

Collective Dipole-Dominated Doping of Transition Metal Dichalcogenides: Orientation and Magnitude Control via the Supramolecular Approach

Ye Wang¹, Sai Manoj Gali², Amine Slassi², David Beljonne², Paolo Samorì¹

¹ University of Strasbourg, CNRS, ISIS UMR 7006, 8 Allée Gaspard Monge, F-67000 Strasbourg, France.

² Laboratory for Chemistry of Novel Materials, Université de Mons, Place du Parc 20, 7000 Mons, Belgium.

ye.wang@unistra.fr

Molecular doping is a powerful, tuneable, and versatile method to modify the electronic properties of 2D transition metal dichalcogenides (TMDs). While electron transfer is an isotropic process, dipole-induced doping is a collective phenomenon in which the orientation of the molecular dipoles interfaced to the 2D material is key to modulate and boost this electronic effect, despite it is not yet demonstrated. A novel method toward the molecular functionalization of mono-to-few layer TMDs relying on the molecular self-assembly of metal phthalocyanine and the orientation-controlled coordination chemistry of axial ligands is reported here. It is demonstrated that the subtle variation of position and type of functional groups exposed on the pyridinic ligand, yields a molecular dipole with programmed magnitude and orientation which is capable to strongly influence the opto-electronic properties of TMDs. In particular, experimental results revealed that both p- and n-type doping can be achieved by modulating the charge carrier density up to $4.8 \times 10^{12} \text{ cm}^{-2}$. Density functional theory calculations showed that the doping mechanism is primarily resulting from the effect of dipole-induced doping rather than charge transfer. The strategy to dope TMDCs is a highly modulable and robust, and it enables to enrich the functionality of 2D materials-based devices for high-performance applications in optoelectronics. [1]

References

[1] Y. Wang, S. M. Gali, A. Slassi, D. Beljonne, P. Samorì, *Adv. Funct. Mater.* 2020, 30, 2002846.

Figures

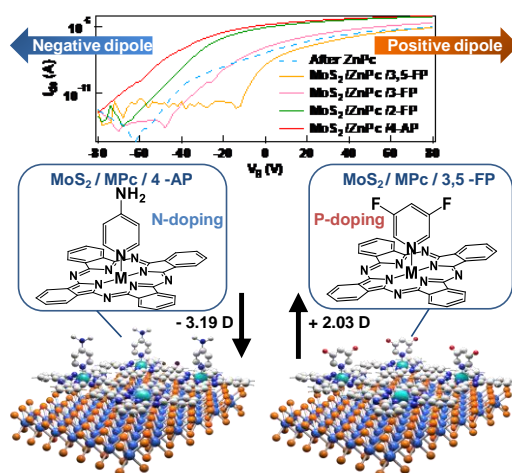


Figure 1: Collective dipole-induced doping of monolayer MoS₂ by axial ligands.