Investigation of Quantum Tunneling Effects in Gap Plasmons Using Collapsible Nano-Fingers

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Plasmonic nanostructures have recently received attention because of their ability to concentrate light to small volumes. This property gives rise to numerous potential applications in optical communication, disease diagnosis, and chemical sensing. Therefore, it is important to investigate nano-plasmonic hot spots both theoretically and experimentally. While it has been theoretically predicted that the optimal hot spot is a sub-5 nm gap between two metallic particles, due to the difficulties in fabrication of sub-5 nm structures, most of the studies on hot spots at that scale have either been theoretical only or based on ensembles of multiple hot spots, i.e. dense-packed nano-particles separated by spacers, which do not necessarily behave the same as each individual hot spot. Moreover, the optimal gap size is a result of competition between classical electromagnetic interaction and electron tunneling inside gap plasmonic nanostructures. However, little is known about how electron tunneling strength can be modified by gap materials, which directly decide the tunneling barrier height.

Last year, we have reported the successful fabrication of sub-5 nm gap plasmonic structures with precisely controlled nano-gaps by using collapsible nano-fingers [1]. The basic schematic is shown in Figure 1. The gap size is well defined by twice the thickness of the ALD dielectric layer. Now we have improved the fabrication procedure, so that we can have more controllable finger collapse driven by capillary forces. In addition, we used mother mold made by interference lithography to fabricate large area finger dimer pairs. The aspect ratio and straightness of the nano-fingers' sidewall determine the success of collapse process. Low aspect ratio or tapered sidewall will prevent the fingers from collapsing, while fingers with too high aspect ratios result in a random collapse. Figure 2 shows SEM images of the collapsible nano-fingers before and after collapse. The nano-fingers collapsed uniformly in a dimer configuration.

After realizing well-controlled finger collapse as well as a high collapse rate over large areas, we successfully tuned the tunneling barriers for electrons by using different ALD dielectric material to cover the nano-fingers. As shown in Figure 4, the absorption spectra of the collapsed nano-fingers using TiO_2 as gap material have been characterized, revealing how tunneling can tune the plasmonic enhancement at the hot spot. The optimal gap size is determined according to the strongest absorption peak. The peak absorption strength and resonant wavelengths of gap plasmon with TiO_2, WO_3, and SiO_2 spacers are compared, shown in Figure 3 and Figure 5. The corresponding tunneling barrier heights are 0.89 eV, 1.65 eV, and 4.20 eV, respectively. In this way, we realized direct control of tunneling strength by simply changing the material between metallic nanoparticles. Lower tunneling barrier heights result in a larger optimal gap sizes (Figure 3). Moreover, in contrary to classical theory, a redshift of the plasmon frequency with increasing gap size is observed when it narrows to sub-5 nm range (Figure 5).

We propose a simple, phenomenological model to describe the electric field enhancement at the gap center point between two nanoparticles for different separation distances in order to interpret and correlate the experimental observations with quantum based model incorporating electron tunneling and nonlocality, which can ultimately guide the effective design of potential applications.

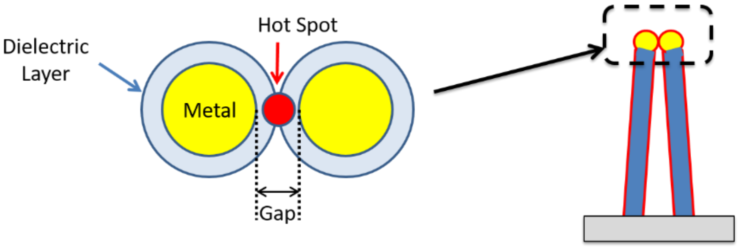


Figure 1. Schematic of controllable hot spots created using nano-fingers.

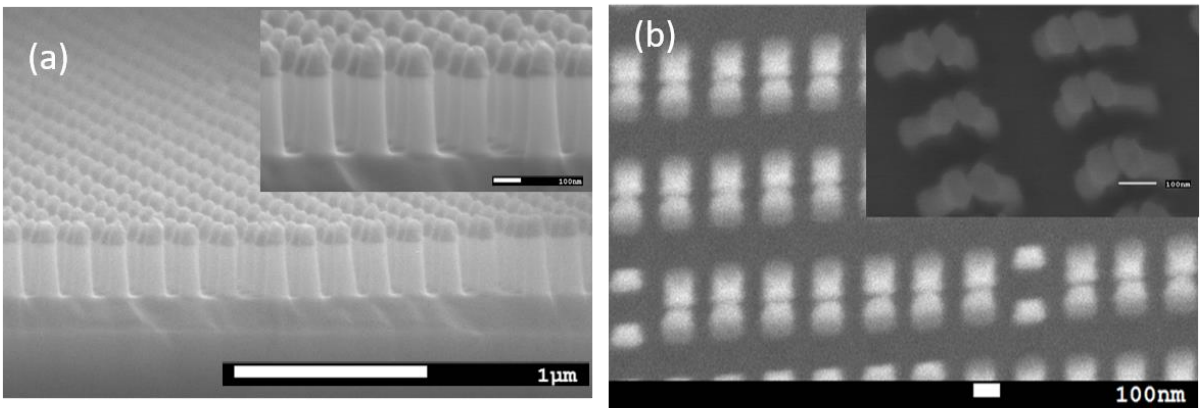


Figure 2. SEM images of collapsible nano-fingers in large area: (a) before collapse (b) after collapse (The ALD TiO_2 layer is about 2 nm thick, which cannot be observed under SEM)

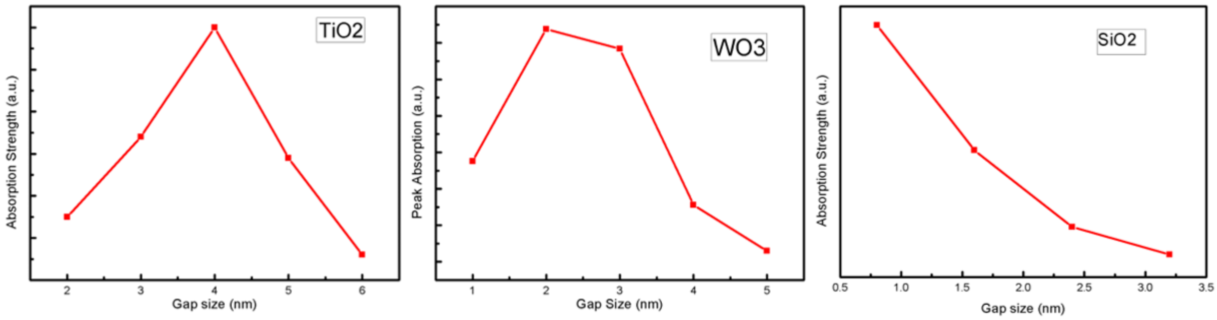
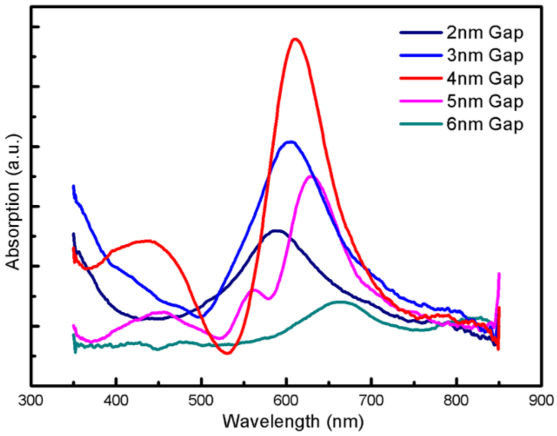


Figure 3. Peak absorption strength vs. gap size for TiO_2, WO_3 and SiO_2.



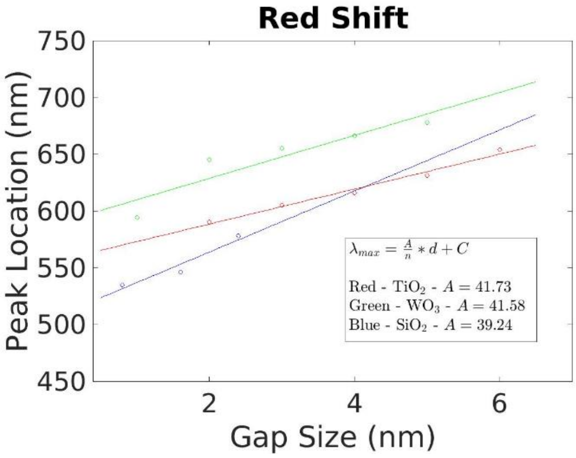


Figure 4 (Left). Absorption spectrum of collapsible nano-fingers using TiO_2 as gap material
Figure 5 (Right). Resonance frequency vs. gap size for different gap material (experiment data points and theoretical fitting)

References

[1] B. Song, Y. Yao, Y. Wang, H. Liu, Y. Li, S. Cabrini, A. Schwartzberg, D. Olynick, S. Cronin, W. Wu, The 60th International Conference on Electron, Ion, and Photon Beam Technology and nanofabrication