

Plasma Process for Generating Movable Sulfur Vacancies in MoS₂ Layers and Fabricating Analogue Ionically-Coupled Multi-Terminal Memristors

Da Li, Jeongseop Yoon, Byunghoon Ryu, and Xiaogan Liang*

Mechanical Engineering Department, University of Michigan, Ann Arbor, MI 48109

Memristors have been extensively studied as important components for constructing artificial neural networks for neuromorphic computing applications.¹ The current memristor-based neural networks based on transition metal oxides are still mainly driven by electronic effects (*e.g.*, voltage distributions), while ionic-interactions, as observed in biological networks, could effectively reduce the power consumption of neural networks and also result in a higher analogue computation precision.² To emulate such sophisticated interactions, including synaptic competition and cooperation, ionically-coupled multi-terminal memristors are needed.^{3, 4, 5} Recently, memristors based on 2D semiconductors (*e.g.*, MoS₂ and WSe₂) were demonstrated.^{3,4} Such 2D semiconductor memristors exhibit low threshold fields for initiating memristive switching and analogue switching characteristics. The memristive switching behavior of such memristors has been attributed to multiple mechanisms.⁴ One of these mechanisms is the interaction between movable ionic defects in 2D semiconductor layers and the Schottky Barriers at semiconductor/metal interfaces.⁴ This mechanism could be further employed to produce ionically-coupled memristors. Therefore, it is highly desirable to develop materials processing and fabrication approaches capable of generating a high areal density of movable ionic defects in 2D semiconductor layers.

Here, we report that a plasma treatment can form a high density of movable S vacancies in few-layer MoS₂ layers (Fig. 1 (a)). The memristors made of such plasma-treated MoS₂ layers exhibit analogue pulse-programmed switching characteristics with a good linearity in switching courses. In addition, multiple such memristor channels can be ionically coupled and could be further exploited for emulating complex synaptic interactions in biological systems.

Fig. 1 (b) shows the SEM image of a plasma-treated MoS₂ memristor after electrical stimulation (*i.e.*, 5000 20V, 2ms pulses). Three Auger electron spectroscopy (AES) sampling areas denoted by red boxes in Fig. 1 (b) are chosen to track the spatial migration of S-vacancies after electrical stimulation. Specifically, the atomic ratios of S/Mo atoms measured within these sampling areas are plotted in Fig. 1 (c) (*i.e.*, 1.74 for Area 1; 1.93 for Area 2; 2.11 for Area 3). This AES result indicates that plasma treatment can generate a significant number of movable S vacancies, which can migrate under the influence of an electric field. Fig. 1 (d) shows the corresponding analogue pulse-programmed switching characteristics of the device with a good linearity and a relatively high on/off ratio (> 6). Fig. 2 (a) shows the optical micrograph of a representative multi-terminal MoS₂ memristor, in which a lithographically patterned few-layer MoS₂ flake and five Ti/Au electrodes labelled with “0”, “1”, “2”, “3”, “4” form four branch memristors (*i.e.*, 0-1, 0-2, 0-3, and 0-4 memristors). Fig. 2 (b) displays three conductance states of the 0-2 memristor represented by the small-signal I-V characteristic curves in different programming conditions (black curve: initial state; red curve: measured after a train of 1000 +12V, 2ms voltage pulses was applied to the 1-0 memristor; blue curve: measured after another train of 2000 -20V, 2ms pulses was applied to the 1-0 memristor). This work preliminarily shows that different branch memristors can be ionically coupled to each other through controlling the spatial distribution of S vacancies around their common Schottky junctions. In addition, we have demonstrated that such ionic coupling effects can be also achieved by the field-driven migration

of doped ions in MoS₂ layers. The specific methods for fabricating and characterizing analogue multi-terminal MoS₂ memristors will be explained in the presentation.

This work leveraged the unique in-plane transport properties of 2D layered semiconductors for memristive electronics applications and advanced the nanofabrication capability for controlling memristive switching behaviors.

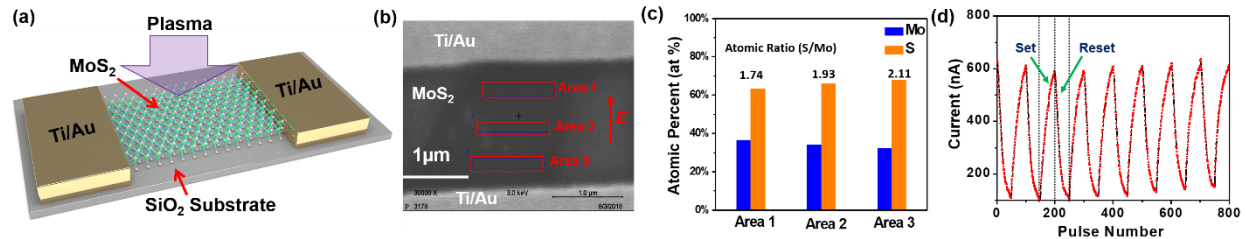


Fig. 1 (a) Schematic illustration of the generation of moveable sulfur vacancies in MoS₂ layers through plasma treatment; (b) SEM image of a representative plasma-treated MoS₂ memristor after electrical stimulation, in which the red boxes denote the AES sampling areas, and the red arrow indicates the electric field direction of applied pulses; (c) Atomic ratios of S/Mo atoms in three sampling areas mentioned in (b); (d) Analogue pulse-programmed switching characteristics of the plasma-treated MoS₂ memristor, which was modulated by a sequences of set/reset trains (set trains: 50 +5V, 5ms pulses; reset trains: 50 -3V, 2ms pulses).

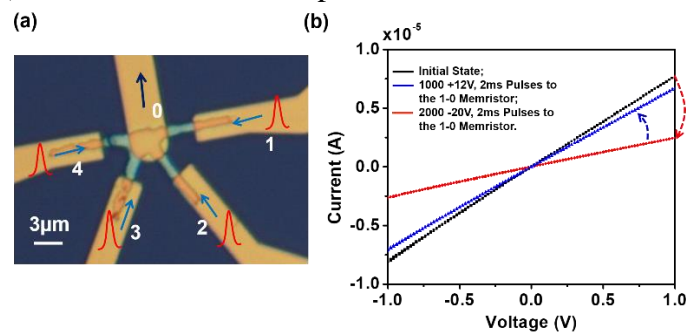


Fig. 2 Demonstration of a multi-terminal MoS₂ memristor consisting of four ionically coupled branches: (a) optical micrograph of the multi-terminal MoS₂ memristor, in which a lithographically patterned MoS₂ flake in together with five Ti/Au electrodes labelled with “0”, “1”, “2”, “3”, “4” form four memristor branches (*i.e.*, 0-1, 0-2, 0-3, and 0-4 memristors), each channel length is 3 μ m; (b) small-signal I-V curves measured from the 0-2 memristor at its initial state (black curve), after application of 1000 +12V, 2ms pulses to the 1-0 memristor (red curve), and after application of 2000 -20V, 2ms pulses to the 1-0 memristor (blue curve).

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