## Photoexcitation in Optical Assembling of Dye-Doped Polystyrene Microparticle at Solution Interfaces

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Gaining control on the particle-particle interactions and self-assembled structures is a key milestone for colloidal and material sciences [1]. Currently, several strategies have been described to achieve such control, however, they lack the spatiotemporal resolution required at the microscale. In this work, we have demonstrated high potential of unexplored optical absorption force in modifying the particle-particle interactions and the structural order of light absorbing particle assemblies at the microscale.

By tightly focusing an intense non-resonant 1064 nm laser (focus size  $\sim 1 \ \mu m$ ) at D<sub>2</sub>O solution interface of the 1 µm dye-doped polystyrene (DPS) microparticles (MPs), a single concentric circle (CC)-like assembly was prepared, which diameter expanded outside of the focus to more than 10  $\mu$ m. This is similar to our previous report for bare PS MPs, which is called "optically evolved assembling" [2]. Thereafter 488 nm resonant widefield (WF) illumination was introduced to excite an electric transition of the dye in DPS MPs, exerting optical absorption force on the MPs. Under photoexcitation, the absorption force pushes the MPs towards the interface and triggers opposite morphology changes, depending on the physicochemical properties of interface; quick rearrangement from CC to hexagonal close packing (HCP) and explosive dispersion of assembly at air/solution (A/S) and glass/solution (G/S) interfaces, respectively (Figure 1). The rapid morphology changes were not observed with bare PS MPs, which supports that the photoexcitation of the dye mediates mutual attractive and repulsive interactions between MPs at A/S and G/S interface, respectively. We explain the observations by considering not only the optical absorption force but also coupling with other non-optical forces (e.g., capillary force, dipole-dipole interaction or electrostatic repulsion) at solution interfaces [3]. Due to the inherent spatiotemporal properties of light and the electronic transition of materials, the optical absorption force is a unique element to control particle-particle interaction and modify the structural order of particle assemblies at an interface.



**Figure 1. A schematic illustration of the rapid morphology changes under photoexcitation.** An optically prepared CC-like assembly at solution interfaces (middle), photoexcitation-induced quick rearrangement to HCP assembly at A/S interface (right) and explosive dispersion at G/S interface (left).

**References.** [1] J. Zhang *et al.*, *Chem. Soc. Rev.* **2017**, *46*, 5551. [2] H. Masuhara, K. Yuyama, *Annu. Rev. Phys. Chem.* **2021**, *72*, 565. [3] R. McGorty *et al.*, *Mater. Today* **2010**, *13*, 34.