Supplementary Materials for

Long-distance optical pulling of nanoparticle in a low index cavity using a single plane wave

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The PDF file includes:

Supplementary Text
Figs. S1 to S5
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Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/6/21/eaaz3646/DC1)

Movie S1
Supplementary Text

1. Derivation of optical force on a spherical nanoparticle

We consider a spherical nanoparticle (NP) under a linearly polarized plane wave. For simplicity, we set the center of the NP at the origin of the coordinate system; the plane wave has the electric field oscillating in the x-direction and it propagate in the positive z-direction. In this case, due to symmetry, the radiative pressure applies an optical force on the NP in the z-direction. We recall the generalized form of the optical force \( F_z \) in z-direction introduced by Salandrino et al.:\((12)\)

\[
F_z = F_{ss} + F_{ts},
\]

\[
F_{ss} = \frac{\pi \epsilon_0}{k_0^2} \sum_{n=1}^{\infty} \sum_{m=0}^{n} \left\{ C_{nm} \left[ (1 + \delta_{m,0}) \Im \left( a_{enm}^{*} e_{e(n+1)m} + a_{enm}^{*} b_{enm}^{*} e_{e(n+1)m} \right) + (1 - \delta_{m,0}) \Im \left( a_{enm}^{*} e_{o(n+1)m} + a_{enm}^{*} b_{enm}^{*} e_{o(n+1)m} \right) \right] \right\}
\]

\[
F_{ts} = \frac{\pi \epsilon_0}{2k_0^2} \sum_{n=1}^{\infty} \sum_{m=0}^{n} \left\{ C_{nm} \left[ (1 + \delta_{m,0}) \Im \left( a_{enm}^{*} e_{e(n+1)m} + a_{enm}^{*} b_{enm}^{*} e_{e(n+1)m} \right) + (1 - \delta_{m,0}) \Im \left( a_{enm}^{*} e_{o(n+1)m} + a_{enm}^{*} b_{enm}^{*} e_{o(n+1)m} \right) \right] \right\},
\]

where \( C_{nm} = \frac{(n+m)!\sqrt{2(n+2)(n+m+1)}}{(n-m)!\sqrt{2(n+3)(2n+1)}} \) and \( D_{nm} = \frac{(n+m)!2m}{(n-m)!2n+1} \).

Here, \( F_{ss} \) is the optical force induced by the scattered field, \( F_{ts} \) is the optical force induced by the scattered field-incident field interactions. \( a_{o/e nm} \) and \( b_{o/e nm} \) are, respectively, the complex amplitude of the electric and magnetic multipole coefficients of the spherical harmonics with the degree of \( n \) and the order of \( m \) in the incident electromagnetic field. The subscripts ‘o’ and ‘e’ depict the corresponding multipole coefficients of the spherical harmonics with \( \cos(m\phi) \) and \( \sin(m\phi) \), respectively. The superscript ‘s’ depicts the corresponding multipole coefficient in the scattered field. One can find the definition of these variables in Ref. \((12)\). On the other hand, the incident plane wave described above has the following properties:

\[
a_{enm} = 0 \text{ and } b_{omn} = 0 \text{ at } m \neq 1,
\]

\[
a_{onm} = 0 \text{ and } b_{onm} = 0 \text{ at all } m.
\]

Also, the scattered field and the incident field have the relationship as:

\[
a^{s}_{o/e nm} = \alpha_{n} a_{o/e nm},
\]

\[
b^{s}_{o/e nm} = \beta_{n} b_{o/e nm},
\]

where \( \alpha_{l} = \frac{m_{np} \psi_{l}'(\gamma_{np}) \psi_{l}(m_{np} \gamma_{np}) - \psi_{l}(\gamma_{np}) \psi_{l}'(m_{np} \gamma_{np})}{m_{np} \xi_{l}'(\gamma_{np}) \psi_{l}(m_{np} \gamma_{np}) - \xi_{l}(\gamma_{np}) \psi_{l}'(m_{np} \gamma_{np})} \).
\[ \beta_i = -\frac{\psi_i'(ny_p)\psi_i(m_{np}ny_p) - m_{np}\psi_i'(ny_p)\psi_i'(m_{np}ny_p)}{\xi_i'(ny_p)\psi_i(m_{np}ny_p) - m_{np}\xi_i'(ny_p)\psi_i'(m_{np}ny_p)}. \]

Here, \( \alpha_i \) and \( \beta_i \) are the Lorenz-Mie single sphere scattering coefficients, \( \psi_i \) and \( \xi_i \) are Ricatti-Bessel Functions. The prime means differentiation, and \( m_{np} \) and \( y_{np} \) are, respectively, the refractive index ratio and the size parameter of the NP defined in the main text. By substituting equations (A5) and (A6) into equations (A2) and (A3), we can rewrite equation (A1) to obtained the reduced form of the optical force:

\[
F_n = \frac{\pi\varepsilon_0}{2k_0^2} \sum_{n=1}^{\infty} \left\{ C_{n1} \text{Im} \left( \begin{array}{c}
2a_n a_{en1} a_{n+1} a_{e(n+1)}^* + 2\beta_n b_{on1} b_{n+1} b_{o(n+1)}^* \\
+ a_{en1} a_{n+1} a_{e(n+1)}^* + \alpha_n a_{en1} a_{e(n+1)}^* \\
+ b_{on1} \beta_{n+1} b_{o(n+1)}^* + \beta_n b_{on1} b_{o(n+1)}^* \\
+ D_{n1} \text{Im} \left( \begin{array}{c}
2a_n a_{en1} \beta_{n+1} b_{o(n+1)}^* \\
+ a_{en1} \beta_{n+1} b_{o(n+1)}^* \\
+ a_n a_{en1} b_{o(n+1)}^* \\
+ D_{n1} \text{Im} \left( a_{en1} b_{o(n+1)} (2a_n \beta_{n+1} + \beta_n + \alpha_n) \right) \end{array} \right) \right) \right\},
\]

\[ (A7) \]

The multipole term at \( n = 1 \) dominates in equation (A7), and then we can approximate (A7) with more convenient conventions of the spherical harmonics by Mackowski as: (13)

\[
F_n \approx \frac{\pi\varepsilon_0}{k_0^2} \left\{ \begin{array}{c}
\frac{12}{5} \text{Im} \left( p_{11} p_{21}^* 2(2a_k \alpha_z^* + \alpha_z + \alpha_z^*) \right) \\
+ q_{11} q_{21}^* 2(2\beta_1 \beta_2^* + \beta_1 + \beta_2^*) \\
+ \frac{4}{3} \text{Im} \left( p_{11} q_{11}^* (2 \beta_1 \beta_1^* + \alpha_1 + \beta_1^*) \right) \end{array} \right\},
\]

\[ (A8) \]

where \( a_{e11} = 2p_{11}, \ a_{e21} = 2p_{21}, \ b_{o11} = 2i q_{11}, \) and \( b_{o21} = 2i q_{21}. \) Here, \( p_{11}, p_{21}, q_{11}, \) and \( q_{21} \) are, respectively, the electric dipole, the electric quadrupole, the magnetic dipole, and the magnetic quadrupole modal coefficients of spherical harmonics in the incident planewave defined in equation (1) in Ref. (13).

On the other hand, the internal field of a spherical cavity facing a linearly polarized planewave can have the 1st (\( n=1 \) and \( m=1, \) dipole) and 2nd (\( n=2 \) and \( m=1, \) quadrupole) order modal coefficients of spherical harmonics as follows (13):
electric dipole: \( e_d = -\left(\frac{3}{4}\right) \frac{im_c}{m_c \xi_1'(\gamma_c) \psi_1(m_c \gamma_c) - \xi_1(\gamma_c) \psi'_1(m_c \gamma_c)} \),

magnetic dipole: \( m_d = -\left(\frac{3}{4}\right) \frac{im_c}{m_c \xi_1'(\gamma_c) \psi_1(m_c \gamma_c) - m_c \xi_1(\gamma_c) \psi'_1(m_c \gamma_c)} \),

electric quadrupole: \( e_q = \left(\frac{5}{12}\right) \frac{m_c}{m_c \xi_2'(\gamma_c) \psi_2(m_c \gamma_c) - \xi_2(\gamma_c) \psi'_2(m_c \gamma_c)} \),

magnetic quadrupole: \( m_q = \left(\frac{5}{12}\right) \frac{m_c}{m_c \xi_2'(\gamma_c) \psi_2(m_c \gamma_c) - m_c \xi_2(\gamma_c) \psi'_2(m_c \gamma_c)} \).

When we consider the optical force on an NP that is inside a spherical cavity which faces a linearly polarized planewave (see Fig. 1 in the main text), the term \( p_{11}, p_{21}, q_{11}, \) and \( q_{21} \) in the optical force (A8) should be replaced with the modal coefficients of electromagnetic field inside the cavity. With the 1\(^{st}\) order scattering approximation, the replacement can be possible with \( e_d, m_d, e_q, \) and \( m_q \) described in equation (A9), which yields equation (1) in the main text.
2. Scattering coefficient terms

Fig. S1. Scattering coefficient terms as a function of the refractive index ratio ($m_{np}$) and the size factor ($\gamma_{np}$) of NP. The real part and the imaginary part of the scattering coefficients are put inside the log function to effectively show their signs (i.e., positive or negative). (a) The real part of $\delta_{ee}$ is negative; note we put $-\delta_{ee}$ in the log function. (b) The imaginary part of $\delta_{ee}$ is mostly
positive. (c) The real part of $S_{em}$ is mostly positive. (d) The imaginary part of $S_{em}$ is positive. In (b) and (c), the white areas correspond to the regimes with negative signs. In (a), the complex refractive indices of Au, Si, and SiO$_2$ are plotted when $n_c = 1$.

3. Modal coefficient terms of the internal fields inside the cavity

Fig. S2. Modal coefficient terms of the internal fields inside the cavity as a function of the refractive index ratio of the cavity ($m_c$). (a) The real part and (b) the imaginary part of $e_d e_a^*$. (c) The real part and (d) the imaginary part of $e_d m_{d}^*$. In (a) to (d), (red) $\gamma_c = 0.9425$, (magenta) 1.492, (blue) 2.042, (green) 2.591, (cyan) $\pi$. When $n_m =1$ and $r_c = 150$ nm, the trend of wavelength ($\lambda$) is noted in (b) and (c). The insets illustrate the optical configuration of the cavity. In (a) and (d), the solid black lines with arrow ends depict the region where the optical condition in Table 1 in the main text can be satisfied. In (a), the inset illustrates the optical configuration of a spherical cavity to excite internal fields under the incidence of linearly polarized plane wave.
4. Pump-probe optical scattering imaging for the confirmation of nanobubble

We use a pump-and-probe-based optical scattering imaging technique to confirm the formation of nanobubble around SiO₂-Au core-shell (CS) nanoparticles (see Fig. S3).(22,23) The pump laser (800 nm, femtosecond pulsed laser with a repetition rate of 80.7 Mhz and the power of 690 mW – the same as used in the NP movement experiment) is used to thermalize the CS NP in water to create a nanobubble. The probe laser (533 nm, continuous laser) is co-axial with the pump laser, and then a high-sensitivity camera (HX-7, the ISO rating of 8000) is used to capture the scattered probe light at a certain solid angle via a mechanical pinhole.(23) In the experiment, we minimize the size of the mechanical pinhole and reduce the power of the probe beam (green laser, 533 nm) to 5 mW until the camera detects no signal of scattered probe light without the pump beam (see Figs. S3a and c). This offset process can greatly enhance the sensitivity of detecting the increment of scattered light from NPs. Besides, the sensitivity of the camera (ISO 8000) is much higher than normal cameras (ISO 200 to 1600). According to our calculations, at the wavelength of 533 nm, the NP encapsulated by a nanobubble (i.e., supercavitation) will have higher scattering cross-sections than that without the nanobubble (see Fig. S3e). It is noted that the pump beam is filtered out using a short pass filter so it cannot reach the photodetector arrays in the camera. Indeed, we observe that the camera detects the scattered probe light when the pump beam illuminates, and these are shown as glowing dots (see Figs. S3b and d), which is in contrast to the case without the pump laser in Fig. S3c. The change of scattered light at a certain solid angle is only possible when the optical configuration near the CS NP has been changed upon the pump laser. By considering the facts that the pump laser is at the plasmonic resonance peak of the CS NP in water and the power density of laser is above the reported threshold to create a nanobubble (21), we conclude that the change of the intensity of scattered probe light is from the formation of nanobubble around the CS NP. This is also well supported by the calculated result in Fig. S3e.
Fig. S3. Pump-probe optical scattering imaging for the confirmation of nanobubble formed around SiO$_2$-Au core-shell nanoparticles. (a) and (b) Schematics of pump-and-probe optical scattering imaging setup (a) without pump laser and (b) with pump laser. (c) and (d) Optical images from the experiment (c) without pump laser and (d) with pump laser. (e) Calculated scattering cross-section of core-shell nanoparticle with (dot lines) or without (solid) the nanobubble (radius of 150 nm). The inset illustrates the configurations of NP in the nanobubble or the NP in water.
5. Analyzed velocities for representative cases of optical pulling

![Diagram](Light → Focal plane)

**Fig. S4. Tracked positions and the maximum speeds of representative cases of optical pulling motions.** (a) The positions of optically pulled NPs as a function of time for 14 representative cases observed in the experiment. The time interval between two nearest identical symbols is 200 µs. Each color shows the tracking history of an NP which moves from right to left against the light propagation direction. (b) The maximum speed in each case of the pulled NPs. The dotted line indicates the average value.

6. Thermophoretic force of optically pulled CS NP in a nanobubble

When the CS NP excited at the surface plasmon resonance (SPR) gets encapsulated by a nanobubble with the configuration to enable the optical pulling force, there is a temperature difference on the surface of NP along the moving axis (see Fig. 4a in the main text). The NP surface close to the nanobubble interface can have a lower temperature than that of the other side of the NP. This temperature difference ($\Delta T$) across the NP can induce a thermophoretic force ($F_{z}^{th}$) on the NP against the direction of the incident light. Therefore, it is important to check the order-of-magnitude of $F_{z}^{th}$, which can be quantified by (28):

$$F_{z}^{th} = \frac{9\pi \eta^2 k_s \Delta T}{2\rho k_{np} T},$$

(A10)

where $\eta$ is the viscosity of steam, $k_s$ is the thermal conductivity of steam, $\rho$ is the mass density of steam, $k_{np}$ is the thermal conductivity of NP, and $T$ is the temperature of steam. Here, we estimate $F_{z}^{th}$ at two limiting cases of $T$: one is the spatial average temperature of steam in the nanobubble ($T \approx 400$ K, $\eta = 1.3 \times 10^{-5}$ kg·m⁻¹s⁻¹, $\rho = 1.44$ kg·m⁻³, and $k_s = 27 \times 10^{-3}$ W·m⁻¹K⁻¹), and the other is the local temperature of steam around the surface of the NP ($T \approx 700$ K, $\eta = 2.6 \times 10^{-5}$ kg·m⁻¹s⁻¹, $\rho = 7.7$ kg·m⁻³, and $k_s = 60 \times 10^{-3}$ W·m⁻¹K⁻¹). In this way, we can estimate the
order-of-magnitude of $F_{th}^z$ on the NP. We also assume that $k_{np}$ of the CS NP is the volumetric average of the thermal conductivity values of the SiO$_2$ core and Au shell: $k_{np} = 131$ W·m$^{-1}$K$^{-1}$. Actually, the heat conduction efficiency of the CS NP is likely closer to that of the Au as it is a continuous shell that can spread heat fast via its high metallic thermal conductivity. For $\Delta T$, we use a finite element method to calculate the temperature profile on the surface of the NP by solving the momentum, energy, and continuity equations for the NP-in-nanobubble in water system, and then $\Delta T$ is estimated by taking the difference between the averaged surface temperatures of the left and right hemispheres of the NP. The heat generation rate in the Au shell of the CS NP is calculated based on the optical absorption rate of the shell under the light illumination. As a result, we find that $\Delta T$ is 0.5 ~ 1.0 K, which leads to $F_{th}^z$ to have the order-of-magnitude of $10^{-16}$~$10^{-15}$ N as shown in Fig. S5.

**Fig. S5.** Calculated thermophoretic force on a CS NP as a function of nanobubble size. The inset shows the schematic of geometrical configuration of NP in nanobubble where the NP contacts the nanobubble interface at the light-incoming side (i.e., the same configuration in Fig. 4b in the main text).

**Caption for Movie S1**

Long distance optical pulling of Au-silica core-shell NP-in-nanobubble structure. Field of view size: 1004 µm × 110 µm. The Gaussian beam propagates from left to right. Time is in msec.