Twisted $MoSe_2$ Homobilayer Behaving as a *Heterobilayer*

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Heterostructures (HSs) formed by artificially twisted transition metal dichalcogenide (TMDC) monolayers (1Ls) have shown great prospect in modulating electronic and optical properties. In semiconducting HSs, the interlayer charge (CT) and energy transfer (ET) processes control the carrier relaxation pathways. Recently, researchers have studied the twist angle dependency in the CT process. However, a similar understanding about the ET process remains elusive. In this work, we show an unusual ET coupling between the twisted molybdenum diselenide ($MoSe_2$) homobilayer formed by the unconventional method of combining the chemical vapor deposition (CVD) and mechanical exfoliation (Exf.) techniques. This ET coupling results in ~8× photoluminescence (PL) enhancement from the HS area at room temperature (300 K). Using a series of optical and electron spectroscopy techniques at 5 K and 300 K, complementing by the density functional theory (DFT) calculations, we show that the large twist angle (57°) and a lattice parameter mismatch of ~1.2% between the two layers effectively suppress the interlayer CT process. Which, allows the ET process to take over the carrier relaxation channels. Thus, the electronically decoupled homobilayer behaves as a *heterobilayer*, i.e., each layer works independently. This work lays the foundation in understanding the complex ET process in a twisted TMDC HS, which is necessary for next generation opto/electronic device applications.