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## Supporting Information



**Figure S1.** TEM images of silica-coated AgNS (AgNS@SiO<sub>2</sub>): (a) 4-BBT labeled (AgNS<sub>4-BBT</sub>@SiO<sub>2</sub>), and (b) 4-CBT labeled (AgNS<sub>4-CBT</sub>@SiO<sub>2</sub>).



**Figure S2.** PA spectra of different kinds of nanoparticles (AgNS, AuNP and AuNR, 150 pM each) under various laser wavelength (680-900 nm) with D.I. water as a control.



**Figure S3.** (a) SERS spectra of different kinds of nanoparticles (AgNS, AuNP and AuNR, 150 pM each), labeled with 4-FBT (1 mM). (b) SERS intensity profile (386 cm<sup>-1</sup> of 4-FBT) of each nanoparticle without normalization. SERS spectra were obtained using portable-Raman system by 785-nm photoexcitation, 30-mW laser power, and light acquisition time of 5 s



**Figure S4.** Full-range SERS spectra of  $AgNS_{4-FBT}$ @SiO<sub>2</sub> (green line),  $AgNS_{4-BBT}$ @SiO<sub>2</sub> (blue line) and  $AgNS_{4-CBT}$ @SiO<sub>2</sub> (red line). SERS spectra were obtained using portable-Raman system by 785-nm photoexcitation, 15-mW laser power, and light acquisition time of 3 s



**Figure S5.** PA spectra of AgNS@SiO<sub>2</sub> at various concentrations under different laser wavelength. The range of wavelength is 680-950 nm, the concentration of AgNS@SiO<sub>2</sub> is from 1000 pM to 15.8 pM, and 1% BSA solution in PBS as a control.



Figure S6. Relative SERS intensity profile of  $AgNS@SiO_2$  with incubation time at various conditions for stability of our nanoprobes. 1075 cm<sup>-1</sup> of 4-FBT peak were used for analysis of this data.



Figure S7. Size and distribution of each AgNS@SiO<sub>2</sub> measuring by nanoparticle tracking analysis (NTA)



**Figure S8.** (a) PA spectrum with various concentration of AgNS@SiO<sub>2</sub> for showing PA sensitivity in vivo skin of rat. (b) SERS intensity about 1075 cm<sup>-1</sup> of 4-FBT with various concentration of AgNS<sub>4-FBT</sub>@SiO<sub>2</sub> for showing SERS sensitivity in vivo skin of rat. (c) SERS spectrum with various concentration of AgNS<sub>4-FBT</sub>@SiO<sub>2</sub>. SERS spectra were obtained using portable-Raman system by 785-nm photoexcitation, 60-mW laser power, and light acquisition time of 10 s



**Figure S9.** *In vivo* PA detection of axillary SLN of a rat with AgNS@SiO<sub>2</sub> mixture (1:1:1 ratio, 1 nM, 100  $\mu$ L). (a) Control PA MAP image of a rat's left axillary region. (b) Post-injection PA MAP image of a rat's left thigh after 40 min, (c) 80 min, (d) 120 min, and (e) 160 min of injections.



**Figure S10.** *In vivo* 3D PA images of axillary of a rat with AgNS@SiO<sub>2</sub> mixture (1:1:1 ratio, 1 nM, 100  $\mu$ L). (a) Control PA MAP 3D image of a rat's left axillary region. (b) Post-injection PA MAP 3D image of a rat's left thigh after injection (30 min). (c) Differences of PA SLN 3D images between (a) and (b).



**Figure S11.** *In vivo* detection of axillary SLN of a rat with AgNS@SiO<sub>2</sub> mixture (ratio: 1:3:5, 1 nM, 100  $\mu$ L). (a) Control PA MAP image of a rat's left axillary region. (b) Post-injection PA MAP image of a rat's left thigh after injection. (c) Photograph of control and the injected SLN. (d) *Ex vivo* PA image of control and the injected SLN (Bottom)