Engineering Excitons, Polaritons, and Defects for 2D Semiconductor Optoelectronics

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Because of their direct bandgaps and atomic-scale thickness, monolayer transition metal dichalcogenides (TMDs) have emerged as appealing materials for both classical and quantum optoelectronics. These 2D semiconducting materials, such as MoS_2 and its related compounds, can support diverse non-trivial optical phenomena such as valley-selective exciton-polaritons and quantum emission from localized excitonic defects. Harnessing these interesting features for applications requires controlled tuning and manipulation to tailor the optical phenomena. The exposed 2D surface of these layered materials provides an active playground for manipulating the optical and electronic environment of these materials [1]. Here, we present examples of intentional engineering of TMD optical phenomena, ranging from strong light-matter interactions to light emission from localized defects.

An interesting example of optical engineering of TMDs is found in the strongly-coupled exciton-polaritons that preserve the valley degree of freedom of TMDs [2]. The valley-sensitive properties of these hybrid light-matter quasiparticles can persist to room temperature with emission polarization that can be tuned using microcavity parameters familiar from photonics. Although dynamics of these novel quasiparticles are sensitive to the optical environment, in many applications charge dynamics are of prime importance. Coating with highly-customizable organic molecules leverages the exposed 2D surface of these materials for a versatile approach for control of optical phenomena. We show that phthalocyanine-MoS_2 heterojunctions have optical response that depends sensitively on the metal core atom of the molecule. This behaviour can be associated with charge transfer form d-orbitals based on experiments and electronic structure calculations [3]. Measurements have revealed strong quenching of MoS_2 defect emission when in a phthalocyanine-MoS_2 heterostructure. This effect is likely linked to the metal core serving as a large donor of charge. The defect quenching does not occur in MoS_2 layers treated with metal-free Pc molecules, underlining the importance of the metal core in heterostructure charge transfer dynamics. The ability to influence optical response and defect emission using configurable organic molecules highlights the potential for optoelectronic engineering in low-dimensional heterostructures.

References

[1] T. LaMountain, E. J. Lenferink, Y.-J. Chen, T. K. Stanev, N. P. Stern, Frontiers of Physics 13 (2018) 138114.
[2] Y.-J. Chen, T. K. Stanev, J. D. Cain, V. P. Dravid, and N. P. Stern. Authors, Nature Photonics, 11 (2017) 431.
[3] S. Amsterdam, T. K. Stanev, et al., in press.

Figures

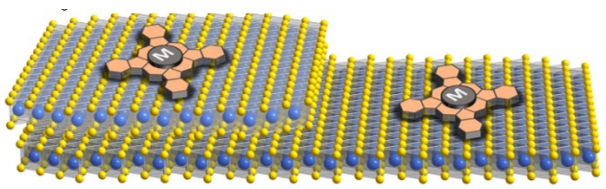

Figure 1: Illustration of bilayer and monolayer materials with metal-core phthalocyanine organic dyes deposited on top.