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One-step Synthesis of Metal/Semiconductor Heterostructure NbS₂/MoS₂

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ABSTRACT

Chemical vapor deposition (CVD) has proven its surpassing advantages, such as larger scale, interlayer orientation control and clean interface, in the synthesis of transitional metal dichalcogenide (TMDC) semiconductor/semiconductor van der Waals (vdW) heterostructures. However, it is suffering problems of high melting points and low chemical reactivity of metal oxide feedstocks in the preparation of high-quality metal/semiconductor (M/S) TMDC vdW heterostructures. Here, for the first time, we report the synthesis of the M/S TMDC vdW heterostructure NbS₂/MoS₂ via a one-step halide-assisted CVD method, which effectively overcomes the drawbacks of metal oxide precursors. What's more, this one-step method provides the high quality and clean interface of the NbS₂/MoS₂ heterostructure, which has been proved by the transmission electron microscopy characterization. A mechanism that MoS₂ finishes the growth first and subsequently serves as a superior substrate for the growth of NbS₂ is proposed. This novel method will open up new opportunities in the syntheses of other M/S TMDC vdW heterostructures and facilitate the research of the TMDC M/S interface.

1. Introduction

Van de Waals (vdW) heterostructures,^{1, 2} vertically stacked by diverse two-dimensional (2D) components, have shown great potentials on novel physical applications³⁻⁵ and next generation optoelectronic devices such as atomically thin P-N junction.⁶⁻⁹ Benefiting from the weak interlayer vdW interaction, engineering of vdW heterostructures can be realized via controlling the chemical components,^{7, 10-12} layer numbers¹³ and interlayer stacking orders,¹⁴ leading to a transition of designing new materials from a traditionally methodology-driven motivation to the one driven by the function of materials. Thanks to the atom-thin nature, various physical properties, and numerous analogues in the family (>40),¹⁵ 2D transitional metal dichalcogenides (TMDCs) have been employed to construct the artificial vdW heterostructures^{11, 16} with unique functionalities.

Chemical vapor deposition (CVD) has become a dominant method to synthesize the TMDC vdW heterostructures with large size, such as ReS₂/WS₂¹⁷ and MoSe₂/WSe₂.¹⁰ However, up to now, only a hand of TMDC vdW heterostructures have been prepared by this method, and nearly all of them are focused on Semiconductor/Semiconductor (S/S) vdW heterostructures,^{7, 10, 17, 18} leaving Metal/Semiconductor (M/S) counterparts unexplored. This may be primarily ascribed to the following reasons: 1) High melting points and low chemical reactivity of metal oxides (e.g. Nb₂O₅ and Ta₂O₅), make metallic TMDCs hard to synthesize.¹⁹ 2) Crystal structures mismatch.¹⁸ It is relatively easier to grow vdW heterostructures for two components that have the same crystal structure, e.g. 2H-MoS₂ and 2H-WS₂. Yet for different crystal structures, e.g. 2H and 1T, the two components should be meticulously selected to achieve the epitaxial growth.

The fast separation of the photogenerated charge carriers of M/S heterostructures, due to the variant electrical features, makes 2D M/S vdW heterostructures²⁰ very fascinating in the optoelectronic and photocatalytic applications²¹⁻²³. Recently, a two-step CVD method concerning the selenization of sequent deposited Nb₂O₅ and WO₃ to produce M/S

$\text{NbSe}_2/\text{Nb}_x\text{W}_{1-x}\text{Se}_2$ was adopted reluctantly owing to the drawbacks of Nb_2O_5 as metal precursors.²⁴ As known, such method will lead to interlayer alloying, bring relative high defect and grain boundary concentration,²⁵ and finally result in a poor interface. Therefore, to synthesize vdW heterostructures with high-quality interface becomes challenging but profound as it is the key to understand the layer¹³ or orientation-dependent¹⁶ physical properties of vdW heterostructures, exemplified by the ultra-low-frequency Raman spectrum²⁶ and interlayer excitons.

Here, we demonstrate the synthesis of high-quality 2D TMDC M/S vdW heterostructure $\text{NbS}_2/\text{MoS}_2$ by one-step halide-assisted CVD method, which provides a sharp atomic M/S interface. Firstly, through mixing the metal oxide precursor with the alkaline metal halide salt²⁷, we successfully produced a 20 μm single crystalline monolayer niobium disulfide (NbS_2), as confirmed by the Atomic Force Microscopy (AFM). Raman spectrum of few-layer sample with a size up to 60 μm reveals the 3R-stacking order of the obtained NbS_2 . Further developing the growth recipe, we obtained the 2D M/S TMDC vdW heterostructure $\text{NbS}_2/\text{MoS}_2$, which was substantiated by Optical Microscopy (OM), Scanning Electron Microscopy (SEM), X-ray Photoelectron Spectroscopy (XPS) and Photoluminescence (PL) Tests. Transmission Electron Microscopy (TEM) characterization provided firm evidence of the high quality of this $\text{NbS}_2/\text{MoS}_2$ heterostructure, and the periodic superlattice originated from well-aligned orientation affirmed the clean interface of the obtained heterostructure. TEM further revealed the R-stacking order between the MoS_2 and NbS_2 layer, which is favoured in pristine CVD-grown NbS_2 . With the aid of OM and Raman characterization, we figured out that, due to the difference of growth temperature, only when the growth of MoS_2 is completed, NbS_2 begins the epitaxial growth on MoS_2 . This is the first time that high-quality 2D M/S TMDC vdW heterostructure has been prepared by one-step CVD method. Our work is believed to speed up the syntheses of other M/S TMDC vdW heterostructures and provide a platform for the research into the TMDC M/S interface.

Figure 1. Synthesis of large-area layered NbS₂ single crystal. (a) Schematic of the CVD setup. (b-d) Optical images of one, two and three layered NbS₂, inset of (b) shows AFM and the thickness of 1.1 nm indicates the monolayer nature of (b). All the scale bars are 10 μ m. (e-g) Raman spectra of samples with different thicknesses (one, two and three layers). (f, g) Zoom-in of E₂ and A₁ peaks in (e), presents the phonon stiffening and softening characteristics.

2. Results and Discussions

2.1. Halide-assisted CVD Synthesis of NbS₂

At first, we demonstrate the synthesis of intrinsic NbS₂. The CVD setup is depicted in **Figure 1a**: under the gas flow of argon, the gaseous sulfur was introduced to the central high temperature zone (800 $^{\circ}$ C), and reacted with the evaporated metal precursors (mixture of niobium pentoxide, Nb₂O₅ and sodium chloride, NaCl). NbS₂ layers were then deposited on the 285 nm SiO₂/Si substrate. By using argon gas with the flow rate of 60 standard cubic

centimetre per minute (sccm), monolayer NbS₂ with the size up to 20 μm was synthesized, shown in **Figure 1b**. To the best of our knowledge, this is the largest monolayer single-crystal NbS₂ that has ever been produced by CVD method. As proposed in the previous report,^{27, 28} the reaction between Nb₂O₅ and NaCl, forming a metaloxychloride at high temperature, which leads to a lower melting point and higher chemical reactivity than Nb₂O₅, contributes to this better result. AFM height topography (inset of **Figure 1b**) shows that the thickness of the as-grown NbS₂ is about 1.1 nm, which confirms the monolayer nature of our sample. Further by applying different flow rates, we could also achieve the synthesis of bilayer (**Figure 1c**) and tri-layer (**Figure 1d**) NbS₂, which has a largest area (60 μm) with a thickness around 3.0 nm. In order to confirm the crystal quality and stacking order of this sample, Raman spectra using a 532nm laser as light source were performed. As shown in **Figure 1e**, three peaks at 311.2 cm⁻¹, 343 cm⁻¹, and 381.2 cm⁻¹ were found to be the characteristic peaks of 3R NbS₂, corresponding to the E₁, E₂ and A₁ vibration modes.²⁹ This result fits very well with the spectra reported previously.^{30, 31} Worth to mention, the reported phonon stiffening and softening features of E₂ and A₁ modes from thicker to thinner NbS₂,³² which results from the dielectric screening of long-range Coulomb interactions, could also be observed in our result (**Figure 1f** and **1g**). The origin of the peak located at 179.4 cm⁻¹ is still unclear, which might come from higher-order Raman processes.

2.2. Synthesis and Characterization of NbS₂/MoS₂

The successful synthesis of high-quality NbS₂ atomic layer sheds light on the fabrication of the 2D TMDC M/S vdW heterostructures. As MoS₂ shares the same anionic atom with NbS₂, we choose MoS₂ as the substrate to achieve the epitaxial growth of NbS₂, forming the 2D TMDC M/S vdW heterostructure NbS₂/MoS₂ (schematically presented in **Figure 2a**). In order to grow MoS₂, MoO₃ powder was placed at the up flow of the Nb₂O₅/NaCl mixtures, leaving a distance about 1.5 cm (**Figure S1**). Under the growth temperature of 800 °C and 60 sccm argon gas, we

successfully obtained this vertical stacking heterostructure (**Figure 2b**). As we can see from **Figure 2b** and **2c**, the distinct contrasts between NbS₂ and MoS₂ in both OM and SEM images, which results from different light absorption and conductivity, confirm that this is a vertical NbS₂/MoS₂ heterostructure rather than a bilayer MoS₂. To further ascertain this, XPS test was performed at the thicker area (marked with a circle in **Figure S2a**). As shown in **Figure 2d-2f**, the peaks at 228.8 eV and 232.0 eV are ascribed to the 3d_{5/2} and 3d_{3/2} orbitals from Mo,³³ and 203.2 eV and 206.0 eV are attributed to Nb 3d_{5/2} and 3d_{3/2} core levels,³⁴ while 162.9 eV and 164.1 eV are corresponded to S 2p_{3/2} and 2p_{1/2} core levels. All these peaks match very well with the reported data. Besides, by integrating the areas of the peaks, the obtained ratio of the anion S to the cations (Nb and Mo), which is 2.125, further demonstrates the stoichiometry of this NbS₂/MoS₂ heterostructure. To provide evidence of physical properties (i.e., optical properties), PL single spectra and mapping were conducted on the pure MoS₂ and heterostructure (region 1 and 2 in **Figure 2b**) respectively. From **Figure 2g**, it clearly shows that in region 2 (heterostructure), the PL of the bottom-layer MoS₂ has been quenched because of the fast charge transfer process,³⁵ which confirms the existence of metal/semiconductor interface. It is noted that the small change in PL single spectra (**Figure 2h**) of the pure MoS₂ and uncovered MoS₂ (region 1) indicates a low dopant ratio (less than 0.5%) of Nb in the bottom-layer MoS₂.³⁶ Hence, with the aid of OM, SEM, XPS and PL, 2D M/S vdW heterostructure NbS₂/MoS₂ was successfully identified.

Figure 2. NbS₂/MoS₂ vdW heterostructure. (a) Schematic of NbS₂/MoS₂ vdW heterostructure. OM (b) and SEM (c) images, show the evident contrast between NbS₂ and MoS₂ owing to the difference in light absorption and conductivity. Scale bar: 10 μ m. (d-f) XPS peaks of Mo 3d (c), Nb 3d (d) and S 2p (e) from NbS₂/MoS₂ vdW heterostructure on SiO₂/Si substrate. **The ratio of anion (S) to cations (Mo and Nb) is 2.125, via performing peak area integration, which shows good stoichiometry in this heterostructure.** (g) Photoluminescence (PL) mapping of area marked with dashed rectangle in (b), shows the PL of MoS₂ was quenched in the heterostructure (region 2). (h) PL single spectra of pure MoS₂, region 1 and 2 in (b). The small change in PL of pure MoS₂ and 1 indicate low dopant ratio of Nb in MoS₂ (region 1 in (g)).

To further determine the atomic structure and stacking of the 2D NbS₂/MoS₂ vdW heterostructure, we conducted annular dark-field (ADF) imaging in an aberration-corrected scanning transmission electron microscope (STEM). **Figure 3a** shows the morphology of the as-transferred sample in a low-magnification Z-contrast image. In STEM's Z-contrast image, the intensity is directly related to the atomic number and the thickness of the sample,³⁷ which can therefore be used to discriminate different atoms and thickness. As Mo(Z=42) and Nb(Z=41) have very close atomic number, a NbS₂ monolayer would exhibit almost the same contrast as monolayer MoS₂. Thus, by analysing the intensity and comparing it with that of the vacancy area, we can firstly distinguish different layer numbers (1L, 2L and 3L) of the sample in specific area (**Figure S3**). Furthermore, the detailed atomic structure of different areas can be revealed by high-resolution STEM Z-contrast imaging and corresponding fast

Figure 3. Atomic structure of the NbS₂/MoS₂ vdW heterostructure. (a) Low-magnification ADF-STEM image of the sample, where the monolayer area was MoS₂, the bilayer area was monolayer (1L) NbS₂ grown on top of 1L MoS₂; (b) ADF-STEM image acquired from the bilayer area in (a), which shows the periodic Moiré pattern. Scale bar: 5 nm; (c) FFT pattern obtained from (b). In the magnified view of the FFT pattern (the inset in black dash line), the reciprocal lattices of NbS₂ and MoS₂ are marked by green and orange circles, respectively; (d) Top view and side view of the atomic model of the NbS₂/MoS₂ vdW heterostructure with an R stacking orientation. (e) The atomic-resolution ADF-STEM image recorded at the lower right corner of (b). (f) The simulated STEM image based on an R-type stacking model in (d), which is consistent with (e). Scale bar of (e) and (f): 2 nm. The supercell of the NbS₂/MoS₂ heterostructure is demarcated by the red dashed rhombuses in (b, d, e and f), with a lattice constant around 6.6 nm.

Fourier transform (FFT) pattern. **Figure 3b** is a large scale ADF image of the bilayer region, from which we can observe Moiré pattern with a periodicity of around 6.6 nm, highlighted by the red dashed rhombus. The corresponding fast Fourier transformed (FFT) image of **Figure 3b** shows two different sets of hexagonally arranged spots. Careful analysis of these spots reveals their lattice constants (parameter: a): one corresponding to NbS₂ of 3.33 Å while the other corresponding to MoS₂ of 3.21 Å, within instrumental error.³⁸ In addition, the two sets of well-aligned spots in **Figure 3c** demonstrate that the NbS₂ and MoS₂ layers are aligned with

no twist angle. In order to further resolve the stacking order of the bilayer heterostructure, we applied inverse FFT of NbS₂ and MoS₂ respectively (**Figure S4c** and **S4d**). From the same orientation of NbS₂ and MoS₂ in the zoom-in images (**Figure S4e** and **S4f**), R stacking of the bilayer heterostructure was preliminarily confirmed. Via using the reported method³⁹, we compared the experimental image (**Figure 3e**) with the simulation results (**Figure S5**) of both R (**Figure 3d**) and H stacking NbS₂/MoS₂ bilayer models. From intensity profiles of several areas (**Figure S6**), we found the simulation image of R stacking is consistent with experimental image quantitatively, while the simulated H stacking image is apparently different. This confirms the R-stacking order between NbS₂ and MoS₂ in our heterostructure.

Figure 4. Proposed mechanism for the vertical heterostructure growth: At first, MoS₂ grows and forms a full triangle. (a) Optical image and (e) schematic structure of MoS₂; (b) and (f) NbS₂ on the top nucleates and grows at the corners of bottom-layer MoS₂; (c) and (g) Then top-layer NbS₂ at these corners grows up and merges at the edge, while the core of MoS₂ on the bottom remains uncovered; (d) and (h) Finally, the growth of NbS₂ continues and achieves the full coverage of the bottom MoS₂. Scale bar: 10 μ m.

From **Figure S2a** and **S2b**, we found that the second-layer NbS₂ started the growth from one or two triangle corners, leaving the other uncovered. This is not like a one-step growth behaviour, but more like a two-step process. Via regulating the time for growth, we can achieve different morphologies of this heterostructure: (1) NbS₂ on the top firstly nucleates and grows at the

corners of bottom-layer MoS₂ (**Figure 4b**); (2) Then top-layer NbS₂ at these corners grows up and merges at the edge, while the core of MoS₂ on the bottom remains uncovered (**Figure 4c**); (3) Finally, the growth of NbS₂ continues and achieves the full coverage of the bottom MoS₂ (**Figure 4d**). On the basis of these observations, the possible mechanism for the heterostructure growth could be understood as follows (schematically shown in **Figure 4e-4h**): Firstly, MoS₂ nucleates at low temperature (650-750°C), and grows into a monolayered single-crystal triangle. The growth of MoS₂ takes about several minutes until the precursors MoO₃ have been consumed. Then the growth temperature is increased to 800°C, the growth of NbS₂ is initiated. The second-layer NbS₂ preferably nucleates at the corner of the triangular MoS₂ owing to its defects-rich nature,¹⁰ and then grows inward the central part, finally forms the vertical NbS₂/MoS₂ heterojunction. Worth to mention, this behaviour is different with one-step synthesis of WS₂/MoS₂ vertical heterojunction,⁷ where WS₂ starts the growth from the center of growing MoS₂ on the bottom. This might be ascribed to the lower chemical reactivity of Nb₂O₅ in comparison with WO₃, and thus higher growth temperature. Hence, NbS₂ will grow after MoS₂ finishes the growth while WS₂ starts earlier.

Several interesting phenomena will help to understand this process: (1) NbS₂ may start the nucleation along the edge, which is also defect-rich, and then form a type-I like structure defined in the previous report¹⁰ (**Figure S2c**). This further strengthens that the growth of NbS₂ initiates only after that of MoS₂ brings to an end. (2) Another interesting growth behaviour is the grain boundary crossing, shown in **Figure S2d**, which would not happen in the growth of second-layer MoS₂ between two adjacent MoS₂ triangles. Both support the growth mechanism we propose: The bottom-layer MoS₂ grows first due to the low nucleation temperature, then the second-layer NbS₂ nucleates and grows from the defect-rich corners or edges of the as-grown MoS₂, inward the center, finally forms the M/S vdW heterostructure NbS₂/MoS₂.

In order to further examine the mechanism aforementioned, Raman spectra and mapping were carried out using a 532nm laser. The characteristic E₁, E₂, and A₁ peaks of pure NbS₂, could

Figure 5. Raman spectra and mapping analysis of NbS₂/MoS₂ vdW heterostructure. (a) Optical image of NbS₂/MoS₂ vdW heterostructure, scale bar: 5 μm; (b) Raman spectra taken from pristine NbS₂ (red curve), NbS₂/MoS₂ vdW heterostructure (blue curve) and the exposed MoS₂ (black curve). Raman mapping at 342.7 cm⁻¹ (c) (E₂ mode of NbS₂) and 402.6 cm⁻¹ (d) (A₁ mode of MoS₂) further confirm that NbS₂ is seated at one corner of bottom-layer MoS₂, scale bar: 3 μm.

be found at 306.7 cm⁻¹, 342.7 cm⁻¹, and 377.6 cm⁻¹ (red spectrum in **Figure 5b**), respectively. The two characterized, in-plane E₂ and out-of-plane A₁ vibration modes of pure MoS₂ are located at 382.0 cm⁻¹ and 402.6 cm⁻¹ (black spectrum in **Figure 5b**, the frequency difference is 20.6 cm⁻¹, indicating a monolayer MoS₂⁴⁰), which were collected from the NbS₂ uncovered area of the heterostructure. The blue curve in **Figure 5b** shows the Raman spectrum of the NbS₂/MoS₂ vdW heterostructure, in which all the characteristic peaks from MoS₂ and NbS₂ can be found. One interesting thing should be noted that two adjacent peaks, A₁ peak of NbS₂ and E₂ peak of MoS₂ are overlapped and merged into one peak at 379.2 cm⁻¹ in this spectrum. The

multipeak fitting analysis was performed to figure out the individual peaks (**Figure S7**), in which the A_1 peak of NbS_2 is located at 377.1 cm^{-1} and E_2 peak of MoS_2 is located at 380.1 cm^{-1} . **Figure 5c** and **5d** present the Raman mapping of the area (marked with a square in **Figure 5a**) of the NbS_2/MoS_2 vdW heterostructure. Raman mapping from NbS_2 E_2 peak located at 342.7 cm^{-1} shows the NbS_2 sitting at one corner of MoS_2 , as shown in **Figure 5c**, while Raman mapping collected from MoS_2 A_1 peak located at 402.6 cm^{-1} confirms the single-crystal MoS_2 triangle (**Figure 5d**). These mappings match very well with the optical image in **Figure 5a**. All the Raman tests further consolidate the mechanism we propose above.

3. Conclusion

In conclusion, we report the synthesis of largest area monolayer ($20\text{ }\mu\text{m}$) and few-layer ($60\text{ }\mu\text{m}$) NbS_2 , and for the first time, the preparation of 2D M/S TMDC vdW heterostructure NbS_2/MoS_2 , which is benefited from the contribution of sodium chloride on the reduction of melting point and the improvement of chemical reactivity of Nb_2O_5 . Also, it is the difference in chemical reactivity between NbS_2 and MoS_2 precursors that results in the sequent growth, leading to the formation of this vertical heterostructure. TEM characterization offers firm evidence that shows the high quality, R-stacking order and clean interface of the obtained NbS_2/MoS_2 heterostructure. We believe this work could be developed into the syntheses of other 2D TMDC M/S heterostructures and will accelerate the research in the TMDC M/S interface.

4. Methods

4.1 Synthesis and characterization of large-area layered NbS_2

Atmospheric Pressure CVD Chamber is schematically depicted in **Figure 1a**, in which, detailly, the heating setup is a Lindberg/Blue M Mini-Mite tube furnace (TF55035C-1, digital, single segment), and the fused quartz tube is 1000 mm long with an Outside Diameter (O.D.) of 1 inch. Mixed powders of Nb_2O_5 and NaCl with a ratio around 9:1 were used as metal precursor,

placed in an alumina crucible and located in the zone where temperature was ramped to 800 °C in 16 min and hold for 10 min, while sulfur feedstock was placed at the upstream of the mixture with temperature ranging from 120 °C to 200 °C. Under the carrier gas of 60~80 sccm Ar, sulfur vapor was flowed in and reacted with metal mixture, and then layered NbS₂ will be deposited on the substrate (285nm SiO₂/Si), which was placed 3~5 mm above the metal precursor with the polished side facing down. After the reaction is finished, the furnace was naturally cooled down to room temperature.

4.2 Synthesis of NbS₂/MoS₂ and time regulation

We used the same growth temperature (800 °C), gas flow rate (60 sccm Ar), and CVD setup for the synthesis of NbS₂/MoS₂. To first grow MoS₂, MoO₃ powder was put at the upper stream of the metal mixture of Nb₂O₅ and NaCl with a distance of 1.5 cm (**Figure S1**). Unlike the control of thickness via gas flow rate in intrinsic NbS₂, we regulate the holding time at the temperature of 800 °C ranging from 6 min to 12 min, to achieve different coverage of NbS₂ on MoS₂.

4.3 XPS, SEM and Raman characterizations

XPS tests were carried out via a PHI Quantera II spectrometer with a monochromatic Al-K α radiation ($h\nu = 1486.6$ eV), calibrated with C 1s core level (binding energy: 284.8 eV). SEM characterization was performed under the JOEL JSM-7600F system with an accelerating voltage of 15 kV. Raman spectra were recorded by a Witec system equipped with 100x objective lens and a grating of 1800 lines/mm under 532 nm laser excitation.

4.4 Transfer and STEM characterization and image simulation

The TEM samples were prepared with a poly (methyl methacrylate) (PMMA) based transfer method. First, the Si wafer with the as-grown CVD samples were spin-coated with PMMA at 3,000 r.p.m. for 60 s and then baked at 150 °C for 3 min. Afterward, PMMA-coated sample was peeled off by etching away the underneath SiO₂ in 1% HF solution. Next, the floating PMMA film was transferred to the distilled water and scooped up by a TEM grid. The TEM grid was

heated at 50 °C for 5 hours, and then submerged into acetone for about 24 h to remove PMMA. Finally, the TEM grid was rinsed by isopropyl alcohol (IPA) and dried in air.

ADF-STEM images were obtained using a probe-corrected Titan ChemiSTEM (FEI, USA) which was operated at an accelerating voltage of 200 kV. The convergent semi-angle for illumination was set to be about 21.4 mrad with a probe current of 79 pA. The collection angle is about 43.4 to 200 mrad for ADF-STEM imaging. The atomic-resolution ADF-STEM images were processed with an improved Wiener filtering method to partly remove the background noise. STEM–ADF image simulations were carried out using QSTEM⁴¹ software, with the input parameters being set according to the experimental conditions. False-color images were performed by ImageJ to improve visual inspection.

Supporting Information

This material is available free of charge via the Internet at <http://pubs.acs.org>.

Supporting Information:

Figure S1: CVD setup for the NbS₂/MoS₂ synthesis;

Figure S2: OM of NbS₂/MoS₂ heterostructure with different morphologies;

Figure S3: Intensity line profiles of few-layer NbS₂/MoS₂ sample;

Figure S4: Atomic structure of NbS₂ and MoS₂ heterostructure;

Figure S5: Comparison of the STEM experimental image with simulation images;

Figure S6: Quantitative analysis of STEM intensity profiles;

Figure S7: Multipeak fitting analysis of the Raman peak.

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Q. F. and X. W. contributed equally to this work. The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. Q. F. and J. Z. proposed the idea and Q.F. prepared the materials. J. X. performed the Raman spectra and mapping tests. Q. Z. helped to obtain the SEM images. X. W. and D. L. conducted the STEM characterization. C. Z. and X. W. were involved in other characterizations including XPS. Q. F. and X. W. analyzed the data and composed the manuscript under the instruction of F. L., Z. S., C. J. and Z. L..

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