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Supplementary information

Stand-off trapping and manipulation of sub-10 nm objects and biomolecules using opto-thermo-electrohydrodynamic tweezers

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Stand-off Trapping of sub-10 nm Objects and Biomolecules using Opto-Thermo-Electrohydrodynamic Tweezers

Chuchuan Hong, Sen Yang, Justus C. Ndukaife

The power dissipation density in the gold film is modeled using equation (1) shown below in COMSOL Multiphysics while taking into account the Gaussian distribution of the incident laser beam and exponential decay of the light in the gold film.

$$Q(\mathrm{r}) = P_0 A \frac{\alpha_{Au}}{\pi\sigma^2} \mathrm{e}^{-(\frac{x^2+y^2}{2\sigma^2})} e^{-\alpha_{Au} z} \quad (1)$$

,where P_0 is incident laser power, A is the absorption of the nanohole array. The attenuation coefficient in the nanohole array at the illumination wavelength is given by $\alpha_{Au} = \frac{4\pi\kappa}{\lambda} = 8.91 \times 10^7\ m^{-1}$, where κ is the imaginary part of the refractive index of gold. The lattice constant of the nanohole array is 590 nm, while the thickness of the gold film is 120 nm. This gold nanohole array is numerically simulated in Lumerical FDTD using one unit cell with periodic boundary conditions to find the absorption spectrum.

The temperature distribution around the nanohole array and the surrounding fluid is obtained by solving the heat conduction equation given by:

$$-\mathrm{k}\nabla^2 T + \rho C_p \mathbf{u} \cdot \nabla T = Q \quad (2)$$

In the first term on the left-hand side, k is the thermal conductivity of the materials (glass, gold, or water). T is the temperature in Kelvin, and ∇T is the temperature gradient. The second term is the convection term, which depends on $\mathbf{u}$, the velocity of the fluid. ρ is the density of the fluid and C_p is the specific heat capacity at constant pressure. Q is the heat source density, which is calculated from equation (1). The outer temperature on the outer surface of the boundaries was set to $T = 293.15\ K$.

Inorder to find the electric field distribution in the fluid, we solved the Poisson equation below:

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\varepsilon_0}, \text{ with } \mathbf{E} = -\nabla \mathrm{V} \quad (3)$$

to find the electric field distribution in the vicinity of the nanohole array. The calculated electric field distribution is used in the microfluidic flow simulation.

In the microfluidic flow simulation, we focus on two main flows that enables trapping in OTET: the electrothermoplasmonic (ETP) flow and the a.c. electro-osmosis flow.

Firstly, the ETP flow arise from the localized heating of the fluid to establish a gradient in the permittivity and electrical conductivity in conjunction with an applied a.c. electric

field. The volumetric body force induced in the fluid known as the electrothermal force density, f_{ET} is given by [1]:

$$f_{ET} = \rho_e E - \frac{1}{2}|E|^2 \nabla \varepsilon_m \quad (4).$$

After perturbative expansion in the limit of small temperature gradient, the force density is transformed into [2] :

$$f_{ET} = \frac{1}{2} Re\{\frac{\varepsilon(\alpha-\beta)(\nabla T.E)E^*}{\sigma+i\omega\varepsilon} + \frac{1}{2}\varepsilon\alpha|E|^2\nabla T\} \quad (5)$$

,where $\alpha = {}^{1}/_{\varepsilon}\frac{\mathrm{d}\varepsilon}{\mathrm{d}T} = -0.004\ \mathrm{K}^{-1}$ and $\beta = {}^{1}/_{\sigma}\frac{\mathrm{d}\sigma}{\mathrm{d}T} = 0.02\ \mathrm{K}^{-1}$. ε and σ stands for the permittivity and conductivity of the fluid, respectively. ω is the a.c. field frequency, while ∇T is the temperature gradient calculated from solving the heat equation stated in equation (2).

Subsequently, we solved the Navier-Stokes equation using laminar flow module in COMSOL Multiphysics given by:

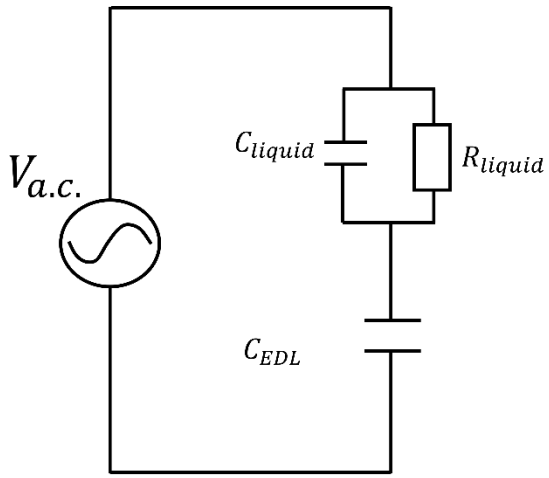
$$\rho_0[\mathrm{u}(\mathbf{r})\cdot\nabla]\mathrm{u}(\mathbf{r}) + \nabla p(\mathbf{r}) - \eta\nabla^2\mathrm{u}(\mathbf{r}) = \mathbf{F} \quad (6),$$

,where body force $\mathbf{F}$ is given by electrothermal force f_{ET}. The contribution from a.c. electro-osmotic flow is modelled by prescribing a slip boundary condition given by the Smoluchowski slip velocity around the nanohole array.

The Smoluchowski slip velocity $\vec{\mathbf{u}}$ is given by: $\vec{\mathbf{u}} = \mu_{eo}\overrightarrow{\mathbf{E}_t}$, where $\mu_{eo} = -\frac{\varepsilon_r\varepsilon_0\zeta}{\mu}$ is the electroosmotic mobility, ε_r is the relative permittivity, ε_0 is the permittivity of free-space, ζ is the zeta potential of the electrical double layer at the nanohole array and fluid interface, and μ is the dynamic viscosity of the liquid. $\overrightarrow{\mathbf{E}_t} = \vec{\mathbf{E}} - (\vec{\mathbf{E}}\cdot\vec{\mathbf{n}})\vec{\mathbf{n}}$ is the tangential component of the electric field, where $\vec{\mathbf{E}}$ is calculated by solving equation 3.

The zeta potential is a.c. frequency-dependent and increases as a.c. frequency reduces [3,4]. The a.c. frequency-dependent zeta potential is determined from the equivalent circuit diagram below.

The equivalent circuit depicting the impedance between the gold film and the fluid (water) is shown in Figure s1. The water medium is modeled as a resistor and capacitor connected in parallel. The electrical double layer (EDL) induced on the surface of the gold nanohole array is modeled as a capacitor with frequency-dependent capacitance.



Supplementary Fig. 1: A schematic of the equivalent circuit across the microfluidic channel. The liquid is composed of a parallel connection of a capacitor and resistor. The electrical double layer (EDL) is considered as a single capacitor connected in series with the liquid's impedance.

The zeta potential is obtained by calculating the voltage drop across C_{EDL}.

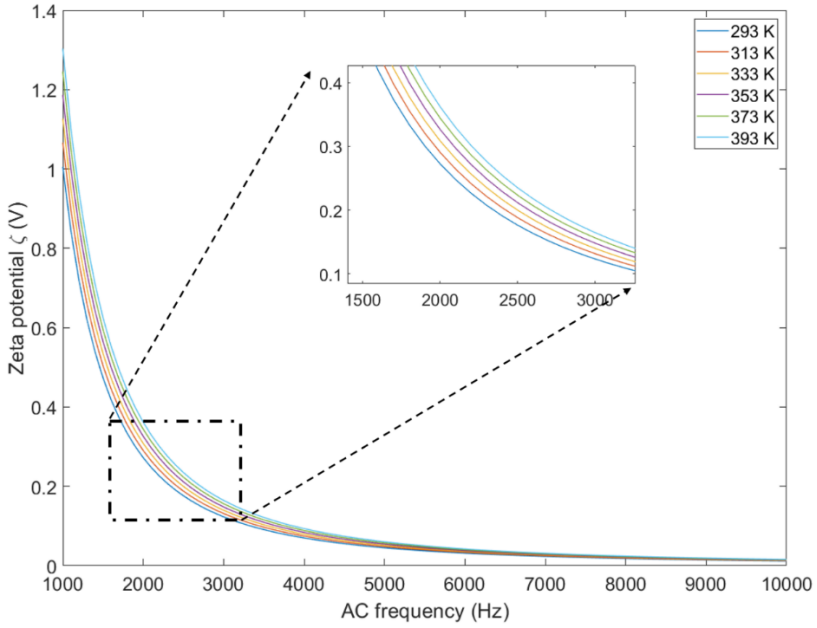
$$\zeta = V_{AC}\frac{Z_{EDL}}{Z_{EDL}+Z_{liquid}} \quad (7)$$

In an RC circuit, we calculate the impedance of the EDL as: $Z_{EDL} = \frac{1}{j\omega C_{EDL}}$ (8), and impedance of liquid (water) $Z_{liquid} = \frac{R_{liquid}}{1+j\omega R_{liquid}C_{liquid}}$ (9). The capacitance of either EDL or water can be obtained by:

$\mathrm{C} = \varepsilon_r\varepsilon_0 \times Area/h$ (10). Here ω is the a.c. frequency, $j = \sqrt{-1}$, ε_r is relative permittivity and ε_0 is the permittivity of free-space. Both the liquid and the EDL have the same surface area, which is the area of the channel. The height of the liquid is the height of channel, which equals 120 μm. The thickness of the EDL is given by the Debye length [4,5]:

$$\lambda_D = \sqrt{\frac{\mu\varepsilon KT}{2\sigma z^2 e}} \quad (11)$$

Here, μ and σ are the mobility and electrical conductivity of the liquid, respectively. ε is permittivity, K is Boltzmann constant, T is temperature, e is the net charge of an electron, and z is the electrolyte charge number. The measured mobility and electrical conductivity of liquid were used in the calculation to find that the Debye length = 10.9 nm. The zeta potential as a function of a.c. field frequency is depicted in Figure s2.



Supplementary Fig. 2: Zeta potential versus AC frequency in the range of 1 kHz to 10 kHz under different temperatures.

As the a.c. frequency drops to below 3 kHz, the zeta potential rises rapidly, which means that the strength of the a.c. electro-osmotic flow (ACEO) increases. This would cause the trapping position to shifted further from the edge of the nanohole array as the a.c. field frequency is reduced, which is in agreement with experimental observations.

Movies:

Supplementary video 1:

Fast transport, trapping, and release of a single BSA protein molecule.

This video shows the rapid transport of the single BSA protein molecule towards the edge of the nanohole array, and the single BSA protein molecule is trapped approximately 8.6 μm away from the edge of the nanohole array pattern within 3 seconds. We see that the protein molecule is stably trapped at the trapping location.

Supplementary video 2:

Dynamic manipulation on a single BSA protein molecule.

In this video, we translate the laser beam relative to the nanohole array, and the BSA protein molecule is moving along the edge that is parallel to the direction that the laser spot is moving relative to the nanohole array.

Supplementary video 3:

Frequency-dependent tuning of trapping position.

By changing the a.c. frequency, the distance between the position of the trapped object and the edge of the nanohole array is tuned.

Supplementary video 4:

Sorting 100 nm Polystyrene particle from 20 nm polystyrene particle by changing the a.c. frequency.

Initially, both 100 nm and 20 nm polystyrene beads were trapped near using a 2.5kHz a.c. frequency. Subsequently, the a.c. frequency was increased to 3.5 kHz, thereby changing the trapping location. We then increased the a.c. frequency to 4 KHz, resulting in the release of the 100 nm polystyrene bead from the trap, while the 20 nm beads stay trapped.

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