Spin-valley reconstruction of dipolar interlayer excitons in WSe_2/WS_2 moiré heterobilayers

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Abstract

Two-dimensional semiconducting heterobilayers have emerged as a powerful platform for exploring correlated electronic phenomena by trapping charge carriers and excitons in a periodic moiré potential. In particular, interlayer excitons in WS_2/WSe_2 heterobilayers experience strong on-site Coulomb repulsion, forming interacting bosonic states that give rise to a dipolar exciton series [1] and provide an experimental platform for simulating Bose-Hubbard physics [2]. Despite these advances, the microscopic origin of these dipolar exciton states—especially the spin–valley configuration of the excited dipolar ladder states—remains poorly understood.

Here, we investigate the origin of dipolar ladder exciton peaks in R-stacked WS_2/WSe_2 heterobilayers using polarization-resolved magneto-optical spectroscopy at cryogenic temperature. Power-dependent measurements reveal that, in addition to the single moiré-trapped ground-state interlayer exciton with spin-triplet configuration ($IX1^{(T)}$), two distinct biexciton states (IX2) emerge from bosonic interactions. Polarization-resolved and magnetic-field-dependent spectroscopy further show that these states correspond to interlayer biexciton species formed by combining the ground-state $IX1^{(T)}$ with an additional interlayer exciton arising from transitions involving either the lower or the upper spin-split conduction bands, yielding $IX2^{(T,T)}$ and $IX2^{(T,S)}$, respectively. The observed energy splitting of ~22 meV between $IX2^{(T,T)}$ and $IX2^{(T,S)}$ agrees well with the conduction-band spin splitting in monolayer WS_2 [3]. These results clarify the microscopic origin of previously observed moiré-trapped dipolar exciton series and highlight the role of spin degrees of freedom in bosonic interactions, opening new opportunities for exploring spin-dependent Bose-Hubbard physics in dipolar exciton systems.

References

[1] Park, H., Zhu, J., Wang, X. et al. Dipole ladders with large Hubbard interaction in a moiré exciton lattice. Nat. Phys. 19, 1286-1292 (2023).
[2] Lian, Z., Meng, Y., Ma, L. et al. Valley-polarized excitonic Mott insulator in WS2/WSe2 moiré superlattice. Nat. Phys. 20, 34–39 (2024).
[3] Kośmider, et al.. Large spin splitting in the conduction band of transition metal dichalcogenide monolayers. Phys. Rev. B 88, 245436 (2013).

Figures

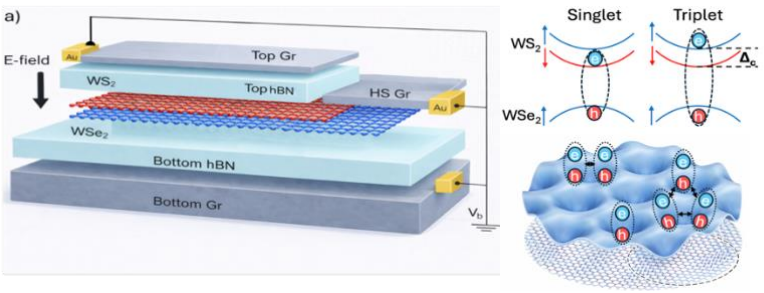

Figure 1: (a) Schematic of the dual-gated hBN-encapsulated R-stack WS_2/WS_2 heterobilayer device. (b) Spin-resolved band alignment showing triplet and singlet interlayer excitons. (c) Moiré potential landscape with localized interlayer excitons.