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Precise structure control of three-state nanomechanical DNA origami devices

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ABSTRACT

Precise structure switching between all of the three forms of three-state nanomechanical DNA origami devices has been accomplished. A nanomechanical DNA origami device called DNA origami pliers, which consists of two levers of 170-nm long, 20-nm wide, and 2-nm thick connected at a Holliday-junction fulcrum, takes three conformations: closed parallel, closed antiparallel, and open cross forms. They were previously applied to construct detection systems for biomolecules in single-molecular resolution by observing the structure switching between cross form and one of the other two forms under atomic force microscope (AFM). We redesigned DNA origami pliers in this study to let them freely switch between all of the three states including parallel–antiparallel direct switching without taking cross form. By the addition of appropriate switcher strands to the solution, hybridization and dehybridization of particular binder strands that fix the levers into predetermined state were selectively triggered as programmed in their sequence. Circuit structure switching through all of the three states in both of the two opposite direction was even successful with the new design.

1. Introduction

Rapid development of DNA nanotechnology has realized construction of various nanomechanical molecular devices that can change their shapes through specific interaction with molecules of interest in single-molecule manner [1,2]. Various excellent designs have been proposed, such as rotatory DNA nanomachines utilizing B–Z DNA transition [3], switching between PX–JX_2 parallel crossover motifs [4–7], or DNA pinching devices such as DNA tweezers [8–11] and DNA scissors [12,13].

DNA origami [14], in which long single-stranded DNA is folded into designed nanostructure with the aid of many short staple strands, opened a way to construct not only simple nanomechanical structures such as DNA origami boxes [15,16] but further complicated molecular machines with specific functions that can even be described as molecular robots [17]. The clamshell-shaped nanorobot that targets cancer cells [18] and transcription regulation system mediated by mechanical operation of tubular DNA origami [19] are typical successful examples.

We also have recently developed a nanomechanical DNA origami device (DNA origami pliers and DNA origami forceps) by joining two stick-like components of 170-nm long (levers of the pliers) at a fulcrum [20,21]. DNA origami pliers thus can take three conformations: cross form in which the two levers are not tied to each other and are in X-shape connected at the fulcrum, parallel and antiparallel forms in which two levers are aligned horizontally by the addition of the second or more bridges between the levers (Scheme 1). We applied them to construct detection systems for biomolecules in single-molecular resolution by observing the structure switching of DNA origami pliers under AFM. In the previous studies, the structure switching was only achievable between cross and one of the other two forms, although far useful systems may be accomplishable if we can directly and freely switch between all of the three forms. Nanomechanical DNA devices reported so far in fact usually take only two states; most of the cases combinations of restrained and relaxed structures, and the switching is often one way.

In this study, we designed new bridging strands between the two levers of DNA origami pliers to let them directly switch between parallel and antiparallel forms without taking cross from (Scheme 1). With the aid of precisely programmed bridging strands and switcher strands, reversible and circuit structure switching through all of the three forms was successfully accomplished.

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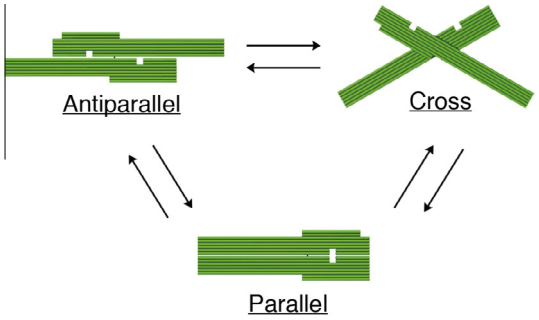


Scheme 1. Circuit structure switching between the three states of DNA origami pliers.

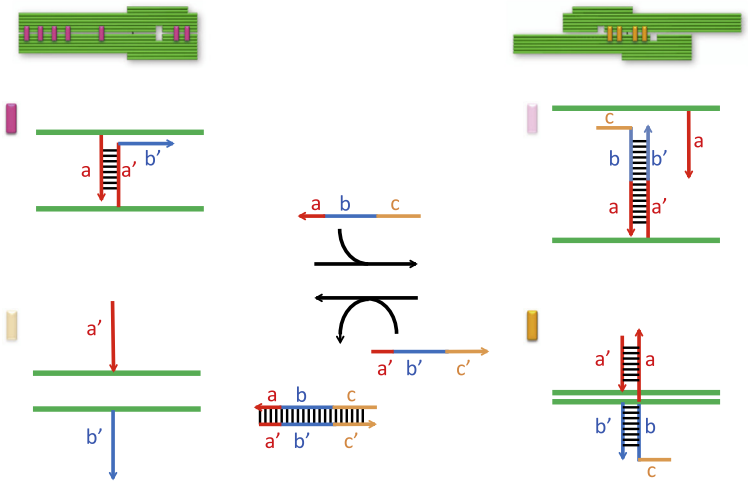
2. Material and methods

2.1. Material

Staple and switcher strands were purchased from Integrated DNA Technologies (IA, USA) under standard desalting grade and used without further purification. M13mp18 ssDNA (Takara, Japan) was used for the DNA origami scaffold.

2.2. Preparation of nanomechanical DNA origami devices

Formation of DNA origami pliers in parallel form (initial state) was performed with M13mp18 ssDNA (4 nM), staple strands



Scheme 2. Structures of the switcher and the binder strands for direct switching between parallel and antiparallel DNA origami pliers.

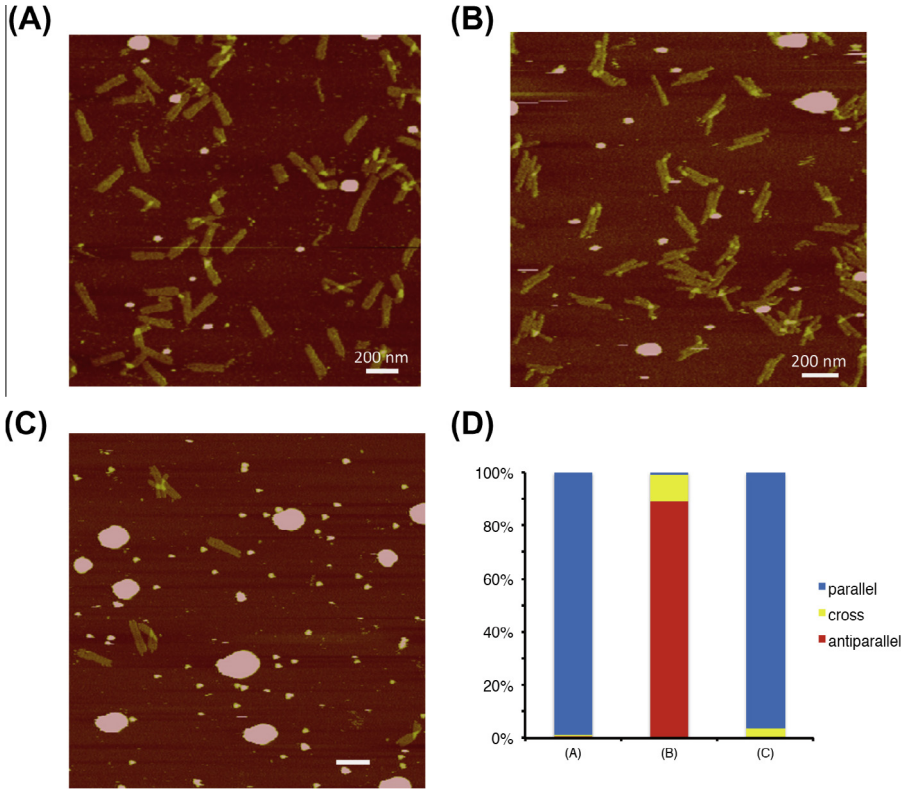


Fig. 1. Direct structure switching between parallel and antiparallel DNA origami pliers. (A) An AFM image of initial parallel DNA origami pliers deposited on mica. (B) An AFM image of DNA origami pliers after the addition of the switcher *cba* that triggers parallel to antiparallel transition. (C) An AFM image of retrieved parallel DNA origami pliers by the addition of *a′b′c′*. (D) Proportion of each form of DNA origami pliers observed in AFM images.