Supporting Information for

Interconnecting Gold Islands with DNA Origami Nanotubes

Baoquan Ding, Hao Wu, Wei X‡, Zhao Zhao, Yan Liu, Hongbin Yu, Hao Yan

Experimental Section:

Materials: All oligonucleotides were purchased from Integrated DNA Technologies (www.idtdna.com). The origami staple strands were ordered in 96-well plates that were normalized to 100μM, and were used without further purification. The thiolated strands were purified by denaturing PAGE. The concentration of each strand was estimated by measuring the UV absorbance at 260 nm. M13 viral DNA waspurchased from New England Biolabs, Inc. (catalog number N4040S). The droplet contact angle was measured with the contact angle meter (FM40 Easy Drop, Krüss GmbH, Germany). Typically, the wafer sample was fixed on the stage holder first. Water droplets were deposited manually with syringes. The image of the droplet was captured with a camera and analyzed with Easy Drop software.

Self-assembly of DNA origami nanotube: The design of the DNA origami tubes is the same as previously described by L. A. Stearns *et al*[1]. The strand structure is shown in Figure S6. The sequences of unmodified helper strands are listed in Table S1 and the sequences of the thiolated strands are listed in Table S2. A molar ratio of 1:10 between viral, ssDNA M13 and short unmodified staple strands was used. A molar ratio of 1:1 ratio for the thiolated strands to the viral DNA was used. Origami tubes were assembled in 1x TAE-Mg^{2+} buffer (40 mM Tris, 20 mM acetic acid, 2 mM EDTA, 12.5 mM $Mg(OAc)_2$, pH 8.0) by cooling slowly from 90°C to room temperature. The origami tubes and excess staples were then separated by EtBr stained, 1.5% agarose gel (running buffer 1x TAE-Mg^{2+} buffer, 10 V/cm), running in an ice-water bath. The band corresponding to the origami tubes was excised and extracted from the gel using Freeze N' Squeeze columns (Bio-Rad, USA). After purification, the tiles were then concentrated using 100 kDa Microcon centrifugal filter devices (Millipore, USA). The final concentration of origami tiles was estimated according to the dsDNA absorbance at 260 nm and the calculated extinction coefficient (http://biophysics.idtdna.com/).

Substrate patterning for the alignment of DNA origami: We used Piranha and BOE for wafer pre-cleaning. Piranha ingredients are 70% concentrated sulfuric acid and 30% hydrogen peroxide. BOE is Buffered Oxide Etch which is a 20:1 mixture of hydrofluoric acid and ammonium fluoride. When cleaning wafers, first we soaked the wafers in Piranha for 10 minutes and then soaked them in BOE for 5 minutes, followed by distilled water washing and N_2 drying. Samples were stored in a Nitrogen box. . The spin coater we used was a LS-8000 LAB SPINNER from Metron Systems Inc. For HMDS, the spin parameter was 5000 RPM for 30 seconds; for PMMA (2% in anisole) it was 4000 RPM for 30 seconds. Specifically, before pattern definition, a monolayer of HMDS was spin-coated with 5000RPM for 30 seconds followed by an 80 nm thick layer of PMMA resist (2% PMMA in Anisole, 4000RPM for 30 seconds). The desired patterns were formed by different arrangements of nanodots. The sizes of these dots ranging from 30 nm to 90nm were created by varying the dosage of the Electron-beam exposure (JEOL 6000 FS). After the removal of exposed PMMA, 2 nm of Cr and 10 nm of Au layers were thermally evaporated followed by the lift-off process.

Interconnecting gold islands with DNA origami tubes: Thiolated DNA strands in the purified origami tube were reduced with 20 mM TCEP for 1 hour before use. Extra TCEP and the reduced small molecules were removed using a Microcon centrifuge device (100 kDa). Typically, 400 µl of 1×TAE Mg^{2+} buffer was added to 100 µl DNA tube solutions and centrifuged at 3000 RPM for 10 minutes. The process was repeated three times. The gold island patterned substrate was cut into 5 mm × 20 mm small pieces. Each piece was then immersed into a tube filled with freshly reduced DNA origami solution and incubated for 20 hours or longer. The volume of origami solution was usually 100 µL with a concentration ranging from 1 to 10 nM. The substrate was then thoroughly rinsed with water, dried under a stream of nitrogen gas, and imaged in air in tapping modeatomic force microscope (TM-AFM, Veeco V).

AFM imaging: Typically, AFM imaging has been done with tapping-in-buffer mode for most of the DNA nanostructures in the DNA nanotechnology community. In most cases, the DNA nanostructures were deposited on a mica surface and imaged in buffer. For our system, the sample was on a Si surface and we not only needed to image the DNA tubes, but also the gold surface island. The diameter of the DNA tubes is about 7 nm; the gold islands' diameter ranges from 40 nm to 80 nm with a height of 20 to 30 nm. We found that high resonant frequency (~300 kHz) tips gave us the best images for both DNA tubes and gold islands. So tapping-in-air was used for our imaging process. The AFM tip is ultrasharp 15 series (NSC15) purchased from MikroMasch (www.spmtips.com) with resonant frequency ~ 325 kHz and force constant ~ 46 N/m.

General discussion:

We did observe that some tubes are met at branch points in the space between spots. These could be the result of the reduced thiolated groups bonding with each other to form the disulfide bonds in the solution. Another possibility is that the tubes aggregate through stacking interaction at the ends. In both cases, we believe that the concentration of DNA origami tubes plays an important role in effecting the degree of aggregation. Higher concentrations of the tubes will increase the chances that they interact with each other in solution. In Fig.5 of the main text, we used a lower concentration (1 nM) of DNA tubes and the aggregating effect was decreased.

The length of the linker strand is important here. If the length is short, considering the bending and twisting of the tube structure, the thiolated groups may not be long enough to reach the gold island. If the linker strands are too long, then the thiolated groups can get to the gold islands that were not designed for the tube to reach. For example, the tube could reach the gold islands in the diagonal direction in the gold islands square lattice.

We used a 1 to 1 ratio for the thiolated strands to the scaffold viral DNA. Since we purify the tube structures with agarose gels, a ratio that is higher than 1 to 1 should also work. We chose the 1 to 1 ratio with the purpose of not wasting the purified thiolated DNA strands. We did try a higher ratio of 3 to 1. After purification with an agarose gel, we did not observe any difference as compared to the 1 to 1 ratio. We did not try higher ratios without purification. We think the extra thiolated strands would attach to the gold islands and may affect the binding of tubes.

We also observed some multiply bridged spots. We think the multiple bridging tubes are mainly due to independent attachments. Before the surface deposition, we used gels to purify the single tubes. The side-to-side pre-aggregating in solution could only happen after the reduction of thiolated groups and will form disulfide bonds thus lower the chance for the thiolated groups to bind to the gold islands. One reason that there is multiple bridging is that after one bridging happened, the local area became hydrophilic and the accessibility for the next tube was then increased.

Table S1. Sequences of unmodified helper strands used to assemble the origami nanotube. To prepare nanotubes bearing thiolated single strand extensions, helper strands were replaced by 3' thiol group modified DNA strands (Table S2) and annealed with the remaininghelper strands.

Name	Sequence (5'-3')
1	TGTGTAGGTAAAGAATAGTAGTAGCATTCCTCAGAACCGCCA
2	CATCAATTCTACTATTCAAAAGGGTGAGATAGATTAGAGCCG
3	AACCAGAGCCACCAACCCTCAGAGCCGCAGCTGAAAAGGTGG
4	CCCTCAGAGCCACCCCGGAACCGCCTCCAAGTATTAGACTTT
5	TCAATAGATAATACATTTGAGGATTTAGCTCAGAGCCGCCAC
6	ACAAACAATTCGACAGCACTAACAACTAAAAGGCCGGAGACA
7	CCTTATTAGCGTTTAGCATTGACAGGAGATGGTCAATAACCT
8	AAAATATCTTTAGGAACTCGTATTAAATATCAAAATCACCGG
9	TTATTAATTTTAAAAGTTGAAAGGAATTCCGTTCTAGCTGAT
10	GTCAAATCACCATCTCATTTGGGGCGCGCACCAGAACCACCA
11	CCAGAGCCGCCGCCGCCATCTTTTCATACCTTTGCCCGAACG
12	GTTTAGCTATATTTAATATGATATTCAAGAGGAAGGTTATCT
13	CACCGTAATCAGTAAGCGCAGTCTCTGATCTGGAAGTTTCAT
14	GTTGGCAAATCAACAGTTTGAGTAACATTTCGGTCATAGCCC
15	TGATTATCAGATGAAGCAAATGAAAAATTGGAGCAAACAAGA
16	AAATTAATGCCGGAGATACATTTCGCAAGTTGAGGCAGGTCA
17	AAGCCAGAATGGAAGCGACAGAATCAAGTATCATCATATTCC
18	GACGATTGGCCTTGTTTTCATCGGCATTTATCATTTTGCGGA
19	ACAAAGAAACCACCTATCAAACCCTCAATTGAGAGATCTACA
20	TGCTGAACCTCAAAAGAAGGAGCGGAATTTTGCCTTTAGCGT
21	AAGGCTATCAGGTCGATTCCCAATTCTGATAAATCCTCATTA
22	CAGACTGTAGCGCGATATTCACAAACAACGAACGAGTAGATT
23	TAGTTTGACCATTAGAGGGTAGCTATTTTCAATATCTGGTCA
24	TCCATATAACAGTTATTGCCTGAGAGTCCTAAAGCATCACCT
25	GGGAATTAGAGCCAGGGGTCAGTGCCTTCATTTTTGCGGATG
26	CAACCGATTGAGGGAGTATTAAGAGGCTAGCTTCAAAGCGAA
27	CGCTGAGAGCCAGCTGGCAATTCATCAAACCATCGATAGCAG
28	GTACCTTTTACATCACGTGGCACAGACATCAGCTCATTTTTT
29	GAATCGATGAACGGACTAAAGTACGGTGATTTACCGTTCCAG
30	TTCTGAAACATGAAAGGGAAGGTAAATAAATATACAGTAACA
31	TAAGCGTCATACATACGTCACCAATGAATATAATCCTGATTG
32	TTTGGATTATACTTAGTATTAACACCGCGCATGTCAATCATA
33	GAGGTGAGGCGGTCCTGAATAATGGAAGAGCACCATTACCAT
34	TAATAAGTTTTAACGCAAAATCACCAGTGGTTAGAACCTACC
35	ATATCAAAATTATTGAACGAACCACCAGCCAAAAACAGGAAG
36	CCATTAAAAATACCTGCACGTAAAACAGGACTTGAGCCATTT
37	GTATAAACAGTTAATTATCACCGTCACCAAATAAAGAAATTG
38	CGTAGATTTTCAGGTGCGCGAACTGATAACGTTAATATTTTG
39	CTATTAGTCTTTAATTTAACGTCAGATGTTGACGGAAATTAT
40	TTAAAATTCGCATTAACTCCAACAGGTCTTCGGAACCTATTA

41	TCATTAAAGGTGAATGCCCCCTGCCTATAGGATTAGAGAGTA
42	ATTGTATAAGCAAATTTTGATAAGAGGTGAGTAACAGTGCCC
43	TGTACCCCGGTTGATGCTGAATATAATGCAGGAGTGTACTGG
44	GCTTAGAGCTTAATTAATCAGAAAAGCCCAGAAGATAAAACA
45	GTTTTAAATATGCATAATCGTAAAACTACTGCAACAGTGCCA
46	TAGCAAGGCCGGAAGGCTTTTGATGATACTGTAGCTCAACAT
47	CCTTTAATTGCTCCTATTTAAATTGTAAGCCCTAAAACATCG
48	CCAGACCGGAAGCAAAATTTTTGTTAAAATATTTTTGAATGG
49	GAAAGCGTAAGAATGGGAGAAACAATAAAAAAGGGCGACATT
50	TAACCCACAAGAATCCAGACGTTAGTAACCAAAAGGAATTAC
51	AACCAATAGGAACGCGCGTTTTAATTCGGAGACTCCTCAAGA
52	GTTTTGTCGTCTTTTGAGTTAAGCCCAATTATCAAAATCATA
53	GCAACTGTTGGGAAGCAACACTATCATACGTAACGATCTAAA
54	AAGACTTCAAATATCCATCAAAAATAATAACCCTTCTGACCT
55	GGTCTGAGAGACTAATCCTGAGAAGTGTGGCCTCTTCGCTAT
56	GAGGCATAGTAAGAGGGCGATCGGTGCGTTTTATAATCAGTG
57	AGGCCACCGAGTAAAGTCAATAGTGAATTAATAAGAGCAAGA
58	AAGACGCTGAGAAGAAGAGTCTGTCCATGCCATTCAGGCTGC
59	GACGACGATAAAAAGCAAAGCGCCATTCCACGCAAATTAACC
60	GTTGTAGCAATACTCCTTGAAAACATAGCGAAGCCCTTTTTA
61	TTTTCCCTTAGAATTCTTTGATTAGTAACGGCACCGCTTCTG
62	AAGTTTTGCCAGAGTCCAGCCAGCTTTCTAACATCACTTGCC
63	TGAGTAGAAGAACTGCTTCTGTAAATCGTTACCAGAAGGAAA
64	GAGTGAATAACCTTCAAACTATCGGCCTGACAGTATCGGCCT
65	TAGCGTCCAATACTTTTGAGGGGACGACTGCTGGTAATATCC
66	AGAACAATATTACCTGGAAACAGTACATCCAAAAGAACTGGC
67	ATTACCTTTTTTAAGCCAGCCATTGCAAGGGCGCATCGTAAC
68	CCCCCTCAAATGCTACGTTGGTGTAGATCAGGAAAAACGCTC
69	ATGGAAATACCTACATTAATTACATTTAGTTAGCAAACGTAG
70	ATCAAGAAAACAAAATTTTGACGCTCAACGGCGGATTGACCG
71	ATAAATCAAAAATCTCCGTGGGAACAAATCGTCTGAAATGGA
72	TTATTTACATTGGCCTGAGCAAAAGAAGTAAAAGAAACGCAA
73	TTCATTTCAATTACAGATTCACCAGTCATGTGAGCGAGTAAC
74	AAGCAAAGCGGATTTCATCAACATTAAACACGACCAGTAATA
75	AAAGGGACATTCTGAGTTACAAAATCGCACAATCAATAGAAA
76	TGCTTTGAATACCAGCCAACAGAGATAGTCGCGTCTGGCCTT
77	CCTGTAGCCAGCTTGCATCAAAAAGATTCAGTACCAGGCGGA
78	AACCCGTCGGATTCAGGTCTTTACCCTGTATAGCCCGGAATA
79	TAATGGGATAGGTCTTAAACAGTTCAGATACCGCCACCCTCA
80	CGTGCATCTGCCAGGCGGAATCGTCATACAGAGCCACCACCC
81	CAGGAAGATCGCACGGGGTAATAGTAAAGAACCCATGTACCG
82	GTGCCGGAAACCAGCCAAAATAGCGAGACTACAACGCCTGTA
83	AACAATGAAATAGCGCCCTCATAGTTAGACCCTCGTTTACCA
84	GCATTCCACAGACAAATAGCTATCTTACCGATAGCTTAGATT
85	AGAAAAGTAAGCAGGTCACCAGTACAAAGGCTTTTGCAAAAG

86	TAACACTGAGTTTCATAGCCGAACAAAGTCGCTATTAATTAA
87	CCGAGGAAACGCAAAGCAAGCCCAATAGATGTTTAGACTGGA
88	TCATTTTCAGGGATTAATAACGGAATACAAATCAATATATGT
89	ATGATTAAGACTCCAGAACCGCCACCCTAATATTCATTGAAT
90	GAACCGCCACCCTCTTATTACGCAGTATACAATTTCATTTGA
91	AAAATACATACATACTCAGGAGGTTTAGAAACGAGAATGACC
92	GGTGTATCACCGTAAAGGTGGCAACATAATGATGAAACAAAC
93	AGACACCACGGAATAGGGTTGATATAAGACTATTATAGTCAG
94	TAAGTGCCGTCGAGAAGTTTATTTTGTCGCAGAGGCGAATTA
95	ATTCATATGGTTTAAGCGGGGTTTTGCTAAGAGGAAGCCCGA
96	GAAGGATTAGGATTCCAGCGCCAAAGACCGGATTCGCCTGAT
97	AACGGTACGCCAGACCTTTTTAACCTCCTAATATCAGAGAGA
98	GAACGCGAGGCGTTGCATCGGAACGAGGAGGCTGGCTGACCT
99	TACGCCAGCTGGCGGCAGATACATAACGATGAATTTTCTGTA
100	AGCAGCGAAAGACATTAGCGAACCTCCCCCAACATGTAATTT
101	CTGCCCGCTTTCCACTTGACAAGAACCGGGATCGTCACCCTC
102	CACATTCAACTAATAAAGGGGGATGTGCGGATTTTAGACAGG
103	AGGCAGAGGCATTTCCATCACCCAAATCCGTGCCAGCTGCAT
104	TCATCAAGAGTAATGTCGGGAAACCTGTAAGTTTTTTGGGGT
105	CGAGGTGCCGTAAACATATTTAACAACGGACTTGCGGGAGGT
106	TAATTGAGAATCGCGCACTAAATCGGAATTGCGTTGCGCTCA
107	AAATCAACGTAACACTAACTCACATTAACCCTAAAGGGAGCC
108	CCCGATTTAGAGCTTATAAAGCCAACGCTATTTTGCACCCAG
109	CAAATTCTTACCAGTGACGGGGAAAGCCGTAAAGCCTGGGGT
110	CCCTGACGAGAAACCGGAAGCATAAAGTGGCGAACGTGGCGA
111	GAAAGGAAGGGAAGAAGCCTGTTTAGTAGCTAACGAGCGTCT
112	ATAATTACTAGAAAAAAGCGAAAGGAGCCGCTCACAATTCCA
113	GATGGTTTAATTTCGTGAAATTGTTATCGGGCGCTAGGGCGC
114	TGGCAAGTGTAGCGGCGTTAAATAAGAAATAAACAGCCATAT
115	GTGTGATAAATAAGGTCACGCTGCGCGTCGTAATCATGGTCA
116	CGATTTTAAGAACTACCGAGCTCGAATTAACCACCACACCCG
117	CCGCGCTTAATGCGCTAAATTTAATGGTTTTTTTGTTTAACG
118	TTTCATCTTCTGACCCGCTACAGGGCGCGCAGGTCGACTCTA
119	AAGAAAAATCTACGAAGCTTGCATGCCTGTACTATGGTTGCT
120	TTGACGAGCACGTAAAAACTTTTTCAAAAGAGAATAACATAA
121	ACAAAGAACGCGAGTAACGTGCTTTCCTACGACGTTGTAAAA
122	TATTACAGGTAGAAGGGTTTTCCCAGTCCGTTAGAATCAGAG
123	CGGGAGCTAAACAGTAAATGCTGATGCATTAACTGAACACCC
124	TATATAACTATATGGAGGCCGATTAAAGTGCAAGGCGATTAA
125	GTTGGGTAACGCCAAGATTCATCAGTTGTTTCAGCGGAGTGA
126	CGACGGCCAGTGCCTTAATAAAACGAACCGAATAATAATTTT
127	GAGGATCCCCGGGTGGCTCATTATACCACTCCAAAAGGAGCC
128	TAGCTGTTTCCTGTAACTTTAATCATTGTTCGAGGTGAATTT
129	CACAACATACGAGCACCAGAACGAGTAGCCGACAATGACAAC
130	GCCTAATGAGTGAGAAGCTGCTCATTCATCGGTCGCTGAGGC

131	TTTGAAGCCTTAAAAGGCCGCTTTTGCGGATATTCATTACCC
132	TTGCAGGGAGTTAATCAAGATTAGTTGCTCAACAGTAGGGCT
133	CTACAATTTTATCCCATAACCGATATATGTGAATAAGGCTTG
134	AACCATCGCCCACGTGAATCTTACCAACTCATATGCGTTATA
135	TTCCAGAGCCTAATTACCGATAGTTGCGTAAATTGGGCTTGA
136	CTTAAACAGCTTGATTGCCAGTTACAAATAAACACCGGAATC
137	TATTTATCCCAATCTTTATCAGCTTGCTTGAATTACCTTATG
138	TTTAATTGTATCGGCAAATAAGAAACGATTGAAATACCGACC
139	TCAAAAATGAAAATCTCCAAAAAAAAGGGTCAGGACGTTGGG
140	TTCACGTTGAAAATAGCAGCCTTTACAGTATATTTTAGTTAA
141	AAACAGGGAAGCGCAACTAAAGGAATTGTAACGGAACAACAT
142	GAATAGAAAGGAACATTAGACGGGAGAAAATCCAATCGCAAG
143	TGAACAAAGTCAGAACAACTTTCAACAGAGATTTAGGAATAC
144	TGGGATTTTGCTAAGGGTAATTGAGCGCGGCTTAGGTTGGGT
145	GCCCACTACGTGAATCGAGCCAGTAATAATCCGGTATTCTAA
146	TAATGAATCGGCCACAGACCAGGCGCATGTAGCAACGGCTAC
147	AGATGAACGGTGTAACGCGCGGGGAGAGCTATCAGGGCGATG
148	TTTACGAGCATGTATTTGATGGTGGTTCAGGCGAAAATCCTG
149	CGGAGATTTGTATCTGGTTTGCCCCAGCCGAAATCGGCAAAA
150	TCCCTTATAAATCAGAACAAGAAAAATACTTATCATTCCAAG
151	AATAGATAAGTCCTAAAGAATAGCCCGATGAGAGAGTTGCAG
152	CGACCTGCTCCATGTCACCGCCTGGCCCGATAGGGTTGAGTG
153	TTGTTCCAGTTTGGCTAATGCAGAACGCATCGAGAACAAGCA
154	AACAACATGTTCAGAACAAGAGTCCACTAGTGAGACGGGCAA
155	TCAATCATAAGGGATTTTTCTTTTCACCATTAAAGAACGTGG
156	ACTCCAACGTCAAAAAGTAATTCTGTCCATTACCGCGCCCAA
157	ACCGACAAAAGGTAGGGCGAAAAACCGTGCGGTTTGCGTATT
158	GGGCGCCAGGGTGGACCGAACTGACCAAATGAGGAAGTTTCC
159	CAGCTGATTGCCCTTTACTTAGCCGGAATACGAAGGCACCAA
160	CAAGCGGTCCACGCATCGCCTGATAAATCTAAAACACTCATC
161	GAAACCAATCAATAATTATACCAAGCGCGAAACAAAGTACAA
162	TTTGACCCCCAGCGATCGGCTGTCTTTCATATCCCATCCTAA
163	AACGGGTATTAAACGGCAAAAGAATACATGTGTCGAAATCCG
164	CCTAAAACGAAAGACAAGTACCGCACTCGCCTGTTTATCAAC
165	AGCCGTTTTTATTTATACGTAATGCCACCGAGGCGCAGACGG
166	ATTAAACGGGTAAATCATCGTAGGAATCAGACGACGACAATA
167	TAGCAAGCAAATCACTAAAGACTTTTTCCTTTGAAAGAGGAC
168	AGAGGCTTTGAGGAGATATAGAAGGCTTAGAGAATATAAAGT

Table S2. Sequences of helper strands functionalized with thiolated groups. Strand names refer to unmodified helper strands listed in Table S1. The 33 nucleotideregion extended from the tubes is highlighted in red.

Name	Sequence (5'-3')
s09	TTATTAATTTTAAAAGTTGAAAGGAATTCCGTTCTAGCTGATTAATAATAATAATAATAATAATAATAATAATAA-S
s10	GTCAAATCACCATCTCATTTGGGGCGCGCACCAGAACCACCATAATAATAATAATAATAATAATAATAATAATAA-S
s12	GTTTAGCTATATTTAATATGATATTCAAGAGGAAGGTTATCTTAATAATAATAATAATAATAATAATAATAATAA-S
s13	CACCGTAATCAGTAAGCGCAGTCTCTGATCTGGAAGTTTCATTAATAATAATAATAATAATAATAATAATAATAA-S
s155	TCAATCATAAGGGATTTTTCTTTTCACCATTAAAGAACGTGGTAATAATAATAATAATAATAATAATAATAATAA-S
s156	ACTCCAACGTCAAAAAGTAATTCTGTCCATTACCGCGCCCAATAATAATAATAATAATAATAATAATAATAATAA-S
s160	CAAGCGGTCCACGCATCGCCTGATAAATCTAAAACACTCATCTAATAATAATAATAATAATAATAATAATAATAA-S
s166	ATTAAACGGGTAAATCATCGTAGGAATCAGACGACGACAATATAATAATAATAATAATAATAATAATAATAATAA-S

References:

1.) Stearns, L. A.; Chhabra, R.; Sharma, J.; Liu, Y.; Petuskey, W. T.; Yan, H. and Chaput, J. C. *Angew. Chem. Int. Ed.***2009**, 45, 8494-8496.

Figure S1. Additional AFM images used for thestatistical analysisreported in Figure 3. All sample substrates were fabricated using the same pattern, and prepared by incubation in 2 nM DNA origami solution for 20 hours. a) 80 nm gold island, 4 thiolated strands at each end; b) 80 nm gold island, 2 thiolated strands at each end; c) 80 nm gold island, 1 thiolated strand at each end; d) 60 nm gold island, 4 thiolated strands at each end; e) 60 nm gold island, 2 thiolated strands at each end; f) 40 nm gold island, 2 thiolated strands at each end, some 30- 40 nm gold islands may have dissociated from the substrate during the lift-off and sonication cleaning process.

Figure S1.

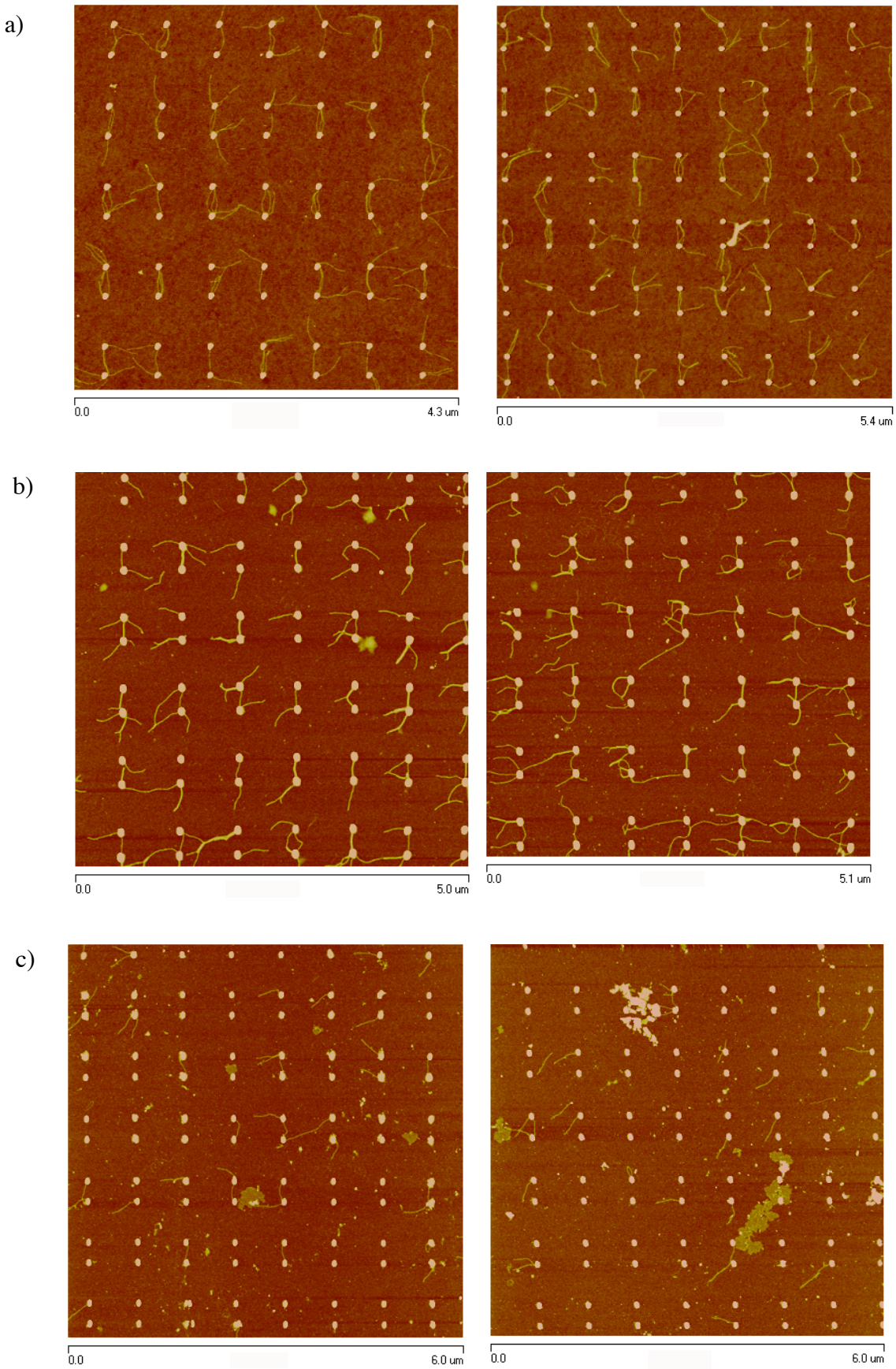
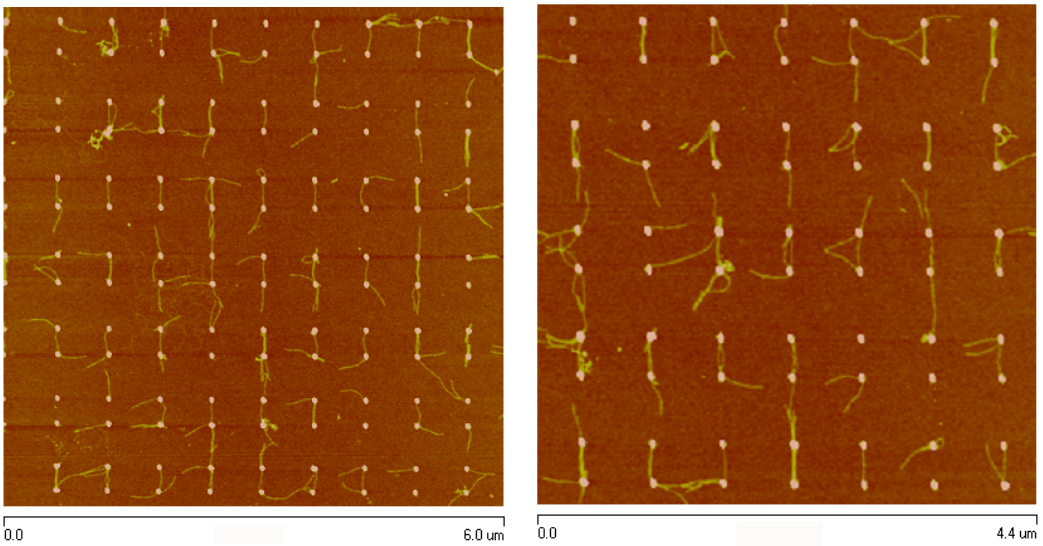
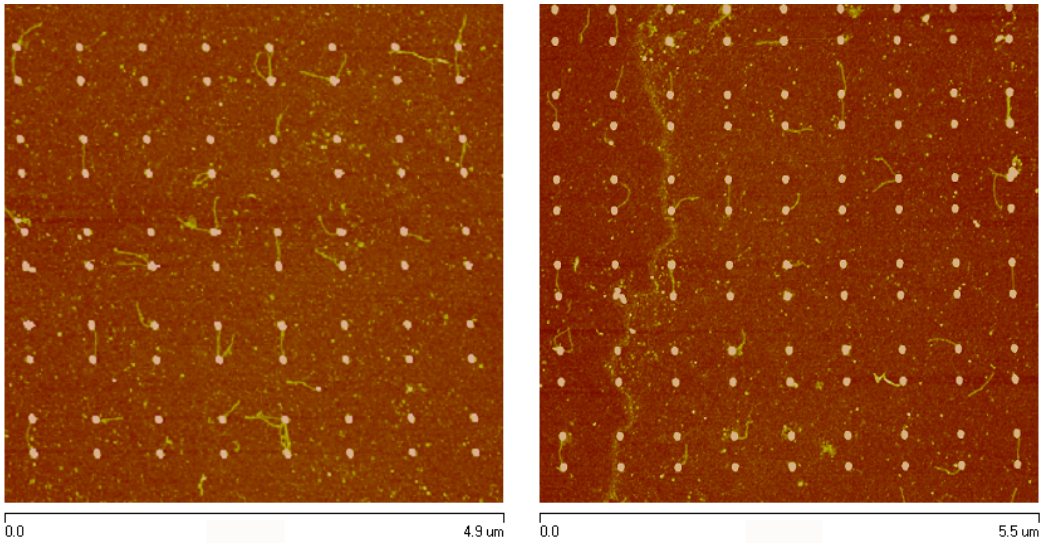


Figure S1, continued.

d)



e)



f)

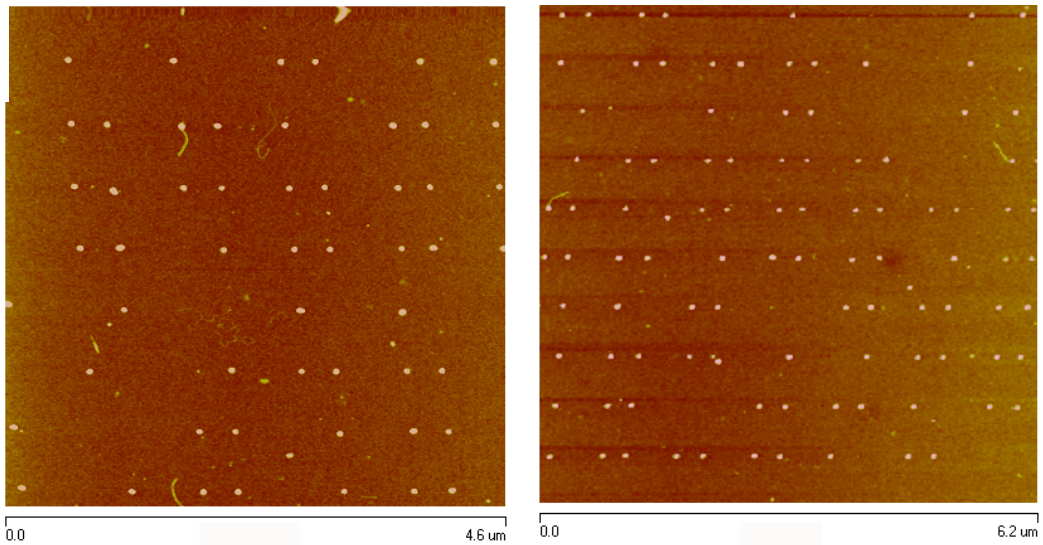


Figure S2. AFM images for 60 nm gold islands incubated with 2 nM DNA origami tubes that each contained 4 thiolated DNA strands at both ends for 40 hours. Longer incubation time enhances the binding efficiency and improves the number of connections.

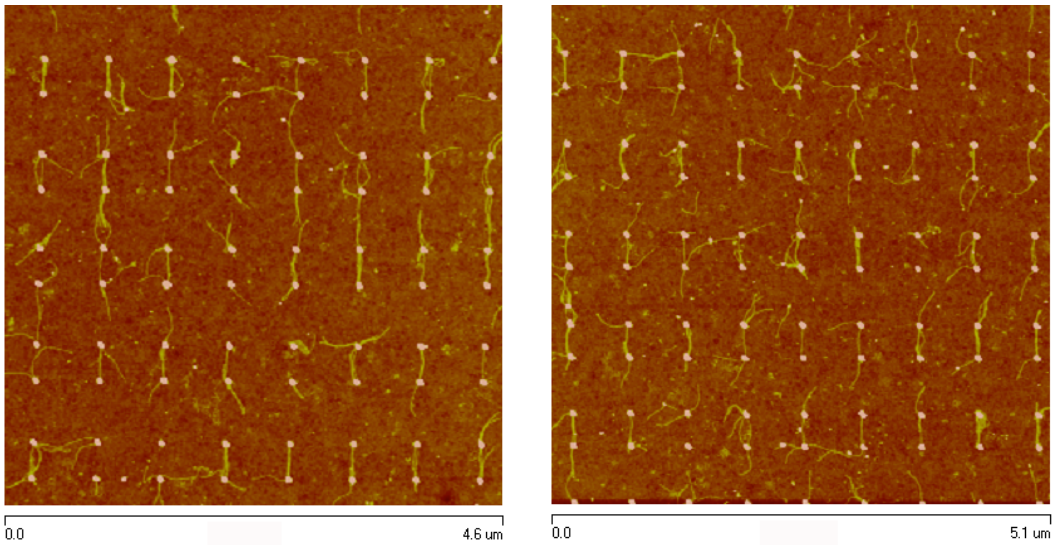


Figure S3. Additional AFM image used for the analysis shown in Figure 5. The gold islands are 60 nm; DNA origami tubes each contain 4 thiolated strands at both ends; the incubation time is 20 hours in 2 nM purified origami solution.

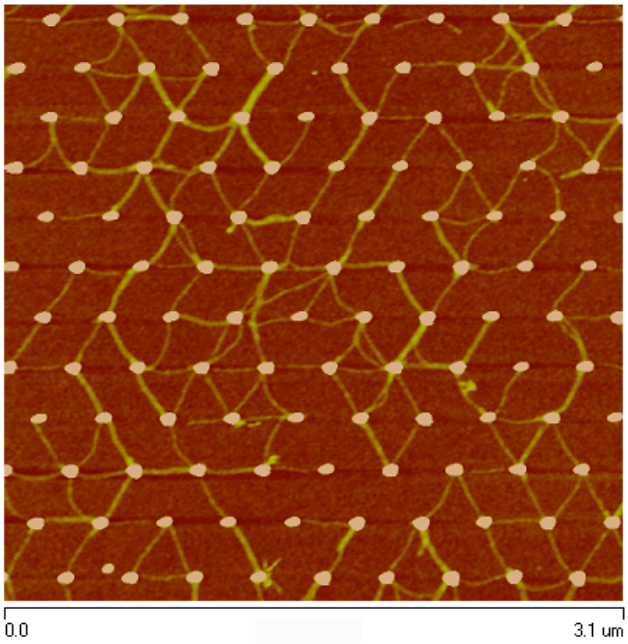


Figure S4. AFM images of two different areas (phase images) show that the DNA nanotube concentration affects the binding efficiency. The gold islands are 60 nm; DNA origami tubes each contain 4 thiolated strands at both ends; the incubation time was 20 hours in 10 nM purified origami solution. Higher concentration of origami enhances the binding efficiency of origami tubes to gold islands.

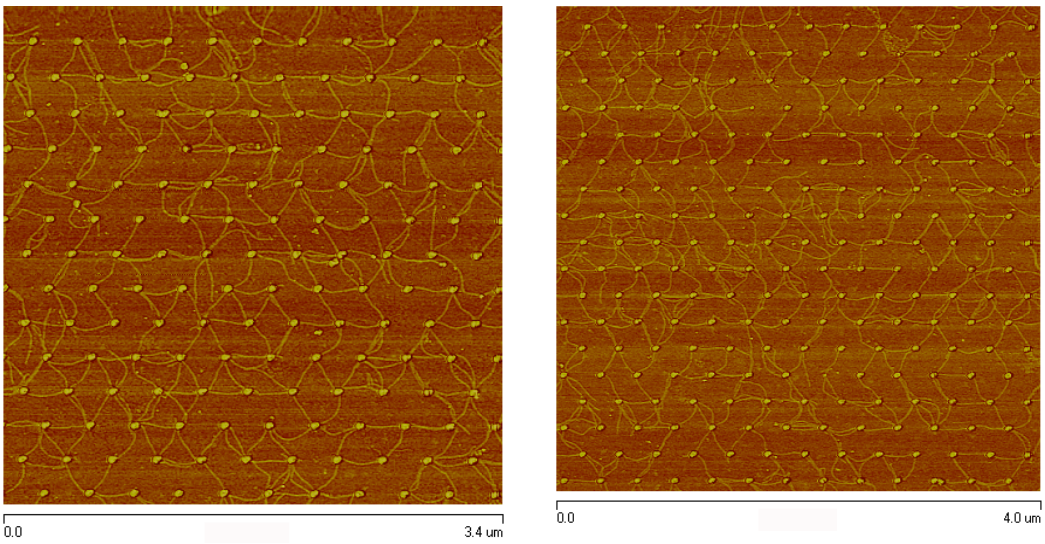


Figure S5. Additional AFM images for complex nanostructures formed by interconnecting groups of gold islands with DNA origami tubes. a) Triangles; b)"Z" shapes; c) hexagons; d) squares.

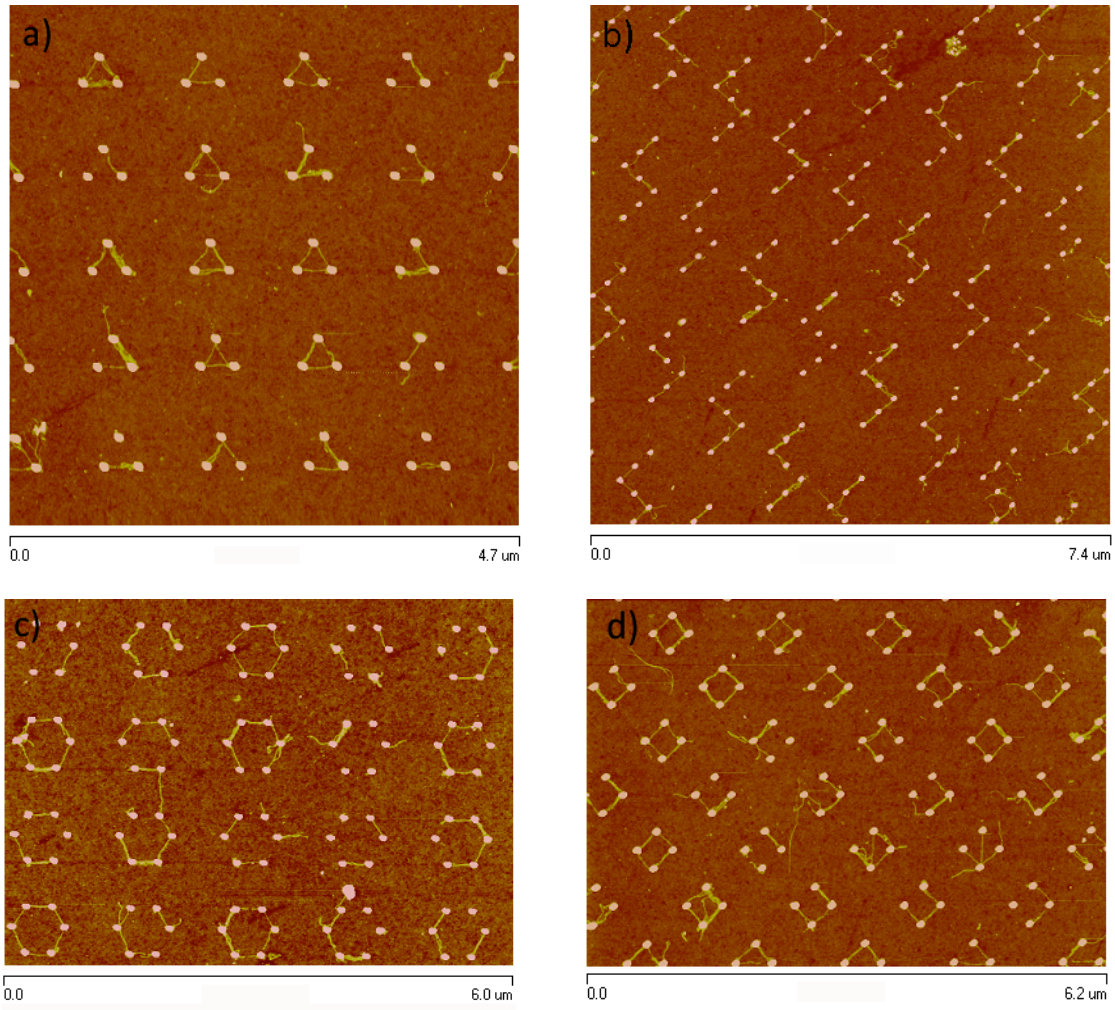
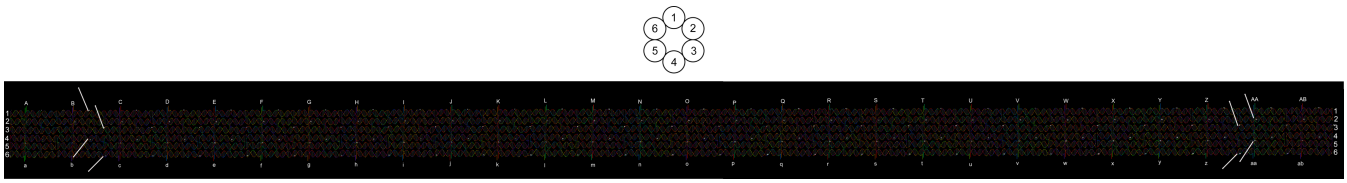
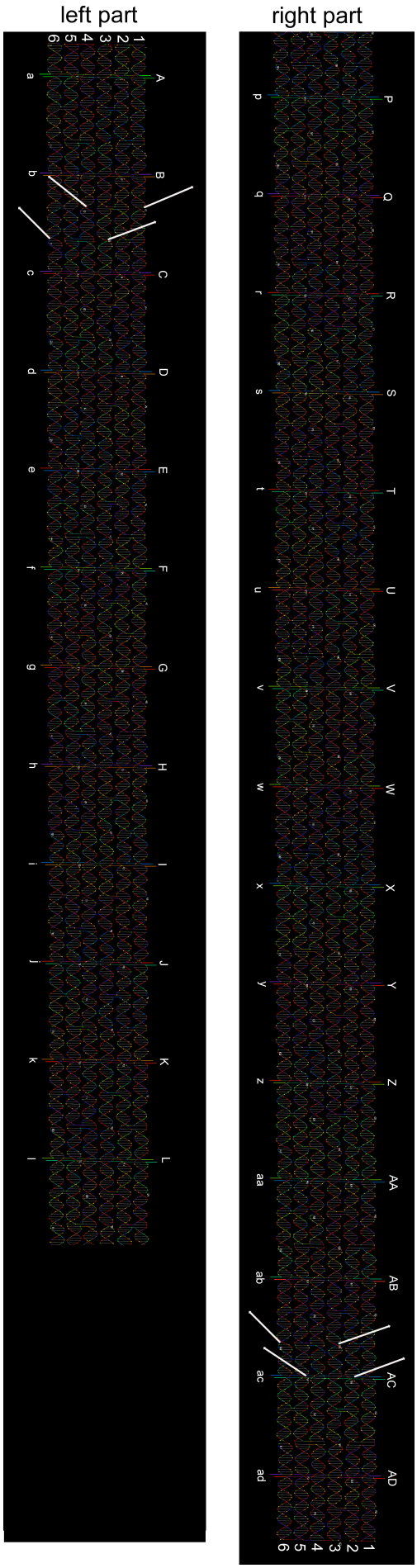


Figure S6. Structure layout of the DNA origami nanotube. The structure was designed using our in house software Tiamat that can be downloaded from http://yanlab.asu.edu/Resources.html. The structure is a six-helix bundle when viewed from the helix axis. To allow the readers to view the 3D structure in a 2D plane, the structure was rearranged in 2D as shown below. Helix 1 and 6 are connected at crossover points at position A and A', B and B' and so on. The picture shown on the next page provides better resolution image for zoom-in view by separating the structure into two panels. The white lines indicate the position of the thiol modification. The numbering of the strands in the structure corresponds to the numbering of the sequences shown in Table S1.




left part
right part