# Density dependence of orientational order in one-patch particles

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Spherical patchy colloidal particles show various orientationally-ordered phases due to its well-defined interaction anisotropy when the particles are closely-packed and can only rotate [1, 2]. Such purely orientational order is unique to the system, cannot be attained with shape anisotropy, and useful as an alternative route to positional order [3] for designing self-assembled mesostructures. In the experimental system, however, short-range interparticle interaction must be repulsive to avoid irreversible aggregation by van der Waals attraction. Thus, small vibrational motion is allowed, and rotational motion of a particle cannot be completely decoupled from the translational motion. In addition, particles often possess slight shape anisotropy in experiment. Thus, the degree of packing, i.e. particle density in a system, should affect orientational order in such a system.

In this contribution, therefore, we study the density dependence of orientational order in one-patch particles, both in experiment and numerical simulation. The particles are fairly closely-packed between flat substrates, and the sample thickness is up to a few particle layers; the equilibrium orientational order at a density has been already studied [2]. With decreasing particle density, orientationally-ordered structures change, as shown e.g. in Figure 1 for tetragonal bilayer: The two-dimensionally spanned structure by inter-patch bonding in Figure 1a transforms into isolated clusters in Figure 1b. Similar structural transition was also observed in hexagonal bilayer. The corresponding simulation suggests that this transition is predominantly due to the increase in the entropic contribution of translational motion. For closer-packing at higher density than e.g. in Figure 1a, on the other hand, small shape anisotropy of the particles dominates orientational order: e.g. in hexagonal monolayer, homeotropic alignment of the particles against substrates were observed, to maximize particle density in the two-dimensional monolayer.

Our study demonstrates that orientational order in patchy particles can show drastic transition even by the small change in particle density: In a fairly dense system allowing only small vibrational translational motion, the coupling between the degrees of freedom in translational and orientational motion plays a significant role in determining the ordered phases. For tight confinement in denser system, small shape anisotropy determines the order.

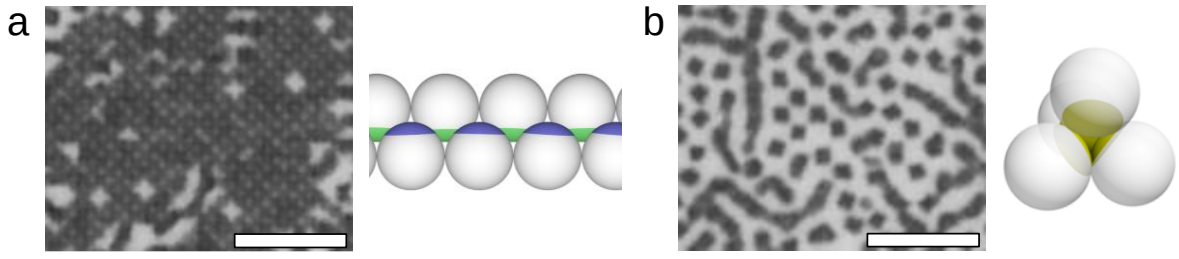


**Figure 1** Density dependence of orientationally-ordered structures in a tetragonal bilayer. (a) Bilayer structure. The schematic drawing is its side view. (b) Tetrahedral tetramers with linear clusters. The schematic drawing is the tetramer. In the microscopy images opaque patches are observed but the spherical bodies of particles are not resolved. Particle diameter is 1.5 μm and scale bars are 10 μm in the microscopy images.

[1] e.g., Z. Preisler *et al*., *Soft Matter*, **10**, 5121 (2014).
[2] Y. Iwashita & Y. Kimura, *Soft Matter*, **10**, 7170 (2014); Y. Iwashita & Y. Kimura, submitted.
[3] e.g., Q. Chen *et al*., *Nature*, **469**, 381 (2011); D.Z. Rocklin & X. Mao, *Soft Matter*, **10**, 7569 (2014).