Maskless Micro/Nanopatterning and Bipolar Electrical Rectification of MoS2 Flakes Through Femtosecond Laser Direct Writing Processing

*Pei Zuo,**† Lan Jiang,\*,† Xin Li, † Mengyao Tian, † Peng Ran, † Bo Li, † Yongfeng Lu,§*

†Laser Micro/Nano Fabrication Laboratory, School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, P.R. China, §Department of Electrical and Computer Engineering, University of Nebraska-Lincoln, Lincoln, NE 68588-0511, USA

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**ABSTRACT**: MoS2 micro/nanostructures are desirable for tuning electronic properties, developing the required functionality, and improving existing performance levels of a multilayer MoS2 device. This paper presents a useful method to flexibly microprocess multilayer MoS2 flakes through femtosecond laser pulse direct writing. This method can be used to directly fabricate regular MoS2 nanoribbon arrays with ribbon widths of 179, 152, 116, 98, 77 nm, and arbitrarily pattern MoS2 flakes to form micro/nanostructures such as single nanoribbons, labyrinth arrays, and cross structures. 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 MoS2. This bonding can be attributed to the roughness of defect-sites and edges of micro/nanostructures that contain numerous unsaturated edge sites and highly active centers. In addition, electrical tests of the field-effect transistor fabricated from the femtosecond laser pulse direct written MoS2 nanoribbon arrays revealed the following characteristics: output and transfer characteristics exhibited strong rectification (not going through zero and bipolar conduction) of drain–source current. This rectification is supposedly attributed to the coordinate structures and p-type chemical doping of oxygen molecules on MoS2 nanoribbon arrays. This study demonstrated the ability of femtosecond laser pulses to directly induce micro/nanostructures of, property changes in, and novel device properties of two-dimensional materials, which may enable new device applications in the future.

**INTRODUCTION**

Transition metal dichalcogenides (TMDCs), comprising representative layered materials such as graphene, have been widely studied as a family of new-type semiconducting materials with extraordinary mechanical, electrical, and optical properties, and are promising for extensive applications.[1–6](#_ENREF_1" \o "Chhowalla, 2016 #3) Molybdenum disulfide (MoS2), a representative material belonging to the TMDC group, has a lattice structure such that a layer of molybdenum atoms is sandwiched between two layers of sulfur atoms.[7, 8](#_ENREF_7" \o "Behura, 2015 #48) Unlike graphene, which possesses zero band gap, MoS2 is a semiconductor with a sizable energy band gap:[5](#_ENREF_5" \o "Mak, 2013 #14), [9](#_ENREF_9" \o "Lu, 2016 #49) Bulk MoS2 possesses an indirect energy gap of approximately 1.2 eV and monolayer MoS2 possesses a direct band gap of approximately 1.8 eV.[3](#_ENREF_3" \o "Nam, 2013 #2) Therefore, MoS2 can serve as a substitute of graphene, and it has excellent potential in semiconductor-related applications of layered materials such as thin-film transistors,[1](#_ENREF_1" \o "Chhowalla, 2016 #3), [10](#_ENREF_10" \o "Cho, 2018 #6) integrated circuits,[11, 12](#_ENREF_11" \o "Cheng, 2014 #7) complementary inverters,[13](#_ENREF_13" \o "Yoo, 2018 #8) photodetectors,[14](#_ENREF_14" \o "Pak, 2018 #10) light-emitting diodes,[15, 16](#_ENREF_15" \o "Woo, 2018 #12) photovoltaics,[17, 18](#_ENREF_17" \o "Zhang, 2018 #14) superconductors,[19, 20](#_ENREF_19" \o "Chen, 2018 #16) and chemical or biological sensors.[21, 22](#_ENREF_21" \o "Kumar, 2017 #18)

To achieve desirable electronic properties and required functionality and improve the existing performance of MoS2 devices, a microfabrication process is usually required to fabricate large arrays of orderly arranged MoS2 micro/nanostructures, commonly involving patterning and etching.[3](#_ENREF_3" \o "Nam, 2013 #2), [23](#_ENREF_23" \o "Zhao, 2016 #1) Hence, research on MoS2 patterning with ordered micro/nanostructures, controlled structure size, and device application with new features is essential. Several approaches to patterning MoS2 materials or fabricating micro/nanostructures of MoS2 deposited on substrates have been attempted, including tape exfoliation,[24, 25](#_ENREF_24" \o "Huang, 2017 #20) chemical vapor deposition (CVD),[26, 27](#_ENREF_26" \o "Cong, 2018 #21) thermal decomposition of thiosalts,[28](#_ENREF_28" \o "Liu, 2012 #23) van der Waals epitaxial growth,[29, 30](#_ENREF_29" \o "Pak, 2017 #25) patterned MoO3 sulfurization,[31, 32](#_ENREF_31" \o "Xue, 2016 #27) CVD in plasma-treated areas,[33, 34](#_ENREF_33" \o "Chen, 2016 #30) block copolymer lithography,[35](#_ENREF_35" \o "Han, 2017 #28) reactive ion etching (RIE) and deposition in plasma-treated areas,[3](#_ENREF_3" \o "Nam, 2013 #2) lithography and stamping,[23](#_ENREF_23" \o "Zhao, 2016 #1) lithography and RIE,[36, 37](#_ENREF_36" \o "Jung, 2014 #31) and continue wave (CW) laser direct processing.[38, 39](#_ENREF_38" \o "Cao, 2013 #33) Among them, CVD in plasma-treated areas, block copolymer lithography, lithography and deposition in plasma-treated areas, lithography and stamping, lithography and RIE, and CW laser direct processing can realize control of the location, shape, and size of MoS2 micro/nanostructures, whereas in lithography, plasma surface treatment, and RIE, chemical organics (such as photoresist) and inorganic or organic masks are used combined processes are required, and the integral process is relatively complex. CW laser direct processing is a one-step process and is simple, no special atmosphere system and mask are needed, cost is relatively low, and flexibility and controllability are high. However, CW laser processing is a thermal process, and the strong thermal effect causes thermal oxidization of the materialsand leads to a large recasting layer; hence, processing precision is insufficient. Femtosecond (ultrafast) laser pulse direct processing is another laser processing technology. Femtosecond laser pulses have ultrashort pulse width, ultrahigh power density, and a nonlinear nonequilibrium processing feature, and they can realize multiphoton absorption and nonthermal effects. Moreover, in addition to all the advantages of CW laser processing, femtosecond laser direct processing possesses high precision.[40](#_ENREF_40" \o "Jiang, 2018 #65)

In this study, we proposed a useful method of utilizing femtosecond laser pulse direct writing to modify multilayer MoS2 flakes, directly fabricate regular MoS2 nanoribbon arrays with different ribbon widths, and arbitrarily pattern MoS2 flakes to form different MoS2 micro/nanostructures. Moreover, the laser-fabricated MoS2 structures were chemically and physically bonded with numerous oxygen molecules in the air. This bonding can be attributed to the roughness of defect sites and long edges of the nanoribbons that may contain numerous unsaturated edge sites and highly active centers. The field-effect transistor (FET) fabricated from the femtosecond laser pulse direct written MoS2 nanoribbon arrays was tested, and the output and transfer characteristics of the FET exhibited strong rectification (not going through zero and bipolar conduction) of drain–source current. This rectification is supposedly attributed to the coordinate structures and p-type chemical doping of oxygen molecules on the MoS2 nanoribbon arrays, which may cause the transition of n-type channels to p-type channels or properties similar to the pn junction. The proposed method is mask-free and simple and has high flexibility, strong controllability, and high precision. Moreover, the method indicated the ability of femtosecond laser pulses to directly induce micro/nanostructures/patterns of, property changes in, and new device features of two-dimensional materials.

**RESULTS AND DISCUSSION**

In this study, multilayer MoS2 flakes were mechanically exfoliated from a natural crystal by using an adhesive tape and deposited on 300-nm SiO2/Si substrates. The thickness of MoS2 flakes selected for experiments was several to dozens of nanometers. The schematic of our method of fabricating MoS2 nanoribbons/patterns is shown in Figure 1a. The method relies on the formation and regular removal of nanostructures/material through femtosecond laser pulse irradiation. When a femtosecond laser pulse beam is focused and irradiated on the material surface, surface plasmons (SPs) can be induced on the material surface. The interaction of SPs with the incident laser field can lead to the formation of initial grating structures on the material. These initial grating structures can assist the coupling of the SPs and incident laser field, leading to further formation of final grating structures with deep gaps.[41](#_ENREF_41" \o "Huang, 2009 #35) The depth of the gaps of the laser-induced grating structures is influenced by the number and energy of ultrafast laser pulses on unit area of material and can be on a micrometer scale.[40](#_ENREF_40" \o "Jiang, 2018 #65), [42, 43](#_ENREF_42" \o "Shuangshuang, 2011 #37) The MoS2 used in our experiment was a nanoscale thin flake. Therefore, sufficiently slowing the controlling scan speed of the laser beam (i.e., sufficiently large laser pulse number on unit area of MoS2) can control the gaps penetrating the whole MoS2 thin flake, enabling surface nanogratings to become independent nanoribbons. Figure 1b displays the optical comparison of pristine MoS2 with femtosecond laser-processed (FLP) MoS2. The optical image of the MoS2 flake showed an obvious color change after femtosecond laser irradiation, indicating a change in thickness or surface roughness. Figure 1c depicts the results of atomic force microscopy (AFM) of the MoS2 flake in Figure 1b. The result indicates a reduction in the outline height of the FLP-MoS2 flake, which suggests a reduction in the thickness of MoS2 by femtosecond laser irradiation. Figure 1d displays the high-resolution AFM image of FLP-MoS2, which indicates the formation of MoS2 grating structures.



Figure 1. Principle and preliminary characterization of this method. a) Schematic of femtosecond laser direct wiring processing to induce MoS2 nanoribbons array. b) Optical comparison of pristine MoS2 (upper part) with FLP-MoS2 (lower part). c) AFM and d) high resolution AFM of the FLP-MoS2 flake.

The physical morphology of the FLP-MoS2 micro/nanostructures was further characterized through scanning electron microscopy (SEM), as shown in Figure 2. Figure 2a–e presents regular MoS2 nanoribbon arrays with ribbon widths of approximately 179, 152, 116, 98, and 77 nm. Figure 2f displays two independent MoS2 nanoribbons with widths of 56 and 420 nm. Figure 2g and h shows MoS2 patterned structures obtained through material removal by using femtosecond laser direct writing. The patterned structures were revealed to be a labyrinth array and a cross structure, which indicated the flexible processing capability of femtosecond laser for arbitrary patterns.



Figure 2. FLP-MoS2 micro/nanostructures and patterns. a–e) SEM images of regular MoS2 nanoribbon arrays with widths of 179, 152, 116, 98, and 77 nm. f) SEM images of two independent MoS2 nanoribbons with widths of (up) 56 and (down) 420 nm. SEM images of g) MoS2 labyrinth array and h) MoS2 cross structure.

To prove the formation of MoS2 nanoribbons rather than surface structures on MoS2, energy dispersive X-ray (EDX) analysis was performed. Figure 3a–c shows the SEM images of the edge of a FLP-MoS2 flake and its S and Si EDX mapping images. The grey part denotes MoS2 and the black part denotes the substrate; the regular striped structures were induced by irradiation through femtosecond laser pulses (Figure 3a). Figure 3b depicts the clearly separated MoS2 nanoribbons, and Figure 3c shows clear gaps between these MoS2 nanoribbons. Figure 3d–f displays the SEM image of MoS2 nanoribbon arrays and their S and Si EDX mapping images, which indicates the separation of MoS2 nanoribbons, thus evidencing the formation of MoS2 nanoribbons rather than surface structures on MoS2.



Figure 3. Proof of the formation of separated MoS2 nanoribbons. a) SEM image of the edge of a FLP-MoS2 flake and its b) S and c) Si EDX mapping images. d) SEM image of MoS2 nanoribbon arrays and their e) S and f) Si EDX mapping images.

Raman and X-ray photoelectron spectroscopy (XPS) characterizations were conducted to investigate the change in chemical properties of the FLP-MoS2 flakes. Figure 4a presents the Raman spectra of the pristine MoS2 (black line) and FLP-MoS2 (red line). The absence of Raman peaks at 820 cm−1 indicated that MoO3 did not form and thus, no thermal oxidation occurred for FLP-MoS2[44](#_ENREF_44" \o "Paradisanos, 2014 #38) due to the nonthermal effect of femtosecond laser processing.[40](#_ENREF_40" \o "Jiang, 2018 #65) At 520 cm−1, the Raman peak of SiO2 was not detected on the pristine MoS2 flake. However, a Raman peak of SiO2 was detected on the FLP-MoS2 flake, which indicated that the substrate was detected on the FLP-MoS2 flake.[45, 46](#_ENREF_45" \o "Serincan, 2004 #39) Moreover, this observation proved the separation of MoS2 nanoribbons and penetrating gaps between them, which was consistent with the results in Figure 3. XPS Mo, O, and S spectra of pristine MoS2 and FLP-MoS2 are illustrated in Figure S1, Figure 4b and c, and Figure S2, respectively. Figure S1a presents the Mo 3d spectra for pristine MoS2, which exhibits three peaks at 227.2, 229.3, and 232.4 eV, respectively assigned to the S 2s orbital of divalent sulfur, Mo4+ 3d5/2 orbit of tetravalent Mo, and 3d3/2 orbitals of tetravalent Mo. However, in the Mo 3d spectra for FLP-MoS2 (Figure 4b), in addition to the three peaks observed for pristine MoS2, a new peak was observed at approximately 235 eV, which was assigned to the Mo6+ 3d3/2 orbital of hexavalent Mo. The Mo6+ in FLP-MoS2 was attributed to the Mo–O bonds formed through oxygen bonding to the unsaturated Mo bonds, defect sites, or edges of nanoribbons,[47, 48](#_ENREF_47" \o "Wei, 2014 #19) generated from the damage and material removal of MoS2 during femtosecond laser pulse writing. Figure S1b depicts the O 1s spectra of pristine MoS2, which exhibits two peaks at 532.6 and 533.4 eV, attributed to the divalent oxygen of Si–O bonds and the nonvalent oxygen of oxygen molecules physically adsorbed on the MoS2 surface (O2/MoS2), respectively.[49–51](#_ENREF_49" \o "Hollinger, 1981 #23) The divalent oxygen of Si–O bonds was derived from the 300-nm SiO2/Si substrate, and the oxygen molecules on the MoS2 surface could be attributed to the intrinsic defects and edges of the flake, which were active sites capable of physical adsorption. However, in O 1s spectra for FLP-MoS2 (Figure 4c), except the strong and dominant peak assigned to divalent oxygen of Si–O bonds as shown in pristine MoS2, it revealed stronger peak of nonvalent oxygen of O2/MoS2, indicating more oxygen molecules physically adsorbed on MoS2 surface. Moreover, the spectra revealed a new peak at approximately 530.5 eV, which was assigned to the divalent oxygen of Mo–O bonds[52, 53](#_ENREF_52" \o "Colton, 1978 #25); this finding was consistent with the result of the XPS Mo 3d spectra. These results indicated that more oxygen atoms and molecules bonded chemically and physically to FLP-MoS2. This can be attributed to the roughness of the defect sites on the MoS2 nanoribbons and the long edges of the nanoribbons, which contained numerous unsaturated edge sites. These highly active centers can physically and chemically bond with adsorbates such as O2 molecules.[54–56](#_ENREF_54" \o "Nan, 2014 #15) The oxygen absorption on MoS2 can not only change the chemical valence of MoS2, but also, according to previous reports, can lead to p-type doping effect on MoS2 with O2 as an electron acceptor and MoS2 as an electron donor.[54](#_ENREF_54" \o "Nan, 2014 #15)



Figure 4. Raman and XPS spectra of FLP-MoS2. a) Raman spectra of pristine MoS2 (black line) and FLP-MoS2 (red line). XPS peak-split results of b) Mo 3d and c) O 1s spectra of FLP-MoS2.

To evaluate the electronic properties of the resultant MoS2 nanoribbon arrays, we fabricated a back gate FET by using MoS2 nanoribbon arrays (NbA) on SiO2/p+ Si substrates as the channel, with 5 nm Ti and 75 nm Au as the source and drain electrodes, respectively, through electron beam lithography (EBL), metal evaporation deposition, and a lift-off process. The electrical measurements of the fabricated FET were recorded using a Keithley 4200 semiconductor characterization system in air and at room temperature. The schematic of the structure and measurement of the fabricated MoS2 NbA-FET is shown in Figure 5a. Figure 5b presents the SEM image of the MoS2 nanoribbon arrays used for fabricating the FET with a channel length, *L* ≈ 7.3 μm; integral channel width, *W* ≈ 3.6 μm; a ribbon number of approximately 38; and gate dielectric thickness, *d* = 300 nm. Moreover, for comparison, a pristine MoS2 flake was fabricated as a back-gated FET. Figure 5c shows the drain–source current (*IDS*) versus drain–source voltage (*VDS*) characteristics of this NbA-FET under different gate voltages (*VG*) ranging from −15 to 15 V. For this output characteristic curve, the drain–source current changed linearly with the drain–source voltage, which indicated a nearly ohmic contact for this FET device. In addition, the output curve did not go through zero and exhibited a strong rectification of the drain–source current, which was different from the output characteristic curve of the FET fabricated using a pristine or undamaged MoS2 flake, as shown in Figure S3a. This change may be attributed to the p-type chemical doping of oxygen molecules on MoS2 nanoribbon arrays, which might cause transition of n-type channel to p-type channel or properties similar to a pn junction.[3](#_ENREF_3" \o "Nam, 2013 #2)

Figure 5d shows the drain–source current (*IDS*) versus gate voltage (*VG*) characteristics of this NbA-FET under different drain–source voltages (*VDS*) ranging from 6 to 10 V. The transfer curve of the NbA-FET was a nonmonotone variation and exhibited a strong rectification of the drain–source current, which was different from the transfer characteristic curve of the FET fabricated using the pristine/undamaged MoS2 flake. As shown in Figure S3b, the transfer characteristic curve of the pristine/undamaged MoS2 FET exhibited n-type conduction. However, the transfer characteristic curve of the MoS2 NbA-FET exhibits bipolar conduction, similar to p-n type transfer conduction (Figure 5d), with current chopping by a small voltage range. To further evaluate the electronic properties of this device, the on/off ratio, carrier mobility (μ), and subthreshold swing (SS) were calculated. The on/off ratio of the device was calculated to be 1.2 × 102 (p-type segment) and 1.7 × 103 (n-type segment) at a drain–source voltage (*VDS*) of 10 V. The carrier mobility (μ), based on results of previous reports,[23](#_ENREF_23" \o "Zhao, 2016 #1) is calculated as follows:

 (1)

where *L* and *W* are the length and width of FET channel, respectively; *d*, *ε0*, and *εr* are the thickness, vacuum permittivity, and relative permittivity of the grid dielectric layer, respectively; can be calculated from the *IDS*–*VG* curve. For this FET device, *L* ≈ 3.6 μm and *W* ≈ 7.3 μm; the grid dielectric layer was SiO2 with thickness *d* ≈ 300 nm, and its vacuum permittivity and relative permittivity were *ε0* ≈ 8. 85 × 10−12 F/m and *εr* ≈ 3.9 F/m,[23](#_ENREF_23" \o "Zhao, 2016 #1) respectively. Hence, the carrier mobility of this device was calculated to be approximately 2.5 × 10−3 cm2 V−1 s−1. The SS is calculated using the following equation (Equation 2, which was derived from previous reports:[57](#_ENREF_57" \o "Wang, 2017 #41)

 (2)

According to Equation 2, the SS of this device was approximately calculated to be 21 V/dec. The obvious strong rectification behavior of *IDS*–*VDS* and *IDS*–*VG* of the MoS2 NbA-FET indicated a change in the property of the material and new device properties. This change may enable new device applications. In addition, moderate modification of MoS2 flakes by femtosecond laser pulses would tune the n-type electronic properties of MoS2 FET, as shown in Figure S3. The on/off ratio of MoS2 FET was increased by a magnitude of two, and as shown in Figure S4, the drain–source current (*IDS*) increased quicker and reached saturation faster with the increase in gate voltage (*VG*).



Figure 5. Electrical test of MoS2 NbA-FET. a) Schematic of the structure and measurement of fabricated MoS2 NbA-FET. b) SEM image of MoS2 nanoribbon arrays used for fabricating FET. c) Output and d) transfer characteristic curve of the fabricated MoS2 NbA-FET.

**CONCLUSIONS**

Femtosecond laser pulse direct writing was used to nonthermally modify multilayer MoS2 flakes, induce separated MoS2 nanoribbon arrays, and arbitrarily pattern MoS2 flakes to form different MoS2 micro/nanostructures. Optical microscopy, AFM, and SEM were performed to characterize the physical micromorphology of the laser-processed MoS2 flakes. EDX mapping indicated the separation of MoS2 nanoribbons, which proved the formation of MoS2 nanoribbons rather than surface structures on MoS2. Moreover, Raman spectra indicated the nonthermal effect of femtosecond laser processing and proved the separation of MoS2 nanoribbons and the penetrating gaps among the nanoribbons. XPS spectra indicated that more oxygen molecules bonded chemically and physically to FLP-MoS2, which was attributed to the roughness of the defect sites on MoS2 nanoribbons and the long edges of the nanoribbons that contained numerous unsaturated edge sites and possessed highly active centers. An MoS2 NbA-FET was fabricated, and electrical tests were conducted to evaluate the electronic properties of the fabricated MoS2 nanoribbon arrays. Results indicated that the output and transfer characteristic curves exhibited strong rectification (not going through zero and bipolar conduction) of drain–source current, which was different from that of the FET fabricated using the pristine/undamaged MoS2 flake. This finding may be attributed to the coordinate structures and p-type chemical doping of oxygen molecules on MoS2 nanoribbon arrays, which might cause transition of n-type channel to p-type channel or properties similar to pn junction. Finally, the on/off ratio, carrier mobility, and SS were calculated. The proposed method indicated the ability of femtosecond laser pulses to directly induce two-dimensional nanostructures, property change in the materials, and new device properties, which may enable future enable new device applications.

**METHODS**

**Femtosecond laser pulse processing**: The light path setup of our femtosecond laser pulse processing system was reported in our previous study.[8](#_ENREF_8" \o "Zuo, 2018 #5)

**Characterization of MoS2 nanostructures**: Optical images were obtained using an Olympus microscope. The AFM characterization was performed using a SPM-960 atomic force microscope. The SEM images and EDX mapping spectra were obtained using a Hitachi scanning electron microscope. Raman spectra were obtained using a Renishaw InVia Reflex spectrometer with a 532 nm light source. XPS was conducted using a PHI Quantera X-ray photoelectron spectrometer.

**Device fabrication and electrical characterization**: Multilayer MoS2 flakes were placed on p+Si/SiO2 (300 nm) substrates and processed by femtosecond laser pulses. Then the substrates were spin-coated with poly methyl methacrylate (PMMA) solution. The source and drain electrodes were fabricated using three main steps: standard EBL was performed by Zeiss Supra55 SEM and Raith pattern generator to pattern the PMMA masks; 5 nm Ti and 75 nm Au were deposited through electronic beam evaporation; a lift-off process was performed in acetone. The electrical measurements of the fabricated MoS2 FET were recorded using a Keithley 4200 semiconductor characterization system in air and at room temperature.

**ASSOCIATED CONTENT**

**Supporting Information.** The following supporting information is available free of charge on the ACS Publications website:

Supplementary XPS Mo, O, and S spectra of pristine MoS2, XPS S spectra of FLP-MoS2, and supplementary electrical test results of MoS2 FETs with moderate surface modification by femtosecond laser pulses (PDF).

**AUTHOR INFORMATION**

**Corresponding Author**

\*E-mail (Lan Jiang): [@bit.edu.cn](mailto:jianglan@bit.edu.cn)

**Author Contributions**

All authors contributed to the writing of the manuscript. The final version of the manuscript was approved by all authors.

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**Notes**

The authors declare no competing financial interest.

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**Table of Contents**



**Supporting Information**

**Maskless Micro/Nanopatterning and** **Bipolar Electrical Rectification of MoS2 Flakes through Femtosecond Laser Direct Writing Processing**

Pei Zuo,1 Lan Jiang,\*,1 Xin Li, 1 Mengyao Tian,1 Peng Ran, 1 Bo Li, 1 Yongfeng Lu,2

*1Laser Micro/Nano Fabrication Laboratory, School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, P.R. China,*

*2Department of Electrical and Computer Engineering, University of Nebraska-Lincoln, Lincoln, NE 68588-0511, USA*

*\*Address correspondence to [@bit.edu.cn](mailto:jianglan@bit.edu.cn)*

**Supplementary figures and analysis.**



Figure S1. XPS spectra of pristine (P-) MoS2. XPS peak-split results of a) Mo 3d, b) O 1s, and S 2p spectra of P-MoS2.



Figure S2. XPS peak-split results of S 2p spectra of FLP-MoS2.

***Surface moderate modification of MoS2 flakes through femtosecond laser pulse processing for FET (-1 and -2).*** Figures S3 and S4 show the electrical test results for MoS2 FET-1 and MoS2 FET-2 before and after surface modification.

Figure S3a and c shows the drain–source current (*IDS*) versus drain–source voltage (*VDS*) characteristics of MoS2 FET-1 before and after surface moderate modification under different gate voltages (*VG*) ranging from −10 to 10 V. For these output characteristic curves, the drain–source current changed linearly with the drain–source voltage, indicating a nearly ohmic contact for these FET devices. Figure S3b and d shows the drain–source current (*IDS*) versus gate voltage (*VG*) characteristics of MoS2 FET-1 before and after surface moderate modification under different drain–source voltages (*VDS*) ranging from 0.05 to 0.5 V. The transfer characteristic curve of pristine/undamaged and surface-moderate-modified MoS2 FET exhibited n-type conduction. To further compare the change in electronic properties of this device, the on/off ratio, carrier mobility (μ), and subthreshold swing (SS) were calculated. For pristine/undamaged MoS2 FET-1, the on/off ratio, μ, and SS were respectively 2.84 × 103, 13.87 cm2 V−1 s−1, and 14.3 V/dec; for surface modified MoS2 FET-1, the on/off ratio, μ, and SS were respectively 1.85 × 105, 1.55 cm2 V−1 s−1, and 9.4 V/dec. These results indicated that after the femtosecond laser modification process, the on/off ratio of MoS2 FET-1 increased by a magnitude of two, whereas the carrier mobility decreased. The increase of on/off ratio might be attributed to the thinning effect of femtosecond laser on the MoS2 flake.[1](#_ENREF_1" \o "Chen, 2013 #42) The decrease in μ might be attributed to the defect states on laser-modified MoS2. This decrease can reduce the portion of mobile carriers in the conduction band and increase the portion of trapped carriers that do not contribute to charge transport, thereby leading to the degradation of the effective mobility.[2](#_ENREF_2" \o "Bertolazzi, 2017 #4)



Figure S3 Electrical test of MoS2 FET-1 before and after surface modification. a) Output and b) transfer characteristic curve of MoS2 FET-1 before surface modification. c) Output and d) transfer characteristic curve of MoS2 FET-1 after surface modification. e) SEM image of MoS2 FET-1 after surface modification.

Figure S4a and c shows the drain–source current (*IDS*) versus drain–source voltage (*VDS*) characteristics of MoS2 FET-2 before and after surface moderate modification under different gate voltages (*VG*) ranging from −10 to 10 V. For these output characteristic curves, the drain–source current changed linearly with the drain–source voltage, indicating a nearly ohmic contact for these FET devices. Figure S4b and d indicates the drain–source current (*IDS*) versus gate voltage (*VG*) characteristics of MoS2 FET-2 before and after surface moderate modification under different drain–source voltage (*VDS*) ranging from 0.75 to 1.5 V. The transfer characteristic curves of pristine/undamaged and surface-moderate-modified MoS2 FET exhibited n-type conduction. To compare the change in electronic properties of this device, the on/off ratio, μ, and SS were calculated. For pristine/undamaged and surface-modified MoS2 FET-2, the on/off ratio and μ were unchanged at 1.13 × 105 and 3.5 × 10−2 cm2 V−1 s−1, respectively, whereas SS was modified from 18.5 to 11.7 V/dec. However, the drain–source current (*IDS*) of surface modified MoS2 FET-2 increased quicker and reached saturation faster with the increase in gate voltage (*VG*) compared with the pristine/undamaged MoS2 FET-2.



Figure S4 Electrical test of MoS2 FET-2 before and after surface modification. a) Output and b) transfer characteristic curve of MoS2 FET-2 before surface modification. c) Output and d) transfer characteristic curve of MoS2 FET-2 after surface modification. e) SEM image of MoS2 FET-2 after surface modification.

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