



# Van der Waals epitaxial growth and optoelectronics of a vertical MoS<sub>2</sub>/WSe<sub>2</sub> p–n junction

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## Abstract

Two-dimensional (2D) transition metal dichalcogenides (TMDs) have attracted extensive attention due to their unique electronic and optical properties. In particular, TMDs can be flexibly combined to form diverse vertical van der Waals (vdWs) heterostructures without the limitation of lattice matching, which creates vast opportunities for fundamental investigation of novel optoelectronic applications. Here, we report an atomically thin vertical p–n junction WSe<sub>2</sub>/MoS<sub>2</sub> produced by a chemical vapor deposition method. Transmission electron microscopy and steady-state photoluminescence experiments reveal its high quality and excellent optical properties. Back gate field effect transistor (FET) constructed using this p–n junction exhibits bipolar behaviors and a mobility of 9 cm<sup>2</sup>/(V·s). In addition, the photodetector based on MoS<sub>2</sub>/WSe<sub>2</sub> heterostructures displays outstanding optoelectronic properties ( $R = 8$  A/W,  $D^* = 2.93 \times 10^{11}$  Jones, on/off ratio of  $10^4$ ), which benefited from the built-in electric field across the interface. The direct growth of TMDs p–n vertical heterostructures may offer a novel platform for future optoelectronic applications.

**Keywords** MoS<sub>2</sub> · WSe<sub>2</sub> · Chemical vapor deposition (CVD) · Vertical heterostructure · Optoelectronic transistor

## 1 Introduction

In the past decades, two-dimensional (2D) transition metal dichalcogenides (TMDs) layered semiconductors have been considered as promising candidates for next-generation electronics and optoelectronics. This is due to their unique properties such as atomically thin thickness, and to their electronic, photonic, and mechanical properties [1–6]. Moreover, the heterostructures that are composed of various 2D layered TMDs materials have attracted more

and more attention as a consequence of their diversified band alignments [5]. For example, type-I materials are usually used for light-emitting diodes [7], type-II have an important application in photovoltaic devices [8, 9], and type-III are widely used in the tunneling field effect transistors [10]. Until now, there have been diverse ways to synthesize the heterostructures, including mechanical stacking, the solution method, and chemical vapor deposition (CVD) [12–14], in which the CVD approach can easily synthesize materials with high production yield and large area. Many van der Waals heterostructures have been successfully composed by the CVD method and can be used in integrated optoelectronic applications [15–17]. For instance, Li et al. reported high-performance optoelectronic devices based on van der Waals vertical MoS<sub>2</sub>/MoSe<sub>2</sub> heterostructures with enhanced photoresponsivity of 36 A/W and a remarkable detectivity of  $4.8 \times 10^{11}$  Jones [16]. Wu et al. demonstrated the vapor growth of WSe<sub>2</sub>/WS<sub>2</sub> heterostructures with distinct nonlinear optical properties by controlling crystallographic alignments in the form of AA and AB stacking [15]. However, although there is demand for mass production of high-quality vertical p–n heterostructures for use in integrated devices,

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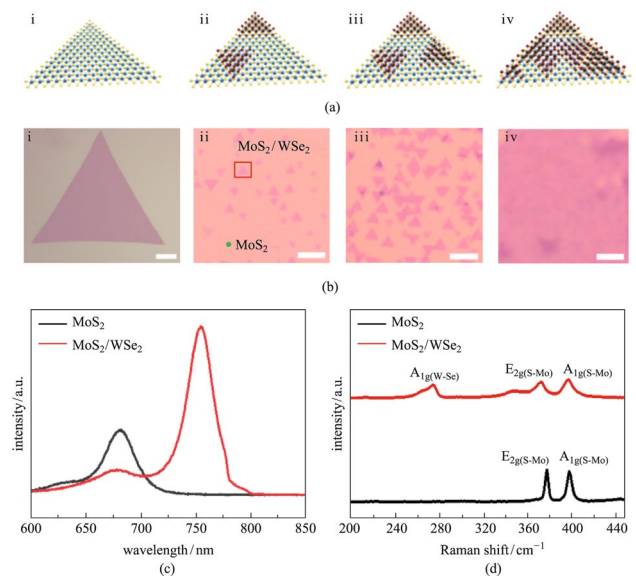
there are still few reports on them because synthesizing the non-alloyed heterojunctions with sharp interface by CVD method is difficult. Most of the devices developed so far are based on single-material [18–21], but their performance is limited by defects and small light absorption cross-sections [21, 22]. Therefore, the controllable synthesis of vertical p–n heterostructures is very important for future integrated electronic and optoelectronic applications.

In this work, we report production of vertical  $\text{MoS}_2/\text{WSe}_2$  p–n heterojunctions through a two-step CVD method and explore its application in photoelectric devices. The optical measurements and scanning transmission electron microscopy (STEM) demonstrate that the heterostructures are of high quality, without alloying. Electrical measurements exhibit considerable photon–electron conversion efficiency, photoresponsivity, detectivity, and stable light response. The study thus provides an alternative approach to design various TMDs heterostructures for future optoelectronic applications.

## 2 Experimental details

### 2.1 Synthesis of $\text{MoS}_2/\text{WSe}_2$ vertical heterostructures

The vertical  $\text{MoS}_2/\text{WSe}_2$  heterostructures were synthesized by a two-step growth CVD approach. In the first step,  $\text{MoS}_2$  template was synthesized using a salt-assisted method. Salt could reduce the evaporation temperature of  $\text{MoO}_3$ , thereby reducing the difficulties for production of  $\text{MoS}_2$  with large-area. An alumina boat containing S powder was placed at upstream of a furnace. Then, another boat with mixed powder of NaCl and  $\text{MoO}_3$  (99.99%, Alfa Aesar) was positioned at the central region of furnace, on top of which three pieces of  $\text{SiO}_2/\text{Si}$  substrates was placed for deposition. Before heating, the system was cleaned by pure Ar flow for 30 min at a rate of 80 standard cubic centimeters per minute (sccm). The furnace was heated to 810 °C within 30 min at 60 sccm, with Ar as the carrier gas, then maintained at that temperature for a further 10 min. Finally, the furnace naturally cooled down to room temperature. For the second step, to synthesize vertical  $\text{WSe}_2/\text{MoS}_2$  heterostructures, a boat with as-grown  $\text{MoS}_2$  templates was placed at downstream of furnace for subsequent deposition of  $\text{WSe}_2$ . The  $\text{WSe}_2$  source powder (99.99%, Alfa Aesar) was put at the central heating area of the furnace. After the cleaning procedure as described above, the furnace was heated to 1020 °C within 30 min at 60 sccm and maintained at that temperature for 3 min. After the reaction, the furnace cooled down to room temperature naturally.



**Fig. 1** Growth morphology and the optical characterizations of as-grown  $\text{MoS}_2/\text{WSe}_2$  heterostructures. Schematic views of heterostructure growth (**a**<sub>i</sub>–**a**<sub>iv</sub>) and their corresponding optical images (**b**<sub>i</sub>–**b**<sub>iv</sub>). Scale bar: 20  $\mu\text{m}$ . **c** PL spectra of monolayer  $\text{MoS}_2$  and  $\text{MoS}_2/\text{WSe}_2$  heterostructure. **d** Raman spectra of  $\text{MoS}_2$  and  $\text{MoS}_2/\text{WSe}_2$  heterostructure

### 2.2 Characterizations

The morphologies of heterostructures were characterized using an optical microscopy (Zeiss Axio Scope A1), an atomic force microscope (AFM, Bruker Multimode 8), and a scanning transmission electron microscope (JEM ARM200F). The optical measurements (Photoluminescence and Raman) were conducted using a confocal microscope (WITec, alpha-300) with a 532 nm laser focused by an objective.

### 2.3 Device fabrication and optoelectronic measurements

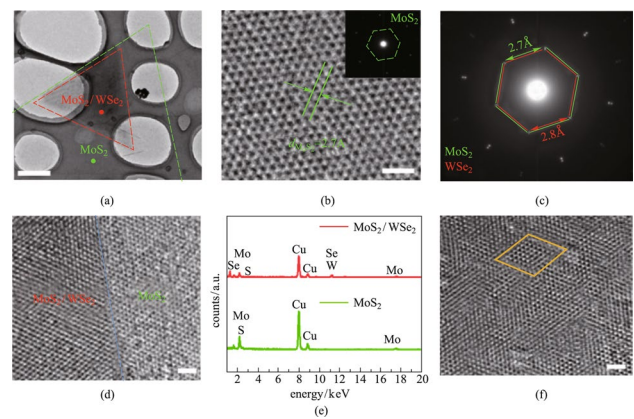
Standard e-beam lithography (EBL, Raith 150) and metal thermal evaporation were performed to fabricate the Au/Cr (10 nm/50 nm) electrodes on the as-grown heterostructures with a lift-off approach. The electric and optoelectronic properties of the heterostructures were measured in vacuum with Lake Shore Probe Station and Agilent B1500A semiconductor analyzer at room temperature.

## 3 Results and discussion

The  $\text{MoS}_2/\text{WSe}_2$  heterostructures were grown by a two-step CVD method. Figure 1a shows a schematic diagram of the atomic growth model, and Fig. 1b exhibits a picture of the heterostructure under the optical microscope. The schematic

view and optical image of MoS<sub>2</sub> grown on SiO<sub>2</sub>/Si substrate are shown in Figs. 1a<sub>i</sub> and 1b<sub>i</sub> (Additional file 1: Fig. S1 for detail) respectively. The lateral size of monolayer MoS<sub>2</sub> triangles ranged from 20 to 200 μm. The red square in Fig. 1b<sub>ii</sub> indicates the small WSe<sub>2</sub> triangles that was grown on MoS<sub>2</sub> in the second step, and the area marked by the green spots is the underlying MoS<sub>2</sub>. The evolution of morphology of WSe<sub>2</sub> on MoS<sub>2</sub> after 2, 4, and 6 min growth is shown in Figs. 1a<sub>ii</sub>–a<sub>iv</sub> and 1b<sub>ii</sub>–b<sub>iv</sub>, respectively, with the whole WSe<sub>2</sub> covered eventually. The optical image clearly indicated that WSe<sub>2</sub> on MoS<sub>2</sub> had only 0° and 60° stacking modes. Atomic force microscopy showed that MoS<sub>2</sub> and WSe<sub>2</sub> were both monolayers (Additional file 1: Fig. S2). Figure 1c shows the photoluminescence (PL) spectra of MoS<sub>2</sub> and MoS<sub>2</sub>/WSe<sub>2</sub> heterostructures. The peaks at 680 and 755 nm correspond to the A excitons of MoS<sub>2</sub> and WSe<sub>2</sub>, respectively. The PL intensity of MoS<sub>2</sub> becomes quenched in the heterostructure because of the charge transfer in the MoS<sub>2</sub>/WSe<sub>2</sub> interface, with type-II band alignment, as shown in Fig. 4f. Figure 1d shows the Raman spectra of MoS<sub>2</sub> and MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure. The peak positions of MoS<sub>2</sub> locate at 379 cm<sup>-1</sup> (E<sub>2g(S-Mo)</sub>) and 397 cm<sup>-1</sup> (A<sub>1g(S-Mo)</sub>), respectively, while that of the heterostructure locate at 270 cm<sup>-1</sup> (A<sub>1g(W-Se)</sub>), 372 cm<sup>-1</sup> (E<sub>2g(S-Mo)</sub>), and 400 cm<sup>-1</sup> (A<sub>1g(S-Mo)</sub>). PL and Raman results are consistent with MoS<sub>2</sub> and WSe<sub>2</sub> properties previously reported, proving the successful synthesis of MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure.

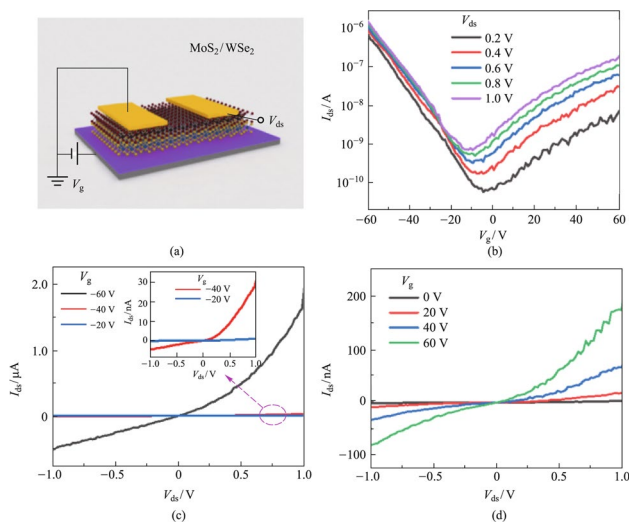
The MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure grown on SiO<sub>2</sub>/Si was transferred to a copper net (see the supporting information for the detail transfer method), and then the microstructure and chemical composition of the MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure were analyzed by transmission electron microscopy (TEM). Figure 2a gives the low-resolution scanning transmission electron microscope (TEM) image of MoS<sub>2</sub>/WSe<sub>2</sub> vertical heterostructure. The triangular area marked by green dash line is the bottom layer of MoS<sub>2</sub>, and the red triangle is the heterojunction area. Figure 2b is the high-resolution transmission electron microscope (HRTEM) image of the bottom MoS<sub>2</sub>, with a lattice spacing of 2.7 Å, corresponding to the (100) crystal plane of the 2H phase MoS<sub>2</sub>. The inset is the selected area electron diffraction (SAED) pattern of MoS<sub>2</sub>. The SAED of the MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure is shown in Fig. 2c. Two sets of regular hexagon diffraction pattern are displayed clearly. The polygonal diffraction patterns marked by green and red lines have a crystal plane spacing of 2.7 Å and red 2.8 Å, corresponding to MoS<sub>2</sub> and WSe<sub>2</sub> (100) crystal planes, respectively. Figure 2d shows the area at the interface of the heterostructure under the HRTEM. The right side is MoS<sub>2</sub>, and the left side is the heterostructure. The atomic structure is clear, which proves that the high-quality MoS<sub>2</sub>/WSe<sub>2</sub> vertical heterojunction with a clear interface was successfully synthesized. The TEM energy-dispersive X-ray photoelectron spectroscopy (EDS) of different regions



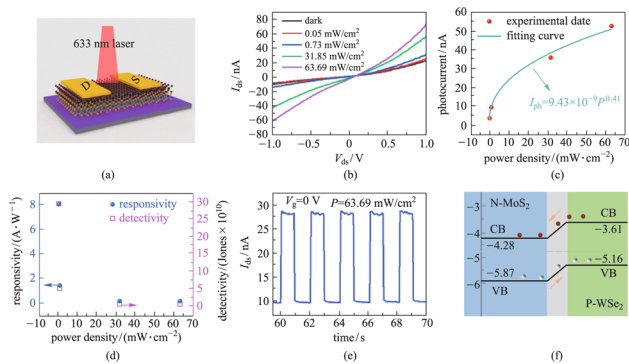
**Fig. 2** High-resolution atomic characterizations of MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure. **a** Low-resolution TEM image of transferred MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure on a Cu grid. Scale bar: 1 μm. **b** HRTEM image of MoS<sub>2</sub>. Scale bar: 1 nm. **c** SAED patterns of MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure. **d** Atomic resolution scanning TEM image of the heterostructure interface. Scale bar: 2 nm. **e** EDS spectra of the MoS<sub>2</sub>/WSe<sub>2</sub> vertical heterostructure. **f** HRTEM image of heterostructure region. Scale bar: 2 nm

is shown in Fig. 2e. The red line shows several peaks for elements such as W, Se, Mo, S, and Cu (from the copper mesh), corresponding to the MoS<sub>2</sub>/WSe<sub>2</sub> heterojunction. The green line only shows Mo, S, and Cu peaks, corresponding to the MoS<sub>2</sub>. The results agrees with The X-ray photoelectron spectroscopy (XPS) measurement of the heterojunction given in Additional file 1: Fig. S3. Figure 2f is the HRTEM image in the heterojunction region, the moiré patterns can be clearly observed due to the interlayer coupling between MoS<sub>2</sub> and WSe<sub>2</sub>. This observation further demonstrates the successful synthesis of the high-quality, well-defined MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure.

To further investigate the charge transport properties of the vertically stacked MoS<sub>2</sub>/WSe<sub>2</sub> van der Waals heterostructures, back-gate field-effect transistors (FET) were fabricated based on MoS<sub>2</sub>/WSe<sub>2</sub> p–n heterojunctions. Figure 3a displays the device structure in which Ti/Au electrodes are deposited on WSe<sub>2</sub>, as the drain and source electrodes. The transport characteristic of the heterostructure device given in Fig. 3b shows ambipolar behavior, which is attributed to the p-type WSe<sub>2</sub> and the n-type MoS<sub>2</sub>. Figure 3b shows that, when a negative gate voltage ( $V_g$ ) is applied, the WSe<sub>2</sub> layer is turned on and the device exhibits p-type behavior. Conversely, when the MoS<sub>2</sub> layer is on state and the device shows n-type behavior while the gate voltage was positive. At the same time, the current increases when the drain-source voltage  $V_{ds}$  changes from 0.2 to 1 V. The diagram indicates that the threshold voltage is  $-10$  V, and the on/off ratio is  $10^4$ . The mobility could be obtained using the expression  $\mu = g_m L / (WC_0 V_{ds})$ , where  $g_m$  represents the transconductance,  $L = 17.3$  μm and  $W = 54.5$  μm are the length and the width of



**Fig. 3** Electronic characterization of MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure transistors. **a** Schematic configuration. **b** Transfer characteristic curves. **c, d**  $I_{ds}$ – $V_{ds}$  output characteristics curves at various gate voltages ranging from  $-60$  to  $60$  V



**Fig. 4** Optoelectronic device performance of MoS<sub>2</sub>/WSe<sub>2</sub> vertical heterostructure. **a** Schematic illustration of the device. **b**  $I_{ds}$ – $V_{ds}$  curves across the heterojunction under 633 nm laser illumination with different incident powers.  $V_{ds}$  ranges from  $-1$  to  $1$  V. **c** Dependence of device photocurrent on illumination power densities. **d** Photoreponsivity and detectivity of the photodetector at various illumination power densities ranging from 0 to 63.69 mW/cm<sup>2</sup>. **e** Photocurrent response of the device; the laser excitation is turned on and off by a chopper working at 1 Hz ( $V_{ds} = 1$  V,  $V_g = 0$  V,  $P = 63.69$  mW/cm<sup>2</sup>). **f** Band structures and the charge transfer process at the interface of the MoS<sub>2</sub>/WSe<sub>2</sub> p–n heterojunction

the device, respectively.  $C_0$  is the capacitance per unit area for the 280 nm thick SiO<sub>2</sub>. Based on the data shown in Fig. 3b, the calculated mobility is 9 cm<sup>2</sup>/(V·s). Figure 3c, d show the corresponding output characteristics of MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure. The inset in Fig. 3c is the enlarged view of the region marked by the red circle. The drain-source current  $I_{ds}$  decreases as the  $V_g$  varies from  $-60$  to  $-20$  V, and increases as  $V_g$  raises from 0 to 60 V. All the device measurements were conducted under vacuum at room temperature.

Benefiting from the large built-in electric field at the p–n heterojunction interface and the resulting high separation efficiency of photogenerated carriers, p–n heterostructures are widely used in optoelectronic applications. Under illumination of 633 nm laser light, the photovoltaic and photoresponse characteristics of the MoS<sub>2</sub>/WSe<sub>2</sub> vertical heterostructures were examined further. The photodetector based on MoS<sub>2</sub>/WSe<sub>2</sub> vertical heterojunction is schematically illustrated in Fig. 4a, with drain-source voltage  $V_{ds}$  applied to the heterojunction device. Figure 4b shows the  $I_{ds}$ – $V_{ds}$  curves at various illumination intensities ranging from 0 to 63.69 mW/cm<sup>2</sup> ( $V_g = 20$  V). More electron–hole pairs are generated as the illumination intensity increases, resulting in a greater photocurrent. In Fig. 4c, the photocurrent intensities are evaluated at a gate bias of 20 V. Other photocurrent data under different gate voltages are shown in Additional file 1: Fig. S4a. The photocurrent intensity ( $I_{ph}$ ) is defined as  $I_{ph} = I_{light} - I_{dark}$ , where  $I_{light}$  ( $I_{dark}$ ) is current measured under light (dark state). The relation between  $I_{ph}$  and laser power density  $P$  can be expressed as  $I_{ph} = aP^\alpha$ . The fitted parameters of  $a$  and  $\alpha$  obtained from Fig. 4c are  $9.43 \times 10^{-9}$  and 0.41, respectively. The responsivity ( $R$ ) can be calculated using  $R = I_{ph}/(PA)$ , where  $A$  is the effective area of the device channel. The calculated responsivity of the device is up to 8 A/W ( $P = 0.05$  mW/cm<sup>2</sup>,  $V_{ds} = 1$  V, device area of 943.85  $\mu\text{m}^2$ ), which is greater than that of monolayer MoS<sub>2</sub> and WSe<sub>2</sub> devices previously reported [24–26]. In addition, the value of  $R$  can reach up to 41 A/W at 60 V gate voltage (Additional file 1: Fig. S4b). To characterize the sensitivity of a photodetector, the device detectivity ( $D^*$ ) is used.  $D^*$  can be calculated using the formula  $D^* = RA^{1/2}/(2eI_{dark})^{1/2}$ , where  $e$  is the electron charge, assuming  $D^*$  is primarily affected by the shot noise of the dark current [26, 27]. According to this formula,  $D^*$  can reach a maximum value of  $2.93 \times 10^{11}$  Jones as shown in Fig. 4d. Detectivity at various gate voltages can be found in Additional file 1: Fig. S4c. The photoresponse speed of the MoS<sub>2</sub>/MoSe<sub>2</sub> heterostructure photodetector was investigated further by turning on and off the laser beam (633 nm, 63.69 mW/cm<sup>2</sup>). Figure 4e displays exemplary photocurrent curves for the photodetector, showing excellent stability and reliability of on/off switching behavior ( $V_{ds} = 1$  V,  $V_g = 0$  V,  $P = 63.69$  mW/cm<sup>2</sup>). Figure 4f exhibits the band structure of the MoS<sub>2</sub>/WSe<sub>2</sub>. It is shown that a typical type-II heterostructure is formed. When the laser beam was focused on the heterojunction, the electrons in WSe<sub>2</sub> were transferred to MoS<sub>2</sub>, whereas the holes in MoS<sub>2</sub> were transferred to WSe<sub>2</sub>. This results in the photogating effect, wherein the charges modify the conductance of the WSe<sub>2</sub> channel. The high responsivity and the excellent reliability indicate that the designed MoS<sub>2</sub>/WSe<sub>2</sub> p–n heterojunction may achieve ultrasensitive photodetection in the future.

## 4 Conclusions

In this work, we developed a controllable two-step growth method to prepare vertically stacked MoS<sub>2</sub>/WSe<sub>2</sub> heterostructures. Furthermore, PL Spectroscopy, Raman spectroscopy, and HRTEM were used to characterize the obtained p–n heterojunctions. The results indicate that the samples are of high crystal quality. A p–n field effect transistor was fabricated based on the heterostructures. The bipolar behavior was observed and the device exhibited high carrier mobility of up to 9 cm<sup>2</sup>/(V·s). Under the laser illumination, excellent photodetection properties was obtained with the photoresponsivity approaching 8 A/W and detectivity of 2.93 × 10<sup>11</sup> Jones. Meanwhile, stable on/off photoswitching was also demonstrated. All the results show that the achieved MoS<sub>2</sub>/WSe<sub>2</sub> p–n heterojunctions have great potential in integrated optoelectronic applications.

**Supplementary Information** The online version contains supplementary material available at <https://doi.org/10.1007/s12200-022-00041-4>.

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**Author contributions** YX, SLC, and ALP conceived the idea and participated in the data analysis. YX performed the material growth, optical characterization and drafted the manuscript. All authors read and approved the final manuscript.

## Declarations

**Competing interests** The authors declare that they have no competing interests.

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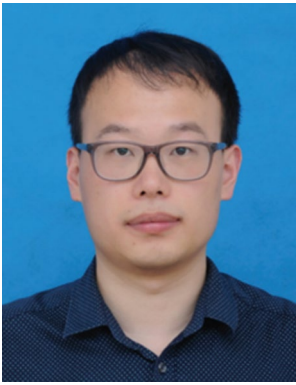
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