

Conformational Change and Biocatalysis-Triggered Spectral Shift of Single Au Nanoparticles

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Supporting Information

1. Experimental Section

Oligonucleotides:

HPLC purified oligonucleotides were obtained from Sangon Biotechnology Co., Ltd. (Shanghai, China).

Reagents:

GNP stoste with an average diameter of 50 nm was obtained from TED PELLA, Inc. (USA). The GNP solution was prepared by diluting the GNP stoste with deionized water. Benzidine (DAB) was purchased from AccuStandard (USA). Hemin was obtained from Sigma Aldrich (USA). The stock solution of slightly soluble DAB was prepared in acidic deionized water (adjusted with concentrated hydrochloric acid), the pH of the stock solution was adjusted back to 7 with 1 M NaOH after thorough dissolution. The stock solution of hemin (5 mM) was prepared in DMSO, stored in dark at -20 °C and diluted to the required concentration with HEPES buffer solution (25 mM). H₂O₂ and Cetyltrimethylammonium bromide (CTAB) were of analytical grade, and used as received without further purification. All oligonucleotide stock solutions were prepared with deionized water from a Millipore system and quantified using UV-vis absorption spectroscopy.

Instrument:

Circular dichroism (CD) spectra were measured on a Jasco J-810 spectropolarimeter (Jasco, Japan). Dark field images were acquired on a Nikon inverted microscope Eclipse Ti-U equipped with a colored CCD (Nikon, DS-FI1-U2). The NPPR scattering spectra were recorded using SP2556 spectrograph mounted on the microscope, and a 512B_excelon EMCCD was used as the detector (Princeton Instruments, USA). Glass slides were cleaned under PSD-UV4 ozone system (Novascan Technologies) before use. All tests were carried out at room temperature.

Modification of GNPs on glass slide:

The GNPs were modified on glass slide for dark field observation. Before modification, the microscope slide was first wiped with ethanol-immersed absorbent cotton, followed by sonication in ethanol for 2 h to remove the dust particles. Then, the slides were thoroughly rinsed with Milli-Q water, blow-dried with nitrogen, stepped by a treatment with Ultra-Violet/Ozone cleaner for 15 min in order to further remove the surface organic contaminants. The cleaned slide was then immersed into 1 mM CTAB solution for 1 h, followed by successive rinsing with ethanol and blow-drying with nitrogen. The freshly prepared diluted GNPs solution (~ 0.02 nM) was dropped onto the CTAB-covered microscope slide for 10 min, then rinsed with water and dried with nitrogen. The GNP-modified glass slides were prepared and stored for further use. The step of CTAB treatment is to change the surface charge character of the glass slide, in order to achieve a high adsorption efficiency of negatively charged GNPs. To eliminate the possible influence of CTAB on further modification of GNP with biomolecules, the GNP-modified glass slide were treated with Ultra-Violet/Ozone cleaner for another 15 min to remove (at least partly) CTAB on the glass slide. It is worthy to note that the modified GNPs will stably attached on the slide glass even after rinsing and CTAB removing procedure, proved by the unchanged DFM image after all modification steps.

2. Supporting Figures

Figure S1. The transmission electron microscopic image of GNPs.

Figure S2. Statistical histogram of the plasmonic peak position of 21 GNPs with green scattering color.

FigureS3. Circular dichroism spectra of the G-rich DNA in bulk solution with different potassium concentration.

Figure S4. The NPPR scattering spectra of the G-DNA-modified GNPs with different size. (a) GNP-G-DNA with NPPR peak centered at 549 nm; (b) GNP-G-DNA with NPPR peak centered at 558 nm; (c) GNP-G-DNA with NPPR peak centered at 566 nm; (d) GNP-G-DNA with NPPR peak centered at 541 nm.

Figure S5. The NPPR scattering spectra of the random DNA-modified GNP.

Figure S6. The NPPR scattering spectra of GNP-R-DNA in 0.1 mM DAB and 100 μM H_2O_2 solution after the hemin treatment step.

Figure S7. The NPPR scattering spectra of different immobilized GNP-DNAzyme with 0.1 mM DAB in (a) 500 μM H_2O_2 . (b) 100 μM H_2O_2 , respectively.

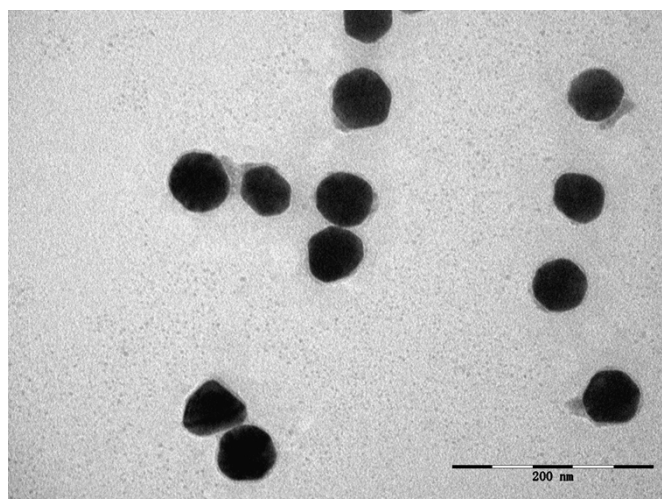


Figure S1. The transmission electron microscopic image of GNPs.

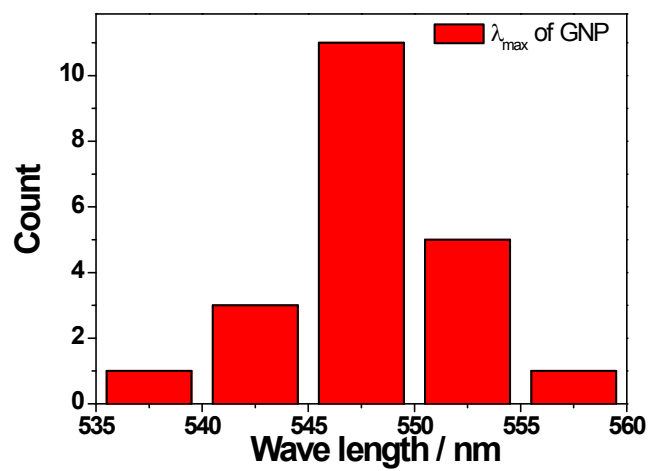


Figure S2. Statistical histogram of the plasmonic peak position of 21 GNPs with green scattering color.

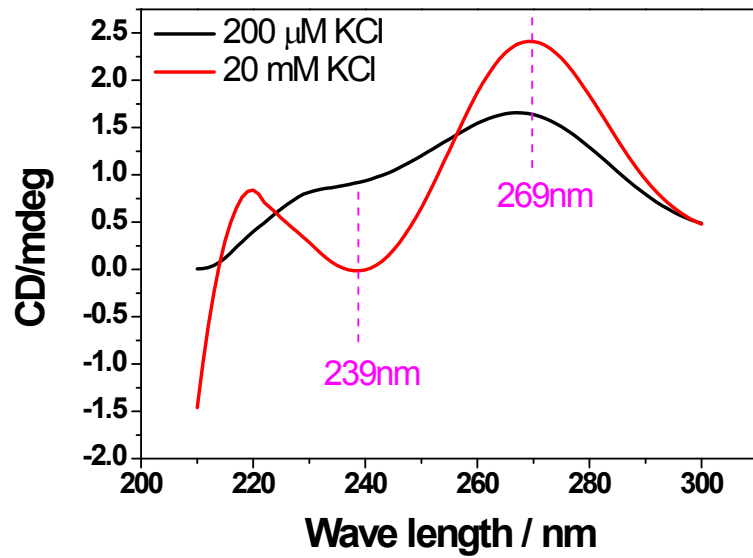


Figure S3. Circular dichroism spectra of G-rich DNA in bulk solution with different potassium concentration.

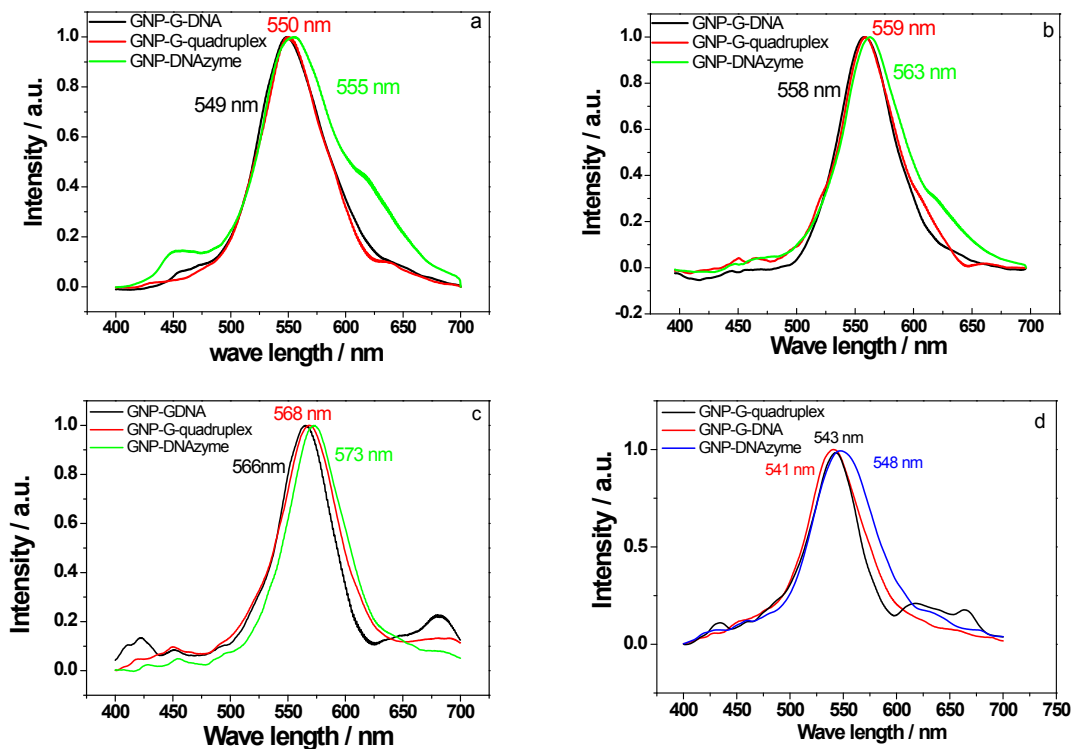


Figure S4. The NPPR scattering spectra of the G-DNA-modified GNPs with different size. (a) GNP-G-DNA with NPPR peak centered at 549 nm; (b) GNP-G-DNA with NPPR peak centered at 558 nm; (c) GNP-G-DNA with NPPR peak centered at 566 nm; (d) GNP-G-DNA with NPPR peak centered at 541 nm.

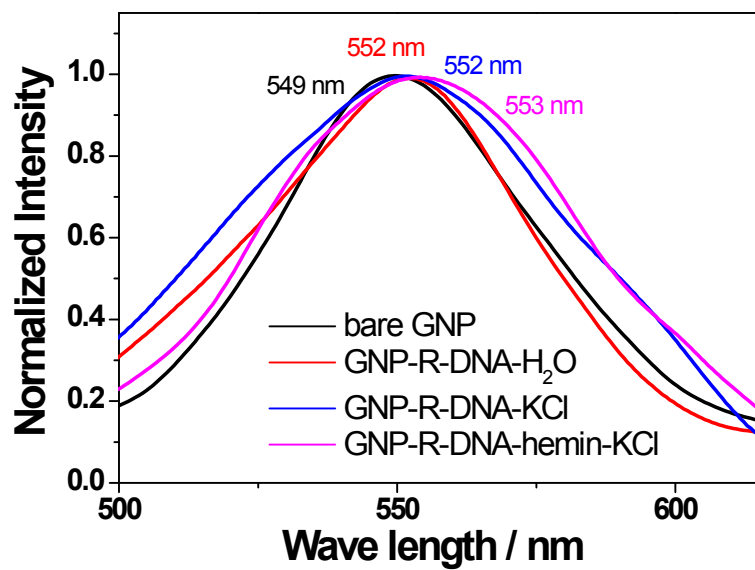


Figure S5. The NPPR scattering spectra of the random DNA-modified GNP.

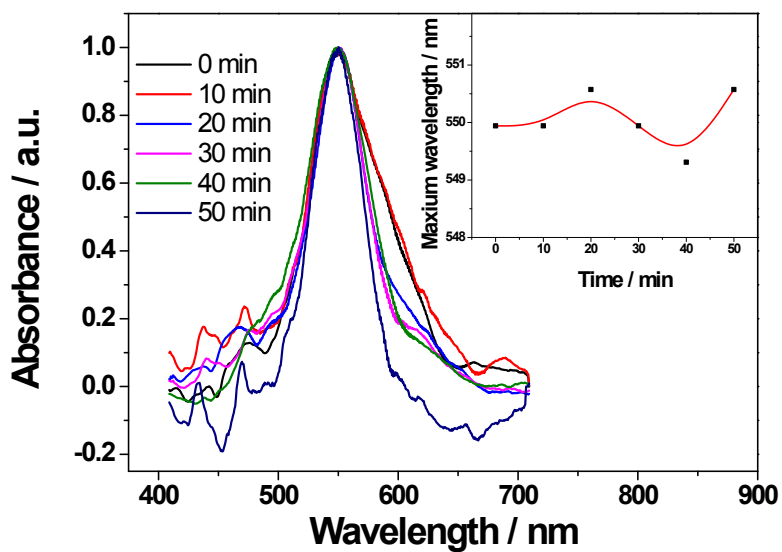


Figure S6. The NPPR scattering spectra of GNP-R-DNA in 0.1 mM DAB and 100 μM H₂O₂ solution after the hemin treatment step.

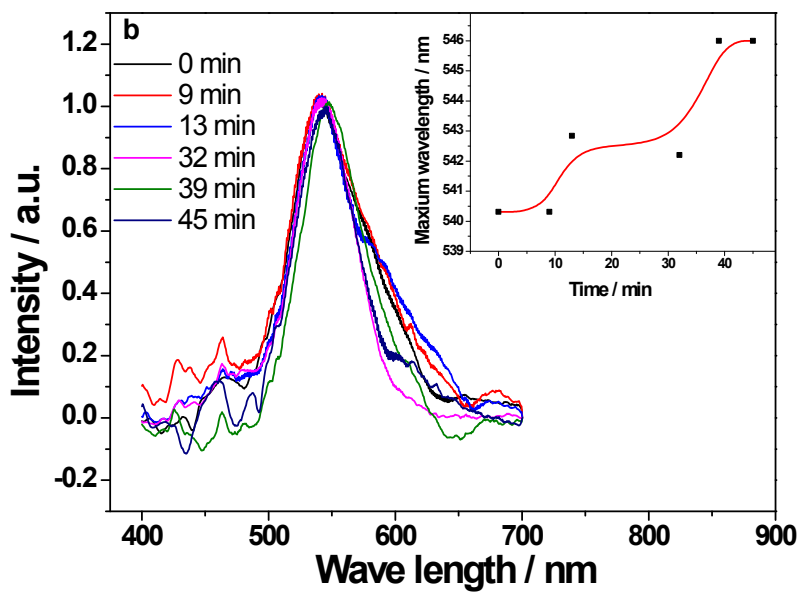
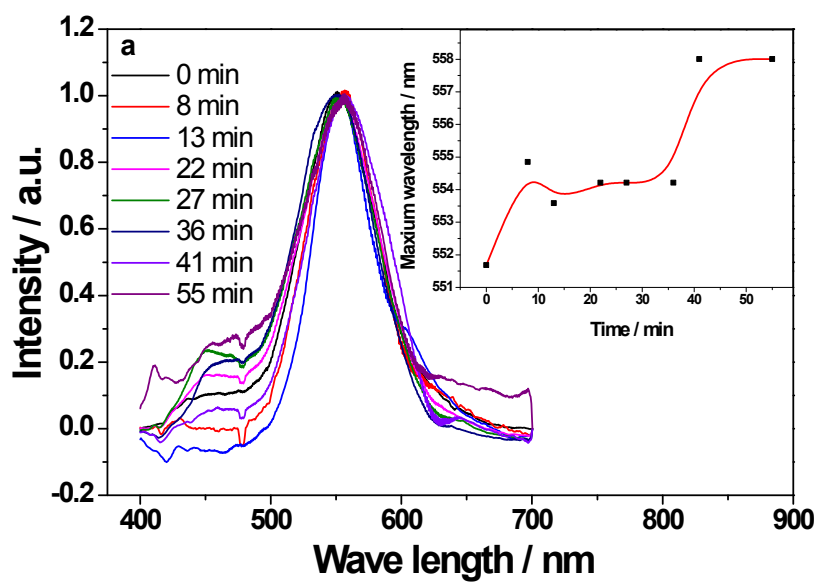


Figure S7. The NPPR scattering spectra of different immobilized GNP-DNAzyme with 0.1 mM DAB in (a) 500 μM H_2O_2 . (b) 100 μM H_2O_2 , respectively.