Exciton spectroscopy and diffusion in MoSe$_2$-WSe$_2$ lateral heterostructures encapsulated in hexagonal boron nitride

Dorian Beret$^{1,+}$, Ioannis Paradisanos$^{1,+}$, Ziyang Gan$^{2,+}$, Emad Najaﬁdehaghani$^2$, Antony George$^{2,3}$, Tibor Lehner$^1$, Johannes Biskupek$^4$, Shivangi Shree$^5$, Ana Estrada-Real$^1$, Delphine Lagarde$^1$, Jean-Marie Poumirol$^6$, Vincent Paillard$^6$, Kenji Watanabe$^7$, Takashi Taniguchi$^8$, Xavier Marie$^1$, Ute Kaiser$^4$, Pierre Renucci$^1$, Laurent Lombez$^1$, Andrey Turchanin$^{2,3,4}$ and Bernhard Urbaszek$^{11}$

$^1$Université de Toulouse, INSA-CNRS-UPS, LPCNO, 135 Avenue Rangueil, 31077 Toulouse, France
$^2$Friedrich Schiller University Jena, Institute of Physical Chemistry, 07743 Jena, Germany
$^3$Abbe Centre of Photonics, 07745 Jena, Germany
$^4$Ulm University, Central Facility of Electron Microscopy, D-89081 Ulm, Germany
$^5$Department of Physics, University of Washington, Seattle, WA, USA.
$^6$CEMES-CNRS, Université de Toulouse, Toulouse, France
$^7$Research Center for Functional Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan
$^8$International Center for Materials Nanoarchitectonics, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

Chemical vapor deposition (CVD) allows lateral edge epitaxy of transition metal dichalcogenide heterostructures with potential applications in optoelectronics. Critical for carrier and exciton transport is the quality of the two materials that constitute the monolayer and the nature of the lateral heterojunction. Important details of the optical properties were inaccessible in as-grown heterostructure samples due to large inhomogeneous broadening of the optical transitions. Here we perform optical spectroscopy at T = 4 K and also at 300 K to access the optical transitions in CVD grown MoSe$_2$-WSe$_2$ lateral heterostructures that are transferred from the growth-substrate and are encapsulated in hBN. Photoluminescence (PL), reflectance contrast and Raman spectroscopy reveal considerably narrowed optical transition linewidth similar to high quality exfoliated monolayers. In high-resolution transmission electron microscopy (HRTEM) we find near-atomically sharp junctions with a typical extent of 3 nm for the covalently bonded MoSe$_2$-WSe$_2$. In PL imaging experiments we find effective excitonic diffusion length that are longer for WSe$_2$ than for MoSe$_2$ at low T=4 K, whereas at 300 K this trend is reversed.

Introduction.— Research on semiconducting transition metal dichalcogenide (TMDs) monolayers [1–3] and their heterostructures is motivated by new collective effects of the electronic system [4–6] and the potential for new optoelectronic and quantum technology devices [7–17]. The optical and electronic properties of these materials can be tuned by combining different monolayers (MLs) via van der Waals stacking to create vertical heterostructures [18–20]. But interestingly, tuning of optical properties of TMDs can also be achieved while staying in the ultimate monolayer limit. Recent progress is based on innovative growth techniques such as for Janus monolayers with different top and bottom chalcogen [21–23] as well as in lateral heterostructures (LHs) [24, 25] within the monolayer plane with an atomically-sharp 1D interface which exhibits p-n junction characteristics [26, 27]. Examples of potential applications for LHs are photodetectors [28], p-n junction diodes [24, 29, 30], photovoltaic [29], electroluminescent [29] and quantum devices [31].

LHs can be fabricated following one-step [24, 25, 27, 30] or multiple-step [32, 33] growth processes either by physical vapor deposition (PVD) or by chemical vapor deposition (CVD). Electron beam lithography has also been used for the fabrication of LHs [34]. One-step CVD approaches with suitable growth conditions are simpler and have the advantage to grow large area TMD LHs at lower temperatures [24]. Accessing the quality of the monolayer heterojunction is so far mainly based on electron microscopy techniques. A depletion width on the order of few nanometers is reported using scanning tunneling microscopy and spectroscopy techniques [35]. The electronic structures and band alignments of TMD LHs have been calculated using density functional theory [36] and the formation of interface excitons is predicted by tight-binding models, as well as effective mass models [37].

To further access carrier dynamics and excitonic properties at the interface optical spectroscopy is needed as a powerful and non-invasive tool. But in as-grown CVD samples details are masked due to the large inhomogeneous broadening reported for the optical transitions. The vast majority of optical characterization experiments performed on LHs are reported at room temperature and a detailed investigation at cryogenic temperatures is not reported so far. At cryogenic temperatures, a precise estimation of the material quality near and across the junction are possible, as thermal broadening effects are reduced and defects can be detected through luminescence of localized states. An important target is investigating the consequences of connecting MoSe$_2$, a material dominated by bright spin-allowed exciton transitions [38, 39], with WSe$_2$ which has a very similar bandgap but shows very rich and
striking emission features based on dark, spin-forbidden exciton complexes [40].

Here we perform low-temperature optical spectroscopy and microscopy experiments in CVD-grown MoSe$_2$-WSe$_2$ monolayer LHs [24]. We first lift the LHs from the growth substrate. This shows that transfer of the lateral heterostructure to other substrates is possible for device processing. Second, we encapsulate the LHs in high quality exfoliated hBN flakes [41] (Fig. 1a). Encapsulation of the TMD monolayer in high quality hexagonal boron nitride (hBN) [41] is crucial to access the intrinsic optical properties of exfoliated and CVD grown flakes [42–50]. We report optical transition linewidth of the LH monolayer ($\approx 5$ meV at $T=4$ K) comparable to high quality exfoliated layers. Our step-by-step scans across the heterojunctions in optical spectroscopy experiments show an abrupt change from WSe$_2$ to MoSe$_2$. In atomic-resolution transmission electron microscopy on our samples we find a transition with a nm-sharp junction from MoSe$_2$ to WSe$_2$. The structural quality is also demonstrated in Raman spectroscopy. Photoluminescence (PL) imaging experiments allow us to investigate excitonic transport governed by the different effective lifetime of the exciton species for MoSe$_2$ and WSe$_2$ at $T = 4$ K and 300 K.

Experimental results.— Our MoSe$_2$-WSe$_2$ lateral monolayer heterojunction is grown by CVD synthesis that we reported recently [24]. A schematic representation of the encapsulated MoSe$_2$-WSe$_2$ structure is shown in Fig. 1a. A HAADF-STEM image recorded at the boundary region of the MoSe$_2$-WSe$_2$ structures. The image was recorded at an operating voltage of 200 kV, see supplement. The atomically resolved HAADF image allows the detection of the roughness of the interface at sub-nm resolution due to exploiting of the Z-contrast, where lighter Mo atoms show darker contrast than heavier W atoms [51]. Thus, MoSe$_2$ appears darker and WSe$_2$ appears brighter in contrast. From these measurements we estimate the transition width of the boundary region between MoSe$_2$ and WSe$_2$ to be as narrow as $\approx 3$ nm.

We use water-assisted deterministic transfer to pick up as-grown, CVD LHs using polydimethylsiloxane (PDMS) and deterministically transfer and encapsulate them in hBN [49, 52]. We use these encapsulated samples for all optical spectroscopy measurements.

We first analyze the PL and differential white light reflectivity spectra of the individual monolayer areas.
(away from the heterojunction), collected at T=5 K. Fig. 1c shows superimposed PL (black) and reflectivity (red) spectra of MoSe$_2$ (top) and WSe$_2$ (bottom) monolayers after transfer and encapsulation in hBN. We measure in PL a neutral exciton (A$_{1s}$) linewidth of 7 meV in MoSe$_2$ and 5.5 meV in WSe$_2$ (Fig. 1c). These FWHM values are comparable to high-quality exfoliated MoSe$_2$ and WSe$_2$ MLs [15, 45–47]. We note that homogeneous and inhomogeneous broadening effects contribute to the total optical transition linewidth of any TMD ML [19]. The homogeneous part is defined by the radiative lifetime and is of the order of 1 meV. Thus, any substantially larger linewidth at low temperature will provide the inhomogeneous contribution due to imperfections in the ML or the direct environment [53]. MoSe$_2$ monolayers exhibit two pronounced PL emission peaks at 1.665 eV and 1.635 eV, assigned to neutral neutral (A$_{1s}$) and charged (T) excitons, respectively [54]. In the case of WSe$_2$, the A$_{1s}$ transition is located at 1.734 eV with the singlet trions (T) and dark excitons (X$^D$) lying 35 meV and 40 meV below A$_{1s}$, consistent with previous reports [55]. Additional peaks appear at lower energies, attributed to contributions from neutral, charged biexcitons and localized emission from defects. In Fig. 1c strong excitonic resonances appear in differential reflectivity for both materials with negligible Stokes shift in energy compared to the PL emission, which is a sign of negligible exciton localization effects for these spectra. Clear signatures of B$_{1s}$ exciton states are also observed in both materials. For WSe$_2$ the appearance of the A$_{2s}$ excited exciton state, which has a larger Bohr radius, shows the good quality of the CVD-grown monolayers of the LHs [56].

To optically examine the interface between the two materials, we perform linescans across several heterojunctions, see supplement. In particular, we collect the Raman, PL and reflectivity spectra within our detection spot while moving the sample over a ≈ 10 µm distance with a step size of ≈ 150 nm using attocube nanopositioners. Typical Raman PL and reflectivity scans across a heterojunction are shown in the contour plots of Fig. 2a,b,c. A black dashed line indicates the position of the heterojunction in each contour plot. As a common feature for all three contour plots, we observe abrupt changes in the optical spectra as we scan across the lateral heterojunction, as a result of distinctly different phonon energies and exciton transition energies in the two materials.

Now we discuss Raman spectroscopy results in Fig. 2a for ML MoSe$_2$ and WSe$_2$, collected at T=4 K using an excitation laser wavelength of A=633 nm. The main Raman peaks of MoSe$_2$ and WSe$_2$ can be identified in the contour plot (detailed Raman spectra can be found in the Supplementary Material, section B). For ML MoSe$_2$ we observe the A$_1'(\Gamma)$ phonon at 241 cm$^{-1}$ and the E$'$(\Gamma) at 291 cm$^{-1}$ [57], while a strong peak at 458 cm$^{-1}$ has been associated to other peaks to form triplets [58]. Interestingly, we also observe a strong peak at 531 cm$^{-1}$, recently assigned to multi-phonon processes associated with either both K and M point phonons or a combination of Γ point phonons [58]. The observation of this Raman peak is a signature of resonant excitation with an excited exciton state in MoSe$_2$. WSe$_2$ phonons are spectrally different compared to MoSe$_2$. The degenerate A$_1'(\Gamma)/E'(\Gamma)$ phonons are located at 250 cm$^{-1}$ and, similar to MoSe$_2$, a strong and recently discovered peak at 495 cm$^{-1}$ is also observed here and attributed to multi-phonon processes at K and
FIG. 3: Photoluminescence imaging. (a) Exciton diffusion measurements at a temperature of $T=4$ K for the MoSe$_2$ monolayer region (blue) and WSe$_2$ (red). In the inset we show the MoSe$_2$ diffusion spot compared to the WSe$_2$ spot. The laser spot diameter (black) is shown to indicate over which area excitons are initially generated, with small intensity oscillations due to Airy discs visible in logarithmic scale (b) same as (a) but at $T = 300$ K.
FIG. 4: **Photoluminescence imaging scans at T= 4 K.** (a) Amplitude of the PL profile when crossing the lateral interface (b) Squared width of the PL profile when crossing the lateral interface. The insets show results from a simple numerical model that captures the main features.

M points or combination of Γ point phonons [58]. The Raman analysis and the assignment of the Raman peaks further confirms high quality CVD-grown MLs. As the $A_1'(\Gamma)$ and $E'(\Gamma)$ modes remain practically constant in the Raman linescans (Fig. 2a), we tentatively infer that there are not any significant variations in the electron density [59] and lattice structure [60] close to the heterojunction.

The PL linescan in Fig. 2b shows emission from the main exciton transitions in WSe$_2$ and MoSe$_2$ (indicated by arrows) and a clear change in transition energy is discernible as we go across the lateral junction, with similar energies as for the individual spectra shown in Fig. 1c. Due to the extreme sensitivity of the PL emission energy spectrum on the defect concentration and dielectric environment [19], we see spectral shifts
between spectra taken at different positions. This makes the PL contour plot appear less smooth than the Raman and reflectivity linescans.

The reflectivity linescan shown in Fig. 2c shows a clear change in the main exciton energies as we scan from one material to the other. The main neutral exciton $A_{1g}$ feature in MoSe$_2$ in reflectivity is at an energy of 1.665 eV, whereas for WSe$_2$ this transition energy is clearly shifted to 1.734 eV. Reflectivity is less sensitive to the local defect and dielectric environment and therefore the measured transition energies remain comparatively constant from one spectrum to the other.

**Exciton diffusion.**—A PL imaging experiment is used to probe the effective diffusion length in the two TMD materials that are connected at the junction. First, we concentrate on results away from the lateral junction i.e. neither the excitation nor the PL emission profile have any spatial overlap with the lateral junction. The experiment consists in a local photogeneration of excitons with a He:Ne laser focused on a spot size of about 0.7 $\mu$m (see black curve in Fig. 3) and with an excitation power of 5 $\mu$W. The generated spatial gradient of the exciton concentration (i.e. chemical potential gradient) induces lateral diffusion of excitons which is probed by recording the spatial PL profile with a CMOS camera, see supplement, where we intergate over the full spectral range of the monolayer emission. Fig. 3a shows the PL profiles obtained at T=4 K on WSe$_2$ (red line) and MoSe$_2$ (blue line). We clearly observe that PL emission occurs over a larger spot diameter than the laser excitation [61, 62]. At this temperature WSe$_2$ shows a longer effective diffusion length than MoSe$_2$. This difference is also visible in the inset by directly comparing the images of the PL profile from the two materials.

Interestingly, this behavior is reversed at T=300 K where MoSe$_2$ shows a longer effective diffusion length than WSe$_2$ (see Fig. 3b). This is possibly linked to dark excitonic states which have longer lifetimes (PL emission times) [63, 64]. Indeed, the lowest energy state of the conduction band is a dark state in WSe$_2$ while it is a bright state in MoSe$_2$. It has therefore been reported that their PL intensity evolution with the temperature shows opposite trends: WSe$_2$ is darker at low temperature and brighter at room temperature as compared to MoSe$_2$ [65]. The contribution of dark exciton states with longer lifetime would increase the effective diffusion length as compared to MoSe$_2$, as dark exciton emission is negligible for MoSe$_2$ [38, 39]. Therefore, when we compare the two materials WSe$_2$ has a longer diffusion length at 4 K and a shorter one at T=300 K.

We then proceed to another experiment where we scan the laser excitation spot across the lateral WSe$_2$-MoSe$_2$ junction and record a PL image at each location, with a step size of about 100 nm between each acquisition. For the measurements in Fig. 4 we have chosen an interface region which gives uniform PL images, for more statistics on PL images see supplement. For our scan across the lateral interface, we fit for each image the PL profile by a Gaussian distribution from which we extract the amplitude and the width $w^2$. The evolution of these two parameters is displayed in Fig. 4 for the measurement at T=4 K. The amplitude (proportional to the PL intensity) decreases as we go from MoSe$_2$ to WSe$_2$. The width $w^2$ of the PL profile shows a more complex behavior, similar to an oscillation. We thus performed a modeling of the experiment by solving a two-dimensional diffusion equation in the two materials with a finite element method, see supplement. The modeling results are displayed in the insets in Fig. 4. They show qualitative agreement with the experimental data and indicate a smooth transition from one material to the other, in agreement with our TEM microscopy data in Fig. 1b. Our basic model reproduces the data when we consider as the only input information that WSe$_2$ has a longer effective exciton lifetime and a lower radiative efficiency than MoSe$_2$ at T=4 K. This scenario is consistent with the involvement of dark states for WSe$_2$ emission and might lie at the origin of the observed change in width $w^2$ of the PL profile as a function of scan distance.

**In conclusion,** we have performed detailed spectroscopic studies on CVD grown lateral MoSe$_2$-WSe$_2$ heterostructures. As an important step towards device processing and for accessing the intrinsic optical quality of the junction, we have first transferred the sample from its original growth substrate and then encapsulated the lateral heterostructures in top and bottom flakes of high quality hBN. Our experiments give access to the excitonic structures at cryogenic temperatures, with neutral exciton transition linewidth of the order of 5 meV. In exciton diffusion experiments we show that the MoSe$_2$ and WeSe$_2$ exciton transport show opposite trends in temperature dependent experiments, as dark excitons contribute to the PL signal in WSe$_2$ and not in MoSe$_2$. In future studies on these high-quality samples a clearer identification of interlayer excitons at the lateral junction is the target. Here an important consideration is the large spot diameter (order of 500 nm) of the excitation and detection spot in PL, whereas the lateral heterostructure border is only a 1-3 nm wide stripe (as we see in TEM) within this spot. So to increase the sensitivity for detecting photons originating from the junction region, it would be desirable to perform optical spectroscopy with higher spatial resolution. One promising direction here is to perform tip enhanced PL [66–68].
Optical microscope image and linescan description

An optical microscope image of the MoSe$_2$/WSe$_2$ lateral heterostructure (LH) and a schematic representation of the linescan process used in the optical spectroscopy experiments are shown in Fig. 5a,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 [49, 52]. The optical contrast between MoSe$_2$ and WSe$_2$ is different, allowing the direct visualization of the heterojunctions, shown with white dashed lines in Fig. 5a. In Fig. 5b we show a schematic representation of the process for the Raman, PL and Reflectivity linescans (Figure 2 in the main text). We position the excitation laser ($\lambda = 633$ nm, diffraction-limited spot diameter of 1 $\mu$m) on the area of WSe$_2$ and we use piezo-controlled nanopositioners to move the sample with steps of $\approx 150$ nm. After each step, a single -Raman, photoluminescence, reflectivity- spectrum is collected and finally all spectra are plotted as contour representations in Figure 2 of the main text.

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. 6a,b,c shows individual Raman, PL and reflectivity spectra of MoSe$_2$ (bottom), junction (middle) and WSe$_2$ (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 $\approx 1$ $\mu$m and 6 $\mu$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 amplitude of the differential reflectivity signal also depends on thin-film interference effects related to the thickness of top, bottom hBN and SiO$_2$ layers [69].

The main Raman peaks of MoSe$_2$ (Fig. 6a) include the $A_1^\prime(\Gamma)$ phonon at 241 cm$^{-1}$ and the $E'(\Gamma)$ at 291 cm$^{-1}$ [57], while a strong peak at 458 cm$^{-1}$ has been associated two other peaks to form triplets, see supplementary material of [58]. 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 $\Gamma$ point phonons [58]. The observation of this phonon is a signature of resonant excitation with an excited exciton state. WSe$_2$ phonons are spectrally different compared to MoSe$_2$. The degenerate $A_1^\prime(\Gamma)/E'(\Gamma)$ phonons are located at 250 cm$^{-1}$ and, similar to MoSe$_2$, a strong and recently discovered peak at 495 cm$^{-1}$ is also observed here and attributed to multi-phonon processes at K and M points or combination of $\Gamma$ point phonons [58]. 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 MoSe$_2$ and WSe$_2$ monolayers exhibit pronounced peaks (Fig. 6b), associated with neutral neutral ($X^0$) and charged ($X^T$) excitons. Additional peaks appearing at lower energies, especially in WSe$_2$ 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 A1s exciton state (see Fig. 6c). Clear signatures of B1s states are also observed in both materials while for WSe$_2$ the appearance of the A2s 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.
For the investigation of lateral transport, we used an experimental procedure very similar to the one described in part. The excitation is based on a HeNe laser with an excitation spot size of about 0.7μm and an excitation power of 5μW. 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, 2π], 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 2D classical diffusion equation with the use of the Matlab toolbox pdetool (partial differential equation toolbox). We set two zones having the same diffusion coefficient of 1 cm$^2$/s but different lifetime of $\tau_1 = 100$ ps and $\tau_2 = 400$ ps. To take into account the contribution of dark and bright states, different radiative efficiency $\eta$ are taken into account with a ratio of $\eta_1/\eta_2 = 10$. The laser excitation spot is assumed gaussian, and we model the PL profile for each position of the laser spot. The PL profiles $I_{PL}(r)$ are fitted by a gaussian distribution $I_{PL}(r) = A \exp(-r^2/w^2)$ where $w^2$ reflects the broadening of the PL profile (i.e. diffusion). We used Neumann boundary conditions with $dI_{PL}(r)/dr = 0$ to simulate ideal interface with no recombination.

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

**Acknowledgements**

Toulouse acknowledges partial funding from ANR IXTASE, ANR HiLight, NanoX project 2DLight, the Institut Universitaire de France, and the EUR grant ATRAP-2D NanoX ANR-17-EURE-0009 in the framework of the "Programme des Investissements d’Avenir", the Institute of quantum technology in Occitanie IQO and a UPS excellence PhD grant. Growth of hexagonal boron nitride crystals was supported by JSPS KAKENHI (Grants No. 19H05790, No. 20H00354 and No. 21H05233). The Jena group received financial support of the Deutsche Forschungsgemeinschaft (DFG) through a research infrastructure grant INST 275/257-1 FUGG, CRC 1375 NOA (Project B2), SPP2244 (Project TU149/13-1) as well as DFG grant TU149/16-1. This project has also received funding from the joint European Union’s Horizon 2020 and DFG research and innovation programme FLAG-ERA under grant TU149/9-1. (‡) D.B., I.P. and Z.G. contributed equally to this work. (†) T.L. now at Karlsruhe Institute of Technology, Laboratory for Electron Microscopy, 76131 Karlsruhe, Germany.
FIG. 7: Characterization of lateral heterostructures (a,b) Optical microscopy image of as grown MoSe$_2$-WSe$_2$ 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.
FIG. 8: Examples of PL imaging in different sample areas. We show more examples of typical PL emission spots in MoSe$_2$ (left), on the junction between the two materials (middle), and in WSe$_2$ (right) for two different sample temperatures as marked on the panels for each line.
atomically thin semiconductors. Nature Photonics 11, 497 (2017).

[15] Scuri, G. et al. Large excitonic reflectivity of monolayer mose2 encapsulated in hexagonal boron nitride. Phys. Rev. Lett. 120, 037402 (2018).

[16] Hong, X. et al. Ultrafast charge transfer in atomically thin mos 2/ws 2 heterostructures. Nature nanotechnology 9, 682 (2014).

[17] Turunen, M. et al. Quantum photonics with layered 2D materials. Nature Reviews Physics 1–18 (2022).

[18] Geim, A. K. & Grigorieva, I. V. Van der waals heterostructures. Nature 499, 419–425 (2013).

[19] Shree, S., Paradisanos, I., Marie, X., Robert, C. & Urbaszek, B. Guide to optical spectroscopy of layered semiconductors. Nature Reviews Physics 3, 39–54 (2021).

[20] Andrei, E. Y. et al. The marvels of moiré materials. Nature Reviews Materials 6, 201–206 (2021).

[21] Lu, A.-Y. et al. Janus monolayers of transition metal dichalcogenides. Nature nanotechnology 12, 744–749 (2017).

[22] Zheng, T. et al. Excitonic dynamics in janus mosse and wsse monolayers. Nano Letters 21, 931–937 (2021).

[23] Petrić, M. M. et al. Raman spectrum of janus transition metal dichalcogenide monolayers wsse and mosse. Physical Review B 103, 035414 (2021).

[24] Najafidehbaghani, E. et al. 1D p–n junction electronic and optoelectronic devices from transition metal dichalcogenide lateral heterostructures grown by one-pot chemical vapor deposition synthesis. Advanced Functional Materials 2101086 (2021).

[25] Huang, C. et al. Lateral heterojunctions within monolayer mose 2–wse 2 2D semiconductors. Nature materials 13, 1096–1101 (2014).

[26] Duan, X. et al. Lateral epitaxial growth of two-dimensional layered semiconductor heterojunctions. Nature nanotechnology 9, 1024–1030 (2014).

[27] Gong, Y. et al. Vertical and in-plane heterostructures from ws 2/mos 2 monolayers. Nature materials 13, 1135–1142 (2014).

[28] Wu, W. et al. Self-powered photovoltaic pyroelectric detector established on lateral monolayer mos2-ws2 heterostructures. Nano Energy 51, 45–53 (2018).

[29] Pospischil, A., Furchi, M. M. & Mueller, T. Solar-energy conversion and light emission in an atomic monolayer p–n diode. Nature nanotechnology 9, 257–261 (2014).

[30] Sahoo, P. K., Memaran, S., Xin, Y., Balicas, L. & Gutiérrez, H. R. One-pot growth of two-dimensional lateral heterostructures via sequential edge-epitaxy. Nature 553, 63–67 (2018).

[31] Wang, J., Li, Z., Chen, H., Deng, G. & Niu, X. Recent advances in 2D lateral heterostructures. Nano-Micro Letters 11, 1–31 (2019).

[32] Li, M.-Y. et al. Epitaxial growth of a monolayer ws2-mos2 lateral pn junction with an atomically sharp interface. Science 349, 524–528 (2015).

[33] Zhang, Y. et al. Edge-epitaxial growth of 2D nbs2-ws2 lateral metal-semiconductor heterostructures. Advanced Materials 30, 1803656 (2018).

[34] Mahjouri-Samani, M. et al. Patterned arrays of lateral heterojunctions within monolayer two-dimensional semiconductors. Nature communications 6, 1–6 (2015).

[35] Chu, Y.-H. et al. Atomic scale depletion region at one dimensional mose2-ws2 heterointerface. Applied Physics Letters 113, 241601 (2018).

[36] Guo, Y. & Robertson, J. Band engineering in transition metal dichalcogenides: Stacked versus lateral heterostructures. Applied Physics Letters 108, 233104 (2016).

[37] Lau, K. W., Gong, Z., Yu, H., Yao, W. et al. Interface excitons at lateral heterojunctions in monolayer semiconductors. Physical Review B 98, 115427 (2018).

[38] Lu, Z. et al. Magnetic field mixing and splitting of bright and dark excitons in monolayer mose2. Physical Review B 105, 015017 (2019).

[39] Robert, C. et al. Measurement of the spin-forbidden dark excitons in mos2 and mose2 monolayers. Nature communications 11, 1–8 (2020).

[40] Yang, M. et al. Relaxation and darkening of excitonic complexes in electrostatically doped monolayer wsse: Roles of exciton-electron and trion-electron interactions. Phys. Rev. B 105, 085302 (2022).

[41] Taniguchi, T. & Watanabe, K. Synthesis of high-purity boron nitride single crystals under high pressure by using ba-bn solvent. Journal of Crystal Growth 303, 525 – 529 (2007).

[42] Rhodes, D., Chae, S. H., Ribeiro-Palau, R. & Hone, J. Disorder in van der waals heterostructures of 2D materials. Nature materials 18, 541 (2019).

[43] Raja, A. et al. Dielectric disorder in two-dimensional materials. Nature nanotechnology 14, 832–837 (2019).

[44] Cadiz, F. et al. Excitonic linewidth approaching the homogeneous limit in mosb-based van der waals heterostructures. Phys. Rev. X 7, 021026 (2017).

[45] Ajayi, O. A. et al. Approaching the intrinsic photoluminescence linewidth in transition metal dichalcogenide monolayers. 2D Materials 4, 031011 (2017).

[46] Wierzbowksi, J. et al. Direct exciton emission from atomically thin transition metal dichalcogenide heterostructures near the lifetime limit. Scientific reports 7, 12383 (2017).

[47] Stier, A. V. et al. Magnetooptics of exciton rydberg states in a monolayer semiconductor. Physical Review Letters 120, 057405 (2018).

[48] Shree, S. et al. High optical quality of mos2 monolayers grown by chemical vapor deposition. 2D Materials 7, 015011 (2019).

[49] Paradisanos, I. et al. Controlling interlayer excitons in mos 2 layers grown by chemical vapor deposition. Nature communications 11, 1–7 (2020).

[50] Martin, E. W. et al. Encapsulation narrows and preserves the excitonic homogeneous linewidth of exfoliated monolayer mose2. Physical Review Applied 14, 021002 (2020).

[51] Liu, J. Scanning transmission electron microscopy and its application to the study of nanoparticles and nanoparticle systems. Journal of electron microscopy 54, 251–278 (2005).

[52] Jia, H. et al. Large-scale arrays of single-and few-layer mos 2 nanomechanical resonators. Nanoscale 8, 10677–10685 (2016).

[53] Moody, G. et al. Intrinsic homogeneous linewidth and broadening mechanisms of excitons in monolayer transition metal dichalcogenides. Nature communications 6, 8315 (2015).

[54] Shree, S. et al. Observation of exciton-phonon coupling in mose 2 monolayers. Physical Review B 98, 035302 (2018).

[55] Wang, G. et al. In-plane propagation of light in transition
metal dichalcogenide monolayers: optical selection rules. 

**Physical review letters** **119**, 047401 (2017).

[56] Delhomme, A. *et al.* Magneto-spectroscopy of exciton rydberg states in a cvd grown wse2 monolayer. *Applied Physics Letters* **114**, 232104 (2019).

[57] Soubelet, P., Bruchhausen, A. E., Fainstein, A., Nogajewski, K. & Faugeras, C. Resonance effects in the raman scattering of monolayer and few-layer mose 2. *Physical Review B* **93**, 155407 (2016).

[58] McDonnell, L. P., Viner, J. J., Rivera, P., Xu, X. & Smith, D. C. Observation of intravalley phonon scattering of 2s excitons in mose2 and wse2 monolayers. *2D Materials* **7**, 045008 (2020).

[59] Sohier, T. *et al.* Enhanced electron-phonon interaction in multivalley materials. *Physical Review X* **9**, 031019 (2019).

[60] Dadgar, A. *et al.* Strain engineering and raman spectroscopy of monolayer transition metal dichalcogenides. *Chemistry of Materials* **30**, 5148–5155 (2018).

[61] Cadiz, F. *et al.* Exciton diffusion in wse2 monolayers embedded in a van der waals heterostructure. *Applied Physics Letters* **112**, 152106 (2018).

[62] Kulig, M. *et al.* Exciton diffusion and halo effects in monolayer semiconductors. *Phys. Rev. Lett.* **120**, 207401 (2018).

[63] Zhang, X.-X., You, Y., Zhao, S. Y. F. & Heinz, T. F. Experimental evidence for dark excitons in monolayer wse 2. *Physical review letters* **115**, 257403 (2015).

[64] Robert, C. *et al.* Fine structure and lifetime of dark excitons in transition metal dichalcogenide monolayers. *Physical review B* **96**, 155423 (2017).

[65] Wang, G. *et al.* Spin-orbit engineering in transition metal dichalcogenide alloy monolayers. *Nature communications* **6**, 1–7 (2015).

[66] Lee, H. *et al.* Tip-enhanced photoluminescence nanospectroscopy and nano-imaging. *Nanophotonics* **9**, 3089–3110 (2020).

[67] Darlington, T. P. *et al.* Imaging strain-localized excitons in nanoscale bubbles of monolayer wse2 at room temperature. *Nature Nanotechnology* **15**, 854–860 (2020).

[68] Zhang, S. *et al.* Nano-spectroscopy of excitons in atomically thin transition metal dichalcogenides. *Nature Communications* **13**, 1–8 (2022).

[69] Robert, C. *et al.* Optical spectroscopy of excited exciton states in mos2 monolayers in van der waals heterostructures. *Phys. Rev. Materials* **2**, 011001 (2018).