Harnessing the Charge Transport through Films of Liquid-Phase Exfoliated TMDs

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During the last five years, the scientific community has witnessed a tremendous progress on liquid-phase exfoliation (LPE) of Transition Metal Dichalcogenides (TMDs),[1] in combination with chemical approaches adopted to finely tune their properties.

Here, we describe an unprecedented molecular strategy to increase interflake electronic connectivity in TMD networks, taking advantage of their inherent sulfur vacancies (V_S),[2] generated during the exfoliation process, and their healing by means of thiolated molecules.

The starting MS_2 dispersions (MoS_2, WS_2 and ReS_2)[3] were drop-casted onto interdigitated gold electrodes, supported on Si-SiO_2, and successively exposed to solutions of thiolated molecules under inert atmosphere. We focussed our attention on the nature of the healing molecules and explored, *via* multiscale characterizations, the effects coming from monothiolated molecules (Thiophenol, TP) and dithiolated molecules (1,4-Benzenedithiol, BDT). We found that MS_2-based devices treated with BDT exhibit superior electrical performance, in terms of charge carrier mobility μ_{FE} and I_{ON}/I_{OFF}.

Such an increased performances can be ascribed to a better interflake connectivity within the networks, attained by means of difunctional molecules which can bridge, mainly *via* edges and secondarily *via* basal plane, adjacent TMD nanosheets, thereby generating percolation pathways for the efficient charge transport within the film.

Remarkably, this work paves the way to the *ex situ* functionalization of TMD networks *via* molecular approaches and defect engineering, allowing a fine tailoring of the resulting physicochemical properties by proper design of the functionalizing species.

References

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[3] Kelly A.G. *et al.*, Science, 356 (2017), 69-73

Figures

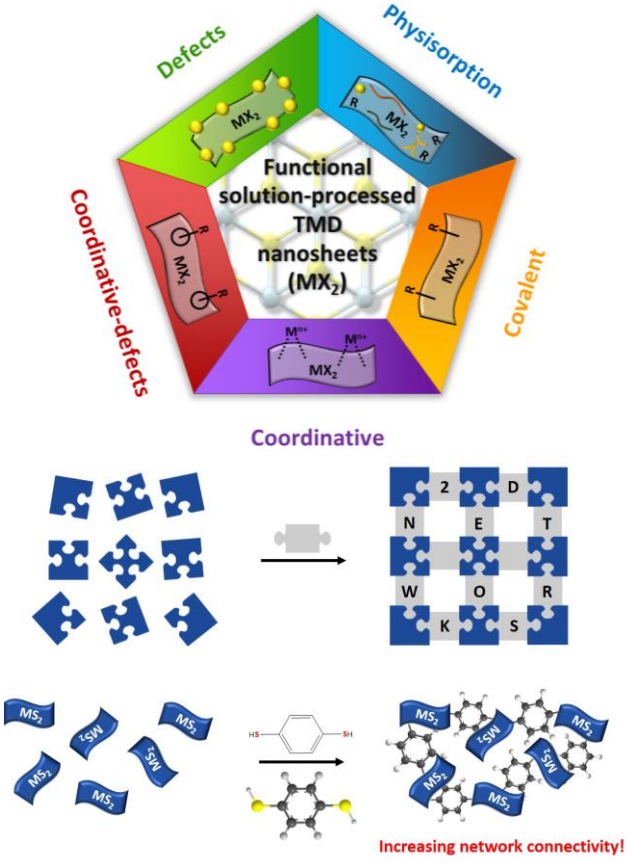

Figure 1: Main functionalization strategies for LPE TMDs (top) and sketch illustrating the approach (coordinative-defects) adopted to increase the connectivity of TMD networks *via* healing of V_S (defect-engineering) by thiolated molecules.