

Preface

The focus of this thesis lies in the use of soft-matter self-assembly as a design tool for the bottom-up manufacture of functional inorganic nanoarchitectures. Several routes are explored, starting from structure formation of hybrid materials on the 10-nm length scale to their application in photonic and optoelectronic devices. Considering the underlying principles and accomplished results, the outline of the thesis is structured as follows:

Chapters 1–4:

Introduction to fundamental aspects of experimental and techniques.

Chapters 5–7:

Material morphology control and implementations in thin-film architectures.

Chapters 8–10:

Photonic structures designed by soft-matter self-assembly.

The fundamental principles of soft-matter self-assembly are discussed in [Chap. 1](#), including the driving mechanisms for structure formation in block copolymers (BCP), colloidal and hybrid systems, as well as their implementation in soft nanotechnology. The optics of thin films and interfaces is the subject of [Chap. 2](#), an important aspect of many results presented in the following chapters. Dye-sensitised solar cells were a main device platform for the implementation and validation of the proposed material functionality. [Chapter 3](#) is therefore devoted to explaining the main principles of this photovoltaic concept and describing current challenges for further efficiency improvements. The final chapter of the introductory part, [Chap. 4](#), then represents an overview of the material fabrication and characterisation techniques that were employed in the course of this work.

Experimental results of the doctoral research are presented in [Chaps. 5–10](#). In [Chap. 5](#) the developed routes for BCP-induced structure control of inorganic nanomaterials are described. These findings were of central importance for many applications presented in the following. The morphological control that is offered by soft-matter co-assembly allows to finely control crystallisation and electronic properties of mesoporous networks. This is explored in [Chap. 6](#), which contains the study of crystal growth in BCP-derived TiO₂ nanostructures during high temperature calcination and its effect on the performance of dye-sensitised solar cells. The sacrificial use of organic material suffers from considerable shrinkage,

occurring during the several fabrication steps, from solution processing of the hybrid mixture to the crystallisation of the inorganic material at high temperatures. In [Chap. 7](#), a protocol is developed that enables to overcome this drawback and allows the formation of sufficiently thick films without the typically occurring crack formation and delamination.

Soft-matter self-assembly allows fine tuning of inorganic material formation on the 10–500 nm length scale and therefore enables to design material architectures with interesting photonic properties. This is the motivation for [Chaps. 7–10](#). Control over sub-wavelength properties, such as pore dimensions, pore volume and film thickness of the resulting inorganic films enabled the stacking of individual mesoporous layers into a multilayer lattice of alternating high and low refractive index, a mesoporous distributed Bragg reflector (MDBR). A BCP-based route to tunable MDBRs is presented in [Chap. 8](#). The combination of BCP and colloidal self-assembly was used to establish a TiO_2 electrode architecture, where a high surface area mesoporous underlayer is coupled to an optically and electronically active three-dimensional photonic crystal. This concept is demonstrated in [Chap. 9](#), including the experimental characterisation of such a photonic electrode architecture in terms of light harvesting and device performance in a dye-sensitised solar cell. Detailed control over the pore architecture allowed the fabrication of dielectric thin films with such ultralow refractive indices that enable the incorporation of high refractive index photocatalytic TiO_2 nanocrystals. A new materials strategy for self-cleaning antireflective coatings is presented in [Chap. 10](#), which is compatible with low processing temperatures and therefore applicable for a variety of substrate materials.

Finally, [Chap. 11](#) concludes the work by summarising the results and identifying possible directions for future studies.

Inorganic Nanoarchitectures by Organic Self-Assembly

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