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## Self-assembly of flexible and rigid structures: from colloidal molecules to lattices

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# Summary

Self-assembly offers a promising route to create complex structures and materials using simple building blocks. Through, colloidal self-assembly, we can understand the governing principles of the self-assembly process and unlocking its potential in diverse applications in materials science, photonics and electronics. This summary provides an overview of the key findings and insights presented in Chapters 2, 3, and 4. These chapters delve into the self-assembly of colloidal molecules driven by electrostatic forces, the controlled conformational flexibility of colloidal molecules, and the self-assembly of flexible colloidal lattices, respectively.

In chapter 2, we explored the self-assembly of colloidal molecules driven by electrostatic forces. We discovered that the shape and size ratio of colloidal particles play a crucial role in controlling their valence. By using positively charged cubes and negatively charged spheres, we achieved high yields of different types of colloidal molecules with coordination numbers of  $AB_6$ ,  $AB_4$ , and  $AB_2$ . Monte Carlo simulations helped us determine the appropriate size ratios for obtaining each molecule type, and we found that electrostatic repulsion between the spheres was important for leveraging the templating effect of the cubes. We also observed that the spheres preferentially absorbed onto the faces of the cubes, preventing the formation of irregular clusters shapes. Monte Carlo simulations aided in determining the size ratios required for each molecule type, and the importance of electrostatic repulsion between spheres was identified. This approach offers several advantages, such as valence control through geometric factors, versatility in material choice, and magnetic separation of excess particles. The method's simplicity and robustness make it applicable to different template geometries and driving mechanisms, opening avenues for creating novel materials and model systems.

In Chapter 3, we demonstrated the self-assembly of colloidal molecules with directional bonds and controlled conformational flexibility using spherical and cubic particles. By changing of the size ratio of spheres to cubes, we achieved high yields of flexible colloidal molecules with well-defined coordination numbers. The critical factor for restricting sphere motion to the cube's face was identified as the size ratio ( $\alpha$ ) and the DNA patch size between the sphere and cube. As  $\alpha$  increased, sphere motion became confined to a single face of the cube. A microscopical model revealed the creation of an effective free-energy landscape due to the curvature variation of the cube, limiting the probability of sphere crossing at edges and corners. Furthermore, we demonstrated the reversible switching of sphere motion on the cube's surface using temperature. These flexible colloidal molecules act as basic units for higher-order structures and offer valuable insights

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into the design of materials with controlled conformational flexibility. Additionally, our findings unlock possibilities for the development of controllable functional devices and machines.

In chapter 4, we explored the self-assembly of binary mixtures of colloidal particles functionalized with surface-mobile DNA into flexible square colloidal lattices. When using equal-sized spheres, the lattice transitioned between square and hexagonal configurations. However, unequal-sized spheres and spheres-cubes systems maintained a square arrangement with flexibility. The combination of spheres and cubes resulted in a square lattice with limited angular motion due to confined sphere movement. Cluster growth analysis showed that the system with equal-sized spheres exhibited the fastest growth, while the system with spheres and cubes took the longest to form square lattices. The findings emphasize the influence of particle shape, size, and reconfiguration on growth dynamics and lattice arrangement, providing insights for designing flexible colloidal materials.

Overall, the chapters presented in this thesis contribute to our understanding of self-assembly processes in binary colloidal systems. It also sheds light on how the shape, size, and number ratio of colloidal particles impact the final structure of colloidal molecules in both electrostatic and DNA functionalized colloidal assembly, and as well as flexible colloidal lattices. This work showcases the potential for creating novel flexible materials with tailored properties. The research findings also provide fundamental insights into the governing mechanism of self-assembly and route to the development of functional materials and devices with controlled properties and behavior.