Electrical switching between exciton dissociation to exciton funneling in MoSe₂/WS₂ heterostructure

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The heterostructure of monolayer transition metal dichalcogenides (TMDCs) provides a unique platform to manipulate exciton dynamics. The ultrafast carrier transfer across the van der Waals interface of the TMDC hetero-bilayer can efficiently separate electrons and holes in the intralayer excitons with a type II alignment, but it will funnel excitons into one layer with a type I alignment. In this work, we demonstrate the reversible switch from exciton dissociation to exciton funneling in a MoSe₂/WS₂ heterostructure, which manifests itself as the photoluminescence (PL) quenching to PL enhancement transition. This transition was realized through effectively controlling the quantum capacitance of both MoSe₂ and WS₂ layers with gating. PL excitation spectroscopy study unveils that PL enhancement arises from the blockage of the optically excited electron transfer from MoSe₂ to WS₂. Our work demonstrates electrical control of photoexcited carrier transfer across the van der Waals interface, the understanding of which promises applications in quantum optoelectronics.
two-dimensional (2D) semiconductors are promising candidates for light-harvesting and optoelectronic applications due to their strong light–matter interaction from excitonic responses. Their atomically thin nature further enables engineering exciton dynamics and energy relaxation pathways through ultrafast carrier transfer across 2D van der Waals (vdW) interfaces. In particular, a vdW heterostructure can, respectively, dissociate electrons and holes into separate layers or funnel excitons to one layer with a type II or type I band alignment. It is highly desirable to achieve both functions in a single device in an electrically reconﬁgurable way. However, to the best of our knowledge, this has not been demonstrated yet. Here we demonstrate reversible electrical switching between exciton dissociation and funneling in a MoSe2/WS2 heterostructure device. We show that the electron transfer from MoSe2 to WS2 can be blocked by efﬁcient gating of the LaF3 substrate, leading to a transition between photoluminescence (PL) quenching to PL enhancement for the MoSe2 A exciton emission. The ability to electrically control interlayer charge transfer pathways ushers in application concepts, such as light switch and energy steering.

Results
Charge transfer in the MoSe2/WS2 heterostructure. We construct the MoSe2/WS2 heterostructure on the LaF3 substrate through a layer-by-layer dry transfer technique, and the heterostructure is also capped by a thin layer of hexagonal boron nitride (BN) on the top. A typical MoSe2/WS2 heterostructure on the LaF3 substrate is shown in Fig. 1a. The overlapped region of the monolayer MoSe2 and WS2 forms the MoSe2/WS2 heterojunction. We use few-layer flakes of graphene to contact both the monolayer MoSe2 and WS2, and a schematic of the device is shown in Fig. 1b. The heterostructure can be gated through the LaF3 substrate as the back gate, which provides efﬁcient control of doping through the double layer, as schematically shown in Fig. 1b. Typical PL spectra for different regions of the device are shown in Fig. 1c, with the continuous wave (CW) laser excitation centered at 2.331 eV and a power of 100 µW. Without applying any gate voltage, the PL from the MoSe2/WS2 heterojunction (red line in Fig. 1c) exhibits both the PL of the MoSe2 A exciton resonance (~1.979 eV) and MoSe2 A exciton resonance (~1.548 eV), compared with that of the monolayer WS2 (blue line in Fig. 1c) and the monolayer MoSe2 (black line in Fig. 1c), respectively (see Supplementary Note 4). This simultaneous quenching of PL at both MoSe2 and WS2 A excitons was observed in all the heterostructures we constructed, including three MoSe2/WS2 heterostructures on SiO2/Si substrate and seven heterostructures on LaF3 in the absence of the gate voltage (see Supplementary Notes 1 and 7). It thus suggests a type II alignment for the as-prepared MoSe2/WS2 heterostructures, and the PL quenching is a result of the optically excited electron transferred to the MoSe2 layer and hole transferred to the WS2, according to the band alignment shown in the inset of Fig. 1c. It is interesting to note that the quenching of MoSe2 A exciton PL is signiﬁcantly less than that of the WS2 A exciton in the heterojunction region. While the integrated PL of the WS2 A exciton in the heterojunction is quenched by more than one order of magnitude smaller, the integrated PL intensity of MoSe2 A exciton in the heterojunction is only slightly quenched, being ~70% of that from the monolayer MoSe2 (Fig. 1c). The signiﬁcantly less quenching of MoSe2 A exciton PL can be understood from the relative band alignment shown in the inset of Fig. 1c. In the type II alignment conﬁguration, the conduction band minimum (CBM) of the WS2 is only slightly lower than that of the MoSe2 according to the theoretical calculations. The thermal equilibrium of the two CBMs at room temperature therefore allows a certain population of electrons in the CBM of the MoSe2 even though the CBM of the WS2 is the lower energy state for electrons in the heterojunction region. The small energy difference between the two CBMs offers the opportunity for us to apply an efﬁcient electrostatic gating to manipulate the optically excited carrier transfer across the MoSe2/WS2 interface. We achieve that by using the LaF3 as the ionic back gate, which has been proven to efﬁciently gate 2D materials though a double layer (schematically shown in Fig. 1b).

Gate-dependent PL enhancement in MoSe2/WS2. To reveal the effect of gate-controlled carrier transfer across the heterojunction, we then investigate the PL spectra around the MoSe2 A exciton resonance as a function of the gate voltage for both monolayer MoSe2 (Fig. 2a) and MoSe2/WS2 heterojunction (Fig. 2b) (see Supplementary Note 5). The CW laser excitation centered at 2.0 eV (620 nm) with a power of 100 µW was used to obtain the PL spectra shown in Fig. 2a, b. This excitation photon energy is

![Fig. 1 Monolayer MoSe2/WS2 heterostructure device. a Optical microscopic image of the monolayer MoSe2/WS2 heterostructure, capped with a few-layer h-BN layer. Scale bar: 5 µm. b Schematic of the MoSe2/WS2 heterostructure device, contacted by few-layer graphene electrodes and gated by the ion substrate LaF3. c Typical room temperature PL spectra from regions of the monolayer MoSe2 (black), monolayer WS2 (blue), and MoSe2/WS2 heterojunction (red), with no gate voltage applied. Inset: schematic representation of the type II band alignment of the MoSe2/WS2 heterostructure.](image-url)
large enough to excite A excitons in both monolayer MoSe$_2$ (1.548 eV) and WS$_2$ (1.979 eV). The PL intensity from the WS$_2$ A exciton is drastically quenched in the heterojunction, and we focus on the PL intensity of the MoSe$_2$ A exciton for both the monolayer (Fig. 2a) and heterojunction region (Fig. 2b). We can see from the color plots (Fig. 2a, b) that, although the MoSe$_2$ A exciton PL intensity is weaker in MoSe$_2$/WS$_2$ heterojunction (Fig. 2b) than in the monolayer MoSe$_2$ (Fig. 2a) for the gate voltage from $-2$ to $-1$ V, the PL is stronger in the heterojunction than in the monolayer MoSe$_2$ at the gate voltage $>0$ V. This relative PL ratio from quenching to enhancement transition is clearly illustrated in the PL spectra in Fig. 2d, which combine the line cuts of Fig. 2a, b at the gate voltage $-2$ and $4$ V. To better understand the PL behavior change, we define the PL enhancement factor (EF) as $EF = I_{\text{Heter}} / I_{\text{Monolayer}}$, where $I_{\text{Heter}}$ ($I_{\text{Monolayer}}$) is the integrated MoSe$_2$ A exciton PL intensity in the MoSe$_2$/WS$_2$ heterojunction (monolayer MoSe$_2$). EF as a function of the gate voltage for the photoexcitation centered at 2.0 eV is shown in Fig. 2e (black dots), which shows that EF is almost a constant between the gate voltage of $-2$ to $-1$ V (EF ~ 0.6) but quickly rises to ~1.8 at the gate voltage 0 V, and it remains largely a constant as the gate voltage is further increased.

It is interesting to note that this observation is sensitive to the excitation photon energy, and the results are distinctively different for the CW photoexcitation of the same power (100 µW) but centered at 1.797 eV (690 nm), which is below the A exciton resonance energy of WS$_2$ but above that of MoSe$_2$. At the gate voltage of $-4$ V, we observe PL quenching at MoSe$_2$ A exciton resonance in the heterojunction (Fig. 2d), similar to the scenario with the photoexcitation at 2 eV (Fig. 2c). However, as we increase the gate voltage to 4 V, we do not observe the PL enhancement of the MoSe$_2$ A exciton in the heterojunction, even though the PL intensity is quite close to that of the monolayer MoSe$_2$ (Fig. 2d). A detailed gate-dependence study of the photoexcitation centered at 1.797 eV also results in quantitative EF as shown in Fig. 2e (red dots), which shows a similar step function behavior as the case of...
photoexcitation centered at 2.0 eV, but the maximum value of EF is smaller and never exceeds 1.

PL excitation (PLE) spectroscopy of EF in MoSe2/WS2. Since the observed PL EF at MoSe2 A exciton resonance is sensitive to the excitation photon energy, we then perform a detailed PLE spectroscopic study. The integrated PL intensity at the MoSe2 A exciton resonance for monolayer MoSe2 (black) and MoSe2/WS2 heterojunction (red) regions at the gate voltage of −2 V (a) and the gate voltage of 4 V (b), respectively. Figure 3c, d are PL enhanced factor for the gate voltage of −2 V (c) and 4 V (d), respectively.

Previous work has shown that the LaF3 back gate should be at least >100 times more efficient than the silicon back gate with 300 nm thermal oxide.

Discussion

The experimental observation can be understood theoretically by considering the gate-dependent carrier distribution in the heterojunction. Taking into account the quantum capacitance of the monolayer MoSe2 and WS2, for the device configuration shown in Fig. 1b, the effective capacitance model can be schematically shown as the inset of Fig. 4 (MoSe2 being the bottom layer, and detailed derivation in Supplementary Note 3). Here CQ1(CQ2) are the quantum capacitance of monolayer MoSe2 (WS2), CG1 is the geometry capacitance between MoSe2 and the LaF3 back gate, and CG2 is the geometry capacitance between MoSe2 and WS2. For qualitative understanding, we consider zero-temperature case here (see Supplementary Note 2 for the discussion of the finite temperature case, which does not qualitatively change the picture). Owing to the large energy difference between the VBMs of MoSe2 and WS2, the hole transfer from WS2 to MoSe2 plays the central role in the PL enhancement. As shown in Fig. 4, when the gate voltage is at point A (e.g., −2 V for the device 2 shown in Fig. 2), both the MoSe2 and WS2 layers are intrinsic and with the quantum capacitance of zero. As a result, the gate voltage is dropped only on the quantum capacitance and the band alignment is determined by the work function of each layer. The type II alignment (shown at point A in Fig. 4) determines that the optically excited electron in MoSe2 will transfer to WS2, reducing the electron density in the MoSe2 layer in the heterojunction, compared to the case of the bare monolayer MoSe2. In addition, with (on-resonance excitation) and without (off-resonance excitation) the hole transfer from WS2 to MoSe2,
The electron density in the MoSe2 layer in the heterojunction is always less than the hole density. As a result, the electron is the minor carrier that determines the available MoSe2 A exciton density. The reduced electron density thus leads to the quenching of MoSe2 A exciton PL in the heterojunction.

When the WS2 layer starts to get electron-doped (point B), the number of optically excited electrons transferred from the MoSe2 to WS2 in the heterojunction region will be modulated by the gate voltage. For simplicity, we can use the off-resonance excitation as an example. The charge transfer from MoSe2 to WS2, \( \Delta Q \), can be obtained from the following equation according to the effective capacitance model33 (inset of Fig. 4):

\[
\frac{Q - \Delta Q}{C_{Q1}} = \frac{\Delta Q}{C_{Q2}} + \frac{\Delta Q}{C_{G2}},
\]

where \( Q \) is the total charge of optically excited electrons in the MoSe2 layer of the MoSe2/WS2 heterojunction. Reorganization of Eq. (1) results in the expression of \( \Delta Q \) as:

\[
\Delta Q = \frac{1}{1 + \frac{C_{Q2}}{C_{Q1}} + \frac{C_{G2}}{C_{G1}}},
\]

For gate voltage smaller than that of point B, \( C_{Q1} = 0 \) and hence \( \Delta Q = Q \), which indicates that ~100% of the optically excited electron in MoSe2 layer of the heterojunction region is transferred to WS2. As a result, PL quenching of the MoSe2 layer in the heterojunction is similar to that of A point (similar EF). As we move forward from point B, however, electron transfer will be less efficient due to the finite \( C_{Q1} \) (i.e., finite density of states (DOS) at Fermi level in MoSe2) and the PL quenching will be less significant. As the gate voltage is increased to point C, the doping further increases and the Fermi level is aligned with the conduction band of MoSe2. Assuming a similar effective electron mass \( m^* \) in WS2 and MoSe2, we have \( C_{Q1} = C_{Q2} = C_{G2} \), where \( C_{G2} = \frac{\varepsilon}{d} \) is the DOS in 2D. Since \( C_{Q2} \gg C_{G2} \) (see Supplementary Note 2), from Eq. (2), we found that \( \Delta Q \approx 0 \) and optically excited electron transfer from MoSe2 to WS2 is blocked. The EF of MoSe2 A exciton PL will therefore again be largely a constant, with the value of 1 (Fig. 4) for the off-resonance excitation in the ideal scenario.

The electron transfer in the on-resonance scenario can be understood in a similar fashion (see Supplementary Note 2), with similar PL quenching (EF < 1) from point A to B. However, when MoSe2 is sufficiently doped (point C), optically induced holes in the MoSe2 layer become the minor carrier that determines the MoSe2 A exciton density. For the on-resonance excitation, the WS2 layer is also excited and we have nearly 100% of the optically excited holes transfer from WS2 to MoSe2. Therefore, the A exciton density in the MoSe2 layer in the heterojunction is enhanced, giving rise to the PL enhancement with a largely constant EF > 1. We thus conclude that, for both the off-resonance and on-resonance excitation, the qualitative gate dependence of EF will be of the form shown in Fig. 4. Particularly, EF will show an abrupt increase around specific gate voltage (region II) and remain largely constant on either side. On the low voltage side (region I), EF should be <1; and on the high gate voltage side (region III), EF = 1 for the off-resonance excitation and EF > 1 for the on-resonance excitation.

The theoretical prediction in Fig. 4 is in excellent agreement with our experimental observation in Fig. 2e. The experimentally observed EF as a function of the gate voltage can be clearly divided into three regions, similar to a step function for both the on-resonance and off-resonance excitation as predicted by Fig. 4.

The EF for the on-resonance excitation (photoexcitation at 2.0 eV) in region III shows an EF ~ 2.0, while the EF for the off-resonance excitation (photoexcitation at 1.797 eV) in the region III is about 0.8. The EF of <1 for the off-resonance case is probably due to decreased quantum efficiency in the heterojunction from the different dielectric environment.

The consideration of the finite temperature effects is included in Supplementary Note 2, and it gives qualitatively similar results as in Fig. 4. Interestingly, we found that, for large enough gate voltage, the charge accumulated on the \( C_{G2} \) will give rise to a large energy shift between the MoSe2 and WS2, which switches the type II alignment to a type I alignment configuration, as shown schematically by the inset at point E in Fig. 4. The efficient ionic gating thus not only allows the control of optically excited carrier transfer across the atomically sharp interface but also leads to the possibility of modifying the alignment type reversibly. The associated fundamental understanding will enable quantum
optoelectronics based on transition metal dichalcogenide (TMDC) vdW heterostructures.

Method

Device fabrication. The MoSe2/WS2 heterostructure devices were fabricated through a layer-by-layer dry transfer technique. More specifically, each of the monolayer TMDC was sequentially transferred to the LaF3 substrate, and a final BL film was used to cap the heterostructure. Two pieces of few-layer graphene were used as the electrodes to contact the monolayer MoSe2 and WS2 layers separately, and both were grounded during the measurements, as schematically shown in Fig. 1b. The final devices were annealed in vacuum at 100°C for 5 h.

Optical measurements. All the optical measurements in this work were performed at room temperature. The micro-PL measurements were performed with a homemade confocal microscope, in which the excitation lasers were focused to a spot size of ~2 μm. The PLE spectra were taken with a supercontinuum white laser (Fianium), and the filtered light (with bandwidth ~4 nm) was used as the excitation source.

Data availability

The data that support the findings of this study are available from the authors on reasonable request, see “Author contributions” for specific data sets.

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Author contributions

S.-F.S. conceived the experiment. Y.M. and Z.Lian fabricated the devices. Y.M. and T.W. performed the measurements. S.-F.S., T.W., Y.M., C.J., S.M., Z.Li and F.S. analyzed the data. T.T. and K.W. grew the BN crystals. S.-F.S. supervised the project. S.-F.S., T.W., Y.M., Z.Lian and S.-F.S. acknowledge support from NYSTAR through Focus Center-NY–RIPE Contract C150117. The device fabrication was supported by the Micro and Nanofabrication Clean Room (MNCR) at Rensselaer Polytechnic Institute (RPI). K.W. and T.T. acknowledge support from the Elemental Strategy Initiative conducted by the MEXT, Japan and the CREST (JPMJCR15F3), JST. F.S. acknowledges support from the National Natural Science Foundation of China (No. U1732273). S.-F.S. also acknowledges the support from a KIP grant from RPI and a VSP grant from NHMFL.

Competing interests

The authors declare no competing interests.

Additional information

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