Lithographically Driven Nanoscale Assembly of DNA Nanostructures

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The chemical specificity of biomolecules makes them intriguing candidates for assembly of functional nanostructures. DNA, in particular, seems well-suited to this endeavor due to its unique base pairing and to the nearly infinite combinations of nucleotide sequences. These properties confer high potential to the use of self-assembled DNA nanostructures as scaffolds for ordered assembly of functional nano-objects. Recently, there has been significant progress in this area, with solution-based assembly of metallic nanoparticles[1-3] and carbon nanotubes[4] on DNA scaffolds. In order to build functional devices from these components, however, it is first necessary to direct the assembly of DNA on macroscopic substrates, with control over location and orientation. In this work we present highly specific nanoscale assembly of DNA molecules and DNA nanostructures through biochemical functionalization of lithographically patterned surfaces.

Using electron beam and nanoimprint lithography (following a previously published, high throughput method[5]) we produce sub-10 nm metal dots arranged in multiple configurations on Si or glass substrates. We have developed techniques for the selective functionalization of these patterns - at the single nanodot level - with a variety of biomolecules. In the frame of this work, single stranded DNA is attached to the dots either by direct thiolation or through avidin/biotin recognition. Each step of the biochemical functionalization is monitored by EPI-Fluorescence Microscopy performed on biomolecules tagged with different fluorophores. Highly selective binding on the dots – with no non-specific binding - has been achieved (Fig. 1). Restriction enzymes cleavage studies confirm that single-stranded DNA attached to the dots maintains its native conformation.

We exploit Watson-Crick base pairing between complementary DNA strands to bind DNA nanostructures to lithographically patterned substrates using the functionalized dots as anchors. Triangular DNA origami (about 120 nm on a side), synthesized with an A30 strand (a single strand of DNA comprising 30 adenines) on each corner, were bound to nanodot trimers (Fig. 2) with side length matching the origami dimensions and functionalized with complementary T30 (thymine) strands. Hybridization between the complementary strands on the DNA nanostructures has been achieved, resulting in the ordered placement of the origami on the nanodot patterns, as demonstrated by Atomic Force Microscopy (AFM), both in liquid (Fig. 3) and in air (Fig. 4).

This approach can be applied to direct the self-organization of different nano-objects onto appropriately patterned and functionalized surfaces. Furthermore DNA origami can be synthesized with many single strands of DNA with different sequences in various locations. They can work as anchoring points to selectively drive the assembly of different nanocomponents (e.g. carbon nanotubes, inorganic nanorods, quantum dots, etc.) functionalized with the appropriate complementary DNA strand, achieving high precision over the relative location on the origami surface. In this way, highly complex arrangements can be created with high resolution and high throughput, opening the possibility for the realization of functional devices at molecular dimension.

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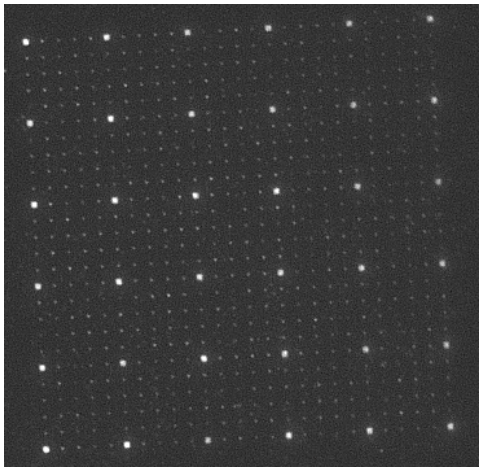

Figure 1. EPI-fluorescence image of the nanopatterned dots functionalized with a Cy3 labeled dsDNA: exitation 550nm , emission 568nm. Every fifth sub-50nm dot, a 1mm register dot is clearly visible, and brighter in the fluorescence image. Exposure time: 300ms.

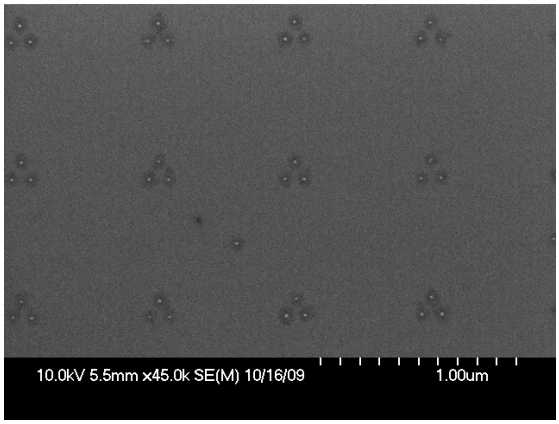

Figure 2. SEM of nanodot trimers for the binding of triangular DNA origami.

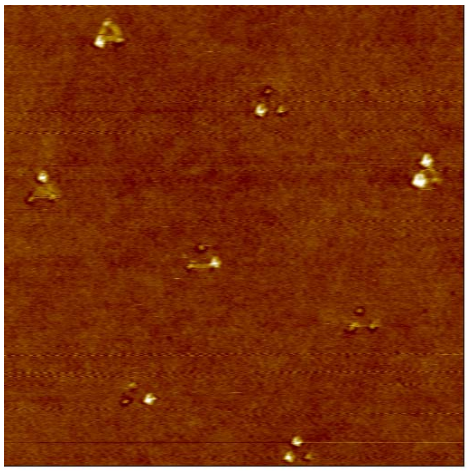

Figure 3. Liquid AFM image of DNA origami selectively bound to nanodot trimers. Image size: 1.9μm.

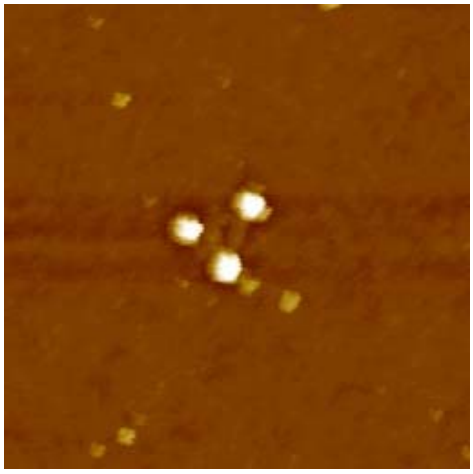

Figure 4. Dry AFM image of DNA origami on nanodot trimers. (The origami structure remains intact after drying.). Image size: 900 nm.