Characterization of the near-field and convective transport behavior of micro and nanoparticles in nanoscale plasmonic optical lattices

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Here, we report the characterization of the transport of micro- and nanospheres in a simple two-dimensional square nanoscale plasmonic optical lattice. The optical potential was created by exciting plasmon resonance by way of illuminating an array of gold nanodiscs with a loosely focused Gaussian beam. This optical potential produced both in-lattice particle transport behavior, which was due to near-field optical gradient forces, and high-velocity ($\sim \mu\text{m/s}$) out-of-lattice particle transport. As a comparison, the natural convection velocity field from a delocalized temperature profile produced by the photothermal heating of the nanoplasmonic array was computed in numerical simulations. This work elucidates the role of photothermal effects on micro- and nanoparticle transport in plasmonic optical lattices. Published by AIP Publishing. [<http://dx.doi.org/10.1063/1.4948775>]

I. INTRODUCTION

The optical trapping of microscale and nanoscale particles was pioneered by Ashkin *et al.* in the early 1970s, and has become a standard tool for micro- and nanoscale optical manipulation.^{1,2} Plasmonic optical tweezers have drawn much research attention in recent years.^{3–5} For conventional optical tweezers based on the far-field focusing technique, the spatial confinement of the light field is inherently limited by diffraction, and the magnitude of the trapping force drops dramatically (following an $\sim a^3$ law for nanoparticle of radius a) when the particle size is much smaller than the wavelength of the light.⁶ Plasmonic-based, near-field optical manipulation has been developed to overcome these diffraction-imposed limitations, and has been successfully applied to trap dielectric and metallic nanoparticles, and bacteria.^{7–10} In liquid environments, these efforts have culminated in the recent demonstration of the tweezing of a single protein molecule and measurement of mechanical vibration to reveal its identity with the extraordinary acoustic Raman technique.^{11–15}

Technical advances in conventional optical trapping techniques have enabled the creation of microscale and nanoscale one-, two-, and three-dimensional periodic potentials (or optical lattices).^{16–18} Using such techniques and by leveraging the principle of optical fractionation in microfluidic environments, optical sorting by particle size or index of refraction has been demonstrated.^{17,18} A scaled-down version of such periodic potentials can be produced using periodic plasmonic nanostructures.^{19,20} Cuche *et al.* recently demonstrated that near-field optical forces can produce negative refraction effects on the trajectory of nanoparticles in a plasmonic crystal created in a patterned metallic film.¹⁹ The same group of authors also reported on trapping and transport over a periodic array of gold nanodiscs, under illumination with a loosely

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focused Gaussian beam.²⁰ Single-particle transport and multiple-particle trapping have been demonstrated for dielectric nanospheres with a diameter of 100 nm.

Study of the transport of nanoparticles with plasmonic optical tweezers is inevitably complicated by the metallic plasmonic nanostructures used in plasmonic optical tweezers, which can act as a heat source upon illumination, leading to a photothermal effect. Quidant's group used a Green's function approach to calculate both the temperatures of plasmonic nanostructures of various shapes, including nanospheres and nanodiscs,^{21,22} and fluidic convection velocities, on the order of $\sim$ nm/s, near single isolated plasmonic nanostructures.²³ Using the same approach, Baffou *et al.* showed that the temperature profile of a plasmonic nanodisc array could be calculated by summing over the contributions of each individual nanodisc; these calculations were validated experimentally.²⁴ Toussaint's group calculated the convection flow induced by an array of nanoantennas, and concluded that the optically absorptive, thermally conductive indium tin oxide (ITO) could efficiently distribute the thermal energy to generate an increase in convection velocity.²⁵ As a result, high fluidic velocities of the order of $\sim$ μ m/s could be achieved. They also demonstrated that the optically and thermally induced forces could be tuned to generate various distinct states of trapping, including lateral delocalization, and hexagonally closed-packed clusters.²⁶ The photothermal effects could also be combined with an AC electrical field to enable electrokinetic manipulation, allowing for microparticle sorting; this sorting was achieved by tuning the frequency of the AC field to create a strong electrothermal fluid flow, with a velocity as high as 50 μ m/s.²⁷ Here, we demonstrate the interplay between the in-lattice micro and nanoparticle transport, driven by the near-field plasmonic forces, and the natural convection field. A simple square plasmonic optical lattice was used to confer in-lattice transport, similar to our previous work.²⁰ The optical lattice used here also produced a delocalized temperature profile. These two features distinctly differed from Toussaint's work on the arrays of bowtie nanoantennas.²⁵

II. EXPERIMENTAL SETUP

A. Optical setup

The experimental setup was modified from a commercial optical trapping kit (OTKB1, Thorlabs).²⁰ A fiber-coupled diode laser 980 nm beam (PL980P330J) was expanded via beam expansion, and loosely focused using an oil immersion microscope objective (NA = 1.25, 100 $\times$, Nikon) to excite the plasmonic sample. The loose focusing was achieved by partially overfilling the back aperture of the microscope objective. The spot size σ of the Gaussian beam was obtained by fitting the transverse intensity distribution, $I = (P/2\pi\sigma^2) \exp(-r^2/2\sigma^2)$, of the CCD camera (DCU224, Thorlabs) image, where r is the azimuthal radius and P is the total optical power. The spot size σ was 4.0 μ m, and corresponded to the full width half maximum, $H = 2(2\ln 2)^{1/2}\sigma = 9.4 \mu$ m (see Figure 1). Fluorescent excitation was achieved at 470 nm, using a light emitting diode (LED) light source (Touchbright, Korea), and the fluorescent image was captured using the same objective, in conjunction with a dichroic mirror (DM) and an emission filter (41001, Chroma Technology). Fluorescently labeled nanospheres, with a diameter of 500 nm, were dispensed on the sample, and the motion of the fluorescent particles was recorded using a CCD camera. A custom-designed Matlab program was then used to adjust the threshold of the image, and to extract the particle trajectory data. The motion was recorded using the CCD camera, with a frame rate of 10 frames per second.

B. Fabrication of the plasmonic nanostructure

To create the two-dimensional plasmonic array, metallic nanostructures were patterned on an indium tin oxide (ITO)-coated cover glass, using electron beam lithography, thermal evaporation of a 40-nm gold layer, and the lift-off technique. No adhesion layer was used to minimize the damping of localized surface plasmon resonance. In these experiments, a 22×22 array of nanodiscs was used, and each unit cell contained a nanodisc with a diameter $d = 550$ nm; the closest distance between the centers of pairs of nanodiscs was $p = 750$ nm. In order to confer

FIG. 1. Experimental setup. (a) Schematic view of the setup. A Gaussian beam with a wavelength of 980 nm was used to excite the surface plasmon resonance of the nanostructures, through a microscope objective ($100\times$ NA = 1.25, Nikon). The inset shows an SEM micrograph of the nanostructures. The simple square array consisted of a 22×22 square lattice of gold nanostructures. Each primitive cell consisted of one gold nanodisc with a thickness of 40 nm and a diameter of ~ 550 nm. The period of the square lattice was 750 nm. Two polarization directions of the incident light were used; $\mathbf{E}_1$ and $\mathbf{E}_2$ denote the polarization directions of the excitation 45° and 0° to the x axis, respectively. (b) Illustration of nanoparticle transport over the nanoplasmonic array illuminated with a Gaussian beam.

delocalized transport within the lattice, the distance between the nanodiscs and the diameter of a nanodisc should be roughly comparable to the diameter of the nanoparticles being trapped.²⁸

C. Simulation of the electromagnetic field intensity distribution

The time-averaged electric field intensity enhancement $|E|^2/|E_{inc}|^2$ distribution was simulated using a commercial software package (Lumerical Solutions Inc., Canada), where E_{inc} is the electrical field of the incident electromagnetic field in the simulation. Surface plasmon resonance on a primitive cell was excited by either 0° or 45° linearly polarized light. Each primitive cell consisted of one gold nanodisc with a thickness of 40 nm and a diameter of 550 nm, as indicated by the black dashed circles in Figure 2. Periodic boundary conditions were imposed to extract the results. The adsorption cross-section, σ_{abs} , was also calculated.²⁹ The adsorption cross section of the gold nanostructure was obtained by a custom-written script designed to illuminate one nanodisc with a plane wave with uniform intensity I_{inc} , with periodic boundary conditions imposed on the finite domain time difference (FDTD) simulations, and to calculate the power P_{abs} being adsorbed, such that, $\sigma_{abs} = P_{abs}/I_{inc}$.

D. Temperature profile measurement and analytical calculation

We have previously measured the temperature profile near the optical lattice using the temperature-sensitive fluorescent dye, Rhodamine B (Sigma Aldrich).³⁰ Briefly, the fluorescent intensity of the dye versus temperature was measured to obtain a calibration curve in a microfluidic channel of $\sim 8\ \mu\text{m}$ height, positioned in a uniformly heated microscope incubator (Living Cell Instrument, Korea) mounted on an inverted microscope (Leica Microsystems). The microfluidic chip used for calibration was prepared from polydimethylsiloxane, by soft lithography. Due to photothermal heating, the temperature profile was obtained by placing deionized water with the dye on the illuminated nanoplasmonic array, recording the CCD image for fluorescent intensity change, and calculating the temperature from the calibration curve. The temperature profile measured at optical power ~ 10 mW at different heights from the substrate surface is displayed in Figures 3(a) and 3(b). Note that the nanodisc array severely interfered with the fluorescent intensity, so we were unable to measure the temperature within the lattice.

FIG. 2. Simulated, time-averaged electric field intensity enhancement $|E|^2/|E_{inc}|^2$ distribution. (a) The intensity distribution shows an enveloped expanded Gaussian. (Inset) Surface plasmon resonance on a primitive cell was excited by 0° polarized light (relative to the x axis). The intensity profile was recorded at a plane 290 nm above the substrate. (b) Same as (a), with the exception that the array was excited using 45° polarization light, and the excitation formed fringed patterns. The black dashed circles indicate the rims of the nanodiscs.

We also calculated the temperature profile using an analytic formalism. Baffou *et al.* showed that the temperature profile of a plasmonic nanodisk array can be calculated by summation over the contribution of each individual nanodisk.²⁴ The uniform temperature approximation (UTA) was employed for the temperature profile; this was justified because the thermal conductivity of the gold was much greater than the thermal conductivities of the surrounding water medium and the glass layer, i.e., $\kappa_{\text{gold}} \gg \kappa_{\text{water}}$, $\kappa_{\text{gold}} \gg \kappa_{\text{glass}}$. We also neglected the contribution of the ITO layer, since it is much thinner than the underlying glass substrate. The temperature on the surface of the j th nanodisk was obtained using the formula,

$$\Delta T_j(\vec{r}) = \sum_{\substack{k=1 \\ k \neq j}}^{N_{\text{total}}} \frac{q_k}{4\pi\bar{\kappa}|\vec{r}_j - \vec{r}_k|}, \quad (1)$$

where q_j is the total heat of the j th nanodisk resulting from the photothermal heating, given by $q_j = \sigma_{\text{abs}} I$, and $\bar{\kappa}$ is the averaged thermal conductivity of the medium and the substrate, i.e., $\bar{\kappa} = (\kappa_{\text{glass}} + \kappa_{\text{water}})/2$. Here, we used $\kappa_{\text{glass}} = 1.38 \text{ W/mK}$ and $\kappa_{\text{water}} = 0.6 \text{ W/mK}$.²⁴ A similar formula was used to calculate the rise in temperature on the surface of the nanodiscs. Equation (1) can be written in the following more explicit form

$$\Delta T^{\text{ext}}(x, y, z) = \frac{1}{4\pi\bar{\kappa}} \frac{\sigma_{\text{abs}} P}{2\pi\sigma^2} \sum_{l,m} \frac{\exp\left[-(x_{l,m}^2 + y_{l,m}^2)/2\sigma^2\right]}{\left[(x - x_{l,m})^2 + (y - y_{l,m})^2 + z^2\right]^{1/2}}. \quad (2)$$

Here, the intensity I and the heat power q_j delivered by the j th nanodisk were used

$$q_j = \sigma_{\text{abs}} I(x_{l,m}, y_{l,m}) = \frac{P\sigma_{\text{abs}}}{2\pi\sigma^2} \exp\left[-(x_{l,m}^2 + y_{l,m}^2)/2\sigma^2\right]. \quad (3)$$

For a two-dimensional $N \times N$ square lattice, two indices, l and m , are needed to specify the location of the nanodisk for the summation (N is an even integer). The position of the nanodisk with indices (l, m) is denoted as

FIG. 3. Measured and calculated temperature profiles. (a) Measured temperature profiles at heights 0 μm , 5 μm , and 10 μm from the substrate surface, under 0° polarization. The dashed line marks the edge of the optical lattice. (b) Same as (a) but for 45° polarization. (c) Calculated temperature profile at heights 0 μm , 5 μm , and 10 μm from the substrate surface, under 0° polarization. Both calculations from the analytic theory and the COMSOL simulation are displayed. (d) Same as (c) but for 45° polarization. (e) Calculated temperature distribution in the z -direction from the lattice center and edge. Both calculations from the theory and the COMSOL simulation are displayed. (f) Same as (e) but for 45° polarization.

$$(x_{l,m}, y_{l,m}) = (lp, mp) - \left(\frac{N-1}{2}p, \frac{N-1}{2}p \right) \quad l, m = 0, 1, 2, \dots, N-1, \quad (4)$$

with the convention that the origin of the coordinate system is at the center of the lattice. Figures 3(c)–3(f) show the calculated temperature profile from a simple square array excited by illumination with 0° and 45° polarization for a total incident power $P = 10 \text{ mW}$, at different heights from the substrate.

E. Out-of-lattice particle velocity measurements

Microspheres with diameters of 2 μm and 4 μm were used for the particle velocity measurements outside the nanoplasmonic array, similar to the method used by Toussaint's group.²⁵ These microspheres, rather than nanospheres with a diameter of 500 nm, were used because they yielded consistent results in the power range of interest, especially at low optical powers. Measurements performed using nanospheres with a diameter of 500 nm suffered from the

excessive Brownian motion of the nanospheres. Time-lapse movies were taken, and data were post-processed using a centroid algorithm.²⁹ The length for the transport was typically $\sim 8 \mu\text{m}$ outside the nanoplasmonic arrays, and data were averaged over eight measurements from various incident directions.²⁹ In general, the microspheres entering the optical lattice were repelled by the axial convective flow, and eventually exited the optical lattice.²⁶ The particle velocity data are displayed in Figure 4. As a comparison, the particle velocity for a substrate with only an ITO layer was also measured and was significantly lower than the velocity of particles with an optical lattice.

F. Calculated natural convection velocity field

The natural convection velocity can be calculated using the Boussinesq approximation

$$\nabla \cdot [-\kappa_{\text{water}} \nabla T(\vec{r}) + \rho_{\text{water}} c_p T(\vec{r}) \vec{u}(\vec{r})] = 0, \quad (5a)$$

$$-\nu \nabla^2 \vec{u}(\vec{r}) = g \rho_{\text{water}} \beta(T) [T(\vec{r}) - T_0] \hat{z}, \quad (5b)$$

$$\nabla \cdot \vec{u}(\vec{r}) = 0, \quad (5c)$$

where $T(\vec{r})$ and $\vec{u}(\vec{r})$ are the spatial temperature and velocity fields, respectively, and g is the gravitational acceleration. Here, we also assumed that the medium is incompressible and in a

FIG. 4. In-lattice and out-of-lattice particle transport. The out-of-lattice particle velocity was measured using 2- μm and 4- μm microspheres placed outside the optical lattice produced using: (a) 0° and (b) 45° polarization, respectively. For comparison, the averaged in-lattice drift (along the trajectories shown in Figs. 6 and 7) velocity, particle velocity for ITO alone, and numerically calculated natural convection fluidic velocity at height 2- μm are also displayed.

steady-state condition. The values used for the water density and ambient temperature were $\rho_{\text{water}} = 998 \text{ kg m}^{-3}$ and $T_0 = 20^\circ\text{C}$, respectively. Both the thermal expansion coefficient, $\beta(T)$, and the viscosity, ν , of the water were considered temperature-dependent.³¹ The ratio of heat transfer by convection versus advection, i.e., $|\rho c_p \bar{u} \cdot \nabla T|/|\kappa_{\text{water}} \nabla^2 T|$ in the energy, Eq. (5a), yields the thermal Peclet number, $Pe_T = \rho c_p u H^2 / \kappa_{\text{water}}$, which for our system is always much smaller than one. Accordingly, the effect of fluid flow on the temperature field can be neglected and Eq. (5a) can be simplified to $\nabla^2 T(\bar{r}) = 0$. An order of magnitude estimate for the natural convection velocity from Eq. (5b) is $u \sim H^2 g \beta / \nu$, which is in the order of $\mu\text{m/s}$, in agreement with the maximal velocities in the simulation in Fig. 5.²⁹ Herein, a thermal expansion coefficient $\beta = 10^{-4} \text{ K}^{-1}$, kinematic viscosity $\nu = 10^{-6} \text{ Pa s}$, and $\Delta T \sim 20\text{K}$ were taken.

The velocity field was numerically solved using COMSOL, with the following simplifications: (1) an axisymmetric geometry was used so as to solve a two-dimensional instead of a three-dimensional problem; (2) the discrete heat power generation (W) within each of the nanodiscs was replaced with a continuous heat flux (W/m^2) by dividing Eq. (3) by the unit cell $A_{\text{cell}} = p^2$; (3) the square nanodisc array was replaced with a circular continuous heat-generation region bearing the same active area, i.e., $R_{\text{eff}} \approx 9.2\mu\text{m} = Np/\sqrt{\pi} \sim 9.2\mu\text{m}$; (4) the thermal problem, which is decoupled from the hydrodynamic one, was solved within a small fluidic domain only, instead of also resolving the thermal problem within the substrate, by modifying the heat flux term to account for the parallel thermal resistance circuit, wherein the generated heat can dissipate into both the fluid and wall domains

$$q''(r) = \left[\frac{P\sigma_{\text{abs}}}{2\pi\sigma^2} e^{-r^2/2\sigma^2} \right] \left(\frac{\kappa_{\text{water}}}{\kappa_{\text{water}} + \kappa_{\text{glass}}} \right) \frac{1}{A_{\text{cell}}}. \quad (6)$$

This approach is identical in using $q''(r) = \frac{1}{2} \left[\frac{P\sigma_{\text{abs}}}{2\pi\sigma^2} e^{-r^2/2\sigma^2} \right] \frac{1}{A_{\text{cell}}}$ with an effective thermal conductivity, which is an average between that of the fluid and substrate, i.e., $\bar{\kappa} = (\kappa_{\text{glass}} + \kappa_{\text{water}})/2$, as used in the analytical approach.²⁴ The agreement between the analytical and numerical solutions was confirmed via the temperature profiles in Figure 3. The hydrodynamic problem was then solved using Eqs. (5b) and (5c) to yield the velocity field shown in Figure 5.

FIG. 5. Numerical simulation of the temperature (color) and velocity (vector) fields. The thermal and hydrodynamic boundary conditions are indicated. σ is the heat flux density. Axisymmetric geometry was used.

FIG. 6. Representative trajectory of (a) single 500-nm-diameter nanospheres entering the edge of the array were attracted to the central region of the simple square lattice generated by 0° polarization light. Each trajectory was collected separately and compiled into this figure. The frame-by-frame points here were obtained via image processing of a movie recorded at 10 frames per second. The green and blue trajectories are representative of non-channeling states (shown for comparison). (b) Same as (a), but with 45° polarization light excitation. For all data presented here, the total incident laser power was ~ 1.5 mW.

III. RESULTS AND DISCUSSION

In this work, we characterized both in-lattice transport of nanoparticles and out-of-lattice microparticle transport. We performed experiments on a simple square lattice, using linearly polarized light. Two different directions of polarization were used: one along the diagonal of the square lattice and the other aligned along the edge of the square lattice. We first characterized the transport behavior of single nanospheres with a diameter of 500 nm. As in our previous work, the Gaussian beam was used to conveniently create a field gradient, such that a particle moving close to the edge of the optical lattice will subsequently move toward the center of the optical lattice. Figure 6 shows a representative set of particle trajectories for two different polarization directions (0° and 45° directions). In both cases, the nanoparticles roaming around the edge of the optical potential were typically attracted toward the center of the lattice. To provide a quantitative comparison of the observed transport process with that of free diffusion, we defined the square of the (two-dimensional) distance R^2 as $R^2 = (x - x_0)^2 + (y - y_0)^2$, where x and y are the frame-by-frame coordinate points of each trajectory, and x_0 and y_0 are the coordinates of the initial (edge) point of the trajectory. Figs. 7(a) and 7(b) show plots of the square of the distance as a function of time for the trajectories shown in Figs. 6(a) and 6(b). As a comparison, the expectation value of the square of the free diffusion distance versus time, which is given by $\langle R^2(t) \rangle = 2Dt$,³² is also plotted with the corrected diffusion coefficient $D = \gamma k_B T / 3\pi\eta d$, where k_B , T , η , d , and γ are the Boltzmann constant, the temperature, the viscosity of the medium, the diameter of the spheres, and Faxén's correction coefficient due to wall effect, respectively. The correction factor due to wall effect to the fifth order of (d/z) is given by^{33,34}

FIG. 7. The square of distance versus time for 500-nm-diameter nanospheres in a simple square lattice generated using: (a) 0° and (b) 45° polarized light. The analysis is based on the particle trajectories displayed in Figure 6, and presented using the same color code. The black dashed line shows the expected values for the square of the corrected diffusion distance for a sphere of the same size. For all data presented here, the total incident laser power was ~ 1.5 mW.

$$\gamma = 1 - \frac{9}{16} \left(\frac{d}{2z} \right) + \frac{1}{8} \left(\frac{d}{2z} \right)^3 - \frac{45}{256} \left(\frac{d}{2z} \right)^4 - \frac{1}{16} \left(\frac{d}{2z} \right)^5, \quad (7)$$

where z is the distance between the particle's center and the substrate. At $T = 298$ K, we used $\eta = 8.9 \times 10^{-4}$ Pa s for the viscosity of the water,³⁵ $z = 290$ nm, and $d = 500$ nm, yielding the diffusion coefficient $D = 0.98 \mu\text{m}^2/\text{s}$. Note the height, z , is the summation of the radius of the nanoparticle and the thickness of the nanodisc.

We also computed the effective in-lattice velocity and passage time for the single trajectory data displayed in Figs. 6(a) and 6(b). The effective in-lattice velocity is defined as $v_{\text{eff}} = R/T_p$, where R is the distance from the edge to the center and the passage time, T_p , is defined as the time from particle entry at the edge of the array until it reaches the center of the array. The calculated in-lattice velocity ranges from $1.8 \mu\text{m/s}$ to $3.7 \mu\text{m/s}$ for 0° polarization and from $2.0 \mu\text{m/s}$ to $2.9 \mu\text{m/s}$ for 45° polarization.²⁹ These values are significantly higher than the values of the computed natural convection velocity at comparable optical power, as displayed in Figure 4. Note that the velocity measurements for in-lattice and out-of-lattice particles are obtained at different locations. As shown in Figure 5, the simulation showed that the convective velocity decreases as the particles approach the center of the optical lattice, so the convective velocity at the center is expected to be even smaller than the values measured out of lattice. This indicates that the in-lattice transport indeed predominantly arises from the near-field optical force rather than from the convective fluidic drag.

A striking feature in Figure 4 is that out-of-lattice particle transport velocities were in the order of $\sim \mu\text{m/s}$, which is roughly an order of magnitude higher than the numerically calculated natural convective fluidic velocity. The latter were calculated $2 \mu\text{m}$ above the substrate and $\sim 15.2 \mu\text{m}$ radially away from the array center, which corresponds to the same region as the $2 \mu\text{m}$ and $4 \mu\text{m}$ microparticle velocity measurements.²⁹ We therefore rule out photothermal convection as the dominating cause for this transport. Alternatively, we propose that it adds up to another transport mechanism that is related to the optical evanescent field produced by the ITO layer. The movement of microparticles due to the evanescent field of a glass-water interface was reported by Kawata's work.³⁶ In our setup, the thin 40 nm-thick ITO layer had index of refraction ~ 1.7 at 980 nm,³⁷ which is higher than the water (with index refraction ~ 1.3) and glass (with index refraction ~ 1.5), and therefore can serve as a dielectric planar waveguide.³⁸ This mechanism may also explain why the velocities of $2 \mu\text{m}$ microspheres (with and without a plasmonic nanoarray) are consistently higher than those of $4 \mu\text{m}$ microspheres (Fig. 4), since they are subjected to a higher force from the evanescent field, whose intensity exponentially decays along the z direction. The transport velocity of particles with plasmonic nanoarrays is higher than those with ITO only, which means that the plasmonic nanoarray further enhances transport by coupling the near-field optical gradient forces to the optical evanescent field of the planar waveguide.³⁹

Several alternatives were checked to reconcile between the numerically computed natural convection velocity and the experimentally measured particle velocity, but the efforts were futile. First, the temperature rise cannot be simply increased by an order of magnitude, which in turn will accordingly increase the convection velocity, since this would be inconsistent with the measured temperature profile depicted in Figures 3(a) and 3(b). Second, we tried to account for the effect of the ITO layer, both in terms of its relatively high thermal conductivity and radiative absorption. The numerical simulations indicated a small temperature increase (a maximum of ~ 3 K) when using an experimentally determined adsorption coefficient,³⁷ which cannot explain neither this difference nor the velocity difference between the nanoplasmonic array with ITO and the ITO layer only, as seen in Figure 4.²⁹

Differences between the current work and a previous work by Toussaint's group²³ include identification of a different mechanism for the high fluidic velocity of out-of-lattice transport, which was determined here, by carefully comparing photothermal convection from simulation. Essentially, we obtained a maximal velocity $\sim 0.8 \mu\text{m/s}$ at the location $z = 80 \mu\text{m}$ above the center of the lattice,²⁹ which is of the same order of magnitude as Toussaint's results. They display

maximal velocity at a height of $>50\ \mu\text{m}$ above the array and compare these values to the measured particle velocity on the substrate surface. We used the same region on the substrate surface to compare the measured results with simulation. Moreover, Toussaint's group used a wide range of tracer microparticles size ($1\text{--}20\ \mu\text{m}$), while we refined the measurement by using two definite sizes ($2\ \mu\text{m}$ and $4\ \mu\text{m}$) of microspheres. Our convectional velocity calculation used a temperature profile consistent with the measurement results in Figures 3(a) and 3(b), while Toussaint's group did not present any measured temperature profile. These refinements led us to rule out the photothermal convection as the dominant mechanism for the out-of-lattice transport.

Second, their array consisted of well separated antenna, which do not support the long-range transport of nanoparticles over the array. In contrast, the parameters in our work were chosen, such that the nanodisc diameter and lattice spacing were very close to the nanoparticle under trapping, to support the in-lattice transport, similar to the work by Grier^{16,17} and Dholakia.¹⁸ In addition, the extent of temperature distribution quantified by the confinement factor ζ was very different. For an assembly of M nanostructures, the temperature increase experienced by a given nanostructure j is given by $\Delta T_j^s + \Delta T_j^{\text{ext}}$, where ΔT_j^s and ΔT_j^{ext} are both self-contributed and contributed by other $M-1$ nanostructures. The confinement factor ζ is defined as $\Delta T_j^s/\Delta T_j^{\text{ext}}$, i.e., the ratio between ΔT_j^s and ΔT_j^{ext} . Baffou *et al.* showed that either the localized or the delocalized temperature profiles can be obtained, depending on the detailed lattice parameters, such as lattice spacing, length of the nanostructure, and dimensionality. In Toussaint's work, the laser beam was tightly focused on a diffraction-limited spot with a size of $H \sim 0.61\ \lambda/\text{NA}$, for illumination ($\lambda = 860\ \text{nm}$, $\text{NA} = 0.6$). The confinement factor for a Gaussian illumination of full-width half maximum H , can be estimated using the formula given by Baffou *et al.*, $\zeta = p^2/3HL$. Here L and p are the characteristic length of the plasmonic nanostructure and the distance between the plasmonic nanostructure for an infinite array, respectively.²⁴ Using $L \approx 100\ \text{nm}$, $p = 425\ \text{nm}$, and $H = 860\ \text{nm}$, we estimated the confinement factor in Toussaint's experiments to $\zeta = 0.7$; we could therefore conclude that their temperature distribution was strongly localized, which is consistent with their calculated temperature profile. In contrast, our temperature distribution was highly delocalized, with a confinement factor of $\zeta \approx 0.07$, using $L \sim d/2 = 550\ \text{nm}$, $p = 750\ \text{nm}$, and $H = 9.3\ \mu\text{m}$.

IV. CONCLUSION

In conclusion, the described study enabled detailed characterization of the transport of nanoparticles in a two-dimensional simple square nanoscale plasmonic lattice. The results explicitly showed that at low optical power, the delocalized transport was indeed induced by the near-field effects of the nanoplasmonic optical lattice. In addition, the delocalized temperature distribution was computed both analytically and numerically and found it to be in fair agreement with the measured temperature distribution. The resulting fluidic convection velocity was then calculated and found to be an order of magnitude smaller than the measured particle out-of-lattice velocities ($\sim \mu\text{m}$). Thus, particle movement was proposed to be mainly due to the enhanced evanescent field of the plasmonic array confined within the ITO layer. Our hope is that this work will serve as a benchmark experiment to elucidate the role of photothermal convection in nanoparticle transport in plasmonic optical lattices. The experimental demonstration of plasmonics-induced transport and photothermal heating in this work will be useful for a growing number of applications in nanobiotechnology. The plasmonic optical lattice will be an ideal tool for trapping and manipulating viruses and vesicles.^{5,40,41} The size of a wide repertoire of known viruses and important biological entities, in the micrometer to nanometer ranges, match well with the trapping ranges of plasmonic optical lattices. Extracellular vesicles hold great potential for diagnostic and therapeutic applications. Similarly, integration of a lab on a chip device with a plasmonic optical lattice will generate reliable disease prediction tools, which avoid the use of time-consuming and cumbersome processes involving ultracentrifugation. Another possible application is to concentrate and support the optofluidic cultivation of photosynthetic bacteria, using an enhanced evanescent plasmonic field.^{42–44} Excessive

photothermal heating may adversely affect the microbial growth, and must be controlled, for example, by engineering proper heat-sinks.⁴⁵ The work reported here represents a significant step toward these goals.

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- ¹A. Ashkin, J. M. Dziedzic, J. E. Bjorkholm, and S. Chu, *Opt. Lett.* **11**, 288 (1986).
- ²D. Grier, *Nature* **424**, 810 (2003).
- ³M. L. Juan, M. Righini, and R. Quidant, *Nat. Photonics* **5**, 349 (2011).
- ⁴T. Shoji and Y. Tsuboi, *J. Phys. Chem. Lett.* **5**, 2957 (2014).
- ⁵J. S. Huang and Y. T. Yang, *Nanomaterials* **5**, 1048 (2015).
- ⁶W. H. Wright, G. J. Sonek, and M. W. Berns, *Appl. Rev. Lett.* **63**, 715 (1993).
- ⁷G. Volpe, R. Quidant, G. Badenes, and D. Petrov, *Phys. Rev. Lett.* **96**, 238101 (2006).
- ⁸M. Righini, A. S. Zelenina, C. Girard, and R. Quidant, *Nat. Phys.* **3**, 477 (2007).
- ⁹M. Righini, G. Volpe, C. Girard, D. Petrov, and R. Quidant, *Phys. Rev. Lett.* **100**, 186804 (2008).
- ¹⁰T. Shoji *et al.*, *J. Am. Chem. Soc.* **135**, 6643 (2013).
- ¹¹Y. Pang and R. Gordon, *Nano Lett.* **12**, 402 (2012).
- ¹²A. Kotnala and R. Gordon, *Biomed. Opt. Express* **5**, 1886 (2014).
- ¹³A. A. Al Balushi and R. Gordon, *ACS Photonics* **1**, 389 (2014).
- ¹⁴A. A. Al Balushi and R. Gordon, *Nano Lett.* **14**, 5787 (2014).
- ¹⁵S. Wheaton, R. M. Gelfand, and R. Gordon, *Nat. Photonics* **9**, 68 (2015).
- ¹⁶P. T. Korda, M. B. Taylor, and D. G. Grier, *Phys. Rev. Lett.* **89**, 128301 (2002).
- ¹⁷K. Ladavac, K. Kasza, and D. G. Grier, *Phys. Rev. E* **70**, 010901 (2004).
- ¹⁸G. C. MacDonald, G. C. Spalding, and K. Dholakia, *Nature* **426**, 421 (2003).
- ¹⁹A. Cuche, B. Stein, A. Canguier-Durand, E. Devaux, C. Genet, and T. W. Ebbesen, *Nano Lett.* **12**, 4329 (2012).
- ²⁰K. Y. Chen, A. T. Lee, C. C. Hung, J. S. Huang, and Y. T. Yang, *Nano Lett.* **13**, 4118 (2013).
- ²¹G. Baffou, R. Quidant, and J. F. Garcia de Abajo, *ACS Nano* **4**, 709 (2010).
- ²²G. Baffou, R. Quidant, and C. Girard, *Phys. Rev. B* **82**, 165424 (2010).
- ²³J. S. Donner, G. Baffou, D. McCloskey, and R. Quidant, *ACS Nano* **5**, 5457 (2011).
- ²⁴G. Baffou *et al.*, *ACS Nano* **7**, 6478 (2013).
- ²⁵B. J. Roxworthy, A. M. Bhuiya, S. P. Vanka, and K. C. Toussant, Jr., *Nat. Commun.* **5**, 3173 (2014).
- ²⁶B. J. Roxworthy, K. D. Ko, A. Kumar, K. H. Fung, E. K. C. Chow, G. L. Liu, N. X. Fang, and K. C. Toussaint, Jr., *Nano Lett.* **12**, 796 (2012).
- ²⁷J. C. Ndukaife *et al.*, *ACS Nano* **8**, 9035 (2014).
- ²⁸M. Pelton, K. Ladavac, and D. Grier, *Phys. Rev. E* **70**, 031108 (2004).
- ²⁹See supplementary material at <http://dx.doi.org/10.1063/1.4948775> for Matlab codes and additional data.
- ³⁰D. Ross, M. Gaitan, and L. E. Locascio, *Anal. Chem.* **73**, 4117 (2001).
- ³¹F. P. Incropera and D. P. DeWitt, *Fundamentals of Heat and Mass Transfer*, 4th ed. (John Wiley & Sons, New York, 1996).
- ³²F. Reif, *Fundamentals of Statistical and Thermal Physics*, 1st ed. (McGraw-Hill, New York, 1965).
- ³³A. Pralle, E.-L. Florin, E. H. K. Stelzer, and J. K. H. Hörber, *Appl. Phys. A* **66**, S71 (1998).
- ³⁴J. Happel and H. Brenner, *Low Reynolds Number Hydrodynamics* (Kluwer, Dordrecht, 1991).
- ³⁵J. Kestin, M. Sokolov, and W. A. Wakeham, *J. Phys. Chem. Ref. Data* **7**, 941 (1978).
- ³⁶S. Kawata and T. Sugirua, *Opt. Lett.* **17**, 772–774 (1992).
- ³⁷H. Kim *et al.*, *J. Appl. Phys.* **86**, 6451–6461 (1999).
- ³⁸A. Yariv, *Quantum Electronics* (John Wiley & Sons, New York, 1988).
- ³⁹S. Murai, M. A. Verschuuren, G. Lozano, G. Pirruccio, S. R. K. Rodriguez, and J. Gómez Rivas, *Opt. Express* **21**, 4250–4262 (2013).
- ⁴⁰S. Jane Flint, W. Enquist, V. R. Racaniello, and A. M. Skalka, *Principles of Virology* (ASM Press, Washington, 2009).
- ⁴¹E. Van der Pol, A. N. Böing, P. Harrison, A. Sturk, and R. Nieuwland, *Pharm. Rev.* **64**, 676–705 (2012).
- ⁴²M. D. Ooms, L. Bajin, and D. Sinton, *Appl. Phys. Lett.* **101**, 253701 (2012).
- ⁴³D. Erickson, D. Sinton, and D. Psaltis, *Nat. Photonics* **5**, 583–590 (2011).
- ⁴⁴R. A. Anderson, *Algal Culturing Technique*, 1st ed. (Academic Press, London, 2005).
- ⁴⁵K. Wang, E. Schonbrun, P. Steinvurzel, and K. B. Crozier, *Nat. Commun.* **2**, 469 (2011).