

# Hole mobility enhancement in monolayer WSe<sub>2</sub> p-type transistors through molecular doping

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**Abstract** Two-dimensional (2D) transition metal dichalcogenide (TMDC) semiconductor materials exhibit extraordinary electrical properties, holding promise for the realization of next-generation complementary metal-oxide-semiconductor (CMOS) devices at ultimate scaling. However, constrained by effective device doping strategies, the hole mobility and device performance of tungsten diselenide (WSe<sub>2</sub>) p-type transistors, especially monolayer chemical vapor deposition (CVD)-grown WSe<sub>2</sub>, have not met expectations. In this paper, an effective performance enhancement of monolayer WSe<sub>2</sub> p-type transistor was achieved through a molecular doping strategy. Synthesizing monolayer WSe<sub>2</sub> directly on SiO<sub>2</sub> back-gated substrates and leveraging energy band alignment design, 4-nitrobenzenediazonium tetrafluoroborate (4-NBD) molecular dopant with a concentration of 10 mM was utilized to modulate the Fermi level position of monolayer WSe<sub>2</sub> for hole doping. The devices demonstrated a more than 98% increase in hole mobility, reaching up to 97 cm<sup>2</sup> · V<sup>-1</sup> · s<sup>-1</sup> while maintaining the current on/off ratio of 10<sup>8</sup>. Monolayer p-type WSe<sub>2</sub> transistors with 1 μm channel length exhibit a high drive current surpassing 176 μA · μm<sup>-1</sup>, exceeding previous CVD-WSe<sub>2</sub> devices with similar channel length. This straightforward and effective approach to improving the electrical performance of WSe<sub>2</sub> transistors paves the way for advanced logic technologies based on transition metal dichalcogenide semiconductors.

**Keywords** CVD-grown WSe<sub>2</sub>, molecular doping, 4-NBD, p-type transistors, hole mobility

## 1 Introduction

Two-dimensional (2D) transition metal dichalcogenides (TMDCs) offer a wide range of tunable bandgaps and extraordinary electrical properties, rendering them highly attractive for various applications, especially in electronic devices [1,2]. Theoretical considerations suggest that 2D materials possess far superior mobility compared to silicon at extremely scaled thicknesses, presenting potential advantages for ultimate transistor scaling for advanced logic devices [3,4]. Recently, molybdenum disulfide, as the most typical n-type TMDC material, has made remarkable progress in wafer-scale material synthesis of chemical vapor deposition and device performance optimization [5–7]. To further promote advancement of 2D material complementary metal-oxide-semiconductor (CMOS) technology, p-type 2D TMDC transistor technologies with matched threshold voltage and device performance are very necessary and desired. Among the prime candidates for p-type transistor technologies, WSe<sub>2</sub> stands out as one of the best candidates due to its more balanced conduction and valence band edges to different work function metals and impressive hole mobility [8–10]. Nevertheless, previous studies have encountered challenges related to mobility degradation, particularly concerning scalable chemical vapor deposition (CVD) monolayer WSe<sub>2</sub> [11–14].

Controlled channel doping techniques offer a practical means to modulate the electrical properties of 2D transistors [15,16]. Different from the complex doping process technologies established in the traditional semiconductor industry, such as ion implantation and thermal diffusion, the ultrathin nature

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and expansive surface area of 2D materials make them sensitive to the surrounding environment and thus amenable to modulation of surface charge transfer doping. In the previous studies, substitution doping and chemical adsorption were reported frequently. Partial substitution of W atoms with Nb achieves p-type character, the doping process needs to be done during WSe<sub>2</sub> growth while it is easy to over-dop resulting in very weak gate dependence. Besides, electrostatic doping is the most commonly used doping technique in 2D device technology. However, the doping concentration of electrostatic doping is limited by the dielectric breakdown, and it also requires additional electrode terminals and power supply, presenting challenges in the miniaturization of transistor technology. Typically, gases with oxidizing characteristics like NO<sub>x</sub>, H<sub>2</sub>O, and O<sub>2</sub> have been utilized as doping agents to enhance p-type device performance, as reported in previous studies [13,17]. However, physical adsorption gases rapidly desorb and revert to their original state under vacuum conditions, while chemical adsorption may alter the original band structure, leading to device performance or electrostatic control degradation [12–14,18]. To achieve effective enhancement of p-type transistor performance, one promising method involves using redox-active molecules capable of integer electron transfer reactions with TMDCs as dopants, yielding more robust and non-destructive doping effect [19,20].

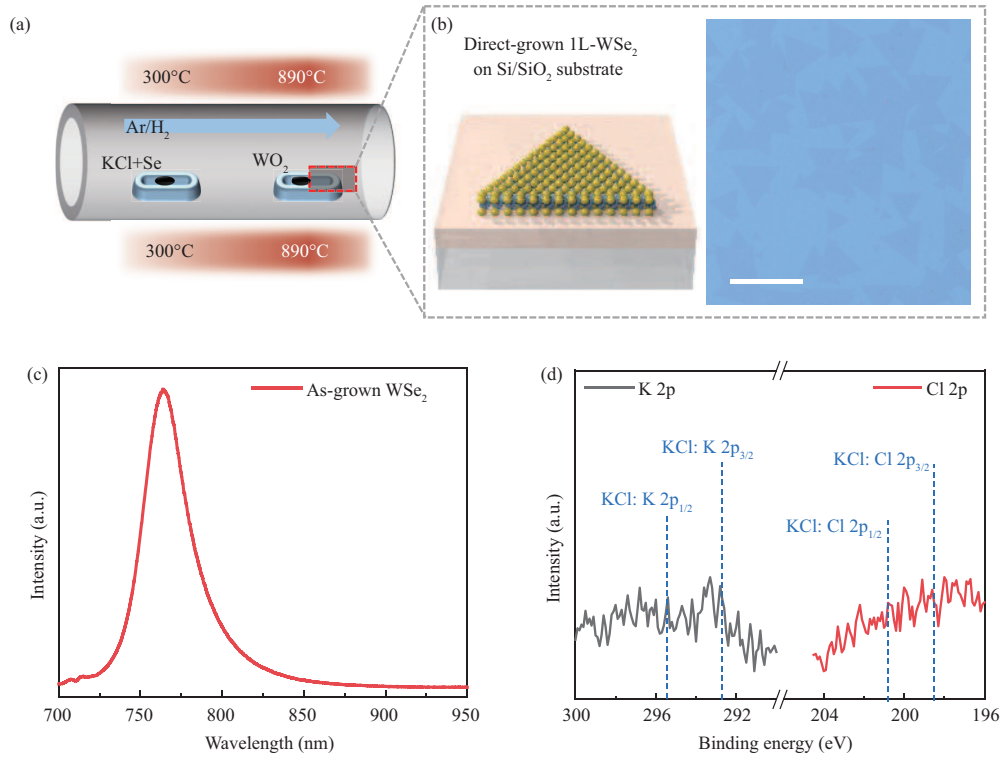
In this study, we demonstrated an effective chemical doping approach for low-pressure chemical vapor deposition (LPCVD) direct-grown monolayer WSe<sub>2</sub> field-effect transistors (FETs) employing 4-nitrobenzenediazonium tetrafluoroborate (4-NBD) p-dopants. Validation of efficient surface charge transfer effects was achieved through Raman and X-ray photoelectron spectroscopy (XPS) spectral analyses. The field-effect hole mobility of WSe<sub>2</sub> transistors increased significantly from the pristine 49 to 97 cm<sup>2</sup> · V<sup>-1</sup> · s<sup>-1</sup>, marking a remarkable 98% enhancement under the doping concentration of 10 mM. Importantly, unlike the degradation in off-state current reported in previous studies [14,18] due to chemical doping, this study achieved improvements in the off-state current while maintaining a high on/off ratio of up to 1.2 × 10<sup>8</sup>. Moreover, a high drive current exceeding 176 μA · μm<sup>-1</sup> was achieved on the WSe<sub>2</sub> transistors with a 1 μm channel length, surpassing previously reported monolayer CVD p-type WSe<sub>2</sub> devices with the same channel length [19,21–23]. This strategy offers a straightforward and effective means to boost the electrical performance of 2D p-type WSe<sub>2</sub> transistors, showcasing great potential in fabricating 2D CMOS applications.

## 2 Results and discussion

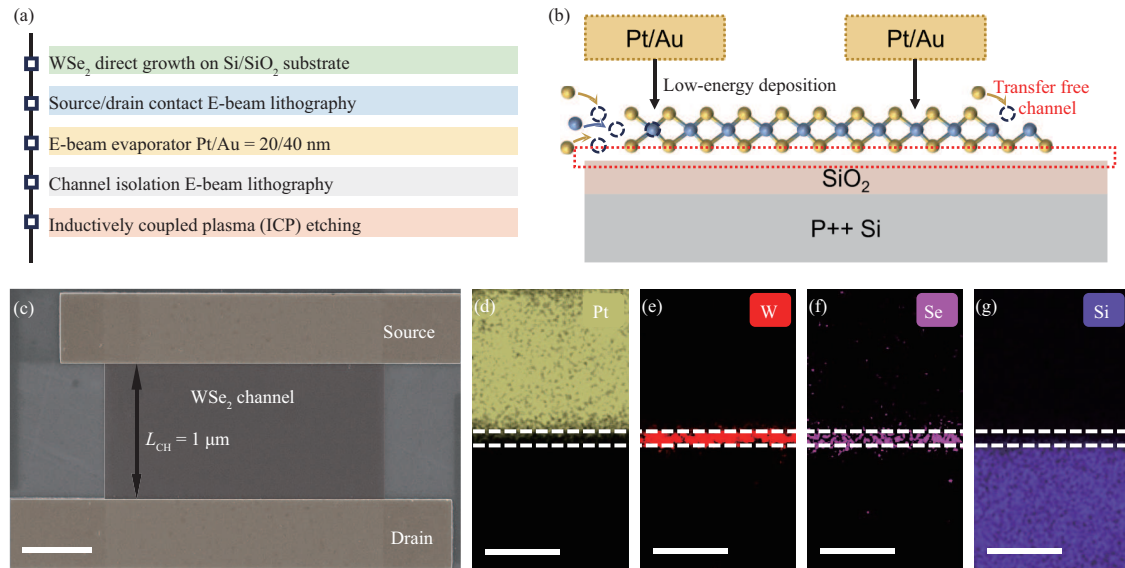
Monolayer WSe<sub>2</sub> was grown by molten-salt-assisted LPCVD process using WO<sub>2</sub> and Se precursors on a silicon substrate covered by 10 nm SiO<sub>2</sub> grown by atomic layer deposition (ALD). During the growth, 100 sccm of Ar was introduced as the carrier gas and 3 sccm H<sub>2</sub> as the reducing gas to reduce WO<sub>2</sub> to WO<sub>2-x</sub>. An additional potassium chloride (KCl) was added to WO<sub>2</sub> powder to further reduce the formation energy of WSe<sub>2</sub> growth, as depicted in Figure 1(a). The morphology of molten salt-assisted LPCVD monolayer WSe<sub>2</sub> and an optical microscope photograph of the as-grown monolayer WSe<sub>2</sub> are shown in Figure 1(b), with a single crystal domain size of 80 μm. Photoluminescence (PL) spectra in Figure 1(c) exhibit a strong photoluminescence peak at 764 nm as the excitation of direct bandgap, suggesting a pure phase of monolayer WSe<sub>2</sub> with a bandgap of 1.6 eV [24,25]. To exclude the doping of KCl during the growth process, the high-resolution X-ray photoelectron spectroscopy spectrum in Figure 1(d) of Cl 2p and K 2p was measured, where no evident peak was observed at 198.3, 200.3, 292.7, and 295.5 eV, indicating no KCl residue in WSe<sub>2</sub> film during the LPCVD process.

A key feature of this study is that WSe<sub>2</sub> can be grown directly on SiO<sub>2</sub> substrates without any transfer process, indicating that the proposed method is compatible with silicon-based substrates, providing a feasible path for integrating two-dimensional materials into existing silicon-based electronic devices. The device fabrication process in this study follows the integration process flow as shown in Figure 2(a). All patterning processes use standard electron-beam lithography. The substrate is low-resistance silicon with 10 nm ALD-SiO<sub>2</sub> back-gate dielectric. The source/drain contact was metalized with 20/40 nm Pt/Au at a deposition rate of 0.1–0.3 Å · s<sup>-1</sup>, following the lift-off process. Inductively coupled plasma (ICP) was used to etch and define the active region. To minimize organic residues during fabrication and reduce carriers scattering, the transfer-free channel and low-energy source-drain metal evaporation process are crucial steps in achieving high mobility transistors, as depicted in Figure 2(b).

The scanning electron microscope (SEM) image of the channel region in Figure 2(c) shows no apparent process chemical residues in the channel and source/drain contact areas, benefiting from the transfer-

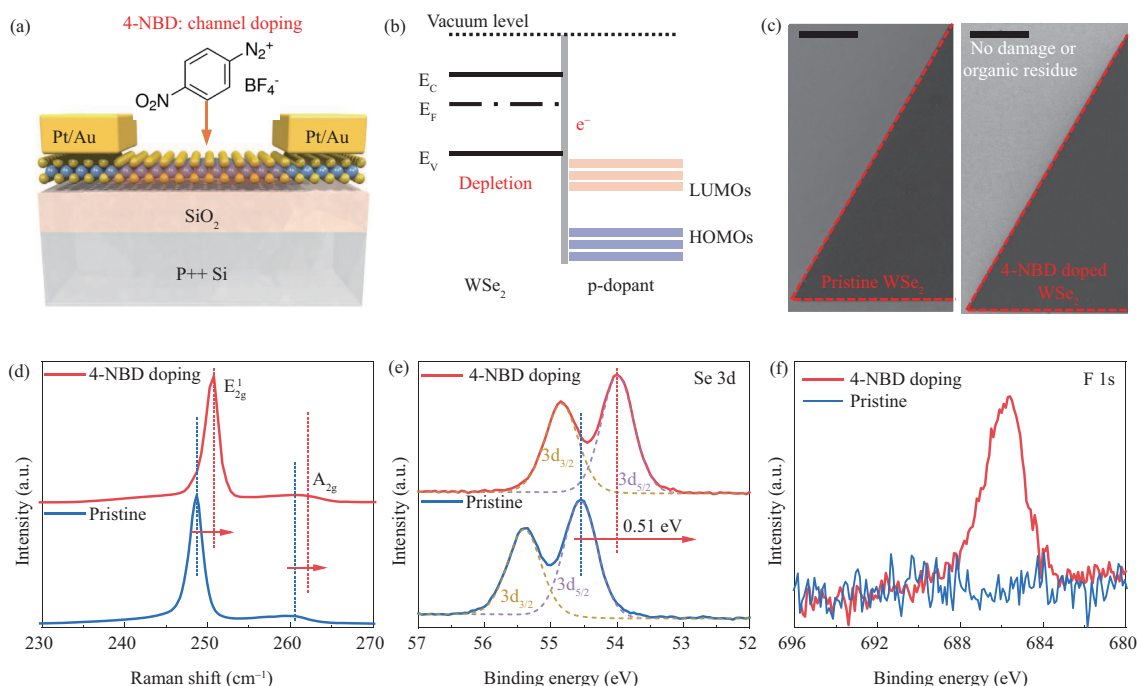


**Figure 1** (a) Schematic of molten-salt-assisted LPCVD direct-growth of 1L-WSe<sub>2</sub> on Si/SiO<sub>2</sub> substrate; (b) schematic of 1L-WSe<sub>2</sub> structure on Si/SiO<sub>2</sub> substrate and corresponding optical micrograph of triangular grains (scale bar, 100 μm); (c) PL spectra of as-grown monolayer WSe<sub>2</sub>; (d) high-resolution XPS spectra of Cl/K for as-grown WSe<sub>2</sub>, the absence of Cl 2p and K 2p indicating no KCl residue in WSe<sub>2</sub> film during molten-salt-assisted LPCVD process.



**Figure 2** (a) Key process steps for fabricating WSe<sub>2</sub> transistors; (b) schematic of a transfer-free WSe<sub>2</sub> transistor with low-energy deposited Pt/Au contacts on a 10 nm SiO<sub>2</sub> dielectric; (c) SEM image of a WSe<sub>2</sub> transistor with a channel length of 1 μm (scale bar, 500 nm); (d)–(g) cross-sectional EDX elements mapping of the contact region, detailing the interface of Pt contact and SiO<sub>2</sub> dielectric with monolayer WSe<sub>2</sub> channel (scale bar, 10 nm).

free channel material. A clean and sharp interface between contact metal and WSe<sub>2</sub> as well as back gate dielectric and WSe<sub>2</sub> is established due to the transfer-free WSe<sub>2</sub> channel and low-energy metal deposition. The energy dispersive X-ray spectroscopy (EDX) mapping of Pt, W, Se, and Si elements reveals the spatially resolved elemental distributions with an abrupt change across the neighboring films shown in Figures 2(d)–(g), further confirming the clear interfaces and absence of damage within the WSe<sub>2</sub>



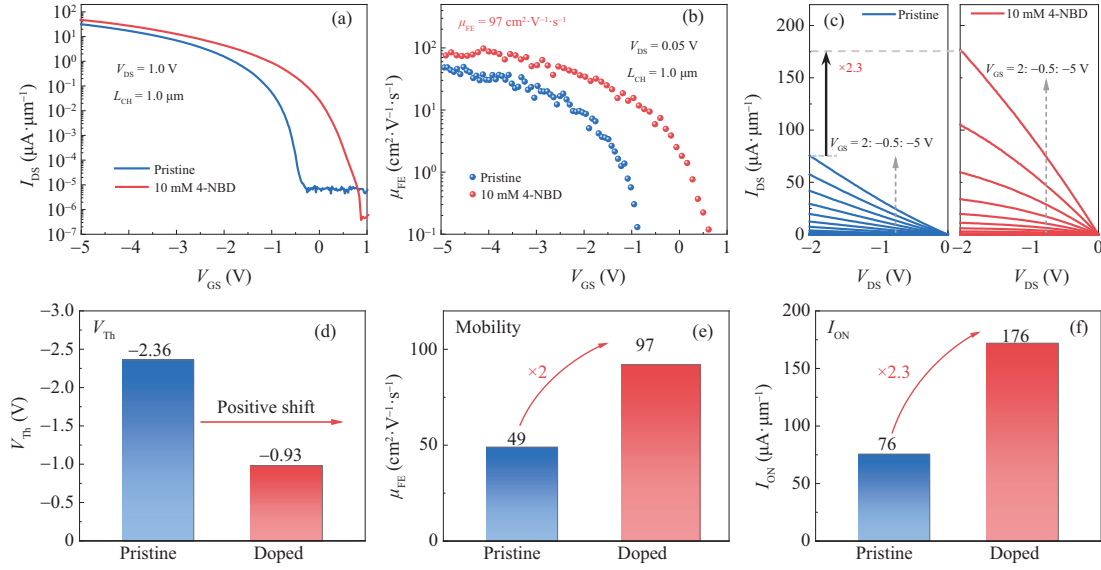
**Figure 3** (a) Schematic view of 4-NBD channel doped 1L-WSe<sub>2</sub> transistor structure and chemical structure of 4-NBD; (b) band alignment of WSe<sub>2</sub> and 4-NBD p-dopant; (c) SEM images of pristine (left) and 4-NBD doped (right) WSe<sub>2</sub> (scale bar, 15  $\mu$ m); (d) Raman spectra of 4-NBD doped and pristine WSe<sub>2</sub>; high-resolution XPS spectra of Se 3d (e) and F 1s (f) for pristine and 4-NBD doped WSe<sub>2</sub>.

film, all serving as the basis for achieving high mobility devices.

The subsequent molecular doping was performed on the back-gated WSe<sub>2</sub> devices, employing the selected 4-NBD molecular structure as illustrated in Figure 3(a). The fabricated WSe<sub>2</sub> FETs were immersed in an acetone solution of 4-NBD with concentrations of 10 mM and kept in solution for 10 min at room temperature without heating, following rinse in the isopropanol to wash off the supererogatory dopants. Figure 3(b) displays the energy levels of WSe<sub>2</sub> and 4-NBD [26]. The highest occupied molecular orbital energy of 4-NBD is lower than the valence band edge of WSe<sub>2</sub>, which induces electrons from WSe<sub>2</sub> to 4-NBD, resulting in the p-doping in WSe<sub>2</sub>. As depicted in Figure 3(c), SEM images displayed no discernible damage or organic residues during the WSe<sub>2</sub> doping process.

Raman spectroscopy and XPS were performed to further investigate the impact of surface charge transfer effect in 4-NBD doped WSe<sub>2</sub>. Figure 3(d) shows the Raman spectrum (using a laser with the wavelength of 514 nm) of the pristine WSe<sub>2</sub> (blue curve), exhibiting the E<sub>2g</sub><sup>1</sup> peak at 248.69 cm<sup>-1</sup> and the A<sub>2g</sub> peak at 260.53 cm<sup>-1</sup>. A comparison with the spectrum of doped WSe<sub>2</sub> (red curve) revealed both peaks significantly shifted towards higher wavenumbers, indicating decreased electron-phonon scattering due to p-doping. The p-doping effect is also confirmed by XPS. The peak shape of the Se 3d core level shows no apparent changes, suggesting no chemical structure alterations or damage in the WSe<sub>2</sub> channel [26]. While the comparison of Se 3d core level doublet (3d<sub>5/2</sub> and 3d<sub>3/2</sub>) of pristine WSe<sub>2</sub> and 4-NBD doped WSe<sub>2</sub> in Figure 3(e) shows a 0.51 eV shift towards lower binding energy in the doped WSe<sub>2</sub>. This shift is attributed to band bending in WSe<sub>2</sub> due to charge transfer from WSe<sub>2</sub> to 4-NBD, consistent with the Fermi level moving closer to the valence band [26]. Furthermore, the appearance of F 1s core level in doped WSe<sub>2</sub> indicates effective doping of WSe<sub>2</sub>, as shown in Figure 3(f).

The direct comparison of the electrical performance of the devices demonstrates the effectiveness of the doping effect. Figure 4(a) illustrates the transfer characteristics for pristine and doped monolayer WSe<sub>2</sub> transistors ( $V_{DS} = -1$  V) measured under a vacuum environment of below 10<sup>-5</sup> torr at room temperature. The pristine WSe<sub>2</sub> transistor exhibits p-type characteristics with an on/off ratio of >10<sup>6</sup> and an on-state current of 76  $\mu$ A  $\cdot$   $\mu$ m<sup>-1</sup> at  $V_{DS} = -2$  V. The pristine WSe<sub>2</sub> transistor did not exhibit n-type behavior under positive gate bias. The intrinsic high current of WSe<sub>2</sub> is a result of transfer-free processes, which mitigate the damage to WSe<sub>2</sub>. After the introduction of 10 mM 4-NBD doping, the doped device shows a significant positive shift in threshold voltage while improving both the on-state and off-state current, exhibiting a high on/off ratio exceeding 10<sup>8</sup>, over an order of magnitude enhancement.



**Figure 4** (a) Comparison of transfer characteristics of pristine (blue curve) and 10 mM 4-NBD doped (red curve) WSe<sub>2</sub> transistors with channel length of 1  $\mu$ m and channel width of 1  $\mu$ m at  $V_{DS} = -1$  V; (b) field-effect mobility extraction of WSe<sub>2</sub> transistors at  $V_{DS} = -0.05$  V; (c) output characteristics comparison of WSe<sub>2</sub> transistors with channel length of 1  $\mu$ m (for the pristine WSe<sub>2</sub> FET, the range spans from  $V_{GS} = 1$  to  $-5$  V in steps of  $-0.5$  V); (d) histograms of the threshold voltage shift from pristine to 10 mM 4-NBD doped WSe<sub>2</sub> FETs at  $V_{DS} = -0.05$  V; histograms showing the optimization for 10 mM 4-NBD doped WSe<sub>2</sub> FETs of mobility ( $V_{DS} = -0.05$  V) (e) and  $I_{ON}$  ( $V_{GS} = -5$  V,  $V_{DS} = -2$  V) (f), respectively.

Figure 4(b) presents the extracted field-effect hole mobility under low electric field conditions ( $V_{DS} = -0.05$  V). The effective field-effect carrier mobility ( $\mu_{FE}$ ) was calculated by using

$$\mu = \frac{G_m L}{C_{ox} V_{DS} W}, \quad (1)$$

where  $G_m = dI_{DS}/dV_{GS}$  as the transconductance obtained from the transfer curve at  $V_{DS} = -0.05$  V; the  $C_{ox}$  is the capacitance per unit of the 10 nm thick SiO<sub>2</sub>, the value of  $C_{ox}$  is 0.345  $\mu\text{F cm}^{-2}$ ;  $V_{DS}$  is the applied voltage of drain;  $L$  and  $W$  are the effective length and width of the channel. After the chemical doping process, the maximum carrier mobilities achieved by the 10 mM 4-NBD doped transistor is 97  $\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ , which is over 98% higher than the pristine mobility of 49  $\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ . Notably, the 10 mM 4-NBD doping significantly increases the hole mobility of WSe<sub>2</sub>. The chemical doping increases the carrier concentration and reduces the influence of CVD-WSe<sub>2</sub> defects. Furthermore, the p-type doping reduces the contact resistance, thereby increasing the hole mobility [20, 26, 27]. Output characteristics of the transistors are shown in Figure 4(c); the blue and red curves are pristine and 10 mM 4-NBD doped transistors, respectively. The output drain current of the doped 1  $\mu$ m channel length and 1  $\mu$ m channel width reaches 176  $\mu\text{A} \cdot \mu\text{m}^{-1}$  at a drain bias of  $-2$  V and gate voltage of  $-5$  V, marking a 2.3-fold increase compared to the pristine WSe<sub>2</sub> transistors under the same bias voltage, far exceeding the most of previous studies such as NO<sub>x</sub> doping [23], 2D contact [28], or novel metal contact for monolayer CVD p-type WSe<sub>2</sub> transistors [29–31]. Furthermore, the contact resistance is reduced from 8.64 to 5.86  $\text{k}\Omega \cdot \mu\text{m}$  under the same carrier density after doping treatment by transmission line method (TLM), indicating an improvement of the metal-semiconductor interface. Figures 4(d)–(f) present a quantitative comparison of key parameters between the pristine WSe<sub>2</sub> field-effect transistor devices and 4-NBD-doped WSe<sub>2</sub> field-effect transistor devices, visually illustrating the performance enhancement brought about by doping. Figure 4(d) shows the positive shift from  $-2.36$  to  $-0.93$  V of threshold voltage at  $V_{DS} = -1$  V due to the p-type doping of WSe<sub>2</sub>. The effective field-effect carrier mobility and  $I_{ON}$  mark 2- and 2.3-times increase for 10 mM doped WSe<sub>2</sub> FETs, respectively, showing a significant improvement as shown in Figures 4(e) and (f). Advanced logic devices demand high on/off ratios and high mobility. The 10 mM 4-NBD doped WSe<sub>2</sub> device exhibits a high on/off ratio of  $10^8$  and a high hole mobility of 97  $\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ , achieving high on-state current and extremely low off-state current (below  $5 \times 10^{-7}$   $\mu\text{A} \cdot \mu\text{m}^{-1}$ ). Furthermore, Table 1 shows the comparison of the doped WSe<sub>2</sub> performance in this study with previous studies [26, 32], demonstrating the superiority of device mobility and on/off ratio in this study.



**Table 1** Comparison of the doped WSe<sub>2</sub> performance

Ref.	Channel	Mobility ( $\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ )	On/off ratio	$I_{\text{ON}}$ ( $\mu\text{A} \cdot \mu\text{m}^{-1}$ )	EOT <sup>a)</sup> (nm)
[32]	Solution-processed WSe <sub>2</sub>	0.1	$4 \times 10^4$	—	—
[26]	CVD-WSe <sub>2</sub> 1L	82	$1.0 \times 10^7$	—	90
This work	CVD-WSe <sub>2</sub> 1L	97	$1.2 \times 10^8$	176	10

a) EOT denotes equivalent oxide thickness.

### 3 Conclusion

In conclusion, we successfully demonstrated an effective chemical doping approach for LPCVD direct-grown monolayer WSe<sub>2</sub> FETs by employing 4-NBD p-dopants. The effectiveness of surface charge transfer was confirmed through comprehensive Raman and XPS spectral analyses. The field-effect hole mobility of WSe<sub>2</sub> transistors exhibited a significant increase from the initial 49 to 97  $\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ , representing an impressive 98% enhancement under the 10 mM doping concentration. Importantly, unlike the degradation in off-current reported in previous literature due to chemical doping, this study achieved improvements in off-state current while maintaining a high on/off ratio of up to  $10^8$ . Moreover, a high drive current exceeding 176  $\mu\text{A} \cdot \mu\text{m}^{-1}$  was achieved on the WSe<sub>2</sub> transistors with a 1  $\mu\text{m}$  channel length. This strategy offers a straightforward and effective means to boost the PFET electrical performance of 2D WSe<sub>2</sub>, showcasing great potential in fabricating 2D CMOS circuit applications.

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