

Supplementary Information : Exciton spectroscopy and unidirectional transport in MoSe₂-WSe₂ lateral heterostructures encapsulated in hexagonal boron nitride

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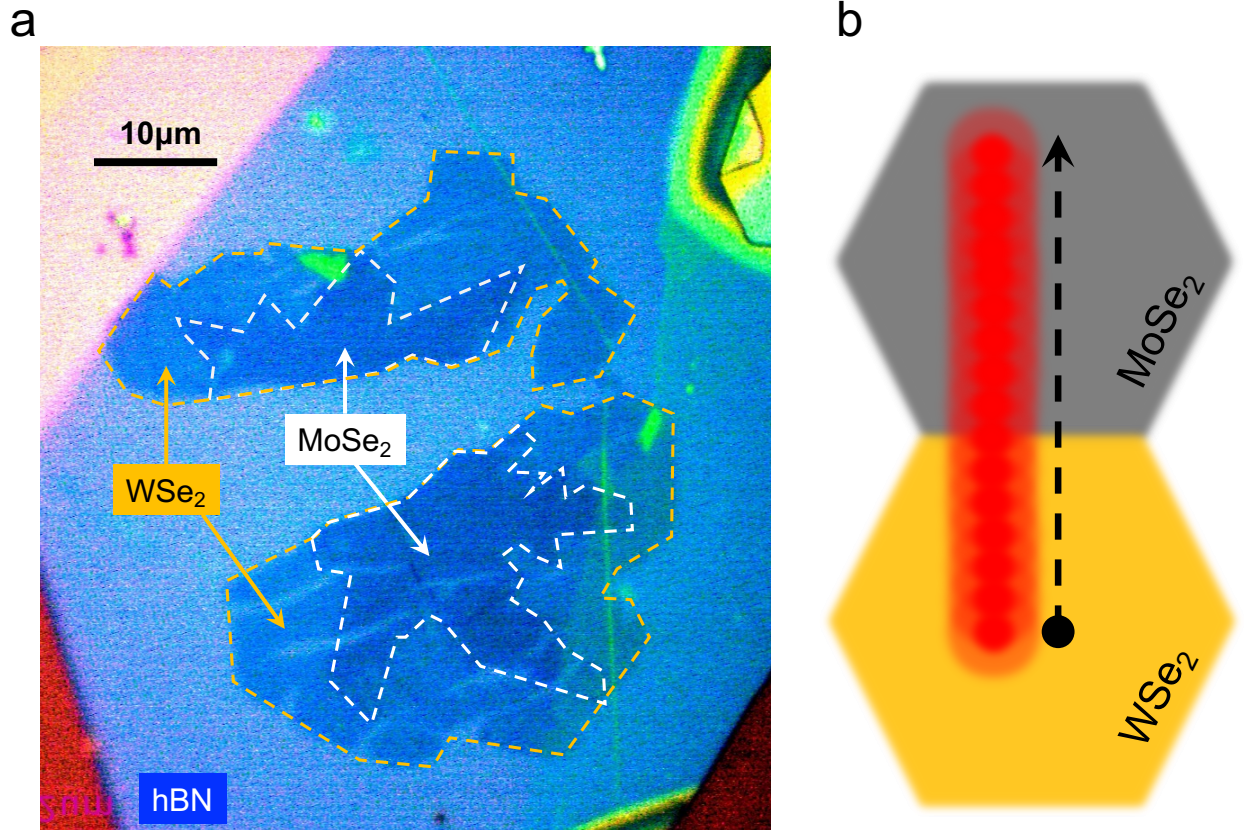
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Supplementary Note 1. Optical microscope image and linescan description

An optical microscope image of the MoSe₂/WSe₂ lateral heterostructure (LH) and a schematic representation of the linescan process used in the optical spectroscopy experiments are shown in Fig. 1a,b. We use water-assisted deterministic transfer to pick up as-grown, chemical vapor deposition (CVD) LHs using polydimethylsiloxane (PDMS) and deterministically transfer and encapsulate them in hBN [1, 2]. The optical contrast between MoSe₂ and WSe₂ is different, allowing the direct visualization of the heterojunctions, shown with white dashed lines in Fig. 1a. In Fig. 1b we show a schematic representation of the process for the Raman, PL and Reflectivity linescans (Figure1 in the main text). We position the excitation laser ($\lambda = 633$ nm, diffraction-limited spot diameter of $1 \mu\text{m}$) on the area of WSe₂ and we use piezo-controlled nanopositioners to move the sample with steps of ≈ 150 nm. After each step, a single -Raman, photoluminescence, reflectivity- spectrum is collected and finally all spectra are plotted as contour representations in Figure1 of the main text.

Supplementary Note 2. Optical spectroscopy

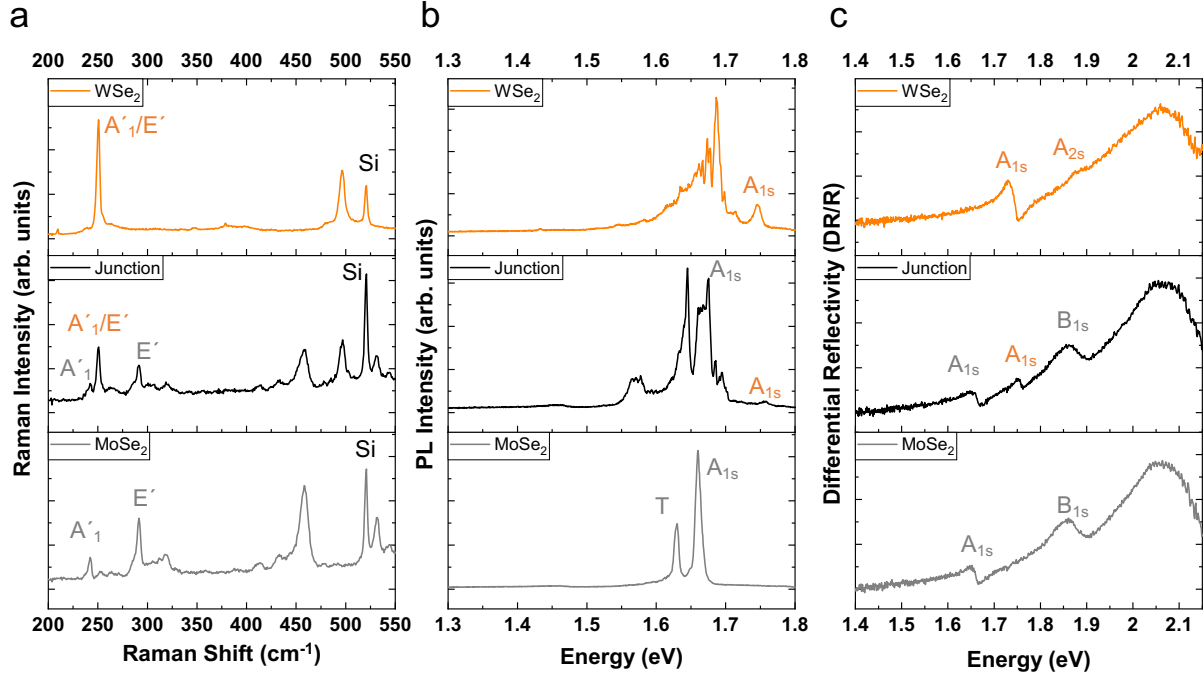
Raman, PL and differential white light reflectivity spectra are collected at T=5 K in a closed-loop liquid helium (LHe) system. Fig. 2a,b,c shows individual Raman, PL and reflectivity spectra of MoSe₂ (bottom), junction (middle) and WSe₂ (top), after transfer and encapsulation in hBN. For the Raman and PL experiments we use a 633 nm HeNe laser as an excitation source with a spot size diameter of $\approx 1 \mu\text{m}$ and $6 \mu\text{W}$ power. In reflectivity we use a tungsten-halogen white light source with a power of a few nW to collect the intensity reflection coefficient of the sample with the monolayer (R_{ML}) and the reflection coefficient of the substrate (R_S) so that $\Delta R = (R_{ML} - R_S)/R_S$. The spectral shape and



Supplementary Figure 1. (a) Optical image of the hBN-encapsulated LH. Orange and white dashed lines indicate the boundaries of WSe₂ and MoSe₂, respectively. (b) Schematic representation of the Raman, PL and Reflectivity linescan process across the heterojunction of MoSe₂/WSe₂ LH. The laser is initially positioned on WSe₂ and a series of sequential steps (step size ≈ 150 nm) follows with a single signal acquisition after each step.

amplitude of the differential reflectivity signal also depends on thin-film interference effects related to the thickness of top, bottom hBN and SiO₂ layers [3]

The main Raman peaks of MoSe₂ (Fig. 2a) include the A₁'(Γ) phonon at 241 cm⁻¹ and the E'(Γ) at 291 cm⁻¹ [4], while a strong peak at 458 cm⁻¹ has been associated two other peaks to form triplets, see supplementary material of [5]. A strong peak at 531 cm⁻¹ was recently assigned to multi-phonon processes either associated with both K and M point phonons or combinations of Γ point phonons [5]. The observation of this phonon is a signature of resonant excitation with an excited exciton state. WSe₂ phonons are spectrally different compared to MoSe₂. The degenerate A₁'(Γ)/E'(Γ) phonons are located at 250 cm⁻¹ and, similar to MoSe₂, a strong and recently discovered peak at 495 cm⁻¹ is also observed here



Supplementary Figure 2. (a) Raman, (b) photoluminescence and (c) reflectivity spectra of MoSe₂ (bottom), on the junction (middle) and WSe₂ (top).

and attributed to multi-phonon processes at K and M points or combination of Γ point phonons [5]. As expected, when the laser spot is on the junction, Raman spectra display a superposition of the individual spectral signatures. We did not identify any measurable shift or broadening of the Raman peaks when we scanned across the junction. PL spectra of MoSe₂ and WSe₂ monolayers exhibit pronounced peaks (Fig. 2b), associated with neutral (X^0) and charged (X^T) excitons. Additional peaks appearing at lower energies, especially in WSe₂ and at the junction, possibly due to an ensemble of localized emission from defects. Further experiments are necessary to examine any possibility of the formation of interlayer excitons. Strong excitonic resonances appear in reflectivity for both materials with negligible Stokes shift between emission and absorption of the A_{1s} exciton state (see Fig. 2c). Clear signatures of B_{1s} states are also observed in both materials while for WSe₂ the appearance of the A_{2s} excited state further supports the good quality of the CVD-grown monolayers. Similar to Raman and PL spectroscopy, on the junction we observe a superposition of the available states from both materials.

Supplementary Note 3. Diffusion experiment and model

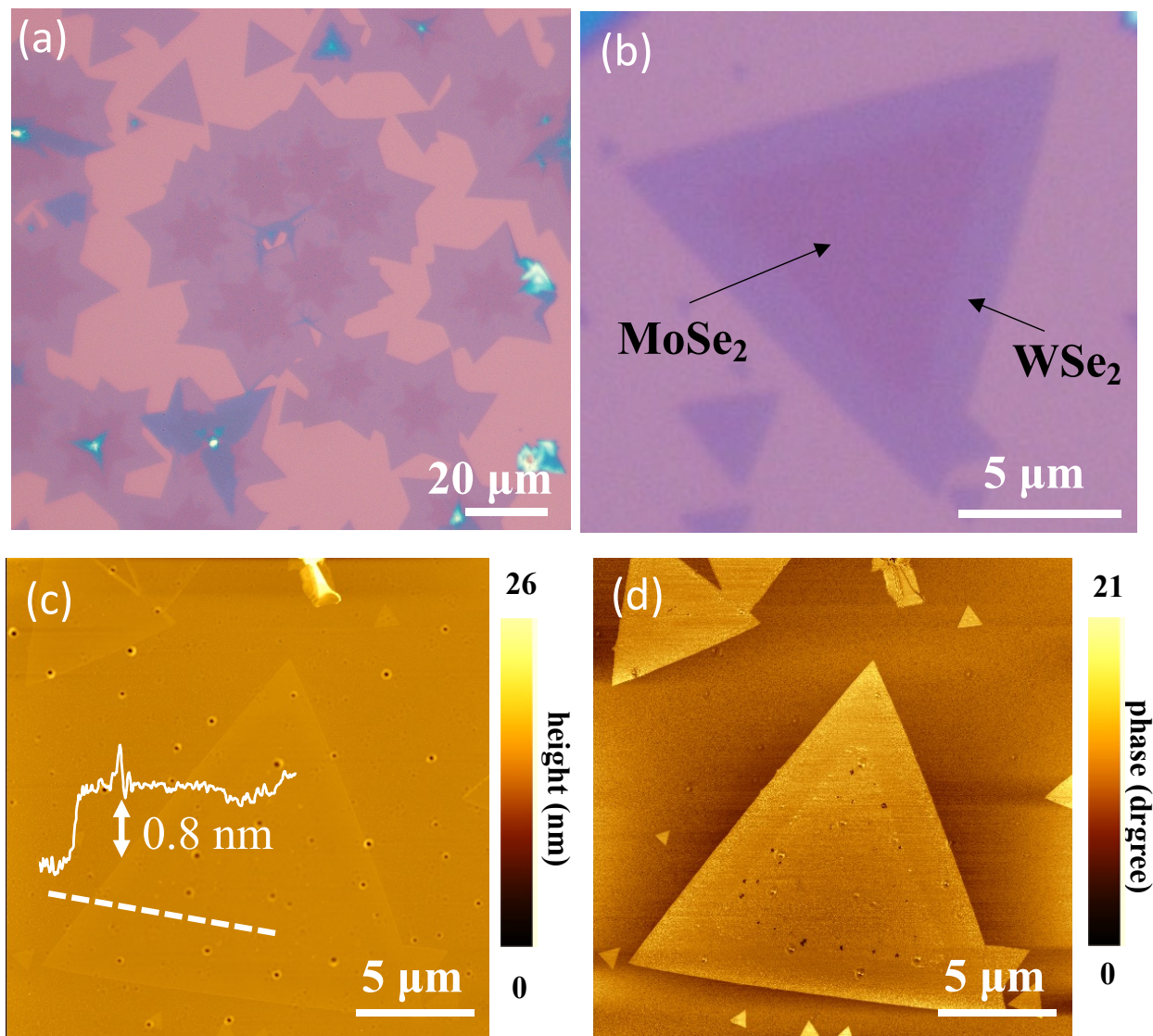
For the investigation of lateral transport, we used an experimental procedure very similar to the one described in Supplementary Note 1. The excitation is based on a HeNe laser with an excitation spot size of about $0.7\mu\text{m}$ and an excitation power of $5\mu\text{W}$. The PL images are recorded by a Hamamatsu Fusion-BT CMOS camera. The PL profiles are plotted in polar coordinates. To do so, we integrate over all the polar angles $[0;2\pi]$, the data are therefore $I_{PL}(r)$ where r is the radius from the maximum PL intensity.

To model the scanning part over the junction (see Fig.4 in the main text), we numerically solved the classical diffusion equation with the use of the Matlab toolbox *pdetool* (partial differential equation toolbox).

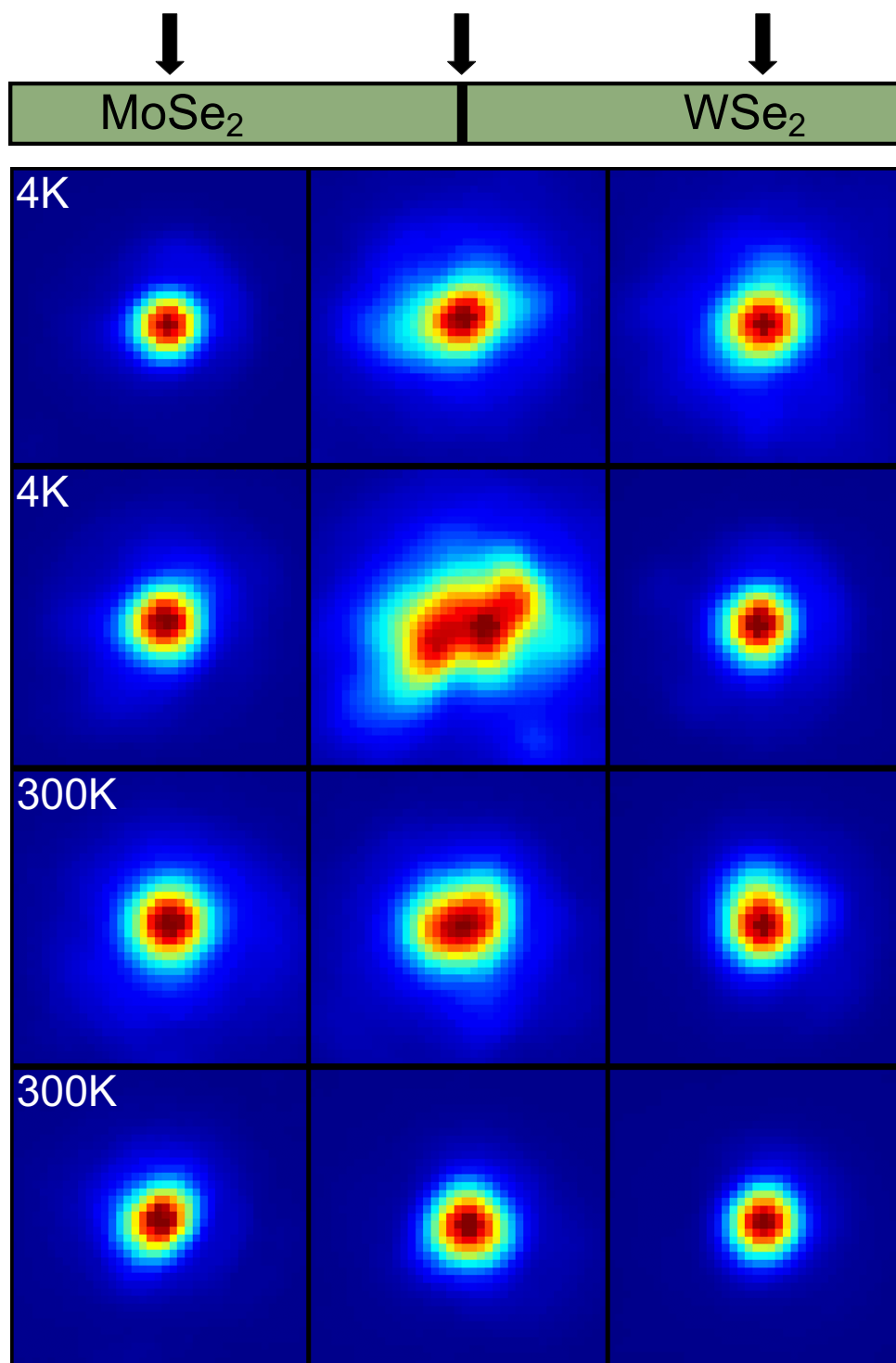
Supplementary Note 4. Electron microscopy

For STEM investigation, the samples were transferred to Quantifoiltm grids using PMMA assisted transfer protocol. The High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image was acquired with a Thermofisher Talos 200X microscope operated at 200 kV.

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Supplementary Figure 3. **Characterization of lateral heterostructures** (a,b) Optical microscopy image of as grown MoSe₂-WSe₂ LHs (c,d) AFM height and phase image of as grown LHs. The height profile in the inset of (c) shows monolayer thickness of 0.8 nm.



Supplementary Figure 4. **More examples of PL imaging** We show more examples of typical PL emission spots in MoSe₂ (left), on the junction between the two materials (middle), and in WSe₂ (right) for two different sample temperatures as marked on the panels for each line.