Extraordinary Sunlight Absorption and One Nanometer Thick Photovoltaics Using Two-Dimensional Monolayer Materials

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ABSTRACT: Graphene and monolayer transition metal dichalcogenides (TMDs) are promising materials for next-generation ultrathin optoelectronic devices. Although visually transparent, graphene is an excellent sunlight absorber, achieving 2.3% visible light absorbance in just 3.3 Å thickness. TMD monolayers also hold potential as sunlight absorbers, and may enable ultrathin photovoltaic (PV) devices due to their semiconducting character. In this work, we show that the three TMD monolayers MoS2, MoSe2, and WS2 can absorb up to 5–10% incident sunlight in a thickness of less than 1 nm, thus achieving 1 order of magnitude higher sunlight absorption than GaAs and Si. We further study PV devices based on just two stacked monolayers: (1) a Schottky barrier solar cell between MoS2 and graphene and (2) an excitonic solar cell based on a MoS2/WS2 bilayer. We demonstrate that such 1 nm thick active layers can attain power conversion efficiencies of up to ∼1%, corresponding to approximately 1–3 orders of magnitude higher power densities than the best existing ultrathin solar cells. Our work shows that two-dimensional monolayer materials hold yet untapped potential for solar energy absorption and conversion at the nanoscale.

KEYWORDS: Monolayer materials, graphene, transition metal dichalcogenides, solar energy, sunlight absorption, photovoltaics

Following the pioneering work from Geim and colleagues showing the mechanical exfoliation of graphene with the Scotch tape method,1 monolayers of BN,2 hybrid graphene-BN,3 MoS2,4,5 MoSe2,6 and WS2,7 have been prepared by exfoliation and chemical vapor deposition. While technical barriers toward large-scale synthesis of monolayer materials continue to be overcome, studies of micrometer-scale flakes complemented by theoretical calculations have already highlighted a host of novel optical and electronic properties in these materials.8–11 For example, the three transition metal dichalcogenides (TMDs) MoS2, MoSe2, and WS2 undergo a crossover from indirect to direct gap when going from bilayer to monolayer,12 resulting in enhanced monolayer photoluminescence.6,7,13 Although graphene has been used in optoelectronics and photovoltaics (PV) as a transparent contact, it displays an extraordinary absorbance of 2.3% in the visible considering its thickness of only 3.3 Å.14 Such an absorbance is equivalent to that of approximately 20 nm thick Si or 5 nm thick GaAs, namely, two of the most commonly used absorbers in solar cells.15 This high optical absorption of graphene and other two-dimensional monolayers makes such materials appealing for solar energy conversion.16,17 Although it may appear counterintuitive to use graphene as a key active material in PV given its transparency and metallic behavior, the idea of coupling a semiconducting TMD monolayer with graphene to create a bilayer Schottky barrier solar cell is viable. In addition, solar cell active layers obtained by stacking two different TMD monolayers are also possible. The high carrier mobility of monolayer materials, including values of over 200 000 cm2/V·s for suspended graphene8 and up to 200 cm2/V·s in monolayer MoS2,9,10 also make them appealing for PV applications. Such high mobilities coupled to the possibility of making ultrathin solar cells can lead to monolayer-based PV with very low series resistance, large voltages, and near-optimal I–V curves.

Here we study the feasibility and predict the performance of 1 nm thick solar cells based on bilayers of MoS2/graphene or two stacked TMD monolayers. First, we use a combination of first principles calculations based on density functional theory (DFT) and the GW-Bethe Salpeter method18–20 to compute accurate absorbance spectra for monolayers of MoS2, MoSe2, WS2, and graphene, showing quantitative agreement with available experimental measurements for MoS2 and graphene. Our calculations show that a single TMD monolayer with subnanometer thickness can absorb as much sunlight as 50 nm of Si and generate electrical currents as high as 4.5 mA/cm2.
Next, we compute the band alignment in bilayers of MoS$_2$/graphene and MoS$_2$/WS$_2$ and demonstrate that both these interfaces can realize PV operation by formation of a Schottky barrier and a type-II heterojunction, respectively. Using conservative assumptions, we compute that such bilayer solar cells can reach over 1% power conversion efficiency (PCE) in just 1 nm thickness, thus packing a power density of up to 2.5 MW/kg, a value that is far superior to any known energy conversion or storage device. These calculations illustrate new avenues for nanoscale solar energy conversion using TMD monolayers and graphene.

Accurate computation of optical absorption in TMD monolayers (Figure 1a) is challenging due to a number of technical reasons. In this work, we employ DFT calculations, the GW method,$^{19}$ and the Bethe–Salpeter equation (BSE)$^{20}$ to obtain the macroscopic dielectric tensor including an accurate account of electron–electron and electron–hole interactions, as well as spin–orbit and semiconductor state effects. The calculations were carried out using the VASP$^{21,22}$ and Yambo$^{23}$ codes and are detailed in the Supporting Information.

Figure 1b compares our computed absorbance for a monolayer of MoS$_2$ with the experimental measurements of Mak et al.$^{12}$ The quantitative agreement observed between the computed and experimental absorbance highlights the accuracy of our approach. Figure 1c shows the absorbance calculated for the three TMD monolayers MoS$_2$, MoSe$_2$, and WS$_2$, compared to the absorbance of graphene (as computed here, in good agreement with the flat 2.3% value measured by Nair et al.$^{14}$) and to the incident AM1.5G solar spectrum.$^{24}$ We predict that TMD monolayers possess a high absorbance of 5–10% in the visible, and can thus capture a significant fraction of incident sunlight in a subnanometer thickness. Table 1 compares the absorbed photon flux $J_{\text{abs}}$ in graphene, the three TMD monolayers studied here, and 1 nm thick layers of materials commonly used in thin-film inorganic (Si and GaAs) and organic (P3HT polymer) solar cells, whose bulk absorption spectrum was taken from experimental data in the literature.

For each material, $J_{\text{abs}}$ is calculated using the absorbance $A$ from the integral:

$$J_{\text{abs}} = \epsilon \int_{E_g}^{\infty} A(E) J_{\text{ph}}(E) \, dE$$

(1)

where $E_g$ is the incident photon flux (units of photons/cm$^2$·s·eV), $A\text{E}_E$ the incident photon flux (units of photons/cm$^2$·s·eV), and $E$ is the photon energy. By multiplying by the elementary charge $\epsilon$, the absorbed photon flux is expressed as the equivalent short-circuit electrical current density (units of mA/cm$^2$) in the ideal case when every photon is converted to an electron–hole pair in a PV device, so that $J_{\text{abs}}$ sets the upper limit for the contribution of the single material to the solar cell short-circuit current. We remark that this choice of units represents nothing more than a convenient way to quantify sunlight absorption and is fully equivalent to expressing the results in terms of the absorbed photon flux.

Our results indicate that subnanometer thick graphene and TMD monolayers can absorb photon fluxes equivalent to short-circuit currents of 2–4.5 mA/cm$^2$, while 1 nm thick Si, GaAs, and P3HT all generate currents in the 0.1–0.3 mA/cm$^2$ range. For example, approximately 15 nm of GaAs or 50 nm of Si are needed to absorb the same fraction of sunlight as a TMD monolayer such as MoSe$_2$. While the absorbance of graphene is regulated by the fine structure constant,$^{14}$ we discuss here the origin of the large absorbance in TMD monolayers. The valence and conduction bands energetically close to the gap in TMD monolayers are dominated by localized $d$ states of the transition metal atoms.$^{13,26}$ Within an independent-particle treatment, the high optical absorption at visible energies in
TMD monolayers can be explained by dipole transitions with large joint density of states and oscillator strengths between localized $d$ states with strong spatial overlap; such transitions are dipole-allowed in a regime of weak spin–orbit coupling, as exhibited by MoS$_2$ and other TMD systems. This observation that a major role is played by strong dipole transitions between localized $d$ states is supported by recent work of Britnell et al., although we note that such an independent-particle picture is only partially complete. In particular, excitonic effects in TMD monolayers resulting from the poor screening of electrons and holes due to the vacuum surrounding the monolayer, give rise to a strong mixing of electron–hole configurations in the excited-state wave function. This results in a constructive superposition of the oscillator strengths for transitions at low energies near the absorption onset, as also found in other semiconductors. To quantify the different contributions giving rise to the large optical absorption at visible energies for the case of MoS$_2$, we compare two different approaches for computing the absorbance in Figure S1: the independent-particle absorbance computed using DFT with the random phase approximation (DFT-RPA) which includes only dipole transitions, and the absorbance shown in Figure 2b obtained using BSE which explicitly includes electron–hole interactions. As shown in Figure S1, excitonic effects increase the absorbance at visible energies by a factor of $\sim 2$ compared to DFT-RPA. This result suggests that, while part of the reason for the strong absorption in monolayer MoS$_2$ is due to the dipole transition between localized $d$ orbitals contributing to visible absorbances in the $2$–$5\%$ range, another important contribution arises from excitonic coupling of such transitions and is responsible for increasing the absorbance further up to $5$–$10\%$. A similar behavior is found for the other TMD monolayers studied in this work.

It is important to compare the absorption in graphene and TMD monolayers with those of their bulk counterparts—respectively, graphite and bulk TMDs—composed by stacked multilayers held together by van der Waals forces. We focus in this comparison on visible photon energies in the $1$–$2.5$ eV range of key relevance for photovoltaics, and for TMD monolayers analyze the case of MoS$_2$ for which accurate absorption experiments are available for the bulk. Monolayer MoS$_2$ has an absorbance of $A \approx 5$–$10\%$ in a thickness $\Delta z = 6.5$ Å. Although strictly speaking one cannot define a macroscopic absorption coefficient in the layer-normal direction for a single layer of MoS$_2$ (since by definition this quantity should be averaged over several unit cells of the material), the equivalent absorption coefficient $\alpha$ for monolayer MoS$_2$ can be obtained as $\alpha = A/\Delta z = 1.5 \times 10^6$ cm$^{-1}$. Similar values are found for the case of graphene ($A = 2.3\%$, $\Delta z = 3.3$ Å, and thus $\alpha = 0.7 \times 10^6$ cm$^{-1}$). These absorption coefficients are higher than those found in bulk MoS$_2$ and graphite at visible energies of up to $2.5$ eV: for bulk MoS$_2$, experimental measurements suggest $\alpha$ values in the $0.1$–$0.6 \times 10^6$ cm$^{-1}$ range, while for graphite experiments suggest $\alpha$ values of $0.2$–$0.4 \times 10^6$ cm$^{-1}$ (see ref 31). The absorption values are thus higher by a factor of $2$–$3$ for both graphene and MoS$_2$ monolayers compared to their bulk counterparts. To allow a direct comparison of monolayer and bulk MoS$_2$ with our calculation approach, we computed the absorption coefficient of bulk MoS$_2$ using BSE (see Figure S2 in Supporting Information) and find excellent agreement with the experimental absorption values of $0.1$–$0.6 \times 10^6$ cm$^{-1}$ in ref 30. Experimental measurements for bulk GaAs—which is used here as a reference material with high visible absorption—also show $\alpha$ values in the $10^5$–$10^6$ cm$^{-1}$ range. We note that, despite the linear scaling in absorbance versus number of layers for up to $4$–$5$ layers of MoS$_2$ and graphene, the different values of optical absorption in the bulk versus monolayer forms

Figure 2. MoS$_2$/graphene interface and Schottky barrier solar cell. (a) The MoS$_2$/graphene solar cell described in this work. M1 and M2 are, respectively, low and high workfunction metals. The MoS$_2$/graphene junction is shown enclosed in a dashed box. The polarity of the electrodes is also shown. (b) Band alignment at a MoS$_2$/graphene interface, as predicted using DFT. $p$-SB is the hole Schottky barrier; also shown are the valence and conduction band edges ($E_v$ and $E_c$, respectively) of MoS$_2$ as well as the quasi-Fermi levels for electrons and holes under illumination ($E_{FF}$ and $E_{FF}p$ shown as red dashed lines), together with the direction of electron and hole diffusion. $V_{\text{max}}$ is the maximum open-circuit voltage. (c) PDOS of a MoS$_2$/graphene interface. The energy is referenced to the Fermi energy. (d) Absorbance of the MoS$_2$/graphene interface and its composing monolayers, computed within the independent particle approximation using DFT.
suggest that after stacking a sufficiently high number of layers a “bulk-like” behavior emerges. For graphene, deviation from this linear dependence of absorbance versus number of layers has been observed beyond 4–5 layers,14 while for TMD this linear dependence has been shown only for up to 3–4 layers.12 We suggest that further work is necessary to establish the difference and the transition between monolayer and bulk behavior in layered materials.

The exceptional sunlight absorption of graphene and TMD monolayers discussed so far suggests the possibility to design 1-nm-thick solar cells based on just two stacked monolayers, a topic we develop in the remainder of the paper. The first device we examine is based on a bilayer of MoS2/graphene as the active layer material. Figure 2a shows a possible geometry for a solar cell formed using a bilayer of MoS2/graphene interfaced to a high workfunction metal on the MoS2 side and a low workfunction metal on the graphene side. Since graphene is a (semi)metal and MoS2 is a semiconductor, in order for such a device to work electron–hole pairs generated in either material composing the interface should be separated through the formation of a Schottky barrier (SB). We employ DFT combined with the lineup method32 to study the formation of a SB at an interface between graphene and a defect-free, undoped layer of MoS2 (see Supporting Information). The first step in this calculation is the determination of the work functions of MoS2 and graphene. Using DFT, we computed a workfunction value of $\phi_{\text{MoS2}} = 5.2$ eV for monolayer MoS2, in agreement with the recent experimental observation of ohmic contact between n-type MoS2 and Au,4 and a workfunction of $\phi_G = 4.25$ eV for graphene, which is close to commonly measured values in the 4.3–4.6 eV range. Our calculations further predict the formation of a SB of 1.2 eV for holes to diffuse from graphene to MoS2, thus enabling the design of SB solar cells between (preferably p-type) MoS2 and graphene. Charge separation occurs by injecting photogenerated electrons from the conduction band of MoS2 to graphene, while holes photogenerated in the valence band of MoS2 cannot diffuse to graphene due to the large SB (see Figure 2b). Notably, electron injection from MoS2 to graphene upon illumination as predicted here has been observed by recent experiments in MoS2/graphene phototransistors.33 Under these operating conditions, the maximum open circuit voltage $V_{OC,max}$ is set by the difference between the SB and the built-in potential, given that holes are extracted at the VBM in MoS2 (namely, the maximum quasi-Fermi hole level that can be reached under illumination). We estimate a relatively small $V_{OC,max} \approx 0.3$ eV, obtained as the difference between the computed SB of 1.2 eV and the built-in potential $\phi_{\text{MoS2}} - \phi_G \approx 0.9$ eV.34

We remark that our calculation assumes that the metallic electrode contacting the MoS2 monolayer is placed sufficiently far (i.e., at least one screening length away) from the MoS2/graphene junction (Figure 2a), so that the Fermi energy near the contact recovers its value in monolayer MoS2 unaffected by the interface dipole induced by graphene. This assumption justifies the alignment shown in Figure 2b, where band bending in MoS2 occurs in a direction parallel to the monolayer when moving away from the junction area toward the electrode contacting MoS2. Our proposed geometry with the electrodes spatially separated from the junction further reduces the risk of leakage currents potentially occurring if the bilayer solar cell is placed between two metallic electrodes separated by just 1 nm. While our calculations suggest that a p-type doping is preferable in MoS2, some experiments have shown that MoS2 deposited on SiO2 shows n-type behavior.4 However, recent calculations suggest that the doping type in MoS2 may not be intrinsic but rather due to impurities or defects at the SiO2 surface,35 as further supported by experiments reporting p-type behavior in MoS2.36

Figure 2c–d shows other important aspects of the MoS2/graphene bilayer proposed here. Upon formation of the interface, our calculations suggest a redistribution of the ground state charge in the system, as also concluded in a recent study by Ma et al.36 Despite this effect, the projected density of states (PDOS) shows that the electronic states of graphene and MoS2 do not hybridize near the Fermi energy (Figure 2c). In addition, the absorbance at visible photon energies (computed here using DFT within the independent particle approximation44 due to the large size of the simulation cell) is equal to the sum of the absorbances of isolated graphene and the MoS2 monolayer (Figure 2d). Using the absorbance spectrum in Figure 2d and eq 1, we estimate a maximum short-circuit current of $J_{sc} = 4.3$ mA/cm2 for a MoS2/graphene active layer. This value differs slightly from the sum of the monolayer currents in Table 1 (5.9 mA/cm2) due to the use of different levels of theory (BSE in Table 1 and independent-particle in Figure 2d); for our purpose here it suffices to establish that a maximum short-circuit current of $\sim 4.5$ mA/cm2 can be achieved. We estimate a range of PCE values for the MoS2/graphene solar cell in Figure 2a by using $J_{sc} = 4.5$ mA/cm2 combined with: (1) for the lower PCE limit, an open circuit voltage $V_{OC} = 0.1$ V and a small fill factor (FF) of 0.3, namely, a reasonable value for a “poor” solar cell device with high series and low shunt resistances, and (2) for the upper PCE limit, $V_{OC} = 0.5$ V and FF = 0.6 as a reasonable FF value in a device with effective carrier transport.37 In all cases, a conservative value of 0.7 is assumed for the internal quantum efficiency (IQE, namely the fraction of absorbed photons extracted as carriers at the contacts), a value significantly lower than the best Si and GaAs solar cells achieving IQE close to unity. This approach corresponds to assuming that only 70% of the absorbed photons contribute to the current, resulting in a short-circuit current $J_{SC} = 0.7 \cdot J_{sc}$ in the solar cell device. We compute the PCE under AM1.5G illumination by dividing the product $J_{SC} \times V_{OC} \times FF$ through the incident power of 100 mW/cm²,38 resulting in a PCE range of 0.1–1.0% for a bilayer of MoS2/...
MoS$_2$/WS$_2$ interface and excitonic solar cell. (a) DFT bandstructure of the MoS$_2$/WS$_2$ bilayer. Shown in red below the plot is the charge density associated with the wave functions for the Γ-points and bands marked as 1–3. At the K point, the VBM stems from states in WS$_2$ and the CBM from states in MoS$_2$, while the VBM at Γ shows contributions from both layers. The atoms are shown as yellow (S), blue (Mo), and green (W) spheres. (b) PDOS of the MoS$_2$/WS$_2$ bilayer, showing type-II alignment consistent with the charge density plots shown in a. The energy is referenced to the Fermi energy, and the arrows indicate PDOS features in the VBM due, respectively, to electronic states at the K and Γ points of the Brillouin zone. (c) Absorbance spectra of the MoS$_2$/WS$_2$ bilayer and of the individual composing layers, computed using BSE.

Despite the relatively low efficiency compared to thicker active layers, the power generated by a unit volume or mass of active layer material (power density) in a 1% efficient MoS$_2$/graphene solar cell would be incredibly high. Table 2 compares the power density of a MoS$_2$/graphene solar cell to the power densities of ultrathin solar cells based on GaAs and Si with current records of thickness and efficiency. We estimate that a MoS$_2$/graphene bilayer with a thickness of 0.9 nm, a weight of 3.9 mg/m$^2$, and an efficiency of 0.1%–1.0% as derived above would achieve a power density of 1.0–10.0 MW/L or 0.25–2.5 MW/kg. Such values are higher by approximately 1–3 orders of magnitude compared to existing record solar cells, and higher than any known energy generation and conversion device.$^{39,40}$ We remark that, although power density is not a conventional figure of merit in PV, it is an important metric to understand the ultimate power generation limits in solar cells achieving the smallest possible thickness, as well as to estimate the energy achievable from a unit volume or weight of active layer material.$^{42}$ For example, Table 2 indicates that a 1% efficient solar cell based on MoS$_2$/graphene is 30 times less efficient than the best 1 μm thick device based on GaAs, but using a thickness 1000 times smaller; this corresponds to a generated power per unit volume (or equivalently, per unit thickness) higher by a factor of ~30 for MoS$_2$/graphene compared to GaAs.

We highlight the fact that the PCE (and thus power density) values estimated here for a MoS$_2$/graphene solar cell are well-grounded: the absorbed photon flux $J_{ab}$ is estimated from accurate calculations of the absorbance closely matching experimental results (Figure 1b), the chosen $V_{OC}$ values are moderate (0.1–0.5 V) and within the range estimated above using DFT, and a realistic range of FF values$^{37}$ of 0.3–0.6 as well as a moderate IQE value of 0.7 were assumed in our calculations. Rather conservative values have been chosen for all the quantities composing the PCE, and the high power densities obtained are the sole consequence of the high absorbance for very small thickness in graphene and TMD monolayers.

We next estimate the feasibility and performance of another possibility for 1-nm-thick PV, this time constituted by an interface between the two semiconducting monolayers MoS$_2$ and WS$_2$, which would need to form a type-II heterojunction to enable exciton dissociation and charge separation.$^{17}$ Different from the MoS$_2$/graphene interface, the interaction between two TMD monolayers leads to significant changes in the bandstructure and absorption spectrum compared to the isolated monolayers. In particular, the DFT bandstructure of the MoS$_2$/WS$_2$ interface shows the formation of an indirect gap due to the interaction of antibonding $p_z$ orbitals from S atoms in the two TMD monolayers, resulting in an increase of the VBM at Γ (Figure 3a). The VBM at K shows contributions only from in-plane d orbitals of WS$_2$, while the CBM at K is contributed only by d$_{xz}$ orbitals of MoS$_2$, which would need to form a type-II heterojunction to enable the formation of a charge-transfer exciton shared by the two layers. Consistent with this picture, a type-II alignment favorable for solar cell operation is achieved at the MoS$_2$/WS$_2$ interface, where WS$_2$ behaves as the donor and MoS$_2$ as the acceptor, as shown by the analysis of the PDOS in Figure 3b.$^{43}$ Although the VBM states near the Γ point are an exception as they stem...
from both monolayers and may act as recombination centers, we expect the key photoexcitation at visible photon energies to involve states at the K point and thus conclude that the observed band alignment would lead to effective PV operation. Owing to the interlayer interaction leading to the formation of an indirect gap, the optical absorption spectrum of the bilayer obtained from BSE shows significant differences from the sum of the spectra of the two composing layers (Figure 3c). In particular, the bilayer absorbance is lower than the sum of the absorbances of the individual layers, and the absorption spectrum shows the formation of a lower absorption edge compared to the composing layers, thus confirming the presence of charge-transfer excitons. Using eq 1, we estimate a maximum short circuit current $I_{sc} \approx 3.5$ mA/cm$^2$ for the MoS$_2$/WS$_2$ bilayer (close to the value for isolated MoS$_2$), and we extract from the PDOS a maximum open circuit voltage of $\sim 0.4$ V, equal to the band gap of the type-II interface (see Figure 3b). Using an IQE value of 0.7, FF values in the 0.3–0.6 range as above, and a $V_{oc}$ of 0.5–1 V, we estimate PCE values of 0.4−1.5% for a bilayer of MoS$_2$/WS$_2$ with a 1.2 nm thickness and a weight of only 7.9 mg/cm$^2$, resulting in ultrahigh power densities similar to the MoS$_2$/graphene case (see Table 2).

The PV efficiencies computed here could be increased using a number of strategies in a real device. For example, an increase by a factor of 2 in the efficiency compared to what is estimated here can be gained using a double pass of light in the active layer, as can be obtained by employing a back metallic contact in the absence of light interference effects. In addition, since the absorbance of graphene and MoS$_2$ has been measured to in the 1.5−8999. Owing to the interlayer interaction leading to the formation of a lower absorption edge compared to the composing layers, thus confirming the presence of charge-transfer excitons. Using eq 1, we estimate a maximum short circuit current $I_{sc} \approx 3.5$ mA/cm$^2$ for the MoS$_2$/WS$_2$ bilayer (close to the value for isolated MoS$_2$), and we extract from the PDOS a maximum open circuit voltage of $\sim 0.4$ V, equal to the band gap of the type-II interface (see Figure 3b). Using an IQE value of 0.7, FF values in the 0.3–0.6 range as above, and a $V_{oc}$ of 0.5–1 V, we estimate PCE values of 0.4−1.5% for a bilayer of MoS$_2$/WS$_2$ with a 1.2 nm thickness and a weight of only 7.9 mg/cm$^2$, resulting in ultrahigh power densities similar to the MoS$_2$/graphene case (see Table 2). The PV efficiencies computed here could be increased using a number of strategies in a real device. For example, an increase by a factor of 2 in the efficiency compared to what is estimated here can be gained using a double pass of light in the active layer, as can be obtained by employing a back metallic contact in the absence of light interference effects. In addition, since the absorbance of graphene and MoS$_2$ has been measured to in the 1.5−8999. Owing to the interlayer interaction leading to the formation of a lower absorption edge compared to the composing layers, thus confirming the presence of charge-transfer excitons. Using eq 1, we estimate a maximum short circuit current $I_{sc} \approx 3.5$ mA/cm$^2$ for the MoS$_2$/WS$_2$ bilayer (close to the value for isolated MoS$_2$), and we extract from the PDOS a maximum open circuit voltage of $\sim 0.4$ V, equal to the band gap of the type-II interface (see Figure 3b). Using an IQE value of 0.7, FF values in the 0.3–0.6 range as above, and a $V_{oc}$ of 0.5–1 V, we estimate PCE values of 0.4−1.5% for a bilayer of MoS$_2$/WS$_2$ with a 1.2 nm thickness and a weight of only 7.9 mg/cm$^2$, resulting in ultrahigh power densities similar to the MoS$_2$/graphene case (see Table 2). The PV efficiencies computed here could be increased using a number of strategies in a real device. For example, an increase by a factor of 2 in the efficiency compared to what is estimated here can be gained using a double pass of light in the active layer, as can be obtained by employing a back metallic contact in the absence of light interference effects. In addition, since the absorbance of graphene and MoS$_2$ has been measured to.

**ASSOCIATED CONTENT**

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<td>Methods: Details of the DFT, GW, and BSE calculations. Figure S1: Comparison of independent-particle and BSE absorption spectra of monolayer MoS$_2$, Figure S2: Absorption coefficient of bulk MoS$_2$. This material is available free of charge via the Internet at <a href="http://pubs.acs.org">http://pubs.acs.org</a>.</td>
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(31) Taft, E. A.; Philipp, H. R. Phys. Rev. 1965, 138, A197 The absorption coefficient $\alpha$ of graphite was obtained from the optical conductivity $\sigma$ in Figure 6 using $\alpha = (4\pi/(c\cdot n))\sigma$, where $n \approx 2.2$ is the refractive index of graphite.


(34) We remark that this result is prone to variations in a real device depending on Fermi-level pinning at the MoS2/metal interface and on device processing conditions, so that a quantitatively accurate limit of the open-circuit voltage can only be predicted when all such variables are defined.


(43) We note that, although the band alignment is inferred here from a DFT calculation, the type-II alignment is retained both at the GW and BSE levels of theory and the BSE optical gaps are numerically close to the DFT gaps due to a compensation of errors at the DFT level. This situation is also found in other monolayer systems. Hence, correcting the DFT gaps to the GW or BSE gaps would not change the type-II alignment for the MoS2/WS2 interface.
