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Properties of organic-inorganic hybrids

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Summary

In this thesis, I investigated *Properties of Organic-Inorganic Hybrids* in terms of *Chemistry, Connectivity and Confinement*. Organic-inorganic hybrids are materials that consist of organic and inorganic components that are combined into a single compound. Organic and inorganic compounds generally have different physical properties. While organic compounds can provide structural flexibility and easy processing, inorganic compounds are known for their robust electronic and magnetic properties, their wide range of band gaps and bandwidths, and thermal stability. Combining both components into a single hybrid compound gives rise to materials with properties that are associated with both components. Moreover, as the variety of organic and inorganic materials that can be implemented into a hybrid structure is very large, organic-inorganic hybrids represent a large set of materials with very distinct physical properties.

The main goal of my research was to gain a better fundamental understanding of structure-property relations in organic-inorganic hybrids. Thus, I wanted to increase the understanding of how displacements in the crystal structure can change fundamental physical properties in organic-inorganic hybrid compounds. I think that this understanding could eventually lead to tools that can be used to design materials with properties for desired applications.

The *Chemistry* of organic-inorganic hybrids is important to tune the materials properties. The organic and inorganic components build up the structure together. Changing either of the two components as a direct influence on the structure of the other component and hence the properties of the compound. I have shown that increasing the size of the organic cation directly influences the structure of the inorganic part and consequently changes the band gap (Chapter 3). By changing the inorganic part, I have shown that structurally similar compounds can have very distinct mechanisms for ferroelectricity (Chapter 6). Moreover, I observed that synthesis conditions play a major role in the end product. The presence of water can lead to secondary phases (Chapter 4), and inert/ambient conditions and the choice of solvent can determine what hybrid phase will form (Chapters 8 and 9).

The *Connectivity* of the inorganic lattice plays a major role in the materials properties. The inorganic lattice often takes the shape of a 3-dimensional network of corner-sharing MX_6 -octahedra, with M a divalent metal and X a halide. I showed that the band gap of the material is directly related to the dimensionality of the inorganic network, *i.e.* 3-dimensional structures, 2-dimensional sheets and 1-dimensional ribbons, as well as to the connectivity of the MX_6 -octahedra, *i.e.* corner-sharing, edge-sharing and face-sharing

(Chapter 4). In addition, I observed that replacing the divalent metal with a trivalent metal gives rise to a 0-dimensional structure of face-sharing MX_6 -octahedra that is related to a polar phase transition (Chapter 5). Moreover, I showed that different connectivity of Jahn-Teller distorted MX_6 -octahedra gives rise to different magnetic interactions (Chapter 9).

Confinement also plays a role in organic-inorganic hybrids. Here, I distinguish between two forms of confinement: physical confinement (*i.e.* confined in space) and electronic confinement. I observed that micropatterning a 2-dimensional hybrid influences the microstructure and the optical properties. Confining the hybrid in a few micron wide channel induces a strong preferential orientation of the crystallites. In addition, the larger grain sizes compared to non-patterned films enhanced the photoluminescence lifetime (Chapter 7). Moreover, I reported that face-sharing MX_6 -octahedra in layered structures create a quantum confinement effect leading to effectively 1-dimensional behavior as shown by the electronic structure (Chapter 3).

To summarize, I have synthesized both new and previously reported organic-inorganic hybrids, studied their crystal structures and measured fundamental physical properties. Subsequently, I have investigated structure-property relations. My findings in this thesis add to the understanding of the chemistry, structures and physical properties of these hybrid materials. Furthermore, I have reported a few tools that are of use for direct tuning of certain physical properties of organic-inorganic hybrids for desired applications.