Electronic Supplementary Information

DNA Reusability and Optoelectronic Characteristics of Streptavidin-Conjugated DNA Crystals on a Quartz Substrate†

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Materials and Methods

O_2 Plasma Exposure. The treatment of oxygen (O_2) plasma on quartz changes the surface properties from hydrophobic to hydrophilic. The O_2 plasma cleaner (Femto Science, Model: Cute) was used in the present study. The chamber was evacuated and then the oxygen plasma cleaning process commenced at a power of 50 W, base pressure of 5×10^{-2} torr, oxygen flow rate 45 sccm, working oxygen pressure $\sim 7.8 \times 10^{-1}$ torr, and plasma generation time 10 min.

DNA Crystal Growth on a Quartz Substrate. Synthetic oligonucleotides of DNA, purified by high performance liquid chromatography (HPLC), were purchased from BIONEER (Daejeon, Korea). The complexes were formed by mixing a 1× TAE/Mg^{2+} (40 mM Tris base, 20 mM acetic acid, 1 mM EDTA (pH 8.0), and 12.5 mM magnesium acetate) buffer solution which contains an equimolar mixture of 8 different DX strands. For annealing, the O_2 plasma treated quartz substrate along with the DNA strands were inserted into an AXYGEN-tube with a total sample volume of 250 μL which was then placed in a styrofoam box with 2L of boiling water and cooled slowly from 95 to 25°C over a period of 24 hours to facilitate the hybridization process. During the annealing process, the DX strands formed DX crystals on a given substrate and consequently these crystals completely covered the quartz substrate. We prepared the sample with a concentration of 50 nM, which was well above the saturation concentration of 10 nM for the full coverage of DX crystals.

Streptavidin binding to DXB crystals. Biotinylated oligos were purchased from BIONEER (Daejeon, Korea). Streptavidin was purchased from Rockland Inc. (PA, USA). A 200 nM solution of streptavidin was prepared in deionized water. A 1:1 ratio of streptavidin-DXB was prepared by directly pippetting streptavidin solution in the test tube.

AFM imaging and Raman spectra. For AFM imaging, a substrate assisted grown sample was placed on a metal puck using instant glue. 30 μL 1× TAE/Mg^{2+} buffer was added onto the substrate and another 10 μL of 1× TAE/Mg^{2+} buffer was dispensed into the silicon nitride AFM tip (Veeco Inc., USA). AFM images were obtained by a Multimode Nanoscope (Veeco Inc., USA) in the fluid tapping mode. Before measuring the Raman spectra, the samples were rinsed with deionized water, followed by gental blowing with nitrogen gas to remove chemical residues on the surface. The measurements were performed at room temperature with a confocal Raman microscope (WITec, alpha 300 R) at 532 nm.

Electrical Measurement. The electrical properties of DNA, DXB, and DXB+SA crystals were measured with a semiconductor parameter analyzer (4200-SCS, Keithley Instruments Inc., USA). To prepare the device, the samples were gently rinsed with deionized water and allowed to dry naturally. Then, the silver paste was deposited on the DNA crystal surface to define the metal contact with a channel length of ~ 1 mm.

Optical Measurement. Optical transmission measurements in the near-infrared (NIR), visible (Vis), and ultraviolet (UV) regions (3300 ~ 175 nm) were carried out with a spectrophotometer (Varian Cary 5G). The spectrophotometer is equipped with two light sources: a deuterium arc lamp (NIR and Vis) and a quartz W-halogen lamp (UV). It employs two detectors: a cooled PbS detector for the NIR region and a photomultiplier tube for the Vis and UV regions. The spectrophotometer measures the frequency-dependent light intensity passing either through the vacuum or through a sample. The present experiment used wavelengths of 1200 nm to 190 nm.

Fig. S1. Sequence map of the double-crossover (DX) tiles.

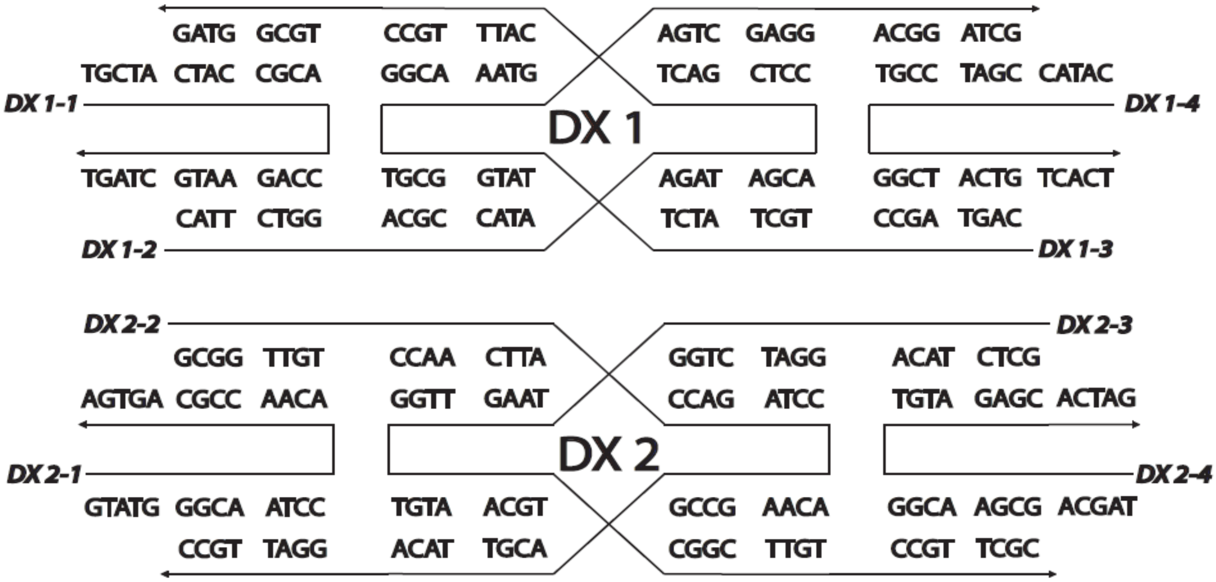

Table S1. Sequence pool for the double-crossover (DX) tiles

Strand Name	Number of Nucleotides	Sequence (5' to 3')
DX1-1	26	TGCTA CTACCGCA CCAGAATG CTAGT
DX1-2	48	CATTCTGG ACGCCATA AGATAGCA CCTCGACT CATTTGCC TGCGGTAG
DX1-3	48	CAGTAGCC TGCTATCT TATGGCGT GGCAAATG AGTCGAGG ACGGATCG
DX1-4	26	CATAC CGATCCGT GGCTACTG TCACT
DX2-1	26	GTATG GGCAATCC ACAACCGC AGTGA
DX2-2	48	GCGGTTGT CCAACTTA CCAGATCC ACAAGCCG ACGTTACA GGATTGCC
DX2-3	48	GCTCTACA GGATCTGG TAAGTTGG TGTAACGT CGGCTTGT CCGTTCGC
DX2-4	26	TAGCA GCGAACGG TGTAGAGC ACTAG

Fig. S2. Sequence map of the DX-biotin (DXB) tiles.

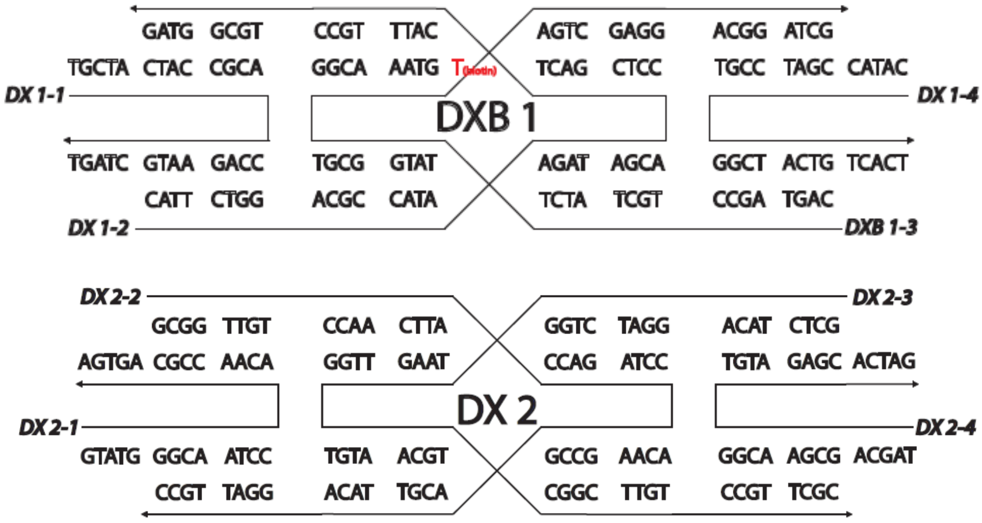

Table S2. Sequence pool for the DX-biotin (DXB) tiles.

Strand Name	Number of Nucleotides	Sequence (5’ to 3’)
DX1-1	26	TGCTA CTACCGCA CCAGAATG CTAGT
DX1-2	48	CATTCTGG ACGCCATA AGATAGCA CCTCGACT CATTTGCC TGCGGTAG
DXB1-3	48	CAGTAGCC TGCTATCT TATGGCGT GGCAAATG/ibiodT/ AGTCGAGG ACGGATCG
DX1-4	26	CATAC CGATCCGT GGCTACTG TCACT
DX2-1	26	GTATG GGCAATCC ACAACCGC AGTGA
DX2-2	48	GCGGTTGT CCAACTTA CCAGATCC ACAAGCCG ACGTTACA GGATTGCC
DX2-3	48	GCTCTACA GGATCTGG TAAGTTGG TGTAACGT CGGCTTGT CCGTTCGC
DX2-4	26	TAGCA GCGAACGG TGTAGAGC ACTAG

Calculation of reusable DNA strand amount:

- The size of the substrate: $5 \times 5\ \mathrm{mm}^2$
- The dimension of unit DX tile: 12 nm (length) × 4 nm (width)
- Effective dimension of single DX tile (due to electrostatic repulsion force among backbones): 12 nm (length) × 6 nm (width)
- One substrate has two surface sides.
- Single side of the substrate can be covered with N (length) × M (width) tiles.

  $N = 5\ \mathrm{mm}/6\ \mathrm{nm} = 5\times10^{-3} / 6\times10^{-9} = 8.33\times10^{5}$ tiles

  $M = 5\ \mathrm{mm}/12\ \mathrm{nm} = 5\times10^{-3} / 12\times10^{-9} = 4.17\times10^{5}$ tiles

- Total number of tiles for covering one side: N (length) × M (width) = 3.47×10^{11} tiles/side
- Total number of tiles for covering two sides: 2 × N (length) × M (width)

  = $2\times3.47\times10^{11}$ tiles/two sides = 6.94×10^{11} tiles/two sides = $\sim7\times10^{11}$ tiles/two sides

  = **$\sim7\times10^{11}$ tiles/substrate**

- The sample with a 10 nM concentration of DNA has the following number of DNA strands:

  = 10×10^{-9} × Avogadro # / L = $10\times10^{-9} \times 6.023\times10^{23}$ strands/L = 6.023×10^{15} strands /L

  = 6.023×10^{9} strands /μL

- Total DNA volume in single test tube: 250 μL/test tube
- Total number of DNA strands in one test tube:

$$\frac{6.023\times10^{9}\ strands}{250\ \mu L} \times \frac{250\ \mu L}{1\ test\ tube} = \frac{1.506\times10^{12}\ strands}{1\ test\ tube}$$

- A total of eight kinds of DNA strands were used for constructing DX lattices which has two types of DX tiles.

  For DX-1 tile, $\frac{1.506\times10^{12}\ tiles}{1\ test\ tube}$ exist

  For DX-2 tile, $\frac{1.506\times10^{12}\ tiles}{1\ test\ tube}$ exist

- Total number of DX tiles in a test tube (10 nM, 250 μL);

  = $2 \times 1.506\times10^{12}$ tiles/test tube = 3.012×10^{12} tiles/test tube

  = **$\sim 30\times10^{11}$ tiles in a test tube**

- Total number of times for full coverage of one substrate (both sides) with DX tile:

$$\frac{30\times10^{11}\ \mathit{tiles}}{1\ \mathit{test\ tube}}\times\frac{1\ \mathit{substrate}}{7\times10^{11}\ \mathit{tiles}}=\frac{4.29\ \mathit{substrate}}{1\ \mathit{test\ tube}}$$

⇒ 250 μL of one test tube [10 nM] can cover ~4 substrates. (~4 times available)

- Therefore ~2.3 nM of DNA are needed for full coverage on a given substrate.

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