High temperature stability of MoS_2 probed by combining thermal desorption spectroscopy and atomic layer deposition

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Introduction

Defects engineering in 2D materials is crucial to improve the device performance as well as to modify the material properties. Especially for MoS_2, controlling sulfur vacancy has still been intense issue even though many healing processes using adsorbates have already been proposed. This is largely due to the limited understanding on the defect formation. Recently, detailed evolution of sulfur vacancy in MoS_2 on Au and in suspended MoS_2 has been reported by using STM and in-situ TEM during the high temperature annealing in vacuum [1,2]. However, both conditions are different from the standard device circumstances, that is, MoS_2 on insulating SiO_2/Si substrate, where oxygen exists around MoS_2. To make matters worse, no measurement method for sulfur vacancy with atomic resolution is available. Here, it is well known that Al_2O_3 during atomic layer deposition (ALD) adsorbed only at the defect sites, which will allow us to detect defect evolution with the assistance of surface topology observed macroscopically by AFM. In this study, we investigate the desorption behavior of species from MoS_2 flake on SiO_2/Si wafer by thermal desorption spectroscopy (TDS) up to elevated temperature of ~1000°C. Subsequently, the defect evolution was traced by 2-nm ALD Al_2O_3.

Experimental

The large amount of molybdenum disulfide flakes was prepared onto clean SiO_2/Si substrate. Then, TDS measurement was carried out from 25°C up to ~1000°C at 20 K/min in UHV. Then, 2-nm Al_2O_3 (10 cycles) was deposited on MoS_2 by ALD, followed by AFM measurement. Alternatively, the structural and electrical changes were inspected for monolayer MoS_2 by Raman and PL measurements after TDS.

Results & Discussion

Fig. 1 shows TDS spectra for M/z = 32 (S, O_2), 64 (SO_2). It is clear that there are two dominant desorption peaks at ~400°C and ~650°C for SO_2. Since the desorption of O_2 was observed from 100°C to 500°C, it is considered that S atom in MoS_2 reacted with O_2 and desorbed as SO_2 at ~400°C and ~700°C. Finally, single S atom started to desorb directly at ~900°C. These results suggest that the oxygen adsorption on the MoS_2 surface is critical for S desorption as SO_2 at low temperature and the high temperature stability of MoS_2.

Fig. 2 shows the coverage ratio of ALD Al_2O_3 on MoS_2 flakes as a function of maximum TDS temperature. The coverage increased even at 355°C, which is consistent with the detection of SO_2 desorption in TDS. Full coverage is observed after the annealing to 640°C, at which the morphological difference between the step and terrace became blurred, indicating a wide spread defect formation on MoS_2 surface. **Fig. 3** shows PL measurements at different maximum TDS temperatures. The exciton peak intensity increased up to ~630°C because O_2 chemisorbed at defect sites [3]. However, at ~835°C, it was drastically reduced, and defect-related peak appeared [4]. It should be noted that two distinct Raman peaks in monolayer MoS_2 did not broaden even after annealing up to ~835°C, suggesting that crystallinity of MoS_2 is still preserved.

In summary, ALD-assisted TDS experiment clearly demonstrated defects evolution of MoS_2/SiO_2 system under UHV annealing condition. Surprisingly, MoS_2 shows high temperature stability even on the SiO_2/Si substrate at least up to 630°C. Since S was desorbed as SO_2 at low temperature, the reduction of O_2 around MoS_2 will be the key to prevent from defect formation.

Reference:

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[2] Q. Chen, *et al.*, Nano lett. 2017, 17, 5502.
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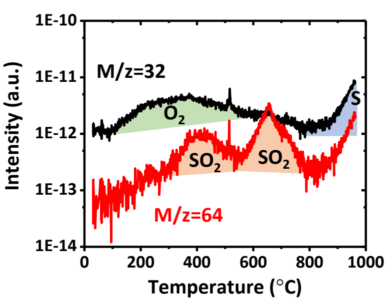

Fig. 1 TDS Spectrum of bulk MoS_2.

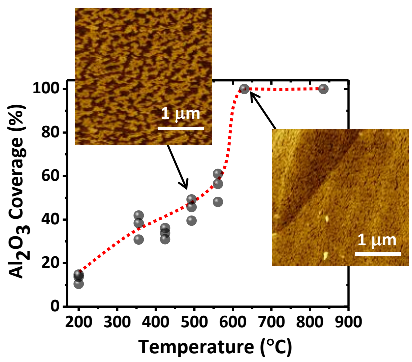

Fig. 2 Coverage ratio of ALD-Al_2O_3 on bulk MoS_2 annealed at different temperatures.

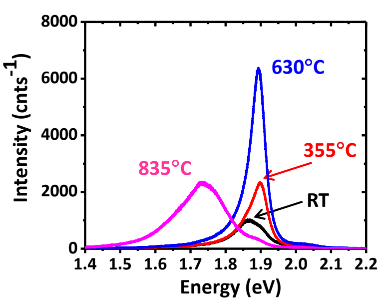

Fig. 3 PL Spectra of monolayer MoS_2 annealed at different temperatures.