

Direct Observation of Strong Spin-Layer Coupling in Bilayer MoS₂

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This study presents groundbreaking findings on the spin-layer locking effect within bilayer molybdenum disulfide (MoS₂), extending the domain of valleytronics beyond monolayer transition metal dichalcogenides (TMDCs). Utilizing a comprehensive experimental approach, the research delves into the distinction between spin-valley and spin-layer locking effects, facilitated by the unique electronic band structures of 2D semiconductor materials. By employing circularly polarized light, the study selectively excites electrons in specific valleys or layers, enabling the observation of the degree of polarization (DOP) which quantifies the extent of spin-valley and spin-layer coupling. Significantly, the research underscores the novel application of valleytronics in bilayer TMDCs, challenging the previously held belief that high DOP is exclusive to monolayers with lattice inversion asymmetry. Through meticulous sample preparation and measurement, including the use of suspended MoS₂ bilayers to exclude substrate influences, this study experimentally confirms that DOP in bilayer MoS₂ stems from spin-layer coupling rather than substrate effects. The findings are bolstered by Raman spectroscopy and photoluminescence (PL) analysis at both room and cryogenic temperatures, revealing that DOP values are consistently higher in bilayer MoS₂ across all temperatures, indicative of the robustness of the spin-layer locking effect. Furthermore, the investigation explores the role of substrate-induced strain at 4 K, elucidating its impact on DOP through thermal expansion coefficient disparities between SiO₂ and MoS₂. This nuanced understanding of strain effects and the associated symmetry breaking contributes to the observed increase in DOP, highlighting the complexity of electron dynamics in bilayer structures. This work not only affirms the presence of the spin-layer locking effect in bilayer MoS₂ but also expands the potential for valleytronics applications to include bilayer TMDCs. The experimental evidence provided opens new avenues for research in TMDCs-based valleytronics, suggesting a broader applicability of this emerging field beyond its initial scope.

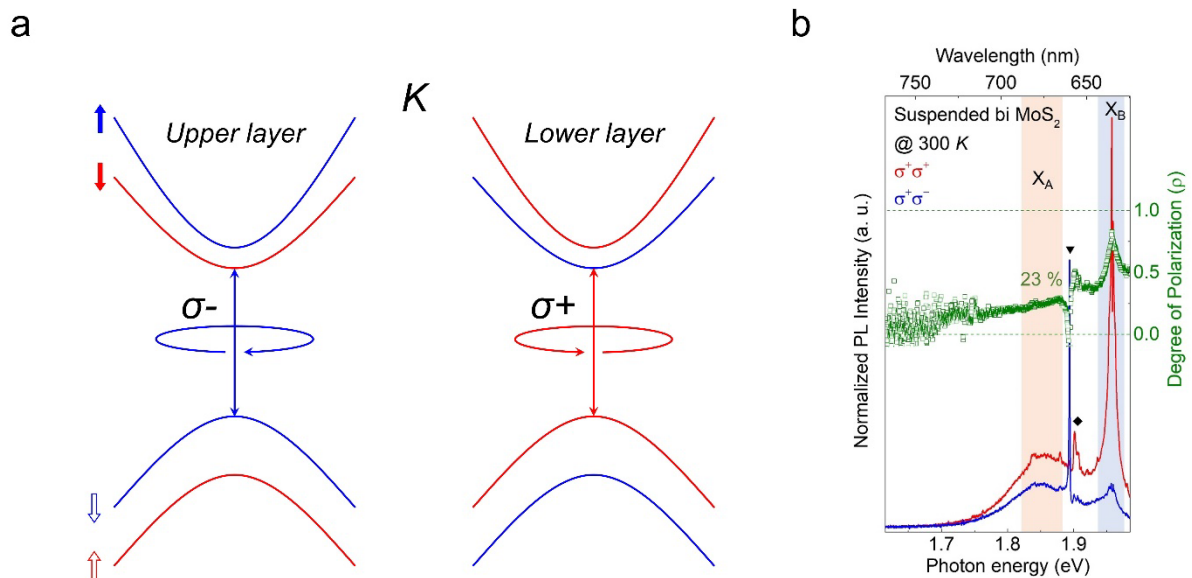


Fig. 1. (a) Schematic presentation for the interaction between circularly polarized light and the electronic states in the K/K' valleys of the stratified layers of bilayer MoS₂. (b) The PL spectra and DOP for suspended bilayer MoS₂. The observation of DOP in bilayer MoS₂, free from substrate influence, provides clear evidence that DOP originates from spin-layer coupling.