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Stoichiometric Control of DNA-Grafted Colloid Self-Assembly
THI VO, VENKAT VENKATASUBRAMANIAN, SANAT KUMAR, Columbia University, BABJI SRINIVASAN, Indian Institute of Technology - Gandhinagar, SUCHETAN PAL, YUGANG ZHANG, OLEG GANG, Brookhaven National Lab
— There have been recent surges of interest in understanding the self-assembly of DNA-grafted colloids into different crystallographic lattices, namely CsCl, AlB_2, Cr_3Si, and Cs_6C_{60}. Conventional approaches view the number of grafted linkers and effective size of each colloid as the major governing design parameters. It is generally assumed that the mixed stoichiometries need to match those defined by the target structures in order to obtain the desired lattice. Thus, contributions from stoichiometry are considered secondary and its exact effects on lattice formation remains an open question. Theoretical extensions to the popular complementary contact model show that the equilibrium lattice structure can be tuned through direct control of stoichiometry. Our results are also validated through experimental observations of the equilibrium crystal morphologies at differing stoichiometric ratios. These findings strongly suggest that stoichiometry is a new handle that can be used to control DNA-grafted colloidal self-assembly.

Thi Vo
Columbia Univ

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