Supporting Information

Aptamer-tagged DNA Origami for Spatially Addressable Detection of Aflatoxin B1

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1. Materials and methods

1.1. Materials and reagents

Single-stranded *M13mp18* DNA was purchased from New England Biolabs (Catalog number: #N4040S). All unmodified staple strands and thiol-modified complementary ssDNA were synthesized and purified by Shanghai Sangon Biotech Inc. Magnesium acetate tetrahydrate ($C_4H_6MgO_4$•$4H_2O$), sodium citrate tribasic dehydrate ($C_6H_5Na_3O_7 \cdot 2H_2O$), Tris (2-carboxyethyl) phosphine hydrochloride (TCEP), chloroauric acid ($HAuCl_4$) were purchased from Aladdin. Bis (p-sulfonatophenyl) phenylphosphine dihydrate dipotassium salt (BSPP), sodium chloride, aflatoxin B1 were bought from Sigma-Aldrich. All other reagents are of analytical grade and obtained from commercial sources.

1.2. Instruments

A PCR machine (SelectCycler Ⅱ Thermal Cycler) was used to prepare DNA origami. The images of the DNA origami were obtained with an atomic force microscope (AFM) (DIMENSION ICON, Bruker Inc. USA) in ambient conditions. The morphologies of the AuNPs were characterized with field emission scanning electron microscope (SM-7800F, JEOL, Tokyo, Japan). The UV-vis spectra, hydrodynamic sizes and zeta potentials of AuNPs before/after modifications were measured using UV-VIS spectrophotometer (UV-2550, SHIMADZU) and Zetasizer Nano (Malvern Instruments Ltd), respectively.

1.3. DNA sequences

DNA origami substrates were prepared according to the method reported by Rothemund[1] with minor modifications.

The sequences of the staple DNA strands are listed as below (From 5' end to 3' end):

t11s18h,D1, AATACTGCGGAATCGTAGGGGGTAATAGTAAAATGTTTAGACT

t11s28h,E1, TCTTTGATTAGTAATAGTCTGTCCATCACGCAAATTAACCGTT

t11s8h,F1, CAGAAGGAAACCGAGGTTTTTAAGAAAAGTAAGCAGATAGCCG

t1s10g,B2, GACGGGAGAATTAACTCGGAATAAGTTTATTTCCAGCGCC

t1s12i,C2, TCATATGTGTAATCGTAAAACTAGTCATTTTC

t1s14i,D2, GTGAGAAAATGTGTAGGTAAAGATACAACTTT

t1s16i,E2, GGCATCAAATTTGGGGCGCGAGCTAGTTAAAG

t1s18i,F2, TTCGAGCTAAGACTTCAAATATCGGGAACGAG

t1s20g,G2, GAATACCACATTCAACTTAAGAGGAAGCCCGATCAAAGCG

t1s22i,H2, TCGGGAGATATACAGTAACAGTACAAATAATT

t1s24i,A3, CCTGATTAAAGGAGCGGAATTATCTCGGCCTC

t1s26i,B3, GCAAATCACCTCAATCAATATCTGCAGGTCGA

t1s28i,C3, CGACCAGTACATTGGCAGATTCACCTGATTGC

t1s2i,D3, CGGGGTTTCCTCAAGAGAAGGATTTTGAATTA

t1s30g,E3, TTGACGAGCACGTATACTGAAATGGATTATTTAATAAAAG

t1s4i,F3, AGCGTCATGTCTCTGAATTTACCGACTACCTT

t1s6i,G3, TTCATAATCCCCTTATTAGCGTTTTTCTTACC

t1s8i,H3, ATGGTTTATGTCACAATCAATAGATATTAAAC

t2s11g,A4, AGAAAAGCCCCAAAAAGAGTCTGGAGCAAACAATCACCAT

t2s13g,B4, ACAGTCAAAGAGAATCGATGAACGACCCCGGTTGATAATC

t2s15f,C4, ATAGTAGTATGCAATGCCTGAGTAGGCCGGAG

t2s17f,D4, AACCAGACGTTTAGCTATATTTTCTTCTACTA

t2s1g,E4, GATAAGTGCCGTCGAGCTGAAACATGAAAGTATACAGGAG

t2s21g,F4, CCTGATTGCTTTGAATTGCGTAGATTTTCAGGCATCAATA

t2s23g,G4, TGGCAATTTTTAACGTCAGATGAAAACAATAACGGATTCG

t2s25f,H4, AAGGAATTACAAAGAAACCACCAGTCAGATGA

t2s27f,A5, GGACATTCACCTCAAATATCAAACACAGTTGA

t2s3g,B5, TTTGATGATTAAGAGGCTGAGACTTGCTCAGTACCAGGCG

t2s5f,C5, CCGGAACCCAGAATGGAAAGCGCAACATGGCT

t2s7f,D5, AAAGACAACATTTTCGGTCATAGCCAAAATCA

t3s10g,E5, GTCAGAGGGTAATTGATGGCAACATATAAAAGCGATTGAG

t3s14e,F5, CAATATGACCCTCATATATTTTAAAGCATTAA

t3s16e,G5, CATCCAATAAATGGTCAATAACCTCGGAAGCA

t3s18g,H5, AACTCCAAGATTGCATCAAAAAGATAATGCAGATACATAA

t3s20g,A6, CGCCAAAAGGAATTACAGTCAGAAGCAAAGCGCAGGTCAG

t3s24e,B6, TAATCCTGATTATCATTTTGCGGAGAGGAAGG

t3s26e,C6, TTATCTAAAGCATCACCTTGCTGATGGCCAAC

t3s28g,D6, AGAGATAGTTTGACGCTCAATCGTACGTGCTTTCCTCGTT

t3s30g,E6, AGAATCAGAGCGGGAGATGGAAATACCTACATAACCCTTC

t3s4e,F6, TGTACTGGAAATCCTCATTAAAGCAGAGCCAC

t3s6e,G6, CACCGGAAAGCGCGTTTTCATCGGAAGGGCGA

t3s8g,H6, CATTCAACAAACGCAAAGACACCAGAACACCCTGAACAAA

t4s11g,A7, GCAAATATTTAAATTGAGATCTACAAAGGCTACTGATAAA

t4s13g,B7, CGTTCTAGTCAGGTCATTGCCTGACAGGAAGATTGTATAA

t4s15f,C7, CAGGCAAGATAAAAATTTTTAGAATATTCAAC

t4s17f,D7, GATTAGAGATTAGATACATTTCGCAAATCATA

t4s1g,E7, TAGCCCGGAATAGGTGAATGCCCCCTGCCTATGGTCAGTG

t4s21g,F7, GCGCAGAGGCGAATTAATTATTTGCACGTAAATTCTGAAT

t4s23g,G7, GATTATACACAGAAATAAAGAAATACCAAGTTACAAAATC

t4s25f,H7, TAGGAGCATAAAAGTTTGAGTAACATTGTTTG

t4s27f,A8, TGACCTGACAAATGAAAAATCTAAAATATCTT

t4s3g,B8, TTTAACGGTTCGGAACCTATTATTAGGGTTGATATAAGTA

t4s5f,C8, CTCAGAGCATATTCACAAACAAATTAATAAGT

t4s7f,D8, GGAGGGAATTTAGCGTCAGACTGTCCGCCTCC

t5s10g,E8, GATAACCCACAAGAATGTTAGCAAACGTAGAAAATTATTC

t5s14e,F8, TTAATGCCTTATTTCAACGCAAGGGCAAAGAA

t5s16e,G8, TTAGCAAATAGATTTAGTTTGACCAGTACCTT

t5s18g,H8, TAATTGCTTTACCCTGACTATTATGAGGCATAGTAAGAGC

t5s20g,A9, AACACTATCATAACCCATCAAAAATCAGGTCTCCTTTTGA

t5s24e,B9, AATGGAAGCGAACGTTATTAATTTCTAACAAC

t5s26e,C9, TAATAGATCGCTGAGAGCCAGCAGAAGCGTAA

t5s28g,D9, GAATACGTAACAGGAAAAACGCTCCTAAACAGGAGGCCGA

t5s30g,E9, TTAAAGGGATTTTAGATACCGCCAGCCATTGCGGCACAGA

t5s4e,F9, CCTTGAGTCAGACGATTGGCCTTGCGCCACCC

t5s6e,G9, TCAGAACCCAGAATCAAGTTTGCCGGTAAATA

t5s8g,H9, TTGACGGAAATACATACATAAAGGGCGCTAATATCAGAGA

t6s15g,B10, ATAAAGCCTTTGCGGGAGAAGCCTGGAGAGGGTAG

t6s17f,C10, TAAGAGGTCAATTCTGCGAACGAGATTAAGCA

t6s25g,E10, TCAATAGATATTAAATCCTTTGCCGGTTAGAACCT

t6s27f,F10, CAATATTTGCCTGCAACAGTGCCATAGAGCCG

t6s5g,H10, CAGAGCCAGGAGGTTGAGGCAGGTAACAGTGCCCG

t6s7f,A11, ATTAAAGGCCGTAATCAGTAGCGAGCCACCCT

t7s10g,B11, ATAAGAGCAAGAAACATGGCATGATTAAGACTCCGACTTG

t7s14e,C11, ATGACCCTGTAATACTTCAGAGCA

t7s16e,D11, TAAAGCTATATAACAGTTGATTCCCATTTTTG

t7s18g,E11, CGGATGGCACGAGAATGACCATAATCGTTTACCAGACGAC

t7s20g,F11, GATAAAAACCAAAATATTAAACAGTTCAGAAATTAGAGCT

t7s24e,G11, ACAATTCGACAACTCGTAATACAT

t7s26e,H11, TTGAGGATGGTCAGTATTAACACCTTGAATGG

t7s28g,A12, CTATTAGTATATCCAGAACAATATCAGGAACGGTACGCCA

t7s30g,B12, GAATCCTGAGAAGTGTATCGGCCTTGCTGGTACTTTAATG

t7s4e,C12, GCCGCCAGCATTGACACCACCCTC

t7s6e,D12, AGAGCCGCACCATCGATAGCAGCATGAATTAT

t7s8g,E12, CACCGTCACCTTATTACGCAGTATTGAGTTAAGCCCAATA

t8s17g,G12, TAATTGCTTGGAAGTTTCATTCCAAATCGGTTGTA

t8s27g,A1, CGCGAACTAAAACAGAGGTGAGGCTTAGAAGTATT

t8s7g,C1, AGCCATTTAAACGTCACCAATGAACACCAGAACCA

t9s10h,D1, TATCTTACCGAAGCCCAAACGCAATAATAACGAAAATCACCAG

t9s16e,E1, ACTAAAGTACGGTGTCGAATATAA

t9s18g,F1, TGCTGTAGATCCCCCTCAAATGCTGCGAGAGGCTTTTGCA

t9s20h,G1, AAAGAAGTTTTGCCAGCATAAATATTCATTGACTCAACATGTT

t9s26e,H1, ACCACCAGCAGAAGATGATAGCCC

t9s28g,A2, TAAAACATTAGAAGAACTCAAACTTTTTATAATCAGTGAG

t9s30h,B2, GCCACCGAGTAAAAGAACATCACTTGCCTGAGCGCCATTAAAA

t9s6e,C2, CCATTAGCAAGGCCGGGGGAATTA

t9s8g,D2, GAGCCAGCGAATACCCAAAAGAACATGAAATAGCAATAGC

t-10s17h,E2, ACCAACCTAAAAAATCAACGTAACAAATAAATTGGGCTTGAGA

t-10s27h,F2, AACTCACATTATTGAGTGTTGTTCCAGAAACCGTCTATCAGGG

t-10s7h,G2, ACGACAATAAATCCCGACTTGCGGGAGATCCTGAATCTTACCA

t-12s19h,C3, CCTGACGAGAAACACCAGAACGAGTAGGCTGCTCATTCAGTGA

t-12s29h,D3, ACGTGGACTCCAACGTCAAAGGGCGAATTTGGAACAAGAGTCC

t-12s9h,E3, TGCTATTTTGCACCCAGCTACAATTTTGTTTTGAAGCCTTAAA

t-1s10e,F3, AGAGAATAACATAAAAACAGGGAAGCGCATTA

t-1s12i,G3, AGGGATAGCTCAGAGCCACCACCCCATGTCAA

t-1s14e,H3, ATTTTCTGTCAGCGGAGTGAGAATACCGATAT

t-1s14i,A4, CAACAGTTTATGGGATTTTGCTAATCAAAAGG

t-1s16e,B4, ATTCGGTCTGCGGGATCGTCACCCGAAATCCG

t-1s16i,C4, GCCGCTTTGCTGAGGCTTGCAGGGGAAAAGGT

t-1s18g,D4, CGACCTGCGGTCAATCATAAGGGAACGGAACAACATTATT

t-1s18i,E4, GCGCAGACTCCATGTTACTTAGCCCGTTTTAA

t-1s20e,F4, ACAGGTAGAAAGATTCATCAGTTGAGATTTAG

t-1s22i,G4, CGCGTCTGATAGGAACGCCATCAACTTTTACA

t-1s24e,H4, CAGTTTGACGCACTCCAGCCAGCTAAACGACG

t-1s24i,A5, AGGAAGATGGGGACGACGACAGTAATCATATT

t-1s26e,B5, GCCAGTGCGATCCCCGGGTACCGAGTTTTTCT

t-1s26i,C5, CTCTAGAGCAAGCTTGCATGCCTGGTCAGTTG

t-1s28g,D5, TTTCACCAGCCTGGCCCTGAGAGAAAGCCGGCGAACGTGG

t-1s28i,E5, CCTTCACCGTGAGACGGGCAACAGCAGTCACA

t-1s2i,F5, CCTTTTTTCATTTAACAATTTCATAGGATTAG

t-1s30e,G5, CGAGAAAGGAAGGGAAGCGTACTATGGTTGCT

t-1s4e,H5, TTATCAAACCGGCTTAGGTTGGGTAAGCCTGT

t-1s4i,A6, TTTAACCTATCATAGGTCTGAGAGTTCCAGTA

t-1s6e,B6, TTAGTATCGCCAACGCTCAACAGTCGGCTGTC

t-1s6i,C6, AGTATAAAATATGCGTTATACAAAGCCATCTT

t-1s8g,D6, TTTCCTTAGCACTCATCGAGAACAATAGCAGCCTTTACAG

t-1s8i,E6, CAAGTACCTCATTCCAAGAACGGGAAATTCAT

t-2s11g,F6, CCTCAGAACCGCCACCCAAGCCCAATAGGAACGTAAATGA

t-2s13g,G6, AGACGTTACCATGTACCGTAACACCCCTCAGAACCGCCAC

t-2s15f,H6, CACGCATAAGAAAGGAACAACTAAGTCTTTCC

t-2s17f,A7, ATTGTGTCTCAGCAGCGAAAGACACCATCGCC

t-2s1g,B7, AAAACAAAATTAATTAAATGGAAACAGTACATTAGTGAAT

t-2s21g,C7, GCTCATTTTTTAACCAGCCTTCCTGTAGCCAGGCATCTGC

t-2s23g,D7, GTAACCGTCTTTCATCAACATTAAAATTTTTGTTAAATCA

t-2s25f,E7, ACGTTGTATTCCGGCACCGCTTCTGGCGCATC

t-2s27f,F7, CCAGGGTGGCTCGAATTCGTAATCCAGTCACG

t-2s3g,G7, AGAGTCAAAAATCAATATATGTGATGAAACAAACATCAAG

t-2s5f,H7, ACTAGAAATATATAACTATATGTACGCTGAGA

t-2s7f,A8, TCAATAATAGGGCTTAATTGAGAATCATAATT

t-3s10g,B8, AACGTCAAAAATGAAAAGCAAGCCGTTTTTATGAAACCAA

t-3s14e,C8, GTTTTGTCAGGAATTGCGAATAATCCGACAAT

t-3s16e,D8, GACAACAAGCATCGGAACGAGGGTGAGATTTG

t-3s18g,E8, TATCATCGTTGAAAGAGGACAGATGGAAGAAAAATCTACG

t-3s20g,F8, TTAATAAAACGAACTAACCGAACTGACCAACTCCTGATAA

t-3s24e,G8, TGTAGATGGGTGCCGGAAACCAGGAACGCCAG

t-3s26e,H8, GGTTTTCCATGGTCATAGCTGTTTGAGAGGCG

t-3s28g,A9, GTTTGCGTCACGCTGGTTTGCCCCAAGGGAGCCCCCGATT

t-3s30g,B9, TAGAGCTTGACGGGGAGTTGCAGCAAGCGGTCATTGGGCG

t-3s4e,C9, GATTAAGAAATGCTGATGCAAATCAGAATAAA

t-3s6e,D9, CACCGGAATCGCCATATTTAACAAAATTTACG

t-3s8g,E9, AGCATGTATTTCATCGTAGGAATCAAACGATTTTTTGTTT

t-4s11g,F9, AGGTTTAGTACCGCCATGAGTTTCGTCACCAGGATCTAAA

t-4s13g,G9, AGCGTAACTACAAACTACAACGCCTATCACCGTACTCAGG

t-4s15f,H9, TAGTTGCGAATTTTTTCACGTTGATCATAGTT

t-4s17f,A10, GTACAACGAGCAACGGCTACAGAGGATACCGA

t-4s1g,B10, GAGCAAAAGAAGATGAGTGAATAACCTTGCTTATAGCTTA

t-4s21g,C10, GTTAAAATTCGCATTAATGTGAGCGAGTAACACACGTTGG

t-4s23g,D10, GGATAGGTACCCGTCGGATTCTCCTAAACGTTAATATTTT

t-4s25f,E10, AGTTGGGTCAAAGCGCCATTCGCCCCGTAATG

t-4s27f,F10, CGCGCGGGCCTGTGTGAAATTGTTGGCGATTA

t-4s3g,G10, ACATAGCGCTGTAAATCGTCGCTATTCATTTCAATTACCT

t-4s5f,H10, GTTAAATACAATCGCAAGACAAAGCCTTGAAA

t-4s7f,A11, CCCATCCTCGCCAACATGTAATTTAATAAGGC

t-5s10g,B11, TCCCAATCCAAATAAGATTACCGCGCCCAATAAATAATAT

t-5s16e,D11, AACAGCTTGCTTTGAGGACTAAAGCGATTATA

t-5s18g,E11, CCAAGCGCAGGCGCATAGGCTGGCAGAACTGGCTCATTAT

t-5s20g,F11, ACCAGTCAGGACGTTGGAACGGTGTACAGACCGAAACAAA

t-5s26e,H11, TGCTGCAAATCCGCTCACAATTCCCAGCTGCA

t-5s28g,A12, TTAATGAAGTTTGATGGTGGTTCCGAGGTGCCGTAAAGCA

t-5s30g,C12, CTAAATCGGAACCCTAAGCAGGCGAAAATCCTTCGGCCAA

t-5s6e,D12, GTGTGATAAGGCAGAGGCATTTTCAGTCCTGA

t-5s8g,E12, ACAAGAAAGCAAGCAAATCAGATAACAGCCATATTATTTA

t-6s13f,F12, ACAGACAGCCCAAATCTCCAAAAAAAAATTTCTTA

t-6s15c,G12, CGAGGTGAGGCTCCAAAAGGAGCC

t-6s17f,H12, ACCCCCAGACTTTTTCATGAGGAACTTGCTTT

t-6s23f,A1, CGGCGGATTGAATTCAGGCTGCGCAACGGGGGATG

t-6s25c,B1, TGGCGAAATGTTGGGAAGGGCGAT

t-6s27f,C1, TGTCGTGCACACAACATACGAGCCACGCCAGC

t-6s3f,D1, TCCCTTAGAATAACGCGAGAAAACTTTTACCGACC

t-6s5c,E1, GTTTGAAATTCAAATATATTTTAG

t-6s7f,F1, AATAGATAGAGCCAGTAATAAGAGATTTAATG

t-7s10g,G1, GCCAGTTACAAAATAATAGAAGGCTTATCCGGTTATCAAC

t-7s18g,A2, AAAACACTTAATCTTGACAAGAACTTAATCATTGTGAATT

t-7s20g,B2, ACCTTATGCGATTTTATGACCTTCATCAAGAGCATCTTTG

t-7s28g,D2, TTCCAGTCCTTATAAATCAAAAGAGAACCATCACCCAAAT

t-7s30g,E2, CAAGTTTTTTGGGGTCGAAATCGGCAAAATCCGGGAAACC

t-7s8g,G2, GCGCCTGTTATTCTAAGAACGCGATTCCAGAGCCTAATTT

t-7s8g,G2, GCGCCTGTTATTCTAAGAACGCGATTCCAGAGCCTAATTT

t-8s15f,H2, CGGTTTATCAGGTTTCCATTAAACGGGAATACACT

t-8s17c,A3, GGCAAAAGTAAAATACGTAATGCC

t-8s25f,B3, TCTTCGCTATTGGAAGCATAAAGTGTATGCCCGCT

t-8s27c,C3, GCGCTCACAAGCCTGGGGTGCCTA

t-8s5f,D3, TTCTGACCTAAAATATAAAGTACCGACTGCAGAAC

t-8s7c,E3, TCAGCTAAAAAAGGTAAAGTAATT

t-9s10g,F3, ACGCTAACGAGCGTCTGGCGTTTTAGCGAACCCAACATGT

t-9s20g,H3, TGGTTTAATTTCAACTCGGATATTCATTACCCACGAAAGA

t-9s30g,B4, CGATGGCCCACTACGTATAGCCCGAGATAGGGATTGCGTT

ts-rem1,D4, GCGCTTAATGCGCCGCTACAGGGC

t-5s2e-t6s23c-3T,A6, TTAATTAATTTTTTACCATATCAAA

t-7s4e-t8s25c-2T,B6, TTAATTTCATCTTAGACTTTACAA

t-9s6e-t10s27c-1T,C6, CTGTCCAGACGTATACCGAACGA

t-11s8e-t12s29c-0T,D6, TCAAGATTAGTGTAGCAATACT

t-5s12e-t6s3c-3T,E6, TGTAGCATTCCTTTTATAAACAGTT

t-7s14e-t8s5c-2T,F6, TTTAATTGTATTTCCACCAGAGCC

t-9s16e-t10s7c-1T,G6, ACTACGAAGGCTTAGCACCATTA

t-11s18e-t12s9c-0T,H6, ATAAGGCTTGCAACAAAGTTAC

t-5s22e-t6s13c-3T,A7, GTGGGAACAAATTTCTATTTTTGAG

t-7s24e-t8s15c-2T,B7, CGGTGCGGGCCTTCCAAAAACATT

t-9s26e-t10s17c-1T,C7, ATGAGTGAGCTTTTAAATATGCA

t-11s28e-t12s19c-0T,D7, ACTATTAAAGAGGATAGCGTCC

The blue colored staple strands (t1s14i, D2) were replaced by the AFB1 probe in the designed origami.

Sequences of the AFB_1 capture probe:

GTTGGGCACGTGTTGTCTCTCTGTGTCTCGTGCCCTTCGCTAGG

CCCACTTTTTGTGAGAAAATGTGTAGGTAAAGATACAACTTT[1] [2]

DNA stand complements with the AFB_1 probe:
AGAGACAACACGTGCCCAACAAAAAA-$(CH_2)_3$-SH

2. Preparation of DNA origami substrate

The single-strand *M13mp18* DNA and the staple strands (including AFB_1 probe) were mixed at a molar ratio of 1:10 in a 1×TAE~Mg^{2+} buffer (40 mM Tris, 20 mM acetic acid, 1 mM EDTA, 12.5 mM magnesium acetate). The mixture was annealed from 95°C to 20°C in PCR (1°C /100s). [1] The DNA origami was washed with centrifugal filters for three times to remove excess staple strands (washing buffer: 1×TAE~Mg^{2+} buffer). [3]

3. Synthesis of AuNPs

AuNPs were synthesized by citrate reduction method[4]. Briefly, 2 mL $HAuCl_4$ (1 wt%) was mixed with 198 mL deionized water in a round-bottom flask equipped with a reflux condenser. When the solution was heated to a boil, 4 mL sodium citrate (1%) was rapidly injected under stirring. The mixture was boiled for another 30 min until its color turned to deep red.

4. Preparation of the AuNP-complementary ssDNA conjugates

AuNP-DNA conjugates were synthesized according to Ding's route[5]. Briefly, 15 mg BSPP was added into 50 mL AuNPs suspension (1.0 μg/mL), stirring overnight at room temperature. Solid sodium chloride was gradually added until the color of the solution changes from wine red to lilac. The suspension was centrifuged at 10,000 rpm for 15 min to pellet the AuNPs. After washing with a solution containing 1 mL BSPP (2.5 mM) and 1 mL CH_3OH, the pellet was dispersed in 1 mL BSPP (2.5mM). After activated by TCEP (10 mM) for 2 hours, thiol-modified DNA (100 μM) was incubated with AuNPs at a molar ratio of 200:1 in 0.5×TBE~50 mM NaCl buffer for 40 hours at room temperature. After that, the products were collected and washed three-time with 0.5×TBE~50 mM NaCl buffer by centrifugation at a speed of 13,000 rpm. The concentration of AuNPs-DNA conjugates was estimated according to the Wolfgang Haiss's approach.

5. AFB1 detection with origami substrate

The AFB1 probe-tagged DNA origami was incubated with AFB_1 at different molar ratio (1:100, 1:10, 1:1, 1:0) for 30 min at room temperature [6]. The corresponding final concentrations are 80, 8, 0.8 and 0 ng/mL, respectively. Then, AuNPs-DNA conjugates were added into the mixture and slowly cooled down from 43°C to 20°C at a rate of 0.5°C per second in a PCR machine [5]. During this process, all un-occupied probes can react with the AuNP-DNA conjugates.

6. AFM measurement

A 10-μL aqueous sample was dropped onto freshly cleaved mica, which was pre-treated with Mg^{2+} ions to enable the easy adsorption of DNA origami, drying at ambient conditions for 1 hour [7]. The dried samples were scanned with AFM at ScanAsyst-mode in air.

7. Gel electrophoresis

The samples were pre-mixed with NAS and then loaded in 1.5% agarose gels. After running for three hours at 80 V, [8, 9] the gels were photographed using a digital camera system (Chemi XR5).

8. Characterization of AuNPs

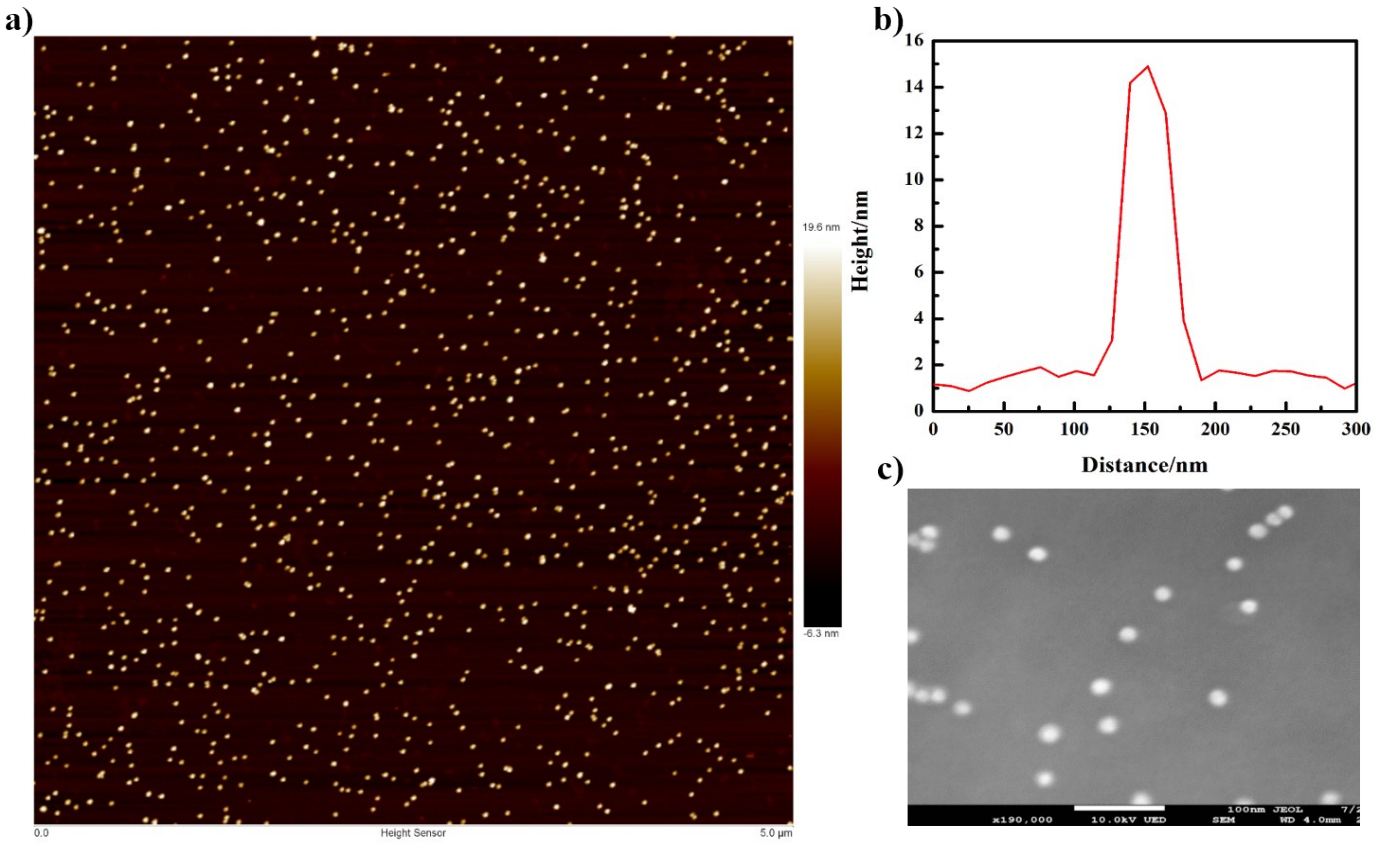


Figure S1. a) AFM and c) FE-SEM images of AuNPs; b) is an AFM section curve showing the size of a AuNP.

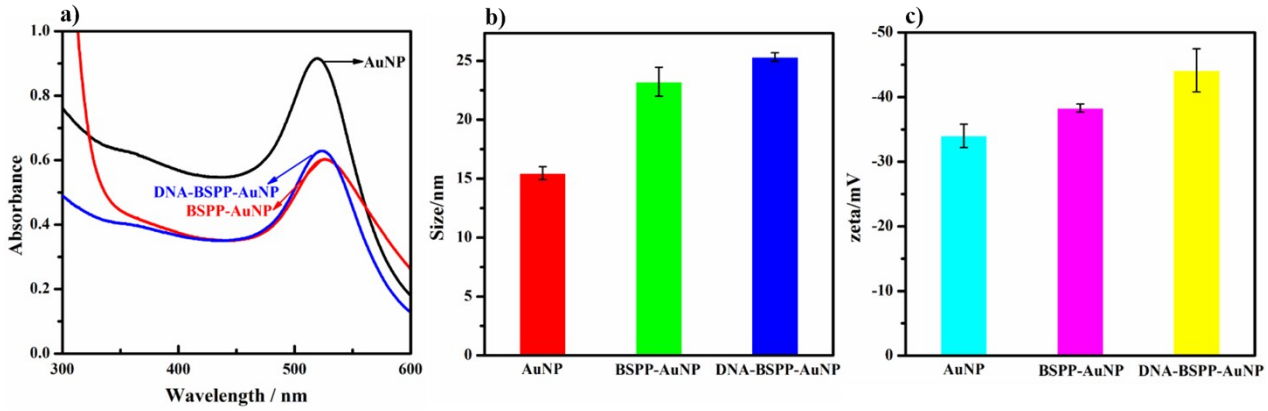


Figure S2. UV-vis (a), hydrodynamic size (b) and zeta potential (c) of AuNPs before and after modified with BSPP and SH-DNA. Data in b) and c) are presented as the average ± standard deviation from three independent measurements.

As shown in the AFM image and the FESEM micrograph (Fig. S1), the as-prepared AuNPs are uniformly dispersed on the substrates and the average size of them is around 15 nm. The surface coating of BSPP and DNA causes the red-shift and significant decrease of the absorption peak (Fig. S2a). Both hydrodynamic size and zeta potential of AuNPs increase after surface modification with BSPP and DNA (Fig. S2b and S2c). The results clearly indicate the successful surface coating of BSPP and DNA on the AuNPs.

9. Relative sites between the origami and the anchored AuNP

Figure S3 illustrates the AFM images of the pure DNA origami and the DNA origami-AuNP complex. The attached AuNP could be located at the angle, side and center of the triangle DNA origami substrate. Since DNA origami is a highly addressable substrate, the possible resting site of the AuNP could be estimated based on the sizes of the origami and AuNP, as well as the lengths of the probe and complementary strands. As shown in Scheme S1, A is the designed site of the AFB1 probe. After hybridized with the AuNP-conjugated ssDNA, the AuNP could freely move in a circle with the A site as the center. The radius of the circle should be the length of linking strand plus the diameter of the AuNP. Thus, the relative sites observed in the AFM images could be well explained by the above calculation.

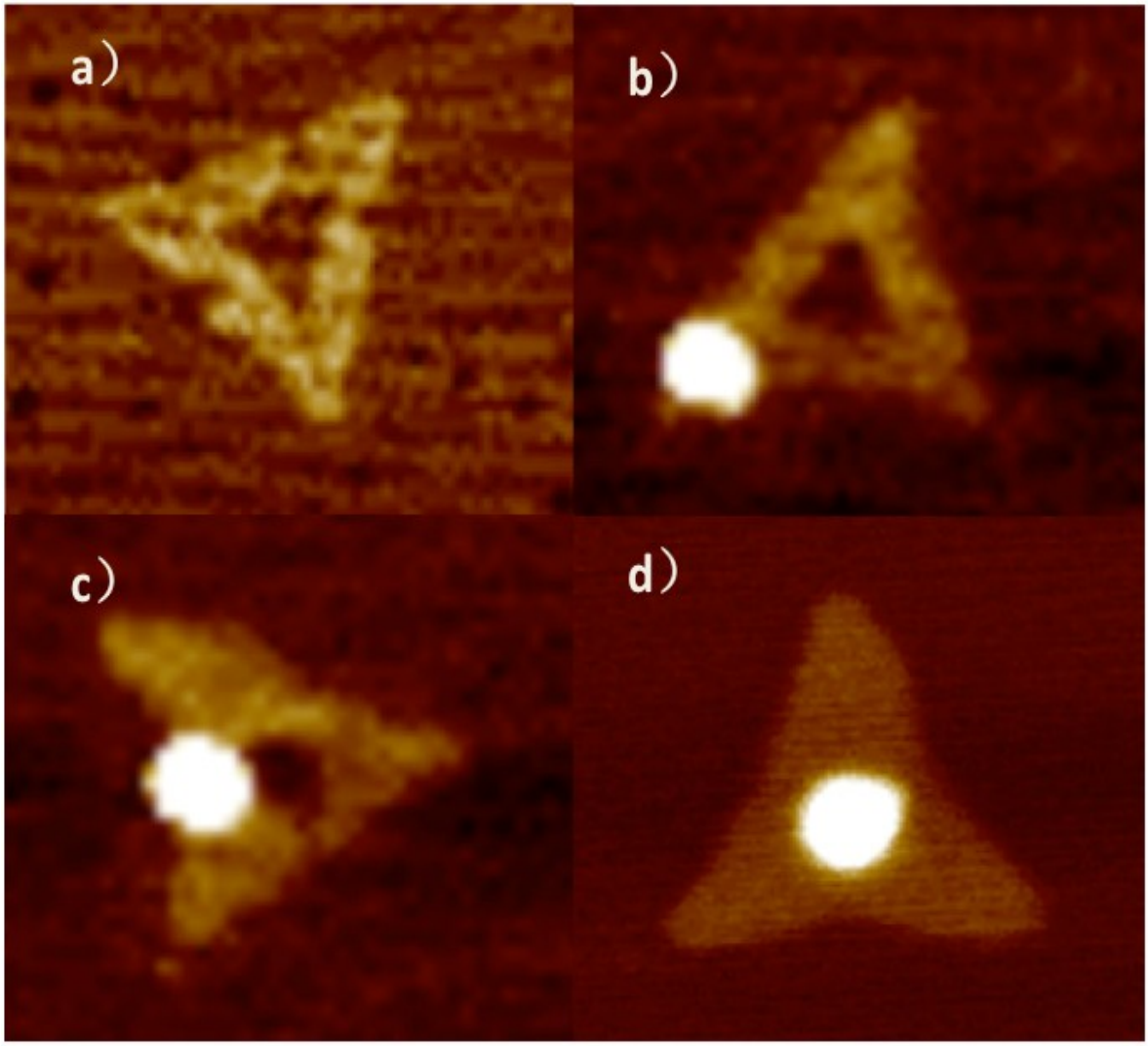
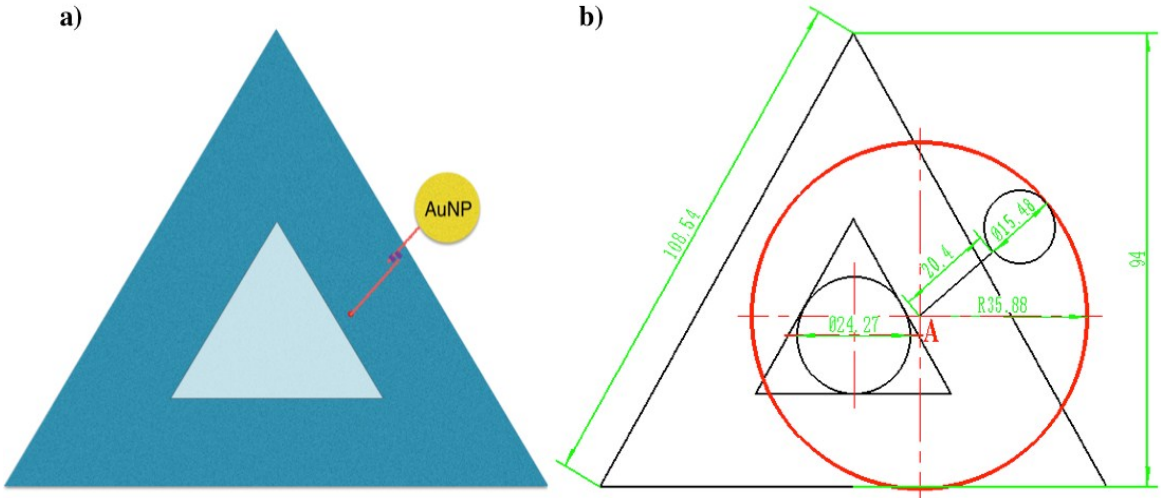


Figure S3. AFM images of the DNA origami and the DNA origami-AuNP complex. a) DNA origami with an AFB1 probe; DNA origami substrates with an angle- (b), side- (c) and center-attached (d) AuNP, respectively.



Scheme S1. Schematic drawing illustrating the possible sites of the origami-anchored AuNP.

Table S1. Binding rates of ssDNA-AuNPs conjugates on DNA origami templates in the detection systems containing different concentrations of AFB1.

Column	AFB1 Concentration	Mean	SD
a	0 ng / mL	64.65%	0.87
b	0.8 ng / mL	52.13%	1.93
c	8.0 ng / mL	47.43%	1.52
d	80.0 ng / mL	37.88%	2.45

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