Charge Transport in MoS$_2$/WSe$_2$ van der Waals Heterostructure with Tunable Inversion Layer

Manh-Ha Doan,$^{†*}$‡ Younggio Jin,$^{†*}$‡ Subash Adhikari,$^{†*}$‡ Sanghyub Lee,$^{†*}$‡ Jiong Zhao,$^{†*}$‡ Seong Chu Lim,$^{†*}$§ and Young Hee Lee$^{†*}$

$^{†}$Center for Integrated Nanostructure Physics, Institute for Basic Science (IBS), Suwon 16419, Republic of Korea
$^‡$Department of Energy Science and $^§$Department of Physics, Sungkyunkwan University, Suwon 16419, Republic of Korea

Supporting Information

ABSTRACT: Despite numerous studies on two-dimensional van der Waals heterostructures, a full understanding of the charge transport and photoinduced current mechanisms in these structures, in particular, associated with charge depletion/inversion layers at the interface remains elusive. Here, we investigate transport properties of a prototype multilayer MoS$_2$/WSe$_2$ heterojunction via a tunable charge inversion/depletion layer. A charge inversion layer was constructed at the surface of WSe$_2$ due to its relatively low doping concentration compared to that of MoS$_2$, which can be tuned by the back-gate bias. The depletion region was limited within a few nanometers in the MoS$_2$ side, while charges are fully depleted on the whole WSe$_2$ side, which are determined by Raman spectroscopy and transport measurements. Charge transport through the heterojunction was influenced by the presence of the inversion layer and involves two regimes of tunneling and recombination. Furthermore, photocurrent measurements clearly revealed recombination and space-charge-limited behaviors, similar to those of the heterostructures built from organic semiconductors. This contributes to research of various other types of heterostructures and can be further applied for electronic and optoelectronic devices.

KEYWORDS: vdW heterostructure, charge transfer, inversion layer, tunneling, recombination, photocurrent

Van der Waals (vdW) heterostructures have emerged as an attractive research topic for both fundamental science and applied physics.$^{1,2}$ Among the fabricated vdW heterostructures reported recently, the MoS$_2$/WSe$_2$ structure has attracted much attention because its band offset exists as a type II, staggered gap, which can be used for many electronic and optoelectronic applications such as photovoltaic devices, light emitters, tunnel diodes, and field-effect transistors (FETs).$^{3-7}$ Although the p–n diode behavior is usually observed in the current–voltage characteristics of this heterojunction, the underlying physical mechanisms related to interfacial charge transfers, formation of charge depletion/inversion layers, which play a crucial role for optimizing the device performance, have not been fully understood. For example, in the MoS$_2$/WSe$_2$ heterojunction, the rectifying behavior of the I–V curves is governed by the interlayer recombination of free carriers,$^3$ in good contrast with other reports of the simple p–n junction model with the flat band at the interface.$^1$–$^6$

The vdW heterostructure devices have been usually fabricated right after the stacking process without efficient thermal treatments$^3$–$^6$ except in a few cases.$^7$–$^8$ In the vdW junctions made by a manually stacking process, air gaps and/or residues usually exist at the interface, regardless of the so-called wet or dry transfer methods.$^3$–$^9$ These residues result in a poor interfacial contact between the stacked layers. In such a case, the interlayer coupling is not strong, resulting in an insufficient charge transfer. A flat band model is then reasonable to explain, for example, electronic transport via a monolayer heterojunction or multilayer heterojunction with weak coupling. However, in an ideal interface with strong coupling, charge transfer occurs at the interface, resulting in the band bendings, and then the flat band model for multilayer heterojunction is no longer valid, which may involve complicated charge depletion and charge inversion layers.$^{10}$–$^{16}$

Constructing a heterointerface with a strong interaction is certainly advantageous in searching for interesting physics and useful applications. For example, in the epitaxially grown heterojunctions with III–V semiconductors or oxides, charge transfers and couplings at the interface give rise to emergent phenomena such as ultrahigh electron mobility, superconductivity, quantum Hall effects, and so on. Most of these

Received: January 2, 2017
Accepted: March 14, 2017
Published: March 14, 2017

DOI: 10.1021/acsnano.7b00021
ACS Nano 2017, 11, 3832–3840
observations are based on the formation of a two-dimensional (2D) electron/hole gas at the interface, where the electrons or holes are free to move in planar directions but are tightly confined in the perpendicular direction. While there have been several studies on strong interlayer couplings in the vdW heterojunctions as well as ultrafast charge transfers in these structures, to our knowledge, the formation of the charge inversion layer and its effects on the charge transport and photocurrent generation in the vdW heterojunctions have not been observed yet.

In this work, we construct a prototype multilayer MoS$_2$/WSe$_2$ heterojunction by exfoliating each material and manually stacking them. A strongly coupled interface was established after thermal annealing, confirmed by cross-sectional transmission electron microscopy, Raman spectroscopy, and electrical transport measurements. The measured carrier density in the pristine and overlapped channels allowed us to conclude the staggered heterojunction with the band bending distributed within a few nanometers on the MoS$_2$ side and formation of a charge inversion layer at the WSe$_2$ surface. Effects of the inversion layer on the charge carrier transport and photocurrent of the junction are further studied by the backgate and source/drain voltage modulations.

RESULTS AND DISCUSSION

Figure 1a,b shows a typical scanning electron microscopy (SEM) image and the schematic illustration of our fabricated device. A MoS$_2$ flake with a thickness of ~10 nm was stacked on top of a ~70 nm thick WSe$_2$ layer exfoliated in advance on SiO$_2$/Si substrate (see Supporting Information, Figure S1). By intentionally choosing the MoS$_2$ and WSe$_2$ flakes with the rectangular shapes and stacking them perpendicularly, we can make the devices in which electronic transport in the pristine and overlapped channels can be examined in the same flakes for fair comparison, and electronic transport through the MoS$_2$/WSe$_2$ junction can also be investigated by selecting the source and drain electrodes, as shown in Figure 1b. The interface regions of the sample after annealing were studied using cross-sectional transmission electron microscopy (TEM), as presented in Figure 1c. The atomically contacted and buried interface was observed (Supporting Information, Figures S1 and S2), which ensures the effectiveness of our thermal treatment.

Figure 1d shows the Raman spectra of the sample at the different positions of pristine MoS$_2$ and WSe$_2$ on SiO$_2$ and MoS$_2$/WSe$_2$ overlapped regions. On the pristine MoS$_2$ side, two strong distinct peaks appear near 382 and 408 cm$^{-1}$, corresponding to the respective E$_{2g}$ and A$_{1g}$ modes. WSe$_2$ shows a weak broad peak centered at ~250 cm$^{-1}$. These peaks, in particular, the MoS$_2$ peak, were significantly reduced at the overlapped region even though the MoS$_2$ layer is located on top of WSe$_2$. One may attribute these to variation of the light intensity reflected at the MoS$_2$/SiO$_2$ and MoS$_2$/WSe$_2$ interfaces due to the difference of refractive indices between SiO$_2$ and WSe$_2$. However, we believe that this is not the dominant factor in our system as we used a SiO$_2$/Si substrate with a 300 nm thick SiO$_2$ layer; an enhancement of the Raman peak intensity in the overlapped region is expected because of the higher refractive index of WSe$_2$ (n ~ 3) compared to that of SiO$_2$ (n ~ 1.46). Although the exact mechanism has not been clarified, such intensity reductions are ascribed to strong interfacial coupling between two materials due to the fact that we clearly observed the peak splitting into two parts from both A$_{1g}$ and E$_{2g}$ peaks of MoS$_2$ in the overlapped region: an original one with an unmodified peak position and a new shifted one. To get information on the charge transfer and strain effect, each peak of the overlapped MoS$_2$/WSe$_2$ region was deconvoluted into two components, as shown in the right
Figure 2. Transport phenomena in the pristine and overlapped WSe$_2$ and MoS$_2$ channels. (a) Current-voltage characteristics of the pristine (open symbols) and overlapped channels (filled symbols). (b) Band diagrams of the MoS$_2$/WSe$_2$ heterojunction. Because of asymmetric unintentional doping in MoS$_2$ ($\sim 3.2 \times 10^{15}$ cm$^{-2}$) and WSe$_2$ ($3.8 \times 10^{15}$ cm$^{-2}$), the band bending in WSe$_2$ is stronger than that in MoS$_2$ and an inversion layer is formed on the WSe$_2$ side. (c,d) Transfer characteristics of the pristine and overlapped channels at $V_{ds} = 1$ V. The threshold gate bias shifts, and additional broad shoulders are observed in the overlapped channels shown in the insets.

Panel: The blue shift by 3 cm$^{-1}$ in the A$_{1g}$ peak is understood by phonon stiffening due to electron transfer from MoS$_2$ to WSe$_2$. Some portion of overlapped MoS$_2$/WSe$_2$ still retains unaltered. This implies that the charge transfer in MoS$_2$ takes place partially. The peak near 404 cm$^{-1}$ (black) is not certain but could be contributed by a blue-shifted peak of WSe$_2$. The deconvoluted E$_{1g}^{1}$ peak also has two components: one is red-shifted by 4 cm$^{-1}$, indicating tensile strain. Another peak remains unchanged, revealing similar behavior observed in the A$_{1g}$ peak analysis. The tensile strain further supports contribution of overlapped WSe$_2$ in the A$_{1g}$ peak (black).

The tensile strain in the overlapped region could be introduced after thermal annealing due to the difference in thermal expansion coefficients of MoS$_2$ and WSe$_2$. The thermal expansion coefficients of MoS$_2$ are significantly smaller than those of WSe$_2$ in both a and c directions. Therefore, the MoS$_2$ layers in the overlapped region are expanded during thermal annealing. Expansion residues may exist in MoS$_2$ when the system is cooled to room temperature, resulting in the observed tensile strain. It may involve complex processes which require further studies combining Raman and TEM investigations. Note that the area ratio of the unchanged peak to the changed peak for both A$_{1g}$ and E$_{1g}^{1}$ modes is about 2.5, similar to each other. The area ratio is interpreted by the thickness ratio, $t_1/t_2 = 2.5$, and the total thickness of MoS$_2$, $t = t_1 + t_2 = 10$ nm. Thus, the estimated thickness of the charge-depleted MoS$_2$ side is $\sim 2.8$ nm. The validity of this estimation is further considered from the electronic transport result in the next section. This partial modification of the peak position was not observed in the analysis of the WSe$_2$ peak near 250 cm$^{-1}$ (see Supporting Information, Figure S3). It is, therefore, a sign that the thick WSe$_2$ layer is fully charge-depleted.

To know quantitatively the amount of charges transferred at the interface, we conducted the transfer characteristics for each channel in the same device. Figure 2a shows the current-voltage $(I-V)$ characteristics of the pristine and overlapped MoS$_2$ and WSe$_2$ channels. In these measurements, the potential $(V_{ds})$ was applied only to the two investigated terminals. For reasonable comparisons, the current density is used for presentation in all figures, which is obtained by dividing the measured current by the channel area. The strongly coupled interface between MoS$_2$ and WSe$_2$ renders a huge drain current quenching in the overlapped channels by 2 orders of magnitude compared to the pristine ones. This is ascribed to the reduced free carrier density at each channel by the charge transfer at the interface. The pristine WSe$_2$ and MoS$_2$ FETs show the n-type and p-type behaviors with the threshold gate biases of about 5 and $\sim 40$ V, respectively, as shown in Figure 2c,d. The respective carrier concentrations for the pristine channels are estimated, $p_{WSe}$ $(MoS_2) = q^{-1}C_g|V_{dd}| = 3.8 \times 10^{11}$ (3.1 $\times 10^{10}$) cm$^{-2}$, with $q = 1.6 \times 10^{-19}$ C and $C_g = 1.23 \times 10^{-8}$ F cm$^{-2}$ for the 300 nm thick SiO$_2$ layer. The insets show a shoulder at the small current regime in the overlapped channels that involves the shift of the threshold bias due to the interfacial charge coupling and tunneling for both devices. The shoulder observed in the transfer curve corresponds to the current at the gate bias where the conduction (valence) band of WSe$_2$ (MoS$_2$) is aligned to that of the MoS$_2$ (WSe$_2$) layer (see Supporting Information, Figure S4). In such a case, increasing the negative (positive) gate bias results in the reduction of the current since holes (electrons) flow through the n- (p-) channel.

The reduced on-current density and threshold bias shifts are due to the reduced free carrier density at the interface. From a $\sim 20$ V downshift of the threshold bias for WSe$_2$ at the overlapped channel (Figure 2c), the carrier density change in

DOI: 10.1021/acsnano.7b00021
ACS Nano 2017, 11, 3832–3840

ACS Nano

Article
the WSe2 layer is about $1.5 \times 10^{12} \text{ cm}^{-2}$ using the formula $\Delta \rho = q^{-1}C_{\text{A}}\Delta V$. The similar value of $\Delta V \sim 20 \text{ V}$ was also observed in the overlapped MoS2 channel even though it is located on top of WSe2 (Figure 2d). This implies that the gating effect on the MoS2 layer is also very effective, and thus the screening effect of the bottom WSe2 layer is negligible. It can be understood by the charge depletion on the WSe2 side, in good contrast with previous reports of large screening by the bottom layer with insufficient charge depletion.26,27 The amount of $\sim 1.5 \times 10^{12}$ electrons per square centimeters transferred from MoS2 to WSe2 is half the electron density in pristine MoS2 but larger than the hole concentration in pristine WSe2. Therefore, the p-type carrier of WSe2 is completely converted to the n-type at the overlapped region. The transferred electrons are confined near the surface of WSe2, with a typical depth of $\sim 2 \text{ nm}$, which is usually observed in common oxide or III–V semiconductor interfaces.28 Taking into account strong Coulomb interaction and layer confinement in vdW heterostructures, we believe that the inversion layer width in our system is less than 2 nm.29,30 The upper bound of the density of the excess electrons in this region is calculated to be greater than $5.6 \times 10^{18} \text{ cm}^{-3}$. This value is higher than the estimated effective density of states in the conduction band of WSe2 at room temperature, $N_C = 2\left(\frac{2\pi m^*_e T}{\hbar^2}\right)^{3/2} \approx 4.7 \times 10^{18} \text{ cm}^{-3}$, where $m^*_e \approx 0.34m_0$ is the effective mass of electron in WSe2,31 $T$, $k_B$, and $h$ are temperature, Boltzmann, and Planck constants, respectively. Therefore, the aligned Fermi level of the junction in equilibrium locates above the conduction band of WSe2 at the interface, and consequently, a 2D electron inversion layer is established at the surface of WSe2, as shown in Figure 2b.

To see effects of the charge inversion layer at the interface on the electronic transport through the heterojunction, we provided $I$–$V$ characteristics of the MoS2/WSe2 diode, as shown in Figure 3. Here, the forward bias corresponds to the applied positive voltage on the p-WSe2 side. A clear rectifying behavior of a conventional p–n junction diode is observed similar to the previous reports,7–7 as seen in the inset of Figure 3a. However, when plotted in a log-scale current, we observed two distinct linear regimes in the forward bias region; $0.05 \text{ V} < V_{ds} < 0.2 \text{ V}$ with an ideality factor of $\sim 1.9$ and $0.3 \text{ V} < V_{ds} < 0.6 \text{ V}$ with an ideality factor of $\sim 2.8$, which is clearly visible in the log–log graph in Figure 3b. The current is directly proportional to the applied voltage with a slope of 1 in the first regime, as shown in Figure 3b. This simply implies a direct tunneling via the thermionic emission.32,35 The current increases more rapidly in the second voltage regime because at high source–drain voltages, the potential barrier at the junction originating from the formation of the inversion layer becomes narrower and more triangular, consequently leading to the Fowler–Nordheim (FN) tunneling, evidenced by the replotted $I$–$V$ curve in Figure 3c, following the FN tunneling.34 The related band diagram is shown in Figure 3d. At very high voltage, region III, the current is limited by the injected charges, called the space-charge-limited region.35,56

We further analyze for estimation of the depletion layer thickness in the MoS2 side. Under small applied voltages, the direct tunneling current is proportional to the voltage, expressed by37

$$I \propto V \exp\left(-\frac{2d\sqrt{2m^* \phi}}{h}\right) \tag{1}$$

where $d$, $m^*$, and $\phi$ are the tunneling thickness, effective electron mass, and tunneling barrier; $h$ is the reduced Planck constant. The slope of 1 in Figure 3b simply assures the direct tunneling. The FN tunneling current is widely modeled by the FN law35

$$I \propto V^2 \exp\left(-\frac{4d\sqrt{2m^* \phi^3}}{3h\varepsilon}\right) \tag{2}$$

From two above eqs 1 and 2, the plot of $\ln(I/V^2)$ versus $1/V$ should show a linear regime with a negative slope for the FN tunneling and a logarithm regime for the direct tunneling, as shown in Figure 3c. The threshold voltage of the FN tunneling,
From the slope of the FN region, \( \frac{4d^2/\alpha}{3h} = 1.5 \) S, where \( m^* = 0.5m_0 \) is taken from the literature for the effective mass of the electron in MoS\(_2\).\(^{39}\) We get \( d \approx 1.8 \) nm. This tunneling barrier width is established from the MoS\(_2\) side, as shown in Figure 3d, which is slightly smaller than the charge depletion layer width of 2.8 nm estimated from Raman data and FN analysis above.

It is worth noting that from the doping concentrations of MoS\(_2\) and WSe\(_2\), the depletion layer widths located in the MoS\(_2\) and WSe\(_2\) sides can be calculated by using a depletion model for the conventional p−n junction heterostructures\(^{40,41}\).

\[
\begin{align*}
\chi_p &= \sqrt{\frac{2N_p e_f V_t}{\phi}} \\
\chi_n &= \sqrt{\frac{2N_n e_f V_t}{\phi}}
\end{align*}
\]

Here, \( N_d \approx 3.1 \times 10^{18} \) cm\(^{-3}\) and \( N_i \approx 5.4 \times 10^{16} \) cm\(^{-3}\), which are calculated, \( N_d(d) = n(p)/V_{\text{MobS}_2(WSe}_2) \), from the electron and hole doping concentrations and thicknesses of MoS\(_2\) and WSe\(_2\), \( n(p) \approx 3.1 \times 10^{12} \) (3.8 × 10\(^{11}\)) cm\(^{-3}\) for MoS\(_2\) and WSe\(_2\), respectively;\(^{42}\) and \( V_{th} \approx 0.33 \) V is the built-in potential at the junction which is already extracted from the FN tunneling curve above. With these, we obtained \( \chi_p = 92 \) nm, exceeding the thickness of WSe\(_2\) (70 nm). This implies that the WSe\(_2\) side is completely charge-depleted. Meanwhile, the obtained \( \chi_n = 1.5 \) nm, which is slightly smaller than the estimated value (1.8–2.8 nm) from the Raman data and FN plots, implying the partial charge depletion of MoS\(_2\). While this analysis is reasonable to explain the charge transfer and inversion layer formation, the total calculated depletion width (93.5 nm) is larger than the whole thickness of our device (80 nm). This suggests that the conventional p−n heterojunction model, which is based on the diffusion theory for the intentionally doped inorganic semiconductors with high-carrier mobility,\(^{40,41}\) may not be suitable to describe electronic transports in the vdW junctions. Because of the strong interfacial charge coupling, carrier transports in our unintentionally doped MoS\(_2\)/WSe\(_2\) heterojunction are rather similar to those of the one built from organic semiconductors whose depletion width is relatively thin.\(^{43,44}\)

Another advantage of our vdW heterostructure device is tunability of the interface characteristics by the back-gate modulation which allows for investigating the effect of the 2D inversion layer on the transport property of the junction. Figure 4a shows that the source−drain current increases with two slopes in the regions I and II under the positive back-gate bias because the charge inversion layer in the WSe\(_2\) side is still kept in these regions. At negative gate biases, the depletion layer width becomes narrower in the WSe\(_2\) side and the charge inversion layer disappears, revealing the monotonic current modulation, different from the positive gate bias. From the transfer characteristics, we observed distinct gate bias regimes (Figure 4b). At a low source−drain forward voltage (0.1 V, red line), the current is governed by recombination between the free holes in WSe\(_2\) and free electrons in MoS\(_2\). The drain current then gets a maximum value as the flows of the holes and electrons are matched. At a high source−drain voltage (1.0 V, blue line), the recombination still takes place under the negative gate bias. However, in the positive gate bias side, the current flowing through the junction is dominated by transports of the free electrons in MoS\(_2\) due to tunneling, which is much higher compared to the recombination current. This is one of the advantages of our device as the high on-current leads to a high rectifying factor, which has not been achieved in the reported vdW heterojunctions yet.\(^{45}\) The rectifying factor can be improved in the optimized configuration. For example, using asymmetric electrodes, a low work function metal at the n-MoS\(_2\) side and a high work function one at the p-WSe\(_2\) side can increase the rectifying factor significantly due to efficient carrier injections. The linear graphs (Figure 4c,d) clearly demonstrate the recombination and tunneling.
Figure 5. Photocurrent generation in the heterostructure. (a) $I$–$V$ curves in dark and under white-light illumination. The inset shows a zoomed-in image near the $V_g$ position. (b) Gate-bias-dependent $I$–$V$ curves under light illumination. (c) Short-circuit current ($I_{sc}$) and open-circuit voltage ($V_{oc}$) as a function of gate bias. (d) Gate-bias-dependent photocurrent under the different source–drain biases. (e,f) Band diagrams of the junction under light illumination with the different source–drain voltages and gate biases in the regimes of recombination-limited (RL) and space-charge-limited (SCL) photocurrents. The red and blue circles denote the photogenerated carriers in the WSe$_2$ and MoS$_2$ layers.

Figure 5e,f. At small source–drain voltages, $V_{ds} < V_{oc}$, the photogenerated electrons in MoS$_2$ and photogenerated holes in WSe$_2$ diffuse to the source and drain in the opposite direction with the injected carriers resulting in a negative net photocurrent as in Figure 5e. Applying the negative gate biases renders the enhanced interlayer recombination, and therefore the photocurrent and open-circuit voltage are low as most of the photogenerated carriers are lost. The recombination processes are possible in both mechanisms including Shockley–Read–Hall (defect-assisted recombination) or bimolecular recombination between electrons and holes. The fact that we can clearly modulate the recombination by the back-gate bias implies the resemblance of the bimolecular one. This is reasonable because the binding energy of the interlayer exciton in the vdW heterostructure is hundreds of meV. The maximum recombination corresponds to the minimum values of $J_{sc}$ and $V_{oc}$. Under the positive gate biases, $J_{sc}$ and $V_{oc}$ are higher compared to those in the negative gate biases because of the reduced interlayer recombination due to the existence of the inversion layer. At very high positive gate biases, $J_{sc}$ and $V_{oc}$ are saturated by the space-charge limit. On the other hand, at large source–drain bias, $V_{ds} > V_{oc}$, the photogenerated carriers are drifted in the same direction with the applied electric field. The photocurrent is then positive. The maximum recombination now induces the highest photocurrent since the reduced interlayer recombination forces the photogenerated electrons and holes to move in the same direction with the injected carriers.

Because of the nanometer-scale depletion layer width and the inversion layer located in the WSe$_2$ side, the photocurrent behaviors in our heterojunction are seemingly distinct from the conventional p/n junction structures. The photovoltaic effect with an open-circuit voltage, $V_{oc} \approx 0.34$ V was observed in our device, as in Figure 5a. This value is in excellent agreement with the obtained $V_{bi}$ or tunnel barrier height extracted above. Several distinct behaviors are observed in the real photocurrent, $J_{ph} = J_{light} - J_{dark}$ (Figure 5a): (i) persistent increase of the current at reverse drain voltages, (ii) transition between negative and positive currents at $V_{ds} = V_{oc}$, and (iii) rapid increase of the current at high forward drain voltages. These features cannot be explained by the Shockley diode model but rather are similar to the mechanism observed in the heterojunctions with strong Coulomb interaction such as organic photovoltaic devices.

Because the appearance/disappearance of the charge inversion layer can be controlled by the back-gate bias, the photocurrent generated in the heterojunction is modulated by the gate voltage, as shown in Figure 5b. Under positive gate biases, the junction shows the better factors for a photovoltaic device, including $V_{oc}, J_{sc}$, and fill factor compared to those in the case of applying the negative gate biases. Figure 5c summarizes $J_{sc}$ and $V_{oc}$ values as a function of the gate bias. The deep valleys of $J_{sc}$ and $V_{oc}$ appear at $V_g \sim -18$ V, where the electron and hole currents are matched (as shown in Figure 4c,d). A non-monotonic fluctuation is observed at the gate bias corresponding to the value for the interlayer tunneling in the overlapped WSe$_2$ channel ($V_g \sim -10$ V). Depending on the source–drain voltage and gate bias, the photocurrent can be negative or positive and the minimum negative photocurrent under small $V_{ds}$ transforms to a maximum positive value when $V_g$ increases (Figure 5d).


dependence because the internal electric field in the device helps dissociate electron–hole pairs into free charges.\textsuperscript{55,56} Applying a reverse/forward source–drain voltage can increase/decrease the charge separation efficiency by changing the magnitude of the internal field. In Figure 6, we present the photocurrent of our MoS\textsubscript{2}/WSe\textsubscript{2} heterojunction under different gate biases. Here, $V_G$ is the compensation voltage defined as $J_{ph}(V_G) = 0$, and $V$ is the applied source–drain voltage. The dependence of $J_{ph}$ on $V_G$ under negative and positive gate biases are well fitted with the recombination-limited (RL) and space-charge-limited (SCL) photocurrent mechanisms, which are defined as $I_{ph} = qG\left(\frac{\mu_{h(e)}v_{f(h,e)}}{\epsilon\mu_{h(e)}V}\right)^{0.25}$ and $I_{ph} \leq \left(qG\right)^{0.75}\left(\frac{\mu_{h(e)}v_{f(h,e)}^2}{\epsilon\mu_{h(e)}V}\right)^{0.25}$, respectively.

![Figure 6. Photocurrent dependence on the source–drain voltage under different gate biases. Here, $V_G$ is the compensation voltage defined as $J_{ph}(V_G) = 0$, and $V$ is the applied source–drain voltage.](image)

**CONCLUSION**

We have demonstrated that the strong interfacial charge coupling renders robust interlayer tunneling of charge carriers at the overlapped region of the MoS\textsubscript{2}/WSe\textsubscript{2} vdW heterostructure. An unusually narrow depletion width of a few nanometers is formed in the MoS\textsubscript{2} side, and the charge inversion layer is created on the WSe\textsubscript{2} side. The current transport through the heterointerface is governed by the presence of an inversion layer, in which single carrier tunneling and electron–hole recombination rely on the carrier densities in the two layers. Recombination-limited and space-charge-limited photocurrents were observed in the MoS\textsubscript{2}/WSe\textsubscript{2} heterojunction by controlling the applied gate bias.

Our results suggest important guidelines for the design and optimization of electronic and optoelectronic devices based on vdW structures. For example, one should consider the reduced recombination window but not fall into the space-charge-limited regime for the solar cell, while the light-emitting devices should show better performance under the condition for the optimized bimolecular recombination. The strong interlayer recombination regime also provides opportunities to fabricate IR emitters/detectors with tunable wavelengths by gate modulation. In terms of device design for band-to-band tunneling field-effect transistors based on vdW junctions, an insertion of thin insulator layers at the interface may be necessary to prevent band bending due to interfacial charge transfer. The formation of the 2D inversion layer in our structure contributes to research of quantum physics phenomena emerging from the vdW interfaces such as many-body effect, interface-induced superconductivity, quantum Hall effect, and so on.\textsuperscript{58,59}

**METHODS**

**Device Fabrication.** The MoS\textsubscript{2}/WSe\textsubscript{2} heterostructure device was fabricated on the SiO\textsubscript{2}/Si substrate by using a common dry transfer method.\textsuperscript{5} The thin flakes of MoS\textsubscript{2} and WSe\textsubscript{2} were mechanically exfoliated from the commercial bulk forms (2D Semiconductors, America) using scotch tape. The WSe\textsubscript{2} flakes were first exfoliated on an highly p-doped Si substrate with a 300 nm thick SiO\textsubscript{2} layer. MoS\textsubscript{2} thin flakes were then exfoliated on a PMMA/PVA-coated SiO\textsubscript{2}/Si substrate. The PVA layer was dissolved in water, leaving the MoS\textsubscript{2} flakes embedded in the PMMA. Selected MoS\textsubscript{2} flakes were picked up by a metal bar with a circular hole and stacked on top of WSe\textsubscript{2} by an aligned transfer system integrated with a microscope. To fabricate the transistor device, the electrode patterns were defined using an electron-beam lithography system, and Cr/Au (20/70 nm) metals were deposited by electron-beam-based evaporation at high vacuum. The fabricated device was then annealed at 350 °C for 5 h in Ar\textsubscript{2}/H\textsubscript{2} environment to enhance interfacial contact between the MoS\textsubscript{2} and WSe\textsubscript{2} layers as well as metal electrode/channel contacts prior to measurements.

**Device Characterization.** Thickness and interfacial morphology of the MoS\textsubscript{2}/WSe\textsubscript{2} junctions were investigated by cross-sectional transmission electron microscopy (JEM ARM 200F, JEOL), which also allows for observations of composition mapping and line profiles across the interface using energy-dispersive spectroscopy (EDS) mode. Raman scattering measurements were carried out at room temperature using a 532 nm laser as an excitation source. Electrical characteristics of the fabricated device were conducted at high vacuum ($\sim$10\textsuperscript{-6} Torr) with a Keithley-4200SCS parameter analyzer in dark condition or under white light irradiation with an optical power of $\sim$4 mW/cm\textsuperscript{2}.

3838

DOI: 10.1021/acsnano.7b00021
ACS Nano 2017, 11, 3832–3840
ASSOCIATED CONTENT

 Supporting Information
 The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsnano.7b00021.

 Figures S1–S4 showing cross-sectional TEM images and EDS profile data of the MoS2/WSe2 interface, Raman analysis of the sample in the WSe2-related peaks, and the interlayer tunneling in the electronic transport of the overlapped channels (PDF)

 AUTHOR INFORMATION

 Corresponding Author
 *E-mail: leeyoung@skku.edu.

 ORCID

 Jiong Zhao: 0000-0002-7411-0734
 Seong Chu Lim: 0000-0002-0751-1458
 Young Hee Lee: 0000-0001-7403-8157

 Notes
 The authors declare no competing financial interest.

 ACKNOWLEDGMENTS

 This work was supported by the Institute for Basic Science (IBS, E51104).

 REFERENCES


