Role of the *Direct to Indirect* Bandgap Crossover in the '*Reverse*' Energy Transfer Process

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Van der Waals (vdW) heterostructures (HSs), composed of layered materials, have demonstrated significant potential for advancing (opto)electronic applications. In the type-II transition metal dichalcogenide (TMDC) HSs, long-range energy transfer (ET) occurs via the dipole-dipole coupling (Förster type). This ET happens from the acceptor to donor materials due to the resonant overlap of the high-lying absorption states [1]. To investigate this, we studied a HS made by the 1L of tungsten disulfide (WS_2) and 1L-5Ls of molybdenum disulfide (MoS_2), with hexagonal boron nitride (hBN) as a charge-blocking interlayer (Fig. a), using photoluminescence (PL), photoluminescence excitation (PLE), time-resolved PL (TRPL) and density functional theory (DFT) calculations. At room temperature, PL enhancement has been observed in the neutral exciton of WS_2 (Fig. c) in the 1L WS_2-hBN-1L MoS_2 (1L HS) and 1L WS_2-hBN-2L MoS_2 (2L HS) regions compared to isolated WS_2 emission. This enhancement confirms an efficient ET from the MoS_2 B excitonic level to the WS_2 A excitonic level. The increase in layers changes the MoS_2 bandgap from direct to indirect, promoting more carrier scattering from the K valley. Consequently, the carrier population decreases and ET becomes less effective. Our results also prove that this reverse ET happens at a faster timescale than the intervalley scattering (K+ ↔ K-). Overall, this work helps to understand the interlayer processes for better development of the (opto)electronic devices.

References

[1] Arka Karmakar et. al., Nano Lett. 12 (2023), 23.

Figures

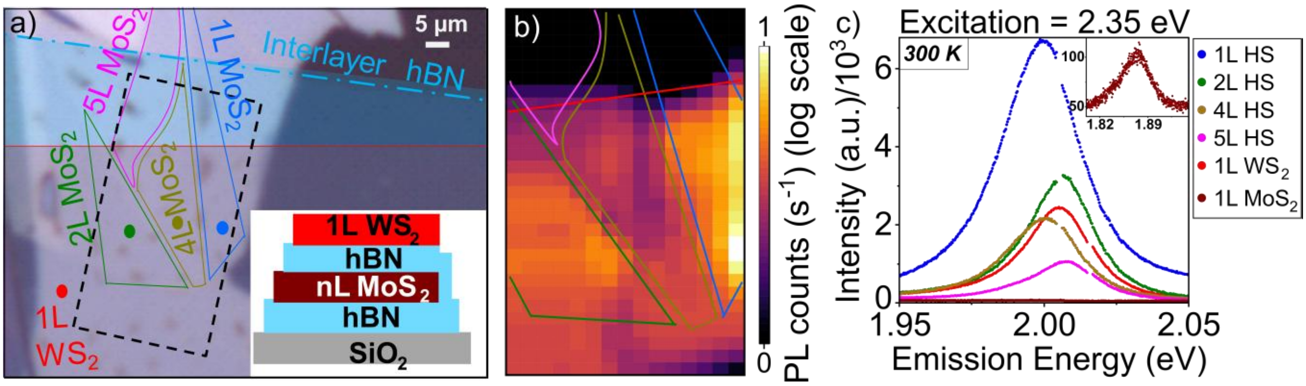

Figure : a) Optical image of the sample with cross-sectional schematics in which different areas of the sample are depicted in various colours. The black box indicates the region selected for PL mapping. b) PL intensity mapping of the black box region at 300 K with excitation energy 2.35 eV, shows the change of enhancement due to the ET process in the multilayer regions of HS. c) PL spectra of all HS regions measured at room temperature, with the inset showing the PL spectrum from the 1L MoS_2 region.