Controlling Optical Coherence in Semiconductors with Electric and Magnetic Fields. A Summary:

The proposed basic research of this OPUS project is set in the context of **light-matter interaction** in solids and is focussed on **optical coherence** in two-dimensional (2D) materials; spanning from classical **quantum wells** (QWs), through **monolayers of transition metal dichalcogenides** (TMDs) to emerging new materials like, **2D magnets**. In direct band-gap semiconductors, the elementary electronic excitations are **excitons**: bound electrons and holes, with their relative motion correlated by the Coulomb attraction. They are usually created by shining a laser onto the sample. Their lifetime is short, spanning from pico- to micro-seconds, depending on the host environment and the exciton recombination is accompanied by photon emission. Thus, over the last 60 years, fundamental properties of excitons have been investigated via **optical spectroscopy**. This vibrant and long-standing research on semiconductor optics is driven by their relevance in opto-electronics impacting our daily life. For decades, epitaxial growth provides nano-structures, where a smaller bandgap material is surrounded by a higher bandgap one. Such built potential wells restrict the motion of the charge carriers in one, two or three dimensions in so-called quantum wells, wires and dots, respectively, and their energies consequently become quantized. **Engineering the quantum confinement of excitons** is a key method to control the light-matter interaction dynamics in semiconductors, where the electron-hole overlap integral to large extent governs the **radiative lifetime T_1**. Another relevant observable in the optics of excitons is their **coherence**. For ensembles, it describes the phase relation within the exciton polarization field. This phase relation, inherited from the excitation laser, also disappears for single excitons owing to **dephasing** processes, for example interactions with the lattice vibrations, i.e. phonons, usually being faster than T_1. Conversely, a single exciton mimics a quantum two-level system and its coherence reflects the degree of the quantum superposition of an excitonic qubit, which is characterized by a **dephasing time T_2**.

The first objective of this OPUS project is to **control the coherent dynamics and couplings of excitons in innovative semiconductor devices via dynamical tuning of their quantum confinement**. In epitaxy, the latter is achieved by the material composition. It is therefore intrinsically fixed by the growth design. Moreover, it suffers from the disorder of the alloy composition, as well as the fluctuating sizes and shapes of the trapping potential on an atomic scale. These limitations can be overcome in novel semiconductor devices proposed here used to dynamically control the quantum confinement, allowing us to vary the dimensionality from 2D and 1D electron gases to 0D, when strictly one exciton is confined within the potential trap. In quantum nano-electronics a confirmed strategy to confine individual electrons in so-called electrostatically defined quantum dots is by fabricating planar metallic gates on top of a two-dimensional high mobility electron gas in a pure GaAs/AlGaAs heterostructure and controlling the transport in a single-electron regime.

Because of their charge neutrality, confining the excitons electrostatically is even more challenging. This has very recently been accomplished by exploiting the DC Stark effect and enhanced polaronic (exciton in a presence of charge carriers) interactions in **a gated $MoSe_2$ monolayer** (D. Thureja et al. Nature **606**, 298 (2022)). Such a representative TMD material hosts excitons of high binding energy E_B of around 200 meV. They can thus withstand strong electric field gradients beyond $10^3 V \cdot \mu m^{-2}$, otherwise leading to the exciton break-down and ionization in standard semiconductors, like gallium arsenide.

Moreover, when encapsulated in a high quality hexagonal boron nitride (hBN), the electronic disorder experienced by the $MoSe_2$ excitons can be largely suppressed, preventing their spatial localization, permitting long diffusion lengths and thus enabling the **quantization of their centre of mass momentum**. To generate strong in-plane electric field gradients one needs to furnish hBN/$MoSe_2$/hBN heterostructures with contacts and gates on the top and bottom side of a stack. This can nowadays be achieved by **electron beam lithography and metal deposition**, combined with **stacking techniques** to fabricate van der Waals heterostructures – the strategy which will also be pursued in this OPUS project.

To demonstrate the variable exciton confinement and the emergence of their coherent coupling confined at neighbouring quantized levels, we will use **coherent ultrafast nonlinear spectroscopy**. The 2D excitons in $MoSe_2$ display a sub-ps radiative lifetime. An increase of T_1 and T_2 times when varying the gate voltages within a lateral p-i-n junction will be a sought-after signature of their variable quantum confinement and the crossover from two- towards one-dimensional character. Similarly, by using CrSBr individual layers, which is a 2D magnetic van der Waals material hosting robust excitons, we will demonstrate a variable exciton spatial configuration and confinement by applying vectorial magnetic fields, within barely 1 Tesla range.

This challenging **project thus opens the field of exciton coherent control under tunable confinement** via electric and magnetic fields. The project will benefit from my engaged collaborations with the world-wide recognized experts in that field, specifically with prof. Ataç Imamoglu from ETH Zurich, and prof. Jonathan Finley and dr Nathan Wilson at Walter Schottky Institute, TU Munich.