This version of the Supporting Information replaces the version published on 20th July 2017. A change has been made to the M1 sequence as an error was present in the previous version.

One DNA Strand Homo-Polymerizes into Defined Nanostructures

Mo Li[1], Hua Zuo (左华)[2*], Jinwen Yu[1], Xinfeng Zhao (赵新锋)[1,3], Chengde Mao (毛成德)[1*]

[1]Department of Chemistry, Purdue University, West Lafayette, IN, 47907, USA

[2]College of Pharmaceutical Sciences, Southwest University, Chongqing, 400716, China

[3]Key Laboratory of Resource Biology and Biotechnology in Western China, Ministry of Education; College of Life Science, Northwest University, Xi'an 710069, China

Supporting Information

Experiment details

Sequence Design. All sequences are designed by a computer software SEQUIN (Seeman, N. C. *J. Biomol. Struct. Dyn.* **1990**, *8*, 573-581) and all the schemes were drawn by a computer software Tiamat [Williams, S., Lund, K., Lin, C., Wonka, P., Lindsay, S., and Yan, H. (2008) Tiamat: A three-dimensional editing tool for complex DNA structures, The 14th International Meeting on DNA Computing, Prague, Czech Republic]. The requirement of sequence complementary is outlined in Figure 4a. During the sequence design, we use SEQUIN to make sure the required sequence complementarities outlined in Figure 4a and avoid undesired sequence complementarities. As long as the sequence complementarities are satisfied, the exact DNA sequences do not matter.

Oligonucleotides. All DNA strands were purchased from IDT, Inc. and purified by denaturing PAGE. DNA sequences were listed below:

M1: 5'-AGATGCGATTACTGACTGCCTCGATAGCGCCTGTACACCG TTTTCGGACA TGTGGCGCTATCGGCATCTAGGCAGTCAGTAATC-3'

M2: 5'-GATGCCTGACTGCCTACGATAGCGCCTGTACACCGTTTTCGGACATGTGG CGCTATCGGCATCTAGGCAGTCAG-3'

M3: 5'-AGATGCCTGACTGCCTCGATAGCGCCTGCGCACCGTTTTCGGACGCGTGGC GCTATCGGCATCTAGGCAGTCAG-3'

M4: 5'-GCAGCGCCCTCGTCTATCCGTCGCTGCATTACTTGCTCCGCCTGTACACCG TTTTCGGACATGTGGCGGAGAACTACAAGTAATACGGATAGACGAGGG-3'

M5: 5'-CTGACTGCCTACGATAGCGCCTGTACACCGTTTTCGGACATGTGGCGCTAT CGGCATCTAGGCAGTCAG-3'

M6: 5'-CTGACTGCCTACGATAGCGCCTGTTGTCCGACGTCGGACAACAGGCGCTAT CGGCATCTAGGCAGTCAG-3'

T1 5'-GCAGCGCCCTCGTCTATCCGTCGCTGCATTACTTGCTCCGCC-3'

T2 5'-GGCGGAGAACTACAAGTAATACGGATAGACGAGGG-3'

T3 5'-ACATGTGGCGGAGAACTACAAGTAATACGGATAGACGAGGG-3'

List of DNA nanostructures and their component DNA strands:

1D ladder: **M1**;

2D array: **M2**;

1D chain: **M3**;

3D prism: **M4**;

Reference triangle: **T1** + **T2**;

Reference prism: **T1** + **T3**.

Formation of DNA complex

For 1D and 2D structures: DNA strand was dissolved in TAE/Mg^{2+} buffer and a mica plate (Ted Pella, Inc.) was immersed into the solution. The solution was slowly cooled down from 95 °C to 22 °C over 48 hours in a water bath. TAE/Mg^{2+} buffer contained 40 mM tris base (pH 8.0), 20 mM acetic acid, 2 mM EDTA, and 12.5 mM magnesium acetate.

For 3D triangular prism and controls: DNA strands were combined in TAE/Mg^{2+} buffer and were incubated at 95 °C for 5 min, 65 °C for 30 min, 55 °C for 30 min, 37 °C for 30 min, 22 °C for 30 min. The solution was stored at 4 °C.

For the samples in Fig. 4, the Mg^{2+} concentration in TAE/Mg^{2+} buffer was adjusted to 20 mM. There were three methods for sample preparations. Method 1 – Quench (22 °C): DNA solutions was incubated at 95 °C for 5 min, then quenched at 22 °C immediately and followed by incubation at 22 °C for 2 hours. Finally the DNA samples were stored at 4 °C. Method 2 – Quench (37 °C): DNA solutions were incubated at 95 °C for 5 min, then quenched at 37 °C immediately and followed by incubation at 37 °C for 2 hours. Finally, the DNA samples were stored at 4 °C. Method 3 – Annealed: DNA solutions were incubated at 95 °C for 5 min, 65 °C for 30 min, 55 °C for 30 min, 37 °C for 30 min, 22 °C for 30 min. The solutions were stored at 4 °C.

Polyacrylamide Gel Electrophoresis

10% denaturing PAGE gel was prepared with 19:1 acrylamide / bisacrylamide solution, 8 M urea, and TBE buffer, containing 89 mM tris base (pH 8.0), 89 mM boric acid and 2 mM EDTA.

The gel was run at 55 °C under 650 V on Hoefer SE 600 electrophoresis system and then was stained with ethidium bromide (Sigma). The major band was cut under UV light and was eluted out.

5% native PAGE gel was prepared with 19:1 acrylamide / bisacrylamide solution and TAE/Mg^{2+} buffer. The gel was run at 4 °C. Then stained with Stains-All (Sigma) and scanned by an HP scanner (Scanjet 4070 Photosmart).

AFM imaging.

DNA 1D and 2D structures were first imaged in air. The mica annealed with the solution was taken out and washed by 25 μL 2 mM $Mg(Ac)_2$. Then dried by compressed air. Imaging was performed on a Bruker Multimode 8 AFM at SCANASYST-AIR mode with ScanAsyst-air Nitride probe (Bruker). Fourier transformation was done by Gwyddion, a modular program for scanning probe microscopy [David Nečas and Petr Klapetek. "Gwyddion: an open-source software for SPM data analysis." *Open Physics* **10**, 181-188 (2012).]. DNA 1D ladder was also scanned in fluid. The mica annealed with the solution was taken out and 25 μL TAE/Mg^{2+} buffer was added onto the mica surface. Then imaged under SCANASYST-FLUID mode with ScanAsyst-fluid+ probe (Bruker).

DNA 3D triangular prism was scanned in air. 25 μL 1 mM $NiCl_2$ solution was deposited onto fresh cleaved mica surface for 30 seconds to enhance DNA particle absorption, and then was blown away with compressed air. 5 μL annealed solution was dropped on the substrate and incubated for 1 min. The mica surface was washed with 25 μL 2 mM $Mg(Ac)_2$ before imaging in air.

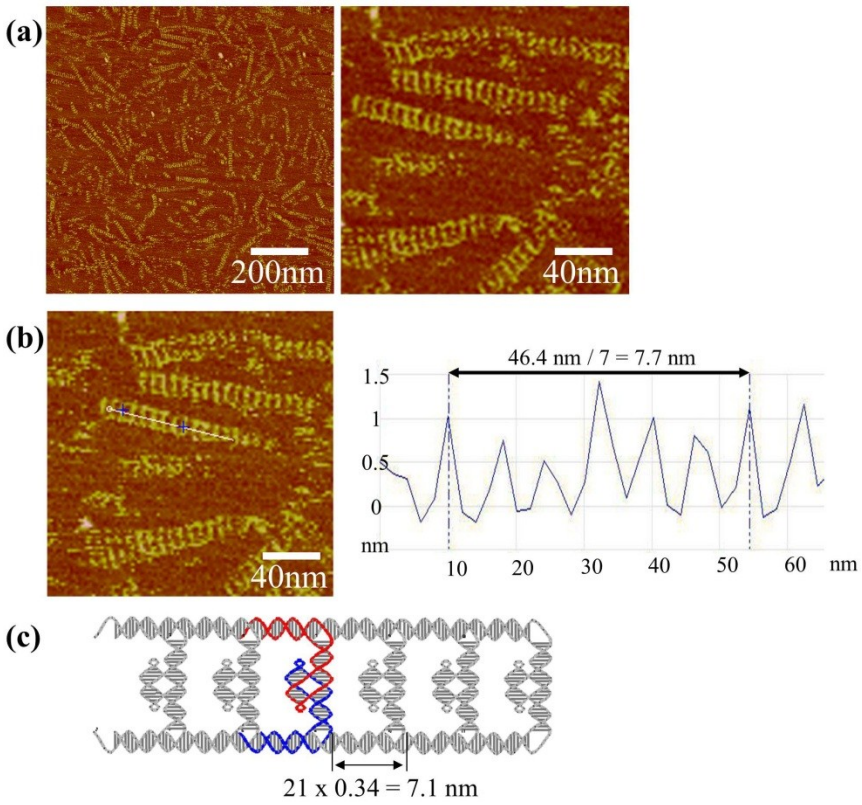

Figure S1. AFM imaging of 1D ladders assembled from M1 in fluid. (a) A large field view of AFM image (left) and its zoom-in image (right). (b) A section analysis of a ladder along the line indicated in the image (left). (c) Calculation of the repeating distance along the ladder.

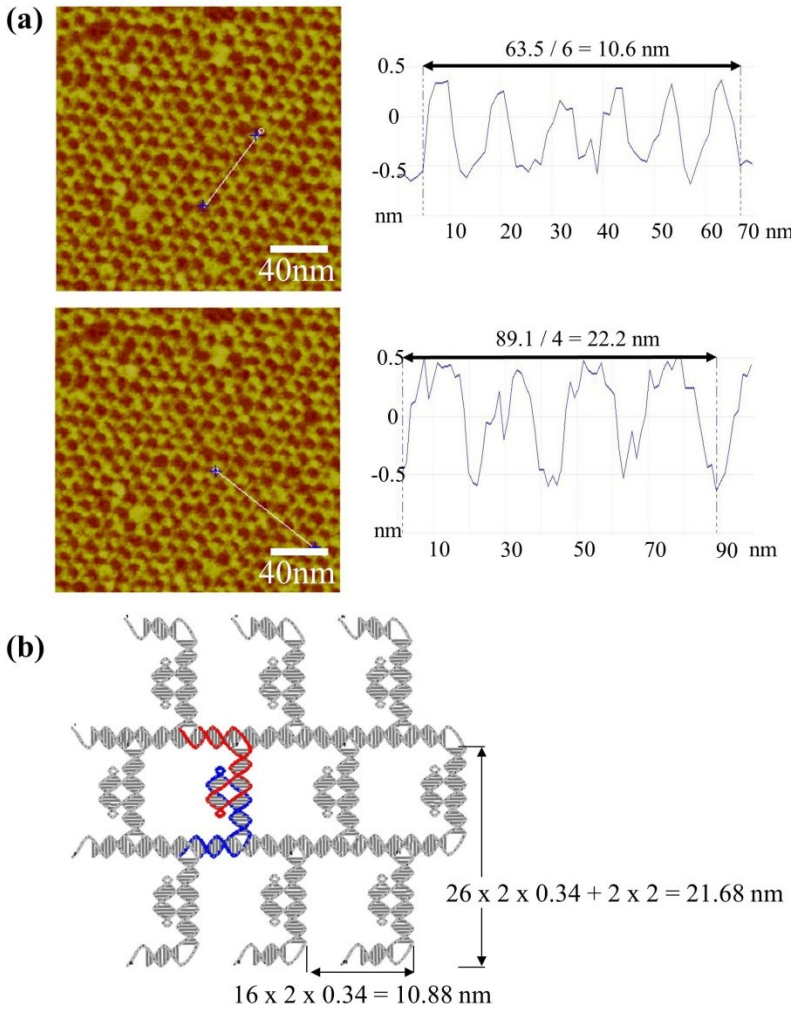

Figure S2. Section analysis of M2 -- 2D arrays assembled from M1. (a) The height profiles (right) are along the lines indicated on the AFM images (left). (b) The calculation of the repeating distances.