G-9-04 Extended Abstracts of the 2020 International Conference on Solid State Devices and Materials, VIRTUAL conference, 2020, pp473-474

Electrochemically-modulated photoluminescence of single-layer MoS_2 for bio-imaging

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Abstract

Visualizing biological activity in real time poses a novel way for biosensing. Single-layer Molybdenum disulfide (MoS_2) shows strong photoluminescence (PL) under photo-excitation due to its semiconducting nature with direct bandgap. In this work, we demonstrate pH-sensitive photoluminescence of single-layer MoS_2 grown by chemical vapor deposition (CVD). While CVD MoS_2 grown from MoS_2 powder did not have a strong pH sensitivity, MoS_2 grown from MoO_2 powder exhibited a sensitivity to the solution pH in its PL intensity. Next, we utilized the single-layer MoS_2 as the active layer of an imaging sensor to monitor activity of individual bacteria by the MoS_2 PL. For the demonstration, we employed Lactobacillus (LGG). It is one of the most studied probiotic bacteria which consumes glucose and produces lactic acid by fermentation. The bacteria were placed on a single-layer MoS_2 and its PL intensity was monitored in real time.

1. Introduction

As a new tool for biosensing with a high sensitivity, two-dimensional (2D) materials have been gaining a wide attention due to its high specific surface area and atomically flat surface. Electrical conductivity of graphene has been utilized to detect pathogenic bacteria in human mouth. Raman imaging of graphene has been demonstrated to monitor production of immunoglobulins through observations of pH distribution at the vicinity of human embryonic kidney cells. Ttransition metal dichalcogenide represented by molybdenum disulphide (MoS_2) is a promising platform for versatile applications in biosensing. The semiconducting properties of MoS_2 coupled with its large surface area allows for highly sensitive detection of external environments. Due to its semiconducting nature, MoS_2 field effect transistor (FET) has revealed a 74-fold higher sensitivity than that of graphene FETs. MoS_2 has demonstrated its potential for a variety of biosensing. Especially for cells, MoS_2 FET has shown its ability to sense cancer cell activity though monitoring hydrogen peroxide.

Because of its direct band-gap property, single-layer MoS_2 has exhibited strong photoluminescence (PL) under photo-excitation, and the PL intensity has been demonstrated to change with the internal electron density [1]. Its optical absorption and PL have been sensitively affected by adsorbates and pH change in the solution [2, 3]. Althogh

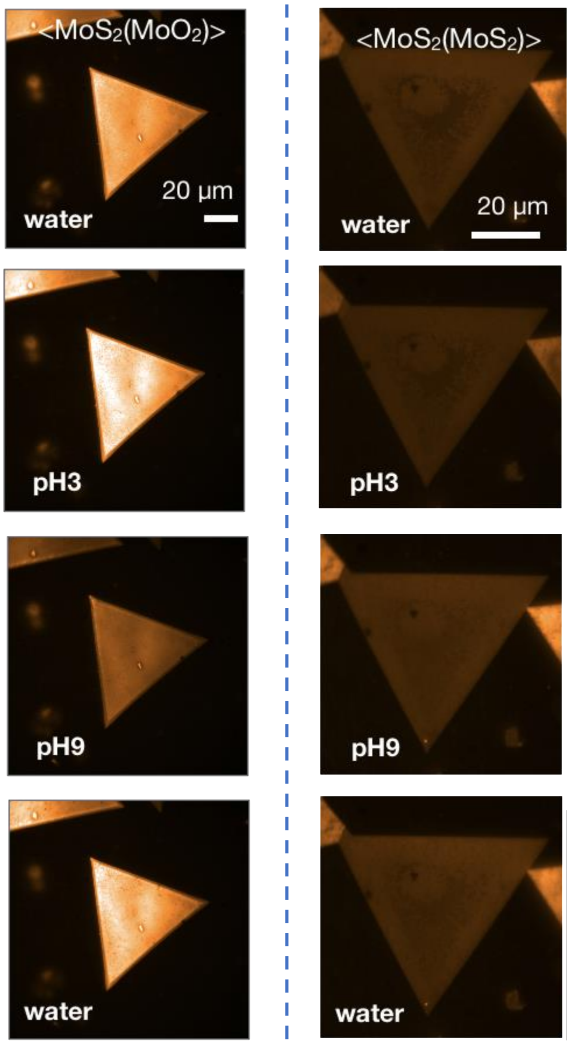

Fig. 1 Photoluminescence images of MoS_2 under aqueous solution with various pH. These MoS_2 were grown by chemical vapor deposition with a source material of either MoO_2 or MoS_2.

MoS2 FET is used widely as an active layer for electrical biosensing with a high sensitivity, the optical properties have not been employed fully. Futhermore, while chemical vapor deposition (CVD) is a promising method for synthesizing large and high quality MoS_2, optical properties of CVD-grown MoS_2 under physiological solution is not clear. Understanding the effect of the pH on PL of MoS_2 can lead us to utilize MoS_2 as a novel optical biosensing platform.

2. Result and Discussions

The PL properties can be employed to visualize the spatial distribution of biological moieties. For the sake of this, in this work, we employed single-layer MoS_2 as an active layer for PL based imaging sensor [4]. First, we demonstrate their PL responses to solution pH under aqueous condition by means of PL spectroscopy and imaging. MoS_2 with different Mo sources and additive such as MoS_2, MoO_2 and NaCl was synthesized. It was found that the PL response was highly affected by the Mo sources. While MoS_2 made from MoO_2 showed the highest modulation of PL by solution pH, MoS_2 made from MoS_2 did not show significant response (Fig. 1). Furthermore, we demonstrated enhanced pH-sensitivity by UV/O_3 treatments. Spectral analysis on PL and X-ray photoelectron spectroscopy revealed that the PL responses to solution pH were not directly correlated to the defect density (S/Mo ratio) of MoS_2, but likely correlated to its state of defects and initial carrier density. These findings allow us to utilize MoS_2 as an optical sensing platform. Next, we observed that PL intensity of MoS_2 was modulated by the concentration of lactic acid in aqueous solution. For this measurement, we fabriated elecrochemical cell for MoS_2 as shown in Fig. 2a. In this setup, we can apply elecrtrochemical potential into the MoS_2 to mudulate its electron density and PL intensity to enhance the contrast in the PL imaging and sensitivity to the environmental condition. Using this setup, the adhesion of Lactobacillus (LGG) on single-layer MoS_2 was visualized by PL imaging under phosphate buffer (PB) with a good contrast between the locations with and without LGG (Fig. 2c). It indicates that production of lactic acid by LGG can be visualized through MoS_2 PL [5].

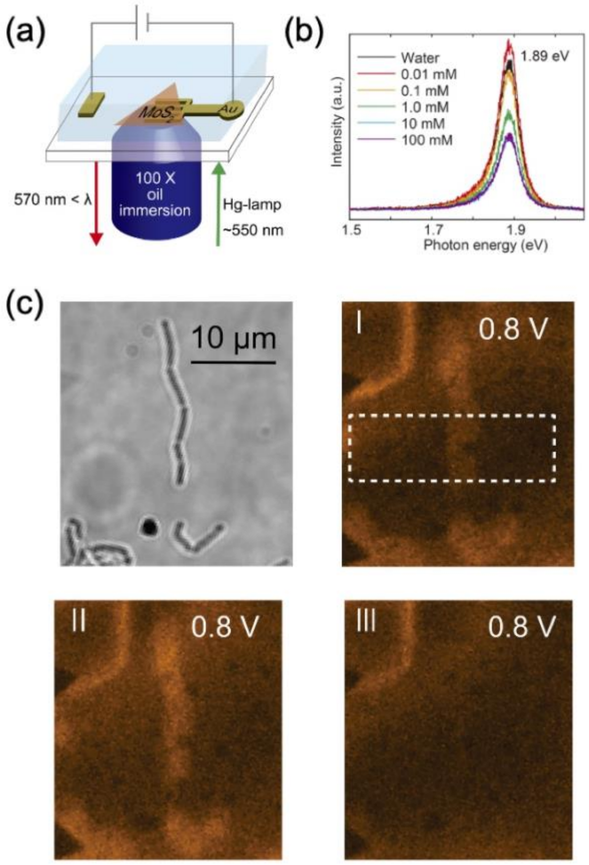

Fig. 2 (a) schematic of experimental setup for PL imaging with an electrochemical cell. MoS_2 was excited through an oil immersion lens from the bottom over the cover glass and the PL was collected by the objective lens. (b) PL spectra of MoS_2 under lactic acid solution with various concentrations. (c) Optical and PL images of LGG on MoS_2. The location where LGG exists shows higher intensity than other regions without LGG. During the incubation of LGG, the contrast of the PL image increased (I- II). However, the intensity disappeared after stopping providing sugars to the LGG (III), indicating the production of lactic acid by LGG might stop.

3. Conclusions

We demonstrate pH-sensitive photoluminescence of single-layer MoS_2 grown by chemical vapor deposition (CVD). While CVD MoS_2 grown from MoS_2 powder did not have a strong pH sensitivity, MoS_2 grown from MoO_2 powder exhibited a sensitivity to the solution pH in its PL intensity. Next, we utilized the single-layer MoS_2 as the active layer of an imaging sensor to monitor activity of individual bacteria by the MoS_2 PL. For the demonstration, we employed Lactobacillus. We successfully observed an intensity contrast in the PL images which is caused by the bacteria on a single-layer MoS_2. This work proved a new way to visualized biological environment at the vicinity of MoS_2 surface. Thus, it can open a new door for the wide usage of MoS_2 PL for biosensing.

Acknowledgements

The research was supported by JSPS KAKENHI Grant Number 25706012 and 16H05973.

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