

Chapter 2

Motivation and Outline

To continue along the lines of thoughts of George Whitesides, the primary focus of this thesis is on fabrication or, to be more precise, the generation of symmetric surface patterns via colloidal crystallization. As such, the self-assembly process of colloidal particles to form highly ordered, two-dimensional crystals is at the heart of the projects introduced in this thesis.

As briefly touched on in the introduction, colloidal self-assembly offers several attractive benefits for the creation of surface patterns. First, colloids mostly consist of cheap materials and can be synthesized with enormous precision with respect to their size and size distribution. Second, they are able to assemble into two-dimensional arrays with an astonishingly high overall order that can subsequently be transferred to nanostructure arrays when the monolayer is used as a mask in a lithographic process. As the colloids can be synthesized in a broad size range down to several tens of nanometers, the diffraction limit is conveniently circumvented and structure arrays with feature sizes as small as several nanometers can be created. This lithographic process does not require any sophisticated equipment. Compared to electron beam lithography or a focused ion beam, both of which are able to create arbitrary structures with similar resolution, colloidal lithography is a parallel process in which all structures are created simultaneously during the lithographic process. In contrast, both other mentioned techniques are serial processes that literally “write” one structure after another. Hence, colloidal lithography is both cheaper and faster—and in principal amendable to arbitrary large surfaces.

One of the main drawbacks of the colloidal lithography process is its inherent restriction to limited structural designs that are pre-set by the arrangements of the colloids in the monolayer. One major research direction is therefore the extension of accessible structures that can be created using colloids as masks. Originally, triangular structures that reflect the shape of the interstitial sites of a densely-packed monolayer were at the forefront of colloidal lithography and an immense amount of work has been designated to investigate and exploit their plasmonic properties; especially in sensing applications.

Only in the last few years has the field widened to target more complex organizations of colloids. First, the controlled creation of non-close-packed monolayers was a milestone for applications in lithography as the area shaded by the colloids could be used in lithographic processes by introduction of rotations and angular evaporation and etching steps. Thus, a variety of different surface structures are now available in symmetric arrays by colloidal lithography. On the other hand, several approaches have been introduced for the assembly of binary crystals consisting of a close-packed monolayer of large spheres with small spheres present in the interstitial sites formed by the large spheres. As both number and size of the small spheres can be adjusted separately, binary monolayers possess a rich variety of possible conformations and crystal structures compared to single particle monolayers.

The motivation of this thesis is to further contribute to flexibility and versatility of colloidal lithography. Several methods for the creation of colloidal crystals with different complexities are presented and subsequently used in lithographic approaches to introduce novel structural features or functions to nanostructure arrays. Even though several applications for the structured arrays prepared in this thesis can be envisioned and are briefly touched on in the individual chapters, the aim of the work is not the detailed exploitation of the structures in sophisticated applications. Instead, structures and fabrication processes are presented and made available to a broad range of scientists by the simplicity of the colloidal crystallization approach.

The thesis is organized as follows. First, a short survey about the theoretical foundations of processes relevant to this thesis is presented ([Chap. 3](#)). These involve the synthesis of colloids, forces involved in their stabilization and crystallization and experimental methods to yield two-dimensional crystals. Furthermore, a comparison of colloidal lithography to other nanopatterning methods is given along with a short introduction on the interaction of metal nanostructures with light that lead to a resonance phenomenon called plasmon resonances. Following the theoretical section will be a short introduction to methods that have been used in this thesis ([Chap. 4](#)).

The results and discussion chapter is generally divided into three parts: synthesis of functional colloidal particles, monolayer crystallization processes and different lithographic approaches. The three main parts are further divided into subchapters that present the individual projects that are subject of this thesis. As several projects require synthesis as well as crystallization and lithography, the different chapters are generally interconnected. The unifying motive for all projects is the use of colloidal particles to generate symmetric structure arrays.

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