

6 Supplementary Information

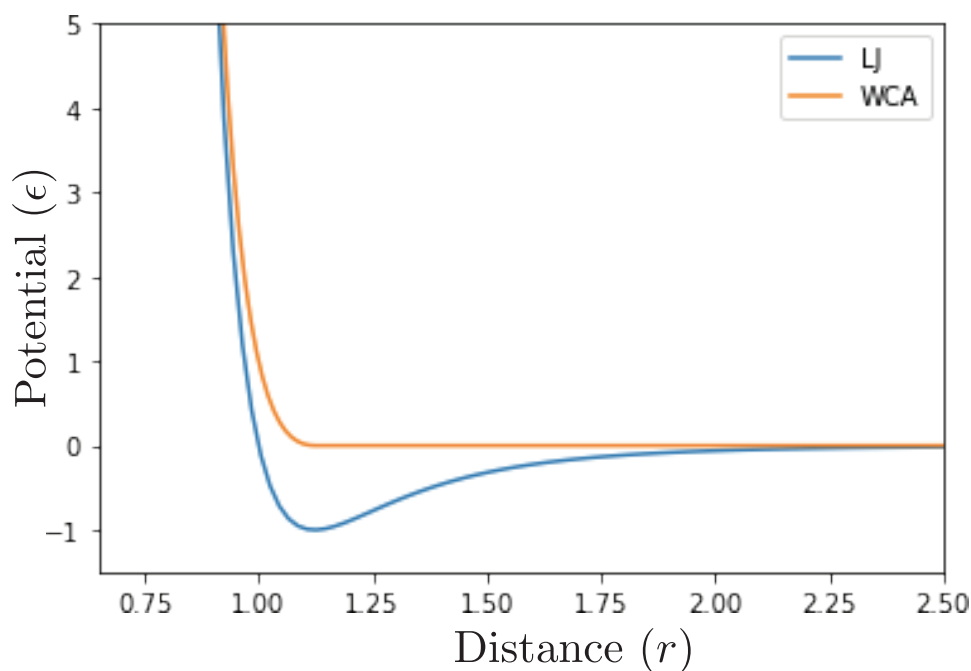


Fig. S1 The attractive Lennard-Jones Potential, and the purely repulsive Weeks–Chandler–Anderson Potential (WCA). The WCA potential corresponds to a LJ potential shifted by ϵ and with $r_{\text{cut}}^{\text{WCA}} = 2^{1/6}\sigma$.

S1.1 The Weeks–Chandler–Anderson Potential

To create the Weeks–Chandler–Anderson (WCA) potential used to model interactions,⁵⁴ we start with the Lennard-Jones (LJ) potential, as shown in Figure S1.

The Lennard-Jones pair potential is given by:

$$V_{ij}^{\text{LJ}}(r) = 4\epsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r} \right)^{12} - \left(\frac{\sigma_{ij}}{r} \right)^6 \right]. \quad (1)$$

The units of interaction strength ϵ_{ij} are $\epsilon_{ij} = \epsilon$ between all particle pairs, the units of diameter for all beads in the system are $\sigma_{ij} = \sigma$. The WCA potential corresponds to a LJ potential shifted by ϵ and with $r_{\text{cut}}^{\text{WCA}} = 2^{1/6}\sigma$:

$$U_{ij}^{\text{WCA}}(r) = \begin{cases} V_{ij}^{\text{LJ}}(r_{\text{cut}}^{\text{WCA}}) + \epsilon & r < r_{\text{cut}}^{\text{WCA}}, \\ 0 & r \geq r_{\text{cut}}^{\text{WCA}}. \end{cases} \quad (2)$$

This creates a potential wherein all interactions are purely repulsive, as shown in Fig. S1. This potential is used because it is the most similar to the purely repulsive, volume excluding interactions of the experimental hard–sphere colloids that are the subject of this computational investigation. It should be noted, however, that the interaction potential is an additional avenue for inducing an even richer diversity of phase behavior. As previous studies have shown, the range, strength, and relative positioning of the potential wells are all independent parameters that can be varied to tune self–assembly behavior.⁶

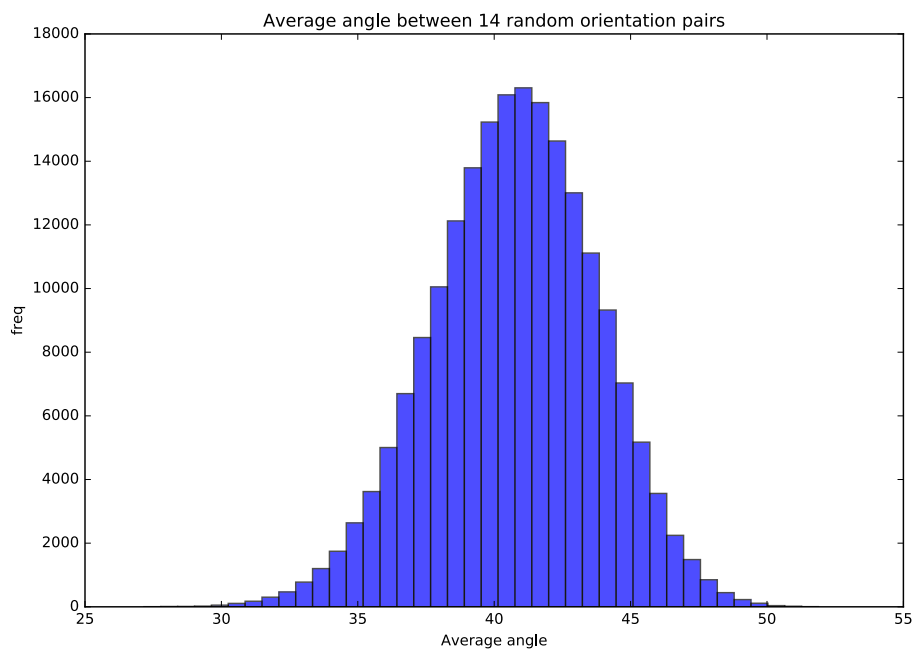


Fig. S2 Histogram of the average minimal rotation angle between 14 randomly-generated pairs of cluster orientations, taking into account the underlying octahedral symmetry of the cluster. Each average is taken over the minimal rotation angle between a randomly-generated orientation and 14 randomly-generated partner orientations. This averaging procedure was performed 200,000 times to generate the above histogram.