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# Dislocation-driven growth of $WS_2/WSe_2$ quantum well superlattices

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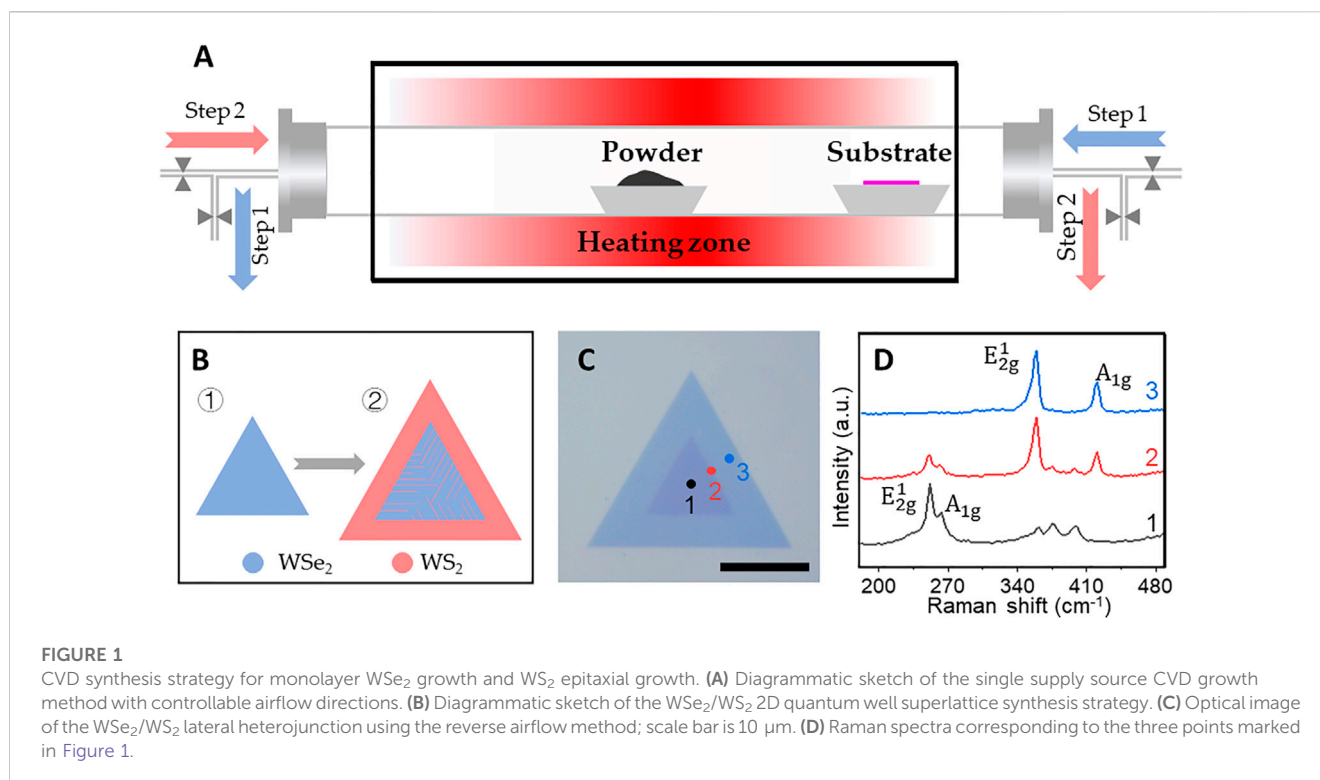
The synthesis of two-dimensional lateral heterojunctions with nanoscale characteristic width and sharp interfaces remains challenging. The quantum confinement effects are still difficult to create on 2D materials since widths smaller than 5 nm are necessary for quantum confinement effects and quantum well applications. In this study, we demonstrated the growth of a sub-2-nm tungsten sulfide quantum well array in a monolayer of tungsten selenide, driven by the climb of mismatch dislocation in a heterointerface due to the lattice mismatch. Width-controllable 2D quantum well superlattices are theoretically formed by the mismatch dislocation-driven growth mechanism, according to our analysis. Thus, abundant photonic electronic properties can be obtained in 2D quantum well superlattices formed at varied lateral heterointerfaces, which will support the study of topological insulators and superconductors.

## KEYWORDS

chemical vapor deposition (CVD), lateral epitaxial growth, monolayer  $WSe_2$ , 2D quantum well superlattices, misfit dislocation-driven growth

## 1 Introduction

Two-dimensional (2D) lateral heterojunctions and superlattices with a series of novel properties have been generated and reported recently (Geim and Grigorieva, 2013; Liu et al., 2014; Gong et al., 2015; Li et al., 2015; Zhang et al., 2017; Chen et al., 2018; Liu et al., 2019; Shi et al., 2020; Zhang et al., 2021). Available growth methods produce featured widths in the scale of hundreds of nanometers or, at best, at micrometer scale and commonly result in rough and defective interfaces with extensive chemical intermixing (Li et al., 2015; Liu et al., 2015; Zhang et al., 2017; Shi et al., 2020). Quantum wells in conventional semiconductors with their unique electronic structure and quantum confinement have important applications in quantum cascade lasers, solar cells, high-electron mobility transistors, and infrared photodetectors (Faist et al., 1994; Gudiksen et al., 2002). In a 2D system, quantum well structures may be best generated by laterally sandwiching a nanoscale strip of a 2D semiconductor between two strips of another 2D semiconductor with a different band gap. It is theoretically necessary to control the width of the 2D quantum well comparable to the de Broglie wavelength of the carrier (the sub-10-nm regime and, ideally, less than



5 nm) so that quantum size effects occur. However, available growth methods only produce features with widths in the micrometer or, at best, 100 nm scale. Therefore, implementing quantum confinement effects in 2D systems remains challenging. In recent years, several studies have reported observations of quantum wells in 2D systems. Han et al. (2018) and Zhu et al. (2020) reported the fabrication methods of one-dimensional MoS<sub>2</sub> quantum channels in WSe<sub>2</sub>-MoS<sub>2</sub> and MoSe<sub>2</sub>-MoS<sub>2</sub> hetero-systems, respectively.

Here, we report the growth of high-quality sub-2-nm-wide quantum wells within semiconductor monolayers, making use of the lattice mismatch between two semiconducting materials in the 2D lateral heterojunctions. The growth was controlled by individual misfit dislocations formed at the lateral heterointerface between a WSe<sub>2</sub> and a WS<sub>2</sub>. Atomic resolution scanning transmission electron microscopy (STEM) images revealed that these tungsten sulfide quantum wells were less than 2 nm in width and formed fully coherent lateral interfaces with the tungsten selenide monolayer matrix without extended defects.

These results showed that the insertion of metal and S atoms into the dislocation cores induced dislocation climb, with concomitant selective substitution of Se atoms around the dislocation core by S atoms. The substitution process, driven by the local strain field, led to the growth of WS<sub>2</sub> quantum well arrays laterally sandwiched in the WSe<sub>2</sub> monolayers.

Theoretically, the mismatch dislocation at the interface needed to be formed in intervals to release strain. Repetition of the misfit dislocation-driven growth mechanism formed parallel WS<sub>2</sub> quantum wells, and these parallel 2D narrow nano-bands were quantum well superlattices. Considering the large variety of 2D materials, 2D quantum well superlattices in varied lateral heterojunctions will display diverse photonic electronic properties; therefore, controllable width 2D quantum well superlattices are expected to be fabricated.

## 2 Experimental details

### 2.1 Growth of monolayer WSe<sub>2</sub>

A quartz boat loaded with WSe<sub>2</sub> powder (~1 g) was located at the center heating zone of the furnace, as shown in Figure 1A, and a clean silica (~300 nm)/Si substrate was located at the terminal of the furnace as the growth substrate. The horizontal distance between the powder and the silicon wafer was kept between 11 and 15 cm. During the process of temperature increase, the growth substrate was placed upstream of the source, and the airflow was set to 300 sccm (along the “step 1” arrow in Figure 1A) for 15 min to remove the undesired oxygen and water from the tube. The Ar airflow was then kept at 50 sccm. When the central heating zone reached the target temperature of 1150°C, the 50 sccm airflow with the transferred direction (along the “step 2” arrow in Figure 1A) ensured the growth substrate was located downstream of the source and then maintained for 10 min for growth. Next, the airflow direction was reversed (along the “step 1” arrow in Figure 1A) to stop the growth process and allow it to naturally cool down to room temperature. The entire process occurred under ambient pressure in an argon atmosphere.

### 2.2 Growth of WSe<sub>2</sub>/WS<sub>2</sub> 2D quantum well superlattices

To obtain the WSe<sub>2</sub>/WS<sub>2</sub> 2D quantum well, the monolayer WSe<sub>2</sub> was chosen as a seed to further the epitaxial growth of the monolayer WS<sub>2</sub>. At the same time, quantum wells started forming at the interfaces of WSe<sub>2</sub>/WS<sub>2</sub> lateral heterojunctions. For the epitaxial growth of WS<sub>2</sub>, the WS<sub>2</sub> powder and prepared monolayer WSe<sub>2</sub>

(substituted for clean substrate and  $\text{WSe}_2$  powder) were placed at the same position in another specified CVD system similar to the system used for  $\text{WSe}_2$  growth. Likewise, during the period of temperature increase, the growth substrate was placed upstream of the source and the airflow (along the “step 1” arrow in Figure 1A) was set to 300 sccm for 15 min, and then reduced to 100 sccm and kept at that airflow rate. When the central heating zone reached the target temperature of  $1120^\circ\text{C}$ , the airflow was set to 100 sccm in the transferred direction (along the “step 2” arrow in Figure 1A), ensuring the growth substrate was located downstream of the source, and then maintained for 5 min for  $\text{WS}_2$  epitaxial growth. After that, the airflow direction was reversed (along the “step 1” arrow in Figure 1A) to stop the epitaxial growth process and allow it to naturally cool down to room temperature. The entire process occurred under ambient pressure in an argon atmosphere.

## 2.3 Characterization

Optical microscopy and STEM were used to characterize the morphologies of the nano-heterojunctions. The Raman spectra were performed using a Renishaw confocal Raman system excited by a 488-nm laser at room temperature. The applied laser power was set to 5%, and the exposure time was approximately 0.1 s. STEM imaging and EELS analysis were carried out using a Thermo Scientific Themis Z 3.2 system equipped with a cold field-emission gun and a mirror lens corrector operating at 300 kV. All STEM experiments were completed at room temperature. The strain distribution of the nanostructure was calculated based on the geometric phase analysis method using the FRWRtools plugin for DigitalMicrograph ([www.physik.hu-berlin.de/en/sem/software/software\\_frwrtools](http://www.physik.hu-berlin.de/en/sem/software/software_frwrtools)).

The STEM samples were prepared using a poly-methyl-methacrylate (PMMA)-assisted method. PMMA was spin-coated onto the heterojunction sample until the PMMA layer fully covered the heterojunction sample and then baked at  $180^\circ\text{C}$  for 3 min. Afterward, the wafer was immersed in a saturated 0.2 M NaOH solution to etch the  $\text{SiO}_2$  layer. After the sample coated with PMMA was separated from the silicon wafer, it was transferred into fresh deionized water, repeatedly, to wash away residual contaminants and then fished by a conventional lacey carbon film TEM grid. The as-transferred sample was dried naturally in the ambient environment and then immersed in acetone overnight to clean up the PMMA coating layers. All the STEM samples were baked at  $150^\circ\text{C}$  for 6 h to avert unexpected contamination from hydrocarbon.

## 3 Results and discussion

### 3.1 Lateral heterojunction with 2D quantum wells

Figure 1B is the synthesis strategy diagrammatic sketch of the  $\text{WSe}_2/\text{WS}_2$  2D quantum well, and Figure 1B① and Figure 1B② show the idealized schematic sketch of the monolayer  $\text{WSe}_2$  and the  $\text{WSe}_2/\text{WS}_2$  2D quantum well, respectively. The  $\text{WS}_2$  quantum well was inserted into the monolayer  $\text{WSe}_2$ , which started to grow from the  $\text{WSe}_2/\text{WS}_2$  interface, as depicted in Figure 1B②. The width of the quantum well in Figure 1B② is exaggerated to make the display easy to

recognize and understand. Figure 1C is the optical image of the monolayer  $\text{WSe}_2/\text{WS}_2$  lateral heterojunction synthesized on a silica/silicon substrate; it shows two concentric regions with slightly different optical contrast due to the refractive index and thickness difference between tungsten selenide and tungsten sulfide (Henrie et al., 2004; Blake et al., 2007). Here, the nanoscale  $\text{WS}_2$  2D quantum wells formed at the  $\text{WSe}_2/\text{WS}_2$  interface but were difficult to recognize in the optical image due to the low enlargement factor of optical microscopy. To further explore the spatial modulation of structural and optical properties in  $\text{WSe}_2/\text{WS}_2$  lateral heterostructures, the confocal Raman microscope was used to characterize the nanostructure. We picked up three points marked “1,” “2,” and “3” in Figure 1C for the Raman study, where “1” (black point), “2” (red point), and “3” (blue point) are located at the internal tungsten selenide, the  $\text{WSe}_2/\text{WS}_2$  interface, and the epitaxial tungsten sulfide area, respectively. The Raman spectra at different locations showed distinct differences: the internal region Raman spectrum had a significant peak at  $250\text{ cm}^{-1}$  (black line in Figure 1D), consistent with the  $A_{1g}$  resonance pattern of  $\text{WSe}_2$ , and the peripheral region Raman spectrum had two significant peaks, at  $350\text{ cm}^{-1}$  and  $419\text{ cm}^{-1}$  (blue line in Figure 1D), corresponding to the  $E_{2g}^1$  and  $A_{1g}$  resonance patterns of  $\text{WS}_2$ . Significantly, the Raman spectrum of the  $\text{WSe}_2/\text{WS}_2$  interface showed both the resonance modes of  $\text{WSe}_2$  and the resonance modes of  $\text{WS}_2$  (red line in Figure 1D), which indicate that tungsten selenide and tungsten sulfide coexist well within the same triangular domain.

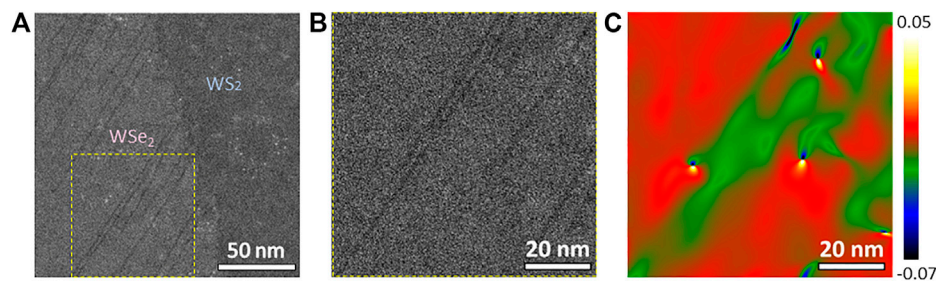
### 3.2 Strain analysis in a $\text{WSe}_2/\text{WS}_2$ 2D quantum well

These  $\text{WS}_2$  quantum wells extending from the  $\text{WSe}_2/\text{WS}_2$  interface, mostly distributed in parallel, were generally observed in the lateral  $\text{WSe}_2/\text{WS}_2$  heterojunction samples, as shown in the STEM image in Figure 2A. Figure 2B is the enlarged view of Figure 2A, and the clear boundaries between  $\text{WSe}_2$  and  $\text{WS}_2$  in the  $\text{WSe}_2/\text{WS}_2$  2D quantum well are apparent. Also, the strain distribution corresponding to Figure 2B is given in Figure 2C. The perfect lattice from the monolayer  $\text{WSe}_2$  was used as a reference for calculating the strain. In the perfect monolayer, the lattice constant of  $\text{WSe}_2$  was  $\sim 4\%$  larger than that of  $\text{WS}_2$ , which indicates that the  $\text{WS}_2$  quantum well had high and uniform tensile strain along its growth direction, leading to the observed dislocation-free transverse interface. However, the situation was different in the direction perpendicular to the growth direction, where there was a comparatively larger lattice mismatch between  $\text{WS}_2$  and  $\text{WSe}_2$  ( $\sim 4.3 \pm 0.5\%$ ), as revealed by the calculated strain distribution spectrum.

### 3.3 Atomic-scale sharp $\text{WS}_2$ 2D quantum well

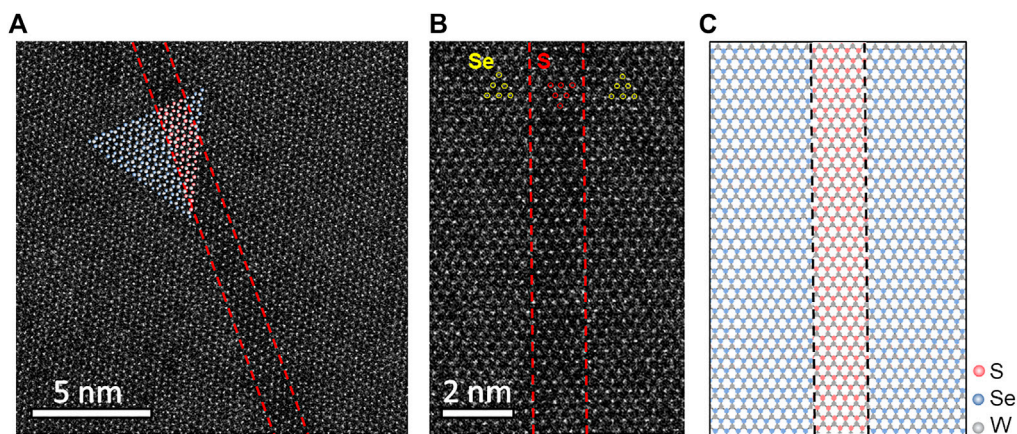
Figure 3A shows a 2D quantum well in which the monolayer of the  $\text{WS}_2$  nanoribbons is laterally sandwiched between two monolayer  $\text{WSe}_2$  nanoribbons. As shown in the STEM image in Figure 3A, the Se and S atomic sites are distinguished from the image contrast due to different atomic numbers (Krivanek et al., 2010; Zhou et al., 2012), where the  $\text{WS}_2$  quantum well shows a lower image intensity than the surrounding monolayer  $\text{WSe}_2$ , and the  $\text{WS}_2/\text{WSe}_2$  interface is marked by red dashed lines.





**FIGURE 2**

Strain analysis of  $WS_2$  2D quantum wells at the interface between  $WSe_2$  and  $WS_2$ . (A) STEM image of  $WS_2$  quantum wells with uniform width at the  $WSe_2/WS_2$  lateral heterojunction interface. (B) Enlarged view of (A). (C) Corresponding strain distribution of (B).



**FIGURE 3**

Atomic-scale sharp  $WS_2$  2D quantum well embedded in monolayer  $WSe_2$ . (A) STEM image of the  $WSe_2/WS_2$  lateral heterojunction interface; the red dashed line highlights the interface between  $WSe_2$  and  $WS_2$ . (B) High-resolution STEM-ADF image of a  $WS_2$  quantum well; the red dashed line along the armchair orientation highlights the interface between  $WSe_2$  and  $WS_2$ . The yellow and red circles mark the Se and S atoms, respectively. (C) Atomic structural model of the  $WS_2$  quantum well corresponding to (B). Pink, blue, and gray spheres represent the S, Se, and W atoms, respectively.

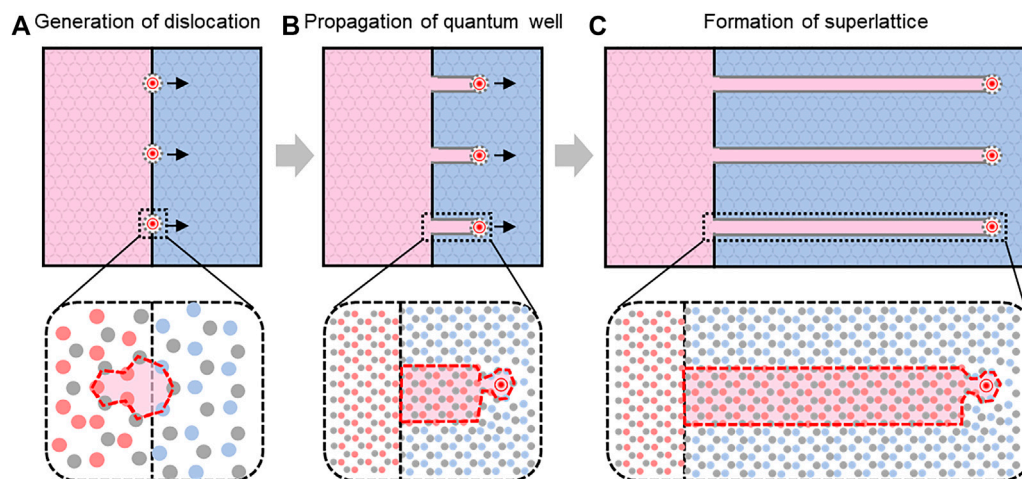
The high-resolution STEM-annular dark-field (ADF) image of a  $WS_2$  quantum well is shown in Figure 3B; the yellow and red circles mark Se and S atoms, respectively. It was observed that the  $\sim 1.2$  nm  $WS_2$  quantum well was embedded in a single layer of  $WSe_2$  and growing along the hexagonal armchair orientation. Correspondingly, Figure 3C is the idealized atomic structural model of the  $WS_2$  quantum well according to Figure 3B. The interface is highlighted by the red and black dashed lines in Figures 3B,C, respectively. It is obvious that there are no mismatches or defects at the interface, which means a fully coherent lateral interface was formed.

### 3.4 Formation mechanism of $WS_2$ quantum wells at the $WSe_2/WS_2$ lateral interface

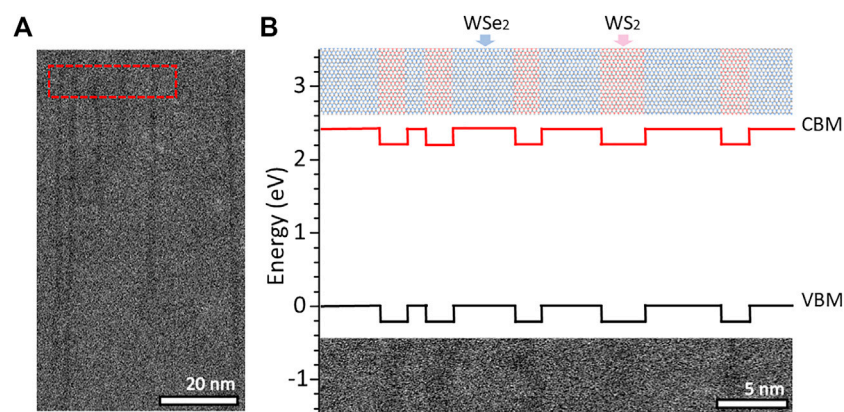
Similar to film growth, strain relaxation at the epitaxial interface with intrinsic lattice mismatch produces mismatch dislocation once the critical width is exceeded (Matthews and Blakeslee, 1974; Jain et al., 1997). For the lateral  $WSe_2/WS_2$  heterostructures, the mismatch

dislocation array spaced  $\sim 8$  nm in the zigzag orientation was expected to alleviate the lattice strain due to the presence of  $\sim 4\%$  lattice mismatch between  $WSe_2$  and  $WS_2$ . Through careful observation, it was found that  $WS_2$  quantum wells relied on a dislocation core for growth at the lateral  $WSe_2/WS_2$  heterointerface in general, and further growth was considered driven by misfit dislocation climb. As shown in Figure 4A, the insertion of one W atom and two S atoms from the gas source was considered initiation of the dislocation-driven growth as such dislocation drove the W and S atoms to invade the  $WSe_2$  along the hexagonal zigzag orientation, which is consistent with our experimental results. According to statistical analysis, the width of  $WS_2$  quantum wells is about 1.2 nm, which is equivalent to the width of the four  $WS_2$  unit cells. A four-unit-cell-width and a one-unit-cell-width  $WS_2$  nano-seed, shaded in light red in Figure 4B, are shown penetrating to the  $WSe_2$  monolayer to propagate the quantum well.

Theoretically, the formation of the mismatch dislocation at the interface was required per each  $\sim 8$  nm interval to release strain. After repeating the aforementioned growth mechanism, parallel  $WS_2$  quantum wells were formed, and these parallel 2D narrow nano-bands were quantum well superlattices, as shown in Figure 4C.



**FIGURE 4**  
Diagrammatic sketch of the formation mechanism of  $WS_2$  quantum wells at the interface between  $WSe_2$  and  $WS_2$ . (A) Generation of dislocation. (B) Propagation of quantum wells. (C) Formation of the  $WS_2$  superlattice.



**FIGURE 5**  
 $WS_2$  quantum superlattice. (A,B) Atomic structure model of the  $WSe_2/WS_2$  superlattice and the calculated band structure. The red and black solid lines represent the valence band maximum (VBM) and the conduction band minimum (CBM), respectively.

### 3.5 Promising 2D quantum well superlattices with atomically sharp lateral interfaces

According to the aforementioned results, there is potential to generate a 2D quantum superlattice, consisting of equally spaced 2D quantum wells, if growth conditions are precisely controlled. The  $WS_2$  quantum well grown by this mechanism formed a type II band arrangement with the surrounding monolayer  $WSe_2$ , as shown in Figure 4B.

Meanwhile, the n-type doped ultranarrow  $WS_2$  nanoribbons were expected to be conductors due to the type II band alignment. Nanoribbons of higher carrier concentrations can be obtained by this process of modulation doping as ionizing donors are distributed in the  $WSe_2$  region and thus reduce Coulomb scattering in  $WS_2$ . Furthermore, the quantum wells shown in Figure 5A are not evenly spaced, and such a phenomenon could be attributed to the periodic array that was not initially formed at the interface. The bottom area in Figure 5B is an enlarged display

of the red dotted frame in Figure 5A. There are different energy band distributions in the region of tungsten sulfide and tungsten selenide. From Figure 5A and Figure 5B, it can be concluded that the length distribution of quantum wells ranges from ten nanometers to a few microns, while the spacing between the quantum wells is about 8 nm, which is basically consistent with the previous theoretical analysis results. High-quality 2D semiconductor quantum well superlattices may be prepared by a misalignment drive mechanism if the growth parameters of the chemical vapor deposition (CVD) system are stably controlled.

## 4 Conclusion

To summarize, we successfully synthesized sub-2-nm quantum well arrays in  $WSe_2/WS_2$  lateral heterojunctions. High-resolution STEM images showed the atomically sharp lateral interfaces between the

WS<sub>2</sub> quantum well and monolayer WSe<sub>2</sub>. Among them, ~4% lattice mismatch between WSe<sub>2</sub> and WS<sub>2</sub> monolayers led to misfit dislocation arrays with an average spacing of 8 nm at the lateral heterointerface. After that, dislocations drove the corresponding atoms to penetrate along the hexagonal armchair orientation, which plays a key role in the efficient growth of quantum wells. According to the results of the WSe<sub>2</sub>/WS<sub>2</sub> lateral heterojunction system, such quantum well superlattice structures are expected to be formed in varied lateral heterojunctions. Considering the large variety of 2D materials, 2D quantum well superlattices in varied lateral heterojunctions will display abundant photonic electronic properties, which provide new roads for the study of topological insulators and superconductors.

## Data availability statement

The original contributions presented in the study are included in the article/[Supplementary Material](#); further inquiries can be directed to the corresponding author.

## Author contributions

HY: conceptualization, methodology, formal analysis, resources, visualization, and writing—original draft. LZ: methodology and formal analysis. ZH: conceptualization and visualization. TZ: methodology, and writing—review and editing. SZ: investigation, validation, and formal analysis. XZ: laboratory resources. ZA: investigation. XL: visualization. BZ: conceptualization, writing—review and editing, supervision, and project administration. All authors have read and agreed to the published version of the manuscript.

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## Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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## Supplementary material

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