

## **Reference for Mr. Jakub Jasinski's PhD thesis Control of the Excitonic Properties in Transition Metal Dichalcogenide based Structures**

The dissertation manuscript of Mr. Jakub Jasinski is entitled "Control of the Excitonic Properties in Transition Metal Dichalcogenide based Structures".

Before entering the content of the thesis, I would like to mention that Mr Jasinski's work has been well-recognized by the community. To date, he is 1<sup>st</sup> author in two publications that are directly related to this thesis (Chapters 3, 4.1), a further one is submitted and available as preprint. A further manuscript (relating to Chapter 4.2) is in the making. In addition, he contributed to three further papers as coauthor. He presented his work at various international conferences.

Let me now summarize the thesis:

Chapter 1 introduces the basics while referring to the state of the art, good overview of transition metal dichalcogenide (here abbreviated as "TMD") structures depending on layer configuration. Similarly, the intriguing electronic structure with its indirect to direct band gap transition is introduced here. Importantly for the thesis, a detailed overview on the physics of excitons and related charged excitons is given. This includes also dark and bright exciton and interlayer excitons. I miss the discussion of spin-orbit coupling, as it is the reason for many of the physical effects discussed afterwards. Also, I have the impression that the literature is cited in a little biased way. For example, the strong SOC in TMDs has first been proposed theoretically (Schwingenschlögl), or the direct band gap character of MoS<sub>2</sub> has been discussed at the same time by two groups (both papers are cited, but at different locations).

Chapter 2 introduces the experimental methodology, which relies heavily on the methods of optical spectroscopy. In 2.1, the physical background of the employed techniques (Raman, photoluminescence) including the analysis of signal shapes is summarized. Particular attention is paid to the impact of strain on the spectra, which comes both from phonons and by the optoelectronic structure itself. Both effects are then explored in the remaining chapters. Chapter 2.2 is very technical and important for the reproducibility of this work. It will also serve as a useful introduction to the field to new students.

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Chapters 3 relates to a publication of the candidate and as such has already been peer-reviewed by external experts. Here, particular geometric features of GaAs are taken to impose strain on single-layer TMDs. This process works, but is not always straight-forward and a thorough analysis of the samples seems to be very important. Strain is then probed by two techniques: By the Raman shifts (coupling to phonons) and by excitonic transitions. I would have liked a comment on the impact of the GaAs substrate on the exciton binding energy, and on the interactions between single-layer MoS<sub>2</sub> and GaAs substrate.

In Chapter 4 the intriguing interplay between valley-coupled triplet excitons (Dexter-like transfer) is discussed both for intervalley coupling of WSe<sub>2</sub> (4.1, published) and in hybrid structures of WSe<sub>2</sub> with 2D perovskites of type-II band alignment (manuscript in preparation). This effect has not been covered by the literature to a large extent and is a little hard to grasp. Therefore, collaboration with theoretical partners (Malic group) was required to describe the experimental findings, which match, but not yet perfectly. In particular for heterostructures. This chapter offers an additional control parameter for these structures, namely optical polarization coupling to the valley polarization of the materials.

Chapter 5 is the final research chapter, where the tunability of excitons using an electric field in MoSe<sub>2</sub> bilayer is investigated. For that purpose, a rather complex device has been developed that allowed electrostatic gating using graphene of the encapsulated MoSe<sub>2</sub> homobilayer stack. The system offers a rich physics with various exciton types that can be manipulated by the external field. Among them, quadrupolar excitons, emerging due to dipole-dipole coupling of excitons, is probably the most remarkable observation.

The thesis is well structured and well written. The figures are of excellent quality and also well described in self-contained captions. Notably, the work is written in a way that it will help new scientists to enter the field. The science of Mr. Jasinski's thesis as presented in Chapters 3 and 4.1 has already been successfully reviewed in the scope of peer-review when publishing the work in journals that are well recognized in the field. The remaining results are of excellent quality and already deeply interpreted. Therefore, I assume that they will be published soon and without problems.

I enjoyed reading the thesis, even though there are minor shortcomings that can be fixed easily. The work appears to be unique and reflects the deep understanding of a matured PhD student who has become expert in a complex field. In particular the coverage of the very technical device making and optical measuring towards thorough and rather deep theoretical analysis of the results is remarkable. I recommend the acceptance of the dissertation. Although no top-ranked publications emerged so far from this thesis, I consider the work to be excellent. I am looking forward to the defense. If the defense will be as excellent as thesis manuscript I recommend to consider the predicate "with distinction".



Prof. Thomas Heine