Gate-tunable carbon nanotube–MoS_2 heterojunction p-n diode

Deep Jariwala[a], Vinod K. Sangwan[a], Chung-Chiang Wu[a], Pradyumna L. Prabhumirashi[a], Michael L. Geier[a], Tobin J. Marks[a,b,1], Lincoln J. Lauhon[a], and Mark C. Hersam[a,b,c,1]

Departments of [a]Materials Science and Engineering, [b]Chemistry, and [c]Medicine, Northwestern University, Evanston, IL 60208

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The p-n junction diode and field-effect transistor are the two most ubiquitous building blocks of modern electronics and optoelectronics. In recent years, the emergence of reduced dimensionality materials has suggested that these components can be scaled down to atomic thicknesses. Although high-performance field-effect devices have been achieved from monolayered materials and their heterostructures, a p-n heterojunction diode derived from ultrathin materials is notably absent and constrains the fabrication of complex electronic and optoelectronic circuits. Here we demonstrate a gate-tunable p-n heterojunction diode using semiconducting single-walled carbon nanotubes (SWCNTs) and single-layer molybdenum disulfide as p-type and n-type semiconductors, respectively. The vertical stacking of these two direct band gap semiconductors forms a heterojunction with electrical characteristics that can be tuned with an applied gate bias to achieve a wide range of charge transport behavior ranging from insulating to rectifying with forward-to-reverse bias current ratios exceeding 10^4. This heterojunction diode also responds strongly to optical irradiation with an external quantum efficiency of 25% and fast photoresponse <15 μs. Because SWCNTs have a diverse range of electrical properties as a function of chirality and an increasing number of atomically thin 2D nanomaterials are being isolated, the gate-tunable p-n heterojunction concept presented here should be widely generalizable to realize diverse ultrathin, high-performance electronics and optoelectronics.

2D transition metal dichalcogenide | single layer MoS_2 | van der Waals heterostructure | rectifier | photodetector

When two semiconductors with an opposite carrier type contact one another, charge transfer occurs across their interface and creates a potential difference determined by the doping profile. In bulk semiconductor p-n junctions, the doping level is primarily controlled via diffusion or implantation of substitutional impurities, which implies minimal control over the doping profile following device fabrication. In contrast, atomically thin semiconductors can be electrostatically doped by applying a bias to a capacitively coupled gate electrode (1–3). The atomically thin structure of these materials also enables doping modulation of the overlying layers in a vertically stacked heterostructure (4). For example, this strategy allows gapless graphene to be used in field-effect tunneling devices in combination with other layered materials (4, 5). Vertical 2D heterostructures have also been used to create high-performance Metal Oxide Semiconductor Field Effect Transistors (MOSFETs) (6), tunneling field-effect transistors (FETs) (4), barristors (7), inverters (8), and memory devices (9, 10), in addition to facilitating the study of novel physical phenomena in layered materials (11–14). Similarly, in-plane graphene heterostructures and controlled doping have served as the basis for unique 2D devices (15–18). Although the nearly perfect 2D structure and low density of states in graphene provide advantages in some heterostructure devices, its gapless nature prevents the formation of a large potential barrier for charge separation and current rectification despite efforts to create in-plane p-n homojunctions by split gating (19). In particular, the lack of distinct monolayer semiconductors with complementary (p and n) polarities has precluded the realization of a gate-tunable heterojunction p-n diode.

In this report, we demonstrate the fabrication and operation of a gate-tunable p-n heterojunction diode using semiconducting single-walled carbon nanotubes (s-SWCNTs) (p-type) (20) and single layer (SL)-MoS_2 (n-type) (21, 22). Fig. 1*A* shows a false-colored SEM of a representative device. A lower-magnification optical micrograph of the same device in Fig. 1*B* shows the n-type (SL-MoS_2) FET, p-n s-SWCNT/SL-MoS_2 heterojunction, and p-type (s-SWCNT) FET from top to bottom. The device fabrication begins by depositing e-beam lithographically defined gold electrodes on SL-MoS_2 flakes that are exfoliated on 300-nm SiO_2/Si wafers (Fig. 1 *C, I*). The SL-MoS_2 FET is electrically isolated by patterning 30-nm alumina via atomic layer deposition (ALD) followed by transfer and patterning of a sorted s-SWCNT thin film (23, 24) (Fig. 1 *C, II*) to yield the final device (Fig. 1 *C, III*). *SI Text* provides further details on the device fabrication.

The ultrathin nature of the heterojunction allows gate tunability of the diode electrical characteristics. Fig. 2*A* shows the output plots of a representative device under different gate biases. The device transitions from a nearly insulating behavior at $V_G = 70$ V to a poorly rectifying state ($r^2 < 60\%$ for fits to the Shockley diode equation) at $V_G = 40$ V to a highly rectifying diode for $V_G \leq 0$ V ($r^2 > 99\%$). For the heterojunction diodes, V_D refers to the bias on the s-SWCNT electrode such that $V_D > 0$ corresponds to forward bias while the electrode in contact with SL-MoS_2 is grounded. The transfer plot further demonstrates the gate tunability of the current through the p-n heterojunction (Fig. 2*B*). The transfer characteristics of the junction (green)

Significance

The p-n junction diode is the most ubiquitous and fundamental building block of modern electronics, with far-reaching applications including integrated circuits, detectors, photovoltaics, and lasers. With the recent discovery and study of atomically thin materials, opportunities exist for adding new functionality to the p-n junction diode. Here we demonstrate that a p-n heterojunction diode based on atomically thin MoS_2 and sorted semiconducting carbon nanotubes yields unprecedented gate tunability in both its electrical and optical properties, which is not observed in the case of bulk semiconductor devices. In addition to enabling advanced electronic and optoelectronic technologies, this p-n heterojunction diode provides new insight into charge transport and separation at atomically thin heterointerfaces.

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[1]To whom correspondence may be addressed. E-mail: t-marks@northwestern.edu or m-hersam@northwestern.edu.

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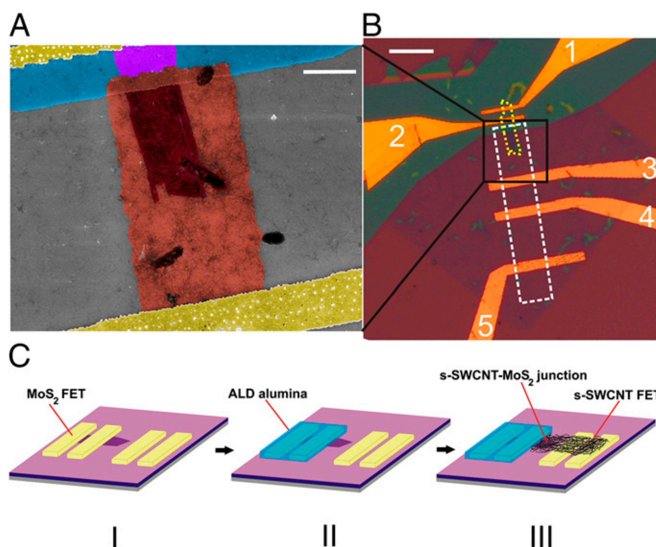

Fig. 1. Microscopy and fabrication of the s-SWCNTs/SL-MoS$_2$ p-n heterojunction diode. (*A*) False-colored SEM image of the heterojunction diode. (Scale bar, 2.5 μm.) The yellow regions at the top and bottom are the gold electrodes. The patterned alumina (blue region) serves as a mask for insulating a portion of the SL-MoS$_2$ flake (violet region). The pink region is the patterned random network of s-SWCNTs (p-type) in direct contact with the exposed part of the SL-MoS$_2$ flake (n-type) to form the p-n heterojunction diode (dark red). (*B*) Optical micrograph showing the device layout at a lower magnification. The dashed yellow boundary indicates the SL-MoS$_2$ flake, whereas the dashed white rectangle denotes the patterned s-SWCNT film. Electrodes 1 and 2 form the n-type (SL-MoS$_2$) FET, which is insulated by the patterned alumina film (cyan). Electrodes 2–3 form the p-n heterojunction, whereas 3–4 and 4–5 form p-type s-SWCNT FETs. (Scale bar, 10 μm.) (*C*) Schematic of the fabrication process: (*I*) SL-MoS$_2$ FET and an extra pair of electrodes are fabricated via e- beam lithography on 300 nm SiO$_2$/Si. The Si substrate acts as the global back gate. (*II*) The MoS$_2$ FET is insulated by patterning an alumina film in a liftoff process, followed by (*III*) transfer and patterning of the s-SWCNT network to yield the final device configuration consisting of a top contact SL-MoS$_2$ FET, bottom contact s-SWCNT FET, and p-n heterojunction.

show an unusual gate voltage dependence, which we refer to as "antiambipolar" behavior. In particular, the maximum conductance occurs near $V_G = 0$, which is the opposite of conventional ambipolar behavior that shows a minimum conductance near $V_G = 0$. The current on/off ratio exceeds 10^4 in the transfer plots, which is suitable for advanced logic applications (*SI Text*). The transfer characteristics of the heterojunction can be qualitatively viewed as a superposition of the p-type s-SWCNT (red) and n-type SL-MoS$_2$ (blue) FET transfer plots. However, the net current through the heterojunction is smaller than the SL-MoS$_2$ and s-SWCNT FET transfer characteristics due to the additional resistance at the junction.

This unique transfer characteristic can be viewed as originating from an FET channel consisting of two p and n semiconductors in series. The change in the resistance of each component with gate bias affects the net series resistance leading to the resulting antiambipolar transfer characteristic of the junction. Thus, the junction transfer curve has two off states, representing the depleted state of SL-MoS$_2$ and s-SWCNTs. Furthermore, the contact resistances of the s-SWCNT film and SL-MoS$_2$ with Au are relatively small compared with the sheet/channel resistances (25, 26) and thus have negligible effect on the junction characteristics (see *SI Text* for output plots of s-SWCNT and SL-MoS$_2$ FETs). It is worth noting that the maximum of the junction transfer curve does not exactly coincide with the intersection of the SL-MoS$_2$ and s-SWCNT transfer curves. This offset can be attributed to hysteresis in the transfer plots and the fact that the SL-MoS$_2$ FET is encapsulated in alumina, which is known to shift threshold voltage in SL-MoS$_2$ FETs (21).

An important parameter in characterizing diode characteristics is the ratio of the forward current, $|I_f|$, to the reverse current, $|I_r|$, at the same bias magnitude. Fig. 2*C* shows that $|I_f|/|I_r|$ varies by over five orders of magnitude as a function of gate voltage. Between the two gate bias extremes, the s-SWCNT/SL-MoS$_2$ heterojunction transitions from an n-n$^+$ junction at V_G = 80 V to a p-n junction at $V_G = -80$ V. The large band gap of SL-MoS$_2$ (>1.8 eV) (27) compared with that of the s-SWCNTs (∼0.7 eV) allows electrostatic depletion of SL-MoS$_2$ to a lightly n doped (n$^-$) or nearly intrinsic insulating state, thereby leading to $|I_f|/|I_r|$ values exceeding 10^4 at $V_G = -80$ V. On the other hand, the small band gap of the s-SWCNTs allows electrostatic inversion from p-doping to n-doping at large positive V_G, resulting in poor $|I_f|/|I_r|$ values for $V_G > 60$ V. Gate tunable p-n homojunction diodes have been previously fabricated by split gating of individual SWCNTs. However, such homojunctions only allow control over the built-in voltage via differential biases in split gates (28, 29). The present gate tunable p-n heterojunction, on the other hand, is fundamentally different as it has a built-in potential at zero gate bias as evident from the rectifying current (I)-voltage (V) (*I-V*) characteristics (Fig. 2*A*). Furthermore, in this heterojunction, the gate is used to simultaneously tune the doping concentrations of both semiconductors, thereby allowing tunability in the built-in voltage and rectification ratios.

To further understand the gate-dependent modulation of the heterojunction *I-V* curves, we fit them to the Shockley diode equation. The best fit to the Shockley diode equation is observed for $V_G = -40$ V. For other V_G values, either the diode ideality factor (n) is >2 or the fits are poor ($r^2 < 60\%$) (*SI Text*). The disorder at the interface of random network SWCNT films [see atomic force microscopy (AFM) images in *SI Text*] and SL-MoS$_2$ possibly leads to more recombination, resulting in larger ideality factors compared with the nearly ideal diode behavior in WSe$_2$/InAs heterojunctions that have a uniform 2D interface (30). The gate dependence of the present diode behavior enables gate-tunable rectifier circuits (Fig. 2*D*), which is a unique feature of the s-SWCNT/SL-MoS$_2$ heterojunction device that has not been observed in conventional bulk semiconductor diodes (see *SI Text* for further details on measurement techniques).

Both SL-MoS$_2$ and s-SWCNTs have direct band gaps (31, 32) and exhibit signatures of bound excitonic states in their absorption spectra (2, 31). Therefore, photocurrent generation is expected on optical irradiation of p-n heterojunctions based on these materials. To that end, scanning photocurrent microscopy was used to spatially map the local photoresponse of the s-SWCNT/SL-MoS$_2$ heterojunction device (Fig. 3*A*). The regions of large negative photocurrent lie in the heterojunction area outlined by the SL-MoS$_2$ flake (purple) overlapping with the patterned s-SWCNT film (red). No measurable photocurrent is observed from the nonoverlapping regions of either the s-SWCNT film, SL-MoS$_2$, or the electrical contacts (see *SI Text* for additional details), indicating that the photocurrent measured under uniform illumination is generated by the vertical heterojunction. Photocurrent from Au contacts to s-SWCNT films was not observed in our recent study (33). For the case of two-terminal SL-MoS$_2$ devices, photocurrent has been observed at the contacts due to band bending (34). However, the band bending at the contacts is opposite to that of the junction, which suppresses the local near-contact photocurrent because charge neutrality cannot be maintained at the injection level used. The spectral dependence of the photocurrent (Fig. 3*B*) corresponds to the absorption peaks of SL-MoS$_2$ (27) and S_{22} (24) peaks of s-SWCNTs, which demonstrates that this unique heterointerface can induce carrier separation following exciton and/or free carrier generation in either material. The photocurrent generated in the visible portion of the

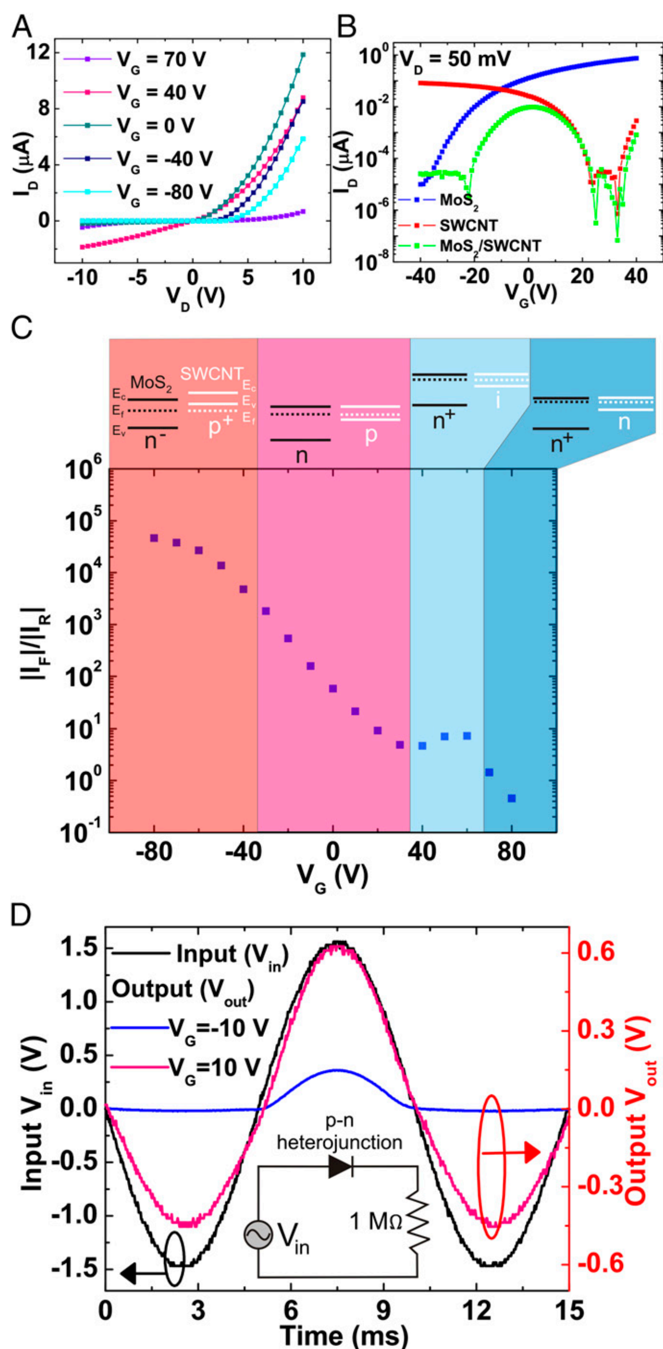

Fig. 2. Electrical properties of the s-SWCNT/SL-MoS_2 p-n heterojunction diode. (*A*) Gate-tunable output characteristics showing the transition from a nearly insulating state at V_G = 70 V to a conductive state with relatively poor rectification at V_G = 40 V to a highly rectifying diode behavior at negative gate voltages. (*B*) Transfer characteristics of the p-n junction (green), showing an antiambipolar characteristic, which is qualitatively a superposition of the transfer characteristics of the p-type s-SWCNT FET and n-type SL-MoS_2 FET. (*C*) Forward-to-reverse current ratio (at a heterojunction bias magnitude of 10 V) as a function of gate bias. The labels at the top show the corresponding band diagrams for the s-SWCNT/SL-MoS_2 p-n heterojunction. At a high positive gate bias, the formation of an n^+-n junction implies a low rectification ratio that transitions into an n^+-i junction (plateau region in the plot) with reducing V_G. The rectification ratio then rises with decreasing gate bias due to the formation of a p-n junction. (*D*) Demonstration of gate-tunable rectification using the p-n heterojunction diode. The *y* axis on the left shows the input voltage, whereas the *y* axis on the right shows the output voltage across the series resistor (1 MΩ). As a function of the gate bias, the device evolves from a nonrectifying resistor-like state at V_G = 10 V (magenta) to a diode-like rectifying state at V_G = -10 V (blue).

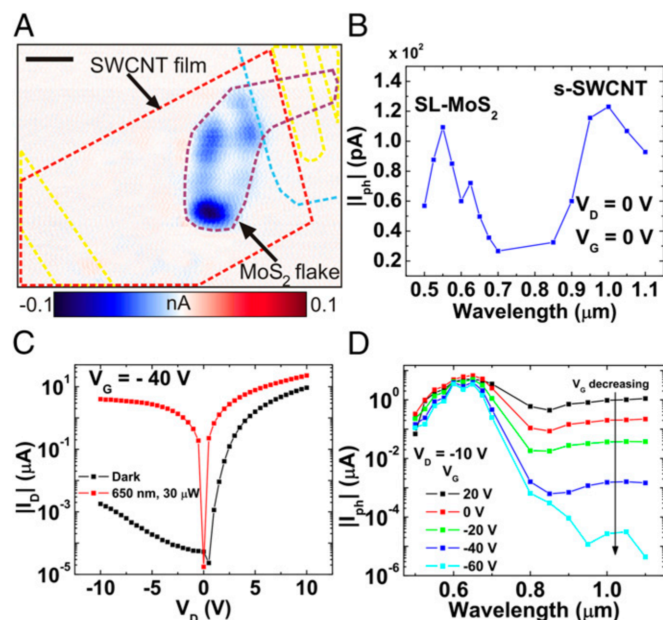

Fig. 3. Photoresponse of the p-n heterojunction. (*A*) Scanning photocurrent micrograph of a representative heterojunction device acquired at V_D (s-SWCNT electrode), $V_G = 0$ V showing the outlines of the SL-MoS_2 flake (purple dashed line) and the patterned s-SWCNT film (red dashed line) acquired at 700 nm with 20-μW power. Regions of large negative photocurrent (blue) are observed in the overlapping junction region. The patterned alumina and electrodes are indicated by cyan and yellow dashed lines, respectively. (*B*) Photocurrent spectrum of the junction under global illumination and zero bias conditions. The photocurrent magnitude is highest at the characteristic absorption energies of both SL-MoS_2 and s-SWCNTs. The photocurrent spectrum is acquired at the same incident power (30 μW). (*C*) Output curve of the same device in the dark and under global illumination at 650 nm. (*D*) Photocurrent spectral response can be tuned with the gate voltage. With decreasing gate voltage, the increased p-doping of the nanotubes and concomitant decreased n-doping of MoS_2 leads to a lower photocurrent in the near infrared region.

spectrum also likely has contributions from both SL-MoS_2 and the S_{33} transitions of s-SWCNTs because both absorb in that range of wavelengths (see *SI Text* for absorbance spectrum of SWCNTs).

To further illustrate the photoresponse of the s-SWCNT/SL-MoS_2 heterojunction, both output (*I-V*) and transfer curves (*I-V_g*) were acquired under global illumination at a series of wavelengths. A representative comparison of the dark and illuminated *I-V* curves at $V_G = -40$ V reveals that the photocurrent increases by four orders of magnitude at a heterojunction reverse bias of −5 V (Fig. 3*C*). Fig. 3*D* also shows the gate voltage-dependent photocurrent values at a heterojunction reverse bias of −10 V (Fig. 3*D*). As the gate voltage becomes more negative, the relative contribution from the s-SWCNT portion of the spectrum decreases, which is consistent with the s-SWCNT/SL-MoS_2 heterojunction becoming a p^+-n^- junction. Because the depletion region in a p-n junction extends farther into the side with lower doping/majority carrier concentration, the junction almost entirely lies in the SL-MoS_2 as it is depleted at negative V_G, leading to a reduced photocurrent contribution from the s-SWCNTs as observed in Fig. 3*D*. If we consider the junction from a molecular perspective used to describe organic semiconductor heterojunctions, one might also expect a change in the rate of charge transfer due to changes in band offsets [or Highest Occupied Molecular Orbitals (HOMO) and Lowest Unoccupied Molecular Orbitals (LUMO) levels] across the heterojunction.

With a strong photoresponse, the s-SWCNT/SL-MoS_2 heterojunction can be exploited as a photodetector. Diode-based

Fig. 4. Photodetection using the p-n heterojunction diode. (*A* and *B*) Time-dependent photoresponse of the p-n heterojunction showing fast rise and decay times of ∼15 μs. (*C*) EQE as a function of reverse bias for the heterojunction at 650 nm. EQE increases linearly with reverse bias from 0 to −5 V with the highest EQE of 25% occurring at −10 V. (*D*) Spectrally dependent responsivity (*R*) of the photodiode in linear (blue) and logarithmic (red) scales. A large responsivity is observed for the absorption wavelengths of SL-MoS_2 compared with s-SWCNTs because the diode is being operated at $V_G = -40$ V (depletion mode of SL-MoS_2).

photodetectors are known for their fast photoresponse times compared with phototransistors because the photoexcited carriers must only traverse a distance equal to the depletion width of the junction. Indeed, we observe a fast photoresponse (<15 μs) from the s-SWCNT/SL-MoS_2 heterojunction as seen in Fig. 4 *A* and *B*. Importantly, this photoresponse time is orders of magnitude smaller than recently reported heterojunction phototransistors based on graphene (35). Furthermore, the s-SWCNT/SL-MoS_2 heterojunction photoresponse time reported here is limited by the rise time of the preamplifier used in these experiments, and thus 15 μs represents an upper bound.

The external quantum efficiency (EQE) of the s-SWCNT/SL-MoS_2 heterojunction photodetector is also noteworthy (∼25% at $V_D = -10$ V) and compares favorably to other recently reported 2D nanomaterial heterostructures (Fig. 4*C*) (35–37). The EQE is calculated as $EQE = \frac{I_{ph}/e}{P/h\nu} \times 100$, where I_{ph}, e, P, and $h\nu$ represent the photocurrent, electronic charge, incident optical power, and photon energy, respectively. The linear rise in EQE at low reverse biases (Fig. 4*C*) is similar to the behavior of quantum dot (QD)-based photodiodes (38), which is related to changes in the depletion region. Here, given the negligible thickness of SL-MoS_2 compared with the expected depletion width (39), we attribute the increasing EQE to an increase in the rate of charge transfer across the heterojunction interface due to the change in band alignment rather than to an increase in the depletion width. The spectral responsivity (R) of the heterojunction photodetector is presented in Fig. 4*D*. The highest R exceeds 0.1 A/W at a wavelength of 650 nm, which is comparable to other nanostructured diode-based photodetectors in the literature (40). The combination of high R and fast photoresponse time presents distinct advantages over currently available organic and QD photodiodes (38).

In conclusion, we demonstrated a gate-tunable p-n heterojunction diode through the integration of p-type s-SWCNTs and n-type SL-MoS_2. The ultrathin nature of the constituent materials implies that both components can be modulated by a capacitively coupled gate bias, thereby enabling wide tunability of charge transport from a nearly insulating state to a highly rectifying condition with forward-to-reverse bias current ratios exceeding 10^4. When operated as a three-terminal device, the p-n heterojunction diode also shows antiambipolar behavior with current on/off ratios greater than 10^4, suggesting its utility in advanced logic applications. Furthermore, because s-SWCNTs and SL-MoS_2 are direct band gap semiconductors, the p-n heterojunction diode serves as an effective photodetector with fast photoresponse <15 μs. By combining other chirality resolved s-SWCNTs (41) with the growing list of 2D semiconductor nanomaterials (42–45), the p-n heterojunction diode can be generalized to a wide range of electronic and optoelectronic applications.

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