Electronic Supplementary Information

1.1 Materials

All oligonucleotides were used as purchased from Sangon Biotech Co., Ltd. (Shanghai, China) without further purification. T4 DNA Ligase and Exonuclease I were purchased from Takara Biotechnology Co., Ltd. (Dalian, China). Ethylene diamine tetraacetate, urea, magnesium acetate tetrahydrate, boric acid, acetic acid, tris(hydroxymethyl)-aminomethane (Tris), acrylamide bis-acrylamide were purchased from Sigma Aldrich Corporation. AFM cantilevers were from Bruker Corporation. Mica sheets for AFM were from Nanjing Zhongjingkeyi Technology Co., Ltd. Water (18 MΩ• cm) was from a Milli-Q Ultrapure Water Purification System.

1.2 Preparation of circularised DNA

The synthetic strategy is shown in Figure S1. A mixture of 3.5 µM circularisable 5'-phosphorylated oligonucleotide (35µL) and 4.5 µM splint oligonucleotide (45µL) was annealed from 95 °C to room temperature over 3 h. Then 10 µL T4 DNA ligase (350U/µL) containing 66 mM Tris-HCl (pH 7.6), 6.6 mM $MgCl_2$, 10 mM DTT, 0.1 mM ATP and 3.3 µM [32P]-$Na_4P_2O_7$, was added and the mixture was incubated at 16 °C for 16 hours. The sample was then heated at 65 °C for 10 min to inactivate T4 DNA ligase and then cooled via ice bath for 5 min. Then 10 µL of exonuclease-I (5U/µL) was added and the mixture was incubated at 37 °C for 30 min to digest both linear splint and un-ligated oligonucleotides. Exonuclease-I was inactivated by heating at 80 °C for 5 min and then suddenly cooled via ice bath for 5 min. Prior to loading the sample to the denaturing polyacrylamide gel, 50 µL deionised formamide was added.

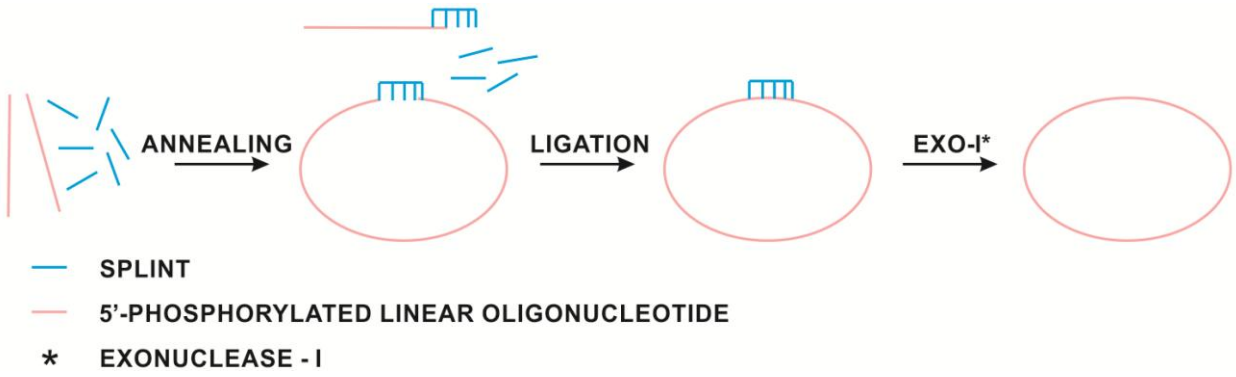


Fig. S1 Schematic representation of preparation of a circularised DNA template.

1.3 Purification of circularised DNA via denaturing PAGE

The circularised DNA strands were undergone 12% denaturing polyacrylamide gel (19:1 acrylamide/bisacrylamide, 4.2 M urea) electrophoresis in 1 × Tris-borate-EDTA (TBE) buffer (89 mM tris-HCl, 89 mM boric acid, 2 mM EDTA, pH 8.0) at room temperature. After the gel electrophoresis was carried out at 75V (constant voltage) for 2 h, the bands were cut from the denaturing gel, macerated finely and eluted in a TE buffer containing 500 mM ammonium acetate, 10 mM tris-HCl and 1 mM EDTA (pH 7.6). Finally the purified DNA was dissolved in sterilised water.

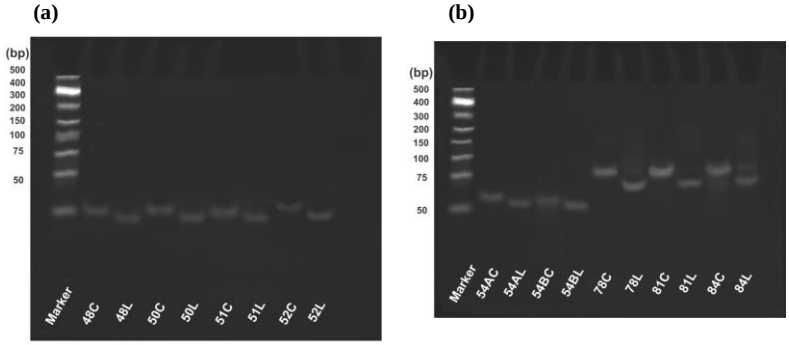


Fig. S2 Denaturing PAGE gel (12%) analysis for all the circularised strands, where numbers show the "base pairs" in the corresponding lanes, label C after numbers represents "circular" and label L after numbers represents "linear".

1.4 Synthesis of self-assembled nanostructures

Each designed set of DNA strands was mixed in stoichiometric amounts of 0.5 μM with 1×Tris-Acetate-EDTA-Mg^{2+} (TAE-Mg^{2+}) buffer (40 mM Tris-Acetate, 2 mM EDTA, 25 mM $Mg(Ac)_2$, pH 8.5) to a final volume of 20μL. The oligonucleotide mixture was annealed in a thermocycler programmed to cool from 95°C to 4°C following the set of conditions: 95°C to 65°C by 1°C every 150 seconds, 64°C to 50°C by 0.1°C every 10 min (for 48- to 54-nt rings; and every 7 min for 78- to 84-nt rings), 49°C to 5°C by 1°C every 150 seconds, then hold at 4°C.

1.5 Native polyacrylamide gel electrophoresis (PAGE)

Gels contained 10% polyacrylamide (19:1 acrylamide/bisacrylamide) and TAE-Mg^{2+} buffer. The loading TAE/Mg^{2+} buffer (1 μL) containing 50% glycerol, 0.2% tracking dye of Bromophenol Blue and Xylene Cyanol FF, and 0.15 μM DNA complex. Gels were run in the presence of 1× Tris-Acetate-EDTA (TAE) (40 mM Tris-Acetate, 2 mM EDTA, pH 8.5) buffer on an electrophoresis unit (Biorad) in cold water bath using a constant voltage of 65V for 2 h. After electrophoresis, the gels were stained with GelRed for 30 min and then imaged with digital camera. One example of the native gel for constructing triangle-Δ16 tile step by step is shown in Figure S3.

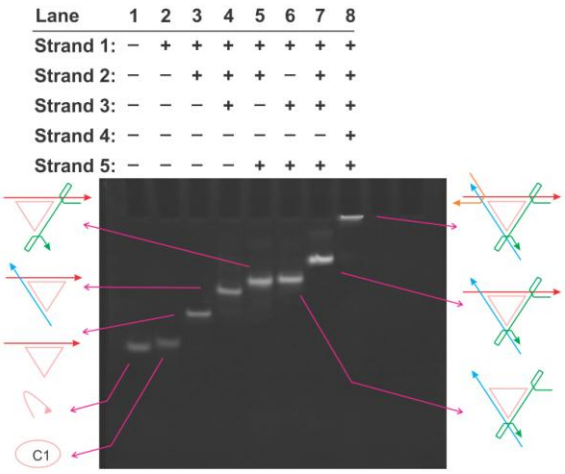


Fig. S3 Native PAGE gel (10%) analysis for constructing triangle-Δ16 tile step by step.

1.6 Qualitative statistics of production yield of nanotubes

We define the nanotube's production yield as imaging statistics of nanotube assemblies, i.e. the frequency of observation of nanotubes found in all experiments of this project. Generally we conducted AFM testing on 6 × 6 μm^2 area 4~6 times for good samples, and did a qualitative statistical analysis of how frequently nanotubes were observed. NT-9 was the most frequently imaged, NT-4, NT-7, and NT-8 were the second, NT-1 was the third, NT-2, NT-3, NT-5 and NT-6 were the fourth.

1.7 Atomic force microscopy (AFM)

To prepare the samples for AFM imaging, 5 μL of the annealed mixture was deposited onto freshly cleaved mica and the solution drop was left to stay for 2 min in order to allow nanostructures to be adsorbed onto the surface. Mica was washed by 100 μL of sterilised water two times and excess water was removed with filter paper. To avoid time-dependent sample degradation, imaging was conducted within 24 h. AFM images were acquired in air and at room temperature. The samples were scanned with the FastScan-C probe in the ScanAsyst mode in air, and with the ScanAsyst-Fluid+ probe in PeakForce Mapping in Fluid-HiS mode on a FastScan AFM (Bruker Inc.).

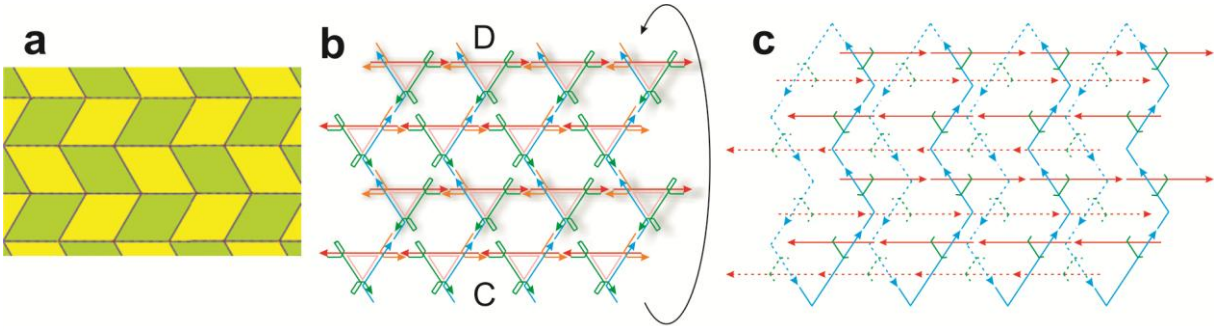
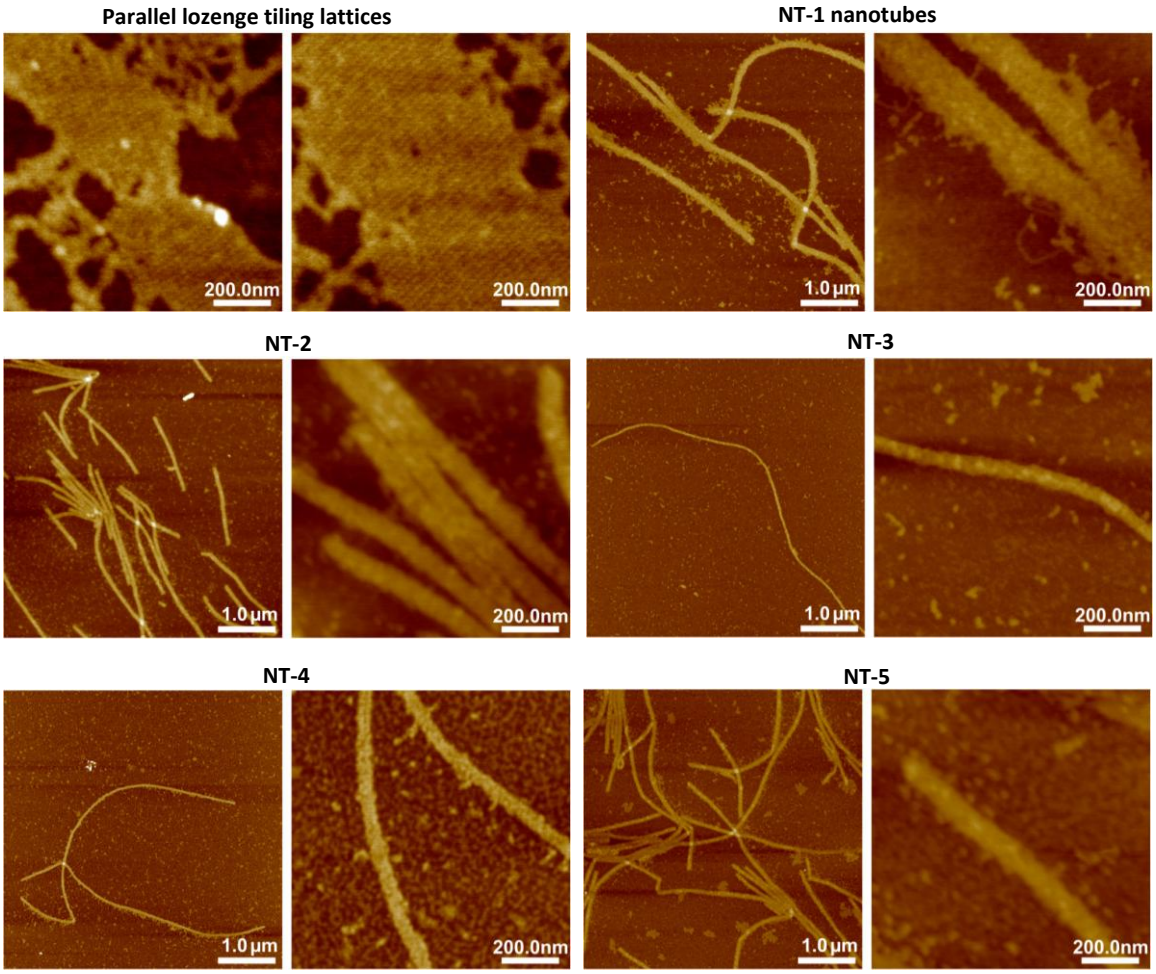


Fig. S4 Schematic antiparallel lozenge tiling structures suggested: (a) mathematical model showing antiparallel lozenge tiling, (b) antiparallel lozenge tiling lattice, (c) forming an antiparallel lozenge tiling nanotube from (b). The curved black arrow head in (b) points to the folding direction forming the nanotube (c).

1.8 Additional AFM images



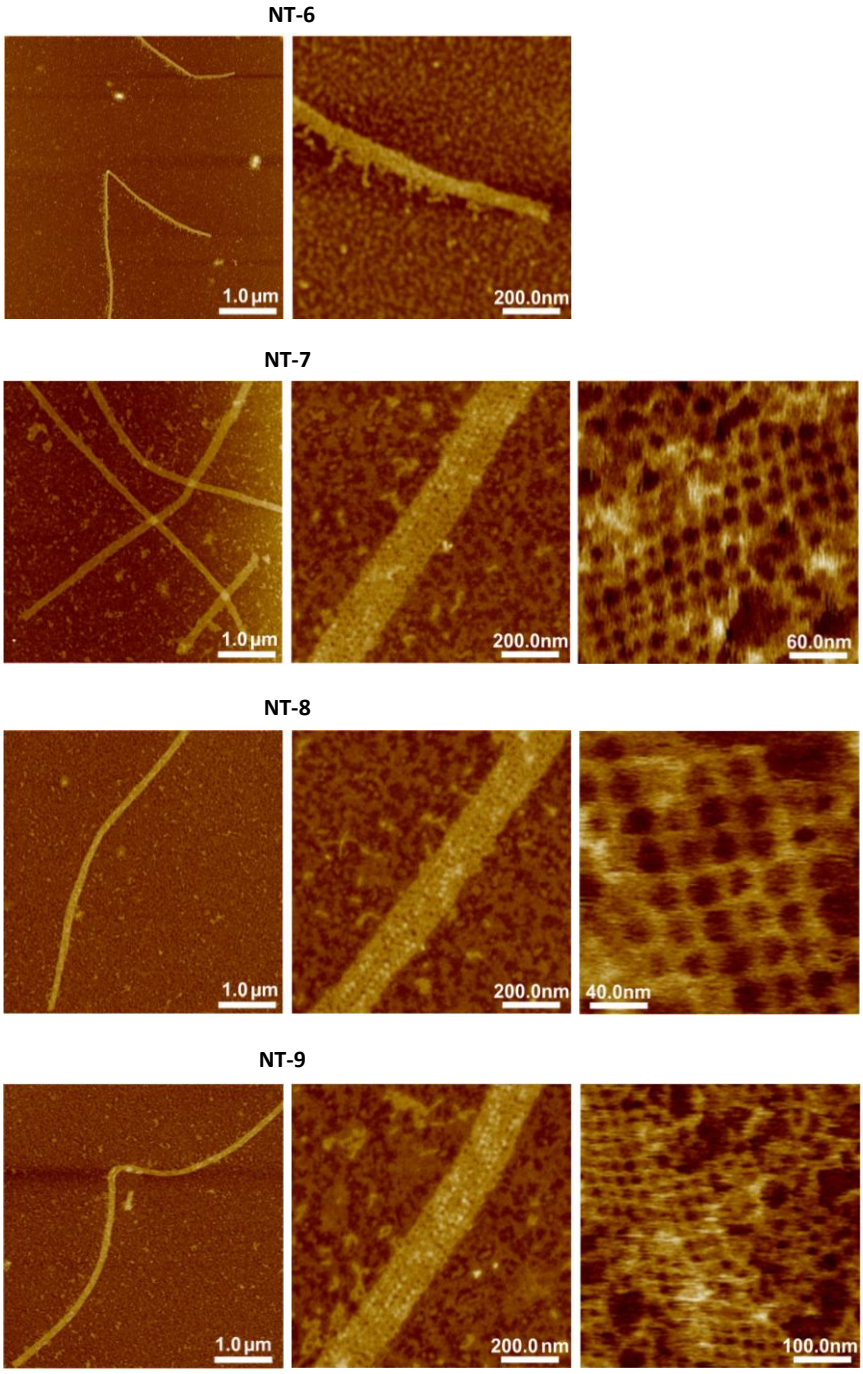


Fig. S5 More AFM images of the designed nanotubes. All images were captured in air except the three right panels of NT-8 to NT-9 respectively in fluid.

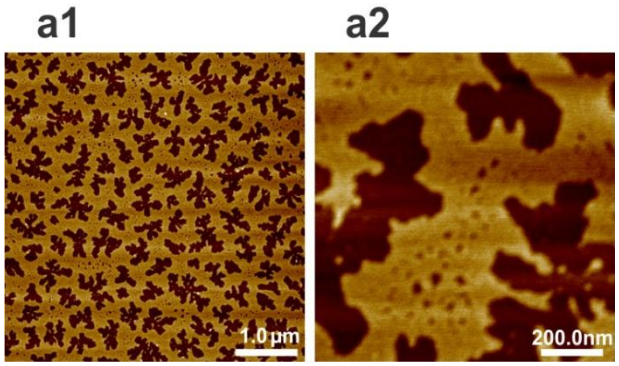
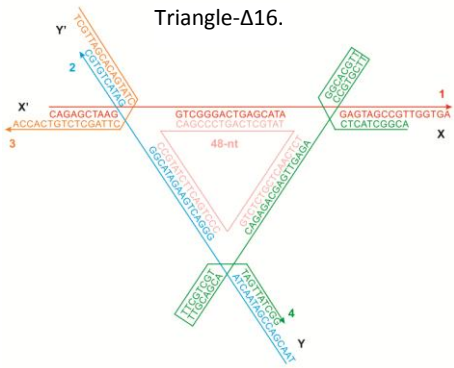
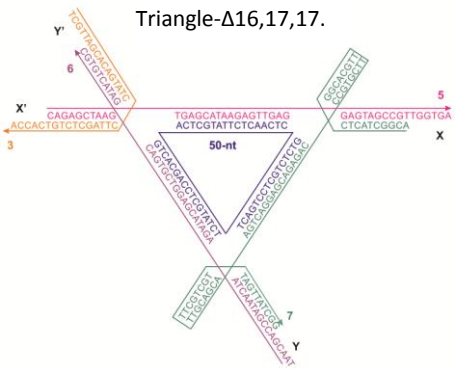
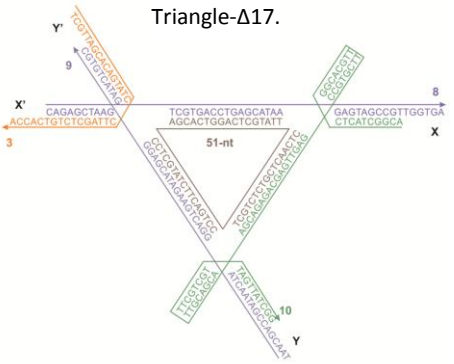


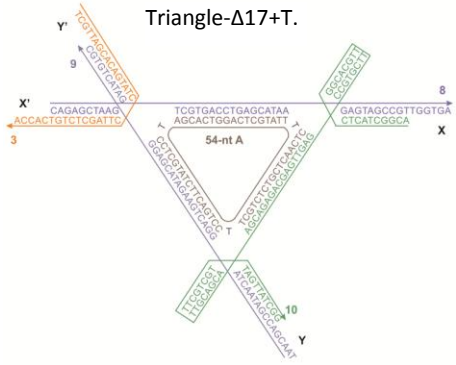
Fig. S6 AFM images of a control experiment using linear 48-nt in triangle-Δ16 at progressively increasing resolution, where a2 is the zoom-in view of a1.

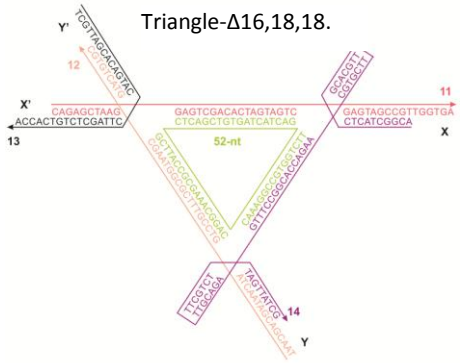
1.9 Designs of DNA triangle tiles forming nanotubes

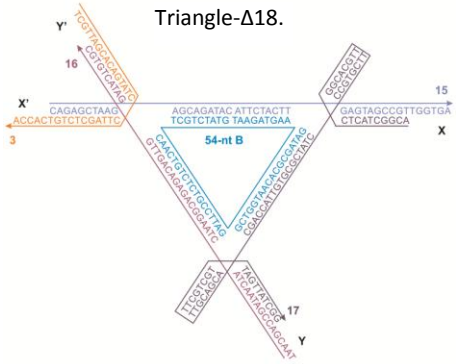












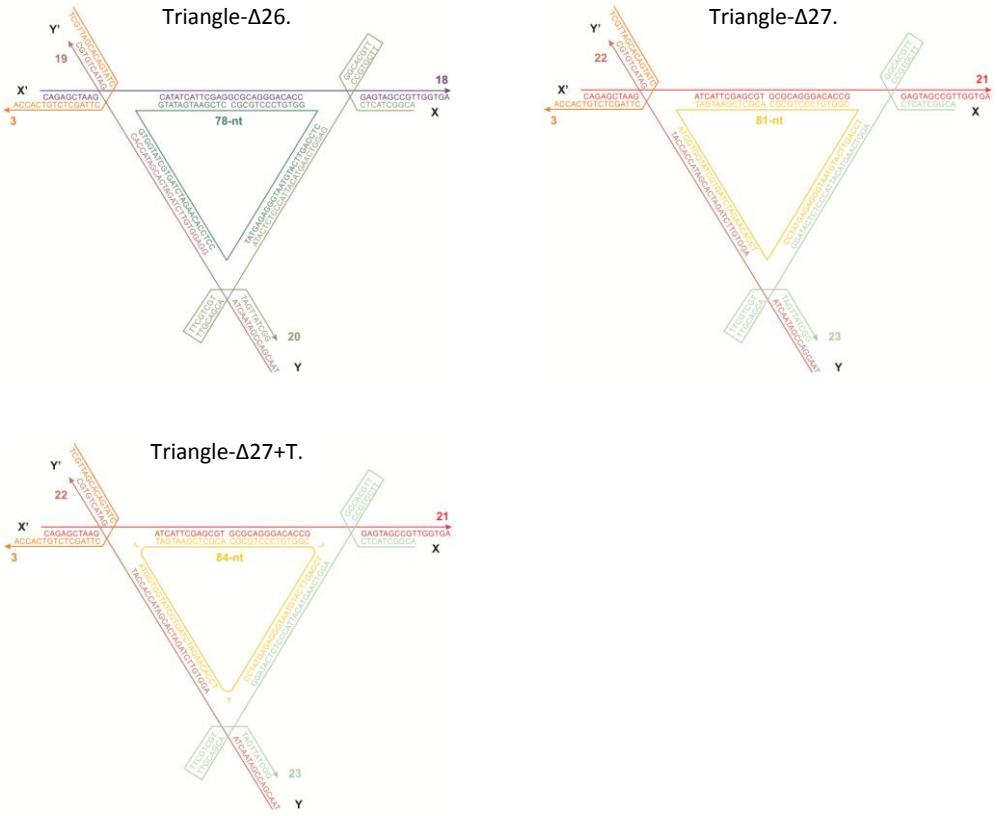


Fig. S7 Designs of triangle-Δn tiles, where "n" shows side length of the triangle. Pairs of X, X' and Y,Y' are complementary sticky ends respectively.

1.10 Oligonucleotide sequences used in this study

Table S1 Sequence codes of linear template strands (5' phosporylation)

Template ID*	Sequence (5' to 3')
48nt (NT-1)	ACTCGTATTCTCAACTCGTCTCTGCCCTGACTTCTATGCCCAGCCCTG
50nt (NT-2)	ACTCGTATTCTCAACTCGTCTCTGCTCCTGACTTCTATGCTCCAGCACTG
51nt (NT-3)	ACTCGTATTCTCAACTCGTCTCTGCTCCTGACTTCTATGCTCCAGCACTGG
52nt (NT-5)	CTCAGCTGTGATCATCAGTTCTGGTGCCGGAAACCAGGCAAAGCGCCATTCG
54nt-A (NT-4)	ACTCGTATTTCTCAACTCGTCTCTGCTTCCTGACTTCTATGCTCCTAGCACTGG
54nt-B (NT-6)	TAAGATGAAGATAGCGCACAATGGTCGGATTCCGTCTCTGTCAACTCGTCTATG
78nt (NT-7)	CGCGTCCCTGTGGCTCCAGTTCATGTAATGGGAGAGTATCCTCCACAAGATCTAGTGCTATGGTGGTATAGTAAGCTC
81nt (NT-8)	CGCGTCCCTGTGGCTCCAGTTCATGTAATGGGAGAGTATCCTCCACAAGATCTAGTGCTATGGTGGTATAGTAAGCTCGCA
84nt (NT-9)	CGCGTCCCTGTGGCTTCCAGTTCATGTAATGGGAGAGTATCCTTCCACAAGATCTAGTGCTATGGTGGTATTAGTAAGCTCGCA
(*) Asterisk shows respective nanotubes arising from corresponding template strands.	

Table S2 Sequence codes of splint strands

Splint ID	Sequence (5' to 3')
48nt splint	ATACGAGTCAGGGCTG
50nt splint	ATACGAGTCAGTGCTG
51nt splint	ATACGAGTCCAGTGCT

52nt splint	CACAGCTGAGCGAATGGCGC
54nt-A splint	ATACGAGTCCAGTGCT
54nt-B splint	TCATCTTACATAGACG
78nt splint	CAGGGACGCGGAGCTTACTA
81nt splint	CAGGGACGCGTGCGAGCTTA
84nt splint	CAGGGACGCGTGCGAGCTTA

Sequence codes of staple strands

Table S3 Sequence codes of staple strands (IDs correspond to their respective labels in the triangles of Figure S7)

ID	Sequence (5’ to 3’)
1	AGTGGTTGCCGATGAGATACGAGTCAGGGCTGGAATCGAGAC
2	CGTGTCATAGGGCATAGAAGTCAGGGATCAATAGCCAGCAAT
3	ACCACTGTCTCGATTCCTATGACACGATTGCT
4	GGCTATTGATTGCTGCTTTTGCAGCACAGAGACGAGTTGAGACCGTGCTTTTGCACGGCTCATCGGCA
5	AGTGGTTGCCGATGAGAGTTGAGAATACGAGTGAATCGAGAC
6	CGTGTCATAGCAGTGCTGGAGCATAGAATCAATAGCAGCAAT
7	GCTATTGATTGCTGCTTTTGCAGCAAGTCAGGAGCAGAGACGCGTGCTTTTGCACGCTCATCGGCA
8	AGTGGTTGCCGATGAGAATACGAGTCCAGTGCTGAATCGAGAC
9	CGTGTCATAGGGAGCATAGAAGTCAGGATCAATAGCCAGCAAT
10	GGCTATTGATTGCTGCTTTTGCAGCAAGCAGAGACGAGTTGAGCCGTGCTTTTGCACGGCTCATCGGCA
11	AGTGGTTGCCGATGAGGATGATCACAGCTGAGGAATCGAGAC
12	CGTGTCATGCGAATGGCGCTTTGCCTGATCAATAGCAGCAAT
13	ACCACTGTCTCGATTCCATGACACGATTGCT
14	GCTATTGATTCTGCTTTTGCAGAGTTTCCGGCACCAGAACTCGTGCTTTTGCACGCTCATCGGCA
15	AGTGGTTGCCGATGAGTTCATCTTACATAGACGAGAATCGAGAC
16	CGTGTCATAGGTTGACAGAGACGGAATCATCAATAGCCAGCAAT
17	GGCTATTGATTGCTGCTTTTGCAGCACGACCATTGTGCGCTATCCCGTGCTTTTGCACGGCTCATCGGCA
18	AGTGGTTGCCGATGAGCCACAGGGACGCGGAGCTTACTATACGAATCGAGAC
19	CGTGTCATAGCACCATAGCACTAGATCTTGTGGAGGATCAATAGCCAGCAAT
20	GGCTATTGATTGCTGCTTTTGCAGCAATACTCTCCCATTACATGAACTGGAGCCGTGCTTTTGCACGGCTCATCGGCA
21	AGTGGTTGCCGATGAGGCCACAGGGACGCGTGCGAGCTTACTAGAATCGAGAC
22	CGTGTCATAGTACCACCATAGCACTAGATCTTGTGGAATCAATAGCCAGCAAT
23	GGCTATTGATTGCTGCTTTTGCAGCAGGATACTCTCCCATTACATGAACTGGACCGTGCTTTTGCACGGCTCATCGGCA