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Engineering Self-limiting Assembly via Programmable Misfit of DNA Origami Particles

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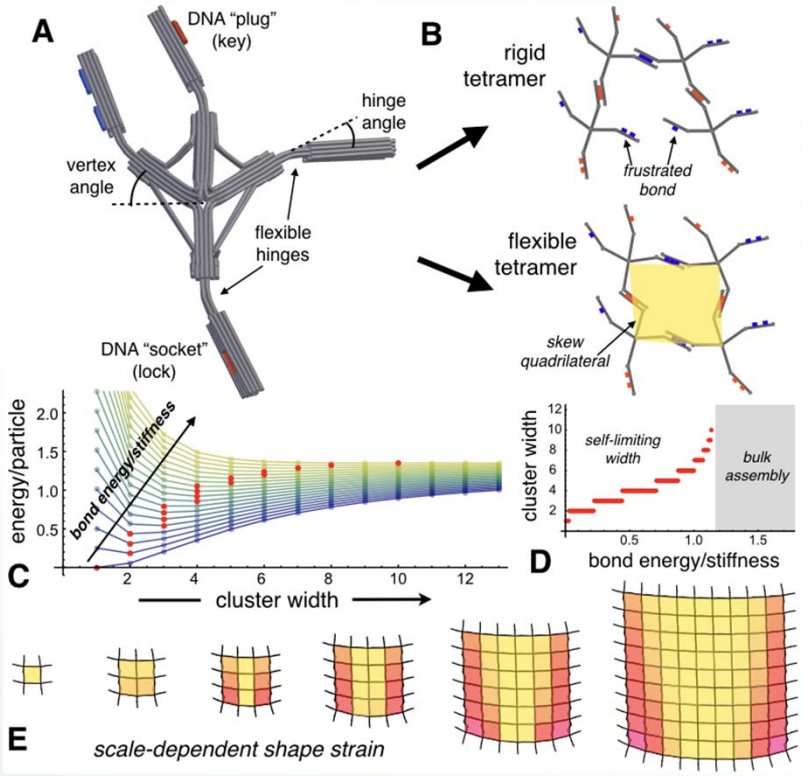
The assembly of identical sub-units into larger, yet finite-sized, superstructures is ubiquitous and functionally vital in biology, from photonic nanostructures in butterfly wings to size-regulated protein fibers. An emerging paradigm to achieve finite architectures in synthetic systems is the incorporation of *geometric frustration* into the assembly process. Like an ill-fitting jigsaw puzzle, such assemblies require mechanical deformation of building blocks, which in turn, feeds back into self-limiting equilibrium domains.

To realize particles with the control over inter-particle misfit *and* shape deformability needed for size-selective assembly, a new class of 'frustrated vertex' particles was designed for synthesis via state-of-the-art DNA origami techniques. Angular geometry of vertices and pre-bent hinges frustrate the 'lock and key' bonds between particle arms, leading to skewed local packing incompatible uniform planar tiling. Computational modeling verifies the accumulation of hinge bending with cluster size, which gives rise to equilibrium domains whose finite width is tunable via strength of DNA-mediated bonds.



(A) Finite-element simulation of DNA origami vertex particle, cylinders depicting base-paired double strands; (B) Angular geometry frustrates bonds and requires cluster deformation; Size-dependent energy/particle (C) predicts self-limiting domains (D) due to the accumulation of shape strain (E)

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