated DNA *A* and *B* together, and the 2nd strategy used NPs functionalized with thiolated DNA *F* (see the sequence section for details). For the 1st strategy, thiolated DNA *A* and *B* were added to the AuNPs with molar ratio of 150:1 between DNA and AuNPs, respectively. For the 2nd strategy, thiolated DNA *F* were added to the AuNPs with molar ratio of 300:1 between DNA and AuNPs. After 1 h, the sodium dodecyl sulfate (SDS) and phosphate buffer (PB, 100 mM) were added to the solution to bring to a concentration of 0.01% for SDS and 10 mM for phosphate. After another 1 h, 2 M NaCl was gradually added over 5 hours to a final concentration of 0.3 M. Then the solution was put on rotating apparatus overnight. The DNA functionalized AuNPs were purified by centrifuging for 1 h at 20,000 rcf to remove the supernatant containing unreacted DNA. After repeating the centrifugation procedure three times, the AuNPs were finally resuspended in 0.1 M PBS buffer. The concentration of the AuNPs was measured by UV-vis spectroscope.

Fabrication of DNA/magnetic beads conjugates: Streptavidin capped on magnetic beads (MBs, ~1 μm) used in this experiment were purchased from New England Biolabs. Biotin-modified ssDNA *A'* and buffer (12.5 mM Mg^{2+}, 1×TAE) were mixed with MBs, and rotated at room temperature for 2 hours. After that, the mixture was rinsed several times to remove the unattached ssDNA *A'*.

Transmission electron microscope (TEM) image: The carbon coated copper grids were glow discharged for 30 s before use. The 5 μL sample solution was dropped onto a carbon coated copper grid and deposited for 20 min, and then the excess solution was slowly wicked away with filter paper. After rinsing by buffer (12.5 mM Mg^{2+}, 1×TAE) twice, the sample was negatively stained by 5 μL 2% (weight/volume) uranyl acetate aqueous solution for 15 s and the excess staining solution was immediately removed. The sample was imaged in JEOL-2100 TEM operating at 200 kV.

DLS measurement: The instrument we used was Malvern Zetasizer Nano-ZSE, which was equipped with a 633 nm laser source and based on the principle of dynamic light scattering (DLS) to measure the hydrodynamic size of nanoclusters.

Brief protocol: In the 1st strategy, we firstly capped both sulfhydryl DNA *A* and *B* on the AuNPs. In step 1, the modified AuNPs called *p-AB* were added into the solution containing *s-A'* surface. AuNPs could bind with the surface of MBs tightly by the interactions between sticky end *A* and *A'*. After rotating for 12 h at room temperature, the supernatant was removed and the sample was rinsed four times with buffer (12.5 mM Mg^{2+}, 1×TAE). In step 2, we added ssDNA *B'C'* to the rinsed solution at a molar ratio of 3:1 for ssDNA *B'C'* : AuNPs. The solution was shaken at 1,000 rpm for 12 h at 35 °C and then rinsed four times with method similar as mentioned above. In this procedure, complementary base-pairings between *B'C'* and *B* only occurred on the top hemisphere of the AuNPs and was not affected by severe steric hindrance. After sufficient reaction time, we added the substituted chain *A''* (molar ratio of *A''* and *A'* was 5:1), which contained three more complementary bases than *A*, to separate patchy particles from the surface. Then, we put the solution of octahedral DOFs which had two vertices stretched out with DNA chains *F* into the solution we obtained from step 3 at a 1:3 molar ratio, and shook at 1,000 rpm for 12 h at 38 °C (step 4). After 12 h of adequate reaction, the dimes were produced.

In the 2nd strategy, the DOFs had one specific vertex stretching out DNA sticky ends *O*, which were complementary with DNA *O'* on the surface MBs (Figure 3a). In step 1, 5 μL (10 nM) DOFs were added into MBs solution and reacted for at least 12 h to be installed on *s-O'* surface. Another four vertices in the middle-plane were designed to 'active' (*F*) for grasping the AuNPs. We added AuNPs functioned with strand *F'* into the solution at a 10:1 molar ratio for AuNPs and DOFs. The mixture was shaken at 1,000 rpm for 12 h at 35 °C in step 2. After that, the solution was rinsed for several times. Finally, 'fuel DNA' *O''* was put into the solution to release the clusters, which were then separated from the surface by magnets.

DNA sequences: DNA oligonucleotides we used were ordered from Sangon Biotech (Shanghai, China). The thiolated DNA strands were purified by HPLC, while the other DNA strands were purified by PAGE. The DOFs we designed have totally six vertices, labeled from Octa Ⅰ to Octa Ⅵ (as shown in the Figure S7). Each vertex stretched out four sticky ends for assembly with NPs. The sequences were listed as follows (5' to 3').

(1). DNA sequences used in the 1st strategy

The DOFs used in the 1st strategy had two 'active' vertices: Octa Ⅱ and Octa Ⅳ.

Name	Sequence
A	ATTGGATTGGAAGTATTTTTTTTTTTTTTT-C6H12-SH

B	CTCTCTACACTATCTTTTTTTTTTTTTTTT-C6H12-SH
B'C'	AGATAGTGTAGAGAGAGTATTGATAAGGAT
A'	CTTGTGTCTACTTCCAATCCAATTTTTTTTTTTTTTTT-Biotin
A''	ATTGGATTGGAAGTAGACACAAGAA
Octa Ⅱ-C_1	AATAGCAATAGCACCAGAAGGAAACCTAAAGCCACTGGTAATTTATCCTTATCAATACT
Octa Ⅱ-C_2	GACAGGAGGTTGAAACAAATAAATCCGCCCCCTCCGCCACCCTTATCCTTATCAATACT
Octa Ⅱ-C_3	CAGAATCAAGTTTCGGCATTTTCGGTTAAATATATCACCAGTTTATCCTTATCAATACT
Octa Ⅱ-C_4	TCATATGGTTTACGATTGAGGGAGGGAAACGCAATACATACATTATCCTTATCAATACT
Octa Ⅳ-C_1	GCTCACAATTCCGTGAGCTAACTCACTGGAAGTAATGGTCAATTATCCTTATCAATACT
Octa Ⅳ-C_2	GGCCCTGAGAGAAGCAGGCGAAAATCATTGCGTAGAGGCGGTTTATCCTTATCAATACT
Octa Ⅳ-C_3	TTTGCGGATGGCCAACTAAAGTACGGGCTTGCAGCTACAGAGTTATCCTTATCAATACT
Octa Ⅳ-C_4	CTTAAACAGCTTATATATTCGGTCGCTTGATGGGGAACAAGATTATCCTTATCAATACT

(2). DNA sequences used in the 2nd strategy

The DOFs used in the 2nd strategy had five vertexes: Octa Ⅰ, Octa Ⅱ, Octa Ⅲ, Octa Ⅳ and Octa Ⅴ.

Name	Sequence
O'	Biotin-TTTTTTTTTTTTTTTCTCTCTTCTATCCTAACCTTCAT
O''	ATGAAGGTTAGGATAGAAGAGAG
F'	SH-C6H12- TTTAGTATTGATAAGGAT
Octa Ⅰ-O_1	AGAGCCTAATTTGATTTTTTGTTTAAATCCTGAAATAAAGAATTTTTTTTTATGAAGGTTAGGATA
Octa Ⅰ-O_2	TGTAGCATTCCAACGTTAGTAAATGAAGTGCCGCGCCACCCTTTTTTTTTTTATGAAGGTTAGGATA
Octa Ⅱ-F_1	AATAGCAATAGCACCAGAAGGAAACCTAAAGCCACTGGTAATTTATCCTTATCAATACT
Octa Ⅱ-F_2	GACAGGAGGTTGAAACAAATAAATCCGCCCCCTCCGCCACCCTTATCCTTATCAATACT
Octa Ⅱ-F_3	CAGAATCAAGTTTCGGCATTTTCGGTTAAATATATCACCAGTTTATCCTTATCAATACT
Octa Ⅱ-F_4	TCATATGGTTTACGATTGAGGGAGGGAAACGCAATACATACATTATCCTTATCAATACT
Octa Ⅲ-F_1	CAACGCTCAACAGCAGAGGCATTTTCAATCCAATGATAAATATTATCCTTATCAATACT
Octa Ⅲ-F_2	ATCAAAATCATATATGTAAATGCTGAACAAACACTTGCTTCTTTATCCTTATCAATACT
Octa Ⅲ-F_3	TGATTGCTTTGAGCAAAAGAAGATGAAATAGCAGAGGTTTTGTTATCCTTATCAATACT
Octa Ⅲ-F_4	AACGGGTATTAAGGAATCATTACCGCCAGTAATTCAACAATATTATCCTTATCAATACT
Octa Ⅳ-F_1	GCTCACAATTCCGTGAGCTAACTCACTGGAAGTAATGGTCAATTATCCTTATCAATACT
Octa Ⅳ-F_2	GGCCCTGAGAGAAGCAGGCGAAAATCATTGCGTAGAGGCGGTTTATCCTTATCAATACT
Octa Ⅳ-F_3	TTTGCGGATGGCCAACTAAAGTACGGGCTTGCAGCTACAGAGTTATCCTTATCAATACT
Octa Ⅳ-F_4	CTTAAACAGCTTATATATTCGGTCGCTTGATGGGGAACAAGATTATCCTTATCAATACT
Octa Ⅴ-F_1	CAAATGCTTTAAAAAATCAGGTCTTTAAGAGCAGCCAGAGGGTTATCCTTATCAATACT
Octa Ⅴ-F_2	CTTCATCAAGAGAAATCAACGTAACAGAGATTTGTCAATCATTTATCCTTATCAATACT
Octa Ⅴ-F_3	AAAGATTCATCAGGAATTACGAGGCATGCTCATCCTTATGCGTTATCCTTATCAATACT
Octa Ⅴ-F_4	AAACGAAAGAGGGCGAAACAAAGTACTGACTATATTCGAGCTTTATCCTTATCAATACT

(3). Staple DNA for octahedral DOFs (Octa-1 to Octa-120)

Octa-1	TCAAAGCGAACCAGACCGTTTTATATAGTC
Octa-2	GCTTTGAGGACTAAAGAGCAACGGGGAGTT
Octa-3	GTAAATCGTCGCTATTGAATAACTCAAGAA
Octa-4	AAGCCTTAAATCAAGACTTGCGGAGCAAAT
Octa-5	ATTTTAAGAACTGGCTTGAATTATCAGTGA
Octa-6	GTTAAAATTCGCATTATAAACGTAAACTAG
Octa-7	AGCACCATTACCATTACAGCAAATGACGGA
Octa-8	ATTGCGTAGATTTTCAAAACAGATTGTTTG
Octa-9	TAACCTGTTTAGCTATTTTCGCATTCATTC
Octa-10	GTCAGAGGGTAATTGAGAACACCAAAATAG
Octa-11	CTCCAGCCAGCTTTCCCCTCAGGACGTTGG
Octa-12	GTCCACTATTAAAGAACCAGTTTTGGTTCC
Octa-13	TAAAGGTGGCAACATAGTAGAAAATAATAA
Octa-14	GATAAGTCCTGAACAACTGTTTAAAGAGAA
Octa-15	GGTAATAGTAAAATGTAAGTTTTACACTAT
Octa-16	TCAGAACCGCCACCCTCTCAGAGTATTAGC
Octa-17	AAGGGAACCGAACTGAGCAGACGGTATCAT
Octa-18	GTAAAGATTCAAAAGGCCTGAGTTGACCCT
Octa-19	AGGCGTTAAATAAGAAGACCGTGTCGCAAG
Octa-20	CAGGTCGACTCTAGAGCAAGCTTCAAGGCG
Octa-21	CAGAGCCACCACCCTCTCAGAACTCGAGAG
Octa-22	TTCACGTTGAAAATCTTGCGAATGGGATTT
Octa-23	AAGTTTTAACGGGGTCGGAGTGTAGAATGG
Octa-24	TTGCGTATTGGGCGCCCGCGGGGTGCGCTC
Octa-25	GTCACCAGAGCCATGGTGAATTATCACCAATCAGAAAAGCCT
Octa-26	GGACAGAGTTACTTTGTCGAAATCCGCGTGTATCACCGTACG
Octa-27	CAACATGATTTACGAGCATGGAATAAGTAAGACGACAATAAA
Octa-28	AACCAGACGCTACGTTAATAAAACGAACATACCACATTCAGG
Octa-29	TGACCTACTAGAAAAAGCCCCAGGCAAAGCAATTTCATCTTC
Octa-30	TGCCGGAAGGGGACTCGTAACCGTGCATTATATTTTAGTTCT
Octa-31	AGAACCCCAAATCACCATCTGCGGAATCGAATAAAAATTTTT
Octa-32	GCTCCATTGTGTACCGTAACACTGAGTTAGTTAGCGTAACCT
Octa-33	AGTACCGAATAGGAACCCAAACGGTGTAACCTCAGGAGGTTT
Octa-34	CAGTTTGAATGTTTAGTATCATATGCGTAGAATCGCCATAGC
Octa-35	AAGATTGTTTTTTAACCAAGAAACCATCGACCCAAAAACAGG
Octa-36	TCAGAGCGCCACCACATAATCAAAATCAGAACGAGTAGTATG
Octa-37	GATGGTTGGGAAGAAAAATCCACCAGAAATAATTGGGCTTGA

Octa-38	CTCCTTAACGTAGAAACCAATCAATAATTCATCGAGAACAGA
Octa-39	AGACACCTTACGCAGAACTGGCATGATTTTCTGTCCAGACAA
Octa-40	GCCAGCTAGGCGATAGCTTAGATTAAGACCTTTTTAACCTGT
Octa-41	CCGACTTATTAGGAACGCCATCAAAAATGAGTAACAACCCCA
Octa-42	GTCCAATAGCGAGAACCAGACGACGATATTCAACGCAAGGGA
Octa-43	CCAAAATACAATATGATATTCAACCGTTAGGCTATCAGGTAA
Octa-44	AACAGTACTTGAAAACATATGAGACGGGTCTTTTTTAATGGA
Octa-45	TTTCACCGCATTAAAGTCGGGAAACCTGATTTGAATTACCCA
Octa-46	GAGAATAGAGCCTTACCGTCTATCAAATGGAGCGGAATTAGA
Octa-47	ATAATTAAATTTAAAAAACTTTTTCAAACTTTTAACAACGCC
Octa-48	GCACCCAGCGTTTTTTATCCGGTATTCTAGGCGAATTATTCA
Octa-49	GGAAGCGCCCACAAACAGTTAATGCCCCGACTCCTCAAGATA
Octa-50	GTTTGCCTATTCACAGGCAGGTCAGACGCCACCACACCACCC
Octa-51	CGCGAGCTTAGTTTTTCCCAATTCTGCGCAAGTGTAAAGCCT
Octa-52	AGAAGCAACCAAGCCAAAAGAATACACTAATGCCAAAACTCC
Octa-53	ATTAAGTATAAAGCGGCAAGGCAAAGAAACTAATAGGGTACC
Octa-54	CAGTGCCTACATGGGAATTTACCGTTCCACAAGTAAGCAGAT
Octa-55	ATAAGGCGCCAAAAGTTGAGATTTAGGATAACGGACCAGTCA
Octa-56	TGCTAAACAGATGAAGAAACCACCAGAATTTAAAAAAAGGCT
Octa-57	CAGCCTTGGTTTTGTATTAAGAGGCTGACTGCCTATATCAGA
Octa-58	CGGAATAATTCAACCCAGCGCCAAAGACTTATTTTAACGCAA
Octa-59	CGCCTGAATTACCCTAATCTTGACAAGACAGACCATGAAAGA
Octa-60	ACGCGAGGCTACAACAGTACCTTTTACAAATCGCGCAGAGAA
Octa-61	CAGCGAACATTAAAAGAGAGTACCTTTACTGAATATAATGAA
Octa-62	GGACGTTTAATTTCGACGAGAAACACCACCACTAATGCAGAT
Octa-63	AAAGCGCCAAAGTTTATCTTACCGAAGCCCAATAATGAGTAA
Octa-64	GAGCTCGTTGTAAACGCCAGGGTTTTCCAAAGCAATAAAGCC
Octa-65	AATTATTGTTTTCATGCCTTTAGCGTCAGATAGCACGGAAAC
Octa-66	AAGTTTCAGACAGCCGGGATCGTCACCCTTCTGTAGCTCAAC
Octa-67	ACAAAGAAATTTAGGTAGGGCTTAATTGTATACAACGGAATC
Octa-68	AACAAAAATAACTAGGTCTGAGAGACTACGCTGAGTTTCCCT
Octa-69	CATAACCTAAATCAACAGTTCAGAAAACGTCATAAGGATAGC
Octa-70	CACGACGAATTCGTGTGGCATCAATTCTTTAGCAAAATTACG
Octa-71	CCTACCAACAGTAATTTTATCCTGAATCAAACAGCCATATGA
Octa-72	GATTATAAAGAAACGCCAGTTACAAAATTTACCAACGTCAGA
Octa-73	AGTAGATTGAAAAGAATCATGGTCATAGCCGGAAGCATAAGT
Octa-74	TAGAATCCATAAATCATTTAACAATTTCTCCCGGCTTAGGTT
Octa-75	AAAGGCCAAATATGTTAGAGCTTAATTGATTGCTCCATGAGG
Octa-76	CCAAAAGGAAAGGACAACAGTTTCAGCGAATCATCATATTCC

Octa-77	GAAATCGATAACCGGATACCGATAGTTGTATCAGCTCCAACG
Octa-78	TGAATATTATCAAAATAATGGAAGGGTTAATATTTATCCCAA
Octa-79	GAGGAAGCAGGATTCGGGTAAAATACGTAAAACACCCCCCAG
Octa-80	GGTTGATTTTCCAGCAGACAGCCCTCATTCGTCACGGGATAG
Octa-81	CAAGCCCCCACCCTTAGCCCGGAATAGGACGATCTAAAGTTT
Octa-82	TGTAGATATTACGCGGCGATCGGTGCGGGCGCCATCTTCTGG
Octa-83	CATCCTATTCAGCTAAAAGGTAAAGTAAAAAGCAAGCCGTTT
Octa-84	CAGCTCATATAAGCGTACCCCGGTTGATGTGTCGGATTCTCC
Octa-85	CATGTCACAAACGGCATTAAATGTGAGCAATTCGCGTTAAAT
Octa-86	AGCGTCACGTATAAGAATTGAGTTAAGCCCTTTTTAAGAAAG
Octa-87	TATAAAGCATCGTAACCAAGTACCGCACCGGCTGTAATATCC
Octa-88	ATAGCCCGCGAAAATAATTGTATCGGTTCGCCGACAATGAGT
Octa-89	AGACAGTTCATATAGGAGAAGCCTTTATAACATTGCCTGAGA
Octa-90	AACAGGTCCCGAAATTGCATCAAAAAGATCTTTGATCATCAG
Octa-91	ACTGCCCTTGCCCCGTTGCAGCAAGCGGCAACAGCTTTTTCT
Octa-92	TCAAAGGGAGATAGCCCTTATAAATCAAGACAACAACCATCG
Octa-93	GTAATACGCAAACATGAGAGATCTACAACTAGCTGAGGCCGG
Octa-94	GAGATAACATTAGAAGAATAACATAAAAAGGAAGGATTAGGA
Octa-95	CAGATATTACCTGAATACCAAGTTACAATCGGGAGCTATTTT
Octa-96	CATATAACTAATGAACACAACATACGAGCTGTTTCTTTGGGG
Octa-97	ATGTTTTGCTTTTGATCGGAACGAGGGTACTTTTTCTTTTGATAAGAGGTCATT
Octa-98	GGGGTGCCAGTTGAGACCATTAGATACAATTTTCACTGTGTGAAATTGTTATCC
Octa-99	CTTCGCTGGGCGCAGACGACAGTATCGGGGCACCGTCGCCATTCAGGCTGCGCA
Octa-100	TCAGAGCTGGGTAAACGACGGCCAGTGCGATCCCCGTAGTAGCATTAACATCCA
Octa-101	TTAGCGGTACAGAGCGGGAGAATTAACTGCGCTAATTTCGGAACCTATTATTCT
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Octa-103	TGTCGTCATAAGTACAGAACCGCCACCCATTTTCACAGTACAAACTACAACGCC
Octa-104	CGATTATAAGCGGAGACTTCAAATATCGCGGAAGCCTACGAAGGCACCAACCTA
Octa-105	AACATGTACGCGAGTGGTTTGAAATACCTAAACACATTCTTACCAGTATAAAGC
Octa-106	GTCTGGATTTTGCGTTTTAAATGCAATGGTGAGAAATAAATTAATGCCGGAGAG
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Octa-109	TGATTATCAACTTTACAACTAAAGGAATCCAAAAAGTTTGAGTAACATTATCAT
Octa-110	ACATAACTTGCCCTAACTTTAATCATTGCATTATAACAACATTATTACAGGTAG
Octa-111	GTAGCGCCATTAAATTGGGAATTAGAGCGCAAGGCGCACCGTAATCAGTAGCGA
Octa-112	TTATTTTTACCGACAATGCAGAACGCGCGAAAAATCTTTCCTTATCATTCCAAG
Octa-113	TTTCAATAGAAGGCAGCGAACCTCCCGATTAGTTGAAACAATAACGGATTCGCC
Octa-114	GGGCGACCCCAAAAGTATGTTAGCAAACTAAAAGAGTCACAATCAATAGAAAAT
Octa-115	AGCCGAAAGTCTCTCTTTTGATGATACAAGTGCCTTAAGAGCAAGAAACAATGA
Octa-116	GTGGGAAATCATATAAATATTTAAATTGAATTTTTGTCTGGCCTTCCTGTAGCC

Octa-117	CCCACGCGCAAAATGGTTGAGTGTTGTTCGTGGACTTGCTTTCGAGGTGAATTT
Octa-118	ATGACCACTCGTTTGGCTTTTGCAAAAGTTAGACTATATTCATTGAATCCCCCT
Octa-119	TCCAAATCTTCTGAATTATTTGCACGTAGGTTTAACGCTAACGAGCGTCTTTCC
Octa-120	GGGTTATTTAATTACAATATATGTGAGTAATTAATAAGAGTCAATAGTGAATTT

2. Supporting figures

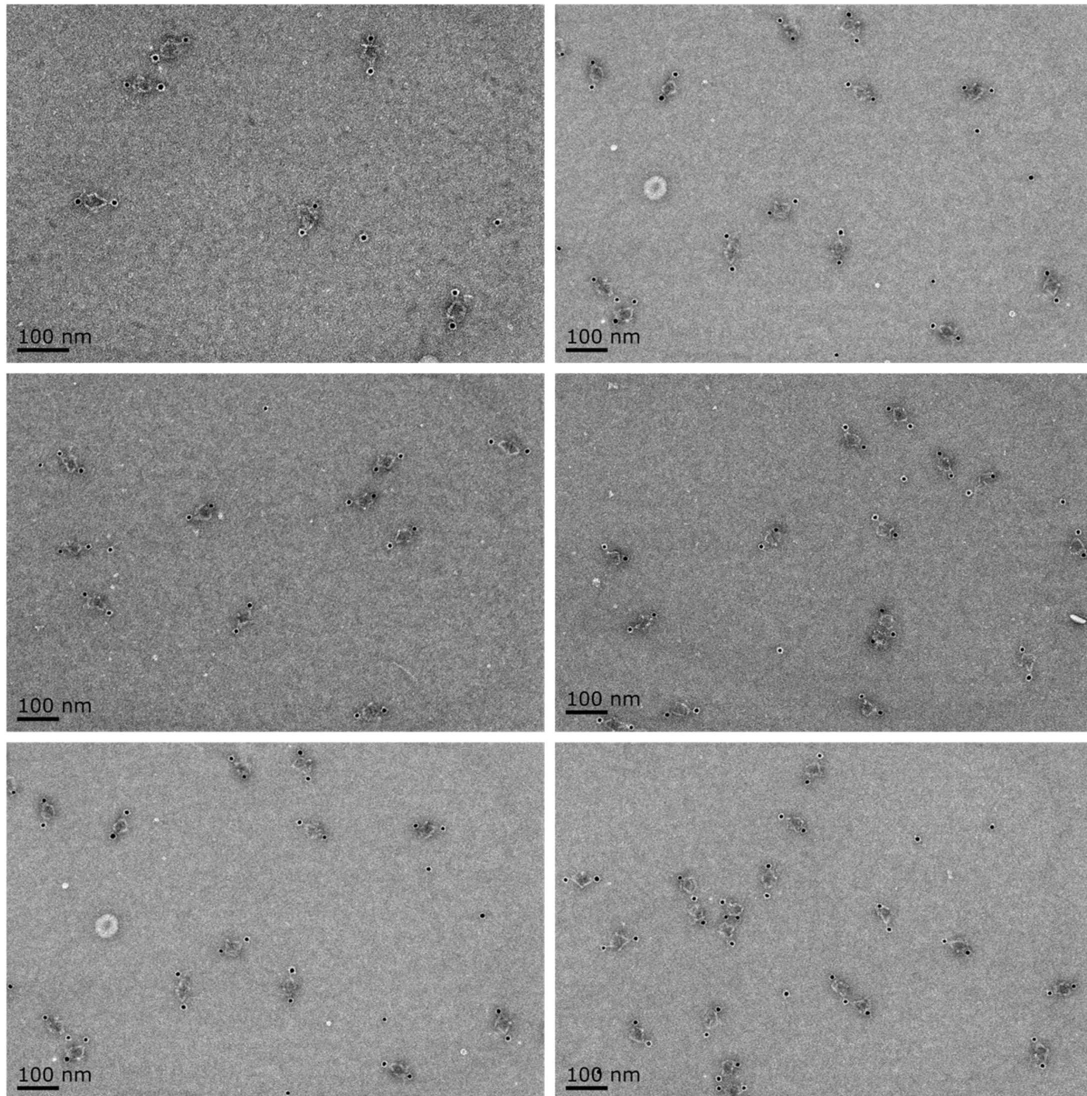

Figure S1. Representative negative stained TEM images for dimers fabricated by the 1st strategy.

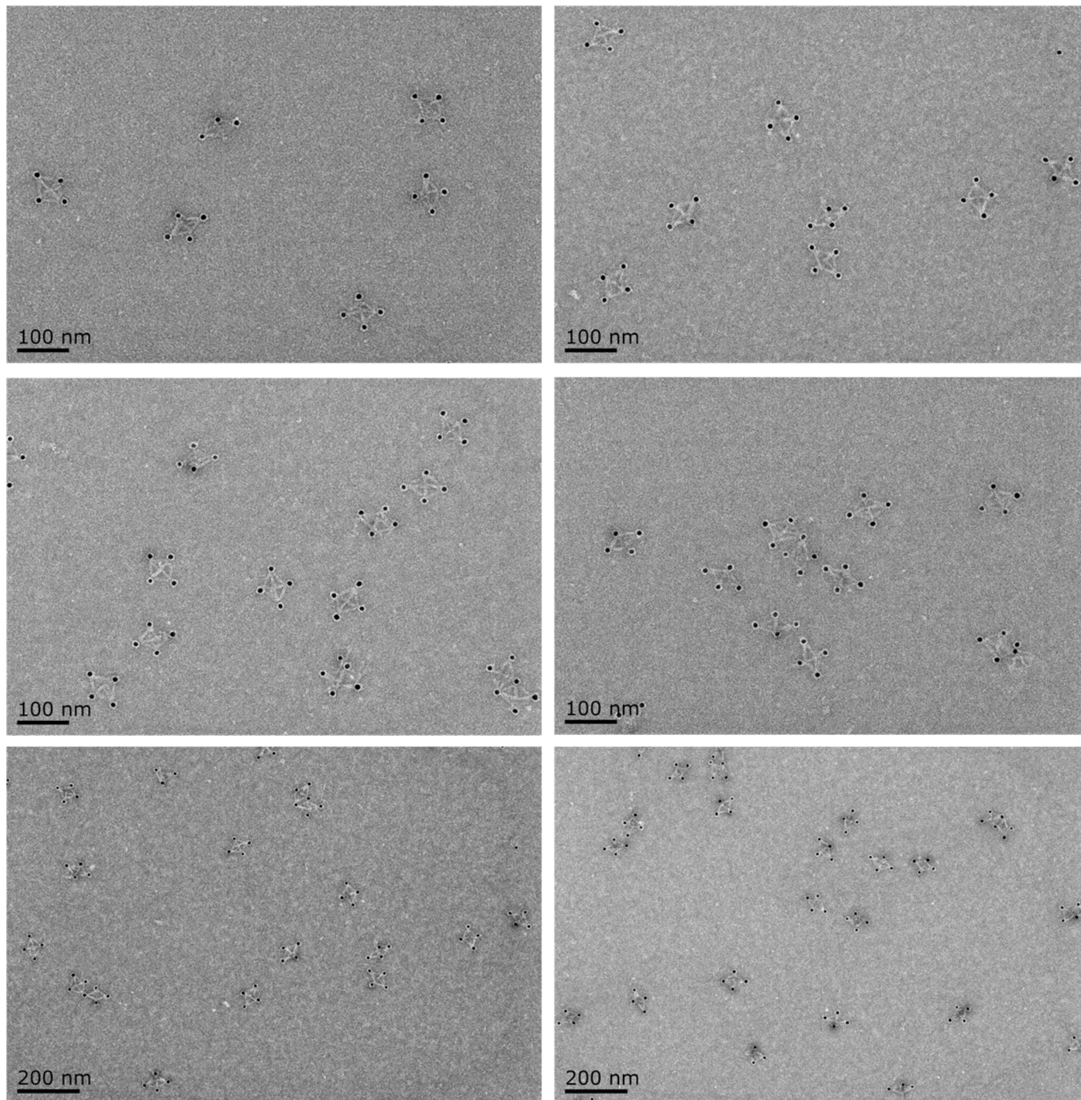

Figure S2. Representative negative stained TEM images for tetramers fabricated by the 2nd strategy.

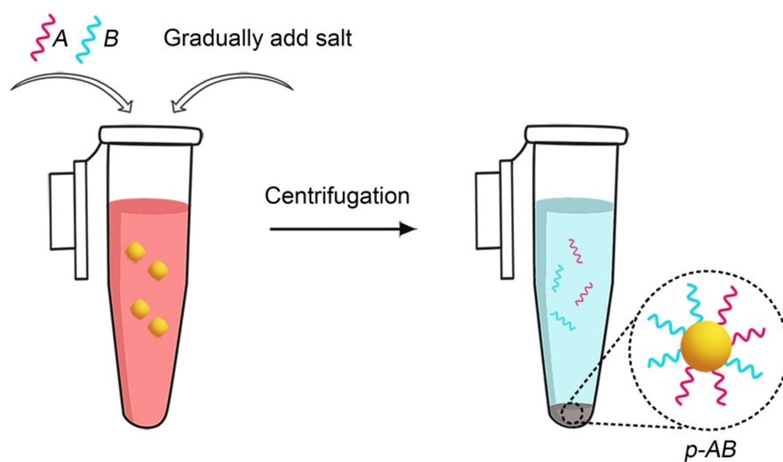

Figure S3. Schematic diagram of DNA functionalized on Au nanoparticles. Salt (NaCl) were gradually added to the tube, after mixing gold nanoparticles (yellow spheres) with DNA (*A* and *B*) and aging for two hours. Another 12 hours aging time were needed before centrifuging and rinsing the DNA functionalized nanoparticles (*p-AB*).

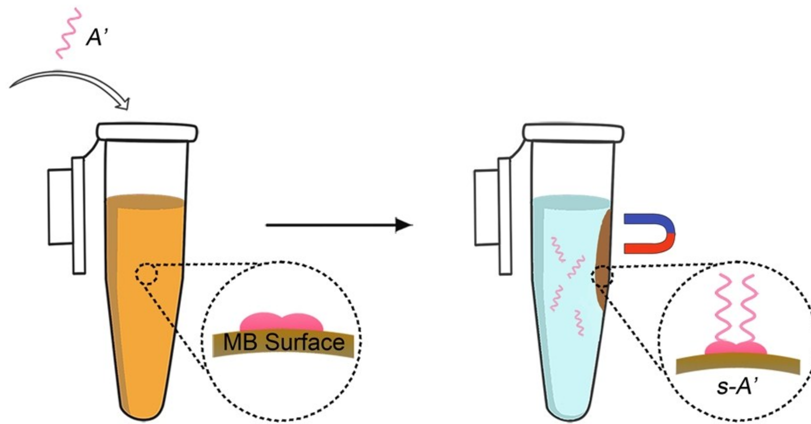

Figure S4. Schematic diagram of DNA functionalized on magnetic beads (MBs). MBs with surface encoded with streptavidin were purchased without further modification. DNA sequences with end contained biotins (*A'*) were mixed with these MBs to attach DNA on the surface (*s-A'*). By applying magnetic field, excess DNA-biotin sequences could be washed out.

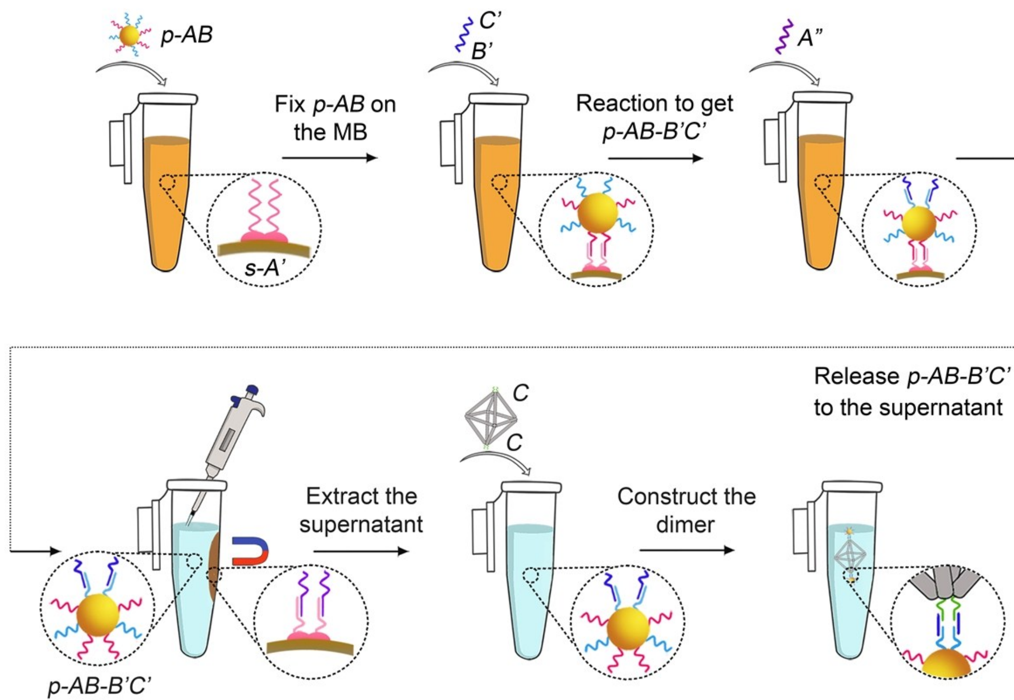

Figure S5. Schematic illustration for dimers assembly realized by the 1st strategy. First, we put the AuNPs *p-AB* into the tube containing MBs surface *s-A'*. After rotating for 12 h at room temperature, *p-AB* were fixed on the surface *s-A'* tightly. The supernatant of the tube was removed and the sample was rinsed four times with buffer. Second, we added ssDNA *B'C'* to the tube at a molar ratio of 3:1 for ssDNA *B'C'* : *p-AB*. The tube was shaken at 1,000 rpm for 12 h at 35 °C and then rinsed four times with method similar as mentioned above. After that, we added the substituted chain *A''* at a molar ratio of *A''* and *A'* for 5:1 to release patchy particles *p-AB-B'C'* from the *s-A'* to the supernatant. Finally, we put the octahedral DOFs into the supernatant at a 1:3 molar ratio with the patchy particles *p-AB-B'C'*. After been shaken at 1,000 rpm for 12 h at 38 °C, the tube was filled with freshly produced dimer.

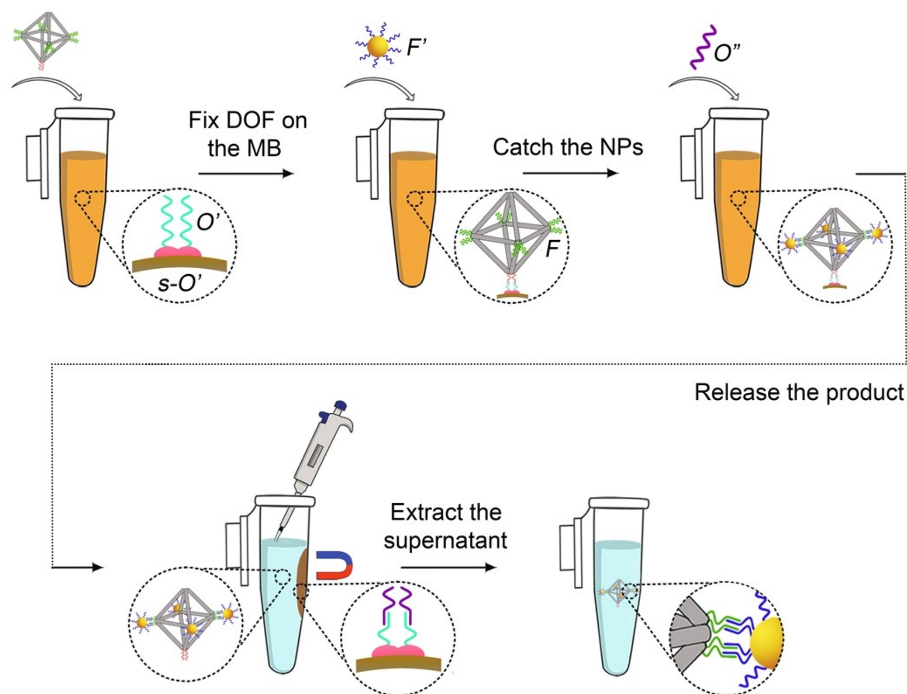

Figure S6. Schematic illustration for dimers assembly realized by the 2nd strategy. First, the DOFs were added into surface *s-O'* solution and reacted for 12 h. Second, AuNPs *p-F'* was added into the solution at a 10:1 molar ratio for AuNPs and DOFs. After the tube was shaken at 1,000 rpm for 12 h at 35 °C, the solution was rinsed for several times. Finally, 'fuel DNA' *O''* was put into the solution to release the clusters to the supernatant.

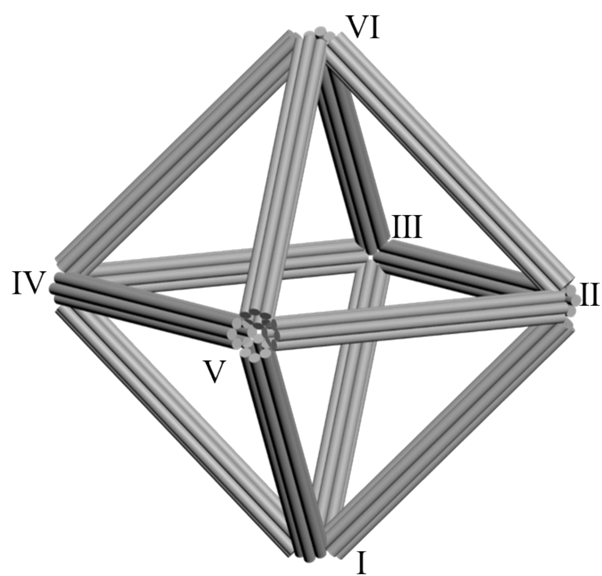

Figure S7. Schematic diagram of the octahedral DOF. The DOF we designed has totally six vertices, as labeled from Octa Ⅰ to Octa Ⅵ.