Supplementary Materials

Polydispersity and Gelation in Concentrated Colloids with Competing Interactions

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MOLECULAR DYNAMIC SIMULATION ALGORITHM

In simulations, the starting columnar phase is obtained by replicating a colloidal column on a hexagonal lattice (Fig. S1). The column is cut out from a face-centered-cubic crystal along the orientation (001). The inter-column/lamella separation is modified such that the overall volume fraction is 0.35 same as the experimental situation.

To introduce a polydispersity into the columnar phase, we notice that the experimentally obtained columnar structures are built by crystalline clusters which were formed spontaneously even before the microcrystalline gel occurs. In the study of Auer and Frenkel [24], it is found that in a crystallizing hard sphere system, fractionation occurs during the nucleation and crystal nuclei have a narrower size distribution than in the metastable fluid. With this knowledge, it is plausible to assume that the initial columnar structures are composed of sub-crystalline domains. Particles in each sub-crystalline domain are identical in size, and the sub-crystalline domains may be distinct from each other in terms of the size of their constituent particles.

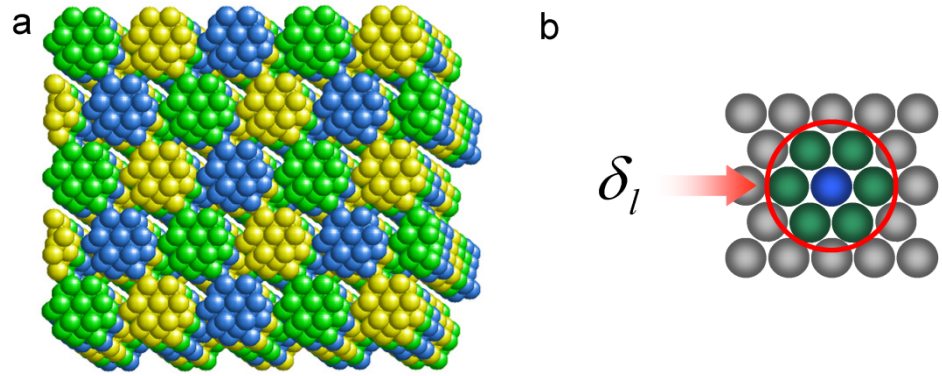

Fig. S1 a: The schematic illustration of the starting columnar phase for simulations. The number of particles in the columnar phase is 3500. Particles are colored by size. In a monodisperse system, all particles are same in size. In a polydisperse system, the columns are constructed by sub-crystalline domains. Particles in the same sub-crystalline domain are identical in size. b: Illustration of the local polydisperisty: only a central particle (blue) and its nearest neighbors (green) are included for the calculation.

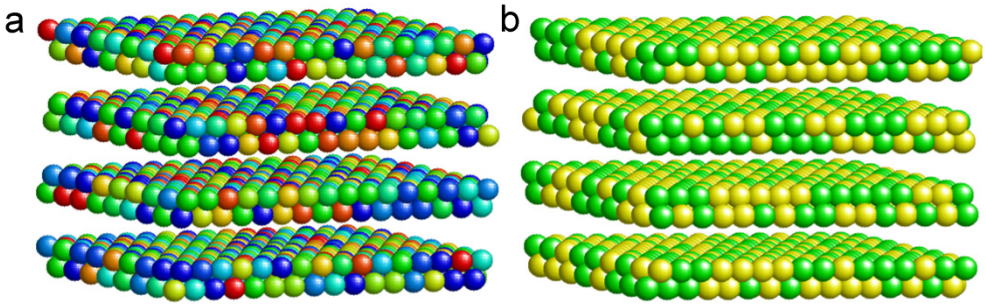

FIG. S2 Schematic illustration of starting lamellar phases. a: In a polydisperse system. B: In a binary system. Particles are colored by size. In the simulations, the number of particles in the lamellar is 4000.