



DNA Nanotechnology

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Amphiphilicity-driven self-assembly is a bottom-up supramolecular approach for the design of well-defined

nanostructures.^[10] Recently, DNA-based amphiphiles have

emerged as a unique building block for the creation of DNA

nanostructures.^[11] The most remarkable feature of the DNA

nanostructures obtained using this strategy is the dense

display of single-stranded DNA (ssDNA) on the surface of

the nanostructure, allowing them to act as a DNA-based

template for the organization of functional molecules through

DNA hybridization. Usually, DNA-based amphiphiles have

flexible chains as the hydrophobic part, which self-assemble

into micellar nanostructures.^[12] We have shown that the

incorporation of a large π -surface as the hydrophobic domain

drives the assembly in a lamellar fashion, leading to the formation of vesicles^[13] and nanosheets^[14] However, the

design of chiral nanostructures via DNA-based amphiphile

self-assembly is not well explored.^[15] We envisioned that the

incorporation of a hydrophobic π -surface that has a propeller

conformation (chiral geometry), such as hexaphenylbenzene

(HPB), could potentially induce a helical twist in the lamellar

organization and direct the assembly into a DNA-decorated,

chiral nanostructure, such as a helically twisted ribbon.

Derivatives of HPB have received great attention in recent

years due to the atropisomerism associated with the rotation

of $C(sp^2)-C(sp^2)$ bonds connecting the radial benzene rings with the core benzene ring.^[16] Hence, HPB can adopt two

chiral propeller conformations, which include the conforma-

tion with all radial benzene rings tilted in clockwise direction

and the other conformation with an anticlockwise tilt

(Scheme 1). Moreover, one of the chiral propeller conforma-

tions can be favored in the self-assembled state by the

incorporation of a chiral moiety on HPB due to the chirality

transfer.^[17] This has been explored for the design of chiral

nanostructures of HPB.^[18] Herein, the design and synthesis of DNA-based amphiphiles derived from the hybrid of HPB and

ssDNA is reported, and their self-assembly into helically

twisted ribbons is demonstrated. Transfer and long-range

expression of molecular chirality of ssDNA to the HPB core

in the self-assembled state bias one of the chiral propeller conformations of HPB that resulted in the exclusive formation of twisted ribbon with left-handed (M) helicity. The

potential of DNA-decorated chiral nanoribbon as a reversible

template for the construction of 1D chiral plasmonic nano-

phosphoramidite chemistry. Details of the synthesis of 2,^[19]

DNA1, and DNA2 are provided in the Supporting Informa-

tion. Both DNA1 and DNA2 have the same hydrophobic

HPB segment conjugated to a 9-mer DNA (5'-

Amphiphiles (DNA1 and DNA2) were synthesized using

structures is also demonstrated (Scheme 1).

DNA-Decorated, Helically Twisted Nanoribbon: A Scaffold for the Fabrication of One-Dimensional, Chiral, Plasmonic Nanostructures

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Abstract: Crafting of chiral plasmonic nanostructures is extremely important and challenging. DNA-directed organization of nanoparticle on a chiral template is the most appealing strategy for this purpose. Herein, we report a supramolecular approach for the design of DNA-decorated, helically twisted nanoribbons through the amphiphilicity-driven self-assembly of a new class of amphiphiles derived from DNA and hexaphenylbenzene (HPB). The ribbons are self-assembled in a lamellar fashion through the hydrophobic interactions of HPB. The transfer of molecular chirality of ssDNA into the HPB core results in the bias of one of the chiral propeller conformations for HPB and induces a helical twist into the lamellar packing, and leads to the formation of DNAwrapped nanoribbons with M-helicity. The potential of the ribbon to act as a reversible template for the 1D chiral organization of plasmonic nanomaterials through DNA hybridization is demonstrated.

he creation of chiral plasmonic nanostructures is extremely important due to their potential applications in areas ranging from material science^[1] to medicine^[2] to nanotechnology.^[3] The bottom-up approach using directed self-assembly of nanoparticles on a chiral template is the most appealing strategy for this purpose.^[4] Different nanostructures derived from the self-assembly of DNA,^[5] peptide,^[6] and small molecules^[7] have been efficiently applied as templates for the chiral organization of plasmonic nanomaterials, and undoubtedly DNA-based templates are the most powerful and viable among them. This is because DNA nanostructures offer the unique opportunity of DNA-directed spatial addressability, which permits the design of extremely complex plasmonic nanostructures that are otherwise difficult to achieve.^[8] Nanostructures of DNA are typically designed using the principles of DNA nanotechnology.^[9] Though this strategy allows the design of chiral templates of any geometry, scalability and the complex design principles involved are two major concerns. Hence, the development of a simple, yet efficient, bottom-up strategy for the design of DNA-based chiral templates is highly demanding.

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Scheme 1. Synthesis of DNA-based amphiphiles through phosphoramidite chemistry. Conditions: a) CH_2CI_2 , diisopropylamine, 2-cyanoethyl N,N-diisopropylchlorophosphoramidite, RT, 2 h, 80%. Schematic for the self-assembly of DNA-based amphiphiles into DNA-decorated, twisted nanoribbons with *M*-helicity. DNA-directed chiral organization of AuNPs and AuNRs is also shown.

TCGCACCCA-3' for DNA1 and 5'-TCTACATTA-3' for DNA2) and the only structural difference between them is the difference in the sequence of DNA (Scheme 1). Selfassembly was achieved by annealing the amphiphile $(1 \mu M)$ in Tris-HCl buffer (50 mm, pH 7.4) containing NaCl (1 mm) at 90°C for 5 min and allowing to cool to room temperature. Denaturing PAGE (20%) analyses of DNA1 and DNA2 show no migration, suggesting the spontaneous formation of DNA nanostructures with a dense display of ssDNA on their surface (Supporting Information, Figure S3). The UV/Vis spectra of DNA1 (Figure 1b inset) and DNA2 (Supporting Information, Figure S4a) at 20°C show the characteristic absorption of DNA at 260 nm and HPB at 275 nm. Interestingly, no significant change in absorption was observed upon increasing the temperature from 20°C to 70°C, suggesting that the monomeric species formed at high temperature have a similar electronic behavior as the aggregated state.

At 10 °C, **DNA1** aggregates showed an induced circular dichroism (ICD) signal in the absorption region corresponding to HPB. An ICD couplet with a first positive Cotton effect at 284 nm, followed by a negative Cotton effect at 230 nm with zero crossing at 267 nm was observed (Figure 1a). Temperature-dependent CD studies showed a gradual disappearance of the ICD signal upon increasing the temper-



Figure 1. Temperature-dependent a) CD and b) emission spectra of **DNA1**. Inset of emission spectrum shows the corresponding changes in absorption spectrum. c) Zoomed-out and d) zoomed-in TEM, e) AFM, and f) SEM images of **DNA1**.

ature from 10°C to 90°C and its complete disappearance at 90 °C. Importantly, a CD signal observed at 90 °C matches the inherent CD signal of the ssDNA segment of DNA1. These observations suggest that the transfer of molecular chirality of ssDNA into the HPB core results in a bias towards one of the chiral propeller conformations of HPB^[20] and leads to the formation of optically active aggregates, which, upon an increase temperature, disassemble into the corresponding monomeric species. Similar chiroptical behaviors were observed for DNA2 (Supporting Information, Figure S4b), which implies that the sequence of DNA has no effect on the self-assembly and chirality induction. Fluorescence spectra of DNA1 and DNA2 show the characteristic emission of HPB at 334 nm with quantum yields of 0.17 and 0.19, respectively. Interestingly, a gradual decrease in emission intensity was observed for DNA1 (Figure 1b) and DNA2 (Supporting Information, Figure S5) upon increasing the temperature from 20°C to 90°C, due to the disassembly of the aggregates. These results indicate that the enhanced emission for the aggregates of DNA1 and DNA2, compared to the corresponding monomeric species, is due to the restricted rotation of intramolecular C(sp²)-C(sp²) bonds of HPB in the aggregated state.^[21] It is also important to note that no shift was observed in the absorption and emission maxima of HPB in the aggregated state when compared with the corresponding monomeric species, suggesting that the chiral tilt con-

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formation of HPB is retained and no planarization takes place in the aggregated state.

Transmission electron (TEM), atomic force (AFM), and scanning electron (SEM) microscopic analyses of DNA1 (Figure 1 c-f) and DNA2 (Supporting Information, Figures S8 and S9) revealed the formation of helically twisted nanoribbons, and most importantly the ribbons have M-helicity. However, at very few places flat ribbons were also observed, which could be attributed to the loss of helical twist under the experimental conditions of microscopic analyses.^[22] Ribbons of DNA1 were several hundred nanometers to few micrometers in length. The average helical pitch of the ribbon was 500 nm. The breadth of the ribbons was in the range of 50-100 nm. The thickness of the ribbons was found to be approximately 11 nm, which is approximately twice the molecular length (bilayer distance) of DNA1 (approximately 12 nm). In the case of **DNA2**, the length of the ribbon was also in the range of several hundred nanometers to few micrometers. The average helical pitch of the DNA2 ribbon was 450 nm. The breadth and thickness of the DNA2 ribbons were 50-100 nm and approximately 11 nm, respectively. Based on optical, chiroptical, and microscopic data, a plausible molecular model for the assembly of the amphiphiles is proposed in Scheme 1.

The potential of the ribbon to act as a chiral template for the organization of plasmonic nanomaterials through DNA hybridization was explored. Ribbons of DNA1 were used as the representative template. For this purpose, gold nanoparticles (AuNPs) with average diameters of approximately 30 nm and approximately 10 nm were synthesized and their surface was modified with ssDNA of sequence 5'-TGGGTGCGAA-3', complementary to the DNA on the surface of the ribbon.^[8c] The assembly of AuNPs on DNA1 ribbons was achieved by annealing a 1:1 molar solution of AuNPs (1 μм) and DNA1 ribbons (1 μм) from 35 °C to 4 °C in $0.5 \times$ Tris/Borate/EDTA (TBE) buffer containing 50 mM NaCl. TEM images clearly show the 1D assembly of approximately 30 nm (Figure 2a) and approximately 10 nm (Figure 2b) AuNPs along the template. Furthermore, AuNPs are organized on either faces of the ribbon in a left-handed helical manner, which demonstrates the DNA-directed assembly of AuNPs. In accordance with this, CD studies show an ICD signal in the surface plasmon frequency of AuNPs (approximately 10 nm) at 520 nm (Figure 2 c). However, the non-bisignated nature of the ICD signal suggests that no interparticle surface plasmon coupling occur in the AuNP assembly. In support of this, no shift was observed in the electronic absorption maximum for the chiral AuNP assembly when compared with the absorption maximum of the corresponding non-assembled AuNPs (Figure 2d). Notably, significant quenching (76%) of fluorescence was observed for DNA1 after the formation of the DNA1-AuNP assembly (Figure 2d, inset). This could be due to the electronic interaction between the AuNPs and the HPB stacks.^[13] Another unique feature of the chiral template is its thermo-responsive nature. Temperature-dependent CD studies show a gradual decrease in the ICD signal at 520 nm and its complete disappearance at 60 °C (Figure 2 c). Accordingly, the fluorescence of the DNA1 ribbon was almost restored



Figure 2. TEM images of **DNA1**–AuNPs assembly of a) approximately 30 nm and b) approximately 10 nm AuNPs. c) CD spectra of **DNA1**–AuNP (approximately 10 nm) assembly at 20°C and 60°C. d) Comparison of absorption spectrum of AuNPs (approximately 10 nm) alone and after their assembly onto a **DNA1** ribbon. Inset shows the corresponding changes in emission spectrum at different temperatures.

(90%) to the emission intensity of **DNA1** at 60°C (Figure 2d, inset). These results reveal that the dissociation of the AuNPs from the template with increasing temperature is due to the melting of the duplex DNA between the template and AuNPs, as well as the disassembly of the template itself.

To establish the universal nature of the template, we have also demonstrated the potential of the ribbon for the chiral assembly of gold nanorods (AuNRs). To this end, AuNRs with an average length of 40 nm and average width of 8 nm were prepared^[23] and the surface was functionalized with ssDNA complementary to the ssDNA on the surface of the ribbon.^[24] Self-assembly of AuNRs and DNA1 ribbons was achieved using the same protocol described for AuNP assembly. Interestingly, TEM images display the formation 1D organization of the AuNRs (end-to-end) preferably along the longitudinal direction of the ribbon (Figure 3a). Assembly of the rods across the longitudinal axis of the ribbon (sideby-side) is rarely seen (Figure 3b). Unlike **DNA1**-AuNPs, no clear helical organization is seen for the DNA1-AuNR assembly. This might be due to the partial straightening of the helical ribbon upon AuNR binding due to the long length of AuNRs compared to AuNPs. Accordingly, the UV/Vis absorption spectrum of the assembly shows a decrease in intensity and broadening of the longitudinal surface plasmon band of AuNR centered at 752 nm, indicating the coupling of longitudinal plasmon oscillation (Supporting Information, Figure S16a). Whereas the transverse plasmon absorption band at 521 nm remains nearly unaffected. This is a characteristic absorption behavior for the end-to-end assembly of AuNRs.^[25] As in the case of AuNPs, a significant quenching of fluorescence (72%) of **DNA1** was observed after the assembly of the AuNRs on the ribbon due to the DNA-

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Figure 3. a and b) TEM images of **DNA1**–AuNR assembly. c) CD spectra of **DNA1**–AuNR assembly at 20°C and 60°C. d) TEM image of **DNA1** ribbon after the assembly with AuNR-modified noncomplementary DNA.

mediated electronic interaction of AuNRs and HPB (Supporting Information, Figure S16b). Furthermore, the emission was almost restored (79%) to the emission intensity of DNA1 at 60°C upon increasing the temperature of the DNA1-AuNR assembly to 60°C. CD analysis of the DNA1-AuNR assembly shows a bisignated CD couplet at the longitudinal surface plasmon resonance position of AuNR (Figure 3c). A negative Cotton effect at 790 nm, followed by a positive Cotton effect at 665 nm with a zero crossing at 740 nm was observed. As observed with the DNA1-AuNP assembly, the intensity of the CD signal gradually decreased with the rise in temperature and completely disappeared at 60 °C. It is to be noted that the DNA1-AuNR assembly exhibits plasmon coupled CD signal, whereas the DNA1-AuNP assembly shows an ICD signal without interparticle plasmon coupling. This difference in chiroptical behavior can be attributed to the strong dipolar coupling of AuNRs compared to AuNPs.^[2] In order to further confirm that the assembly of the AuNRs is indeed due to sequence-specific DNA hybridization, control experiments were carried out with AuNRs functionalized with ssDNA (5'-TCTACATTA-3'), which is noncomplementary to the ssDNA on the surface of the ribbon. As expected, no assembly of AuNRs on the ribbon was observed (Figure 3d).

Our results show that DNA-based amphiphiles are a unique class of supramolecular building blocks for the design of DNA-decorated, chiral nanostructures and the selfassembly of DNA-based amphiphiles is primarily driven by hydrophobic interactions. Though the structural diversity of the nanostructures that can be achieved by the self-assembly of DNA-based amphiphiles is limited, the simplicity of the design and straightforward synthesis make this approach of interest. The exclusive formation of DNA-decorated, chiral nanostructures with one handedness would find potential applications in the fields of chiral sensing, enantioselective synthesis, and photonic materials. Moreover, this approach permits the easy integration of functional domains of interest as hydrophobic segments into the DNA nanostructures. To our knowledge, this is the first report demonstrating the design of a helically twisted ribbon with one handedness from the self-assembly of a DNA-based amphiphile that can act as a universal template for the fabrication of 1D chiral plasmonic nanomaterials.

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Conflict of interest

The authors declare no conflict of interest.

Keywords: amphiphiles · chirality · DNA nanostructures · plasmonic nanomaterials · supramolecular chemistry

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DNA-Decorated, Helically Twisted Nanoribbon: A Scaffold for the Fabrication of One-Dimensional, Chiral, Plasmonic Nanostructures



Scarlet ribbons: This work reports the amphiphilicity-driven self-assembly of DNA-hexaphenylbenzene conjugates into DNA-decorated, helically twisted





nanoribbons, which can act as a universal template for the fabrication of 1D chiral plasmonic nanomaterials.