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Ultrafast Hot-carrier Photovoltaics of Type-I Monolayer Heterojunctions in the Broad Spectral Ranges

Ji Ho Sung,1,2,† Soonyoung Cha,1,4,† Hoseok Heo,1,2 Sangwan Sim,4 Juho Kim,1,3 Hyunyong Choi,4,* and Moon-Ho Jo,1,2,3,*

1Center for Artificial Low Dimensional Electronic Systems, Institute for Basic Science (IBS), Pohang 37673, Republic of Korea.
2Division of Advanced Materials Science, Pohang University of Science and Technology (POSTECH), Pohang 37673, Korea.
3Department of Materials Science and Engineering, Pohang University of Science and Technology (POSTECH), Pohang 37673, Korea.
4School of Electrical and Electronic Engineering, Yonsei University, Seoul 120-749, Korea

†These authors contributed equally to this work, *To whom correspondence should be addressed: hychoi@yonsei.ac.kr (H. Choi) and mhjo@postech.ac.kr, (M.-H. Jo)

ABSTRACT

Strong interlayer photoresponses in monolayer (ML) semiconductor stacks, such as substantial light absorption and charge separation across interlayer band alignments, suggest potentials for two-dimensional photovoltaics (PVs). Here, we report an interlayer PV conversion in a type-I ML heterojunction by ultrafast interlayer transfer of photoexcited hot carriers in the broad spectral ranges. Specifically, low-energy photoexcitation on a stack of a narrow-bandgap ($E_g$) Bi$_2$Te$_3$ few-layer and a large-$E_g$ MoS$_2$ ML, permits interlayer transfer of transient hot carriers from the Bi$_2$Te$_3$ layer to the excitonic states of the neighbouring MoS$_2$ ML within a time scale of ~ 70 fs, producing interlayer charge separation. Thereby we achieve substantial conversion efficiency from a MoS$_2$ ML with visible to infrared light illumination.
KEYWORDS: Two-dimensional materials, Photovoltaics, Transition-metal dichalcogenide, Hot carrier injection, Ultrafast charge transfer
Efficient light-to-electricity conversion in the broad spectral range is an important step forward in producing next-generation photovoltaics (PVs). Transition-metal chalcogenides (TMChs) exhibit various electronic structures in the atomically thin regime, ranging from a vanishing energy bandgap ($E_g$) in topological insulators to distinct singularities near the band edges in large-$E_g$ semiconductor monolayers (MLs). Those two-dimensional (2D) electronic structures provide diverse excitation and relaxation routes of photoexcited quasiparticles that are spectrally and kinetically distinctive. Such photoresponses can be manipulated in the artificial 2D stacks, where dissimilar TMCh MLs are bound to form new sets of electronic superstructures or band alignments. Previous studies reported that the type-II heterojunctions in large-$E_g$ ML stacks induce charge separation across the band alignments by ultrafast interlayer transfer of photoexcited carriers that are generated by direct-$E_g$ optical transitions in constituent MLs. However, in principle, interlayer charge separation can be also generated in an alternative route by low-energy photo-excitations in other types of the 2D band alignments in designer TMCh ML stacks. Here, we report a type-I 2D stack, composed of a large-$E_g$ MoS$_2$ ML and a narrow-$E_g$ Bi$_2$Te$_3$ few-layers. Therein, the transient hot carriers in Bi$_2$Te$_3$ few-layers that are excited by the sub-$E_g$ (of MoS$_2$ ML) photons are transferred to the $A$ and $B$ excitonic states of the MoS$_2$ ML on an ultrafast time scale of $\sim$70 fs, to drive interlayer charge separation. As a result, the stack operates as a 2D excitonic PV cell in the broad excitation ranges down to near infrared. In our PV cells, the few-layer Bi$_2$Te$_3$ (or Sb$_2$Te$_3$) serves as an efficient hot carrier reservoir in the broad absorption range, which is qualitatively different from usual metals for hot carrier excitations; optically, it possesses a fairly large absorption cross-section from the near-infrared to the ultraviolet range, and electrically, it efficiently creates low-energy hot carriers, whose transient time scale ($\sim$10 ps) is much slower than in normal metals of
These long lived energetic carriers in Bi$_2$Te$_3$ (or Sb$_2$Te$_3$) are attributed to suppressed electron-phonon scattering process with its narrow $E_g$ of ~ 160 meV, which is an order of magnitude larger than the highest phonon energy (~23 meV). The MoS$_2$ ML serves as an efficient photon absorber in the visible range (a direct $E_g$ of 1.89 eV), with the distinct excitonic states. The charge separation at the 2D heterointerfaces in our work is reminiscent of charge transfer excitons at the donor/acceptor heterointerfaces in organic heterojunctions. Although it is perceived that both heterojunction systems suffer from large exciton binding energy due to the poorly screened Coulomb potential, our strategy toward the spectral extension of PV conversion with ultrafast hot carrier transfer may suggest another pathway to achieve higher conversion efficiency as ultrathin planar PV platforms of large-areas.

MoS$_2$ MLs and Bi$_2$Te$_3$ (or Sb$_2$Te$_3$) few-layers (7 nm in thickness or 7 quintuple layers) were separately crystallized by vapor transport synthesis in a 12-in hot-wall quartz tube. Vertical stacks of such 2D crystals were fabricated by a mechanical transfer method on SiO$_2$/Si substrates (Figure 1a) – see also Figure S1. Multiple electrical contacts were made by e-beam lithography in such a way that one can selectively collect photoexcited carriers (photocurrent, $I_{ph}$) within the individual layers or across the stack in a spectroscopic manner by local illumination with a supercontinuum laser. For example, we identified $A$ and $B$ excitonic transitions from the top MoS$_2$ ML in the $I_{ph}$ spectra by local illumination at the MoS$_2$ ML-metal contact area under the finite bias voltage ($V_b$), as in Figure S2, ensuring the crystal quality. We expected a type-I band alignment (the inset of Figure 1b) in the stack due to the fact that the work function of chemical vapor deposition grown few-layers Bi$_2$Te$_3$ (~4.3 eV) is larger than the electron affinity of MoS$_2$ (~4.0 eV), and found such a signature from the rectifying $I_{dark}$-$V$
characteristics (Figure 1b). This band offset can be electrically tuned by applying the $V_g$. The forward $I_{dark}$ of the stack is strongly enhanced with the positive $V_g$, due to the $n$-type semiconductor character of MoS$_2$ ML. Of principal interest in this work is the interlayer charge separation upon light illumination (the PV responses), which can be spectrally captured with a pair of MoS$_2$ and Bi$_2$Te$_3$ contacts. Figure 1c shows the $I_{ph}$-$V$ characteristics upon the local illumination of monochromatic light on the junction area at various excitation energies; at $h\nu = 2.33$ eV the PV responses are clearly marked by the distinct open circuit voltage ($V_{oc}$) and short circuit current ($I_{sc}$). We found that the PV response persists under the light illumination down to the near IR range of 1.17 eV, which is far below the $E_g$ (1.89 eV) of MoS$_2$ MLs. This indicates that the interlayer charge separation is efficiently achieved across the Bi$_2$Te$_3$-MoS$_2$ junction in the broad absorption range. At $h\nu < 1.89$ eV, because the top MoS$_2$ MLs are completely transparent, the charge separation is solely achieved by the low-energy ($i.e., h\nu < E_g$ of MoS$_2$ MLs) carrier excitation in the bottom Bi$_2$Te$_3$ layers, followed by interlayer transfer. That is, provided that the photoexcited electrons in Bi$_2$Te$_3$ layers efficiently transfer to MoS$_2$ MLs, leaving holes behind, the spatial charge separation is established across the heterointerfaces (Figure 1d). Hereafter, we termed these photoexcited electrons in Bi$_2$Te$_3$ layers as hot electrons (carriers), because the excitation photon energy in this study is far greater than the $E_g$ of Bi$_2$Te$_3$ ($\sim$160 meV). At $h\nu > 1.89$ eV, the photocarrier generation is additionally made by the interband optical transitions in the top MoS$_2$ MLs, followed by the interlayer transfer of holes, to establish the interlayer charge separation. In both cases, we observed the similar magnitude of $V_{oc}$ ($\sim 0.2$ eV), which is very close, but slightly larger than the $E_g$ of Bi$_2$Te$_3$ layers. This observation is consistent with the PV principle that the $V_{oc}$ of the stack-type cells is fundamentally limited by the quasi-Fermi level difference between Bi$_2$Te$_3$ layers and MoS$_2$ MLs.$^1$ Qualitatively the same
PV responses were observed in MoS$_2$ MLs/Sb$_2$Te$_3$ few-layers stacks, which is another narrow-$E_g$ semiconductor with the similar band alignment (Figure S2-4).

The observed interlayer charge transfer characteristics was investigated in more details by ultrafast optical pump and white-light probe spectroscopy (Figure 2).$^{43,44}$ We choose the pump-photon energy ($h\nu_{\text{pump}} = 0.99$ eV) on MoS$_2$/Bi$_2$Te$_3$ stacks to be far below the $E_g$ of MoS$_2$, such that the pump pulse generates hot carriers solely in the Bi$_2$Te$_3$ layers. Two sets of experiments were performed. In the first experiment (the upper panel of Figure 2a), we determined the transient absorption responses ($\Delta T/T_0 < 0$, where $\Delta T$ is the pump-induced transmission change, and $T_0$ is the transmission without the pump) of Bi$_2$Te$_3$ only (red dots) and the $\Delta T/T_0$ of MoS$_2$/Bi$_2$Te$_3$ stacks (black dots), measured at the probe-photon energy $h\nu_{\text{probe}}$ of 1.74 eV. Because $h\nu_{\text{probe}}$ is also below (above) the $E_g$ of MoS$_2$ (Bi$_2$Te$_3$), one can expect that the photoresponses of Bi$_2$Te$_3$ layers should be different from those of MoS$_2$/Bi$_2$Te$_3$ stacks.$^7$ By comparing the decay dynamics of the transients in the two sets of samples, the effect of the photoexcited hot carrier transfer time from Bi$_2$Te$_3$ to MoS$_2$ can be definitively determined. They show similar rising dynamics within the time-resolution of our probe pulse ($\sim 50$ fs).$^7$ While the Bi$_2$Te$_3$ transient shows a single-exponential decay of 6.5 ps, the MoS$_2$/Bi$_2$Te$_3$ stacks exhibit an extremely fast $\tau_1$ of 70 fs with decreased differential absorption, where the transient can fit with fast ($\tau_1$) and slow ($\tau_2$) decay components in a bi-exponential function. Then, the transient relaxes with a slow $\tau_2$ of 6.5 ps. Here, we emphasize that this rapid absorption quenching ($\tau_1 \approx 70$ fs) occurs only in the MoS$_2$/Bi$_2$Te$_3$ stacks, and no such fast decay component was observed in the Bi$_2$Te$_3$ layers. The measured slow decay time is consistent with previous studies of slow cooling dynamics in the Bi$_2$Te$_3$ layers and fast decay time of $\sim 70$ fs is even faster than the time needed to
recover Fermi-Dirac distribution in the Bi$_2$Te$_3$ layers.$^{30,31}$ The fact that no pump-probe signal was observed in MoS$_2$ MLs (blue dots in the lower panel of Figure 1a) further corroborates that the hot electron transfer takes place only in the MoS$_2$/Bi$_2$Te$_3$ stacks. In the second experiment, this hot-carrier transfer was confirmed by scanning the probe-photon energy across the $A$ and $B$ exciton energies of MoS$_2$ MLs. Figure 2b shows the spectrally resolved dynamics in the MoS$_2$/Bi$_2$Te$_3$ stacks (black circle) as a function of the pump-probe delay. Under the same pump energy and fluence condition, the stack shows negative transmission changes ($-\Delta T/T_0$) over the probe photon energies, i.e., increased absorption. In particular, we observed that the absorption changes in the stacks are significantly reduced at two specific probe energies, which are exactly matched with the $A$ and $B$ exciton energies of MoS$_2$, that is, 1.89 and 2.1 eV, respectively.$^{46}$ The dynamics of Bi$_2$Te$_3$ are spectrally featureless,$^{47}$ regardless of the pump-probe delay (red circle), which can be regarded as the intralayer pump-induced hot carrier generation and cooling within the Bi$_2$Te$_3$ layers.$^{26,48}$ Thus, our observations in the pump-probe measurements provide direct evidence of the charge separation, whose ultra-fast kinetics originates from the photoexcited hot electron transfer from the Bi$_2$Te$_3$ layers to the $A$ and $B$ excitonic population states of the MoS$_2$ MLs (Figure 2c).

Figure 3a is the $I_{ph}$ spectrum (the red trace) collected with a pair of MoS$_2$ and Bi$_2$Te$_3$ contacts under zero-bias voltage ($V_b$), thus representing the spectral $I_{sc}$ of the stacks – see also Figure S2 for the MoS$_2$ and Sb$_2$Te$_3$ stacks, which shows essentially the same spectral responses. We clearly verified that the PV $I_{sc}$ persist down to the near IR range of 0.75 eV, comparing with the $I_{ph}$ spectra collected by local illumination at the metal-MoS$_2$ ML contacts (thus representing the metal junction photoresponses of the MoS$_2$ ML alone). This observation is consistent with the
suggested interlayer charge separation mechanism of Figure 1c and d. The interlayer PV responses at the heterointerfaces are also examined by the spatial mapping of the $I_{sc}$ in the stack (Figure 3b-d) by scanning a focused laser – see also Figure S3. We immediately identified that the $I_{sc}$ generation is localized in the stack region over the spectral range, suggesting the interlayer charge separation is the key element to the PV conversion, although the $I_{sc}$ intensity is not homogeneously distributed over the stack region, presumably due to the non-uniform contact of the two layers during the manual transfer.49

Earlier reports on monolayer semiconductor PV cells are typically based on either intralayer (lateral) or interlayer (vertical) $p$-$n$ junctions.19-25 By contrast, the heterojunction cells operating by the hot carrier injection across the type-I van der Waals interfaces has been rarely addressed. We discuss the light-to-power conversion of our stacks (of the effective junction area of 50 $\mu m^2$) in comparison with earlier works of ML $p$-$n$ junctions. Figure 4a is the PV $I$-$V_b$ characteristics under various light illumination intensities at $h\nu = 2.33$ eV. The inset is the generated power ($P_{pv}$), which is quadratic to the applied $V_b$. We measure the maximum power to be over 600 pW from the MoS$_2$/Bi$_2$Te$_3$ stacks (and 5,000 pW from the MoS$_2$/Sb$_2$Te$_3$ stacks in Figure S4) under 170 $\mu W$ light illumination. Typically, the generated power is ranged from 100 pW to 1 nW from more than 10 devices, and is comparable to those of the earlier MoS$_2$ intra-ML $p$-$n$ PV cells.50,51,52 Considering the measured power loss due to the lateral charge transport in our stacks, it suggests that the charge separation is reasonably efficient by hot carrier transfer in the stack. Remarkably, such PV responses persist down to $h\nu = 1.17$ eV, as in Figure 4c, with a $P_{pv}$ of 30 pW under 880 $\mu W$ light excitation. Last, we report the external quantum efficiency, $\eta_{ext} = (I_{ph}/e)/(P_{opt}/h\nu)$, when $V_b = 0$, $I_{ph} \sim I_{sc}$. Figure 4b and d are such EQEs for the MoS$_2$/Bi$_2$Te$_3$ stack upon light illumination
above and below the $E_g$ of MoS$_2$ MLs, which are electrically tuneable by the $V_g$. The maximum EQE was measured to be $\sim 10^{-4}$ at $h\nu = 2.33$ eV. It also persists at $h\nu = 1.17$ eV, albeit with values lower by two orders of magnitude. Note that these EQE values can be converted to photon conversion efficiency (PCE), defined as $PCE = (J_{ph} \cdot hc)/(I_{hv} \cdot \lambda e)$, of 20% at the excitation of 2.33 eV and 0.2% for 1.17 eV, where $J_{ph}$ is the photocurrent density, $h$ is the Planck constant, $c$ is the speed of light, $I_{hv}$ is the areal light intensity, $\lambda$ is the wavelength, and $e$ is the elementary charge. Rather poor quantum efficiency of our device is due to lateral charge collection, and it can be overcome by vertical charge collection scheme made with MoS$_2$/Bi$_2$Te$_3$ vertical stack between two graphene layers.$^{25}$ Recent experimental finding suggests clever way of extending lifetime ($\sim 50$ ps) of excited carriers in topological insulators by photoinduced electron-hole asymmetry which can be used for high-efficiency 2D hot-carrier photovoltaic cells.$^{53}$

**METHODS**

**Materials: Vapour Transport Synthesis of 2D Bi$_2$Te$_3$ and Sb$_2$Te$_3$ crystals**

2D Bi$_2$Te$_3$ and Sb$_2$Te$_3$ crystals of few QLs were grown by vapour transport synthesis from solid powder precursors. High-purity Bi$_2$Te$_3$ or Sb$_2$Te$_3$ powders were placed in alumina boats and loaded in the centre of a 12-in. hot-walled quartz-tube furnace. SiO$_2$ (300 nm)/Si wafers as the growth substrates were also placed 10 cm downstream from the tube centre, considering the temperature gradient along the tube furnace. Prior to heating, the furnace was evacuated to $10^{-3}$ Torr and purged with high-purity nitrogen (N$_2$) for 30 min to eliminate residual oxygen. Then, the furnace was heated to 530 °C (for Sb$_2$Te$_3$) or 540 °C (for Bi$_2$Te$_3$) for 30 min under 5 sccm of high-purity argon (Ar) flow as a carrier gas under a total pressure of 0.1 Torr.
Spatially and spectrally resolved photocurrent spectroscopy

For the spectral photocurrent measurements, we used a broadband supercontinuum laser (450 nm ≤ λ ≤ 2000 nm) combined with a monochromator for the high-resolution spectra. During the wavelength scanning, the photocurrent was measured by a lock-in technique with a chopper frequency of 500Hz and subsequently normalized to the photon flux. The chopped laser beam was focused by a microscopic lens (NA = 0.8) and illuminated the ML channel region of the devices.

Ultrafast pump-probe spectroscopy

To perform the ultrafast pump-probe spectroscopy, femtosecond pulses at photon energy of 1.55 eV were generated by a 250 kHz Ti:sapphire regenerative system (Coherent RegA9050). The laser output pulse was separated into two pulses by a ratio of 7:3. The 70% laser pulse was injected into an optical parametric amplifier (Coherent OPA 9850) to generate infrared pump pulses at a photon energy of 0.99 eV. The other was focused into a 1 mm-thick sapphire disk to create a white-light supercontinuum source, which served as a probe pulse. To suppress the scattering-induced artefact, the pump and probe pulses were focused on the sample with non-collinear geometry. The spot size of our pump and probe beams were 100 µm and 50 µm, respectively. The spectrally-resolved transmission of probe pulses was obtained by Si photodetector and monochromator (Newport 74125), and the pump-induced transient differential signal was recorded using a lock-in amplifier (SR850) with a chopping frequency of 10 kHz. During measurements, the pump-probe delay was controlled using a computer-controlled mechanical delay stage (Newport M-IMS300LM).
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Supporting Information Available: This material is available free of charge via the Internet at http://pubs.acs.org.
Figure 1. (a) Schematic diagram and an optical image of the vertically stacked MoS$_2$/Bi$_2$Te$_3$ heterointerface. Scale bar is 5 μm. (b) Dark I-V curves of the MoS$_2$/Bi$_2$Te$_3$ device at various back gate voltages measured across the heterojunction with a pair of MoS$_2$ and Bi$_2$Te$_3$ contacts. Inset: Energy band alignment (type-I alignment) of the heterojunction. (c) Photoresponse characteristics under 2.33 eV (left y-axis), 1.38 eV, and 1.17 eV (right y-axis) light illumination at the heterointerface of the MoS$_2$/Bi$_2$Te$_3$ vertical stack device. (d) Band alignments and charge separation at the MoS$_2$/Bi$_2$Te$_3$ heterointerface. Upon light illumination of $h \nu < 1.89$ eV (the $E_g$ of MoS$_2$ ML), interlayer charge transfer is attributed solely to the sub-$E_g$ excited hot carrier transfer across the two layers. At $h \nu > 1.89$ eV light illumination, interlayer charge separation is dominated by the intralayer interband transitions of the MoS$_2$ ML followed by charge transfer.

Figure 2. (a) Time-resolved ultrafast transmission changes ($-\Delta T/T_0$) in the MoS$_2$/Bi$_2$Te$_3$ stacks (black), Bi$_2$Te$_3$ layers (red), and MoS$_2$ MLs (blue). The measurements were performed at room temperature with $h \nu_{\text{probe}}$ of 1.74 eV (below the $E_g$ of MoS$_2$ ML) under a pump fluence of 970 µJ/cm$^2$. The solid lines (red and black) are the exponential fits multiplied by a Heaviside step function. (b) Spectrally resolved $-\Delta T/T_0$ spectra of the MoS$_2$/Bi$_2$Te$_3$ heterojunction (black) and Bi$_2$Te$_3$ layers (red) after 0.99eV optical excitation. Note that the transient spectra in the MoS$_2$/Bi$_2$Te$_3$ heterojunctions exhibit pronounced dips at A and B excitonic resonances (dashed lines), which are direct evidence of interlayer charge transfer from the Bi$_2$Te$_3$ layers to the MoS$_2$ MLs. (c) Illustration for hot carrier transfer from Bi$_2$Te$_3$ layers to A and B excitonic states of the MoS$_2$ ML in the pair momentum space.

Figure 3. (a) Optical power normalized local $I_{\text{ph}}$ spectra of a MoS$_2$/Bi$_2$Te$_3$ heterojunction device
under zero-bias voltage with the pair of MoS$_2$ and Bi$_2$Te$_3$ contacts (red line) and two MoS$_2$
contacts (blue line). $I_{ph}$ under zero-bias voltage between two MoS$_2$ contacts is generated in
metal-MoS$_2$ junction area. (b) Optical image of the MoS$_2$/Bi$_2$Te$_3$ stack device. (c-d) Scanning $I_{sc}$
images of the MoS$_2$/Bi$_2$Te$_3$ heterojunction device under (c) 2.33 eV and (d) 0.94 eV light
illumination. The $I_{sc}$ is generated on the entire area of MoS$_2$/Bi$_2$Te$_3$ heterojunction which shows
that $I_{sc}$ by vertical interlayer transport dominates, rather than by the lateral transport.

Figure 4. (a,c) $I-V$ curves under (a) 2.33 eV and (c) 1.17 eV optical excitation of the
MoS$_2$/Bi$_2$Te$_3$ stacks with various laser powers. Inset: Generated PV power ($P_{pv} = I_{ds}V_{ds}$) as a
function of $V_b$ for different excitation optical powers. (b,d) External quantum efficiency under (b)
2.33 eV excitation for $V_b = 0$ at various gate voltages and (d) under 1.17 eV excitation.
REFERENCES


It is indeed a rather complex process due to a nontrivial band structures of Bi$_2$(Sb$_2$)Te$_3$ few MLs, as another subject of our future study, yet to be precisely determined including both carrier scattering and phonon scattering with/without topological surface states. At the moment, it is beyond to this study.


Figure 1. (a) Schematic diagram and an optical image of the vertically stacked MoS\(_2\)/Bi\(_2\)Te\(_3\) heterointerface. Scale bar is 5 µm. (b) Dark \(I-V\) curves of the MoS\(_2\)/Bi\(_2\)Te\(_3\) device at various back gate voltages measured across the heterojunction with a pair of MoS\(_2\) and Bi\(_2\)Te\(_3\) contacts. Inset: Energy band alignment (type-I alignment) of the heterojunction. (c) Photoresponse characteristics under 2.33 eV (left \(y\)-axis), 1.38 eV, and 1.17 eV (right \(y\)-axis) light illumination at the heterointerface of the MoS\(_2\)/Bi\(_2\)Te\(_3\) vertical stack device. (d) Band alignments and charge separation at the MoS\(_2\)/Bi\(_2\)Te\(_3\) heterointerface. Upon light illumination of \(h\nu < 1.89\) eV (the \(E_g\) of MoS\(_2\) ML), interlayer charge transfer is attributed solely to the sub-\(E_g\) excited hot carrier transfer across the two layers. At \(h\nu > 1.89\) eV light illumination, interlayer charge separation is dominated by the intralayer interband transitions of the MoS\(_2\) ML followed by charge transfer.
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Figure 4. (a,c) $I$-$V$ curves under (a) 2.33 eV and (c) 1.17 eV optical excitation of the MoS$_2$/Bi$_2$Te$_3$ stacks with various laser powers. Inset: Generated PV power ($P_{pv} = I_{ds}V_{ds}$) as a function of $V_b$ for different excitation optical powers. (b,d) External quantum efficiency under (b) 2.33 eV excitation for $V_b = 0$ at various gate voltages and (d) under 1.17 eV excitation.

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