Modelling defective MoS_2 and Au interfaces in 2D memristors by combining DFT with Green's function surface calculations

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Atomically thin single-layer molybdenum disulfide (MoS_2) is a two-dimensional material with highly desirable mechanical, electronic, and optical properties. In addition, its reduced dimensionality and ultra-thin size could enable the fabrication of a new generation of very compact and low-power devices going beyond the conventional CMOS technology.
In this context, 2D memristors for neuromorphic computing applications [1] based on single-layer MoS_2 present several advantages with respect to conventional devices based on transition metal oxides: good flexibility, high transparency, and the potential to work when applying low voltages (0.1 - 0.2 V), thus allowing the fabrication of very compact and energy-efficient devices.
However, the working mechanism of 2D memristors is still far from being understood: the resistive switching may non-trivially depend on several factors, such as device architecture, metal electrodes, and the quality of the MoS_2 film. Small and extended defects in MoS_2 films, which are introduced during the material growth, are thought to play a crucial role not just in the properties of MoS_2 itself but also in the quality of the metal contact.
Indeed, at present, MoS_2 films are far from being pristine, and there is still much debate around the role of defects in the physics of the device.
Thus, to shed light onto the physics of metal-MoS_2 interfaces, we carry out atomistic computer simulations in the framework of density functional theory (DFT). We employ surface calculations based on the Green's function formalism to construct realistic interfaces and to compute surface properties [2]. To model the metal electrode, we choose Au as it is commonly one of the most popular choices, and it has been successfully used to develop 2D memristors [3]. As we aim to bridge the gap between materials' properties and device physics, we investigate the effect of common defects on MoS_2 (i.e., vacancies and substitutions) on the physics and chemistry of Au-MoS_2 interfaces [4].
To the best of our knowledge, this is the first attempt to thoroughly model defective MoS_2 interfaces with DFT coupled with the Green's function formalism.
Ultimately, our study constitutes the first step of a more comprehensive multi-scale modelling approach, in which the aim is to construct a full atomistic-to-device level model that can aid us in elucidating the working mechanism of 2D memristors based on single-layer MoS_2.

[1] EU H2020 NeurONN Project, www.neuronn.eu.
[2] S. Smidstrup et al. "First-Principles Green's Function Method for Surface Calculations: A Pseudopotential Localized Basis Set Approach," Phys. Rev. B, 96, 195309, **2017**.
[3] R. Ge et al., "Atomristor: Nonvolatile Resistance Switching in Atomic Sheets of Transition Metal Dichalcogenides," *Nano Lett.*, 18, 434-441, **2017**.
[4] G. Boschetto et al. "Ab Initio Computer Simulations on Interfacial Properties of Single-Layer MoS_2 and Au Contacts for Two-Dimensional Nanodevices" ACS Appl. Nano Mater. Just published, **2022**.