Light-Assisted, Templated Self-Assembly Using a Photonic-Crystal Slab

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Supporting Information

ABSTRACT: We experimentally demonstrate the technique of light-assisted, templated self-assembly (LATS). We excite a guided-resonance mode of a photonic-crystal slab with 1.55 μm laser light to create an array of optical traps. We demonstrate assembly of a square lattice of 520 nm diameter polystyrene particles spaced by 860 nm. Our results demonstrate how LATS can be used to fabricate reconfigurable structures with symmetries different from traditional colloidal self-assembly, which is limited by free energetic constraints.

KEYWORDS: Optical trapping, self-assembly, photonic crystal, nanomanipulation

The force of light on objects provides tremendous flexibility for nanoscale manipulation. While conventional optical tweezers use the optical gradient force of a focused laser,1−3 recent work has leveraged the strong field gradients near microphotonic devices for particle trapping.4−15 However, such work has focused on trapping single or few particles. We have proposed to use optical forces near microphotonic devices for a fundamentally different purpose: to assemble periodic arrays of nanoparticles resembling synthetic, reconfigurable two-dimensional (2D) crystals.16,17 Our approach, called light-assisted, templated self-assembly (LATS), exploits photonic-crystal slabs to create resonantly enhanced optical forces orders of magnitude larger than radiation pressure.16,17 Here we provide the first experimental demonstration of LATS, assembling a square array of over 100 polystyrene particles near a silicon photonic-crystal slab. Our method, ideally suited for on-chip integration, should provide a platform for flow-through, serial fabrication of 2D or 3D-nanostructured materials, all-optically tunable photonic devices, and lab-on-a-chip applications.

The LATS process is shown schematically in Figure 1. Light is incident from below on a photonic-crystal slab, which consists of a silicon device layer patterned with a periodic array of air holes. The slab is designed to support guided-resonance modes, electromagnetic modes for which the light intensity near the slab is strongly enhanced.18 Our previous work has theoretically predicted19 that when the incident laser is tuned to the wavelength of a guided-resonance mode, nanoparticles will be attracted toward the slab. The attractive, optical force arises from a strong electric-field gradient just above the slab surface. In addition, the nanoparticles will experience lateral optical forces due to the electromagnetic field structure of the guided-resonance mode, resulting in the assembly of a nanoparticle array.

Unlike traditional colloidal self-assembly for which free energy minimization results in hexagonal, close-packed structures, our process is not subject to such constraints. In this paper, we experimentally demonstrate the formation of a square lattice as one such example. Indeed, the use of light to drive the system dramatically alters the underlying potential landscape, potentially allowing for the formation of a range of complex lattices16,17 and multiparticle clusters.19 Nanoparticle arrays assembled using LATS can be viewed as “programmable
The magnetic field from the light source with the trapping device and the background, oriented at 45° with respect to the camera. (a–c) Sequential snapshots taken with the light beam on. (d) Snapshot taken after the beam is turned off.

We have explored the dependence upon power, wavelength, and particle diameter. The size of the cluster, that is, the number of particles trapped, decreased in a monotonic fashion as the power was decreased, or as the wavelength was tuned away from resonance. We also attempted to assemble two other sizes of polystyrene particles, 380 nm and 1 μm. We were able to assemble a square lattice of 380 nm particles. The trapped particles exhibited more Brownian motion when compared to the 520 nm particles. We did not observe trapping with 1 μm optical matter turning the laser on and off will reversibly assemble or disassemble the structure. Moreover, exciting different resonance modes of the photonic crystal, by adjusting the wavelength of the input laser, should allow different crystalline structures to be formed.

Light-driven assembly of multiparticle patterns has previously been achieved using structured light fields generated by interference fringes, holography, spatial light modulators, or other methods. Our approach differs crucially from previous work in that it exploits near-field, rather than far-field, effects. Rather than generating a structured light beam via free-space optics, we use a simple, Gaussian input beam. The structured light field responsible for trapping is generated by the interaction of light with the photonic-crystal device. Here, we demonstrate the method using an external laser incident on a photonic-crystal slab. Ultimately, however, LATS could be carried out using a photonic-crystal laser, allowing the integration of the light source with the trapping device and making our approach highly suitable for on-chip integration. We thus expect a wide range of applications from all-optically tunable photonic devices, to materials assembly, to biological trapping and manipulation.

We designed, fabricated, and characterized a photonic-crystal slab for use in the LATS process. An electron micrograph is shown in Figure 2a. The device was fabricated in silicon using electron-beam lithography and reactive ion etching (see Supporting Information Methods). The dimension and spacing of the holes was designed to support doubly degenerate guided-resonance modes near 1.55 μm. The lattice constant a is 860 nm, and the hole radius is 0.174 Å (150 nm). The thickness of the silicon device layer is 250 nm. The magnetic field profile resulting from an x-polarized, incident plane wave is shown in Figure 2b. Fields were calculated using the three-dimensional finite-difference time-domain method (FDTD).

Figure 2. Square lattice photonic crystal. (a) SEM image of photonic-crystal slab. The scale bar in the inset is 1 μm. (b) A 3D FDTD simulation of the magnetic field (|H|) for a normally incident, x-polarized plane wave. Circles represent the positions of holes; four unit cells are shown. (c) Measured transmission spectrum (log scale).

Figure 3. Light-assisted, templated self-assembly of 520 nm diameter particles above a photonic-crystal slab. The square lattice of the slab is visible in the background, oriented at 45° with respect to the camera. (a–c) Sequential snapshots taken with the light beam on. (d) Snapshot taken after the beam is turned off.
particles. This is likely due to the increased scattering cross-section, which results in a net repulsive force away from the slab.

Each site of the square lattice may be viewed as an optical trap. We used particle-tracking software to analyze particle motion for fully assembled clusters (see Supporting Information Methods). Figure 4a shows the recorded particle positions extracted from a 20 s video. The incident light was polarized along the x-direction of the lattice. The figure shows that the particles tend to stay above the holes in the photonic crystal with some variation in position over time. Each blue ellipse represents a fit to the data in a single unit cell. It can be seen that the variation in particle position increases at the edge of the trapping region due to the reduction in power away from the center of the beam.

The stiffness of each trap can be determined from the variance in particle position. Figure 4b,c shows histograms of the in-plane stiffness values extracted from the videos. The stiffness of each trap was normalized to the local intensity in that unit cell (see Supporting Information Methods). We observe that the power-normalized stiffness over the array of traps is normally distributed, both for the parallel and perpendicular stiffness. The mean parallel stiffness is lower than the perpendicular stiffness, as shown in Table 1 (0° angle).

We observe that the trap stiffness can be tuned by rotating the direction of incident light. When the incident light is polarized at 45° with respect to the lattice directions, the stiffness values are approximately the same in the x- and y-directions (Table 1). This is to be expected, since the incident light excites both of the doubly degenerate modes with equal strength. At 90°, the stiffness in the parallel direction (y) is again lower than in the perpendicular (x) direction. The ability to tune the stiffness with incident light indicates the strong optical nature of our traps. The mean values of stiffness are comparable to those reported elsewhere in the literature for single particle traps.

Using the Stokes drag method (see Supporting Information Methods), we experimentally estimated the maximum force exerted on the particles by the traps to be 0.3 pN.

To understand how the optical forces result in the observed nanoparticle patterns, we calculated the force numerically (see Supporting Information Methods). Figure 5a shows the force on a 520 nm diameter particle whose bottom edge is 25 nm above the surface of the photonic crystal slab for x-polarized light. The background color represents the vertical force, where a negative force indicates attraction toward the slab. There are two regions above and below the hole where the force is slightly repulsive, but at any other position within the unit cell the particle is attracted to the slab. The arrows represent the in-plane forces.

To determine the equilibrium position of the trapped particle, we calculate the optical potential. Given the relative size of the particles (260 nm radius) and holes (150 nm radius) in our experiment, the particle can be partially drawn into the hole by the attractive vertical force. We calculate the optical potential as a function of x−y position. For each x−y position, the vertical height of the particle is as small as possible, given the geometrical constraints (see inset in Figure 5b). The result is shown by the blue line (“total potential”) in Figure 5b. It can be seen that the stable equilibrium position is in the center of the hole (x = 0, y = 0), which is in agreement with experiments.

The ability of the particle to sink into the hole is a key factor in determining the equilibrium positions. For comparison, the red, dashed line (“in-plane potential”) in Figure 5b shows the optical potential calculated at a constant z-height (bottom edge of particle 25 nm above the slab surface). Two local minima are observed at the edges of the hole, which are indicated by green, dashed lines. From inspection of Figure 5a, it can be seen that these points correspond to locations where the in-plane forces are zero. However, these two minima are not stable equilibrium positions of the total potential (Figure 5b).

From our experiments, we determined that the threshold intensity for trapping was 134 μW per unit cell (see Supporting Information Methods). For this intensity, the calculated potential depth is 4.5/k_B T.

In summary, we have experimentally demonstrated the technique of light-assisted, templated self-assembly (LATS). Our technique uses the resonantly enhanced near field of a photonic-crystal slab to create periodically spaced optical traps. Nanoparticles in solution are attracted to the slab and form ordered arrays. We have observed the assembly of 100 polystyrene particles with 520 nm diameter in a square lattice using 64 mW of incident power. We have measured the trapping stiffness as a function of incident polarization, and we have shown that the equilibrium trapping positions can be predicted via calculation of the optical forces.

Our technique can be extended to assemble larger numbers of particles by designing the photonic-crystal slab to reduce the input power per area required for trapping. The input beam can then be spread over a larger area, resulting in a larger cluster. One approach to reducing the required power per area is to use a mode with a higher quality factor, resulting in higher near-field intensity. Another approach is to use slot-confinement effects to strongly localize the field in the trapping regions, as we have previously studied theoretically.17

The LATS approach can be used to assemble complex structures with symmetries not constrained by the typical free energetic constraints; here we have demonstrated just one. We
envision that judicious template design will allow the assembly of a variety of lattice types with complex unit cells. Moreover, changing the wavelength of the incident beam to excite a different resonance can be used to reconfigure the particle arrangement. The use of metal nanoparticles or quantum dots such effects may yield rich assembly behavior in alternate particle systems.

We anticipate that our technique will find applications in the fabrication of metamaterials and other photonic devices. For example, postassembly polymerization could be used to transfer 2D nanoparticle arrays to another substrate. Our preliminary results indicate that it is also possible to assemble 3D arrays in situ.

LATS should also allow a variety of dynamic, real-time applications. One example is the use of the assembled, reconfigurable “photonic matter” as an all-optically tunable transmission filter. Other applications, based on particle dynamics in the 2D optical potential, include particle sorting and ratchet behavior. We also expect that LATS may be extended for batch processing of biological objects, providing a novel tool for reconfigurable control over spatially mediated biological interactions.

Finally, LATS naturally lends itself to compact integration on-chip. By fabricating the photonic-crystal device in an active material, the light source could be integrated with the trapping device, allowing for system miniaturization.

**REFERENCES**