Epitaxial Hexagonal Boron Nitride: from Color Centers to Wafer Scale Heterostructures

In recent years, hexagonal boron nitride (hBN) has established its position among two dimensional materials as an irreplaceable component of ultrathin heterostructures. Owing to its high durability, wide bandgap, and flat surface with ideally no dangling bonds, it was found to improve the performance and provide protection for other van der Waals crystals. The material has also gathered a lot of attention due to its potential use in quantum technological applications. Point defects in the atomic structure of hexagonal boron nitride can be optically active. The emission from such color centers can be highly sensitive to local environmental properties like strain, electric and magnetic field, making them promising candidates for nanosensors.

This thesis addresses the topic of hBN applications and properties in two ways: on a macro- and nano-scale. In the studies, we use large-area, ultrathin material grown by Metalorganic Vapor Phase Epitaxy (MOVPE) to fabricate heterostructures with other two-dimensional materials.

Chapters from 1 to 4 present the motivation, the theoretical background, and the overview of studied materials, as well as fabrication and characterization methods. Chapter 5 describes the experimental setups used and the developed large-area transfer technique, which is key to other elements of the work.

In chapter 6, we present a combined heteroepitaxial growth method aimed at the large-area fabrication of high optical quality monolayers of molybdenum diselenide ($MoSe_2$) - a representative of 2D material semiconductors. We tested the utility of epitaxial hBN as a substrate that improves the performance of the subsequently grown monolayer crystal. Our studies include two approaches: $MoSe_2$ growth on as-grown and on transferred hBN layers. Morphology, structural, and optical spectroscopy studies unveil a high uniformity and repeatability of fabricated samples. Photoluminescence measurements performed at low temperatures show a high optical quality of the grown $MoSe_2$ layers manifested by narrow and resolved excitonic lines. An additional transfer step of the hBN layer onto a silicon/silicon dioxide substrate allowed us to perform electrical gating of the structures, which enabled control of the well-resolved $MoSe_2$ charged-to-neutral exciton intensity ratio.

In chapter 7, we delve into the nanoscale interactions between 2D magnets and color centers in epitaxial hBN. We show the fabrication scheme and challenges related to the stability of the heterostructures. Optical studies allow us to characterize defects typically found in untreated epitaxial layers. Spectroscopy measurements performed with varying temperature resulted in an intriguing observation of a change in defect-related emission energy and a cut-off correlated with an antiferromagnetic-paramagnetic phase transition in a 2D magnet, chromium thiophosphate. Further studies performed in an external magnetic field showed additional interesting effects. At a magnetic field corresponding to the so-called spin-flop transition in a 2D magnet, the intensity of the hBN color center emission drastically drops, and its energy slightly shifts. We discuss the origin of the observed phenomena, considering mechanical strain and/or changes in an electric and magnetic field as possible explanations. Studies of the hBN/2D magnet heterostructures are supplemented by the results of polarization-resolved optical probing. We present the angular dependencies collected at various magnetic fields for the Raman signal of a 2D magnet and the hBN defect-related emission, which provides valuable information about the symmetry of the electronic states involved in the investigated optical properties.

The thesis is concluded in chapter 8, pointing out opportunities for further development and applications of the presented research.

The described results show that hBN is a prospective material that can already be scaled up and used in macro-sized heterostructures and devices. Simultaneously, hBN provides a great platform for studying basic physical phenomena related to 2D semiconductor systems on the nanoscale. The presented findings, considering color centers in hBN and their interaction with 2D magnets, open a wide path to applications related to quantum technologies and sensing, while the combined growth of $MoSe_2$ on hBN holds great promise for scaling up van der Waals heterostructure fabrication.