From Many-Body Interactions to Collective Behaviour in Core-Shell Microgels

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Core-shell microgels, composed of a solid core covalently bonded to a crosslinked polymer shell, are versatile stimuli-responsive colloids with promising applications in soft matter and materials science.[1] Yet, a microscopic, temperature-resolved understanding of their effective interactions–and of how these govern collective behaviour–remains incomplete. The challenge becomes particularly acute in dense suspensions, where resolving all microscopic degrees of freedom proves computationally prohibitive and coarse-grained effective interaction potentials become essential. For soft, deformable particles such as microgels, however, these interactions are inherently environment-dependent: crowding, compression, shrinking, and interpenetration can all induce significant deviations from a simple pairwise picture. In this talk, we present a numerical framework to generate realistic core-shell microgels with tunable crosslinker concentration and shell-to-core ratio, reproducing the structural features of experimental silica-pNIPAM systems across the volume phase transition (VPT). We then determine their effective interactions as a function of temperature, uncovering a clear many-body signature: three-body contributions, while smaller than pair interactions, are not negligible and change qualitatively across the VPT, being attractive at low temperature, repulsive at high temperature, and nearly absent near the transition.[2] Guided by these microscopic insights, we develop coarse-grained descriptions for dense suspensions in which effective pairwise approximations become inadequate. Using a machine-learning framework trained on fine-grained simulation data, we construct computationally efficient many-body effective potentials that encode the environmental dependence of the underlying interactions and remain predictive at high packing fractions.[3] These models accurately reproduce the structural and thermodynamic properties of dense core-shell microgel assemblies, providing a route toward predictive and architecture-aware modelling of collective behaviour in responsive soft-matter systems.

Keywords:
Microgels, Molecular Simulation, Machine Learning

References:

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