ABSTRACT

We present preliminary results from an experimental study of optically-assisted assembly. Interference patterns, formed by the intersection of two coherent laser beams, result in periodic one-dimensional potential wells (“optical trenches”). Polystyrene spheres become trapped in these potential wells and subsequently self-assemble into a two-dimensional periodic structure. The spacing between optical trenches is adjusted dynamically, which offers the freedom to dynamically control the lattice constant, offering a recipe for defect-free assembly which begins with annealing at large lattice constant and subsequent compression into a close-packed structure.

Keywords: Optical tweezers, laser trapping, self-assembly, one-dimensional optical lattice

1. INTRODUCTION

Optical micromanipulation techniques have been continuously developing since the work of Ashkin\(^1\) in the 1970s. Light has become\(^2\) a highly practical, flexible, non-invasive tool for positioning, measuring, and controlling microscopic matter from the scale of hundreds of microns down to ångströms. A huge diversity of utilization has emerged, ranging from physics\(^3,6\) and material science\(^7,8\) to microfluidics\(^9,10\) and medical science\(^11,12\). However, emphasis has largely remained upon manipulation of either individual particles or a very limited number of particles (typically not more than two). In this paper, we report the application of a laser micromanipulation system for defect-free two-dimensional assembly of an ensemble of polystyrene spheres.

Alternative methods for assembly of two-dimensional periodic structures have been in high demand recently, with much of the interest centered upon applications in electronics and photonics\(^13\). Unfortunately, although a variety of techniques – from liftoff or subtractive photolithography and e-beam writing, to particle self-assembly – have proven successful in preparing such structures, there persists a trade-off between cost and defect density\(^14\). In this letter, we report the optically-assisted assembly of polystyrene spheres with diameters of three micrometers, emphasizing a self-healing characteristic of (tunable) optically-bound assemblies: in particular, we examine the merger of interstitial particles into the periodic superstructure, and make comparisons to methods of self-assembly.

There are many optical geometries which can provide for laser micromanipulation, ranging from something as simple as a single, tightly focused Gaussian beam\(^15\) (“optical tweezers”) to systems involving interferometers\(^16\), computer-generated diffraction patterns\(^17,18\), and spatial light modulators\(^19,20\). In our experiments, we generate a periodic optical field (a one-dimensional interference pattern) from two tightly focused coherent laser beams, which originate from a single laser beam. The spacing of the interference maxima is given by \(d = \frac{\lambda}{2\sin(\theta/2)}\), where \(d\) is the period, \(\lambda\) is the wavelength of the incident laser, and \(\theta\) is the angle between the two beams. This interference provides strong gradients along one direction. Weaker, two-dimensional gradients are generated simultaneously by focusing the input, describable by the usual Gaussian intensity profile: \(I = I_0 e^{-\rho^2}\), where \(I\) is the light intensity, \(I_0\) is the intensity at beam center and \(\rho\) is the distance from the beam center, in units of the beam waist diameter.

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We apply this light field to dielectric colloids—here, polystyrene spheres suspended in water. The polarizability of the spheres implies a position-dependent energy within the optical lattice. The strong gradient force will organize the spheres onto periodic lines and the weak gradient force in the normal direction will prevent the particles from diffusing out along each line.

The direct optical forces can be approximately written as:

$$F_{\text{grad}} = 2\pi a^3 \frac{\sqrt{\varepsilon_0}}{c} \left( \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right) \nabla |S|,$$

(1)

And

$$F_r = \frac{8}{3} \pi (ka)^4 a^2 \frac{\sqrt{\varepsilon_0}}{c} \left( \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right)^2 |S|,$$

(2)

where $F_{\text{grad}}$ is the gradient force, $F_r$ is the radiation force resulting from backscattering of light, $S$ is the Poynting vector of the light field, $a$ is the radius of the polystyrene particle, $k$ is the wave vector of the incident light, $\varepsilon$ and $\varepsilon_0$ are the dielectric constants of the particle and the water, respectively. Although Harada and Asakura derived these for Rayleigh particles, it was shown that these forms remain quite good approximations for particles sizes approaching the wavelength, at least in the paraxial limit.22

The role of radiation pressure in our traps is predominantly longitudinal—pushing the particles against the far wall of the sample cell. As our primary point of interest has to do with organization along the transverse directions, it is reasonable to focus attention upon the gradient force only. Assuming the intensity does not vary along the optic axis over the scale of the trapped particles, a zeroth-order approximation for the restoring force acting against lateral motion, is:

$$|F_{\text{grad}}| = 16\pi a^3 \frac{\sqrt{\varepsilon_0}}{c} \left( \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right) \left| I_0 \left( \frac{\pi}{d} \right) \sin \delta \right| \hat{x} - \hat{\rho} | \hat{\rho} |,$$

(3)

where $\hat{x}$ and $\hat{\rho}$ are the in-plane unit vectors perpendicular to the interference fringes and along the radial direction, respectively, $d$ is the spacing of interference fringe maxima and $\delta$ is the phase factor across the transverse direction. Noting that $\rho$ is measured in units of the beam waist, the $\hat{\rho}$ component, over the size of the particles, is much smaller than the transverse $\hat{x}$ force component. The factor of $(\varepsilon - \varepsilon_0)/(\varepsilon + 2\varepsilon_0)$ shows that a large potential barrier will be experienced by particles having significant refractive index contrast with respect to the surrounding medium.

2. EXPERIMENTAL INSTRUMENTATION

In most other approaches to laser micromanipulation, sensitivity to aberrations presents significant experimental challenges during alignment. We have constructed an interferometric apparatus that allows the optical lattice constant to be easily reconfigured, without any re-alignment. Shown in Fig. 1, the stage, derived from a General Electric x-ray diffractometer, has an internal cam designed to yield $\theta / 2\theta$ scans, but here is utilized to provide symmetric, tunable angles of incidence for two input beams. The large-circle goniometer is computer-controlled for precise angular control, down to 0.05° increments. The incoming beam is divided with a beam splitter, indicated as BS in Fig. 1, in such a way that the two beams are reflected from an equal number of optical surfaces (involving two sets of identical mirrors and prisms), focused by a single short focal length lens ($f = 18$ mm) down to 60 microns to increase intensity, and meet each other at the specimen surface with the same $s$-polarization. A translation stage, indicated as TS, in one branch is used to adjust the optical delay of one of the beams, so as to equalize the optical path lengths of the two beams: under these conditions the coherence of the two beams is maximized, and yields approximately equal intensity for each of the interference maximums. As the $\theta / 2\theta$ system rotates, the incident angles of the two light beams are tuned, but kept...
symmetrical, with the net result of continuously adjusting the period of the interference pattern, while all optics are automatically kept aligned.*

We use an argon laser (Spectra-Physics 260) operating in the TEM$_{00}$ mode, at a wavelength of 514.5 nanometers, and a maximum output power of about 2.0 W. The laser beam was first collimated with a long focal length lens (focal length 1500 mm), as shown in Fig. 2. After splitting the beam and focusing through the optics onto the sample stage shown in Fig. 1, the interference pattern was formed in the middle of a microfluidic sample cell.

The sample cell was made from two microscope cover slips (each 83 micron thick). Before making the cell, the surfaces of the cover glass were soaked and washed with de-ionized water. We deposit approximately 5 micro-liters of colloidal solution (3-micron polystyrene spheres suspended in de-ionized water) onto one cover slip before sealing the cell; the thickness of the fluid layer was 80 microns. The polystyrene spheres, purchased from Interfacial Dynamics Corporation, were surface pre-treated to be negatively charged with a surface charge density of 8.3 µC/cm$^2$ to avoid aggregation or attachment to the walls of the sample cell.

Images of the colloidal solution were collected via a digital video microscope equipped with a Mitutoyo long working-distance 50× objective lens. The output of the CCD was fed to a computer with frame grabber for image analysis. The frame grabber was externally synchronized, controlling the frame rate. This external synchronization was also configured to ensure that no frames were unintentionally dropped from the video processing. A notch filter, whose bandwidth is 10 nm at normal incidence, was placed in front of the CCD to remove the normally incident laser light. Only laser light scattered to high angles by the colloidal particles can pass the notch filter and enter the imaging system. Because the particle diameters are comparable to the wavelength of the laser light, strong diffraction results, providing a high-contrast self-illumination of the colloidal particles. As shown in Fig. 3(a), the high-angle scattered light emphasizes the edges of the poly-spheres, and yields images of the spheres with bright centers. As a comparison, shown in Fig. 3(b), another image was taken with the identical microscope objective lens using illumination from a conventional white light; here, all laser light, both straight incidence and scattered from the spheres, was eliminated by a band-reject filter which was effective at all spatial angles. It is clearly shown that the contrast between particles obtained with laser self-illumination, in Fig. 3(a), is greatly enhanced over methods utilizing conventional, external illumination. Although the spheres appeared with a more uniform brightness under the conventional illumination, the clarity of the sphere boundary under laser illumination improved the accuracy of automated particle identification and position measurement.

* As the angle increases, the position of the interference fringe shifts longitudinally. However, this changing of the position is within the focal depth of the final lens. The lens is set on another translation stage, which is used to adjust the longitudinal position of the lens when a larger dynamic angular range is needed.
For the studies described in this paper, the sample cells were orientated vertically. Without any incident laser beam, the spheres were observed to drift downward at a speed of approximately 100 microns/hour, which is negligible over the time scales of our experiments: gravitational force would only drag the polystyrene spheres down over much longer periods than the 10 second intervals described here. The interference fringes were also formed along the vertical direction. Therefore, in the horizontal direction along the surface of the sample cell, the spheres were trapped in the potential barriers, predominantly by the $\hat{x}$ component of the gradient force described in the equation (3). The particles assembled into vertical lines in the interferometric laser field, as shown in Fig. 4 (frame 1). In our initial set-up, the extent of the organization was limited by optical aberrations; however, organization in optical trenches has recently been demonstrated over macroscopic areas (over more than 1mm²).

The sequence of frames in Fig. 4 demonstrates both the balance of forces involved in our two-beam geometry, and our ability to translate ensembles along the horizontal direction. At time $T = 0.2$ s, as shown in frame 2, one of our two beams was blocked, and then unblocked at $T = 4.33$ s, as shown in frame 7. The optical trenches exist only when both beams are present, and therefore no significant gradient force existed in the horizontal dimension from frame 2 to frame 7. On the other hand, an unbalanced radiation pressure, due to the remaining beam, did exist. What is observed is that the organized lines of aligned polystyrene spheres relaxed, under Brownian motion, while they were also pushed rightward by the radiation force of the remaining light beam. The lines were promptly reorganized after the second laser beam was reapplied at $4.33$ s. Although the relaxation and re-organization times depend upon both laser power and the definition of the optical structures, we note from the frames shown below that the spheres moved from random positions into the light traps with a speed greater than 10 µms⁻¹.

* The lines appeared tilted approximately 6 degrees from the vertical; however, this is an artifact of the orientation of the CCD camera, which was slightly off the vertical direction.
3. RESULTS

The importance of forming widely spaced lines of particles before assembling them into close-packed two-dimensional structures lies in establishing organizational order while there is still a significant free volume available, which is critical to “annealing” out defects from otherwise periodic assembled structures. The fact that defects are initially present during self-organization is, statistically, unavoidable. However, effective annealing is possible so long as the disorder is not “quenched” into the system. It is the tunable nature of the lattice constant and of the particle-trench interactions (as well as the particle-particle interactions) that offers the possibility of defect-free assembly, wherein organization first proceeds under conditions providing significant free volume, and the ensemble is annealed as it proceeds, in stepwise fashion, towards close packing.

Unfortunately, in traditional templated-assembly processes, there is no free-volume left in the assembly process. For example, to fabricate a hexagonal structure, one can start with the assembly of poly-spheres into a line-like pattern whose line spacing is \( \frac{\sqrt{3}}{2} \) \( R \), where \( R \) is the sphere radius. Inevitably, some spheres might dislocate from the lattice structure. In our approach, there is dynamically configurable control over the angle between the two incident light beams, hence, control over the lattice constant; this provides a key degree of freedom in the optically-assisted assembly process. By symmetrically adjusting the incident angle of the both laser beams, the spacing of the lines can be increased to a value larger than \( 4R \), which leaves significant interstitial space, and with it a path for removal of dislocated spheres. After all defects are “annealed” out of the line-pattern, the defect-free line-like patterns can be configured back into the hexagonal structure simply by adjusting the line-spacing back to the original \( \frac{\sqrt{3}}{2} R \) value.

In this work, we introduced some defects intentionally in a low-quality line-like pattern, as shown in Fig. 5. The incident angles of both beams were then adjusted to about 5.5 degrees, which increased the spacing between the interference fringes to 5 micrometers. The fringe spacing was large enough that there was free-volume to create a path allowing any dislocated particles to drift away (due to Brownian motion and/or an optical force). Once dislocated spheres were moved out of the desired area, the spacing of the optical trenches was then adjusted back to 2.60 microns and all particles then assembled into a close-packed structure. Fig. 6 shows that the dislocated spheres in Fig. 5 were able to merge into the line-like structure while at a larger line spacing. These same lines, after the dislocated sphere disappeared, were then assembled into a close-packed structure, as shown in Fig. 7.
When the lattice constant is smaller, particle-particle interactions constrain point defects, over very long time scales, to very small motions within a many-body cage formed by the ensemble, as annealing would require the collective motion of many particles. In Fig. 6, the lattice constant has been adjusted so as to highlight the behavior of a single interstitial polystyrene sphere, indicated with an arrow, over easily accessible time scales. Since the dislocated sphere is initially located at an interference minimum, it is at an energetically unstable position. The transverse gradient force from the interference field tends to pull the sphere into the lattice (defect healing) along the direction of the arrow. However, from $T = 0\ s$ to $T = 5\ s$, the sphere was observed to fluctuate within the unstable position, constrained by the surrounding ensemble. Starting from 5 seconds, we observe in detail how this unstable equilibrium was broken: the sphere eventually merged into a stable equilibrium position (an intensity maximum) through correlated motion with two neighbors (which, in turn, required the motion of other spheres along that line). In particular, the frame for $T = 6\ s$, it is seen that the two nearest neighbors of the defect were pushed to the right slightly, away from their equilibrium positions along the intensity maximum. Under the influence of optical restoring forces, while the defect moved to the right, merging with the line, the two neighbors moved to the left, back into the center of the line after $T = 6.5\ s$. Note that the probability of time-reversed fluctuations into the defected state is reduced by lower energy of the ordered state. Note also that, in Fig. 6, the time interval among the first 7 frames was 1 second and was 0.067 second for the next 9 frames, providing a sense of both the relative stability provided by the many-body cage and the time scale over which fluctuations affect order. Further work is aimed at exploring the spatio-temporal range of correlated motion as a function of lattice parameter.
After “annealing” out all the defects, the angle \( \theta \) between the two incident beams was increased, so that the spacing between lines dropped to \( \frac{\sqrt{3}}{2} R \) (2.6 microns, in this case). This created a close-packed hexagonal pattern, shown in Fig. 7. It is observed that there are still some dislocations around the periphery of the crystal. This results from the details of the optical trenches generated in this initial set-up. At the end of the interference fringes, the \( \hat{\rho} \) component of the gradient force is larger than that close to the center of the pattern. Therefore, the force points radially inward, distorting the line-like shape of the assembled structure. However, since the overall size of the optically assisted assembly ensemble is significantly larger than that of any discontinuity at the edge of the structure, the lattice distortions along the periphery did not result in any significant effects upon the observed ensemble-averaged properties of the structures.\(^{24}\) Note again that much larger ensembles have been demonstrated in optical fringes. Although the size of the ensemble shown here is limited, the essential point is clear enough: unlike many self-assembly methods, the tunable lattice constants associated with optical assembly allows defects to be healed before the final pattern forms, so that, as shown in Fig. 7, no defects remain inside the final assembly of the close-packed hexagonal structure.

4. FURTHER WORK

We have reported an experimental study of optically assisted assembly of colloidal polystyrene spheres associated with an alternative optical trapping approach. Significantly different from the conventional laser tweezing techniques, we developed a \( \theta / 2 \theta \) adjustable trapping setup which offers simplified dynamical control of the spacing...
between the structures. Moreover, we have explored two critical advantages of optically assisted assembly: 1. Free volume can be tuned for annealing. 2. Control over lattice constants also allows for the assembly into structures other than the close-packed pattern. For example, a square lattice results when the line spacing is equal to the sphere diameter, a point that requires detailed consideration of the particle-particle interaction. A detailed study of dependence upon particle size will also be forthcoming; for some large particle sizes the equilibrium positions may be centered between intensity maxima. Moreover, there are various sorts of “optical binding” which might play a role in determining the equilibrium positions within such assemblies.

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