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Student Paper

Atomic Force Microscopy of DNA Self-Assembled Nanostructures for Device Applications

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DNA nanotechnology, which relies on Watson-Crick hybridization, is a versatile self-assembly process whereby a variety of complex nanostructures can be fabricated with sub-lithographic features.[1] Adopting this technology, 10^{12} identical devices can be synthesized to have hundreds of components with 1nm resolution. Example nanostructures include: 1) DNA motifs [2], 2) two-dimensional DNA crystals [3], and DNA origami [4]. Currently, this technology is being adopted towards electronic, optical, and opto-electronic devices.[5]

During self-assembly, a long DNA "scaffold" strand is systematically folded into DNA origami nanostructures when hybridized with shorter DNA "staple" strands. Examples of DNA origami fabricated in this study are shown in Fig. 1. As a test vehicle to explore the viability of DNA-based electronic and optical devices, DNA-nanotubes were designed and synthesized following a similar procedure outlined by Shih *et al.* [6] and functionalized for site-specific incorporation of nanoparticles. Specifically, DNA-nanotubes were functionalized to arrange quantum dots and gold nanoparticles into linear arrays. The DNA-nanotubes, consisting of a bundle of six parallel duplex DNA strands, are an example of three-dimensional DNA origami.

With a known but pseudo-random base sequence, single-stranded DNA from the M13 bacteriophage was used as the scaffold strand. A random but known base sequence is imperative to ensure that the scaffold strand has unique, addressable oligonucliotides along its backbone. Addressable oligonucliotides: 1) prevent the formation of undesired secondary structures, and 2) promote the site-specific hybridization of designer staple strands. In this study, 170 unique staple strands were designed in-house and obtained commercially from a synthetic DNA manufacturer. When a subset of staple strands is end-functionalized with biotin, nanoparticles can be decorated at predefined locations along the length of the DNA nanotube. The resulting nanotubes then possess biotin groups to which streptavidin coated nanoparticles attach via the biotin-streptavidin conjugate. Functionalized staple strands were chosen to produce a single row of biotin binding sites with a spacing of either 15 nm or 30 nm along the DNA nanotubes.

Negatively charged DNA is electrostatically attracted to negatively charged mica surfaces in the presence of divalent cations. In addition, removing the excess staple strands and drying the sample promotes adhesion between the DNA nanotubes and the mica surface. Once the samples were prepared for characterization, atomic force microscopy (AFM) was used to image the DNA nanotubes before and after attachment of quantum dots and gold nanoparticles. Fig. 1(c) shows multiple DNA nanotubes, while Fig. 2 shows an individual DNA nanotube that has been decorated with streptavidin-coated quantum dots. The DNA nanotube of Fig. 2(a) possesses 29 biotin binding sites spaced 15 nm apart. The average number of observed quantum dots attached to the nanotubes was substantially less than 29. The DNA nanotube of Fig. 2(b) possesses 15 biotin binding sites spaced 30 nm apart. An average of 9 quantum dots were observed to attach to these DNA nanotubes.

In comparison, Fig. 3 shows an individual DNA nanotube with 29 biotin binding sites, spaced at 15 nm, to which gold nanoparticles have been attached. In this case, only three gold nanoparticles are attached, substantially less than the 29 biotin binding sites the nanotube possessed. A number of factors could account for the difference between the number of binding sites and the number of attached nanoparticles, including: (1) crowding of nanoparticles, which prevents further nanoparticle attachment; (2) hiding of biotin within the DNA origami structure of the nanotube, which prevents access of nanoparticles to biotin; and (3) poisoning of biotin binding sites by streptavidin not bound to the nanoparticles.

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Using AFM, the fabrication of DNA self-assembled nanoparticle arrays has been confirmed. The ability of AFM to determine the number and location of nanoparticles attached to DNA origami is invaluable in determining the causes of assembly defects, such as those described in this study. AFM will be an indispensable tool in transforming DNA-based self-assembly into a reliable means of fabricating nanodevices.

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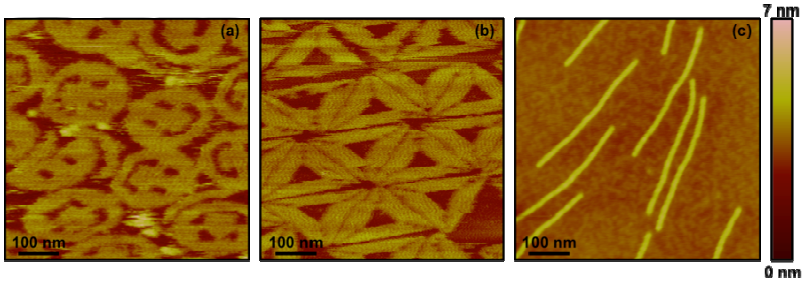

Fig. 1: AFM micrographs of DNA origami: (a) smiley faces; (b) triangles; and (c) nanotubes fabricated at Boise State University.

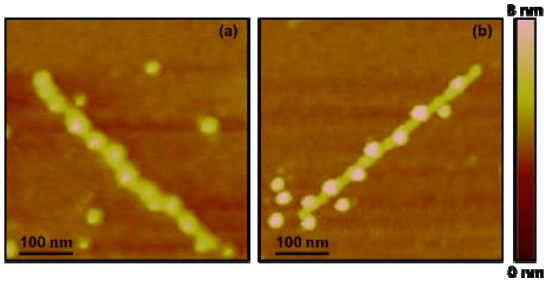

Fig. 2: AFM micrographs of DNA Origami nanotube decorated with quantum dots; In (a) the DNA nanotube possesses biotin binding sites with a 15 nm spacing; In (b) the DNA nanotube possesses biotin binding sites with a 30 nm spacing.

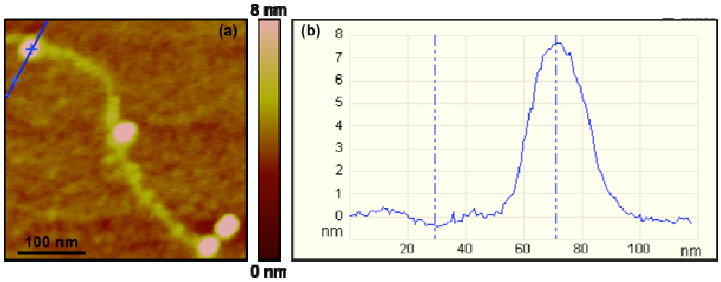

Fig. 3: (a) AFM micrograph of DNA origami nanotube decorated with gold nanoparticles; (b) AFM height profile across the gold nanoparticle along the path indicated in (a).