

**Supporting Information**  
**of**  
**Optical Force-Induced Dynamics of Assembling,**  
**Rearrangement, and Three-Dimensional Pistol-like Ejection**  
**of Microparticles at Solution Surface**

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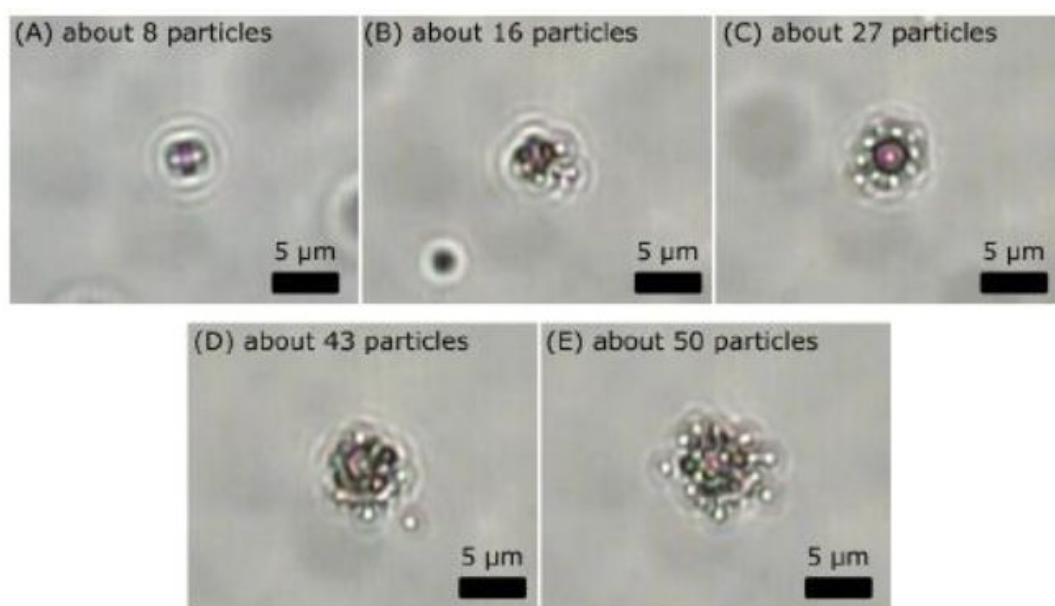
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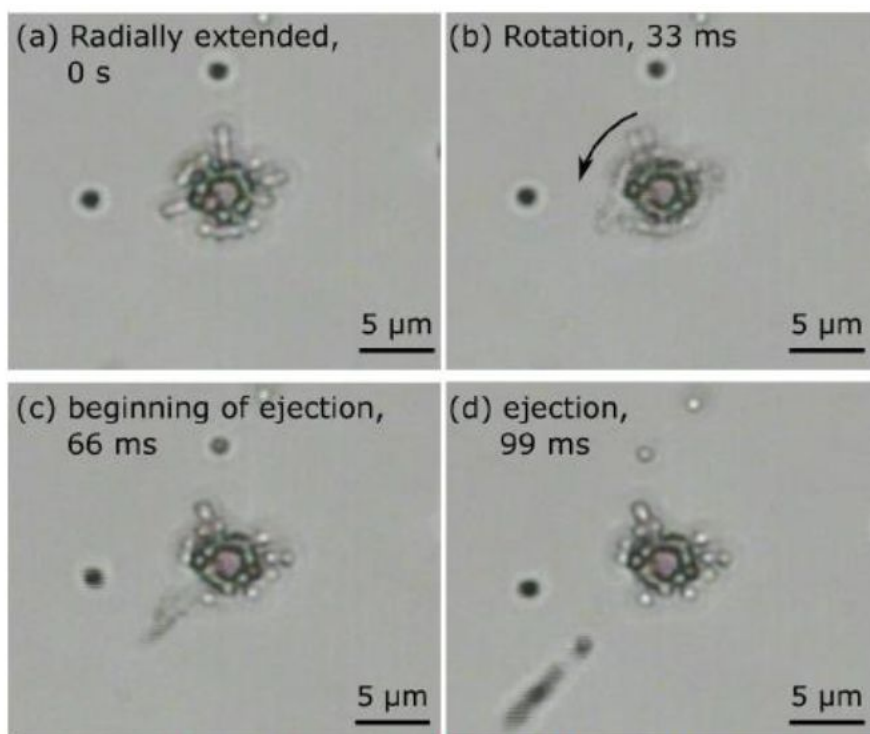
Total 7 pages including this cover page

5 Figures and Captions (Figure S1~S5)

3 Videos attached separately as Supporting Information

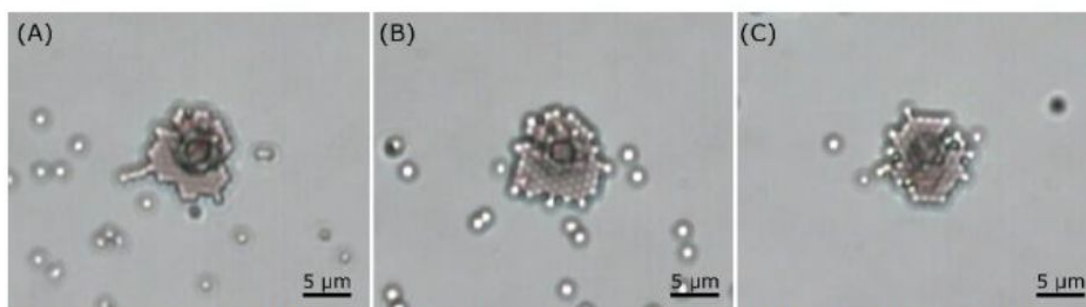


**Figure S1.** Transmission images of 1  $\mu\text{m}$  PS MPs assembly formed by optical trapping. The number of the trapped particles are counted by carefully watching the video frame by frame, and its number is indicated in each image.

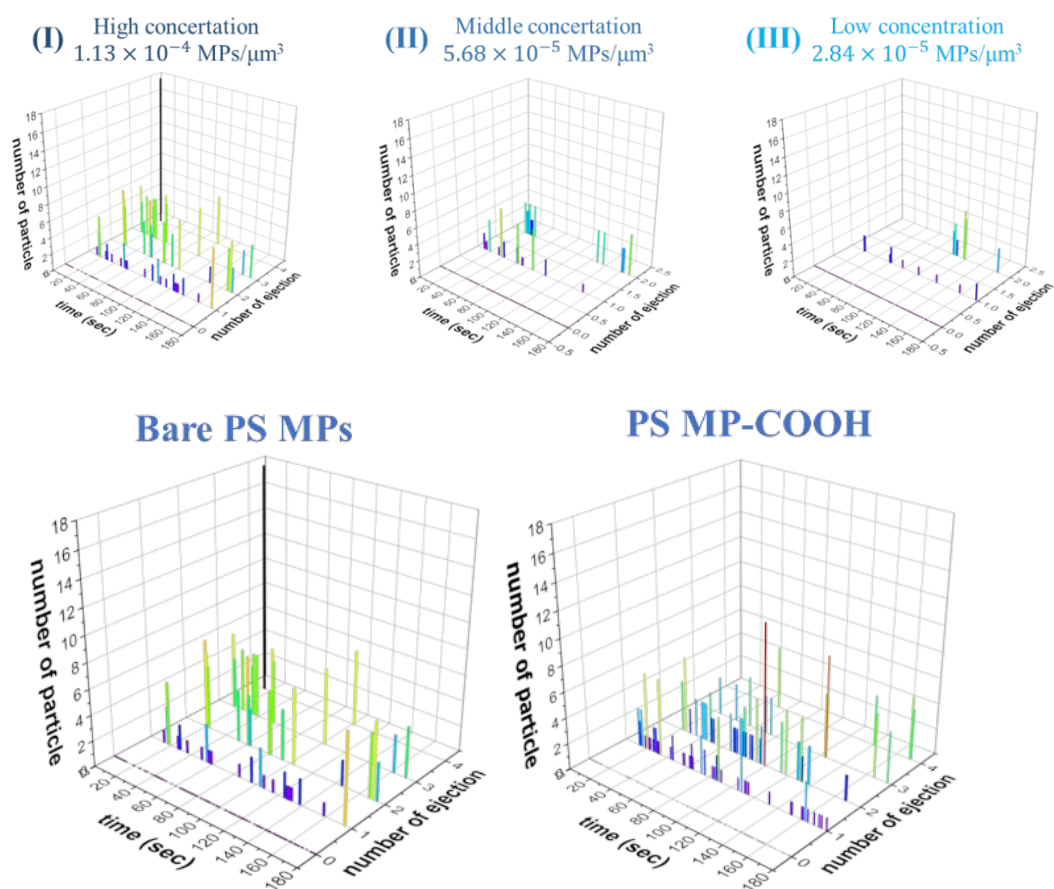


**Figure S2.**

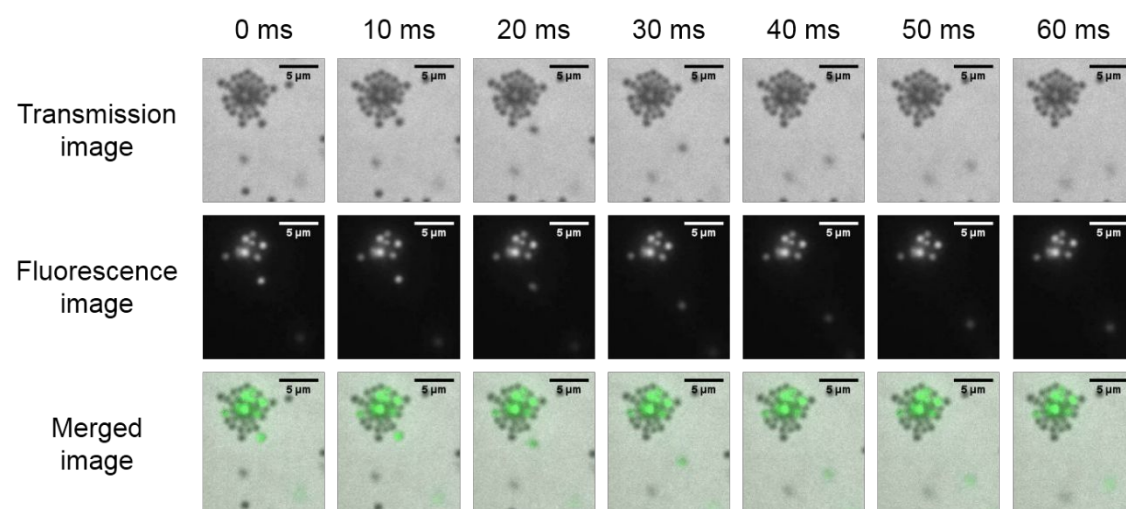
Transmission images of one typical example of the pistol-like ejection. (a) Radially extended alignment is formed outside the assembly. (b) The assembly is dynamic and the whole assembly is sometimes rotated, which direction is shown by the curved arrow. (c) Then the radially extended alignment is ejected out. (d) Four particles are ejected far away from the assembly.



**Figure S3.** Transmission images showing the hexagonal structure with horn-like alignment.



**Figure S4.** The number of the PS MPs per the ejection event is plotted as functions of irradiation time and the number of the ejection event observed in one frame, which is examined for different concentration of PS MPs. The color of the bars is same to that of Figure 4.



**Figure S5.** Transmission, fluorescence, and their merged images acquired with the overlapped configuration of Figure 1: Detection Path b). We can clearly see that the particle is ejected from the assembly.

**SI Video 1**

One ejection behavior and the rearrangement from the CC to HCP structures of the 1  $\mu\text{m}$  PS MPs at 1.4 W laser irradiation.

**SI Video 2.**

Overlaid images of transmission and backscattering of the pistol-like ejection of the 1  $\mu\text{m}$  PS MPs from their assembly observed at 1.4 W laser irradiation.

**SI Video 3.**

A movie of Figure 4 (Pistol-like ejection of the 1  $\mu\text{m}$  PS MPs from their assembly observed at 1.4 W laser irradiation).