Fabrication of Transition Metal Dichalcogenide Photovoltaic Devices Using Surface-Charge Transfer (SCT) Doping Mechanism

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Emerging atomically layered transition metal dichalcogenides (TMDCs, *e.g.*, WSe_2, WS_2, and MoS_2) exhibit ultra-high light absorption coefficients over a broad range of wavelengths (NUV-vis-IR) and have been envisioned to enable next-generation ultrathin flexible photovoltaic (PV) devices with excellent performance and low cost.[1-3] In order to fully leverage the superior photonic properties of TMDCs for practical photovoltaic applications, new doping (or charge transfer) techniques are needed for fabricating TMDC-based PV devices with reliable built-in junctions that can efficiently separate photo-generated e-h pairs. Our group previously reported MoS_2 PV devices made by using plasma-assisted implantation.[4] However, this plasma doping process significantly degrades the transport properties of TMDC materials. Chen *et al*. recently reported the fabrication of TMDC PV devices based on WSe_2/MoS_2 heterojunctions that can serve as p-n diode junctions for facilitating the charge transfer.[5] However, the fabrication of such vertically-stacked TMDC bilayer heterostructures over large areas is still a challenge. Therefore, new doping (or charge transfer) methods, which do not need detrimental implantation processes and are compatible to large-area nanomanufacturing processes, are strongly demanded.

Here, we report a new upscalable doping technique capable of forming permanently stable built-in p-n junctions in pristine WSe_2 photoactive layers and resulting in excellent PV performance. This new doping method employs regular deposition processes to coat WSe_2 surfaces with thin-film materials with deterministic work functions (Φ), which can induce reliable surface-charge transfer (SCT) processes at WSe_2/thin-film material interfaces and result in p-n junctions with deterministic band alignments. Such a highly relevant correlation between work functions of chosen thin-film materials and resultant band alignments of p-n junctions is attributed to the 2-D nature of TMDC surfaces that have an extremely low density of interfacial charge traps and can effectively suppress the Fermi-level-pinning effect.

Fig. 1a illustrates a WSe_2 PV device consisting of a vertically stacked high-Φ electrode/few-layer WSe_2/low-Φ electrode/ITO structure. **Fig. 1b** shows the schematic band diagram of this WSe_2 PV device. At the thermodynamically equilibrium state, the SCT processes at hig-Φ electrode/WSe_2 and WSe_2/low-Φ electrode interfaces can induce p- and n-doping in the WSe_2 photoactive region, respectively, and form a p-n junction. **Fig. 2** displays photovoltaic characteristics of a set of our WSe_2 PV devices with (a) no low-Φ electrode; (b) Φ_L = 4.26 eV; (c) Φ_L = 4.30 eV; (d) Φ_L = 3.63 eV. All devices have the same high-Φ electrode (*i.e.*, Au, Φ_H = 5.1 eV). The measurement was under 532 nm laser illumination (P = 283 mW/cm^2). Fig. 2 shows that Φ_L values can greatly affect the performance of WSe_2 PV devices. The smallest Φ_L value (*i.e.*, 3.63 eV) results in the best PV performance, in terms of V_{oc}=0.37V, I_{sc}=1.2μA (J_{sc}=105 mA/cm^2), FF=0.50, and PCE=6.9% under 532 nm laser illumination (P = 283 mW/cm^2).

This work provides important scientific insights for enhancing the PV responses of TMDCs. The presented SCT-based doping processes can potentially enable the large-manufacturing of TMDC-based ultrathin-film PV cells.

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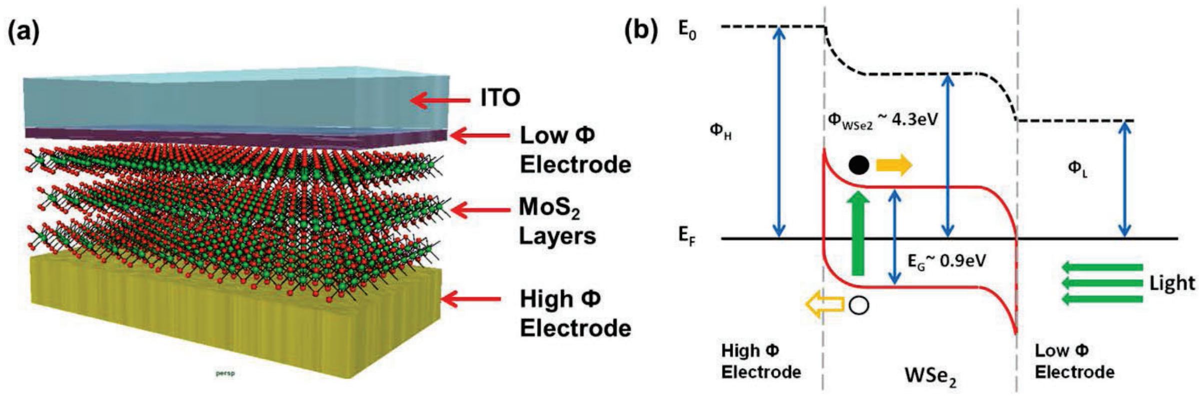

Fig. 1 (a) Illustration of a WSe_2 photovoltaic (PV) device consisting of a vertically stacked high Φ electrode/WSe_2/low Φ electrode/ITO structure; (b) Schematic band diagram of this PV device.

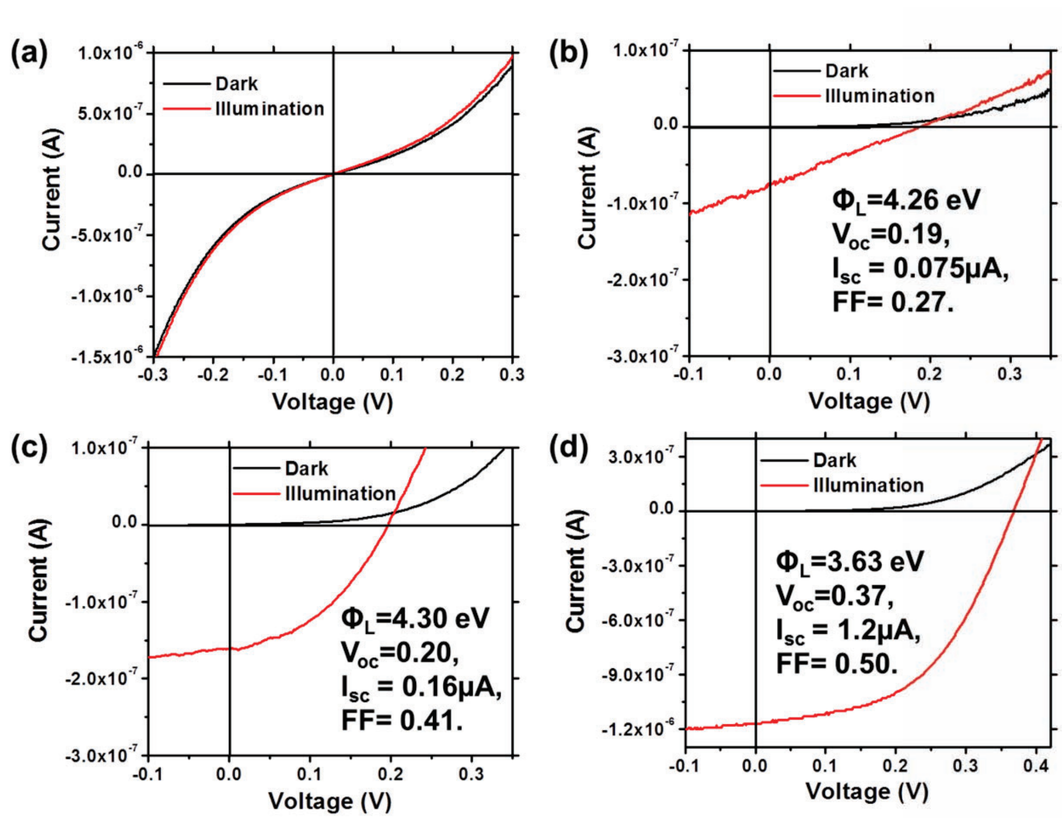

Fig. 2 Photovoltaic characteristics of WSe_2 PV devices with (a) no low-Φ electrode; (b) Φ_L = 4.26 eV; (c) Φ_L = 4.30 eV; (d) Φ_L = 3.63 eV. All devices have a high-Φ electrode (*i.e.*, Au, Φ_H = 5.1 eV). The measurement was under 532 nm laser illumination (P = 283 mW/cm^2).

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