Supplementary Information

Laser-Assisted Photothermal Heating of a Plasmonic Nanoparticle-Suspended Droplet in a Microchannel

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1. Finite Element Analysis of Microdroplet Temperature in a Microchannel

Upon the calculation of the spectral absorptance distribution, the total volumetric photothermal absorption rate can be calculated by $q'''(z) = \frac{d}{dz}\left[\int_{\lambda_{\min}}^{\lambda_{\max}} \tau A_\lambda(z) P_\lambda d\lambda\right]$, where τ is the transmittance of the PDMS microchannel cover, and P_λ is the spectral laser power density. The laser power density was assumed to be wavelength-dependent with Gaussian distribution that has a full-width at half maximum of 3 nm. In order to calculate the temperature distribution of a microdroplet-in-microchannel system, the present study numerically solved the heat transfer equation using a commercial finite element analysis package (COMSOL Multiphysics). The FEA domain is illustrated in **Figure S1a**, where a nanoparticle-suspended droplet is placed in the center and mineral oil is filled in the remaining of the PDMS microchannel. The heat transfer equation can be written as

$$\rho c \frac{\partial T(\hat{\mathbf{r}}, t)}{\partial t} = \nabla \cdot (k \nabla T) + q'''(\hat{\mathbf{r}}) \qquad \text{* MERGEFORMAT (2)}$$

Where ρ is the density, c is the specific heat, and k is the thermal conductivity. It should be noted that a microdroplet was assumed to be stationary and natural convection within the microdroplet was not considered, as the Rayleigh number was found to be $\ll 1$. Since the volume concentration of nanoparticles under consideration is very low (i.e., less than 1 %), thermophysical properties of the nanofluid droplet were assumed to be the same as those of pure water. **Figure S2** shows the computation results for GNS and GNR suspension cases, demonstrating the capability of photothermal heating in a microchannel due to the enhanced light absorption upon the localized surface plasmon excitation.

2. Video Clip for Bubble Formation in a Microdroplet

The supplemental video 1 shows local bubble formation in a ~30nL GNR PNF droplet of ~20.0 ppm GNR concentration as a result of laser heating at 10x magnification. The GNR/QD solution was not well dispersed, leading to coalescing of vapor and induced flow within the microchannel. The medium surrounding the droplet is mineral oil and the laser power density is ~200 $W\cdot cm^{-2}$.

3. Video Clip for Convection in a Microdroplet

Supplemental video 2 shows local convection within a ~30 nL GNS PNF droplet of ~0.5 ppm GNS concentration as a result of laser heating at 20x magnification. The surrounding GNS particles, as well as the small agglomerations, can be seen being drawn into a circulating column of fluid centered about the laser spot of ~550 $W\cdot cm^{-2}$ power density.

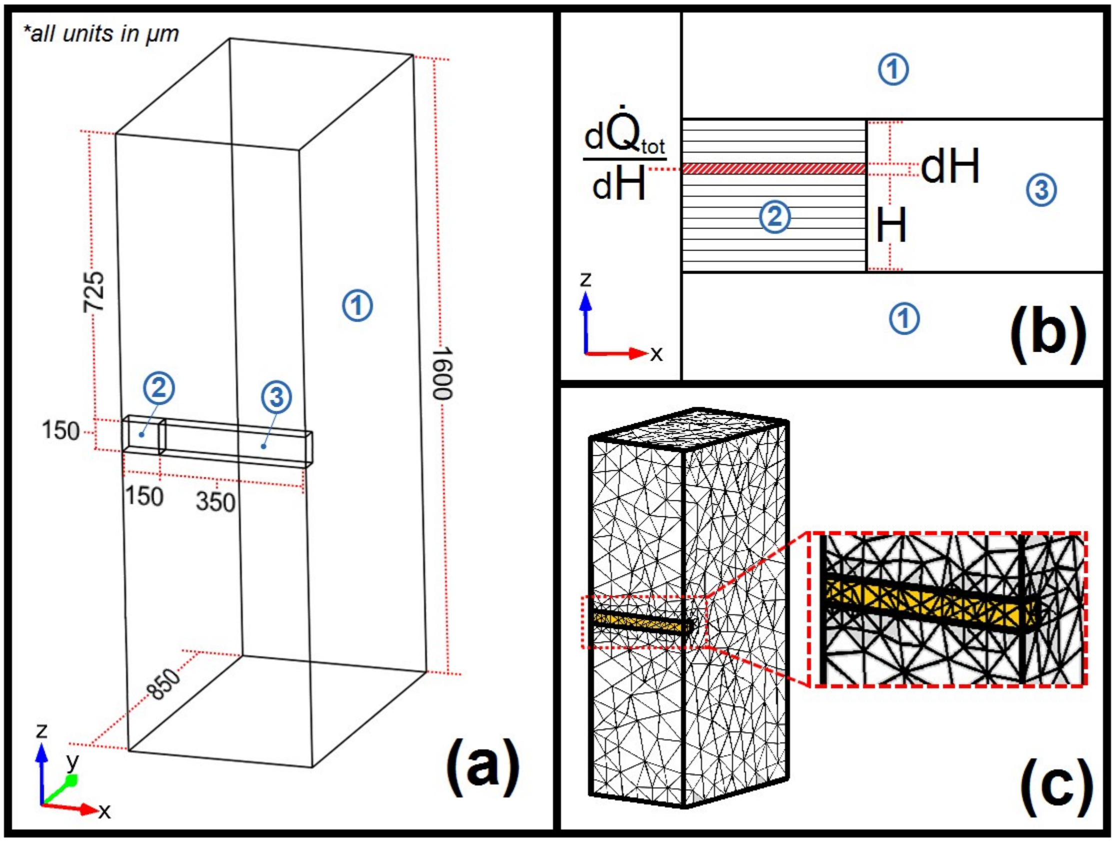

Figure S1. (a) Full FEA model in COMSOL with dimensions and subdomains: PDMS, water, and mineral oil USP labeled 1,2,3 respectively. **(b)** The discretization process of the volumetric heat generation in the microdroplet. **(c)** Fully meshed FEA model in COMSOL: 2807 tetrahedral elements and 106729 degrees of freedom.

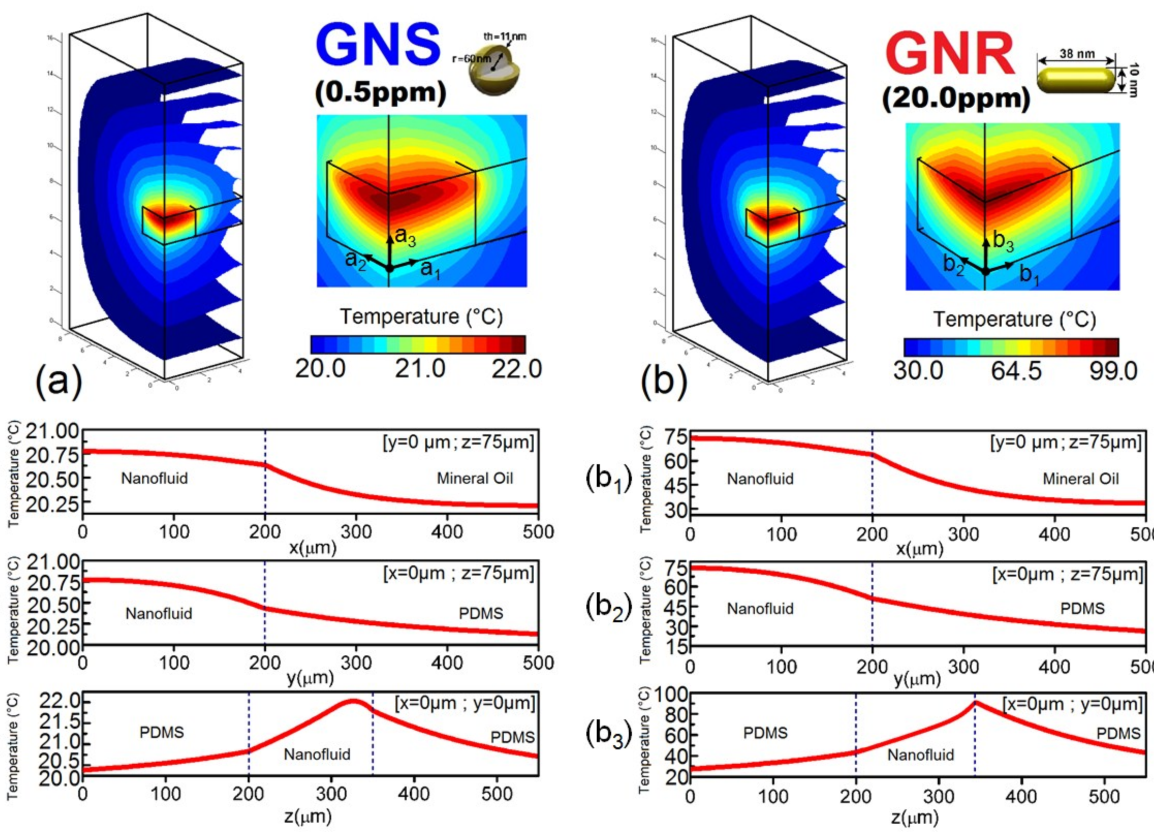

Figure S2. Simulated temperature steady-state distribution under 500W·cm^{-2} and 450W·cm^{-2} respectively for **(a)** 0.5ppm GNS concentration **(b)** 20.0ppm GNR concentration. **Top-to-bottom:** 3D isothermal plot, x-axis temperature profile (a_1,b_1), y-axis temperature profile (a_2,b_2), and z-axis temperature along microchannel center (a_3,b_3).