学位論文の要約

題目 Artificially controllable nanodevices constructed by DNA origami technology: photofunctionalization and single molecule analysis

(DNA オリガミ法を使った操作可能なナノデバイスの構築：その光機能化と一分子観察)

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General introduction: overview of DNA origami as biomaterials and applications

DNA origami was emerging as a useful tool not only for the structural nanotechnology but also exhibiting greater potential in nanomaterial science. Because of specific recognition and biocompatible property, DNA has been utilized for constructiing programmed nanostructures in 2D and 3D. Furthermore, the DNA origami can be taken as platform for the selective functionalization with small molecule complexes, biomolecules and nanoparticles. And also mechanical nanodevices based on DNA origami technique have been realized and biological-system orientated applications have been growing in great efforts.

In this thesis, it was mainly discussed about the photofunctionalization and single molecule analysis based on DNA origami methodology and high-speed AFM.

Chapter 1: Single-molecule visualization of the hybridization and dissociation of photoresponsive oligonucleotides and their reversible switching behavior in a DNA nanostructure

A single molecule observation system based on DNA frame was developed to directly visualize the hybridization and dissociation of photoresponsive oligonucleotides. A pair of pseudocomplementary oligonucleotides containing azobenzene moieties (Azo-ODNs) was introduced two parallel dsDNA separately which were located in the cavity of DNA frame. Two dsDNAs contacted at the central position (X-shape) were dissociated into parallel shape after UV irradiation. The reversible hybridization (parallel shape to X-shape) could be switched by visible light irradiation. The reversible switching behaviors were both imaged by high-speed AFM successfully.

Chapter 2: Direct observation of dual-switching behaviors corresponding to the state transition in a DNA nanoframe

A dual-switching system was constructed: a frame-shaped DNA platform with three parallel dsDNAs carrying Azo-ODNs and G-telomeric repeats together. Photoirradiation and K^+ were employed as input stimuli to regulate interactions among three dsDNAs in a logical manner. Three states: Association State-1, Relaxations State and Association State-2, were defined in correspondence with the configuration conversion in the nanoframe. Cascading transformation from

photoinduced dissociation to G-quadruplex formation in bulk solution was implanted successfully. The direct dual-switching behaviors were visualized using high-speed AFM.

The above two observation systems exhibited a general strategy for investigating various kinds of DNA-based structural changes or DNA-related switches to in single-molecule level resolution.

Chapter 3: Photo-controllable DNA origami nanostructures assembling into predesigned multiorientational Patterns

By employing Azo-ODNs as photoregulative linkers for assembling instead of for single photoreaction observation, a series of photocontrollable DNA nanostructures were assembled into predesigned patterns. A hexagonal-shaped DNA origami was employed as assembling unit tethered with Azo-ODNs. The reversible assembly and disassembly switched by UV and visible light irradiation was examined by agarose gel electrophoresis and fluorescent spectroscopy. The assembled oligomers without facing orientation were obtained in linear or curved patterns. By regulating the numbers and the positions of Azo-ODNs, the hexagonal oligomers with facing orientation could also be controlled and obtained. In this part, it was realized that the construction together with photofunctionalization in a meanwhile.

Chapter 4: Dynamic assembly/disassembly processes of photoresponsive DNA origami nanostructures directly visualized on a lipid membrane surface

Here it was shown that the direct visualization of the interactions between two DNA origami unit switched by photoirradiation using high-speed AFM. The dynamic processes of assembly/disassembly between two hexagonal-shape DNA structures were directly observed in real-time on a lipid membrane surface. The observation relies on controlled interactions between bilayer components and cholesterol modified hexagonal unit. This is the first report on the real-time imaging of nano/mesoscale interactive behaviors of DNA origami. This surface-dependent strategy should facilitate the studies of interactive and functional behaviors of various biomateirals.

Chapter 5: Programmed placement of gold nanoparticles onto a slit-type DNA origami scaffold

DNA origami method affords the possibility of positioning of functional molecules and nanomaterials precisely and selectively. Here not limited to photofunctinalization, gold nanoparticles (AuNPs) have been chosen and placed on a 2D slit-type DNA nanostructure at the predesigned position in different patterns. The arrangement of AuNPs guided by slit cavity was dependent on the position of thiolated staples. The approach presented here should facilitate the studies of interactive and functional behaviors of DNA nanostructures.

Above all, combining precise manipulation with various kinds of functionalization of different molecules, DNA origami has gradually become a useful tool for the investigation of chemical/biochemical interactions in defined nanospace.