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Supporting Information

Enhanced Photovoltaic Performances of Graphene/Si Solar Cells by Insertion of MoS_2 Thin Film

Yuka Tsuboi[†], Feijiu Wang[†], Daichi Kozawa[†], Kazuma Funahashi[‡], Shinichiro Mouri[†], Yuhei Miyauchi[†], Taishi Takenobu[‡] and Kazunari Matsuda[†]*

[†]*Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan*

[‡]*Department of Advanced Science and Engineering, Waseda University, Shinjuku-ku, Tokyo 169-8555, Japan*

*Email: matsuda@iae.kyoto-u.ac.jp

S1. MoS_2 film fabrication

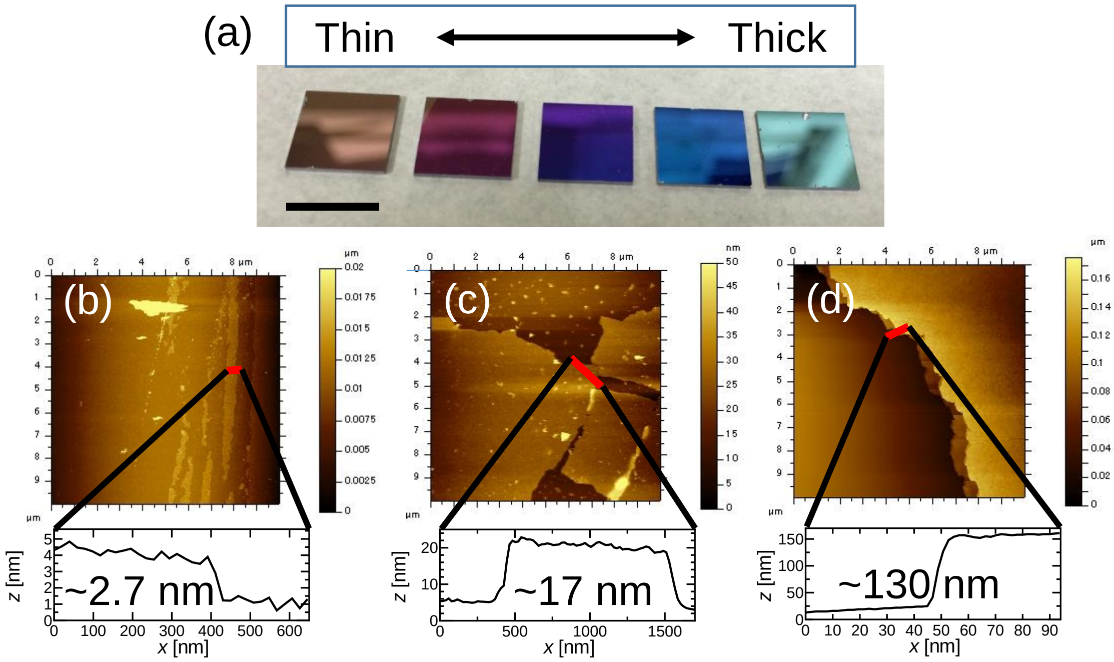

Figure S1. (a) Optical image of as-grown MoS_2 films with various layer thicknesses on SiO_2/Si substrates, as fabricated by the CVD method. The scale bar is 1 cm. The thickness of the MoS_2 films was controlled *via* the MoO_3 deposition time. We observed that MoS_2 film with a thickness of ~2/3 of the original MoO_3 film is fabricated by CVD process in the case of MoO_3 with an averaged thickness of 30 nm. The leftmost image is an SiO_2/Si substrate. (b)–(d) AFM image of an MoS_2 film. The height profiles are measured at the step edge between the MoS_2 film and the SiO_2/Si substrate. The thickness of the MoS_2 films was varied from 2.7 to 130 nm. For solar cells in Figure 3, we used films with ~17-nm thickness, as shown in (c).

S2. Process-flow of MoS_2 film transfer

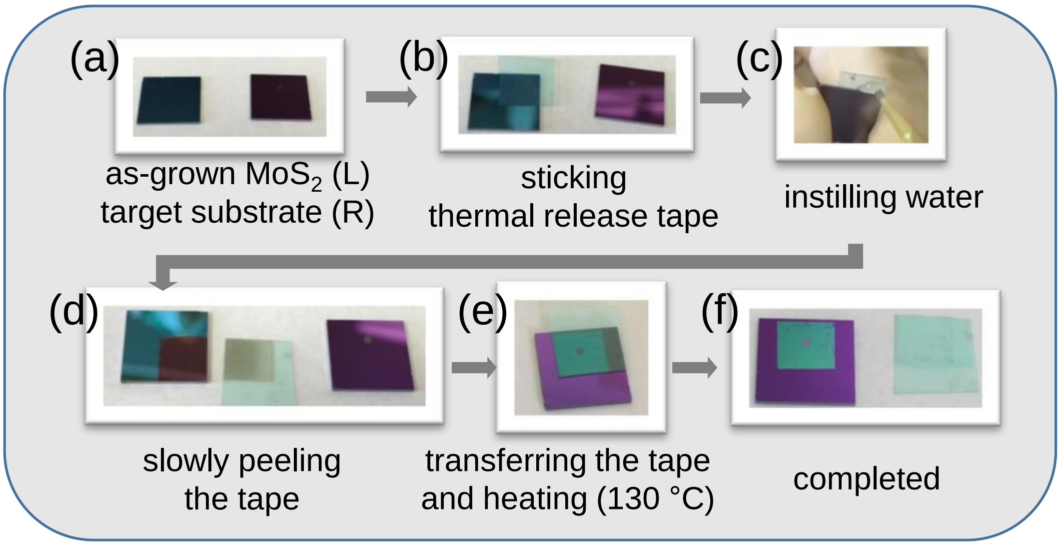

Figure S2. (a)–(f) The process-flow of as-grown MoS_2 film transfer. This method was conducted using thermal release tape (REVALPHA; No. 3195MS, Nitto Denko). The sticking MoS_2 film with the thermal release tape (left in (b)) was easily released by a drop of water, as shown in (c). After the tape with MoS_2 was transferred to the target substrate, the tape was removed by heating the substrate on a hotplate.

S3. Characterization of stacked graphene/MoS_2 film

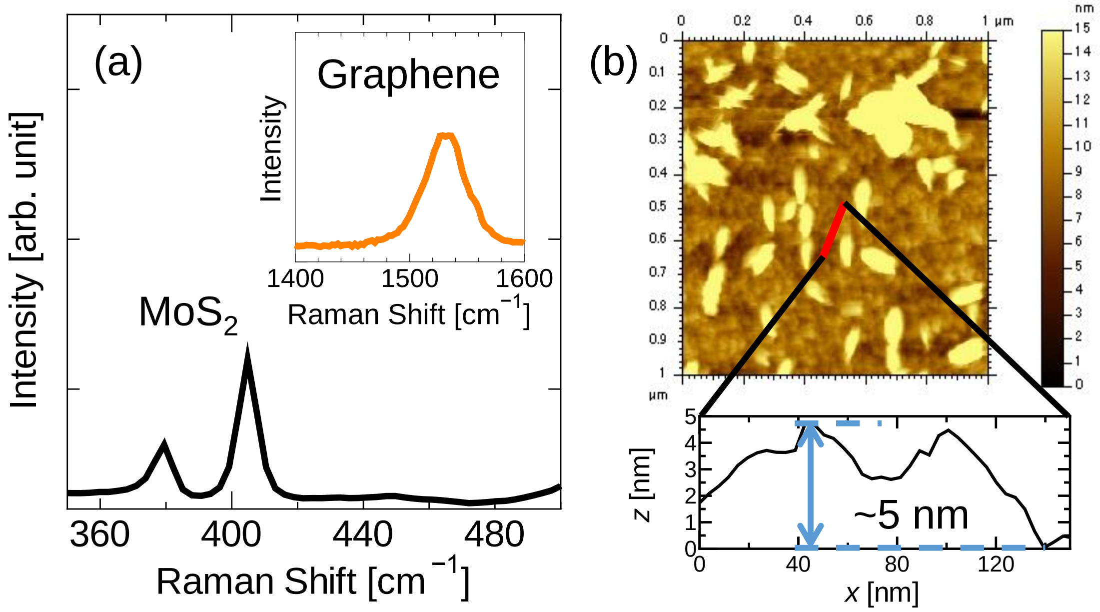

Figure S3. (a) Raman spectrum of the vertical stacking of graphene/MoS_2. (b) AFM image of a stacked graphene/MoS_2 film and its height profile along the red line in the image. The surface roughness of the graphene/MoS_2 film decreases in comparison with that of the MoS_2 film (Figure 2(a)).

S4. Photovoltaic parameters of graphene/MoS_2/*n*-Si solar cells

Cell Type	V_{OC} [V]	J_{SC} [mA/cm^2]	FF	η [%]	R_s [Ω·cm^2]
MLG/*n*-Si	0.38	6.76	0.17	0.44	17
MLG/MoS_2/*n*-Si	0.41	13.1	0.25	1.35	30
BLG/MoS_2/*n*-Si	0.51	21.4	0.55	5.98	11
TLG/MoS_2/*n*-Si	0.54	28.2	0.53	8.02	8.8

Table S1. Photovoltaic parameters of monolayer-graphene/*n*-Si and graphene/MoS_2/*n*-Si solar cells with monolayer-graphene (MLG), bilayer-graphene (BLG) and trilayer-graphene (TLG), as evaluated from experimentally obtained *J*–*V* curves. The open-circuit voltage V_{OC}, short-circuit current density J_{SC}, fill factor FF, photovoltaic efficiency η, and series resistance R_S are shown. Here, the R_S values were estimated from the slopes of the *J*–*V* curves.[S1]

S5. Transmission spectra and sheet resistance of graphene

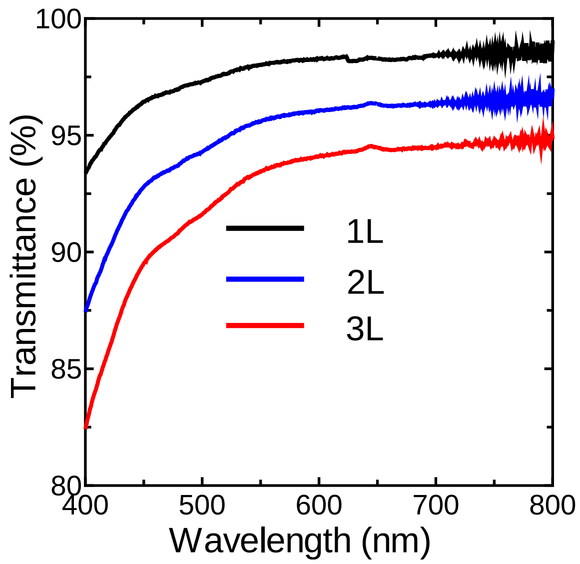

Figure S4. Transmission spectra of graphene films used in this study as a function of layer numbers.

Optical transmission spectra of graphene films used in this study are shown in Fig. S4. Sheet resistances of each sample were also measured by four-probe method. The sheet resistances drastically decreased from 41 kΩ/sq. in monolayer (1L), 4.9 kΩ/sq. in bilayers (2L), to 1.5 kΩ/sq. in trilayers (3L).

S5. The incident photon-to-current conversion efficiency (IPCE) spectrum of graphene/MoS_2/*n*-Si solar cells

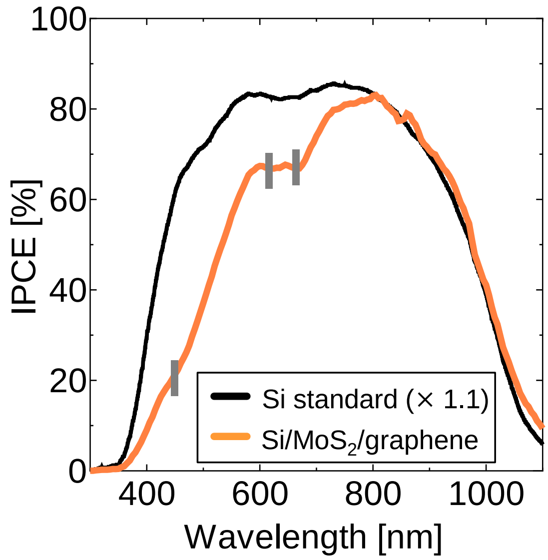

Figure S5. IPCE spectra of a trilayer-graphene/MoS_2/*n*-Si solar cell and a standard Si solar cell. The spectrum of the standard Si solar cell is normalized by the values at 800 nm. The dip structures were observed in the IPCE spectrum of graphene/MoS_2/*n*-Si in the shorter-wavelength region (400–700 nm). The positions of these dip structures are consistent with the absorption peaks of the MoS_2 film shown in Figure 2(b), which suggests that the generated carriers in the *n*-Si layer are the primary contributors to the photovoltaic current in the cell, whereas the MoS_2 functions as a shading layer at the point of light absorption.

S6. Carrier transport properties of an MoS_2 film, as measured by an electric double-layer transistor

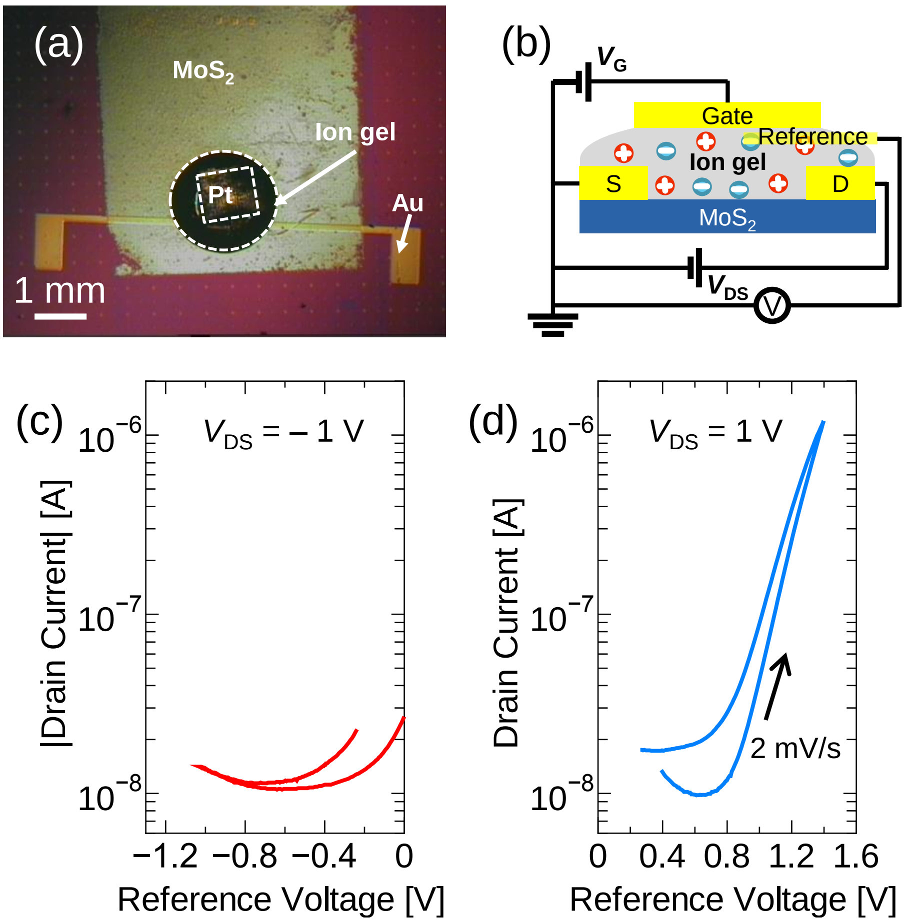

Figure S6. (a) Optical image of CVD-grown MoS_2 electric double-layer transistor and (b) A schematic illustration of the transistor, in which an ionic gel ([EMIM][TFSI] with PS-PMMA-PS) as a gate electrolyte, Pt as a top-gate electrode, and Ni/Au source and drain electrodes were used.[S2] (c)(d) Transfer characteristics measured by changing the voltage of the Pt top-gate electrodes. Current resulting from electron transfer was confirmed (blue line), whereas current resulting from hole transfer was not (red line).

Reference

S1. Miao, X.; Tongay, S.; Petterson, M. K.; Berke, K.; Rinzler, A. G.; Appleton, B. R.; Hebard, A. F., High Efficiency Graphene Solar Cells by Chemical Doping. *Nano Lett.* **2012**, 12, 2745-2750.

S2. Pu, J.; Yomogida, Y.; Liu, K.-K.; Li, L.-J.; Iwasa, Y.; Takenobu, T., Highly Flexible MoS_2 Thin-Film Transistors with Ion Gel Dielectrics. *Nano Lett.* **2012**, 12, 4013-4017.