Electronic Supplementary Material (ESI) for Nanoscale.

WS_2 Monolayer Based Light Emitting Devices in a Vertical p-i-n Architecture

Dominik Andrzejewski, Eric Hopmann, Michèle John, Tilmar Kümmell, Gerd Bacher

Supporting Information

1. Photoluminescence dependence on adjacent layers

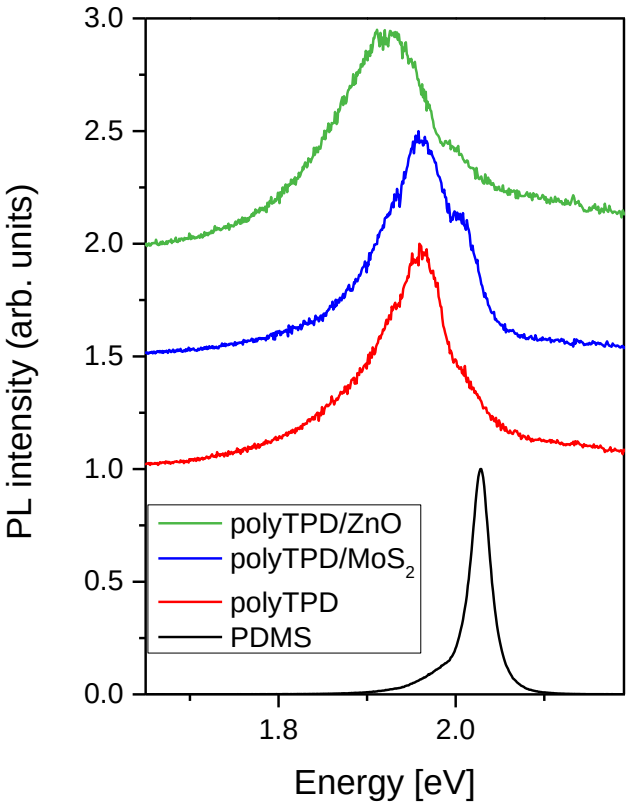

Figure S1. Change of PL emission during the process chain, showing the influence of the adjacent layers. Black: WS_2 on PDMS, Red: WS_2 on poly-TPD, Blue: WS_2 between poly-TPD and MoS_2 multilayer, Green: WS_2 between poly-TPD and ZnO-NP layer.

The spectra are normalized and vertically shifted for clarity.

The PL analysis of the active WS_2 monolayer at different stages of the process chain indicates the influence of the different surrounding on the luminescence spectrum. The exfoliated flake on PDMS (black) exhibits luminescence dominated by the A exciton with a maximum at 2.015 eV. In contrast, after WS_2 transfer onto TPD (red) we see two lines with maxima at 2 eV and 1.95 eV

that we attribute to the A-exciton, slightly shifted in energy possibly by the different dielectric surrounding, and the negatively charged X^-, which is shifted by another 50 meV towards lower energy. While the MoS_2-ESL on top of the WS_2 layer (blue) does not change the PL significantly with respect to the PL on poly-TPD, the PL in the sample with ZnO as ESL is clearly different: The PL maximum is here at 1.91 eV and the emission is significantly broadened to 130 meV. Possible reasons could be bound excitons that are enhanced by the ZnO/WS_2 interface, an enhanced contribution of trions, an energy shift due to the Quantum Confined Stark effect or a combination thereof.

2. Dependence of the EL signal on the MoS_2 thickness in the type II design.

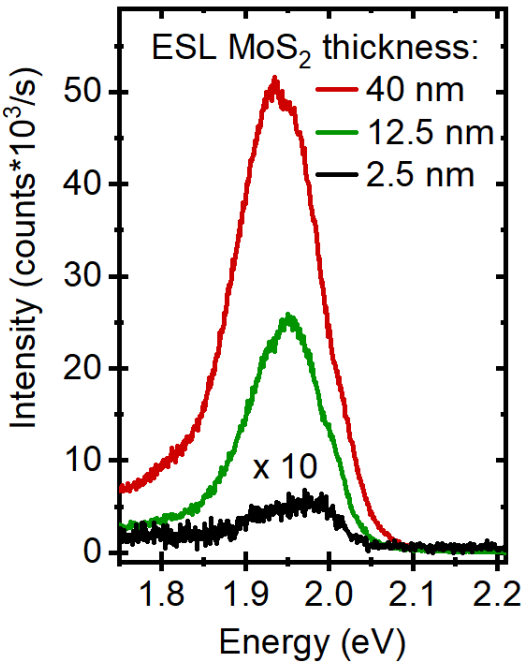

Figure S2. Electroluminescence spectra of WS_2 p-i-n devices with MoS_2 ESL layers of different thickness at 5.5 V. The intensity increases with increasing number of MoS_2 layers with a simultaneous peak shift to lower energies.

We studied the influence of the MoS_2-based electron supporting layer thickness on the electroluminescence of type II devices. As a striking result, only very weak EL is found for thin (2.5 nm) MoS_2-ESL. Obviously, the influence of the contacts is leading to a hole transfer to the contact and thus to an EL quenching [Ref. S1]. With increasing thickness, the electroluminescence becomes stronger, but with a distinct redshift. Here, we assume hole

transfer from WS_2 to the adjacent layers (poly-TPD and MoS_2), enhancing the probability for negatively charged excitons (trions) in the WS_2.

3. Estimation of internal quantum efficiency of the WS_2 monolayers

The collection efficiency of our setup was estimated by introducing a reflecting substrate (gold layer with reflectance of 80 % at a wavelength of 532 nm) and measuring the detected counts in the setup for a defined laser power.

The detected photon number per time can be written generally as

$$P_{det}/h\nu_{det} = (P_{exc}/h\nu_{exc}) \cdot IQE \cdot A \cdot \beta$$

Here $P_{det}/h\nu_{det}$ and $P_{exc}/h\nu_{exc}$ represent the photons per time detected in the setup and exciting the sample, respectively, with $P_{det}(P_{exc})$ as detected (exciting) power. A is the absorbance of the sample at the excitation wavelength, IQE the internal quantum efficiency in the sample and β the collection efficiency of the setup. In case of the reference with 80% reflection, IQE*A is set to 0.8 and β can be determined from measuring the counts in the detector and the power of the exciting laser at the position of the sample. From this, we determine a collection efficiency $\beta = 10^{-3}$. When characterizing the WS_2 monolayers by PL, the IQE can be calculated from the laser power, the detected intensity and the absorption in the monolayer that we assume to be 2% at the excitation wavelength of λ=532 nm [Ref. S2].

References

S1 U. Bhanu, M. R. Islam, L. Tetard and S. I. Khondaker, Scientific Reports **4**, 5575 (2014).

S2 H. Hill, A.Rigosi, C. Roquelet, A. Chernikov, T. Berkelbach, D. Reichman, M. Hybertsen, L. Brus and T. Heinz, Nano letters **15,** 2992 (2015)