

## Supporting Information

# Growth of Metal-Semiconductor Core-Multishell Nanorods with Optimized Field Confinement and Nonlinear Enhancement

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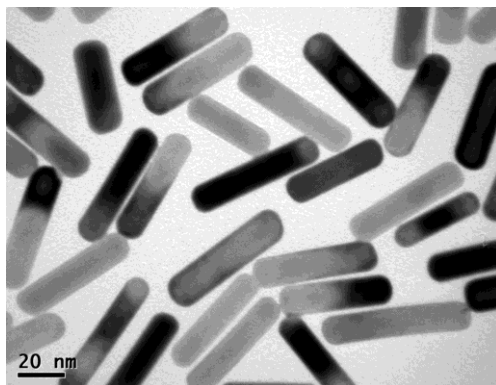
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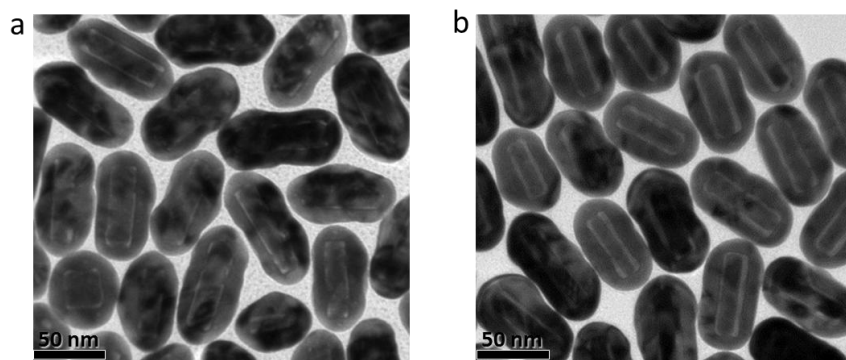
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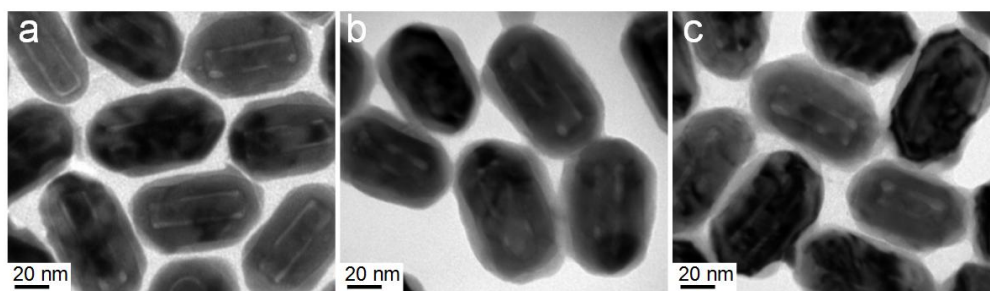
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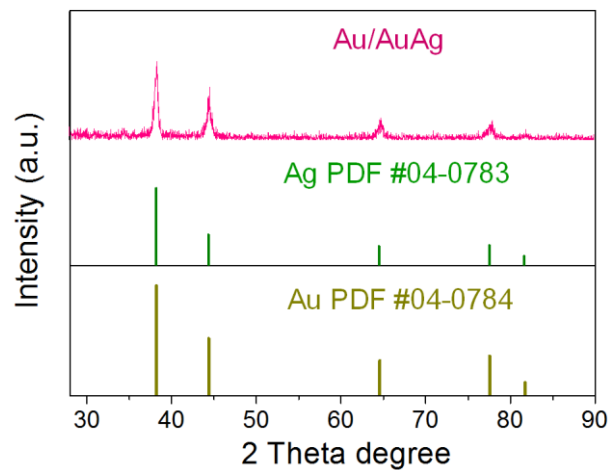
**Figure S1.** TEM image of the Au nanorods.



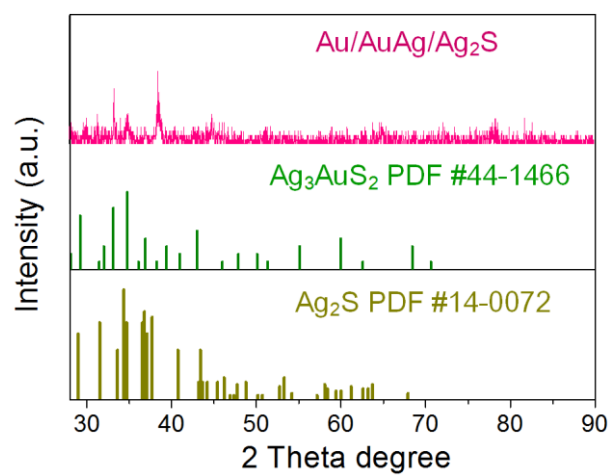
**Figure S2.** TEM images of Au/AuAg NRs with different gaps distances.



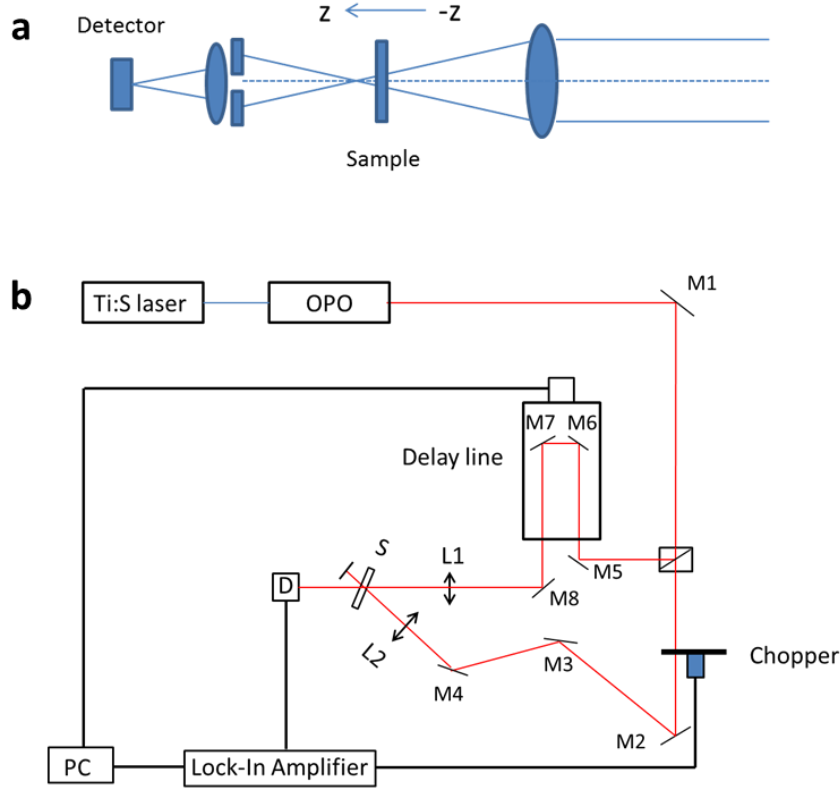
**Figure S3.** TEM images of Au/AuAg/Ag<sub>2</sub>S NRs with different Ag<sub>2</sub>S thicknesses.



**Figure S4.** XRD patterns of Au/AuAg, Au (PDF #04-0784) and Ag (PDF #04-0783).



**Figure S5.** XRD pattern of Au/AuAg/Ag<sub>2</sub>S and the standard patterns of Ag<sub>2</sub>S (PDF #14-0072) and Au<sub>3</sub>AuS<sub>2</sub> (PDF #44-1466).



**Figure S6.** Experimental set-up of Z-scan (a) and Pump-probe measurement (b).

The effective NLA coefficient  $\beta_{\text{eff}}$  and the NLR index  $\gamma$  can be calculated by the following relationships:

$$T_{\text{OA}} = \sum_{m=0}^{\infty} \frac{(-q_0)^m}{(1 + z^2/z_0^2)^m (1+m)^{3/2}} \quad (1)$$

$$\frac{T_{\text{CA}}}{T_{\text{OA}}} = 1 + \frac{4\Delta\phi_0 z/z_0}{[(z/z_0)^2 + 9][(z/z_0)^2 + 1]} \quad (2)$$

where  $q_0 = \beta_{\text{eff}} I_0 L_{\text{eff}}$  and  $\Delta\phi_0 = \gamma k I_0 L_{\text{eff}}$ .  $I_0$  is the peak irradiance at the focus ( $z=0$ ).  $L_{\text{eff}}$  is the effective thickness of the samples,  $k=2\pi/\lambda$  is the wave vector of the laser radiation,  $z_0$  is the Rayleigh length of the Gaussian incident beam.