Repetitive Nanoprinting Technique for Producing Vertically Stacked Transition Metal Dichalcogenide Heterostructure Arrays and Photo-Response Devices

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Semiconducting transition metal dichalcogenides (TMDCs, *e.g.*, WSe_2, WS_2, and MoS_2) are attractive materials for making the next generation ultra-thin optoelectronic devices due to their superior electronic, photonic, and mechanical properties.[1-2] The generation of reliable built-in potentials in TMDC layers is essential for fabricating new working photo-response devices based on semiconducting TMDCs. Currently, it is still a challenge to generate reliable built-in potentials in such layered materials *via* doping methods because most available doping methods cause detrimental damage to layered material structures. Recently, it has been preliminarily demonstrated that mechanically stacked (or epitaxially grown) WSe_2/MoS_2 heterostructures can generate sizable built-in potentials for facilitating the separation of photo-generated e-h pairs (or excitons) and resulting in high photo-responsivity without inducing any detrimental damage to TMDC structures.[3] However, we still lack a top-down nanofabrication approach capable of producing large arrays of such attractive TMDC heterostructures.

Here, we present an upscalable nanofabrication technique capable of producing uniform multilayer TMDC heterostructure arrays into device sites. Specifically, using this technique, we have demonstrated the fabrication of uniform multilayer WSe_2/MoS_2 heterostructure arrays. These heterostructures can be used for making photo-response devices.

Fig. 1 schematically illustrates the presented nanofabrication technique for producing TMDC heterostructure arrays. More specifically, this technique involves two critical sub-processes. Firstly, our previously developed nanoimprint-assisted shear exfoliation (NASE) technique is used to exfoliate prepatterned TMDC features with uniform thicknesses from a bulk TMDC stamp onto a PDMS transfer stamp (Steps 1-4).[4] These TMDC features are subsequently transfer-printed into ordered device sites with adhesion layers (*e.g.*, metallic electrodes) on the substrate (Steps 5, 6). The repetitive operation of this process for printing different TMDC materials can generate vertically stacked TMDC heterostructure arrays. **Fig. 2** displays the optical micrographs (OMs) of (a) uniform WSe_2 mesa arrays exfoliated from a bulk WSe_2 stamp onto a PDMS transfer stamp, (b) the same WSe_2 mesa arrays transfer-printed onto prepatterned Au contacts on a SiO_2 substrate, and (c) MoS_2 mesa arrays subsequently transfer-printed on top of WSe_2 structures, forming uniform vertically-stacked multilayer WSe_2/MoS_2 heterostructure arrays. Such heterostructures have been used for making photo-response devices. **Fig. 3** (a) shows the OM of a representative photo-response device consisting of vertically stacked $Au/WSe_2/MoS_2/ITO$ layers. **Figs. 3b, 3c** exhibits the I-V characteristics of this device measured under dark condition and 532 nm laser illumination, respectively. Under the laser illumination, the device exhibits large photocurrents at negative biases, and also an observable photovoltaic effect (V_{oc}=0.35 V, J_{sc}=27.8 mA/cm^2, FF=0.17 and PCE= 0.6%).

This work advanced the critical nanofabrication technology to leverage the superior optoelectronic properties of TMDCs for practical photonic device applications.

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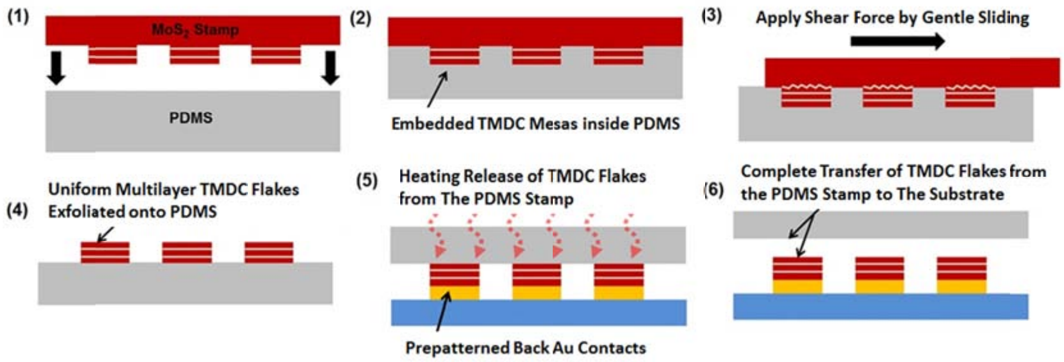

Fig. 1 Schematic steps for printing uniform multilayer TMDC arrays into device sites on the substrate: The whole process consists of two critical sub-processes – Steps (1)-(4): nanoimprint-assisted shear exfoliation (NASE) of pre-patterned TMDC mesas from a bulk TMDC stamp onto a PDMS transfer stamp; Steps (5) to (6): transfer-printing of TMDC mesas from the PDMS stamp onto the device sites on the final substrate. The repetitive application of this process for printing different TMDC flakes can generate vertically stacked TMDC heterostructures.

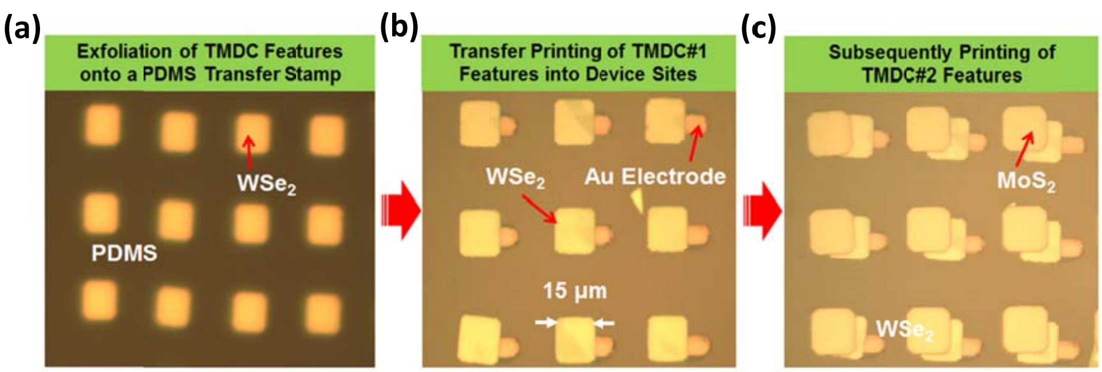

Fig. 2 Optical micrographs of (a) NASE-produced WSe_2 mesa arrays on a PDMS transfer stamp, (b) WSe_2 mesa arrays transfer-printed onto the prepatterned Au contacts on the final device substrate, and (c) MoS_2 mesas subsequently printed on top of WSe_2 mesas, resulting in vertically stacked WSe_2/MoS_2 heterostructure arrays.

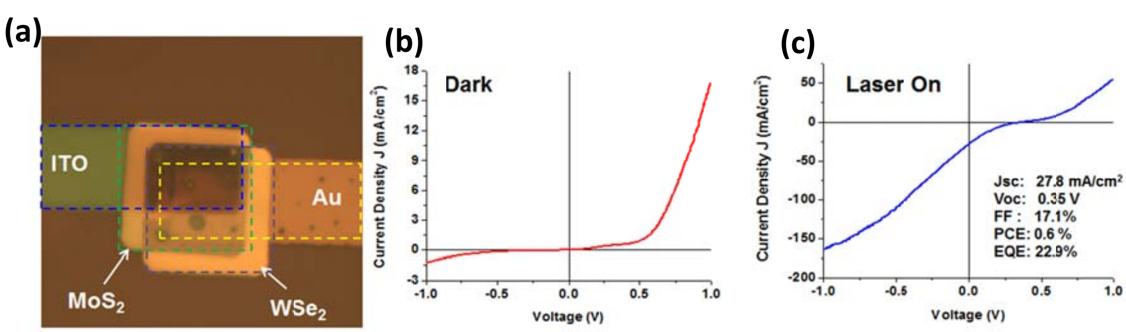

Fig. 3 (a) Optical micrograph of a vertically stacked $Au/WSe_2/MoS_2/ITO$ photo-response device. I-V characteristics of this device measured (b) under no illumination and (c) under 532 nm laser illumination ($P = 200\ mW/cm^2$).

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