

## ***Supporting Information***

### **Laser Illumination-Induced Dramatic Catalytic Activity Change on Au Nanospheres**

Zhongju Ye,<sup>a</sup> Lin Wei,<sup>b</sup> Lehui Xiao,<sup>\*a</sup> and Jianfang Wang<sup>c</sup>

<sup>a</sup> State Key Laboratory of Medicinal Chemical Biology, Tianjin Key Laboratory of Biosensing and Molecular Recognition, College of Chemistry, Nankai University, Tianjin, 300071, China

<sup>b</sup> Key Laboratory of Phytochemical R&D of Hunan Province, College of Chemistry and Chemical Engineering, Hunan Normal University, Changsha, 410081, China

<sup>c</sup> Department of Physics, The Chinese University of Hong Kong, Shatin, Hong Kong SAR, China

\* Corresponding author

Email: lehuixiao@nankai.edu.cn

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## Chemicals and Materials

Trisodium citrate, AgNO<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). HAuCl<sub>4</sub>·3H<sub>2</sub>O was obtained from Aladdin (Shanghai, China). 3-aminopropyltriethoxysilane (APTES) was obtained from J&K Scientific Ltd. Chemical Company (Beijing, China). Amplex Red was obtained from Invitrogen Ltd. (Paisley, UK). All other chemicals not mentioned here were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Unless otherwise noted, all experiments were carried out at room temperature and all aqueous solutions were prepared with deionized (DI) water.

## Fabrication of GNPs and Characterizations

The spherical gold nanoparticles (GNPs) used in this experiment with a size of 59.0±1.8 nm were synthesized based on a seed-mediated growth method which is similar to the procedure described before with minor modifications.<sup>1</sup> Herein, 18 nm GNPs were used as the seed for the synthesis of larger size GNPs. Briefly, 1.03 mL of 24.28 mM HAuCl<sub>4</sub> was mixed with 98.97 mL of DI water, and then the mixture was heated to 110 °C. As the temperature reached 110 °C, 10 mL of 1.455 × 10<sup>-2</sup> M trisodium citrate was rapidly injected to the boiling solution. The mixture was vigorously stirred and refluxed for 20 min. The color of the mixture would gradually change from colorless to pale red, pale purple, and finally to the color of wine red. After the color stays the same, the colloidal solution was kept stirring at room temperature until the solution was cooled down. The colloidal solution was stored at 4 °C before use.

Typically, to synthesize larger size GNPs (60 nm), the as-prepared 18 nm GNPs were used as the seed. 10 mL of the seed GNPs solution was injected to a mixture including 40 mL DI water and 30 mL of 0.13 M trisodium citrate. The obtained mixture was then heated to 120 °C and followed by gradually adding 20 mL of 2.35 × 10<sup>-3</sup> M HAuCl<sub>4</sub> to the hot solution for 40 min under gently stirring. As the reaction completed, the solution was kept stirring to cool down to room temperature and stored at 4 °C before use. Ultraviolet-visible (UV-vis) absorption spectra were collected by a Shimadzu UV-2450 spectrophotometer. The size of these GNPs was characterized by transmission electron microscopy (TEM, JEM 1230, JEOL, Japan).

## Elongation and Characterization of GNPs with Controllable Laser Melting

Laser-assisted morphology modulation and characterization of the single gold nanoparticle were done at a home-built imaging system based on a Nikon Ti-U inverted epi-fluorescence microscope (Japan), including dark-field and single-molecule fluorescence imaging systems, as illustrated in Figure 1. Scattering spectrum and polarization characterizations of the nanoparticle before and after elongation were obtained based on the dark-field imaging system. GNPs were firstly injected into a flow channel made by amine-functionalized cover glass. The excess reactants were removed by washing with DI water three times. To modulate the shape of GNPs at single-particle level, the wavelength of the laser (532 nm) was selected to match the UV-vis absorption peak of GNPs. The laser beam (d=2 mm) was expanded around 10× and then reflected into the back port of the microscope. Then it was collected by a 100× oil objective (NA 0.5-1.3) and focused to the sample at the focal plane. The scattered signals generated by the GNPs before and after modulation were collected by the same objective and finally projected onto a true-color complementary metal oxide semiconductor camera (CMOS, Digiretina 16, Tuscan photonics Co., Ltd) that was mounted on the right port of the microscope. Scattering spectra of individual nanoparticles before and after laser melting were measured by a transmission grating beam splitter with 100 lines per mm (Edmund Scientific, Barrington, NJ) that was mounted on the front of the camera. The images were analyzed by Image J (<http://rsbweb.nih.gov/ij/>).

## Single-Nanoparticle Catalysis Experiments

Single-molecule fluorescence measurements were performed on the single-molecule imaging system. The laser beam was reflected by a 565 nm dichroic mirror. The excited fluorescence from the molecules were filtered by a 605/55 nm band-pass filter (Semrock, USA). The fluorescent signals of the resorufin molecules were collected by a 100× oil objective (NA 0.5-1.3) and then captured by an EMCCD (Andor iXon ultra 897). More than 4000 frames were captured to track the catalytic trajectory and the exposure time was set to 50.0 ms. Reactants with different concentrations was injected into the channel in a flow rate at 5  $\mu\text{L}/\text{minute}$ . All the movies were processed and analyzed with matlab and Image J.

Figures

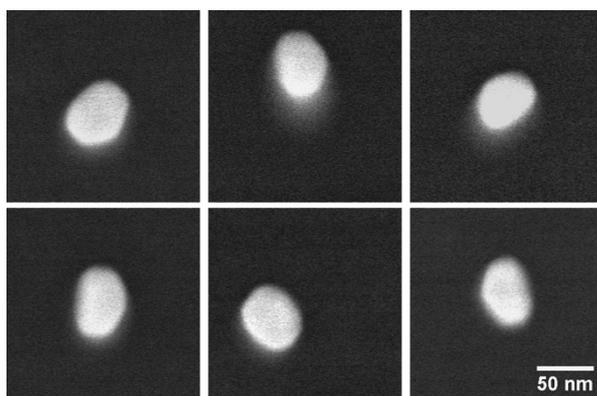


Fig. S1 The SEM results of the particle after laser melting. Scale bar 50 nm.

## References

- 1 L. Wei; Z. J. Ye; Y. L. Xu; B. Chen; E. S. Yeung; L. H. Xiao, *Anal. Chem.* 2016, **88**, 11973.