DNA Assembly on Patterned Surfaces

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A great deal of progress has been made since the notion of using DNA as a structural element in nanotechnology was first articulated.[1,2] DNA nanostructures can be designed to assume a variety of complex shapes, with sizes extending to a range accessible by conventional photolithography.[3] Complex DNA structures are now being explored as scaffolds for the assembly of functional nanomaterials. While solution-based assembly of metallic nanoparticles (NPs)[4,5] and carbon nanotubes (CNTs)[6] on DNA scaffolds has been demonstrated, their integration into circuits and other complex organizations requires selective placement on solid substrates with control over position and orientation. In this work we present techniques based on the combined use of lithographic patterning and bio-molecular assembly to produce highly ordered, self-assembled arrangements of nano-objects.

One focus of our work is the DNA assembly of one-dimensional (1D) structures onto lithographically patterned surfaces. We use a rigid 1D DNA motif as a model for functional materials such as CNTs and semiconducting nanorods, and study their assembly on arrays of sub-10 nm metal dots arranged in pairs, which are fabricated by electron-beam and nanoimprint lithography.[7] We have developed technique to selectively functionalize these nanodots with a variety of biomolecules.[8] In this work, short single stranded DNA (ss-DNA) oligomers are attached to the dots using a thiol-based chemistry. We designed a 60 nm-long double crossover (dx) DNA rod with a length which matches the distance between the dots and whose ends are functionalized with an complementary strand. We tag these dxDNA structures with a fluorophore, and we monitor the binding to the substrates by epifluorescence microscopy. The arrangement of the dots enables us to resolve the binding to a single nanodot-pair (Fig. 1). By varying the length of the ss-DNA, we are also able to study the effects of polyvalent binding.

A second experiment involves the lithographic placement of DNA origami, which can be used as scaffolds to organize smaller nanocomponents (e.g. NPs, CNTs, nanorods). Using nanoimprint lithography, we chemically pattern hydrophilic regions on hydrophobic substrates, producing shapes that exactly match those of the origami. A buffer solution is used in which the DNA origami are complexed with salt ions, rendering them highly hydrophilic. Figure 2 shows the selective binding of DNA origami having different shapes to Si substrates with high precision over position and orientation, as demonstrated by Atomic Force Microscopy (AFM). In addition, we synthesize origami with various anchoring points (single strands of DNA), which facilitates the attachment of multiple NPs on both sides of a single origami scaffold with precise positional control. This is possible by optimizing the hybridization conditions in order to shield the electrostatic repulsion. Different kinds of NP/DNA complexes can be selectively bound to different locations on both sides of the origami scaffold, as shown in Fig. 3.

Combining lithographic patterning with DNA-mediated assembly enables us to leverage the best features of both: positional control and chemical specificity at the molecular level. Such site specific biomolecular strategies are the most promising avenue toward complex circuits comprising functional nanomaterials.

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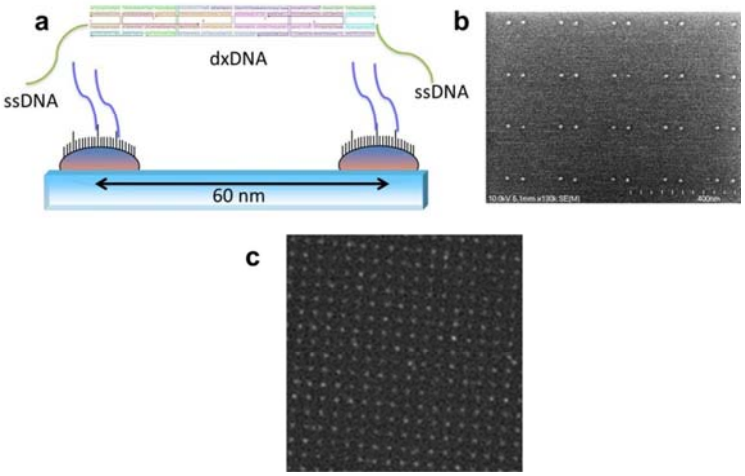

Figure 1. Binding dxDNA to nanodot dimers. (a) Binding scheme. (b) SEM of nanodot dimer array. The dot diameter is ~ 7 nm, and the intra-pair spacing is 60 nm. The distance between dimers is 200 nm. (c) Fluorescence microscopy image of dxDNA on nanodot dimer array.

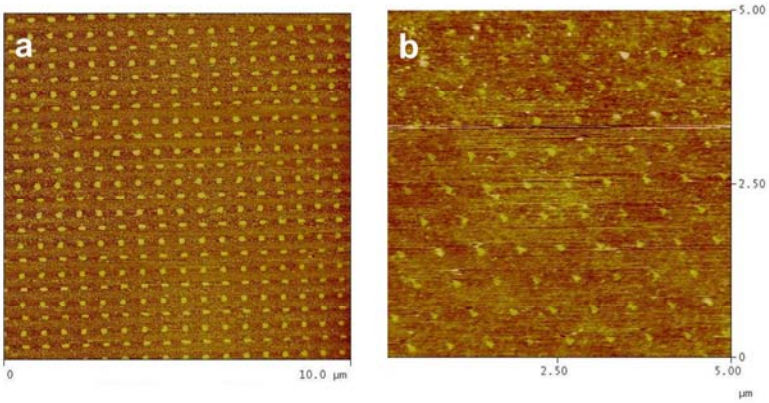

Figure 2. Liquid AFM images of DNA origami bound to NIL-defined hydrophilic shapes. (a) Origami rectangles. (b) Origami triangles.

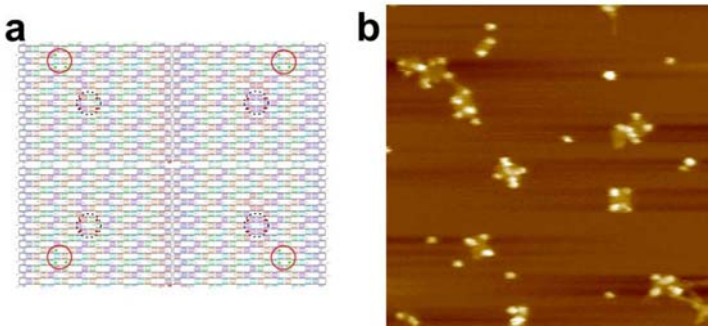

Figure 3. Au nanoparticles bound to both sides of an origami rectangle. (a) Schematic showing locations of binding anchors. Anchors on the "top" and "bottom" of the origami are indicated by solid outline circles and dashed outline circles, respectively. (b) AFM image of Au naoparticles on origami rectangles showing up to 8 NPs on a single scaffold.