Exciton and Trion Dynamics in Bilayer MoS$_2$

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2D transition metal dichalcogenide (TMD) semiconductors$^{[1–10]}$ such as molybdenum disulphide (MoS$_2$), have attracted tremendous attentions owing to their unique properties, such as strong interactions with light$^{[1–4]}$, layer-dependent energy gaps$^{[5,6]}$, electrically tunable exciton dynamics$^{[7,8]}$, tightly bound trions$^{[9,10]}$, and so on. The electronic band structure of MoS$_2$ strongly depends on the layer number and layer-stacking sequences$^{[11–13]}$. Especially, monolayer MoS$_2$ owns the most distinct properties comparing to few-layer counterparts, as there is no interlayer interaction and reduced screening effect$^{[14]}$ for few-layer TMDs, interlayer interaction, screening effect, quantum confinement, and crystal symmetry jointly determine their electronic structures$^{[11–13]}$ which gives rise to direct band gap emerging in monolayer MoS$_2$ and indirect band gap in few-layer MoS$_2$ at room temperature$^{[15]}$. Most of previous studies and findings are limited to monolayers$^{[6,8,9]}$. However, few-layer structures, particularly bilayer structure, are extremely important, since they offer us unique platforms to investigate the fundamental phenomena arising from the interlayer van der Waals interactions, which can enable many new optoelectronic devices based on heterostructures$^{[16]}$. Owing to the direct band nature, the photoluminescence (PL) intensity from monolayer MoS$_2$ can be electrically tuned by up to two orders of magnitude, which enables the control of exciton and trion dynamics in monolayer MoS$_2$ at room temperature;$^{[6,9]}$ in contrast, the PL spectra from bilayer MoS$_2$ could not be tuned by electric field at room temperature owing to its indirect band gap manner,$^{[7]}$ which makes the exciton and trion dynamics in bilayer MoS$_2$ still underexplored.

In this paper, we demonstrate the valley control of exciton and trion dynamics in bilayer MoS$_2$, via the comodulations by both temperature and electric field. We found that as temperature decreases from 300 to 100 K, the valley of the conduction band at Λ point (named as Λ valley) moves down relatively to the valley at K point (named as K valley) in monolayer MoS$_2$, while the Λ valley rises up relatively to the K valley in bilayer MoS$_2$ (Figure 1). This opposite temperature dependence of the valley movements in mono- and bilayer MoS$_2$ can significantly change the photocarrier relaxation pathways in their PL processes, which leads to more than twice faster increasing of the measured PL intensity from bilayer MoS$_2$ than that from monolayer MoS$_2$ as temperature decreases. More importantly, the rising up of the Λ valley in bilayer MoS$_2$ at low temperature offers the electrical tunability of the K–Λ direct PL transition, enabling the exploration of the exciton and trion dynamics in bilayer MoS$_2$. The trion binding energy of bilayer MoS$_2$ was firstly measured to be 27 meV at 83 K, which is smaller than the measured trion binding energy of 39 meV in monolayer MoS$_2$. Our findings provide insight into exciton and trion dynamics in bilayer MoS$_2$ and enable new applications in photonics and optoelectronics.$^{[1,2,17]}$ Moreover, the comodulation technique by both temperature and electric field provides a novel method to explore the fundamental phenomena in few-layer 2D semiconductors.

We calculated the band structures of mono- and bilayer MoS$_2$ at various temperatures (Figure 1) within density functional theory (DFT) molecular dynamics using Perdew–Wang (PW) generalized gradient approximation (GGA) based on a real-space numerical atomic orbital code.$^{[18]}$ From the simulation results, as temperature decreases from 300 to 100 K, the Γ peak in the valence band (named as Γ peak) of 2L MoS$_2$ significantly moves down relative to the K peak, which drives the indirect band structure of bilayer MoS$_2$ at room temperature approaching direct band structure at the temperature range of 50–250 K (Figure 1 and Figure S1, Supporting Information). Meanwhile, Λ valley moves down relatively to K valley in monolayer MoS$_2$, while Λ valley moves up relatively to K valley in bilayer MoS$_2$ (Figure 1a,c), which drives the direct band structure of monolayer MoS$_2$ at room temperature approaching indirect band structure at the temperature range of 50–210 K (Figure 1 and Figures S1...
This opposite movement of the valleys with temperature can also significantly change the photocarrier relaxation pathways in mono- and bilayer MoS\(_2\),\(^{17}\) since photoexcited electrons and holes will always prefer the low energy states. Hence, the weight of the photoelectrons relaxed into the K valley will decrease in monolayer MoS\(_2\) as temperature decreases (Figure 1b), while conversely in bilayer MoS\(_2\), more portion of photoelectrons will relax into K valley at lower temperature (Figure 1d).

Similarly, the holes in valence band follow the same rules as shown in Figure 1. In order to investigate this opposite temperature dependence of the photocarrier pathways, we carried out temperature dependent PL measurements on both mono- and bilayer MoS\(_2\) samples as a comparison.

**Figure 1.** Calculated band structures of mono- and bilayer MoS\(_2\) and the schematic of their photocarrier relaxation pathways at 100 and 300 K. a,c) Band structure of monolayer (labeled as “1L”) and bilayer (labeled as “2L”) MoS\(_2\). The solid black arrows indicate the moving directions of Λ valley and Γ peak as temperature decreases (K point is fixed). b,d) Schematic of the photocarrier relaxation pathways in 1L (b) and 2L (d) MoS\(_2\). The orange and green lines indicate the VBM and CBM, respectively. The green circle “e” stands for electrons and orange circle “h” stands for holes. The dashed and solid lines present the situations at 100 and 300 K, respectively.

and S2, Supporting Information). This opposite movement of the valleys with temperature can also significantly change the photocarrier relaxation pathways in mono- and bilayer MoS\(_2\),\(^{17}\) since photoexcited electrons and holes will always prefer the low energy states. Hence, the weight of the photoelectrons relaxed into the K valley will decrease in monolayer MoS\(_2\) as temperature decreases (Figure 1b), while conversely in bilayer MoS\(_2\), more portion of photoelectrons will relax into K valley at lower temperature (Figure 1d). Similarly, the holes in valence band follow the same rules as shown in Figure 1. In order to investigate this opposite temperature dependence of the photocarrier pathways, we carried out temperature dependent PL measurements on both mono- and bilayer MoS\(_2\) samples as a comparison.

**Figure 2** shows the results of temperature dependent PL measurements from mono- and bilayer MoS\(_2\) samples. The low temperature PL measurements were carried out with a Horiba Yvon T64000 micro-Raman/PL system equipped with a Linkam liquid nitrogen chamber, using a 532 nm green laser for excitation. In the experiment, the low temperature chamber was cooled down from room temperature (298 K) to near liquid nitrogen temperature (83 K), with a step of 30 K. It is clear to see that the PL intensity increases with the decrease of temperature for both mono- and bilayer samples, which are due to the suppressed nonradiative decays at low temperature.\(^{17}\) The PL peak location shows a blue shift with the decrease of temperature, which can be explained by the Varshni relation.\(^{19}\) Yet the increasing PL intensity for mono- and bilayer MoS\(_2\) shows different trends. From 298 to 233 K, the PL intensity of monolayer MoS\(_2\) is stronger than that of the bilayer MoS\(_2\) sample, owing to the direct band gap in monolayer MoS\(_2\) and the indirect band gap in bilayer MoS\(_2\) at this relatively high temperature range. However, from 203 K, the PL intensity from bilayer MoS\(_2\) surpasses that from monolayer and reaches almost twice the intensity from the monolayer MoS\(_2\) sample at 83 K, as indicated in Figure 2c. A similar trend was also observed from another batch of mono- and bilayer MoS\(_2\) samples (Figure S3, Supporting Information). At a low temperature of 83 K, the much faster rising of the PL intensity from bilayer MoS\(_2\) compared to monolayer can be explained with the tuning of band structure with decreasing temperature. More specifically, the K–K carrier recombination pathway is suppressed with the moving down of Λ valley in monolayer MoS\(_2\), while the K–K carrier recombination pathway is strengthened with the rising up of the Λ valley in bilayer MoS\(_2\).

In contrast to an exciton, a trion (charged exciton) has an extra charge with nonzero spin, which can be used for spin manipulation.\(^{20,21}\) More importantly, the density of trions can be electrically tuned by the gate voltage, offering...
remarkable optoelectronic applications. Recently, tightly bound trions have been observed in monolayer MoS$_2$ at room temperature, which is of considerable interest for the fundamental studies of many-body interactions, such as carrier multiplication and Wigner crystallization. However, trions have not been observed in bilayer MoS$_2$, since the PL spectra in bilayer MoS$_2$ could not be tuned at room temperature owing to its indirect band gap nature at room temperature. Fortunately, we could use temperature to tune the valley positions in bilayer MoS$_2$ and make its electronic band structure approaching direct band gap manner, which offers the electrical tunability of the exciton and trion dynamics in bilayer MoS$_2$ at low temperature.

Using back-gated metal–oxide–semiconductor (MOS) devices, we demonstrate the tunability of exciton and trion dynamics in bilayer MoS$_2$ at low temperature, with the comodulations by both temperature and electric field. We used mechanical exfoliation to transfer a MoS$_2$ flake (with mono- and bilayer MoS$_2$) onto a SiO$_2$/Si substrate (275 nm thermal oxide on n$^+$ doped silicon). The MoS$_2$ flake was placed near a gold electrode that was prepatterned on the substrate. Another thick graphite flake was similarly transferred to electrically bridge the MoS$_2$ flake and the gold electrode, forming a MOS device. This fabrication procedure kept the MoS$_2$ samples free from chemical contaminations by minimizing the post-processes after the MoS$_2$ flake was transferred. In the measurement, the gold electrode is grounded, and the n$^+$ doped Si substrate functions as a back gate providing uniform electrostatic doping in the MoS$_2$ (Figure 3b). In the experiment, we tuned the back gate voltage from 50 to $-50$ V. For the monolayer MoS$_2$, obvious gate-dependent PL spectra were observed at both 298 and 83 K (Figure 3c,d). In the PL spectra, the higher-energy emission peak at $\approx 1.92$ eV is attributed to neutral exciton (A) emission, and the lower-energy emission peak at $\approx 1.88$ eV (Figure 3c,d) is attributed to negative trion (A$^-$) emission, which is consistent with previously report. MoS$_2$ sample is an n-type semiconductor owing to the initial electron doping, which makes the negative trion PL peak dominant at zero back gate voltage (Figure 3c,d). As the back gate voltage $V_g$ was changed from $-50$ to 50 V, positive charges were injected to monolayer MoS$_2$ layer sample and makes the doping level close to neutral at $-50$ V. Therefore, the exciton spectral weight was increasing with the injection of positive charges by back gate voltages and negative trions (A$^-$) will be converted to excitons (A). The conversion can be represented as $\text{A}^- + h \rightarrow \text{A}$, where $h$ represents a hole. In monolayer MoS$_2$, most photocarriers will recombine through the K–K transitions at 298 K. When temperature is down to 83 K, A valley slightly moves down relatively to K valley, which reduces the weight of the photoelectrons relaxed into K valley. However, as the photoelectrons still remain with a moderate amount in K valley, the conversion from exciton (A) to negative trion (A$^-$) will not be significantly influenced, so gate-dependent PL spectra could be observed from monolayer MoS$_2$ at both 298 and 83 K as shown in Figure 3c,d, respectively.

On the other hand, we did not observe obvious gate-dependent PL spectra from bilayer MoS$_2$ at 298 K.
At 298 K, bilayer MoS$_2$ has an indirect band gap and the quasi-Fermi level locates within the lower-energy $\Lambda$ valley, but not the higher-energy K valley. The electric field from the back gate will only tune the photoelectron density within the $\Lambda$ valley, but not within the K valley. Since the main PL peak in bilayer MoS$_2$ comes from the direct K–K transition, the electric field would not affect the main PL K–K emission at 298 K (Figure 3e inset). This situation changes when the MOS device was cooled down to 83 K, at which the clear gate-dependent PL spectra emerged (Figure 3f). Two clear emission peaks, located at $\approx$1.91 and $\approx$1.88 eV, respectively, could be observed via back gate modulation. The higher-energy peak at $\approx$1.91 eV is attributed to exciton (A) emission and
the lower-energy peak at ≈1.88 eV is attributed to the negative trion (A⁻) emission. The trion binding energy[29] is the energy difference of these two peaks A and A⁻. The emerging gate-dependent PL spectra in bilayer MoS₂ at 83 K come from the rising up of the Λ valley relatively to the K valley. When the energy of Λ valley becomes comparable to that of K valley, the weight of photoelectrons relaxed to K valley will be highly enhanced (Figure 3f inset), which leads to the electrical tunability of the PL and the exciton and trion dynamics in bilayer MoS₂ at 83 K.

In order to investigate the detailed exciton and trion dynamics in mono- and bilayer MoS₂, we measured their PL spectra under various back gate voltages at different temperatures ranging from 298 down to 83 K. All the PL spectra are fitted using Lorentzian function to extract the exciton and trion components (Figures S4 and S5, Supporting Information). In monolayer MoS₂, the intensity of trions exhibits a large gate dependence, while the intensity of trions approximately preserves when the back-gated voltage is changed from −50 to 50 V at 83 K (Figure 4a), which is consistent with previous report.[9] For bilayer MoS₂, the back gate voltage will have an obviously larger influence on the spectral weight of trions than that of excitons at 83 K (Figure 4b), which could be related to the initial carrier density of K valley in bilayer MoS₂.[17] The trion binding energies of mono- and bilayer MoS₂ are measured to be 39 and 27 meV at 83 K (Figure 4c,d), respectively. The lower trion binding energy in bilayer MoS₂ could be due to the reduced quantum confinement.[9,30] For 2L MoS₂ at both 298 and 263 K, the PL spectra can only be fitted using one peak and this peak is attributed to the emission of excitons, according to the temperature evolution of exciton and trion peak energies for 2L MoS₂ (Figure 4d and Figure S5b, Supporting Information).

This is because most of the photoexcited electrons relax to the Λ valley rapidly, making the neutral excitons dominant in the K–K transition (Figure 3c inset). We find that the peak positions of exciton and trion emissions in both mono- and bilayer can be fitted well (solid lines in Figure 4d) using a standard semiconductor band gap dependence[7,31] of \( E_T(T) = E_T(0) - S\hbar / \omega \left( \cosh \left( \frac{\hbar \omega}{2kT} \right) - 1 \right) \), where \( E_T(0) \) is the ground-state transition energy at 0 K, \( S \) is a dimensionless coupling constant, and \( \hbar \omega \) is an average phonon energy.

**Figure 4.** Exciton and trion dynamics in mono- and bilayer MoS₂, at different back gate voltages and temperatures. a,b) PL intensity of emission peaks from excitons (“A”) and trions (“A⁻”) as a function of gate voltages, from 1L (a) and 2L (b) MoS₂ at 83 K. c) PL peak energy of “A” and “A⁻” emissions as a function of gate voltages for 1L and 2L MoS₂ at 83 K. d) PL peak energy as a function of temperature. For 2L, the “A” peak can only be fitted out below 233 K. All the peaks are fit to Lorentzians by multipeak fitting (see the Supporting Information). The solid lines are the fitting curves using a standard semiconductor band gap dependence of \( E_T(T) = E_T(0) - S\hbar / \omega \left( \cosh \left( \frac{\hbar \omega}{2kT} \right) - 1 \right) \), where \( E_T(0) \) is the ground-state transition energy at 0 K, \( S \) is a dimensionless coupling constant, and \( \hbar \omega \) is an average phonon energy.
energy. From the fits, we extract for excitons (trions) the $E_g = 1.921$ (1.883) eV, $S = 1.668$ (1.488), $\hbar \omega = 26.92$ (21.29) meV in monolayer MoS$_2$, and $E_g = 1.909$ (1.883) eV, $S = 2.223$ (1.887), $\hbar \omega = 28.99$ (23.81) meV in bilayer MoS$_2$.

In conclusion, we successfully used comodulation technique by both temperature and electric field to probe the exciton and trion dynamics in bilayer MoS$_2$. From numerical calculations, we show that the band structure evolution of bilayer MoS$_2$ is from indirect at room temperature toward direct band structure as temperature decreases, while monolayer MoS$_2$ shows an adverse trend. This opposite temperature dependence of the band structure evolution in mono- and bilayer MoS$_2$ can significantly change the photocarrier relaxation pathways in their PL processes, which leads to more than twice faster increasing of the measured PL intensity from bilayer MoS$_2$ than that from monolayer MoS$_2$ as temperature decreases. More importantly, this indirect-to-direct transition trend in bilayer MoS$_2$ at low temperature provides the electrical tunability of the K–K direct PL transition, which enables the exploration of exciton and trion dynamics in bilayer MoS$_2$. The trion binding energy of bilayer MoS$_2$ was then measured to be 27 meV at 83 K, which is smaller than the measured trion binding energy of 39 meV in monolayer MoS$_2$. Our results pave a new way to enable new excitonic devices using bilayer MoS$_2$.

**Experimental Section**

Device Fabrication and Characterization: Mechanical exfoliation was used to transfer a MoS$_2$ flake onto a SiO$_2$/Si substrate (275 nm thermal oxide on n$^+$-doped silicon), near a prepatterned Au electrode. The Au electrodes were patterned by conventional photolithography, metal deposition, and lift-off processes. Another thick graphite flake was similarly transferred to electrically bridge the Au electrodes, forming a MOS device. All PL measurements were conducted using a T64000 micro-Raman system equipped with a charge-coupled device (CCD) and InGaAs detectors, along with a 532 nm Nd:YAG laser as the excitation source. For low temperature measurements, the sample was placed into a microscope-compatible chamber with a low temperature controller (liquid nitrogen as the coolant). The electrical bias was applied using a Keithley 4200 semiconductor analyser.

**Trion Binding Energy Extraction:** The binding energies of excitons and trions are extracted from the measured PL spectra using multipoint Lorentz fitting, which has been successfully used by Shan and co-workers$^{[9]}$ and Xu and co-workers.$^{[29]}$ Through Lorentz fitting, we can clearly see two peaks in each measured PL spectra. The higher-energy peak (A) is attributed to the neutral exciton emission, and the lower-energy peak (A') is due to the trion emission.$^{[9,26]}$ From the gate-dependence of these two peaks, we know the trion is negatively charged trion. The trion binding energy is the energy difference of these two peaks A and A$^\prime$.

**Band Structure Simulation:** The band structures of 1–2 L MoS$_2$ were calculated at different temperatures within DFT molecular dynamics calculation using PW generalized gradient approximation based on a real-space numerical atomic orbital code.$^{[18]}$ A double numerical polarized basis set was used with a k-point set of $25 \times 25 \times 1$. All electrons are included in the calculation.

A vacuum space of at least 30 Å was kept to avoid mirror interactions. The temperature dependence of the electronic structure is based on modeling the effects of thermal lattice expansion and the electron–phonon interaction. Before performing the DFT molecular dynamics calculation, the total number of particles, the system’s volume, and the absolute temperature become constant and the system reaches an equilibrium state, after 10 ps relaxation (called the canonical NVT ensemble). At a certain temperature, molecular dynamics simulations at this temperature are conducted first to determine the lattice parameters; and then band structure is calculated using ab initio method based on the lattice parameters.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**References**


Supporting Information

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Supporting Information for

Exciton and trion dynamics in bilayer MoS\textsubscript{2}

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S1. Calculation of the band structures at different temperatures

We calculated the band structures of 1-2L MoS$_2$ under different temperatures within density-functional theory (DFT) using Perdew-Wang (PW) generalized gradient approximation (GGA) based on a real-space numerical atomic orbital code. A double numerical polarized basis set was used with a $k$-point set of 25×25×1. All electrons are included in the calculation. A vacuum space of at least 30 Å was kept to avoid mirror interactions. The temperature dependence of the electronic structure is based on modelling the effects of thermal lattice expansion and the electron-phonon interaction. Before performing the DFT molecular dynamics calculation, the total number of particles, the system’s volume and the absolute temperature become constant and the system reaches an equilibrium state, after 10 ps relaxation (called the canonical NVT ensemble). At a certain temperature, molecular dynamics simulations at this temperature are conducted first to determine the lattice parameters; and then band structure is calculated using ab initio method based on the lattice parameters.

**Figure S1 | Temperature induced evolution of the Λ valley and Γ peak.** (a), calculated energy difference between Λ valley ($E_{C\Lambda}$) and K valley ($E_{CK}$) in the conduction band (b) and energy difference between Γ peak ($E_{V\Gamma}$) and K peak ($E_{VK}$) in the valence band of 1-2L MoS$_2$, as a function of temperature. The dashed lines are guides for the eye. Based on above calculation results, 1L MoS$_2$ owns a direct band gap at the temperature range of ~210-300 K
or < 50 K and an indirect band at the temperature range of ~50-210 K; in contrast, 2L MoS$_2$ shows a direct band at the temperature range of ~50-250 K.

Wang$^{[2]}$ et al has demonstrated that when thick MoS$_2$ is thinned down to 1L, the Λ valley of conduction band moves up and the Γ hill of valence band moves down, because of the reduction of interlayer coupling. Please note that above simplified simulation is based on the DFT calculation of band structures at absolute zero temperature.

Figure S2 | Molecular structure of MoS$_2$. "a" is the distance between S-S atoms; "b" is the distance between S-Mo atoms, "c" is the distance between two layers.

In the real case, temperature can modulate the lattice parameters (Figure S2), $a$, $b$, $c$, significantly. It should be noted that the in-plane thermal expansion (modulation of “$a$” and “$b$”), also has distinct effects on the band structure, although the in-plane thermal expansion is typically much smaller compared to “$c$” expansion.$^{[3]}$ For bilayer MoS$_2$ in the real case, the moving trends of Λ valley and Γ hill are not only affected by the interlayer coupling (“$c$”), but also the modulation of in-plane thermal expansion (“$a$” and “$b$”) as well as electron-phonon interactions at elevated temperature. In addition, please note that the temperature-induced modulation of band structure is also dependent on the material itself. For instance, compared with 2L MoSe$_2$, 2L MoS$_2$ has totally opposite temperature dependence of photoluminescence due to different band structure evolution as a function of temperature.$^{[4]}$

Typically, in order to simplify the simulation process, some DFT calculations only consider
interlayer coupling and ignore the influence of in-plane thermal expansion.\cite{2,4} Also, people assume that thermal expansion causes the interlayer spacing to increase monotonously with temperature, thus reducing the interaction between the layers. In the real case, for few-layer MoS$_2$, the temperature-induced modulation of the interlayer spacing is not monotonous.\cite{5} It is impossible to find the temperature modulation of such complex systems analytically; therefore, we use MD simulation to circumvent this problem by using numerical methods. From our MD simulation (Figure S1), the evolution of band structure as a function of temperature is indeed not monotonous. In the temperature range of 300 - 100 K, our simulation results can reasonably explain our experimental findings.
Figure S3 | Temperature dependence of PL intensity from another MoS$_2$ sample with mono-and bi-layers. (a), measured PL spectra of the 1L MoS$_2$ under various temperatures. (b), measured PL spectra of the 2L MoS$_2$ in the same spectral range. (c), PL peak intensity of the 1-2L samples as a function of temperature.
Figure S4 | Exciton and trion dynamics as a function of back gate voltage in the mono- and bi-layer MoS₂ at 83 K. (a)-(b), measured PL spectra of 1L (a) and 2L (b) MoS₂ samples under different back gate voltages, with Lorentzian fittings (solid violet curves are the measured raw data, solid red lines are the exciton (A) components, solid black lines are the trion (A⁻) components, and solid green lines are the cumulative fitting results). The dashed lines are added as guides to the eye for the peak energies of excitons (A, red line) and trions (A⁻, black line).
Figure S5 | Exciton and trion dynamics as a function of temperature, in mono- and bi-layer MoS₂. In order to clearly see the exciton (A) and trion (A̅) peak, the PL spectra under back gate voltages of 50 and -50V are plotted in the same panel. The dashed lines are added as guides to the eye for the peak energies of excitons (A, red line) and trions (A̅, black line).
References


