Electronic Supplementary Information

Hairpin Embedded DNA Lattices Grown on a Mica Substrate†

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Fig. S1. Schematic diagrams of four possible hairpin sites on a DX tile.

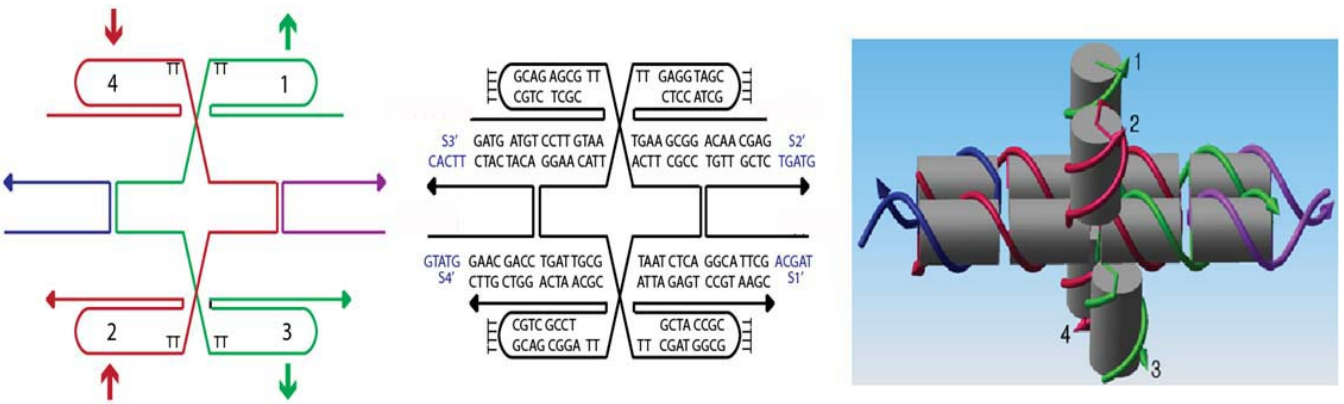

Fig. S2. Schematic diagrams of DX tiles participating in DNA crystals, (a) DX-NH, (b) DX-1SH, (c) DX-2SH, (d) DX-1DH and (e) DX-2DH. Complementary sticky end pairs are indicated by Sn and Sn' in sequence drawings (blue). 1s in parentheses after the tile name indicates the existence of a hairpin in the designated positions shown in **Fig. S1**. For instance, CS4 (1,0,0,1) has hairpins at the 1 and 4 sites.

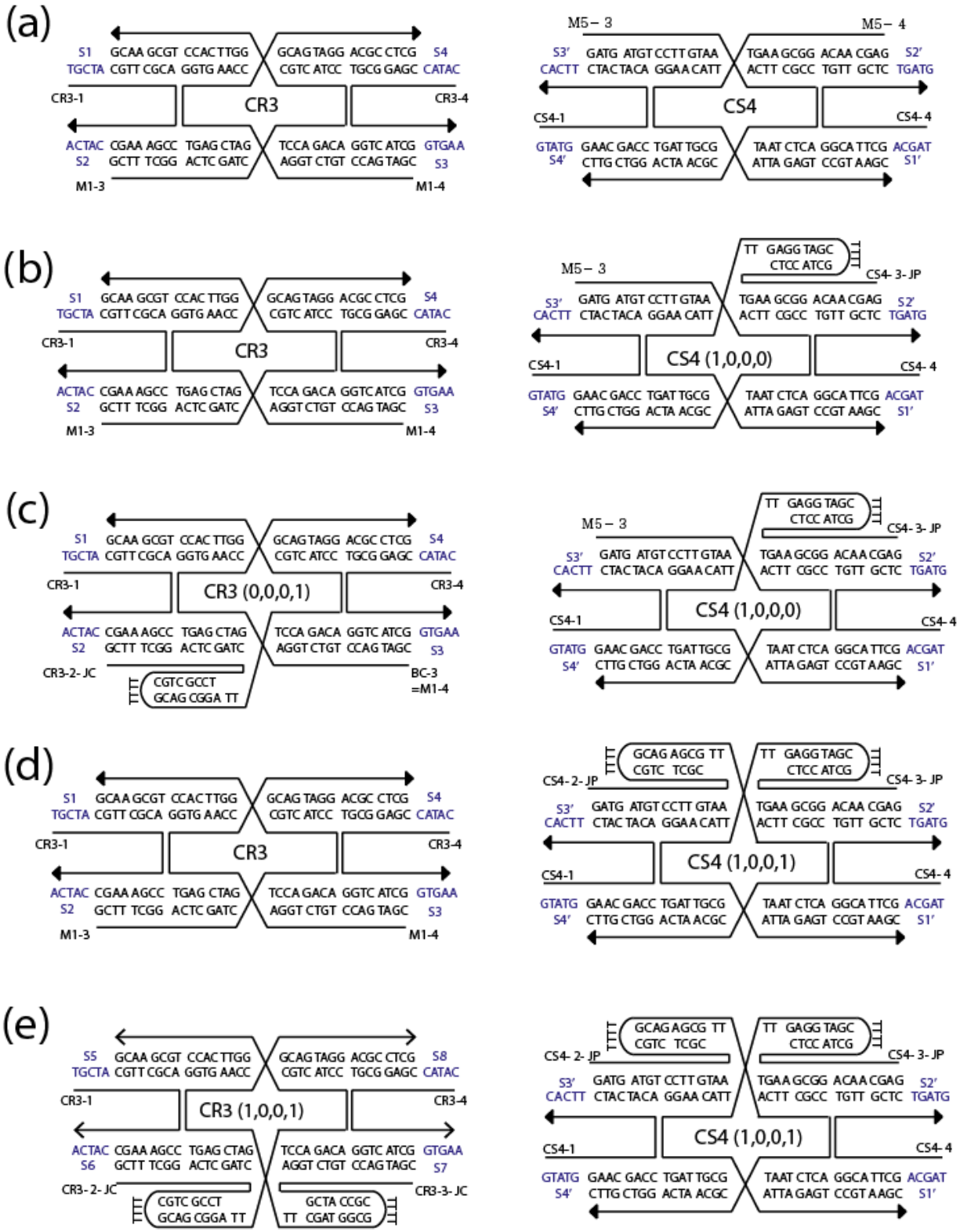

Table S1. Sequencing pool for DX tiles.

Strand name	No. of Nucleotides	DNA Base Sequences (5' to 3')
CR3-1	26	TGCTA CGTT CGCA CCGA AAGC CATCA
CR3-2-JC	70	GCTT TCGG ACTC GATC TCCG CTGC TTTT GCAG CGGA TT TCCA GACA CCTA CTGC GGTT CACC TGCG AACG
CR3-3-JC	70	CGAT GACC TGTC TGGA GCTA CCGC TTTT GCGG TAGC TT GATC GAGT GGTG AACC GCAG TAGG ACGC CTCG
CR3-4	26	CATAC CGAG GCGT GGTC ATCG GTGAA
CS4-1	26	GTATG GAAC GACC ACAT CATC TTCAC
CS4-2-JP	70	GATG ATGT CCTT GTAA CGCT CTGC TTTT GCAG AGCG TT ACTT CGCC ACTC TAAT CGCA ATCA GGTC GTTC
CS4-3-JP	70	GAGC AACA GGCG AAGT CTCC ATCG TTTT CGAT GGAG TT TTAC AAGG TGAT TGCG ATTA GAGT CCGT AAGC
CS4-4	26	TAGCA GCTT ACGG TGTT GCTC TGATG
M1-3	48	GCTT TCGG ACTC GATC TCCA GACA CCTA CTGC GGTT CACC TGCG AACG
M1-4	48	CGAT GACC TGTC TGGA GATC GAGT GGTG AACC GCAG TAGG ACGC CTCG
M5-3	48	GATG ATGT CCTT GTAA ACTT CGCC ACTC TAAT CGCA ATCA GGTC GTTC
M5-4	48	GAGC AACA GGCG AAGT TTAC AAGG TGAT TGCG ATTA GAGT CCGT AAGC

Table S2. Individual tile names and their composition.

	CR3	CS4	CR3 (1,0,0,1)	CR3(0,0,0,1)	CS4(1,0,0,0)	CS4(1,0,0,1)
Strand 1	CR3-1	CS4-4	CR3-1	CR3-1	CS4-4	CS4-4
Strand 2	M1- 3	M5-4	CR3-2-JC	CR3-2-JC	CS4-3-JP	CS4-3-JP
Strand 3	M1- 4	M5-3	CR3-3-JC	M1-4	M5-3	CS4-2-JP
Strand 4	CR3-4	CS4-1	CR3-4	CR3-4	CS4-1	CS4-1

Table S3. Detailed tile combinations in five different crystals: DX-NH, DX-1SH, DX-2SH, DX-1DH, and DX-2DH.

DX-NH	CR3	CS4
Strand 1	CR3-1	CS4-4
Strand 2	M1-3	M5-4
Strand 3	M1-4	M5-3
Strand 4	CR3-4	CS4-1

DX-1SH	CR3	CS4(1,0,0,0)
Strand 1	CR3-1	CS4-4
Strand 2	M1-3	CS4-3-JP
Strand 3	M1-4	M5-3
Strand 4	CR3-4	CS4-1

DX-2SH	CR3(0,0,0,1)	CS4(1,0,0,0)
Strand 1	CR3-1	CS4-4
Strand 2	CR3-2-JC	CS4-3-JP
Strand 3	M1-4	M5-3
Strand 4	CR3-4	CS4-1

DX-1DH	CR3	CS4(1,0,0,1)
Strand 1	CR3-1	CS4-4
Strand 2	M1-3	CS4-3-JP
Strand 3	M1-4	CS4-2-JP
Strand 4	CR3-4	CS4-1

DX-2DH	CR3(1,0,0,1)	CS4(1,0,0,1)
Strand 1	CR3-1	CS4-1
Strand 2	CR3-2-JC	CS4-2-JP
Strand 3	CR3-3-JC	CS4-3-JP
Strand 4	CR3-4	CS4-4

Table S4. Unit conversion between monomer (C_m) and molecular concentrations (C_{Mol}).

	Number of nucleotides (nt)	Molecular weight (325 Da. per nt)
Unit DX-NH	296	96,200 Da.
Unit DX-2SH	340	110,500 Da.
Unit DX-2DH	384	124,800 Da.

Conversion equation:

(Molarity) × (Avogadro number per mole) × (Number of nts per unit tile) × (Molecular weight per nt) × $(\frac{1.66 \times 10^{-24}\ \text{g}}{1\ \text{Da.}})$ × $(\frac{1\ \text{liter}}{1000\ \text{cm}^3})$

Ex.) 1 nM of DX-NH = $\frac{1\ \text{nmole}}{1\ \text{liter}} \times \frac{6.02 \times 10^{23}}{1\ \text{mole}} \times 296\ \text{nts} \times \frac{325\ \text{Da.}}{1\text{nt}} \times \frac{1.66 \times 10^{-24}\ \text{g}}{1\ \text{Da.}} \times \frac{1\ \text{liter}}{1000\ \text{cm}^3}$

$= 0.962 \times 10^{-7}\ \text{g/cm}^3$

Molecular concentration (C_{Mol}) of each DNA crystal (units: 10^{-7} g/cm^3).

	1 nM	5 nM	10 nM	20 nM
DX-NH	0.962	4.806	9.62	19.24
DX-2SH	1.104	5.521	11.042	22.084
DX-2DH	1.247	6.235	12.471	24.943

Fig. S3. AFM images of DX crystals (DX-NH, DX-1SH, DX-2SH, DX-1DH and DX-2DH) with C_m = 200 nM annealed in free solution. Scan sizes are 1 × 1 μm^2.

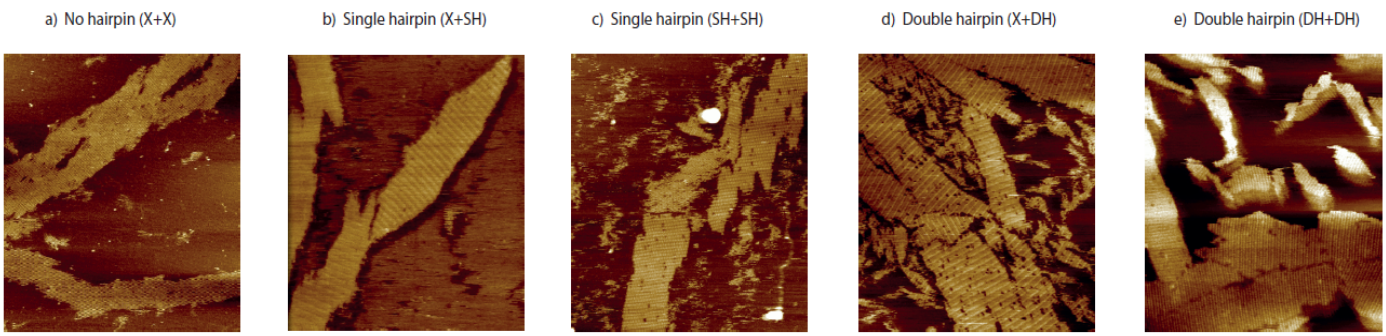

Fig. S4. AFM images and a schematic of a long hairpin lattice (DX-1LH). DX-1LH has hairpins twice longer than a hairpin used in **Fig. 1**. and **Fig. S2**. The lattice is self-assembled into extended 2D crystalline arrays with highly ordered structures on a mica substrate. (a) An AFM image of substrate-assisted grown hairpin-embedded DX-1LH lattices (Scan size, 1 × 1 μm^2). (b) An AFM image of DX-1LH annealed in free solution (Scan size, 1 × 1 μm^2). (c) Base sequences of DX-1LH motifs, one has no hairpin while the other has double hairpins positioned in upward and downward directions.

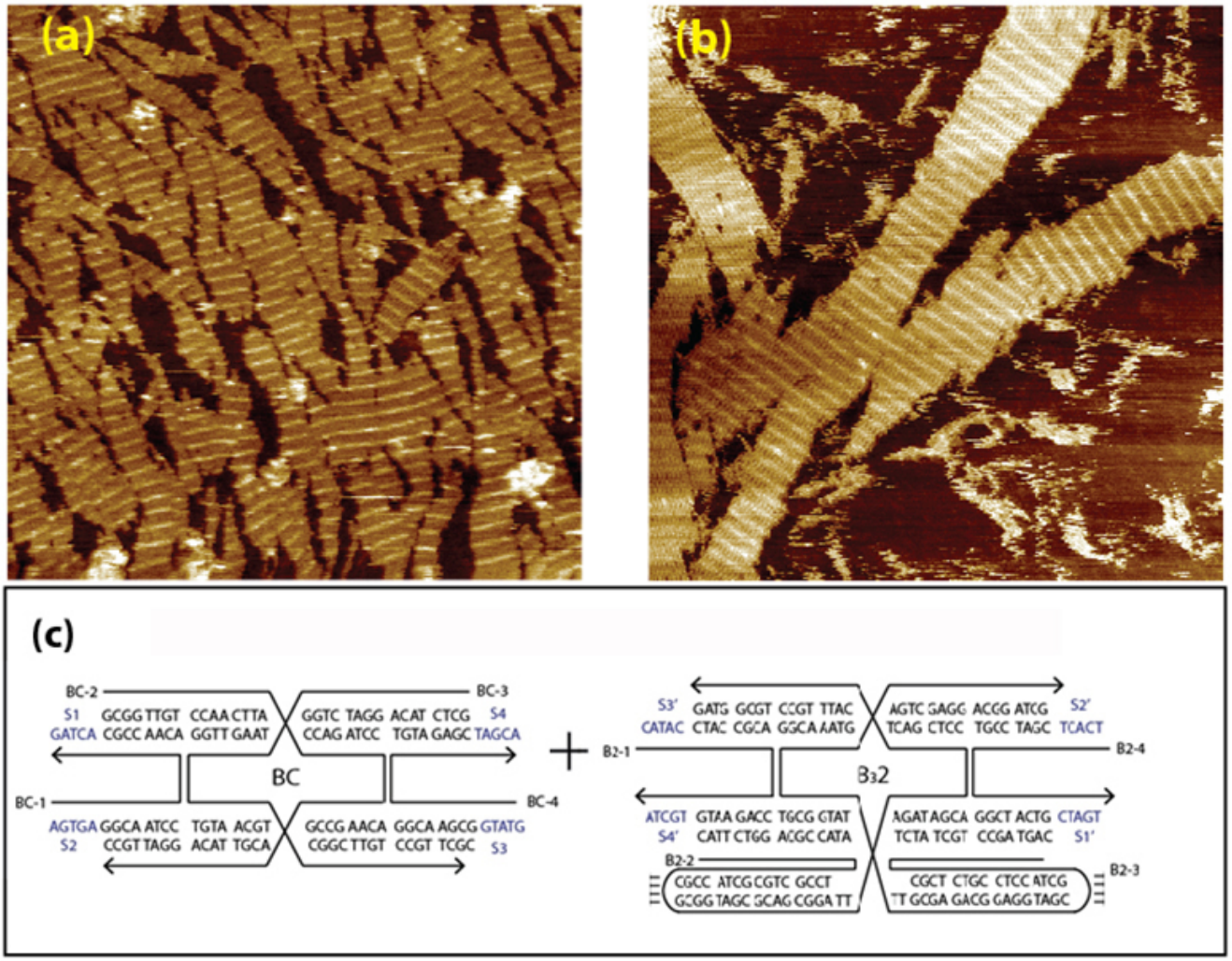

Fig. S5. AFM height analysis of substrate-assisted grown DX-1LH and DX-1DH. Average heights of DX-1LH and DX-1DH measured from the bare DX lattices are 1.1 nm and 0.5 nm respectively shown in (a) and (b). In addition, average heights of DX-1LH and DX-1DH measured from the surface of the substrate are 2.5 nm and 2.0 nm respectively. The height differences between measured and theoretical values are due to AFM tip pressure during collecting data and suppression by electrostatic interactions between DNA molecules and the given substrate. Although we have certain limitations to measure heights of hairpins precisely, we can observe the height differences of different length of hairpins like DX-1LH and DX-1DH easily.