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Thesis title: Unraveling the Nature of Excitons and their Interactions through Time-Resolved Photoemission and Optical Spectroscopy

**Research aim:** Excitons – a coulomb bound electron hole pair, dominates the optoelectronic response in two dimensional semiconductors. Optical techniques have provided useful information about the spectroscopic features of the exciton but lack the ability to resolve their momentum coordinate. Resolving the excitons in the momentum space will reveal important information, such as their direct or indirect nature, their wave function, their size, the nature of their interactions and their impact on the quasiparticle bandstructure. In this thesis we aim to study excitons in a variety of van der Waals semiconductor systems using novel ultrafast photoemission and optical spectroscopic techniques to tackle some of the open questions regarding their nature, their interaction, and their impact on the quasiparticle bandstructure.

**Material and method:** We employed ultrafast optical spectroscopic techniques and novel multidimensional photoemission spectroscopy to study the excitons in the different two-dimensional semiconductor systems. We employed time resolved angle resolved photoemission spectroscopy to study the interlayer excitons in Molybdenum disulfide/Tungsten diselenide heterostructures and to observe the impact of strong excitonic field in the monolayer Tungsten disulfide. We also used micro transient absorption spectroscopy to study the exciton-exciton annihilation process in bilayer Black phosphorus.

**Result:** Using time resolved angle resolved photoemission spectroscopy, we were able to resolve the momentum coordinates of the electrons and the holes of the interlayer excitons in Molybdenum disulfide/Tungsten diselenide heterostructures. By simultaneously resolving the momentum coordinates of the exciton bound electrons and the holes we were able to determine the size of the interlayer excitons and their confinement within the moiré unit cell. We found that the interlayer exciton size (diameter = 5.2 nm) was comparable to moiré lattice constant (6.1 nm) but was tightly confined within the moiré unit cell. We also measured the changes in the quasiparticle bandstructure in the presence of high density of excitons in monolayer Tungsten disulfide, demonstrating the first experimental evidence of Floquet effects driven by other bosonic fields such as excitons. At high exciton density, we observed the predicted Mexican hat like dispersion of the valence band resulting from the hybridization between the exciton dressed conduction band replica and the bare valence band. We also reproduce the spectroscopic features associated with the predicted BEC to BCS transition in non-equilibrium excitonic insulators. Finally, using micro transient absorption spectroscopy we were able to resolve the unique exciton-exciton annihilation process in bilayer black phosphorus. We were able to tune the interaction between the anisotropic excitons in black phosphorus from 1D like at low density to 2D like at high density.

**Conclusion:** We used novel photoemission and optical spectroscopic techniques to unravel the nature of the excitons and their interactions which paves the way for uncovering novel non-equilibrium phenomena in two dimensional materials.