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**A Directional Entropic Force Approach to Assemble Anisotropic Nanoparticles into Superlattices**[1] BYEONGDU LEE, Argonne Natl Lab, TAO LI TEAM, KAYLIE YOUNG, GEORGE C. SCHATZ, CHAD A. MIRKIN COLLABORATION[2], MICHAEL ENGEL, PABLO F. DAMASCENO, SHARON C. GLOTZER COLLABORATION[3] — We introduce a directional entropic force approach (DEFA) for controlling the assembly of anisotropic nanoparticles into crystalline lattices. The method relies on surfactant micelle-induced depletion interactions to assemble anisotropic gold nanoparticles into reconfigurable, non-close-packed (open) superlattices in solution. The anisotropic nanoparticles align along their flat facets to maximize entropy, and therefore minimize the free energy of the system, leading to assemblies with long-range order. Importantly, our experimental work complements recent theoretical work that proposes directional entropic forces between nanoparticle facets as a viable means for thermodynamically assembling nanoparticle superlattices. The experimental work herein uses depletants to create strong attractive forces that can drive assembly of reversible superlattices with tunable spacing in solution. These directional entropic forces are analogous to the directional bonding between atoms in molecules. The resulting crystalline superlattices are therefore shape-dependent. We show that the electrostatic and depletion interactions combine to determine the lattice spacing, and can be tuned independently with surfactant concentration and ionic strength to reconfigure the lattice constant. .

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