EQUAISE

Enabling Quantum Information by Scalability of Engineered Quantum Materials

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In Short

- Investigate the electronic properties of bilayer structures including a transition-metal dichalcogenide nanobubble or nanowrinkle and a monolayer.
- Assess the ability of these systems to act as single-photon emitters.
- Study the role of strain in the electronic properties of transition-metal dichalcogenide heterobilayers.

Transition metal dichalcogenide (TMDC) monolayers have emerged as the most promising class of next-generation semiconductors. Their exceptional light-matter interaction properties, their mechanical flexibility, and their ability to be integrated into nanostructured architectures make them ideal candidates as single-photon emitters for emerging quantum information technologies. The experimental efforts that are relentlessly going on in this field need a robust complement from theory in order to rationalize the behavior of the material with respect to the large number of degrees of freedom that can be applied to modulate their properties. To create single-photon emitters based on TMDC monolayers, an effective strategy that was recently proposed experimentally consists of modifying their structure upon strain until a “nanobubble” is formed [1]. A schematic representation of such a system is provided in Fig. 1. In the lab, this is realized by pumping H_2 molecules between two TMDC sheets. While effective simulations, such as those included in Refs. [1,2], provide a helpful platform to interpret the experimental results, first-principles calculations based on density-functional theory (DFT) can offer unprecedented insight to rationalize and predict the behavior of the material as a function of its structural deformations without the need for empirical parameters.

In the first phase of the project, we studied mechanically deformed TMDC-based nanostructures such as free-standing nanowrinkles as well as nanobubbles (see Fig. 1, top panel). We investigated the electronic properties of these systems focusing in particular on their band structure and their atomically projected density of states. The former was unfolded in the Brillouin zone of the pristine TMDC

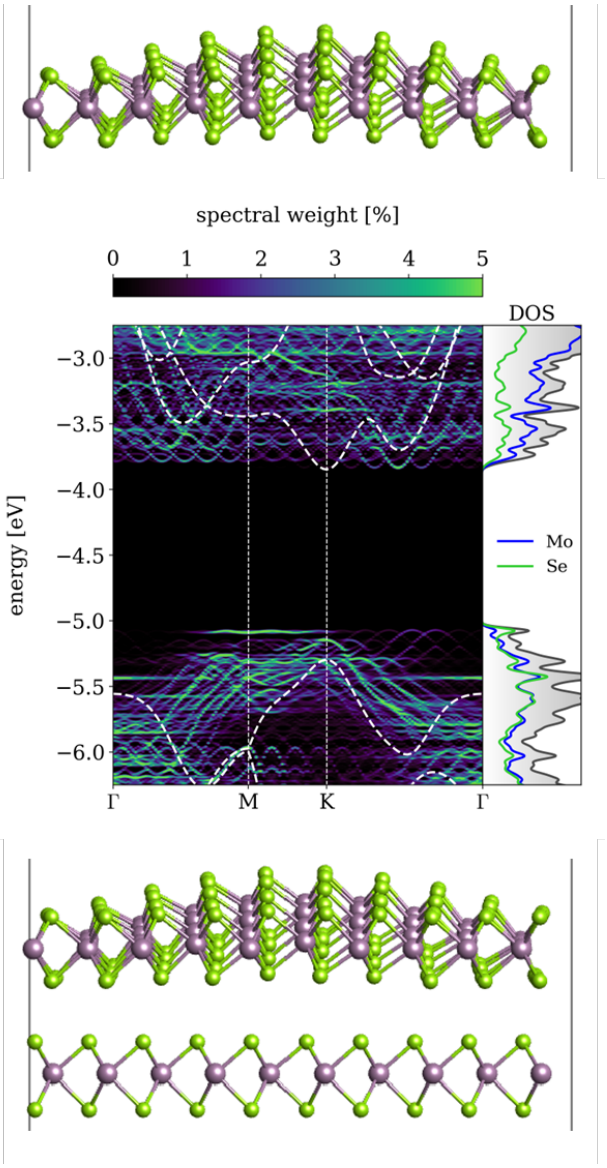

Figure 1: *Ball-and-stick representation of a $MoSe_2$ nanobubble in its unit cell (top) together with its band structure and atom-projected density of states (DOS), bottom panel. The white dashed lines represent the bands of the flat monolayer under 10% strain and the shades of color indicates the spectral weight of the electronic states of the nanobubble with the same strain as the monolayer downfolded in the Brillouin zone of the flat monolayer. The bottom panel shows the same layer with nanobubble plus additional flat layer*

monolayer for a clearer visualization (Fig. 1, middle panel). The presence of a flat band at the top of the valence band of such a system is a clear indication of the favorable characteristics of this system as a single photon emitter. The studies on quasi-one-

dimensional nanowrinkles showed equally intriguing characteristics.

In order to deepen this analysis and to better reflect the experimental samples, in which the nanobubbles and nanowrinkles are deposited on a flat layer of the same material, we plan to model such systems (Fig. 1, bottom panel) in the framework of DFT. In parallel, to provide a more accurate estimate of band gap and band edges, we will perform GW calculations to include the quasi-particle correction in the previously obtained DFT results for the free-standing structures.

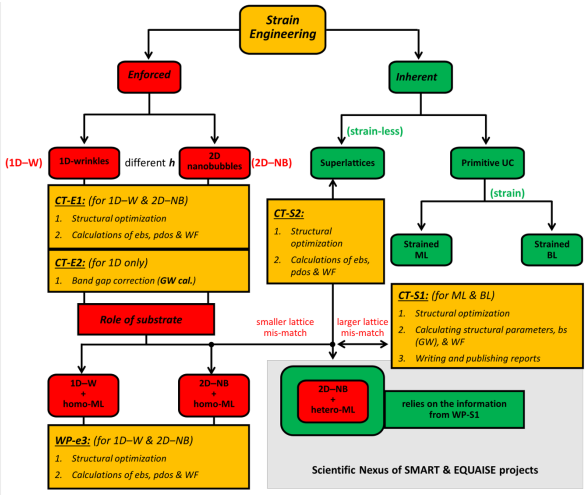

Figure 2: *Breakdown of the sub-tasks, summary and connections between the calculations planned in this project.*

Within the related granted project (SMART), we will explore the role of strain in the electronic and optical properties of TMDC heterobilayers. In previous work, we showed that TMDC heterostructures may exhibit significantly different characteristics in comparison with their isolated components [6,7]. The breakdown of task and the connections between the two parts of the project are sketched in Fig. 2. Taking $MoTe_2$ as reference platform, in light of its ideal band-gap size for telecommunication and quantum-information applications, we will consider heterostructures formed by this material TMDC monolayers with larger lattice parameters than $MoTe_2$. Given the different lattice parameters of the aforementioned systems, strain will play a crucial role in the formation of the heterobilayers. We aim to purposely use this parameter in connection with the unit cell size to tune the electronic structure of the two-dimensional interfaces in order to achive the optimal conditions for band-gap size and optical response in view of the target applications.

The whole work proposed in this application will be performed in the framework of DFT using the package Quantum Espresso [3] and Vienna Ab initio Simulation Package (VASP). This research will be carried out in close collaboration with a number of partners with complementary experimental and theoretical background from Germany and abroad. In the framework of these consortia, our *ab initio* results will provide parameters for model Hamiltonian calculations on realistic nanostructures, which are not affordable for quantum-mechanical, atomistic simulations. Moreover, our findings will contribute to the understanding of the experimental characterization of the nanostructures and, in a feedback loop with several cooperation partners, to the optimization of their design on the nanoscale.

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More Information

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Project Partners

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DFG Subject Area

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