Supplementary Information

Tuning optical trapping of metal-dielectric hybrid nanoparticles under ultrafast pulsed excitation: A theoretical investigation

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Theoretical Formulation:

The trapping forces and corresponding potentials along the axial directions are calculated using the dipole approximation for Gaussian beam profile. The forces acting on the trapped dielectric particles are the gradient force and the scattering force. While in the case of metallic particles, there is an additional contribution from the absorption forces. Under tight focusing conditions, the axial forces around the geometric focus can be expression as follows [1]:

$$\mathrm{F}_{axial,gradient}(z;r) = -\frac{2\pi n^w a^3}{c}\,\mathrm{Re}[\alpha_0]\,\frac{8Z/(k\omega_0{}^2)}{(1+4Z^2)^2}\,\frac{2P_{peak/avg}}{\pi\omega_0{}^2}\exp[-2R^2] \quad (1)$$

$$\mathrm{F}_{axial,scattering}(z;r) = \frac{8\pi n^w k^4 a^6}{3c}\,|\alpha_0|^2\,\frac{2P_{peak/avg}}{\pi\omega_0{}^2(1+4Z^2)}\exp[-2R^2] \quad (2)$$

$$\mathrm{F}_{axial,absorption}(z;r) = \frac{4k\pi n^w a^3}{c}\,\mathrm{Im}[\alpha_0]\,\frac{2P_{peak/avg}}{\pi\omega_0{}^2(1+4Z^2)}\exp[-2R^2] \quad (3)$$

where $P_{peak/avg}$ is peak/average power under pulsed/continuous-wave (CW) excitations, respectively: $P_{peak} = \frac{P_{avg}}{f\times\tau}$; $R = \frac{r}{\omega_0}$ and $Z = \frac{z}{k\omega_0^2}$ are reduced co-ordinates. Here, ω_0 is spot size at geometric focus which is $\omega_0 = \frac{0.82\lambda}{NA}$; NA is the numerical aperture of the objective, $k = \frac{2\pi n^w}{\lambda}$ is the propagation vector. The polarizability is a significant factor determining the magnitude as well as the direction of force acting on the trapped particle. For the hybrid particles, the effective polarizability per unit volume is expressed in terms of the polarizabilities for core and shell [2]:

$$\alpha_0 = \frac{\alpha'}{V} = \frac{\left(n^{s^2} - n^{w^2}\right)\left(n^{c^2} + 2n^{s^2}\right) + f^3(2n^{s^2} + n^{w^2})(n^{c^2} - n^{s^2})}{\left(n^{s^2} + 2n^{w^2}\right)\left(n^{c^2} + 2n^{s^2}\right) + f^3(2n^{s^2} - 2n^{w^2})(n^{c^2} - n^{s^2})} \quad (4)$$

where, n^c, n^s, and n^w are the refractive index of core, shell, and water (medium), respectively, $f = \frac{a_c}{a_s}$ is the ratio of the radius of core (a_c) to that of shell (a_s). In the limit $n^c = n^s = n^p$ the expression for the polarizability per unit volume will yield that for homogeneous nanoparticle. V in the above formula is $4\pi n^{w^2}\epsilon_0 a_s^3$. In case of silver particles, the refractive index is expressed as $n^p = n_0^p + i\kappa_0^p$; n_0^p is a linear real part of refractive index and κ_0^p is a linear imaginary part which is directly related to the absorptivity [3]. For dielectric particles, the imaginary part does not significantly contribute because absorption is negligible, whereas, in the case of silver particles, absorption is significant, and the overall contribution of the imaginary part is more than the real part of the refractive index. For silver nanoparticles refractive index can be calculated as: $n_0^s = \sqrt{\frac{\epsilon_1}{2} + \frac{\sqrt{\epsilon_1^2 + \epsilon_2^2}}{2}}$, $k_0^s = \frac{\epsilon_2}{2n_0^p}$, and σ_0^s is absorptivity of the particle $\sigma_0^s = \frac{4\pi\kappa_0^p}{\lambda}$; where $\epsilon_1 = Re[\epsilon]$, $\epsilon_2 = Im[\epsilon]$ and $\epsilon = \varepsilon_\infty - \frac{\omega_p{}^2}{\omega^2 + i\gamma_c\omega}$ according to the corrected Drude-Lorentz model for silver nanoparticles [4]. All the relevant parameters used in simulations are listed in table 1 in main text.

Total force acting on the particle in the case of dielectric and metallic particles along axial direction can be written as:

$$F_{total,dielectric,axial}(z; r = 0) = \mathrm{F}_{axial,gradient}(z; r = 0) + \mathrm{F}_{axial,scattering}(z; r = 0) \quad (5)$$

$$F_{total,metallic,axial}(z; r = 0) = \mathrm{F}_{axial,gradient}(z; r = 0) + \mathrm{F}_{axial,scattering}(z; r = 0) + \mathrm{F}_{axial,absorption}(z; r = 0) \quad (6)$$

Numerical integration of the force expressions yields the potential well associated with the optical trap as given below:

$$U_{axial}(r;\, z) = -\int F_{axial}(r;\, z)\, dz \quad (7)$$

In addition to this, the role of nonlinear effects is investigated by incorporating the third, fifth, and seventh order nonlinear refractive indices for silver particles, and second-order nonlinear refractive indices for dielectric particles. The total refractive index, including optical nonlinearity, can be calculated from the given expression for silver particles:

$$n^p = n_0^p + n_2^p \times I_{peak/avg}(r,z) + n_4^p \times I_{peak/avg}^2(r,z) + n_6^p \times I_{peak/avg}^3(r,z) + i\,(k_0^p + k_2^p \times I_{peak/avg}(r,z) + k_4^p \times I_{peak/avg}^2(r,z) + k_6^p \times I_{peak/avg}^3(r,z)) \quad (8)$$

for polystyrene nanoparticles:

$$n^p = n_0^p + n_2^p \times I_{peak/avg}(r,z) \quad (9)$$

Here, $I_{peak/avg}$ is peak and average intensity of the focused Gaussian beam which can be described as [5]:

$$I_{peak/avg}(z;r) = \left(\frac{2P_{peak/avg}}{\pi\omega_0^2}\right)\frac{1}{1+(2Z)^2}\exp[-2R^2] \quad (10)$$

Since the nonlinear refractive index of water is very small, it does not contribute significantly to the linear refractive index for low average power and can be neglected for both CW and pulsed excitations (*i.e.* $n^w \approx n_0^w$) [5].

References:

1. Y. Harada and T. Asakura, "Radiation forces on a dielectric sphere in the Rayleigh scattering regime," Opt. Commun. 124(5-6), 529–541 (1996).
2. B. S. Kim, and T. R. Lee, "The Development of Smart, Multi-Responsive Core@Shell Composite Nanoparticles," In Nanoparticles Technology; InTech, (2015).
3. A. Devi, and A. K. De, "Generalized description of the nonlinear optical force in laser trapping of dielectric nanoparticles," Phys. Rev. Res. 2, 043378 (2020).
4. A. Devi, and A. K. De, "Generalized Lorenz-Mie theory for the reversal of optical force in a nonlinear laser trap," Phys. Rev. A 102, 023509 (2020).
5. A. Devi, and A. K. De, "Theoretical investigation on nonlinear optical effects in laser trapping of dielectric nanoparticles with ultrafast pulsed excitation," Opt. Express 24 (9), 21485-21496 (2016).

Figures and figure captions:

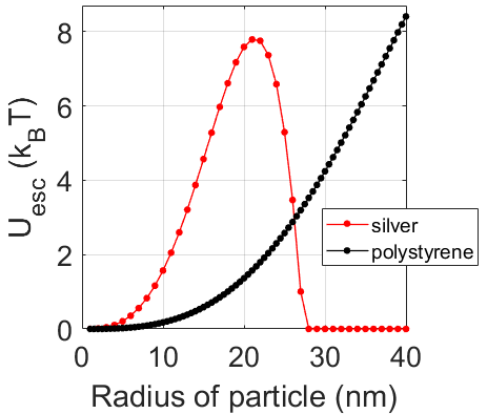

Fig. S1. Plots of escape potential against particle size at 100 mW average power under CW excitation for fixed NA 1.4.

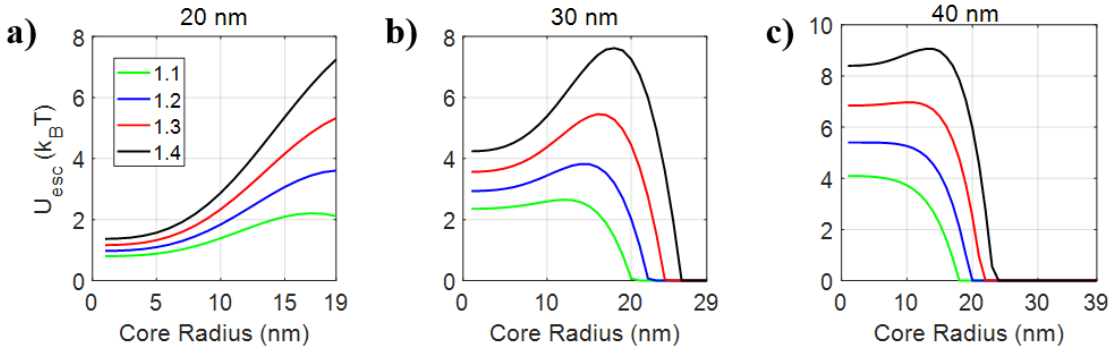

Fig. S2. Plots of escape potential against core radius by fixing shell radius a) 20 nm, b) 30 nm, and c) 40 nm for silver-polystyrene nanoparticles under CW excitation at 100 mW average power.

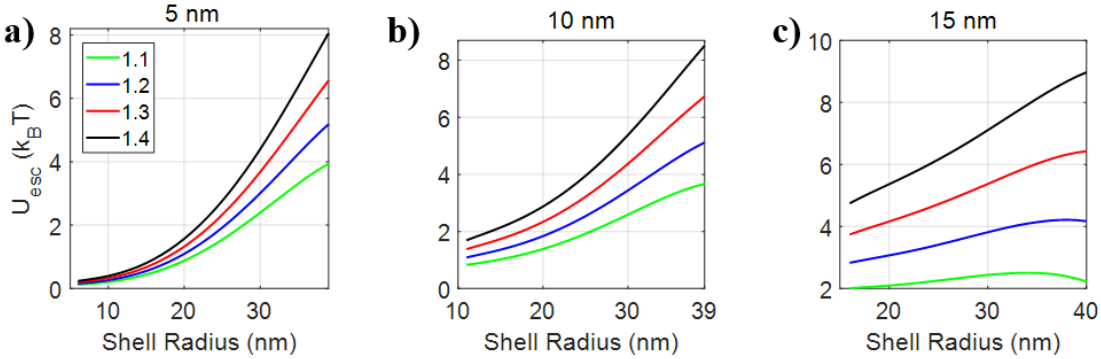

Fig. S3. Plots of escape potential against shell radius by fixing core radius a) 5 nm, b) 10 nm, and c) 15 nm for silver-polystyrene nanoparticles under CW excitation at 100 mW average power.

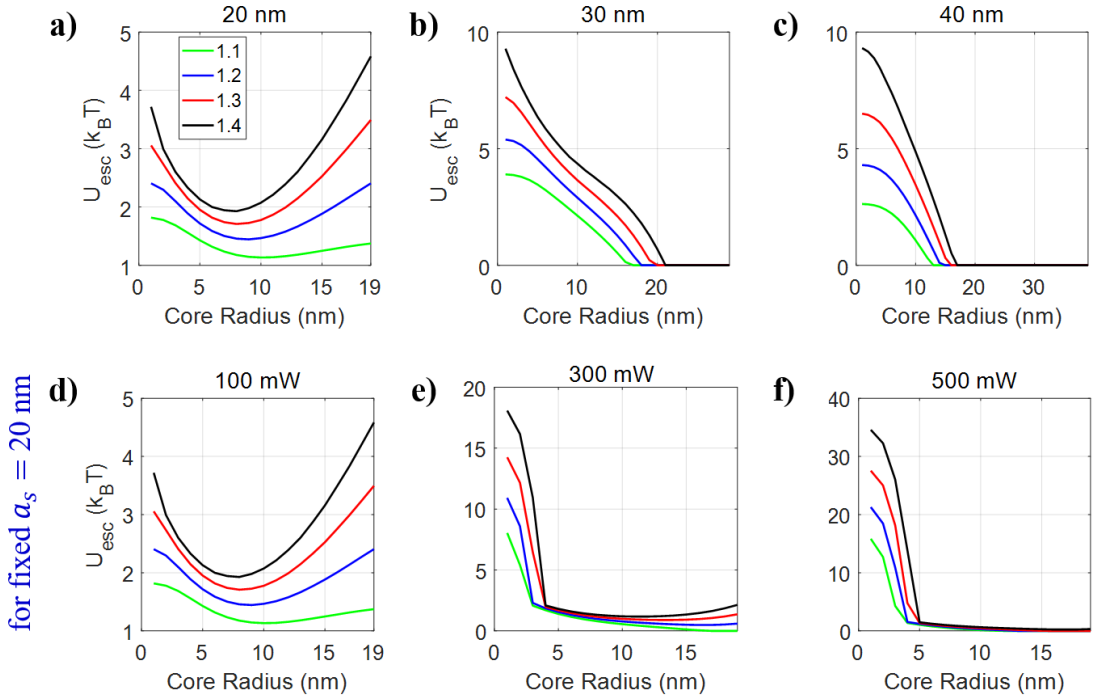

Fig. S4. Plots of escape potential against core radius by fixing shell radius a) 20 nm, b) 30 nm, and c) 40 nm at 100 mW average power, and d) 100 mW, e) 300 mW, and f) 500 mW for fixed shell radius 20 nm for silver-polystyrene nanoparticles under pulsed excitation.

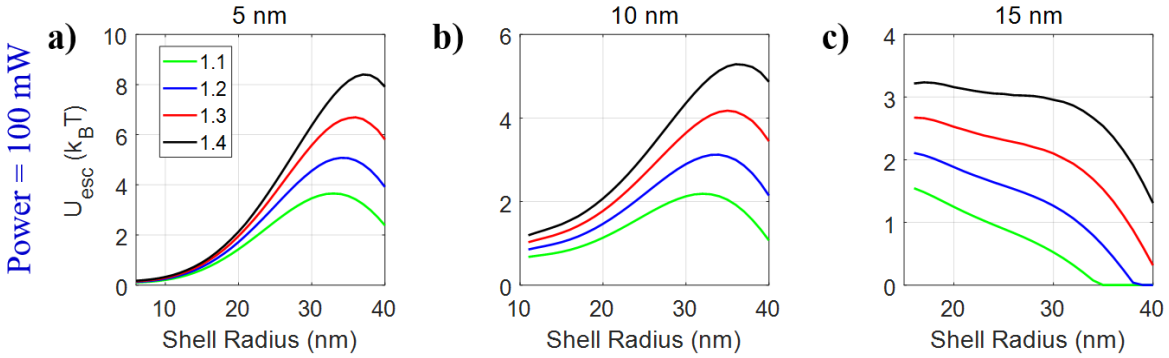

Fig. S5. Plots of escape potential against shell radius by fixing core radius a) 5 nm, b) 10 nm, and c) 15 nm at 100 mW average power for silver-polystyrene nanoparticles under pulsed excitation.