

Correlation of Atomic Structure and Luminescence of Two-dimensional MoSe₂/WSe₂ In-plane Nanodot Heterostructures

Saiphaneendra Bachu¹, Steffi Y. Woo², Benjamin Huet^{1,3}, Nicholas Trainor¹, Danielle Reifsnnyder Hickey^{1,4}, Joan M. Redwing^{1,3}, Mathieu Kociak², Luiz H.G. Tizei^{2,*}, Nasim Alem^{1,3,*}

¹Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA, USA

²Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS-UMR 8502, Orsay, , France

³2D Crystal Consortium (2DCC), Materials Research Institute (MRI), The Pennsylvania State University, University Park, PA, USA

⁴Department of Chemistry, The Pennsylvania State University, University Park, PA, USA

*Corresponding authors: luiz.galvao-tizei@universite-paris-saclay.fr, nua10@psu.edu

Spatially confined material systems such as nanowires, quantum wells and thin film superlattices are widely studied for their unique light emission properties, making them attractive candidates for emerging optoelectronic applications such as photodetectors, light emitting diodes, lasers, etc. [1]. Two-dimensional (2D) transitional metal dichalcogenides (TMDs) are an excellent choice to add to the growing list of spatially confined material systems because, in the form of monolayers, they become direct band gap semiconductors displaying high binding energy excitons, which makes them excellent light emitters [2]. These materials can be further engineered to get localized light emission by means of point defects, folds, and Moiré superlattice formation by preparing vertical heterostructures [3]. A novel method to possibly achieve localized emission from TMDs is to form in-plane monolayer TMD heterostructures wherein one of the phases is spatially confined resulting in nanodot/matrix type interfaces. Price *et al.* have recently proposed that it is possible to achieve quantum confinement from such structures by carefully tuning the size and shape of the TMD nanodots which could lead to localized light emission and potentially single photon emission (SPE) [4]. However, this has not been experimentally realized yet.

In this work, we study the MoSe₂/WSe₂ (nanodot/matrix) in-plane heterostructures synthesized on *c*-plane sapphire substrates using metal organic chemical vapor deposition (MOCVD) process. As-grown heterostructures are then encapsulated with hBN before being transferred onto Quantifoil Au TEM grids using the method outlined in the ref. [5]. The encapsulated hBN-MoSe₂/WSe₂-hBN stacks are then examined using scanning/transmission electron microscopy (S/TEM) imaging and cathodoluminescence (CL) spectroscopy to correlate the atomic structure of heterostructures and the light emission properties. Here, hBN encapsulation aides in reducing the roughness of TMD layer, thus reducing the excitonic line widths [5]. It also permits the creation of a larger number of electron-hole pairs that leads to enhanced TMD light emission in CL. CL spectroscopy performed in the STEM mode benefits from the high spatial resolution so it is possible to investigate the effect of nanoscale structural heterogeneities on the light emission properties of the heterostructure [6]. This is in contrast to conventional optical measurements which provide averaged results over large areas from such structures. The CL experiments are performed at low temperature (~ 150 K) using a modified Nion HERMES-S200 (ChromaTEM) microscope operated at 60 kV and fitted with an Attolight Mönch CL system.

Figure 1a shows an annular dark field-STEM (ADF-STEM) image of hBN-MoSe₂/WSe₂-hBN stack transferred on a Quantifoil Au TEM grid. Owing to the atomic number dependent contrast of ADF-STEM images, MoSe₂ nanodots (red arrows) appear darker in comparison to WSe₂ matrix (blue arrow) [6]. Furthermore, we see negligible contrast from hBN for the same reason. Next, a summed CL spectrum obtained from the dashed yellow box in Figure 1a displays two emission peaks (Figure 1b). While the peak at ~720 nm corresponds to WSe₂, the peak at ~800 nm could be attributed to MoSe₂ [7]. The relatively high intensity of the MoSe₂ peak could indicate the nanodots are more luminescent than the matrix. The presence of MoSe₂ nanodots is further confirmed using

STEM-energy dispersive spectroscopy (STEM-EDS) mapping, as presented in Figure 2. Figure 2c and 2d clearly show the spatial separation of Mo and W signals, whereas Se is present everywhere in Figure 2b. STEM-EDS maps are acquired from an unencapsulated heterostructure sample. The presentation will further explore the atomic structure at the heterostructure interfaces, and elucidate the effects of size and shape of dots, interface quality and presence of defects on the emission characteristics of heterostructures [8].

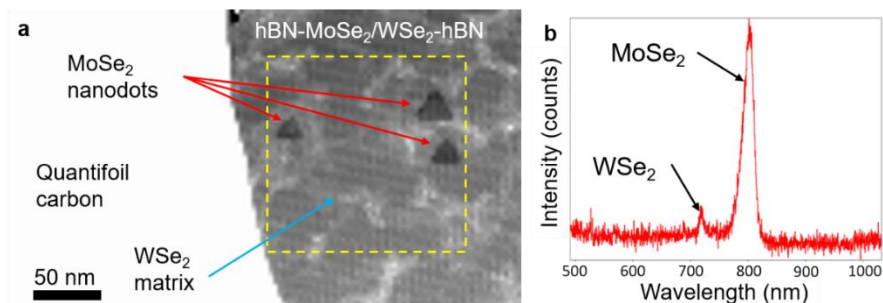


Figure 1 (a) ADF-STEM image of hBN-MoSe₂/WSe₂-hBN stack suspended over holey carbon Quantifoil Au TEM grid showing the triangular MoSe₂ nanodots embedded in WSe₂ matrix forming the MoSe₂/WSe₂ in-plane heterostructures, and (b) summed CL spectrum obtained from the dashed yellow box in (a) showing characteristic light emission peaks of MoSe₂ nanodots and WSe₂ matrix.

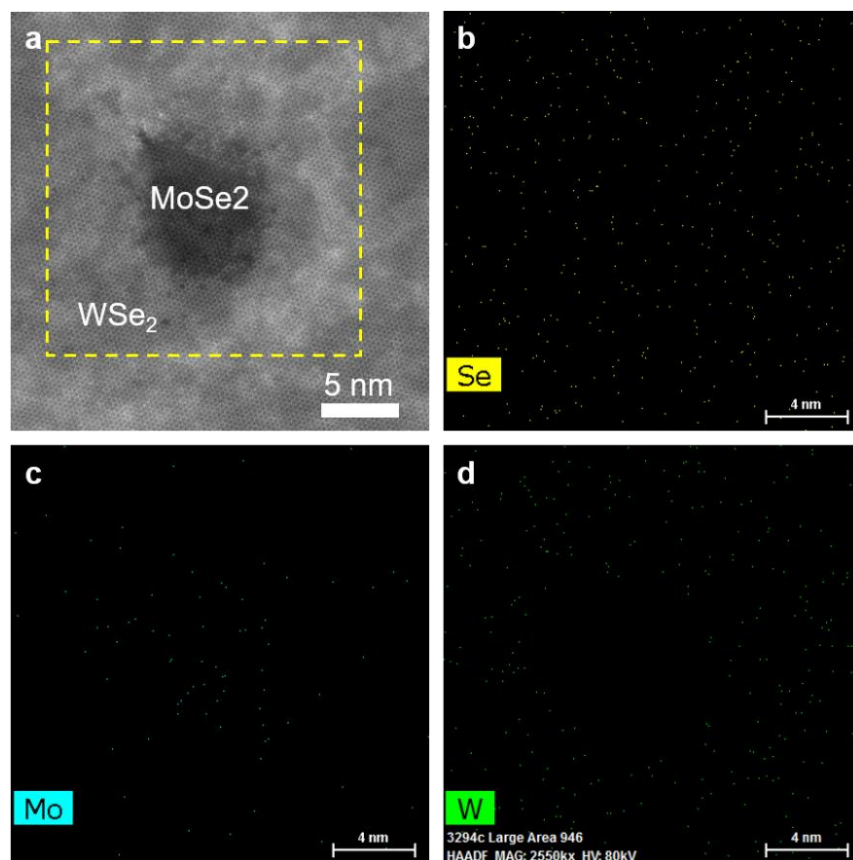


Figure 2 (a) Atomic resolution ADF-STEM image of unencapsulated MoSe₂/WSe₂ in-plane heterostructure and (b-d) elemental STEM-EDS maps obtained from the dashed yellow box in (a) clearly confirming the spatial separation of MoSe₂ nanodot and WSe₂ matrix with little intermixing.

References:

- [1] Fox, Mark, and Radu, Ispasoiu, Springer Handbook of Electronic and Photonic Materials (2017) : 1.
- [2] Mak, Kin Fai, et al., Physical Review Letters 105.13 (2010) : 136805.
- [3] Michaelis de Vasconcellos, Steffen, et al., Physica Status Solidi B (2022) : 2100566
- [4] Price, Christopher C., et al., ACS nano 13.7 (2019) : 8303-8311.
- [5] Shao, Fuhui, et al., arXiv preprint arXiv :2202.04483 (2022).
- [6] Bonnet, Noémie, et al., Nano Letters 21.24 (2021) : 10178-10185.
- [7] Tongay, Sefaattin, et al., Applied Physics Letters 104.1 (2014): 012101.
- [8] This work was supported by the National Science Foundation (NSF), in part under the CAREER program (DMR-1654107) and in part by the Penn State 2DCC-Materials Innovation Platform (2DCC-MIP) under NSF cooperative agreement DMR-1539916. This project has also been funded in part by the National Agency for Research under the program of future investment TEMPOS-CHROMATEM (reference no. ANR-10-EQPX-50), the JCJC grant SpinE (reference no. ANR-20-CE42-0020), and by the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101017720 (EBEAM)