

# Advancing hybrid inorganic/organic systems for future photovoltaics

## Electronic properties of multi-layered hybrid inorganic/organic systems: the example of bilayer MoS<sub>2</sub> with pyridine as intermedium

Wenxiao Liang<sup>1</sup>, Ronaldo Rodrigues Pelá<sup>2</sup>, Claudia Draxl<sup>1</sup>, <sup>1</sup> Institut für Physik and CSMB, Humboldt-Universität zu Berlin

<sup>2</sup> Supercomputing Department, Zuse Institute Berlin

### In Short

- Tri-layer systems composed of an organic molecule embedded between two transition-metal-dichalcogenide layers represent a new promising material class for optoelectronic nanodevices.
- Strong exciton couplings, chemical versatility, and tunable electronic properties are expected to figure among their attractive features.
- However, the real benefits of this material class remain largely unknown, as they have not been deeply investigated.
- The project will address this question. We will employ the *GW* method, regarded as the state-of-the-art for computing the electronic structure of materials, to accurately evaluate the electronic properties of bilayer MoS<sub>2</sub> with pyridine as intermedium, chosen as a benchmark case of this new material class.

Hybrid inorganic/organic systems (HIOS) consisting of a two-dimensional (2D) transition metal dichalcogenide (TMDC) monolayer interfaced with a  $\pi$ -conjugated organic molecule have emerged as a promising platform for exploring excitonic phenomena, charge transfer mechanisms, and orbital hybridization at the nanoscale [1]. Notably, the modulation of the dielectric environment in TMDC monolayers through molecular functionalization has been demonstrated to induce significant band renormalization, a topic extensively investigated by our group [2]. Despite the immense potential of such hybrid architectures for tunable electronic and optoelectronic applications, a critical challenge persists: organic molecules adsorbed on TMDC surfaces are unstable. A promising strategy to counteract this issue consists of encapsulating the organic molecules within a TMDC bilayer. While multilayered inorganic TMDC heterostructures have been widely studied, the fundamental characteristics of this material class, hybrid tri-layer systems involving two TMDC monolayers and an organic molecule, remain largely unexplored.

These properties are critical for tailoring the performance of materials in emerging optoelectronic applications such as solar cells, photo-detectors, and light-emitting devices.

In this work, we aim to predict the electronic properties of a material belonging to this class, namely molybdenum disulfide (MoS<sub>2</sub>) monolayers with a  $\pi$ -conjugated organic molecule (pyridine) positioned as an intermedium. This is intended to be a prototype case, as pyridine is one of the smallest  $\pi$ -conjugated organic molecules, and MoS<sub>2</sub> is the simplest, yet most studied, TMDC. The insights gained from this representative case will serve as a foundation for investigating more complex systems belonging to the same family.

Since this material has not yet been synthesized, our study will have significant predictive value. Therefore, to ensure high accuracy, we will employ the state-of-the-art methodology to study electronic properties, the *GW* approach, known for results in excellent agreement with experiments. By obtaining the electronic structure, we will then be able, in the second step, to investigate the optical properties of our benchmark material and assess its potential for optoelectronic applications. Additionally, our calculations will also enable us to comprehend which factors contribute to either enhanced or diminished performance regarding optoelectronic applications. This understanding can, in turn, guide future optimization efforts.

All calculations will be performed with the `exciting` code [3], a full-potential all-electron package. `exciting` employs the linearized augmented plane wave method, the gold standard for solving electronic structure problems, and can also achieve the highest numerical precision in *GW* calculations [4], two aspects necessary for predictive quality.

### WWW

<https://sol.physik.hu-berlin.de/>

### Reference

- [1] P. Puschnig, P. Amiri, C. Draxl, *Phys. Rev. B* **86**, 085107 (2012). doi:10.1103/PhysRevB.86.085107
- [2] I. Gonzalez Oliva, F. Caruso, P. Pavone, C. Draxl, *Phys. Rev. Mater.* **6**, 054004 (2022). doi:10.1103/PhysRevMaterials.6.054004

- [3] A. Gulans, S. Kontur, C. Meisenbichler, D. Nabok, P. Pavone, S. Rigamonti, S. Sagmeister, U. Werner, C. Draxl, *J. Phys.: Condens. Matter* **26**, 363202 (2014). doi: 10.1088/0953-8984/26/36/363202
- [4] D. Nabok, A. Gulans, C. Draxl, *Phys. Rev. B* **94**, 035418 (2016). doi:10.1103/PhysRevB.94.035418

#### Funding

China scholarship council

#### DFG Subject Area

3.21-02