Asymmetrical Three Dimensional Plasmonic Nanostructures with Multiple Resonance Modes

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Multiple resonance modes in plasmonic nanostructures provide sharp resonance peaks with high intensity compared to conventional localized surface plasmon resonance (LSPR), which could be used as highly sensitive biosensors. Non-concentric ring/disk cavity, nanoparticle clusters, dolmen nanostructure, and asymmetrical heptamers could produce Fano resonance. These nanostructures resulted in hybrid coupling between superradiant dipole modes and subradiant high order modes in two dimensions (2D) and had limited sensitivity. In this study, novel three dimensional (3D) plasmonic nanostructures composed of nanopillars inside nanoholes to generate three different plasmonic layers are proposed. The nanostructures provide the hybrid coupling effect of LSPR, Fano resonance, and Fabry-Perot cavity modes with high sensitivity.

Quasi 3D plasmonic nanostructures with Au nanoholes on the top and Au nanodots at the bottom showed hybrid plasmonic coupling of the LSPR and Fabry-Perot cavity modes. As shown in Fig. 1, quasi 3D symmetrical plasmonic nanoholes with 300 nm width, 535 nm pitch, and 450 nm depth showed measured multiple resonance peaks and high sensitivity of 218 nm/refractive index unit (RIU). The sensitivity of the quasi 3D plasmonic nanostructures could be further improved by using Au asymmetrical nanoholes as the top layer because of the higher electromagnetic field intensity compared to symmetrical nanostructures. As shown in Fig. 2, quasi 3D asymmetrical plasmonic nanoholes show a higher measured sensitivity of 910 nm/RIU.

To generate sharp resonance peaks with higher sensitivity, 3D plasmonic nanostructures are proposed. Figure 3 (a) shows the schematic diagram of 3D plasmonic nanostructures composed of Au nanosquare as the top layer, Au asymmetrical nanohole as the middle layer, and Au asymmetrical nanoring as the bottom layer. Unlike the non-concentric ring/disk cavities in 2D, these 3D plasmonic nanostructures allow hybrid plasmonic coupling of multiple modes. Simulated extinction spectra of such 3D plasmonic nanostructures is shown in Fig. 3(b). Fano resonance at 706 nm with small full width at half maximum of 6 nm was observed and higher sensitivity of 1075 nm/RIU was achieved at 1033 nm due to the hybrid coupling of the LSPR, Fano resonance, and Fabry-Perot cavity modes.

Reversal nanoimprint lithography will be further developed to fabricate the 3D asymmetrical plasmonic nanostructures. The sharp Fano resonance peak and highly sensitive resonance peak at 1033 nm will be useful for detecting live cancer cells and tumor biomarkers at low concentrations.



Figure 1: (a) Schematic diagram of quasi 3D symmetrical nanoholes. (b) Micrograph of quasi 3D symmetrical nanoholes with 300 nm width, 535 nm pitch, and 450 nm depth. (b) Measured extinction spectra of quasi 3D symmetrical nanoholes.



Figure 2: (a) Micrograph of quasi 3D asymmetrical nanoholes with 300 nm linewidth, 535 nm pitch, and 450 nm depth. (b) Measured extinction spectra of quasi 3D asymmetrical nanoholes.



Figure 3: (a) Schematic diagram of 3D plasmonic nanostructures composed of Au nanosquare as top layer, Au asymmetrical nanohole as middle layer, and Au asymmetrical nanoring as bottom layer. (b) Simulated extinction spectra of 3D plasmonic nanostructures.