Harnessing the Charge Transport in Covalently Interconnected TMD Networks

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Solution-processed semiconducting transition metal dichalcogenides (TMDs) are at the centre of an ever-increasing research effort in printed (opto)electronics[1]. However, device performance is limited by structural defects resulting from the exfoliation process and poor inter-flake electronic connectivity.

We developed a new molecular strategy to simultaneously heal sulfur vacancies (V_S) in solution-processed MS_2 (M = Mo, W, and Re) and increase the inter-flake electronic connectivity by means of dithiolated molecular systems. By taking advantage of π-conjugated dithiolated molecules (HS-R-SH), we proved *via* diverse multiscale analysis the simultaneous: i) healing of V_S to restore the MS_2 crystal structure and decrease the related stoichiometric deficiencies acting as charge scattering centres, ii) the covalent bridging of adjacent flakes, resulting in an enhanced charge carrier transport through an interconnected network.

Our approach represents an innovative and universal functionalization method capable of improving the performance of devices based on solution-processed MS_2 for large-area electronic applications. In particular, we implemented this strategy in liquid-gated thin-film transistors (LG-TFTs), boosting their characteristics by one order-of-magnitude and reaching state-of-the-art electrical performance characterized by competing field-effect mobilities (μ_{FE}) and I_{ON} / I_{OFF}, along with the fastest switching speed reported to date for devices of this kind[2]. Moreover, covalently interconnected MS_2 networks show additional unique features, such as improved water stability and mechanical robustness.

REFERENCES

[1] Wang *et al.*, Nature Nanotechnology, 7 (2012), 699-712
[2] Kelly *et al.*, Science, 356 (2017), 69-73

FIGURES

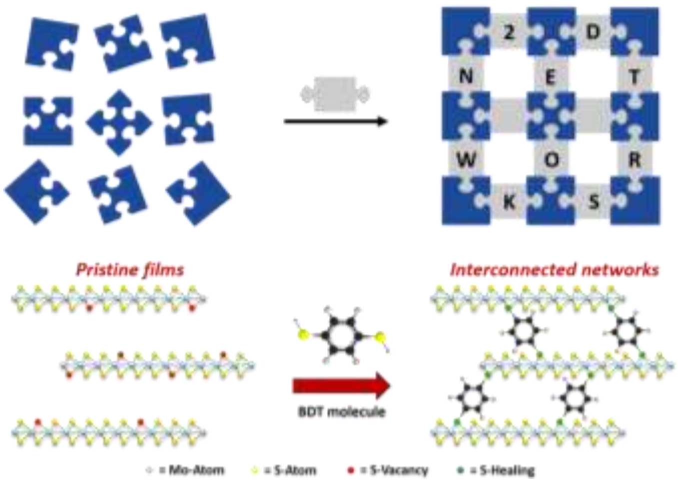

Figure 1: Sketch of V_S healing mechanism in MS_2 films by means of dithiolated molecules to form networks.