

Supplementary Information

Vertical Heterostructures of MoS₂ and Graphene Nanoribbons by Two-Steps Chemical Vapor Deposition for High-Gain Photodetectors

Rozan Mohamad Yunus,^a Hiroko Endo,^b Masaharu Tsuji,^{a,b} and Hiroki Ago^{a,b,c,}*

^a Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Fukuoka

816-8580, Japan

^b Institute for Materials Chemistry and Engineering (IMCE), Kyushu University, Fukuoka

816-8580, Japan

^c PRESTO, Japan Science and Technology (JST), Saitama 332-0012, Japan

Experimental

CVD growth of graphene nanoribbon (GNR) and its heterostructure with MoS₂

We report a two-step CVD process to obtain heterostructure of MoS₂ on GNRs, with a supporting substrate of SiO₂/Si. In the first step, GNR was grown by ambient CVD on Cu/MgO(100) and transferred onto SiO₂/Si substrate.²¹ After transferring the GNRs onto SiO₂/Si substrate, MoS₂ was grown on the GNRs by a second CVD process using MoO₃ and S powder, as shown in Fig. S1. High purity MoO₃ (0.3 g; from Aldrich; 99% purity) was placed in a quartz sample holder at the zone 1 of the furnace. A transferred GNRs on SiO₂/Si Substrate was placed at the zone 2. Sulfur powder was gradually heated by heating ribbon and carried by Ar (500 sccm) into the furnace. The furnace was gradually heated from room temperature to 900-950 °C for zone 2 and 3 and 600-650 °C for the zone 1. After keeping for 30 - 60 minutes, the furnace was naturally cooled down to room temperature.

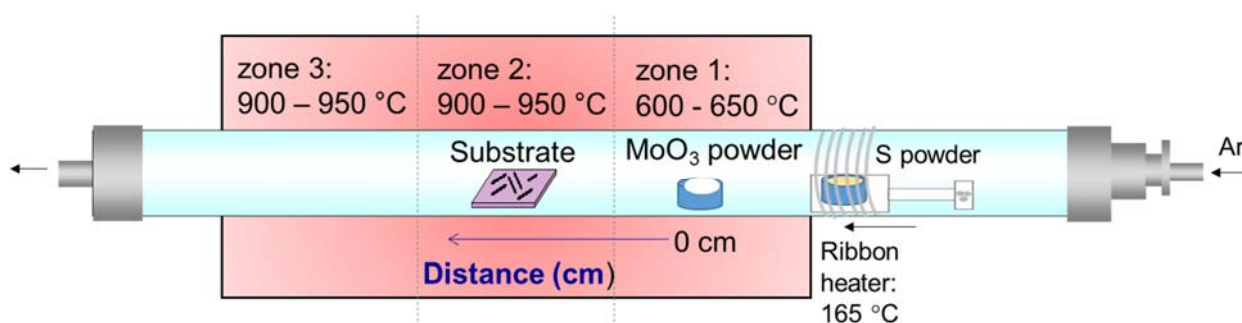


Figure S1. Schematic illustration of CVD setup to grow MoS₂/GNR heterostructure. Several reaction conditions have been studied such as MoO₃ temperature (600-650 °C), growth temperature, and distance of the GNR substrate from MoO₃ powder.

Characterizations

The as-grown GNR samples were studied by AFM (Bruker Nanoscope V), and then the GNRs were transferred onto a SiO₂ (300nm)/Si substrate by using a standard polymer-mediated transfer technique. The transferred GNRs were then characterized by AFM, SEM (Hitachi S-4800),

and confocal Raman spectroscope (Tokyo Instruments, Nanofinder 30) with a 532 nm excitation. The as-grown MoS₂/GNRs sample were measured by AFM, SEM, and Raman spectroscopy. TEM images were measured for the transferred MoS₂/GNRs on a Quantifoil grid using a JEOL 2100F at an acceleration voltage of 200 kV.

Back-gated-FET device measurement

Back-gated FET devices were fabricated with channel lengths between 200 nm to 1000 nm. The Si back plane and the SiO₂ layers are used as the back-gate electrode and gate dielectric, respectively. The EB lithography was used to pattern the source and drain contacts followed by metallization by electron beam evaporation of Au/Ti (40 nm/5 nm). Carrier transport properties were measured at room temperature under vacuum (typically 4×10^{-4} Pa) with a semiconductor parameter analyzer (Keysight Technologies B1500A). Photoinduced current modulation was measured by illuminating a device with a 532 nm laser beam at different power density. The typical power of the illuminating laser was 4.73 mW when an optical filter (optical density (OD)=1) was used (40 W/m²).

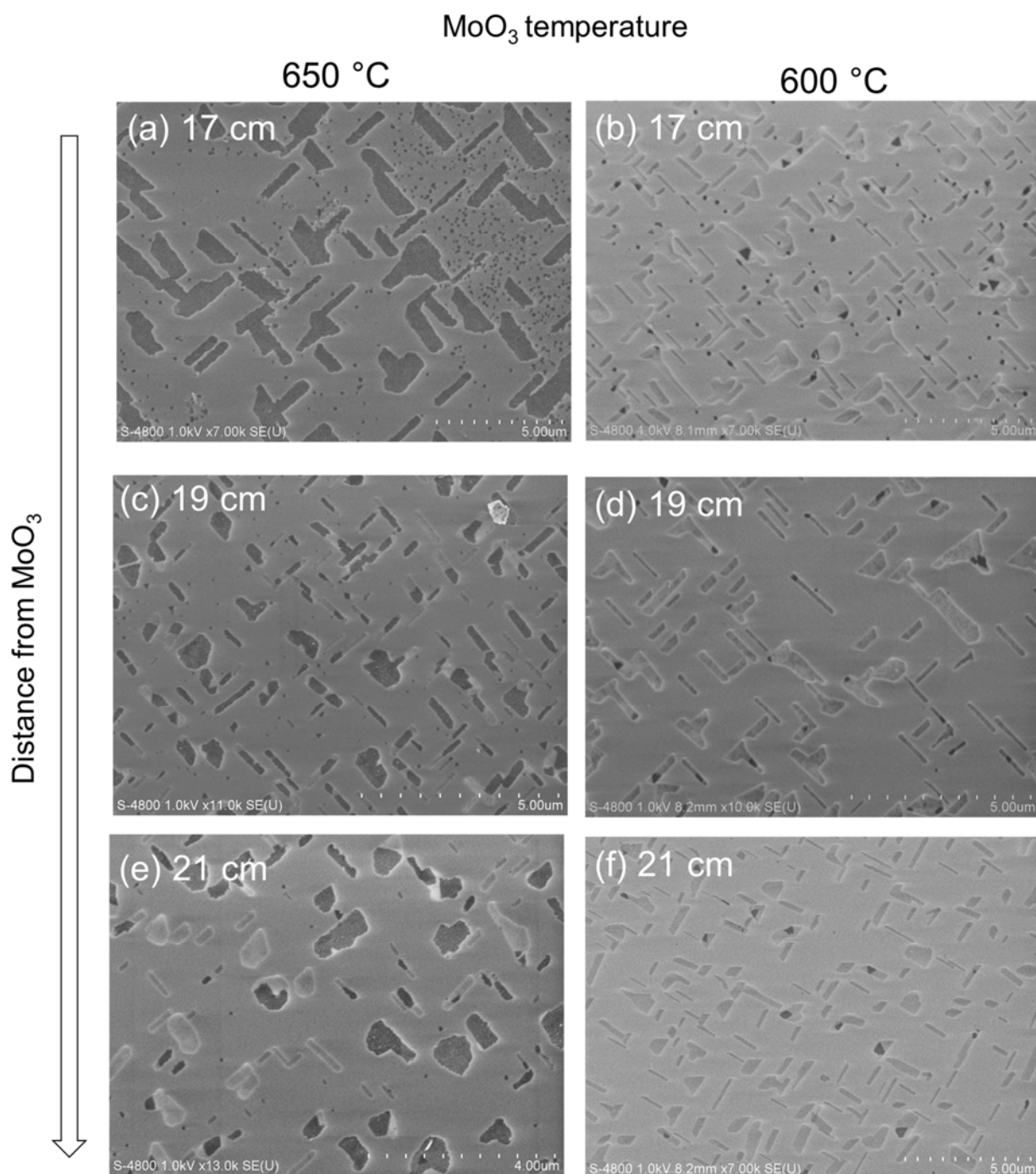


Figure S2. SEM images of GNRs with different coverage of MoS₂. (a-f) The MoS₂ coverage can be controlled by the distance between a GNRs/SiO₂ substrate and the MoO₃ precursor as well as by decreasing the MoO₃ temperature from 650 to 600 °C.

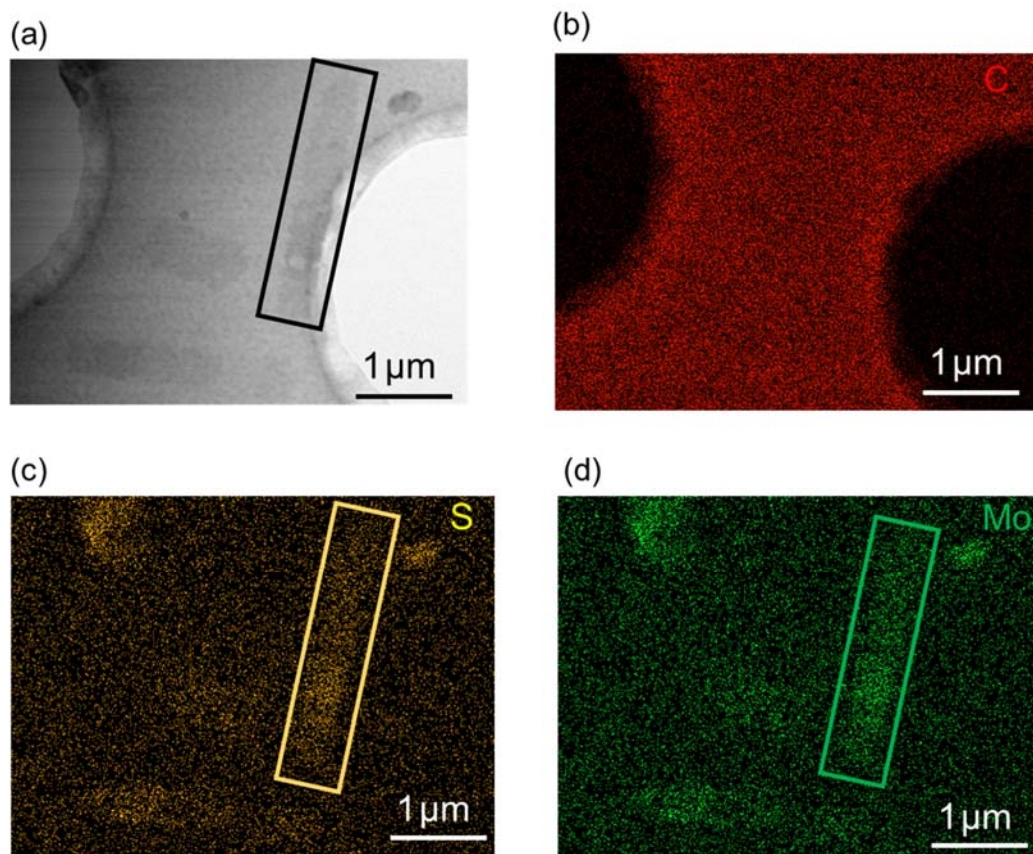


Figure S3. (a) TEM images of three MoS₂/GNRs deposited on a TEM grid. (b-d) EDX elemental maps for C, S, and Mo. By comparing the images (a-d), Mo and S are found locally at the positions marked by rectangular, while C is hard to observe from the color contrast due to the carbon supported grid used for the TEM grid. The signal of S and Mo atoms are very weak because of the thin MoS₂ layer deposited on GNR.

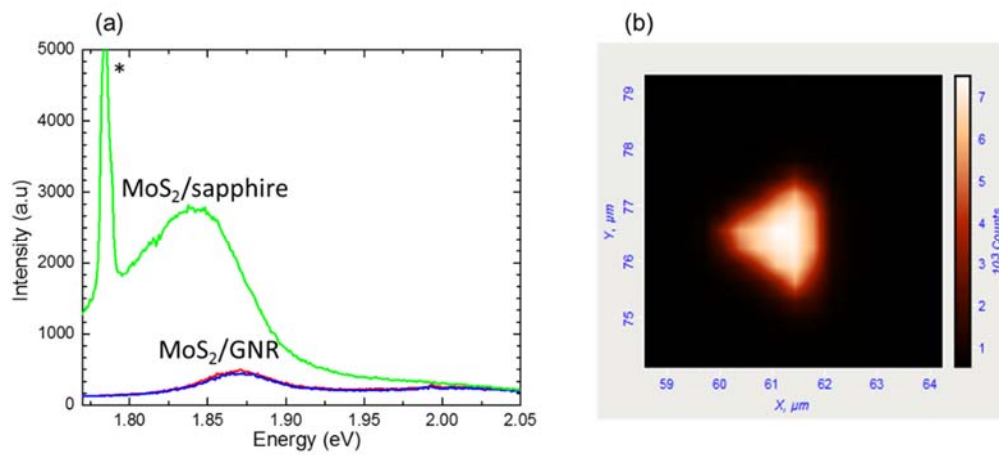


Figure S4. (a) Red and blue spectra: PL spectra of MoS₂ grown on GNR taken at the positions 1 and 2 marked in Figure 2. Green spectrum: PL from MoS₂ grown on a sapphire substrate. The PL quenching effect by GNR is observed. Asterisk (*) indicate a peak from the sapphire substrate. (b) PL mapping image of a single-layer MoS₂ domain grown on sapphire.