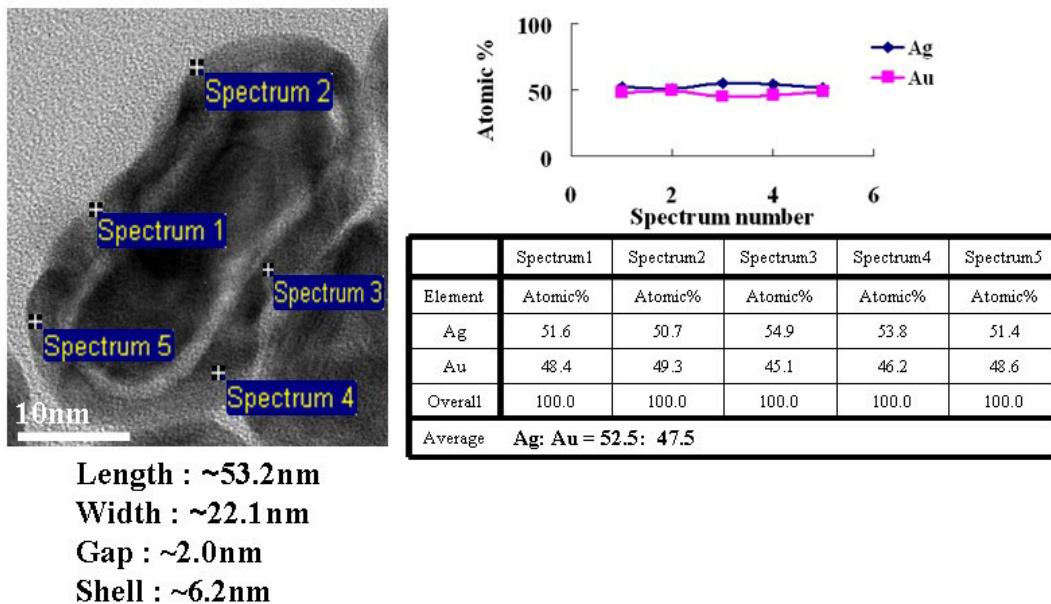


*Supplementary Materials*

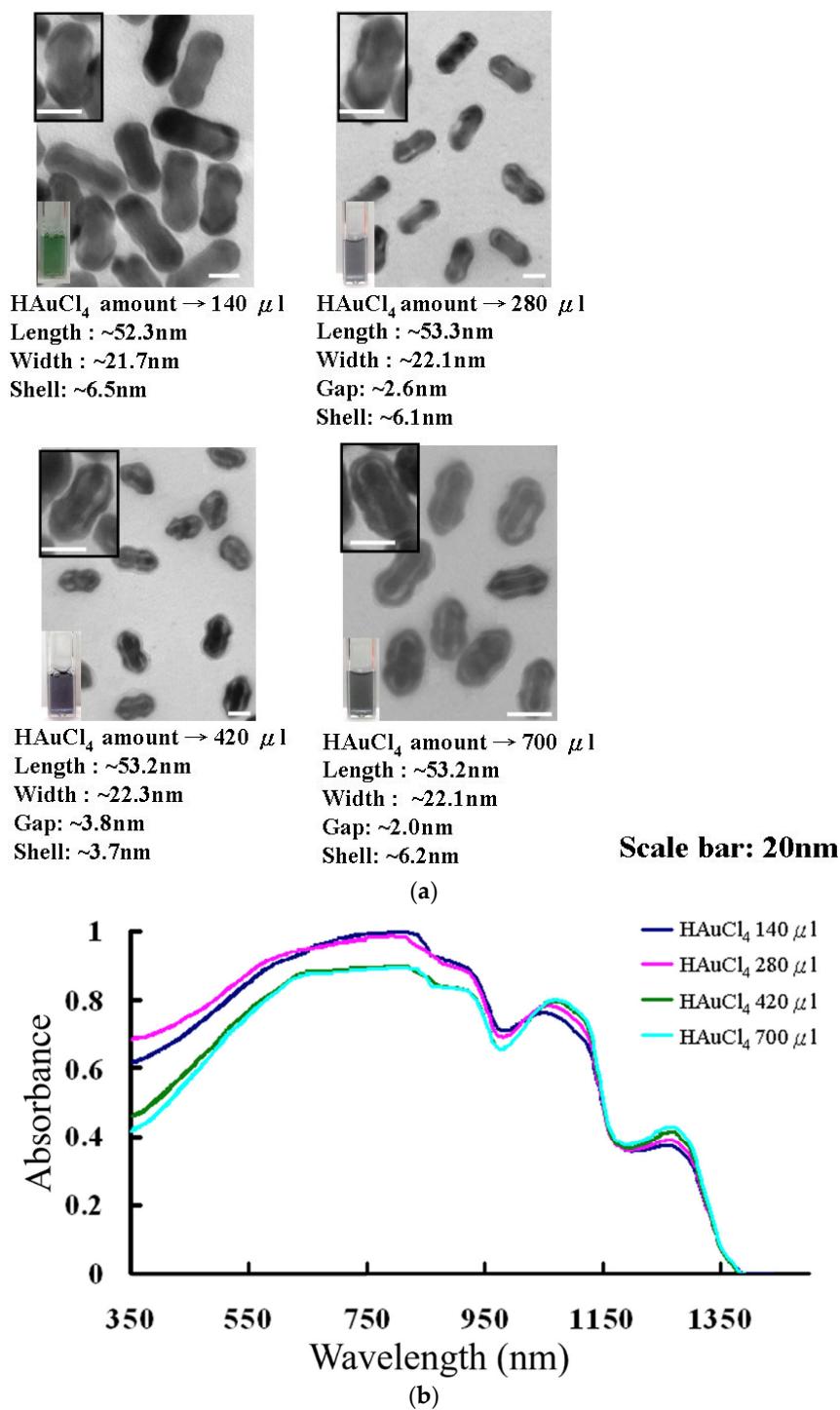
# Plasmon Resonant Two-Photon Luminescence Inducing Photosensitization and Nonlinear Optical Microscopy In Vivo by Near-Infrared Excitation of Au Nanopeanuts

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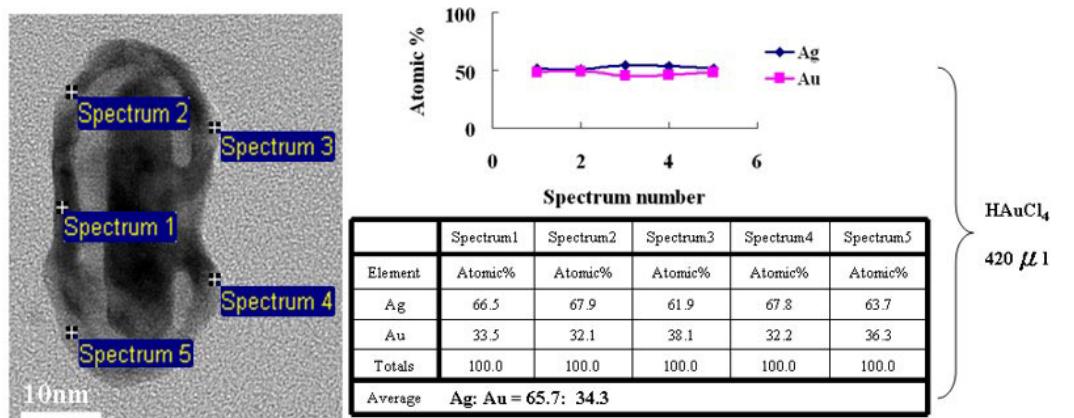
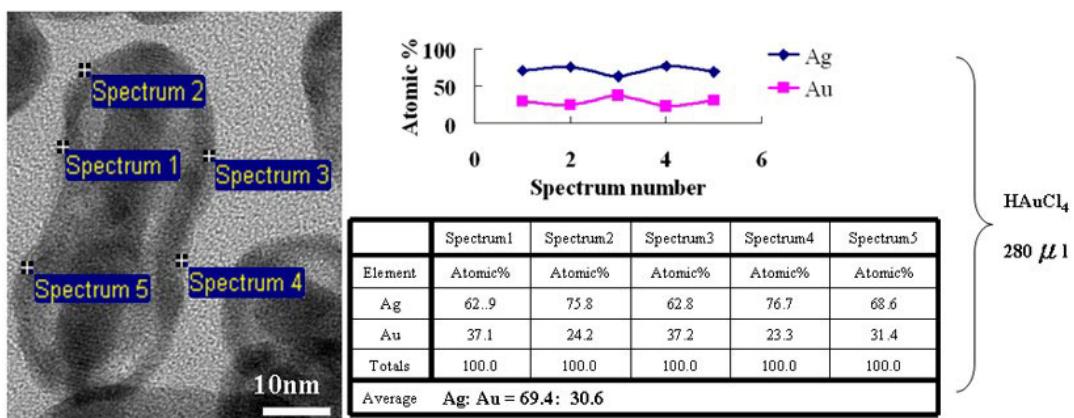
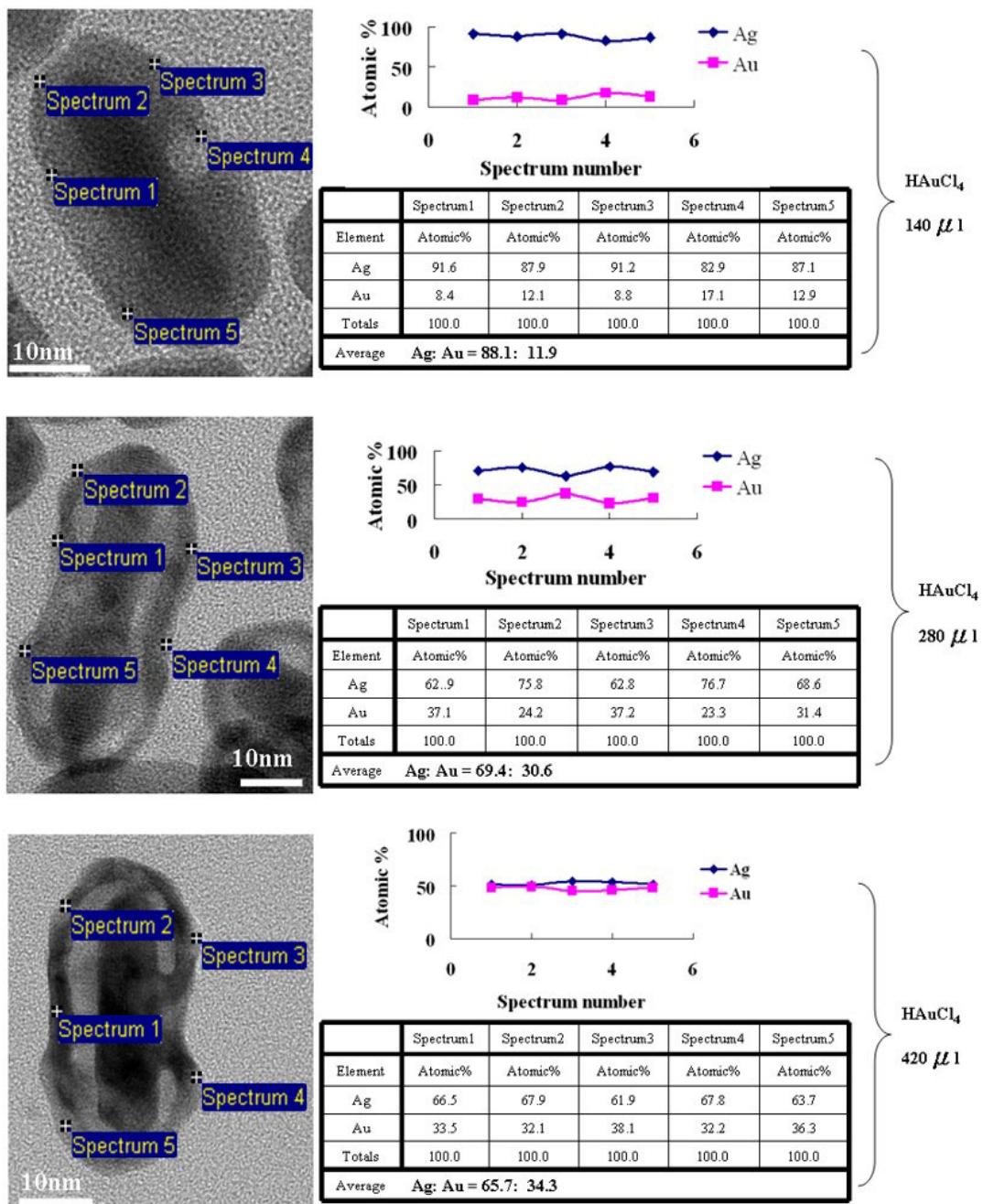
nanopeanuts obtained by adding different volumes (140, 280, and 420  $\mu\text{L}$ ) of HAuCl<sub>4</sub> into Au NR@Ag colloidal solutions containing CTAB and AA. Spectra 1~5 shown for each nanopeanut represent different spot sites analyzed in the Ag/Au shell.



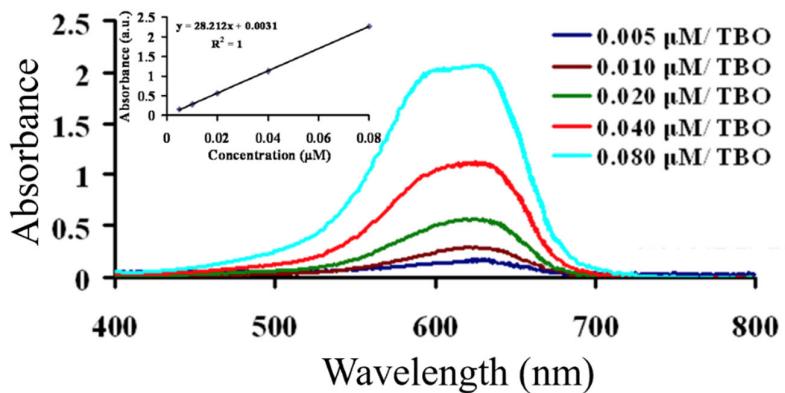
**Figure S1.** The high-resolution transmission electron microscope (HRTEM) image and energy dispersive X-ray (EDX) analysis of Au nanopeanuts. Spectra 1-5 represent different sites analyzed in the Ag/Au nanoshell. Au nanopeanuts were prepared by adding 700  $\mu\text{L}$  of HAuCl<sub>4</sub> into Au NR@Ag colloidal solution containing CTAB and AA..



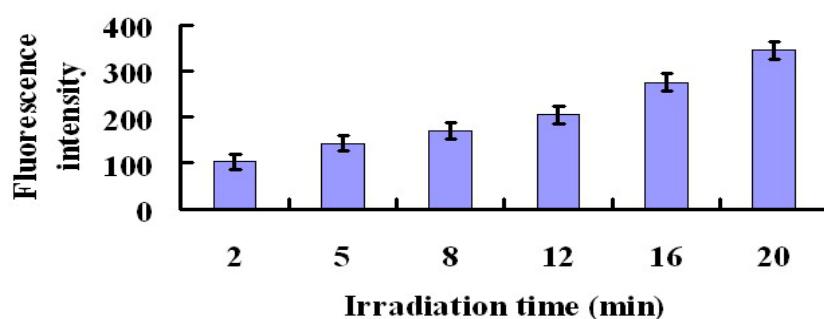
**Figure S2.** (a) TEM images and (b) UV-Vis-NIR spectra of Au nanopeanuts obtained from the addition of different volumes (140, 280, 420, and 700  $\mu\text{L}$ ) of HAuCl<sub>4</sub> into Au NR@Ag colloidal solutions containing CTAB and AA. Insets in each figure present the corresponding colloidal color and the amplified TEM of the single nanopeanut.



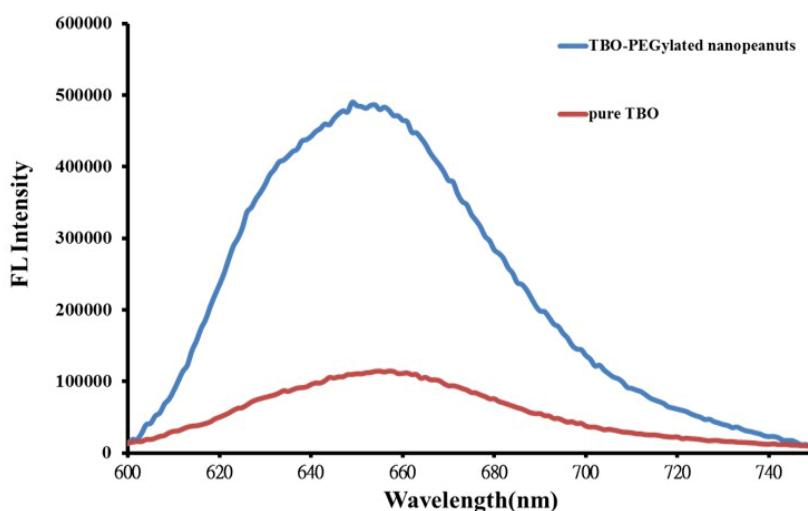
**Figure S3.** HRTEM images and EDX analysis of Au nanopeanuts obtained by adding different volumes (140, 280, and 420  $\mu$ L) of HAuCl<sub>4</sub> into Au NR@Ag colloidal solutions containing CTAB and AA. Spectra 1~5 shown for each nanopeanut represent different spot sites analyzed in the Ag/Au shell.



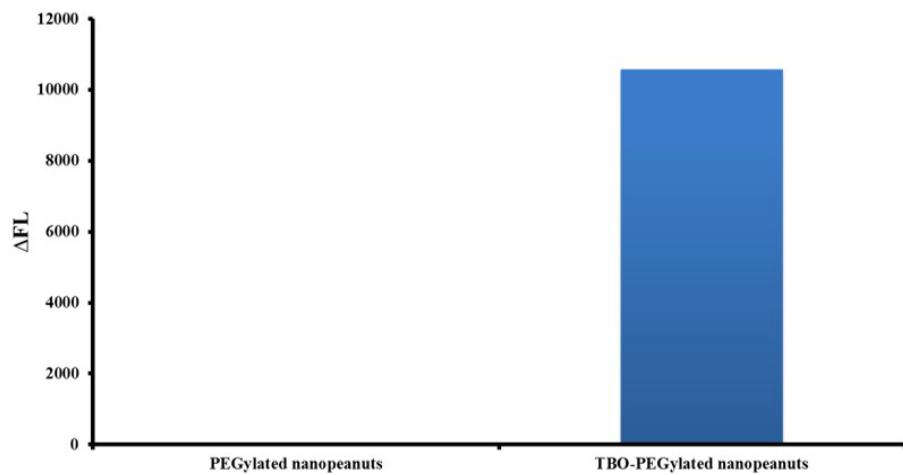
**Figure S4.** UV-Vis spectra taken as a function of TBO concentration. The inset shows the standard linear calibration curve of TBO.



**Figure S5.** Fluorescence intensity of Singlet Oxygen Sensor Green mixed with TBO-PEGylated nanopeanuts (20 μg/mL) exposed to a 633-nm (200 mW/cm<sup>2</sup>) diode laser for different irradiation times.



**Figure S6.** Fluorescence intensity of TBO-PEGylated nanopeanuts and free TBO measured from a conventional HORIBA Fluoromax-4 spectrometer by selection of excitation wavelength 580 nm of xenon arc-lamp. The TBO concentration was fixed at 0.23 μM for both TBO-PEGylated nanopeanuts and free TBO molecules



**Figure S7.** Fluorescence intensity of Singlet Oxygen Sensor Green mixed with nanopeanuts ( $20 \mu\text{g/mL}$ ) exposed to 1230 nm femtosecond Cr: forsterite laser ( $104 \text{ mW/cm}^2$ ) for 3 min.