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# 1 Centimeter-Scale and Visible Wavelength 2 Monolayer Light-Emitting Devices

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13 **Keywords** transition metal dichalcogenide, electroluminescence, WS<sub>2</sub>,  
14 chemical vapor deposition, monolayer display, visible emission

15

## 16 **ABSTRACT**

17 **Monolayer two-dimensional transition metal dichalcogenides**  
18 **(TMDCs) have shown great promise for optoelectronic applications**  
19 **due to their direct bandgaps and unique physical properties. In**  
20 **particular, they can possess photoluminescence quantum yields (PL**  
21 **QY) approaching unity at the ultimate thickness limit, making their**  
22 **application in light-emitting devices highly promising. Here we**  
23 **synthesize and characterize large-area WS<sub>2</sub> grown via chemical**  
24 **vapor deposition (CVD) for visible (red) light-emitting devices. We**  
25 **perform detailed optical characterization of the synthesized films,**  
26 **which show peak PL QY as high as 12%. Electrically pumped**  
27 **emission from the synthetic WS<sub>2</sub> is achieved utilizing a transient-**  
28 **mode electroluminescence device structure, which consists of a**  
29 **single metal-semiconductor contact and alternating gate fields to**

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1 **achieve bipolar emission. Utilizing this aforementioned structure,**  
2 **we demonstrate a centimeter-scale ( $\sim 0.5 \text{ cm}^2$ ) visible (640 nm)**  
3 **display fabricated using TMDCs to showcase the potential of this**  
4 **material system for display applications.**

## 5 **1. Introduction**

6 Two-dimensional (2D) transition metal dichalcogenides (TMDCs), such  
7 as  $\text{WS}_2$  and  $\text{MoS}_2$ , offer versatile layer-dependent properties that have  
8 facilitated an emergent field of technological advancements.<sup>[1-4]</sup> TMDCs  
9 scaled down to the monolayer limit are of particular interest, at which point  
10 these materials exhibit direct bandgaps and several properties suitable for  
11 optoelectronic applications.<sup>[5-7]</sup> This class of materials does not require  
12 lattice-matching to form heterostructures and is amenable to bandgap  
13 engineering *via* various methods such as strain.<sup>[8-10]</sup> Importantly, these  
14 materials can exhibit near-unity photoluminescence (PL) quantum yields  
15 (QY),<sup>[11]</sup> even in the presence of defects as long as the particles are kept in  
16 the neutral exciton form.<sup>[12]</sup> PL QY is defined as the ratio of photons emitted  
17 to photons absorbed, and is a key figure of merit as it directly dictates the  
18 final efficiency when the materials are made into light-emitting devices or  
19 photovoltaics. Various micro-scale light-emitting devices have been  
20 demonstrated using TMDCs.<sup>[13-16]</sup> However, a challenge has been the  
21 requirement for simultaneous formation of low-resistance contacts to both  
22 electrons and holes in the same device. In one specific architecture, we  
23 recently demonstrated efficient bipolar carrier injection using transient-mode

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1 operation through a single Schottky contact.<sup>[17]</sup> The device effectively acts as  
2 a light-emitting capacitor, with minimal dependence on the contact metal to  
3 the semiconductor. This device structure was shown to work with exfoliated  
4 monolayers of MoS<sub>2</sub>, WS<sub>2</sub>, MoSe<sub>2</sub>, and WSe<sub>2</sub>. Furthermore, large-area (3 mm  
5 × 2 mm) emission was demonstrated using WSe<sub>2</sub> monolayers grown by  
6 chemical vapor deposition (CVD). Although the WSe<sub>2</sub> devices were bright,  
7 their emission is in the near infrared regime (750 nm peak emission).  
8 Recently, millimeter-scale WS<sub>2</sub> devices operated in the visible wavelength  
9 regime were demonstrated using a vertical architecture with quantum dots  
10 and polymers as electron/hole injection layers, but a more efficient device is  
11 still demanded.<sup>[18]</sup> In this regard, we report a centimeter-scale, bright, visible  
12 light-emitting device based on WS<sub>2</sub> monolayers synthesized *via* CVD<sup>[19-21]</sup>  
13 using a simple capacitor structure. The CVD-synthesized material exhibits a  
14 respectable peak PL QY of approximately 10%, without a droop at high  
15 injection levels. Using this material, we fabricate a transient-  
16 electroluminescence (t-EL) device and characterize its performance; in  
17 particular, its efficient light emission at high injection levels. Finally, we also  
18 fabricate a sixteen-pixel display with bright red EL emission (640 nm, peak  
19 output power 14 μW cm<sup>-2</sup>), which is visible in ambient room lighting.

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## 21 **2. Results and Discussion**

22 **Figure 1a** shows the PL spectra of the synthesized WS<sub>2</sub> measured over  
23 a pump dynamic range of 5 orders of magnitude (the inset shows the

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1 normalized PL spectra). The spot-to-spot variation is  $\sim 30$  meV in peak  
2 energy (Figure S1). The corresponding pump-power dependence of PL QY  
3 over a pump dynamic range of over 5 orders of magnitude is shown in Figure  
4 1b. Notably, the CVD-grown monolayer does not show a reduction in PL QY at  
5 high pump powers.<sup>4</sup> For the case of exfoliated  $WS_2$ , a droop is observed at  
6 the high pump power regime, which is attributed to biexcitonic  
7 recombination.<sup>[4,12]</sup> The absence of this behavior has been previously  
8 observed in CVD-grown  $WSe_2$  and  $MoS_2$ ,<sup>[22,23]</sup> however, the underlying  
9 mechanism is unknown, although it could be due to differences in  
10 background doping or the presence of strain as a result of the growth  
11 process.<sup>[12,22]</sup> Importantly, the lack of droop in the PL QY of CVD  $WS_2$  at high  
12 injection levels makes the material a highly attractive candidate for an  
13 electroluminescent device.

14 Growth conditions were optimized in order to maximize the monolayer  
15 coverage. Specifically, growth of  $WS_2$  was conducted in a two-heating zone  
16 furnace (Figure S2, details in Methods), with growth substrates,  $WO_3$  and KBr  
17 loaded into the downstream zone, and sulfur loaded into the upstream zone.  
18 The upstream zone temperature, in conjunction with the residual heat from  
19 the downstream zone, was used to control the sulfur vapor pressure and  
20 consequently the lateral coverage of  $WS_2$ . The optical images in **Figure 2a**  
21 show that the monolayer coverage increases with the upstream zone  
22 temperature (sulfur temperature), with peak coverage achieved at a sulfur  
23 temperature of 55 °C. Note that thickness of the film is identified by optical

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1 contrast and confirmed by atomic force microscopy and Raman spectroscopy  
2 (Figure S9). At higher sulfur temperatures, the material is predominantly bi/  
3 multilayer. The PL QY is within a few percent for all sulfur temperatures  
4 (Figure 2b), indicating that changes in coverage do not considerably affect  
5 the optical quality, although the samples grown with a sulfur temperature of  
6 55 °C generally show a higher QY. Figure 2c shows centimeter-scale WS<sub>2</sub>  
7 monolayer film after optimizing the synthesis conditions, i.e. using a sulfur  
8 temperature setpoint of 55 °C. The film exhibits ~90% monolayer coverage  
9 across the substrate. The corresponding PL image (Figure 2d) shows uniform  
10 emission where the monolayer is present. The dark regions (~10% coverage)  
11 in the PL image are attributed to bi/multilayers or pinholes, which could be  
12 improved in the future by other growth methods such as metal-organic CVD  
13 (MOCVD).

14 A simple device structure, which we previously used to achieve EL in  
15 2D semiconductors, was utilized here (shown schematically in **Figure 3a**).<sup>[17]</sup>  
16 In short, a single Al electrode (source) is fabricated on the monolayer which  
17 is grown directly on a p-doped silicon substrate with a 90 nm thick SiO<sub>2</sub> layer  
18 as the gate oxide. The 90 nm oxide layer was selected because it results in a  
19 higher extraction efficiency of the emitted light.<sup>[24]</sup> The device is operated  
20 using a bipolar square wave applied to the gate electrode while the source  
21 electrode is grounded. While the gate is held at a negative (positive)  
22 potential, holes (electrons) accumulate within the WS<sub>2</sub> layer. As the gate  
23 voltage ( $V_g$ ) is switched, holes (electrons) exit the device while electrons

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1 (holes) enter, resulting in the formation of excitons near the contact edge  
2 and their subsequent recombination and light emission.<sup>[17]</sup> It is important to  
3 note that the contact material used for the source, as well as the resulting  
4 Schottky barrier, are inconsequential to the overall efficiency as the injection  
5 during the transient is driven by tunneling through a thinned barrier (the  
6 corresponding band diagrams during the operation cycle are shown in Figure  
7 S4). Specifically, during the transient, if the voltage polarity is switched very  
8 fast (slew rate  $\sim 1$  V/ns), the voltage does not get dropped across the gate  
9 oxide as the semiconductor cannot be charged quickly enough. Instead, the  
10 applied voltage is dropped at the metal-semiconductor contact, resulting in a  
11 large band-bending within few nanometers of the metal contact. This  
12 subsequently results in efficient injection of charge carriers through  
13 tunneling, overcoming the problem of Schottky contacts.

14 For the WS<sub>2</sub> device presented here, the emission extends  $\sim 3$   $\mu\text{m}$  from  
15 the semiconductor contact edge. Due to the operation mechanism described  
16 above, injected carriers (*e.g.*, electrons) diffuse inward while the other type  
17 of charge (*e.g.*, holes) stored in the steady-state exit the semiconductor  
18 through the contact or recombine with incoming carriers. After the  $V_g$   
19 transient, before the system reaches steady-state the injected carrier density  
20 along the semiconductor is always higher near the source contact than away  
21 from the contact. Therefore, the recombination rate is higher near the  
22 contact. Note that the emission length depends on the interplay between  
23 several parameters, including the radiative lifetime, mobility (for free

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1 carriers), diffusivity (for excitons), and contact barrier height for both type of  
2 carriers.

3 Figure 3b shows a fabricated  $WS_2$  t-EL device. The source contact is  
4 patterned as an array of electrodes with a line spacing of  $6\ \mu\text{m}$  to maximize  
5 the emission area, and the EL measurements are performed in vacuum.  
6 When operated, the device shows near-uniform emission over the entire  
7 device area (corresponding EL image shown in Figure 3c).<sup>[17]</sup> The EL emission  
8 is in good accordance with the PL emission spectrum of the as-grown  
9 material, with minimal differences in peak position and spectral shape  
10 (Figure 3d). Device stability was tested by operating the device continuously  
11 at a frequency of 800 kHz with a  $V_g$  of  $\pm 45\text{V}$  for 11.5 hours (Figure 3e). The  
12 device continuously emits light throughout the long measurement period,  
13 although variation in intensity is observed. The operation mechanism of this  
14 device is verified using time-resolved EL (TREL) measurements, as shown in  
15 **Figure 4**. Pulsed emission predominantly occurs during negative-to-positive  
16 voltage transients and is consistent with the behavior which would be  
17 expected from a device with a lower Schottky barrier height to electrons  
18 (Figure S5).

19 To further characterize the transient-mode operation of the device, we  
20 investigated the dependence of EL on two primary parameters, specifically  
21 frequency and  $V_g$ . By operating the device at higher frequencies, the number  
22 of transients per second increases and the total light emitted by the device  
23 increases proportionally. Under gate frequencies ranging from 1 kHz to 100

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1 kHz, the device shows almost no variation in the emitted photon count per  
2 cycle (**Figure 5a**). It should be noted that higher frequency operation was  
3 limited by the parasitics in the device and the bandwidth of the power source  
4 used to drive the device. The EL as a function of  $V_g$  is shown in Figure 5b and  
5 indicates that the EL increases linearly with  $V_g$  once past the turn-on voltage  
6 ( $V_t$ ) of approximately 15 V (Figure 5b). The  $V_t$  is dependent on the bandgap  
7 ( $E_g$ ) of the semiconductor, the gate oxide thickness, and the parasitic  
8 impedances in the device. In the future, the onset voltage can be reduced by  
9 thinning the gate oxide, with the theoretical lower bound being  $V_t \sim E_g/2q$ , as  
10 discussed in ref 17. A challenge with the extraction of efficiency in t-EL  
11 devices is the lack of ability to accurately measure the electron-hole current  
12 injection per cycle. However, the overall external efficiency of the device can  
13 be roughly approximated as the ratio of emitted photons per cycle to the  
14 accumulated electron ( $n_0$ ) and hole ( $p_0$ ) concentrations in the device  
15 according to:<sup>[17]</sup>

$$16 \quad \eta_e = \frac{\text{photons/cycle}}{(n_0 + p_0) \times A}$$

17 where  $A$  is the device area. The total carrier concentration injected by the  
18 device is given by:<sup>[17]</sup>

$$19 \quad (n_0 + p_0) = \frac{C_{ox}(2V_g - E_g/q)}{q}$$

20 where  $C_{ox}$  is the gate oxide capacitance (38.4 nF cm<sup>-2</sup>) and  $q$  is the electron  
21 charge. Finally, the external efficiency is converted to an internal value by  
22 correcting for the fraction of light which is able to escape from the

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1 semiconductor to free space.<sup>[24]</sup> The device shows a peak internal efficiency  
2 of approximately 4% across the tested frequencies and shows a near-linear  
3 increase with respect to  $V_g$  (Figure 5c).

4 To demonstrate the scalability of  $WS_2$  for display applications, we  
5 fabricated a centimeter-scale and visible wavelength display utilizing TMDCs.  
6 A sixteen-pixel display was directly fabricated on a 7 mm  $\times$  7 mm as-grown  
7  $WS_2$  monolayer (**Figure 6a**). Each pixel is 1.2 mm  $\times$  1.2 mm, consisting of an  
8 array of source contacts designed to maximize the contact edge length to  
9 capitalize upon emission over the entire pixel area (Figure 6b). The entire  
10 display shares a common gate electrode (p++ Si/SiO<sub>2</sub>, 90 nm thickness). The  
11 device was then packaged in a standard chip carrier in which each pixel was  
12 directly wire-bonded, and then measured in vacuum. The pixels show similar  
13 performance to the devices shown in Figure 3 and can output a maximum  
14 power density of  $\sim 14 \mu W cm^{-2}$  at a  $V_g$  of  $\pm 28 V$  and frequency of 250 kHz  
15 (Figure 6c). Individual pixels can be selectively turned on by grounding the  
16 appropriate source contact (Figure 6d). The display was then operated to  
17 dynamically display the letters C-A-L under ambient room lighting and  
18 recorded using a commercial camera, showcasing the potential for TMDC-  
19 based light-emitting displays (Figure 6e-g).

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### 21 **3. Conclusions**

22 In summary, we have demonstrated a centimeter-scale and visible  
23 wavelength light-emitting display from CVD-grown  $WS_2$  monolayer

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1 semiconductors. The device operates in the transient mode and exhibits  
2 bright red emission, despite the emission layer being only 0.7 nm in  
3 thickness. The work highlights the potential use of monolayer  
4 semiconductors for ultrathin displays, taking advantage of their high  
5 luminescence quantum yields. Future work includes exploration of other  
6 monolayer semiconductors and/or strain-engineering for green, blue, and  
7 white light emission. Furthermore, the contacts and gate dielectrics can also  
8 be scaled down to monolayer or few-layer thicknesses by using appropriate  
9 2D materials, thus enabling the entire device to be at the ultimate thickness  
10 limit.

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## 1 **4. Experimental Section**

### 2 **Materials Growth**

3  $WS_2$  was grown *via* CVD on 90 nm Si/SiO<sub>2</sub> substrates. Growth was done in a  
4 two-heating zone furnace (Daepoong Industry, 50602). Substrates were first  
5 cleaned in acetone and isopropyl alcohol under sonication, then loaded into  
6 the downstream zone. Tungsten oxide (WO<sub>3</sub>) and potassium bromide (KBr)  
7 were added into an alumina combustion boat (Coorstek) at a 1:1 mass ratio  
8 and loaded in front of the substrates. A second alumina boat containing 1  
9 gram of sulfur was placed in the center of the upstream zone. Upon loading,  
10 the quartz tube was vacated, and Ar was introduced at 200 sccm, at which  
11 the pressure of the setup was adjusted to 10.0 torr. The upstream zone was  
12 first ramped to 55 °C, and subsequently the downstream zone to 800 °C. The  
13 upstream zone temperature, in conjunction with the residual heat from the  
14 downstream zone, was used to control the sulfur vapor pressure and  
15 consequently the lateral coverage of  $WS_2$ . The optimization results are shown  
16 in Figure S6. Once the temperature of the downstream zone was stabilized,  
17 Ar flow was lowered to 35 sccm, and H<sub>2</sub> was introduced at 20 sccm. The  
18 pressure was lowered to 2.2 torr, and growth was carried out for 22 mins.  
19 Upon completion of growth, H<sub>2</sub> flow was stopped and the furnace was opened  
20 to rapidly cool down the system.

### 21 **Device Fabrication**

22 Devices were patterned using conventional photolithographic techniques.  
23 However, alkaline developers were found to severely degrade the PL QY of

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1 the material as well as cause spalling of the grown film. Therefore, a tri-layer  
2 photolithographic process consisting of MMA EL9 (140 °C, 10 min bake)/ LOR-  
3 5A (140 °C, 10 min bake)/ OiR-906 (90 °C, 1 min bake) was used. The  
4 photoresist was exposed and developed using an unmodified process, and an  
5 additional development step in 50% acetone/50% methanol (by volume) was  
6 used to transfer the image to the MMA. Contacts were deposited by thermal  
7 evaporation of 40 nm thick Aluminum, and liftoff was performed in room  
8 temperature acetone.

### 9 **Optical and Electrical Characterization**

10 Photoluminescence (PL) and electroluminescence (EL) measurements were  
11 conducted using a custom-built micro-PL instrument described in detail in a  
12 prior work.<sup>11</sup> All PL and EL measurement signals were passed through a 550  
13 nm dielectric long-pass filter, dispersed by a  $f = 340$  mm spectrometer with  
14 a 150 mm  $g^{-1}$  grating, and then detected by a Si charge-coupled device  
15 (CCD) (Andor iDus BEX2-DD). Prior to each measurement, the CCD  
16 background was obtained and subtracted from the resulting acquisition. All  
17 measurements were collected via either a 50 $\times$  (numerical aperture 0.55) or  
18 a 10 $\times$  (numerical aperture 0.25) objective lens. Steady state PL was  
19 measured using an Ar<sup>+</sup> laser (Lexel 95) with a 514.5-nm line for excitation.  
20 The power density was adjusted *via* neutral filters and monitored by a  
21 photodiode power sensor (Thorlabs S120C). External PL QY calibration and  
22 the extraction of internal QY was conducted as outlined in our previous  
23 study.<sup>11,21</sup> Time-resolved PL was measured using a monochromated 514 nm

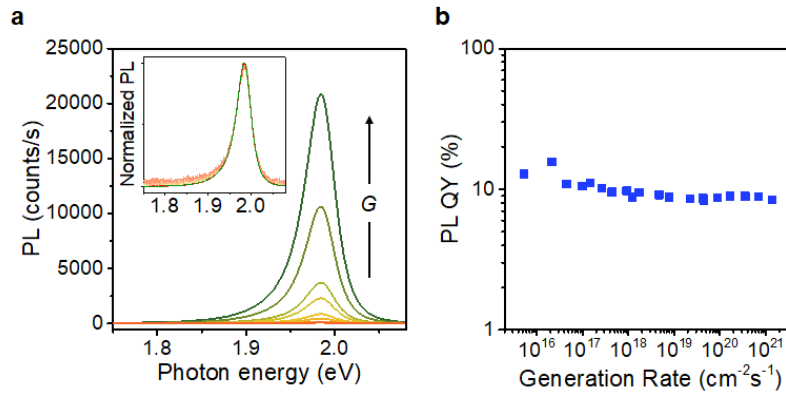
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1 line from a pulsed supercontinuum laser (Fianium WhiteLase SC-400) as an  
2 excitation source. The resulting signal was detected *via* a single-photon  
3 counting avalanche photodiode (ID Quantique) with a time-correlated single-  
4 photon counting (TCSPC) module (Becker-Hickl GmbH). Time-resolved EL was  
5 measured using an identical collection module. PL and EL imaging were  
6 conducted with a fluorescence microscopy setup and a CCD detector (Andor  
7 Luca) was used for image acquisition. For PL imaging only, a 470 nm LED  
8 excitation source was utilized. For the centimeter-scale film imaging, a CMOS  
9 camera with a telephoto lens equipped with a 550-nm colored glass long-  
10 pass filter was used. Macroscopic photographs of the EL device was taken  
11 with a commercial camera with a single exposure. All EL device  
12 measurements were done under vacuum at room temperature. Transistor  $I_d$ -  
13  $V_g$  characteristics were obtained using an Agilent B1500A semiconductor  
14 parameter analyzer (Figure S8).

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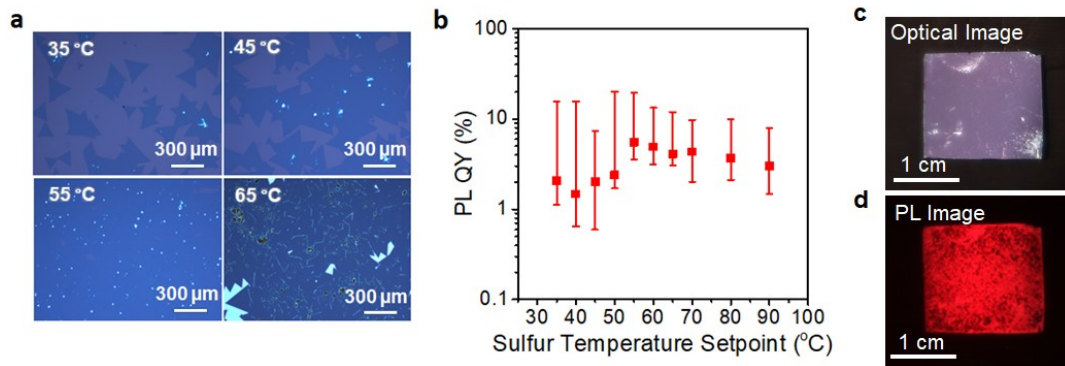


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2 **Figure 1.** a) The PL spectra of the synthesized WS<sub>2</sub> measured over a pump  
3 (*i.e.*, generation rate  $G$ ) dynamic range of 5 orders of magnitude from  $G =$   
4  $10^{16}$  to  $10^{21}$   $\text{cm}^{-2}\text{s}^{-1}$ . The inset is the normalized PL spectra which show a  
5 centered emission energy of 1.97 eV. b) The pump power dependence of the  
6 PL QY for a typical CVD WS<sub>2</sub> sample.

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2 **Figure 2.** a) Optical microscope images of CVD-grown WS<sub>2</sub> monolayers  
3 grown using varying sulfur temperatures. b) PL QY as a function of sulfur  
4 temperature. Each error bar shows the maximum, median (dots), and  
5 minimum values of the PL QY across 25 random spots on a single sample, for  
6 a given temperature. A distribution of PL peak energies is shown in Figure  
7 S3. c) Optical image of a CVD-grown WS<sub>2</sub> monolayer film on a 90 nm Si/SiO<sub>2</sub>  
8 substrate with a sulfur temperature of 55 °C. d) Macroscopic PL imaging of  
9 the CVD-grown sample under excitation by a 470 nm LED.

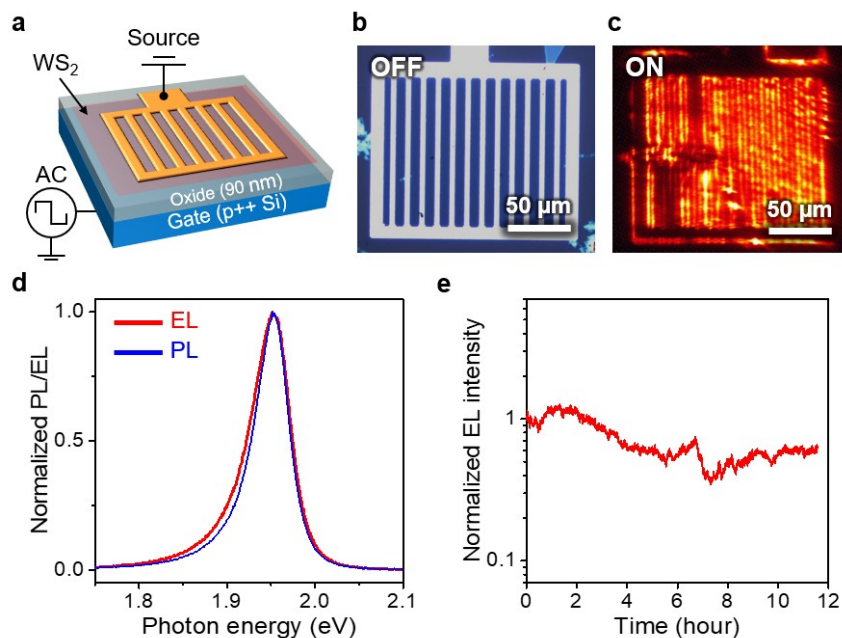
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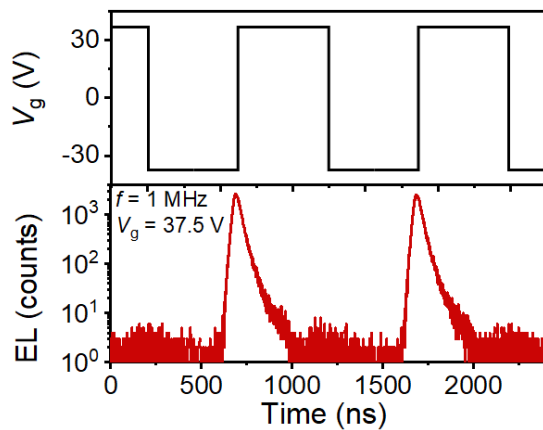
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3 **Figure 3.** a) Schematic of the t-EL device. b) Optical microscope image of  
4 the fabricated t-EL device. c) EL image of the device, showing emission is  
5 concentrated near the metal contacts. Taken under  $V_g = \pm 28$  V and  $f = 200$   
6 kHz. d) PL (at  $G = 10^{20}$  cm<sup>-2</sup>s<sup>-1</sup>) and EL ( $V_g = \pm 28$  V and  $f = 200$  kHz) spectra  
7 of the CVD-grown monolayer WS<sub>2</sub> device. e) Device stability measurement  
8 for the t-EL device taken under continuous operation for approximately 12  
9 hours under vacuum.

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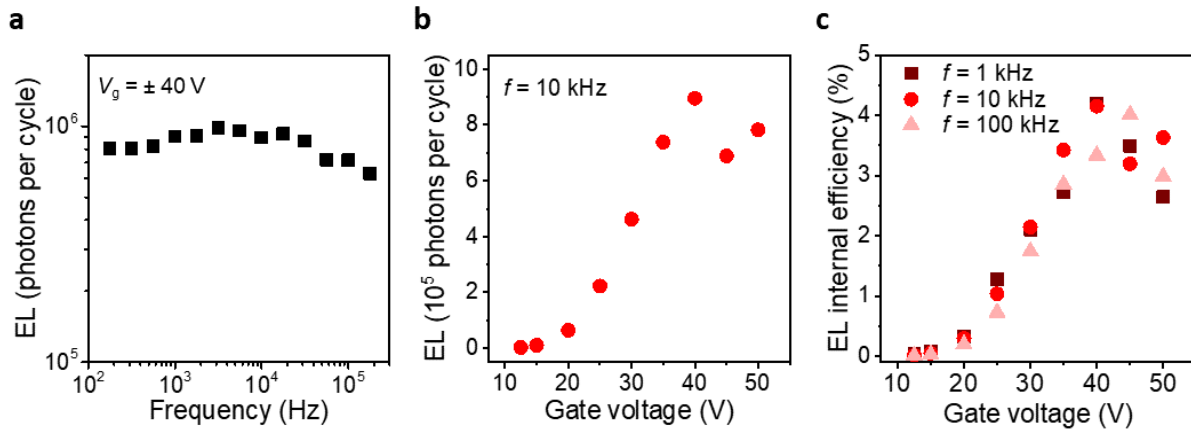
2 **Figure 4.** TREL measurement of the  $WS_2$  t-EL device with the corresponding  
3  $V_g$  profile shown in the upper panel. Note that the measurement is limited by  
4 the slew rate of the amplifier, resulting in a discrepancy between the TRPL  
5 (shown in Figure S7) and TREL lifetimes.

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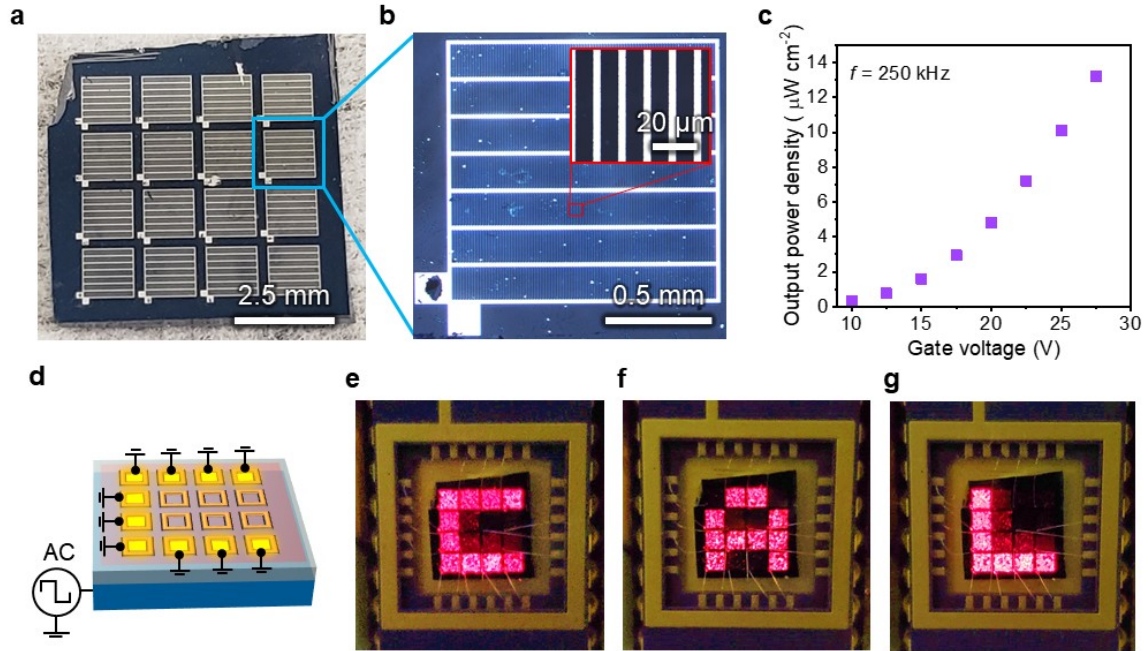
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2 **Figure 5.** a) Frequency dependence and b) voltage dependence of EL for the  
3 CVD-grown WS<sub>2</sub> t-EL device. c) EL internal efficiency of the device as a  
4 function of  $V_g$ .

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2 **Figure 6.** a) Photograph of the sixteen-pixel t-EL display. b) Optical  
3 microscope image of an individual pixel. c) Output power density of a single  
4 pixel as a function of  $V_g$ . d) The operation principle of the sixteen-pixel t-EL  
5 display. e-g) Photograph of the device sequentially displaying the letters C-A-  
6 L, respectively.

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1 **ASSOCIATED CONTENT**

2 **Supporting Information**

3 Supporting Information is available from the Wiley Online Library or from the  
4 author

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9 05Ch11231.

10 **Conflict of Interest**

11 The authors declare no conflict of interest

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1 **Large-area and visible Wavelength Monolayer Light-Emitting**  
2 **Devices.** We demonstrate a centimeter-scale and visible wavelength light-  
3 emitting display from CVD-grown WS<sub>2</sub> monolayer semiconductors. The  
4 device operates in the transient mode and exhibits bright red emission,  
5 despite the emission layer being only 0.7 nm in thickness. The work  
6 highlights the potential use of monolayer semiconductors for ultrathin  
7 displays, taking advantage of their high luminescence quantum yields.

8

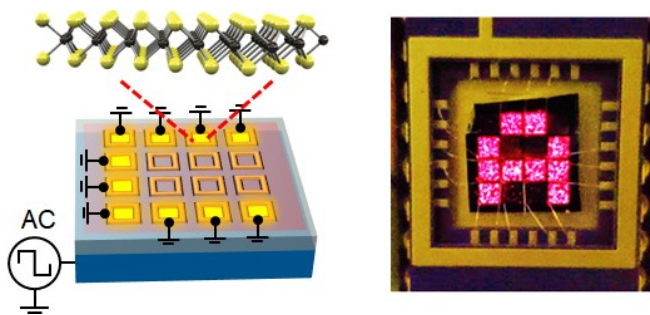
9 *Joy Cho<sup>1,2</sup>, Matin Amani<sup>1,2</sup>, Der-Hsien Lien<sup>1,2</sup>, Hyungjin Kim<sup>1,2</sup>, Matthew Yeh<sup>1,2</sup>,*  
10 *Vivian Wang<sup>1,2</sup>, Chaoliang Tan<sup>1,2</sup>, and Ali Javey<sup>1,2,\*</sup>*

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12 **Centimeter-Scale and Visible Wavelength Monolayer Light-Emitting**

13

### Devices



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