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Energy transfer mechanisms in stacks composed of a monolayer transition metal dichalcogenide and a Ruddlesden-Popper 2D perovskite

Mechanizmy przekazu energii w heterostrukturach złożonych z monowarstwy dichalkogenku metalu przejściowego oraz dwuwymiarowego perowskitu Ruddlesden-Popper

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For many decades, heterostructures (HS) have been used to build devices with desired properties. Initially, HS were grown by epitaxial techniques, which required that the lattice constants of the constituent materials have similar values. This severely limited the number of materials that could be used together. Following the discovery of graphene in 2004, a new type of HS composed of van der Waals materials has emerged. Offering a remarkable degree of flexibility due to the relaxed lattice constant matching requirement, van der Waals stacks have become excellent candidates for a new generation of optoelectronic devices.

One example of a phenomenon important for future applications is the excitation transfer between the layers of the constituent materials in the HS. This can occur via two main pathways: charge transfer (CT) or energy transfer (ET). In this thesis, I compare the results obtained in the optical spectroscopy experiments and theoretical calculations toward understanding the mechanism of excitation transfer in HS composed of two different van der Waals materials - a 2D hybrid organic-inorganic perovskite and a monolayer transition metal dichalcogenide (TMD). The investigated stacks include: PEA_2PbI_4/WS_2, where PEA stands for phenylethylammonium ($PEPI/WS_2$), $PEA_2PbI_4/MoSe_2$ ($PEPI/MoSe_2$), and $BA_2PbI_4/MoSe_2$, where BA is butylammonium ($BAPI/MoSe_2$).

The footprints of the excitation transfer in the investigated stacks were manifested in the photoluminescence (PL), reflectivity, PL excitation and time-resolved PL measurements. All the experiments were conducted at a temperature of 5 K. The CT was evidenced by monitoring the excess carrier concentration by estimating the charged-to-neutral exciton PL intensity ratio in TMD. The signatures of the ET were observed as resonances in the PL excitation spectra in TMD. According to the density functional theory calculations performed by a collaborating group from the University of Dresden, all three HS exhibit a particular type II band alignment, where the hole transfer between perovskite and TMD is allowed, while the electron transfer is blocked. Based on the comparison of the results obtained in the experiments and DFT calculations, I concluded that in $PEPI/WS_2$ and $BAPI/MoSe_2$ stacks, both CT and ET occur, while $PEPI/MoSe_2$ stack exhibited signatures of only CT.

The obtained results have shown that the specific excitation transfer pathway in 2D perovskite/TMD stacks can be closed or opened by simply using different organic spacer compounds in a perovskite material. The easy manipulation of the excitation transfer holds great promise for future applications of these HS in novel, ultrathin electronics.

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