# **Light propagation in optical trapping assembling of colloidal particles at an interface**

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#### **ABSTRACT**

We conduct the optical trapping and assembling of polystyrene particles at the glass/solution interface by utilizing tightly focused 1064 nm laser of high power. Previously we reported that this leads to form the assembly sticking out horns consisting of single row of aligned particles through light propagation. Here, we demonstrate the laser power dependence of this phenomenon. With increasing the laser power, the particles are started to distribute around the focal spot and form the assembly larger than focal spot. The shape of the assembly becomes ellipse-like and the color at the central part of the assembly in transmission images is changed. This indicates that the assembly structure is changed, and trapping laser is started to propagate through the adjoining particles leading to horn formation. Strong laser power is necessary to elongate the horns and to align them straightly. We expect that this study will offer a novel experimental approach for assembling and crystallization of nanoparticles and molecules exclusively by optical trapping.

**Keywords:** Optical trapping, colloidal assembly, glass/solution interface, light propagation, multiple scattering

### **1. INTRODUCTION**

Upon laser focusing, particles fall into an optical potential and are trapped three-dimensionally at a focal spot<sup>1</sup>. Twodimensional assembly and crystalline structure can be formed by focusing the laser at an interface, which was first demonstrated by Burns et al<sup>2</sup>. A lot of experimental studies at the interface have been reported by utilizing various light  $\text{fields}^{2-19}$  and these assemblies are trapped only inside the irradiated area. Optical trapping and light propagation are usually studied as different phenomena, however we recently demonstrated the novel optical assembling which is enabled by the interplay of optical trapping and light propagation<sup>20</sup>. In particular we found that, when the laser propagates through the formed assembly, the assembly is expanded outside the irradiated area with sticking out rows of linearly aligned particles like horns. Here, we demonstrate the laser power dependence of this assembling phenomenon and show how the particles gather and form the assembly to stick out the horns with increasing the laser power.

# **2. SAMPLE AND OPTICAL SETUP**

Figure 1 shows the sketch of the sample chamber and optical setup. This optical setup is conventional and similar to that reported previously from our groups<sup>20-22</sup>. Polystyrene particles (Polyscience, mean diameter 500 nm) are a target for optical trapping and assembling. The particles are suspended in water and concentration are about  $3.6 \times 10^8$  particles/ml. The sample solution is sandwiched by the glass substrates (Matsunami) with a spacer (Electron Microscopy Sciences, thickness 120 μm). This sample is set on the stage of inverted microscope (Nikon Eclipse Ti). The 1064 nm continuous wave laser (Spectra-Physics, Nd<sup>3+</sup>:YVO<sub>4</sub> laser, J201-BL-106C) is introduced to objective lens (Olympus UPlanSApo, 0.95 numerical aperture,  $40 \times$  magnification) for the optical trapping. A diameter of the laser before the objective lens is about 9 mm. We use the linearly polarized light and conduct the laser power dependence in this study. Transmission images are monitored by CCD (charge-coupled device) camera with halogen lamp focused by condenser lens (0.52 numerical aperture) to the sample. The frame rate of the camera is 30 fps. 750 nm short-pass filter (Semrock: FF01- 750/SP-25) is inserted before the CCD camera to cut the 1064 nm laser from the transmission images.

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Optical Trapping and Optical Micromanipulation XIII, edited by Kishan Dholakia, Gabriel C. Spalding, Proc. of SPIE Vol. 9922, 99221R · © 2016 SPIE · CCC code: 0277-786X/16/\$18 · doi: 10.1117/12.2235272



Figure 1. An illustration of setup for optical trapping and assembling of polystyrene particles at glass/solution interface. The left-up inset shows the sketch of sample chamber sandwiched by glass substrates. The trapping laser is focused at the interface between upper-glass substrate and solution.

# **3. RESULTS AND DISCUSSION**

#### **3.1 The assembly before showing horns**



Figure 2. Transmission images observed during optical trapping at the glass/solution interface with relatively low laser power. Under this condition, the horns are not observed. (a,b,c) The laser power is 15, 50, and 100 mW, respectively. The direction of linearly polarized light is vertical. The laser is turned on at 0 s. The length of the black bars is 10 μm.

Here we demonstrate the optical trapping with increasing the laser power from 15 to 100 mW. The particles are started to distribute around the focal spot in Figure 2a. The laser power of 15 mW is not enough to stably trap the particles in the focal spot. The distribution area of the particles is determined by the balance between the optical potential and Brownian motion. When the laser power is 10 mW, it is hard to continuously trap the particles around the focal spot although the particle diffusion seems to be biased by the optical potential. With the laser power of 5 mW, it is difficult to collect the particles around the focal spot.

Figure 2b and c show the images obtained during optical trapping with laser power of 50 and 100 mW, respectively. The particles are attracted to the focal spot, and the size of the formed assembly is larger than that of focus. We consider this is because of multiple scattering in the assembly. The multiple scattering expands the effective irradiation area out from the focus with enlarging the assembly size. In addition, we can find that the assembly shape becomes to be ellipse reflecting the laser beam profile. The profile for linearly polarized light is elongated along the direction of polarization because of the tightly focusing. The assembly is started to form, while the horns are not observed yet.



#### **3.2 The assembly sticking out the horns**

Figure 3. Transmission images observed during optical trapping at the glass/solution interface with relatively high laser power. Under this condition, the horns are observed. (a,b,c) The laser power is 300, 500 mW, and 1.4 W, respectively. The direction of linearly polarized light is vertical. The laser is turned on at 0 s. The length of the black bars is 10 μm.

Here we show how the assembly develops the horns. Figure 3a indicates the images of optically trapped assembly with laser power of 300 mW. The assembly size is increased with the irradiation time and assembly is elongated along the polarization direction. After this growth process, the color at the central part of the assembly in transmission image is suddenly changed (for example from the image at 5 min 34 s to the image at 5 min 36 s in Figure 3a). As we discussed in our previous paper through transmittance spectra,<sup>20</sup> we consider that this color change corresponds to the structure change of the assembly. The particles are moving dynamically, and this structure/color change behavior is also dynamic.

After the growth of the assembly is almost completed, we can apparently observe the short and unstable horns sticking out from the assembly (for example see the images after 8 min in Figure 3a). The horn formation is not always observed and dynamic and stochastic even upon its observation. It should be noted that when we monitor the horns, the color at the assembly center is always changed. This fact allows us to say the assembly structure is important to stick out the horns through light propagation.

Figure 3b and 3c represent the optically formed assembly with horns with laser power of 500 mW and 1.4 W, respectively. The particles are assembled faster, and the color change is observed earlier with increasing the laser power. Also the horns appear rapidly, and the length of the horns becomes longer with increase in laser power. The particles constituting the horns are straightly well-aligned with laser power of 1.4 W. The horn can be extended out about 10 μm from the assembly, which should be difficult to be explained by only considering the irradiation area of the focal spot.



#### **3.3 Assembly and horn formation mechanism depending on laser power**

Figure 4. Mechanism for optical trapping evolved assembly with horns depending on laser power. (a) 15 mW to 100 mW: the particles are gathered and elongated along the linear polarization. (b) 300 mW and 500 mW: tetragonal structure is formed and it starts to develop the short horns. (c) 1.4 W: trapping laser propagates through the adjoining particles to extend the horns outside of assembly. The light red indicates the effective irradiated area expanded due to multiple scattering. The dark red line indicates the trapping light propagation along the adjoined particles in the structure. The direction of the polarization is vertical and the dashed-line ellipse denotes the size of focal spot.

We have previously proposed the possible mechanism for assembly formation with horns. In Figure 4, here we discuss about the mechanism based on the laser power dependence. The laser beam profile for linearly polarized light is elliptical because of the tightly focusing. With increasing the laser power from 15 mW up to 1.4 W, the focal spot size of laser beam slightly increases because of the Gaussian profile of laser beam (see the size of dashed-line ellipse in Figure 4a to 4c). According to the simple estimation of width of the Gaussian beam (see supporting information in previous paper<sup>20</sup>), the width is about only two times extended although the laser power is a hundred times magnified.

When the laser power is about 15 mW, the motion of the particles are started to be biased by optical potential and they are distributed around the focal spot as time proceeds. When the laser power is increased up to 100 mW, more particles are gathered to the assembly. Its size is extended with expanding the effective irradiated area out of focal spot due to multiple scattering. This area is represented by light-red in Figure 4. The elliptical assembly is formed reflecting the laser beam profile especially when the laser power is larger than around 100 mW (Figure 4a (v)).

When the laser power is 300 mW, tetragonal structure is formed by filling up the particles in elliptical shape of focal spot (Figure 4b (iv)). This structure formation can be observed through color change in transmission images and we consider that the assembly is now ready to propagate the light. After the structure formation, we can apparently observe the short horn (Figure 4b (v)). The particles are always dynamic, and it is difficult to continuously observe the color change and horn formation.

When the laser power is about 1.4 W, the assembly grows faster and the elliptical shape (Figure 4c (ii)) as well as structure/color change (Figure 4c (iii)) can be observed earlier compared with lower laser power. Then the trapping laser propagates through adjoining particles from the center to the outside of the assembly (Figure 4c (iii) to (v)). As we reported previously, the trapping light propagation was observed in the backscattering images of trapping laser along the horns. This optical trapping and light propagation are interactively evolved to stick out the horns from the assembly until the laser intensity at the horn edge becomes comparable to the intensity required for overcoming thermal fluctuations. From the results in Figure 2a, the laser power at the horn edge may correspond to about 15 mW.

## **4. CONCLUSION**

We have demonstrated the laser power dependence of optical trapping and assembling of polystyrene particles at glass/solution interface with the linearly polarized light. We have described that strong laser power is necessary to form the structure for propagating the trapping laser out from the assembly. This light propagation enables to extend the horns out from the assembly, leading to propagate the trapping laser further through horns. This assembly dynamics is due to the interactive evolution of optical trapping and light propagation.

We point out that this horn formation will be more efficiently achieved by tuning the trapping laser wavelength to electronic transition of its component nanoparticles. One of the authors (T.K.) already proposed the resonant light enhances the optical force<sup>23</sup> so that propagated resonant laser may be able to selectively trap the dye-doped particles at the edge of the horns. This resonance is also achieved when the structure of assembled particles gives a photonic band edge matching to the laser wavelength. Electric field will be confined and enhanced in the structure, leading to further extension of the assembly and its horns.

We consider that light propagation in optical trapping assembling at interfaces of glass/solution and air/solution is generally applied to molecules and their clusters. As a suggestive experiment, we have demonstrated optical trapping induced crystallization of L-phenylalanine at the air/solution interface<sup>24</sup>. One single plate-like crystal with the size of a few tens micrometer is formed, and its center is trapped at the focus. A framework structure can be found from the focus to the edge of the single crystal, which is fabricated during the crystal growth. When colloidal particles are added in the solution, we observed the particles are gathered around the edge of the framework despite the laser is not illuminated directly. This implies that the laser is efficiently propagated through the framework to form the attractive potential at the crystal edge. By switching off the laser the crystal was started to diffuse at the interface. When the laser was focused again to the crystal, the focus traces the framework and the crystal was pulled back to the original position. The framework should be thicker and receives stronger trapping force compared to other parts. Now we understand that, the trapping laser passes through the framework to grow the crystal thicker with extending the light propagation, leading to further and further crystal growth. We expect that the present work will offer a novel experimental approach for assembling and crystallization of nanoparticles molecules, proteins and DNA exclusively by optical force due to light propagation.

#### **ACKNOWLEDGEMENTS**

The present work is partly supported by MOE-ATU Project (National Chiao Tung University) of the Ministry of Education, Taiwan to H.M., and by grants from Ministry of Science and Culture, Taiwan to K.Y. (MOST 103-2113-M-009-022-MY2) and to H.M. (MOST 105-2811-M-009-022). T.K. deeply acknowledges support from the Japan Society for the Promotion of Science Postdoctoral Fellowships for Research Abroad and thanks also Prof. Hajime Ishihara for his kind understandings and strong support.

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