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Supporting Information

Heterogeneous Interface Adsorption of Colloidal Particles

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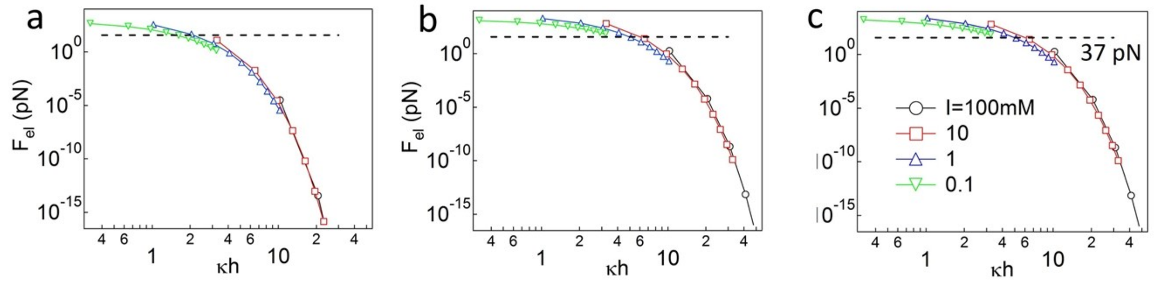

Figure S1. Comparisons of the magnitude of the charge-image charge interaction force (a) with that of the electrostatic interaction between the particle and the oil-water interface (b). (c) Summation of the two interaction forces. The dashed line indicates the optical trapping force that pushes the trapped particle toward the interface.

The surface potential of the image charge is given by $\psi_{image} = \frac{2k_BT}{e}\sinh^{-1}\left[\frac{\varepsilon_W - \varepsilon_{oil}}{\varepsilon_W + \varepsilon_{oil}}\sinh\left(\frac{e\psi_p}{2k_BT}\right)\right]$, where k_B is the Boltzmann constant, T is the temperature, e is the elementary charge, ε_W and ε_{oil} are the dielectric constant of water and oil, respectively, and ψ_p is the surface potential of particle. Due to the relatively small value of ε_{oil} compared to ε_W, the magnitude of the image charge is almost same to the charge on the particle ($\psi = \psi_p \approx \psi_{image} \approx -80mV$). Based on the Derjaguin approximation, the electrostatic interaction force between two spheres is given by $F_{el} = 64 \times 10^3 \pi R N_A I \kappa^{-1} k_B T \Upsilon_0^2 e^{-\kappa h}$, where R is the particle radius, N_A is the Avogadro's number, I is the ionic strength, κ^{-1} is the Debye screening length, h is the surface-to-surface distance, and $\Upsilon_0 = \tanh\left(\frac{e\psi}{4k_BT}\right)$ is the Gouy-Chapman parameter [1]. Because the image charge is formed in the oil phase at the same distance of the charge in the aqueous phase, the charge-image charge interaction force can be calculated by $F_{image} = 64 \times 10^3 \pi R N_A I \kappa^{-1} k_B T \Upsilon_0^2 e^{-2\kappa h}$. As shown in Figure S1, the magnitude of the charge-image charge interaction forces was significantly lower than that of the electrostatic interaction forces between the particle and the interface particularly at salt concentrations of $I \geq 1$ mM, where the large number of particles were adsorbed to the oil-water interface. This result demonstrates that the particle adsorption behaviour for the negatively charged particles to the interface is mainly determined by the electrostatic repulsion between the particle and the interface.

[1] P. C. Hiemenz and R. Rajagopalan, *Principles of colloid and surface chemistry*, CRC press, 1997.