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SUPPORTING INFORMATION

Thermophoresis of Gold Nanorods from Surface Enhanced Raman Scattering and Real-Time Rayleigh Scattering in Solution

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Figure S1 displays the extinction spectrum of gold nanorod SERS-substrates measured in a cuvette with a path length of 1 cm using an Ocean Optics USB 2000 optical fiber spectrometer. The resonant absorbance occurs at 800 nm. Here, OD refers to optical density.

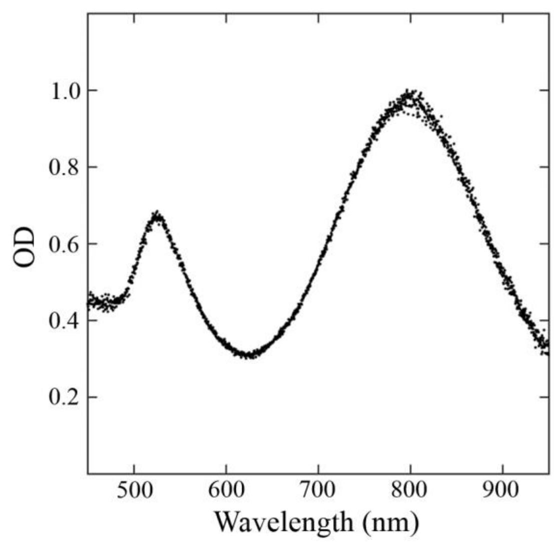

Fig. S1 Extinction spectrum of gold nanorods SERS-substrates.

Figure S2 displays SERS signal decreasing as excitation power is increased, then returning to the original value with no hysteresis when excitation power is decreased.

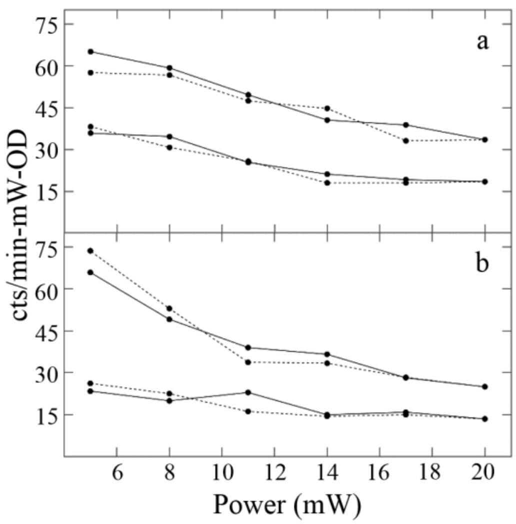

Fig. S2 The SERS intensity decreasing with increasing excitation power, then returning to the original value with no hysteresis for (a) two modes of CTAB and (b) two modes of DOPC. In each case the 1450 cm^{-1} CH_2 scissor mode is the top plot and the 718/760 cm^{-1} symmetric headgroup stretch is the lower plot.

Timescale of Radiation Pressure:

Gucciardi et al. derive the radiation force acting on a nanorod from a Gaussian beam as:

$$F_{rad} = \frac{n_m \sigma_{ext}}{c} I$$

$$I = \frac{2 n_m P}{\pi w_0^2}$$

where P is the incident power, w_0 is the beam waist, n_m is the refractive index of the medium, σ_{ext} is the extinction cross section, and c is the speed of light.[1] We use P = 10 mW which is a typical value for our incident power, w_0 = 1 μm which is the diameter of our scattering volume, and n_m = 1.3296 which is the refractive index of water at 785 nm. We use σ_{ext} = 8.4e-16 m^2 as was measured for gold nanorods which are similar to the ones used here.[2]

We assume no preferential orientation for our nanorods and therefore, model the nanorods (50 nm x 160 nm) as spheres with radius R = 80 nm when calculating drag. The Stokes drag force on a spherical particle in a viscous fluid is:

$$F_{drag} = -6\pi\eta R v$$

where η is the fluid viscosity, R is the particle radius, and v is the particle's velocity. We use η = 8.9e-4 Pa-s which is the viscosity of water at room temperature.

Therefore, the equation of motion $ma = F_{rad} - F_{drag}$ can be rewritten as $\ddot{x} + \frac{6\pi\eta R}{m}\dot{x} = \frac{F_{rad}}{m}$ and solved for *x(t)*. Assuming a nanorod is initially at rest, the time it takes to travel 1 μm under radiation pressure and drag is $O(10^{-2})$ seconds.

Figure S3 displays the effect of the excitation wavelength on Rayleigh scattering for 641 nm (off-resonance) and 785 nm (on-resonance) excitation beams. As seen in Figure S1, absorbance for gold nanorods used here is lower at 641 nm. Thus, there is less localized heating caused by 641 nm excitation compared to 785 nm, leading to a higher steady-state concentration of nanorods in the focal volume. Data was fit to exponential decays, and time constants of 26.8 (with 95% confidence intervals of 19.3 to 44.0) and of 14.3 (with 95% confidence intervals of 12.3 to 17.2) were calculated for excitations of 641 nm and 785 nm, respectively.

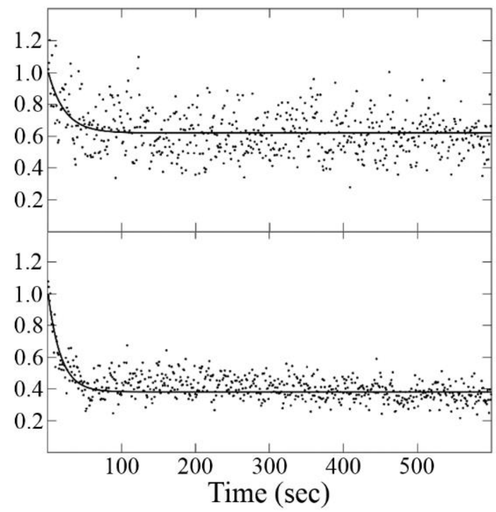

Fig. S3 Rayleigh scattering from 641 nm (top) and 785 nm (bottom) excitation, both for 16 mW, displaying the difference in real-time depletion of nanorods in the focal volume on-resonance and off-resonance.

1. B. Fazio, C. D'Andrea, A. Foti, E. Messina, A. Irrera, M. G. Donato, V. Villari, N. Micali, O. M. Marago and P. G. Gucciardi, *Sci Rep-Uk*, 2016, **6**.
2. H. Liao and J. Hafner, *Chem Mater*, 2005, **17**, 4636-4641.