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Electronic Properties of a 1D Intrinsic/*p*-Doped Heterojunction in a 2D Transition Metal Dichalcogenide Semiconductor

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Abstract

Two-dimensional (2D) semiconductors offer a convenient platform to study 2D physics, for example, to understand doping in an atomically thin semiconductor. Here, we demonstrate the fabrication and unravel the electronic properties of a lateral doped/intrinsic heterojunction in a single-layer (SL) tungsten diselenide (WSe₂), a prototype semiconducting transition metal dichalcogenide (TMD), partially covered with a molecular acceptor layer, on a graphite substrate. With combined experiments and theoretical modeling, we reveal the fundamental acceptor-induced *p*-doping mechanism for SL-WSe₂. At the 1D border between the doped and undoped SL-WSe₂ regions we observe band bending and explain it by Thomas-Fermi screening. Using atomically resolved scanning tunneling microscopy and spectroscopy, the screening length is determined to be in the few nm range, and we assess the carrier density of intrinsic SL-WSe₂. These findings are of fundamental and technological importance for understanding and employing surface doping, *e.g.*, in designing lateral organic-TMD heterostructures for future devices.

Keywords: organic-TMD heterostructure, charge transfer, lateral intrinsic/*p*-doped heterojunction, Thomas-Fermi screening, scanning tunneling microscopy/spectroscopy, photoemission spectroscopy

Recent progress in two-dimensional (2D) electronic materials has brought fascinating prospective applications in next-generation electronic and optoelectronic devices closer to realization.¹ A remaining key challenge is finding robust means to controllably select the majority charge carrier type and tune the Fermi level position (*i.e.*, doping) in the 2D semiconductor, as required by the particular device type.² Among various chemical and physical methods,³⁻¹³ the deposition of organic molecular layers on 2D materials¹⁴⁻¹⁶ is a promising doping approach to avoid structural damage, thereby retaining the desired optical and electrical properties of the 2D materials, as such organic layers are mainly stabilized through non-covalent interactions.¹⁷ Previous studies have revealed that the deposition of suitable molecules can be used to modify the energy levels and the charge density of graphene¹⁸⁻²¹ and semiconducting transition metal dichalcogenides (TMDs)^{14,15} through charge transfer (CT). The situation is more complicated when the heterostructure comprises more than two stacked layers. For example, when graphene is covered with a strong molecular acceptor, it can be *p*-doped or *n*-doped depending on the underlying substrate.¹⁸ Electrostatic interactions (*e.g.*, screening and dipole fields) in multi-layer systems can significantly impact the interfacial properties.²²⁻²⁴ The use of organics also leverages the well-developed organic semiconductor technologies for flexible electronic devices.^{2, 25,26}

Although much work has been done to understand the electronic properties in vertical²⁷⁻²⁹ and lateral³⁰⁻³² 2D-TMD heterojunctions, there is still a lack of studies of the heterojunctions at the atomic scale.³³ In particular, the lateral doping of a single layer with a formation of a 1D interface between doped and intrinsic regions is not understood. An ultimate goal is to fabricate lateral heterojunctions with atomically sharp 1D interfaces to facilitate carrier or energy transport for superior device performance, for example, by selective deposition of a dopant material onto well-defined regions of a TMD layer.⁵

In this study, we explain how a molecular acceptor layer on top of a single-layer (SL) tungsten diselenide (WSe₂) acts as *p*-type dopant, and reveal the electronic structure at a prototypical 1D interface (a line) between *intrinsic* and *p-doped* WSe₂ (Figure 1a). Using a combination of low-temperature scanning tunneling microscopy/spectroscopy (STM/STS) and photoemission spectroscopy (PES) in conjunction with theoretical modeling we explore the molecule/2D semiconductor hybrid on graphite as substrate. The fluorinated fullerene C₆₀F₄₈ is chosen as the molecular acceptor as its electron affinity (5.2 eV³⁴) is higher than the work function (WF) of the

SL-WSe₂ (4.6 eV), which should enable CT. Indeed, we find that negative charge is transferred to the acceptor, however, it mainly originates from the graphite substrate while the SL-WSe₂ becomes polarized by the CT dipole field. The semiconductor layer becomes *p*-doped, as evidenced by a 0.7 eV shift of the Fermi level towards the valence band. This doping is further corroborated by the changed transconductance of a field effect transistor (FET) with WSe₂ as semiconductor, with and without surface-adsorbed C₆₀F₄₈.

Because the molecules adsorb in close-packed islands above a critical coverage, *p*-doped (covered with molecules) and intrinsic (bare) WSe₂ regions co-exist next to each other. The border between the two forms a line, *i.e.*, a 1D interface between *p*-doped and undoped semiconductor regions (Figure 1a). Using spatially resolved STS, the Thomas-Fermi (TF) screening length (or Debye length) at such a 1D interface is determined to be in the nm range. This knowledge is important when considering the use of lateral *p-n* junctions of 2D semiconductors in future electronic devices.

Results

Figure 1b shows a typical large-scale STM image of the CVD-grown SL-WSe₂ on graphite (**Methods** section).³⁵ As revealed by the atomically resolved image in Fig. 1c, the WSe₂ surface away from the edge is clean, smooth and uniform. The WSe₂ unit cell is highlighted by a black rhombus with a lattice constant of 3.4 ± 0.1 Å; a typical moiré superstructure arising from the lattice mismatch between graphite and WSe₂ is highlighted by a blue rhombus. A proposed structural model of the moiré cell is given in the inset in Fig. 1c, corresponding to a (3×3) supercell of WSe₂ with a (4×4) supercell of graphite. The bright half supercell is assigned to Se atoms lying on top of the underlying C atoms; while the dim half is attributed to the Se atoms located at the hollow sites of the graphite substrates. From the dI/dV curve (inset in Fig. 1b), the electronic bandgap of the SL-WSe₂ is deduced to be 1.95 ± 0.05 eV, consistent with previously reported values.^{24, 36}

C₆₀F₄₈ molecules were subsequently deposited onto the clean SL-WSe₂/graphite surface. As shown in Fig. 1d, the molecules aggregate to form close-packed ordered islands with 0.5 ML coverage. The C₆₀F₄₈ unit cell is highlighted by the black rhombus in the bottom left inset in Fig.

1d, with a lattice constant of 1.22 ± 0.04 nm. A similar close-packed $C_{60}F_{48}$ phase is also observed for the $C_{60}F_{48}$ assembly on bare graphite surface.³⁷

To obtain insight into the electronic properties at the $C_{60}F_{48}/WSe_2$ /graphite heterostructure, STS measurements were carried out. Figure 2a shows a close-packed molecular island (right) with bare WSe_2 (left). Fig. 2b (8×8 nm²) is an enlarged part of Fig. 2a near the $C_{60}F_{48}$ island edge (white square). Unlike the uniform WSe_2 surface observed far away from the molecular island or before molecular deposition, a contrast variation is visible close to the island (the upper right corner of Fig. 2b) indicating that the electronic properties of the WSe_2 layer are modified. A series of STS spectra assembled in Fig. 2c were obtained at the positions denoted by colored triangles in Fig. 2a and 2b. The dI/dV spectra obviously vary as a function of the distance (d) from the $C_{60}F_{48}$ island edge. As highlighted by the dashed lines in Fig. 2c, the valence band maximum (VBM) and the conduction band minimum (CBM) shift significantly as the tip moves from the pristine WSe_2 ($d > 5$ nm) towards the $C_{60}F_{48}$ island edge ($d = 0$ nm). As illustrated in Fig. 2d, the VBM shifts upward to the Fermi level from +0.84 eV to +0.22 eV ($\Delta U_{VBM} = 0.62$ eV), while the CBM shifts upwards as well from -1.10 eV to -1.48 eV ($\Delta U_{CBM} = 0.38$ eV), consequently implying a bandgap decrease of $\Delta E_g = 0.24$ eV.

The data in Fig. 2 thus strongly suggest that the bare WSe_2 is intrinsic and it is *p*-doped when covered by $C_{60}F_{48}$, while a region of band bending forms between the two. It might be straightforward to presume that electron transfer from WSe_2 to $C_{60}F_{48}$ causes the *p*-doping of the 2D TMD semiconductor. But previous observations for a molecular acceptor adsorbed on graphene have highlighted the importance of the underlying substrate in tuning the electronic structure of a 2D material,¹⁸ and thus the graphite substrate must be included in our considerations in due course.

To understand the origin of the CT, we calculated the differential charge density (DCD) across the organic-TMD heterostructure including the graphite substrate, as well as those of reference systems. The side-views of the DCD isosurfaces for $C_{60}F_{48}$ on graphite, SL- WSe_2 /graphite, and SL- WSe_2 alone are plotted in Figure 3a, 3c and 3e, respectively. A (7×7) graphene supercell and a ($3\sqrt{3} \times 3\sqrt{3}$) WSe_2 supercell were used (**Methods**). The adsorption of the $C_{60}F_{48}$ molecule on a double bond in between hexagons was used in the calculations (SI). The DCD plots reveal that the $C_{60}F_{48}$ molecule is negatively charged in all cases, and the electrons predominantly

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3 accumulate at the lower half of the molecule. In turn, all substrates are positively charged.
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5 However, the sandwiched WSe₂ single-layer in Fig. 2c remains essentially charge-neutral but
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7 becomes polarized, as the top Se layer facing the molecule is hole-accumulated, while the bottom
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9 facing graphite is electron-accumulated.

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11 To provide a more quantitative picture, the plane-integrated DCD, $\Delta\rho(z)$ (red), and the
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13 cumulative CT, $\Delta Q(z)$ (blue) are plotted in Fig. 3b, 3d and 3f; the latter describes how much
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15 charge has been transferred from the bottom to the top at position z . The total amount of CT
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17 (ΔQ_{total}) from graphite to C₆₀F₄₈ reaches -0.40 e per molecule in Fig. 3b, and reduces slightly to -
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19 0.38 e with the SL-WSe₂ interlayer in Fig. 3d. Two pronounced peaks can be observed in Fig. 3d:
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21 the top one at -0.38 e represents the CT from WSe₂/graphite to C₆₀F₄₈, and the lower one at -
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23 0.25 e is that from graphite to C₆₀F₄₈/WSe₂. The difference of these two peaks, -0.13 e , is then
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25 the contribution from the WSe₂ interlayer, which is only half of that from the graphite substrate.
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27 Consequently, the graphite contributes about two thirds of the charge to the molecular acceptor
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29 while the SL-WSe₂ contributes only one third, contradicting the straightforward presumption of
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31 dominant TMD-molecule CT. When the graphite substrate is removed, the CT from the WSe₂ to
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33 C₆₀F₄₈ is only -0.29 e as seen in Fig. 3f. The underlying graphite substrate thus plays an
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35 important role in defining the electronic structure of the heterojunction, as speculated above.

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37 Further calculations suggest that the C₆₀F₄₈/WSe₂/graphite heterojunction has a CT-induced
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39 interfacial dipole moment of 2.65 $e\text{\AA}$, which is much larger than that of C₆₀F₄₈/graphite without
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41 the WSe₂ interlayer (1.70 $e\text{\AA}$) and that of for C₆₀F₄₈/WSe₂ without the graphite substrate (1.31
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43 $e\text{\AA}$) (SI). The dipole field induced by the CT at the heterojunction can significantly impact the
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45 electronic properties (*e.g.*, energy level alignment) and is linked with the CT. In fact, the
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47 bandgap decrease of the WSe₂ interlayer observed by STS spectra (Fig. 2c) is induced by the
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49 vertical electric field, known as Stark effect.^{38,39}

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51 To substantiate the conclusions from STM/STS and theoretical modeling, and to highlight the
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53 effect of coexisting differently intrinsic/doped WSe₂ patches in area-averaging experimental
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55 techniques, we performed PES measurements to determine changes of WF (Fig. 4a) as well as
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57 valence band (Fig. 4b and 4c) and core level spectra (Fig. 4d) as a function of C₆₀F₄₈ coverage.
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59 The spectra of the clean graphite substrate are shown as black dotted lines for comparison. The
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WF of graphite (4.5 eV) increases by less than 0.1 eV when covered with SL-WSe₂ (0 Å),

indicating negligible electrostatic interaction. After the adsorption of $C_{60}F_{48}$, the WF increases significantly (Fig. 4a). The WF change saturates at a nominal coverage Θ of 8 Å, which is identified as $C_{60}F_{48}$ monolayer coverage. The total WF increase of 1.56 eV with a $C_{60}F_{48}$ monolayer indicates the formation of an interfacial dipole induced by the CT across the heterostructure, consistent with theoretical modeling.

In Fig. 4b, the SL-WSe₂ shows two distinct features in the valence region at 1.5 eV and 3.9 eV binding energy (BE). The VB edge of the pristine SL-WSe₂, namely the lower binding energy onset of the first peak, is at 1.17 eV, slightly larger than the value of the VBM determined by STS (0.84 eV). The most probable reason for the difference is that the VBM of SL-WSe₂ is located at the K point as determined by ARPES,⁴⁰ however, our VB spectra measured at normal emission are corresponding to electronic states around the Γ point. The VB edge around the Γ point is energetically deeper than that at the K point. Therefore, it is reasonable that the VB edge measured by PES, corresponding to the region surrounding the Γ point, is further away from the Fermi level than the VBM determined by STS.

The intensities of the two distinct peaks reduce with increasing $C_{60}F_{48}$ coverage. Broadening and BE shifts can be better seen in the zoom-in spectra in Fig. 4c after background-subtraction and normalization. The pristine WSe₂ feature at 1.5 eV BE (red) initially shifts 0.1 eV to lower BE with the adsorption of $C_{60}F_{48}$. Concomitantly, a shoulder emerges at the lower BE side and becomes dominant as the $C_{60}F_{48}$ coverage increases. At full monolayer coverage (8 Å) a single peak at 0.8 eV (orange) remains instead of the pristine WSe₂ peak. With accurate fitting, we find that a third contribution (green) is present for the partially covered WSe₂ (e.g., 6 Å coverage), which is located halfway between the pristine (red) and the $C_{60}F_{48}$ -covered (orange) WSe₂ peaks, and is about twice as broad as the other two peaks. The spectral weight of this contribution firstly increases with increasing $C_{60}F_{48}$ coverage and decreases again while approaching monolayer coverage. In accordance with the band bending region existing near the $C_{60}F_{48}$ island edge (Fig. 2), we assign this contribution to the transition area between the $C_{60}F_{48}$ covered and uncovered WSe₂. Meanwhile, the peak feature (orange) arising from the $C_{60}F_{48}$ -covered WSe₂ is constant at 0.8 eV BE from the very beginning (e.g., 4 Å), indicating an abrupt change for the *p*-doped WSe₂ to the maximum, instead of a gradual shift with the increasing coverage. The 0.7 eV shift of the valence levels towards the Fermi level is identical to the BE shift observed in the core levels, e.g.,

W 4f in Fig. 4d and Se 4d in SI, *i.e.*, all electronic levels shift in parallel due to electrostatic reasons (the CT-induced dipole field). This value is consistent with that of 0.62 eV (ΔU_{VBM}) obtained from STS spectra in Fig. 2.

The STM/STS and PES experiments clearly demonstrate that the intrinsic and *p*-doped WSe₂ patches coexist next to each other, and the border between them forms a 1D (line) intrinsic/*p*-doped interface in the semiconducting WSe₂ interlayer. The gradual band bending over several nm observed from the spatially resolved STS spectra (Fig. 2d) is due to in-plane electrostatic screening. Using the non-linear Thomas-Fermi theory to interpret dielectric screening in a semiconductor,⁴¹ the change of the potential as a function of distance is $\Delta U \propto e^{-d/L_{\text{TF}}}$, where L_{TF} is the TF screening length (SI). Fitting the curve in Fig. 2d with this equation (SI), L_{TF} of 2.3±0.7 nm is deduced in the intrinsic WSe₂ side to the hole dopant edge. The in-plane screening length in a 2D semiconductor is a key parameter for applications in electronic devices, and a reliably determined value can aid device design.

Finally, an FET was fabricated to evaluate the carrier densities and the transport properties (SI) of SL-WSe₂ layer without and with C₆₀F₄₈. Figure 5a shows the optical microscope image of a back-gate SL-WSe₂ FET on a highly *p*-doped silicon substrate with 300 nm SiO₂ (**Methods**). Transfer characteristics of the FET before and after C₆₀F₄₈ deposition were investigated by the source-drain current *versus* gate voltage, namely $I_{\text{sd}}-V_{\text{g}}$ curve, as shown in Fig. 5b. An ambipolar characteristic is revealed by the inserted logarithmic plot, as I_{sd} increases with both negative and positive voltages. A much faster current increase is observed in the negative region compared to the positive side, suggesting a hole dominated transport behavior of the intrinsic SL-WSe₂. By linear extrapolation, the threshold voltage V_{th} of the as-made FET device is determined to be -51.6 V for hole transport. After the deposition of 20 nm C₆₀F₄₈, V_{th} for holes shifts to -36.6 V, indicating an effective hole-doping. This phenomenon is consistent with the above theoretical calculations shown in Fig. 3e and 3f, where charge transfer from SL-WSe₂ to C₆₀F₄₈ is possible without underlying substrate, analogy to the case with an insulator substrate like 300 nm SiO₂/Si. The carrier (hole) concentration can be estimated by using the equation $N_{\text{h}} = -C_{\text{i}}(V_{\text{g}} - V_{\text{th}})/e$, where C_{i} is the gate capacitance per unit area ($1.15 \times 10^{-4} \text{ Fm}^{-2}$ for the 300 nm SiO₂ layer). The hole concentration at $V_{\text{g}} = -55 \text{ V}$ is derived to be $2.4 \times 10^{11} \text{ cm}^{-2}$ for the as-made SL-WSe₂, and it increases to $1.2 \times 10^{12} \text{ cm}^{-2}$ after C₆₀F₄₈ doping.

If the dopant density is known, the TF screening length of a semiconductor can also be estimated by $L_{TF} = \sqrt{\epsilon k_B T / e^2 N_h}$, where $\epsilon = \epsilon_r \epsilon_0$ is the dielectric constant (SI). Here, we use $\epsilon_r = 7.25$ for WSe₂,⁴² $T = 300$ K and $N_h = 2.4 \times 10^{11} \text{ cm}^{-2}$ obtained from the FET data. This yields $L_{TF} = 1.7$ nm for intrinsic SL-WSe₂ on a SiO₂/Si substrate at room temperature, which is in good agreement with the value determined by STM/STS on a graphite substrate at 77 K. On the other hand, the carrier density can be estimated from the known L_{TF} , as $N_h = kT\epsilon/e^2 L_{TF}^2$. By using $L_{TF} = 2.3 \pm 0.7$ nm obtained from STS, we have $N_h = (4 \pm 2) \times 10^{10} \text{ cm}^{-2}$ at 77 K, which would increase by fourfold to reach the range of 10^{11} cm^{-2} at room temperature, comparable to the value for the intrinsic SL-WSe₂ on SiO₂/Si obtained from the FET. We thus find high reliability of carrier density and screening length values determined by spatially resolved STM/STS and those derived from transport measurements. We suggest that STS can be used in general to estimate the carrier density in 2D semiconductors with laterally doped structures, through the determination of the TF screening length at a 1D interface in a lateral heterojunction.

Conclusion

The electronic properties of a prototypical molecule/TMD heterostructure on a graphite substrate were elucidated by STM/STS and PES measurements combined with first-principles calculations. The semiconducting SL-WSe₂ can be efficiently *p*-doped by an adsorbed C₆₀F₄₈ layer, and a lateral intrinsic/*p*-doped heterojunction is thereby realized in SL-WSe₂. C₆₀F₄₈ acts as an electron acceptor, however, the negative charge originates mainly from the graphite substrate. In fact, the WSe₂ monolayer is strongly polarized by the CT dipole field between graphite and the acceptor layer, and it becomes significantly *p*-doped, accompanied by a bandgap narrowing. The 1D doped/undoped interface features a band bending region that was spatially resolved by STM/STS mapping, and identified as a transition feature in surface-averaged PES spectra. The Thomas-Fermi screening length of intrinsic SL-WSe₂ on the graphite substrate was determined to be 2.3 nm at 77 K by STM/STS, consistent with the value of 1.7 nm at room temperature derived from transport measurements in an FET. The comprehensive understanding of the doping mechanism in 2D semiconductors described here is of fundamental importance for developing this technique further. Likewise, it is of technological relevance, as 2D TMD materials and their

heterojunctions (lateral and vertical) have fascinating prospective applications in future flexible and wearable devices.

Methods

Sample preparations for STM and PES measurements

The monolayer WSe₂ samples were synthesized by chemical vapor deposition (CVD) method using WO₃ powders as precursor. More details about the CVD growth can be found in previous reports.³⁵ The sample used for STM measurements was directly grown on a graphite substrate to avoid contaminations introduced by chemical transfer. The overall coverage of SL-WSe₂ is ~10%, estimated by performing statistical analysis over the graphite surface. For PES measurements, a homogeneously large-scale SL-WSe₂ film with size over 5×7 mm² and coverage higher than 80% was synthesised on sapphire substrates first and then transferred by a wet chemical method to the graphite substrate. All the WSe₂ samples were degassed overnight at 300 °C - 450°C with base vacuum better than 10⁻⁹ mbar before ultrahigh vacuum STM and PES measurements. The cleanliness and quality of the WSe₂ crystals were confirmed by STM imaging, STS spectra and XPS wide scan spectra (SI).

C₆₀F₄₈ (Nano-C, 99.6%) molecules were deposited from a Knudsen effusion cell with a typical deposition rate of ~0.04 ML/min. Here, one monolayer (ML) refers to one full monolayer C₆₀F₄₈ in close-packed configuration. The deposition rate of C₆₀F₄₈ was monitored by a quartz-crystal microbalance (QCM) during evaporation, and the overall coverages were further calibrated by taking statistic over large-scale STM images or monitoring the attenuation in intensity of the C 1s peak during PES measurements. During the deposition, the substrates were held at room temperature and the samples were subsequently transferred *in-situ* to the attached STM chamber or PES analysis chamber for analysis.

Characterization

LT-STM studies were performed in an Omicron high-resolution STM interfaced to a Nanonis controller at the National University of Singapore.^{6, 24, 36} All STM/STS measurements were recorded at 77K. An electrochemically etched tungsten tips were used and bias voltages were applied to the STM tip. Tip bias voltage was used in the paper for the convenience to compare

with PES spectra. STM images were recorded by using constant current mode with a tunnelling current I_{Tip} of about 0.05 nA. For dI/dV spectra, the tunnelling current was obtained by a lock-in amplifier, with a modulation of 625 Hz and 40 mV.

All the PES measurements in this study were conducted at Humboldt Universität zu Berlin. For UPS, a Scienta He discharge lamp ($h\nu = 21.21$ eV) was used. XPS was conducted using a Scienta DAR 400 x-ray source. The Mg K_{α} line (1253.69 eV) was used for excitation. The emitted electrons were detected with a Specs Phoibos 100 hemispherical energy analyzer in fixed analyzer transmission mode and with an acceptance angle of $\pm 7^{\circ}$. The sample was biased at -10V when measuring the secondary electron cut-off (SECO) to overcome the work function of the analyzer. A clean gold crystal was used to calibrate the work function of the detector and to determine the energy resolution (150 meV) of the system. The reproducibility of all energy values in this study was within 0.1 eV.

Computation

The first-principles calculations were carried out based on density functional theory (DFT), employing Projector augmented wave pseudopotentials^{43,44} and the Perdew-Burke-Ernzerhof form⁴⁵ of the exchange-correlation functional, as implemented in the Vienna *ab initio* simulation package code.⁴⁶ The experimental graphite lattice constant of 2.46 Å was used. The SL-WSe₂ on a single-layer graphene was optimized by using the van der Waals density functional method (optB88-vdW).^{47,48} We employed an energy cutoff of 400 eV for plane waves, and the criterion for total energy convergence was set to 10^{-4} eV. All atoms were relaxed during geometry optimization until the magnitude of forces was less than 0.01 eV/Å. For the plots shown in Fig. 3, a $(3\sqrt{3}\times 3\sqrt{3})$ WSe₂ supercell on a (7×7) graphene supercell (with stress of $\sim 0.3\%$) including a C₆₀F₄₈ molecule and at least 17 Å of vacuum was used. The systems without the SL-WSe₂ interlayer and without graphene substrate were also calculated for comparison. Note that the molecular density in these models is only half of the close-packed phase shown in Figs 1 and 2, more calculations had been performed to understand the effect of the molecular density on the total amount of the charge transfer which will be presented in a paper under preparation.³⁸ The calculations based on a model including a 3-layer graphite slab are shown in SI.

Device

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3 A monolayer WSe₂ was mechanically exfoliated from bulk WSe₂ crystals using a scotch tape and
4 transferred onto degenerately *p*-doped silicon wafers coated with 300 nm SiO₂. The source and
5 drain electrodes were precisely patterned on the flake using conventional e-beam lithography
6 technique, followed by thermal evaporation of Pd(20 nm)/Au(50 nm) as the metal contacts. After
7 liftoff the as-made device was wire bonded onto a leaded chip carrier and loaded in a custom-
8 designed high vacuum system ($\sim 10^{-8}$ mbar) for *in situ* electrical measurements. The transfer
9 characterization ($I_{\text{sd}}-V_{\text{g}}$) of the FET devices was carried out by an Agilent 2912A source measure
10 unit at room temperature.
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20 21 **Acknowledgements**

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43 **Author Contributions**

44
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50 **Additional information**

51
52 Supporting Information Available: Adsorption configurations of C₆₀F₄₈ molecules; Theoretical
53 calculations on different systems; XPS full spectrum of the clean WSe₂/Graphite substrate; Se 3d,
54 C 1s and F 1s core levels for C₆₀F₄₈/SL-WSe₂/graphite; Derivation of the change of the
55 electrostatic potential in the SL-WSe₂ on Graphite; Deduce the TF screening length by fitting the
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spatial dI/dV spectra; The carrier density for SL-WSe₂ estimated by the TF screening length; Derivation of Thomas Fermi Screening length in semiconductors; Transport properties of the SL-WSe₂ layer uncovered and covered with C₆₀F₄₈; PL spectra of the SL-WSe₂ layer on graphite with and without C₆₀F₄₈. This material is available free of charge via the Internet at <http://pubs.acs.org>.

Competing financial interests

The authors declare no competing financial interests.

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Figure 1. Fabrication of organic/TMD/graphite heterojunction. (a) A schematic model of the C₆₀F₄₈/SL-WSe₂/graphite heterostructure. (b) A large-scale STM image of a SL-WSe₂ flake on graphite (250×250 nm²; V_{Tip} = -3.1 V). Inset: STS spectrum reveals a 1.95 eV bandgap for the clean SL-WSe₂ (set point: V_{Tip} = -0.9V, I_{Tip} = 100 pA). (c) Moiré superstructure is visible on the SL-WSe₂ (6×6 nm²; V_{Tip} = -0.8V). The proposed model corresponds to the moiré superstructure is shown in the inset, where the Se atoms on the top/hollow sites of the graphite are represented by gold/green. Blue and grey atoms represent W and C respectively. (d) 0.5 ML C₆₀F₄₈ molecules form close-packed islands on the SL-WSe₂/graphite surface (80×200 nm²; V_{Tip} = -3.1 V). The zoom-in of the close-packed island is shown in the bottom left inset in (d) (6×6 nm²; V_{Tip} = -3.1 V).

158x215mm (300 x 300 DPI)

Figure 2. 1D intrinsic/p-doped heterojunction in SL-WSe₂. (a) A molecular-resolved STM image shows a close-packed $C_{60}F_{48}$ island (0.5 ML) on the SL-WSe₂ (20×20 nm²; V_{Tip} = -3.1 V), and (b) is the close-up corresponding to the white dotted square in the bare SL-WSe₂. Contrast variation is observed close to the molecular island edge (8×8 nm², V_{Tip} = -0.9 V). (c) dI/dV spectra recorded at the positions denoted by the multicolored triangles in panel (a) and (b) (set point: V_{Tip} = -0.9 V, I_{Tip} = 100 pA). The variation of the WSe₂ bandgap is getting pronounced as the STM tip approaches to the $C_{60}F_{48}$ island. (d) Schematic diagram shows the band bending of the SL-WSe₂ as a function of the distance from the molecular island, due to the TF screening effect.

138x111mm (300 x 300 DPI)

Figure 3. Calculations of the charge transfer. (a), (c) and (e) are side views of DCD isosurfaces of a C60F48 molecule on a single-layer graphene, SL-WSe2/graphene, and SL-WSe2, respectively (isovalue: ± 0.001 $e/\text{\AA}^3$). A (7×7) graphene supercell and a $(3\sqrt{3} \times 3\sqrt{3})$ WSe2 supercell were used. Gray, green, brown and pearl gray balls represent W, Se, C and F atoms, respectively. Blue (red) represents electron accumulation (depletion) regions. (b), (d) and (f) represent the corresponding plots of the plane-averaged $\Delta\rho(z)$ (red) and $\Delta Q(z)$ (blue).

138x296mm (300 x 300 DPI)

Figure 4. PES spectra of the C60F48/WSe2/graphite heterostructure. (a) The secondary cut-off shifts to higher kinetic energies with the increase of C60F48 coverage, indicating a WF increase. (b) Valence band spectra show that the sharp WSe2 peaks are pronounced at around 1.5 eV and 4 eV before molecular deposition (0 Å), and strongly attenuated for coverages higher than 4 Å. The dashed black line in (a) and (b) represents the clean graphite for comparison. (c) Zoom-in spectra of the valence region after Shirley-background subtraction and normalization. Three different contributions with fixed energetic positions can be clearly separated. The red peak stems from the uncovered WSe2, the orange peak is shifted by 0.7 eV to lower BE and stems from the WSe2 covered by C60F48, and the green peak stems from the transition region between covered and uncovered WSe2. (d) W 4f core levels shift by 0.7 eV to lower BE with C60F48 coverage increasing.

131x73mm (300 x 300 DPI)

Figure 5. Transport measurements on a SL-WSe₂ FET without and with C60F48. (a) Optical image of the as-fabricated WSe₂ device. (b) Transfer characteristics (I_{sd} - V_g) of the SL-WSe₂ device before (black) and after 20 nm C60F48 deposition (red) with $V_{sd} = 1$ V. Inset: logarithmic plot of the transfer curve.

70x29mm (300 x 300 DPI)

61x29mm (300 x 300 DPI)