



# Anomalous electrical bistability in lateral grain rich polycrystalline molybdenum disulfide thin films

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## ABSTRACT

Lateral-grain- rich polycrystalline MoS<sub>2</sub> films have been prepared using sulfurization of the Mo-coated glass substrate. The goal of such synthesis is to achieve electrical bistability without fabricating a sandwiched structure that generally utilizes other materials, such as graphene or N-vinylcarbazole. These films show a symmetrical three-stage electrical behavior that follows a different charge transport mechanism. In a single voltage sweep, both on and off stages appear. The intermediate stage obeys a trap-controlled space charge limited current mechanism, whereas the ON stage follows a Schottky emission model. The overall charge transport mechanism was explained on the basis of contributions from dangling bonds, stoichiometric defects, atmospheric adsorbates, s-vacancies, and free charge carriers. Under an assumption of the high contribution of the leakage current, the simulated I-V curve shows two different sections. Such a trend was observed in the experimental I-V curves.

## 1. Introduction

2D materials possess special electrical, chemical, and physical properties mainly due to enhanced surface area, weak forces between layers, and quantum confinement effects in specific dimensions [1,2]. Because of these specific properties they have been deeply investigated in past few decades, and in particular, their potential applications in high speed electronics, energy conversion devices, and bio-sensing can be revolutionary [3]. In a line of investigation of the layered 2D compounds, transition metal dichalcogenides (TMDC) are the top choice candidates due to an indirect to direct transition within a band in the case of their monolayer counterparts. Several different methods have been proposed to grow TMDC layers as well as nanocrystals [4]. Among them, ion-intercalation and exfoliation, chemical vapor deposition, and colloidal chemical synthesis approaches are often utilized [5]. A non-injection method [5] has been reported that does not require the pre-synthesis of precursors, for the growth of MoS<sub>2</sub>, a key material in the category of TMDCs. MoS<sub>2</sub> has exhibited its applicability in a range of applications including electronic transistors, batteries, photovoltaics, and catalysts. Note that the transition metals have a partially occupied d sub-shell (different level of filling of bands), and hence possess a range of electronic properties [6]. MoS<sub>2</sub> structure consists of Mo atoms in one plane that are sandwiched by sulfur atoms (See Fig. 1a). Hence, the aggregate of three layers form a single layer of MoS<sub>2</sub>. In this monolayer, Mo and S atoms are bonded by the strong covalent

interactions whereas the interactions between sulfur layers are relatively weak due to van der Waals force [6]. One of the great advantages of MoS<sub>2</sub> is enhanced light confinement (about an order of magnitude greater than graphene) [7]. The endless possibilities of the applications of 2D material are beyond the scope of this manuscript. However, some of the key areas including electronics, sensors, and energy are of particular interest. Magnetic bistability [8], optical bistability [9], and electrical bistability are some of the recently observed novel behaviors of 2D materials. Among them resistance switching phenomena [10] based on electrical bi-stability have been investigated and resonant tunneling diodes were specifically explored in order to facilitate ultra-fast nonvolatile memory operation. The key advantages of nonvolatile memory elements are long time storage, low energy operation, and large density storage [10]. These can be used for potential computer devices that have high performance storage and low power consumption. Non-volatile memories can also be applied to the 'off computing system' in order to reduce the energy consumption.

Many different types of memory elements including ferroelectric RAM, magnetoresistive RAM, Phase change RAM, and Resistance RAM have been investigated in the past few decades under a goal of increasing performance [10]. In the case when resistance is controlled by electrical stimulation, the resulting technology has emerged as a NAND flash technology alternative [11]. The low power consumption, fast switching, and scalability are key features of this technology. A series of candidates is considered as promising in this category. These promising

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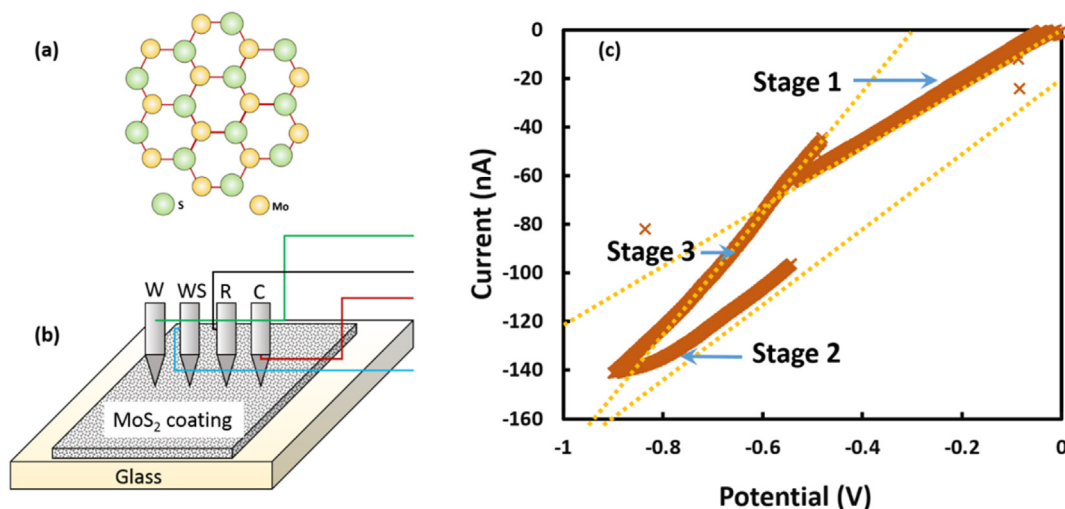


Fig. 1. (a) Crystal structure of MoS<sub>2</sub> (b) Schematic diagram of four stage current-potential measurements and (c) experimental i-v behavior showing multistage conduction.

candidates include: graphene oxide (GO), perovskite, polymer, metal oxides, transition metal di-chalcogenides, and other composite materials [11,12]. Among them, TMDCs have some advantages including three dimensional stacking capability, flexibility, ease of synthesis, and low cost [11].

A continuous reduction of flash memory scale (~less than 10 nm) causes the exploration of a new materials and processes in order to provide other methods for high density memory devices [13]. Chalcogenide materials and their phase change memory property under the application of potential or electrical signal and caused by local temperature elevation, can also be utilized [13]. An electrical current readout can distinguish the crystalline and amorphous phases, based on their conductivity and band structure. A suitable design of programming pulse can also result in ultrafast ~1 ns switching [13]. Electrical bistability in the case of MoS<sub>2</sub> is less common. In one case of a sandwich structure of MoS<sub>2</sub> sheet based device, fabrication steps utilize the coating of a 1:1 mixture of MoS<sub>2</sub> and poly (N-vinylcarbazole) PVK in chlorobenzene [5]. A multilevel resistive switching device based on MoS<sub>2</sub>-graphene oxide, exhibited nice nonvolatile memory performance, based on the modulation of the density of trapped space charges [11]. However, a simpler method is essential to develop, where electrical bistability is possible for MoS<sub>2</sub> based devices. In order to address this challenge a single-step fabricated MoS<sub>2</sub> sample is examined and associated electrical bistability (see: Fig. 1b and c) is studied for further improvement and device applications.

## 2. Experiments

In order to grow the layer of MoS<sub>2</sub>, the authors utilized Mo coated glass (Mo thickness was ~280 nm). The Mo-coated glass was acquired from Guardian Glass S.A (Dudelange - G.D. of Luxembourg). The Mo-coated glass was properly cleaned and dried with nitrogen and subsequently sulfurized by annealing in an evaporated elemental sulfur environment. Elemental sulfur (99.99%) placed on a quartz boat, heated in a tube furnace, and Mo-films exposed to the elemental sulfur at 600 °C for 3 h. The alumina tube was purged with argon for 30 min to displace air, prior to sulfurization. After sulfurization, films were allowed to cool naturally and subsequently characterized. The morphology and grains of MoS<sub>2</sub> layers was examined using a field emission scanning electron microscope (Hitachi S-4800 SEM) with a tungsten filament based field emission gun at 5kV accelerating voltage and emission current ~18 μA. Energy dispersive X-ray spectroscopic analysis (EDS) was utilized for the chemical analysis and elemental mapping of the MoS<sub>2</sub> layers. An Oxford (X-Max) EDAX detector unit

attached to the SEM, was utilized for this. Most of the EDS analyses were performed at ~20 kV accelerating voltage and high probe current. The X-ray diffraction (XRD) of the MoS<sub>2</sub> film was carried out using a Miniflex XRD (CuK<sub>α</sub> = 1.54059 Å, Rigaku, Akishima-shi, Tokyo, Japan) in the 2θ range of 20–90° with a step size of 0.015° and a dwell time of 1° min<sup>-1</sup>. The diffraction pattern was analyzed using Rigaku PDXL2 analysis software and indexing was done with a standard JCPDS card. Raman examinations were performed using a portable Raman spectrometer (Raman system: make R 3000 QE) with a 785 nm laser excitation at power ~130 mW. The Raman spectrophotometer wavelength stability was good as it has less than 1 cm<sup>-1</sup> drift for over a 12 h period.

## 3. Results and discussion

### 3.1. Morphology and phase examinations

In Fig. 2, SEM images and elemental mapping information of MoS<sub>2</sub> film grown on glass support, are shown. The low magnification SEM image indicates (Fig. 2a) the formation of an adherent film of MoS<sub>2</sub> on glass substrate. The image indicated that a uniform continuous MoS<sub>2</sub> film formed at the glass substrate. To confirm the uniformity of the film, we mapped the film for the elemental composition over a large area. A small section of the EDS map is shown in Fig. 2e-g clearly shows the composition of the film is uniform and continuous. The layered nature of the MoS<sub>2</sub> film is evident when the magnification of the image increased to a higher level as shown in Fig. 2b and e. Inset in Fig. 2c is a view of the uniform coating of MoS<sub>2</sub> on the glass substrate. Highly oriented grains of MoS<sub>2</sub> are easily evident in Fig. 2e. A close examination of Fig. 2a suggests the formation of flaky sheets that are partially supported from the original MoS<sub>2</sub> matrix (see arrows). The length of these lateral sheets varies from 100 nm to 800 nm. The thickness of the lateral sheets were estimated from the location where they were found to be vertical to the sample. Elemental maps were obtained for the area shown in Fig. 2e using an EDS detector, and they show a uniform distribution of Mo and S. These results confirm the hypothesis of uniform deposition of a MoS<sub>2</sub> layer on glass substrate. Similar structure and morphology of MoS<sub>2</sub> have been reported by various groups [14,15]. Note that the film quality and grain size also depends on the method of growth, sulfurization, and time of deposition. It was observed that MoS<sub>2</sub> film gets smoother from a granular structure as the deposition time i.e. the thickness of the film increased [15]. Interestingly, in our present study, the well-defined precipitate structures were observed in the MoS<sub>2</sub> film, as shown in Fig. 2c (as well in the

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