Exciton Formation in Transition Metal Dichalcogenides Monolayers

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Robust excitons dominate the optical properties of atomically thin semiconductors based on transition-metal dichalcogenides (TMDs). The use of fully encapsulated hexagonal boron nitride (hBN) and charge-tunable (CT) TMD monolayers (MLs) allow the electrostatic doping of the MLs, thereby substantially improving the control of exciton complexes. Exciton relaxation and formation dynamics in TMDs have been extensively studied by time-resolved optical spectroscopies.[1] Nonetheless, a crucial question persists: What is the exciton formation mechanism, and how does this process occur in two-dimensional semiconductor systems? This study addresses this fundamental problem through polarization-dependent micro-photoluminescence (PL) studies performed at cryogenic temperatures (4K) on fully hBN-encapsulated and CT TMD monolayers close to the neutrality point. The results of our experiments performed on both WSe_2 and MoS_2 MLs clarify the role played by the two potential formation mechanisms: a) geminate and b) bimolecular (figure 1). The geminate exciton formation process corresponds to the monomolecular annihilation of the photogenerated correlated electron-hole pair. In contrast the non-geminate formation results from the random bimolecular binding of two free charges, losing all correlation between the excitation photon and the electron-hole pair of the exciton.[2]
For a laser excitation energy below the band gap, we show that the geminate mechanism prevails as expected, whereas above the band gap, both geminate and bimolecular phenomena coexist. These results bring precious information on the exciton formation mechanism in 2D semiconductors, which is crucial for the optoelectronic applications of these materials. Additional striking phenomena will be further discussed.

References

[1] Trovatello et al. Nat Commun 11, 5277 (2020)
[2] P. W. M. Blom et al. Phys. Rev. Lett. 71, 3878 (1993)

Figures

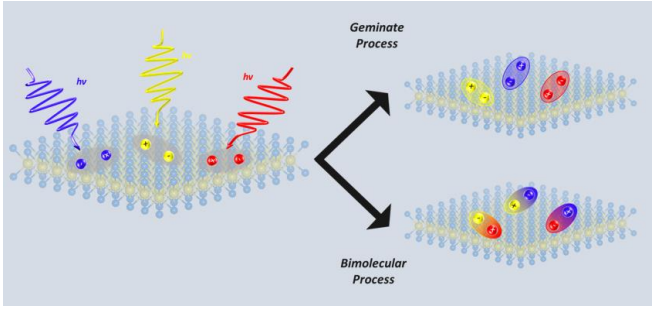

Figure 1: The two characteristic exciton formation processes in TMD monolayers: a) Geminate and b) Bimolecular.