Supplementary Information

Optofluidic transport and manipulation of plasmonic nanoparticles by

thermocapillary convection

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Methods and materials details:

2D Tracking: The 2D tracking method was implemented in Matlab R2012b. The method is based on the algorithm developed by Parthasarathy¹ to find the center of an intensity peak in a pixelated image with sub-pixel precision. In the present case, the center of intensity is the position of the gold nanoparticle. The working principle of the algorithm can be described as follows. First, for every pixel a line is drawn along the local intensity gradient.

The center of the radially symmetric intensity distribution is the point minimizing the distance from all these lines. This point is then calculated using an explicit formula derived from the minimization condition¹. In a first step, we converted all frames of the recorded movie to grayscale images (i.e. intensity images) and selected the particle of interest. Then starting from the first

frame, a region of interest is cropped from the image. On this cropped image, the algorithm of Parthasarathy¹ is applied to find the particle position. After that, the process is repeated for every single frame of the recorded movie. Finally, the laser position is also calculated with the algorithm of Parthasarathy¹.

3D Tracking and Calibration: Like the 2D tracking method, the 3D tracking method was implemented in Matlab R2012b. The lateral position of the particle is determined as described in the previous section. When going through every single frame of the recorded movie, also the information about the third dimension, i.e. the distance from the image plane, is extracted. This is done by applying the method described by Speidel et al.² using the diffraction rings to measure the distance from the focal plane. For all calculations, the outermost diffraction ring has been used. To relate the diffraction ring radius to the distance from the focal plane and thus the liquid surface, a calibration with a single nanoparticle at known distances from the image plane is done beforehand. For that purpose, the stage was moved in discrete 2.52 μ m steps away from the focal plane (Speidel *et al.*²). The slope of the linear dependence of the diffraction ring radius on the particle's distance from the focal plane was determined to be 0.432 ± 0.005. The data points and the corresponding linear fit are shown in Figure SI1.



Figure SI1: Calibration of the 3D Tracking. The fitted line is shown in red and the measured data is plotted in black. The stage position is measured from the first data point. At two positions A and B the corresponding gray scale dark field images are depicted exemplarily.

Particle Motion in a Viscous Fluid : An 80 nm gold nanoparticle in PEG solution can be assumed to move with the same velocity as the fluid. According to Holyst et al. ³, nanoparticles of 80 nm diameter experience a macroscopic value of viscosity in aqueous PEG solution. The Reynolds number Re can be approximated by ⁴

$$Re = \frac{\rho \, v \, l}{\eta} = \frac{1125.3 \, kg \, m^{-3} \times 1 \mu m \, s^{-1} \, \times 80 \, nm}{4 \, Pa \, s} \approx 10^{-11}$$

where ρ is the density of the fluid, v the velocity of the particle relative to the liquid, l the characteristic length scale (i.e. the particle diameter) and η the viscosity of the liquid. Since $Re \ll 1$, the drag force is given by Stokes' law $F_{Drag} = 6 \pi \eta R (u - v)$. Here R is the radius of the nanoparticle, and v and u are the velocity of the nanoparticle and the fluid, respectively. Comparing the particle's inertia to this force gives reaction times of less than 1 ns. Thus it can be assumed that the particle moves with the same velocity as the fluid.

Optical Forces: The simulated optical force corresponding to the particle positions from Figure 2a are shown in Figure SI2. For the simulation, the simulation toolbox developed by Nieminen et al. has been used^{5–7}. Throughout the particle motion, the optical force is orders of magnitude smaller than the drag force $F_{Drag} = 6 \pi \eta R(u) \approx 3 pN$, where η is the viscosity, R the radius of the nanoparticle and u the velocity of the fluid. For distances of a few micrometers, the optical forces are completely negligible.



Figure SI2: Absolute values of the optical force exerted on a nanoparticle traveling along the path shown in figure 2a.

Deviation from a Straight Line: When tracking particles between 10 μ m and 30 μ m distance from the laser spot (see Figure SI4), the deviation from a straight radial line is below \pm 280 nm.



Figure SI3: The motion of three different particles are tracked separately. The particle tracks are shown as a dark blue lines.

Simulation Parameters: To identify and illustrate the dependence of the simulation results on the system parameters, Figure SI4 shows the simulation results for different parameter sweeps. Here, the fluid velocity at the surface with respect to the distance from the laser focus (cf. Figure 3 in the main text) is taken as a reference. Variations of the friction parameter *k* and the surface tension gradient γ (defined here as the first partial derivative of the surface tension with respect to temperature) have different effects on the shape of the curve. Thus, they are decoupled and can be determined in a fit uniquely. For comparison, the case of $k = 0 N s^{-1} m^{-3}$, i.e. no friction, is also shown. Variations of both the heat conductivity κ and the viscosity η of the PEG solution have similar effects compared to a variation of γ . Therefore, differences from the fitted and the expected value of γ can also be attributed to uncertainties in the estimation of κ and η .



Figure SI4: The figure shows different parameter sweeps for (a) the slip parameter k, (b) the surface tension gradient γ , (c) the heat conductivity κ and the viscosity η . Note that only the parameter specified in the plot is varied while all other parameters stay constant.

Flow Field Tracking: The velocity field of the nanoparticles have been determined using the following data processing routine: First, the traces of all nanoparticles are calculated using the two dimensional tracking technique. Then the region of interest, i.e. the vicinity of the laser beam configuration, is divided into small rectangular bins. For each rectangle all particles passing through it are determined. Once these particle traces have been found, the mean velocity of these particles passing through the rectangle is calculated. If no particle has passed through a specific rectangle, this rectangle is omitted. The data from all these rectangular bins then constitutes the flow field.

Image Overlay: An alternative representation of the particle movement shown in Fig 4. Additionally, the movies corresponding to Fig. 4 (a)-(c) (Fig. SI5 (a)-(c)) can be found online.



Figure SI5: An alternative representation of the particle movement from Fig 4. All images (a)-(c) have been created by overlaying the five images shown in Fig. 4 (a)-(c) respectively. In (c), the particle positions has been numbered chronologically.

A Game of Pong: A movie of a game of pong with gold nanoparticles can be found online. The

different laser spots have been created with the Spatial Light Modulator. The position switching

was done manually according to the movement of the particle, like a player would do in a "real"

game of pong. For better illustration, the movie has been speeded up by a factor of 4.

References:

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