Interlayer Energy Transfer in Type-II 2D Heterostructure

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Two-dimensional (2D) transition metal dichalcogenides (TMDs) are believed to be the fundamental building blocks of next generation optoelectronic device applications [1]. Interlayer charge (CT) and energy transfer (ET) are the two main photocarrier relaxation pathways in the vertical TMD heterostructures (HSs). The interlayer CT process can be easily blocked by placing a thin dielectric medium between the 2D layers [2]. Whereas, the interlayer ET process can survive up to several tens of nm [3]. That's why understanding the interlayer ET process is necessary for designing the TMD HS-based optoelectronic applications. In this talk, I'll present our recent results [4,5] on the effect of resonance overlaps between the *optical bandgaps* and *high-lying* excitonic states in TMD HSs in the interlayer ET processes.

Resonant overlaps between the optical bandgaps of monolayers (1Ls) of molybdenum diselenide ($MoSe_2$) and rhenium disulfide (ReS_2) led us to observe an unprecedented interlayer ET process which dominates over the traditional fast interlayer CT process in type-II HSs [4]. Without a charge blocking medium, namely the hexagonal boron nitride (hBN), we observe ~3.6 times enhanced $MoSe_2$ photoluminescence (PL) emission from the HS area as compared to the 1L area alone. We report that this observed $MoSe_2$ HS PL enhancement is due to the nonradiative ET from the ReS_2 to $MoSe_2$ layer. After introducing different thicknesses of interlayer hBN, we observed more than one order of magnitude higher $MoSe_2$ PL emission at a separation of ~13.5 nm distance. The distance dependence data showed that the ET follows the $1/d^2$ dependency, proving a 2D-to-2D coupling dominated in the HS.

1Ls of molybdenum disulfide (MoS_2) and tungsten diselenide (WSe_2) have overlaps between the high-lying excitonic states. These resonant overlaps allowed us to observe an unconventional ET from the WSe_2 B excitonic level to MoS_2, resulting ~1.9 times enhanced MoS_2 emission from the HS area [5] at the cryogenic temperature (8 K) with hBN as an interlayer medium. With increasing temperature this PL enhancement starts to decrease and at 100 K it completely vanishes due to the increased electron-phonon scattering. This work [5] showed that an interlayer ET process from the *lower-to-higher* bandgap material happens at a rate faster than the intervalley scattering but slower than the intravalley scattering purely due to the resonant overlap between the B excitonic levels in two materials. These works open a new door to study and understand the interlayer ET processes at ultrafast timescale to create a comprehensive understanding for developing the next generation optoelectronic devices.

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