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Temperature-Dependent Phonon Scattering and Photoluminescence in Vertical MoS₂/WSe₂ Heterostructures

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Abstract: Transition metal dichalcogenide (TMD) monolayers and their heterostructures have attracted considerable attention due to their distinct properties. In this work, we performed a systematic investigation of MoS₂/WSe₂ heterostructures, focusing on their temperature-dependent Raman and photoluminescence (PL) characteristics in the range of 79 to 473 K. Our Raman analysis revealed that both the longitudinal and transverse modes of the heterostructure exhibit linear shifts towards low frequencies with increasing temperatures. The peak position and intensity of PL spectra also showed pronounced temperature dependency. The activation energy of thermal-quenching-induced PL emissions was estimated as 61.5 meV and 82.6 meV for WSe₂ and MoS₂, respectively. Additionally, we observed that the spectral full width at half maximum (FWHM) of Raman and PL peaks increases as the temperature increases, and these broadenings can be attributed to the phonon interaction and the expansion of the heterostructure's thermal coefficients. This work provides valuable insights into the interlayer coupling of van der Waals heterostructures, which is essential for understanding their potential applications in extreme temperatures.

Keywords: transition metal dichalcogenides; heterostructures; phonon scattering; thermal quenching; thermal activation energy



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1. Introduction

Transition metal dichalcogenides have recently garnered significant interest due to their scalability and thickness-dependent electrical and optical properties [1]. The evolution of bulk TMDs to atomically thin 2D layered structures is characterized by their transition from indirect to direct bandgap semiconductors [2]. Such a transition of bandgaps has significant implications for their optoelectronic properties and potential applications [3–6]. These 2D semiconducting materials, MX₂ (M = Mo, W; X = S, Se), are exemplified by compounds such as molybdenum disulfide (MoS₂) and tungsten diselenide (WSe₂) with direct bandgaps of 1.9 eV and 1.6 eV, respectively [7]. Beyond monolayers, vertically-stacked heterostructures based on the van der Waals force provide a fascinating platform for investigating novel physical phenomena [8]. With the assistance of an artificial stacking arrangement, electronic band engineering of heterostructures can be achieved, resulting in the modulation of optical properties via interlayer coupling effects [9].

Heterostructures composed of two-dimensional materials have been shown to exhibit various intriguing phenomena, including ultrafast charge transfer [10], high PL quantum yield [11], and interlayer valley physics [12]. These properties make them potential candidates for applications in field-effect transistors [13], electrocatalysis [14], and photodetectors [15]. The thermodynamic stability of these heterostructures plays a critical role in their optoelectronic properties [16]. A comprehensive understanding of the correlation between phonon scattering and heterostructure interfacial coupling is imperative to achieve the optimal performance of such structures [17,18]. Raman spectroscopy is an essential

tool to investigate the interaction between electrons and phonons in 2D materials and their heterostructures [19]. Research on temperature-dependent Raman spectroscopy of few-layered WS₂ provides valuable insights into their intrinsic phonon scattering processes and thermal properties [20]. Several early experiments investigated the impact of temperature on the phononic properties of few-layered TMDs, including MoS₂, WS₂, MoSe₂, and others [21]. These studies have demonstrated that temperature-dependent behaviors of phononic modes in these nanosheets are attributed to the effects of thermal expansion and anharmonic resonance [22]. Moreover, some studies investigated the impact of interlayer coupling on the Raman spectra of TMDs heterostructures, which demonstrated a strong dependence on the stacking orientation of the constituent monolayers [23,24].

This work provides a systematic investigation into the temperature-dependent Raman characteristics of a heterostructure composed of MoS₂ and WSe₂ monolayers. Raman spectroscopy measurements were conducted over the controlled temperature range of 79 to 473 K to investigate the in-plane (E_{2g}^1) and out-of-plane (A_{1g}) vibrational modes of the heterostructure. Experimental observations indicate that both Raman modes display a nearly linear dependence across the entire temperature range, alongside a commensurate variation in their full width at half maximum. The primary focus of this study is on attempting to understand the variation of interlayer interactions, particularly temperature-dependent phonon interactions, and the thermal expansion coefficients of the heterostructure under high vacuum conditions. Furthermore, temperature-dependent photoluminescence was also observed and utilized to determine the activation energy of the PL emissions caused by thermal quenching. This work provides a platform to investigate the photodynamics of low-dimensional materials and also helps to understand the electronic and photonic properties of van der Waals heterostructures.

2. Materials and Methods

The synthesis of 2D transition metal dichalcogenides has revolutionized modern techniques, such as physical vapor deposition (PVD) [25], chemical vapor deposition (CVD) [26], electrodeposition [27], and thermal synthesis [28], facilitating their advancement and enhancing their capabilities for diverse applications. In this work, a MoS₂ monolayer was grown by a chemical vapor deposition method using sulfur (S) and molybdenum oxide (MoO₃) powders. MoO₃ and S powder were put in separate boats at the center of a fused quartz tube located in a furnace. Subsequently, a SiO₂/Si wafer was suspended on one of the boats, and the temperature was ramped up to 810 °C for 30 min and maintained for 10 min. When it cooled down to room temperature, triangular-shaped MoS₂ monolayers were successfully prepared. Further details can be found in our previous work [29]. The synthesis procedure was conducted at atmospheric pressure using Ar gas as a carrier agent with a flow rate of 70 sccm. The same CVD procedure was employed to grow WSe₂ utilizing tungsten oxide (WO₃) and selenium (Se) powder as source materials, with the furnace temperature kept constant at 950 °C and naturally allowed to cool. A comprehensive description of the synthesis procedures and a schematic depiction of the CVD setup can be found in Supplementary Section S1. A simple wet transfer technique was employed to prepare the MoS₂/WSe₂ heterostructures [30]. Further details about the transfer process and the schematic illustration can be found in Supplementary Section S2. The heterostructure underwent annealing in a vacuum furnace at 70 °C for 12 h to eliminate water molecules and promote better crystallinity.

For spectral measurements of materials, PL and Raman spectra were analyzed using an iHR550 Raman spectrometer from Horiba with a laser excitation of 532 nm. An objective lens providing 50 times magnification was utilized to enable a more detailed analysis of the sample's features. The diameter of the laser spot was about 1 μm. The spectrometer was equipped with gratings of 300 g mm^{−1} and 1200 g mm^{−1} to facilitate accurate analysis of the spectral data. Furthermore, a temperature-controlled cryostat (THMS-600, Linkam Scientific Instruments Ltd., Redhill, UK) was utilized to investigate the temperature-dependent behavior of the heterostructure (Supplementary Section S3).

The cryostat utilized liquid nitrogen to attain the lowest temperature of the boiling point of liquid nitrogen. Samples were loaded into the cryostat full of nitrogen to avoid any impact from the surface adsorption of gas molecules and to enable precise temperature control during measurements.

3. Results and Discussion

Figure 1a shows the schematic view of the MoS₂/WSe₂ heterostructure. The MoS₂ and WSe₂ monolayers were in a 2H phase, behaving as semiconductors with direct band gaps. Figure 1b shows the scanning electron microscopy (SEM) image of the as-prepared MoS₂/WSe₂ heterostructure, where the top WSe₂ monolayer has been transferred onto the bottom MoS₂ monolayer. The scale bar is 10 μ m. Figure 1c shows the optical image of heterostructures, where monolayer and heterostructure regions can be observed. The monolayer thicknesses of MoS₂ (region 1) and WSe₂ (region 2) were determined to be 0.8 nm and 0.76 nm, respectively, using atomic force microscopy (AFM) as described in Supplementary Section S1. The height profiles of both monolayer regions are plotted in Figure 1d. Additional AFM results of the heterostructure (region 3) with a 0.39 nm interlayer distance can be found in Supplementary Section S1.

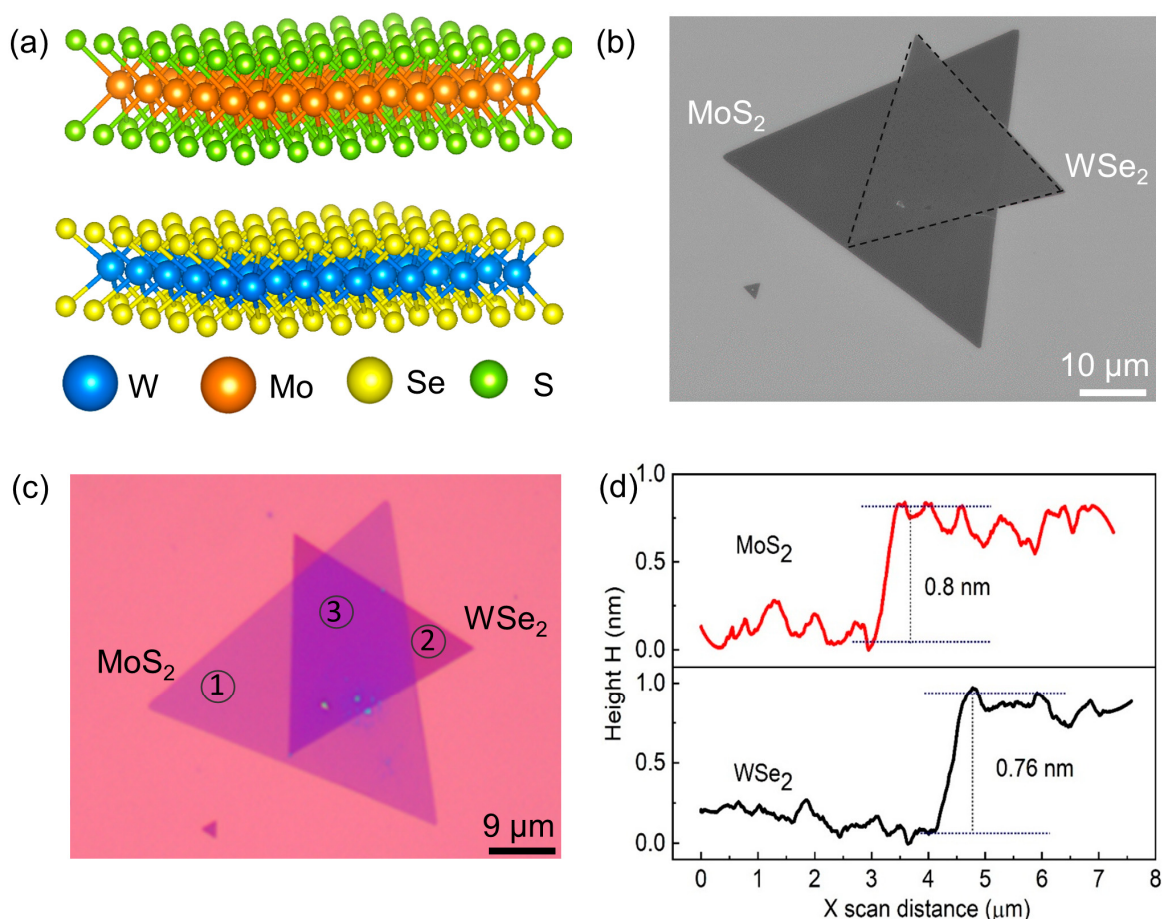


Figure 1. Structure and morphology of MoS₂/WSe₂ heterostructure. (a) Schematic of MoS₂ and WSe₂ monolayers. (b) SEM and (c) optical images of the vertically stacked heterostructure with scale bars of 10 and 9 μ m, respectively. (d) Height profiles of MoS₂ and WSe₂ monolayers confirmed by AFM.

Raman spectroscopy was employed to characterize the structure and spectral properties of the MoS₂/WSe₂ heterostructure. The sample was put into a vacuum annealing furnace to anneal the heterostructure sample and mitigate the influence of defects and gas adsorption (Supplementary Section S2). Moreover, to avoid any local heating of the sample, which can alter peak positions in the Raman spectra, the excitation power was

carefully controlled at 1.2 mW. The Raman spectra in Figure 2a display two distinct peaks corresponding to the longitudinal (E) and transverse (A) modes of monolayer regions in the $\text{MoS}_2/\text{WSe}_2$ heterostructure. The Raman modes of E_{2g}^1 and A_{1g} for MoS_2 (red curve) show a separation of about 18 cm^{-1} , which is consistent with previously reported works on monolayer MoS_2 [31].

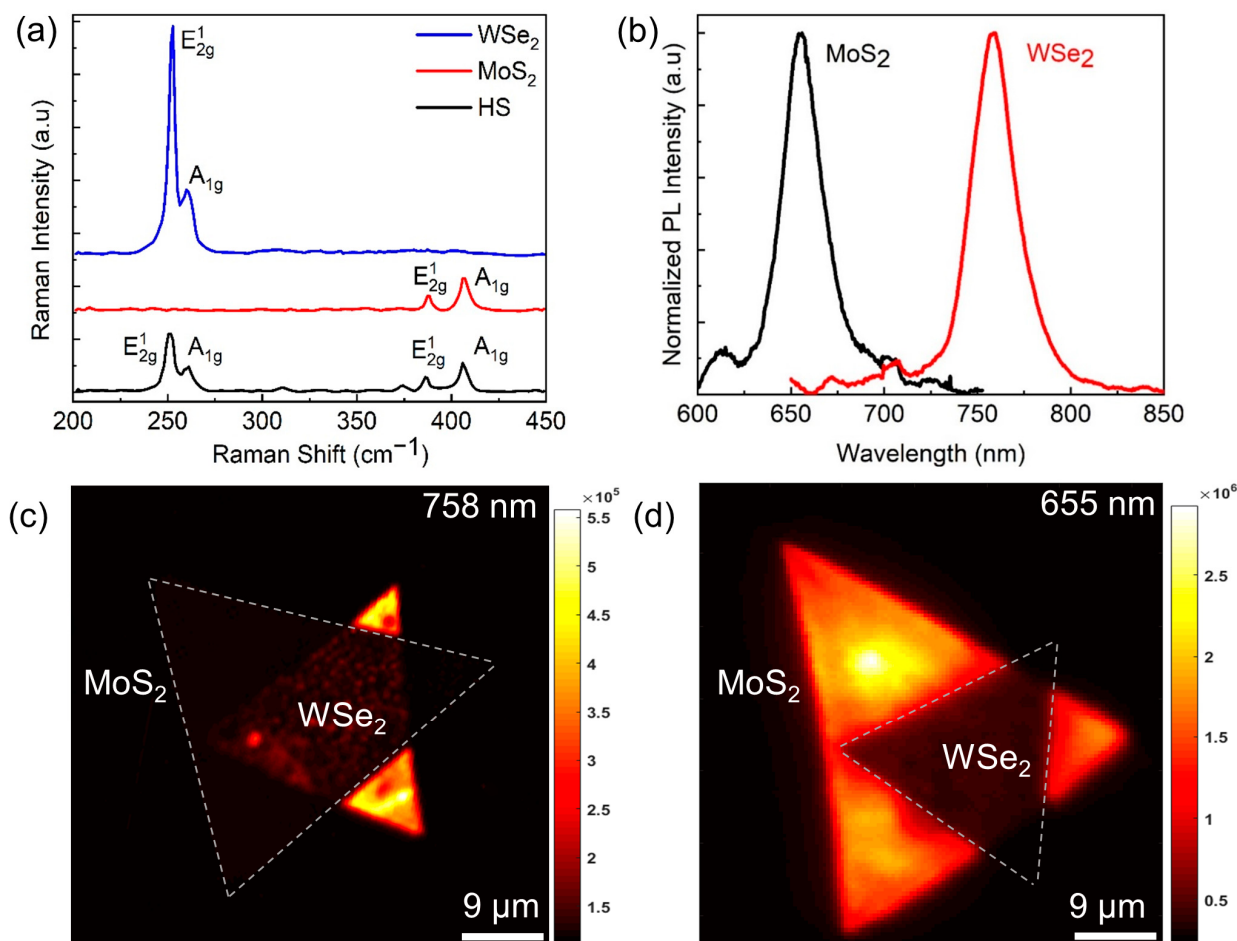


Figure 2. Raman and PL spectra of $\text{MoS}_2/\text{WSe}_2$ heterostructures. (a) Raman shifts of MoS_2 and WSe_2 monolayers and their heterostructures. (b) PL spectra of MoS_2 and WSe_2 monolayers with the main peaks at 655 nm and 758 nm, respectively. Mapping images of PL intensity of heterostructure samples at a wavelength of (c) 758 nm and (d) 655 nm.

The transverse and longitudinal modes were detected at 252 cm^{-1} and 260 cm^{-1} , respectively, in the monolayer WSe_2 region of the heterostructure, consistent with previous studies [32]. The Raman spectra acquired from the heterostructure region (black curve) shown in Figure 2a exhibit identical vibrational modes, as observed in both monolayers. Nevertheless, a marginal reduction is observed in the intensity of these modes, which is attributable to enhanced excitation light scattering caused by the presence of the heterostructure. The $\text{MoS}_2/\text{WSe}_2$ heterostructure manifests a type II band alignment, which facilitates effective charge transfer, yet concurrently reduces the PL intensity by spatially separating the valence and conduction bands. The photoluminescence peaks exhibited by the heterostructure in Figure 2b are attributed to the “A” excitons, corresponding to the direct bandgap transitions at the K-point of the Brillouin zone in both MoS_2 and WSe_2 . For the MoS_2 spectrum, an additional peak at higher energy near 2.02 eV was observed, corresponding to the “B” exciton formed due to transitions between the spin-orbit split valence band and the conduction band [33]. However, the emission strength of the B exciton

is relatively weaker than the A exciton due to its lower oscillator strength, which resulted from an indirect transition.

PL mapping was performed at specific wavelengths to assess the photoemissions of MoS₂/WSe₂ heterostructures. As illustrated in Figure 2c,d, the resulting PL emission maps at 758 nm and 655 nm provide valuable insight into the distribution of PL intensity from MoS₂ and WSe₂, respectively, in the heterostructure. Remarkably, the photoluminescence maps obtained in this study demonstrate a stronger emission signal emanating from the MoS₂ monolayer, which can be ascribed to its elevated radiative recombination rate and binding energy. Importantly, these findings align with previous investigations of similar systems [34]. Moreover, photoluminescence measurements were conducted for both MoS₂ and WSe₂ in the monolayer position as well as the heterostructure region. Notably, the peak intensities at 655 nm and 758 nm in the heterostructure's region 3 exhibit significant quenching compared to the monolayer regions (1 and 2). This behavior can be attributed to the ultrafast charge transfer phenomenon observed in atomically thin MoS₂/WSe₂ heterostructures. Detailed discussions can be found in Supplementary Section S4 for a more in-depth analysis of these findings. All Raman and PL spectra in Figure 2 were measured at room temperature using a 532 nm laser of 1.2 mW.

To better comprehend the vibrational modes and interlayer coupling in MoS₂/WSe₂ heterostructures, temperature-dependent Raman measurements ranging from 79 to 473 K, as illustrated in Figure 3a,b, enable insights into their thermal behavior. The Raman findings indicate strong scattering intensities of the E_{2g}^1 and A_{1g} modes for MoS₂/WSe₂ heterostructures with increasing temperatures. Thermal expansion of the lattice is known to trigger a decline in the vibrational frequency with increasing temperature, thereby inducing a downward shift of the Raman modes towards lower frequencies, as corroborated by the outcomes shown in Figure 3a,b. Remarkably, the uniform redshift across all Raman modes with increasing temperature implies a systematic alteration in the vibrational characteristics of the heterostructure. At high temperatures, it looks as though split peaks appear apart from the main peaks, but actually, we are observing the temperature-dependent enhancement and widening of inconspicuous peaks at low temperatures. To visualize these changes, Figure 3c illustrates the temperature-dependent peak positions of the Raman modes. Our experimental data exhibit a linear decline with temperature and are well-fitted by a linear function.

Elevated temperatures lead to increased atomic vibrations in the lattices of WSe₂ and MoS₂, which causes an increase in the scattering rate and a reduction in the lifetime of vibrational modes. These effects contribute to the broadening of the Raman spectra, as demonstrated by the increase in full width at half maximum (FWHM) observed in Figure 3d. The FWHM of the WSe₂ region in the heterostructure is particularly significant, perhaps due to its lower binding energy, which leads to the higher scattering of lattice modes at higher temperatures (indicated in Figure 3d). When the temperature rises, the thermal energy of the lattice vibrations increases, resulting in an increased number of phonon scattering events. In addition, the thermal energy of the lattice vibrations in 2D materials also increases, resulting in a greater number of phonon scattering events. This, in turn, leads to an increase in the FWHM of the Raman peak associated with the A_{1g} mode. In MoS₂, this effect is more pronounced for the A_{1g} mode compared to the E_{2g}^1 mode due to the latter's less sensitive in-plane vibrations of Mo and S atoms, as illustrated in Figure 3d. It is noteworthy that slight deviations observed at certain temperatures may be attributed to slight instrumentation instability in our Raman setup. The differences in Raman peak broadening between the E_{2g}^1 and A_{1g} modes of WSe₂ and MoS₂ provide valuable information about the behavior of these materials under varying thermal conditions.

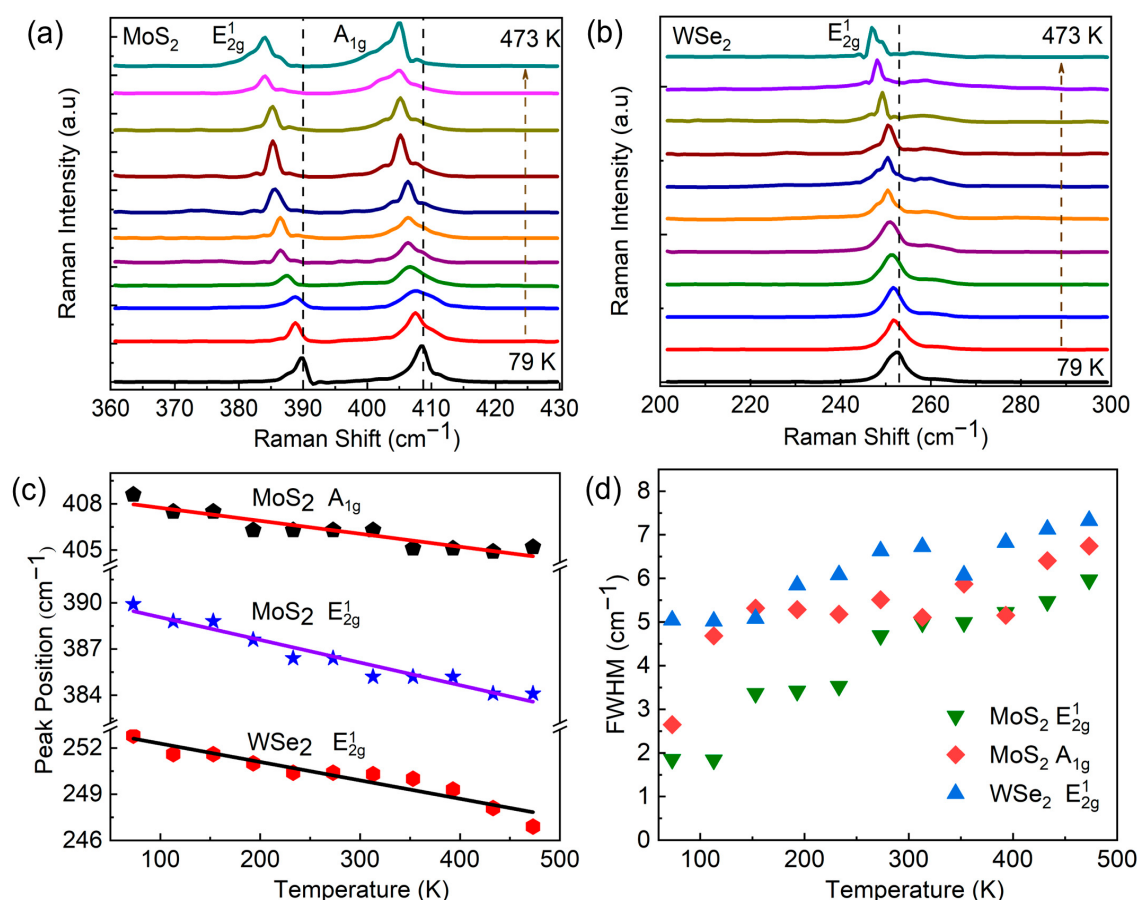


Figure 3. Temperature-dependent Raman spectra of MoS₂/WSe₂ heterostructures. (a,b) The variation of Raman modes of MoS₂ and WSe₂ at varying temperatures of 79–473 K. The vertically black and orange dashed lines are drawn to highlight specific Raman modes and indicate temperature increase, respectively. (c) Temperature-induced Raman shifts of three characteristic peaks showing linear fitting curves. (d) The full width at half maximum of Raman peaks at different temperatures.

In addition to its effects on the Raman spectra, temperature can also influence the photon emissions of the MoS₂/WSe₂ heterostructures. The aspect of temperature-dependent photoluminescence in heterostructures is crucial in device physics to improve their operation performance. The combined effect of thermal and optical energies that contribute to the de-trapping of carriers and desorption of adsorbates (such as O₂ and H₂O) could result in changes in the conductivity of the material, as well as the optical emission properties, as discussed elsewhere [35]. In the case of heterostructures, temperature-dependent PL spectroscopy has revealed new and interesting effects, including an anomalous redshift, which could be related to thermal quenching, defect engineering, and strain. Several studies have explored and linked the temperature-dependent PL spectra to the underlying band structure, electronic properties, and potential device applications of heterostructures. To investigate thermal quenching and the underlying band structure, photoluminescence measurements of the heterostructure were conducted over a range of temperatures (79 K to 473 K), as illustrated in Figure 4. The results of the photoluminescence measurements reveal a significant temperature dependence in both the peak position and intensity of the spectrum. As the temperature increases, both the WSe₂ and MoS₂ peaks experience redshifts towards longer wavelengths, as demonstrated in Figure 4a,b. The reduction in photoluminescence intensity with increasing temperature is a well-known phenomenon observed in both two-dimensional transition metal dichalcogenides and bulk semiconductors, attributable to the amplification of non-radiative recombination, thermal activation of electrons and holes, and phonon-assisted recombination.

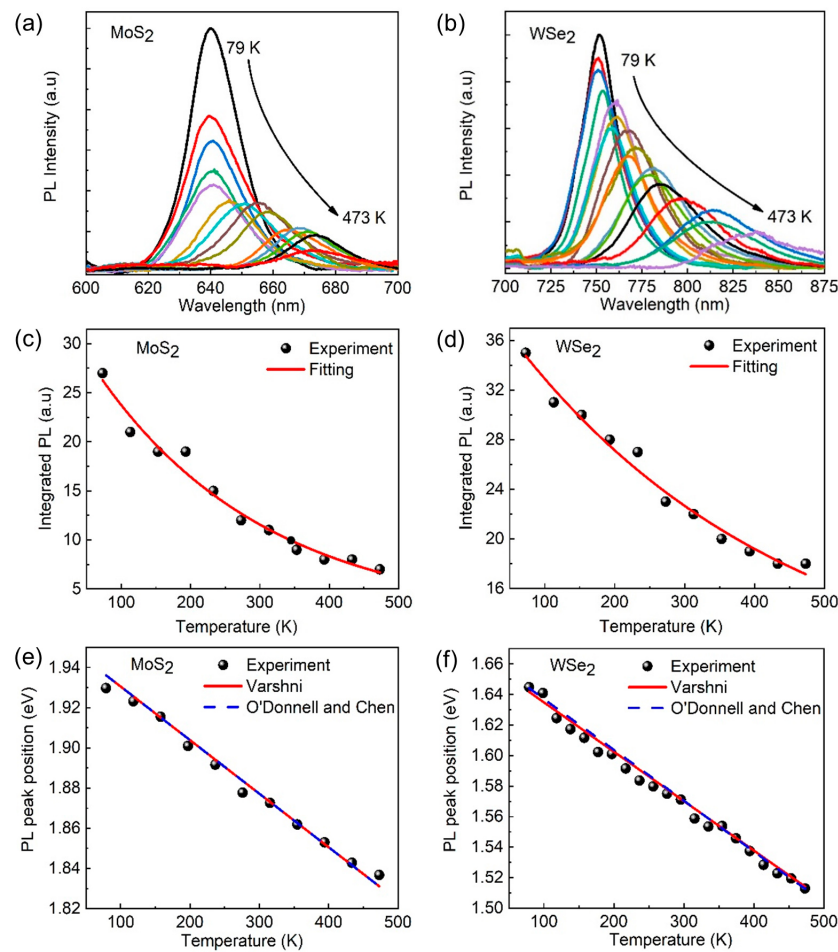


Figure 4. Temperature-dependent photoluminescence. (a,b) PL spectra of MoS₂ and WSe₂ at various temperatures. The peak position shifts red, and the intensity decreases as temperature increases from 79 to 473 K. (c,d) Integrated PL intensities of corresponding regions of MoS₂ and WSe₂, respectively. Solid lines depict experimental data, while the scattered points show the corresponding Arrhenius fitting. (e,f) Temperature-dependent shifting of PL peaks detected for MoS₂ and WSe₂ regions in the heterostructures. Red lines and dotted blue lines represent fitting curves for Equations (2) and (3).

Here, the investigation of the temperature dependence of photoluminescence intensity in MoS₂/WSe₂ heterostructures revealed that the Arrhenius formula (Equation (1)) provides a good fit for the experimental data. This formula correlates the photoluminescence quenching rate with the thermal activation energy for non-radiative recombination, as demonstrated in Figure 4c,d. These findings offer insights into the fundamental processes involved in optoelectronic devices and emphasize the significance of carefully controlling temperature effects in such systems [36].

$$I(T) = \frac{A}{1 + C e^{(-E/k_B T)}} \quad (1)$$

Here, in Equation (1), the temperature-dependent PL intensity (I) is the main variable, along with the density of quenching centers (A) and the strength of electron–phonon coupling (C). The thermal quenching process is characterized by the thermal activation energy (E), and the Boltzmann constant (k_B) is utilized in the formula to relate temperature and energy. The thermal activation energies of MoS₂ (82.6 meV) and WSe₂ (61.5 meV) were determined via Arrhenius fitting of the experimental data presented in Figure 4c,d. The two materials exhibit different thermal activation energies, E , due to differences in the bandgap, effective mass, and mechanical strain. The distinct crystal structures and chemical bond-

ing may also contribute to different effective masses in the MoS₂/WSe₂ heterostructure, resulting in varying carrier mobilities and activation energies. The observed differences in the thermal activation energies between MoS₂ and WSe₂ can ultimately be attributed to material-specific factors, including band structures, carrier dynamics, and mechanical forces. As shown in Figure 4e,f, the photoluminescence peak position, corresponding to the direct excitonic transition energy in MoS₂/WSe₂, is affected by temperature variations. An increasing temperature causes a redshift in the PL peak, indicating a reduction in the energy gap of the heterostructure [37]. This energy gap refers to the energy of the lowest allowable exciton state, which corresponds to the energy required to create an electron–hole pair in the system. The decreased energy gap shifts the lowest allowable exciton state to lower energy. This effect is commonly observed in various materials, including semiconductors and insulators. Density functional theory (DFT) calculations are performed in Supplementary Section S4. The relationship between temperature and the PL peak position is typically described by empirical equations, such as the one proposed by Varshni [38], given in Equation (2):

$$E_g(T) = E_g^0 + \alpha \frac{T^2}{T + \beta} \quad (2)$$

In this equation, E_g^0 represents the band gap at a temperature of absolute zero, where $T = 0$ K; β is a constant approximating the Debye temperature of the material; and α is the coefficient representing the change in bandgap energy with temperature. The Varshni fitting shown in Figure 4e,f provides a good agreement with the experimental data, yielding bandgap values of $E_0 = 1.94$ eV and 1.64 eV for MoS₂ and WSe₂, respectively. In the same way, the O'Donnell and Chen formula, as given in Equation (3), is another model to fit the temperature-dependent PL and obtain the bandgap of the material [39].

$$E_g = E_g^0 - S < \frac{h}{2\pi} > \left[\cot \left(\frac{h\omega}{4\pi k_B T} \right) - 1 \right] \quad (3)$$

Here, the parameter E_g^0 gives the bandgap at zero temperature, while S gives information on electron–phonon coupling, and ω is the photon frequency. The fitting curves in Figure 4e,f for both MoS₂ and WSe₂ peak position shifting versus temperature are well-matched to the experimental data. The band gap values obtained using the O'Donnell and Chen formula for MoS₂ and WSe₂ are 1.93 eV and 1.65 eV, respectively, which are consistent with the results from the Varshni fitting, as well as with previously reported observations [40]. These temperature-dependent observations of phonon scattering and the interlayer coupling of heterostructures provide a significant influence on the performance of heterostructure devices; for example, the thermal noise is serious in photodetectors, and the carrier mobility of transistors is highly influenced by the environmental temperature.

4. Conclusions

Our study provides important insights into the temperature-dependent photoluminescence and bandgap characteristics of the MoS₂/WSe₂ heterostructure. These findings expand our understanding of this material system and have potential implications for various technological applications. We observed that both the longitudinal and transverse modes of MoS₂ and WSe₂ exhibit a linear variation in peak position and full width at half maximum with temperature. The quenching of the photoluminescence in the heterostructure is attributed to non-radiative recombination mechanisms. The behavior of the photoluminescence with temperature is similar to that observed in bulk MoS₂ and WSe₂. Furthermore, we found that differences in the bandgap, effective mass, and mechanical strain of MoS₂ and WSe₂ lead to different thermal activation energies, with values of 82.6 meV and 61.5 meV, respectively. Overall, our study contributes to the existing knowledge based on MoS₂/WSe₂ heterostructures and provides important insights into their temperature-dependent optoelectronic properties, which can enlighten the design and optimization of various devices.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/nano13162349/s1>: Section S1. CVD preparation of MoS₂ and WSe₂ and their corresponding AFM analysis. Section S2. Preparation of MoS₂/WSe₂ heterostructure. Section S3. Experimental setup of Raman and PL spectroscopy. Section S4. PL Measurement and Details of the DFT Calculation of MoS₂/WSe₂ Hetrostructure. References [41–50] are cited in the Supplementary Materials.

Author Contributions: W.A. and M.H. prepared the samples. W.A. and Y.X. performed the spectral experiments. W.A. and Y.L. analyzed and processed experiment data. W.A. and Z.L. designed the experiment and wrote the manuscript. Y.L. and Z.L. offered the funding. W.A. and Y.L. contributed equally as the first authors. All authors have read and agreed to the published version of the manuscript.

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