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Shape Alloys of Nanorods and Nanospheres from Self-Assembly

JAIME MILLAN, University of Michigan, XINGCHEN YE, University of Pennsylvania, MICHAEL ENGEL, University of Michigan, JUN CHEN, BENJAMIN DIROLL, University of Pennsylvania, SHARON GLOTZER, University of Michigan, CHRIS MURRAY, University of Pennsylvania — Mixtures of anisotropic nanocrystals promise a great diversity of superlattices and phase behaviors beyond those of single-component systems. However, obtaining a colloidal shape alloy in which two different shapes are thermodynamically co-assembled into a crystalline superlattice has remained a challenge. Here we present a joint experimental-computational investigation of two geometrically ubiquitous nanocrystalline building blocks—nanorods and nanospheres—that overcome their natural entropic tendency towards macroscopic phase separation and co-assemble into three intriguing phases over centimeter scales, including an AB2-type binary superlattice. Monte Carlo simulations reveal that although this shape alloy is entropically stable at high packing fraction, demixing is favored at experimental densities. Simulations with short-ranged attractive interactions demonstrate that the alloy is stabilized by interactions induced by ligand stabilizers and/or depletion effects. An asymmetry in the relative interaction strength between rods and spheres improves the robustness of the self-assembly process. Reference: Ye, Millan, Engel, Chen, Diroll, Glotzer, Murray, Nano Letters 13, 4980 (2013).

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