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Growth and Applications of Surface Oxides on Two-Dimensional Transition Metal Dichalcogenides

Mahito Yamamoto

WPI Center for Materials Nanoarchitechtonics (WPI-MANA)
National Institute for Materials Science
1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan
Phone; +81-29-851-3354, Email; Yamamoto.Mahito@nims.go.jp

Abstract

Surface oxidation of two-dimensional (2D) transition metal dichalcogenides is of increasing interest for applications in electronics and optoelectronics. Here I will review our recent work on homogeneous and thickness-controlled oxidation of 2D tungsten diselenide (WSe_2). Then I will report on the impact of the surface oxides on the transport properties of the underlying WSe_2. Possible electronic and optoelectronic applications will also be discussed.

1. Introduction

Growing surface oxides is a key process in the fabrication of semiconductor devices, i.e., metal oxide semiconductor field effect transistors (MOSFETs). On silicon, for example, one can simply grow a homogeneous oxide film with excellent dielectric and insulating properties just by oxygen treatments, which is one reason why silicon dominates other semiconductors as a building block of electronic devices.

Recently, two-dimensional (2D) crystals of transition metal dichalcogenides (TMDCs) such as MoS_2 and WSe_2 have emerged as a new class of semiconductors with potential as "post-silicon" materials. Indeed, 2D TMDCs have been used to fabricate FETs with comparable or even better performances than silicon FETs [1], regardless of their atomically thin bodies. However, in contrast to silicon, homogeneous growth of surface oxides on 2D TMDCs is seemingly difficult because of the absence of the dangling bonds, hampering their practical use in electronics. Toward the progress of post-silicon electronics based on 2D TMDCs, a controlled surface oxide fabrication process is essential.

Here I will first introduce our work on homogeneous and thickness-controlled oxidation of 2D WSe_2. Then, as a first step for MOSFET applications of the surface oxide, I will discuss how the surface oxides influence the transport properties of the underlying WSe_2. Lastly I will present potential electronic/optoelectronic applications based on atomically thin heterostructures of the surface oxide and WSe_2.

2. Results

Self-limiting layer-by-layer oxidation. We find oxygen treatments of 2D TMDCs result in negligible oxidation at low temperatures, while in etching of the surfaces at high temperatures [2]. Instead, ozone (O_3) exposure leads to uniform and controllable surface oxidation of 2D WSe_2 at relatively low temperatures [3]. Below 100 °C, the O_3 treatment results in the formation of oxides, WO_x ($x \leq 3$) at the edges and triangular oxide islands on the surface (Fig. 1a). With further exposure, the oxides grow laterally and coalesce to each other (Fig. 1b), ultimately forming a uniform film on top (Fig. 1c). However, oxidation does not progress to the underlying layers. At 200 °C, the surface layers are oxidized in the layer-by-layer regime, up to trilayers. Using Raman and photoluminescence spectroscopy, we find that the underlying WSe_2 is mechanically decoupled from the top oxide, promising MOSFET applications.

Fig. 1. Atomic force microscopy images of bilayer and 4-layer WSe_2 on SiO_2 after O_3 exposure at 70 °C for (a) 0.5, (b) 1, and (c) 1.5 hours.

Controlled electron transfer doping. Next we investigate the transport properties of 2D WSe_2 with the surface oxides grown by O_3 exposure [4]. We find atomic layers of WSe_2 with the oxides are hole-doped, with the dopant concentrations being environmentally "tunable" from the degenerate to the nondegenerate regime. Right after the O_3 treatment at 100 °C, the WSe_2 underlayers are degenerately hole-doped and almost metallic due to the electron transfer from WSe_2 to high-electron affinity WO_x (Fig. 2a). However, upon air exposure the electron transfer is suppressed and, hence, the dopant concentration is reduced down to the

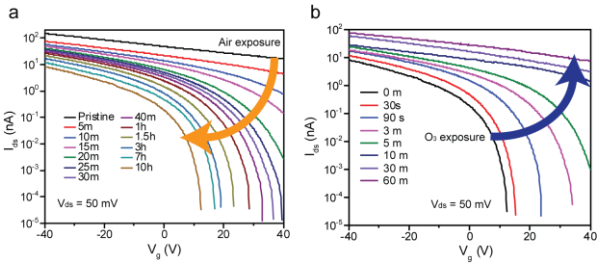

Fig. 2. Transfer curves of a single-layer WSe_2 FET with the surface oxide after consecutive (a) air and (b) O_3 exposure.

degenerate regime (Fig. 2a). The doping concentration can be retrieved by O_3 exposure at room temperature (Fig. 2b).

Controlled electron trapping. We find the surface oxides grown on WSe_2 also serve as tunable electron trapping layers. Pristine 2D WSe_2 FETs fabricated on SiO_2 with Ti/Au contacts show ambipolar transport with higher conduction in electron transport. However, after O_3 oxidation, the electron transport is suppressed even at high positive gate voltages. Furthermore, the FETs show hysteresis upon the gate sweep and the hysteresis window increases with the maximum gate voltages. Also, white LED illumination results in photocurrents that increase with illumination time and persist even after turning off the light. The controllable charge trapping properties suggest the application in nonvolatile electronic/optoelectronic memory (Fig. 3).

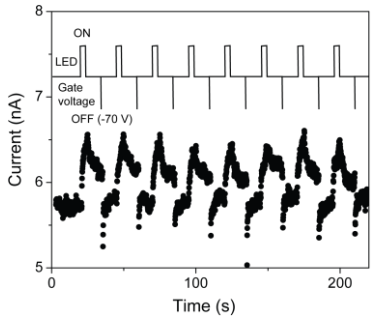

Fig. 3. Optoelectronic switch based on single-layer WSe_2 with the surface oxide.

3. Conclusions

In this work, we present a simple and reliable method to grow homogeneous and thickness-controlled oxide films on 2D WSe_2 for the first time. The oxide film acts as a tunable charge transfer and trapping layer for the 2D WSe_2 FET. Our work may serve as a first step toward the development of 2D semiconductor-based MOSFETs but also have important implications for other electronic and optoelectronic applications including nonvolatile memory, photodetectors, and gas sensors.

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