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Entropically patchy particles: Shape-driven self assembly of hard colloids[1]

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Although the structural diversity of colloidal fluids and crystals has grown substantially in recent years, it still aspires to that of atomic and molecular systems. Ionic colloidal crystals and binary nanoparticle superlattices, by exploiting electrostatic interactions in mixtures of particles of opposite charge, have substantially broadened the diversity of structures beyond those obtainable in traditional hard sphere systems, but rely on energetic interactions as well as entropy for their stability. Likewise, "traditional" patchy particles with sticky interactions exploit explicit attractive interactions for assembly. Here we explore the role of shape and entropy in phase transitions of hard particle fluids, in the absence of all other interactions. Using computer simulations, we show that particle shape alone can suffice to produce a rich diversity of colloidal crystals, quasicrystals, glasses and mesophases through thermodynamic self-assembly whose complexity rivals that of atomic analogues. We compare self-assembled phases of hard polyhedra with their dense packings obtained from small unit cell compressions, and show the packings tend to be less structurally complex than the assemblies. Based on our findings, we present design rules for anisotropic hard, facetted colloids as "entropically patchy particles" for self assembly.

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