Gate-tunable contact-induced Fermi-level shift in semimetal

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Low-dimensional semimetal–semiconductor (Sm-S) van der Waals (vdW) heterostructures have shown their potentials in nanoelectronics and nano-optoelectronics recently. It is an important scientific issue to study the interfacial charge transfer as well as the corresponding Fermi-level shift in Sm-S systems. Here we investigated the gate-tunable contact-induced Fermi-level shift (CIFS) behavior in a semimetal single-walled carbon nanotube (SWCNT) that formed a heterojunction with a transition-metal dichalcogenide (TMD) flake. A resistivity comparison methodology and a Fermi-level catch-up model have been developed to measure and analyze the CIFS, whose value is determined by the resistivity difference between the naked SWCNT segment and the segment in contact with the TMD. Moreover, the relative Fermi-level positions of SWCNT and two-dimensional (2D) semiconductors can be efficiently reflected by the gate-tunable resistivity difference. The work function change of the semimetal, as a result of CIFS, will naturally introduce a modified form of the Schottky–Mott rule, so that a modified Schottky barrier height can be obtained for the Sm-S junction. The methodology and physical model should be useful for low-dimensional reconfigurable nanodevices based on Sm-S building blocks.

Significance

Resistivity comparison methodology was developed to measure and analyze the contact-induced Fermi-level shift (CIFS) as well as the interfacial charge transfer in low-dimensional semimetal–semiconductor (Sm-S) systems. The Fermi-level catch-up model was further built to depict the gate-tunable CIFS in such Sm-S systems. The Schottky barrier height for the Sm-S junction can be modified by introducing the CIFS term. The progress in this work will have an important role in promoting the research of Sm-S junctions, which are essential building blocks for future low-dimensional nanodevices.

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After junction formation, the contact-induced Fermi-level shift in semimetals is sensitive to the gate potential (V_g), which can be attributed to their specific band structures and the quantum-capacitance-related Fermi-level shift. The transfer curves of metallic SWCNT and graphene show relative high resistivity when their Fermi levels are set around the Dirac point by V_g (27, 28). In addition to the gate modulation, interfacial charge transfer will also introduce a Fermi-level shift of such semimetals and a corresponding resistance change in the Sm-S systems. Fig. 1 schematically illustrates the band alignment of a semimetal SWCNT in contact with semiconductors, in which the CIFS δμ_Sm of the semimetal depends on the work function difference before contact (Δφ = φ_Sm − φ_S). When the Fermi level of SWCNT is lower than that of the semiconductor, Δφ > 0, electrons will transfer from semiconductor to semimetal, inducing N doping and a positive δμ_Sm for SWCNT, as shown in Fig. 1B. In the case of Δφ < 0, δμ_Sm < 0 and P doping is induced to SWCNT, as shown in Fig. 1C. In all these cases, the Fermi level of semimetal shifts away from its Dirac point, which brings a change of resistivity. Therefore, if the resistivity difference of semimetal with and without semiconductor contacting can be measured, behaviors of the interfacial charge transfer as well as the gate-tunable CIFS δμ_Sm of the semimetal could possibly be revealed efficiently. Therefore, the resistivity comparison methodology is suggested and developed to study these issues.

A 1D-2D mixed-dimensional system was constructed to investigate the CIFS and interfacial charge transfer in Sm-S heterostructures. Fig. 2A sketches the device structure and presents a scanning electron microscopy (SEM) image of such a device, where a stacked SWCNT-TMD heterostructure is wired by a series of well-designed electrodes. The fabrication procedures mainly include dry transfer techniques of CNT and TMD, and e-beam lithography (EBL). Briefly, an individual ultralong suspending SWCNT was identified, picked up, and then transferred to the Si/SiO_2 substrate by two tungsten tips. A dry transfer method via polydimethylsiloxanes (PDMS) stamp was then used to stack a mechanically exfoliated MoS_2 flake onto the CNT. The Ti/Au (5/50 nm) electrodes were fabricated by using EBL, electron beam evaporation, and lift-off procedures. Raman spectroscopy (Fig. 2B) gives the identification for the metallic SWCNT (29, 30), MoS_2 and WSe_2 (31). The prominent radial breathing mode (RBM) peak verifies that the CNT is single walled, and the absence of the D peak around 1,350 cm⁻¹ indicates the high quality of the CNT structure.

The as-fabricated device provides an effective solution for the comparative studies on CNT with and without MoS_2 contacting. The CNT is divided into segments by the Ti/Au electrodes. The five electrodes from B to F define four CNT segments with different lengths. All the transport measurements were conducted under vacuum at room temperature. The quasi-1D electronic

Results

Resistivity Comparison Methodology. It is known that the transport behaviors of low-dimensional semimetals are sensitive to the gate potential (V_g), which can be attributed to their specific band structures and the quantum-capacitance-related Fermi-level shift. The transfer curves of metallic SWCNT and graphene show relative high resistivity when their Fermi levels are set around the Dirac point by V_g (27, 28). In addition to the gate modulation, interfacial charge transfer will also introduce a Fermi-level shift of such semimetals and a corresponding resistance change in the Sm-S systems. Fig. 1 schematically illustrates the band alignment of a semimetal SWCNT in contact with semiconductors, in which the CIFS δμ_Sm of the semimetal depends on the work function difference before contact (Δφ = φ_Sm − φ_S). When the Fermi level of SWCNT is lower than that of the semiconductor, Δφ > 0, electrons will transfer from semiconductor to semimetal, inducing N doping and a positive δμ_Sm for SWCNT, as shown in Fig. 1B. In the case of Δφ < 0, δμ_Sm < 0 and P doping is induced to SWCNT, as shown in Fig. 1C. In all these cases, the Fermi level of semimetal shifts away from its Dirac point, which brings a change of resistivity. Therefore, if the resistivity difference of semimetal with and without semiconductor contacting can be measured, behaviors of the interfacial charge transfer as well as the gate-tunable CIFS δμ_Sm of the semimetal could possibly be revealed efficiently. Therefore, the resistivity comparison methodology is suggested and developed to study these issues.
structures of metallic CNTs give small and constant DOS between the two von Hove singularities, as sketched in Fig. 2C (32, 33). And there is an additional narrow gap about several tens of millielectronvolts (∼k_BT) at the Dirac point due to curvature and strain (34). Such specific band structures make a small on/off ratio that could always be observed in transfer characteristics of metallic SWCNT devices, as shown in Fig. 2D. The length-dependent resistance derived from Fig. 2D was further plotted in Fig. 2 E, Inset, and the contact resistance (SI Appendix, Fig. S3), the channel resistivity (Fig. 3A) as well as the mean-free path of electrons (Fig. 2E) can be extracted from the intercepts and slopes according to the transfer length method (35). More information about the transfer length method can be found in Materials and Methods. The CNT segment covered by MoS_2 was wired by two electrodes A and B at the two naked parts. As shown in Fig. 2C, the resistance between electrodes A and B is equivalent to the sum of the contact resistance between SWCNT and Ti/Au, the resistance of intrinsic naked SWCNT, and the resistance of SWCNT in contact with MoS_2. Note that the resistance induced by the scattering at the junctions between covered CNT and naked CNT was neglected due to the linear dispersion relationship of the metallic SWCNT and the mismatched momentum between its two conducting channels (36–38). The Dirac electrons can pass through the barrier through Klein tunneling without backscattering (Fig. 2C) (39–41), which could be verified by the micrometer-long mean-free path of electrons (Fig. 2E) and the perfect linear output characteristics of the CNT segment defined by electrodes A and B (SI Appendix, Fig. S4 C and D). Therefore, the resistance as well as the resistivity (Fig. 3B) of the covered CNT can be calculated by subtracting the other items as shown in Fig. 2C from the resistance between A and B. The resistivity of the CNT with and without MoS_2 contacting is comparatively plotted in Fig. 3C.

Some critical information of the CIFS can be obtained from the V_g-dependent contact-induced resistivity difference of the
semimetal in such a 1D-2D Sm-S system (Fig. 3 A–C). First, the shape of the two resistivity plots shows obvious differences. The \( \rho_{CNT}(V_g) \) plot of the intrinsic SWCNT (blue curve in Fig. 3 A and C) without semiconductor contacting shows a maximum value at the Dirac point, while there is a maximum value and an additional minimal value in the comparative plot for the SWCNT in contact with MoS\(_2\) (green curve in Fig. 3 B and C). The significant change of the resistivity characteristics indicates a complicated behavior about the \( V_g \)-dependent CIFS in the semimetal SWCNT. Second, the two comparative plots have the same maximum resistivity at the Dirac points. Since the CIFS changes the Fermi-level position of the semimetal SWCNT rather than the band structure, the maximum value of the resistivity of the two SWCNT parts should be the same and appear at the position of their Dirac points. This could be used to verify the correctness of our resistivity measurement method. Third, the contact-induced resistivity difference \( \delta \rho \) is also plotted in Fig. 3C (red curve). The value of \( \delta \rho \) can be either positive or negative depending on the gate potential, which indicates that the CIFS depends on the gate potential and the two original Fermi levels in the Sm-S system may experience a complicated catch-up procedure under the gate modulation, which is discussed next.

**Fermi-Level Catch-Up Model.** From the respective band structures of the semimetal and semiconductor components, the gate-dependent CIFS \( \delta \mu_{Sm} \) can be theoretically predictable for a
Sm-S junction. Before junction formation, the Fermi-level positions ($\mu$) and the work functions ($\phi = 0 - \mu$) of the two materials can be independently controlled by $V_g$ through the electrostatic capacitance effect following $d\phi (e \cdot dV_g) = d\mu (e \cdot dV_g) = C_g (C_g + C_Q)$, where $C_g$ and $C_Q$ are the gate capacitance and the quantum capacitance, respectively (42). Since the metallic SWCNT has a constant DOS (constant $C_g$) and the carriers in an N-type MoS$_2$ follow the Boltzmann distribution at room temperature (1), the derivatives of $\phi$ are

$$\frac{d\phi_{\text{CNT}}}{dV_g} = -A$$  \hspace{1cm} [1]$$

$$\frac{d\phi_{\text{MoS}_2}}{dV_g} = -\frac{kT}{V_g + B}.$$  \hspace{1cm} [2]$$

The work function difference between the CNT and MoS$_2$, defined as $\Delta\phi = \phi_{\text{CNT}} - \phi_{\text{MoS}_2}$, can thus be derived as

$$\Delta\phi = C + kT \ln (V_g + B) - AV_g,$$  \hspace{1cm} [3]$$

where $A$, $B$, and $C$ are all constants determined by materials and device structures. The curve drawn by Eq. 3 opens down like a downward parabola. Fig. 3 D1, E1, and F1 shows three representative cases for $\Delta\phi$ and the doping states of the CNT after contact with MoS$_2$. Note that the orange and blue shaded areas represent the contact-induced P-type and N-type doping, respectively. Fig. 3 D1 shows the case of $\Delta\phi > 0$ for all $V_g$, which indicates that the Fermi level of the CNT is lower than that of MoS$_2$ and electrons are injected into the CNT from MoS$_2$ after contacting. The contact thus introduces an equivalent N-type doping to the CNT and induces a Fermi-level lifting ($\delta\mu_{\text{SM}} > 0$). Fig. 3 E1 presents the case of $\Delta\phi < 0$ for all $V_g$. The CNT feels equivalent P-type doping and $\delta\mu_{\text{SM}} < 0$, and the whole panel is marked in orange. Fig. 3 F1 shows that the equation $\Delta\phi = 0$ has two solutions $\alpha$ and $\beta$. In the orange area ($V_g < \alpha$ and $V_g > \beta$), $\Delta\phi < 0$ and $\delta\mu_{\text{SM}} < 0$, holes are injected into the CNT inducing equivalent P-type doping. In the blue area ($\alpha < V_g < \beta$, $\Delta\phi > 0$ and $\delta\mu_{\text{SM}} > 0$, it turns to N doping. At $\alpha$ and $\beta$, $\Delta\phi = 0$, $\delta\mu_{\text{SM}} = 0$, and there is no charge transfer. Therefore, the CIFS $\delta\mu_{\text{SM}}$ has a similar dependency on $V_g$ to that of $\Delta\phi$ in the above three cases, which can be further verified by Eq. 9 in Materials and Methods.

The contact-induced resistivity difference $\rho$ can also be deduced from the as-established $V_g$-dependent CIFS $\delta\mu_{\text{SM}}$. The $\rho - V_g$ curve of intrinsic CNT shown in Fig. 3A was used as a reference for theoretical deduction, with the orange and blue color blocks representing hole and electron conducting, respectively. Fig. 3 D2, E2, and F2–F4 lists five representative cases of $V_g$-dependent $\rho$ with (green curves) and without (blue curves) contacting as well as the corresponding $\rho$ (red curves). In these panels, the top two color blocks indicate the intrinsic CNT conducting type and the bottom color blocks present the contact-induced doping type. Fig. 3 D2 and E2 correspond to the cases of $\delta\mu_{\text{SM}} > 0$ and $\delta\mu_{\text{SM}} < 0$, respectively. There are three more cases for the $\pm$ switching of $\delta\mu_{\text{SM}}$, including the Dirac point located in the N-doping region (Fig. 3 F3), the left P-doping region (Fig. 3 E2), and the right P-doping region (Fig. 3 F4). These qualitative deductions about $\rho - V_g$ are based on three basic principles. First, the position of the Dirac point shifts to the left when the CNT is N doped to and to the right when it is P doped. Then the two transfer curves intersect at the point $X$, where the naked and MoS$_2$-covered CNTs have different conducting types but the same resistivity, revealing that their Fermi levels are symmetric with respect to the Dirac point. Second, when the conducting type and the contact-induced doping type of the CNT are the same, the resistivity decreases; otherwise, the resistivity increases. This can be clearly seen by comparing the top and bottom block colors. The same color corresponds to a negative $\delta\rho_{\text{CNT}}$, and the different color corresponds to a positive $\delta\rho_{\text{CNT}}$. Third, the green and blue $\rho_{\text{CNT}}$ curves intersect at points $\alpha$ and $\beta$, where $\delta\mu_{\text{SM}} = 0$ and there is no contact-induced doping to the CNT. For a better understanding, Movies S1–S5 demonstrate the five cases.

The correctness of the theoretical analysis can be verified with experimental results. It can be found that almost all features of Fig. 3 F3 are in accord with the experimental results shown in Fig. 3C, including the shape of the curves and the five feature points $X$, $\alpha$, $\beta$, DP, and DP'. The theoretical analysis about CIFS $\delta\mu_{\text{SM}}$ and contact-induced resistivity difference $\rho$ in the CNT is built on the $V_g$-dependent Fermi-level misalignment $\Delta\phi$ between the CNT and MoS$_2$. Their band structures’ difference shows different modulation efficiencies on Fermi-level shifting (Eqs. 1 and 2) and their Fermi levels thus experience a catch-up process during gate modulation. Hence, such a theory is vividly named the Fermi-level catch-up model. Fig. 3 C is further divided into six regions by the five feature points for more specific discussion. The corresponding band diagrams of the naked and MoS$_2$-covered CNTs are sketched in Fig. 3H. Fig. 3 H, i indicates that the Fermi level of the CNT is higher than that of MoS$_2$ ($\Delta\phi < 0$), the P-type CNT receives a hole injection, the P doping is enhanced, and the conductivity is improved. In Fig. 3 H, ii and iii, MoS$_2$’s Fermi level catches up with and surpasses the CNT at $\alpha$. Fig. 3 H, ii represents that the P-type SWCNT accepts electron injection, the P doping is weakened, and the conductivity is reduced. Then the conductive type switching occurs in Fig. 3 H, iii and iv. Entering Fig. 3 H, v, the N-type SWCNT accepts electron injection, making the N type enhanced and conductivity improved. From Fig. 3 H, v to vi, the CNT’s Fermi level overtakes MoS$_2$ at $\beta$. The N-type SWCNT receives hole injection again in Fig. 3 H, vi with weakened N doping and reduced conductivity. Therefore, when $V_g$ is scanned from $-30$ to $50$ V, the Fermi levels of the CNT and MoS$_2$ experience a process of overtaking and being overtaken, which leads to twice switching of the CIFS direction and can also be visually discriminated by the background colors of the band diagrams. Fig. 3C shows that the Dirac point of the CNT is located in the N-doping region (Fig. 3 F3) and the other cases could possibly be realized by changing the doping level of the CNT and MoS$_2$. For instance, the case presented in Fig. 3 F4 has been experimentally implemented by introducing extra N doping to the SWCNT/MoS$_2$ junction; the Fermi level of the CNT reaches its Dirac point at negative $V_g$, and all the features of the $\rho(V_g)$ curves shown in SI Appendix, Fig. S6 agree well with the prediction. More scenarios could be theoretically accessible via employing appropriate semimetals and semiconductors.

The Fermi-level catch-up process around point $\alpha$ was further verified by Kelvin probe force microscopy (KPFM). The surface potential images of the MoS$_2$ around the Sm-S junction and the corresponding line profiles are presented in Fig. 4, in which $V_g$ varies from $-30$ to $20$ V in steps of $10$ V. It can be found that the surface potential difference between the Sm-S junction and the MoS$_2$ background can be efficiently modulated by $V_g$. The potential drops about $10$ mV at $V_g = -30$ V on the lower-$V_g$ side of $\alpha$. This originates from the negatively charged MoS$_2$ surface induced by the electron transfer from SWCNT to MoS$_2$, since the Fermi level of the SWCNT is higher than
that of MoS$_2$ at this region. Their Fermi levels meet at $\alpha$ around -10 to 0 V, where the potential difference disappears (the third and fourth panels in Fig. 4). A reversed contrast of the Sm-S junction appears in the potential images at $V_g = 10$ and 20 V, which can be attributed to the Fermi level of MoS$_2$ overtaking the SWCNT, inducing a positively charged MoS$_2$ surface around the CNT. These results qualitatively agree well with the $\rho (V_g)$ curves shown in Fig. 3C. More detailed information about the measuring system and sample can be found in Materials and Methods and SI Appendix.

**Determination of the Relative Fermi-Level Positions.** By combining the resistivity comparison methodology and the Fermi-level catch-up model, the $V_g$-dependent relative Fermi-level positions of the SWCNT and semiconductor can be efficiently obtained. For the two-component system, the intuitive rule can be summarized from Fig. 3 C and H. The X point should be set as a critical point. On its lower-$V_g$ side, larger resistivity corresponds to higher Fermi-level position; while on the higher-$V_g$ side, the reverse applies. This strategy has great abilities in resolving the Fermi-level positions of the SWCNT and semiconductor can be efficiently. For the two-component system, where several 2D semiconductors are stacked on different segments of a single SWCNT. Since different 2D materials have the same CNT as a reference, their relative Fermi-level positions can thus be determined by the rules of the two-component system. As an embodiment, SWCNT/MoS$_2$ and SWCNT/WSe$_2$ heterojunctions were fabricated on the same metallic SWCNT as shown in Fig. S10A. The CIFS in the SWCNT induced by the two MoS$_2$ flakes was further studied and the comparative resistivities are plotted in SI Appendix, Fig. S9C. The relative Fermi-level positions and the Fermi level catch-up process of the three materials can thus be deduced, which is sketched in SI Appendix, Fig. S9D. Compared with 8 nm MoS$_2$, the Fermi level of 3L MoS$_2$ is lower at $V_g = -30$ V, but higher at $V_g = 30$ V. This reveals that the 3L MoS$_2$ owns a higher conduction band edge and a lower valence band edge, which is consistent with previous reports (43). All the results show that the as-developed strategy has great abilities in resolving the Fermi-level positions as well as providing rich information on the Fermi-level catch-up process in the low-dimensional systems.

**Discussion**

The CIFS in semimetal is prevalent in Sm-S systems for their finite DOS. Similar CIFS behaviors as metallic SWCNT have also been observed in MoS$_2$-covered graphene by using the resistivity comparison methodology. The detailed information about the graphene/MoS$_2$ specimen and the $\rho (V_g)$ curves with four feature points (X, $\beta$, DP, and DP$^*$) are shown in SI Appendix, Fig. S10. Considering the $C_Q$ of graphene (7 $\mu$F/cm$^2$) (44), the modulation of its Fermi level should be within ±50 mV in our experiment. In this range, the variation of $C_Q$ is not significant and Eqs. 1 and 2 also apply; thus the comparative resistivity plots in SI Appendix, Fig. S10D approximate the theoretical deduction (Fig. 3 F3). However, the feature point $\alpha$ is absent in this $V_g$ range, which can be attributed to the different band structure between
the graphene and SWCNT and the screening effect induced by underlying graphene. Because of the CIFS behavior in semimetal, it is necessary to introduce a modification to the Schottky–Mott rule for the low-dimensional Sm-S junctions. The barrier height at the Sm-S interface should be expressed as \( \Phi_B = (\phi_{Sm} - \delta \mu_{Sm}) - \chi_S \), where the CIFS \( \delta \mu_{Sm} \) is a modification term for semimetal work function and actually reflects the direction of the built-in electric field in the semiconductor (SI Appendix, Fig. S11C). The barrier height depends on the gate potentials, since both the work functions of semimetal \( \phi_{Sm} \) and \( \delta \mu_{Sm} \) are gate tunable. The modified rule for Sm-S junctions is significantly different from that for the traditional M-S junctions and it could serve as a fundamental theoretical rule for analyzing low-dimensional nanodevices.

**Materials and Methods**

**Origin of CIFS \( \delta \mu_{Sm} \) and Modification to the Schottky–Mott Rule.** The low-dimensional semimetal with half-filled energy band has finite DOS near the pristine Fermi level, which means that the charge transfer could lead to a significant shift of Fermi level relative to the band. This phenomenon is called the quantum capacitance effect (14), which is defined as

\[
C_0 = e^2 \frac{dQ}{d\mu} = e^2 \nu \frac{dn}{d\mu},
\]

where \( Q \), \( \mu \), and \( n \) are the net charge concentration, the chemical potential (Fermi level), and electron number density in low-dimensional materials. It can be simplified as \( C_0 = e^2 \nu \text{DOS}(\mu) \) at 0 K, and a more complex expression should be given as

\[
C_0(\mu, T) = e^2 \int d\mu' \nu \text{DOS}(\mu') \left| \frac{\partial f(\mu', T)}{\partial \mu'} \right|
\]

at a certain temperature \( T \) due to the thermal broadening of Fermi distribution. That is, the quantum capacitance is positively correlated with DOS.

When a low-dimensional semimetal with finite DOS contacts a semiconductor, the misaligned Fermi level leads to spontaneous charge transfer at the interface to achieve thermodynamic equilibrium. Carrier depletion or accumulation occurs in the semiconductor at the interface; meanwhile, the transferred charge will naturally lead to the Fermi-level shift of the semimetal, making the Schottky barrier height (SBH) of the low-dimensional Sm-S junction deviate from the description of the classical Schottky–Mott rule, as shown in Fig. 1B. A modification term of semimetal work function \( \delta \mu_{Sm} \) needs to be added.

The cases shown in Fig. 1 do not consider the influence of impurities, defects, and other interface states. The charge transfer process satisfies the charge conservation:

\[
\Delta Q_{Sm} + \Delta Q_i = 0.
\]

After contact, electrons transfer from semiconductor to (semi)metal, because of the higher work function of (semi)metal. Using the depletion layer approximation (1), the total charge transferred is

\[
\Delta Q_i = eN_0 W_i = \sqrt{e\varepsilon_0 N_0 V_i},
\]

where \( e \) is the electronic charge, \( N_0 \) is the free electron density, and \( W_i \) is the depletion width.

**Conclusion**

In summary, we fully discussed the Fermi-level shift behavior in low-dimensional Sm-S junctions represented by SWCNT/MoS\(_2\), SWCNT/WSe\(_2\), and graphene/MoS\(_2\). An efficient resistivity comparison methodology to study the CIFS as well as the interfacial charge transfer has been developed. The Fermi-level misalignment between the semimetal and the semiconductor. A simple but effective rule was further developed to efficiently determine the relative Fermi-level positions between CNT and 2D materials at specific gate voltages, which is applicable for both two- and multicOMPONENT systems. In addition, the CIFS in semimetal introduces a modification to the Schottky–Mott rule, giving an accurate expression for the Schottky barrier height of the Sm-S junction. The progress in this paper has an important role in promoting the research on Sm-S junction, which will be the essential building blocks for future low-dimensional nanodevices.
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Device Fabrication. The high-quality ultralong CNTs were grown in suspension using iron as catalyst and ethylene as carbon source. The metallic single-walled CNTs were selected by combining optical and electrical measurements and then transferred onto the target location on the Si/300-nm SiO2 substrate using two tungsten tips. For the device shown in Fig. 2A, the dry transfer method via PDMS was used to stack the mechanically exfoliated MoS2 flake onto the bottom CNT; while the wet method via PDMS/polyvinyl alcohol was used to pick up MoS2 and WSe2 flakes successfully and then construct the heterojunctions shown in Fig. 5A with one step transfer. To achieve clean and tight van der Waals interface, the sample was annealed under vacuum (400 mTorr) at 350 °C for 1 h. Electrodes Ti/Au (5/50 nm) were fabricated through EBL electron beam evaporation, and lift off. Another annealing process was performed to improve the contacts.

Characterization. Electrical measurements were carried out on a semiconductor analyzer (Agilent B1500A) combined with a Lakeshore CRX-4K under vacuum. SEM images were obtained on an FEI Nova NanoSEM 450 with accelerating voltage at 1 kV. Atomic force microscopy (AFM) images were carried out on a Bruker icon in tapping mode. Raman spectroscopy was performed on a Horiba Jobin Yvon LabRAM HR 800 with a 532-nm focused laser.

KPFM measurements were performed under ambient conditions at room temperature and 10% relative humidity (RH). The potential images were obtained using Bruker Multimode 8 in tapping mode. The direct current (DC) bias applied compensates the contact potential difference (CPD) between the tip and sample, \( V_{GS} = V_{TD} \). The CPD can be expressed as \( V_{CPD} = (\phi_{CNT} - \phi_{MoS2}) / q \), where \( q \) is elementary charge unit. The KPFM was conducted in amplitude modulation mode and the lift height was set to 100 nm. The probe is affected by the gate potential during the test, which limits the \( V_{GS} \) range that can be applied to ~30 to 20 V in our experiment.

Data Availability. All study data are included in this article and/or SI Appendix.

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