Giant Resonance Shifts upon Plasmonic Coupling of Complex Nanoscale Geometries

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Nano-plasmonic systems are known for their versatility, modularity, and rapid detection. They have been proposed for use in research fields such as biosensing, electronic switches, and environmental studies. We previously pioneered a novel nanoplasmonic sensing platform based on randomized gold nanomushrooms on the SiO₂ substrate, achieved by a simple gold film deposition and one-step annealing and reactive ion etching process. These nanomushrooms have been applied for *in vitro* detection of biofilms, chemotherapy drugs, and nucleic acids.^{1,2}

In this work, we demonstrate ultrasensitive nanoplasmonic sensing by the formation of gold nanomushroom and gold nanoparticle clusters (**Figure 1a**) to enable local surface plasmon resonance (LSPR). The LSPR coupling efficiency was optimized based on the nanoparticle diameter, ranging from 4 to 200 nm (**Figure 1b**). We found that 40 nm nanoparticles provide the largest resonance shift, and the accompanying spectrum shift is correlated with the gold nanoparticle concentration (**Figure 1c**). Due to the strong coupling, we are able to detect a ~14 nm resonance shift with 100 pM gold nanoparticles, which is far more sensitive than other competitive LSPR systems. This sensitive and concentration dependent coupling was explained by finite-difference time-domain (FDTD) simulations (**Figure 1d**) and will be further used in gold nanoparticle-labeled biochemistry assays for biosensing applications, such as SARS-CoV-2.

¹ Riccardo Funari, Nikhil Bhalla, Kang-Yu Chu, Bill Söderström, and Amy Q. Shen, ACS Sensors **3** (8), 1499 (2018).

² Nikhil Bhalla, Shivani Sathish, Casey J. Galvin, Robert A. Campbell, Abhishek Sinha, and Amy Q. Shen, ACS Applied Materials & Interfaces **10** (1), 219 (2018).



Figure 1. (a) SEM images of the gold nanomushroom-gold nanoparticle clusters. The schematic shows the LSPR coupling. (b) Plasmonic resonance shift vs. nanoparticle diameter. (c) Experimentally measured resonance shift vs. nanoparticle concentration. (d) FDTD simulation for the gold nanomushroom-gold nanoparticle coupling at various concentrations (1-6 nanoparticles per nanomushroom).