

Fabrication of Vertically-Stacked MoS₂-Based Thin-Film Photovoltaic Devices

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2D layered transition metal dichalcogenides (LTMDs) have been envisioned for making new ultra-thin-film photovoltaic (PV) devices with a unique combination of excellent photovoltaic performance, superior flexibility, long life time, and low manufacturing cost.^[1, 2] Especially, a single semiconducting LTMD (*e.g.*, WSe₂, WS₂, and MoS₂) layer (~0.5 nm thick) can absorb as much sunlight as 50 nm of Si (or 12 nm of GaAs) and generate currents as high as 4.5mA/cm². Therefore, 2D LTMD films hold a significant potential to be used for making ultrathin flexible photovoltaic (PV) cells with 1-3 orders of magnitude higher power densities than the best existing thin-film solar cells.^[1-2] In spite of such an optimistic anticipation, the current LTMD-based PV cells still exhibit relatively poor PV performance,^[2-5] which is attributed to the lack of proper fabrication and doping processes for tailoring MoS₂ layers into optimized PV device architectures.

To address this challenge, we developed a fabrication route combining transfer-printing methods^[6] and plasma-assisted doping processes,^[7] which can create PV devices consisting of vertically stacked Au/p-type MoS₂/n-type MoS₂/ITO structures (**Fig. 1**). Such vertically stacked PV devices exhibit reasonably good (and promising further improved) PV performance, in terms of short-circuit photocurrent density (J_{sc}) up to 29 mA/cm² that is comparable to J_{sc} values of exquisite III-V semiconductor PV cells, power-conversion efficiencies (PCEs) up to 3.53% that is the highest PCE among LTMD-based thin-film PV cells (**Fig. 2**), and extrinsic quantum efficiencies (EQEs) > 40% over a broad wavelength range (300 to 800 nm) (**Fig. 3a**). In addition, such a vertically stacked ITO/plasma-doped MoS₂/metal structure can also be used for making thin-film photodetectors with a high responsivity (> 100 mA/W at zero bias for $\lambda = 300$ to 800 nm) (**Fig. 3b**) and a high light/dark current ratio ($10^2 - 10^4$ at negative biases) (**Fig. 3c**).

This work provided important scientific insights and materials processing methods for creating new thin-film photovoltaic devices and other relevant optoelectronic components based on emerging 2D materials.

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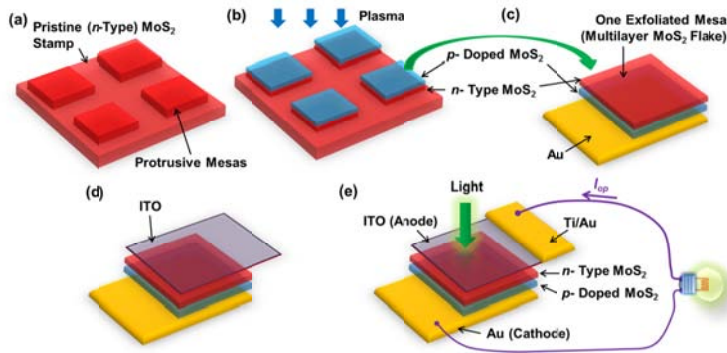


Fig. 1 Fabrication of PV devices consisting of vertically stacked Au/p-type MoS₂/n-type MoS₂/ITO structures by using transfer-printing in combination with plasma doping.

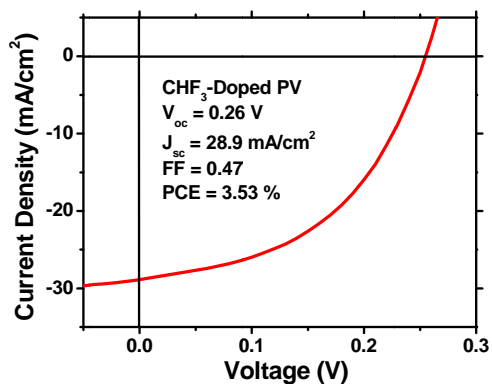


Fig. 2 J-V characteristic curve, measured under illumination of AM1.5G simulated sunlight (power density, 100 mW/cm²), of a plasma-doped MoS₂ PV device.

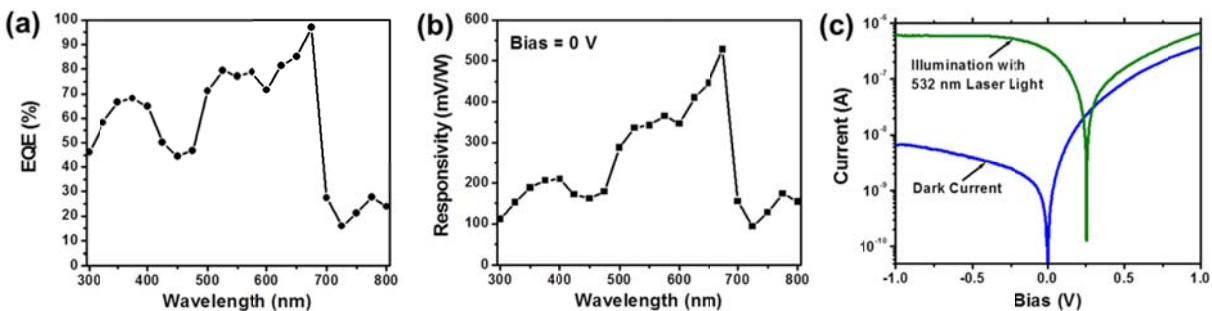


Fig. 3 EQE (a) and responsivity (b) values of a PV device with MoS₂ thickness of 120 nm measured at wavelengths $\lambda = 300$ to 800 nm. (c) shows the I-V characteristic curves under dark (blue curve) and illumination (532 nm laser light with excitation power of 203 mW/cm²) (green curve) conditions. This PV diode exhibits high light/dark current ratios (10^2 to 10^4) at reverse biases, suitable for photodetector applications.