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Anisotropy, multivalency and flexibility-induced effects in colloidal systems

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Summary

IN this thesis, we have studied the impact of particle shape anisotropy, multivalent interactions and flexibility on systems of colloidal particles. The study of these effects was motivated first by a desire to understand the rich underlying physics that governs these systems and second, because of their possible relevance to understanding processes found in biology. For example, to shed light on how shape changes affect the biological function^{100–104} of biopolymers²⁵¹ and proteins.^{252,282} Additionally, colloidal particles can be employed for medical applications, such as for targeted drug-delivery.^{7–9,114–117} In addition to biological and medical examples, there exists a great variety of applications where colloidal particles can serve as model systems.^{17–19,127–129,195}

A general introduction to several aspects of soft matter and specifically, colloid science that are relevant to this thesis are given in **Chapter 1**.

In **Chapter 2**, we report on our discovery that colloidal dimer particles have preferred orientations with respect to the substrate above which they diffuse, due to electrostatic repulsion between substrate and particle, which is greatly affected by the anisotropic shape of the dimer particles. Our results highlight the rich dynamics that nonspherical particles exhibit in the proximity of walls and can aid in developing quantitative frameworks for the dynamics of arbitrarily-shaped particles in confinement.

On top of the effects of a fixed and anisotropic particle shape, we have studied colloidal systems that have internal degrees of freedom that allow for shape changes. We have extensively characterized the properties of colloid-supported lipid bilayers^{85,110–112} (CSLBs), that consist of solid micron-sized colloidal particles surrounded by a fluid lipid bilayer. In **Chapter 3**, we have first characterized how the formation of CSLBs can be optimized, leading to CSLBs that have a high colloidal stability and a homogeneous bilayer that is fully fluid. Then, in **Chapter 4**, we have studied the multivalent interactions between DNA linker-functionalized CSLBs that can form flexible bonds. We have found that linker depletion effects^{84,126} can be used to limit the valency of self-assembled clusters made of CSLBs.

By taking advantage of the ability to limit the valency of clusters of CSLBs using low DNA linker concentrations, we have formed flexibly-linked colloidal chains of three to six CSLBs, as discussed in **Chapters 5–6**. These can serve as model system for e.g. biopolymers²⁵¹ and intrinsically disordered proteins,^{252,282} in which shape changes affect the biological function of the compound.^{100–104} We have compared

experimental and simulated data to predictions based on polymer theory. Moreover, we have quantitatively measured the full diffusion tensor of these flexible chains and have found marked flexibility-induced effects, such as a Brownian quasiscallop mode, where translational displacements are coupled to shape changes and a second timescale set by the flexibility of the chain, which leads to a faster relaxation of the influence of particle shape on diffusion.

In **Chapter 7** we have assembled flexible rings of four and six CSLBs, that mimic ring polymers,³⁰² which are of great interest due to their unique topology^{302,306} and their diffusive^{306,307} and rheological behavior.^{303,305} We have quantified the differences between flexible chains, loops and domino lattices, where we found a lower flexibility and a higher diffusivity of the loops and lattices compared to the chains. We argued that the flexibility of the loops and lattices scales as the number of excess floppy modes per bond. This scaling could have implications for the flexibility of floppy colloidal materials.

Finally, we have provided a short outlook in **Chapter 8** on how to synthesize flexibly-linked particles with directional interactions. Because of their unique combination of directional interactions with flexible bonds, we hope that these type of particles will lead to a higher degree of control over the self-assembly of reconfigurable colloidal structures in future studies.

In conclusion, our work demonstrates the rich dynamics and possibilities for applications of reconfigurable colloidal systems. Using experiments and simulations, we have found marked flexibility-induced effects in the behavior of reconfigurable colloidal structures. We hope our findings further the study of the diffusivity of flexible objects found in complex mixtures relevant in, for example, the cosmetic, pharmaceutical and food industries, as well as in biological systems. Our results may have implications for understanding both the diffusive behavior and the most likely conformations of macromolecular systems in biology and industry, such as polymers, single-stranded DNA and other chain-like molecules.