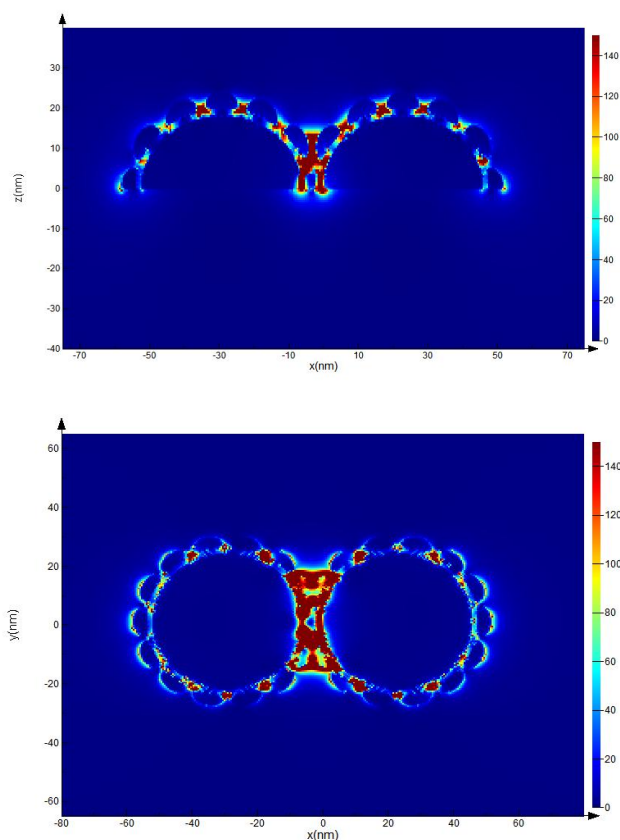


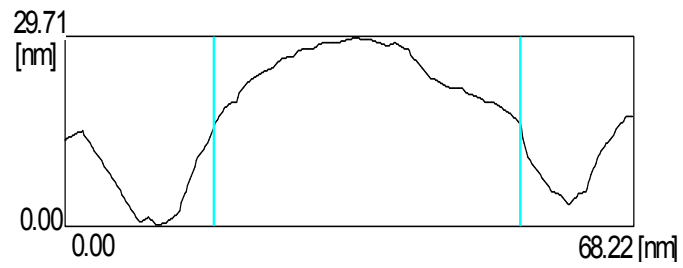
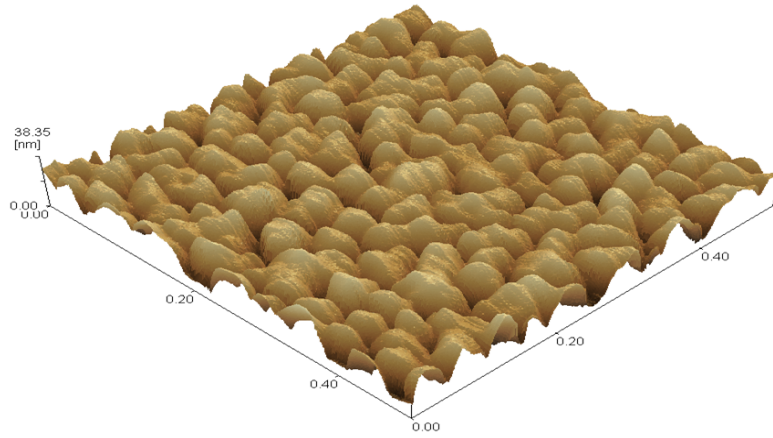
Supplementary Information for  
“**Tunable Plasmon Resonance and Enhanced Second Harmonic  
Generation and Upconverted Fluorescence of Hemispheric-Like  
Silver Core/Shell Islands**”

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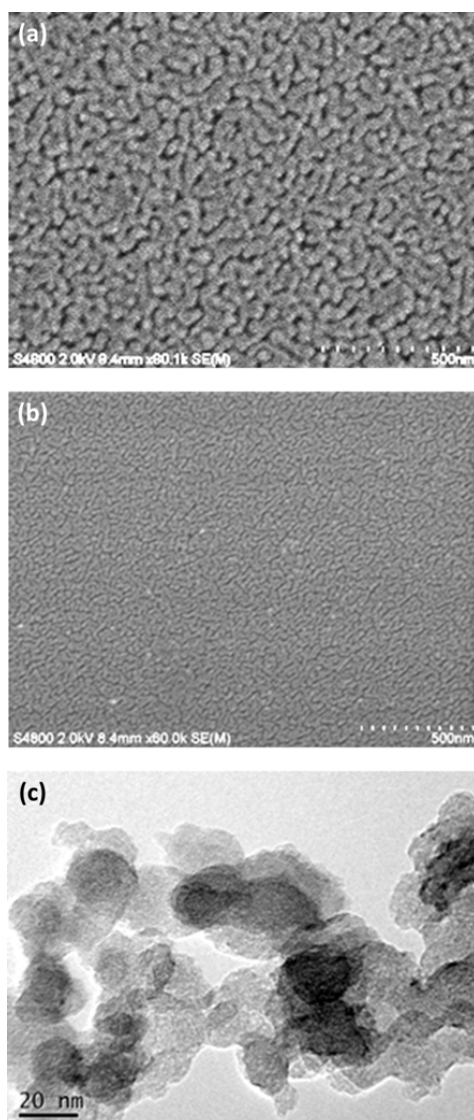
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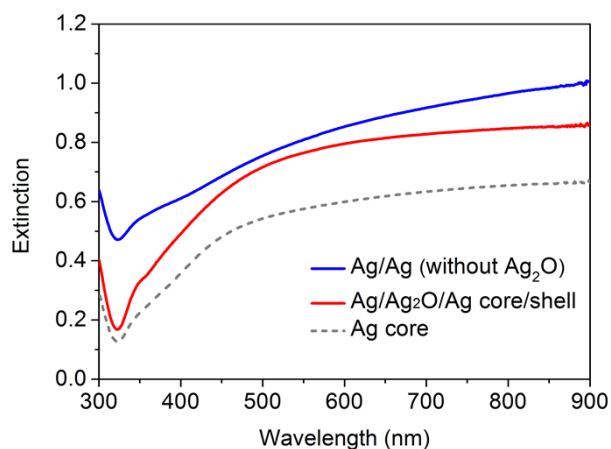
**Figure S1.** Local field enhancements of Ag/Ag<sub>2</sub>O/Ag core/shell islands with two coupled cores at the plasmon resonance wavelength. It demonstrates stronger local field enhancement in the gap of two silver cores.



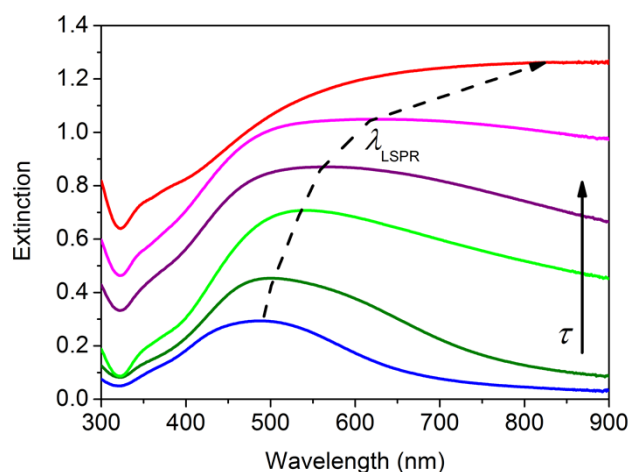
**Figure S2.** 3D AFM image of silver island films near the percolation (lateral sizes:  $500 \times 500 \text{ nm}^2$ ) and cross-sectional height profile of silver islands. The hemispheric-like geometry of the islands is demonstrated and the percolating aggregation of the islands is also observed.



**Figure S3.** (a) SEM image of the near percolating silver islands sputtered by using SBC-12 system, which have larger granular sizes and are used as core islands. (b) SEM image of the near percolating silver islands sputtered by using K575XP system, which have smaller granular sizes and are used as shell islands. (c) TEM image of the Ag/Ag<sub>2</sub>O/Ag core/shell island films.



**Figure S4.** Extinction spectra of the near percolating Ag core ( $\tau_{\text{core}} = 1.03$ , grey line), Ag/Ag<sub>2</sub>O/Ag core/shell (red line), and reference Ag/Ag without Ag<sub>2</sub>O (blue line) island films. Ag/Ag<sub>2</sub>O/Ag islands also demonstrate optical features similar to Ag core sample owing to the percolating effect, but the reference sample Ag/Ag without Ag<sub>2</sub>O separation layer exhibits behavior of conductive film (the extinction significantly increases with wavelength in the near infrared regime).



**Figure S5.** Extinction spectra of the silver island films prepared with different deposition time ( $\tau_{\text{core}}$ ). The central LSPR peak ( $\lambda_{\text{LSPR}}$ ) red-shifts increases from  $\sim 480$  nm to longer than 800 nm with increase of  $\tau_{\text{core}}$ .