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Chemically tuned p- and n-type WSe₂ monolayers with higher carrier mobility for advanced electronics

From the advent of atomically thin transition metal dichalcogenides (TMDCs), such as MoS₂, WS₂, and WSe₂, they have been attracting great interest due to their promising and unique properties [1]. Especially, some TMDCs are known to show semiconducting properties with tunable band gap depending on the combination of transition metal and chalcogen atoms. The superior electrical and optical properties of these semiconducting TMDCs have been already reported by demonstrating various devices, such as transistors, logic gates, and optical sensors [2]. Although both p-type and n-type materials are required for the fabrication of p-n junction and logic gate circuits, which are the important working units in advanced electronics, most of semiconducting TMDCs exhibit n-type or ambipolar behavior. Therefore, the lack of p-type TMDCs is retarding the progress of device applications of TMDC materials.

To control over the electrical polarity of semiconducting TMDCs, several approaches have been reported. For example, partial substitution of Mo (or W) atoms with other metal atoms was proved to have a doping effect. However, the device showed heavy degenerated property with a weak gate voltage dependence [3]. Also, deposition of metal nanoparticles was reported to give p-doping effect on WSe₂ but the nanoparticles on WSe₂ flake were found to quench the photoluminescence (PL), which is a critical disadvantage for optical application [4]. As an alternative, chemical doping can be a promising method [5]. Chemical doping of TMDCs using specific molecules was found to have an efficient doping effect without degradation of optical property. Although the usefulness of chemical doping has been demonstrated, the controlled p- and n-type transport with a single TMDC has not been reported so far.



Figure 1: Chemical structures of 4-NBD (a) and DETA (b) molecules. Transfer curves of pristine and doped WSe₂ (c) and the maximum carrier mobilities (d).

In this work, we demonstrate the doping of chemical vapor deposition (CVD) grown-WSe₂ for selective conversion from ambipolar to p- or n-type semiconductors. This was done by using 4-nitrobenzenediazonium tetrafluoroborate (4-NBD) and diethylene triamine (DETA) molecules as p- or n-type dopants, respectively (see Fig. 1a,b). After each doping process, WSe₂ showed clear p- or n-type transfer curves (Fig. 1c). In addition, the carrier mobility showed significant increase up to $10^3 \sim 10^4$ times after the doping, with maximum values of 82 cm²V⁻¹s⁻¹ and 25 cm²V⁻¹s⁻¹ for holes and electrons, respectively (Fig. 1d). Various spectroscopic measurements were performed by using PL, Raman and X-ray photoelectron spectroscopy (XPS) to understand the chemical doping effects. In addition, density functional theory (DFT) calculation was performed for further understanding of the charge transfer between WSe₂ and the dopant molecules.

To demonstrate the device application of our chemically doped WSe₂, field-effect transistors (FETs) with pand n-type WSe₂ channels were integrated to fabricate a complementary metal-oxide-semiconductor (CMOS) inverter, as illustrated in Fig. 2a. The device showed sharp inverting between input and output voltage (Fig. 2b), with extremely low power consumption (~170 pW) and voltage gain up to ~10. Finally, we fabricated p-n junction within single WSe₂ grain. The spatially controlled p- and n-doping of single WSe₂ grain could be realized by using lithography technique. The optical image of the final device is shown in Fig. 2c. The spatially controlled p-n junction showed clear rectification behavior (Fig. 2d), which is a strong evidence of p-n junction formation. The device also exhibited optical response under laser illumination, suggesting potential application to optoelectronics (Fig. 2e). We believe our chemical doping strategy offers a versatile approach to control the property of TMDC materials for various practical applications [6].



Figure 2: Schematic illustration of a CMOS inverter (a) and the measured V_{out} - V_{in} relationship (b). Optical image of a p-n junction device (c) and the obtained output curves (d) and optical response (e).

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