Light-Matter Interactions in Low-Dimensional Materials

The thesis you are about to read is the culmination of my three years as a PhD student in the Structured Electromagnetic Materials group at the Technical University of Denmark (DTU). Contained herein is a compilation of the results obtained from the research projects I have been involved in during these three years. The projects have been focused on experimental characterisation of the optical properties of plasmonic structures and two-dimensional (2D) materials, and especially how we can tailor these properties. We begin with an introduction to the optical properties of metals. This leads us to the concept of collective electron excitations called plasmons. We especially focus on the localised surface plasmons hosted by nanoscale metallic structures, and we review their properties in single and coupled configurations. We conclude this part with a presentation of the experimental investigation into the interaction between spherical gold nanoparticles and single-crystalline gold substrates, which are known to host electronic surface states. We detect no change, however, in the optical properties from these states, when we compare to a polycrystalline reference substrate. We ascribe this to the weak effect induced by the surface states compared to other factors such as the surface roughness. Following this, we introduce the family of 2D materials known as the transition metal dichalcogenides (TMDCs). We review their basic electronic and optical properties, and we explore the strong light-matter interaction in these atomically thin materials dominated by electron-hole complexes known as excitons. We also see how it is possible to theoretically model the optical response of such a 2D material on a substrate with the transfer-matrix method, and we use this to extract the dielectric permittivity of WS2 mono- and multilayers. Having the basic theory in place, we demonstrate how it is possible to manipulate the emission energy of excitons in WSe2 monolayers, encapsulated between flakes of hexagonal boron nitride (hBN), by nanostructuring the dielectric environment. This is explained by the reduced binding energy of the excitons from the dielectric screening, and the change in the material band structure from the presence of the hBN. We find that the emission energy increases with decreasing screening from the hBN, an effect that appears to be dependent on whether the metal atom is molybdenum or tungsten. TMDCs, and the cavities are plasmonic structures. In the case of weak coupling, we report photoluminescence quenching in MoS2 when coupled to semi-continuous, thin gold films, known as percolation films. This quenching is ascribed to the introduction of nonradiative pathways by the percolation film. We furthermore show a 50% reduction in the reflected signal, which could stem from enhanced absorption in the MoS2 layer, since we observe a drastically different response without the presence of the 2D material. We then continue to discuss the basic theory of strong coupling, after which we present the experimental confirmation of strong coupling for mono- and multilayers of WS2 coupled to ultra-thin, single-crystalline gold nanodisks. In the monolayer case, we explore this coupling both in scattering and reflection measurements, where we argue that the latter is representative of the absorption properties of the coupled system. For the multilayer, we show a record-high figure of merit for our system consisting of an open plasmonic cavity coupled to a TMDC. We conclude the chapter by briefly exploring the temperature dependence of the optical properties of such a system. Finally, a summary and ideas for future work is provided. Since an outlook is given for the individual projects as they are presented, the outlook is comprised of my thoughts on how to continue the work presented here, if more time was available.

We then move on to discussing the coupling of emitters and cavities in the weak and strong-coupling regimes. The emitters are here represented by the excitons in TMDCs, and the cavities are plasmonic structures. In the case of weak coupling, we report photoluminescence quenching in MoS2 when coupled to semi-continuous, thin gold films, known as percolation films. This quenching is ascribed to the introduction of nonradiative pathways by the percolation film. We furthermore show a 50% reduction in the reflected signal, which could stem from enhanced absorption in the MoS2 layer, since we observe a drastically different response without the presence of the 2D material. We then continue to discuss the basic theory of strong coupling, after which we present the experimental confirmation of strong coupling for mono- and multilayers of WS2 coupled to ultra-thin, single-crystalline gold nanodisks. In the monolayer case, we explore this coupling both in scattering and reflection measurements, where we argue that the latter is representative of the absorption properties of the coupled system. For the multilayer, we show a record-high figure of merit for our system consisting of an open plasmonic cavity coupled to a TMDC. We conclude the chapter by briefly exploring the temperature dependence of the optical properties of such a system. Finally, a summary and ideas for future work is provided. Since an outlook is given for the individual projects as they are presented, the outlook is comprised of my thoughts on how to continue the work presented here, if more time was available.

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