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**Polymer-tethered nanoparticle “shape amphiphiles”: A new class of macromolecular building block for self-assembly**

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Fabricating complex ordered structures from nanoparticles requires controlling nanoparticle interactions for self-assembly over multiple length scales. Here we exploit both building block shape and interaction anisotropy for self-assembly, and explore the use of polymer “tails” attached to nanoparticle “head groups” to create a new kind of amphiphile that self assembles into structures like those seen in surfactant and block copolymer systems, but with important differences arising from nanoparticle shape, and tethered nanoparticle geometry and topology. We investigate the impact of nanoparticle size polydispersity and show that it can both help and hinder formation of certain complex phases. Using simulation, we investigate tethered spheres, rods, cubes, triangles, and other shapes, and provide design rules for the predicted self-assembly of a range of chiral and achiral structures, including helical scrolls, gyroid, square arrays, and ionic crystal-like structures.