Preface

Semiconductors that are thinned down to a few atomic layers can exhibit novel properties beyond those encountered in bulk forms. Transition-metal dichalcogenides (TMDs) such as MoS_2, WS_2, and WSe_2 are prime examples of such semiconductors. They appear in layered structure that can be reduced to a stable single layer where remarkable electronic properties can emerge. Monolayer TMDs have a pair of electronic valleys which have been proposed as a new way to carry information in next-generation devices, called valleytronics. However, these valleys are normally locked in the same energy level, which limits their potential use for applications.

This dissertation presents the optical methods to split their energy levels by means of coherent light-matter interactions. Experiments were performed in a pump-probe technique using a transient absorption spectroscopy on MoS_2 and WS_2 and a newly developed XUV light source for time- and angle-resolved photoemission spectroscopy (TR-ARPES) on WSe_2 and WTe_2. Hybridizing the electronic valleys with light allows us to optically tune their energy levels in a controllable valley-selective manner. In particular, by using off-resonance circularly polarized light at small detuning, we can tune the energy level of one valley through the optical Stark effect. At larger detuning, we observe a separate contribution from the so-called Bloch-Siegert effect, a delicate phenomenon that has eluded direct observation in solids. The two effects obey opposite selection rules, which enables us to separate the two effects at two different valleys.

Monolayer TMDs also possess a strong Coulomb interaction that enhances many-body interactions between excitons, both bonding and nonbonding interactions. In the former, bound excitonic quasiparticles such as biexcitons play a unique role in coherent light-matter interactions where they couple the two valleys to induce opposite energy shifts. The latter are found to exhibit energy shifts that effectively mimic the Lennard-Jones interactions between atoms. Through these works, we demonstrate new methods to optically tune the energy levels of electronic valleys in monolayer TMDs.

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