Supporting Information for:

DNA-Functionalized Gold Nanoparticles with Toehold-Mediated Strand Displacement for Nucleic Acid Sensors

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SUPPORTING TABLE

Table S1. Fitted parameters of equation 1 for the melting curves of different combinations of DNA-AuNPs.

Density		Fitted Parameters				
Probe	Seq. 2	A_1	A_2	X_0	p	s
220	260	-0.01278	0.211	48.24	0.602	1.026
154	260	-0.00059	0.129	47.52	0.577	0.997
121	260	-0.01179	0.396	47.48	0.183	1.024
220	202	0.00135	0.195	39.39	0.546	0.983
154	202	0.000523	0.133	37.78	0.287	0.947

SUPPORTING FIGURES

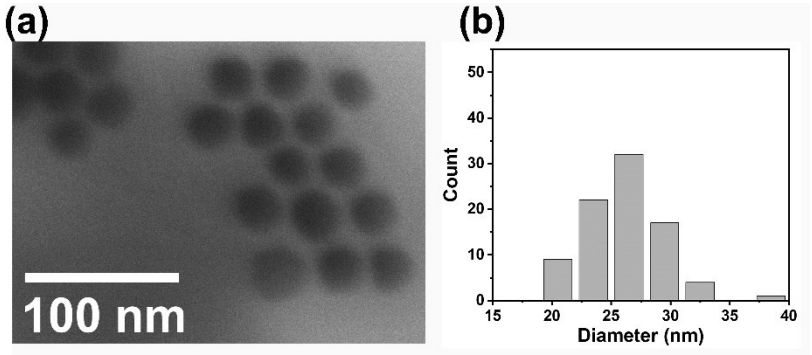

Figure S1. (a) Scanning transmission electron microscope image of DNA-AuNPs and (b) their size distribution.

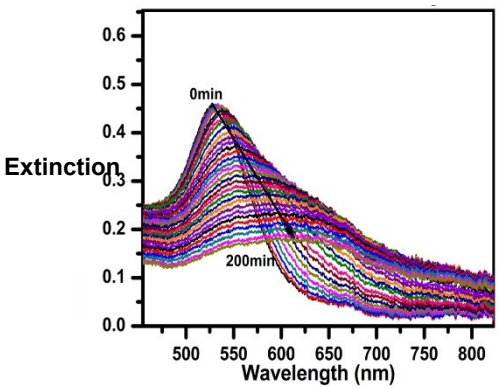

Figure S2. Representative plot of the extinction spectra evolution during the aggregation of DNA-AuNPs.

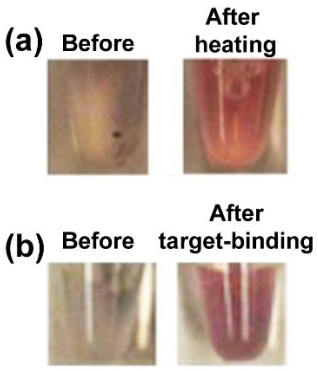

Figure S3. Photographs of the samples before and after (a) thermal melting and (b) incubation with the target at room temperature. The after photos were taken at the end of the respective experiments.

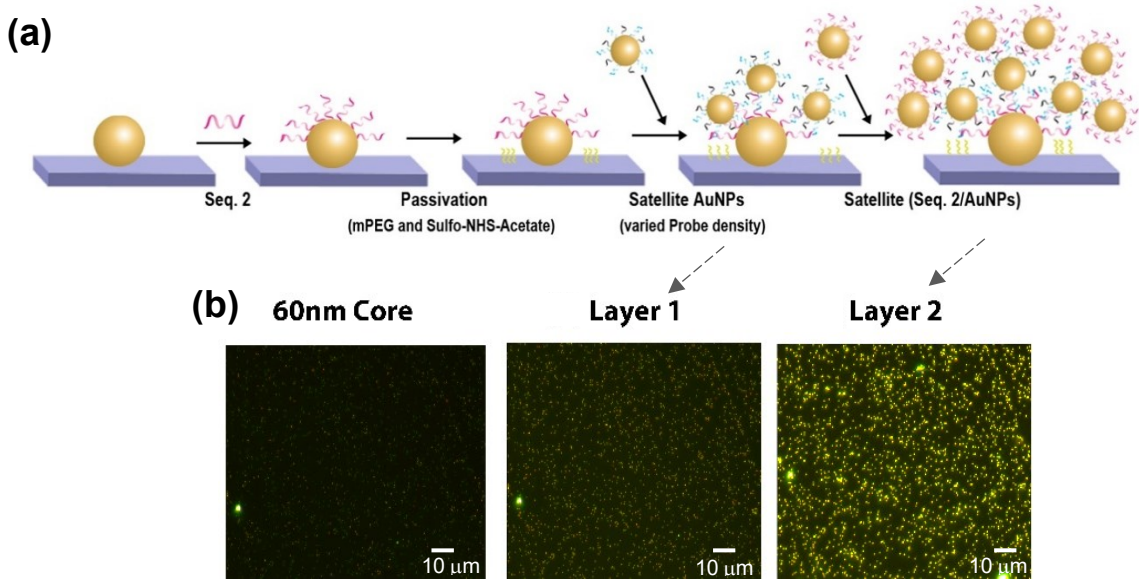

Figure S4. Layer-by-layer fabrication of core-satellite AuNP assemblies: (a) schematic of the procedure and (b) dark-field images captured during the assembly process.

SUPPORTING EXPERIMENTAL

Fabrication of core-satellite assemblies as chip-based sensors

The procedure was adapted from previously work[1] and briefly described below.

Deposition and DNA-functionalization of core AuNPs on amino-modified substrate

Citrate-capped AuNPs (60 nm) were first immobilized on an aminopropyl-trimethoxysilane- (APTMS) treated coverslip. The anchored core particles were then functionalized with Seq. 2 using 10 μL of solution containing 0.01 M phosphate buffer (PB), 1.25 M NaCl, 0.1% SDS and DNA (2-5 μM) for 2 hours. The coverslip was rinsed with water and dried with compressed air.

To prevent non-specific binding of the satellite nanoparticles on the amino-modified glass coverslip, the remaining amino groups were passivated in 10 mM mPEG-Succinimidyl Valerate (LaysanBio) solution in 0.1 M bicarbonate buffer (pH of 8.3) for 4 hours. Then the coverslip was treated with 0.2 M solution of sulfo-NHS-acetate (Sulfosuccinimidyl Acetate, Thermo Scientific) in 0.1 M bicarbonate buffer (pH 8.3) for 4 hours. The sample was rinsed with water and dried with compressed air.

Layer-by-layer assembly of satellite nanoparticles

Colloidal solutions of Probe- and Seq. 2-functionalized 30-nm AuNPs were prepared as in solution experiments. To self-assemble the first layer of satellite nanoparticles, a volume of ~45 μL of 0.6 nM of Probe-AuNP in 0.01 M PB, 0.4 M NaCl and 0.01% SDS was incubated with core AuNPs on the coverslip for 45-60 min. The solution was replaced with buffer (0.01 M PB, 0.4 M NaCl) and the second set of satellite AuNPs (Seq. 2-AuNP) was added and incubated similarly. Each layer of the assembly process was monitored by darkfield microscopy. The coverslip samples were stored in 0.01 M PB and 0.13 M NaCl until sensing experiments.

Sensing of DNA-210

Micromolar of DNA-210 was introduced to the sample in 0.01 M PB and 0.13 M NaCl. A SecureSeal chamber (Grace Bio-Labs) was used for incubation. The dark-field images were captured using a Nikon TE2000 equipped with a Jenoptik CCD camera. Image analysis was carried out using Igor as established previously.[1]

Electron microscopy

A Quanta 3D electron microscope in the scanning transmission mode at 25 kV was employed for morphological characterization.

Reference

1. Le, N. H.; Nguyen, B. K.; Ye, G.; Peng, C.; Chen, J. I. L., Tuning the Sensing Performance of Multilayer Plasmonic Core–Satellite Assemblies for Rapid Detection of Targets from Lysed Cells. ACS Sensors 2017, 2 (11), 1578-1583.