Quantum crystal structures and phase transitions from first principles

Vibrations of atomic nuclei in crystals are commonly approximated to second order in the atomic displacements. However, this harmonic approximation for the phonons is known to break down in certain scenarios. For example, a displacive phase transition can introduce sizeable anharmonic terms in the potential energy of the nuclei.

In this thesis you will explore a phase transition occurring in layered transition metal dichalcogenides (TMDs), called a charge density wave (CDW) instability. It is characterized by a spatial modulation of the atomic positions and the electronic charge density.

A new theoretical approach has recently been develop to describe anharmonic vibrational properties of crystals, called the Stochastic Self-Consistent Harmonic Approximation (SSCHA). It comprises a minimization of the quantum free energy landscape, to optimize all relevant degrees of freedom of the crystal structure, based on input from first-principles calculations within the density functional theory (DFT) framework.

You will employ this approach to characterize CDW-driven changes in the crystal structure of a TMD material, e.g. tantalum disulfide (TaS_2), as a function of temperature. You will furthermore explore the effect of dimensionality on the CDW behavior by comparing bulk and monolayer structures.