Supporting Information for:

Gold Suprashells: Enhanced Photothermal Nanoheaters with Multiple LSPR for Broadband SERS

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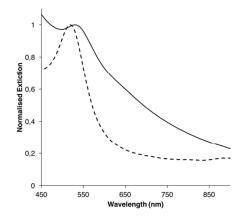
Figure S1. Extinction spectrum of the 20 nm gold nanoparticles building blocks used here to assemble gold suprashells (doted lines, (λ_{max} = 519 nm)), and G1 suprashells (continuous line).
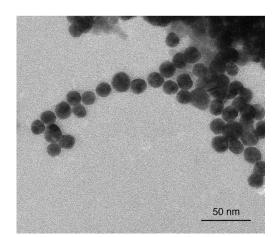

Figure S2. Representative TEM image of anisotropic assemblies of nanoparticles in gold suprashells (G3). The inter-particle distance was calculated as the average distance between 150 nanoparticle pairs measured in several images.
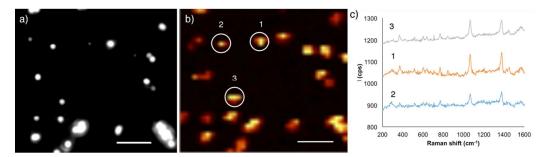
Figure S3. SERS mapping of suprashells modified with 2-naphthalenethiol; a), b) Correlated dark-field microscopy (DFM) image and SERS map of G5 suprashells adsorbed onto a glass slide; c) Representative SERS spectra of single suprashells (highlighted with a circle in (b)). SERS images were generated from analysis of the peak intensity at ~1064 cm^{-1} with respect to the background signal with the brightest points corresponding to intensities >300 counts/s (cps). Scale bars: 5 μm.

Experimental: Glass slides were cleaned by ultrasonic treatment in acetone for 5 min followed by rinsing with ethanol and deionized water, and drying with nitrogen. The slides were then immersed in Hellmanex^{TM} for at least 1 h, rinsed with abundant water and dried with nitrogen. Subsequently the slides were immersed in a 1% (v/v) solution of poly(diallyldimethylammonium chloride) (PDDA) for 30 min. The slides were then rinsed with deionized water and dried with nitrogen. 100 μL of the suprashell dispersion was placed onto the substrates for 10 min. The nanoparticles were adsorbed onto the PDDA-covered slide due to the electrostatic interaction between the citrate-capped nanoparticles and positively charged glass surface. The slides were then covered with 1 μM 2-naphtalenethiol solution for 10 minutes, rinsed with water and dried with nitrogen. Correlated dark-field and SERS imaging was performed on two different microscopes with reference marks on the slide surface used to identify different regions. Raman maps were obtained using a confocal WITec Alpha300R instrument at 633 nm excitation. All maps were acquired using a 100× objective (Olympus MPlan, NA = 0.9). Areas up to 20 × 20 μm in size were imaged in ~0.6 μm steps. An incident laser power of ~0.9 mW, spot size ~0.6 μm and signal integration time of 1 s was used. The SERS maps were created by plotting the difference between the maximum and minimum intensities in the 980-1100 cm^{-1} window, targeting the peak at 1064 cm^{-1} and a preceding background region of the spectrum.
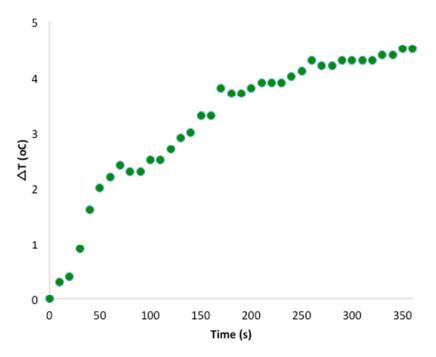
Figure S4. Increase in temperature with time when exciting G5 gold suprashells with a 514 nm CW laser (27 mW).
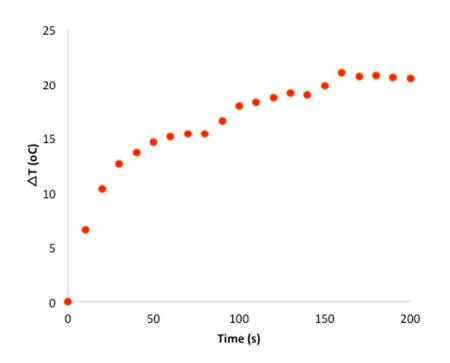

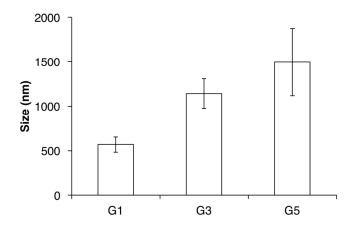**Figure S5.** Increase in temperature with time when exciting G5 gold suprashells with a 785 nm CW laser (235 mW).

Figure S6. Size distribution of G1, G3 and G5 suprashells obtained with dynamic light scattering (DLS). Error bars are the standard deviation.
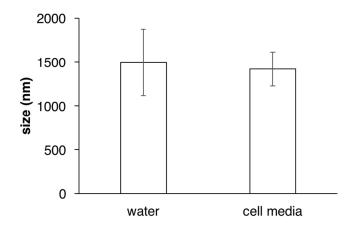

Figure S7. DLS size distribution of G5 dispersed in water or cell media. Error bars are the standard deviation.