

Montag, 24.06.2024 um 15:15 Uhr
R87, Wilhelm-Klemm-Str. 10

Exciton phenomena in atomically-thin semiconductors

Dr. Roberto Rosati

Department of Physics
Philipps-Universität Marburg

Atomically thin two-dimensional (2D) semiconductors such as the transition metal dichalcogenides (TMD) monolayers have been recently a very active research field. Their reduced dimensionality and weak screening lead to a remarkably strong Coulomb interaction, with tightly bound excitons stable even at room temperature. The excitons in TMDs form a rich landscape, including bright, momentum-dark, and spin-dark excitons, with the interplay between different excitonic species dominating the overall response. These properties make TMDs an ideal platform for exploring exciton optics, dynamics and transport phenomena. Here we study the excitonic transport combining a fully microscopic and material-specific many-particle theory with experiments in different TMDs nanodevices as well as related heterostructures.

After being optically excited, the bright excitons scatter with phonons to form hot dark excitons [1,2], whose excess energy allows peculiar transient optics and transport, including fast and even negative diffusion [3]. The energy of excitons changes in a valley-dependent way with strain, which allows to control the exciton diffusion [4] and drift, even inducing the anti-funneling driven by dark excitons [5], as well as to fingerprint spectral features and their valley [6]. Stacking two monolayers into vertical heterostructures allows interlayer excitons, whose electron and hole are spatially separated in two different monolayers. The resulting spatial dipole allows nonlinear fast anomalous diffusion via dipole-dipole repulsion, even controllable electrically for hybrid excitons, whose interlayer component can be increased by electric fields [7].

Two monolayers grown in the same plan can bind covalently to form lateral heterostructures [8]. Here we investigate the many-particle processes behind the formation of charge-transfer (CT) excitons, which are spatially-separated analogously to interlayer excitons but so far with a much more elusive optical observation. We microscopically demonstrate how CT excitons can be tuned via interface- and dielectric engineering, i.e. band offset, junction width and dielectric environment [8]. Investigating the photoluminescence spectra after excitation at the interface of a hBN-encapsulated lateral MoSe₂-WSe₂ heterostructure, we predict the appearance of a low-energy peak emitted from CT excitons, cf. the purple box in Fig. 1(a) [8]. This occurs for junction widths w smaller than the Coulomb-induced Bohr radius (cf. thick blue and thin orange line in Fig. 1(a)) and for large-enough dielectric constant of the environment. The theoretical prediction is compared with experimental low-temperature photoluminescence measurements showing emission in the bound CT excitons energy range, cf. the purple box in Fig 1(b) [8]. Overall, our work presents a significant step towards a microscopic understanding of optical properties of technologically promising 2D materials.

References

- [1] Wallauer et al. *Nano Lett.* **21**, 5867 (2021)
 [2] D. Schmitt et al., *Nature* **608** (2022) 499
 [3] R. Rosati, et al., *Nanoscale* **13**, 19966 (2021)
 [4] R. Rosati, R. Schmidt et al., *2D Mater.* **8**, 015030 (2021)
 [5] R. Rosati, R. Schmidt et al., *Nat. Commun.* **12**, 7221 (2021)
 [6] A. Kumar, D. Yagodkin, et al., preprint at *arXiv:2312.07332* (2023)
 [7] F. Tagarelli et al., *Nat. Photonics* **17**, 615 (2023)
 [8] R. Rosati et al., *Nat. Commun.* **14**, 2438 (2023)

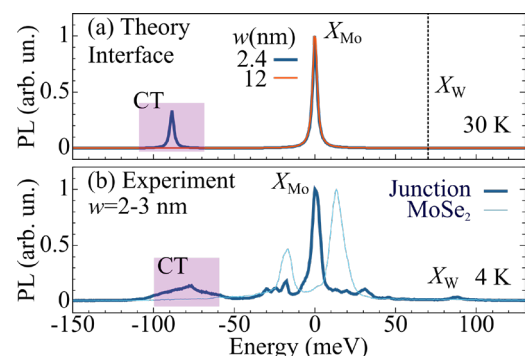


Fig.1. Theoretical (a) and experimental (b) photoluminescence spectrum (PL) after excitation at the interface of a hBN-encapsulated lateral MoSe₂-WSe₂ heterostructures, showing a low-energy feature attributed to CT excitons (purple box). Larger junction width w (thin orange in (a)) or excitation on the MoSe₂ side (thin cyan in (b)) do not give rise to CT excitons.