

Optical properties of thin layers of transition metal dichalcogenides

Abstract

The research reported in the thesis entitled '*Optical properties of thin layers of transition metal dichalcogenides*' focuses on physical phenomena which emerge in the limit of two-dimensional (2D) miniaturisation when the thickness of fabricated films reaches an atomic scale. The importance of such man-made structures has been revealed by the dynamic research on graphene: a single atomic plane of carbon atoms arranged in honeycomb lattice. Graphene is intrinsically gapless and therefore mainly explored with respect to its electric properties. The investigation of semiconducting materials which can also display the hexagonal crystal structure and which can be thinned down to individual layers, bridges the concepts characteristic of graphene-like systems (K-valley physics) with more conventional properties of semiconductors. This has been indeed demonstrated in a number of recent studies of ultra-thin films of semiconducting transition metal dichalcogenides (sc-TMD). Particularly appealing, from the point of view of optical studies, is a transformation of the bandgap alignment of sc-TMD films, from the indirect bandgap bulk crystals to the direct bandgap system in single layers. The presented thesis work provides a comprehensive optical characterisation of thin structures of sc-TMD crystals. The manuscript is divided into five parts: three main chapters with a preceding introduction and the appendix reporting the supplementary studies of another layered material: hexagonal boron nitride.

Introduction. The fundamental properties of the investigated crystals are presented, especially those which are important from the point of view of optical studies. The discussion includes information on the crystal structure, Brillouin zone and electronic band structure. Also, the general description of the samples' preparation process and experimental set-up is provided.

Chapter 1. Basic optical characterisation of excitonic resonances in mono- and multi-layers of sc-TMDs. The optical response, as seen in the reflectance and luminescence spectra of thin sc-TMD is analysed (mostly for MoSe₂ and WSe₂ materials). The impact of the number of layers and temperature on the optical resonances is studied and interpreted in details. The complementary time-resolved study is also presented.

Chapter 2. Zeeman spectroscopy of excitonic resonances in magnetic fields. The evolution of the optical resonances in an external magnetic field, applied perpendicularly to the layers of sc-TMD materials is investigated. Based on these results, a phenomenological model is developed aiming to describe the linear with magnetic field contributions to the energy of individual electronic states in fundamental sub-bands of sc-TMD monolayers. Furthermore, the effects of optical pumping are investigated in WSe₂ monolayers, which can be tuned by tiny magnetic fields.

Chapter 3. Single photon sources in thin sc-TMD flakes. The uncovering of localised narrow lines emitting centres at the edges of thin exfoliated sc-TMD flakes is discussed. The optical

investigations provide information on their fundamental properties. The presented study covers a broad range of topics, such as the impact of temperature and magnetic field on the optical response of the emitting centres, analysis of their polarisation properties and excitation spectra as well as photon correlation measurements.

Appendix A. Single photon emitters in boron nitride crystals. Hexagonal boron nitride also belongs to the family of layered materials, but it exhibits much larger band gap than semiconducting transition metal dichalcogenides. Narrow lines emitting centres have been observed in boron nitride structures, which reveal multiple similarities to defect centres in wide gap materials. They are characterised in a similar manner as the emitting centres in WSe₂.