



Supporting Information

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Supplemental Information

**DNA Tile Directed Self-assembly of Quantum Dots into Two-
dimensional Nanopatterns**

Jaswinder Sharma, Yonggang Ke, Chenxiang Lin, Rahul Chhabra, Qiangbin Wang,

Jeanette Nangreave, Yan Liu* and Hao Yan*

Department of Chemistry & The Biodesign Institute

Arizona State University

Tempe, AZ 85287, USA

E-mail: hao.yan@asu.edu; yan_liu@asu.edu

Experimental Methods

1. Assembly of DNA scaffold

All DNA strands were purchased from IDTDNA (www.idtdna.com) and purified by denaturing PAGE, except the biotin- and Cy5-labelled DNA strands were purified by HPLC. The DX-DNA arrays were assembled by mixing equimolar quantities of all the constituting strands at a concentration of 1 μM in 1 x TAE- Mg^{2+} buffer (40 mM Tris, 20 mM Acetic acid, 2 mM EDTA and 12.5 mM Magnesium acetate, pH 8.0). The mixture was cooled slowly from 90 $^{\circ}\text{C}$ to room temperature. This annealed sample was used further for TEM, AFM and confocal fluorescence imaging studies.

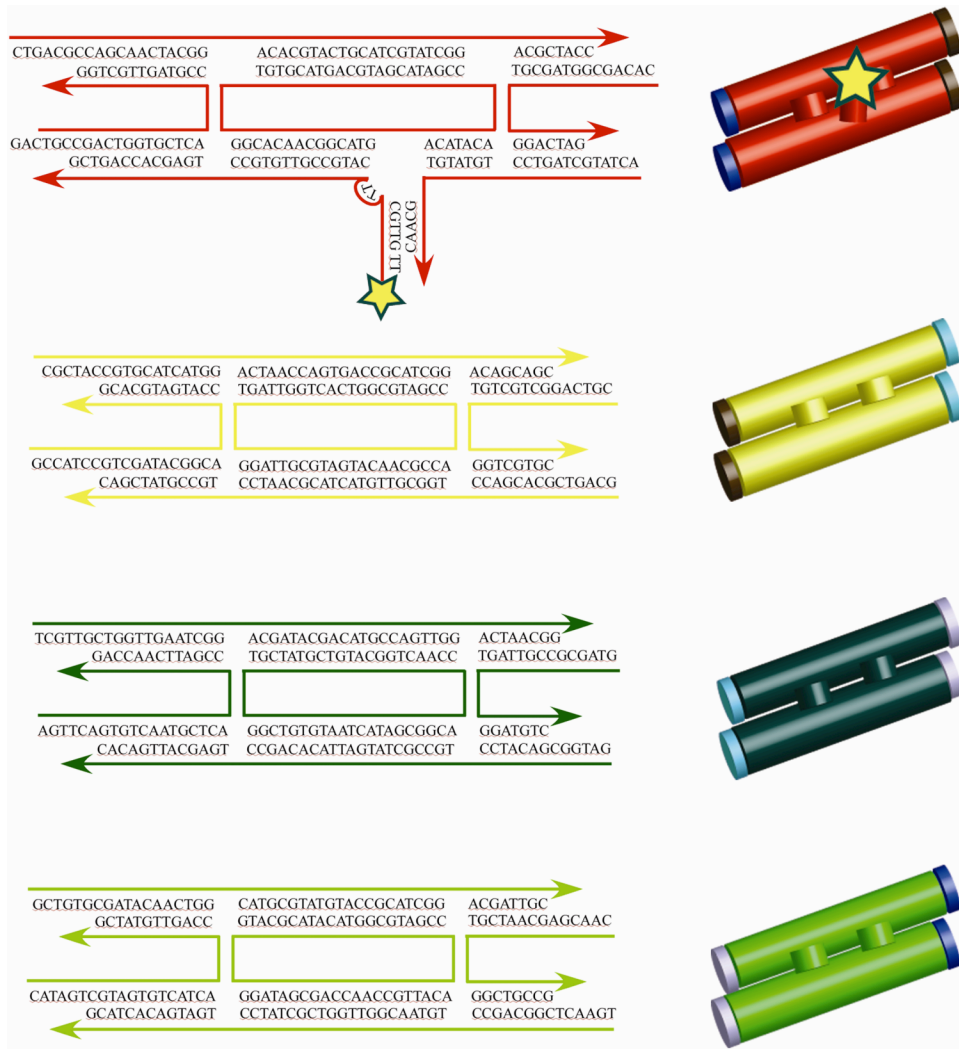


Figure S1. Sequences used in the self-assembly of DX-ABCD nanoarrays.

2. TEM studies

The TEM sample was prepared by dropping sample solution (3 μL) of biotinylated DX-DNA arrays on carbon-coated grid (400 mesh, Ted pella). Before depositing the sample, the grids were glow discharged using Emitech K100X machine. After one minute, the sample was wicked from the grid by touching its edge with a piece of filter paper. The grid was washed by touching it quickly with a drop of water and wicking out the excess water with filter paper. Streptavidin modified quantum dot solution 3 μL 0.2 μM (Invitrogen Corp., Qdot® 545 ITK™ streptavidin conjugate, CAT # Q10091MP, see <https://catalog.invitrogen.com/index.cfm?fuseaction=viewCatalog.viewProductDetails&productDescription=31566> for details of this product) was added to the grid and incubated for 10 minutes. Thereafter, excess quantum dot solution was wicked away with filter paper. To remove nonspecifically bound quantum dots from the TEM grids, the grids were washed once more with water. TEM studies were conducted by using a Philips CM12 transmission electron microscope, operated at 80 kV in the bright field mode. The high resolution TEM and EDX study, was collected using JEOL 2010F, operated at 200 kV.

3. Preparation of the sample for AFM imaging

DX-DNA arrays (15 μL , 1 μM) were mixed with 1 μL of STV-QDs (2 μM) and incubated for 30 minutes at room temperature. We added less amount of STV-QDs to < 50% saturation in the aim to avoid too many free QDs in solution. The incubated sample was then washed with 1 x TAE-Mg²⁺ buffer using Microcon centrifugal filter devices (100,000 MWCO, at 300x g speed, for 10 min) in the aim to get rid of unbound STV-QDs and loose DNA strands. After washing, the sample was reconstituted in 1 x TAE-Mg²⁺ buffer.

4. AFM imaging

DX-DNA arrays (2 μL) were deposited onto a freshly cleaved mica (Ted Pella, Inc.) and left to adsorb for 2 mins. Imaging buffer (1 x TAE-Mg²⁺, 400 μL) was added to the liquid cell and the sample was imaged in tapping mode in liquid on a Pico-Plus AFM (Molecular Imaging, Agilent Technologies) with NP-S tips (Veeco, Inc.).

5. Confocal fluorescent microscopy

Biotinylated DX-DNA arrays were assembled in the same way as regular one except that the central strand of DX-B tile was labeled by a red fluorescence dye (Cy5). 10 μL of 1 μM dye-labelled array was mixed with 1 μL of 2 μM Qdot-545 streptavidin conjugates (Invitrogen) and incubated for 10 minutes. The sample (2.5 μL) was deposited on a glass slide and immediately covered by an 18x18 mm² cover slip (the solution was spread over the entire covered area) for imaging. All the fluorescence microscope images were taken using a Zeiss® LSM 5 DUO (Carl Zeiss) laser scanning confocal microscope. The sample was scanned at a selected confocal plane and sequentially through the “green” and “red” channels. The color of the channels and the superimposed color were assigned

by a program associated with Zeiss® LSM 5 and may not reflect the true emission color of the fluorophores. A 90x90 μm^2 framed image was taken with 512x512 pixel resolution.

The set-up parameters for imaging are as described below. To image Qdot-545 (green channel), an excitation wavelength of 405 nm (Diode laser) was employed, which is reflected by a HFT 405/488/543/633 dichroic mirror and focused by an oil immersed PL APO 100.0x/1.40 DIC objective lens to irradiate the sample. The emitted photons were collected by the same objective, transmitted through the same dichroic mirror, filtered by a spectra-photometer (bandpass: 508-561 nm) and focused onto a 156 μm pinhole before reaching the detector in the green channel. For the Cy5 dye, the same set-up was utilized with changing of the excitation light source (633 nm, He/Ne laser), the spectra-photometer bandpass (657-754 nm) and pseudo-color of the channel (red).

A rectangular shape region of 11x15 μm^2 was selected from the imaged area to be photobleached. This region was constantly irradiated by a focused 405 nm laser beam at the power of 0.9 mW for 81 s. Images were taken using the same sequential scanning set-up as above with 9 s intervals during the photobleaching process. The relative intensity of red and green fluorescence within the bleached region was measured and plotted against the time course by a program associated with Zeiss® LSM 5.

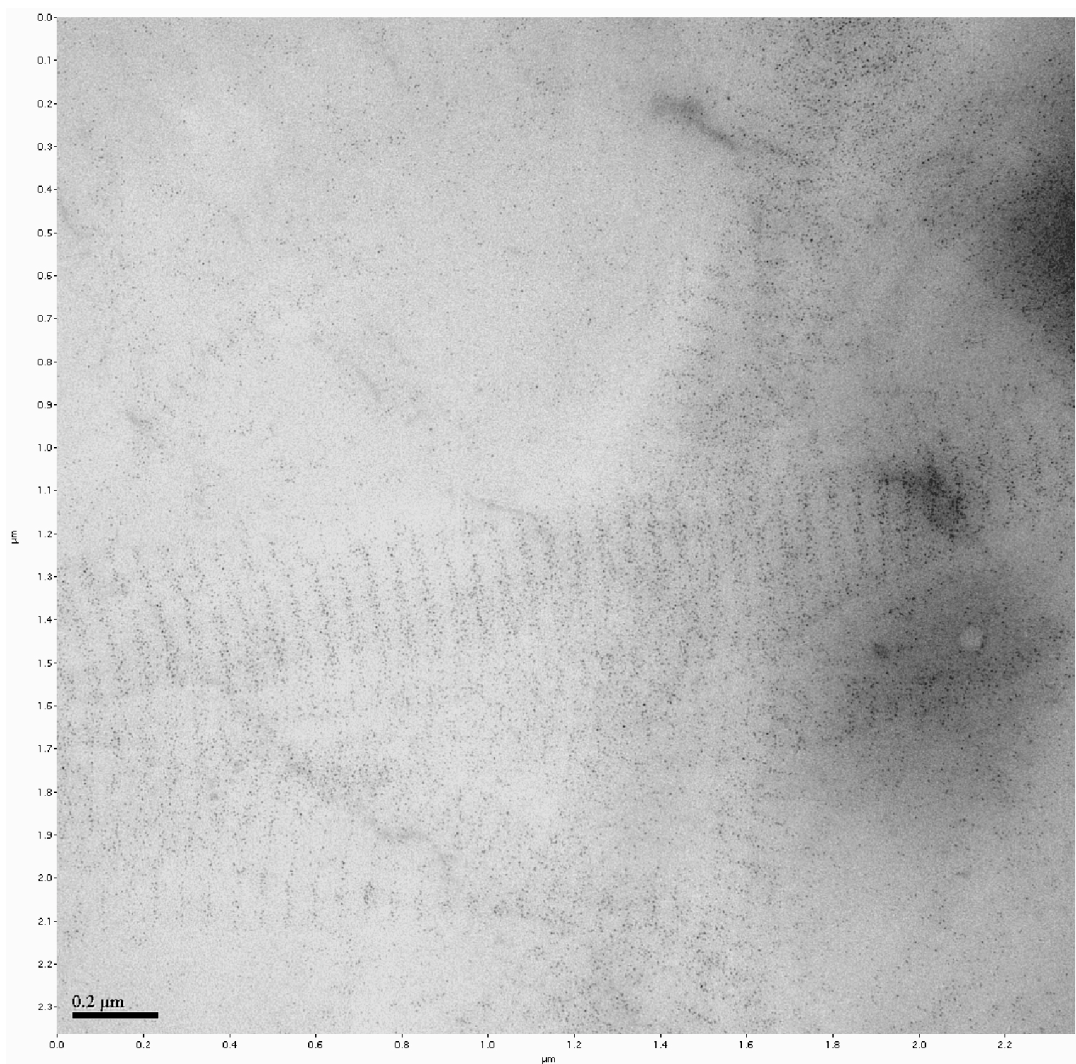
6. Further comments on important experimental conditions

Sample preparation for TEM analysis: We believe washing steps in the sample preparation play a crucial role in the patterning of quantum dots by DNA tile arrays. After deposition of DNA arrays on plasma treated TEM grids, the sample was left to adsorb for 2-3 minutes before washing off the extra salts with ultrapure water. Presence of salts may aggregate streptavidin coated quantum dots. Upon addition of quantum dots, the sample was incubated for 8-10 minutes before washing again with water. Washing step helps to get rid of free quantum dots which can hinder regular patterning of quantum dots on DNA tile arrays.

Sample preparation for AFM analysis: In AFM characterization, 10 μL DNA arrays sample was incubated with 1 μL stock solution of quantum dots for 30 minutes. Following incubation, the sample was exchanged with 1 \times TAE-Mg²⁺ buffer solution and purified using Microcon column (MWCO 100k). The purified sample was then deposited on freshly cleaved mica and scanned using AFM cantilever. Buffer exchange turns out to be a crucial step in sample preparation for AFM analysis. Efforts to pursue AFM characterization without buffer exchange led to no success of AFM imaging as DNA arrays did not adsorb onto the mica surface. We anticipate that there must be salts present in the stock solution of quantum dots that hinder the adsorption of quantum dots-DNA arrays on the mica surface. Upon buffer exchange and thus removing salts from the sample, quantum dots tagged DNA arrays could easily stick to the mica surface and scanned using AFM cantilever in tapping mode.

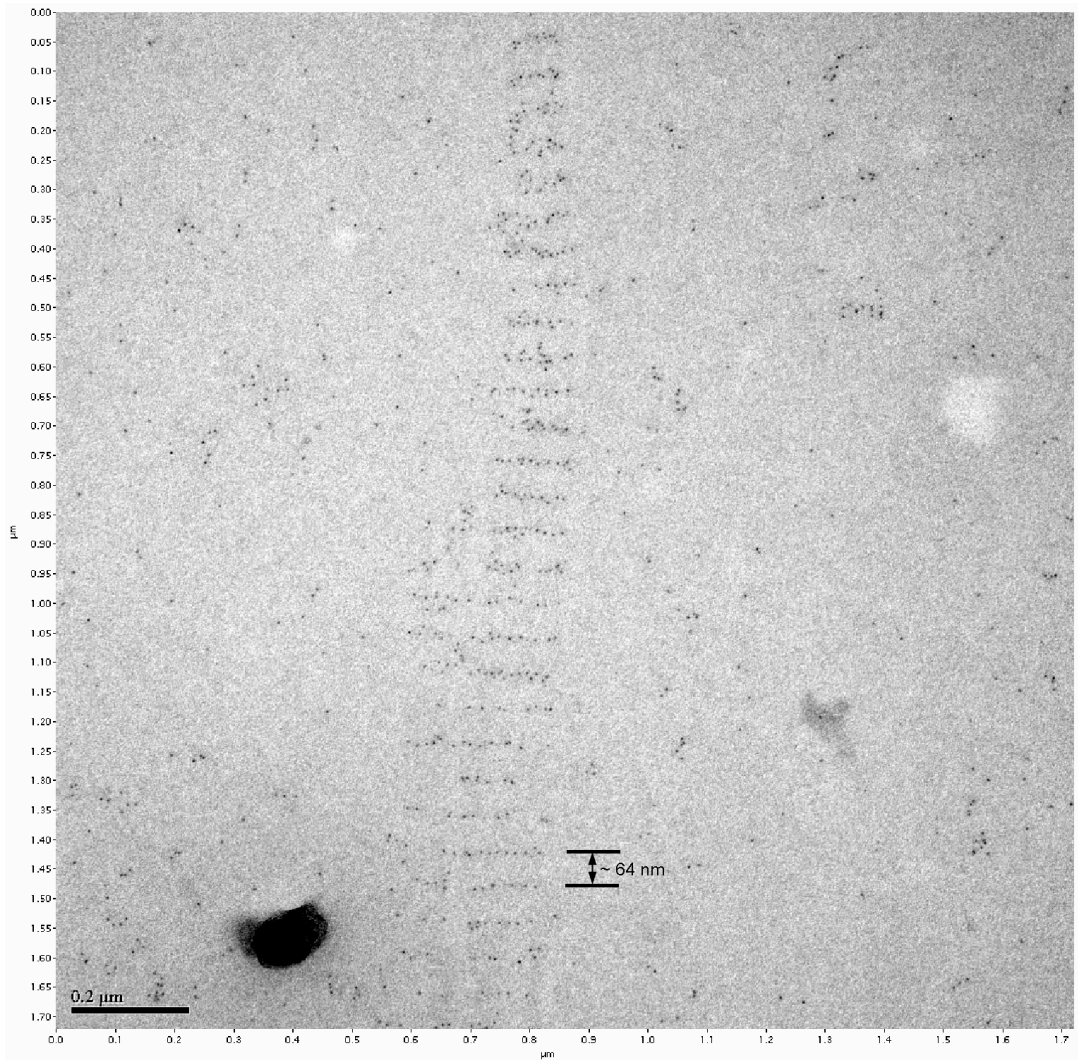
Figure S2: Additional TEM images showing alignment of quantum dots on the ABCD arrays.

a)



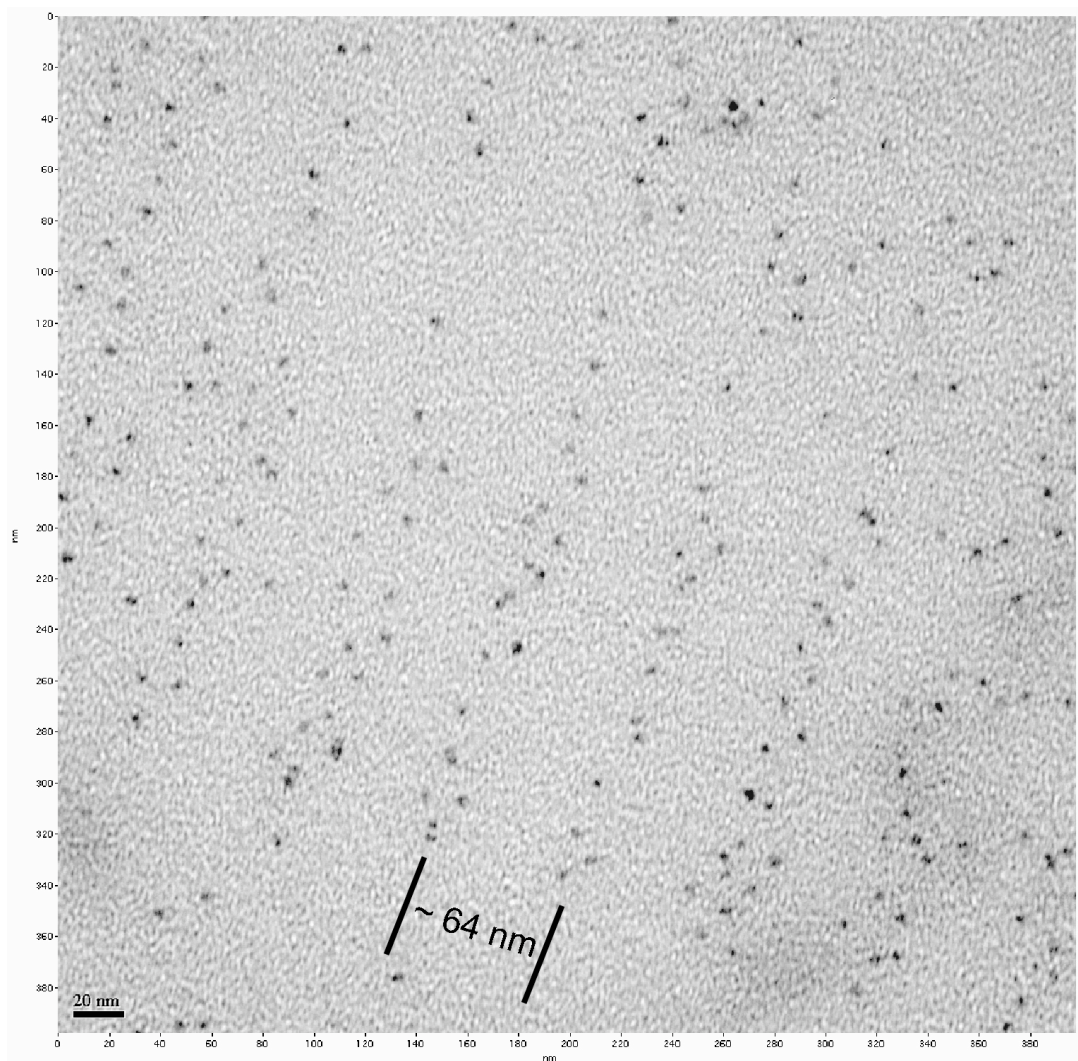
Zoom-out TEM image

b)



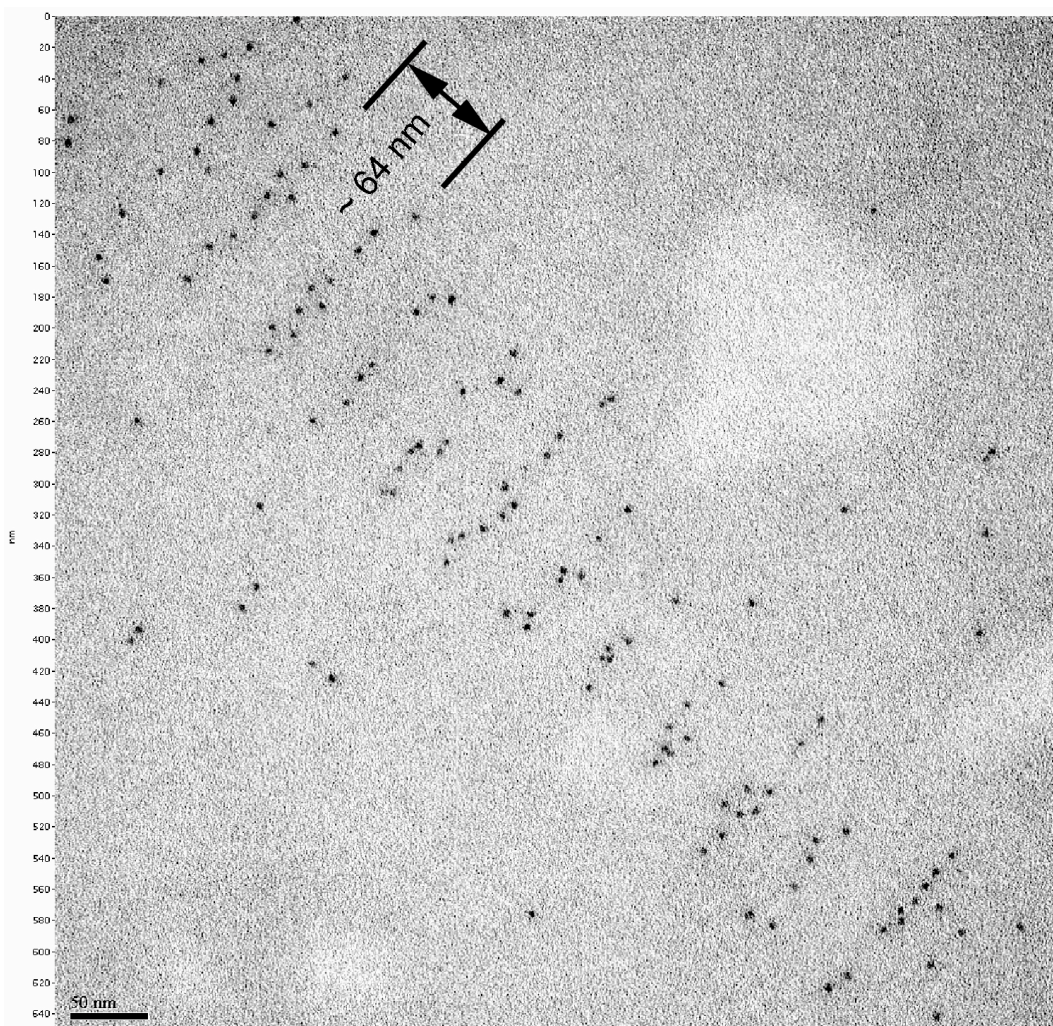
Zoom-out TEM image

c)



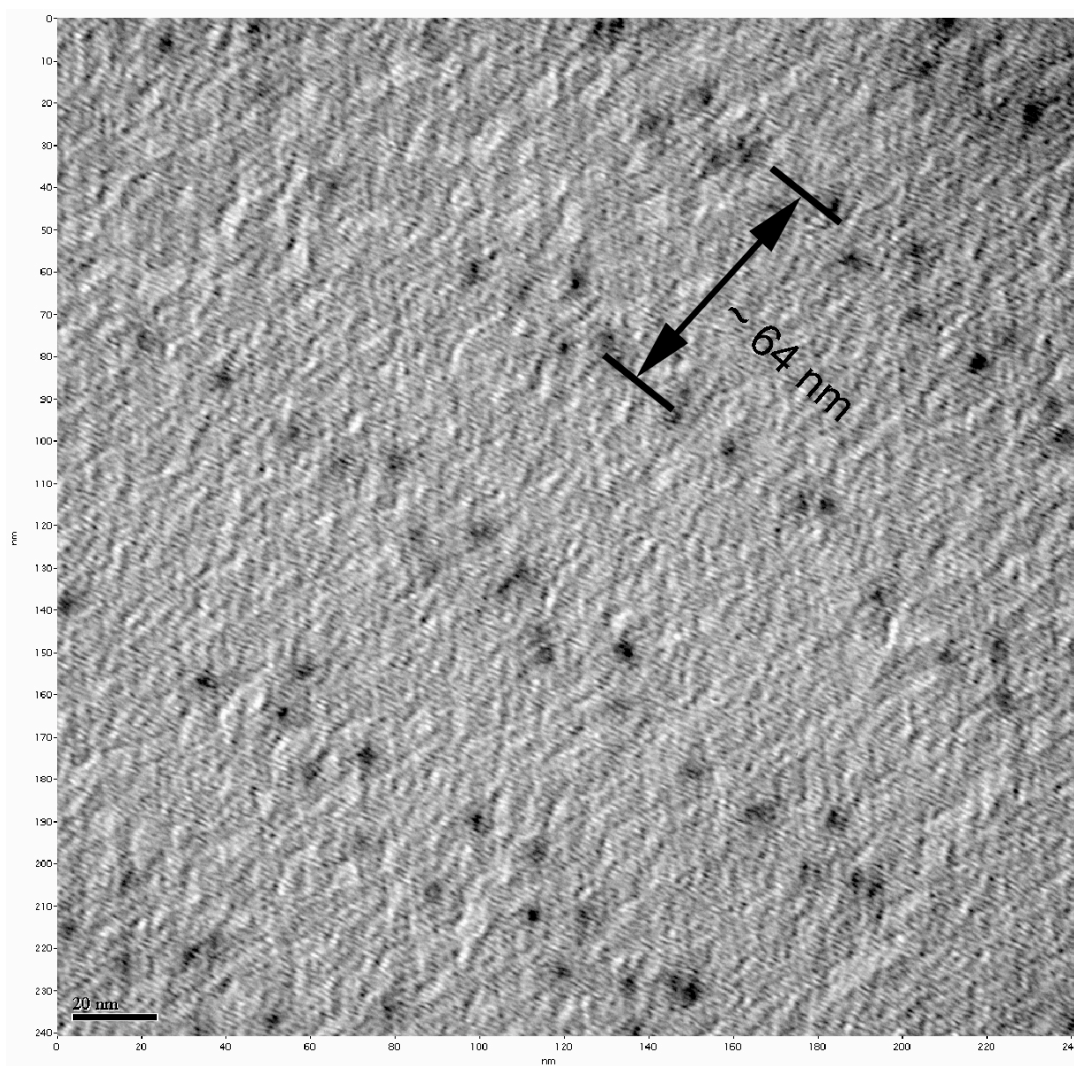
Zoom-in TEM image

d)



Zoom-in TEM image

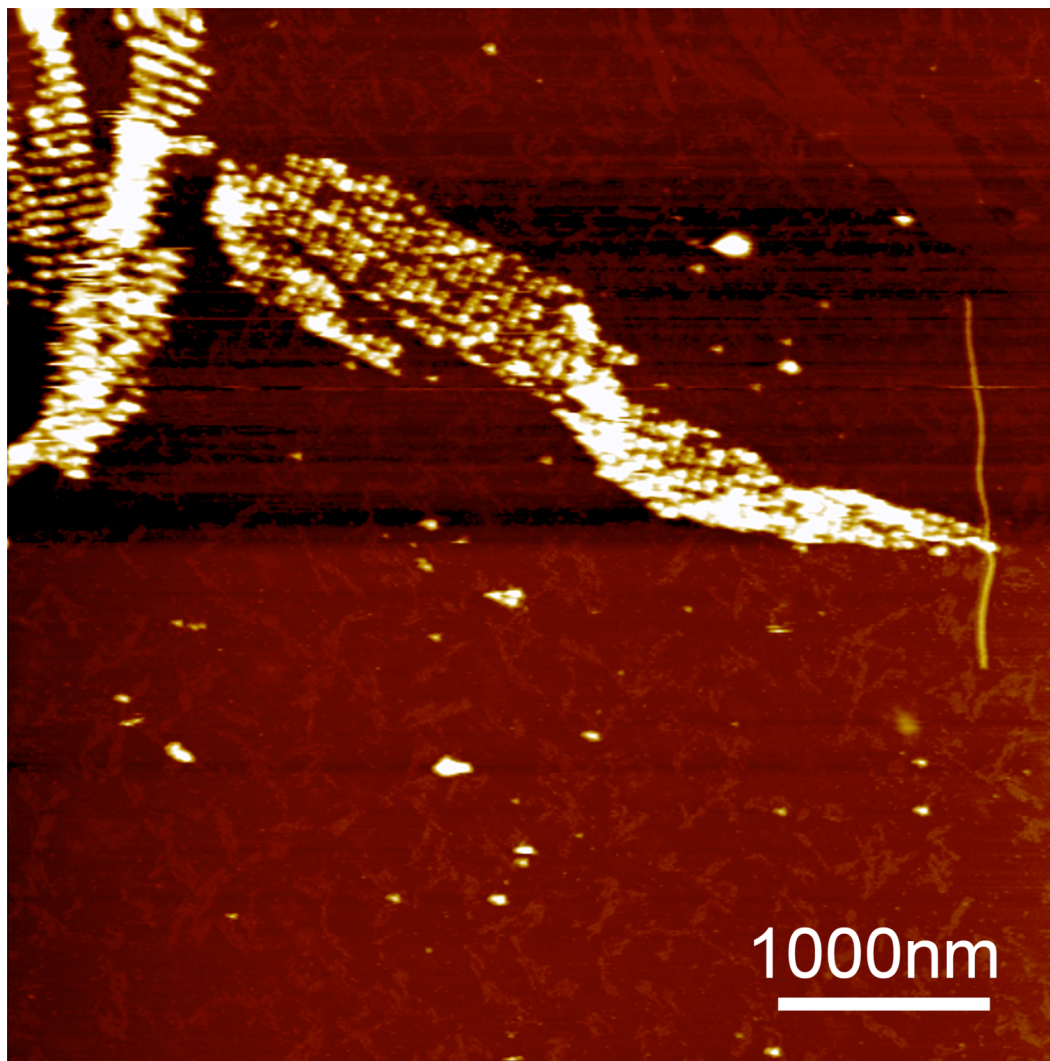
e)



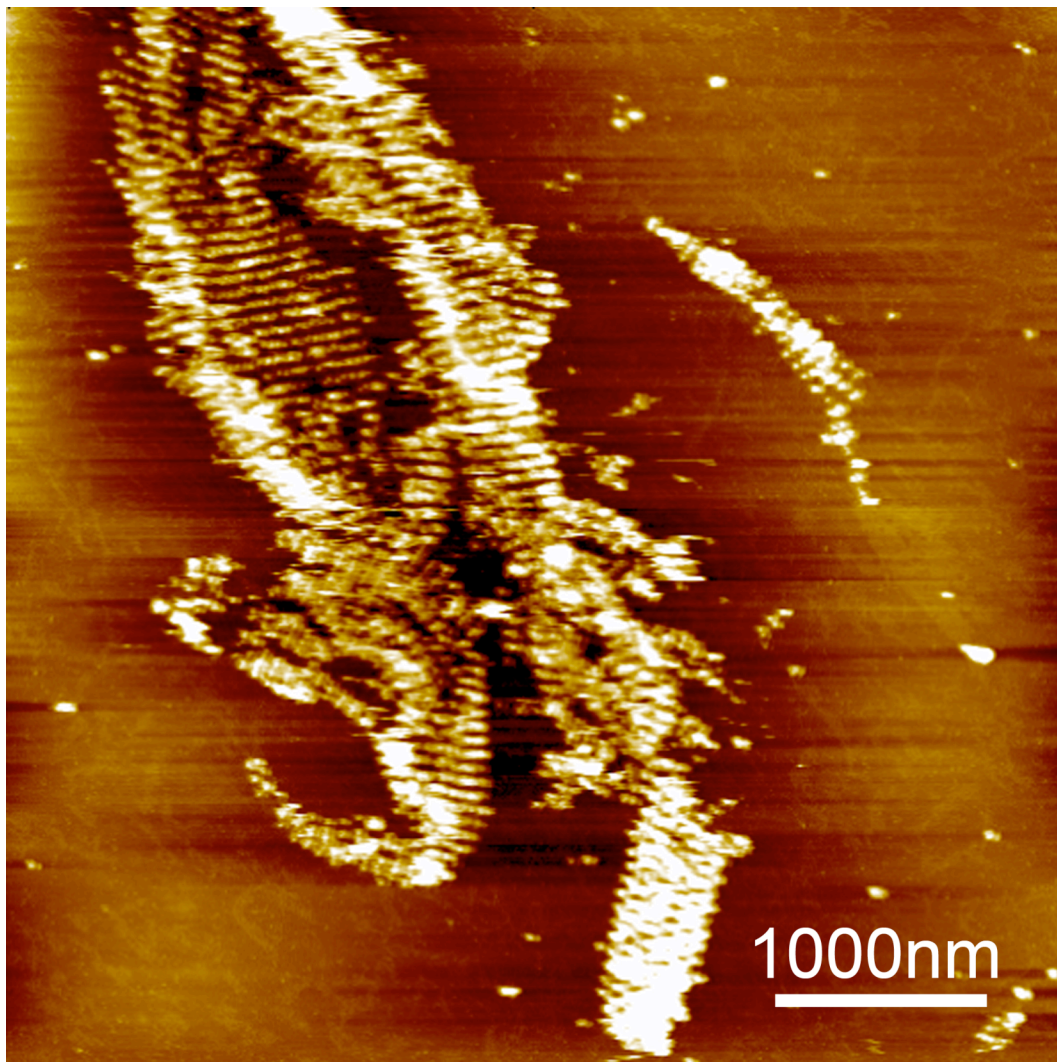
Zoom-in TEM image

Figure S3: Additional AFM images showing alignment of quantum dots on the DX-ABCD arrays.

a)



b)



c)

