Supporting Information

Photo-switchable patterning of gold nanoparticles along 3D DNA nanotubes

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I. Materials and reagents

Acetic acid, urea, boric acid, ethylenediaminetetraacetic acid disodium salt dihydrate (EDTA), formamide, magnesium chloride hexahydrate, StainsAll*, tris(hydroxymethyl)aminomethane (Tris), *N*,*N*,*N*',*N*'-tetramethylethylenediamine, acrylamide, bis-acrylamide,ammonium persulfate and glycerol were used as purchased from Sigma Aldrich. Vectabond[™] reagent was purchased from Vector Laboratories. 1000Å nucleoside-derivatized LCAA-CPG solid supports with loading densities of 25-40 µmol/g, reagents used for automated DNA synthesis were purchased from BioAutomation. Sephadex G-25 (super fine DNA grade) was used as purchased from Amersham Biosciences. 1 X TA-Mg²⁺ buffer was composed of 45 mM Tris, 7.6 mM MgCl₂, with pH adjusted to 7.8 using glacial acetic acid. 1 X TBE buffer was composed of 90 mM Tris and boric acid, 1.1 mM EDTA, with a pH of ~ 8. Microscopic glass coverslips (12mm) were purchased from Bragg & Co. Azobenzene phosphoramidite was purchased from Glen Research and used as received. Gold nanoparticles with a mean diameter of 5 nm and stabilized in citrate buffer, Bis(p-sulfonatophenyl)phenylphosphine dihydrate dipotassium salt (BSPP) and tris(2-carboxyethyl)phosphine (TCEP) were purchased from Sigma Aldrich. FAM and Dabcyl dually-labeled DNA cpLS, biotin-labeled DNA RS3-biotin and thiol-labeled DNA RS1-SH were purchased from Sangon Biotech Shanghai. All other regular DNA strands were purchased from Integrated DNA Technologies.

II. Instrumentation

Fluorescence gel scanning was performed on a Fujifilm FLA-9000 scanner. Fluorescence measurements were conducted on HORIBA JobinYvon[™] FluoroMax-4 spectrofluorometer. Standard solid-phase oligonucleotide synthesis was performed on BioAutomation MerMade MM6 DNA synthesizer. UV-Vis measurements were carried out on Shimadzu[™] UV-1700 PharmaSpec UV-Vis spectrophotometer. UV/vis reversible switching process was performed using A 100W Xenon Arc Lamp (Newport Model 71232) as a light source. PAGE Gel electrophoresis experiments were carried out on apolyacrylamide 20 X 20 cm Maxi Vertical electrophoresis apparatus (MV-20DSYS). Confocal fluorescence imaging was performed on

Laser Confocal Scanning Microscope (Leica TCS SP5) with magnification of 63X. Agarose gel electrophoresis was performed on Bio-Rad Mini-Sub Cell GT Cell Apparatus with a 7 X 7 cm gel tray. Melting temperature measurement was performed on HP 8453 Spectrometer with Peltier Temperature Controller 89090A. Matrix-Assisted Laser Desorption-Ionization Time-of Flight Mass Spectrometry (MALDI-TOF MS) was performed on Bruker Autoflex Mass Spectrometer.

III. DNA sequences

Table S1. Sequences of DNA strands for the construction of DNA nanotube

	Sequence $(5' \rightarrow 3')$	ε ₂₆₀
		[L/(mole ·cm)]
TL	TATTGGTTTGV*TGACCAATAACACAAATCGGV*TCAGTAATCTCTTGAAG GTAV*GGAAACGACA	710200
Т	TATTGGTTTGTGACCAATAACACAAATCGGTCAGTAATCTCTTGAAGGTA GGAAACGACA	710200
CS1	TCGGCAGACTCTACTTGGTGCAAACCAATATGTCGTTTCCGGCGGCGTTA AATAGACGCGGCCA	608300
CS2	CGGTGCATTTAGTCGTGTCGCCGATTTGTGTTATTGGTCACGCGAATCAT GCGTACTCGT	559300
CS3	CCATAGCTTTCTCAGCAGCGAAAATACCTTCAAGAGATTACTGAGTCTTG GAGTCGGATTGAGC	621800
cpLS1	AGTCTGCCGAGGATTTCCAGCTACATTCGG(FAM)TTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTT	766300
cpLS3	AAATGCACCGGGATTTCCAGCTACATTCGG(FAM)TTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTT	762800
cpLS2	TGAGAAAGCTATGGTTAGGACCTCCATCCGTTTTAGCATCACGTCACCGA TGGCCGCGTCTATT	604300

Tt	CAAACCAATATGTCGTTTCC	189800
RS1	TAACGCCGCCTTTTTTCGCTGC	185700
RS1-SH	(5'-SH)TTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTT	394300
RS2	ACTCCAAGACTTCGACACGACT	209600
RS3	ATGATTCGCGTTCACCAAGTAG	216200
RS3-	(Biotin)ACTCCAAGACTTCGACACGACT	209600
biotin		
LS2	TCGGTGACGTGAGGTCCTAA	195700
Azo-LS13	TTGAAATTCACCTGTTCCTAGCAACAGACCGCACTTTATGATAGCAA(Azo)	665600
	GC(Azo)TA(Azo)GGTTGTAG CTGGAAATCC	

* V represents terphenyl molecules

(a) General Procedures for Solid-Phase DNA Synthesis:

DNA synthesis was performed on 1 μ mole scale, starting from the required nucleotide modified 1000 Å LCAA-CPG solid support. Azobenzene phosphoramidite was site-specifically coupled onto the growing oligonucleotide chain with a prolonged coupling time of 10 min and 3 couplings. The coupling efficiency was monitored by the trityl concentration level. All sequences were fully deprotected in concentrated ammonium hydroxide at 55 °C over 8 h.

(b) Purification:

Crude DNA strands were purified on 15% polyacrylamide/8M urea polyacrylamide gels at constant current of 30 mA for 2 h (0.5 h at 250 V followed by 1.5 h at 500 V), using 1 X TBE buffer. After electrophoresis, the gels were wrapped in plastic film and placed on a fluorescent TLC plate and then illuminated with a UV lamp at 254 nm. The bands were excised quickly and the selected gel pieces were crushed and incubated in 12 mL of sterile water at 55 °C for over 12 h. DNA samples were concentrated to about 1mL, desalted using Sephadex G-25 column chromatography. Quantification is carried by UV/Vis analysis at 260 nm.

IV. Irradiation apparatus setup for the photo-isomerization experiment

UV (300 nm < λ < 400 nm) or visible light (λ > 400 nm) irradiation was performed with Newport Oriel Apex Illuminator (100 W Xe, Ozone free). UV light was obtained with a filter (50.8 X 50.8 mm Newport Color Glass Filter, FSQ-UG1, UV bandpass) and visible light was obtained with a filter (50.8 X 50.8 mm Newport Color Glass Filter, FSQ-GG420, Cut-on 420 nm). The duration of UV irradiation is 3 min while duration of visible light irradiation is 1 min.

V. Characterization of azobenzene-inserted DNA strand Azo-LS and its duplex

The successful insertion of azobenzene moieties into DNA strand was confirmed by both UV-vis absorption and high resolution MALDI-TOF mass spectrometry studies. The melting temperature T_m was determined by a thermal denaturation experiment. The photoisomerization of azobenzene from *trans* to *cis* was tested with a Newport Arc Lamp.



Figure S1. UV-vis absorbance spectra for azobenzene-inserted DNA strand Azo-LS(red curve), regular DNA strand (black curve) and 4-hydroxy azobenzene (blue curve).



Figure S2. MALDI-TOF MS analysis of purified Azo-LS. Calculated: 19929.16 g/mol; found: 19927.4074 g/mol.

Determination of the melting temperature Tm of RdsLS2Azo

Thermal denaturation experiment was used to determine the melting temperature T_m of azobenzene-inserted strand Azo-LS. 0.5 μ M Azo-LS in 1 X PBS buffer was heated at 1 °C/min from 15 °C to 80 °C with a 1 °C interval and hold for 1 min at each degree. This experiment was performed twice. The melting temperature **Tm** was calculated (first derivative) to be 45 °C as shown in Figure S3.



Figure S3. The melting curve of Azo-LS. Experiment carried out in $1 \times$ PBS buffer. Data interval: 1°C from 15 °C to 80 °C; rate: 1°C/min; hold: 1 min.

Photoswitching efficiency of Azo molecules

The fluorescence change in the steady state experiments in Figure 1d in the manuscript is used to determine the photoswitching efficiency of the azobenzene between *trans* and *cis* forms. Firstly, the fluorescence intensity of FAM in fully opened and closed state of the duplex (without azobenzene molecules) were found to be 3955410 a.u. and 1596550 a.u. respectively in Figure S4. In figure 1d in the manuscript, the average fluorescence signals of

FAM in designed HPAzo-dsLS system after UV (red dots) and Vis (green dots) light irradiation were found to be ~ 2488407 a.u. and 1548362 a.u. respectively.

Photoisomerization efficiency =
$$\frac{F_{\text{open state}} - F_{\text{closed state}}}{F_{\text{fully closed state}} - F_{\text{fully open state}}} \times 100\%$$

According to the above calculation, the photoisomerization efficiency in our design system is found to be \sim 40 % which is highly comparable with the photoisomerization efficiency of the azobenzene molecule reported in the literatures.¹



Figure S4. Fluorescence spectra of the fully opened and fully closed state of the DNA duplex (without azobenzene molecules).



Figure S5. Fluorescence emission changes of FAM in control sample cpLS2+LS2. FAM excited at 490 nm and emission collected at 520 nm.

To convert the relative fluorescence quenching into a distance change, we simply applied the idea of Förster theory to determine the distance change of the DNA building HPAzo-dsLS in response to sequential UV and Vis irradiation. The distance change between Fluorescein and Dabcyl was calculated based on the theory reported by Glossman-Mitnik and coworkers.²

The FRET efficiency between Fluorescein and Dabcyl was defined as

$$E_{fret} = \frac{R_0^6}{R_0^6 + R^6}$$

Where R_0 is the FRET radius of the FRET pair, R is the separation of the FRET pair. In this instance, R_0 for fluorescein and Dabcyl was 4.697 nm.

For a fluorophore and quencher pair, the fluorescence emission efficiency was defined as

$$E_{FE} = 1 - E_{fret}$$

$$= 1 - \frac{R_0^6}{R_0^6 + R^6}$$

$$= \frac{R^6}{R_0^6 + R^6}$$

In our design, when nanotube was in fully closed state, the separation between fluorescein and Dabcyl assumed to be 0 nm. When nanotube was in fully open state, the separation was 10.5 nm.

According to the fluorescence changes in steady state fluorescence emission experiment, R could be calculated which corresponded to the DNA building block HPAzo-dsLS stretched distance as shown in Figure S6. Those calculated distance deviated a bit from the expected theoretical value 10.5 nm because only ~ 40 % of photoisomerization efficiency in our design system.



Figure S6. Distance separation between FAM and Dabcyl in hybridized HPAzo-dsLS by alternating UV and Vis irradiation.

VI. Self-assembly of the triangular rung (R) and DNA nanotubes

The assembly of azobenzene-incorporated DNA nanotubes started with the preparation of triangular-shaped DNA rung (R) from a closed DNA template (T). The DNA template T was prepared following a previously reported procedure.³ By mixing T with equivalent amount of three complementary DNA strands (CS1, CS2 and CS3) and three short DNA strands (RS1, RS2 and RS3) sequentially, R was formed quantitatively with short sticky-end overhangs (Scheme S1).



Scheme S1.Schematic representation showing theformation of the triangular-shaped DNA rung (R) from a closed DNA template (T).

The self-assembly of DNA nanotubes was carried out according to the previously reported methods by Sleiman and coworkers.^{3–5} Typically, 77.4 pmoles of the purified T and equimolar amounts of strands CS1, CS2, CS3, RS1, RS2, RS3 and 3 μ L of 1X TA-Mg²⁺ buffer

were combined and dried completely with EppendorfTM Concentrator *Plus* and re-dissolved in 30 µL of autoclaved water. The mixture was first heated to 95 °C and then slowly cooled down to 4 °C over ~ 5 h. This would generate a triangular rung **R** (Scheme S1). Then equimolar amounts of Azo-LS13 and cpLS1, Azo-LS13 and cpLS3, LS2 and cpLS2 were also combined in 10 µL of 0.1X TA-Mg²⁺ buffer and annealed from 95 °C to 4 °C over ~ 5 h to form 3 pairs of DNA linking strands. Finally, two solutions were mixed and annealed from 60 °C to 4 °C over 3 h to form the desired DNA nanotubes.

VII. Characterization of azobenzene-inserted DNA nanotubes

(a) TEM characterization

Transmission electron microscopy (TEM) was carried out on a FEI/Philips Tecnai 12 BioTWIN Transmission Electron Microscope under vacuum with an operation mode of 120 kV. Typically 15 µL (5 µL X 3 times) of the assembled nanotube solution (consisting of 9.63 pmoles of T strand and equimolar amount of the rest strands with an overall concentration of 5.2 µM) was deposited on carbon films on 200 mesh copper grids. The drying was accelerated by Eppendorf[™] Concentrator Plus and the freshly dried sample was stained by being placed with the sample side covered on a drop of 10 µL of NanoVan[™] solution for 1 min and dried with Kimwipe tissue followed by being placed on a drop of 10 µL of deionized water for another 1 min. Then the sample was roughly dried with Kimwipe tissue and dried in Eppendorf[™] Concentrator Plus for 10 min and then moved into a desiccator prior to imaging.

(b) Confocal fluorescence imaging on coverslips

The coverslips used for confocal imaging were treated following the reported protocol by Sleiman and coworkers.⁴ The 22 X 22 mm micro cover glasses from VWRTM were immersed in Piranha solution (70%/30% concentrated sulfuric acid/ 30% H_2O_2) for at least 2 h and then rinsed with de-ionized water 3 times with sonication for 10 min each time. The coverslips were then rinsed with HPLC grade acetone 3 times with sonication for 10 min each time and then dried at 120°C to remove any trace of water. The dried coverslips were immersed in 0.5

mL of VectaBondTM dispersed in 25 mL of HPLC grade acetone for 2 min followed by being rinsed with deionized water. After drying with N₂ flow, the coverslips were immersed in 20 mL of 0.1 M sodium bicarbonate solution containing 50 mg of biotin-mPEG-SVA (average MW=5000 g/mol) in 20 mL solution for 3 h. Then the coverslips were gently rinsed with biograde water (from 1st Base Pte Ltd) 3 times and dried with N₂ flow. 50 μ L of 1 mg/mL avidin in PBS solution was dispersed in 10 ml PBS buffer and the coverslips were immersed in the avidin solution for 3 h. After that coverslips were rinsed gently with biograde water 3 times and dried under N₂ flow. The dried coverslips were placed on clean aluminum foil in a Petri dish and the biotin (on RS3 strand) anchored nanotube samples (5.2 μ M in 30 μ L) were dropped onto the coverslips and left with lid covered for 3 h.Then the coverslips were rinsed gently with biograde water and immersed in 6 mL of 1 mg/ml staining solution (V-carbozole) for 2 h. Then the coverslips were rinsed with biograde water 3 times and were ready for confocal imaging.



Figure S7. Fluorescence emission changes of FAM in control nanotube. FAM excited at 490 nm and collected at 512 nm.



Figure S8. Fluorescence gel scanning analysis of DNA nanotubes. Lane 1: Azobenzenetethered DNA nanotubes labeled with FAM and Dabcyl, but not irradiated; Lane 2: Azobenzene-tethered DNA nanotubes labeled with FAM and Dabcyl, with UV irradiation for 3 min; Lane 3: Control DNA nanotubes labeled with FAM and Dabcyl, with UV irradiation for 3 min; Lane 4: Control DNA nanotubes labeled with FAM and Dabcyl, but not irradiated. Excited at 488 nm.



(a)



(c)



(b)



Figure S9. Representative confocal fluorescence images of Azo-NTs (a) before and (b) after UV. Scale bar is 5 μ m. Representative TEM images of Azo-NTs (c) before and (d) after UV. Scale bar is 1 μ m.

VIII. Persistence length determination

Persistence length measurements were performed on the nanotube images obtained in confocal fluorescence microscopic experiments (N= 30). The wormlike chanin model was used in which the correlation between a given nanotube's contour length (L) and its average squared end-to-end distance <R²>was related to the persistence length (p) in the following equation.

$$< R^{2} > = 4pL[1 - \frac{2p}{L}(1 - e^{-L/2p})]$$

We plotted $\langle R^2 \rangle$ against *L*for AzoNTs before and after UV irradiation which fitted well to this function with p as a free parameter (**Figure S10**). We found that the resulting persistence length of linear AzoNTs was 29.5 ± 2.3 µm. while that of bending AzoNTs was 11.2 ± 3.4 µm. These results strongly suggested that the persistence lengths of AzoNTs before and after irradiation were distinguishable and induced by UV irradiation.



Figure S10. Average squared end-to-end distance $\langle R^2 \rangle$ plotted against contour length L for azobenzene-inserted DNA nanotubes (a) before and (b) after UV (300 nm $\langle \lambda \rangle$ 400 nm) irradiations. Lines are least-squares fits to the data using the 2D Kratky-Porod Model.

IX. Construction of gold nanoparticles-anchored DNA nanotubes

(a) Preparation of gold nanoparticle mono-conjugated DNA strand

The procedure for preparing DNA mono-functionalized gold nanoparticles was adapted from the protocol reported by Taton.⁶ Typically, 10 mL of citrate-stabilized gold nanoparticles with a mean diameter of 5 nm was mixed with 2 mg of Bis(psulfonatophenyl)phenylphosphine dihydrate dipotassium salt (BSPP) and shaken for over 16 h at low speed at room temperature. Then the mixture was centrifuged at ×500 g for 30 min and the supernatant was discarded and the residue precipitate was dissolved in 100 µL of 0.5 mM BSPP solution. Then 50 µL of methanol was added and mixed thoroughly and the mixture was centrifuged at ×10000 g for 30 min. Then the supernatant was discarded and the residue precipitate was dissolved in 100 µL of 0.5 mM BSPP solution and 11 µL of 5× TBE buffer was added. The BSPP-passivated gold nanoparticles were quantitated by UV absorbance at 520 nm.

Disulfide-functionalized DNA strand was first treated by tris(2-carboxyethyl)phosphine (TCEP) for 1h and then mixed with BSPP-passivated gold nanoparticles. 1M NaCl solution was added until the final concentration of NaCl reached 0.05 M. Then the mixture was shaken for 12h at low speed at room temperature.

(b) Purification of gold nanoparticle-DNA monoconjugates

The DNA mono-functionalized gold nanoparticles were separated from the bare nanoparticles by 4% agarose gel electrophoresis in 0.5 X TBE buffer at a constant voltage of 100 V for 45 min. The desired band was cut off and crushed and incubated in MilliQ water at 4 °C for 12 h. The extracted gold nanoparticle-DNA conjugates were concentrated with filter membrane with a molecular cutoff of 1000 g/mol.



Figure S11. Agarose gel electrophoresis isolation of gold nanoparticle-DNA monoconjugates. Lane 1: Isolated AuNP-RS1; Lane 2: reaction mixture; Lane 3: bare gold nanoparticles.

To confirm the band excised from the agarose gel containing only one DNA strand per AuNP, we conducted additional experiments to quantify the unlabeled DNA strand bound to gold nanoparticle by measuring the UV absorbance at 260 nm and at 520 nm. The coverage quantification was based on a UV-visible spectroscopy method reported by Hutchison and coworkers⁷ and performed on NanoDrop 1000 spectrophotometer. KCN treatment is used to decompose the purified AuNP-RS1 conjugates in order to eliminate the UV/vis absorbance at 260 nm.

$A_{260} dAuNP-RS1 = A_{260} RS1 + A_{260} dAuNP$	equation (1)
A ₂₆₀ RS1 = A ₂₆₀ dAuNP-RS1 - A ₂₆₀ dAuNP	equation (2)

Firstly, to determine the concentration of AuNP in AuNP-RS1 sample, the UV spectrum of AuNP-RS1 conjugate before KCN treatment was measured with absorbance 0.036 at 520 nm, which corresponded to 3.6×10^{-8} mol/L. Next, AuNP-RS1 was treated with potassium cyanide (KCN) solution at pH 12 for 8 h to decompose into KAu(CN)₂ and RS1 strands. The absorbance of this decomposed mixture (A_{260} dAuNP-RS1) was measured at 260 nm and is

found to be 0.023 which is contributed by both RS1 strands and the decomposed AuNPs (equation 1).

Next, to determine the absorbance of AuNP core contributed at 260 nm after KCN treatment, calibration curve of the decomposed AuNPs was obtained. Different concentrations of bare AuNP solution were treated with potassium cyanide (KCN) solution at pH 12 for 8 h to decompose into KAu(CN)₂. The absorbances of these KAu(CN)₂ at 260 nm (A_{260} dAuNP) were measured and plotted against bare AuNP concentration and a linear regression of the calibration curve was obtained (Figure S12a). From the decomposed AuNP absorbance-concentration calibration curve, the absorbance of AuNP core contributed at 260 nm:

A260_{dAuNP} = 0.0269 + (3.6×10⁻⁸)* 507500 = 0.02096

From equation 2, the absorbance contributed by RS1 at 260 nm:

 $A260_{RS1} = 0.023 - 0.02096 = 0.00204$

Meanwhile, different concentrations of RS1-SH strand were prepared and their absorbances at 260 nm were also measured and plotted against corresponding DNA concentrations (Figure S12b). A good linear regression of the calibration curve was also obtained. From the RS1 absorbance-concentration calibration curve, we could get the DNA concentration:

 $[RS1-SH] = (0.00204-0.0003063)/41250 = 4.20 \times 10^{-8} \text{ mol/L}$

Then the number of DNA strand per gold nanoparticle:

$$N = (4.20 \times 10^{-8}) / (3.6 \times 10^{-8}) = 1.17$$



Figure S12. UV/vis absorbance calibration curve at 260 nm of (a) AuNPs and (b) RS1-SH strand.

(c)Preparation of gold nanoparticle-anchored DNA nanotubes

The procedure for preparing gold nanoparticles-tethered DNA nanotubes was slightly different. First, 19.25 pmole of the purified T and equimolar amounts of the rest corresponding strands except the AuNPs-functionalized DNA strand was combined and dried completely with Eppendorf[™] Concentrator Plus and re-dissolved in 3 µL of autoclaved water. Please note, TA-Mg²⁺ buffer should be avoided because DNA-functionalized gold nanoparticles easily precipitated in Mg²⁺ buffered solutions. Then the AuNPs-functionalized DNA strand was added to the mixture and the mixture was heated to 60 °C and slowly cooled down to 4 °C over about 3 h.



Scheme S2. Schematic representation for the construction of gold nanoparticles-anchored azobenzene-inserted DNA nanotubes.

X. Characterizations of AuNP-anchored DNA nanotubes

TEM was carried out using a Philips Technai 12 operated at an acceleration voltage of 120 kV. TEM samples were deposited directly on a 200 mesh carbon coated copper grids, and allowed to adsorb for 2 min. After that, the excess solution was removed with a filter paper, followed by being placed on a drop of 10 μ L of deionized water for another 30 s. Then the sample was roughly dried with filter paper and dried in air for 30 min and then moved into a desiccator prior to imaging.



Figure S13. TEM images of gold nanoparticles-anchored azobenzene-tethered DNA nanotubes in (a) closed state and (b) open state after UV irradiation.



Figure S14. The observed (red color) and theoretical (blue color) inter-particle distance of AuNP-Azo-NTs in open and closed states.

XI. References

- 1 H. Kang, H. Liu, J. A. Phillips, Z. Cao, Y. Kim, Y. Chen, Z. Yang, J. Li and W. Tan, *Nano Lett.*, 2009, **9**, 2690–2696.
- 2 M. Alvarado-González, M. Gallo, P. Lopez-Albarran, N. Flores-Holguín and D. Glossman-Mitnik, *J. Mol. Model.*, 2012, **18**, 4113–4120.
- 3 F. a Aldaye, P. K. Lo, P. Karam, C. K. McLaughlin, G. Cosa and H. F. Sleiman, *Nat. Nanotechnol.*, 2009, **4**, 349–352.
- P. K. Lo, P. Karam, F. a Aldaye, C. K. McLaughlin, G. D. Hamblin, G. Cosa and H. F.
 Sleiman, *Nat. Chem.*, 2010, 2, 319–328.
- 5 P. K. Lo, F. Altvater and H. F. Sleiman, J. Am. Chem. Soc., 2010, **132**, 10212–10214.
- 6 T. A. Taton, in *Current Protocols in Nucleic Acid Chemistry*, John Wiley & Sons, Inc., Hoboken, NJ, USA, 2002, p. 12.2.1-12.2.12.
- 7 B. L. Baldock and J. E. Hutchison, *Anal. Chem.*, 2016, **88**, 12072–12080.