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Making DNA competitive: a new strategy to improve the self-assembly properties of DNA-coated particles MIRJAM LEUNISSEN[1], FOM Institute AMOLF, BORTOLO MOGNETTI[2], DAAN FRENKEL[3], University of Cambridge — We present a new approach to widen the normally very narrow temperature window for equilibrium self-assembly (e.g. crystallization) of DNA-coated particles. Using Monte Carlo simulations, we first show that not only enthalpic but also entropic effects - due to the multi-bond character of the DNA-mediated interactions - play an important role in the overall binding properties of the particles. We then outline a new strategy that exploits the competition between different types of inter-particle DNA linkages to achieve a temperature-dependent switching of the dominant bond type. Depending on the length ratio of the DNA constructs, the bond switching is either energetically driven or controlled by a combinatorial entropy gain, which arises from the large number of possible binding partners for each DNA strand. Importantly, the resulting particle interaction is less strongly temperature dependent than in "conventional" systems with only one bond type, thus enhancing the experimental control over self-assembly. Finally, we will also show that in general stable gas-liquid separation is expected to occur only for particles smaller than a few tens of nanometers, which suggests that nanoparticles and micrometer-sized colloids will follow different routes to crystallization.

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