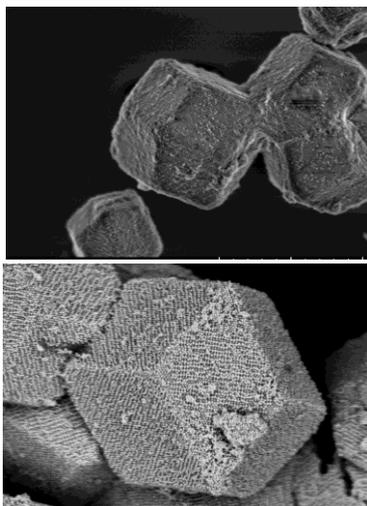


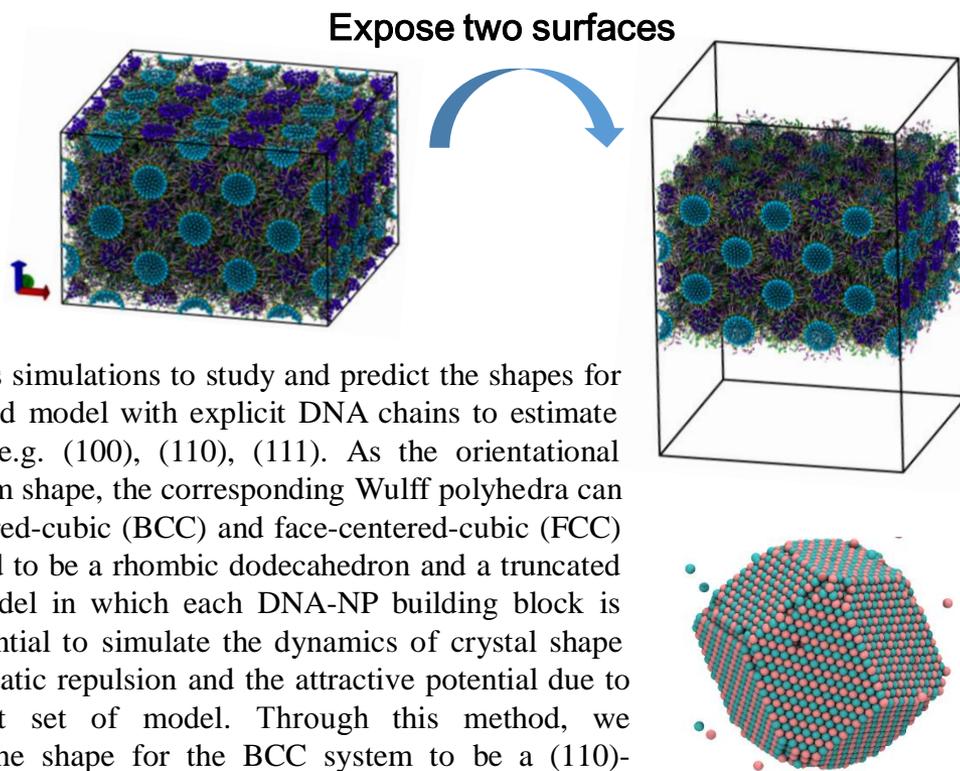
DNA-mediated nanoparticle crystallization into Wulff polyhedra

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DNA hybridization has proven promising to mediate versatile crystallization product of nanoparticles (NPs). Here, we showed that through very slow cooling, DNA functionalized nanoparticles can assemble into superlattices with a specific crystal habit, providing a nanoscale analogue to the crystallization behavior exhibited by conventional atomic crystals.



Here we apply multi-scale models with molecular dynamics simulations to study and predict the shapes for such systems. We firstly use a scale-accurate coarse-grained model with explicit DNA chains to estimate surface energy ratios for different surface orientations, e.g. (100), (110), (111). As the orientational dependence of the surface energy determines the equilibrium shape, the corresponding Wulff polyhedra can be calculated accordingly. For the symmetry of body-centered-cubic (BCC) and face-centered-cubic (FCC) of DNA-NP superlattices, the Wulff polyhedra is predicted to be a rhombic dodecahedron and a truncated octahedron respectively. Secondly, we use a colloidal model in which each DNA-NP building block is represented by a single bead with effective pair-wise potential to simulate the dynamics of crystal shape formation. The repulsive potential comes from the electrostatic repulsion and the attractive potential due to complementary DNA strands is validated by the first set of model. Through this method, we reproduce the polyhedra growth in silico, and confirm the shape for the BCC system to be a (110)-enclosed rhombic dodecahedron. However, due to defects including twinning and stacking faults in the lattice, the FCC system does not show any uniform shape except triangular features with (111) and (100) facets. The simulated crystal habits of both BCC and FCC system are consistent with experiments.