Tuning Optoelectronic Properties of Transition Metal Dichalcogenides via Dopant-Defect Interactions

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Abstract: Two-dimensional transition metal dichalcogenides (TMDs) have garnered significant attention for their potential applications in optoelectronics, sensing, and quantum computing, owing to their unique electronic and optical properties. Substitutional doping of TMDs with metal atoms has been widely employed to control charge carrier concentrations and exciton dynamics. However, these materials are prone to defects, such as chalcogen vacancies, which can alter their optoelectronic properties and strongly interact with introduced dopant atoms. Understanding these dopant-defect interactions is critical for optimizing TMDs for next-generation electronic and photonic applications. In this study, we investigate how dopant-defect interactions influence the optoelectronic properties of TMDs using photoluminescence (PL) and transient absorption spectroscopy. Additionally, we correlate these findings with field-effect transistor measurements and atomic-scale scanning transmission electron microscopy imaging to gain a deeper understanding of how these interactions affect exciton dynamics.

Our work reveals that doping monolayer MoS_2 with Rhenium atoms during metal-organic chemical vapor deposition reduces sulfur vacancies by an order of magnitude, enhancing both free electron density and mobility. Similarly, vanadium doped WS_2, synthesized via atmospheric pressure chemical vapor deposition, shows enhanced PL followed by a decrease in negative trions when doped at concentrations below 0.2 atom%. However, higher vanadium concentrations result in dopant-defect coupling which can lead to the formation of mid-gap states and quenching of PL, indicating a delicate balance between dopant concentration and defect interaction. Our findings offer a comprehensive framework for controlled synthesis and substitutional doping of TMDs, which can be utilized to fine-tune their optical, electronic, and magnetic properties for advanced applications, including catalysis, dilute magnetic semiconductors, and quantum photonic devices.