Supporting Online Material

Material and methods:

Complex Design, Assembly and Characterization. The design of the 4x4 tiles and superstructure assemblies analyzed here were based on the structure of immobile 4-arm branched junctions. The subsequence used for all bulged loops was T_4. Sequences were designed with the program SEQUIN (*S1*) to minimize the chance of undesired complementarity and sequence symmetry. The strand sequences for the molecules used here are given below. Custom oligonucleotides were purchased from Integrated DNA Technology (www.idtdna.com) and purified by PAGE. Complexes were formed by mixing a stoichiometric quantity of each strand, as estimated by OD_{260} in 20 mM Tris (pH 7.6), 2 mM EDTA, 12.5 mM $MgCl_2$. The final concentration of DNA was between 0.5 and 1.0 μM, and the final volume was 10 - 50 μL. Oligo mixtures were cooled slowly from 90 °C to 20 °C in a heating block over approximately 16 hours to facilitate hybridization. Non-denaturing polyacrylamide gel electrophoresis and thermal profile experiments were described previously (*S2*).

AFM Imaging. A 5 μL sample was spotted on freshly cleaved mica (Ted Pella, Inc.) and left to adsorb to the surface for 3 min. then 30 μL 1xTAE/Mg buffer was placed onto the mica. Imaging was performed under 1xTAE/Mg in a fluid cell on a Multimode NanoScope IIIa, using NP-S tips (Veeco Inc.).

SEM Imaging. All SEM images were taken at 10 -15 kV (accelerating voltage) with e-beam spot size = 3 in a FEI XL30 Thermal Field Emitter Scanning Electron Microscope

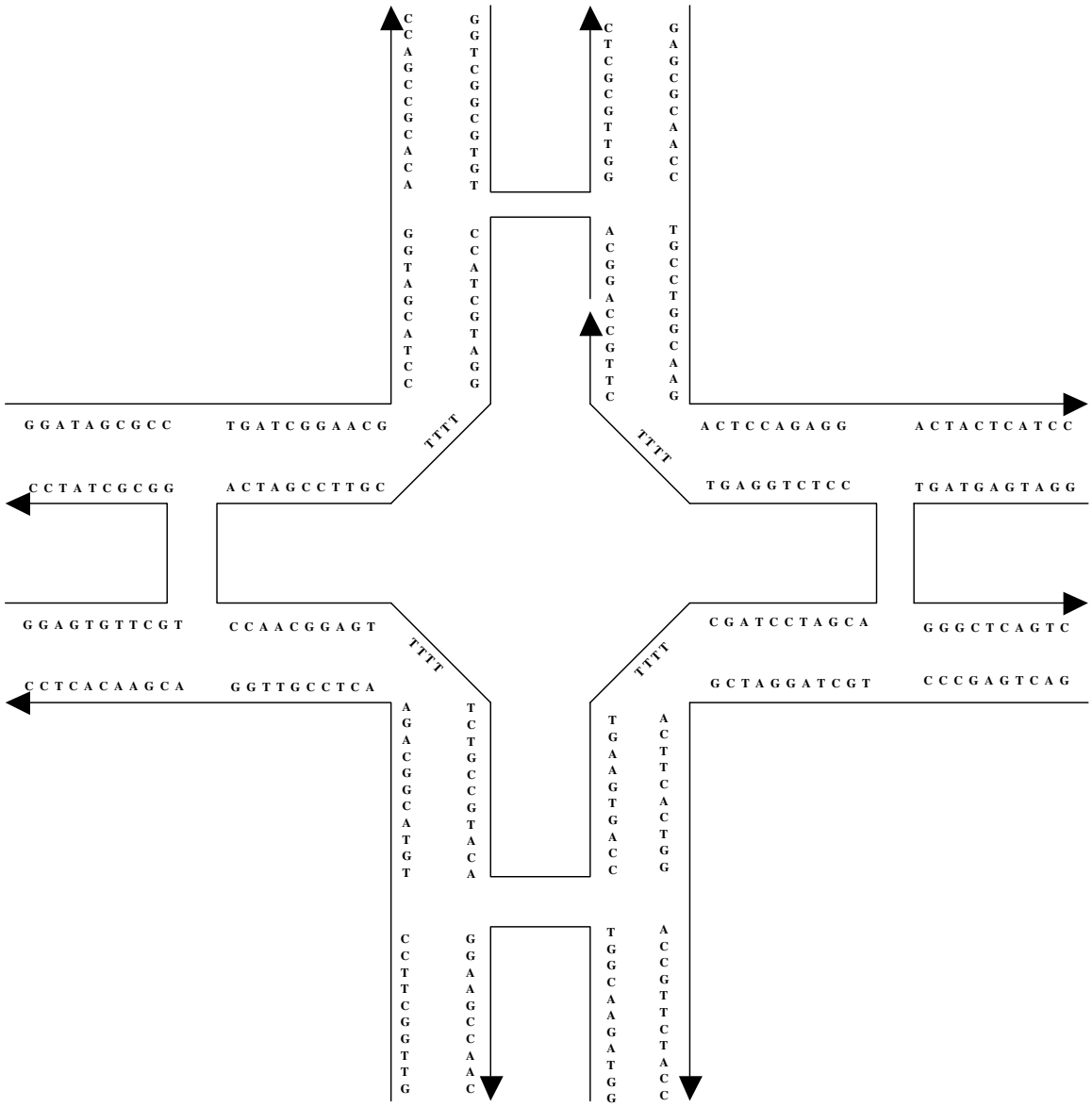
made by Philips. Chamber pressure was between 1.0 X 10^{-7} and 1.0 X 10^{-6} mbar. All samples were prepared on doped n-type silicon substrate with resistivity = 2.0 X 10^{-5} Ωm and 500 nm thermal oxide layer prepared using the RCA cleaning process without the oxide stripping step.

Streptavidin binding to self-assembled DNA nanogrids: Biotinyalated oligo was purchased from www.idtdna.com. Streptavidin was purchased from Rockland Inc. (Gilbertsville, PA, USA). 4x4 nanogrids containing biotinylated oligo were annealed as described in the complex assembly section in supplementary information. DNA complex:streptavidin ratio is optimized as 1μM:1μM. After adding streptavidin to the DNA nanogrids, the solution was incubated for overnight at 4 °C before imaging.

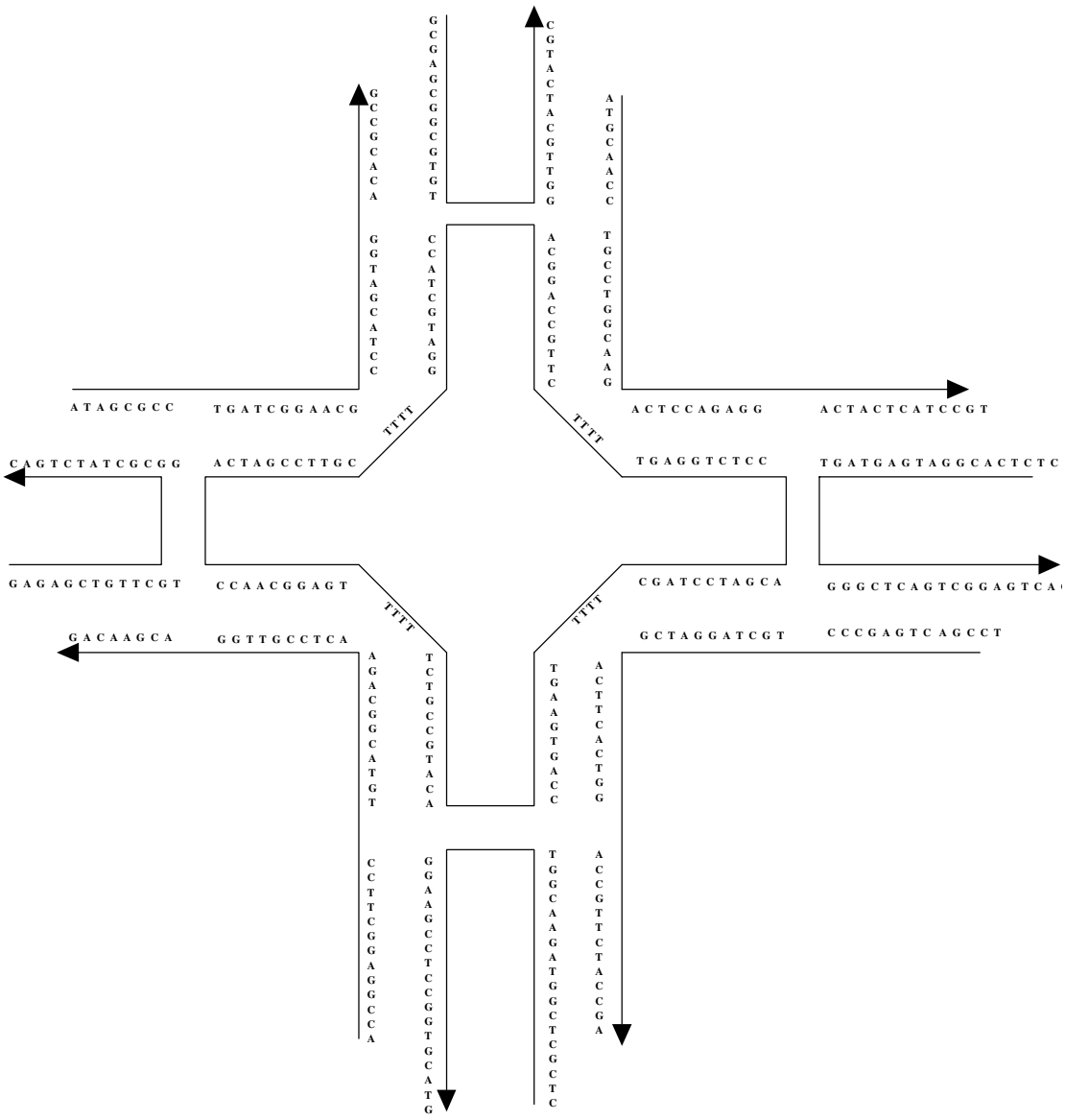
Two-Step Metallization. 1). First, the 4x4 ribbon lattice was seeded with silver using the glutaraldehyde method (*S3*). Annealed DNA was incubated with 0.2% glutaraldehyde in 1xTAE-Mg buffer on ice for 20 minutes, then at room temperature for 20 minutes, then the sample was loaded into a Slide-A-Lyzer Mini Dialysis unit (Pierce, Rockford, IL) and dialysised overnight at 4°C in 1 L of 1xTAE Mg buffer. The published method was modified in that the silver seeding was done in aqueous solution for 20 minutes instead of on substrate. Aldehyde-derivatized DNA was incubated in the dark with a 0.1 M solution of $AgNO_3$ in 25% ammonia buffer (pH 10.5) at room temperature for 30–90 minutes, then 10 μL was deposited onto silicon substrate, and excess reagent was rinsed off with distilled water. 2). In the second step, HQ SILVER^{TM}-EM Formulation (www.nanoprobes.com) was used according to the manufacturer's instructions. One unit

of initiator (A) was mixed with a unit of moderator (B) and a unit of activator (C), then freshly deposited 10 μL of this mixture onto silicon substrate for 5 minutes. Finally excess reagent was rinsed off again with distilled water.

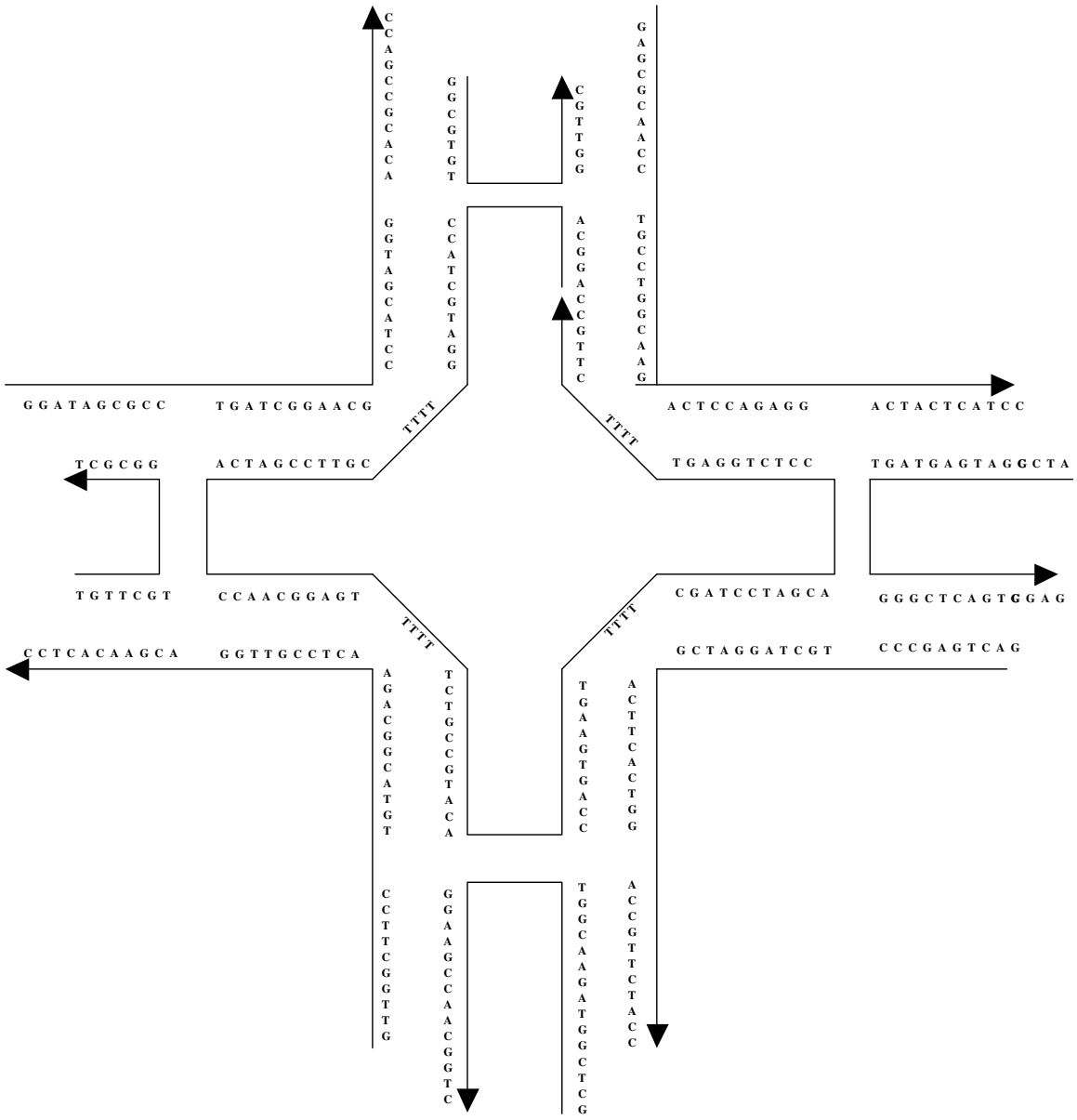
DNA strand structure and sequences used in Fig. 1A and Fig. S1:



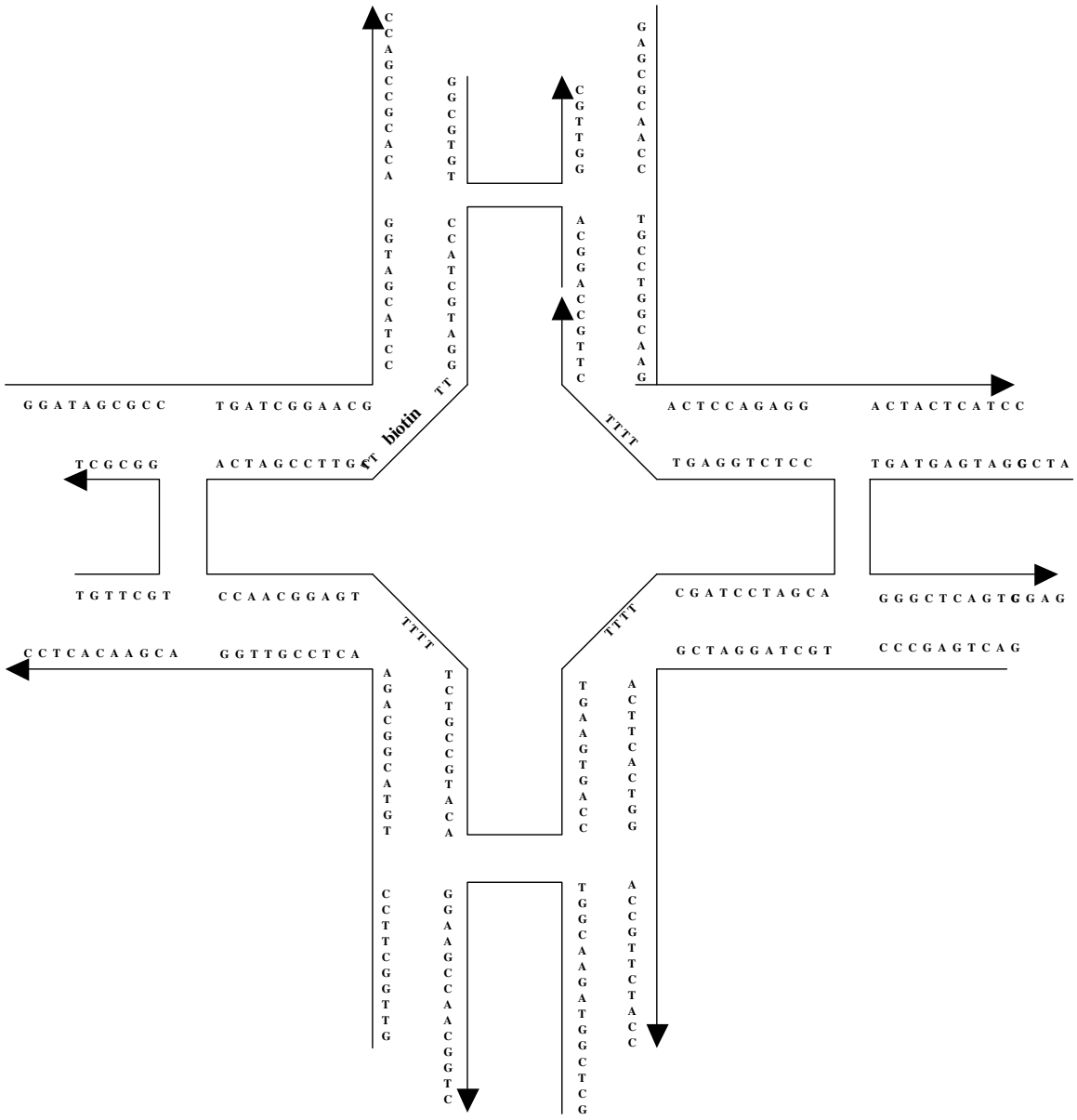
DNA strand structure and sequences used in Fig. 1B & Fig. 3:



DNA strand structure and sequences used in Figure 1C:



DNA strand structure and sequences used in Fig. 2:



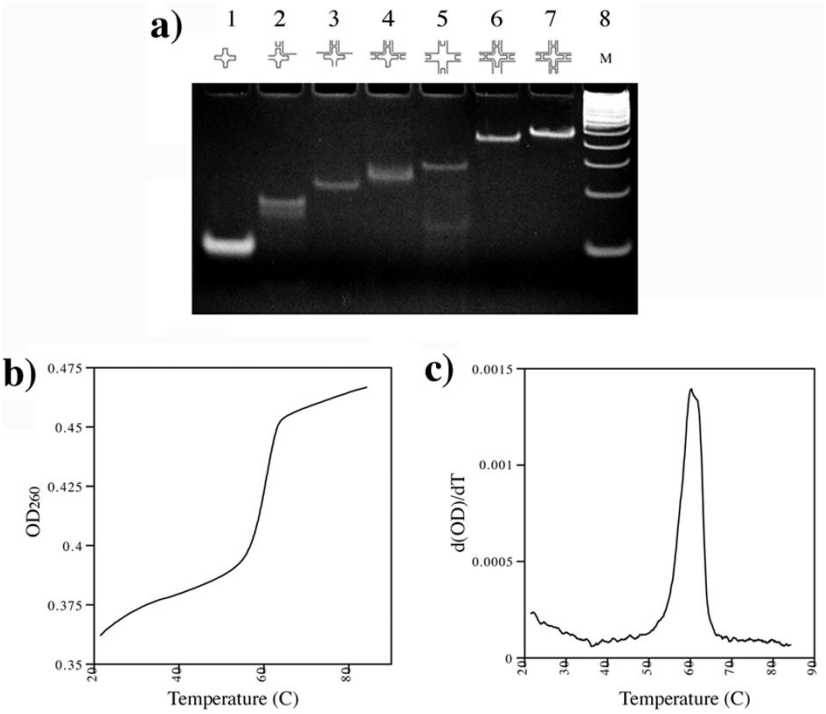


Fig. S1 shows characterization of the 4x4 tile structure using non-denaturing electrophoresis and thermal transition experiments. **a).** An 8% polyacrylamide gel (ethidium bromide stained) showing association complexes between various equimolar combinations of the 4x4 DNA complex component strands. Equimolar mixtures at 1 μM concentration per included strand were annealed and run on the gel at room temperature. Strands included in the annealings are indicated in the drawing above each lane. Lane 8 contains 50 bp DNA ladder size markers. The 4x4 complex runs as a single band on non-denaturing gels, without any higher molecular weight byproducts (from unexpected base-pairings between two or more complexes) nor lower molecular weight byproducts (from dissociated complex), indicating the 4x4 tile complex is a stable structure in the buffer used. **b)**. Thermal transition profile. The left panel shows the relative change in optical density at 260 nm as a function of temperature. The right panel shows the first derivative of the 4x4 complex melting data. The results show that the 4x4 complex melts cooperatively, as a single transition, with T_m= 60 °C.

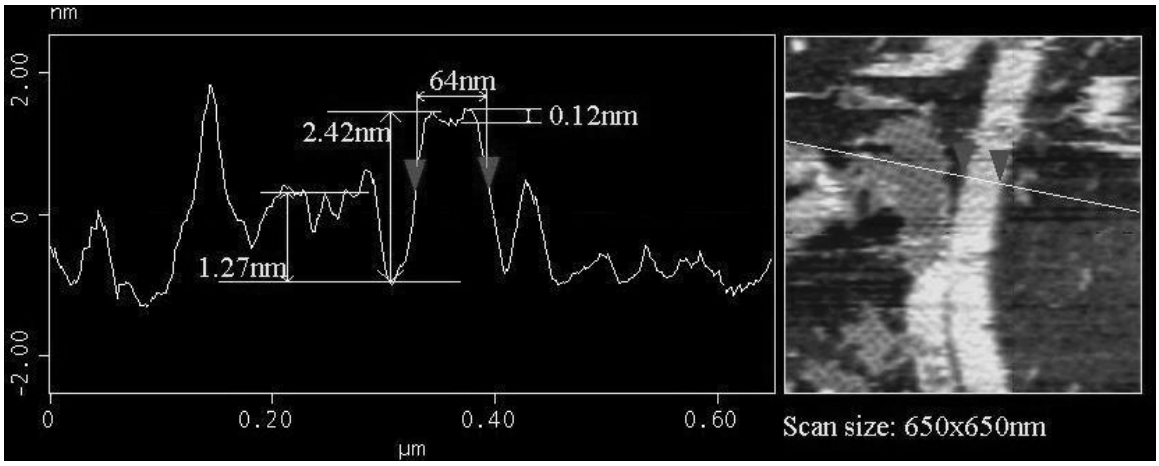


Fig. S2 shows a cross-section height profile analysis of an AFM image which contains 4x4 nanoribbon and single layered 4x4 flat lattice. The AFM image clearly shows that the nanoribbon structure has two layers compared to the flat lattice. Also the edges of the ribbon appear slightly higher (~0.12 nm) than the middle, indicating a finite radius of curvature for the squashed tube structure.

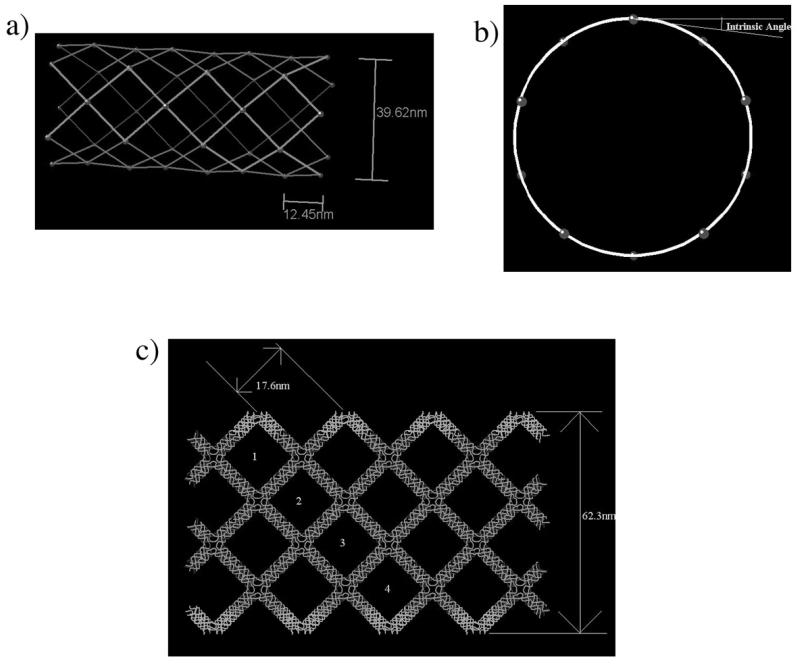


Fig. S3 illustrates a model of the 4x4 nanoribbon structure. a) Side view of a proposed tube structure where each dot represents the center of a 4x4 tile (blue and red dots are on separate ring layers). b) End-on view of a tube showing 5 tiles in the terminal ring. From this model we can calculate an intrinsic angle of ~18 degrees. c) Overhead view of a tube squashed onto the surface of the substrate and forming a ribbon with four diagonal square cavities and ~62.3nm width. Note the saw-tooth edge formed by folding one row of 4x4 tiles along a diagonal running through their most flexible region, the TTTT loops between adjacent arms. The jagged edge along with the 45 degree diagonal containing four cavities are typically observed in high resolution AFM images of the ribbons.

Fig. S4 is a 10x10 um AFM image showing a wider sampling of 4x4 nanoribbons without metallization. The average ribbon length is ~5 um and we also see ribbons larger than 10 um.

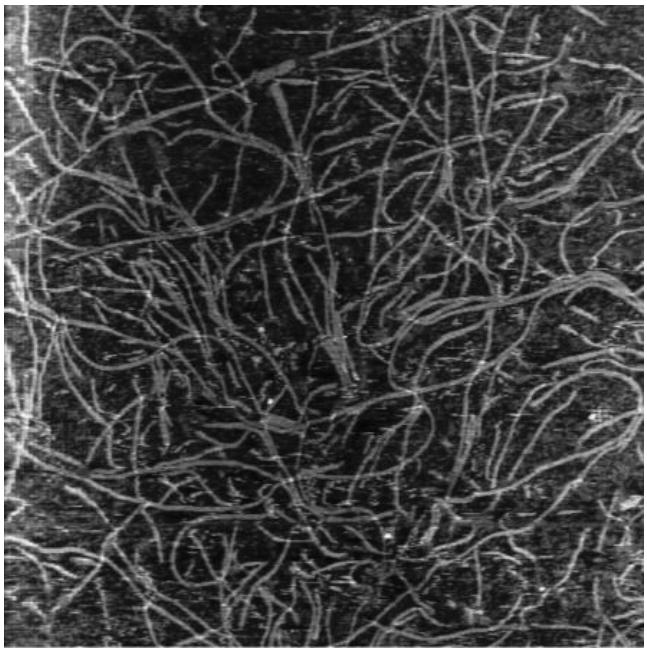
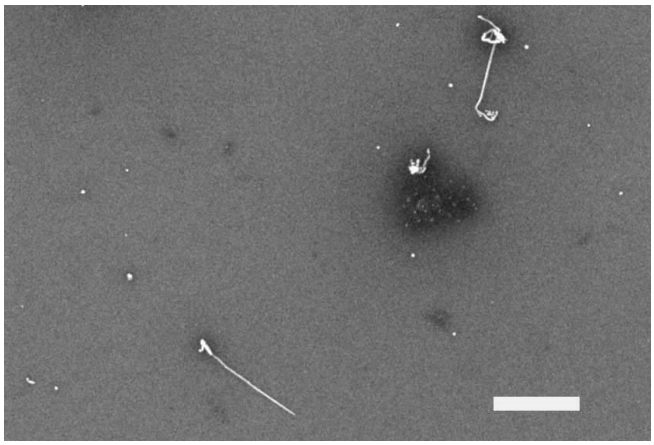


Fig. S5 is an SEM image showing two metallized 4x4 nanowires. The scale bar is 2 um. Due to the rinsing process, the metallized nanowires are less dense on substrate.



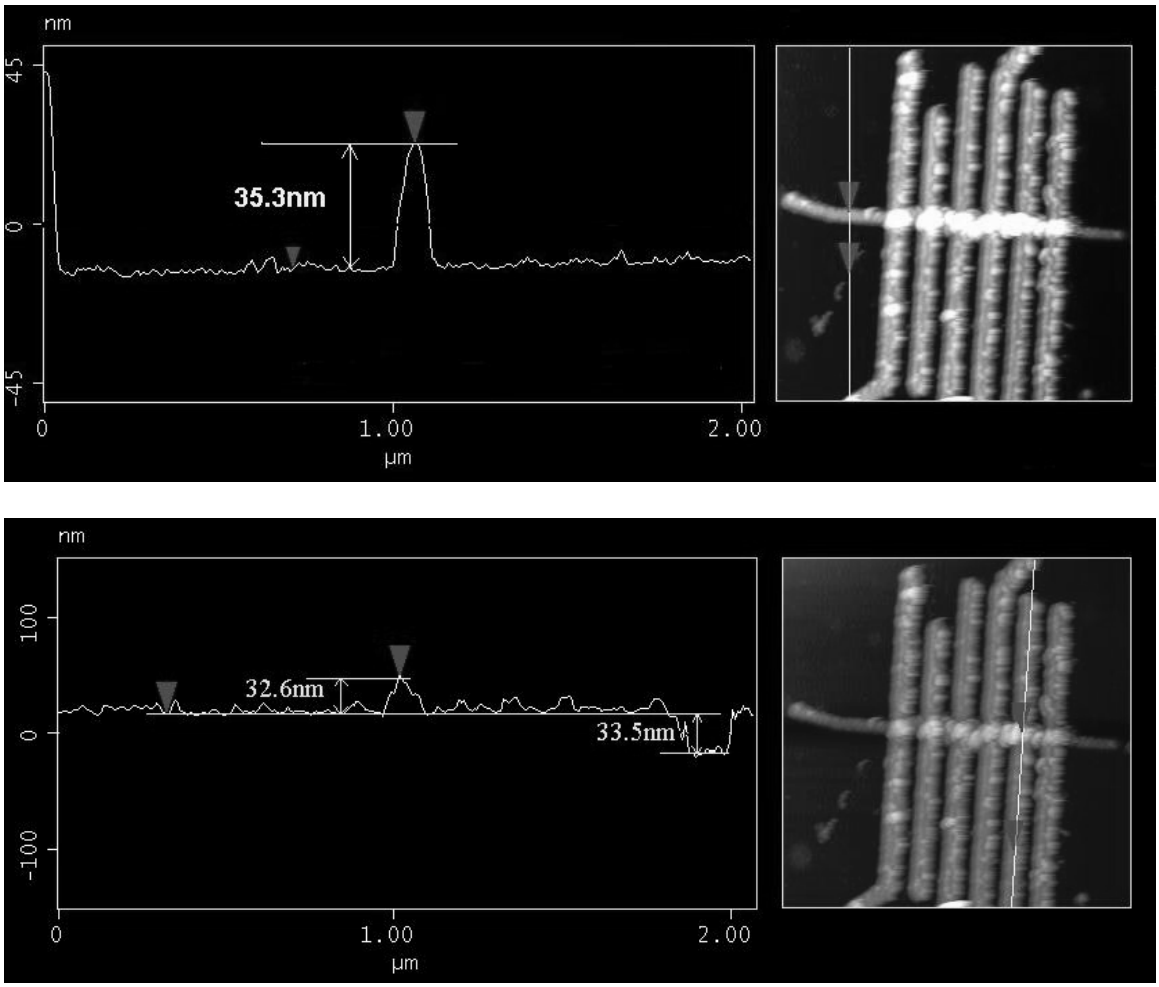


Fig. S6 shows AFM height profile measurements for the silver metallized 4x4 nanowire over gold electrode on silicon substrate. It clearly shows that the height of the nanowire is ~35 nm. SEM measurements (which gives more accurate measurements on the lateral direction) of the width of this nanowire is ~43 nm. After silver metallization, the 4x4 nanoribbon is less likely to be flattened on solid substrate compare to the non-metallized nanoribbon, causing the increase of the height and decease of the width.

Reference:

S1. N. C. Seeman, *J. Biomol. Struct. Dyns.* **8**, 573 (1990).
S2. T. H. LaBean, *et al.*, *J. Am. Chem. Soc.* **122**, 1848 (2000).
S3. K. Keren, *et al.*, *Science*, **297**, 72 (2002).