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Monolayer MoS₂ field-effect transistors patterned by photolithography for active matrix pixels in organic light-emitting diodesHyeokjae Kwon¹, Sourav Garg², Ji Hoon Park¹, Yeonsu Jeong¹, Sanghyuck Yu¹, Seongsin M. Kim², Patrick Kung¹ and Seongil Im¹

Two-dimensional molybdenum disulfide (MoS₂) has substantial potential as a semiconducting material for devices. However, it is commonly prepared by mechanical exfoliation, which limits flake size to only a few micrometers, which is not sufficient for processes such as photolithography and circuit patterning. Chemical vapor deposition (CVD) has thus become a mainstream fabrication technique to achieve large-area MoS₂. However, reports of conventional photolithographic patterning of large-area 2D MoS₂-based devices with high mobilities and low switching voltages are rare. Here we fabricate CVD-grown large-area MoS₂ field-effect transistors (FETs) by photolithography and demonstrate their potential as switching and driving FETs for pixels in analog organic light-emitting diode (OLED) displays. We spin-coat an ultrathin hydrophobic polystyrene layer on an Al₂O₃ dielectric, so that the uniformity of threshold voltage (V_{th}) of the FETs might be improved. Our MoS₂ FETs show a high linear mobility of approximately $10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, due to a large grain size around $60 \mu\text{m}$, and a high ON/OFF current ratio of 10^8 . Dynamic switching of blue and green OLED pixels is shown at $\sim 5 \text{ V}$, demonstrating their application potential.

npj 2D Materials and Applications (2019)3:9; <https://doi.org/10.1038/s41699-019-0091-9>

INTRODUCTION

Molybdenum disulfide (MoS₂), a two-dimensional (2D) transition metal dichalcogenide (TMD) semiconductor has attracted much attention from many researchers owing to its interesting physical properties and potentials for future nanoscale electronics.^{1–4} Like graphene, those 2D semiconductors are easily formed by mechanical exfoliation using scotch tapes in general, while it also displays high ON/OFF current ratio and good subthreshold swing in a field-effect transistor (FET) form with n -type conduction unlike graphene, which shows no bandgap.^{5–14} Since 2D MoS₂ obtained by mechanical exfoliation limits the length scale only to a few micrometers, researchers have studied on how to fabricate such 2D MoS₂ in a large scale,^{15–20} which would open the gate toward more practical applications enabling conventional photolithographic device/circuit patterning. Chemical vapor deposition (CVD) has thus become one of mainstream fabrication techniques to achieve large-scale MoS₂ 2D sheets. Many of success on large-scale MoS₂ monolayer sheets have been reported.^{21–23} Yet, reports on conventional photolithographic patterning for large-scale 2D MoS₂-based devices with decently high mobilities are still rare,^{24–26} to the best of our limited knowledge. For instance, CVD-grown monolayer MoS₂ FETs might have never been used for both switch and driver of organic light-emitting diodes (two FETs in an OLED pixel for active matrix: see the circuit details in Supporting Information Figure S1), while exfoliated TMD-based FETs were barely used once with poor yield as a proof-of-concept-type demonstration.²⁷ According to previous works, low threshold voltage would be an important issue for OLED pixel switching.^{26,27}

In the present work, we have fabricated CVD-grown large-scale MoS₂ FETs with patterned bottom gate and investigated their applications toward analog OLED pixel as low voltage switching and high current driving FETs. In particular, we successfully spin-coated an ultrathin hydrophobic polystyrene (PS) polymer layer on Al₂O₃ dielectric before the CVD MoS₂ monolayer was transferred onto the gate-patterned Al₂O₃ on glass, so that the uniformity of threshold voltage (V_{th}) of FETs might be improved. Our MoS₂ FET shows a linear mobility of $\sim 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and an ON/OFF current ratio of 10^8 as patterned by photolithography on glass, and those values are regarded quite high or comparable to those of previous CVD MoS₂ FETs^{28–32} because a single grain of our CVD MoS₂ monolayer appears as large as $\sim 60 \mu\text{m}$ in side dimension. Among 48 MoS₂-based FETs realized from the same CVD MoS₂ wafer, 38 devices successfully work (yield $\sim 80\%$) operating at $\sim 5 \text{ V}$ with 50 nm-thick Al₂O₃ bottom gate dielectric. Dynamic/static switchings of blue and green OLED pixels were nicely demonstrated using the CVD monolayer MoS₂ FETs.

RESULTS AND DISCUSSION

According to scanning electron microscope (SEM) images of Fig. 1a–c, sufficiently large area and continuous MoS₂ monolayer film was achieved (Fig. 1b). For the CVD growth, we used a single precursor source consisting of MoS₂ powder at a high growth temperature $\sim 800 \text{ }^\circ\text{C}$ with a temperature of $900 \text{ }^\circ\text{C}$ for the powder precursor,³³ instead of the combination of MoO₃ and S, which is more often reportedly used (see Methods section for more details).^{15–17} From a more magnified view of the film region, a grain boundary is distinguished better as indicated by dashed

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Received: 27 August 2018 Accepted: 28 January 2019

Published online: 19 February 2019

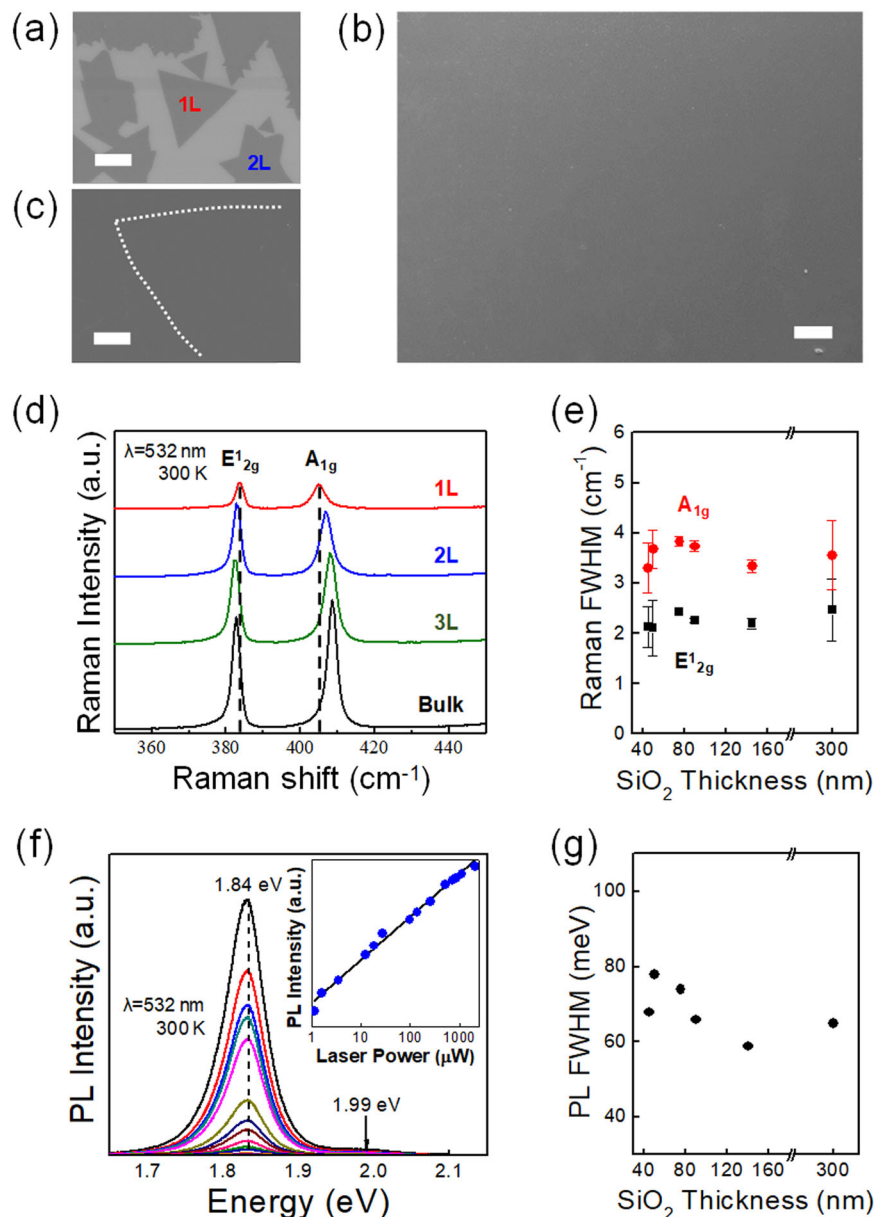


Fig. 1 Scanning electron microscope (SEM) image of chemical vapor deposition (CVD)-grown MoS₂. **a** Triangular shape MoS₂ located near periphery, **b** whole area-covering MoS₂ monolayer near central region. Each scale bar in **a** is 20 μm and in **b** is 200 μm . **c** Grain boundary is indicated by dashed line. Scale bar is 20 μm . **d** Raman spectra of 1L, 2L, and 3L CVD MoS₂, and bulk. **e** Raman peak full width at half maximum (FWHM) of 1L MoS₂ grown on differently thick SiO₂. **f** PL spectra of 1L CVD MoS₂ under varied laser power (inset). **g** Photoluminescence (PL) spectra FWHM plot obtained from varied SiO₂ thickness

outline in Fig. 1c. The average individual grain size is 2000 μm^2 . At the periphery of the substrate, characteristic triangular-shaped monolayer MoS₂ with side dimensions from 30 to 60 μm were observed (Fig. 1a). In Fig. 1a we can also see how the monolayers start to join to ultimately form the continuous film. These results were obtained from all the thermally oxidized SiO₂/Si substrates we used. Continuous film area approaches to a few mm². Micro-Raman spectroscopy was used to confirm the monolayer nature of the MoS₂ grown as the separation between the E¹_{2g} and A_{1g} Raman peaks is known to be characteristic of the number of MoS₂ layers.^{34–36} The spectra obtained over the film regions (Fig. 1b) are shown in the red “1L” curve of Fig. 1d, with a peak separation of 20.2 cm^{-1} . For further confirmation, we have put the spectra for bilayer (2L, blue, 23.2 cm^{-1} peak separation) and trilayer (3L, green, 24.2 cm^{-1} peak separation) MoS₂ spots that we could sometimes find at the very outer periphery of the substrates,

where MoS₂ nucleation might not be so controllable resulting in more vertical growth (supporting photo image of the outer periphery is seen in Figure S2). We also included the Raman spectrum from bulk MoS₂ powder (black line), which exhibited a 25.1 cm^{-1} peak separation. To assess the quality of the MoS₂ monolayer films,^{37–39} we plot in Fig. 1e the full width at half maximum (FWHM) of the E¹_{2g} and A_{1g} Raman peaks for the material grown on the various thicknesses of SiO₂. The FWHM was essentially independent of the oxide thickness, staying in the range of 2–2.5 and 3.5 cm^{-1} for the E¹_{2g} and A_{1g}, respectively. For comparison, FWHM values reported to date for the E¹_{2g} and A_{1g} peaks of CVD-grown monolayer have been in the ranges of 3.5–4.2 and of 5–7 cm^{-1} respectively.^{35,40,41} Furthermore, MoS₂ monolayers exfoliated from geological materials exhibit a FWHM of 3.7 cm^{-1} for E¹_{2g} peak in report.⁴² Therefore, the FWHM data we obtained may indicate that our CVD-grown monolayer MoS₂ is

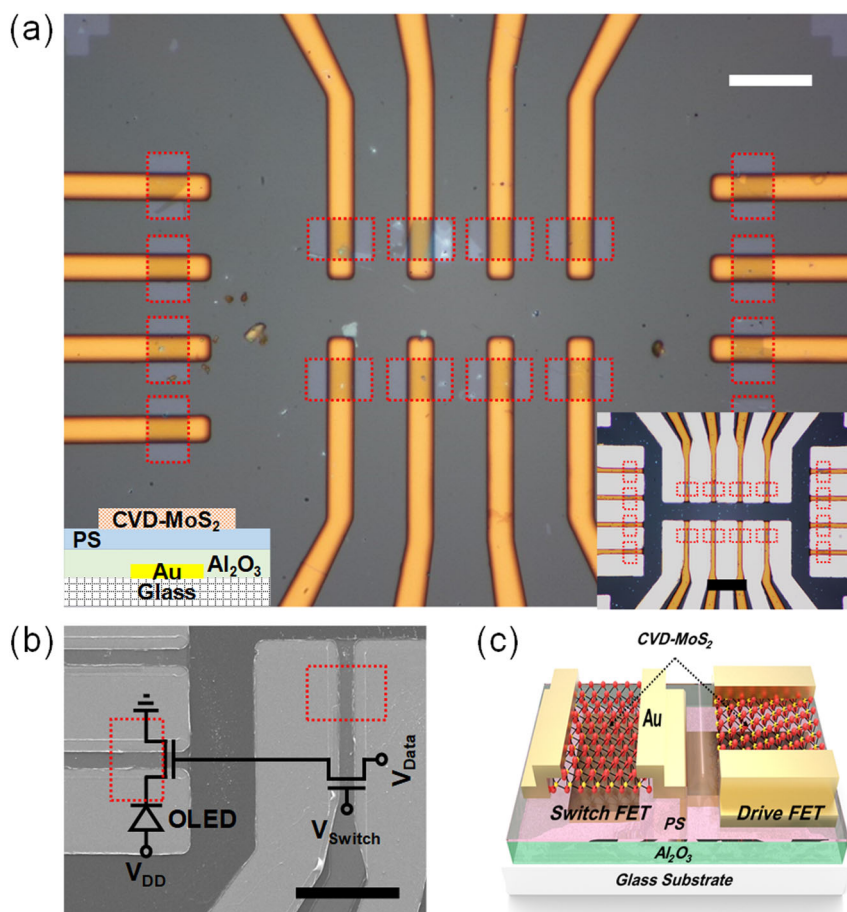


Fig. 2 **a** Optical microscope (OM) images of patterned chemical vapor deposition (CVD)-MoS₂ channels (red dotted rectangles) on patterned bottom gate (left inset for cross section) and OM images of field-effect transistors (FETs; right inset with patterned Au S/D electrode). **b** Scanning electron microscope (SEM) image of switching and driver FETs for an organic light-emitting diode (OLED) pixel. Circuit for an OLED pixel is overlaid on SEM image. **c** Schematic three-dimensional (3D) view of those FETs for the OLED pixel circuit. Hydrophobic polystyrene (PS) layer on oxide dielectric is worth to note in our FETs (schematic 3D and cross-sectional two-dimensional). All scale bars are 20 μm

comparable to or potentially better in crystalline quality than reported ones.⁴³ We might attribute such good crystalline quality along with large-scale continuous monolayer MoS₂ to a high growth temperature over ~800 °C with a temperature of 900 °C for the powder precursor, since the higher temperature enhances surface diffusion of adatoms and lateral crystal growth, and therefore smooth coalescence into a continuous film. The optical properties of our MoS₂ monolayer films were also characterized using power-dependent photoluminescence (PL) measurements at room temperature, as shown in Fig. 1f. A strong emission related to the A-exciton in MoS₂ was observed at 1.84 eV. The B-exciton-related peak could also be distinguished at 1.99 eV. The peak separation of 150 meV is due to the splitting of valence band.^{2,42,44} The peak positions did not shift with exciton power. The inset of Fig. 1f shows that the luminescence emission intensity (I_{PL}) follows a power relationship with incident laser excitation power (I_L): $I_{\text{PL}} = nI_L^a$.⁴⁵ The exponent a indicates the type of radiative recombination process the PL originates from, with an exponent of 1 corresponding to free exciton recombination. In our samples, a slope of 0.79 (as a) was obtained, which indicates the recombination is close to excitonic although there is some degree of defect-related recombination.^{46,47} We have also compared the FWHM of the main luminescence peak for the MoS₂ films grown on the various thicknesses of SiO₂ as shown in Fig. 1g. The FWHM was found to range from 60 to 80 meV, which is close to the best reported value (60 meV) to date (50 meV is for the suspended MoS₂ monolayer²).¹⁵

The large-scale MoS₂ monolayer was transferred onto a 50 nm-thick Al₂O₃ dielectric, beneath which a patterned Au gate was located already. Our MoS₂ was then aligned and patterned on the gate as channels by conventional photolithography including O₂ plasma dry etching. Figure 2a displays such patterned MoS₂ channels (red dotted-line rectangles) and gates in optical microscope (OM) images along with a schematic cross section (left inset) while completed devices (16 FETs) with Au S/D electrodes are also shown in the right inset as another OM image. As stated in the introduction part, we particularly spin-coated an ultrathin hydrophobic PS layer on Al₂O₃ dielectric before the transfer of CVD MoS₂ monolayer, expecting and targeting a relatively uniform V_{th} distribution of MoS₂ FETs. But we also fabricated another set of MoS₂ FETs with bare Al₂O₃ without PS layer, so to compare the two types of FET devices with and without PS layer. Capacitance-voltage (C-V) measurements were thus conducted for two different dielectrics (see Figure S3 for the values of 8.46 nm PS/50 nm Al₂O₃ and 50 nm Al₂O₃). In addition, another PS layer was utilized as a technical sacrificial cover to transfer monolayer MoS₂ during these device fabrications, and this second PS layer is peeled-off from MoS₂ after transferring onto dielectric, while the ultrathin PS layer on Al₂O₃ stays firm because it covalently bonded on Al₂O₃ surface (see fabrication details in Methods section and Supporting Information, Figure S4.) According to the OM images of channels in Fig. 2a, most of the monolayer channels appear clean without any distinguishable specks except a few channels, which might contain nonuniform

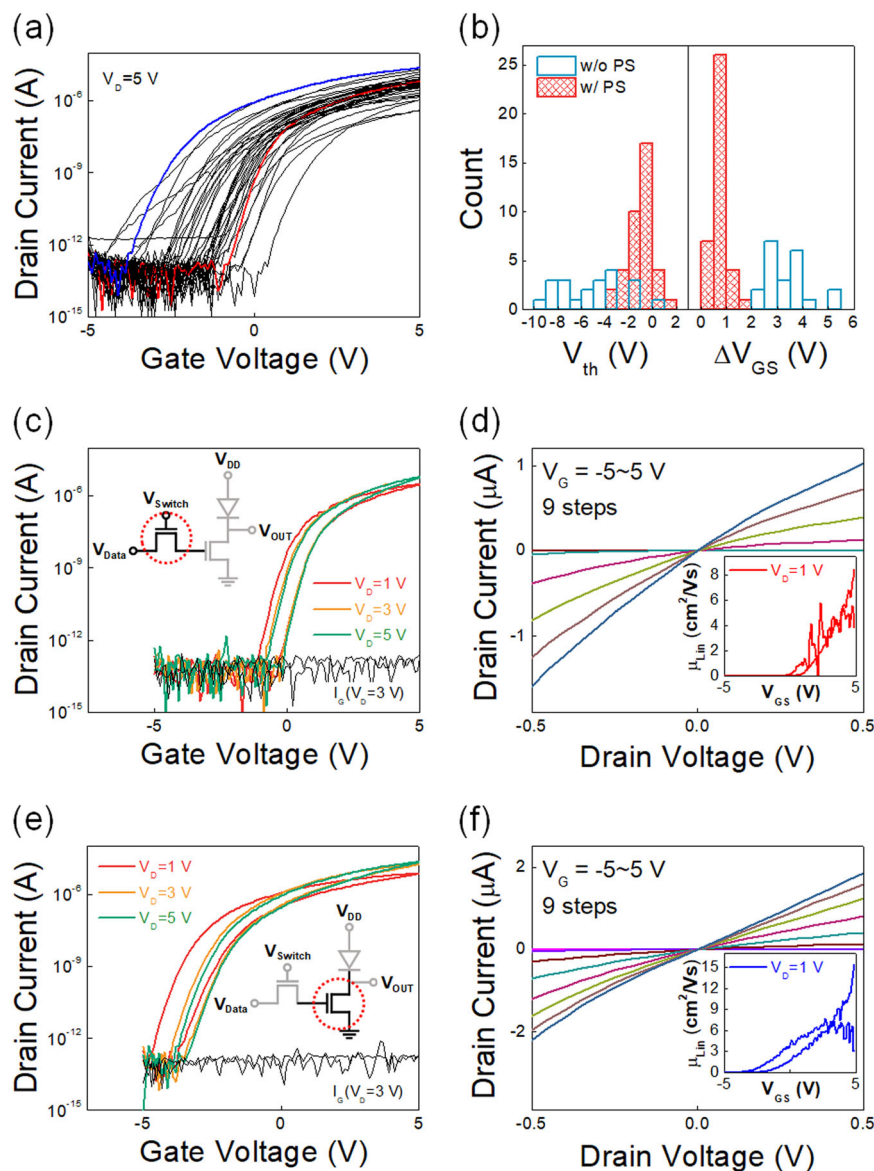


Fig. 3 **a** Transfer curves of total 38 chemical vapor deposition (CVD) MoS₂ field-effect transistors (FETs) with polystyrene (PS) layer obtained at $V_D = 5$ V (without including hysteresis). Blue and red curve FETs were selected for driver and switching devices in the pixel operation. **b** Histograms of the V_{th} and ΔV_{GS} from 38 FETs (w/ PS) and 22 FETs (w/o PS). **c** Transfer curves of switching driver FETs obtained at $V_D = 1, 3,$ and 5 V (including hysteresis). Switching FET is indicated in the inset organic light-emitting diode pixel circuit by red dotted circle. **d** Output curve of switching FET and inset linear mobility plot. **e** Transfer curves of driver FET obtained at $V_D = 1, 3,$ and 5 V (including hysteresis). Driver FET is indicated in the inset circuit by red dotted circle. **f** Output curve of driver FET and inset linear mobility plot

thickness (bilayer or trilayer) regions. Figure 2b is an SEM image from two MoS₂ FETs with S/D electrodes as example devices for switch and driver of an OLED pixel circuit (as described in the overlaid inset circuit). The red rectangles in Fig. 2b identify the channel area as partially covered by S/D region while Fig. 2c illustrates the same circuit as a three-dimensional drawing.

As we fabricated 48 MoS₂ FETs with PS layer (type 1, FET with PS/Al₂O₃ bilayer for gate dielectric) and 32 FETs without PS layer (type 2, FET with Al₂O₃ only), 38 FETs operated among total 48 while 22 FETs appeared alive among total 32 devices (~80% vs. ~70% in yield for type 1 vs. type 2). Figure 3a shows drain current-gate voltage (I_D - V_{GS}) transfer characteristics of such 38 FETs with PS layer (type 1), and the transfer curves from 22 FETs without PS (type 2) were put to Supporting Information, Figure S5a and b. We compare device uniformity between FETs with PS layer and without in Fig. 3b, where the histograms of V_{th} and gate hysteresis (ΔV_{GS}) are plotted as obtained from those two types of FETs. The V_{th} distribution

of FETs with PS layer appears quite uniformly centered (near -1 V) while that of FETs without is relatively scattered, and the ΔV_{GS} of the former devices appears mostly smaller (as ~ 1 V in average) than that of the latter (approximately more than 3 V). Failed or low-performance FETs seem to originate from the transfer failure of MoS₂ monolayer, which includes tearing and possible wrinkling during transfer whether on PS/Al₂O₃ or bare Al₂O₃ surface. It is, however, regarded that the hydrophobic PS layer surface certainly improves the device uniformity and causes hysteresis reduction due to trap density reduction at the MoS₂ channel/dielectric interface. Among the 48 FETs of Fig. 3a, we selected two FETs for switch and driver, which have red and blue curves, respectively for active OLED pixel operation. Figure 3c shows transfer characteristics of switching MoS₂ FET (inset circuit), where ΔV_{GS} (~ 1 V) and V_{th} (~ -1 V) of the device are displayed along with ON I_D current of a few μ A and high ON/OFF I_D ratio of $\sim 10^8$ at a drain voltage (V_{DS}) of 1, 3, and 5 V. Gate leakage was ~ 100 fA only. Output (I_D - V_{DS})

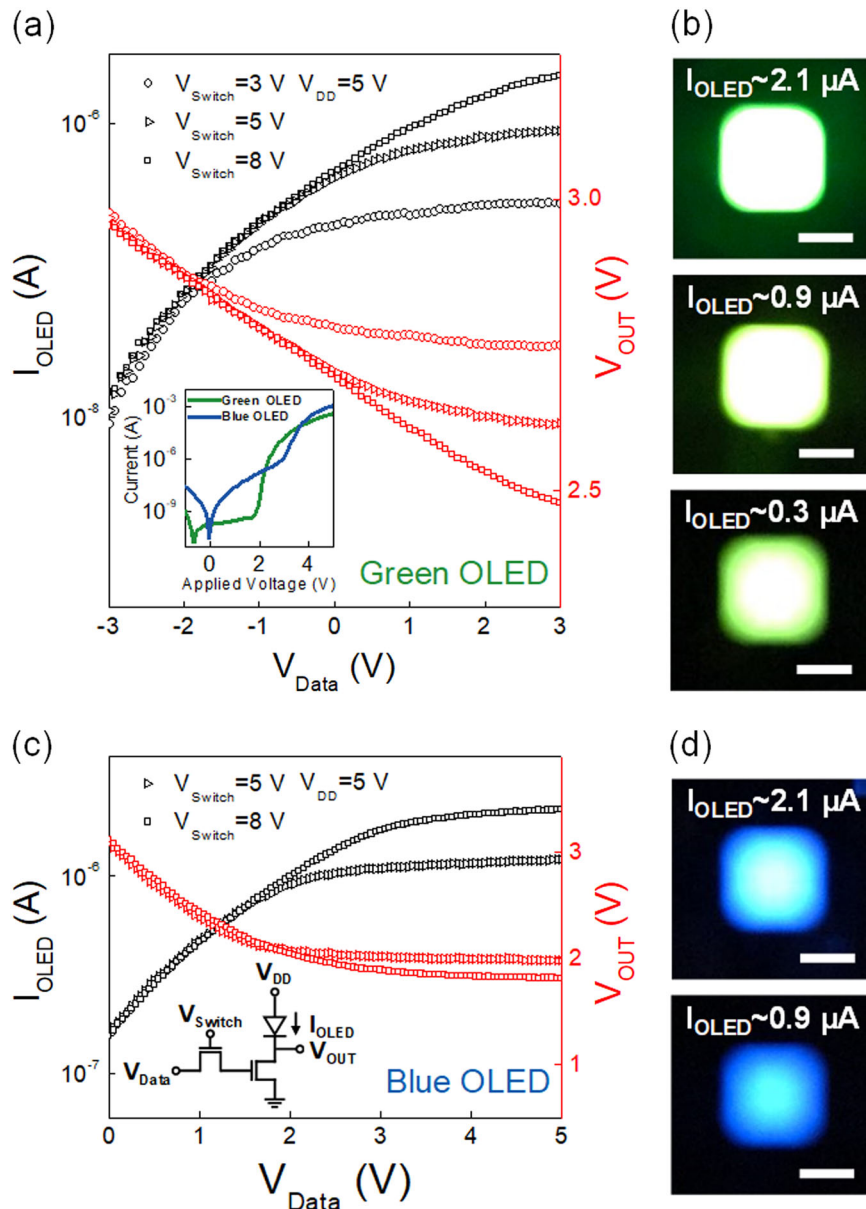


Fig. 4 **a** Organic light-emitting diode (OLED) current (left y-axis) and output voltage (right y-axis) plots obtained from the pixel circuit by V_{Data} sweep for green OLED (under $V_{\text{Switch}} = 3, 5,$ and 8 V). Inset I - V curves were obtained from green and blue OLED diodes. **b** Each V_{Switch} condition causes different emission intensity and I_{OLED} . **c** I_{OLED} and V_{OUT} plots obtained from the pixel circuit by V_{Data} sweep for blue OLED ($V_{\text{Switch}} = 5$ and 8 V). Inset pixel circuit is seen for analog OLED pixel operation. **d** Each V_{Switch} condition causes different blue emission intensity and I_{OLED} . All scale bars are $50\ \mu\text{m}$

characteristics of the switching device are shown in Fig. 3d where ohmic behavior between Au and S/D is confirmed. Inset linear mobility plot of Fig. 3d shows that the average mobility of switching FET is as high as $\sim 8\text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. On the one hand, from the type 1 FET group in Fig. 3a we chose another device (blue transfer curve) for driving FET whose V_{th} is a little deviated to more (–) direction from central region. Our selection was according to a natural expectation that this FET would draw more current for OLED operation, although such V_{th} deviation may be undesirable in producing uniform devices. This deviation may come from bilayer- or trilayer-containing MoS_2 channel. Figure 3e displays the transfer characteristics of driving MoS_2 FET (inset circuit), where ΔV_{GS} ($\sim 1\text{ V}$) and V_{th} ($\sim -4\text{ V}$) of the device are shown along with high I_{D} current of more than $5\ \mu\text{A}$ at 5 V of V_{DS} and high ON/OFF I_{D} ratio of $\sim 10^8$ at $1, 3,$ and 5 V of V_{DS} . Output characteristics of the driver FET are quite similar to those of

switching device as shown in Fig. 3f. Inset mobility plot of Fig. 3f shows the linear mobility of driving FET as $\sim 15\text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. In fact, maximum mobility of $\sim 15\text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and average of $8\text{--}10\text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ were achieved from our type 1 FETs. Those decently high mobility values might come from large MoS_2 grain size ($\sim 60\ \mu\text{m}$), which is relatively larger than channel length ($L = 4.5\ \mu\text{m}$) and width ($W = 10\ \mu\text{m}$).

Based on the device performances of the switching and driving FETs, an analog pixel operation for a single OLED was attempted with a circuit (the inset of Fig. 4c) under voltage conditions: a fixed supplied voltage (V_{DD}), varied switching voltages (V_{Switch}), and varied data voltages (V_{Data}). Figure 4a shows OLED current (I_{OLED}) and output voltage (V_{OUT}) with respect to V_{Data} sweep, which was taken to turn on a green OLED (inset displays I - V curve characteristics of green and blue OLED). It is no doubt that high V_{Switch} ($= 8\text{ V}$) and V_{Data} ($= 3\text{ V}$) cause (or drive) high I_{OLED} with most

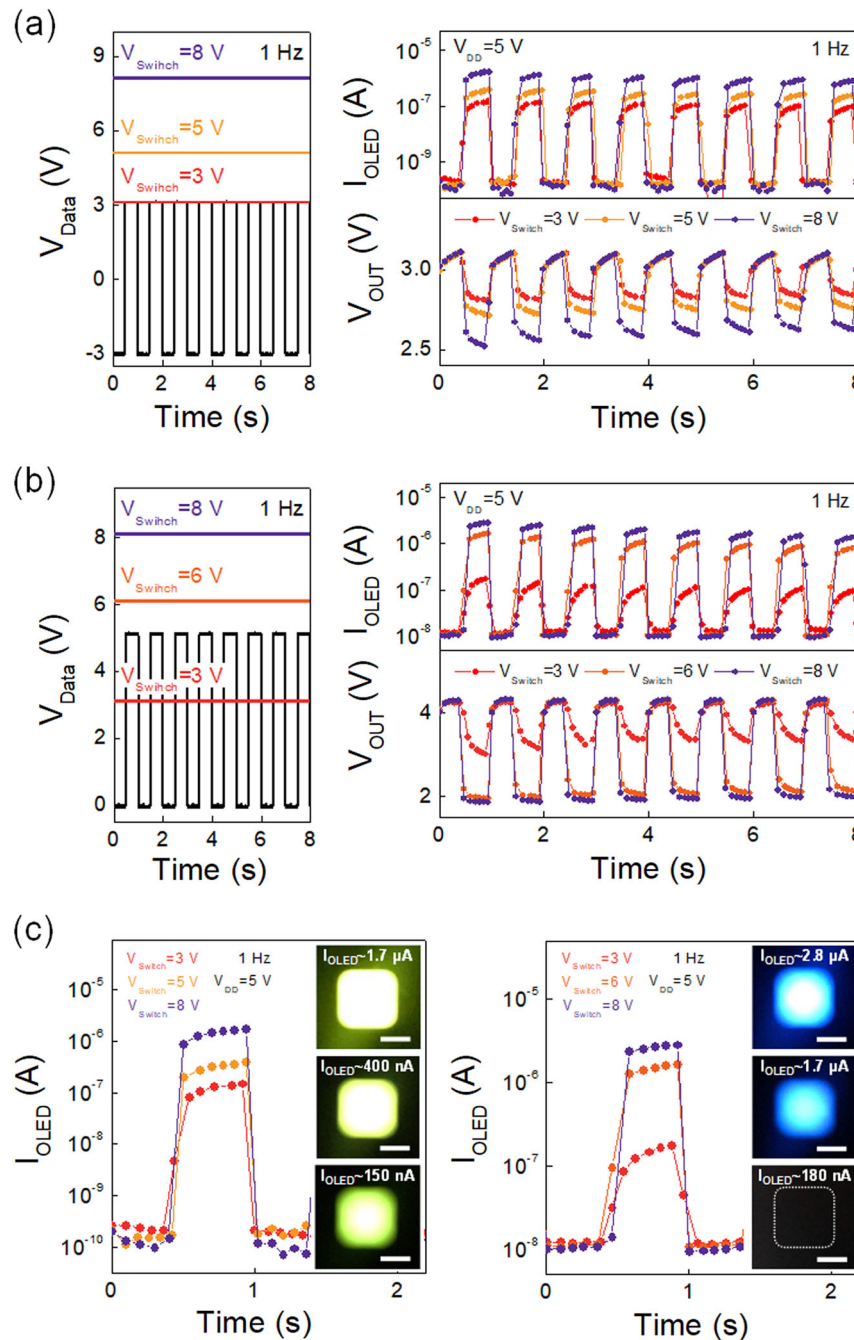


Fig. 5 Time domain plots of organic light-emitting diode (OLED) current and output voltage. **a** For ON (light) and OFF (dark) switching in the pixel circuit of green OLED, V_{Data} was modulated between -3 and 3 V at 1 Hz under three different V_{Switch} = conditions 3 , 5 , and 8 V. **b** For ON and OFF switching in the blue OLED pixel circuit, V_{Data} was modulated between 0 and 5 V at 1 Hz under three different V_{Switch} = conditions 3 , 6 , and 8 V. **c** Dynamic emission properties of green vs. blue OLED pixels. Even under smaller I_{OLED} current, green light appears brighter than blue emission. Under $V_{\text{Switch}} = 3$ V, blue OLED cannot be turned on. All scale bars are $50 \mu\text{m}$

intense brightness among three different pixel displays of Fig. 4b. Medium and low I_{OLED} are also displayed with V_{Switch} of 5 and 3 V showing their respective brightness in Fig. 4b. The value of $V_{\text{DD}} - V_{\text{OUT}}$ matches with the applied voltage to the OLED pixel itself. Similar behavior of analog pixel operation was observed from a single blue OLED with two different switching voltages of 5 and 8 V as shown in Fig. 4c, d. However, it is recognized from blue OLED pixel operation that we might need higher V_{Switch} to brighten up the blue light than that for the case of green OLED. According to Fig. 4d, I_{OLED} values of blue OLED are higher than

those of green OLED for V_{Switch} ($=8$ and 5 V) while blue lights appear much dimmer than green lights.

Dynamic ON/OFF switching of OLED pixels was attempted as shown in Fig. 5a, b, which are according to their static light-emitting performances in Fig. 4a–d. Figure 5a shows the I_{OLED} and V_{OUT} dynamics in green OLED pixel circuit as obtained by temporally modulating V_{Data} (between -3 and 3 V at 1 Hz). ON and OFF switching was observed from all the switching voltages ($=3$, 5 , and 8 V), and of course, the green emission intensity appears distinguishable according to each switching voltage. For blue OLED pixel operation of Fig. 5b, almost the same dynamics

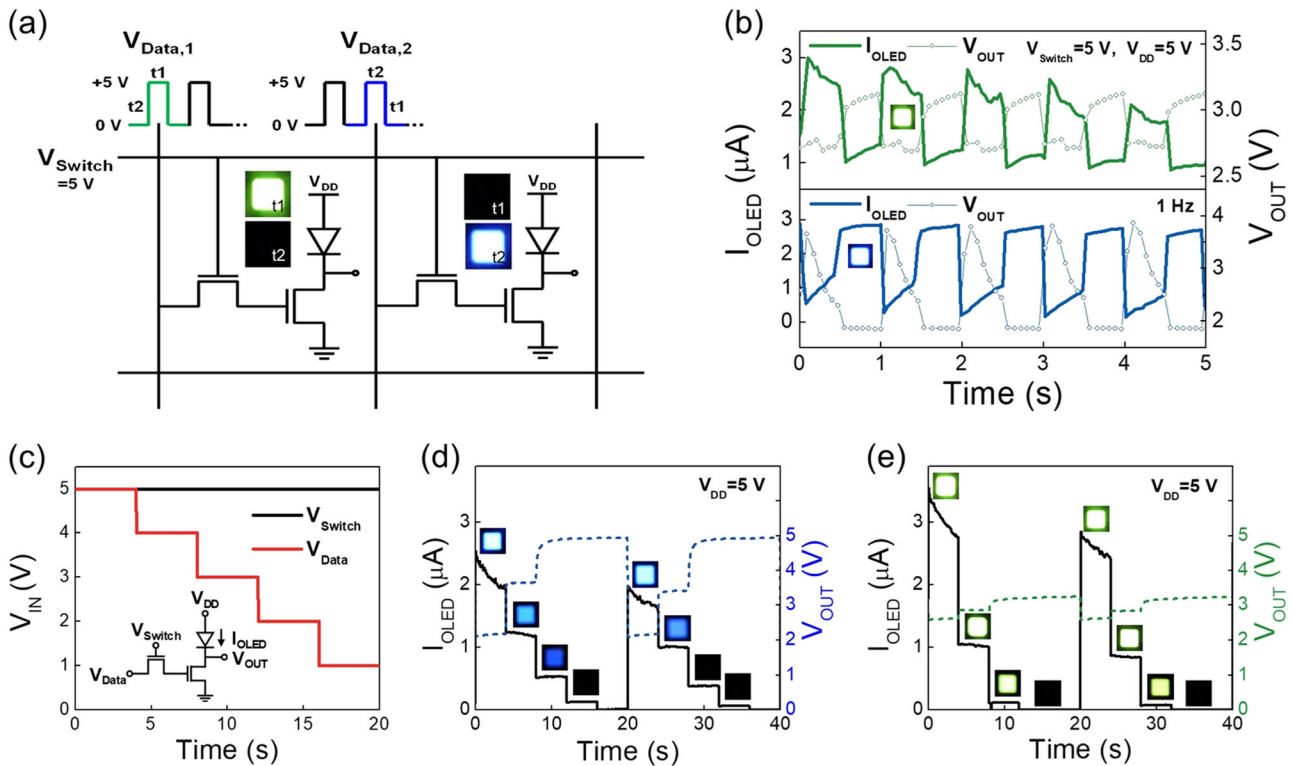


Fig. 6 **a** Circuitry for digital alternate switching of 2×1 two-color organic light-emitting diode (OLED) pixel array, and **b** $I_{\text{OLED}}/V_{\text{OUT}}$ dynamics for each color OLED under constant $V_{\text{DD}} = V_{\text{Switch}} = 5$ V, periodic $V_{\text{Data},1} = 5$ and 0 V ($V_{\text{Data},2} = 0$ and 5 V, synchronously). **c** Time- V_{IN} design plot for grayscale analog pixel switching. **d, e** Time domain I_{OLED} and V_{OUT} results for grayscale analog OLED pixel (blue and green)

was expected and observed from the circuit, however, in fact a low V_{Switch} of 3 V could not turn on the blue light at its I_{OLED} ($\sim 0.18 \mu\text{A}$) while 6 and 8 V turn on the OLED without problem. Based on the I_{OLED} results of Fig. 5a, b, dynamic OLED emissions for green and blue lights are displayed in Fig. 5c for more detailed comparison between the two OLEDs. According to Fig. 5c, the blue light appears completely dark at I_{OLED} ($\sim 0.18 \mu\text{A}$) and 3 V switching voltage while the green emission is still very bright at the same voltage and even lower I_{OLED} ($\sim 0.15 \mu\text{A}$). Such emission difference in both dynamic and static operations comes from their respective emission efficiencies since green OLED emission displays a higher efficiency than blue ones in general. On the one hand, an obvious delay in V_{OUT} dynamics (Fig. 5b) for blue emission was observed under 3 V of V_{Switch} . It is probably because I - V curve characteristics of blue OLED is different from that of green OLED. According to the inset plots of Fig. 4a, blue OLED should have higher resistance (dV/dI) in the range of 1–3 V than green OLED device. Such higher resistance (R) might cause longer RC delay under the same parasitic capacitance (C) condition. Nevertheless, it is regarded that our CVD MoS_2 monolayer channel FETs overall show strong potentials to function as the gate switch and I_{OLED} driver for analog OLED pixel circuits.

Based on above switching dynamics results from Fig. 5a–c, we further conducted two types of more advanced dynamic switching: digital switching of 2×1 OLED pixel array and grayscale analog switching of single blue/green OLED pixel. These demonstrations were carried out under fixed V_{DD} and V_{Switch} ($V_{\text{DD}} = V_{\text{Switch}} = 5$ V), which appears to be a proper condition for pixel display in Fig. 5c when two alternating V_{Data} pulses are used between 0 and +5 V. Figure 6a shows 2×1 OLED pixel array operation employing four CVD MoS_2 FETs, where green OLED is ON with $V_{\text{Data},1}$ of +5 V when blue one is OFF with $V_{\text{Data},2}$ of 0 V at a certain time t_1 , and vice versa at time t_2 . Such alternating digital operations are displayed with I_{OLED} and V_{OUT} from each OLED pixel

circuit (green and blue) as shown in Fig. 6b. Grayscale analog pixel operation was designed as plotted in time- V_{IN} ($=V_{\text{Data}}$), and in this case V_{IN} decreases with 1 volt step in 4 s. According to resultant time- I_{OLED} plots in Fig. 6d, e, grayscale pixel display appears quite clear in each step of I_{OLED} . For each color, four different intensities of OLED are obtained while green color is much brighter than blue one as expected. Another interesting to note is that V_{OUT} difference in analog green OLED driving is much minimal compared to that in analog blue driving. In other words, small V_{OUT} difference causes large difference in I_{OLED} for green OLED. Such difference between green and blue pixel would be again attributed to the I - V curve origins of green and blue OLED diode in the inset of Fig. 4a. Supporting Information, a video file (two MoS_2 OLED.avi) is also prepared to demonstrate the digital switching of 2×1 pixel array and grayscale analog switching of single blue/green OLED pixel.

In summary, we have fabricated CVD-grown a few mm large-scale MoS_2 FETs and investigated their low voltage OLED pixel circuit applications. In particular, when CVD MoS_2 monolayer was transferred onto gate-patterned Al_2O_3 dielectric on glass, an ultrathin hydrophobic PS layer was used as a moderating medium between MoS_2 and Al_2O_3 , to improve the uniformity of V_{th} and reduce the gate hysteresis in FETs. Our MoS_2 FET shows its average linear mobility of $8\text{--}10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and a high ON/OFF I_{D} ratio of 10^8 as patterned by photolithography on glass, and those values are regarded quite high or comparable to previous reports for CVD MoS_2 FETs^{28–32} because a single grain of our CVD MoS_2 monolayer appears as large as $\sim 60 \mu\text{m}$ in side dimension. Among 48 MoS_2 -based FETs, 38 devices successfully work (yield $\sim 80\%$) operating at ~ 5 V. Dynamic switching of blue and green OLED pixels was nicely demonstrated using the CVD monolayer MoS_2 FETs in two ways: 2×1 digital pixel array switching and grayscale analog switching of single blue/green OLED pixel. We conclude

that our CVD MoS₂ FETs are very promising as backpanel devices for future active matrix OLED pixel display.

METHODS

MoS₂ CVD growth

The 2D MoS₂ material was grown by CVD in a quartz tube furnace. A single precursor source was used, consisting of MoS₂ powder.³³ Instead of the combination of MoO₃ and S, which is more often reportedly used. The precursor was placed in a quartz boat at the center of a tube while the substrates were placed downstream. We did not use any substrate surface treatment to promote nucleation prior to growth. The growth was performed at a pressure of 10 mbar under 20 sccm Ar flow, with the furnace temperature ramped to 950 °C, for a duration of 20 min. This high temperature leads to the thermal evaporation of the MoS₂ precursor, which nucleates and re-deposits onto the substrates located at a cooler region of the furnace (estimated to be ~200 °C lower). The substrate temperature (~800 °C with a temperature of 900 °C for the powder precursor) is therefore also higher than those reported when a combination of MoO₃ and S was used. We believe this higher growth temperature leads to enhanced surface diffusion during growth and therefore, in combination with the use of a single precursor, contributes to the larger area MoS₂ monolayer film obtained here. Finally, the furnace was allowed to cool down naturally. Thermally oxidized silicon substrates were used in this work. The substrates were oxidized prior to growth under an O₂/Ar at 1000 °C at an atmospheric pressure for the durations of 60, 120, 180, and 240 min, resulting in an oxide thickness of approximately 45, 75, 90, and 145 nm, respectively, as confirmed by ellipsometry. In addition, we have used commercially available 300 nm SiO₂/Si wafer for comparison. To the best of our knowledge, this represents the first comparative study of MoS₂ growth on different oxide thickness, while the majority of reported data was on thick 300 nm oxide.

Transfer process

As-grown MoS₂ on the SiO₂/p⁺-Si substrate was spin-coated by PS layer (molecular weight = 280,000, Sigma-Aldrich) at 3500 rpm for 60 s. The sample was annealed at 85 °C for 15 min. After baking, the resultant PS/MoS₂/SiO₂/p⁺-Si structure was dipped into deionized (DI) water, so that the PS/MoS₂ membrane might be separated from the SiO₂/p⁺-Si substrate and floated on the surface of DI water. The PS/MoS₂ membrane was picked up and transferred onto the target (ultrathin PS/gate-patterned atomic layer deposition (ALD) Al₂O₃/glass or gate-patterned ALD Al₂O₃/glass) substrate. After the transfer to the target substrate, the sample was baked at 85 °C for 1 h and 180 °C for 30 min. The top PS residues were peeled-off by using toluene.⁴⁸ (Supporting Information, Figure S4)

Device fabrication

The glass substrates were cleaned in acetone and ethanol using an ultrasonicator for 15 min. The Au/Ti (20 nm/10 nm) bilayer was deposited by DC magnetron sputtering system and patterned for bottom gate-electrodes by photolithography and lift-off process (lift-off layer: LOR 3A, Micro Chemical, and photoresist layer: AZ GXR-601, AZ electronic materials). For dielectric layers, ultrathin PS/50 nm-thick Al₂O₃ (or 50 nm-thick Al₂O₃ only) layer was prepared by ALD system and PS coating. For hydrophobic PS layer spin-coating, dimethyl chlorosilane-terminated PS (Polymer Source, Product No. P3881-SSiCl) was initially dissolved in toluene (Aldrich) solvent, and the oxygen plasma cleaning (150 W, 50 sccm, 20 s) was applied to the prepared Al₂O₃ dielectric surface. The PS-brush solution (10 mg/mL) was spin-coated onto the Al₂O₃ surface and then heated at 170 °C for 48 h inside a vacuum oven, so that the ultrathin PS layer may be covalently bonded to the plasma-treated Al₂O₃ surface.^{49,50} As stated in the above transfer process, as-grown monolayer was transferred from SiO₂/p⁺-Si substrate to patterned bottom gate substrate using PS transfer method (Supporting Information, Figure S4).⁴⁸ After transfer, CVD MoS₂ flakes were patterned by photolithography and O₂ plasma dry etching process. For the source (S) and drain (D) ohmic contact electrodes, Au (50 nm) was deposited and patterned using conventional photolithography and lift-off processes. Finally, we annealed the device at 250 °C in N₂ ambient to improve the S/D contact for MoS₂ channel.

Measurements

The device current-voltage (*I*-*V*) characterizations were carried out by a semiconductor parameter analyzer (HP 4155C, Agilent Technologies) and

the capacitance-voltage (*C*-*V*) measurements were carried out by a LCR meter (HP4284A, Agilent Technologies). Electrical dynamics were investigated with a function generator (AFG 310, Sony/Tektronix). SEM images were taken with an Field Emission SEM (JSM-7800F, JEOL Ltd.). OLED sample pixels were supplied from Samsung Display Co.

DATA AVAILABILITY

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

ACKNOWLEDGEMENTS

The authors acknowledge the financial support from NRF (NRL program: Grant No. 2017R1A2A1A05001278, SRC program: Grant No.2017R1A5A1014862, vdWMRC center). J.H.P. acknowledges this research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (NRF-2017R1A6A3A11035872). S.M.K. acknowledges travel support from NSF for US-Korea collaboration.

AUTHOR CONTRIBUTIONS

H.K. performed the fabrication and measurements of the MoS₂ transistors and demonstration of the AMOLED pixel, S.G. and P.K. performed the synthesis and characterization of the MoS₂ films, S.M.K. performed Raman and PL measurement, J. H.P. and Y.J. helped with PS layer, S.Y. helped with 3D figure scheme techniques, and S.I. designed whole device experiments. All authors discussed the results.

ADDITIONAL INFORMATION

Supplementary Information accompanies the paper on the *npj 2D Materials and Applications* website (<https://doi.org/10.1038/s41699-019-0091-9>).

Competing interests: The authors declare no competing interests.

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