Electronic Supplementary Information (ESI)

Plasmonic Bimetallic Nanodisk Arrays for DNA Conformation Sensing

Thanh Thi Van Nguyen,^a Xiaoji Xie,^b Jiahui Xu,^b Yiming Wu,^b Minghui Hong^c and Xiaogang Liu^{*,b,d,e}

^a Advanced Materials for Micro- and Nano-Systems Programme, Singapore-MIT Alliance, Singapore 117576. ^b Department of Chemistry, National University of Singapore, 3 Science Drive 3, Singapore 117543. ^c Department of Electrical and Computer Engineering, National University of Singapore, Singapore 117576. ^d The N.1 Institute for Health, National University of Singapore, 28 Medical Dr. #05-COR, Singapore 117456. ^e Joint School of National University of Singapore and Tianjin University, International Campus of Tianjin University, Fuzhou, 350207, P. R. China

DNA sequences

Name	Description	Sequence	
Single-stranded DNA detection			
а	Capture DNA	5'-[thiolC6]AAACGGGAGGCAGTG-3'	
b	Capture DNA	5'-ATAAATCACTCAAAA[thiolC3]-3'	
t	Target DNA	3'-GCCCTCCGTCACTATTTAGTGAGT-5'	
ť	Non- complementary target DNA	5'-ACCTGGAGGAGTATTGCGGAGGAAGGT-3'	
	I	Different DNA length strands	
a1	Capture DNA	5'-[thiolC6]CTAGAAGGACCTCTT-3'	
b1	Capture DNA	5'-ATTCGATCCTTTCTA[thiolC3]-3'	
t1	24-base target DNA	5'-AAAGGATCGAATAAGAGGTCCTTC-3'	
t2	48-base target DNA	5'-AAAGGATCGAATCCGTTAAGACGAGGCAATCATGCA AAGAGGTCCTTC-3'	
t3	72-base target	5'-AAAGGATCGAATCCGTTAAGACGAGGCAATCATGCA	
.5	DNA	TATATTGGCCGCTTT AGCGACAACAAGAGGTCCTTC-3'	
t4	96-base target DNA	5'-AAAGGATCGAATCCGTTAAGACGAGGCAATCATGCA TATATTGGCCGCTTTAGCGACAACCTCATGTACGTATGTAGGATCCGAAAGAGGTCC TTC-3'	
		Different DNA configuration strands	
a2	Capture DNA	5'-[ThiolC6]TTTTTTTTTTTTTTT-3'	
b2	Capture DNA	5'-TTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTT	
1L#	Linear strand	5'-A17TCGGATCCTACGTAGGAATTA19-3'	
	Linear strand	5'-A18TCGGATCCTATCGAACTGCATAAGCGCACTCGTA	
2L#		GGAATTA18-3'	
3L#	Linear strand	5'-A20 TCGGATCCTACATACGTACATGAGCATTACTCGA	
		ACTGCATAAGC GCACTCGTAGGAATTA19-3'	
1B#	Bulge strand	5'-A15 TCGGATCCTA AAA CGTAGGAATTA16-3'	
2B#	Bulge strand	5'-A15 TCGGATCCTA <u>AAA</u> TCGAACTGCATAAGCGCACT <u>AAA</u> CGTAGGAATT A15-3'	
3B#	Bulge strand	5'-A15 TCGGATCCTA AAA CATACGTACATGAGCATTAC AAATCGAACTGC TAAGCGCACTAAACGTAGGAATTA15-3'	
1K#	Linker strand	3'-AGCCTAGGATGCATCCTTAA-5'	
2K#	Linker strand	3'-AGCCTAGGATAGCTTGACGTATTCGCGTGAGCATCC	

		TTAA-5'
ЗК#	Linker strand	3'-AGCCTAGGATGTATGCATGTACTCGTAATGAGCT
		TGACGTATTCGCGT GAGCATCCTTAA-5'
G-Q	G-quadruplex	5'-A15TGGG(T ₂ AG ₃) ₃ A15-3'



Figure S1. Schematic presentation of the fabrication process: (i) photoresist coating, (ii) LIL exposure and development, (iii) metal coating, and (iv) lift-off.



Figure S2. LSPR peak shifts versus the refractive index of surrounding media for different nanodisk structures. 30Au and 30Ag nanodisk structures were fabricated for comparison. The measured points at n = 1.0 (ambient air) are not showed in the refractive index range.



xy section at half height of the nanodisk

Figure S3. FDTD simulated field distribution of (a) 20Ag10Au and (b) 10Ag20Au nanodisks at xy and xz sections, respectively.



Figure S4. Extinction spectra of (a) 10Ag20Au and (b) 15Ag15Au nanodisk arrays after each of modification step and (c) The LSPR shift enhancement by gold nanoparticle attachments of 3 different nanodisk structures.



Figure S5. SEM images of 20Ag10Au nanodisk arrays after attaching DNA-modified gold nanoparticles with (a) 1 pM and (b) 100 nM concentration of target ssDNA.



Figure S6. The sensitivity of the plasmon enhancement platform for ssDNA sensing. Black triangle: the LSPR peak shifts at various concentrations of target ssDNA after excluding the effects of nonspecific-binding. Blue curve: quantitative response curve for DNA-modified gold nanoparticles binding to DNA-modified nanodisks upon the DNA hybridization. Note: the sensitivity is calculated according to Langmuir adsorption equation: $\Delta R = \Delta R_{\text{max}} \frac{K_a \times C[DNA]}{1 + K_a \times C[DNA]}$, where $\Delta R = \Delta \lambda_{\text{max}}$,

LSPR shift at a given concentration, ΔR_{max} is the maximum LSPR response at high concentrations, K_a is the surface-confined thermodynamic affinity constant, and C[DNA] is the concentration of target DNA solution.



Figure S7. LSPR extinction spectra of 20Ag10Au nanodisk structures illustrate nonspecific bindings of DNA modified nanodisks interacting with (a) non-complementary target DNA, (b) DNA-modified gold nanoparticles, (c) non-complementary target DNA and DNA-modified gold nanoparticles.



Figure S8 (a) Experimental extinction spectra of 20Ag10Au nanodisk arrays at each step in the surface modification with different length linker DNAs: (from left to right) 24-base, 48-base, 72-base, and 96-base DNA. (b) FDTD simulation of plasmonic interaction between a 20Ag10Au nanodisk and a gold nanoparticle with the gap between them equating to the length of linker DNAs, (from left to right) 7.2 nm, 14.4 nm, 21.6 nm, and 28.8 nm respectively.



Figure S9. Extinction spectra of 20Ag10Au nanodisk arrays after DNA-modified gold nanoparticle attachment by G-quadruplex DNA in the presence of (a) K^+ and (b) Na⁺ buffer; and (c) the corresponding LSPR shifts.