**Supporting Information**

# Non-volatile resistive switching mechanism in single-layer $MoS_2$ memristors: Insights from ab initio modelling of Au and $MoS_2$ interfaces

Gabriele Boschetto,[1] Stefania Carapezzi[1], and Aida Todri-Sanial[1,2]

[1]Laboratory of Computer Science, Robotics, and Microelectronics, University of Montpellier, CNRS, 161 Rue Ada, 34095 Montpellier, France
[2]Department of Electrical Engineering, Eindhoven University of Technology, Groene Loper 3, 5612 AE Eindhoven, Netherlands

E-mail: gabriele.boschetto@lirmm.fr; aida.todri@lirmm.fr

## 1. Stability of defects in free-standing $MoS_2$

We assessed the effect of S vacancies, Au substitution, and different combinations of both on free-standing monolayer $MoS_2$ by computing formation energies of selected defects.

***Table S1.*** *Formation energies of selected defects in free-standing $MoS_2$.*

| **Defect** | **$E_F[MoS_2]$ (eV)** | **$E_F[V_S]$ (eV)** |
|---|---|---|
| $V_S$ | 2.75 | - |
| $V_{S2}^A$ | 5.45 | 2.69 |
| $V_{S2}^P$ | 5.48 | 2.73 |
| $Au_S$ | 3.48 | 0.72 |
| $Au2_{S2}^A$ | 7.27 | 4.52 |
| $Au2_{S2}^P$ | 6.64 | 3.88 |

The structural stability of defects can be assessed by computing their formation energies (see Table S1). To compute such a quantity, we considered two cases: i) a defect-free $MoS_2$, and ii) a $MoS_2$ sample with a $V_S$ density of ~2%, in line with typical defect densities found in CVD-$MoS_2$. Formation energies computed with the latter approach ($E_F[V_S]$) give us an estimate of how likely it is to generate new S vacancies (and Au to fill such vacancies) when the material is already defective, as it is the case in $MoS_2$ nanodevices. Formation energies computed with defect-free $MoS_2$ ($E_F[MoS_2]$) confirmed that $V_S$ is the most thermodynamically stable defect ($E_F[MoS_2]$ = 2.75 eV), in accordance with previous studies. Additionally, it is worth noting that $E_F[MoS_2]$ of an axial ($V_{S2}^A$) and an in-plane S di-vacancy ($V_{S2}^P$) is lower than the sum of $E_F[MoS_2]$ for two infinitely separated S vacancies, which suggests the slight tendency of $V_S$ to form clusters. We also found that $Au_S$ has a higher $E_F[MoS_2]$ (3.48 eV) than a simple S vacancy, which is the outcome of both the removal of an S atom and the addition of an Au atom from/to the defect-free $MoS_2$. We could also observe the tendency of Au atoms to cluster in-plane rather than axially. When considering the material with a $V_S$ density of ~2%, we were able to confirm that additional S vacancies are slightly more likely to form in the proximity of

an already existing vacancy. Indeed, $E_F[V_S]$ for one additional S vacancy is slightly lower than $E_F[MoS_2]$ for the single S vacancy. Moreover, we found that the formation energy of $Au_S$ significantly decreases ($E_F[V_S]$ = 0.72 eV), making this the most energetically favourable defect. Once again, we found that Au atoms tend form small clusters in-plane rather than axially out-of-plane.

**2. Partial Charges of Selected Interfaces**

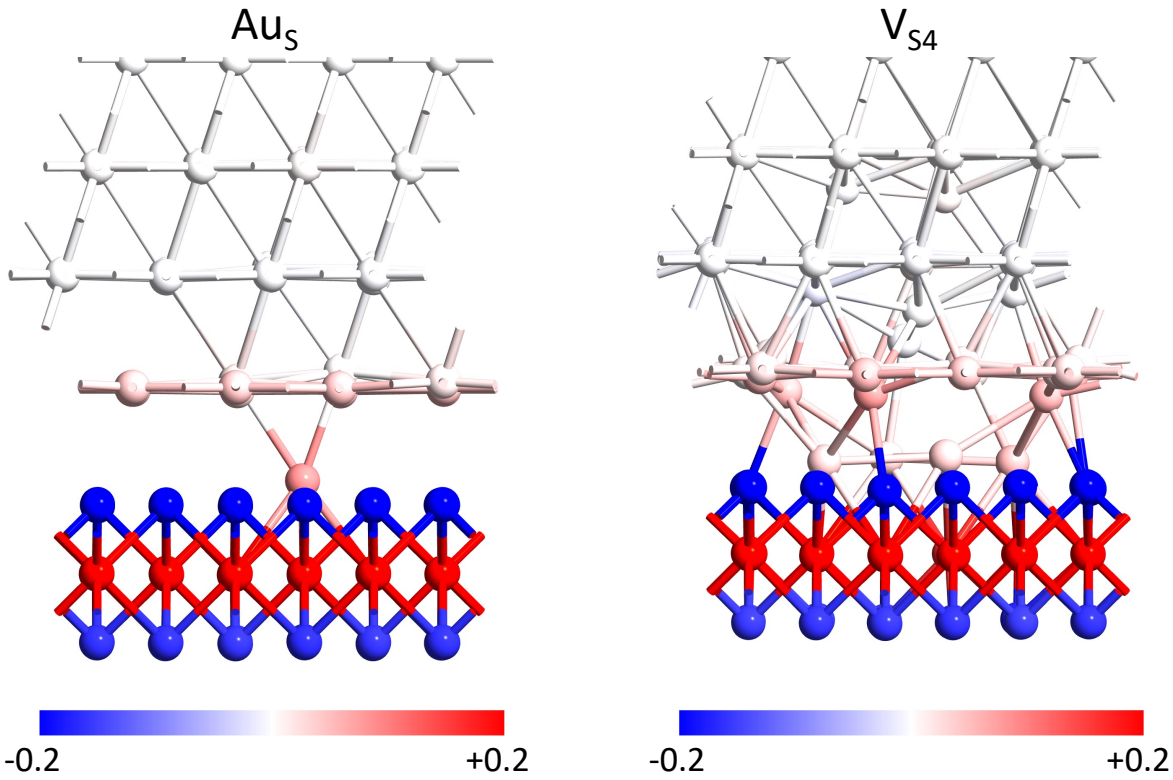


***Figure S1.*** *Mulliken population analysis of $Au_S$ and $V_{S4}$ interfaces. Negative partial charges are in blue, whereas positive partial charges in red.*