

Chapter 2

Direct Observation of Single Hybridization and Dissociation of Photoresponsive Oligonucleotides in the Designed DNA Nanostructure

2.1 Introduction

Hybridization is the process that two complementary single-stranded nucleic acid molecules (DNA or RNA) associate together into a single double-stranded helix through specific base pairing. This process is reversible and the duplex can be controlled to dissociate by regulating the reaction conditions. To direct observation of the hybridization/dissociation reaction at single molecule level is still a challenge and important issue for the basic nanoscience and nanotechnology. Single molecules exhibit different characteristic, which is difficult to identify one by one from the ensemble state in bulk conditions. Individual molecules can behave heterogeneously, depending on the physical state of the molecules and the local environment around them [1–3].

DNA nanostructures have already been employed as scaffolds for enzyme reaction analysis and also the visualization of chemical reactions using various analysis methods [4–9]. Direct visualization of the association and dissociation of DNA duplex would also be quite fascinating. However, currently there is no direct and viable method to realize it under physical conditions. Our group have developed a single-molecule observation platform for the investigation of DNA–protein and structural DNA strand interactions in real-time using high-speed atomic force microscopy (AFM) [10–13]. In this chapter, a photocontrolled DNA nanostructure was constructed. And the dynamic hybridization and dissociation behaviors of a pair of pseudocomplementary photoresponsive oligonucleotides were tried to observe on this DNA nanostructure at single molecule level.

Azobenzene is one of photoresponsive molecules and gained popular attention of researchers because of its most intriguing property: reversible photoisomerization in a wavelength-dependent manner. Planer trans-form azobenzene molecules can isomerize to the non-planer cis-form by the photoirradiation of UV light ($330\text{ nm} \leq \lambda \leq 380\text{ nm}$) and the reverse transformation can easily switched by irradiation with visible light ($\lambda \geq 400\text{ nm}$) [14]. Asanuma and his colleagues have

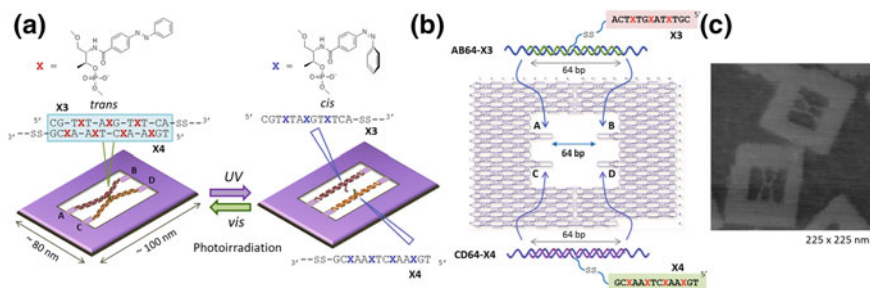


Fig. 2.1 Single-molecule observation system for the hybridization and dissociation of a photoresponsive DNA duplex containing azobenzene molecules. **a** Two dsDNAs carrying photoresponsive domains were assembled into a frame structure in X-shape because the photoresponsive domain hybridized to form a duplex in the *trans*-form of azobenzene moieties. After UV irradiation, the photoresponsive domain dissociated because of the photoisomerization from *trans*- to *cis*- form, resulting the two dsDNAs in separated-shape. **b** Two dsDNAs containing photoresponsive domain (X3 and X4) separately were located at specific sites (A–B and C–D) in the DNA frame using the corresponding ssDNAs as sticky ends. **c** AFM image of the assembled structure. [Single-Molecule Visualization of the Hybridization and Dissociation of Photoresponsive Oligonucleotides and Their Reversible Switching Behavior in a DNA Nanostructure/Endo, Masayuki; Yang, Yangyang; Suzuki, Yuki; Hidaka, Kumi; Sugiyama, Hiroshi/Angew. Chem. Int. Ed, 51/42. Copyright (c) [2012] [copyright owner as specified in the Journal]. (<http://onlinelibrary.wiley.com/doi/10.1002/anie.201205247/abstract>)

developed a series of azobenzene-tethered oligonucleotides, in which the photoresponsive performances were investigated comprehensively and also some further applications were successfully carried out [14–16]. It has already been found out that azobenzene molecules can reversibly regulate the hybridization of a double-stranded DNA by switching the photoirradiation. However, the dynamic process of single conformational changes of DNA strand has not been fully addressed yet.

A pair of pseudocomplementary DNA strands containing azobenzene moieties (photoresponsive domains) was introduced into a DNA nanoframe that has already been employed to image DNA–enzyme interactions and observe G-quadruplex formation. As shown in Fig. 2.1, the photoresponsive domain is consist of two pseudocomplementary oligonucleotides in which one contains three azobenzenes (X3) while the other is tethered with four azobenzenes (X4). They can form the duplex when azobenzene molecules are in *trans*-form, which will be induced to dissociate in the *cis*-form after the UV irradiation. Two double-stranded DNAs (dsDNAs) carrying photoresponsive domain were assembled into the cavity (approximately 40 nm × 40 nm) of DNA nanoframe. The reversible switching performances of photoresponsive domain can be identified by a global structural change of two dsDNA as an X-shape and as a separated-shape in the DNA nanoframe, respectively.

2.2 Experimental Section

2.2.1 *Synthesis of Oligonucleotides Containing Photoresponsive Domains*

The single DNA strand “AB64-3-SS” (32 m, $\sim 100 \mu\text{M}$, 20 μL) tethered with disulfide bond at the 5' terminal were mixed with 5 mM DTT and 0.1 M Tris–HCl buffer (pH 8.0). After mixing uniformly, the mixture was incubated at 37 °C for 2 h. The thiol-attached oligonucleotide (ODN-1) was purified by HPLC [linear gradient using 2–30 % acetonitrile/water (20 min) containing 20 mM ammonium formate, Nacalai Cosmosil C18 reversed-phase column (7.5 $\times$ 150 mm), 2.0 mL/min, 260 nm]. After the purification, the reduced thiol-attached oligonucleotide was directly treated with 5, 5'-dithiobis (2-nitrobenzoic acid) (DTNB) (1 μL , 50 mM DMF solution) at 40 °C for 1 h during the evaporation of HPLC solvent. The oligonucleotide–nitrobenzoic acid conjugate (ODN-2) was purified by HPLC [the same conditions as described above] (see Fig. 2.2).

The azobenzene tethered oligonucleotide (“Azo-3X-SS”, 40 μM , 20 μL), which also had the disulfide protection at the 3' terminal, was also treated with 5 mM DTT and 0.1 M Tris–HCl buffer (pH 8.0) at 37 °C for 2 h for the de-protection. The thiol-attached oligonucleotide (ODN-3) was purified by HPLC [the same conditions as described above].

Finally, the same equivalent ODN-2 and ODN-3 were mixed and put at 40 °C for 1 h under the evaporation of HPLC solvent. The concentrated mixture was analyzed and purified by HPLC [the same conditions as described above] and finally lyophilized to get the azobenzene modified oligonucleotide (AB64-X3). The final product was confirmed by denaturing polyacrylamide gel electrophoresis. The method for the coupling of another strand CD64-3-SS with Azo-4X-SS was the same as the AB64-X3.

2.2.2 *Preparation of the DNA Frame (Fig. 2.1b)*

The DNA frame structure was prepared according to the previous method. Briefly, the DNA frame was assembled in a 20 μL solution containing 10 nM M13mp18 single-stranded DNA (New England Biolabs), 50 nM staple strands (5 eq), 20 mM Tris buffer (pH 7.6), 1 mM EDTA, and 10 mM MgCl_2 as following the previous study. The mixture was annealed from 85 to 15 °C at a rate of $-1.0 \text{ }^\circ\text{C/min}$.

2.2.3 *Introduction of Two dsDNAs Containing Photoresponsive Domains into the DNA Frame*

The pre-assembled dsDNAs containing photo-responsive domains [20 nM (two equivalents)] were incorporate into the DNA frame (10 nM) by heating at 40 °C and

then cooling to 15 °C at a rate of -0.5 °C/min using a thermal cycler. The sample was purified using gel-filtration (GE sephacryl-300). The assembled structures were observed by AFM, and the yield of incorporation was counted manually.

2.2.4 Photoirradiation to the dsDNA-Attached DNA Frame

Photoirradiation for the sample was performed using Xe-lamp (300 W, Ashahi-spectra MAX-303) with band-path filter (10 nm—HW); 350 and 450 nm for the UV-light and the visible-light, respectively. The sample containing 10 nM dsDNA-attached DNA frame, 20 mM Tris-HCl (pH 7.6), 10 mM MgCl_2 was irradiated at 25 °C for 15 min for both UV- and visible-light.

2.2.5 High-Speed AFM Imaging of the dsDNAs in the DNA Frame

High-speed AFM images were obtained on a fast-scanning AFM (Nano Live Vision, RIBM, Tsukuba, Japan) using a silicon nitride cantilever (Olympus BL-AC10EGS) cooperated with light source (Olympus U-RFL-T). The sample (2 μL) was absorbed on a freshly cleaved mica plate for 5 min at room temperature, and then washed with the buffer solution for the observation of single photo-switching directly. Scanning was performed in the same buffer solution using a tapping mode.

2.3 Results and Discussion

The preparation of photoresponsive DNA nanoframe followed two steps, including (1) assembly of DNA frame as observation scaffold and (2) introduction of two dsDNAs carrying azobenzene moieties into the nanostructure. The assembled structures were confirmed by AFM (Fig. 2.1b, c). Two dsDNAs were located in the DNA nanoframe and connected at the center by the hybridization of the photoreponsive domain in X-shape.

2.3.1 Direct Imaging of the Single Photoresponsive Duplex by AFM

Figure 2.3 illustrates the molecular modeling of azobenzene modified DNA duplex. The azobenzene molecules in *trans*-form were intercalated into the duplex.

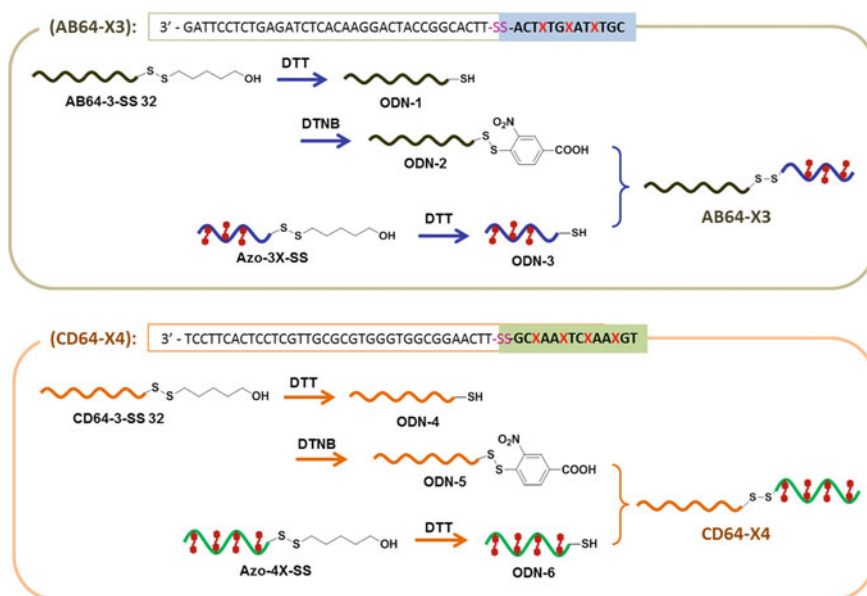


Fig. 2.2 Schematic drawings for oligonucleotides containing photoresponsive domains. The *red X* and *red hexagonal* pairs are defined as azobenzene molecules tethered with oligonucleotides. [Single-Molecule Visualization of the Hybridization and Dissociation of Photoresponsive Oligonucleotides and Their Reversible Switching Behavior in a DNA Nanostructure/Endo, Masayuki; Yang, Yangyang; Suzuki, Yuki; Hidaka, Kumi; Sugiyama, Hiroshi/Angew. Chem. Int. Ed., 51/42. Copyright (c) [2012] [copyright owner as specified in the Journal]. (<http://onlinelibrary.wiley.com/doi/10.1002/anie.201205247/abstract>)

The total length of the photoresponsive domains in duplex formation was estimated to be 10.5 nm including 17 base pairs (5.8 nm) and two organic linkers (4.7 nm). Two double stranded DNA (dsDNAs) were both 64-m (~20nm), which is fitted into the frame cavity in a tensed state with four connectors (A–B) and (C–D). Ideally they were placed parallelly in a distance of 15 nm before introducing photoresponsive domains.

After the two dsDNAs being coupled with photoresponsive domains separately, the hybridized photoresponsive domain was supported by tensed dsDNAs in the middle point as the X-shape, meaning that the photoresponsive domain was also in a relatively tensed state imaged directly by AFM. Four representative AFM images of DNA nanoframe were shown in Fig. 2.3, in which the single photoresponsive duplex on each structure was clearly identified between two dsDNAs. And also the observed lengths of hybridized photoresponsive duplex between two dsDNAs were very close to the estimated length.

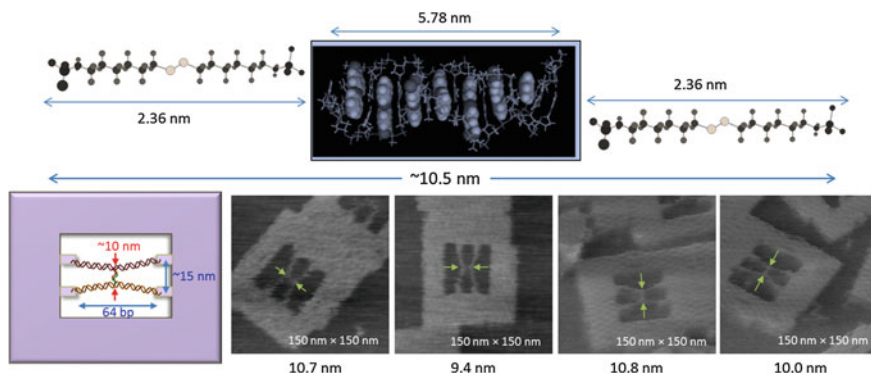


Fig. 2.3 Molecular modeling of azobenzene-tethered photoresponsive domains and AFM images of final assembled nanostructures in which the single photoresponsive duplex were clearly identified in each nanostructure

2.3.2 Photoirradiation of Oligonucleotides Containing Photoresponsive Domains

The photoisomerization performances of the photoresponsive domains were firstly examined by using two light sources that belong to (1) Xe-lamp and (2) high-speed AFM. The UV–visible spectroscopy was carried out for both UV and visible light irradiation under different time. The single-stranded strand, Azo-4X-SS, was chosen for the characterization. Firstly, the photoirradiation was carried out by using the Xe-lamp (Asahi-spectra) (Fig. 2.4a, b). Under UV irradiation (350 nm by band path filter) with different time, the absorbance of azobenzene molecules at 330 nm reduced gradually and a small increase at 440 nm occurred. After switching to visible light (450 nm by band path filter), the absorbance changed reversibly. The reversible absorbance change of azobenzene molecules indicated that Xe-lamp is able to initiate the photoisomerization of azobenzenes between *trans*- and *cis*-form. Next photoirradiation to oligonucleotide was also investigated by using another light source which is equipped together with high-speed AFM for single molecule analysis. Similar changes of the absorbance of azobenzenes were observed on UV–Vis spectras as expectedly (Fig. 2.4c, d). As a result, both of the light sources can successfully induce the photoreaction of photoresponsive domain.

2.3.3 Photoirradiation of Photoresponsive Nanoframe in Solution

The final assembled nanoframes were confirmed by AFM and ~80 % of X-shaped DNA nanoframes were obtained because of the hybridization of photoresponsive

domain containing azobenzene moieties in the *trans*-form (Fig. 2.5a). UV-light irradiation ($\lambda = 350$ nm by band path filter) from Xe-lamp was firstly introduced to nanoframe solution for 15 min at 25 °C. The irradiated sample was then imaged by AFM to confirm the structural change directly, by which separated-shaped nano-frames were observed and the distribution ratio increased from 16 to 26 % (Fig. 2.5b). The changes demonstrated that the hybridized photoresponsive domains were dissociated by the photoisomerization of azobenzenes from *trans*-form to *cis*-form. The UV-irradiated sample was further treated with visible-light ($\lambda = 450$ nm by band path filter) for 10 min at 25 °C. The ratio of X-shaped frame recovered from 64 to 74 % (Fig. 2.5c), representing that the reverse hybridization proceeded by the *cis*- to *trans*-isomerization. From the graph in Fig. 2.5d, it can be seen that the ratio variations of X-shape and separated-shape are consistent with each other during photoirradiation. The photoresponsive domain in DNA nanostructure worked correctly in solution in a wavelength-dependent manner.

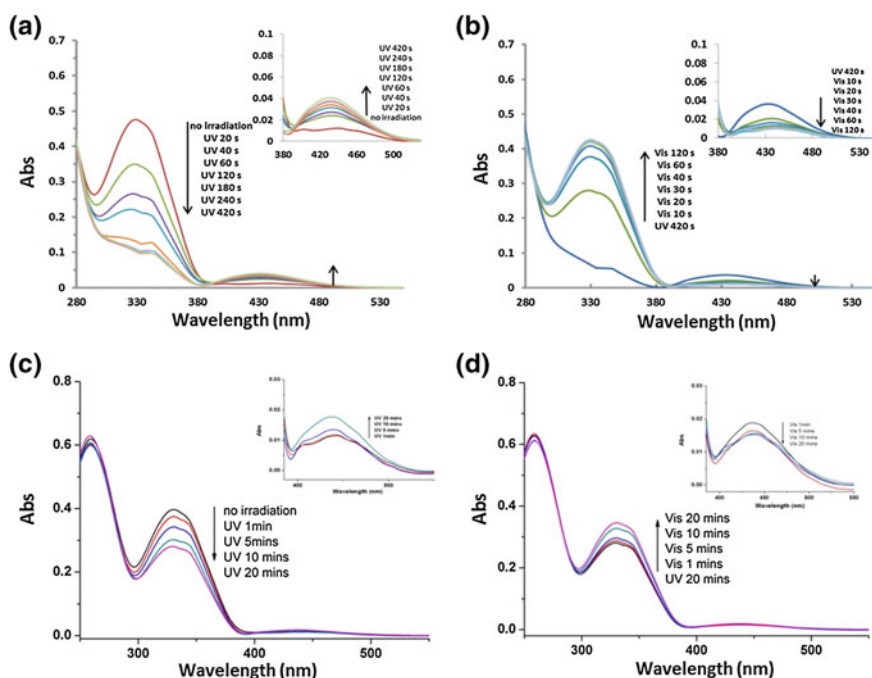


Fig. 2.4 UV-Vis spectra of unmodified Azo-4X-SS under different irradiation wavelength and irradiation time by using (1) light source of Xe-lamp (**a** and **b**) and (2) light source equipped on high-speed AFM (**c** and **d**). Conditions: 10 mM, pH 7.6 Tris-HCl buffer, UV light (350 nm) and visible-light (450 nm) by band path filter, room temperature. [Single-Molecule Visualization of the Hybridization and Dissociation of Photoresponsive Oligonucleotides and Their Reversible Switching Behavior in a DNA Nanostructure/Endo, Masayuki; Yang, Yangyang; Suzuki, Yuki; Hidaka, Kumi; Sugiyama, Hiroshi/Angew. Chem. Int. Ed, 51/42. Copyright (c) [2012] [copyright owner as specified in the Journal]. (<http://onlinelibrary.wiley.com/doi/10.1002/anie.201205247/abstract>)

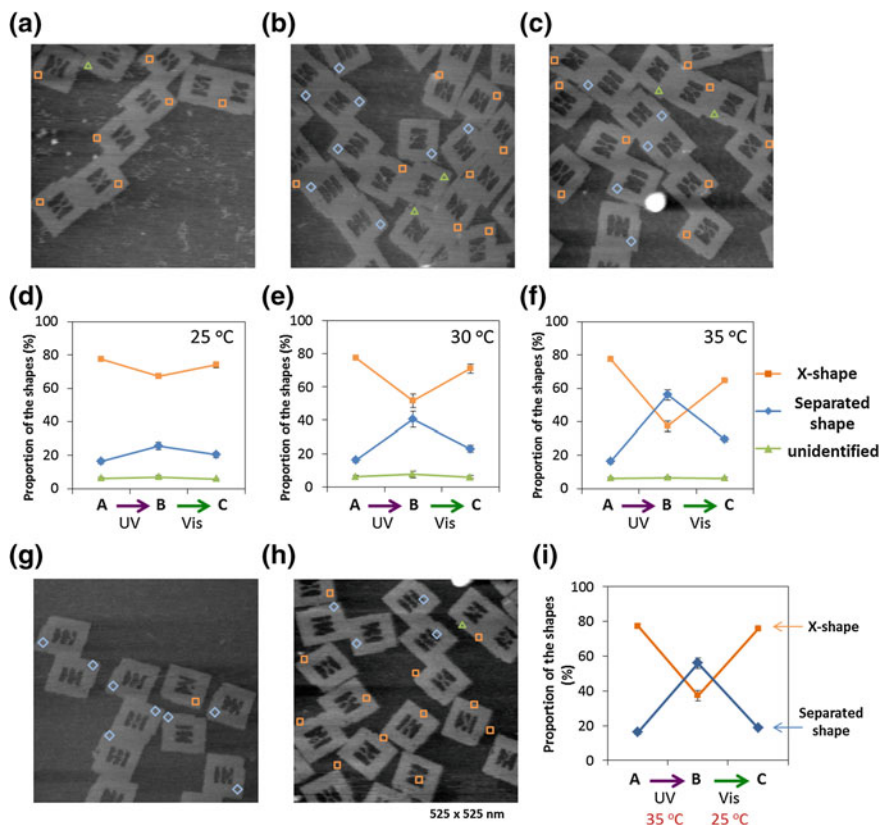


Fig. 2.5 Photoregulative dissociation and hybridization of photoresponsive domains in the DNA nanostructure. AFM images of DNA frames **a** after the assembling two dsDNAs containing pseudocomplementary strands; **b** after UV irradiation for 15 min at 25 °C; **c** after visible-light irradiation for 15 min at 25 °C of the sample from **(b)**. X-shape = orange rectangle; separated-shape = blue diamond; unidentified shapes = green triangle. **d–f** Temperature-dependent distribution ratio of X-shapes and separated-shapes after UV and visible-light irradiation at 25 °C (**d**), 30 °C (**e**), and 35 °C (**f**). A = after assembly, B = after UV irradiation, and C = after UV then visible-light irradiation. **g, h** Temperature-controlled photoirradiation for the effective dissociation and hybridization of the photoresponsive domains. AFM images after UV irradiation at 35 °C (**g**) and with subsequent visible-light irradiation at 25 °C (**h**). **i** Percentage of X-shapes and separated-shapes under the predetermined photoirradiation temperatures. [Single-Molecule Visualization of the Hybridization and Dissociation of Photoresponsive Oligonucleotides and Their Reversible Switching Behavior in a DNA Nanostructure/Endo, Masayuki; Yang, Yangyang; Suzuki, Yuki; Hidaka, Kumi; Sugiyama, Hiroshi/Angew. Chem. Int. Ed., 51/42. Copyright (c) [2012] [copyright owner as specified in the Journal]. (<http://onlinelibrary.wiley.com/doi/10.1002/anie.201205247/abstract>)

Besides the wavelength, it has already been found out that reaction temperature is also the major factor for the photoregulative hybridization and association [16]. To evaluate the temperature effect on photoresponsive domain in DNA nanoframe

during photoirradiation, three temperatures: 25, 30 and 35 °C were examined through one cycle of irradiation: UV-15 min and vis-10 min. The ratios of X-shape and separated-shape were analyzed, respectively (Fig. 2.5d–f). It can be clearly seen that higher reaction temperature exhibited higher conversion to separated-shape (56 % at 35 °C with UV light treatment), indicating that higher temperature favored the dissociation of photoresponsive domain under UV irradiation. However, it was observed that higher temperature weakened the re-formation of X-shape by the hybridization of photoresponsive domain with visible light, meaning that lower temperature is better for the hybridization of already dissociated domain. From these results, it was anticipated that controlled reaction temperature is necessary to obtain efficient dissociation and re-hybridization. (1) 35 °C for UV and (2) 25 °C for visible light were finally employed as reaction temperature for one-cycle photoirradiation. The AFM images of irradiated solution were shown in Fig. 2.5g, h respectively. From the percentage changes of X-shape and separated-shape shown in Fig. 2.5i, the UV-vis irradiated sample almost returned to the initial state completely under controlled temperature.

2.3.4 Direct Observation of Dissociation of Photoresponsive Domains in Nanoframe Under UV Irradiation

In bulk solution conditions, it was verified that this photoregulative DNA nanoframe is not only wavelength-dependent but also temperature-dependent. For single molecule analysis by high-speed AFM, constant temperature is preferable for observation. In this case, the AFM observation system was set up at 25 °C and run through whole observing process during photoirradiation, which is much lower than the melting temperature of the photoresponsive domain, to exclude the temperature effect on its dissociation.

Firstly, a freshly prepared solution of DNA nanoframe assembled with photoresponsive domain was loaded onto a clear mica surface. After clear AFM images of DNA nanoframe in X-shape were obtained, the UV light ($\lambda = 330\text{--}380\text{ nm}$) was switched by band path filter and directly introduced to the cantilever stage on high-speed AFM, which can initiate the *trans*- to *cis*-isomerization during the scanning. The time-lapsed AFM images representing the two dsDNAs changing from X-shape to separated-shape on nanoframe were shown in Fig. 2.6, indicating the dissociation of photoresponsive domain by direct UV irradiation. Interestingly, the dissociation of each single nanoframe occurred at different moments separately from 30 to 70 s. In single molecule resolution, single event of different single molecules were captured by high-speed AFM. Also, some dsDNAs on nanoframe did not show any response to photoirradiation, which may be attributed to the strong interactions between DNA molecules and mica surface, resulting the suppression of the movement of dsDNAs.

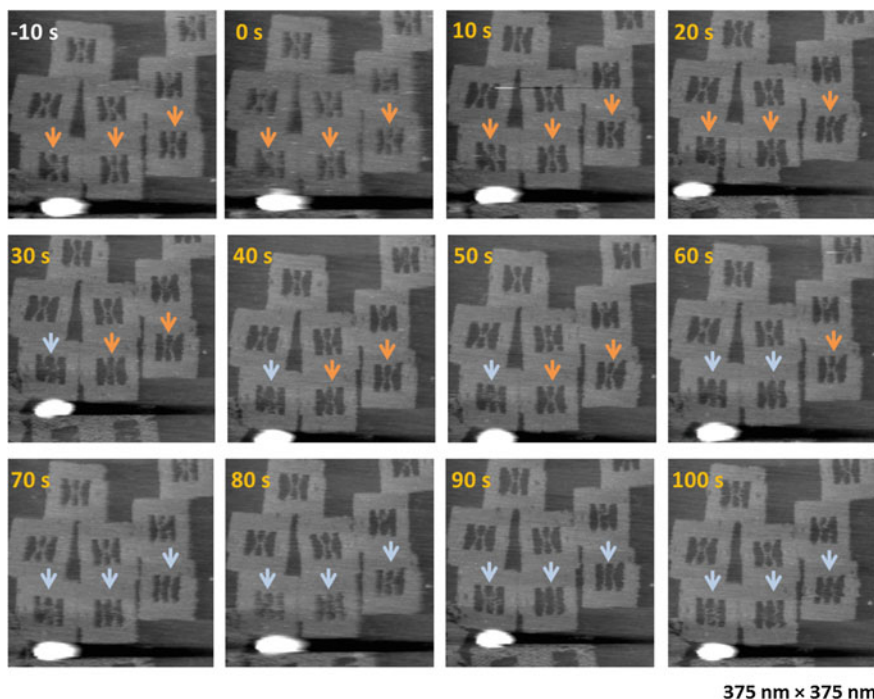


Fig. 2.6 Dissociation of the photoresponsive domain in the DNA frame during UV irradiation. AFM scanning was 0.1 frames per second. X-shape = orange arrow; separated-shape = blue arrow. [Single-Molecule Visualization of the Hybridization and Dissociation of Photoresponsive Oligonucleotides and Their Reversible Switching Behavior in a DNA Nanostructure/Endo, Masayuki; Yang, Yangyang; Suzuki, Yuki; Hidaka, Kumi; Sugiyama, Hiroshi/Angew. Chem. Int. Ed., 51/42. Copyright (c) [2012] [copyright owner as specified in the Journal]. (<http://onlinelibrary.wiley.com/doi/10.1002/anie.201205247/abstract>)

2.3.5 Direct Observation of Association of Photoresponsive Domains in Nanoframe Under Visible Light Irradiation

Under UV irradiation, the dissociation behaviors of single pair of photoresponsive domain on DNA nanoframe were directly visualized in real-time. Next it is tried to observe the reverse hybridization movements with the irradiation of visible light. The sample pretreated with UV light was loaded on the mica surface and the expected nanoframes in separated-shape were imaged. Visible light ($\lambda = 440\text{--}470\text{ nm}$) was then introduced by band path filter for the *cis*- to *trans*-isomerization of azobenzene molecules in the nanostructures. As shown in Fig. 2.7, the two separated dsDNAs were associated at the central position into X-shape because of the hybridization of photoresponsive domain (100–120 s). And also the

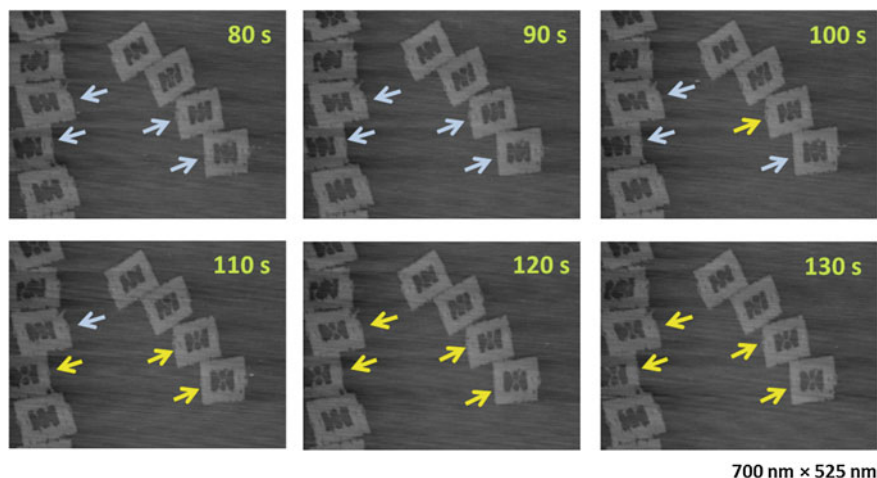


Fig. 2.7 Hybridization of the pre-UV irradiated photoresponsive domain in the DNA frame during visible-light irradiation. AFM scanning rate was 0.1 frames per second. X-shape = yellow arrow; separated-shape = blue arrow. [Single-Molecule Visualization of the Hybridization and Dissociation of Photoresponsive Oligonucleotides and Their Reversible Switching Behavior in a DNA Nanostructure/Endo, Masayuki; Yang, Yangyang; Suzuki, Yuki; Hidaka, Kumi; Sugiyama, Hiroshi/Angew. Chem. Int. Ed, 51/42. Copyright (c) [2012] [copyright owner as specified in the Journal]. (<http://onlinelibrary.wiley.com/doi/10.1002/anie.201205247/abstract>)

hybridization on different nanoframes occurred separately at different time points when observing the events at single molecule resolution (Fig. 2.8).

The photoisomerization of azobenzene as well as its derivatives is extremely rapid, occurring on picosecond timescales¹⁷, which can be ignored as compared with the reaction time-scale for the hybridization and dissociation of the azobenzene-modified oligonucleotides. Therefore, the rate determining process of dynamic interacting between two dsDNAs should be the hybridization and dissociation of photoresponsive domains. The first UV-induced dissociation occurred at 30 s while the first re-hybridization was captured at 110 s, representing that the slower hybridization of two single DNA strands requires more time for the correct contact and complete association.

2.3.6 Reversible Photoswitching of Photoresponsive Domains Repeatedly in a Single DNA Nanoframe

Besides the direct visualization of hybridization and dissociation of photoresponsive domains on DNA nanoframe separately on mica surface, the reversible activities of a single pair were also successfully obtained by alternating UV–Vis–UV directly on the AFM stage. The X-shaped and separated-shaped DNA

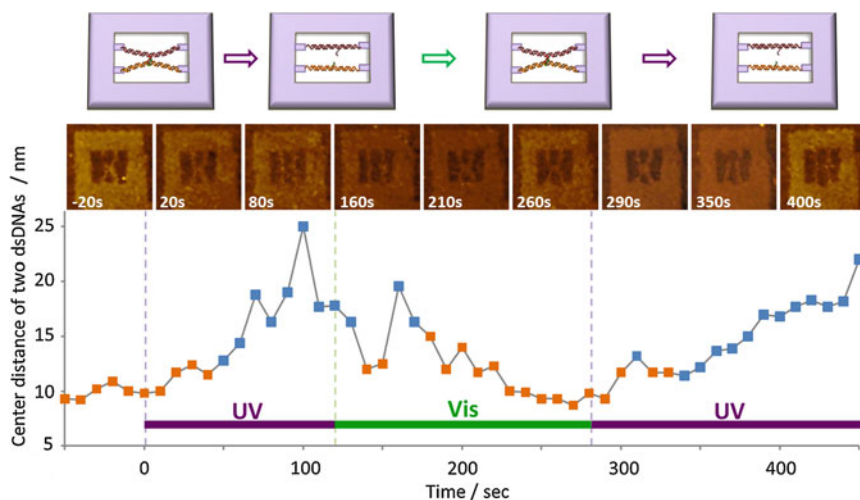


Fig. 2.8 Photoswitching activities of single pair of photoresponsive domain in the DNA frame. The closest distances between two dsDNAs were plotted by alternating photoirradiation in UV (purple) and visible-light range (green) during AFM scanning. The dissociation and hybridization of photoresponsive domains were monitored repeatedly. [Single-Molecule Visualization of the Hybridization and Dissociation of Photoresponsive Oligonucleotides and Their Reversible Switching Behavior in a DNA Nanostructure/Endo, Masayuki; Yang, Yangyang; Suzuki, Yuki; Hidaka, Kumi; Sugiyama, Hiroshi/Angew. Chem. Int. Ed. 51/42. Copyright (c) [2012] [copyright owner as specified in the Journal]. (<http://onlinelibrary.wiley.com/doi/10.1002/anie.201205247/abstract>)

nanoframe could be repeatedly observed by high-speed AFM. As shown in Fig. 2.8, here the closest distance between two dsDNAs was analyzed to determine the state of photoresponsive domains. The first dissociation occurred at 50 s after introducing UV-irradiation and after that the two dsDNAs maintained a separated shape for over 90 s. After irradiation switching from UV to visible irradiation at 120 s, a quick association was captured at 140–150 s but dissociated at the next 10 s; it was assumed that this was occurred randomly such as incomplete hybridization. The photoresponsive domain further associated stably at 180 s, indicating that one cycle of the reversible photoswitching was accomplished. Next it took 60 s (at 340 s) to dissociate again at the second round of UV irradiation. As a result, the reversible dynamic behaviors of photoresponsive domain were successfully visualized continuously from the global structural changes of dsDNAs.

Normally photo-damage is one of unavoidable issues for the photoirradiation especially in the UV range. Here, the appearances of both DNA nanoframe and the incorporated dsDNAs in the nanoframe should be maintained intact through all irradiation processes in solution as well as on the mica surface. The photoirradiation conditions employed in this study should be moderate to maintain DNA safely.

2.4 Conclusion

Based on DNA origami technique a single molecule observation nanosystem are demonstrated here to observe the hybridization and dissociation of photoresponsive oligonucleotides directly by using high-speed AFM. The dynamic behaviors of photoresponsive duplex containing azobenzene molecules were captured by recording the movement and global change of two dsDNAs in the DNA nanoframe. By switching UV and visible-light irradiation, the dissociation and hybridization of the photoresponsive domain occurred reversibly in solution as well as on flatten mica surface. DNA-based nanoscaffolds can be employed as promising observation platform for other kinds of switching behaviors in nanometer-sized resolution and also in subsecond timescale by using high-speed AFM.

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