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Dear Editor,

We would like to submit enclosed manuscript entitled "Maskless micro/nanopatterning and bipolar electrical-rectification of MoS_2 flakes through femtosecond laser direct writing", which we wish to be considered for publication in "*ACS Applied Materials & Interfaces*". No conflict of interest exits in the submission of this manuscript, and manuscript is approved by all authors for publication. I would like to declare on behalf of my co-authors that the work described is original research that has not been published previously, and not under consideration for publication elsewhere, in whole or in part. All the authors listed have approved the manuscript that is enclosed.

Molybdenum disulfide (MoS_2) is a typical member of the transition metal dichalcogenides family, a kind of graphene-like two-dimensional materials which are one of the most popular materials recently. MoS_2 micro/nanostructures are desirable for tuning electronic properties, developing required functionality, or improving existing performance of MoS_2 devices. Hence, it is crucial to conduct research on MoS_2 patterning with ordered micro/nanostructures, controlled structure-size, and device application having new features.

This work presents a useful method to flexibly microprocess multilayer MoS_2 flakes through femtosecond laser pulse direct writing, which can directly fabricate regular MoS_2 nanoribbon arrays with ribbon widths of 179, 152, 116, 98, and 77 nm, and arbitrarily pattern MoS_2 flakes to form micro/nanostructures such as single nanoribbon, labyrinth array, and cross structure. This method is mask-free and simple, and has high flexibility, strong controllability, and high precision. Moreover, numerous oxygen molecules are chemically and physically bonded to laser-processed MoS_2, attributed to roughness defect-sites and edges of micro/nanostructures that contain numerous unsaturated edge-sites and highly active centres. In addition, electrical tests of the field effect transistor fabricated from prepared MoS_2 nanoribbon arrays reveal new interesting features: output and transfer characteristics exhibit strong rectification (not going through zero and bipolar conduction) of drain−source current, which is supposedly attributed to the coordinate structures and p-type chemical doping of oxygen molecules on MoS_2 nanoribbon arrays. This work demonstrates the ability of femtosecond laser pulses to directly induce micro/nanostructures, property changes, and new device-properties of two-dimension materials, which may future enable new device applications.

The manuscript is considered appropriate for *"ACS Applied Materials & Interfaces"* for the following reasons: 1) *MoS_2 micro/nanostructures* are fabricated through femtosecond laser pulse direct writing, and the used method is mask-free and simple, and has high flexibility, strong controllability, and high precision for 2D material microprocess; 2) oxygen molecules are chemically/physically bonded to laser-processed MoS_2 with numerous unsaturated edge-sites and highly active centres, resulting in *chemical surface modification of MoS_2*; 3) *strong electrical-rectification of MoS_2* with physical nanostructures and chemical modification, similar to p-n type transfer conduction, which may future enable new electronic device applications.

Associated Supporting Information including XPS spectra and electrical test of MoS_2 FETs with moderate surface modification is also enclosed for review and publishing.

The following is a list of five possible reviewers for your consideration:

- Minlin Zhong E-mail: zhml@tsinghua.edu.cn
- Minghui Hong E-mail: elehmh@nus.edu.sg
- Emmanuel Stratakis E-mail: stratak@iesl.forth.gr
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We deeply appreciate your consideration of our manuscript, and we look forward to receiving comments from the reviewers. If you have any queries, please don't hesitate to contact me at lixin02@bit.edu.cn.

Thank you and best regards,

Yours sincerely,

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