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Unified Theoretical Framework for Shape Entropy in Colloids[1]
GREG VAN ANDERS, N. KHALID AHMED, DAPHNE KLOTSA, MICHAEL ENGEL, SHARON C. GLOTZER, Department of Chemical Engineering, University of Michigan — Entropy has long been known to order colloidal systems ranging from dense suspensions of hard particles to dilute suspensions of spheres in the presence of smaller polymeric depletants. We present a framework for treating directional entropic forces (DEFs) in systems of colloidal shapes. By introducing an effective potential of mean force and torque we demonstrate that the microscopic origin of the entropic ordering of anisotropic shapes is the emergence of DEFs that tend to align neighboring particles. We define and compute these forces and show that, at the onset of ordering, they are on par with traditional depletion interactions, as well as and other forces contributing to assembly in nanocolloidal systems. By retaining only steric interactions, we allow the comparison of the role of shape to other forces present in experimental systems. Well-known cases involving spheres arise as the limit of "zero shape." Our results apply to monodisperse systems and mixtures of hard particles of arbitrary shape and to systems of hard particles with traditional depletants. As such, we present a single theoretical framework that unifies the ordering of arbitrary shapes due to entropy alone, incorporating the well-known works of Kirkwood, Onsager, and Asakura and Oosawa.

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