Transport and Trapping in Two-Dimensional Nanoscale Plasmonic Optical Lattice

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Supporting Information

ABSTRACT: We report the transport and trapping behavior of 100 and 500 nm diameter nanospheres in a plasmon-enhanced two-dimensional optical lattice. An optical potential is created by a two-dimensional square lattice of gold nanostructures, illuminated by a Gaussian beam to excite plasmon resonance. The nanoparticles can be guided, trapped, and arranged using this optical potential. Stacking of 500 nm nanospheres into a predominantly hexagonal closed pack crystalline structure under such a potential is also reported.

KEYWORDS: Plasmonics, optical lattice, optical sorting, optical tweezer, Brownian motion, microfluidics

Recent advances in optical trapping techniques have enabled the creation of one-, two-, and three-dimensional periodic potentials (or optical lattices) at the microscale. These optical potentials have led to numerous interesting fundamental studies and technical applications including the thermal ratchet, the washboard potential, kinetic lock-in, optical sorting of microparticles, and the optical crystallization and binding of microparticles. The optical trapping and manipulation of microscale and nanoscale particles was pioneered by Ashkin et al. in the early 1970s. Far-field optical trapping techniques, such as traditional optical traps, face trapping size limitations based on the diffraction limits; in particular, considering an object of radius \( a \), the trapping efficiency drops rapidly (following a \( \sim a^3 \) law) . To overcome these diffraction limits, researchers have developed near-field optical trapping techniques; the optical manipulation of nanoparticles has been demonstrated using waveguides, ring resonators, photonic crystal resonators, and plasmonic metallic nanostructures. Compared to photonic structures made from dielectric materials, metallic plasmonic nanostructures allow deep subwavelength concentration of the light and resonant enhancement of the optical field intensity. Trapping of a wide range of objects such as bacteria, micro- and nanoparticles, and even as small as a single protein molecule has been demonstrated. Metallic plasmonic structures also present an opportunity for large-scale integration of lab-on-a-chip applications. Recently, optical sorting of metallic nanoparticles based on negative refraction effect of a plasmonic crystal has been reported. Here we present a more direct approach to create a periodic potential by simply illuminating a two-dimensional array of plasmonic nanostructures and study the transport and trapping behavior of the nanospheres under such optical potentials. Our experiment has been inspired by the pioneering work conducted by Dholakia’s group and is essentially a scaled-down incarnation of their washboard optical potential created by a Bessel-Gaussian beam, albeit through the application of plasmonics.

To create our two-dimensional plasmonic array, we pattern metallic nanostructures on an ITO-coated (indium tin oxide) cover glass by electron beam lithography, thermal evaporation, and adhesion layer technique. Adhesion layer is not used in order to minimize the damping of localized surface plasmon resonance. We define our periodic potential by using a lattice and associate each lattice point with a primitive cell. In this experiment, we have chosen a square lattice with period \( 1 \) \( \mu m \), and each primitive cell contains four disks of diameter \( D = 200 \) nm. Typically, \( 7 \times 7 \) arrays are used for the data presented here, as shown in Figure 1a. To excite the plasmonic resonance, we have built a custom optical setup by modifying a commercially available optical trap kit (OTKB, Thorlabs Inc.; for details, see Supporting Information). An infrared diode laser (PL980P330J, Thorlabs) with a wavelength of 980 nm and a fiber output is fed into and only partially overfills the rear aperture of a high numerical aperture oil immersion microscope objective (NA = 1.25, \( \times 100 \), Nikon) for the beam to be loosely focused on the gold nanostructure array. The objective in conjunction with a dichroic filter set (41001, Chroma Technology) is also used for the imaging of fluorescent polystyrene nanospheres (Invitrogen) using 470 nm LED light (Touchbright) as an excitation source. The

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scattering from the trapping laser is rejected by using a long pass filter (FM01, Thorlabs). Two nanosphere sizes with diameters \( d_1 = 500 \text{ nm} \) and \( d_2 = 100 \text{ nm} \) are used in our experiments. These nanospheres are dispersed in deionized water and are dispensed on the gold nano array sample. The motion of the nanospheres is then recorded using a charge-coupled device (CCD) camera (DCU224C, Thorlabs Inc.).

Our array is illuminated with an expanded Gaussian beam with an intensity profile defined by \( I = I_0 \exp(-2r^2/w^2) \), where \( r \) is the azimuthal radius and the spot size \( w \) is \( \sim 7.4 \mu \text{m} \). We have performed finite-difference time-domain (FDTD) simulations of the electromagnetic field distribution within a primitive cell. The structure is designed to resonate at a wavelength of 980 nm. The representative optical intensity profiles of a primitive unit cell simulated by FDTD method at 55, 90, and 290 nm above the substrate are shown in Figure 1b. Note that the plasmon modes of each nanodisk within a primitive cell couples with each other due to the small gap (\( \sim 48 \text{ nm} \)) between the two neighboring nanodisks. Within each cell at 90 nm above the substrate, four pronounced trapping sites are produced under excitation by linearly polarized light. The density of these trapping sites is therefore \( 4 \text{ sites}/\mu \text{m}^2 \), which is more than 20 times the density of a previous diffraction-limited two-dimensional optical lattice created by a holographic technique that was reported in the literature.\(^7\) The center of the Gaussian beam is aligned with the center of the array. With such an illumination profile, the representative optical intensity profiles can be calculated based on the intensity data from the FDTD simulation, as shown in Figure 1c. The optical lattice has a spatially dense periodic potential from each primitive cell with a spatially slowly varying Gaussian envelope.

Single particle trajectories are recorded in our experiment and the images are then processed frame by frame with the Matlab program to extract each particle trajectory.\(^8\) Representative results are displayed in Figure 2a,b for nanospheres with diameters of 500 and 100 nm, respectively (see also the Supporting Information for movies). Because we can intuitively understand that the particles tend toward the lowest potential energy region in the central region of the maximum optical intensity, we expect them to be attracted to and trapped within the central region of the lattice and this is indeed what we observed. In general, the particles roaming around the optical lattice have a finite probability of being captured by the lattice. The particles, when captured by the lattice, exhibit a running state and are always attracted from the edge toward the central region of the lattice, regardless of the different routes taken, as shown in Figure 2a,b. This is very similar to the behavior that was previously reported for a washboard type potential with a Bessel-Gaussian beam shape.\(^7\)

Note that the presence of the plasmonic structure enables the trapping of nanospheres at such low optical power. This is especially striking if we estimate the optical potential energy \( W \) to be \( 4 \times 10^{-3} k_B T \) in the absence of the plasmonic structure for the nanospheres of diameter 100 nm (\( d \ll \lambda \)) in the center of the expanded Gaussian beam (see Supporting Information). Here \( k_B T \) is the thermal energy at room temperature and as a rule of thumb the optical potential at least \( \sim 10 k_B T \) is needed.
for stable trapping. Such an optical potential is too small to guarantee any observable trapping without plasmonic nanostructures. A control experiment is done on a bare glass without plasmonic nanostructures and with the same Gaussian illumination and no trapping of nanospheres of both sizes is observed because the optical power is too small to create a significant optical gradient force for trapping.

For quantitative comparison of the observed transport process with that of free diffusion, we define the square of the (two-dimensional) distance \( R^2 = (x - x_0)^2 + (y - y_0)^2 \), 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. We plot the square of the distance versus time in Figure 2a,c,d for the same trajectories that were shown in Figure 2a,b. As a comparison, the expectation value of the square of the free diffusion distance versus time, which is given by \( \langle R^2(t) \rangle = 2D_t t \), is also plotted with the free diffusion coefficient \( D_t = k_BT/\pi \eta d \), where \( k_B \), \( T \), \( \eta \), and \( d \) are the Boltzmann constant, the temperature, the viscosity of the medium, and the diameter of the spheres, respectively.33 At \( T = 298 \) K, we use \( \eta = 8.9 \times 10^{-4} \) Pa s for the viscosity of the water34 to obtain the diffusion coefficient \( D_t = 0.98 \) \( \mu^2/m/s \) when \( d = d_1 = 500 \) nm, and \( D_t = 4.9 \) \( \mu^2/m/s \) when \( d = d_2 = 100 \) nm. The observed transport process is obviously faster than that of free diffusion. For each trajectory displayed in Figure 2a,b, we can define the effective drift velocity \( v_{\text{eff}} \) by \( v_{\text{eff}} = R/T_r \), where \( R \) is the distance from the edge to the center and \( T_r \) is the corresponding passage time.35 With such a definition, the observed effective velocity ranges from 3.1 to 5.7 \( \mu m/s \) for nanospheres of diameter 500 nm and from 3.6 to 8.5 \( \mu m/s \) for nanospheres of diameter 100 nm (for details, see Supporting Information). Here, we did not observe significant difference in the velocity of two kinds of spheres. A plausible explanation might be that the stronger optical force experienced by larger spheres is compensated by larger frictional force due to Stoke drag, which is proportional to the particle diameter.34 We point out that a quantitative theoretical analysis involves not only calculating the optical potential from the plasmon electromagnetic field but also solving the problem of a particle in a Gaussian enveloped periodic potential. Although it is beyond the scope of this work, we believe that our single particle trajectory experiment can be best treated and simulated with the path integral formulation of the Fokker–Planck equation.36

Multiple particle trapping in the central region of the array is also observed for both particle sizes. Successive images extracted from a representative motion video of the particle are displayed in Figure 3a,b for nanospheres with diameters of 500 and 100 nm, respectively. For the 500 nm diameter nanospheres, the particles are tightly trapped by the plasmonic nanostructures, and stacking of the nanospheres is observed (see Supporting Information for movies). Similar particle stacking has been observed with a three-dimensional optical trap.40 Closer examination actually reveals that the nanospheres form a predominantly closely packed (hexagonal) crystalline structure, which is similar to that observed in the case of surface plasmon excitation.32 This is understandable since the particles tend to smear out the fine feature of the nanoscale potential as shown in Figure 1c for the optical intensity 290 nm above the substrate so the hexagonal phase bears no resemblance in terms of symmetry to the underlying optical potential, which has square crystalline structures. Similar symmetry breaking has also been observed for entropically driven self-assembly of colloidal particles on patterned substrates.37 Also, we observe slight fluorescent intensity fluctuations consistent with the result reported.38 In contrast, nanospheres of diameter 100 nm are more loosely trapped. Because of resolution limit, we can not count exact number of the trapped 100 nm particle nor can we clearly observe particle stacking.

In conclusion, we have shown that a plasmonic enhanced optical lattice can be used to guide and arrange nanoparticles. The experiments reported here can be extended to the study of other stochastic transport phenomena, such as the kinetic lockin of nanoparticles,9 and the realization of optical matter crystallization and binding.12 In principle, we can also achieve nanoparticle sorting based on optical fractionation.9,11 Compared to holographic or interference techniques,9,10 the plasmonic optical lattice has subwavelength spatial features and is considerably easier to set up with less complicated optical components and is therefore more suitable for integration with microfluidic systems for lab-on-a-chip applications.30 Finally, the scaling down of our experiment also has far reaching implications. Recently, several theoretical works have also explored the feasibility of creating plasmon-enhanced single-atom optical traps39–41 and optical field confinement down to the atomic scale has been reported.42 Scaling down of the plasmonic optical lattice certainly requires considerable engineering effort. It will obviously be necessary to litho-

**Figure 3.** Multiple particle trapping of nanospheres. (a) Successive fluorescent images of the trapped 500 nm nanospheres are displayed here. The laser illumination is initially turned on at \( t = 0 \) s. As time progresses, the particles accumulate in the central region and form predominant (hexagonal) closed pack structures. Finally, at \( t = 34 \) s, the laser illumination is turned off and the particles disperse and escape from the array. The total incident laser power is \( \sim 4.75 \) mW. (b) Same as (a) but for 100 nm diameter nanospheres. At \( t = 0 \) s, the laser illumination is turned on. At \( t = 4 \) s, the first particle is captured and is trapped in the central region. As time progresses, the particles accumulate in the central regions but are only loosely trapped. When the laser illumination is turned off at \( t = 46 \) s, the particles escape from the array. The total incident laser power is \( \sim 3.5 \) mW (see Supporting Information for movies).
graphically reduce the feature sizes of the devices. Also, the device surfaces must be passivated to prevent unwanted adsorption of the trapped objects and appropriate thermal management is required to prevent excessive ohmic heating as a result of light illumination.\(^{43}\) However, we believe that the results of these efforts will be very rewarding; these optical lattices will ultimately enable atomic physicists to study collective coherent interactions and explore novel many-body physics problems.

**ASSOCIATED CONTENT**

**Supporting Information**

Additional information, figure, tables, and movies. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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This article provides supplementary information to the main text of the manuscript. In Section I, we present the details of the experimental setup. In Section II, we present the detailed data extracted from the single particle trajectory.
I. Experimental setup

1. Experimental setup

Figure S1. Experimental setup. A fiber-coupled laser diode (PL980P330J) of wavelength 980 nm is expanded through beam expansion, directly through a long pass filter M1 (FM01, Thorlabs) and finally loosely focused by an oil immersion microscope objective (NA=1.25, ×100, Nikon) to excite the plasmonic sample. The spot size $w$ of the Gaussian beam, defined by $I=I_0 \exp(-2r^2/w^2)$, is obtained by fitting the transverse intensity distribution of the CCD camera (DCU224, Thorlabs) image. The fluorescent image is taken with same objective in conjunction with dichroic mirror (DM) and emission filter (41001, Chroma Technology) under the fluorescent excitation at 470 nm from LED light source (Touchbright). The motion is recorded with the CCD camera with either 10 or 15 frames per second. A three dimensional stage is used to move and align the sample with respect to the incident Gaussian beam. The mechanical design is modified from a commercial optical trap kit (OTKB1, Thorlabs). All the optical and optomechanical components such as lens, mirrors and 30 mm cage mounts are purchased from Thorlabs Inc.

II. Notes on theoretical Analyses

1. Effective drift velocity and passage time for single particle trajectory

Here we tabulate the effective drift velocity and passage time for the single trajectory data displayed in Figure 2(a) and (b). The effective drift velocity is defined as $v_{\text{eff}} = R / T_p$ where the
passage time, $T_p$, is defined as the time for the particle to enter from the edge of the array and reach the center of the array.$^2$

<table>
<thead>
<tr>
<th>Color</th>
<th>Distance (µm)</th>
<th>Passage time (sec)</th>
<th>Effective drift velocity (µm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blue</td>
<td>3.1</td>
<td>1</td>
<td>3.1</td>
</tr>
<tr>
<td>Cyan</td>
<td>4.5</td>
<td>0.8</td>
<td>5.7</td>
</tr>
<tr>
<td>Green</td>
<td>3.1</td>
<td>1</td>
<td>3.1</td>
</tr>
<tr>
<td>Black</td>
<td>4.2</td>
<td>1</td>
<td>4.2</td>
</tr>
<tr>
<td>Red</td>
<td>3.0</td>
<td>0.9</td>
<td>3.3</td>
</tr>
</tbody>
</table>

Table S1. List of parameters of single particle trajectory for nanospheres of diameter 500 nm in Figure 2(a) in the main text. The distance, passage time and effective drift velocity are displayed here. Color here refers to the color code used in labeling the trajectory in Figure 2(a).

<table>
<thead>
<tr>
<th>Color</th>
<th>Distance (µm)</th>
<th>Passage time (sec)</th>
<th>Effective drift velocity (µm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blue</td>
<td>3.7</td>
<td>0.67</td>
<td>5.6</td>
</tr>
<tr>
<td>Cyan</td>
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<tr>
<td>Green</td>
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<td>0.40</td>
<td>8.5</td>
</tr>
<tr>
<td>Black</td>
<td>4.6</td>
<td>1.27</td>
<td>3.6</td>
</tr>
<tr>
<td>Red</td>
<td>3.3</td>
<td>0.47</td>
<td>7</td>
</tr>
</tbody>
</table>

Table S2. List of parameters of single particle trajectory for nanospheres of diameter 100 nm in Figure 2(b) in the main text. The distance, passage time, and effective drift velocity are displayed here. Color here refers to the color code used in labeling the trajectory in Figure 2(b).
2. Information for video files

We tabulate the content of the four video files attached.

<table>
<thead>
<tr>
<th>Movie files</th>
<th>Content</th>
</tr>
</thead>
<tbody>
<tr>
<td>S2</td>
<td>Representative single particle trajectory movie of nanosphere of diameter 500 nm</td>
</tr>
<tr>
<td>S3</td>
<td>Representative single particle trajectory movie of nanosphere of diameter 100 nm</td>
</tr>
<tr>
<td>S4</td>
<td>Multiple particle trapping movie of nanosphere of diameter 500 nm</td>
</tr>
<tr>
<td>S5</td>
<td>Multiple particle trapping movie of nanosphere of diameter 100 nm</td>
</tr>
</tbody>
</table>

Table S3. The content of movie files. Video are compressed to reduce the file size and adjusted in brightness and contrast to show the underlying nanostructure arrays used. Typically, two arrays are displayed in the movie and only the upper array is illuminated with laser beam and shows trapping behavior.

References

1 Optical trap kit manual, Thorlabs website.